Energy gap at the induced spin-flip transition in $ErFeO_3$

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The dependence of the energy gap in the spin-wave spectrum at the spin-flip transition in $ErFeO_3$ on the magnetic field and the temperature has been investigated by microwave spectroscopy. It has been established that the gap at the point of completion of the spontaneous spin flip transition is equal to 26 ± 2 GHz and remains unchanged in magnetic fields with strengths up to 10 kOe. It has been conducted from a comparative analysis of the experimental results and various theoretical models of the formation of energy gaps that the gap measured in the present work is specified by magnetoelastic and dipolar interactions, while the contribution of the longitudinal oscillations of the magnetization is insignificant.

One of the most characteristic effects associated with the dynamics of magnetic phase transitions of the second kind is the energy gap in the spectrum of a soft resonance mode. In particular, investigations of the antiferromagnetic resonance in rare-earth orthoferrites have shown that the gaps at the spin-flip transitions characteristic of these compounds amount to at least several tens of gigahertz. Interest in this phenomenon has also been aroused by the fact that, according to the conventional theory of phase transitions of the second kind,[1] the frequencies of soft magnetic-resonance modes should vanish at the points of completion of spin-flip transitions.

There have been many theoretical investigations which examined the origin of these gaps, but nearly all of them can be reduced to two models. One of them (see, for example, Ref. 2) initially assumes that the sublattices are magnetized to saturation at the temperatures of spin-flip transitions. Therefore, all the high-frequency effects in an ordered system of spins are determined mainly by the transverse high-frequency susceptibility with maintenance of the magnetization of the sublattices: $\mathbf{M}_1^2=\mathbf{M}_2^2$ ($\mathbf{M}_1$ and $\mathbf{M}_2$ are the magnetizations of a two-sublattice antiferromagnet). On the other hand, the other model (see, for example, Ref. 3) is based on consideration of the longitudinal oscillations and reveals that just these oscillations are responsible for the appearance of the experimentally observed energy gaps and that the magnitude of the effect is directly related to the value of the longitudinal susceptibility. Until recently, these approaches were regarded as alternatives, each of them having been confirmed experimentally, but under different conditions. This prompted us to conduct additional investigations under conditions which would make it possible to compare the experimental data with both models. On the basis of these measurements, it was asserted that these models are not, in fact, alternatives[4] and that each of them can claim to provide a more or less faithful description of an experiment only over a definite (specific for each compound) range of temperatures T and magnetic fields H. Complete agreement between the authors of both theoretical models and of the present work was achieved from this standpoint during a subsequent discussion of the results reported in Ref. 4 together with the results presented in this paper.

The experiments in Ref. 4 were performed on $TmFeO_3$ and $YbFeO_3$. These rare-earth orthoferrites are antipodes in the sense that they have different kinds of soft modes producing the spectra containing the gap that was the subject of investigation. One special feature of rare-earth orthoferrites is the presence of two resonance subsystems, viz., the iron and rare-earth ion subsystems, with characteristic frequencies of the ν_σ and ν_R soft modes. Since the model described in Ref. 3 was devised for the dynamics of only one magnetic subsystem, it was first tested on $YFeO_3$, in which the yttrium rare-earth ion is nonmagnetic, although rare-earth orthoferrites for which $\nu_\sigma \ll \nu_R$ or $\nu_\sigma \gg \nu_R$ are suitable for this purpose. Then the experimentally observed soft mode may also be regarded as being due to only one spin subsystem. $TmFeO_3$ and $YbFeO_3$, respectively, are just such orthoferrites. However, the resonance subsystems of iron and the rare-earth ion in other orthoferrites do not have such strongly differing frequencies, and they are coupled to a greater or lesser degree. To ascertain the possibility of extending the conclusions from Ref. 4 to other rare-earth orthoferrites, experiments similar to those described in Refs. 3 and 4 must be performed on a compound for which $\nu_\sigma \sim \nu_R$. $ErFeO_3$ is such a compound. However, this situation is actually more typical, since in most of the other orthoferrites the iron and rare-earth ion subsystems are coupled quite strongly.[5]

In the present work we investigated the soft magnetic-resonance mode in the vicinity of the same $\Gamma_2(F_x, G_z) \rightleftarrows \Gamma_{24}(F_{xz}, G_{xz})$ transition (F and G are components of the ferromagnetism vector $\mathbf{F}=\mathbf{M}_1+\mathbf{M}_2$ and the antiferromagnetism vector $\mathbf{G}=\mathbf{M}_1-\mathbf{M}_2$) on which the model described in Ref. 3 was tested in experiments with $YFeO_3$ and $DyFeO_3$ and in the subsequent experiments with $TmFeO_3$ and $YbFeO_3$ in Ref. 4. The measurements were performed in the following manner. The spectrum of the soft mode was first constructed in the region of the $\Gamma_2(F_x, G_z) \rightleftarrows \Gamma_{24}(F_{xz}, G_{xz}) \rightleftarrows \Gamma_4(F_z, G_x)$ spin-flip transition under spontaneous conditions ($H=0$, the transition was caused by a temperature change). The results of these measure-

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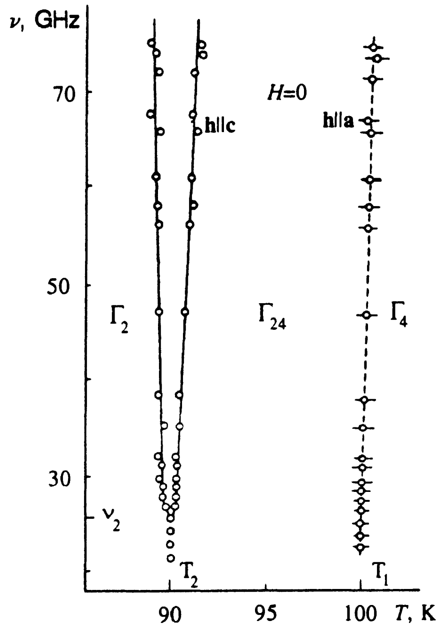

FIG. 1. Spectrum of the soft magnetic resonance mode for the spontaneous Γ_2–Γ_{24}–Γ_4 transition in $ErFeO_3$ (T_1 and T_2 are the temperature boundaries for spin-flip transitions corresponding to a phase transition of the second kind; ν_2 is the value of the gap at the point of the Γ_2–Γ_{24} transition). The experimental points at $\nu<\nu_2$ are associated with absorption on the wings of the resonance lines.

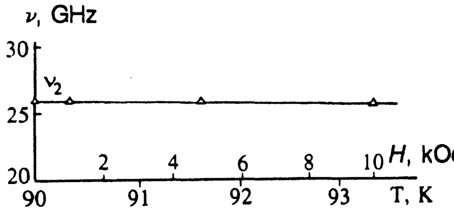

FIG. 2. Dependence of the energy gap at the point of the Γ_2–Γ_{24} transition in $ErFeO_3$ on the magnetic field **H**‖ **a** and the temperature.

ments are shown in Fig. 1. As is seen from this figure, the soft mode is recorded in the usual form only in the vicinity of T_2, i.e., at the point of the $\Gamma_2-\Gamma_{24}$ transition, while a broad single line is observed at T_1 over the entire working range of frequencies. According to Refs. 2 and 5, a spectrum of such form is attributable specifically to the fact that at T_2 the soft mode is formed mainly by oscillations of the iron spins owing to the strong dynamic interaction of the Er and Fe subsystems, while at T_1 it is formed by oscillations of the spins of the rare-earth ions. Since in the temperature range investigated there is strong damping in the rare-earth ion subsystem, the absorption lines from the Γ_{24} and Γ_4 phases are not resolved. This, in turn, precludes direct determination of the gap at T_1. The measured value of the gap $\nu_2=26\pm2$ GHz at T_2 is in good agreement with the calculations in Ref. 2, according to which $2\pi\nu_2\sim140$ GHz. As a whole, it may be stated that the spectrum in Fig. 1 for the spontaneous Γ_2–Γ_{24}–Γ_4 transition is described well by the model in Ref. 2.

What happens to this gap in a magnetic field? An answer to this question is, in fact, provided by ascertaining which of the two mechanisms under discussion, i.e., the mechanism in Ref. 2 or the mechanism in Ref. 3, predominates in forming the gap. There is actually a clear-cut test for the latter: ν_2 should increase in a field, since, according to Ref. 3, $\nu_2\sim\sqrt{(\chi_\parallel/\chi_\perp)}H_{tz}$ ($\chi_\parallel$ and $\chi_\perp$ are, respectively, the longitudinal and transverse susceptibility, and H_{tz} is the transition field). The geometry of our experiment was exactly the same as in Ref. 3 and 4: **H**‖ **a**, **h**‖ **c** (**a** and **c** are axes of the crystal, and **h** is the magnetic component of the high-frequency field). The imposition of **H**‖ **a** shifts the Γ_2–Γ_{24} transition toward higher temperatures: when $H=0$, $T_2=90$ K, while in a field $H=10$ kOe, the transition occurs already at $T\sim93.4$ K.

The measurements in a field were performed in the following manner. Accurate ($\pm5'$) orientation parallel to the **a** axis in the *ac* plane was achieved in the preparatory stage. The procedure used to orient the sample parallel to the axis was the same as that described in detail in Ref. 6. After the sample was oriented parallel to the axis, the gap was measured at fixed values of the field by constructing frequency-temperature plots as the temperature was scanned. We note that the need for accurate orientation of the field parallel to the axis is of decisive importance. Otherwise, as control experiments demonstrated, the gap increases with increasing H, the increase being stronger, the larger the angle between the direction of the field and the axis. This is due to suppression of the phase transition in an oblique field, since only the Γ_{24} canted phase is realized in the sample at any temperature in this case. This effect can be erroneously interpreted as a manifestation of the mechanism in Ref. 3, one of whose main properties is an increase in the gap in a field. The gap was determined in 1.5, and 10-kOe fields. Within the range of accuracy of the measurements, no increase in the gap was detected in these fields (see Fig. 2). It may therefore be concluded that it is described by the model in Ref. 2 in the case of an induced spin-flip transition as well. At first glance the absence of a dependence of the gap on the field (temperature) discovered here contradicts the conclusions in Refs. 3 and 6. However, now, after the experiments in Ref. 4 and a comparative analysis of the latter together with the results and the measurements in Ref. 3, the same conclusion that was drawn in reference to the similar behavior of the gaps in $TmFeO_3$ and $YbFeO_3$ should be drawn here. In the temperature range considered ($T\lesssim100$ K), the longitudinal susceptibility and, accordingly, the contribution of the longitudinal oscillations to the value of the gap are imperceptibly small. Here the gaps are formed mainly by the dynamic interaction of different oscillatory subsystems of the magnet, viz., the elastic and dipolar interactions of iron and the rare-earth ion etc. In particular, in $ErFeO_3$ the value is determined by the magnetoelastic and dipolar contributions.[2] It should, however, be noted that just as the model in Ref. 2 ignores the longitudinal oscillations of the magnetization, the theory in Ref. 3 disregards the dynamic interactions just indicated. Therefore, neither theory alone can claim to be a complete and universal theory for interpreting experimental results. Since the contributions provided by these models to the value of the gap are additive, the partial contributions or the dominant role of some definite mechanism can be assessed in each specific case. Therefore, it would be appropriate to ascertain the magnitude of the effect that would be expected in our experiment, if the main contribution to the formation of the gap were made by the longitudinal oscillations of the magnetization in accordance with the role assigned to them by the theory in Ref. 3. An evaluation using quantitative data from this work reveals that an 8.4-GHz increase in the value of the gap should have been observed in a field $H=10$ kOe. This greatly exceeds the error in our measurements.

Thus, on the basis of the experiments with $TmFeO_3$, $YbFeO_3$, and $ErFeO_3$, it may be concluded that the absence of a dependence of the gap on the field (temperature) is more likely a general property of all rare-earth orthoferrites, in which spin-flip transitions occur at temperatures T_1 and T_2 that are considerably lower than the ordering temperatures of iron $T_N \sim 640$–740 K and accordingly in comparatively small fields. This fact does not depend on the ratio between the resonant frequencies ν_σ and ν_R or, therefore, on the degree of dynamic interaction between the oscillatory subsystems of iron and the rare-earth ion. With consideration of the measurements performed on $YFeO_3$ and $DyFeO_3$ in Ref. 3, it may be added that the models discussed here do not contradict one another and make independent contributions to the formation of the gaps. In addition, at $T \to 0$ the main role is played by the transverse oscillations of magnetizations M_1 and M_2, which were taken into account by the theory in Ref. 2, while the longitudinal oscillations[3] are decisive at $T \to T_N$, at which $\chi_\parallel / \chi_\perp \to 1$.

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