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Kerr and four-wave mixing spectroscopy at the band edge of one-dimensional photonic crystals

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Kerr and four-wave mixing spectroscopy is shown to be a powerful technique to quantify the strong enhancement of the third-order optical nonlinear susceptibilities at the band edge of photonic crystals. Local field factors of about 5 are demonstrated for crossed Kerr effect and a narrow resonance peak observed for the conjugate reflectivity. Moreover, a reduction of the effective nonlinear susceptibility of the four-wave mixing process with increasing pump intensities is measured, which is due to different Kerr-induced blueshifts of the band edge for forward and backward pump beams and signal and conjugate beams. This observation definitely demonstrates the need for considering all the nonlinear processes for the optimization of nonlinear photonic crystals for a given application in optical signal processing. © 2005 American Institute of Physics.

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Due to their potential use in the development of new devices for optical signal processing, photonic crystals (PC) are presently widely studied both from the fundamental and applicative points of view.1,2 Besides passive circuits, PC microchips will need the realization of active functions such as optical switching. With respect to this need, third-order nonlinear optics will play a crucial role for the generation of optical switching. With respect to this need, third-order nonlinear susceptibilities of the PC near its band edge or at a defect in order to benefit from light localization. The sample used in this study consisted of two superposed Cd0.75Mn0.25Te–Cd0.40Mg0.60Te semiconductor quarter-wave stacks [see Fig. 1(a)]. The back stack (20 pairs deposited on a CdMnTe buffer first deposited on the CdZnTe substrate) was designed to reflect the incident light transmitted by the front stack (30 pairs) whose band edge was therefore exactly at the center wavelength of the back Bragg mirror. The refractive indices were \( n_{1} = 3.088 \) and \( n_{2} = 2.585 \) for Cd0.75Mn0.25Te and Cd0.40Mg0.60Te, respectively. The thicknesses of Cd0.75Mn0.25Te (\( e_{1} \) and \( e'_{1} \)) and Cd0.40Mg0.60Te (\( e_{2} \) and \( e'_{2} \)) layers were \( e_{1} = 54 \) nm and \( e_{2} = 64.6 \) nm for the front stack (the studied one-dimensional PC) and \( e'_{1} = 57.3 \) nm and \( e'_{2} = 68.5 \) nm for the back Bragg mirror. This design was chosen to automatically create the backward pump beam at the band edge of the PC for the four-wave mixing process. As a consequence of this design, for a lossless photonic crystal all the beams would be totally reflected for wavelengths inside the band gap of the PC or of the back Bragg mirror. As seen in Fig. 1(b), this is not the case at low photon wavelengths.
where band absorption occurs at the electronic band edge (EBE), of Cd$_{0.75}$Mn$_{0.25}$Te located at 620 nm and lowers the reflectivity. Strong losses occur also even below this EBE at an intensity of 0.50 GW/cm$^2$. In Figs. 2(a) and 2(b), the pump reflectivity (stars) and the signal reflectivity without and with the pump pulse present (closed and open triangles respectively) are plotted as a function of the laser-wavelength. The phase conjugate spectra appear in Figs. 2(c) and 2(d). As demonstrated in Figs. 2(a) and 2(b), the PC band edge represented by the hole in the reflectivity spectra is blueshifted for both signal and pump beams when the pump pulse is present on the sample. Moreover, this blueshift is almost proportional to the pump intensity ($\Delta \lambda_B = 0.3$ nm and 0.4 nm and $\Delta \lambda_B = 0.5$ nm and 0.8 nm with a precision of ±0.1 nm for $I_p = 0.22$ GW/cm$^2$ and $I_p = 0.50$ GW/cm$^2$, respectively). It must also be stressed that within the experimental errors the signal blueshift is twice that experienced by the pump beam due to the degeneracy of Kerr effect in the latter case. As these blueshifts, it is possible to evaluate the refractive index change by using the expression of the band edge wavelength $\lambda_B$ given in Ref. 16.

$$\lambda_0/\lambda_B = \arccos[-T_{12}\cos(\pi/N) + R_{12}],$$  \hspace{1cm} (2)

where $\lambda_0 = 4n_1\epsilon_1 = 4n_2\epsilon_2$ is the center wavelength of the band gap, and $T_{12} = 4n_1n_2/(n_1+n_2)^2$ and $R_{12} = (n_1-n_2)^2/(n_1+n_2)^2$ are the transmission and reflexion coefficients of the interfaces, respectively. Indeed, at high intensities, the refractive indices are modified ($n_i = n_i + \Delta n_i$ with $\Delta n_i$, the nonlinear refractive index change for $i=1, 2$). As $n_1 = n_2$, the change in the band edge wavelength is mainly related to the modification of $\lambda_0$ and is given, in first approximation, by

$$\Delta \lambda_B/\lambda_B = \Delta n_1/n_1 + \Delta n_2/n_2)/2.$$  \hspace{1cm} (3)

As the band edge wavelength of the PC is very near the electronic band gap of Cd$_{0.75}$Mn$_{0.25}$Te and far from that of Cd$_{0.40}$Mg$_{0.60}$Te, only the change of $n_1$ is of importance for determining the optical band edge blueshift of the PC. The nonlinear refractive index change for the crossed Kerr effect is then $\Delta n = -0.007$ and $\Delta n = -0.004$ for pump beam intensities of $I_p = 0.50$ GW/cm$^2$ and $I_p = 0.22$ GW/cm$^2$, respectively. The corresponding third-order susceptibility is then enormous ($\chi^{(3)}_{PC} = -1.2 \times 10^{-11}$ esu) which explains why these large changes in reflectivity occur with only small input intensities. Moreover using the value $\chi^{(3)} = -4 \times 10^{-11}$ esu calculated for the third order nonlinear susceptibility of bulk Cd$_{0.75}$Mn$_{0.25}$Te (Ref. 17) and assuming equal local field factors for all wavelengths, Eq. (1) gives the value $f = 4.2$ for the mean local field factor, in good agreement with that calculated using the transfer matrix method.

As expected from the high value of the measured local field factor, the four-wave mixing process was strongly enhanced at the band edge of the PC. This is illustrated in Figs.
as a function of the pump intensity. The value is high and the peak effective third-order nonlinear susceptibility plotted intensity. This last behavior is illustrated in Fig. 3 showing the effective nonlinear susceptibility strongly depends on the pump intensities and that the maximum value of the measured effective nonlinear susceptibilities. This observation demonstrates that all the possible nonlinear processes must be taken into account for an optimized design of nonlinear photonic crystals for optical signal processing.

In conclusion, Kerr and four-wave mixing spectroscopy have been used in order to experimentally quantify the strong enhancement of the corresponding third order nonlinear susceptibilities. Our results demonstrate local field factors larger than four for a low contrast one-dimensional PC and a narrow peak for the phase conjugate reflectivity due to light localization. Moreover, a reduction of the enhancement of the phase conjugate reflectivity was demonstrated at high laser intensities due to different Kerr-induced blueshifts experienced by the forward and backward pump beams on one hand and signal and conjugate beams on the other hand. This result definitely demonstrates that all the possible nonlinear processes must be taken into account for an optimized design of nonlinear photonic crystals for optical signal processing.

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