

Short Communication

Photoinduced Deformation in the Tl_4SnSe_3 Single Crystals

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mStudies of the deformation for the ternary chalcogenide crystals Tl_4SnSe_3 was performed under illumination of the two type of pulsed lasers: nanosecond Nd:YAG laser generating at 1064 nm and nanosecond CO_2 laser generating at 10.6 μm and its fourth harmonic at 2.65 μm . The dilatometry studies have shown nonlinear dependence of laser stimulated deformation versus the pump power density. The dependence has an opposite feature which may indicate on an excitation of the different systems by these two lasers: electronic and phonon ones. The possible mechanisms are based on the excitations of the electron and phonon subsystem and formation of the photopolarized states which change the deformation.

Keywords: chalcogenide crystals; photostimulated deformation; Tl_4SnSe_3 Single Crystals

1. INTRODUCTION

Recently one can observe an enhanced interest to the photoinduced effects in the complex chalcogenide crystals [1]. This is caused by a fact that these crystals possess large polarizabilities and contribution of the electron-phonon anharmonicities [2]. Additionally there are a lot of trapping levels due to intrinsic defect states [3] which may favor the laser stimulated effects. Particular interest

present the ternary and quaternary compounds possessing the chalcogenide anions [4]. The latter allow to perform the measurements in the wide spectral IR range (up to 20 mm) which open a new opportunity for the use of the thallium containing compounds.

Due to high electron-phonon anharmonicity in the such kind of crystals [5-8] one can expect that illumination by the external laser may favor not only an increase of the nonlinear optical coefficients however also of the mechanical coefficients including the elastic constants.

2. EXPERIMENTAL METHODS

The Tl_4SnSe_3 is formed in the $\text{Tl}_2\text{Se-SnSe}$ quasibinary system and congruently melted at 706 K [9]. Standardized crystallographic data of them presented in Table 1. A synthesis of Tl_4SnSe_3 was carried out from binary thallium (I), and tin (II) chalcogenides in ratio 2:1. For obtaining the binaries high purity elemental solids (Tl—99.99 wt.%, Sn—99.998 wt.%, Se—99.99 wt.%) were used.

All synthesis passed in vacuum evacuated to 0.133 Pa quartz ampoules. Crystal was grown by the Bridgman method. The temperatures of the zones were maintained with ± 0.5 K accuracy. The solidification rate was 2–2.4 mm/h, and the temperature gradient at the solid–liquid interface was 2–4 K/mm. The crystal was annealed in the growth zone at 550 K for 4 hours and cooled to room temperature at a rate within 30 K/h.

Binary and ternary compounds were characterized by differential thermal analysis (DTA) (Chromel–Alumel thermocouple, two-coordinate recorder) and X-ray powder diffraction (XRD) (DRON 4 powder diffractometer, Cu Ra radiation). The X-ray diffraction (XRD) powder patterns of compounds (see Fig. 1) were in good agreement with those calculated based on reported structure data (see Table 1).

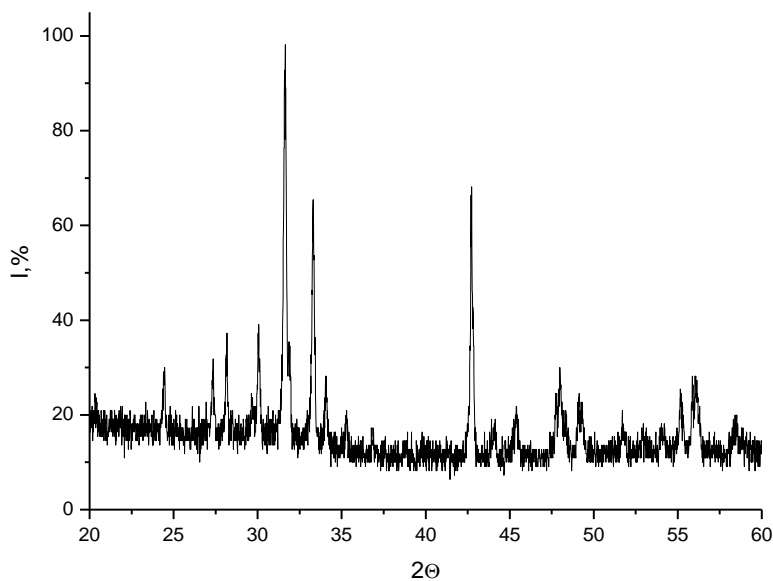


Figure 1. XRD pattern of Tl_4SnSe_3

Obtained single crystal was gray with metallic-like color. It didn't contain any admixtures and cavities. The crystal is stable on air and don't react with water and ethanol, slowly soluble in concentrated nitric acid.

The measurements of the photoinduced dilatometer were done using method described in the details in the ref. 10. As photoinducing lasers were used microsecond CO₂ lasers with frequency repetition about 20 Hz as well as 15 ns Nd:YAG lasers with frequency repetition about 100 Hz.

3. CRYSTALLOCHEMISTRY ASPECT.

The coordination environment of atoms of metallic component and distance to the selenium atoms in the structure of compound Tl₄SnSe₃ is shown in Fig. 2. Atoms Sn are situated within the octahedra with selenium atoms are slightly displaced along the main axis and possess asymmetric location within the immediate environment of the atoms of anions which may be responsible for nonlinear optical effects. Octahedron of polyhedra around the atoms Sn are merged into the structure of nodes and create 3d channels (Fig.3a - Fig.3b parallel and perpendicular to the main axis) in which the atoms are arranged Tl. As shown in Fig.2 and Fig.3, Tl atoms possess more asymmetric environment than atoms Sn, which also indicates their huge potential for the effect described by third rank tensors.

Table 1. Standardized crystallographic data for Tl₄SnSe₃ (SG *P4/ncc* (130); $a = 8.522$, $b = 8.522$, $c = 12.722$ Å)

Site	Wyck.	x	y	z
Tl*	16g	0.125	0.5777	0.0974
Sn*	4c	1/4	1/4	0.2931
Se1	4c	1/4	1/4	0.0372
Se2	8f	0.4129	0.5871	1/4

$$\text{Tl}^* = 0.935\text{Tl} + 0.065\text{Sn}$$

$$\text{Sn}^* = 0.74\text{Sn} + 0.26\text{Tl}$$

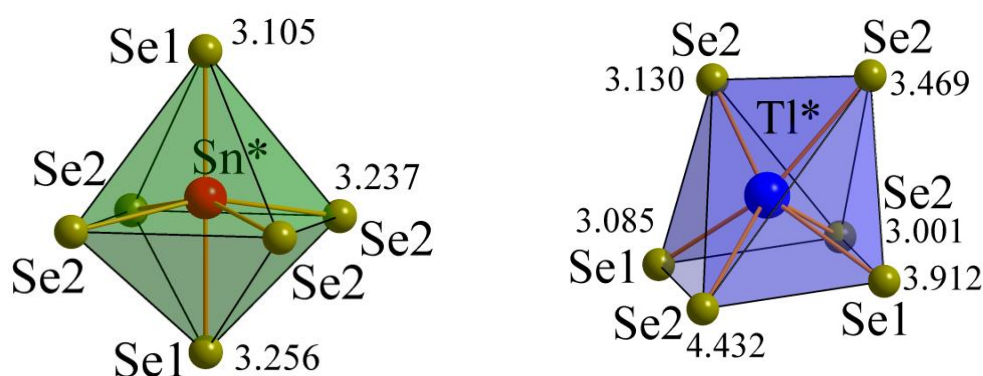


Figure 2. Coordination of the principal metallic atoms and distances to the Se atoms for the Tl₄SnSe₃

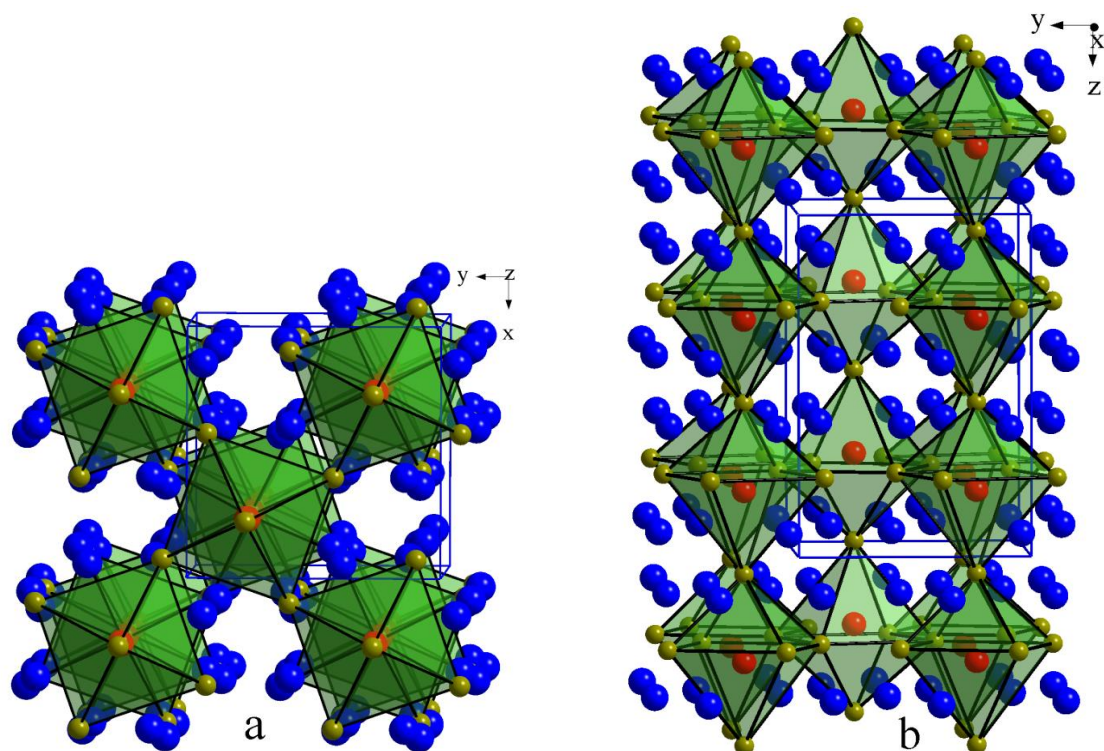


Figure 3. The package of atoms Tl in the voids between the selenium octahedral surrounding Sn.

4. RESULT AND DISCUSSION

The photoinduced dilatometry measurements were performed using the dilatometer which has performed the direct deformation measurements during the illumination. The red line corresponds to illumination by the microsecond CO₂ laser at wavelength 10.6 μm and the green line corresponds to the illumination by the 2.65 μm laser. The laser geometry was chosen to excite the maximally the trapping levels both electronic [11] as well as phonon ones [12].

Following the Fig. 4 one can clearly see that there are two different pump power dependences of the studied crystals. The first one corresponds to the occurrence of the deformation maximum for the 10.6 μm laser illumination and the second one to the occurrence of the corresponding minimum for 2.65 μm at 0.25 mJ/cm². Such behavior may indicate excitation of the different subsystem in the observed dependences. The Nd:YAG laser excite mainly the electronic subsystem and the CO₂ laser excite mainly the phonon subsystem [13]. The principal mechanisms are optical Kerr effects, mechanoelastic and piezoelectricity together with elasto-opticity [14]. The sign of the corresponding electron-phonon anharmonicities [10] will be here different which may cause the additional effects. The occurrence of the maximum and minimum at the almost the same energy densities may confirm the principal role of the electron-phonon anharmonicities which may play here a decisive role due to interaction with the defect induced trapping levels [16]. It is crucial the effect was independent on the light polarization and the defocusing in accordance with the ref. [17]. The relative changes of the surface temperature did not exceed 3-4 K. Following the general photoinduced experiments such

temperature enhancement could not change substantially the photostimulated optical constants [18]. The photoinduced deformation is occurred due to the electron-phonon part of the corresponding electromechanical effects described by the third-order tensor.

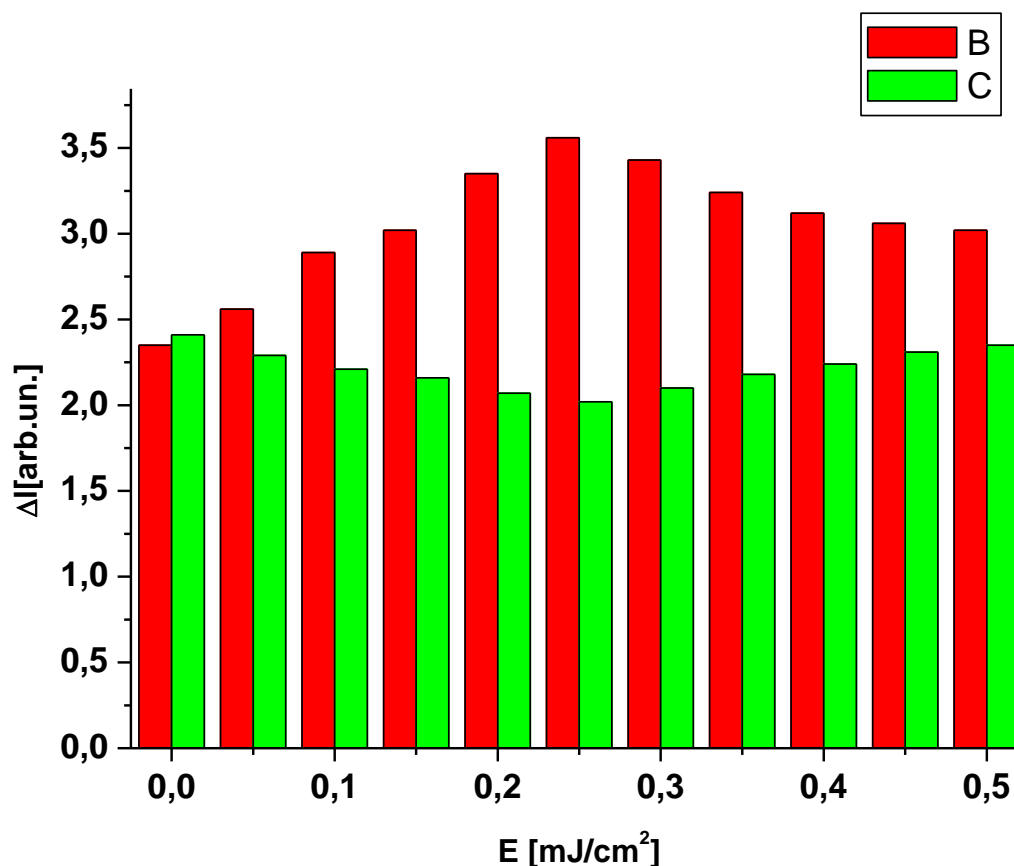


Figure 4. Photoinduced deformation perpendicular to the light propagation for the crystals. B-Nd:YAG excitation; C-CO₂ laser excitation.

It is crucial that the process is completely reversible and disappears after the switching off of the photoinduced laser beams. Moreover, the form of the beam sequence did not play any significant role in the observed effects.

5. CONCLUSIONS

During the dilatometer studies of the Tl_4SnSe_3 single crystals it was found nonlinear dependence of deformation versus the pump power density. The dependence has an opposite feature which may indicate on an excitation of the different systems by these two lasers. The principal mechanisms are related to the excitations of the different subsystems by the lasers. The photoinduced deformation is occurred due to the electron-phonon part of the corresponding electromechanical effects described by the third-order tensor. It is crucial that the process is completely reversible and disappears after the switching off of the photoinduced laser beams.

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