# Antiferromagnetic resonance in the cubic crystals FeGeG and CrGeG

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Antiferromagnetic resonance (AFMR) has been investigated in the range 1.7 to 15 K in the spin-flop state of monocrystals of garnets in which the octahedral sublattice is formed by $Fe^{3+}$ or $Cr^{3+}$ ions ($T_N \approx 13$ K). The experimental angular variations of the AFMR field are described by theoretical relations that take account of magnetoelastic interaction [A. G. Berezin and V. G. Shavrov, Zh. Eksp. Teor. Fiz. **72**, 2362 (1977) (this issue) [Sov. Phys. JETP **45**, 1242 (1977)]]. The best agreement of theory with experiment is obtained with the following values of the effective anisotropy and magnetostrictive fields (4.2 K): for CrGeG, $H_{A1} = 38.1 \pm 0.5$, $H_1 = 15 \pm 1.5$, $H_2 = 11.2 \pm 1.5$ Oe; for FeGeG, these values are, respectively, $15.3 \pm 0.3$, $3.6 \pm 0.7$, $1.3 \pm 0.7$ Oe. The observed temperature variations of the anisotropy and magnetoelastic-interaction constants give evidence in favor of a single-ion mechanism for these interactions in cubic crystals.

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In compounds with the garnet structure, in which the magnetic 3*d* ions occupy only octahedral interstices of the oxygen lattice, antiferromagnetic ordering is observed at sufficiently low temperatures. At present there are a considerable number of papers on neutron-diffraction investigation of the magnetic structure and on study of the magnetic susceptibility and of the specific heat of antiferromagnetic garnets (see, for example, the review by Belov and Sokolov[1]).

Study of antiferromagnetic garnets by the method of antiferromagnetic resonance (AFMR) is of interest from at least two points of view. First, this is apparently the simplest method of obtaining information about the anisotropy of these compounds, since it is impossible to determine the value of the sublattice-flip field of a cubic antiferromagnet by measurements of the magnetization. Second, theoretical calculations carried out by Berezin and Shavrov[2] indicate an influence of magnetoelastic interaction on the antiferromagnetic resonance spectrum in cubic crystals. They showed that the magnetoelastic gap is anisotropic, and that its angular dependence in general is different from the angular dependence of the gap caused by magnetocrystalline anisotropy.

In the present work, AFMR was investigated in the spin-flop state of garnet monocrystals in which the octahedral sublattice is formed by magnetic ions $Fe^{3+}$ or $Cr^{3+}$. Since in Ref. 3 a difference had been observed in the nature of the specific-heat anomaly near the Néel temperature ($T_N$) between polycrystalline garnets $Cd_3Cr_2Ge_3O_{12}$ ($T_N = 12.55$ K,[3] $a_0 = 12.205$ Å) and $Ca_3Cr_2Ge_3O_{12}$ ($T_N = 13.5$ K,[3] $a_0 = 12.263$ Å), we measured the AFMR parameters on crystals of both series and detected no qualitative difference. The results presented below pertain to the garnet $Cd_3Cr_2Ge_3O_{12}$ (CrGeG), which has a narrower resonance-absorption line (~ 500 Oe). Because there has already been a short communication[4] regarding the detection and study of AFMR in $Ca_3Fe_2Ge_3O_{12}$ (FeGeG) with $T_N = 12.55$ K,[5] for this specimen we present only the results of a comparison of the experimental angular variations of the resonance field $H_0$, as obtained by us earlier,[4] with the theoretical calculations.[2]

## EXPERIMENTAL METHOD

The measurements were made over the frequency range 23.5–50 GHz, on a direct-amplification radiospectrometer with a pass-through cylindrical resonator, whose construction permitted retuning within the indicated frequency range. Standard klystron generators were used as sources of microwave power. The uncertainty of the frequency measurement was $2 \cdot 10^{-3}$. In measurement of the frequency variation of the AFMR, the external magnetic field $H$ was produced by a superconducting solenoid at 50 kOe, into which tightly fitted the resonator with the crystal glued to its ground bottom. In order to obtain the angular dependence of the resonance field, the crystal, oriented in an appropriate manner, was placed at the center of the resonator by means of a special quartz mount, and a magnetic field was produced by a superconducting Helmholtz coil, which could be rotated about the resonator. Because the resonator was tuned to the $H_{011}$ mode, the condition $\mathbf{H} \perp \mathbf{h}_{microwave}$ was satisfied in both cases. Serving as a marker for the magnetic field was the narrow line of the EPR signal from the radical $\alpha$, $\alpha'$-diphenyl-$\beta$-picrylhydrazyl (DFPH), placed near the specimen under study. The AFMR and EPR signals were recorded on an *xy*-recorder.

The temperature interval 1.7–4.2 K was obtained, as usual, by pumping of liquid-helium vapors. Measurements in the range 4.2–20 K were made in a shorted waveguide section with a vacuum jacket. Here use was made of a spectrometer connected in a bridge circuit, and the temperature of the specimen was controlled by an Allen-Bradley carbon thermometer. The accuracy of the measurement and regulation of the temperature was 0.1 K.

The FeGeG and CrGeG monocrystals were grown in the magnetism problem laboratory of Moscow State Uni-

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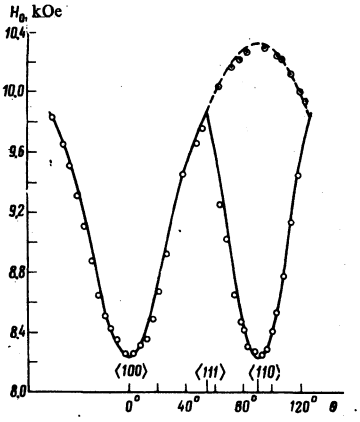


FIG. 1. Angular variation of the resonance field in the (110) plane for CrGeG. Points, experiment at frequency 26.35 GHz and $T = 4.2$ K. Solid line, calculation by formulas (6) and (8) in Ref. 2; dotted line, by formula (10).

versity, by B. V. Mill'. For obtaining AFMR, well faceted crystals of dimension 1 and 2 mm were selected, whose shape was nearly spherical. From several crystals, little spheres were made, which after polishing were annealed at 1200 °C for 24 hours. No important difference, however, was observed between the AFMR parameters of these specimens and those of the unworked specimens. The orientation of the crystals was done by an x-ray method with an accuracy no worse than 0.5°.

## RESULTS AND DISCUSSION OF THEM

As has been shown[2], with $\mathbf{h}_{\text{microwave}} \perp \mathbf{H}$ there should be excited, in the AFMR spectrum of a cubic antiferromagnet, the high-frequency branch of the oscillations

$$(\omega/\gamma)^2 = H_0^2 + H_E H_{A1} f_1(x) + H_E \tilde{H}_1 + H_E \tilde{H}_2 f_2(x). \quad (1)$$

Here $H_E$ is the exchange field, $H_{A1}$ is the anisotropy field (corresponding to the first anisotropy constant $K_1$), and $\tilde{\mathbf{H}}_1 = \frac{1}{2}(H_1 + H_2)$ and $\tilde{\mathbf{H}}_2 = \frac{1}{2}(H_1 - H_2)$ are the effective magnetostrictive fields with $H_1 = 9\lambda_{100}^2(C_{11} - C_{12})/M_0$ and $H_2 = 18\lambda_{111}^2 C_{44}/M_0$, where $C$ and $\lambda$ are the elastic constants of the crystal and the magnetostriction constants; $M_0$ is the sublattice magnetization. The functions $f_1(x)$ and $f_2(x)$ describe the angular variation of, respectively, $K_1$ and the anisotropic part of the magnetoelastic gap: for the (001) plane, $x = \cos 4\varphi$; for the (110) plane, $x = \sin^2\theta$. Because the accuracy of our experiment did not permit determination of more than three parameters from the angular variations of $H_0$, we disregarded in (1) the contribution of the Dzyaloshinskii interaction and of the hyperfine field (an isotropic term $H_E H_N$), and also the second anisotropy constant, which gives a contribution of the form $H_E H_{A2} f(x)$.

1. *Antiferromagnetic resonance in* CrGeG. Figures 1 and 2 show the angular variation of the resonance field in CrGeG at 4.2 K, at frequency 26.35 GHz, in the planes (110) and (100). The experimental values of $H_0$, shown on the graph by circles, indicate that in CrGeG the ⟨100⟩ axis is an axis of easy magnetization; that is, $K_1 > 0$. Furthermore, in the angle interval 60–120° in the (110) plane, and for all directions in the (100), two maxima of the resonance absorption are observed; in the (100) plane, the lower-field resonance is independent of the direction of the external magnetic field. The high high-field resonance is less intense, with a broader absorption line, and is practically unobservable near the directions of degeneracy: ⟨111⟩ for the (110) plane and ⟨100⟩ for the (100) plane.

Double-field resonance similar to that described above was observed by Eastman *et al.*[6] in a low-anisotropy cubic antiferromagnet, $TlMnF_3$ doped with cobalt. According to their measurements,[6] these two resonances correspond to two nonequivalent equilibrium positions of the sublattice magnetization in the spin-flop plane. In fact, analysis of the equilibrium spin configurations in CrGeG shows that when $H$ lies in the (100) plane, the lower-field resonance is due to a sublattice magnetization directed along the ⟨100⟩ axis. This axis corresponds to the "absolute" minimum of the anisotropy energy (a stable state) and to the angular function $f(x) = 1$ in (1) (see formula (2) in Ref. 2). The higher resonance corresponds to a sublattice magnetization perpendicular to the ⟨100⟩ axis and is characterized by a metastable state, with a relative minimum of the anisotropy energy and with the angular function $f(x) = \cos 4\varphi$ (formula (4) in Ref. 2).

When the field lies in the (110) plane, in the angle interval $0 \leq \theta \leq 54.7°$ a single resonance should be observed, corresponding to a unique minimum of the anisotropy energy, with the angular functions given in formula (6) in Ref. 2; but for $54.7° \leq \theta \leq 90°$, a stable and metastable state again occur, (8) and (10) in Ref. 2.

In Figs. 1 and 2, the solid and dotted lines correspond to the values of the resonance field for the stable and metastable resonances according to formulas (2), (4), (6), (8), and (10) in Ref. 2. We obtained the best agreement with experiment with the following values of the effective fields in (1):

$$H_E H_{A1} = 9.53; \quad H_E H_1 = 3.8; \quad H_E H_2 = 2.8 \text{ kOe}^2.$$

By using the experimental value of the exchange field $H_E = 250$ kOe,[3] we find

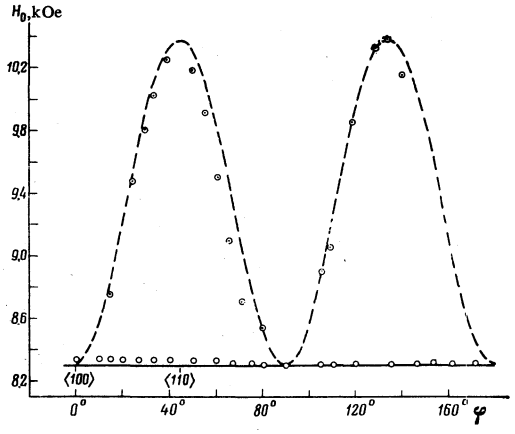


FIG. 2. Same as Fig. 1, but for the (100) plane. Solid and dotted lines, calculation by formulas (2) and (4) in Ref. 2.

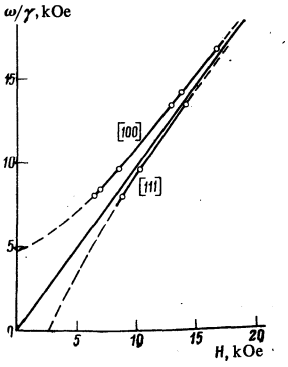


FIG. 3. Frequency-field relations for AFMR in CrGeG at 4.2 K, along principal axes of the crystal. Points, experiment. Dotted curves, calculation by formula (1). Solid line, EPR for $g = 1.980$.

$H_{A_1}=38.1\pm0.5$; $H_1=15\pm1.5$; $H_2=11.2\pm1.5$ Oe.

Since for a cubic crystal with $K_1 > 0$ the field $H_{A1} = 2K_1/M_0$, and $M_0 = g\mu_B S$, the value found for $H_{A1}$ gives for the anisotropy constant (per $Cr^{3+}$ ion): $K_1 = 27 \cdot 10^{-1}$ $cm^{-1}$. We remark that the sign of $K_1$ in CrGeG agrees with the results of investigation of the EPR of octahedral $Cr^{3+}$ in a matrix of diamagnetic garnets, which give for the parameter of the axial crystalline field in the spin Hamiltonian the values $D = 0.35$ $cm^{-1}$ (YGaG[7]) and $D = 0.209$ $cm^{-1}$ (YAlG[8]).

Figure 3 gives the frequency-field relation for AFMR in CrGeG at 4.2 K along the principal axes of the crystal, [100] and [111]. It is evident that the experimental data (points) agree well with the calculated, which were obtained by use of the values found for $H_{A1}$, $H_1$, and $H_2$.

It was established earlier[9] that in the AFMR spectrum of cubic $RbMnF_3$ there is observed an isotropic gap $H_N H_E$, caused by the hyperfine anisotropy field $H_N$, which leads to a strong temperature dependence of the resonance field ($H_N \propto 10/T$). Our measurements of the AFMR field in CrGeG in the interval 1.7–12 K indicate a very weak dependence of $H_0$ on temperature, in any event for 1.7-4.2 K. This is obviously explained by the small value of the hyperfine-interaction constant, which cannot be determined from the data on EPR of $Cr^{3+}$ in garnet[7,8], and also by the insignificant content of the isotope $Cr^{53}$ in natural chromium (< 10%).

Supposing that the change of $H_E(T)$ corresponds to a Brillouin function for $S = 3/2$, we calculated the temperature dependence of the magnetoelastic-interaction constants $B_1(T)$ and $B_2(T)$ and of the anisotropy constant $K_1(T)$ in CrGeG. Figure 4 shows the experimental values of $B_1$, $B_2$, and $K_1$ (divided by the values of 0 K) as functions of the relative magnetization $m(T) = M(T)/M(0)$.

According to the single-ion theory, in cubic crystals at low temperatures $K_1$ varies with temperature as $m^{10}$.[10] This relation is represented by curve 1 in Fig. 4. It is seen that the experimental values for $K_1$ in CrGeG agree well with the theoretical relation.

The temperature dependence of the magnetoelastic-interaction constants, within the framework of the same model, has the form[11]

$$\frac{B(T)}{B(0)} = \frac{I_{l+1}\{L^{-1}[m(T)]\}}{I_{l}\{L^{-1}[m(T)]\}}, \qquad (2)$$

where $I_{l+1/2}$ is the modified Bessel function, and where $L^{-1}$ is the inverse Langevin function of the relative magnetization (Curve 2 in Fig. 4). The experimental values of $B_1$ and $B_2$ (circles in Fig. 4) do not agree with the theoretical relation (2), in any event in the low-temperature range ($T < 8$ K). But it may be remarked that our determinations of $B_1$ and $B_2$ give evidence in favor of the single-ion mechanism of magnetoelastic interaction in CrGeG rather than of the two-ion, which is described by an $m^2(T)$ dependence (the dotted line in Fig. 4). As regards the temperature range near $T_N$, where the anisotropy is small, the error in the determination of $H_1$ and $H_2$ increases to such an extent that a calculation of $B_1$ and $B_2$ has no meaning.

We remark that a resonance-absorption line in CrGeG is observed also at $T > T_N$. From the paramagnetic-resonance field in CrGeG, we found the value $g = 1.980 \pm 0.005$, which agrees well with the results of EPR impurity $Cr^{3+}$ ions in diamagnetic garnets.[7,8]

2. *Antiferromagnetic resonance in* FeGeG. In accordance with our preliminary data on measurement of AFMR in a garnet with $Fe^{3+}$ in octahedra,[4] FeGeG is an undistorted cubic antiferromagnet with $K_1 < 0$. Processing of the experimental angular dependencies at 4.2 K (see Fig. 1 in Ref. 4) according to formula (1) gives the following values of the effective fields:

$H_E H_{A_1}=5.95$; $H_E H_1=1.45$; $H_E H_2=0.53$ $kOe^2$.

According to the measurements of Allain and Lecomte,[12] the sublattice-flip field in FeGeG is $H_E = 404$ kOe. With allowance for the Brillouin dependence $H_E(T)$, we get for 4.2 K.

$H_{A_1}=15.3\pm0.3$; $H_1=3.6\pm0.7$; $H_2=1.3\pm0.7$ Oe.

Since for $K_1 < 0$ the field $H_{A1} = -4K_1/3M_0$, the value found for $H_{A1}$ gives for the anisotropy constant (per $Fe^{3+}$ ion) $K_1 = -26.4 \cdot 10^{-4}$ $cm^{-1}$.

The temperature variation of $K_1$ for FeGeG, shown in

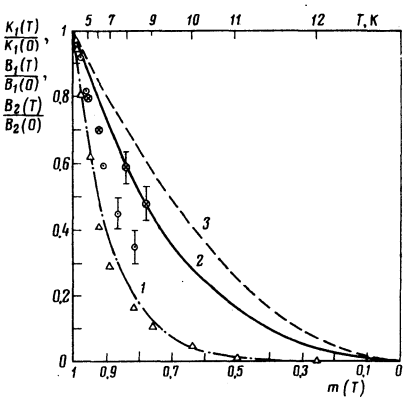


FIG. 4. Anisotropy constants $K_1$ and magnetoelastic-interaction constants $B_1$ and $B_2$, normalized with respect to $T = 0$ K, as functions of the relative magnetization $m(T)$ for CrGeG. Points, experiment: △, $K_1$; ○, $B_1$; ⊗, $B_2$. Curves, theory: 1, $m^{10}$; 2, formula (2); 3, $m^2$.

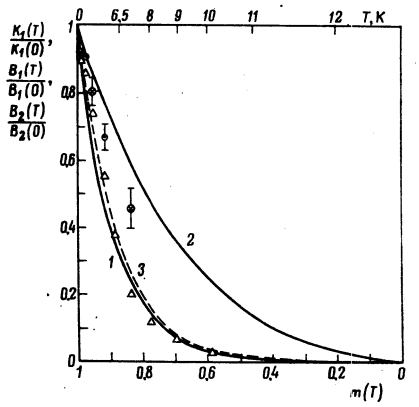


FIG. 5. Same as Fig. 4, but for FeGeG. Points, experiment: Δ, $K_1$; ○, $B_1$; ⊗, $B_2$. Curves: 1, single-ion theory; 2, formula (2); 3, EPR data.[13]

Fig. 5 (Δ), is well described by the single-ion theory (Curve 1). It should be noted that our results agree with the $K_1(T)$ variation obtained earlier[13] from EPR data on octahedral $Fe^{3+}$ ions in garnet (the dotted line in Fig. 5). But the absolute values of $K_1$ differ significantly; at 4.2 K, EPR gives $K_1 = -64 \cdot 10^{-4}$ cm$^{-1}$. For quantitative agreement it is apparently necessary, in the processing of the angular dependences of the AFMR spectrum, to take $K_2$ into account. (We note that in the garnet structure, according to Ref. 14, $K_2/K_1 \approx 3$ to 4 for octahedral $Fe^{2+}$, $Co^{2+}$, and $Ni^{2+}$ ions.)

In Fig. 5 the circles represent the magnetoelastic-interaction constants $B_1$ and $B_2$ in FeGeG. As in the case of CrGeG, so also in FeGeG the constants $B_1$ and $B_2$ are apparently of single-ion nature. From resonance measurements on FeGeG at $T > T_n$ we obtained $g = 2.003 \pm 0.001$, which agrees within the limits of error with EPR data on impurity $Fe^{3+}$ ions in octahedral sites of a garnet.[13]

In contrast to the chromium garnet, in FeGeG we observed no resonance connected with metastable states of the magnetization vector, although according to Ref. 2 it should occur when the external field lies in the (110) plane. A possible reason for this is the low intensity of the high-field resonance. Thus investigation of AFMR in garnets makes it possible to obtain information about the equilibrium spin configurations, the anisotropy, and the magnetoelastic interactions in antiferromagnetic garnets. In this respect, static magnetic methods, for crystals with high symmetry, are apparently much less informative.

Our measurements on two antiferromagnetic garnets with $Fe^{3+}$ and $Cr^{3+}$ ions in octahedra show that in these low-anisotropy compounds, an important role is played by magnetoelastic interactions (the magnetostriction constants of the specimens investigated are of the same order as in ferrimagnetic YIG). In contrast to uniaxial antiferromagnets, the effective anisotropy and magnetostrictive fields differ insignificantly.

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[1]K. P. Belov and V. I. Sokolov, Usp. Fiz. Nauk 121, 285 (1977) [Sov. Phys. Usp. 20, (1977)].
[2]A. G. Berezin and V. G. Shavrov, Zh. Eksp. Teor. Fiz. 72, 2362 (1977) (this issue) [Sov. Phys. JETP 45, 1242 (1977)].
[3]T. V. Valyanskaya, B. V. Mill', and V. I. Sokolov, Fiz. Tverd. Tela (Leningrad) 18, 1212 (1976) [Sov. Phys. Solid State 18, 696 (1976)].
[4]K. P. Belov, B. V. Mill', V. I. Sokolov, and O. I. Shevaleevskiĭ, Pis'ma Zh. Eksp. Teor. Fiz. 20, 98 (1974) [JETP Lett. 20, 42 (1974)].
[5]K. P. Belov, G. Ronninger, V. I. Sokolov, T. D. Hien, and B. V. Mill', Fiz. Tverd. Tela (Leningrad) 12, 1761 (1970) [Sov. Phys. Solid State 12, 1393 (1970)].
[6]D. E. Eastman, M. W. Shafer, and R. A. Figat, J. Appl. Phys. 38, 5209 (1967).
[7]J. W. Carson and R. L. White, J. Appl. Phys. 32, 1787 (1961).
[8]L. J. Schwee and J. R. Cunningham, J. Appl. Phys. 37, 449 (1966).
[9]D. T. Teaney, M. J. Freiser, and R. W. H. Stevenson, Phys. Rev. Lett. 9, 212 (1962).
[10]H. B. Callen and E. R. Callen, J. Phys. Chem. Solids 27, 1274 (1966).
[11]E. R. Callen and H. B. Callen, Phys. Rev. 129, 578 (1963).
[12]Y. Allain and M. Lecomte, Solid State Commun. 14, 119 (1974).
[13]P. Novák, V. Havlíček, B. V. Mill', V. I. Sokolov, and O. I. Shevaleevskiĭ, Solid State Commun. 19, 631 (1976).
[14]M. D. Sturge, F. R. Merritt, J. C. Hensel, and J. P. Remeika, Phys. Rev. 180, 402 (1969); M. D. Sturge, E. M. Gyorgy, R. C. LeCraw, and J. P. Remeika, Phys. Rev. 180, 413 (1969).

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