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Bragg resonance-enhanced coherent anti-Stokes Raman scattering in a planar photonic band-gap waveguide

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A planar periodically corrugated waveguide, consisting of a grating and a mirror and integrating a hollow waveguide and a photonic band-gap structure into a single compact optical element, is shown to enhance coherent anti-Stokes Raman scattering (CARS) in molecular nitrogen. Cross-correlation measurements, intended to visualize group-delay effects in the photonic band-gap waveguide and performed with picosecond Nd : YAG laser pulses, and also the measured dependence of CARS efficiency on the thickness of the waveguide layer, indicate 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. Copyright © 2002 John Wiley & Sons, Ltd.

INTRODUCTION

Structures with a spatially modulated refractive index open up ways to modify, control and engineer the dispersion of optical materials,[1] offering, in particular, new solutions to the problems of non-linear optics and spectroscopy.[2,3] Such structures often allow non-linear optical interactions to be phase- and group-velocity matched.[4–12] The idea of using the dispersion of structures with a spatially periodic modulation of the refractive index to phase match non-linear optical interactions has been discussed since the pioneering work of Bloembergen and Sievers.[4] Physically, this approach to phase matching rests on the generalized momentum conservation for periodic structures,[5] which allows a Bragg resonance to be achieved for a non-linear optical wave-mixing process in a situation when the mismatch of the wavevectors of light beams involved in the non-linear interaction becomes equal to a multiple of the reciprocal lattice vector. It is clear from this condition that the period of the structure should be of the order of the optical wavelength to make such phase matching possible, which implies that photonic band gaps (PBGs), arising from the strong coupling of forward and backward waves, become essential for the propagation and phase matching of light pulses. This relation between the spatial period of the structure and the optical wavelength is also important in understanding the difference between phase-matched non-linear optical processes in PBG structures and quasi-phase-matched wave mixing in periodically poled materials.[13–15] Quasi-phase matching implies that the phase mismatch is compensated by a jumpwise phase shift, introduced through the inversion of the sign of the non-linear optical susceptibility, while PBG structures allow the phase mismatch to be compensated on the spatial scale of the order of optical wavelength.

Analysis of band-edge phase matching in one-dimensional PBG structures[6–8] shows that such structures offer several remarkable opportunities. In particular, owing to local-field enhancement effects occurring within a limited spectral range, the efficiency of second-harmonic generation in such structures grows as a function of the non-linear interaction length even faster than in the case of perfect phase matching.[6,8] Previous experimental results on multilayer samples are very encouraging,[9–11] showing that, in many important situations, PBG structures allow the

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phase-matching problem to be solved in an efficient and simple fashion. As shown by Berger,[12] PBG materials also offer a very elegant way of extending the concept of phase matching to two dimensions.

Waveguide regimes of non-linear optical interactions in PBG structures add more physical aspects to the phase-matching problem, as the dispersion of waveguide modes provides more degrees of freedom in reducing the phase mismatch of light pulses involved in the non-linear optical process. In particular, the integration of a PBG structure with a hollow waveguide, attainable in a planar corrugated waveguide consisting of a pair of gratings or a grating and a mirror,[16] allows the efficiency of four-wave mixing in gases filling the waveguide to be improved[17] and opens up new horizons in short-pulse generation and frequency conversion. Although the opportunity of using waveguides with a periodically modulated refractive index to phase-match non-linear optical processes was elucidated a long time ago,[18] recent achievements in material processing technology[19] and a rapid progress with laser sources, permitting shorter and shorter pulses to be generated, as well as recent advances in the theory of non-linear optical interactions in waveguides with a periodically modulated refractive index,[20,21] make such waveguides very promising for the creation of compact and efficient elements for the frequency conversion of low-energy laser pulses.

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

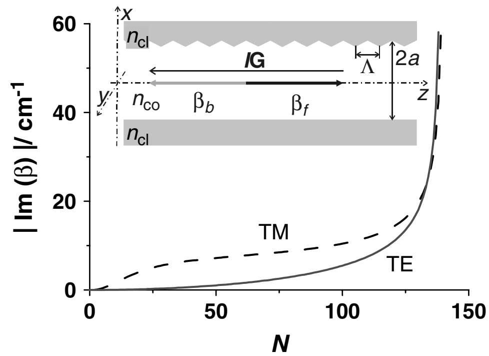

Figure 1. The imaginary part of the propagation constant β for a hollow planar waveguide with aluminum-coated walls as a function of the mode index N for $2a = 44$ µm and $\lambda = 0.64$ µm. The inset shows a mirror and diffraction grating combined to form a hollow planar corrugated waveguide: Λ is the period of the grating, $2a$ is the separation between the grating and the mirror, n_{co} is the refractive index of the waveguiding layer, and n_{cl} is the refractive index of the cladding.

COUPLED MODES IN A HOLLOW PLANAR PBG WAVEGUIDE

The waveguide structure and the general idea

To implement experimentally the main idea of this work, we employed a waveguide structure consisting of a mirror and a diffraction grating (inset in Fig. 1). Both optical elements forming this waveguide were aluminum coated. As shown in our previous studies,[16,17] such a structure integrates a hollow waveguide and a one-dimensional photonic crystal, combining the advantages of these optical elements. 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 with the use of the approaches similar to those developed in earlier studies,[22,23] permitting high-order harmonic generation experiments,[24] and opening up ways to improve the sensitivity of non-linear optical methods of gas-phase analysis.[25,26] Similarly to gas-filled hollow fibers, the waveguide regime of non-linear optical interactions in our structure improves the efficiency of wave-mixing and harmonic-generation processes relative to the regime of tightly focused pump beams owing to a radical increase in the interaction length. On the other hand, a periodic perturbation of the refractive index introduced by the diffraction grating (see the inset in Fig. 1) gives rise to photonic band gaps, 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 up new ways of phase and group-velocity matching in non-linear optical interactions through 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 non-linear optical process. Finally, the period and the profile of the grating are the main means of controlling the dispersion of the periodic structure.

The basic relations

Our analysis of the dispersion of light waves involved in non-linear optical interactions in a hollow planar PBG waveguide was based on coupled-mode equations.[27] We assumed that waveguide modes propagate along the z-direction, and the diffraction grating introduces a small spatially periodic perturbation $\Delta\varepsilon(x, y, z)$ of the dielectric function near one of the waveguide walls. The total dielectric function is then written as

$$\varepsilon(x, y, z) = \varepsilon_0(x, y) + \Delta\varepsilon(x, y, z) \quad (1)$$

where $\varepsilon_0(x, y)$ is the unperturbed part of the dielectric function, which defines the waveguide structure.

In accordance with the general approach of the theory of coupled modes,[27] the light field in a perturbed waveguide is represented as a linear combination of the eigenmodes of the unperturbed waveguide:

$$E = \sum A_m(z)E_m(x, y)\mathrm{e}^{i(\omega t - \beta_m z)} \quad (2)$$

where ω is the radiation frequency, β_m is the propagation constant of the mth mode of the unperturbed waveguide and $E_m(x, y)$ is the field distribution in the (x, y) plane in the mth waveguide mode. The amplitude $A_m(z)$ of the mth mode depends on the propagation coordinate due to the perturbation of the dielectric function introduced by the grating. Positive and negative values of β_m correspond to the modes propagating in the positive and negative directions of the z-axis, respectively. The field distributions $E_m(x, y)$ in waveguide modes are orthonormalized in accordance with the following condition:

$$\int E_k^*(x, y)E_m(x, y)\mathrm{d}x\mathrm{d}y = \delta_{km} \quad (3)$$

Periodic perturbation of the dielectric function gives rise to an additive to the polarization of the medium, $\Delta P = \Delta\varepsilon(x, y, z)E$, opening the channel of energy exchange between waveguide modes. In the case of a waveguide consisting of isotropic materials, modes with different polarizations remain uncoupled.[27] The photonic band gap in the dispersion relation and transmission spectrum of the considered waveguide structure arises owing to a strong coupling of forward and backward waveguide modes with propagation constants β_f and β_b, with the reciprocal lattice constant G involved in momentum conservation, leading to a Bragg resonance.

Owing to the z-periodicity of the perturbation of the dielectric function, the quantity $\Delta\varepsilon(x, y, z)$ can be represented as a Fourier series:

$$\Delta\varepsilon(x, y, z) = \sum_{m\neq 0} \varepsilon_m(x, y) \exp\left(-im\frac{2\pi}{\Lambda}z\right) \quad (4)$$

where m is the order of the Fourier component and Λ is the modulation period of the dielectric function (inset in Fig. 1).

Substituting Eqn (2) for the field into the wave equation, multiplying the resulting expression by $E_k^*(x, y)$ and performing integration in x and y with the use of Eqns (3) and (4) within the framework of the slowly varying amplitude approximation,[27] we arrive at

$$\frac{\mathrm{d}}{\mathrm{d}z}A_k = -i\sum_l\sum_m K_{kl}^{(m)}B_l\mathrm{e}^{i(\beta_k+\beta_l-m2\pi/\Lambda)z} - i\sum_l\sum_m K_{kl}^{(m)}A_l\mathrm{e}^{i(\beta_k-\beta_l-m2\pi/\Lambda)z} \quad (5)$$

$$\frac{\mathrm{d}}{\mathrm{d}z}B_k = i\sum_l\sum_m K_{kl}^{(-m)}A_l\mathrm{e}^{-i(\beta_k+\beta_l-m2\pi/\Lambda)z} + i\sum_l\sum_m K_{kl}^{(-m)}B_l\mathrm{e}^{-i(\beta_k-\beta_l-m2\pi/\Lambda)z} \quad (6)$$

where A_k and B_l are the amplitudes of counterpropagating modes and

$$K_{kl}^{(m)} = \frac{\omega^2}{2c^2\beta_k}\int E_k^*(x, y)\varepsilon_m(x, y)E_l(x, y)\mathrm{d}x\mathrm{d}y \quad (7)$$

is the coefficient of coupling of the kth and lth modes. This coefficient, as can be seen from Eqn (7), is determined by the spatial overlapping of the fields in waveguide modes and the area of perturbed dielectric function.

Waveguide modes with indices k and l are strongly coupled when the Bragg resonance condition is satisfied. This condition is written as $\beta_k + \beta_l = 2\pi m/\Lambda$ for counterpropagating modes and $\beta_k - \beta_l = 2\pi m/\Lambda$ for copropagating modes. Let us consider the case of strongly coupled counterpropagating waves assuming that Bragg resonance conditions are satisfied for a single value of m. Then, omitting all the terms on the right-hand sides of Eqns (5) and (6) with mode indices other than m and introducing notations $A_k' = A_k\mathrm{e}^{-i(\beta_k-m\pi/\Lambda)z}$ and $B_k' = B_k\mathrm{e}^{i(\beta_k-m\pi/\Lambda)z}$, we arrive at

$$\frac{\mathrm{d}}{\mathrm{d}z}A_k' + i(\beta_k - m\pi/\Lambda)A_k' + i\sum_l K_{kl}^{(m)}B_l' = 0 \quad (8)$$

$$\frac{\mathrm{d}}{\mathrm{d}z}B_k' - i(\beta_k - m\pi/\Lambda)B_k' - i\sum_l K_{kl}^{(-m)}A_l' = 0 \quad (9)$$

Equations (8) and (9) provide a background for our numerical analysis of the transmission and dispersion of a hollow planar PBG waveguide. The results of such an analysis are presented in the following section.

Numerical simulations: transmission spectra and the group velocity

The use of the approach described in the previous section allows us to find complex propagation constants for a hollow planar PBG waveguide by searching for the eigenvalues of $2M \times 2M$ square matrices of the relevant characteristic equations, where M is the number of modes of an unperturbed planar waveguide (a waveguide with no corrugation), determined from the cut-off condition.[28]

Figure 1 shows the imaginary part of the propagation constant as a function of the mode index at the wavelength $\lambda = 0.63$ μm for TE modes (transverse electric modes, with the electric field directed along the *y*-axis in the inset in Fig. 1) and bulk TM modes (transverse magnetic modes, with the electric field having non-zero projections on the *x*- and *z*-axes) of a hollow planar waveguide consisting of an aluminum mirror and an aluminum-coated 2400 grooves mm^{-1} grating located at a distance $2a = 44$ µm from the mirror. As can be seen from Fig. 1, the imaginary part of the propagation constant increases with increase in the mode index, leading to weakening of mode coupling around the photonic band gap. We shall therefore ignore the contribution of modes with indices above the cut-off to the transmission and dispersion of the waveguide.

The inset in Fig. 2 presents the spectral dependencies 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, owing 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. 2, agree qualitatively 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. 2). 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 constants, as can be seen from Eqn (7), depend on the Fourier amplitudes of the periodic profile of the grating, and also on

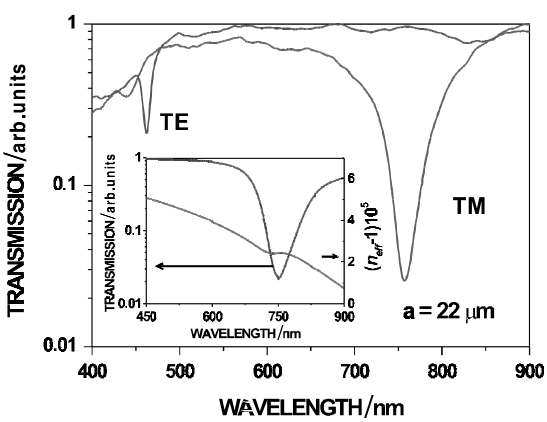

Figure 2. Transmission spectra measured for TM and TE modes of a planar corrugated hollow waveguide consisting of a 2400 grooves mm^{-1} aluminum-coated grating and an aluminum mirror with $2a = 44$ µm. The inset shows the results of calculations for the transmission spectrum and the spectral dependence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

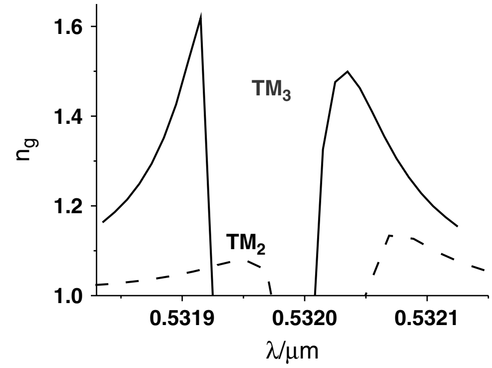

Figure 3. The spectral dependence of the group index n_g for the TM_2 (solid line) and TM_3 (dashed line) modes of a hollow planar corrugated waveguide with a 1200 grooves mm^{-1} diffraction grating and $2a = 22$ µm.

the spatial overlapping of light fields in waveguide modes and the area of perturbed dielectric function.

The main purpose of our experimental studies described in the following sections 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 non-linear medium in our experiments and the pressure of this gas never exceeded the atmospheric pressure, the coherence length for CARS-type FWM 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 owing 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. 3, which displays the wavelength dependence of the group index for TM_2 and TM_3 modes of a hollow planar PBG waveguide with a mirror–grating gap $2a = 22$ µm within the spectral range corresponding to the second harmonic of the Nd : YAG laser radiation. The solid and dashed lines in Fig. 3 represent the group indices for the TM_2 and TM_3 modes, respectively. The group velocity of light pulses, as can be seen from the results presented in Fig. 3, decreases considerably in this spectral range owing to the PBG effect. This increases the mean density flux of electromagnetic radiation in the waveguide, thus leading to the enhancement of non-linear optical processes.

EXPERIMENTAL

A diagram of the experimental set-up employed to investigate the CARS process in a gas medium filling a hollow planar PBG waveguide is shown in Fig. 4. We studied a two-color CARS process leading to the generation of a signal at the frequency $\omega_{CARS} = 2\omega_1 - \omega_2$ (Fig. 5), where ω_{CARS} is

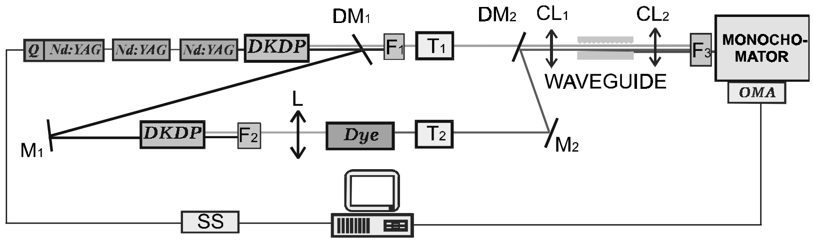

Figure 4. Diagram of the experimental set-up for studying coherent anti-Stokes Raman scattering in a hollow planar corrugated waveguide: M_1, M_2, rotating mirrors; DM_1, DM_2, dichroic mirrors; F_1–F_3, sets of optical filters; OMA, optical multichannel analyzer; SS, synchronization system; CL_1, CL_2, cylindrical lenses; L_1, spherical lens; T_1, T_2, telescopes.

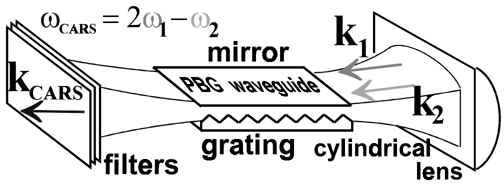

Figure 5. Diagram of coherent anti-Stokes Raman scattering in a planar corrugated photonic band-gap waveguide consisting of a mirror and a grating.

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 20 mJ in two Nd:YAG amplification stages. 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 a second DKDP crystal. This second-harmonic beam was then used to pump a sulforhodamine 101 dye laser. This dye laser radiation served as the second pump beam in the CARS process (the frequency ω_2).

The pump beams with 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 (Figs 4 and 5) 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^{-1} aluminum-coated diffraction gratings were used to create a hollow waveguide. The length of the waveguides used in our experiments was 5 cm. The distance between the waveguide walls was varied from 22 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 a wavelength of dye laser radiation of 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 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.

RESULTS AND DISCUSSION

To characterize the enhancement of the CARS process involving Raman-active transitions of molecular nitrogen in an 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 beams, but in the regime of cylindrical focusing of pump beams in the absence of a waveguide. Figure 6 presents the CARS enhancement ratio η_w/η_f measured as a function of the distance 2*a* between the

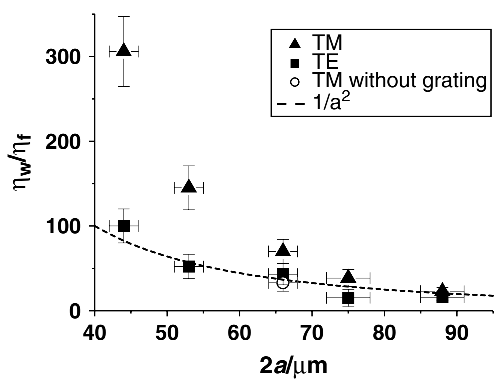

Figure 6. The ratio of the efficiency η_w of the CARS process in a hollow planar PBG waveguide to the efficiency η_f of the CARS process with the same energies of pump beams, but for cylindrically focused pump beams in the absence of a waveguide as a function of the distance 2*a* between the waveguide walls. The CARS signal is related to Raman-active transitions of molecular nitrogen in an atmospheric pressure air.

mirror and the 1200 grooves mm^{-1} grating, forming a planar PBG waveguide. Triangles show this ratio for TM waveguide modes and 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 geometric factors. In contrast to FWM processes in gas-filled hollow fibers, when this geometric enhancement ratio scales as $1/a^4$, where a is the fiber inner diameter,[25,26] 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. 6.

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. 6, with the enhancement attainable in the waveguide regime due to purely geometric factors. A much higher CARS enhancement ratio, as is seen from Fig. 6, 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 higher order TM modes. The electromagnetic energy density in the waveguide increases under these conditions, which leads to the enhancement of non-linear optical processes.

The results of CARS experiments correlate well with cross-correlation measurements intended to visualize group-delay effects in the PBG waveguide and performed with picosecond pulses produced by an Nd:YAG laser. A diagram of cross-correlation experiments is sketched in the inset to Fig. 7. Picosecond pulses produced by a passively mode-locked and actively Q-switched Nd:YAG laser system[29] were employed to generate the second harmonic in a KDP crystal. This second-harmonic radiation was then coupled into the PBG waveguide. Rotating the polarization of second-harmonic radiation, we were able to excite TE and TM mode families of the PBG waveguide. The cross-correlation function was measured by mixing second-harmonic radiation coming out of the waveguide with a time-delayed pulse of 1.06 μm radiation in another KDP crystal. The results of these cross-correlation measurements are presented in Fig. 7. The second-harmonic pulse transmitted through TM modes of the PBG waveguide is noticeably delayed in time with respect to the pulse transmitted through TE modes, indicating a considerable influence of group-delay effects for TM guided modes, which agrees well with our theoretical predictions. The time shift of the cross-correlation traces recorded for TE and TM modes ranged from 5 to 20 ps in our experiments, depending on the geometry of mode excitation.

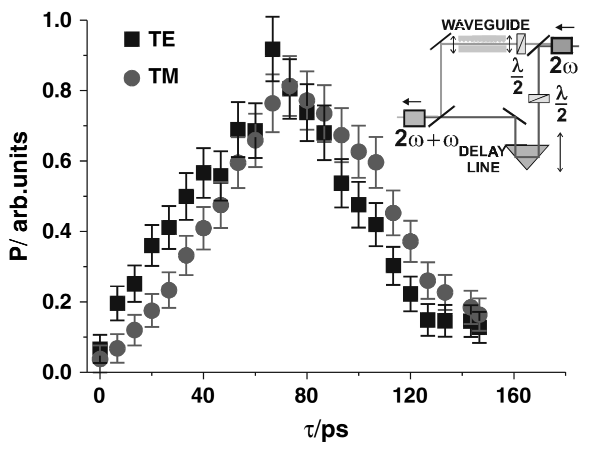

Figure 7. Cross-correlation traces for light pulses transmitted through TE (squares) and TM (circles) modes of the planar corrugated PBG waveguide. The inset shows the diagram of cross-correlation experiments.

The above-described cross-correlation measurements can provide only a qualitative demonstration of group-delay effects, since regimes with smaller numbers of guided modes excited in the PBG structure would be necessary to make quantitative judgments. However, the results of these experiments indicate that the improvement in CARS efficiency achieved in our PBG waveguide experiments in the case of TM modes may be attributed, at least partially, to the decrease in the group velocities of pump fields. There are, of course, several other physical factors leading to FWM enhancement under the conditions of our experiments. One of these factors may be related to local field enhancement in plasmon TM modes, which increases the efficiency of non-linear 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. As shown in earlier work,[30] ionization effects may considerably change phase-matching conditions in high-order non-linear optical processes. Investigation of these effects is in progress.

CONCLUSION

The results of the experimental and theoretical studies presented in this paper demonstrate the possibility of substantial enhancement of FWM processes in a gas medium placed 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 non-linear optical process. The idea of integrating advantages of a waveguide and a PBG structure in one optical component, on the other hand, can be extended to two dimensions by creating a waveguide consisting of two gratings whose grooves are perpendicular to each other. The method of enhancement of non-linear optical processes demonstrated in this paper opens up new possibilities for improving the sensitivity of non-linear 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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