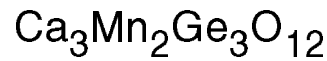


Letters to the Editor

Light-induced optical absorption in the garnet



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Light-induced change in the optical absorption of the garnet $\text{Ca}_3\text{Mn}_2\text{Ge}_3\text{O}_{12}$ has been found. The magnitude of the light-induced optical absorption does not depend on the polarization state of the exciting illumination. The effect persists for a long time at low temperatures and can be attributed to light-induced generation or redistribution of Mn^{4+} ions in the crystal.

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Photoinduced effects occurring in dielectric crystals, in particular, in yttrium iron garnet, may be classified according to whether they depend on the polarization state of the exciting light or not. Different physical mechanisms are responsible for the first and second type of photoinduced effects [1].

Until now, only photoinduced effects depending on the polarization state of the exciting irradiation were reported for the garnet $\text{Ca}_3\text{Mn}_2\text{Ge}_3\text{O}_{12}$. Illumination with linearly polarized light changes the optical anisotropy [2–5] and influences the process of magnetization reversal of the sublattices induced in the antiferromagnetic state by a magnetic field [6,7].

In the present paper we report the results of an experimental observation of a photoinduced effect in calcium manganese germanium garnet (CaMnGeG), which is independent of the polarization state, namely, light-induced optical absorption (LIOA).

The effect of illumination on the optical absorption of CaMnGeG was studied with use of an optical double-beam setup. The beam of the He-Ne laser with the wavelength $\lambda = 633$ nm and a power

of several milliwatts was split into two beams. The sample was irradiated by the first beam, which served at the same time as a transmission probe, while the second beam passed by the sample. Each beam was chopped alternately with a frequency of about 170 Hz by means of a mechanical modulator and hit the photocathode of a photoelectric multiplier. At the beginning, the intensity of the second beam was adjusted to the same intensity as one of the beams that passed through the sample. As the transmission of the sample changed under illumination, the intensity of the first beam also changed and an alternating electric signal appeared at the photoelectric multiplier. This signal is proportional to the change in the transmission of the sample. It was measured by means of a lock-in amplifier.

The sample, a plate of thickness $d \approx 130$ μm , was cut from a single crystal of CaMnGeG, which was polished mechanically and annealed at a temperature of approximately 1 000 °C to relax the internal stresses which appeared during polishing. At the temperature $T \approx 520$ K a twin domain structure arises in CaMnGeG because of a Jahn-Teller phase transition from the cubic to the tetragonal

state [8–10]. Therefore, a thermal treatment [9] is needed to increase the average size of the domains in the sample to approximately 1 mm^2 . A special sample holder with a diaphragm was used to select a single-domain region of the sample. The measured beam passed through a domain in which the tetragonal c axis was perpendicular to the plate; i.e., the measurements of the LIOA were carried out for a sample cut perpendicularly to the tetragonal axis. The sample was immersed into a helium cryostat in a vacuum.

The light-induced variation in the transmission of CaMnGeG was measured as a function of the illumination time. The curves which were measured at several temperatures are shown in Fig. 1, where ΔI is the change in intensity of the transmitted light as a result of the illumination of the crystal, and I is the intensity of the transmitted light measured at the beginning of the illumination of the sample. The measurements show that illumination increases the optical absorption of CaMnGeG .

As the optical absorption of CaMnGeG increases with increasing temperature, too [11], it is necessary to separate the genuine LIOA from the increase in the optical absorption due to heating of the sample under illumination. For this purpose the change in the temperature of the sample under illumination was determined from the known temperature dependence of the linear birefringence [9], using a domain with the c axis in the plane of the sample. The measurements were carried out at an initial temperature of the sample (before illumination) of $T = 200 \text{ K}$, because there is no photoin-

duced linear birefringence above a temperature of 180 K (Refs. 2–5). The time dependences of the sample temperature are shown in the inset in Fig. 1 for heating of the sample during illumination and for cooling after the illumination is switched off. The curves were measured for an intensity of 1.68 and 0.86 W/cm^2 , respectively. As can be seen from a comparison of the dependences shown in Fig. 1 and in the inset in Fig. 1, the time constants for heating and thermal relaxation are much smaller than the time constants for induction and relaxation of LIOA. Measurements of the change in birefringence as a result of illumination at low temperatures allow us to conclude that the time constants for heating and thermal relaxation remain approximately the same, and that heating of the sample by light increases insignificantly. The large difference between the two processes allows us to separate them unambiguously. Furthermore, the measurements of LIOA were carried out at a much lower intensity, 0.16 W/cm^2 . In this case the increase in the temperature of the sample under illumination was less than 1 K and the change in the optical absorption caused by heating of the sample was not essential.

As can be seen from Fig. 1, the saturation value of the LIOA depends on temperature. The temperature dependence of the light-induced change of the transmission of CaMnGeG , measured after reaching saturation, is shown in Fig. 2. The light-induced change in the optical absorption coefficient was calculated using the relation

$$\Delta\alpha = -[\ln(1 - \Delta I/I)]/d$$

and is also plotted in Fig. 2 as a function of temperature. The change in the absorption coefficient under illumination reaches a value of about 30 cm^{-1} at a low temperature but decreases with

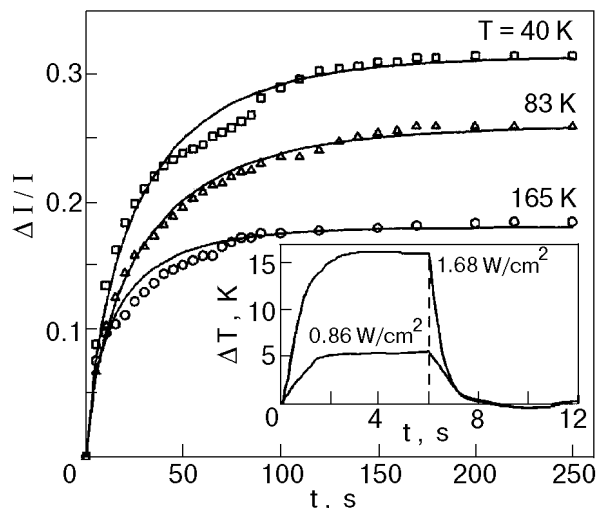


Fig. 1. Change of the optical transmission as a function of the time of illumination measured for the garnet $\text{Ca}_3\text{Mn}_2\text{Ge}_3\text{O}_{12}$ at temperatures $T = 40, 83,$ and 165 K ($I = 0.16 \text{ W/cm}^2$, $\lambda = 633 \text{ nm}$, $d \approx 130 \text{ mm}$). The inset shows the time dependences of the temperature change of the sample at $I = 0.86$ and 1.68 W/cm^2 .

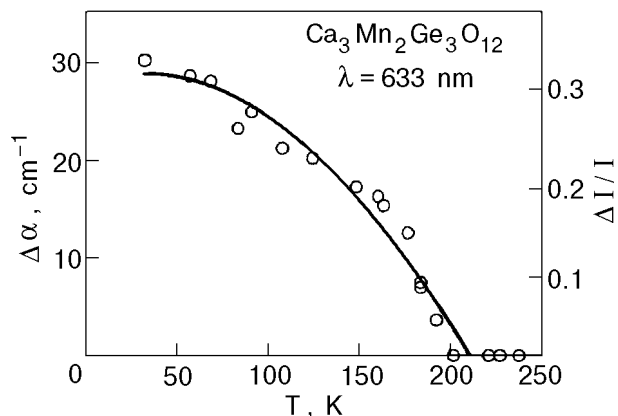


Fig. 2. Temperature dependence of the saturation value of the light-induced optical absorption for the garnet $\text{Ca}_3\text{Mn}_2\text{Ge}_3\text{O}_{12}$.

increasing temperature. LIOA is not observed above $T = 200$ K. It should be noted that the photoinduced linear birefringence in CaMnGeG disappears at $T = 180$ K (Refs. 2–5).

The saturation value of the LIOA is approximately the same for illumination of the crystal with linearly and circularly polarized light as well as unpolarized light. Hence, the magnitude of the LIOA does not depend on the polarization of the inducing light.

The most probable cause for the change in the optical absorption of CaMnGeG under illumination is the generation of Mn^{4+} ions due to optically induced transfer of electrons from Mn^{3+} ions to traps (photodoping). In accordance with Refs. 12 and 13, Mn^{4+} ions can be located in the octahedral positions in CaMnGeG. The local octahedral oxygen surrounding of the Mn^{4+} ion in $Ca_3Mn_2Ge_3O_{12}$ differs from that of the Mn^{3+} ion, because the Mn^{4+} ion has an orbitally nondegenerate ground state, and because it is not a Jahn-Teller ion. Local distortions and stresses of crystal lattices should accompany an appearing of Mn^{4+} ions in CaMnGeG. The Mn^{4+} ions placed within the changed oxygen octahedra in the lattice of CaMnGeG can be the light-induced absorption centers, which are responsible for the increase of absorption. It is known that Mn^{4+} ions in the octahedral position of the garnet lattice (e.g., in the garnet $Ca_3Ga_2Ge_3O_{12} : Mn$) have absorption bands in the visible range of the optical spectra [14]. The optical absorption can also increase due to charge transfer transitions between Mn^{3+} and Mn^{4+} ions.

A further conceivable mechanism is the change in the optical absorption of CaMnGeG by light-induced redistribution among already existing Mn^{4+} in the crystal. A similar model was proposed for the explanation of LIOA in the garnet $Y_3Fe_5O_{12}$ [15,16]. The photoinduced change of the optical absorption in this garnet was interpreted in terms of different properties of Fe^{2+} and Fe^{4+} ions at the sites that are near and far from an impurity ion or a vacancy. The compensation for the electrical charge of the Mn^{4+} ions is accomplished by vacancies of Ge^{4+} ions in the crystal [12,13]. If it is

assumed that the optical properties of Mn^{4+} ions differ at the sites that are near and far from Ge vacancies, then the optical absorption of CaMnGeG can change as a result of the light-induced redistribution of these ions between the sites near and far from the vacancies.

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