

Spontaneous phase transitions and optical anisotropy in manganese germanium garnet ($\text{Ca}_3\text{Mn}_2\text{Ge}_3\text{O}_{12}$)

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A study is made of the linear birefringence Δn in single-domain MnGeG crystals over a wide range of temperatures including the Jahn-Teller phase transition from the cubic to the tetragonal phase ($T_c = 516$ K) and the transition to the magnetically ordered state ($T_N = 13.4$ K). A visual study is made of the twinned domain structure arising at the transition of the crystal to the tetragonal state, and a technique is proposed for coercing thin slabs of MnGeG into a single domain. It is shown that in a MnGeG crystal with a small tetragonal lattice distortion the lattice contribution Δn^0 to the birefringence is proportional to the tetragonal distortion of the crystal: $\Delta n^0 = K(c/a - 1)$, with $K = 4.4$. The $\Delta n^0(T)$ curve is compared with the theory, and it is concluded that one must take into account the anisotropy of the thermal expansion of the crystal in addition to the Jahn-Teller mechanism in order to explain the temperature dependence of the tetragonal distortion of MnGeG. It is shown that the Jahn-Teller transition in MnGeG is a second order phase transition, and the critical exponent β for this transition is found to be 0.5 ± 0.03 . The magnetic contribution Δn_M to the linear birefringence is separated out, and its temperature dependence $\Delta n_M(T)$ near T_N is used to obtain the critical exponent $2\beta = 0.84 \pm 0.04$. It is shown that the temperature derivative of the fluctuation contribution to Δn_M has a logarithmic temperature dependence.

It has been shown^{1,2} that crystals with the garnet structure containing the ions Mn^{3+} or Cu^{2+} exhibit the cooperative Jahn-Teller effect, which leads to a lowering of the symmetry of the crystal from cubic to tetragonal. In the garnet $\text{Ca}_3\text{Mn}_2\text{Ge}_3\text{O}_{12}$ the transition from the cubic phase (space group $Ia3d-O_h^{10}$) to the tetragonal phase (space group $I4_1/a-C_{4h}^6$) occurs at a temperature² $T_c = 525$ K. In manganese germanium garnet (MnGeG) the tetragonal distortion of the lattice and the Jahn-Teller nature of the ion Mn^{3+} give rise to a many-sublattice canted antiferromagnetic structure³ upon magnetic ordering. According to the specific-heat data, in polycrystalline samples⁴ the transition to the magnetically ordered state occurs at a temperature $T_N = 13.85$ K.

In the present paper we study the optical anisotropy arising as a result of the Jahn-Teller transition from the cubic to the tetragonal state in the garnet $\text{Ca}_3\text{Mn}_2\text{Ge}_3\text{O}_{12}$ and follow its changes with decreasing temperature and on the transition of the crystal to the magnetically ordered state. Because the linear birefringence is proportional to the order parameter of the phase transition or to a bilinear function of the order parameter,⁵ one can use polarization optics to study the structural and magnetic phase transitions in the crystals and to determine the critical exponents. Our present study of the temperature dependence of the linear birefringence has yielded new information on the structural and magnetic phase transitions in MnGeG. The ability to make precision measurements of the linear birefringence is especially useful in studying the transition to the tetragonal phase, where the magnitude of the birefringence is directly proportional to the tetragonal distortion of the crystal. Because of the nonferrodistortive nature of the ordering of the local deformations in MnGeG, the macroscopic strain of the

crystal is small (the ratio of the lattice constants at liquid helium temperatures is $c/a \approx 1.003$)² and direct methods of measuring it are less exact. Furthermore, the transition to the tetragonal phase in MnGeG is accompanied by a twinning of the crystal, which complicates experimental techniques which rely on the study of macroscopic characteristics of the sample. Polarization optics permits visual observation and study of the twinned domain structure. We also succeeded in coercing thin slabs of MnGeG into a single-domain state, and these slabs were used in the studies. The measurements of the linear birefringence were made in the coerced (single-domain) samples.

1. SAMPLE PREPARATION AND MEASUREMENT TECHNIQUES

The visual observations of the twinned domain structure and the measurements of the linear birefringence were done in MnGeG slabs 30–50 μm thick, cut perpendicular to the fourfold axis of the cubic crystal. The birefringence of light was measured in single-domain slabs that had been chemically polished in orthophosphoric acid at a temperature of 100 °C. Single-domain samples with a fixed orientation of the C_4 axis were obtained on the transition from the cubic to the tetragonal phase under directional compression or extension of the crystal. For this purpose the slab to be coerced into a single domain was cemented to a substrate made of a material with a markedly different coefficient of thermal expansion from that of MnGeG, e.g., fused quartz; the stresses created on heating were estimated to reach $5 \cdot 10^8$ dyn/cm². Photo 2a in Fig. 1 shows a mechanically polished slab of MnGeG with C_4 axis parallel to the extended surface after being coerced by this method into a single domain. The twinned domain structure which existed in this same sample

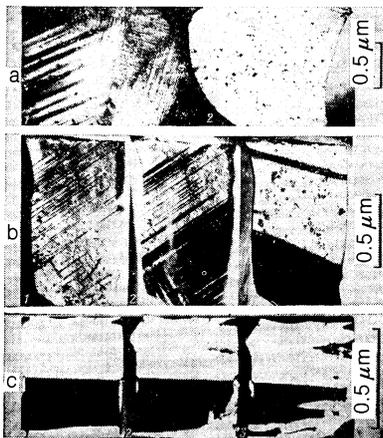


FIG. 1. Twinned domain structure in MnGeG slabs cut in the $(001)_{\text{cub}}$ plane. a) Mechanically polished slab: 1) before coercion into single domain, 2) after coercion; $l = 44 \mu\text{m}$, $T = 295 \text{ K}$. b) Chemically polished slab: 1, 2) initial state; 3) after heating above T_c and rapid cooling; $l = 34 \mu\text{m}$, $T = 295 \text{ K}$. c) Change of the domain structure in a chemically polished slab under uniaxial compression along the $[001]_{\text{cub}}$ direction (photos 2 and 3); photo 1 shows the sample in the initial unstressed state; $l = 36 \mu\text{m}$, $T = 295 \text{ K}$. The scale in the figure is in millimeters.

prior to coercion is shown by photo 1a.

Good results on the coercion of chemically polished samples into a single-domain state were obtained on the transition of unstressed slabs from the cubic to the tetragonal phase at high cooling rates $dT/dt \approx 150 \text{ K/min}$. In this case a slab of area $\sim 1 \text{ mm}^2$ often went into a single domain or contained 2 or 3 large domains (photos 3b and 1c in Fig. 1).

The visual observation and photography of the domain structure were done in polarized white light with the aid of a polarization microscope. The linear birefringence Δn was measured for laser light with $\lambda = 6328 \text{ \AA}$ for $\mathbf{k} \perp C_4$ and $\mathbf{E} \parallel [011]_{\text{cub}}$ by the Sénarmont compensation method. To compensate the phase difference we used a mica quarter-wave plate. The birefringence Δn in MnGeG crystals around $35 \mu\text{m}$ thick could be measured to an accuracy of $\pm 2 \cdot 10^{-6}$. In the measurements of the temperature dependence of the birefringence in MnGeG, such a high accuracy of measuring Δn was necessary only for $T < 20 \text{ K}$, where there was an appreciable magnetic contribution to the linear birefringence and, furthermore, the temperature could be measured to high accuracy. For $T > 20 \text{ K}$ the birefringence Δn was measured to an accuracy of $\pm 5 \cdot 10^{-5}$.

For measurements in the temperature range $6 < T < 300 \text{ K}$ we used an optical helium cryostat. For measuring the linear birefringence and for the visual observation of the twinned domain structure in the region between room temperature and 550 K the sample was placed inside a thermostatic cell. The temperature was measured by means of a copper-constantan thermocouple for $T > 20 \text{ K}$ and by a carbon resistance thermometer for $T < 20 \text{ K}$. The uncertainty in the thermocouple measurements of the temperature was less than 1 K over the entire interval $20 < T < 550 \text{ K}$. In the region $6 < T < 20 \text{ K}$ the error in determining the absolute value of the temperature was less than 0.5 K , while the uncertainty in determining the change in temperature was less than 0.03 K .

2. TWINNED DOMAIN STRUCTURE

In the structural phase transition from the cubic to the tetragonal phase in MnGeG, three types of crystalline domains form, in accordance with the three possible choices of tetragonal axis along one of the fourfold axes of the cubic crystal. Figure 1 shows the twinned domain structure observed in mechanically and chemically polished slabs cut in the $(001)_{\text{cub}}$ plane. The photographs taken in crossed polaroids with $\mathbf{k} \parallel [001]_{\text{cub}}$ and $\mathbf{E} \parallel [110]_{\text{cub}}$ (all the photographs in Fig. 1 except 2b) show dark domains corresponding to regions of the crystal in which the tetragonal axis is perpendicular to the plane of the slab ($C_4 \parallel [001]_{\text{cub}}$). In the bright domains the tetragonal axis lies in the plane of the slab ($C_4 \parallel [100]_{\text{cub}}$ and $C_4 \parallel [010]_{\text{cub}}$). To obtain contrast between the domains with $C_4 \parallel [100]_{\text{cub}}$ and $C_4 \parallel [010]_{\text{cub}}$ a quarter-wave plate was inserted in the optical circuit (photo 2b). The mechanically polished slab of MnGeG exhibited a complex multilayer domain structure as a result of the formation of shallow, "non-through" domains (domains which do not pass all the way through the slab) near the surface, with C_4 axis in the plane of the slab. These domains are due to stresses arising as a result of the disruption of the surface of the crystal by the mechanical polishing. In the chemically polished slabs the domain structure was simpler, often with large "through" domains having boundaries that passed through the slab from one extended surface to the other.

Our visual observations of the domain structure and analysis of the polarization of light transmitted through domains of different types enabled us to determine the orientation of the domain walls in the crystal. In the $(001)_{\text{cub}}$ slabs of MnGeG the walls between domains with $C_4 \parallel [100]_{\text{cub}}$ and $C_4 \parallel [010]_{\text{cub}}$ are perpendicular to the plane of the slab, while the tetragonal axes form an angle of about $\pi/4$ with the plane of the wall. In this case the domain boundaries lie in the $(110)_{\text{cub}}$ or $(\bar{1}10)_{\text{cub}}$ planes. The domain walls separating regions with $C_4 \parallel [001]_{\text{cub}}$ and $C_4 \parallel [100]_{\text{cub}}$ (or $C_4 \parallel [010]_{\text{cub}}$) form an angle of about $\pi/4$ with the plane of the slab and are parallel to the $(101)_{\text{cub}}$, $(\bar{1}01)_{\text{cub}}$, $(011)_{\text{cub}}$, and $(0\bar{1}1)_{\text{cub}}$ planes. Upon the transition from the cubic to the tetragonal phase, planes of the $\{110\}$ type are coherent twinning planes. The domain walls observed in the $(001)_{\text{cub}}$ slabs of MnGeG are coherent.

When the crystal is heated, the contrast between the domains is reduced on account of a decrease in the birefringence, which is proportional to the tetragonal distortion of the crystal. However, the large value of the birefringence in MnGeG permits visual observation of the domain structure in the slabs at temperatures up to $T \approx 510 \text{ K}$ for $T_c = 516 \text{ K}$. Near the transition temperature the ferroelastic properties of MnGeG come into play. In the interval $(T_c - T) < 30\text{--}40 \text{ K}$ we observed motion of the domain boundaries. On slow heating we observed in this same temperature interval the twinning of samples initially in a single-domain state. The twinning of the crystals made it necessary to repeatedly coerce the sample into a single-domain state by rapid cooling during the measurements of the birefringence near T_c . In the immediate vicinity of the transition we were unable to fix the single-domain state of the crystal, and the birefringence was

measured in individual "through" domains. The motion of the domain boundaries and the twinning of single-domain slabs at temperatures close to T_c may be due to internal stresses in the crystal.

Mechanical compression of a chemically polished $(001)_{\text{cub}}$ slab of MnGeG perpendicular to its extended surface at room temperature caused the domain boundaries to move and gave rise to surface domains analogous to those observed in the mechanically polished slabs (photos 2c and 3c in Fig. 1). The threshold pressure for producing the change in the domain structure in this slab at room temperature is about 10^8 dyn/cm². The resulting domain structure remained unchanged when the external pressure was removed.

3. TEMPERATURE DEPENDENCE OF THE LINEAR BIREFRINGENCE

Figure 2 shows the temperature dependence of the linear birefringence $\Delta n(T)$ of MnGeG over a wide range of temperatures including the structural and magnetic phase transitions. The section of the $\Delta n(T)$ curve near T_N is shown in greater detail and in a larger scale in Fig. 3. The birefringence Δn becomes nonzero at the transition of the crystal from the cubic to the tetragonal phase and grows smoothly as the temperature is lowered. The $\Delta n(T)$ curve shown in Fig. 2 exhibits three characteristic regions. As the temperature is lowered in the interval $270 < T < 516$ K, there is initially a rapid increase in Δn , then a slower increase, and finally an approach to saturation. At temperatures $18 < T < 190$ K there is a linear region on the $\Delta n(T)$ curve. Near T_N the birefringence again exhibits a rapid increase, which is naturally attributed to the magnetic ordering of MnGeG.

The complex nature of the $\Delta n(T)$ curve suggests the presence of several contributions to the birefringence. In the high-temperature region $T > (2-3)T_N$ the magnetic contribution to Δn is absent and the birefringence is determined by the lattice contribution Δn^0 . In optically uniaxial crystals

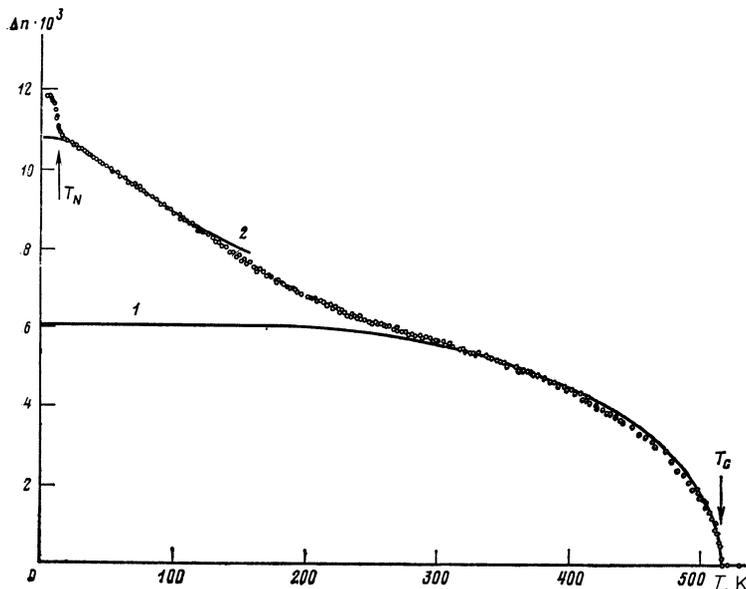


FIG. 2. Temperature dependence of the linear birefringence in MnGeG in a temperature region including the structural ($T_c = 516$ K) and magnetic ($T_N = 13.4$ K) phase transition. Curve 1 is $\Delta n_{\text{IT}}^0(T)$ as calculated from Eq. (3) for $\Delta n_{\text{IT}}^0(0) = 6.1 \cdot 10^{-3}$. Curve 2 is $\Delta n^0(T)$ as calculated from the relation $\Delta n^0(T) = [8.34 - 5.57 \cdot 10^{-4} T^{3/2} + 2.45 \text{th}(50/T)] \cdot 10^{-3}$.

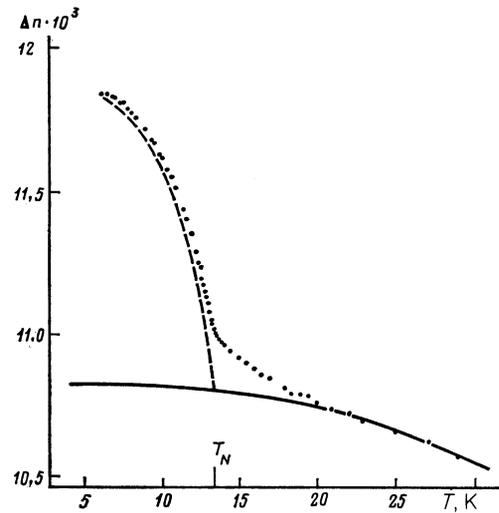


FIG. 3. Temperature dependence of the linear birefringence in the neighborhood of the Néel temperature. The solid curve shows $\Delta n^0(T)$ as calculated from the relation $\Delta n^0(T) = [8.34 - 5.57 \cdot 10^{-4} T^{3/2} + 2.45 \text{th}(50/T)] \cdot 10^{-3}$. The dashed curve was obtained by subtracting from $\Delta n(T)$ the fluctuational magnetic contribution $\Delta n_M'(T)$.

belonging to the intermediate crystal systems (trigonal, hexagonal, or tetragonal), the temperature dependence of the birefringence can be approximated by the expression

$$d(\Delta n^0) = A \frac{da(T)}{a} + C \frac{dc(T)}{c}, \quad (1)$$

where A and C are temperature-independent coefficients. Near the structural phase transition from the cubic to the tetragonal phases, however, one expects that the linear birefringence in the small-strain limit will be directly proportional to the tetragonal distortion of the crystal. Then near T_c the function $\Delta n^0(T)$ in MnGeG can be described more simply than in (1) by the relation

$$\Delta n^0(T) = K\eta(T), \quad (2)$$

where $\eta = (c/a - 1)$ is the degree of tetragonal distortion of the lattice and K is a temperature-independent coefficient of

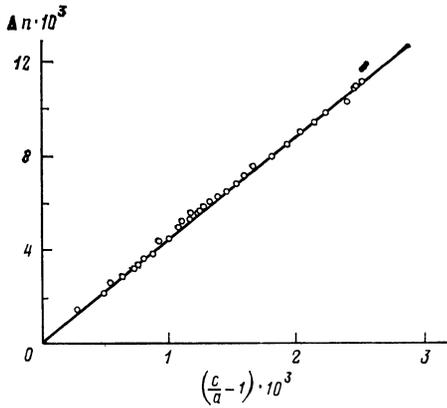
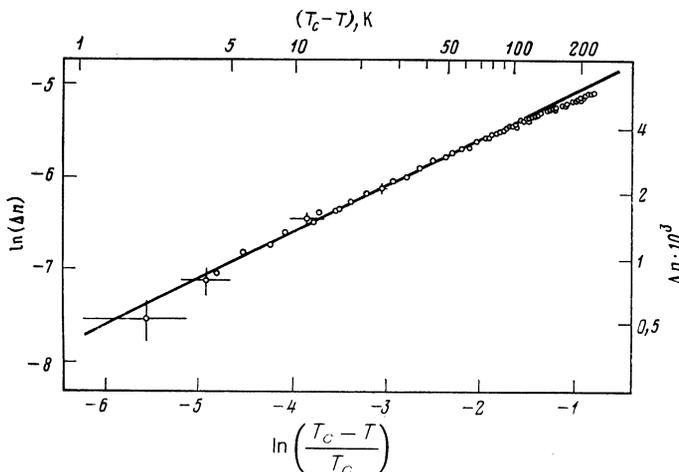


FIG. 4. Linear birefringence versus the degree of tetragonal distortion in MnGeG. The open circles correspond to temperatures $T > T_N$, the filled circles to $T < T_N$.

proportionality. Using the data of x-ray studies² on the temperature dependence of the lattice parameters of MnGeG, one can construct the function $\Delta n(\eta)$ by comparing the values of Δn and η at the same temperatures. The resulting linear behavior of $\Delta n(\eta)$ (Fig. 4) implies that the linear birefringence in MnGeG is directly proportional to the tetragonal distortion of the lattice. Proportionality between Δn and η is not observed just near the phase transition but is maintained over a wide temperature interval $T_N \lesssim T < T_c$. This may be due to the small macroscopic strain of the MnGeG lattice. From the slope of the straight line (Fig. 4) we find the coefficient of proportionality to be $K = 4.4 \pm 0.2$.

It should be noted that proportionality between the temperature-induced changes in the birefringence and the tetragonal distortion of the lattice is not unambiguous evidence of a photoelastic mechanism for the birefringence in MnGeG. The temperature-induced changes in the birefringence could be caused in this case by either a photoelastic or a thermo-optical mechanism, although the latter is not a consequence of the change in the lattice parameter of the crystal but is determined by the isochoric electron-phonon interaction.⁶ Without having measurements of the photoelastic effect we cannot separate the contributions from these two mechanisms to the temperature-induced changes of Δn in MnGeG.



For the transition from the cubic to the tetragonal phase, η can be regarded as the order parameter of the phase transition. The fact that the birefringence and the tetragonal distortion of the crystal are proportional in MnGeG enables one to determine the critical exponent β in the expansion

$$\eta = B[(T_c - T)/T_c]^\beta (1 + \dots)$$

from the temperature dependence of the birefringence $\Delta n(T)$. Figure 5 shows $\ln(\Delta n)$ versus $\ln[(T_c - T)/T_c]$. In the temperature interval $(T_c - T) \lesssim 100$ K the experimental points conform well to a straight line, from the slope of which we find $\beta = 0.5 \pm 0.03$. This value of β is in good agreement with the value of the exponent in the Landau theory of second order phase transitions. The absence of a jump on the $\Delta n(T)$ curve at $T \rightarrow T_c$ and the exponent value $\beta = 0.5$ indicate that the Jahn-Teller phase transition in MnGeG is a second order transition. A dependence $\Delta n(T) \sim \varepsilon^\beta$ with $\beta = 0.5$ [$\varepsilon = (T_c - T)/T_c$] holds well in MnGeG as $T \rightarrow T_c$, at least to temperatures $(T_c - T) \approx 1$ K. Thus, the region in which critical fluctuations play a role near the Jahn-Teller phase transition in MnGeG is narrow, no wider than $\varepsilon \approx 2 \cdot 10^{-3}$. The suppression of critical fluctuation near T_c can be attributed to a long-range nature of the interactions causing the phase transition. Similar behavior has been found for the ferrodistorive Jahn-Teller phase transition in TbVO_4 , for which a critical exponent $\beta = 0.5 \pm 0.02$ was obtained, in contrast to DyVO_4 , where $\beta = 0.34 \pm 0.02$ (Ref. 7).

Because the relation $\Delta n(T) = K\eta(T)$ holds in MnGeG over a wide temperature interval $T_N \lesssim T < T_c$ because the birefringence Δn is measured to greater accuracy than the available values of the tetragonal distortion, it makes sense to compare the $\Delta n(T)$ curve with the theory, after having considered the possible mechanisms for changes in η . Near the transition from the cubic to the tetragonal phase the temperature-induced changes in the parameter η of the crystal are due to the phase transition.

To describe the temperature dependence of the contribution Δn_{JT}^0 to the linear birefringence, let us use the expression obtained in Ref. 8 for the temperature dependence of the deformation of the crystal in the case of a Jahn-Teller phase transition. Then the temperature dependence of Δn_{JT}^0 is given by the equation

FIG. 5. Logarithm of the linear birefringence versus the logarithm of the reduced temperature in MnGeG near the transition from the tetragonal to the cubic phase.

$$\Delta n_{JT}^0(T) = \Delta n_{JT}^0(0) \operatorname{th} \left[\frac{\Delta n_{JT}^0(T) T_c}{\Delta n_{JT}^0(0) T} \right]. \quad (3)$$

The expression we have used for the temperature dependence of the macroscopic strain of the crystal holds rigorously for a ferrodistorptive Jahn-Teller ordering. For a nonferrodistorptive ordering it holds for the sublattice, and the temperature dependence of the macroscopic strain of the crystal can in general be more complex. However, for a nonferrodistorptive ordering the temperature dependence of the macroscopic strain can be described satisfactorily by the above expression if the contributions to the macroscopic strain and the temperature dependences of the individual Jahn-Teller sublattices are identical. Expression (3) gives a good description of the experimentally observed temperature dependence of the linear birefringence in MnGeG in the interval $300 \lesssim T < 516$ K.

Near T_c

$$\frac{\Delta n_{JT}^0(T) T_c}{\Delta n_{JT}^0(0) T} \ll 1$$

and we have the approximate expression

$$\Delta n_{JT}^0(T) \approx 3^{1/2} \Delta n_{JT}^0(0) (1 - T/T_c)^{1/2}. \quad (4)$$

Using the linear part of the curve of $\ln(\Delta n)$ versus $\ln[(T_c - T)/T_c]$ (Fig. 5), we find $\Delta n_{JT}^0(0) \approx 6 \cdot 10^{-3}$. Figure 2 (line 1) shows the curve $\Delta n_{JT}^0(T)$ calculated from (3) with the value $\Delta n_{JT}^0(0) = 6.1 \cdot 10^{-3}$ for which the best agreement is obtained between the calculated curve and the experimental points.

The deviation of the experimental points from the curve calculated from (3) for $T \lesssim 300$ K can be explained by taking into account the contribution of the anisotropy of the thermal expansion to the change in the tetragonal distortion of the crystal. The temperature dependence of the linear birefringence Δn_α^0 , which is proportional to this contribution, can be expressed by the equation

$$\Delta n_\alpha^0(T) = K \int [\alpha_c(T) - \alpha_a(T)] dT, \quad (5)$$

where α_c and α_a are the coefficients of thermal expansion of the crystal along the c and a axes. It is natural to assume that the contribution of the anisotropy of the thermal expansion to the change in η with temperature is small near T_c , where the crystal is almost cubic, and increases with the increase in η as the temperature is lowered. Using the data of Ref. 2, we can estimate the derivative

$$\frac{d}{dT} \Delta n_\alpha^0(T) = K [\alpha_c(T) - \alpha_a(T)]. \quad (6)$$

From the temperature dependence of the lattice parameters of MnGeG, we find the coefficient $\alpha \approx 10^{-5}$ for $T > T_c$. Because the constant c changes little with temperature² for $T < 180$ K and because $\Delta n(T)$ is nearly linear, we can estimate the value of

$$\frac{d}{dT} \Delta n_\alpha^0$$

from (6), assuming $\alpha_c(T) = 0$ and $\alpha_a(T) = \alpha$. We find the value

$$\frac{d}{dT} \Delta n_\alpha^0 \approx -4.4 \cdot 10^{-5} \text{ K}^{-1}.$$

The experimental value is

$$\frac{d}{dT} \Delta n \approx -2.3 \cdot 10^{-5} \text{ K}^{-1}$$

for the linear part of $\Delta n(T)$. Thus the anisotropy of the thermal expansion can give an appreciable contribution (comparable to that of the cooperative Jahn-Teller effect) to the change in the tetragonal distortion of the crystal, and $\Delta n_\alpha^0(T)$ can be comparable to $\Delta n_{JT}^0(T)$ at low temperatures. The growth of Δn with decreasing temperature at $T < 300$ K can be attributed to the contribution of the thermal expansion to the change in η .

The macroscopic strain of the MnGeG lattice due to the cooperative Jahn-Teller effect is small. At liquid-helium temperatures the tetragonal distortion of the crystal reaches a value² of $\eta = 2.6 \cdot 10^{-3}$. By comparison, η reaches a value of 0.2 in spinels containing the Mn^{3+} ion.⁹ In the garnet $\text{NaCa}_2\text{Cu}_2\text{V}_3\text{O}_{12}$, the cooperative Jahn-Teller effect causes a tetragonal distortion of the lattice with $\eta = 1.5 \cdot 10^{-2}$ at liquid-helium temperatures, even though T_c is lower in CuVG (250 K) than in MnGeG. In crystals with a large macroscopic Jahn-Teller lattice distortion the anisotropy of the thermal expansion is imperceptible against the background of the strong temperature dependence of $\eta(T)$ due to the cooperative Jahn-Teller effect. In MnGeG, in contrast, the tetragonal lattice distortion due to the cooperative Jahn-Teller effect is of the same order of magnitude at low temperatures as the change in the tetragonal distortion due to the anisotropy of the thermal expansion. Therefore, in explaining the temperature dependence of $\eta(T)$ in MnGeG one cannot neglect the anisotropy of the thermal expansion. Neglect of the contribution of the anisotropy of the thermal expansion to the temperature-induced changes in η in MnGeG might be the reason for the lack of agreement between the experimental and theoretical $\eta(T)$ curves that was found in Ref. 2. At the same time, in the garnet $\text{NaCa}_2\text{Cu}_2\text{V}_3\text{O}_{12}$, in which this contribution is less noticeable against the background of the larger macroscopic Jahn-Teller distortion of the lattice, the experimental and theoretical $\eta(T)$ curves are found to be in satisfactory agreement.²

4. SPONTANEOUS MAGNETIC BIREFRINGENCE

The onset of magnetic ordering in MnGeG leads to the growth of the linear birefringence in the vicinity of $T_N = 13.4$ K (Figs. 2 and 3). Isolating the magnetic contribution to the linear birefringence always involves difficulties in connection with the extrapolation of the lattice part of the birefringence to the temperature region in which the magnetic ordering is observed. To separate out the magneto-optical contribution to Δn one could extrapolate the lattice part of the birefringence into the temperature region $T \sim T_N$ with the aid of the relation (2) and the data on the change of the lattice parameters with temperature. However, the lack of sufficiently detailed information on how the lattice parameters change with temperature in the neighborhood of T_N does not allow one to separate out the magneto-optical contribution to the required degree of accuracy. An attempt to use the Debye approximation for extrapolating the lattice contribution into the low-temperature region in order to sep-

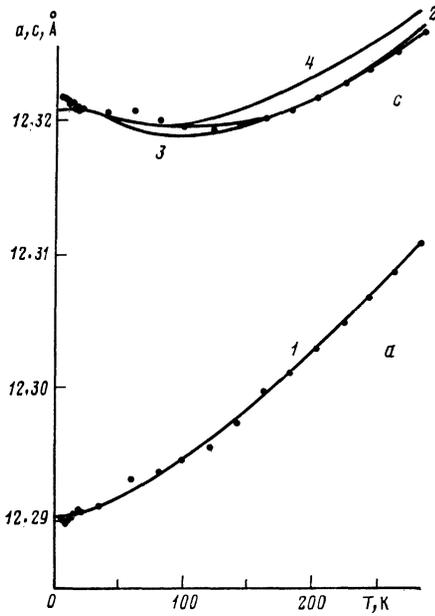


FIG. 6. Temperature dependence of the parameters a and c in MnGeG. The points show the experimental values of a and c from Ref. 2. Curve 1 is the function $a(T) = a(0) + a_1 T^{3/2}$ with $a(0) = 12.2903 \text{ \AA}$ and $a_1 = 4.37 \cdot 10^{-6} \text{ \AA/K}^{3/2}$. Curve 2 was calculated from the relation $c(T) = a(T)[1 + \Delta n(T)/K]$ with $K = 4.36$ and with $a(T)$ corresponding to curve 1. Curves 3 and 4 were calculated from the relation $c(T) = c(0) + c_1 T^{3/2} + c_2 \text{th}(c_3/T)$, where the values for curve 3 are $c(0) = 12.3119 \text{ \AA}$, $c_1 = 2.8 \cdot 10^{-6} \text{ \AA/K}^{3/2}$, $c_2 = 0.0089 \text{ \AA}$, $c_3 = 50 \text{ K}$, and those for curve 4 are $c(0) = 12.3139 \text{ \AA}$, $c_1 = 2.8 \cdot 10^{-6} \text{ \AA/K}^{3/2}$, $c_2 = 0.0069 \text{ \AA}$, $c_3 = 50 \text{ K}$.

arate out the magnetic contribution has also met with failure, indicating that this approximation is too crude for describing the thermal expansion of MnGeG. Therefore, we have attempted to find an empirical law for the change in the lattice parameters a and c with temperature in the region $T \gg T_N$ and, using relation (2), to extrapolate $\Delta n^0(T)$ into the temperature region $T \sim T_N$. The magnetic contribution Δn_M separated out in this way includes not only the magneto-optical contribution but also a magnetostrictive contribution and a magnetic correction to the thermo-optical contribution.

Figure 6 gives the experimental temperature dependence of the lattice parameters a and c of MnGeG from Ref. 2.²⁾ We have found that the temperature dependence of a in the interval $15 < T < 250 \text{ K}$ is described well by the power law $a(T) = a(0) + a_1 T^{3/2}$, where $a(0) = 12.2903 \text{ \AA}$ and $a_1 = 4.37 \cdot 10^{-6} \text{ \AA/K}^{3/2}$ (curve 1 in Fig. 6). By substituting the experimental values of $\Delta n(T)$ and the values of $a(T)$ corresponding to curve 1 in Fig. 6 into Eq. (2), we find the $c(T)$ curve (curve 2 in Fig. 6) that should obtain if Δn is proportional to the tetragonal distortion of the crystal. The experimental points from Ref. 2 are in fair agreement with the calculated curve at a coefficient of proportionality $K = 4.36$. The behavior of $c(T)$ in the interval $15 < T < 250 \text{ K}$ can be described by the equation $c(T) = c(0) + c_1 T^{3/2} + c_2 \text{th}(c_3/T)$ [curve 3 in Fig. 6 was calculated for $c(0) = 12.3119 \text{ \AA}$, $c_1 = 2.8 \cdot 10^{-6} \text{ \AA/K}^{3/2}$, $c_2 = 0.0089 \text{ \AA}$, and $c_3 = 50 \text{ K}$], although the accuracy of such a description is somewhat poorer than that obtained for

the parameter $a(T)$. By varying $c(0)$ and c_2 one can obtain good agreement with curve 2 in the temperature interval $15 < T < 100 \text{ K}$ (curve 4 in Fig. 6). We note that curves 3 and 4 practically coincide for $T < 20 \text{ K}$ [$c^{(4)} - c^{(3)} < 3 \cdot 10^{-5} \text{ \AA}$ for $T < 20 \text{ K}$], where $\text{th}(c_3/T) \approx 1$.

Knowing how the parameters $a(T)$ and $c(T)$ change with temperature, one can use relation (2) to find an expression for the temperature dependence of that part of the linear birefringence which is not due to the magnetic ordering of the crystal. Substituting the values of $a(T)$ and $c(T)$ corresponding to curves 1 and 4 (Fig. 6) into Eq. (2), we find $\Delta n^0(T) = [8.34 - 5.57 \cdot 10^{-4} T^{3/2} + 2.45 \text{th}(50/T)] \cdot 10^{-3}$.

The $\Delta n^0(T)$ curve calculated from this relation is shown in Figs. 2 and 3.

Subtracting from $\Delta n(T)$ the lattice contribution $\Delta n^0(T)$, we get the temperature dependence of the magnetic linear birefringence of MnGeG. The magnetic contribution Δn_M thus isolated contains a part due to the long-range magnetic order and a fluctuational part:

$$\Delta n_M = \kappa \langle s \rangle^2 + \Delta n_M^f. \quad (7)$$

The linear birefringence due to the long-range magnetic order is proportional to the square of the magnetic moment of the sublattice and goes to zero at the Néel point. The fluctuational part, which is due to the short-range magnetic order, has a maximum at $T = T_N$ and decreases with distance from T_N on both sides of this point. Assuming that the temperature dependence of the fluctuational contribution Δn_M^f is not very different for $T < T_N$ and $T > T_N$, one can separate out the contribution to Δn_M due to the long-range magnetic order¹⁰

$$\Delta n_M(T_N - \Delta T) - \Delta n_M(T_N + \Delta T) = B \left(\frac{T_N - T}{T} \right)^{2\beta} (1 + \dots), \quad (8)$$

and determine the critical exponent β . Figure 7 shows a plot of (8) in logarithmic coordinates. For $T_N - T \lesssim 3 \text{ K}$ the points conform well to a straight line, from the slope of which we get $2\beta = 0.84 \pm 0.4$. This value of the critical exponent β is in good agreement with the values of β found from the temperature dependence of the birefringence in the cubic $3d$ Heisenberg antiferromagnets RbMnF_3 , KNiF_3 , and KCoF_3 .^{10,11}

The fluctuational contribution to the magnetic linear birefringence in MnGeG is observed above the Néel point in the temperature region $T - T_N \lesssim 8 \text{ K}$. Figure 7 shows a plot of the derivative

$$\frac{d}{dT} \Delta n_M(T)$$

versus $\ln|(T - T_N)/T_N|$ for $T > T_N$. The experimental points conform well to a straight line over the entire temperature interval in which Δn_M^f is nonzero for above the Néel point. Thus in MnGeG the derivative

$$\frac{d}{dT} \Delta n_M(T)$$

exhibits a logarithmic temperature dependence. No deviation from a logarithmic dependence was observed in the region $T \gtrsim T_N + 0.1 \text{ K}$.

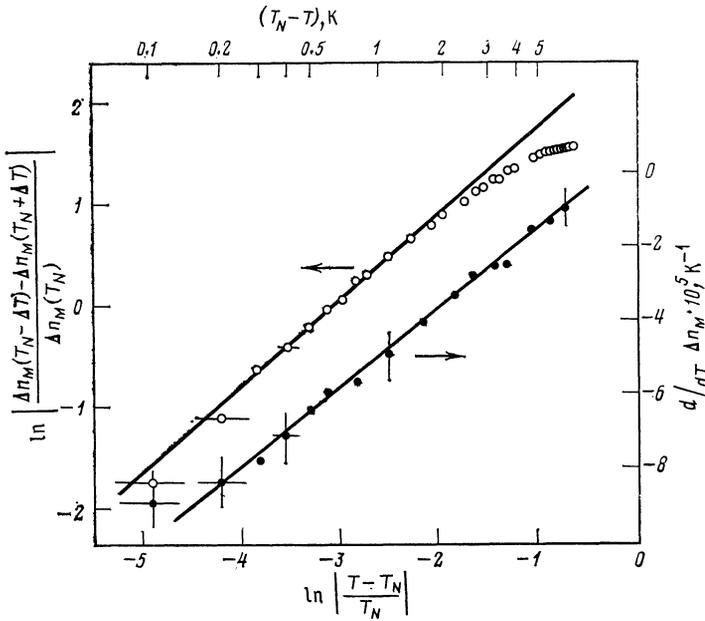


FIG. 7. Logarithm of the contribution to Δn from the long-range order for $T < T_N$ (the open circles) and the temperature derivative of the fluctuational part of the magnetic contribution for $T > T_N$ (filled circles) versus the logarithm of the reduced temperature.

The experimental curve of $\Delta n_M(T)$ for $T > T_N$ was also processed in accordance with the formula

$$\frac{\Delta n_M(T) / \Delta n_M(T_N)}{1 - \Delta n_M(T) / \Delta n_M(T_N)} = B' \left(\frac{T - T_N}{T_N} \right)^\nu, \quad (9)$$

which was proposed for the fluctuational part of the magnetic linear birefringence in Ref. 12. The experimental points on curve (9) plotted in logarithmic coordinates conform to a straight line in the interval $T - T_N \lesssim 3$ K, but the value of the critical exponent of the correlation length found from the slope of the linear region is too high: $\nu = 1.1 \pm 0.2$.

In concluding the section on the magnetic linear birefringence in MnGeG, let us estimate the magneto-optical contribution to Δn_M . We can estimate Δn_M^{MO} from the relation

$$\Delta n_M^{\text{MO}}(T) = \Delta n(T) - K\eta(T)$$

by taking the experimental value (which includes the change in the lattice parameters due to magnetostriction) for the degree of tetragonal distortion of the crystal below the Neel point. At $T = 7$ K we have² $\eta = 2.57 \cdot 10^{-3}$, and $\Delta n_M = 10.1 \cdot 10^{-4}$. Accordingly, $\Delta n_M^{\text{MO}} \approx 5.8 \cdot 10^{-4}$ at $T = 7$ K. This estimate shows that the magneto-optical contribution to the linear birefringence in MnGeG is comparable to the contribution proportional to the magnetostrictive change in $\eta(T)$.

CONCLUSION

In conclusion, let us briefly summarize the main results and inferences of this study.

We have shown by direct visual observation that the transition from the cubic to the tetragonal phase in MnGeG crystals is accompanied by the formation of three types of crystalline twins with tetragonal axes along one of the four-fold axes of the cubic crystal and with coherent domain boundaries lying in twinning planes of the $\{110\}$ type. The twinned domain structure which arises reflects the ferroelas-

tic properties and can change under the influence of a uniaxial stress. We have proposed a technique for coercing thin slabs of MnGeG into a single-domain state.

We have measured the linear birefringence in single-domain slabs of MnGeG over a wide range of temperature including the structural and magnetic phase transitions. We have shown that in MnGeG, a crystal with a small tetragonal lattice distortion, the lattice contribution to the linear birefringence is proportional to the tetragonal distortion of the crystal. We have found the coefficient of proportionality between Δn and η as 4.4 ± 0.2 .

We have shown the Jahn-Teller transition from the cubic to the tetragonal state in MnGeG is a second order transition and have found the value $\beta = 0.5 \pm 0.02$ for the critical exponent of the transition.

We have compared the temperature dependence of the lattice contribution to the linear birefringence with the theory and have demonstrated the necessity of taking into account the anisotropy of the thermal expansion of the crystal in addition to the Jahn-Teller mechanism in explaining the temperature dependence of the tetragonal distortion of MnGeG.

We have separated out the magnetic contribution to the linear birefringence. By processing the magnetic contribution due to the long-range magnetic order, we were able to determine the critical exponent $2\beta = 0.84 \pm 0.04$ for the transition to the magnetically ordered state. We have shown that the temperature derivative of the fluctuational part of the magnetic contribution has a logarithmic temperature dependence.

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