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"PREMATURE" DISAPPEARANCE OF ANTIFERROMAGNETIC RESONANCES IN HEMATITE

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The temperature dependence of AFMR in hematite ($\alpha - Fe_2O_3$) is studied experimentally for the external magnetic field lines parallel or perpendicular to the axis of easy magnetization. The experimental conditions are $H = 0 - 200$ kOe, $T = 77–293°K$ and $\lambda_{unf} \approx 8$ mm or 4 mm. As expected, with increase of temperature the resonance field strengths decrease, but considerably earlier than the Morin point $T_M \approx 260°K$ the resonance intensity at a frequency $\nu_r = 37.7$ GHz unexpectedly drops to zero. This "premature" disappearance of resonances occurs at $T_D^\perp \approx 205°K$ in a perpendicular field and at $T_D^\parallel \approx 240°K$ in a parallel field. No such pronounced effect is observed at a frequency $\nu_r = 72$ GHz.

A phenomenological calculation of the dynamics of the hematite magnetic system is performed by taking into account the "second anisotropy" constant for the antiferromagnetic vector **l**. Introduction of this constant leads to duality of the $\omega_{res}(H)$ dependences near those values of H for which transition from a phase with $l_z \neq 0$ to a phase with $l_z = 0$ occurs and, as a consequence, to the formation of "gaps" in the dependences. The calculated temperature dependence of the gaps can explain the "premature" disappearance of resonances (for both magnetic field orientations) observed at $\nu_r = 37.7$ GHz and also the absence of a similar anomaly for $\nu_r = 72$ GHz. It follows from the calculation, moreover, that the strength of the parallel "spin-flop" field in hematite $H_\parallel^t$ is not equal to ω_0/γ (ω_0 is the AFMR frequency in zero field) as observed in ordinary antiferromagnets with a small second anisotropy constant. Satisfactory agreement with the experimental data is obtained for the following values of the effective field strengths: $2H_E = 8960$ kOe; $H_D = 22.7$ kOe; $H_{A_4}(77°K) = 0.382$ kOe; $H_{A_2}(77°K) = 0.222$ kOe. It is concluded that at 77°K the phase transition in hematite in a perpendicular field $H_\perp^t = 130$ kOe and at higher temperatures is a first-order transition; however, the discontinuity of the magnetic moment ($\Delta m^{\perp t}_{(77°K)} \sim \sigma_{(300°K)}/8$) and the gap in the AFMR spectrum ($\omega^{\perp t}_{(77°K)}/\gamma \approx 6$ kOe) are relatively small.

1. INTRODUCTION

INVESTIGATIONS of the static and dynamic properties of hematite ($\alpha - Fe_2O_3$, rhombohedral structure), performed during the last 3–4 years in a large number of laboratories[1-15] on artificial single crystals, have greatly advanced our understanding of the features of the behavior of this most popular among all antiferromagnets (AF). Particularly interesting details were observed in the investigation of the low temperature ($T < T_M = 262°K$[11]) phase of the hematitie, in which it is an AF with "easy axis" (EA) anisotropy. It has turned out that, besides the known "flipping of the sublattices" in the parallel field, a magnetic phase transition is possible also in a magnetic field of definite magnitude perpendicular to the EA. This transition is accompanied by a change in the character of the magnetization curve $m_\perp(H_\perp)$ (from a dependence in the form $m_\perp = \chi_\perp^{(1)} H_\perp$ at $H_\perp \ll H_\perp^t$ to a dependence in the form $m_\perp = \sigma^{(2)} + \chi_\perp^{(2)} H_\perp$ at $H_\perp^t$) and the appearance of resonance absorption at relatively low frequencies[9,13]. It was observed that with increasing temperature, the sharpness of this transition, as determined from the $m_\perp(H_\perp)$ curve, increases with increasing temperature from that of liquid nitrogen to the Morin point T_M: at $T = 77°K$, a jump of the derivative $dm_\perp/dH_\perp$ is observed in the transition field $H_\perp^t$, whereas near T_M, a quasidiscontinuous increase of the magnetic moment $m_\perp$ itself takes place in a field $H_1^t(T)$.

In the first approximation, the picture of the phase transition in a perpendicular magnetic field, especially at low temperatures, can be well described by the phenomenological theory that takes into account only three interactions-- exchange with parameter B, anisotropy parameter a, and Dzyaloshinskiĭ interaction with parameter β[9,16,17]: with increasing field, rotation of the antiferromagnetic axis (AF axis) occurs from the EA, which is parallel to the C_3 axis of the crystal, to the basal plane, and in the critical field $H_\perp^t$ the AF axis falls in the basal plane. An experimental study of the resonance predicted by this theory, however, leads to a value $\beta(77°K) = 30$ kOe[9], whereas from the measurements of $m_\perp(H_\perp)$ at practically the same temperature it follows that $\beta(110°K) = 22.7$ kOe[12], which coincides with the value $\beta(293°K) = 22$ kOe obtained from resonance[2] and static[1] experiments in the high temperature (easy-plane) phase of the hematite ($T > T_M$). In addition, the simplest theory does not predict any change in the qualitative characteristics of the aforementioned transition in a perpendicular field with increasing temperature.

Cinader, Flanders, and Shtrikman[11], in a study of the phase transition induced by a perpendicular field in the immediate vicinity of the Morin point ($T \lesssim T_M$), have shown by numerical calculation that the break is observed in the $m_\perp(H_\perp)$ plot for a field $H_\perp = H_\perp^t$ when account is taken of fourth-order anisotropy for the AF vector. It also follows from the calculation that with decreasing temperature the magnitude of the jump decreases. Levitin and Shchurov[12] measured in detail the $m_\perp(H_\perp, T)$ curves for the region $H_\perp = 0$–150 kOe and $T = 120$–$291°$K, and attempted to describe analytically the change observed by them in the character of the transition with changing temperature, likewise by introducing a second anisotropy constant[18] 1).

In order to determine the connection between the temperature change of the character of the discussed transition and the resonance absorption near the critical point, and also to find the causes of the quantitative difference between the values of $\beta(77°K)$ obtained by simplest reduction of the resonance and static experiments, we have undertaken an investigation of the temperature dependence of the resonance absorption in the easy-axis hematite at the microwave wavelengths $\lambda = 8$ and 4 mm.

It was observed that at $\lambda = 8$ mm and $H \perp C_3$, an increase of temperature leads not only to the expected smooth decrease of the resonance field, but also to a sharp drop in the intensity of the resonance in a narrow temperature interval near $T_V^\perp \sim 200°$K, i.e., long before the Morin point $T_M = 262°$K is reached2).

As shown by us below, this phenomenon ("premature" vanishing of the resonance) is closely connected with the temperature change of the character of the transition in hematite. In particular, this phenomenon is sufficiently well described quantitatively if account is taken of the "second anisotropy constant" in the calculation of the dynamics of the system. At the same time, it is possible to bring in agreement many of the previously determined experimental characteristics of the hematite, and furthermore predict the "premature" vanishing of the resonance in the frequently investigated case when the external magnetic field is parallel to the EA. This effect was also observed experimentally.

2. PHENOMENOLOGICAL THEORY

1. We consider first in as general a form as possible the influence of the energy terms of order higher than the second on the static and dynamic properties of hematite in the easy-axis state (i.e., when $T < T_M$), placed in a magnetic field perpendicular to EA C_3 ($C_3 \parallel z$; $H \parallel x$). We shall adhere here to the calculation sequence used in[13], and write down the magnetic energy of the crystal, following[19] and being interested only in effects that are isotropic in the basal plane, in the form $\mathscr{E} \equiv 2M_0\mathscr{H}$:

$$\mathscr{H} = \frac{B}{2}\mathbf{m}^2 + \frac{a_1}{2}(l_x^2 + l_y^2) - \beta(m_x l_y - m_y l_x) + \frac{b}{2}m_z^2$$
$$+ \frac{C}{4}\mathbf{m}^4 + \frac{D}{2}\mathbf{m}^2 l_z^2 - \frac{a_2}{4}l_z^4 - f l_z^2(m_x l_y - m_y l_x)$$
$$+ \frac{G}{2}\mathbf{m}^2(m_x l_y - m_y l_x) + \frac{e}{2}(m_x l_y - m_y l_x)^2 - \mathbf{mH}. \qquad (1)$$

Here $2M_0$ is the saturation magnetization of the crystal; $\mathbf{m}$ and $\mathbf{l}$ are ferromagnetic and antiferromagnetic vectors (see[13]); B and C are the exchange-interaction constants; a_1, b, and β are the second-order relativistic-interaction constants; D and G are the constants of the mixed interaction (exchange-relativistic interaction); a_2, f, and e are the fourth-order relativistic-interaction constants. In accordance with the chosen form for writing down the energy (which differs somewhat from that used by us[13]), all these constants are the effective fields of the corresponding interactions and therefore, like the external magnetic field H, are measured in Oersteds (Gausses). The fourth-order terms with coefficients C, G, and e will henceforth be neglected, since they contain the small quantity m raised to a sufficiently high power.

As before[9,13] we are interested in the temperature region $T < T_M \ll T_N$, and we therefore assume the sublattices to be magnetized to saturation; assuming an isotropic g factor, this leads to

$$\mathbf{m}^2 + \mathbf{l}^2 = 1; \qquad (\mathbf{ml}) = 0. \qquad (2)$$

2. From the equations of motion for $\mathbf{m}(t)$ and $\mathbf{l}(t)$ (see[13]) it follows that when $\mathbf{H} = (H, 0, 0)$ the equilibrium values of the components m_y, m_z, and l_x vanish, and their small deviations oscillate at a frequency ω_{21} in the first phase ($l_z \neq 0$) and ω_{22} in the second phase ($l_z = 0$), see below.

For the other three components we can easily derive

$$\dot{m}_x = -l_z F, \qquad (3a)$$
$$\dot{l}_y = l_z(\Phi - H), \qquad (3b)$$
$$\dot{l}_z = m_x F - l_y(\Phi - H), \qquad (3c)$$

where

$$F(m_x, l_y) \equiv \partial\mathscr{H}'/\partial l_y \equiv \mathscr{H}_l'; \qquad \Phi(m_x, l_y) - H \equiv \partial\mathscr{H}'/\partial m_x \equiv \mathscr{H}_m';$$

$$\mathscr{H}'(m_x, l_y) = -\frac{a_2}{4} + \frac{B + D + a_2}{2}m_x^2 + \frac{a_1 + a_2}{2}l_y^2 - (\beta$$
$$+ f)m_x l_y - \frac{D + a_2}{2}m_x^2 l_y^2 - \frac{2D + a_2}{4}m_x^4 - \frac{a_2}{4}l_y^4$$
$$+ f m_x l_y(m_x^2 + l_y^2) - m_x H. \qquad (4)$$

(This is obtained from (1) by making the substitutions $m_y = m_z = l_x = 0$ and $l_z^2 = 1 - m_x^2 - l_y^2$) Equation (3c) owing to condition (2), follows from (3a) and (3b).

Examination of small deviations ($m_x \to m_x + \mu_x$; $l_y \to l_y + \lambda_y$; $l_z \to l_z + \lambda_z$; $\mu_x, \lambda_y, \lambda_z \sim e^{i\omega t}$) from the equilibrium positions in the first and second phases leads to the following system of homogeneous equations3) for μ_x, λ_y, and λ_z with the following coefficients:

μ_x	λ_y	λ_z
$i\omega - l_z\mathscr{H}'_{lm}$	$-l_z\mathscr{H}'_{ll}$	$-\mathscr{H}_l'$
$l_z\mathscr{H}'_{mm}$	$i\omega + l_z\mathscr{H}'_{ml}$	$\mathscr{H}_m'$
$\mathscr{H}'_l + m_x\mathscr{H}'_{lm} - l_y\mathscr{H}'_{mm}$	$-(\mathscr{H}'_m + l_y\mathscr{H}'_{ml} - m_x\mathscr{H}'_{ll})$	$i\omega$

(5)

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[1] It should be noted that the need for taking into account this constant in the description of the behavior of low-temperature hematite (in a parallel field) was also pointed out by Bessez, Mozzish, and Searle [10].

[2] This phenomenon was observed independently and simultaneously by Rudashevskii and Miznov, using a spectrometer with a constant magnetic field ($H_{max} = 110$ kOe), see [15].

[3] Here and throughout (with the exception of fhe figures) we write for brevity ω in lieu of ω/γ, where γ is the magnetomechanical ratio. In the numerical calculations for hematite we use a value of 2 for the g-factor [6].

the expression for $\mathscr{H}'_{ll}$. In view of this, with practically no sacrifice in accuracy, we have henceforth put throughout, for simplicity, C = D = f = 0, and undertook in this approximation a detailed calculation of the dynamics of the system under consideration.

In addition, since in hematite, owing to the relation $a_1 \ll \beta \ll B$, the phenomena of interest to us take place at values $m \lesssim a_1/\beta \ll 1$, all the subsequent calculations were made under the assumption that $l_z^2 = 1 - m_x^2 - l_y^2 \approx 1 - l_y^2$. It was frequently convenient (especially for the case $H \perp C_3$) to express all quantities as functions of $l_1 = l_y^{\mathrm{I}}(H)$, rather than H ($l_y^{\mathrm{I}}$ is the y component of the AF vector in the first phase).

4. Hematite at $T < T_M$, $H \perp C_3$. Only the terms with B, β, a_1, a_2, and b, and the condition (10), were taken into account; we introduced the parameter $\kappa(T) \equiv H_0^2/a_2B$, where $H_0^2 \equiv a_1B - \beta^2$.

Phase with $l_z \neq 0$:

$$m_1 = (\beta l_1 + H)/B, \quad \beta H = a_2Bl_1(1 + \varkappa - l_1^2), \tag{12a}$$

$$(\omega_{11}{}^{\perp})^2 = a_2B(1 - l_1^2)(1 + \varkappa - 3l_1^2), \tag{12b}$$

$$(\omega_{21}{}^{\perp})^2 = a_2B(1 + \varkappa - l_1^2) + H^2 = H(H + \beta / l_1). \tag{12c}$$

Phase with $l_z = 0$

$$m_x = (\beta + H)/B, \quad l_y \approx 1, \tag{13a}$$

$$(\omega_{12}{}^{\perp})^2 = \beta H - H_0^2, \tag{13b}$$

$$(\omega_{22}{}^{\perp})^2 = H(H + \beta). \tag{13c}$$

We express the energy difference of these two phases as functions of l_1:

$$\mathscr{H}_1 - \mathscr{H}_2 = {}^1/_4 a_2(1 - l_1)^2[3l_1^2 + 2l_1 - (2\varkappa + 1)].$$

The unexpected simplicity of this expression makes it possible to obtain the subsequent results in explicit form.

A transition from the first phase ($l_z \neq 0$) to the second ($l_z = 0$) will occur when $\mathscr{H}_1 - \mathscr{H}_2 = 0$, i.e., either when $l_1 = 1$ if $\kappa > 2$, or else when

$$l_1 = l_y{}^{\mathrm{t}} \equiv (\sqrt{4 + 6\varkappa} - 1)/3, \tag{14}$$

if $\kappa \leq 2$, for at this value of κ the quantity $l_y^{\mathrm{t}} < 1$ (we recall that $l_1 \equiv l_y(H)$ cannot be larger than unity).

The relations described by formulas (12) and (13) are shown in Fig. 2 (the construction was made for the values of B, β, a_1 and a_2 determined below from a comparison with experiment at T = 77°K, where $\kappa = 1.46 < 2$, and at T = 215°K, where $\kappa = 0.5$). The most characteristic parameters of the phase transition (in this case—of first order!) are determined by the following formulas, which are valid when $\kappa \leq 2$:

$$H_{\perp}{}^{\mathrm{t}} = a_2B[9\varkappa + 4 + (2 + 3\varkappa)\sqrt{4 + 6\varkappa}]/27\beta, \tag{15a}$$

$$(\omega_{11}{}^{\perp \mathrm{t}})^2 = 2a_2B[(2 + 3\varkappa)^2 - (9\varkappa - 2)\sqrt{4 + 6\varkappa}]/27, \tag{15b}$$

$$(\omega_{12}{}^{\perp \mathrm{t}})^2 = a_2B[(2 + 3\varkappa)\sqrt{4 + 6\varkappa} - (18\varkappa - 4)]/27, \tag{15c}$$

$$\Delta m \equiv m_2{}^{\mathrm{t}} - m_1{}^{\mathrm{t}} = \beta(4 - \sqrt{4 + 6\varkappa})/3B, \tag{15d}$$

$$H_{\perp}{}^{(1)} = 2a_2B(\varkappa + 1)^{3/2}/3^{3/2}\beta, \quad H_{\perp}{}^{(2)} = H_0{}^2/\beta, \tag{15e}$$

$$\omega_{11}|_{H_{\perp} \to 0} = \omega_0 \equiv [(a_1 + a_2)B - \beta^2]^{1/2} \equiv [a_2B(\varkappa + 1)]^{1/2}, \tag{16}$$

(see also Fig. 1).

If the relative magnitude of the different interactions were such that $\kappa > 2$, then the transition from the first phase to the second would occur without jumps in the quantities m_x and l_y (i.e., by means of a second-order phase transition), with

$$l_y{}^{\mathrm{t}} \approx 1;$$
$$H_{\perp}{}^{\mathrm{t}} = H_{\perp}{}^{(1)} = H_{\perp}{}^{(2)} = H_0{}^2/\beta;$$
$$\omega_{11}{}^{\perp \mathrm{t}} = \omega_{12}{}^{\perp \mathrm{t}} = 0 \tag{17}$$

In this case there can exist a temperature T* such that $\kappa(T^*) = 2$ (at this temperature a change takes place in the order of the phase transition); then, for temperatures $T > T^*$ but close to T^* (such that $2 - \kappa(T) \ll 1$), the relations (15b, c, d) can be readily expanded in series:

$$\omega_{11}{}^{\perp \mathrm{t}} \approx 2\omega_{12}{}^{\perp \mathrm{n}} \approx \approx (a_2B)^{1/2}(2 - \varkappa)/2, \tag{18a}$$

$$\Delta m \approx \beta(2 - \varkappa)/4B. \tag{18b}$$

Thus, when the temperature rises from 0°K, a change in the order of the phase transition induced by a perpendicular magnetic field can take place in the AF type hematite. As soon as such a change takes place (the transition becomes of the first order), a gap $\omega_{12}^{\perp \mathrm{t}}$ appears in the dependence of the AFMR frequency on the field and increases with temperature, first slowly (see formula (18a)) and then more and more rapidly (formula (15c)).

The ambiguity of the dependence of the AFMR frequency on the field in the segment from $H_{\perp}^{(2)}$ to $H_{\perp}^{(1)}$ (at $\kappa < 2$) is evidence of the presence of metastable states separated by an energy barrier, as is typical

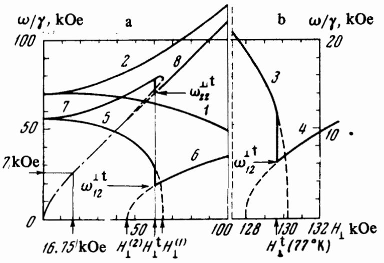

FIG. 2. AFMR spectrum in a perpendicular field for an easy-axis AF with Dzyaloshinskii interaction and with allowance for the second anisotropy constant. The plotting was in accordance with formulas (12) and (13) for hematite. The parameter values B = 8960 kOe and β = 22.7 kOe were obtained by reducing the results of static measurements of Voskanyan, Levitin, and Shchurov [12], while the parameters $a_1(T)$ and $a_2(T)$ were obtained by reducing the experimental resonance data (see Figs. 4 and 6 below). The first and second subscripts of ω denote the number of the branch and the number of the phase, respectively. Curve 1 – $\omega_{11}{}^{\perp}$, 2 – $\omega_{21}{}^{\perp}$, 3 – $\omega_{11}{}^{\perp}$, 4 – $\omega_{12}{}^{\perp}$, 5 – $\omega_{11}{}^{\perp}$, 6 – $\omega_{12}{}^{\perp}$, 7 – $\omega_{21}{}^{\perp}$, 8 – $\omega_{22}{}^{\perp}$, For curves 1 and 4, a_1 = 0.382 kOe, a_2 = 0.222 kOe, and T = 77°K; for curves 5 – 8, a_1 = 0.173 kOe, a_2 = 0.234 kOe, T = 215°K.

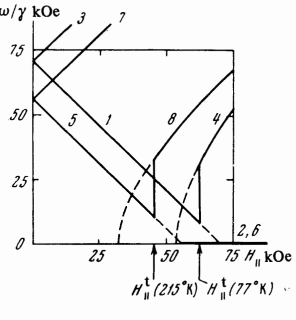

FIG. 3. AFMR spectrum in a parallel field, plotted under the same conditions as in Fig. 2. Curve 1 – $\omega_{11}{}^{\parallel}$, 2 – $\omega_{12}{}^{\parallel}$, 3 – $\omega_{21}{}^{\parallel}$, 4 – $\omega_{22}{}^{\parallel}$, 5 – $\omega_{11}{}^{\parallel}$, 6 – $\omega_{12}{}^{\parallel}$, 7 – $\omega_{21}{}^{\parallel}$, 8 – $\omega_{22}{}^{\parallel}$. For curves 1 – 4, a_1 = 0.382 kOe, a_2 = 0.222 kOe, T = 77°K; for 5 – 8, a_1 = 0.173 kOe, a_2 = 0.234 kOe, T = 215°K.

where

$$\mathcal{H}'_{mm} \equiv \frac{\partial^2 \mathcal{H}'}{\partial m_x^2} = B + D + a_2 - (D + a_2) l_y^2 - 3(2D + a_2) m_x^2 + 6 f m_x l_y,$$

$$\mathcal{H}'_{ll} \equiv \frac{\partial^2 \mathcal{H}'}{\partial l_y^2} = a_1 + a_2 - (D + a_2) m_x^2 + 6 f m_x l_y - 3 a_2 l_y^2, \qquad (6)$$

$$\mathcal{H}'_{ml} \equiv \frac{\partial^2 \mathcal{H}'}{\partial m_x \partial l_y} = -[(\beta + f) + 2(D + a_2) m_x l_y - 3f(m_x^2 + l_y^2)].$$

The equilibrium values $m_x \equiv m_1(H)$ and $l_y \equiv l_1(H)$ for the first phase ($l_z \neq 0$) are given, as follows from (3a) and (3b), by the equations

$$F(m_1, l_1) = 0, \qquad \Psi(m_1, l_1) = H. \qquad (7)$$

The energy in the equilibrium state is determined in this case by the expression

$$\mathcal{H}_1 = -\frac{a_2}{4} - \frac{B + D + a_2}{2} m_1^2 + \frac{a_1 + a_2}{2} l_1^2 + \frac{3}{4} (2D + a_2) m_1^4 - \frac{a_2}{4} l_1^4 + \frac{D + a_2}{2} m_1^2 l_1^2 - 2 f m_1^3 l_1.$$

For the lower AFMR frequency branch in the first phase ($l_z \neq 0$; $\mathcal{H}'_l = \mathcal{H}'_m = 0$) we obtain from (5)

$$\omega_{11}^2 = l_z^2 \Delta(m_1, l_1), \qquad (8a)$$

where

$$\Delta(m_1, l_1) \equiv \mathcal{H}_{mm}' \mathcal{H}_{ll}' - (\mathcal{H}_{ml}')^2. \qquad (8b)$$

The first index of ω denotes the branch number and the second the phase number.

Differentiating the system (7) with respect to H, we obtain expressions that also contain the function $\Delta(m_1, l_1)$:

$$\frac{dm_1}{dH} = \frac{\mathcal{H}_{ll}'(m_1, l_1)}{\Delta(m_1, l_1)}; \qquad \frac{dl_1}{dH} = -\frac{\mathcal{H}'_{ml}(m_1, l_1)}{\Delta(m_1, l_1)}.$$

The second phase ($l_z = 0$; $l_y \Phi - m_x F = l_y H$) is characterized by the relations

$$(B - a_1) m_x - \beta \frac{1 - 2m_x^2}{\sqrt{1 - m_x^2}} = H, \qquad l_y = \sqrt{1 - m_x^2};$$

$$\mathcal{H}_2 = \frac{a_1}{2} - \frac{B - a_1}{2} m_x^2 - \frac{\beta m_x^3}{\sqrt{1 - m_x^2}},$$

$$\omega_{12}^2 = (\beta m_x - a_1 l_y) \left[(B - a_1) l_y + \beta m_x \frac{3 - 2m_x^2}{\sqrt{1 - m_x^2}} \right].$$

As expected, by starting from the form of $\mathcal{H}$ (see (1)), allowance for the fourth-order terms does not affect the characteristics of the second phase when $H \ll B$ (with just this purpose in view, we chose to write down the fourth-order terms in a form different from that used by Levitin and Shchurov in[18]).

3. We now analyze the change of the quantities m_1, l_1, and ω_{11}^2 with increasing external magnetic field and the relative contribution made to this change by different fourth-order terms (with the coefficients C, D, a_2, and f). We shall take into account here the fact that we know the exact expressions when $C = D = a_2 = f = 0$ (these are given in[9,13]).

By regarding l_1 formally as an independent variable, we know that with increasing l_1 the quantity ω_{11}^2 can vanish as a result of the vanishing of either l_z or Δ. If $l_z = 0$ initially with increasing l_1, then a continuous transition to the second phase takes place, since the latter is in fact characterized by the condition $l_z = 0$.

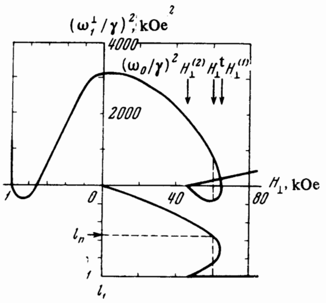

FIG. 1. Illustration of the dependence of the frequency of the first AFMR branch on the magnitude of the perpendicular field in an easy-axis AF with the Dzyaloshinskiĭ interaction and with account of the fourth-order interactions. The curves were plotted for a concrete case (hematite, T = 215°K), but they reflect the general type of the regularities discussed in Sec. 2, Item 3.

On the other hand, if at first $\Delta = 0$, then with further increase of l_1 the quantity ω_{11}^2 becomes smaller than zero, thus evidencing absolute instability of the solutions of the system (7), and a jumplike transition to the second phase must take place. A general idea of the dependence of ω_{11}^2 on l_1 and $H_\perp$ is given by the curves in Fig. 1, although they have been plotted for the concrete case of hematite (see below).

The quantity Δ is determined by expressions (8b) and (6). When H = 0 we have $m_x = l_y = 0$ and

$$\Delta|_{H=0} = \omega_{11}^2|_{H=0} = (a_1 + a_2)(B + D + a_2) - (\beta + f)^2. \qquad (9)$$

This quantity is larger than zero, since we are considering a stable easy-axis temperature phase of the hematite ($T < T_M$), but decreases with increasing temperature (mainly as a result of the decrease of a_1). If we assume, for example, that $D > 0$, $a_2 > 0$, and $f < 0$, then a situation is possible wherein Δ vanishes with increasing H, by virtue of the simultaneous increase of m_x and l_y (see formulas (6)!) at smaller values of m_x and l_y than those at which $l_z = (1 - m_x^2 - l_y^2)^{1/2}$ vanishes. This situation occurs all the sooner for hematite, the larger C, D, a_2, and f, or the smaller $\Delta|_{H=0}$, i.e., the higher T.

Let us estimate the relative contribution of different fourth-order terms to the value of Δ. We note here that we should assume apriori that $a_1 \sim \beta$ (since the nature of these second-order interactions is the same). For hematite, on the other hand, experiment yields $\beta \gg a_1$ (this circumstance apparently is worthy of a special analysis from the microscopic point of view). We therefore have

$$B \sim C \gg D \sim \beta \gg a_1 \sim a_2 \sim f. \qquad (10)$$

The relative contribution of the fourth-order terms can be estimated by taking for the first phase the values of m_x and l_y obtained by us[9,13] with allowance for only the second-order terms in the crystal energy:

$$m_x \approx a_1/\beta \text{ and } l_y \approx 1 \text{ for } H_\perp \approx H_\perp^t. \qquad (11)$$

Substituting (11) in (6) and (8b) and taking (10) into account, we can easily show that, from among all the fourth-order terms, the largest contribution to Δ is made by the term $a_2 l_z^4/4$, owing to the term $3a_2 l_y^2$ in for first-order phase transitions. If the time of transition of the system through this barrier is small, then the AFMR, by virtue of the presence of the gap $\omega_{12}^{\perp t}$, will be observed only at frequencies larger than a certain value. In addition, it can be easily shown that $d\omega_{12}^{\perp t}/d\kappa < 0$ when $2 \geq \kappa > -\frac{1}{2}$, and therefore, since $d\kappa/dT < 0$ in hematite (see below), $\omega_{12}^{\perp t}$ increases with temperature, and if the resonance is observed at constant frequency, it should vanish when a definite temperature is reached. This is precisely the effect which was observed experimentally (see[15] and Fig. 4).

The quantities $H_{\perp}^{(1)}$ and $H_{\perp}^{(2)}$ determine the field beyond which the corresponding phases are absolutely unstable. In the field interval between these two extreme values ($H_{\perp}^{(1)} - H_{\perp}^{(2)} \approx 2.2$ kOe at $T = 77°K$), hysteresis is possible in the $m_{\perp}(H_{\perp})$ dependence, and also the conservation of one of the resonances. However, in the concrete case of hematite, the system overcomes the energy barrier apparently quite rapidly, and therefore under ordinary conditions (for example, pulsed fields with a growth rate smaller than 50 kOe/msec), no such effects are observed.

5. Hematite at $T < T_M$, $H \parallel C_3$. All the phenomena are considered by us for $T \ll T_N$, so that we assume for simplicity that $\alpha \equiv \chi_{\parallel}/\chi_{\perp} \approx 0$ (this agrees quite well with the experimental data[12]).

Phase with

$$\text{c } l_z \neq 0:\ \mathbf{m} = 0;\ l_x = l_y = 0;\ l_z = 1,$$
$$\omega_{11,21}^{\parallel} = \omega_0 \mp H;$$
$$\omega_0 \equiv [(a_1 + a_2)B - \beta^2]^{1/2}$$
$$\equiv [a_2 B(\varkappa + 1)]^{1/2}. \tag{19}$$

Phase with

$$\text{c } l_z = 0:\ m_y = l_x = 0;\ m_x$$
$$\approx \beta/B;\ m_z \approx H/(B + b);\ l_y \approx 1,$$
$$\omega_{12}{}^{\parallel} = 0; \quad \omega_{22}{}^{\parallel} = (H^2 - H_0^2)^{1/2}. \tag{20}$$

The two phase energies ($\mathscr{H}_1^{\parallel}$ and $\mathscr{H}_2^{\parallel}$) become equal when

$$H_{\parallel}{}^{t} = [(a_1 + {}^1\!/_2 a_2)B - \beta^2]^{1/2}$$
$$\equiv [a_2 B(\varkappa + {}^1\!/_2)]^{1/2}. \tag{21}$$

Plots of (19) and (20) are illustrated in Fig. 3. Since

$$\omega_{11}{}^{\parallel t} = [a_2 B(\varkappa + 1)]^{1/2} - [a_2 B(\varkappa + {}^1\!/_2)]^{1/2}, \tag{22}$$

it can be easily shown that $d\omega_{11}^{\parallel t}/d\kappa < 0$, i.e., the gap in the $\omega(H_{\parallel})$ dependence exists at any temperature $T < T_M$ and increases with increasing temperature (if it is assumed that B, β, and a_2 do not depend on the temperature in first approximation, and $d\kappa/dT < 0$, as is the case for hematite).

6. The Morin point T_M, i.e., the temperature of the spontaneous reorientation of the AF vector, is obviously determined by the requirement $H_{\perp}^{t} = H_{\parallel}^{t} = 0$. Expressions (15a) and (21) for $H_{\perp}^{t}$ and $H_{\parallel}^{t}$ then lead to the following equation for T_M: $(a_1 + a_2/2)\,B - \beta^2 = 0$, i.e., $\kappa(T_M) = -\frac{1}{2}$, where it must be assumed that B, β, a_1, and a_2 are known functions of the temperature. At the Morin point, the frequency gaps reach the following values (which obviously hold when $H = 0$):

$$\omega_{11}{}^{\perp t} = \omega_{12}{}^{\perp t} = \omega_{21}{}^{\perp t} = \omega_{21}{}^{\parallel t} = \omega_{22}{}^{\parallel t} = (a_2 B/2)^{1/2} \equiv \omega_M;$$
$$\omega_{12}{}^{\parallel t} = \omega_{22}{}^{\perp t} = 0.$$

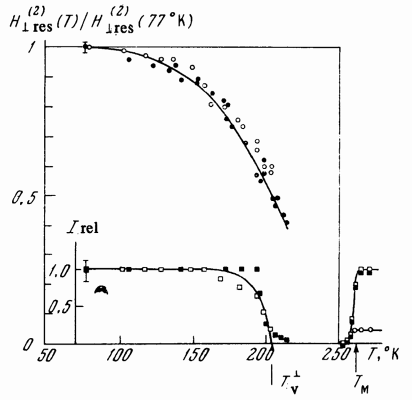

FIG. 4. Temperature dependence of AFMR in hematite in a field perpendicular to the C_3 axis ($H^{(2)}_{\perp res}$ – field of second absorption peak, I_{rel} – relative intensity of this peak), $\nu_r = 37.7$ GHz: ● – $H^{(2)}_{\perp res}$, sample No. 1; ○ – $H^{(2)}_{\perp res}$, sample No. 2; ■ – I_{rel}, sample No. 1; □ – I_{rel} sample No. 2; ◒, ◪ – corresponding data of Rudashevskii [14] for resonance in the easy-plane phase

3. EXPERIMENT

1. Apparatus. We studied the temperature dependence of the AFMR in hematite in the temperature range from 77 to 300° K with the aid of a reflex radio spectrometer with a pulsed magnetic field, described in[9,13]. The required sample temperature was maintained constant by directly regulating the current in a heater wound around the waveguide, and was measured with a chromel-copel thermocouple. The working junction of the thermocouple was glued to the outer wall of the waveguide as close as possible to the sample. The absence of an appreciable temperature difference between the working junction and the sample was verified with the aid of an analogous thermocouple, the junction of which was glued directly to the sample. The accuracy with which the temperature was maintained and measured was ± 1° K.

2. Samples. We used for the measurements samples meauring about 1 × 3 × 3 mm. These were cut from single crystals grown in different laboratories; sample No. 1—from a single crystal grown by V. M. Skovikov (Institute of General and Inorganic Chemistry); sample No. 2—single crystal grown by R. A. Voskonyan (Crystallography Institute), i.e., sample No. 2 was of the same origin as that used in[12] for static measurements. Each of the samples had a clearly pronounced natural face parallel to the basal plane of the crystal. Therefore the accuracy with which they were glued to the waveguide, which determines the accuracy of orientation of the external magnetic field relative to the principal directions of the crystal, is estimated to be ± 1°.

In the investigation of AFMR in a field perpendicular to C_3 axis, the azimuthal orientation of the field in the basal plane was not determined by us. As shown by investigations of E. G. Rudashevskiĭ and S. V. Mizonov (private communication), the temperatures of the "premature" vanishing of the resonance (see[15]) may differ slightly (up to 10°) for different field directions in the basal plane (for example, along the U_2 axis and at a right angle to it). It was not our purpose, however, to investigate these fine details.

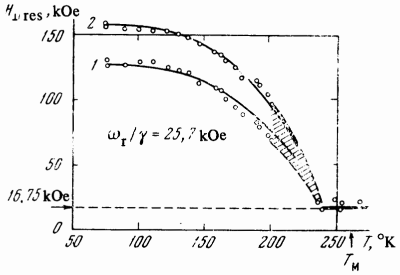

FIG. 5. Temperature dependence of the resonance fields for the case $H \perp C_3$ at a frequency $\nu_r = 72$ GHz (resonance-field measurement error ± 3 kOe). In the shaded region, the obtained resonance absorption does not lend itself to a simple and unambiguous reduction. The solid lines 1 and 2 are the result of calculation by formulas (12a) and (12b) for the low-field resonance (1) and by formula (13b) for the high-field resonance (2).

3. Resonance in perpendicular field ($H \perp C_3$). Figure 4 shows the temperature dependence of the main characteristics of the AFMR (at the working frequency $\nu_r = 37.7$ GHz) in hematite in the case when the external magnetic field was perpendicular to the C_3 axis of the crystal. At T = 77° K, two peaks are observed (in fields $H_{\perp res}^{(1)}$ and $H_{\perp res}^{(2)}$), which shift with increasing temperature, as expected, towards lower field values. The width of each of the peaks, roughly measured at half the height, is estimated by us to be 4 kOe and does not depend very strongly on the temperature. The peaks are separated from one another slightly, and therefore in Fig. 4, for simplicity, we show the temperature dependence of the resonance field of only the second (strong-field) peak of the resonance absorption. Long before the Morin point is reached, the intensity of both peaks decreases quite rapidly to zero: for sample No. 1 this "premature" vanishing of the resonance occurs st $T_V^{\perp} = 210 \pm 4$° K, and for sample No. 2 at $T_V^{\perp} = 202 \pm 4$° K. The rate of vanishing of the resonance with changing temperature is also slightly different for the two samples, but by virtue of the very low accuracy with which we determined the intensities of the peaks (simply from their height at constant microwave power at the spectrometer input), this difference cannot be described quantitatively. At temperatures exceeding 250° K, absorption again appears, corresponding to the usual resonance in the easy-plane phase[14].

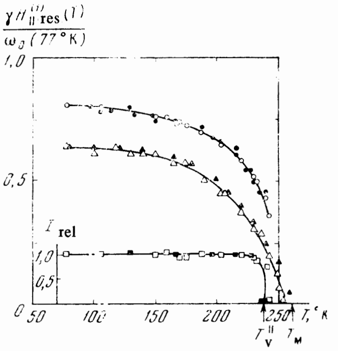

FIG. 6. Temperature dependence AFMR in hematite in a field parallel to the C_3 axis ($H_{\parallel res}^{(1)}$ – field in which a maximum of resonance absorption is observed; I_{rel} – relative intensity of this absorption): $\omega_0/\gamma | 77°K = 67 \pm 4$ kOe, $\nu_r = 37.7$ GHz, ● – $H_{\parallel res}^{(1)}$, sample No. 1, ○ – $H_{\parallel res}^{(1)}$, sample No. 2, ■ – I_{rel}, sample No. 1, □ – I_{rel}, sample No. 2; $\omega_0/\gamma | 77°K = 67 \pm 4$ kOe, $\nu_r = 72$ GHz, △ – $H_{\parallel res}^{(1)}$, sample No. 2, $\omega_0/\gamma | 77°K = 75 \pm 5$ kOe, $\nu_r = 70$ GHz, ▲ = $H_{\parallel res}^{(1)}$, data of [6].

The measurement of the temperature dependence of the positions of these peaks at double the frequency $\nu_r = 72$ GHz reveals a qualitatively different picture (Fig. 5): no clear cut vanishing of the resonance with increasing temperature is observed. In the region 200° < T < 240° K, the picture of the resonance absorption becomes more complicated. The number of peaks apparently becomes larger than two, and since their distances from one another begin to become comparable with their widths, it is difficult to obtain an unambiguous interpretation of the experimental data. In Fig. 5, this region is shown by the shaded section. With further increase of the temperature, the picture again becomes clearer: above 240° K there is observed one rather narrow ($\Delta H < 1$ kOe) absorption peak, the position of which does not depend on the temperature and is in good agreement with the formula $\omega = \gamma\sqrt{H(H + H_D)}$, which is characteristic in first approximation of AFMR in the easy-plane phase of the hematite[2].

4. Resonance in parallel field ($H \parallel C_3$). As a rule, the resonance of the easy-axis AF is investigated precisely at this orientation of the external field. Fonez and Williamson investigated the temperature dependence of $H_{\parallel res}$ in the low-temperature (easy-axis) hematite in the wavelength ranges $\lambda_r \sim 4$ and 2 mm[6]. We performed similar measurements in the ranges $\lambda_r \sim 4$ and 8 mm (see Fig. 6).

The agreement of the results at $\lambda_r \sim 4$ mm is almost complete—if we compare the temperature dependences of the resonance fields referred to 77° K. The difference in the absolute values ($H_{\parallel res} = 50$ kOe in[6] and $H_{\parallel res} = 41$ kOe in our case) is not surprising, since the magnitude of the anisotropy field (and consequently also of the resonance field) in hematite can differ for samples of different origin, owing to its strong dependence on the impurities[8,10].

At $\lambda_r \sim 8$ mm, the resonance absorption in a parallel field has a qualitative singularity similar to that described above for a perpendicular field: at a temperature $T_V^{\parallel} = 240 \pm 2$° K, its intensity drops rapidly to zero. Details of this effect, observed in both our samples, are given in the caption of Fig. 6.

4. DISCUSSION OF RESULTS

1) All these experimental facts can be described quite well within the framework of the calculation performed in Sec. 2. The quantitative reduction was performed by us as follows:

a) The static data $\chi_{\perp}^{(2)} = 1.95 \times 10^{-5}$ (± 5%) cm³/g and $\sigma^{(2)} = 0.442$ (± 5%) G – cm³/g of[12] were used in conjunction with the formulas $\chi_{\perp}^{(2)} = 2M_0/B$ and $\sigma^{(2)} = 2M_0\beta/B$ to determine the parameters B = 8960 kOe (±%) and β = 22.7 kOe (± 10%).

It was assumed that their magnitude does not change with temperature in the range 0–300° K, in good agreement with the data of[1,2]. For the saturation magnetization we assumed the pure-spin value $2M_0$ = 175 G – cm³/g.

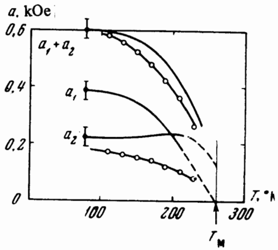

FIG. 7

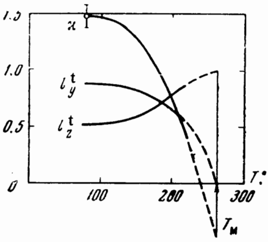

FIG. 8

FIG. 7. Temperature dependences of the first (a_1) and second (a_2) anisotropy constants in the hematite, obtained by reducing the data of Figs. 4 and 6 by means of formulas (13b) and (19) –solid lines. Light circles – results of Levitin and Shchurov [18], obtained under the assumption that the transition between the phases is realized in a field $H_\perp^{(1)}$ and not in $H_\perp^t$ (see Fig. 2).

FIG. 8. Temperature dependences of the parameter $\kappa \equiv (a_1 - \beta^2/B)/a_2$, and also of the y and z components of the AF vector in the transition field $H_\perp^t$ (T) for hematite

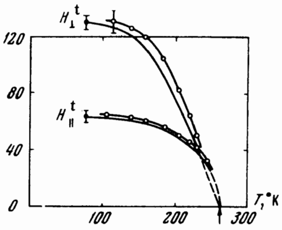

FIG. 9

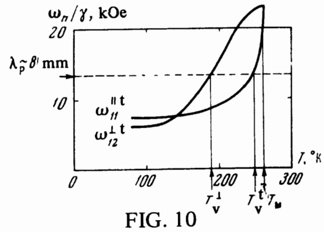

FIG. 10

FIG. 9. Comparison of the $H_\perp^t$(T) and $H_\parallel^t$ (T) dependences calculated from formulas (15a) and (21) (solid lines) with the experimental data of [12] (light circles) for hematite. The ordinate represents the magnetic field in kOe.

FIG. 10. Temperature dependence of AFMR frequencies at the transition points between the phases, calculated from formulas (15c) and (22): $\omega_{12}^{\perp t}$ (T) – value of the AFMR frequency, first branch, second phase, at a field perpendicular to the C_3 axis, in which the transition between the phases with $l_z \neq 0$ and $l_z = 0$ takes place at a temperature T. $\omega_{11}^{\parallel t}$ (T) – value of the AFMR frequencv, first branch, first phase, in a field parallel to the C_3 axis, in which a transition between the phases $l_z \neq 0$ and $l_z = 0$ takes place at the temperature T. The intersection of these curves with the horizontal line, corresponding to the working frequency, determines the temperature at which the vanishing of the corresponding resonance should take place (compare with Figs. 4 and 6). The ordinate represents the magnetic field in kOe.

b) Using the formula (13b), we obtained from the curve $H_{\perp\text{res}}^{(2)}(T)$ (Fig. 4) the $a_1(T)$ dependence (Fig. 7), assuming $H_{\perp\text{res}}^{(2)}(77°K) = 136$ kOe (± 5%)[9].

c) Using formula (19), we plotted from the $H_{\parallel\text{res}}^{(1)}(T)$ curve at $\nu_r = 37.7$ GHz (Fig. 6) the temperature dependence of $(a_1 + a_2)$, assuming $\omega_0(77°K) = 70$ kOe (± 5%). This quantity, for the purpose of greater generality of the subsequent results, was obtained by simple averaging of the experimental values given by various authors (naturally, for different hematite crystals): Fonez and Williamson[6]: 75 ± 5 kOe, Voronkov and Gromzin (private communication): 72 ± 5 kOe, Rudashevskiĭ and Mizonov (private communication): 66.3 ± 1 kOe, and the present investigation (Fig. 6): 67 ± 4 kOe.

The $a_1(T)$ and $a_2(T)$ plots obtained in this manner are shown in Fig. 7, where they are also compared with the corresponding results of[18] is given by $a_1 + a_2 \to a$ and $a_2 \to -b$). The noticeable difference between the curves for $a_2(T)$ is apparently connected with the fact that in[18] it was assumed that the real transition between the phases with $l_z \neq 0$ and with $l_z = 0$ occurs in hematite at perpendicular values $H_\perp = H_\perp^{(1)}$ (see formula (15e)), and not $H_\perp = H_\perp^t$ (see (15a)).

2. Now, knowing the main parameters of the theory (B, β, a_1, and a_2) and their temperature dependence, we can quantitatively calculate all the experimentally measured quantities. It is first useful to construct the temperature dependences of such an important parameter as $\kappa \equiv (a_1 - \beta^2/B)/a_2$, and also the critical values of the y and z components of the AF vector (l_y^t and l_z^t), at which a jumplike transition to the second phase (where $l_z = 0$ and $l_y \cong 1$) takes place with increasing field perpendicular to the C_3 axis (see Fig. 8).

After plotting, with the aid of expressions (15a) and (21) and of the data of Figs. 7 and 8, the temperature dependences of the phase-transition fields $H_\perp^t(T)$ and and $H_\parallel^t(T)$, we obtain curves that agree satisfactorily with the experimental data of Voskanyan, Levitan, and Shchuzov[12] (see Fig. 9). We note here once more that in the case considered by us the magnitude of the field $H_\parallel^t$ of "sublattice flipping" does not coincide with that value of the parallel field $H_\parallel^{(1)} = \omega_0$, at which one of the frequencies of the ordinary AFMR tends to zero. For example, in hematite $H_\parallel^t(77°K) \cong 63$ kOe, whereas $\omega_0(77°K) \cong 70$ kOe (see Fig. 3). The agreement between these two quantities takes place only in the absence of an appreciable contribution of the second anisotropy constant ($a_2 \ll a_1$).

3) The phenomena of "premature" vanishing of the resonance both for perpendicular and parallel orientations of the external magnetic field can also be analyzed quantitatively. Figure 10 shows the temperature dependences of the "gaps" $\omega_{12}^{\perp t}$ and $\omega_{11}^{\parallel t}$ in the AFMR spectra, calculated from formulas (15c) and (22) (see also Figs. 2 and 3). By drawing through these curves a horizontal line corresponding to the operating frequency (in our case $\omega_r = 13.5$ kOe), we obtain the temperature values $T_v^\perp = 190°K$ and $T_v^\parallel = 248°K$, at which vanishing of the resonance absorption should take place in fields perpendicular and parallel to the EA. This is in satisfactory agreement with the experimental data (Figs. 4 and 6).

By virtue of the fact that in plotting $a_1(T)$ and $a_2(T)$ we started from the curves $H_{\perp\text{res}}^{(2)}(T)$ and $H_{\parallel\text{res}}^{(1)}(T)$ at $\omega_r = 13.5$ kOe ($\lambda_r \sim 8$ mm), which experience a break in the temperature region near 220°K (Figs. 4 and 6), the curves of Figs. 7–10 have a very low accuracy at $220° < T < T_M$. In particular, the value of $\omega_{12}^{\parallel t}(T_M) = \omega_{11}^{\parallel t}(T_M) \equiv \omega_M$ was obtained after extrapolating the function $a_2(T)$ to $T = T_M$ (see Fig. 7), which cannot be performed without ambiguity from our experimental data alone. However, it is obvious that as the "gaps" $\omega_{12}^{\perp t}$ and $\omega_{11}^{\parallel t}$ in the AFMR spectrum increase with increasing temperature, they cannot exceed a certain value ω_M. In our case $\omega_M = 23$ kOe, i.e., it is such that the "premature" vanishing of the resonance should not take place already at $\lambda_r \sim 4$ mm. This agrees with experiment (see Figs. 5 and 6). The reduction of the data of[11] for $H_\perp^t(T)$ and

$H_{\parallel}^{t}(T)$ near T_M gives for ω_M a somewhat larger value: $a_2(T_M) \sim 0.45$ kOe, which leads to $\omega_M \sim 45$ kOe. Therefore it is possible that the region of zero intensity of AFMR at $H \perp C_3$ and $\lambda_r \sim 4$ mm still exists, but obviously it is very small (not more than several degrees K).

4) From an examination of Fig. 10 (and also of Fig. 2b) we draw one more conclusion. In hematite, the phase transition in a perpendicular field at 77° K, as well as in temperatures close to T_M, is apparently a first-order transition ($\kappa'_{77^\circ K} < 2$—see Fig. 8), but the degree to which it is a first order transition, i.e., in particular, the magnitude of the magnetization jump in the transition field $H_{\perp}^{t}$, is small (see formulas (15d) and (18b)). Therefore it is experimentally difficult to determine the order of the transition at 77° K from the $m_{\perp}(H_{\perp})$ curve (all the more, since the sharpness of the jump can be greatly decreased by unaccounted for interactions), but this can be quite readily done with the aid of resonance measurements at low frequencies, for example, using the $\omega_{12}^{\perp t}(T)$ plot of Fig. 10.

5) Satisfactory agreement with the experimental data (both static and dynamic) is attained for $T = 77^\circ K$ at values $\beta = 22.7$ kOe, and $a_1 = 0.382$ kOe, which differ from those obtained in[9] ($\beta' = 30$ kOe, $a' = 0.54$ kOe) on the basis of a reduction of the resonance data with the aid of the formulas of the simplest theory (without allowance for the second anisotropy constant). Nonetheless it turns out that the degree of quantitative agreement between theory and experiment on the angular dependence of the resonance field (Fig. 7 in[13]) remains practically unchanged. As already noted, expressing the fourth-order anisotropy energy in the form $-(\frac{1}{4})a_2 l_z^4$ does not change any of the formulas for the phase with $l_z = 0$. This includes the validity of the formula (22) of[13], which connects the z and x components of the magnetic field in which the second absorption peak is observed. Introduction of the second anisotropy constant decreases both $\beta(77^\circ K)$ and $a(77^\circ K)$. Therefore the coefficients of Eq. (22) of[13] change so little, that the curve 2 of Fig. 7 in[13] fits the experimental points as well as before.

As to the phase diagram in the $(H_x H_z)$ plane, the introduction of a term with a_2 complicates the picture. It can be shown that for low temperatures ($\sim 77^\circ K$) the boundary between the phases with $l_z \neq 0$ and with $l_z = 0$ in the $(H_x H_z)$ plane begins to differ noticeably from the surrounding only for directions that are very close to the $z \parallel C_3$ axis. On the other hand, with increasing temperature, the region of these directions increases. A complete quantitative analysis of the phase diagram, i.e., of the $H_z^t(H_x^t, T)$ dependence, can be performed in this case apparently only with the aid of numerical methods.

5. CONCLUSION

The frequency analysis method used in Sec. 2 to clarify the influence of high-order terms on the resonance AF spectrum is of considerably general interest. In particular, it is possible to draw on its basis definite conclusions concerning the possibilities of experimentally observing the resonance in a perpendicular field in a large number of easy-axis AF with a strong Dzyaloshinskiĭ interaction (for example, in CoF_2—see[20]). The fact that for hematite it is possible to derive explicit expressions by means of this analysis is connected with the unusual relation between the magnitudes of the main interactions in this substance ($a_1 \ll \beta \ll B$). The quantitative agreement between our experimental data on the resonance and the calculated ones for the temperature range $77^\circ K < T < 220^\circ K$ makes it desirable to carry out an analogous investigation (primarily at frequencies larger than 72 GHz) in the immediate vicinity of the Morin point, which makes it possible to refine the character of the functions $a_1(T)$ and $a_2(T)$ near T_M.

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