Statics and Linear Dynamics of Orthoferrites—Phase Transition of the "One-and-One-Halfth Kind"

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A phenomenological calculation of the static and dynamic properties of orthorhombic antiferromagnets, with effective fields that satisfy the relations H_A, $H_D \ll 2H_E$, is carried out by taking into account two "bilinear" and three "biquadratic" anisotropy constants. A satisfactory agreement is obtained between the experimentally determined and the calculated orientational dependence of the antiferromagnetic resonance (AFMR) fields in $YFeO_3$ at $T = 300°K$ and $\nu = 128$ GHz. Consideration is given to a critical point of phase transitions of the second kind, at which the phase transition with respect to the field is characterized by an infinite discontinuity of the differential susceptibility while the magnetic moment changes continuously (a phase transition of the "one-and-one-halfth kind"). An experimental investigation of the dependence of the differential susceptibility on field and temperature justifies the supposition that such a transition exists in $SmFeO_3$.

1. INTRODUCTION

THE dependence of antiferromagnetic resonance (AFMR) frequencies on the value of the magnetic field and on its orientation, in orthoferrites and in compounds of the orthorhombic syngony related to them, has been investigated in recent years in a whole series of laboratories[1-5]; and this work has led to an understanding of the basic characteristics of the effect of various interactions on the magnetostatics and the linear magnetodynamics of these systems. Thus observation of AFMR in $TmFeO_3$ near the temperature of reorientation of the AF-vector[1] and interpretation of these results[2,3] have revealed the important role of the "second" anisotropy constants (which, by analogy with "biquadratic" exchange[6], we shall call the "biquadratic" anisotropy constants). The effect of biquadratic anisotropy on the statics of orthoferrites in small fields, near the reorientation region, has been investigated in considerable detail[7-10]. Recent precision measurements of the static magnetic properties of $YFeO_3$ in strong fields and related calculations made by Jacobs, Burne, and Levinson[11], as well as available quantitative static characteristics of rare-earth orthoferrites (for example, $SmFeO_3$[12,13]), show that allowance for biquadratic anisotropy is necessary for consistent description of the experimental data even far from the reorientation temperatures (or in the absence of any, as in $YFeO_3$). For the purpose of explaining the effect of biquadratic anisotropy on the magnetostatics and dynamics of orthoferrites over a range of temperatures not necessarily close to the reorientation region, we have investigated AFMR in $YFeO_3$ and $SmFeO_3$ and the temperature dependence of the value of the phase-transition field in $SmFeO_3$. In our phenomenological calculation of the static and dynamic properties of orthoferrites, for a broad range of values of the external field $H \ll 2H_E$ and for arbitrary orientation of it in the (ac) plane of the crystal, biquadratic anisotropy has been taken into account in a form corresponding to symmetry requirements (as was done also by Belov and others[8] in their analysis of the phenomenon of reorientation in $SmFeO_3$).

2. BASIC EQUATIONS

We use the two-sublattice model of an antiferromagnetic orthoferrite belonging to space group D_{2h}^{16}. We write the magnetic energy of the system at temperature T (sufficiently low in comparison with T_N), and for arbitrary orientation of the external field H in the (ac) plane (the most interesting case), in the form

$$\mathcal{E} \equiv 2M_T\mathcal{H}, \tag{1a}$$

$$\mathcal{H} = \tfrac{1}{2}E\mathbf{m}^2 - D(m_zl_x - m_xl_z) - \mathbf{mH} + \tfrac{1}{2}a_1l_x^2 + \tfrac{1}{2}c_1l_z^2 + \tfrac{1}{4}a_2l_x^4 + \tfrac{1}{4}c_2l_z^4 + \tfrac{1}{2}fl_x^2l_z^2. \tag{1b}$$

Here $\mathbf{m} \equiv (\mathbf{M}_1 + \mathbf{M}_2)/2M_T$ and $\mathbf{l} \equiv (\mathbf{M}_1 - \mathbf{M}_2)/2M_T$ are the ferro- and antiferromagnetic vectors, respectively; $\mathbf{M}_1$ and $\mathbf{M}_2$ are the magnetic moments of the sublattices; M_T is their value at the given temperature T, such that $M_0 - M_T \ll M_0$; the x, y, z axes are directed along the axes [a], [b], and [c] of the crystal ($a < b < c$); $\mathbf{H} = (H\cos\theta, 0, H\sin\theta)$—see Fig. 1a. We neglect anisotropy of the g-factor. The Dzyaloshinskiĭ interaction energy should, in accordance with symmetry requirements, be written in the form

$$\mathcal{H}_D = -d_1m_zl_x + d_2m_xl_z \equiv -D(m_zl_x - m_xl_z) - A_{xz}(m_zl_x + m_xl_z),$$

but from experimental data on $YFeO_3$[11] and $SmFeO_3$[12], in these materials (and in a whole series of other orthoferrites) $A_{XZ} \ll D$, and therefore we shall neglect the term containing A_{XZ}. Because the expression for the energy has been written in the normalized form (1a, b), the coefficients that appear in it are measured in oersteds (gausses). $\frac{1}{2}E$ and D are the effective symmetric and antisymmetric exchange fields ($E \equiv 2H_E$; $D \equiv H_D$; $E > 0$; $D > 0$); $A \equiv \{a_1, c_1, a_2, c_2, f\}$ are the effective bilinear (a_1 and c_1) and biquadratic (a_2, c_2, and f) anisotropy fields. Being interested only in the region of sufficiently low temperatures, we shall make use of the conditions

$$\mathbf{m}^2 + \mathbf{l}^2 = 1, \qquad \mathbf{ml} = 0, \tag{2}$$

which follow from $|\mathbf{M}_1|^2 = |\mathbf{M}_2|^2 = M_T^2$.

For all orthoferrites, experiment shows[14] that the inequalities

$$|A| \ll D \ll E, \tag{3}$$

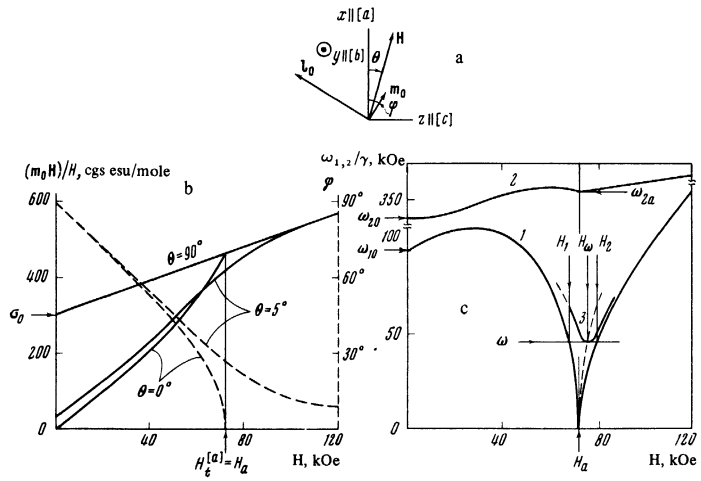

FIG. 1. Analysis of the statics and dynamics of orthorhombic antiferromagnets with large Dzyaloshinskiĭ interaction. a: Coordinate axes and basic variables of the problem. b: Static characteristics of the system when $\Lambda_a > 0$ ([a] is the easiest axis); calculation for the specific case of $YFeO_3$ at 4.2°K (see [11]). c: AFMR spectrum (g = 2). Curves 1 and 2: the functions $\omega_1(H)$ and $\omega_2(H)$ at $\theta = 0$, calculated for the specific values E = 12 800 kOe, D = 140 kOe, A_0 = 0.68 kOe, A_a = 1.2 kOe that follow from paper [11] for $YFeO_3$ at 4.2°K. In the calculation of $\omega_2(H)$, the values $A_x = A_z = -7.5$ kOe have been used in addition, for illustration. Curve 3: calculation of the function $\omega_1(H)$ for disorientation angle $\theta = \theta_\omega = 0.85°$, at which the curve $\omega_1(H)$ in field $H = H_\omega \approx 2H_a - H_1$ has a minimum equal to operating frequency $\omega = 8\cdot10^{11}$ sec^{-1}.

are satisfied; these significantly facilitate the calculation for an arbitrary type of anisotropy, since because of them, by analogy to the situation in hematite[15], the parameter $m \ll l \sim 1$ is small over the whole field range of practical interest, from zero to $H \sim (AE)^{1/2}$ or $H \sim AE/D$. This fact permits us, in writing the anisotropy energy, to restrict ourselves to terms containing only components of the AF-vector l. The specific results obtained below are valid also under the more general condition

$$|A|,\ D \ll E. \tag{3a}$$

We find the stationary state ($\mathbf{m}_0$, l_0) and the resonance frequencies for small oscillations of the quantities $\mu \equiv \mathbf{m} - \mathbf{m}_0$, $\lambda \equiv l - l_0$ from the equations of motion for m and l; these automatically take account of the conditions (2):

$$\gamma^{-1}\dot{\mathbf{m}} = -[\mathbf{m}\mathbf{H}_m] - [l\mathbf{H}_l],\quad \gamma^{-1}\dot{l} = -[\mathbf{m}\mathbf{H}_l] - [l\mathbf{H}_m];$$
$$\mathbf{H}_m \equiv -\partial\mathcal{H}/\partial\mathbf{m},\quad \mathbf{H}_l \equiv -\partial\mathcal{H}/\partial l. \tag{4}*$$

3. THE STATIC CASE

Taking account of conditions (3), we get for $\mathbf{m}_0$ and l_0 (see Fig. 1a)

$$\mathbf{m}_0 = (m_0\cos\varphi,\ 0,\ m_0\sin\varphi),\quad l_0 \approx (\sin\varphi,\ 0,\ -\cos\varphi), \tag{5}$$

where the functions $\varphi(H, \theta)$ and $m_0(H, \theta)$ are given by the equations

$$[D + H\cos(\varphi - \theta)]H\sin(\varphi - \theta) = E(A_0\sin^2\varphi + A_a\cos^2\varphi)\sin\varphi\cos\varphi, \tag{6a}$$

$$m_0 = [D + H\cos(\varphi - \theta)]/E. \tag{6b}$$

Here and below we use the notation

$$A_0 \equiv c_1 + f - a_1 - a_2 \equiv -A_c,\quad A_x \equiv a_1 + a_2,$$
$$A_a \equiv c_1 + c_2 - a_1 - f,\quad A_z \equiv c_1 + c_2. \tag{6c}$$

We shall introduce at once the following quantities, which are convenient for what is to follow:

$$H_a \equiv {}^1/_2[-D + (D^2 + 4EA_a)^{1/2}],\ \text{i.e.}\ H_a^2 + DH_a \equiv EA_a; \tag{7a}$$

$$H_c \equiv {}^1/_2[-D + (D^2 + 4EA_c)^{1/2}],\ \text{i.e.}\ H_c^2 + DH_c \equiv EA_c; \tag{7b}$$

$$A_2 \equiv A_a - {}^1/_2A_0 = a_2 + c_2 - 2f. \tag{7c}$$

We shall consider below only systems with initial (at H = 0) state

$$\mathbf{l}_{00} \approx (1, 0, 0),\quad \mathbf{m}_{00} \approx (0, 0, D/E),$$

this is the case in $YFeO_3$ and in most orthoferrites at temperatures below the reorientation range. To facilitate the obtaining of numerical values of A_0 and A_a from static measurements, we note that in using the results of Jacobs and others[11] it is necessary to set $A_0 = H_{k2} - 2H_{k4}$, $A_a = H_{k2}$.

From (6a, b) follow the well-known results shown in Fig. 1b. Here

$$2M_T/E = \chi_m,\quad D = \sigma_0/\chi_m,\quad EA_a = \sigma_0^2/\chi_l\chi_m, \tag{8}$$

where χ_m and χ_l are the initial susceptibilities along the original directions of the ferro- and antiferromagnetic vectors (that is, in the case of $YFeO_3$, along the axes [c] and [a] respectively).

4. DYNAMICS

Equations (4) are the starting point for a standard calculation of the resonance frequencies of the systems under consideration, for arbitrary value of the field H and for arbitrary orientation θ of the field in the (ac) plane. The result can be expressed in a form quite convenient for analysis or for calculation. The two AFMR frequencies $\omega_{1,2}(H, \theta)$ are determined by the equation

$$(\omega/\gamma)^4 - (U + V)(\omega/\gamma)^2 + (UV - W) = 0; \tag{9a}$$

where

$$U \equiv HEm_0 - E(A_0\sin^2\varphi + A_a\cos^2\varphi)\cos 2\varphi + 2EA_2\sin^2\varphi\cos^2\varphi, \tag{9b}$$

$$V \equiv DEm_0 - E(A_x\sin^2\varphi + A_z\cos^2\varphi) + EA_2\sin^2\varphi\cos^2\varphi + H^2\sin^2(\varphi \tag{9c}$$

$$W \equiv 2H^2\sin(\varphi - \theta)\{EA_0\sin\theta\cos^3\varphi + EA_0\cos\theta\sin^3\varphi +$$
$$+ [DEm_0 - E(A_x\sin^2\varphi + A_z\cos^2\varphi) + 4EA_2\sin^2\varphi\cos^2\varphi]\sin(\varphi - \theta)\}. \tag{9d}$$

The values of φ and m_0 as functions of H and θ are given by equations (6a, 6b). It follows from the expressions (6) and (9) that for complete description of the static behavior of the system, two combinations (A_0 and A_a) of the five anisotropy constants involved are sufficient, whereas the dynamics requires four: A_0, A_a, A_x, and A_z. This shows up even in the easily calculated special cases: for $\theta = \pi/2$ and $H \ll E$,

$$\varphi = \pi/2,\quad (\omega_1/\gamma)^2 = H^2 + DH + EA_0,$$
$$(\omega_2/\gamma)^2 = D^2 + DH - EA_x; \tag{10a}$$

for $\theta = 0$ and $H_a \leq H \ll E$,

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

$$\varphi = 0, \quad (\omega_1/\gamma)^2 = H^2 + DH - EA_x, \quad (\omega_2/\gamma)^2 = D^2 + DH - EA_x. \tag{10b}$$

For quantitative determination of A_x and A_z, it is necessary to measure ω_{20} and ω_{2a} (see Fig. 1c); in actual materials, these as a rule lie in the hard-to-reach submillimeter range. In general, the values of A_x and A_z affect also the values of ω_1 for $0 < H < H_a$; but as is shown by a numerical calculation that we have made by means of (9), this effect, which reaches a maximum at $\varphi \sim 45°$, is very small. For example, for materials of the type of $YFeO_3$, the difference in ω_1 at $H \sim 50$ kOe, for $A_x \approx A_z$ equal to -7.5 kOe and to -15 kOe, does not exceed 1%.

5. ANALYSIS OF SPECIAL CASES

In strong fields ($H \sim H_a$) parallel to the initial (at $H = 0$) direction of the AF-vector, it follows from (6a) that a phase transition occurs from the "xz-phase" (in which, for $H \neq 0$, both the x- and the z-components of the AF-vector are nonzero) to the "z-phase", in which the AF-vector is directed strictly along the z axis, while $m_{x_0} = (D + H)/E$. This transition, depending on the relation between the magnitudes of the different interactions, may be a transition of the second or of the first kind (with respect to the field). To find this relation, we set $\theta = 0$ in (6a), denote $\cos\varphi$ by ζ, and obtain for H

$$H = \{-D + [D^2 + 4E(A_0 + A_2\zeta^2)\zeta^2]^{1/2}\}/2\zeta. \tag{11}$$

It is easy to show (by analyzing, by means of (6a), the behavior of the differential susceptibility $\chi_x \equiv dm_x/dH$ at $\theta = 0$) that the nature of the transition is determined by the sign of the derivative $dH/d\zeta$ at the point ($H = H_a$, $\zeta = 1$), through which the curve $H(\zeta)$ necessarily passes by virtue of the definition (7a) of the quantity H_a. In calculating this derivative, we shall introduce the quantity Λ_a (in general temperature-dependent), whose sign coincides with the sign of the derivative under consideration:

$$\Lambda_a(T) \equiv {}^1/_2 DH_a + EA_2 \equiv H_a^2 + {}^3/_2 DH_a - EA_0. \tag{12}$$

Then by use of the equation of state (6a) and the results of the calculation of the dynamics of the system, we arrive at the following conslusions.

a) $\Lambda_a > 0$, $\theta = 0$. The transition $xz \rightarrow z$ is a transition of the second kind and occurs at field $H_t = H_a$. As parameter of the transition we may take the angle φ (or l_{x_0}), which for $0 \leq H \leq H_a$ is determined by equation (11). Near the critical field, this parameter depends on the field in the typical fashion

$$\varphi \approx l_{x_0} \approx \left[\frac{2H_a + D}{\Lambda}(H_a - H)\right]^{1/2} \quad \text{for } H_a - H \ll \frac{\Lambda_a}{2H_a + D}. \tag{13}$$

The dependence of the AFMR frequencies on the value of the field, as follows from formulas (9a–d) for $\theta = 0$, differs slightly (at small H) from that obtained by Cinader[16]; it is depicted in Fig. 1c. We note again the characteristic points of this figure (see also (10)):

$$(\omega_{10}/\gamma)^2 = EA_0 = \sigma_0^2/\chi_m\chi_\perp, \tag{14a}$$

$$(\omega_{20}/\gamma)^2 = D^2 - EA_x, \tag{14b}$$

$$(\omega_{2a}/\gamma)^2 = D(H_a + D) - EA_x. \tag{14c}$$

b) $\Lambda_a > 0$, $\theta \neq 0$. As follows from (6a), the component l_{x_0} does not vanish for any values of H. Consequently, in the (H_x, H_z) plane the point (H_a, 0) is an isolated phase-transition point of the second kind (with respect to the field). Near this point there occurs a characteristic relationship (see Fig. 1c). Thus for small $\theta \ll 1$, on expanding the formal function $\theta(H_a, \varphi)$ in a series of powers of $\varphi \ll 1$ by means of (6a), we get

$$\varphi(H_a, \theta) \approx [H_a(H_a + D)\theta/\Lambda_a]^{1/2}. \tag{15}$$

Similar cube-root dependences are encountered in the investigation of other isolated phase-transition points of the second kind (for example, in the (T, H) plane at $T = T_c$ in ferromagnets, or at $T = T_N$ in antiferromagnets with weak ferromagnetism, where $l_{y_0}(T_N, H_x) \sim H_x^{1/3}$; see[17]).

The range of fields H and angles θ near the point (H_a, 0) is of particular interest from the experimental point of view, since the AFMR frequencies necessary for observation in this range are not too high. The field and orientation dependences of the lower AFMR frequency near this point can be obtained by expanding the functions θ and ω_1^2 as series in $h \equiv H - H_a \ll H_a$ and in $\varphi \ll 1$ by means of the general relations (6) and (9) (see Fig. 1c):

$$h_2 \equiv H_2 - H_a = \omega^2/\gamma^2(2H_a + D), \tag{16a}$$

$$h_\omega \approx h_2/2, \tag{16b}$$

$$h_1 \approx -h_2/2; \tag{16c}$$

$$\theta_\omega \approx 2^{1/2}(\omega/\gamma)^2/3H_a(H_a + D)(3\Lambda_a)^{1/2}. \tag{16d}$$

The last expression gives the limiting angle θ_ω of inclination of the magnetic field to the direction of the easy axis of the crystal (in the (ac) plane), beyond which observation of AFMR at a given frequency ω becomes impossible (if the resonance line is infinitely sharp). In an actual case, with allowance for finite complex susceptibilities (see[18]), "fade-out" of the absorption is delayed until slightly larger angles.

c) $\Lambda_a < 0$. In this case, for $\theta = 0$, the transition $xz \rightarrow z$ occurs as a transition of the first kind at field H_t (see Fig. 2). The angle φ changes discontinuously at this field from φ_t to zero. Other characteristics of the system also undergo discontinuous changes. It can be shown that, by analogy to the situation in hematite[15],

$$H^{(2)} < H_t < H^{(1)}, \tag{17}$$

where $H^{(2)} = H_a$ is the lower instability field. The upper field after instability, $H^{(1)}$, is determined by the condi-

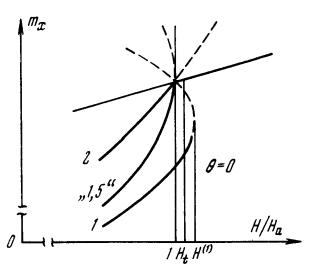

FIG. 2. Possible types of magnetization curve of the system near the phase-transition field H_t[a]. Curve 1 occurs when $\Lambda_a < 0$ and characterizes a phase transition of the first kind (with resepct to the field). Curve 2 occurs when $\Lambda_a > 0$: a phase transition of the second kind. Curve "1.5" occurs when $\Lambda_a = 0$: phase transition of the "one-and-one-halfth kind"; at the point $H = H_a$ the function $m_x(H)$ has a singularity $\sim (H_a - H)^{1/2}$, the function $\chi_x(H)$ a singularity $(H_a - H)^{-1/2}$.

tion $dH/d\zeta = 0$ (see (11)), and the transition field H_t is determined from the condition that the difference of energies of the two phases shall vanish at the transition point:

$$\mathcal{H}_{xz} - \mathcal{H}_z = 0.$$

d) For answering the question of the kind of transition, practical use of the parameter Λ_a described in the form (12) is not altogether convenient. Experimentally, it is easy to measure the values of σ_0, χ_m, χ_l, and the transition field H_t (which is not equal to H_a if $\Lambda_a < 0$). It therefore makes sense to introduce the quantity

$$\Lambda_{exp} \equiv H_t^2 + \frac{3}{2}\frac{\sigma_0}{\chi_m} H_t - \frac{\sigma_0^2}{\chi_l \chi_m}, \qquad (18)$$

which in general is larger than or equal to the quantity Λ_a (see (12), (8), and (17)). Then $\Lambda_{exp} > 0$ will be the criterion for a transition of the second kind (in this case $H_t = H_a$ and $\Lambda_{exp} = \Lambda_a$), whereas $\Lambda_{exp} < 0$ will be the criterion for a transition of the first kind (since in this case $\Lambda_a < 0$ because $\Lambda_a < \Lambda_{exp}$)[1)].

The possibility is not excluded that there might be a crystal in which, at a certain temperature T_b, the parameter $\Lambda_a(T)$ vanishes: $\Lambda_a(T_b) = 0$. This limiting case is curious, since for $T = T_b$ the field-induced phase transition xz → z possesses properties in a certain sense intermediate between those of phase transitions of the second and of the first kind. In accordance with the definition introduced in the book of Landau and Lifshitz[19], T_b is a critical point of phase transitions of the second kind. We shall consider this case in somewhat greater detail.

e) For small departures of the field direction from the axis [a] and of the field magnitude from the value H_a—more exactly, for those values of $\theta \ll 1$ and of $h \equiv H - H_a \ll H_a$ for which $\varphi \ll 1$—we can obtain from (6a) and (9a—d)

$$H_a(H_a + D)\theta \approx [(2H_a + D)h + \Lambda_a\varphi^2 + P\varphi^4]\varphi, \qquad (19a)$$

$$(\omega_1/\gamma)^2 \approx (2H_a + D)h + 3\Lambda_a\varphi^2 + Q\varphi^4,$$
$$P \equiv {}^3/_8 DH_a - \Lambda_a, \quad Q \equiv \mathcal{F}(T)\Lambda_a + H_a({}^{15}/_8 D - {}^2/_3 H_a) \qquad (19b)$$

(here $\mathcal{F}(T)$ is a definite function of the effective fields).

Under conditions that permit neglect of the terms containing φ^4, and for $\Lambda_a > 0$, it is easy to derive the relations (13), (15), and (16) given above. They describe an isolated point of phase transition of the second kind. The corresponding relations for $\Lambda_a = 0$ have the form

$$\varphi(H_a, 0) \approx \left[\frac{8(2H_a + D)}{3DH_a}(H_a - H)\right]^{1/4}, \quad \varphi(H_a, \theta) \approx \left[\frac{8(H_a + D)}{3D}\theta\right]^{1/5},$$
$$\theta \sim \omega^{5/2}, \quad h_1, h_\omega \sim \omega^2.$$

Thus at $T = T_b$, when $\Lambda_a = 0$, the point $(H_a, 0)$ in the (H_x, H_y) plane is, as before, an isolated point of phase transition, at which the transition parameter φ, with increase of field, tends continuously toward zero at the transition field $H_t = H_a$; but its approach to zero from the smaller-field side occurs not along a parabola, but along a curve $(H_a - H)^{1/4}$. Therefore, although the magnetic moment m_x of the system (a quantity proportional to the first derivative of the thermodynamic potentials with respect to the field) also undergoes no discontinuity at $H = H_a$, the differential susceptibility dm_x/dH becomes infinite at this field (from the low-field side), whereas in an ordinary transition of the second kind it should experience only a finite jump. This intermediate situation characterizes a so-called critical point of phase transitions of the second kind (in our case, in the (H, T) plane; see[19]). By analogy with[20] we may, for brevity, call this transition with respect to the field when $\Lambda = 0$ a phase transition of the "one-and-one-halfth kind" (see Fig. 2). By using the results of paragraphs b), c), and e) of this section, one can show that in accordance with the general theory[19], the $H_t(T)$ curve undergoes a discontinuity of the second derivative at the point $T = T_b$.

6. ANTIFERROMAGNETIC RESONANCE IN $YFeO_3$ AND $SmFeO_3$

a) Apparatus. We have investigated the absorption of microwave radiation, of wavelengths $\lambda \sim 2$, 4, 6, and 8 mm, in monocrystals of $YFeO_3$ ($T_N = 644°K$) and of $SmFeO_3$ ($T_N = 674°K$), as a function of the value of the field and of its orientation with respect to the crystalline axes, by means of a reflection radiospectrometer, with a pulsed magnetic field ($H_{max} = 300$ kOe; duration of the field, from zero to zero, $\tau_{00} \sim 10$ msec). The specimen being investigated was placed near the shorted end of a waveguide insert, on a circular table of diameter 2.5 mm, whose surface formed part of the inner surface of the wide wall of a waveguide of 8-millimeter range. Rotation of the table about a horizontal axis was accomplished by means of a worm gear that insured an accuracy of setting of the angle of rotation no worse than 20′ of arc. The whole waveguide insert, of length about 500 mm, was fastened on the Dewar cap, which could be moved along a cylindrical surface. The horizontal axis of this surface went through the center of the specimen and made a right angle with the axis of rotation of the table. The maximum possible angle of inclination of the waveguide insert to the vertical axis (which coincided with the axis of the pulsed solenoid) was 4°; the accuracy of setting of the angle was no worse than 10′.

b) Specimens. A monocrystal of $YFeO_3$ was kindly provided to us by the Semiconductor Institute, Academy of Sciences, USSR (Leningrad); it belonged to the same group of specimens as those used by Yudin and others others[21]. The monocrystal of samarium orthoferrite used for resonance measurements was grown by the method of crucibleless zone fusion with radiation heating[22]. The content of iron of various valences was monitored according to the sharpness of the edge of the window of transparency in the infrared part of the spectrum. Practially all the iron was in the trivalent state. The absorption coefficient (α) at wavelength 1.2 microns was 12 cm^{-1}. The $SmFeO_3$ monocrystal on which the measurements of differential susceptibility were made was grown from a solution (oxides of the rare earth in oxides of lead and boron) in the melt; it was kindly provided to us by J. Marechal (France). All specimens had approximately the shape of a cube of edge about 3 mm. The critical fields of the $SmFeO_3$ specimens, measured dynamically (by AFMR) and quasistatically (by the dif-

[1)] In hematite (α-Fe_2O_3) at $T < T_m$, as follows from[15], an analogous experimental criterion for the kind of transition, in a field perpendicular to C_3 (at fixed T), is the sign of the quantity $\Lambda \equiv 3(\chi_\perp^{AF} - \chi_\perp^{WF})H_{\perp t} - 2\sigma_0^{WF}$.

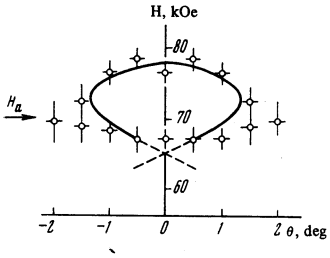

FIG. 3. Orientational dependence of the AFMR field at frequency ν = 128 GHz in $YFeO_3$ at T = 300°K. The value of the field H_a was determined (with absolute error ±2.5 kOe) from the position of the unresolved absorption maximum at frequency 37 GHz. The curve was drawn according to formulas (19a, b) with P = Q = 0, by use of static data of Yudin and others [23]. On the dotted part of this curve the condition $\varphi \ll 1$ is poorly satisfied.

ferential susceptibility), agreed within the limits of absolute error of the field measurements (± 5%).

The fastening of the specimens to the table was performed in a constant magnetic field, in such a way that their spontaneous magnetic moment (and consequently the axis [c] for $YFeO_3$ and the axis [a] for $SmFeO_3$) was perpendicular to the surface of the table. The fundamental measurements of the orientational dependence of the resonance fields were made by rotation of the waveguide insert without change of the position of the specimen with respect to the waveguide.

c) Results. Figure 3 shows the orientational (in the ac plane) dependence of the values of the magnetic field at which maxima of the absorption of microwave energy occur in $YFeO_3$, with frequency ν = 128 GHz. At room temperature, the width ΔH of each of the two resonance lines, when the field was oriented strictly along the axis [a], that is when $\theta = 0°$ (±10′), was 1.5 ± 0.5 kOe. With inclination of the field to the axis [a], the lines draw closer together, and at $\theta_\omega^{exp} = 1.5°$ they are almost completely fused. In the angle interval 1.5 to 2.5°, a single absorption maximum is observed; its intensity diminishes with increase of $|\theta|$ and vanishes for $|\theta| \geq 2.5°$. At frequencies corresponding to wavelengths ~ 4, 6, and 8 mm, even at $\theta = 0°$ an unresolved absorption maximum is observed; according to[18], it is a field-dependent absorption at the edge of the line belonging to the real branch $\omega_1(H)$ of the AFMR. This maximum at $\theta = 0$ occurs at the phase-transition field H_t, the measured value of which (with absolute error ± 2.5 kOe), at 300°K, is shown in Fig. 3.

In $SmFeO_3$ at 300°K and $\theta = 0$, at frequency 129 GHz, two wide ($\Delta H \sim 6$ kOe) lines are observed, at field values 46 and 60.5 kOe; with increase of the disorientation angle θ, they begin to draw closer together, but more slowly than in $YFeO_3$. The resolution is maintained up to $\theta = 4°$.

d) Discussion. The phenomenological calculation made above enables us to describe the experimental results quantitatively with sufficient accuracy (for $YFeO_3$, the better, the closer to 0°K). By using the data on static measurements available in the literature,[11,21,12], our data for the transition fields, and the deductions of Section 5, we construct the following table:
Use of the experimental data of Gorodetsky and others[9] for $YFeO_3$ gives the same values of θ_ω^{calc}.

There can be no doubt that in $YFeO_3$ at both temperatures, what occurs at field $H = H_t$ is a phase transition of the second kind (at $\theta = 0$, of course). The availability of sufficiently accurate static data enables us not only to calculate the limiting angle θ_ω (by formula (16d)), but also to construct, for fixed operating frequency ω, the whole curve of the orientational dependence $\theta(h)$ of the resonance fields; this is easily obtained by eliminating the variable φ from equations (19a, b) for P = Q = 0. This construction has been made in Fig. 3. The dotted part of the curves corresponds to values of φ for which the condition $\varphi \ll 1$ is poorly satisfied, and therefore neglect of the terms containing P and Q in (19a, b) is not legitimate.

The failure to resolve the absorption line when $\lambda \sim 4$, 6, and 8 mm in $YFeO_3$, even when the field was oriented along the axis [a] with the greatest accuracy possible in our apparatus ($\theta \leq 10'$), is easily explained by the strong frequency dependence of the value of the splitting Δh and of the limiting angle θ_ω ($\Delta h \equiv h_2 - h_1 \sim \omega^2$; $\theta_\omega \sim \omega^3$—see (16a, c, d)) and by the relatively low sharpness of the resonance lines of the specimens investigated. For $SmFeO_3$, a quantitative interpretation of the orientation curve is very difficult. The absence of sufficiently accurate static data makes it impossible to estimate the basic parameter of the problem, Λ_{exp}, with reasonable accuracy. An attempt to calculate Λ_{exp} (see Table) from the data of Belov and collaborators[12] leads to a relatively small value for it. Expression (16d)—in which, for application to the case of $SmFeO_3$ at T = 300°K, it is sufficient to replace H_a by H_c and Λ_a by Λ_c—shows that the value of the limiting angle θ_ω increases with decrease of Λ_c. Therefore the significantly different character of the orientation curve in $SmFeO_3$ as compared with $YFeO_3$ (see Table) agrees qualitatively with theory.

7. PHASE TRANSITION WITH RESPECT TO FIELD IN $SmFeO_3$

In samarium orthoferrite at H = 0 and at temperatures from 0°K to the reorientation range ($T_R \approx 480°K$), the configuration l ∥ [c], m ∥ [a] exists. In a field H ∥ [c], there is completed at $H = H_t$ a "quasiflop of the sublattices", that is a phase transition xz → x. The value of H_t, as follows from the experiments of Belov and collaborators[12], in small static fields depends strongly on temperature. It seemed of interest to follow the dependence $H_t(T)$ to lower temperatures. We did

Crystal	T, °K	cgs emu/mole $10^4\chi_m$	cgs emu/mole $10^4\chi_l$	cgs emu/mole σ_0	H_t, kOe	Λ_{exp}, kOe²	Kind of transition	θ_ω at ν = 128 GHz Theory	θ_ω at ν = 128 GHz Expt.
$YFeO_3$	300	23.2	34	232	70±2.5	8600(±10%)	Second	1.3°	1.5°
	4.2	21.8	49	305	72.5±1	11800(±10%)	Second	0.85°	—
$SmFeO_3$	300	21.1	25	262	50±2	~0±800	Second	—	>4°

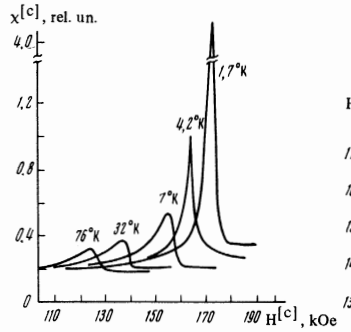

FIG. 4

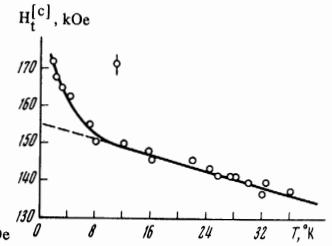

FIG. 5

FIG. 4. The results of induction measurements of the field and temperature dependence of the differential susceptibility of $SmFeO_3$ point to a change of character of the phase transition with lowering of the temperature.

FIG. 5. Temperature dependence of the value $H_t^{[c]}$ of the xz → x phase-transition field in $SmFeO_3$, according to measurements of the differential susceptibility. There is a similar temperature variation of the value of the AFMR field for ν = 37 GHz and **H** ∥ [c].

this on the monocrystalline specimen of $SmFeO_3$ described above: at various temperatures from 1.7 to 300°K, we measured by the induction method the dependence of the differential susceptibility along the axis [c] on the value of the pulsed field.

The results of analysis of the oscillograms are shown for several temperatures in Fig. 4. Besides the expected increase of H_t with lowering of temperature, one notices the abrupt change of character of the behavior of $\chi^{[c]}(H)$ near H_t at the lowest temperatures. The increase in the height of the peak in $\chi^{[c]}$ and its abrupt contraction permit us to state that the phase transition xz → x at field H_t changes from continuous to discontinuous with respect to the magnetic moment. The fact that a similar transformation of the character of the transition occurs on approach to the "compensation point" of $SmFeO_3$ (see[23]; $T_{comp} \approx 4.2$°K) is not surprising. At the compensation point, the assumed absence of a spontaneous magnetic moment makes the phase transition under discussion similar to the classical flop of sublattices in pure antiferromagnets, which is known to be a transition of the first kind. The transition must remain discontinuous also in a certain interval above T_{comp}, until at a certain $T = T_b$ the jump in the magnetic moment at field H_t decreases to zero. Thus this temperature T_b separates the regions of phase transitions of the first and of the second kinds with respect to the field. Right at $T = T_b$, the transition with respect to the field may possess the features of the phase transition of the "one-and-one-halfth kind" discussed in Sec. 5. The form of the temperature dependence $H_t(T)$ for $SmFeO_3$, shown in Fig. 5, is still another argument in favor of this deduction, since at some temperature near 4.2°K the value of d^2H_t/dT^2, in accordance with the prediction of the theory, undergoes a discontinuity (or at least changes abruptly). For determination of the specific value of T_b and, in particular, of its difference from T_{comp}, and also for quantitative analysis of the phenomenon (calculation of Λ and of the jump in d^2H_t/dT^2 at $T = T_b$), we are undertaking to improve appreciably the accuracy of the measurements.

8. CONCLUSIONS

Phenomenological calculation of the statics and dynamics of orthorhombic antiferromagnets, with interactions subject to the conditions $|A|, D \ll E$, with allowance for bilinear and biquadratic anisotropy in a form corresponding to the requirements of the crystal symmetry, enables us to obtain convenient expressions for qualitative and quantative analysis of the properties of antiferromagnets of this class. For a complete quantitative description of the statics of these materials, two combinations (A_a and A_c) of the five anisotropy constants involved are sufficient; but description of the linear dynamic properties requires introduction of two additional combinations (A_x and A_z). The kind of magnetic phase transition (with respect to the field) that occurs at a certain value of the magnetic field, parallel to the axis that is easy for the AF-vector ([a] or [c] outside the reorientation region), is determined by the sign of the parameter Λ_{exp}, which is made up of experimentally measured magnetic characteristics of the crystal (see (18)): for $\Lambda_{exp} > 0$, the transition is of the second kind; for $\Lambda_{exp} < 0$, it is of the first kind. In the case $\Lambda > 0$, the orientational dependence of the resonance fields near the isolated point of phase transition of the second kind in the (H_x, H_z) plane can be described by relatively simple expressions, which are convenient for predicting the results of dynamic measurements for most orthoferrites.

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