Magnetoanisotropic properties of the exchange-noncollinear antiferromagnet $Mn_3Al_3Ge_3O_{12}$

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The magnetic and magnetostriction properties of an antiferromagnet with a triangular magnetic structure—the single-crystal garnet MnAlG ($Mn_3Al_2Ge_3O_{12}$)—are investigated. For $T < T_N$ an anisotropy is observed which causes flipping of the "spin triangle" in an external magnetic field. At $T = 1.8$ K the spin-flop field for $\mathbf{H} \parallel [001]$ is 22 kOe. The effect of realignment of the domain structure on the angular and field dependences of the magnetostriction of the MnAlG crystal is studied. The Andreev and Marchenko exchange symmetry theory is used to discuss the experimental results.

Among the cubic antiferromagnets (AF) with the garnet structure MnAlG, in which Mn^{2+} ions occupy dodecahedral positions, is of special interest. According to neutron-diffraction data,[1,2] in this garnet, below $T_N = 6.65$ K,[3] the spins are oriented along the ⟨211⟩ axis and form a triangular configuration in the (111) plane. Analysis of the exchange interactions in MnAlG by the molecular-field method shows[3] that the noncollinear AF structure of this garnet is caused by the distinctive topology of exchange coupling of the weakly-anisotropic dodecahedral Mn^{2+} ions (nearest neighbors form a triangle) and the exchange interactions, comparable in magnitude, of the magnetic ions in the first, second and third coordination spheres. Thus, MnAlG is a noncollinear AF in the exchange approximation and due to its small magnetocrystalline anisotropy is a convenient system for experimental study.

In this work we investigate the magnetic and magnetocrystalline properties of single-crystal MnAlG with the goal of explaining the behavior of an exchange-noncollinear AF in an external magnetic field. Our view is that the available experimental data can be interpreted according to the exchange symmetry theory of Andreev and Marchenko.[4] As a sensitive and informative probe of the response of the spin subsystem of the crystal to an external magnetic field we use the degree of magnetostriction.

The magnetic susceptibility χ and the magnetostriction $u \equiv \Delta l / l$ are measured on single-crystal MnAlG samples of dimension $1.5 \times 1.5 \times 1.5$ mm^3, grown by the Czochralski method in air in a platinum crucible. According to the DTA-TG data, MnAlG melts congruently at 1370 °C, and the melt consumes oxygen intensively, yielding it back on crystallization of the garnet. Crystal growth is accompanied by a pronounced vaporization of GeO_2, but the liberation of O_2 on the crystallization surface makes it unstable.

Magnetic susceptibility was measured in the temperature interval 1.8–70 K with a vibromagnetometer. A capacitive transducer in a high-stability cryogenic oscillator, operating at a 1.5 MHz frequency, was used to measure the magnetostriction. The construction of the apparatus allowed measurement of the longitudinal magnetostriction $u_\parallel$ in a superconducting solenoid to 60 kOe and the transverse $u_\perp$ in a superconducting magnet, performed in a Helmholtz coil geometry (fields up to 40 k Oe), as well as the thermal expansion. Crystals were oriented by the x-ray method to 1° accuracy.

The χ-behavior of a crystal of MnAlG below T_N can be described by two independent components $\chi_\parallel$ and $\chi_\perp$ which correspond to the susceptibility parallel and perpendicular to the vector normal $\mathbf{n}$ to the "triangular spins" plane (Figure 1). On the basis of this we can assume that the χ-anisotropy in the plane of the triangle is small and can be neglected. Then for arbitrary orientation of the external magnetic field $\mathbf{H}$ the susceptibility of a single-domain crystal along the $\mathbf{H}$-direction has the form

$$\chi_H = \chi_\perp + (\chi_\parallel - \chi_\perp)\cos^2\alpha, \qquad (1)$$

where α is the angle between $\mathbf{n}$ and $\mathbf{H}$.

However, for $H = 0$ the crystal appears to have multiple domains; four types of AF domains exist, in which the normal $\mathbf{n}$ is directed along the 4-fold axes of a third-order BCC lattice. In addition, for each orientation of $\mathbf{n}$ there are two AF domains, connected with time-inversion operations. When a field is added there remain only those domains in which the direction of maximum χ makes the smallest angle with $\mathbf{H}$. Our investigation for $T < T_N$ shows that the MnAlG

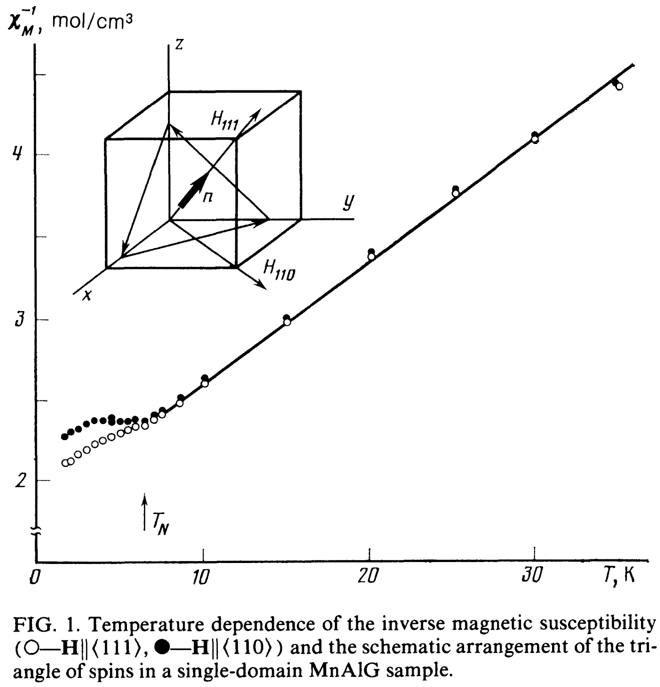

FIG. 1. Temperature dependence of the inverse magnetic susceptibility (O—$\mathbf{H}\parallel\langle 111\rangle$, ●—$\mathbf{H}\parallel\langle 110\rangle$) and the schematic arrangement of the triangle of spins in a single-domain MnAlG sample.

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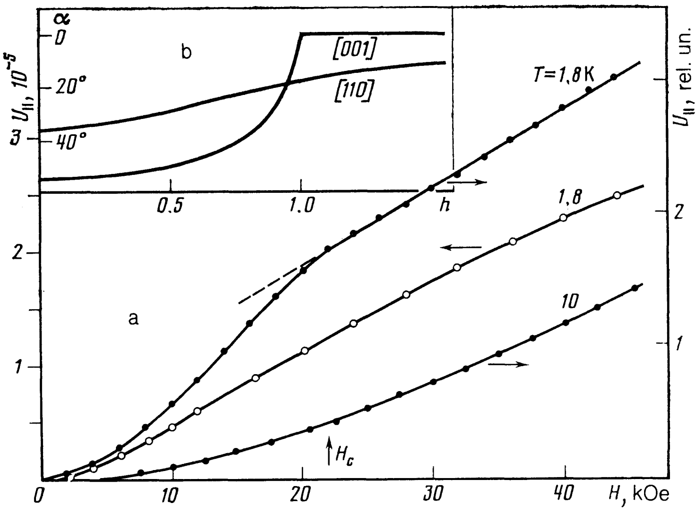

FIG. 2. Experimental isotherms of the longitudinal magnetostriction (a) and theoretical dependence of the angle between **n** and **H** on $h = H/H_c$ (b) for a field orientation **H**$\|\langle 001\rangle$ (●) and **H**$\|\langle 110\rangle$(○).

crystal stays single-domained in a field of $H \approx 1$ kOe.

In Figure 1 the temperature dependence of the inverse molar susceptibility along the $\langle 111\rangle$ and $\langle 110\rangle$ crystal directions is shown. It is evident that the χ-anisotropy arises below $T = 6.3$ K, corresponding, obviously, to T_N. This is confirmed by our measurements of thermal expansion of a MnAlG crystal, which show a sharp anomaly in Δl for $T = 6.3 \pm 0.2$ K. At temperatures $T > T_N$ the dependence of χ^{-1} on T follows the Curie-Weiss law with a Curie temperature $\Theta p = -(25 \pm 2)$ K, which within the error limits agrees with data for a polycrystalline MnAlG sample.[3] At the same time, below T_N for $H \gtrsim 1$ kOe the measured sample appears to be single-domained; $\chi_{111} = \chi_{\|}$ and $\chi_{110} = \chi_{\perp} + 2/3\,(\chi_{\|} - \chi_{\perp})$, that is, the greatest susceptibility is associated with **n**$\|$**H**. The observed χ-anisotropy of MnAlG is characterized by the following values for the parameter $\eta = (\chi_{\|} - \chi_{\perp}) \times \chi_{\perp}^{-1}$: at 4.2 K, $\eta = 0.15$ and grows to $\eta = 0.26$ at $T = 1.8$ K. We note that with the values cited η gives an adequate theoretical description of the experimental antiferromagnetic-resonance (AFMR) spectra in a MnAlG crystal.[5]

Due to χ-anisotropy, an external magnetic field brings about a reorientation of the Mn^{2+} magnetic moments relative to the crystal axes to a state with **n**$\|$**H**. To first order we can assume that in fields significantly less than the effective MnAlG exchange field $2H_E = 210$ kOe,[3] the reorientation of the spin triangle takes place without change in the relative orientation of the magnetic moments in the triangle. To experimentally observe the effects of the reorientation we carried out magnetostriction measurements on a crystal of MnAlG.

In Figure 2a isotherms of the longitudinal magnetostricton $u_{\|}$ are shown for field orientations **H**$\|\langle 001\rangle$ and **H**$\|\langle 110\rangle$. For **H**$\|\langle 110\rangle$ below T_N a critical field H_c is observed on the isotherms, at which the nonlinear dependence of $u_{\|}$ on H in a weak field changes to a linear one. We believe that this is connected with a change in the contributions to the measured magnetostriction. For $H < H_c$ the magnetostriction is anisotropic and is due to a fundamental reorientation of the triangle of spins. For $H > H_c$ the exchange striction, connected with a change in the relative orientation of the magnetic moments in the triangle, begins to play a decisive role.

Therefore, H_c corresponds to the spin-flop field, that is, the field at which the process of reorientation of the magnetic structure to the state of maximum χ along the **H**-direction is completed. The value $H_c = 22$kOe ($T = 1.8$K) that we have determined for **H**$\|\langle 001\rangle$ agrees with the size of the spin-flop field obtained by using the experimental AFMR data for MnAlG of Prozorova *et al.*[5]

For **H**$\|\langle 110\rangle$ there is no linear region on the $u\|(H)$ isotherm, which demonstrates the "drag" of the spin-flop process up to fields $H > 50$ kOe. The differences in magnetic behavior of a MnAlG crystal along the $\langle 001\rangle$ and $\langle 110\rangle$ directions are easily understood by looking at the theoretical dependence of α on h, where $h = H/H_c$, for two directions of **H**.

From an analysis of the Lagrangian for MnAlG written according to the exchange symmetry theory,[4] it follows that for **H**$\|\langle 001\rangle$ the spin-flop is completed in a field

$$H_c = [(\sqrt{3} - 1)\lambda(\chi_{\|} - \chi_{\perp})^{-1}]^{1/2}, \qquad (2)$$

where λ is the anisotropy parameter. For **H**$\|\langle 110\rangle$ the rotation to the state with $\alpha = 0$ is determined by the relationship

$$\sin\alpha = \lambda[\sqrt{3}\lambda + 2(\chi_{\|} - \chi_{\perp})H^2]^{-1}, \qquad (3)$$

that is, the spin-flop process is complete in an infinite field. Using experimental values of H_c and $\chi_{\|} - \chi_{\perp}$ for 1.8 K, we estimate according to Eq. (2) a value of $\lambda = 0.18\ \mathrm{cm}^{-1}$/ion for the anisotropy parameter.

To study the domain processes and the anisotropy of the magnetostriction in the AF investigated we measured the transverse magnetostriction $u_{\perp}$ of the MnAlG crystal for **H** orientations in the (110) plane. The results showed that the character of the angular dependence of $u_{\perp}$ depends substantially on the precision of the sample orientation; specifically, on the angle and the plane of misorientation of the crystal. This is illustrated in Figure 3 by the $u_{\perp}(\varphi)$ dependence (φ is the angle between the [001] axis and the field direction) for $H = 20$ kOe. Curves 1 and 2 in this figure were obtained on the same sample in two different experiments.

Curve 1, corresponding to a more precise sample orientation, reflects the symmetry of a cubic crystal in the (110) plane: $u_{\perp}$ reaches extreme values for **H** along the $[1\bar{1}0]$ and

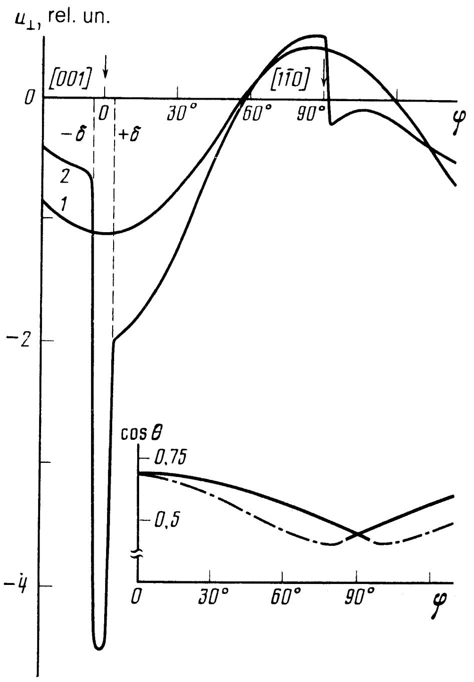

FIG. 3. Angular dependence of the transverse magnetostriction $u_\perp$ (from experiment) and cos θ (θ is the angle between [001] and the vector **n**, $h = 0.6$) for two domains with **n**$\in$(110) (calculation) for **H** lying in the (110) plane. Curves 1 and 2 correspond to different angles of misorientation δ, the dot-dash curve to a metastable domain.

[001] axes and varies monotonically in the interval between these angles. This behavior for $u_\perp(\varphi)$ is in our view connected with the variation in a field of the equilibrium orientation of the triangular spin structure in MnAlG relative to the crystallographic axes. Reconstruction of the domain structure apparently then does not take place.

The $u_\perp(\varphi)$ dependence of the same crystal has a completely different character (curve 2) when there is a large ($\sim 5°$) misalignment of **H**, such that a substantial field exists in the direction of measurement [110]. In this case sharp anomalies are observed in the magnetostriction near [001] and [1$\bar{1}$0]; the largest values of $u_\perp$ in absolute magnitude are reached for **H**$\|$[001]. The experiments show that minima in the $u_\perp(\varphi)$ curve near [001] arise only in fields greater than $H_{cr} = 8$ kOe; their magnitude grows sharply with an increase in H.

The large magnetostriction in MnAlG near [001] is a basis for assuming that this anomaly is connected with a reconstruction of the domain structure of the AF crystal. In fact, a magnetic field in the (110) plane stabilizes domains for which **n** lies in that plane. For **H**$\|$[001] all four types of domain become equivalent, and therefore even a component **H**$\|$[110] which is not large will stabilize the domains for which **n** does not lie in the (110) plane. Such "skewed" domains are stable in the range of angles $1 \pm \delta$ near [001], where δ is the maximum angle of deviation of **H** from the (110) plane.

The size of δ characterizes the half-width of the minimum in the magnetostriction near [001], which, as our measurements show, remains practically constant for all $H \geqslant 8$ kOe. Using the experimental value $\delta \approx 5°$, we can estimate the threshold field for domain structure reconstruction in MnAlG for **H**$\|$[110]: $H_t = H_{cr} \sin\delta = 700$ Oe.

From the dependence of $u_\perp$ on φ it follows that the magnetic states of the crystal for **H** inclined at $\pm\delta$ from [001] (on different sides of the [001] axis) are different; i.e., part of the skewed domains are in a metastable state, vanishing gradually or abruptly in proportion to the increase in φ. In part, the discontinuity in $u_\perp$ near [110] could be related to the loss of stability in the skewed domains, because the sign of the component of **H** along [110] changes.

Another possible reason for the discontinuity of $u_\perp$ for **H** near [110] could be the reconstruction of domains with **n** vectors lying in the same plane as **H**. A basis for this suggestion lies in results of a numerical calculation using the cos $\theta(\varphi)$ dependence of the Lagrangian (θ is the angle between [001] and **n**) for two domains with **n** vectors lying in the same plane as H. From Figure 3, in which the cos $\theta(\varphi)$ dependence for $H = 0.6\ H_c$ is shown (the dot-dash curve corresponds to a metastable domain), we see that near [1$\bar{1}$0] the domains differ from each other, while near [001] they are practically indistinguishable.

Thus, the investigations caried out make it possible to establish the basic parameters characterizing the behavior of the exchange-noncollinear AF in an external magnetic field (anisotropy of the magnetic susceptibility, spin-flop field, threshold field for domain reconstruction) and to show several of the magnetic properties related to the MnAlG domain structure. In conclusion we emphasize again that the domain reconstruction processes in the AF garnet MnAlG in an external field largely determine the magnetoanisotropic properties of the MnAlG crystal and further study will undoubtedly be of interest.

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