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Effect of light illumination on antiferromagnet–metamagnet phase transitions in the garnet $Ca_3Mn_2Ge_3O_{12}$

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The effect of linearly polarized light illumination on the metamagnetic phase transition in the antiferromagnetic garnet $Ca_3Mn_2Ge_3O_{12}$ is studied. The crystal is exposed to light propagating both along the tetragonal axis [001] and along the [100] direction. In both cases, a change of the field H_t of the metamagnetic phase transition is observed under illumination, and this change depends on the orientation of plane of polarization of the light with respect to the crystal axes. In the first case, $\mathbf{k} \parallel \mathbf{H} \parallel [001]$, the value of H_t decreases on exposure to light with the polarization $\mathbf{E} \parallel [110]$ and increases on exposure to light with the polarization $\mathbf{E} \parallel [1\bar{1}0]$. In the second case, $\mathbf{k} \parallel \mathbf{H} \parallel [100]$, the value of H_t decreases irrespective of the orientation of the plane of polarization of the light with respect to the crystal axes. However, the magnitudes of the change of H_t are different for light with the polarization $\mathbf{E} \parallel [011]$ and with the polarization $\mathbf{E} \parallel [0\bar{1}1]$. The change of the field of the metamagnetic phase transition in the second case was much larger than in the first case. A phenomenological theory of the photomagnetic effects observed in the antiferromagnetic garnet $Ca_3Mn_2Ge_3O_{12}$ is developed. It is shown that the effect of light illumination on the metamagnetic phase transition is related to the photoinduced magnetic moment in this antiferromagnet. The magnetic moment induced by linearly polarized light in the garnet $Ca_3Mn_2Ge_3O_{12}$ is detected experimentally by means of a SQUID magnetometer.

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Introduction

The study of possibility of controlling the magnetic state of crystals by using light illumination is attracting interest in terms of both a better insight into the nature of the effect and its application. Light-induced phase transitions have been observed in some magnetically ordered crystals [1]. For instance, the linearly polarized light illumination of yttrium iron garnets results in a spin-reorientation phase transition as a result of the photoinduced change of crystalline magnetic anisotropy [2,3]. The changes in magnetic and electronic states in response to light irradiation was recently observed in manganites with colossal magnetoresistance [4–7]. In these compounds the light induces a phase transition from an insulating antiferromagnetic (AFM) state to a conducting ferromagnetic one. At the moment, the mechanism of the light-induced phase transition in manganites remains unclear. The transition is thought to be a associated with the photoinduced melting of the charge-ordered state caused by optical transitions with charge transfer between Mn^{3+} and Mn^{4+} ions.

The light-induced modification of the magnetic state related to the redistribution of the Mn^{3+} and Mn^{4+} ions in the crystal lattice due to the photoinduced charge transfer between the ions was recently observed in another manganese oxide, namely in the AFM garnet $Ca_3Mn_2Ge_3O_{12}$ [8]. Linearly polarized light alters the field of the metamagnetic (MM) phase transition. The effect of light on the phase transition in the garnet $Ca_3Mn_2Ge_3O_{12}$ is related to the light-induced magnetic moment that arises in the AFM state.

The mechanisms of the photoinduced phase transitions in the above manganese oxides have some common features; in particular, the optical transitions with charge transfer between the Mn^{3+} and Mn^{4+} ions are of great importance. A comprehensive elucidation of the mechanisms of the photoinduced phase transitions in manganese oxides is of obvious interest for the physics of photoinduced phenomena in magnets and is also necessary for application of these compounds. The photoinduced effects observed in manganese oxides could be used in devices for the storage and processing of information.

The paper reports experimental data on the effect of illumination on the first-order MM phase transitions induced by magnetic field in the AFM garnet $Ca_3Mn_2Ge_3O_{12}$. The experiments were carried out for two directions of propagation of the inducing light and two directions of magnetic field with respect to the crystallographic axes: along the tetragonal axis [001] ($\mathbf{k} \parallel \mathbf{H} \parallel$ [001]) and normal to this axis, i.e., along the crystallographic direction [100] ($\mathbf{k} \parallel \mathbf{H} \parallel$ [100]). It was found that the effect of light on the MM phase transition in the above two cases differs substantially. A phenomenological theory has been developed to describe the observed photoinduced effects.

Experimental technique

The effect of light irradiation on the first-order MM phase transitions in calcium–manganese–germanium garnet (CaMnGeG) was investigated by means of magnetooptical and magnetometric techniques. The field dependences of the angle of rotation of the plane of polarization of the light and the field dependences of the magnetization were measured. Visual observations of the two-phase domain structure formed during the MM phase transition were also performed. The samples under study were plates of several tens of microns in thickness. The single crystal plates were cut perpendicular to a direction of type [100]. The elastic stresses generated in the plate surface layers due to mechanical polishing were removed by annealing at a temperature of about 1000 °C as well as by chemical polishing in orthophosphoric acid. It is known that the CaMnGeG crystals display the Jahn–Teller phase transition from the cubic to a tetragonal phase at $T \approx 500$ K [9]. This transition results in the formation of crystal twins in the low-symmetry phase [9–11]. A special thermal treatment was used to obtain single-domain samples [10]. As a result of the treatment, single-domain plates with the tetragonal axis oriented perpendicular or parallel to the plate surface were obtained.

In the magnetooptical and visual experiments the sample was placed on a holder in an optical helium cryostat and was kept in vacuum. Temperature was measured by means of a resistance thermometer with an accuracy of about 0.1 K. A superconducting magnet produced a magnetic field that was perpendicular to the plate surface and parallel to the direction of light propagation.

The optical scheme of the experimental setup for visual observation of the two-phase domain structure is shown in Fig. 1,*a*. The light from filament lamp (1) passes through polarizer (5), the sample (7), and analyzer (9). The sample image is projected by the objective (8) onto the photocathode of a TV camera (10) and is displayed on a monitor (11) and saved by a videorecorder (12). To ensure that the magnetic state of the crystal under study remains unchanged, the light flux density is decreased to 0.01 W / cm^2 by means of the filters. To investigate the effect of light illumination on the MM phase transition, the sample is exposed to the light of a helium–neon laser (15) with a wavelength $\lambda = 633$ nm and a light flux density of about 0.1 W / cm^2. The optical system is also equipped with rotary mirrors (13) and (14) and with a field diaphragm (3) having a certain shape which allows local irradiation of a chosen area of the sample. The diaphragm image on the sample surface is formed by the lens (4).

When the field dependences of the angle of rotation of the plane of polarization were measured,

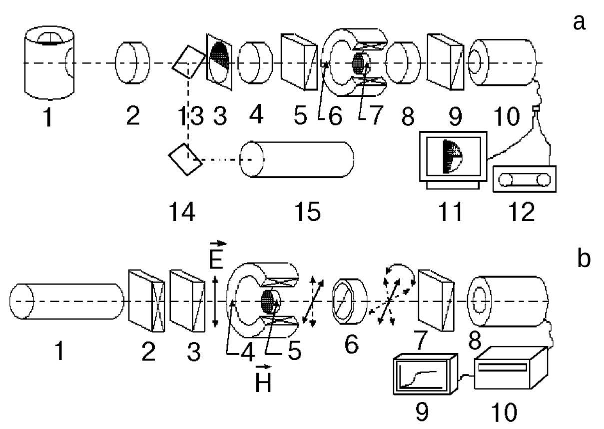

Fig. 1. The optical system of the experimental setup for visual observation of the domain structure: 1 — filament lamp; 2, 4, 8 — lenses; 3 — field diaphragm; 5 — polarizer; 6 — solenoid; 7 — sample; 13, 14 — mirrors; 15 — helium–neon laser (*a*). The optical system for measuring the angle of rotation of the plane of polarization of the ligth: 1 — helium–neon laser; 2 — light attenuator; 3 — polarizer; 4 — solenoid; 5 — sample; 6 — modulator; 7 — analyzer; 8 — photomultiplier; 9 — recorder; 10 — lock-in amplifier (*b*).

it should have been taken into account that several magnetooptical effects (the Faraday effect, Cotton–Mouton effect, and the linear magnetooptical effect (LMOE) [12]) arise in a magnetic field in the antiferromagnet CaMnGeG. Therefore, the incident linearly polarized light became elliptically polarized even in the case when it passed through the crystal along the tetragonal axis. In our experiments we measured the angle of rotation Φ of the ellipse axis of the transmitted light with respect to the plane of polarization of the incident light. The angle Φ depended on the Faraday rotation and linear birefringence. When the measuring beam of light passed through the crystal along the [100] direction, i.e., perpendicular to the tetragonal axis, the plane of polarization of the incident light was chosen parallel to the crystallographic direction [010] or [001]. In this case, the contribution of crystalline birefringence to the modification of the polarization of the transmitted light was minimal. As a rule, the light ellipticity was slight (about 1°) in our experiments. That made possible to use a modulation technique with light modulation in the plane of polarization and synchronous detection of the signal to measure the angle Φ (Fig. 1,*b*). The measurement of field dependences $\Phi(H)$ and the light illumination of the sample were carried out with the use of helium–neon laser of wavelength $\lambda = 633$ nm. The light flux density used for illumination of the sample was about 0.1 W / cm^2. To measure the field dependences $\Phi(H)$, the flux density of the laser beam was attenuated down to 0.01 W / cm^2.

The field dependences of the magnetization and photoinduced magnetic moment were measured by means of a commercial SQUID magnetometer (Quantum Design MPMS-5).

Experimental results

Before studying the effect of light illumination on the MM phase transitions, we examined these transitions in an unexposed crystal. The data from the magnetooptical and visual studies were used to construct the *H–T* magnetic phase diagrams of CaMnGeG for two orientations of external magnetic field: **H** ∥ [001] and **H** ∥ [100] (Fig. 2). In both cases the transition from the AFM to the MM state is a first-order phase transition at low temperatures. For **H** ∥ [100] the first-order AFM–MM

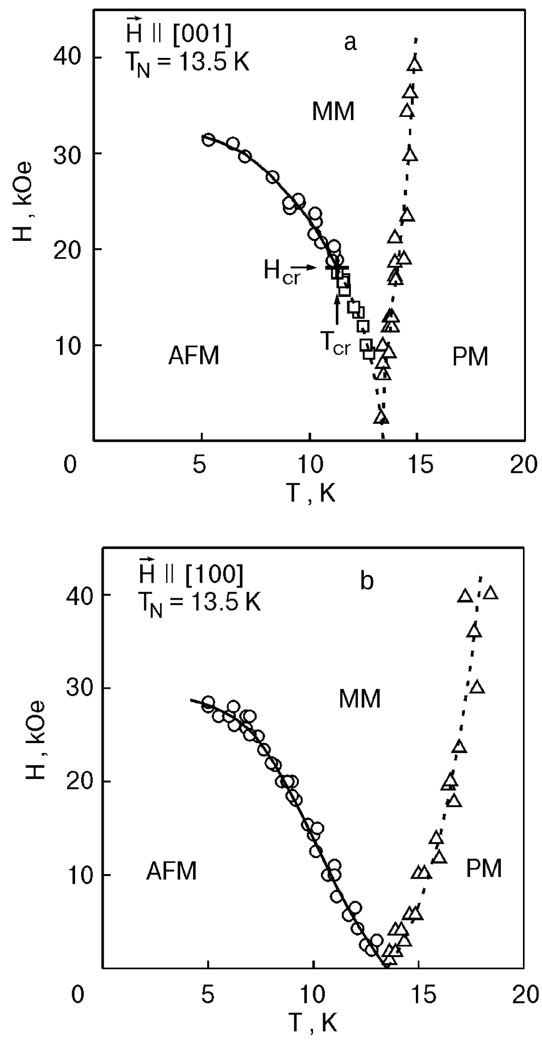

Fig. 2. Magnetic phase diagrams of the garnet $Ca_3Mn_2Ge_3O_{12}$ for **H** ∥ [001] (*a*) and **H** ∥ [100] (*b*). The solid and dotted lines correspond to the first- and second-order phase transitions, respectively.

phase transition is observed in the whole temperature range $T < T_N \approx 13.5$ K where T_N is the Neel temperature. In the case **H** ‖ [001], the H–T phase diagram exhibits a critical point, $H_{cr} \approx 18$ kOe, $T_{cr} \approx 11.5$ K, at which the line of first-order phase transitions passes into a line of second-order ones. Because the effect of light illumination on the first-order MM phase transition was studied, in the case **H** ‖ [001] the experiments were performed at $T < T_{cr}$.

In the first experiment we studied the effect of light irradiation on the MM phase transition induced by a magnetic field **H** ‖ [001]. In this case the directions of the inducing light and the measuring light beam were parallel to the [001] crystal axis. The effect of light on the MM phase transition was first studied visually. For this purpose a single domain AFM state was prepared by applying a magnetic field [12]. The process of monodomainization was monitored visually through the LMOE. After the process was completed and the magnetic field was switched off, the upper half of the sample was exposed to laser light with the polarization **E** ‖ [110] whereas the lower one to light with the polarization **E** ‖ [$1\bar{1}0$]. Thereupon we observed visually the magnetic-field-induced phase transition from the AFM to the MM state. The two-phase domain structure formed during the transition in the exposed sample is shown in Fig. 3. The AFM–MM interphase wall is indicated by the dashed line, and the boundary between the crystal parts exposed

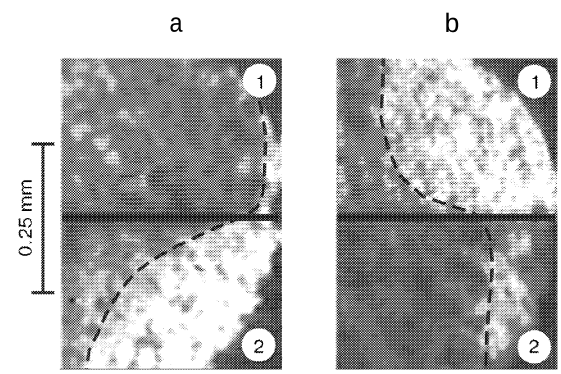

Fig. 3. The two-phase domain structure formed in the $Ca_3Mn_2Ge_3O_{12}$ plate during the MM phase transition after exposure of the crystal to linearly polarized light. The AFM–MM interphase wall is denoted by a dashed line. The sample temperature $T = 7$ K; the applied magnetic field **H** ‖ [001]. Part 1 (above the solid heavy line) exposed to light with the polarization **E** ‖ [110] and part 2 (below the solid heavy line) to light with the polarization **E** ‖ [$1\bar{1}0$] (*a*); part 1 exposed to light with the polarization **E** ‖ [$1\bar{1}0$] and part 2 to light with the polarization **E** ‖ [110] (*b*).

to the light with the different polarizations is shown by the solid heavy line. As is evident from Fig. 3,*a*, the transition from the AFM to the MM state in the upper part of the sample occurs before that in the lower part. To be sure that the difference in the transition fields between the upper and lower parts of the sample is caused by the light illumination and not accidental factors (internal mechanical stresses, temperature gradient, etc.), in the second stage of the experiments the upper part of the sample was exposed to light with the polarization **E** ‖ [$1\bar{1}0$] and the lower one to light with the polarization **E** ‖ [110]. In this case we observed the inverse effect, namely, the magnetic-field-induced phase transition from the AFM to the MM state in the lower part of the sample occurs before that in the upper part (Fig. 3,*b*). Thus the visual observation permits us to conclude that exposure of the crystal to light with the polarization **E** ‖ [110] stimulates the MM phase transition in the garnet $Ca_3Mn_2Ge_3O_{12}$, while exposure to light with the polarization **E** ‖ [$1\bar{1}0$] inhibits the transition.

To determine the value of the photoinduced change of the phase transition field, ΔH_t , we measured the field dependences of the angle rotation, $\Phi(H)$, shown in Fig. 4. The dependences were measured in the same area of the sample (~ 100 μm in diameter) exposed first to light with the polarization **E** ‖ [110] and then to light with the polarization **E** ‖ [$1\bar{1}0$]. In both cases the intensity and the duration of the exposure were the same. The jump in the $\Phi(H)$ curve (Fig. 4) corresponds to a first-order MM phase transition. The transition is accom-

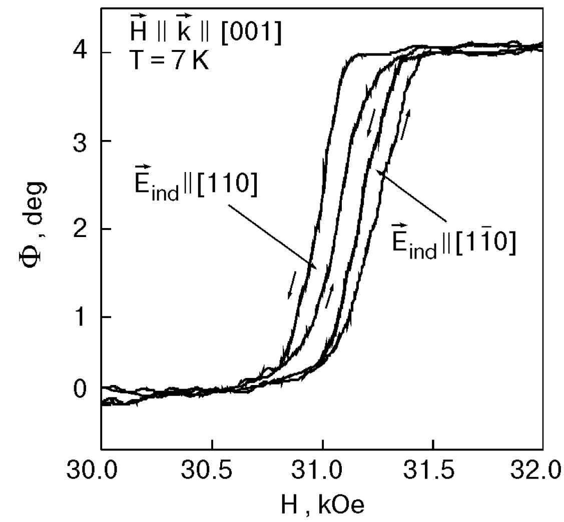

Fig. 4. The field dependences of the rotation angle measured in the case **k** ‖ **H** ‖ [001] in a sample area of about 100 μm in diameter exposed beforehand to linearly polarized light with the polarization **E** ‖ [110] or **E** ‖ [$1\bar{1}0$]. The sample temperature $T = 7$ K.

panied by relatively small hysteresis. The difference in transition fields, $2\Delta H_t = H_{t2} - H_{t1}$, between the cases of exposure of the crystal to light with polarization **E** ∥ [110] and **E** ∥ [1$\bar{1}$0] is about of 180 Oe at the temperature $T = 7$ K, i.e., $\Delta H_t \approx 90$ Oe. The values H_{t1} and H_{t2} were determined from the midpoint of the section of sharp change of the rotation angle on the curve $\Phi(H)$ measured with an increasing (or decreasing) magnetic field for **E** ∥ [110] and **E** ∥ [1$\bar{1}$0] ($H_t = (H' + H'')/2$ (see Figs. 5 and 6)). The MM phase transition field in the unexposed sample, H_t, was equal to ~ 31.2 kOe. This is close to the value of $(H_{t1} + H_{t2})/2$.

In the second experiment we investigated the effect of light illumination on the MM phase transition induced by a magnetic field **H** ∥ [100]. In this case the studies were performed by means of magnetooptical and magnetometric techniques. The field dependences of the angle of rotation of the plane of polarization and the field the dependences of the magnetization were measured. The direction of propagation of the inducing light was parallel to the crystal axis [100]. In the magnetooptical experiments, the direction of propagation of the measuring light beam was also parallel to the [100] axis.

At **k** ∥ **H** ∥ [100] we also observed a photoinduced change of the MM phase transition field. The value $\Delta H_t = H_{t1} - H_{t0}$ ($H_t = (H' + H'')/2$) also depended on the polarization of the inducing light. However, exposure to light with any linear polarization always resulted in a decrease of the MM transition field. The field dependences of the rotation angle, $\Phi(H)$, measured at the temperature $T = 11$ K, are shown in Fig. 5. The jump in the $\Phi(H)$ curves corresponds to the MM phase transition. As it is evident from Fig. 5, after exposing the crystal to light with the polarization **E** ∥ [011] and **E** ∥ [0$\bar{1}$1], the phase transition occurs at a lower field that in the unexposed crystal. The photoinduced decrease in the transition field for **E** ∥ [0$\bar{1}$1] was larger than that for **E** ∥ [011]. It is noted that the different magnitudes of the rotation angle Φ in the MM state on the curves shown in Fig. 5 are related to the appearance of photoinduced linear birefringence in the illuminated crystal [13].

Figure 6 shown the field dependences of the magnetization measured at the temperature $T = 11$ K in the unexposed sample and in the sample exposed to light with the polarization **E** ∥ [011]. It is also seen in Fig. 6 that a decrease of the MM transition field is observed after illumination of the sample.

In the case **E** ∥ [0$\bar{1}$1] (larger magnitude of ΔH_t), the values of the photoinduced changes in the transition field, ΔH_t, at different temperatures were determined from the field dependences, $\Phi(H)$, measured in the temperature range 7–13 K. The dependence $\Delta H_t(T)$ is shown in Fig. 7. As is seen in Fig. 7, the value of ΔH_t increases with decreasing temperature, peaks at $T = 10.5$–11 K, and then decreases with further reduction in temperature. In the case under consideration the maximum value of ΔH_t is about of 1.2 kOe. This is much larger than in

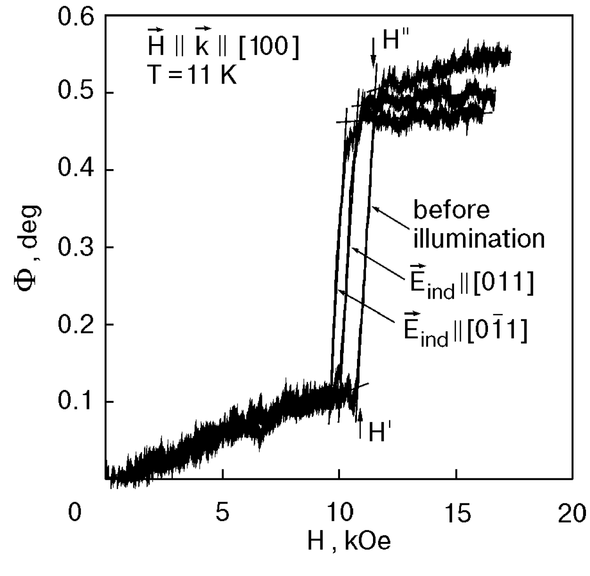

Fig. 5. The field dependences of the rotation angle measured in the case **k** ∥ **H** ∥ [100] in a sample area about 100 μm in diameter exposed beforehand to linearly polarized light with the polarization **E** ∥ [011] or **E** ∥ [0$\bar{1}$1] as well as in the unexposed sample. The temperature of the sample $T = 11$ K.

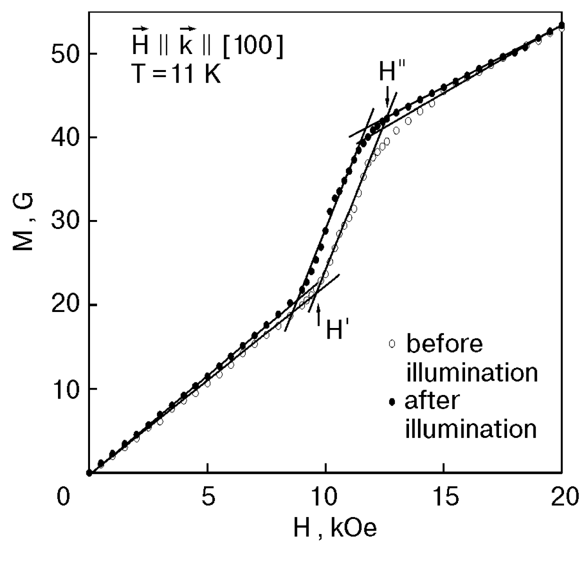

Fig. 6. The field dependences of the magnetization measured in a field **H** ∥ [100] at a temperature $T = 11$ K. ❍ — unexposed sample; ● — the sample was preliminarily exposed to linearly polarized light with polarization **E** ∥ [011]. The direction of propagation of the inducting light **k** ∥ [100].

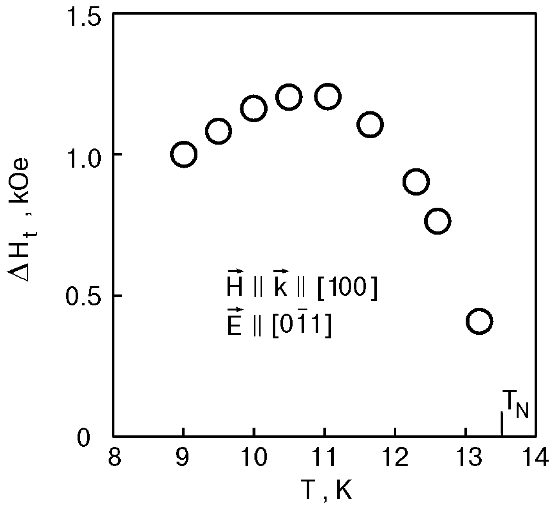

Fig. 7. The temperature dependence of the change in the MM phase transition field caused by light irradiation in the case **k** || **H** || [100] and **E** || [0$\bar{1}$1].

the previous case where the inducing light propagated along the tetragonal axis. The sign of the ΔH_t remains unchanged in the whole temperature range studied.

Discussion

The experimental investigations demonstrated that the exposure of the garnet $Ca_3Mn_2Ge_3O_{12}$ to linearly polarized light propagating along [001] the direction resulted in an increase or a decrease in the MM phase transition field, depending on the polarization of the inducing light. On exposure of the crystal to light propagating along [100] the direction, the MM phase transition field decreases without regard to the of the polarization inducing light. The reason why the light irradiation affects the MM phase transition in the CaMnGeG may be magnetic moment stimulation by the light. Depending on the direction of the photoinduced magnetic moment, the AFM phase in the magnetic field is more or less stable in the exposed crystals than in the unexposed crystal, and that results in the variation in the MM phase transition field.

Let us consider a phenomenological model for the appearance of a magnetic moment in this garnet under illumination. It is known that on exposure of the crystal to light, its internal energy per unit volume varies as follows [14]:

$$\Delta F = -(1/16\pi)\,[d(\omega\varepsilon_{ik})/d\omega]\; E_i\, E_k\,, \qquad (1)$$

where ω is the light frequency, E is the electric field strength of the light wave, and ε_{ik} is the dielectric tensor of the crystal. The change in the internal energy under illumination can generate a light-induced magnetic moment ${}^{ph}\mathbf{m}$ in the crystal. The value and the direction of the photoinduced magnetic moment are dependent on the polarization of the inducing light as well as on the crystalline and magnetic symmetry of the crystal [15,16]. Using (1), we can derive an expression for ${}^{ph}\mathbf{m}$. To do this, we first expand the ΔF series in **H**, restricting to the second-order terms, and then we differentiate the resulting expression with respect to **H**. As a result we obtain:

$${}^{ph}m_l = C_{lik}\, E_i\, E_k + B_{lmik}\, H_m\, E_i\, E_k\,, \qquad (2)$$

where C_{lik} and B_{lmik} are the first and the second derivatives of the expression $(1/16\pi)[d(\omega\varepsilon_{ik})/d\omega]$. Using (2), we can derive an expression for the magnetic moment ${}^{ph}\mathbf{m}$ induced by linear by polarized light propagating along the [001] direction. For this purpose, we rewrite Eq. (2) in the following form:

$${}^{ph}m_l = {}^{ph}m_l^{(1)} + {}^{ph}m_l^{(2)}\,. \qquad (3)$$

In this expression ${}^{ph}m_l^{(1)} = C_{lik}\, E_i\, E_k$, where C_{lik} is an axial c-tensor symmetric in i and k, and ${}^{ph}m_l^{(2)} = B_{lmik}\, E_i\, E_k\, H_m = {}^{Ph}\Delta\chi_{lm} H_m$, where B_{lmik} is a polar i-tensor symmetric on two pairs of indices i, k and l, m. Let us obtain first ${}^{ph}m_l^{(1)}$. Taking into account that CaMnGeG belongs to the point magnetic group $4'/m$, the tensor matrix C_{lik} can be written in the form:

$$C_{l\mu} = \pm \begin{bmatrix} 0 & 0 & 0 & C_{14} & C_{15} & 0 \\ 0 & 0 & 0 & -C_{15} & C_{14} & 0 \\ C_{15} & -C_{15} & 0 & 0 & 0 & C_{14} \end{bmatrix}. \qquad (4)$$

The reduction of indices was used in (4). In this expression, «+» and «–» correspond to the two time-inverted antiferromagnetic states, AFM^+ and AFM^-, and $C_{15} = C_{xxz} = C_{yyz} = C_{xzx}$ and $C_{14} = C_{yzx} = C_{xzy} = C_{xyz}$. If the inducting light propagates along the [001] direction, the components ${}^{ph}\, m_l^{(1)}$ can be written as follows:

$$\begin{aligned} {}^{ph}\, m_x^{(1)} &= 0 \\ {}^{ph}\, m_y^{(1)} &= 0 \\ {}^{ph}\, m_z^{(1)} &= \pm\,(C_{zxx}E_xE_x - C_{zyy}E_yE_y + 2C_{zxy}E_xE_y)\,. \end{aligned} \qquad (5)$$

To determine the second term ${}^{ph}\, m_l^{(2)}$ in Eq. (3), we calculate ${}^{ph}\Delta\chi_{lm}$ taking into consideration the fact that the Laue crystal class of the garnet under study is C_4 :

$$\chi_{lm} = \begin{bmatrix} B_{11} & B_{12} & B_{13} & 0 & 0 & B_{16} \\ B_{12} & B_{11} & B_{13} & 0 & 0 & -B_{16} \\ B_{31} & B_{31} & B_{33} & 0 & 0 & 0 \\ 0 & 0 & 0 & B_{44} & B_{45} & 0 \\ 0 & 0 & 0 & -B_{45} & B_{44} & 0 \\ B_{61} & -B_{61} & B_{63} & 0 & 0 & B_{66} \end{bmatrix} \begin{bmatrix} E_1 & E_1 \\ E_2 & E_2 \\ 0 & \\ 0 & \\ 0 & \\ 2E_1 & E_2 \end{bmatrix} =$$

$$= \begin{bmatrix} B_{11}E_1^2 + B_{12}E_2^2 + 2B_{16}E_1E_2 & B_{61}E_1^2 - B_{61}E_2^2 + 2B_{16}E_1E_2 & 0 \\ & B_{12}E_1^2 + B_{11}E_2^2 - 2B_{16}E_1E_2 & 0 \\ & & B_{31}E_1^2 + B_{31}E_2^2 \end{bmatrix}. \quad (6)$$

Then, using (6), we can derive an expression for the magnetic moment ${}^{ph}m_l^{(2)}$:

$${}^{ph}m_l^{(2)} = \begin{bmatrix} B_{11}E_1^2 + B_{12}E_2^2 + 2B_{16}E_1E_2 & B_{61}E_1^2 - B_{61}E_2^2 + 2B_{16}E_1E_2 & 0 \\ & B_{12}E_1^2 + B_{11}E_2^2 - 2B_{16}E_1E_2 & 0 \\ & & B_{31}E_1^2 + B_{31}E_2^2 \end{bmatrix} \begin{bmatrix} H_x \\ H_y \\ H_z \end{bmatrix}, \quad (7)$$

where

$$\begin{aligned} {}^{ph}m_x^{(2)} &= (B_{xxxx}E_x^2 + B_{xxyy}E_y^2 + 2B_{xxxy}E_xE_y)H_x + (B_{xyxx}E_x^2 - B_{xyxx}E_y^2 + 2B_{xxxy}E_xE_y)H_y \\ {}^{ph}m_y^{(2)} &= (B_{xxyy}E_x^2 + B_{xxxx}E_y^2 - 2B_{xxxy}E_xE_y)H_y + (B_{xyxx}E_x^2 - B_{xyxx}E_y^2 + 2B_{xxxy}E_xE_y)H_x \\ {}^{ph}m_z^{(2)} &= B_{zzxx}(E_x^2 + E_y^2)H_z\ . \end{aligned} \quad (8)$$

It should be noted that the matrix of the tensor B_{lmik} is determined only by the crystal symmetry of CaMnGeG. Therefore, the direction of the moment ${}^{ph}m_l^{(2)}$ is independent of the magnetic state of the crystal. If we know the expressions for ${}^{ph}m_l^{(1)}$ and ${}^{ph}m_l^{(2)}$, we can determine the magnetic moment ${}^{ph}\mathbf{m}$ induced by light propagating along the [001] direction. Upon introducing the azimuthal angle φ measured from the axis $\mathbf{x} \parallel [100]$, the components of this magnetic moment, ${}^{ph}m_x$, ${}^{ph}m_y$, ${}^{ph}m_z$, can be given as follows:

$${}^{ph}m_x = (B_{xxxx}\cos^2\varphi + B_{xxyy}\sin^2\varphi + B_{xxxy}\sin 2\varphi)|E|^2H_x + H_y(B_{xyxx} + B_{xxxy})|E|^2(\sin 2\varphi + \cos 2\varphi)\ ;$$

$${}^{ph}m_y = (B_{xxyy}\cos^2\varphi + B_{xxxx}\sin^2\varphi - B_{xxxy}\sin 2\varphi)|E|^2H_y + H_x|E|^2(B_{xyxx}\cos 2\varphi + B_{xxxy}\sin 2\varphi)\ ; \quad (9)$$

$${}^{ph}m_z = B_{zzxx}|E|^2H_z \pm |E|^2(C_{zxx}\cos 2\varphi + C_{zxy}\sin 2\varphi)\ .$$

The signs «±» correspond to the antiferromagnetic states AFM$^+$ and AFM$^-$. At $\mathbf{k} \parallel \mathbf{H} \parallel [001]$, field components H_x and H_y are equal zero. Therefore, ${}^{ph}m_x = 0$, ${}^{ph}m_y = 0$ and ${}^{ph}m_z = B_{zzxx}|E|^2H_z \pm$ $\pm |E|^2(C_{zxx}\cos 2\varphi + C_{zxy}\sin 2\varphi)$ in this case.

If the inducing light propagates along the [100] direction, the induction of a magnetic moment is also allowed by the symmetry. Repeating the line of calculation made for the previous case, we can derive expressions for the photoinduced magnetic moment components:

$${}^{ph}m_x = (H_xB_{xxxx} + H_yB_{xyxx})|E|^2\cos^2\varphi + H_yB_{xxzz}|E|^2\sin^2\varphi + (H_zB_{xzxz} \pm 2C_{xxz})|E|^2\sin 2\varphi\ ;$$

$${}^{ph}m_y = [(H_xB_{xyxx} + H_yB_{yyxx})|E|^2\cos^2\varphi + H_yB_{yyzz}|E|^2\sin^2\varphi] + (H_zB_{yzxz} \pm 2C_{yxz})|E|^2\sin 2\varphi\ ; \quad (10)$$

$${}^{ph}m_z = H_y B_{zzxx}|E|^2 + (H_y B_{yzxz} + H_x B_{xzxz})|E|^2\sin 2\varphi \pm C_{zxx}|E|^2\cos^2\varphi\ .$$

Here, the angle φ is measured from the axis **y** ∥ [010], and the signs «±», as in the previous case, correspond to the two antiferromagnetic states AFM^+ and AFM^-. At **k** ∥ **H** ∥ [100], field components H_y and H_z are equal zero. Therefore, ${}^{ph}m_x = H_x B_{xxxx}|E|^2 \cos^2\varphi \pm 2C_{xxz}|E|^2 \sin 2\varphi$, ${}^{ph}m_y = H_x B_{xyxx}|E|^2 \cos^2\varphi \pm 2C_{yxz}|E|^2 \sin 2\varphi$ and ${}^{ph}m_z = H_x B_{xzxz}|E|^2 \sin 2\varphi \pm C_{zxx}|E|^2 \cos^2\varphi$, i.e. in this case all components of photoinduced magnetic moments are not equal zero.

Experimental verification of the fact that a magnetic moment is really induced by linearly polarized light in $Ca_3Mn_2Ge_3O_{12}$ came from the magnetic measurements. The photoinduced magnetic moment was measured by means of a SQUID magnetometer. The uniformly magnetized crystal was irradiated along the [001] direction by helium–neon laser light with a wavelength $\lambda = 633$ nm and a light flux density of about 0.1 W / cm^2. The plane of polarization the inducing light was close to the (110) plane of crystal. The magnetization was measured along the axis **z** ∥ [001]. The sample was in a magnetic field $H_z = 6$ kOe at a temperature $T = 5$ K. The change of the magnetization of crystal under illumination, i.e., the photoinduced magnetic moment,

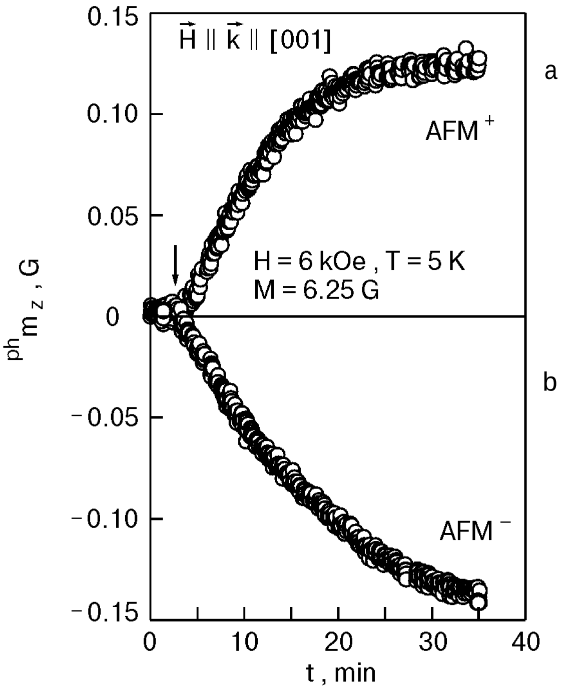

Fig. 8. The time dependences of the photoinduced magnetic moment in the garnet $Ca_3Mn_2Ge_3O_{12}$ exposed to linearly polarized light for two time-inverted antiferromagnetic states AFM^+ and AFM^-. The direction of propagation and the polarization of the inducing light are **k** ∥ [001] and **E** ∥ [110], respectively. The sample temperature $T = 5$ K, and the applied magnetic field is equal to 6 kOe and is oriented along the [001] axis.

was measured. Figure 8 presents the kinetics of the induced magnetic moment measured for the two antiferromagnetic states of the sample, AFM^+ and AFM^-. It follows from (9) that the value of ${}^{ph}m_z$ changes when the antiferromagnetic state is modified from AFM^+ to AFM^-. Indeed, the same exposure of the crystal induced magnetic moments of opposite directions in the AFM^+ and AFM^- states (Fig. 8). The absolute magnitude of the photoinduced magnetic moment upon saturation was 0.12 and 0.13 G in the AFM^+ and AFM^- states, respectively. Thus the absolute magnitudes of the photoinduced magnetic moments in the antiferromagnetic states AFM^+ and AFM^- are almost identical. One can suggest that the polarization-independent magnetic moment ${}^{ph}m_z^{(2)}$ is appreciably less than ${}^{ph}m_z^{(1)}$ or is equal to zero. It should be noted that in the case **k** ∥ [001] the photoinduced magnetic moment was not found in the MM state.

In the case **k** ∥ [100], the magnetic measurements also revealed a photoinduced magnetic moment. In AFM state the value of ${}^{ph}\mathbf{m}$ was lower than in the previous case. As in the above case, the photoinduced magnetic moments in the antiferromagnetic states AFM^+ and AFM^- are almost the same in value and opposite in direction. Thus, in the case considered, the value of the magnetic moment ${}^{ph}m_x^{(2)}$ is also much lower than ${}^{ph}m_x^{(1)}$ or is also equal to zero. The absolute magnitude of the moment was approximately 0.05 G at $T = 5$ K and $H = 6$ kOe. In the case under consideration, the magnetic measurements also revealed a photoinduced magnetic moment in the MM state. Its value was approximately 0.25 G at $T = 5$ K and $H = 35$ kOe.

Using the values of ${}^{ph}\mathbf{m}$ measured with the SQUID magnetometer, one can estimate the change in the MM phase transition field, ΔH_t, caused by the light irradiation in CaMnGeG. The energy of a magnet in a magnetic field can be given in the form of expansion in powers of H [17]:

$$E = E_0 - m_i^0 H_i - \chi_{ij} H_I H_j + \dots , \qquad (11)$$

where E_0 is the energy of the magnet in the absence of magnetic field; m^0 is the spontaneous magnetic moment; χ_{ij} is the magnetic susceptibility. The photo induction of a magnetic moment results in a change of the energy of the magnet by a quantity ${}^{ph}m_i H_i$. Taking this addition into consideration and restricting ourselves to the second-order term in the expansion in H, we can rewrite (11) as follows:

$$E = E_0 - m_i^0 H_i - \chi_{ij} H_i H_j - {}^{ph}m_i (H) H_i . \qquad (12)$$

where ${}^{ph}m_i(H) = {}^{ph}m_i^{(1)} + {}^{ph}\Delta\chi_{ij} H_i$. Then, the energies of the AFM and MM states in both cases considered above can be written as follows:

$$ {}^{A}E = {}^{A}E_0 - {}^{A}\chi_{ii} H_i^2 - {}^{ph}m_i^A(H)H_i ; \qquad (13)$$
$$ {}^{M}E = {}^{M}E_0 - m_i^0 H_i - {}^{M}\chi_{ii} H_i^2 - {}^{ph}m_i^M(H)H_i .$$

In (13) the notations A and M refer to the AFM and MM phases, respectively. Equating the energies of the AFM and MM states in the point of MM phase transition for unexposed and exposed crystal and solving the obtained set of equations, we find the following expression for the photoinduced change in the MM transition field:

$$\Delta H_t = -\frac{{}^{ph}m_i^M(H_t) - {}^{ph}m_i^A(H_t)}{2({}^{M}\chi_{ii} - {}^{A}\chi_{ii}) + m_i^0/H_t} \qquad (14)$$

where H_t is the field of the MM transition in the unexposed crystal.

Using expression (14), we can estimate the value of ΔH_t in both cases considered **k** ∥ **H** ∥ [001] and **k** ∥ **H** ∥ [100]. In the first case no photoinduced magnetic moment was found in the MM state by means of the SQUID magnetometer. By substituting into Eq. (14) ${}^{ph}m_z^M \approx 0$, ${}^{ph}m_z^A \approx \pm 0.12$ G as well as $({}^{M}\chi_{zz} - {}^{A}\chi_{zz}) = -3.6\cdot10^{-4}$ and $m_z^0 \approx 47$ G, we obtained $\Delta H_t \approx \pm 150$ Oe. The estimated value ΔH_t is somewhat (less than two times) higher than the experimental magnitude $\Delta H_t \approx \pm 90$ Oe. However, taking into account the errors in the determination of the parameters substituted in (14), the agreement between the experimental and the calculated values of the ΔH_t is considered satisfactory. It should be mentioned that ${}^{M}\chi_{zz}$, ${}^{A}\chi_{zz}$, and m_z^0 were determined from the field dependence $M(H)$ (m_z^0 was determined by extrapolating the linear field dependence of the magnetization in the MM state to $H = 0$).

In the case **k** ∥ **H** ∥ [100], one has $({}^{M}\chi_{xx} - {}^{A}\chi_{xx}) = -9\cdot10^{-4}$ and $m_x^0 \approx 23$ G at the temperature $T = 11$ K (see Fig. 6), as well as ${}^{ph}m_x^M \approx 0.25$ G, ${}^{ph}m_x^A \approx \pm 0.05$ G. By substituting these parameters into Eq. (14), we obtain two values for the change of the transition field $\Delta H_t \approx -1.2$ kOe and $\Delta H_t \approx -0.8$ kOe. These could correspond either to the two antiferromagnetic states AFM$^+$ and AFM$^-$ or to the exposure of crystal by the light with the two polarizations **E** ∥ [011] and **E** ∥ [0$\bar{1}$1]. The estimated values are in a good agreement with the experimental values $\Delta H_t \approx -1.2$ kOe and $\Delta H_t \approx -0.7$ kOe that were obtained for illumination of the crystal by light with the polarizations **E** ∥ [011] and **E** ∥ [0$\bar{1}$1] at the temperature $T = 11$ K. As can be seen from Fig. 7, the magnitude of ΔH_t decreases at higher and lower temperatures. Apparently, the decrease ΔH_t as the temperature increases from 11 K to T_N is related to the decrease of the photoinduced magnetic moment near the Neel temperature. Some decrease of ΔH_t at temperatures $T < 10.5$ K can be explained by a decreasing absolute value of $(\chi_M - \chi_A)$ while the photoinduced magnetic moment reaches saturation. For instance, one obtains a calculated value $\Delta H_t \approx -1$ kOe at the temperature $T = 9$ K.

Conclusion

It follows from a comparison of the experimental results obtained and the results of a theoretical consideration that the change of the MM transition field induced by linearly polarized light in the garnet $Ca_3Mn_2Ge_3O_{12}$ is due to the induction of a magnetic moment under illumination. The appearance of the photoinduced magnetic moment can be explained by the redistribution of the Mn^{4+} ions between the magnetic sublattices in the crystal. The garnet $Ca_3Mn_2Ge_3O_{12}$ contains Mn^{4+} ions in a low concentration [13]. In the ground state these ions are uniformly distributed between the sublattices. The illumination of the crystal by linearly polarized light leads to a nonuniform distribution of the Mn^{4+} ions between the sublattices as a result of optical transitions with charge transfer [13,16]. As a result of the redistribution, the magnetic sublattices become nonequivalent and a photoinduced magnetic moment appears.

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