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On Increasing the Conversion Efficiency to Second-Harmonic for Undoped and Doped ZnO Nanocomposites

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Abstract: ***Theoretical investigation of frequency transformation for the case of pure ZnO films, ZnO: Ag (1.6 at.%) and ZnO: Cu (4.7 at.%) enables increasing conversion efficiency while taking phase effects into consideration. We will use the constant-intensity approximation of fundamental radiation. This analytical method also allows to approximately measure the maximum of second--harmonic intensity for the case of ZnO films with silver and copper impurities. We will analyze the parameters that restrict the efficiency of frequency conversion. We can calculate the coherent length of crystal converter at given pump intensity. These methods of investigation for second-harmonic generation in pure and doped ZnO films can be useful in the research area of other film.***

Keywords: nanocomposite film; dopant; second-harmonic generation method; constant-intensity approximation. PACS Number(s): 42.65.-k, 42.70.a, 42.70.Mp, 61.46. – w

I. INTRODUCTION

The nanosize structures have affected the compact devices greatly. It requires a new approach which relies on using the thin dielectric and semiconductor films rather than the bulk materials. The technology for making bulk crystal is comparatively more expensive due to the fact that it requires a higher level of technology for the processes of growing and preparing crystals and takes a longer time period to process.[1].

Thin films are based on ZnO films. The reason for using Zinc Oxide films are being used is because of their electrical and optical properties, in particularly, high nonlinear quadratic susceptibility and their better chemical stability and non-toxicity. Recently, the technology of similar structures has added to the interest in using this approach. The reason for this interest is because of the possibility of frequency conversion in these media due to the relevant second order susceptibility (close to 10 pm/V) under the nonlinear tensor. Besides, these crystals are capable of depositing the layers over different kinds of substrates [2-3]. The ZnO films are optimal to be used in solar cells, gas sensors, electronic and optoelectronic devices, acoustic wave and piezoelectric devices [4]. Recent quality improvement in the conductivity of epitaxial ZnO crystals have caused more interest to use this composites in the short wavelength light emitters and transparent electronics [5, 6]. Zinc oxide has the energy gap equal to 3.37 eV at room temperature. This results for their use in order to decrease the influence of the UV region of solar spectrum. These composites are also more appropriate because of high resistance to high energy radiation and this makes them suitable for space application and more stable when subjected to chemical etching resulting in smaller devices [7]. ZnO is a very important II-VI semiconductor composite and by changing its gap energy band could be used in other areas. Adding any metal will shift the band gap which results in establishing new energy levels for semiconductor composites [8, 9]. Properties of ZnO films are derived according to added metals like Ag, Cu, V and others [6, 10-12]. We also know (from [13]) that the doping metals from group I (Ag and Cu) are diffused rapidly in the semiconductor composites. We can see from [6, 10-11] that the diffusion of copper or silver will change the characteristics of its structure and therefore their other physical properties. The concentration of Cu will affect the energy gap, optical absorption and thickness for the ZnO composites [4]. According to experience the Cu doped ZnO films have higher electrical resistance [14-17]. Akio et al. investigated electron trap level of Cu doped ZnO. The resistance for these composites is high and increase with higher Cu concentration [18]. Impurities like Ag or Cu with different concentrations in doped ZnO have been studied by photosensitivity, luminescent properties and electrical stability and are being applied in different applications. ZnO has a lot of application in the field of communication as acoustics wave devices[19]. The electromechanical coefficient, d_{33}, equal to 110 pC/N is being reported to achieved for the V-doped ZnO films and will give us tentative interpretations. In [20] the investigation of second-harmonic generation (SHG) on optical properties of these films under the influence of impurities has been carried out. The theory for SHG has been developed recently and is the popular method to study the nonlinear optical properties of the different composites in SHG. The properties of SHG for diagnosing are based on dependency of nonlinear optical response (which is described by nonlinear polarization $P(2\omega)$) on symmetry of material over the tensorial characteristic of the relation.

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The analysis of the interaction between laser radiation and matter at SHG, specifically the Maker beats, allows us to study the different parameters of nonlinear media and more importantly the component of nonlinear susceptibilities of them. It is known that the intensity of second-harmonic is a function of the direction of the polarization of pump wave.

The same way is being applied to study the X-ray structure analysis of crystallographic axes definition, planes for monocrystals and quality of surface [21]. It should be mentioned that in order to study the micro- and nanostructure we should have the tensor component for nonlinear susceptibility.

This work will carry out the analysis of nonlinear interaction for SHG under the constant- intensity approximation [22] for the case of pure ZnO and doped ZnO nanocomposite materials (ZnO:Ag and ZnO:Cu) while considering the dissipation and phase change of all interacting waves. The effect of impurities on the nonlinear optical propertiers has been investigated. Also the factors that restrict the efficacy of conversion frequency are being studied. This is our new work in the cycle of works that is devoted to the study of nonlinear optical characteristics for the case of zinc oxide films [23].

II. THEORY

The theoretical investigation of ZnO structures are carried out while considering the phase effects at nonlinear wave interaction and under the constant-intensity approximation. In addition to this, the coefficients for Fresnel transmission, refraction phenomena in the film and different values for second-order nonlinear susceptibility for ZnO films, $\chi_{eff}^{(2)}$, [20, 24], walk off effect and different impurities for two different SHC polarization configurations will be taken into considerations which are the result of the experiments [24].

While considering these we will choose parameters to be able to satisfy the conditions for current experiment that regards the ZnO film [24]. For fundamental beam we have chosen the output from Q-switched Nd:YAG laser that is being generated at wavelength of 1064 nm and has a duration of 16 ps for its pulses and the range of repetition lying in 10 Hz. The fundamental intensity relies in the range of 1 to 10 GW/cm^2. The ZnO films that were used had the thickness of 600 to 1200 nm. The polarization of this incoming beam, as said before, was varied from s to p.

Due to possessing tensor component in ZnO structure, the experimental observation of the second-harmonic signal will be p-polarized all the time even though the incoming beam might have s or p polarization [25].

We will have one degree of resolving power for the incident angle.

We also should analyze the doubling of the frequency for steady state using the coupled system of equation while the constant-intensity approximation is being applied. We will take an isotropic, absorbing nonlinear pure ZnO film and ZnO with impurities layered over the substrates [26].

Based on the existing and known reduced equation system under the constant-intensity approximation for the output of film structure ($z = \ell$) and without taking reflections into account we will have the following expression for transmitted second-harmonic intensity, [27]

$$I_2(l) = \gamma_2^2 I_{10}^2 t_{af}^4 t_{fs}^2 t_{sa}^2 \frac{(\sin^2 x + \mathrm{sh}^2 y)}{\rho} \exp\left[-(\delta_2 + 2\delta_1)l\right], \quad (1)$$

where

$$\rho^2 = a^2 + b^2, \quad a = 2\Gamma^2 + \frac{\Delta^2}{4} - (\delta_2 - 2\delta_1)^2/4,$$

$$b = \Delta(\delta_2 - 2\delta_1)/2, \quad \Gamma^2 = \gamma_1\gamma_2 I_{10}, \quad I_j = A_j A_j^*,$$

$$x = \rho^{1/2}\ell\cos(\varphi/2), \quad y = \rho^{1/2}\ell\sin(\varphi/2),$$

$$\varphi = \mathrm{atan}(b/a).$$

Here

$$\gamma_1 = \frac{8\pi^2\chi_{eff}^{(2)}}{\lambda_1 n(\omega_1)}, \quad \gamma_2 = \frac{4\pi^2\chi_{eff}^{(2)}}{\lambda_2 n(\omega_2)}.$$

Denotes stands for the second-order coupling coefficients of the waves at the frequencies ω_1 and $\omega_2 = 2\omega_1$, δ_j $(j = 1, 2)$ are signifies the absorption coefficients, $\chi_{eff}^{(2)}$ is the effective second-order nonlinear susceptibility for ZnO films, $\lambda_{1,2}$ are wavelengths of pump and generated waves, $n(\omega_{1,2})$ show the refractive indices in this zinc oxide films at frequencies ω and 2ω, respectively, and t_{af}, t_{fs}, t_{sa} stand for the Fresnel transmission coefficient for air-film boundary for fundamental beam, the Fresnel transmission coefficients for film-substrate and substrate-air boundaries for harmonic beam, respectively. $\Delta = k_2 - 2k_1$ signifies the phase mismatch between interacting waves and for the case of s - polarized fundamental beam it takes place the next relation for Δ [27, 20]

$$\frac{\Delta^{s-p}}{2}\ell = \frac{2\pi}{\lambda_1}\ell\left[n_{2\omega}(\theta)\cos\theta_{2\omega} - n_{10}\cos\theta_{1\omega}\right], \quad (2)$$

and for p - polarized fundamental beam

$$\frac{\Delta^{s-p}}{2}\ell = \frac{2\pi}{\lambda_1}\ell\left[n_{2\omega}(\theta)\cos\theta_{2\omega} - n_{1\omega}(\theta)\cos\theta_{1\omega}\right], \quad (3)$$

where θ is the incident angle.

For the case of s - polarized fundamental beam the effective second-order nonlinear susceptibility for ZnO films, $\chi_{eff}^{(2)}$, will be equal to

$$\chi_{eff}^{(2)} = \chi_{zxx}^{(2)}\sin\theta_{2\omega}$$

and for p - polarized fundamental beam $\chi_{eff}^{(2)}$ will be equal to:

$$\chi_{eff}^{(2)} = \chi_{zxx}^{(2)}(\cos\theta_{2\omega}\sin 2\theta_{2\omega} + \sin\theta_{2\omega}\cos^2\theta_{\omega}) + \chi_{zzz}(2)\sin\theta_{2\omega}\sin^2\theta_{\omega},$$

where θ_{ω}, $\theta_{2\omega}$ are the fundamental and generated beams refractive angles which are being determined by the law of diffraction.

We can see from (1) that when the absorption coefficient is small the harmonic intensity will be periodic function over the nonlinear interaction length ℓ.

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For the case of phase mismatch that happens when Δ is zero we will have, following (1), the optimum value for nonlinear medium length for which the conversion efficiency will be maximum. This can be calculated as follows [27]

$$\ell_{opt} = \frac{\operatorname{atan}\left(\dfrac{2\Gamma^2 - \dfrac{(\delta_2 - 2\delta_1)^2}{4}}{\delta_1 + \delta_2/2}\right)}{\sqrt{2\Gamma^2 - \dfrac{(\delta_2 - 2\delta_1)^2}{4}}}. \qquad (4)$$

When $\delta_2 = 2\delta_1$ the optimum value of length will be given by: for case $\delta_2 = 2\delta_1$ the optimum value ℓ_{opt} is determined as follows:

$$\ell_{opt} = \frac{\operatorname{atan}\left(\dfrac{2\Gamma^2 + \dfrac{\Delta^2}{4}}{\delta_2}\right)}{2\Gamma^2 + \dfrac{\Delta^2}{4}}, \qquad (5)$$

We can see that the optimum length of crystal is a function of pump intensity, I_{10}, (via its dependency on Γ) and dissipation in medium.

We will have $\ell_{opr} = 0.5\pi/(2\Gamma_1^2 + \Delta^2/4)$ for optimum length when Δ is zero. In case of $\delta_{1,2} = 0$ the optimum length is expressed by $\ell_{opr} = 0.5\pi/(2\Gamma_1^2 + \Delta^2/4)$.

While the constant-field approximation is in the rule ($\gamma_1 = 0$ and $\delta_j = 0$) then we can derive the well-known expression of coherent length $\ell_{opt}^{CFA} = \pi/4 = k_2 - 2k_1$.

For the sake of better investigation of optimum value of the efficiency of frequency conversion in case of doped and undoped ZnO films of laser radiation, we are going to do some analytic calculation of equation (1). The Sellmeier model has been used to calculate the refractive indices of undoped ZnO [1].

III. RESULTS AND DISCUSSION

We are going to approximate the expected transformation power of the second-harmonic according to relation (1). For this purpose we should be aware of thickness, refractive index (at least Sellmeier coefficients) of the ordinary and extraordinary rays and absorption coefficient for the studied samples of films depending over the wavelength in order to solve precisely the analysis and give the recommendations. Currently there are different data, unfortunately, that is a huge variation among studied cases. Optical features of doped ZnO films are still at the stage of studying and definition.

Based on Eq. (1) Fig 1-4 depicts the dynamic process for frequency transformation in second-harmonic for the case of pure ZnO [3, 20] and doped ZnO structure (ZnO:Ag and ZnO:Cu) [6, 10-11]. For the structure the refraction phenomena have also been considered besides all other parameters like different values of dissipation for the fundamental wavelength and harmonic wave frequencies, different concentrations of impurities in ZnO, walk off effect and Fresnel coefficients for transmission for two different configurations of pump wave polarization ($s_\omega - p_{2\omega}$ and $p_\omega - p_{2\omega}$), which are being mentioned in [27]. The case of two polarization configuration is being used in the experiment [20].

Fig 1 depicts the dependency of intensity for second-harmonic $I_2(l)$ over the length of film length. We have six versions of conversion for the case of ZnO films where the only difference among them is the intensity of pump (done by Nd:YAG) that are radiating at wavelength in 1.064 mcm over two different polarization configuration while θ was equal to 35 degrees. We can see from the behavior of curves, that differs from monotonous behavior under the constant-intensity approximation, that there is an optimum value of a crystal length for which the conversion efficiency is the highest. The solid and dotted curves 3 are representing the dependency of $I_2(l)$ for θ =45^0. As we increase the incident angle more (up to 50^0), the efficiency will also be maximum which is in agreement with Figs. 2 and 3. Fig. 1 shows that the coherent length, ℓ_{opt}, (5) that corresponds to having maximum efficiency, is about 460 nm. To make a better comparison we can mention that the experimental value of ZnO is 950 nm for its length when the pump intensity is 4 GW/cm^2 [20]. By decreasing the film thickness of ZnO from 950 nm to the coherent length, 460 nm, (2.065 times) it will be possible to raise the efficiency ~ 2.012 times when θ =35^o (see solid curve 2).

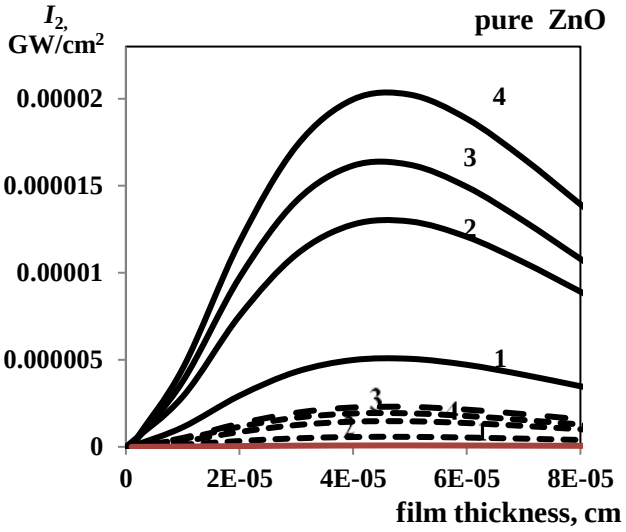

Fig. 1a. Dependences of intensity of second-harmonic for the radiation energy of pump wave (λ=1064 nm), $I_2(l)$, on film thickness, *l*, calculated in the constant-intensity approximation : a) For pure ZnO films at pump intensity of I_{10}= 2.5 GW/cm^2 (dotted and solid curves 1), 4 GW/cm^2 (dotted and solid curves 2 and 4), 5 GW/cm^2 (dotted and solid curves 3) and for the incident angle of θ = 35^o (dotted and solid curves 1, 2 and 4), 45^o(dotted and solid curves 3).

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Fig. 1b shows the analogical relations of frequency conversion, $I_2(l)$, in ZnO:Ag (1.6 at.%) and ZnO:Cu (4.7 at.%) over the length of crystal. We can see from the curves that the incident angle is θ =35^0 for the situation in which pump intensity is 4 GW/cm and the coherent length is 200 nm for ZnO:Cu (4.7 at.%) and it is 340 nm for ZnO:Ag (1.6 at.%) film. This is the result of the values of quadratic susceptibility $\chi_{eff}^{(2)}$ [20], which are 1.43*10^{-5} pm/V and 0.83*10^{-5} pm/V respectively and coherent length (Equations (4) and (5)).

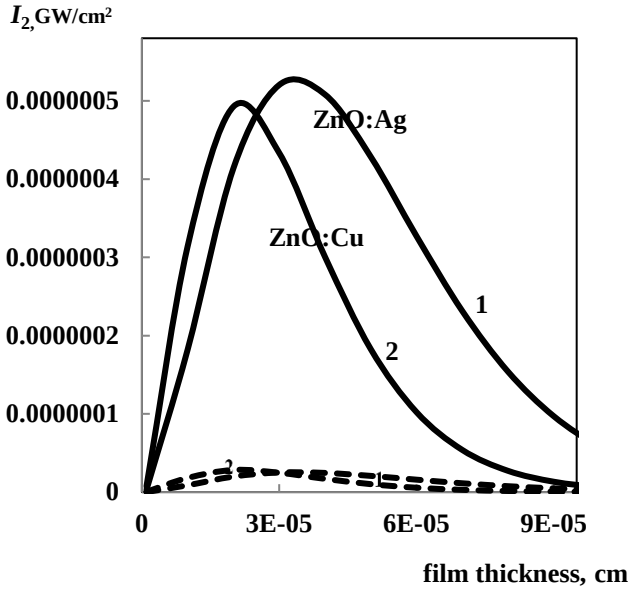

Fig. 1b. Dependences of intensity of second-harmonic for the radiation energy of pump wave (λ=1064 nm), $I_2(l)$, on film thickness, *l*, calculated in the constant-intensity approximation. b For ZnO: Cu (4.7 at.%) films (curves 1) and ZnO: Ag (1.6 at.%) (curves 2) at pump intensity of I_{10}= 4 GW/cm^2 and at the incident angle of θ = 35º.

For nonlinear media, in reality, that possess more nonlinearity the transformation maximum will occur on smaller lengths. On the other hand the authors of [20] had mentioned "…..in the case of Cu doping this decrease of $\chi_{eff}^{(2)}$ is more noticeable due to the difference in sizes of Ag and Cu ions incorporated into ZnO lattice." By having knowledge over the exact parameters of studied samples we can make better conclusion over the role that the size and nonlinearity of Cu and Ag impurities play in transformation efficiency (refractive index, at least Sellmeier coefficients for ordinary and extraordinary rays and absorption coefficients for studied films). The same thing is also possible to explain the violation with pure ZnO film sample.

The results for the analysis of the transformation efficiency is depicted in Fig. 2 ($I_2(\theta)$) for the case of three different values of pump intensity when the film thickness is almost 400 nm. We can see from these dependencies that they have maxima when θ is 50^0 and their minimum occurs when the normal incidence of pump wave lacks the non-central symmetry in this direction. The observed angular dependency is because of the symmetry that crystalline has for the case in which the preferred axis exists (in 002 direction) [1, 25, 28]. To compare better the observed angular dependency in experiment has a maxima close to 40-55^0 [20]. The reason that the optimal angular values are different is because there are no exact values for the parameters of the task used in experiment.

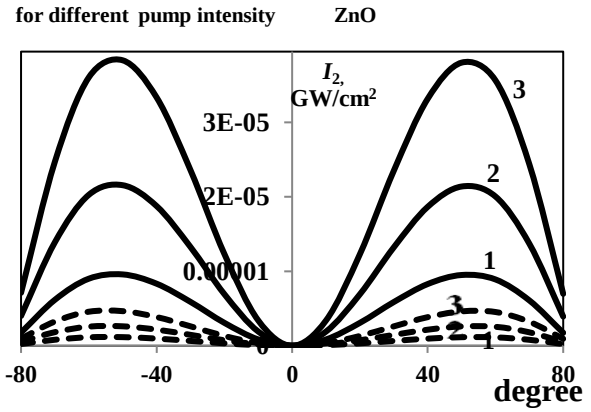

Fig. 2. Dependences of second-harmonic intensity (λ=532 nm) on the incident angle, $I_2(\theta)$, for pure ZnO films calculated in the constant-intensity approximation for the film thickness of *l* =400nm and at the next values of pump intensity of I_{10}= 3 GW/cm^2 (dotted and solid curves 1), 4.5 GW/cm^2 (dotted and solid curves 2), 6 GW/cm^2 (dotted and solid curves 3).

Fig. 3 depicts the result of the analyze for frequency transformation ($I_2(\theta)$) for four different values of film thickness 300 nm, 460 nm, 600 nm and 950 nm [20] for two different polarization states ($s_\omega - p_{2\omega}$ and $p_\omega - p_{2\omega}$) when the pump intensity is 4 GW/cm^2 (pump intensity being applied in the experiment). The maximum intensity is observed at optimum angle of 50^0 (curve 1-4). Thickness of the film is related to the value of the coherent length at 4 GW/cm^2 and is equal to 450-460 nm when the angle varies in the interval of 35^0 to 45^0 (see solid and dotted curves 3). We can see that for the coherent length (equal to 460 nm) the maximum of intensity for second-harmonic will occur.

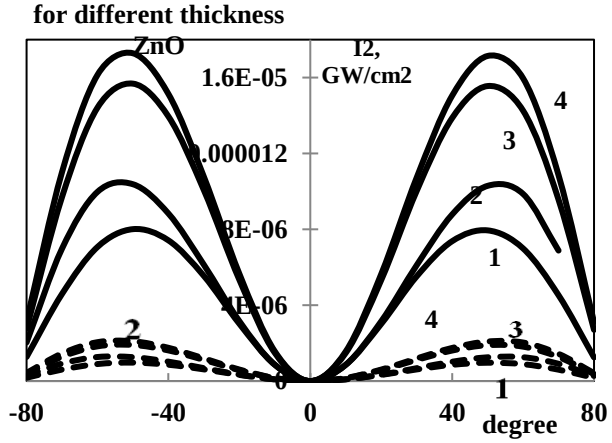

Fig. 3. Dependences of second-harmonic intensity (λ=532 nm) on the incident angle, $I_2(\theta)$, for pure ZnO films calculated in the constant-intensity approximation at pump intensity of I_{10}= 4 GW/cm^2 for the next values of film thickness of *l* = 300 nm (dotted and solid curves 3), 400 nm (dotted and solid curves 1), 600 nm (dotted and solid curves 2), 950 nm (dotted and solid curves 4).

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Fig 4 shows the angular dependency of SHG intensity for (a) ZnO, (b) ZnO:Ag (1.6 at.%) and (c) ZnO:Cu (4.7 at.%). The fundamental beam has s and p polarization and will produce p polarization beam. The solid curves stand for the calculation in the constant-intensity approximation based on relation (1) for these three samples of doped films. The dotted curves (upper for $p_\omega - p_{2\omega}$, lower for $s_\omega - p_{2\omega}$ polarization configurations) show the experimental results which are obtained from [20].

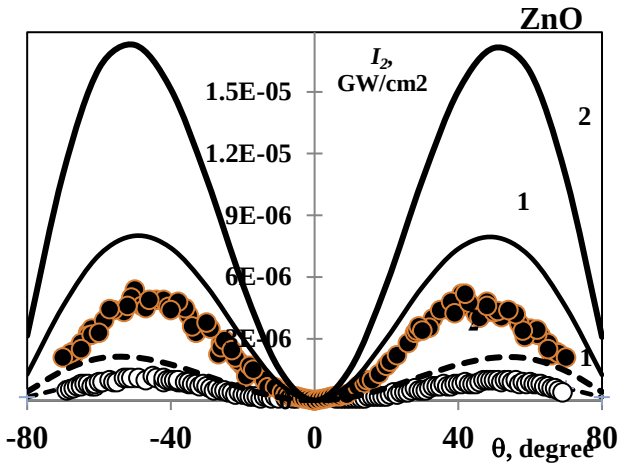

Fig. 4a. Dependences of second-harmonic intensity (λ=532 nm) on the incident angle, $I_2(\theta)$, calculated in the constant-intensity approximation at pump intensity of I_{10}= 4 GW/cm^2. The experimental results, [20], are presented by dots: upper dots for $p_\omega - p_{2\omega}$, lower dots for $s_\omega - p_{2\omega}$ polarization configurations: a) For pure ZnO films at $l = \ell_{opt}$ =460 nm (dotted and solid curves 2) and 950 nm [20] (dotted and solid curves 1).

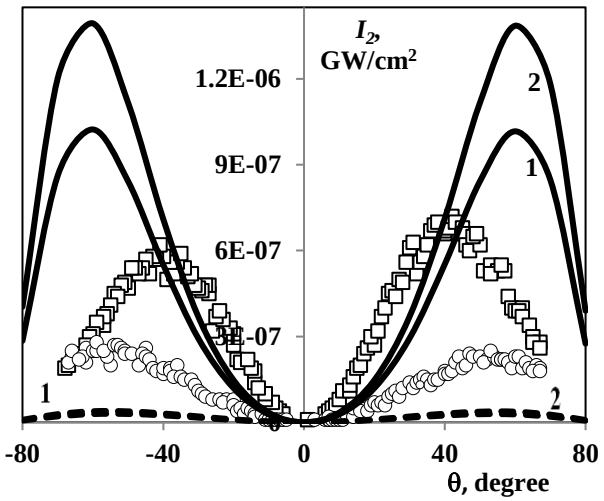

Fig. 4b. Dependences of second-harmonic intensity (λ=532 nm) on the incident angle, $I_2(\theta)$, calculated in the constant-intensity approximation at pump intensity of I_{10}= 4 GW/cm^2. The experimental results, [20], are presented by dots: upper dots for $p_\omega - p_{2\omega}$, lower dots for $s_\omega - p_{2\omega}$ polarization configurations: b) For ZnO:Ag (1.6 at.%) films at $l = \ell_{opt}$ =340 nm (dotted and solid curves 2) and 515 nm [20] (dotted and solid curves 1).

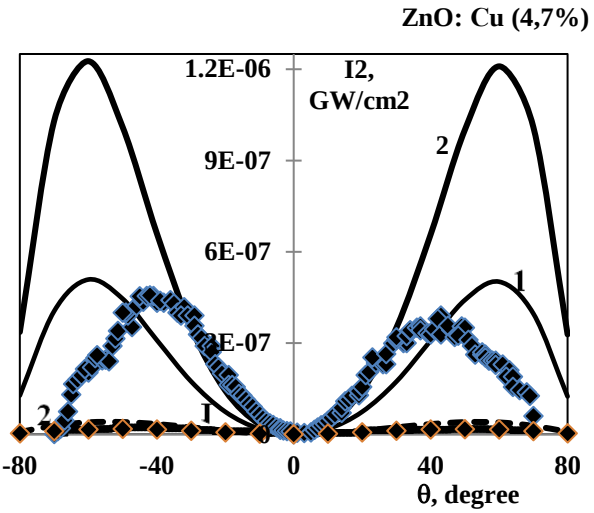

Fig. 4c. Dependences of second-harmonic intensity (λ=532 nm) on the incident angle, $I_2(\theta)$, calculated in the constant-intensity approximation at pump intensity of I_{10}= 4 GW/cm^2. The experimental results, [20], are presented by dots: upper dots for $p_\omega - p_{2\omega}$, lower dots for $s_\omega - p_{2\omega}$ polarization configurations: c) For ZnO:Cu (4.7 at.%) films at $l = \ell_{opt}$ =200 nm (dotted and solid curves 2) and 450 nm [20] (dotted and solid curves 1).

A similar angular dependency will be observed for c-oriented ZnO, ZnO:Ag, and ZnO:Cu films with its maximum located at 50^0-57^0 as usual. We will get the best agreement for the optimal angle between theory and experiments only for the case of pure ZnO film. It is possible to describe this via precise value of refractive indices for ordinary and extraordinary rays, coefficient of extinction and etc for the sample of ZnO film under considerations. For ZnO:Ag (1.6 at.%) the information of the similar case ZnO:Ag (2 at.%) has been gathered [10] and for the case of ZnO:Cu (4.7 at.%) we have chosen the parameters related to the available data of the crystal of ZnO:Cu (2 at.%) and (4-6 at.%) [6, 11]. Because of this the values of optimum angle will vary.

Based on Eq. (1) we see a 30% variation from experiment than theory itself for the chosen parameters for the intensity of second-harmonic. The similar differences may be on different circumstances. This could be because of lack of information over some of required parameters during experiment. For the case of pure ZnO films this can be stated because of lack of exact information of dissipation.

The curves 1 and 2 of Figs. 4 a), b) and c) were calculated for two different couples with different film thicknesses. To compare these curves it can be see that the more transformation efficiency is observed at coherent length for each case.

IV. CONCLUSION

The investigation of frequency transformation in ZnO, ZnO:Ag (1.6 at.%) and ZnO:Cu (4.7 at.%) while considering the phase effects allows us to find solutions for improving the conversion efficiency.

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Namely for the given values of pump intensity we will be able to find the optimal value for the length of the converter crystal which is the coherent length of films. This analytical method also enables us to approximately measure the expected second-harmonic intensity for the ZnO films. The results for this research will be helpful to elaborate the doped ZnO films.

In this work the method for analyzing the second-harmonic generation in pure and doped ZnO films could be used to investigate other doped nanocomposites.

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