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Phase Effects at Second-Harmonic Generation in ZnO/PMMA Nanocomposite Films

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Abstract: In this paper analysis of constant-intensity approximation of nonlinear interaction of the second-harmonic generation in ZnO/PMMA nanocomposite films for different concentrations of ZnO was carried out with regard to the losses and phase changes of all the interacting waves. The investigated samples were manufactured on the basis of ZnO nanoparticles embedded into polyvinylchloride polymeric matrix (PMMA) by the method of electrochemical deposition. The main goal of work is the exploration of the ZnO morphology and parameters of the second order susceptibilities. It is verified that the surface effects in ZnO/PMMA structures will give a larger contribution than the volume effects. The factors restricting the efficiency of the process of frequency conversion have been analyzed.

Key words: Nanocomposite film, polyvinylchloride, second-harmonic generation, constant-intensity approximation, frequency conversion.

1. Introduction

The modern development of the microelectronics, quantum electronics and nonlinear optics requires development of new perspective nonlinear materials. As is known, it is possible to create the new sources of coherent optical radiation by the methods of nonlinear optics using crystals, which is distinguished among other transparent crystals in this region of spectrum, for its extremely high nonlinearity.

Tunable parametric sources of coherent radiation permit, in combination with frequencies mixing effects, can extend considerably, the region of tuning for wavelengths of laser radiation. Thus, for instance, by using summary frequency generation, receiving tunable laser radiation in short-wave range of spectrum is possible right up to VUV radiation. Harmonic generation is interesting for solving a number of the applied issues in UV and the EUV (extreme UV) regions of spectrum, and in obtaining the photons of high energy.

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The current trend to miniaturization of the devices is accompanied by technological elaboration of nanosize structures. It promoted development of the new approach, namely use of thin dielectric and semiconductor films instead of the bulk materials. It should be noted that the technology of the bulk crystal synthesis is relatively expansive since it requires highly developed technology for crystal growth methods and long processing time. Exploration of semiconductors and dielectrics by harmonic generation method yields information on morphology and structure of these compounds [1].

The thin films of ZnO (zinc oxide) are of interest for devices of nanoscale optoelectronics. Choosing zinc oxide films are because of some considerations. The first reason is relevant second order susceptibility (up to 10 pm/V) [2]. Furthermore these crystals have excellent optical and photomechanical properties [1], and ability to deposit the layers on different kinds of substrates [3]. A series of works for instance Refs. [4-6] are dedicated to the theory of nonlinear interaction in the thin films of zinc oxide, in particular, at SHG (second-harmonic generation). In these works the authors make an analysis of frequency conversion, in particular, in the CFA (constant-field approximation) [7, 8].

In CFA, the coherent length of nonlinear medium depends absolutely on mismatch of wave vectors. The amplitude and phase of the basic radiation are constant. Such simplification is valid only on the initial stage of interaction when the effect of the excited harmonic wave on basic radiation wave could be ignored, i.e. the pump depletion does not exist. As a result, a number of important qualitative features of nonlinear process will be lost. For analysis of the nonlinear process, it is possible to use the direct numerical account of reduced equations. However, the development of analytical method will enable us to obtain the concrete analytical expressions and determine the optimum parameters of a task for the purpose of obtaining the maximum efficiency of conversion.

Simultaneous calculation of phase change and losses of interacting waves is possible to make in the CIA (constant-intensity approximation) [9, 10] with regard to the reverse reaction of the excited wave on pump wave. Besides, in this approximation it is found that coherent length depends on such parameters of a task as basic radiation intensity and losses in the medium in addition to mismatch of interacting waves.

In this work there are carried out the analysis of CIA of nonlinear interaction of SHG in ZnO/PMMA nanocomposite films for different concentrations of ZnO with regard to the losses and phase changes of all the interacting waves. It is verified that the surface effects in ZnO/PMMA structures will give a larger contribution than the volume effects. The factors restricting the efficiency of the process of frequency conversion have been analyzed.

2. Experiment

PVC (Polyvinylchloride) gel (dissolved in tetrahydrofuran) with thickness of 80 nm was deposited onto the surface of glass/SnO_2 substrates by centrifugation. After drying under vacuum at room temperature, the substrates were used as a cathode for deposition of ZnO. The electrochemical deposition has been performed with three electrode configurations: graphite as anode, Ag/$AgCl_3$ electrode as reference electrode and glass/SnO_2/PVC as cathode. Total area of working electrodes (cathode) was 1 × 1 cm^2. At electrodeposition we used aqueous solution of $Zn(NO_3)_2$. The electrodeposition was carried out potentiostaticallyat -0.9, -1.2 V, -1.28 V and -1.35 V vs. Ag/AgCl for 1-2 hours.

3. Theory

Let’s analyze the process of redoubling the frequency of laser radiation (at frequency ω_1) in the noncentrosymmetric ZnO structures in case of first type of the scalar phase matching. At nonlinear conversion theoretical analysis of waves interaction is made with the use of the known system of the reduced equations, describing SHG (at frequency ω_2) with account for linearly losses in a structure [7, 8].

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

where $A_{1,2}$ stand for the complex amplitudes of pump wave and its second-harmonic at frequencies $\omega_{1,2}$ ($\omega_2 = 2\omega_1$), k_1 , k_2 are values of wave vectors of pump and second-harmonic, respectively, $\Delta = k_2 - 2k_1$ is phase mismatch, $\gamma_{1,2}$ signify nonlinear coupling coefficients and $\delta_{1,2}$ are coefficients of absorption for waves at frequencies $\omega_{1,2}$, respectively.

We assume that the ω_2 field is not present at the input, only the amplitude A_1 of the input ω_1 field must be taken nonzero, so that boundary conditions will become

$$A_1(z=0) = t_{af}^{1s} \cdot A_{10} \exp(i\varphi_{10}), \quad A_2(z=0) = 0 \quad (2)$$

where, $z=0$ corresponds to the input of crystal, t_{af}^{1s} is the Fresnel transmission coefficient for the boundary of air-film for fundamental beam (1 s) [4] and φ_{10} is an initial phase of pump wave at the entry of the medium.

We next find the solution that satisfies the appropriate boundary conditions. The Eq. (1) in the CIA of fundamental radiation ($I_1(z) = I_1(z=0) = I_{10}$) for the boundary conditions (2) possess the next solution for complex amplitude of second-harmonic at the output of the structure ($z = \ell$) [9, 10]

$$A_2(\ell) = -i\gamma_2 \cdot t_{fs}^{2p} \cdot t_{sa}^{2p} \cdot \left(t_{af}^{1s}\right)^2 A_{10}^2 \ell \,\mathrm{sinc}\lambda\ell \exp\left[2i\varphi_{10} - \left(\delta_2 + 2\delta_1 - i\Delta\right)\ell/2\right], \quad (3)$$

where, we have introduced the quantities

$$\lambda^2 = 2\Gamma^2 - \left(\delta_2 - 2\delta_1 + i\Delta\right)^2/4,$$

$$\Gamma^2 = \gamma_1\gamma_2\left(t_{af}^{1s}\right)^2 I_{10},$$

$$\mathrm{sinc}x = \sin x/x, \qquad I_j = A_j A_j^*,$$

$$\gamma_1 = \frac{8\pi^2\chi_{1,eff}^{(2)}}{\lambda_1 n_\omega}, \quad \gamma_2 = \frac{4\pi^2\chi_{2,eff}^{(2)}}{\lambda_2 n_{2\omega}}.$$

Here $\chi_{1,2eff}$, $\lambda_{1,2}$ and $n_{\omega,2\omega}$ are effective quadratic nonlinear coefficients, wavelengths and refractive indexes in this nanocomposite films at frequencies $\omega_{1,2}$, respectively, and t_{fs}^{2p} , t_{sa}^{2p} are the Fresnel transmission coefficients for film-substrate-air system for second-harmonic beam (2p) [4].

As it follows Eq. (3), the basic and harmonic field's energy interchange takes place periodically and as a result, spatial variations of the second-harmonic field is observed. This time, the minimums of harmonic intensity beating, as an analysis shows in CIA, depend on nonlinear susceptibilities of the crystal. This fact permits to define the nonlinear susceptibilities of substances by a simple way more precise than in the CFA [11].

From Eq. (3) the optimum length ($\ell_{opt} = \lambda^{-1}\arctan(\lambda/\delta_2)$, $\lambda^2 = 2\Gamma^2 + \Delta^2/4$) of structure-converter may be received. At this length for the process of frequency conversion a harmonic signal will be at maximum. According to the theoretical analysis made in CIA in contrast to the results of the CFA, an optimum length of crystal, i.e. coherent length of nonlinear medium depends on pump intensity I_{10} and losses in a medium [9-10]. In case of $\delta_{1,2}$ =0 the optimum length is expressed by $\ell_{opt}^{'} = 0.5\pi/(2\Gamma^2 + \Delta^2/4)$.

For γ_1 =0 and δ_j =0 from Eq. (3) we have an expression for conversion efficiency in the constant-field approximation.

The transmitted SH intensity can be defined from Eq. (3). The efficiency $\eta_2(\ell)$ for conversion of power from the ω1 wave to the ω2 wave can be defined by $\eta_2(\ell) = I_2(\ell)/I_{10}$ and by neglecting reflections, we see that for the values given above, conversion efficiency is $(\delta_2 = 2\delta_1)$

$$\eta_2(\ell) = \gamma_2^2 I_{10}\ell^2\left(t_{af}^{1s}\right)^2\left(t_{fs}^{2p}\right)^2\left(t_{sa}^{2p}\right)^2 \mathrm{sinc}^2\lambda'\ell\exp\left(-2\delta_2\ell\right) \quad (4)$$

where

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

t_{af}^{1s} is the Fresnel transmission coefficient (air-film system) for second-harmonic (2p) beam [4].

To study the ways of increasing frequency conversion efficiency in ZnO structure of laser radiation, we will make the numerous calculation of the analytical expression for conversion efficiency of Eq. (4) received in CIA.

Under considered case, the nonlinear material is a crystalline particle inside the polymeric film. For this case, in Ref. [4], the notion of equivalent thickness of nonlinear medium inside PMMA is being used. Here, $d_{ZnO}^{equivalent}$ shows the thickness which could have ZnO nanocrystals without PMMA as host

$$d_{ZnO}^{equivalent} = \frac{\%wt_{ZnO}}{100 - \%wt_{ZnO}}\frac{\rho_{PMMA}}{\rho_{ZnO}}\ell_{PMMA+ZnO} \quad (5)$$

where, ρ_{PMMA} , ρ_{ZnO} are densities of PMMA and ZnO, respectively, $\%wt_{ZnO}$ is the weight concentration of ZnO nanocrystals inside a polymeric matrix and $\ell_{PMMA+ZnO}$ is the thickness of ZnO/PMMA nanocomposite film.

In Eq. (4) instead of ordinary thickness we used introduced equivalent thickness, according to Ref. [4]. As a result, Eq. (4) can be written as

$$\eta_2(\ell_{eff}) = \gamma_2^2 I_{10} \ell_{eff}^2 \left(t_{af}^{1s}\right)^2 \left(t_{fs}^{2p}\right)^2 \left(t_{sa}^{2p}\right)^2 \operatorname{sinc}^2 \lambda' \ell_{eff} \exp\left(-2\delta_2 \ell_{eff}\right) \quad (6)$$

where

$$\ell_{eff} = d_{ZnO}^{equivalent} / \cos\theta,$$

$$\frac{\Delta'}{2} = \frac{2\pi}{\lambda_1} \ell \left(n_{2\omega} \cos\theta_{2\omega} - n_\omega \cos\theta_\omega\right) [12].$$

Here θ is the incidence angle of laser beam, $\theta_\omega, \theta_{2\omega}$ are the refractive angles for fundamental and second-harmonic waves, $n_{2\omega}, n_\omega$ are the refractive indices of the nonlinear material at frequencies 2ω and ω. The refractive indices of ZnO have been evaluated according to Sellmeier model [13].

4. Results and Discussion

In Figs. 1-7, the dynamic process of frequency conversion is shown to the second-harmonic in ZnO/PMMA structures. The dependencies are spent taking into account due to the analysis in CIA phase effects at nonlinear wave interaction, the refraction phenomena in the structure, different values of losses at fundamental wavelength and harmonic wave frequencies and different concentration ZnO in structures and Fresnel transmission coefficients.

The parameters of the task are chosen according to the conditions of existing experiments for the given ZnO structure [1, 4, 14-16]. As a fundamental beam, there are considered the output beam of a Q-switched Nd doped yttrium aluminum garnet laser generating at λ = 1,064 nm with 16 ps pulse duration and 10 Hz repetition rate. The fundamental beam has the s-polarization. The absolute value of the effective quadratic susceptibility for this nanocomposite films is experimentally measured in the work [4]. In bulk nonlinear materials, increasing of ZnO concentration leads to the growth of a quadratic susceptibility. In case of the nanocomposite films the opposite effect takes place. The surface effects in the films are dominant when compared to volume effects because of the size of the characteristic materials which is being reduced to nanoscale level [4].

At the ZnO concentration of 5% wt. and less, these values of the second order susceptibility $\chi_{1,2eff}$ lie in the range from 4 to 6 pm/V and are higher than analogical value for ZnO bulk which is about 2.5 pm/V. The thickness of the investigated samples is changed in the diapason (1 ÷ 14) mcm [4, 6]. Experimental value of pump intensity was changed in the range (5 ÷ 23) GW/cm^2 [4]. In work [14], the value for the quadratic susceptibility of ZnO/PMMA film has been obtained and is equal up to 30 pm/V, in Ref. [15] the maximal value of the second order susceptibility was found to be approximately equal to 83 pm/V.

In Refs. [13, 16] Sellmeier's coefficients are experimentally established for ZnO. Using these values of coefficients, we will calculate the ordinary and extraordinary refractive indices in case of SHG.

Because of lack of experimentally measured value of Sellmeier's coefficients for ZnO/PMMA at different weight concentrations (of ZnO nanocrystalls inside a polymeric matrix), the same order of coefficients was supposed for the considered concentrations.

The numerical calculations of the analytical Eq. (6) obtained in CIA on length of nonlinear interaction ℓ at different value of parameters are displayed in Fig. 1 for ZnO/PMMA 1. 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. With decrease of pump intensity (compare curves 1 and 2) and increases of losses (compare curves 2 and

3), the conversion efficiency falls down. In addition, the period of spatial beatings and optimum length of crystal increase. The latter is explained by the fact that at lower values of pump intensity for achievement of maximum conversion longer geometrical lengths of crystal are required. As is seen in figure, an increase in length of a sample approximately twice would permit to increase conversion efficiency several times. For the ZnO concentration of 3% wt. at I_{10} = 23 GW/cm^2, according to expression obtained in CIA, maximum efficiency is equal to 0.0037 (curve 2) and at pump intensity I_{10} = 17 GW/cm^2 maximum efficiency will be 0.0025 (curve 1).

The dependencies of conversion efficacy $\eta_2(\theta)$ are cited in Fig. 2 for Zno/PMMA 2. Analogical behavior of $\eta_2(\theta)$ takes place for ZnO/PMMA 1. The variation of length of nonlinear interaction in experiment was carried out by means of the rotation of the ZnO/PMMA 1st and 2nd structure, allowing changing of the incidence angle θ. The difference of these structures depends on the variation in the dimensions of ZnO nanocrystals. Let us consider the behavior of curves in Fig. 2. With decrease of pump intensity and ZnO concentration the conversion efficiency (at corresponding losses existing in experiment) falls down (compare curves 3, 4, 5 and 1, 2, 4). As the concentration of ZnO increase from 5% to 15%, an efficiency of frequency conversion increases almost 2 times. This results are from the fact that on one side, with growth of ZnO concentration the absolute value of the effective quadratic susceptibility decreases and on the other side, nanocrystal generates stronger second-harmonic wave due to the larger interaction thickness of the nonlinear medium (see Figs. 5 and 6). This fact was noted earlier in Ref. [4].

In Fig. 3 that is resulted the dependencies of conversion efficacy to second-harmonic on the reduced phase mismatch of interacting waves $\Delta\ell$. As one can expect, the curve for $\eta_2(\Delta\ell)$ depends on several parameters. As is seen in figure the increase of conversion efficiency is observed by rising of weight concentrations of ZnO (curves 2, 4 and 5), pump intensity (curves 1 and 2) and decrease of losses (curves 3

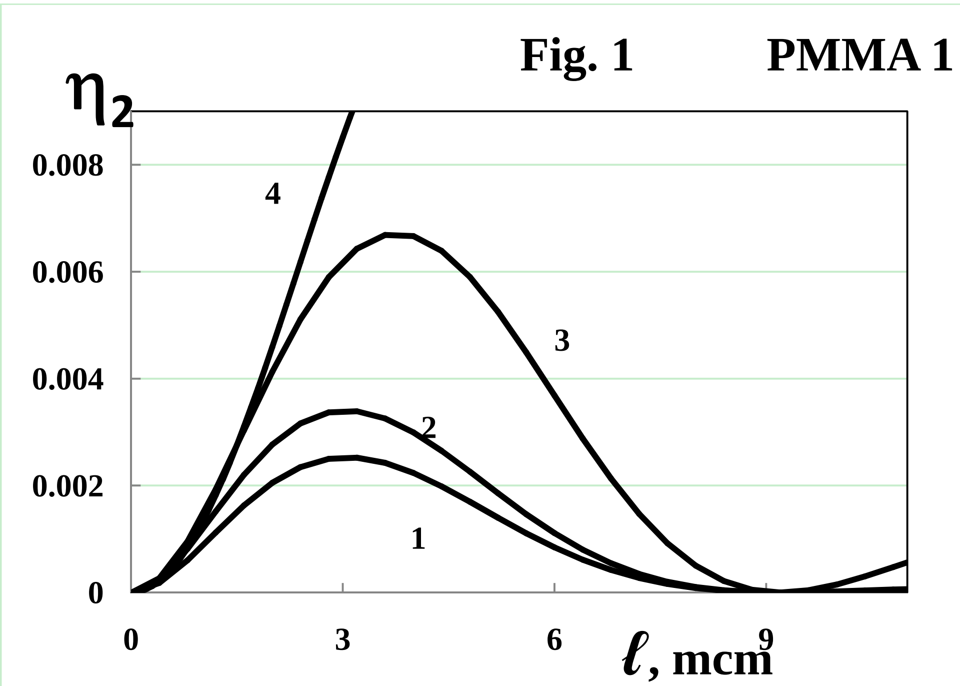

Fig. 1 Dependencies of conversion efficiency of pump wave (λ = 1.064 mcm) to the wave of the second harmonic η_2 on lengths for ZnO/PMMA nanocomposite films ℓ calculated in CIA (curves 1-3) and CFA (curve 4) for Δ=1.2 × 10^4 cm^{-1}, pump intensity of I_{10} = 17 GW/cm^2 (curves1 and 4), 23 GW/cm^2 (curves 2 and 3), and $\delta_2 = 2\delta_1$=0 (curve 4), 0.1 × 10^4 cm^{-1} (curve 3), 0.2 × 10^4 cm^{-1} (curves 1 and 2).

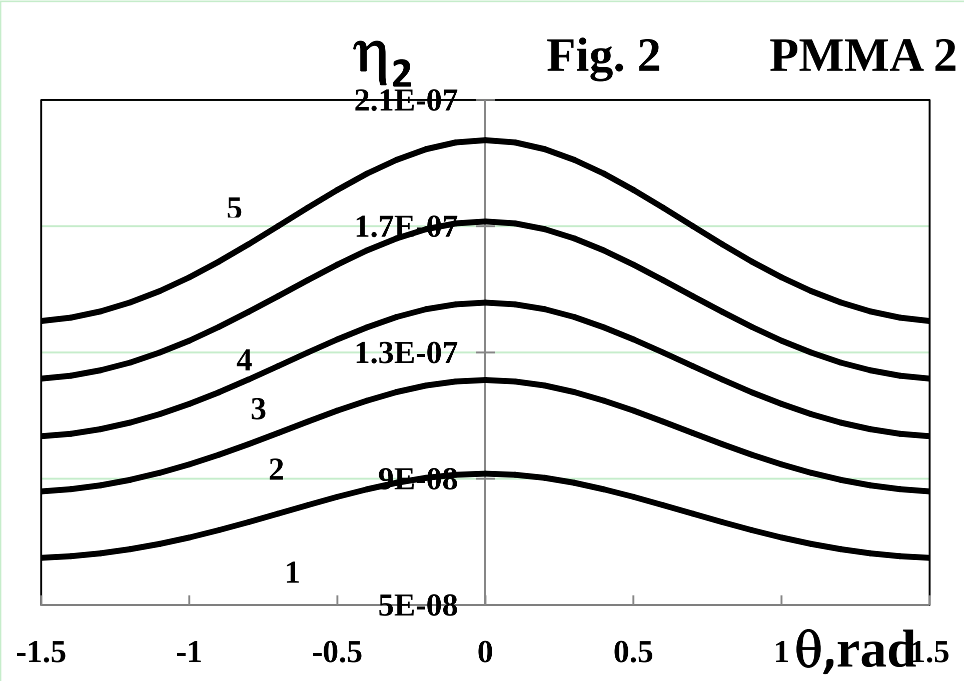

Fig. 2 Dependencies of conversion efficiency of pump wave to wave of second harmonic η_2 calculated in CIA versus the incidence angle θ for ZnO/PMMA 2 nanocomposite films with different ZnO concentration: 5% (curve 1), 10% (curve 2) and 15% (curves 3-5) at $\Delta = 1.2 \times 10^4$ cm^{-1}, pump intensity of I_{10} = 17 GW/cm^2 (curve 3), 20 GW/cm^2 (curves 2-4) and 23 GW/cm^2 (curve 5), $\delta_2 = 2\delta_1$ = 0.045 × 10^4 cm^{-1} (curve 1), 0.06 × 10^4 cm^{-1} (curve 2) and 0.09 × 10^4 cm^{-1} (curves 3-5). The thickness of all films is equal to 1 mcm [4].

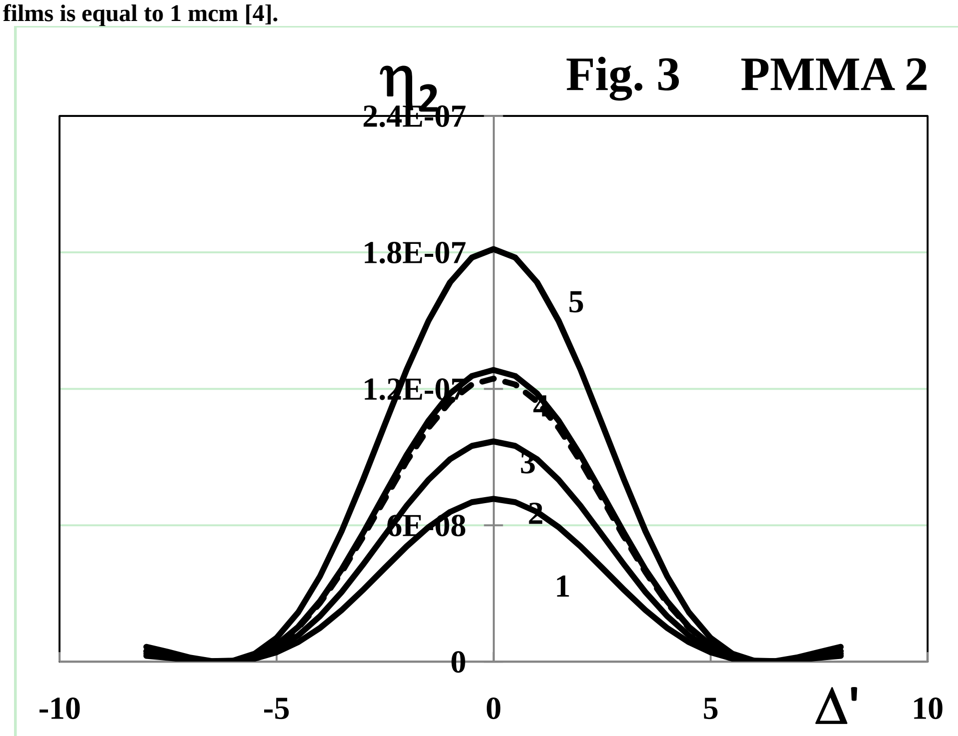

Fig. 3 Dependencies of conversion efficiency of pump wave to wave of second harmonic η_2 as a function of the phase mismatch $\Delta' = \Delta\ell$ calculated in CIA for ZnO/PMMA 2 nanocomposite films with different ZnO concentration: 5% (curves 1 and 2), 10% (curves 3 and 4) and 15% (curves 5) at pump intensity of I_{10}= 23 GW/cm^2 for $\delta_2 = 2\delta_1$ = 0.045 × 10^4 cm^{-1} (curves 1 and 2), 0.06 × 10^4 cm^{-1} (curve 4), 0.09 × 10^4 cm^{-1} (curve 5) and 0.9 × 10^4 cm^{-1} (curve 3). The thickness of all films is equal to 1 mcm [4].

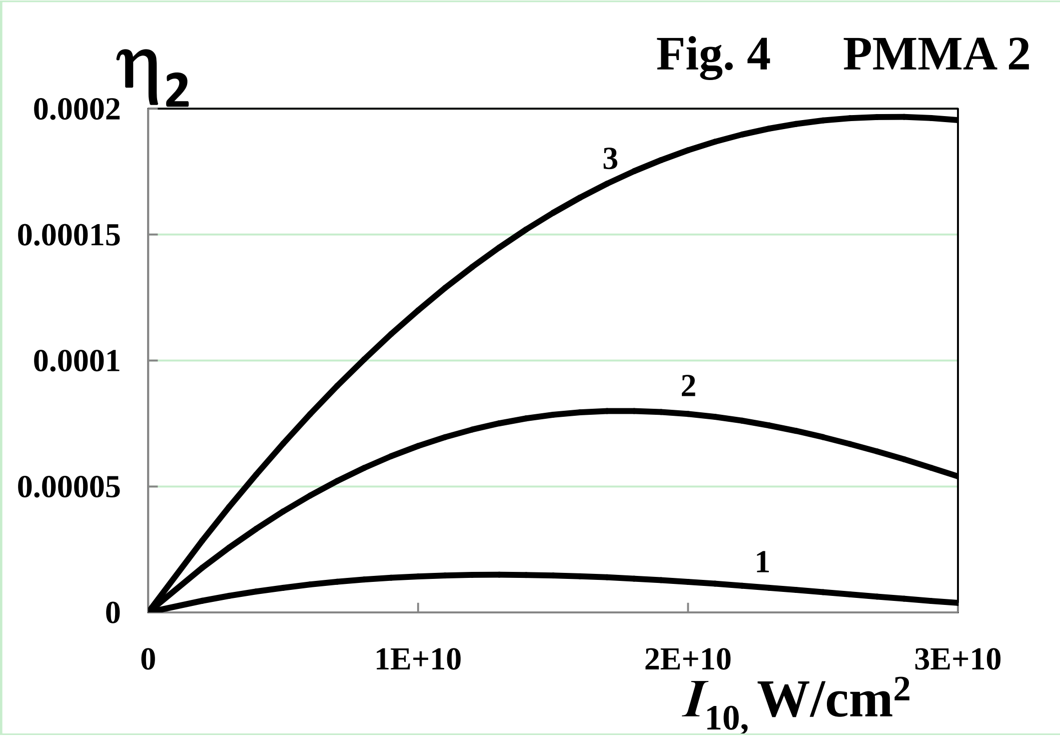

Fig. 4 Dependencies of conversion efficiency of pump wave to wave of second harmonic η_2 as a function of pump wave intensity calculated in CIA for ZnO/PMMA 2 nanocomposite films with ZnO concentration of 5%, $\delta_2 = 2\delta_1 = 0.045 \times 10^4$ cm^{-1} at $\Delta' = \Delta\ell$ = 30.14 (curve 1), 23 (curves 2 and 3), $d_{ZnO}^{equivalent}$ = 40 mcm (curve 3) and 50 mcm (curves 1 and 2). The thickness of all films is equal to 1 mcm [4].

and 4). The width of angular phase matching is reduced with decrease both in pump intensity (comparing curves 1-2) and weight concentrations of ZnO (comparing curves 2, 4 and 5).

In Fig. 4 the dependencies of conversion efficacy η_2 on the pump intensity I_{10} for two values of the equivalent thickness and phase mismatch are presented for Zno/PMMA 2. Analogical behavior of conversion efficacy $\eta_2(I_{10})$ takes place for ZnO/PMMA 1. As is seen in figure, laser radiation maximum converts, efficiently, into wave of second-harmonic at optimum value of pump intensity which decreases with increasing of length of nonlinear interaction (see curves 2 and 3). The latter is explained by the fact that at lower values of length of nonlinear interaction to achieve maximum conversion efficiency, higher level of pump intensity will be required. From comparison of curves 1 and 2 it is seen that with 1.3 times' decrease of phase mismatch the efficiency will increase ~5.36 times.

Let us consider practical example of the nonlinear interaction of optical waves at SHG in ZnO/PMMA compounds [4]. As is known, in this compound the crystal ZnO responsible for nonlinear properties of a structure and PMMA plays the role of matrix. Authors of this work use nonlinear interaction in ZnO, which is connected to relevant second order susceptibility (up to 10 pm/V) and which strongly depend on degree of the crystallinity of the film and the grain boundaries [1]. In the process of doubling, the generation of radiation takes place on a wavelength of the order of 0.532 mcm. Let us estimate the efficiency of conversion to this wavelength for experimentally realized value of intensity of laser generation which is changed in the range from 5 to 23 GW/cm^2. It should be noted that, even for input intensities in the range of 23 GW/cm^2, no structural changes or damages of the samples occurred [4].

The dependencies of the analytical expression for intensity of the second harmonic per square of input laser intensity I_2 / I_{10}^2 got in CIA on the incidence angle θ displayed in Fig. 5 for ZnO/PMMA 1 and Fig. 6 for ZnO/PMMA 2. The change in angle, in the given diapason, would permit to vary ℓ_{eff} on nanometer scale. When the ZnO concentration is 3% wt. in ZnO/PMMA 1, the effective length, (ℓ_{eff}), is changed in the range of (4÷ 5.2) nm (curve 1), at 7%, this range is about 10.6-12.67 nm (curve 2) and at concentration of 16%, it is changed in the range of 26-32 nm (curve 3). Thus, the numerical calculation of the efficacy obtained in CIA confirms the next fact that at higher concentrations of ZnO, the films generate stronger second harmonic signal due to the larger interaction length of the nonlinear medium.

According to expression obtained in CIA for optimum length of $\ell_{1,opt} = \lambda_1^{-1} \arctan(\lambda_1 / \delta_2)$, the coherent length of the structure is equal to 3.2 mcm (for the ZnO concentration of 3% wt. at I_{10}=23 GW/cm^2 incurve 2 of Fig. 1). The length ℓ_{eff} of nonlinear interaction for nanocomposite in experiment [4] (at ZnO dimension ℓ = 1 mcm) corresponds to the initial part of considered theoretical dependence of $\eta_2(\ell)$, where efficiency is low. An increase in length of a sample would permit to increase conversion efficiency of $\eta_2(\ell_{eff})$.

Let us estimate the dependencies of the ratio I_2 / I_{10}^2 for ZnO/PMMA 1 and ZnO/PMMA 2 nanocomposite films on its ZnO weight concentrations calculated by authors in CIA (dashed curves 1 and 3, correspondingly). Here, the analogical

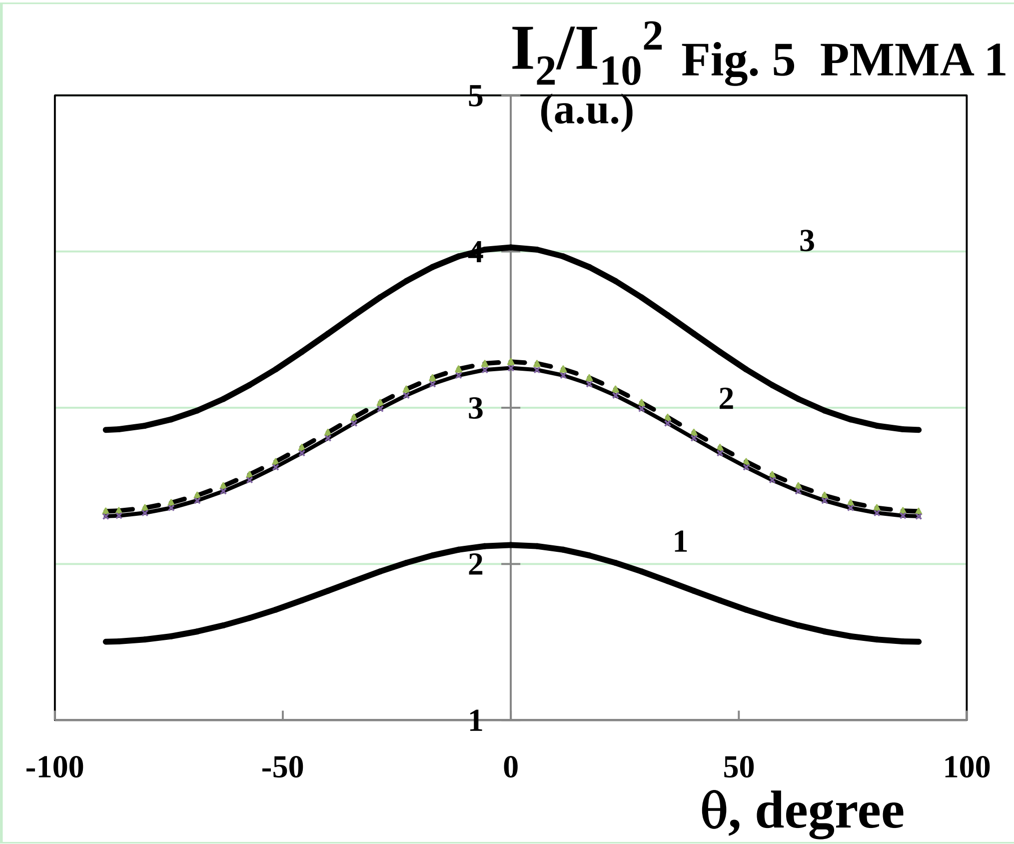

Fig. 5 Dependencies of intensity of the second harmonic per square of input laser intensity I_2 / I_{10}^2 got in CIA on the incidence angle θ for ZnO/PMMA 1nanocomposite films with different ZnO concentration: 3% (curve 1), 7% (curve 2), 12% (curve 3) and 16% (curves 4) at pump intensity I_{10} = 20 GW/cm^2, Δ = 1.2 × 10^4 cm^{-1}, $\delta_2 = 2\delta_1$ = 0.2 × 10^4 cm^{-1} (curve 1), 0.22 × 10^4 cm^{-1} (solid curve 2), 0.299 × 10^4 cm^{-1} (dashed curve 2), 0.28 × 10^4 cm^{-1} (curve 3). The thickness of all films is equal to 1 mcm [4].

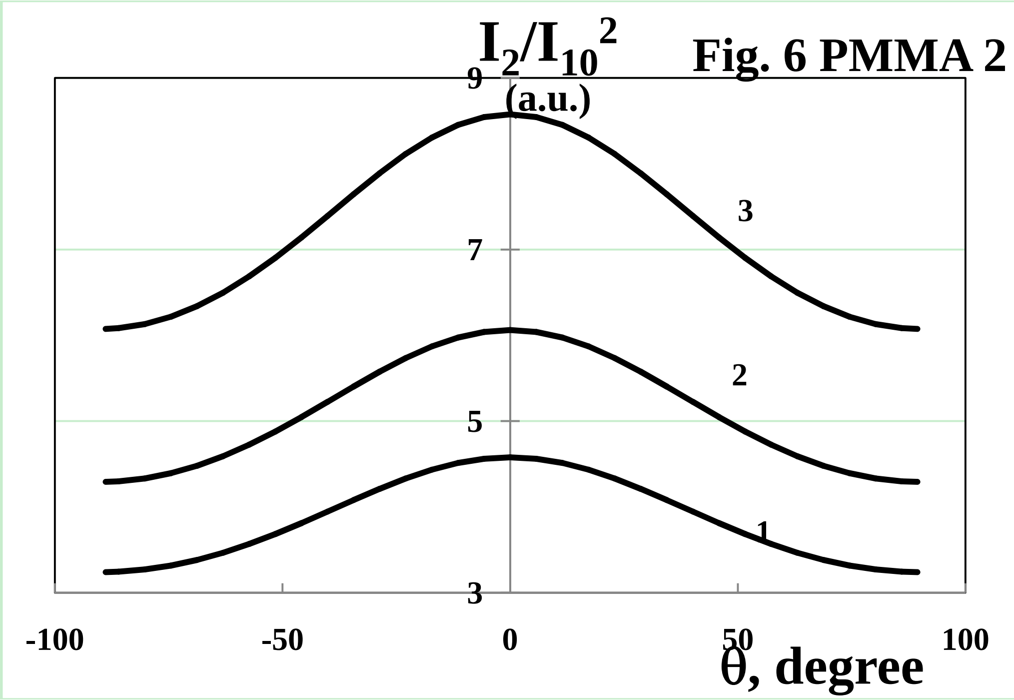

Fig. 6 Dependencies of intensity of the second harmonic per square of input laser intensity I_2/I_{10}^2 got in CIA on the incidence angle θ for ZnO/PMMA 2 nanocomposite films with different ZnO concentration: 5% (curve 1), 10% (curve 2) and 15% (curve 3) at pump intensity I_{10} = 20 GW/cm^2, Δ=1.2 × 10^4 cm^{-1}, $\delta_2 = 2\delta_1$ = 0.045 × 10^4 cm^{-1} (curve 1), 0.06 × 10^4 cm^{-1} (curve 2), 0.09 × 10^4 cm^{-1} (curve 3). The thickness of all films is equal to 1 mcm [4].

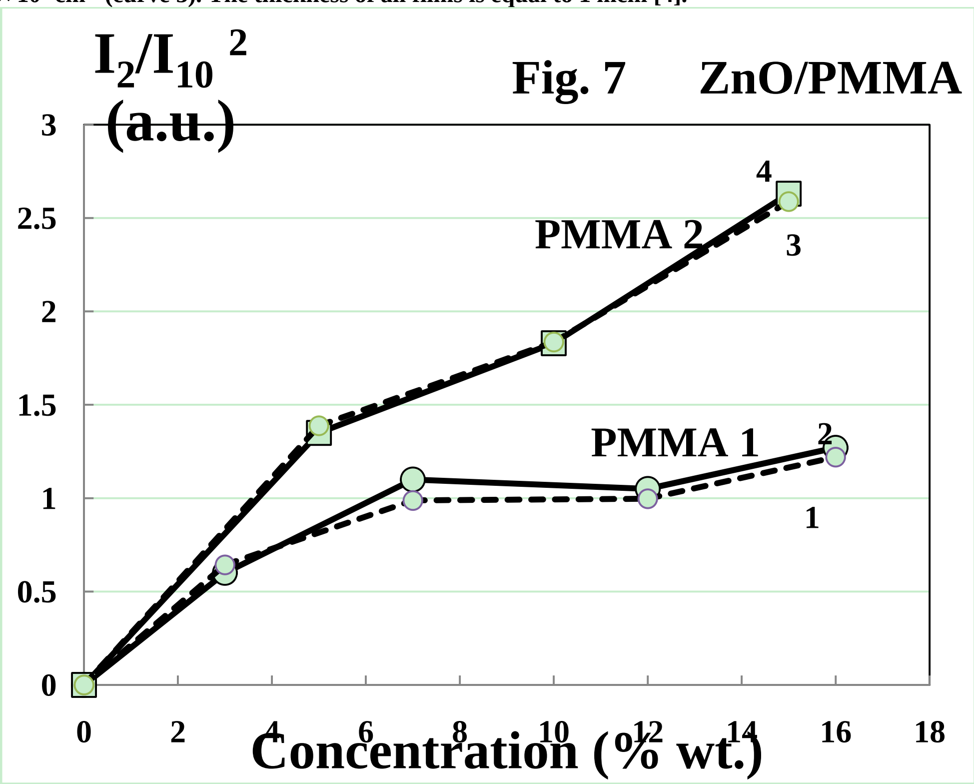

Fig. 7 Dependencies of the ratio I_2/I_{10}^2 for ZnO/PMMA 1 and ZnO/PMMA 2 nanocomposite films on its ZnO weight concentrations calculated in the CIA (dashed curves 1 and 3, correspondingly). Experimental results are given (solid curves 2 and 4) from Ref. [4]. The thickness of all films is equal to 1 mcm [4].

experimental results are given (solid curves 2 and 4) from Ref. [4] (see Fig. 7). As is seen from comparison of secures, the good agreement between experimental and theoretical results are observed (dashed and solids curves 1 and 2).

It should be noted that analysis in the CIA shows that on ZnO/PMMA 1 nanocomposite films, the maximum conversion might be obtained, for example, at length ℓ_{eff} of the nonlinear medium equal to 3.5 mcm and at the pump intensity of 23 GW/cm^2 than the case of experiment (material thickness 1 mcm [4]) (see Fig. 1). Also, an increase of pump intensity up to 20 GW/cm^2 would permit to obtain for ZnO/PMMA 2 the maximum efficiency at length ℓ_{eff} = 50 mcm which exceeds the experimental values approximately ~ 6 times (see curve 2 of Fig. 4 and curve 1 of Fig. 2).

5. Conclusions

Thus, theoretical investigation of frequency conversion in ZnO/PMMA structures with account for phase effects allows one to reveal the ways of increasing conversion efficiency. Namely at the given values of the length of crystal converter, it is possible to calculate the optimum value of pump intensity. As well, at chosen pump intensity of laser radiation it is possible to calculate the coherent length of a crystal converter. The analytical method also permits to estimate the expected conversion efficiency on different wavelengths of laser radiation. Thus, the numerical calculation of the efficacy obtained in CIA confirms the next fact that at higher concentrations of ZnO the films generate stronger second harmonic signal due to the larger interaction length of the nonlinear medium.

The results of carried out researches will be useful at elaborations of the modern devices of the quantum electronics and in the nanoscale optoelectronic circuitry. Method of analysis of SHG in ZnO/PMMA structures, developed in the present work, may be used for investigation of other nanocomposite films.

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