Study of the Antiferromagnetic Resonance Spectrum in $BaMnF_4$

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The two branches of antiferromagnetic resonance are detected and studied at 4.2°K in an orthorhombic antiferromagnet, with the direction of the static magnetic field along the principal axes of the crystal. It is shown that both branches are well described by the formulas for the two-sublattice collinear model. The temperature dependence of the gap and of the linewidth is studied for the high-frequency branch.

HOLMES, Eibschütz, and Guggenheim[1] studied the temperature dependence of the magnetic susceptibility of a single crystal of $BaMnF_4$; they found that below $T \approx 25°K$ this material undergoes a transition to an antiferromagnetic state. The behavior of the susceptibility, however, was very unusual: with lowering of the temperature, the crystal remains magnetically isotropic down to 32°K, although the susceptibility goes through a diffuse maximum at a temperature $T_{max} \approx 50°K$. Below 32°K there occurs an anisotropy of the susceptibility: the susceptibility parallel to the b axis of the crystal approaches zero, while the susceptibility perpendicular to this axis goes through a minimum at $T \approx 25°K$ and becomes equal to $\chi_\perp = 3 \cdot 10^{-2}$ csgemu/mol. at $T \to 0$.

The susceptibility behavior described, as was shown theoretically by Lines[2] and as follows from experiments on $BaNiF_4$ and $BaFeF_4$[3,4], is characteristic of materials in which the exchange interaction occurs only between magnetic ions lying in a single layer. In the present research, crystals of $BaMnF_4$ were synthesized, and antiferromagnetic resonance (AFMR) was studied in them for the purpose of explaining the possible peculiarities in AFMR resulting from the presence of a layered structure.

As was shown by Keve, Abrahams, and Bernstein[5], $BaMnF_4$ is a rhombic crystal. Its crystal symmetry is described by the space group C_{2v}^{12}. The elementary cell contains four Mn^{++} ions. The lattice constants are: a = 5.9845 Å, b = 15.098 Å, c = 4.2216 Å.

Because the stratification of the crystal must basically affect the temperature dependence of the sublattice magnetizations, one might think that the dependence of the AMFR frequency on magnetic field at low temperatures would be governed by the same formulas as for a three-dimensional antiferromagnet with the same crystallographic symmetry. The theory of AFMR for a rhombic crystal was developed in papers of Keffer and Kittel[6], Yosida[7], Ubbink[8], and Gorter[9] (see also the review article[10]). In these papers it was shown that the energy of a rhombic antiferromagnet can be described in the form

$$E = {}^1/_2 B\mathbf{m}^2 + {}^1/_2 a_x l_x^2 + {}^1/_2 a_y l_y^2 - \mathbf{mH} \qquad (1)$$

(here $\mathbf{m} = \mathbf{M}_1 + \mathbf{M}_2$, $\mathbf{l} = \mathbf{M}_1 - \mathbf{M}_2$; $\mathbf{M}_i$ is the magnetization of a sublattice). If one introduces the effective anisotropy fields $H_{A_1} = {}^1/_2 a_x l$ and $H_{A_2} = {}^1/_2 a_y l$ and exchange field $H_E = {}^1/_2 Bl$, then one obtains the following formulas for the AFMR frequencies in the directions perpendicular to the axis of antiferromagnetism: for $\mathbf{H} \parallel x$,

$$(\nu_1/\gamma)^2 = 2H_{A_1}H_E + H^2, \qquad (2a)$$

$$(\nu_2/\gamma)^2 = 2H_{A_2}H_E; \qquad (2b)$$

and for $\mathbf{H} \parallel y$,

$$(\nu_1/\gamma)^2 = 2H_{A_1}H_E, \qquad (3a)$$

$$(\nu_2/\gamma)^2 = 2H_{A_2}H_E + H^2. \qquad (3b)$$

If the field is applied along the z axis, then at a field H_c^* there occurs a flopping of the sublattices. If $H_{A_1} < H_{A_2}$, then H_c^* is determined by the expression

$$H_c^* = \left[\frac{2H_{A_1}H_E}{1 - \chi_\parallel/\chi_\perp}\right]^{1/2}. \qquad (4)$$

For $\mathbf{H} \parallel z$ and $H > H_c^*$, the formulas for AFMR have the form

$$(\nu_1/\gamma)^2 = H^2 - 2H_{A_1}H_E, \qquad (5a)$$

$$(\nu_2/\gamma)^2 = 2(H_{A_2} - H_{A_1})H_E. \qquad (5b)$$

The formulas for the AFMR frequencies when $\mathbf{H} \parallel z$ and $|\mathbf{H}| < H_c^*$ are unwieldy and will not be quoted here.

METHOD AND SPECIMENS

AFMR in $BaMnF_4$ was studied in a magnetic spectrometer described in paper[11]. The resonance absorption line was recorded by means of an xy-recorder on the basis of the change of a microwave signal reflected from the crystal, as a function of the magnitude of the magnetic field. Measurements were made in the interval from 4.2 to 35°K, for which a vacuum cryostat was used[12]. The temperature was measured by means of a carbon thermometer, calibrated at the points 4.2, 27.25, and 77.5°K. The accuracy of the measurement and maintenance of the temperature was 0.5°K.

The $BaMnF_4$ crystals were grown by fusion of single-crystal MnF_2 and BaF_2 in an atmosphere of HF. The melting point of $BaMnF_4$ is $T_{mp} = 740 \pm 10°C$. The single crystals were grown in an atmosphere of He, since HF destroys this compound at temperatures below 400°C. The crystals have a rosy color; they are fragile and easily cleave along a (010) plane. The single crystals were oriented on x-ray apparatus, and then parallelepipeds were cut from them with edges ~1 mm, parallel to the principal axes of the crystal with accuracy 3°. The specimens were glued to the copper end-cap of a waveguide with BF-6 glue. The crystals with

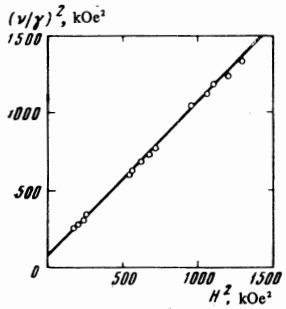

FIG. 1

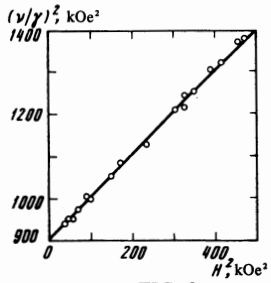

FIG. 2

FIG. 1. Dependence of the square of the resonance frequency of the low-frequency branch on the square of the applied magnetic field for **H** ∥ a.

FIG. 2. Dependence of the square of the resonance frequency of the high-frequency branch on the square of the applied magnetic field for **H** ∥ c.

the smallest number of defects were chosen on the basis of x-ray photographs and of the resonance spectra.

RESULTS AND DISCUSSION

1. Resonance Branches at T = 4.2°K

The AFMR spectrum was studied for directions of the static field along the principal axes of the crystal, at frequencies from 45 to 100 GHz, in magnetic fields up to 40 kOe. The experimental results are shown in Figs. 1–3.

As is seen from Figs. 1 and 2, within the limits of experimental error a linear relation between ν^2 and H^2 is fulfilled, in agreement with formulas (2a) and (3b). A smaller value of the gap is obtained for a field directed along the a axis of the crystal. Thus in the notation used above, the x and y axes correspond to the a and c axes of the crystal. By extrapolation of the straight lines in Figs. 1 and 2, we obtained the following values of the energy gaps in the AFMR spectrum:

$$H_{AE_1} = \sqrt{2H_{A_1}H_E} = 9.2 \pm 0.2\ \text{kOe}, H_{AE_2} = \sqrt{2H_{A_2}H_E} = 30.1 \pm 0.2\ \text{kOe}.$$

The value γ = 2.8 GHz/kOe obtained (g = 2.0) is usual for Mn^{++} ions.

Figure 3 shows the AFMR spectrum in the case in which the field is directed along the b axis. At $H = H_c^*$ there was observed the so-called "orientational resonance", connected with reorientation of 1 from the easy axis b to the middle axis a. To this resonance correspond the points on the vertical straight line at $H = H_c^*$. From Fig. 3 it is seen that $H_c^* = 9.8 \pm 0.2$ kOe; this, according to formula (4), is consistent with the value of the gap H_{AE_1} as well as with the value $H_c^* = 10$ kOe

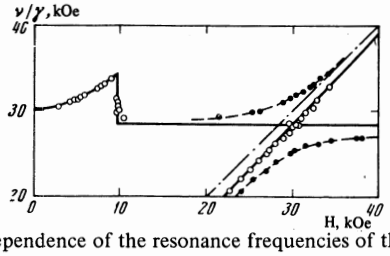

FIG. 3. Dependence of the resonance frequencies of the AFMR branches on applied magnetic field for **H** ∥ b. The dashed-dotted line shows the paramagnetic dependence $\nu = \gamma H$

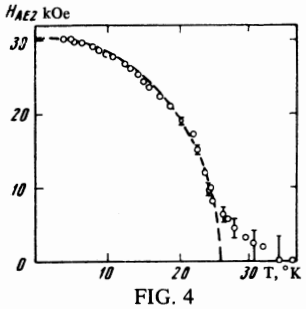

FIG. 4

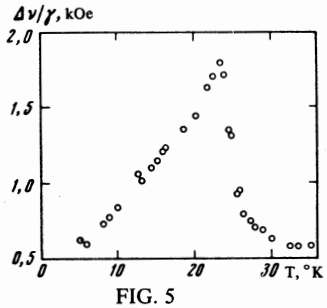

FIG. 5

FIG. 4. Temperature dependence of the gap for the high-frequency branch.

FIG. 5. Temperature dependence of the line-width for the high-frequency branch.

obtained in paper[1]. The line-width ΔH of the "orientational resonance" amounted to ~1 kOe. The continuous curves in Fig. 3 were calculated with formulas (5a) and (5b), with the values of the constants given above.

At small deviations of **H** from the easy axis, splitting of the AFMR branches was observed. The dark points in Fig. 3 show experimental data obtained with an inclination of **H** by an angle ~10° in the bc plane. They agree well with the dotted lines, which correspond to the theoretical formula[10]

$$\frac{H_x^2}{(\nu/\gamma)^2 + 2H_{A_1}H_E} + \frac{H_y^2}{(\nu/\gamma)^2 + 2(H_{A_1} - H_{A_2})H_E} = 1$$

for inclination of **H** by an angle of 12°. Such a phenomenon has been observed in antiferromagnets with anisotropy of the "easy plane" type[12,13].

2. Temperature Measurements

Investigation of the temperature dependence of the gap in the low-frequency branch (2a) was impeded by splitting of the AFMR branches, in consequence of inaccuracy in directing the static field along the principal axes of the crystal. Therefore we present here only the results of the temperature measurements of the gap for the high-frequency branch with **H** ∥ c, since in this case (see formula (3b)) the branches do not intersect.

The AFMR line was observed over the broad temperature interval from 4.2 to 35°K. For each temperature, the value of H_{AE_2} was calculated by formula (3a). The temperature dependence is shown in Fig. 4. The dotted curve in this figure is a normalized Brillouin function for S = 5/2, so chosen as to agree as well as possible with the experimental points in the range 4.2–24°K. It was found that for best fit it was necessary to choose the value of T_N in the Brillouin function as 25.6 ± 0.5°K.

Figure 5 shows the temperature dependence of the AFMR line-width. Because the AFMR frequency depends nonlinearly on the field, the experimental values of the line-width ΔH were converted to equivalent values of width in the frequency expression

$$\Delta\nu/\gamma = \gamma H \Delta H/\nu,$$

which are given in Fig. 5. It is seen that $\Delta\nu/\gamma$ increases with temperature; but that at a temperature about 2° below T_N, defined as was indicated above, it begins to fall abruptly. Noncoincidence of the position of the maximum of $\Delta\nu/\gamma$ with the temperature T_N has been

observed also in $CsMnF_3$[14], but no explanation of this has been found.

Notice the "tails" on the $H_{AE}(T)$ and $\Delta\nu/\gamma(T)$ curves in the range 25—33°K. It is possible that this is due to the fact, mentioned above, that $BaMnF_4$ is a two-dimensional antiferromagnet. In the three-dimensional antiferromagnets investigated up to now, H_{AE} always decreased abruptly on approach to T_N and vanished at $T = T_N$. The only exceptions were antiferromagnets with weak ferromagnetism, in which the induction of antiferromagnetism by a field at $T > T_N$ can manifest itself in the presence of H_{AE} above T_N (see, for example[15]); but in the case of $BaMnF_4$, both static data[1] and and our data on AFMR have proved the absence of weak ferromagnetism. Smearing out of the transition point because of imperfection of the specimens should be accompanied by strong broadening of the line above the transition point.

One may suppose that the observed "tails" are due to a characteristic of two-dimensional antiferromagnets and indicate that in such antiferromagnets, over a quite wide range of temperatures above the transition point, there exist long-lived regions of fluctuations of the short-range order. Such fluctuations may manifest themselves in the fact that the resonance frequency will differ from the paramagnetic above T_N. No theory of this phenomenon exists at present, but qualitative considerations regarding the possibility of such an effect have been presented in a number of papers (see, for example,[2,16]).

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