

# The role of precession and longitudinal oscillations of the magnetization in the spin reorientation dynamics of $\text{Fe}_3\text{BO}_6$

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We have carried out an experimental study of the soft magnetic-resonant mode in  $\text{Fe}_3\text{BO}_6$  in the vicinity of the orientational transitions induced by the magnetic field  $H$ . The results are interpreted in terms of ideas about the energy gap at the transition point as a result of a dynamic interaction of various oscillatory subsystems of the magnet. We find that in  $\text{Fe}_3\text{BO}_6$  the longitudinal oscillations of the magnetization contributes to the magnitude of the gap even in the limit  $H \rightarrow 0$ , which does not follow from the existing theory. © 1996 American Institute of Physics. [S1063-7761(96)02102-2]

## 1. INTRODUCTION

At present a number of experimental papers (e.g., Refs. 1 and 2) have appeared, from which it follows that the contributions of precession and longitudinal oscillations of the magnetization to the reorientation dynamics must always be considered as coexisting and competing. This conclusion, natural for experiments at temperatures  $T > 0$  (i.e., under real conditions), did not arise until the spin-wave model<sup>3,4</sup> and thermodynamic model,<sup>5,6</sup> each describing the dynamics of spin reorientation in a different way, were confirmed by experiments carried out under substantially different conditions. These conditions were dictated by the properties of the specific magnets used in the measurements. And although in almost all cases these were rare-earth (RE) orthoferrites with practically identical iron ordering temperatures  $T_N = 620\text{--}640$  K, the difference of their static characteristics in the reorientation region turned out to be decisive. Comparison of the results of corresponding experiments revealed that the experiments independently confirming the two models were carried out in different temperature and magnetic field ranges. As a result, it turns out that the spin-wave model provides a quite good description of the experimentally observed dynamics only of spontaneous transitions or those induced by a relatively small field ( $H < 10$  kOe at reorientation temperatures  $T_{IR} \ll T_N$  (Refs. 1 and 2), whereas the thermodynamic model was first confirmed (in  $\text{YFeO}_3$ ) for  $T_{IR}/T_N > 0.3\text{--}0.6$  in fields  $H > 60$  kOe (Ref. 5). This latter circumstance allows us to treat the important role of the longitudinal susceptibility as a strong-field effect. However, our analysis of all of the experiments which have examined the dynamics of induced reorientation shows that the manifestation of the longitudinal oscillations in the resonant properties is independent of the magnitude of  $H$ , whereas the absolute value of the growth of the gap in the presence of a field predicted by the theory<sup>5,6</sup> unconditionally grows with increasing field strength. The redistribution of the contributions to the dynamics from precession and the longitudinal oscillations correlates instead with the magnitude of the relative reorientation temperature  $\tau_{IR} = T_{IR}/T_N$ . A characteristic parameter for magnets where spontaneous reorientation is real-

ized may be the ratio of the spontaneous spin reorientation temperature to the ordering temperature of the corresponding spin system (the relative spontaneous reorientation temperature)  $\tau_{SR} = T_{SR}/T_N$ . In the instance of  $\text{Fe}_3\text{BO}_6$  we will show that the longitudinal oscillations at the relatively large value of  $\tau_{SR}$  intrinsic to this compound give an appreciable contribution to the dynamics even as  $H \rightarrow 0$ .

## 2. BACKGROUND INFORMATION ABOUT $\text{Fe}_3\text{BO}_6$ , AND THE EXPERIMENTAL TECHNIQUE

The compound  $\text{Fe}_3\text{BO}_6$ , isomorphic to rare-earth orthoferrites for  $T < T_N = 508$  K, is a weak orthorhombic ferromagnet which undergoes the reorientation  $\Gamma_2(F_x, G_z) - \Gamma_4(G_x, F_z)$  at  $T = T_{SR} = 415$  K. Here  $F_{x,z}, G_{x,z}$  are the components of the ferromagnetism  $\mathbf{F} = \mathbf{M}_1 + \mathbf{M}_2$  and antiferromagnetism  $\mathbf{G} = \mathbf{M}_1 - \mathbf{M}_2$  vectors;  $\mathbf{M}_1$  and  $\mathbf{M}_2$  are the magnetizations of the iron sublattices. In contrast to rare-earth orthoferrites, reorientation in  $\text{Fe}_3\text{BO}_6$  takes place via a first-order phase transition, i.e., without the formation of the canted phase  $\Gamma_{24}(F_{xz}, G_{xz})$  (Ref. 7). This latter phase can be brought into being only by a field. In what follows, we will be interested in two cases:  $\mathbf{H} \parallel \mathbf{c}$  for  $T < T_{SR}$  and  $\mathbf{H} \parallel \mathbf{a}$  for  $T > T_{SR}$ , which correspond to the second-order phase transitions  $\Gamma_{24} - \Gamma_4$  and  $\Gamma_{24} - \Gamma_2$  at the completion points of the induced reorientation. It was specifically in transitions of this type that these theoretical models were tested in experiments with rare-earth orthoferrites.<sup>1,2,5,6</sup> It is of fundamental importance here to have the field accurately aligned relative to the axes. Such an accurate alignment can be achieved by the method described in Ref. 5 and is based on the strong sensitivity of the soft-mode spectrum of the antiferromagnetic resonance to the deviation of the field from the crystalline axes in the  $ac$  plane. In our experiments an accuracy of 10–20 arcmin sufficed.

The  $\text{Fe}_3\text{BO}_6$  sample about 2.5 mm<sup>3</sup> in volume had a shape similar to that depicted in the inset to Fig. 1. It was cemented to the  $ac$  plane at the center of a piston closing off a rectangular waveguide with SHF oscillations of the type  $H_{10}$ . The optimal mutual orientation of the ferromagnetism vector  $\mathbf{F}$  and the magnetic vector of the SHF field,  $\mathbf{h}$ , for

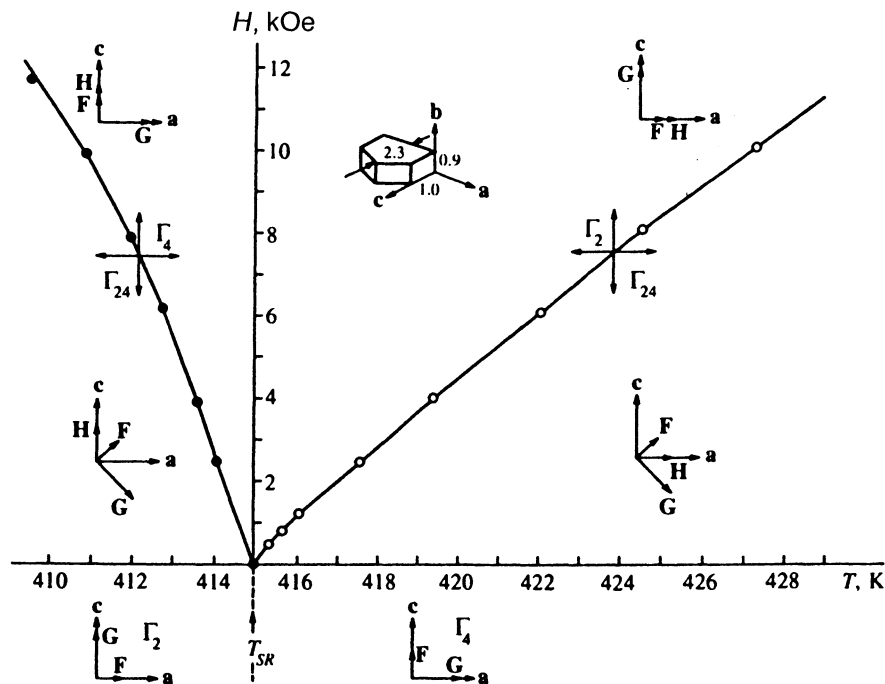


FIG. 1.  $H-T$  phase diagrams and structures of the field-induced phase transitions in  $\text{Fe}_3\text{BO}_6$ :  $\Gamma_{24}-\Gamma_4$  in a field  $\mathbf{H}\parallel\mathbf{c}$  (●) and  $\Gamma_{24}-\Gamma_2$  in a field  $\mathbf{H}\parallel\mathbf{a}$  (○). The structure of the spontaneous transition  $\Gamma_2-\Gamma_4$  is shown below;  $T_{\text{SR}}$  is the spontaneous transition temperature. The insert shows the form and characteristic linear dimensions of the sample in mm.

recording the soft ferromagnetic mode is  $\mathbf{F}\perp\mathbf{h}$ . However, in order to be able to make measurements at both transition points  $\Gamma_{24}-\Gamma_4$  and  $\Gamma_{24}-\Gamma_2$  with one mounting of the sample, the vector  $\mathbf{h}$  was set at an angle of  $45^\circ$  to the  $\mathbf{a}$  and  $\mathbf{c}$  axes.

The measurements were performed with the help of a direct-amplification spectrometer in the reflected-power regime. This made it possible to record resonant absorption in two different ways: by scanning the temperature for  $H=\text{const}$  and by scanning the field for  $T=\text{const}$ .

Our main results were obtained by scanning the temperature while holding the field fixed and modulated at a frequency of 39 Hz. In this regime the temperature derivative of the absorption signal was recorded in the range 12–26 GHz. From these recordings we reconstructed the temperature dependence of the magnetoresonant frequencies for each given value of  $H=\text{const}$ . Using the modulation technique allowed us to increase the sensitivity and resolving power of the spectrometer by more than an order of magnitude in comparison with the levels achieved in Ref. 8.

### 3. RESULTS AND DISCUSSION

Figure 1 shows the low-field portion of the  $H-T$  phase diagram of  $\text{Fe}_3\text{BO}_6$  corresponding to the completion points of the reorientations  $\Gamma_{24}-\Gamma_4$  in a field  $\mathbf{H}\parallel\mathbf{c}$  and  $\Gamma_{24}-\Gamma_2$  for  $\mathbf{H}\parallel\mathbf{a}$ , and also the structure of these transitions. This diagram was obtained from the high-frequency measurement data presented in Fig. 2. Each point in Fig. 1 corresponds to the position of the minimum frequency of the soft magnetoresonant mode with respect to temperature and field. The vertical and horizontal arrows indicate the two ways of crossing the phase boundaries in the  $H-T$  phase diagram when recording the absorption signals, i.e., scanning the field and scanning the temperature. The displayed diagram was also obtained in another way. In this technique, each data point in the diagram

is obtained at a given transition field  $H=\text{const}$ . The transition temperature is then determined from the position of the absorption peaks from the wings of the resonance lines. This position, as can be seen from Fig. 2, is independent of the frequency within the limits of measurement error, but is different for each value of  $H$ .

The phase diagram of  $\text{Fe}_3\text{BO}_6$  shown here differs from phase diagrams typical of orthoferrites in the vicinity of the

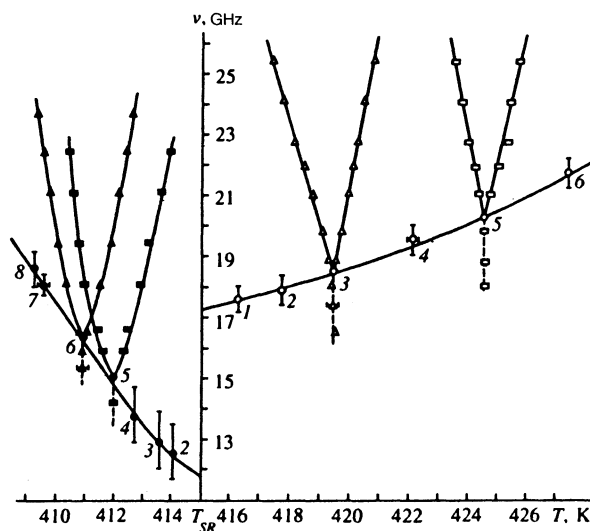


FIG. 2. Temperature dependence of the frequencies of the soft mode and the energy gaps at the reorientation completion points in  $\text{Fe}_3\text{BO}_6$ : the characteristic dependence of the soft mode frequencies in a field  $\mathbf{H}\parallel\mathbf{c}$  with field strength 8 kOe (■), 10 kOe (▲), and in a field  $\mathbf{H}\parallel\mathbf{a}$  with field strength 4 kOe (△), 8 kOe (□); the dependence of the gap at the spin reorientation ( $\Gamma_{24}-\Gamma_4$ ) completion point in fields  $\mathbf{H}\parallel\mathbf{c}$  (●) and  $\mathbf{H}\parallel\mathbf{a}$  (○). Each point in the temperature dependence of the gaps corresponds to the following field values in kOe: 1) 1.25, 2) 2.5, 3) 4, 4) 6, 5) 8, 6) 10, 7) 12, and 8) 12.5. The vertical dashed lines join points corresponding to the peaks of the absorption from the sides of the resonance lines.

$\Gamma_2-\Gamma_4$  transitions in that here the phase boundaries do not intersect. The fact that the curves of the second-order phase transitions converge to one point at  $H=0, T=T_{SR}$ , where the first-order phase transition  $\Gamma_2-\Gamma_4$  occurs, is unique. This means that the spontaneous transition  $\Gamma_2-\Gamma_4$  (its structure is shown in the lower part of Fig. 1) can simultaneously be considered as a second-order phase transition in which the starting and ending points,  $T_1$  and  $T_2$ , of the reorientation coincide. Note that in rare-earth orthoferrites the width of the region of the canted phase  $\Gamma_{24}$  in the spontaneous transitions,  $\Delta T=T_1-T_2$  usually lies within the limits of a few degrees to a few tens of degrees. And since the spontaneous transitions  $\Gamma_{24}-\Gamma_4$  and  $\Gamma_{24}-\Gamma_2$  take place in them via second-order phase transitions,  $\Delta T>0$  is always true. Here we in fact have the situation, where  $\Delta T=0$ . And this, in turn, allows us to assume that the spontaneous transition  $\Gamma_2-\Gamma_4$  in  $\text{Fe}_3\text{BO}_6$  is close to a second-order phase transition.

This same point of view, based on the results of high-frequency<sup>8</sup> and ultrasound<sup>9</sup> measurements, has already been expressed before. In this regard, we call attention to the calculation of the magnetoresonant frequencies in  $\text{Fe}_3\text{BO}_6$  (Ref. 10) from which it follows that at the spontaneous transition point  $\Gamma_2-\Gamma_4$  there is no energy gap and the frequency jump in the soft-mode spectrum characteristic of a first-order phase transition is absent. But in the experimental studies of Ref. 8 a significant energy gap ( $\approx 17.5$  GHz) was detected although no frequency jump was recorded. This latter result, as is now clear, was a result of insufficient resolving power of the technique used in the experiments,<sup>8</sup> and the frequency jump, as the results of our measurements show, exists. However, this is not reflected in the conclusion that here we are dealing with a first-order phase transition close to a second-order phase transition. We draw such detailed attention to this fact because all of the above theoretical models describe the dynamics of magnetic transformations at second-order transition points. Only at such transitions is the “energy background” removed that is associated with the magnetic anisotropy, whose external manifestation is the frequency gap in the spectrum of the soft magnetoresonant mode (it is in general impossible to achieve this at first-order transition points). Thus, conditions are created for observing the fine aspects of the dynamic interaction of different vibrational subsystems of the magnet: the spin subsystem (precession and longitudinal oscillations of the magnetizations of the sublattices), the elastic subsystem the dipole subsystem, paramagnetic, rare-earth, etc. And although in classical terms<sup>11</sup> the soft-mode frequency here should vanish, in fact significant energy gaps are always observed which are a result of the indicated interaction. In what follows we will call them starting gaps if the corresponding second-order phase transition is spontaneous.

Figure 2 displays examples of the reconstructed temperature dependence of the soft-mode frequency in magnetic fields of different magnitude and orientation. The minimum frequency of each such dependence is the energy gap  $\nu_{IR}$  at the completion point of the induced spin reorientation  $\Gamma_{24}-\Gamma_4$ , respectively  $\Gamma_{24}-\Gamma_2$ , in  $\text{Fe}_3\text{BO}_6$  for  $\mathbf{H}\parallel\mathbf{c}$ , respectively  $\mathbf{H}\parallel\mathbf{a}$ . From the results of these measurements we have reconstructed the temperature dependence of the gap  $\nu_{IR}$ ,

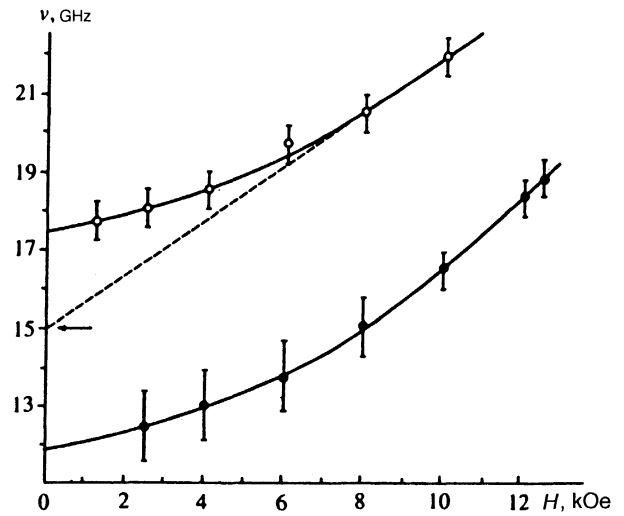


FIG. 3. Field dependence of the gaps at the spin reorientation completion points in  $\text{Fe}_3\text{BO}_6$ :  $\Gamma_{24}-\Gamma_4$  (●) and  $\Gamma_{24}-\Gamma_2$  (○), respectively, in fields  $\mathbf{H}\parallel\mathbf{c}$  and  $\mathbf{H}\parallel\mathbf{a}$ . The arrow indicates the value of the gap 15±1.5 GHz at the spontaneous transition point of the transition  $\Gamma_2-\Gamma_4$ , obtained by extrapolating its field dependence measured in Ref. 12 from the region  $H=40-80$  kOe. The dashed line is the linear extrapolation of the field dependence derived here of the gap on the  $\Gamma_{24}-\Gamma_2$  transition line out of the region  $H>8$  kOe.

which is also shown in Fig. 2. Their extrapolation to the spontaneous transition point ( $\Gamma_2-\Gamma_4$ ) from the low-temperature side gives  $\nu_{SR}=11.8\pm 1.5$  GHz, and from the high-temperature side,  $\nu_{SR}=17.5\pm 0.5$  GHz. Note that the absolute value of the frequency jump on the phase boundary substantially exceeds the error in determining the gaps.

Since the  $H-T$  phase diagram places a definite value of  $H$  in correspondence with each value of the temperature, Fig. 2 in fact reflects the temperature–field dependence of the energy gaps. Thanks to this, these results can also be displayed in a temperature–field plot. This is also of interest because high-field measurement data of  $\text{Fe}_3\text{BO}_6$  in the submillimeter wavelength range, which we will compare our results with below, were published in this form.<sup>12</sup> The field (or in the above sense, temperature–field) dependences of the gaps which we obtained in our experiments are plotted in Fig. 3.

Let us single out the most important results of these measurements.

1. In contrast to all of the previous work, which is cited above, we have discovered that the temperature–field dependence of the energy gaps at the curves of the second-order phase transitions approach the point  $T=T_{SR}, H_{IR}=0$  with nonzero derivatives  $\partial\nu_{IR}/\partial T$  and  $\partial\nu_{IR}/\partial H$ .

2. For all temperatures  $\partial\nu_{IR}/\partial H>0$  holds, whereas  $\partial\nu_{IR}/\partial T<0$  holds in the region  $T<T_{SR}$  and  $\partial\nu_{IR}/\partial T>0$  holds in the region  $T>T_{SR}$ .

3. The temperature–field dependence of the energy gaps at temperatures higher and lower than  $T_{SR}$  is substantially different. Extrapolating it to  $H=0, T=T_{SR}$  gives different values of the gaps  $\nu_{IR}$ , i.e., there is a frequency jump at the spontaneous transition  $\Gamma_2-\Gamma_4$ , and both gaps  $\nu_{SR}$  have significant absolute values.

Let us compare these results with the results of other experiments and presently existing theory.

We comment first on the last of the results noted above. The frequency jump detected at the spontaneous transition point has no direct connection with the aim of the present paper, but it is necessary to take it into account in the interpretation of the experimental data. This effect is unexpected since not only does it not follow from the calculations of the magnetoresonance spectrum of  $\text{Fe}_3\text{BO}_6$ , but its absence is specially grounded in theory.<sup>10</sup> This, even though, as is well known, such a frequency jump is a characteristic sign of a first-order phase transition.

Since a magnetic field transforms a first-order phase transition into a second-order phase transition, according to existing ideas<sup>11</sup> the presence of a field should cause this frequency jump to disappear. What then is the meaning of its continued existence subsequent to such a transformation of the transition in  $\text{Fe}_3\text{BO}_6$ ? It may be postulated that in a comparatively small field, i.e., under the conditions of an induced second-order phase transition, but near a spontaneous first-order phase transition, the former represents a peculiar memory of the latter. Thus, the anisotropy of the energy gap in a magnetic field is induced by the proximity of the first-order phase transition. This influence can be explained by the fact that the spontaneous first-order phase transition in  $\text{Fe}_3\text{BO}_6$  is similar in nature to a second-order phase transition. As a result, the second-order phase transition induced by a comparatively small field is unstable and the more so, the closer (in  $H$  and  $T$ ) it is to the spontaneous first-order phase transition.

Such an explanation is supported by an extrapolation of the temperature–field dependence of the gaps to the spontaneous transition point in the phase diagram from regions in  $T$  and  $H$  that are as far as possible from it. For the  $\Gamma_{24}-\Gamma_2$  transition we will use the results of our own measurements at  $H > 8$  kOe and  $T > 425$  K, extrapolation of which to zero field gives  $\nu_{SR} \approx 15$  GHz (see Fig. 3). For the  $\Gamma_{24}-\Gamma_4$  transition it is preferable to use the results of Ref. 12, in which the field dependence of the gap on the curve of this transition is reconstructed in the intervals  $H = 40-80$  kOe,  $T = 290-380$  K (for comparison, in our measurements we used the intervals  $H = 0-12.5$  kOe,  $T = 409-415$  K). Extrapolating these data to zero field gives practically the same value of the gap at the spontaneous transition point. Given the roughness of this approach, we may nevertheless note that the tendency toward a coincidence of the values of the gaps obtained in this way is logical since this extrapolation is carried out from regions where the second-order phase transition is already quite stable. Also for it the absence of a frequency jump at the phase boundary is a fundamental property. It is precisely this value of  $\nu_{SR}$  that corresponds to the definition of the concept of a starting gap. Its field gradient is  $\approx 0.7$  GHz/kOe (see the dashed line in Fig. 3), whereas extrapolation of the field dependence of the gaps to zero field from the region  $H = 1-3$  kOe for both transitions gives  $\partial\nu_{IR}/\partial H \approx 0.2$  GHz/kOe (the solid lines in this figure). In either case, we have  $\partial\nu_{IR}/\partial H > 0$  even at  $H = 0$ . But in fields of 10–12 kOe the increments in the starting gaps are already a few times greater than the maxi-

imum error in the measurements. So, to what extent do the results obtained here agree with existing theory?

In the thermodynamic approximation<sup>5,6</sup> we have the following first-order expression for the gap:

$$\nu_{IR} = \frac{\gamma}{2\pi} \sqrt{\frac{\chi_{\parallel}}{\chi_{\perp}}} H,$$

where  $\gamma$  is the gyromagnetic ratio, and  $\chi_{\parallel}$  and  $\chi_{\perp}$  are respectively the longitudinal and transverse susceptibilities (with respect to  $\mathbf{G}$ ). Modification of these calculations specifically for  $\text{Fe}_3\text{BO}_6$  does not undercut the main conclusion that follows from this expression, namely that as  $H \rightarrow 0$  the gap at the completion point of the induced reorientation should also vanish. Thus, neither the original nor the modified theoretical model in its presently existing form can explain the experimentally observed dynamics of orientational transitions in  $\text{Fe}_3\text{BO}_6$  as  $H \rightarrow 0$ . However, their fundamental property,  $\partial\nu_{IR}/\partial H > 0$  at  $H_{IR} > 0$ , as evidence of the contribution to the dynamics of the longitudinal oscillations of the magnetization and the relaxation, is not in doubt and is an admissible test in the identification of this contribution in the experiments. As has been shown in a number of previous experiments with rare-earth orthoferrites  $\text{RFeO}_3$  ( $\text{R: Yb, Tm, Er}$ ), for which the relative reorientation temperatures  $\tau_{SR}$  are comparatively small, up to 10 kOe the magnetic field has no effect on the magnitude of the gaps.<sup>1,2</sup> The result  $\partial\nu_{IR}/\partial H = 0$  obtained in these experiments is interpreted as evidence that the contribution of the longitudinal oscillations is unimportant in the presence of the large contribution of the precession, taken into account in the spin-wave approximation.<sup>3,4</sup> The significant starting gaps detected in these experiments at the points of the spontaneous orientational transitions, or those induced by a comparatively small field, are explained by the above-mentioned dynamical interaction between the various subsystems.

A different picture can be expected in  $\text{Fe}_3\text{BO}_6$ , where the relative spontaneous transition temperature  $\tau_{SR}$  is approximately equal to 0.8. As far as this parameter is concerned,  $\text{Fe}_3\text{BO}_6$  would seem to be better than other similar compounds for comparison with thermodynamic theory. The field dependence of the energy gaps in the region  $H = 40-80$  kOe, obtained in Ref. 12, at the corresponding relative temperatures  $\tau_{IR} = 0.5-0.7$ , is excellently described by this theory. As was already noted, it is almost linear, which allows us to extrapolate it to  $H = 0$ . The first of these results, in our opinion, is actually due to the large value of  $\tau_{SR}$ , which, in turn, is the cause of significant value of the factor  $\chi_{\parallel}/\chi_{\perp}$  in the expression for the gap. From the temperature dependence of  $\chi_{\parallel}/\chi_{\perp}$  obtained in Ref. 6 for the transition  $\Gamma_{24}-\Gamma_2$  in the iron subsystem in  $\text{YFeO}_3$  it is possible to estimate the value of this factor for  $\text{Fe}_3\text{BO}_6$ . As a result, for  $T = T_{SR}$  we obtain  $\chi_{\parallel}/\chi_{\perp} \approx 0.7$ . The correctness of turning to the results of Ref. 6 is justified by the fact that in  $\text{YFeO}_3$ , as in  $\text{Fe}_3\text{BO}_6$ , the entire spin dynamics is due only to the iron subsystem, and the structures of the induced orientational transitions in these compounds are absolutely identical.

The significant value of the ratio of susceptibilities  $\chi_{\parallel}/\chi_{\perp}$  in  $\text{Fe}_3\text{BO}_6$  even in zero field casts a different light on

the nature of the energy gap of the spontaneous transition observed here. In the previous experiments with rare-earth orthoferrites of ytterbium, thulium, and erbium, we were dealing with relatively low spontaneous transition temperatures,  $\tau_{SR}=0.01-0.15$ , and that means, with small values of  $\chi_{\parallel}/\chi_{\perp}$  (Refs. 1 and 2). For this reason the corresponding results are well described in the spin-wave approximation, which takes account only of precession while holding the magnitudes of the magnetizations of the sublattices fixed:  $|\mathbf{M}_1|=|\mathbf{M}_2|=\text{const}$ . If we follow the logic of the redistribution of the partial contributions of precession and the longitudinal oscillations as a function of  $\tau_{SR}$ , then we must assume that in  $\text{Fe}_3\text{BO}_6$  even the starting gap is to a significant extent formed as a result of the latter. An external sign of this is the nonzero value of the derivative  $\partial\nu_{IR}/\partial H$  at  $H=0$ . Increasing the field for  $\mathbf{H}\parallel\mathbf{c}$  or  $\mathbf{H}\parallel\mathbf{a}$  only increases the amplitude of this contribution. But in no case is the presence of an external magnetic field a necessary condition for the participation of longitudinal oscillations of the magnetization in the formation of the dynamics of the spin reorientation. And only in this sense is the partial contribution to the magnitude of the gap caused by them a strong-field effect. Of course, this does not follow from the existing thermodynamic model, in which not only do we have  $\nu_{IR}\rightarrow 0$ , but also  $\partial\nu_{IR}/\partial H\rightarrow 0$ , as  $H\rightarrow 0$ . The question of the specific relationship between the contributions of the precession and the longitudinal oscillations, of course, requires special calculations, especially when the expected contributions are comparable. For the time being, the theory gives a satisfactory answer only in the limiting cases: the spin-wave model for  $T_{IR}/T_N\ll 1$  ( $\chi_{\parallel}/\chi_{\perp}\rightarrow 0$ ), and the thermodynamic model, for  $T\rightarrow T_N$  ( $\chi_{\parallel}/\chi_{\perp}\rightarrow 1$ ).

In Ref. 12 the falloff of the gap in  $\text{Fe}_3\text{BO}_6$  and, in contrast, its growth in  $\text{YFeO}_3$  (Ref. 6), with increase of the temperature are explained by the different relative roles of  $\chi_{\parallel}$  and  $H$  in these two experiments. Actually, the real cause is the different structures of the transitions investigated in Refs. 12 and 6: in  $\text{Fe}_3\text{BO}_6$  the gap was measured at the  $\Gamma_{24}-\Gamma_4$  phase transition, for which  $\partial H/\partial T < 0$  holds (see Fig. 1), and in  $\text{YFeO}_3$ , at the transition  $\Gamma_{24}-\Gamma_2$ , for which  $\partial H/\partial T > 0$ .

This difference in the signs of the temperature derivatives of the gap can also be demonstrated in  $\text{Fe}_3\text{BO}_6$ . As can be seen from Fig. 2, the gap grows as the temperature is lowered in the region  $T < T_{SR}$ , where the  $\Gamma_{24}-\Gamma_4$  reorientation takes place, and as the temperature is raised in the region  $T > T_{SR}$ , where the  $\Gamma_{24}-\Gamma_2$  reorientation takes place. Here the role of the magnetic field in both cases is identical—in either temperature interval the gap grows with  $H$ .

The difference in sign of the temperature derivatives of the gaps in  $\text{Fe}_3\text{BO}_6$  for  $\mathbf{H}\parallel\mathbf{c}$  and  $\mathbf{H}\parallel\mathbf{a}$  has no fundamental significance within the context of this paper and only reflects the character of the low-field part of the phase diagram (Fig. 1). But since in a weak magnetic field the  $\Gamma_{24}-\Gamma_4$  and  $\Gamma_{24}-\Gamma_2$  transitions are found in adjacent temperature regions, and in a quantitative relationship, this should not introduce any substantial differences in their dynamics.

Let us turn to the field dependence shown in Fig. 3. Another aspect of the problem follows from the fact that

neither  $\nu_{IR}$  nor  $\partial\nu_{IR}/\partial H$  vanishes at  $H=0$ , and must be taken into account in the description of the dynamics actually observed in the majority of such compounds. It consists in the following. The original theory<sup>5</sup> was developed for  $\text{YFeO}_3$ , in which spontaneous orientational transitions do not occur and the resonance mode in zero field is softened only at the point  $T=T_N$ . Applied to  $\text{YFeO}_3$ , the theory is valid without restrictions. Its extension to  $\text{DyFeO}_3$  (Ref. 6) turns out to be possible only in the region of temperatures substantially in excess of  $T=T_{SR}=40$  K. Thus, in  $\text{DyFeO}_3$  as well as in  $\text{YFeO}_3$  over the entire range of working temperatures, where the role of the longitudinal oscillations becomes noticeable, the transition field is nonzero. The question arises of how to describe the situation in which  $H_{IR}$  vanishes not only at  $T=T_N$ , but at some intermediate point in the interval  $T=0-T_N$ . In compounds with small  $\tau_{SR}$  the contribution of the longitudinal oscillations to the magnitude of the gaps can be manifested only in a comparatively strong field with a corresponding growth of  $T_{IR}$ . This is the case, for example, in  $\text{DyFeO}_3$  (Ref. 6) in fields  $H=H_{IR}>40$  kOe at temperatures  $T=100-400$  K. In this situation, the contribution of the longitudinal oscillations to the dynamics of reorientation can indeed be treated as a strong-field effect. A new aspect in experiments with  $\text{Fe}_3\text{BO}_6$ , to which we have already drawn attention, is that here the spontaneous transition takes place at large  $\tau_{SR}$ , thanks to which even in a field  $H=H_{IR}=0$  the growth of the field  $\Delta\nu_{SR}$  is nonzero.

#### 4. CONCLUSION

From a comparison of the results of our experiment with all previous studies of the low-energy dynamics of orientational transitions, we can draw the following conclusions:

1. The ratio of the spontaneous transition temperature to the spin system ordering temperature  $\tau_{SR}=T_{SR}/T_N$ , which is unique for each specific magnetic, can serve as a qualitative parameter characterizing the ratio of the contributions to the dynamics due to precession and due to the longitudinal oscillations of the magnetization near the spontaneous transition points (i.e., in small field).

2. For a comparatively large value of this parameter, as is the case, for example, in  $\text{Fe}_3\text{BO}_6$ , the derivatives  $\partial\nu_{IR}/\partial T$  and  $\partial\nu_{IR}/\partial H$  can be nonzero even at  $H=0$ . This, in turn, means that at the spontaneous transition point ( $H=0$ ,  $T=T_{SR}$ ) the energy gap can be the result of an interaction of the elastic, dipole, and other oscillatory subsystems of the magnet not only with the precessional, but also with the longitudinal oscillations of the magnetization in the spin subsystem.

3. The starting gap grows with increase of a field of definite orientation as the temperature is either raised above or lowered below  $T_{SR}$ . The first of these dependences is characteristic of the  $\Gamma_{24}-\Gamma_2$  transition, and the second, of the  $\Gamma_{24}-\Gamma_4$  transition. In the latter case, the gap grows as the field increases with simultaneous decrease of the longitudinal susceptibility. Here, for the thermodynamic theory<sup>6</sup> the model situation obtains when the gap grows as a result of the increase of both the transition field and the ratio  $\chi_{\parallel}/\chi_{\perp}$ .

It is completely obvious that different signs of the derivative  $\partial v_{IR}/\partial T$  can be expected for the indicated transitions for the series of rare-earth orthoferrites: 1) for large  $\tau_{SR}$  (e.g., in  $\text{SmFeO}_3$ , where  $\tau_{SR} \approx 0.7$  holds even in a weak field, 2) if the quantity  $\tau_{SR}$  is very small, then in a strong enough magnetic field (e.g., in  $\text{YbFeO}_3$ ,  $\text{ErFeO}_3$ ,  $\text{TmFeO}_3$ ,  $\text{NdFeO}_3$ , etc.).

4. It thus follows that apart from the dependence on the structure of the transition, the thermodynamic model in its present form can adequately describe only the results of high-field experiments, where the main contribution in the expressions for the gap comes from the magnetic field  $H$ .

In the case of small fields and relatively high spontaneous reorientation temperatures, the contributions to the dynamics from precession and longitudinal oscillations of the magnetization can be comparable, and thus neither of the existing theories can separately claim to provide a quantitative description of the experiment. In the final account, some general approach should appear which correctly explains the dynamics for all fields and temperatures, and in which the spin-wave and thermodynamic approximations are limiting cases.

The first step in this direction was taken in Ref. 13, where the spectrum of the coupled magneto-acoustic oscillations was obtained in magnets with both precession and the longitudinal oscillations taken into account. The relevance of a modification of the existing theory is confirmed by the experiments reported in Ref. 14, in which a pronounced correlation was detected between the high-frequency and acoustic characteristics of the metamagnetic transition in  $\text{ErFeO}_3$ , where these characteristics were due to the redistribution of the indicated contributions.

5. The frequency jump at the first-order phase transition  $\Gamma_2 - \Gamma_4$  means that here the total anisotropy energy does not vanish and is different on the two sides of the transition (with respect to temperature). This latter circumstance is the cause of the corresponding anisotropy of the energy gaps in the vicinity of  $T_{SR}$ .

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