Magnetic resonance in the noncollinear antiferromagnet $Mn_3Al_2Ge_3O_{12}$

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Three antiferromagnetic-resonance branches have been observed in the noncollinear antiferromagnet $Mn_3Al_2Ge_3O_{12}$. Experimental and theoretical results are reported on the dependence of the resonance frequencies on the strength and direction of the magnetic field.

Low-frequency branches of spin excitations occur in magnetic materials in which relativistic effects are small in comparison with exchange effects. When relativistic effects are ignored, these branches are Goldstone modes associated with a spontaneous breaking of the symmetry of the exchange interactions. Relativistic effects give rise to finite gaps in the spectrum of these low-frequency oscillations. Since these oscillations experience in the homogeneous case a mechanical spin moment (and also a magnetic moment), as the quantity conjugate to the Goldstone degree of freedom—the rotation angle of the spin space—these gaps determine the magnetic-resonance spectrum of the material. A magnetic resonance has previously been observed only in the simplest collinear structures: in ferromagnets (and ferrimagnets), in which there is a single resonance, and in antiferromagnets, where there are two resonances. In noncollinear structures, in contrast, a magnetic resonance has been observed only in superfluid He^3-*B*, where, however, one spin-excitation branch remains gapless because of the particular degeneracy of the system.

In the present letter we report the observation and study of the behavior in external fields of three magnetic-resonance branches in the noncollinear antiferromagnet $Mn_3Al_2Ge_3O_{12}$.

A magnetic structure has been proposed for $Mn_3Al_2Ge_3O_{12}$ (Fig. 1) by Prandl[1] on the basis of neutron diffraction analysis. Studies of the magnetic properties and of the specific heat carried out by Valyanskaya *et al.*[2] have confirmed this model. The magnetic moments of the manganese lie in the (111) plane and are directed along or opposite [211], [121], and [112] axes; i.e., there is a noncollinear "triangular" 12-sublattice antiferromagnetic ordering. According to the exchange-symmetry theory,[3] a structure of this sort is described by two mutually perpendicular antiferromagnetic vectors. In our case the magnetic cell coincides with the crystal cell, so that these vectors are classified in terms of irreducible representations of the exchange crystal class. Since the magnetic symmetry of the structure is determined by the elements S_6R and σ_d, the exchange crystal symmetry class may be either D_{3d} or O_h (the symmetry group of the crystal in the paramagnetic phase is O_h^{10}). In each case, the antiferromagnetic vectors transform under two-dimensional representations of E_u. Since the structure does not change when the particles are interchanged in a rotation of C_2 around

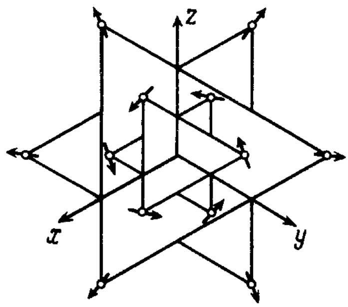

FIG. 1.

the C_4 axis (with the spins remaining in the same direction), the exchange crystal class that we are seeking is O_h.

This high symmetry of the magnetic structure substantially simplifies the problem of interpreting the experimental magnetic-resonance data, since there is only a single relativistic second-order invariant,

$$\lambda\left[l_{1x}l_{2x} - l_{1y}l_{2y} + \frac{\sqrt{3}}{2}\,(l_{1z}^2 - l_{2z}^2)\right], \tag{1}$$

which completely fixes the orientation of the spin system with respect to the crystallographic axes [see Eq. (35) in Ref. 3]. The antiferromagnetic vectors $\mathbf{l}_1$ and $\mathbf{l}_2$ ($\mathbf{l}_1 \perp \mathbf{l}_2$ and $\mathbf{l}_1^2 = \mathbf{l}_2^2 = 1$) are determined by the following transformations upon the interchanges of particles corresponding to rotations of C_4 and C_3: $C_4\mathbf{l}_1 = \mathbf{l}_1$, $C_4\mathbf{l}_2 = -\mathbf{l}_2$,

$$C_3\,\mathbf{l}_1 = -\frac{1}{2}\,\mathbf{l}_1 - \frac{\sqrt{3}}{2}\,\mathbf{l}_2, \qquad C_3 l_2 = \frac{\sqrt{3}}{2}\,\mathbf{l}_1 - \frac{1}{2}\,\mathbf{l}_2.$$

Introducing in the usual way Euler angles θ, φ, ψ, which determine the orientation of the triad of vectors $\mathbf{l}_1$, $\mathbf{l}_2$, and $[\mathbf{l}_1\mathbf{l}_2]$ with respect to the unit vectors x, y, z along the C_4 axes, we can replace (1) by

$$\lambda\left[\frac{1}{2}\sin 2\psi\cos 2\varphi\,(1+\cos^2\theta) + \sin 2\varphi\cos 2\psi\cos\theta + \frac{\sqrt{3}}{2}\sin^2\theta\cos 2\psi\right]. \tag{2}$$

For $\lambda > 0$, the minimum of the anisotropy energy in (2) corresponds to an eightfold-degenerate state (of the type $\cos\theta = 3^{-1/2}$, $\varphi = \pi/4$, $\psi = \pi/2$). In an external field, the spin plane rotates because of the anisotropy of the susceptibility. Since the behavior in an external field and the dynamics can be studied easily in accordance with the theory of Ref. 3, we will write some results here without derivation.

If the external field is directed along the C_3 axis, only two states with a spin plane perpendicular to the field will have a minimum energy (under the condition $\chi_\parallel > \chi_\perp$, where $\chi_\parallel$ is the susceptibility parallel to the direction of $[\mathbf{l}_1\mathbf{l}_2]$, and $\chi_\perp$ is the susceptibility perpendicular to $[\mathbf{l}_1\mathbf{l}_2]$. One of the frequencies corresponding to the oscillations of the spin plane around the field direction in these states is $\nu_1 = \gamma(2\lambda/\sqrt{3}\chi_\parallel)^{1/2}/\pi$; the

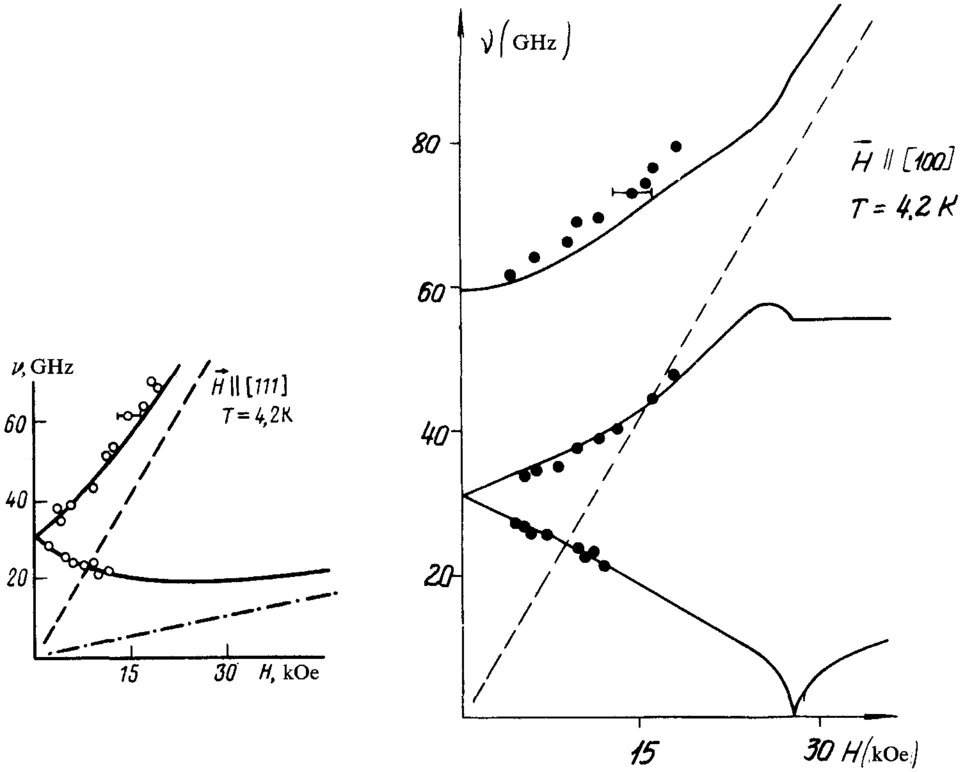

FIG. 2.

two others, corresponding to oscillations of the orientation of the spin plane, are

$$\nu_{2,3} = \sqrt{\nu_0^2 + \left(\frac{1+\eta}{4\pi}\gamma H\right)^2} \pm \frac{1-\eta}{4\pi}\gamma H, \quad \nu_0 = \gamma(2\lambda/\sqrt{3}\chi_\perp)/2\pi,$$

where $\eta = (\chi_\parallel - \chi_\perp)/\chi_\perp$. When a field is applied along the C_4 axis, all eight states are under identical conditions, and a rotation of the spin plane is described by the law $\cos\theta = \left[\sqrt{3} - (\chi_\parallel - \chi_\perp)\gamma^2 H^2/\lambda\right]^{-1}$ (the angles φ and ψ remain fixed). The frequencies are determined by a rather lengthy cubic equation, which we have solved numerically. The rotation of the spin plane terminates in a field $H_c = \left[(\sqrt{3}-1)\lambda/\eta\chi_\perp\right]^{1/2}/\gamma$. In strong fields we have the following results for the resonant frequencies: $\nu_2 = 2(\lambda/\chi_\perp)^{1/2}$

$$\nu_{1,3}^2 = \frac{\sqrt{3}}{2}\nu_0^2 + \frac{1+\eta^2}{2}\left(\frac{\gamma H}{2\pi}\right)^2 \pm \left\{\left[\frac{\sqrt{3}}{2}\nu_0^2 + \frac{1+\eta^2}{2}\left(\frac{\gamma H}{2\pi}\right)^2\right]^2 + \frac{3}{2}\nu_0^2 - \sqrt{3}\eta\left(\frac{\gamma H}{2\pi}\nu_0\right)^2 - \eta^2\left(\frac{\gamma H}{2\pi}\right)^4\right\}^{1/2}.$$

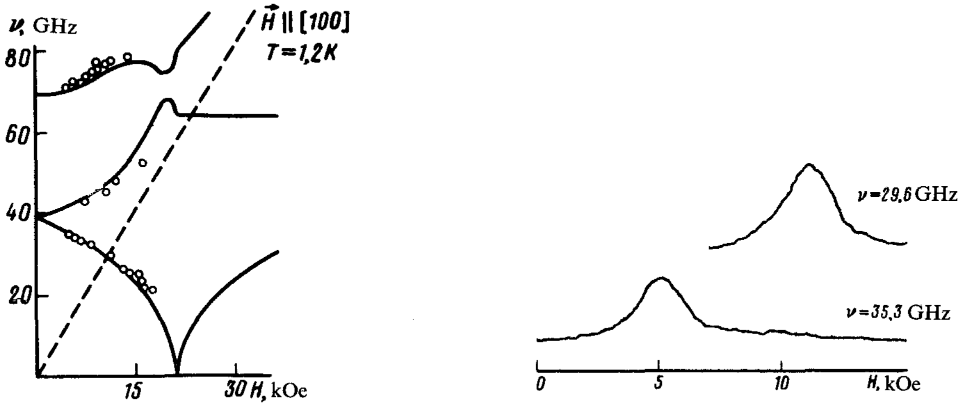

FIG. 3.

We carried out some measurements[1)] on a direct-amplification spectrometer over the frequency range from 20 to 78 GHz, in fields up to 20 kOe, at temperatures of 4.2 and 1.2 K (the Néel temperature is 6.67 K). Figure 2 shows the antiferromagnetic-resonance spectrum for the [111] and [100] directions (at 4.2 K), and Fig. 3 shows the spectrum for 1.2 K. The width of the antiferromagnetic-resonance lines is 5–7 kOe at 4.2 K and about 3 kOe at 1.2 K. We selected the following values for the adjustable parameters of the theoretical curves: $\nu_0 = 31$ GHz and $\eta = 0.126$ for 4.2 K and $\nu_0 = 39$ and $\eta = 0.28$ for 1.2 K.

Regardless of the field direction, we never observe more than three resonant lines. This result is evidence that the spin space easily undergoes a reorientation to a more favorable state. When the sample is rotated around the [110] axis at fixed values of the frequency and of the direction of the static field, we observe the angular dependence of the resonant field, which is characteristic of this situation.

The value of η at 4.2 K agrees with the result found by Sokolov and Kazeĭ, who directly measured both components of the susceptibility tensor in a single crystal.

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[1)]The $Mn_3Al_2Ge_3O_{12}$ single crystals were grown by B. V. Mil'. We thank him for furnishing the samples.

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