OPTICAL SECOND HARMONIC GENERATION IN ZnO FILM:
MULTIPLE-REFLECTION EFFECTS

G. Buinitskaya, I. Kravetsky, L. Kulyuk, V. Mirovitskii, E. Rusu

Institute of Applied Physics, Academy str. 5, Kishinev, Moldova

The second harmonic (SH) generation by ZnO thin film on the glass substrate is investigated. The theoretical model perfectly describing the measured SH intensity as a function of incident angle is developed. It is shown that the optical axis of the effective nonlinear susceptibility of textured film is directed nearly along the normal of its surface.

INTRODUCTION

Optical SH generation is a well known and sensitive method for testing of surfaces, interfaces and thin nonlinear optical (NLO) films [1-5]. The SH generated upon reflection from a surface of NLO material has been treated for the first time by Bloembergen and Pershan [6]. Hase et al. [7] (see [8, 9] also) have extended this analysis taking into consideration that reflected (transmitted) SH beam includes a coherent superposition of the waves multiple reflected by surfaces of NLO thin film. It has been shown that the effect of multiple reflections of both the fundamental and SH waves within the film can not be neglected to agree the calculated SH intensity with measured one.

In this paper, we report the results of investigation of SH generation by thin ZnO film grown on glass substrate. We have measured the SH intensity as a function of incident angle of fundamental beam \( I_{2\omega} (\Theta) \) and developed the theoretical approach for calculation with a high accuracy the nonlinear susceptibility of thin film on a transparent substrate. In contrast to the models mentioned above we have taken into account the effect of multiple reflections of both fundamental and SH waves in the complete system "film + substrate", but not only in the film. It has allowed us to describe qualitatively the characteristic features of measured dependence of the SH intensity.

EXPERIMENT

Transparent and optically homogeneous ZnO textured films were prepared by chemical vapour deposition method [10]. The thickness of investigated ZnO films were 0.1 – 0.3 \( \mu \)m, i.e. approximately on an order of magnitude less than the coherence length of bulk zinc oxide \( (l_{coh}=\lambda_{0}/4[\pi(2\omega) - \pi(\omega)]=3\mu m) \) [11]. The second harmonic measurements in transmission were carried out on a standard experimental setup. A Q-switched YAG:Nd\(^{3+}\) laser operating at \( \lambda_{0}=1064 \) nm (pulse duration 20 ns, repetition rate 14 Hz) was used as a source of fundamental radiation. Laser pulse energy was maintained below 10 mJ in order to avoid optical damage of the film. The geometry of measurement (P-P type) is presented schematically in Fig.1. The measured dependence \( I_{2\omega} (\Theta) \) is shown in Fig.2.

THEORETICAL MODEL

We have considered theoretically the SH generation in the NLO thin film of ZnO compound grown on a transparent and isotropic dielectric substrate (glass). Both fundamental and SH waves propagating into the system are of TH-type, i.e. the vectors of magnetic field in all media are perpendicular to the incident plane as well as vectors of electric field lie in this plane (P-P polarised beams) (see Fig 1).

The following idealizations have been accepted in our model. First of all we have considered a textured film as a plane-parallel monocrystalline slab which is characterized by several effective parameters. These parameters are the refractive indices of the film at both fundamental (\( \omega \)) and SH (2\( \omega \)) frequencies, components of nonlinear susceptibility tensor and the thickness of the film. They should be found by fitting the results of theoretical calculations to the experimental data. Then we have assumed the absorption in system at both frequencies as well as anisotropy of linear optical
properties of the film as negligible. Our aim has concluded in the calculation of angular dependence of SH intensity transmitted through this simplified system. The SH in every medium should obey the wave equation with inhomogeneous term containing the source of polarization distributed in the bulk

\[
H_i' = H_i^{(0)}
\]

\[
E_i = E_i^{(0)}
\]

\[
H_x'^{R} = H_x^{(0)}
\]

\[
H_y'^{R} = H_y^{(0)}
\]

\[
E_z = E_z^{(0)}
\]

\[
\text{air (}\alpha=0) \quad \Theta_i \omega \quad 2\omega \omega
\]

\[
\text{film (}\alpha=1) \quad a \quad H_2^{(1)} H_4^{(1)} H_2^{(1)} H_1^{(1)}
\]

\[
\text{glass (}\alpha=2) \quad b \quad H_2^{(2)}
\]

\[
\text{air (}\alpha=3) \quad \Theta_i \omega \quad 2\omega \omega
\]

\[
H_2^{(3)} \equiv H_2^{(3)}
\]

\[
H_1^{(1)} \equiv H_1^{(1)}
\]

\[\text{Fig 1. Signs } H_i^{(\alpha)} \text{ denote vectors of magnetic field in medium } \alpha; \ i=1,2 \text{ corresponds to the fundamental (}\omega) \text{ and SH (}2\omega) \text{ wave, respectively; } H_i', H_i^T, H_i^R \text{ correspond to incident, transmitted and reflected waves; } \Theta_i \text{ is the angle of incidence of the fundamental wave on the system; } E_i^{(\alpha)} \text{ is the vector of electric field of the incident wave.}
\]

\[\nabla \times \nabla \times E^{(\alpha)} = -\left[ \frac{n_{\alpha}^2}{c^2} \frac{\partial^2 E^{(\alpha)}}{\partial t^2} + \frac{4\pi}{c^2} \frac{\partial^2 P^{(\alpha)}}{\partial t^2} \right]
\]

This polarization is nonlinear because it is quadric on the components of electrical field of fundamental wave. The latter can be represented as a superposition of the waves \( E_i^{(1)} \) and \( E_i^{(1)'} \) which move forwards and backwards, respectively (see Fig.1). Both of them are determined by multiple reflections of the fundamental wave in the complete system. The resulting vector of the electric field lies in the incident plane, i.e. it contains only \( y \) and \( z \) components, whose amplitudes are calculated numerically imposing boundary conditions on components \( H_x^{(\alpha)}(\omega) \) and \( E_y^{(\alpha)}(\omega) \) at all three interfaces of layered media. The general solution of eq. (1) consists of the solution of the homogeneous equation plus one particular solution of the inhomogeneous equation. The expression for tangential components of electric and magnetic vectors of the homogeneous equation solution were

\[H_x^{(\alpha)}(2\omega) = x \left( F_{\alpha} e^{-ik_{\alpha}\mu} + G_{\alpha} e^{ik_{\alpha}\mu} \right) e^{ik_{\alpha}(y-y_0)}
\]

\[E_y^{(\alpha)}(2\omega) = y \left( \frac{c}{2\omega} \right) \left( \frac{k_{\alpha} \omega}{n_{\alpha,2\omega}} \right) \left( F_{\alpha} e^{-ik_{\alpha}\mu} - G_{\alpha} e^{ik_{\alpha}\mu} \right) e^{ik_{\alpha}(y-y_0)}
\]

where \( k_{\alpha y} = 2k_{0 y} = 2k_{1 y} = 2k_{2 y} = 2k_{3 y} = \frac{2\alpha \omega_{alpha}}{c} \sin \Theta_i; \ k_{\alpha z} = \sqrt{\left( \frac{2\omega}{c^2} \frac{n_{\alpha,2\omega}}{2\omega} - k_{\alpha y}^2 \right)} \)
The particular solution of the inhomogeneous equation is nonzero only in the NLO film and can be represented as follows:

\[
E_{\text{NL}}^{(1)}(2\omega) = E^{(1)}(2\omega) = \frac{c}{2\omega} \left[ k_{\text{by}} \left( \gamma_4 e^{-ik_{\text{by}}z} + \gamma_5 + \gamma_6 e^{ik_{\text{by}}z} \right) + k_{\text{h}} \left( \gamma_4 e^{-ik_{\text{h}}z} + \gamma_5 + \gamma_6 e^{ik_{\text{h}}z} \right) \right] e^{i(k_{\text{by}} y - 2\omega t)}
\]

where \( k_{\text{by}} = 2k_{11} \), \( k_{\text{h}} = 2k_{12} = 2\sqrt{\frac{\omega n_{\text{by}}^2 - \omega c^2}{c^2}} \) and \( \gamma_i \) are constants which are determined by substituting vectors \( E^{(1)}(2\omega) \) in equation (1). The boundary conditions, i.e. the continuity of tangential components of vectors of SH fields are \( E^{(1)}(2\omega) = E^{(1)}(2\omega) \), \( H^{(1)}(2\omega) = H^{(1)}(2\omega) \). Their numerical solution has allowed us to determine the amplitudes of SH beam transmitted through the system. Its intensity is equal to

\[ I_{2\omega}(\Theta_i) \propto E^T(2\omega) \cdot \left( E^T(2\omega) \right)^* \]

and has been calculated numerically.

RESULTS

Varying model’s parameters we have achieved a good agreement of calculated dependence \( I_{2\omega}(\Theta_i) \) with measured one (see Fig.3). The best fit has been obtained for the following ratios of components of the effective second-order susceptibility \( d_{33} = 1.9 \) and \( d_{11} = 2.1 \). They are larger than correspondent relations for bulk zinc oxide (0.3 and 0.33, respectively [12]) We have also found that the value of nonlinear refractive index of ZnO film (\( n_2 = 2.053 \)) is very close to the tabulated one. At last the effective thickness of the investigated film has been found about 0.28 \( \mu m \).

Besides we have explained a few important features in the experimental curve (compare Fig.2 and Fig.3). First, the quasi-periodic oscillations which become frequent with increase of \( \Theta_i \) are determined mainly by refractive index and thickness of substrate. Second, the asymmetry in amplitudes and locations of these peaks depends on preferred orientation of micro-crystals composing the film’s texture. Due to the high sensitivity of \( \delta \)-tensor to crystalline orientation we have found out that the principal \( (z) \) axis of the tensor deviates from the normal to the film surface on a small angle (1°, 38') in the incident plane. Third, the fact that SH intensity does not vanish even at \( \Theta_i = 0 \) is explained by diffuse scattering of both fundamental and SH waves on the inhomogeneous structure of the film. And at last two maxima in the envelope curve of experimental dependence are located approximately at \( \pm 50^\circ \) and determined by the film thickness as well as the ratios of \( \delta \)-tensor components. The difference in their heights is also explained by mentioned deviation of principal axis of texture’s \( \delta \)-tensor from the normal. As to another feature in the area of these maxima near \( \pm 56^\circ \) (indicated by arrows in Fig.3) it can be explained by the fact that Brewster angle for glass-air interface is equal to 56°19'. It means that in the P-P geometry of the experimental setup the reflected fundamental wave into substrate is vanished together with the amplitude of correspondent quasi-periodic oscillations.

So, an essential role of multiple reflections of light into complete optical system "film + substrate" on the form of \( I_{2\omega}(\Theta_i) \) dependence of SH generation has been revealed here by proposed theoretical model.

REFERENCES