## Nature of the energy gap in the spin-wave spectrum during spin flop in a magnetic field

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The orthoferrites of yttrium and dysprosium are used for a study of the energy gap in the spectrum of the quasiferromagnetic mode of the antiferromagnetic resonance near the completion of the spin flop induced by a magnetic field, over a broad temperature range (78–400 K). It follows from calculations carried out free of the limitations imposed by the Landau-Lifshitz equation that none of the normal oscillations corresponding to modes of the antiferromagnetic resonance is an oscillation of the order parameter in the transition field. As a result, one observes a significant energy gap of exchange origin. The theory derived here can be used along with the results of a dynamic experiment to reconstruct the temperature dependence of  $\chi_{\parallel}/\chi_{\perp}$  for an orthorhombic antiferromagnet with a weak ferromagnetic moment.

At the completion of the spin-flop transition induced by a magnetic field applied along the antiferromagnetism axis (the a axis of the crystal) in yttrium and dysprosium orthoferrites (YGeO<sub>3</sub> and DyFeO<sub>3</sub>), the frequency of the quasiferromagnetic mode of the antiferromagnetic resonance (which had previously been assumed to be soft in the course of this transition) does not vanish,<sup>1,2</sup> despite the fact that this transition is a second-order phase transition.<sup>3</sup> The energy gap which has been observed is far larger than could be explained by a magnetostriction mechanism.<sup>4</sup> At room temperature for YGeO<sub>3</sub> and DyFeO<sub>3</sub>, for example, the gaps are 107 and 136 GHz, respectively, while the magnetoelastic gap is less than 15-20 GHz according to the data of Refs. 5 and 6 (a similar estimate was given in Ref. 7). It has been suggested that the energy gap is associated with nonconservation of the magnitude of the magnetic moment of the sublattices of the antiferromagnet (i.e., with a parallel susceptibility  $\chi_{\parallel}$ ). That mechanism explained the apparent discrepancy between the observed effect and a general dynamic theorem, according to which the frequency of one of the modes in the spin-wave spectrum must vanish upon a second-order phase transition.

It follows from calculations carried out on the equilibrium state of magnetic moments which undergo changes in magnitude that one of the antiferromagnetic-resonance (AFMR) modes becomes soft during a spin-flop transition induced by a magnetic field. The change which occurs in the magnetic structure, however, is of such a nature that this mode is relaxational in the dynamics.<sup>7</sup> The intensity estimated for the absorption lines of the relaxation mode from the width of AFMR lines is two orders of magnitude lower than the intensity of the ordinary AFMR modes over the entire range of external fields up to the flop field and thus was not seen in the experiments of Refs. 1 and 2. There are two points to be noted here, however. First, the experiments of Refs. 1 and 2 were performed only at room temperature, so it was necessary to carry out measurements over a wide temperature range in order to test the suggestion of Refs. 1 and 7 that a parallel susceptibility plays a role in the formation of a gap in the spin-wave spectrum during a second-order orientational phase transition. Second, it was necessary to establish the mechanism for the formation of the energy gap, i.e., to examine the relationship between the order parameter of the transition of interest and the normal coordinates of the oscillations of the magnetic subsystem of the antiferromagnet, not only in order to generate a prediction regarding the existence of the observed effect in substances with other symmetries but also to prove that this is an unambiguous consequence of the nonconservation of the magnitude of the magnetic moment of the sublattices during the motion.

In this paper we report an effort to solve these two problems: to experimentally study the AFMR spectra near the completion of the spin flop induced by a magnetic field applied along the **a** axis of an orthorhombic crystal, over a wide temperature range, and to identify the mechanism for the formation of the energy gap during the second-order phase transition.

## THEORY

In the calculations we use the thermodynamic potential of orthorhombic crystals<sup>8,1,7</sup>:

$$\Phi(\mathbf{M}, \mathbf{L}) = \Phi_{0}(\mathbf{L}^{2}) + \frac{1}{2}B\mathbf{M}^{2} + \frac{1}{2}D(\mathbf{ML})^{2} + d(M_{x}L_{z} - M_{z}L_{x}) + \frac{1}{2}a_{1}L_{x}^{2} + \frac{1}{2}a_{2}L_{y}^{2} + \frac{1}{2}a_{3}L_{z}^{2} + \frac{1}{4}a_{11}L_{x}^{4} + \frac{1}{4}a_{22}L_{y}^{4} + \frac{1}{4}a_{33}L_{z}^{4} + \frac{1}{2}a_{12}L_{x}^{2}L_{y}^{2} + \frac{1}{2}a_{13}L_{x}^{2}L_{z}^{2} + \frac{1}{2}a_{23}L_{y}^{2}L_{z}^{2} - \mathbf{MH}.$$
(1)

Expression (1) ignores the interaction of the rare earth and iron magnetic subsystems (the R-Fe interaction), since according to Refs. 1 and 7 the energy gap in the spin-wave spectrum at the point of the second-order phase transition results from exchange (determined by the parallel susceptibility of the magnetic moments of the iron sublattices), so it should be observed in a situation of this sort in any compound with similar symmetry, regardless of any additional assumptions regarding the interaction of the iron sublattices with the other subsystems.

Incorporating the parallel susceptibility  $\chi_{\parallel}$ , which is described by the parameter D in the thermodynamic potential, has the consequence that during a spin flop in the **ac** plane induced by a magnetic field **H**||**a** the vectors **M** and **L** are not perpendicular to each other:  $\mathbf{ML} \neq 0$ . Consequently, a systematic symmetry description lacks one of the conservation laws (**ML** = 0) which is a direct consequence of the

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use of the Landau-Lifshitz equations of motion for each sublattice of a two-sublattice antiferromagnet. A discrepancy of this sort indicates the need to analyze the symmetry in the equations of motion beyond the customary symmetry analysis. Otherwise, the theory may lead to some nonphysical results.

A complete incorporation of the symmetry leads to the following equations, in the exchange approximation<sup>9,10</sup>:

$$\dot{\mathbf{m}} = \gamma_{1} [\mathbf{M}_{0} \mathbf{H}_{M}] + \gamma_{2} [\mathbf{L}_{0} \mathbf{H}_{L}] + \tau_{1}^{-1} \mathbf{H}_{M},$$

$$\dot{\mathbf{l}} = \gamma_{2} [\mathbf{L}_{0} \mathbf{H}_{M}] + \gamma_{3} [\mathbf{M}_{0} \mathbf{H}_{L}] + \tau_{2}^{-1} \mathbf{H}_{L},$$
(2)

where  $\mathbf{m} = \Delta \mathbf{M} = \mathbf{M} - \mathbf{M}_0$ ,  $\mathbf{l} = \Delta \mathbf{L} = \mathbf{L} - \mathbf{L}_0$ ,  $\mathbf{M}_0$  and  $\mathbf{L}_0$ are the equilibrium ferromagnetism and antiferromagnetism vectors,  $\mathbf{M} = \mathbf{M}_1 + \mathbf{M}_2$ ,  $\mathbf{L} = \mathbf{M}_1 - \mathbf{M}_2$ ,  $\mathbf{M}_1$  and  $\mathbf{M}_2$  are the sublattice magnetizations,

$$\mathbf{H}_{\mathbf{M}} = \frac{\partial \Delta \Phi}{\partial \Delta \mathbf{M}}, \quad \mathbf{H}_{L} = \frac{\partial \Delta \Phi}{\partial \Delta \mathbf{L}}, \quad \Delta \Phi = \frac{1}{2} \sum_{i,j=1}^{N} \alpha_{ij'} \Delta X_{i'} \Delta X_{j'},$$

 $\mathbf{X}' = \{\mathbf{M}, \mathbf{L}\}\$  is a six-dimensional vector,  $\alpha'_{ij} = \partial^2 \Phi / \partial X'_i \partial X'_j$  is a stability matrix, and  $\tau_1$  and  $\tau_2$  are phenomenological relaxation parameters. Equations (2) become the Landau-Lifshitz equations when we set  $\gamma_1 = \gamma_2 = \gamma_3$  and  $\tau_1^{-1} = 0$ .

For our purposes, the weak-field approximation (fields much weaker than the spin-flip fields) is sufficient in deriving the theory. In this case we can adopt the model assumption  $L^2 = \text{const.}$  This condition alters the form of the equations of motion, since the relation  $L^2 = L_0^2$  necessarily implies  $\gamma_3 = 0$  (Ref. 7).

In the case of interest here,  $\mathbf{H} \| \mathbf{a}$ , it is convenient to switch to the variables  $\mathbf{M}$ ,  $\theta$ , and  $\varphi$  in the static and dynamic equations of the magnetic subsystem; here the angle  $\varphi$  specifies the position of the vector  $\mathbf{L}$  in the **ac** plane, and the angle  $\theta$  determines the excursion of the vector  $\mathbf{L}$  from this plane. When we use the condition  $\mathbf{L}^2 = L_0^2$ , we then find that we are left with only five unknown variables  $X_i$  in this problem:  $M_x$ ,  $M_y$ ,  $M_z$ ,  $\theta$ , and  $\varphi$ . The static changes in the structure during a flop in the **ac** plane involve the three variables  $M_x$ ,  $M_z$ , and  $\varphi$ .

Let us examine the increment in thermodynamic potential (1) which stems from treating small deviations of the vector **M** and **L** from their equilibrium values at  $H \ge H_f$  ( $H_f$ is the field at which the flop is completed):

$$2\Delta\Phi = \sum_{i,j=1}^{n} \alpha_{ij} \Delta X_i \Delta X_j = \alpha_{11} (\Delta M_x)^2 + \alpha_{22} (\Delta M_y)^2 + \alpha_{33} (\Delta M_z)^2 + \alpha_{44} (\Delta\theta)^2 + \alpha_{55} (\Delta\phi)^2 + 2\alpha_{35} \Delta M_z \Delta\phi, \quad (3)$$

where

$$\hat{\alpha} = \{ \alpha_{ij} \} = \partial^2 \Phi(\mathbf{M}, \theta, \varphi) / \partial X_i \partial X_j,$$

$$\alpha_{11} = \alpha_{22} = \chi_{\perp}^{-1}, \quad \alpha_{33} = \chi_{\parallel}^{-1}, \quad \alpha_{44} = \chi_{\perp} (\Delta_{2a} - \Delta_{1a} + HH_D),$$

$$\alpha_{55} = \chi_{\perp} [-\Delta_{1a} + (H + H_D) \alpha_{35}], \quad \alpha_{35} = \chi_{\perp} \chi_{\parallel}^{-1} (H + H_D) - H,$$

$$\chi_{\perp}^{-1} = B, \quad \chi_{\parallel}^{-1} = B + DL_0^2, \quad H_D = dL_0,$$

$$\Delta_{1a} = \chi_{\perp}^{-1} L_0^2 [a_3 - a_1 + (a_{33} - a_{13}) L_0^2],$$

$$\Delta_{2a} = \chi_{\perp}^{-1} L_0^2 [a_2 - a_1 + (a_{23} - a_{13}) L_0^2] + H_D^2.$$

To find an explicit expression for the order parameter as

a function of the variables characterizing the magnetic subsystem, we diagonalize  $\hat{\alpha}$ . We know<sup>11</sup> that a symmetric matrix can be put in diagonal form through an orthogonal transformation of the basis. In the new basis, of the eigenvectors  $\mathbf{Y}_i$ , the correction to the thermodynamic potential,  $\Delta \Phi$ , becomes

$$\Delta \Phi = \sum_{i=1}^{3} \lambda_i (\Delta \mathbf{Y}_i)^2, \qquad (4)$$

where  $\lambda_i$  are the eigenvalues of the matrix  $\hat{\alpha}$ , and  $\mathbf{Y}_i$  are the corresponding eigenvectors.

To find the  $\lambda_i$  we solve the characteristic equations

$$\alpha_{11} - \lambda)(\alpha_{22} - \lambda)(\alpha_{44} - \lambda)[\lambda^2 - \lambda(\alpha_{33} + \alpha_{55}) + \delta] = 0, \quad (5)$$

where  $\delta \equiv \alpha_{33}\alpha_{55} - \alpha_{35}^2$ . It is not difficult to see that  $\delta$  vanishes in the field  $H = H_f$ , and a single vanishing solution of Eq. (5) for  $\lambda$  arises here. Knowing the set of eigenvalues  $\lambda_i$ , we can determine the corresponding eigenvectors (in the space of the variables  $\mathbf{M}, \theta, \varphi$ ):

$$\lambda_{1} = \lambda_{2} = \chi_{\perp}^{-1}, \quad \lambda_{3} = \chi_{\parallel}^{-1} (1 + a^{2}), \quad \lambda_{4} = \alpha_{44}, \quad \lambda_{5} = 0,$$
  
$$\mathbf{Y}_{1} = M_{x}, \quad \mathbf{Y}_{2} = M_{y}, \quad \mathbf{Y}_{3} = M_{z} + a\varphi, \quad \mathbf{Y}_{4} = \theta, \quad \mathbf{Y}_{5} = aL_{0}^{-2}M_{z} - \varphi,$$
  
$$a = \chi_{\perp} (\beta H + H_{D}), \quad \beta = 1 - \chi_{\parallel} / \chi_{\perp}. \quad (6)$$

The physical meaning of the zero eigenvalue which appears at the point of the second-order phase transition is obvious: Small deviations in the direction of the eigenvector  $\mathbf{Y}_5$ , which corresponds to  $\lambda_5 = 0$ , yield no change in  $\Delta \Phi$ , so the system has no "stiffness" against small static displacements in this direction, and the vector  $\mathbf{Y}_5$  itself turns out to be the order parameter associated with the transition.

We now seek the normal coordinates of small oscillations of the magnetic subsystem (normal modes). For this purpose we solve equations of motion (2), using  $\gamma_3 = 0$ . Using the dynamic matrix<sup>9</sup>  $\hat{\gamma}$ , we can rewrite Eqs. (2) as

$$\Delta \dot{\mathbf{X}}' = \mathbf{\hat{\gamma}} \hat{\alpha}' \Delta \mathbf{X}'. \tag{7}$$

The normal coordinates are eigenvectors of the matrix  $\hat{v} = \hat{\gamma} \hat{\alpha}'$  which correspond to the eigenvalues, the AFMR frequencies. When the matrix  $\hat{v}$  is diagonalized, two pairs of complex-conjugate eigenvalues arise:  $i\omega_1^{(\pm)}$  and  $i\omega_2^{(\pm)}$ . These eigenvalues correspond to quasiferromagnetic and quasiantiferromagnetic modes of the AFMR. In addition, there is one real eigenvalue,  $\omega_R$ , which is a relaxation mode. It has been shown<sup>7</sup> that the frequency of the relaxation mode vanishes in the field  $H_f$  (a soft "relaxer"), while the frequencies of the ordinary magnetic modes are nonzero. The eigenvectors (normal coordinates)  $\mathbf{Z}_i$  corresponding to various eigenvalues of the matrix  $\hat{v}$  take the following form in the transition field (in the basis of the vectors  $\mathbf{Y}_i$ ):

It can be seen from (8) that at  $H = H_f$  the normal coordinate of the relaxation mode,  $Z_R$ , is the same as the order parameter of the transition,  $Y_5$ . It is for this reason that the relaxation mode is soft. With regard to the two other modes (quasiferromagnetic and quasiantiferromagnetic) on the other hand, we note that their normal coordinates do not

even contain the order parameter, so the frequencies  $\omega_1$  and  $\omega_2$  do not vanish at the point of the second-order phase transition. The values of the frequency  $\omega_1$ , of the quasiferromagnetic AFMR mode (which had previously been assumed to be soft in the course of this transition), is given at the transition point by the expression<sup>7</sup>

$$\omega_{01} = \gamma_2 (\chi_{\parallel}/\chi_{\perp})^{\frac{1}{2}} H_f + (\gamma_1 - \gamma_2) (\chi_{\perp}/\chi_{\parallel})^{\frac{1}{2}} (H_f + H_D).$$
(9)

Consequently, a more thorough analysis of the symmetry of the problem in the thermodynamic potential and the equations of motion implies that the ordinary AFMR modes are not qualitatively different from each other: The normal oscillations are not the same as an oscillation of the order parameter in either case. It should be noted here that if we ignore the parallel susceptibility in the calculations and use the Landau-Lifshitz equations, we find that the frequency of the quasiferromagnetic AFMR mode must vanish at the point of the phase transition, in contradiction of existing experimental data.<sup>1-3</sup>

We believe that a final confirmation of the theory can be found if we are able to detect an energy gap in the  $\omega_1(H)$ spectrum after the completion of the spin flop induced by a magnetic field over a wide temperature range and if we can use expression (9) to reconstruct the function dependence  $\chi_{\parallel}(T)$ .

This calculation approach makes it possible in principle to explain the mechanism for the formation of any energy gap in a spin-wave spectrum due to a second-order phase transition and to specify which branch of the spectrum is soft. For this purpose, we should write the normal coordinates found by diagonalizing the matrix  $\hat{v}$  in the basis of eigenvectors of the stability matrix  $\hat{\alpha}$ . That mode whose normal coordinate is the order parameter (i.e., which is the same as the eigenvector of the matrix  $\hat{\alpha}$  which corresponds to the zero eigenvalue) is the soft mode. Here is is necessary to pay strict attention to ensure that the symmetry of the equations of motion corresponds to the symmetry of the problem. Incorporating a parallel susceptibility without a decay, for example, would lead to the result that the relaxation mode simply drops out of the calculations, and the soft mode disappears from the AFMR spectrum, despite the fact that the flop is a second-order phase transition. This is the nonphysical result which we mentioned above.

## **EXPERIMENTAL RESULTS AND DISCUSSION**

The experiments were carried out on a direct-amplification spectrometer over the frequency range 75–400 GHz ( $\lambda = 4$ –0.75 mm), over the magnetic-field range 3–130 kOe, at temperatures from 78 to 400 K. As samples we selected float-zoned single crystals of yttrium and dysprosium orthoferrites,<sup>12</sup> which have been studied quite thoroughly at room temperature.<sup>1–3</sup> Yttrium orthoferrite is a convenient model compound for studying orthoferrites since it contains the nonmagnetic ion Y<sup>3+</sup>, and all the magnetic properties are determined by the interaction of the iron sublattices.

As was mentioned in the preceding section, however, the energy gap in which we are interested here, which appears in the spin-wave spectrum at the point of a secondorder phase transition, results primarily from exchange, so it should be observed regardless of any additional interactions with other subsystems (elastic, rare-earth, etc.). It was for this reason that we decided on dysprosium orthoferrite,  $DyFeO_3$ , in which (in contrast with the situation in  $YFeO_3$ ) a decrease in the temperature is accompanied by significant structural changes in the AFMR spectra due to the onset of an interaction of the rare earth and iron subsystems.<sup>13</sup>

In the experiments we determined the positions of the lines in which the incident electromagnetic radiation is absorbed by the sample as a function of the applied magnetic field, in the orientation  $H \parallel a$ , at various temperatures. A regulation system kept the sample temperature constant to within  $\pm 0.5$  K throughout the experiment. The experimental procedure and the procedure for precisely aligning the sample in the magnetic field are described in detail in Ref. 7. Here we will simply mention that the symmetry of the experimental problem imposes some fairly severe requirements on the precision of the orientation of the sample in the magnetic field which can be achieved at the maximum separation of the AFMR absorption lines near the field  $H_f$  at  $H < H_f$  and  $H > H_f$  at the given frequency. With an essentially continuous frequency spectrum (the radiation sources were backward-wave tubes), this method yields some very good results. The best precision achieved in the alignment of the sample in the magnetic field was  $\pm$  3'. The actual orientation precision in the present experiments was  $\leq 15'$ .

Our measurements of the AFMR spectra showed that the positions of the experimental points on the  $\omega_1^2(H)$  curve for the quasiferromagnetic mode in fields 10% or more above  $H_f$  is essentially independent of the deviation of the **a** axis of the sample from the magnetic field direction within  $\pm 2^\circ$ . For this reason, a very slight degradation of the orientation (by a few arc minutes) does not lead to an error in the determination of the gap (expressed in kiloersteds)  $\Delta_{01} = (\omega_{01}/\gamma_0)^2$  since  $\Delta_{01}$  is found by extrapolating the Hdependence of  $(\omega_1/\gamma_0)^2$  from the region  $H > 1.1H_f$  to the field  $H_f$ . The transition field was determined to within 1° from the tails of the absorption lines (nonresonant absorption<sup>14</sup>).  $H_f$  was found to be independent of the disorientation of a sample within the same limits ( $\pm 2^\circ$ ). Figure 1



FIG. 1. Frequency of the quasiferromagnetic AFMR mode versus the magnetic field near the completion of the spin flop at various temperatures.  $a-DyFeO_3$ ;  $b-YFeO_3$ .

shows some illustrative results on  $\omega_1^2/\gamma_0^2$  as a function of Hnear the completion of the spin flop at various temperatures. We clearly see that a significant energy gap in the AFMR spectrum forms at  $H = H_f$  over the entire temperature range studied. At the temperature T = 78 K (at which  $\Delta_{01}$  is at a minimum), the value of  $\Delta_{01}$  in DyFeO<sub>3</sub> is nearly an order of magnitude greater than  $\Delta_{me}$ , due to the magnetoelastic interaction:  $\Delta_{me} \approx 40$  kOe<sup>2</sup>,  $\Delta_{01} \approx 300$  kOe<sup>2</sup>. In YFeO<sub>3</sub>, the ratio  $\Delta_{01}/\Delta_{me}$  reaches 10<sup>2</sup> (see Ref. 7 regarding the calculations of  $\Delta_{me}$ ). Clearly, the magnetostriction mechanism contributes only very slightly to the formation of the observed energy gap.

Figure 2 shows the temperature dependence of the energy gap for the compounds studied. It is an easy matter to explain why  $\Delta_{01}$  falls off more rapidly with the temperature for DyFeO<sub>3</sub> than for YFeO<sub>3</sub> on the basis of the theory proposed here. Specifically, it follows from the basic assumption that the value of  $\Delta_{01}$  is determined primarily by the product of  $\chi_{\parallel}$  and  $H_f^2$ , so the dependence  $\Delta_{01}(T)$  is steeper in DyFeO<sub>3</sub>, in which  $H_f$  falls off sharply with decreasing temperature (Fig. 1a), than it is in YFeO<sub>3</sub>, in which  $H_f$  remains essentially constant over the temperature range studied.

It was shown in Ref. 7 that in the yttrium orthoferrite the kinetic coefficients  $\gamma_1$  and  $\gamma_2$  differ from each other and from the gyromagnetic ratio  $\gamma_0 = ge/2mc$  by no more than 1–2% at room temperature. Using this result and relation (9), we find the following expression for  $\chi_{\parallel}/\chi_{\perp}$ , which holds to within  $(\Delta \gamma / \gamma_0)^2$ , where  $\Delta \gamma = \gamma_1 - \gamma_2$ :

$$\chi_{\parallel}/\chi_{\perp} = \Delta_{01}/H_f^2 - 2\Delta\gamma \left(H_f + H_D\right)/\gamma_0 H_f.$$
(10)

At room temperature, where  $\Delta_{01}^{1/2}$  is a few tens of kiloersteds, the corrections for the small difference in kinetic coefficients are only a few percent, and we can ignore the second term in (10). With decreasing temperature, however, the contribution of the first term in (10) falls off, and the second term becomes important. In DyFeO<sub>3</sub>, this effect should be seen more vividly because of the decrease in  $H_f$ and the simultaneous increase in  $H_d$  at low temperatures (more on this below).

A comment is in order here since the  $Dy^{3+}$  ion is magnetic, while expression (9) was derived without consideration of the R-Fe interaction. As a rule, temperatures above 50 K are regarded as quite high for a rare earth subsystem (the magnetic ordering temperature of the  $Dy^{3+}$  ions is  $T_{N2} \approx 5$  K). Nevertheless, it follows from our study of the



FIG. 2. The energy gap  $\Delta_{01} = (\omega_{01}/\gamma_0)^2$  in the spectrum of the quasiferromagnetic AFMR mode in the field at which the spin flop is completed, as function of the temperature. O—YFeO<sub>3</sub>, **●**—DyFeO<sub>3</sub>.



FIG. 3. Temperature dependence found for the ratio  $\chi_{\parallel}/\chi_{\perp}$  from an analysis of the experimental data with help of expression (10). O—YFeO<sub>3</sub>, • — DyFeO<sub>3</sub>. (See the text proper for an explanation.)

AFMR spectra in DyFeO<sub>3</sub> that the effect of the R-Fe interaction is significant up to temperatures above 100 K. By virtue of the exchange enhancement, a leading role is played here by the parameter  $\tau$ , which is a phenomenological coefficient in the thermodynamic potential in the  $H_x L_z$  term, which describes the anisotropic part of the R-Fe interaction.<sup>8-15</sup> This effect is manifested as an increase in the effective Dzyaloshinskiĭ field, which increases to a level at liquidnitrogen temperature which is three times its value at room temperature. It has also been observed that for  $T \ge 78$  K it is not legitimate to ignore the intensification factor  $\eta_x$ , which characterizes the effect of the exchange field of the rare earth ions on the weak ferromagnetic moment of the iron sublattice. An expression was accordingly derived for the energy gap  $\Delta_{01}$  in the R-Fe interaction case. The corrections for this interaction, which can be estimated by analyzing the AFMR spectra obtained at various temperatures, are found to be significantly smaller than the effect on the energy gap of the difference in the kinetic coefficients at low temperatures.

Taking all these comments into account, we derived the temperature dependence of the ratio of the parallel and perpendicular susceptibilities for the two compounds (Fig. 3). The behavior  $\chi_{\parallel}(T)/\chi_{\perp}(T)$  agrees well with the general physical picture of the behavior found for the parallel and perpendicular susceptibilities in molecular-field theory.<sup>16</sup> For example, an extrapolation of the dependence  $\chi_{\parallel}(T)/\chi_{\perp}(T)$  to the Neél temperature yields a value of approximately unity (the dashed line in Fig. 3), and at low temperature this ratio is small.

The comparatively large experimental error at low temperatures is a consequence of the corrections (which we have already mentioned) for the difference in the kinetic coefficients  $\gamma_i$  (the magnitude of this error corresponds to  $\Delta\gamma/\gamma_0 \approx 1\%$ ). Although our experiments do not furnish a direct method for determining  $\chi_{\parallel}$ , we nevertheless find a quantity which is directly proportional to it from the AFMR spectra, with a known proportionality factor (except at low temperatures, where for magnetic R ions it is necessary to make a correction, which can easily be estimated from the same spectra, and where  $\Delta\gamma$  becomes significant because of the decrease in the energy gap).

## CONCLUSION

Let us summarize the results of this paper.

1. An experimental study has been carried out on the AFMR spectra of yttrium and dysprosium orthoferrites near the completion of the spin flop induced by a magnetic field applied along the **a** axis of the orthorhombic crystal. Over the entire temperature range studied (78-400 K), the results reveal a significant energy gap in the spectrum of the quasiferromagnetic AFMR mode at the point of the second-order phase transition. This gap increases with increasing temperature.

2. Explicit expressions have been derived for the order parameter of this transition and also for the normal coordinates of the oscillations corresponding to the AFMR modes at the point of the transition. These calculations were based on the thermodynamic potential which incorporates the symmetry of the problem most fully and on the basis of some thermodynamic equations of motion free of the limitations imposed by the Landau-Lifshitz equation.

3. It has been established that in this transition the AFMR mode which was previously regarded as soft is not an oscillation of the order parameter. It has also been established that the order parameter itself for the phase transition studied here is a more complex entity, consisting of a combination of several components characterizing the magnetically ordered state of the system.

4. The present experiments completely confirm the fact that the energy gap observed in the spectrum of the quasiferromagnetic AFMR mode in the course of the second-order phase transition is due primarily to the parallel susceptibility and is therefore of exchange origin. The ratio  $\chi_{\parallel}/\chi_{\perp}$  has been found as a function of the temperature for an orthorhombic antiferromagnet with a weak ferromagnetic moment.

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- <sup>1</sup>A. M. Balbashov, A. G. Berezin, Yu. Gufan, *et al.*, Pis'ma Zh. Eksp. Teor. Fiz. **41**, 391 (1985) [JETP Lett. **41**, 479 (1985)].
- <sup>2</sup>A. M. Balbashov, A. G. Berezin, P. Yu. Marchukov, *et al.*, Kratkie Soobshcheniya po Fizike 7, 3 (1986).
- <sup>3</sup>A. M. Balbashov, A. G. Berezin, Yu. M. Gufan, *et al.*, Pis'ma Zh. Eksp. Teor. Fiz. **43**, 205 (1986) [JETP Lett. **43**, 259 (1986)].
- <sup>4</sup>A. S. Borovik-Romanov and E. G. Rudashevskiĭ, Zh. Eksp. Teor. Fiz. **47**, 2095 (1964) [Sov. Phys. JETP **20**, 1407 (1964)].
- <sup>5</sup>A. M. Kadomtseva, A. P. Agafonov, M. M. Lukina, et al., Zh. Eksp. Teor. Fiz. **81**, 700 (1981) [Sov. Phys. JETP **54**, 374 (1981)].
- <sup>6</sup>K. P. Belov, A. K. Zvezdin, A. M. Kadomtseva, and I. B. Krynetskiĭ, Zh. Eksp. Teor. Fiz. **67**, 1974 (1974) [Sov. Phys. JETP **40**, 980 (1974)].
   <sup>7</sup>A. M. Balbashov, A. G. Berezin, Yu. F. Gufan, *et al.*, Zh. Eksp. Teor. Fiz. **93**, 302 (1987) [Sov. Phys. JETP **66**, 174 (1987)].
- <sup>8</sup>K. P. Belov, A. K. Zvezdin, A. M. Kadomtseva, and R. Z. Levitin, Orientatsionnye perekhody v redkozemel'nykh magnetikakh (Orientational Transitions in Rare Earth Magnetic Materials), Nauka, Moscow, 1979, Ch. 3.
- <sup>9</sup>Yu. M. Gufan, Zh. Eksp. Teor. Fiz. **60**, 1537 (1971) [Sov. Phys. JETP **33**, 831 (1971)].
- <sup>10</sup>E. G. Rudashevskiĭ, in: Tez. dok. na 16-ĭ Vsesoyuzn. konf. po fizike magnitnykh yavleniĭ (Proceedings of the Sixteenth All-Union Conference on the Physics of Magnetic Phenomena), Tula, 1983, p. 150.
- <sup>11</sup>V. A. Il'in and E. G. Poznyak, *Lineĭnaya algebra (Linear Algebra)*, Nauka, Moscow, 1984, Ch. 7.
- <sup>12</sup>A. M. Balbashov, A. Ya. Chervonenkis, A. V. Antonov, and V. B. Bakhteuzov, Izv. Akad. Nauk SSSR. Ser. Fiz. 35, 1243 (1971).
- <sup>13</sup>A. M. Balbashov, A. A. Volkov, S. P. Lebedev, *et al.*, Zh. Eksp. Teor. Fiz. **88**, 974 (1985) [Sov. Phys. JETP **61**, 573 (1985)].
- <sup>14</sup>F. B. Hagedorn, E. M. Gyorgy, R. C. Le Craw, *et al.*, Phys. Rev. Lett. **21**, 364 (1968).
- <sup>15</sup>A. K. Zvezdin, V. M. Matveev, A. A. Mukhin, and A. I. Popov Redkozemel'nye iony v magnitouporyadochennykh kristallakh (Rare Earth Ions in Magnetically Ordered Crystals), Nauka, Moscow, 1985, Ch. 14.
- <sup>16</sup>J. S. Smart, Effektivnoe pole vteorii magnetizma (Effective Field Theories of Magnetism), Saunders, Philadelphia, 1966 (Russ. transl. Mir, Moscow, 1968).

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