

Magnetic Resonance Phenomena in Ferrites

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The resonant frequencies of the rotations of domain magnetization in ferrimagnetic materials are calculated by the use of the molecular field approximation, taking into account the effects of anisotropy, external field, and differences in magnetization and g values between the two sublattices. Resonance is found to occur in two frequency bands: one in a microwave band, which various workers have investigated experimentally, and one in the infrared, predicted earlier by Kaplan and Kittel. It is necessary to discuss critically the physical nature of the anisotropy field entering the equations; when this is done, an excellent agreement is found with the measurements of Yager *et al.* and Healy on single crystals of nickel ferrite. Finally, the relation between the theory presented here and experiments on sintered samples of ferrite is discussed.

INTRODUCTION

IN recent years several experiments have been performed on ferrites to find resonance phenomena analogous to those known to occur in ferromagnetic substances.¹⁻⁷ Two bands of resonance frequencies have been identified: a radio frequency band, probably corresponding to oscillations of the Bloch walls of the sample,⁴ and a microwave band, probably due to domain rotations.⁴ In addition, the theory of exchange resonance predicts a third frequency band in the infrared,⁸ but this has not yet been verified experimentally.

The theory of the Bloch wall resonance is beyond the scope of these remarks, but the microwave and the predicted infrared resonance should both be due to the collective interactions of spins within each domain, and should, therefore, both be explainable by the same physical considerations. Most of the experiments on the microwave resonance, however, were done before the theory of ferrimagnetic resonance was worked out in detail,⁸⁻¹² and so the theoretical interpretation of these phenomena was given at the time and has since been discussed in terms borrowed from the theory of ferromagnetic resonance.¹³ Essentially, it has been argued that the resonance is due to the motion of the net magnetization of the domain precessing in an aligning field consisting of an equivalent magnetocrystalline anisotropy

field, taken to be the same as the anisotropy field determined from torque measurements on a saturated crystal, plus any externally applied magnetic field. The object of this paper is to derive an expression for the resonant frequencies to be expected in ferrimagnetic materials, to discuss the role played by the equivalent anisotropy field, and to compare this theory with experiment.

THEORY

To derive the equations of motion, we suppose that the ferrite possesses two oppositely directed magnetic sublattices, of magnetization \mathbf{M}_1 and \mathbf{M}_2 . In the molecular field approximation, each lattice can be regarded as subjecting the other to an effective exchange field H_E proportional to its own magnetization: that acting on lattice 1 is

$$\mathbf{H}_{E1} = -\lambda\mathbf{M}_2, \quad (1a)$$

and similarly

$$\mathbf{H}_{E2} = -\lambda\mathbf{M}_1. \quad (1b)$$

We assume that \mathbf{M}_1 is directed along the positive z axis, which we take to be a direction of easy magnetization of the ferrimagnetic crystal. The sample is taken as spherical, so that demagnetization effects can be neglected. We assume further that there is an externally applied field H_0 , along the z axis, an anisotropy field H_{A1} , which tends to orient \mathbf{M}_1 in the positive z direction, and a field H_{A2} acting on \mathbf{M}_2 in the opposite sense. We neglect the effects of damping, but we assume that the splitting factor, g , is not necessarily the same for the two lattices.⁸ The equations of the motion are of the form

$$\partial\mathbf{M}/\partial t = \gamma\mathbf{M} \times \mathbf{H}.$$

Defining $M_+ = M_x + iM_y$, and similarly for other quantities, we find for the precession of lattice 1,

$$\dot{M}_{1+} = -i\gamma_1(H_{1z}M_{1+} - H_{1+}M_{1z}). \quad (2)$$

In this situation, H_+ acting on each lattice is due entirely to the exchange field H_E of the other lattice;

¹ J. L. Snoek, Philips Tech. Rev. 8, 353 (1948); Nature 160, 90 (1947); Physica 14, 207 (1948); H. G. Beljers and J. L. Snoek, Philips Tech. Rev. 11, 313 (1949).

² Yager, Galt, Merritt, and Wood, Phys. Rev. 80, 744 (1950).

³ C. Kittel, J. phys. radium 12, 292 (1951).

⁴ G. T. Rado, Revs. Modern Phys. 25, 81 (1953).

⁵ Wijn, Gevers, and van der Burgt, Revs. Modern Phys. 25, 91 (1953).

⁶ D. W. Healy, Jr., Phys. Rev. 86, 1009 (1952).

⁷ Okamura, Torizuka, and Kojima, Phys. Rev. 88, 1425 (1952).

⁸ J. Kaplan and C. Kittel, J. Chem. Phys. 21, 760 (1953).

⁹ F. Keffer and C. Kittel, Phys. Rev. 85, 329 (1952).

¹⁰ R. K. Wangsness, Phys. Rev. 86, 146 (1952).

¹¹ C. Kittel, Phys. Rev. 82, 565 (1951); T. Nagamiya, Progr. Theoret. Phys. (Japan) 6, 342 (1951).

¹² We follow Néel in using the term *ferrimagnetic* to describe phenomena characteristic of the magnetic structure of ferrites. Ferromagnetism and antiferromagnetism can both be regarded as special cases of ferrimagnetism.

¹³ C. Kittel, Phys. Rev. 73, 155 (1948).

in fact, from (1),

$$H_{1+}M_{1z} = H_{E2z}M_{2+}, \quad H_{2+}M_{2z} = H_{E1z}M_{1+}.$$

In the limit of small oscillations, $H_{E1z} = H_{E1}$ and $H_{E2z} = -H_{E2}$, so we shall drop the subscript z and write (2) as

$$\dot{M}_{1+} = -i\alpha_1\gamma[(H_0 + H_{A1} + H_{E1})M_{1+} + H_{E2}M_{2+}].$$

Similarly,

$$\dot{M}_{2+} = -i\alpha_2\gamma[(H_0 - H_{A2} - H_{E2})M_{2+} - H_{E1}M_{1+}], \quad (3)$$

where

$$\alpha_1 = \frac{1}{2}g_1, \quad \alpha_2 = \frac{1}{2}g_2, \quad \text{and} \quad \gamma = e/mc.$$

The frequency at which such a system will resonate if driven is the frequency of free oscillation ω_0 . Setting M_{1+} and M_{2+} each proportional to $e^{i\omega_0 t}$ and solving the resulting secular equation gives

$$\begin{aligned} -2\omega_0/\gamma = & (\alpha_1 + \alpha_2)H_0 + \alpha_1H_{A1} - \alpha_2H_{A2} + \alpha_1H_{E1} \\ & - \alpha_2H_{E2} \pm \{[(\alpha_1 - \alpha_2)H_0 + \alpha_1H_{A1} \\ & + \alpha_2H_{A2} + \alpha_1H_{E1} + \alpha_2H_{E2}]^2 \\ & - 4\alpha_1\alpha_2H_{E1}H_{E2}\}^{1/2}. \quad (4) \end{aligned}$$

In the special case of antiferromagnetism we have $\alpha_1 = \alpha_2 \sim 1$, $H_{A1} = H_{A2} = H_A$, and $H_{E1} = H_{E2} = H_E$. Equation (4) then reduces to $\omega_0/\gamma = H_0 \pm [H_A(H_A + 2H_E)]^{1/2}$, as given earlier.¹¹ In the ferromagnetic case, $\alpha_2 = H_{E1} = 0$ and the above resonance condition gives $\omega_0/\gamma = \alpha_1 \times (H_0 + H_A)$ in agreement with the theory of ferromagnetic resonance.¹³

In most of the applications in which one is interested, the square root in (4) can be expanded. Corresponding to the upper sign we get

$$\begin{aligned} -\omega_0/\gamma = & \alpha_1H_{E1} - \alpha_2H_{E2} \\ & + \frac{(\alpha_1^2H_{E1} - \alpha_2^2H_{E2})H_0 + (\alpha_1^2H_{E1}H_{A1} + \alpha_2^2H_{E2}H_{A2})}{\alpha_1H_{E1} - \alpha_2H_{E2}}, \quad (5) \end{aligned}$$

and to the lower,

$$\begin{aligned} -\frac{\omega_0}{\gamma} = & \frac{\alpha_1\alpha_2}{\alpha_2H_{E2} - \alpha_1H_{E1}} [(H_{E2} - H_{E1})H_0 \\ & + H_{A1}H_{E2} + H_{A2}H_{E1}], \quad (6) \end{aligned}$$

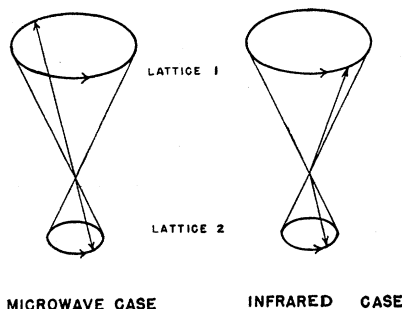


FIG. 1. Precessional motion of magnetic lattices in a ferrite for the two resonance cases treated. H_0 is parallel to axis of cones.

where in evaluating the root we have assumed that $\alpha_1H_{E1} < \alpha_2H_{E2}$ and that even the difference between these quantities is large compared to H_0 , H_{A1} , and H_{A2} .

Equation (5) is, except for the corrections due to anisotropy and external field, the same as the formula of Kaplan and Kittel⁸ for the infrared resonance. The resonant frequency (6), on the other hand, is of a different order of magnitude, falling in the range of microwave phenomena which we discussed earlier, for ω_0 is of the order of γH_0 or γH_A .

Let us now see what sort of lattice motions take place in these two ranges of frequencies. If we substitute (5) and (6) back into (3), we find that

$$M_{2+} \sim (\alpha_2/\alpha_1)M_{1+} \quad (\text{infrared}),$$

and

$$M_{2+} \sim -(H_{E1}/H_{E2})M_{1+} \quad (\text{microwave}).$$

These motions are respectively in phase and in opposition to each other and are illustrated in Fig. 1. Clearly, the microwave motion flexes the bond coupling the two lattices very little, in contrast to the infrared motion which flexes it to the greatest possible degree. This accounts for the large difference between the two frequency ranges and shows why the frequency of the microwave oscillation is roughly independent of the quantities H_{E1} which describe the bond.

DISCUSSION

We can compare the predictions of Eq. (6) with available experimental data. Before doing this, however, we must examine the relation of H_{A1} and H_{A2} , as they appear in the resonance expression, to the so-called equivalent anisotropy field, designated as H_{AO} , which has been measured for nickel ferrite from the hysteresis loops of single crystals¹⁴ and from the variation with crystal orientation of the external field necessary for ferrimagnetic resonance.^{2,6}

Ordinarily, H_{AO} is defined in terms of the torque required to cause a small angular deviation of the magnetization of a specimen away from a direction of easy magnetization. The torque is proportional to the anisotropy constant and to the angular deviation θ and is written as $H_{AO}M_s\theta$, where M_s , the saturation magnetization, is $M_1 - M_2$. But we can express it also in terms of H_{A1} and H_{A2} , each of which is assumed to act on the magnetization of the corresponding lattice so as to tend to align it with the axis of easy magnetization (that is, to orient M_1 in the positive z direction and M_2 in the negative one). In these terms, the torque is equal to $(M_1H_{A1} + M_2H_{A2})\theta$. Thus, we have

$$M_1H_{A1} + M_2H_{A2} = H_{AO}(M_1 - M_2). \quad (7)$$

The use of (1) then allows us to write (6) as

$$\omega_0 = \gamma\alpha_1\alpha_2 \frac{M_2 - M_1}{\alpha_2M_1 - \alpha_1M_2} (H_{AO} + H_0). \quad (8)$$

¹⁴ Galt, Matthias, and Reméika, Phys. Rev. **79**, 391 (1949).

We can now compare the predictions of (8) with that of resonance experiments on single crystals of nickel ferrite. To do this we shall take M_1 equal to 865 and M_2 to 600 cgs so that $M_s = 265$. Further $g(\text{Ni}^{2+}) = 2.20$ and $g(\text{Fe}^{3+}) = 2.00$, corresponding to a pure inverse spinel structure, which gives⁸ $\alpha_2 = 1.00$ and $\alpha_1 = 1.03$. We may evaluate H_{AO} from the expression for the crystal anisotropy energy density, for small angular deviations,

$$f = pk_1\theta^2, \quad (9)$$

where k_1 is the first-order anisotropy constant of the crystal. Equation (9) follows from a standard expression for the anisotropy energy if $p=1$ for positive k_1 and $p=-\frac{2}{3}$ for negative k_1 as in the present case of ferrites in which the (111) direction is an easy direction of magnetization. The torque as given by (9) is

$$T = \partial f / \partial \theta = (4/3)k_1\theta.$$

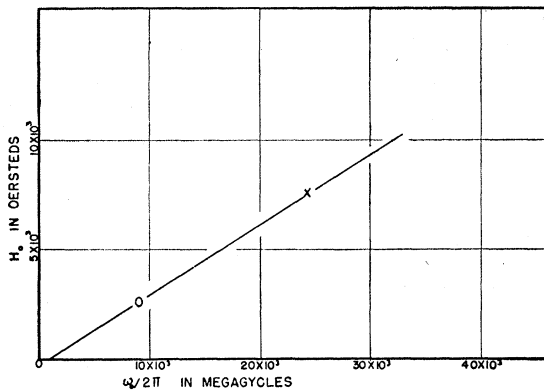


FIG. 2. Ferrimagnetic resonance conditions in single crystals of nickel ferrite. X—experimental data of Yager *et al.* (see reference 2); O—data of Healy; solid line is plot of present theory.

The anisotropy field H_{AO} is then equal to

$$H_{AO} = (4/3)(k_1/M_s). \quad (10)$$

If we insert the previously measured value² of $k_1 = -6.27 \times 10^4$ ergs/cm³, Eq. (10) gives $H_{AO} = 315$ oersteds. A plot of (8) is given in Fig. 2, together with the available experimental data. The agreement is excellent, considering the probable accuracy of the constants used and the apparent variability⁶ of the effective anisotropy constant from one crystal specimen to another. Further, the slope of the theoretical line in Fig. 2 represents a calculated g -value of 2.21, as compared with 2.25 and 2.19 as measured by Healy and Yager, respectively.

In addition to the data cited above for resonance in single crystals, we may compare the present results with the measurements of Okamura *et al.*⁷ on spheres cut from sintered nickel ferrite and manganese ferrite. Figure 3 shows this comparison, where H_{AO} in each case has been taken to give the best fit with the experi-

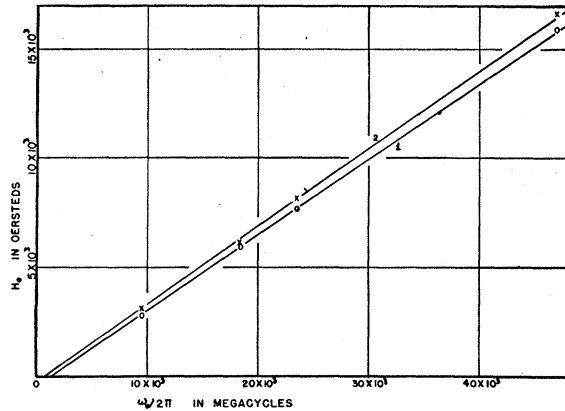


FIG. 3. Ferrimagnetic resonance conditions in sintered ferrites. X—experimental data of Okamura *et al.*⁷ in nickel ferrite; O—data in manganese ferrite (same author); present theoretical prediction in each case is represented by solid curve.

mental points. In the case of sintered nickel ferrite, a value of $\alpha_2 = 1.00$ has been used,¹⁵ corresponding to our choice of $g(\text{Fe}^{3+}) = 2.00$, and α_1 has been adjusted to give the correct experimental slope. With M_1 and M_2 assumed the same as for the single crystals, the data yield a g factor for nickel in sintered nickel ferrite of 2.07.

A similar comparison is possible for the data⁷ in sintered manganese ferrite. Again the experimental points are shown along with the theoretical curve computed for $H_{AO} = 278$ oersteds, as given by Okamura, under the assumption that $\alpha_1 = \alpha_2 = 1.00$, corresponding to observation,¹⁵ and that $g(\text{Fe}^{3+}) = g(\text{Mn}^{2+}) = 2.00$. (In this case the values of M_1 and M_2 do not enter the resonance condition.) The agreement is again satisfactory.

CONCLUSIONS

In view of the agreement with experiment of these ideas on the nature of the equivalent anisotropy field as applied to resonance in ferrites, it is of interest to speculate on the expected resonance frequencies in the closely related antiferromagnetic materials. From Eq. (7) it is found that H_{A1} and H_{A2} are appreciably smaller than H_{AO} . In fact, if we take $H_{A1} = H_{A2}$, it is found that $H_{A1} = 90$ for sintered nickel ferrite. If we assume that the anisotropy fields in antiferromagnetic materials are of this same order of magnitude, the resonance frequencies to be expected may be somewhat lower than those predicted using a value of $H_{AO} \sim 500$,¹¹ thus offering the possibility of observing antiferromagnetic resonance under less extreme experimental conditions than usually anticipated.

It may further be noticed that the internal field observed by Okamura⁷ of 507 oersteds for sintered

¹⁵ F. W. Lancaster and W. Gordy, *J. Chem. Phys.* **19**, 1181 (1951); L. R. Maxwell and T. R. McGuire, *Revs. Modern Phys.* **25**, 279 (1953).

nickel ferrite is significantly larger than that in the ferrite single crystals (Fig. 2), i.e., 315 oersteds. The difference may be attributed to the presence of internal strains and to demagnetization at the grain boundaries in the polycrystalline samples, the additional internal field of this origin being large enough to overcome the effects of the lower apparent magnetocrystalline ani-

sotropy fields which must result from the random orientation of crystallites away from directions of easy magnetization.¹⁶

¹⁶ After this work was completed an article by R. Wangness [Phys. Rev. **91**, 1085 (1953)] appeared in which is given a formula equivalent to (6) except that it does not contain the distinction between the phenomenological anisotropy field and that which enters the resonance expression.

The Change of Ferromagnetic Curie Points with Hydrostatic Pressure*

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The effect of pressure on the Curie temperature has been measured for 13 ferromagnetic materials, comprising 4 elements (Fe, Co, Ni, Gd), 7 metallic alloys, a ferrite, and a perovskite. Curie points ranged from 16°C (Gd) to 1120°C (Co), and pressures up to 9000 atmos. The results are in disagreement with two common forms of the interaction curve. The result obtained for Fe is not in agreement with the prediction of Kornetzki, based on volume magnetostriction measurements between 20°C and 100°C.

I. INTRODUCTION

Importance of Interatomic Distance

THE effect of pressure is considered to be that resulting from the change produced in interatomic distance, which is known to be an important parameter in the strength of magnetic interaction. Slater¹ first pointed out that ferromagnetism occurs only in those elements in which there is a large ratio of interatomic distance, D , to diameter of unfilled inner shell, d (usually a d shell, but in Gd it is the $4f$ shell). An accurate calculation of the magnetic interaction is still impossible at

the present stage of the theory, but the several theoretical approaches all agree on certain qualitative features of the interaction curve (i.e., the curve showing the strength of the interaction as a function of interatomic distance).

Sommerfeld and Bethe² discussed the type of wave function necessary to give a positive exchange integral in the Heisenberg theory, and they drew a curve (Fig. 1) to show qualitatively the features of this positive range. The band theory and Zener's theory both agree that ferromagnetism is not possible for very small or very large values of D , but the width of the ferromagnetic range is still an unsettled question. The salts of ferromagnetic elements, in which D is large, are paramagnetic rather than weakly ferromagnetic. On the other hand, Zener³ has pointed out the importance of conduction electrons in coupling the d -shell spins. The salts may be paramagnetic because of the absence of conduction electrons.

The Forrer Correlation

Forrer⁴ has made a correlation of magnetic interaction with interatomic distance, D , for Mn, Cr, Fe, Co, Ni and their compounds in which these elements retain their characteristic moments and in which coupling does not occur through intermediate atoms by the superexchange mechanism. He finds that as D increases the interaction changes, for each element, from anti-ferromagnetic to ferromagnetic and then to paramagnetic. The ferromagnetic range is roughly 10 percent

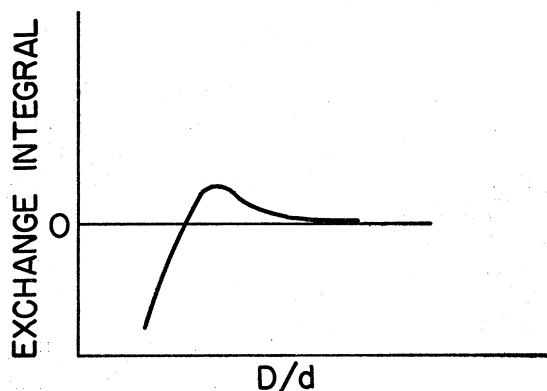


FIG. 1. Bethe interaction curve.

* Submitted in partial fulfillment of the requirements for the degree of Ph.D. at the University of Chicago. The work was done at the laboratory of the Institute for the Study of Metals, Chicago, Illinois.

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¹ J. C. Slater, Phys. Rev. **36**, 57 (1930).

² A. Sommerfeld and H. A. Bethe, *Handbuch der Physik* (J. Springer, Berlin, 1933), second edition, Vol. 24, No. 2, p. 596.

³ C. Zener, Phys. Rev. **81**, 440 (1951).

⁴ R. Forrer, Ann. Physik **7**, 605 (1952).