

# MAGNETISM AND MAGNETIC MATERIALS



HOTEL WESTWARD HO  
Phoenix, Arizona  
November 13 -16 1961



## MAGNETICS AND MAGNETIC MATERIALS EXHIBIT

*Council Room*

*WESTWARD HO HOTEL*

A Magnetic and Magnetic Materials exhibit has been arranged with some of the nation's outstanding firms engaged in research, manufacture, and application of magnetic materials, components, and equipment. The exhibits will be open each day of the Conference. The exhibit hours are 0900-1730 except the last day when the exhibits will close at 1630.

	Booth
<b>AMP, Inc. - Magnetics Division, Harrisburg, Pa.</b>	7
All magnetic logic including shift registers, counters and decoding networks showing operating 10 bit and 150 bit shift registers.	
<b>The Arnold Engineering Company, Marengo, Ill.</b>	4-6
Permanent Magnets of Alnico and Ceramic; Sillectron transformer cores; High Permeability tape wound cores of Deltamax, Permalloy, Supermalloy, Supermendur, as well as Bobbin cores; Powder cores of Molybdenum, Permalloy, Carboonyl Iron and Sendust; Barium Titanate Transducers; Special magnetic materials.	
<b>F. W. Bell, Inc., Columbus, Ohio</b>	14
Magnetic field measuring equipment.	
<b>Bell Telephone Laboratories, INC., Murray Hill, N. J.</b>	3
Research in Magnetism	
<b>The Carpenter Steel Company, Reading, Pa.</b>	18
Carpenter alloys for electronic, magnetic and electrical applications.	
<b>Crucible Steel Company of America, Pittsburgh, Pa.</b>	13
Permanent Magnets	
<b>Digital Equipment Corporation, Maynard, Mass.</b>	12
Magnetic core and memory testers and accessories.	
<b>Electromagnetics Corporation, Hudson, Mass.</b>	23
<b>Encyclopaedia Britannica, San Diego, Calif.</b>	15
<b>General Electric Research Laboratory and Magnetic Materials Section, Schenectady, N. Y. and Edmore, Mich.</b>	Kachina Lounge
Magnetic materials and phenomena.	
<b>Harvey-Wells Corporation - Magnetics Division, Framingham, Mass.</b>	1
Electromagnets, power supplies, NMR equipment.	
<b>Indiana General Corporation, Valparaiso, Ind.</b>	20 & 21
Permanent Magnets, Ferrites, Technical Ceramics, Memory Products.	
<b>Instrument Systems Corporation, College Point, N. Y.</b>	26
Hall generators, Hall devices and systems, Gaussmeters, Fluxmeters, Magnetic storage drums, Magnetic Amplifiers, Saturable Reactors, Ferrite components.	
<b>Leyman Corporation - Magnetics Division, Cincinnati, Ohio</b>	19
Plastiform magnets and magnetic materials.	
<b>Lumen, Inc., Joliet, Ill.</b>	11
Ferrotracer, Miniature coil winder, demonstration of self-balancing Magnetic Amplifier technique.	
<b>Magnetic Shield Division - Perfection Mica Company, Chicago, Ill.</b>	10
Magnetic Shielding Alloys.	
<b>Radio Frequency Laboratories, Inc., Boonton, N. J.</b>	2
Magnetic field measuring equipment, Magnet charging and Treating equipment.	
<b>Textron, Inc. - Dalmo Victor Company Division, Belmont, California</b>	22
Dalmo Victor Model SAVM-1, stabilized absolute Vector Magnetometer with probes for a variety of applications.	
<b>Ugine Industries, Inc., New York, N.Y.</b>	17
Magnet assemblies, Magnetic clutches, Magnetizers, Permanent Magnets, New scientific equipment.	
<b>U.S. Naval Ordnance Laboratory, Silver Spring, Md.</b>	8 & 9
<b>Varian Associates - Instrument Division, Palo Alto, California</b>	24 & 25
Electromagnet systems, nuclear magnetic resonance fluxmeters.	

The Seventh Annual Conference on Magnetism and Magnetic Materials is being held under the joint sponsorship of the American Institute of Electrical Engineers and the American Institute of Physics. The Institute of Radio Engineers, the Metallurgical Society of A.I.M.E. and the Office of Naval Research are cooperating with the sponsoring societies.

It is intended that the Conference will bring together in a stimulating environment those individuals who are interested in basic and applied work in magnetism.

This program contains abstracts of the invited and contributed papers. Discussion will be invited following the presentation of each of the papers.

The Conference is to be held November 13-16, 1961 in the Hotel Westward Ho, Phoenix, Arizona.

Inquiries should be directed to P. B. Myers, Local Chairman, Motorola, Inc., 5005 E. McDowell Road, Phoenix, Arizona.



## SUMMARY OF ACTIVITIES

Time	Session	Title	Locations*
<b>Monday</b>			
0800-		Registration	KR
0930-	I	General Session	TH
1400-	IIA	Magnetic Devices	TH
1400-	IIB	Internal Fields	TU
0900-1730		Magnetics Exhibit	CR
<b>Tuesday</b>			
0900-	IIIA	Thin Films - 1	TH
0900-	IIIB	Spin Configurations and Anisotropy	TU
2000-	IVA	Thin Films - 2	TH
2000-	IVB	Magnetization	TU
0900-1730		Magnetics Exhibit	CR
<b>Wednesday</b>			
0900-	VA	Oxides - 1	TH
0900-	VB	Soft Magnetic Materials	TU
1400-	VIA	Rare Earths	TU
1400-	VIB	Devices and Phenomena	TH
0900-1730		Magnetics Exhibit	CR
1730-		Cocktail Hour and Banquet	
<b>Thursday</b>			
0900-	VIIA	Antiferromagnetism and Resonance	TH
0900-	VIIB	Permanent Magnets and Micromagnetics	TU
1400-	VIIIA	Alloys and Compounds	TH
1400-	VIIIB	Oxides - 2 and Crystals	TU
0900-1630		Magnetics Exhibit	CR

\*KR - Kachina Room  
 TH - Thunderbird Room  
 TU - Turquoise Room  
 CR - Council Room

### Preregistration

A preregistration form accompanies this program. Everyone is urged to preregister to save time and aid the Conference Committee in its planning. Make checks payable to the Conference on Magnetism, and mail to D. L. Fresh, Motorola Inc., P. O. Box 5409, Phoenix, Arizona. SINCE THE ADDRESS GIVEN ON THIS FORM WILL BE USED IN MAILING YOUR CONFERENCE PROCEEDINGS, BE SURE TO GIVE AN ACCURATE AND SUFFICIENT ADDRESS.

### Registration and Proceedings

The registration desk will be located in the Kachina Room of the Hotel Westward Ho. It will be open from 0800 to 1600 on Monday, November 13, and from 0830 to 1630 on the other days of the Conference.

The registration fee will be \$10. Each registrant will receive a copy of the Proceedings which will appear as a separate issue of the Journal of Applied Physics to be published in the Spring of 1962.

### Hotel Reservations

A block of rooms has been reserved at the Hotel Westward Ho. It is anticipated that the Conference attendance will be larger than the number of reserved rooms and the

overflow will be allocated to neighboring facilities. Transportation will be provided to the Hotel Westward Ho. Rooms at the headquarters hotel will be assigned on a first-received basis. Please utilize the enclosed form or mention the Conference when requesting information. If difficulties develop, please contact the Local Chairman or F. R. Gleason, Motorola Inc., P. O. Box 5409, Phoenix, Arizona, Telephone: 945-6311.

### Cultural and Recreational Activities

Because of the many natural and resort attractions of Phoenix and the Valley of the Sun, sessions will be held on Tuesday, November 14, in the morning and evening. The afternoon will be open for sightseeing, either on an individual basis or in organized tours. Further information on places of interest and how to get there will be provided at the Conference.

In addition, a program for wives and families of attendees is being arranged. Those who have not received information on this may contact the Local Chairman or Mrs. D. L. Fresh, 5802 Calle Del Media, Scottsdale, Arizona.

### Social Gatherings and Banquet

On Wednesday evening, November 15, the social activities of the Conference will be climaxed by a banquet in the Thunderbird Room of the Hotel Westward Ho. The banquet will be preceded by a cocktail hour starting at 1730 hours. The banquet fee this year has been reduced to \$6.00. To avoid disappointment it is recommended that you place your order with the preregistration form.

The principal speaker at the banquet will be Thomas L. Martin, Jr., Dean of the College of Engineering at the University of Arizona, Tucson, who will discuss current problems in Engineering education. Dean Martin has a wide reputation for innovation and constructive action to meet the challenges facing education today.

### Information for Speakers

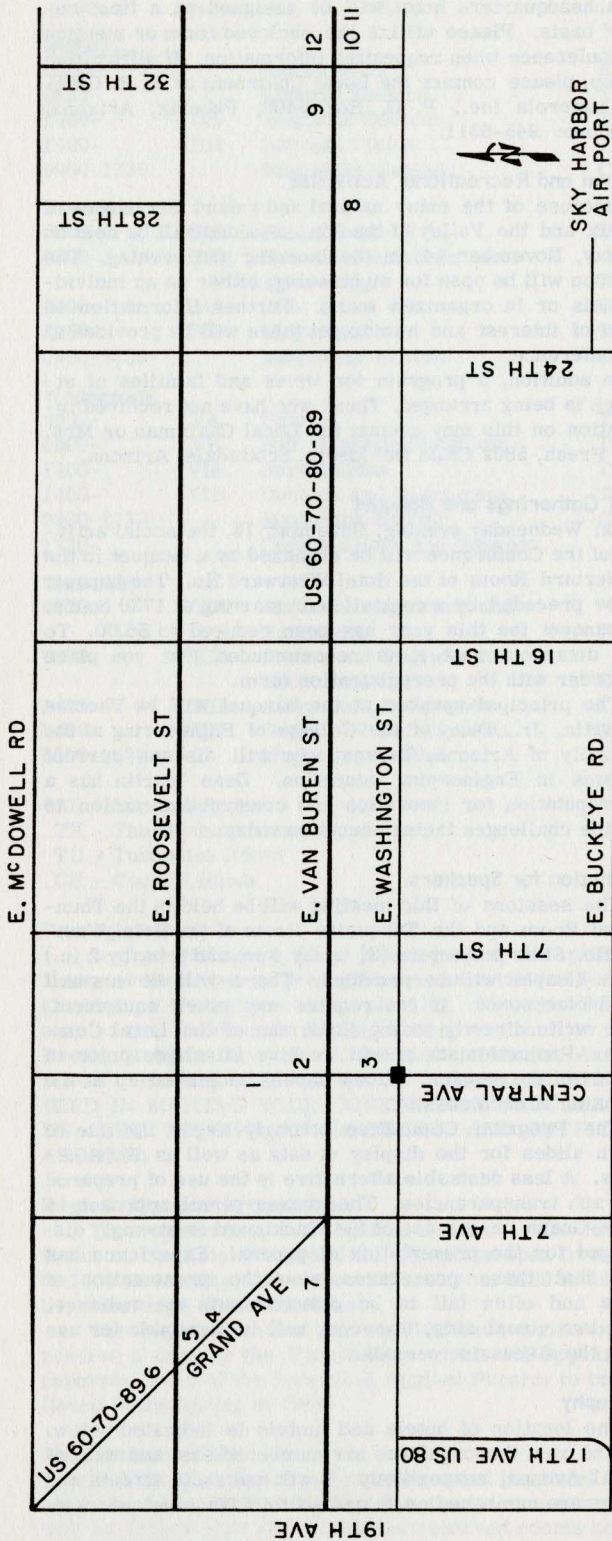
The sessions of this meeting will be held in the Thunderbird Room and the Turquoise Room of the Hotel Westward Ho. Slide projectors (3½ in. by 4 in. and 2 in. by 2 in.) and Vu-Graphs will be provided. There will be a small lighted blackboard. If you require any other equipment, please write directly to the Chairman of the Local Committee. Projectionists should receive all slides prior to the start of the session. Slides should be picked up at the conclusion of each session.

The Program Committee strongly urges the use of lantern slides for the display of data as well as of mathematics. A less desirable alternative is the use of prepared Vu-Graph transparencies. The grease pencil approach to the Vu-Graph or any use of the blackboard is strongly discouraged for the presentation of papers. Experience has shown that these procedures slow the presentation of papers and often fail to be effective with the audience. These two visual aids, however, will be available for use during the discussion periods.

### Geography

The location of hotels and motels is indicated below. East and west thoroughfares are numbered east and west of Central Avenue, respectively. North and south streets and avenues are numbered north and south of Washington Street.





- |                           |                             |
|---------------------------|-----------------------------|
| 1. Westward Ho            | 618 North Central Avenue    |
| 2. Sahara                 | 401 North 1st Street        |
| 3. Adams                  | North Central at East Adams |
| 4. Egyptian               | 765 Grand Avenue            |
| 5. Bali Hi                | 1515 Grand Avenue           |
| 6. Del Webb's HiWay Inn   | 1735 Grand Avenue           |
| 7. Desert Inn             | 950 West Van Buren Street   |
| 8. Desert Hills           | 2745 East Van Buren Street  |
| 9. Del Webb's HiWay House | 3148 East Van Buren Street  |
| 10. Continental Caravan   | 3323 East Van Buren Street  |
| 11. Desert Sky            | 3541 East Van Buren Street  |
| 12. Sands                 | 3320 East Van Buren Street  |



**CONFERENCE ON MAGNETISM AND  
MAGNETIC MATERIALS**

PHOENIX, NOVEMBER 13-16, 1961

*Hotel Westward Ho*

**Technical Program**

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**MONDAY, NOVEMBER 13, 1961**

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0930 Hours

Session I          Thunderbird Room

**GENERAL SESSION**

L. R. Bickford, Jr., Presiding

1. MODIFICATION OF SCREW SPIN STRUCTURE DUE TO ANISOTROPY ENERGY AND APPLIED FIELD (INVITED)  
T. Nagamiya  
Osaka University
2. MAGNETIC DISORDER AS A FIRST ORDER PHASE TRANSITION (INVITED)  
C. P. Bean and D. S. Rodbell  
General Electric Research Laboratory
3. HIGH MAGNETIC FIELD RESEARCH (INVITED)  
Benjamin Lax  
Lincoln Laboratory
4. SUPERCONDUCTING MATERIALS AND HIGH MAGNETIC FIELDS (INVITED)  
J. E. Kunzler  
Bell Telephone Laboratories, Incorporated
5. THE STATE OF THE ART OF MAGNETIC MEMORIES (INVITED)  
Q. W. Simkins  
International Business Machines Incorporated



1400 Hours

Session IIA      Thunderbird Room

## MAGNETIC DEVICES

W. L. Shevel, Jr., Presiding

6. PROPERTIES OF MAGNETIC FILMS FOR MEMORY SYSTEMS (INVITED)  
E. M. Bradley  
International Computers and Tabulators (Engineering) Ltd.
7. MAGNETO-OPTIC READOUT FOR COMPUTER MEMORIES  
R. L. Conger  
United States Naval Ordnance Laboratory
8. MAGNETO OPTICALLY MEASURED HIGH SPEED SWITCHING OF SANDWICH FILM ELEMENTS  
J. C. Suits and E. W. Pugh  
IBM Research Center
9. SWITCHING PROPERTIES OF MULTILAYER THIN FILM STRUCTURES  
A. Kolk, L. Douglas, and G. Schrader  
The National Cash Register Company
10. MAGNETOSTATIC INTERACTIONS BETWEEN THIN MAGNETIC FILMS  
Harrison W. Fuller and Donald L. Sullivan  
Laboratory for Electronics, Incorporated
11. PERMALLOY-SHEET TRANSFLUXOR-ARRAY MEMORY  
G. R. Briggs and J. W. Tuska  
RCA Laboratories
12. PROPOSAL FOR AND DEMONSTRATION OF MAGNETIC DOMAIN-WALL STORAGE AND LOGIC  
Donald O. Smith and J. M. Ballantyne  
Lincoln Laboratory
13. THRESHOLD PROPERTIES OF PARTIALLY SWITCHED FERRITE CORES  
V. T. Shahan and O. A. Gutwin  
IBM Research Center
14. FLUX REVERSAL IN FERRITE CORES UNDER THE EFFECT OF A TRANSVERSE FIELD  
Kam Li  
RCA Laboratories
15. APPROXIMATE SOLUTION OF THE EQUATIONS OF MAGNETIZATION REVERSAL BY COHERENT ROTATION  
R. F. Elfant  
IBM Research Center  
F. J. Friedlaender  
Purdue University
16. ALL MAGNETIC LOGIC ELEMENTS USING STRAINED PERMALLOY WIRE  
H. R. Irons  
United States Naval Ordnance Laboratory
17. A PERMANENT MAGNET TWISTOR MEMORY ELEMENT OF IMPROVED CHARACTERISTICS  
E. J. Alexander, J. C. McAlexander, R. J. Salhany, and L. H. Young  
Bell Telephone Laboratories, Incorporated

1400 Hours

Session IIB      Turquoise Room

## INTERNAL FIELDS

P. J. Wojtowicz, Presiding

18. NUCLEAR RESONANCE IN FERROMAGNETIC ALLOYS (INVITED)  
Toshimoto Kushida and A. H. Silver  
Ford Motor Company  
Yoshitaka Koi and Akira Tsujimura  
Tokushima University
19. CRYSTALLINE ELECTRIC FIELDS IN SPINEL-TYPE CRYSTALS  
V. J. Folen  
United States Naval Research Laboratory
20. THE AXIAL CRYSTAL FIELD IN CORUNDUM AND THE SUSCEPTIBILITY OF  $Ti_2O_3$   
S. Geschwind  
Bell Telephone Laboratories, Incorporated
21. HYPERFINE FIELDS IN TRANSITION METAL AND RARE EARTH IONS  
A. J. Freeman  
Ordnance Materials Research Office  
R. E. Watson  
AVCO R. A. D.
22. HYPERFINE MAGNETIC FIELD AND ELECTRON SPIN RELAXATION IN THE  $Sm^{+3}$  ION IN EUROPIUM IRON GARNET  
M. E. Caspari, S. Frankel, and G. T. Wood  
University of Pennsylvania
23. THE de HAAS van ALPHEN EFFECT IN ZINC MANGANESE ALLOYS  
F. T. Hedgcock and W. B. Muir  
The Franklin Institute Laboratories
24. ANISOTROPY OF THE HYPERFINE INTERACTION IN MAGNETITE  
E. L. Boyd and J. C. Slonczewski  
IBM Research Center
25. THE ATOMIC MOMENTS AND HYPERFINE FIELDS IN  $Fe_2Ti$  AND  $Fe_2Zr$   
C. W. Kocher and P. J. Brown  
Brookhaven National Laboratory
26. INTERNAL MAGNETIC FIELDS IN NICKEL RICH NICKEL-COBALT ALLOYS  
Lawrence H. Bennett and Ralph L. Streever, Jr.  
National Bureau of Standards



0900 Hours

Session IIIA Thunderbird Room

## THIN FILMS - 1

C. J. Kriessman, Presiding

27. THE INDUCED ANISOTROPY OF EVAPORATED FILMS FORMED IN MAGNETIC FIELD (INVITED)  
Minoru Takahashi  
Tohoku University
28. LABYRINTH DOMAINS IN PERMALLOY FILMS  
K. J. Harte and D. O. Smith  
Lincoln Laboratory
29. BITTER PATTERNS OF EPITAXIALLY GROWN SINGLE CRYSTAL THIN FILMS OF IRON AND NICKEL  
H. Sato, R. S. Toth, and R. W. Astrue  
Ford Motor Company
30. CONSTRICTION OF HARD DIRECTION HYSTERESIS LOOPS IN THIN PERMALLOY FILMS  
S. Middelhoek  
International Business Machines Corporation
31. A THEORETICAL MODEL FOR PARTIAL ROTATION  
H. Thomas  
International Business Machines Corporation
32. SUPPORT AND EXTENSION OF THE ROTATIONAL MODEL OF THIN FILM MAGNETIZATION  
J. E. Schwenker and T. R. Long  
Bell Telephone Laboratories, Incorporated
33. INHOMOGENEOUS COHERENT MAGNETIZATION ROTATION IN THIN MAGNETIC FILMS  
K. D. Leaver and M. Prutton  
International Computers and Tabulators (Engrg.) Ltd.
34. KERR MAGNETO-OPTIC STUDIES OF THE ANGULAR DISPERSION OF THE EASY AXIS OF PERMALLOY FILMS  
Robert W. Olmen and Sidney M. Rubens  
Remington Rand Univac
35. ELECTRON MICROSCOPE STUDY OF THE ROUGHNESS OF PERMALLOY FILMS USING SURFACE REPLICATION  
A. Baltz  
The Franklin Institute Laboratories
36. THE EFFECT OF SUBSTRATE CLEANNESS ON PERMALLOY THIN FILMS  
John S. Lemke  
Remington Rand Univac
37. ANISOTROPY SOURCES FOR ELECTRODEPOSITED PERMALLOY FILMS  
M. Lauriente and J. Bagrowski  
Westinghouse Electric Corporation
38. FACTORS INFLUENCING COERCIVE FIELD VALUES IN SPUTTERED PERMALLOY FILMS  
A. J. Noreika and M. H. Francombe  
Philco Research Division

0900 Hours

Session IIIB Turquoise Room

## SPIN CONFIGURATIONS AND ANISOTROPY

I. S. Jacobs, Presiding

39. MAGNETOELECTRIC EFFECTS IN ANTIFERROMAGNETICS (INVITED)  
George T. Rado  
United States Naval Research Laboratory
40. THE LATTICE THEORY OF SPIN CONFIGURATIONS (INVITED)  
E. F. Bertaut  
Nuclear Center and Laboratoire d'Electrostatique et de Physique du Metal
41. MAGNETIC STRUCTURE WORK AT THE NUCLEAR CENTER OF GRENOBLE  
E. F. Bertaut, A. Delapalme, F. Forrat, G. Roul, F. de Bergevin, and R. Pauthenet  
Laboratoire d'Electrostatique et de Physique du Metal
42. WEAK FERROMAGNETISM OF SOME ORTHOFERRITES  
D. Treves and S. Alexander  
Bell Telephone Laboratories, Incorporated
43. APPEARANCE OF WEAK FERROMAGNETISM IN FINE PARTICLES OF ANTIFERROMAGNETIC MATERIALS  
W. J. Schuele and V. D. Deetscreek  
The Franklin Institute Laboratories
44. THE MAGNETIC STRUCTURE OF MANGANESE CHROMITE  
L. M. Corliss and J. M. Hastings  
Brookhaven National Laboratory
45. MAGNETIC TRANSITIONS IN CUBIC SPINELS  
N. Menyuk, A. Wold, D. B. Rogers, and K. Dwight  
Lincoln Laboratory
46. NEUTRON DIFFRACTION STUDIES ON EUROPIUM METAL  
C. E. Olsen, N. G. Nereson, and G. P. Arnold  
Los Alamos Scientific Laboratory
47. NEUTRON DIFFRACTION STUDY OF MAGNETIC ORDERING IN THULIUM  
W. C. Koehler, J. W. Cable, E. O. Wollan, and M. K. Wilkinson  
Oak Ridge National Laboratory
48. THE NATURE OF ONE-ION MODELS OF THE FERRIMAGNETIC ANISOTROPY  
Peter J. Wojtowicz  
RCA Laboratories
49. THE TEMPERATURE DEPENDENCE OF THE MAGNETOCRYSTALLINE ANISOTROPY OF FACE-CENTERED CUBIC COBALT  
D. S. Rodbell  
General Electric Research Laboratory



2000 Hours

Session IVA Thunderbird Room

## THIN FILMS - 2

H. Rubinstein, Presiding

50. ELECTRODEPOSITION OF MAGNETIC MATERIALS (INVITED)  
I. W. Wolf  
General Electric Company
51. ROTATABLE MAGNETIC ANISOTROPY IN COMPOSITE FILMS  
J. M. Lommel and C. D. Graham, Jr.  
General Electric Research Laboratory
52. UNIDIRECTIONAL HYSTERESIS IN THIN PERMALLOY FILMS  
W. D. Doyle, J. E. Rudisill, and S. Shtrikman  
The Franklin Institute Laboratories
53. FERROMAGNETIC RESONANCE IN SINGLE CRYSTAL NICKEL FILMS  
M. Pomerantz, J. F. Freedman, and J. C. Suits  
IBM Research Center
54. STRATIFICATION IN THIN PERMALLOY FILMS  
R. J. Prosen, J. O. Holmen, B. E. Gran, and T. J. Cebulla  
Minneapolis-Honeywell Research Center
55. FERROMAGNETIC RESONANCES IN THIN FILMS  
D. Chen and A. H. Morrish  
University of Minnesota
56. ISOTROPIC STRESS MEASUREMENTS IN EVAPORATED PERMALLOY FILMS  
G. P. Weiss and D. O. Smith  
Lincoln Laboratory
57. HALL EFFECT DETERMINATION OF PLANAR STRESS IN FERROMAGNETIC FILMS  
R. L. Coren  
Philco Research Division
58. STRESS EFFECTS ON THE MAGNETIC PROPERTIES OF EVAPORATED SINGLE CRYSTAL NICKEL FILMS  
J. F. Freedman  
IBM Research Center
59. MAGNETOELASTIC BEHAVIOR OF THIN FERROMAGNETIC FILMS AS A FUNCTION OF COMPOSITION  
E. N. Mitchell and G. I. Lykken  
University of North Dakota

2000 Hours

Session IVB Turquoise Room

## MAGNETIZATION

S. Foner, Presiding

60. STATISTICAL MECHANICS OF FERROMAGNETISM (INVITED)  
Herbert Callen  
University of Pennsylvania
61. MAGNETIZATION OF LOCALIZED STATES IN METALS (INVITED)  
P. A. Wolff  
Bell Telephone Laboratories, Incorporated
62. MAGNETIZATION OF NICKEL AT LOW TEMPERATURES (INVITED)  
E. W. Pugh and B. E. Argyle  
IBM Research Center
63. TEMPERATURE DEPENDENCE OF YIG MAGNETIZATION  
Irvin H. Solt, Jr.  
Fairchild Semiconductor
64. NUCLEAR MAGNETIC RESONANCE IN THE INSULATING FERROMAGNET  $\text{CrBr}_3$   
A. C. Gossard, V. Jaccarino, and J. P. Remeika  
Bell Telephone Laboratories, Incorporated
65. FERROMAGNETIC RESONANCE IN  $\text{CrBr}_3$   
J. F. Dillon, Jr.  
Bell Telephone Laboratories, Incorporated
66. PRINCIPLES AND PROCEDURES FOR PRECISION MEASUREMENTS OF MAGNETIC PROPERTIES  
A. Arrott, C. H. Haakana, and J. E. Noakes  
Ford Motor Company



0900 Hours

Session VA Thunderbird Room

## OXIDES - 1

L. M. Corliss, Presiding

67. ACOUSTIC LOSSES IN FERROMAGNETIC INSULATORS (INVITED)  
R. C. LeCraw, E. G. Spencer, and E. I. Gordon  
Bell Telephone Laboratories, Incorporated
68. RARE EARTH RUTHENATES  
R. Aleonard, E. F. Bertaut, M. C. Montmory, and R. Pauthenet  
Laboratoire d'Electrostatique et de Physique du Metal
69. SUBSTITUTIONS OF DIVALENT TRANSITION METAL IONS IN YTTRIUM IRON GARNET  
S. Geller, H. J. Williams, R. C. Sherwood, and G. P. Espinosa  
Bell Telephone Laboratories, Incorporated
70. LOW TEMPERATURE ANISOTROPY OF MANGANESE-IRON FERRITES  
Wilfred Palmer  
IBM Research Center
71. PREPARATION AND PROPERTIES OF FERROSPINELS CONTAINING  $Ni^{+3}$   
M. W. Shafer  
IBM Research Center
72. PERMINVAR CHARACTERISTICS OF NICKEL-CADMIUM FERRITES WITH SMALL ADDITIONS OF MOLYBDENUM AND COBALT  
H. Lessoff and A. P. Greifer  
Radio Corporation of America
73. VANADIUM IRON OXIDES  
A. Wold, D. Rogers, R. J. Arnott, and N. Menyuk  
Lincoln Laboratory
74. X-RAY AND MAGNETIC STUDIES OF  $CrO_2$  SINGLE CRYSTALS  
W. H. Cloud, D. S. Schreiber, and K. R. Babcock  
E. I. du Pont de Nemours and Company
75. CATION-CATION THREE-ELECTRON BONDS  
John B. Goodenough  
Lincoln Laboratory
76. SOME SUPERPARAMAGNETIC PROPERTIES OF FINE PARTICLE  $\delta$   $FeOOH$   
A. W. Simpson  
The Plessey Co., Ltd.
77. FINE-GRAINED FERRITES. II.  $Ni_{1-x}Zn_xFe_2O_4$   
W. W. Malinofsky, R. W. Babbitt, and G. C. Sands  
United States Army Signal Research and Development Laboratory

0900 Hours

Session VB Turquoise Room

## SOFT MAGNETIC MATERIALS

P. A. Albert, Presiding

78. RECENT DEVELOPMENTS IN SOFT MAGNETIC ALLOYS (INVITED)  
Edmond Adams  
United States Naval Ordnance Laboratory
79. MAGNETIC PROPERTIES OF TAPE WOUND CORES OF HIGH-PURITY 3 PER CENT SILICON IRON WITH THE (110) [001] TEXTURE  
J. L. Walter  
General Electric Research Laboratory
80. MAGNETIC ANISOTROPY OF THE DEMAGNETIZED STATE  
Jean-Claude Barbier and Bernadette Ferlin-Guion  
Laboratoire d'Electrostatique et de Physique du Metal
81. ON THE TWO EFFECTS OF CHANGES IN TENSION  
Osamu Yamada  
Laboratoire d'Electrostatique et de Physique du Metal
82. TEXTURED 6.5 PER CENT SILICON-IRON ALLOY  
G. Facaros  
Westinghouse Materials Manufacturing Department
- R. G. Aspden  
Westinghouse Research Laboratories
83. GRAIN-SIZE EFFECTS IN ORIENTED 48 PER CENT NICKEL-IRON CORES AT 400 CYCLES  
M. F. Littmann, E. S. Harris, and C. E. Ward  
Armco Steel Corporation
84. ANISOTROPY IN ROLLED PERMALLOY TAPE  
E. M. Gyorgy and D. Treves  
Bell Telephone Laboratories, Incorporated
85. A METHOD OF MAGNETIC ANNEALING AND SOME ELEVATED TEMPERATURE MEASUREMENTS ON VANADIUM PERMENDUR TYPE ALLOYS  
R. E. Burket, D. M. Stewart, E. C. Falkowski, and R. H. Johns  
Allegheny Ludlum Steel Corporation
86. THE EFFECTS OF LOW TEMPERATURE NEUTRON IRRADIATION ON THE MAGNETIC PROPERTIES OF IRON-NICKEL ALLOYS  
A. I. Schindler  
United States Naval Research Laboratory
- R. H. Kernohan  
Oak Ridge National Laboratory
87. COOLING RATE EFFECT ON INITIAL PERMEABILITY OF 4-79 MOLY-PERMALLOY  
A. A. Lykens  
The Carpenter Steel Company
88. LOW ANGLE X-RAY SCATTERING FROM  $MnS$  IN SILICON STEELS  
Bani R. Banerjee and R. E. Lenhart  
Crucible Steel Company of America
- W. H. Robinson  
Carnegie Institute of Technology



## 1400 Hours

Session VIA      Turquoise Room

## RARE EARTHS

J. F. Dillon, Presiding

89. FERRIMAGNETIC RESONANCE IN TERBIUM DOPED YTTRIUM IRON GARNET (INVITED)  
L. R. Walker  
Bell Telephone Laboratories, Incorporated
90. FAR INFRARED SPECTRA OF MAGNETIC MATERIALS (INVITED)  
M. Tinkham  
University of California
91. MAGNETOCRYSTALLINE ANISOTROPY OF RARE EARTH IRON GARNETS (INVITED)  
R. F. Pearson  
Mullard Research Laboratories
92. THE CONTRIBUTIONS OF RARE EARTH IONS TO THE ANISOTROPY OF IRON GARNETS  
B. A. Calhoun, M. J. Freiser, and R. F. Penoyer  
IBM Research Center
93. SPINWAVE LINEWIDTHS IN RARE-EARTH SUBSTITUTED YTTRIUM IRON GARNET  
P. E. Seiden  
IBM Research Center
94. HIGH FIELD LOW TEMPERATURE MAGNETIZATION STUDIES OF PRASEODYMIUM SUBSTITUTED YTTRIUM IRON GARNET  $5\text{Fe}_2\text{O}_3 \times \text{Pr}_2\text{O}_3(3-X)\text{Y}_2\text{O}_3$   
Warren E. Henry  
Lockheed Missiles and Space Company  
Gerard Villers and Jean Lories  
Centre National de la Recherche Scientifique
95. PARAMAGNETIC RESONANCE OF  $\text{Gd}^{+++}$  IN YTTRIUM GALLIUM GARNET  
L. Rimai and G. A. Demars  
Raytheon Company
96. NEUTRON MAGNETIC SCATTERING FROM RARE EARTH IONS  
R. E. Watson  
AVCO, R. A. D.  
A. J. Freeman  
Ordnance Materials Research Office
97. HIGH TEMPERATURE SUSCEPTIBILITY OF GARNETS: EXCHANGE INTERACTIONS IN YIG AND LuIG  
Peter J. Wojtowicz  
RCA Laboratories

## 1400 Hours

Session VIB      Thunderbird Room

## DEVICES AND PHENOMENA

C. L. Hogan, Presiding

98. A COAXIAL FERRITE PHASE SHIFTER FOR HIGH POWER APPLICATIONS  
A. S. Boxer and R. S. McCarter  
Bell Telephone Laboratories, Incorporated
99. SLOW-WAVE UHF FERRITE PHASE SHIFTERS  
N. G. Sakiotis and D. E. Allen  
Motorola Incorporated
100. EFFICIENT FREQUENCY DOUBLING FROM FERRITES AT THE 100-WATT LEVEL  
A. S. Risley and I. Kaufman  
Space Technology Laboratories, Incorporated
101. A MAGNETO-DYNAMIC MODE FERRITE AMPLIFIER  
Roy W. Roberts and Robert Schell  
Melabs  
Bert A. Auld  
Stanford University
102. A MINIATURIZED FERRIMAGNETIC HIGH POWER CO-AXIAL DUPLEXER-LIMITER  
J. Clark and J. Brown  
Sperry Microwave Electronics Company
103. DESIGN CONSIDERATIONS FOR BROADBAND SYMMETRICAL "WYE" TUNABLE FERRITE CIRCULATORS IN TEM MODE STRIP TRANSMISSION LINE  
D. S. Elkort  
Narda Microwave Corporation
104. NONLINEAR EFFECTS IN FERRITE FILLED REDUCED SIZE WAVEGUIDE  
F. S. Hickernell, B. H. Auten, and N. G. Sakiotis  
Motorola Incorporated
105. MILLIMETER WAVE PARAMETRIC AMPLIFICATION WITH ANTIFERROMAGNETIC MATERIALS  
R. A. Moore  
Westinghouse Electric Corporation
106. A MICROWAVE MAGNETIC MICROSCOPE  
R. F. Soohoo  
Lincoln Laboratory
107. SAMPLING MAGNETOMETER BASED ON THE HALL EFFECT  
H. H. Wieder  
United States Naval Ordnance Laboratory
108. NEW TYPE OF FLUX-GATE MAGNETOMETER  
William A. Geyger  
United States Naval Ordnance Laboratory
109. WAVEFORM OF THE TIME RATE OF CHANGE OF TOTAL FLUX FOR MINIMUM CORE LOSS  
H. L. Schenk and F. J. Young  
Westinghouse Research Laboratories
110. IRON LOSSES IN ELLIPTICALLY ROTATING FIELDS  
R. D. Strattan and F. J. Young  
Carnegie Institute of Technology
111. LOW FREQUENCY LOSSES AND DOMAIN BOUNDARY MOVEMENTS IN SILICON IRON  
Daniel A. Wycklendt  
Allis-Chalmers Manufacturing Company
112. PROPERTIES OF RESET CORES IN RADAR PULSE TRANSFORMERS  
Rueben Lee  
Westinghouse Electric Corporation



0900 Hours

Session VIIA

Thunderbird Room

## ANTIFERROMAGNETISM AND RESONANCE

M. T. Weiss, Presiding

113. PERIODIC ADIABATIC TIME VARIATIONS (INVITED)  
H. Suhl  
University of California, San Diego
114. FIELD DEPENDENT MAGNETIC SUSCEPTIBILITY AND  
AND MAGNETOSTRICTION IN ANTIFERROMAGNETIC  
CoO, MnO AND NiO SINGLE CRYSTALS  
T. R. McGuire and W. A. Crapo  
IBM Research Center
115. PULSED CRITICAL FIELD MEASUREMENTS IN MAGNETIC  
SYSTEMS  
Simon Foner and Shou-Ling Hou  
National Magnet Laboratory
116. ANTIFERROMAGNETIC RESONANCE IN  $MnTiO_3$   
J. J. Stickler and G. S. Heller  
Lincoln Laboratory
117. ANTIFERROMAGNETIC AND PARAMAGNETIC RESONANCE  
IN  $CuF_2 \cdot 2H_2O$   
M. Peter and T. Moriya  
Bell Telephone Laboratories, Incorporated
118. FERROMAGNETIC RESONANCE LOSS IN LITHIUM  
FERRITE AS A FUNCTION OF TEMPERATURE  
R. T. Denton and E. G. Spencer  
Bell Telephone Laboratories, Incorporated
119. IMPURITY ION EFFECTS IN THE FERRIMAGNETIC  
RESONANCE OF ORDERED LITHIUM FERRITE  
A. D. Schnitzler, V. J. Folen, and G. T. Rado  
United States Naval Research Laboratory
120. FERROMAGNETIC RESONANCE MAGNON DISTRIBUTION  
IN YTTRIUM IRON GARNET  
T. J. Matcovich, H. S. Belson, N. Goldberg, and C. W. Haas  
Remington Rance Univac
121. ON THE POSSIBILITY OF OBTAINING LARGE AMPLITUDE  
TUBE RESONANCE IN VERY THIN FERRIMAGNETIC  
DISKS  
F. R. Morgenthaler  
Massachusetts Institute of Technology
122. MICROWAVE RESONANCE LINEWIDTH IN SINGLE  
CRYSTALS OF COBALT SUBSTITUTED MANGANESE  
FERRITE  
R. W. Teale  
Mullard Research Laboratories
123. DC EFFECTS IN FERROMAGNETIC RESONANCE IN  
THIN FERRITE FILMS  
W. Heinz and L. Silber  
Polytechnic Institute of Brooklyn

0900 Hours

Session VIIB

Turquoise Room

## PERMANENT MAGNETS AND MICROMAGNETICS

F. E. Luborsky, Presiding

124. EXCHANGE ANISOTROPY - A REVIEW (INVITED)  
W. H. Meiklejohn  
General Electric Research Laboratory
125. PERMANENT MAGNETIC PROPERTIES OF IRON-COBALT  
PHOSPHIDES  
K. J. de Vos, W. A. J. J. Velge, and M. G. v. d. Steeg  
N. V. Phillips' Gloeilampenfabrieken
126. STUDIES OF HIGH COERCIVITY COBALT-PHOSPHOROUS  
ELECTRODEPOSITS  
J. S. Sallo and J. M. Carr  
Minneapolis-Honeywell Research Center
127. THE FORMATION OF MONOCRYSTALLINE ALNICO  
MAGNETS BY SECONDARY RECRYSTALLIZATION  
METHODS  
E. Steinort  
Centro Magneti Permanenti, S. P. A.  
E. R. Cronk, S. J. Garvin, and H. Tiderman  
Thomas & Skinner, Incorporated
128. SATURATION MAGNETIZATION OF SWAGED Mn-Al  
M. A. Bohlmann  
Indiana General Corporation
129. STUDY OF PARTICLE ARRANGEMENTS AND MAGNETIC  
DOMAINS ON THE SURFACE OF PERMANENT MAGNETS  
K. J. Kronenberg  
Indiana General Corporation
130. ON THE POSSIBILITY OF DOMAIN WALL NUCLEATION  
BY THERMAL AGITATION  
Amikam Aharoni  
The Weizmann Institute of Science
131. FAILURE OF THE LOCAL-FIELD CONCEPT FOR  
HYSTERESIS CALCULATIONS  
William Fuller Brown, Jr.  
University of Minnesota
132. SINGLE-DOMAIN MAGNETIZATION CURVES FOR THE  
GENERAL ELLIPSOID  
Clark E. Johnson, Jr.  
University of Minnesota
133. THE DEPENDENCE OF AVERAGE INTERACTION  
FIELDS ON INTENSITY OF MAGNETIZATION IN  
ASSEMBLIES OF FERROMAGNETIC PARTICLES  
G. Bate  
International Business Machines Corporation
134. THE EFFECT OF ANGULAR VARIATIONS IN FIELD  
ON FINE-PARTICLE REMANENCE  
Eric D. Daniel  
Memorex Corporation  
R. Noble  
M. S. S. Recording Company
135. REMANENT TORQUE STUDIES IN POLYCRYSTALLINE  
 $BaFe_{12}O_{19}$   
P. J. Flanders and S. Shtrikman  
The Franklin Institute Laboratories



1400 Hours

Session VIIIA Thunderbird Room

## ALLOYS AND COMPOUNDS

J. A. Osborn, Presiding

136. AGE-HARDENED GOLD PERMALLOY AND GOLD PERMINVAR  
E. A. Nesbitt and E. M. Gyorgy  
Bell Telephone Laboratories, Incorporated
137. PARAMAGNETIC BEHAVIOR OF IRON-RICH IRON-VANADIUM ALLOYS  
Sigurds Arajs, R. V. Colvin, Henry Chessin, and J. M. Peck  
United States Steel Corporation
138. MAGNETIC CHARACTERISTICS OF HYDROGENATED HOLMIUM  
Y. Kubota and W. E. Wallace  
University of Pittsburgh
139. NEW MODIFIED  $Mn_2Sb$  COMPOSITIONS SHOWING EXCHANGE INVERSION  
T. A. Bither, P. H. L. Walter, W. H. Cloud, T. J. Swoboda, and P. E. Bierstedt  
E. I. du Pont de Nemours and Company
140. MAGNETIC STRUCTURES OF  $Mn_2As$  AND  $Mn_2Sb_{0.7}As_{0.3}$   
A. E. Austin and E. Adelson  
Battelle Memorial Institute  
W. H. Cloud  
E. I. du Pont de Nemours and Company
141. STRUCTURAL AND MAGNETIC PROPERTIES OF COPPER-SUBSTITUTED MANGANESE-ALUMINUM ALLOYS  
Makoto Sugihara and Ichiro Tsuboya  
Nippon Telegraph and Telephone Public Corporation
142. MAGNETIC SUSCEPTIBILITY AND INTERNAL FRICTION OF TETRAGONAL MANGANESE-COPPER ALLOYS CONTAINING 70 PER CENT MANGANESE  
A. E. Schwaneke and J. W. Jensen  
Bureau of Mines
143. ANOMALOUS MAGNETIC MOMENTS AND TRANSFORMATIONS IN THE ORDERED ALLOY  $FeRh$   
J. S. Kouvel and C. C. Hartelius  
General Electric Research Laboratory
144. ALLOYS OF THE FIRST TRANSITION SERIES WITH PALLADIUM AND PLATINUM  
S. J. Pickart  
United States Naval Ordnance Laboratory  
R. Nathans  
Brookhaven National Laboratory
145. NEUTRON DIFFRACTION INVESTIGATIONS OF FERROMAGNETIC PALLADIUM AND IRON GROUP ALLOYS  
J. W. Cable, E. O. Wollan, W. C. Koehler, and M. R. Wilkinson  
Oak Ridge National Laboratory
146. MAGNETIC PROPERTIES OF MIXED VANADIUM-CHROMIUM SULFIDES  
K. Dwight, R. W. Germann, A. Wold, and N. Menyuk  
Lincoln Laboratory
147. MAGNETIC MOMENT OF  $Co-Cu$  SOLID SOLUTIONS WITH 40 TO 85 PER CENT  $Cu$   
E. Kneller  
IBM Research Center
148. MAGNETIC SUSCEPTIBILITIES AND EXCHANGE EFFECTS IN FOUR ORGANIC FREE RADICALS  
J. H. Burgess, R. S. Rhodes, M. Mandel, and A. S. Edelstein  
Stanford University

1400 Hours

Session VIIIB Turquoise Room

## OXIDES - 2 AND CRYSTALS

R. L. White, Presiding

149. TEMPERATURE STABLE MICROWAVE HYBRID GARNETS  
Gordon R. Harrison and L. R. Hodges, Jr.  
Sperry Microwave Electronics Company
150. MAGNETIC PROPERTIES OF YTTRIUM-GADOLINIUM-ALUMINUM-IRON GARNETS  
E. A. Maguire and J. J. Green  
Raytheon Company
151. NICKEL ALUMINUM GALLIUM FERRITES FOR USE AT HIGH SIGNAL LEVELS  
J. W. Nielsen and Joel Zneimer  
Airtron Division of Litton Industries
152. RESONANCE PROPERTIES OF SINGLE CRYSTAL HEXAGONAL FERRITES  
C. R. Buffler  
Varian Associates
153. HEXAGONAL FERRITES FOR USE AT X- TO V-BAND FREQUENCIES  
G. P. Rodrigue and J. E. Pippin  
Sperry Microwave Electronics Company
154. SPIN WAVE EXCITATION IN PLANAR FERRITES  
I. Bady  
United States Army Signal Corps Research and Development Laboratory  
E. Schlömann  
Raytheon Company
155. INVESTIGATION OF SPIN-WAVE INTERACTIONS BY THE PARALLEL PUMPING TECHNIQUE  
J. J. Green and E. Schlömann  
Raytheon Company
156. SUBSIDIARY ABSORPTION EFFECTS IN FERRIMAGNETICS  
J. H. Saunders and J. J. Green  
Raytheon Company
157. HIGH-POWER CHARACTERISTICS OF SINGLE-CRYSTAL FERRITES WITH PLANAR ANISOTROPY  
Samuel Dixon, Jr.  
United States Army Signal Research and Development Laboratory
158. THE EFFECT OF GROWTH CONDITIONS AND SUBSEQUENT THERMAL AND MECHANICAL TREATMENT ON THE FERRIMAGNETIC RESONANCE LINEWIDTH OF LITHIUM FERRITE SINGLE CRYSTALS  
J. W. Nielsen, D. A. Lepore, J. Zneimer, and G. B. Townsend  
Airtron Division of Litton Industries
159. THE GROWTH OF YTTRIUM-IRON GARNET ON A SEED FROM A MOLTEN SALT SOLUTION  
R. A. Laudise, R. C. Linares, and E. F. Dearborn  
Bell Telephone Laboratories, Incorporated
160. COBALT FERRITE CRYSTAL GROWTH FROM THE TERNARY FLUX SYSTEM  $Na_2O-CoO-Fe_2O_3$   
W. Kunmann and A. Wold  
Lincoln Laboratory  
E. Banks  
Polytechnic Institute of Brooklyn
161. GROWTH OF SINGLE-CRYSTAL HEXAGONAL FERRITES CONTAINING  $Zn$   
Arthur Tauber, Robert O. Savage, Richard Gambino, and Charles Whinfrey  
United States Army Signal Research and Development Laboratory



# SESSION I

## GENERAL SESSION

L. R. BICKFORD, JR., Presiding

### 1. MODIFICATION OF SCREW PIN STRUCTURE DUE TO ANISOTROPY ENERGY AND APPLIED FIELD (Invited)

T. NAGAMIYA

Department of Physics  
Osaka University  
Osaka, Japan

The screw structure of spins in layer crystals, first predicted by Yoshimori, has found its example in quite a number of substances, notably in rare earth metals such as Dy, Ho, Er, Tm, and Tb. The spin structures found in these rare earth metals are, however, not the simple screw structure and they change with temperature. For example, the spin structure in Er is such that the spin vectors of the consecutive hexagonal layers rotate on a narrow cone at temperatures below 20°K, rotate on a plane oblique to the hexagonal axis between 20°K and 52°K, and between 52°K and 80°K (the Néel temperature) they distribute sinusoidally along the hexagonal axis showing no regularly arranged component perpendicular to this axis. Such strange behavior can be understood as being due to the effect of the anisotropy energy, without which the spin system would have shown a simple screw arrangement. A detailed theory, including the consideration of different anisotropy energies for the series of rare earth metals, has been developed by Miwa and Yosida (Tokyo) and Kitano (Osaka) to account for the observed results. It explains all the essential features of the spin structure in rare earth metals, except a rather rapid change of the period of spin arrangement with temperature observed for Dy, Ho, and Er in certain temperature ranges. (Elliott and Kaplan have developed similar theories independently but in less detail.)

Application of a magnetic field also modifies the spin structure. This was considered in detail by the present writer (independently of Herpin and Mériel and Enz, but more extensively and rigorously) for a simple system at absolute zero in which the plane of the layers is stabilized by the anisotropy energy of uniaxial symmetry and a screw is realized in the absence of external field. If the anisotropy energy is large, the spin vectors remain in the plane with increase in the field applied within the plane, but the screw structure changes abruptly to a sine wave structure whose amplitude decreases with increasing field and finally vanishes above a certain critical field (spins becoming parallel to the field). This occurs, however, when the turn angle of the screw is not in the neighborhood of 90° and 180°. If the turn angle is in the neighborhood of 90° or 180°, transition first occurs to an antiferromagnetic state and then to a sine wave state. In the case of weak and intermediate anisotropy energy, there is a range of field in which the spin vectors rotate on an elliptic cone whose axis is along the field direction and whose ellipticity increases with increasing field, finally the cone becoming planar and the spin arrangement melting into the sine wave one.



## 2. MAGNETIC DISORDER AS A FIRST ORDER PHASE TRANSITION (Invited)

C. P. BEAN and D. S. RODBELL  
General Electric Research Laboratory  
Schenectady, New York

The exchange interaction that gives rise to magnetism depends upon interatomic separation. If the lattice is deformable, then a spontaneous distortion will occur in the magnetic state. We have calculated, in the molecular field approximation, the properties of a system in which the exchange energy dependence is given by  $T_C = T_0 (1 + \beta (v - v_0)/v_0)$ .  $T_C$  is the Curie temperature appropriate to a lattice volume  $v$  while  $v_0$  is the equilibrium volume in the absence of magnetic interactions. The course of the magnetization with temperature of such a system depends upon the steepness,  $\beta$ , of the exchange interaction dependence on interatomic distance, the compressibility,  $K$ , and upon  $T_0$ . The behavior may be the usual second order transition to paramagnetism, but it can in fact become a first order transition with the properties usually associated thereto, e.g., latent heat and discontinuous density change. We define a parameter as

$$\eta = 40 KkKT_0\beta^2 [j(j+1)]^2 / [(2j+1)^4 - 1]$$

in which  $N$  is the number per unit volume of magnetic ions of angular momentum  $j\hbar$  while  $k$  is the Boltzmann constant. With  $\alpha_1$  the lattice thermal expansion coefficient,  $P$  the externally applied pressure, then for  $\eta > 1 + \beta (\alpha_1 T_C - PK)$  the transition is of first order. We have also calculated the "phase diagram" within the framework described above and have determined the boundaries in the  $(P, T)$  plane between ferro-, antiferro- and paramagnetism. The theory will be compared to some real materials.

## 3. HIGH MAGNETIC FIELD RESEARCH (Invited)

BENJAMIN LAX  
Lincoln Laboratory & National Magnet Laboratory\*  
Massachusetts Institute of Technology  
Cambridge 39, Massachusetts

The use of high magnetic fields as a research tool for a wide variety of physical phenomena is clearly recognized today and the subject of an International Symposium at Cambridge this fall. Another important step in promoting research with the help of large magnetic fields has been the sponsorship of the M.I.T. National Magnet Laboratory by the Air Force. This paper will review the highlights of the conference which included papers on research in plasma physics, low temperatures, solid state and the latest developments for generating high magnetic fields. The plans and objectives of the National Magnet Laboratory and description of the physical facilities will be presented. In addition, a brief review will be given of a number of experiments already performed in the existing Magnet Laboratory at M.I.T.

\*Supported by the Air Force through the Air Force Office of Scientific Research.

## 4. SUPERCONDUCTING MATERIALS AND HIGH MAGNETIC FIELDS (Invited)

J. E. KUNZLER  
Bell Telephone Laboratories, Incorporated  
Murray Hill, New Jersey

The observation that superconducting  $Nb_3Sn$  can sustain current densities of  $150,000 \text{ amp/cm}^2$  in magnetic fields exceeding 88 kgauss has stimulated extensive studies of several potential materials of interest for magnet construction. The feasibility of building a superconducting magnet that operates with characteristics consistent with measurements made on samples of the winding material has been demonstrated using a Mo-Re alloy. Nb-Zr alloys are currently the most likely material for superconducting magnets operating at fields as large as 80-100 kgauss, while  $Nb_3Sn$  "wire" shows promise as magnet material for fields of at least 200 kgauss. Information resulting from investigations of these materials will be summarized and the evidence for associating the "hard" superconducting properties of high-field superconductors with dislocations will be reviewed.

## 5. THE STATE OF THE ART OF MAGNETIC MEMORIES (Invited)

Q. W. SIMKINS  
International Business Machines Incorporated  
Poughkeepsie, New York

A broad range of magnetic memories finds extensive use in today's data processing equipment. The most significant factors in evaluating a memory for a given application are reliability, cost, size, speed and function. Ferrite core memories with capacities of  $10^3$  to  $10^6$  bits and with cycle times as short as  $0.5 \mu\text{sec}$  are in use. Recent developments in this area, including partial switching, 2 cores/bit, and new core fabrication techniques, will be discussed.

There have been extensive development efforts recently in many forms of magnetic metal film devices. Many geometries, modes of operation, fabrication techniques, substrate materials, and film compositions are being used. A comparison of some of the resulting devices will be made, and the effects of the factors listed above will be discussed. The application of metal film memory devices will be considered and a comparison drawn with ferrite devices.

Techniques for achieving special purpose functions, such as nondestructive readout, read only and associative memory using magnetic elements, will also be described and compared.



## SESSION IIA

# MAGNETIC DEVICES

W. L. SHEVEL, JR., Presiding

### 6. PROPERTIES OF MAGNETIC FILMS FOR MEMORY SYSTEMS (Invited)

E. M. BRADLEY

International Computers and Tabulators (Engineering) Ltd.

Stevenage, Hertfordshire, England

It is now well known that magnetic films can be deposited by vacuum evaporation in which the anisotropy energy expressed as a function of  $\theta$ , the angle between the easy axis and the magnetisation, is given by:  $E = K \sin^2 \theta$  and further that the reversal process can be a coherent rotation of electron spins.

The techniques of using this type of material in a computer storage matrix are reviewed. It is pointed out that the square hysteresis loop which is exhibited in the easy direction is very difficult to use as the basis for a matrix selected storage element; however several schemes for selection in which one of the selection lines applies a field along the easy axis and the other along the hard axis have been proposed. These have the advantages of very fast switching times and large tolerances in the selection fields and/or film parameters.

It has thus been possible to define the required properties of the magnetic film for use in matrix storage devices, and the effects of the process of deposition on these properties is discussed in detail.

It is shown that the addition of a small quantity of cobalt typically 3 per cent to the nickel iron alloy of approximately 80/20 composition gives a film whose properties are a marked improvement on the binary nickel iron alloy. It is shown how the  $H_K$  parameter varies with substrate temperature and cobalt content for the ternary alloy and that it is important that the film be of such composition that it is not magnetostrictive.

The compositions for zero magnetostrictive Ni Fe Co alloys are given. It is shown that the easy axis angle dispersion over a large area film deposited on an aluminum plate (4 in. x 3 in.) is a function of the processing conditions and in particular the variation of angle dispersion with cobalt content in the range 0 - 5 per cent is given together with the effect of substrate temperature, film thickness, vapour beam angles of incidence. Finally the variation of easy direction coercivity, which controls the tolerances in the digit current in a matrix device, with film thickness is discussed.

### 7. MAGNETO-OPTIC READOUT FOR COMPUTER MEMORIES\*

R. L. CONGER

United States Naval Ordnance Laboratory  
Corona, California

Experimental measurements have shown that non-destructive magneto-optic readout from magnetic film computer elements by means of the longitudinal Kerr effect, can operate at rates of at least one million bits per second. If the steady light source of the magneto-optic apparatus is sufficiently bright, a signal to noise ratio of ten or better can be obtained from the photo multiplier readout tube. Since the reversal time of thin ferromagnetic films is less than a microsecond, writing speed can be as fast as readout. Information readout from a magnetic film can be obtained by scanning with a high intensity light spot. Spurious variations in surface reflectivity of the films are sufficiently small, so that parts of the film magnetized in opposite directions can be clearly determined.

Experiments with an electronic strobing magneto-optic method make it possible to observe the details of the magnetization reversal process in very small parts of a film with a time resolution of the order of a nanosecond. Since this strobing process involves sampling and integration, the information band width is too small to be used for digital computer readout. However, this method is useful in studying fine details of the magnetization reversal process. Measurements show that the reversal is repeatable in the different parts of the film.

\*Work supported by the Navy Bureau of Weapons.

### 8. MAGNETO-OPTICALLY MEASURED HIGH SPEED SWITCHING OF SANDWICH FILM ELEMENTS

J. C. SUITS and E. W. PUGH

IBM Research Center  
Yorktown Heights, New York

To achieve high packing density and low drive currents, individual thin film memory elements are being designed with smaller and smaller areas. This in turn requires that they be made correspondingly thinner in order to maintain low self-demagnetizing fields. A reduction in linear dimensions by a factor of  $n$ , therefore, results in an  $n^2$  reduction in integrated output signal. Sandwich elements, in which two films magnetized in a head-to-tail fashion are placed on either side of a drive line, have been proposed as a means for utilizing thicker films without having excessive internal demagnetizing fields.<sup>1</sup> In this paper a magneto-optical method for making high speed switching measurements on such elements is described together with switching results on 1 mm diameter films *before* and *after* being incorporated into the proposed sandwich configuration.

Sandwich elements were fabricated by first evaporating a row of permalloy bits on glass, followed in succession by a layer of silicon oxide for electrical insulation, an aluminum conducting strip, more silicon oxide and finally a second row of permalloy bits on top of the first.



Switching curves were taken after each step in the fabrication by pulsing the bits either with an external strip line or with the evaporated conducting strip. Read-out was obtained by means of the Kerr magneto-optical effect. The photomultiplier output was passed through a narrow band-pass filter tuned to the strip line pulse repetition rate. This technique allowed switching measurements to be made on one mm diameter bits as thin as 150 Å with switching times as short as 10 nanoseconds.

The following results were noted: (1) a considerable improvement in the remanence of thick bits was observed when the bits were in the sandwich configuration, (2) when the demagnetizing fields were taken into account, the domain wall coercive force for single bits was found to be independent of thickness, (3) the switching speed of single bits was markedly dependent upon film thickness even when all bits were thin enough to remain fully saturated in their remanent state, (4) the effect of the nearby conductor (the 6,000 Å aluminum strip) was to reduce the switching speed but only by a negligible amount, and (5) in many cases a substantial improvement in switching speed was observed for the sandwich bits as compared with the single bits.

<sup>1</sup>See survey article by A. V. Pohm and E. N. Mitchell, IRE Transactions on Electronic Computers, Sept. (1960).

## 9. SWITCHING PROPERTIES OF MULTILAYER THIN FILM STRUCTURES

A. KOLK, L. DOUGLAS, and G. SCHRADER  
The National Cash Register Company  
Hawthorne, California

Thin films of magnetic materials which differ widely in their coercivities have been found to display a square hysteresis loop with no discontinuities when deposited in direct contact with one another. This is in contrast to the discontinuous hysteresis characteristic normally found in gross heterogeneous structures.

The total saturation flux was found to be approximately the sum of the expected values of flux for each of the individual layers, but the coercive force could be adjusted to values which were not obtainable with the separate materials of an equivalent thickness.

The structures usually consisted of a layer of material possessing a moderate coercivity (15 to 30 oe) and a second layer of 80 Ni, 20 Fe permalloy. The coercivity of the composite was found to be between the coercivities of the individual components at the total thickness of the composite.

The switching response to a step function magnetic drive has been studied. The switching data indicated a highly complicated switching mechanism, but it was observed that the inherently high switching speeds of the individual materials were preserved.

The data indicated that there were changes in switching mechanism as the amount of overdrive was increased. It was observed, however, that even at low values of overdrive, switching coefficients ( $S_w$ ) were obtained which were substantially lower than those normally associated with domain wall motion.

The use of pulse sampling techniques made it possible

to examine the switching response of the films studied in much more detail than has heretofore been possible.

## 10. MAGNETOSTATIC INTERACTIONS BETWEEN THIN MAGNETIC FILMS\*

HARRISON W. FULLER and DONALD L. SULLIVAN  
Computer Products Division  
Laboratory for Electronics, Incorporated  
Boston, Massachusetts

This paper discusses some magnetostatic interactions between two thin films that are separated by a small distance between parallel film planes.

The external field of a domain wall in one film is calculated using a simple wall model, and the effect of this field on a second adjacent film is determined. The conditions are investigated under which the wall field of the first film together with an external field can cause nucleation of a domain in the second film.

The interaction energy between a movable wall in one film and a stationary wall or a linear imperfection in a second adjacent film is calculated using simple models. The equilibrium position of the movable wall as a function of an externally applied field is then determined. Means for electrically modifying the degree of interaction are investigated.

Experimental results are presented illustrating the aforementioned interactions. The nucleation of long narrow domains in one film is achieved by the use of a domain wall, a scratch imperfection or a film edge in a second adjacent film. The influence of a stationary wall in one film on the controlled wall motion of a wall in a second film is demonstrated. A comparison of theory and experiment is made.

Mention is made of how the subject interactions might be utilized in new information storage devices.

\*This work was supported in part by contracts with the Electronics Research Directorate of the Air Force Cambridge Research Laboratory, and the Information Systems Branch of Office of Naval Research.

## 11. PERMALLOY-SHEET TRANSFLUXOR-ARRAY MEMORY

G. R. BRIGGS and J. W. TUSKA  
RCA Laboratories  
Princeton, New Jersey

Annealed, molybdenum-permalloy sheets in the thickness range of 1/8 - 1 mil have been photoetched to form transfluxor arrays utilizing the inhibited-flux<sup>1,2</sup> mode of storage. This mode permits a fast memory cycle and yields useful storage characteristics despite non-ideal hysteresis loop properties of the storage medium and despite eddy current effects.

Operating results will be presented for single elements, of the permalloy-sheet transfluxor type, switched in the time range of 20 nsec-10  $\mu$ sec. Measurements have been obtained using both word-organized and X - Y



coincidence methods of selection. These include: a minimum  $S_w = 0.16$  oe  $\mu\text{sec}$  (1/8 mil thick element switched in 30 nsec), ONE- to ZERO-state information signal ratios of 5 to 16, and an operating temperature range of  $-70^\circ\text{C}$  to  $+150^\circ\text{C}$  without drive-current or strobe-point compensation.

Small-signal properties of the output circuits of single elements have been studied. This study indicates that more than 1000 elements can be sensed per sense winding without a serious attenuation or a delay problem.

Nondestructive sensing has been demonstrated, utilizing reading-current pulses of shorter-than-standard duration. For a 1 mil thick element, nondestructive sensing is possible with 0.8  $\mu\text{sec}$  reading pulses.

The photoetching process for forming permalloy sheet transfluxor arrays will be described. This process is capable of high dimensional precision. Experimental arrays fabricated include 128-element sheets for X-Y coincidence use and 1152-element sheets for word-organized application. The latter permit a memory bit density of 250,000 per in<sup>3</sup>.

Methods of coating and heat-treating the arrays have been found for controlling mechanical stressing of the permalloy to produce uniform magnetic properties. Using these methods, a ONE-state output uniformity of  $\pm 3$  per cent has been obtained, for all the elements of typical experimental 128-element arrays fabricated using 0.5 mil thick permalloy.

A photofabricating process for forming windings on the permalloy memory sheets has been developed also and will be described. Because of the thinness of the sheets, the windings can be formed linking the element apertures. This process has been applied successfully to small experimental arrays.

<sup>1</sup>J. A. Baldwin, Jr. and J. L. Rogers, J. Appl. Phys. **30**, 585 (1959).

<sup>2</sup>W. W. Lawrence, Jr., Proc. Eastern Joint Computer Conference, 101 (1956).

## 12. PROPOSAL FOR AND DEMONSTRATION OF MAGNETIC DOMAIN-WALL STORAGE AND LOGIC

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If a ferromagnetic domain configuration is stable, so is its negative, and this fact forms the basis of the usual magnetic representation of binary digital information. However, if a domain configuration is stable, so is the one which corresponds to a reversal of the sense of rotation of the magnetization  $M$  within the walls separating the domains; hence, alternatively, the sense of wall rotation can be used to represent binary information. The two methods of representation are essentially different since different aspects of spatial symmetry are involved. A practical method of utilizing wall-rotation information coding can be effected by means of strips of thin magnetic film having a uniaxial magnetic easy axis perpendicular to the long dimension of the strip and nearby current conductors that generate magnetic fields in specified directions at specified

times and places. A shift register consists of a regular up-and-down domain pattern with  $M$  perpendicular to the long strip axis; no information is contained in the domain pattern itself, which only serves to spatially separate the information-bearing walls. Shifting is accomplished by two-phase clock pulses on a pair of shifting windings. Readout requires the use of the operation of conditional erase, accomplished as follows: if two walls of the same sense are brought together, they form a double wall which can subsequently be separated into two walls; if the walls are of the opposite sense, they will destroy one another on being brought together. Thus, erasure occurs on condition that two walls have opposite sense. Read-out then consists of conditional erase between a data wall and a wall of known sense read-in for the purpose of read-out. Fan-out can be achieved with a Y strip. A wall is moved from the single to the double strip. Furthermore, if data walls from two independent converging strips are juxtaposed in a third strip, the logical operations AND and OR can be performed by appropriate use of the operations of conditional erase and the additional operation of unconditional erase; unconditional erase erases two walls regardless of sense. Finally, a method of complementing information can be conceived by considering the details of flux closure within a domain wall.

Preliminary experiments on the various logical and storage operations have been performed by manually controlling the current flowing through an appropriate configuration of wires placed closely under a thin strip of evaporated Permalloy film, and the sequence of events was observed on the top side of the film using the Bitter technique. Walls of known sense were read into the films with sets of parallel wires that make angles between  $50^\circ$  and  $75^\circ$  with the strip of film, the smaller angles being required for the thinner films. It was found that the success of the operation of conditional erase depends strongly on the uniformity and thickness of the films, with very thin films around 50 A thick giving the widest operating margins. The field required to erase a double wall formed from two walls of the same sense is, for a reasonably uniform film 50 A thick, about 20 oe, which is at least 9 times that required to bring together and erase a pair of walls of opposite sense. The same wire configuration used for shifting is employed without difficulty to fan out information at a junction.

\*Operated with support from the U. S. Army, Navy, and Air Force.

## 13. THRESHOLD PROPERTIES OF PARTIALLY SWITCHED FERRITE CORES

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A switching threshold relaxation effect has been observed which is detrimental to high speed selection of partially switched (time limited switching) ferrite memory cores. This paper discusses these threshold effects as determined by measurements on cores of one to five oersted coercivity. Two aspects of switching threshold are



apparent: a static threshold ( $H_0$ ) which is effective for times long after switching, and a relaxation effect which is a reduction in threshold immediately after switching. This reduction is marked and limits the amplitude of digit disturb-field that the core may withstand to a small fraction of the coercive force. A first approximation to the threshold dependence on time may be given as:  $H_t = H_0 [1 - \exp(-pt)]$ , where  $1/p$  is the relaxation time, for  $t > 0$ . This representation is reasonably accurate for most ferrite cores, although a sum of exponentials is necessary for an improved description.

The ratio of static threshold to coercivity ( $H_0/H_c$ ) is influenced by duration and polarity (relative to switching field) of the disturb field, amplitude of switching field, flux state, ambient temperature, and composition. The static threshold at the 50% flux state resulting from a 10 nsec switching field may be only half the value of the threshold after switching to the 50% state with a 2  $\mu$ sec field. The relaxation time depends on the above mentioned parameters (except relative polarity) and, in addition, is strongly linked with the coercivity of the sample, the lower coercivity samples having the longer relaxation times. Measurements at ambient temperatures ranging from liquid nitrogen temperature up to near the Curie temperature ( $\sim 250^\circ\text{C}$ ) show the product of coercivity and relaxation time to be nearly constant. For a given core, relaxation time is maximum at the 40-50% flux state and approaches zero at the major loop remanent flux states. Numbers for a representative copper manganese ferrite core are 1.6 oe coercivity, 0.22  $\mu$ sec relaxation time and 1.3 oe static threshold at the 50% flux state obtained with a 0.2  $\mu$ sec switching field. Measurements of all the above-mentioned dependencies are reported in this paper.

#### 14. FLUX REVERSAL IN FERRITE CORES UNDER THE EFFECT OF A TRANSVERSE FIELD\*

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It is known that flux reversal in ferrites involves three switching mechanisms, namely, domain wall motion, non-uniform rotation and uniform rotation. This paper demonstrates that a steady transverse field in conjunction with set and reset pulses is capable of separating these mechanisms into three regions of flux dispersion. Each region is designated by flux components  $\phi_1$ ,  $\phi_2$  and  $\phi_3$ , with corresponding characteristic transverse fields  $H_1$ ,  $H_2$  and  $H_3$ . The output waveform of the longitudinal sense signal during flux reversal is characterized by its first peak, second peak and the long tail. The fifty per cent value of the first and second peaks occur at the same characteristic transverse fields  $H_1$  and  $H_2$ , respectively. The experimental results agree with theoretical predictions in which less flux is switched by domain wall motion at higher drive currents.

The demagnetizing field in the transverse direction depends strongly on the thickness of the ferrite core. The characteristic transverse fields  $H_1$ ,  $H_2$ , and  $H_3$  increase to higher values as the thickness decreases. If the transverse field affects appreciably the distribution of flux

reversal mechanism in favor of rotation over domain wall motion, the first and second peaks should increase at low transverse fields. Experimentally, the first and second peaks remain practically constant over region 3. Furthermore, the characteristic transverse fields are found to be independent of the amplitude of circumferential drive. Therefore, the transverse field acts to eliminate the slow reversal mechanism component first but does not change the mechanisms appreciably. A proper transverse field has the effect of speeding up the switching time of a ferrite core without sacrificing the amplitude of the output signal due to the fast reversal components.

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#### 15. APPROXIMATE SOLUTION OF THE EQUATIONS OF MAGNETIZATION REVERSAL BY COHERENT ROTATION\*

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Starting with the modified Landau-Lifshitz equation, Gyorgy and Hagedorn<sup>1</sup> have obtained a set of three non-linear differential equations which describe the coherent rotational process of flux reversal in square-loop ferrite cores. In the formulation of these equations the effects of crystalline anisotropy have been neglected. The effective field was assumed to be the vector sum of the applied, demagnetizing and exchange fields; however, it was shown that the exchange field can be neglected.

This paper presents an approximate solution of these equations for  $\alpha > 0.1$ , where  $\alpha$  is the phenomenological damping constant. It is shown that for  $\alpha > 0.1$

$$m_r = \frac{H_a}{4\pi M_S \alpha} \left(1 - e^{-4\pi\alpha\tau}\right) \operatorname{sech}$$

$$\left[ \frac{H_a}{M_S} \tau \left(\alpha + \frac{1}{\alpha}\right) + \tanh^{-1} m_i - \frac{H_a}{4\pi M_S \alpha^2} \left(1 - e^{-4\pi\alpha\tau}\right) \right]$$

and

$$m_z = \tanh \left[ \frac{H_a}{M_S} \tau \left(\alpha + \frac{1}{\alpha}\right) + \tanh^{-1} m_i - \frac{H_a}{4\pi M_S \alpha^2} \left(1 - e^{-4\pi\alpha\tau}\right) \right]$$

where  $H_a$  is the applied field

$M_S$  is the saturation magnetization

$$m_i = \frac{M_i}{M_S} \text{ where } M_i = M_z \Big|_{t=0}$$

$$\tau = \frac{\gamma M_S t}{1 + \alpha^2} \text{ where } \gamma \text{ is the gyromagnetic ratio.}$$

For  $\frac{H_a}{4\pi\alpha^2 M_S} \ll 1$  these equations lead to the important results that



$$H_a T_s = - \frac{2\alpha}{\gamma} \tanh^{-1} m_i$$

and

$$\left( \frac{dM_z}{dt} \right)_{\max} = \frac{\gamma}{\alpha} M_s H_a$$

where  $T_s$  is the time to switch a fixed percentage of the total flux (usually 90 per cent) and  $\left( \frac{dM_z}{dt} \right)_{\max}$  is the maximum rate of change of magnetization. The maximum output voltage can be found from  $\left( \frac{dM_z}{dt} \right)_{\max}$  as

$$\left( \frac{d\phi}{dt} \right)_{\max} = (e_o)_{\max} = 4\pi A_T \frac{\gamma}{\alpha} M_s H_a \text{ abvolt}$$

where  $A_T$  is the total cross sectional area of the core.

Thus it is shown that  $T_s$  is inversely proportional to  $H_a$  and the  $\left( \frac{dM_z}{dt} \right)_{\max}$  is linearly dependent on  $H_a$ .

\*This investigation was supported in part by the National Science Foundation.

<sup>1</sup>E. M. Gyorgy and F. B. Hagedorn, "Uniform Rotational Flux Reversal of Ferrite Toroids," J. Appl. Phys. 30, 1368-1375 (1959).

## 16. ALL MAGNETIC LOGIC ELEMENTS USING STRAINED PERMALLOY WIRE

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A system for performing digital logic which employs permalloy wire, solenoid windings, resistors, and a two phase current pulse source has been built and operated successfully. The re-entrant hysteresis loop of a torsionally strained permalloy wire provides sufficient power gain for a fan out of three. A simple balanced input circuit assures unidirectional information flow and also provides means for obtaining certain logic functions.

Each logic element consists of bias and read solenoids 0.6 in. long wound in bifilar fashion, one or several .05 in. solenoids to which logic inputs are applied, an output resistor, and a permalloy wire core. A buffer zone, the magnetization of which is controlled by the read current, separates adjacent logic elements to minimize interactions between elements.

The logic elements are arranged in groups A and B and are supplied by a two phase source so that in phase 1 group A receives a bias pulse and its logic inputs and B receives a read pulse to produce logic outputs. In phase 2, group A receives a read pulse and B receives a bias pulse and the logic outputs from A. The bias pulse supplies most of the energy needed to switch the core material to the 'one' state of magnetization. The logic input must supply only the energy needed to assist the bias in the reversal of a .05 in. section of the permalloy wire; the bias pulse then

completes the switching of the element. If a logic input is not present, the bias cannot start the switching process. The read currents supply all of the energy to switch the cores to the 'zero' state of magnetization and provide sufficient output to control several following elements (the bias coil is used as the output winding).

The balanced input winding consists of two .05 in. solenoids spaced 0.1 in. apart and connected in series opposition. Due to the cancellation of the voltages produced by each coil, no voltage appears at the input terminals during read-out, and unidirectional information flow is assured. The spacing between the two input coils permits the winding that is properly poled to assist the bias and start the switching of the element in spite of the presence of the opposing field of the balancing input coil.

Logic functions are readily obtained by using multiple input windings either superposed or adjacent. A simple *exclusive or* circuit is formed by two superposed input windings of the balanced type so arranged that when both inputs are present their magnetic fields cancel, but when either input is present alone, switching occurs. Because of the uniformity of the components used, it has been found that such cancellation is reliable.

Majority logic elements where threshold type operations are necessary have also proven successful. The threshold can be provided by the starting field required to nucleate the switching process or by a bias current in an additional solenoid.

The magnetic material used was a 5-79 permalloy wire 0.003 in. in diameter placed under a torsional strain of  $2\pi$  radians per inch. Eddy currents in the wire limit the switching to  $15\mu s$  or  $50\mu s$  depending on the magnitude of the bias current. The read out occurs in  $3\mu s$ .

These elements have been used to construct flip-flops, binary counters and a full serial adder. The adder required only three logic elements and two delay elements. The circuits operated successfully when the pulse currents from the power supply were varied by  $\pm 20\%$ . The simplicity and expected reliability of these elements and the ease with which logic functions can be realized should make them competitive with other devices where the slow operating speed is not objectionable. Because an open flux structure (as opposed to a toroidal structure) is used in these elements, it is believed that similar techniques will have applications in circuits employing evaporated magnetic films.

## 17. A PERMANENT MAGNET TWISTOR MEMORY ELEMENT OF IMPROVED CHARACTERISTICS

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The basic twistor memory element employed in permanent magnet twistor memories has been redesigned to obtain higher and more uniform outputs at a faster switching speed. The desired characteristics were obtained by:

1. Enhancing the drive field by halving the bit length. The same drive current applied to a single turn solenoid of half the original bit length produces an approximately doubled drive field at the twistor bit.



2. Replacing the copper return wire in the twistor pair with a reverse wrapped twistor wire. Thus the twistor bit contains the same amount of magnetic material and upon interrogation produces two series-aiding outputs. (A balanced transmission line is also obtained.)

3. Annealing the twistor wire to lower the coercive force and to increase the squareness ratio of the molypermalloy tape. Stability of the magnetic material is also achieved.

The chief result of these and other changes is that for the same solenoid read current, the net switching field at the bit is 2.5 times larger than in earlier designs.

A 26000 bit card changeable permanent magnet twistor memory utilizing the new element design has been constructed. The twistor wires employed in this memory had coercive forces with a spread of  $\pm 30\%$ . This variation in parameters was virtually obscured by the high driving field and highly uniform switching outputs which resulted. If twistor wires with very low coercive forces are used, the magnetic fields due to information storage and such stray fields as the earth's magnetic field may partially switch the twistor bit and produce either a low one or a false zero. Therefore it is necessary to set a lower limit on the coercive force of acceptable wire. The effects of interaction fields are further reduced by employing appropriate shielding techniques in the memory plane.

In comparison to previously reported permanent magnet twistor memories, the new design is characterized by twice the speed (less than 2.5 microseconds cycle time), 1.5 times the bit density, 2.5 times the output voltage level, 2.5 times the signal to noise ratio, and 7 times the signal discrimination area.

## SESSION IIB INTERNAL FIELDS

P. J. WOJTOWICZ, Presiding

### 18. NUCLEAR RESONANCE IN FERROMAGNETIC ALLOYS (Invited)

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The internal magnetic fields,  $H_i$ , seen at the nuclei in ferromagnetic alloys are investigated using nuclear magnetic resonance.  $\text{Co}^{59}$  resonances in Co-rich alloys (Co-Fe, -Ni, -Cu, -Cr, -Mn, -Al) have fine structures which depend on the kind and the concentration of impurity metals. Similar structures are found for the  $\text{Fe}^{57}$  resonance in Fe-rich alloys. The  $\text{Co}^{59}$  resonance is also observed in Fe-rich Co-Fe alloys, and it is found that  $H_i$  seen at the Co site is lower by about 50 koe than that at the Fe site. The  $\text{Co}^{59}$  line width is about 400 kc for (Fe + 1% Co) alloy.  $\text{Cu}^{63}$  and  $\text{Cu}^{65}$  resonances are found in ferromagnetic Fe and Co alloys.  $H_i$  at the Cu nucleus is 217.7 koe at  $0^\circ\text{C}$  for the Fe-Cu alloy and 157.5 koe at  $9^\circ\text{C}$  for Co-Cu alloy. The temperature and the pressure dependences of these lines have been studied.

### 19. CRYSTALLINE ELECTRIC FIELDS IN SPINEL-TYPE CRYSTALS

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Because of the important effects of crystalline electric fields on ferrimagnetic properties, i.e., magnetocrystalline anisotropy, magnetostriction, resonance linewidth and  $g$ -value, it is apparent that the determination of such fields in spinel-type crystals would be useful in the interpretation of these properties. In this paper, the first electron paramagnetic resonance (EPR) measurements of crystalline electric fields acting on ferric ions in the spinel-type crystals is reported. These measurements include the first studies of "order-disorder" effects on paramagnetic resonance in any material. Specifically, the EPR measurements were obtained for the ferric ion in "ordered" and "disordered" monocystals of lithium aluminate ( $\text{Li}_{0.5}\text{Al}_{2.5}\text{O}_4$ ) which is isomorphous<sup>1</sup> with lithium ferrite ( $\text{Li}_{0.5}\text{Fe}_{2.5}\text{O}_4$ ). In the "disordered" aluminate, the EPR linewidths in general were found to be very broad (300 gauss), whereas in the "ordered" aluminate the linewidths were comparatively narrow (15 gauss).

The broadness of the EPR linewidths in the



"disordered" aluminate prevented quantitative determination of the crystalline electric fields. From comparison of the "ordered" and "disordered" linewidths, we deduce that there may be large fluctuations in the crystalline electric fields in disordered spinels. Such fluctuations may be relevant to the problems of the large ferromagnetic resonance linewidths observed in disordered spinels.

In the "ordered" aluminate, the observed EPR spectrum was found to correspond to that of four ions per unit cell having trigonal site symmetry along the [111] crystallographic directions. This symmetry is in accord with the symmetry of the tetrahedral site<sup>1</sup> so that it is concluded that the ferric ions have a substantial preference for the tetrahedral sites. The spectrum corresponding to the octahedral sites, which have twofold distortion axes along the [110] directions, was not observed in the aluminate. For the tetrahedral site the measured values of the crystalline electric field parameters  $|D|$  and  $|a-F|$  are  $0.104 \text{ cm}^{-1}$  and  $0.0166 \text{ cm}^{-1}$ , respectively, and the measured  $g$ -value is 2.006.

The relation of these and additional measurements of the crystalline electric fields to the interpretation of magnetocrystalline anisotropy in ferrites will be discussed.

<sup>1</sup>P. B. Braun, *Nature* 170, 1123 (1952).

## 20. THE AXIAL CRYSTAL FIELD IN CORUNDUM AND THE SUSCEPTIBILITY OF $\text{Ti}_2\text{O}_3$

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$\text{Ti}_2\text{O}_3$  shows an anomalously small susceptibility at low temperature,<sup>1</sup> orders of magnitude less than what one might expect from the unpaired  $d$  electron of  $\text{Ti}^{+3}$ . In addition, neutron diffraction data indicate a very small localized magnetic moment. It has been suggested that a strong direct covalent-type interaction between the  $\text{Ti}^{+3}$  ions pairs their spins, and results in the vanishingly small moment observed.<sup>2</sup> It should be noted that the sign of the axial crystal field in the corundum lattice is such as to leave as a ground state a nonmagnetic doublet for a single  $d$  electron such as is found in  $\text{Ti}^{+3}$ . This sign of the axial crystal field is deduced from paramagnetic resonance data of  $d^n$  ions in  $\text{Al}_2\text{O}_3$  and especially from recent work to be here reported on  $\text{Ru}^{+3}$  in  $\text{Al}_2\text{O}_3$ . If the  $\text{Ti}^{+3}$  were completely ionic, both  $g_{\parallel}$  and  $g_{\perp}$  would be equal to zero. However, paramagnetic resonance of  $\text{Ti}^{+3}$  in  $\text{Al}_2\text{O}_3$  has been observed<sup>3</sup> with  $g$  values of  $g_{\parallel} = 1.067$  and  $g_{\perp} \leq 0.1$ . For these values to be consistent with sign of the axial field it is necessary to assume an orbital reduction factor  $k = 0.5$ , i.e., complete covalency. This would then seem to support the covalent pair bond idea, in line also with the observed metallic nature of  $\text{Ti}_2\text{O}_3$ .

<sup>1</sup>P. H. Carr and S. Foner, *J. Appl. Phys.* 31, 344S (1960).

<sup>2</sup>J. B. Goodenough, *J. Appl. Phys.* 31, 359S (1960).

<sup>3</sup>L. S. Kornienko and A. M. Prokhorov, *JETP*.

<sup>4</sup>K. W. H. Stevens, *Proc. Roy. Soc.* 219, 542 (1953).

## 21. HYPERFINE FIELDS IN TRANSITION METAL AND RARE EARTH IONS

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Mössbauer and NMR measurements have now been made of the hyperfine fields in a wide variety of magnetic materials; in the few cases for transition metals in which the sign was determined the internal field was found to be negative, i.e., opposed to the direction of magnetization. However, for some ions of the first transition series and for almost all the rare earths the hyperfine fields are expected to be positive (but to date no such sign determinations have been made). This is so because of the large (and positive) contribution due to the orbital angular momentum of the unpaired (magnetic) electrons. For the rare earth ions the angular momentum of the  $4f$  electrons is almost completely unquenched (the reverse of the transition metal case) and this contribution to the field at the nucleus is much larger than the (negative) field from the polarization of the core  $s$  electrons via the Fermi contact term (except for  $\text{Gd}^{+3}$  and  $\text{Eu}^{+2}$ ). These matters are discussed on the basis of analytic Hartree-Fock calculations which were carried out for eight trivalent ions of the series and comparison is made with experiment. Also discussed is: (1) the relation of the spin orbit parameter calculated from the many-electron potential function to the result obtained from the usual  $\langle r^{-3} \rangle$  approximation and (2) the resulting effect of this on the accurate determination of nuclear magnetic moments which uses this information.

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## 22. HYPERFINE MAGNETIC FIELD AND ELECTRON SPIN RELAXATION IN THE $\text{Sm}^{+3}$ ION IN EUROPIUM IRON GARNET\*

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The rotation and the attenuation of the integral angular correlation of the 1415-122 keV  $\gamma - \gamma$  cascade in  $\text{Sm}^{152}$  following the  $K$  capture of  $\text{Eu}^{152}$  in polycrystalline samples of Europium iron garnet was measured with and without magnetizing field perpendicular to the plane containing the counters from  $-50^\circ\text{C}$  to above the Néel temperature. The consistent interpretation of these results requires the presence of time dependent hyperfine interactions of short relaxation or correlation times. The existence of these time dependent perturbations was confirmed by differential angular correlation experiments. The rotation of the angular correlation obtained below the Néel temperature with a weak magnetizing field perpendicular to the counter plane is due to the average uncompensated component of the effective hyperfine field  $H_{\text{eff}}^Z$  in the direction of the



magnetizing field. In order to obtain the correct value of  $H_{\text{eff}}^Z$  the effect of the time dependent perturbations on the rotation must be taken into account. The magnitude, sign and temperature dependence of  $H_{\text{eff}}^Z$  from  $-50^\circ\text{C}$  to the Néel point was in excellent agreement with the expectation value of  $H_{\text{eff}}^Z$  calculated from perturbation theory under the assumption that the electronic configuration of the  $\text{Sm}^{+3}$  ion is perturbed below the Néel temperature by the known magnetic exchange field  $H_{\text{exch}}$  acting on the  $\text{Eu}^{+3}$  ions in EuIG in the direction of the magnetizing field. In this calculation the ground state of the  $\text{Sm}^{+3}$  ion was given by Russell-Saunders coupling. Crystalline field and saturation effects were neglected and the magnetic hyperfine interaction Hamiltonian did not take into account the effect of exchange coupling between the 4f shell electrons and inner core s electrons. The excellent agreement between theory and experiment strongly indicates: (1) The electronic configuration of  $\text{Sm}^{152}$  following the K capture of  $\text{Eu}^{152}$  reaches that of the ground state of the  $\text{Sm}^{+3}$  ion during the times relevant in this experiment; (2) the hyperfine magnetic field in the  $\text{Sm}^{+3}$  ion in EuIG can be calculated satisfactorily from molecular field theory; (3) the indirect exchange coupled contribution to the magnetic hyperfine field in  $\text{Sm}^{+3}$  is very small compared with the direct contribution. The magnitude of the magnetic hyperfine field was calculated from the rotation data and was found to be  $4 \times 10^6$  oe at room temperature. Under the assumption that the contribution of the quadrupole hyperfine interaction in the time dependent perturbation could be neglected relative to the magnetic contribution, the electron spin relaxation time at room temperature was found to be  $4 \times 10^{-12}$  sec. The data is consistent with a temperature variation of the electron spin relaxation time which is inversely proportional to the absolute temperature over the experimentally determined temperature range.

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### 23. THE de HAAS van ALPHEN EFFECT IN ZINC MANGANESE ALLOYS

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Observations of the long period de Haas van Alphen effect in pure zinc and in a zinc alloy containing 0.01 atom per cent manganese have been made to determine whether the Fermi surface of the alloy changes in the temperature region where the zinc manganese alloy exhibits an electrical resistance minimum.

On the basis of a nearly free electron interpretation it has been shown that: (1) within the experimental error the Fermi surface of pure zinc and the 0.01 atom per cent zinc manganese alloy are identical; (2) the number of electrons added or subtracted from the conduction band on alloying cannot be greater than 1 electron per impurity atom; (3) if the resistance minimum is due to a change in the density of states in the conduction band, this change is less than 0.0015 per cent; (4) if the ionic state of the manganese ions changes in the temperature range of the resistance minimum then less than half of the ions are involved.

In order to obtain a consistent interpretation of the variation of the amplitude of the de Haas van Alphen oscillations in the alloy it was necessary to introduce a new term into the collision damping (Dingle) factor. This term has the form of an internal field with a value of approximately 25 koe at  $1.7^\circ\text{K}$ . It should be noted that an "internal field" of this order of magnitude is required by Bailyn for his theory of the thermal powers of alloys having a resistance minimum.

### 24. ANISOTROPY OF THE HYPERFINE INTERACTION IN MAGNETITE

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This paper describes effects of anisotropy of the hyperfine interaction on nuclear magnetic resonance in a ferromagnetic or ferrimagnetic material. It is shown how the line shape in a polycrystalline sample depends on whether the excitation is through domains or domain walls. The absorption spectrum of  $\text{Fe}^{57}$  in powdered magnetite ( $\text{Fe}_3\text{O}_4$ ) is observed and the results show that, in this case, the excitation is predominantly through domains. They also establish a value for the hyperfine anisotropy on octahedral sites.

For an octahedrally coordinated nucleus in magnetite, the nuclear precession frequency  $\nu$  is expected, phenomenologically, to have the form

$$\nu = \nu_{\perp} + (\nu_{\parallel} - \nu_{\perp}) \cos^2 \theta,$$

where  $\theta$  is the angle between the magnetization vector  $M$  and the trigonal axis of local symmetry, and  $\nu_{\parallel}$  and  $\nu_{\perp}$  are constants.

A powder sample presents a density  $\rho(\nu)$  of resonance frequencies because the values of  $\theta$  are distributed. One limiting case is that of random orientation of  $M$  with respect to the crystal axes. It leads to a line shape, well known in nonferromagnetic NMR and EPR, having a peak at  $\nu = \nu_{\perp}$  and a step discontinuity at  $\nu = \nu_{\parallel}$ . One might expect such a line shape from single domain particles in which the shape anisotropy dominates the magnetocrystalline anisotropy.

Other limiting cases for  $\rho(\nu)$  are presented by domain wall excitation. One finds characteristic line shapes having a peak at  $\nu = \nu_{\perp}$  and a peak at a second frequency  $\nu_a$  lying between  $\nu_a$  and  $\nu_{\perp}$ . The value of  $\nu_a$  depends on the angle between the local trigonal axis and the wall plane. These line shapes are easily distinguished from that due to random orientation of  $M$ .

Two lines were observed by continuous wave techniques in the isotopically enriched sample of magnetite at temperatures of 100 to  $300^\circ\text{K}$ . One sharp line is attributed to isotropic hyperfine interaction on tetrahedral sites. The shape of the second line fits the theory for random orientation and is attributed to hyperfine anisotropy on octahedral sites. The resonance pattern is linear in  $T^{3/2}$  down to a temperature  $T$  of  $100^\circ\text{K}$ , below which absorption is weak. The extrapolated internal fields at  $0^\circ\text{K}$  are  $H(\text{tet.}) = 508$  K oe,  $H_{\perp}(\text{oct.}) = 489$  K oe, and  $H_{\parallel}(\text{oct.}) = 477$  K oe. At no temperature was evidence of domain wall observed.



The octahedral frequencies are considered to be mean values characteristic of  $\text{Fe}^{+2}$  and  $\text{Fe}^{+3}$ . The discrepancy between the measured anisotropy,  $H_{\perp}(\text{oct.}) - H_{\parallel}(\text{oct.}) = 12$  kilogauss and the calculated dipolar value of 19 kilogauss is attributed to specific crystal field effects in  $\text{Fe}^{+2}$ .

## 25. THE ATOMIC MOMENTS AND HYPERFINE FIELDS IN $\text{Fe}_2\text{Ti}$ AND $\text{Fe}_2\text{Zr}^*$

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$\text{Fe}_2\text{Ti}$  and  $\text{Fe}_2\text{Zr}$  are members of the group of inter-metallic compounds possessing structures of the Laves type. The former crystallizes in the hexagonal Laves phase structure ( $C_{14}$ ), the latter in the cubic structure ( $C_{15}$ ). The literature contains little or no information on the magnetic properties of alloys of this type containing two transition elements. The present neutron diffraction and Mössbauer studies were undertaken not only to remedy this situation but also to investigate the dependency of the internal magnetic fields of the iron atoms on their magnetic moment.

Saturation magnetization measurements show both compounds to be ferromagnetic. In the case of  $\text{Fe}_2\text{Ti}$  where the iron atoms are located on two non-equivalent sites, the diffraction studies lead to an assignment of  $0.10 \pm 0.03 \mu_B$  for the 2 (a) sites and  $0.20 \pm 0.02 \mu_B$  for the 6 (h) sites of the space group  $P6_3/\text{mmc}$ . The total of  $0.35 \mu_B/\text{molec.}$  agrees quite well with  $0.34 \mu_B/\text{molec.}$  expected from the measured magnetization. In the case of  $\text{Fe}_2\text{Zr}$  the magnetic scattering gives a value of  $1.34 \pm 0.06 \mu_B$  for each of the iron atoms, which is to be compared with  $1.28 \mu_B$  given by the saturation magnetization. In both materials the assignment of any moment to the Ti or Zr atoms could be ruled out. All of these moments from the neutron diffraction work have been evaluated using the iron metal magnetic form factor.

The Mössbauer patterns of both of these materials show unusual features. For  $\text{Fe}_2\text{Ti}$  two non-symmetric absorption lines were observed using an unsplit source of  $\text{Co}^{57}$  in copper. The average width of the lines, source plus absorber, was found to be  $0.350 \pm 0.010$  mm/sec with their center shifted by  $-0.186 \pm 0.006$  mm/sec compared to stainless steel. While the principal origin of the  $\text{Fe}_2\text{Ti}$  Mössbauer pattern lies in electric quadrupole effects, the apparent lack of symmetry can be explained by the presence of a small additional Zeeman splitting of the iron nuclei levels. Estimates of the size of these magnetic fields are obviously approximate since they involve an analysis of the differences in peak shapes. An upper limit of 10 kgauss is suggested by the data. In spite of the small moments found on the iron atoms in  $\text{Fe}_2\text{Ti}$  by the neutron work, this value for the nuclear field is still somewhat low. Scaling down the pure iron values in proportion to their relative moments would lead to fields of 15 kgauss and 30 kgauss for each of the sites.

For  $\text{Fe}_2\text{Zr}$ , a more typical absorption pattern is found. A result of the somewhat more cubic environment of the iron atoms is the absence of significant quadrupole effects. The Zeeman splitting at room temperature gives a value of  $190 \pm 10$  kgauss for the internal field at the iron sites.

This agrees quite well with what is expected from an inter-comparison of the magnetic moments of the iron atoms in  $\text{Fe}_2\text{Zr}$  and  $\alpha\text{-Fe}$ .

Freeman and Watson<sup>1</sup> have noted that the nuclear magnetic fields are quite sensitive to changes in the mean radius of the 3d electrons. All other terms contributing to the nuclear fields should be proportional to the magnetic moment of the iron atoms. Interpreted along such lines, the above results would indicate that in spite of the marked changes in the number of unpaired electrons in the Fe atoms in these materials, no significant change has occurred in the radial dependence of the 3d electrons from that in metallic iron. This is of particular interest because of the marked change in the environment of the iron atoms in  $\text{Fe}_2\text{Ti}$  and  $\text{Fe}_2\text{Zr}$  compared to body-centered iron.

\*Work performed under the auspices of the U. S. Atomic Energy Commission.

<sup>1</sup>A. J. Freeman and R. E. Watson, Phys. Rev. Letters 5, 498 (1960).

## 26. INTERNAL MAGNETIC FIELDS IN NICKEL RICH NICKEL-COBALT ALLOYS

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The nuclear magnetic resonances of both the  $\text{Ni}^{61}$  nuclei and the  $\text{Co}^{59}$  nuclei have been observed in a series of ferromagnetic nickel alloys containing up to two atomic per cent cobalt. The samples were powders with diameters of up to ten microns, and were annealed at  $1050^\circ\text{C}$ . The room temperature value of the magnetic field at the site of the  $\text{Co}^{59}$  nucleus in any of these alloys is found to be about 112 koe, considerably smaller in magnitude than the magnetic field at the nickel site in either the alloy or in pure nickel, which is 170 koe, assuming a  $\text{Ni}^{61}$  nuclear moment of 0.30 nuclear magnetons,<sup>1</sup> or at a cobalt site in pure cobalt, which is about 212 koe. The resonance frequency of the cobalt, about 113 Mcps, increases roughly in proportion to the increase in saturation magnetization as the temperature is lowered. Neither the cobalt resonance frequency nor the nickel resonance frequency changes with cobalt concentration as rapidly as the change in saturation magnetization would suggest. The line widths of both the nickel and cobalt resonances increase with concentration, and the intensities diminish even more rapidly with increasing cobalt concentration.

Three different nuclear resonance spectrometers were used in this research. A high frequency and a low frequency super regenerative oscillator permitted visual presentation of the resonances. Line shapes were studied using recorder presentation with a marginal regenerative oscillator.

<sup>1</sup>J. W. Orton, P. Auzins, and J. E. Wertz, Phys. Rev. 119, 1691 (1960).



# SESSION IIIA

## THIN FILMS - 1

C. J. KRIESSMAN, *Presiding*

### 27. THE INDUCED ANISOTROPY OF EVAPORATED FILMS FORMED IN A MAGNETIC FIELD (Invited)

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The origin of the induced magnetic anisotropy in evaporated films formed at normal incidence in magnetic field is not yet clear. A systematic and quantitative investigation, therefore, was undertaken, in order to clarify this origin in iron and nickel-iron films. Some of the following three factors are considered to be possibly the origin: (1) trace of fibrous structure or anisotropic aggregation of crystallites or anisotropic shape of crystallites, (2) stress induced in the limited parts of the film, (3) intrinsic stress induced in the interior of the film.

The following experiments were performed to determine which is main among the above three factors: (1) the temperature dependence of  $K_u$ , (2) the relation of  $K_u$  to film thickness, substrate temperature and substrate materials, and (3) both the change of  $K_u$  with isothermal magnetic annealing and the relation of  $K_u$  to composition have been experimentally studied.

First, any appreciable fibrous structure was not detected by electron diffraction. The temperature dependence of  $K_u$  in the range between 200°C and -180°C for films which were evaporated at substrate temperature 20°C could be explained neither by the crystal anisotropy nor by the atoms pair orientation theory. The anisotropy in question, therefore, may originate from the other factors.

Second, if the stress induced in the limited parts of a film causes the anisotropy,  $K_u$  should remarkably depend on the film thickness. If this stress arises from the difference in thermal expansion coefficient between film and substrate, materials of the latter should largely influence the value of  $K_u$ . The experimental results are as follows: (1) the values of  $K_u$  are considerably scattered in both iron and nickel-iron films formed at 20°C, while  $K_u$  is likely to decrease with increasing thickness in case of films formed at 300°C, (2)  $K_u$  remarkably decreases with increasing substrate temperature, (3) any appreciable difference in the value of  $K_u$  was not detected with the exchange of substrate material. These results suggest that a very small part of the uniaxial anisotropy might be caused by the stress induced in the limited parts of films but a greater part originates from another factor.

Putting the above facts together, the structure defects formed during evaporation, that is, such imperfections as vacancies, dislocations, impurity atoms, etc., may be the most possible origin, if these imperfections are directionally aligned. In order to study much more about the nature of the imperfection alignment, the following experiments were done.

The change of  $K_u$  with isothermal treatment alternated by heatings with and without magnetic field at temperatures between 100 and 300°C was measured. The films were formed on a silica substrate at 20°C. The magnetic field applied along the direction of difficult magnetization.  $K_u$  decreases with duration of the magnetic heat treatment and, at last, the direction of easy magnetization was completely exchanged with the direction of difficult magnetization at the beginning. It should be noticed from this experiment that the activation energy required to change the easy direction is about 1 eV and that the direction of easy magnetization at the beginning is elastically recovered as soon as the heat treatment without magnetic field begins.

If the structural imperfection model is possible as the origin, it is important to examine whether some relation exists between the magnetostriction and the imperfection alignment or not. The results showed that the maximum and minimum of  $K_u$  appeared at compositions with 60 and 80 per cent nickel, respectively. These compositions nearly correspond to the maximum and minimum of magnetostriction, respectively.

It is suggested from the present study that the main part of the anisotropy likely to originate from some directional alignment of structural imperfections which is presumed to have some connection with the magnetostriction.

### 28. LABYRINTH DOMAINS IN PERMALLOY FILMS

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Bitter pattern studies of Permalloy films in reversing fields have revealed that flux reversal may occur in a non-coherent manner in which alternating switched and unswitched regions form a labyrinth-like pattern.<sup>1</sup> Conditions for quasi-static switching by labyrinths are described: for a reversing field at an angle  $\alpha$  to the easy axis, reversal takes place by parallel wall displacement for  $\alpha < \alpha_c$ , and by labyrinths for  $\alpha > \alpha_c$ ;  $\alpha_c$  depends on thickness and preparation variables and varies from 0 to about 60°. Labyrinths may propagate from either the film edge or the interior and are seen as long slender domains which develop from the tip (rather than laterally) leaving behind regions of unswitched material. The threshold for labyrinth switching is found, in non-inverted films (that is, wall coercive force at  $\alpha = 0$  less than the anisotropy field), to lie below the coherent rotation threshold by an amount which increases with decreasing film thickness. Characteristic patterns in thin and thick films with and without external fields present are presented. Some qualitative features of labyrinth domains are discussed, including their stability and their relationship to anisotropy dispersion. Upon repeated reversals, labyrinths are found to reoriginate at approximately the same locations, suggesting local low switching thresholds. A model for labyrinth propagation is presented in which the film is assumed to have randomly located regions of low anisotropy which act as switching centers. Magnetostatic coupling of the switched portion of a labyrinth to an adjacent low anisotropy region about to switch accounts for the propagating feature of



labyrinths; this coupling, along with that of the switching center to the surrounding unswitched material, leads to a direction of propagation as a function of thickness in reasonable agreement with experiment and is partially successful in predicting the labyrinth threshold and its thickness dependence.

\*Operated with support from the U. S. Army, Navy and Air Force.

<sup>1</sup>D. O. Smith, J. Appl. Phys. 32, 70S (1961).

## 29. BITTER PATTERNS OF EPITAXIALLY GROWN SINGLE CRYSTAL THIN FILMS OF IRON AND NICKEL.

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The single crystal thin film technique has many advantages chiefly because single crystals of some material which are very difficult to obtain in bulk form can be easily and quickly grown in the film form. In order to utilize this technique advantageously for the study of magnetic materials, the characteristics of the epitaxially grown single crystal thin films should be known. For example, it is already known that the anisotropy constants of some epitaxially grown single crystals have a higher value than bulk material when the film remains on substrates. In order to know the origin of such deviations in magnetic properties from the bulk as well as to utilize this technique in the study of ferromagnetic crystals, basic domain structures of epitaxially grown single crystals of iron and nickel were investigated by the Bitter pattern technique.

Single crystal thin films of both materials were obtained by evaporation onto freshly cleaved MgO single crystals held at an appropriate temperature in a vacuum of  $10^{-5}$  mm Hg or less. The nature of the resulting films was examined using reflection electron diffraction. From the mode of epitaxy, the structure of the domains and the direction of the moments were determined with respect to the orientation of the MgO substrate. Since the easy direction for magnetization is [100] for iron and [111] for Ni, these materials constitute good examples of two cases where the moments lie inside and outside the plane of the film.

The Bitter patterns were obtained with the metal films on the substrate. Because the cleavage surface of the substrate is not perfectly flat, but contains rather large steps as well as tear lines, these defects are clearly observable in the powder patterns. These defects give rise to complications in the domain structures which are important for understanding the effect of the single crystal substrate. The relation between such imperfections and the resulting patterns will be discussed. In addition, the patterns showed signs of the existence of matching strain between the metal film and the substrate.

In general, thick films ( $\sim 1000$  Å) show structures which are similar in nature to those of bulk as would be expected. This is especially apparent in the case of iron, since the magnetic moments lie in the plane of the film. However, as the films become thinner, the domain

structures deviate in a systematic way from the typical shape and become much finer. Furthermore, the sign of the existence of the internal strain becomes more pronounced. In the case of Ni, it is very difficult to obtain domain patterns because the moments do not lie in the film. However, as the film becomes thinner, visible domains can be nucleated which are very similar to those of thin iron films. Visible domains are also nucleated by the application of a magnetic field or by a stress on the film.

## 30. CONSTRUCTION OF HARD DIRECTION HYSTERESIS LOOPS IN THIN PERMALLOY FILMS

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According to single domain theory, thin permalloy films with uniaxial anisotropy should have straight hard direction M-H loops without hysteresis. Practically, however, the M-H loops of almost all films are open, when the ac field is applied exactly in the hard direction and the earth's magnetic field is carefully compensated. Moreover, films thicker than about 800 Å often show hard direction hysteresis loops which are constricted at the origin.

As is well known, magnetization reversal in the hard direction is associated with the splitting up of the film into a great number of long domains parallel to the easy axis. Whether the walls separating these domains are of the Bloch or Néel type depends upon the strength of the applied field. It will be shown that the constricted loops can theoretically be constructed when, in addition to the anisotropy energy and the energy of the magnetization in the applied field, the energy of these walls is considered. Bitter observations of the reversal process confirm the theory, showing that the constriction is actually a result of Néel-Bloch transitions.

## 31. A THEORETICAL MODEL FOR PARTIAL ROTATION

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In order to study the partial rotation process observed in thin ferromagnetic films<sup>1</sup> theoretically, the magnetization reversal of a simple model is investigated. The model consists of an alternating set of infinite slabs, which are cut at an angle of  $45^\circ$  to the easy direction of a uniaxial anisotropy, with no exchange forces between them, and whose anisotropy constants  $K_1$  and  $K_2$  differ slightly. The magnetic field is applied parallel to the slabs in the plane, which contains the slab normal and the easy direction and which corresponds to the plane of the ferromagnetic film. The magnetization direction is restricted to this plane. This condition replaces the form anisotropy of the film. In order to take into account the finite film thickness, the magnetostatic coupling between the infinite slabs is reduced by a factor depending on the ratio slab width/film thickness.



For this model, the Stoner-Wohlfarth model of coherent rotation is unstable against arbitrarily small differences in the anisotropy constants of adjacent slabs. At approximately the average critical field value of the Stoner-Wohlfarth theory, there occurs a transition to an intermediate state with half of the slabs reversed, the other half not reversed. The transition to the reversed single domain state occurs at a second critical field, which is 1.4 times the Stoner-Wohlfarth value for  $K_1 = K_2$ , and increases with increasing difference between the anisotropy constants. This model thus confirms the essential features of the interpretation of the partial rotation process given in reference 1.

<sup>1</sup>S. Methfessel, S. Middelhoek, H. Thomas, J. Appl. Phys. 32 (1961).

### 32. SUPPORT AND EXTENSION OF THE ROTATIONAL MODEL OF THIN FILM MAGNETIZATION

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Many workers have used the rotational model for magnetic behavior first proposed by Stoner and Wohlfarth as a yardstick with which to measure thin film materials at their disposal. This paper describes the behavior of experimental magnetic material some of which behaves in most respects as the model predicts. An attempt is made to describe deviations from the simple model in terms of an ensemble of such models whose easy axes are distributed through some small angle. The magnetic material under investigation is electrodeposited permalloy in the form of a 1-micron thick cylindrical film on the surface of a 5-mil diameter wire.

If one represents the field applied to the material by a point in an  $h_x, h_y$  plane, then a critical examination of the material's behavior in terms of the model can be obtained by permitting the applied field to follow various trajectories in this plane and simultaneously observing the resultant magnetization. M-H loops taken on certain trajectories agree closely with predicted behavior. Of particular interest is the appearance of cusps on the M-H loops when measured along trajectories inclined at small angles with respect to the hard direction. Such cusps are seen only with material which is quite uniformly oriented, i.e., the dispersion of easy axis orientations is small.

The predicted boundary on this  $h_x, h_y$  plane within which reversible behavior of M obtains is represented by the astroid,  $h_x^{2/3} + h_y^{2/3} = 1$ . The actual boundary has been measured by several techniques. The manner in which domain wall processes enter into the observed behavior is implied by the measured boundaries. This leads to a proposed explanation of the well-known variation of the dynamic threshold for switching in the presence of transverse fields.

### 33. INHOMOGENEOUS COHERENT MAGNETIZATION ROTATION IN THIN MAGNETIC FILMS

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In studying the mode of reversal of thin magnetic films under inhomogeneous applied fields it has been observed that a form of coherent magnetization rotation can occur in which the orientation of the magnetization is inhomogeneous. The particular case of interest was that in which the field was produced by the current in a thin strip conductor close to a continuous, thin, uniaxial magnetic film.

A 0.5 cm wide strip line was laid over the surface of the film and so close to it that the field dropped off very sharply at the strip edge. The strip was laid along the easy direction, and between it and the film were placed several fine wires, also parallel to the easy direction and each at a different distance from the center of the strip. These wires were used as pickup lines, the return line being placed in zero field. The drive strip was supplied with current at 400 cps and each pickup line was connected in turn to the vertical amplifier of an M-H loop plotter.

The films investigated were made of Gyrallloy evaporated on to aluminium substrates, but the effects observed are expected to occur in any uniaxial magnetic film which exhibits coherent magnetization rotation in homogeneous fields. The film thicknesses range from 500 Å to 6000 Å, and all the films were magnetically uniaxial.

For all the pickup lines, straight-line hysteresis loops were observed, just as for the whole film. But the value of  $H_k$  derived from the loop was seen to depend upon the lateral position of the pickup line. This value increased monotonically with the distance of the line from the drive-strip center.

If the magnetization immediately under the drive strip were to rotate coherently and homogeneously, free poles would appear at the walls dividing this region from the rest of the film. The demagnetizing field arising from these poles would tend to become infinitely large close to the walls, and would oppose the applied field. This is inconsistent with homogeneous coherent spin rotation. In fact the observations suggest that the magnetization at the center of the strip is rotated into the hard direction at a lower field than the magnetization off-center. Also, at any given applied field, the magnetization at any given point under the strip lies at an angle to the easy direction which is governed by its distance from the center of the strip. It is interesting to note that this occurs in a region within which the applied field is homogeneous.

The inhomogeneous rotation has also been observed in steady fields by means of the Kerr effect. The magnetization distribution under a 0.1 cm wide strip measured magneto-optically appears to be similar to that under the wider line.



#### 34. KERR MAGNETO-OPTIC STUDIES OF THE ANGULAR DISPERSION OF THE EASY AXIS OF PERMALLOY FILMS

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The longitudinal Kerr magneto-optic effect has been used to study the angular dispersion of the easy axis in thin permalloy films which have been vacuum-deposited in the presence of a magnetic field. Two different methods are employed corresponding to the hysteresis loop methods and the pulse methods which are generally used for observing dispersion. The Kerr methods yield values of the maximum dispersion angle,  $\alpha_{\max}$ , instead of  $\alpha_{90}$  or  $\alpha_q$ , the angles for 90% and 50%, respectively, of maximum remanent magnetization obtained by hysteresigraphic methods. The values of  $\alpha_{\max}$  obtained by the Kerr methods range from 7° to 12°, and the two methods agree within the experimental error. The values of  $\alpha_{90}$  obtained with a hysteresigraphic method and with a pulse technique for the same samples also agree within expected error. There is reasonable agreement between the values of  $\alpha_{\max}$  obtained with the Kerr methods and values of  $\alpha_{\max}$  estimated from the hysteresigraphic and the pulse data. The latter estimates cannot be made with precision, however. Film composition and substrate temperature have been found to significantly affect  $\alpha_{90}$  and  $\alpha_{\max}$ ; rate of deposition and substrate treatment have been found to have relatively small effect.

Observations have been made of the breakup of a previously saturated permalloy film into fine domains in the presence of a transverse field (applied in the difficult direction) less than the anisotropy field,  $H_K$ . The transverse field for breakup,  $H_0$ , has been shown to be the same as the reversible limit measured by a hysteresis loop technique for a number of permalloy film specimens. The transverse loop coercive force,  $H_C$ , has also been found to be approximately equal to  $H_0 = \alpha H_K$  for most of the films measured. The ratio  $M_r/M_s$  for the transverse loop has not been found to be well correlated with  $\sin \alpha_{\max}$  for the sample studied. In general, however, these findings agree with experimental observations and conclusions of others based on the applicability of the Stoner-Wohlfarth theory to domain rotation.

#### 35. ELECTRON MICROSCOPE STUDY OF THE ROUGHNESS OF PERMALLOY FILMS USING SURFACE REPLICATION\*

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A study of the surface roughness of 17% Fe-83% Ni films evaporated at normal and oblique incidence with respect to a substrate and the correlation with magnetic properties has been completed. The results which are in substantial agreement with those reported by Smith,<sup>1</sup> et al. were obtained by an entirely different technique. The films were replicated by various techniques in an effort to rule out any artifacts which might be introduced by the

replicating material. By this method, films, regardless of thickness, can be examined, while a study such as conducted by Smith,<sup>1</sup> et al. is limited by the inability of electrons to penetrate thick specimens. All films were evaporated at room temperature onto carbon coated glass micro-cover slides.<sup>2</sup>

Electron micrographs taken of replicas of thick Permalloy films evaporated at normal incidence to the substrate exhibited generally a randomly oriented granular structure. Although an alignment of surface structure was observed occasionally, in general such alignment was not seen. In the cases which showed alignment, no correlation between the direction of the shadow casting and the direction of surface structure was found.

For oblique incidence films, the degree of alignment was found to be dependent upon the angle of incidence and the thickness of the films. A directional alignment of 100 Å films evaporated at 55° was rather obvious. The chains of particles were perpendicular to the incident beam as observed by Smith<sup>1</sup> and Konig<sup>3</sup>. The average particle size was approximately 200-250 Å. Films 250 Å thick evaporated at 60° were somewhat smoother than the 100 Å films with an average particle size of approximately 100 - 150 Å. Chains of aligned particles, however much shorter than the ones observed in the 100 Å films, could be seen.

The tendency of Permalloy films to become smoother with thickness was evident on films evaporated to 800 Å thickness at 40° incidence. Here an alignment of chains which are very short can be observed. These chains do not run in a particular direction. Rather clusters of chains can be seen running in one direction in one area and in another direction in another area. No explanation for this observation can be given. The same phenomenon was also observed when an alignment of the topography was noticeable on normal incidence films.

In addition to the visual inspection of the films, a statistical evaluation of the micrographs has been made which shows clearly a high degree of alignment for films up to 250 Å thick evaporated at angles other than normal to the substrate.

In general, the magnetic data agreed with results obtained by Smith,<sup>1</sup> Cohen,<sup>4</sup> and Kambersky<sup>5</sup> including a 90° change in the easy direction for angles > 70°.

The most important magnetic data was a serious discrepancy for oblique incidence films between the magnetic thickness, i.e., thickness determined from magnetic moment measurements assuming a value of  $I_s$ , and the optical thickness as determined by a multiple beam interferometer. This is in contrast to the normal incidence films for which good agreement was found.

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<sup>1</sup>Smith, D. O. et al., *J. Appl. Phys.* **31**, 1755 (1960).

<sup>2</sup>Bradley, D. E., *Brit. J. Appl. Phys.*, **5**, 65 (1954).

<sup>3</sup>Konig, H. and Helwig, G., *Optik*, **6**, 111 (1950).

<sup>4</sup>Cohen, M. S., *J. Appl. Phys.*, **32**, 87S (1961).

<sup>5</sup>Kambersky, V. et al., *Czech. J. Phys.*, **B11**, 171 (1961).



## 36. THE EFFECT OF SUBSTRATE CLEANNES OF PERMALLOY THIN FILMS

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The use of mechanical and chemical cleaning techniques in the production of a clean and smooth microslide glass substrate surface for the vacuum deposition of uniaxial thin permalloy film elements has been studied. The principal cleaning techniques intensively investigated were: (1) a mild detergent wash followed by a vapor degreasing cycle in isopropyl alcohol; (2) a chalk paste scrub followed by a distilled water rinsing cycle and a forced hot air drying process; and (3) a hydrofluoric acid etching followed by a distilled water rinsing cycle and a forced air-drying process. The surface condition of the cleaned microslide glass was assessed with electron micrographs of pre-shadowed carbon replicas (magnification 88,000X). The micrographs show that the chalk-cleaned glass substrate has the smoothest surface. The variously cleaned substrates were then used for the vacuum deposition of 1600 Å 4 mm diameter permalloy film elements (melt composition of 83% nickel, 17% iron). This work was carried out in a  $10^{-6}$  mm Hg vacuum system. Fifteen evaporations were made with a total of 54 elements per evaporation. The permalloy film elements deposited on the differently cleaned glass substrates were then examined by means of a 1000 cycle hysteresis loop apparatus and the following measurements were made: (1) coercive force,  $H_c$ , (2) saturation flux,  $\phi_s$ , (3) orientation of the anisotropy axis,  $\theta$ , (4) magneto-elastic strain coefficient,  $\eta$ , and (5) anisotropy field,  $H_k$ . Dispersion measurements of the "easy" axis were made on a 1000-cycle crossed field hysteresis loop apparatus. Measured in this manner, the chalk-cleaned glass substrates yielded consistently the lower average value of  $H_c$ , i.e., 1.4 oe. The average value of  $H_k$  for the three different cleaning techniques was 2.5 oe. Within the total range of values of coercive force and the anisotropy field, the chalk-cleaned and the acid-etched glass substrates yielded the same values, i.e.,  $H_c \pm 0.15$  oe,  $H_k \pm 0.15$  oe. Measurements of angular dispersion varied from  $8^\circ$  to less than  $1^\circ$  with an average value of less than  $3^\circ$ . The chalk-cleaned substrates yielded the lowest values of angular dispersion.

## 37. ANISOTROPY SOURCES FOR ELECTRODEPOSITED PERMALLOY FILMS

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Experimental evidence has been found to indicate that electrodeposited iron-nickel films are sensitive to the geometric anisotropy of the chain-like growth of crystallites that compose the conducting film of the substrate. Chromium-gold films were vacuum evaporated on glass slides at normal and oblique incidence. Using Wolfe's solution as an electrolyte, Permalloy films were electrodeposited without a magnetic field. Uniaxial anisotropy was found for all the

specimens, including the controls, with the exception of those specimens plated without a magnetic field on normal incidence substrates. Angles as small as  $5^\circ$  were found sufficient to influence this anisotropy. From the knowledge that evaporation at oblique incidence creates a geometric anisotropy by a process of self-shadowing of the crystallites, it is believed that this geometric anisotropy is replicated in the magnetic film by epitaxy. In support of this theory, directional polishing of the substrates was also found to induce an anisotropy, although in these cases usually with higher  $H_k$  values. The epitaxy model can be helpful for applying data from evaporating techniques to plating techniques. Magnetic films in this study were from 3,000 to 4,000 Å thick. Their magnetic characteristics compared favorably to evaporated films on B-H tests measured at 400 cps. Conclusions are that in this experiment, the anisotropy induced by the magnetic field is separate from the one induced by the substrate.

## 38. FACTORS INFLUENCING COERCIVE FIELD VALUES IN SPUTTERED PERMALLOY FILMS

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An investigation has been made of the experimental factors influencing wall and anisotropy coercive field values,  $H_c$  and  $H_k$ , of permalloy films prepared by cathodic sputtering. Since oblique incidence effects are absent in this process,<sup>1</sup>  $H_c$  and  $H_k$  are determined primarily by film homogeneity and substrate temperature.

Depending upon the experimental conditions used, the composition of the deposited film can either be maintained constant, or can change progressively during sputtering from the Ni-rich to the Fe-rich side of the non-magnetostrictive composition. In the second case, the extent to which a homogeneous composition is achieved depends upon the ease with which diffusion and mixing can proceed during film growth. It appears that chemical inhomogeneity, resulting from incomplete mixing, leads to the appearance of an anomalous maximum in the  $H_c$  versus thickness curve within the range 600-1000 Å. This maximum is similar in character to that observed for evaporated films and has previously been identified with the transition from Néel to Bloch type domain walls.

$H_k$  is found to be relatively insensitive to average film composition and homogeneity, but varies systematically with substrate temperature. The equilibrium temperature attained at the substrate after a few seconds, and the resulting values of  $H_k$ , can be effectively controlled by regulating the electrode potential,  $V$ , and current,  $I$ . For a given combination of  $V$  and  $I$ , no variation of  $H_k$  with thickness above 300 Å is observed.

Electron microscope studies reveal that, depending on the cathode constitution and the experimental conditions, the crystal size and texture of the films vary widely. The largest and most perfect crystallites are obtained by reducing oxygen contamination to a minimum. These conditions also lead to improved film homogeneity and reduced coercive force values.

<sup>1</sup>M. H. Francombe and A. J. Noreika, J. Appl. Phys. 32, 97 S(1961).



# SESSION IIIB

## SPIN CONFIGURATIONS AND ANISOTROPY

I. S. JACOBS, *Presiding*

### 39. MAGNETOELECTRIC EFFECTS IN ANTIFERROMAGNETICS (INVITED)

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Spin-ordered materials may exhibit a magnetic moment which is proportional to an applied electric field and an electric moment which is proportional to an applied magnetic field. Both the experimental and theoretical investigations of these recently discovered magnetoelectric (ME) effects are still in an early stage. In this paper a comprehensive discussion is given of the present knowledge of ME effects. The specific topics covered are listed below under the headings of (I) Thermodynamics and Symmetry, (II) Atomic Mechanism, and (III) Experimental Studies on  $\text{Cr}_2\text{O}_3$ .

(I) Thermodynamics and Symmetry. The thermodynamic potential and the derivation of the constitutive relations; a necessary condition for the occurrence of ME effects;<sup>1,2</sup> digression on piezomagnetic effects;<sup>1,3</sup> some aspects of the magnetic symmetry of crystals, especially the transformation properties of axial vectors and the role of time reversal as a symmetry operation; non-existence of ME effects in paramagnetics;<sup>1,2</sup> antiferromagnetic  $\text{Cr}_2\text{O}_3$  as an illustrative example of a material which may exhibit ME effects;<sup>2</sup> thermodynamic and symmetry considerations involved in a proposed "piezomagnetolectric" effect.<sup>4</sup>

(II) Atomic Mechanism.<sup>5</sup> Theory of the temperature dependence of the magnetoelectric susceptibilities of a two-sublattice antiferromagnet; introduction of a fictitious magnetic field; parity considerations and the role of the position-independent part,  $\vec{E}^c$ , of the crystalline electric field; the combined action of  $\vec{E}^c$ , the applied electric field, and the spin-orbit coupling as a possible quantum-mechanical mechanism of ME effects; order of magnitude estimates and comparison with experiments.

(III) Experimental Studies on  $\text{Cr}_2\text{O}_3$ . A) Electrically induced ME effect: experimental method and early observations;<sup>6</sup> measurements of the magnetoelectric susceptibilities ( $\alpha$ 's) on oriented monocrystals;<sup>7</sup> experimental finding that the temperature dependence of the  $\alpha$ 's is strongly anisotropic;<sup>7</sup> determination of the algebraic signs of the  $\alpha$ 's;<sup>8</sup> role of demagnetization corrections.<sup>8</sup> B) Magnetically induced ME effect<sup>8</sup>: experimental method and measurements on oriented monocrystals; experimental finding that for a given crystallographic orientation both ME effects yield the same temperature dependence of  $\alpha$ ; determination of the algebraic signs of the  $\alpha$ 's. C) Structure sensitive effects<sup>9</sup>: experimental finding that the  $\alpha$ 's of certain crystals (grown by the flame fusion method) vary from sample to sample in magnitude but not in temperature dependence; observation of magnetic annealing effects on the magnitudes and signs

of the  $\alpha$ 's; interpretation on the basis of antiferromagnetic regions, e.g., "domains" whose spin configurations are different, e.g., by  $180^\circ$ .

<sup>1</sup> L. D. Landau and E. M. Lifshitz, *Electrodynamics of Continuous Media* (Addison-Wesley Publishing Company, Inc., Reading, Massachusetts, 1960), p. 119. (English translation of a 1958 Russian Edition.)

<sup>2</sup> I. E. Dzyaloshinskii, *Soviet Phys.-JETP* **10**, 628 (1960)

<sup>3</sup> I. E. Dzyaloshinskii, *Soviet Phys.-JETP* **6**, 621 (1958)

<sup>4</sup> G. T. Rado, to be published

<sup>5</sup> G. T. Rado, *Phys. Rev. Letters* **6**, 609 (1961)

<sup>6</sup> D. N. Astrov, *Soviet Phys.-JETP* **11**, 708 (1960)

<sup>7</sup> V. J. Folen, G. T. Rado, and E. W. Stalder, *Phys. Rev. Letters* **6**, 607 (1961)

<sup>8</sup> G. T. Rado and V. J. Folen, paper presented at the International Conference on Magnetism and Crystallography Kyoto, Japan, September, 1961; to be published in *J. Phys. Soc. Japan*. A more extensive account of this work will be published elsewhere.

### 40. THE LATTICE THEORY OF SPIN CONFIGURATIONS (INVITED)

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In the frame of a given crystallographic symmetry a matrix  $\zeta(\mathbf{k})$  is constructed, the eigenvectors and eigenvalues of which are directly related to the spin configuration and magnetic exchange energy respectively. For Bravais lattices the matrix equation reduces to Villian's equation<sup>1</sup>.

When chemical and magnetic unit cells coincide, the eigenvectors of  $\zeta(\mathbf{k})$  are shown to be identical to the "basis of irreducible representations" used by Russian authors<sup>2,3</sup> in the construction of an invariant Hamiltonian.

The usefulness of both methods will be discussed. The matrix theory is more general in the sense that it remains valid for magnetic cells different from the chemical one. Although the theory presented here starts from conventional crystallographic symmetry, it contains all the configurations possible in the so-called magnetic groups and even those, not contained there (helical configurations).

The theory includes isotropic as well as anisotropic (crystalline field, dipolar, pseudodipolar, antisymmetric) coupling in the order 2 approximation.

Various examples of magnetic structures in the fields of perovskites, ilmenites, spinels, garnets etc. will be presented.

<sup>1</sup> J. Villain, *J. Phys. Chem. Sol.* **11**, 303 (1959)

<sup>2</sup> I. E. Dzialoshinski, *J. Phys. Chem. Sol.* **4**, 241 (1958)

<sup>3</sup> E. A. Turov and V. E. Nays, *J. Met. USSR*, **11**, 161 (1961)



#### 41. MAGNETIC STRUCTURE WORK AT THE NUCLEAR CENTER OF GRENOBLE

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The magnetic structures of FeRh, Cr<sub>3</sub>Se<sub>4</sub> and UCo<sub>4</sub> will be reported.

FeRh, with a small excess of Rh has an interesting first order transition from antiferromagnetic at low temperatures to ferromagnetic above a critical temperature T<sub>c</sub> which in the sample investigated is 60°C.

In Cr<sub>3</sub>Se<sub>4</sub> the determination of the magnetic structure is able to solve the problem of the distribution of bivalent and trivalent cations in defect structures in which an ordering of holes occurs.

UCo<sub>4</sub> is an antiferromagnetic structure (T<sub>N</sub> = 12°K) in which only super-superexchange occurs.

#### 42. WEAK FERROMAGNETISM OF SOME ORTHOFERRITES

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Sensitive static torque measurements were carried out on single crystals of MFeO<sub>3</sub>, M=La, Lu, Y. The purpose of this investigation was to determine if weak ferromagnetism in these materials is caused by single ion magnetocrystalline anisotropy or by anisotropic exchange interaction. The linear part of the torque, measured in the various crystallographic planes, as a function of field gives the ferromagnetic component; the quadratic terms give relations between the susceptibility tensor coefficients, the cubic and higher terms yield additional information on the interactions responsible for weak ferromagnetism.

The following symmetry considerations show a priori which cubic and higher terms in the torque can be nonzero. The free energy F is expanded in a series in the applied field H:

$$F = \sum M_i H_i + \frac{1}{2} \sum X_{ij} H_i H_j + \sum C_{ijk} H_i H_j H_k + \dots$$

Using the fact that F must be invariant under all the symmetry operations of the magnetic point group appropriate to these crystals (the ferromagnetic component is in the z direction), one finds that the only cubic coefficients that may be nonzero are C<sub>xxs</sub>, C<sub>yyz</sub>, C<sub>zzz</sub>. The expression for the torque is found using the relation M<sub>i</sub> = δF/δH<sub>i</sub>.

The instrument used was a sensitive recording torque magnetometer. The crystals were mounted in a current carrying coil that enabled the suppression of the linear term in the torque. This increased the sensitivity by several orders of magnitude, so that the coefficients of the cubic terms of the torque could be experimentally evaluated.

The ferromagnetic component, the susceptibility and cubic coefficients were calculated assuming a two sublattice system, for the anisotropic exchange model and also for the single ion magnetocrystalline anisotropy model. The comparison of the measured coefficients to those

calculated for the two models indicates that the anisotropic exchange mechanism leads to better agreement with the experimental results.

#### 43. APPEARANCE OF WEAK FERROMAGNETISM IN FINE PARTICLES OF ANTIFERROMAGNETIC MATERIALS

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Very recently Néel has suggested that antiferromagnetic materials in fine particle form should, under certain conditions exhibit some interesting magnetic properties, i.e., superparamagnetism and weak ferromagnetism. A detailed theoretical analysis has been given by Néel<sup>1,2,3</sup>. In this paper we report our experimental work carried out to obtain evidence which would bear out Néel's suggestions.

Fine particles of some antiferromagnetic materials were prepared by techniques previously reported for the preparation of ultra fine ferrite particles<sup>4,5</sup>. The moment of the material was measured as a function of the external field. The material was then heated above the Néel temperature, cooled in a field of 10,000 gauss and the moment of the material again measured. The magnetic moment vs field curves have identical slopes before and after heat treatment. The samples cooled through the Néel point in a magnetic field have an additional magnetic moment which is field independent. Reheating above the Néel point and cooling without a field eliminated this additional moment. This additional magnetic moment was found to increase as the temperature of measurement decreased and it is reasonable to assume that this additional magnetic moment should depend on temperature in the same way as the sublattice magnetization does. We would speculate that this may be a direct measurement of the temperature dependence of the sublattice magnetization in antiferromagnetic materials.

<sup>1</sup> L. Néel. Comptes Rendus **252**, 4075 (1961).

<sup>2</sup> L. Néel. Comptes Rendus **253**, 9 (1961).

<sup>3</sup> L. Néel. Comptes Rendus **253**, 203 (1961).

<sup>4</sup> W. J. Schuele and V. D. Deetscreek, J. Appl. Phys. **32**, 255S (1961).

<sup>5</sup> A. E. Berkowitz and W. J. Schuele, J. Appl. Phys. **30**, 134S (1959).

#### 44. THE MAGNETIC STRUCTURE OF MANGANESE CHROMITE\*

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Numerous attempts have been made to explain the anomalous magnetic properties of chromites and of mixed spinels containing chromium on the basis of the theoretical models of Néel and of Yafet and Kittel. Recent calculations by T. A. Kaplan and co-workers, using a classical Heisenberg model, have shown that over an appreciable range of the ratio of the BB to the AB interaction, the ground state



is a ferrimagnetic spiral, i.e. a conical spin configuration in which the transverse components progress in spiral fashion along a fixed direction in the crystal. The object of the present study is to determine the suitability of the various proposed theoretical models in the case of manganese chromite, for which rather complex magnetic behavior has been observed by means of neutron diffraction.

From diffraction patterns taken at room temperature it is readily established that  $\text{MnCr}_2\text{O}_4$  is a normal spinel with less than one per cent of the  $\text{Mn}^{+2}$  ions present on B-sites. The Curie temperature, as determined from diffraction data is  $\sim 43^\circ\text{K}$ . Below this temperature the magnetic contributions to the fundamental spinel peaks arising from aligned spins increases as the temperature is lowered and is effectively saturated at about  $20^\circ\text{K}$ . At  $18^\circ\text{K}$ , additional sharp peaks appear at positions which cannot be indexed either on the original unit cell or on any reasonably enlarged cell. These extra reflections persist with unchanged intensities down to  $4.2^\circ\text{K}$ . No change in either the positions or intensities of the fundamental lines is observed in going through the transition. Above  $18^\circ\text{K}$  a broad, diffuse peak is present in the region where the principal extra lines develop. This diffuse peak decreases with increasing temperature, but is still observable above the Curie point. Application of a magnetic field along the neutron scattering vector decreases the magnetic contributions to the fundamentals, but increases the intensities of the extra reflections.

Analysis of the data shows that while little difficulty is encountered in explaining the observed saturation moments, the Néel model and the Yafet-Kittel model fail to account for major qualitative features of the diffraction results. Agreement is not significantly improved by modifications of the Néel model in which reversed spins (either random or ordered) are introduced, or in which some of the spins remain paramagnetic even at low temperatures. On the other hand, all the qualitative aspects of the diffraction results can be explained in terms of the ferrimagnetic spiral model of Kaplan and co-workers. The theoretical calculations give reasonably good quantitative agreement with the positions and intensities of the extra lines as well as the intensities of the fundamentals, while retaining the expected values for the individual ionic moments and the macroscopic saturation moment. Although some discrepancies still remain, there is every reason to believe that these can be removed by further refinement of the theory and by the extension of experimental measurements to single crystals.

\*Research performed under the auspices of the U. S. Atomic Energy Commission.

#### 45. MAGNETIC TRANSITIONS IN CUBIC SPINELS

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The magnetization curves of the cubic spinels  $\text{MnCr}_2\text{O}_4$ ,  $\text{CoCr}_2\text{O}_4$ ,  $\text{MnV}_2\text{O}_4$  and  $\text{CoV}_2\text{O}_4$  have been obtained between  $4.2^\circ\text{K}$  and their Curie points. All the curves indicate the presence of a magnetic transition within this temperature range, and the magnetic moments of the samples at  $4.2^\circ\text{K}$

are inconsistent with collinear (Néel-type) spin configurations. The presence of these transitions is in accord with the theory of spin configurations in normal cubic spinels. According to this theory, a ferrimagnetic non-collinear (spiral) spin configuration at low temperature cannot persist to the paramagnetic region, as the spin configuration is always of the Néel type if the sample is ferrimagnetic immediately below the highest transition temperature. Furthermore, in addition to a Néel-spiral transition, there is the possibility of a second transition between different types of spiral configurations. However, double transitions have not been observed in any of the materials investigated.

The magnetization measurements on  $\text{MnCr}_2\text{O}_4$  are in agreement with both the ground state theory and the neutron diffraction measurements of Corliss and Hastings on the same material. However, there are quantitative features of the other magnetization curves which do not appear to be consistent with the theory. The nature of these inconsistencies and their possible significance are discussed.

\*Operated with support from the U. S. Army, Navy and Air Force.

#### 46. NEUTRON DIFFRACTION STUDIES ON EUROPIUM METAL\*

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Neutron diffraction investigations have been made on metallic europium which show the metal to be antiferromagnetic with a Néel temperature of  $88^\circ\text{K}$ . Analysis of the magnetic reflections are consistent with a helical spin structure having a helix axis in the  $[110]$  direction and a pitch of  $\sim 3.6 a_0$ . Within experimental error between  $20^\circ\text{K}$  and  $88^\circ\text{K}$  the intensities of the magnetic reflections are proportional to  $(T_n - T)^{1/2}$  where  $T_n$  is the Néel temperature. Below  $20^\circ\text{K}$  they tend to become independent of temperature. Within experimental error the helix pitch is not a function of temperature as evidenced by the magnetic and lattice line positions being independent of temperature below  $88^\circ\text{K}$ .

\*Work performed under the auspices of the United States Atomic Energy Commission.

#### 47. NEUTRON DIFFRACTION STUDY OF MAGNETIC ORDERING IN THULIUM

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Magnetic measurements on polycrystalline thulium have revealed the existence of a magnetic ordering transition at a temperature variously reported as  $51^\circ\text{K}$ ,<sup>1</sup> or  $60^\circ\text{K}$ ,<sup>2</sup> to what has been described as an antiferromagnetic state. At low temperatures, Rhodes, Legvold, and Spedding<sup>1</sup> noted



behavior of the magnetization suggestive of a tendency toward ferromagnetism, and Davis and Bozorth<sup>2</sup> deduced a Curie point of 22°K from hysteresis measurements. The highest moment reported was 0.5 Bohr magneton per atom. Warren Henry<sup>3</sup> has observed 3.4 Bohr magnetons per atom in fields of 70,000 oe.

Neutron diffraction measurements have been made on a single crystal specimen of thulium and the results of these measurements show that in the temperature range from approximately 38°K to 56°K the substance is an anti-ferromagnet with the simple oscillating z-component type structure which has previously been reported for the high temperature structure in erbium.<sup>4</sup> Below 38°K there are observed in the diffraction pattern additional satellite reflections which suggest that the sinusoidal modulation is modified below this temperature. At 4.2°K the normal lattice reflections show an increase in intensity in zero field which is consistent with a mean magnetic moment per thulium atom of approximately one Bohr magneton directed parallel to the c axis. The fundamental period of the modulation remains sensibly constant at a value corresponding to 3.5 Å spacings over the entire range of temperatures.

<sup>1</sup> B. L. Rhodes, S. Legvold, and F. H. Spedding, Phys. Rev. **109**, 1547 (1958).

<sup>2</sup> D. D. Davis and R. M. Bozorth, Phys. Rev. **118**, 1543 (1960).

<sup>3</sup> W. E. Henry, J. Appl. Phys. **31**, 323S (1960).

<sup>4</sup> Cable, Wollan, Koehler, and Wilkinson, J. Appl. Phys. **32**, 49S (1961).

#### 48. THE NATURE OF ONE-ION MODELS OF THE FERROMAGNETIC ANISOTROPY

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A great deal of progress has been made toward the understanding of the origin and characteristics of the ferromagnetic anisotropy through the introduction and development of the so-called "one-ion models". Comprising the results of several authors<sup>1</sup>, the one-ion models propose that the observed anisotropy is simply the sum over all crystal sites of single-ion anisotropies, the origin of which lies in the interaction of the individual magnetic ions with the crystalline electric field. The central assumption which permits the independence of the ions and the consequent additivity of single-ion anisotropies is that the near neighbor exchange interactions can be represented by a Weiss molecular field. In this paper the nature of the one-ion models is examined from the point of view of formal statistical mechanics. The purpose here is to establish the relationship between one-ion theories and rigorous statistical mechanics, to derive a formulation for correction terms, and to determine the conditions under which one-ion theories can be expected to be applicable.

Within the minor restriction that the Hamiltonian of the system be composed of isotropic pair exchange interactions plus single-ion anisotropy operators alone, the formal theory demonstrates that the anisotropic component of the free energy cannot in general be decomposed into a sum of single-ion contributions. It is shown however, that a one-

ion theory based on the use of the correct single particle density operator will be adequate in certain restricted circumstances. The correction terms to this one-ion theory are found to consist of correlations or fluctuations in the thermal expectation values of the anisotropy operators of pairs and higher multiplets of ions and thus represent n-ion contributions. A criterion for the importance of these corrections in particular cases has been constructed in terms of a comparison of the magnitudes of the anisotropy energy and the Curie temperature. It is further noted that the use of the molecular field approximation in the theory not only neglects the above n-ion contributions, but also completely ignores the effects of the exchange-induced correlations on the single ion behavior.

<sup>1</sup> K. Yosida and M. Tachicki, Prog. Theor. Phys. (Kyoto) **17**, 331 (1957); W. P. Wolf, Phys. Rev. **108**, 1152 (1957); J. C. Slonczewski, Phys. Rev. **110**, 1341 (1958) and J. Appl. Phys. **32**, 253S (1961).

#### 49. THE TEMPERATURE DEPENDENCE OF THE MAGNETOCRYSTALLINE ANISOTROPY OF FACE-CENTERED CUBIC COBALT\*

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Single crystals of face centered cubic cobalt have been examined between 4.2 and 850°K. The usual transformation to the hexagonal structure below 700°K is avoided by using two special forms of samples: (1) thin films evaporated onto MgO substrates, and (2) the precipitated cobalt-rich phase in a 2% Co-Cu single crystal. In both cases the fcc cobalt is stabilized by and has a close correspondence with the host lattice.

Standard ferromagnetic (electron-spin) resonance techniques have been used to determine the magnetocrystalline anisotropy parameters  $K_1/M$  and  $K_2/M$  over the temperature range indicated and, in addition, the spectroscopic splitting factor,  $g$ , is found to be  $2.06 \pm .03$  independent of temperature.

The temperature dependence of the anisotropy constants is in accord with the relation  $K(T)/K(0) = [M(T)/M(0)]^n$  where for  $M(T)/M(0)$  we have taken the determination by Jaccarino<sup>1</sup> of the temperature dependence of the cobalt nuclear magnetic resonance frequency which we assume to be proportional to the magnetization. The results that are obtained indicate that the power  $n$  of the dependence noted is nearly 10 for  $K_1$ .

\*This work was supported in part by Wright Air Development Division, Air Research and Development Command, United States Air Force.

<sup>1</sup> V. Jaccarino, Bull. Am. Phys. Soc. II **4**, 461 (1959).



# SESSION IVA

## THIN FILMS - 2

H. RUBINSTEIN, Presiding

### 50. ELECTRODEPOSITION OF MAGNETIC MATERIALS (Invited)

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A discussion of the electrodeposition of alloys of the Ni-Fe-Co system is presented. Solutions from which these metals are deposited are described and the role of their constituents discussed. The various applications for these deposited materials is reviewed pointing out their advantages and shortcomings. A detailed description of the electrodeposited thin film is given. Finally, some of the remaining problems in understanding their behaviour are discussed.

### 51. ROTATABLE MAGNETIC ANISOTROPY IN COMPOSITE FILMS

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While investigating the influence of metal-coated substrates on the anisotropy of pure ferromagnetic films, we have prepared films with a uniaxial anisotropy of 15,000 ergs/cm<sup>3</sup>. This is 15 times greater than that found in films of pure nickel evaporated in a magnetic field at normal incidence on glass substrates. This anisotropy also differs from the usual *M*-induced anisotropy in that the easy axis can be rotated by the application of a sufficiently large field.

Proesen, Holmen, and Gran<sup>1</sup> have reported similar behavior in nickel-iron permalloy films evaporated in the presence of oxygen. They have found evidence of the existence of nickel oxide in their films and have interpreted the rotatable anisotropy in terms of an exchange anisotropy between nickel and nickel oxide. The present experiments were performed on nickel films evaporated and measured in a vacuum of less than 10<sup>-9</sup> mm Hg; hence, they should contain little or no nickel oxide. The films we have made differ from others in that the substrate was a molybdenum-covered glass disc.

A possible interpretation is that nickel and molybdenum form an antiferromagnetic solution or compound and that this is in exchange contact with ferromagnetic nickel. Nickel films evaporated onto substrates covered with copper do not show this rotatable anisotropy; it is known that nickel-copper alloys are not antiferromagnetic. Films of nickel-iron have been evaporated on glass substrates and on glass substrates covered with molybdenum or copper. None of these films exhibited the rotatable anisotropy.

This anisotropy has been measured with a torque magnetometer and a hysteresis loop-tracer. At low fields a

unidirectional anisotropy is found, i.e., the film has a constant remanent magnetization below the coercive force. At fields above  $H_C$  the rotatable anisotropy appears. At still higher fields the rotatable anisotropy begins to disappear and there is a large increase in the rotational hysteresis. At the highest fields which we have, 2500 oe, the rotational hysteresis has almost disappeared and only the small, 1000 erg/cm<sup>3</sup>, uniaxial anisotropy which is a result of magnetic annealing remains. After annealing, the rotatable anisotropy is smaller than in as-deposited films and the field range in which the easy axis may be rotated is decreased.

The magnetic annealing anisotropy was present in all the films we made; it does not appear to be associated with residual gases in the evaporation system. The following precautions were followed to make the films as clean as possible: evaporation at 2 · 10<sup>-10</sup> mm Hg in a system having a liquid helium finger, rapid evaporation from a molten source, and evaporation onto metal-covered surfaces.

<sup>1</sup>R. J. Proesen, J. O. Holmen, and B. E. Gran, J. Appl. Phys. 32, 91S (1961).

### 52. UNIDIRECTIONAL HYSTERESIS IN THIN PERMALLOY FILMS\*

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A study of torque curves in thin Permalloy films, and the field dependence of the rotational hysteresis in particular, has revealed that the results are dependent on the value of the reduced coercive force  $h_c = H_C/H_K$  where  $H_C$  is the coercive force in the easy direction and  $H_K$  is the anisotropy field. Further, the films can be classified according to their torque curves in one of two categories, depending on whether  $h_c \leq .5$  or  $\geq .5$ .

Previously,<sup>1</sup> the case of  $h_c \leq .5$  has been discussed where both theoretically and experimentally the situation appears relatively straightforward. In that region, the results agree very well with the theoretical formulae derived by Shtrikman and Treves,<sup>2</sup> S-T.

The extension of the work to the case  $h_c \geq .5$ , to be reported here, has shown that the coherent rotation theory of Stoner and Wohlfarth,<sup>3</sup> S-W, to which the S-T model reduces in this range, does not adequately describe the experimental results.

The behavior of a 1400 A, 77 per cent Ni Permalloy film with  $h_c = .9$ , is discussed as a typical example. The torque curves for applied fields  $h > .5$ , using the reduced rotation, are generally complex and the discrepancies with the S-W model widespread. For  $h < .5$ , if the film is previously saturated, the torque has a period of  $2\pi$ , in agreement with the S-W model. However, when  $.3 < h < .5$ , it is irreversible and the hysteresis is unidirectional, occurring around the easy direction of the bulk of the film. This effect can be understood very well if it is assumed that the film contains small regions with negative anisotropy,  $-K_2$ , where  $K_2$  is of the order of  $K_1$ , the anisotropy of the bulk of the film. The observations of Smith<sup>4</sup> indicate the existence of such regions and have been suggested by him to account for films with  $h_c > 1$ .



Representing the first order effect of the bulk of the film, in the saturated state, as an effective field  $\bar{H}_{dc}$  along the hard axis of the negative anisotropy region, the calculated angular dependence of the torque, shows a unidirectional hysteresis about the easy axis of the bulk of the film. If the torque due to the interaction of the applied field with the magnetization of the bulk of the film is superimposed, the result is in good agreement with experiment. According to the proposed model, the direction about which the loss occurs should be opposite to the direction of  $\bar{H}_{dc}$  and therefore of the magnetization of the bulk of the film. This has been verified experimentally. The magnitude of the unidirectional loss, observed in a series of films whose  $h_c$  varied from .4 to 1.5, has been found to increase sharply above  $h_c = .8$ . This is indicative of a similar increase in the total volume fraction of negative anisotropy regions, in agreement with Smith's results.

\*This research was supported by the United States Air Force under Contract No. AF33(616)-6298, monitored by the Wright Air Development Division.

\*On leave of absence from the Weizmann Institute of Science, Rehovoth, Israel.

<sup>1</sup>Doyle, Rudisill and Shtrikman, J. Appl. Phys. Sept. 1961. Also paper presented at International Conference on Magnetism and Crystallography, Japan, 1961.

<sup>2</sup>Shtrikman and Treves, J. Phys. Rad. 20, 286 (1959).

<sup>3</sup>Stoner and Wohlfarth, Phil. Trans. Roy. Soc., London, 290A, 599 (1948).

<sup>4</sup>Smith, J. Appl. Phys., 32, 705 (1961).

### 53. FERROMAGNETIC RESONANCE IN SINGLE CRYSTAL NICKEL FILMS

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Ferromagnetic resonance measurements have been carried out on single crystal nickel films obtained by epitaxial evaporation on NaCl under various evaporation conditions. Resonance measurements on thin films by previous investigators have yielded anomalous values of  $4\pi M$  which have tentatively been attributed to stress in the films. In this paper it is shown that there is a second factor which must be taken in consideration to explain the shift in resonance peak.

Films that were grown at high temperatures (400°C) and in good vacuum ( $10^{-10}$  mm Hg) exhibited an apparent saturation magnetization only 60-70% of the bulk value. This is in accord with the results of ferromagnetic resonance experiments of Kuriyama et al.,<sup>1</sup> who attempted to explain their results by the presence of large tensile stresses in the films. However, electron microscope studies of films grown under the above conditions indicated that the films formed discrete, well-separated islands of nickel. An approximate calculation based on the observed island size indicated that the demagnetizing factor (perpendicular to the film) for such films would be 60-70% of the value for a continuous planar film. This implies that the shift in the resonance peak was caused not by stresses but by a lowering of the demagnetizing field resulting from the discrete particle growth.

To further substantiate this, continuous films grown at lower temperatures (300°C) and in technical vacuum ( $10^{-5}$  mm Hg) exhibited a magnetization approximately 60% higher than the bulk material values. X-ray diffraction indicated the presence of large compressive stresses in these films. That the increased magnetization was induced by these compressive stresses was convincingly shown by the decrease of the saturation magnetization to bulk value when the stress was relieved by the introduction of water vapor into the cavity. No such change in the external field at resonance was observed for the discontinuous films under a similar exposure to water vapor.

<sup>1</sup>M. Kuriyama, H. Yamanouchi and S. Hosoya, J. Phys. Soc. Japan, 16, 701 (1961).

### 54. STRATIFICATION IN THIN PERMALLOY FILMS

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Microstructural analysis of vapor deposited permalloy films indicates the existence of layers parallel to the substrate surface. This stratification occurs in films formed by a non-interrupted deposition process and is believed to be related to the diffusion of oxygen through the film during deposition.

The analysis of these films has included etch studies using a dilute Mirrofe solution. The films are etched slowly and the undissolved portion is weighed on a microbalance. Markedly differing etch rates are obtained at distinct thickness levels and changes in these etch rates are accompanied by changes in the 60-cycle hysteresis loop characteristics. In the etch of the final 1500 Å of film, the coercive force is found to vary with thickness as  $T^{-4/3}$  as predicted by Néel; likewise, the anisotropy field,  $H_K$ , obeys a similar law.

Films deposited on substrates containing an excess amount of adsorbed oxygen possess rotatable anisotropy.<sup>1</sup> In the etching process, these films lose their rotatable anisotropy property becoming first isotropic and then anisotropic. The field necessary to saturate the film in the planar direction is constant through the rotatable region; it then decreases through the isotropic range of thickness and becomes equal to the anisotropy field in the anisotropic region. In the isotropic range the remanence remains essentially constant.

Torque magnetometer measurements reveal a distinct change in saturation magnetization as layers of the films are removed by etching. Attempts are being made to relate these changes in the saturation magnetization to the Bitter patterns of film cross sections obtained by the Craik technique. Further evidence for stratification includes: (1) the observation of two ferromagnetic resonance lines which vary in a consistent way as the film is etched, (2) cross sectional studies with the electron microscope in which the topography has been enhanced by preferential etching, and (3) chemical analysis of the various layers as determined polarographically.



## 55. FERROMAGNETIC RESONANCES IN THIN FILMS\*

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Ferromagnetic resonance absorption in thin films has been studied at 24 kMc with the steady magnetic field applied parallel to the plane of the film. The majority of investigations were made with permalloy films. Permalloy films less than 400 Å in thickness always exhibit only one resonance peak. Films of greater thickness frequently possess more than one peak, the usual number being two. The observation of two resonance peaks in some permalloy films has been reported earlier by van Itterbeek, et al.<sup>1</sup> The spin wave model does not account for these extra resonances. When a film with two peaks was partially etched, the smaller peak disappears. It therefore appears likely that there is a stratification in the films, formed either in the evaporation process, or by oxidation of the film surface.

Measurements have been made on permalloy films prepared under the conditions necessary to produce rotatable anisotropy.<sup>2</sup> Such films are likely to be stratified. For the films that are anisotropic (about 1000 Å thick) there is a secondary peak on the low field side of the main peak. For films that possess rotatable anisotropy (> 2500 Å) the secondary peak is found on the high field side of the main peak.

In order to examine this stratification idea, a theoretical and experimental study of films with two layers of permalloy or other ferromagnetic material separated by a nonmagnetic layer has been made. In the theoretical model, it is assumed that there is a dipole-dipole coupling between the ferromagnetic layers. The separation of the resonance peaks was calculated in terms of the coupling constants. In the experiments, the two layers of permalloy are separated by a layer of SiO of various thicknesses. Data obtained from some films are in accord with the theoretical model. However, the absorption curve of other films is complicated, with four or more peaks being observed. Analysis of such curves is therefore difficult. It is likely that the multiple peaks are the result of stratification within each layer of the two layer films.

Films with two layers, one Co and the other Fe in one case, and one Ni and the other Fe in another case, have also been investigated. The resonance peaks are found to shift as compared to the peaks for a single-layer film of one of the materials, again in accord with our simple model.

\*This research was supported in part by the U. S. Air Force through the Air Force Office of Scientific Research (ARDC).

<sup>1</sup>A. van Itterbeek, G. Forrez, J. Smits and J. Witters, *J. Phys. Radium* **21**, 81 (1960).

<sup>2</sup>R. J. Prosen, J. O. Holmen and B. E. Gran, *J. Appl. Phys.* **32**, 91S (1961).

## 56. ISOTROPIC STRESS MEASUREMENTS IN EVAPORATED PERMALLOY FILMS

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Isotropic stress measurements in Permalloy films have been carried out as a function of thickness  $t$ , rate of deposition  $R$ , and substrate temperature  $T_s$ . The measurements are made by clamping one end of a thin strip of mica or glass substrate and observing the deflection during deposition of the free end. Tensile, compressive or zero stress is observed depending on  $t$ ,  $R$  and  $T_s$ . For example, for  $t = 1000 \text{ Å}$ ,  $R = 50 \text{ Å/min}$ , the stress is tensile for  $25^\circ\text{C} \leq T_s \leq 100^\circ\text{C}$ , becomes compressive for  $100^\circ\text{C} \leq T_s \leq 325^\circ\text{C}$ , and again becomes tensile up to the upper experimental limit of  $T_s = 350^\circ\text{C}$ . These results extend the work of previous investigators<sup>1-4</sup> who have only reported the presence of tensile stress in various metallic films evaporated at the pressures used in this study ( $\approx 10^{-5} \text{ mm Hg}$ ); compressive stress in Al evaporated at  $10^{-4} \text{ mm Hg}$  on copper substrates has been observed by Murbach and Wilman.<sup>3</sup> The observed stresses are thought to be due basically to the surface energy of the small crystallites which make up these polycrystalline films. As limiting examples, tensile stress could arise from the process of reducing the surface energy between neighboring crystallites which are in contact but which are each bonded to the substrate,<sup>5</sup> while compressive stress could arise from the surface energy of an isolated crystallite bonded to the substrate. Correlation between the stress and magnetic properties has been observed. For example, for films of 85% Ni - 15% Fe, made as in the example above, a rapid decrease in wall coercive force from  $\approx 50 \text{ oe}$  to  $10 \text{ oe}$  occurs as  $T_s$  is increased through  $100^\circ\text{C}$ , the temperature at which the stress is changing from tension to compression.

\*Operated with support from the U. S. Army, Navy and Air Force.

<sup>1</sup>E. C. Crittenden and R. W. Hoffman, *Phys. Rev.* **78**, 349 (1950).

<sup>2</sup>R. W. Hoffman, F. J. Anders and E. C. Crittenden, *J. Appl. Phys.* **24**, 231 (1953).

<sup>3</sup>H. P. Murbach and H. Wilman, *Proc. Phys. Soc. (London)* **B66**, 905 (1953).

<sup>4</sup>R. W. Hoffman, R. D. Daniels and E. C. Crittenden, *Proc. Phys. Soc. (London)* **B67**, 497 (1954).

<sup>5</sup>D. O. Smith, M. S. Cohen, and G. P. Weiss, *J. Appl. Phys.* **31**, 1755 (1961).

## 57. HALL EFFECT DETERMINATION OF PLANAR STRESS IN FERROMAGNETIC FILMS

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Hall effect measurements on a ferromagnetic film can be used to examine the normal anisotropy and the distribution of stress in the plane of the film. The measured Hall voltage exhibits a smooth transition from low to high field



regions rather than the sharp crossover, at technical saturation, which is expected of a single domain. Since the extraordinary or ferromagnetic Hall effect depends upon the magnetic anisotropy perpendicular to the plane one can attribute this difference of behavior to a domain structure describable by a distribution of normal anisotropies. The difference between the single and multi-domain Hall plots depends upon the nature of this distribution and from certain features of the experimentally obtained difference curve one can infer the type and extent of the distribution. If the anisotropy distribution is due to a corresponding range of strains superimposed on the geometric anisotropy of the film one can also estimate the extent and form of the function describing the planar stresses. It is shown that the existence of these distributions results in a difference between the fields at which the Hall measurements indicate technical saturation and that at which the normal anisotropy distribution is peaked. A correction, derivable from the particular distribution function must therefore be applied to the interpretation of Hall effect data.

## 58. STRESS EFFECTS ON THE MAGNETIC PROPERTIES OF EVAPORATED SINGLE CRYSTAL NICKEL FILMS

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Nickel single crystal films were grown by evaporation from a tungsten filament onto heated (100) surfaces of NaCl, prebaked for one hour at 500°C. When measured on the NaCl, such films yield anomalously large values for the crystalline anisotropy constant,  $K_1$ , as well as for  $K_1$ , the perpendicular anisotropy, i.e., the anisotropy resulting from rotation out of the plane of the film. These values decrease to normal bulk values when the films are floated off the rock salt and picked up on glass.

It has been shown by direct X-ray diffraction, using  $\text{CoK}_\alpha$  radiation and a Geiger tube goniometer, that the as-evaporated films exist in a highly strained condition. The strain causes a distinct shift of the diffraction peaks and results in a tetragonal distortion of the cubic symmetry. The parameters of the unit cell changes from  $a_0 = 3.524 \text{ \AA}$  for unstrained nickel to  $a = b = 3.500 \text{ \AA}$  in the plane of the film and  $c = 3.546 \text{ \AA}$  normal to the film. The stress causing this strain is elastic in character, and is relieved by floating the film off the rock salt, as indicated by the return of the cubic unit cell,  $a_0 = 3.524 \text{ \AA}$ .

An estimate of the stress made through the use of bulk elasticity data, yields a compressive stress of  $\sigma = 1.5 \times 10^{10} \text{ dyne/cm}^2$  acting in the plane of the film, with an accompanying Poisson's extension normal to the film. The magnitude and direction of the stress vector indicates that it results from the difference in thermal expansion between the nickel film and the NaCl substrate, rather than from epitaxial stresses or an intrinsic stress, i.e., stresses resulting from defects grown into the film during evaporation.

This externally applied stress system influences the magnetic state of the film by contributing to the total energy of the system through a magnetoelastic interaction. Employing the five-constant magnetostrictive equation, it is shown that a planar compressive stress of this magnitude

does result in an anomalously high crystalline anisotropy value, as well as an anomalously high value of the anisotropy perpendicular to the plane of the film.

## 59. MAGNETOELASTIC BEHAVIOR OF THIN FERROMAGNETIC FILMS AS A FUNCTION OF COMPOSITION\*

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Grand Forks, North Dakota

A series of films of composition varying from 70% Ni, 30% Fe to 90% Ni, 10% Fe were evaporated at a pressure the order of  $10^{-5}$  mm Hg onto a heated glass substrate (300°C) in the presence of a magnetic orienting field in the plane of the film. The composition was controlled by placing a charge of the selected composition on the filament in an amount just sufficient to make the desired 3000 Å film when all of the material was evaporated.

The magnetoelastic behavior of the films was studied by measuring

$$\eta = \Delta H_k / (\Delta \ell / \ell)$$

where  $\Delta H_k$  is the change in the anisotropy field upon the application of a stress sufficient to strain the film an amount  $\Delta \ell / \ell$  (typically  $4 \times 10^{-5}$ ).  $\eta$  was found to decrease linearly with composition to a first approximation with a slope of  $26.5 \times 10^{-5}$  oe per per cent nickel with  $\eta$  being zero for a film composed of 81.5% Ni, 18.5% Fe.

The value of  $\eta$  was compared with that of  $\Lambda_m$  for bulk material<sup>1</sup> where  $\Lambda_m$  is the change in the magnetization divided by the applied stress in the case where the stress approaches zero and where the bulk material is biased by an applied field sufficient to yield the maximum change in  $\Lambda$ . Using bulk data for the saturation magnetization and Young's Modulus the value of  $\eta$  measured here yields a value for  $\Lambda$  which is too large by an order of magnitude. The discrepancy is discussed.

\*This work is supported in part by a grant from the National Science Foundation.

<sup>1</sup>R. M. Bozorth and H. J. Williams, *Revs. Modern Phys.* **17**, 72-8 (1945).



# SESSION IVB MAGNETIZATION

S. FONER, Presiding

## 60. STATISTICAL MECHANICS OF FERROMAGNETISM (Invited)

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A statistical mechanical theory of ferromagnetism confronts all the general difficulties of a strongly interacting many-body problem, compounded with the special peculiarities of the commutation relations of spin operators. Rigorous series developments at low temperatures (the spin wave treatment of Dyson) and at high temperatures have been given, but the interesting intermediate region has been treated only by heuristically-appealing cluster approximations. Recently the Green function method (Zubarev) and the diagrammatic technique based on multivariate cumulant expansions (Brout, Horwitz and Callen) have been applied to the problem. The physical ideas underlying the various approximations and the nature of the results will be discussed.

## 61. MAGNETIZATION OF LOCALIZED STATES IN METALS (Invited)

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A study has been made of the magnetic properties of dilute solutions of Fe, Co, and Ni in various non-magnetic 4d series elements and alloys. In some cases the iron group ions possess a localized magnetic moment which manifests itself as an inverse temperature dependence in the susceptibility of the solution. The occurrence of the moment is determined by the valence electron concentration in the solute element (or alloy). As a function of this quantity one finds portions of the 4d series in which localized moments are strictly absent, interspersed by regions (one centered near Mo, the other in the vicinity of Rh and Pd) in which magnetization occurs and at whose edges the moments appear almost discontinuously.

This behavior may be understood in terms of a theoretical model in which the magnetization is ascribed to a virtual level (of the type proposed by Friedel) of the iron group atom. Polarization occurs when the virtual level lies near the Fermi level and is sufficiently sharp in energy. A self-consistent Hartree-Fock calculation indicates that under these circumstances the impurity atom develops an exchange potential which splits the virtual level, causing it to have different energies for spin-up and spin-down electrons and thus giving rise to magnetization. The model gives a qualitative description of the experiments and, in particular, is able to account for the fact that the impurity atoms sometimes carry a moment that is a fraction of a Bohr magneton. It also predicts an anomalously large susceptibility and residual resistance for those alloys in which the moments are just beginning to appear.

## 62. MAGNETIZATION OF NICKEL AT LOW TEMPERATURES (Invited)

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The temperature dependence of the magnetization of various nickel single crystals has been studied by a pyromagnetic technique from 2°K to 100°K. By this method the change in magnetization of the sample resulting from a change in temperature is recorded directly, and an entire magnetization versus temperature curve may be obtained without changing the external field or altering the positioning and crystallographic orientation of the sample. Magnetization changes as small as one part in  $10^6$  can be detected near liquid helium temperature.

Measurements on these samples have been made in applied fields ranging from 3,000 oe to 14,000 oe and for various crystallographic orientations. These studies not only provide new data relating to the technical approach to saturation but also clearly illustrate the effect of an external field in inhibiting the thermal excitation of spin waves.

The various studies on nickel have revealed that the magnetization versus temperature curves obey neither the  $T^{3/2}$  law predicted by simple spin wave theory nor the  $T^2$  law of the collective electron model. These results will be discussed in terms of various theoretical models including a spin wave calculation containing applied field and anisotropic field energy terms.

## 63. TEMPERATURE DEPENDENCE OF YIG MAGNETIZATION

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The temperature dependence of the saturation magnetization,  $4\pi M_S$ , has been measured for pure, polished YIG spheres in the temperature range from 2 to 50°K. The technique used involved measuring the change in magnetic field separation of the 210 and 220 magnetostatic modes. These modes were chosen since: (1) their separation is linearly proportional to the magnetization ( $H_{210} - H_{220} = 4\pi M_S/5$ ), (2) they have the same second-order shift in line position due to propagation effects, and (3) they have the same shift in line position due to the magnetic anisotropy. This method offers a high degree of precision since the line widths are less than 1 gauss and the separation of the two modes is the order of 500 gauss.

Data were taken for several samples and with the external magnetic field oriented in both the [100] and [111] directions. The measurements yielded  $M_S = M_0 [1 - \alpha T^{(1.85 \pm 0.1)}]$ . The experimental error is sufficiently small so that a fit of the data to the  $T^{3/2}$  curve predicted by simple spin-wave theory is definitely excluded. Since the decimal exponent has little physical significance, the experimental curve has been fitted to the higher order spin-wave correction terms. These results will be presented.

\*The experimental work was done while the author was with Hughes Research Laboratories, Malibu, California.



#### 64. NUCLEAR MAGNETIC RESONANCE IN THE INSULATING FERROMAGNET $\text{CrBr}_3$

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The recently discovered insulating ferromagnet<sup>1</sup>  $\text{CrBr}_3$  ( $T_C = 37^\circ\text{K}$ ,  $\mu_S = 3 \mu_B/\text{Cr}^{+3}$  ion) more closely approximates the model of ferromagnetism on which spin wave theory is based than do previously known ferromagnets.  $\text{Cr}^{53}$  nuclear magnetic resonance (NMR) has been observed in this compound and has been used to test spin wave predictions. A precise measurement of the temperature dependence of the saturation magnetization has been obtained by observing the temperature dependence of the  $\text{Cr}^{53}$  NMR in zero applied magnetic field. Comparison of these results with predictions of spin wave theory, modified to include effects of known magnetocrystalline anisotropy of  $\text{CrBr}_3$ , have been made. A satisfactory least squares fit to the predicted form has yielded the values of the exchange interactions between nearest and next nearest neighbor  $\text{Cr}^{+3}$  ions:  $J_{\text{nearest}} = 5.44^\circ\text{K}$ ,  $J_{\text{next nearest}} = 0.88^\circ\text{K}$ . These values are consistent with those predicted from the paramagnetic Curie temperature on the basis of molecular field theory. Fits to forms which neglected effects of anisotropy and zone boundary effects on the spin waves due to the discreteness of the lattice were poorer.

The nuclear resonance frequency extrapolated to  $0^\circ\text{K}$  is 58.096 Mcps, corresponding to a field at the Cr nuclei of 241,460 oe at  $0^\circ\text{K}$ . Only those nuclei in the bulk of the ferromagnetic domains, and not those in the domain walls, participate in the resonance. A quadrupole splitting of the three allowed transitions for  $I=3/2$  is observed. The magnetic field at the nucleus is found to be antiparallel to the magnetization. The use of both Cr and Br NMR to investigate the superexchange predominantly responsible for the magnetic interactions in this crystal will be discussed.

<sup>1</sup>Chiro Tsubokawa, J. Phys. Soc. Japan 15, 1664 (1960).

#### 65. FERROMAGNETIC RESONANCE IN $\text{CrBr}_3$

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Murray Hill, New Jersey

Spin resonance has been observed in single crystals of anhydrous chromium tribromide in both the ferromagnetic and the paramagnetic temperature regions. Frequencies of 20 to 27 kMcps were used. At  $1.5^\circ\text{K}$  a g-value of 2.006 is found along with a uniaxial anisotropy,  $K=9.4 \times 10^5 \text{erg/cm}^3$ . Line widths as narrow as 3.5 oe have been observed at  $1.5^\circ\text{K}$ . The line width increases monotonically with temperature up to at least  $300^\circ\text{K}$  where its value is about 450 oe. The crystals are very soft, and the line width is degraded by any handling or manipulation of them.

#### 66. PRINCIPLES AND PROCEDURES FOR PRECISION MEASUREMENTS OF MAGNETIC PROPERTIES

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The advantages of using spherical samples for determining the intrinsic properties of ferromagnetic materials are considered, i.e., the absolute value of saturation magnetization can be determined with the accuracy of the measurement of applied fields and problems of sample placement are minimized. The determination of  $d\sigma/dp$  and compressibility are discussed. It is shown that for a spherical sample  $d\sigma/dV$  can be obtained directly from a measurement of an applied field. An absolute measurement of susceptibility above the Curie temperature of a homogeneous ferromagnetic material can be determined from the ratio of a solenoid current to the displacement of any instrument which has a linear response to magnetic moment. Apparatus for determining changes of Curie temperatures and saturation magnetization to parts per million are described. Pressure effects are measured without resort to high pressure techniques. Sensitivity, convenience, and accuracy are obtained using operational amplifiers in integrating circuits to reduce magnetic moment measurements to voltage measurements with precision potentiometers. Results of measurements for dilute alloys of vanadium in iron are given.



# SESSION VA

## OXIDES - I

L. M. CORLISS, Presiding

### 67. ACOUSTIC LOSSES IN FERROMAGNETIC INSULATORS (Invited)

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Studies of acoustic losses in ferrites and garnets using single crystal spherical resonators will be described. Resonant modes have been observed in spheres of yttrium iron garnet (YIG) at  $\sim 9$  Mcps and room temperature with Q's of  $\sim 10^7$ . This is approximately six times greater than any other known material at the same frequency and temperature. These high Q's have now been observed in both shear and compressional modes.<sup>1</sup> In contrast to YIG, the highest Q observed at this frequency and temperature in any ferrite single crystal is in a high resistivity magnesium ferrite and is  $\sim 10^3$  lower. High resistivity cobalt ferrite single crystals are lower than YIG by  $\sim 10^5$ . Low resistivity ferrites were not measured because of possible complications introduced by a stress induced  $\text{Fe}^{++} = \text{Fe}^{+++}$  relaxation mechanism.<sup>2</sup> An important question to be answered is the reason for the apparent "mechanical" uniqueness of YIG among the ferromagnetic insulators. Its "magnetic" uniqueness is relatively well understood.

It should be noted that the above Q in YIG may be only a lower bound. Contact losses have been eliminated by levitation. However, due to complicating factors such as the relatively large and variable dislocation density (revealed by etching) and surface defects, we are not yet certain whether the measured Q's are characteristic of perfect YIG. The intrinsic Q could be higher than the above figure and still be consistent with the present results.

<sup>1</sup>R. C. LeCraw, E. G. Spencer, and E. I. Gordon, Phys. Rev. Letters 6, 620 (1961). In this reference only a certain compressional mode showed the unusually high Q.

<sup>2</sup>D. F. Gibbons, J. of Appl. Phys. 28, 810 (1957).

### 68. RARE EARTH RUTHENATES

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and R. PAUTHENET

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Pyrochlores  $\text{A}_2\text{B}_2\text{O}_7$  bear a great resemblance with the  $\text{CaF}_2$  or  $\text{UO}_2$  structure. They may be derived from  $\text{U}_4\text{O}_8$  ( $= 4\text{UO}_2$ ) by suppressing one O atom and by ordering A and B atoms in a doubled unit cell. Following the work of R. S. Roth<sup>1</sup> on  $\text{Ti}_2\text{T}_2\text{O}_7$  where T is a rare earth (Nd, Sm, Gd, Yb) or Y, pyrochlores were prepared with  $\text{Ru}^{+4}$  and  $\text{Ir}^{+4}$  replacing  $\text{Ti}^{+4}$  ( $\text{RuO}_2$  and  $\text{IrO}_2$  crystallize in the rutile

structure  $\text{TiO}_2$ ) with the hope to get more insight into the behavior of 4d and 5d elements.<sup>2,3</sup>

Susceptibility measurements have been undertaken from 2°K to 1300°K on various ruthenates. After deduction of the theoretical contribution of the rare earth, a Curie constant is deduced for the  $\text{Ru}^{+4}$  ion which is near to one instead of the value  $C_m = 3$ , expected for a  $d^4$  state with  $g = 2$  and  $j = 2$ .

<sup>1</sup>R. S. Roth, J. Res. NBS, 56, 17 (1956).

<sup>2</sup>F. Bertaut, F. Forrat, M. C. Montmory, C. R. Ac. Sc., Paris 249, 829 (1959).

<sup>3</sup>M. C. Montmory, F. Bertaut, C. R. Ac. Sc., Paris, 252 (1961).

### 69. SUBSTITUTIONS OF DIVALENT TRANSITION METAL IONS IN YTTRIUM IRON GARNET

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Various amounts of the divalent ions of manganese, iron, cobalt and nickel have been substituted for trivalent iron in yttrium iron garnet. Electrical balance was accomplished by the simultaneous substitution of tetravalent silicon and in some cases by tetravalent germanium. Under these conditions, the  $\text{Mn}^{+2}$  ion prefers octahedral sites; the amount of  $\text{Mn}^{+2}$  ion which may be substituted in the dodecahedral sites in a garnet of formula  $\{\text{Y}_{3-x}\text{Mn}_x\}[\text{Fe}_2](\text{Fe}_{3-x}\text{Ge}_x)\text{O}_{12}$  is considerably less than  $x = 1.0$  claimed by Tauber, et al.<sup>1</sup>

The great affinity of silicon for the tetrahedral sites in the garnets results in the reduction of trivalent to divalent iron at high temperatures (see also ref. 2) even in air atmosphere when appropriate amounts of finely divided silica are present. In  $\{\text{Y}_3\}[\text{Fe}^{+3}_{2-x}\text{Fe}_x^{+2}](\text{Fe}^{+3}_{3-x}\text{Si}_x)\text{O}_{12}$  the maximum attainable value of x appears to be 0.5; in  $\{\text{Y}_{3-x}\text{Fe}_x^{+2}\}[\text{Fe}_2](\text{Fe}_{3-x}\text{Si}_x)\text{O}_{12}$ , the maximum attainable value of x is less than 0.1. Thus in the case of the  $\text{Fe}^{+2}$  ion also crystalchemical and magnetic investigation indicates that the  $\text{Fe}^{+2}$  ions prefer the octahedral sites when substituted in yttrium-iron garnet. The most likely mechanism through which broadening or ferrimagnetic resonance lines occurs when  $\text{Fe}^{+2}$  ions are present in yttrium-iron garnet then must involve valency-interaction among  $\text{Fe}^{+3}$  and  $\text{Fe}^{+2}$  ions in the octahedral sites and therefore involve  $-\text{Fe}^{+3}-\text{O}^{2-}-\text{O}^{2-}-\text{Fe}^{+2}-$  chains. (See also ref. 3 and pertinent references therein.)

Of the four divalent ions here discussed, the one found to be by far the easiest to substitute into yttrium iron garnet is cobalt. At the time of this writing the garnet  $\{\text{Y}_3\}\text{Co}_2\text{FeGe}_2\text{O}_{12}$  has been made and we intend to determine whether further substitution is feasible. Magnetic and crystallographic measurements have been made on a number of cobalt substituted yttrium-iron garnet specimens and the results of these measurements will be discussed. The divalent cobalt ions also appear to prefer the octahedral sites in yttrium-iron garnet.

The maximum divalent nickel ion substitution we have attained is represented by the formula  $\{\text{Y}_3\}[\text{Ni}_{0.5}\text{Fe}_{1.5}]$



( $\text{Fe}_{2.5}\text{Si}_{0.5}\text{O}_{12}$ ). As indicated, the  $\text{Ni}^{+2}$  ions also appear to prefer octahedral sites.

A refined procedure for the preparation of the polycrystalline garnets will be described and it will be demonstrated that the exercise of great care in these preparations is of considerable importance.

- <sup>1</sup>A. Tauber, E. Banks and H. H. Kedesdy, *J. Appl. Phys.* **29**, 385 (1958).  
<sup>2</sup>K. A. Wickersheim, R. A. Lefever and B. M. Hanking, *J. Chem. Phys.* **32**, 271 (1960); R. A. Lefever and A. B. Chase, *ibid.*, 1575 (1960).  
<sup>3</sup>E. G. Spencer, R. C. LeCraw and R. C. Linares, Jr. *Phys. Rev.*, to be published, September 1961.

## 70. LOW TEMPERATURE ANISOTROPY OF MANGANESE-IRON FERRITES

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The magnetic anisotropy of the composition series  $\text{Mn}_x\text{Fe}_{3-x}\text{O}_4$ ,  $x \leq 1.80$ , has been measured by the torque method between 4.2 and 77°K. Penoyer and Shafer<sup>1</sup> found in an anisotropy study of this series between 77 and 313°K that for  $1.00 < x \leq 1.80$ ,  $K_1$  is negative and arises predominantly from  $\text{Fe}^{+++}$  ions on octahedral sites. An anisotropy of  $-5.5 \times 10^{-2} \text{ cm}^{-1}$  per  $\text{Fe}^{+++}$  ion has been obtained from measurements of  $K_1$  at 4.2°K. Using the theory of Yosida and Tachiki,<sup>2</sup> which attributes this anisotropy to the splitting of the  $\text{Fe}^{+++}$  levels in the cubic crystalline field, a splitting parameter of  $2.2 \times 10^{-2} \text{ cm}^{-1}$  was calculated. A maximum in the temperature dependence of the  $\text{Fe}^{+++}$  ion anisotropy was observed at 75°K. As  $x$  is reduced from 1.00 to 0.40,  $|K_1|$  decreases as a result of the addition of  $\text{Fe}^{++}$  on octahedral sites. The positive contribution to  $K_1$  per  $\text{Fe}^{++}$  ion is strongly temperature dependent at low temperature, almost doubling in magnitude between 77 and 4.2°K and showing a maximum temperature dependence at 25°K. The contribution per ion increases with decreasing  $\text{Fe}^{++}$  concentration, and a value at 4.2°K of  $1.4 \text{ cm}^{-1}$  per  $\text{Fe}^{++}$  ion, approximately twenty-five times the magnitude of the contribution per  $\text{Fe}^{+++}$  ion, is estimated for  $\text{MnFe}_2\text{O}_4$ . Because of the differences in sign, magnitude and temperature dependence of the  $\text{Fe}^{++}$  and  $\text{Fe}^{+++}$  ion contributions, values of  $K_1$  in this composition region show a minimum at some temperature below 110°K. In  $\text{Mn}_{0.80}\text{Fe}_{2.20}\text{O}_4$  the low temperature  $\text{Fe}^{++}$  contribution is sufficient to change the sign of  $K_1$  from negative to positive at 34°K. Several possible origins of the anomalously large  $\text{Fe}^{++}$  ion anisotropy will be discussed.

<sup>1</sup>Penoyer and Shafer, *J. Appl. Phys.* **30**, 315S (1959).

<sup>2</sup>Yosida and Tachiki, *Progr. Theoret. Phys.* **17**, 331 (1957).

## 71. PREPARATION AND PROPERTIES OF FERROSPINELS CONTAINING $\text{Ni}^{+3}$

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Nickel-iron spinels with compositions between  $\text{NiFe}_2\text{O}_4$  and  $\text{Ni}_2\text{FeO}_4$  have been prepared under oxygen pressures up to 2000 psi and at temperatures between 250° and 950°C.

Good agreement was found between the measured lattice parameters of the resulting spinel phases and the values obtained by extrapolating from the previously determined spinel series between  $\text{Fe}_3\text{O}_4$  and  $\text{NiFe}_2\text{O}_4$ . Spinel compositions with lattice parameters as low as 8.2955 Å were obtained; this compares with 8.3885 for  $\text{Fe}_3\text{O}_4$  and 8.3394 for  $\text{NiFe}_2\text{O}_4$ .

As in the case of the spinel solid solution series between  $\text{Fe}_3\text{O}_4$  and  $\text{NiFe}_2\text{O}_4$ , the composition of these nickel rich spinels was found to depend strongly on both the oxygen pressure and the temperature. These relationships have been determined and are presented in terms of temperature-composition and pressure-composition diagrams. Since it had been shown that there is essentially no measurable deviation from Végard's law between  $\text{Fe}_3\text{O}_4$  and  $\text{Ni}_2\text{FeO}_4$ , lattice parameter measurements were a convenient method of determining the compositions of the spinel phase.

The fact that single phase nickel-iron spinels can be prepared with nickel concentrations greater than  $\text{NiFe}_2\text{O}_4$  indicates that  $\text{Ni}^{+3}$  replaces  $\text{Fe}^{+3}$  in the spinel lattice. Of the several possible configurations for a composition such as  $\text{Ni}_2\text{FeO}_4$ , there is strong evidence from magnetic moment measurements that on octahedrally ligated spinel sites the preferred arrangement is the  $[\text{Ni}^{+2}\text{Ni}^{+3}]$  ion pair. The corresponding decrease in moment as  $\text{Ni}^{+3}$  substitutes for  $\text{Fe}^{+3}$  between  $\text{NiFe}_2\text{O}_4$  and  $\text{Ni}_2\text{FeO}_4$  agrees very well with that predicted by the Néel antiparallel array theory. A lowering of the Curie temperature was also observed in this compositional range.

## 72. PERMINVAR CHARACTERISTICS OF NICKEL-CADMIUM FERRITES WITH SMALL ADDITIONS OF MOLYBDENUM AND COBALT

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During an investigation into low-loss ferrites, it was found that nickel-cadmium ferrite with minor additions of molybdenum and cobalt exhibited wasp-waisted hysteresis loops. After annealing in a magnetic field, the loops then exhibited rectangular hysteresis with sixty-cycle squareness ratios approaching unity. As in the case of zinc substitutions in nickel ferrite, the coercive force and Curie temperature decreased with increasing cadmium content with no evidence by X-ray diffraction or polishing techniques of a second phase in samples containing up to 20 mole per cent cadmium. The coercive force and squareness ratio remained constant over the temperature range of approximately 100 to 400°C. When the slight additions of



molybdenum and/or cobalt were not in the formulation, the wasp-waisted loop was not present. Furthermore, partial and complete substitutions of zinc for cadmium did not exhibit the "perminvar" effect.

The properties, in general, are discussed in terms of the Néel-Taniguchi model of directional ordering. The uniaxial anisotropy responsible for the perminvar characteristics may be due to the migration of the cobalt ions to the preferred octahedral sites that establish an axis of symmetry along a particular [111] direction. The cadmium, having a larger ionic radius than zinc, probably has to "squeeze" into the spinel lattice; the consequent "stretching" of the lattice then permits easier diffusion and ordering of cobalt ions. The role of molybdenum is difficult to define but may be due to creation of cation vacancies in a manner similar to creation of vacancies by excess ferric oxide. Another possibility is that molybdenum's fluxing action allows ease in the diffusion of cobalt ions.

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### 73. VANADIUM IRON OXIDES

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An investigation in the ternary system Fe-V-O has been made of the chemical, crystallographic and magnetic properties of the corundum  $\text{FeVO}_3$ , and the spinels  $\text{FeV}_2\text{O}_4$  and  $\text{Fe}_2\text{VO}_4$ .  $\text{VeFO}_3$  has been reported in the literature as  $\text{Fe}^{+2}\text{V}^{+4}\text{O}_3$  and antiferromagnetic at room temperature. However, the study reported in this paper shows that these results are incorrect. Oxidation of the spinel  $\text{FeV}_2\text{O}_4$  results in the formation of some  $\text{FeVO}_3$ . Conversely, reduction of  $\text{FeVO}_3$  ultimately forms  $\text{FeV}_2\text{O}_4 + \text{Fe}$ . Therefore both the iron and vanadium are trivalent in the compound  $\text{Fe}^{+3}\text{V}^{+3}\text{O}_3$ . Furthermore,  $\text{FeVO}_3$  has a small magnetic moment at room temperature which persists up to approximately 445°K as well as an anomalous rise in the magnetic moment below 150°K. This rise is accompanied by a change in the relative intensity of several lines in the x-ray pattern of the material, which may indicate a shift in the position of the metal atoms. However, no change in the symmetry was observed. The magnetic properties of  $\text{FeV}_2\text{O}_4$  and  $\text{Fe}_2\text{VO}_4$  are also studied. The magnetization curves of both these materials indicate the existence of a magnetic transition between the Curie point and 4.2°K.

\*Operated with support from the U. S. Army, Navy and Air Force.

### 74. X-RAY AND MAGNETIC STUDIES OF $\text{CrO}_2$ SINGLE CRYSTALS

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The oxygen parameter,  $u$ , in  $\text{CrO}_2$ , which has a rutile crystal structure, was determined from x-ray measurements on a single-crystal sphere approximately 0.3 mm in diameter. The (hko) reflections for which  $h+k$  is odd were used because chromium ions do not contribute to these reflections. The best fit between observed and calculated structure factor amplitudes was obtained for  $u = 0.3045 \pm .0002$ . Each chromium ion has six nearest-neighbor oxygen ions which would be equidistant from the chromium ion for  $u = 0.3044$ . Within the experimental error the six chromium-oxygen distances in  $\text{CrO}_2$  are equal.

Using  $u = 0.3045$ , a lattice sum of the tetragonal term of the crystal field potential was carried out to a distance of 10 Å from the central chromium ion. The term began to converge to a positive value. A negative value would be required to remove the degeneracy of  $\text{Cr}^{+4}$  in a manner which empties the  $t_{2g}$  orbitals parallel to the  $c$  axis, as proposed by Goodenough.<sup>1</sup> Cation-cation exchange along the  $c$  axis should, therefore, not be considered negligible in  $\text{CrO}_2$ . This conclusion is confirmed by single-crystal measurements of  $a$  and  $c$  vs. temperature which, in agreement with published measurements on  $\text{CrO}_2$  powders,<sup>2</sup> show that  $a$  increases from 4.422 Å to 4.436 Å and  $c$  decreases from 2.918 Å to 2.913 Å as the temperature is increased from room temperature to 160°C. The decrease in  $c$  with increasing temperature, which is sharpest near the 121°C Curie temperature, indicates the presence of strong magnetic exchange interactions in the  $c$  direction.

A semiquantitative determination of the magnetic anisotropy of  $\text{CrO}_2$  was obtained from magnetization curves of an 80-micron single-crystal sphere with the applied field in various crystallographic directions. Although the small size of the sample restricted the accuracy of the measurements, the following conclusions can be made: the directions of easy magnetization lie in (100) planes at an angle of 30°-40° to the  $c$  axis, but the energy required to magnetize parallel to  $c$  is not much greater (ca.  $8 \times 10^4$  ergs/cc at 77°K and  $4 \times 10^4$  ergs/cc at 300°K). The hard directions are [110] directions. The energy required to magnetize in these hard directions is approximately  $3 \times 10^5$  ergs/cc at 77°K and  $2 \times 10^5$  ergs/cc at 300°K. These conclusions are in agreement with neutron diffraction measurements on  $\text{CrO}_2$  powder<sup>3</sup> which show that the  $\text{Cr}^{+4}$  moments lie at an angle of approximately 40° to the  $c$  axis.

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<sup>1</sup>J. B. Goodenough, Phys. Rev. **117**, 1442 (1960).

<sup>2</sup>K. Siratori, S. Iida, J. Phys. Soc., Japan **15**, 2362 (1960).

<sup>3</sup>E. Adelson, A. E. Austin, C. M. Schwartz, private communication.



## 75. CATION- -CATION THREE-ELECTRON BONDS

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Several types of cation-sublattice d-electron ordering at a transition temperature  $T_t < T_{mp}$ , where the melting point  $T_{mp}$  is primarily determined by ordering of the outer s and p electrons, have been identified in transition-metal compounds: (1) ordering of two cations of the same atom in different valence states to stabilize the Madelung energy, as in  $Fe_3O_4$ , (2) Jahn-Teller ordering, as in  $Mn_3O_4$ , (3) electron-orbit ordering below an electron-spin ordering temperature due to spin-orbit coupling, as in FeO, (4) homopolar cation- -cation bonding, as in  $VO_2$ , (5) bonding-band  $\Rightarrow$  metallic-band transitions, as in  $\gamma Mn$  or in  $\alpha = \gamma Fe$ , (6) ordering of the s - p covalent portion of the bonding between a transition-metal atom and an interstitial atom, as in CrN, (7) s-p polarization ordering for a small cation in a large interstice, as in  $BaTiO_3$ , and (8) low-spin-state  $\Rightarrow$  high-spin-state transitions, as may occur in  $LaCoO_3$ . To this list can be added (9) three-electron cation- -cation bonding, as in stoichiometric FeS.

There are two necessary conditions for three-electron cation- -cation bonding: (1) cation- -cation separations must be  $R_{tt} \approx R_c$ , where  $R_c$  is a critical distance such that for  $R_{tt} < R_c$  the cation-sublattice d electrons are "collective" and for  $R_{tt} > R_c$  the cation-sublattice d electrons are localized," (2) more than one electron per cation d orbital (for high-spin-state cations) and the possibility of electron ordering to give 3/2 electron per d orbital that overlap near-neighbor d orbitals either within a cation-sublattice plane or along a linear cation chain.

The physical characteristics to be associated with three-electron bonding are: (1) displacement of cations from the center of symmetry of their interstice towards one another to form clusters that satisfy the criterion of three electrons per bond, (2) ferromagnetic spin correlations between bonded atoms and a maximum contribution to the atomic moment from the electrons participating in the three-electron bonds of  $0.5 \mu_B$  per bond, (3) partial quenching of the orbital contribution to the atomic moment by bond formation, and (4) sharp changes in electrical resistivity associated with trapping of electrons in the three-electron bonds.

The crystallographic, magnetic, and electrical changes that occur at  $T_\alpha$  in stoichiometric FeS are interpreted in terms of  $Fe^{+2}$  -  $Fe^{+2}$  three-electron bonding within basal planes below this temperature.

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## 76. SOME SUPERPARAMAGNETIC PROPERTIES OF FINE PARTICLE $\delta FeOOH$

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In 1939 Glemser and Gwinner<sup>1</sup> showed that if a freshly prepared aqueous precipitate of ferrous hydroxide was

rapidly oxidized, by hydrogen peroxide for example, a strongly magnetic black precipitate was obtained which they believed to be an unusual crystallographic form of ferric oxide and denoted  $\delta Fe_2O_3$ . More recently Francombe and Rooksby<sup>2</sup> and others<sup>3</sup> have shown that the precipitate is ferric oxihydroxide  $\delta Fe_2O_3 \cdot H_2O$  or  $\delta FeOOH$  with a structure based on the ferrous hydroxide structure  $Fe(OH)_2$ . X-ray analysis shows that  $\delta FeOOH$  is essentially a closed packed hexagonal array of oxygen ions with  $Fe^{+3}$  ions randomly dispersed over the octahedral and tetrahedral sites with a preference for the former. It was shown that on warming  $\delta FeOOH$  the  $Fe^{+3}$  ions orders onto the octahedral sites with the formation of goethite ( $\alpha FeOOH$ ). Further heating produces hematite ( $\alpha Fe_2O_3$ ). It was suggested that the magnetic properties are due to superexchange between  $Fe^{+3}$  ions on octahedral and tetrahedral sites, presumably in a similar manner to that in cubic  $\gamma Fe_2O_3$ . A program of work was started to see if the magnetic data would support this picture.

A range of differing samples of  $\delta FeOOH$  have been prepared by varying the temperature of preparation over a region 25 to 85°C and also by subsequently heating the powders so formed at temperatures up to 150°C. The powders were examined by x-rays and by an electron microscope and found to be hexagonal platelets with their c axes perpendicular to the plane with a mean diameter of 200A and mean thickness of the order 20A.

The powders were pressed into cylindrical compacts and their magnetization measured with fields of up to 9000 oe both at room temperature and at liquid nitrogen temperature. At room temperature the magnetization for most samples fell with decreasing fields to zero or to a very low value at zero applied fields indicating that the materials were superparamagnetic. This was confirmed by plotting the magnetization at both temperatures against the applied field divided by the absolute temperature when, except for low fields, the points tended to lie on a single curve. At 77°K all samples showed a finite remanence and had coercivities lying between 100 and 400 oe. By suitable extrapolation the saturation magnetization at 77°K was calculated and values as high as 27 emu/g were observed. It was found that for a given sample the saturation magnetization decreased smoothly with increasing temperature of the heat treatment while maintaining the chemical formula  $FeOOH$ .

The temperature above which the material becomes superparamagnetic, the blocking temperature, was measured by recording the remanent magnetization as a function of temperature as the sample warmed up in a dewar from liquid nitrogen temperature. The curves showed pronounced tails and therefore the blocking temperature had to be defined as the temperature at which the remanent magnetization had fallen to 1 per cent of the value of the saturation magnetization at 77°K. The blocking temperature throughout all the samples varied from about 200 to 350°K.

<sup>1</sup>O. Glemser and E. Gwinner, Z. Anorg. Chem. 240, 163 (1939).

<sup>2</sup>M. H. Francombe and H. P. Rooksby, Clay Minerals Bulletin, 21, 1 (1959).

<sup>3</sup>J. D. Bernal, O. R. Dasgupta, and A. L. Mackay, Clay Minerals Bulletin, 21, 15 (1959).



## 77. FINE-GRAINED FERRITIES. II. $Ni_{1-x}Zn_xFe_2O_4$

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A series of fine-grained single-phase ferrites were prepared according to the formula  $Ni_{1-x}Zn_xFe_2O_4$ , where  $x$  took the values 0, 0.33, 0.50, and 0.67. A previously-described process<sup>1</sup> combining the so-called flame-spraying and hot-pressing techniques was used to obtain grain sizes of approximately 0.1 micron in the densified bodies.  $\mu'$  and  $\mu''$  were measured to 1000 Mc, with grain size and composition as parameters. Measurements were also made to 3800 Mc on a ferrite annealed through the critical size for multi-domains, confirming the previously-reported<sup>1</sup> theory that magnetic poles on the domain walls are the source of the microwave peak at about 2000 Mc.

In addition, the temperature dependence of initial permeability  $\mu_0$  was studied. The temperature coefficient of  $\mu_0$  was found to increase with grain size. For  $x = 0$ , a six-fold improvement in temperature stability was found in a core with a grain size of 0.08 micron as compared with a core of normal grain size, both having about the same  $\mu_0$ . While the  $\mu_0$ -vs-temperature curves were of various shapes generally, still a set of ferrites made up of differing compositions and grain sizes were obtained having linear temperature dependences. Of practical interest are those with slopes of  $-220$  ppm/ $^{\circ}C$ ,  $0$  ppm/ $^{\circ}C$  (NPO), and  $220$  ppm/ $^{\circ}C$ .

Microwave properties were studied also. Because zero field ( $H_{dc} = 0$ ) measurements showed that fine-grains, i.e., below the critical size for multi-domains, eliminated the so-called microwave loss peak in the rf dispersion, it could be expected that a decrease in the low field loss should occur. To confirm this,  $\mu'$  and  $\mu''$  in the low field region were measured on thin toroidal samples, where dc fields up to 5000 oe were applied. The frequency was varied from 1 to 4 kMc, having the effect of sharpening the resonance phenomenon. The samples were then annealed, and the low field loss was measured as a function of grain size. In addition, microwave measurements of resonance loss vs rf power were made at X-band using a cavity technique. Although normal critical fields  $h_c$  were generally found, one sample with  $x = 0.67$  showed an anomalous peak, increasing  $h_c$  and significantly extending its power-handling capability.

<sup>1</sup>W. W. Malinofsky and R. W. Babbitt, J. Appl. Phys. 32, 2375 (1961).

## SESSION VB SOFT MAGNETIC MATERIALS

P. A. ALBERT, Presiding

### 78. RECENT DEVELOPMENTS IN SOFT MAGNETIC ALLOYS (INVITED)

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The present status of soft magnetic alloys used in power generation, communication, and special devices applications will be reviewed with emphasis on future prospects for their improvement. Although the study of microscopic structure of magnetic materials has led to important advances, present and future improvements require an understanding of the fundamental processes of magnetization and a study of submicroscopic and atomic influences.

The bulk of present day magnetic materials are based on alloys of iron with silicon, aluminum, nickel and cobalt in varying proportions. Their quality has been constantly upgraded by better control of purity and composition through improvements in melting, processing and annealing practices. Some examples of how each of these factors affect the ultimate magnetic properties of soft magnetic alloys will be given. Brief summaries will also be presented on the magnetic properties of current soft magnetic materials and how they relate to modern devices. Some interesting properties of magnetic alloys based on iron, silicon and/or aluminum will be reported. The development of a hardenable, corrosion and abrasion resistant non-magnetic alloy based on a ductile intermetallic compound TiNi will also be disclosed.

Most of the current approaches to the development of new magnetic alloys utilize the now-available rare earth metals since their measured uncompensated spin values are close to the theoretical values. Recent work on magnetic alloys based on the various rare earth metals will be reviewed with emphasis on  $AB_5$  compounds of gadolinium with iron and cobalt. The work shows the existence of a three sub-lattice structure in which the spin arrangements vary from  $A\uparrow, 5B\downarrow$  in the case of  $GdCo_5$  to  $A\uparrow 2B\uparrow 3B\downarrow$  in the case of  $GdFe_5$ .

### 79. MAGNETIC PROPERTIES OF TAPE WOUND CORES OF HIGH-PURITY 3 PER CENT SILICON IRON WITH THE (110)[001] TEXTURE

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In general, the most useful soft magnetic materials are characterized by high residual and saturation flux density, intrinsic saturation at low magnetizing force, high maximum permeability, low power losses, and low coercive force.

Alloys of nickel and iron, such as Deltamax, have most of the desirable magnetic properties. These alloys are



used mostly in low power applications because saturation flux density is relatively low.

Oriented silicon iron alloys have higher saturation flux density than Deltamax and are used extensively in high-power applications for this reason. However, silicon iron also has lower residual induction and slightly lower induction at low magnetizing force than does Deltamax. Also, ac coercive force and losses are higher than found for Deltamax. Particularly this is true for laminations less than about 0.006 in. thick for use in low power applications. In the range of thickness up to 0.006 in., oriented silicon iron does not compare with Deltamax.

Recent discoveries have made it possible to obtain sharp (110) [001] textures in high-purity 3 per cent silicon iron; this combination of conditions should result in improved magnetic properties when compared with commercial silicon iron. Magnetic tests of wound cores, made from tapes 0.001 in. thick, showed the extent of the improvement in properties. AC excitation and losses at frequencies of 60,400 and 1000 cps were equivalent to Deltamax. Residual induction at 60 cps was 16,700 gauss and at 1000 cps, 19,000 gauss. Induction at 2 oe varied from 18,300 gauss to 20,000 gauss, while the ratio  $B_1/B_2$  was 0.91 and 0.97 at 60 and 1000 cps, respectively. For the Deltamax cores induction at 2 oe was 14,500 gauss at 60 cps and 15,100 gauss at 1000 cps. The coercive force of the high-purity silicon iron cores was as low as 0.24 oe at 60 cps and 0.62 oe at 1000 cps.

Performance characteristics of the high-purity silicon iron cores and the Deltamax cores in a high-wave magnetic amplifier were determined at the same three frequencies. The load current output from the silicon iron cores was as much as 22 per cent greater than was obtained from the Deltamax cores; the high-purity silicon iron cores delivered approximately 50 per cent more power.

It is concluded that high-purity silicon iron could be used to advantage in any application where Deltamax type materials are now used.

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## 80. MAGNETIC ANISOTROPY OF THE DEMAGNETIZED STATE

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An isotropic polycrystalline substance, demagnetized in a slowly decreasing alternating field, is in general magnetically anisotropic. The magnetization curve  $J(H)$  in a given direction will depend on the angle  $\phi$  between  $H$  and the direction of demagnetization.

We have compared the two magnetization curves for  $\phi = 0$  and  $\phi = \frac{\pi}{2}$ . Their difference is a maximum for a field of 0.75  $H_C$ , and can be more than 60 per cent of  $J$ . This effect does not exist in substances with uniaxial magnetic anisotropy like hexagonal cobalt, or in samples with large internal stresses. We have also investigated the variation of the anisotropy of the demagnetized state of a sample which was made progressively more uniaxial by an increasing stress.

Furthermore, if a complete demagnetization is followed by a partial demagnetization in the perpendicular direction, this partial demagnetization only modifies the initial state if a certain critical field is exceeded. We have determined some characteristics of this critical field.

O. Yamada has shown that the  $J(H)$  curve at a given temperature depends on the change in temperature of the sample between demagnetization and measurement. We have shown that there is a great similarity between the influence of changes in temperature and changes in stress.

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## 81. ON THE TWO EFFECTS OF CHANGES IN TENSION

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If a ferromagnetic sample initially demagnetized under tension  $\sigma_a$  is subjected to a weak magnetic field, maintained from then on constant, then subjected to a change in tension from  $\sigma_a$  to  $\sigma_b$ , one observes in general a change in the magnetization, no matter whether it is increasing or decreasing tension.

The change of the magnetization due to a change of tension comprises two parts: the first effect and the second effect.

The first effect appears in all of the processes of wall displacement and naturally appears during the hysteresis cycle performed after saturation, while the second effect is present mainly in the Rayleigh region.

The effect of an increase in the tension and the effect of a decrease in the tension are both equal in the case of the first effect, while they are in general different for the second effect.

The first and the second effect always produce some change of the magnetization and are accompanied by the appearance of unsymmetrical hysteresis cycles.

The first effect is caused by the displacement of 180° walls, in contrast to the second effect which is associated with 90° walls.

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## 82. TEXTURED 6.5 PER CENT SILICON-IRON ALLOY

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Sheet of an iron base alloy containing 6.5 per cent silicon and second phase inclusions was prepared by hot rolling and pack rolling. Two structures were obtained by varying the rate of heating to the final annealing temperature. A slow rate of heating yielded {110} secondary recrystallization and a fast rate the primary recrystallization and normal grain growth structure. Only during the slow rate of heating were inclusions effective in suppressing normal grain growth and in promoting growth of {110} secondaries.

The secondary recrystallization texture was of the



{110}<001> type. The normal grain growth texture had a similar degree of alignment of <100> directions with the rolling direction and a near random distribution of planes of the <100> zones parallel to the rolling plane. Similar dc and ac magnetic characteristics were observed for these two structures since they had the same degree of alignment of <100> directions (easy directions of magnetization) with the rolling direction

### 83. GRAIN-SIZE EFFECTS IN ORIENTED 48 PER CENT NICKEL-IRON CORES AT 400 CYCLES

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The constant-current, flux-reset test at 400 cycles has been used widely to evaluate core material for magnetic amplifier applications. An important criterion by this test is the "gain" or ratio of flux change to change in dc control magnetizing force. It is well known that gain is related to the squareness ratio  $B_r/B_m$  of the hysteresis loop.

This study shows that gain is also very sensitive to the grain structure as well as the crystal orientation of the core material. Material having a homogeneous structure has high gain, whereas if partial secondary grain growth has occurred, resulting in a large difference in grain size for the material, the gain can be reduced by a factor 1.5 to 3 times. The effect was artificially simulated by combining two cores of different grain size. Individually, the cores had high gain but together they had very low gain. The cause is ascribed to the difference in coercive force between the large and small grains. The conclusion is reached that high gain is obtainable from material consisting entirely of either large or small grains of proper crystal orientation.

### 84. ANISOTROPY IN ROLLED PERMALLOY TAPE

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The anisotropy induced by cold rolling has been investigated using a recording torque magnetometer. The material was gold permalloy which was cold rolled from 0.014 in. to 0.000125 in. and then heat treated for two hours in a dry hydrogen atmosphere. The torque curves obtained could be decomposed into a hard and soft component. For fields below 200 oe the torque curves due to the hard component have a tangent-like shape with the peak a few degrees from the hard direction, while the torque curves due to the soft component have the regular shape corresponding to uniaxial magneto-crystalline anisotropy. Both components are uniaxial with the easy direction along the roll direction.

The hard component is independent of the heat treatments up to about 900°C and has an equivalent anisotropy field of about 200 oe. The high apparent anisotropy of the hard component and its independence of the heat treatment suggest that this component arises from a magnetostatic

effect. By etching the tape it has been shown that the hard component is due to the surface structure of the tape. The surface structure is probably the result of the rolling process.

The soft component is strongly dependent upon the heat treatment subsequent to the cold work and almost vanishes at an annealing temperature of about 700°C. This decrease is consistent with the ordered pair mechanism suggested by Chikazumi<sup>1</sup>. The small residual soft component at high annealing temperatures is very sensitive to the rate of quenching. This behavior may indicate the presence of partial ordering and some degree of crystal orientation, but a detailed interpretation cannot be given at present. The value of the soft component is in fair agreement with the measured value of the threshold for rapid rotational flux reversal<sup>2</sup>.

<sup>1</sup>S. Chikazumi, J. Appl. Phys. 29 S, 346 (1958)

<sup>2</sup>E. A. Nesbitt and E. M. Gyorgy, J. Appl. Phys. 32 S, 1305 (1961)

### 85. A METHOD OF MAGNETIC ANNEALING AND SOME ELEVATED TEMPERATURE MEASUREMENTS ON VANADIUM PERMENDUR TYPE ALLOYS

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A method of magnetic annealing 2V Permendur (2% V, 49% Co, 49% Fe) stamped rings and laminations in zero externally applied magnetic field is discussed. It is shown that an internal magnetic field biasing technique yields highly improved magnetic properties.

This method involves magnetizing the samples when they are still in a hard condition, i.e., as stamped or low temperature (500 to 600°C) annealed, and then subsequently annealing them at the normal time and temperature employed for the development of optimum magnetic properties (800-900°C). No external magnetic field is applied during any of the anneals.

This technique utilizes the internal magnetic field which remains after magnetizing the samples at room temperature. It is shown that a marked improvement may be obtained in 1-DU type laminations although the samples are unstaked after the initial magnetization (before annealing). Apparently, although the samples are partially self-demagnetized, there is still a sufficiently high internal field remaining to produce a self-magnetic annealing effect during the anneal. It is also shown that the cooling rate employed in this anneal is important—just as it is in conventional magnetic annealing.

Normal dc and 60 cycle magnetic properties are presented for .014 in. ring and 1-DU samples from a number of heats. The data indicate that the effect is manifested by a two to three fold increase in the dc  $\mu_{max}$  and a 35 to 50% increase in the rectangularity of the dc hysteresis loop ( $B_r/B_m$ ). It does not necessarily cause a decrease in the dc coercive force or the 60 cycle watt loss.

In addition to these data, some magnetic properties are presented for a number of 4 mil wound cores of Supermen-



dur in the temperature range from room temperature to 850°C. These data indicate that upon first exposure to the elevated temperature that many hours are required for the material to come to a stable condition as determined by the magnetic properties measured at temperature. The data also clearly indicate that the nominal magnetic saturation (as expressed by the flux density at three oersteds) decreases to a minimum at around 450°C, increases to a secondary maximum at around 550°C, and then decreases monotonically as the Curie temperature is approached.

## 86. THE EFFECTS OF LOW TEMPERATURE NEUTRON IRRADIATION ON THE MAGNETIC PROPERTIES OF IRON-NICKEL ALLOYS

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Iron-nickel alloys of six different compositions were irradiated in the Oak Ridge Graphite Reactor for seven weeks at a temperature of 90°K. Sixty-cycle hysteresis loops were obtained both during the irradiation and the subsequent warm-up to room temperature. Following one room-temperature anneal isochronal annealing studies were also made. In general, it was found that the coercive force, remanence and hysteresis loop shape were modified only slightly as compared with room temperature irradiations of similar samples.<sup>1</sup> The largest irradiation-induced change noted before annealing was found for supermalloy. The coercive force increased about 40 per cent while the remanence decreased by 10 per cent. However, since no constriction in the hysteresis loop was found, it appears unlikely that significant short-range directional order occurred. The isochronal annealing studies were obtained by heating the samples for one hour at 25°C intervals and then making room temperature hysteresis loop measurements. In all cases, up to annealing temperatures of 350°C, the coercive force continued to increase while the remanence continued to decrease. At annealing temperatures greater than 350°C a reversal in behavior was found with both the coercive force and remanence approaching their pre-irradiation values.

\*Oak Ridge National Laboratory is operated by Union Carbide Corporation for the U.S. Atomic Energy Commission.

<sup>1</sup>A. I. Schindler, E. I. Salkovitz, J. Appl. Phys. 31, 245S (1960)

## 87. COOLING RATE EFFECT ON INITIAL PERMEABILITY OF 4-79 MOLY-PERMALLOY

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Unpublished work by Chegwidan and Ashworth<sup>1</sup> has revealed the separate effects of Ni and Mo on the optimum cooling rate ( $CR_{opt.}$ ) for highest initial permeability in 4-79 Moly-Permalloy. This paper shows the combined effects of Ni and Mo and provides an empirical equation for calculating the  $CR_{opt.}$  for highest 60 cycle permeability at 40 gauss flux density ( $B_{40}\mu$ ). Cold rolled strip (.014 in. in thickness) was obtained from 13 heats of Carpenter Steel Company HyMu 80 with Ni between 79.29 to 80.63% and Mo between 4.08 to 4.44%. Prior to running cooling rate studies, duplicate ring lamination samples from each heat were thoroughly purified in  $H_2$  at 2050°F to remove carbon and other non-metallics<sup>2</sup>. This was done so as to obtain the true effects of Ni and Mo on the  $B_{40}\mu$ -cooling rate relationship. Magnetic measurements were made after each 4 hours at temperature until the  $B_{40}\mu$  stabilized. For this phase an average cooling rate of 150°F/hour through the critical ordering temperature range (1112°F to 572°F) was employed. Due to the method of cooling (forced air), the rate was not constant but decreased with decreasing temperature. As annealing time increased,  $B_{40}\mu$  gradually increased and then stabilized between 30,500 and 43,250. This variation is attributed to differences in  $CR_{opt.}$  between heats.

The stabilized samples, when repeatedly heated to 2050°F in  $H_2$  and immediately cooled at different rates between 60 and 810°F/hour, displayed an  $CR_{opt.}$  between 100 to 600°F/hour. As the  $CR_{opt.}$  decreased, the peak of the  $B_{40}\mu$ -cooling rate curve became more abrupt. With the aid of an IBM 650 computer, the following regression equation was calculated:  $CR_{opt.} = 26201.187 + 365.036 (\% Ni) - 633.289 (\% Mo)$ . The standard deviation of the difference between the observed and the predicted values was calculated to be 45.03°F/hour. One observes that Mo has nearly twice the influence as Ni on the  $CR_{opt.}$  but in an opposing direction.

<sup>1</sup>R. M. Bozorth, Ferromagnetism, (D. Van Nostrand Company, Inc., 1956), 4th printing, p. 137.

<sup>2</sup>W. A. Klawitter and A. A. Lykens, "The Effect of Hydrogen Flow In Annealing Permalloy Type Magnetic Alloys", not published to date.

## 88. LOW ANGLE X-RAY SCATTERING FROM MnS IN SILICON STEELS

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In silicon steels, a dispersed MnS impurity phase is believed to pin grain boundaries, inhibit grain growth, and



permit the desired secondary recrystallization for optimum orientation, grain size, and magnetic properties of the finished material. Although May and Turnbull<sup>1</sup> theoretically postulated the role of this precipitate, no experimental evidence has yet been obtained to establish the size distribution of this phase.

Therefore the size distribution of this precipitate was explored by a low-angle x-ray scattering technique. A low-angle scattering spectrometer using a Johansson-focussing monochromator was employed. Several careful experiments determined the role of surface effects, and this information was used to avoid these effects in the final experiments.

Laboratory heats of 3.25 per cent silicon steel compositions, containing varying amounts of sulfur were prepared by vacuum induction melting of high-purity components. After appropriate rolling to hot bands, cold rolling plus intermediate annealing reduced the materials to 0.014 in. thick sheets. These were further cold rolled, and finally pack rolled to foil thickness of 0.5 mil.

Using a sulfur-free sample as reference standard for residual-surface scattering, a plot of  $\ln(I_{sc})$  vs  $(2\theta)^2$  showed the MnS particles to be 20 Å to 150 Å in diameter. This size distribution explains the difficulty in detecting these particles by ordinary electron microscope techniques.

<sup>1</sup>J. E. May, D. Turnbull, Trans. AIME, **212**, 769 (1958).

## SESSION VIA RARE EARTHS

J. F. DILLON, Presiding

### 89. FERRIMAGNETIC RESONANCE IN TERBIUM DOPED YTTRIUM IRON GARNET (Invited)

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Dillon found that the replacement of a small fraction of the yttrium irons in YIG by rare earth ions produced remarkable effects upon the field for ferrimagnetic resonance at helium temperatures. The resonance field varied rapidly with the direction of the applied magnetic field, making, in some cases, excursions of several thousand oersteds within a few degrees. This variation indicates a strong angular dependence of the magnetic properties of the terbium sublattice. This, in turn, implies that the energy levels of the terbium ion in crystalline and exchange fields of comparable strength will depend considerably upon the direction of the latter. Kittel, in fact, suggested that the sharpest excursions of the field were due to near-crossings of the two lowest levels.

A fairly successful attempt has been made to account for the observed resonant behavior of terbium doped YIG. Crystal exchange fields with some adjustable parameters were chosen and the corresponding terbium energy levels and wave functions were calculated. These were used to find the dc magnetization and rf susceptibility of the terbium sublattice and hence the resonant field of the whole magnetic system. Good qualitative agreement was found for all directions of the applied field. This indicates that the general structure of the two lowest levels as a function of the direction of the exchange field has been satisfactorily accounted for. Quantitative agreement is somewhat less satisfactory. This may be ascribed to the fact that only four out of ten possible parameters in the field have been optimized.

### 90. FAR INFRARED SPECTRA OF MAGNETIC MATERIALS\* (Invited)

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The far infrared spectral region (100-1500 microns or 7-100  $\text{cm}^{-1}$ ) corresponds to  $kT$  for  $T = 10\text{-}150^\circ\text{K}$  or  $\beta H$  for  $H = 7 \times 10^4 - 10^6$  oe. Thus, materials with characteristic temperatures or fields of these orders of magnitude might be expected to have interesting far infrared spectra. To date, we have studied far infrared resonance spectra in several antiferromagnets and several of the ferrimagnetic rare earth iron garnets.

Antiferromagnets have a resonance frequency  $\omega_0 = \gamma\sqrt{H_A(2H_E + H_A)}$ , where  $H_E$  is the exchange field and  $H_A$



the anisotropy field. If  $H_E$  is found from  $\chi_{\perp}$ ,  $H_A$  can be found from  $\omega_0$ , and compared with theory. We have done this with  $FeF_2$ ,  $MnO$ , and  $NiO$ , which have resonances for  $T \approx 0$  at frequencies of  $52.7 \text{ cm}^{-1}$ ,  $27.5 \text{ cm}^{-1}$ , and  $36.6 \text{ cm}^{-1}$ , respectively. As the temperature is raised, these frequencies fall, reaching zero at  $T_N$ . When a magnetic field is applied along the easy-axis of  $FeF_2$ , there is a first-order Zeeman splitting, from which a  $g$ -value of 2.25 was determined. In  $MnO$  and  $NiO$ , however, there is an easy [111] plane. In this case the resonant mode is non-degenerate and correspondingly the Zeeman effect is second-order and unobservably small.

The exchange coupling between the rare earth and iron ions in rare earth iron garnets is typically of order  $10\text{-}50 \text{ cm}^{-1}$  ( $15\text{-}70^\circ\text{K}$ ), while the iron ions are coupled strongly together, corresponding to their Curie temperature of about  $550^\circ\text{K}$ . Because anisotropy breaks down selection rules, transitions in which a single rare earth ion spin flips in the exchange field of the iron may be observed at far infrared frequencies. However, we also observe exchange resonances of the sort envisaged in the theory of Kaplan and Kittel (1953) in which the entire sublattice of rare earth ions precess together and induce a corresponding precession of the iron sublattice. Since the frequency of such a resonance depends on the rare earth sublattice magnetization, it is quite temperature dependent. This allows the exchange resonance to be distinguished from the single ion absorption, whose frequency is essentially independent of temperature for temperatures well below the Curie point. To obtain quantitative agreement with experiment, the Kaplan-Kittel theory must be generalized to take account of anisotropy energy. A detailed study of  $YbIG$  has been completed. At  $1.5^\circ\text{K}$ , with the magnetization along the [111] easy direction, we find single ion resonances at  $23.4$  and  $26.4 \text{ cm}^{-1}$  and an exchange resonance at  $14.1 \text{ cm}^{-1}$ . The latter rises in frequency as the temperature is raised, whereas the former frequencies are essentially independent of temperature up to  $60^\circ\text{K}$ , where the intensity becomes too low for observation. Studies of other garnets are in progress, and will be reported.

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## 91. MAGNETOCRYSTALLINE ANISOTROPY OF RARE EARTH IRON GARNETS (Invited)

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This paper will review recent anisotropy data, deduced from static and resonance measurements, on the rare earth iron garnets of formula  $5Fe_2O_3 \cdot 3M_2O_3$  where  $M$  is  $Y$ ,  $Gd$ ,  $Yb$ ,  $Er$ ,  $Sm$ ,  $Tb$ ,  $Dy$  and  $Ho$  respectively. Both the high temperature behaviour (up to  $300^\circ\text{K}$ ) which can be expressed in terms of the anisotropy constants  $K_1$  and  $K_2$  and the anomalous anisotropy properties which occur at liquid helium temperatures are discussed. The results are interpreted in terms of the single ion model which considers the anisotropy resulting from the effect of the combined crystal and exchange fields of the garnet on the energy levels of the

individual magnetic ions, and the extent to which this model can describe the anisotropy of rare earth iron garnets will be briefly discussed.

## 92. THE CONTRIBUTIONS OF RARE EARTH IONS TO THE ANISOTROPY OF IRON GARNETS

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The examination of the angular dependence of the free energy of rare earth ions in garnets provides a powerful means of testing the applicability of "one ion" models for the description of the magnetic properties of these materials. When the free energy surfaces are as convoluted as those of the rare earth ions, quasi-static measurements of the torque acting on the crystal<sup>1</sup> as the magnetization is rotated in symmetry planes can be interpreted much more straightforwardly than can microwave resonance measurements.<sup>2</sup> Torque curves have been obtained on several crystals of compositions  $R_xY_{3-x}Fe_5O_{12}$  and  $R_xGd_{3-x}Fe_5O_{12}$  where  $R$  represents a rare earth element between  $Dy$  and  $Yb$ , and  $x$  lies between 0.06 and 0.30. The measurements have been made from room temperature to  $1.6^\circ\text{K}$  in fields up to  $25,000$  oe using a new torque balance. The novel features of this balance will be described briefly.

The observed torque curves show two striking features: (1) complex shapes with regions of very sharp curvature, and (2) an intrinsic dependence of the torques on the magnitude of the applied field. Thulium is unique among the rare earth ions in that the torque curves for  $Tm$  garnets are well represented by the usual first order cubic anisotropy constant,  $K_1$ . For the other rare earth ions, the torque curves are represented by the coefficients of a Fourier expansion in the angle  $\theta$  between the applied field and a symmetry direction, instead of the usual cubic anisotropy constants. The use of orthogonal functions has the advantage that coefficients already determined are not altered by the inclusion of higher order terms. The torque curves for garnets containing  $Dy$  and  $Yb$  include terms up to at least  $\sin 20\theta$ . There are significant differences in the torque curves obtained for  $Dy$  and  $Yb$  in  $YIG$  and  $GdIG$ . The torque curves for the  $Dy$  garnets indicate that the anomalies observed in the resonance study of this ion are not due to a crossing of the energy levels of the  $Dy$  ion.<sup>3</sup> The origin of the field dependence of the torque and its relation to the angular dependence of the magnetic moments of the rare earth ions will be discussed.

<sup>1</sup>R. F. Pearson and R. W. Cooper, *J. Appl. Phys.* **32**, 265S (1961).

<sup>2</sup>J. F. Dillon, Jr. and J. W. Nielsen, *Phys. Rev.* **120**, 105 (1960).

<sup>3</sup>C. Kittel, *Phys. Rev.* **117**, 681 (1960).



### 93. SPINWAVE LINEWIDTHS IN RARE EARTH SUBSTITUTED YTTRIUM IRON GARNET

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The temperature dependence of the spinwave linewidths have been measured for a series of yttrium iron garnets substituted with the order of one atomic per cent of the rare earths and the results fitted to the theory of de Gennes, Kittel, and Portis.<sup>1</sup> The spinwave measured is the z-directed degenerate spinwave associated with the main resonance nonlinear effect. It is believed that the spinwave linewidth is more reliable than the uniform precession linewidth in these materials since many other mechanisms that affect the uniform precession linewidth will not affect the short wavelength spinwaves. In most cases the data for the spinwave linewidths do show a better fit to the theory of linewidth in these materials than the data for the uniform precession linewidths.

A new technique has been developed that eliminates the need for extensive point by point measurements previously necessary to obtain  $\Delta H_K$ . The new technique records the saturation curve directly, allowing one to obtain the threshold field for the nonlinear effect from an extrapolation done on the recording.

Low temperature data taken on Er, Ho, and Tm substituted yttrium iron garnet from 4.2 to 30°K show approximately a  $T^{3/2}$  dependence for  $\Delta H_K$  which using the theory of de Gennes, Kittel, and Portis, indicates a  $T^{-3/2}$  dependence for the rare earth ion relaxation time. In the temperature region from 70 to 300°K the temperature dependence for  $\Delta H_K$  is very close to  $T^{-1}$  for Er, Sm and Tm, indicating a rare earth ion relaxation time that is independent of temperature. For Ho the data in the higher temperature range yield  $T^{-2.6}$  for  $\Delta H_K$  resulting in approximately a  $T^{-3/2}$  dependence for the rare earth ion relaxation time over both temperature ranges in this material. Measurements are presently being extended to other rare earth ions and higher temperatures.

<sup>1</sup>P. G. de Gennes, C. Kittel, and A. M. Portis, Phys. Rev., 116, 323 (1959).

### 94. HIGH FIELD LOW TEMPERATURE MAGNETIZATION STUDIES OF PRASEODYMIUM SUBSTITUTED YTTRIUM IRON GARNET $5\text{Fe}_2\text{O}_3 \cdot X \text{Pr}_2\text{O}_3 \cdot (3-X)\text{Y}_2\text{O}_3$

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An investigation of the magnetization of  $5\text{Fe}_2\text{O}_3 \cdot X \text{Pr}_2\text{O}_3 \cdot (3-X)\text{Y}_2\text{O}_3$  has been carried out in the liquid helium range and in magnetic fields up to 95,000 gauss,\* in which  $X = 0.25, 0.50, 0.75, 1.00, 1.25, 1.50$  and  $1.75$  moles. At 95,000 gauss and 4.2°K saturation is approached, except for  $X = 1.75$ . At this concentration, the magnetization

is linear in magnetic field between 20,000 gauss and 90,000 gauss with a slope of  $2 \times 10^{-5}$  Bohr magnetons per atom per gauss. The magnetization at 90,000 and 4.2°K is nearly linear in the concentration of praseodymium. The magnetization per formula is given by

$$\bar{\mu}_F = 9.6 + 3.75 X$$

in Bohr magnetons formula. An apparent maximum is reached at a concentration somewhat less than  $X = 1.50$ , the magnetization at 1.75 being 11.2 Bohr magnetons per garnet formula. The magnetization results here obtained indicate an effect of ion size on the distribution of ions in the garnet sublattice.

\*The high magnetic fields were provided at the Low Temperature Laboratory of the University of California, Berkeley.

### 95. PARAMAGNETIC RESONANCE OF $\text{Gd}^{+++}$ IN YTTRIUM GALLIUM GARNET

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We have investigated the EPR spectrum of yttrium gallium garnet single crystals<sup>1</sup> doped with gadolinium, present in the form of  $\text{Gd}^{+++}$ , substituting for yttrium ions. The experiments were performed at a frequency of 35 kMcps.

For an arbitrary orientation of the magnetic field there are six inequivalent ion sites, each having three mutually perpendicular two-fold axes of symmetry. The analysis of the angular dependence of the EPR spectrum shows that two of these axes lie in a (100) plane, the third being of necessity the [100] direction normal to this plane. The two axes in this plane are parallel (within 1°) to the two [110] directions in this plane.

The various possibilities for combining three such directions yield six inequivalent ions in a cubic unit cell, for a general orientation of the applied field  $H$ . The angular dependence of the spectrum may be described by the following spin Hamiltonian, for which the coordinate axes are the two-fold axes and the z direction is a [110]:

$$H = b_{20}Y_{20} + b_{2,2}(Y_{2,2} + Y_{2,-2}) + b_{40}Y_{40} + b_{4,4}(Y_{4,4} + Y_{4,-4}) + b_{60}Y_{60} + b_{6,4}(Y_{6,4} + Y_{6,-4}) + g\beta H_0 S$$

The coefficients  $Y_{L,M}$  are polynomials in the components of the spin  $S$ , that transform like the corresponding spherical harmonics. The coefficients  $b_{L,M}$  are those used by Geschwind.<sup>2</sup> We choose as z the [110] direction for which the splitting due to the fourth order terms is maximum. At room temperature we obtain (only relative signs are important):

$$g = 1.992 \quad b_{20} = .0440 \text{ cm}^{-1} \quad b_{2,2} = -.0269 \text{ cm}^{-1} \\ b_{40} = 4.21 \times 10^{-3} \text{ cm}^{-1} \quad b_{4,4} = 2.11 \times 10^{-3} \text{ cm}^{-1} \quad b_{60} = -2.0 \times 10^{-4} \text{ cm}^{-1} \\ b_{6,4} = -3.5 \times 10^{-4} \text{ cm}^{-1}$$

The ratio of the fourth order coefficients indicates that these are not axes of cubic symmetry. It is interesting to note, however, that if one rotates around z by  $\alpha = 36.7^\circ$  the



correct ratio is obtained in the new system of axes. This is still true for the less accurately known 6th order coefficients. Thus, it seems that the 4th and 6th order terms in (1) correspond essentially to a cubic field with one axis parallel to a [110] direction and the other two in the (110) plane, one of these being very close to the [111] as indicated by the value of  $\alpha$ .

A discussion of these results insofar as they relate to calculations of the magneto-crystalline anisotropy in gadolinium iron garnet will be presented.

<sup>1</sup>Crystals were prepared by Mr. C. Quadros of Harvard University.

<sup>2</sup>S. Geschwind and J. P. Remeika, Phys. Rev. **122**, 757 (1961).

## 96. NEUTRON MAGNETIC SCATTERING FROM RARE EARTH IONS

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Unlike the case for the iron series transition metals, where the orbital angular momentum of the 3d electrons is almost completely quenched, the magnetic scattering of neutrons from the rare earths (except for europium and gadolinium) arises from both the spin and orbital contributions to the magnetization. Using recently determined Hartree-Fock wave functions for the trivalent rare earth ions we have determined both the spin and orbital contributions to the magnetic form factors. Asymmetries due to the non-spherical 4f spin densities were determined in the manner of Freeman<sup>1</sup> and Weiss and Freeman<sup>2</sup> whereas for the orbital scattering the methods of Trammell<sup>3</sup> and Blume<sup>4</sup> were followed in the calculation. In order to estimate the contribution of the core electrons to the scattering we also carried out a spin or exchange polarized Hartree-Fock calculation for Gd<sup>3+</sup> and from the resultant spin density determined the way this changes the purely 4f form factor.

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<sup>1</sup>A. J. Freeman, Phys. Rev. **113**, 169 (1959) and Acta Cryst. **12**, 261 (1959).

<sup>2</sup>R. J. Weiss and A. J. Freeman, J. Phys. Chem. Solids **10**, 147 (1959).

<sup>3</sup>G. T. Trammell, Phys. Rev. **92**, 1387 (1953).

<sup>4</sup>M. Blume, Phys. Rev. (to appear).

## 97. HIGH TEMPERATURE SUSCEPTIBILITY OF GARNETS: EXCHANGE INTERACTIONS IN YIG AND LuIG

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Power series expansions of the high temperature susceptibility and its inverse in ascending powers of the

reciprocal temperature have been obtained through the use of an extension<sup>1</sup> of the method of Rushbrooke and Wood<sup>2</sup>. The Heisenberg form of exchange is adopted, and in this paper, interactions between neighboring spins from different sublattices (a-d exchange) only are included. The coefficients in the series are derived for general spins and arbitrary g-factors on the two sublattices. The calculations have been carried out to terms including the fifth power of the exchange divided by the temperature; the molecular field theory by contrast is rigorously valid only to the first power term of its expansion.

The inverse susceptibility series have been used to determine the magnitudes of the a-d exchange interactions in the ferrimagnets, yttrium iron garnet (YIG) and lutetium iron garnet (LuIG). The intra-sublattice exchange interactions are assumed to be zero. This is in keeping, however, with an increasing suspicion on the part of numerous investigators that these interactions play a minor role in the garnets and have been seriously overestimated by molecular field considerations. The recent experimental data of Aléonard<sup>3</sup> was treated by two separate procedures. First, the Curie constant was taken to be fixed at the spin-only value,  $C = 43.78$ . A least-squaring method then gave the following for the a-d exchange:  $J/K = -35.7^\circ\text{K}$  for YIG and  $-37.2^\circ\text{K}$  for LuIG. Secondly, both C and J were adjustable, and a least-squaring procedure yielded.  $C = 43.3$ ,  $J/K = -35.0^\circ\text{K}$  for YIG and  $C = 42.5$ ,  $J/K = -34.5^\circ\text{K}$  for LuIG. In both cases the deviation of the theory from experiment was about 1-2 per cent (within the range of the practical convergence of the truncated series). For YIG,  $J/K = -35.0^\circ\text{K}$  corresponds to a value of the Landau-Lifschitz stiffness constant, A, of  $6.1 \times 10^{-7}$  erg/cm, and to a value of the spin-wave dispersion relation constant, D of  $1.15 \times 10^{-28}$  erg/cm<sup>2</sup>. These compare favorably with the values  $A = 4.4 \times 10^{-7}$  and  $D = 0.85 \times 10^{-28}$  obtained from the low temperature heat capacity<sup>4</sup>, and with the value  $D = 0.99 \times 10^{-28}$  obtained from spin-wave spectra<sup>5</sup>. It is concluded that these results support the hypothesis of relatively weak intra-sublattice exchange in the garnets.

<sup>1</sup>P. J. Wojtowicz, J. Appl. Phys. **31**, 265S (1960).

<sup>2</sup>G. S. Rushbrooke and P. J. Wood, Mol. Phys. **1**, 257 (1958).

<sup>3</sup>R. Aléonard, J. Phys. Chem. Solids **15**, 167 (1960).

<sup>4</sup>Kunzler, Walker and Galt, Phys. Rev. **119**, 1609 (1960); S. S. Shinozaki, Phys. Rev. **122**, 388 (1961).

<sup>5</sup>E. H. Turner, Phys. Rev. Lett. **5**, 100 (1961); R. C. LeCraw and L. R. Walker, J. Appl. Phys. **32**, 167S (1961).



# SESSION VIB

## DEVICES AND PHENOMENA

C. L. HOGAN, Presiding

### 98. A COAXIAL FERRITE PHASE SHIFTER FOR HIGH POWER APPLICATIONS\*

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The low and high-power behavior of a coaxial ferrite-loaded transmission line has been studied experimentally at microwave frequencies. Longitudinal magnetic biasing fields both above and below the cut-off range ( $\frac{\omega}{\gamma} - 4\pi M_S \leq H_{dc} \leq \frac{\omega}{\gamma}$ ) were explored, the two regions yielding devices with markedly different characteristics. The mechanical structure consisted of a rigid  $\frac{7}{8}$ -in. coaxial line fully loaded with Trans-Tech 1-103 ferrite together with appropriate quarter-wave dielectric transformers.

(1) Operation below cut-off ( $0 \leq H_{dc} < \frac{\omega}{\gamma} - 4\pi M_S$ ),  
 $f = 3000$  Mc

These parameters permitted construction of a compact, light-weight phase shifter producing  $360^\circ$  of phase shift in 10 inches of active length upon application of only 100 oe. A figure of merit of 180 deg/db or more was maintained up to a peak power of 10 kw.

(2) Operation above resonance ( $H_{dc} > \frac{\omega}{\gamma}$ ),  $f = 1350$  Mc

With these parameters, a device was built that is capable of handling at least several hundred kilowatts of rf peak power while maintaining a figure of merit of 1000 or more. The loaded coaxial line provided  $360^\circ$  of phase shift in 24 inches of active length when biased from 950 to 1350 oe.

The high-power and loss characteristics of the latter device permit application to electronic steering of the transmitter beam in a linear array antenna where relatively few phase shifters suffice. The low-field device is suited for beam control in a planar array where large numbers of relatively small phase shifters of medium peak-power capability are needed.

\*Supported by the Bureau of Ships, Department of the Navy, Contract No. NObsr-77570.

### 99. SLOW-WAVE UHF FERRITE PHASE SHIFTERS\*

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The adaptability of periodic coaxial, periodic strip, and helical transmission lines for use in UHF ferrite phase shifters is discussed. In general, it was found that the periodic strip transmission line was best suited for use in a UHF ferrite phase shifter. Gains of up to 5 times in the phase shift per unit length were realized through the use of

the slow-velocity striplines rather than the conventional strip transmission lines.

It is concluded that, even though considerable gains in the phase shift per unit length can be realized with slow velocity structures, it does not necessarily follow that better phase shifters can be realized through their use. The disadvantages of the slow-velocity strip transmission line ferrite phase shifter, in comparison to the conventional strip transmission line ferrite phase shifter, is brought out by the lower merit factors (degrees of phase shift per unit absorption) produced by the slow-velocity structures, and the larger cross-sectional areas required by the slow-velocity structures.

The volt-ampere-microsecond product for a phase shifter is defined, and results of calculations are discussed for  $360^\circ$  phase shifters utilizing slow-velocity and conventional stripline structures, showing that the switching volt-ampere-microsecond product of the slow-velocity stripline phase shifter can be 2.5 times greater than that of the conventional transmission line.

The real and imaginary parts of the permeability have been measured for a very large percentage of the commercially available ferrites and garnets. Results of this study are given over the frequency spectrum of 400-1000 Mc. From these spectra, the materials are categorized into one or more of five possible low-loss phase shifting regions. The regions are defined according to the various possible behaviors of the ferrimagnetic loss mechanisms upon the applied magnetic field strength.

A brief summary of the permeability and loss mechanisms of ferrimagnetic materials in the UHF range is presented together with experimental data which partially confirm the theory. Methods for the proper choice of material for phase shifting devices are discussed in terms of the material constants and the desired operating frequency range.

The paper stresses the importance of the ferrimagnetic material which is chosen as a phase shifting medium for any particular device design at frequencies below 1000 Mc.

\*This work was supported by the Air Force Cambridge Research Center under contract AF 19(604)-6171.

### 100. EFFICIENT FREQUENCY DOUBLING FROM FERRITES AT THE 100-WATT LEVEL

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Efficient frequency doubling at the 100-watt level from 8.5 to 17 Gc has been accomplished with disk-shaped ferrite specimens in a doubly-resonant cavity. Within the tolerances imposed by the uniformity of the material the performance is repeatable for specimens of the same type of ferrite, but of different batches. It is felt that this represents an improvement in the state of the art, since in earlier work:<sup>1</sup> (1) efficient frequency doubling was reported possible only with input power levels of the order of 10 kw; (2) experimental results, though outstanding in efficiency, could not be correlated with theoretical expectations based on line width and  $M_S$  criteria; (3) repeatability for specimens of different batches was sometimes difficult.



From results of experimental studies of harmonic generation of a more basic nature,<sup>2</sup> we selected single crystal manganese ferrite as the most promising material. As an example of the results, we achieved 18 watts (17 Gc) delivered to an external load, for 155 watts of total drive power (8.5 Gc), of which 125 watts were absorbed by the ferrite.

An important aspect of this experiment was the development of a cavity-ferrite system that satisfies three conditions simultaneously: (1) to maximize the fundamental field intensity, the loaded cavity is resonant to the drive frequency  $\omega$ ; (2) the ferrite is at ferromagnetic resonance at  $\omega$ ; (3) the cavity is resonant to the output frequency  $2\omega$ . This could not be achieved with the usual doubly-resonant cavity, where the entire resonant volume of one mode is common to that of the other. In the present cavity, the  $2\omega$  mode is confined to a fraction of the cavity structure, whereas the  $\omega$  fields occupy the entire volume. By putting a variable short in the region where only the  $\omega$  fields exist, the  $\omega$ -resonant frequency can be varied without altering the resonance at  $2\omega$ . The pronounced detuning effect (at  $2\omega$ ) of the large volume of ferrite necessary for efficient generation is easily compensated for in this new cavity.

For fixed susceptibility  $\chi'$  at ferromagnetic resonance, the conversion efficiency in frequency doubling increases linearly with input power. Although our basic measurements<sup>2</sup> showed an increase in linewidth with drive level, they, nevertheless, showed a continuing increase in conversion efficiency. This would indicate that efficiencies higher than stated above should be possible at higher drive levels. Actually, we found the limit in power level of our experiment given by relaxation oscillation phenomena similar to those observed by M. T. Weiss.<sup>3</sup>

<sup>1</sup> J. H. Melchor, W. P. Ayres, and P. H. Vartanian, Proc. Radio Engrs. **45**, 644 (1957).

<sup>2</sup> D. D. Douthett, I. Kaufman, and A. S. Risley, "Microwave Harmonic Generation by Ferrimagnetic Crystals." Accepted for publication in J. Appl. Phys.

<sup>3</sup> M. T. Weiss, J. Appl. Phys., **31**, 103S (1960).

## 101. A MAGNETO-DYNAMIC MODE FERRITE AMPLIFIER

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A ferrite amplifier has been operated using two magneto-dynamic modes for the signal and idler resonant circuits. This is in contrast to the more usual electromagnetic cavity type resonance or the ferrite magnetostatic modes. These magneto-dynamic modes, which have been described by Heller<sup>1</sup> and by Coleman and Steier<sup>2</sup> for ferrite cylinders, result from the coupling of a cavity type resonance with a magnetostatic resonance. The coupling or mixing is strongest when the two unperturbed resonant frequencies are the same.

For the amplifier described in this paper a single crystal yttrium iron garnet sphere was used. The lowest order cavity mode has a resonant frequency of about 18.5

kmc for this size sphere. The magnetic bias field applied was  $17 \text{ kMc}/\gamma$  oersteds. The resulting magneto-dynamic modes were resonant at 21 kMc and 14 kMc, respectively, for the two branches of the mode tuning curve.

Coleman and Steier have pointed out some of the advantages of these modes for ferrite amplification. These are: (1) ease of coupling, (2) lack of interference from other resonant modes.

This second advantage can be of considerable importance. In the amplifier described in this paper both the signal and the idler mode lay outside of the limits of the Walker magnetostatic mode spectrum. This not only eases the problem of selectively coupling into the signal circuit but also minimizes one potential source of noise, that of signal coupling to a non-thermally excited magnetostatic mode.

The amplifier described in this paper operated at a signal frequency of 21 kMc. Parallel pumping was used at a pump frequency of 35 kMc. Additional information on both the amplifier performance and the magneto-dynamic modes is given in the paper.

<sup>1</sup> G. S. Heller, "Ferrite Loaded Cavity Resonators," MIT Lincoln Lab Report M 82-5, May 6, 1959.

<sup>2</sup> W. H. Steier, P. D. Coleman, J. Appl. Phys., **30**, 1454 (1959).

## 102. A MINIATURIZED FERRIMAGNETIC HIGH POWER COAXIAL DUPLEXER-LIMITER\*

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The device to be described in this presentation is a miniaturized high power coaxial component performing within a single package the functions of circulation, power limiting, and preselection. Attention is principally directed to the limiting and preselection functions which are accomplished through the use of a gyromagnetic coupler which differs basically from those previously reported in that the second order non-linear process rather than the first order process is the basis for its operation. This limiter has been tested independently at power levels as high as 8kw and found to exhibit a flat leakage output less than 200 mw and a spike leakage energy of about 0.5 erg. The limiter is mechanically or electrically tunable over a frequency range greater than ten percent in C-band. At a fixed setting the 3db bandwidth is about 20 Mc. The recovery time of the limiter is determined by the feedback of energy from the garnet spin system. Experimental values of this quantity will be presented. The duplexer-limiter package operates at peak powers up to 40 kw into a 2:1 antenna mismatch. Input vswr at the transmitter port is less than 1.35. The device is tunable from 5.4 to 5.9 kMc. Insertion loss from the transmitter port to the antenna port is less than 0.7 db. Low power insertion loss from antenna port through the limiter to the receiver port is 1.0 db.

\*This work was supported by the U.S. Army Signal Corps, Fort Monmouth, New Jersey.



### 103. DESIGN CONSIDERATIONS FOR BROADBAND SYMMETRICAL "WYE" TUNABLE FERRITE CIRCULATORS IN TEN MODE STRIP TRANSMISSION LINE

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This paper describes a differential dielectric and ferrite loading technique which has been successfully employed in the design of a series of broadband tunable "wye" type ferrite circulators in dielectric filled strip transmission line propagating the dominant TEM mode. These circulators are capable of being rapidly tuned (external dc electromagnet) over octave bandwidths, in the 400 Mcs to 8 kMc frequency range, and exhibit typical forward insertion loss characteristics in excess of 20 db.

The basic physical and mathematical design equations governing the use of this technique are initially described in terms of conventional lumped parameter low frequency network equivalent circuits. These transmission line relationships are then described in terms of the particular microwave frequency non-reciprocal rf propagation characteristics exhibited by this class of symmetrical "wye" circulators in TEM mode strip transmission line. It is shown that through the application of a mode distortion technique employed in the design of non-reciprocal ferrite devices in coaxial transmission line propagating the dominant TEM mode, it has been possible to achieve a differential rf phase propagation characteristic at the ferrite dielectric interface located at the transmission line junction of the circulator. A dielectric ring loading technique is also described. This technique is shown to eliminate the frequency sensitivity exhibited by the junction of this type of circulator, and also maintain the proper rf field phase distribution in the vicinity of the ferrite dielectric interface. It is shown that by varying the external applied dc field through the ferrite as a function of input rf frequency, the conditions required for achieving attenuation ratios in excess of 35:1 db over the previously indicated bandwidths may be satisfied. First order approximations of the propagation equations which govern the performance of this type of symmetrical, non-reciprocal transmission line structure are presented in terms of the general mathematical synthesis employed in analyzing the performance of equivalent limited bandwidth symmetrical wye structures. Finally typical performance data for an L-Band, S-Band and C-Band Broadband Structure are presented.

### 104. NONLINEAR EFFECTS IN FERRITE FILLED REDUCED SIZE WAVEGUIDE\*

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Ferrite filled waveguides of arbitrarily small cross-section can support two distinct modes of propagation, the normal dominant mode and the birefringent modes. The presence of these modes is dependent upon the relation between the off diagonal component  $\kappa$ , and the diagonal component  $\mu$  of the permeability tensor. In the region  $\mu^2 - \kappa^2 > 0$ ,

$\kappa < 0$ , of the dominant mode, the ferrite material behaves as low loss high index of refraction dielectric. It is characterized by the intense concentration of the r-f magnetic field as  $\mu$  and  $\kappa$  approach equivalent numerical values. The region of the birefringent modes from  $\mu^2 - \kappa^2 = 0$  to  $\mu = 0$  is similarly characterized by an intense concentration of rf field but with higher losses related to a reduction in the velocity of propagation. Nonlinear effects are observed in both modes of propagation.

Nonlinear effects were investigated at 9375 Mc in rectangular and circular waveguide with transverse and axial applied magnetic fields respectively. The cross sectional area of standard X-band waveguide was reduced down to as low as 1/10 its original value, maintaining approximately the same aspect ratio. Factors investigated were insertion loss, threshold power level, spike characteristics, dynamic range of limiting and breakdown power level. These are discussed with relation to waveguide size, frequency, ferrite loading geometry, ferrite material parameters and temperature.

Thresholds as low as 3 watts peak were observed using standard polycrystalline ferrite materials. Spike widths of less than 40 nanoseconds were observed with spike amplitude decreasing as much as 8 db over a 20 db limiting range. It was possible to realize useable limiter configurations whose low power losses were less than 1 db.

The application of these effects in the design of a compact X-band limiter are mentioned. The advantages and limitations of devices utilizing these effects are also discussed.

\*This work was supported by the U.S. Navy Department, Bureau of Ships, under contract NObSr 81244.

### 105. MILLIMETER WAVE PARAMETRIC AMPLIFICATION WITH ANTIFERROMAGNETIC MATERIALS

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The use of material with natural magnetic resonances in the millimeter and submillimeter wave bands makes possible electro-magnetic interaction without an extremely large applied magnetic field. Antiferromagnetic materials most suitable for microwave application  $\text{Cr}_2\text{O}_3$  and  $\text{MnF}_2$  display resonances at approximately 160 and 250 kMc.

In the presence of a magnetic field applied parallel to the sublattice magnetizations the uniform precessional mode resonates at different frequencies for oppositely directed circularly polarized excitation. A perturbation analysis has been conducted to determine the parametric response of the oppositely directed uniform precessional modes to longitudinal pumping. For materials with a sufficiently narrow linewidth very close coupling between the uniform precessional mode and an external coaxial, strip-line or waveguide signal circuit is possible. Calculations have been conducted on the response of an antiferromagnetic medium in a silver cavity resonant to the pump frequency. The pump power absorbed by the cavity at the threshold of amplification for  $\text{Cr}_2\text{O}_3$  would be  $2(10)^4$  watts.



Narrower line width materials are needed for efficient anti-ferromagnetic millimeter and submillimeter amplification.

A more efficient utilization of the pump power and a simplified structure for millimeter wave application results when the pump fields are supported by a dielectric rod mode resonance. If the rod is placed across the narrow dimension of rectangular waveguide, the broad metallic wall forms the conducting end plates of the  $H_{01}$  mode. The signal  $T_{01}$  waveguide mode couples to the appropriately directed circularly polarized uniform precessional resonance of the antiferromagnetic material on the rod.

## 106. A MICROWAVE MAGNETIC MICROSCOPE

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A microwave magnetic probe has been developed which is capable of measuring the spatial variations of the magnetic properties of magnetic materials. In contrast to other forms of microscopes, this one measures the spatial variations of the important magnetic properties of a sample directly. By a selective microwave resonance technique, the gyromagnetic ratio, magnetization, line width, exchange constant, surface and anisotropic energies, etc. of small portions of a ferromagnet may be determined selectively. Actual experimental results using this probe will be presented to illustrate its possible utility.

By placing the magnetic sample against the outer face of the back wall of a  $TE_{10n}$  cavity which has a small centered hole, only that portion of the material that is directly opposite the opening is exposed to the microwave radiation from the cavity. By varying the static magnetic field applied to the sample, the resonance spectrum of that portion of the sample may be determined. From this result, many of the important magnetic properties of this selected portion may be ascertained. The magnetic properties of other portions of the material may likewise be obtained by shifting these portions against the cavity hole successively. In this way, the spatial variation of the magnetic properties of the entire sample may be determined. These results may be used, among other things, to study the macroscopic structure and nature of imperfections of magnetic materials.

To date, it has been possible to measure the magnetic properties of a thin Permalloy film,  $\sim 1000$  Å placed outside the back wall of an x-band cavity with the film excited to resonance through a  $25 \mu$  hole. This represents a sample to cavity volume ratio of one part in  $10^{13}$ . For bulk insulating magnetic materials, the sensitivity could be further increased, i.e. it would be possible to examine even smaller portions of such materials selectively. Since the hole at the back wall of the cavity acts essentially like a waveguide beyond cutoff, the back wall must be made thin compared to the hole diameter to avoid excessive attenuation.<sup>1</sup> By using a cavity made of superconducting material, it is anticipated that portions of materials as small as 200 Å in cross-section may be examined.

To increase the sensitivity of the microscope, it is desirable to modulate the static magnetic field by a small

alternating field and detecting the modulating signal. This is so because the crystal noise decreases with frequency and that narrow-band amplification may then be used. However, at the high sensitivity required for the microscope, microphonic oscillations of the cavity walls give rise to undesirable signals at the modulation frequency. Means to overcome this will be briefly discussed.

\*Operated with support from the U.S. Army, Navy, and Air Force.

<sup>1</sup>R. F. Soohoo, Quarterly Progress Report on Solid State Research, Lincoln Laboratory, M.I.T. (15 April 1961), p. 62.

## 107. SAMPLING MAGNETOMETER BASED ON THE HALL EFFECT

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The design, construction, and performance criteria of a stroboscopic Hall effect magnetometer are described in detail. The purpose of this device is the measurement and recording of the amplitude, waveform, and direction of periodic magnetic fields of arbitrary waveshape and the mapping of the magnetic fields surrounding a radiating magnetic dipole. The magnetometer is based upon the pulsed operation of semiconductor intermetallic thin film Hall effect detectors. Narrow, rectangular current pulses are applied to a Hall plate of a thin film of evaporated indium antimonide. These current pulses are phase modulated, causing the periodic magnetic field orthogonal to the Hall plate to be sampled and scanned point by point. The resultant Hall voltage, which is orthogonal to both the pulse current and the magnetic field produces, after suitable integration and amplification, a wave form which is a low-frequency replica and synthesis of the parameters of the high-frequency magnetic field. Pulsed operation of thin film Hall effect detectors provides an increase in sensitivity over cw operation of conventional Hall devices. This is due, in part, to the improved thermal dissipation of the film, the reduction in joule heating, and the greatly improved noise discrimination because of stroboscopic sampling. A sensitivity of better than  $10^{-4}$  volts/gauss, over a frequency range between 100 cps to 5 Mc and an angular orientation sensitivity of  $10^{-5}$  volts/degree have been obtained in devices built to date. Linearity to better than 2 per cent is retained between the Hall voltage output amplitude and magnetic fields ranging from  $10^{-3}$  gauss to  $10^4$  gauss. Control of the evaporation process in order to obtain desired impurity doping for temperature compensation of the InSb film will be described, as well as details of the magnetic circuit and pulse drive apparatus. Results obtained in the stroboscopic mapping of magnetic fields surrounding various radiators will be illustrated in addition to field intensity profiles for a number of radio frequency resonators.



## 108. NEW TYPE OF FLUX-GATE MAGNETOMETER

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The use of an ordinary toroidal core without air-gap as a flux-gate magnetometer has been overlooked. A magnetometer using such a core as the field-sensitive element with semicircularly wound and differentially connected second-harmonic detector windings has been developed. The semicircle portions of a nickel-iron-alloy ring core act here like two separate cores, corresponding to the two parallel nickel-iron-alloy strips or scrolls of conventional forms of flux-gate elements, as far as second-harmonic flux components are concerned. By using tape-wound or laminated (washer-type) Supermalloy cores having i.d.-o.d. ratios in the range from 0.85 to 0.98, a sensitivity of 1,000  $\mu$  amp/oe or 1 v/oe can be achieved. Such "ring-core flux-gate elements" make it possible to: (1) use ultrathin, 1/8-mil tape and correspondingly high excitation frequencies, 10 to 40 kc, (2) make "point measurements" by reducing the core diameter to 0.5 in., or less, (3) minimize the magnetizing-current requirements, (4) obtain linear characteristics, (5) eliminate memory effects, (6) facilitate matching of the magnetic characteristics of the two active parts of the flux-gate element which belong here to the same core, (7) detect very small changes in the earth's magnetic field, and (8) apply multiple detector windings on a common core. The power requirements of a portable magnetometer, operated from a 6 v battery, have been reduced to 50 mw by combining the ring-core flux-gate element with a switching-transistor magnetic-coupled multi-vibrator in such a way that the oscillation frequency is solely determined by the parameters of the ring core and its excitation windings.

## 109. WAVEFORM OF THE TIME RATE OF CHANGE OF TOTAL FLUX FOR MINIMUM CORE LOSS

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In testing magnetic materials core losses are measured when the time rate of change of total flux,  $d\phi/dt$ , in the core varies sinusoidally since the design engineer usually bases his calculations on sine wave values. The ASTM standard method of measuring core loss using an Epstein frame is designed<sup>1</sup> so that  $d\phi/dt$  is sinusoidal for values of peak induction up to 12 to 15 kilogauss depending on the material being tested. When  $d\phi/dt$  is not sinusoidal as is generally the case at flux densities above 15 kilogauss, a correction is applied to the observed core loss to obtain the sine wave core loss values. In order to avoid the use of correction factors, much effort has been directed toward maintaining a sinusoidal  $d\phi/dt$  at these high flux densities.<sup>2-5</sup>

Although it may seem intuitive that the core loss for a given flux density is a minimum when  $d\phi/dt$  is sinusoidal, measurements and a simple analysis indicate otherwise. It is the purpose of this work to show that the core loss is smaller if the time rate of change of total flux in the core

follows a square wave than it would be if the time rate of change of total flux followed a sine wave with the peak induction being the same in both cases. By a variational method it is shown that for a given value of peak flux density the ratio of square wave to sine wave eddy current losses is  $8/\pi^2$ .

The ratio of square wave to sine wave core losses is measured as a function of induction for a fixed frequency and as a function of frequency for a fixed induction using a HIPERSIL specimen (cube on edge orientation). A feedback circuit<sup>4</sup> is used to prevent distortion in the waveform. When the measurement is made at 60 cps the ratio is about 0.90 for flux densities below 12.5 kilogauss and rises with increasing flux density to about 0.96 at 17.5 kilogauss. For an induction of 5 kilogauss the ratio decreases from about 0.92 at 30 cps to 0.87 at 120 cps.

<sup>1</sup> ASTM Standards, A34 (1958)

<sup>2</sup> W. Cormack, Trans. South African Inst. Elect. Engrs. **38**, 257, (1947)

<sup>3</sup> S. C. Leonard and R. L. Tenbroeck, Trans. Am. Inst. Elect. Engrs. **76**, pt. 3, 118 (1957)

<sup>4</sup> J. McFarlane and M. J. Harris, J. Inst. Elec. Engrs. (London) **105A**, 395 (1958)

<sup>5</sup> W. P. Harris and I. L. Cooter, J. Research Natl. Bur. Standards **60**, 509 (1958)

## 110. IRON LOSSES IN ELLIPTICALLY ROTATING FIELDS

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Power frequency losses in silicon-iron alloys due to an elliptically rotating magnetic field have been measured by a calorimetric technique and predicted approximately using a simple model. Previously reported experiments for measuring these fail to approach saturation and have doubtful field uniformity. A thin disc-shaped specimen is placed in the elliptically rotating field of a set of two phase air-cored rectangular Helmholtz-type coils. The 60 cps rotational loss is calculated from the initial rate of specimen temperature rise sensed by a copper-constantan thermocouple attached to the disc. The thermocouple output is amplified and recorded with an accuracy of  $10^{-3}$  °C. The field coil system produces fields uniform to 2 per cent in a 1.0 in. diameter region and of strengths up to 250 oe which is sufficient to approach saturation in 0.005 in. thick by 0.875 in. diameter discs. The losses have been measured for grain oriented and single crystal materials with all materials showing a decrease in loss for an increase in flux density for large nearly circular fields. This effect is more pronounced for rotation in the (100) plane than in the (110) plane. The losses for elliptical magnetization may be approximated with a simple model composed of a mixture of lossless domain rotation, eddy current loss, and alternating hysteresis loss proportional to an alternating flux. This model is intended only for the calculation of rotational hysteresis losses and not to explain the origin of these losses. The calculated and experimental losses agree for all amplitudes and eccentricities of magnetization to within 40 per cent for rotation in the (100) plane with the worst discrepancy for circularly rotating magnetization.



## 111. LOW FREQUENCY LOSSES AND DOMAIN BOUNDARY MOVEMENTS IN SILICON IRON

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Earlier observations of very low frequency losses in (110) [001] textured silicon iron have been extended to include losses produced by excitation with a sinusoidal field intensity. Excitation frequencies of 0.005 cps to 10 cps and flux density amplitudes of 10, 15 and 17 kilogauss were used in these measurements. The downward concavity of the loss-frequency curve near zero frequency, previously reported for sinusoidal induction, is greatly exaggerated by imposition of a sinusoidal field intensity.

To further investigate these low frequency loss anomalies, low amplitude, high frequency fields were impressed on the material during cyclic magnetization at low frequencies. The superposed field, most often of 320 cps or 1000 cps frequency, had an amplitude considerably smaller than the coercive force of the material. Waveforms of the gross, low frequency magnetization and the incremental, high frequency magnetization were accurately controlled by high gain, direct coupled, feedback amplifiers. The ratio of the B amplitude to the H amplitude for the high frequency components varied greatly within a cycle of magnetization.  $|B|/|H|$  was also found to vary with the peak value and waveform of the gross induction. Most significantly, the ratio increased greatly with frequency of the gross magnetization at very low frequencies. These changes can be attributed to an increase in the number of domain walls in simultaneous motion when the speed of gross magnetization is increased.

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## 112. PROPERTIES OF RESET CORES IN RADAR PULSE TRANSFORMERS

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This paper outlines recent work done on large uncut toroidal cores of Hipersil and Supermendur. Permeability and equivalent loss resistance are measured for both kinds of material. With forced cooling, large increments of core induction can be utilized without overheating the windings. Under these conditions, and with a reset core, pulse permeability is much higher than it is in cut cores, and therefore alters pulse transformer design radically.

Previously reported work was done on Molybdenum Permalloy at maximum core induction of 10 to 13 kilogauss. Tests reported in this paper were conducted at inductions up to 24 kilogauss. In this range the pulse permeability of fully oriented Hipersil was greater than that of Supermendur, and was in the neighborhood of 6,000 compared to approximately 1,500 for cut cores of 2 mil thickness in both cases. Core loss of Hipersil, calculated from the measurements of exciting current was found to be 50 per cent greater than Supermendur. It further appears from these tests that it would be practicable to operate Hipersil with excursions of induction up to 30 kilogauss and Supermendur

up to 40 kilogauss. By taking full advantage of all factors and precautions in design, it is possible to reduce pulse transformer weight by 80 per cent with a reset core. The reduced weight includes that of a bias choke used in the resetting circuit.



# SESSION VIIA ANTIFERROMAGNETISM AND RESONANCE

M. T. WEISS, *Presiding*

## 113. PERIODIC ADIABATIC TIME VARIATIONS (Invited)

H. SUHL

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It is shown that by modulating certain parameters of a system of weakly interacting constituents, the nature of the interaction can be radically changed: It can be made to vanish in first order, or its sign can be reversed. As a particular case we discuss the suppression of instability in ferromagnetic resonance by modulation of the magnetizing field, the signal frequency, or both. In that case the modulation has the effect of minimizing the interaction between spin waves.

Some more speculative aspect of this method will also be discussed.

## 114. FIELD DEPENDENT MAGNETIC SUSCEPTIBILITY AND MAGNETOSTRICTION IN ANTIFERROMAGNETIC CoO, MnO AND NiO SINGLE CRYSTALS

T. R. MCGUIRE and W. A. CRAPO  
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We have studied domain effects in antiferromagnetics by investigating the behavior of the magnetic susceptibility ( $\chi$ ) and the magnetostriction ( $\Delta\ell/\ell$ ) as a function of magnetic field.  $\chi$  is increased when the electron spin axes are aligned more nearly perpendicular to the applied field either by rotation or by the growth of a domain. These same effects cause magnetostriction because the spin axis orientation is closely related to the crystallographic distortion found in antiferromagnetics.

CoO, MnO and NiO single crystals were annealed at high temperatures to relieve strains and then cooled without stressing. A large magnetostriction was observed in each sample, and was attributed to domain reorientation. Values found at 77°K and 14,000 gauss are  $\Delta\ell/\ell = -9 \times 10^{-6}$  along [110] for MnO,  $\Delta\ell/\ell = -15 \times 10^{-6}$  along [110] for NiO and  $\Delta\ell/\ell = 6 \times 10^{-6}$  for CoO. Our results for CoO are in agreement with values reported by Nakamichi and Yamamoto<sup>1</sup> at lower magnetic fields.

The susceptibility showed large changes with field in only two of the materials studied. The increase in  $\chi$  at 77°K as H varied from 7000 to 21,000 gauss was about 13% for MnO and NiO but less than 1% for CoO. This small change in  $\chi$  and the relatively large magnetostriction in CoO may be due to the relatively large lattice distortion in this material compared to MnO or NiO. In MnO and NiO the field dependence of  $\chi$  can be used to evaluate the anisotropy.

From the model of Keffer and O'Sullivan<sup>2</sup> we calculate an in-the-plane anisotropy,  $K_2 \approx 10^4$  ergs/cc for MnO and  $K_2 \approx 10^3$  ergs/cc for NiO. This model assumes that the spin rotation is confined to (111) planes. The magnetostriction in these materials means that other types of domain behavior are probably present, for example, domain wall motion, and this would modify our  $K_2$  values. In both MnO and NiO we observed a field hysteresis of the susceptibility. Néel has suggested that such an effect is the result of weak spontaneous moments caused by the lack of regular arrangements of antiparallel spins in domain walls.

We wish to acknowledge that the CoO and NiO single crystals were grown by Dr. E. J. Scott of the Naval Ordnance Laboratory while the MnO sample was provided by Dr. Y. Nakazumi of the Tochigi Chemical Company, Osaka.

<sup>1</sup>T. Nakamichi and M. Yamamoto, J. Phys. Soc. Japan 16, 126 (1961).

<sup>2</sup>F. Keffer and W. O'Sullivan, Phys. Rev. 108, 637 (1957).

## 115. PULSED CRITICAL FIELD MEASUREMENTS IN MAGNETIC SYSTEMS

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Massachusetts Institute of Technology  
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Pulsed field antiferromagnetic resonance (AFMR) has been quite successfully applied to several uniaxial antiferromagnets. An alternate technique which requires only magnetic susceptibility measurements is "spin-flop" which is equivalent to AFMR at zero frequency. Recently Jacobs<sup>1</sup> has examined spin-flop in MnF<sub>2</sub> at 4.2°K by measuring M vs H in pulsed fields. We have arranged a carefully balanced pair of coils to measure dm/dt versus H (or t). With careful alignment of uniaxial antiferromagnets in the pulsed field, a  $\Delta M$  or 0.1 gauss can be observed with pulsed fields of 100 k gauss. Further improvement has been achieved with passive filters. The technique has been applied to measurements of the critical field,  $H_C$ , in Cr<sub>2</sub>O<sub>3</sub>, (Cr<sub>2</sub>O<sub>3</sub>)<sub>0.9</sub> · (Al<sub>2</sub>O<sub>3</sub>)<sub>0.1</sub> from 4.2°K up to about 0.9 T<sub>N</sub>. Using the relation  $H_C = [2\lambda K/(1-\alpha)]^{1/2}$ , where  $\lambda$  is the exchange constant, K is the anisotropy energy, and  $\alpha$  is the ratio of parallel to perpendicular susceptibility obtained from independent static susceptibility measurements at low magnetic fields, the quantity  $2\lambda K$  has been calculated and compared to the AFMR data. These values of  $2\lambda K$  versus temperature are in complete agreement with the earlier reported AFMR results<sup>2</sup> for these systems. The method has also been applied to measurements in antiferromagnetic MnF<sub>2</sub> and CoF<sub>2</sub> and metamagnetic CoCl<sub>2</sub> and FeCl<sub>2</sub>. The results of these measurements will be summarized, applications to other magnetic systems will be reviewed, and limitations of the technique will be discussed.

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\*\*Supported by the Air Force through the Air Force Office of Scientific Research.

<sup>1</sup>I. S. Jacobs, J. Appl. Phys. 32, 61S (1961).

<sup>2</sup>S. Foner, J. Appl. Phys. 32, 63S (1961), J. phys. radium 20, 336 (1959).



## 116. ANTIFERROMAGNETIC RESONANCE IN $\text{MnTiO}_3$

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Resonance has been observed in a single crystal of  $\text{MnTiO}_3$ , one of the class of the ilmenites being investigated at this laboratory, which has reportedly an antiferromagnetic Curie temperature of  $41^\circ\text{K}$ .<sup>1</sup> Measurements of resonant field as a function of temperature were made at frequencies from 70 to 140 kMcps using a simple waveguide spectrometer. With the applied magnetic field directed along the *c* axis and at frequencies of 75, 84 and 105 kMcps, the resonant field goes to zero at some temperature in the range from 40 to  $60^\circ\text{K}$  corresponding to the transition from the lower to the upper antiferromagnetic mode. At 140 kMcps, the variation of resonant field with temperature exhibits no zero field resonance showing, therefore, that 140 kMcps is above the zero field resonant frequency.

The data are consistent with the usual resonance equations

$$\omega/\gamma = \sqrt{2\text{H}_E\text{H}_A} \pm \text{H}$$

for the two sublattice model of an antiferromagnetic uniaxial crystal with a critical field,  $\sqrt{2\text{H}_E\text{H}_A}$ , of approximately 46 kilogauss and a *g*-factor close to the free electron spin value.

\*Operated with support from the U. S. Army, Navy, and Air Force.

<sup>1</sup>Y. Ishikawa, J. Phys. Soc. Japan, 13, 1298 (1958).

## 117. ANTIFERROMAGNETIC AND PARAMAGNETIC RESONANCE IN $\text{CuF}_2 \cdot 2\text{H}_2\text{O}$

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We have studied the paramagnetic and antiferromagnetic spectrum in  $\text{CuF}_2 \cdot 2\text{H}_2\text{O}$ . The crystal structure was determined by Geller and Bond who found a monoclinic layer structure with Cu surrounded octahedrally by 2 fluorines and 2 oxygens in the (101) planes. The short Cu-Cu distance is found along the *c* axis. The neutron diffraction data by Abrahams, together with the present work, indicate that the alignment of the Cu spins lie almost along the *c* axis. Along this axis, the spins are parallel, whereas within the (101) planes they are antiparallel. The two antiferromagnetic resonance frequencies,  $w_1$  and  $w_2$ , which are expected for orthorhombic or lower symmetry, were found to occur at almost identical frequencies:  $w_1 = 95.84 \pm .05$  kMc and  $w_2 = 95.91 \pm .05$  kMc.

The field dependence has been predicted from the models of Ubbink and Yosida as well as the zero field lines, and the paramagnetic *g* tensor; a close fit to the observed data has been obtained. The paramagnetic *g* tensor for the only magnetic site occurring was found to have its principal directions along the monoclinic axis, and in the *ac* plane at nearly 45 degrees from the *c* axis. The *g* values were  $g_1 = 2.07$ ,  $g_2 = 2.08$ ,  $g_3 = 2.47$  (at  $T = 47^\circ\text{K}$ ).

A discussion of the superexchange interaction and anisotropy energy is given. From the measured *g* tensor,

the wave functions of ground and excited states are deduced. The ground state wave function is nearly confined to the (101) plane and the strong antiferromagnetic coupling in this plane and the ferromagnetic coupling in the *c* direction can be understood. The anisotropy energy is caused by the dipole interaction and the anisotropic superexchange interaction of pseudo dipolar type. The former has been calculated numerically as:

$$E_d = -.020 \alpha^2 - 0.112 \gamma^2 + 0.030 \alpha \gamma$$

where  $\alpha$ ,  $\beta$  and  $\gamma$  are the direction cosines of the direction of antiferromagnetic ordering with respect to the *a*, *b*, *z* coordinate system with *z* perpendicular to the *a* and *b* axes. The antiferromagnetic resonance measurement shows that the easy direction is in the *ac* plane and  $3^\circ 30'$  from the *c* axis, and that the antiferromagnetic anisotropy is nearly tetragonal. This information makes it possible to deduce the anisotropy energy arising from anisotropic exchange interaction, and hence the value of the total anisotropy. From the perpendicular susceptibility data by Sherwood and Williams and the total anisotropy, the zero field resonance can be predicted to be 130 kMc. The agreement with experiment is not unreasonable in view of the reduction of this frequency by the zero point oscillations of the spin waves.

## 118. FERROMAGNETIC RESONANCE LOSS IN LITHIUM FERRITE AS A FUNCTION OF TEMPERATURE

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Measurements have been made of the intrinsic linewidth of lithium ferrite,  $\text{Li}_{0.5}\text{Fe}_{2.5}\text{O}_4$ , at microwave frequencies as a function of temperature from  $290^\circ\text{K}$  through a low temperature linewidth maximum in the neighborhood of  $30^\circ\text{K}$ . The value of the linewidth maximum is highly anisotropic in much the same way as the low temperature linewidth maximum which has been reported for nickel ferrite.<sup>1</sup> The linewidth maxima at  $30^\circ\text{K}$  are 16 oe in the [111] direction and 2.5 oe in the [100] direction. The data of this paper were taken using parallel pump measurement techniques. The advantage of these measurements is that the role of scattering mechanisms is minimized and the resulting linewidth is the intrinsic linewidth for long-wavelength spin waves.

Data on the loss of lithium ferrite are of particular interest since it is one of the ferrites which in stoichiometric proportions would have all of the iron ions in the ferric state. It has been shown that the presence of ferrous ions in ferrites causes a large increase in linewidth; however, in lithium ferrite the contribution of ferrous ions to the linewidth should be low enough so that the loss due to any ferrous ions which are present can be separated from the loss of ferric lattice. The results of these preliminary investigations can be interpreted in terms of ferrous ion relaxation mechanisms which have been proposed in the literature.

<sup>1</sup>Yager, Galt, and Merritt, Phys. Rev. 26, 1203 (1955).



## 119. IMPURITY ION EFFECTS IN THE FERRIMAGNETIC RESONANCE OF ORDERED LITHIUM FERRITE

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Ordered lithium ferrite and yttrium iron garnet (YIG) differ from all other ferrimagnetic oxides in two structural properties which have an important influence on ferromagnetic resonance phenomena; in pure crystals of these materials all the magnetic ions are identical and form an ionically ordered structure. The latter property, at least, may lead us to expect some similarity between the effect of impurity ions on the ferromagnetic resonance of ordered lithium ferrite and YIG. In the present paper we report experimental results on the resonance linewidth ( $\Delta H$ ),  $g$  factor and magnetocrystalline anisotropy of ordered lithium ferrite containing impurity ions. Our analysis of these results shows that the expected similarity actually exists.

Measurements were made on ordered monocrystals at a frequency of 24,200 Mcps and at temperatures ranging from 4.2°K to 300°K. The temperature dependence of the  $g$ -factor was found to be proportional to  $1/T$  at "high" temperatures (50°K  $\leq T \leq$  300°K) but less pronounced at "low" temperatures ( $T < 50^\circ\text{K}$ ). The  $\Delta H$  vs  $T$  curves exhibit a maximum ( $\Delta H = 5, 24$  and  $14$  oe at 4.2°K, 50°K and 300°K, respectively) in the transition region ( $T \approx 50^\circ\text{K}$ ) for all crystallographic directions which are not in the vicinity of [110]. Both of these effects can be interpreted on the basis of the theory of rapidly relaxing impurity ions proposed by Kittel and co-workers<sup>1</sup> in connection with rare earth iron garnets and doped YIG. This interpretation is plausible because a spectroscopic analysis indicates the presence of about 0.1% Mn, 0.1% Ni and 0.01% Co in our samples, and because certain ions<sup>2</sup> of these metals may be rapidly relaxing in ferrites. Ferrous ions are presumably not responsible for these effects because an extensive oxidation treatment did not alter the two effects mentioned above. The role of ferrous ions in the anomalous  $\Delta H$  peak observed in the [110] direction of our ordered samples was investigated earlier.<sup>3</sup>

We find that the impurity ions in ordered lithium ferrite affect not only  $g$  and  $\Delta H$ , but also produce anomalous anisotropy peaks of the kind first observed by Dillon<sup>4</sup> in garnet structures. The previously mentioned extensive oxidation did not influence these peaks. To calculate the resonance field vs. angle curve, we use Kittel's<sup>5</sup> theory and supplement it with the specific assumptions that the effective impurity ions are on tetrahedral sites, where the uniaxial crystalline electric field is trigonal, and that their ground state has a non-accidental degeneracy. We assume that this particular degeneracy is removed when the exchange field is rotated away from a direction perpendicular to the trigonal field. Our calculations show that peaks should exist in the [110] and [112] directions of the (110) plane, and that the amplitude of the former is twice that of the latter. Analysis of our experimental data confirms both of these predictions as well as the other details of the resonance field vs angle curve.

<sup>1</sup>C. Kittel, Phys. Rev. **115**, 1587 (1959); P.-G. de Gennes, C. Kittel, and A. M. Portis, Phys. Rev. **116**, 323 (1959).

<sup>2</sup>R. L. White, Phys. Rev. Letters **2**, 465 (1959).

<sup>3</sup>A. D. Schnitzler, V. J. Folen, and G. T. Rado, J. Appl. Phys. **31**, 348S (1960).

<sup>4</sup>J. F. Dillon, Jr., Phys. Rev. **105**, 759 (1957); **111**, 1476 (1958).

<sup>5</sup>C. Kittel, Phys. Rev. Letters **3**, 169 (1959); Phys. Rev. **117**, 681 (1960).

## 120. FERROMAGNETIC RESONANCE MAGNON DISTRIBUTION IN YTTRIUM IRON GARNET

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The ferromagnetic resonance frequency is dependent on the number of magnons present. There is, therefore, a small shift in frequency as a function of applied power. This shift depends on both shape and magnetocrystalline anisotropy. The relation previously obtained<sup>1</sup> for the shift in resonance frequency for an ellipsoidal sample has been extended to include the effect of the dipole-dipole interaction. The shift in the resonance frequency can be expressed in the form

$$\Delta f = a n_0 + \sum_{\mathbf{k} \neq 0} b_{\mathbf{k}} n_{\mathbf{k}},$$

where  $n_0$  is the number of  $\mathbf{k} = 0$  magnons and where  $n_{\mathbf{k}}$  is the number of magnons with wave vector  $\mathbf{k}$ . Values of  $a$  and the  $b_{\mathbf{k}}$ 's for a sphere aligned with the dc magnetic field along the [100] (hard) direction and the [111] (easy) direction have been evaluated. Techniques for measuring these small shifts to an accuracy of 10 kc at X band have been developed. Measurements have been made on 5 single crystal yttrium iron garnet spheres with the dc magnetic field in both the hard and easy directions. The rf magnetic fields were in the range of 1 to 10 millioersted, which is below the onset of non-linear effects. It was observed that  $\Delta f$  is directly proportional to the square of the rf field, positive in the hard direction, and negative in the easy direction. The shifts were typically about 100 kc for rf fields of 10 millioersted. By relating  $n_0$  to the half-width and the rf magnetic field, the measurements in conjunction with the above equation can be used to determine  $n'/n_0 \sum_{\mathbf{k} \neq 0} \alpha_{\mathbf{k}} b_{\mathbf{k}}$ , where  $n'$  is the total number of  $\mathbf{k} \neq 0$  magnons and  $\alpha_{\mathbf{k}}$  is the fraction of all the  $\mathbf{k} \neq 0$  spin waves with wave vector  $\mathbf{k}$ . For each sample this quantity has been determined to be negative for the easy direction. If one assumes that the  $\mathbf{k} \neq 0$  magnons are degenerate with the  $\mathbf{k} = 0$  magnons, the  $b_{\mathbf{k}}$ 's are negative only in the region  $k < k_{\max}/2$  where  $k_{\max}$  is the largest  $k$  that a magnon degenerate with the  $\mathbf{k} = 0$  magnon can have. Therefore, the dominant contribution to the above sum must come from magnons with  $k < k_{\max}/2$ . This conclusion is not materially affected by assuming other reasonable energy distributions of the magnons. The results will be compared with the surface pit scattering theory of Sparks, Loudon, and Kittel.<sup>2</sup>

<sup>1</sup>Matcovich, Belson, and Goldberg, J. Appl. Phys. **32**, 163S, (1961).

<sup>2</sup>Sparks, Loudon and Kittel, Phys. Rev. **122**, 791, (1961).



121. ON THE POSSIBILITY OF OBTAINING LARGE AMPLITUDE RESONANCE IN VERY THIN FERRIMAGNETIC DISKS\*

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In this paper, a theoretical prediction is made that the uniform resonance and the spinwaves in a very thin ferrimagnetic disk can be made nondegenerate. If this condition can be achieved experimentally, the usual second order spinwave instabilities<sup>1</sup> will be forbidden and cannot limit the component of the magnetization occurring at the resonance frequency ( $\omega_0$ ). The precession cone angle ( $\theta$ ) should then increase, in response to the usual rf driving field, until limited by third order instabilities involving spinwaves with frequencies equal to  $3/2\omega_0$ . The new instability threshold is independent of frequency except for very small  $\omega_0$  and even then is approximately given by

$$\theta_{\text{crit}} = 2 \sqrt[3]{\Delta H_k / M}$$

where  $\Delta H_k$  is the half linewidth of the unstable spinwave and  $M$  is the saturation magnetization.

The state of nondegeneracy is to be achieved in the following manner: a thin disk, magnetized in a direction perpendicular to the plane, is excited by two transverse positive-circularly-polarized pulsed rf magnetic fields of specified amplitude. One of these fields has a frequency equal to  $\omega_0$  the other a frequency detuned from  $\omega_0$  by an amount comparable to the width of the resonance. As  $\theta$  increases from zero during the ensuing transient, there will occur a point at which the familiar second order coupled spinwaves become unstable (the state of nondegeneracy has not yet been reached). Since there is a time lag before these modes can effect the uniform precession, the cone angle will continue to increase. When some larger value of  $\theta$  has been attained the degeneracy between the spinwaves and the resonance is removed; the previously unstable spinwaves are then decoupled from the uniform resonance.

The degeneracy is removed because, as the author has already shown theoretically,<sup>2</sup> the resonance frequency of the thin disk should lie below the main spinwave band provided  $\theta > \sqrt{2N_t}$ , where  $N_t$  (the transverse demagnetizing factor) is by assumption very small. This level of excitation would create the desired state of nondegeneracy, if it were not for the fact that a range of degenerate spinwaves (separate from the main band) still exists. These degenerate modes may be destroyed, however by introducing the additional transverse rf field. The existence of this auxiliary field raises the threshold of nondegeneracy somewhat but this effect is not serious. It is obvious, however, that if a stable level of the resonance is to be maintained this new threshold must be lower than that governing the previously mentioned third order spinwave instabilities. This last consideration places a maximum permissible value on  $N_t$ . This value is dependent on the amplitude and frequency of the auxiliary field. For the conditions specified in this paper, it is given by

$$N_t \leq \frac{9}{10} \left( \frac{\Delta H_k}{M} \right)^{\frac{2}{3}} - 3 \left( \frac{\Delta H_0}{M} \right)$$

where  $\Delta H_0$  is the half linewidth of the uniform precession.

Details of the theory will be presented and applied to the case of single crystal YIG as well as certain of the mixed ferrimagnetic garnets.

Since the resonant frequency of the disk is a function of  $\theta$ , a discussion of the buildup transient of the cone angle is also included.

<sup>1</sup>H. Suhl, Proc. Inst. Radio Engrs., 45, 1271 (1956).

<sup>2</sup>F. R. Morgenthaler, Doctoral Thesis Department of Electrical Engineering, Massachusetts Institute of Technology Cambridge, Mass. (May 1960).

122. MICROWAVE RESONANCE LINEWIDTH IN SINGLE CRYSTALS OF COBALT SUBSTITUTED MANGANESE FERRITE

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Measurements will be reported of the microwave resonance linewidth,  $\Delta H$ , for small, polished, spherical samples of single crystals with the composition  $\text{Co}_x\text{Mn}_{1-x}\text{Fe}_2\text{O}_4$ , where  $0 \leq x \leq 0.1$ . Accurate chemical analyses are available. The Curie temperature of the material increases with  $x$  and lies between 550°K and 575°K.

The substitution of cobalt for manganese increases  $\Delta H$  rapidly. At room temperature and 17,000 Mcps  $\Delta H [100] = 37 + 4,000 x$  oersteds and  $\Delta H [111] = 31 + 3,000 x$  oersteds, where  $\Delta H [100]$  is the value of  $\Delta H$  measured with the static magnetic field,  $H$ , applied along a crystallographic [100] direction and similarly for  $\Delta H [111]$ . The corresponding results at 9600 Mcps are  $\Delta H [100] = 19 + 2680 x$  oersteds and  $\Delta H [111] = 9 + 2150 x$  oersteds. Results for other crystallographic directions lie between those for [100] and [111] hence it can be seen that at this temperature the linewidth due to the cobalt substitution is anisotropic by about 20 per cent.

Measurements at 17,000 Mcps show that in the temperature range 250°K to 500°K  $\Delta H$  is fairly independent of temperature, though the anisotropy in  $\Delta H$  increases steadily as the temperature is reduced. Between 250°K and 80°K  $\Delta H$  broadens increasingly rapidly with reducing temperature and at the lower end of this range sharp maxima in  $\Delta H$  as a function of crystallographic direction appear at 20° from a [100] direction in a (110) plane. These maxima are probably associated with the maxima in the field for resonance at these points observed at liquid helium temperatures and reported previously.<sup>1</sup>

In view of the small temperature variation of  $\Delta H$  over a wide range of high temperatures it seems that the divalent cobalt ion does not act here, as has been suggested,<sup>2</sup> like the rapidly relaxing rare earth ions in the iron garnets. This conclusion is supported by the variation of the  $g$ -factor for these crystals with  $x$  which was reported previously,<sup>1</sup> this variation is much too slow to fit the expression appropriate if the cobalt ion relaxed rapidly.

The measurements show that the effect of cobalt upon  $\Delta H$  is considerably larger than that calculated by Haas and Callen<sup>3</sup> who assumed that the cobalt ions scatter energy from the uniform mode to spin waves of short wavelength.



The results of experiments on disc samples, where the effect of such scattering should be much smaller than in the spheres will also be reported.

<sup>1</sup>R. W. Teals and M. J. Hight, *J. Appl. Phys.* **32**, 140S, (1961).

<sup>2</sup>R. L. White, *Phys. Rev. Lett.*, **2**, 465 (1959).

<sup>3</sup>C. Warren Haas and Herbert B. Callen, *Phys. Rev.* **122**, 59, (1961).

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### 123. DC EFFECTS IN FERROMAGNETIC RESONANCE IN THIN FERRITE FILMS\*

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The dc "Hall-effect" voltage which accompanies ferromagnetic resonance has been observed in thin films of ferrites. This voltage, which is associated with both the Hall and magneto-resistance effects, is in qualitative agreement with the theory<sup>1,2</sup>, but larger by an order of magnitude. The curves of voltage vs static magnetic field are primarily of absorption form, and reverse with reversal of the static field. The magnitude of the voltage is proportional to the microwave power; the voltage observed on a 1000 Å film of magnetite was one millivolt at a microwave field level of 100 oe.<sup>2</sup> Because the effects depend on the microwave current density in the film, it is necessary to use films of high conductivity; our observations were made on films of magnetite and nickel ferrous ferrite. In these materials the ferromagnetic resonance linewidth is extremely large and the permeability small, even at resonance, so the dc voltage per unit power is far smaller than in metal films. In order to obtain sufficiently high microwave magnetic fields with the available power, the samples were placed in a cavity, near a point of maximum magnetic field. It was thus possible to observe ferromagnetic resonance simultaneously by the usual microwave methods and by the dc signal. Observation of ferromagnetic resonance by the dc voltage would appear to be possible in films too thin to examine by the conventional microwave technique.

\*This work was supported by; United States Army Signal Research and Development Laboratory, Office of Naval Research, Air Force Office of Scientific Research.

<sup>1</sup>M. H. Seavey, *J. Appl. Phys.* **31**, 216S (1960).

<sup>2</sup>H. J. Juretschke, *J. Appl. Phys.* **31**, 1401 (1960).

## SESSION VIIB PERMANENT MAGNETS AND MICROMAGNETICS

F. E. LUBORSKY, *Presiding*

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### 124. EXCHANGE ANISOTROPY - A REVIEW (Invited)

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The term exchange anisotropy was coined to describe the exchange interaction across the interface between two magnetic materials. This interaction was first discovered in fine particles of cobalt having an antiferromagnetic cobaltous oxide shell. A compact of the fine particles exhibited a shifted hysteresis loop, a  $\sin \theta$  torque curve and a non-vanishing rotational hysteresis at magnetic fields greater than  $2K/M_S$ . This system of cobalt-cobaltous oxide had a unidirectional anisotropy (not uniaxial), although this is not a unique property of exchange anisotropy. Various workers have discovered this interaction in other powders, in thin films and in bulk metallic materials. Some of these workers have used this exchange anisotropy to explain improved properties of fine particle permanent magnets, a memory effect in a mixed ferrimagnetic spinel, rotatable anisotropy in thin films, and the reverse magnetization of certain deposits in the earth's crust. Examples of this exchange interaction between antiferromagnetic-ferromagnetic, ferrimagnetic-ferromagnetic, and antiferromagnetic-ferrimagnetic systems are reviewed. The interfacial conditions, such as the relative spin axis directions, necessary to obtain the interaction between the two magnetic systems are discussed, as well as the problem of determining whether the asymmetrical hysteresis loop is a minor loop due to applying insufficient magnetic field, or a shifted hysteresis loop created by an exchange interaction. A model is presented which yields non-vanishing rotational hysteresis for magnetic fields greater than  $2K/M_S$  as has been found in a number of systems where the exchange interaction is believed to be present.

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### 125. PERMANENT MAGNETIC PROPERTIES OF IRON-COBALT PHOSPHIDES

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According to Berak and Neumann<sup>1</sup> the ternary diagram Fe-Co-P contains a quasi-binary section with  $Fe_2P$  and  $Co_2P$  as final components. Preliminary structural investigations have led to the conclusion that the phase relations in this section will probably have to be modified. The compounds  $Fe_2P$  and  $Co_2P$  have been reported by several investigators to be weakly magnetic at room temperature.<sup>2,3</sup>



As against this it was found that alloys of the composition  $(\text{Fe}, \text{Co})_2\text{P}$  have a moderate magnetic moment and an appreciable magnetic anisotropy. A method is described whereby high intrinsic coercive forces can be obtained in fine particles. Anisotropy measurements, demagnetization curves and  $\sigma$ -T curves are shown as a function of the Fe-Co content for alloys prepared by different methods. The possibility of utilizing these materials as permanent magnets is discussed.

<sup>1</sup>Berak and Neumann, *Archiv für das Eisenhüttenwesen*, **21**, 327 (1950).

<sup>2</sup>H. Le Chatelier and S. Wologdine, *Comptes Rendus*, **149**, 709 (1909).

<sup>3</sup>S. Zemczuzny and J. Schepelew, *Zeitschr. f. anorg. Chem.* **64**, 245.

## 126. STUDIES OF HIGH COERCIVITY COBALT-PHOSPHOROUS ELECTRODEPOSITS

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Previous studies of high coercivity electrodeposits have been done in the Co-Ni-P and Co-P systems. In these cases coercivities of 400 oe to 800 oe were found and the deposits were shown to have a single-domain platelet-like structure. X-ray studies showed the crystallographic uniaxial easy direction of magnetization to be in the plane of the film.

Further studies in the Co-P system have been undertaken and coercivities in the range of 1000 oe to 1300 oe have been obtained in the plane of the film. In this case, the crystallographic easy direction of magnetization is normal to the plane of the film. Electron micrographs of the surface of these samples show a fairly large grained (two microns) deposit. No subdivision of these grains is found when conventional replication techniques are employed.

Bitter patterns of the surface of these deposits were obtained utilizing the Craik technique. The demagnetized surface produces a large elliptical pattern. The magnetized (remanence state) surface also produces a pattern, indicating that the easy direction of magnetization is out of the plane of the film. The patterns obtained on this surface indicate the existence of a rod-like structure rather than a platelet structure, the major axis of the rod being normal, or nearly normal to the plane of the film.

Most samples in the 400 oe to 800 oe region consist of platelets. In this case, shape anisotropy cannot contribute to the coercivity and only crystalline anisotropy need be considered. The Stoner-Wohlfarth model predicts  $B_r/B_m = 0.5$  for a random compact of such platelets. Dc hysteresis loops of the samples show a good agreement with this prediction. The higher coercivity materials have a rod-like structure, and shape anisotropy may contribute to the higher coercivities observed. Samples having coercivities of 1200 oe to 1300 oe in the plane of the film have  $B_r/B_m$  of 0.3 to 0.4. Experimental samples having  $H_c$  of 500 oe and consisting of rods, have  $B_r/B_m$  of 0.2. Theoretical shape anisotropy models predict similar  $B_r/B_m$  ratios for

materials having an easy direction of magnetization at 70° or 80° from the applied field.

All of the results support the rod-like model of this material. The possibility of improved properties is discussed in the light of this model.

## 127. THE FORMATION OF MONOCRYSTALLINE ALNICO MAGNETS BY SECONDARY RECRYSTALLIZATION METHODS

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A brief review of the progress of the "Alnico Art" is given tracing early attempts at increasing energy yield by changes in composition, to present day techniques of promoting polycrystalline growth parallel to the magnetic axis. Energy product values have steadily risen to the present commercially available level of 7.5 mega-gauss-oersteds. Since research has apparently reached a temporary impasse in further improvement in polycrystalline Alnico magnets, except by an uneconomic fabrication from a much larger initial casting, or expensive foundry mold techniques, an attempt to take advantage of the 12.0 mega-gauss-oersted energy product available from single crystals of Alnico is an obvious solution. Two known methods of producing single crystals sufficiently large to be of practical value are described and the obvious economic and production problems discussed.

New work on a third method is described in which normal foundry techniques are used, but the formation of a single monocrystal of the entire magnet is promoted by secondary recrystallization. It is shown that if a sufficiently large grain edge strain can be induced in a polycrystalline aggregate, resulting in secondary recrystallization, formation of a single large crystal is possible. It is also shown that if a sufficiently large internal stress can be introduced by both mechanical and thermal means, the required grain-edge shift can be accomplished. The additions of normally prohibitive amounts of either carbon, nitrogen, manganese, or other "gamma-phase" promoting elements will serve to provide the necessary mechanical strain, and methods of controlling their effect on the magnetic results are shown. The thermal stresses are accomplished by a simple maintenance of 60-80°C gradient across the magnet poles. This process has yielded single crystals approaching 1/4 lb. in size whose energy product is 11.0 mega-gauss-oersteds.

The practical nature of this process is discussed, and possible mass production techniques outlined.



## 128. SATURATION MAGNETIZATION OF SWAGED Mn-Al\*

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Two years ago remarkable properties were reported for a new alloy based on manganese and aluminum.<sup>1</sup> The value for saturation magnetization in Mn-Al had been extrapolated to infinite magnetizing field and was given as 6200 gauss. Our measurements indicate that the extrapolated figure is too low. Magnetization values of over 6700 gauss have been measured at 10,000 oe and extrapolations of these values to infinite fields give over 7300 gauss for  $4\pi M_S$  at room temperature. The material having the high saturation magnetization had an energy product of  $4.0 \times 10^6$  gauss-oe.

To reach the high values, it was necessary to adhere to processes and procedures having narrow limits. Saturation magnetization depended directly on composition, homogenizing, quenching rate, annealing and deformation. The latter was most critical because a preferred axis of orientation is developed by this process. Mn-Al is very brittle, but the brittleness problem was circumvented by swaging the material in suitable jackets.

Micrographs of domain patterns show the proportion of magnetic material present after the various treatments. Rockwell tests indicate that hardness is directly related to  $4\pi M_S$ .

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\*Work was sponsored by the Aeronautical Systems Division of the Air Research and Development Command of the U. S. Air Force.

<sup>1</sup>A. J. J. Koch, et al., J. Appl. Phys. 31S, 75S (1961).

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## 129. STUDY OF PARTICLE ARRANGEMENTS AND MAGNETIC DOMAINS ON THE SURFACE OF PERMANENT MAGNETS\*

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Some permanent magnet materials are compacts of very small particles. Their properties are based on the magnetic peculiarities of these particles which have been studied for many years. The properties of compacts depend also greatly on the mutual magnetic influences between particles or groups of them. Packing arrangement, packing density, orientation, separating layers, dislocations, slip lines, etc. influence the magnetic properties of such materials. Little is known about these factors in many cases. It seems necessary to determine them more clearly in order to study their correlation and effects.

Features of the surfaces of fine particle compacts can be made visible to give clues on the above mentioned factors and domain arrangements. The sizes of the details involved require the resolving power of a good electron microscope. Special replicating techniques have been developed to combine shadow-graphing and colloid application on replicas of rough and porous surfaces.

A relatively simple method is described which makes the surface details visible and reveals magnetic phenomena simultaneously.

Examples are presented of such surface investigations on iron-cobalt particle magnets with particular particle groupings becoming visible. Surfaces of fine crystalline barium ferrite magnets show evidence of magnetic domains many times smaller than found or assumed existing before.

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\*Work was sponsored by the Aeronautical Systems Division of The Air Research and Development Command of the U. S. Air Force.

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## 130. ON THE POSSIBILITY OF DOMAIN WALL NUCLEATION BY THERMAL AGITATION

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Very fine particles are known to reverse their magnetization spontaneously, passing the energy barrier by means of thermal activation. In the same way one could argue that small portions of larger particles could be spontaneously reversed, thereby serving as nuclei for reversed domains. These can then grow, because it is known that the energy favors subdivision into domains. In treating such a possibility, however, one should take into account the additional barrier for forming a wall around the "nucleus," which is usually left out in standard domain theory calculations when the wall energy is used. This energy is rather small but the barrier for formation of the wall is large.

The model studied is a small sphere, centrally located inside a spherical particle, the magnetization in which starts to rotate gradually from the z direction in which the rest of the particle is magnetized. The angle between magnetization and the z axis is assumed to vary linearly in the radial coordinate, from zero on the surface of the inner sphere, to its maximum value in the center. This value eventually reaches  $\pi$ , thus completing a wall around a reversed domain of radius zero, which should then start to grow by further supply of energy.

Studying the energy barrier for such a process, it is seen that the main contribution is from the exchange energy, which practically forbids the process. The probability for a domain wall to nucleate at room temperature is thus shown to be negligibly small, thus justifying the neglect of temperature effects in Brown's equations.

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## 131. FAILURE OF THE LOCAL-FIELD CONCEPT FOR HYSTERESIS CALCULATIONS

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The concept of the "local" or "effective" field is legitimate and useful in linear problems, such as the



molecular theory of nonpolar dielectrics.<sup>1</sup> The local field is the field produced at the position of one dipole by the external sources and all the other dipoles; it is useful to the extent that it can be approximated by a simple formula such as that of Lorentz.

The local-field concept has also been used in theories of magnetic hysteresis based on the Preisach model.<sup>2</sup> In this model the elementary dipoles have rectangular hysteresis loops centered about a nonzero field intensity; the local-field concept enters if this last quantity is interpreted as the field of other dipoles. Such use of the local-field concept is not legitimate and can lead to serious errors. The vertical jump in a rectangular hysteresis curve occurs at a field at which the system becomes unstable. When two or more particles interact, the effect of the interaction is not simply to shift the instability fields of each particle by an amount equal to the field of the other particles. The interaction may result in an instability with respect to simultaneous rotations of the different dipole moments through related but different angles; the local-field calculation considers only rotations of a single dipole moment with the others held fixed. Therefore coercivities calculated by the local-field method may be too high.

This principle is illustrated by the "chain of spheres" model of Jacobs and Bean.<sup>3</sup> As a further illustration, the instability field  $H_C$  has been calculated for a cylindrical particle, of various length-to-diameter ratios  $p^*$ , consisting of  $n$  ( $= 2$  or  $3$ ) identical single-domain cylinders end-to-end, with magnetic coupling between the parts; the applied field is longitudinal. Both the rigorous calculation (1) and the local-field method (2) give values of  $h_c = H_C/M_S$  ( $M_S =$  spontaneous magnetization) between that (3) for a single-domain cylindrical particle of ratio  $p^*$  and that (4) for independent particles of ratio  $p = p^*/n$ ; but the local-field method gives too high a value. For  $p^* = 6$  the four values are, in the order (1) to (4): ( $n = 2$ ) 4.25, 4.63, 5.02, 3.87; ( $n = 3$ ) 3.59, 3.97, 5.02, 2.86. For  $n = 3$ , instability occurs first with respect to a simultaneous rotation of the end moments through one angle and of the middle moment through a different angle of opposite sign; the local-field value of  $h_c$  is a good approximation to a different mode of reversal, in which the end moments rotate oppositely while the middle moment remains unchanged, and for which, at  $p^* = 6$ ,  $h_c = 3.94$ .

<sup>1</sup>W. F. Brown, Jr., *Encyclopedia of Physics*, Vol. 17 (Springer-Verlag, Berlin, 1956), p. 47 ff.

<sup>2</sup>For references see D. F. Eldridge, *J. Appl. Phys.* **32**, 247S-249S (1961).

<sup>3</sup>I. S. Jacobs and C. P. Bean, *Phys. Rev.* **100**, 1060-1067 (1955).

### 132. SINGLE-DOMAIN MAGNETIZATION CURVES FOR THE GENERAL ELLIPSOID

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Single-domain magnetization curves have been extended to the general ellipsoid. The calculation was carried out on an IBM 705 Computer and was an iterative minimization

of the energy as a function of the direction of magnetization for an arbitrary applied field. The results are similar to those of Stoner and Wohlfarth for ellipsoids of revolution. Tables containing reduced magnetization as a function of the magnitude and direction of the applied field and shape will be made available.

### 133. THE DEPENDENCE OF AVERAGE INTERACTION FIELDS ON INTENSITY OF MAGNETIZATION IN ASSEMBLIES OF FERROMAGNETIC PARTICLES

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The hysteresis properties of assemblies of fine particles depend on the properties of the individual particles and on the magnetic interaction between them. As the interaction fields vary with position in the sample and change in size and direction as the state of magnetization changes, we must discuss average interaction fields and examine how they depend on the intensity of magnetization of the sample as a whole. The method which has been developed is as follows: first, the graph of decreasing remanence as a function of increasing demagnetizing field is obtained; then, the specimen is brought to a chosen point on this curve and the increasing remanence values acquired as a result of temporarily-applied magnetizing fields are measured. The two curves, one showing how the remanence decreased and the other how it regained its former value, are compared at their (remanence) mid-points. The difference between the magnetizing and the demagnetizing fields required to attain these points is taken as a measure of the average interaction field in the sample at that intensity. This procedure is repeated at different intensities, and a graph is obtained of the average interaction field as a function of intensity from maximum-remanence down to zero-remanence. Oriented and unoriented specimens of  $\gamma$   $Fe_2O_3$  with packing densities of 20% and 40% (by volume) have been examined by this method and gave similar results. An average interaction field which is negative and of over 200 oe was found near maximum-remanence. The field rapidly decreased and ultimately became zero at zero-remanence. The field required to attain the remanence mid-point in the forward (magnetizing) direction was found to be constant over a wide range of magnetization intensities for the un-oriented sample and increased only slowly with decreasing remanence for the other samples. The results can be explained by means of the Preisach diagram which has been calculated for each specimen from the experimental curves.



### 134. THE EFFECT OF ANGULAR VARIATIONS IN FIELD ON FINE-PARTICLE REMANENCE

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The Stoner-Wohlfarth model is used to calculate the remanence of a randomly oriented assembly of single-domain particles when the applied field changes in direction. The cases considered are: (1) fields applied successively in directions differing by multiples of  $\pi/2$ , and (2) fields rotated through multiples of  $\pi/2$ . Some consideration is also given to the anhysteretic remanence when the unidirectional and alternating fields are orthogonal. Experimental results obtained on discs of  $\gamma$  ferric oxide recording tape are found to be in reasonably good qualitative agreement with the calculations.

\*With Ampex Corporation, Redwood City, California when this work was initiated.

### 135. REMANENT TORQUE STUDIES IN POLYCRYSTALLINE $\text{BaFe}_{12}\text{O}_{19}$ \*

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Measurements of the anisotropy distribution in polycrystalline  $\text{BaFe}_{12}\text{O}_{19}$  were carried out using the remanent torque technique recently tested on  $\gamma\text{-Fe}_2\text{O}_3$  powders.<sup>1,2</sup> Because  $\text{BaFe}_{12}\text{O}_{19}$  has a high uniaxial crystal anisotropy one has  $K = 3.3 \times 10^6 \text{ erg/cm}^3 \gg 1.6 \times 10^5 = I_s^2$  and therefore one is justified in applying a picture in which interparticle interactions have been neglected. Because magnetostatic effects can be neglected one expects also that in this case the remanent torque curves will be an intrinsic property of the material; i.e., samples with different hysteretic properties should display the same remanent torque curves.

To test this, measurements were carried out on two  $\text{BaFe}_{12}\text{O}_{19}$  randomly oriented powder samples whose hysteresis loops differed considerably. As expected, however, the torque curves are very similar.

The results of the analysis of the torque curves together with the coercive force of the samples studied are given in Table I. Instead of reproducing the anisotropy distribution curve derived from the torque curves, only the average anisotropy  $K$  and the half width at half value  $K$  is given. It is interesting to compare the experimental results with theoretical prediction for an assembly of non-interacting single domain particles, Stoner-Wohlfarth model<sup>3</sup> with which Table I has been supplemented. The experimental coercive force is much lower than the theoretical one indicating the importance of noncoherent magnetization processes in the determination of the coercive force, these being more important in sample I than in II. The agreement is much better when the anisotropy is considered indicating that the remanent torque curves can be

described by the Stoner-Wohlfarth<sup>3</sup> model to a reasonable approximation. This method should, therefore, be a useful one for the determination of the uniaxial anisotropy of materials available only in polycrystalline form such as manganese-aluminum.

TABLE I

	$H_c$ (Oersteds)	Peak $K$ ( $\text{erg/cm}^3$ )	$K$ Half Width ( $\text{erg/cm}^3$ )
Sample I	1500	$2.1 \pm 0.2 \times 10^6$	$1.0 \pm 0.2 \times 10^6$
Sample II	4000	$2.1 \pm 0.2 \times 10^6$	$1.0 \pm 0.2 \times 10^6$
Stoner-Wohlfarth	8500	$3.3 \times 10^6$	

\*The authors gratefully acknowledge the support by the Air Force Office of Scientific Research of the Air Research and Development Command.

\*\*On leave of absence from the Weizmann Institute of Science, Rehovoth, Israel.

<sup>1</sup>P. J. Flanders and S. Shtrikman, submitted to J. Appl. Phys.

<sup>2</sup>P. J. Flanders and S. Shtrikman, presented before the International Conference on Magnetism, Kyoto, Japan, September 1961.

<sup>3</sup>E. C. Stoner and E. P. Wohlfarth, Trans. Roy. Soc. A240, 599 (1948).



# SESSION VIIIA

## ALLOYS AND COMPOUNDS

J. A. OSBORN, Presiding

### 136. AGE-HARDENED GOLD PERMALLOY AND GOLD PERMINVAR

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This work is an extension of previous work which showed that the addition of gold to permalloy made it possible to cause the precipitation of a gold-rich phase by heat treatment, and thereby to control the coercive force by controlling the formation of the precipitate. This addition of a controlled second phase to permalloy made possible a great improvement in the switching speed of the alloy. Some compositions of gold permalloy had switching speeds four times faster than that of the standard composition. In the present work, some of the details of the precipitation process were studied. It was found that for specimens containing seven per cent gold and annealed at 650°C, the precipitation of the gold-rich phase depended strongly upon cold working. When this alloy was quenched from the solution temperature and annealed at 650°C for 2 hours there was little increase in coercivity and the electron micrographs showed practically no precipitation of particles of 1,000 Å diameter or larger. However, when the material was quenched from the solution temperature, drastically cold rolled and annealed at 650°C, the coercivity was increased substantially and the electron micrographs revealed the presence of many particles having a diameter of 1,000 Å.

Seven per cent gold was also added to the perminvar composition of 43% Ni-34% Fe-23% Co. The alloy behaved somewhat similarly to the gold-permalloy compositions. It indicated the effects of age-hardening in the vicinity of 600°C; square hysteresis loops and fast switching speeds were obtained. However, the threshold for these high switching speeds was much higher than for permalloy and therefore the alloy appeared less desirable for switching applications.

### 137. PARAMAGNETIC BEHAVIOR OF IRON-RICH IRON-VANADIUM ALLOYS

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Recently we have started extensive paramagnetic investigations of iron-rich binary iron alloys. In this paper the experimental results on fourteen iron-vanadium alloys up to 26 atom per cent vanadium will be presented. Iron-vanadium alloys exhibit unusual paramagnetic behavior.

Magnetic susceptibility vs vanadium concentration curves have a maximum at about 10 atom per cent vanadium. The size of this maximum decreases with increasing temperatures. At 1160°K the paramagnetic susceptibility of bcc iron-vanadium alloy containing 10 atom per cent vanadium is approximately twice that of pure bcc iron at the same temperature. The curve of paramagnetic Curie temperatures vs vanadium concentration has a maximum at about 10 atom per cent vanadium. The magnetic moment per iron atom decreases gradually with amount of vanadium in iron. The paramagnetic susceptibility behavior of iron-vanadium alloys with low vanadium content obeys the Curie-Weiss law. Higher vanadium concentrations cause deviations from this law which appear as an additional temperature independent paramagnetism superimposed on the Curie-Weiss behavior. Possible explanations for the above-described phenomena will be given.

### 138. MAGNETIC CHARACTERISTICS OF HYDROGENATED HOLMIUM

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The published work of Stalinski<sup>1</sup> for the La-H and Ce-H systems and unpublished work<sup>2</sup> in this Laboratory on the Dy-, Ho- and Yb-H systems have shown in each case that metallic conduction disappears when the metal is fully hydrogenated. This behavior together with the observed stoichiometries indicates that these hydrides are saline in nature, the hydrogen present existing as an anion. Various evidence indicates, further, that: (1) the hydride anion is formed by consumption of electrons in the conduction band, and (2) in the fully hydrogenated material the conduction band is completely depopulated. Since exchange effects in the lanthanide metals occur in the main via the conduction electrons, progressive alterations in the magnetic behavior are expected as hydrogen is absorbed by the metal. For example, cooperative magnetic phenomena are expected to be greatly suppressed in the hydrogenated metal as compared to the element. To ascertain whether this is indeed the case a series of investigations involving the lanthanide hydrides is being carried out. The present study dealing with the holmium-hydrogen system is the first of these.

Magnetization-temperature data have been obtained over the temperature range 4 to 300°K for alloys designated as HoH<sub>X</sub>, in which X = 1.51, 1.60, 1.82, 2.00, 2.31, 2.48, 2.62, 2.93 and 3.06. From recent crystal structure studies in this Laboratory it is known<sup>3</sup> that the samples with X = 1.82 to 2.31 are single phase and cubic, and their structures are in fact based on the ideal stoichiometry HoH<sub>2</sub>. The samples with X ≥ 2.62 are single phase and hexagonal, and have structures based on HoH<sub>3</sub>. The other three samples are mixtures of the element and the dihydride phase and of the di- and tri-hydride phases.

Above 15°K the single phase materials all obey the Curie-Weiss Law. Magnetic moments of the three dihydrides and three trihydrides are  $9.9 \pm 0.1$  and  $10.0 \pm 0.1 \mu_B$  per holmium ion, respectively. (The moment for the element was found to be  $10.9 \pm 0.2 \mu_B$ .) The several dihydrides exhibit a Néel point at about 9°K, whereas the trihydrides



give no evidence of a Néel point. The latter however exhibit deviations from the Curie-Weiss Law below about 10°K. The observed magnetic behavior is consistent with present notions regarding the electronic nature of the lanthanide hydrides and the mechanism for exchange in the lanthanides.

<sup>1</sup> B. Stalinski, Bull. de l'accad. Pol. des Sci., 5, 1001 (1957); 7, 269 (1959).

<sup>2</sup> T. Peltz and W. E. Wallace.

<sup>3</sup> A. Pebler and W. E. Wallace, submitted to J. Phys. Chem.

### 139. NEW MODIFIED $Mn_2Sb$ COMPOSITIONS SHOWING EXCHANGE INVERSION

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Transformations from ferrimagnetic to antiferromagnetic ordering with decreasing temperature of the type previously reported for the compounds  $Mn_{2-x}Cr_xSb^{1,2}$  have now been observed in manganese antimonides modified with the transition elements V, Co, Cu, and Zn and with Ge and As. In these compounds, as with chromium modified  $Mn_2Sb$ , the exchange inversion temperature shows a dependence upon the concentration of the modifying element but each system has a unique range of operability. These materials all have the same tetragonal crystal structure as  $Mn_2Sb$  ( $P4/nmm$ ). The function of the modifying element in each case appears to be similar to that of chromium, that is, it shrinks the structure in such a way that the interlayer distance along the tetragonal axis is decreased so that exchange inversion can take place. The magnitude of this interlayer distance is a function of the modifying element. For  $Mn_2Sb_{0.95}Ge_{0.05}$  the magnetic structure change is accompanied by a sharp first order change in the tetragonal axis in the temperature range -88° to -75°C. with the ferrimagnetic state appearing at a c axis length of 6.530 Å. The binary compounds  $Mn_2As$ ,  $Cu_2Sb$ , and  $Mn_2Sb$  all have the same tetragonal crystal structure and both  $Mn_2As$  and  $Cu_2Sb$  are antiferromagnetic<sup>3,4,5</sup>. Transitions observed in manganese antimonides containing arsenic are much broader (100° range) than those in the  $Mn_2Sb$  systems modified with the other elements. In the copper modified system, a two phase region appears near the composition  $Cu_{0.2}Mn_{1.8}Sb$  due to the formation of the intermediate cubic compound  $CuMnSb^6$ . For ternary compositions containing two transition elements, it was previously demonstrated that in the case of chromium, this element substitutes for manganese in the Mn(I) position<sup>2</sup> of the  $Mn_2Sb$  structure. In contrast, it has been found for germanium modified  $Mn_2Sb$  that the germanium substitutes for antimony in this structure.

<sup>1</sup> T. J. Swoboda et al., Phys. Rev. Letters 4, 509 (1960).

<sup>2</sup> W. H. Cloud, T. A. Bither, T. J. Swoboda, J. Appl. Phys. 32, 55S (1961).

<sup>3</sup> M. Yuzuri, M. Yamada, J. Phys. Soc. Japan, 15, 1845 (1960).

<sup>4</sup> A. E. Austin, E. Adelson, W. H. Cloud, Submitted to 1961 Conference on Magnetism and Magnetic Materials.

<sup>5</sup> M. Yuzuri, J. Phys. Soc. Japan, 15, 2007 (1960).

<sup>6</sup> H. Nowotny, B. Glatzl, Mon. Chem. 83, 237 (1952).

### 140. MAGNETIC STRUCTURES OF $Mn_2As$ AND $Mn_2Sb_{0.7}As_{0.3}$

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Neutron diffraction studies have been made on powder specimens of  $Mn_2As$  and  $Mn_2Sb_{0.7}As_{0.3}$ . Magnetic susceptibility measurements and specific heat measurements have indicated that  $Mn_2As$  is antiferromagnetic with a Néel temperature of 573°K.<sup>1</sup> Neutron diffraction measurements at room temperature show that the antiferromagnetic unit cell of  $Mn_2As$  has twice the tetragonal axis of the x-ray unit cell. The magnetic structure is the same layered structure as that of the isomorphous  $Mn_{1.9}Cr_{0.1}Sb$  below the exchange-inversion temperature.<sup>2</sup> In order to determine values for atomic moments the neutron diffraction intensities of  $Mn_2As$  powder were compared with those of a powdered specimen of  $Mn_{1.9}Cr_{0.1}Sb$  for which the atomic moments were known from previous single-crystal studies.<sup>3</sup> In  $Mn_2As$  the moments of Mn(I) and Mn(II) were found to be 3.7 and 3.5 Bohr magnetons respectively and to lie perpendicular to the tetragonal axis.

Neutron diffraction studies of  $Mn_2Sb_{0.7}As_{0.3}$  were made at room temperature and 150°C. The exchange-inversion transition in this compound takes place over the 45°C to 130°C temperature range.<sup>4</sup> The transition is the same type of antiferromagnetic-ferrimagnetic transition as has been observed in  $Mn_{1.9}Cr_{0.1}Sb$ .<sup>3</sup> The magnetic moments of Mn(I) and Mn(II) in the room-temperature antiferromagnetic state are 2.3 and 2.8 Bohr magnetons respectively.

In  $Mn_2As$  the Mn(I) atoms are at positions (0,0,0) and ( $\frac{1}{2}, \frac{1}{2}, 0$ ) and the Mn(II) and As atoms are at positions ( $0, \frac{1}{2}, z$ ) and ( $\frac{1}{2}, 0, \bar{z}$ ). Values of  $z_{Mn(II)} = +0.320$  and  $z_{As} = -0.275$  were determined from x-ray powder data.

Since suitable single crystals of  $Mn_2Sb_{0.7}As_{0.3}$  were not available, x-ray studies were made on single crystals of  $Mn_2Sb_{0.8}As_{0.2}$  which has an exchange inversion temperature below room temperature. A first-order transition was observed in the c axis of the unit cell similar to that observed in the  $Mn_{2-x}Cr_xSb$  system.<sup>5</sup> The critical value of c was 6.487Å, a value considerably smaller than the 6.530Å value observed for the  $Mn_{2-x}Cr_xSb$  system.

<sup>1</sup> M. Yuzuri, M. Yamada, J. Phys. Soc. Japan, 15, 1845 (1960)

<sup>2</sup> W. H. Cloud, H. S. Jarrett, A. E. Austin, E. Adelson, Phys. Rev. 120, 1969 (1960).

<sup>3</sup> W. H. Cloud, T. A. Bither, T. J. Swoboda, J. Applied Phys. 32, 55S (1961).

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<sup>5</sup> T. J. Swoboda, et al. Phys. Rev. Letters 4, 503 (1960)



#### 141. STRUCTURAL AND MAGNETIC PROPERTIES OF COPPER-SUBSTITUTED MANGANESE ALUMINIUM ALLOYS

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It has been investigated how the tetragonal structure and its magnetic properties in manganese-aluminum alloys are changed by an addition of copper. The composition range of the specimens used in this study is Al 45 atom per cent, Mn 55-43 atom per cent and the rest Cu 0-12 atom per cent. Each specimen was prepared by melting in air and casting into a bar. First, the specimens were annealed at 900°C for 3 hours to remove their precipitate and quenched into water. Then the specimens were aged at high temperatures. The change of crystal structure of the powdered specimens with the time of aging was investigated by x-ray analysis, and at the same time magnetic properties of the bulk specimens were measured.

The tetragonal phase showing high coercive force is found in the specimens with 0~10 atom per cent Cu after aging at 400-700°C. The specimen with about 12 atom per cent Cu has a ferrimagnetic cubic structure,  $\eta$  phase<sup>1</sup>, in both states, quenched and slow cooled. Generally this  $\eta$  phase is predominant even after aging at any high temperatures. However, some quantity of the tetragonal phase is found particularly after aging at about 500°C. The axial ratio  $c/a$  of the tetragonal phase decreases from 1.30 to 1.16 with the increase of Cu from 0 to about 10 atom per cent, followed by discontinuous change to 1.00, cubic phase.

The tetragonal phase in manganese-aluminum alloys is metastable and it decomposed into a  $\beta$ -Mn and an Al-rich  $\eta$  phase after aging for a longer time. Stability of the tetragonal phase, however, increases with an addition of copper. The bulk intrinsic coercive force,  $H_c$ , increases with aging, accompanied by an increase of the tetragonal phase. The highest value of  $H_c$  is obtained for the specimen without copper after aging at about 400°C. Generally  $H_c$  and  $I_r$  monotonically decrease with increase of copper content.

<sup>1</sup> I. Tsuboya and M. Sugihara, J. Phys. Soc. Japan **16**, 571 (1961)

#### 142. MAGNETIC SUSCEPTIBILITY AND INTERNAL FRICTION OF TETRAGONAL MANGANESE-COPPER ALLOYS CONTAINING 70 PER CENT MANGANESE

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The relationship of the structure to the magnetic susceptibility and internal friction is described for a 70 per cent manganese-30 per cent copper alloy having a tetragonal structure at temperatures up to 140°C. This alloy is typical of those having face centered tetragonal crystal

structures and containing from 60 to 80 per cent manganese.

The tetragonal structure results after partial precipitation of alpha manganese by aging the alloy. The 70 per cent alloy exhibits a reversible fct to fcc transition at temperatures from 20 to 140°C., depending on the amount of aging. The magnetic susceptibility follows essentially a Curie-Weiss law behavior, but with a change in slope of the temperature-susceptibility line at the structure transition point. The low stress internal friction of these alloys shows a resonance peak at the temperature of the structure change.

Additional information on aging experiments by the Bureau of Mines for 70 and 80 per cent manganese alloys are presented as evidence that quenching of alloys containing more than 80 per cent manganese from solid solution temperatures does not prevent changes typical of alloys aged to initiate precipitation of alpha manganese. Thus the fct-fcc transformation that others have associated with the disappearance of antiferromagnetic ordering also is associated with an early stage of the alpha manganese precipitation.

#### 143. ANOMALOUS MAGNETIC MOMENTS AND TRANSFORMATIONS IN THE ORDERED ALLOY FeRh

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Magnetic and electrical measurements were made over a wide temperature range on an iron-rhodium alloy (52 atom per cent Rh) having CsCl-type atomic order. From 77°K up to 350°K, this alloy has a low magnetic susceptibility ( $\approx 1 \cdot 10^{-4}$  emu/g) which is essentially independent of field and temperature, whereas just above 350°K, its magnetization rises rapidly with field to a saturation value of about 110 emu/g. This abrupt magnetic transition (with a small temperature hysteresis) is in qualitative accord with the results of Fallot and Hocart.<sup>1</sup> At still higher temperatures, the saturation magnetization is observed to decrease in a manner typical of a ferromagnet with a normal Curie temperature of 675°K, in disagreement with the precipitous decrease reported previously.<sup>1</sup> The electrical resistivity drops suddenly by about 30 per cent as the temperature is increased through 350°K but shows a more gradual Curie temperature behavior at 675°K.

The abrupt onset of ferromagnetism in this alloy at about 350°K has been recently found to be accompanied by a one per cent increase in volume,<sup>2</sup> indicating that this transformation is indeed first-order. Moreover, according to a very recent neutron diffraction study,<sup>3</sup> this transformation in FeRh constitutes a change from an antiferromagnetic to a ferromagnetic state. The experimental results presented here are consistent with this first-order antiferromagnetic-ferromagnetic transformation and are also indicative of unusual values for the atomic moments in this material. The saturation magnetization values obtained in the ferromagnetic range (350 to 675°K) extrapolate to a value at 0°K of 130 emu/g, which is equivalent to about  $1.9\mu_B$  for the average atomic moment; a similar result for the average atomic moment is deduced from paramagnetic data taken



above 675°K. If the magnetization of the alloy is associated entirely with the iron, the moment of each iron atom would have to be about  $3.9\mu_B$ . It seems more likely that the atomic moment of iron in FeRh is considerably less than this value and that each rhodium atom carries a substantial moment.

Preliminary measurements, in collaboration with P. E. Lawrence, indicate metamagnetic (antiferromagnetic-to-ferromagnetic) transitions in FeRh induced by large pulsed fields at temperatures below 350°K.

<sup>1</sup>M. Fallot and R. Hocart, *Revue scientifique* **8**, 498 (1938).

<sup>2</sup>F. de Bergevin and L. Muldower, *Comptes rendus* **252**, 1347 (1961).

<sup>3</sup>F. de Bergevin and L. Muldower, *Bull. Amer. Phys. Soc.* **6**, 159 (1961).

#### 144. ALLOYS OF THE FIRST TRANSITION SERIES WITH PALLADIUM AND PLATINUM\*

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Several ordered ferromagnetic alloys of iron group metals with palladium and platinum have been investigated by neutron diffraction methods to determine whether these higher transition series elements can possess a localized magnetic moment. The compositions investigated were FePd<sub>3</sub>, MnPt<sub>3</sub>, and CrPt<sub>3</sub>, all of which show a high degree of order of the Cu<sub>3</sub>Au type. We find the following results: in FePd<sub>3</sub> at room temperature the palladium moment is  $0.3 \pm 0.1\mu_B$ , the iron moment is  $2.4 \pm 0.1\mu_B$ ; in MnPt<sub>3</sub> at 77°K the platinum moment is  $0.0 \pm 0.1\mu_B$ , the manganese moment is  $4.0 \pm 0.2\mu_B$ ; in CrPt<sub>3</sub> at room temperature the platinum moment is  $-0.4 \pm 0.1\mu_B$ , the chromium moment is  $2.7 \pm 0.1\mu_B$ . These results were determined by combining the difference in moment of the two atomic species, which is related to the magnetic component of the superlattice intensities, with the total moment as determined from saturation magnetization measurements.<sup>1</sup> The magnetic contribution to the scattering was measured by application of an 8500 oe field normal to the scattering planes. The values given above depend of course upon the magnetic form factors used for the 3d, 4d, and 5d shell electrons. In the present study we used the form factor for Fe determined by polarized neutron measurements<sup>2</sup> and the Mn<sup>2+</sup> form factor<sup>3</sup> for Cr and Mn. We assumed further that the 4d form factor found<sup>4</sup> for Mo<sup>3+</sup> was appropriate for Pd and that at the lowest Bragg reflection the Pt form factor was effectively zero. Since the 4d and especially the 5d form factors fall off much more rapidly than the 3d, these latter assumptions are not overly critical. We are indebted to Dr. D.W. Ernst of the Naval Ordnance Laboratory for furnishing the alloys.

\*Work performed under the auspices of the U. S. Atomic Energy Commission.

\*\*Guest Scientist, Brookhaven National Laboratory.

<sup>1</sup>M. Fallot, *Ann. Phys. Paris* **10**, 291 (1938); M. Auwarter and A. Kussmann, *Ann. Phys.* **7**, 169 (1950); E. Friedrich and A. Kussmann, *Phys. Zeit.* **36**, 185 (1935).

<sup>2</sup>R. Nathans, C. G. Shull, G. Shirane, and A. Andresen, *J. Phys. Chem. Solids* **10**, 138 (1959).

<sup>3</sup>L. Corliss, N. Elliott, and J. Hastings, *Phys. Rev.* **104**, 924 (1956).

<sup>4</sup>M. K. Wilkinson, E. O. Wollan, H. R. Child, and J. W. Cable, *Phys. Rev.* **121**, 74 (1961).

#### 145. NEUTRON DIFFRACTION INVESTIGATIONS OF FERROMAGNETIC PALLADIUM AND IRON GROUP ALLOYS

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In order to account for the magnetic properties of alloys it becomes important to determine the individual magnetic moments of the constituent atoms. This determination can be accomplished by the combination of neutron diffraction and magnetic induction measurements. Such measurements were made on the following ferromagnetic alloys: Pd<sub>3</sub>Fe, PdFe, Pd<sub>3</sub>Co, PdCo, Ni<sub>3</sub>Co, and NiCo. The average moment values were obtained from magnetic induction measurements while the differences in the atomic moments were determined from either the ferromagnetic diffuse scattering of the disordered alloys or the superlattice reflections of the ordered alloys. In some cases, short-range-order effects were encountered and these will be discussed.

#### 146. MAGNETIC PROPERTIES OF MIXED VANADIUM-CHROMIUM SULFIDES

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Samples of Cr<sub>7</sub>S<sub>8</sub> have been reported to possess metallic conductivity and a superlattice structure. By using a quenching technique, we have prepared single-phase samples of pure Cr<sub>7</sub>S<sub>8</sub> which are semiconductors and which do not evidence a superlattice structure. The magnetization curve for our samples shows a broad peak in the neighborhood of 60°K, the magnetic moment gradually vanishing by 4.2°K. This type of magnetic behavior is at variance with that previously reported.

In addition, we have prepared pure, single-phase samples of the analogous compound V<sub>7</sub>S<sub>8</sub>, and find the magnetic properties to be different. In particular, the temperature variation of the magnetic moment appears to be ferrimagnetic in character, with a Curie temperature of about 70°K. The magnetization is extremely small, though about twice the peak value for our Cr<sub>7</sub>S<sub>8</sub>.

In order to illuminate the difference between the above two samples, we have prepared samples of several members of the stoichiometric series V<sub>x</sub>Cr<sub>1-x</sub>S. Such mixtures can be prepared as single-phase materials for values of x at least as small as 0.2 and at least as large as 0.9, al-



though neither VS nor CrS exist in stoichiometric form. The magnetic properties of these compounds show striking similarities to those of  $\text{Cr}_7\text{S}_8$  for small  $x$ , and to  $\text{V}_7\text{S}_8$  for large  $x$ . Some implications of the magnetic data regarding the origins of such behavior are considered.

\*Operated with support from the U. S. Army, Navy and Air Force.

#### 147. MAGNETIC MOMENT OF Co-Cu SOLID SOLUTIONS WITH 40 TO 85 PER CENT Cu

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In the binary system Co-Cu no thermodynamically stable solid solutions exist at any temperature in the concentration range between 5 and 90% Co. The following results give strong evidence that it is possible to obtain metastable solid solutions in this concentration range by simultaneous evaporation of copper and cobalt on a substrate, which is kept at room temperature or at a lower temperature.

Measurements of the intrinsic magnetization  $I_0$  of 1000 A Cu-Co films with 40 to 85% Cu give for the as evaporated films a linear dependence of  $I_0$  as a function of copper concentration.  $I_0$  vanishes at about 80% Cu. Within the accuracy of the measurements this is in quantitative agreement with the dependence of  $I_0$  on copper concentration, calculated for solid solutions under the assumption that the mean spin moment corresponds to the average number of holes in the 3d-shell. For an alloy with 66% Cu the Curie temperature is about 770°K.

After annealing of the films for one hour at 700°C the intrinsic magnetization is increased to values which correspond to mixtures of almost pure copper and a ferromagnetic phase containing 90% Co and 10% Cu. This is in agreement with the equilibrium properties of the alloys expected from the phase diagram of the system.  $I_0$  of annealed alloys is within the accuracy of the experiment independent of temperature between 77 and 700°K, thus indicating a Curie temperature much above this range, which is presumably the Curie temperature of the 90 Co-10 Cu phase at 1318°K.

Electron diffraction pictures show that the films have fcc structure before and after annealing.

#### 148. MAGNETIC SUSCEPTIBILITIES AND EXCHANGE EFFECTS IN FOUR ORGANIC FREE RADICALS\*

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We have measured the mass susceptibilities of the four free radicals: 1,1 diphenyl-2-picryl hydrazyl (DPPH), 1,3 bis-diphenylene-2-phenyl allyl (BDPA), picryl-n-amino carbazyl (PAC), and Wurster's blue perchlorate (WB) at temperatures ranging from 1.5°K to 300°K. These suscep-

tibilities were obtained primarily from the intensities of the EPR absorption line observed at various frequencies from 25 Mcps to 10 kMcps. In the case of BDPA some points were obtained from Knight Shifts of the proton NMR absorption. At high temperatures all four materials follow a Curie-Weiss law  $\chi = C/(T-\theta)$  with negative values of  $\theta_H$  (see Table I). In WB, PAC, and BDPA the susceptibility reaches a maximum at a temperature  $T_m$  (see Table I). Below  $T_m$  it decreases by an order of magnitude but more gradually than in typical antiferromagnetic materials. At still lower temperatures in WB and PAC the susceptibility again follows a Curie-Weiss law with positive values of the Weiss constant  $\theta_L$  (see Table I) indicating weak ferromagnetic coupling. The ratio of the Curie constant in this region to that at higher temperatures is about 1:80 for WB and 1:60 for PAC. The situation in DPPH is different in that no susceptibility maximum separates the two Curie-Weiss law regions and the Weiss constant  $\theta$  is negative in both regions. In agreement with earlier work we find a transition region between 77°K and 4.2°K in which the Curie constant decreases by about a factor of two. The susceptibility does appear to reach a maximum at about 2°K.

In other materials susceptibility behavior similar to that reported here has been explained in terms of exchange coupled pairs or triplets of paramagnetic centers. The susceptibility peak predicted by these models, however, is not sharp enough to fit the free radical data. The sharpness of the susceptibility peak, the large decrease in the Curie constant and the low temperature paramagnetism suggest the formation of larger clusters of spins. Another theory based on a model of random magnetic dilution predicts susceptibility behavior at low dilution qualitatively like that observed here. The magnetic concentration did not exceed 0.96 spins per free radical molecule in any of our samples.

TABLE I

	$\theta_H$	$\theta_L$	$T_m$
WB	$-39^\circ \pm 15^\circ\text{K}$	$0.6^\circ \pm 0.1^\circ\text{K}$	180°K
PAC	$-130^\circ \pm 10^\circ\text{K}$	$2.1^\circ \pm 0.1^\circ\text{K}$	75°K
BDPA	$-2^\circ \pm 1^\circ\text{K}$		6°K
DPPH	$-15^\circ \pm 10^\circ\text{K}$	$-0.4^\circ \pm 0.3^\circ\text{K}$	2°K

\*Work supported by the Air Force Office of Scientific Research.



# SESSION VIII B

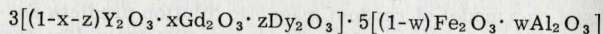
## OXIDES - 2 AND CRYSTALS

R. L. WHITE, Presiding

### 149. TEMPERATURE STABLE MICROWAVE HYBRID GARNETS\*

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Microwave garnets may be temperature stabilized by ionic substitution or what is sometimes called "molecular engineering." It is most desirable that the magnetization of the material exhibit temperature stability provided that in obtaining this stability the linewidth, g-factor, and Curie temperature of the material are still suitable for microwave applications. Most rare earth garnets possess compensation points in their magnetization vs temperature characteristics. These compensation points when suitably controlled by ionic substitutions can be used to produce microwave garnet materials whose magnetizations are quite temperature independent in the region from -25°C to +125°C. The microwave properties of the following compositions have been investigated:



for  $x = 0$  to 1.00,  $z = 0$  to 0.1, and  $w = 0$  to 0.1.

The magnetizations of these compositions have been studied from -195°C to the Curie temperatures. The compensation points of the magnetization are controlled by varying the rare earth and aluminum content of the material. The result is a series of materials possessing magnetizations in the region of 1200 to 300 gauss which vary no more than  $\pm 10\%$  over the temperature range of -25°C to 125°C. The other associated microwave properties of these compositions are as follows: linewidth (25°C, X-band) 35 to 800 oe;  $g_{eff}$ -factor (25°C, X-band) 2.10 to 1.80; Curie temperature, 200°C to 285°C. The thresholds for nonlinear effects in these materials are relatively high particularly for those compositions containing dysprosium. Many temperature stable microwave components have been developed for high power applications using these materials.

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### 150. MAGNETIC PROPERTIES OF YTTRIUM-GADOLINIUM-ALUMINUM-IRON GARNETS\*

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Polycrystalline garnets with the general composition  $Gd_{3-x}Y_xFe_{5-y}Al_yO_{12}$  were considered for this investigation.

In this structure the magnetic moment of the rare earth ions on the c sites is coupled antiferromagnetically to the resultant moment of the a-d sublattice ( $Fe^{+3}$  and  $Al^{+3}$ ). At low temperatures, where there is sufficient alignment of the c sublattice in the exchange field of the a-d sublattice, it is possible to have a compensation point, a point of zero magnetization, if the moment of the c ion is greater than the average a-d moment. For  $Gd_3Fe_5O_{12}$  such a compensation point occurs at room temperature. As a consequence of this compensation point,  $Gd_3Fe_5O_{12}$  has a temperature region where the saturation magnetization is independent of temperature ( $dM_S/dT = 0$ ). By the substitution of non-magnetic yttrium ( $Y^{+3}$ ) for gadolinium ( $Gd^{+3}$ ) and/or non-magnetic aluminum ( $Al^{+3}$ ) for iron ( $Fe^{+3}$ ), it is possible to shift this compensation point and the region where the temperature variation of the magnetization is zero. Measurements of the ferromagnetic resonance linewidth and saturation induction were made on materials in which  $x = 0$  to 3 and  $y = 0$  to 2. As predicted by the sublattice model, the compensation point and region of  $dM/dT = 0$  are shifted by the substitution of  $Y^{+3}$  and  $Al^{+3}$ . As an example, for  $x = 0$ ,  $y = 1.00$ ,  $4\pi M_S$  is equal to 600 gauss and  $dM_S/dT = 0$  at  $T = 20^\circ C$ . The Curie temperature of this composition is about 250°C and the ferromagnetic resonance linewidth is 100 oe at  $T = 20^\circ C$ . Observations of the temperature dependence of the magnetization and linewidth will be reported for the other compositions. Special emphasis will be given to those compositions which are of most technical importance; namely, those having low  $4\pi M_S$  and  $\Delta H$ ,  $dM/dT = 0$ , near room temperature, and a high Curie temperature.

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### 151. NICKEL ALUMINUM GALLIUM FERRITES FOR USE AT HIGH SIGNAL LEVELS

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Several investigators<sup>1,2,3</sup> have reported that nickel ferrite and nickel aluminum ferrites must be subjected to very high rf fields before non-linear effects set in. This behavior, plus their high Curie temperatures, make nickel ferrite and nickel aluminum ferrites attractive for microwave devices operating at high signal levels. However, in the nickel aluminum ferrites the compensation point, which occurs at  $x \approx 0.67$  in  $NiFe_{2-x}Al_xO_4$ , leads to excessively high  $g_{eff}$  factors for compositions from  $x \approx 0.3$  to 0.7. Thus nickel aluminum ferrites with saturation magnetizations near 1200 gauss and lower which are required at C-band and below, are not suitable for devices.

This problem has been solved by an extension of the work of Maxwell and Pickart.<sup>4</sup> They found that gallium substitutes in the tetrahedral sites in nickel ferrite while aluminum substitutes in the octahedral sites. Thus a simultaneous substitution of Ga and Al will shift the compensation point to very high concentrations of substituents, and the proper composition in the system  $NiFe_{2-(x+y)}Ga_xAl_yO_4$  will have no compensation point at all. However, the effect



of Ga on the Curie temperature is the same as that of aluminum. This is in contrast to zinc which lowers the Curie temperature more severely. The saturation magnetization will decrease as Ga and Al are added to the system in a manner which depends upon: (1) the ratio of Ga to Al, (2) the tenacity with which Ga and Al stay in the tetrahedral sites respectively, (3) the general weakening of magnetic interactions as the non-magnetic ion concentration increases, especially the A-B interaction.

The important properties of this system have been measured. Saturation magnetization, ferromagnetic resonance linewidth, Curie temperature,  $g_{\text{eff}}$ , dielectric loss, and the threshold for onset of non-linear effects are reported as a function of composition. At room temperature, substitutions of equal amounts of Al and Ga ( $x = y$ ) up to  $x + y = 1.2$  effect a linear decrease in saturation magnetization while  $g_{\text{eff}}$  stays constant at 2.4. Substitutions with  $2x = y$  show a faster decrease in magnetization, also linear, but  $g_{\text{eff}}$  remains small enough that these compositions are suitable for device use. Linewidths are near 200 oe for compositions suitable for devices operating at C-band and below.

The  $\text{NiFe}_{2-(x+y)}\text{Ga}_x\text{Al}_y\text{O}_4$  system appears to have the following advantages:

1. Saturation magnetization can be varied from over 2000 gauss down to 700 gauss while maintaining Curie temperatures of 200°C or more.

2. The substituents used,  $\text{Al}_2\text{O}_3$  and  $\text{Ga}_2\text{O}_3$ , are extremely stable and do not contribute to electrical conductivity through valence exchange mechanisms.

3. The system lends itself readily to the preparation of iron deficient compositions which exhibit higher resistivities than stoichiometric bodies.<sup>5</sup>

<sup>1</sup>E. Schlömann, J. H. Saunders, and M. H. Sirvetz, I.R.E. Trans. On Microwave Theory and Techniques **8**, 96 (1960).

<sup>2</sup>I. Reingold, S. Dixon, Jr., and J. L. Carter, U. S. Army Signal Research and Development Laboratory Technical Report 2143, Jan. 1960.

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<sup>4</sup>L. R. Maxwell and S. J. Pickart, Phys. Rev. **92**, 1120 (1953).

<sup>5</sup>L. G. Van Uitert, J. Chem. Phys. **23**, 1883 (1955).

## 152. RESONANCE PROPERTIES OF SINGLE CRYSTAL HEXAGONAL FERRITES

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Ferromagnetic resonance measurements have been performed on single crystal samples of the hexagonal ferrites  $\text{BaFe}_{12}\text{O}_{19}$ ,  $\text{Zn}_2\text{Y}$ , and  $\text{Co}_2\text{Y}$ . Values of linewidth,  $g$  value, and anisotropy field are given as a function of temperature from 77°K to 600°K at various frequencies between 8 kMc and 75 kMc.  $\text{Zn}_2\text{Y}$  is singularly interesting because it probably exhibits the narrowest room temperature linewidth of any of the highly anisotropic ferrites. Linewidths as low as 18 oe, with anisotropy fields of 9000 oe are

reported. The theory of the resonance behavior is developed and compared to the experimental results.

## 153. HEXAGONAL FERRITES FOR USE AT X- TO V-BAND FREQUENCIES

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A series of uniaxial materials have been prepared and studied in which the effective anisotropy field can be controlled and varied over the range of 0 to 12,700 oe. Different materials of this series have been used to construct resonant isolators at V-band and lower frequencies with either very small applied magnetic fields or no external magnetic field whatsoever. By using various members of this series prepared to the proper composition, magnetless isolators can be made at any frequency (linewidth permitting) below about 40 kMc. The linewidth of these materials is sufficiently small to permit their use in phase shift devices at the higher frequencies. The materials are solid solutions of the Ni W and Co W compounds and have the chemical formula  $\text{BaO} \cdot (2-x)\text{NiO} \cdot x\text{CoO} \cdot 7.8\text{Fe}_2\text{O}_3$  with  $x$  varying from 0 where the anisotropy field is 12,700 oe to 1.0 where the anisotropy field is vanishingly small. Control of the magnetocrystalline anisotropy is achieved by balancing the negative first order anisotropy constant of Co W against the positive first order anisotropy constant of Ni W. Resonance studies have been made of the temperature dependence of the net resultant anisotropy field and the effect of temperature on the balancing of anisotropies. By using oriented polycrystalline samples of these materials the effective internal anisotropy field can be used to augment or replace externally applied fields. The anisotropy fields and temperature characteristics of this series of materials will be discussed as will their application to various practical devices.

## 154. SPIN WAVE EXCITATION IN PLANAR FERRITES

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Planar ferrites are ferrites with a preferential plane of magnetization. The large crystalline anisotropy encountered in many of these materials markedly affects the rf magnetic field at which spin-wave instability sets in. We have calculated the instability threshold under various assumed experimental conditions for the case in which the dc magnetic field is applied in the preferential plane.

If the rf magnetic field is applied parallel to the dc field, the presence of the anisotropy tends to decrease the instability threshold. At low dc fields the critical rf field is reduced by a factor of  $4\pi M / (4\pi M + H_a)$ . Here  $4\pi M$  is



the saturation magnetization and  $H_a$  is the anisotropy field, and we have assumed that the saturation magnetization and the spin-wave decay rates are the same in the two situations compared. The anisotropy also modifies the general shape of the "butterfly curve" representing the critical rf field as a function of the dc field.

If the rf magnetic field is applied perpendicular to the dc field, and the dc field adjusted to ferromagnetic resonance, spin-wave instability can occur through the first- or the second-order nonlinear process. The first-order process is important at low signal frequencies and involves spin waves whose frequencies equal half the signal frequency. Conversely, the second-order process is important at high signal frequencies and involves spin waves whose frequencies equal the signal frequency. The characteristic frequency separating the first- and second-order regions is dependent on the anisotropy. It increases with increasing anisotropy for all spheroids magnetized along their axes, but decreases for a disc cut parallel to the preferential plane. The instability threshold in the first- and second-order region has been calculated as a function of frequency for the case of a sphere and for several values of  $4\pi M$  and  $H_a$ . The results show that the spin waves excited in the second-order region do not necessarily propagate along the direction of the dc field (as is the case in the absence of anisotropy). The frequency dependence of the critical rf field differs markedly from that obtained in the absence of anisotropy.

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#### 155. INVESTIGATION OF SPIN-WAVE INTERACTIONS BY THE PARALLEL PUMPING TECHNIQUE\*

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The subharmonic generation of spin waves at high power levels has been used as a research tool for probing magnon-magnon and magnon-phonon interactions. Useful information can be obtained from measurements of the instability threshold as a function of the dc field, from measurements of the susceptibility beyond the threshold, and from the transient response to a pulsed rf field.

Measurements of the instability threshold have shown that in pure yttrium-iron garnet (YIG) the spin-wave relaxation rate depends quite sensitively on the magnitude as well as the direction of the propagation vector. By contrast, similar measurements on YIG containing appreciable amounts of rare earth impurities indicate that the contribution of the rare earth ions to the relaxation rate is independent of the propagation vector.

Measurements of the high-power susceptibility as a function of dc field at a given power level have been performed at various frequencies. The measured curves show a well defined structure which can largely be attributed to relaxation processes in which two parametrically excited magnons coalesce into a third magnon under conservation of energy and momentum.

Under the influence of a pulsed rf field the spin-wave population initially builds up at a rate proportional to the difference between the actual rf field and the critical field. Under some conditions a persistent amplitude modulation of the power reflected from the microwave cavity containing the sample has been observed. Measurements of the modulation frequency as a function of power level will be reported.

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#### 156. SUBSIDIARY ABSORPTION EFFECTS IN FERRI-MAGNETICS\*

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Cavity perturbation techniques have been used to study the subsidiary absorption in the following polycrystalline materials: (1) yttrium iron garnet, (2) yttrium iron garnet with partial substitution of aluminum for iron, (3) nickel ferrite, and (4) nickel ferrite with partial substitution of aluminum for iron. Using perpendicular pumping we have found that the values of  $\Delta H_K$  (5 - 10 oe) inferred from the threshold fields are of the same order of magnitude as those previously obtained by resonance saturation experiments. Parallel pumping was used on several of the garnet samples and the values of  $\Delta H_K$  obtained coincided with those obtained from perpendicular pumping. The dependence of the critical field on the dc field for parallel pumping indicates the presence of rare earth impurities. This would account for the difference in values of  $\Delta H_K$  found in these polycrystalline samples and good single crystal yttrium iron garnet. At power levels several db above the subsidiary absorption threshold a power dependent loss appears at zero applied dc field. The power level at which this loss appears is independent of the sample shape whereas the threshold for subsidiary absorption as measured by perpendicular pumping does depend on the sample shape. In a single crystal sample of yttrium iron garnet it was possible to make measurements up to 24 db above threshold. It was found that the power dependence of the zero field loss is similar to that of the subsidiary absorption.

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## 157. HIGH-POWER CHARACTERISTICS OF SINGLE-CRYSTAL FERRITES WITH PLANAR ANISOTROPY

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Single crystal ZnY ( $\text{Ba}_2\text{Zn}_2\text{Fe}_{12}\text{O}_{22}$ ) was used at X-band frequencies in an investigation of the variation of the magnetic susceptibility with microwave magnetic field. In this material, the large anisotropy field that tends to keep the magnetization in the easy plane, reduces the field required for ferromagnetic resonance. The low-level linewidth varied from 150 to 20 oe. These measurements indicate that this material has significant applications in microwave devices, such as isolators, circulators, and power limiters. The variation of the magnetic susceptibility with the microwave magnetic field for single-crystal ferroplanar ( $\Delta H = 20$  oe) has been compared with that of single crystal, yttrium-iron-garnet. The critical field and corresponding spin-wave resonance linewidth has been determined. Measurements on the material with a linewidth of 150 oe indicate that as the power level is increased, there is a shift of 600 oe toward the high-field side in the applied dc field required for resonance. This effect could be used in the design of a new type power limiter. It has not been determined if this shift in the applied dc field required for resonance is an intrinsic characteristic of the material.

## 158. THE EFFECT OF GROWTH CONDITIONS AND SUBSEQUENT THERMAL AND MECHANICAL TREATMENT ON THE FERRIMAGNETIC RESONANCE LINEWIDTH OF LITHIUM FERRITE SINGLE CRYSTALS

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The purpose of this paper is to describe how ferrimagnetic resonance linewidths less than 3.5 oe can be achieved in lithium ferrite crystals at room temperature and 5 kMc. Experiments directed toward understanding the "large and unexpected effects"<sup>1</sup> of heat treatment and polishing on the linewidth are also described.

The crystals of lithium ferrite were grown from molten salts under several conditions of composition and ambient. Crystals grown under the same conditions and processed in the same way exhibited the same linewidths, but the changes in linewidth with processing were unusual. For example, crystals grown from PbO in air pass through a minimum in  $\Delta H$  with increasing surface perfection. This minimum occurs at 6 to 7 oe and at a point where several surface pits can be clearly seen. As the surface is further polished  $\Delta H$  climbs to over 10 oe.

Spheres from the same run were also given heat treatment in air at 820°C.  $\Delta H$  again passed through a minimum as time at temperature was increased. The value of  $\Delta H$  at the minimum depended upon the original state of polish, but spheres polished to a minimum  $\Delta H$  and a sphere polished

further to a fine finish, which possessed a  $\Delta H$  of 11 oe, exhibited the same minimum  $\Delta H$  of about 5 oe after heat treatment. The minimum occurred at about 3 hours at 820°C. Spheres from crystals grown in oxygen which were heat treated in oxygen behaved similarly, but the minimum linewidth observed was less than 4 oe.

These results show the overwhelming effect of surface condition on  $\Delta H$  in lithium ferrite spheres. The minimum observed during polishing must be evidence for the removal of strains caused by rough grinding through removal of material. This is followed by the introduction of high surface disorder on a submicroscopic scale caused by the final polish. This result is in sharp contrast to YIG which shows a continuous decrease in  $\Delta H$  with surface improvement.

The decrease from 6 to 7 oe (the polishing minimum) to 4 to 5 oe with heat treatment is the result of oxidation of the sphere. This, too, is essentially a surface effect. Measurements of electrical conductivity of lithium ferrite slabs showed that heating in air sharply reduced the conductivity but only to a depth of about .05mm in the time required for  $\Delta H$  of a sphere to reach a minimum under the same conditions. The initial resistivity of the crystals was less than 100 ohm-cm. Thus the 6 to 7 oe  $\Delta H$  observed before heat treatment is unusually low compared to that observed in other spinels with similar resistivities.

The increase in  $\Delta H$  with prolonged heat treatment is not understood, but the possibility of  $\text{Li}_2\text{O}$  evaporation is under study. These experiments will be described in detail.

Further experiments on spheres with very high resistivities and the technique for attaining high resistivities will be reported. Such spheres have exhibited linewidths as low as 3.3 oe at 5 kMc and 28°C.

<sup>1</sup>A. D. Schnitzler, V. J. Folen, and G. T. Rado, J. App. Phys. Supp. 31, 348S (1960).

## 159. THE GROWTH OF YTTRIUM-IRON GARNET ON A SEED FROM A MOLTEN SALT SOLUTION

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Yttrium-iron garnet has been crystallized on a seed crystal from molten  $\text{BaO-xB}_2\text{O}_3$  where x is 0.61. Two methods were used: (1) Slow cooling of a melt in which a rapidly rotating seed was suspended, and (2) growth in a temperature gradient where excess yttrium-iron garnet was maintained in a hotter part of the system and a rapidly rotating seed was suspended in a cooler region. The geometry of the furnace, crucible, baffle, stirrer and circulator system required to produce controlled growth in each of these systems is described and the nature of the rate limiting step in each of the systems is discussed. The dependence of growth rate in the [110] direction on stirring rate, cooling rate, and solvent weight is presented. Good quality growth with rates as high as 50-75 mil/day was achieved in favorable cases.



## 160. COBALT FERRITE CRYSTAL GROWTH FROM THE TERNARY FLUX SYSTEM $\text{Na}_2\text{O}-\text{CoO}-\text{Fe}_2\text{O}_3$

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It is shown that the simple ferrite spinels ( $\text{M}^{+2}\text{Fe}_2\text{O}_4$ ) crystallize readily from the ternary flux system  $\text{MO}-\text{Na}_2\text{O}-\text{Fe}_2\text{O}_3$ . The liquidus-solidus phase diagrams for the pseudo-binary of  $\text{Na}_2\text{Fe}_2\text{O}_4-\text{CoFe}_2\text{O}_4$  and the ternary  $\text{Na}_2\text{O}-\text{CoO}-\text{Fe}_2\text{O}_3$  are presented. A general discussion of flux growth of ferrite crystals of both the simple spinel type and the more complex layered structures ( $\text{BaFe}_{12}\text{O}_{19}$ ) is given:

\*Operated with support from the U. S. Army, Navy, and Air Force.

## 161. GROWTH OF SINGLE-CRYSTAL HEXAGONAL FERRITES CONTAINING Zn

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Single crystals of ZnY ( $\text{Ba}_2\text{Zn}_2\text{Fe}_{12}\text{O}_{22}$ ), ZnW ( $\text{BaZn}_2\text{Fe}_{16}\text{O}_{27}$ ), ZnZ ( $\text{Ba}_3\text{Zn}_2\text{Fe}_{24}\text{O}_{41}$ ), Zn<sub>2</sub>Y (X) ( $\text{Ba}_4\text{Zn}_2\text{Fe}_{30}\text{O}_{60}$ ), Zn<sub>2</sub>S (V) ( $\text{Ba}_2\text{Zn}_2\text{Fe}_{28}\text{O}_{46}$ ), as well as  $\text{ZnFe}_2\text{O}_4$ , have been grown using a flux by recrystallization from the melt in platinum crucibles. Equilibrium and crystallization data indicate that the fluxing compound is  $\text{Na}_2\text{FeO}_4$ . Crystallization of these compounds is considered within the quaternary system  $\text{Na}_2\text{FeO}_4 - \text{Fe}_2\text{O}_3 - \text{BaO} - \text{ZnO}$ . The compositions investigated all lie on three different planes. The different crystalline composition fields intersecting these planes are generally defined.

Mixtures of oxides and/or carbonates are heated in platinum crucibles to 1250 to 1350°C and cooled at 2° to 4°C/hr. to 1050°C. Subsequently the crucibles are removed from the furnace and cooled to room temperature in air. Leaching of the crystals is accomplished with boiling, dilute (10 to 20% by volume) nitric acid.

Crystals obtained from this system are characteristically black with highly reflecting natural faces and easy, almost perfect basal cleavage. Morphologically, ZnY crystals were generally tabular with well-developed basal pinacoid(s), varying from hexagonal prismatic to pyramidal. The density of the ZnY crystals was found to be  $5.44 \pm .04 \text{ g/cm}^3$  (x-ray density  $5.46 \text{ g/cm}^3$ ) even though thin sections, transparent and red in color for thicknesses of .005 in., of these crystals reveal 5-20 $\mu$  octahedral inclusions of a second phase.

Ferromagnetic resonance line width made on discs ground from these crystals at 9kMcps has been found to be as low as 15 oe by other workers.<sup>1,2</sup> High power experi-

ments employing spheres ground from crystals having line widths of 200 - 300 oe exhibit a large shift (600 oe at 2000 watts peak power applied to the cavity) in the applied dc field.<sup>2</sup>

\*Deceased.

<sup>1</sup>C. R. Buffer - Varian Associates.

<sup>2</sup>S. Dixon - USASRD.



## AUTHORS INDEX

AUTHOR	PAPER NUMBER	AUTHOR	PAPER NUMBER
Adams, E. . . . .	78	Brown, W.F., Jr. . . . .	131
Adelson, E. . . . .	140	Buffler, C.R. . . . .	152
Aharoni, A. . . . .	130	Burgess, J.H. . . . .	148
Aleonard, R. . . . .	68	Burket, R.E. . . . .	85
Alexander, E.J. . . . .	17	Cable, J.W. . . . .	47, 145
Alexander, S. . . . .	42	Calhoun, B.A. . . . .	92
Allen, D.E. . . . .	99	Callen, H. . . . .	60
Arajs, S. . . . .	137	Carr, J.M. . . . .	126
Argyle, B.E. . . . .	62	Caspari, M.E. . . . .	22
Arnold, G. P. . . . .	46	Cebulla, T.J. . . . .	54
Arnott, R. J. . . . .	73	Chen, D. . . . .	55
Arrott, A. . . . .	66	Chessin, H. . . . .	137
Aspden, R.G. . . . .	82	Clark, J. . . . .	102
Astrue, R.W. . . . .	29	Cloud, W.H. . . . .	74, 139, 140
Auld, B. A. . . . .	101	Colvin, R.V. . . . .	137
Austin, A.E. . . . .	140	Conger, R.L. . . . .	7
Auten, B.H. . . . .	104	Coren, R.L. . . . .	57
Babbitt, R.W. . . . .	77	Corliss, L.M. . . . .	44
Babcock, K.R. . . . .	74	Crapo, W.A. . . . .	114
Bady, I. . . . .	154	Cronk, E.R. . . . .	127
Bagrowski, J. . . . .	37	Daniel, E.D. . . . .	134
Ballantyne, J.M. . . . .	12	Dearborn, E.F. . . . .	159
Baltz, A. . . . .	35	de Bergevin, F. . . . .	41
Banerjee, B.R. . . . .	88	Deetscreek, V.D. . . . .	43
Banks, E. . . . .	160	Delapalme, A. . . . .	41
Barbier, J. . . . .	80	Demars, G.A. . . . .	95
Bate, G. . . . .	133	Denton, R.T. . . . .	118
Bean, C.P. . . . .	2	de Vos, K.J. . . . .	125
Belson, H.S. . . . .	120	Dillon, J.F. . . . .	65
Bennett, L.H. . . . .	26	Dixon, S., Jr. . . . .	157
Bertaut, E.F. . . . .	40, 41, 68	Douglas, L. . . . .	9
Bierstedt, P.E. . . . .	139	Doyle, W.D. . . . .	52
Bither, T.A. . . . .	139	Dwight, K. . . . .	45, 146
Bohlmann, M.A. . . . .	128	Edelstein, A.S. . . . .	148
Boxer, A.S. . . . .	98	Elfant, R.F. . . . .	15
Boyd, E.L. . . . .	24	Elkort, D.S. . . . .	103
Bradley, E.M. . . . .	6	Espinosa, G.P. . . . .	69
Briggs, G.R. . . . .	11	Facaros, G. . . . .	82
Brown, J. . . . .	102	Falkowski, E.C. . . . .	85
Brown, P.J. . . . .	25	Ferlin-Guion, B. . . . .	80

AUTHOR	PAPER NUMBER	AUTHOR	PAPER NUMBER
Flanders, P.J. . . . .	135	Hou, S. . . . .	115
Francombe, M.H. . . . .	38	Irons, H.R. . . . .	16
Frankel, S. . . . .	22	Jaccarino, V. . . . .	64
Freedman, J.F. . . . .	53, 58	Jensen, J.W. . . . .	142
Freeman, A.J. . . . .	21, 96	Johns, R.H. . . . .	85
Freiser, M.J. . . . .	92	Johnson, C.E., Jr. . . . .	132
Friedlaender, F.J. . . . .	15	Kaufman, I. . . . .	100
Folen, V.J. . . . .	19, 119	Kernohan, R.H. . . . .	86
Foner, S. . . . .	115	Kneller, E. . . . .	147
Forrat, F. . . . .	41	Kocher, C.W. . . . .	25
Fuller, H. W. . . . .	10	Koehler, W.C. . . . .	47, 145
Gambino, R. . . . .	161	Koi, Y. . . . .	18
Garvin, S.J. . . . .	127	Kolk, A. . . . .	9
Geller, S. . . . .	69	Kouvel, J.S. . . . .	143
Germann, R.W. . . . .	146	Kronenberg, K.J. . . . .	129
Geschwind, S. . . . .	20	Kubota, Y. . . . .	138
Geyger, W.A. . . . .	108	Kunmann, W. . . . .	160
Goldberg, N. . . . .	120	Kunzler, J. E. . . . .	4
Goodenough, J.B. . . . .	75	Kushida, T. . . . .	18
Gordon, E.I. . . . .	67	Laudise, R.A. . . . .	159
Gossard, A.C. . . . .	64	Lauriente, M. . . . .	37
Graham, C.D., Jr. . . . .	51	Lax, B. . . . .	3
Gran, B.E. . . . .	54	Leaver, K.D. . . . .	33
Green, J.J. . . . .	150, 155, 156	LeCraw, R.C. . . . .	67
Greifer, A.P. . . . .	72	Lee, R. . . . .	112
Gutwin, O.A. . . . .	13	Lemke, J.S. . . . .	36
Gyorgy, E.M. . . . .	84, 136	Lenhart, R.E. . . . .	88
Haakana, C.H. . . . .	66	Lepore, D.A. . . . .	158
Haas, C.W. . . . .	120	Lessoff, H. . . . .	72
Harris, E.S. . . . .	83	Li, K. . . . .	14
Harrison, G.R. . . . .	149	Linares, R.C. . . . .	159
Hastings, J.M. . . . .	44	Littmann, M.F. . . . .	83
Harte, K.J. . . . .	28	Lommel, J.M. . . . .	51
Hartelius, C.C. . . . .	143	Long, T.R. . . . .	32
Hedgcock, F.T. . . . .	23	Loriers, J. . . . .	94
Heinz, W. . . . .	123	Lykens, A.A. . . . .	87
Heller, G.S. . . . .	116	Lykken, G.I. . . . .	59
Henry, W.E. . . . .	94	Maguire, E.A. . . . .	150
Hickernell, F.S. . . . .	104	Malinofsky, W.W. . . . .	77
Hodges, L.R., Jr. . . . .	149	Mandel, M. . . . .	148
Holmen, J.O. . . . .	54	Matcovich, T.J. . . . .	120



AUTHOR	PAPER NUMBER	AUTHOR	PAPER NUMBER
McAlexander, J.C. . . . .	17	Robinson, W.H. . . . .	88
McCarter, R.S. . . . .	98	Rodbell, D.S. . . . .	2, 49
McGuire, T.R. . . . .	114	Rodrigue, G.P. . . . .	153
Meiklejohn, W.H. . . . .	124	Rogers, D. . . . .	45, 73
Menyuk, N. . . . .	45, 73, 146	Roult, G. . . . .	41
Middelhoek, S. . . . .	30	Rubens, S.M. . . . .	34
Mitchell, E.N. . . . .	59	Rudisill, J.E. . . . .	52
Montmory, M.C. . . . .	68	Salhany, R.J. . . . .	17
Moore, R.A. . . . .	105	Sallo, J.S. . . . .	126
Morgenthaler, F.R. . . . .	121	Sakiotis, N.G. . . . .	99, 104
Moriya, T. . . . .	117	Sands, G.C. . . . .	77
Morrish, A.H. . . . .	55	Sato, H. . . . .	29
Muir, W.B. . . . .	23	Saunders, J.H. . . . .	156
Nagamiya, T. . . . .	1	Savage, R.O. . . . .	161
Nathans, R. . . . .	144	Schell, R. . . . .	101
Nereson, N.G. . . . .	46	Schenk, H.L. . . . .	109
Nesbitt, E.A. . . . .	136	Schindler, A.I. . . . .	86
Nielsen, J.W. . . . .	151, 158	Schlömann, E. . . . .	154, 155
Noakes, J.E. . . . .	66	Schnitzler, A.D. . . . .	119
Noble, R. . . . .	134	Schrader, G. . . . .	9
Noreika, A.J. . . . .	38	Schreiber, D.S. . . . .	74
Olmen, R.W. . . . .	34	Schuele, W.J. . . . .	43
Olsen, C.E. . . . .	46	Schwaneke, A.E. . . . .	142
Palmer, W. . . . .	70	Schwenker, J.E. . . . .	32
Pauthenet, R. . . . .	41, 68	Seiden, P.E. . . . .	93
Pearson, R.F. . . . .	91	Shafer, M.W. . . . .	71
Peck, J.M. . . . .	137	Shahan, V.T. . . . .	13
Penoyer, R.F. . . . .	92	Sherwood, R.C. . . . .	69
Peter, M. . . . .	117	Shtrikman, S. . . . .	52, 135
Pickart, S.J. . . . .	144	Silber, L. . . . .	123
Pippin, J.E. . . . .	153	Silver, A.H. . . . .	18
Pomerantz, M. . . . .	53	Simkins, Q.W. . . . .	5
Prosen, R. J. . . . .	54	Simpson, A.W. . . . .	76
Prutton, M. . . . .	33	Slonczewski, J.C. . . . .	24
Pugh, E.W. . . . .	8, 62	Smith, D.O. . . . .	12, 28, 56
Rado, G.T. . . . .	39, 119	Sooahoo, R.F. . . . .	106
Remeika, J.P. . . . .	64	Solt, I.H., Jr. . . . .	63
Rhodes, R.S. . . . .	148	Spencer, E.G. . . . .	67, 118
Rimai, L. . . . .	95	Steeg, M.G.v.d. . . . .	125
Risley, A.S. . . . .	100	Steinort, E. . . . .	127
Roberts, R.W. . . . .	101	Stewart, D.M. . . . .	85

AUTHOR	PAPER NUMBER	AUTHOR	PAPER NUMBER
Stickler, J.J. . . . .	116	Walker, L.R. . . . .	89
Strattan, R.D. . . . .	110	Wallace, W.E. . . . .	138
Streever, R.L., Jr. . . . .	26	Walter, J.L. . . . .	79
Sugihara, M. . . . .	141	Walter, P.H.L. . . . .	139
Suhl, H. . . . .	113	Ward, C.E. . . . .	83
Sullivan, D.L. . . . .	10	Watson, R.E. . . . .	21, 96
Suits, J.C. . . . .	8, 53	Weiss, G.P. . . . .	56
Swoboda, T.J. . . . .	139	Whinfrey, C. . . . .	161
Takahashi, M. . . . .	27	Wieder, H.H. . . . .	107
Tauber, A. . . . .	161	Wilkinson, M.K. . . . .	47, 145
Teale, R.W. . . . .	122	Williams, H.J. . . . .	69
Thomas, H. . . . .	31	Wojtowicz, P.J. . . . .	48, 97
Tiderman, H. . . . .	127	Wold, A. . . . .	45, 73, 146, 160
Tinkham, M. . . . .	90	Wolf, I.W. . . . .	50
Toth, R.S. . . . .	29	Wolff, P.A. . . . .	61
Townsend, G.B. . . . .	158	Wollan, E.O. . . . .	47, 145
Treves, D. . . . .	42, 84	Wood, G.T. . . . .	22
Tsuboya, I. . . . .	141	Wycklendt, D.A. . . . .	111
Tsujimura, A. . . . .	18	Yamada, O. . . . .	81
Tuska, J.W. . . . .	11	Young, F.J. . . . .	109, 110
Velge, W.A.J.J. . . . .	125	Young, L.H. . . . .	17
Villers, G. . . . .	94	Zneimer, J. . . . .	151, 158



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