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Resonant second-harmonic generation in a GaAs photonic crystal waveguide

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Second-harmonic generation (SHG) measurements in reflection on a GaAs/$Al_{0.25}Ga_{0.75}As$ photonic crystal waveguide show a resonant enhancement when the pump beam is frequency and momentum matched with the photonic modes in the slab. The enhanced SH signal is observed in the form of resonant peaks, unlike in linear reflectance spectra. The observations are in very good agreement with a full 3D calculation of the anisotropic mode dispersion in the photonic crystal slab. The present results open the way towards realizing the extraordinary enhancement of SHG which was recently predicted [A. R. Cowan and J. F. Young, Phys. Rev. B **65**, 085106 (2002)], and also demonstrate the potential of SHG as a nonlinear spectroscopic tool for optical studies of photonic crystals.

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The nonlinear properties of photonic crystals (PC's) are becoming a new and exciting area of research in nonlinear optics.[1] The tailoring of photonic states and of their dispersion relations due to the spatial variation of the dielectric constant offers extraordinary possibilities for the realization of large nonlinearities, phase-matching conditions, diffraction processes and anisotropy, resonant effects, etc. Focusing on quadratic nonlinearities, second-harmonic generation (SHG) with phase matching due to form birefringence was first demonstrated in GaAs/Alox superlattices[2] and more recently in nanostructured silicon.[3] Nonlinear diffraction in two-dimensional (2D) PC's with spatially varying $\chi^{(2)}$ was predicted[4] and observed in periodically poled lithium niobate.[5] Various schemes for achieving enhanced SHG due to phase matching *and* an increased density of photonic states have been proposed in 1D microcavities[6] and periodic systems.[7] Phase matching was also demonstrated in colloidal 3D PC's.[8]

Among the great variety of possible structures, photonic crystals embedded in planar waveguides (also known as PC slabs)[9] are intensively investigated in order to achieve a full control of light propagation in 3D. The lithographic definition of the photonic pattern results in a precise control of the 2D lattice, which may also contain linear and point defects acting as channel waveguides and microcavities, respectively. A photonic crystal slab may support a small number of truly guided modes, which lie below the light line of the cladding material and are ideally lossless;[10] however the majority (sometimes all) of the photonic modes lie above the light line and are quasiguided, since they are subject to radiation losses due to coupling to leaky waveguide modes. Quasiguided modes can be probed by an external beam incident on the surface[11] in a reflectance measurement. The latter technique has proved to be a very useful and flexible method to measure the dispersion of the photonic bands.[11–15] The coupling strength of a quasiguided mode to the external beam is small: indeed, the signature of photonic modes in linear reflectance spectra consists in weak spectral features which may have widely differing line shapes (peaks, dips, or dispersivelike). It was recently predicted that SHG in reflection from the surface of a PC waveguide should be resonantly enhanced when the frequency of the pump and/or the second-harmonic beam coincides with that of a photonic mode of the slab.[16,17] This is due to a strong increase of the electromagnetic field energy in the core of the waveguide. The enhancement of SHG can reach up to six orders of magnitude when resonance at the pump and at the SH frequency is simultaneously achieved, thereby realizing an effective phase-matching condition for SHG.

In this paper we report on SHG measurements on a GaAs PC slab, which demonstrate the enhancement of the SH signal when the pump frequency is resonant with a photonic mode. The appearance of a quasiguided mode in the nonlinear process is strongly enhanced as compared to the linear reflectance spectrum and it allows us to perform nonlinear spectroscopy of photonic states with greatly increased sensitivity. The observations are interpreted according to a full 3D calculation of the photonic mode energies and dispersion in the PC slab. The present results show the potential of nanoscale structures such as PC slabs in producing large enhancement of SHG, in line with the expectations of Ref. 16. They also demonstrate the interest of SHG as a spectroscopic tool for optical studies of photonic crystals, in analogy to other nonlinear techniques (e.g., resonant two-photon absorption) for the optical spectroscopy of crystalline solids.[18]

The sample employed in the present study is an air/GaAs/AlGaAs waveguide (nominally 500-nm GaAs, 1.5-μm $Al_{0.25}Ga_{0.75}As$) grown on [001]-oriented GaAs substrate and patterned with a square lattice of 500-nm period; the unit cells contain air rings with 12% air fraction. The fabrication procedure consists of epitaxial growth, electron-beam and x-ray lithography, and reactive-ion etching.[19] A scanning-electron micrograph of the sample is shown in the inset to Fig. 1; the etch depth is about 1 μm. Variable-angle linear reflectance on this sample yielded the dispersion of photonic bands in the 0.5–1.1 eV energy range.[15] SHG measurements

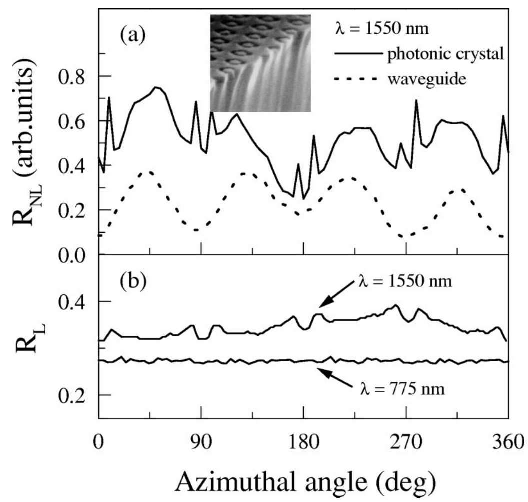

FIG. 1. (a) Nonlinear second-harmonic reflectance R_{NL} vs azimuthal angle ϕ ($\theta = 45°$, p polarization of the pump) of the photonic crystal (solid line) and of the bare waveguide (dotted line). Inset: scanning-electron micrograph of the sample. (b) Linear reflectance R_{L} of the photonic crystal ($\theta = 45°$, p polarization) at the pump and harmonic frequencies.

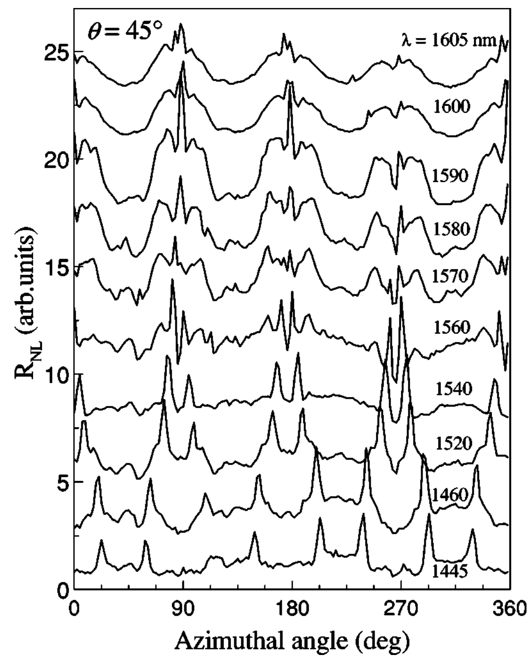

FIG. 2. Nonlinear second-harmonic reflectance vs azimuthal angle ϕ (at $\theta = 45°$, p polarization of the pump) of the photonic crystal for the indicated pump wavelengths. The different curves are offset for clarity.

in reflection and diffraction geometry were previously performed at the wavelengths of a Ti:sapphire laser.[20] However, the pump frequency was too high to induce resonance effects on the photonic modes.

In the present experiment, instead, a femtosecond optical parametric oscillator pumped by a Ti:sapphire laser has been employed. The source is continuously tunable between 1450 and 1610 nm and delivers pulses of up to 2.5 nJ at a repetition rate of 80 MHz. The laser beam is intensity and polarization controlled and the light spot on the sample is of ~ 40 μm diameter. The pump intensity I_{ω} is monitored by an InGaAs photodiode. The setup allows us to vary the angle of incidence θ between $\sim 30°$ and $60°$, and to change the orientation angle ϕ of the plane of incidence with respect to the [100] direction of the GaAs substrate. The reflected radiation is collected by a 1-mm core optical fiber whose end is positioned in front of a cooled photomultiplier (PMT) with maximum sensitivity between 700 and 800 nm. A combination of color and wide-band interference filters prevents undesired radiation wavelengths to reach the detector. The amplitudes of the PMT $I_{2\omega}$ and photodiode I_{ω} signals, averaged over ~ 50 oscilloscope traces, are taken for each angle ϕ by modulating the pump beam with a mechanical chopper. From these data the nonlinear reflectance $R_{\mathrm{NL}} = I_{2\omega}/I_{\omega}^2$ is obtained. No absolute calibration is performed, however the radiant sensitivity of our apparatus is almost constant in the spectral range investigated.

Measurements were performed at fixed angles of incidence θ by varying the azimuthal angle ϕ by one full turn for the two input polarizations. Similar measurements were also performed on the unpatterned waveguide. Figure 1(a) illustrates a typical comparison (at $\theta = 45°$, p polarization of the pump, and $\lambda = 1550$ nm) between R_{NL} vs ϕ on the PC (solid line) and on the bare waveguide (dotted line). The nonlinear response of the PC results from the superposition of two effects, namely, a bulk SH modulation given by the GaAs core layer (similar to that of the unpatterned waveguide) and a resonant pattern associated with the photonic lattice. The former reflects the fourfold symmetry axis of (001)-oriented GaAs surface, and is a well-known result of SH generation with input p polarization.[21] The SH peaks arise from a resonance of the pump beam with quasiguided modes in the PC slab and have the same fourfold symmetry of the square photonic lattice. In general, a full rotation around the azimuth draws a circular path in the Brillouin zone around the Γ point. When the pump radiation is resonant with a photonic mode at the same in-plane wave vector, spectral structures are expected.[16] Due to the symmetry of the square Bravais lattice of the PC, each pair of peaks occurs four times, therefore the photonic resonances occur a total of eight times *per* revolution. This effect is highly amplified in the nonlinear response as compared to the linear one, as is evident by the sharp and well resolved peaks of Fig. 1(a) compared with the weak structures in the linear spectrum of Fig. 1(b) for $\lambda = 1550$ nm. No structures are observed in the linear spectrum at the harmonic frequency. Measurements with input s polarization (not shown) differ from those of Fig. 1(a) only by the waveguide which is displaced by $45°$.[21]

The nonlinear response of the system exhibits a rich behavior as a function of the incident photon energy. Figure 2 shows several scans on the PC vs ϕ taken for $\theta = 45°$ and p polarization with pump wavelengths ranging from 1445 to 1605 nm. On increasing the pump wavelength, the two peaks in each pair approach each other and merge around $\lambda = 1580$ nm, marking the edge of the photonic band, and split again at longer wavelengths. All SH resonances are seen to

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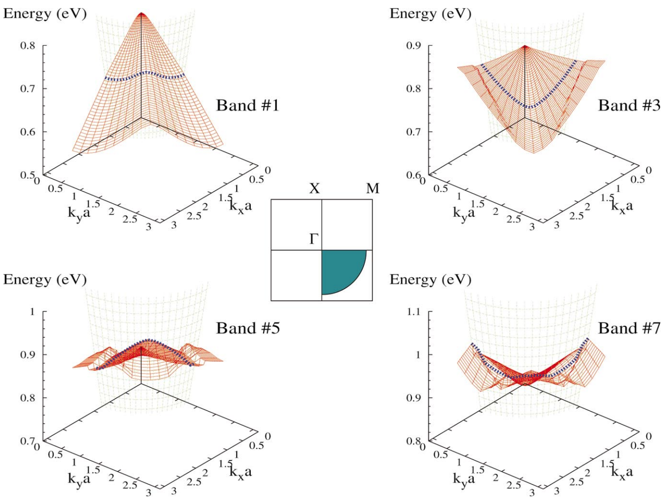

FIG. 3. (Color) Dispersion of the first, third, fifth, and seventh photonic bands (in red) in the region of the Brillouin zone shown as a blue area. The plots show also the light cone at an incidence angle $\theta = 45°$ (in green) and the intersection between the band dispersion and the light cone (blue line).

have the form of pronounced peaks, unlike in linear reflectance spectra[11–15] [see also Fig. 1(b)]. This important feature of the nonlinear process is explained as follows. The field energy in the waveguide core is strongly increased when the external field has the same frequency and the same in-plane wave vector of a quasiguided mode, giving rise to an enhancement of the nonlinear polarization and hence of the SH signal. As a consequence, the resonant energy of a quasiguided mode should be taken at the peak maximum. This is a remarkable difference of SHG over linear reflectance spectra, which exhibit different line shapes depending on the underlying interference pattern.[11–15]

The photonic band dispersion has been calculated by a 3D approach[22] which consists of an expansion on the basis of guided modes of an effective wave guide, where each layer has a spatially averaged dielectric constant. The method yields all photonic modes, including the quasiguided modes above the cladding light line. The dispersion of the photonic bands is strongly anisotropic. As an example, in Fig. 3 we show the dispersion of the first, third, fifth, and seventh photonic bands for the investigated sample, in the region of the Brillouin zone indicated by the blue area. The bands in Fig. 3 are of course repeated with 90° periodicity. The four panels also show the dispersion of light in air at an incidence angle $\theta = 45°$, corresponding to the experimental results in Figs. 1 and 2: the photonic resonances in SHG as a function of azimuthal angle ϕ trace the intersection (blue lines) between the band dispersion and the light cone at the specified angle of incidence θ. Notice that a photonic band cannot be characterized by a polarization label (except along the high-symmetry directions Γ-X and Γ-M of the square lattice). This fact implies that any band has the proper polarization component to be coupled with a p polarized pump. Due to the low air fraction of the lattice, it turns out that all bands occur in pairs with nearly degenerate energy values. In the following we concentrate on bands 3 and 5, which fall in the investigated energy range.

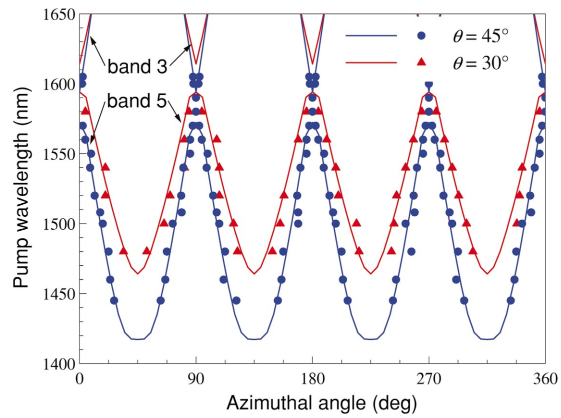

FIG. 4. (Color) Photonic band diagram vs azimuthal angle ϕ for different angles of incidence $\theta = 30°$ and 45° as calculated (solid lines) and measured (symbols).

Figure 4 illustrates the comparison between calculated and experimental photonic band dispersion for two different angles of incidence: the calculated values correspond to the blue lines in Fig. 3. The agreement is very satisfactory and demonstrates the reliability of the nonlinear optical technique as well as of the theoretical method. Most of the experimental points correspond to band 5, while only a few of them (for $\lambda = 1600$ and 1605 nm) are associated with band 3. Notice that the calculated band diagrams display a narrow photonic gap whose central energy and width have a slight variation with angle of incidence θ. On the contrary, by following two adjacent resonance peaks in Fig. 2 with increasing wavelengths, one observes that they merge at approximately the band-gap energy and then split again without disappearing. The absence of an experimental photonic gap follows from the linewidth of the resonances.

In general, the radiative linewidth of the photonic states is a crucial parameter for the coupling of the external radiation field with quasiguided modes. The *intrinsic* radiative linewidth of the quasiguided modes, which would persist in an ideal structure with infinitely deep holes and no disorder, can be calculated within the finite-basis expansion method by taking into account coupling to leaky waveguide modes by Fermi's golden rule. The resulting values for the intrinsic linewidth are lower than 0.2 nm for the present sample. This follows from both the weak refractive index contrast in the planar GaAs/AlGaAs waveguide, and the very low air fraction of the 2D lattice.[23] These values are much lower than the spectral linewidths of the SH peaks in Figs. 1 and 2, which are of the order of 30 nm. The large observed linewidths follow from three experimental factors: (a) the short laser pulse duration which produces a finite bandwidth of the pump beam (~25 nm); (b) inhomogeneous broadening and disorder in the sample; (c) the effect of sampling a small area and hence a finite number of unit cells of the PC, thereby giving a finite dispersive power to the photonic lattice. All these factors are likely to play a role in the present experiment, however effect (a) is probably dominant. The large experimental broadening is the main reason why the enhancement of SH resonances (typically three to four times with respect to the background) is much smaller than predicted in theoretical work.[16]

In summary, we have demonstrated experimentally the novel possibilities offered by nonlinear interaction of light with photonic structures in terms of exploiting resonance effects in reflection; similar effects are also expected in a diffraction geometry.[20] The spectral resonance of incoming light with photonic bands provides a strong increase of the SH generated signals. This phenomenon is analogous to ''giant two-photon absorption'' by excitations in solids, where a resonance with an intermediate state leads to an enhancement of the transition probability.[18] Resonant SHG allows us to map the anisotropic band dispersion with high sensitivity; the measured photonic dispersion is in excellent agreement with the results of a full 3D calculation of the quasiguided mode dispersion. The line shape of a single SHG resonance is observed in the form of a peak. This may allow us in principle to probe the radiative line width of the quasiguided modes. In the present experiment we have demonstrated the enhancement due to resonance with the pump beam only. This preludes to the phenomenon predicted by Cowan and Young,[16] i.e., the occurrence of a double resonance with both pump and second harmonic, or in general with any parametrically generated radiation, which should provide much greater enhancement factors.

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[1] See papers in *Nonlinear Optics of Photgonic Crystals*, edited by C.M. Bowden and A.M. Zheltikov, Feature issue J. Opt. Soc. Am. B **19**, 1961 (2002).

[2] A. Fiore, V. Berger, E. Rosencher, P. Brevetti, and J. Nagle, Nature (London) **391**, 463 (1998).

[3] P.K. Kashkarov, L.A. Golovan, A.B. Fedotov, A.I. Efimova, L.P. Kuznetsova, V.Y. Timoshenko, D.A. Sidorov-Biryukov, A.M. Zheltikov, and J.W. Haus, J. Opt. Soc. Am. B **19**, 2273 (2002).

[4] V. Berger, Phys. Rev. Lett. **81**, 4136 (1998).

[5] N.G.R. Broderick, G.W. Ross, H.L. Offerhaus, D.J. Richardson, and D.C. Hanna, Phys. Rev. Lett. **84**, 4345 (2000).

[6] V. Berger, J. Opt. Soc. Am. B **14**, 1351 (1997); V. Pellegrini, R. Colombelli, I. Carusotto, F. Beltram, S. Rubini, R. Lantier, A. Franciosi, C. Vinegoni, and L. Pavesi, Appl. Phys. Lett. **74**, 1945 (1999).

[7] M. Centini, C. Sibilia, M. Scalora, G. D'Aguanno, M. Bertolotti, M. J. Bloemer, C.M. Bowden, and I. Nefedov, Phys. Rev. E **60**, 4891 (1999); G. D'Aguanno, M. Centini, M. Scalora, C. Sibilia, Y. Dumeige, P. Vidakovic, J.A. Levenson, M.J. Bloemer, C.M. Bowden, J.W. Haus, and M. Bertolotti, *ibid.* **64**, 016609 (2001); Y. Dumeige, I. Sagnes, P. Monnier, P. Vidakovic, I. Abram, C. Mériadec, and A. Levenson, Phys. Rev. Lett. **89**, 043901 (2002).

[8] J. Martorell, R. Vilaseca, and R. Corbalan, Appl. Phys. Lett. **70**, 702 (1997).

[9] See papers in *Photonic CrystalStructures and Applications*, edited by T.F. Krauss and T. Baba, Feature section, IEEE J. Quantum Electron. **38**, 724 (2002).

[10] S.G. Johnson, S. Fan, P.R. Villeneuve, J.D. Joannopoulos, and L.A. Kolodziejski, Phys. Rev. B **60**, 5751 (1999).

[11] V.N. Astratov, D.M. Whittaker, I.S. Culshaw, R.M. Stevenson, M.S. Skolnick, T.F. Krauss, and R.M. De La Rue, Phys. Rev. B **60**, R16 255 (1999).

[12] D. Coquillat, A. Ribayrol, R.M. De La Rue, M. Le Vassor d'Yerville, D. Cassagne, and C. Jouanin, Phys. Status Solidi A **183**, 135 (2000).

[13] V. Pacradouni, W.J. Mandeville, A.R. Cowan, P. Paddon, J.F. Young, and S.R. Johnson, Phys. Rev. B **62**, 4204 (2000); A.R. Cowan, P. Paddon, V. Pacradouni, and J.F. Young, J. Opt. Soc. Am. A **18**, 1160 (2001).

[14] M.C. Netti, A. Harris, J.J. Baumberg, D.M. Whittaker, M.B.D. Charlton, M.E. Zoorob, and G.J. Parker, Phys. Rev. Lett. **86**, 1526 (2001).

[15] M. Galli, M. Agio, L.C. Andreani, L. Atzeni, D. Bajoni, G. Guizzetti, L. Businaro, E. Di Fabrizio, F. Romanato, and A. Passaseo, Eur. Phys. J. B **27**, 79 (2002).

[16] A.R. Cowan and J.F. Young, Phys. Rev. B **65**, 085106 (2002).

[17] In the context of 1D gratings, a similar phenomenon was considered by R. Reinisch and M. Nevière, Phys. Rev. B **26**, 5987 (1982).

[18] E. Hanamura, Solid State Commun. **12**, 951 (1971); G.M. Gale and A. Mysyrowicz, Phys. Rev. Lett. **32**, 727 (1974); J.K. Pribram, G.L. Koos, F. Bassani, and J.P. Wolfe, Phys. Rev. B **28**, 1048 (1982).

[19] F. Romanato, L. Businaro, E. Di Fabrizio, A. Passaseo, M. De Vittorio, R. Cingolani, M. Patrini, M. Galli, D. Bajoni, L.C. Andreani, F. Giacometti, M. Gentili, D. Peyrade, and Y. Chen, Nanotechnology **13**, 644 (2002).

[20] A.M. Malvezzi, F. Cattaneo, G. Vecchi, M. Falasconi, G. Guizzetti, L.C. Andreani, F. Romanato, L. Businaro, E. Di Fabrizio, A. Passaseo, and M. De Vittorio, J. Opt. Soc. Am. B **19**, 2122 (2002).

[21] N. Bloembergen and P.S. Pershan, Phys. Rev. **128**, 606 (1962); J. Ducuing and N. Bloembergen, Phys. Rev. Lett. **10**, 474 (1963).

[22] L.C. Andreani and M. Agio, IEEE J. Quantum Electron. **38**, 891 (2002).

[23] H. Benisty, H. Benisty, D. Labilloy, C. Weisbuch, C.J.M. Smith, T.F. Krauss, D. Cassagne, A. Beraud, and C. Jouanin, Appl. Phys. Lett. **76**, 532 (2000).