Ferromagnetism at Very High Frequencies. IV. Temperature Dependence of the Magnetic Spectrum of a Ferrite*

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The magnetic spectrum of a sintered ferrite, previously studied by the authors at room temperature and found to contain an rf resonance, attributed to domain wall displacements, and a microwave resonance, attributed to domain rotations, has now been observed at 77°K, 195°K, and 411°K. By using single-domain particles of this material embedded in wax, the rotational resonance was observed separately. The principal aspect of the experimental results is the fact that with increasing temperature the resonance frequencies decrease, the fractional change being about the same in all three cases and exceeding that of the saturation magnetization. A semiquantitative theoretical interpretation of this fact is given. Auxiliary experiments, performed as an aid in interpreting the magnetic spectrum, include measurements of the spectroscopic splitting factor g at various temperatures. The deviation of the apparent g factor values from g=2, as well as their temperature dependence, was found to decrease with increasing resonance field, i.e., increasing resonance frequency.

1. INTRODUCTION

N a previous paper¹ of this series it was shown that I N a previous paper- or this series is the magnetic spectrum² of a certain ferrite contains two resonances at room temperature. The corresponding dispersion mechanisms were identified by comparing, in the region between zero and 10,000 Mc/sec, the magnetic spectrum of solid samples with that of singledomain particles made from the same sintered material. It was concluded that one of the resonances is due to the Larmor precession of domains in the internal magnetic field, a mechanism previously invoked by Snoek³ and others⁴ in interpreting⁵ the single resonance (or relaxation) found by them in various substances at rf or microwave frequencies. The other resonance, however, had not been observed previously. We showed it to be due to oscillations of domain walls and thus obtained experimental support for the concept of domain wall inertia. This inertia had been calculated by Döring,⁶ and a generalized treatment was given by Rado.7

In the present paper we report on an extension of our work to temperatures other than room temperature. We begin with the measurements of the temperature dependence of the magnetic spectrum in solid samples and single-domain particles (Sec. 2A) and then describe

2. EXPERIMENTAL METHODS AND RESULTS A. Magnetic Spectrum The methods described in paper III have been

adapted to measure the complex permeability, $\mu = \mu_1$ $-i\mu_2$, as a function of frequency as well as temperature. As in the room temperature experiments, no static magnetic field was used, and precautions were taken to ensure that (1) the alternating field was sufficiently

some auxiliary experiments on the temperature dependence of the g factor (Sec. 2B) and the saturation magnetization and coercive force (Sec. 2C). These auxiliary experiments are limited in scope since they were designed primarily to aid in the interpretation of the magnetic spectrum. Nevertheless, the g factor work is of independent interest because it casts doubt on the validity of some previous measurements made on polycrystalline materials. In Sec. 3 we attempt a theoretical interpretation of the magnetic spectrum. While the polycrystalline nature of our samples makes it extremely difficult to give a quantitative treatment of the problem, it will be shown that the principal*aspects of the temperature dependence can be explained in a semiquantitative manner.

The same magnetic samples were used in the present work as in paper III. They consist of a commercial ferrite, "ferramic A," which is composed of several oxides but is primarily magnesium ferrite. The precise chemical composition of the samples is not important for our purposes because our conclusions are based on comparisons of various magnetic properties measured on the same material. It should be noted, furthermore, that the two resonances which we observed at room temperature are not a unique property of ferramic A. We have found similar effects in pure magnesium ferrite, lithium ferrite $[(Li_{0.5}Fe_{0.5})Fe_2O_4]$, and nickel ferrite prepared at this laboratory. The investigations on these and other materials will be reported at a later date

^{*} A preliminary account was given at the New York meeting of the American Physical Society, January 31, 1952 [Phys. Rev. 86, 599 (1952)].

¹Rado, Wright, and Emerson, Phys. Rev. 80, 273 (1950), referred to as III.

² The term "magnetic spectrum" is used throughout the present paper to denote the frequency dependence of the complex initial permeability. ³ J. L. Snoek, Physica 14, 207 (1948); Nature 160, 90 (1947);

J. L. Shole, Physica 14, 207 (1946), Nature 100, 50 (1947),
H. G. Beljers and J. L. Snoek, Philips Tech. Rev. 11, 313 (1949).
J. B. Birks, Proc. Phys. Soc. (London) B63, 65 (1950);
Brockman, Dowling, and Steneck, Phys. Rev. 77, 85 (1950);
Welch, Nicks, Fairweather, and Roberts, Phys. Rev. 77, 403 (1950).

⁵ A discussion of the interpretations proposed in references 3 and 4 is given in references 1 and 10.

⁶ W. Döring, Z. Naturforsch. **3a**, 374 (1948). For a simplified calculation, see R. Becker, J. phys. et radium **12**, 332 (1951); C. Kittel, Phys. Rev. **80**, 918 (1950). ⁷ G. T. Rado, Phys. Rev. **83**, 821 (1951).



FIG. 1. $\mu_1 - 1$ versus frequency for solid ferramic A at various temperatures.

small to justify referring to μ as the *initial* permeability,⁸ (2) dimensional effects were excluded (thin sample method), and (3) the effect of wave-guide types of modes (i.e., other than TEM) was negligible. The measured permeabilities represent, of course, average values of a position-dependent permeability, so that from this point of view there is no essential distinction between the high frequency case and the static case, as long as the variations in local properties caused by the domain structure of the (macroscopically isotropic) samples are on a scale which is small compared to λ , the wavelength in the material. This requirement is clearly fulfilled in our case because the thickness of each sample is small compared to λ .

The apparatus was modified for the low temperature measurements by surrounding the end of the coaxial line containing the sample with a cylindrical jacket and filling the latter with liquid air or a mixture of solid CO_2 and alcohol. Measurements with a thermocouple attached to either side of the sample showed that the temperature did not exceed that of the fixed points by more than 3°C. During the actual rf measurements, however, the thermocouple was attached only to the side of the sample that was backed by a short. To avoid

dielectric effects due to water droplets, we inserted a very thin Teflon window into the line at some distance from the sample and forced helium through this section of line. Tests showed that reflections from the window and other disturbances were negligible. The high temperature data were taken by inserting the end of the line into a furnace.

As noted in paper III, the formulas that were used for evaluating μ_1 and μ_2 are based on the assumption that the ohmic losses in the line are negligible. However, under the "thin-sample conditions" previously stated, the line losses may be taken into account by replacing Eq. (III-2) by

$$\mu_2 = \left(\frac{1}{\xi} - \frac{1}{\xi'}\right) \frac{1}{k_0 L}.$$
 (1)

In deriving this equation we assumed that the measurements are confined to the first few standing wave minima near the sample, a requirement that ensures that the line losses may be regarded as a small perturbation. As before, L is the sample thickness, k_0 the wave number in free space, and ξ the voltage standing wave ratio (VSWR). The quantity ξ' is the VSWR of the empty line and represents the effect of line losses. This correction has been applied to all our results, including those of paper III. It can be shown that the expression for μ_1 [Eq. (III-1)] is not affected by small



FIG. 2. μ_2 versus frequency for solid ferramic A at various temperatures.

⁸ For nonlinear effects in a ferrite, see J. J. Went and H. P. J. Wijn, Phys. Rev. 82, 269 (1951); Physica 17, 976 (1951). See also the comments in reference 19.

line losses because the shift of the standing wave minimum is measured relative to an empty line characterized by the same ohmic losses as the line containing the sample.

The measured magnetic spectrum of the solid samples is shown in Figs. 1 and 2 for various temperatures, the two sets of curves representing $\mu_1 - 1$ and μ_2 , respectively, as a function of frequency. It is seen that two resonances were observed at each of the four temperatures (77°K, 195°K, 300°K, 411°K) used in the experiments and that at the highest temperature the two resonances begin to merge.⁹ As stated in paper III, we attribute the rf resonance (occurring at 43 Mc/sec at 300°K) to domain walls and the microwave resonance (occurring at 1400 Mc/sec at 300°K) to domain rotations. The corresponding mean resonance frequencies, which we take to be the positions of the peaks of the μ_2 curve, will again be denoted by ω_0'' and ω_0' , respectively. Both of these frequencies are seen to decrease by a factor of about five as the temperature is increased in the range stated. This fact appears to be one of the principal aspects of the experimental results, and the appropriate numerical data will be given later (Sec. 3) in tabulated form. Another interesting aspect of the results is that the total static initial permeability (which is primarily due to domain walls) varies much more with temperature than the contribution of the rotations. It may also be noted that μ is almost independent of temperature (in the range stated) at frequencies around 200 Mc/sec, a fact which may be of some practical interest.

Finally, we note that Figs. 1 and 2 do not contain any data for frequencies less than about 5 Mc/sec, even though some measurements (in addition to those



FIG. 3. $\mu_1 - 1$ versus frequency for 70 percent (by weight) mixture of ferramic A powder and wax at various temperatures.

⁹ It will be shown in a later publication that this effect may be related to the apparent coalescence, at room temperature, of the two resonances in ferrites containing zinc. See G. T. Rado, Revs. Modern Phys. (scheduled for January, 1953, issue).



FIG. 4. μ_2 versus frequency for 70 percent (by weight) mixture of ferramic A powder and wax at various temperatures.

reported in paper III) were made in this range. This omission is due to the fact that in this range of relatively low frequencies the permeability appears to depend on the thermal history of the material in an unexpected manner. Thus, the value of $\mu_1 - 1$ at 1000 cps, which is essentially the static value, was measured to be 18.6, 12.6, and 9.8 at temperatures of 300°K, 195°K, and 77°K, respectively; these values agree satisfactorily with those obtained by extrapolating the curves of Fig. 1 to very low frequencies. However, after the material was allowed to reach room temperature again, the remeasured values of μ_1 at 300°K and 77°K were found to be about 40 percent higher than the original values, and only small changes were observed when the temperature was recycled. It appears that this effect is caused in some way by the exposure of the material to very low temperatures, because in an interval of 18 months we did not detect any changes in the 1000-cps permeability of samples that had been kept at room temperature. Although a more detailed investigation of this effect might prove interesting, it can be stated that we did not find evidence of this effect at the frequencies ($\gtrsim 5$ Mc/sec) included in the graphs of Figs. 1 and 2.

The measured magnetic spectrum of the samples containing a mixture of small particles of ferramic A(70 percent by weight) and wax is shown in Figs. 3 and 4 for various temperatures, the two sets of curves representing μ_1 —1 and μ_2 , respectively, as a function of frequency. The particles are essentially single domains, as shown in paper III. Data are given for frequencies above 200 Mc/sec only, and the single resonance observed at each temperature is attributed to domain rotations. The absence of the rf resonance in these samples, as demonstrated by the room temperature studies, has been the main reason for our conclusion that the rf resonance observed in the solid samples

TABLE I. Apparent g factors for ferramic A measured at variosu frequencies^a and temperatures.

	Frequency (Mc/sec)			
Temperature (°K)	9200	24,000		
77	2.67	2.14		
195	2.48	2.06		
300	2.26	2.01 ^h		

^a Okamura, Torizuka, and Kojima, Sci. Rep. Tohoku Univ. **A2**, 663 (1950), found that the g factor of Ni-ferrite at room temperature increases with increasing frequency. However, their samples were not saturated and the sphere sizes may have been too large. ^b It may be noted that Yager, Merritt, and Guillaud, Phys. Rev. **81**, 477 (1951), obtained g = 2.03 and g = 2.06 for two Mg-ferrite samples having different M_s . Their measurements were performed at room temperature, the frequency was 24,164 Mc/sec, and the g values represent estimated extrapolations to zero diameter spheres.

is due to domain walls. At low temperatures, however, there is seen to be an indication of a subsidiary peak in μ_2 at about 1000 Mc/sec; this peak is not spurious, and it may be due to rudimentary walls, impurities, or some unknown causes, but the effect is relatively small and will not be considered further at this time. Returning to the main effect, the mean resonance frequency, to be denoted by Ω_0' , is seen to decrease as the temperature is increased, this behavior being similar to that of ω_0'' and ω_0' . The static value (measured at 1000 cps) of $\mu_1 - 1$ was measured to be 0.65 at room temperature.

B. Spectroscopic Splitting Factor

Numerous measurements of the spectroscopic splitting factor g have been reported in the literature, and they have resulted in repeated confirmations of Kittel's formulas for the frequency of ferromagnetic resonance absorption in the presence of a strong magnetic field.^{10,11} However, only Healy¹² appears to have studied the dependence of g on temperature in ferrites, and his experiments were limited to a single frequency ($\sim 10,000$ Mc/sec). In the present work, we used less elaborate apparatus but measured the temperature dependence of g at three frequencies. Since the techniques involved in resonance experiments are well known, we shall describe but briefly our particular procedure.

The samples were small spheres¹³ of ferramic A. Four to eight spheres, appropriately separated from each other, were attached to one end plate of a transmission-type, half-wave, rectangular cavity which operated at about 3000, 10,000, or 24,000 Mc/sec and was surrounded by a suitable cooling jacket. The cavity was supplied with microwave energy and situated between the polefaces of an electromagnet (see Sec. 2C)

in such a way that the static field was perpendicular to the microwave field as well as to the direction of microwave propagation. A microwave spectrum analyzer was used as the detector at each of the frequencies mentioned. The experiment consisted of measuring the static field at which the cavity transmission was a minimum. For observations involving fields on either side of resonance, small adjustments of the frequency were made in order to prevent the small, but detectable, detuning of the cavity caused by the real part of the effective permeability. The g factor was obtained from Kittel's¹⁴ relation for a sphere, $\omega = \gamma H$, where ω and H are the values, at resonance, of the circular frequency and static field, respectively, and γ is given by ge/2mc; the numerical value of $\gamma/2\pi$ is approximately 2.80 Mc/(sec oersted) for g=2. It is well known that Kittel's formula applies only in the limiting case where the wavelength in the sample is very large compared to the sphere diameter. Since the relevant electromagnetic problem has never been solved, even to a first approximation,¹⁵ for the case of a reasonably small sphere having an anisotropic permeability and since the samples are, furthermore, inhomogeneous (on account of cavities, etc.), we shall regard the measured g values as *apparent* g factors. To test the importance of the size effect, we measured the apparent g factor at 77°K and 300°K on spheres having various sizes. At 9200 Mc/sec and 300°K, g varied by 2 to 3 percent when the sphere diameter was reduced from 0.080 to 0.010 inches.

The final results, assembled in Table I, were obtained at 77°K, 195°K, and 300°K, using 0.010-inch diameter spheres at frequencies of 9200 and 24,000 Mc/sec. Additional measurements were made at 3000 Mc/sec, but the data could not be interpreted because the material is definitely not saturated at the low resonance field required, and because the frequency falls into a region of natural resonance (see Fig. 2). It should be mentioned that even in the 9200-Mc/sec experiment the material is not completely saturated, so that the large deviation of the apparent g factor from the value g=2, as well as its pronounced temperature dependence, cannot be regarded as a basic effect. (Similarly, caution should be used in all other cases involving porous materials before ascribing undue theoretical significance to g factor measurements performed at 10,000 Mc/sec.) There is, in fact, no assurance that our 24,000-Mc/sec data refer to the true g factor, and higher resonance fields (i.e., higher frequencies) might have to be used to achieve a more complete saturation and thus elucidate this point. For our purposes (see Sec. 3), however, it is sufficient to state, on the basis of Table I, that the deviations of the *true* g factor from the value g=2, as well as the temperature dependence of g, are very small or nonexistent.

¹⁰ For a review of this subject, see C. Kittel, J. phys. et radium

^{12, 332 (1951).} ¹¹ This type of resonance is sometimes referred to as "induced" esonance to distinguish it from the "natural" resonances observedr (e.g., in Sec. 2A) in the absence of a static field. ¹² D. W. Healy, Tech. Rept. No. 135 (August 15, 1951), Cruft

Laboratory, Harvard University. See also Phys. Rev. 86, 1009 (1952)

¹³ The spheres were made by the method of W. L. Bond, Rev. Sci. Instr. 22, 344 (1951).

¹⁴ C. Kittel, Phys. Rev. 73, 155 (1948).

¹⁵ See the remarks of H. B. G. Casimir at the end of Kittel's paper, reference 10.

C. Saturation Magnetization and Coercive Force

An oscillation magnetometer, similar to that described by Griffiths and MacDonald,16 was used to measure the saturation magnetization M_s of ferramic A at 77°K, 195°K, and 300°K. The sample is a flat circular cylinder (cut from the material used for making the rings for the rf experiments), having a diameter of 0.917 inch and a thickness of 0.051 inch, and is mounted on a polished glass rod whose moment of inertia is such that the period of oscillation in the fields used (~2000 to 5500 oersteds) ranged from $\frac{1}{4}$ to $\frac{1}{2}$ second. At the low temperatures, the glass rod is arranged to oscillate in a bath of liquid air or a mixture of solid CO_2 and ether. Tests in a water bath at room temperature showed that friction effects are negligible in liquids of these viscosities. The period of oscillation is measured by reflecting a light beam from a mirror attached to the glass rod and using a photocell which is connected to a Sanborn low frequency amplifier and recorder. The electromagnet, used also in the g factor work, has a sufficiently uniform field throughout the extent of the large pole pieces (90 square inches in area). A motor-generator set supplies the current, a stability of ± 0.05 percent being assured by a Fluke regulator. By using a calibrated flip coil with a ballistic galvanometer, the field versus current relation was found to be linear in the range of interest. The reproducibility of the final results is better than ± 2 percent, while the absolute error, estimated by measuring M_s for very thin nickel disks, is less than 5 percent and seems to be partially due to the error in measuring the volume of porous samples. It therefore appears that the accuracy of the instrument is comparable to that of Griffiths and MacDonald, the disadvantage of using a liquid bath and porous samples with a relatively small axial ratio (~ 18) being compensated by the advantage of using the recorder and other improvements. The experimental results, shown in Table II, represent M_s referred to a unit volume of sample, rather than solid material.

Finally, the coercive force H_c of the samples containing a mixture of single-domain particles and wax was measured at 77°K and 300°K by means of a ballistic method. The ratio $(H_c)_{77}/(H_c)_{300}$ was found to be equal to $(M_s)_{77}/(M_s)_{300}$. This indicates that the coercive force of the particles is primarily due to shape anisotropy, an interesting (and somewhat surprising) fact in view of the conclusion (Sec. 3) that the microwave resonance frequency is determined by both crystalline and shape anisotropy in this case.

3. THEORETICAL INTERPRETATION

The resonances observed in the magnetic spectrum of the solid samples are undoubtedly composed of many individual resonances whose frequencies might be represented by some kind of distribution function. A calculation of this function appears to be prohibitively complicated and would require considerable information about the size, shape, and position of the crystallites and inclusions in the sample. Similar difficulties (treatment of magnetic interactions, geometry of the particles) would exist in the case of the single-domain particles. It is not yet possible, therefore, to predict the widths and shapes of the absorption "lines," or even the exact magnitude of the mean resonance frequencies, from a (supposed) knowledge of the saturation magnetization, crystalline anisotropy, damping constant, and other basic properties of the material.

As shown in Table II, however, there are two remarkably simple experimental facts concerning the temperature dependence of the mean resonance frequencies in this material. (a) In the range¹⁷ between 77°K and 300°K, the three frequencies ω_0'' , ω_0' , and Ω_0' depend on temperature in approximately the same way. (b) This common temperature dependence is qualitatively similar to, but quantitatively more pronounced than, that of the saturation magnetization. In the interpretation attempted below, we shall consider these two facts only.

We begin by discussing the nature of the internal fields. It is well known^{1,3} that

$$H_i = \omega_0' / \gamma, \tag{2}$$

where H_i is the internal field responsible for the rotational resonance. Since $\omega_0'/2\pi \sim 10^3$ Mc/sec, Eq. (2) yields $H_i \sim 300$ oersteds. To account for this field in terms of internal stresses, we must assume that they are of the order of $|M_{s}H_{i}/\lambda_{s}|$, where λ_{s} is the saturation magnetostriction. Using the value $\lambda_s \approx -2 \cdot 10^{-6}$ measured on ferramic A, we obtain about 10^{10} dynes/cm² $(\approx 10^2 \text{ kg/mm}^2)$ for the required stresses. In view of the brittleness of ferramic A, and the fact that the x-ray diffraction lines at room temperature are quite sharp,¹ it is very unlikely that stresses of this magnitude exist in our material at room temperature. While this argument does not apply to ω_0'' , and may not¹⁸ apply to Ω_0' , the temperature dependence of these frequencies was found to be approximately the same as that of ω_0' , so that the role of the stresses is probably unimportant in all three cases. It seems reasonable, therefore, to simplify the problem by completely neglecting the effect of internal stresses. The contributions to H_i of crystalline anisotropy and internal demagnetizing fields, on the other hand, must both be taken into account. This necessity was indicated in paper III in a qualitative way and will now be formulated quantitatively on the basis of room temperature data. If only anisotropy

 $^{^{16}}$ J. H. Griffiths and J. R. MacDonald, J. Sci. Instr. 28, 56 (1951).

¹⁷ We do not discuss the data taken at 411°K because at temperatures approaching the Curie point (≈ 550 °K in ferramic A) the distinction between domains and domain walls becomes meaningless.

¹⁸ The diffraction lines in the case of the single-domain particles are not very sharp (see reference 1), a fact which may be due to the small size of the particles or the presence of stresses.

TABLE II. Mean resonance frequencies and saturation magnetization for ferramic A.

Temper-	$\omega_0''/2\pi$		$\omega_0'/2\pi$		$\Omega_0'/2\pi$		M s	
°K	Mc/sec	%	Mc/sec	%	Mc/sec	%	cgsm	%
77	100	100	3900	100	5000	100	140	100
195	71	71	2700	69	3600	72	123	88
300	43	43	1400ª	36ª	2300	46	105	75

* This low value is probably due to the relatively pronounced overlap of the two resonances at this temperature; at 411°K the overlap is even more pronounced.

fields were important, we would expect $\Omega_0' = \omega_0'$, rather than the observed $\Omega_0' > \omega_0'$; and if only demagnetizing fields were important, we would expect $\Omega_0'/2\pi$ $\leq 2\pi M_s(\gamma/2\pi) \approx 1800$ Mc/sec, the limiting value appropriate for infinitely separated, long, thin, cylindrical particles of single-domain size, whereas experimentally $\Omega_0'/2\pi = 2300$ Mc/sec, as measured on particles that have been ground so that their shape should be roughly spherical rather than cylindrical. Finally, we allow for the possibility that torques due to exchange fields, which are known to be important inside domain walls, may affect the rotation of the spins within any domain that contains sufficiently small regions of inhomogeneous magnetization.

On the basis of the above statements we can describe rotations by writing Eq. (2) in the form

$$\omega_0' = \gamma f(K, M_s, E_J), \tag{3}$$

with a similar equation for Ω_0' . Here f is some unknown function whose temperature dependence is determined by K, M_s , and E_J (and whose form may be different if ω_0' is replaced by Ω_0'), K is the first-order anisotropy constant, and E_J is the exchange energy density. While M_s can be measured on polycrystalline samples (see Sec. 2C), an unambiguous and reasonably accurate determination of K requires a single crystal; owing to the effect of magnetic interactions, only the order of magnitude of K can be measured¹⁹ on a polycrystal. Since ferramic A is not available in the form of single crystals, K must be treated as an unknown parameter. The quantity E_J cannot be measured directly, and an indirect measurement is not now possible because of insufficiencies in the statistical aspects of the theory of ferromagnetism at all but very low temperatures. However, the molecular field approximation leads to the proportionality²⁰

$$E_J \propto M_s^2 \tag{4}$$

for the temperature dependence of E_J . This relation²¹

follows directly from the Néel²² theory of the ferrites because in magnesium ferrite (MgOFe₂O₃) all the magnetic ions are of the same kind (Fe^{+++}) ; the relatively small net magnetization is ascribed to the presence of an excess number of Fe+++ ions on the "octahedral" lattice sites of the incompletely inverted spinel structure. Using the relation (4) and noting that f must have the dimensions of a magnetic field, we can equate f to the product of M_s (or $K^{\frac{1}{2}}$, or K/M_s) and some function whose temperature dependence is determined by the dimensionless ratio K/M_s^2 . Without loss of generality, we may therefore replace Eq. (3) by

$$\omega_0' \propto g M_s F(K/M_s^2), \tag{5}$$

where F is some unknown function. The value of Fcan be derived in a few special cases. If, for example, magnetic interactions are entirely neglected, H_i is known to be given by $2K/M_s$ for a substance in which K is positive, so that in this case

$$F \propto K/M_s^2$$
, (6a)

a relation that applies equally well to the case of a uniformly magnetized, single-domain particle which is free of imperfections and spherical in shape. If, on the other hand, the effect of demagnetizing fields outweighs that of anisotropy, we expect $H_i \propto M_s$, so that in this case

$$F = \text{constant},$$
 (6b)

with respect to temperature. In the general case, however, we cannot progress beyond Eq. (5) unless we make some assumption about the form of F or the temperature dependence of K/M_s^2 .

To describe wall displacements,²³ we use the equation^{1,6}

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

where α is the stiffness constant²⁴ of a domain wall and m_w is its inertia (or apparent mass per unit wall area). In principle, α can be obtained from the static initial susceptibility due to wall displacements, χ_0'' , by using the formula¹

$$\alpha = M_s^2 / \chi_0'' d. \tag{8}$$

In practice, however, this determination is not possible because in a polycrystal there is no way of accurately separating χ_0'' from the measured $(\chi_0)_{total}$ and because the average domain size d is difficult to ascertain. It is therefore necessary to obtain α from theoretical considerations. Before doing this, we recall that the mechanism responsible for impeding wall motion in a given substance determines not only α but also the

¹⁹ G. T. Rado and A. Terris, Phys. Rev. 83, 177 (1951). ²⁰ The proportional sign (α) is used throughout the present paper to indicate proportionality of two quantities in the course

of temperature changes. ²¹ Several papers which do not explicitly consider temperature effects, e.g., the review of static domain theory by C. Kittel, Revs. Modern Phys. 21, 541 (1949), give the impression that E_J is independent of temperature. This may be a fair approximation near room temperature for materials having a high Curie point (e.g., silicon iron), but in most cases the relation (4) is probably a better approximation.

²² L. Néel, Ann. phys. 3, 137 (1948).

²³ It should be pointed out that the possibility of the existence of domain walls in ferrites, which are ferrimagnetic (see reference 22), was proved beyond doubt by the observations of J. K. Galt, Phys. Rev. 85, 664 (1952) on a single crystal of magnetite. As noted by Néel (reference 22), the theory of ferrimagnetic walls is

expected to be quite similar to that of ferromagnetic walls. ²⁴ See Eq. (11) of reference 1; the restoring force acting on a unit wall area during a small displacement, X, is given by αX .

coercive force. In ferramic A there are air-filled cavities, and probably other nonmagnetic inclusions, so that the coercive force (and thus α) may well be due to Néel's²⁵ disperse fields or to Kersten's surface tension effect,²⁶ depending on whether the size of the inclusions is comparable to, or much smaller than, the wall thickness. It is also possible that the size and irregularity of sufficiently small crystallites plays a role, as discussed qualitatively by Döring.²⁷ In view of the many detailed models which may have to be treated, the complications of the problem, and the unavailability of a precise description of the structure of our material, it is clearly necessary to keep the discussion as general as possible. Remembering that the effect of internal stresses is probably small, we therefore express each of the quantities α and g^2m_w by some function whose temperature dependence is determined by K, M_s , and E_J . On the basis of Eqs. (7) and (4) and a dimensional argument similar to that used in connection with Eq. (5), we obtain

$$\omega_0^{\prime\prime} \propto g M_s \Phi(K/M_s^2), \qquad (9)$$

where Φ is some unknown function. It is instructive to calculate Φ for a special case. If the size of the inclusions is very much smaller than the wall thickness, a straightforward expansion of the restoring force obtained from Néel's²⁸ model leads to $\alpha \propto K/\delta^2$, where δ is the wall thickness parameter. In deriving this expression we have neglected, for simplicity, the contribution of magnetic energy in comparison with that of exchange and anisotropy energy, a step justified by the supposed smallness of the inclusions. Using the same model, we find that m_w depends on the position of the wall. However, it can be shown by means of rough calculations and the properties of ferramic A that this effect may be neglected because it depends primarily on the small change of magnetic energy in a moving wall as it passes through an inclusion. Consequently we use the usual expression for m_w and write it in the form⁷ $m_w \propto 1/g^2 \delta$ for any given type of wall. On the basis of Eqs. (4) and (7) and the relation $\delta \propto (E_J/K)^{\frac{1}{2}}$, we therefore obtain in this case

$$\Phi \propto (K/M_s^2)^{\frac{3}{4}}.$$
 (10)

In the general case, however, we cannot progress beyond Eq. (9) unless we make additional assumptions, as stated in connection with Eq. (5).

It has been assumed implicitly that there is neither a change of phase (or atomic order) nor a change in the sign of K in the temperature range under consideration. We now assume, in addition, that K/M_s^2 is independent

of temperature between 77°K and 300°K. This assumption represents merely a rough first approximation, as discussed later, and its usefulness arises from the fact that no other reasonable assumption leads to a definite prediction on the basis of Eqs. (5) and (9). (In view of the difficulties outlined above, the situation would not be markedly improved if the temperature dependence of K/M_s^2 were known.) Our assumption leads to the simple result

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$$\omega_0'' \propto \omega_0' \propto \Omega_0' \propto gM_s, \tag{11}$$

which is seen to agree quantitatively with "fact (a)" mentioned near the beginning of this section. However, the agreement between the relation (11) and "fact (b)" is only qualitative because the results of Sec. 2B show that the temperature dependence of g, if it exists at all, is too small to account for the observation that the temperature dependence of the resonance frequencies is more pronounced than that of M_s .

The most probable explanation of the residual discrepancy is that K decreases with increasing temperature somewhat faster than M_s^2 . Such a behavior is to be expected if the known thermal characteristics of nickel ferrite¹² provide a guide to those of ferramic A. In an attempt to decide this question experimentally, we measured the static initial permeability of the single-domain particles and found it to decrease by at most one percent (which is about equal to the experimental error) between 77°K and 300°K. Since we are concerned here with rotations, one is tempted to conclude from this measurement that K/M_s^2 is independent of temperature in the range specified. However, the experimental data refer to an apparent permeability because of the (large) demagnetizing field of the particles, so that the observed temperature dependence of the permeability is necessarily less pronounced than the true dependence.²⁹ It is therefore possible that K/M_s^2 does, in fact, decrease somewhat as the temperature is increased. If this is the correct explanation of "fact (b)," however, then it is by no means clear why the functions F and Φ should have the same temperature dependence, a required behavior if we are to explain "fact (a)" without assuming that K/M_s^2 is constant. The best we can do at present is to surmise that F and Φ do have nearly the same temperature dependence in the limited range under discussion because in some special cases both of these functions vary less rapidly than K/M_s^2 ; this can be seen by comparing Eqs. (6a) and (6b), which represent the limiting cases for F, with Eq. (10), which represents one particular case for Φ . Finally, we summarize the discussion by noting that our interpretation, based on the relation (11) supplemented by the above remarks, appears to account semiquantitatively for the measured temperature dependence of the resonance frequencies.

We wish to thank M. Maloof for assistance in solving problems connected with the apparatus.

²⁹ This is similar to the air-gap effect in magnetic circuits.

²⁵ L. Néel, Annales Univ. Grenoble 22, 299 (1946); Physica 15,

 <sup>225 (1949).
&</sup>lt;sup>26</sup> For an improved calculation, see L. Néel, Cahiers phys. 25, 21 (1944). This paper also contains the prediction of "Néel spikes" for the case where the inclusions are much larger than

the wall thickness. ²⁷ W. Döring, Z. Naturforsch. 4a, 605 (1949). ²⁸ Reference 26, pp. 16–18.