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Coherent Anti-Stokes Raman Scattering of Slow Light in a Hollow Planar Periodically Corrugated Waveguide

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Enhancement of coherent anti-Stokes Raman scattering (CARS) by molecular nitrogen in a hollow planar periodically corrugated waveguide is experimentally detected. The measured dependence of CARS efficiency on the thickness of the waveguide layer indicates that CARS enhancement under these conditions is at least partially due to the decrease in the group velocity of pump pulses around the photonic band gap. *© 2002 MAIK "Nauka/Interperiodica".*

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Remarkable properties of structures with a periodically modulated refractive index are opening ways to modify and control the dispersion of optical materials [1, 2]. Periodic and quasi-periodic structures, such as multilayer mirrors, microstructure fibers, as well as two- and three-dimensional periodic arrays generally referred to as photonic crystals [2, 3], are now actively used to shape, transfer, and control ultrashort light pulses [4, 5], as well as to create optical fibers of new types [6–8] and to develop optical switches, couplers, filters, and other optical components [2]. Structures with a periodically modulated refractive index often allow nonlinear-optical interactions to be phase-matched [9–12]. Physically, phase matching in photonic band-gap (PBG) structures is based on the generalized momentum conservation [9, 10] involving the reciprocal lattice vector of a periodic structure. Waveguides with a periodically perturbed refractive index offer a convenient way of extending this approach to large lengths of nonlinear-optical interaction [13], providing more degrees of freedom in reducing the phase mismatch of light pulses involved in a nonlinear-optical process.

In this paper, we will demonstrate the possibility to considerably increase the efficiency of four-wave mixing (FWM) in a gas medium filling a hollow planar corrugated waveguide. The main difference of our experimental approach from the methods used in earlier nonlinear-optical experiments in PBG waveguides (see, e.g., [13]) is that a gas filling the waveguide layer of a hollow planar waveguide serves as a nonlinear medium in our experiments. The coherence length under these conditions considerably exceeds the waveguide length (which was on the order of several centimeters in our studies). Four-wave mixing efficiency can be improved in such a situation, as will be shown below, due to the field enhancement in a PBG waveguide related to group-velocity lowering for one or several pump waves around the PBG edge.

To experimentally implement the main idea of this work, we employed a waveguide structure consisting of a mirror and a diffraction grating. Both optical elements forming this waveguide were aluminum-coated. As shown in our previous studies [14, 15], such a structure integrates a hollow waveguide and a one-dimensional photonic crystal, combining the advantages of both of these optical components. On the one hand, high-power laser radiation can be coupled into such a waveguide, allowing ultrashort pulses to be produced due to self-phase modulation and high-order stimulated Raman scattering using the approaches similar to those developed in [16, 17] and permitting high-order harmonic generation experiments [18]. Similar to gas-filled hollow fibers, the waveguide regime of nonlinear-optical interactions in our structure improves the efficiency of wave-mixing and harmonic-generation processes relative to the regime of tightly focused pump beams due to a radical increase in the interaction length. On the other hand, a periodic perturbation of the refractive index introduced by the diffraction grating gives rise to photonic band gaps (Fig. 1), which substantially change the dispersion properties of light fields with respect to the case of a gas medium in a conventional gas cell or waveguide modes in a gas-filled hollow fiber. The created waveguide opens new ways of phase and group-velocity matching in nonlinear-optical interactions through the independent control of three main dispersion components: material dispersion, dispersion of waveguide modes, and dispersion of a periodic structure. The material dispersion of the gas filling the waveguide can be varied by changing the gas composition and the gas pressure. The waveguide dispersion can be changed by varying the thickness of the waveguiding layer and by choosing appropriate materials for waveguide walls and a set of waveguide modes involved in the nonlinear-optical process. Finally, the period and the profile of the grating are the main knobs to control the dispersion of the periodic structure.

Our analysis of the dispersion of light waves involved in nonlinear-optical interactions in a hollow planar PBG waveguide was based on the equations of coupled-mode theory [1]. The use of this approach allowed us to find complex propagation constants for a hollow planar PBG waveguide by searching for the eigenvalues of $2N \times 2N$ square matrices of the relevant characteristic equations, where N is the number of modes of an unperturbed planar waveguide (a waveguide with no corrugation), determined from the cut-off condition. The inset in Fig. 1 presents the spectral dependences of the transmission coefficient and the effective refractive index for the lowest order bulk mode TM_2 of the hollow planar PBG waveguide calculated with the use of coupled-mode equations. The photonic band gap arises in this spectral region, as follows from the results of our calculations, due to the strong coupling of the lowest order bulk mode TM_2 with surface plasmon modes TM_0 and TM_1. The results of these calculations, as can be seen from Fig. 1, qualitatively agree with the experimental data. Our method of calculations allows the position of the photonic band gap in the transmission spectrum of a hollow planar PBG waveguide to be reproduced with reasonable accuracy (Fig. 1). However, the absolute values of the transmission coefficient, effective refractive index, and group velocity obtained with the use of the above-described approach can be considered as very rough estimates only, since these absolute values are highly sensitive to the coupling coefficients, which are not known with sufficient accuracy. These coupling coefficients depend on the Fourier amplitudes of the periodic profile of the grating, as well as on the spatial overlapping of light fields in waveguide modes and the area of a perturbed dielectric function.

The main purpose of our experimental studies was to demonstrate the possibility of enhancing coherent anti-Stokes Raman scattering (CARS) using the created hollow PBG waveguide. Since a gas filling the waveguide layer between the grating and the mirror serves as a nonlinear medium in our experiments and the pressure of this gas never exceeded the atmospheric pressure, the coherence length for CARS-type four-wave mixing processes should considerably exceed the waveguide length, which was typically on the order of several centimeters in our experiments. The efficiency of FWM processes can be increased under these conditions due to field-enhancement effects, which are characteristic of PBG structures and which are related to a decrease in group velocities of light pulses around photonic band gaps in such structures. This effect is illustrated in Fig. 2, which displays the wavelength dependence of the group index for TM_2 and TM_3 modes in a hollow planar PBG waveguide with a mirror–grating gap $2a = 22$ µm within the spectral range corresponding to the second harmonic of a Nd:YAG-laser radiation. The solid and dashed lines in Fig. 2 represent the group indices for the TM_2 and TM_3 modes, respectively. The

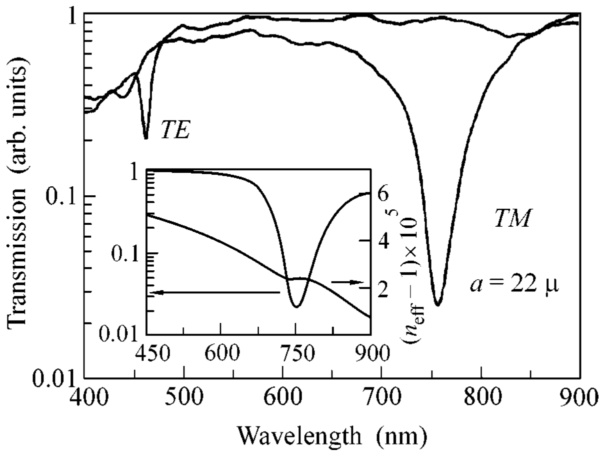

Fig. 1. Transmission spectra measured for *TM* and *TE* modes of a planar corrugated hollow waveguide consisting of a 2400-grooves/mm aluminum-coated grating and an aluminum mirror with $2a = 44$ µm. The inset shows the results of calculations for the transmission spectrum and the spectral dependence of the effective refractive index for the TM_2 mode of this waveguide.

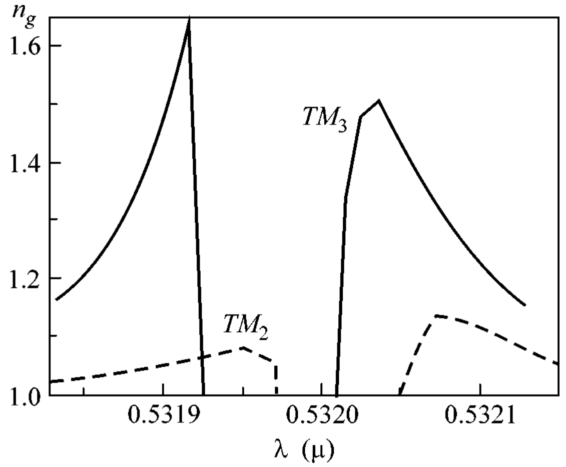

Fig. 2. The spectral dependence of the group index $n_g = c/v_g$ (c is the speed of light in vacuum and v_g is the group velocity in the waveguide structure) for the TM_2 (solid line) and TM_3 (dashed line) modes of a hollow planar corrugated waveguide with a 1200-grooves/mm diffraction grating and $2a = 22$ µm.

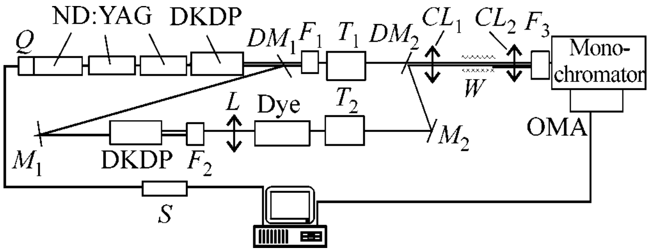

Fig. 3. Diagram of the experimental setup for studying coherent anti-Stokes Raman scattering in a hollow planar corrugated waveguide: M_1, M_2, rotating mirrors; DM_1, DM_2, dichroic mirrors; F_1–F_3, sets of optical filters; OMA, optical multichannel analyzer; *S*, synchronization unit; CL_1, CL_2, cylindrical lenses; *L*, spherical lens; T_1, T_2, telescopes; and *W*, waveguide.

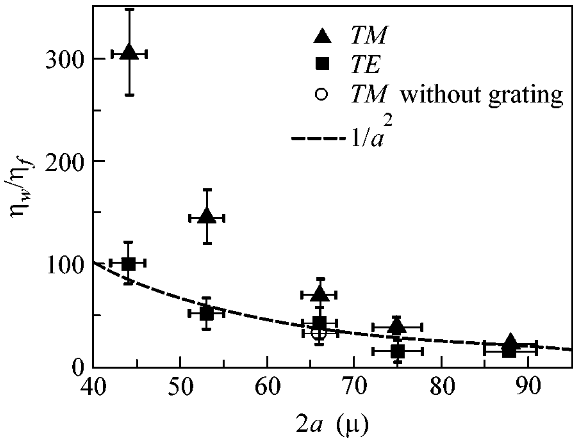

Fig. 4. The ratio of the efficiency *w* of the CARS process in a hollow planar PBG waveguide to the efficiency η_f of the CARS process with the same energies of pump pulses, but for cylindrically focused pump beams in the absence of a waveguide, as a function of the distance $2a$ between the waveguide walls. The CARS signal is related to Raman-active transitions of molecular nitrogen in atmospheric-pressure air. Triangles show the CARS enhancement ratio for *TM* waveguide modes, while the squares correspond to *TE* modes. The circle shows the CARS enhancement ratio for *TM*-polarized radiation in a waveguide with a second mirror instead of the diffraction grating (an unperturbed waveguide). The dashed line represents the scaling law $1/a^2$, which describes the increase in the efficiency of the CARS process in a planar waveguide with no corrugation relative to the efficiency of the same CARS process in cylindrically focused beams due to purely geometrical factors.

group velocity of light pulses, as can be seen from the results presented in Fig. 2, decreases considerably in this spectral range due to the PBG effect. This increases the mean density flux of electromagnetic radiation in the waveguide, leading to the enhancement of nonlinear-optical processes.

A diagram of the experimental setup employed to investigate the CARS process in a gas medium filling a hollow planar PBG waveguide is shown in Fig. 3. We studied a two-color CARS process leading to the generation of a signal at the frequency $\omega_{CARS} = 2\omega_1 - \omega_2$, where ω_{CARS} is the frequency of the CARS signal and ω_1 and ω_2 are the frequencies of the pump waves. A *Q*-switched Nd:YAG laser, generating 15-ns pulses of 1.064-μm radiation, was employed as a master oscillator. The laser pulses produced by this oscillator were amplified up to about 30 mJ in two Nd:YAG amplification stages. The fundamental radiation was then converted into the second harmonic using a DKDP crystal. The second harmonic produced in this crystal served as one of the pump beams in the CARS process (the frequency ω_1). Fundamental radiation that remained frequency-unconverted at the output of the DKDP crystal was separated from the second harmonic with a dichroic mirror DM_1 and was employed to generate the second harmonic in the second DKDP crystal. This second-harmonic beam was then used to pump a sulforhodamine 101 dye laser. Dye-laser radiation served as the second pump beam in the CARS process (the frequency ω_2). The pump beams with the frequencies ω_1 and ω_2 were brought into spatial coincidence with a dichroic mirror DM_2 and were coupled into a hollow planar corrugated waveguide by a cylindrical lens CL_1 with a focal length of 9 cm. The energy of the second-harmonic pulse was 8 mJ, while the energy of dye-laser radiation was equal to 0.8 mJ. Aluminum-coated mirrors and 1200- and 2400-grooves/mm aluminum-coated diffraction gratings were used to create a hollow waveguide. The length of the waveguides used in our experiments was equal to 5 cm. The distance between the waveguide walls was varied from 22 up to 88 μm.

The frequency ω_2 of dye-laser radiation was chosen in such a way as to satisfy the condition of Raman resonance $\omega_1 - \omega_2 = \Omega$ with a Raman-active transition of molecular nitrogen with $\Omega = 2331$ cm^{-1}. This condition was met with the wavelength of dye-laser radiation equal to 0.607 μm. The wavelength of the CARS signal related to molecular nitrogen in the atmospheric-pressure air filling the hollow PBG waveguide was then equal to 0.473 μm. This signal was collimated with a cylindrical lens CL_2 and separated from the pump beams with a set of optical filters. Then, we let the CARS signal pass through a monochromator and detected the signal at the output of the monochromator with the use of an optical multichannel analyzer.

To characterize the enhancement of the CARS process involving Raman-active transitions of molecular nitrogen in atmospheric-pressure air in a hollow planar PBG waveguide, we compared the efficiency η_w of this process in the waveguide with the efficiency η_f of the same process with the same energies of pump pulses, but in the regime of cylindrical focusing of pump beams in the absence of a waveguide. Figure 4 presents the CARS enhancement ratio η_w/η_f measured as a function of the distance $2a$ between the mirror and the 1200-grooves/mm grating, forming a planar PBG waveguide.

Triangles show this ratio for *TM* waveguide modes, while the squares correspond to *TE* modes. The circle shows the CARS enhancement ratio for *TM*-polarized radiation in a waveguide with a second mirror instead of the diffraction grating (an unperturbed waveguide). A planar waveguide obviously provides an increase in the efficiency of any FWM process relative to the efficiency of the same FWM process in cylindrically focused beams due to geometrical factors. In contrast to FWM processes in gas-filled hollow fibers, when this geometrical enhancement ratio scales as $1/a^4$, where d is the fiber inner diameter [19, 20], a planar waveguide provides FWM enhancement scaling as $1/a^2$. This scaling law of FWM enhancement due to purely geometric factors is shown by the dashed line in Fig. 4.

The enhancement of the CARS process in the case of *TE* modes in our experiments virtually coincided, as can be seen from the data presented in Fig. 4, with the enhancement attainable in the waveguide regime due to purely geometric factors. A much higher CARS enhancement ratio, as is seen from Fig. 4, can be achieved for *TM* modes of a planar PBG waveguide, when the maximum enhancement ratio relative to the case of cylindrically focused beams may be as high as 300. These higher values of CARS enhancement ratios attainable for *TM* modes are due to the fact that the frequencies ω_1 and ω_2 of the second harmonic and dye-laser radiation fall within the range of strong coupling between the lowest order bulk mode TM_2 and one of the higher order *TM* modes. The electromagnetic energy density in the waveguide increases under these conditions, which leads to the enhancement of nonlinear-optical processes.

Since the CARS signal intensity is proportional to the product of group indices of pump fields, the decrease in the group velocity of the second-harmonic field by a factor of 1.2–1.4 and the lowering of the group velocity of dye-laser radiation by a factor of 1.1–1.2 result in the enhancement of the CARS process by a factor of 1.6–2.4. Although these estimates are, of course, very rough since the group velocities of pump waves are known only approximately, they indicate that the improvement in CARS efficiency achieved in our PBG-waveguide experiments in the case of *TM* modes may be, at least partially, attributed to the decrease in the group velocities of pump fields. Quantitative discrepancies between the experimental values of the CARS enhancement ratio and the above estimates for this ratio may be indicative of other physical factors leading to FWM enhancement under conditions of our experiments. One of these factors may be related to local field enhancement in plasmon *TM* modes, which increases the efficiency of nonlinear-optical wave mixing, leading to energy transfer to the waveguide modes that provide the dominant contribution to the FWM process. Another group of factors includes effects changing the material component of dispersion, e.g., the excitation and ionization of the gas medium filling the waveguide.

The results of experimental and theoretical studies presented in this paper demonstrate the possibility of a substantial enhancement of four-wave mixing processes in a gas medium in a hollow planar corrugated waveguide due to field enhancement effects related to the decrease in the group velocity of one or several pump fields around the photonic band gap. The enhancement of the CARS process achieved in our experiments, performed with a planar waveguide structure consisting of a metal mirror and a diffraction grating, can be considerably increased by optimizing the parameters and the geometry of the waveguide for a specific set of waveguide modes involved in a nonlinear-optical process. The method of enhancement of nonlinear-optical processes demonstrated in this paper opens new possibilities for improving the sensitivity of nonlinear-optical gas-phase analysis, promoting ultrashort-pulse formation with the use of self-phase modulation and high-order stimulated Raman scattering, and increasing the efficiency of high-order harmonic generation and wave mixing in gas-filled hollow waveguides.

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