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FERROMAGNETIC RESONANCE

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Sommaire. — Cette communication passe en revue les travaux expérimentaux et théoriques dans le domaine de l'absorption de résonance ferromagnétique. L'effet lui-même a été découvert expérimentalement par Griffiths en 1946. On montre qu'il est prévu par les équations de résonance classique, puis on en esquisse une interprétation par la Mécanique quantique. On discute les vérifications expérimentales des conditions de résonance pour des corps de différentes formes. On étudie la liaison entre les expériences de résonance en ondes très courtes et les mesures ordinaires du coefficient gyromagnétique. On établit l'équation donnant le point d'antirésonance. On discute diverses influences sur la largeur des raies. On résume les récentes études expérimentales de Blœmbergen, Bickford et Yager et Galt. Le mémoire se termine par une théorie de l'influence de la fréquence sur la perméabilité initiale des ferrites qui est en accord avec les résultats expérimentaux récents.

I. — Historical Review.

Ferromagnetic resonance absorption was discussed theoretically (Gans and Loyarte [1]; Dorfmann [2]) before its experimental discovery by Griffiths [3] in the Clarendon Laboratory in Oxford in 1946. In particular the theoretical paper by Landau and Lifshitz [4] written in Kharkov in 1935 provides a foundation from which, after certain essential modifications, many of the later results can be derived. After the first observation of ferromagnetic resonance the development of the subject was perhaps more strongly affected by contemporary work in the fields of nuclear and electronic paramagnetic resonance, than by the earlier thinking on the subject.

Resonance Equations. — The pioneer paper by Griffiths gives the results of resonance absorption experiments at several microwave frequencies on thin films of iron, cobalt and nickel. If his results are interpreted in terms of the Larmor equation connecting resonance frequency and applied field :

$$\omega_0 = g \, \frac{e}{2 \, mc} \, H, \tag{1}$$

we are led to values of the spectroscopic splitting factor g which are considerably too high, sometimes as high as 12, while the g-value for electron spin is 2.00.

The derivation of Equation (1) is based on the equation of motion

$$\frac{\mathrm{d}\mathbf{J}}{\mathrm{d}t} = \mathbf{M} \times \mathbf{H}, \qquad (2)$$

where \mathbf{J} is the angular momentum per unit volume and \mathbf{M} is the magnetization. We suppose that

$$\mathbf{M} = \mathbf{\mathbf{\gamma}} \mathbf{J},\tag{3}$$

where γ is the magnetomechanical ratio and is equal to $\frac{e}{mc}$ for electron spin. If we set **H** equal to the applied field H_z we have

$$j \omega M_x = \gamma M_y H_z, \qquad (4)$$

$$j\omega M_{\gamma} = -\gamma M_x H_z; \tag{5}$$

these equations may be solved for M_x , giving

$$(-\omega^2 + \gamma^2 H_z^2) M_x = 0, \tag{6}$$

so that the resonance frequency is given by

$$\omega_0 = \gamma H_z$$

This means that the energy absorption from an incident r - f field of frequency ω_0 will go through a maximum when the static field intensity is equal to $\frac{\omega_0}{\gamma}$. We have neglected demagnetizing effects in reaching the above result.

In the actual experimental arrangement employed by Griffiths and most subsequent workers there are appreciable demagnetizing effects which act to shift the resonance frequency. We consider a ferromagnetic sheet whose surface is the plane y = o, and which is subjected to a strong static field H_z . The essential point is that we must explicitly take into account the demagnetizing field $H_y = -4\pi M_y$ which results when in the course of the precessional motion of the spin system there occurs a magnetization component normal to the plane of sheet. The equations of motion become

$$j \omega M_{y} = \gamma (M_{y}H_{z} + 4\pi M_{y}M_{z}) = \gamma B_{z}M_{y}, \quad (7)$$

$$\omega M_{\gamma} = -\gamma M_x H_z, \qquad (8)$$

giving [5, 6]

j

$$\omega_0 = \gamma (B_z H_z)^{\frac{1}{2}} = g \frac{e}{2 mc} (B_z H_z)^{\frac{1}{2}}.$$
 (9)

When this equation is used to interpret the results

of Griffiths we find g-values often in the vicinity of 2.1 to 2.2, which is somewhat closer to our original expectation. The results are exhibited in figure 1. A typical resonance curve is shown in figure 2, from the work of Yager and Bozorth [7] on Supermalloy.



Fig. 1. — Summary of data of Griffiths on ferromagnetic resonance in plane specimens : comparison of effective g values as calculated using H and $(BH)^{\frac{1}{2}}$ in the resonance equation. Notice that $(BH)^{\frac{1}{2}}$ gives g values independent of frequency and close to the free electron value 2.

The resonance frequency depends in general on the form of the specimen, and we have a simple result only when the specimen is in the form of a general ellipsoid, and here only when the dimensions of the specimen are small enough in relation to the skin depth and to the wavelength in the material so that the r - f field penetrates uniformly and so that the phase of the r - f field is substantially the same throughout the specimen. In the case of a plane specimen these requirements may be relaxed, however, for geometrical reasons. The resonance frequency in an ellipsoid with demagnetizing factors N_x , N_y , N_z with the static magnetic field in the z-direction is given by

$$\omega_0 = \gamma \left\{ \left[H_z + (N_x - N_z) M_z \right] \left[H_z + (N_y - N_z) M_z \right] \right\}^{\frac{1}{2}}.$$
 (10)

This relation reduces in the case of a sphere to

$$\omega_0 = \gamma H_z, \tag{11}$$

which was first verified experimentally by Hewitt [8]. The predictions of Equation (10) regarding parallel and perpendicular orientations of a plane specimen were verified by Kittel, Yager and Merritt [9]. Other experiments with spherical specimens of ferrites have been performed by Beljers [20] and Yager, Galt, Merritt, Wood and Matthias [26].

The resonance frequency in a ferromagnetic single cristal will depend on the angle the magnetization makes with the crystal axes, as a result of the magnetocrystalline anisotropy energy. In the (001) plane of a cubic crystal we have

$$\omega_{\theta} = \gamma \left\{ \begin{bmatrix} H_{z} - (N_{x} - N_{z})M_{z} + \frac{2K_{1}}{M_{z}}\cos 4\theta \end{bmatrix} \times \left[H_{z} - (N_{y} - N_{z})M_{z} + \frac{i\Lambda_{1}}{M_{z}}\left(\frac{3}{2} + \frac{1}{2}\cos 4\theta\right) \right] \right\}^{\frac{1}{2}}, \quad (12)$$



Fig. 2. — Resonance curve for Supermalloy, according to Yager and Bozorth; the apparent permability is plotted on a logarithmic scale.

where θ is the angle between the z-axis and the [100] direction. In the (011) plane we have, according to Bickford [10]

$$\omega_{0} = \gamma \left\{ \begin{bmatrix} H_{z} + (N_{x} - N_{z})M_{z} \\ + \frac{K_{4}}{M_{z}}(2 - \sin^{2}\theta - 3\sin^{2}2\theta) \end{bmatrix} \\ \times \begin{bmatrix} H_{z} + (N_{y} - N_{z})M_{z} \\ + \frac{2K_{1}}{M_{z}}\left(1 - 2\sin^{2}\theta - \frac{3}{8}\sin^{2}2\theta\right) \end{bmatrix} \right\}^{\frac{1}{2}}$$
(13)

where θ is the angle between the z-axis and a [100] direction. It must be emphasized that the above equations are valid only when the static field is sufficiently large that the magnetization may be considered to be oriented approximately in the direction of the static field; that is, we must have $H_z \gg \frac{K}{M_z}$. The anisotropy effect in a single crystal was first verified experimentally by Kip and Arnold [11], who worked in the (001) plane of a Si — Fe crystal. They did experiments at two wavelengths, near 1 cm and 3 cm. At 1 cm $H \gg \frac{K}{M}$, and the predictions

of the theory were confirmed; but at 3 cm $H \approx \frac{K}{M}$ and the simple theory is no longer applicable. At 3 cm two resonance peaks were found for certain orientations, and this result was explained satisfactorily by considering the deviations of the magnetization from the direction of the static field.

Thus far our discussion has been based on classical theory. One expects classical theory to be applicable as the quantum numbers of the entire system are extremely large. It is, however, reassuring that quantum-mechanical calculations have been carried out which lead to the same resonance conditions. A most exhaustive investigation of the quantum-mechanical problem has recently been made by Van Vleck [12], who discusses the shape and anisotropy effects as well as considering line width and q-values. This treatment is of particular value in exhibiting the approximations which must be made in order that the quantum theory results be equal to the classical results. Other quantummechanical treatments have been given by Polder [13], Richardson [14], and Luttinger and Kittel [15].

The latter paper gives perhaps the simplest method by which one can see that the Zeeman eigenvalues are in fact displaced by demagnetizing effects in the manner predicted by classical theory, as in Equation (10). The classical macroscopic Hamiltonian of the system is

$$H = -M_z H_z V + \frac{1}{2} \left(N_x \ \mathcal{H}_x^2 + N_y M_y^2 + N_z M_z^2 \right) V, \quad (14)$$

where V is the volume of the specimen. This equation may be rewritten to give the quantum-mechanical Hamiltonian

$$H = aJ_z + bJ_x^2 + cJ_y^2, (13)$$

where the J's are angular momentum operators. We let H operate on the function

$$\psi = \sum_{m=-j}^{j} p_m u_m^j, \tag{16}$$

where the u_m^i are eigenfunctions of the operators J^2 , J_x . We are thus led to a difference equation on the p's; the difference equation is then approximated by a differential equation. The differential equation is essentially that for a harmonic oscillator, and gives equally spaced eigenvalues with the separation

$$\Delta E = g \,\mu_{\rm B} \left\{ - [H_z + (N_x - N_z)M_z] \\ \times [H_z + (N_y - N_z)M_z] \right\}^{\frac{1}{2}}, \qquad (17)$$

which is just the classical result.

Theory of g and g'. — It was a surprising and puzzling result of the ferromagnetic resonance

experiments that they led to g values appreciably higher than the values obtained from gyromagnetic experiments, such as the Barnett and Einstein-de Haas experiments. The situation now appears somewhat improved, as it has been shown that the resonance experiments do not in principle measure precisely the same quantity as the gyromagnetic experiments. The theoretical situation is not yet entirely satisfactory, and on the experimental side there is an unfortunate lack of agreement in the results of gyromagnetic experiments carried out by different observers. It would be of considerable importance to resolve the existing gyromagnetic discrepancies.

We now look more deeply into the philosophy underlying the two types of experiment. In a gyromagnetic experiment we determine the magnetomechanical factor g' by the equation

$$g' \frac{e}{2\,mc} = \frac{\Delta M}{\Delta J},\tag{18}$$

where M is the magnetization and J the angular momentum per unit volume. For convenience we shall imagine that we are always dealing with unit volume of material. Now the total ΔJ is zero by conservation of angular momentum, but we actually observe the angular momentum ΔJ_{lat} of the lattice, while

$$\Delta J_{\text{tot}} = \Delta (J_{\text{lat}} + J_{\text{spin}} + J_{\text{orb}}) = 0, \qquad (19)$$

the terms J_{spin} and J_{orb} being the angular momenta associated with the spin and orbital motion of the electrons. Now we may similarly decompose the magnetization :

$$\Delta M_{\rm tot} = \Delta (M_{\rm lat} + M_{\rm spin} + M_{\rm orb}), \qquad (20)$$

but M_{lat} may be neglected as the angular velocity of the lattice is always negligible in comparison with the electronic angular velocity. We have then the result that g' is determined by the equation

$$g' \frac{e}{2 mc} = \frac{\Delta (M_{\rm spin} + M_{\rm erb})}{\Delta (J_{\rm spin} + J_{\rm orb})}.$$
 (21)

Now we know that

$$\frac{d_{\rm spin}}{d_{\rm spin}} = \frac{e}{mc},\tag{22}$$

$$\frac{M_{\rm erb}}{L_{\rm orb}} = \frac{e}{2\,mc},\tag{23}$$

so that if we imagine

$$J_{\rm orb} = \varepsilon J_{\rm spin}, \qquad (24)$$

where $z \leq 1$, we have the result

$$g' \cong 2 - \mathfrak{l}. \tag{25}$$

In a resonance experiment we determine the spectroscopic splitting factor g by the equation

$$s \frac{e}{2mc} = \frac{\Delta M}{\Delta J}.$$
 (26)

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Now in a magnetic dipole transition

$$\Delta J_{\rm tot}^z = \hbar, \tag{27}$$

while for first-order approximation wave functions including spin-orbit interaction

$$\Delta J_{\rm spin}^z = h, \tag{28}$$

$$\Delta J_{\rm orb}^{z} = \varepsilon \hbar, \qquad (29)$$

so that we must have

$$\Delta J_{\rm Iat}^{z} = -\Delta J_{\rm orb}^{z} = -\varepsilon \hbar. \tag{30}$$

Therefore

$$s \frac{e}{2 m c} = \frac{\Delta U_{\rm tel}}{\Delta J_{\rm tel}} \cong \frac{\Delta (M_{\rm spin} + M_{\rm orb})}{\Delta J_{\rm spin}}$$
(31)

which should be compared with Equation (28) for q'. We have further

$$g \cong 2 + \hat{\epsilon} \tag{32}$$

so that g + g' = 4, or

$$g-2\cong 2-g',\tag{33}$$

as given independently by Van Vleck [12] in considerable generality and by the present author [16] for a particular model.

Equation (33) is approximately verified by experiment to the extent that g is usually greater than 2 while g' is usually less than 2, but there remains the unsolved problem that g - 2 is usually greater than 2 - g'. Numerical results are given in Table I.

TABLE 1.

Comparison of representative experimental values of spectroscopic splitting factor (g) and magnetomechanical ratio (g') for ferromagnetic substances (1).

	Microwave Resonance	Gyromagnetic Experiments
	g.	<i>g</i> '.
[ron	2.12-2.17	1.93
Cobalt	2.22	1.87
Nickel	2.2	1.92
Magnetite	2.20	1.93
Heusler alloy	2.01	2.00
Permalloy	2.07-2.14	1.91
Supermalloy	2.12-2.20	-

(1) The values of g' are taken from the summary by S. J. BARNETT, *Proc. Am. Acad.*, 1944, **75**, 109. The values of g are based largely on J. H. E. Griffiths, reference [3] and unpublished data kindly communicated by Dr. Griffiths; also on various papers and unpublished measurements by W. A. Yager and his collaborators, to which detailed reference is made in the text. The value of g for magnetite is from L. R. BICKFORD, *Phys. Rev.*, 1949, **76**, 137. For Permalloy, g is from Kip and Arnold, unpublished.

We have made above a statement about the properties of the first order wave functions for a state with no orbital degeneracy, but including spin-orbit coupling as a perturbation. For a proof of the statement the reader is referred to the papers just cited, and also to the paper by Gorter and Kahn [17]. The original suggestion that g might differ from g' in the ferromagnetic case was made by Polder [13] and Van Vleck [18].

The quantity ε which determines the amount of orbital momentum mixed in with the spin is given by an expression of the form

$$\varepsilon = -\frac{C\lambda}{\Delta},\tag{34}$$

where C is a constant of the order of unity, λ is the spin-orbit coupling constant, and Δ is an energy level separation of the order of the separation in the solid of energy levels which were coincident in the free ion. The coupling constant λ is positive for an electronic shell less than half full, and negative for a shell more than half full. We then expect ε to be positive, and g therefore > 2, for paramagnetic salts containing Fe++, Co++, Ni++, and Cu++ ions, and this is generally confirmed by experiment; ε is also positive for the ferromagnetic metals Fe, Co and Ni, and the alloys thereof which have been investigated. In Heusler alloy (Cu₂AlMn) $g \cong q' \cong 2$, and this is what would be expected on the naive picture that Mn++ ions are here the principal carriers of the magnetic moment, as Mn++ has a half-filled 3 d shell.

« Anti-resonance » point. — It was pointed out by Yager [23] that when the results of a resonance experiment are plotted as $\log \mu_{\rm R} vs H$ a welldefined minimum is frequently exhibited at low field strengths, as well as the usual maximum at higher fields. For a plane specimen with the static field parallel to the plane, Yager gives in the limit of small damping

$$\omega_{\min} = \gamma B_z, \tag{35}$$

which may be compared with the familiar expression for the point of maximum μ_{R} :

$$\omega_{\mathrm{max}} = \frac{\gamma}{2} \left(B_z H_z \right)^{\frac{1}{2}}.$$
 (36)

The point of minimum $\mu_{\rm R}$ corresponds physically fo $4 \pi M_x$ being equal to H_v , but 180° out of phase, so that the microwave induction $B_x \simeq 0$; in other words, we have a field but no flux. The expression tor the minimum is of considerable use in situations where it is not convenient to determine the saturation magnetization by ordinary methods; one may then solve Equations (35) and (36) for both M_v and γ , as was done for example by Yager and Merritt [19].

It is easy to derive the general condition for the minimum. The r - f susceptibility is, neglecting damping,

$$\chi_{v} = \frac{\chi_{0}}{1 - \left(\frac{\omega}{\omega_{0}}\right)^{2}},\tag{37}$$

where ω_0 is given by Equation (10) and

$$\chi_0 = \frac{M_z}{M_z + (\Lambda_w - \Lambda_z) M_z};$$

now μ_x will be zero when $\chi_x = -\frac{1}{4\pi}$, so that the condition for $\mu_x = 0$ becomes

$$\omega_{\min}^2 = (\mathbf{I} + 4\pi\chi_0)\omega_0^2, \qquad (38)$$

or

$$\omega_{\min} = \gamma \{ [H_z + 4\pi H_z + (N_x - N_z)M_z] \times [H_z + (N_y - N_z)M_z] \}^{\frac{1}{2}}.$$
(39)

Special cases follow :

Plane,
$$II$$
 parallel
plane : $\omega_{\min} = \gamma B_z$; (40)
Plane, II perpen-

dicular plane :
$$\omega_{\min} = \gamma \left[H_z (H_z - 4\pi M_z) \right]^{\frac{1}{2}};$$
 (41)

Sphere :
$$\omega_{\min} = \gamma [(H_z + 4\pi M_z)H_z]^2$$
. (42)

The minimum in the plane parallel case is illustrated by figure 3, which was obtained with Heusler alloy by Yager and Merritt [19].



Fig. 3. -- Ferromagnetic resonance in Heusler alloy, according to Yager and Merritt; note the minimum in the apparent permeability.

Line width. — The first task in the investigation of line width is to set up a suitable form of a relaxation expression to be included in the equations of motion, a form which will give an adequate representation of the experimental results. The first attempt was to write

$$\frac{\mathrm{d}\mathbf{M}}{\mathrm{d}t} = \gamma \,\mathbf{M} \times \mathbf{H} - \lambda' (\mathbf{M} - \chi_0 \,\mathbf{H}), \qquad (43)$$

but this was shown by Yager (1949) to fit the expe-

rimental results at one frequency as H is varied only if λ' is taken proportional to H. It is more satisfactory to write

$$\frac{\mathrm{d}\mathbf{M}}{\mathrm{d}t} = \gamma \,\mathbf{M} \times \mathbf{H} - \lambda \left[\frac{(\mathbf{H}\mathbf{M})\,\mathbf{M}}{M^2} - \mathbf{H}\right]; \qquad (44)$$

it is easily verified that the relaxation term is always perpendicular to M, so that the absolute magnitude of the total magnetization does not vary. The constant λ has the dimensions of a frequency and will be called the *relaxation frequency*. The relation between λ' and λ is readily seen to be given by

$$\lambda' M_z \cong \lambda H_z. \tag{45}$$



Fig. 3 a. — Line width in nickel and Supermalloy, as a function of temperature (Bloembergen).

In the interpretation of experimental results in ferromagnetic resonance one must always bear in mind that the actual resonance curves may be broadened not only by genuine relaxation effects, but also by crystalline anisotropy effects in polycrystalline specimens, and by local and surface strains and inhomogeneities in both polycrystalline and single crystal specimens; in alloys there may also be « unnatural » sources of broadening associated with variations in the order-disorder structure.

Experimental values of γ determined by Yager are given in Table II. Studies by Bloembergen will be referred to below.

The progress which has been made in the theoretical explanation of line width consists in showing that the several mechanisms for which calculations have been made fail entirely to account for the observed width. Akhieser [32] has calculated spinspin and spin-lattice relaxation effects on the basis of magnetic dipole interactions between electrons; this calculation leads to relaxation frequencies much lower than observed; Van Vleck [12] has confirmed this tendency with a calculation which bears rather more directly on the experimental situation than does Akhieser's calculation, which has been criticized by Polder [13].

TABLE II.

Experimental values of Relaxation Frequency λ , from Measurements by W. A. Yager.

Frequency			λ	
Material.	(Me).	M_s .	H_z (res).	~ (sec -1).
Supermalloy $\begin{cases} 2 \\ \end{array}$	ý 23932	630	4835	$1.13 imes 10^8$
	(9311	630	1043	1.97×10^{8}
Nickel	23906	490	5325	3.2×10^{8}
Heusler alloy	»388o	01č	584o	1.3 × 108

It is not surprising that magnetic dipole interactions do not account for the observed line-width, as it is well known that such interactions fail to account for the observed magnetostriction, which also results from interaction between magnetization direction and elastic strain.

Exchange interaction. — The effect of exchange forces on ferromagnetic resonance has recently been treated [24]. In a uniformly magnetized specimen exchange forces have no effect on the resonance frequency, as a consequence of the fact that the magnetization operator $\sum_i S^x_i$ commutes with the exchange operator $\sum_{i=i}^{j} \mathbf{S}_i \mathbf{S}_j$; in classical language, the Weiss molecular field $\mathbf{H}_{ov} = q\mathbf{M}$ is always parallel to the magnetization M, and hence the torque $\mathbf{M} \times q\mathbf{M}$ must vanish. But in metallic specimens the microwave field penetrates only to a depth of 10⁻⁵ to 10⁻⁴ cm, so that the r - fcomponent of the magnetization is non-uniform, and the exchange energy will play a role in determining the resonance frequency and line width. The effects will in general be of importance only in pure metals at low temperatures, where the skin depth is considerably smaller than at room temperatures.

We write the exchange energy density as [25]

$$f_{\rm ex} = \left(\frac{A}{M_s^2}\right) (\nabla \mathbf{M} \nabla \mathbf{M}), \qquad (46)$$

where $A = \frac{2JS^2}{a} = 2.0 \times 10^{-6}$ ergs : cm for iron. The shift for the plane parallel arrangement is found to be

$$H_z = \frac{2A}{\delta^2 M_s},\tag{47}$$

where ∂ is the classical depth for permeability $2\mu_2$; here $\mu = \mu_1 - j \mu_2$. There are contributions to the line width of the same order of magnitude as the field shift. At room temperature the effect in pure iron is only of the order of 10 cersteds at 2 400 mc: s, but may be of the order of 100 to 1000 cersteds at liquid hydrogen temperature.

II. — Recent Experimental Results.

Line width and g-values at elevated temperatures. — N. Bloembergen [22] has recently investigated the temperature dependance of the g-value and the line width in nickel and supermalloy from room temperature to above the Curie point. This



Fig. 4. — Resonance curves vs. temperature for Supermalloy, according to Bloembergen.

marks the first observation of resonance in the paramagnetic region above the Curie point of normal ferromagnetic materials. Bloembergen found that the g-value is independent of temperature within the limit of error (2 per 100), with g = 2.12 for supermalloy and 2.20 for nickel.

The results of the line width determinations at 24 400 mc : s are plotted in figure 3 a. The width $\Delta \omega$, which is equal to $\frac{I}{T_2}$ in Bloembergen's notation, is half the width in frequency between the points where the imaginary part of the susceptibility has dropped to half its maximum value. The absorption is found to be continuous across the Curie

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point in both materials. The Curie point is 358° C in nickel and about 400° C in supermalloy. The resonance curves in supermalloy at various temperatures are plotted in figure 4. The plane parallel arrangement was used.

Magnetite crystal at low temperatures. — L. R. Bickford [10] has studied ferromagnetic resonance in an artificial crystal of magnetite (Fe₃O₄) at low temperatures, and from microwave



Fig. 5. — Anisotropy constant K_1 of magnetite vs. temperature, from microwave measurements by Bickford.

measurements alone he has determined the crystalline anisotropy constant as a function of temperature; he has also contributed to theun ders-



Fig. 6. — Dependence of resonance field on crystal orientation, for magnetite at a fixed frequency (Bickford).

tanding of the transition which magnetite undergoes at -160° C (113° K), characterized by anomalous effects in specific heat, magnetization and electrical conductivity.

At room temperature, values of g = 2.12and $K_1 = -1.10 \times 10^5$ ergs; cc were obtained. It was found that below about $-_{90}$ ° C the absolute value of K_1 decreases with decreasing temperature, as shown in figure 5, reaching zero at about $-_{143}$ ° C. Between $-_{143}$ ° C and the transition point K_1 is positive and increases with decreasing temperature. The *g*-value was found to decrease gradually with temperature. A typical plot of the angular variation of the resonance field is given in figure 6.

The behavior of magnetite in the resonance experiments below the transition indicates that the magnetic symmetry is uniaxial in this region. Below the transition the magnetic axis is the [100] direction most nearly parallel to a strong magnetic field applied to the crystal as it is cooled through the transition. At temperatures not far below the transition it was found to be possible to change the magnetic axis from one [100] direction to another by means of a strong magnetic field.

Nickel ferrite crystals. — Yager, Galt and their collaborators [26] have recently carried out resonance experiments on small spheres of nickel ferrite NiO-Fe₂O₃ cut out from single crystals. The



Fig. 7. — Resonance curve for nickel ferrite single crystal in form of sphere 0.10 cm diameter (Yager, Galt, Merritt, Wood and Matthias).

frequency was about 24 000 mc : s, and the observations were made at room temperature. The anisotropy constant was determined from the variation of resonance field as a function of crystallographic orientation. The easy direction was found to be [111], as in magnetite, and $K_1 = -6.0 \times 10^4 \text{ ergs} : \text{cc.}$

The first sphere measured was 0.10 cm in diameter, and it was found here that for every orientation of the crystal the resonance consisted of a main line and one satellite on either side, figure 7. The satellites are explained as caused by cavity-type electromagnetic resonances in the sphere. The idea is that as we go up the permeability resonance curve we pass through values of $(\varepsilon \mu)^{\frac{1}{2}}$ sufficiently

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large so that the half-wavelength of the radiation in the material is of the order of the diameter of the specimen, and near this point we expect a cavity type resonance. It is not possible to verify this explanation directly, as the dielectric constant ε is not known for nickel ferrite, but the order of



Fig. 8. — Same as figure 7, except that sphere is 0.038 cm in diameter. Difference between curves is ascribed to electromagnetic cavity-type resonance effects in the larger sphere.

magnitude of the required wavelength in the material appears to be reasonable. The explanation can be tested, however, by measurements on a much smaller sphere, as for a sufficiently small sphere at the same frequency one would not expect cavity resonances to be possible. Measurements on a sphere about 0.038 cm in diameter showed in fact only a single resonance peak, figure 8.

III. — Frequency Dependence of Initial Permeability.

Our recent understanding of the ferromagnetic resonance effect enables us to explain the principal aspects of the frequency dependence of the permeabiliy of ferromagnetic materials under conditions of normal usage, in which there is no static or biasing field present. The discussion is particularly simple in the case of high-resistivity ferromagnetic substances, such as the ferrites, as here we may neglect entirely eddy current effects, even on a microscopic scale. We first discuss frequency dependence in such substances; it is also assumed that the measurements are interpreted in such a way that the true magnetic permeability is obtained, quite separate from dielectric effects, especially geometrical resonance effects.

The average permeability of a polycristalline material may be written as

$$\mathbf{1} - \mathbf{I} = 4 \pi (\chi_{\text{rot}} + \chi_{\text{disp}}), \tag{48}$$

where $\chi_{\rm rot}$ is the average susceptibility associated with rotation of magnetization within the domains, and $\chi_{\rm disp}$ is the average susceptibility associated with displacement of domain boundaries. At very low frequencies both $\chi_{\rm rot}$ and $\chi_{\rm disp}$ will contribute to the permeability; the two terms will, however, depend on frequency in different ways. In some cases it is possible experimentally to separate the two types of dispersion.

The rotational susceptibility is determined largely by the crystalline anisotropy energy, or in some cases by strain energy and the magnetostrictive constants. In either situation there will be an effective anisotropy energy density K which determines the magnitude of the rotational susceptibility, according to the classical relation

$$\chi_{\rm r,t} \approx \frac{M_{\rm s}^2}{K},\tag{49}$$

where M_s is the saturation magnetization. There may be in special cases demagnetizing effects which will change the susceptibility appreciably, but we shall suppose that the average rotational susceptibility at zero frequency in a randomly-oriented polycrystalline specimen is given by

$$\chi_{\rm rot}^0 = \frac{M_s^2}{3\,K}.\tag{30}$$

This definition corresponds to the use of $\frac{2K}{M_s}$ as the effective anisotropy field.

The frequency variation is given, using Equations (44) and neglecting all demagnetizing effects, by

$$\frac{\chi_{\rm ret}(\omega)}{\chi_{\rm rot}^{0}} = \frac{\omega_{0}^{2} + \left(\frac{\lambda}{\chi'}\right) \left(j\omega + \frac{\lambda}{\chi'}\right)}{\omega_{0}^{2} + \left(j\omega + \frac{\lambda}{\chi'}\right)^{2}},\tag{31}$$

where χ' is an average rotational susceptibility approximately equal to χ^{0}_{rot} ; and the resonance frequency in the anisotropy field is

$$\omega_0 = \gamma \left(\frac{2K}{M_s}\right) = \frac{2\gamma M_s}{3\chi_{r-1}^0} \,. \tag{52}$$

The sharpness of the resonance depends on the value of the ratio

$$\frac{\omega_0}{\frac{\lambda}{\lambda_{\rm rot}^0}} = \frac{2\gamma M}{3\lambda}; \tag{33}$$

that is, if the resonance frequency in the field M_s , is much greater than the relaxation frequency λ the resonance will be well defined, and vice versa. In actual materials intermediate cases occur most commonly, but sometimes the rotational dispersion resembles a resonance process. In the two limiting further cases the Equation (51) assumes the form : $\lambda \ll \gamma M_s$; resonance

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$$\frac{\chi_{\rm r-1}(\omega)}{\chi_{\rm rot}^0} \simeq \frac{1}{1 - \left(\frac{\omega}{\omega_0}\right)^2},\tag{54}$$

 $\lambda \gg \gamma M_{s}$; relaxation

$$\frac{\chi_{r+1}(\omega)}{\chi_{rot}^0} \cong \frac{1}{1+j\frac{\omega\chi_{rot}^0}{2}}.$$
(53)

The displacement susceptibility at low frequencies is determined by the restoring force tending to return the boundary wall to its equilibrium position, and by the number of walls intercepting a line of unit length. We suppose that the restoring force per unit area of wall is $-q\Delta x$; the magnetizing pressure is ${}_{2}M_{s}(\Delta H)$, so that

$$\Delta x = \frac{2M_s(\Delta H)}{q} \cdot \tag{56}$$

The magnetization change is

$$\Delta M = 2 M_s \eta \,\Delta x,\tag{57}$$

where r_i is the average number of boundaries per unit length. Thus

$$\chi^{0}_{\rm disp} = \frac{4M_s^2 \gamma}{q} \cdot \tag{58}$$

It was first shown by Landau and Lifshitz [4] that there is a frictional term in the equation of motion of a boundary, of such a nature that in the absence of a restoring force the wall velocity is given by

$$\frac{v}{H} = (\gamma^2 M_s^2 + \lambda^2) \frac{\left(\frac{A}{K}\right)^{\frac{1}{2}}}{\lambda M_s}, \qquad (59)$$

where $\left(\frac{A}{K}\right)^{\frac{1}{2}}$ is the usual wall-thickness parameter; it is near the order of magnitude of the wall thickness, but somewhat smaller [25, 30]. A simple derivation of Equation (59) has been given Kittel [38].

The complete equation of boundary motion is

$$\Delta H = \left(\frac{q}{2}\frac{\Delta x}{M_s}\right) + \frac{j\omega\Delta x}{G},\tag{60}$$

where in Equation (59)

$$G = \frac{v}{H} = \left(\frac{\gamma^2}{4}M_{\lambda}^2 + \lambda^2\right) \frac{\left(\frac{A}{K}\right)^2}{\lambda M_s}.$$
 (61)

We have then

$$\Delta x = \frac{\Delta H}{\frac{q}{2M_{\star}} + \frac{j\omega}{G}}, \qquad (62)$$

or

$$\chi_{\rm disp} = \frac{2M, \gamma}{\frac{q}{2M} + \frac{j\omega}{G}}; \tag{63}$$

$$\frac{\chi_{\rm disp}}{\chi_{\rm disp}^{0}} = \frac{1}{1 + j \circ \frac{\chi_{\rm disp}^{0}}{2M_{\rm e}G_{i}}} \cdot$$
(64)

This result [31] shows that the displacement susceptibility has a frequency dependence of the relaxation type, with relaxation frequency

$$\Lambda = \frac{2 M_s \tau_1 G}{\chi_{\rm disp}^0} \,. \tag{65}$$

We estimate the order of magnitude of \ by taking $M_{
m s} \sim 10^3$, $\tau_{\rm i} \sim 10^3$, $\chi^0_{
m disp} \sim 10^2$; and a rough estimate gives $G \sim 10^3$ cm : s : cersted, so that $\Lambda \sim 10^7$ cycles : s, although in high permeability materials Λ may be very much less than one Mc : s.



Fig. 9. - Frequency dependence of permeability in magnesium ferrite, according to Welch, Nicks, Fairweather and Roberts.

The most complete measurements available to the author on frequency dependence in ferrites are these of Welch, Nicks, Fairweather and Roberts [29] on magnesium ferrite. Their results are shown in figure 9. One may also refer to work by Birks [21].

Relationship between frequency and permea**bility.** — We have from Equation (52)

$$\omega_0 \chi_{\rm rot}^0 = \frac{2 \,\gamma \, M_{\star}}{3} = \left(\frac{g \, e}{3 \, mc}\right) M, \tag{66}$$

where ω_0 is the rotational resonance frequency and γ is the magnetomechanical ratio. This relation was first stated by Snoek [28], although in incomplete form, as he does not point out that it is not the total susceptibility, but only the rotational susceptibility, with which the equation is concerned. Now at high frequencies we are often concerned only with the rotational part of the susceptibility, as the displacement contribution often drops out first. Equation [66] therefore

conveys the interesting and technically important result that the product of the « maximum usable frequency » by the « effective high-frequency permeability » of a material is essentially a constant, dependent for practical purposes only on the saturation magnetization. We have approximately

$$(\mu_{rf} - \mathbf{I})f_0 \approx 2B_s \,\mathrm{Mc.} \tag{67}$$

This relation is plotted in figure 10, and there are included several experimental data, which are seen to be in fair agreement with the theory.



Fig. 10. — Plot for several materials of maximum usable frequency vs. effective high frequency permeability.

We pause to note that for $\gamma M_s \gg \lambda$,

$$G \cong \gamma^2 M_s rac{\left(rac{A}{K}
ight)^{rac{1}{2}}}{\lambda},$$

and using Equation (65),

$$\frac{\Lambda}{\lambda} \simeq \left(\frac{\gamma^2 M_{\lambda}^2}{\lambda^2}\right) \left[\frac{2 \eta \left(\frac{A}{K}\right)^{\frac{1}{2}}}{\chi_{\rm disp}^0}\right]; \tag{68}$$

if we take as a numerical example $\frac{\gamma M_s}{\lambda} = 3$, $\eta = 10^3$.

$$\left(\frac{A}{K}\right)^{\frac{1}{2}} = 3 \times 10^{-6}$$
, and $\chi^{0}_{dips} = 10^{2}$, we have
 $\frac{\Lambda}{\lambda} \sim 6 \times 10^{-4}$ (69)

which suggests that A may often be less than λ ; further, for the same numerical values,

$$\frac{\Lambda}{\omega_0} \sim 2 \times 10^{-3}, \tag{70}$$

so that the displacement susceptibility may drop out with increasing frequency before the rotational susceptibility. We note further that Equation (65) gives a relation involving $\Lambda \chi^{\rm disp}_{\rm usp}$ which is parallel in function to the relation (66) involving $\omega_0 \chi^{\rm p}_{\rm rel}$, but appreciably more complicated and thus less useful.

Rado, Wright and Emerson [37] have suggested independently in connection with their measurements on a magnesium ferrite "Ferramic A" that the permeability may be broken down into rotational and displacement contributions. In their material the wall damping is unusually low, and they accordingly find definite effects of the wall inertia term calculated by Döring [27]. This marks the first time that the inertial effects of wall motion have been observed. The condition for the resolution of the inertial term is given as, approximately,

$$\left(\frac{\lambda}{\gamma M_s}\right)^2 < \frac{\delta \eta}{2 \pi \chi_{\rm disp}^0} \cdot$$
(71)

Dispersion in conducting materials. — In conducting materials the wall movements are damped out for the most part at lower frequencies [33] than in the ferrites. This is the result of eddy currents in the material. At high frequencies the magnetization is expected to be attribuable principally to domain rotation. One may cite the measurements of Johnson, Rado and Maloof [34], for example, who find that in iron 200 mc : s magnetization by rotation is the dominant effect. Another confirmation is offered by the reasonably good agreement of Equation [67] with the data on conducting materials.

It seems probable then that the higher frequency relaxation effects are associated with rotational relaxation, rather than with eddy current effects previously suggested by several workers [35], including the present author [36]. It is also unlikely that effects of the inertia of domain boundaries [27] are important here, as the frictional terms in the equation of motion of a Bloch wall are dominant in conducting materials and serve to eliminate wall motion at relatively low frequencies.

Acknowledgments. – I wish to express my indebtedness to my colleague W. A. Yager for his kindness in making available various unpublished results, and thanks are also due to Professor J. H. Van Vleck and Dr N. Bloembergen for friendly discussions of the ferromagnetic resonance problem. The writing of this paper has been facilitated by the constant assistance of J. G. Walker and Miss Phyllis Brown. The drawings were kindly produced by the Research Drafting Department under E. H. Hasbrouck.

Remarque de M. Bates. — I only wish to say that Dr Standley at Nottingham has found a preliminary value of g for MnAs by the method of ferromagnetic resonance of about 2.6; it is certainly greater than 2.5.

Remarque de M. Casimir. — Je propose d'adopter les notations de M. Kittel en désignant par g la valeur trouvée dans les expériences de résonance gyromagnétique et par g' les valeurs données par les expériences Einstein de-Haas ou Barnett.

Remarque de M. Roberts. — I would like to make three remarks on Dr Kittel's contribution : 1° Regarding g values. I have obtained in some magnesium and other ferrites, in powder form, with samples of a few mm³ dispersed across the wave guide cross-section, two resonance peaks as the transverse static field is increased. These two peaks are usually unequal in height and very broad, and suggest a splitting of energy levels of the magnetic ions in these ferrites. The direct interpretation of the results of high frequency measurements in terms of the g-factor alone can therefore lead to erroneous values. 2° Regarding the experimental line widths obtained in nickel and supermalloy, I wonder whether this could be accounted for in terms of a magnetic zero point energy such as Prof. Stoner has found, in effect, to be operative in preventing the complete parallel alignement of all spins at 0° K. 3° I should like to add new results to those shown in Dr Kittel's figure 9.



These are shown in my figure A and refer to a range of Mg ferrites in which Fe is gradually replaced by Al. It is found in all cases that these curves can be matched by a relaxation term plus two damped resonance terms like Frenkel's damped resonance, these two terms being nearly equal and opposite in low frequency magnitude and differing slightly in resonance and damping frequencies. To complete the picture, a third Frenkel term should be added, to correspond with those spins which give the resultant low frequency magnetisation. The other two Frenkel terms then correspond with the two groups of Fe⁺⁺⁺ spins which mutually cancel at low frequencies.

Remarque de M. Stoner. — I should like to report very briefly on some work at Leeds by Millership, Webster and others on the measurement of initial permeability of metallic ferromagnetics in the form of wires over a frequency range down to a wave length of 3 cm. It is essential to measure both the " resistive " and " inductive " permeabilities from which the " real " and " imaginary " components of the complex permeability may be determined. For iron the curves fit roughly, but not well, on the theoretical curves obtained by Becker. The fit is no better when a further theoretical term corresponding to resonance is introduced. For nickel, there are clear indications of resonance effects, no doubt because the anisotropy field frequency is in the range examined, while for iron it is too high. The work on some five ferromagnetic metals and alloys is nearing completion.

Remarque de M. Casimir. — There have been made some measurements at Delft by Miss van Leeuwen and pupils in cooperation with Kronig. Preliminary results have been published in "Physica" and one may look for further results.

Remarque de M. Went. — I should like to say a few words on the role of the rotational processes

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and wall displacements in the magnetization process if sintered ferrites. It is well known that in metallic ferromagnetics the major contribution to the permeability at low amplitudes is due to reversible wall displacements. We have found (Went and Wijn, to be published) that in sintered ferrites the rotational processes are mainly active in the low amplitude region. The observations are the following. The initial permeability is virtually constant up to the frequency where precessional resonance sets in. While at larger amplitudes and low frequencies the permeability is usually considerably higher as a result of (irreversible) wall displacements, we find that, in many materials, at large amplitudes and frequencies above roughly 100 Kc : s the permeability decreases to a value exactly equal to the initial permeability. We have found that the gradual disappareance of the contribution of the wall displacements as a function of frequency can be described by a single relaxation time. Furthermore we have found that the occurence of the damping of the wall dispacements is closely connected with the existence of volume magnetostriction.

Remarque de M. Casimir. -1° I should like to add a few comments to Went's remarks. He has particularly stressed that he has only done measurements on sintered specimens and it seems that even there the wall's contribution of initial susceptibility is negligible only for certain ferrites and it seems that those ferrites have always considerable volume magnetostriction. This suggest a mechanism of freezing in connected with volume magnetostriction. 2º In connection with Bozorth's remark that the size of a specimen in a resonant cavity has to be small compared to the wavelenght I should like to point out that it has to be small compared to the wavelenght inside the specimen; this wavelength may be considerably shorter than that in vacuum. The problem of calculating the corrections when this is not the case is rather difficult to solve when the susceptibility is anisotropic, as in the case for gyromagnetic resonance; so far we have not been able to find a solution and not even the value of the first coefficient in a series development in powers of $\frac{R}{5}$.

Remarque de M. Shockley. — Dr Roberts has remarked that values of q as high 2.6 to 3.0 have been obtained in Mg ferrite powder. It may be possible that these large values are due in part to magnetostatic effects which tend to act as an increased restoring force.

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