

# Dynamic properties of $\text{YbFeO}_3$ in the vicinity of an orientational phase transition

N. K. Dan'shin, S. V. Zherlitsyn, S. S. Zvada, G. G. Kramarchuk, M. A. Sdvizhkov,  
and V. D. Fil'

*Physicotechnical Institute, Academy of Sciences of the Ukrainian SSR, Donetsk*

(Submitted 24 April 1987)

*Zh. Eksp. Teor. Fiz.* **93**, 2151–2160 (December 1987)

High-frequency and acoustic investigations of ytterbium orthoferrite were made in the vicinity of an orientational phase transition. A soft magnetic resonance mode associated with oscillations of the magnetic moments of  $\text{Yb}^{3+}$  ions was observed. The energy gaps at the points of spontaneous and field-induced transitions were determined. The influence of the magnetoelastic interaction on the anomalies of the velocity and attenuation of sound were determined in the vicinity of these transitions. It was found that the magnetoelastic coupling is enhanced by a magnetic field. A study was made of the influence of misorientation of a magnetic field on the magnetic resonance spectrum, as well as on the velocity and attenuation of sound. It is concluded that the magnetoelastic interaction was not the main mechanism responsible for the formation of the experimentally observed energy gaps in the spectrum of the soft magnetic resonance mode of  $\text{YbFeO}_3$ .

## 1. INTRODUCTION

A typical feature of rare-earth orthoferrites (with the space group  $D_{2h}^{16}$ ) is the presence of two magnetic subsystems in the form of a slightly canted antiferromagnetic structure of  $\text{Fe}^{3+}$  spins and an ensemble of paramagnetic rare-earth ions. The interaction between them is responsible for a great variety of static and dynamic properties and also for orientational phase transitions. In the majority of orthoferrites these transitions involve continuous rotations of the weak ferromagnetic moment of the iron sublattices and of the induced moment in the rare-earth subsystem from the crystal axis  $c$  to the axis  $a$  (as a result of cooling) via a canted phase. The onset and completion of a spontaneous orientational phase transition occur at temperatures  $T_1$  and  $T_2$  specific to each rare-earth orthoferrite. In the range  $T > T_2$  the phase has the  $\Gamma_4$  symmetry, whereas in the range  $T < T_1$  the phase is  $\Gamma_2$  (Ref. 1). The canted phase  $\Gamma_{24}$  exists in the range  $T_1 - T_2$ .

A characteristic feature of the reorientation process in  $\text{YbFeO}_3$  is that the polarization of the magnetic moments of  $\text{Yb}^{3+}$  in the low-temperature phase ( $T < T_1$ ) is described not by one but by two irreducible representations of the  $D_{2h}^{16}$  group, which are  $\Gamma_2$  and  $\Gamma_8$  (Ref. 2). An admixture of  $\Gamma_8$  is due to a considerable increase of the interaction in the rare-earth subsystem as a result of cooling. We shall not ignore this fact, but bear in mind that this admixture was not observed explicitly in our experiments, which justifies labeling the orientational phase transition in the form typical of the majority of rare-earth orthoferrites:  $\Gamma_2 - \Gamma_{24} - \Gamma_4$ .

The dynamics of an orientational phase transition in orthoferrites with relatively high transition temperatures ( $T_{1,2} > 50$  K) is governed primarily by the iron subsystem. At the limits of an orientational phase transition corresponding to the temperatures  $T_1$  and  $T_2$ , which are second-order phase transition (FT-2) points, a quasiferromagnetic mode of an antiferromagnetic resonance (AFMR) of the  $\text{Fe}^{3+}$  spins becomes softer.<sup>3,4</sup> The influence of the rare-earth ions on the dynamic properties reduces to a temperature-dependent renormalization of the AFMR frequencies by the parameters of the R-Fe interaction (R is a rare-earth ion).

This softening may be limited by the interaction with the rare-earth modes if the splitting of the main doublet (quasi-doublet) of the rare-earth ion is considerably less than the characteristic AFMR frequencies. This occurs, for example, in the case of  $\text{HoFeO}_3$  (Ref. 3) and clearly also in  $\text{ErFeO}_3$  (Ref. 4). However, in the case of other ferrites in which an orientational phase transition occurs at relatively high temperatures ( $T_{1,2} < 10$  K) such AFMR mode softening is not observed.<sup>5,6</sup> In the latter orthoferrites the loss of stability necessary for the onset of the transition may be due to softening of one of the rare-earth modes.<sup>7</sup> In the case of  $\text{YbFeO}_3$  the  $\Gamma_2 - \Gamma_4$  reorientation occurs at a temperature lower than in the case of other rare-earth orthoferrites ( $\sim 6-8$  K). Therefore, it would be of interest to investigate the dynamics of this transition in order to identify the nature of the soft mode and the influence of this mode on the effects associated with the magnetoelastic interaction. A more general aim of the present experiments was to obtain information on the theoretically predicted<sup>8</sup> mutual influence of magnons and phonons which should be manifested most strongly at the points of spin reorientation transitions.

## 2. EXPERIMENTAL METHOD

The magnetic resonance spectrum was determined using a direct-amplification reflection microwave spectrometer operating at frequencies in the range 14–79 GHz. Effective wide-band tuning was achieved by a nonresonator method using interchangeable short-circuited rectangular waveguides. The waveguide cross section selected for each frequency interval was such that only the fundamental waveguide mode  $\text{TE}_{10}$  was propagated. Samples were bonded to a rotatable stage located at the center of a short-circuiting plunger. Rotation of this stage altered the orientation of the crystal axes relative to the magnetic component of a high-frequency field  $\mathbf{h}$ . The absorption of the microwave power was determined at fixed frequencies while temperature was varied. A static magnetic field was applied in the plane of rotation of the stage. The main magnetic resonance experiments were carried out on a spherical sample with a diameter of 0.8 mm. The preliminary results of these experiments were published earlier.<sup>9</sup>

Measurements of the relative changes in the phase velocity  $\Delta s/s$  and in the attenuation  $\Delta\alpha$  of sound were made using a self-balancing bridge circuit whose operation was similar to that of the circuit described in Ref. 10. One of the measured parameters ( $\Delta s/s$  in our case) was processed by a microcomputer and then plotted directly as a function of temperature when the latter was varied slowly ( $\sim 0.02$  K/min). A magnetic field up to 35 kOe was created in a small superconducting solenoid. In a weak field (up to 3 kOe) it was possible to tilt within a narrow range ( $\sim 1^\circ$ ) the resultant magnetic field vector, relative to the crystallographic axes of a sample, by means of Helmholtz coils. Acoustic vibrations of frequencies 50–250 MHz were excited by LiNbO<sub>3</sub> piezoelectric transducers at the fundamental frequency of  $\sim 50$  MHz. The wave vector of sound  $\mathbf{k}$  was oriented (to within  $0.5^\circ$ ) along the  $c$  axis for all the polarizations. Measurements using longitudinal and transverse sound with the  $\epsilon\|\mathbf{b}$  polarization of the vibrations were carried out using a cylindrical sample with a diameter  $\sim 5$  mm and of length  $\sim 6$  mm, whereas in the  $\epsilon\|\mathbf{a}$  polarization the length of the acoustic path was reduced because the effect was stronger. In the latter case we used a disk with rounded edges: the thickness of the disk was  $\sim 1$  mm and its diameter was 5 mm. The technique used in the acoustic measurements on insulating crystals, such as the investigated rare-earth orthoferrite, was described in Ref. 11.

All samples were cut from the same single crystal grown by the floating zone method in which radiation heating was employed. The orientation of a sample was determined and final checks were carried out using a DRON-3 x-ray diffractometer.

### 3. SOFT MAGNETIC RESONANCE MODE NEAR AN ORIENTATIONAL PHASE TRANSITION IN YbFeO<sub>3</sub>

We shall first consider the high-frequency properties of YbFeO<sub>3</sub> in the case of a spontaneous orientational phase transition ( $H = 0$ ). Typical resonance absorption curves are shown in Fig. 1. It is worth noting that by selecting the polarization of the oscillations of the hf field as  $\mathbf{h}\|\mathbf{c}$  or  $\mathbf{h}\|\mathbf{a}$  we could excite selectively a magnetic resonance in the vicinity of  $T_1$  or  $T_2$ . A number of records of this type, obtained at several fixed frequencies  $\nu$ , were used to plot the temperature dependence of the magnetic resonance frequency shown in Fig. 2. The minimum resonance frequencies of the spontaneous transition corresponded to the temperatures  $T_1$  and  $T_2$ , which we regarded as the limits of the orientational phase transition. They represented energy gaps in the magnetic resonance spectrum when the reorientation process begins

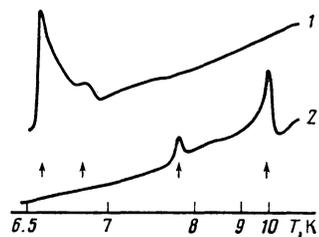


FIG. 1. Temperature dependence of hf absorption in the vicinity of the spontaneous  $\Gamma_2$ - $\Gamma_4$  orientational transition in YbFeO<sub>3</sub> at 52 GHz, obtained using different polarizations of  $\mathbf{h}$ : 1)  $\mathbf{h}\|\mathbf{c}$ , 2)  $\mathbf{h}\|\mathbf{a}$ . The arrows identify the positions of the magnetic resonance lines.

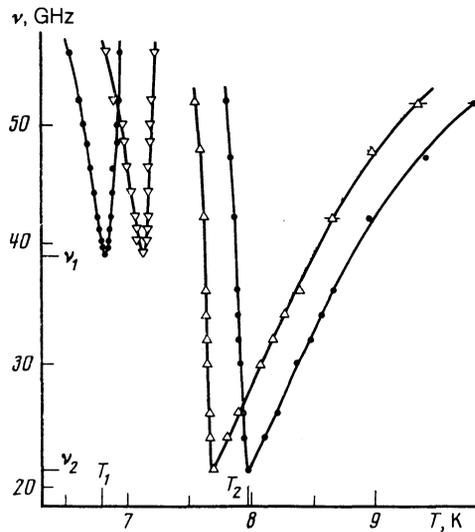


FIG. 2. Temperature dependence of the frequencies of the soft magnetic resonance mode of YbFeO<sub>3</sub> observed in spontaneous  $\Gamma_2$ - $\Gamma_4$  (●) and field-induced  $H = 3$  kOe transitions:  $\Gamma_2$ - $\Gamma_{24}$  in a field  $\mathbf{H}\|\mathbf{a}$  (▽) and  $\Gamma_{24}$ - $\Gamma_4$  in a field  $\mathbf{H}\|\mathbf{c}$  (Δ).

and ends:  $\nu_1(T_1)$  and  $\nu_2(T_2)$ . At operating frequencies in the range  $\nu < \nu_{1,2}$  we observed a signal associated with the absorption at the edge of a resonance line. This was confirmed by the vanishing of the intensity when the frequency was lowered by an amount just equal to the width of an absorption line. Therefore,  $\nu_1(T_1)$  and  $\nu_2(T_2)$  were the gaps in the spectrum and not frequencies below which one could not resolve too closely spaced absorption lines. When the field  $\mathbf{h}$  was inclined to the  $c$  axis in the  $ac$  plane, the intensity of the resonance lines decreased monotonically in the range  $T < T_1$ , whereas in the  $\mathbf{h}\|\mathbf{a}$  orientation they disappeared completely. For  $\mathbf{h}\|\mathbf{b}$  the intensity of the signal was an order of magnitude less than for  $\mathbf{h}\|\mathbf{c}$ . Hence, the magnetic susceptibility of the phase  $\Gamma_2$  obeyed  $\chi_{cc} \gg \chi_{bb}$ ,  $\chi_{aa} = 0$ . The resonance branch observed at  $T > T_2$  was excited preferentially by the field  $\mathbf{h}\|\mathbf{a}$ . This branch was characterized by  $\chi_{aa} \gg \chi_{bb}$ ,  $\chi_{cc} = 0$ .

The influence of a static magnetic field applied in the  $ac$  plane was as follows. When the orientation of the static field was  $\mathbf{H}\|\mathbf{a}$ , the  $\Gamma_2$ - $\Gamma_{24}$  transition shifted toward higher temperatures ( $\partial T_1/\partial H_a = 0.97$  K/kOe) and the  $\Gamma_{24}$ - $\Gamma_4$  transition was suppressed (Fig. 3). The value of the gap at the transition point  $T_1(H_a)$  was not affected (Fig. 2), whereas the resonance branch in the vicinity of  $T_2$  rose even in a field of  $H > 0.2$  kOe beyond the working frequency range. The field  $\mathbf{H}\|\mathbf{c}$  induced the  $\Gamma_{24}$ - $\Gamma_4$  transition which shifted toward lower temperatures ( $\partial T_2/\partial H_c = -0.092$  K/kOe). The gap observed in the case of this field-induced transition, as in the preceding case, was identical with the gap observed for the spontaneous transition. This orientation of the field suppressed the transition in the vicinity of  $T_1$ , so that the corresponding soft magnetic resonance mode disappeared (in fields  $H > 1$  kOe). The experimental dependence in Fig. 3, corresponding to the exact orientation of the field along the  $a$  and  $c$  axes, represented in fact the  $H$ - $T$  phase diagram of YbFeO<sub>3</sub> in the vicinity of the  $\Gamma_2$ - $\Gamma_4$  transition.

A marked reduction in the resonance frequencies on approach to the characteristic temperatures of the orienta-

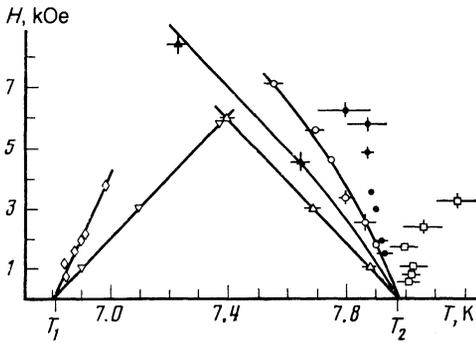


FIG. 3. Relationship between the critical values of the temperature and the magnetic field corresponding to the minimum of the magnetic resonance frequency when the field was inclined in the  $ac$  plane by various angles  $\varphi$ . Slant relative to the  $a$  axis:  $0^\circ$  ( $\nabla$ ),  $5^\circ$  ( $\bullet$ ). Tilt relative to the  $c$  axis:  $0^\circ$  ( $\Delta$ ),  $1^\circ$  ( $\blacktriangle$ ),  $3^\circ$  ( $\circ$ ),  $5^\circ$  ( $\bullet$ ),  $10^\circ$  ( $\square$ ).

tional phase transition indicated that a soft magnetic resonance mode characterized by  $m_1M$  was excited in our experiments. Symmetry analysis<sup>12</sup> indicated that the  $\Gamma_2-\Gamma_{24}-\Gamma_4$  orientational phase transition could soften a quasiferromagnetic mode of AFMR associated with the iron subsystem or one of the rare-earth modes with the same oscillation symmetry ( $m_1M$ ). An investigation of the submillimeter spectrum of  $YbFeO_3$  indicated that the minimum frequency of the quasiferromagnetic mode in the vicinity of the orientational phase transition was  $\sim 20 \text{ cm}^{-1}$  (Ref. 5). This demonstrated unambiguously that the resonance branch we observed in the millimeter range was not an AFMR mode.

The slight changes in the frequency of this branch reported in Ref. 5 could be attributed to softening of the AFMR, which however was limited by the interaction with the rare-earth mode of lower frequency. This interaction could give rise to a branch of coupled oscillations (as found, for example, in the case of  $HoFeO_3$ —Ref. 3) in the form of a soft mode during an orientational phase transition. This could be the resonance mode observed in our experiments. Then, as in the case of high-temperature transitions, the dynamics of the iron subsystem should play the main role.

However, the formation of a soft mode as a result of cooling could occur in accordance with a different mechanism.<sup>7</sup> This mechanism involves a change in the role of the rare-earth subsystem: its contribution to the high-frequency properties becomes mainly dynamic rather than relaxational. In this situation the oscillations of the rare-earth moments are coupled to a greater or lesser extent to oscillations of the iron spins because of the R-Fe interaction. This may be the reason for the anomaly in the temperature dependence of the frequency of the quasiferromagnetic mode observed in Ref. 5. The actual ratio of the dynamic contributions of iron and ytterbium to the observed soft mode can be determined by developing a theoretical model which would allow for both mechanisms and by obtaining experimental results throughout the full range of the resonance spectrum of  $YbFeO_3$ . Nevertheless, since a temperature dependence  $\nu_{AFMR}(T)$  near an orientational phase transition is unusual for a soft mode,<sup>5</sup> preference should be given to the mechanism of formation of the spectrum associated with an increase of the role of the dynamics of the rare-earth subsystem as a result of cooling.

The absolute values of the energy gaps of the soft mode

at the orientational phase transition points were determined in our experiments. In the case of just the spontaneous transitions, two possible mechanisms of formation of the gaps are known at present: the magnetoelastic<sup>8</sup> and the relaxational.<sup>13</sup> The relaxational mechanism<sup>13</sup> can give rise to a gap in the AFMR spectrum because of relaxation in the rare-earth subsystem. Since the soft mode we investigated was not associated with the AFMR, the relaxation mechanism could be ignored. Our case probably corresponded to the situation considered in Ref. 13, when the dynamics of the rare-earth subsystem could be described by purely relaxational equations of motion. An additional nonactivated branch of oscillations then appeared (when no allowance was made for other gap mechanisms), which had the soft-mode symmetry. Therefore, we analyzed only the role of the magnetoelastic interaction of phonons and magnons when considering the formation of energy gaps.

Going back to the field-induced transitions we ask the question: what happens to the soft mode in an oblique field? It is known<sup>1</sup> that in rare-earth orthoferrites exhibiting the high-temperature  $\Gamma_2-\Gamma_4$  reorientation slanting  $H$  from the  $a$  or  $c$  axis in the  $ac$  plane converts  $RFeO_3$  to the canted phase  $\Gamma_{24}$ . This destroys the distinction between the phases and the orientational phase transition disappears. In the resonance experiments this is manifested as follows: the slightest tilt of the field greatly increases the minimum frequency of the quasiferromagnetic AFMR mode, which is no longer soft if  $\varphi \neq 0$  (Refs. 14–16). The results of hf experiments on  $YbFeO_3$  in an oblique field are plotted in Figs. 3–5. Bearing in mind the comments made above, we can see that the curves shown in Fig. 3 and obtained for  $\varphi \neq 0$  are no longer the  $\Gamma_2-\Gamma_{24}$  and  $\Gamma_{24}-\Gamma_4$  second-order phase transition lines, but represent simply the positions of the minimum magnetic resonance frequency obtained for a given field and temperature. The sensitivity of this characteristic “memory” of the phase transition to the field direction is demonstrated in Fig. 4. We can see that whereas in the case of the exact orientation  $H||a$  or  $H||c$  the minimum frequencies (gap) are practically

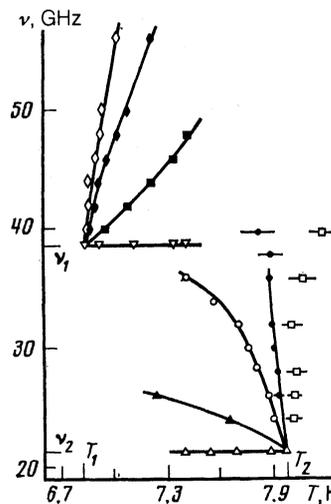


FIG. 4. Temperature dependences of the energy gaps in the case of transitions induced in fields  $H||a$  ( $\nabla$ ) and  $H||c$  ( $\Delta$ ), and of the minimum resonance frequencies in the case of fields tilted in the  $ac$  plane by various angles  $\varphi$ . Tilt relative to the  $a$  axis:  $1^\circ$  ( $\blacksquare$ ),  $3^\circ$  ( $\blacklozenge$ ),  $5^\circ$  ( $\diamond$ ). Tilt relative to the  $c$  axis:  $1^\circ$  ( $\Delta$ ),  $3^\circ$  ( $\circ$ ),  $5^\circ$  ( $\bullet$ ),  $10^\circ$  ( $\square$ ).

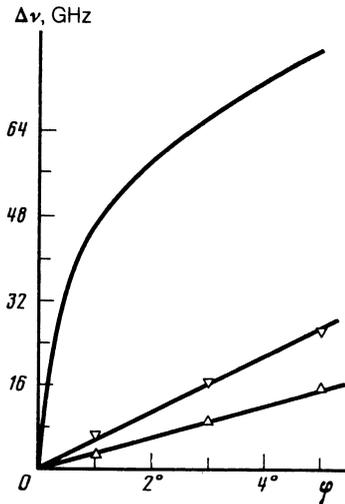


FIG. 5. Angular dependence of the increments in the minimum resonance frequencies of  $\text{YbFeO}_3$ , and in a magnetic field  $H = 5 \text{ kOe}$  slanted relative to the  $\mathbf{a}$  axis ( $\nabla$ ) at temperatures  $T \geq T_1$  relative to the  $\mathbf{c}$  axis ( $\Delta$ ) at  $T \leq T_2$ . The continuous curve represents the results of a calculation of the increment of the frequency gap of  $\text{TmFeO}_3$  (Ref. 14) when a field  $H = 7 \text{ kOe}$  was slanted relative to the  $\mathbf{a}$  axis at temperatures  $T \geq (T_1 = 81 \text{ K})$ .

independent of the field, an oblique field suppresses both transitions (at  $T_1$  and  $T_1$ ) and gives rise to a strong dependence of the minimum magnetic resonance frequencies on the parameters mentioned above.

We compared the dependence of the frequency gap on the field direction obtained in the present experiments with the corresponding calculations for  $\text{TmFeO}_3$  (Ref. 14) by plotting in Fig. 5 the increments of the gaps of  $\text{TmFeO}_3$  and  $\text{YbFeO}_3$  as a function of  $\varphi$ . The results indicate that the gap of  $\text{YbFeO}_3$  is much less sensitive to the field direction than  $\text{TmFeO}_3$ , particularly in the case of small angles  $\varphi$ . This quantitative difference could be due to the different nature of the soft mode in  $\text{TmFeO}_3$  and  $\text{YbFeO}_3$ . Moreover, the calculations of Ref. 14 are made on the assumption that there is no gap for  $\varphi = 0$ , i.e., no allowance is made for any mechanisms of activation at the phase transition point, whereas our results were obtained with actual large "seed" gaps  $\nu_1$  and  $\nu_2$  (Fig. 4), observed also for  $\varphi = 0$ .

#### 4. MAGNETOACOUSTIC EFFECTS NEAR AN ORIENTATIONAL PHASE TRANSITION IN $\text{YbFeO}_3$

It follows from the calculations of Ref. 8 that the magnetoelastic interaction is the main factor responsible for the energy gap in the spectrum of magnons at the orientational phase transition point. The interaction of a soft magnetic mode with an active acoustic mode should reduce the velocity of a quasiaoustic wave to zero when the wave vector is  $\mathbf{k} \rightarrow 0$ . In the case of orthoferrites the expected features in the behavior of the velocity of sound are quite familiar (see, for example, Ref. 17). In accordance with the general theory of second-order phase transitions, the velocity of longitudinal sound changes abruptly at a phase boundary and the dependence of the velocity of transverse sound ( $\mathbf{k} \parallel \mathbf{c}$ ,  $\epsilon \parallel \mathbf{b}$ ) on the external parameter causing the transition has a kink. The velocity of a second transverse acoustic wave ( $\mathbf{k} \parallel \mathbf{c}$ ,  $\epsilon \parallel \mathbf{a}$ ) should exhibit resonance features at the phase transition points. It should be pointed out that, although the experi-

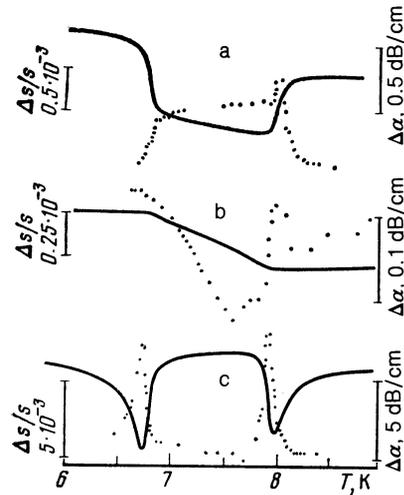


FIG. 6. Temperature dependences of the relative change in the velocity  $\Delta s/s$  (continuous curves) and in the attenuation  $\Delta\alpha$  (points) of sound with the wave vector  $\mathbf{k} \parallel \mathbf{c}$ , determined in the vicinity of the spontaneous orientational phase transition in  $\text{YbFeO}_3$  for different acoustic wave polarizations and frequencies: a)  $\epsilon \parallel \mathbf{c}$ , cylindrical sample with the axis along  $\mathbf{c}$ , frequency of sound 169.9 MHz,  $s(4.2 \text{ K}) = 6.0 \times 10^5 \text{ cm/sec}$ ; b)  $\epsilon \parallel \mathbf{b}$ , same cylindrical sample, frequency of sound 162.9 MHz,  $s(4.2 \text{ K}) = 3.7 \times 10^5 \text{ cm/sec}$ ; c)  $\epsilon \parallel \mathbf{a}$ , disk-shaped sample with the normal along the  $\mathbf{c}$  axis, frequency of sound 54.3 MHz,  $s(4.2 \text{ K}) = 3.3 \times 10^5 \text{ cm/sec}$ .

mental results of Refs. 17–20 agree qualitatively with the theoretical predictions, none of the investigated orthoferrites exhibit changes in the sound velocity in excess of  $\sim 10^{-2}$ . Among the various factors which can be responsible for such a small change in the velocity are sample inhomogeneities (in their structure and in the temperature field), the influence of domain walls, and nonzero values of  $\mathbf{k}$  which are usually encountered in acoustic measurements.

Figure 6 shows the experimental temperature dependence of  $\Delta s/s$  and  $\Delta\alpha$  obtained for various acoustic modes of  $\text{YbFeO}_3$  in the vicinity of the spontaneous orientational phase transition. The results plotted in this figure are in qualitative agreement with the published results of acoustic experiments near similar orientational phase transitions in  $\text{ErFeO}_3$  (Refs. 17, 18, and 20) and in  $\text{TmFeO}_3$  (Refs. 19 and 20).

The anomalies of the velocity and of the attenuation of that acoustic wave which interacts with magnons are of the greatest interest to us. The wave in question is transverse and its polarization is  $\epsilon \parallel \mathbf{a}$ . We shall call it the active acoustic mode. It is clear from Fig. 6 that the anomalies of the velocity of the active mode at the phase transition point are resonant, but they amount to just  $\sim 0.5\%$  (for comparison, in the case of  $\text{ErFeO}_3$  the corresponding value is  $\sim 0.2\%$ , whereas for  $\text{TmFeO}_3$  it is  $\sim 3.2\%$ ). We must begin by pointing out that the working frequencies of sound used in our experiments were much lower than the gaps observed in the magnon spectrum at  $\nu_1$  and  $\nu_2$ . On the assumption of the magnetoelastic origin of the gaps ( $\nu_{1,2} \sim \nu_{me}$ ) this would correspond to the long-wavelength approximation ( $\nu \ll \nu_{me}$ ) for which the dispersion law of the quasiaoustic mode should be quadratic.<sup>8</sup> However, a change in the frequency of sound by a factor of 5 (50–250 MHz) in our experiments failed to reveal any frequency dependence of the quantity  $\Delta s/s$ . Hence we concluded that the magnetoelastic gaps

$\nu_{me 1,2}$  corresponding to the interaction of the soft mode with phonons were considerably smaller than the observed energy gaps  $\nu_{1,2}$ . In other words, the magnetoelastic interaction of magnons and phonons was "masked" by some other gap formation mechanism. In this case the sound frequencies could correspond to values of  $k$  which were far too large so that our results were outside the limits of the quadratic part of the dispersion curve. As pointed out earlier, this is pronounced in the limit  $k \rightarrow 0$  (Ref. 8). One should also bear in mind some shift of the attenuation maxima relative to the phase transition points ( $\Delta T \sim 0.15$  K near  $T_2$ —see Fig. 6), corresponding to extrema of the velocity of sound of the magnetic resonance frequencies. Such a shift is not predicted by the usual theory of the magnetoelastic interaction<sup>8</sup> and could be the result of activation of additional mechanisms governing the characteristic features of the propagation of an active acoustic mode near the reorientation points.

The insufficiently exact approach to the phase transition points when an external parameter (in our case, temperature) was varied was an obvious factor contributing to errors, so that it had to be allowed for in the measurements. It should be stressed that the traces in Fig. 6 represent continuously recorded functions of temperature (the discrete steps were simply due to the digital analysis and equal  $\sim 3 \times 10^{-3}$  K) when heating or cooling was slow. Therefore, the transition points could not have been missed. As pointed out above, the factors which could significantly suppress the anomalies of the velocity of the active mode could be an inhomogeneity of a sample and of the temperature field in it, as well as the presence of domains. It would be quite difficult to determine experimentally the influence of such factors. We suppressed domains by attempting to induce a single-domain state by applying a magnetic field parallel to the  $c$  axis (as shown earlier, the  $H \parallel c$  field shifted the  $\Gamma_{24} - \Gamma_4$  transition to lower temperatures, but did not affect the soft mode gap). It was found that the magnetic field did indeed enhance the magnetoelastic interaction (Fig. 7). The anomalies of  $\Delta s/s$  and  $\Delta\alpha$  varied reversibly and approximately linearly with the field, reaching (at  $H \sim 35$  kOe) values which were almost an order of magnitude higher than the corresponding values in zero field. Although the actual reason for the enhancement of the magnetoelastic coupling due to the application of the field was not clear, this enhancement made it possible to exclude immediately sample inhomogeneity as a possible reason for the constraints on the magnetoelastic anomalies of sound, because the field could not alter structural or temperature inhomogeneities. The reversibility and the absence of saturation of the anomalies on increase in the field were also evidence that domains played an unimportant role. The application of an additional static field normal to the axis of the superconducting solenoid allowed us to establish that in the main experiments (Fig. 7) the angle between the  $c$  axis and the direction of the solenoid field was  $0.5^\circ$ . In the case of the exact  $H \parallel c$  orientation the magnitude of the effect, i.e., the changes  $\Delta s/s$  and  $\Delta\alpha$ , increased by a further  $\sim 25\%$ . It should be pointed out that on application of a magnetic field to  $\text{ErFeO}_3$  the changes in the velocity of the active mode became smaller.<sup>18</sup> Therefore, along the  $\Gamma_{24} - \Gamma_4$  phase transition line the energy gap in the magnon spectrum was constant and the change in the velocity of transverse acoustic waves characterized by

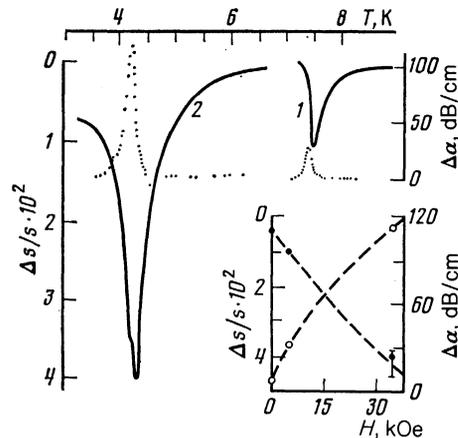


FIG. 7. Temperature dependences of the relative changes in the velocity  $\Delta s/s$  (continuous curves) and attenuation  $\Delta\alpha$  (points) of transverse sound ( $k \parallel c$ ,  $\epsilon \parallel a$ ) in magnetic fields: 1) 5 kOe; 2) 35 kOe. The field was inclined relative to the  $c$  axis in the  $ac$  plane by an angle  $\sim 0.5^\circ$ . The sample was a disk with the normal along the  $c$  axis and the frequency of sound was 54.3 MHz. The inset shows the field dependence of  $\Delta s/s_{\max}$  and  $\Delta\alpha_{\max}$  for a field inclination between  $H$  and  $c$ .

$\epsilon \parallel a$  increased with the field. A change in the magnetoelastic coupling should have been accompanied by a corresponding change in the magnetoelastic gap  $\nu_{me}$ .

This apparently contradictory (from the theoretical point of view) result can be explained by going back to the earlier hypothesis that the gaps due to the magnetoelastic interaction  $\nu_{me 1,2}$  were much smaller than the energy gaps  $\nu_1$  and  $\nu_2$  actually observed experimentally. In this case the change in  $\nu_{me}$  under the influence of the field should have practically no effect on the change in the total gap at the phase transition point and some other mechanism must have dominated the formation of the gap at the orientational phase transition in  $\text{YbFeO}_3$ .

The magnetoelastic interaction in the case of the  $\Gamma_{24} - \Gamma_4$  orientational phase transition was subjected to a detailed theoretical analysis in Ref. 21 in the specific case of  $\text{ErFeO}_3$ . However, this analysis ignored the role of the rare-earth subsystem, whereas in the case of  $\text{YbFeO}_3$  this subsystem plays the decisive role in the formation of a soft mode interacting with an active acoustic mode. Moreover, since the moment induced at the  $\text{Yb}^{3+}$  ions increases as a result of cooling, this is accompanied by an increase in the role of the dipole-dipole interactions which influence the magnetoelastic coupling. Therefore, investigations of various aspects of the magnetoelastic interaction in the case of the orientational phase transition in  $\text{YbFeO}_3$  will require a special theoretical analysis allowing for the results obtained in the present study.

We conclude by mentioning one further experimental observation which could not be explained. When hf sound was excited, the temperature dependences of the attenuation of the active mode exhibited sharp (compared with similar behavior at 50 MHz) additional peaks  $\Delta\alpha(T)$  near  $T_1$  and  $T_2$ . These anomalies were exhibited by all the samples and were affected by strong magnetization reversal or thermal cycling involved in experiments carried out at liquid helium temperatures.

Experimental curves showed no hysteresis. The influ-

ence of possible nonlinear effects was avoided by making measurements at the lowest possible power transmitted by a sample. Control experiments carried out with sound amplitude twice as large as the working amplitude failed to reveal any nonlinear effects (including harmonics). Therefore, they should be also excluded from among the possible factors that could limit the magnetoelastic interaction in  $\text{YbFeO}_3$ .

## 5. CONCLUSIONS

We carried out a comprehensive investigation of the dynamics of an orientational phase transition in  $\text{YbFeO}_3$ . A comparison of the results obtained in the submillimeter<sup>5</sup> and millimeter (Ref. 9 and the present study) ranges of wavelengths was used to consider the nature of the observed soft mode. It was concluded that it was entirely or partly due to vibrations of the rare-earth magnetic subsystem. An important result was determination of the energy gaps of the soft mode at the limits of the reorientation transition by the direct magnetic resonance method. A study was made of the anomalies in the propagation of acoustic waves in the vicinity of the orientational phase transition. It was found that an external magnetic field, oriented exactly along the easy axis, enhanced the magnetoelastic interaction but did not alter the energy gaps. This observation, together with the small changes in the velocity of the active acoustic mode, led to the conclusion that the magnetoelastic gap due to the interaction of phonons with the soft mode was considerably less than the energy gaps found experimentally. Consequently, the magnetoelastic mechanism was not the only one which governed the gaps in the magnon spectrum at the orientational phase transition points, and the problem of the origin of the energy gaps  $\text{YbFeO}_3$  requires further study. The development of a suitable theoretical approach should make it possible to use  $\text{YbFeO}_3$  in determination of the contribution of various mechanisms to the formation of the observed gaps. It would be of fundamental importance to allow for the specific nature of the dynamics of the orientational phase transition in  $\text{YbFeO}_3$  manifested by softening of the rare-earth mode. Since for various reasons it is not possible to measure the energy gaps in the case of high-temperature orientational phase transitions, the results obtained for yt-

terbium orthoferrite would then provide unique data.

The authors are deeply grateful to I. M. Vitebskiĭ, A. A. Mukhin, A. A. Stepanov, and D. A. Yablonskiĭ for valuable discussions of the results.

- <sup>1</sup>K. P. Belov, A. K. Zvezdin, A. M. Kadomtseva, and R. Z. Levitin, *Oriental Transitions in Rare-Earth Magnetic Materials* [in Russian], Nauka, Moscow (1979).
- <sup>2</sup>Yu. P. Cherenkov, V. P. Plakhtii, and A. V. Kovalev, Preprint No. 897 [in Russian], Institute of Nuclear Physics, Leningrad (1983).
- <sup>3</sup>G. V. Kozlov, S. P. Lebedev, A. A. Mukhin, *et al.*, Proc. Twenty-Fourth All-Union Conf. on Low Temperature Physics, Tbilisi, 1986 [in Russian], Part 3, p. 28.
- <sup>4</sup>N. Koshizuka and K. Hayashi, *J. Magn. Magn. Mater.* **31-34**, 569 (1983).
- <sup>5</sup>K. B. Aring and A. J. Sievers, *J. Appl. Phys.* **41**, 1197 (1970).
- <sup>6</sup>S. Venugopalan, M. Dutta, A. K. Ramdas, and J. P. Remeika, *Phys. Rev. B* **31**, 1490 (1985).
- <sup>7</sup>I. M. Vitebskiĭ, N. K. Dan'shin, N. M. Kovtun, and M. A. Sdvizhkov, *Zh. Eksp. Teor. Fiz.* **90**, 1118 (1986) [*Sov. Phys. JETP* **63**, 652 (1986)].
- <sup>8</sup>I. E. Dikshstein, E. A. Turov, and V. G. Shavrov, *Dynamic and Kinetic Properties of Magnetic Materials* [in Russian], Nauka, Moscow (1986).
- <sup>9</sup>N. K. Dan'shin, G. G. Kramarchuk, and M. A. Sdvizhkov, *Pis'ma Zh. Eksp. Teor. Fiz.* **44**, 85 (1986) [*JETP Lett.* **44**, 107 (1986)].
- <sup>10</sup>V. D. Fil', P. A. Bezuglyĭ, E. A. Masalitin, and V. I. Denisenko, *Prib. Tekh. Eksp. No. 3*, 210 (1973).
- <sup>11</sup>I. M. Vitebskiĭ, S. V. Zherlitsyn, A. I. Zvyagin, A. A. Stepanov, and V. D. Fil', *Fiz. Tverd. Temp.* **12**, 1108 (1986) [*Sov. J. Low Temp. Phys.* **12**, 626 (1986)].
- <sup>12</sup>V. G. Bar'yakhtar, I. M. Vitebskiĭ, and D. A. Yablonskiĭ, *Zh. Eksp. Teor. Fiz.* **76**, 1381 (1979) [*Sov. Phys. JETP* **49**, 703 (1979)].
- <sup>13</sup>V. G. Bar'yakhtar, I. M. Vitebskiĭ, Yu. G. Pashkevich, and V. L. Sobolev, *Fiz. Tverd. Tela (Leningrad)* **26**, 1786 (1984) [*Sov. Phys. Solid State* **26**, 1080 (1984)].
- <sup>14</sup>F. B. Hagedorn, E. M. Gyorgy, R. C. Le Craw, J. C. Hensel, and J. P. Remeika, *Phys. Rev. Lett.* **21**, 364 (1968).
- <sup>15</sup>V. I. Ozhogin, V. G. Shapiro, K. G. Gurtovoi, E. A. Galst'yan, and A. Ya. Chervonenkis, *Zh. Eksp. Teor. Fiz.* **62**, 2221 (1972) [*Sov. Phys. JETP* **35**, 1162 (1972)].
- <sup>16</sup>A. M. Balbashov, A. G. Berezin, Yu. M. Gufan, G. S. Kolyadko, P. Yu. Marchukov, I. V. Nikolaev, and E. G. Rudashevskii, *Pis'ma Zh. Eksp. Teor. Fiz.* **41**, 391 (1985) [*JETP Lett.* **41**, 479 (1985)].
- <sup>17</sup>G. Gorodetsky and B. Lüthi, *Phys. Rev. B* **2**, 3688 (1970).
- <sup>18</sup>G. Gorodetsky, V. Lüthi, and T. J. Moran, *Int. J. Magn.* **1**, 295 (1971).
- <sup>19</sup>G. Gorodetsky, S. Shaft, and B. M. Wanklyn, *Phys. Rev. B* **14**, 2051 (1976).
- <sup>20</sup>A. N. Grishmanovskii, V. V. Lemanov, G. A. Smolenskii, A. M. Balbashov, and A. Ya. Chervonenkis, *Fiz. Tverd. Tela (Leningrad)* **16**, 1426 (1974) [*Sov. Phys. Solid State* **16**, 916 (1974)].
- <sup>21</sup>I. E. Dikshstein, V. V. Tarasenko, and V. G. Shavrov, *Fiz. Tverd. Tela (Leningrad)* **19**, 1107 (1977) [*Sov. Phys. Solid State* **19**, 644 (1977)].

Translated by A. Tybulewicz