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On Influence of Temperature and Doped Concentrations on the Frequency Conversion Efficiency in Erbium-Doped Zinc Oxide Films

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Abstract: The effect of different concentrations of erbium as the doping element with different values of the temperature on the nonlinear optical properties of ZnO films was considered. Analytical analysis was carried out with the consideration of phase effects accompanying the study process. For this purpose, the constant intensity approximation was used. The analytical method also allows us to estimate the effect of impurities on the nonlinear optical properties of films by method of the third harmonic generation and the maximum value of the intensity of the third harmonic in pure and doped by erbium ZnO films. It is possible to calculate for specific values of the pump intensity the coherent length of a nonlinear frequency converter. Analytical analysis indicates that the increase in intensity of the third harmonic in the vicinity of the coherence length values offset by significant losses taking place at such high order, the interaction lengths 600 nm. It is shown that that the introduction of erbium the intensity of the third harmonic is significantly increased. This occurs when the surface of the samples has the lowest degree of roughness when there is a delocalization of the excitons. With increasing impurity of erbium greater than 2% the surface becomes porous, increasing the surface roughness of the obtained samples. The proposed method of increasing the efficiency of the process is to improve the technology of production of films with small losses.

Keywords: Zinc Oxide, Dopant, Harmonic Generation, Constant-Intensity Approximation

1. Introduction

During last years the subject of finding materials with higher and quicker nonlinearity for the case of optical switching and application in sensor protection has been very active. The nonlinear properties of materials are the key feature ruling this area and ZnO, due to its strong nonlinearity e.g. third order susceptibility, is one of the pioneer semiconductors [1]. They are also good candidate for being used at solar cell devices [2], luminescent materials [3] and etc.

The semiconductor wafers are of importance for optoelectronics and alike. Important feature and parameters for ZnO thin film including their optical, electrical and structural features are extensively studied by a number of groups [1-6].

Some of the methods used to study the parameters for materials include second-harmonic generation (SHG) [7-8], third-harmonic generation (THG) [9-13], nonlinear interferometry [17], optical phase conjugation [18], degenerate four-wave mixing (DFWM) [19] and the Z-scan technique [20]. Due to existence of high frequency the THG will be able to show coherent electronic nonlinearity more clearly. This THG metod also allows us to investigate the absorption edge for the material without any undesired change in sample at the presence of strong laser beam.

The big and small sizes of grains happens for substartes during various temperatures which makes more dominant irregularities. Variation of irregularities can affect the optical features of studied ZnO films.

It is being proved that the electronic and luminescence properties of ZnO could be derived using deposition parameters, applied temperature and impurities [2, 7, 8, 10-11, 13]. It should be mentioned though that there are not enough report regarding the systematic study of impurities which affects the nonlinear properties of frequency conversion in particular.

The effect of different impurities (Al, Cu, Sn and Vn) over the microstructure of Zinc Oxide thin films has been extensively studied [6, 7, 14-16]. It shows that as the density of Erbium in ZnO: Er increase the effective nonlinearity, $\chi_{eff}^{(3)}$, gets bigger [11-12] when compared to the case of other impurities like silver or copper [7].

In this paper we are going to investigate the efficiency of frequency conversion while using the constant-intensity approximation [21] for ZnO with and without added Er as impurity for different elaborated temperatures and impurity concentrations while considering dissipation and changes in the waves of interacting waves. The effects coming from impurities are being studied and limiting factors for frequency conversion are discussed.

2. Theory

First we will investigate theoretically the structure of ZnO by having phase effects, refraction phenomena in the films and walk off effect in mind for the case of nonlinear interaction of waves during the generation of direct third-harmonic.

We solve the famous system of reduced equation for the coupled amplitude for direct third harmonic generation while using the constant-intensity approximation and neglecting reflections which will get us the relation for third harmonic intensity. We are going to take Fresnel transmission coefficient for the case of fundamental and harmonic beams for three different boundaries of studied layer structure of samples.

We can get the following relation for the output of this layer structure of samples (air-film-substrate-air) (z=ℓ), [22]

$$I_{3\omega}^{f}(z) = (t_{af}^{\omega})^6 \cdot (t_{fs}^{3\omega})^2 \cdot (t_{sa}^{3\omega})^2 \cdot \gamma_3 \times$$

$$\Gamma^2 I_{\omega 0} \cdot (\gamma_1 \rho_3)^{-1} \cdot e^{-(\delta_3+3\delta_1)z} \cdot (sin^2 \propto_3 + sinh^2 \beta_3), \quad (1)$$

In which

$$\rho_3^2 = a_3^2 + b_3^2; \Gamma_3^2 = \gamma_1 \gamma_3 I_{\omega 0}^2,$$

$$a_3 = 3\Gamma_3^2 + \left[\Delta_f^2 - (\delta_3 - 3\,\delta_1)^2\right]/4,\ b_3 = \Delta_f(\delta_3 - 3\,\delta_1)/2;$$

$$\propto_3 = \sqrt{\rho_3} z \cos {}^{\varphi}/_{2};\ \ \beta_3 = \sqrt{\rho_3} z \sin {}^{\varphi}/_{2};\ \varphi = atan\frac{b_3}{a_3}.$$

Here

$$\gamma_1 = \frac{3\pi^2}{n_1 \lambda_\omega} \chi_{eff}^{(3)}, \gamma_3 = \frac{3\pi^2}{n_3 \lambda_\omega} \chi_{eff}^{(3)},$$

Signifies the third order coupling coefficients of the waves at the frequencies $\omega_{1,3}$ ($\omega_1 = \omega$ and ω_3=3ω), δ_j (j=1, 3) is stand for the absorption coefficients, $\chi_{eff}^{(3)}$ stands for the effective third order nonlinear susceptibility for pure and doped zinc oxide films, λ_ω is the signifies the wavelength of $I_{\omega 0}$ initial value pump wave, $n_{\omega,3\omega}$ are the refractive indices in these films at frequencies ω and 3ω, respectively, and t_{af}, t_{fs}, t_{sa} are 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. $\Delta_f = k_3 - 3k_1$ is the phase mismatch between interacting waves for films. We have to emphasize that the glass substrate has lower affection on THG which allows us to ignore it.

We have done the analytical investigation for the third-harmonic generation to establish the condition of current experiment which include ZnO and due to this fundamental investigation of Q-switched Nd: YAG laser. This laser operates at 1064 nm with pulses lasting 15 ps (it offers 1.62 mJ/pulse); its intensity has been about 2 GW/cm^2. Thin pure and doped ZnO films with thickness of 200 to 270 nm are used.

For the case of phase mismatch for interacting waves Δ_f in film, the next relation occurs

$$\Delta_f\,\ell = \frac{6\pi}{\lambda_1}\ell(n_3^f cos\theta_3^f - n_1^f cos\theta_1^f), \quad (2)$$

where $\theta_{1,3}^f$ stands for the refractive angles of fundamental and third harmonic waves of the pure and doped ZnO films when the frequencies are equal to ω and 3ω (and θ is the incident angle on film for laser beam). The refractive indices for Er-doped ZnO films have been evaluated through transmission coefficient according to [13].

3. Results and Discussion

Different effects over the nonlinear optical properties of ZnO have been studied due to different amount of concentration of impurities. The process for the dynamic nonlinear transformation is being depicted in Figs. 1-4 which follows from Eq. (1) and it is a result for the case of pure and doped ZnO structures.

As impurities a rare earth element, Erbium, has been considered under the different atomic concentration of 2%, 3%, 5% and 7% for specific set of elaborated temperatures. In addition, all remaining parameters from the amount of dissipation of the pumped and harmonic wave frequencies to variation of density of impurities for ZnO, walk off effect and Fresnel coefficient of pumped and harmonic waves are being counted; there are also refraction effects which will play some role in our research.

First of all we will calculate the expression (1) analytically in order to find the optimal value for the efficiency of frequency conversion for the case of Laser radiation to third order for both pure ZnO and ZnO with added impurities (in this case Erbium).

We first have to know the thickness, refractive index and absorption coefficients (of the waves at the frequencies $\omega_{1,3}$) to be able to study those films [1, 11-13] and their relation to the wavelengths. By doing this we will have the ability of more precise analysis and prediction of results. But now due to existence of different values results will not be clearly defined. For example for ZnO structures that have impurities embedded to them there are not exact set of information which cause uncertainty while their study. This information is already under the study.

We can vary the length of the nonlinear interaction of the film simply by rotating the sample. The thickness for the film will vary in the interval of 200-270 nm during the experiment which brings us the possibility of variation of the incidence angle θ. Samples of added Erbium (5%) are elaborated also under the different values of temperatures (400°C, 450°C and 500°C) in experiment.

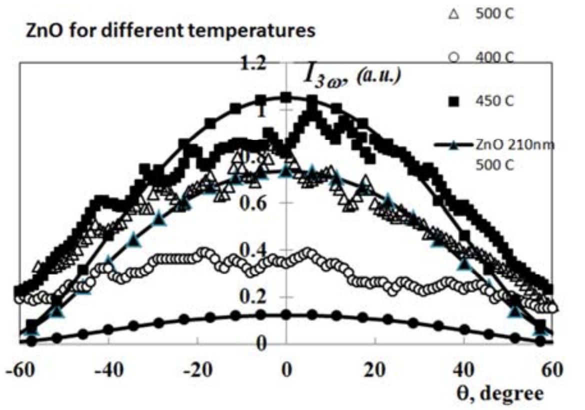

Fig. 1a. *Dependences of intensity of third harmonic $I_{3\omega}$ for the radiation energy of pump wave (λ=1064 nm) on the angle θ, calculated in the constant-intensity approximation for pure ZnO films at pump intensity of $I_{\omega 0}$ =2 GW/cm^2; elaborated temperatures 400°C (curve with black circles dots), 450°C (curve with small black squares) and 500°C (curve with triangles); the film thickness of l=270 nm (curve with black circles dots), 220 nm (curve with small black squares) and 210 nm (curve with triangles). The dotted curves with circles dots, small black squares and triangles come from the experimental results based on the information obtained from reference [13].*

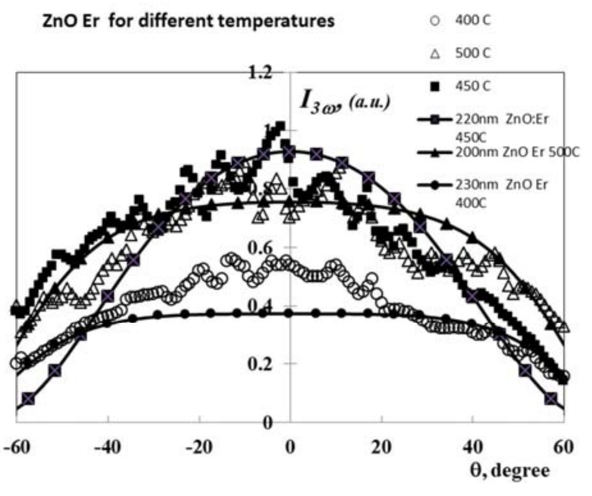

Fig. 1b. *Dependences of intensity of third harmonic $I_{3\omega}$ for the radiation energy of pump wave (λ=1064 nm) on the angle θ, calculated in the constant-intensity approximation for ZnO: Er (5%) films at pump intensity of $I_{\omega 0}$ =2 GW/cm^2; elaborated temperatures 400°C (curve with black circles dots), 450°C (curve with small black squares) and 500°C (curve with triangles); the film thickness of l=230 nm (curve with black circles dots), 220 nm (curve with small black squares) and 200 nm (curve with triangles). The dotted curves with circles dots, small black squares and triangles come from the experimental results based on the information obtained from reference [13].*

Fig. 1a shows the dependency of intensity over the third harmonic based on (1) as a function of the length of the ZnO film (i.e. rotating the angle θ). The dotted curves come from the experimental results based on the information obtained from reference [13] but circle dots, here, signify elaborated temperature of pure ZnO when it is equal to 400°C, small black squares are representing the case with 450°C and triangles show for 500°C. Theoretical results are in good agreement with experimental ones. Under the chosen parameters we can expect to see 20 to 30% variation for the maximum of intensity for third harmonic between theory and experiment. These variations might happen on several occasions and the main reason is the lack of knowledge over the required information regarding the experiment.

Fig. 1b also shows the analogous relation of frequency conversion derived from (1) in ZnO: Er (5%) over the length of the film (that is rotating angle θ): circle dots correspond to 400°C, small black squares are representing the case with 450°C and triangles again show for 500°C. We will have 3 versions of conversion for ZnO and ZnO: Er (5%) films for three different elaborated temperatures (done in experiment [13]) and this factor (temperature) is the key factor of differentiation between them. It can be seen from the curves, which is different than monotonous one, that the maxima of conversion efficiency occur when the temperature equals to 450°C. The authors of Ref. 13 state that the existence of this optimal elaborated temperature can be explained by the grain structures of film sample. In this case a smooth and homogeneous surface will be obtained experimentally by the same authors. By comparing the curves 3 in both figures we can see from [13] that the introduction of erbium significantly increases the nonlinear susceptibility of the film due to the generation of additional polarizability centers, which themselves depend on the degree of roughness, that is, the quality of the optical surface. The analysis showed that by increasing the concentration of impurity, Erbium, from 0 to 5% the third harmonic signal will be intensified for about 161.83 times.

Fig. 2a and 2b picture the dependency of intensity of the third harmonic over the ZnO and ZnO (5%) film length, respectively. Among three versions of conversion available for ZnO films and it is the intensity of $I_{\omega 0}$ by Nd: YAG which makes the difference between them. By observing the behavior of the curves we will be able to see the maxima of conversion efficiency which will be the highest for intensity of 2.4 GW/cm^2 ((highest intensity has been achieved in the experiment [13] is equal to $I_{\omega 0}$ =2 GW/cm^2). We can see clearly from the Fig 2a and 2b that the efficiency will be improved about 1.73 times for the cases of ZnO and ZnO: Er (5%) films when we increase the pump intensity to 2.4 GW/cm^2 (curves 3) (experimental value has been 2 GW/cm^2 - curves 2). Further increase in the pump intensity will allow us to go even beyond and obtain higher conversion efficiency. By analyzing more it is seen that the optimal value for the pump intensity is very close to the values higher than experimental value of 2 GW/cm^2.

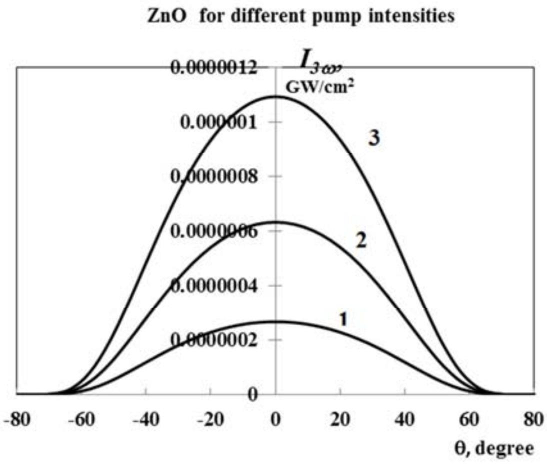

Fig. 2a. *Dependences of intensity of the third harmonic $I_{3\omega}$ on the angle θ for ZnO film at the film thickness of l=220 nm and elaborated temperature 450°C for the pump intensity of $I_{\omega 0}$ =1.5 GW/cm^2 (curve 1), 2 GW/cm^2 (curve 2), 2.4 GW/cm^2 (curve 3).*

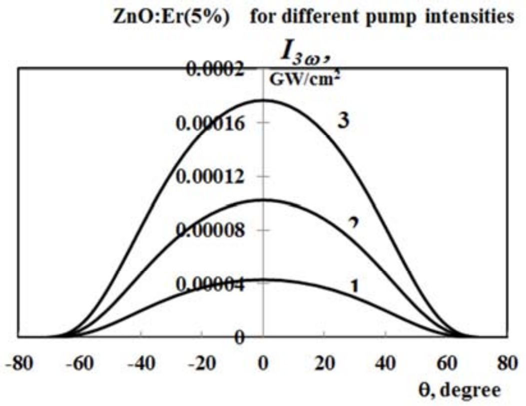

Fig. 2b. *Dependences of intensity of the third harmonic $I_{3\omega}$ on the angle θ for ZnO: Er (5%) film at the film thickness of l=220 nm and elaborated temperature 450°C for the pump intensity of $I_{\omega 0}$ =1.5 GW/cm^2 (curve 1), 2 GW/cm^2 (curve 2), 2.4 GW/cm^2 (curve 3).*

In order to calculate the optimal length appropriate for achieving maximal third harmonic amplitude we will use the formula (for $\delta_3 = 3\delta_1$) which is obtained from (1)

$$\ell_{coh} = \lambda_1^{-1}\text{atan}(\lambda_1/\delta_3), \text{ where } \lambda_1^2=3\Gamma_3^2 + \Delta^2/4.$$

According to this relation, at the pump intensity of 2 GW/cm^2 and elaborated temperature of 450°C the value of ℓ_{coh} =650 nm for the case of ZnO and will be 609.67 nm for Zno: Er (5 %). As the thickness of pure and doped ZnO increase beyond the experimental length of the sample (220 nm), the efficiency will also increase. Analytical studies show though that increasing the intensity of harmonic for the coherent length will be compensated by considerable dissipation in samples over the large length of interaction (~600 nm). One way to increase the harmonic intensity is to obtain films with lower absorption.

Figs. 3a (for ZnO) and 3b (ZnO: Er) draw the analyze for frequency transformation dependency $I_{3\omega}$ when the elaborated temperature is 450°C and pump intensity of 2 GW/cm^2 (the same intensity employed in experiment [12, 13]) for two different values of concentrations of Erbium atoms (0% and 5%) as a function of the thicknesses of films. The different character of dependency is obvious. By analyzing the transformation efficiency in ZnO films we will observe that the dependencies have one maximum at the film thickness is about 120-240 nm (see curves 1-4) and two maxima at the film thickness is 60 nm (curve 5). For the latter case we see the transformation of energy occurring in lateral from the central maximum (curve 5). We get the maximum intensity for the film with the thicknesses when it is equal to 60 nm (it should be mentioned that the thickness of pure and doped ZnO lies in the range of 180-240 nm in experiment). By maximizing the thickness of the pure ZnO film to values higher than 60 nm (120 nm, 160 nm, 240 nm and so on), our calculations give a decrease in the efficiency at the corresponding coherent length is 600nm.

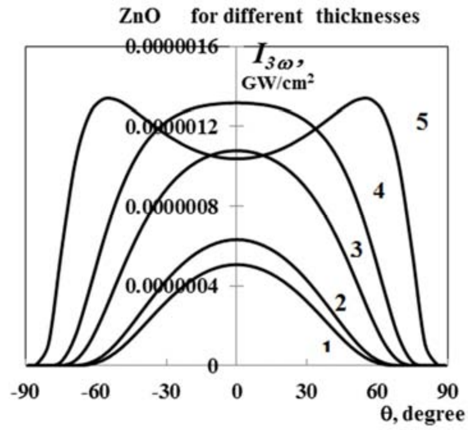

Fig. 3a. *Dependences of intensity of third harmonic $I_{3\omega}$ for the radiation energy of pump wave (λ=1064 nm) on the angle θ, calculated in the constant-intensity approximation for pure ZnO films at pump intensity of $I_{\omega 0}$ =2 GW/cm^2, elaborated temperatures 450°C at the film thickness of l=240 nm (curve 1), 220 nm (curve 2), 160 nm (curve 3), 120 nm (curve 4) and 60 nm (curve 5).*

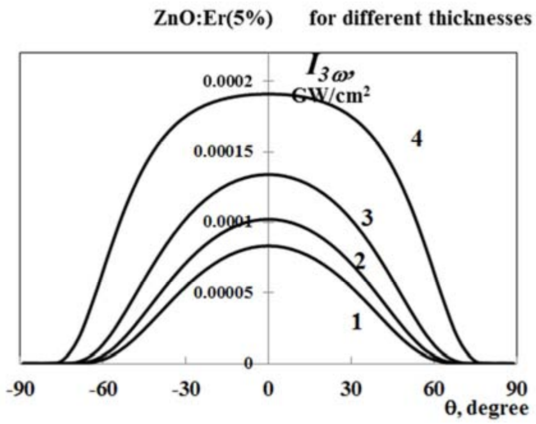

Fig. 3b. *Dependences of intensity of third harmonic $I_{3\omega}$ for the radiation energy of pump wave (λ=1064 nm) on the angle θ, calculated in the constant-intensity approximation for ZnO: Er (5%) films at pump intensity of $I_{\omega 0}$ =2 GW/cm^2, elaborated temperatures 450°C at the film thickness of l=240 nm (curve 1), 220 nm (curve 2), 190 nm (curve 3) and 130 nm (curve 4).*

The analytical studies done by us show that the increase in the intensity of the third harmonic around the coherent length will be compensated via high experimental dissipation over this large length of the interaction (~600 nm). By analogous studies for the case of ZnO films with impurities we will get the following result (see Fig. 3b). By comparing four curves for different thicknesses (130 nm, 190 nm, 220 nm and 240 nm) we will see minimization in the thicknesses from 240 nm to 130 nm cause increasing the efficiency for about 2.3 times for ZnO: Er 5%) film. As it is mentioned before the behavior in ZnO can be explained by considerable dissipation in samples over the large length of interaction.

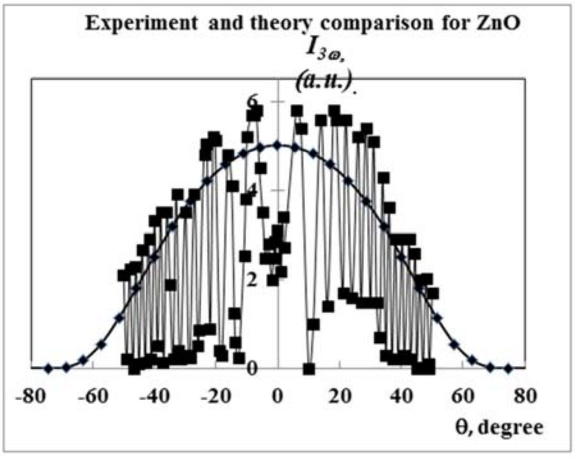

Fig. 4a. *Dependence of intensity of third harmonic $I_{3\omega}$ for the radiation energy of pump wave (λ=1064 nm) on the angle θ, calculated in the constant-intensity approximation for pure ZnO films at pump intensity of $I_{\omega 0}$ =2 GW/cm^2, elaborated temperatures 450°C at the film thickness of l=200 nm. Here there are dependence with oscillations corresponding to results of experiment [12].*

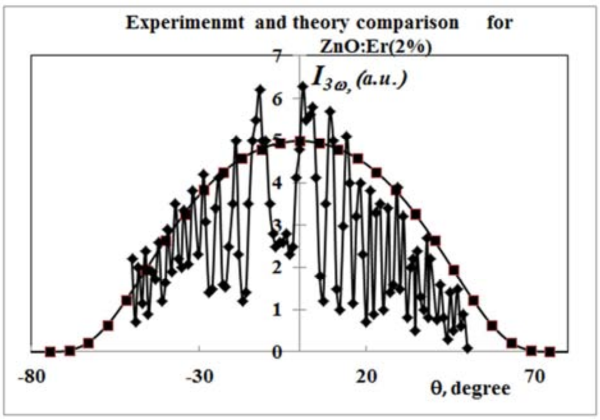

Fig. 4b. *Dependence of intensity of third harmonic $I_{3\omega}$ for the radiation energy of pump wave (λ=1064 nm) on the angle θ, calculated in the constant-intensity approximation for ZnO: Er (2%) films at pump intensity of $I_{\omega 0}$ =2 GW/cm^2, elaborated temperatures 450°C at the film thickness of l=180 nm. Here there are dependence with oscillations corresponding to results of experiment [12].*

Figs. 4a (for ZnO), 4b (ZnO: Er 2%) and 4c (for ZnO: Er: 3%, 5% and 7%) depicts the analysis of frequency transformation based on relation (1) when the pump intensity equals to 2 GW/cm^2 for four different values of Erbium concentration (solid curves). The dependences of the oscillations corresponding to the results of experiments [12] are illustrated in this figure. Contrary to the case of silver or copper in Zinc Oxide addition of erbium cause the increase in cubic nonlinearity of the nanocomposite. Strong third harmonic signal is achieved for the film with good quality of crystals for concentration of 2%. For samples with different Er concentration experimentally observed film quality deterioration.

Apparently, this is due to the fact that with 2% erbium doped surface of film is homogeneous, excitons are not localized at the interface and the main reason for the growth of nonlinear optical response is the excitonic resonance. With increasing impurity the surface becomes porous, the main role is played by surface excitons. They are localized at the interface, which leads to a decrease in the susceptibility of the dielectric film, and consequently to reduce the harmonic signal.

Lamrani and others [12], referring to Castaneda et al [23], explained the absence of the Maker beats in the case of 3-7% impurity concentration of erbium as a consequence of the small value of the film thickness as compared with the corresponding coherence length for the occasion. But in the cases of pure ZNO and doped zinc oxide with Er (2%) the length of the films in the experiment three times less (200 nm and 180 nm) than the corresponding value of the coherence lengths, which, according to (1), equal to 650 nm (0%) and 609.608 nm (2%), but Maker beats, according to Lamrani et al, are observed.

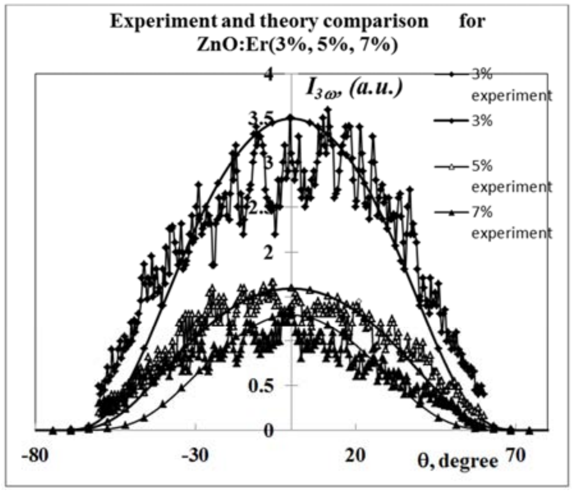

Fig. 4c. *Dependence of intensity of third harmonic $I_{3\omega}$ for the radiation energy of pump wave (λ=1064 nm) on the angle θ, calculated in the constant-intensity approximation for ZnO: Er (3%) at film thickness of l=240 nm, ZnO: Er 5% at film thickness of l=220 nm and ZnO: Er 7% at film thickness of l=210 nm films at pump intensity of $I_{\omega 0}$ =2 GW/cm^2, elaborated temperatures 450°C at the film thickness of l=240 nm. Here there are dependences with oscillations corresponding to results of experiment [12].*

So we can come to conclusion, mentioned earlier by authors of experiment [12, 13] that good homogenous quality of surface films is important besides the high quality obtaining crystallinity. Furthermore, in order to achieve maximal efficiency of frequency transformation it is very important to employ samples with lower coefficient of absorption.

4. Conclusion

The effect of different concentration of impurities over the nonlinear optical properties of ZnO has been investigated. By doing theoretical studies of frequency conversion for ZnO and ZnO: Er and also taking phase effect into account, we will be able to improve the conversion efficiency. We can calculate the optimum value of length of the crystal converter (that is coherent length). The analytical method also allows us to approximately measure the expected intensity at THG for ZnO films with impurities. Studying THG under the constant intensity approximation proves the fact that third harmonic intensity for Er-doped ZnO films will be bigger in magnitude compared to the case of ZnO film or Ag and Cu doped ZnO films cases. This fact has been mentioned experimentally [12, 13]. The samples that contain lower erbium concentration demonstrate a uniform surface covered with grains and intense third harmonic signal. This result depends on the concentration of impurities of Er and used elaborated temperature.

We conclude that the good homogenous quality of a surface of film is an important factor beside the quality of crystal samples. Furthermore, in order to get better efficiency of transformation, one must use samples with thickness of order of corresponding coherent length. However analytical studies show that as the intensity of harmonic increase will be compensated by dissipations coming from higher length of interaction(~600 nm) One way to increase the harmonic intensity is to obtain films that have lower amount of absorption.

The results of the research will be of importance in frequency conversion over the ZnO films with other embedded impurities.

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