Quantum Electronics **32** (1) 49 – 53 (2002)

PACS numbers: 42.65.Ky; 42.70.Qs
DOI: 10.1070/QE2002v032n01ABEH002125

Second harmonic generation in photonic-crystal one-dimensional media

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***Abstract*. SHG is analysed in photonic crystals of two types (crystals consisting of quarter-wavelength GaAs layers alternating with silica layers and AlAs/GaAs crystals) depending on the number of structure periods and the pump intensity. Calculations show that in photonic crystals with the difference in the refractive indices $\Delta n = 0.585$, SHG occurs only in the reflected light and in two directions for a periodic structure with $\Delta n = 2$.**

Keywords: *photonic crystal, second harmonic generation, conversion efficiency, density of optical modes.*

1. Introduction

The problem of frequency doubling of femtosecond pulses in photonic crystals is of interest for increasing the efficiency of the pump radiation conversion to the second harmonic, as well as for the study of nonlinear-optical properties of periodic structures called photonic crystals. Photonic crystals represent periodic structures consisting of dielectrics selected in such a way that the density of electromagnetic field modes in these structures has a band gap for all directions of the wave vector. As a rule, a photonic crystal is assumed to be a three-dimensional structure [1, 2]; however, one-dimensional structures also feature interesting properties, which are typical for 'true' photonic crystals. The presence of the band gap in a photonic crystal opens up new possibilities for controlling optical processes occurring in them [3 – 5].

Much attention is currently paid to the growth of various periodic structures of a complicated structure and to the development of mathematical models for their description. The interaction of femtosecond light pulses with one-dimensional periodic structures possessing cubic and quadratic nonlinearities has been studied in many papers [3 – 12].

In a periodic one-dimensional structure, in which one of the layers is a crystal with quadratic nonlinearity, SHG for transmitted and reflected pump radiation is possible. The nonlinear interaction of the waves depends substantially on phase relations between the pump wave and harmonic, the dispersion of the refractive index in a nonlinear medium, as well as on the mismatch between phase and group velocities in a nonlinear crystal. To solve this problem correctly, one should know the transmission and reflection spectra at the frequencies of the interacting waves, the dispersion relations, and the density of modes in the optical spectrum of a photonic crystal.

The temporal characteristics of the second harmonic and the transmitted pump radiation, which are typical for SHG of femtosecond pulses, namely, the shortening of the pulse duration in the quasi-static approximation at low conversion efficiency and broadening of pulses due to the reverse effect of the second harmonic on the pump [13], should be also manifested in periodic structures.

It was shown in papers [8, 10] that the SHG efficiency in a photonic crystal is several orders of magnitude higher than in a nonlinear crystal of the same length, and it increases with increasing the difference between the refractive indices of layers comprising its period and with increasing the crystal length. We also demonstrated [11] a high SHG efficiency in a periodic structure with a nonlinear GaAs crystal compared to that in a crystal of the same length.

The aim of this paper is to calculate SHG, linear transmission and reflection spectra at the pump and second-harmonic frequencies, and the density of optical modes for quarter-wavelength periodic structures of two types (AlAs/GaAs and fused silica with GaAs) upon pumping by a femtosecond Nd glass laser as functions of the number of periods of the photonic crystal and the pump intensity.

2. Linear properties of photonic crystals at the pump and second-harmonic frequencies

To find the dependences of the transmission (T) and reflection (R) coefficients at the pump and second-harmonic frequencies on the number $N_{\rm st}$ of periods of the structure, we used the characteristic matrices of the layers with different refractive indices [14]. We employed the Chebyshev functions of second kind, whose argument β satisfied the dispersion relation

$$\cos\beta = \cos\varphi_1\cos\varphi_2 - {}^1/_2(p_1/p_2 + p_2/p_1)\sin\varphi_1\sin\varphi_2, \qquad (1)$$

where $\varphi_j = (\omega/c)l_j(n_j^2 - \alpha^2)^{1/2}$; $p_j = n_j\cos\theta_j$; $\alpha = n_i\sin\theta_i$; n_j, l_j are the refractive index and the thickness of the jth layer, respectively; and θ_i, θ_j are the angles of incidence of

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Received 2001 8 May 2001; revision received 11 October 2001
Kvantovaya Elektronika **32** (1) 49 – 53 (2002)
Translated by M.N.Sapozhnikov

radiation at the entrance of and exit from the layer; n_i is the refractive index of the medium from which radiation is incident on the structure.

The density of optical modes is defined as a derivative with respect to the frequency from the effective wave number $k_{\rm eff}$ of the structure [15–17]

$$\rho(\omega) = \frac{{\rm d}k_{\rm eff}}{{\rm d}\omega}. \quad (2)$$

To calculate $\rho(\omega)$, we should find the phase shift of the waves caused by the properties of the given photonic crystal. Let us represent the amplitude coefficients $t(\omega)$ and $r(\omega)$ in the form $t = {\rm e}^{{\rm i}\varphi}\sqrt{T}, r = {\rm e}^{{\rm i}\psi}\sqrt{R}$, where φ and ψ are phases acquired by a plane wave after its transmission and reflection, respectively. The effective wave number $k_{\rm eff}$ for the structure of length $D = (l_1 + l_2)N_{\rm st}$ is introduced so that the phase is determined by the expression $\varphi = k_{\rm eff}D$.

By representing the transmission coefficient in the form $t = u + {\rm i}v$, we can determine φ from the expression $\tan(k_{\rm eff}D) = v/u$ and obtain the dispersion relation

$$k_{\rm eff}(\omega) = \frac{1}{D}\tan^{-1}\left[\frac{v(\omega)}{u(\omega)}\right].$$

In this case, the density of optical modes will have the form

$$\rho(\omega) = \left|\frac{{\rm d}k_{\rm eff}}{{\rm d}\omega}\right| = \frac{1}{D}\left|\frac{u'v - v'u}{u^2 + v^2}\right|, \quad (3)$$

where the primes denote derivatives from the real and imaginary parts of the amplitude transmission coefficient. The densities $\rho_\omega, \rho_{2\omega}$ of optical modes are calculated by expressions (1)–(3), in which the calculated amplitudes and phases of transmission coefficients and their derivatives were used.

The dependence of linear properties on the number $N_{\rm st}$ of periods was analysed for a photonic crystal consisting of a nonlinear GaAs crystal (with refractive indices $n_1(\omega) = 3.45$, $n_1(2\omega) = 3.1$) with different dielectrics: fused silica ($n_2(\omega) = 1.45$, $n_2(2\omega) = 1.46$) and AlAs ($n_2(\omega) = 2.865$, $n_2(2\omega) = 2.9$). In the spectral regions between band gaps, the additional calculation was performed by expressions from [17]. The results of calculations of transmission coefficients and $\rho_\omega, \rho_{2\omega}$ were in good agreement. It follows from the calculated spectra that the band gap increases with increasing the difference between the refractive indices. The increase in the number of periods in the photonic crystal has no effect on the band gap but results in the increase in the number of maxima of the transmission coefficient T and of the minima of the reflection coefficient R.

The dependences of the transmission coefficient at the fundamental (T_ω) and doubled ($T_{2\omega}$) frequencies and the densities $\rho_\omega, \rho_{2\omega}$ of optical modes on the number $N_{\rm st}$ of periods for two photonic crystals are shown in Fig. 1. One can see from Fig. 1a that for the photonic crystal with $\Delta n = n_2 - n_1 = 0.585$ and $N_{\rm st} = 10$, the transmission coefficients for the pump and second harmonic are almost the same: $T_\omega = 0.26$ and $T_{2\omega} = 0.248$, which should be mani-

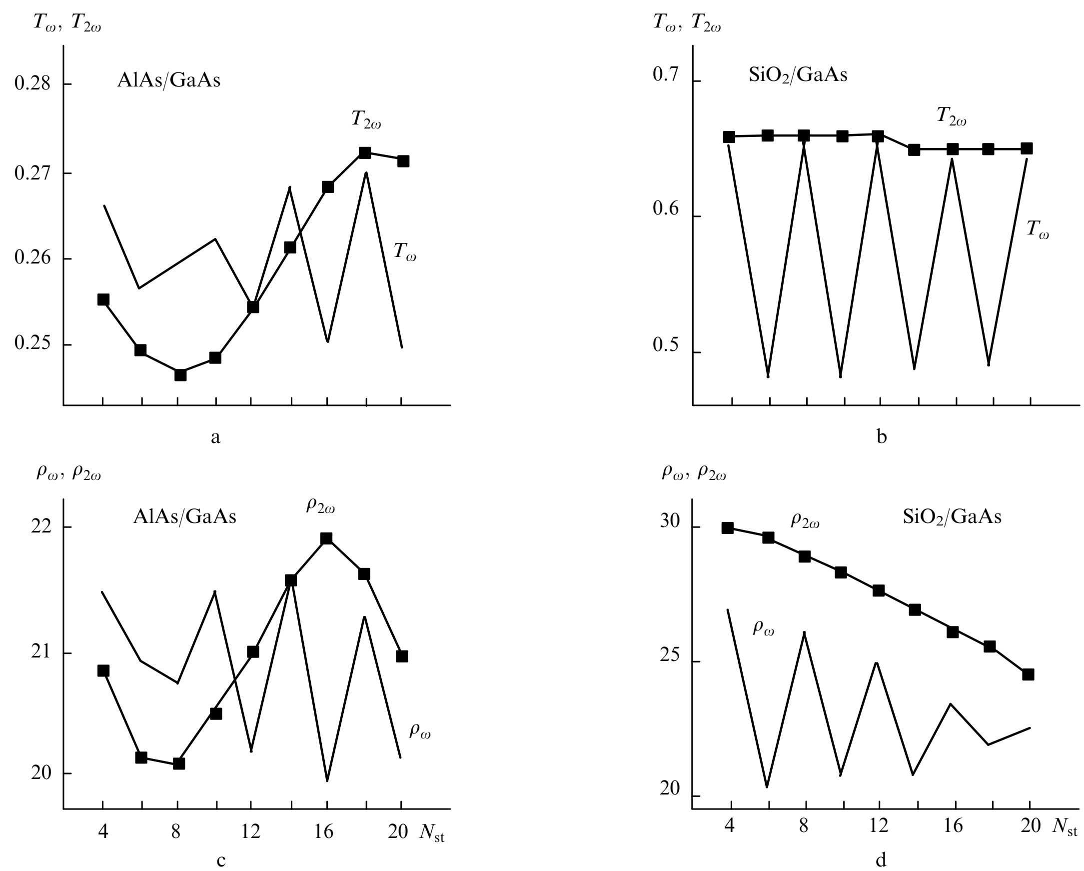

Figure 1. Dependences of transmission coefficients T_ω, $T_{2\omega}$ (a, b) and densities of optical modes ρ_ω and $\rho_{2\omega}$ normalised to the speed of light (c, d) at the fundamental and doubled frequencies on the number of periods of photonic crystals based on AlAs/GaAs (a, c) and SiO$_2$/GaAs layers (b, d).

fested in SHG predominantly in reflection. As the number N_{st} of periods increases, transmission at the doubled frequency monotonically increases, whereas at the fundamental frequency it exhibits oscillations. The densities $\rho_\omega, \rho_{2\omega}$ behave similarly (Fig. 1c); the density $\rho_{2\omega}$ has a maximum at $N_{st} = 16$.

For the photonic crystal with $\Delta n = 2$ (Figs 1b, d) the transmission coefficients for the pump and the second harmonic for ten layers of the structure are 0.5 and 0.7, respectively. Therefore, both the transmitted and reflected waves should be present in the second harmonic. The densities $\rho_\omega, \rho_{2\omega}$ decrease with increasing number of periods.

3. Nonlinear interaction of the pump and second-harmonic waves in a photonic crystal

The numerical simulation of SHG by femtosecond pulses was performed for two photonic crystals considered above and a nonlinear GaAs crystal, which has a cubic lattice and the nonlinear susceptibility $\chi^{(2)}$ at 1060 nm that exceeds $\chi^{(2)}$for a KDP crystal by a factor of 500 [18]. A system of equations in second-order partial derivatives was used in the second approximation of the dispersion theory for the pump and second-harmonic waves [11, 13]. The medium was assumed inertialess down to duration 10^{-14} s.

The field in the medium was written in the form

$$E(z,t) = E_1^+(z,t)\exp(\mathrm{i}k_1 z - \mathrm{i}\omega t) + E_1^-(z,t)\exp(-\mathrm{i}k_1 z - \mathrm{i}\omega t) + E_2^+(z,t)\exp(\mathrm{i}k_2 z - 2\mathrm{i}\omega t) + E_2^-(z,t)\exp(-\mathrm{i}k_2 z - 2\mathrm{i}\omega t) + \text{c.c.}$$

By using Maxwell equations and the approximation of slowly varying envelope for the amplitudes of the pump and second-harmonic waves, we obtain the system of equations describing SHG in a nonlinear crystal

$$-\mathrm{i}D_1\frac{\partial^2 E_1^+}{\partial t^2} + \frac{\partial E_1^+}{\partial z} + \frac{1}{u_1}\frac{\partial E_1^+}{\partial t} = -\mathrm{i}\gamma_1 E_2^+ E_1^{+^*}\mathrm{e}^{\mathrm{i}\Delta kz},$$

$$-\mathrm{i}D_1\frac{\partial^2 E_1^-}{\partial t^2} - \frac{\partial E_1^-}{\partial z} + \frac{1}{u_1}\frac{\partial E_1^-}{\partial t} = -\mathrm{i}\gamma_1 E_2^- E_1^{-^*}\mathrm{e}^{-\mathrm{i}\Delta kz},$$

$$-\mathrm{i}D_2\frac{\partial^2 E_2^+}{\partial t^2} + \frac{\partial E_2^+}{\partial z} + \frac{1}{u_2}\frac{\partial E_2^+}{\partial t} = -\mathrm{i}\gamma_2 E_1^{+^2}\mathrm{e}^{-\mathrm{i}\Delta kz}, \qquad (4)$$

$$-\mathrm{i}D_2\frac{\partial^2 E_2^-}{\partial t^2} - \frac{\partial E_2^-}{\partial z} + \frac{1}{u_2}\frac{\partial E_2^-}{\partial t} = -\mathrm{i}\gamma_2 E_1^{-^2}\mathrm{e}^{\mathrm{i}\Delta kz},$$

where $\gamma_i = 2\pi\chi^{(2)}\omega_0^2/k_i c^2; i = 1,2; \omega_0$ is the pump frequency; $u_{1,2}$ and $k_{1,2}$ are the group velocities and wave vectors of the fundamental and second harmonics, respectively; $\Delta k = 2k_1 - k_2$ is the mismatch of the wave vectors of the pump and second-harmonic waves in the nonlinear crystal; and

$$D_i = \frac{1}{2}\frac{\partial k_i^2}{\partial \omega_i^2}$$

is the group-velocity dispersion for the pump and second-harmonic waves in the nonlinear crystal and dielectric.

The system of equations (4) assumes the use of the algorithm of the solution presented in paper [11]. However, in this paper we used the method for calculation of the amplitudes of a spatial lattice similar to that proposed in papers [5, 6]. The accuracy of numerical calculations was increased by using the Fourier-transform parameter $N = 512$.

The systems of equations (4) was solved by representing the pump by the Gaussian pulse $E(z=0,t) = E_0\exp\{-2 \times \ln 2[(t-t_0)/\tau_p]^2\}$, where E_0 is the pump-pulse amplitude; $\tau = 100$ fs is the pump-pulse duration; and t_0 is the position of the pump pulse on the time axis. The following normalisation of variables was used: $z' = z/\lambda_0$, $t' = t/\tau_p(\lambda_0$ is the pump wavelength, the intensities of the pump and second-harmonic waves were normalised to the intensity 1 GW cm^{-2}. The optical lengths of a nonlinear layer (l_1) and dielectric (l_2) were assumed equal to $0.25\lambda_0$. The mismatch of the phase velocities of the pump and second-harmonic waves in the nonlinear crystal under initial conditions was calculated from the expression

$$\Delta k = \frac{4\pi}{\lambda_0}[n_1(\omega) - n_1(2\omega)].$$

The numerical calculation of the system of equations (4) gave the dependences of the intensities of transmitted ($I_g^+ = |E_2^+|^2$) and reflected ($I_g^- = |E_2^-|^2$) second-harmonic pulses, as well as of their durations τ_g^+, τ_g^- and conversion efficiencies $\eta^\pm$, calculated as the ratio of the second-harmonic energy for transmitted ($W_g^+ = I_g^+\tau_g^+$) and reflected ($W_g^- = I_g^-\tau_g^-$) waves to the pump energy W_0, on the number N_{st} of periods of the lattice and the pump intensity I_0.

Fig. 2 shows the results of numerical simulations obtained for two above-mentioned photonic crystals at the initial pump density 100 GW cm^{-2}. One can see from Fig. 2a that transmission of the pump radiation decreases with increasing number of periods in the photonic crystal, the reflected wave being dominant. The SHG efficiency η^- has a maximum for a photonic crystal with $N_{st} = 10$ (Fig. 2c), the ratio of energies of the reflected and transmitted waves being ~ 35.

For a photonic crystal with $n = 2$, the transmitted pump wave dominates over the reflected wave (Fig. 2b), and the SHG efficiency has a maximum for $N_{st} = 8$. The maximum SHG efficiency for the counterpropagating wave is 1.75 times lower than that for the copropagating wave, and it is observed for $N_{st} = 10$ (Fig. 2d). The total SHG efficiency is lower by a factor of three than that for a lattice consisting of AlAs/GaAs layers.

Fig. 3a shows the dependences of the duration of reflected and transmitted SHG pulses τ_g normalised to the pump-pulse duration on the pump amplitude E_0, and Fig. 3b presents similar dependences for the SHG efficiency for the AlAs/GaAs photonic crystal for $N_{st} = 10$. One can see that as the pump amplitude increases by an order of magnitude, the SHG efficiency η^- increases by a factor of 90 (Fig. 3b); for small values of efficiencies, the second-harmonic pulse is $\sqrt{2}$ times shorter than the pump pulse, in accordance with the analytic estimate applied for these conditions [13], as well as with calculations [11]. In this case, the durations of the transmitted and reflected pulses almost coincide.

One can see from Fig. 3a that the duration of the second-harmonic pulses increases with increasing the

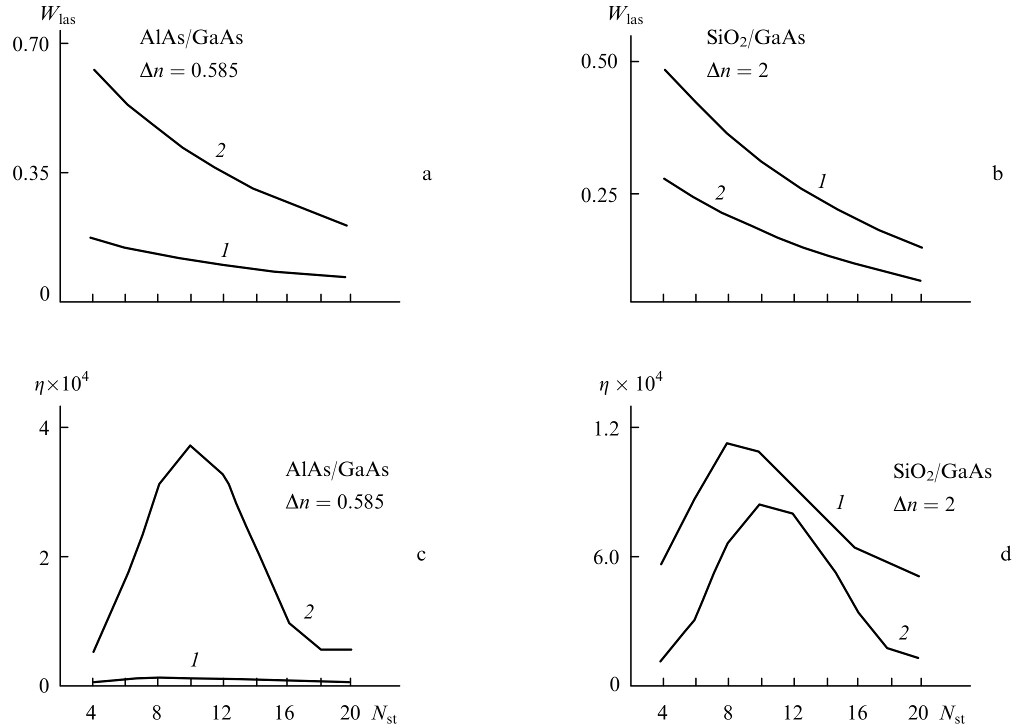

Figure 2. Dependences of energies W_{las} of transmitted (*1*) and reflected (*2*) pump radiation (a, b) and of the SHG efficiency η (c, d) on the number of periods N_{st} for photonic crystals consisting of layers of silica and GaAs (a, c) and AlAs/GaAs (b, d).

pump-pulse amplitude, the duration of the reflected pulse increasing in a greater extent. Such behaviour of the duration of second-harmonic pulses is caused by the nonlinear phase modulation due to the reverse effect of second-harmonic waves on the pump [13].

4. Discussion of calculation results

We calculated numerically the intensities of copropagating and counterpropagating second-harmonic waves, the SHG efficiency, and the pulse durations for photonic crystals consisting of a nonlinear crystal and different dielectrics as functions of N_{st} and E_0. For both photonic crystals studied, the SHG efficiency has a maximum for $N_{st} = 8 - 10$, in agreement with the data [11]. For the pump wavelength used in calculations, SHG occurs only in reflection light for the photonic crystal with AlAs/GaAs layers ($\Delta n = 0.585$) and in both directions in the case of the periodic structure with $\Delta n = 2$ (GaAs and silica). The SHG efficiency for the transmitted wave exceeds that for the reflected wave by a factor of 1.75. The SHG efficiency in reflection for the first photonic crystal is three times higher than the total SHG efficiency in both directions for the second photonic crystal.

Let us compare, similarly to [8, 10], the SHG efficiencies in a photonic crystal and a nonlinear crystal of the same length by using analytic estimates for the second-harmonic amplitude in the femtosecond-pulse field in the case of the quasi-static interaction of the second harmonic and the pump [13]. The results of calculations of the SHG efficiency and second-harmonic pulse duration show that such a SHG regime took place in the nonlinear crystal (the reverse effect of the second harmonic on the pump was negligible and the second-harmonic pulse duration decreased by a factor of $\sqrt{2}$). In this case, we can use the expression for the second-harmonic amplitude [13] and estimate the SHG efficiency

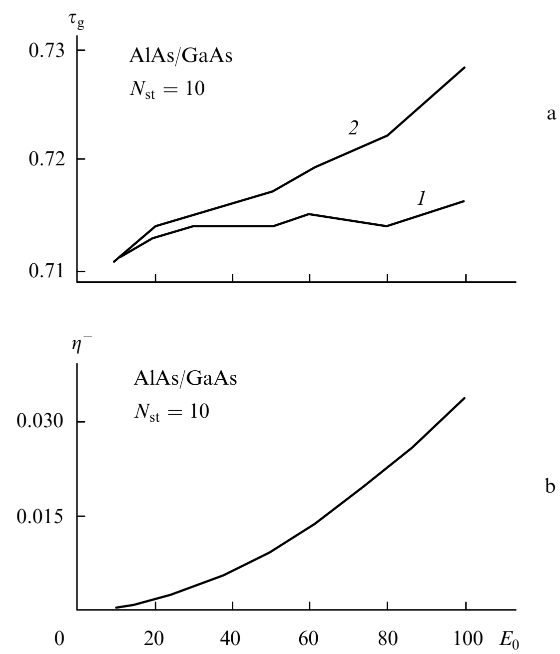

Figure 3. Dependences of the duration τ_g (a) of transmitted (*1*) and reflected (*2*) pulses and of the SHG efficiency η^- for the reflected pulse (b) on the pump amplitude E_0.

for a crystal whose length is equal to the total length of nonlinear layers of the photonic crystal for nonlinearity parameters used in calculations with the help of the expression

$$\eta \approx (\gamma d)^2 I_0 \frac{\sin^2(\Delta kl/2)}{(\Delta kl/2)^2},$$

where γ is the nonlinear conversion coefficient and d is the crystal length.

Thus, for a nonlinear crystal with the length equal to the length of a photonic crystal with $N_{st} = 10$, the SHG efficieny η is approximately 200 times lower at the pump intensity $I_0 = 100$ GW cm^{-2} and 400 times lower at $I_0 = 400$ GW cm^{-2} than for the photonic crystal (Fig. 2c). A comparable value of η was obtained in SHG [8, 11] and SRS [12] calculations.

Analysis of numerical simulations of SHG in photonic crystals of two types showed that, when the difference between the refractive indices of the photonic crystal was great and the number layer was large, the SHG efficiency was lower than it follows from papers [8, 10]. This is explained by the interference properties of the photonic crystal, as one can see from the analysis of spectral and dispersion characteristics of periodic structures with different dielectrics. Let us compare the results of calculations for the linear and nonlinear problems.

According to the definition of $\rho_\omega, \rho_{2\omega}$, one can see that ρ_ω^{-1} and $\rho_{2\omega}^{-1}$ determine the group mismatch of the interacting pump and second-harmonic waves in the SHG process in a periodic structure, similarly to the group mismatch in a nonlinear crystal:

$$\Delta u = \left(\frac{\partial k}{\partial \omega}\right)^{-1}_{2\omega} - \left(\frac{\partial k}{\partial \omega}\right)^{-1}_{\omega}. \qquad (5)$$

The values of $\rho_\omega, \rho_{2\omega}$, for $N_{st} = 10$ shown in Figs. 1c, d give $\Delta u^{-1} = 0.7 \times 10^{-13}$ s cm^{-1} for the photonic crystal with AlAs/GaAs and $\Delta u^{-1} = -3.73 \times 10^{-12}$ s cm^{-1} for GaAs and silica, i.e., the photonic crystal of the second type has anomalous dispersion. From this, we can calculate the group delay $l_g = \tau_p |\Delta u^{-1}|$, which determines the interaction length of the pump and second-harmonic waves until their complete mismatch in space due the dispersion of the medium. We assumed in calculations that $\tau_p = 100$ fs. The effective interaction length for the AlAs/GaAs photonic crystal is $l_g = 1.4$ cm, whereas for the SiO_2/GaAs photonic crystal, $l_g = 0.27$ cm. In both cases, the nonlinear interaction length was substantially shorter than the group delay length (5), which is related to the energy and phase effects of interaction of the waves in SHG. Nevertheless, the solution of the system of equations (4) showed that photonic crystals with a greater group-delay length (AlAs/GaAs) have a higher SHG efficiency.

5. Conclusions

Our analysis of the spectral properties of photonic crystals consisting of the nonlinear crystal and different dielectrics has demonstrated the possibility of the efficient use of photonic crystals for SHG. It follows from the analysis that transmission and reflection coefficients determine the relation between the transmitted and reflected second-harmonic waves, while the density coefficients of optical modes give the estimate of the SHG efficiency.

The SHG efficiency in a periodic structure with $N_{st} = 10$ is several hundreds times greater than that in a nonlinear crystal of the same length. SHG only in reflected light in media with a low lattice contrast makes the periodic structure promising for using as a cavity nonlinear mirror in a femtosecond laser.

Acknowledgements. The author thanks S.Ya.Kilin for useful comments in the discussion of the paper.

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