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Photoinduced effects in Bi_2Te_3-$CaBr_2$-$PbCl_2$ optical fibres

1. Introduction

Bi_2Te_3-$CaBr_2$-$PbCl_2$ optical fibres promise to be perspective materials for IR optoelectronics and quantum electronics due to possibility of their use in different branches of IR optoelectronics, holography and IR quantum electronics materials engineering, particularly as a material for IR fibres /1-3/. The range of light transmission T of the obtained glasses falls within the range 0.65-40 μm at the level 45 - 70%. They possess high photomechanical stability (up to 5.12 GW/cm^2) /1/.

These fibres, due to peculiarities of their chemical content, could be interesting also for IR photoinduced effects, particularly for the nonlinear optics, particularly to the photoinduced optical second harmonic generation (SHG) and two-photon absorption (TPA).

Some physical and chemical properties of the mentioned compounds were described by several authors, but to our knowledge, there is no a work devoted to influence of external IR laser light on their basic optical properties, particularly on nonlinear optical susceptibilities. The lasers that could be actual for the such kinds of illumination are the CO_2 lasers with the wavelengths about 10.6 μm. It is understandable that the doubled frequency signal will have frequency of 5.3 μm.

2. Investigation method

The absolute values of the SHG were more than 20% less comparing to χ_{222} tensor for Ag_3AsSe_3 single crystals. The SHG signal strongly increases within the 125 - 149 K temperature range and achieves its maximum value at 149.6 K. Femtosecond probe-pump measurements indicate on a fast response of the measured fibres (up to the 1.24 ps) at spectral wavelength about 1.06 μm. In the mid-IR spectral range the nature of

photoinduced changes of the optical properties of glasses remains unknown. As a consequence in the present work we perform the photoinduced nonlinear optics measurements in the mentioned fibres in order to test a possibility to use this material for IR nonlinear optics electronics. To clarify mechanisms and origin of the photoinduced effects and contribution of particular structural clusters as well of phonon and electronic subsystems we perform *ab initio* electron structure calculations with simultaneous molecular dynamics geometry optimizations. Especially we clarify role played by IR harmonic and anharmonic electron-phonon interactions in the observed phenomena, particularly during the high-resolved femtosecond temporary effects.

3. Results and discussion

To clarify main physical mechanisms of the observed phenomena we carry out both experimental and theoretical quantum chemical and molecular dynamics simula-

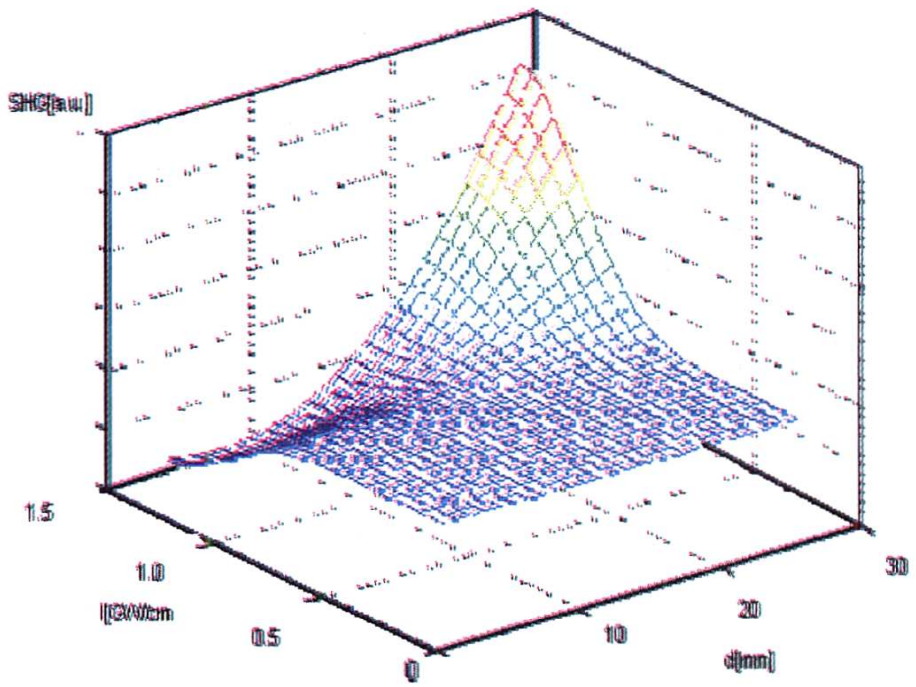

Fig. 1. Dependence of the photoinduced SHG versus the photoinducing beam power I and the fiber diameter d at T=149 K

tions and clarify role of particular structural clusters in observed phenomena. We have carried out measurements of two-photon absorption (TPA) and second harmonic generation (SHG) in the IR-region from 5.8 to 13 μm. CO_2-laser (λ=10.6 μm) and parametrically generated wavelengths (5.7 – 12.4 μm) were used as a source of pumping

light. We have found that with an increase of photoinducing power, the SHG for probe CO_2 laser increases and we have revealed a substantial sublinear dependence of the output SHG maximum on pump-probe time delay about 1.7 ps. Spectral positions of the TPA maxima are strongly depended on the pump power. Contrary to the SHG behavior, for the TPA we observe at least two time delayed maxima: at 1.3...1.8 ps and 4.5 ps. We explain these dependencies within a framework of the quantum chemical approach with taken into account IR to photoinduced anharmonic electron-phonon interaction. We have revealed that Bi-Te tetrahedra play a key role in the observed photoinduced non-linear optics effects. The obtained results show that the mentioned fibres can be used as promising materials for picosecond IR non-linear optics processes. We have also revealed essential dependence of the observed photoinduced SHG on the diameter of the optical fibres (see Fig. 1) and we have found that the optimal diameter for the maximal SHG is about 26 μm.

Simultaneously the investigated glasses are promising materials for IR femtosecond Q-switched quantum electronics.

We perform complex theoretical investigations of IR influence on nonlinear optical susceptibilities. Particularly we investigate the optical SHG, described by the polar third rank tensors, and TPA, described by imaginary part of fourth-rank nonlinear optical tensors, in order to clarify the physical mechanisms responsible for the photoinduced changes in the middle infrared spectral range.

4. Conclusions

We have demonstrated a possibility of using the Bi_2Te_3-$CaBr_2$-$PbCl_2$ glasses as promising materials for the IR optical fibres. We have revealed that the mentioned glasses possess good photoinduced SHG and TPA properties in the mid IR spectral region where generate CO_2 lasers. Moreover we have found that choosing an appropriate fibre diameter and IR photoinducing beam there exist a possibility to enhance the output nonlinear optical susceptibility. These fibres could serve as devices for simultaneous transformation and transmission of the IR signals.

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Summary

We have carried out at the first time measurements of photoinduced optical second harmonic generation in the infrared spectral region (λ=10.6 μm). The highest possible photoinduced output SHG is achieved for the optical fibre diameter about 27 μm and at the photoinducing power about 1.45 GW/cm^2