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Optically induced effects in ZnO nanocrystallites on different substrates

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Using a bicolor coherent treatment with a Nd-YAG laser with wavelength 1.32 μm and its second harmonic generation, we have investigated ZnO nanocrystaqllites deposited on different substrates: MgO, ZnO, CdO and BaO. It was shown that the maximum value of the second order susceptibilities are achieved for the MgO substrates. The performed molecular dynamics and quantum chemical simulations indicate on a principal role of the substrate in the observed dependences of the second-order optical susceptibilities.

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Keywords: ZnO, Optical effects, Second order susceptibility

1. Introduction

Recently one can observe an enhanced interest to ZnO films and nanocrystallites during search and design of new materials for quantum electronics [1-4]. However in all the investigations the second order optical effects, particularly optical second harmonic generation were related with the degree of orientation of the ZnO films. In the present work we will investigate influence of different substrates on the output second-order optical susceptibilities. For the investigations we have chosen ceramics of MgO, ZnO, CdO and BaO.

2. Experimental methods

The samples were deposited using laser molecular epitaxy. As a source laser excimer XeF laser generating at 218 nm was used. The pulses of 8-10 GW/cm^2 were used for deposition of ZnO films on the heated substrates of Si<111>, LiNbO3 and MgO. The deposition rate about 40 nm/s were used. The homogeneity of the film was performed using the TEM and SEM methods. The film thickness were changed within the 0.2 – 5 μm.

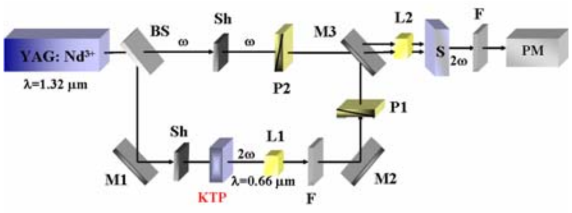

Fig. 1. Main schema of measurement set-up.

The photoinducing pulsed Nd :YAG laser providing the 1320 nm wavelength operates with a pulse duration of 40 ps and a frequency of 100 Hz (see Fig. 1). The optical second harmonics are generated by a single phase-matched KTP crystal. The pumping beam consists of a combination of the fundamental and the doubled frequency wavelengths. The ratio (**r**) of their intensities is changed from 15 to 70 by varying the KTP crystal angle from 3 to 17 degree with respect to the incident fundamental beam. A beam-splitter is used for the independent control of the incident light from the fundamental Nd :YAG laser. The polarisation of the incident bicolour coherent light on the sample is performed with the **Glahn** polariser. A synchronisation of the chopper with the Nd-YAG laser is ensured for a simultaneous light interruption. At the same time, the light source supplies a probing non-monochromatic beam slavered to the chopper whose desired frequency can attain 100 kHz The polarisation of the incident light is done with the polariser before irradiation of the sample. The monitoring of changes in the optical transmission spectrum is performed with the monochromator of about 2 nm spectral resolution directly connected with the **CCD** camera.

3. Results and discussion

In the Fig. 2 principal results concerning the pump dependences of the photoinduced SHG for different substrates of the ZnO are presented. One can see substantially better results for the MgO substrate compared to ZnO, BaO and CdO ones. We have established that maximum SHG is achieved at **r**=20 for all the substrates.

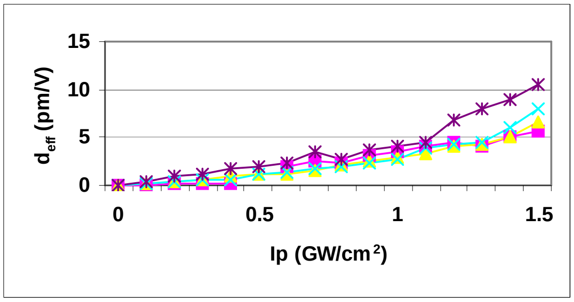

Fig. 2. Dependence of the output second order susceptibilities versus the pump power densities for different substrates: stars-MgO; X – ZnO; triangles – BaO; squares –CdO..

One can expect appearance of reconstructed near-the-surface structure possessing structural conformations corresponding to thermodynamically metastable (or even unstable) states. Usually optimization of the structure with inclusion of different molecular dynamics method consists in variation of principal structural parameters (bond lengths, angles, torsion angles etc.) to find a structural conformation corresponding to minimum of total energy. For the case of the ZnO we have the bulk-like perfect crystallites to which one can apply principles of long-range ordered translation symmetry. However, the long-range ordering usually is break in the interface region.

To perform structural optimization of such structure we have done the appropriate MD simulations following Ref. 5. The basic principle of approach in a use of equilibrium thermodynamics with non-equilibrium perturbation created by surrounding disordered background. The method may be applicable for many semiconductors and take into account a superposition of several structural fragments renormalized by appropriate weighting factors. Main principle of the approach consists in a choosing of several coordinated layers (usually not more than 6 nm) possessing perfect long-range ordering. To the border crystalline layers are added 2-3 layers which are structurally disordered. The geometry optimization is performed between the ordered crystalline layer and disordered ones assuming a fixed atomic positions for the crystalline layers.

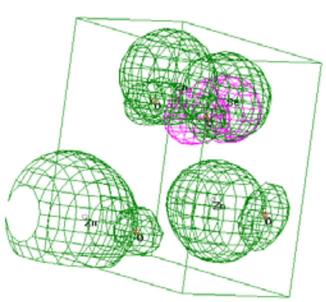

Fig. 3. Electrostatic charge density distribution in the ZnO crystallites deposited on the BaO ceramic substrate at pump power density equal to about 0.8 GW/cm^2.

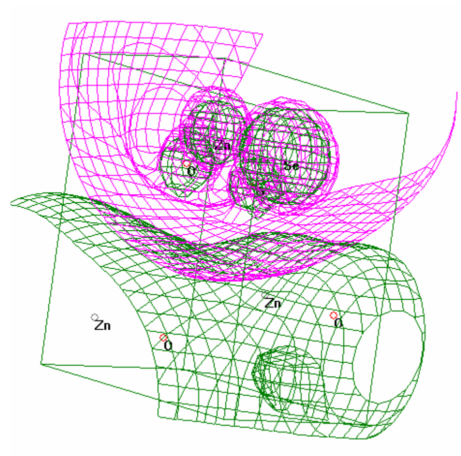

Fig. 4. The same at the pump power density equal to about 0.6 GW/cm^2 for CdO susbtrate.

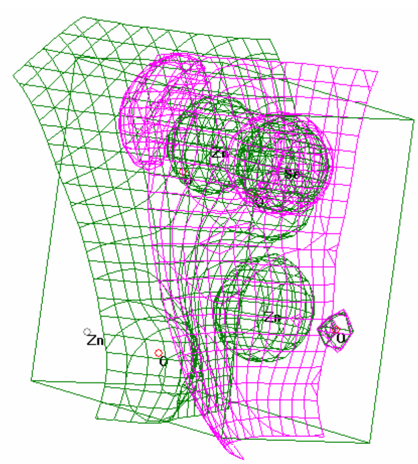

Fig. 5. The same at the pump power density 0.6GW/cm^2 for MgO substrate..

From the Fig. 3-5 one can clearly see how change of the substrate vary the electrostatic potentials indicated in the figures by different colors. This fact play a crucial role in the understanding of the bicolor coherent interactions.

4. Conclusions

Using coherent bicolor interactions we have investigated the SHG in the ZnO films deposited on different substrates: ZnO, MgO, CdO and BaO ceramics. We have shown that the MgO substrate is more preferable for the achievement the susceptibility up to 56 pm/V. Such giant susceptibility may be explained by forming of the hybrid interface structure substantially enhancing the output second-order susceptibility. At the same time the effect was maximal at T=261 K and at film thickness about 2.75 μm.

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