Magnetic Materials for Millimeter Wave Applications*

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Summary—Magnetic materials of interest at millimeter wavelengths can be grouped under four headings: 1) conventional polycrystalline ferrites with cubic crystal structure, 2) yttrium iron garnet and similar single crystals, 3) ferrites having hexagonal crystal structures, and 4) antiferromagnetic materials.

The conventional polycrystalline ferrites have been used in constructing millimeter wave phase shifters, circulators, and Faraday rotation isolators.

Yttrium iron garnet single crystals are distinguished by their extremely small damping and resultant narrow ferromagnetic resonance absorption line, of the order of 0.3 oersted at *X*-band frequencies. The small damping of the precessing magnetization makes this material useful as a relatively high V, tunable, resonant circuit with *Q*'s of the order of 3000.

Magnetic materials with hexagonal crystal structure whose preferred direction of magnetization lies along the *C* axis are said to be uniaxial, and their effective internal anisotropy fields range from virtually zero to greater than 30,000 oersteds. The uniaxial materials are useful in compact, light weight resonance load isolators, harmonic suppressors, etc., in any application where their large effective internal field can be used to advantage.

The antiferromagnetic materials have extremely large effective internal fields and can be used in resonance devices in the frequency range from 2 millimeters into the submillimeter region.

Introduction

MAGNETIC MATERIALS have been used in a wide variety of applications at microwave frequencies, and it is perhaps natural for those familiar with microwave systems and components to think of extending their application to the millimeter and submillimeter range. The existing considerable background of information on the fundamental properties of magnetic substances should serve as the basis for the realization of a broad range of millimeter and submillimeter devices. To point up the potentialities of these materials, the most pertinent characteristics of the different classes of magnetic materials will be reviewed and their possible areas of application discussed.

The applicable magnetic materials can be grouped into four somewhat arbitrary classes. These are 1) conventional isotropic polycrystalline ferrites, 2) yttrium iron garnet and similar materials in single crystal form, 3) highly anisotropic ferrites having hexagonal crystal structure, and 4) highly anisotropic antiferromagnetic materials. Each of these classes will be discussed separately.

The operation of nearly all microwave magnetic devices can be described in terms of the effective susceptibilities of the material for circularly polarized RF radiation. Fig. 1 shows the variation of the real and imaginary components of the effective susceptibility as a function of applied dc magnetic field. Microwave ferrite devices can be grouped into two classes, those operating with the magnetic material biased near the region of ferromagnetic resonance, and those operating with the material well away from resonance. The resonance devices include load isolators, harmonic generators, many of the switches, filters, and resonators themselves. Devices operating outside the resonance region include Faraday rotation isolators, phase shifters, circulators, phase shift type switches and filters.

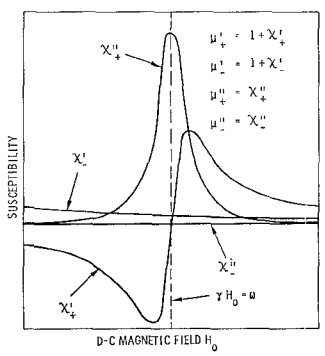

Fig. 1—Real and imaginary parts of effective susceptibilities for positive (χ_+) and negative (χ_-) senses of circular polarization.

The operation of a resonance isolator, for example, is based on the fact that through proper design a wave propagating in one direction is attenuated by an amount corresponding to the peak of χ_+'', while the wave propagating in the opposite direction will be passed with only negligible attenuation characteristic of χ_-''.

Off resonance devices base their operation in the different effective permeability which the material exhibits to positively and negatively circularly polarized waves. Two waves of different polarization experience a differential phase shift as a result.

When considering millimeter and submillimeter wave applications several difficulties arise that preclude a simple extension of microwave techniques. First there is the common problem of the extremely small dimensions and exacting mechanical tolerances encountered at the higher frequencies. In resonance applications there is the further difficulty that the ferromagnetic resonance absorption peak occurs for an isotropic, spherically shaped sample at a frequency given by

$$\omega = \gamma H_0,$$

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where γ is the gyromagnetic ratio (usually 2.8 Mc/oersted) and H_0 is the applied dc magnetic field. A dc field of approximately 10,000 oersteds would be required for frequencies of the order of 28,000 Mc. For higher frequencies proportionately larger fields would be required. Clearly such fields are inconveniently large. For differential phase shift type devices it can be shown that the difference between the magnetic susceptibilities $(\chi_+' - \chi_-')$ grows smaller as the frequency is increased, and ultimately limits the performance of either Faraday rotation or nonreciprocal phase shift devices.

Isotropic Ferrites

Because of the large dc field required to bias them to resonance at millimeter wave frequencies, conventional isotropic, polycrystalline ferrites are usually restricted to Faraday rotation[1] devices at these wavelengths.

Materials intended for phase shift applications at high frequencies should exhibit low over-all loss, *i.e.*, low dielectric loss and low off resonance magnetic losses. The magnetization should preferably be large to achieve a high degree of activity (or rotation) in the material and the Curie temperature should be relatively high to impart some degree of temperature stability. Materials like nickel ferrite, nickel zinc ferrite, and magnesium manganese ferrite are quite suitable.

At millimeter wave frequencies Faraday rotation isolators are commercially available that typically have attenuation ratios of the order of 20 db to 1 db over approximately 2-Gc bandwidths in the frequency ranges from 26 to 75 Gc. Above 75 Gc the performance of these devices begins to deteriorate, but ratios of the order of 10 to 1 are obtained up to about 140 Gc.

If a solenoid is used to supply the small dc biasing field required for rotation devices, they can be made into tunable isolators,[2] off-on switches,[3] or modulators.[4,5] Because of the configuration normally used in such devices they are rather susceptible to high power heating, and their comparative complexity results in some difficulties in fabrication for millimeter wavelengths.

A slightly different type of off resonance device is the Y-junction circulator. Using magnesium manganese ferrite with an applied field of only 200 oersteds Thaxter and Heller[6] constructed circulators for operation at 70 and at 140 Gc.

Still another application of isotropic ferrites to millimeter wave devices involves their use as harmonic generators. Ayers[7] reported frequency doubling from 4 millimeters to 2 millimeters with conventional ferrite. Fifty watts of peak power were obtained at 2 millimeters from 4 millimeters excitation. One important material parameter for this application appears to be the ratio of the saturation magnetization to the resonance linewidth of the material or $4\pi M_s/\Delta H$.[8] While this means of harmonic generation is capable of large output powers, it requires the application of sufficiently large magnetic fields to bias the material to resonance at the fundamental frequency.

Yttrium Iron Garnet Crystals

Single crystals of yttrium iron garnet (YIG) exhibit the narrowest ferromagnetic resonance linewidth of any known material. Highly polished single crystals of highest purity material exhibit linewidths of less than 0.3 oersted. Such narrow linewidths are indicative of very small damping of the spin system,[9] or exceptionally long relaxation times $(T = 2/\gamma\Delta H)$.

Because of its narrow linewidth the material itself can be used as a high-Q resonator with unloaded Q's of the order of 3000 at frequencies ranging from 5 Gc to greater than 70 Gc.[10]

At microwave frequencies YIG crystals have been used as low loss, tunable band-pass filters[11] where the center frequency of the device is tuned by varying the applied field. These crystals have been used as gyromagnetic couplers,[12] limiters,[13] and in a wide variety of similarly operating devices. The operation of all these devices requires that the material be biased to ferromagnetic resonance; hence in the millimeter range very large applied magnetic fields would be necessary. Because of the unique properties of this material, however, it appears that in some instances the effort expended in obtaining the high fields might be justifiable.

The ability of the garnet crystals to act as high-Q resonators and to store RF energy for periods of time of the order of their relaxation time has been used as the basis of a pulsed millimeter wave generator. The operation of the pulsed generator has been reported by Elliott,

[1] C. L. Hogan, "The Ferromagnetic Faraday effect at microwave frequencies and its applications—the microwave gyrator," *Bell Sys. Tech. J.*, vol. 31, pp. 1–31; January, 1952.

[2] M. T. Weiss and F. A. Dunn, "A 5-mm resonance isolator," IRE Trans. on Microwave Theory and Techniques (*Correspondence*), vol. MTT-6, p. 331; July, 1958.

[3] E. H. Turner, "A fast ferrite switch for use at 70 kMc," IRE Trans. on Microwave Theory and Techniques, vol. MTT-6, pp. 300–303; July, 1958.

[4] P. A. Rizzi, "Microwave properties and applications of ferrite rotators," *Microwave J.*, vol. 1, pp. 26–34; November and December, 1958.

[5] C. E. Barnes, "Broad-band isolators and variable attenuators for millimeter wavelengths," IRE Trans. on Microwave Theory and Techniques, vol. MTT-9, pp. 519–523; November, 1961.

[6] J. B. Thaxter and G. S. Heller, "Circulators at 70 and 140 kMc," Proc. IRE (*Correspondence*), vol. 48, pp. 110–111; January, 1960.

[7] W. P. Ayres, "Millimeter-wave generation experiment utilizing ferrites," IRE Trans. on Microwave Theory and Techniques, vol. MTT-7, pp. 62–65; January, 1959.

[8] A. S. Risley and I. Kaufman, "Efficient frequency doubling from ferrites at the 100-watt level," *J. Appl. Phys.*, vol. 33, pp. 1269–1270; March, 1962.

[9] R. C. Le Craw, R. C. Fletcher, and E. G. Spencer, "Electron spin relaxation in ferromagnetic insulators," *Phys. Rev.*, vol. 117, pp. 955–963; February, 1960.

[10] D. Douthett and I. Kaufman, "The unloaded Q af a YIG resonator from X-band to 4 millimeters," IRE Trans. on Microwave Theory and Techniques (*Correspondence*), vol. MTT-9, pp. 261–262; May, 1961.

[11] P. S. Carter, Jr., "Magnetically tunable microwave filters employing single crystal garnet resonators," 1960 IRE International Convention Record, pt. 3, pp. 130–135.

[12] R. W. DeGrasse, "Low-loss ferromagnetic coupling through single crystal garnets," *J. Appl. Phys.*, vol. 30, p. 155S–156S; April, 1959.

[13] J. Brown, "Ferrimagnetic limiters," *Microwave J.*, vol. 4, pp. 74–79; November, 1961.

Schaug-Pettersen, and Shaw[14] who obtained an output frequency of 32 Gc with an input frequency near 4 Gc. In this generator input energy is stored in the spin system and subsequently frequency converted through the action of an external pulsed magnetic field. The output frequency bears no integral relation to the input frequency in this generator, and it is continuously variable through adjustment of the magnitude of the pulsed field. It is important to the operation of this device that the field pulse have a rise time which is small compared to the relaxation time of the material. For this reason materials with the longest possible relaxation time are needed, and thus far YIG is the only satisfactory material.

Hexagonal Ferrites

To overcome the high field requirements of millimeter wave resonance devices, highly anisotropic materials seem to offer a promising solution. All magnetic materials exhibit a preferred direction of magnetization with respect to the crystallographic axes. This preference is phenomenologically described by the magnetocrystalline anisotropy energy. Since this anisotropy energy results in a torque on the magnetization, it is usually represented by an effective anisotropy field defined to have the magnitude and direction that would be required of an external field to produce the same torque on the magnetization.

The so-called hexagonal ferrites are a class of materials that exhibit a high degree of anisotropy. The most promising hexagonal ferrites for the millimeter range are those exhibiting uniaxial anisotropy where the preferred direction of magnetization is along the C axis of the crystal. In a polycrystalline sample in which the C axes of all individual crystallites are aligned along the same direction in space, the bulk medium has an effective macroscopic anisotropy field. This anisotropy field can be used in place of externally applied fields.

Through efforts at many laboratories uniaxial hexagonal ferrites have been developed to such a point that their anisotropy fields can now be controlled over wide ranges, and through careful preparation materials with suitably stable temperature characteristics can be obtained.

The first of the highly anisotropic uniaxial materials investigated by Ferroxdure[15] and Beljers[16] pointed out that this material had a large internal magnetic field which caused resonance to occur in the millimeter wavelength range, and it was subsequently demonstrated[17] that in the absence of an externally applied field resonance occurred in the vicinity of 50 Gc. Weiss and Dunn[2] constructed the first millimeter wave isolator using Ferroxdure. With applied fields of less than 4000 oersteds isolation ratios of the order of 20 to 1 were obtained in the vicinity of 55 Gc.

As a result of subsequent research efforts on hexagonal ferrites, materials are now available with anisotropy fields ranging from less than 6000 oersteds to greater than 50,000 oersteds. These materials should permit the realization of resonance devices from K band up through the 2-millimeter region. The most highly developed hexagonal ferrites can be grouped into two families according to their crystal structure and following the notation of the Philips workers[18] are referred to as the M and W compounds.

The M compounds are isostructural with Ferroxdure which has the chemical formula $BaO \cdot 6Fe_2O_3$. This compound has an effective anisotropy field of approximately 17,000 oersteds and a room temperature saturation magnetization of nearly 5000 gauss. If the barium in this compound is replaced by strontium the anisotropy field increases to 19,000 oersteds and the saturation magnetization remains essentially unchanged. Since the effective anisotropy field of uniaxial compounds is given by

$$H_{\text{anis}} = \frac{2K_1}{M_s},$$

where K_1 is the first-order anisotropy constant and M_s the magnetization, the anisotropy field can be controlled by varying either the anisotropy constant or the magnetization. It is found that by substituting nonmagnetic aluminum for iron in the M structure one can achieve a reduction in the saturation magnetization and a consequent increase in the effective anisotropy field of the material. In this manner the effective anisotropy field of the strontium compound can be increased from 19,000 oersteds to greater than 50,000 oersteds.[19] This variation of anisotropy field with aluminum substitution is shown in Fig. 2. It can be seen that the anisotropy field increases continuously with increasing aluminum content due to the decrease in magnetization. This decrease in magnetization is accompanied by a decrease in the Curie temperature of the material. Hence there is a practical limit to such aluminum substitution, since excessive substitution results in excessively low Curie temperature, and produces a material whose properties are highly temperature sensitive.

The W materials have the chemical formula, $BaO \cdot 2MeO \cdot 8Fe_2O_3$, where Me is some divalent transition element.

[14] B. J. Elliott, T. Schaug-Petttersen, and J. H. Shaw, "Pulsed millimeter-wave generation using ferrites," IRE Trans. on Microwave Theory and Techniques, vol. MTT-9, pp. 92–94; January, 1961.

[15] J. J. Went, G. W. Rathenau, E. W. Gorter, and E. W. Van Osterhaut, "Ferroxdure, a class of new permanent magnet materials," *Philips Tech. Rev.*, vol. 13, pp. 194–208; January, 1952.

[16] H. G. Beljers, "Faraday effect in magnetic materials with traveling and standing waves," *Philips Res. Repts.*, vol. 9, p. 131; 1954.

[17] M. T. Weiss and P. W. Anderson, "Ferromagnetic resonance in ferroxdure," *Phys. Rev.*, vol. 98, pp. 925–926; May, 1955.

[18] G. H. Jonker, H. P. J. Wijn, and P. B. Braun, "Ferroxplana hexagonal ferro-magnetic iron-oxide compounds for very high frequencies," *Philips Tech. Rev.*, vol. 18, pp. 145–154; November, 1956.

[19] D. J. DeBitteto, F. K. DuPré, and E. G. Brockman, "Highly anisotropic magnetic materials for millimeter wave applications," *Proc. Symp. on Millimeter Waves*, Polytechnic Inst. of Brooklyn, N. Y., pp. 95–108; 1959.

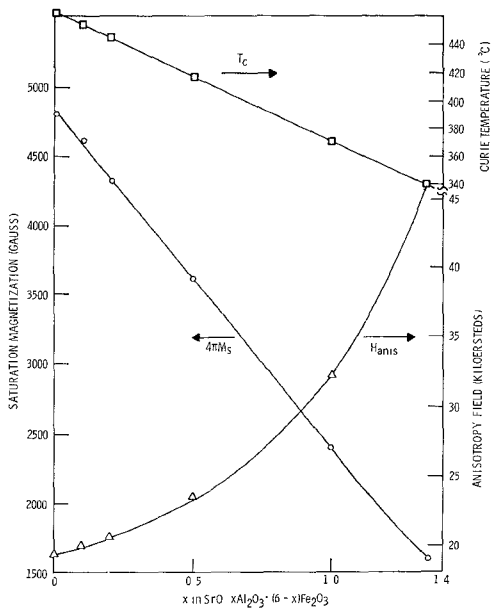

Fig. 2—Anisotropy field, saturation magnetization, and Curie temperature of substituted strontium M compound as a function of aluminum content.

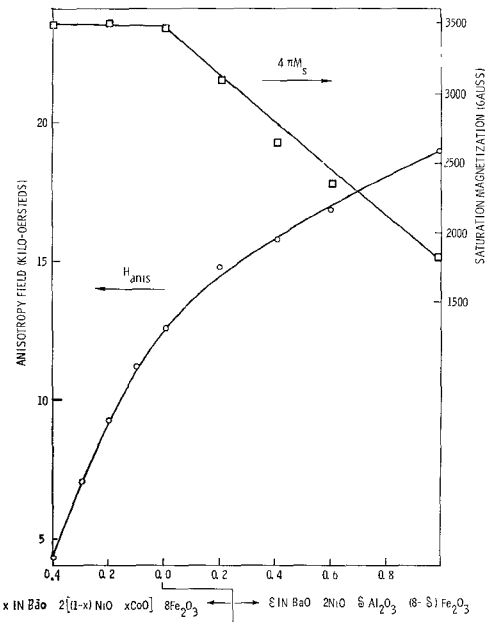

Fig. 3—Anisotropy field and saturation magnetization of substituted Ni_2W as a function of cobalt and aluminum content.

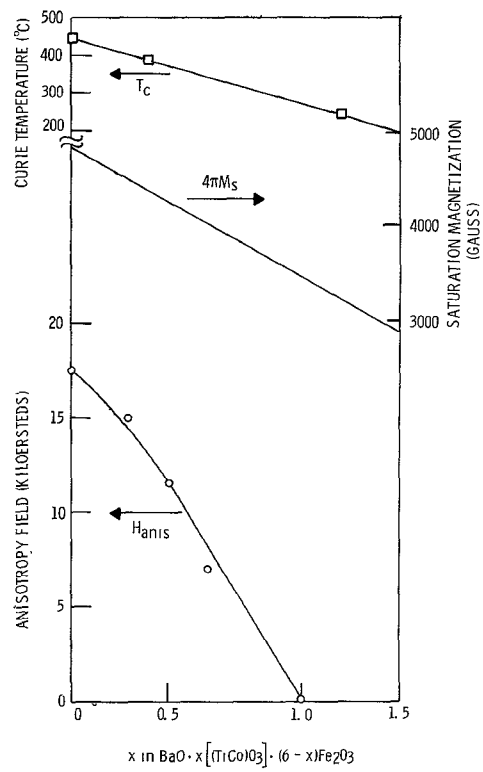

Fig. 4—Anisotropy field, saturation magnetization, and Curie temperature as a function of cobalt content in the substituted barium M compound.

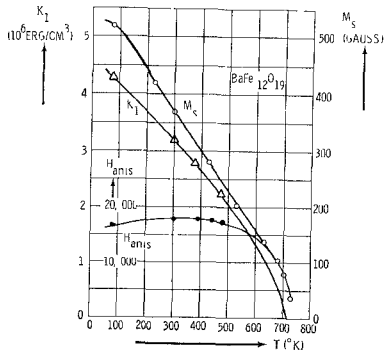

Fig. 5—Saturation magnetization, anisotropy constant, and anisotropy field of barium M compound as a function of temperature.

Ni_2W (Me = Ni) is a uniaxial material with a positive anisotropy constant; Co_2W has a negative anisotropy constant. By preparing solid solutions of Co_2W in Ni_2W it is possible to obtain uniaxial materials having anisotropy fields controllable over the range of 12,700 oersteds to 0 oersteds.[20] As in the M compounds, the substitution of aluminum for iron in W materials results in a reduction of $4\pi M_s$ and a consequent increase in anisotropy field. Fig. 3 shows the variation of anisotropy field and room temperature saturation magnetization with both cobalt and aluminum substitution in the W compounds.

The cobalt substitution can also be used to lower the anisotropy field of the M compound. In this case, however, since divalent cobalt is substituted for trivalent iron, a simultaneous substitution of quadrivalent titanium and cobalt must be carried out to preserve valence balance.[21] The variation of the anisotropy field, Curie temperature, and saturation magnetization of this material with titanium and cobalt substitution is shown in Fig. 4.

Since the center frequency of resonance devices using these materials varies directly with anisotropy field, this internal field must be independent of temperature over the range of operating temperatures of the device. Fig. 5 shows[22] that both the anisotropy constant K_1 and the magnetization M_s of the barium M compound decrease with increasing temperature. Both vanish at the Curie temperature. Since the effective anisotropy field is given by $2K_1/M_s$ its temperature dependence will be determined by the relative temperature variation of these two parameters. In the region from room temperature up to 150°C this material exhibits a fairly high degree of temperature stability, the anisotropy field varying by no more than a few hundred oersteds.

[20] G. P. Rodrigue, J. E. Pippin and M. E. Wallace, "Hexagonal ferrites for use at x- to v-band frequencies," *J. Appl. Phys.*, vol. 33, pp. 1366–1368; March, 1962.

[21] D. J. DeBitetto, F. K. DuPré, and W. Krautkopf, "Ferrorimagnetic resonance and anisotropy fields in A and TiCo substituted ferroxdure," *Bull. Am. Phys. Soc.*, vol. 7, Ser. 2, p. 54; January, 1962.

[22] J. Smit and H. P. J. Wijn, "Ferrites," John Wiley and Sons, Inc., New York, N. Y., p. 205; 1959.

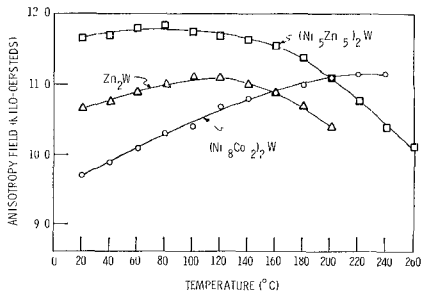

Fig. 6—Anisotropy field as a function of temperature for nickel zinc W, zinc W, and nickel cobalt W compounds.

Approximately 200°C below the Curie temperature, however, the anisotropy field begins to drop off rapidly with increasing temperature and for this reason Curie temperatures too near expected operating temperatures should be avoided in practical devices.

In the W compounds the substitution of zinc for nickel can be used with or without a simultaneous substitution of cobalt to achieve an added degree of temperature stability in the material. Fig. 6 shows the variation of anisotropy field with temperature in three different W compounds. It is seen here that over the temperature range from approximately 0°C to approximately 125°C the anisotropy field of these compounds can be made very stable. This is most notable in the $(Ni_{0.5}Zn_{0.5})_2W$ material.

While the hexagonal ferrites are generally associated with resonance devices in the millimeter range, they can also be used in Faraday rotation devices, as for example in Y circulators requiring no external magnetic field. In such applications it is essential that the material be prepared and operated in such a manner as to make the dielectric losses and magnetic losses negligible. Magnetic losses can be made negligible by operating at such frequencies that the effective anisotropy field biases the material well below resonance (at frequencies well above the natural resonant frequency). Dielectric losses can be minimized through careful preparation, and loss tangents

$$\left(\tan \delta_r = \frac{\epsilon_r''}{\epsilon_r'}\right)$$

of less than 0.001 have been obtained on these hexagonal ferrites.

Antiferromagnetic Materials

Antiferromagnetic materials[23] comprise another class of highly anisotropic materials that are applicable to resonance devices in the millimeter and submillimeter range. An antiferromagnetic material is essentially composed of two interpenetrating lattices of identical spins with adjacent spins aligned antiparallel. The net magnetization of the medium is zero, since each lattice com-

[23] See *e.g.*, B. Lax and K. J. Button, "Microwave Ferrites and Ferrimagnetics," McGraw-Hill Book Company, Inc., New York, N. Y., p. 253 ff; 1962.

pensates the magnetization of the other. Two important quantities which characterize these materials are the exchange field H_E, which causes the antiparallel alignment of spins, and the effective anisotropy field which causes the spin alignment to occur preferentially along certain crystallographic directions. As in other magnetic materials antiferromagnets exhibit a resonant absorption of electromagnetic radiation. The resonant frequency, when the material is under the influence of a dc applied field H_{app}, is given by the equation

$$\omega \pm = \gamma[\sqrt{2H_E H_{anis}} \mp H_{app}].$$

Thus the antiferromagnetic medium behaves as though it were biased by an internal field of $\sqrt{2H_E H_{anis}}$. The $\pm$ signs indicate two possible modes of resonance which are circular polarization sensitive, and thereby permit the fabrication of nonreciprocal devices.

Since H_E is usually of the order of 10^6 oersteds, and H_{anis} is of the order of 10^3 to 10^5 oersteds, the effective fields are quite large, and the resonant frequencies quite high.

The Lincoln Laboratory[24] group has constructed resonance isolators in the range of 150 to 200 Gc using Cr_2O_3, and the operating characteristic of such a device is shown in Fig. 7. The effective field of this material is approximately 60,000 oersteds.

Table I lists this and other antiferromagnetic compounds together with their Néel temperature, effective field, and zero field resonant wavelength. It is seen that these substances can be used well into the submillimeter range.

To observe antiferromagnetic resonance it is necessary to operate at a temperature well below the Néel temperature of the material. In Table I it is seen that this temperature is considerably below room temperature for most antiferromagnetic materials with the exception of those having extremely high exchange fields, *i.e.*, nickel oxide. Since the Néel temperature is a measure of the strength of the exchange coupling between spins, it is to be expected that only those materials with extremely large effective fields will have Néel temperatures above room temperature. Thus antiferromagnetic materials suitable for application to the millimeter range must be operated at 77°K or below.

Exchange resonances similar to those of antiferromagnets occur in all ferrimagnetic materials which in essence are composed of two or more nonidentical interpenetrating lattices of antiparallel spins. Studies of exchange resonances in rare earth iron garnets[25] indicate resonances occurring in the far infrared (10^2 microns).

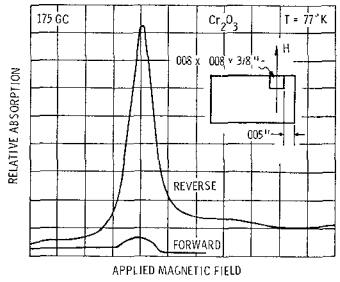

Fig. 7—Typical characteristics of nonreciprocal resonant absorption in single crystal of Cr_2O_3. Ratio of about 20 to 1 obtained with applied field of 3300 oersteds. (After Heller,[24])

TABLE I
ANTIFERROMAGNETIC RESONANCE DATA

Material	T_N(°K)	H_{eff} (oe)	λ_{res} ($H_{app}=0$, $T=0$°K)
$MnTiO_3$	65	$\sim 5.2\ 10^4$	≤ 2 mm
Cr_2O_3	307	5.9×10^4	1.9 mm
MnF_2	67.7	9.4×10^4	1.3 mm
MnO	120	3×10^5	364 μ
NiO	523	4×10^5	274 μ
FeF_2	78.4	5×10^5	190 μ

CONCLUSION

Based on present knowledge of the properties of magnetic devices it appears that several of the magnetic materials will prove useful at millimeter and submillimeter wavelengths.

In the millimeter range isotropic ferrites are even today widely used in Faraday rotation devices, while the hexagonal ferrites are being increasingly developed for application to resonance devices in the range of 10 millimeters to 1 millimeter.

At the still shorter submillimeter wavelengths the exchange resonances of antiferromagnetic and ferrimagnetic materials might be put to practical use at the expense of low temperature operation.

ACKNOWLEDGMENT

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[24] G. S. Heller, J. J. Stickler and J. B. Thaxter, "Antiferromagnetic materials for millimeter and submillimeter devices," *J. Appl. Phys.*, vol. 32, pp. 307S–312S; March, 1962.

[25] M. Tinkham, "Far infrared spectra of magnetic materials," *J. Appl. Phys.*, vol. 33, pp. 1248–1253; March, 1962.