RESONANCE OF DOMAIN WALLS IN COBALT FERRITE

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Submitted to JETP editor August 8, 1960

J. Exptl. Theoret. Phys. (U.S.S.R.) **40**, 441-447 (February, 1961)

A resonance dispersion of magnetic permeability due to domain-wall displacement processes is obtained in a monocrystal of cobalt ferrite. Resonance is observed at frequency 360 Mc/sec. A comparison is made with the theoretically calculated resonance frequency.

INTRODUCTION

THE dependence of the magnetic permeability of ferrites on the frequency at radio and microwave frequencies has two characteristic ranges, one due to motion of domain walls, and the range of natural ferromagnetic resonance. The nature of the dependence in the first of these ranges has not actually been established experimentally; however, a number of experiments[1,2] indicate that resonance dispersion occurs here.

It is known that in weak magnetic fields the magnetization is determined by ferromagnetic-domain wall-displacement processes. Döring[3] showed theoretically that a moving wall possesses an effective mass. The wall is bound to an equilibrium position by a quasi-elastic force. From the presence of a mass and an elastic bond it follows that the wall has a characteristic frequency of oscillation. Consequently, there should be observed a resonance dispersion of magnetic permeability, caused by wall-displacement processes.

In the presence of inertia $m\ddot{X}$, elastic restoring force $-\alpha X$, and frictional force $-\beta\dot{X}$, the equation of motion of a 180-degree wall under the influence of a pressure $2HI_s$ exerted by a magnetic field H, parallel to the wall, takes the form

$$m\ddot{X} + \beta\dot{X} + \alpha X = 2HI_s. \tag{1}$$

On solving this equation for the case of an alternating field $H = H_0e^{i\omega t}$, we get for the magnetic permeability the well known resonance formula

$$\chi(\omega) = \chi_0 \frac{1}{1 - \omega^2/\omega_0^2 + i\omega/\omega_1}, \tag{2}$$

where $\omega_0 = \sqrt{\alpha/m}$, $\omega_1 = \alpha/\beta$, and χ_0 is the initial magnetic susceptibility:

$$\chi_0 = 4I_s^2/\alpha l \tag{3}$$

(since $\chi = 2I_sX/Hl$, where l is the mean width of a domain).

In the present work, the resonance character of the dispersion associated with wall motion is established for cobalt ferrite.

METHOD AND EXPERIMENTAL RESULTS

The magnetic dispersion associated with wall-displacement processes was observed by Rado, Wright, and Emerson[1] in polycrystalline iron-magnesium ferrite (Ferramic A). They attributed the increase of the real part of the magnetic permeability, μ', of the specimen with increase of the frequency of the alternating magnetic field to the resonance character of this dispersion. However, they pointed out that μ' does not become less than unity, as should be the case with resonance, perhaps because of the superposition of a rotational resonance at a near frequency. Miles, Westphal, and von Hippel[2] observed the same sort of dispersion in polycrystalline nickel ferrite and nickel-zinc ferrite and in a monocrystal of nickel ferrite. They likewise obtained no values of μ' less than unity.

We constructed a coaxial line of square section. Such a line possesses all the advantages of the usual coaxial line: absence of dispersion, presence of a single TEM mode. It is important that the form of the magnetic lines of force is approximately square. Therefore if one cuts from a monocrystal a specimen in the form of a square frame, with sides along axes of easiest magnetization, then the alternating magnetic field will be parallel to the directions of the magnetization within the domains. By the same token, superposition of a natural ferromagnetic resonance is excluded, however near in frequency it may be to the resonance being observed. It is to be expected that for cobalt ferrite, because of its high magnetic anisotropy, the resonance associated with rotation of the magnetization vector in the magnetic-anisotropy field will lie in the millimeter-wave range and therefore will be in general unobservable in our experiment.

In the construction of the measuring line, we started from the specimen dimensions and as large as possible a frequency range. The outer dimensions of the coaxial were 10×10 mm, the inner 6×6 mm. The distance the probe could be moved along the line was 450 mm. The detector head was made in the form of a half coaxial resonator. Preparation of an assortment of attachments to the line, of various lengths (waveguide sections, in which the specimen is placed), made possible the measurement of the input resistances in the frequency range 200 to 300 Mc/sec. In the calculation of the input resistance, the specimen may be considered isotropic, since the magnetic lines of force of the high-frequency field are parallel to a single crystallographic direction.

To test the accuracy of performance of the apparatus, the magnetic permeability of a frame of Plexiglas was measured at several frequencies; as was to be expected, it was found equal to unity.

Monocrystals of cobalt ferrite were obtained by the method of Verneuil, in an oxyhydrogen flame. The composition of the ferrite, according to chemical analysis, was $Co_{0.94}Fe^{++}{}_{0.12}Fe^{+++}{}_{1.96}O_4$. Because of the presence of divalent iron, the crystals have a quite high electrical conductivity, which leads to difficulties in the observation of their magnetic susceptibility at high frequencies. We did not succeed in growing, by this method, crystals free from divalent iron.

To decrease the conductivity, the monocrystals were annealed in oxygen at 900° C. Divalent iron oxidized to trivalent, and the electrical conductivity decreased. By such an anneal, sufficiently prolonged, we succeeded in raising the specific resistance to 10^4 ohm cm.

Four square frames, of identical dimensions, were made. The outside dimensions of the frames were 10×10 mm, the inside 6×6 mm. (The inner opening was made by means of ultrasound.) The plane of the frames coincided to within tenths of a degree with the crystallographic plane (100), and the sides of the frames to within 2° with a direction of the type [100], i.e., with axes of easiest magnetization. The crystallographic directions were determined by x-ray diffraction [“epigrams” — Laue patterns by reflection — were taken]. The thickness of the frames was 3 mm.

Two frames made from the same crystal were annealed for six hours, two others for seven days. The resistivity of the first two frames rose to the value 10^3 ohm-cm (the chemical composition changed to $Co_{0.94}Fe^{++}{}_{0.06}Fe^{+++}{}_{2.00}O_4$), of the other two to the maximal value 10^4 ohm-cm.

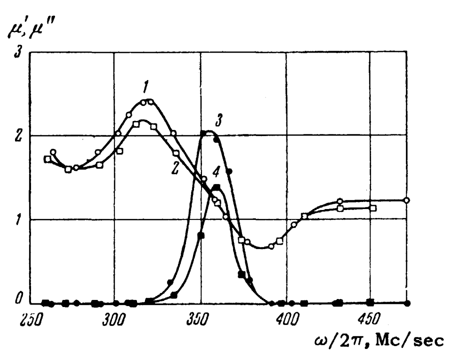
For determination of the direction of magnetization and of the domain width, powder patterns on the frames were observed. For this purpose the surface of the frames was polished (mechanical polishing), and a drop of a ferromagnetic suspension was placed upon it. The powder patterns were observed under the microscope. The domain picture was photographed, and from the photographs the domain width was determined.

A photograph of the powder patterns for a frame annealed for six hours shows parallelism of the domains to the sides of the frame. The domain width (distance between walls) is from 3.7×10^{-3} to 6.4×10^{-3} cm. The domain width in frames annealed for seven days is an order of magnitude smaller, i.e., 3 to 4×10^{-4} cm. Thus in all the frames, parallelism of the alternating magnetic field to the direction of the domain magnetization was attained (with the indicated precision). An exception is the corners of the frames, where both the field and the domain configuration are more complicated. However, these corner regions occupy a small part of the whole body of the specimen, since the sides of the frames are narrow (2 mm). For this reason all the domains in the frames may be considered 180° ones, and the alternating magnetic field may be considered parallel to the domain walls.

The measurement was carried out over a range of frequencies that includes the whole region of noticeable absorption. The first two frames (with domain dimension 3.7 to 6.4×10^{-3} cm) showed a maximum of the magnetic losses at about 360 Mc/sec. Measurements on these were carried out over the range 250 to 450 Mc/sec. In the whole range of frequencies accessible to us (200 to 3000 Mc/sec), no absorption was observed for the other two frames, with the small domain dimension (2 to 3×10^{-4} cm).

The figure shows the behavior of the real (μ') and imaginary (μ'') parts of the magnetic permeability for the first two frames. The curves have a clear resonance character: μ' goes through a maximum and a minimum, in which the value of μ' is less than $\mu'(\infty)$; μ'' goes through a maximum.

We shall compare the curves obtained and formula (2). We first determine ω_0 and ω_1. This can be done by various methods: from the position of the maximum of μ'', from the intersection of



Dependence of the complex magnetic permeability $\mu = \mu' - i\mu''$ of a monocrystal of cobalt ferrite on the frequency of the alternating magnetic field: 1, μ' for frame No. 1; 2, μ' for frame No. 2; 3, μ'' for frame No. 1; 4, μ'' for frame No. 2.

μ' with the straight line $\mu'(\infty)$, from the position of the maximum and the minimum of μ'. The various methods of determination from the two curves give for the two frames, with deviation not exceeding 1%, the following values: $\omega_0 = 2\pi \times 360$ Mc/sec, $\omega_1 = 2\pi \times 2150$ Mc/sec.

On using these values and plotting the theoretical resonance curve according to formula (2), we discover that the experimental curve is narrower than the theoretical. This fact remains obscure. On the contrary, we should expect a broader experimental resonance because of different values of the parameter α for different walls, in consequence of the structure-sensitivity of this parameter.

As additional verification of the fact that the resonance obtained was not "instrumental," the measurements were repeated on the same specimens in the presence of a constant external magnetic field of intensity 10 000 oe. The magnitude of the field was sufficient to make the specimen a single domain. In this case no resonance was observed. Coaxial attachments of different length were also used. Identical results were obtained.

CALCULATION OF THE EFFECTIVE MASS OF A WALL

In order to compare the experimental values of ω_0 and ω_1 with the theoretical, it would be necessary to calculate α, β, and m. Inasmuch as we do not have sufficient quantitative information about inclusions and strains in our specimens, a calculation of the structure-sensitive parameter α according to a theoretical formula is not possible. However, it is possible by use of formula (3) to determine α from the value of the initial magnetic permeability, which we evaluated independently on one of the two frames ($\mu_0 = 2$). As regards the damping parameter β, which is a measure of the energy losses associated with wall motion, its physical nature is not yet clear, since the mechanism of losses in ferrites is not clear. Therefore we will not try to calculate ω_1 but will calculate ω_0. For this purpose we will calculate the effective mass m of the wall.

Döring[3] considered a domain wall in uniform motion in a cubic crystal and expressed its energy W in the form of a power series in the velocity v:

$$W = W_0 + mv^2/2 + \ldots. \tag{4}$$

The first term of the series is the energy of the wall at rest. The term linear in v drops out, since the energy is independent of the sign of the velocity. The quadratic term, if the remaining terms of the series are negligible, represents the kinetic energy of the moving wall. It is natural to regard the coefficient of $v^2/2$ as the effective mass of the wall.

The expansion (4) is carried out, essentially, in terms of a quantity λw^2, where $\lambda = K/2\pi I_s^2$ (K is the magnetic anisotropy constant), and w is a dimensionless velocity, $w = v\hbar n/2K\delta$ (n = number of spins in unit volume; $\delta = (Aa^2 n/4K)^{1/2}$, a parameter describing the wall thickness; a = constant of the cubic lattice; A = mean value of the exchange integral). Thus for validity of the expansion (4) two conditions are necessary: $\lambda \ll 1$, which is satisfied for a whole series of materials, and $w \le 1$. Döring assumed that the velocity was limited by damping processes.

Rado[4] extended Döring's derivation to the case of a field alternating in time, i.e., to the case of a wall in oscillatory motion. He obtained the same expression for the mass that Döring had obtained (under the condition $\lambda \ll 1$). However, the series expansion is now carried out in terms of the two dimensionless parameters

$$p = \gamma H_0/\omega \ll 1, \qquad q = \omega/\gamma(2K/I_s) \ll 1,$$

where γ is the magnetomechanical ratio and H_0 is the amplitude of the external field $H = H_0 e^{i\omega t}$.

Smallness of the parameter p means that the frequency of oscillation of the external field is much larger than the frequency of the Larmor precession in the external field. Smallness of the parameter q means that the frequency of oscillation of the external field is much smaller than the Larmor resonance frequency for rotation in the internal anisotropy field. Both these conditions insure smallness of the velocity of the wall motion without allowance for damping processes, and consequently the legitimacy of neglecting terms above the quadratic in the expansion of the energy

W as a power series in the velocity v (equivalently, the mass is independent of the velocity). Rado remarked that the condition $\lambda \ll 1$ is not essential (it merely simplifies the calculation, making it possible to obtain an analytic expression for the mass) and can be dropped.

Since in our case the wall oscillates with the frequency ω of the external field, we will follow Rado[4] in the calculation of the mass. We are interested in the special case of a 180-degree wall. However, it is simpler (formally) to consider a 90-degree wall, as Rado did. The mass of a 180-degree wall is equal to twice the mass of a 90-degree wall.

The energy W of a wall layer in a cubic crystal, associated with 1 cm^2 of its surface, is given by the formula

$$W = \int_{-\infty}^{\infty} d\xi\delta\left\{\frac{Aa^2n}{4}\left[\frac{1}{1-\alpha^2}\left(\frac{\partial\alpha}{\partial\xi}\right)^2\frac{1}{\delta^2}+(1-\alpha^2)\left(\frac{\partial\varphi}{\partial\xi}\right)^2\frac{1}{\delta^2}\right]\right.$$
$$+K[(1-\alpha^2)\alpha^2+(1-\alpha^2)^2\cos^2\varphi\sin^2\varphi]$$
$$+[2\pi I_s^2(\alpha-\alpha_\infty)^2+I_sH_0e^{i\omega t}(1-\alpha^2)^{1/2}$$
$$\left.\times\, 2^{-1/2}(\sin\varphi-\cos\varphi)]\right\}, \tag{5}$$

where the first term is the exchange-interaction energy, the second is the magnetic anisotropy energy, and the third is the energy of interaction with the magnetic field. The dimensionless parameter ξ is $(x-X)/\delta$, where X is the coordinate of the center of the wall layer; φ and θ $(\alpha \equiv \cos\theta)$ are the polar angles of the direction of the mean magnetization in the wall layer, with respect to the normal to the wall as polar axis. The magnetic field is oriented as in Rado's paper[4] [Fig. 1, formula (16)].

Following Rado,[4] we seek a solution for α and φ in the form

$$\alpha(\xi, t) = \alpha_0(\xi) + \alpha_1(\xi)\, pe^{i\omega t},$$
$$\varphi(\xi, t) = \varphi_0(\xi) + \varphi_1(\xi)\, pqe^{i\omega t},$$
$$v(t) = 4\pi I_s\gamma\delta V_0 pe^{i\omega t}. \tag{6}$$

Without repeating the calculations,[4] we give the solutions for α_0 and φ_0,

$$\alpha_0(\xi) = 0,\quad \varphi_0(\xi) = \operatorname{arctg} e^{\xi} \tag{7}*$$

and the equation for α_1,

$$\left(\frac{d\alpha_1}{d\xi}\right)^2 - \alpha_1\left(1+\frac{1}{\lambda}-3\sin^2\varphi_0+3\sin^4\varphi_0\right)$$
$$=\frac{V_0}{\lambda}\sin\varphi_0\cos\varphi_0. \tag{8}$$

We now substitute (6) in (5) and expand W in powers of p and q (for our ferrite, at resonance frequency $\omega = 2\pi \times 360$ Mc/sec, if we take $H_0 = 0.1$ oe, we get $p = 0.78 \times 10^{-3}$ and $q = 0.64 \times 10^{-2}$). On neglecting terms $\sim p^4$ in comparison with terms $\sim p^2$ and on neglecting q^2 in comparison with 1, we get

$$W = W_0 + mv^2/2,$$

$$m = \frac{1}{8\pi\gamma^2\delta}\frac{2\lambda}{V_0}\int_{-\infty}^{\infty} d\xi\left\{\left(\frac{d\alpha_1}{d\xi}\right)^2\right.$$
$$\left.+\alpha_1^2\left(1+\frac{1}{\lambda}-3\sin^2\varphi_0\cos^2\varphi_0\right)\right\} \tag{9}$$

or, if we use (7) and change the variable of integration,

$$m = \frac{1}{8\pi\gamma^2\delta}\frac{2\lambda}{V_0}\int_0^{\pi} d(2\varphi_0)\left\{\left(\frac{d\alpha_1}{d2\varphi_0}\right)^2\sin 2\varphi_0\right.$$
$$\left.+\alpha_1^2\left(\frac{1+1/\lambda}{\sin 2\varphi_0}-\frac{3}{4}\sin 2\varphi_0\right)\right\}. \tag{10}$$

In order to calculate m by this formula, it is necessary to know the solution of (8) for α_1. Rado solved this equation after setting $\lambda = 0$. In our case, however, λ is not small ($\lambda = 4$, $K = 4 \times 10^6$ erg/cm^3, $I_S = 400$ oe). Therefore it is not possible to solve (8) analytically. We use a variational method. The solution of (8) corresponds to the problem of searching for the minimum of the following functional:

$$S = \int_0^{\pi} d(2\varphi_0)\left\{\left(\frac{d\alpha_1}{d2\varphi_0}\right)^2\sin 2\varphi_0\right.$$
$$\left.+\alpha_1^2\left(\frac{1+1/\lambda}{\sin 2\varphi_0}-\frac{3}{4}\sin 2\varphi_0\right)-\frac{V_0}{\lambda}\frac{d\alpha_1}{d2\varphi_0}2\varphi_0\right\} \tag{11}$$

(here a change of the variable of integration, as in (10), has already been introduced). Now we seek $\alpha_1(\xi)$ in the form of a series in powers of $\sin 2\varphi_0$ (since α_1 is an even function of ξ),

$$\alpha_1(\xi) = C_1\sin 2\varphi_0 + C_2\sin^2 2\varphi_0 + C_3\sin^3 2\varphi_0 + \ldots \tag{12}$$

We determine the variable parameters C_1, C_2, C_3 from the condition that S be a minimum. Then on substituting (12) in (10), we get for $\lambda = 4$ the following value of the mass of a 90-degree wall (to within 1 in the third figure):

$$m_{90^\circ} = 0.24 / 8\pi\gamma^2\delta. \tag{13}$$

The value of the mass of a 180-degree wall, as has been mentioned already, is twice as large.

It is now still necessary to calculate the wall-layer thickness parameter $\delta = (Aa^2n/4K)^{1/2}$, and for this purpose it is necessary to know n and A. The number of spins in unit volume, n, can be determined according to the formula $n = I_S/\mu_B$, where μ_B is the Bohr magneton. We then get for n the value $n = 4.4 \times 10^{22}$ cm^{-3} ($I_S = 400$ oe). In this way, however, one generally gets a low value of n, since in a ferrite not all the spins are parallel.

*arctg = $\tan^{-1}$.

The mean value of the exchange integral A is determined by the formula

$$A = \frac{2S}{2a^2} \sum_{k \neq i} J_{ik} (x_i - x_k)^2,$$

where J_{ik} is the exchange integral of interacting atoms i and k, and where 2S is the number of spins per atom. On taking account only of interactions with nearest neighbors and on assuming them equal, we get

$$A = SJ\,(\Delta x)^2 z a^{-2},$$

where z is the number of nearest neighbors.

We estimate the exchange integral J from the condition that the kinetic energy at the Curie point is equal to the energy of exchange interaction (for one atom): $2SkT_C = 2JS^2z$. Hence

$$J = kT_c/Sz, \qquad A = kT_c\,(\Delta x)^2/a^2.$$

We take Δx in a ferrite equal to a/4. Consequently $\delta = 3.6 \times 10^{-7}$ cm ($K = 4 \times 10^6$ erg/cm^3, $a = 8.4 \times 10^{-8}$ cm, $T_C = 770°$K).

By formula (13)

$$m_{180°} = 0.48/8\pi\gamma^2\delta = 1.7 \cdot 10^{-10} \text{ g/cm}^2.$$

By formula (3)

$$\alpha_{180°} = 4I_s^2/\chi_0 l = 1.6 \cdot 10^9 \text{ g/cm}^2\,\text{sec}^2$$

($\chi_0 = 1/4\pi$, $l = 5 \times 10^{-3}$ cm; we take the mean value of the domain width, measured on photographs of powder patterns).

We now finally determine the value of the resonance frequency ω_0:

$$\omega_0 = \sqrt{\alpha/m} = 2\pi \cdot 500 \text{ Mc/sec}.$$

In view of the fact that some of the initial quantities in the determination of ω_0 are known only to one figure, the agreement with the experimental value $\omega_0 = 2\pi \times 36$ Mc/sec must be considered completely satisfactory.

The authors express their sincere thanks to K. V. Vladimirskii for advice and for discussion of the results of the research.

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Translated by W. F. Brown, Jr.
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