hotoinduced linear birefringence in a crystal with ooperative ordering of Jahn-Teller distortions

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Photoinduced linear birefringence (PLB) in the tetragonal garnet $Ca_3Mn_2Ge_3O_{12}$, a crystal with cooperative ordering of Jahn-Teller distortions, is observed and studied. The anisotropy of PLB is described phenomenologically and a microscopic mechanism for it is proposed. It is concluded that PLB can be observed in other crystals exhibiting a cooperative Jahn-Teller effect.

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Photoinduced birefringence of light is a well-know phenomenon for a number of semiconductors and ferroelectrics.[1] The photoinduced change in the optical anisotropy of the materials occurs due to the electro-optical effect, arising due to the photoinduced change in the internal electric field of the crystal. Photoinduced birefringence has also been observed in magnetically ordered crystals, in which the irradiation with light gives rise to changes in the magnetic subsystem.[2] In this paper, we report the observation of photoinduced linear birefringence in the tetragonal garnet $Ca_3Mn_2Ge_3O_{12}$, a crystal with cooperative ordering of the Jahn-Teller distortions. As a result of irradiation with linearly polarized light, the optical phase function of the crystal becomes deformed and turns. These changes depend on the orientation of the polarization plane and the direction of propagation of the light relative to the crystallographic axes. Photoinduced linear birefringence was observed at temperatures greatly exceeding the temperature of magnetic ordering in MnGeG ($T_N \cong 13.5$ K) in the absence of external fields.

We studied PLB in thin MnGeG plates ($d \approx 45$ μm) cut out perpendicular to the tetragonal c axis and the a axis of the crystal, for two directions of propagation of light: $\mathbf{k} \| c$ and $\mathbf{k} \| a$. The orientation of the principal axes of the index ellipsoid was determined and the magnitude of the birefringence was measured as a function of the polarization and intensity of the birefringence-inducing light, the irradiation time, and the temperature of the crystal. Part of the measurements were performed simultaneously with the irradiation, using two independent light beams.

The source of birefringence-inducing light was a DKSSh-150 arc lamp with S3S26 and OS14 filters or a helium-neon laser ($\lambda = 6328$ Å). An attenuated laser beam was used when measuring the birefringence. The temporal dependences of PLB had the form of exponential curves. For a light intensity inducing birefringence $I_u \approx 1$ W/cm^{-2}, the time constant was about 2 min. A decrease in I_u in the range 1–0.2 W/cm^2 increased the time constant from 2 to 10 min, but had no effect on the maximum birefringence. Photoinduced linear birefringence was observed in MnGeG at liquid nitrogen temperatures. The effect was not observed at $T \approx 200$ K. Photoinduced linear birefringence remained when the external action was switched off and it also remained

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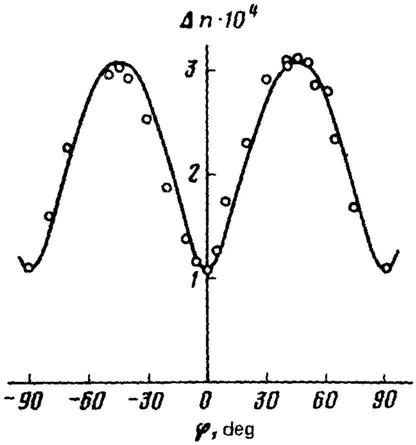

FIG. 1. Photoinduced linear birefringence as a function of the azimuth of the polarization plane of the light inducing the birefringence.

in the dark at liquid nitrogen temperature, without appreciable changes for 20 hours (longer experiments were not performed).

Irradiation of MnGeG with linearly polarized light in the case **k**$\|c$ lowered the optical class of the crystal. The crystal changed from an optically uniaxial crystal to a biaxial crystal. The dependences of birefringence $\Delta n = n_1 - n_2$ (n_1 and n_2 are the principal refractive indices) and of the position of the plane of the optical axes of the crystal on the orientation of the polarization plane of the birefringence-inducing light are presented in Figs. 1 and 2. ϕ is the angle between the a axis of the crystal and the plane of the light inducing PLB, while θ is the angle between the a axis and the plane of the optical axes (to within $\pi/2$). In general, the azimuth of the plane

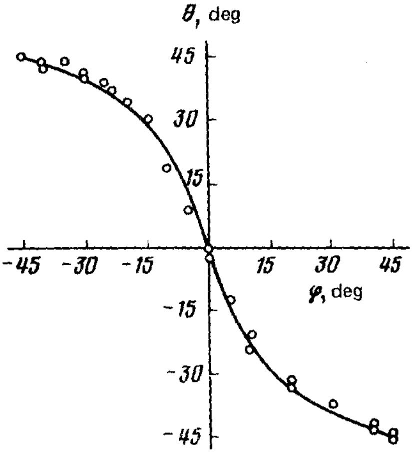

FIG. 2. Azimuth of the plane of the optical axes as a function of the azimuth of the polarization plane of the PLB inducing light.

of the optical axes does not coincide with the azimuth of the polarization plane ($\theta \neq \phi$). They were observed to coincide within the limits of error of the experiment with $\mathbf{E} \| [100],[010]$ or $[110]$. In the case $\mathbf{k} \| a$, irradiation led to rotation of the index ellipsoid. The magnitude of the angle of rotation of the ellipsoid around the a axis reaches 1.5°.

It is logical to assume that the observed PLB effect is attributed to the action of the electric field of the light wave on the state of the crystal. The symmetry of the induced birefringence can be described by a phenomenological expression, which relates the components of the symmetrical part of the dielectric-constant tensor of the crystal and the orientation of the electric field vector $\Delta\, {}^{s}\epsilon_{ij} = C_{ijkl} e_k e_l$, where $\mathbf{e} = \mathbf{E}/E$ is a unit vector. The symmetry of the tensor C_{ijkl} is the same as the symmetry of the tensors describing the Kerr effect and the piezo-optical effect. Since the symmetry of tetragonal MnGeG is described by the point group $4/m$, we find the dependences $\Delta n(\phi)$ and $\theta(\phi)$.

$$\Delta n = \frac{1}{n_0} \left\{ \frac{1}{4} \left[(C_{xxyy} - C_{xxxx}) \cos 2\phi - C_{xxxy} \sin 2\phi \right]^2 + \left[C_{xyxx} \cos 2\phi + \frac{1}{2} C_{xyxy} \sin 2\phi \right]^2 \right\}^{1/2}, \tag{1}$$

$$\theta = \frac{1}{2} \operatorname{arctg} \left\{ \frac{2C_{xyxx} \cos 2\phi + C_{xyxy} \sin 2\phi}{(C_{xxyy} - C_{xxxx}) \cos 2\phi - C_{xxxy} \sin 2\phi} \right\}. \tag{2}$$

Using the experimental values $\Delta n(0) \cong \Delta n_{\min}$ $\Delta n(\pi/4) \cong \Delta n_{\max}$, $\theta(0) \approx 0$, and $\theta(\pi/4) \approx \pi/4$, we determine the phenomenological parameters $C_{xxxy} \approx C_{xyxx} \approx 0$, $|C_{xxyy} - C_{xxxx}| \approx 2n_0 \Delta n_{\min}$, $|C_{xyxy}| \approx 2n_0 \Delta n_{\max}$ and $C_{xyxy}/C_{xxyy} - C_{xxxx} < 0$. Good agreement is observed between the experimental points and the dependences described by Eqs. (1) and (2).

To demonstrate the possibility of using the PLB observed in MnGeG to record an image in this crystal, we recorded the word "CBET." A template containing this word was projected onto the crystal. The word was recorded using light with azimuth $\phi = \pi/4$ in the section of the crystal irradiated beforehand with light with azimuth $\phi = -\pi/4$. When reconstructing the image using light with azimuth $\phi = 0$, a quarter-wave plate was introduced into the optical scheme. The image could be erased using

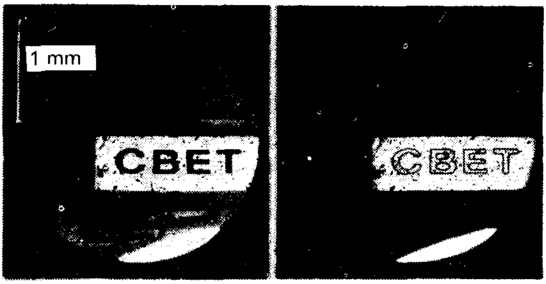

FIG. 3. Image recorded in a $Ca_3Mn_2Ge_3O_{12}$ crystal with the help of PLB.

light with different polarization (re-recording), unpolarized light, or by heating the crystal to $T > 200$ K.

The nature of PLB in MnGeG could be attributed to the cooperative ordering of distortions of local environments of Mn^{3+} ions, caused by the Jahn-Teller (JT) effect, observed in this crystal at $T \lesssim 516$ K. In MnGeG, ordering of JT distortions has a nonferrodistortional nature and it is possible to single out several sublattices with different distortions of the oxygen octahedra. Part of the octahedral positions in the crystal can be occupied by Mn^{4+} ions. The presence of Mn^{4+} ions could be due to the requirement that the crystal be electrically neutral when Ca^{3+} ions replace (during growth) part of the Ge^{4+}ions situated in tetrahedral positions. In the unirradiated crystal, Mn^{4+} ions are distributed with equal probability over the sublattices. The ground state of the Mn^{4+} ion $^4A_{2g}$ does not have orbital degeneracy in the octahedral crystal field and the oxygen octahedra surrounding it are not distorted. The orientation of the orbitals of the Mn^{3+} ion is related to the distortion of the surrounding octahedron, while the probability of electronic transitions depends on the mutual orientation of the orbitals and the direction of polarization of light. Irradiation of MnGeG with linearly polarized light can lead to redistribution of Mn^{4+} ions over the sublattices as a result of electronic transitions with transfer of charge between Mn^{3+} ions from one sublattice and Mn^{4+} ions from another sublattice. This will cause the sublattices to be nonequivalent due to the different number of undistorted octahedra in them, it will lead to anisotropy of the deformation of the crystal, and it will change its optical anisotropy. The times required for the reverse transition to the equally probable distribution of Mn^{4+} ions in the crystal without external action could be long at low temperatures. Aside from the mechanism examined with transfer of charge between Mn^{3+} and Mn^{4+} ions, other mechanisms of capturing a photoelectron are also possible.

The mechanism of PLB observed in MnGeG could be a common mechanism for crystals with a cooperative Jahn-Teller effect. In this case, a wide class of JT crystals is added to the electro-optical and magnetically ordered crystals in which PLB is observed.

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