Ferromagnetic Resonance in Manganese Ferrite Single Crystals*

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The microwave properties of two types of manganese ferrite single crystals have been investigated by means of the ferromagnetic resonance phenomenon from 300°K to 4.2°K and at 24 000, 9100, 5600, and 2800 Mc/sec. The low resistivity of one of the crystals, believed to be related to the presence of divalent iron, led to significantly different microwave behavior. Resonance lines as narrow as 47 oe have been obtained; however the width depends markedly on crystal direction, temperature and microwave frequency. The g-values are close to 2.00 under all conditions. The anisotropy, although frequency insensitive, increases rapidly at low temperatures. The Kramers-Krönig relations between absorption and dispersion are satisfied, and the line shape is roughly Lorentzian. Double resonances have been observed, and it is speculated that this may be a general phenomenon in single-crystal ferrites under suitable conditions.

I. INTRODUCTION

THE ferromagnetic resonance phenomenon in ferrites can be described satisfactorily in terms of the solution of the equation of motion for the precessing magnetization vector. The phenomenological description requires use of the appropriate effective magnetic field acting on the magnetization and of relaxation parameters which determine the line width. The sources contributing to the effective field can be accounted for theoretically, but the origin of the relaxation is not understood at the present time. To this end, it would be of value to have information on the frequency, crystal direction, and temperature dependence of the line width, as well as dependence on ferrite composition.

It is the purpose of this paper to furnish a comprehensive set of data on two types of manganese ferrite single crystals, including magnetic anisotropy, g-value, line width, and their frequency and temperature variations, and to point out new features in the microwave behavior. Earlier a number of microwave experiments on ferrite single crystals have been reported[1–5]; in general the objectives and results were of more limited scope or different emphasis.

In the following sections we describe methods and results of measurements made from room temperature down to 4.2°K and at frequencies from 24 000 Mc/sec to 2800 Mc/sec. Two types of single crystals were used, whose essential difference depended on their respective resistivities. This will be shown to have an important consequence on the line width and its temperature dependence. Finally, an additional resonance, which we believe occurs when the magnetization vector is not lined up with the applied magnetic field, was observed at low temperatures.

* The research reported in this document was jointly supported by the Army, Navy, and Air Force under contract with Massachusetts Institute of Technology.

[1] L. R. Bickford, Phys. Rev. **78**, 449 (1950).
[2] Yager, Galt, Merritt, and Woods, Phys. Rev. **80**, 744 (1950).
[3] D. Healy, Phys. Rev. **86**, 1009 (1952).
[4] Okamura, Kojima, and Torizuka, Science Repts. Research Inst. Tohoku Univ. Ser. A, **4**, No. 1 (1952).
[5] T. Okamura and Y. Kojima, Phys. Rev. **86**, 1040 (1952).

II. EXPERIMENTAL APPARATUS AND TECHNIQUES

The essential components of the basic microwave setup are shown in Fig. 1. Power from a frequency-modulated klystron is sent into a cavity which is placed between the poles of an electromagnet. Both the incident and reflected signals from the cavity are monitored by a pair of directional couplers and crystals. The direct and reflected signals are alternately selected by an electric switch and are viewed simultaneously on an oscilloscope. This permits Q-measurements to be made by comparing incident and reflected powers from the cavity. Sufficient attenuation is inserted into the direct signal arm to balance the reflected power from the cavity. This attenuation is a measure of the reflection coefficient which, in turn, is related to the VSWR and Q's of the cavity.[6] Standard microwave techniques were employed for measuring band widths of cavity resonances and frequency shifts of cavity resonances due to ferrite dispersion. With a stabilized microwave source providing a heterodyning signal, frequency changes of 0.02 Mc/sec could be detected. The power reflected from the cavity could also be recorded automatically. In this scheme the klystron is frequency-stabilized on the cavity, and the magnetic field is swept through its total range.

All X-band measurements were made in a cavity oscillating in the rectangular TE_{101} mode. The sample can be rotated on a dielectric post inserted through the end wall of the cavity. The K-band measurements were taken in a cylindrical cavity oscillating in the circular TE_{111} mode (the degeneracy was removed by a small loop of wire placed over the coupling iris in the E plane of the undesired mode), while the C- and S-band measurements were made with TE_{103} rectangular cavities.

Low-temperature measurements were carried out with the wave guide and cavity surrounded by a double Dewar system. The desired temperatures were obtained by filling the outer Dewar with dry ice and acetone, or

[6] For details see J. O. Artman and P. E. Tannenwald, Technical Report No. 70, Lincoln Laboratory, Massachusetts Institute of Technology, 1954 (unpublished).

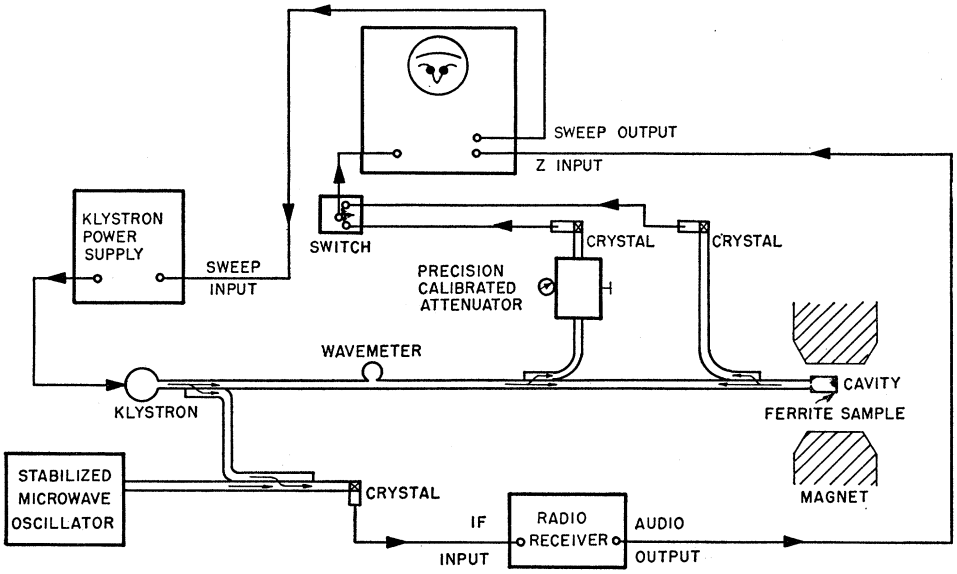

FIG. 1. Experimental setup used at room temperature.

with liquid nitrogen; for the 4.2°K work the cavity and sample were immersed in liquid helium.

The samples employed in this experiment were cut from single crystals of nominal manganese ferrite.[7] One crystal (Mn-Fe ferrite) contained a small (but unknown) amount of ferrous ion as evidenced by the resistivity of the order of 10 ohm-cm;[8] the second contained a small quantity of zinc and had a resistivity of about 10^4 ohm-cm; the third was manganese ferrite of 10^5 ohm-cm resistivity. By tumbling small pieces in an air stream against abrasive material, spheres were ground which ranged in diameter from 0.2 to 0.8 mm. These spheres were oriented and mounted[9] on the sample post by an x-ray back reflection technique in such a way that a crystal [110] axis was parallel to the axis of the post.

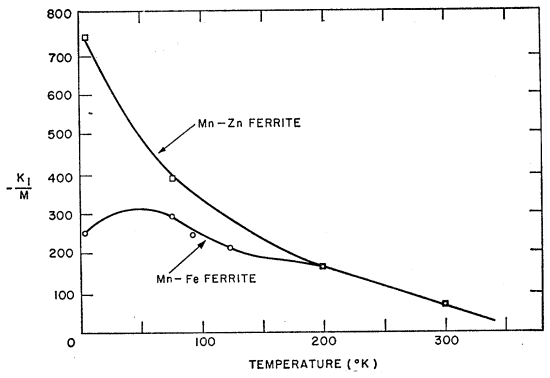

FIG. 2. Temperature variation of first-order anisotropy constant in two ferrites.

[7] Grown by G. W. Clark at Linde Air Products Company.

[8] J. H. Rowen, Bell Sys. **32**, 1333 (1953) has demonstrated the extreme sensitivity of the dc conductivity to iron excess or deficiency from stoichiometry.

[9] Kindly performed by D. Tuomi and E. P. Warekois of Lincoln Laboratory.

With this arrangement, the dc magnetic field (which was always perpendicular to the rf magnetic field) could be pointed along all crystallographic directions in the (110) plane.

III. RESULTS

Reduction of Data

The general resonance condition for a ferromagnetic material subject to an effective magnetic field H_{eff} and a microwave field of frequency ω is[10]

$$\omega=\gamma H_{\text{eff}}.$$

γ, the gyromagnetic ratio, is equal to $g(e/2mc)$. H_{eff} includes effects due to magnetic anisotropy and sample shape demagnetization and is given by

$$H_{\text{eff}}^2=\left[H_a+(N_x-N_z)M+\frac{K_1}{M}f_x(\theta)+\frac{K_2}{M}g_x(\theta)\right]\times\left[H_a+(N_y-N_z)M+\frac{K_1}{M}f_y(\theta)+\frac{K_2}{M}g_y(\theta)\right],$$

where H_a is the externally applied field, M is the saturation magnetization, $N_{x,y,z}$ are the geometrical demagnetizing factors, and $K_{1,2}$ are the first- and second-order anisotropy constants; $f_{x,y}$ and $g_{x,y}$ are functions of θ, the angle between the [100] axis and H_a, calculated by Bickford.[11]

If H_{eff} is eliminated between the two equations above, three unknown quantities will appear: γ (or g), K_1/M, and K_2/M. ω is the fixed operating frequency, and for spheres $N_x=N_y=N_z$. Experimentally, H_a necessary for

[10] C. Kittel, Phys. Rev. **73**, 155 (1948).

[11] L. R. Bickford, Technical Report No. 23, Laboratory for Insulation Research, Massachusetts Institute of Technology, 1949 (unpublished).

resonance is found for different θ's in the (110) plane. Three pairs of experimental quantities are substituted for θ and H_a, and the three equations are solved simultaneously. The three points chosen are the principal crystal axes $\theta=0°$, 55°, and 90° respectively. Experimental location of these points is facilitated because the applied field goes through an extremum at each.

At low microwave frequencies, resonance would occur before spherical samples could be magnetized; consequently at 2800 Mc/sec disk-shaped samples had to be employed. The arrangement in which the magnetic field is parallel to the plane of the disk is undesirable for two reasons. First, slight deviations from parallel surfaces seriously affect the resonance condition and half-width. Second, the internal dc field at which resonance occurs may not be large enough compared to the anisotropy to assure that the magnetization vector is lined up with the applied field.

The arrangement in which H_a is perpendicular to the plane of the disk is more satisfactory for line-width measurements. Clearly this precludes determination of K_1/M, K_2/M, and g; however, values for these parameters may be assumed from C-band results. If M is not known from independent measurements, it can

TABLE I. Ratio of first-order anisotropy constant to the saturation magnetization for Mn-Zn ferrite.

Frequency (Mc/sec)	K_1/M
5600	-73 ± 2
9100	-71 ± 1
24 000	-69 ± 1

be calculated on the basis of these assumptions. Subsequently then, all quantities are known to make the conversion from external to effective internal magnetic field.

Anisotropy

It must be kept in mind that even if K_2/M were as large as K_1/M, its contribution to the anisotropy energy would only be one-ninth that of K_1/M because of the usual normalization of anisotropy energy. Consequently we have attempted to calculate a second-order anisotropy from the data only when the anisotropy is large; even then K_2/M is not of much significance in the manganese ferrites. However, it should be stated that no combination of K_1/M, K_2/M, and g, gave a good fit to the angular variation of H_a. Furthermore, the frequency dependence of the anisotropy is very slight (or absent), and there is essentially no difference between Mn-Zn and Mn-ferrite. So if we are willing to limit ourselves to 10% accuracy, we can make the following simple statement: K_1/M, which is shown as a function of temperature for Mn-Fe ferrite and Mn-Zn ferrite in Fig. 2, is the same at all frequencies, and K_2/M is not significant.

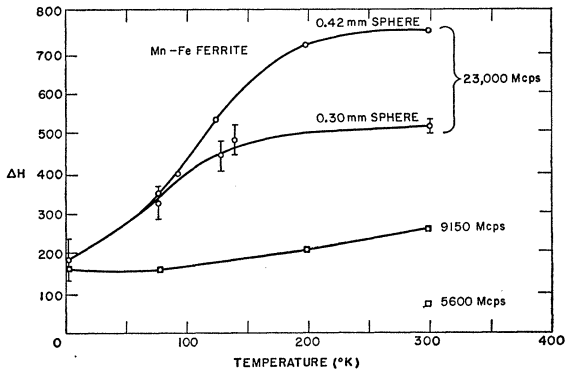

FIG. 3. Temperature dependence of the line width in Mn-Fe ferrite. No size-effect was apparent at 9150 Mc/sec.

Table I gives an example of the small variation of K_1/M with frequency. The largest second-order anisotropy encountered was of the order of $|K_2/M| \cong 2\times10^2$ oe.

g-Values

The value of g is constant at a value of 2.00 at all frequencies and temperatures measured in all three samples. Although occasional minor variations occurred, there was no consistent departure from 2.00. In most cases, g could be measured to an accuracy of ±0.01.

Line Width

The full widths at half-maximum susceptibility (ΔH) are shown plotted in Figs. 3 and 4 for Mn-Fe ferrite and Mn-Zn ferrite respectively. The graphs indicate both the temperature and frequency dependence of the line widths. Furthermore, for the Mn-Fe ferrite, which had the low resistivity, a size effect is apparent at 23 000 Mc/sec. However at 9150 Mc/sec the 0.30- and 0.42-mm spheres gave the same results. In the other samples, spheres ranging in diameter from 0.22 to 0.41 mm were used, where no size effect could be observed.

The Mn-Zn ferrite exhibits remarkably narrow lines at room temperature. The frequency dependence of ΔH is such that the narrowest lines occur at 5600 Mc/sec.

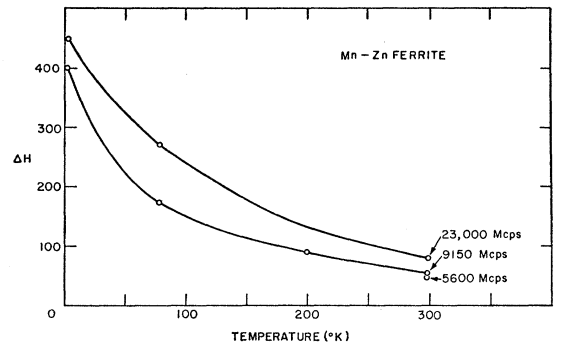

FIG. 4. Temperature dependence of the line width in Mn-Zn ferrite.

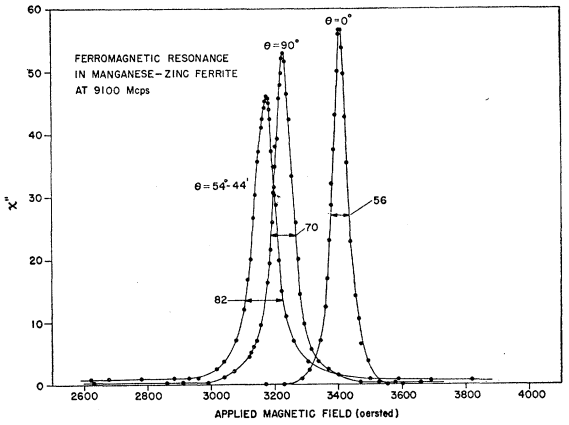

FIG. 5. Resonance absorption in the (110) plane, showing variations in line width.

With decreasing temperature, ΔH slowly increases and near 4.2°K shows a rapid broadening.

Another noteworthy feature is the directional dependence of the line width. The narrowest ΔH's occur in the [100] crystal directions and these are the ones that have been plotted in the graph. However, Fig. 5 shows the three resonance curves measured with the magnetic field applied in the three principal crystal axes. At other temperatures and frequencies the relative difference in line widths is approximately the same. Occasionally the [110] line width has been observed to fluctuate between the [100] and [111] extremes, but we attribute this to experimental uncertainties. Quite generally, the Mn-Zn and Mn ferrites gave identical results. Furthermore, data taken on Mn ferrite by Geschwind[12,13] at the Bell Telephone Laboratories give excellent agreement with the present results.

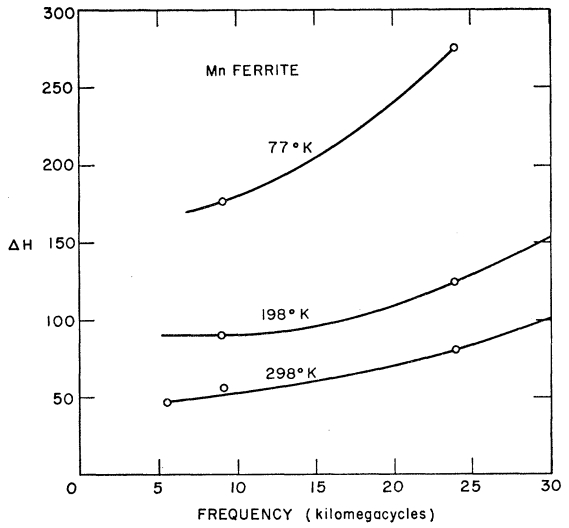

FIG. 6. Frequency variation of the line width in Mn-ferrite at different temperatures.

[12] S. Geschwind (private communication).

[13] Dillon, Geschwind, and Jaccarino, Phys. Rev. (to be published).

The frequency dependence of the Mn ferrite line width at three different temperatures is shown in Fig. 6. The qualitative behavior of ΔH seems to be approximately the same at the three temperatures. Data at 2800 Mc/sec have not been included as yet in the frequency dependence. We have made measurements on an 1/8-inch disk on Mn ferrite 6 mils thick with the magnetic field applied both parallel and perpendicular to the plane of the disk. The results indicate strongly that the line width tends to become broader than at 5600 Mc/sec. However, probably due to nonuniformities or cracks in the disk, the results are not consistent as yet. We hope to clarify this point by further measurements on other samples.

The behavior of Mn-Fe ferrite is completely different, as can be seen in Fig. 3. The lines are broad at room temperature and decrease in width with decreasing temperature. It appears to be a general rule that whenever a ferrite contains Fe^{++}, the temperature dependence of the line width is obscured, or even reversed, by the superposition of an additional loss mechanism.[14]

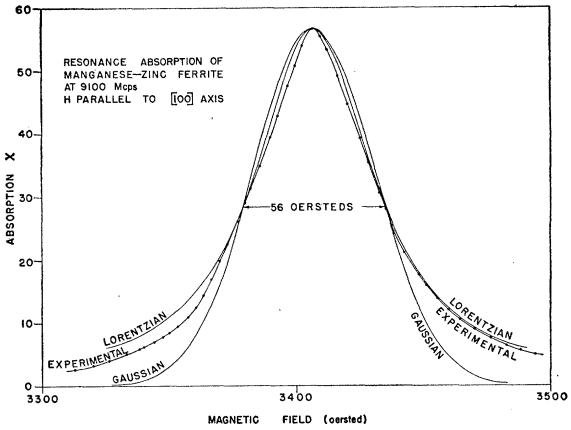

FIG. 7. Comparison of line shapes.

Line Shape

We have tried to fit a theoretical line shape expression to the experimental points measured on Mn-Zn ferrite at 9100 Mc/sec in the [100] direction. It can be seen from Fig. 7 that a Gaussian curve does not fit the data. However, a Lorentzian line gives closer agreement. The Lorentzian curve was expressed through a modification of the Bloch-Bloembergen relations[15] and gives for the absorptive component of the susceptibility:

$$\chi''=4\pi MHH_0\Delta H/[(H^2-H_0^2)^2+H_0^2(\Delta H)^2].$$

We have replaced the damping parameter $1/\gamma T_2$ by $\Delta H/2$, which is accurate to approximately 1% in

[14] Galt, Yager, and Merritt, Phys. Rev. **93**, 1119 (1954) believe that there exist losses in ferrites near the magnetite transition temperature due to a relaxation in short-range order among the divalent and trivalent iron ions. Our data do not show a maximum in the temperature dependence of ΔH.

[15] N. Bloembergen, Phys. Rev. **78**, 572 (1950).

the present case. The resonance field H_0 and the full width ΔH at half-maximum susceptibility were taken from the experimental data, namely $H_0=3407$ oe and $\Delta H=56$ oe. Furthermore, since the saturation magnetization is actually not known for this particular sample, the curves were also matched at their peaks.

Kramers-Krönig Relations

In one particular case, at 9100 Mc/sec on the Mn-Fe ferrite sample, we have made a precise measurement of the ferrite dispersion as well as absorption. According to the Bloch-Bloembergen expressions, the absorptive and dispersive components of the susceptibility are related by

$$\chi'=\chi''(H^2-H_0^2)/H_0\Delta H.$$

However, partly due to the Fe^{++} line broadening in Mn-Fe ferrite, the Bloch-Bloembergen expressions do not describe the data accurately. Therefore we have

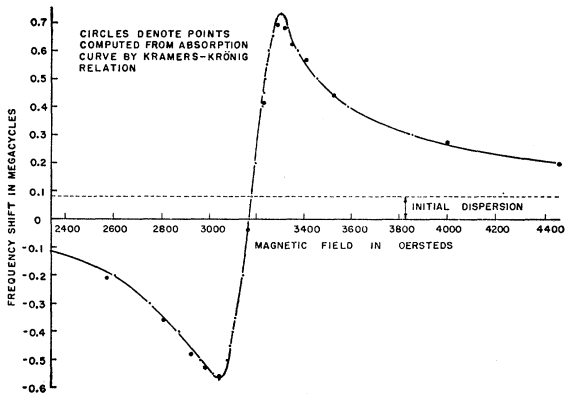

FIG. 8. Frequency shift of microwave cavity due to ferrite dispersion.

made a theoretical comparison between the absorption and dispersion data by way of the more general Kramers-Krönig relations.[16] The solid line in Fig. 8 is drawn through the solid points, which were measured from the frequency shift of the cavity. The heavy dots were computed from the corresponding absorption data. The absolute magnitude of the points has been adjusted arbitrarily. The fit can be regarded as satisfying the Kramers-Krönig relations.

Double Peaks

The behavior of all samples turned out to be rather remarkable at very low temperatures. At 4.2°K the samples of Mn-Zn ferrite and Mn ferrite showed double resonances in and near the hard crystal axes. In the case

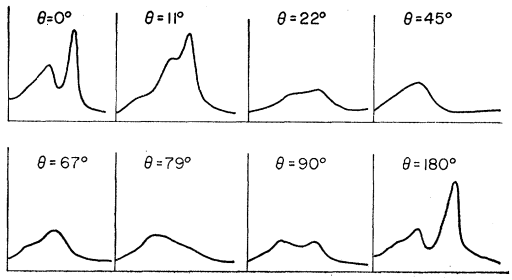

FIG. 9. Absorption as a function of magnetic field applied in various crystal directions in the (110) plane in Mn-Zn ferrite at 4.2°K and 9100 Mc/sec. $\theta=55°$ is the easy axis.

of the Mn-Fe ferrite, double peaks already occurred somewhere below 200°K.

Figure 9 shows the absorption as a function of magnetic field applied in various crystal directions. From measurements at 23 900 Mc/sec, where the usual single resonance is observed in all crystal directions, the anisotropy was found to be $K_1/M=-750$. It has been pointed out previously[17] that whenever the anisotropy becomes of the order of the applied magnetic field necessary for resonance, an additional resonance is possible in the nonaligned state of the magnetization vector. For spherical samples used here, the applied field necessary for resonance is the same as the resonance field in an infinite medium without anisotropy. In a thin disk, however, with the magnetic field applied along the plane of the disk, the external field need only be very small for gyromagnetic resonance and hence resonance in a nonaligned state becomes possible with relatively low anisotropy.[18] For the case of the magnetic field applied perpendicular to the plane of the disk, the situation is somewhat different.[19] In the present case the anisotropy is not sufficiently large to account for the double peaks directly. However, if we admit the possibility that some domain walls still exist, then the dynamic surface charges induced on the walls[20] may

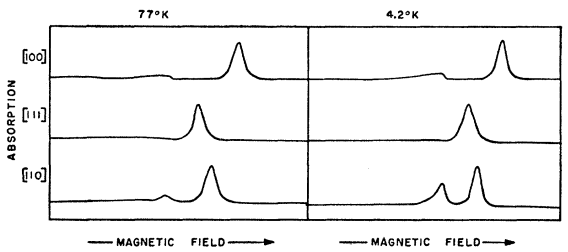

FIG. 10. Appearance of secondary peak at X-band.

[16] For a general discussion, see, e.g., J. H. Van Vleck, in *Propagation of Short Radio Waves*, edited by D. E. Kerr, *Radiation Laboratory Series* (McGraw-Hill Book Company, Inc., New York, 1951), Massachusetts Institute of Technology Radiation Laboratory Series, Vol. 13, Chap. 8.

[17] P. E. Tannenwald and H. J. Zeiger, Phys. Rev. **98**, 1562(A) (1955) and P. E. Tannenwald, Phys. Rev. **99**, 463 (1955).

[18] This is the condition under which A. F. Kip and R. D. Arnold, Phys. Rev. **75**, 1556 (1949), observed double peaks in a sheet of iron single crystal.

[19] See H. Suhl, Phys. Rev. **97**, 555 (1955).

[20] The effect of dynamic demagnetizing fields induced on 180° domain walls was proposed by D. Polder and J. Smit, Revs. Modern Phys. **25**, 89 (1953) and T. Nagamiya, Progr. Theoret. Phys. (Japan) **10**, 72 (1953). The latter used this model to explain Bickford's double peaks in magnetite at low temperatures.

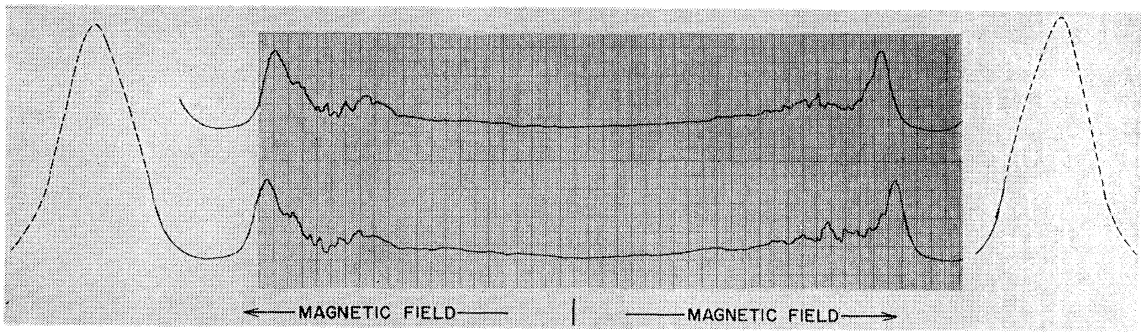

FIG. 11. Fine structure of secondary peak, showing extent to which it is reproducible upon field reversal and upon repetition. Sample is Mn-Fe ferrite single crystal, at 83°K, with field parallel to the [110] axis.

cause a sufficient modification of the simple double resonance condition. We plan to make the appropriate calculations for this type of situation.

The appearance of double peaks is somewhat different in Mn-Fe ferrite. Figure 10 shows the absorption at 77°K and 4.2°K in the three principal crystal axes. The second peak is absent at 23 000 Mc/sec.

Double peaks in ferromagnetic resonance absorption experiments have been reported as early as 1948 when Kip and Arnold showed that a second resonance peak in a sheet of iron single crystal was the result of incomplete alignment of the magnetization vector with the direction of the applied field.[21] Bickford[11] observed rather complex absorption spectra, including double peaks, in magnetite at low temperatures. It appears that double resonances are a quite general phenomenon in ferrites. The evidence from the present experiment suggesting failure of M to line up with H_a plus some remnant domain walls, is as follows:

(1) Double peaks do not occur in the easy crystal direction. (See Figs. 9 and 10.)

(2) Barkhausen-type jumps are observed on the initial rise of the low-field resonance. (See Fig. 11.)

(3) The location of the low-field peak in Mn-Fe ferrite varies with temperature in a similar fashion as the saturation magnetization. (See Fig. 12.)

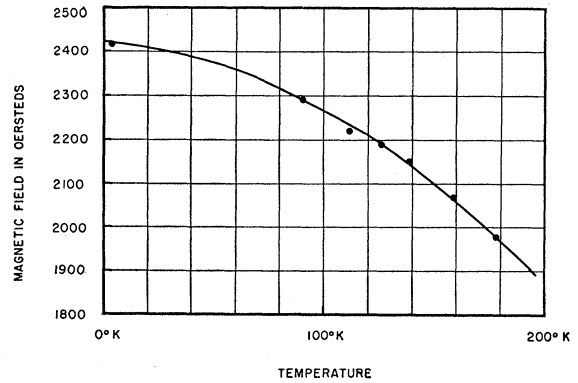

FIG. 12. Location of secondary peak with temperature in the [110] direction at X-band in Mn-Fe ferrite.

CONCLUSION

The most remarkable and at the same time theoretically least understood property of the single-crystal ferrites is the ferromagnetic line width. This investigation shows that lines as narrow as 47 oe can be obtained under suitable conditions of crystal direction, temperature, and microwave frequency in a manganese ferrite single crystal. Yet this is still an order of magnitude larger than can be explained on the basis of existing theories.[22,23] The line width increases at lower temperatures, in a fashion similar to Healy's results on nickel ferrite. The temperature dependence of the line width is reversed for a low-resistivity manganese ferrite, presumably because of an additional loss mechanism involving the divalent iron impurity. This decrease in line width with decreasing temperature is in the same direction as that found by Bickford in magnetite. In general the line width increases with frequency, but there is possibly a minimum in the vicinity of 5600 Mc/sec.

The g-values are close to 2.00 under all conditions. The anisotropy is insensitive to external magnetic field magnitude, but, as expected, increases sharply at lower temperatures. No change in sign was found in the first-order anisotropy constant, and the second-order constant is of relatively small significance. It is true on the other hand that a theoretical anisotropy fit to the experimental data using experimentally determined K_1/M, K_2/M, and g is less than satisfactory.

The Kramers-Krönig relations between absorption and dispersion are satisfied, and the line shape is roughly Lorentzian. It is interesting to note, however, that the areas under the resonance curves in the three principal crystal directions are not equal. (See Fig. 5.)

An increase in anisotropy as well as remnant domain walls are believed to be the basis of a mechanism resulting in secondary resonances, which can occur when

[21] A. F. Kip and R. D. Arnold, Technical Report No. 91, Research Laboratory of Electronics, Massachusetts Institute of Technology, 1948 (unpublished).

[22] C. Kittel and E. Abrahams, Revs. Modern Phys. **25**, 233 (1953).

[23] N. Bloembergen and S. Wang, Phys. Rev. **93**, 72 (1954).

the magnetization vector is not aligned with the applied magnetic field.

ACKNOWLEDGMENTS

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Thermal Conductivity of Indium-Thallium Alloys in the Normal and Superconducting States*†

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Thermal conductivity measurements have been made on three indium-thallium alloys between 0.15° and 0.8°K. Temperature gradients were measured using carbon resistance thermometers. The alloys were of the type which show an increase in thermal conductivity on entering the superconducting state. At temperatures below 1°K the superconducting state thermal conductivity decreases rapidly with decreasing temperature, becoming roughly proportional to T^3 and to the grain size in the specimens at the lowest temperatures. The data are interpreted as meaning that phonon conductivity limited by boundary scattering is important in this region.

I. INTRODUCTION

THE thermal conductivity of metals can be separated into two terms—an electron conductivity K_e and a lattice or phonon conductivity K_g. The total conductivity K is then given by the sum of K_e and K_g. Experimental and theoretical results show that in the liquid helium temperature region K_e may be represented by[1]

$$1/K_e=\rho_0/L_0T+\alpha T^2, \quad (1)$$

where ρ_0 is the residual electrical resistance, L_0 is the Lorenz number $\frac{1}{3}(\pi k/e)^2$, α is a constant which depends on the strength of the phonon-electron interaction, and T is the temperature. The first term on the right-hand side of Eq. (2) represents the contribution to the thermal resistance of scattering of electrons by impurities, and the second term is the resistance due to scattering of electrons by phonons. At a given temperature the relative importance of the two terms depends on the impurity content and on the Debye temperature. For impure metals and alloys at low temperatures impurity scattering is always dominant, but for pure metals with a low Debye temperature phonon scattering may be important. For pure metals, K_e is the order of a few watt units at 1 or 2°K, but this value is reduced by the presence of strains or impurity atoms, and is two or three orders of magnitude smaller for alloys.

In the same temperature region K_g is usually limited by scattering of the phonons by conduction electrons and is proportional to T^2. At lower temperatures the mean free path of the phonons would be comparable to the crystal dimensions and presumably would be determined by the scattering of phonons at crystal boundaries. In this case K_g should have the same temperature dependence as the lattice specific heat and should be given by Casimir's formula.[2] A dependence of K_g on T^3 has been found in superconductors below 1°K [3,4] and in dielectric crystals over a wider temperature range.[5] In most pure metals K_g is very much smaller than K_e, and phonon conductivity is important only in alloys and in metals having a very small number of conduction electrons.

If the additional subscripts s and n denote the superconducting and normal states, then

$$K_s=K_{es}+K_{gs} \quad (2a)$$

$$K_n=K_{en}+K_{gn}, \quad (2b)$$

where K_s and K_n are the total thermal conductivities of the same specimen in the two different states. K_n will be determined by the same factors considered above

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† Supported in part by a grant from the National Science Foundation.

‡ Present address: Department of Chemistry, University of California, Berkeley, California.

[1] For a review of thermal conductivities of metals at low temperature see: J. L. Olsen and H. M. Rosenberg, Advances in Phys. 2, 28 (1953).

[2] H. B. G. Casimir, Physica 5, 595 (1938).

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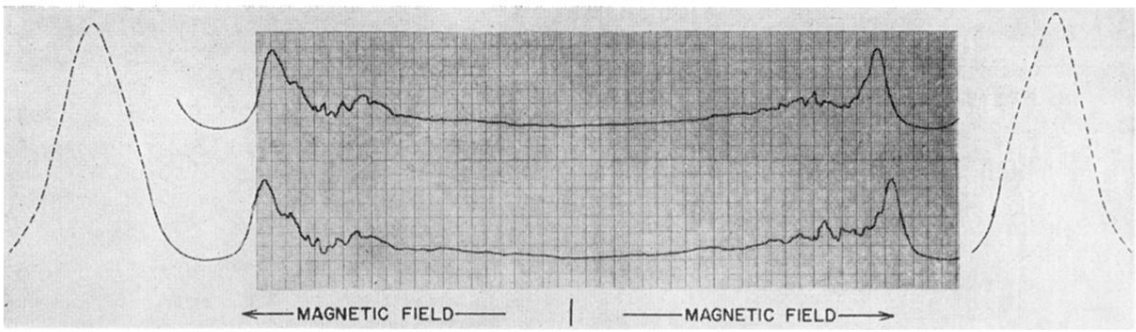

FIG. 11. Fine structure of secondary peak, showing extent to which it is reproducible upon field reversal and upon repetition. Sample is Mn-Fe ferrite single crystal, at 83°K, with field parallel to the [110] axis.