Anomalously large change in sound velocity in erbium orthoferrite

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An explanation is offered for the substantial (25%) decrease in the velocity of a quasielastic wave and for the behavior of the soft quasispin-vibration mode in $ErFeO_3$ near the Γ_2–Γ_{12} orientational phase transition.

A 25% decrease in the transverse sound velocity $\tilde{s}_t$ was recently found[1,2] experimentally in $ErFeO_3$ near the Γ_2–Γ_{12} low-temperature orientational phase transition (at $T=T_3 \simeq 4$ K). This was the first observation of such a large change in $\tilde{s}_t$ in an orthoferrite. In the region of high-temperature orientational phase transitions, both in $ErFeO_3$ (the Γ_4–Γ_{24} orientational phase transition at $T_1 \simeq 100$ K and the Γ_2–Γ_{24} transition at $T_2 \simeq 90$ K) and in other orthoferrites, the maximum change in $\tilde{s}_t$ has ranged from 0.1% to 3% (Refs. 3 and 4). Balbashov *et al.*[5] showed that the rare-earth (f) subsystem plays an important role in the static and dynamic properties of orthoferrites, although this subsystem is in a paramagnetic state. It turns out that the spectrum of orthoferrites consists of four branches, two of which describe vibrations of the f subsystem, while the others describe vibrations of the iron (d) subsystem. Near the orientational phase transition, the soft mode may be either a d mode or an f mode. Magnetoelastic waves in orthoferrites were studied in Ref. 6 without consideration of the f subsystem. It was shown that the reason why the change in $\tilde{s}_t$ near the Γ_2–Γ_{24} transition is small is a limitation imposed on this change by the dipole interaction. However, the 100% change in $\tilde{s}_t$ near the Γ_4–Γ_{24} transition, which was predicted in Ref. 6, is at odds with experiment.[3,4]

In the present letter we offer an explanation of the experimental results on magnetoelastic waves in orthoferrites by taking account of the effect of the f subsystem on the spectrum of magnetoelastic oscillations. In particular, we offer an explanation for the 25% change in $\tilde{s}_t$ observed near the Γ_2–Γ_{12} orientational phase transition in $ErFeO_3$ and the small value of this change near the Γ_4–Γ_{24} transition.

To describe the dynamics of the orthoferrites, we use linearized Landau-Lifshitz equations, Maxwell's equations, and the equations of elastic theory. In the Γ_2 phase the solution of the dispersion relation for the modes of interest here, in the long-wave approximation, is

$$\begin{aligned}
\omega_{\mathrm{I}}^2 &= \omega_{1k}^2 + \omega_{1f}^2 \zeta_{df} + \omega_{4k}^2 \zeta_{de}, \\
\omega_{\mathrm{II}}^2 &= \omega_{1f}^2 (1 - \zeta_{df}), \qquad (1) \\
\omega_{\mathrm{III}}^2 &= \omega_{4k}^2 (1 - \zeta_{df} - \zeta_{de})/(1 - \zeta_{de});
\end{aligned}$$

$$\gamma_{\mathrm{III}} = \omega^2 \omega_E \zeta_{de} (\Lambda_d + \Lambda_f)/[s_4 \omega_{1k}^2 (1 - \zeta_{df})^{1/2} (1 - \zeta_{df} - \zeta_{de})^{3/2}].$$

Here $\omega_{1k}^2 = \omega_{cb}^2 + \omega_{de4}^2 + \omega_{df}^2 + C^2 k^2$ is a quasiantiferromagnetic mode of the d subsystem; ω_{1f}, ω_{4k} are frequencies of vibrations of the rare-earth and elastic subsystems; ζ_{de}, ζ_{df} are parameters of the magnetoelastic and d–f interactions; ω_{df} are the contributions to the spin-wave gap of the d subsystem from the anisotropy in the cb plane, the magnetoelastic coupling, and the d–f coupling, amplified by the homogeneous exchange of this subsystem (ω_E); s_4 is the velocity of transverse sound polarized along the b axis; Λ_d and Λ_f are decay parameters of the d and f subsystems (the latter is renormalized by f–d coupling); and γ_{III} is the decay coefficient of quasielastic branch [$\gamma = \mathrm{Im}(k)$]. The wave vector is in the orientation $\vec{k} \| \vec{Z} \| \vec{c}$. The point of the Γ_2–Γ_{12} orientational phase transition is determined by the condition $\omega_{cb} = 0$ (in this case we have $\zeta_{df} + \zeta_{de} \rightarrow 1$ as $k \rightarrow 0$). Expressions (1) were derived under the condition $\omega_{1k} > \omega_{1f}$ Estimates show that this condition is satisfied near the orientational phase transition of interest in ErFeO$_3$. Results similar to (1)–(2) are found for the Γ_4 phase.

It follows from (1) that near the point of the Γ_2–Γ_{12} orientational phase transition the frequency of the quasielastic branch is a quadratic function of k as $k \rightarrow 0$: $\omega_{\mathrm{III}} = s_4 C k^2 / \omega_{de4}$. On the other hand, we find a velocity $\tilde{s}_4 = \omega_{\mathrm{III}}/k \rightarrow 0$ as $k \rightarrow 0$. Near this transition, in contrast with the Γ_2–Γ_{24} transition, the dipole interaction does not affect the decrease in $\tilde{s}_t$. The quantity γ_{III} is expressed in terms of the decay parameters of the d and f subsystems. The parameter Λ_f is the dominant parameter here, since the f subsystem is in a paramagnetic state. It has been established[5] experimentally that Λ_f falls off substantially with decreasing T. At a large value of γ_{III} it is not possible to find the sound velocity experimentally, because there are no echo signals.[1,3] As the orientational phase transition is approached, the increase in γ_{III} stems from the circumstance that we have $\zeta_{df} + \zeta_{de}$ (the same factor is responsible for the decrease in $\tilde{s}_4$). Near T_1 and T_2, the increase in γ_{III}, because of the approach to the orientational phase transition, occurs against the background of a pronounced decay due to a decay in the f subsystem. The slight increase in the quantity $\zeta_{df} + \zeta_{de}$, with corresponding small changes in $\tilde{s}_4$ and γ_{III}, has the result that the net decay (the background is included) of γ_{III} exceeds a certain critical value which is sufficient to ensure the absence of echo signals. Near the Γ_2–Γ_{12} transition, in contrast, the decay in the f subsystem is substantially weaker. Accordingly, before the absence of echo signals, in the immediate vicinity of T_3, the quantity $\zeta_{df} + \zeta_{de}$ and thus $\tilde{s}_4$ can vary to a much greater extent than near T_1 and T_2. These two factors (the absence of an effect of the dipole interaction at the point T_3 and the sharp decrease in Λ_f with decreasing T) cause the change in $\tilde{s}_4$ near T_3 to be substantially larger than in the region of the high-temperature orientational phase transitions.

According to (1), the soft mode near T_3 is a quasi-rare-earth branch ω_{II}. The magnitude of the activation of this mode at the point of the orientational phase transition is given bv

$$\omega_{\mathrm{II}}(0) = \omega_{1f} [\omega_{de}/(\omega_{df} + \omega_{de})]^{1/2}. \qquad (3)$$

Using experimental results,[5] and assuming that the magnetoelastic coupling constants increase with decreasing T (Ref. 7), we find an estimate $\omega_{\mathrm{II}}(0) \sim 10^2$ GHz of this gap.

This estimate agrees with the experimental result.[1,2]

A pronounced assymmetry in the behavior of the soft mode to the left and right of the point of the Γ_2–Γ_{12} orientational phase transition was observed in Refs. 1 and 2: On the right there is a smooth transition over an interval $\Delta T \sim 1$ K, while on the left there is a sharp transition. This asymmetry may be due to the behavior of the anisotropy constant K_{cb} (the quantity ω_{cb}) near the transition. This constant should be small and should depend only weakly on T on the right of T_3. The reason for this behavior of K_{cb} in erbium orthoferrite is that the f subsystem is nearly in an ordered state at $T \gtrsim T_3$ (an antiferromagnetic ordering of the f subsystem occurs[1,2] at the point T_3, at the same time as the orientational phase transition in the d subsystem). Consequently, the T dependence of the anisotropy constants here becomes different from that near the high-temperature orientational phase transitions. The calculations show that near T_1 and T_2 the constant K_{cb} (and also K_{ac}) depends linearly on T. In the immediate vicinity of T_3, on the right, K_{cb} is essentially independent of T: $K_{cb} \propto \tanh(B/T)$ (B is the f–d exchange constant; we are dealing with the case $T \ll B$). The reason is that the f subsystem is nearly ordered. The specific temperature dependence of the anisotropy constants of the d subsystem can also explain why the low-temperature transition in $ErFeO_3$ is stretched out along T in comparison with the high-temperature transitions. The difference in the temperature dependence of the parameters of the f subsystem on the right and left of T_3 is responsible for the asymmetry of the $\omega_{11}(0)$ gap with respect to this point.

The fact that the Γ_2–Γ_{12} transition is stretched out by an amount $\Delta T \sim 1$ K (as was found independently from calculations) would also facilitate the observation of an anomalously large change in $\tilde{s}_4$ at the point T_3. At the point T_1, in contrast, this situation would be unfavorable for such an observation in the course of the Γ_4–Γ_{24} transition, since in this case we have[6] $\Delta T \sim (10^{-3}–10^{-4})\text{K}^6$.

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