

Antiferromagnetic resonance in the garnet $\text{Ca}_3\text{Fe}_2\text{Ge}_3\text{O}_{12}$

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Antiferromagnetic resonance has been observed for the first time in a compound with garnet structure. Its angular and frequency dependences are investigated at $T = 4.2^\circ\text{K}$.

In the garnet $\text{Ca}_3\text{Fe}_2\text{Ge}_3\text{O}_{12}$, the Fe^{3+} ions occupy only octahedral sites and are isolated by at least two oxygen ions. Magnetic, ^[1,2] calorimetric, ^[3] and neutron-diffraction^[4,5] investigations have shown that this compound is antiferromagnetic at helium temperatures ($T_N = 1.25^\circ\text{K}$).

We have synthesized single crystals of $\text{Ca}_3\text{Fe}_2\text{Ge}_3\text{O}_{12}$ ($a_0 = 12.325 \text{ \AA}$ at room temperatures), in which we have observed antiferromagnetic resonance (AFMR) in the spin-flop state.

The AFMR was investigated in the range 26–46 GHz using a direct-amplification radiospectroscope with a pass-through resonator, the construction of which has made it possible to vary the frequency in the indicated interval. The microwave-power sources were standard klystron generators. The frequency-measurement error was 2×10^{-3} . The magnetic field was produced by a superconducting Helmholtz coil, rotation of which around the resonator made it possible to plot the angular dependences of the AFMR field. The magnetic-field marker was the EPR signal from the radical DPPH. The absorption signal was traced on an x - y plotter.

Small spheres were prepared from the grown single crystals, were polished, and were then annealed at $T = 1200^\circ\text{C}$ for 24 hours. The samples were oriented by x -ray diffraction with accuracy not worse than 0.5° .

For all the investigated crystals (12 samples), the

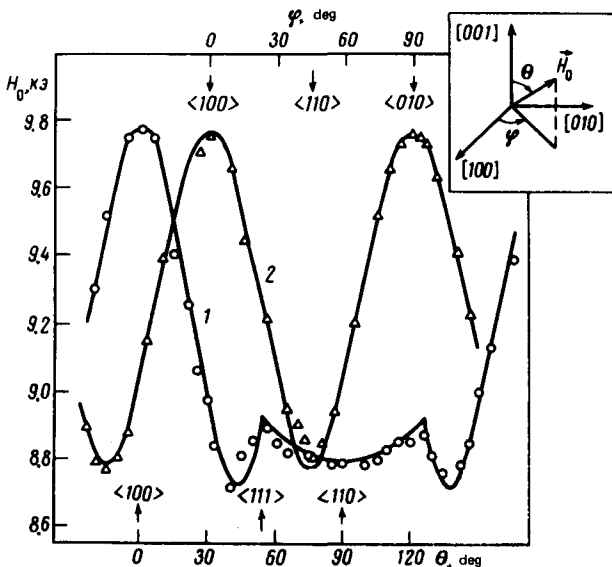


FIG. 1. Angular dependences of the resonance field of $\text{Ca}_3\text{Fe}_2\text{Ge}_3\text{O}_{12}$ at 4.2°K and at 26.73 GHz: 1—(110) plane, 2—(100) plane.

AFMR at 4.2°K was represented by a single symmetrical line, the width of which varied from sample to sample in the range 700–900 Oe. The linewidth increased somewhat when the temperature was lowered to 1.5°C .

Figure 1 shows the angular dependences of the resonance field (H_0) at 4.2°K and at 26.73 GHz in the planes (110) (curve 1) and (100) (curve 2) for a spherical sample of 1.2 mm diameter. The fourfold symmetry of H_0 in the (100) plane and the twofold symmetry in (110) indicates that $\text{Ca}_3\text{Fe}_2\text{Ge}_3\text{O}_{12}$ at 4.2°K is an undistorted cubic antiferromagnet with the easy magnetization directed along the $\langle 111 \rangle$ axes, i. e., the anisotropy constant is $K_1 < 0$.

The dependence of the AFMR frequency of a cubic crystal on the external magnetic field, for $H_0 > \sqrt{2}H_E H_A$ without allowance for the hyperfine interaction (the nuclear spins of both isotopes of iron are close to zero), is of the following form^[6]:

$$(\omega/\gamma)^2 = H_0^2 + 3B(\theta, \phi)H_E H_A, \quad (1)$$

where

$$B(\pi/2, \phi) = -\frac{4 \cos 4\phi}{7 + \cos 4\phi}$$

for the (100) plane and

$$B(\theta, \frac{\pi}{4}) = \begin{cases} -1 + \frac{13}{2} \cos^2 \theta - 6 \cos^4 \theta & \text{for } \theta \text{ between } [001] \text{ and } [111] \\ \frac{(2 - \sin^2 \theta)(3 \sin^2 \theta - 1)}{(2 + \sin^2 \theta)} & \text{for } \theta \text{ between } [111] \text{ and } [110] \end{cases}$$

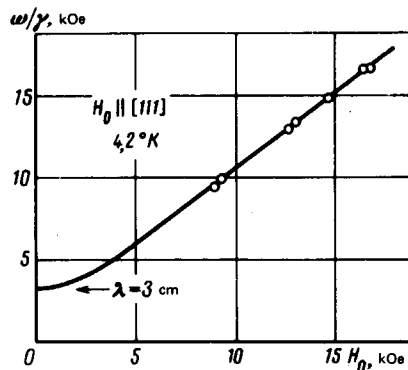


FIG. 2. Dependence of the AFMR frequency on the external field along the $\langle 111 \rangle$ axis at 4.2°K : \circ —experiment, curve—calculation in accordance with (2).

for resonance in the (110) plane. Here θ and ϕ are the polar and azimuthal angles between \mathbf{H}_0 and the corresponding crystallographic axes (Fig. 1), H_E is the exchange field, and H_A is the effective anisotropy field and is equal to $-4K_1/3M_0$ (M_0 is the sublattice magnetization in the ground state).

It is seen from Fig. 1 that the experimental results (points) are well described by a formula similar to (1) (solid lines) with an energy gap H_Δ^2 :

$$(\omega/\gamma)^2 = H_0^2 + 3B(\theta, \phi) H_E H_A + H_\Delta^2. \quad (2)$$

From the dependence of $[(\omega/\gamma)^2 - H_0^2]$ on B we found that the gap $H_\Delta^2 = (3.1 \pm 0.3)$ kOe² is isotropic and remains constant, within the accuracy limits, in the interval 4.2–1.6°K. This gives grounds for assuming that H_Δ^2 is of magnetostriction origin.

For the flopping field we obtained $\sqrt{2H_E H_A} = 3.2$ kOe. Using the experimental value $H_E = 404$ kOe,^[21] we obtained the anisotropy field $H_A = 12.8$ Oe.

The same value of H_E , H_A , and H_Δ^2 give the best agreement between the experimental results and the frequency dependence (2) of the AFMR along fixed crystallographic directions (Fig. 2).

The obtained value of H_A yields an anisotropy constant $K_1 = -22.3 \times 10^{-4}$ cm⁻¹ (per Fe³⁺ ion). This quantity can be compared with $K_1 = -10 \times 10^{-4}$ cm⁻¹ obtained in^[7] for the octahedral sublattice of Y₃Fe₅O₁₂ by the ferromagnetic-resonance method (at $T = 0^\circ\text{K}$). Obviously, this is one-ion anisotropy, the principal mechanism for which in the case of the Fe³⁺ in the garnet structure is connected, according to Geschwind,^[8] with the splitting

of the S state in the cubic and trigonal crystal fields. However, if we use the corresponding crystal-parameters determined from the EPR of the Fe³⁺ ions in diamagnetic gallates and aluminates,^[9] then we get for Ca₃Fe₂Ge₃O₁₂ the value $K_1 = -72 \times 10^{-4}$ cm⁻¹, which is more than three times larger than the value obtained from AFMR. The main cause of this fact is apparently the different degree of distortion of the octahedron surrounding the Fe³⁺. The garnet Ca₃Fe₂Ge₃O₁₂ has a rather symmetrical octahedron: the difference in the length of the edges does not exceed 0.1 Å,^[5] whereas, say, for Y₃Ge₅O₁₂ this difference amounts to 0.3 Å.^[10]

We note in conclusion that we have also observed antiferromagnetic resonance in the garnet Ca₃Cr₂Ge₃O₁₂, the octahedral sublattice of which consists of Cr³⁺ ions.

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