# Ferromagnetism at Very High Frequencies. III. Two Mechanisms of Dispersion in a Ferrite

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The magnetic spectrum of a ferrite is shown to contain two regions of pronounced dispersion. One occurs at radiofrequencies, resembles a resonance, and is proved to be due to domain wall displacements; the other occurs in the microwave range, exhibits typical resonance characteristics, and is attributed to domain rotations. The identification of the dispersion mechanisms is based primarily on a comparison of the complex permeability of solid samples with that of small particles of the same material. Several manifestations of single-domain behavior are reported, and a study of remanence by high frequency methods is presented. The wall effects are interpreted in terms of the concept of apparent wall inertia and it is shown that microscopic eddy currents cannot cause the damping. The rotational effects are interpreted on the basis of crystalline anisotropy. Previously used methods for identifying dispersion mechanisms are discussed.

## I. INTRODUCTION

NUMBER of experiments have shown that in several ferromagnetic oxides a pronounced dispersion of the permeability occurs at radio frequencies<sup>1, 2, \*</sup> or in the microwave range.<sup>3,4</sup> In all these experiments only one region of pronounced dispersion was observed in the magnetic spectrum of each substance studied, although the curve of Welch et al.4 contains one experimental point below 300 Mc/sec. These dispersions were considered to be indicative of a broad resonance and were interpreted on the basis of the domain rotation theory originated by Landau and Lifshitz.<sup>5</sup> Their paper contains the fundamental idea that ferromagnetic anisotropy is equivalent to an internal magnetic field capable of producing a Larmor precession of the electron spins responsible for the spontaneous magnetization. Resonance absorption is to be expected, therefore, when the frequency of an applied alternating field equals the Larmor precession frequency.

While investigating the magnetic spectrum of several ferromagnetic ferrites we ascertained that in general there are *two* regions of pronounced magnetic dispersion at high frequencies. In "ferramic A,"<sup>6</sup> a substance which we have studied in some detail, one region occurs at radiofrequencies ( $\sim$ 50 Mc/sec.) and resembles a broad resonance rather than a relaxation. The other region occurs in the microwave range ( $\sim 2500$  Mc/sec.) and has the appearance of a typical resonance. In the present paper the mechanisms responsible for the two dispersion regions are identified. This identification is based mainly on a comparison of the measured magnetic spectrum of ferramic A with that of very small particles of the same material. Additional evidence used in reaching our conclusions includes measurements of coercive force, the magnetic spectrum in the remanent state, as well as certain x-ray data and electron-microscopic observations.

Our experiments prove that in ferramic A the microwave resonance is caused by domain rotations (Larmor resonance) whereas the radiofrequency dispersion is primarily caused by domain wall displacements. These conclusions differ from the current view which is based on the observation of a single dispersion region and considers rotational processes solely responsible for the magnetic<sup>7</sup> losses in ferrites at frequencies above 1 Mc/sec. However, the existence of wall displacement effects at radiofrequencies is not unexpected. A theoretical treatment of such effects was given by Landau and Lifshitz<sup>5</sup> in the same article which contains the basic theory of domain rotations mentioned above. In paper II of the present series,8 moreover, it was shown that in iron there are some experimental indications for wall displacements at 200 Mc/sec. even though the observed permeabilities are largely due to domain rotations, as first noted<sup>9</sup> in 1947.

Toward the end of this paper we shall discuss the methods used by other investigators in interpreting their experiments<sup>10</sup> on the magnetic dispersion in ferrites, and suggest possible reasons for the lack of evidence concerning wall displacements. In addition, we shall consider some predictions made by Döring<sup>11</sup> in his theoretical work on the apparent inertia of domain walls and show that our results provide some experimental confirmation for his ideas. It appears that

<sup>&</sup>lt;sup>1</sup> J. L. Snoek, Physica 14, 207 (1948); Nature 160, 90 (1947). <sup>2</sup> Brockman, Dowling, and Steneck, Phys. Rev. 77, 85 (1950). \* In the present paper radiofrequencies are not understood to

include microwave frequencies. <sup>a</sup> J. B. Birks, Proc. Phys. Soc. London B 63, 65 (1950). This

paper contains references to Birks' earlier communications. <sup>4</sup> Welch, Nicks, Fairweather, and Roberts, Phys. Rev. 77, 403 (1950).

<sup>&</sup>lt;sup>5</sup> L. Landau and E. Lifshitz, Physik. Zeits. Sowjetunion 8, 153 (1935).

<sup>&</sup>lt;sup>6</sup> Trade name for an iron-magnesium ferrite purchased from the General Ceramics and Steatite Corporation. A chemical analysis of the material used in our experiments gave the following composition: 45 percent Fe, 14 percent Mg, 3 percent Mn, 2 percent Ca, 1 percent Zn; the balance is presumably oxygen.

<sup>&</sup>quot;We are not concerned with losses due to eddy currents and hysteresis.

<sup>&</sup>lt;sup>8</sup> M. H. Johnson and G. T. Rado, Phys. Rev. 75, 841 (1949).

<sup>&</sup>lt;sup>9</sup> Johnson, Rado, and Maloof, Phys. Kev. 71, 322 (1947).

<sup>&</sup>lt;sup>10</sup> For a general discussion of ferromagnetic phenomena at microwave frequencies, see G. T. Rado, *Advances in Electronics* (Academic Press, Inc., New York, 1950), Vol. II, p. 251.

<sup>&</sup>lt;sup>11</sup> W. Döring, Zeits. f. Naturforschung 3a, 374 (1948).

TABLE I. Ferramic A at 1750 Mc/sec. All three sample dimensions are in inches. The dielectric constant,  $\epsilon_1 - i\epsilon_2$ , was measured at eight frequencies (fairly uniformly spaced) in the region of 50 to 10,000 Mc/sec. Throughout this frequency region  $\epsilon_1$  had a constant value of  $8.5 \pm 10$  percent;  $\epsilon_2$  was small throughout this region and did not exceed 0.7.

Outer diameter	Inner diameter	Thickness	μ1	μ2	€1
1.5	0.625	0.108	1.36	2.25	8.9
1.5	0.625	0.025	1.31	2.14	8.9
0.5	0.219	0.025	1.47	2.14	8.2
0.5	0.219	0.115	1.31	2.42	

Döring's mechanism of resonance absorption by domain wall displacements is a possible explanation for our 50 Mc/sec. dispersion. We believe, however, that the high electrical resistivity ( $\sim 10^9$  ohm-cm) of our specimens excludes the possibility, proposed by Becker<sup>12</sup> and Döring, of attributing the fundamental damping processes to the action of microscopic eddy currents.

## **II. METHOD OF MEASUREMENT**

At frequencies between 3.5 and 10,000 Mc/sec. we determined the complex permeability,  $\mu = \mu_1 - i\mu_2$ , and the complex dielectric constant,  $\epsilon = \epsilon_1 - i\epsilon_2$ , by means of coaxial line measurements. Since in ferramic A the skin effect is negligible, the elaborate method and technique<sup>8, 9, 13</sup> developed for the study of metals is not applicable in this case. Instead, we used a simple standing wave method which was developed by O. Halpern<sup>14</sup> and M. H. Johnson with particular reference to thin samples of magnetic insulators and dielectrics.

Our samples were toroids of rectangular cross section, and their radii were so chosen as to match those of the coaxial lines used. The permeabilities were determined by inserting a sufficiently thin sample into the line at a position adjacent to the shorted end, and measuring (1) the voltage standing wave ratio,  $\xi$ , and (2) the displacement, s, of the voltage minimum from its (previously observed) location in the shorted empty line. At the lowest frequencies the line had to be extended by means of coaxial cables. It may also be noted that  $\xi$  was obtained in the usual manner from a measurement of the width of the standing wave pattern near the voltage minimum. If the line losses are neglected it can be shown that

$$\mu_1 - 1 = s/L \tag{1}$$

$$\mu_2 = 1/\xi k_0 L, \qquad (2)$$

where L is the thickness of the sample,  $k_0 = 2\pi/\lambda_0$  is the propagation constant in the empty line, and  $\lambda_0$  is the free-space wave-length. The dielectric constants were determined by placing the sample at a distance of  $\lambda_0/4$ from the shorted end of the line, measuring  $\xi$  and s, and

using Eqs. (1) and (2) with  $\mu_1$  replaced by  $\epsilon_1$ , and  $\mu_2$ replaced by  $\epsilon_2$ .

In order that these equations apply, the quantity  $|\epsilon\mu|(k_0L)^2$  must be negligible compared to unity. This requirement was fairly well fulfilled by proper choice of the sample thicknesses used at the various frequencies. These thicknesses varied from 0.025 to 0.440 inch as the frequency was decreased. To make sure that no serious errors arose we checked our measurements on two (or more) suitable values of L at most frequencies. The resulting permeabilities agreed within the experimental error ( $\pm 5$  percent except for low frequency values of  $\mu_2$ ) and were averaged before being plotted on the final graphs. We also proved that the measured permeabilities are relatively independent of the radial dimensions of the samples. For this purpose we used two coaxial lines characterized by different radii. Table I contains data taken at 1750 Mc/sec. and shows, by means of an example, that our results are independent of all three sample dimensions. Similar comparisons were made at 50 and 3000 Mc/sec. and yielded equally satisfactory results. These experimental precautions are emphasized because they show that our data refer to intrinsic properties of the samples rather than to spurious effects. This point is particularly important since Brockman, Dowling, and Steneck<sup>2</sup> have called attention to the effect of "dimensional resonance" on the dispersion in ferrites.



FIG. 1. Magnetic spectrum of solid ferramic A in the demagnetized state. Data were taken on coaxial lines (circles) and on a G. R. 821A bridge (rectangles). The value of  $(\mu_{stat}-1)$  is 18.6.

and

<sup>12</sup> R. Becker, Physik. Zeits. 39, 856 (1938); Ann. d. Physik (5) 36, 340 (1939). <sup>13</sup> Rado, Johnson, and Maloof, Rev. Sci. Inst. 20, 927 (1949).

<sup>&</sup>lt;sup>14</sup> O. Halpern, M.I.T. Radiation Laboratory Report VII-14S (1942), and unpublished work by M. H. Johnson.

The magnitude of the alternating magnetic field might also be expected to cause uncertainties in the results. However, the coaxial line measurements did not show any field dependence of the permeabilities, so that only *reversible* effects were detected. As an example, we cite measurements at 50 Mc/sec. where both  $\mu_1$  and  $\mu_2$  were constant within  $\pm 3$  percent for a field variation of 50 decibels.

At frequencies between 0.5 and 3.0 Mc/sec. we determined  $\mu_1$  by means of inductance measurements with a General Radio "twin-T impedance-measuring circuit, Type 821A." The samples were toroids of rectangular cross section and were wound with a convenient number of turns of insulated wire arranged in a single layer. Similar coils with a polystyrene core made it possible to check the inductance measurements. Since  $\mu_2$  appeared to be small we did not attempt to determine it accurately by measuring the high frequency resistance of the coils and separating out the eddy current effects due to the windings. It will be seen in the next section that in the frequency range of interest all of the results are based on coaxial line data; the inductance measurements merely served to extend the results to lower frequencies.

Similar inductance measurements on a Maxwell bridge were used to determine  $\mu_1$  at frequencies between 1 and 25 kc. The static reversible permeability,  $\mu_{\text{stat}}$ , was obtained by a slight extrapolation ( $\approx 1$  percent) of the  $\mu_1$  values to zero frequency and zero alternating field. This correction is not significant since it is about equal to the difference between the  $\mu_1$  values measured on different samples.

#### **III. RESULTS AND INTERPRETATION**

#### A. The Magnetic Spectrum

Figure 1 shows the complex permeability of ferramic A as a function of frequency. The measurements refer to the demagnetized state and cover the complete magnetic spectrum of this material. Up to a frequency of about 1 Mc/sec. the permeability is approximately constant and practically equal to the static value,  $\mu_{stat}$ . At higher frequencies  $\mu_2$  reaches a conspicuous maximum in a region ( $\approx 50$  Mc/sec.) where  $\mu_1$  decreases abruptly. It is interesting that  $\mu_1$  also passes through a maximum and becomes, in fact, larger than  $\mu_{stat}$ . This indicates that the r-f dispersion is probably a resonance rather than a relaxation. However, the value of  $(\mu_1 - 1)$ does not become negative at the minimum between the two dispersions. We cannot account for this fact by a simple superposition of two resonance curves; or by a magnetic interaction of the two mechanisms. At microwave frequencies the situation is again straightforward. Both  $\mu_2$  and  $(\mu_1 - 1)$  exhibit a typical resonance behavior (including negative values of  $(\mu_1 - 1)$ ) before approaching, in the vicinity of 10,000 Mc/sec., the value zero.

It is pertinent to inquire into the mechanisms re-

sponsible for the two observed dispersion regions. On the basis of the theoretical considerations presented in the following section we believe that the microwave dispersion is due to the now well-known mechanism of domain rotation resonance in the anisotropy field of the material. The r-f dispersion, on the other hand, may arise from domain wall displacements, or less likely, from domain rotations in those parts of the material where the anisotropy is conceivably very low. We shall now describe the experiments which proved the first alternative to be correct.

#### B. Identification of the Dispersion Mechanisms

In references 8 and 9 it was shown that the use of a static magnetic field parallel to the r-f field provides a possible method for distinguishing between the (relatively field-dependent) wall displacements and the (relatively field-independent) domain rotations in iron. Unfortunately, this method cannot be applied to ferramic A because the r-f dispersion could, in principle, be interpreted as a rotational resonance due to an internal field of about 16 oersteds, corresponding to a Larmor frequency of about 45 Mc/sec. But this field is comparable to the coercive field of the material ( $\approx 3$  oersteds) so that rotations and wall displacements should be about equally field-dependent in the frequency range considered here.

We performed what appears to be a conclusive identification by using the results of the following alternative experiment. Ferramic A (which is a ceramic) was ground up in a ball mill and the relatively large particles were separated from the powder and discarded. The small particles were mixed with paraffin and molded to produce toroidal samples suitable for the coaxial lines and impedance measuring circuits described above.



FIG. 2. Magnetic spectrum of 70 percent (by weight) mixture of ferramic A powder and wax. Data were taken on coaxial lines (circles) and on a G. R. 821A bridge (rectangles). The value of  $(\mu_{stat}-1)$  is 0.65.

Several solidified samples were then used in measuring the magnetic spectrum of a particular mixture containing 70 percent ferramic A by weight (36 percent by volume).

The results, plotted in Fig. 2 show that the r-f dispersion is now absent whereas the microwave resonance is still present. Similar results were obtained with a mixture of somewhat higher concentration (80 percent by weight). These facts clearly indicate that the r-f dispersion, unlike the microwave resonance, cannot be due to ferromagnetic anisotropy or any other fundamental property of the magnetic substance. Consequently the r-f dispersion cannot arise from domain rotations and must be attributed to wall displacements. It should be emphasized that the material used in the powder-wax mixture (Fig. 2) was obtained from the solid material (Fig. 1) without using a heat treatment or changing the chemical composition. The only factors capable of causing differences between the two sets of samples are particle size and shape, magnetic interactions, internal stresses, and possibly small impurities. Our argument is not concerned with these factors because it does not depend on the reason for the absence of the r-f dispersion in the powdered material; the only essential point is the fact of this absence combined with the presence of the relatively unmodified microwave resonance. It is nevertheless interesting to examine the factors mentioned above, and to consider their effects on the two dispersion mechanisms.

### C. Factors that Influence the Dispersions

We shall discuss the *microwave resonance* first. In comparing this resonance in the cases of Figs. 1 and 2 it is necessary to take into account the change in volume concentration from 100 to 36 percent. After this correction is applied the magnitude (and width) of the microwave resonance is quite similar in the two cases if one disregards the peak of  $(\mu_1-1)$  at 1100 Mc/sec. in Fig. 1; it seems possible that this peak is due to the presence of the r-f dispersion. However, the frequency where  $(\mu_1-1)$  is zero (i.e., the frequency of the undamped resonance) turns out to be only twothirds as large in the solid material (2000 Mc/sec.) as in the powdered material (3000 Mc/sec.). This effect may be due to magnetic interactions or to internal stresses.

The interactions between the crystallites of the solid material are very complicated because of wall effects and we shall not attempt to discuss them. It is possible, however, to make a prediction with regard to the powder-wax mixtures. The magnetic interactions between the particles undoubtedly become stronger as the concentration increases. As a result, one may expect<sup>15</sup> the "observed" (or effective) anisotropy of the particles to decrease with increasing concentration. The true crystalline anisotropy should correspond to the limiting case where the particles (assumed to be single crystals as well as single domains) are infinitely separated. Since the resonance frequency is proportional to the effective anisotropy, this explanation is in qualitative agreement with the following observations. We found that in ferrite-wax mixtures containing various concentrations of particles (whose internal stress was presumably constant) the microwave resonance frequency decreases with increasing concentration. This situation is similar to the known dependence<sup>15,16</sup> of the coercive force on the concentration of single-domain particles embedded in a non-magnetic matrix.

The role of the internal stresses is difficult to assess. The stresses in the solid material are small because the observed x-ray diffraction lines are sharp; the stresses in the particles, on the other hand, could not be estimated because the diffuseness of the observed lines is largely due to the smallness of the particle size mentioned below.

It remains to discuss the r-f dispersion. The most probable explanation for the absence of domain wall effects in the powdered material is that most of the particles are sufficiently small to be single domains. Electron-microscopic observations showed that the large majority of the particles is smaller than 0.4 micron in diameter. This value is to be compared with the approximate theoretical estimate of 0.4 micron for the critical diameter characterizing single domain behavior in ferramic A. Our estimate is based on Eq. (6.1.10) of Kittel's<sup>15</sup> review because the critical diameter exceeds the wall thickness [to be calculated in Section IVB] in this material. We used the following constants: saturation magnetization  $(M_s = 99)$ ; Curie temperature  $(T_c \approx 550^{\circ} \text{K})$ ; spacing between the magnetic ions  $(a \sim 4)$  $\times 10^{-8}$  cm); anisotropy constant ( $K \sim 4 \times 10^4$  ergs/cm<sup>3</sup>). The wall energy per unit area was obtained from the approximate expression  $(KkT_c/a)^{\frac{1}{2}}$ , where k is Boltzmann's constant.

These considerations indicate that the particles are likely to be single domains. To obtain further evidence we determined the coercive force of the 70 percent ferramic A-wax mixture. The measured value ( $H_c \approx 50$ oersteds) is considerably larger than that of the solid material ( $H_c \approx 3$  oersteds) but decidedly smaller than the value  $(\sim K/M_s)$  expected on the basis of singledomain behavior. However, it is known<sup>15, 16</sup> that the coercive force of a matrix containing single-domain particles does decrease rapidly with concentration, and it is also known<sup>15</sup> that complicated effects occur if single-domain behavior is not quite reached. Additional evidence consistent with the single-domain interpretation is provided by the fact that the static permeability of the 70 percent mixture changes but little (<1 percent) with the applied field even if the latter is as large as 30 oersteds. Since a very small amount of hysteresis was detected, however, some of the particles

<sup>&</sup>lt;sup>15</sup> C. Kittel, Rev. Mod. Phys. 21, 541 (1949).

<sup>&</sup>lt;sup>16</sup> L. Weil, Comptes Rendus (Paris) 225, 229 (1947).

may have transition properties between single-domain and ordinary behavior. It was also found that the static reversible permeability of the 70 percent mixture was the same at remanence as in the demagnetized state.

We can summarize the situation by stating that all of the evidence presented here favors the single-domain interpretation. It is nevertheless conceivable that the observed effects are due to internal stresses in the small particles; but this possibility is unlikely since ferramic A is rather brittle. A decisive experiment capable of demonstrating single-domain behavior in some substances has recently been reported by Kittel, Galt, and Campbell,<sup>17</sup> who have used nickel particles. However, their method is not applicable to ferramic A because in this substance the quantity  $K/M_s$  is not smaller than, but is comparable to,  $4\pi M_s/3$ . In view of this uncertainty we feel justified in mentioning again that our identification of the r-f dispersion as a domain wall effect does not depend on the single-domain interpretation or on any other detailed theoretical model.

### D. Study of Remanence

Numerous ferromagnetic substances, possessing a sufficiently high anisotropy and reasonably small internal stresses, are known to have the interesting property that the remanence,  $M_r$ , of a polycrystal is roughly equal to half the saturation magnetization,  $M_s$ . The standard explanation<sup>18</sup> of this experimental fact may be formulated as follows. In a demagnetized polycrystal the magnetization vector of each domain is nearly parallel to one of the easy directions of magnetization determined by the crystalline anisotropy. Since in an isotropic polycrystal the axes of the crystallites are oriented at random, it follows that the magnetization vectors can point in all possible directions of the solid angle  $4\pi$ . Consequently the net magnetization of the polycrystal vanishes. It may be noted that on account of internal demagnetizing fields and internal stresses the foregoing statement does not imply a truly random orientation of the magnetization vectors. The application of a sufficiently strong magnetic field causes the magnetization vectors to line up with the field and results in "technical saturation" of the polycrystal. Upon removal of the field the polycrystal assumes the state of remanence. This state must correspond to a minimum in the energy and is reached as a result of domain rotations and wall displacements. The distribution of the vectors is assumed to extend throughout that solid angle of  $2\pi$  which includes the direction of the previously applied field and is located symmetrically with respect to it. To specify this distribution more accurately it is useful to introduce the angle  $\theta$  between the field and an arbitrary magnetization vector. In terms of this angle the above assumption means that each value of  $\theta$  larger than  $\pi/2$  occurring in the demagnetized state is replaced by  $(\pi - \theta)$  at remanence. An elementary calculation can now be used to show that the above assumption leads to  $M_r = 0.5 M_s$ .

It should be pointed out that the remanence problem in "normal" materials (defined by  $M_r \approx 0.5 M_s$ ) is not necessarily as straightforward as this simple theoretical model indicates. In view of the existence of internal demagnetizing fields (which were used by Bozorth<sup>19</sup> to discuss materials where  $M_r \ll 0.5 M_s$  it is clear that even in normal materials the reduction of the field from saturation toward remanence could, in principle, cause  $180^{\circ}$  wall displacements and thus lead to  $M_r$  $< 0.5M_s$ . Wall displacements of this type are not considered in the simple theory because internal demagnetizing fields are neglected so that, even in the presence of stresses, the directions  $\theta$  and  $(\pi - \theta)$  are



FIG. 3. Magnetic spectrum of solid ferramic A in a remanent state. Data were taken on coaxial lines (circles) and on a G. R. 821A bridge (rectangles). The value of  $(\mu_{stat} - 1)$  is 10.9.

energetically equivalent; consequently  $\theta$  never exceeds  $\pi/2$  as the field is decreased from saturation ( $\theta = 0$ ) toward remanence. The fact that the relation  $M_r$  $=0.5M_s$  is so closely fulfilled in many materials shows that the assumption used in the simple theory represents a good approximation. It is therefore interesting to discuss the following investigations which demonstrate by an independent experiment the remarkable applicability of the model described above.

The simple theory leads to the two following predictions for the (reversible) complex permeability. (1) If  $\mu$  is solely due to domain rotations then the value of  $\mu$  at remanence should be equal to that in the demagnetized state; this statement is based essentially on the equality of  $\sin^2\theta$  and  $\sin^2(\pi-\theta)$ . (2) If  $\mu$  is *primarily* due to wall displacements then the value of  $\mu$  at remanence should be substantially smaller than that in the demagnetized state; this statement is based on the (deduced) presence of fewer types of "non-180°-walls" at remanence than in the demagnetized state. These

<sup>&</sup>lt;sup>17</sup> Kittel, Galt, and Campbell, Phys. Rev. **77**, 725 (1950). <sup>18</sup> R. Becker and W. Döring, *Ferromagnetismus* (Verlag. Julius Springer, Berlin, 1939), see pp. 111, 120, 160.

<sup>&</sup>lt;sup>19</sup> R. M. Bozorth, Zeits. f. Physik 124, 519 (1948).

predictions may be tested on ferramic A because in this material  $M_r/M_s \approx 0.6$  and is thus reasonably close to the ideal value of 0.5. We now know, moreover, that the microwave resonance is due to domain rotations whereas the r-f dispersion is primarily due to wall displacements. Consequently the complex  $\mu$  at microwave frequencies should be the same at remanence as in the demagnetized state. Throughout the r-f region, on the other hand,  $\mu$  should be substantially smaller at remanence than in the demagnetized state.

A comparison of the experimental results given in Fig. 1 (demagnetized state) and Fig. 3 (remanence) shows that the theoretical predictions are confirmed in a satisfactory manner. If we now reverse the procedure mentioned above by *assuming* the correctness of the theoretical model of remanence, then the experimental data of Fig. 3 may be regarded as *verifying* our identification of the two dispersion mechanisms.

It is appropriate to mention some experimental details in connection with the magnetic spectrum at remanence. The solid samples of ferramic A rather than the mixtures were used. They were magnetized by passing a large d.c. current briefly ( $\sim 5$  sec.) through a brass rod coaxial with the toroids. In all cases the field was at least 14 oersteds at the outer radius of the sample. Demagnetization was accomplished in a similar way by using a large 60 c.p.s. current. The direction of these fields in the samples was evidently parallel to that of the high frequency field used in the coaxial line. To make sure that the static field was sufficient to saturate the samples we performed the following test. A large d.c. current producing a field of 11 oersteds at the outer radius of the sample was passed through the inner conductor of the coaxial line (using proper precautions to avoid shunting) and  $\mu_1$  was measured while the current was on. At the frequency used for this test (15 Mc/sec.) we obtained  $\mu_1 = 1$  so that the field was indeed sufficient to produce true remanence.

## IV. COMPARISON WITH THEORY

### A. Domain Rotations

In all previously published experiments the single observed dispersion was interpreted by means of relations derived from the domain rotation theory of Landau and Lifshitz.<sup>5</sup> Their fundamental result for the resonance frequency,  $\nu_0'$ , is contained in the equation<sup>20</sup>

$$\nu_0' = (\gamma/2\pi)H_i, \tag{3}$$

where  $H_i$  is the effective internal field due to anisotropy and  $\gamma$  denotes the magneto-mechanical ratio. The numerical value of  $\gamma/2\pi(=ge/4\pi \text{ mc})$  is 2.80 Mc/sec. oersted for a free electron spin (g=2); here g is the Landé splitting factor and e/mc is the specific electronic charge. Landau and Lifshitz were concerned with the special case of a uniaxial crystal exposed to an alternating field applied normal to the preferred axis. When expressed in current notation, their resonance condition (Eq. (41) of their paper) is

$$\nu_0' = (\gamma/2\pi)(2K/M_s),$$
 (4)

where the symbols have the same meaning as elsewhere in this paper.

Snoek<sup>1</sup> applied Eq. (3) to a polycrystal containing randomly oriented crystallites and obtained

$$\nu_0' = (\gamma/2\pi)(M_s/\chi_0')(2/3), \tag{5}$$

where  $\chi_0'$  is the static initial susceptibility for rotations. He found that Eq. (5) (with the measured  $\chi_0$  instead of  $\chi_0'$ ) agrees within a factor of about two with a resonance frequency observed in the r-f region, and gave several possible reasons (neglect of internal stresses, damping, and magnetic interactions) for the residual discrepancy. However, Snoek simply assumed the absence of wall displacements and did not justify using the measured  $\chi_0$  in place of  $\chi_0'$  in Eq. (5). It is therefore conceivable that the resonance discovered by Snoek is due to wall displacements rather than to rotations. Similar remarks apply to the interpretation of the results of Brockman et al.<sup>2</sup> who found a resonance at about 4 Mc/sec. after correcting their data for dimensional effects.

To illustrate the above statements we consider an example taken from our measurements. For ferramic A the measured values of  $M_s$  and  $\chi_0$  are 99 and  $18.6/4\pi$ , respectively; thus Eq. (5) yields a resonance frequency of 124 Mc/sec. if  $\chi_0$ , rather than  $\chi_0'$ , is used. It is interesting that a resonance frequency of this order of magnitude ( $\approx 45$  Mc/sec.) was actually observed (Fig. 1); but this approximate agreement is obviously a coincidence since the 45 Mc/sec. dispersion is definitely due to wall displacements rather than to rotations (Section IIIB). If we had not made microwave measurements, therefore, we might have concluded incorrectly that the 45 Mc/sec. resonance is due to rotations.

The microwave resonance ( $\sim 1300$  Mc/sec.), on the other hand, might be interpreted with Eq. (5) (as written) but this procedure would be of questionable value because  $\chi_0'$  is not known. The measured  $\chi_0$ contains a contribution  $\chi_0^{\prime\prime}$  due to the wall effects (so that  $\chi_0 = \chi_0' + \chi_0''$  and there is no unique way of separating  $\chi_0'$  from  $\chi_0$  on the basis of Fig. 1 alone. It is possible, however, to use Eq. (5) in connection with the data of Fig. 2. Here the measured static initial susceptibility  $(=0.65/4\pi)$  of the mixture is solely due to rotations. Assuming for this purpose, that  $\chi_0'$  is inversely proportional to the volume concentration (36 percent),  $\chi_0' = 0.14$  is obtained. The resonance frequency  $\nu_0'$  calculated this way is 1300 Mc/sec. and should be compared with the value 2500 Mc/sec. measured at the peak of  $\mu_2$  in Fig. 2. It seems probable that this discrepancy is due to the factors mentioned but neglected by Snoek in deriving Eq. (5).

The interpretation of the resonance shown in Fig. 2

 $<sup>^{20}</sup>$  We adopt the convention of denoting quantities characteristic of domain rotations by a single prime (') and quantities characteristic of domain wall displacements by a double prime (").

may be based on Eq. (4) as well. This equation should apply to cubic crystals just as accurately as to uniaxial ones because the anisotropy energy density is the same  $(=K\phi^2)$  in both cases, provided the displacement,  $\phi$ , of a domain magnetization vector from an easy direction is small. On this basis we obtain  $K=4.4\times10^4$  ergs/cm<sup>3</sup> from the resonance at 2500 Mc/sec. This value represents an "effective" anisotropy constant (as mentioned in Section III) because demagnetizing effects have not been considered. The true anisotropy constant may differ from the above value by a factor of perhaps 2 or 3 but for our present purposes (Sections IIIC and IVB) only the order of magnitude of K is needed; the sign of K is not determined. (If K is negative,  $H_i = -4K/3M_s$ .)

It should be mentioned that Birks<sup>3</sup> reported a microwave resonance in several ferrite-wax mixtures. He attributed the effect to domain rotations because the measured permeabilities were relatively insensitive to a static magnetic field applied at an arbitrary angle with respect to the alternating field. In interpreting his data he used the relations of Landau and Lifshitz [Eq. (4)] and Snoek [Eq. (5)]. Birks' results represent the properties of a hypothetical substance obtained by extrapolating measurements performed on ferrite-wax mixtures. The r-f dispersion which we observed in the solid samples (Fig. 1), and proved to be due to wall effects, does not involve any extrapolation. We did not observe this dispersion even in our 80 percent mixture which represents a volume concentration of 49 percent and exceeds the highest concentration used by Birks.

### B. Domain Wall Displacements

The first theoretical treatment of domain wall displacements at high frequencies was given by Landau and Lifshitz.<sup>5</sup> These authors attribute the damping of the wall motion to a certain interaction between the spins. They refer to this interaction as "relativistic interaction" and describe it by a phenomenological constant which is denoted<sup>21</sup> by  $\eta$  and has the same dimensions as  $M_s$ . Their treatment explicitly omits any consideration of hysteresis effects and results in a purely imaginary susceptibility which varies inversely with the frequency.

A somewhat different theory was proposed by Döring.<sup>11</sup> He shows that the motion of a domain wall requires, in general, the existence of an internal demagnetizing field. This field modifies the spin distribution and causes the energy of a moving wall to exceed that of the same wall at rest by a term proportional to the square of the wall velocity. The proportionality constant is interpreted by Döring as an apparent mass (or inertia) characterizing the moving wall. Since Döring (unlike Landau and Lifshitz) does take into account the restoring force acting on a wall, the apparent mass leads him to predict that resonance phenomena will occur when any given type of wall is exposed to an alternating field of the correct frequency.

Neither of these theories has yet been compared quantitatively with the experimental data<sup>1-4</sup> cited although Döring has tentatively suggested that Birks' results may be due to wall displacements as well as to rotations. Some of our experimental results, however, may be used for such a comparison because the r-f dispersion in ferramic A was shown to be primarily a wall effect. In the following presentation we shall follow Döring's theory but treat the damping according to the mechanism of Landau and Lifshitz.

The displacement, x, of an "average" domain wall is given by the equation of motion

$$m(d^2x/dt^2) + \beta(dx/dt) + \alpha x = M_s H, \qquad (6)$$

where *m* is the apparent mass per unit wall area and  $H = H_0 \exp(i\omega t)$  is the applied field; the constants  $\alpha$  and  $\beta$  will be discussed below. Using the abbreviations

$$\omega_0^{\prime\prime} = (\alpha/m)^{\frac{1}{2}} \tag{7}$$

$$\omega_c^{\prime\prime} = \alpha/\beta,\tag{8}$$

as well as the definition of the susceptibility due to wall displacements

$$\chi^{\prime\prime} = x M_{s} / H d, \qquad (9)$$

one obtains from Eq. (6)

and

where

$$\chi'' = \frac{\chi_0''}{1 - (\omega/\omega_0'')^2 + i(\omega/\omega_c'')},$$
 (10)

.,

$$\chi_0^{\prime\prime} = M_s^2 / \alpha d \tag{11}$$

is the static susceptibility and d denotes the average domain size. (We omitted a factor whose magnitude is between one and two on the right-hand side of Eqs. (6) and (9) because its value depends on the type of wall under consideration.) Eq. (10) is a standard form of a resonance and differs from Döring's formulation only in that the solution of Eq. (6) is expressed in terms of  $\chi''$  rather than x.

We consider the resonance frequency first. The "stiffness constant,"  $\alpha$ , measures the restoring force and is given by Eq. (11) in terms of  $\chi_0$ ". The apparent mass, m, was calculated by Döring and represents the fundamental contribution of his theory. Neglecting a factor of the order of unity, we may write his result<sup>22</sup> for 90° walls in the simple form‡

$$m = 1/8\pi\gamma^2\delta,\tag{12}$$

where  $\delta$  is the wall thickness parameter and is given by  $\delta \sim (kT_c/Ka)^{\frac{1}{2}}$ ;  $\gamma$  denotes the magneto-mechanical ratio, as before. Combining Eqs. (7), (11) and (12), we obtain

$$\nu_0'' = (\gamma/2\pi) (M_s/\chi_0'') (8\pi\chi_0''R)^{\frac{1}{2}}, \qquad (13)$$

<sup>&</sup>lt;sup>21</sup> Landau and Lifshitz use the symbol  $\lambda$  instead of  $\eta$ .

<sup>&</sup>lt;sup>22</sup> Döring's theory contains an expansion retaining terms to the order of  $(K/2\pi M_*^2)^2$ . In our case this square is about 0.4. ‡ This factor equals unity if the wall normal is perpendicular

to the spin directions of the adjacent domains.

where  $\nu_0'' = \omega_0''/2\pi$  is the resonance frequency due to wall displacements and R is a dimensionless ratio defined by

$$R = \delta/d. \tag{14}$$

Equation (13) was written to resemble Eq. (5) in all but the last factor. It is interesting to note that these two equations are based on completely different mechanisms and may nevertheless predict the same resonance frequency if R and the static permeability are of the order of unity; this possibility is particularly likely to arise if the measured static susceptibility, rather than  $\chi_0'$ , is used in Eq. (5). To compare Eq. (13) with our results we use  $\chi_0 = 1.48$  (see Fig. 1) because the contribution of the rotations (i.e.,  $\chi_0'$ ) is only about 0.14 as estimated on the basis of Fig. 2 and the known volume concentration of 36 percent. We obtain  $R = 1.5 \times 10^{-3}$ if we take  $\nu_0^{\prime\prime} = 45$  Mc/sec. (i.e., the peak of  $\mu_2$ ) as the experimental resonance frequency. This is a rather arbitrary choice since  $v_0^{\prime\prime}$  should be the frequency where  $(\mu_1 - 1) = 0$ ; but we shall not attach any significance to factors of about 2 in our estimated domain size. The value of  $\delta$  for ferramic A is about  $7 \times 10^{-6}$  cm. Thus Eq. (14) leads to  $d = 5 \times 10^{-3}$  cm; this appears to be of the right order of magnitude because x-ray data indicate that the crystallite size in solid ferramic A is between  $5 \times 10^{-3}$  and  $5 \times 10^{-4}$  cm.

Döring did not consider the damping due to spin-spin interaction but used Becker's<sup>12</sup> theory of microscopic eddy currents to obtain a value for  $\omega_c''$ . This mechanism does not give any appreciable damping for ferramic Abecause in this substance the resistivity ( $\rho \sim 10^9$  ohm cm) is about 10<sup>15</sup> times higher than in iron. For this reason we derive  $\omega_c''$  from the Landau-Lifshitz theory. It should be noted, however, that this is merely a formal step since the damping might influence the mass calculated by Döring; in addition, the damping (i.e.,  $\eta$ ) was introduced phenomenologically as mentioned above. Landau and Lifshitz have shown (their Eq. (29)) that

$$dx/dt = (\gamma M_s \delta/\eta) H. \tag{15}$$

Comparison of this result with Eq. (6) yields  $\beta = \eta/\gamma \delta$ , so that we obtain from Eqs. (8), (11) and (14)

$$\nu_{c}'' = (\gamma/2\pi) (M_{s}/\chi_{0}'') (M_{s}R/\eta)$$
(16)

where  $\nu_c'' = \omega_c''/2\pi$  is the critical damping frequency. From Eq. (6) it follows that the oscillation is less than critically damped if  $\omega_0'' < 2\omega_c''$ ; using Eqs. (13) and (16), this condition may be written

$$\eta/M_{s} < (R/2\pi\chi_{0}^{\prime\prime})^{\frac{1}{2}}.$$
 (17)

We are now in a position to set an upper limit on the unknown constant  $\eta$  by using the relation (17) as an equality and substituting the value  $R = 1.5 \times 10^{-3}$  derived from our r-f experiments which show a resonance effect due to walls. The resulting  $\eta/M_s$  is  $1.3 imes 10^{-2}$  and may be considered satisfactory in that the Landau-Lifshitz theory assumes  $\eta \ll M_s$ . There are, however, two difficulties. (1) As pointed out by Döring, the width of the absorption "line" due to the wall resonance is partly due to the *distribution* of  $\alpha$ -values in an actual material. Consequently the value of n derived above not only describes the basic interactions between the spins but includes all other effects that broaden the line. (2) If we arbitrarily attach a fundamental significance to an experimentally determined  $\eta$ , as is sometimes done, we find that the Landau-Lifshitz theory of spin rotations (their Eq. (40)) predicts a much sharper microwave line (on the basis of  $\eta/M_s = 1.3 \times 10^{-2}$ ) than the one we observed. It therefore appears that the width of the microwave resonance is due even more to experimental effects (inhomogeneities, particle shape, stresses, etc.) than is the r-f resonance. These considerations indicate that the theories<sup>23</sup> available at present do not explain the line widths observed in our experiments on natural ferromagnetic resonance. The resonance frequencies, on the other hand, can be accounted for on the basis of these theories.

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<sup>&</sup>lt;sup>22</sup> C. Kittel, Phys. Rev. **79**, 214 (1950), suggested independently that the dispersion in ferrites be interpreted in terms of domain rotations and wall displacements. He proposed using Landau and Lifshitz' equation (our Eq. (4)) for rotations and a relaxation frequency (similar to our Eq. (16)) for wall displacements, and showed this to be in qualitative accord with the data of Welch *et al.* (reference 4). It should be noted, however, that these data include only one point (100 Mc/sec.) below 300 Mc/sec. As mentioned by one of us during the discussion period for Kittel's talk, our experiments show a wall resonance and had been found to be consistent with Döring's theory (reference 11) of apparent wall inertia.