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Ultraviolet second harmonic generation and sum-frequency mixing in nonlinear-optical polymer photonic-crystal waveguides

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1. Introduction

Research into and applications of nonlinear optical (NLO) processes in photonic crystals (PCs) are very exciting areas. Currently, however, the majority of research and development into PCs is being carried out in passive materials using the photonic bandgap. The combination of PC and NLO is expected to lead to exciting, new active functions and novel high-efficiency NLO applications such as high-efficiency frequency conversion and all-optical switching devices. However, the experimental realization and subsequent analysis of two-dimensional PC slab waveguides formed out of highly nonlinear host material has not yet been achieved because of the difficulty in obtaining high-aspect, high-quality PC profiles in the best-known nonlinear inorganic crystals such as $LiNbO_3$ and KH_2PO_4 (KDP).

Very recently, to overcome the above problems and fabricate a nonlinear PC waveguide, we proposed NLO polymer which exhibits good processability and highly nonlinear optics over a wide frequency range compared to inorganic crystals and semiconductor materials, as a candidate material for realizing nonlinear PC waveguides, and presented the first experimental results on inductively coupled plasma (ICP) etching characteristics of this kind of material.[1] However, in this two-dimensional PC slab waveguide structures formed out of NLO polymer, experimental behaviors of nonlinear optical processes have not been exhibited and investigated so far, which is essential for the realization of novel, high-efficiency nonlinear optical devices.

In this work, for this purpose, we report, for the first time, second harmonic generation (SHG) and sum-frequency mixing (SFM) in the NLO polymer PC waveguides in the ultraviolet region, and the fabrication of the structure suitable for these optical processes, and investigated the optical properties related to their photonic band structures.

2. Experimental

To construct the slab waveguide, a metal cladding layer of silver (thickness t = 500 nm) was vapor-deposited onto a Si substrate. Disperse Red 1 (DR1) doped poly(methylmethacrylate) (PMMA) is the NLO polymer used as the waveguide core layer, and was deposited on the metal cladding by spin coating and curing techniques. The concentration of DR1 in the core was about 10 wt.%. The magnitude of the second order nonlinearity of this material is proportional to the orientational order, as induced by some form of poling technique after all other processes have been completed.

To prevent and minimize plasma etching damage and optical scattering loss for the NLO core layer, the transparent semi-core as the patterning layer, which consists of the PMMA, was deposited on the extremely thin SiO_2 layer which used the etch stop layer on the NLO polymer. To pattern the PMMA, a 150 nm spin-on glass (SOG) hard mask, which has excellent durability in oxygen-based plasmas, was used. Resist patterns formed by electron beam lithography were transferred to the hard mask by ICP etching using CF_4/H_2 plasmas. Finally, the PMMA waveguide semi-core was patterned by ICP etching using the hard mask pattern. A mixture of O_2/Ar etching gas was chosen to minimize lateral etching. Sidewall profiles and surface morphology were evaluated by scanning electron microscopy (SEM). Angle-dependent optical reflectivity measurements of the PC slab waveguides were carried out using a collimated, polarized tungsten-halogen white light source, with reflectivity spectra measured by a Corrected Czerny-Turner spectrometer and a liquid-nitrogen-cooled CCD.

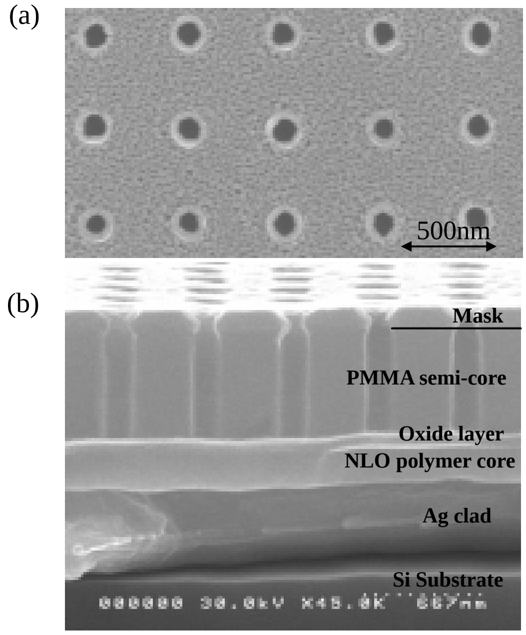

Fig. 1. (a) Plane-view and (b) cross-sectional SEM micrographs of the two-dimensional PC slab waveguides.

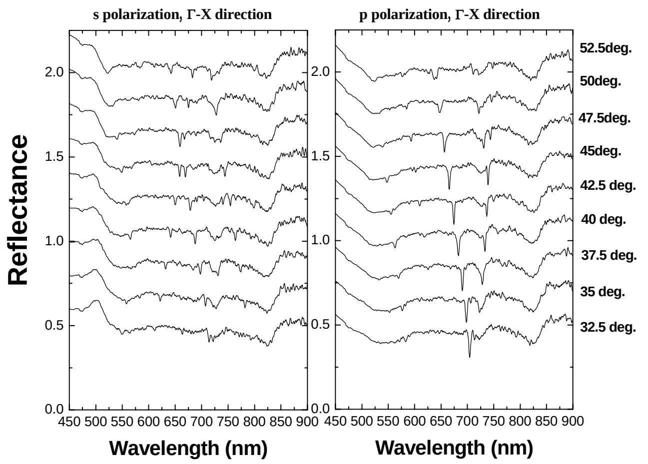

Fig. 2. The observed reflectance spectra for various angles of incidence with s- and p-polarization along the Γ-X lattice direction.

In SHG and SFG measurements, the excitation source we used was an optical parametric amplifier (OPA) in conjunction with a frequency doubled amplified Ti:Sapphire laser (operating at 800 nm). These measurements were performed for varying angles of incidence.

3. Results and discussion

Figure 1 shows plane-view and cross-sectional SEM micrographs of the two-dimensional PC slab waveguide fabricated using the optimal parameters for producing straight sidewalls. The PC were patterned with a square lattice of circular holes of diameter 120 nm and lattice constant 500 nm. As can be seen, excellent etching profiles with straight sidewalls were obtained in the PMMA semi-core layer. Although tapering of the mask edges by direct physical sputtering was observed, since the SOG mask can be removed by buffered HF etching, the does not significantly effect the final PC waveguides.

For this sample ($100\times100\mu m^2$), polarized angular dependent reflectivity measurements were performed along the Γ-X directions, with the s- and p-polarization reflectance spectra shown in Fig. 2. Several sharp dips can be seen in the spectra. These dips originate from resonant phenomena due to surface coupling between external free-photons and in-plane photonic bands at the resonance energies and in-plane wave vectors. The sharp dip structures in each reflectance spectrum therefore provide information about photonic band dispersion, not about the photonic band-gap and its angular dependent on the Bragg peak. The observed resonance wavelength clearly depends on the in-plane propagation lattice direction and the incident angle θ which determines the in-plane wave vector, $k = (2\pi A/\lambda)\sin\theta$ (where A is the lattice constant and θ is the angle of incidence). The photonic band structure of the waveguide was determined from the dispersion curves of the dip positions, and confirmed that a nonlinear photonic band structure was formed. This demonstrates that the PC waveguide structure was etched precisely.

Figure 3 shows the SHG intensity excited by 736 nm

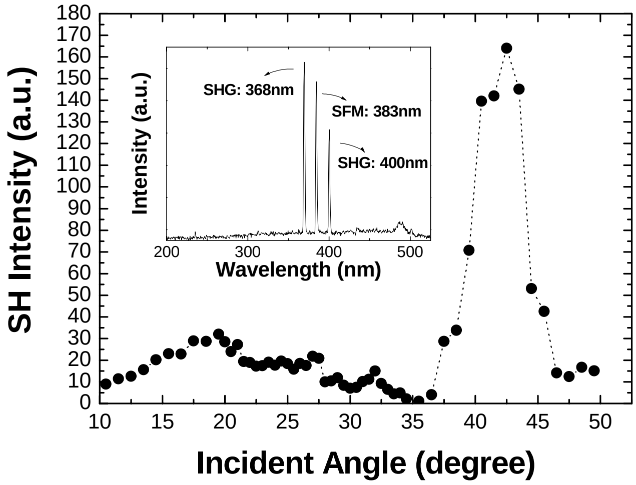

Fig.3. The SHG intensity vs. incident angle. The inset shows the SHG and SFM spectra at 42.5 deg.

OPA with a p-polarization as a function of incident angle, where the signal intensity was normalized by the intensity at the off-resonance angle (35 deg.). The inset shows the SHG and SFM spectra with the angle 42.5 deg. of incidence. The SHG (400nm) was generated by a residual Ti:Sapphire laser (800nm) in the PC waveguide. The ultraviolet SHG (368nm), and SFM (383nm) generated by mixing a Ti:Sapphire laser with OPA (736nm), were clearly observed as can be seen in the spectra. Moreover, the enhancement of SHG was dramatically observed around 42.5 deg. angle of incidence, which originates from the electric field enhancement due to the photonic band resonance. Relativity wide resonance peak was observed in this process. This shows that the dispersion of corresponding photonic band was small in this frequency region, which means that the group velocity of this band is low. These results exhibited a good agreement with the observed photonic band structure in Fig. 2.

4. Conclusion

We successfully fabricated the NLO polymer PC waveguides with straight sidewall at the suboptical wavelength scale, and observed sharp resonances originating from coupling to photonic band modes in the optical reflectance spectra of this waveguide. And finally, we reported, for the first time, second harmonic generation (SHG), sum-frequency mixing (SFM) in the ultraviolet region, and these strong enhancements originate from photonic band resonance in this waveguide, which opens the door to the realization of novel, high-efficiency nonlinear optical devices.

Acknowledgements

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References

[1] S. Inoue, K. Kajikawa, and Y. Aoyagi, Appl. Phys. Lett. 82, 2966 (2003)