The relationship between spin-wave and thermodynamic contributions in the dynamics of orientational transitions

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The high-frequency and acoustic characteristics in a metamagnetic transition in the erbium subsystem of $ErFeO_3$ are found to be correlated. The results of the experiment are explained using the idea of a shift in the role of spin-wave and thermodynamic mechanisms responsible for the motion along the line of the metamagnetic second-order phase transition. © *1994 American Institute of Physics.*

1. INTRODUCTION

This paper deals with the spin-reorientation second-order phase transitions (PT2) characteristics of, among others, most rare-earth (RE) orthoferrites (OF).[1] The traditional approach to describing their dynamics is to use the spin-wave approximation. This approximation embodies the conservation of the absolute value of the sublattice magnetizations, and the resonant properties of the substances are related to the transverse high-frequency susceptibility (the variation of the magnetic moment density is due only to precession).[2] Within this model the dominant feature in forming the energy gaps in the spectra of soft magnetoresonance modes may be the magnetoelastic interaction.[3] However, in the exchange (thermodynamic) approximation, when longitudinal magnetization oscillations play the main role in the dynamics, the energy gap at the transition point can be almost ten times greater than the magnetoelastic contribution.[4] This has been reliably established in experiments with $YFeO_3$ and $DyFeO_3$ (see Refs. 4 and 5). An important feature of this mechanism is that it occurs only when there is an external magnetic field. Here the energy gap is determined primarily by the product $\sqrt{\chi_\parallel/\chi_\perp}\, H_{tr}$, where $\chi_\parallel$ and $\chi_\perp$ are in this case, respectively, the longitudinal and transverse susceptibilities of the iron sublattices, and H_{tr} is the strength of the transition field.

Dan'shin and Kramarchuk[6] were the first to point out that under realistic conditions the different gap formation mechanisms discussed in Refs. 3 and 4 generally compete with each other, always coexist, and contribute independently to the gap value measured experimentally. Here the ratio of the contributions of the transverse and longitudinal oscillations of the magnetic moments of the sublattices depends on the range of temperatures and magnetic field strengths in which the transition exists, i.e., on the specific substance. Redistribution of these contributions has been vividly demonstrated in experiments with $ErFeO_3$ (see Ref. 7), in which temperature variation produces a succession of spontaneous orientational transitions

$$\Gamma_4(F_z,G_x)-\Gamma_{24}(F_{zx},G_{zx})-\Gamma_2(F_x,G_z)-\Gamma_{12}(F_x,G_{zy})$$

(here **F** and **G** are the vectors of ferromagnetism and antiferromagnetism in iron).

This paper studies the low-temperature edge of this sequence, the vicinity of the $\Gamma_2-\Gamma_{12}$ transition encountered at $T=T_{N2}\simeq 4$K. Here antiferromagnetic ordering of erbium along the **c** axis (the z axis) is superposed on the iron reorientation. In the $T\leq T_{N_2}$ range a magnetic field $\mathbf{H}\|\mathbf{c}$ may induce a metamagnetic transition in the erbium subsystem, $C_z^R - F_z^R$ (see Ref. 8), where $\mathbf{C}^R$ and $\mathbf{F}^R$ are the vectors of antiferromagnetism and ferromagnetism in erbium. On the line of this transition we discovered a marked correlation between $\sqrt{\tilde{\chi}_\parallel/\tilde{\chi}_\perp}$ and the gap $\nu_{N2}(H,T)$, where $\tilde{\chi}_\parallel$ and $\tilde{\chi}_\perp$ are, respectively, the longitudinal and transverse high-frequency susceptibilities of the erbium sublattices. According to the theory developed in Refs. 4 and 5, this is because the longitudinal oscillations of erbium magnetization contribute the most to the gap value. However, in contrast to $DyFeO_3$ and $YFeO_3$, in $ErFeO_3$ the magnetoelastic contribution to gap formation is comparable to the contribution of longitudinal magnetization oscillations. In view of this we can expect an appropriate response from the elastic subsystem to the shift in the role of the transverse and longitudinal susceptibilities in the spin dynamics of the given PT2. Eventually, because of the dynamic interaction of the spin and elastic subsystems, their "initial" vibrational spectra must be distorted owing not only to transverse magnetization oscillations[3] but to longitudinal magnetization oscillations.[4] This paper is devoted to studying the implications of this statement.

2. THE EXPERIMENT

Figure 1 depicts a fragment of the H vs T phase diagram of $ErFeO_3$ in a field $\mathbf{H}\|\mathbf{c}$. The diagram has the shape common to metamagnets: a PT2 line for weak magnetic fields, a tricritical point with coordinates $T=T_3$ and $H=H_3$, and a first-order phase transition through an intermediate state for $T<T_3$ and $\partial H/\partial T<0$. More details about the structure of this transition and the description of the phase diagram can be found in Ref. 8. In this paper we analyze the high-frequency and acoustic characteristics of this transition only on the PT2 line, i.e., in the temperature interval from T_3-T_{N2}.

The numerical values of T_3 and T_{N2} are known to be extremely sensitive to impurities and the way the single crystals are grown. The samples used in the high-frequency and

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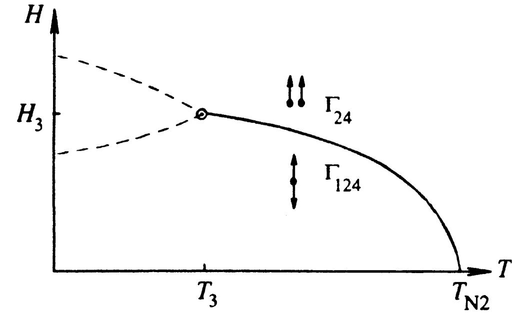

FIG. 1. The phase diagram of $ErFeO_3$ for $\mathbf{H}\|\mathbf{c}$. The solid curve corresponds to PT2, the dashed curves are the intermediate-state boundaries in PT1, and H_3 and T_3 are, respectively, the field strength and temperature of the tricritical point ⊙. The arrows schematically represent the magnetizations of the erbium sublattices.

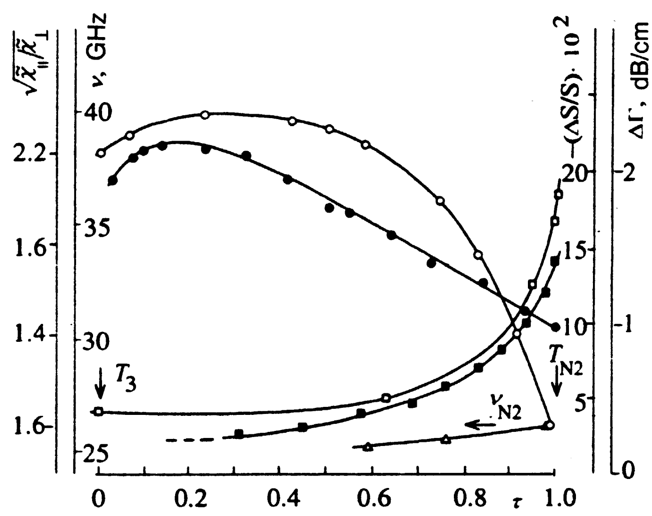

FIG. 2. The temperature–field dependence of the various dynamic parameters on the metamagnetic PT2 line in $ErFeO_3$. Here ● represents $\sqrt{\tilde{\chi}_{\|}/\tilde{\chi}_{\perp}}$, the square root of the ratio of the high-frequency susceptibility; ○ represents the energy gap ν_{N2} for $\mathbf{H}\|\mathbf{c}$; □ represents the relative variation of the speed of the active transverse acoustic mode ($\mathbf{q}\|\mathbf{c}$ and $\varepsilon\|\mathbf{b}$), $\Delta S/S$; △ represents the variation of the decay of longitudinal sound, $\Delta\Gamma$; ■ represents the energy gap ν_{N2} for $\mathbf{H}\|\mathbf{a}$; and $\tau=(T-T_3)/(T_{N2}-T_3)$.

ultrasonic experiments were manufactured from different batches of the initial raw material and their parameters differed somewhat: in the first case $T_{N2}=3.9\pm0.1$ K and $T_3=2.7\pm0.1$ K, and in the second $T_{N2}=4.1\pm0.1$ K and $T_3=2.6\pm0.1$ K. The tricritical-point field strength H_3 for a spherical sample (0.9 mm in diameter) used in magnetoresonance measurements amounted to 4.1 ± 0.1 kOe. For the samples (disks 4 mm in diameter and 1.9 mm high) used in the ultrasonic experiments the field was of the same order of magnitude: 6 ± 0.5 kOe.

High-frequency measurements were done with a wideband microwave spectrometer operating in the 14–80 GHz frequency range, while acoustic measurements were done with a pulsed ultrasonic spectrometer operating in the 25–30 MHz range. The high-frequency and acoustic measurement technique does not differ essentially from that described in detail in Refs. 6 and 9.

3. RESULTS AND DISCUSSION

Figure 2 depicts the set of the dynamic characteristics measured on the metamagnetic transition line as functions of the dimensionless temperature $\tau=(T-T_3)/(T_{N2}-T_3)$. Using a dimensionless temperature permits us to compare the results obtained for samples manufactured from different batches of the initial raw material.

There have already been in-depth theoretical and experimental studies that make it possible to more or less accurately explain the dynamics of spontaneous orientational transitions in rare-earth orthoferrites without leaving the spin-wave approximation. This is due to the "favorable" fact that in most orthoferrites the reorientation of iron occurs at temperatures $T \ll T_{N1}$, where $T_{N1}=620$–740 K is the iron ordering temperature. The same is true of the spontaneous transition Γ_2–Γ_{12} in $ErFeO_3$. It has been firmly established that the soft magnetoresonance mode is caused by vibrations of the RE erbium ions, while the energy gap at the transition point is determined by the ratio of the characteristic frequencies of the rare-earth element and iron and by the magnetoelastic interaction.[3] The measured value of the gap, $\nu_{N2}=26.1\pm0.1$ GHz, agrees with the theoretical estimate, $2\pi\nu_{N2}=140$ GHz, made in Ref. 3. This value of ν_{N2} corresponds to a single point on the phase diagram in Fig. 1, $(H=0, T=T_{N2})$, and is depicted in Fig. 2 at $\tau=1$.

As for the acoustic modes, one should remember that in this transition the transverse acoustic mode with the wave vector $\mathbf{q}$ parallel to the $\mathbf{c}$ axis and the polarization of the shear vector ε parallel to the $\mathbf{b}$ axis (or $\mathbf{q}\|\mathbf{b}$ and $\varepsilon\|\mathbf{c}$) is the active mode, i.e., the one interacting with magnons. The anomalies in the speed of propagation and the absorption of this acoustic mode were first discovered in experiments[10] and later were extensively studied,[9,11] including the theoretical aspects.[3,12] The specific features of these anomalies are the gigantic resonant (asymmetric in T_{N2}) decreases in speed ($\Delta S/S \sim 25\%$) and increase in absorption ($\Delta\Gamma \sim 100$ dB cm). These have never before been detected in a single rare-earth orthoferrite. In Fig. 2, $S=3.98\times10^5$ cm/s is the speed of the active transverse sound far from the transition, which is practically the same for $T \lesssim T_{N2}$. The same figure also depicts the values of the decrease in the speed of the active acoustic mode and the increase in the decay of the longitudinal (inactive) acoustic mode on the PT2 line. Clearly, the maximum variations in the acoustic characteristics of both acoustic modes ($\Delta S/S \sim 20\%$ and $\Delta\Gamma \sim 1.2$ dB cm) correspond to the spontaneous transition point ($\tau=1$). A qualitative explanation of the values of $\Delta S/S$ and $\Delta\Gamma$ is given in Refs. 3 and 12.

Thus, on the whole the dynamics of this spontaneous transition can be satisfactorily described within the spin-wave approximation,[3] which is based on treating as fully as possible the interaction of the various vibrational REOF subsystems: the ordered-iron, the paramagnetic rare-earth, the dipole, the elastic, etc.

Note that within this experimental geometry the acoustic modes under investigation are, respectively, transverse and longitudinal not only in relation to **q** but also to the vector $\mathbf{C}^R$ of antiferromagnetism in erbium. This makes it possible to ask how the longitudinal and transverse oscillations of erbium magnetization are related to the respective acoustic excitations.

When examining a stimulated PT2, we treat the quantities measured at the spontaneous transition point as the initial values. Then the possible effects in which longitudinal magnetization oscillations manifest themselves must be sought in the temperature (field) gradients of the measured parameters. Here we can employ the model of Ref. 3 and the model of Ref. 4. Indeed, although in the present form the theory developed in Ref. 3 deals with spontaneous transitions, it contains no specific field-related effects or restrictions concerning stimulated transitions, and in some cases successfully describes the dynamics of both. On the other hand, the model of Ref. 4 entirely ignores spontaneous transitions, and the effects it predicts occur only in an external magnetic field.

All the temperature curves in Fig. 2 are actually temperature–field curves, i.e., to each value of τ there corresponds a specific field value determined by the phase diagram. In what follows we are not interested in the field value at every point. What is important is the general tendency: the field on the PT2 line increases monotonically as the temperature is lowered.

We first turn to the high-frequency experimental data. The technique for extracting these data is described in detail in Ref. 7. Here we give only the final results and conclusions needed for comparison with the acoustic data. Figure 2 depicts two high-frequency characteristics: the resonant characteristic, the size of the energy gap $\nu_{N2}(T,H)$ in the spectrum of the soft magnetoresonance mode, and the nonresonant characteristic, the square root of the ratio of the high-frequency susceptibilities, $\sqrt{\tilde{\chi}_\parallel/\tilde{\chi}_\perp}\ (H,T)$. The susceptibilities were measured at frequencies $\nu<\nu_{N2}$ by the dielectric resonance method.[13] The curves show that as the field $\mathbf{H}\parallel\mathbf{c}$ grows and the temperature is decreased appropriately, both ν_{N2} and $\sqrt{\tilde{\chi}_\parallel/\tilde{\chi}_\perp}$ increase, with the maximum gradients occurring as $T\rightarrow T_{N2}$ and $H\rightarrow 0$. The energy gap grows from 26.1 GHz at $T=T_{N2}$ to $\sim$38 GHz at the point $T=T_3$, while passing through a moderate maximum. The possible origin of this maximum is discussed in Ref. 7 but is unimportant here. There are more significant features to note, however. First, the temperature variation of ν_{N2} and of $\sqrt{\tilde{\chi}_\parallel/\tilde{\chi}_\perp}$ are clearly correlated. Second, even the lowest operating temperature $T=T_3$ cannot satisfy the condition for the spin-wave approximation, $T/T_{N2}\ll 1$, in the erbium subsystem, since the minimum value T/T_{N2} amounts to approximately 0.7. The third feature follows from a characteristic property of metamagnets: a very high longitudinal susceptibility accompanied by a very low transverse susceptibility. In our case the ratio $\tilde{\chi}_\parallel/\tilde{\chi}_\perp$ reaches a peak value of approximately 7, while in ordinary reorientation of iron sublattices in REOF (see, e.g., Refs. 4 and 6) this ratio cannot, by definition, be greater than unity. Here we are speaking specifically of the susceptibility of the erbium subsystem, since the contribution of iron at liquid helium temperatures is negligible.[14] As a result, if we allow for the fact that the thermodynamic theory[5] satisfies the experiment in the best possible way for large $\chi_\parallel/\chi_\perp$ and T/T_{N1} and that the gap in this model $\sim\sqrt{\chi_\parallel/\chi_\perp}H_{tr}$, we arrive at the following conclusion. The experimentally measured energy gap in the metamagnetic transition in the erbium subsystem is formed with the participation of both transverse and longitudinal magnetization oscillations and is the result of summation of these partial contributions. Here the initial gap at $T=T_{N2}$ and $H=0$ is well-described by the model of Ref. 3, which allows only for transverse magnetization oscillations, while the increase in the gap value in a field $\mathbf{H}\parallel\mathbf{c}$ agrees qualitatively with the theory of Ref. 5, in which the principal contribution to the gap value is related to longitudinal oscillations. Incidentally, in a field $\mathbf{H}\parallel\mathbf{a}$, where the stimulated transition has the same structure as the spontaneous (i.e., Γ_{12}–Γ_2), the initial gap remains practically the same (see Fig. 2) because it is determined, as before, by the transverse oscillations of erbium magnetization.

Now let us study the results of acoustic measurements from this angle. Note that at the spontaneous transition point all the characteristics depicted in Fig. 2 and their temperature (field) gradients are extremal. As the temperature falls and the field strength increases accordingly, the gradients monotonically decrease (in absolute value) and in the τ =0.3–0.5 range all the curves essentially flatten out. If we ignore the insignificant decrease in ν_{N2} and $\tilde{\chi}_\parallel/\tilde{\chi}_\perp$ as the temperature drops into the τ=0–0.3 range, we can speak of a correlation between the dynamic characteristics of the spin and elastic subsystems on the metamagnetic PT2 line in $ErFeO_3$. First we would like to call this fact to the attention of theoreticians studying the dynamics of magnetic orientational transitions. We believe that there is an obvious need to account for the magnetoelastic interaction when describing the dynamics of stimulated orientational transitions. At the present stage we will attempt to explain the results qualitatively. On the whole they agree with the idea of the shift in the role of longitudinal and transverse magnetization oscillations in the formation of reorientation dynamics.

We start with a known fact, the anomaly in the speed of sound in a spontaneous transition caused by the relationship between the transverse acoustic wave and the transverse magnetization oscillations at the natural frequency of the soft magnetoresonance mode (the erbium mode in the given case). Buchel'nikov, Bychkov, and Shavrov[3] have shown the degree of this relationship. In a field $\mathbf{H}\parallel\mathbf{c}$ the longitudinal susceptibility of the erbium subsystem exceeds the transverse susceptibility and increases as the field strength grows and the temperature lowers in the interval from T_{N2} to T_3. But a transversely polarized acoustic wave does not interact with longitudinal magnetization oscillations. Therefore, as the temperature is lowered, the anomaly in the speed of this wave decreases as longitudinal and transverse oscillations exchange roles in creating the dynamics and, consequently, reducing the effect of the spin subsystem on the acoustic subsystem via magnetoelastic interaction.

For the iron subsystem the given temperature range undoubtedly satisfies the spin-wave approximation, since $T_{N2}\ll T_{N1}$. Hence there are practically no longitudinal oscillations of the vectors **F** and **G**. At the same time, the longitudinal components of both vectors can participate in creating the dynamics. We believe that the residual anomaly in the speed of sound in the interval $\tau=0-0.3$, which is about 4% of S, is most likely linked to precisely the transverse oscillations of iron spins. Indeed, in order of magnitude the residual anomaly is comparable to the speed of the active sound in the vicinity of the "high-temperature" reorientation of iron, $\Gamma_2-\Gamma_{24}-\Gamma_4$ at $T=90-100$ K, where $\Delta S/S$ reaches a value of 1.5% (see Ref. 9). The reasons why the speed anomaly in the latter case may be smaller than in the "low-temperature" $\Gamma_{12}-\Gamma_2$ reorientation (which is what we observe here) have been thoroughly analyzed in Ref. 3. However, e.g., in $TmFeO_3$, where the dynamics of the $\Gamma_2-\Gamma_{24}-\Gamma_4$ transition is for all practical purposes determined entirely by the iron sublattices, $\Delta S/S$ reaches an even closer value, $\sim 3\%$ (see Ref. 15).

Many experiments have established that acoustic waves decay (longitudinal waves to a lesser extent than transverse waves) in various spin-reorientation transitions in REOF. Here we are interested in the transverse wave with $\mathbf{q}\|\mathbf{H}\|\mathbf{c}$. How is the decay of this wave related to longitudinal susceptibility? At the spontaneous transition point the decay of this wave is at its maximum. As the temperature falls and the field strength increases, $\Delta\Gamma$ decreases. If the scales along the $\Delta\Gamma$ and $\Delta S/S$ axes are chosen appropriately, one can see that the ways in which $\Delta\Gamma$ and $\Delta S/S$ depend on T and H practically coincide. The most obvious reason for the decrease in the decay of sound as we move closer to the tricritical point is the increase in the rigidity of the magnetic sublattices of erbium (as a result of spontaneous saturation of magnetization as $T\rightarrow 0$ and the effect of the external field $\mathbf{H}\|\mathbf{c}$). Although here the total longitudinal susceptibility grows, the decay of longitudinal sound is most likely linked to its fluctuating part. The longitudinal susceptibility component χ_{zz} can be represented as the sum of two terms, $\chi_{zz}^{\mathrm{L}}+\chi_{zz}^{\mathrm{F}}$, where χ_{zz}^{L} is the jump in susceptibility at the PT2 point predicted by the Landau theory, and χ_{zz}^{F} is the fluctuation increase in susceptibility. For $ErFeO_3$ this is also justified by the fact that the PT2 in question is a combination of two transitions: reorientation in iron and ordering in erbium. The first term reflects the "rigid" part of the susceptibility of erbium, whose ordering via the Er–Fe interaction causes the reorientation of $\mathbf{G}$, while the second term occurs because of fluctuations of F_z^{R} and C_z^{R}. In Ref. 16 it is shown that as we move closer to the tricritical point, the relative contribution of fluctuations to the longitudinal susceptibility χ_{zz} decreases on the PT2 line: theoretically, $\Delta\chi_{zz}^{\mathrm{F}}\Delta\chi_{zz}^{\mathrm{L}}\rightarrow 0$ as $T\rightarrow T_3$ and $H\rightarrow H_3$. But the absolute value of χ_{zz}^{L} grows faster than χ_{zz}^{L}, which in the final analysis augments the role of longitudinal susceptibility in the formation of the energy gap in the spin-wave spectrum, as follows from the theory developed in Refs. 4 and 5.

Finally, the following must be noted. A field $\mathbf{H}\|\mathbf{c}$, in contrast to $\mathbf{H}\|\mathbf{a}$, not only stimulates a transition but also changes the structure of the initial phases. Whereas in the second case the transition, as noted earlier, has the same structure as a spontaneous transition, in the first case ($\mathbf{H}\|\mathbf{c}$) it transforms into $\Gamma_{124}-\Gamma_{24}$. The structure of the initial phases of both iron and the rare-earth element acquires components F_z and F_z^{R} of the ferromagnetism vectors that are longitudinal with respect to $\mathbf{H}$ (these components are characteristic of the Γ_4 representation). Erbium that is spontaneously ordered in an antiferromagnetic structure actually becomes a ferromagnet in a field $\mathbf{H}\|\mathbf{c}$, the difference in sublattice magnetizations being of order F_z^{R}. The variation in the structure of the initial phases may be responsible for the maximum gradients of the characteristics depicted in Fig. 2 exactly at the point of such a transformation.

4. CONCLUSIONS

We have found a correlation between the various dynamic characteristics on the metamagnetic transition line in the erbium subsystem in $ErFeO_3$: the high-frequency susceptibility, the energy gap in the spin-wave spectrum, the speed of the transverse acoustic wave, and the decay of longitudinal sound. Our qualitative analysis of the results points to the need for allowing for magnetoelastic interaction also when describing thermodynamically the spin dynamics of orientational transitions, even more so because within the spin-wave approximation it was only by allowing for the magnetoelastic interaction (in addition to other interactions) that we could explain, qualitatively and often even quantitatively, the large number of purely high-frequency and acoustic properties of orientational transitions in REOF. This factor is also important because reorientation in most REOF occurs in a temperature range where neither the spin-wave approximation nor the thermodynamic approximation works well. In such cases a meaningful explanation of the experimental data requires taking account as many of the various interactions that influence the dynamics formation under real conditions as possible.

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