Effect of anisotropy on field-induced antiferromagnetism in $YFeO_3$

V. I. Ozhogin, V. M. Cherepanov, and S. S.Yakimov

I. V. Kurchatov Institute of Atomic Energy

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The change of the antiferromagnetic order in single-crystal $YFeO_3$ is studied by means of the Mössbauer effect at temperatures near T_N and in magnetic fields between 0 and 30 kOe oriented along the *a* and *c* axes of the crystal. The obtained critical exponents ($\beta=0.36\pm0.1$, $\delta=4.0\pm0.4$, $\gamma\approx\gamma'=1.0\pm0.2$) satisfy the main relation of the scaling hypothesis. Antiferromagnetic order is induced at $T\sim T_N$ in different manners at $H\|a$ and $H\|c$, as predicted by the phenomenological theory with allowance for the anisotropy in the (*a c*) plane.

INTRODUCTION

Yttrium orthoferrite $YFeO_3$ is one of the simplest among the antiferromagnetic (AF) perovskites ABO_3[1], since Y^{3+} is a diamagnetic ion, and the Fe^{3+} ion is in the S state. $YFeO_3$ has been investigated in considerable detail experimentally both with the aid of magnetic[2,3] and neutron-diffraction[4] measurements, as well as by the Mössbauer-effect method[4,5] in a zero field.

Owing to the Dzyaloshinskiĭ interaction, in antiferromagnetic with weak ferromagnetism (AWF), which include $YFeO_3$, an external magnetic field **H** of definite orientation should induce antiferromagnetic order in the critical temperature region ($T\sim T_N$)[6,7]. An investigation of the effect of induction in $YFeO_3$ was of interest also because in the (ac) plane of this crystal (D_{2h}^{16} symmetry) the two principal directions connected with the Dzyaloshinskiĭ interaction are not equivalent from the point of view of anisotropy for the AF vector **l**: the a axis is "easier" than the c axis (but both are very much more easier for **l** than the b axis).

A convenient method of directly observing the induction effect is the use of the Mössbauer effect, as was demonstrated with the weak-magnetism antiferromagnets FeF_3[8] and $FeBO_3$[9]. Measurement with the aid of the Mössbauer effect of the hyperfine field $\mathbf{H}_n$ at the nuclei Fe^{57}, with allowance for the proportionality of this field to the average magnetic moment **M** of the Fe^{3+} ion[4], makes it also possible to investigate the critical behavior of the AWF, and this is of interest from the point of view of the similarity hypothesis[10]. In this case the $H_n(T, H)$ dependence[11] yields more direct information on the relative magnetization of the sublattice than magnetic measurements, inasmuch as the usual relation $\mathbf{l} > \mathbf{m}$ for AWF with large exchange interaction (**m** is the ferromagnetic vector), we have the equality $H_n(T, H)/H_n(0, 0) \equiv h_n = M/M_0 \approx l$ (see[9]).

PHENOMENOLOGICAL THEORY

As already noted, the b axis in $YFeO_3$ is difficult for the AF vector, i.e., the equilibrium magnetizations $\mathbf{M}_i$ (i = 1 and 2 in a two-sublattice model) do not go out from the (ac) plane at $H_b = 0$. We shall therefore describe the critical behavior by four variables (see Fig. 1a; a, b, c → x, y, z); namely, the x and z components of the vectors $\mathbf{m} = (\mathbf{M}_1 + \mathbf{M}_2)/2M_0$ and $\mathbf{l} = (\mathbf{M}_1 - \mathbf{M}_2)/2M_0$; m and *l* are much less than unity near T_N.

The Dzyaloshinskiĭ interaction in orthoferrites can be determined in accordance with their symmetry[12],

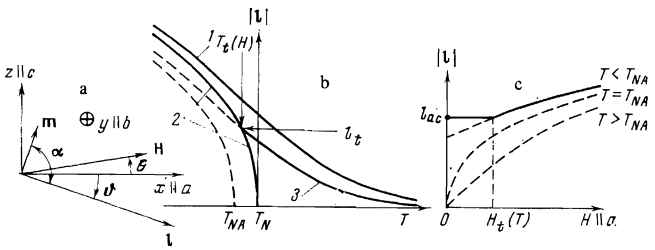

FIG. 1. Illustrating the analysis of the influence of anisotropy on the induction effect in yttrium orthoferrite. a) Variables of the problem. b) Calculated dependence of the AF vector **l** on T: 1) a-phase, **H** || c; 2) a-phase at H = 0 and ac-phase at **H** || a; 3) c-phase, **H** || a. c) Calculated l(H) dependence.

either by the antisymmetrical exchange (**D** || b) and by the single-ion anisotropy A_{12}. Since, however, $D \gg A_{12}$, as shown by low-temperature measurements of the magnetization (for $YFeO_3$ see[3,13]), we neglect A_{12} from now on. Then the thermodynamic potential takes the form

$$\Phi=2M_0\{{}^1/_2E_0l^2+{}^1/_2E_1m^2-D(m_zl_x-m_xl_z) +{}^1/_2Al_z^2+{}^1/_4E_2l^4+{}^1/_4Fm^4+{}^1/_4G(\mathbf{ml})^2-\mathbf{mH}\}. \quad (1)$$

The biquadratic anisotropy, which is important in strong fields at $T \ll T_N$ (when l_z can be of the order of unity), is disregarded here; we neglect the exchange term $Fm^4/4$, putting $m \ll l$. Among the coefficients of the potential, only the quantity E_0 should be regarded as noticeably dependent on the temperature at $T \sim T_N$:

$$E_0-D^2/E_1=\nu(T-T_N)=\nu\Delta T, \quad \nu>0, \quad (2)$$

the remaining coefficients (E_1, E_2, D, A, G) can be regarded as independent of T (and positive).

Minimization of (1) with respect to the variables m_x, m_z, l_x, and l_z leads to the following results:

At **H** || c we have at thermodynamic equilibrium **m** || **H** and **l** || a, therefore $\alpha = \pi/2$ and $\vartheta = 0$ (a-phase). The solution is identical with that of the case of AF with easy-plane anisotropy[6,9]:

$$E_2l_x^3+\nu\Delta Tl_x=DH/E_1, \quad l_z=0; \quad (3a)$$

$$m_z=(Dl_x+H)/E_1, \quad m_x=0. \quad (3b)$$

At **H** || a, in the general case, the balance equations are too complicated. For the sake of simplicity we assume that G is so large that $\alpha = \pi/2$ ($\mathbf{m}\cdot\mathbf{l} = 0$). We then obtain ($l^2 - l_x^2 + l_z^2$):

$$(E_2l^2+\nu\Delta T)l_x=0, \quad (E_2l^2+\nu\Delta T+A)l_z=-DH/E_1, \quad (4a)$$

$$m_x=(-Dl_z+H)/E_1, \quad m_z=Dl_x/E_1. \tag{4b}$$

The magnetization $YFeO_3$ at $T \sim T_N$ for $\mathbf{H} \parallel c$ and $\mathbf{H} \parallel a$ was investigated in detail by Obayashi[2]. However, in the analysis of the data for $\mathbf{H} \parallel a$, the differentiation of Φ was carried out only with respect to the variables m_x and l_z. Physically this signifies the assumption that the induced $H_x \to m_x \to l_z$ occurs independently of whether the spontaneous components m_z and l_x are present or not. This is in fact not so, since the exchange term $E_2 l^4/4 \equiv E^2(l_x^4 + l_z^4 + 2l_x^2 l_z^2)/4$ makes both weakly ferromagnetic (WF) configurations dependent on each other (see Eqs. (4a)).

Equations (4) admit of two solutions:

1) At equilibrium $\mathbf{l} \parallel c$, $\mathbf{m} \parallel a$ ("c-phase"), i.e.,

$$m_z=l_x=0, \quad l_z=-l_c, \quad E_2 l_c^3+(\nu\Delta T+A)l_c=DH/E_1, \quad m_x=(Dl_c+H)/E_1. \tag{5}$$

the equilibrium value of Φ is

$$\Phi_c=\frac{l_c^2}{2}\left[E_0+A+\frac{E_2}{2}l_c^2-\frac{E_1}{D^2}(E_2 l_c^2+E_0+A)^2\right].$$

2) At equilibrium $\mathbf{l}$ and $\mathbf{m}$ have both a and c components ("ac-phase")

$$l^2 \equiv l_0^2=-\frac{\nu}{E_2}\Delta T, \quad l_z=-\frac{DH}{AE_1}, \quad l_x=(l_0^2-l_z^2)^{1/2}; \tag{6a}$$

$$m_x=\frac{H(1+D^2/AE_1)}{E_1}, \quad m_z=\frac{Dl_x}{E_1}, \tag{6b}$$

$$\Phi_{ac}=-\frac{\nu^2(\Delta T)^2}{4E_2}-\frac{H^2(1+D^2/AE_1)}{2E_1}. \tag{6c}$$

The transition between the phases occurs at $\Phi_c = \Phi_{ac}$ (in this case $l_x^{(ac)} = 0$). It follows from (6a) that the relation

$$-\nu\Delta T/E_2=(DH/AE_1)^2$$

is satisfied at the transition point. This relation is the equation of the phase boundary $H_t(T_t)$ on the (H_a, T) plane[1)]. At this boundary we have

$$T_t=T_N\left(1-\frac{E_2D^2H_t^2}{\nu T_N A^2 E_1^2}\right), \quad l_t=\left[-\frac{\nu}{E_2}(T_t-T_N)\right]^{1/2}=\frac{DH_t}{AE_1}, \quad m_t=\frac{H_t(1+D/AE_1)}{E_1}=\left(\frac{A}{D}+\frac{D}{E_1}\right)\left[-\frac{\nu}{E_2}(T_t-T_N)\right]^{1/2}. \tag{7}$$

We can now construct with the aid of (6) and (7) a qualitative picture of the induced antiferromagnetism at $\mathbf{H} \parallel a$—see Fig. 1b; the ac-phase is weak-field and low-temperature, while the c-phase is strong-field and high-temperature (more symmetrical). The transition between them is a second-order phase transition. Equations (5) show that at $\mathbf{H} \parallel a$ the antiferromagnetic order is induced by the method described for the a-phase (for $\mathbf{H} \parallel c$ see (3a)), but the role of the Neél temperature is assumed in this case by the quantity $T_{NA} \equiv T_N(1 - A/\nu T_N)$ (see the dashed lines in Fig. 1b). For $\mathbf{H} \parallel a$ at $T > T_N$, the c-phase is always energywise favored. At $T < T_N$ and $H_a < H_{at}(T)$, owing to the anisotropy energy, the ac-phase is realized. Near T_N, the phase boundary $H_{at}(T)$ is a parabola (within the framework of the Landau-theory approximations). For the entire interval from 0 to T_N, the $H_{at}(T)$ dependence can be plotted, for example, with the aid of the molecular-field theory using the known value $H_t(0°K) = 74$ kOe.[3]

The foregoing calculation is valid only if $m \ll l$. In the ac-phase, it is satisfied practically always, and at $T > T_N$ this calls for $T - T_N \ll D/\nu$ (we use formulas (3) and the relations $D \ll E_1$ and $l \ll 1$).

EXPERIMENT. DISCUSSION OF RESULTS

The measurements of the Mössbauer effect on the Fe^{57} nuclei in single-crystal $YFeO_3$ were made in the temperature interval 300—670°K with a one-channel γ spectrometer operating at a constant velocity of the source (Co^{57} in Cr). The samples, with the a or c axis located in the polished plane, were plates measuring $1 \times 1 \times 0.01$ cm and were placed in an oven secured between the poles of an electromagnet. An automatic regulator maintained the temperature of the investigated sample constant during the measurement process with accuracy not worse than 0.1°C. The temperature drop across the sample was not larger than 0.2°C. The magnetic field perpendicular to the γ quanta was applied in the plane of the sample along one of the crystal axes, a or c. The directions of the crystallographic axes of the sample were determined by x-ray diffraction. The sample was placed in a magnetic field relative to the outer faces of the crystal with accuracy 1°—3°. The magnetic transformation temperature T_N was determined with the aid of the Mössbauer effect from the dependence of the γ-quantum counting rate on the temperature at a fixed source velocity (the thermal-scanning method). It was established that T_N is the same for both investigated samples and equal to 644.0 ± 0.1°K.

Figure 2 shows typical Mössbauer absorption spectra of Fe^{57} nuclei in $YFeO_3$ samples at $T \sim T_N$. The solid curves show the calculated spectra obtained by computer reduction of the experimental data by least squares in the approximation in which the line shape is assumed to be Lorentzian. An analysis of the obtained spectra shows that in $YFeO_3$ there is indeed realized the above-described qualitative picture of its magnetic behavior. Recognizing that the direction of the field $\mathbf{H}_{n1,2}$ is antiparallel to the magnetization $\mathbf{M}_{1,2}$ of the sublattices of the orthoferrite (i.e., practically coincides with 1, if we neglect small ($\psi \sim 0.5°$) canting of the magnetic moments of the sublattices as a result of the Dzyaloshinskiĭ interaction), we can explain the obtained spectra in the following manner:

In the case of a γ-quantum beam direction $\mathbf{k}_\gamma \parallel a$ (Fig. 2a), at $T < T_N$ and $H = 0$ the ratio of the Mössbauer-spectrum components indicates that $\mathbf{l} \parallel a$

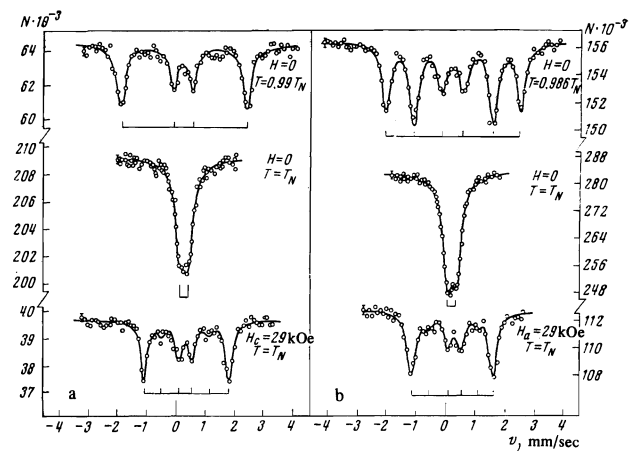

FIG. 2. Influence of external magnetic fields on the Mössbauer absorption spectra of Fe^{57} nuclei in $YFeO_3$ at $T \sim T_N$: a—direction of γ-quantum bean $\mathbf{k}_\gamma \parallel a$, b—$\mathbf{k}_\gamma \parallel c$.

(Fig. 3a). At $T = T_N$ and at $\mathbf{H} \parallel c$ ($\mathbf{H}_c$) the spectrum remains practically unchanged, i.e., the induced magnetic structure is that possessed by $YFeO_3$ at $T < T_N$ (a-phase) (Fig. 3b). On the other hand, in the case $\mathbf{k}_\gamma \parallel c$ (Fig. 2b) at $T < T_N$ and $H = 0$, a spectrum is observed with an intensity ratio 3 : 4 for the two external components. This means that $\mathbf{H}_n \perp \mathbf{k}_\gamma$, and consequently, $\mathbf{l} \parallel a$ (Fig. 3c). At $T = T_N$ and $\mathbf{H} \parallel a$ ($\mathbf{H}_a$) we obtain the same kind of spectrum as at $\mathbf{H}_c$, but with a smaller value of H_n. Since now $\mathbf{H}_n \parallel \mathbf{k}_\gamma$ (i.e., $\mathbf{l} \parallel c$), we conclude that in this case another AWF magnetic-structure configuration that is possible for $YFeO_3$ (the c-phase) is induced.

Finally, in the case when $\mathbf{k}_\gamma \parallel c$, at $T < T_N$ and $0 < H_a < 30$ kOe (Fig. 4), the Fe^{57} nuclei in the $YFeO_3$ sublattices are acted upon by different effective magnetic fields $\mathbf{H}_1$ and $\mathbf{H}_2$. This fact offers evidence that the weak-ferromagnetism configuration of the magnetic structure is such that the vector l occupies an intermediate position between the axes a and c of the crystal (ac-phase). At all values of H, the values of the hyperfine fields H_{n1} and H_{n2} for both sublattices, which can be obtained from the formula (see Fig. 3d)

$$H_{n\,1,\,2}=[(H_1^2+H_2^2)/2-H^2]^{1/2},$$

remain in this case unchanged and equal to $H_{n1} = H_{n2} \approx 133$ kOe (at $T = 637.7°K$). At the same time, the splitting of the resultant Mössbauer spectrum into two Zeeman sextets and the change of the ratio of the intensities of its components with increasing field H indicate that in this case l has rotated from the a axis to the c axis. Thus, in the ac-phase, the field $\mathbf{H}_a$ causes only a rotation of the antiferromagnetic vector, without changing its absolute magnitude.

Figure 5 shows plots of $H_n(T)$ and $H_n(H)$ for both investigated samples at H = 29 kOe. In the entire investigated temperature interval $0.98 \lesssim T/T_N \lesssim 1.02$ we obtain $H_n(T, H_c) > H_n(T, H_a)$. This result agrees with the theoretically-concluded shift of the effective temperature T_{NA} of the magnetic transformation for the c-phase for the region of lower temperatures (see Fig. 1b).

The obtained experimental data make it possible, within the framework of the thermodynamic theory described above (see Eqs. (3) and (4)), to estimate (with allowance for the values $H_n(T = 0°K) \approx 550$ kOe[5] and $D/E_1 \approx 1.1 \times 10^{-2}$[2,3]) the constants E_2, A, and ν:

$$E_2 \approx 60 \text{ kOe}, \quad A \approx 0.6 \text{ kOe}, \quad \nu T_N \approx 600 \text{ kOe}.$$

The value of the anisotropy constant A agrees satisfactorily with the value 0.68 kOe obtained from magnetic measurements[3].

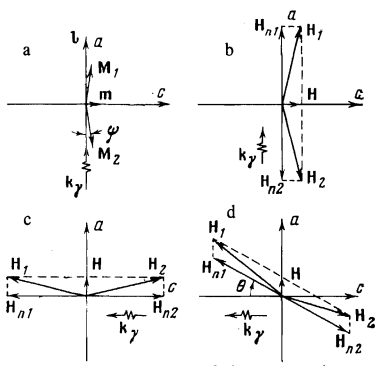

FIG. 3. Schematic representation of the magnetic structure (a) and of the field directions (b–d) at the Fe^{57} nuclei in the $YFeO_3$ sublattices at the different directions of H.

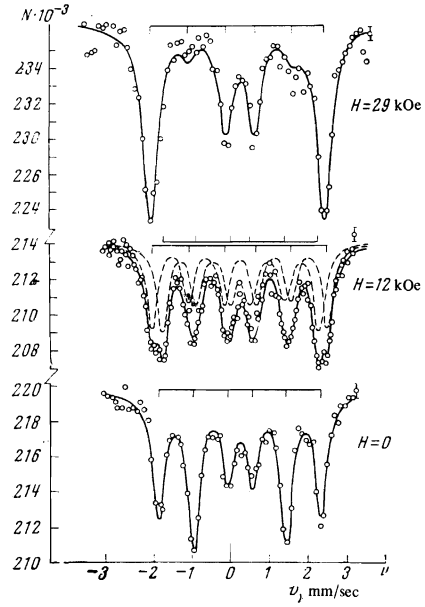

FIG. 4. Change in the form of the Mössbauer spectra at $T < T_N$ with changing field; $T = 0.99T_N$, $\mathbf{k}_\gamma \parallel c$, $\mathbf{H} \parallel a$.

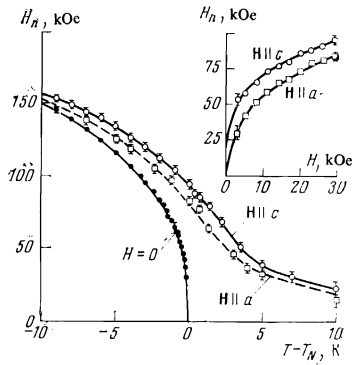

FIG. 5. Dependence of H_n on T and H.

As to the critical exponents, their values $\beta = 0.36 \pm 0.01$ at $0.9 \lesssim T/T_N \lesssim 0.9995$ and $\delta = 4.0 \pm 0.4$ at $3 < H < 30$ kOe, obtained by a power-law approximation of the experimental data for $h_{n,H} = H_n(T, H)/H_n(0, 0)$ (see Fig. 6a, b), in the form

$$h_{n,\,0} \propto (T_N - T)^{\beta}, \quad h_{n,\,H}(T = T_N) \propto H^{1/\delta},$$

differ significantly from the values $\beta = 1/2$ and $\delta = 3$ given by the thermodynamic theory.

We have analyzed our data within the framework of another phenomenological approach—the similarity hypothesis[10], in which the critical exponents satisfy relations such that only any two of the power exponents are independent, and the remainder are expressed in terms of these exponents. In our case the satisfaction of the similarity conditions can be verified by comparing the calculated and measured values of the critical exponents γ and γ', determined from the equations

$$\chi_l^{\perp}(T - T_N)_{H \to 0} \propto (T - T_N)^{-\gamma} \quad T > T_N, \tag{8a}$$

$$\chi_l^{\perp}(T - T_N)_{H \to 0} \propto (T_N - T)^{-\gamma'} \quad T < T_N. \tag{8b}$$

Here $\chi_l^{\perp} \equiv \partial l/\partial H|_{H \to 0}$ is the initial susceptibility of the AWF at $\mathbf{H} \perp \mathbf{l}$.

For an experimental determination of γ and γ' we

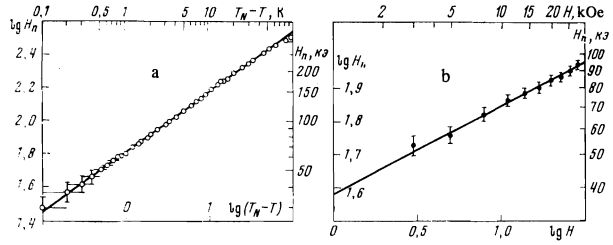

FIG. 6. For use in the determination of the critical exponents β and δ in $YFeO_3$: a) $H_n = A(T_N-T)^{\beta}$, $\beta = 0.36 \pm 0.01$ ($T < T_N$); b) $H_n = BH^{1/\delta}$, $\delta = 4.0 \pm 0.4$ ($T = T_N$, $H \parallel c$).

can use the fact that at a certain distance $\Delta T \sim 10^{-3}T_N$ from T_N, the quantity $l(T, H)$ is practically linearly dependent on H, while the effective values of the exponents γ and γ' change insignificantly. The obtained values $\gamma \approx \gamma' = 1.0 \pm 0.2$ agree well with the values of β and δ and satisfy the similarity-hypothesis condition $\gamma, \gamma' = \beta(\delta - 1)$.

CONCLUSION

The results show that in spite of its limited character, primarily with respect to the quantitative prediction of the critical exponents, the phenomenological theory of phase transitions, based on Landau's work, yields satisfactory description of the induction of the antiferromagnetism by a field not only in the simplest AWF such as $FeBO_3$ [8], but also when account is taken of the crystallographic anisotropy. It is therefore of interest to carry out analogous investigations (including those aimed at measuring the Dzyaloshinskiĭ field H_D) of the induction effect in AWF with qualitative and quantitative relations between the exchange, Dzyaloshinskiĭ interaction, and the anisotropy other than those of $YFeO_3$, and in particular investigations of $FeCO_3$ at $H \perp C_3$ and FeF_2 and $H \perp C_4$.

We are grateful to K. K. Kikoin for constant interest in the work and to L. N. Dem'yanets and L. V. Usov for preparing the $YFeO_3$ single crystals.

[1)]The phase boundary $H_t(T_t)$ for easy-axis antiferromagnets was investigated at $T \approx T_N$ in [14] within the framework of the Landau theory.

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