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# Magnetic Spectra of Ferrites

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A brief discussion is given of the meaning of initial permeability, the principles of its measurement, and the nature of "dimensional resonance." In considering magnetic spectra containing only a single resonance, typical data on sintered ferrites and on finely divided ferrite powders are reviewed, and some of the theoretical interpretations previously advanced are criticized. The investigations involving the two natural resonances found at this laboratory in a commercial Mg-ferrite are then summarized, including resonance in single-domain particles as a method for identifying dispersion mechanisms, experimental evidence for the inertia of domain walls, the nature of internal fields, and the temperature dependence of the natural resonances and of the spectroscopic splitting factor. Using the information obtained from these special studies, the following general hypothesis is advanced: In principle, two natural resonances are present in the magnetic spectrum of any sintered ferrite, and the single resonance sometimes observed represents the coalescence of two resonances. Reasons are given for expecting this coalescence to occur in some materials only, as in certain sintered mixed ferrites containing Zn-ferrite. The new hypothesis is consistent with all presently available data, including new spectra, some containing two resonances, that were recently obtained on pure ferrites at this laboratory. Additional confirmatory experiments are described which provide methods for separating or coalescing two resonances, and permit distinguishing between internal fields due to anisotropy and magnetic interactions. The limitations and possibilities of ferrites for obtaining useful microwave permeabilities are discussed from the new viewpoint, the criteria involved in this problem being shown to differ from those used at radiofrequencies.

## I. INTRODUCTION

It is the purpose of this paper to review the magnetic spectra of special ferrites and to propose a hypothesis, supported by new experiments, with regard to the magnetic spectrum of an arbitrary ferrite.

In order to avoid ambiguities, several definitions are stated below and adhered to throughout the paper. The term "magnetic spectrum" will denote the frequency dependence of the complex *initial* permeability, the frequency range of interest extending, in general, from zero to about $10^4$ megacycles per second (Mc/sec). For the sake of brevity, the word "ferrite" will include any ferromagnetic oxide, as well as the (ferromagnetic or nonferromagnetic) ferrites customarily defined by means of the formula $M\mathrm{Fe_2O_4}$, where $M$ represents a bivalent metallic ion. The expression "natural resonance" will refer to any resonance contained in the magnetic spectrum. This kind of resonance differs from ordinary ferromagnetic resonance, which is sometimes called "induced resonance," because the latter requires the application of a suitable static magnetic field to the material in question. Finally, the adjective "ferromagnetic" will include "ferr*i*magnetic" unless the need for this distinction is specifically noted.

Apart from their theoretical interest, magnetic spectra are important because it is usually the initial permeability that expresses the effect of the magnetic properties of a medium on the propagation of electromagnetic waves. In some particular situations, of course, this is not the case, and these situations will not be discussed here. However, it should be kept in mind that permeabilities other than the initial permeability play a role in the microwave Faraday effect,[1] or any other arrangement in which a static magnetization exists in a ferromagnetic material, as well as in experiments involving alternating fields possessing amplitudes sufficiently large to produce nonlinear effects.[2]

Although the magnetic properties of solids are, in general, most advantageously studied on monocyrstalline specimens, there are two reasons for investigating the magnetic spectra of polycrystalline ferrites. First, satisfactory monocrystals of ferrites are difficult to grow, and so far only a few such crystals of convenient size have been produced. It seems probable, in fact, that for some time to come the major applications of ferrites will involve sintered polycrystals. Second, the very perfection and regularity of monocrystals makes it difficult, as shown later on, to observe the important phenomenon of resonance due to domain walls, and to appreciate the strong influence of irregular internal demagnetizing fields on resonance due to domain rotations.

Accordingly, the present paper deals primarily with polycrystals,[3] and it represents a first attempt at ascertaining the factors that determine the types of natural resonance contained in the magnetic spectrum of an arbitrary ferrite. The discussion is confined to resonance frequencies and related line width problems, and no attempt is made to elucidate the nature of the basic absorption processes beyond the phenomenology usually employed.

## II. MEANING AND MEASUREMENT OF INITIAL PERMEABILITY

The initial permeability of a polycrystal represents an average value of a certain position-dependent quantity.

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[1] F. F. Roberts, J. phys. et radium **12**, 305 (1951); C. L. Hogan, Bell System Tech. J. **31**, 1 (1952).

[2] J. J. Went and H. P. J. Wijn, Phys. Rev. **82**, 269 (1951); H. P. J. Wijn and J. J. Went, Physica **17**, 976 (1951). See also comments in reference 18.

[3] For the magnetic spectrum of *mono*crystals of $\mathrm{Fe_3O_4}$ and $\mathrm{NiFe_2O_4}$ see, respectively, J. K. Galt, Phys. Rev. **85**, 664 (1952), and Galt, Matthias, and Remeika, Phys. Rev. **79**, 391 (1950).

If the magnetization proceeds solely by domain rotations, rather than by domain wall displacements, then this quantity is clearly a special case of the permeability tensor derived by Polder;[4] the effective field $H_e$, occurring in his analysis, is now due to anisotropy and internal magnetic fields. Whatever magnetization processes prevail in any given material, the average value mentioned here is only meaningful if the material is *macroscopically* isotropic and homogeneous. This condition obviously requires, in the case of alternating fields, that the variations in local properties of a sample be on a scale which is small compared to $\lambda_m$, the wavelength in the material. In the permeability measurements performed at this laboratory, the experimental method used prescribes that the thickness of each sample be much smaller than this wavelength, so that the requirement stated above was necessarily fulfilled.

On the basis of these remarks, the initial permeability of a macroscopically isotropic and homogeneous material, possessing no static magnetization, may be defined as the ratio

$$B/H=\mu=\mu_1-i\mu_2, \qquad (1)$$

where $B$ and $H$, representing the magnitudes of the parallel vectors $\mathbf{B}$ and $\mathbf{H}$, respectively, are proportional to exp $i\omega t$, and $H$ is supposed to be sufficiently small to insure $\mu$ being independent of $H$. Here $\omega$ is the circular frequency, and $H$ represents the field inside the specimen, rather than the applied field. The corresponding initial susceptibility $\chi$ is given by

$$M/H=(\mu-1)/4\pi=\chi=\chi_1-i\chi_2, \qquad (2)$$

where $M$ is the magnitude of the magnetization, $\mathbf{M}$.

In principle, the two components of $\mu$ are determined by measuring the reactive and resistive parts of the "impedance" (or related property) of a circuit containing the material in question. To determine $\mu$ uniquely, two sets of impedance measurements must be made unless the dielectric constant, $\epsilon=\epsilon_1-i\epsilon_2$, is known, or the magnetic material is located in a region of the electromagnetic field where the electric intensity is negligibly small. The latter condition is easily fulfilled at sufficiently low frequencies, and at these frequencies it is customary to obtain $\mu$ from the impedance of a coil wound around a ring-shaped core of the ferromagnetic material. If the conductivity of the core material is negligibly small, the well-known formulas

$$\mu_1=L_f/L_0 \qquad (3a)$$

and

$$\mu_2=\Delta R/\omega L_0 \qquad (3b)$$

apply in this simple case. Here $L_f$ and $L_0$ are, respectively, the inductance of the coil with and without the core, and $\Delta R$ is the additional resistance of the coil due to the presence of the core. Appropriate details on bridge circuits and resonance methods for measuring $L_f$, $L_0$, and $\Delta R$ may be found in summary articles.[5]

At sufficiently high radiofrequencies and in the microwave range, on the other hand, the reactive and resistive measurements involve, respectively, the imaginary and real parts of the input impedance of a transmission line, or the resonance frequency and quality factor of a cavity resonator. The input impedance can be determined by standing wave techniques. At this laboratory, measurements have been made over a large frequency range ($\sim$10 to $10^4$ Mc/sec) by a method[6,7] which uses a thin toroidal ferrite specimen of rectangular cross section, the specimen being inserted into a coaxial line at a position of large $H$ (and small $E$) adjacent to the short-circuited termination. By solving the appropriate boundary value problem, it can be shown that for *TEM*-type propagation

$$\mu_1=1+(s/L) \qquad (4a)$$

and

$$\mu_2=\left(\frac{1}{\xi}-\frac{1}{\xi'}\right)\frac{\lambda_0}{2\pi L}, \qquad (4b)$$

where $L$ is the thickness of the sample, $\lambda_0$ is the free-space wavelength, and $s$ is the displacement of the voltage minimum from its (previously observed) location in the shorted empty line. The quantities $\xi$ and $\xi'$ denote the voltage standing wave ratio of the actual line and the empty line, respectively. Equations (4) apply only if the relation

$$|\epsilon\mu|^{\frac{1}{2}}(2\pi L/\lambda_0)\ll 1 \qquad (5)$$

is satisfied, and if the coaxial conductors are highly conducting. These requirements mean, in effect, that the sample is thin compared to a quarter-wavelength in the material, and that the perturbation of the *TEM* mode is small. The "thin-sample method" outlined above, as well as the corresponding method[6] for measuring $\epsilon$, was originated by Halpern[8] in 1942, and the $\xi'$ correction was developed later.[7] In addition to the conceptual simplicity resulting from the "separation" of electric and magnetic effects, the thin-sample method possesses some practical advantages over certain frequently used "thin-sample methods" which require additional data and involve reference to transmission line charts or hyperbolic function tables. A general survey of permeabilities at microwave frequencies, including concepts, methods, and results pertaining to metals, was given by the writer.[9]

For many years it has been known that resonance effects are obtained if the relation $L=\lambda_m/4$ is satisfied in the type of set-up described above, and that these effects are of practical interest. Such a "dimensional resonance" could exist, of course, even if $\mu$ were independent of $\omega$ in the vicinity of the observed resonance,

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[4] D. Polder, Phil. Mag. **40**, 99 (1949).

[5] D. Polder, Proc. Inst. Elec. Engrs. (II) **97**, 246 (1950).

[6] Rado, Wright, and Emerson, Phys. Rev. **80**, 273 (1950).

[7] Rado, Wright, Emerson, and Terris, Phys. Rev. **88**, 909 (1952).

[8] O. Halpern and M. H. Johnson, unpublished work, M.I.T. Radiation Laboratory, 1942.

[9] G. T. Rado, *Advances in Electronics* (Academic Press, Inc., New York, 1950), Vol. II, p. 251.

and thus it should not be regarded as a natural resonance or any other purely magnetic phenomenon. Dimensional effects may be expected to occur in any arrangement where $L$ is comparable to the wavelength in the material. Brockman *et al.*[10] found, in fact, that dimensional resonances can exist at frequencies near the remarkably low value of 2 Mc/sec, the most surprising result of their experiments being the discovery of enormously large apparent dielectric constants in certain ferrites studied at these frequencies.

## III. SPECTRA CONTAINING A SINGLE RESONANCE

### A. Sintered Ferrites

In experiments on several sintered Ni-Zn and Mn-Zn ferrites, Snoek[11] observed a broad natural ferromagnetic resonance at radio frequencies. The typical data reproduced in Fig. 1 appear to be the most complete results published by him and his collaborators. Except for the broad peak in each $\mu_1$ curve, the resonance behavior looks almost like a relaxation because no values of $\mu_1<1$(i.e., $\chi_1<0$) were observed.

Snoek's explanation of the resonance is based on the idea, contained in the basic paper of Landau and Lifshitz,[12] that crystalline anisotropy is equivalent to an effective static magnetic field $H_i$, acting inside each crystallite, and that $H_i$ produces a Larmor precession of the appropriate electron spins. Since the rotation of the spins is subject to damping, it is apparent that resonance absorption occurs in any given crystallite if a small magnetic field, oscillating near the Larmor frequency, is applied to that crystallite along a direction differing from that of $H_i$. Using the simplifying assumptions mentioned below, Snoek found by means of an averaging calculation that the (circular) resonance frequency due to domain rotations, $\omega'_0$, is given by

$$\omega_0'=\gamma(M_s/\chi_0')(2/3), \tag{6}$$

for a polycrystal containing randomly oriented crystallites. Here $M_s$ is the saturation magnetization, $\chi_0'$ is the static initial susceptibility due to domain rotations, and $\gamma$, equal to $ge/2mc$, is the magnetomechanical ratio corresponding to the spectroscopic splitting factor, $g$. For an electron spin, the relevant numerical values are $\gamma/2\pi=2.80$ Mc/(sec oersted), and $g=2$. As pointed out elsewhere[6] (see also Sec. IV), $\chi_0'$ is not necessarily identical with $\chi_0$, the total static initial susceptibility. In deriving Eq. (6), Snoek neglected all magnetic interactions and attributed $H_i$ solely to crystalline (and later to stress-induced) anisotropy. The (first-order) anisotropy constant $K$ does not appear in Eq. (6) because the relations

$$H_i=2K/M_s \tag{7}$$

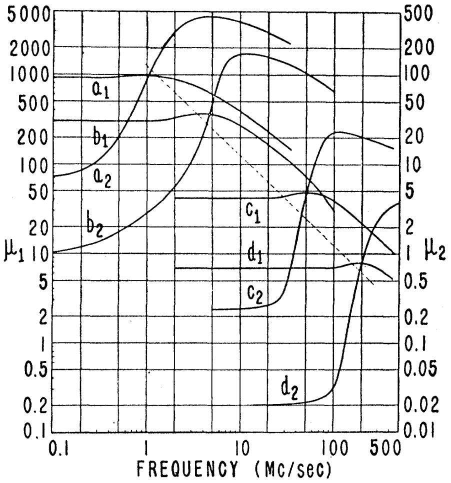

FIG. 1. Magnetic spectrum, at room temperature, of Mn–Zn ferrite (a) and of a number of Ni–Zn ferrites of different composition (b)–(d). The subscripts 1 and 2 for a given material (e.g., a) refer to $\mu_1$ and $\mu_2$, respectively. The dashed line indicates the mean relation between the low frequency value of $\mu_1$ (i.e., essentially $\mu_0$) of a certain ferrite and the critical frequency setting a limit to the range within which that ferrite can be used in filter coils $[\tan(\mu_2/\mu_1)\leq 0.06]$. (Reproduced, with minor alterations of the labeling, from the paper of H. G. Beljers and J. L. Snoek, Philips Tech. Rev. **11**, 313 (1949)).

and

$$\chi_0'=M_s^2/3K, \tag{8}$$

which apply if $K$ is positive, express $H_i$ in terms of $\chi_0'$. It should be noted that the right-hand sides of Eqs. (6) and (8) would have to be multiplied by $[1-(3^{\frac{1}{2}}/\pi)]$ if domain wall displacements were permitted in the theory, because in that case the magnetization vectors at the onset of domain rotations would not be oriented at random but in accordance with the distribution envisaged by Gans.[13] Using Snoek's model, and thus neglecting magnetic interactions, Kittel[14] extended the theory of domain rotations in a polycrystal by deriving an expression for the complex $\chi$. His result is

$$\frac{\chi}{\chi_0'}=\frac{\omega_0'^2+(\lambda/\chi_{00}')[i\omega+(\lambda/\chi_{00}')]}{\omega_0'^2+[i\omega+(\lambda/\chi_{00}')]^2}, \tag{9}$$

where $\chi_{00}'$ is an average rotational susceptibility approximately equal to $\chi_0'$ and $\lambda$ is the relaxation frequency (or damping factor) defined by the last term in the Landau-Lifshitz equation of motion

$$\frac{d\mathbf{M}_s}{dt}=\gamma\mathbf{M}_s\times\mathbf{H}_t-\lambda\left[\frac{(\mathbf{H}_t\cdot\mathbf{M}_s)\mathbf{M}_s}{M_s^2}-\mathbf{H}_t\right], \tag{10}$$

which describes the response of $\mathbf{M}_s$ to the total field $\mathbf{H}_t$. As noted previously,[6,7] it seems probable that Eq. (6), and equations similar to (9), cannot be used for reliably interpreting observed spectra (even when these are

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[10] Brockman, Dowling, and Steneck, Phys. Rev. **77**, 85 (1950).

[11] J. L. Snoek, Philips Tech. Rev. **8**, 353 (1946); Nature **160**, 90 (1947); Physica **14**, 207 (1948); H. G. Beljers and J. L. Snoek, Philips Tech. Rev. **11**, 313 (1949).

[12] L. Landau and E. Lifshitz, Physik. Z. Sowjetunion **8**, 153 (1935).

[13] R. Gans, Ann. Physik V **15**, 28 (1932).

[14] C. Kittel, J. phys. et radium **12**, 332 (1951).

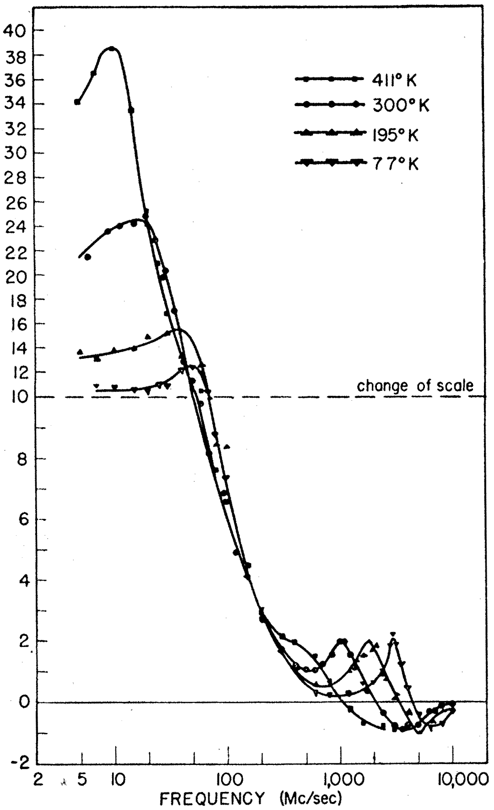

FIG. 2. $\mu_1 - 1$ *versus* frequency in the magnetic spectrum of solid ferramic *A* at various temperatures (see reference 7).

solely due to rotations), because the equations not only neglect the effect of magnetic interactions on the mean $\omega_0'$ but ascribe the over-all line width to damping only, i.e., they do not include the line width caused by a distribution in $\omega_0'$ values.

Snoek assumes that domain wall displacements do not contribute to the linear magnetization at radio frequencies. Although some of Snoek's data agree reasonably well with Eq. (6) and are, therefore, not incompatible with his assumption, it was shown elsewhere[6] (see also Sec. IV) that this approximate agreement does not prove the absence of wall displacements in his experiments or later[2] studies. As shown in Secs. IV and V, moreover, in some ferrites there are actually two resonances, and one of these is due to domain walls. In Sec. V it will be suggested that the apparently single resonance observed by Snoek is due to the presence of Zn-ferrite in his mixed ferrites, and it will be argued that this very presence, which is advantageous in the low radiofrequency region, leads to unsatisfactory magnetic properties at microwave frequencies.

### B. Powdered Ferrites

Shortly after Snoek's first paper appeared, Birks published a note on natural resonance at microwave frequencies in magnetite and gamma-ferric oxide. In these experiments, as in his later work on Ni-Zn ferrite and Mn-Zn ferrite, all of which he summarized in his last paper, Birks[15] used oxides that had not been sintered but consisted of fine powers embedded in wax. Unfortunately, the results given represent derived magnetic properties obtained by extrapolations of observed data on the concentration dependence of oxide-wax mixtures. As shown by unpublished experiments performed at this laboratory, the logarithmic relations used for these extrapolations, being adaptations of Lichtenecker's relations for dielectric mixtures, appear to be of rather limited validity. It seems doubtful, in fact, whether the permeability of a ferromagnetic solid can be obtained by any extrapolation whatsoever. Subsequent work (Sec. IV) has shown that the resonances in single-domain particles, at least, are fundamentally different from those in "solid" substances.

As to the mechanism of microwave magnetization, Birks ascribed it to domain rotations in a manner similar to that used by Snoek in his work on sintered ferrites at radio frequencies. In this connection, mention should be made of experiments at this laboratory which have shown that even in (commercial) mixed ferrites which contain Zn-ferrite, where the single observed dispersion region in sintered samples is at radio frequencies, the location of this region in fine powders is

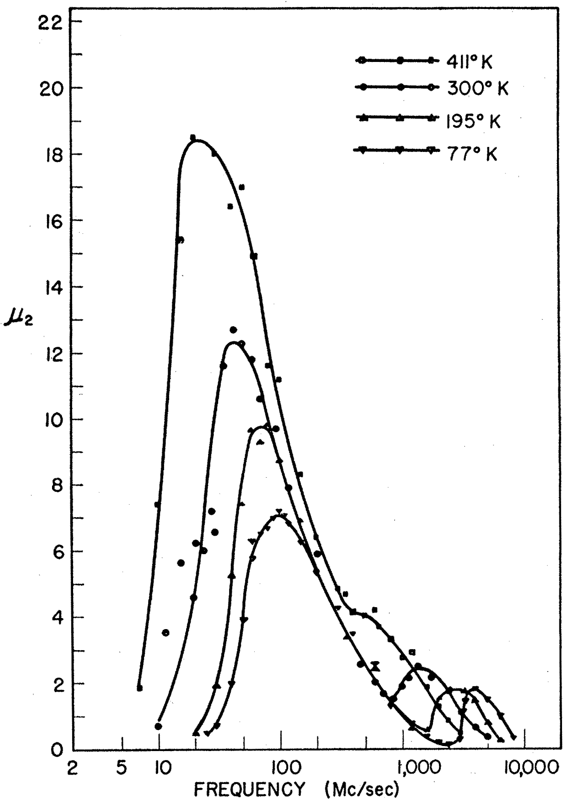

FIG. 3. $\mu_2$ *versus* frequency in the magnetic spectrum of solid ferramic *A* at various temperatures (see reference 7).

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[15] J. B. Birks, Proc. Phys. Soc. (London) **B63**, 65 (1950).

at microwave frequencies. In some cases this behavior may be due to the pronounced stiffness, and hence high resonance frequency, of domain walls in sufficiently small particles; more often, however, this behavior is probably due to the particles being sufficiently small to approach single-domain characteristics so that only rotations can occur (Sec. IV).

## IV. A SPECTRUM CONTAINING TWO RESONANCES

### A. Dispersion Mechanisms

Measurements performed at this laboratory on commercial ferrites of the "ferroxcube" and "ferramic" class have shown that in most of these materials the magnetic spectrum contains only a single natural resonance in the large frequency range investigated. It was particularly interesting to find, therefore, that the magnetic spectrum of "ferramic *A*," a sintered substance composed of several oxides[16] but consisting primarily of magnesium ferrite, contains two natural resonances at room temperature. These are shown by the curves representing the values, at 300°K, of $\mu_1-1$ (Fig. 2) and $\mu_2$ (Fig. 3).

Previous studies[17] conducted at this laboratory had demonstrated for the first time that domain rotations and wall displacements depend on frequency differently, and that at high frequencies ($\geq 200$ Mc/sec, or even lower) the $\mu$ of iron is primarily due to domain rotations and not, as at zero frequency, due to wall displacements. In view of these findings it seemed tempting to conjecture that the two resonances in ferramic *A* arise, respectively, from the two magnetization processes mentioned. To verify this surmise, it was necessary, as in the case of iron, to originate a method for identifying dispersion mechanisms experimentally. In the case of ferramic *A*, the identification was based on the following idea.[6] Domain wall motion is obviously suppressed if the walls are eliminated, and this can be achieved by dividing the material into sufficiently small particles to insure that they are essentially single domains; any resonance observed after this suppression must then necessarily be due to domain rotations, the only other kind of basic magnetization process that is possible if the conventional concept of domain structure is at all meaningful in the material (and at the temperature) in question. This idea proved practicable, and the details of the experiments were described elsewhere.[6,7] Briefly, it was shown that relatively large particles (~0.5 micron) of ferramic *A* exhibit essentially single-domain behavior, and that the measured spectrum of a mixture of these particles and wax clearly indicates (Fig. 4) the *absence* of the radiofrequency resonance (occurring at 43 Mc/sec in the sintered samples) as well as the *presence* of the microwave resonance, the latter occurring, however, at a somewhat higher frequency (2300 Mc/sec) than in the sintered samples (1400 Mc/sec). This result means that in ferramic *A* the rf resonance is due to domain walls, thus providing the first experimental support for the concept of domain wall inertia, and that the microwave resonance is due to domain rotations; these conclusions are not affected by the existence of impurities in the material. When combined with other evidence, the data show that in ferramic *A* the rotational resonance is due not only to crystalline anisotropy but also to internal demagnetizing fields,[6,7] that the effect of internal stresses is probably negligible[6,7] in both resonances, and that the damping is not due to microscopic eddy currents.[6] Furthermore, a measurement of the effective anisotropy[18] indicated, as expected, that the total static initial permeability $\mu_0$ is too small to be accounted for on the basis of rotations only. Finally, the above identification of the dispersion mechanisms is supported by the nature of the observed spectrum at remanence even though this particular argument cannot be regarded as being independent.[6] In this connection, it should be mentioned that the theoretical value of the relative remanence, $M_r/M_s$, might have been based on the "Gans" distribution," but for reasons which are, perhaps, connected with the occurrence of 180° wall displacements, the data seem to favor the distribution assumed in reference 6.

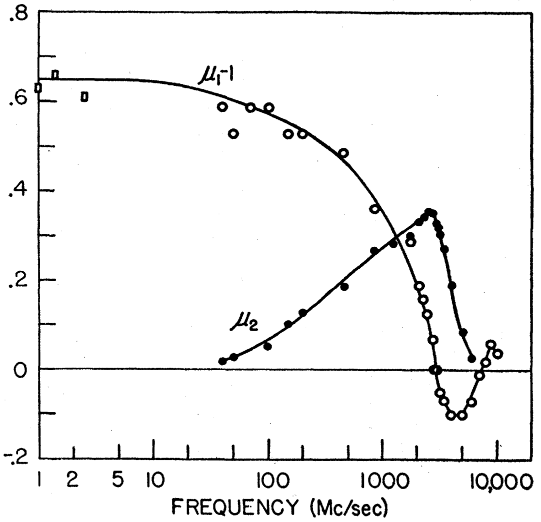

FIG. 4. Magnetic spectrum of 70 percent (by weight) mixture of ferramic *A* powder and wax at room temperature. The powder particles are essentially single-domains (see reference 6).

It was pointed out[6] in analyzing the data that, in general,

$$\chi=\chi'+\chi'', \tag{11}$$

where $\chi''$ is the initial susceptibility resulting from wall displacements. In describing the rotational resonance at room temperature, the method of Snoek

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[16] Ferramic *A*, Type A-34, Batch No. 165, is referred to. In a later shipment, the "ferramic *A*" contained no Mg but more Zn, and the spectrum showed only one pronounced resonance.

[17] Johnson, Rado, and Maloof, Phys. Rev. **71**, 322 (1947); M. H. Johnson and G. T. Rado, Phys. Rev. **75**, 841 (1949); Rado, Johnson, and Maloof, Rev. Sci. Instr. **20**, 927 (1949).

[18] G. T. Rado and A. Terris, Phys. Rev. **83**, 177 (1951).

(Sec. III A) was used. In describing the wall resonance, on the other hand, the equation derived combines the Landau-Lifshitz representation of relaxation effects (see Eq. (10)) with the concept of domain wall inertia. The value, $m_w$, of this inertia (or effective mass per unit wall area) was first calculated by Döring,[19] discussed by Becker[20] and Kittel,[21] and generalized by the writer.[22] The (circular) resonance frequency due to domain wall displacements, $\omega_0''$, is found[6] to be

$$\omega_0''=\gamma(M_s/\chi_0'')(8\pi\chi_0''\delta/d)^{\frac{1}{2}}, \tag{12}$$

where $\delta$ is the wall thickness parameter and $d$ denotes an average domain size. It should be noted that Eq. (12) differs from Eq. (6) only in the last factor (which can easily be of order unity) and in the replacement of $\chi_0'$ by $\chi_0''$, the latter being the static value of $\chi''$. Under certain simplifying assumptions,[6] $\chi''$ is obtained in the form

$$\chi''=\frac{\chi_0''}{1-(\omega/\omega_0'')^2+i(\omega/\omega_c'')}, \tag{13}$$

where the various quantities are given by

$$\omega_0''=(\alpha/m_w)^{\frac{1}{2}}, \quad \omega_c''=\alpha/\beta \tag{14}$$

and

$$\alpha=M_s^2/\chi_0''d, \quad \beta=\lambda/\gamma^2\delta, \quad m_w=1/8\pi\gamma^2\delta. \tag{15}$$

The approximate condition for a wall resonance (rather than a relaxation) to occur is formally expressed by[6]

$$\lambda<\gamma M_s(\delta/2\pi\chi_0''d)^{\frac{1}{2}}. \tag{16}$$

In a monocrystal the perturbing effect of internal demagnetizing fields, and hence the value of $\alpha$, the "stiffness parameter" of the walls, is expected to be relatively small, and $\chi_0''d$ relatively large. The inequality (16) is, therefore, difficult to satisfy (other quantities being assumed the same as in a polycrystal) so that the dispersion in a monocrystal is usually a relaxation, as observed by Galt.[3] Finally, Eqs. (12) through (16) do not include the line width due to a distribution in $\omega_0''$ values, a limitation that is also present in the Eqs. (6) and (9) for rotations.

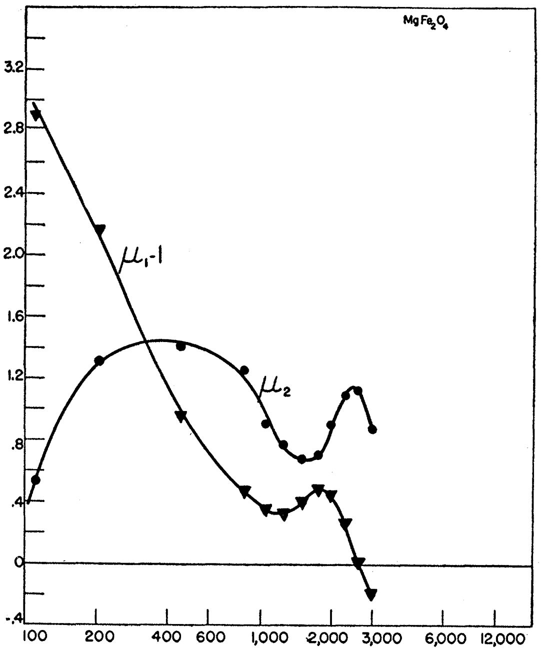

FIG. 5. Magnetic spectrum of sintered $MgFe_2O_4$ sample at room temperature.

### B. Temperature Dependence

The measured magnetic spectrum of the sintered samples of ferramic $A$ at various temperatures, represented by curves of $\mu_1-1$ (Fig. 2) and $\mu_2$ (Fig. 3), shows that two resonances were observed at 77°K, 195°K, 300°K, and 411°K. However, the two resonances begin to merge at the highest temperature investigated, a fact which is probably related to the nature of the spectrum, at room temperature, of mixed ferrites containing Zn-ferrite (Sec. V). For a detailed discussion of the temperature effects the original paper[7] should be consulted. Here it will only be noted that $\mu$ is much more temperature dependent in the low rf range, where it is ascribed primarily to domain walls, than at microwave frequencies, where it is ascribed primarily to rotations; this fact clearly substantiates that two magnetization processes play a role in the spectrum. It is also noteworthy that the spectrum of the single-domain particles at various temperatures, which is not reproduced here, shows that the (single) observed resonance frequency, denoted by $\Omega_0'$ (rather than $\omega_0'$), depends on temperature in a similar way as $\omega_0'$ and $\omega_0''$.

Although it is not yet possible to predict theoretically the widths and shapes of the absorption "lines" in a polycrystal, it is possible, as outlined below, to account for the following two simple experimental facts concerning the temperature dependence of the *mean* resonance frequencies ($\omega_0''$, $\omega_0'$, and $\Omega_0'$) in this material. (a) As the temperature is decreased from 300°K to 77°K, the three frequencies $\omega_0''$, $\omega_0'$, and $\Omega_0'$ increase by approximately the same fractional amount. (b) This common fractional increase is somewhat larger than that of $M_s$ in the range stated.

Assuming, as indicated by various data, that the role of internal stresses is negligible in ferramic $A$, and that, as in the molecular field theory of ferr*i*magnetism[23] for substances possessing only one kind of magnetic ion, the exchange energy varies with temperature as $M_s^2$, it can be shown[7] that the temperature dependence of each of the three resonance frequencies is given by $gM_s\Psi(K/M_s^2)$, where the function $\Psi$ of $K/M_s^2$ is

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[19] W. Döring, Z. Naturforsch. **3a**, 374 (1948).

[20] R. Becker, J. phys. et radium **12**, 332 (1951).

[21] C. Kittel, Phys. Rev. **80**, 918 (1950).

[22] G. T. Rado, Phys. Rev. **83**, 821 (1951).

[23] L. Néel, Ann. phys. **3**, 137 (1948).

different in the three cases and is, moreover, calculable for special situations[7] only. The temperature dependence stated above neglects damping but takes into account anisotropy, exchange forces, and magnetic interactions. If one assumes that, in the range under consideration, the proportionality $K \propto M_s^2$ is valid as a rough approximation, and neither the atomic order nor the sign of $K$ changes, then one obtains

$$\omega_0'' \propto \omega_0' \propto \Omega_0' \propto gM_s, \tag{17}$$

which explains "fact (a)" *quantitatively*. However, "fact (b)" is only *qualitatively* accounted for by (17) because measurements[7] on ferramic $A$ show that the product $gM_s$ does not increase quite as rapidly in the range from 300°K to 77°K as the natural resonance frequencies. It is of independent interest, in fact, that studies[7] of induced resonance indicate that the excess of the apparent $g$ over the value $g=2$, as well as its temperature dependence, decreases in ferramic $A$ with increasing resonance field. The possible reasons for the residual discrepancy between (17) and "fact (b)" were discussed elsewhere.[7]

## V. ON THE SPECTRUM IN THE GENERAL CASE

It may seem pertinent to inquire into the causes responsible for the apparently unique "double resonance" property of ferramic $A$. However, there are two reasons for approaching the problem from a different viewpoint. (1) As shown by the experiments discussed below, ferramic $A$ is not the only substance possessing the double resonance property. (2) There is, at present, no acceptable theoretical argument which indicates that the spectrum of an arbitrary sintered ferrite should not, in principle, contain a wall resonance as well as a rotational resonance; it is only in single-domain particles and in suitably oriented monocrystals that domain theory excludes the possibility of both resonance mechanisms playing a role. The general validity of this statement is not influenced by effects due to internal stresses or cavities in a given material.

For these reasons, it seems natural to propose the hypothesis that the spectrum of any sintered ferrite contains both types of natural resonance and that in some cases these resonances coalesce to give the appearance of a single broad resonance. The phenomenon of relaxation is, of course, considered to be a special case of resonance. On the basis of the new viewpoint, and apart from the question of the physical origin of the "damping" (i.e., relaxation) mechanism described by the phenomenological λ—term in Eq. (10), the principal problem of magnetic spectra consists in accounting for the coalescence that leads to the apparently single resonance discussed in Sec. IIIA. Before suggesting a solution to this problem, it is appropriate to present some recent data, obtained at this laboratory, on magnetic spectra of pure ferrites exhibiting the double resonance effect.

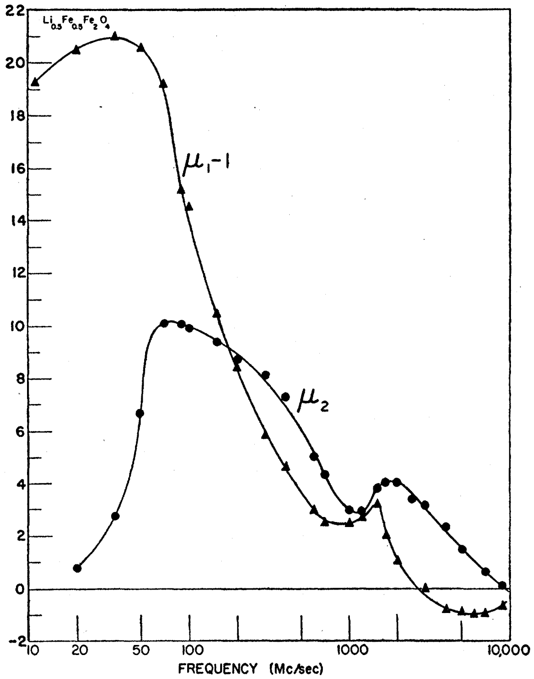

FIG. 6. Magnetic spectrum of sintered $Li_{0.5}Fe_{0.5}Fe_2O_4$ sample at room temperature. The over-all $M_s$ and density are, respectively, 258 and 4.15.

Figures (5) and (6) show the magnetic spectrum, at room temperature, of Mg-ferrite ($MgFe_2O_4$) and Li-ferrite ($Li_{0.5}Fe_{0.5}Fe_2O_4$), respectively. Similar spectra were obtained on Mg-ferrites deviating by ±5 mole percent from the stoichiometric composition and possessing different densities. In all these cases the *general* form of the spectrum is seen to resemble that of ferramic $A$. It should be noted, of course, that these results, as well as those mentioned later, are not necessarily intrinsic characteristics of the materials investigated, and they do not yet cover the whole spectrum; the data represent, instead, limited measurements on samples described by a certain chemical composition, saturation magnetization, density, and sintering treatment. At present, the crucial point is simply the fact, indicated by the figures, that two resonances occur in the spectrum of a pure Mg-ferrite and a pure Li-ferrite. A theoretical explanation for this behavior of these particular substances is not now possible because the anisotropy constant $K$ is not known. The value of $K$ is known,[24] however, for Ni-ferrite ($NiFe_2O_4$), and Eqs. (6) to (8) predict a rotational resonance frequency ($\omega_0'$) of about 840 Mc/sec at 298°K. This frequency is considerably lower than 3000 Mc/sec, the approximate location of the barely resolved microwave resonance in this material at room temperature (Fig. 7). It seems possible, therefore, that this "peak" of $\mu_2$ at 3000 Mc/sec arises from a rotational resonance caused, in part, by internal

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[24] D. W. Healy, Jr., Phys. Rev. **86**, 1009 (1952); Yager, Galt, Merritt, and Wood, Phys. Rev. **80**, 744 (1950).

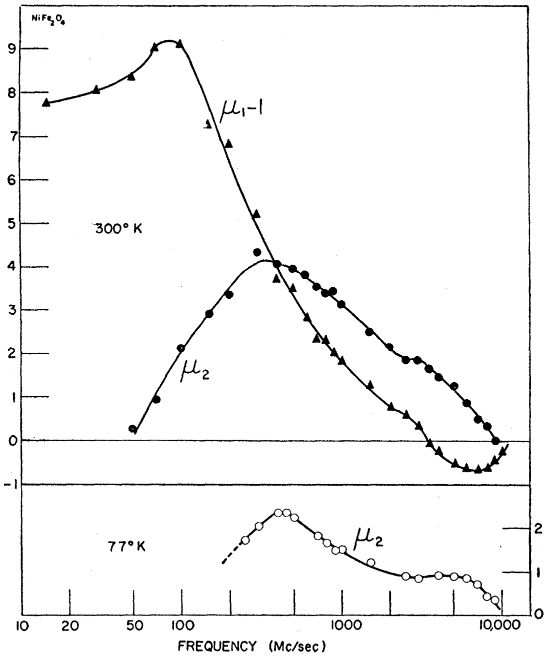

Fig. 7. Upper part: magnetic spectrum of sintered $NiFe_2O_4$ sample at room temperature. Lower part: $\mu_2$ *versus* frequency in the magnetic spectrum of same sample at 77°K. The over-all $M_s$ and density at room temperature are, respectively, 163 and 3.56.

fields due to magnetic interactions. As seen by the lower part of Fig. 7, the separation of the two resonances in Ni-ferrite can be enhanced by cooling the material to 77°K; unlike in the case of ferramic *A*, however, the simple theory[7] of the temperature dependence of $\omega_0'$ and $\omega_0''$ (Sec. IVB) is not applicable here.

The mixed ferrites investigated by Snoek (Sec. IIIA), as well as most commercial ferrites, contain Zn-ferrite as an essential component. According to Snoek's important discovery, which was interpreted by Néel[23] in his theory of ferr*i*magnetism, the presence of Zn-ferrite in a mixture decreases the Curie temperature, thus decreasing $K$ at toom temperature and increasing $\mu_1$ at frequencies below the resonance region. The decrease of $K$ results, moreover, in the (single) resonance region occurring at relatively low frequencies (see Eqs. (6) to (8)). As implied near the beginning of this section, however, neither this fact nor the evidence discussed in Sec. IIIA requires that the resonance in question be necessarily due to rotations only. It seems more reasonable to suppose that the lowering of the anisotropy field causes not only a decrease of the mean $\omega_0'$, but also a relative enhancement of magnetic interactions compared to anisotropy (i.e., a decrease of $K/M_s^2$), and thus brings about a broadening of both resonances. Since the mean wall resonance frequency ($\omega_0''$) is not,[7] in general, expected to depend more strongly on $K$ than the mean $\omega_0'$, the two resonances should coalesce if the Zn-ferrite content of the mixtures is sufficiently high. An alternative explanation of Snoek's single resonance is inherent, of course, in the lack of microwave data in his published results.

The above suggestion concerning the role of Zn-ferrite was confirmed at this laboratory by showing that the addition of enough Zn-ferrite to Mg-ferrite causes a coalescence of the two resonances. An analogous but more informative experiment was performed on a mixture containing 40 mole percent Li-ferrite and 60 mole percent Zn-ferrite. Not only do the two resonances almost coalesce (Fig. 8), but the large decrease of the mean $\omega_0'$ is, in this case, probably due to a decrease of anisotropy rather than magnetic interaction fields. This interpretation is based on the fact that, by the choice of composition, $M_s$ was kept roughly constant (226 compared to 258 in Li-ferrite) while the Curie point was lowered substantially. Similar experiments, in which Gorter's[25] data for obtaining compositions of different Zn-ferrite content but equal $M_s$ are used, should make it possible to distinguish in many other cases between the contribution of $K/M_s$ and $M_s$ to $\omega_0'$. Whenever the change in $\omega_0'$ is small, of course, then the suggested identification is not unique unless the crystallite and inclusion sizes and the density (and hence the coefficient of $M_s$) are about the same in the two compositions under consideration.

To summarize, the hypothesis proposed in this paper is consistent with all presently available data. These include the facts presented in Secs. III to V, the single-

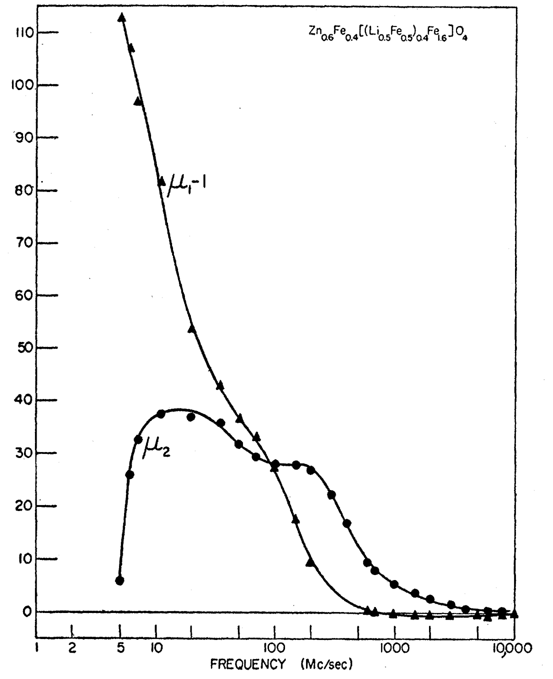

Fig. 8. Magnetic spectrum of sintered

$Zn_{0.6}Fe_{0.4}[(Li_{0.5}Fe_{0.5})_{0.4}Fe_{1.6}]O_4$

sample at room temperature. The over-all $M_s$ and density are, respectively, 226 and 4.72.

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[25] E. W. Gorter, Compt. rend. **230**, 192 (1950).

resonance spectra observed at this laboratory on a suitably prepared Mn-ferrite and Cu-ferrite, as well as other results.[26] While the interpretations proposed here may even apply to magnetic semiconductors other than ferrites, it must be anticipated that, in substances possessing a small $M_s$ value, the role of Kittel's ferri-magnetic effects[27] may have to be considered in the resonance problem. A more detailed understanding of magnetic spectra will probably depend on additional measurements of the magnetic constants ($K$, $M_s$, $\lambda$, etc.) of various materials, and on further studies of relaxation processes and magnetic interactions.

On the basis of the above discussion it appears that high $\mu_1$ values will be difficult to obtain at microwave frequencies. To obtain even $\mu_1=2$ (with $\mu_2 \ll 2$) at $10^4$ Mc/sec, for example, Eqs. (6) to (8) show that a substance characterized by $M_s \gg 430$ (and hence $K \gg 7.5 \times 10^5$ ergs/cc) would be required. Nevertheless, $\mu_1=2$ is clearly more useful than $\mu_1=1$, and, in addition, interesting wave propagation effects can be achieved by operating within a (narrow) resonance region. It is also noteworthy that, contrary to Snoek's theory, the total low frequency $\mu_1$ may still be large even if the microwave $\mu_1$ is small (see Eq. (11)). Furthermore, materials with a low Curie point, such as mixed ferrites containing a large amount of Zn-ferrite, are clearly not satisfactory for ordinary microwave use, but may sometimes be suitable for obtaining low insertion loss in devices based on the Faraday effect.

In conclusion, the writer wishes to thank R. W. Wright, W. H. Emerson, A. Terris, and M. Maloof for their invaluable contributions to the experiments discussed in this paper.

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[26] Welch, Nicks, Fairweather, and Roberts, Phys. Rev. 77, 403 (1950).

[27] C. Kittel, Phys. Rev. 82, 565 (1951).

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# Resonance Phenomena in Ferrites

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ABOVE a certain frequency, of the order of 5–100 Mc/sec, losses of ferrites in an alternating magnetic field are very high. Snoek[1] has ascribed this absorption to rotational resonance of the spins in the internal anisotropy fields, and has related the resonance frequency $f_0$ with the static initial permeability $\mu_0$ by means of the equation

$$f_0(\mu_0-1)=(4/3)\gamma M, \tag{1}$$

with $\gamma$ the gyromagnetic ratio and $M$ the saturation magnetization. Experimentally considerable losses are found at the predicted $f_0$, but the absorption region is very broad and extends to frequencies above 1000 Mc/sec. In this connection one of us[2] has pointed out that in the resonating unmagnetized material magnetic poles will be created and annihilated on a Bloch wall, if the alternating field has a component parallel to it. In some regular arrangements of Weiss domains this effect can, together with demagnetization at the grain boundaries, give rise to high resonance frequencies. This, and also the occurrence of losses near $f_0$, will be illustrated with the following model.

Consider an ellipsoid divided into thin Weiss domains separated by 180° walls perpendicular to one of its principal axes. The ellipsoid is embedded in a medium with the average permeability $\mu(f)$ of the material. For ac fields perpendicular to the walls only the external demagnetization in the field direction plays a role (Fig. 1). The effect is reduced by the presence of the medium by a factor of the order of $\mu$, which is large at relatively low frequencies. The influence of demagnetization and the formation of poles on the walls

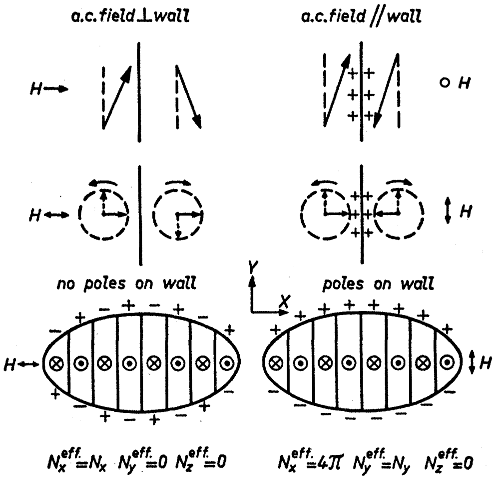

FIG. 1. Effect of Weiss domain structure on ferromagnetic resonance conditions.

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[1] J. L. Snoek, Nature 160, 60 (1947) and Physica 14, 207 (1948).

[2] D. Polder, J. phys. et radium 12, 337 (1951).