

Frequency and Temperature Dependence of the Ferrimagnetic Resonance Linewidth in Disordered Lithium Ferrite

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Experimental results are reported on the temperature (T) dependence and frequency (ν) dependence of the ferrimagnetic resonance linewidth (ΔH) in monocrystals of disordered lithium ferrite. The measurements were made at frequencies ranging from 5.7 to 40 kMc/sec and at temperatures ranging from 4.2 to 425°K. It was found that ΔH in a [100] direction is described by $\Delta H_{[100]} = B\nu e^{T/T_0}$ for $\nu > 12$ kMc/sec and $T > 200^\circ\text{K}$. The values of B and T_0 are 1.5×10^{-10} Oe-sec and 2.5×10^3 °K, respectively. Below 12 kMc/sec at room temperature $\Delta H_{[100]}$ departs from a linear dependence on ν , and below 9 kMc/sec $\Delta H_{[100]}$ increases as ν decreases. In all other crystallographic directions at room temperature the ν -dependence of ΔH was found to be similar to that of $\Delta H_{[100]}$. It was also found that ΔH is anisotropic with a maximum in the [111] direction and a minimum in the [100] direction. The anisotropic part ($\Delta H_{[111]} - \Delta H_{[100]}$) of the linewidth is independent of ν when $\nu > 12$ kMc/sec. ($\Delta H_{[111]} - \Delta H_{[100]}$) is nearly independent of T between 77°K and room temperature, but decreases at an increasing rate as T increases above room temperature. Neither the ν dependence of ΔH in any particular crystallographic direction nor the ν dependence of ($\Delta H_{[111]} - \Delta H_{[100]}$) is described by the simple two-magnon spin-wave scattering theories. A comparison of these ΔH data in disordered lithium ferrite with other ΔH data reported for ultrapure yttrium iron garnet reveals a striking similarity in both the ν dependence and the T dependence. It is also a rather striking fact that at room temperature and a given frequency $\Delta H_{[100]}$ in disordered lithium ferrite is only a factor of 10 greater than ΔH in ultrapure yttrium iron garnet. Some provisional conclusions suggested by the comparisons are discussed.

INTRODUCTION

IN monocrystals of "disordered" lithium ferrite, as well as of other disordered ferrimagnetic oxides with the spinel structure, the only mechanisms that have been definitely established experimentally for relaxation of the uniform mode are associated with the presence of ferrous ions¹ and poorly polished surfaces.² With the discovery of the degeneracy among certain spin waves and the uniform mode,³ it was thought that the problem of explaining the relaxation in disordered ferrites was reduced to merely finding a suitable scattering mechanism. In this connection considerable theoretical attention has been focused upon the disorder among the cations^{4,5} on the octahedral sites of these materials. The initial purpose of our research on lithium ferrite was to determine experimentally whether an effect of ionic ordering on the linewidth does exist in ferrimagnetic oxides. After resolving the unexpected effects associated with production of strain by sample preparation, an ordering effect on the linewidth was reported at the 1959 Conference on Magnetism and Magnetic Materials.⁶ It was reported that the linewidths at all angles and temperatures either increased or were relatively unchanged as a result of ionic ordering, in contradiction to the predictions of the spin-wave scattering theories. Thus, the effect on the linewidth of spin-wave scattering

due to ionic disorder was apparently smaller than other effects associated with the ionic rearrangement. However, at room temperature it was shown that the anisotropic part of the linewidth, present in the disordered state, disappeared upon ordering. This anisotropic part of the linewidth data presented at the 1959 Conference has been interpreted⁵ to be caused by spin-wave scattering associated with fluctuations in the quadratic axial term of the ferric ion spin Hamiltonian in the disordered state. This mechanism was suggested independently by Folen of this Laboratory and subsequently rejected as much too small to account for the observed linewidths.^{7,8} Thus, the sources of the linewidth in disordered lithium ferrite have remained a mystery, and it is apparent that more experimental results are needed to solve this problem.

SAMPLES AND METHODS

Measurements were made on spherical samples polished with "Linde A" powder. The samples were mounted in tunable transmission cavities incorporated in conventional ferrimagnetic resonance measurement systems.⁷ Data were taken at frequencies ranging from 5.7 kMc/sec to 40 kMc/sec and temperatures ranging from 77 to 425°K. All the monocrystalline spheres were heated to 950°C for about 10 min and then quenched in water to establish a disordered cation distribution. Some order-disorder and impurity ion effects in these crystals were investigated earlier.^{6,7,9,10}

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¹ W. A. Yager, J. K. Galt, and F. R. Merritt, *Phys. Rev.* **99**, 1203 (1955).

² R. C. LeCraw, E. G. Spencer, and C. S. Porter, *Phys. Rev.* **110**, 1311 (1958).

³ P. W. Anderson and H. Suhl, *Phys. Rev.* **100**, 1788 (1955).

⁴ A. M. Clogston, H. Suhl, L. R. Walker, and P. W. Anderson, *J. Phys. Chem. Solids* **1**, 129 (1956).

⁵ H. B. Callen and E. Pittelli, *Phys. Rev.* **119**, 1523 (1960).

⁶ A. D. Schnitzler, V. J. Folen, and G. T. Rado, *J. Appl. Phys.* **31**, 348S (1960).

⁷ A. D. Schnitzler, thesis submitted to the University of Maryland, January, 1961 (unpublished). Abstract published in *Dissertation Abstr.* **22**, (1962).

⁸ V. J. Folen, *J. Appl. Phys.* **33**, 1084S (1962).

⁹ V. J. Folen, *J. Appl. Phys.* **31**, 166S (1960).

¹⁰ A. D. Schnitzler, V. J. Folen, and G. T. Rado, *J. Appl. Phys.* **33**, 1293S (1962).

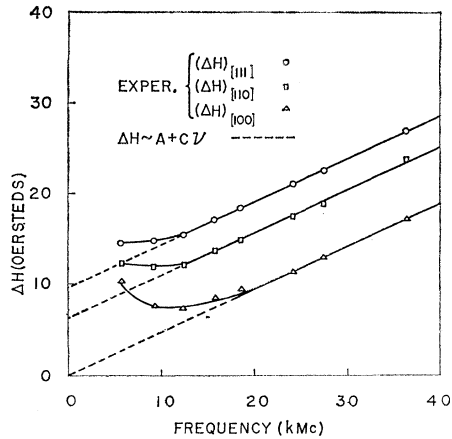


FIG. 1. Frequency dependence of ΔH in the three principal crystallographic directions at room temperature.

EXPERIMENTAL RESULTS

Figure 1 shows the dependence of the linewidth (ΔH) on the frequency (ν) in the three principal crystallographic directions at room temperature. Above approximately 12 kMc/sec the dependence of ΔH on ν is linear, and the rate of increase of ΔH with increasing ν is independent of crystallographic direction. The departure of ΔH from a linear dependence on ν below 12 kMc/sec is probably not due to a three-magnon process¹¹ because it is not supposed to occur in lithium ferrite (where $4\pi M_s = 3720$ Oe at room temperature) above 7 kMc/sec.

Figure 2 shows the dependence of ΔH on the temperature (T) in the three principal crystallographic directions at 27.8 kMc/sec. The magnitude of ΔH decreases as T decreases to 84°K. Data were not obtained below 84°K at 27.8 kMc/sec, but at 24.2 kMc/sec it was observed that in all crystallographic directions a peak in the dependence of ΔH on T exists in the temperature

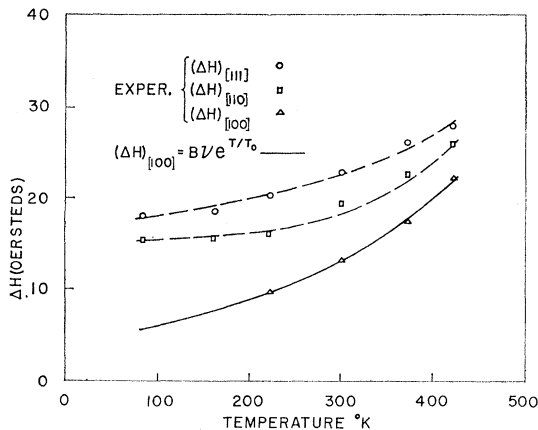


FIG. 2. Temperature dependence of ΔH in the three principal crystallographic directions at 27.8 kMc/sec. B and T_0 are given in the text.

¹¹ E. Schlömann, Phys. Rev. **121** 1312 (1961).

range 4.2 to 63°K. Below 200°K in the [100] direction it was not possible to make accurate measurements of ΔH at the 27.8 kMc/sec due to the appearance of multiple modes. Above 200°K it was found that if the logarithm of the linewidth in the [100] direction, $\log(\Delta H_{[100]})$, is plotted as ordinate and temperature as abscissa, the points fall on a straight line. The dependence of $\Delta H_{[100]}$ on T is, therefore, described by $\Delta H_{[100]} \propto e^{T/T_0}$, where T_0 is a constant given below. With this temperature dependence combined with the linear frequency dependence exhibited in Fig. 1, we may construct an empirical equation for $\Delta H_{[100]}$. Thus,

$$\Delta H_{[100]} = B\nu e^{T/T_0}, \quad (1)$$

where $B = 1.5 \times 10^{-10}$ Oe-sec and $T_0 = 2.5 \times 10^2$ °K were determined from the intercept and slope, respectively, of the graph of $\log(\Delta H_{[100]})$ vs T . It is shown in Fig. 2 that the graph of Eq. (1) fits the data satisfactorily. The validity of Eq. (1) is further demonstrated in Fig. 3

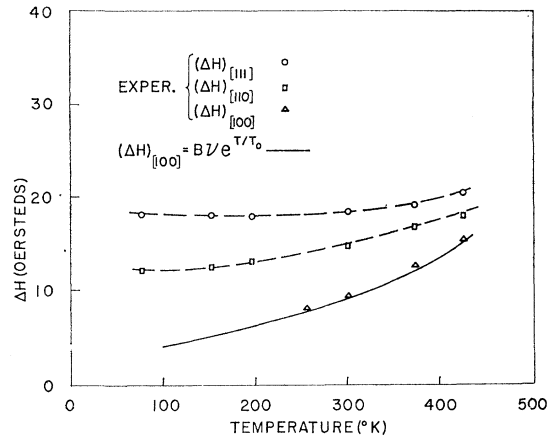


FIG. 3. Temperature dependence of ΔH in the three principal crystallographic directions at 18.7 kMc/sec. B and T_0 are independent of frequency.

where the graph of Eq. (1) and $\Delta H_{[100]}$ vs T are compared at 18.7 kMc/sec. It is interesting to note that the curves shown in Fig. 3 are not as steep as the corresponding curves shown in Fig. 2. This is a consequence of the linear dependence of ΔH on ν shown in Fig. 1. Again at 18.7 kMc/sec, as was found at 27.8 kMc/sec, it was not possible to obtain accurate measurements of $\Delta H_{[100]}$ below 200°K due to the appearance of multiple modes.

The temperature dependence of the anisotropy of the linewidth ($\Delta H_{[111]} - \Delta H_{[100]}$), apparent in Figs. 2 and 3, is shown more clearly in Fig. 4. The anisotropy of the linewidth is nearly independent of T between 77°K and room temperature, but decreases at an increasing rate as T increases to 425°K. Thus, the dependence of $(\Delta H_{[111]} - \Delta H_{[100]})$ on T is not simple, and it appears that data at values of T well above 425°K are needed to determine the explicit form of the dependence. It was

clear in Fig. 1 that at room temperature a dependence of $(\Delta H_{[111]} - \Delta H_{[100]})$ on ν does not exist in the linear range of ΔH vs ν . It is shown in Fig. 4, moreover, that this is also true at other values of T .

DISCUSSION

The theoretical curve in Fig. 5 shows the frequency dependence of ΔH according to the simple two-magnon spin-wave scattering theories^{4,5} which neglect the back reaction of the spin waves on the uniform precession. The frequency dependence of ΔH is similar in both of these theories, depending only on the number of spin-wave modes degenerate with the uniform precession and not on the details of whatever scattering mechanism is invoked. While the frequency dependence shown in Fig. 5 is based on a scattering potential with a "white" spectrum, it is generally true that the number of degenerate spin-wave modes of any particular wave number is a monotonically decreasing function of ν . The value of ΔH was shown in Fig. 1 to be a linearly increasing function of ν above 12 kMc/sec. The linewidth, therefore, cannot be attributed to simple two-magnon scattering. Furthermore, it is clear from Fig. 5 that the anisotropic part of ΔH also is not described by simple two-magnon scattering.

The linear dependence of ΔH on ν suggests a rapidly relaxing impurity ion mechanism. However, in contrast to our observations on ordered lithium ferrite,¹⁰ between 4.2°K and room temperature the g factor in the disordered state exhibits no temperature dependence, and the magnetocrystalline anisotropy is described solely by the first and second order constants, K_1 and K_2 . These observations indicate that the impurity ion effects on ΔH reported for these crystals in the ordered state are not being observed in the disordered state.

Two things may be noted about the ΔH data in the linear range of ΔH vs ν that was presented above. Firstly, the values of $\Delta H_{[100]}$ at room temperature and at a given frequency are only a factor of ten greater than for ultrapure yttrium iron garnet (YIG).¹² Secondly, both the ν dependence and T dependence of $\Delta H_{[100]}$ are similar in the two materials. The origin of ΔH in YIG has been attributed to certain intrinsic spin-spin and spin-lattice interactions.¹³ It seems possible that due to differences in magnitudes of the parameters involved the same interactions that give rise to ΔH in YIG may give rise to a larger (by a factor of 10) ΔH in disordered lithium ferrite.

Alternatively, the ΔH data presented above may indicate the joint relaxation (via the intrinsic interactions mentioned) of the uniform mode and a group of tightly coupled spin-wave modes.¹⁴ This situation may arise (due to the back reaction mentioned earlier) if the

¹² R. C. LeCraw and E. G. Spencer, International Conference on Magnetism and Crystallography, Kyoto, 1961.

¹³ T. Kasuya and R. C. LeCraw, Phys. Rev. Letters **6**, 223 (1961).

¹⁴ P. E. Seiden, J. Phys. Chem. Solids **17**, 259 (1961).

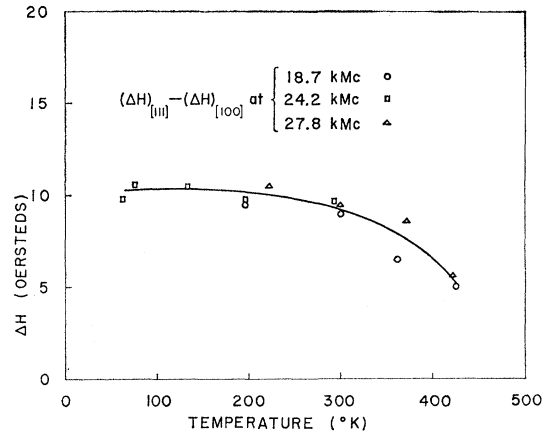


FIG. 4. Temperature dependence of the anisotropy of ΔH at three frequencies in the linear range of ΔH vs ν .

scattering potential is peaked at a particular set of wave numbers.¹⁵ It can be shown that if the relaxation rate of the uniform mode into each of a group of spin-wave modes is large compared to the relaxation rate of each spin-wave mode into other spin waves or into the lattice, then the back reaction must be considered. In this case the relaxation rate of the uniform mode depends linearly on the number of tightly coupled spin-wave modes and on the relaxation rate of the spin-wave modes themselves. Since the ν dependence of the number of spin-wave modes of any particular wave number resembles the theoretical curve shown in Fig. 5 and the ν dependence of the relaxation rate of the spin-wave modes themselves is thought to be linear^{12,13} (above the limiting ν of 7 kMc/sec for the three-magnon process in

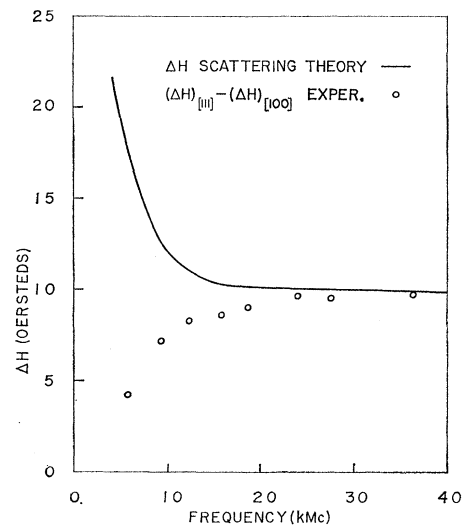


FIG. 5. Relative frequency dependence of ΔH due to spin-wave scattering from a potential with a "white" spectrum. Experimental values of the anisotropy of ΔH are given for comparison. Theoretical curve normalized to approach experimental data for large ν .

¹⁵ Herbert Callen, J. Appl. Phys. **32**, 738 (1961).

lithium ferrite), the above mechanism can account qualitatively for the entire ΔH vs ν curve. Moreover, the T dependence in this case can be the same as in YIG since in both cases it depends essentially on the intrinsic interactions of the spin waves. Whether this is a more nearly correct picture of the relaxation process than the first mechanism¹³ or, indeed, whether the degeneracy has anything at all to do with relaxation of

the uniform mode in disordered ferrites might be answered by making measurements of ΔH on carefully polished samples of different shapes.

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Hybrid Resonance and "Tilted-Orbit" Cyclotron Resonance in Bismuth

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Measurements have been made of microwave absorption in very pure, single-crystal bismuth as a function of static magnetic field, \mathbf{B} at 1.2°K and 70 kMc/sec. The field \mathbf{B} was in the plane of the sample along a crystallographic axis; the microwave electric field \mathbf{E} was either perpendicular or parallel to \mathbf{B} . Except at very low magnetic fields (≤ 100 G), nearly classical skin effect conditions prevail. Onsets of absorption are observed that result from singularities in the effective dielectric coefficient of the bismuth plasma. With \mathbf{E} perpendicular to \mathbf{B} most of the onsets do not correspond to any previously measured type of single-carrier cyclotron resonance in bismuth, but are hybrid resonances which require the presence of at least two distinct carriers. Such hybrid resonances are associated with longitudinal plasma modes. With \mathbf{E} parallel to \mathbf{B} , individual carrier cyclotron resonance is observed for those carriers whose cyclotron-orbit planes are tilted with respect to the magnetic field. Such tilts exist when \mathbf{B} is not parallel to one of the major axes of the electron energy ellipsoids. A close fit to the experimental data is obtained by a theoretical calculation based on electron and hole masses measured in other experiments.

I. INTRODUCTION

CYCLOTRON resonance experiments on metals have proven a powerful tool for determining the effective masses of carriers and their mass anisotropy. So far, two geometries have been utilized in such experiments: The Galt geometry¹ and the Azbel-Kaner geometry.² In the Galt geometry the static magnetic field is applied perpendicular to the surface of the sample, and absorption (or reflection) of circularly polarized radiation that is incident normally on the sample (i.e., along the magnetic field) is measured. By this method it is possible to determine both the effective mass of the carrier and the sign of its charge. This method was used extensively for bismuth under classical skin conditions.^{1,3}

In the Azbel-Kaner geometry the static magnetic field is applied in the plane of the sample and the

surface impedance is measured for normally incident radiation that is linearly polarized either parallel or perpendicular to the static magnetic field. Many experiments have been carried out under anomalous skin conditions; the surface resistance then exhibits oscillations with maxima at harmonics of the carrier frequency and yields the mass of the carriers but not the sign of the charge.

In this paper we report experiments on bismuth in an Azbel-Kaner type geometry but under nearly classical skin effect conditions. In the experiment the plasma frequency of the carriers is much larger than the radiation frequency and the microwave electric field \mathbf{E} was either perpendicular or parallel to the static magnetic field \mathbf{B} . As is shown in detail in Sec. II, when \mathbf{E} is perpendicular to \mathbf{B} and the mass of the carriers is isotropic, cyclotron resonance of individual carriers is shielded out by the large longitudinal depolarizing fields,⁴ which strongly couple the longitudinal and transverse degrees of freedom of the plasma and shift the natural frequencies of the plasma away from the cyclotron frequencies. When two or more carriers of distinct charge-to-mass ratios are present in a high-density plasma, all but one of the shifted frequencies become hybrid frequencies which involve the masses of all carriers. The remaining resonant frequency is near

¹ J. K. Galt, W. A. Yager, F. R. Merritt, B. B. Cetlin, and A. D. Brailsford, *Phys. Rev.* **114**, 1396 (1959).

² M. Ya. Azbel and E. A. Kaner, *Soviet Phys.—JETP* **3**, 772 (1956); as an experimental example, see A. F. Kip, D. N. Langenberg, and T. W. Moore, *Phys. Rev.* **124**, 359 (1961); J. E. Aubrey and R. G. Chambers, *J. Phys. Chem. Solids* **3**, 128 (1957).

³ Preliminary experiments already reported [J. K. Galt, F. R. Merritt, and P. H. Schmidt, *Phys. Rev. Letters* **6**, 458 (1961); J. K. Galt, F. R. Merritt, W. A. Yager, and H. W. Dail, Jr., *ibid.* **2**, 292 (1959)] and additional experiments under way [J. K. Galt (private communication)] indicate that the method is also applicable to metals in which anomalous skin effect conditions exist.

⁴ P. W. Anderson, *Phys. Rev.* **100**, 749 (1955).