Observation of a multicritical singular point on the phase diagram in non-coplanar spin reorientation of $DyFeO_3$

A. M. Balbashov, P. Yu. Marchukov, I. V. Nikolaev, and E. G. Rudashevskiĭ

General Physics Institute, USSR Academy of Sciences

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The compound $DyFeO_3$ undergoes a non-coplanar spin reorientation induced by a magnetic field $\mathbf{H}\|\mathbf{a}$; based on the results of our investigations of the AFMR spectrum of $DyFeO_3$ during this transition, we construct a phase diagram with a tetracritical singular point.

The compound $DyFeO_3$ (dysprosium orthoferrite), a rhombohedral crystal, is known to undergo various types of spin reorientation (SR) when a magnetic field is applied along its **a** axis.[1] For $T > T_M \sim 50$ K (where T_M is the temperature of the Morin transition) a rotation of the antiferromagnetism vector **L** takes place in the crystal's **ac** plane from the phase $\Gamma_4(M_z, L_x)$ into the phase $\Gamma_2(M_x, L_z)$, while for $T < T_M$ this vector rotates in the **bc** plane: $\Gamma_1(L_y) \leftrightarrow \Gamma_{12}(M_x, L_y, L_z) \leftrightarrow \Gamma_2(M_x, L_z)$ (we use the standard notation for the orthoferrite magnetic configurations[1]).

The SRs $\Gamma_4 \leftrightarrow \Gamma_2$ and $\Gamma_1 \leftrightarrow \Gamma_2$ in fields $H_{\rm tr}^{ac}$ and H_{r}^{bc}, respectively, are second-order phase transitions, up until recently, these SRs were studied as separate and independent phenomena,[2,3] although it was shown long ago[4] that in principle a transition $\Gamma_{42} \leftrightarrow \Gamma_{12}$ could be induced by a field $\mathbf{H}\|\mathbf{a}$ from the initial state Γ_4 to canted phases for SR processes in orthoferrites. Based on the available experimental data for $DyFeO_3$ (Refs. 6 and 7), the authors of Ref. 5 predicted that the SR $\Gamma_4 \leftrightarrow \Gamma_{42} \leftrightarrow \Gamma_{12} \leftrightarrow \Gamma_2$ induced by a field $\mathbf{H}\|\mathbf{a}$ can be described in the $(H - T)$ plane by a phase diagram with a multicritical singular point, and that this SR should take place in $DyFeO_3$ in the temperature range $T_M < T < T^*$, where $T^* \sim 60$ K; these authors also calculated the stability conditions for the various magnetic configurations, and described the behavior of the AFMR spectrum. By now the SR predicted in Refs. 4 and 5 was indeed observed in $DyFeO_3$, first by magnetooptic methods[8] and then with AFMR studies.[9]

In this paper we report on our own AFMR investigations of $DyFeO_3$ in the region of the noncoplanar SR $\Gamma_{42} \leftrightarrow \Gamma_{12}$ induced by a field $\mathbf{H}\|\mathbf{a}$, and construct a phase diagram with a multicritical singular point.

Our experiments were carried out using a direct-amplification spectrometer to record sample absorption lines at specified radiation frequencies as a function of magnetic field at constant temperature. Our method of measurement is described in Ref. 10, along with the precise setup with regard to sample orientation in the field.

In Figure 1 we show the AFMR spectrum at various temperatures for fields near the field at which the SR occurs. The two branches we observed, $\nu_1(H)$ and $\nu_2(H)$, correspond to the quasiferromagnetic and quasi-antiferromagnetic modes of the AFMR. It is quite clear (Figs. 1a and 1b) that as the temperature decreases the frequency ν_2 in the field at which the SF occurs decreases, reducing eventually to zero. Corresponding to symmetric vibrations of the quasi-antiferromagnetic mode,[11] this attests to a loss of stability of the original magnetic configuration relative to the motion of the vector **L** out of the plane **ac**. Calculations show[5] that the condition for loss of stability of the phase Γ_{42} during the SR $\Gamma_4 \leftrightarrow \Gamma_{42} \leftrightarrow \Gamma_2$ in a field $\mathbf{H}\|\mathbf{a}$ coincides with the expression for $\nu_2^2(H)$ at $H = H_{\rm tr}^{ac}$.

Thus, the SR $\Gamma_{42} \leftrightarrow \Gamma_{12}$ takes place also in the case where $\nu_2(H)$ reduces to zero before the SR $\Gamma_4 \leftrightarrow \Gamma_2$ has occurred. This situation is illustrated in Fig. 1c: the light circles denote the transition field $H_{\rm tr}^{(1)}$ at which a significant intensity change (a "step") appeared in the trace of the absorption line for the signal passing through the sample. Because the conditions for loss of stability of the phases Γ_{42} and Γ_{12} do not coincide:[5] the SR $\Gamma_{42} \leftrightarrow \Gamma_{12}$ is a first-order phase transition, and therefore it takes place earlier than the reduction of ν_2 to zero. However, we were unable to observe any hysteresis phenomena, and the width of the field step did not exceed 3 kOe.

The phase diagram is shown in Fig. 2. For $T > T^* = 66$ K, the SR in a field $\mathbf{H}\|\mathbf{a}$ is of the usual form: $\Gamma_4 \leftrightarrow \Gamma_{42} \leftrightarrow \Gamma_2$, while the curve AC is a function of $H_{\rm tr}^{ac}(T)$. In the temperature region $T_M < T < T^*$ the SR begins within the **ac** plane, and then at a field $H_{\rm tr}^{(1)}$ the vector **L** passes discontinuously

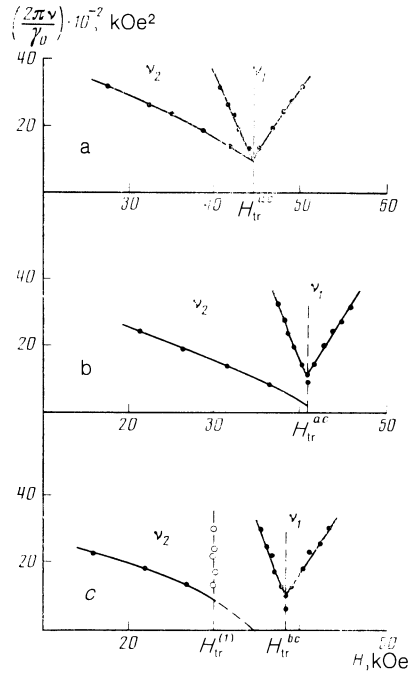

FIG. 1. Dependence of the AFMR frequency on the magnetic field in the vicinity of fields at which SRs occur, for various temperatures: a—74 K, b—66 K, c—60 K (γ_0 is the gyromagnetic ratio).

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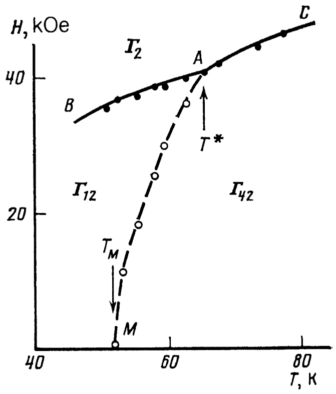

FIG. 2. Phase diagram of $DyFeO_3$ (explanation in text).

from the state Γ_{42} to the state Γ_{12} and the SR takes place in the plane **bc**. The curves MA and BA on the phase diagram are functions of $H_{tr}^{(1)}(T)$ and $H_{tr}^{ac}(T)$. Thus, in this temperature range the SR in a field **H**||**a** has the form $\Gamma_4 \leftrightarrow \Gamma_{42} \leftrightarrow \Gamma_{12} \leftrightarrow \Gamma_2$ and constitutes a combination of two phase transitions, one first-order ($\Gamma_{42} \leftrightarrow \Gamma_{12}$) and one second-order ($\Gamma_{12} \leftrightarrow \Gamma_2$).

For $T < T_M$ (where $T_M = 52$ K was determined from the step in the trace of the absorption line as a function of temperature in the absence of a field, and coincides with the data from Ref. 8) the SR takes place wholly in the crystal's **bc** plane.

At $T = T^*$ (the point A on Fig. 2) the loss of stability relative to the departure of the vector **L** from the plane **ac** occurs at the instant that the phase Γ_2 becomes stable (which is indicated by the reduction of ν_2 to zero, (see Fig. 1b), in which $H_{tr}^{ac} = H_{tr}^{bc} = H_{tr}^{(1)}$. Thus, according to Griffith's classification,[12] the point A on the phase diagram is a three-phase tetracritical singular point, because three phases Γ_{42}, Γ_{12} and Γ_2 are adjacent at this point and four curves of stability-loss converge there (the lines $H_{tr}^{ac}(T)$ and $H_{tr}^{bc}(T)$, and the two loss-of-stability lines for the Γ_{42} and Γ_{12} phases during the phase transition $\Gamma_{42} \leftrightarrow \Gamma_{12}$ which are not shown in Fig. 2). We should point out that the functions $H_{tr}^{ac}(T)$ and $H_{tr}^{bc}(T)$ have previously been measured in the compound $DyFe_{1-x}Al_xO_3$.[13] However, in the phase diagram constructed in Ref. 13, the region MAB (Fig. 2) is shrunk to a vertical line (i.e., $T_M = T^*$), from which it follows that the SR $\Gamma_{42} \leftrightarrow \Gamma_{12}$, which was not observed in Ref. 13, can be induced only by changing the temperature in nonzero magnetic fields.

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