International Journal of Innovative Technology and Exploring Engineering (IJITEE)
ISSN: 2278-3075 (Online), Volume-5 Issue-9, February 2016

Phase Effects at Second Harmonic Generation in Zinc Oxide, Grown on Glass Substrate

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Abstract- Theoretical investigation of frequency conversion in ZnO films laid over glass substrates with account for phase effects has been developed. For this the constant-intensity approximation of fundamental radiation is applied. The numerical calculation of the efficacy obtained in constant-intensity approximation confirms the following that because of dispersion of the second order nonlinear optical coefficients the generated signal decreases for an increasing fundamental wavelength. Furthermore, the zinc oxide films generate stronger second harmonic signal because of the larger interaction length of the nonlinear medium. Method of analysis of second harmonic generation in zinc oxide, grown on glass substrate used in the present work, may be involved for research of other films.

Keywords: nanocompo site film; glass substrate; second harmonic generation; constant-intensity approximation; frequency conversion.

I. INTRODUCTION

In order to be able to process all of optical signals we need to have developed switches for example all optical switches and this requires new nonlinear materials. Among those required candidate one must pay attention to good optical quality (e.g. small amount of dissipation) [1, 2]. Among these ZnO could be considered as an interesting semiconductor with a wide band gap of 3.3 eV at the room temperature, which could be combined with high excitonic gain and binding energy [3]. According to recent studies, ZnO thin films happen to have a strong nonlinear suscepitibilty of second-order which cause for them to be a candidate for generating effective second harmonics [4, 5].

Nanosize structure technology is being used in order to construct miniaturized versions of these devices. This implies deploying new method, which is using of thin dielectric and semiconductor films instead of bulk material itself as the technology of bulk, also, is not very economical because of their requirement of developed technology of crystal growth and time consuming processing. The morphology and structure of these compounds could be informative via exploration of semiconductors and dielectrics by harmonic generation method [6]. It was outlined in work [7] that the studied patterns were manufactured based on ZnO films over a glass substrate using the technology of sputtering ion beam.

Manuscript published on 28 February 2016.
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This work illustrates the theoretical model for the analysis of the experimental data and also is going to discuss preliminary deductions for the structure of the ZnO crystalline relying on measurements of nonlinear optical properties carried out by authors. A couple of works, e.g. [8-10] are dedicated to the theory of nonlinear interaction in the thin films of zinc oxide, particularly, at second harmonic generation (SHG). Authors of these works deploy the analysis of frequency conversion, particularly, the constant-field approximation (CFA) [11-12]. For the case of CFA, the coherent length of nonlinear medium is the total function of mismatch of wave vectors. This approximation will only be acceptable at the initial phase of the interaction for we can ignore the effect of the excited harmonic waves of basic radiation. This will cause losing some information regarding qualitative features of nonlinear process. To analyze the process, we can make direct numerical solution of reduced equation. This will allow us to obtain exact analytical expressions and determining the appropriate parameters in order to get highest conversion efficiency using optimized analytical method.

We can calculate the phase change and losses for interacting waves simultaneously which is the advantage of constant intensity approximation CIA [13-14] while considering the reverse reaction of the excited wave over pump wave. Furthermore, the coherent length in this approach depends on parameters such as basic radiation intensity and dissipation in the medium besides mismatch of interacting waves. We are going to analyze the CIA for nonlinear interaction for generating of second harmonic in ZnO samples laid over glass substrates while considering the losses and phase changes of all interacting waves. It is proved though because of the dispersion regarding second order nonlinear optical coefficients, the generated signal decrease as the fundamental wavelength increase. We are also going to analyze factors causing restriction of efficiency conversion.

II. THEORY

Here we are going to take a closer look at redoubling frequency process for laser radiation with the frequency of ω_1 for ZnO with noncentrosymmetric structure for the case of scalar phase matching of first kind. For nonlinear conversion, the known system of reduced equation is deployed to analyze the interaction of waves to describe second harmonic generation with the frequency of ω_2 while considering linear losses in a structure [11-12]

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$$\frac{dA_1}{dz} + \delta_1 A_1 = -i\gamma_1 A_2 A_1^* \exp(-i\Delta z),$$
$$\frac{dA_2}{dz} + \delta_2 A_2 = -i\gamma_2 A_1^2 \exp(i\Delta z), \quad (1)$$

Here, $A_{1,2}$ are the complex amplitudes of the pump wave and its second harmonic at frequencies $\omega_{1,2}$ ($\omega_2 = 2\omega_1$), k_1, k_2 stand for wave vectors of the pump and second harmonic correspondingly, $\Delta = k_2 - 2k_1$ signifies the phase mismatch, $\gamma_{1,2}$ are the nonlinear coupling coefficients and $\delta_{1,2}$ are coefficients for waves with frequency of $\omega_{1,2}$ correspondingly. Let's assume that for the input the field ω_2 is absent and there is only one amplitude of A_1 field ω_1 with nonzero value which will cause the boundary condition to take the following form

$$A_1(z=0) = t_{af} \cdot A_{10} \exp(i\varphi_{10}), \quad A_2(z=0) = 0, \quad (2)$$

Here, z=0 signifies the input for our crystal, t_{af}^{ω} stands for the Fresnel transmission coefficient for the boundary of air-film for fundamental beam and φ_{10} is the initial phase of pump wave at the entry of the medium.

Then we are going to find a new answer using appropriate boundary conditions. The equations mentioned in (1) is another candidate to be the answer for complex amplitude of second harmonic at the output ($z = \ell$) of the structure in CIA ($I_1(z) = I_1(z=0) = I_{10}$) [13-14]

$$A_2(\ell) = -i\gamma_2 \cdot t_{fs}^{2\omega} \cdot t_{sa}^{2\omega} \cdot \left(t_{af}^{\omega}\right)^2 A_{10}^2 \ell \,\mathrm{sinc}\lambda\ell \exp\left[2i\varphi_{10} - \left(\delta_2 + 2\delta_1 - i\Delta\right)\ell/2\right], \quad (3)$$

Here we have introduced the quantities

$$\lambda^2 = 2\Gamma^2 - \left(\delta_2 - 2\delta_1 + i\Delta\right)^2/4,$$
$$\Gamma^2 = \gamma_1\gamma_2 I_{10} \left(t_{af}\right)^2,$$
$$\mathrm{sinc}x = \sin x / x, \qquad I_j = A_j A_j^*,$$
$$\gamma_1 = \frac{8\pi^2 \chi_{1,eff}^{(2)}}{\lambda_1 n_\omega}, \qquad \gamma_2 = \frac{4\pi^2 \chi_{2,eff}^{(2)}}{\lambda_2 n_{2\omega}}.$$

Where $\chi_{1,2\,eff}^{(2)}$, $\lambda_{1,2}$ and $n_{\omega,2\omega}$ are effective quadratic nonlinear coefficients, wavelengths and refractive indices in this zinc oxide films at frequencies $\omega_{1,2}$, respectively, and $t_{fs}^{2\omega}$, $t_{sa}^{2\omega}$ are the Fresnel transmission coefficients for film-substrate-air system for the second harmonic beam.

As we can see from (3), the basic and harmonic fields energy interchange occur periodically which result in the observation of spatial variations of the second harmonic field. For this, the minimums of harmonic intensity beating, according to analysis achieved from CIA, will depend on nonlinear susceptibility of the crystal. This allows us to define the nonlinear susceptibilities of substances by a simple way more precisely while compared to the case of CFA [15]. From (3) the optimum length of film-converter could be achieved by following formula

$$\ell_{opt} = \lambda^{-1} \arctan(\lambda / \delta_2), \quad (4)$$

Here $\lambda^2 = 2\Gamma^2 + \Delta^2/4$.

A harmonic signal at this length for the frequency conversion will be maximal. Based on theoretical investigation via CIA and comparing it to CFA the optimum length of crystal (coherent length of nonlinear medium) will be affected by the pump intensity, I_{10}, and dissipation in medium [13-14]. For the case of $\delta_{1,2}$=0 the optimum length (the coherent length again) will be given by $\ell'_{opt} = 0.5\pi / (2\Gamma_1^2 + \Delta^2/4)$. In case of $\delta_{1,2}$=0 the optimum length, that is coherent length, is expressed by $\ell'_{opt} = 0.5\pi / (2\Gamma_1^2 + \Delta^2/4)$.

For the case of CFA, (γ_1=0 and δ_j=0), we will get from (3) the relation for conversion efficiency and using (4) the well-known expression for coherent length will be $\ell'_{opt} = \pi / \Delta = k_2 - 2k_1$.

The relation for SH intensity could be obtained from Eq. (3). The conversion efficiency, which is $\eta_2(\ell) = I_2(\ell)/I_{10}$, gives following relation while ignoring reflections (pay attention $(\delta_2 = 2\delta_1)$)

$$\eta_2(\ell) = \gamma_2^2 I_{10} \ell^2 \left(t_{af}^{\omega}\right)^2 \left(t_{fs}^{2\omega}\right)^2 \left(t_{sa}^{2\omega}\right)^2 \mathrm{sinc}^2\lambda\ell \exp\left(-2\delta_2\ell\right), \quad (5)$$

where t_{af}^{ω}, $t_{fs}^{2\omega}$, $t_{sa}^{2\omega}$ - stand for the Fresnel transmission coefficients (air-film-substrate-air system) for fundamental and second harmonic beams.

In order to increase the efficiency of conversion frequency in ZnO film of laser radiation we are going to do some calculation of the analytical expression for conversion efficiency obtained from CIA (5).

For the special case of consideration here we, instead of ordinary thickness, are going to introduce equivalent thickness of nonlinear medium $\ell_{eff} = \ell / \cos\theta$ where ℓ signifies the thickness of ZnO film.

As a result, Eq. (5) will take the following form

$$\eta_2(\ell_{eff}) = \gamma_2^2 I_{10} \ell_{eff}^2 \left(t_{af}^{\omega}\right)^2 \left(t_{fs}^{2\omega}\right)^2 \left(t_{sa}^{2\omega}\right)^2 \mathrm{sinc}^2\lambda'\ell_{eff} \exp\left(-2\delta_2\ell_{eff}\right), \quad (6)$$

where

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$$\lambda'^2 = 2\Gamma^2 + \Delta^2 / 4,$$

$$\frac{\Delta'}{2} = \frac{2\pi}{\lambda_1} \ell \left(n_{2\omega} \cos\theta_{2\omega} - n_\omega \cos\theta_\omega \right) \text{ [16].}$$

Here θ stands for the incidence angle of laser beam, θ_ω, $\theta_{2\omega}$ represent the refractive angles for fundamental and second harmonic waves, correspondingly. The refractive indices for ZnO have been calculated according to Sellmeier model [7].

III. RESULTS AND DISCUSSION

The frequency conversion and its dynamic process are depicted in Figs. 1-6 for the second harmonic in case of zinc oxide which is grown by dual ion beam sputtering method. The dependencies are studied using the nonlinear wave interaction analysis in CIA. Furthermore, Fresnel transmission coefficients, refraction phenomena in the film and different values of $d_{ijk} = \frac{1}{2}\chi^{(2)}_{ijk}$ [17] at three different polarization configurations ($s_\omega - p_{2\omega}, 45_\omega - p_{2\omega}, \ p_\omega - p_{2\omega}$) have been considered which results of the experiment [7] are.

We have chosen parameters the way to fulfill the conditions for current experiment regarding ZnO film. As a fundamental beam, we have considered the output beam of a Q-switched Nd:YAG laser generating at wavelength of 1064 nm with 7 ns pulse width as its duration and repetition rate in the range of 1 - 14 Hz. The polarization of fundamental beam is varied from p_ω (responsible for highest signal) to s_ω (the lowest one). Raman cell can be beneficial here to change the fundamental beam from 1542 to 1907 nm. The **r**esolving power of incidence angle will be 0.01 degree.

The Sellmeier coefficients for ZnO are measured in [18] and we will use them to calculate the ordinary and extraordinary refractive indices for mentioned samples.

The coefficients for dissipation of ZnO films over glass substrates are missing due to lack of experimental data so the same order of coefficients are chosen (which is from the order of ($\sim 10^4$ cm^{-1}).

Fig. 1 shows the numerical calculation of the analytical relation (6) that we have from CIA over the length of nonlinear interaction ℓ for different value of parameters. $p_\omega - p_{2\omega}$ is considered as polarization configuration and the wavelength of the laser radiation is 1064 nm. From behavior of curves differed from monotonous increase in case of CFA (curve 4) it follows that there exists optimum value of crystal length at which conversion efficiency is maximum. By decreasing of pump intensity (compare curves 1 and 3) and increasing the losses (compare curves 2 and 3), the conversion efficiency will decrease. Besides this, the period of spatial beatings and optimum length of crystal will also increase. To explain this we should consider that for lesser value of pump intensity we need higher geometrical length of crystal in order to achieve maximum conversion. As we can see from figure by increasing the pump intensity by three orders we will be able to observe three times of increasing in conversion efficiency approximately. At δ_2 =0.22 × 10^{4}cm^{-1}, based on relation obtained from CIA, the maximum efficiency equates to 5.78 × 10^{-6} when the coherent length of ZnO film is 3.6 mcm (curve 3) and for losses δ_2 =0.3 × 10^4 cm^{-1} the maximum efficiency is 3.48 × 10^{-6} this time for 3.34 mcm coherent length of the same film (curve 2).

For Laser generating at 1064 nm of wavelength we see the dependency of conversion efficiency $\eta_2(\theta)$ in Fig. 2 for three different configurations of Lasers ($s_\omega - p_{2\omega}$, $45_\omega - p_{2\omega}$, $p_\omega - p_{2\omega}$). To vary the length of nonlinear interaction in experiment we rotated the ZnO sample causing angle θ to change. In addition to these, the form of polarization of beam varies from p_ω, 45_ω to s_ω.

Let's see the behavior of curves in Fig 2. By rotating the polarization of pump beam from p_ω to 45_ω or p_ω to s_ω we see the falling of conversion efficiency (compare curves 1, 3 and 1, 5). Here the dependencies between the cases of $s_\omega - p_{2\omega}$ and $45_\omega - p_{2\omega}$ completely coincide (curve 1). By increasing the pump intensity from 1 GW/cm^2 (curve 3) to 1.5 GW/cm^2 (curve 4) and then to 3 GW/cm^2 (curve 5) we observe approximate increase of 3 order of magnitude for efficiency of conversion. This is being mentioned as note in an earlier experiment [7].

Fig 3 has resulted the dependency of conversion efficiency for second harmonic over the incidence angle of pump wave (θ) for three different value of wavelength when the polarization configuration is $p_\omega - p_{2\omega}$. Figure tells us that the generated signal decrease by increasing the fundamental wavelength because of dispersion for second order nonlinear optical coefficients.

Now we want to consider the practical example of the nonlinear interaction for optical waves at second harmonic generation for the ZnO film. During the process of doubling the generation of radiation occurs in experiment for the wavelength of the order of 0.532 mcm. We want to approximately measure the conversion efficiency for three different configuration of polarization varied among $s_\omega - p_{2\omega}$, $45_\omega - p_{2\omega}$ and $p_\omega - p_{2\omega}$.

The dependencies of the analytical relation for signal of the second harmonic achieved in CIA over the incidence angle θ have been displayed in Fig. 4. Changing the angle in the given spectrum permits one to vary ℓ_{eff} on micrometer scale. So, the effective length, ℓ_{eff} , is changed in the range of 0.4÷8 mcm, while considering the fact that the thickness for experiment is 550 nm [7]. The diapason of change of length ℓ_{eff} of nonlinear interaction corresponds to the central part of considered theoretical dependency of $\eta_2(\ell)$

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(see Fig. 1), where efficiency is maximum. From here we will be able to obtain the maximal conversion efficiency, $\eta_2(\ell_{eff})$, via optimal length of sample.

Besides this, the numerical calculation of the efficiency achieved from CIA prove that for smaller wavelength we will have stronger second harmonic signal by film because of dispersion for nonlinear optical coefficients $\gamma_{1,2}$ (this could be seen from curves 1-3).

Let's now compare the simulation results of the azimutal angular dependencies of second-harmonic generation intensity for $0 \div 100^0$ scan, calculated in the CIA (dashed curves 2 and 5) and experimentally measured values (solid curves 1, 3 and 4) [7] (Fig. 4). It is for the case of laser generating at 1064 nm wavelength and three different $(s_\omega - p_{2\omega},\ 45_\omega - p_{2\omega},\ \ p_\omega - p_{2\omega})$ polarization configurations. We see complete matching of theoretical dependency for $s_\omega - p_{2\omega}$ and $45_\omega - p_{2\omega}$ in curve 2. By comparing these curves the theoretical and experimental results will be the same. The best matching between these results will be obtained for more accurate values of task parameter (losses for investigated samples of ZnO films grown on glass substrate, substrate refractive indices at the 1064 nm and others).

Analogical behavior of the azimuth angular dependencies for second-harmonic generation intensity occurs for laser with 1542 nm (Fig. 5) and 1907 nm (Fig. 6) of wavelengths. The similar comparison will not be possible due to lack of analogical experimental dependency for mentioned wavelength. The width of angular phase matching is reduced by increasing the wavelength from 1542 nm to 1907 nm (Figs. 5 and 6). The case of 1064 nm is the one that we get the lowest amount of contribution in the width of angular phase matching (Figs. 3 and 4-6).

IV. CONCLUSION

Thus, theoretical investigation of frequency conversion is taken into consideration with account for phase effects in ZnO films laid over glass substrates. Namely at the given values of the length of crystal converter, it will be possible to calculate the coherent length of nonlinear medium. The analytical method also allows one to calculate the expected conversion efficiency approximately over the different wavelengths of laser radiation. Thus, the numerical calculation of the efficacy obtained in CIA confirms the following that because of dispersion of the second order nonlinear optical coefficients the generated signal decreases for an increasing fundamental wavelength. Besides this, the ZnO films generate stronger second harmonic signal because of the larger interaction length of the nonlinear medium. The result we got here can be used in order to get modern devices of quantum electronics at nanoscale optoelectronic level. Method of analysis for second harmonic generation for the case of ZnO films discussed in the present work may be used to investigate other nanocomposite films.

ACKNOWLEDGEMENT

This work was partially supported by the Science Development Foundations under the President of the Republic of Azerbaijan - **Grant № EIF-2013-9(15)-46/04/1.**

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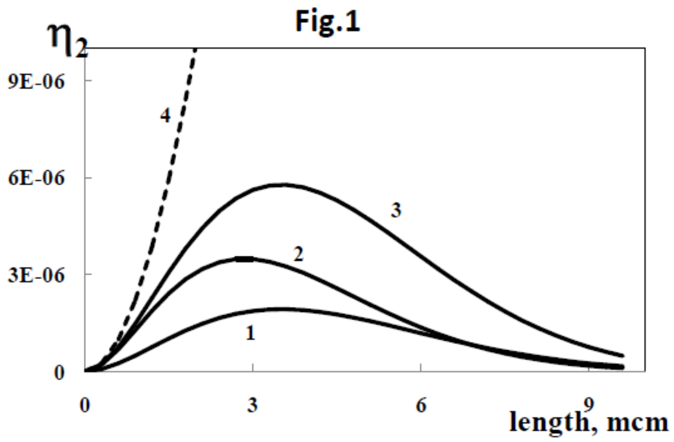

Fig. 1. Dependencies of conversion efficiency of pump wave (λ=1.064 mcm, $p_{\omega} - p_{2\omega}$ polarization configuration) to the wave of the second harmonic η_2 on lengths calculated in the CIA for ZnO films ℓ calculated in CIA (curves 1-3) and CFA (curve 4) for pump intensity of I_{10} = 3 GW/cm^2 (curves 2, 3 and 4), 1 GW/cm^2 (curve 1) and $\delta_2 = 2\delta_1$=0 (curve 4), 0.22*10^4 cm^{-1} (curves 1 and 3), 0.3*10^4 cm^{-1} (curve 2).

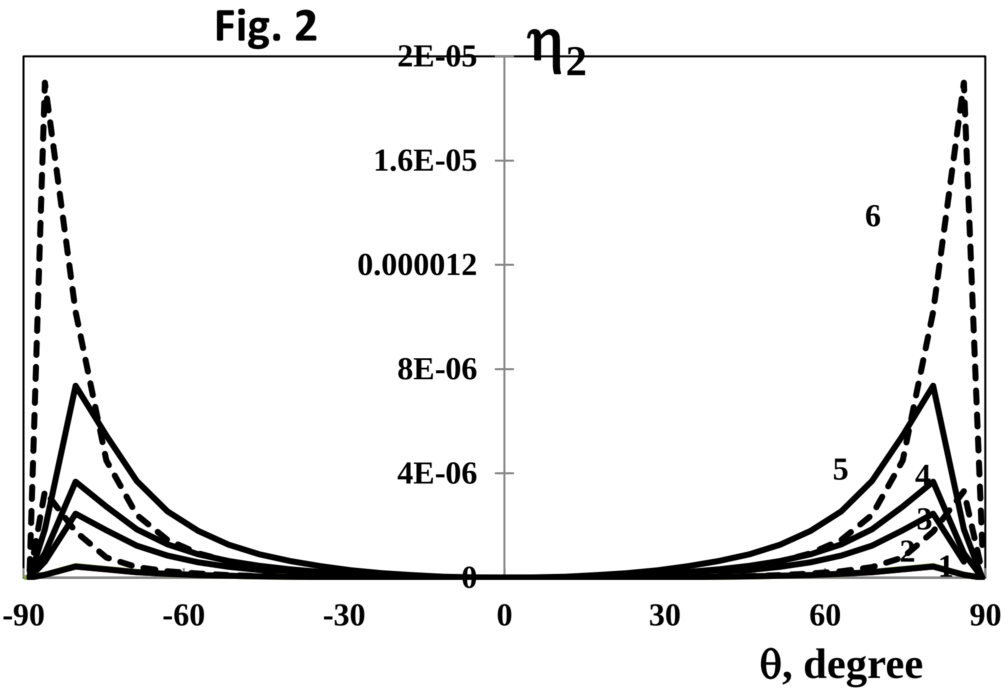

Fig. 2. Dependencies of conversion efficiency of pump wave (λ=1.064 mcm, $p_{\omega} - p_{2\omega}$ polarization configuration) to wave of second harmonic η_2 calculated in CIA versus the incidence angle θ for ZnO films for pump intensity of I_{10} = 1 GW/cm^2 (curves 1, 3 and 6), 1.5 GW/cm^2 (curve 4) and 3 GW/cm^2 (curves 1, 2 and 5) and $\delta_2 = 2\delta_1$=0 (dashed curves 2 and 6), 0.22*10^4 cm^{-1} (curves 1, 3-5).The thickness of all films is equal to 550 nm [7].

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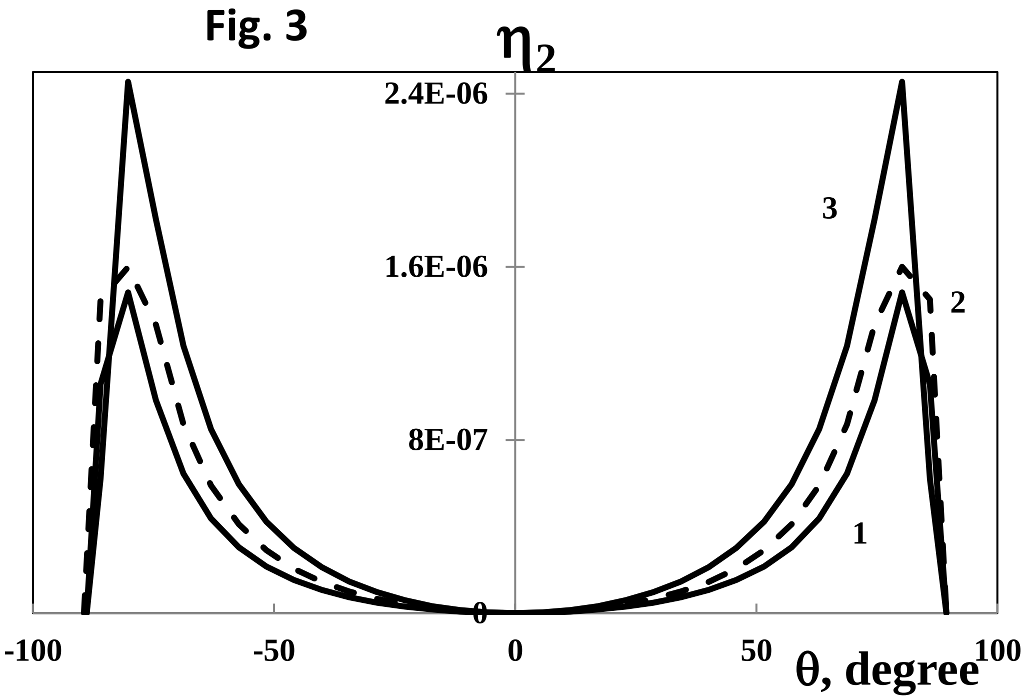

Fig. 3. Dependencies of conversion efficiency of pump wave, where λ is equal to 1.064 mcm (curve 3), 1.542 mcm (curve 1) and 1.907 mcm (curve 2) to wave of second harmonic η_2 calculated in the CIA (for $p_\omega - p_{2\omega}$ polarization configuration) as a function of the incidence angle θ for ZnO films for pump intensity of I_{10} = 1 GW/cm^2 (curves 1- 3) at $\delta_2=2\delta_1=0.22*10^4$ cm^{-1}.The thickness of all films is equal to 550 nm [7].

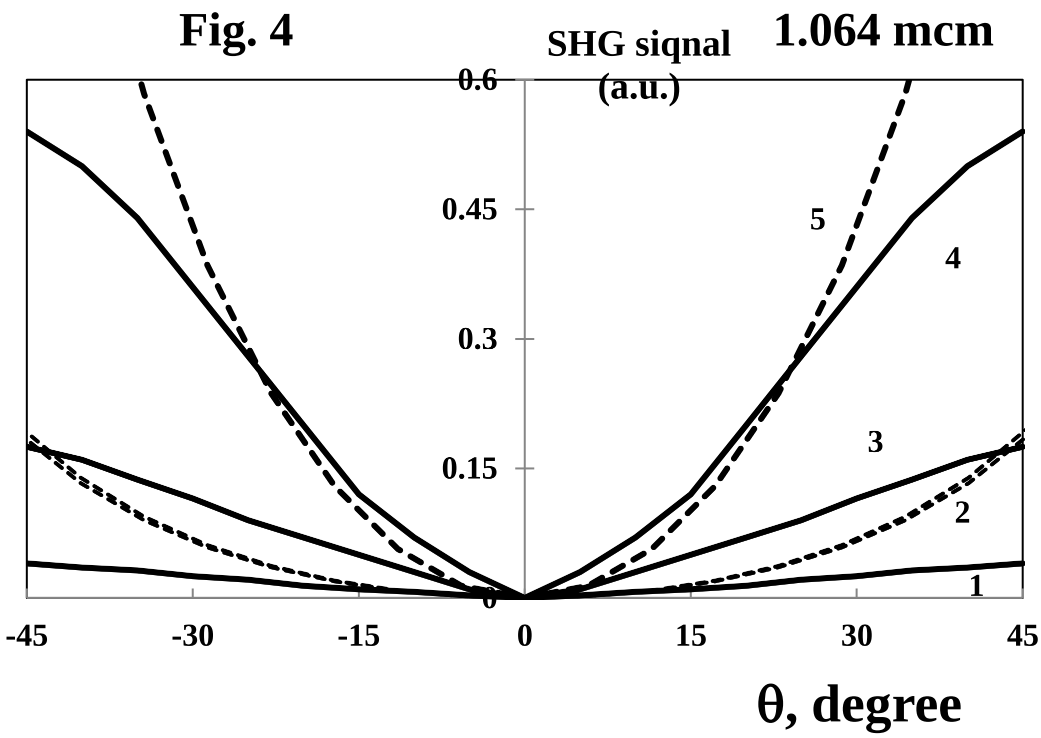

Fig. 4. Dependencies of the analytical expression for intensity of the second harmonic (pump wave λ=1.064 mcm) as a function of the incidence angle θ calculated in the CIA (dashed curves 2 and 5) for ZnO films at $s_\omega - p_{2\omega}$ (curves 1 and 2), $45_\omega - p_{2\omega}$ (curves 2 and 3) and $p_\omega - p_{2\omega}$ (curves 4 and 5) polarization configurations. Experimental results are given (solid curves 1, 3 and 4) from [7]. Here pump intensity of I_{10} = 1 GW/cm^2 (curves 1- 5) at $\delta_2=2\delta_1=0.22*10^4$ cm^{-1}.The thickness of all films is equal to 550 nm [7].

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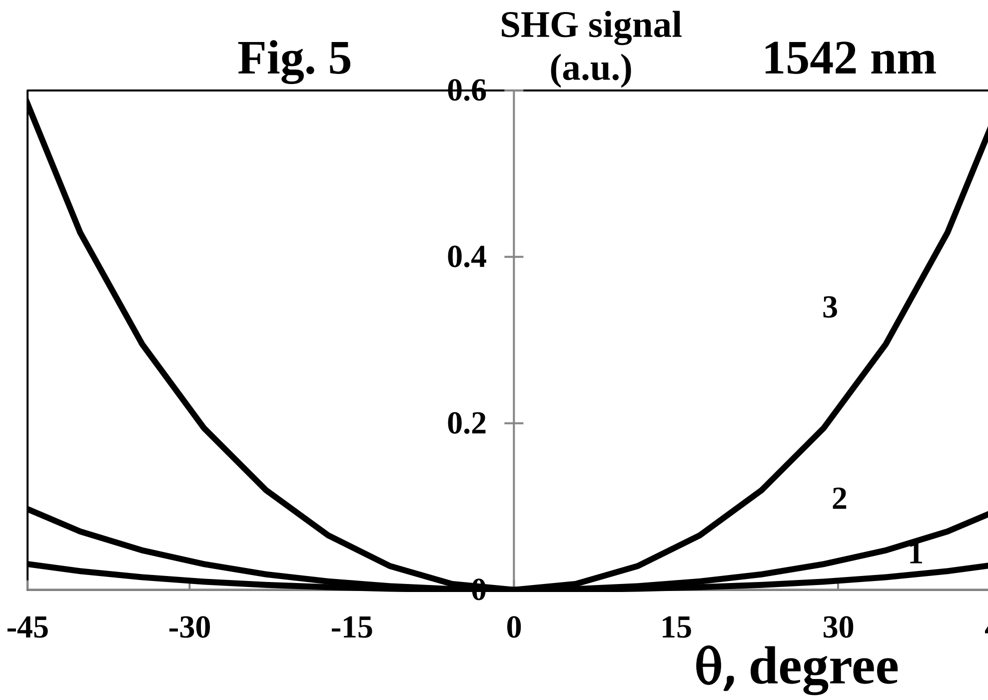

Fig. 5. Dependencies of the analytical expression for intensity of the second harmonic (pump wave λ=1.542 mcm) as a function of the incidence angle θ calculated in the CIA for ZnO films at $s_{\omega} - p_{2\omega}$ (curve 1), $45_{\omega} - p_{2\omega}$ (curve 2) and $p_{\omega} - p_{2\omega}$ (curve 3) polarization configurations at pump intensity of I_{10} = 1 GW/cm^2 and $\delta_2=2\delta_1=0.22*10^4$ cm^{-1}.The thickness of all films is equal to 550 nm [7].

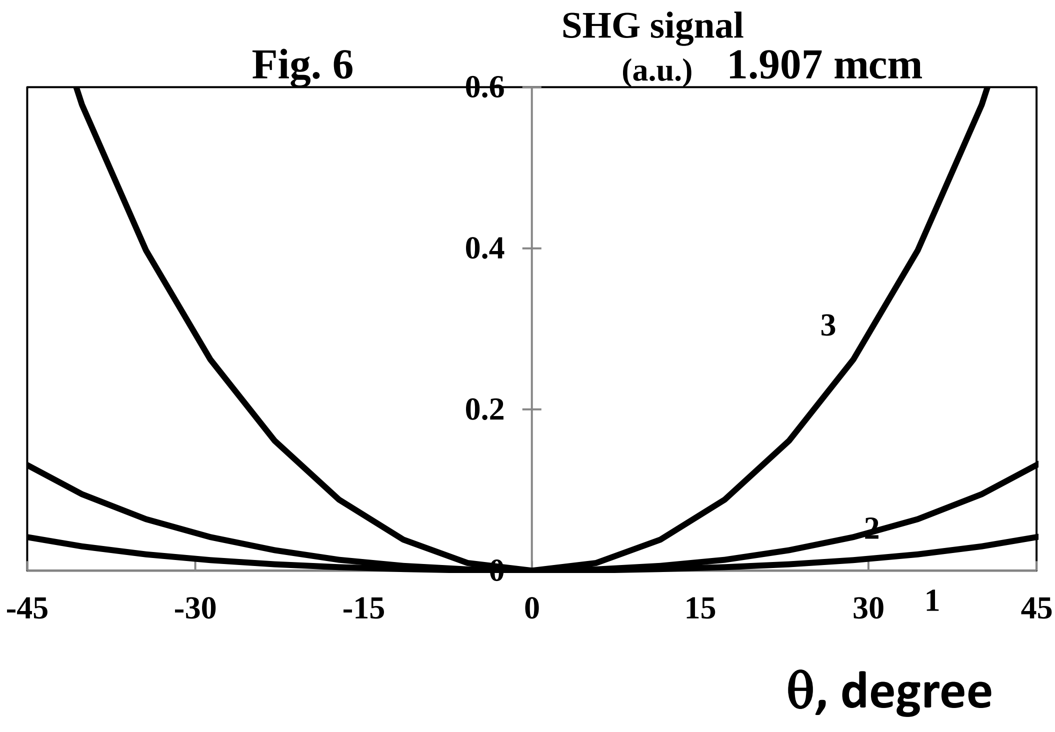

Fig. 6. Dependencies of the analytical expression for intensity of the second harmonic (pump wave λ=1.907 mcm) as a function of the incidence angle θ calculated in the CIA for ZnO films at $s_{\omega} - p_{2\omega}$ (curve 1), $45_{\omega} - p_{2\omega}$ (curve 2) and $p_{\omega} - p_{2\omega}$ (curve 3) polarization configurations at pump intensity of I_{10} = 1 GW/cm^2 and $\delta_2=2\delta_1=0.22*10^4$ cm^{-1}.The thickness of all films is equal to 550 nm [7].

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