

Stimulated Raman scattering of light in a photonic crystal

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Abstract. The results are presented of calculating the stimulated Raman scattering (SRS) of femtosecond pulses in a periodic structure consisting of alternating quarter-wave plates of fused silica and a Nd:KGW crystal. The analysis of the calculations shows that efficient SRS conversion of the pump can be obtained only if the density of the electromagnetic field states is increased by varying the spectral properties of the periodic structure.

1. Introduction

Periodic dielectric structures whose spatial periods are close to the radiation wavelength and whose refractive indices satisfy certain relationships can become so-called photonic crystals, which are promising for applications in new devices employing femtosecond light pulses [1–5]. A characteristic feature of photonic crystals, which they share with semiconductors, is the ability to form a bandgap at some frequencies.

The studies of photonic crystals are motivated by their possible applications in lasers, optical computers, and other devices. Photonic crystals can have either opal-like structures that contain particles exhibiting nonlinear properties or they can represent multilayer crystalline structures. Currently, many papers are devoted to the photonic-crystal growing technology and simulations of the interaction between radiation and periodic structures.

The authors of [4, 5] proposed methods for calculating the interaction between radiation and photonic crystals that employ fast Fourier transform (FFT). Later, we developed a similar approach [6–8] to calculate generation of ultrashort pulses (USP) in a laser with forced mode locking and negative feedback, and to analyse the second-harmonic generation with femtosecond pulses in a photonic crystal. The obtained results have demonstrated the efficiency of the FFT method for calculating the interaction between USP and periodic structures.

In this work, we simulate numerically SRS of a femtosecond pulse in a periodic structure that consists of alternating layers of fused silica and $\text{Nd}^{3+}:\text{KGd}(\text{WO}_4)_2$ (Nd:KGW)

crystal, the latter serving as the scattering medium. The nonlinear properties of this crystal were earlier studied in Refs [9, 10]; it finds applications as a laser medium, and its nonlinear susceptibilities are high enough for the SRS conversion. Depending on the experimental conditions, both lasing and SRS conversion [9] can be observed in this crystal.

However, it is rather difficult to realise efficient SRS conversion in a crystalline nonlinear medium of length ten wavelengths only upon pumping by femtosecond light pulses in the nonstationary regime because the interaction length is small, while the nonstationary SRS-conversion coefficient is proportional to the square root of the scattering medium length [14, 15]. The situation can be remedied because an increase in the density of the electromagnetic field occurring in a photonic crystal, substantially enhances the nonlinear conversion. Earlier, only the possibility of a consideration of the SRS conversion in such systems has been discussed.

The purpose of this work was to simulate numerically the SRS conversion of the femtosecond pulse into the first Stokes component. We have calculated the linear properties of the periodic structure and the nonlinear interaction between the pump pulse, the scattering medium, and the dielectric. Using these data, we analysed the temporal and energy characteristics of the Stokes pulses created in the transmitted and reflected pump light as functions of the position of the pump frequency with respect to the periodic structure bandgap.

2. Formulation of the problem

To study the temporal and energy characteristics of SRS of a 100-fs laser pulse, transmitted and reflected by the photonic crystal, we employed the system of equations containing sources of medium polarisation [11–13] that was complimented by second time derivatives. In this way, we took into account the dispersion properties of both the nonlinear and linear media in the second order of the dispersion theory. As opposed to the problem considered in Ref. [11], in our case the wave that counterpropagates the pump is not the backward SRS wave, as in any nonlinear scattering medium, but rather is created in the field of light reflected by the photonic crystal.

The field inside the photonic crystal is a superposition of waves:

$$\begin{aligned}
 E(z, t) = & E_L^+ \exp(ik_L z - i\omega_L t) \\
 & + E_L^- \exp(-ik_L z - i\omega_L t) + E_S^+ \exp(ik_S z - i\omega_S t) \\
 & + E_S^- \exp(-ik_S z - i\omega_S t) + \text{c.c.},
 \end{aligned}$$

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where $E_{L,S}^{\pm}$, $\omega_{L,S}$, and $k_{L,S}$ are the slowly varying amplitudes, frequencies, and wave numbers of the laser (L) and Stokes (S) waves, respectively. Assuming that $\omega_L - \omega_S = \omega_{21}$, the polarisation of the scattering medium can be written as

$$P_{21}(z, t) = \{q^+(z, t) \exp[i(k_L - k_S)z] + q^-(z, t) \times \exp[-i(k_L - k_S)z]\} \exp[-i(\omega_L - \omega_S)t] + \text{c.c.},$$

where $q^{\pm}(z, t)$ are the medium polarisation amplitudes for the counterpropagating Stokes waves. To analyse the temporal and energy properties of the SRS, we used the following system of equations [7, 11–13]

$$\begin{aligned} -iD_L \frac{\partial^2 E_L^+}{\partial t^2} + \frac{\partial E_L^+}{\partial z} + \frac{1}{u_L} \frac{\partial E_L^+}{\partial t} &= i \frac{\omega_L}{\omega_S} g_{2S}^*(q^+) E_S^+, \\ -iD_L \frac{\partial^2 E_L^-}{\partial t^2} - \frac{\partial E_L^-}{\partial z} + \frac{1}{u_L} \frac{\partial E_L^-}{\partial t} &= i \frac{\omega_L}{\omega_S} g_{2S}^*(q^-) E_S^-, \\ -iD_S \frac{\partial^2 E_S^+}{\partial t^2} + \frac{\partial E_S^+}{\partial z} + \frac{1}{u_S} \frac{\partial E_S^+}{\partial t} &= i g_{2S}^*(q^+) E_L^+, \\ -iD_S \frac{\partial^2 E_S^-}{\partial t^2} - \frac{\partial E_S^-}{\partial z} + \frac{1}{u_S} \frac{\partial E_S^-}{\partial t} &= i g_{2S}^*(q^-) E_L^-, \\ \frac{\partial q^+}{\partial t} &= -\Gamma q^+ + i[g_{1S}^*(E_S^+) E_L^+] + Q^{1/2} F^+(z, t), \\ \frac{\partial q^-}{\partial t} &= -\Gamma q^- + i[g_{1S}^*(E_S^-) E_L^-] + Q^{1/2} F^-(z, t), \end{aligned} \quad (1)$$

where $g_{1S} = \beta(\omega_S)/\hbar$; $g_{2S} = 2\pi\hbar\omega_S g_{1S} N_0$; $\beta(\omega_S)$ is the susceptibility at the Stokes frequency; N_0 is the density of scattering particles; $u_i = c/n_i$ is the group velocity of the i th wave, which is assumed to coincide with the phase one ($i = L, S$); $D_i = (1/2)\partial^2 k_i / \partial \omega_i^2$ are the parameters that determine the group velocity dispersion of the pump and the harmonic in the nonlinear crystal and the dielectric; $\Gamma = 1/T_2$ is the SRS linewidth; T_2 is the dephasing time; F^{\pm} are the normalised statistically independent Gaussian sources of polarisation fluctuations [13]; and Q is the intensity of polarisation fluctuations. As in Ref. [13], we assume that $Q = 2\Gamma/SN_0$, where S is the cross section of the excited volume of the scattering medium.

We solved the system of equations (1) using the method of characteristics for partial first-order differential equations similarly to the algorithm that we earlier developed in Refs [10–12] for calculating the SRS process. We introduced dimensionless variables $tc/\lambda \rightarrow t$ and $z/\lambda \rightarrow z$ (where λ is the pump wavelength) and replaced the linear operator $-iD\partial^2/\partial t^2$ in the Fourier space by the scalar multiplication by $\exp(-iK^2\Delta z/2)$ (where $K = 2\pi/N\Delta z$, N was the Fourier-transform parameter corresponding to the number of points on the frequency grid, and Δz was the dimensionless spatial step). The calculation algorithm followed the general scheme of Ref. [7].

The initial conditions for the waves and the polarisation $q^{\pm}(z, t)$ were specified as

$$\begin{aligned} E_L(z=0, t) &= E_{L0} \exp\{-2 \ln[2((t-t_0)/\tau_L)^2]\}, \\ q^{\pm}(z, t=0) &= 0, \end{aligned}$$

where τ_L is the FWHM duration of the pump pulse and t_0 is the time origin. The boundary conditions for the transmitted and reflected waves were obtained by applying the FFT method to the pump and Stokes waves [7].

As the radiation parameters, we took typical characteristics of a 100-fs 1060-nm neodymium-glass laser. The thickness of the nonlinear layer was l_1 (in units of λ); its other parameters coincided with those of a Nd:KGW crystal: $n_{1L} = 1.968$, $n_{1S} = 1.977$, $T_2 = 5.6$ ps, $N_0 = 4.5 \times 10^{22} \text{ cm}^{-3}$, $\beta(\omega_S) = 2 \times 10^{-25} \text{ cm}^3 \text{ erg}^{-1}$ [10], and $\omega_L/\omega_S = 1.1$. The thickness of the linear layer was l_2 ; its refractive indices coincided with those of silica: $n_{2L} = 1.467$ and $n_{2S} = 1.45$. The total length of the nonlinear medium was $l \approx 0.25\lambda j$, where j was the number of active-medium layers; the number of the structure periods $N_{\text{str}} = 2j$ was varied between 4 and 80. We normalised the intensities of the pump and Stokes waves by 1 GW cm^{-2} .

The nonlinear SRS-amplification coefficient can be estimated from the expression [14, 15]

$$G = \left(\frac{4g_0 J_L \tau_L}{T_2} \right)^{1/2}, \quad (2)$$

where $g_0 = 4\pi g_{1S} g_{2S} / \Gamma c^2$ is the SRS-amplification coefficient equal to 6 cm GW^{-1} , and J_L is the pump intensity. The frequency conversion of femtosecond pulses is a nonstationary process when $\tau_L/T_2 \ll G$. In typical crystals, the transverse relaxation times are of the order of few picoseconds, and this inequality is therefore satisfied. The level of the spontaneous noise in equations (1) was determined by the intensity Q . In the case of the crystal layer, all parameters that define Q are specified, and one can only vary the beam cross section S between 10^{-2} cm^2 and 10^{-6} cm^2 in experiments. In our calculations, the cross section of the beam focused on the crystal surface was 10^{-4} cm^2 .

The earlier analysis of the SRS-conversion efficiency in the nonstationary mode [14–17] has shown that the intensity of the first Stokes component can exceed the intensity of the spontaneous Raman scattering only if $G \geq 1$. To achieve such a high coefficient G in a scattering medium whose length l amounts to only five wavelengths (20 periods of the photonic crystal) the pump intensity must be $J_L = 400 \text{ GW cm}^{-2}$. The calculations were performed for this particular pump intensity. Under these conditions in a nonperiodic scattering medium, the Stokes component is only a few times more intense than the noise radiation. Thus, the increase in the SRS-conversion efficiency in a photonic crystal is due to the interference, which significantly changes the density of states of the pump and Stokes fields.

3. Calculation results

The reflection and transmission spectra of bounded one-dimensional periodic structures were calculated using the method of the transition matrix [18], as applied to an arbitrary layered structure. In this method, one calculates the general characteristic matrix $b^{s,p}$ of the structure from the matrices $M_j^{s,p}$ of individual layers:

$$\begin{aligned} b^{s,p} &= \prod_{j=1}^{N_{\text{str}}} M_j^{s,p}, \\ M_j^{s,p} &= \begin{pmatrix} \cos \phi_j & -i/q_j^{s,p} \sin \phi_j \\ -i/q_j^{s,p} \sin \phi_j & \cos \phi_j \end{pmatrix}, \end{aligned}$$

$$\phi_j = \frac{\omega}{c} l_j (n_j^2 - \alpha^2)^{1/2}, \quad q_j^s = (n_j^2 - \alpha^2)^{1/2},$$

$$q_j^p = -\frac{(n_j^2 - \alpha^2)^{1/2}}{n_j^2}, \quad \alpha = n_i \sin \theta_i,$$

where n_j and l_j are the refractive index and the thickness of the j th layer, respectively (in the calculations, we used the above-given values of refractive indices for the linear and nonlinear media at the pump and Stokes wave frequencies); n_i is the refractive index of the homogeneous medium from which the radiation enters the structure; θ_i is the angle of incidence; and indices s and p refer to the waves whose polarisation is respectively parallel and perpendicular to the incidence plane. Given the general matrix of the structure, we can find the amplitude coefficients of reflection $r^{s,p}$ and transmission $t^{s,p}$:

$$r^s = \frac{b_{11}^s n_i \cos \theta_i + b_{12}^s n_i \cos \theta_i n_i \cos \theta_i - b_{21}^s - b_{22}^s n_i \cos \theta_i}{b_{11}^s n_i \cos \theta_i + b_{12}^s n_i \cos \theta_i n_i \cos \theta_i + b_{21}^s + b_{22}^s n_i \cos \theta_i},$$

$$t^s = \frac{2n_i \cos \theta_i}{b_{11}^s n_i \cos \theta_i + b_{12}^s n_i \cos \theta_i n_i \cos \theta_i + b_{21}^s + b_{22}^s n_i \cos \theta_i}, \quad (3)$$

$$r^p = \frac{b_{11}^p n_i \cos \theta_i + b_{12}^p \cos \theta_i \cos \theta_i - b_{21}^p n_i n_i - b_{22}^p n_i \cos \theta_i}{b_{11}^p n_i \cos \theta_i + b_{12}^p \cos \theta_i \cos \theta_i + b_{21}^p n_i n_i + b_{22}^p n_i \cos \theta_i},$$

$$t^p = \frac{2n_i \cos \theta_i}{b_{11}^p n_i \cos \theta_i + b_{12}^p \cos \theta_i \cos \theta_i + b_{21}^p n_i n_i + b_{22}^p n_i \cos \theta_i}.$$

Here, θ_i is the angle between the surface normal and the radiation beam emerging from a photonic crystal and entering the medium with the refractive index n_i ; b_{mm}^s and b_{mm}^p are elements of the characteristic matrix $b^{s,p}$. Knowing the amplitude coefficients (3), we can calculate the energy coefficients of reflection and transmission:

$$R^{s,p} = |r^{s,p}|^2, \quad (4)$$

$$T^{s,p} = \frac{n_i \cos \theta_i}{n_i \cos \theta_i} |t^{s,p}|^2.$$

It follows from equations (3) that we can change the spectral characteristics of the periodic structure by varying the thickness of the layers, their number, and the refractive indices. Using formulas (4), we calculated the intensity reflection and transmission, $R_{L,S}$ and $T_{L,S}$, at the pump and Stokes frequencies for the case when the pump wave is normally incident on the surface of the photonic crystal.

Fig. 1 shows the reflection spectra of laser and Stokes radiation for three different periodic structures. In the first of them, the pump frequency lies inside the bandgap (maximum reflection), and the Stokes frequency lies at its first sideband peak (Fig. 1a). In the second, the laser frequency is between the second and third-order bandgaps, while the Stokes frequency is to the left of the second-order bandgap (Fig. 1b). In the third, the laser and Stokes frequencies lie between the first and second bandgaps, spanning a few sidebands (Fig. 1c).

The transmission spectra can be obtained by subtracting the functions shown in Fig. 1 from unity. Note that the bandgap structure of the photonic crystal is formed at $N_{\text{str}} \geq 10$; the further increase in the structure thickness

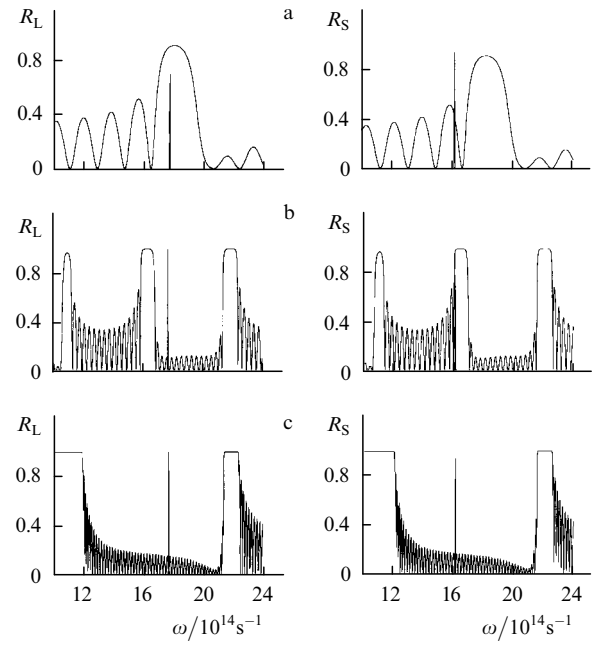


Figure 1. Reflection spectra of periodic structures with parameters $l_1 = 0.21$, $l_2 = 0.06$, $N_{\text{str}} = 10$ (a); $l_1 = l_2 = 0.5$, $N_{\text{str}} = 16$ (b); and $l_1 = l_2 = 0.25$, $N_{\text{str}} = 55$ (c) for the pump (left) and Stokes (right) components.

only insignificantly narrows the bandgaps and increases the number of sideband transmission resonances [5].

Solving the system of equations (1) for the periodic structures whose reflection spectra are shown in Fig. 1, we obtained the intensities $J_{L,S}^{\pm} \sim |E_{L,S}^{\pm}|^2$ and energies $W_{L,S}^{\pm} = \int J_{L,S}^{\pm} dt$ of the first Stokes component copropagating with the reflected and transmitted pump waves, and the same parameters for the pump waves. We did not study the statistical properties of the radiation, which arose due to the quantum polarisation noise. However, the preliminary calculations have shown that different sources F^{\pm} lead to different energies of the transmitted and reflected Stokes waves.

Figs 2 and 3b show the energy W_S^{\pm} of the Stokes component as a function of the number N_{str} of periods contained in the structure. Figs 3a and 3c show the energy of the transmitted and reflected pump pulses and the variation in the Stokes pulse duration. Analysing Figs 1a and 2a, we see that, in a crystal with such spectral characteristics, the energy of the transmitted Stokes radiation is 2.4 times greater than that of the reflected radiation; the ratio of the intensities of the linear spectra at the Stokes frequency is the same. The intensity of the reflected pump is 1.1 times greater than the intensity of the transmitted radiation, which amounts to 0.17 of the initial pump intensity. One can see from Fig. 2a that the Stokes radiation intensity falls off at both higher and lower values of N_{str} .

The energy of the Stokes pulse is significantly higher in the case when the photonic crystal has multiple bandgaps, the corresponding linear reflection spectra being shown in Figs 1b and 1c. One can see from Fig. 2b that the maximum intensity of the reflected Stokes wave is reached for a 16-layer structure with $l_1 = l_2 = 0.5$; this intensity is 4.9 times greater than the intensity of the transmitted wave. We obtained virtually the same parameters of the Stokes wave for the case when $l_1 = l_2 = 0.25$ and $N_{\text{str}} = 55$, although the nonlinear interaction length in Fig. 3 was 1.75 times greater than for

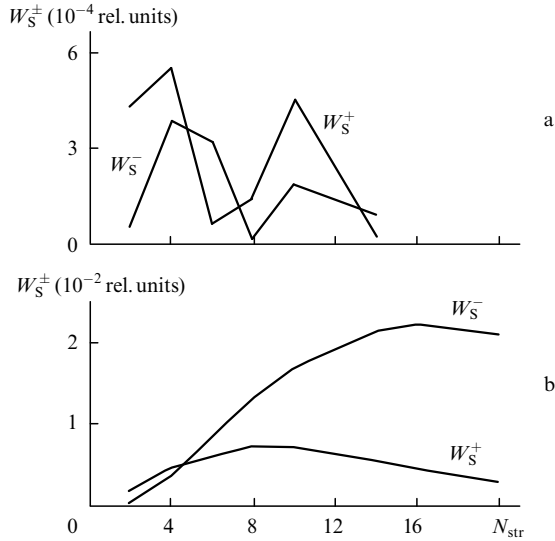


Figure 2. Energy W_S^{\pm} of the Stokes pulses copropagating with the transmitted and reflected pump waves as a function of the number N_{str} of periods contained in the structure; $l_1 = 0.21$, $l_2 = 0.06$ (a) and $l_1 = l_2 = 0.5$ (b).

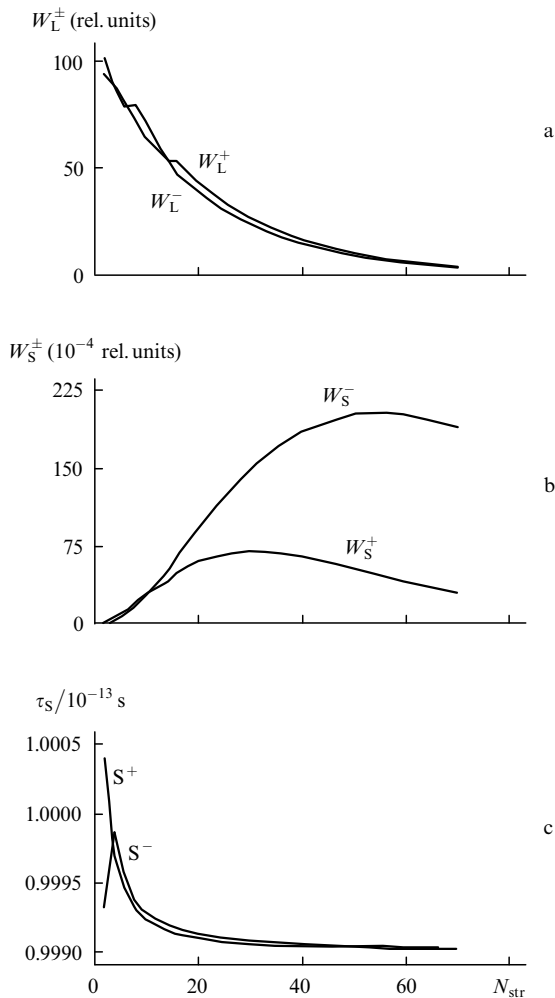


Figure 3. Energy of the transmitted and reflected pump (a) and Stokes (b) waves and the SRS pulse duration τ_S (c) as functions of the number N_{str} of periods.

the one in Fig. 2b. Fig. 4 shows the envelopes of the transmitted and reflected Stokes pulses in the case $l_1 = l_2 = 0.25$. One can see from the figures that the intensity of the reflected wave and, accordingly, the ratio of the reflected intensity to the incident one increase with increasing number of periods in the structure, while the pulse duration remains virtually unchanged. Comparison of the presented calculation results suggests that SRS is primarily affected by the interference properties of the periodic structure.

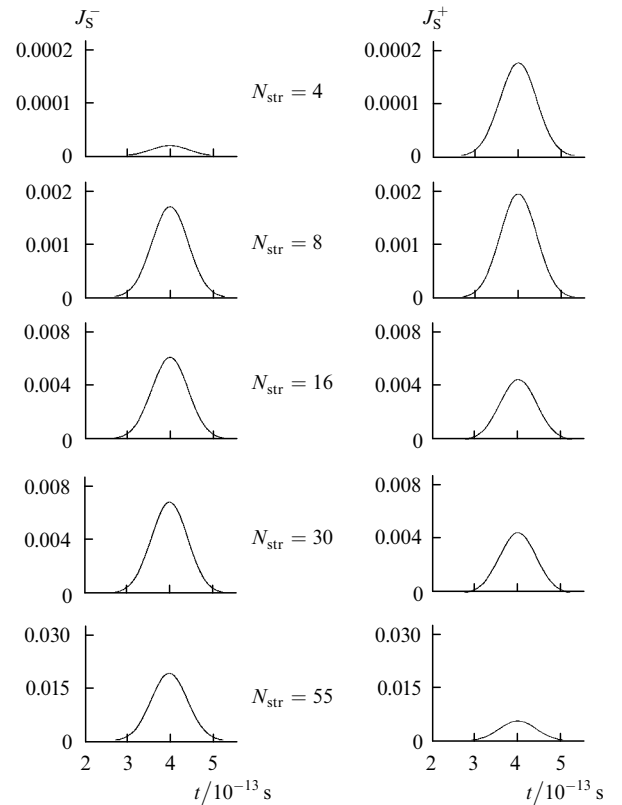


Figure 4. SRS pulses copropagating and counterpropagating with respect to the pump pulse, for different numbers N_{str}

Comparing the density of electromagnetic field states in a photonic crystal with the density of charge-carrier states in semiconductors, we can see why SRS is more efficient in the case when the Stokes frequency lies between the first- and second-order bandgaps (see Figs 1b and 1c). The state density is proportional, first, to the square root of the difference between the Stokes frequency ω_S and the central bandgap frequency ω_g , and, second, to the number of equivalent minima between the bandgaps. When the Stokes frequency lies near the second bandgap, the amplification at the Stokes frequency is enhanced because the difference $\omega_S - \omega_g$ reaches its maximum (Fig. 1b). As for the situation shown in Fig. 1c, the increase in the density of states is caused by the increased number of minima between the first- and second-order bandgaps.

Consider the efficiency of using the periodic structure to convert the frequency of femtosecond pulses in the SRS process. The nonstationary SRS-amplification coefficient calculated according to formula (2) gives $G = 1.36$ for $N_{\text{str}} = 16$ and $J_L = 400 \text{ GW cm}^{-2}$. Therefore, we cannot obtain the Stokes intensity that is more than an order of magnitude

greater than the noise radiation, even if we increase the structure period by a factor of 3.

Estimating the nonstationary SRS-conversion coefficient with the aid of formula $G_\tau = \ln |J_S/J_0|$ (where J_0 is the intensity of the spontaneous noise radiation proportional to Q) [14, 15], we obtain $G_\tau = 9$ in the case of the most efficient conversion (Fig. 2a). This is 7.8 times greater than the value of G given by formula (2). Since the gain is proportional to e^G , this periodic structure provides SRS that is 2000 times more intense than the spontaneous scattering. The total efficiency of the SRS conversion does not exceed 10^{-4} . In this particular scattering medium, the SRS-conversion efficiency grows only insignificantly with increasing pump energy since the energy of the transmitted and reflected pump pulses also grows in this case.

4. Conclusions

We have calculated the SRS frequency conversion of femtosecond pulses in a periodic structure made of a nonlinear Nd:KGW crystal and fused silica. The calculations were performed for three different positions of the pump and Stokes frequencies with respect to the photonic crystal bandgap. They have shown that the efficiency of the pump wave conversion is mainly affected by the spectral properties of the structure.

Comparing the results obtained, we have shown that the SRS-conversion is more efficient in the case of a complex bandgap structure that increases the density of electromagnetic field states of both the pump and Stokes radiation. When the Stokes frequency lies on the bandgap edge, the energy of the reflected wave is approximately five times greater than that of the transmitted one, whereas the duration of Stokes pulses is only insignificantly smaller than the duration of pump pulses. The pump wave is 'trapped' inside the crystal in this case.

Comparing the nonstationary SRS-amplification coefficients of the periodic structure and the scattering medium that has the same interaction length, we found that the amplification coefficient is 7.8 times greater in the former case. Therefore, by selecting the appropriate scattering medium and a dielectric for the periodic structure of a photonic crystal, we can control the frequency conversion of femtosecond pulses in the SRS process.

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