Magnetic Rotation Phenomena in a Polycrystalline Ferrite*

DAVID PARK
Williams College, Williamstown, Massachusetts
(Received August 13, 1954)

In order to calculate the initial susceptibility at high frequencies of a polycrystalline sample of ferrite, one must take account of the interaction between neighboring crystallites, and of the fact that the crystallites differ from each other in their orientation (assumed random) and in their characteristic resonant frequencies. Each crystallite is acted upon by an oblique alternating field, and its magnetization precesses in an elliptical cone. A simple solution of this problem is given by the use of complex variables involving two different imaginaries. It is found that the interaction between crystallites has little effect on the magnetic spectrum other than to raise the effective value of the resonant frequencies, and that the spectrum obtained is not very sensitive to the exact form chosen for the distribution of resonant frequencies among the crystallites.

The resulting expressions for the real and imaginary parts of the susceptibility are compared with the results of some measurements by F. Brown and C. L. Gravel on a lightly-sintered sample of nickel ferrite, assuming that the major part of the susceptibility is due to rotation processes. The agreement is satisfactory and shows among other things that the damping constant need be no larger in these materials than it is found to be in a single crystal.

1. INTRODUCTION

THE magnetic spectrum of unmagnetized polycrystalline samples of ferrite has usually been considered in two ranges, the boundary between them lying roughly at 500 Mc/sec. Above this value, the effects of domain rotation are identified, while below it the permeability is attributed largely to the effects of domain wall motion.[1] Recently, however, an extensive series of experiments[2] has been done on samples of nickel ferrite in which the crystallite sizes are rather small—of the order of a few microns—and there appears to be considerable evidence that rotation phenomena are largely responsible for the permeability over the entire range of frequencies studied (2 to 10 000 Mc/sec).[3] Since these conclusions have been discussed elsewhere,[2] we shall only summarize them briefly here. First, the spectra of grains of single-domain size embedded in wax (where wall motions would not be expected to play any part) are qualitatively similar to, and form part of a continuous progression with, the spectra of samples sintered at progressively higher temperatures. Second, the evidence provided by Snoek's relation between initial susceptibility and resonant frequency[4] (see Sec. 4) is against the hypothesis of wall motion, and third, there is the intuitive objection that the numerous holes, traps and impurities present in any sintered mass would so immobilize any domain walls which might be formed there that they would hardly be expected to react at all to an applied field. This objection ceases to apply, of course, when the crystallites are large enough to contain several domains.

2. THE THEORETICAL MODEL

The theory of domain rotations in a sintered mass of ferrimagnetic material differs from that previously developed for a crystalline sample[5] in three respects. First, the sintered crystallites must be taken to be randomly oriented; second, they will interact magnetically with each other somewhat in the manner of coupled oscillators; and third, the magnetic properties of the material will vary from one crystallite to another, according to their chemical purity, shape, size, state of stress, etc. In what follows we shall suppose that the resonant frequencies characterizing the individual crystallites are distributed in some arbitrary manner. The problem of determining the magnetic response of such a substance is, as it stands, of course quite insoluble, so we shall approximate it by means of the following model, which is capable of exact calculation.

We shall suppose that any given crystallite X is surrounded by several neighbors N, oriented at random, but numerous enough so that their, average magnetization is the same as that characterizing the sample as a whole. Let us suppose that under the influence of an external alternating magnetic field $\mathbf{h}$, whose frequency is ω, this average magnetization is $\mathbf{M}(\omega,t)$. It is clear from symmetry that the average magnetization of a randomly oriented mass cannot have its direction other than parallel to the external magnetizing field $\mathbf{h}$. Clearly also, the averaged field produced by the neighboring crystallites must lie along this line; though it will be directed oppositely to $\mathbf{M}$, since the lines of force external to a magnetized body go round continuously outside it. If a crystallite (or lump of crystallites) N is of size l, it will have a magnetic moment of the order of

* This work was supported by the Sprague Electric Company, North Adams, Massachusetts.

[1] G. T. Rado, Revs. Modern Phys. **25**, 81 (1953). This article reviews the extensive evidence in support of this view, and contains references to most of the earlier work. See also H. P. J. Wijn, dissertation, Leyden, 1953 (unpublished).

[2] F. Brown and C. L. Gravel, Phys. Rev. **95**, 652(A) (1954); see also preceding article [Phys. Rev. **97**, 55 (1954)].

[3] The possibility that the dispersion is all due to rotation was first put forward by members of the Philips group [J. Went and H. Wijn, Phys. Rev. **82**, 269 (1951)].

[4] J. L. Snoek, Physica **14**, 207 (1948).

[5] F. Brown and D. Park, Phys. Rev. **93**, 381 (1954).

Ml^3, and will produce at X a field of the order of Ml^3/l^3, so that the total field acting upon X is given by

$$\mathbf{H}=\mathbf{h}-\rho\mathbf{M}, \tag{1}$$

with ρ, depending on permeability and on crystalline properties, at least of the order of unity.[6] In such a field (we assume a toroidal sample so that $\mathbf{h}$ is not demagnetized) the crystallite X will exhibit a certain rotating magnetization $\mathbf{M}_X$, characteristic of the individual magnetic properties of X. To find the magnetization of a large number of crystallites X, we average this response to the applied field over their resonant frequencies (and, later, over their orientations). But this average $\mathbf{M}$, characterizing the whole mass, must be the same as that which was assumed to characterize the neighbors of any X. This identification will be written in the form of an equation which can then be solved for $\mathbf{M}$. Finally, comparison of this average magnetization with the exciting field $\mathbf{h}$ permits us to find the real and imaginary parts of the susceptibility, which can be compared with experiment.

3. THE SUSCEPTIBILITY

We shall write the equation of motion for the magnetization of the crystallite X in the form given by Landau and Lifschitz:[7]

$$\dot{\mathbf{M}}_X=\gamma\mathbf{M}_X\times\mathbf{H}_T-\frac{\lambda}{M_X{}^2}\mathbf{M}_X\times\mathbf{M}_X\times\mathbf{H}_T, \tag{2}$$

where $\mathbf{H}_T$, composed of the alternating field $\mathbf{H}$ and the fixed anisotropy field $\mathbf{H}_{A0}$ (we assume that there is no fixed external field), is the total magnetic field acting on $\mathbf{M}$, and γ is the effective value of the gyromagnetic ratio for the two-lattice system.[5] Let us adopt a coordinate system $\mathfrak{X}$, with the z-axis chosen to coincide with the direction of $\mathbf{H}_{A0}$. Then if the applied field is not too large, we can neglect its z-component entirely. With respect to these axes, the direction of $\mathbf{h}$ will have some arbitrary orientation, given by the polar angles θ and φ:

$$h_{Xx}=h_0\sin\theta\cos\varphi e^{-i\omega t},\quad h_{Xy}=h_0\sin\theta\sin\varphi e^{-i\omega t}, \tag{3}$$

where the time variation has been put into complex form in order to facilitate the consideration of phase relationships later. It is convenient to consider the x-y plane in the coordinate system $\mathfrak{X}$ as a complex plane in which the y-direction is denoted by the imaginary quantity i. This is, of course, an actual physical plane, as contrasted with that whose imaginary unit is j, and which is merely a computational device. Letting $M_{Xx}+iM_{Xy}=M_{X+}$, and similarly with other quantities, we find that (2) can be expressed in the simple form

$$\dot{M}_{X+}=-i\gamma'(H_{A0}M_{X+}-H_{X+}M_s), \tag{4}$$

where

$$\gamma'=(1-i\alpha)\gamma,\quad \alpha=\lambda/\gamma M_s, \tag{5}$$

and M_s, the saturation magnetization of the crystallite, is the approximately constant value of M_z. Thus the only effect of the damping, as introduced in this way, is to give to the effective value of the gyromagnetic ratio a negative imaginary part in i.

To put in the field $\mathbf{h}$, let us introduce an average alternating susceptibility of the entire sample,

$$\chi_+=M_+/h_+. \tag{6}$$

With this, we can put (1) into (4) to get, for the magnetization of the crystallite X,

$$\dot{M}_{X+}=-i\gamma'[H_{A0}M_{X+}-(1-\rho\chi_+)M_sh_{X+}], \tag{7}$$

where, by (3),

$$h_{X+}=h_{X0+}e^{-i\omega t},\quad h_{X0+}=h_0\sin\theta e^{i\varphi}. \tag{8}$$

Then, if we assume that M_{X+} in the steady state is of the form $M_{X0+}e^{-i\omega t}$, we can readily solve this [with (5)] to get

$$M_{X0+}=\frac{\gamma M_s(1-\rho\chi_+)h_{X0+}}{[ij\omega/(1-i\alpha)]+\omega_X}, \tag{9}$$

where ω_X, the characteristic frequency of X, is given by γH_{A0}.

The susceptibility of X is determined by the storage and dissipation of energy in X as $\mathbf{h}$ goes through its cycle. Thus we need only consider that part of $\mathbf{h}$ which does work on $\mathbf{M}_X$, and the susceptibility can be written as

$$\chi_{X+}=(M_{X0+}h_{X0-})/h_+h_-. \tag{10}$$

When (9) is put into this, we encounter the product

$$h_{X0+}h_{X0-}=h_0{}^2\sin^2\theta.$$

We can now immediately average this over all possible orientations of X. The $\sin^2\theta$ becomes $\frac{2}{3}$, and we find[8]

$$\bar{\chi}_{X+}=\tfrac{2}{3}\gamma M_s(1-\rho\chi_+)\frac{1}{[ij\omega/(1-i\alpha)]+\omega_X}. \tag{11}$$

Now we are to average this over the different resonant frequencies ω_X that X may have. It is certain, of course, that, besides ω_X, other parameters such as γ and M_s will vary from one crystallite to another. However, the most marked effect on the resulting permeability is produced by the statistical distribution in ω_X, and we shall consider only this. Suppose that the distribu-

[6] This problem of averaging is somewhat similar to that encountered by Lorentz in calculating the internal field of an electrically polarized body [H. A. Lorentz, *The Theory of Electrons* (Dover Publications, Inc., New York, 1952), p. 138 and Note 55]. The approach in this paper consists essentially in assuming that there is no appreciable short-range order in the precession of the spins.

[7] L. Landau and E. Lifschitz, Physik. Z. Sowjetunion 8, 153 (1935).

[8] The occurrence of i in formulas (9) and (11) shows that the magnetization, both of X and of the directionally averaged X, precesses in an ellptical cone under the action of the applied field.

tion is according to some law $q(\omega_X)$, where

$$\int_0^\infty q(\omega_X)d\omega_X=1.$$

Then, if we define

$$p(\omega)=\int_0^\infty \frac{q(\omega_X)d\omega_X}{[ij\omega/(1-i\alpha)]+\omega_X}, \tag{12}$$

the average susceptibility of the entire mass will be

$$\chi_+=\tfrac{2}{3}\gamma M_s(1-\rho\chi_+)p(\omega).$$

The hypothesis mentioned earlier—that the average susceptibility of the neighbors of any X is the average susceptibility of all X—equates the two χ_+'s in the expression above, and we find that

$$\chi_+=\tfrac{2}{3}\gamma M_s p(\omega)/[1+\tfrac{2}{3}\rho\gamma M_s p(\omega)].$$

χ_+ is, however, closely related to the measured permeability of the sample, for the latter is defined (again, in terms of energy storage and dissipation) as

$$\chi=\frac{M_x h_x+M_y h_y}{h_x^2+h_y^2}=\frac{1}{h_+h_-}\,\mathrm{Re}_i M_+h_-=\mathrm{Re}_i\chi_+,$$

where Re_i denotes that the real part with respect to i is to be taken, and we have chosen coordinates such that **h** has no z-component. Thus, we have finally

$$\chi=\tfrac{2}{3}\gamma M_s\,\mathrm{Re}_i\{p(\omega)/[1+\tfrac{2}{3}\rho\gamma M_s p(\omega)]\}. \tag{13}$$

As a first orientation, let us consider the case in which the distribution $q(\omega_X)$ is infinitely narrow and centered around some frequency ω_0. In this case, (13) is[9]

$$\chi=\tfrac{2}{3}\gamma M_s \times\frac{(1+\alpha^2)(\omega_0+\tfrac{2}{3}\rho\gamma M_s)-j\alpha\omega}{(1+\alpha^2)(\omega_0+\tfrac{2}{3}\rho\gamma M_s)^2-\omega^2-2j\alpha\omega(\omega_0+\tfrac{2}{3}\rho\gamma M_s)}. \tag{14}$$

It is clear from this, and can be verified to hold for the more general cases to be discussed later, that the principal effect of the intercrystallite interaction is merely to make the frequency at which resonance occurs somewhat greater than ω_0. Since, however, ω_0 (or, in the case of a distribution q, the central value of the distribution) is itself only determinable empirically, we shall henceforth omit the term in ρ and consider ω_0 to represent the increased value. The resultant error in the shapes of the permeability spectra can be expected to be less than that which will result from other assumptions.

Although (14) must be considered an idealization, still it exhibits a number of features which characterize most of the experimental data. Not only is the general shape of the curves (for large enough values of α) the same as those observed, but it is also significant that the asymptotic behavior for large ω is right: writing $\chi=\chi'+j\chi''$, we see that $\chi'\sim\omega^{-2}$ and $\chi''\sim\omega^{-1}$, as observed. The correctness of the asymptotic behavior in this respect is a rather stringent test of any theory. Introducing damping through the use of a relaxation time in the equations of motion,[10] for example, fails to predict this behavior, no matter how the relaxation time is assumed to depend on ω. Equation (14), however, cannot be considered a satisfactory result. First, the data require values of α of the order of 0.8, whereas the measurements of Yager[11] and Healy[12] on single crystals give a value of 0.0045. One would expect it be increased by sintering, but not by so much. Second, the data on χ' are not well fitted for any choice of α; and third, as we have pointed out above, it is not reasonable to expect that the crystallites will all resonate at the same frequency.

In order to continue, we need to assume a distribution function $q(\omega_X)$ which leads to analytically tractable results. A first attempt was made[13] with a rectangular distribution, and the resulting formulas showed a surprising agreement with experiment. Typical curves, together with the assumed distribution function, are shown in Fig. 1. Some qualitative predictions which agree well with the available data[2] are that if A and B are the upper and lower limiting frequencies of the rectangular distribution, then they are the frequencies at which[14] μ' takes its extreme values and they are at the same time the "half-power" points of the curve of μ''. Two other features characteristic of the observed spectra are the great asymmetry of the two extremes of μ' (the asymmetry about $\mu'=1$ is frequently as great as 20 to 1) and the fact that the frequency at which μ' equals unity is usually much greater than that at which μ'' has its maximum (a factor of 10 is not uncommon

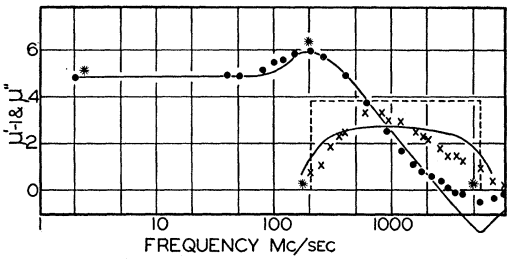

FIG. 1. Data on a sample of nickel ferrite sintered at 1216°C. The circles represent $\mu'-1$ and the crosses represent μ''. The theoretical curves (solid lines) are obtained by assuming a rectangular distribution of crystallite frequencies (dashed line), and are fitted to the data at the points marked with asterisks. This includes an arbitrary scaling of μ so as to fit the data at low frequencies.

[9] If ρ is neglected, this formula becomes identical (except for the sign of j) with one given by Kittel (see reference 1). The formula appears in C. Kittel, J. phys. radium **12**, 332 (1951).

[10] N. Bloembergen, Phys. Rev. **78**, 572 (1950).
[11] Yager, Galt, Merritt, and Wood, Phys. Rev. **80**, 744 (1950).
[12] D. W. Healy, Jr., Phys. Rev. **86**, 1009 (1952).
[13] D. Park, Phys. Rev. **95**, 962(A) (1954).
[14] We write $\mu=1+4\pi\chi=\mu'+j\mu''$.

here). These features are very difficult to understand from the standpoint of wall motions, but both are immediate consequences of our formulas. [For example, the frequency ratio just mentioned is given as approximately $(A/2B)^{\frac{1}{2}}$, which is equal to 3 for the calculation of Fig. 1, as compared with the experimental value of 4.] More exactly, the peak value of μ'' occurs at a frequency of $(AB)^{\frac{1}{2}}$, and the zero of $\mu'-1$ occurs at $[\frac{1}{2}(A^2+B^2)]^{\frac{1}{2}}$. These characteristics also do not follow from the equations of motion in which damping is introduced through a relaxation time. It is interesting to note, however, that Young and Uehling[15] find that experiments on ferromagnetic metals decide in favor of the latter equations.

4. A FOUR-PARAMETER DISTRIBUTION

If we are to avoid nonphysical hypotheses such as that of the rectangular distribution, and at the same time not become involved in analytical work disproportionate to the accuracy of our theory, there is not a very wide range of forms available for q. We shall discuss here the consequences of a distribution of the form

$$q(\omega_X)=\frac{1}{\pi(A-B)}\left(\tan^{-1}\frac{A-\omega_X}{\sigma_1}-\tan^{-1}\frac{B-\omega_X}{\sigma_2}\right), \quad (15)$$

with $\sigma_1 \ll A$ and $\sigma_2 \ll B$. This is a natural generalization of the rectangular distribution; an example is shown in Fig. 2. The conditions are imposed on the σ's so that the contribution to q from values of ω_X equal to zero and below can be neglected. If σ_1 and σ_2 are taken to be equal, we can conveniently represent this as

$$q(\omega_X)=\frac{\sigma}{\pi(A-B)}\int_B^A \frac{d\omega_0}{(\omega_X-\omega_0)^2+\sigma^2} \quad (\sigma \ll B), \quad (16)$$

a superposition of bell-shaped distributions centering about various values of ω_0. If σ_1 is not equal to σ_2 (and it seems usually to be necessary to assume this), we can still write (15) as

$$q(\omega_X)=\frac{1}{\pi(A-B)}\left\{\sigma_2\int_B^\infty \frac{d\omega_0}{(\omega_X-\omega_0)^2+\sigma_2^2} -\sigma_1\int_A^\infty \frac{d\omega_0}{(\omega_X-\omega_0)^2+\sigma_1^2}\right\}. \quad (17)$$

Changing the order of integration, we have first to evaluate (12) using a q of the form $\sigma/\pi[(\omega_X-\omega_0)^2+\sigma^2]$. The result of doing this is rather complicated;[16] we shall give it here in unrationalized form, omitting terms in α^2:

$$\chi_\sigma=\tfrac{2}{3}\gamma M_s\left[\frac{\omega_0-j\alpha\omega}{(\omega_0-j\alpha\omega)^2+(\sigma+j\omega)^2} +\sigma\frac{2\alpha\omega(\omega-\omega_0)+j((\omega-\omega_0)^2+\sigma^2)}{[(\omega-\omega_0)^2+\sigma^2]^2}\right], \quad (18)$$

from which one can verify the asymptotic properties at high frequencies mentioned above, and in the form which it assumes when all terms in α are neglected:

$$\chi_\sigma'=\tfrac{2}{3}\gamma M_s\omega_0\frac{\omega_0^2+\sigma^2-\omega^2}{(\omega^2-\omega_0^2+\sigma^2)^2+4\omega_0^2\sigma^2}, \quad (19)$$

and

$$\chi_\sigma''=\tfrac{2}{3}\gamma M_s\sigma\frac{\omega_0^2+\sigma^2+\omega^2}{(\omega^2-\omega_0^2+\sigma^2)^2+4\omega_0^2\sigma^2}. \quad (20)$$

These can be compared with the expressions[9] which follow from (14), if one neglects ρ and assumes that all the width comes from the damping α:

$$\chi_\alpha'=\tfrac{2}{3}\gamma M_s\omega_0\frac{(1+\alpha^2)^2\omega_0^2-(1-\alpha^2)\omega^2}{[\omega^2-(1-\alpha^2)\omega_0^2]^2+4\alpha^2\omega_0^4}, \quad (21)$$

and

$$\chi_\alpha''=\tfrac{2}{3}\gamma M_s\alpha\omega\frac{(1+\alpha^2)\omega_0^2+\omega^2}{[\omega^2-(1-\alpha^2)\omega_0^2]^2+4\alpha^2\omega_0^4}. \quad (22)$$

It will be noted that these two pairs of equations are very similar, with σ replaced by $\alpha\omega_0$ (in fact, it is in general impossible to distinguish experimentally between effects of α and effects of σ), except that χ_α'' in (22) vanishes with ω, as it should, whereas in (20) one can see the spurious effect of a $q(\omega_X)$ which does not vanish with ω_X.

The integrations in (17) can now easily be carried out to give

$$\chi'=-\frac{1}{6}\frac{\gamma M_s}{A-B}\ln\frac{(\omega^2-A^2+\sigma_1^2)^2+4A^2\sigma_1^2}{(\omega^2-B^2+\sigma_2^2)^2+4B^2\sigma_2^2}, \quad (23a)$$

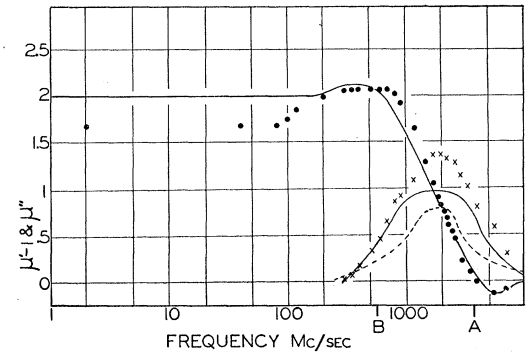

FIG. 2. Data on a sample of nickel ferrite lightly sintered at 960°C. The circles represent $\mu'-1$ and the crosses represent μ''. The theoretical curves (solid line) have been obtained by adjusting the assumed distribution of crystallite frequencies (dashed line) so as to obtain a fit. (There has been no arbitrary scaling.)

[15] J. A. Young, Jr., and E. A. Uehling, Phys. Rev. 94, 544 (1954).

[16] The integral can be evaluated by means of a contour, but since there are here two imaginaries, it is necessary to use some discretion in determining where the singularities lie.

and

$$\chi''=\frac{1}{3}\frac{\gamma M_s}{A-B}\left(\tan^{-1}\frac{A+\omega}{\sigma_1}+\tan^{-1}\frac{A-\omega}{\sigma_1}-\tan^{-1}\frac{B+\omega}{\sigma_2}-\tan^{-1}\frac{B-\omega}{\sigma_2}\right) \quad (23b)$$

$$=\frac{1}{3}\frac{\gamma M_s}{A-B}\left(\tan^{-1}\frac{2A\sigma_1}{\omega^2-A^2+\sigma_1^2}-\tan^{-1}\frac{2B\sigma_2}{\omega^2-B^2+\sigma_2^2}\right). \quad (23c)$$

From these we can find the values at zero frequency,

$$\chi'(0)=\frac{1}{3}\frac{\gamma M_s}{A-B}\ln\frac{A^2+\sigma_1^2}{B^2+\sigma_2^2}, \quad (24a)$$

and

$$\chi''(0)=\frac{2}{3}\frac{\gamma M_s}{A-B}\left(\tan^{-1}\frac{\sigma_2}{B}-\tan^{-1}\frac{\sigma_1}{A}\right). \quad (24b)$$

The imaginary part should, of course, vanish. That it is small is a consequence of the conditions laid on (15). It is often convenient (and accurate enough) to choose $\sigma_1/A=\sigma_2/B$ in order to simplify the analysis generally, and this, in (24b) has the desired effect, although for the wrong reason. We have in this case

$$\chi'(0)=\frac{2}{3}\frac{\gamma M_s}{A-B}\ln\frac{A}{B}, \quad \chi''(0)=0 \quad \left(\frac{\sigma_1}{A}=\frac{\sigma_2}{B}\right). \quad (24c)$$

If (as will generally be the case) $A\gg B$, we find that the minimum of χ' occurs at a frequency

$$\omega_1\approx\frac{A^2+\sigma_1^2}{(A^2-\sigma_1^2)^{\frac{1}{2}}}, \quad \chi'(\omega_1)\approx-\frac{1}{3}\frac{\gamma M_s}{A-B}\ln\frac{A^2+\sigma_1^2}{2A\sigma_1}, \quad (25)$$

while the maximum is at

$$\omega_2\approx(B^2-\sigma_2^2)^{\frac{1}{2}}, \quad \chi'(\omega_2)\approx\frac{1}{3}\frac{\gamma M_s}{A-B}\ln\frac{A^2+\sigma_1^2}{2B\sigma_2}, \quad (26)$$

and, between these, χ' vanishes at a frequency close to $\omega_1/\sqrt{2}$. A measure of the peaking of χ' is given by

$$P=\frac{\chi'(\omega_1)-\chi'(0)}{\chi'(0)}\approx\ln\left(\frac{B^2+\sigma_2^2}{2B\sigma_2}\right)\Big/\ln\left(\frac{A^2+\sigma_1^2}{B^2+\sigma_2^2}\right). \quad (27)$$

There is an important difference between the results of this work and those of the earlier calculation[13] based on the rectangular distribution of frequencies. It is that, when q rises or falls off in a distance σ, it is σ and not α (if α is small enough) that determines the extreme values of χ'. Thus α may be as small as desired, and the data can still roughly be fitted.

It will be convenient to consider specially two frequencies characteristic of the resonance; those at which χ' drops to half its peak and DC values will be called respectively $\omega_{\frac{1}{2}}$ and ω_r. The latter is the "resonant frequency" by which Wijn[1] characterizes a material. If $A\gg B$, these approach the values

$$\omega_{\frac{1}{2}}\approx[2B\sigma_2(A^2+\sigma_1^2)]^{\frac{1}{4}} \text{ and } \omega_r\approx[(A^2+\sigma_1^2)(B^2+\sigma_2^2)]^{\frac{1}{4}}; \quad (28)$$

and in the case which we have discussed above, in which $\sigma_1/A=\sigma_2/B=\lambda$, say, these become

$$\omega_{\frac{1}{2}}\approx\left(\frac{2\lambda}{1+\lambda^2}\omega_1^2\omega_2^2\right)^{\frac{1}{4}} \text{ and } \omega_r\approx(\omega_1\omega_2)^{\frac{1}{2}}. \quad (29)$$

The first of these can be solved to give an estimate of λ:

$$\frac{\lambda}{1+\lambda^2}\approx\frac{1}{2}\left(\frac{\omega_{\frac{1}{2}}^2}{\omega_1\omega_2}\right)^2 \quad (30)$$

in which it is usually permissible to neglect the λ^2, while the extent to which the second is satisfied provides a very simple check on the applicability of this simplified form of the theory to any given set of data.

The quantity ω_r also enters into Wijn's formulation of Snoek's relation[4] between resonant frequency and static permeability. Again in the simplified case, (24c) and (29) give

$$\omega_r(\mu'-1)_0=\frac{8\pi}{3}\gamma M_s\frac{(\omega_1\omega_2)^{\frac{1}{2}}}{A-B}\ln\left(\frac{A}{B}\right).$$

If (as seems to be quite common) λ is in the neighborhood of $\frac{1}{2}$, then it is easily verified that A is close to $\omega_1/\sqrt{2}$, the frequency at which χ' vanishes. Neglecting B in the denominator and making an unimportant approximation in the logarithm now gives

$$\omega_r(\mu_0-1)_0=(8\pi/3)\gamma M_s(2\omega_2/\omega_1)^{\frac{1}{2}}\ln(\omega_1/\omega_2). \quad (31)$$

Without the terms in ω on the right, this is Snoek's relation. The terms in ω provide a factor which is normally very close to unity. Brown and Gravel[2] have applied this formula to the analysis of data obtained by them, and we shall therefore apply it here only to Wijn's data on his sample E of Ferroxcube IV (the only one for which the data are adequate): we find[17]

$$\omega_r=350 \text{ Mc/sec}, \quad \omega_1=2700\sqrt{2} \text{ Mc/sec}, \quad \omega_2=250 \text{ Mc/sec}, \quad M_s=197,$$

where ω_1 has been estimated as $\sqrt{2}$ times the frequency at which χ' vanishes. The factor in ω on the right is in this case 0.98, and explains why Snoek's law in its simple form is so closely obeyed by Wijn's data. In this case, the values of $(\mu'-1)_0$ calculated by (31) and observed are 14 and 12, respectively.

In comparing these formulas with experiment it is necessary to take account of the porosity of the poly-

[17] The constants are given in terms of ordinary frequency, not angular frequency.

crystalline samples—their packing factor (density of sample divided by density of pure crystalline substance) is seldom over 75 percent. It is clear that the saturation magnetization M_s will be reduced in the same ratio, and that the susceptibility will also be correspondingly less, though the effect of the magnetic poles induced on the crystallite boundaries is difficult to evaluate.[18] In the present work we assume that the effect of porosity can be accounted for by using instead of M_s the saturation magnetization of the polycrystalline sample, but we hope to return later to a quantitative discussion of this point.

In comparing theory with experiment there are two considerations which limit us in the choice of data. The first is the fact that we have neglected the effect of the interaction between crystallites on the shape of the resonance curves, so that it will be advantageous to examine data taken on a sample which has been relatively lightly sintered. The second is more serious: the slowness with which χ'' decreases at high frequencies (this is particularly evident in Wijn's data[1]) entails a value of σ_1 which is comparable with A or even greater, contradicting an assumption which was made in order to obtain results in tractable form. It is fortunate that this effect is less pronounced precisely in the ferrite samples which have been sintered at relatively low temperatures;[2] accordingly, we shall pick for comparison Brown's measurements on a sample sintered at 960°C, with a packing factor $p=0.54$. Figure 2 shows the comparison of theory with experiment in this case. The constants used here are,[17] in kMc/sec (10^9 cps):

$$A=4.2, \quad \sigma_1=2.1, \quad B=0.58, \quad \sigma_2=0.29;$$

but the choice could probably be improved upon. In particular, the error in $\chi'(0)$ should not be regarded as significant. Also, the choice of a $q(\omega_X)$ which was more sharply peaked would certainly have brought the theoretical curve of μ'' higher. It is characteristic of the *Ansatz* (15) that the peaks possible with it are not as high as generally required. For pure crystals of nickel ferrite, M_s is[1] 265, and the effective value[5,17] of γ is 1.1×2.80 Mc/sec oersted. The results indicate quite strongly that the mechanism which we have discussed above is in this case the correct one.

5. REMARKS

A detailed discussion, from the above standpoint, of the various experiments which have been performed on resonance phenomena in ferrites is beyond the scope of this paper, though we hope to return to the subject elsewhere. For the present, let us merely note that the mechanism which we have assumed here leads directly to a slightly modified form of Snoek's relation and, conversely, that whenever it is found by experiment that the susceptibility exhibits a resonance which satisfies this relation, there is some likelihood that domain rotation, rather than wall motion, plays the leading part in the resonant process. But there are certain instances[1,19] in which a substance (notably Ferramic A) shows two resonances, of which one, in the rf region, approximately obeys Snoek's law and the other, a much lower peak in the microwave region, does not. If we consider the possibility that the rf resonance is due to domain rotation, we must then look for an alternative explanation for the other peak.

It is possible that the microwave resonance is due to the free gyration of electron spins. The fact that in some materials the magnitude, and even the existence, of the high-frequency peak vary from sample to sample[20] suggests that this resonance is not a property of the crystallites themselves (it has not been reported in single-crystal samples) but of their state of aggregation. Let us suppose, accordingly, that these electrons are trapped in various lattice imperfections.[21] If the trap is effectively a spherical cavity, the electron will experience a field[22] of magnitude $4\pi M_e/3$, where M_e is some effective value of the local magnetization. For want of a better value, we shall use for it M_s, the saturation magnetization of the sintered mass. The electron's spin will resonate at the frequency

$$\omega_e=\gamma(4\pi/3)M_e=11.7M_e \text{ Mc/sec},$$

and the width of the resonance is due more to the lack of sphericity of the trapping region than to any process of dissipation. In support of this hypothesis, there are data[1] at room temperature on Ferramic A ($M_s=99$ oersteds, $\omega_e=1100$ Mc/sec, observed value 1100 Mc/sec), $Li_{\frac{1}{2}}Fe_{\frac{1}{2}}Fe_2O_4$ ($M_s=258$, $\omega_e=3000$, observed value about 2000 Mc/sec) and $NiOFe_2O_3$ ($M_s=163$, $\omega_e=1900$, observed value 3000 Mc/sec). All the observed resonance peaks are at least 1000 Mc/sec wide. Also, in Rado's results on nickel ferrite, the fact that the resonance is barely observable at room temperature but pronounced at 77°K is explainable on the ground that more electrons are trapped at lower temperatures. The same effect is observable in Rado's data on Ferramic A. His measurements on nickel-zinc ferrite, however, do not at all support this view.

If the foregoing explanation of the high-frequency peak is wrong, it is probably for one of two reasons: the effect might be inappreciable,[23] or the spread in the field strengths experienced by the electrons might be so great as not to produce a resonance at all,[24] but only a general distortion of the microwave end of the sus-

[18] D. Polder and J. Smit, Revs. Modern Phys. **25**, 89 (1953).

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

[20] F. Brown (private communication).

[21] I am indebted to F. J. Dyson for this suggestion.

[22] Reference 6, Note 54.

[23] The situation here differs from that in a metal, where the skin effect prevents the magnetic field from penetrating more than a short distance below the surface. In metals, however, the corresponding effect has been observed. [G. T. Rado and J. R. Weertman, Phys. Rev. **94**, 1411(A) (1954).]

[24] It should be noted that in all cases, the microwave resonance peak is far broader than the rf one; the logarithmic presentation of frequencies is deceptive in this respect.

ceptibility curve, and in particular, a lifting of the curve of χ''. Such an effect is apparent in the experiments of Wijn[1] and Brown,[2] and the latter measurements were carried out to a frequency high enough so that one can be sure there is no microwave peak present. In the absence of any theoretical or experimental justification, however, these speculations must remain very tentative.

I am grateful to Fielding Brown and J. Kenneth Moore for communicating to me the results of their measurements and calculations, and for making many contributions to the work presented here.

PHYSICAL REVIEW VOLUME 97, NUMBER 1 JANUARY 1, 1955

Effect of Strong Electrostatic Fields on the Resistance of Tungsten Wires in High Vacua

W. J. DESHOTELS* AND A. H. WEBER
Saint Louis University, St. Louis, Missouri

(Received March 11, 1954; revised manuscript received July 26, 1954)

The effect of a radial electrostatic field upon the resistance of tungsten in high vacua was reinvestigated employing lower pressures and better vacuum technique than in the original experiments of Worthing *et al.* Wires of 0.004-in. and 0.00045-in. diameter were subjected to negative and positive fields (retarding or assisting respectively electron motion from filament to plate) up to 9.0×10^6 and 1.4×10^6 volts/cm, respectively. The resistance abruptly decreased upon application of the electric field, increased slightly with time while the field was constant, and increased abruptly upon removal of the field regardless of the direction of the field. The abrupt increase was usually somewhat less than the abrupt decrease. The abrupt resistance changes satisfied the equation $\Delta R=\alpha E^{\frac{1}{2}}$, where ΔR is resistance change, E is applied field in volts/cm$\times10^{-6}$, and $\alpha=0.46$. A large part of the small resistance change with constant field was due to an observed filament temperature increase resulting from bombardment by the electronic portion of the observed ion-, photo-, and field-emission current. It was found that the photo and ion currents were much larger than the field-emission currents. No observable electrostatic effect was found which could account for the abrupt resistance changes. It has not been possible to offer a theory for the observed effects.

INTRODUCTION

THE effect of a radial electrostatic field upon the resistance of tungsten wires in high vacua was reinvestigated, with lower pressures and better vacuum techniques than in the original experiments.[1–3]

Worthing[1] observed that in the absence of the electric field, the resistance of a hot (2500°K) tungsten filament increased as expected because of the evaporation of the filament. Upon application of electric field strengths greater than about 0.5×10^6 volts/cm, he observed that the resistance abruptly decreased and simultaneously the time-rate of increase of resistance became less than for the no-field case. Estabrook,[2] employing molybdenum filaments at temperatures of 1462 and 1644°K, observed an abrupt increase in resistance upon application of the field along with a lessening of the time-rate of increase of resistance due to evaporation. Vissat,[3] employing tungsten in the temperature range 293°K to 853°K, observed an abrupt increase in resistance upon application of the field, agreeing with Estabrook but not with Worthing. He did not investigate the time rate of change of resistance. In each of these experiments the electric field was applied in cylindrical geometry with the filament positive with respect to the surrounding plate so as to inhibit electron emission from the filament. The pressures ranged from 10^{-5} to 10^{-6} mm Hg obtained with dynamic vacuum systems containing waxed joints. The systems probably were not very well outgassed. The filaments were dc heated, resulting in dc etch[4] of the surfaces and consequently the calculated electric field strengths were inaccurate. (Worthing and Estabrook employed Wimshurst machines as high-voltage sources for their fields.)

There appear to be only two theoretical papers concerning this effect.[5,6] Greibach[5] examined the problem from the thermodynamical point of view and was able to obtain qualitative agreement with Worthing but his calculated values for the resistance changes were but 1 percent of Worthing's experimental values. He attributed this to his assumption of an ideal filament surface, perfectly smooth and clean. Reid,[6] from kinetic theory, supported Estabrook[2] and by proper choice of parameters was able to obtain good agreement with Estabrook's measured resistance changes. However, Vissat's results[3] disagreed with Reid's predictions for tungsten.

In the present investigation, with improved experimental apparatus and technique, more complete information has been obtained concerning the effect of strong

* Now at Jackson and Church, Saginaw, Michigan.
[1] A. G. Worthing, Phys. Rev. **17**, 418 (1921).
[2] G. B. Estabrook, Phys. Rev. **63**, 352 (1943).
[3] P. L. Vissat, Phys. Rev. **64**, 119 (1944).
[4] C. Herring and M. H. Nichols, Revs. Modern Phys. **21**, 185 (1949), p. 99 ff. See also D. B. Langmuir, Phys. Rev. **89**, 911 (1953).
[5] E. H. Greibach, Phys. Rev. **33**, 844 (1929).
[6] W. P. Reid, Phys. Rev. **63**, 359 (1943).