EFFECT OF SPONTANEOUS STRICTION ON ANTIFERROMAGNETIC RESONANCE IN HEMATITE

A. S. BOROVIK-ROMANOV and E. G. RUDASHEVSKIĬ

Physical Problems Institute, Academy of Sciences, U.S.S.R.

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An experimental study is made of the effect of compression and extension in the basal plane upon the dependence of the antiferromagnetic resonance frequency in hematite on the external magnetic field. It is observed that the direction of shift of the line upon deformation of the specimen in the basal plane depends on the mutual orientation of the applied force and of the external magnetic field. A theoretical calculation is carried out on the basis of the complete Hamiltonian for rhombohedral antiferromagnetics possessing weak ferromagnetism, with elastic and magnetoelastic terms taken into account. It is shown that in the resonance spectrum of such a magnetic structure there may be present a gap, independent of the directions, which can be changed by deforming the crystal perpendicular to the threefold axis. The results of the theoretical calculation are in agreement with experiment.

1. INTRODUCTION

As is known, hematite (α-Fe_2O_3) is a rhombohedral antiferromagnetic possessing a weak ferromagnetic moment[1-3]. One of the branches of the spin-wave spectrum in such a magnetic structure should have a very small energy gap in zero external magnetic field H[4,5]. Therefore antiferromagnetic resonance in hematite at frequencies in the centimeter range was observed in fields of the order of a few kilo-oersteds[6,7]. The theoretical dependence of the antiferromagnetic resonance frequency ν (for the low-frequency branch) on an external magnetic field H, applied in the basal plane, has the form[4,5,8]

$$(\nu/\gamma)^2 = H(H + H_D) + H_\Delta^2 \cos 6\varphi, \qquad (1)$$

where γ is the gyromagnetic ratio, H_D is the Dzyaloshinskiĭ field responsible for the weak ferromagnetism, and H_Δ^2 is the hexagonal anisotropy that appears when account is taken of the small anisotropy in the basal plane (ϕ is the angle between the direction of the field and one of the twofold axes).

Experimental studies, carried out on very perfect synthetic monocrystals with narrow absorption lines ($\Delta H \sim 60$ to 150 Oe)[9,10], showed that actually this dependence has the form

$$(\nu/\gamma)^2 = H(H + H_D) + H_\Delta^2, \qquad (2)$$

where $H_D = 22$ kOe, $H_\Delta^2 = 13.7 \pm 0.4$ kOe^2, and H_Δ^2 is independent of the direction of the external magnetic field applied in the basal plane; that is, it is not connected with the hexagonal anisotropy. The hypothesis was proposed[9,10] that the experimentally observed gap H_Δ^2 is caused by spontaneous striction of the hematite, the magnitude of which is quite large[11]. Then there appears in the basal plane a preferred direction, connected with the direction of the external magnetic field by virtue of the fact that the spins in the basal plane, in consequence of the small value of the anisotropy, always set themselves perpendicular to the field, and the direction in which the specimen becomes deformed in consequence of spontaneous striction is connected with the direction of the spins.

In order to test this hypothesis, the experiments described below were performed.[1)] It was expected that subjection of the specimen to a deformation in the basal plane, of the order of magnitude of the spontaneous striction, could appreciably change H_Δ^2.

2. SPECIMENS AND EXPERIMENT

In this work we used synthetic monocrystals of hematite, grown by M. Vihr in the Institute of the Physics of Solids of the Czechoslovak Academy of Sciences. The specimens were thin rectangular plates with dimensions $0.7 \times 0.4 \times 0.05$ mm (cf. Fig. 1). The plane of the plates coincided with the basal plane of the crystal.

The measurements were made on a direct-am-

1)Preliminary results of this work were presented at the Eleventh All-union Conference on the Physics and Technology of Low Temperatures, June 1964, Minsk.

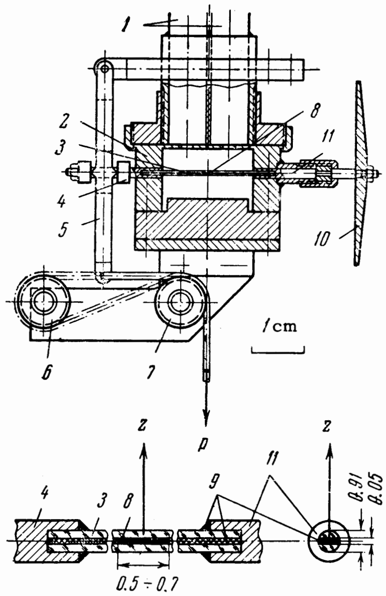

FIG. 1. Apparatus for compression of specimens in the resonator (for symbols, consult the text). The lower part of the figure shows, enlarged, the specimen glued between quartz half-cylinders.

plification magnetic spectrometer with low-frequency modulation of the magnetic field, at wavelengths 0.86, 1.2, and 1.4 cm. As was pointed out above, it was necessary to subject the specimens to a deformation in a direction perpendicular to the threefold axis. At small specimen dimensions, this was quite difficult and required construction of the special apparatus shown in Fig. 1. The high-frequency power was propagated through the waveguides 1 and entered the resonator 2 (H_{011} mode). The high-frequency magnetic field at the center of the resonator was always directed vertically. The constant magnetic field lay in the horizontal plane; thus on rotation of the constant magnetic field in the horizontal plane, its perpendicularity to the microwave field was always preserved.

The specimen 8 was glued with Canada balsam 9 between two quartz half-cylinders 3, which in turn were glued inside the bushings 4 and 11. Special attention was paid to the accuracy of the gluing, in order that the axes of the bushings 4 and 11 might coincide. For this purpose the gluing was carried out in a special press. It should be mentioned that in the course of the work, the gluing was done with various lacquers and glues, but that on hardening they gave appreciable shrinkage (in comparison with the value of the spontaneous striction, $\Delta l/l \sim 10^{-5}$), which led to nonuniform deformation of the specimen and to broadening of the absorption line. The use of Canada balsam proved most convenient because of its fluidity. The broadening of the line was the criterion by which the gluing of the specimen was judged: if the line widths of the specimen, when it was not glued between the quartz half-cylinders and when it was, agreed (to within a few percent), then it could be considered that the specimen was initially not subjected to the effect of compression and extension. With gluing by means of Canada balsam, nonuniform deformations could be removed by a slight heating by hot air (up to +50°C) directly in the resonator.

By means of the lever 5, it was possible to compress or extend (depending on whether the pulling was applied over pulley 6 or 7) the quartz rod, and with it the specimen glued into it, by suspending various weights p. By turning the dial 10, it was possible to set the basal plane of the specimen horizontal and, consequently, to direct the external magnetic field at an arbitrary angle to the applied force, in such a way that both the magnetic field and the force always lay in the basal plane.

3. RESULTS

The measurements were carried out with various mutual orientations of the force and the field, both applied in the basal plane. The curves in Fig. 2 show the dependence of the amount of shift of the resonance line on the external force applied to the affixed quartz rod (λ = 1.4 cm). Similar curves were observed at other wavelengths. The largest observed shift of the absorption line amounted to about ±1000 Oe; but in the range of such large shifts, the relation between the external force and the change of position of the line ceased to be linear, because of insufficiently good transmission of the deformation from the quartz half-cylinders through the thin layer of Canada balsam to the specimen. It was observed that the sign of the shift was different, depending on whether the direc-

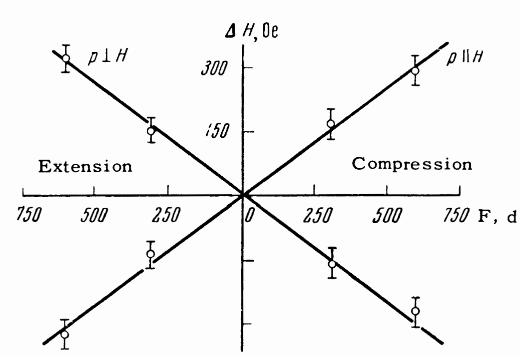

FIG. 2. Dependence of the shift of the antiferromagnetic resonance line on the force applied to the quartz rod, for two mutual orientations of the external magnetic field and the force (the measurements were made at frequency 21.61 Gcps; the unshifted line was observed in field 1.92 kOe).

tion of the external magnetic field was perpendicular or parallel to the direction of the applied force (cf. Fig. 2).

It should be mentioned that in these experiments it was impossible to determine the value of the deformation of the crystal corresponding to a given value of the applied force. This fact was connected with the already mentioned fluidity of the Canada balsam. On the one hand, this fluidity insured the possibility of gluing the specimen into the holder without stress. On the other hand, it led to the result that the deformation of the quartz holder was not completely transmitted to the specimen. As was shown by estimates that were made (cf. below), the transmission coefficient was about 0.1. Furthermore, this fluidity manifested itself in the fact that the initial specimen deformation, produced at the instant of loading, gradually decreased with time if the load was held constant, and the absorption line slowly shifted toward the unloaded position. However, at the time of recording of the absorption line, this shift with time did not exceed 2% of the value of the initial shift.

4. DISCUSSION OF RESULTS

To explain the results obtained, it is necessary to calculate the energy spectrum of the crystal, taking account of the elastic and magnetoelastic energy.

The Hamiltonian for an antiferromagnetic of α-Fe_2O_3 type (space group D_{3d}^6), with allowance for magnetoelastic and elastic terms (without allowance for anisotropy in the basal plane, which is small at room temperature[10]), has the form

$$\begin{aligned}\mathcal{H} &= {}^1/_2 B\mathbf{m}^2 + {}^1/_2 b m_z^2 + {}^1/_2 \tilde{a} l_z^2 + \tilde{q}(l_x m_y - l_y m_x) \\ &+ \lambda_3[4 l_x l_y u_{xy} + (l_x^2 - l_y^2)(u_{xx} - u_{yy})] \\ &+ \lambda_4[2 l_x l_y u_{xz} + (l_x^2 - l_y^2) u_{yz}] + \lambda_5 (l_x^2 + l_y^2)(u_{xx} + u_{yy}) \\ &+ \lambda_6 (l_x^2 + l_y^2) u_{zz} + \lambda_7 [l_x l_z u_{xz} + l_y l_z u_{yz}] \\ &+ \lambda_8 [2 l_x l_z u_{xy} + l_y l_z (u_{xx} - u_{yy})] + {}^1/_2 \mu_1 u_{zz}^2 \\ &+ 2\mu_2 (u_{xx} + u_{yy})^2 + \mu_3[(u_{xx} - u_{yy})^2 + 4u_{xy}^2] \\ &+ 2\mu_4 (u_{xx} + u_{yy}) u_{zz} + 4\mu_5 (u_{xz}^2 + u_{yz}^2) \\ &+ 4\mu_6[(u_{xx} - u_{yy}) u_{yz} + 2u_{xy} u_{xz}] - \sigma_{ij} u_{ij} - \mathbf{mh}. \qquad (3)\end{aligned}$$

As usual (cf., for example,[8]), the z axis is directed along the threefold axis of the crystal, the x axis along one of the axes of second order in the basal plane; $\mathbf{m} = (\mathbf{M}_1 + \mathbf{M}_2)/2M_0$, $\mathbf{l} = (\mathbf{M}_1 - \mathbf{M}_2)/2M_0$; $\mathbf{M}_1$ and $\mathbf{M}_2$ are the sublattice magnetizations ($M_1^2 = M_2^2 = M_0^2$); u_{ij} are the deformations, σ_{ij} are the stresses acting on the crystal; $\mathbf{m}\cdot\mathbf{h} = 2M_0\mathbf{m}\cdot\mathbf{H}$ is the energy of the antiferromagnetic in the external magnetic field (the field is always directed in the basal plane);

$$\tilde{a} = a + \lambda_1'(u_{xx} + u_{yy}) + \lambda_2 u_{zz} = a + \lambda_1' \sum u_{ii} + \lambda_2' u_{zz},$$

$$\tilde{q} = q + \lambda_9 (u_{xx} + u_{yy}) + \lambda_{10} u_{zz} = q + \lambda_9' \sum u_{ii} + \lambda_{10}' u_{zz},$$

where $\Sigma u_{ii} = \Delta v/v$ is the relative change of volume.

By minimizing the expression (3) with respect to the variables u_{ij}, it is possible to find the equilibrium values $u_{ij}^{(0)}$ at given external force p ($\sigma_{ik} = p n_i n_k$, where $\mathbf{n}$ is the unit vector in the direction of p) and at given values of l_i (the values of l_i are determined by the direction of the external magnetic field: $\mathbf{l}$ is always perpendicular to $\mathbf{H}$[1]). We write out some of these, supposing that p = 0 (the spontaneous striction). For $u_{zz}^{(0)}$ we have the following expression:

$$u_{zz}^{(0)} = \frac{\mu_4\lambda_5 - 2\mu_2\lambda_6}{2(\mu_1\mu_2 - \mu_4^2)} + \frac{\mu_4(\lambda_1 - \lambda_5) - 2\mu_2(\lambda_2 - \lambda_6)}{2(\mu_1\mu_2 - \mu_4^2)} l_z^2.$$

In the case of hematite, a > 0, and on the basis of the Hamiltonian written above, $l_z = 0$; but if we take account of terms of higher order in $\mathbf{l}$[1,8], then $l_z \neq 0$, and a spontaneous striction along the axis, proportional to l_z^2, will be observed. Actually, in work of Urquhart and Goldman[11] on the temperature dependence of u_{zz} near the point of transition from the state with weak ferromagnetism to the antiferromagnetic state without weak ferromagnetism (spins directed along the threefold axis), there is observed a small increase, connected with the turning of the spins out of the basal plane (cf. also[10]). However, at room temperature the value of l_z is small, and therefore in further calculations we will consider that $l_z = 0$. We then get the following values for $u_{xx}^{(0)}$ and $u_{yy}^{(0)}$:

$$u_{xx}^{(0)} = \frac{2\mu_4\lambda_6 - \mu_1\lambda_5}{8(\mu_1\mu_2 - \mu_4^2)} + \frac{\lambda_4\mu_6 - 2\lambda_3\mu_5}{8(\mu_3\mu_5 - \mu_6^2)} (l_x^2 - l_y^2), \qquad (4a)$$

$$u_{yy}^{(0)} = \frac{2\mu_4\lambda_6 - \mu_1\lambda_5}{8(\mu_1\mu_2 - \mu_4^2)} - \frac{\lambda_4\mu_6 - 2\lambda_3\mu_5}{8(\mu_3\mu_5 - \mu_6^2)} (l_x^2 - l_y^2). \qquad (4b)$$

In the cited work[11], it was shown that the striction in the direction of the axis of second order in the basal plane depends on the direction of the applied field in the following manner: $\Delta l/l = A \sin^2\phi$, where ϕ is the angle between the magnetic field in the basal plane and the axis of second order. That result agrees with the expression (4a) deduced by us. In fact, for the deformation measured in the experiment we have

$$\left(\frac{\Delta l}{l}\right)_x = u_{xx}^{(0)}(\varphi) - u_{xx}^{(0)}(\varphi = 0)$$

$$= \frac{2\lambda_3\mu_5 - \lambda_4\mu_6}{4(\mu_3\mu_5 - \mu_6^2)} \sin^2\varphi = A\sin^2\varphi. \qquad (4c)$$

Exactly the same expression can be obtained for the case in which $\Delta l/l$ is measured in an arbitrary direction in the basal plane; but in this case ϕ will denote the angle between the direction in which the striction is measured and the magnetic field.

We consider small oscillations of **m** and **l** about the equilibrium values $\mathbf{m}_0$ and $\mathbf{l}_0$, and we determine the characteristic frequencies of these oscillations[4,5,12]. In this we will suppose that sound attentuates strongly at these frequencies and therefore is not propagated in the crystal ($\partial u_{ij}/\partial t = 0$). Then after substitution of the equilibrium values $u_{ij}^{(0)}$ in the characteristic values of the frequencies, we have

$$(\omega_1/\gamma)^2 = H(H+H_D) + 2H_{\Delta 1}^2 + 2H_A'(\mathbf{p})H_E, \quad (5a)$$

$$(\omega_2/\gamma)^2 = 2H_AH_E + H_D(H+H_D) + H_{\Delta 1}^2 + H_A'(\mathbf{p})H_E + H_{\Delta 2}^2 + H_A''(\mathbf{p})H_E; \quad (5b)$$

$$H_D = \tilde{q}/2M_0, \quad 2H_AH_E = \tilde{a}B/4M_0^2, \quad (6a)$$

$$H_{\Delta 1}^2 = \frac{B}{4M_0^2}\frac{4\lambda_3^2\mu_5 + \lambda_4^2\mu_3 - 4\lambda_3\lambda_4\mu_6}{4(\mu_3\mu_5 - \mu_6^2)}(l_x^2 + l_y^2)^2, \quad (6b)$$

$$H_A'H_E = p\frac{B}{4M_0^2}\left[\frac{\lambda_4\mu_3 - 2\lambda_3\mu_6}{4(\mu_3\mu_5-\mu_6^2)}\sin 2\eta \sin(\xi+\varphi) - \frac{\lambda_4\mu_6 - 2\lambda_3\mu_5}{4(\mu_3\mu_5-\mu_6^2)}\sin^2\eta\cos^2(\xi-\varphi)\right], \quad (6c)$$

$$H_{\Delta 2}^2 = \frac{B}{4M_0^2}\frac{4\lambda_6\mu_2 + \lambda_5^2\mu_1 - 4\lambda_5\lambda_6\mu_4}{2(\mu_1\mu_2 - \mu_4^2)}(l_x^2 + l_y^2)^2, \quad (6d)$$

$$H_A''H_E = p\frac{B}{4M_0^2}\left[\frac{\mu_4\lambda_5 - 2\mu_2\lambda_6}{4(\mu_1\mu_2-\mu_4^2)}\cos^2\eta - \frac{\mu_1\lambda_5 - 2\mu_4\lambda_6}{4(\mu_1\mu_2-\mu_4^2)}\sin^2\eta\right]; \quad (6e)$$

ϕ is the angle between one of the axes of second order in the basal plane and the external magnetic field, ξ and η are the azimuthal and polar angles of the unit vector in the direction of the applied force **p**.

Thus we see that in the dependence of the antiferromagnetic resonance frequency on the field, there is in the first branch a gap $2H_{\Delta 1}^2$ independent of its direction, and in the second $H_{\Delta 1}^2 + H_{\Delta 2}^2$ (if we suppose that $l_x^2 + l_y^2 \approx 1$, which is so at room temperature). Such a gap in the first (low-frequency) branch was observed experimentally earlier[9,10]. Formula (5) for p = 0 agrees with formula (2).

We consider the effect of compression and extension on the low-frequency branch of the antiferromagnetic resonance. $2H_A'H_E$ is the gap that appears upon deformation of the specimen. We study three possible mutual orientations of the applied force and the external magnetic field (the magnetic field is always located in the basal plane). From (6c) we have:

1) for $\mathbf{p} \parallel \mathbf{H}$, $\xi - \varphi = 0$, $\eta = \pi/2$,

$$2H_A'H_E = -p(B/4M_0^2)A = -pA/\chi \quad (7a)$$

(A is the amplitude of the spontaneous striction [cf. (4c)], χ is the static susceptibility);

2) for $\mathbf{p} \perp \mathbf{H}$, $\xi - \varphi = \pi/2$, $\eta = \pi/2$,

$$2H_A'H_E = p(B/4M_0^2)A = pA/\chi; \quad (7b)$$

3) for **p** parallel to the threefold axis, $\eta = 0$,

$$2H_A'H_E = 0. \quad (7c)$$

Thus we see that the sign of the shift of the absorption line in the low-frequency branch depends on the mutual orientation of the applied force and the external magnetic field. The deductions of the theory [formulas (7a) and (7b)] are in agreement with the experimental data (Fig. 2). On compression or extension along the threefold axis, the gap does not change (7c). The line may nevertheless shift, since such a deformation may somewhat change H_D[3,10]; but in this case the shift of the line will not obey relations of the form (7a) and (7b).

For the low-frequency branch of the antiferromagnetic resonance, which we have studied, an estimate of the value of $2H_A'H_E$ was made on the basis of the data of Urquhart and Goldman[11]. The shift of the absorption line should exceed that observed by a factor of about ten, if we were able to transmit the deformation completely from the quartz rod to the specimen. This did not occur because of the fluidity, mentioned above, of the Canada balsam. There is, however, complete qualitative agreement. For estimation of the value of $H_{\Delta 1}^2$ it is necessary to know the elasticity-modulus tensor, which is unknown.

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