PHOTOINDUCED ELECTRONIC STRUCTURE AND NONLINEAR SUSCEPTIBILITIES IN Y-Ba-Cu-O CERAMICS

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INTRODUCTION

The appearance of an interesting phenomenon consisting of the photodoping of high temperature superconductivity materials has been discussed in several previous publications [1-6]. Measurements were carried out mainly on $YBa_2Cu_3O_{6+x}$ single crystalline films and have shown that during laser photoexcitation the insulating underdopped samples of $YBa_2Cu_3O_{7-\delta}$ become hightly conductive [7]. Further time-dependent measurements [8,9], have shown an exponential dependence of photocurrent on irradiated light intensity. For light intensities above $6x10^{17}$ photon cm^{-2} the superlinear dependence of photoinduced photocurrent was observed [10]. These effects may be considered as superconductors, driven out of the equilibrium state by the external field. As a consequence, they can undergo transition to either the normal or the superconducting state depending on the initial state of the materials.

The detected peculiarities were explained by the additional carrier transport mechanism [6,7], photoinduced oxygen ordering [11,12], disordering [13], charge transfer with subsequent electron trapping [14] and electron-phonon anharmonicity [15]. The values of resistivity decrease at 50 K were more than 14 orders of magnitude less and photoinduced conductivity was time delayed due to absorption. The concepts of phase separation and metallic droplet formation were used to explain the observed phenomena [10,15]. The similar behaviors of the photoinduced parameters were noticed in Refs. [16,17], where the analogous photostimulated phenomena were observed under the influence of low-power He-Ne, He-Cd, He-Se and nitrogen laser illumination.

In Refs. [10,15,17,18] it was shown that the nonlinear optical methods, particularly second harmonic generation (SHG), are sensitive to the appearance of superconductivity. High sensivity of the SHG to the structural electron-phonon anharmonicity was demonstrated [10]. Particularly, in Ref. [19] it was shown that on approaching near to the phase transition, the SHG 'feels' an appearance of the superconducting state. The essential role of the electron-phonon anharmonicity contributing to the nonsymmetric SHG was demonstrated.

The occurrence of the SHG signal takes place at temperatures higher than those at which the resistivity or magnetic susceptibility begins to arise [20].
In the present work we investigate the influence of photoinducing laser illumination on the second-order nonlinear optical susceptibility of Y-Ba-Cu-O ceramics. The appearance of the SGH signal may indicate unambiguously the presence of the superconducting phase [10,18,19].
These investigations are particularly relevant for the high-resolved photokinetic time regime [15,19]. All the measurements are performed at different points of the investigated ceramic samples to avoid the possibility of an nonuniform distribution of the Y-Ba-Cu-O. There is also the possibility to design hight-time-resolved laser switches. Besides an experimental approach, we also use quantum-chemical calculations taking into account electron-phonon interactions in order to explain the observed effects.
The main goal of our investigation is to study the influence of the external UV illumination on the nonlinear optics parameters of the superconducting Y-Ba-Cu-O ceramics. Such investigations may give valuable information about the origin of high-temperature superconductivity. Besides, these methods may be a promising tool for contact-less detection of superconductivity.

EXPERIMENTAL RESULTS

Experimental methods

As a source of photoinduced changes, we used nitrogen laser (λ=0.337 μm, τ=1-50 ps, W= 45 MW) with pump power varying within 0.1-1.4 GW cm^{-2} where upper power restriction was introduced in order to avoid sample heating. The energy of the pump beam at the specimen surface was checked using a commercial fast-response joulemeter (Genetic Inc., model ED-200). Generally, the equipment is similar to that described in Ref. [15]. The SHG signal was detected using a YAG-Nd pulse laser (W=30 MW, t= 30 ps; frequency of laser pulses rate about 12 Hz).
It was revealed that minimal intensity of the output SHG signal was achieved for parallel polarization of photoinducing and probing light beams. All the measurements were done in the regime of light reflection [15].
The separation between the output SHG (λ = 0.53 μm), photoinducing nitrogen (λ = 0.337 μm) and pumping YAG-Nd (λ = 1.06 μm) was achieved using grating monochromator SMR-3-M (spectral resolution about 0.2 nm mm^{-1}).
The SHG intensity was detected using a RCA 4V photomultiplier. All the measurements were performed in single-pulse regime, with a pulse frequency repetition of 7 Hz. As an intensity standard, a pyrargit single-crystalline cell cut in the plane of the optical axis was used. Time-

dependent SHG signals were measured using the high-time-resolved spectroanalyser A-165.
The investigated specimens had a parallelepiped form with a-surface planes lying within the 5x5 mm surface and roughness better than 0.1 μm.

Photoinduced SHG results
At the beginning, we measured dependences of the photoinduced SHG intensities versus temperature at different photoinducing UV nitrogen laser powers (see Fig. 1). Our measurements have shown that maximal values of the PISHG susceptibility (PhotoInduced SHG) were achieved at pump-probe delaying time about 65 ps. A drastic increase of the photoinduced SHG was achieved at temperatures below 120 K. At higher temperatures, the presence of the non-centrosymmetric surface states in the measured ceramics results. In the appearance of the SHG that indicates that one several kinks in the temperature dependences of the SHG. These kinks may correspond to contributions of different grains [16].
We found that the output SHG signals were increased within the 63-120 K temperature range, achieving saturation below 62 K. The maximal output SHG was equal to about 0.25 pm V^{-1} and was achieved with the photoinducing power of about 1 GW cm^{-2}. For the photoinducing powers higher than 1 GW cm^{-2} the SHG signal decreases (from 0.25 to 0.224 pm V^{-1}). The observed behavior unambiguously shows that the observed photoinduced SHG is caused by the superconducting phase transitions. Because the ceramics possess centrosymmetric symmetry in both phases, one can expect a dominant role of non-centrosymmetric anharmonic electron-phonon interactions in the observed SHG dependences.
Theoretical quantum chemical calculations presented below confirm a substantial influence of the phonon modes on the behavior of the output photoinduced SHG due to achievement of the higher nonuniformity created in density distribution in electronic charge. Competition between photoinducing non-centrosymmetry in electronic charge density distribution and photoinduced power is a main cause of the observed second-order nonlinear optical phenomena. The observed phenomena are caused by photogeneration of carriers from occupied to unoccupied conducting states [15]. Due to changes of the excited dipole matrix elements, there appears local nonlinear optical susceptibility [17]. For calculating the absolute value of the second-order nonlinear optical susceptibility tensor χ_{222} we have measured both transparency and reflection from ceramic specimens.
The duration time of photoinduced SHG signal did not exceed 1.5 ps, *and the further investigation of the photoinduced SHG revealed, that the maximal SHG signal was achieved at pump-probe delaying time of about 68 ps (see Fig. 2).* The existence of the SHG's maximum at $\tau \approx$

65 ps indicates a dominant contribution of electron-phonon interaction to the observed SHG [22]. This temporary asymmetric behavior is very similar to the behaviors that play a part in the photoinduced anharmonic electron-phonon states [21-23]. Besides, quasi-matched phase synchronism conditions play an important role here [24]. We cannot also dismiss completely a possibility that quasi-crystalline interface states may appear [18].

Another very important aspect of the phenomenon is the UV-power dependence of SHG (see Fig. 3). One can see that the maximal increase of the SHG is observed for 0.58-0.71 GW cm^{-2} photoinducing power. For the photoinducing power higher than 1 GW cm^{-2} the output SHG intensity begins to decrease. Such a decrease can be explained by saturation of electron-phonon anharmonic modes effectively contributing to the nonlinear optical susceptibilities. At temperatures higher than 95 K, the SHG signal is very low.

For the ceramics $YBa_2Cu_3O_{6+x}$ with x=0.61, and with increasing temperature, there is a substantial increase of the SHG which indicates, that a superconducting phase appears at the temperature 8-12 K higher, then the temperature, at which superconducting transition occurs. It is necessary to add that for the oxygen content x below 0.61 we have not observed a SHG signal.

In our previous works [20,21], we indicated a possibility that enhanced SHG may occur with heating to the critical points. Corresponding behaviors seem to be very similar to low-temperature structural rearrangement that leads to partial long-range ordering. In the case of ceramics, the long-range ordering disappears and the entire process should be considered within a framework of separate structural clusters.

CONCLUSIONS

We have found experimentally that in Y-Ba-Cu-O ceramics below 120 K with an increasing nitrogen-laser photoinducing power, the SHG maximum output signal increases and achieves its maximum at nitrogen laser power about 1 GW cm^{-2}. We have found that maximal SHG signal is observed for pump-probe delaying time about 65 ps. This indicates a key role of electron-phonon interaction in the observed phenomenon. Using band energy structure calculations, taking into account electron-phonon anharmonicity, we have demonstrated that the dominant role in the observed effect is played by 3dCu-2pO band states effectively interacting with the phonon modes of oxygen near the vacancy. The simulation of spectral dependences of the nonlinear optical susceptibilities unambiguously show the necessity of the appearance of the SHG maximum that is closely connected with the electron-phonon anharmonicity. The observed phenomenon may be used for contact-less detection of the superconducting phase transition.

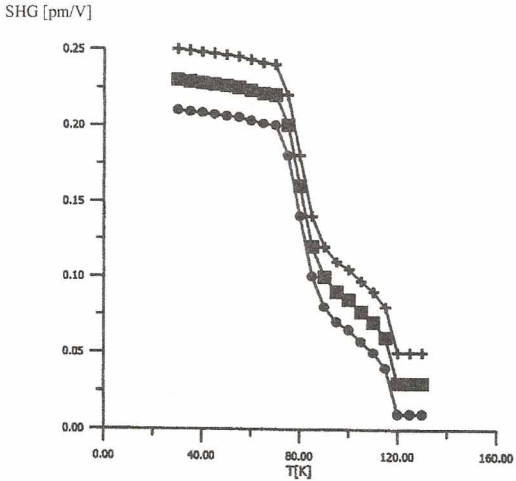

Fig. 1. Dependence of photoinduced SHG as a function of temperature for different nitrogen-laser photon powers: (■) 1.3 GW cm^{-2}, (●) 0.5 GW cm^{-2}, (+) 1 GW cm^{-2}

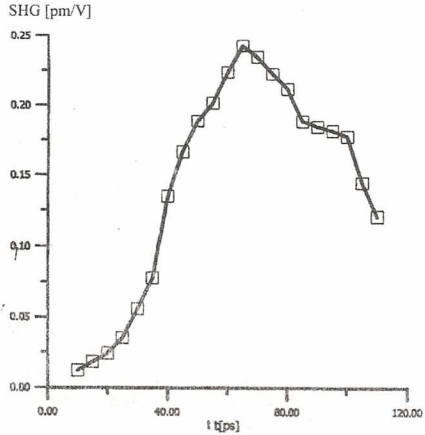

Fig. 2. Dependence of output photoinduced SHG maximum as a function of pump-probe delay at T=77 K. Photoinducing power about 1 GW cm^{-2}

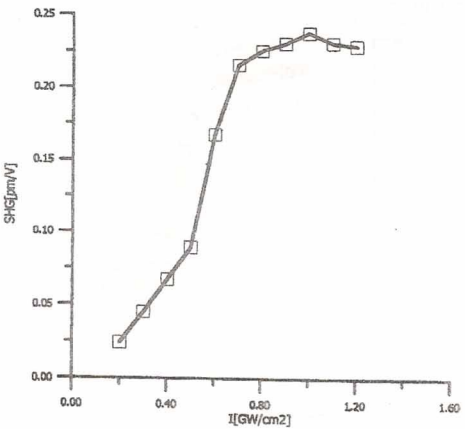

Fig. 3. Dependence of photoinduced SHG at the time delay between the pump and probe beams 68 ps and T = 77 K

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