Magnetic resonance in the intermediate state of $ErFeO_3$

N. K. Dan'shin, N. M. Kovtun, and M. A. Sdvizhkov

Donetsk Physicotechnical Institute, Academy of Sciences of the Ukrainian SSR
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Magnetic resonance is studied experimentally for the intermediate state of erbium orthoferrite during the metamagnetic transition in the Er spin subsystem with simultaneous spin flip in iron. The form of the frequency-field dependences of the magnetic resonance (MR) is shown to depend on the nature of the phase transition—for temperatures below the tricritical point the MR frequency changes abruptly at the phase boundary. There is a range of fields for which resonance occurs in the intermediate state. Oscillations due to the antiferromagnetic and ferromagnetic phases are found to coexist in the intermediate state and correspond to different frequencies which are independent of the external field; moreover, the resonance amplitude for each of the phases depends on their specific volume and also on the field.

INTRODUCTION

The nature of the antiferromagnetic resonance (AFMR) in the intermediate state in antiferromagnetics during the spin-flip transition has recently been studied experimentally.[1–3] According to the theory in Ref. 4, the intermediate state is a thermodynamically stable periodic structure consisting of domains of two phases, between which a phase transition of the first kind occurs. The intermediate state has the distinctive property that the internal magnetic field H_i is independent of the external field H. This has the consequence that the AFMR frequencies in the intermediate state are independent of H if the oscillating magnetic moments in the coexisting domains do not interact dynamically. The intensity of the resonant absorption signal from each phase should be proportional to the relative amount of the phases. The AFMR signal amplitude excited in domains consisting of the starting phase should therefore decrease as H increases, while the signal from the second (growing) phase should become stronger. This behavior was observed experimentally in Ref. 2 for MnF_2 and also in Ref. 3, where the high-frequency branch of the AFMR was studied for $CuCl_2 \cdot 2H_2O$.

In this paper we study magnetic resonance in the intermediate state of erbium orthoferrite $ErFeO_3$, which contains two spin subsystems (Er and Fe). For $T < 636$ K the Fe^{3+} spins from a skew antiferromagnetic (AFM) structure, while the Er^{3+} spins remain paramagnetic down to liquid helium temperatures. For $T \sim 4$ K the Er^{3+} spins assume an AFM ordering along the c crystallographic axis, and this transition induces a spin flip in the Fe^{3+} subsystem.[5] Because of the large anisotropy per Er^{3+} ion, the rate-earth subsystem has metamagnetic properties[6] and can be regarded as a metamagnetic with AFM vector along the c axis. The metamagnetic properties determine the form of the $ErFeO_3$ phase diagram for fields **H** parallel to **c** (Refs. 7, 8). The phase diagram implies that the nature of the phase transition in the metamagnetic depends on the temperature. For T above the critical point T_c, the field-induced spin transition from the AFM to the ferromagnetic (FM) state occurs via a phase transition of the second kind, whereas a first-order transition occurs for $T < T_c$. In the latter case the phase transition for samples of finite size proceeds through an intermediate state.[8,9] The intermediate state in $ErFeO_3$ thus forms when the Er^{3+} ions undergo a metamagnetic AFM→FM transition, which is accompanied by flipping of the Fe^{3+} spins.

We stress that as far as the experimental observation of magnetic resonance in the intermediate state is concerned, metamagnetics are more convenient than antiferromagnetics which have a spin-flip transition but low anisotropy. This is because the intermediate states in metamagnetics have the following properties: 1) for a sample of specified geometry, the intermediate states exist for a wider range of field strengths; 2) the width of the intermediate states relative to the phase transition field is greater; 3) intermediate states exist for angles between the field and the "easy" axis which are considerably greater than the critical angle in ferromagnetics. Another important experimental advantage is that we can go from first-order to second-order phase transition simply by changing the temperature (it is not necessary to reorient the sample or change the direction of the field). This makes it possible to systematically study how the field-frequency dependences of the resonant absorption depend on the nature of the phase transition and thereby gain information about the formation of the intermediate states. We note that the above features of magnetic resonance were discovered experimentally for the metamagnetic phase transition in $FeCl_2$ even before the concept of intermediate state in ferromagnetics had been developed.[10]

EXPERIMENTAL METHOD

The measurements were carried out using a direct-gain radiospectrometer for frequencies $f = 14$–80 GHz, magnetic fields $H = 0$–14 kOe, and temperatures $T = 1.45$–4.25 K. In each specific case we recorded the absorption signals either by sweeping the field with f and $T = \text{const}$ or by varying T with f, $H = \text{const}$. The temperature was measured by a semiconductor resistance thermometer.

The spherical sample of diameter 0.8 mm was bonded to the center of a piston which closed off one end of a rectangu-

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lar waveguide; the a axis of the crystal was parallel to the wide face of the waveguide, and the ac plane coincided with the plane of the piston, in which the external magnetic field rotated. When the waveguide was excited at the fundamental mode, the magnetic component of the microwave field $\mathbf{h}\|\mathbf{a}$ become polarized as required in order to observe the magnetic resonance branches of the in $ErFeO_3$ (Ref. 11). The sample was oriented and glued to the piston on an x-ray diffractometer, which ensured that the b axis of the crystal was normal to the piston to within $\sim 10'$. The sample were analyzed to ensure that no blocks or twinned crystals were present. The sample size was chosen so that the magnetic resonance frequency f stayed well away from the dielectric resonance frequencies[12] f_{de} (for our choice of sample diameter f_{de} was greater than the maximum f considered). In all the experiments **H** was parallel to **c**, as required for the metamagnetic transition in $ErFeO_3$.

MEASUREMENTS AND DISCUSSION

The tricritical point $T = T_c \approx 2.55$ K separates the regions in which the phase transitions of the first and second kind occur. Figure 1a, b shows some typical frequency-field dependences $f(H)$ for two temperatures $T < T_c$ and $T > T_c$. The curves were found by recording the resonant absorption field at various frequencies by sweeping H. The frequency halfwidth of the lines was ~ 6 GHz, while the field width was ~ 1 kOe.

The magnetic field induces a second-order phase transition for $T > T_c$; Fig. 1a shows $f(H)$ for this case. The MR frequencies are seen to become "softer," as is typical for phase transitions of the second kind. The resonant absorption is due to the AFM phase for $H < H_c$ and to the FM phase for $H > H_c$ (here and below, by the AFM and FM phases we mean the corresponding phases for the erbium subsystem). The curves $f(H)$ for this temperature range have the characteristic property that the MR frequencies from the AFM and FM phases are equal at $H = H_c$, i.e., at the phase boundary.

The situation is qualitatively different for $T < T_c$, in which case the field induces a phase transition of the first kind. The absorption traces $I(H)$ in Fig. 2 show that the slope of the resonance lines changes discontinuously at the characteristic fields H_1 and H_2. The absorption for $H < H_1$ is clearly due to the AFM phase, while for $H > H_2$ it is due to the FM phase. The following properties of the resonance curves for $H_1 \leqslant H \leqslant H_2$ are noteworthy: 1) the absorption I depends almost linearly on H for those traces with sufficiently good frequency resolution (e.g., traces a and e); 2) as H increases, the absorption from the AFM phase drops from $I(H_1)$ to zero at $H = H_2$ (trace a), while the absorption from the FM phase increases (trace e); 2) the AFM and FM phases coexist but have different resonant frequencies; 3) the contribution of each of the phases to the absorption is proportional to its specific volume for $H = \text{const}$; 4) the intermediate state exists only for $H_1 \leqslant H \leqslant H_2$. The last conclusion agrees with the results found by NMR in Ref. 8.

The resonance lines from the coexisting phases overlap over a considerable range of frequencies, so that the two signals are added (e.g., Fig. 2b, c). There is a narrow frequency interval (~ 1 GHz) for which this addition produces a weak absorption peak in the intermediate state (Fig. 2c); however, this does not mean that a new branch of the magnetic resonance has been excited. Indeed, this peak is clearly caused by a broadening of the lines in the intermediate state, which makes the dependence $I(H)$ nonlinear.

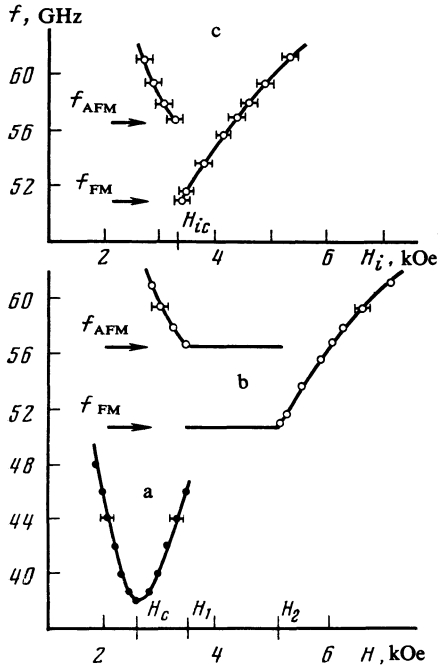

FIG. 1. Field dependence of the resonance frequencies: a) during the second-order phase transition, $T = 3.2$ K, H_c is the critical field for the phase transition; b) during the first-order phase transition, $T = 1.6$ K. The intermediate states exist for $H_1 \leqslant H \leqslant H_2$; f_{AFM} and f_{FM} are the oscillation frequencies of the magnetic moment in the AFM and FM phases; c) recorded at $T = 1.6$ K for the first-order phase transition. $H = H_{ic}$ is the phase-transition field.

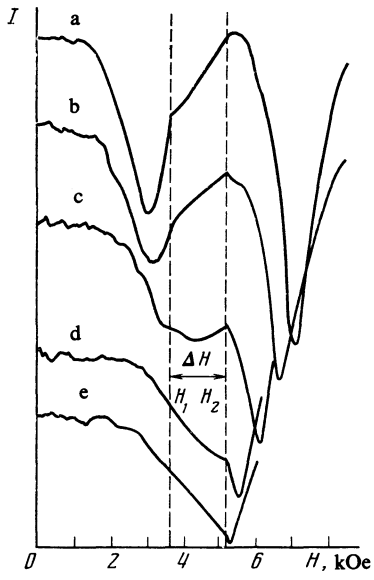

FIG. 2. Traces of the resonant absorption $I(H)$ at $T = 1.6$ K recorded at frequency $f = 60.5$ (a), 59.3 (b), 56.8 (c), 53.4 (d), and 52.4 GHz (e).

The intermediate states are responsible for the following properties of the curves $f(H)$ (Fig. 1b): 1) each field H with $H_1 \leqslant H \leqslant H_2$ corresponds to two resonant frequencies f_{AFM} and f_{FM}; 2) f_{AFM} and f_{FM} are independent of H in the intermediate states. The spectrum in Fig. 1b can also be characterized in terms of the frequency difference $\Delta f = f_{AFM} - f_{FM}$; however, Δf is unrelated to the intermediate states and depends only on the properties of the first-order phase transition. Figure 1c plots f as a function of the internal field $H_i = H - NM$, where $N = 0.33$ is the demagnetizing factor for the spherical sample and M is the magnetization. We determined H_i from data on the magnetization $M(H)$ taken from Ref. 6, because our measurements enabled us to deduce only $\Delta M = \Delta H N^{-1}$, where $\Delta M = M_{FM} - M_{AFM}$. Unlike $f(H)$, the dependence $f(H_i)$ is independent of the shape of the sample, i.e., it has nothing to do with the intermediate states. The values $f(H_i)$ and $f(H)$ coincide only when $N = 0$ (i.e., for samples unbounded along the c axis); in this case the first-order phase transition occurs without the formation of intermediate states.

Our conclusion that the frequencies in the intermediate states are independent of the field was based on an analysis of the absorption traces $I(H)$. This independence could be demonstrated experimentally in a rigorous way by recording the absorption by sweeping the frequency for various fields $H = \text{const}$ between H_1 and H_2. Since our equipment did not enable us to do this, we used the fact that the resonance frequencies are temperature-dependent and varied T instead of f. Although our experimental results can be expressed as usual as field-frequency curves (cf. Refs. 2 and 3, or Fig. 1), it is more convenient to represent them as temperature dependences of the resonance field. The results can then be displayed on the H-T phase diagram.

The phase diagram (Fig. 3) can be constructed from the families of curves $f(H)$ in Fig. 1 and the absorption traces in Fig. 2 recorded at different T. We have already noted that the tricritical point in the phase diagram separates the temperature intervals for which the first- and second-order phase transitions occur. The temperature $T_c = 2.55 \pm 0.05$ K and field $H_c = 3.8 \pm 0.1$ kOe at this point were found by the dielectric resonance method in Ref. 12. The phase diagram for $T < T_c$ was found experimentally by recording the absorption curves $I(H)$ at several frequencies spanning the interval $\Delta f = f_{AFM} - f_{FM}$. For each given T the traces $I(H)$ were used to find the endpoints H_1, H_2 of the field interval for which the intermediate state exists. For example, the family of curves $I(H)$ in Fig. 2 for $T = 1.6$ K gives the results $H_1 \approx 3.5$ kOe, $H_2 \approx 5.2$ kOe. The boundary curve for the second-order phase transition ($T > T_c$) can be found from the frequency-field curves, one of which is shown in Fig. 1a. For each $T = \text{const}$, the minimum frequency corresponds to the critical field H_c. For example, $f(H)$ in Fig. 1a (recorded at $T = 3.2$ K) yields $H_c \approx 2.6$ kOe. This gives one point on the second-order phase transition curve (Fig. 3). Because the second-order phase transition has no direct connection with magnetic resonance in the intermediate state, in our work we merely verified that the values of H_c for several fixed temperatures agreed qualitatively with the phase diagram in Ref. 8.

Figure 3 shows some typical dependences $H_r(T)$ of the resonance field at the two frequencies 37 and 50 GHz. The figure shows that each frequency corresponds to two branches of $H_r(T)$ in the T, H plane, one due to the AFM phase and the other to the FM phase. However, the shape of the curves changes greatly with frequency because they reflect the nature of the phase transition. Thus, for $f = 37$ GHz the AFM and FM branches of $H_r(T)$ merge at a single point on a second-order phase transition curve; by contrast for $f = 50$ GHz $H_r(T)$ corresponds to a first-order phase transition, and resonance occurs in the intermediate state as well as in the AFM and FM phases. The experimental points for the intermediate states were obtained by sweeping T for various $H = \text{const}$ between H_1 and H_2. The absorption $I(T)$ peaked at the two temperatures $T = T_{AFM}$ and $T = T_{FM}$ corresponding to magnetic resonance in the AFM and FM phases. The amplitude of the resonance line depended on the value of H at which the temperature was varied. In addition, the signals from the AFM and FM phases decreased as $H \to H_2$ and $H \to H_1$, respectively.

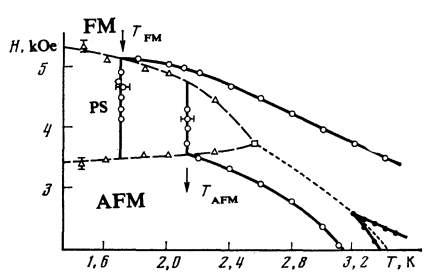

FIG. 3. Phase diagram of $ErFeO_3$: the curves labeled by △ show the lower and upper field bounds H_1, H_2 for which the intermediate states exist; they were found experimentally from $I(H)$ (Fig. 2) recorded at various temperatures; the dashed curve corresponds to the second-order phase transition; □ is the tricritical point; AFM, PS, and FM denote the states of the erbium spins. The temperature dependences of the resonance fields were recorded at $f = 37.0$ GHz (●) and 50.0 GHz (○). T_{AFM} and T_{FM} are the temperatures at which domains in the AFM and FM phases are resonantly excited at a given frequency.

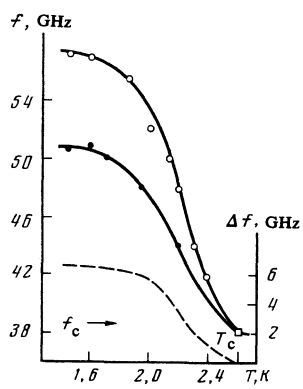

FIG. 4. Temperature dependences of the resonance frequencies f_{AFM} (○) and f_{FM} (●). For each temperature $T < T_c$ there exists a pair of frequencies (f_{AFM} and f_{FM}) and a range of fields $\Delta H = H_2 - H_1$ for which these frequencies are independent of H. The dashed curves shows the temperature dependence of the frequency gap $\Delta f = f_{AFM} - f_{FM}$. The point □ corresponds to T_c and f_c.

The temperature dependences of the resonance fields in Fig. 3 imply that: 1) magnetic resonance in the intermediate state occurs only within a well-defined frequency interval; 2) for each H, the ratio of the signal amplitudes at $T = T_{AFM}$ and T_{FM} is determined by the specific volumes of the AFM and FM phases in the intermediate state; 3) the temperatures T_{AFM} and T_{FM} at which resonance is excited in the AFM and FM phases are independent of the external field. The last conclusion implies that for a given T, the resonance frequencies in the intermediate state are independent of the external magnetic field (see, e.g., Fig. 1b).

The temperature curves for f_{AFM} and f_{FM} in Fig. 4 yield the following conclusions: 1) the frequency gap $\Delta f = f_{AFM} - f_{FM}$ depends on T; 2) the tricritical point in the phase diagram corresponds to a characteristic frequency f_c in addition to T_c and H_c—for $T < T_c$, magnetic resonance in the intermediate state can be excited only at frequency $f > f_c$; 3) magnetic resonance in the intermediate state can be excited for f between f_c and f_0, where $f_0 = f_{AFM}$ $(T = 0)$; 4) the frequency and temperature sweeping methods are interchangeable as far as detecting resonance in the intermediate state is concerned, provided the sweeping ranges of f and T are suitably chosen.

The gap in the spectrum and the existence of the two frequencies f_{AFM} and f_{FM} which are independent of the external magnetic field thus indicate that an internal magnetic field persists in the intermediate state of $ErFeO_3$, and that the magnetic moments of the two coexisting phases oscillate independently.

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[1]V. G. Bar'yakhtar, A. A. Galkin, S. N. Kovner, and V. A. Popov, Zh. Eksp. Teor. Fiz. **58**, 494 (1970) [Sov. Phys. JETP **31**, 264 (1970)].
[2]V. V. Eremenko, A. V. Klochko, and V. M. Naumenko, Pis'ma Zh. Eksp. Teor. Fiz. **35**, 479 (1982) [JETP Lett. **35**, 591 (1982)].
[3]V. V. Eremenko, A. V. Klochko, V. M. Naumenko, and V. V. Pishko, Pis'ma Zh. Eksp. Teor. Fiz. **40**, 219 (1984) [JETP Lett. **40**, 896 (1984)].
[4]V. G. Bar'yakhtar, A. E. Borovik, and V. A. Popov, Zh. Eksp. Teor. Fiz. **62**, 2233 (1972) [Sov. Phys. JETP **35**, 1169 (1972)].
[5]A. M. Kadomtseva, I. B. Krynetskiĭ, and V. M. Matveeva, Zh. Eksp. Teor. Fiz. **79**, 1451 (1980) [Sov. Phys. JETP **53**, 732 (1980)].
[6]G. Gorodetsky, R. M. Hornreich, I. Yaeger, H. Pinto, G. Shachar, and H. Shaked, Phys. Rev. B **8**, 3398 (1973).
[7]V. G. Bar'yakhtar, I. M. Vitebskiĭ, and D. A. Yablonskiĭ, Fiz. Tverd. Tela (Leningrad) **19**, 2135 [Sov. Phys. Solid State **19**, 1249 (1977)].
[8]V. A. Klochan, N. M. Kovtun, G. A. Troitskiĭ, and V. M. Khmara, Zh. Eksp. Teor. Fiz. **81**, 627 (1981) [Sov. Phys. JETP **54**, 335 (1981)].
[9]I. M. Vitebskiĭ and D. A. Yablonskiĭ, Fiz. Tverd. Tela (Leningrad) **19**, 3388 (1977) [Sov. Phys. Solid State **19**, 1978 (1977)].
[10]I. S. Jacobs, S. Roberts, and P. E. Lawrence, J. Appl. Phys. **36**, 1197 (1965).
[11]N. K. Dan'shin, N. M. Kovtun, and A. P. Poluyanenko, Zh. Eksp. Teor. Fiz. **77**, 1058 (1979) [Sov. Phys. JETP **50**, 532 (1979)].
[12]N. K. Dan'shin, N. M. Kovtun, and M. A. Sdvizhkov, in: Proc. All-Union Sem. on Magnetic Phase Transitions and Critical Phenomena, Makhachkala (1984), p. 54.

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