

Two-photon absorption of $Tl_{1-x}In_{1-x}Sn_xSe_2$ nanocrystallites

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Abstract. Novel materials for the infrared two-photon absorption — $Tl_{1-x}In_{1-x}Sn_xSe_2$ single crystals ($x=0.1,0.2$) were grown. Two-photon absorption (TPA) was studied at CO_2 laser wave-length $9.4\mu m$ with pulse duration $1\mu s$. The studies were performed at different temperatures and for the nanocrystallite sizes varying within the $7-200$ nm. The studies have shown that the TPA may be enhanced during the decrease of the nanocrystallite sizes below $50-60$ nm. There exists also some critical x value at which the TPA value begin substantially to increase. The studied nanocrystallites are relatively stable to the infrared laser treatment and are not hygroscopic which allow to use them in different IR optoelectronic devices.

1 Introduction

The search of materials for infrared (IR) light detection in the mid infrared spectral range, (i.e. about $10\mu m$) is very actual task [1–5]. Almost all the studies are related with the use of quantum dot IR photodetectors QDIPs [6]. All these researches require a necessity to use sophisticated and expansive quantum dot-in-a-well technology. However, there exists still a large amount of materials in which due to the processes of additional contribution of the IR induced phonons, including anharmonic ones [7], there occur large reserves for enhanced nonlinear optical susceptibilities. The latter may give substantial contribution to the nonlinear optical effects of the third order [8, 9] which present an active area of research in recent years.

In this paper we present a study study the two-photon absorption (TPA) in the IR wavelength spectral range for the $Tl_{1-x}In_{1-x}Sn_xSe_2$ nanocrystallites ($x=0.1, 0.2$) using as a fundamental CO_2 laser with wavelength $9.4\mu m$ and pulse duration $1\mu s$. The studies were performed at different temperatures and for the nanocrystallite sizes varying within the $7-200$ nm

2 Experimental

The titled single crystals were fabricated by Bridgman–Stockbarger method using solid solution range varying within the $0-20$ mol.% $SnSe_2$. The technological process was proceed in a shaft-type similarly to the method described in Ref.[10]. Finally the such-grown grown single crystals (with diameter 9 mm

and length about 30 mm) were black and possessed a good layered structure which allow to perform their cleaving.

The specimens for linear spectra and two-photon absorption of the $Tl_{1-x}In_{1-x}Sn_xSe_2$ crystals were cleaved from the relatively perfect middle part of the single-crystalline ingot and were shaped as thin plates of thickness varying within the $0.08-0.1$ mm. For the nanocrystallites preparation we have used a method of crystal milling with additional acoustical milling and size separation as described in Ref.[11]

3 Results and discussion

The NC sizes and their size distribution was controlled by JEOL TEM microcopy. Typical NC size distributions is presented in Fig. 1.

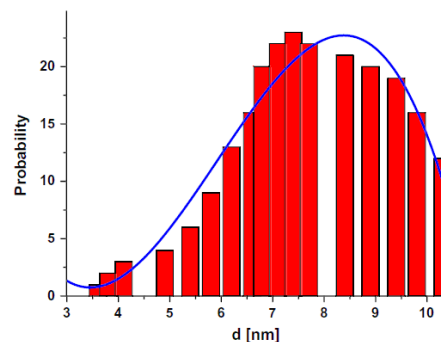


Fig. 1. Typical size distribution of the nanocrystals following the TEM analysis for the series with the lowest sizes

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One can see a huge size dispersion which in addition shows substantial asymmetry. However, the such distribution allows to perform analysis of their optical and nonlinear optical properties.

In Fig. 2 are presented the dependences of the TPA versus the NC sizes at two temperatures: at room (ambient) temperature and the LNT. One can see a drastic increase of the TPA at average sizes below 100 nm at LNT and at below 50 nm at RT. Such differences confirms substantial role of the phonon subsystem for the such kind of the NC. At the same time the enhancement of the TPA was up to four times, which allow to predict that the nano-confined effects favoring high local electric field are dominated here.

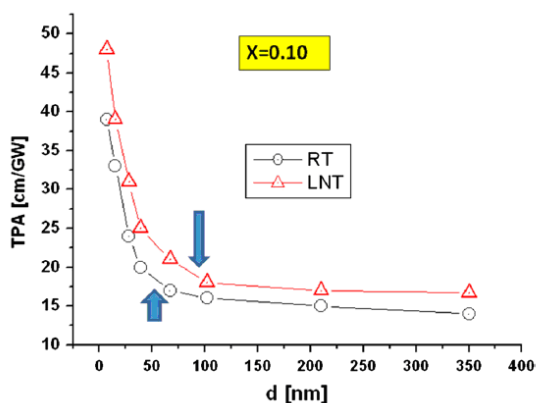


Fig. 2. Size dependence of the TPA for the samples with content $x=0.10$ at RT and LNT temperatures

To explore an influence the nanocrystal composition on the TPA behavior in Fig. 3 are presented the features for $x=0.20$. Here the general TPA dependences versus the sizes is almost the same. However, in this case the role of the temperature is not so crucial. So one can predict that in this case the local disorder defining the local trapping levels is prevailing. It is necessary to add that the illumination of the studied compounds by the corresponding laser pulses did not change the TPA more than 0.2%.

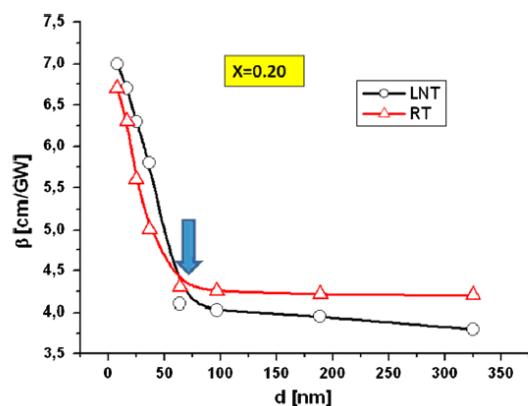


Fig. 3. Size dependence of the TPA for the samples with content $x=0.10$ at RT and LNT temperatures.

The studies of the two-photon absorption for the $Tl_{1-x}In_{1-x}Sn_xSe_2$ single crystals ($x=0.1, 0.2$) and corresponding nanocrystals at $9.4\mu m$ has shown high value of the TPA coefficients for the diagonal tensor components which achieve the value up to 50 cm/GW. The maximal TPA value achieved for diagonal tensor components occurring at the $x=0.1$. It is principal that the TPA decreases with decreasing temperature. Generally it shows sub-stancially non-monotonous dependence versus the Tl content. This fact is a result of two different contributions: the long-range ordering and the mentioned trapping levels. The essential crucial temperature sensitivity behavior may be caused by additional phonon contribution to these localized trapping levels. It is crucial that the effect was stable for the laser beams varying within the wide range of beam spot diameters: from 5 mm up to 3 cm.

The achieved parameters of nonlinear optical effects derived for the wavelengths corresponding to the CO_2 laser illumination allows to predict wide applications of $Tl_{1-x}In_{1-x}Sn_xSe_2$ single crystals as materials for nonlinear optical transformation of the infrared laser beams which is crucial in the infrared LIDAR (Light Detection and Ranging) systems. In particular, the crystals may be effectively used in the photonic gratings [12]. It is necessary to add that during multi-time IR induced treatment the observed photoinduced changes were completely reversible which confirm their high reproduction potential [13–16].

Conclusions

During exploration of the TPA versus the sizes was discovered a began of drastic increase the TPA at nanocrystal average sizes about 100 nm at LNT and at about 50 nm at RT. Such differences confirms substantial role of the phonon subsystem for the such kind of the NC. At the same time the enhancement of the TPA was up to four times, which allow to predict that the nano-confined effects favoring high local electric field are dominated here.

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