Peculiarities of the metamagnetic transition of a garnet single crystal $Ca_3Mn_2Ge_3O_{12}$

Z. A. Kazeĭ, B. V. Mill, and V. I. Sokolov
M. V. Lomonosov Moscow State University

(Submitted 1 February 1980)
Pis'ma Zh. Eksp. Teor. Fiz. **31**, No. 6, 338–342 (20 March 1980)

The existence of a magnetic phase transition, which does not correspond to the conventional model of a metamagnet with a high anisotropy, was determined from measurements of the magnetization of a $Ca_3Mn_2Ge_3O_{12}$ single crystal below $T_N = 13.85$ K.

PACS numbers: 75.30.Kg, 75.30.Gw, 75.50.Ee

Among the antiferromagnetic garnets[1] $Ca_3Mn_2Ge_3O_{12}$ (MnGeG) has a special place. In this compound the Mn^{3+} ions in the octahedral coordination have a doubly degenerate orbital ground state and hence are Jahn-Teller ions. According to the x-ray diffraction data,[2] at the temperature $T_{JT} \approx 400$ K a tetragonal distortion of the crystal lattice occurs in MnGeG ($c/a_{max} \approx 1.003$), which is attributed to the cooperative Jahn-Teller effect. This in turn accounts for the peculiar antiferromagnetic structure of MnGeG below T_c, which is determined from neutron diffraction,[3,4] for the nonlinear behavior of magnetization,[5] and for the anomaly of the specific heat[6] in strong magnetic fields. According to Plumier *et al.*,[7] the spin configurations of MnGeG in strong magnetic fields are associated with the high, local anisotropy of Mn^{3+} ions.

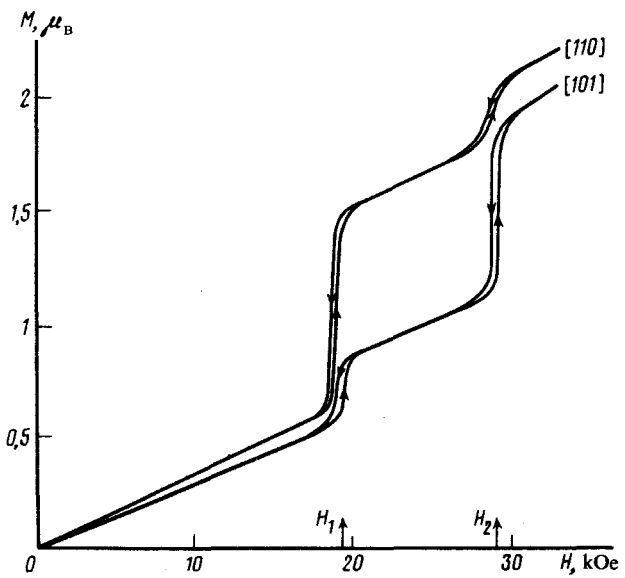

FIG. 1. Magnetization isotherms at 4.2 K of a MnGeG crystal for the direction of the magnetic field along the [110] and [101] axes.

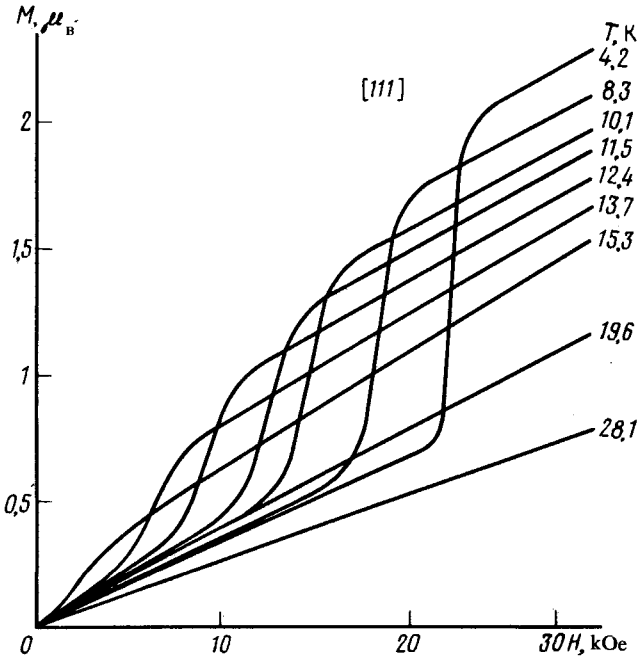

FIG. 2. Magnetization isotherms of MnGeG for the direction of the external magnetic field along the [111] axis.

Until now, the properties of MnGeG have been investigated by using polycrystalline samples. In this paper we report the results of measuring the magnetization of a MnGeG single crystal, which indicate that the metamagnetic transition in this garnet has many peculiarities below T_N.

The $Ca_3Mn_2Ge_3O_{12}$ crystals were grown from a solution in a Pb-GeO_2 melt according to a method proposed by Desvignes and Le Gall.[8] After cooling from 1250 to 1000 °C at the rate of 1.7 deg/h, each platinum crucible containing 30–38 g of the melt (42 mole % PbO, GeO_2, 18% CaO, and 6% Mn_2O_4) produced one garnet crystal 6 to 7.5 g in mass. The measurements were performed in the temperature range 4.2–70 K by using a vibration magnetometer with a superconducting solenoid. The investigated samples, which had the shape similar to a $2.5\times2.5\times2.5$-mm^3 cube, were oriented with an accuracy of $\pm 0.5°$ by using an x-ray method.

Figures 1 and 2 show the magnetization isotherms of the MnGeG crystal, which were plotted by using a X-Y potentiometer, for orienation of the external magnetic field along the ⟨111⟩ and ⟨110⟩ axes. It can be seen that the $M(H)$ dependences have jumps whose number depends on the orientation of H relative to the crystallographic axes. This is attributable to a splitting of the MnGeG crystal below T_{JT} into crystallographic "domains" whose tetragonal distortion axes have different directions, i.e., there are three types of "domains" corresponding to three fourfold axes of the cubic crystal. At 300 K the crystallographic "domains" can be determined from the shape of the diffraction line by means of x-ray diffraction.

It should be noted that the indicated "domain structure" in MnGeG is rather "hard": cooling of the sample below T_{JT} as a result of the action of the external compressive force of 10^7 dyn/cm^2 does not produce any noticeable variations of the $M(H)$ dependences below T_N or of the diffraction pattern at 300 K.

When the direction of H does not coincide with the symmetry axes of the crystal, the $M(H)$ isotherms at 4.2 K display three jumps corresponding to three types of "domains" with different orientations of the c axis relative to the external field. The number of jumps decreases with respect to symmetrical directions. Two jumps are observed for ⟨110⟩ (Fig. 1), since the orientation of the two types of "domains" relative to H is the same. At $H \| \langle 111 \rangle$ one jump remains (Fig. 2), since the c axis in all three types of "domains" form identical angles with H. As seen in Fig. 2, the magnitudes of the jumps and of the corresponding H_c decrease with increasing temperature. At $T > T_N$ the $M(H)$ dependences have the usual paramagnetic nature. The jumps M at 4.2 K are accompanied by a hysteresis for all orientations of the magnetic field (Fig. 2 shows only one change in direction of H), which indicates that there are first-order phase transitions.

Using M_{110} and M_{101}, which were measured along the two ⟨110⟩ axes of the "polydomain" crystal, we calculated the magnetizations of the "single-domain" sample m_{110} and m_{101}, which are nonequivalent in the tetragonally distorted lattice, with

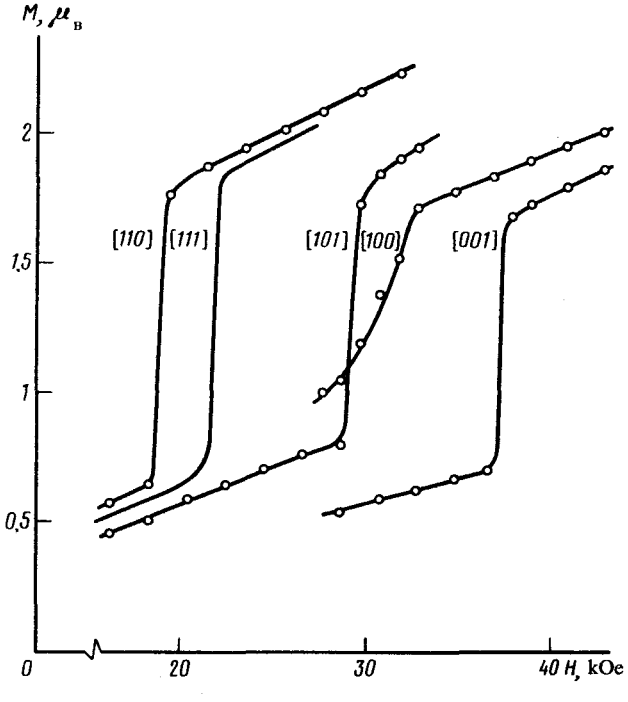

FIG. 3. Dependences of the magnetization on the field at 4.2 K of a "single-domain" MnGeG crystal for different crystallographic directions.

the help of the simple relations:

$$M_{110} = m_{110}\, x_3 + m_{101}\,(x_1 + x_2)\ ,$$

$$M_{101} = m_{110}\, x_2 + m_{101}\,(x_1 + x_3)\ .$$

Here x_1, x_2, and x_3 are parts of the "domains" in which the c axes are oriented, respectively, in the [100], [010], and [001] directions ($x_1 + x_2 + x_3 = 1$). They are determined by the jumps on the magnetization curves ΔM_{110} and ΔM_{101} in the critical fields H_1 and H_2 (see Fig. 1):

$$\frac{x_3}{x_2} = \frac{\Delta M_{110}(H_1)}{\Delta M_{101}(H_1)}\ ;\quad \frac{x_1 + x_2}{x_1 + x_3} = \frac{\Delta M_{110}(H_2)}{\Delta M_{101}(H_2)}\ .$$

The magnetization along the [001] and [100] axes were calculated analogously from the magnetizations measured along two directions such as $\langle 100\rangle$. At $H \| \langle 111\rangle$ the experiment gives the magnetization for the "single-domain" crystal. The results of calculation of the magnetization of MnGeG for different crystallographic directions at 4.2 K are shown in Fig. 3. It can be seen that the critical fields depend greatly on the direction of H in the crystal, whereas the jumps remain virtually constant. It is important that for all the measured directions of H the magnetization after the jump is $\sim 25\%$ of the saturation magnetization $M(0) = 2gs\,\mu_B$ (per MnGeG molecule), i.e., the sublattices do not close completely in MnGeG, as in the ordinary metamagnet with a large (of the order of the exchange energy) anisotropy. It should also be noted that extrapolation of the $M(H)$ dependences for $H > H_c$ to the value $H = 0$ gives $M \neq 0$.

We can assume from the indicated peculiarities of the magnetic behavior of the MnGeG crystal that the magnetism of superexchange orbital ordering, which was examined for the systems with Jahn-Teller ions by Kugel' and Khomskiĭ,[9] is realized in this garnet. At $H > H_c$, however, the experimental $M(H)$ dependences obtained by us differ qualitatively from the theoretical dependences obtained by Kugel' and Khomskiĭ[9] for a completely isotropic spin system. It is likely that the agreement between the theory and experiment can be improved by taking into account the anisotropy of the Mn^{3+} ions, which presumably plays a key role in the variation of the magnetic structure of MnGeG in strong magnetic fields.

In conclusion, we thank K. P. Belov for his attention to this work and D. I. Khomskiĭ, K. I. Kugel', and R. Z. Levitin for useful discussions.

[1]K. P. Belov and V. I. Sokolov, Usp. Fiz. Nauk **121**, 285 (1977) [Sov. Phys. Usp. **20**, 149 (1977)].
[2]Z. A. Zazeĭ, B. V. Mill', and V. I. Sokolov, Pis'ma Zh. Eksp. Teor. Fiz. **24**, 229 (1976) [JETP Lett. **24**, 203 (1976)].
[3]R. Plumier, Sol. St. Comm. **9**, 1723 (1971).
[4]R. Plumier and M. Sougi, J. Phys. **40**, 213 (1979).
[5]K. P. Belov, B. V. Mill', G. Ronniger, V. I. Sokolov, and T. D. Khien, Fiz. Tverd. Tela **12**, 1761 (1970) [Sov. Phys. Solid State **12**, (1970)].
[6]K. P. Belov, T. V. Valyanskaya, L. G. Mamsurova, and V. I. Sokolov, Zh. Eksp. Teor. Fiz. **65**, 1133 (1973) [Sov. Phys. JETP **38**, 561 (1973)].
[7]R. Plumier, D. Esteve, M. Le Comte, and M. Sougi, J. Appl. Phys. **49**, 1525 (1978).
[8]J. M. Desvignes and H. Le Gall, Mater. Res. Bull. **13**, 141 (1978).
[9]K. I. Kugel' and D. I. Khomskiĭ, Pis'ma Zh. Eksp. Teor. Fiz. **23**, 264 (1976) [JETP Lett. **23**, 237 (1976).