The nature of the soft mode at the low-temperature phase transition in $ErFeO_3$

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The nature of the soft mode is examined for the low-temperature spin-reorientation phase transition (SRPT) in $ErFeO_3$. It is pointed out that the dynamical properties of the rare earth subsystem can in principle be different at the low-temperature (~ 4 K) and high-temperature (~ 100 K) SRPT's.

1. INTRODUCTION

The rare earth orthoferrites belong to the most widely used and intensively studied magnetic insulators. Detailed information on the crystal structure and magnetic properties of rare earth orthoferrites can be found in the monograph of Belov *et al.*,[1] and we shall therefore give only a brief discussion of the objects of study.

The spatial symmetry of rare earth orthoferrites is described by the Fedorov group P_{nma}. The unit cell contains four formula units of $RFeO_3$, where R is a rare earth ion. The presence of two interacting magnetic subsystems, the rare earth and the iron, in these compounds makes for the existence of various phase transitions of the spin reorientation type. The general cause of the phase transitions in orthoferrites is a sharp increase in the contribution of the rare earth ions to the thermodynamic potential of the magnetic subsystem as the temperature is lowered; because the magnetic properties of the rare earth ions have an exceptionally high anisotropy, this often leads to a spontaneous change in the orientation of the magnetization of the iron sublattice with respect to the crystallographic axes, i.e., to a phase transition of the spin reorientation type.

In particular, erbium orthoferrite ($ErFeO_3$) exhibits the SRPT, $\Gamma_4 \to \Gamma_{24} \to \Gamma_2$ on decreasing temperature at $T \sim 100$ K (we are using the standard notation[1] for the magnetic configurations). Here the antiferromagnetism vector **G** changes its orientation as follows[1]: $G_x \to G_{xz} \to G_z$. At $T \sim 4$ K there is another phase transition,[2] $\Gamma_2 \to \Gamma_{12}$, at which the vector **G** deviates from the crystallographic axis **c** in the *bc* plane: $G_z \to G_{zy}$.

Since the Néel temperatures of rare earth orthoferrites are ~ 600 K, at temperatures near the SRPT the iron sublattices are magnetized practically to saturation. The rare earth subsystem, on the other hand, because of the weakness of the R–R and R–Fe interactions, can be regarded as paramagnetic all the way down to liquid helium temperatures. Here the influence of the rare earth ions on the magnetic properties of the material reduces mainly to a temperature-dependent renormalization of the magnetic anisotropy constants for the iron sublattices, and this renormalization leads to the SRPT.

From the standpoint of the dynamic properties this means that the antiferromagnetic resonance (AFMR) modes in the rare earth orthoferrites should be due to vibrations of the iron sublattices, while the rare earth sublattices only influence the value of the resonance frequencies and AFMR linewidth. The temperature dependence of the frequencies of the two acoustic AFMR modes in $ErFeO_3$, taken from Ref. 3, is shown in the inset in Fig. 1. For the SRPT $\Gamma_4 \to \Gamma_{24} \to \Gamma_2 (T \sim 100\ \mathrm{K})$, one of these modes (the σ mode) softens, i.e., its frequency becomes anomalously low, in agreement with the vanishing of the effective magnetic anisotropy for the iron sublattices in the *ac* plane.

If the temperature decreases further (within the Γ_2 phase) and the situtation remains as before, then as the temperature approaches $T \sim 4$ K the frequency of the second AFMR mode (the γ mode) should decrease sharply—it is this mode that, by symmetry, can be responsible for the phase transition $\Gamma_2 \to \Gamma_{12}$. Measurements of the γ mode are restricted to temperatures $T > 100$ K, at which no tendency toward softening is observed. There is another possibility, however. At liquid helium temperatures the role of the rare earth subsystem might not reduce merely to a renormalization of the anisotropy constants (and thus of the corresponding AFMR frequencies) for the iron sublattices but could give rise to an additional mode of homogeneous vibrations with the same symmetry as the γ mode of the AFMR. The frequency of this mode would decrease sharply near the low-temperature SRPT, i.e., this would be the soft mode for the phase transition $\Gamma_2 \to \Gamma_{12}$. This second possibility seems more likely in the present case.

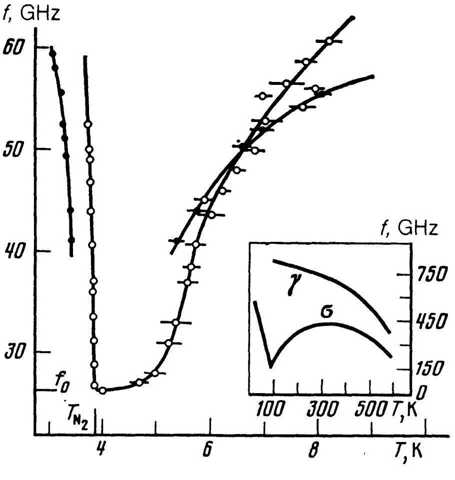

FIG. 1. Temperature dependence of the frequency of the soft magnetic resonance mode near the spontaneous phase transition $\Gamma_{12} \leftrightarrow \Gamma_2$ in $ErFeO_3$: ●) sample No. 1, ○) sample No. 2. The inset shows the temperature dependence of the resonance frequencies of the σ and γ modes in $ErFeO_3$ as obtained in Ref. 3. The softening of the σ mode at $T \sim 90$ K is due to the SRPT $\Gamma_2 \leftrightarrow \Gamma_4$.

We note that a softening magnetic resonance mode in $ErFeO_3$ at $T \sim 4$ K has been observed experimentally,[4] but its nature remains unclear. It was assumed that this was the AFMR γ mode, which is due to vibrations of the iron sublattices (by analogy with the softening of the σ mode at the SRPT $\Gamma_4 \rightarrow \Gamma_2$).

2. EXPERIMENTAL TECHNIQUE AND SAMPLES

The studies were done on a straight-amplification radiospectrometer in the frequency range 14–70 GHz. The sample was placed on a shorting plunger, and the reflected power was sent through a coupler to a crystal detector. The absorption signal was recorded at a fixed frequency as the temperature was scanned. The working temperature range was 1.65–20 K. For excitation of the soft mode at the phase transition $\Gamma_2 \rightarrow \Gamma_{12}$, the magnetic component **h** of the microwave field must have a projection onto the crystallographic axis **a**, as follows directly from symmetry considerations and agrees with the results of measurements.[4] Single-crystal samples, after orientation on an x-ray diffractometer, were affixed to the plunger in such a way that the **a** axis lay along the wide side of the waveguide cross seciton. This gave $\mathbf{h} \| \mathbf{a}$ when the TE_{10} mode was excited in the waveguide.

The present work became feasible after our clarification of the nature of the additional absorption lines, which had earlier prevented a more careful study of the soft mode.[4] It was established[5] that a dielectric resonance, whose modes interact with the soft mode of the magnetic resonance, is excited in $ErFeO_3$. Because this orthoferrite has a large dielectric constant ($\varepsilon \sim 25$–30), the frequency $\nu \sim (d\varepsilon^{1/2})^{-1}$ of the lower boundary of the dielectric resonance spectrum in $ErFeO_3$ will lie lower, for the same sample dimensions d, than in most other magnets (in garnets, for example, $\varepsilon \sim 10$–15). For observation of the magnetic resonance undistorted by the influence of the dielectric resonance, the dielectric resonance must not be excited in the required frequency range. We achieved this condition by using small samples. The main measurements were made on two samples: No. 1, a sphere of diameter 0.8 mm, and No. 2, a sphere of diameter 1.0 mm. For the 1.0 mm sample the dielectric resonance was tuned out at frequencies up to ~ 50 GHz. After the measurements below 50 GHz were made, the sample size was decreased to 0.8 mm, and the lower boundary of the dielectric resonance spectrum was thereby increased to 70 GHz.

3. EXPERIMENTAL RESULTS AND DISCUSSION

The change with temperature of the magnetic resonance frequency in sample No. 2 is shown in Fig. 1. The temperature corresponding to the maximum absorption signal was determined to an accuracy of ~ 0.01 K on the left-hand part of the $f(T)$ curve and to ~ 0.1–0.4 K on the right-hand part. At $T = 3.89 \pm 0.01$ K there is a kink in $f(T)$. This temperature agrees with the temperature of the spontaneous phase transition $\Gamma_2 \rightarrow \Gamma_{12}$ as measured on this same sample by the dielectric resonance method: $T_{N_2} = 3.88 \pm 0.02$ K (Ref. 5). The value of the magnetic resonance frequency at this temperature corresponds to the energy gap of the soft mode at the phase transition: $f_0 = f(T_{N_2}) = 26.1 \pm 0.2$ GHz. Because of the weakness of the signal, we were unable to make measurements at frequencies near the gap in sample No. 1. For sample No. 1 we found $T_{N_2} = 3.5$ K. An important circumstance for explaining the origin of the observed magnetic resonance is the fact that the slope of the $f(T)$ curve decreases as the temperature is raised above T_{N_2}.

If we assume that the observed mode is the softened γ mode, the temperature dependence of which is shown in the inset in Fig. 1, then in order for this dependence to go over monotonically to the dependence observed by us, the slope of our curve should be greater than 9 GHz/K. It follows from Fig. 1 that $\partial f/\partial T$ for sample No. 2 is already less than 9 GHz/K at temperatures $T > 6$ K and shows a tendency to decrease with increasing temperature. The results obtained on sample No. 1 gives no justification whatever for the hypothesis linking this mode to the γ mode.

It can be assumed with a high degree of assurance that the dynamics of the iron sublattices is described satisfactorily by the Landau-Lifshitz equations or the equivalent quantum mechanical equations of motion (see, e.g., Ref. 6). For the rare earth subsystem the situation is more complicated.

The lower Kramers doublet of the Er^{3+} ion in $ErFeO_3$ is separated from the nearest excited level in the crystalline field by an energy gap of 45 cm^{-1}, which corresponds to ≈ 65 K (Ref. 7). One can thus to very good accuracy describe the Er^{3+} ions by an effective spin $s = 1/2$ and an anisotropic g factor. However, to describe the dynamics of the rare earth subsystem, it is not clear beforehand whether to use a) nondissipative equations of motion such as the Landau-Lifshitz equations or b) purely relaxational equations of motion.

In case a) the spectrum of the homogeneous vibrations of the spin system of $ErFeO_3$ includes eight normal modes, in accordance with the number of magnetic sublattices. All these modes were determined in Ref. 6 for the various types of magnetic ordering admissible in rare earth orthoferrites. In particular, the vibrations of the spin system which have the symmetry of the soft mode at the SRPT $\Gamma_2 \rightarrow \Gamma_{12}$ are referred to in Ref. 6 as the modes $E_{12}(2,3)$. These modes are excited by an rf field with polarization $\mathbf{h} \| \mathbf{a}$. The expression for the frequencies of these modes has the structure[7]:

$$\omega_{\pm}{}^2 = {}^1/_2\{\Omega^2 + \omega^2 \pm [(\Omega^2 - \omega^2)^2 + \psi]^{1/2}\}. \tag{1}$$

Here

$$\Omega \sim \gamma [H_E (H_A + H_{\mathrm{Fe-R}})]^{1/2}, \tag{2}$$

$$\omega \sim \gamma [H_{\mathrm{R-Fe}} (H_{\mathrm{R-Fe}} + H_{\mathrm{R-R}})]^{1/2}, \tag{3}$$

$$\psi \sim \gamma^4 H_E H_{\mathrm{Fe-R}} H^2_{\mathrm{R-Fe}}. \tag{4}$$

In (2)–(4) H_E and H_A are the exchange field and anisotropy field for the iron subsystem; $H_{\mathrm{Fe-R}}$, $H_{\mathrm{R-Fe}}$, and $H_{\mathrm{R-R}}$ are, respectively, the effective fields created by the rare earth ions at the iron sublattices, by the Fe^{3+} ions at the rare earth sublattices, and by the rare earth ions at the rare earth ions.

The experimentally observed[3] γ mode of the AFMR did not exhibit a tendency toward softening with decreasing

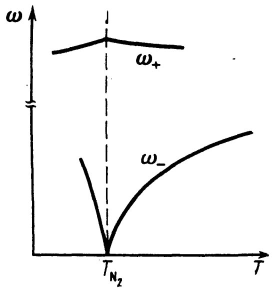

FIG. 2. Schematic temperature dependence of the frequencies of magnetic resonances having the symmetry Γ_{12} of the soft mode for the low-temperature SRPT $\Gamma_2 \leftrightarrow \Gamma_{12}$ in $ErFeO_3$.

temperature (see the inset in Fig. 1), and the $f(T)$ curve observed in the present study becomes flatter with distance from T_{N_2}, especially for sample No. 1 (Fig. 1). Both these circumstances support the idea that the resonance mode in Fig. 1 is not the γ mode of the AFMR but an additional branch $\omega_-(T)$ involving vibrations of the rare earth sublattices. The γ mode would then correspond to the $\omega_+(T)$ branch (Fig. 2).

If situation b) is realized near the low-temperature SRPT in $ErFeO_3$, then the influence of the rare earth subsystem on the spin dynamics would reduce to a temperature-dependent renormalization of the AFMR frequency (this situation obtains at $T \approx 100$ K in the region of the SRPT $\Gamma_4 \to \Gamma_{24} \to \Gamma_2$). In this case the only magnetic resonance mode having the symmetry of the soft mode for the SRPT $\Gamma_2 \to \Gamma_{12}$ would be the γ mode of the AFMR, but in light of the above discussion this mode does not correspond to the soft mode that we observed.

Thus it is most likely that the rare earth subsystem in $ErFeO_3$ is relaxational at high temperatures ($T \approx 10$ K) and dynamic at liquid helium temperatures. As a result, the high-temperature SRPT $\Gamma_2 \to \Gamma_4$ is accompanied by softening of the σ mode, which involves vibrations of the iron sublattices, while the low-temperature SRPT $\Gamma_2 \to \Gamma_{12}$ is associated with the softening of an additional mode ω_-, which arises on cooling below 100 K and involves mainly vibrations of the rare earth subsystem.

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