Investigation of antiferromagnetic resonance in Fe_3BO_6 in strong magnetic fields

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The magnetic field dependence of antiferromagnetic resonance in the orthorhombic ferromagnet Fe_3BO_6 has been measured in the submillimeter electromagnetic wavelength range. The magnetic field was directed along the principal crystallographic axes. The results have been analyzed on the basis of a thermodynamic theory which takes account of a possible variation in the absolute variation of the absolute value of the sublattice magnetic moments. The temperature dependence of the principal internal fields which determine the AFMR frequency of an orthorhombic antiferromagnet with weak ferromagnetism is obtained.

The compound Fe_3BO_6 is an antiferromagnet with weak ferromagnetism at temperatures $T < T_N = 508$ K (Ref. 1). Its crystallographic symmetry is described by the space group D_{2h}^{16}. The Fe^{3+} magnetic moments form a four-sublattice magnetic structure which at temperatures less than the spin–flop transition $T_{sp} = 415$ K is characterized, according to Mal'tsev *et al.*,[2] by ordering of the type $\Gamma_2(m_x, l_z, m'_x, l'_z)$, where **m**, **l** and **m**′, **l**′ are ferro- and antiferromagnetism vectors for the magnetic ions which are in the nonequivalent positions, respectively 8(d) and 4(c). For $T_{sp} < T < T_N$ a magnetic structure of the type Γ_3 (m_z, l_x, m'_z, l'_x) results and in both configurations we have $\mathbf{m} \| \mathbf{m}'$ (Ref. 2). It should be noted that Fe_3BO_6 is the only orthorhombic crystal in which a spin–flip transition is accomplished by a first order phase transition.

The antiferromagnetic resonance (AFMR) spectrum of Fe_3BO_6 was studied earlier in the temperature range 100–510 K in the absence of an external magnetic field.[3,4] It was shown that the dynamic properties of Fe_3BO_6 are described within the framework of the two-sublattice model of an antiferromagnet with weak ferromagnetism. However, these investigations revealed the temperature dependences only of part of the effective internal fields, which determine the AFMR frequencies in orthorhombic antiferromagnets. In order to obtain the values of the whole collection of internal fields it was advisable to study the AFMR behavior of Fe_3BO_6 in an external magnetic field directed along the principal crystallographic directions. In the present work results are given of an experimental investigation of the field dependence of the low-frequency AFMR mode in Fe_3BO_6 in the temperature range 220–380 K in which the Γ_2 magnetic structure is stable in the absence of a field.

For a systematic description of the field dependences of the AFMR frequencies for nonzero temperature, it is essential in the general case to consider changes not only of orientation, but also in the absolute magnitude of the magnetic moments of the sublattices. A thermodynamic theory which takes account of this possibility has been developed.[5-9] The AFMR frequencies obtained on such a model can differ from the frequencies obtained on a constant-moment model. In particular, for an orientational second order phase transition by an external field the frequency of the softening mode ν_1 does not become zero, which was found experimentally in the orthoferrites $YFeO_3$ and $DyFeO_3$ (Refs. 8,9). In Fe_3BO_6, with a structure isomorphic with the orthoferrites, we also found an appreciable energy gap at the analogous phase transition, which at room temperature amounted to 50% of the gap in the absence of a field.

EXPERIMENTAL

The measurements were carried out with the "Solenoid" installation of the high-magnetic field section of the Institute of General Physics of the USSR Academy of Sciences, giving steady magnetic fields up to 130 kOe (Ref. 10). Field calibration with an accuracy up to 1% was achieved for the position of the EPR line in $RbMnF_3$, positioned on the specimen. The AFMR measurement was carried out with the apparatus described by Rudashevsky *et al.*[11] in which backward-wave tubes were used as submillimeter band electromagnetic radiation generators, tunable in frequency (75–400 GHz). A tube previously calibrated with a Fabry–Perot interferometer[12] with reticular mirrors was used for controlling the frequency to an accuracy better than 0.5%.

The radiation was incident through a multimode copper waveguide on the sepcimen located in the center of the solenoid and was recorded by a n-InSb receiver cooled to 4.2 K. The thermal stabilization system achieved a constant specimen temperature to an accuracy of 0.5 K in the range 77–400 K. Polycrystalline Fe_3BO_6 specimens with dimen-

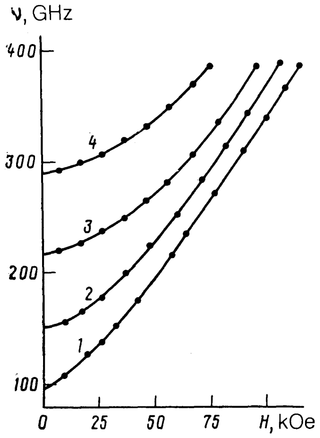

FIG. 1. Dependence of AFMR frequency in Fe_3BO_6 on external magnetic field for the collinear phase ($\mathbf{H} \| \mathbf{a}$) at different temperatures: 1) 380 K; 2) 345 K; 3) 290 K; 4) 220 K.

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TABLE I. Values of effective internal fields and the ratio $\chi_{\parallel}/\chi_{\perp}$ for Fe_3BO_6.

T, K	H_{eff}, kOe				$\chi_{\parallel}/\chi_{\perp}$
	d_2	d_1	A_a	H_c	
220	17.2	13.8	−1.96	–	–
290	16.1	12.3	−1.07	78.0	0.33
345	14.8	10.7	−0.51	55.0	0.37
380	13.8	9.8	−0.08	38.0	0.48

sions $6\times3\times1$ mm^3, oriented perpendicular to the **a** and **c** axes were placed in the center of the magnetic field and perpendicular to the cross section of the waveguide. Since a high accuracy in orienting the specimen relative to the external magnetic field was required in the experiment,[13,14] the apparatus was equipped with a positioning device with which precision orientation could be achieved directly in the magnetic field from the observed effect to an accuracy of 3′ (Ref. 8).

RESULTS AND DISCUSSION

We use the simplest two-sublattice approximation $\mathbf{m}=\mathbf{m}'$, $\mathbf{l}=\mathbf{l}'$ to interpret the experimental results, i.e., will not distinguish ions which are in nonequivalent sites.

The Collinear Phase, H||a

In an external field, applied along the **a** axis (in the direction of the weak ferromagnetic moment **m**), the orientations of the vector **m** and the antiferromagnetism vector **l** do not vary. In that case ($\tilde{\mathbf{h}}\perp\mathbf{m}$, where $\tilde{\mathbf{h}}$ is the magnetic component of the VHF field) only a low-frequency relativistic AFMR branch ν_1 can be excited in the solenoidal field.[15] The experimental magnetic field dependence of the AFMR frequency for this branch for four temperatures is shown in Fig. 1. The results of extrapolating the experimental curves to zero magnetic field agree well with the earlier temperature dependence of the AFMR gap for the low-frequency branch.[3,4]

In the given situation (the external field does not lead to reorientation of the **m** and **l** vectors and the magnitude of the field is much less than the spin–flop transition field), the possibility of a change in the absolute magnitudes of the magnetization of the sublattices can be neglected.[8] As a result of this we used the formula describing the experimental $\nu_1(H)$ dependences obtained by Ozhogin *et al.*,[13] taking account of the symmetrical and antisymmetrical parts of the Dzyaloshinskiĭ interaction:

$$\nu_1^2=(\gamma/2\pi)^2[A_aE+2d_2(d_2-d_1)+(3d_2-2d_1)H+H^2]. \quad (1)$$

Here E is the exchange field, A_a is the anisotropy field, d_1 and d_2 are the Dzyaloshinskiĭ interaction constants (expressed in kOe) written in the form $d_1m_zl_x-d_1m_xl_z$ and γ is the gyromagnetic ratio.

The experimental curves were approximated by the polynomial (1) using the least squares method. The accuracy in reproducing the experimental dependences in this way was better than 3%. This allowed the value of $3d_2-2d_1$ to be found for each of the fixed temperatures and the values of d_1 to be evaluated from the values of d_2 known from static measurements.[1,16] The values of the field d_1 in the Γ_2 phase obtained in this way join on well with the values of d_1 in the Γ_3 phase, which confirms the correctness of the approach taken as a basis.

The value of the anisotropy fields A_a can be determined from Eq. (1) using the value of d_1. Calculations by the method described above of the value of the effective fields d_1 and A_a for different temperatures are shown in Table I.

The Noncollinear Phase, H||c

In a field directed parallel to the **c** axis, reversal in the **ac** plane of the magnetic moments of the sublattices takes place, and for $H>H_c$ a collinear phase with $\mathbf{l}\perp\mathbf{H}$ again arises. The emergence of the collinear phase is a second-order phase transition. The experimental magnetic field dependences of the AFMR frequencies are shown in Fig. 2. It can be seen that in relatively weak fields the frequency first decreases as a function of the external magnetic field, then rises sharply. The minimum value of the AFMR frequency is reached for the limiting frequency for completing the spin–flip transition.

The reason for the gap in the spectrum of low-frequency modes arising at the second-order phase transition is discussed by Balbashov *et al.*[8,9] It lies in the fact that the normal coordinate for this mode does not coincide with the order parameter of the spin–flop transition for non-zero susceptibility in the direction of the antiferromagnetism vector. (The soft mode for which the order parameter and the normal coordinate coincide is then a relaxation mode). Balbashov *et al.*[8] obtained a formula for the value of the energy gap in the threshold field for a weak ferromagnet with antisymmetrical Dzyaloshinskiĭ interaction. We give, without derivation, the formula for the gap in the case of a weak ferromagnet with symmetrical and antisymmetrical parts of the Dzyaloshinskiĭ interaction, obtained in a similar way:

$$\nu_0=\frac{\gamma}{2\pi}(\chi_{\parallel}/\chi_{\perp})^{1/2}[H_c-(d_2-d_1)], \quad (2)$$

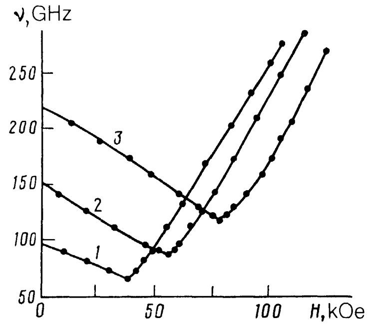

FIG. 2. Dependence of AFMR frequency in Fe_3BO_6 on external magnetic field for the non-collinear phase (H||c) at different temperatures: 1) 380 K; 2) 345 K; 3) 290 K.

where $\chi_{\parallel}$ is the susceptibility in the direction of the antiferromagnetic vector and $\chi_{\perp}$ is the susceptibility in the perpendicular direction.

We note that the value of the gap in Fe_3BO_6 falls with increasing temperature at the same time as the gap in $YFeO_3$ increases. This difference can be explained by noting that in $YFeO_3$ the temperature dependence of the gap is mainly determined by the $\chi_{\parallel}$ (T) relation ($\chi_{\perp}$ and H_c depend weakly on T in the range investigated), while in Fe_3BO_6 there is still a strong dependence of H_c on T.

By determining the values of the threshold field from the experiment, which agree well with the results of static measurement,[17] the value of the energy gap and also d_1 and d_2, the temperature dependence of $\chi_{\parallel}/\chi_{\perp}$ can be calculated (see Table I). The values obtained for this ratio agrees with the results of similar calculations given by Balbashov *et al.*[9] for the orthoferrites $YFeO_3$ and $DyFeO_3$.

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