Magnetic resonance in the orthorhombic antiferromagnet Fe_3BO_6

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Measurements of the temperature dependence of AFMR in Fe_3BO_6 have been carried out at submillimeter wavelengths. In addition to the relativistic branch of AFMR known previously, a new one has been detected and studied. The results obtained are analyzed in terms of the thermodynamic model of second-order phase transitions.

The compound Fe_3BO_6 is antiferromagnetic with weak ferromagnetism at temperatures $T < T_N = 508$ K (Ref. 1). Its crystallographic symmetry is described by the D_{2h}^{16} space group. According to the neutron diffraction data of Ref. 2, the Fe_3BO_6 unit cell contains twelve iron ions, occupying the nonequivalent positions 8*d* and 4*c*, whose magnetic moments form a four-sublattice magnetic structure which at temperatures below the spin-flip transition temperature $T_{sp} = 415$ K is characterized by an ordering of the type $\Gamma_1(F_x, L_{2z}, f_x, l_{1z})$, and for $T_{sp} < T < T_N$, $\Gamma_a(F_z, L_{2x}, f_z, l_{1x})$; in both configurations $\mathbf{F}\|\mathbf{f}$. It should also be noted that Fe_3BO_6 is the only orthorhombic crystal in which the spin flip transition takes place via a first-order phase transition.

The static magnetic properties of Fe_3BO_6 were studied in Refs. 1, 3, and 4. Also studied was the spectrum of a branch of AFMR in Fe_3BO_6 (Refs. 5 and 6) that softens completely as T_{sp} is approached. In addition, Ref. 6 reported the observation of an additional signal which apparently was not due to AFMR. However, it is well known that for noncollinear ferrimagnets[7] such as Fe_3BO_6, the existence of four branches of AFMR should be expected. Two of them are exchange branches, and the corresponding frequencies are usually located in the IR range, and the other two branches are due to relativistic effects and have lower frequencies. The relativistic branches of AFMR have substantially different excitation conditions: one branch, which is usually lower in frequency, is observed when $\mathbf{h}\perp\mathbf{m}$, where $\mathbf{h}$ is the vector of the magnetic component of the microwave field and $\mathbf{m}$ is the vector of the weakly ferromagnetic moment, and the other branch is observed when $\mathbf{h}\|\mathbf{m}$; this permits them to be identified.[8] Thus, certain questions related to the spin dynamics of Fe_3BO_6 remain unstudied.

This paper presents new results of a study of AFMR in Fe_3BO_6 and of its behavior near the spin-flip transition.

EXPERIMENTAL DATA

Fe_3BO_6 single-crystal pellets measuring $\approx 8\times8\times6$ mm^3 were synthesized from a high-temperature solution. The samples to be studied, in the form of plates of thickness ≈ 1 mm and transverse dimensions $\approx 5\times5$ mm^2, were cut out in the *ac* and *bc* planes, respectively, to an accuracy of 1–2°, the natural faceting of the initial single crystals being used.

The temperature measurements were carried out in the 100–510 K range in a thermostat maintaining the temperature uniform to within ~1 K in the sample. The quasi-optical method[9] was used to study AFMR in the 75–550 GHz range, and at frequencies below 75 GHz the studies were done in waveguides using commercial wide-angle standing-wave coefficient meters.

Since the AFMR frequencies of Fe_3BO_6 changed appreciably with temperature, perference was given to the method of temperature scanning at fixed frequencies.[10]

RESULTS AND DISCUSSION

The AFMR spectrum of Fe_3BO_6 is shown in Fig. 1. The AFMR branch located at lower frequency was excited with $\mathbf{h}\perp\mathbf{m}$, which made it possible to identify it as the low-frequency relativistic branch ν_1. The temperature dependence of the frequency for this branch was found to be in good agreement with the data of Ref. 6. The resonance absorption line width for this branch, recalculated by allowing for the temperature dependence of its frequency, was found to be unchanged in the Γ_1 phase and equal to ≈ 6 GHz.

At temperatures greater than T_{sp} and frequencies less than 75 GHz, we studied the AFMR branch that was excited only when $\mathbf{h}\perp\mathbf{m}$; this made it possible to identify it as the low-frequency ν_1 branch in the Γ_3 phase. The general character of the temperature dependence of the frequency of the ν_1 branch in the entire range in which magnetic order exists was identical to the temperature dependence of the threshold field H_c inducing phase transitions between the Γ_1 and Γ_3 phases.[3]

In addition to the low-frequency branch, a new branch of AFMR in Fe_3BO_6, ν_2, was detected and studied (see Fig. 1). It was identified as a second relativistic branch, since, in complete conformity with theoretical calculations, it was excited only when $\mathbf{h}\|\mathbf{m}$. This was manifested in the fact that for a sample of the *ac* cut with the magnetic component of the microwave field oriented strictly either along the *a* axis for

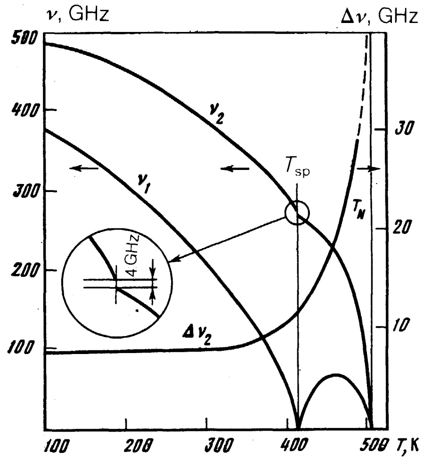

FIG. 1. Temperature dependence of the AFMR spectrum of Fe_3BO_6 and of the resonance absorption line width of the ν_2 branch.

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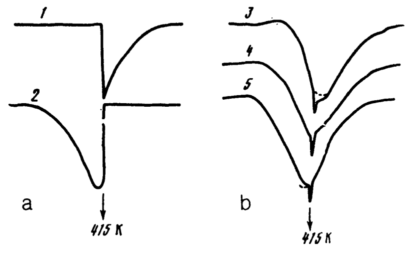

FIG. 2. Recordings of the temperature changes in line shape of the AFMR ν_2 branch near T_{sp}: (a) $\nu = 276$ GHz; 1—$\mathbf{h}\|c$; 2—$\mathbf{h}\|a$; (b) $\mathbf{h}$ at a 45° angle to the a and c axes; 3—$\nu = 271$ GHz, 4—$\nu = 273$ GHz, 5—$\nu = 275$ GHz.

the Γ_1 phase or along the c axis for the Γ_3 phase, a sudden disappearance of the resonance absorption line was observed when T_{sp} was reached, as shown in Fig. 2a. The observed character of the change in the resonance absorption line near T_{sp} resembles the behavior of the AFMR line in CoF_2 in the vicinity of the first-order phase transition induced by an external magnetic field.[11] The small difference between the temperatures of the extreme points of the steep slope of the resonance absorption line was due to the temperature gradient on the sample. Figure 2b shows the observed behavior of the resonance absorption line of the ν_2 branch near T_{sp} when the magnetic component of the microwave field is directed at a 45° angle to the a and c axes. The change in appearance of the resonance absorption line made is possible to detect a gap ≈ 4 GHz in the spectrum of the ν_2 branch at temperature T_{sp} (see the inset in Fig. 1).

The AFMR absorption line width $\Delta\nu_2$ of the ν_2 branch as a function of temperature is shown in Fig. 1. It is evident that, as in other antiferromagnets, the absorption line width of the ν_1 branch is appreciably smaller than that of the ν_2 branch, and as the temperature T_N is approached, a significant broadening of the ν_2 branch is observed.

In a theoretical description of the magnetic properties of Fe_3BO_6, use may be made of the well-known expansion of the thermodynamic potential for the D_{2h}^{16} group[12]:

$$\Phi = \frac{E}{2} m^2 - d_1 m_z l_x + d_2 m_x l_z + \frac{a_1}{2} l_x^2 + \frac{c_1}{2} l_z^2 + \frac{a_2}{4} l_x^4 + \frac{c_2}{4} l_z^4 + \frac{f}{2} l_x^2 l_z^2 - \mathbf{mH}, \quad (1)$$

where $\mathbf{m} \equiv (\mathbf{M}_1 + \mathbf{M}_2)/M_T$ and $\mathbf{l} \equiv (\mathbf{M}_1 - \mathbf{M}_2)/M_T$ are, respectively, the ferromagnetic and antiferromagnetic vectors, $\mathbf{M}_1$ and $\mathbf{M}_2$ are the magnetic moments of the sublattices, M_T is their magnitude at the given temperature, $E/2$ and d_1, d_2 are the effective fields of symmetric and antisymmetric exchange, a_1, c_1 are the effective fields of the bilinear anisotropy and a_2, c_2, f are those of the biquadratic anisotropy, and $\mathbf{H}$ is the external magnetic field. In contrast to orthoferrites, for which the condition $d_1 = d_2$ holds to a good approximation in Fe_3BO_6 an appreciable difference is observed between the components of the effective fields of antisymmetric exchange. Taking this fact into account, one can obtain an analytic form of the dependence of the resonance frequencies on the parameters of thermodynamic potential (1) by using the familiar equations of motion for vectors $\mathbf{m}$ and $\mathbf{l}$. In the notation of Ref. 12, these take the form

$$\nu_1^2 = \gamma^2[-A_a E + 2d_2(d_2 - d_1) + H(3d_2 - 2d_1) + H^2], \quad (2)$$

$$\nu_2^2 = \gamma^2[-A_z E + d_2(d_2 + H)] \quad (3)$$

in the Γ_1 phase and

$$\nu_1^2 = \gamma^2[A_0 E + 2d_1(d_1 - d_2) + H(3d_1 - 2d_2) + H^2], \quad (4)$$

$$\nu_2^2 = \gamma^2[-A_x E + d_1(d_1 + H)] \quad (5)$$

in the Γ_3 phase, where

$$A_0 \equiv c_1 + f - a_1 - a_2, \quad A_a \equiv c_1 + c_2 - a_1 - f,$$

$$A_x \equiv a_1 + a_2, \quad A_z \equiv c_1 + c_2.$$

We shall find the condition determining the stability boundary of the Γ_1 and Γ_3 phases. For this purpose, we shall use the approach developed in Ref. 13 for describing orientational phase transitions. The anisotropic part of the thermodynamic potential (1) can be transformed to

$$\Phi = K_1 \sin^2\varphi + K_2 \sin^4\varphi, \quad (6)$$

where φ is the angle between axis $\mathbf{a}$ and $\mathbf{l}$, and K_1 and K_2 are the first and second anisotropy constants. The difference between the magnitudes of the effective antisymmetric exchange fields d_1 and d_2 gives rise to the folowing additional terms in the anisotropy constants K_1 and K_2:

$$K_1 = d_1(d_1 - d_2)/E + A_0/2, \quad (7)$$

$$K_2 = (d_1 - d_2)^2/2E + (A_a + A_0)/4. \quad (8)$$

The realization of the transition from the Γ_1 to the Γ_3 phase via a first-order transition corresponds to the fulfillment of the conditions[13] $K_2 < 0$ and $K_1 + K_2 = 0$, or

$$d_1^2 - d_2^2 + \frac{E}{2}\left(\frac{A_a + A_0}{2}\right) = 0. \quad (9)$$

Comparing the condition for the first-order phase transition (9) with expressions (2) and (4) for the resonance frequencies of the ν_1 branch, we see that the latter does not undergo a jump at T_{sp}. In the same way, it can be shown that the ν_2 branch at T_{sp} undergoes a jump whose magnitude is given by

$$\nu_2^2(\Gamma_1) - \nu_2^2(\Gamma_3) = \gamma E(a_2 - c_2)/2, \quad (10)$$

i.e., is determined by the difference between small constants of biquadratic anisotropy. The expressions for the threshold fields in the Γ_1 and Γ_3 phases, with allowance for the anisotropy of the effective antisymmetric exchange fields are

$$H_c(\Gamma_1) = {}^1/_2\{-3d_1 + 2d_2 + [(d_1 - 2d_2)^2 - 4EA_0]^{1/2}\}, \quad (11)$$

$$H_c(\Gamma_3) = {}^1/_2\{-3d_2 + 2d_1 + [(d_2 - 2d_1)^2 + 4EA_a]^{1/2}\}. \quad (12)$$

Having made use of our measurements and the results

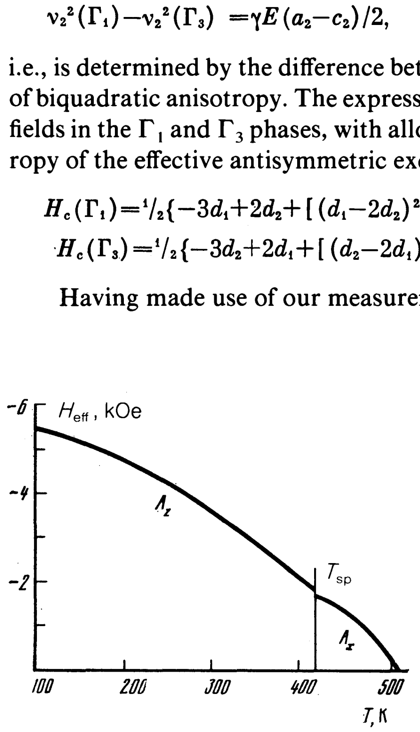

FIG. 3. Temperature dependence of the effective anisotropy fields of Fe_3BO_6 ($A_0 = 12.5$ Oe, $A_a = 19.0$ Oe).

of studies of magnetization, susceptibility, and threshold fields,[1,3,4] we can use Eqs. (2)–(5), (11), and (12) to calculate the temperature dependence of the effective anisotropy fields A_z and A_x throughout the region where Fe_3BO_6 exhibits magnetic order and the values of fields A_a and A_0 at temperature T_{sp} (Fig. 3). At the point T_{sp}, all four effective fields A_x, A_z, A_a, A_0 are in fact determined. Using the identity

$$A_0/2+A_a/2-A_z+A_x=(a_2-c_2)/2 \tag{13}$$

and comparing the values obtained with the magnitude of the jump (10) of the ν_2 branch at T_{sp}, we obtain satisfactory agreement.

Thus the static and dynamic properties of Fe_3BO_6 are adequately described in terms of a two-sublattice model of an antiferromagnet with weak ferromagnetism.

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