DETERMINATION OF THE OPTICAL NONLINEAR COEFFICIENTS SIGNS FOR ZnO FILMS AND SINGLE CRYSTAL

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Abstract. The epitaxial thin films and single crystals of ZnO were investigated by optical second harmonic generation (SHG). A method for characterization the optical nonlinear coefficients of ZnO are proposed. It is shown that the sign of d_{33} nonlinear coefficient is negative. Using the tensorial characteristic of optical SHG crystallographic orientation of noncentrosymmetric hexagonal media can be determined with an error less than $\pm 0.1^0$.

INTRODUCTION

ZnO is attractive material for many applications in optics and optoelectronics. [10]. Unique optical properties and high optical harmonics conversion efficiency of bulk ZnO crystals and films are especially important for integrated optics. Thin films of ZnO provide more efficient harmonic generation than crystals due to the influence of inter-grain boundaries [3,4,9]. This circumstance makes it possible to use the second harmonic generation (SHG) as convenient and sensitive method for testing textures of ZnO films fabricated by different techniques and conditions of growth. In this work, we report the results of comparative study of ZnO single crystal and epitaxial thin films on a sapphire substrate by SHG method.

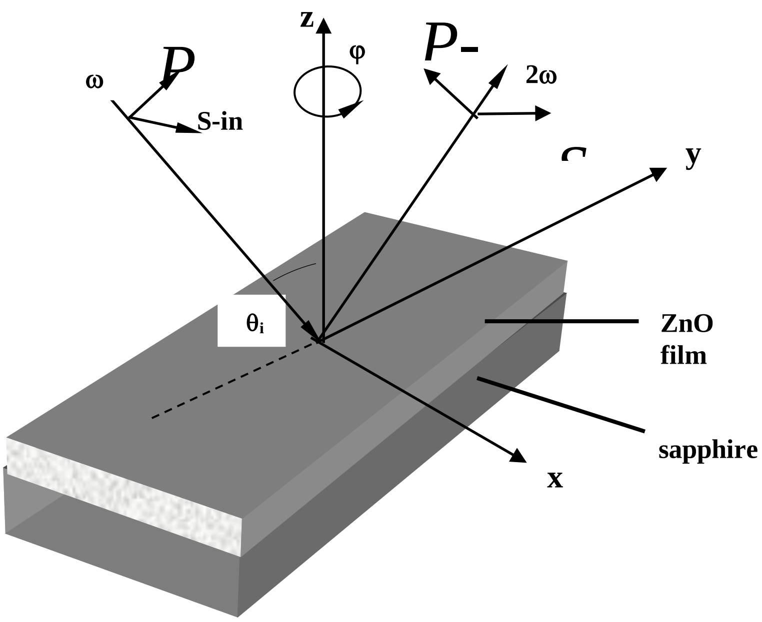
EXPERIMENTAL

Measurements were done using a Ti: sapphire laser radiation (fundamental wavelength λ_ω=780 nm, pulse width 100fs) focused onto the spot of about 100 μm in diameter at $\theta_i = 45^0$ incidences. The SH-radiation was detected with a photomultiplier tube and a standard photon counting system. A rotating a Berek compensator for varied polarization of the fundamental wave (*p*-in or *s*-in), and Glan prisms varied polarization of the SHG waves (*p*-out or *s*-out) were used. The SH intensity as a function of the sample rotation (azimuthal) angle φ were measured for *p*-in, *p*-out, *p*-in, *s*-out, *s*-in, *p*-out, and *s*-in, *s*-out polarization geometries (Fig. 1). The experimental dependencies are shown in Fig.2.

RESULTS AND DISCUSSION

We have developed a theoretical model that allows numerical simulation all measured dependencies [2]. In this model the multiple reflections of light on both fundamental (ω) and SH (2ω) frequencies into nonlinear media were taken into account. Real textured film is a plane-parallel single-crystalline slab, which is characterized by a few effective parameters.

However, bulk sample have only two ratios of independent components of nonlinear susceptibility tensor $\frac{d_{zxx}}{|d_{zzz}|}$ and $\frac{d_{zyy}}{|d_{zzz}|}$ $\frac{d_{yzy}}{|d_{zzz}|} = \frac{d_{xzx}}{|d_{zzz}|}$



In the model corresponding to the film appears three additional parameters such as the effective thickness of the film d_f and an Euler's angle, which determine the preferable orientation of principal C-axes of grains.

Fig.1.Configuration of both fundamental (ω) and SH (2ω) waves in the sample.

The magnitudes of relationships of the nonlinear optical coefficients were determined by the fitting procedure, which has been performed simultaneously for all four dependencies $I_{2\omega}(\varphi)$ measured on each sample.

$$\left(\frac{d_{zxx}}{|d_{zzz}|}\right)_{crystal} = 1{,}333 \qquad \left(\frac{d_{zxx}}{|d_{zzz}|}\right)_{film} = 1{,}327$$

$$\left(\frac{d_{yzy}}{|d_{zzz}|}\right)_{crystal} = 1{,}443 \qquad \left(\frac{d_{yzy}}{|d_{zzz}|}\right)_{film} = 1{,}411$$

One can see that the $\frac{d_{zxx}}{|d_{zzz}|}$ ratio values for crystal and film are practically the same, whereas the $\frac{d_{yzy}}{|d_{zzz}|}$ ratio is lager for the crystal. It has been found that the C-axes of grains in the film are oriented to the [10] direction by angle θ = 4,3^0 (for bulk crystal θ = 0^0). The effective thickness of the film is equal to 60 nm.

It should be noted that there is some ambiguity concerning the sign of the d_{zzz} component in different papers. According [1,11,12] the d_{zzz} sign is positive. Negative values are indicated in [5,7]. The same situation one can reveals in results of theoretical calculations of nonlinear susceptibilities. For example, results of microscopic calculations made by Jha and Bloembergen [6] are in good agreement experimental values only on magnitude, but not coincide on sign. The most successful calculations of the nonlinear susceptibilities apparently have been achieved by Levine [8].

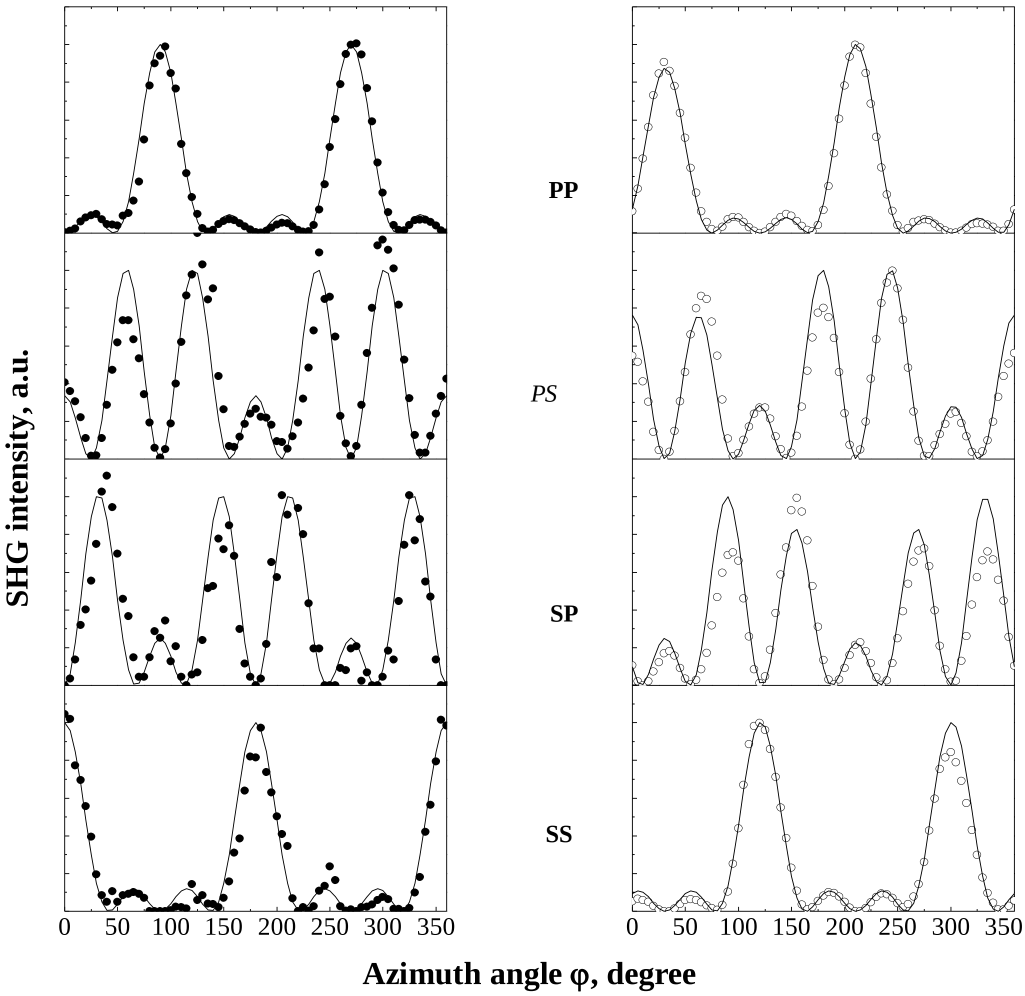


Fig. 2 Azimuthally SH intensities dependence measured in different polarizations for ZnO single-crystal (a) and thin film (b).

CONCLUSIONS

Comparative investigation of nonlinear properties of ZnO thin textured films and single crystal was performed. Unambiguous found that d_{zzz} component of nonlinear susceptibility tensor in both samples is negative. It was also determined the anisotropy of texture showing that nano-grains in the film are preferable oriented in the direction, which deviates from exact [110] crystal direction by an angle of about 4.3^o.

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REFERENCES

1. Bloembergen N., Nonlinear Optics, Benjamin W.A., Inc., New York-Amsterdam, 1965, p.424.
2. Buinitskaya G., Theoretical model of the multiple-reflection effects in optical second harmonic generation (ICMCS-02), Moldova, V.2. 2002, p.109.
3. Cao H., WU J.Y., Ong H.C., Dai J.Y., H. Chang R.P., Appl. Phys. Lett., **73**, (1998), 572.
4. Gang Wang, Kiehne G. T., Wong G. K. L., Ketterson J. B., Liu X., R. Chang P. H., Appl. Phys. Lett., **80**, (2002), 401.
5. Handbook of lasers, Vol.2 (in Russian) Edited by A.M. Prokhorov, Moskow, “Sovetskoe radio”, p.400.
6. Jha S. S., Bloembergen N., Phys. Rev.**171**, (1968), 891.
7. Kurtz S. K., Jerphagnon J., Choy M.M, Nonlinear Dielectric Susceptibilities, (Landolt-Bornstein), New Series, Vol.11,(ed. K.-H. Hellwege), Berlin:Springer-Verlay, 1979.
8. Levine B.F., Phys. Rev. B, **7**, (1973), 2600.
9. Newmann U., Grunwald R., Griebner U., Steinmeyer G., Seeber W., Appl. Phys.Lett.,**84**, (2004) 170.
10. Pearton S.J., Norton D.D., Ip K., Heo Y.W., Steiner T., Prog. Materials Science **50**,(2005) 293
11. Shen Y. R., The Principles of Nonlinear Optics, Willey, New york, (1989), p.558.
12. Sutherland Richard L., Handbook of Nonlinear Optics, (1996), p.68