# CRITICAL FIELDS AND RESONANCE IN AN EASY-AXIS ANTIFERROMAGNET WITH

## DZYALOSHINSKIĬ INTERACTION

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A classical calculation is carried out for the simplest model of an easy-axis antiferromagnet with Dzyaloshinskiĭ interaction of the form  $\beta(m_X l_y - m_y l_x)$ , for arbitrary orientation of the external magnetic field with respect to the axis of easy magnetization. It is shown that in the case  $H_A \ll H_D \ll H_E$  (where  $H_A$ ,  $H_D$ , and  $H_E$  are the anisotropy, Dzyaloshinskiĭ, and exchange fields, respectively), the boundary between the phases with  $l_z \neq 0$  and with  $l_z = 0$  (l is the antiferromagnetic vector) in the ( $H_X$ ,  $H_Z$ ) plane is the circle (18a), whose centerposition and radius depend on the temperature. On this boundary, one of the antiferromagnetic resonance frequencies vanishes. In particular, for  $H \perp z$  the frequency  $\omega_{10} = \gamma H_{C\parallel} (1 - H_X^2/H_{C\perp}^2)^{1/2}$  when  $H_X \leq H_{C\perp}$  and  $\omega_{10} = \gamma H_{C\parallel} (H_X/H_{C\perp} - 1)^{1/2}$  when  $H_X \gtrsim H_{C\perp}$ , where  $H_{C\parallel} = (2H_AH_E - H_D^2)^{1/2}$  and  $H_{C\perp} = (2H_AH_E - H_D^2)/(H_A^2 + H_D^2)^{1/2}$ . An experimental verification of the relations obtained has been undertaken by studying antiferromagnetic resonance in an artificial monocrystal of hematite ( $\alpha - Fe_2O_3$ ) at T = 77 K at wavelengths 4, 6, and 8 mm. Satisfactory agreement with the calculation is obtained with  $2H_A = 0.54$  kOe,  $H_D = 30 \pm 4$  kOe, and  $\frac{1}{2}H_E = 4500$  kOe.

#### 1. INTRODUCTION

 ${f FOR}$  a description of the fundamental properties of a classical two-sublattice antiferromagnet with anisotropy of the "easy axis" (EA) type, for example MnF<sub>2</sub>, knowledge of two of its numerical properties is sufficient in a first approximation: the exchange field H<sub>E</sub> and the anisotropy field HA. With their aid there follows from a simple phenomenological theory, as a rule, satisfactory agreement with experiment, at  $T \ll T_N$ , for the magnetization curves m(H) and for the frequency spectrum  $\omega_{n_0}(H)$ , n = 1 and 2, of antiferromagnetic resonance (AFMR) for parallel and perpendicular orientation of the external magnetic field H with respect to the axis of easy magnetization (see, for example,<sup>[1]</sup>). Furthermore, in principle a calculation of the equilibrium configuration of the sublattice moments is possible for arbitrary orientation of H with respect to the EA<sup>[2]</sup>. Allowance for interactions of higher order-biguadratic exchange, anisotropy in the basal plane, etc.-is necessary in order to describe finer details of the magnetic behavior of an antiferromagnet, for example hysteresis by "collapse" of the sublattices<sup>[3]</sup>, small splitting of the AFMR frequencies, etc.

At the same time, the well-known Dzyaloshinskii interaction (DI) has as a rule not been taken into account in the description of easy-axis antiferromagnets (even if it is allowed by the crystal symmetry), although in order of magnitude its energy is comparable with the anisotropy energy. This is due to the fact that, for example, a DI of the form  $\beta(m_X l_y \pm m_y l_X)$  (in the generally accepted symbols), with  $l_Z = 1$  ( $z \parallel EA$ ), does not manifest itself clearly in small magnetic fields. The necessity for taking it into account in order to describe the behavior of an antiferromagnet in large fields, perpendicular to the EA, was noticed by one of the authors<sup>[4,5]</sup> in a study of the properties of CoF<sub>2</sub>. In particular, it was shown that when

$$H_x := H_{c\perp} \equiv (2H_A H_E - H_D^2) / \gamma H_A^2 + H_D^2$$
(1)

( $H_D$  is the Dzyaloshinskiĭ field), there occurs in CoF<sub>2</sub> a phase transition, connected with the sudden (though still continuous) disappearance of the z component of the antiferromagnetic vector *l*.

A similar anomaly was detected<sup>[6-9]</sup> in investigation of the magnetization curves of low-temperature ( ${f T} < {f T}_M,$  where  ${f T}_M$  is the Morin point) hematite,  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub>, with **H**  $\perp$  EA; in contrast to the high-temperature phase, it is an antiferromagnet with anisotropy of the "easy axis" type. In these researches, the investigation was carried out in a temperature range close to  $T_{M}$  (the point of spontaneous reorientation of the antiferromagnetic vector). In a phenomenological description of the phenomenon, besides the DI, parameters connected with a fourth-order interaction were introduced, and relative agreement with experiment was obtained by choice of their values [6,9]. In an early stage of the calculation, some small quantities were neglected, and this led to definite disagreements with experiment (for example, to the prediction of an unobserved phase transition at fields larger than  $H_{c\perp}$ ).

In the first part of the present paper, a phenomenological calculation is undertaken of the static and dynamic behavior of the simplest of the conceivable antiferromagnetic systems with anisotropy of the "easy axis" type and with a nonvanishing DI. The case of arbitrary orientation of the external field with respect to the EA is considered. The system is described by only three numerical parameters:  $H_E$ ,  $H_A$ , and  $H_D$ ; the calculation, however, is carried out exactly--small quantities are neglected only in a special case and only in the final stage.

In the second part, AFMR is investigated experimentally in an artificial monocrystal of  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> at a temperature T  $\ll$  T<sub>M</sub>, with the aim of clarifying the possibility of describing this antiferromagnet within the framework of the exactly calculated simplest mode.

 $h_x$ 

#### 2. PHENOMENOLOGICAL THEORY

H

1. We consider, from the point of view of classical theory, the simplest antiferromagnet with anisotropy of the "easy axis" type and with nonvanishing DI. We direct the z axis along the EA and the x axis along the projection of the external field H on the basal plane (this does not impair the generality, since we are neglecting anisotropy in the basal plane).

We have in mind application of the results of the calculation to  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub>, which has, according to Turov's<sup>[1]</sup> terminology, an even antiferromagnetic structure with respect to the principal axis; we therefore write the energy of the system in the form

$$= (2M_0)^2 [\frac{1}{2}Bm^2 + \frac{1}{2}a(l_x^2 + l_y^2) - \beta(m_x l_y - m_y l_x) - m_x h_x - m_z h_z].$$
(2)

Here and below we use the notation:  $M_1$  and  $M_2$  are the sublattice magnetizations,

$$|\mathbf{M}_1| = |\mathbf{M}_2| \equiv M_0; \tag{3}$$

m =  $(M_1 + M_2)/2M_0$  is the ferromagnetic vector;  $l = (M_1 - M_2)/2M_0$  is the antiferromagnetic vector; H<sub>E</sub> = BM<sub>0</sub> is the exchange field (B > 0); H<sub>A</sub> = 2aM<sub>0</sub> is the anisotropy field (a > 0); H<sub>D</sub> = 2 $\beta$ M<sub>0</sub> is the Dzyaloshinskiĭ field ( $\beta$  > 0); H = 2M<sub>0</sub>h. For simplicity, anisotropy of the g-factor is ignored.

In order to find both the resonance frequencies and the equilibrium orientation of the sublattices, we use the equations of motion:

$$\frac{2M_0}{\gamma}\dot{\mathbf{m}} = [\mathbf{m}\mathbf{H}_{\mathbf{m}}] + [\mathbf{l}\mathbf{H}_{\mathbf{l}}], \quad \frac{2M_0}{\gamma}\dot{\mathbf{l}} = [\mathbf{m}\mathbf{H}_{\mathbf{l}}] + [\mathbf{l}\mathbf{H}_{\mathbf{m}}], \quad (4)*$$

where  $H_m = -\partial \mathcal{H}/\partial m$ ,  $H_l = -\partial \mathcal{H}/\partial l$ ;  $\gamma$  is the magnetomechanical ratio. Furthermore, we shall take into account that, in consequence of (3),

$$lm = 0, \quad l^2 + m^2 = 1. \tag{5}$$

2. We find the equilibrium state from (4) by setting  $\dot{\mathbf{m}} = \dot{\mathbf{i}} = 0$ . We get

$$\beta m_x l_z - \beta m_z l_x - a l_y l_z = m_y h_z, \tag{6a}$$

$$ul_x l_z + \beta m_y l_z - \beta m_z l_y = m_z h_x - m_x h_z, \qquad (6b)$$

$$0 = m_y h_x, \tag{6c}$$

$$[(B-a)m_{z} - h_{z}]l_{y} + \beta m_{x}m_{z} - \beta l_{x}l_{z} - Bm_{y}l_{z} = 0, \quad (6d)$$

$$(Bm_{x} - h_{x})l_{z} - [(B - a)m_{z} - h_{z}]l_{x} - \beta l_{z}l_{y} + \beta m_{y}m_{z} = 0, \quad (6e)$$

$$[(B-a)m_x - h_x]l_y + \beta(m_x^2 + m_y^2 - l_x^2 - y^2) - (B-a)m_yl_x = 0.$$
(6f)

Since we are considering the general case  $h_X \neq 0$ , it follows from (6c) that  $m_y = 0$ . The remaining equations, with allowance for (5), permit two solutions.

The first of these describes a state with a nonvanishing z component of the antiferromagnetic vector.

$$m_y = 0, \tag{7a}$$

$$l_y = \beta m^2 / a m_x \quad (m^2 = m_x^2 + m_z^2),$$
 (7b)

$$l_x = -m_z l_z / m_x, \qquad (7c)$$

$$l_{z}^{2} = \frac{m_{z}}{m^{2}} (1 - m^{2}) - \frac{r}{a^{2}} m^{2}, \qquad (70)$$

$$h_{\rm x} = \left( B - \frac{\beta^2}{a} - a \frac{m_{\rm z}^2}{m^4} \right) m_{\rm x}, \tag{7e}$$

$$h_{z} = B - a - \frac{\beta^{2}}{a} + a \frac{m_{x}^{2}}{m^{4}} m_{z}.$$
 (7f)

The second solution describes a state with  $l_z = 0$ :

$$m_y = l_x = l_z = 0, \tag{8a}$$

$$l_y^2 = 1 - m^2,$$
 (0D)

$$= (B-a)m_x - \beta(1-m^2-m_x^2)/\gamma 1-m^2, \qquad (8C)$$

$$h_z = (B-a) m_z + \beta m_x m_z / \sqrt{1-m^2}.$$
 (8d)

3. In order to visualize the range of realization of these states, we consider the simple case  $h_Z = 0$ . Then, in agreement with results obtained earlier[5,10], we find that the phase with  $l_Z \neq 0$  is characterized by the relations

$$m_x = \frac{ah_x}{aB - \beta^2}, \quad l_y = \frac{\beta h_x}{aB - \beta^2}, \quad l_z = \left(1 - \frac{h_x^2}{h_{c\perp}^2}\right)^{\eta_z}, \quad (9)$$

where

$$h_{c\perp} = (aB - \beta^2) / \sqrt{a^2 + \beta^2}.$$
 (1a)

Substitution of these expressions into the formula (2) for the energy, and comparison of the resulting value  $\mathcal{H}_{I}$  with the corresponding value  $\mathcal{H}_{II}$  for the phase with  $l_{z} = 0$ , show that the phase with  $l_{z} \neq 0$  occurs when  $h_{z} \leq h_{c\perp}$ . In field  $h_{x} = h_{c\perp}$  there occurs a continuous transition from one state to the other, during which

$$m_{ex} = a / \sqrt{a^2 + \beta^2}, \quad \ell_{ey} = \beta / \sqrt{a^2 + \beta^2}. \tag{10}$$

On increase of the field  $H_x$  from zero to  $H_{C\perp}$ , the equilibrium state of the  $M_1$  and  $M_2$  sublattices, as is easily seen from formula (9), changes in the following manner (see Fig. 1). The vectors  $M_1$  and  $M_2$  move in planes that make (when  $H_x < H_{C\perp}$ ) a constant angle  $\varphi$  with the x axis:

$$\operatorname{tg} \varphi = H_D / H_A. \tag{11}$$

Meanwhile the angle  $\theta$  changes according to the law<sup>1)</sup>

$$\sin \vartheta = H_x / H_{c\perp}. \tag{12}$$

In a field  $H_x = H_{C\perp}$ , the value of  $\sin \theta$  is 1; and upon further increase of the field, the tips of the vectors  $M_1$  and  $M_2$  move along the arcs  $G_1H_{12}$  and  $G_2H_{12}$ , <u>asymptotically</u> approaching the point  $H_{12}$ ; that is, "collapse" of the sublattices does not occur even at very large fields. The usual "collapse" follows from (9)-(12) when  $H_D = 0$ :  $\varphi = 0$ ,  $H_{C\perp} = 2H_E$ ,  $m_{CX} = 1$ ,  $l_c = 0$ . From the expression for  $h_{C\perp}$  the deduction follows that a necessary condition for an "easy-axis" state of the antiferromagnet under consideration in the

FIG. 1. Diagram illustrating the change of equilibrium configuration of the sublattices on increase of the field **H**, applied in a direction perpendicular to the EA (in an antiferromagnet with nonvanishing DI).



<sup>&</sup>lt;sup>1)</sup>Similar results for the case  $H_A \ll H_D \ll H_E$ , which is realized in hematite when  $T \ll T_M$ , were obtained by Cinader and Shtrikman[<sup>11</sup>].

\* $[\mathbf{m}\mathbf{H}_m] \equiv \mathbf{m} \times \mathbf{H}_m$ .

absence of an external magnetic field, in addition to B > 0 and a > 0, is the inequality

$$B - \beta^2 > 0. \tag{13}$$

In general, substances are in principle possible in which  $h_{C\perp} \leq 0$ . Then there exists a situation in which, thanks to the large value of  $\beta$ , even when the constant a has a positive sign, the sublattices arrange themselves in the absence of a magnetic field not along but perpendicular to the principal axis; that is, outwardly the material behaves like an antiferromagnet with anisotropy of the "easy plane" type.

4. On going over to the general case  $h_Z \neq 0$  and  $h_X \neq 0$ , one can conclude that in the antiferromagnet under consideration in a field  $h \leq h_C$  (that is,  $h_X \leq h_{CX}$  and  $h_Z \leq h_{CZ}$ ), there is a state with  $l_Z \neq 0$ . Transition to the phase with  $l_Z = 0$  occurs upon vanishing of the expression (7d). Hence from (7e) and (7f) it is possible to obtain a curve of the critical fields that separate the two phases. In parametric form it is given by the formulas

$$h_{\rm cx} = \left(B - \frac{\beta^2}{a} - a \frac{m_{\rm cz}^2}{m_{\rm c}^4}\right) m_{\rm cx}, \qquad (14a)$$

$$h_{cz} = \left(B - a - \frac{\beta^2}{a} + a \frac{m_{cx}^2}{m_c^4}\right) m_{cz},$$
 (14b)

$$\beta m_c^2 = a m_{cx} \sqrt{1 - m_c^2}, \qquad (14c)$$

where the role of parameters is played by  $m_{CX}$  and  $m_{CZ}$ , which are related by equation (14c). These expressions are sufficient for numerical construction of the function  $h_{CZ}(h_{CX})$  for arbitrary values of  $\alpha$ ,  $\beta$ , and B (satisfying, of course, the inequality (13)). Substitution of the basic condition (14c), which determines the transition between phases, into formulas (8c) and (8d) of the second solution gives the same result for  $h_{CX}$  and  $h_{CZ}$ . Furthermore, it can be shown that when  $h_X = h_{CX}$  and  $h_Z = h_{CZ}$ , the values of the energies of the two phases ( $\mathscr{H}_{IC}$  and  $\mathscr{H}_{IIC}$ ) also coincide. This indicates that the transition between phases occurs in a continuous manner (we recall that we are considering the case  $h_X \neq 0$ ).

The magnetization curves for the two phases (or more accurately, the relations inverse to these) can easily be constructed numerically according to formulas (7e) and (7f) for the phase with  $l_z \neq 0$  and (8c) and (8d) for the phase with  $l_z = 0$ . Here it should be taken into account that the third term in (7e) and the fourth in (7f), proportional to the usually small quantity  $a \ll B$ , gives a contribution that is not small in comparison with the other terms at small values of the magnetization m.

5. This calculation of the equilibrium configuration of the sublattices in a field  $h(h_X, 0, h_Z)$  is completely applicable also to an easy-axis antiferromagnet with odd antiferromagnetic structure with respect to the principal axis (such as  $CoF_2$ ), if the x axis is so chosen that the DI energy is described in the form  $-\beta(m_X l_y + m_y l_X)$ . With such an orientation of the external field, the difference in the form of the DI energy has no effect, since by symmetry  $m_y = 0$  for all fields (the behavior of  $CoF_2$  in a field  $H(H_X, 0, H_Z)$  was considered in detail  $in^{[12]}$ ).

6. As has already been mentioned, this calculation is applicable to the case of arbitrary values of a,  $\beta$ ,

and B within the limits of the inequality (13). Its results, however, are quite difficult to present graphically. Under certain special relations between these basic parameters of the theory, a number of relations are obtained in explicit form. Thus if

$$a \ll \beta \ll B, \tag{15}$$

then, as follows from (7e) and (7f) and from (8c) and (8d), the transition from the first to the second phase occurs for all directions of the field at very small values of the magnetization. For example, when  $h_z = 0$  we have  $m_{CX} = a/\sqrt{a^2 + \beta^2} \approx a/\beta \ll 1$ , and when  $h_X = 0$  we get

$$m_{cz} \approx \sqrt{\frac{a}{B} - \frac{\beta^2}{B^2}} \ll 1, \quad m_{cx} \approx \frac{\beta}{B} \ll 1.$$

The presence of a small parameter  $m \ll 1$  appreciably facilitates the calculation not only of the static, but also of the dynamic properties of an antiferromagnet of the type considered. This is all the more interesting, because the indicated relation between the parameters, as follows from our preliminary communication<sup>[10]</sup>, is satisfied for such a well-known antiferromagnet as  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub>:2H<sub>A</sub> = 0.54 kOe, H<sub>D</sub> = 30 kOe  $\frac{1}{2}$ H<sub>E</sub> = 4500 kOe (at T = 77°K).

In this case we have at the phase boundary in the  $(h_x, h_z)$  plane, from (14c),

$$m_c^2 \approx -\frac{a}{\beta} m_{cx};$$
 (16)

on the other hand, it follows from (8c), (8d), and (15) that

$$m_{cx} \approx \frac{\beta + h_{cx}}{B}, \quad m_{cz} \approx \frac{h_{cz}}{B}.$$
 (17)

If we now substitute (16) and (17) into  $m_c^2 \equiv m_{CX}^2 + m_{CZ}^2$ , we get the equation of the curve in the  $(h_X, h_Z)$  plane that separates the phases with  $l_Z \neq 0$  and with  $l_Z = 0$ :

$$h_{cz}^{2} + \left(h_{cx} - \frac{aB - 2\beta^{2}}{2\beta}\right)^{2} = \left(\frac{aB}{2\beta}\right)^{2}.$$
 (18)

This curve is no other than a circle with center  $S = (aB - 2\beta^2)/2\beta$  on the  $h_x$  axis and radius  $R = aB/2\beta$  (see curve  $T_1$  in Fig. 2). It cuts off intercepts on the z axis  $\pm h_{C||} = \pm (aB - \beta^2)^{1/2}$ , and intercepts on the x axis  $h_{C\perp} = (aB - \beta^2)/\beta$  and  $-h_D = -\beta$ . A physically



FIG. 2. Phase diagrams in the  $(H_X, H_Z)$  plane for an easy-axis antiferromagnet with DI at different temperatures  $T_1 < T_2 < T_3$  and in the case  $H_A \ll H_D \ll H_E$ . The curve  $T_1$  was drawn for the value  $H_{C\perp}/H_{C\parallel} =$ 2.1 characterizing  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> at 77°K.

possible phase diagram in the  $(h_X, h_Z)$  plane, constructed with the symmetry of the system taken into account, must have the form shown by curve  $T_1$  in Fig. 2. There are shown the fundamental characteristics of the phases in the different quadrants of the plane.

The static magnetic properties of the ideal easyaxis antiferromagnet considered by us, in contrast to zero DI (ideal in the sense that other interactions are disregarded), are completely described, as has already been pointed out, by the expressions (7e), (7f), (8c), and (8d). A numerical calculation of the curves  $m_x(h, \theta)$ and  $m_{\mathbf{Z}}(h, \theta)$ , where  $h \equiv (h_{\mathbf{X}}^2 + h_{\mathbf{Z}}^2)^{1/2}$  and where  $\theta$  is the angle between the principal axis and the external magnetic field, was made (see Fig. 3a and Fig. 3b) for an antiferromagnet characterized by the values of the parameters  $H_A$ ,  $H_D$ , and  $H_E$  (see above) determined by us for  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> at T = 77°K from resonance measurements [10]. These curves permit the easy construction both of magnetization curves and of torque curves. The question of their applicability of a real easy-axis modification of the antiferromagnet  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> is discussed below.

7. It is of great interest to study the dynamics of the antiferromagnetic system described, and first of all the frequency spectrum of AFMR,  $\omega_{10}(h)$  and  $\omega_{20}(h)$ . For arbitrary a,  $\beta$ , and B this can be done in explicit form only for fields oriented along the principal directions of the crystal: parallel to the principal axis and perpendicular to it (for a system of CoF<sub>2</sub> type, the results



FIG. 3. Calculated dependence of the (a) transverse and (b) longitudinal components, with respect to the EA, of the total magnetization upon the magnitude of the field, for different orientations of the latter with respect to the EA ( $\theta$  is the angle between H and the EA). The calculation was made for the values  $2H_A = 0.54$  kOe,  $H_D = 30$  kOe, and  $\frac{4}{2}H_E = 4500$  kOe, which follow from resonance measurements for  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> (T = 77°K).

of the calculation were given in <sup>[12]</sup>). When  $a \ll \beta \ll B$ , the calculations in a number of cases are simpler. Thus by solving the system (4) for small deviations from the equilibrium state, which is determined for the different phases by the expressions (7a)-(7f) and (8a)-(8d), one can obtain the functions  $\omega_{1,2}(h, \theta)$ . Construction of them for arbitrary  $\theta$  in the phase with  $l_{\mathbb{Z}} \neq 0$  is, as usual, complicated (therefore the corresponding curves in Fig. 4 are shown approximately by dotted lines). In the remaining cases, however, explicit expressions are obtained.

Phase with  $l_z \neq 0$  (that is,  $h < h_c$ ):

$$\theta = 0; \quad \Omega_{1,2}^2 = (1 \mp \rho)^2, \quad (19)$$

$$\theta = \pi / 2; \quad \Omega_1^2 = 1 - \tau^2 \rho^2, \quad \Omega_2^2 = 1 + \rho^2.$$
 (20)

Phase with  $l_z = 0$  (that is,  $h > h_c$ ):

$$\Omega_{1,2} = \frac{1}{2} \left[ \rho^2 + 2\tau \rho \sin \theta - 1 \mp \sqrt{(\rho^2 - 1)^2 + 4\rho^2 \sin^2 \theta} \right].$$
(21)

Here, for convenience, the relative quantities have been introduced:

$$\Omega_n \equiv \omega_{n0} / \omega_0 \quad (n = 1, 2), \ \omega_0 / 2M_0 \gamma \equiv h_0 \equiv h_{\text{cll}} = \overline{\gamma a B} - \beta^2,$$
  
$$\rho \equiv h / h_0, \quad \tau \equiv \beta / h_0.$$

The curves of Fig. 4 are drawn for the value  $\tau = 0.476$ , which follows from<sup>[10]</sup> for  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> at T = 77°K.

### 3. EXPERIMENT

1. As we have already reported, the resonance absorption near  $H_{C\perp}$  at frequencies  $\omega \ll H_{C\parallel}$ , predicted by formulas (20) and (21) with  $\theta = \pi/2^{23}$ , was actually observed in  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> at T  $\ll$  T<sub>M</sub>. The measurements were made with the aid of a simple reflection radio-spectrometer with a pulsed magnetic field. The latter was produced by discharge of a battery of condensers



FIG. 4. AFMR spectrum for the simplest easy-axis antiferromagnet with nonvanishing DI. The calculation was made for the values of  $H_A$ ,  $H_D$ , and  $H_E$  that occur in  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> at 77°K. The heavy lines are plotted according to formulas (20) and (21). The dotted lines give an approximate representation of the dependence  $\omega(H, \theta)$  for arbitrary  $\theta$  in the phase with  $l_Z \neq 0$ ; the point-and-dash lines represent the non-physical continuation of the dependences (21) into the phase with  $l_Z \neq 0$ .

<sup>&</sup>lt;sup>2)</sup> A similar prediction for the case of a perpendicular field was made independently by Cinader<sup>[13]</sup>.

of capacity C = 5700  $\mu$ F through a solenoid, wound with rectangular copper wire of cross section 1.68 × 2.83 mm<sup>2</sup>. The internal diameter of the solenoid was 19 mm, the external diameter 70 mm, and the height 70 mm. The solenoid was submerged in liquid nitrogen. The largest field that we could use without risk of destroying the solenoid was reached at condenser voltage U<sub>max</sub> = 3.5 kV and was H<sub>max</sub> = 250 kOe. The duration of the pulsed field from zero to zero was  $\tau_{00} \approx 10$  msec.

The field was measured by means of a Rogowski belt, the signal from which was integrated with a semiconductor integrator and fed to one of the inputs of a two-beam oscillograph OK-24 (see Fig. 5a). The field signal was calibrated with the aid of AFMR in a monocrystal of  $Cr_2O_3$  with  $H \parallel C_3$  and  $T = 77^\circ K$ ; it was assumed, in accordance with Foner<sup>[2]</sup>, that  $H_{res}$ = 46.5 kOe at  $\nu$  = 37.7 GHz (see Fig. 5b). The error in the field measurement was ±3%.

The specimen under investigation was clamped near the bottom of a shorted section of an 8-millimeter-band waveguide, on its narrow side or bottom, depending on which polarization of the microwave field was needed for observation of resonance. The part of the waveguide with the specimen was placed in the center of the solenoid, where the extent of 1% field uniformity was about 7 mm. The microwave power reflected from the measurement cell was fed through a directional coupler to a crystal detector, the signal from which was recorded by the second channel of the oscillograph (with sensitivity 1 mm/mV). This same measurement cell was used for measurement in wavelength band 4 and 6 mm.

2. The specimen of monocrystalline  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> on which the fundamental measurements were made<sup>3)</sup> was a parallelopiped of dimensions  $1 \times 3 \times 4$  mm, so cut that the C<sub>3</sub> axis was directed along the edge of length 2 [sic!]mm.

Control experiments (at wavelengths 8 and 4 mm, with  $H \perp C_3$ ) were performed on specimens cut from a monocrystal grown by R. A. Voskanyan (Institute of Crystallography, Academy of Sciences, USSR) and gave (to within the limits of accuracy) the same results.

3. To study the angular dependence of the resonance field, a very simple device was used which made it possible to change the orientation of the specimen within the waveguide. The specimen was glued with Bf glue on a surface of  $3 \times 4$  mm to the end of a rod of antelope bone, which passed through the wide wall of the waveguide. The angle between the field and the  $C_3$  axis was set manually with an accuracy of  $\pm 2^{\circ}$ ; the absence of disturbance to the angular setting was checked visually on the dial after each pulse of the field. Because of the appreciable size of the specimen, it was assumed that in one or another part of the specimen, for any angle of rotation, the microwave magnetic field necessary for excitation of the AFMR polarization was present. To prevent condensation of water and carbon dioxide vapors on the surface of the specimen, the interior of



FIG. 5. Antiferromagnetic resonance in synthetic  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> at liquidnitrogen temperature, observed by means of a reflection radiospectrometer with a pulsed magnetic field. Oscillogram a, dependence of the magnitude of the magnetic field on time. Duration of the pulsed field, from zero to zero,  $\tau_{00} = 9.5$  msec. Oscillograms b, c, and d, microwave detector signals, proportional to the power reflected from the measurement cell and recorded as functions of the time: b, frequency  $\nu = 37.7$ GHz, external field H parallel to the basal plane of the  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> crystal, hmicrowave || H. The narrow peak at the end is ordinary resonance in Cr<sub>2</sub>O<sub>3</sub> with H || C<sub>3</sub> and h<sub>microwave</sub>  $\perp$ H (T = 77°K), used for calibration of the field signal (H<sub>res</sub> = 46.5 kOe). c,  $\nu = 73$  GHz, H  $\perp$  C<sub>3</sub>, h<sub>microwave</sub> || H. d,  $\nu = 37.7$  GHz, H almost parallel to the easy axis of the  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> specimen (to within an accuracy of 2°). Besides the ordinary resonance observed earlier by Foner and Williamson[<sup>14</sup>] (narrow peaks), a broad maximum is visible at large fields.

the measuring cell was separated from the rest of the waveguide circuit by a thin layer of mica and was filled with gaseous helium with a slight excess of pressure.

4. The basic results obtained on the specimen from the monocrystal of V. M. Skorikov are shown in Figs. 5–7. Figure 6 shows satisfactory agreement of the experimental data with the calculated curve  $\omega_{10}(H_X)$ plotted according to formulas (20) and (21) for  $\theta = \pi/2$ with  $H_{C\parallel} = 63 \pm 3$  kOe and  $H_{C\perp} = 128 \pm 4$  kOe (the slight difference from the value of  $H_{C\perp}$  given in<sup>[10]</sup> is within the limits of error of the measurement) and with g-factor equal to 2.

A curious dependence of the resonance fields of hematite in the easy-axis phase is disclosed in Fig. 7.



FIG. 6. Dependence of AFMR frequency on magnitude of the field, perpendicular to the easy axis of  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub>. The curves n = 1 and 2 are plotted according to formulas (20) and (21) with  $\theta = \pi/2$  and with the values H<sub>c|l</sub> = 63 ± 3 kOe, H<sub>c</sub> = 128 ± 4 kOe ( $\lambda_p$  is the operating wavelength of the microwave spectrometer.

<sup>&</sup>lt;sup>3)</sup>The single crystal was grown by V. M. Skorikov (Institute of General and Inorganic Chemistry, Academy of Sciences, USSR) and was kindly given to us by E. G. Rudashevskiĭ.



FIG. 7. Change of the magnitude of the resonance fields in hematite with the angle between the EA and the field H; T = 77°K,  $\nu$  = 37.7 GHz, Z || C<sub>3</sub>. The side of the experimental points corresponds to the errors of measurement. Curve 1 is plotted from the experimental points. Curve 2 is calculated by formula (22) with  $2H_A = 0.54$  kOe, H<sub>D</sub> = 30 kOe  $\frac{1}{2}H_E = 4500$  kOe. The dotted curve is calculated by formula (18a) and corresponds to the boundary between the phases with  $l_Z \neq 0$  and with  $l_Z = 0$  in the (H<sub>x</sub>, H<sub>z</sub>) plane; the dash-dot curve is a nonphysical continuation of the dotted curve.

Displacement of the resonance (curve 2 in Fig. 7) toward the high-field region can be expected from a qualitative analysis of the AFMR spectrum (see Fig. 4). In fact, in observation of resonance at a frequency  $\omega \ll \omega_0$  by scanning of a field oriented at a small (for example,  $\theta = 5.7^{\circ}$ ) angle to the EA, besides the usual AFMR at  $h < h_C(\theta)$  (Foner and Williamson<sup>[14]</sup>), there should occur a broad absorption peak at fields larger than  $h_C(\theta)$ . This circumstance is illustrated by the oscillogram of Fig. 5d.

The dependence of the resonance (curve 2 of Fig. 7) on orientation can be described analytically. Thus from formula (21) for  $\omega_{10}$  (minus sign on the right), we easily find for the field H<sub>2</sub> of the second resonance peak:

$$H_{2z}^{2} = \left(\frac{H_{A}H_{E}}{H_{D}}\right)^{2} + \left(\frac{\omega_{10}}{\gamma}\right)^{2} - \left(H_{2x} - \frac{H_{A}H_{E} - H_{D}^{2}}{H_{D}}\right)^{2} + \left(\frac{\omega_{10}}{\gamma}\right)^{2} \frac{H_{c\perp}H_{2x}}{H_{D}H_{2x} - (\omega_{10}/\gamma)^{2}}.$$
(22)

This expression shows that the curve  $H_{2Z}(H_{2X})$  with  $\omega_{10}$  = const has a singularity at  $H_{2X} = (\omega_{10}/\gamma)^2/H_D$ . As is seen from Fig. 7, the experimental points agree well with curve 2, plotted from formula (22) with  $2H_{A}$ = = 0.54 kOe,  $H_D$  = 30 kOe, and  $\frac{4}{2}H_E$  = 4500 kOe.

#### 4. DISCUSSION OF RESULTS

As follows from what has been stated, the simplest classical model of an easy-axis antiferromagnet with DI describes rather well the dynamics of the low-temperature phase of hematite for  $T \ll T_M$ . Furthermore, it predicts the existence at each given temperature of two phases, with  $l_{\rm Z} \neq 0$  and with  $l_{\rm Z}$  = 0, separated in the ( $H_{\rm X}, H_{\rm Z}$ ) plane by the circle

$$H_{cc}^{2} + \left(H_{cx} - \frac{H_{A}H_{E} - H_{D}}{H_{D}}\right)^{2} = \left(\frac{H_{A}H_{E}}{H_{D}}\right)^{2}.$$
 (18a)

The existence of such a boundary is actually shown already by Fig. 7: the dotted curve is drawn according to formula (18a). It is of considerable interest, however, to construct the boundary line from static data; that is, for  $\omega = 0$ . Similar measurements for temperatures not too far from the Morin point T<sub>M</sub> have already been undertaken<sup>[8,9]</sup>. Their results can be explained, without forcing, within the framework of the model developed.

Actually, the coefficients of (18a) naturally depend on the temperature T. The quantity that changes most rapidly with T, as follows from [15, 16], is the anisotropy field  $H_A(T)$ . The field  $H_D$  varies insignificantly with temperature: it follows from our measurements that  $H_D(77 \circ K) = 30 \pm 4$  kOe, and from the resonance measurements of Rudashevskii and Shal'nikova<sup>[17]</sup> that  $H_D(295^{\circ}K) = 22 \pm 0.2$  kOe. And apparently  $H_E(T)$ changes very little (because of the high value of TN  $\approx$  948°K). In one way or another, the boundary curve  $H_{cz}(H_{cx})$ , while remaining a circle, should become strongly modified with temperature (see curves  $T_1$ ,  $T_2$ , and  $T_3$  in Fig. 2). In particular, when T is very close to  $T_M$ , the boundary curve in the first quadrant of the  $(H_x, H_z)$  plane suggests very much in its character the experimental curve obtained by Kaczér and Shalnikova for  $T_{M}$  –  $T \approx 2^{\circ}$  (see Fig. 4 in<sup>[8]</sup>). So that if we consider only the form of the dependence  $H_{cz}(H_{cx})$ , it is entirely possible to interpret these results without introducing interactions of higher order.

One difficulty, none the less, remains. The curves  $m_x(H_x)$ , as is confirmed in <sup>[9]</sup>, are not continuous at  $H_x = H_{Cl}$ , as they should be if there were full equivalence of the simplest model to the case of  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> at  $T < T_M$  (see formulas (9) and (10)). It is therefore possible that inclusion of higher-order interactions is still necessary for a description of  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub>; this should be carried out, however, within the framework of the calculation of the problem that we have presented above.

In addition, it would be of unquestionable interest to measure carefully the orientational dependence of the magnetization curves of  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> in fields encompassing the critical field, at nitrogen temperature. Another very interesting problem is the investigation of the thermodynamics of the transition upon crossing the boundary curve along different trajectories in the (H<sub>X</sub>, H<sub>Z</sub>) curve.

#### 5. CONCLUSION

The good agreement of the results of a calculation of the dynamics of an easy-axis antiferromagnetic system with nonvanishing DI and of the experimental data on AFMR for low-temperature  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> leads to the conclusion that consistent allowance for DI is necessary for description of any easy-axis antiferromagnetic systems-especially of their behavior in large fields. The expression (1) and the relations (7e), (7f), (8c), and (8d) apparently have a very general character, in particular for systems with arbitrary  $H_A$ ,  $H_D$ , and  $H_E$ . Inclusion of DI in the calculation allows us to consider the processes of "flipping" and "collapse" (now relative) of the sublattice moments from a unified point of view-as field-induced transitions from the phase with  $l_{\mathbf{Z}} \neq 0$  to the phase with  $l_{\mathbf{Z}}$ = 0, which should also take place for arbitrary orientation of the external field with respect to the principal axis.

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