Design and Fabrication of Two-Dimensional Photonic Crystals with Predetermined Nonlinear Optical Properties

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By probing the resonances between a photonic band and an external laser field and their nonlinear changes in angle-resolved reflectivity, we show experimental evidence that the nonlinear optical changes in a two-dimensional photonic crystal waveguide with a Kerr nonlinearity are critically dependent on the dispersion nature and the group velocity of the photonic bands. The results agree well with the behavior predicted from band structures, indicating that the design of nonlinear optical properties of material systems is realistically possible by band dispersion and group velocity engineering.

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Photonic crystals (PhCs) are materials composed of dielectric structures with periodicity on the optical wavelength scale [1–3]. The ability to control the light dispersion relation (photonic band structure) with a great degree of freedom is an issue of fundamental scientific and practical importance and has opened up unprecedented and exciting possibilities in nonlinear optics [4,5]. In particular, interest has grown rapidly in PhCs exhibiting nonlinear optical switching effects as such materials are strong candidates for practical all-optical signal processing devices [6–8]. The optical nonlinearities of conventional nonlinear materials are usually very weak. However, tailoring of the band dispersions in PhC systems can produce larger nonlinearities and minimize the power requirements for switching processes. This arises from the anomalous dispersion characteristics of PhCs, which enhance the electromagnetic field due to their extremely slow group velocities as compared with conventional uniform materials. In this regard, a two-dimensional (2D) PhC waveguide represents one of the most promising PhC structures for efficient nonlinear applications. Unique band dispersions can be designed through control of the 2D PhC configuration and a high optical intensity can be maintained over a long interaction distance.

A variety of optical switching processes based on 2D PhC waveguides have been proposed [7,8]. Although many of these ideas are promising, experimentally determining the performance characteristics of these processes is a great research challenge, especially for highly nonlinear host materials, because of the difficulty in obtaining high-aspect, high-quality 2D PhC profiles. Several experimental studies have been undertaken, mainly concentrating on liquid crystals infused into the air pores of silicon or silica PhCs, which can be tuned by applying an electric field or adjusting the temperature [9,10], and more recently, on semiconductor PhCs tunable by free-carrier injection [11]. These studies employ dynamic tuning of the optical properties of PhCs by modifying their refractive index, but the important issues remain to be solved. In 2D PhC systems combined with highly nonlinear materials, the relationship between the nonlinear optical changes based on third-order nonlinear responses and the band dispersion characteristics has yet to be clarified for the case of an external laser field resonantly coupled to the photonic band. Clarifying the relation between the nonlinear optical response and the nature of the band dispersion at the resonance state and engineering the matter-radiation nonlinear interaction by controlling the band dispersion are essential for the realization of desirable nonlinear applications, such as practical all-optical switching and processing devices with very low operational power and ultrasmall dimensions, which are greatly demanded by photonic information technology. In this study, we experimentally identify nonlinear optical changes arising from modification of the photonic band structure in a 2D PhC waveguide with a Kerr nonlinearity driven by an external laser field resonantly coupled to the photonic band. We reveal a direct relationship between the observed nonlinear responses and the corresponding photonic band structure by angle-resolved reflectivity measurements with reference to experimentally and theoretically determined photonic band structures. Our results verify that the nonlinear dynamics and properties of the waveguide can be enhanced by manipulating the photonic band dispersion and point the way towards nonlinear dynamics engineering in 2D PhC systems.

The 2D PhC waveguide structure with a Kerr nonlinearity is schematically illustrated in Fig. 1(a). The techniques for producing the PhC have been described elsewhere [12–14]. We have identified the nonlinear optical polymer DR1/PMMA, composed of poly(methacrylate) (PMMA) doped with dispersed red one (DR1) as one of the best candidates for nonlinear optical materials applied to 2D PhCs. DR1/PMMA exhibits large nonlinear optical effects and an ultrahigh switching speed due to a large delocalized π electron system [15], and has good

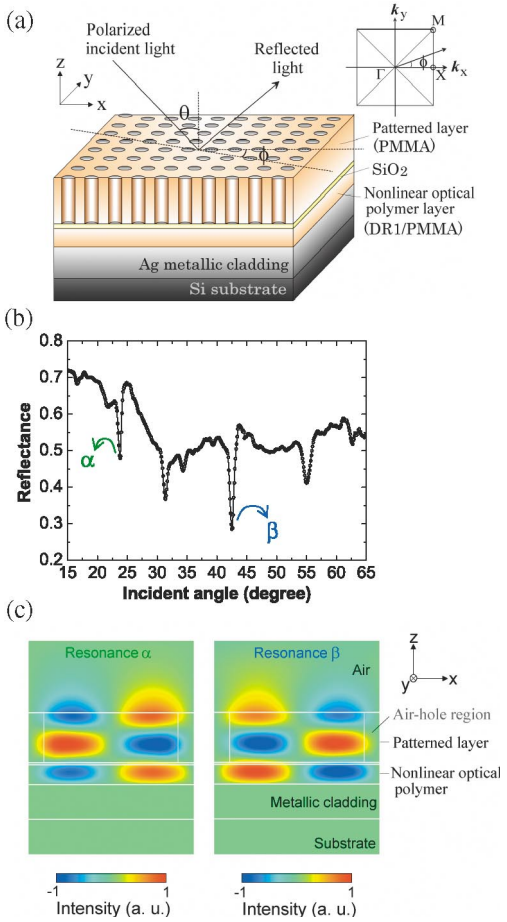

FIG. 1 (color). (a) A schematic showing the experimental geometry of our 2D PhC waveguide. The crystal structure of the patterned layer has a square lattice of circular air holes of diameter 150 nm with a lattice constant of 800 nm. The incident light is transverse electric (TE) polarized. The inset shows the corresponding 2D first Brillouin zone and the high-symmetry lattice points. (b) Measured angular dependence of the linear reflectivity of the 2D PhC waveguide at a fixed wavelength of 800 nm along the Γ-X direction. (c) Electric field distributions in the x-z plane through the center of an air hole of the waveguide at the resonance wavelength (800 nm), and wave vectors k of the resonances α (left) and β (right) calculated by the 3D finite-difference time-domain method. The maximum of the electric field is normalized to unity. Perspective outlines of the structure in the cross section are superimposed on the diagram (white lines).

processability. It is essentially transparent (absorption coefficient, $\alpha < 1\ \mathrm{cm}^{-1}$) at the operation wavelength (800 nm) of our nonlinear experiments [16]. To prevent damage to the molecules in the nanofabrication processes, we have separated the 2D PhC slab from the nonlinear optical polymer layer. The structure, from top to bottom, consists of a patterned PMMA layer, a SiO_2 thin etch stop layer, a layer of nonlinear optical polymer, and a Ag metallic cladding layer on top of a Si substrate. The thicknesses of these layers are 700, 20, 280, and 500 nm, respectively. The 2D PhC slab was patterned with a square lattice of circular air holes using electron beam lithography and inductivity coupled plasma dry etching. The damage to the nonlinear optical materials caused by these patterning processes was minimized by the vertical structural separation of the patterning layer and the nonlinear optical active layer. The metallic cladding enables the modes to exist over a relatively long lifetime in the waveguide core compared with low refractive-index dielectric cladding, due to their stronger confinement, in spite of absorption in the metallic region [14].

To examine the nonlinear optical changes arising from modifications of the photonic bands driven by an external laser field resonantly coupled to the photonic bands, we performed linear and nonlinear angle-resolved reflectivity measurements. These measurements provide a sharp distinction between the different photonic bands by separating the linear components in the angle (or wave vector, k) domain and also convey information on the nonlinear optical responses by monitoring the distinct dynamics of each photonic band in the 2D PhC system. Furthermore, because this technique can separate photonic bands of the same energy, we can investigate the nonlinear optical changes of the different photonic bands without having to consider the energy dispersions of the linear and nonlinear susceptibilities.

In the linear and nonlinear specular angle-resolved reflectivity measurements, the light source used was a mode-locked, cavity-dumped Ti:sapphire laser operating at a wavelength of 800 nm, with a repetition rate of 1 kHz and a pulse duration of 2 ps. The laser beam was divided into two by a beam splitter. One beam from the beam splitter was focused onto the sample ($200 \times 200\ \mu\mathrm{m}^2$) with an achromatic long-focal-length lens ($f = 250$ mm). The beam reflected from the surface of the sample was detected by a Si photodiode and a boxcar integrator after spatial filtering. The other beam from the beam splitter was detected by another Si photodiode and boxcar integrator, which served as a reference to compensate for intensity fluctuations of the laser. Measurements were performed for varying angles of incidence. For simplicity, the in-plane propagation lattice direction was set along the $\Gamma - X(\phi = 0^\circ)$ direction. The inset of Fig. 1(a) shows the 2D Brillouin zone and the symmetry points for a square lattice. All experiments (linear and nonlinear) were performed with TE polarization of the incident light.

Figure 1(b) shows the linear angle-dependent reflectivity of the 2D PhC waveguide at a wavelength of 800 nm, measured with TE polarization along the $\Gamma - X$ lattice direction. A sufficiently low pulse energy was used in this measurement. Several sharp dips (depressions) are distinctly observed in the reflectivity. These dip structures originate from resonant phenomena associated with surface coupling to in-plane photonic bands at resonance energies and in-plane wave vectors [14]. The observed resonance angles have associated in-plane wave vectors, $k = (2\pi/\lambda)\sin\theta$, where λ is the wavelength of the incident light and $\theta = 0^\circ$ is normal incidence. The resonance dip at each resonance angle in the reflectivity is thus directly

connected to the in-plane photonic band mode. Figure 1(c) shows the y components of the electric field of the band modes in the x-z plane at the resonance angles indexed α and β in Fig. 1(b), calculated by the 3D finite-difference time-domain (FDTD) method [14]. This figure shows that the modes in the 2D patterned and the nonlinear active layers are well coupled between the layers and are strongly confined in the waveguide core by the metallic cladding.

Figure 2 shows the angle-dependent reflectivities for different incident peak powers at a wavelength of 800 nm in two different angle ranges in the vicinity of the resonance angles α and β. Clearly, the resonance dip positions in the reflectivities are shifted with increasing incident peak power for both resonance dips. The angular shifts between the incident laser peak powers of 0.05 GW/cm^2 and 1.6 GW/cm^2 at resonances α and β are $+0.06°$ and $-0.30°$, respectively, having different signs. These shifts are accompanied by large nonlinear changes in the reflectivities at the resonance angles of $\Delta R/R = 7.3\%$ and 27.3%, respectively, for the peak power of 0.05 GW/cm^2. These novel and unusual nonlinear phenomena, exhibiting differences in sign and quantity of the nonlinear angular shifts at the resonances, indicate that the nonlinear changes are strongly influenced by the photonic band dispersion properties.

To find the underlying physical mechanism of these results, we examined the photonic band structure of the sample by angle-dependent optical reflectivity measurements in the range 550–950 nm using a collimated tungsten-halogen white light source and a liquid-nitrogen-cooled charge coupled device detector [14]. Figure 3(a) shows the experimental photonic band structure along the Γ-X line compared with the theoretical band structure calculated by the 3D-FDTD method for a model structure identical to the experimental sample structure. The features of the observed band dispersion are in reasonably good agreement with the theoretical curves.

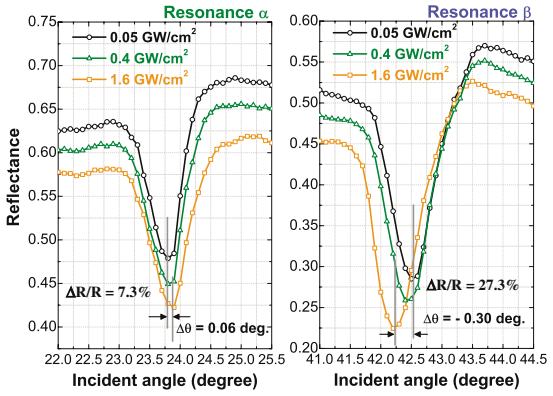

FIG. 2 (color). Angle-dependent reflectivities of the 2D PhC waveguide for three different incident peak powers at two different angle ranges in the vicinity of the resonances α (left) and β (right), measured at a fixed wavelength of 800 nm along the $\Gamma - X$ direction. For clarity, the reflectivities are shifted by -0.03 along the vertical axis with respect to each other. Here, the vertically unshifted reflectivity used as a base is for a peak power of 0.05 GW/cm^2 (black circles).

The experimental and theoretical band structures provide a very clear picture of the relation between the observed nonlinear responses and the photonic band dispersion. In Fig. 3(a), the horizontal black solid line shows the photon energy corresponding to the energy of the incident laser used in the nonlinear experiments. The positions of the in-plane wave vectors k of the incident light at the angles of resonances α and β at this photon energy are indexed as points I and II, respectively. The band dispersion branches shown by the green and blue solid curves correspond to the resonance band branches that contribute to the nonlinear reflectivity changes in the vicinity of α and β shown in Fig. 2. The group velocities v_g of these corresponding bands, estimated by the slope of the dispersion curve ($v_g = \partial\omega/\partial\boldsymbol{k}$), are $v_g = 0.46c$ and $v_g = -0.19c$ at points I and II, respectively, where ω is the angular frequency and c is the light velocity in a vacuum.

We now directly compare the observed nonlinear optical responses with the photonic band structure features in the vicinity of the resonances. The k shift values obtained from the observed resonance angle shifts in Fig. 2 are $\Delta ka/2\pi = +9.5 \times 10^{-4}$ for resonance α and $\Delta ka/2\pi = -3.9 \times 10^{-3}$ for resonance β, where a is the lattice con-

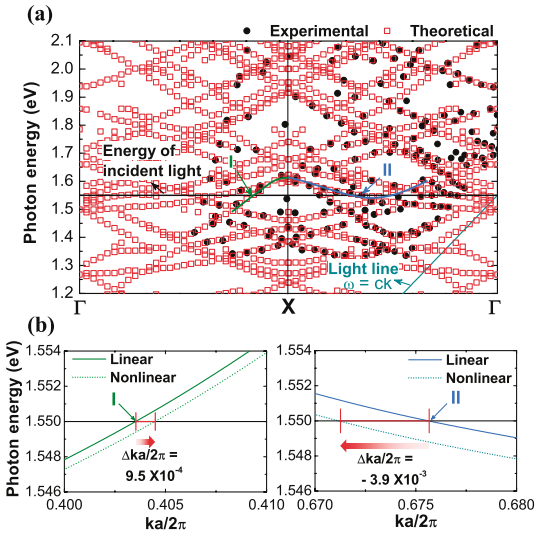

FIG. 3 (color). (a) Experimental photonic band structure of the 2D PhC waveguide along the $\Gamma - X$ line obtained by polarized angular-dependent reflectivity measurements (closed black circles), and theoretical band structure calculated by the 3D finite-difference time-domain method (open red squares). No empirical adjustments of any kind were made in the calculations. Points I and II are the positions of the wave vectors k and photon energies at the resonances α and β. The photonic bands contributing to the nonlinear reflectivity changes in the vicinity of α and β are indicated by the green and blue solid lines, respectively. (b) Detailed theoretical linear band structure (solid lines) in the vicinity of points I (left) and II (right) and the nonlinear band shifts (dashed lines) obtained from an analysis of the observed resonance angle shifts in Fig. 2.

stant. The refractive-index change Δn of the nonlinear optical polymer layer can be estimated by equating the observed Δk amplitude with the value of the refractive-index change in the 3D-FDTD band calculations, giving $\Delta n = +1.7 \times 10^{-3}$ and $\Delta n = +3.7 \times 10^{-3}$ at α and β, respectively. The detailed linear band structure and nonlinear band shifts in the vicinity of I and II are shown in Fig. 3(b). The shifts in the photon energy domain are towards the low photon energy side for both bands because of the positive susceptibility of the nonlinear optical polymer at the energy of the incident laser. On the other hand, the bands are shifted in opposite directions in the k domain, with a positive shift at α and a negative shift at β, which results from the positive and negative slopes of the band dispersion curves at these resonances. The agreement between the band structure features and the resonance angle shifts demonstrates the essential role of the band dispersion character in the nonlinear dynamics.

A direct comparison between the observed nonlinear responses and the dispersion characteristics for the two different band components provides information on the group velocity of the nonlinear optical properties and clarifies the enhancement effect of the nonlinear processes in PhC systems. In the present work, we found basic agreement between the ratio of the inverse group velocities of the photonic bands at I and II, $|\nu_{g\,I}|^{-1}:|\nu_{g\,II}|^{-1} = 1.0{:}2.4$, and the ratio of the refractive index changes at α and β, $\Delta n_\alpha:\Delta n_\beta = 1.0{:}2.2$, as well as between the ratio of the squared inverse group velocities at I and II, $|\nu_{g\,I}|^{-2}:|\nu_{g\,II}|^{-2} = 1.0{:}5.8$, and the ratio of the observed nonlinear resonance angle shifts at α and β, $|\Delta\theta_\alpha|:|\Delta\theta_\beta| = 1.0{:}5.0$. These results clearly demonstrate that the observed nonlinear changes are dominated by the band group velocities at the resonance conditions. The refractive-index change in these processes is considered to be enhanced in proportion to ${\nu_g}^{-1}$ because the electric field intensity of the resonance band mode is enhanced in proportion to ${\nu_g}^{-1}$ based on conservation of energy in the first approximation. The resonance angle shift is enhanced in proportion to ${\nu_g}^{-2}$ because the wave vector shift Δk, which depends on the resonance angle shift, is affected by the combination of the larger refractive-index change ($\propto {\nu_g}^{-1}$), and the larger k vector shift ($\propto {\nu_g}^{-1}$) arising from the slower group velocity, as shown in Fig. 3(b). Possible reasons for the experimental values being slightly lower than those predicted from the band structure are losses in the waveguide and differences in the extent of the resonant coupling for different band components.

Our data give direct evidence that the nonlinear optical changes arising from modifications of the photonic bands by an external laser field resonantly coupled to the photonic band (i.e., by purely optical means) in a 2D PhC waveguide with a Kerr nonlinearity are dominated by the dispersion nature and the group velocity of the photonic bands. Thus, with current technology, active manipulation of these nonlinear processes is a realistic possibility through engineering of the band dispersion and band group velocity characteristics. We note that the features and advantages of PhCs are also common to other nonlinear optical processes, suggesting the possibility of progress in all areas of nonlinear optics.

Although c/ν_g is limited to the order of 1 in conventional waveguides, ν_g in 2D PhC waveguides can be designed with a much greater degree of freedom, for example, extremely slow ν_g values, 30 to 90 times slower than c, have been experimentally observed [14,17]. The enhancement of the nonlinear optical changes resulting from the slower group velocity, such as demonstrated in this research, should lead to dramatic improvements in the performance of all-optical switching devices and will open up new possibilities in applications such as highly efficient dynamic dispersion management devices. Furthermore, the agreement between the observed nonlinear responses and the band dispersion characteristics means we can begin to engineer the nonlinear optical properties of PhC systems by controlling the PhC configuration.

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