Efficient second harmonic generation from large band gap II-VI semiconductor photonic crystal

H. Yang, P. Xie, S. K. Chan, Z. Q. Zhang, I. K. Sou, G. K. L. Wong, and K. S. Wong^{a)}

Department of Physics, Hong Kong University of Science and Technology, Clear Water Bay, Kowloon, Hong Kong, People's Republic of China

(Received 8 February 2005; accepted 5 August 2005; published online 23 September 2005)

Dramatic enhancement of second harmonic generation (SHG) near the photonic band edge was observed in a one-dimensional ZnSe/ZnMgS semiconductor photonic crystal (PC) structure. Over two orders of magnitude increase in SHG intensity was observed at the photonic band edge at ~ 1400 nm compared to the nonphase matching region. The maximum SHG conversion efficiency of 0.8% is observed in only seven micrometers length of crystal. This enhancement came from a combination of large ZnSe second order susceptibility coefficient ($\chi^{(2)}$), high density of optical modes and phase matching of the fundamental and second harmonic waves near the photonic band edge due to modification of the dispersion curve by the PC structure. © 2005 American Institute of Physics. [DOI: 10.1063/1.2061864]

In the last few years, considerable efforts were devoted to the studies of photonic crystals (PCs) because of their unique dispersion properties and potential application in novel optical device to control and manipulate light.^{1,2} The study of nonlinear optical properties such as the second harmonic generation (SHG) is of particular interest in PCs.³ It is well known that for bulk material, the SHG intensity I_{SH} is given by⁴

$$I_{\text{SH}} \propto I_F^2 L^2 (\chi^{(2)})^2 \frac{\sin^2(\Delta k L/2)}{(\Delta k L/2)^2}, \quad (1)$$

where I_F is the pump beam intensity at fundamental frequency ω , L is the sample length, $\chi^{(2)}$ is the second order optical susceptibility of the material, and Δk is the phase-mismatch which is given by $\Delta k = (n_{2\omega} - n_\omega)2\omega/c$. The parameter c is the speed of light in vacuum, $n_{2\omega}$ and n_ω are the refractive indexes at 2ω and ω , respectively. As shown in Eq. (1), one of the fundamental issues in the high conversion efficiency in SHG is the phase matching condition (i.e., $\Delta k=0$), which traditionally is achieved by using birefringent crystal or quasiphasematching structures based on sign reversion of the $\chi^{(2)}$ for every coherent length using electric field poling of ferroelectric materials.⁵⁻⁷ However, one can also utilize the one-dimension (1D) PC structures, which use spatial modulation of the linear refractive index rather than a modulation of the nonlinear susceptibility as quasiphasematching interaction for efficient SHG.⁸ In PCs, the dispersion of the linear materials can be precisely compensated by the dispersion introduced by the geometry of periodic structure, so it can exactly achieve the phase matching condition for harmonic generation in terms of an effective index of refraction. Thus, almost any material with good transmission properties in the wavelengths of interest can be used. As is well known, some semiconductors have very large $\chi^{(2)}$ but lacking birefringent and/or ferroelectric effects,⁹ can be used for fabrication 1D PC to enhance the SHG. Another remarkable property of the PC structure is associated with the transmission resonance that appears close to the band gap, where the optical field is strongly confined which consequently fur-

ther enhances the SHG. As recently shown by Scalora and co-workers,¹⁰⁻¹³ the simultaneous effective refractive index phase-matching and field enhancement at the photonic band edge are achievable in finite 1D PC. Thus, by proper design and choice of material for 1D PC, all the key parameters for efficient SHG indicated in Eq. (1) can be satisfied to produce a dramatic enhancement of second harmonic signal near the photonic band edge.

Some experimental work has been reported for the SHG in the 1D periodic structure based on semiconductor material.^{10,14-16} Dumeige *et al.*¹⁰ demonstrated the SHG in the GaAlAs/AlAs PC structure. However, the exact phase matching cannot be realized in this 1D PC because the index contrast between GaAlAs and AlAs is too small. More recently, the oxidation of AlAs layers to make GaAlAs/ AlO_x PC enabled perfect phase matching to produce very efficient SHG near the photonic band edge and the SH intensity scaling as the fifth power of the structure length instead of usually quadratic dependence was shown.¹⁴ SHG in the visible spectral region was shown in the ZnS/SrF₂ periodic structure where theoretical calculation indicates SH enhancement is mainly due to an increase in optical field intensity at fundamental frequency near the photonic band edge.¹⁵ In this letter, we demonstrate a dramatic enhancement of the SHG in a 1D large band gap II-VI semiconductor PC structure. This large enhancement of SHG is due to the simultaneous advantage of perfect phase matching condition and localization enhancement effect of the electromagnetic field near the photonic band edge in this 1D PC. The maximum conversion efficiency up to 0.8% was obtained in a PC structure composed of ZnSe/ZnMgS multilayer of only a few microns in total thickness, and it has the potential to produce SHG lights from blue to deep red spectral region.

We designed and fabricated a 1D finite semiconductor photonic band gap structure for SHG application. The simulated PC structure consists 25 periods of ZnSe/Zn_{0.8}Mg_{0.2}S for the high/low index layers with layer thickness of 156/131 nm, respectively. The calculation is based on the transfer matrix plane-wave propagation method,^{17,18} assuming nondepletion of the propagating beam. The refractive indexes of the II-VI compounds are based on Ref. 19. The

^{a)}Electronic mail: phkswong@ust.hk

FIG. 1. (a) Experimentally measured (dashed curve) and theoretically calculated (solid line) transmission of the 25 period ZnSe/ZnMgS PC structure versus the incidence wavelengths. The transmission measurement was done with the GaAs substrate removed. (b) The effective refractive index (solid line) and DOM (dashed line) as a function of wavelengths of the same PC. This structure is optimized for SHG at the photonic band edge at ~ 1410 nm. The figure inset shows the detail spectrum of the transmission between 600–750 nm near the second photonic band gap, and the dotted line is the average transmission data points with the Fabry-Perot oscillations filter out to show more clearly the transmission dip centered at ~ 670 nm.

transmission, effective index n_{eff} and density of modes (DOM) for this 1D PC are shown in Fig. 1. The simulation in Figs. 1(a) and 1(b) clearly shows that this ZnSe/ZnMgS PC have perfect phase matching (i.e., $n_{\text{eff},\omega} = n_{\text{eff},2\omega}$) at ~ 1410 nm and it coincides exactly with very high DOM region at the photonic band edge, resulting in efficient SHG at ~ 705 nm.

The design structure is grown on GaAs (100) substrate by molecule beam epitaxy (MBE) technique. The details of the structure of PC were measured by scanning electron microscope (SEM) showing the width of the ZnSe and $\text{Zn}_{0.8}\text{Mg}_{0.2}\text{S}$ layers in the PC were about 156 nm and 131 nm, respectively, as required by the simulation. However, the SEM image showed that the period thickness gradually increases from the substrate interface to the top layer and the difference in thickness between the bottom and top layer is approximately 10%. This slight aperiodicity in this ZnSe/ZnMgS multilayer structure is due to gradual increase in the beam flux during the long growth time used for this sample. Nevertheless, the measured transmission spectrum shown in Fig. 1(a) indicates our sample is a fairly good 1D PC with a clear Bragg stop band and many transmission resonance peaks. The location of the stop band around 1300 nm–1400 nm of the experimental curve is in good agreement with the theoretical result as shown in Fig. 1(a). But the second stop band centered at ~ 680 nm was not be clearly observed and the transmission resonance peaks no longer coincided with the theoretical curve at wavelengths

FIG. 2. The magnitude of the forward second harmonic (bare circle) and backward second harmonic (solid circle) generated from the PC vs different input laser wavelengths. The transmission of the PC is also shown for reference (dotted line).

away from the fundamental band edge as a result of this slight aperiodicity. However, detail inspection of the transmission spectrum (see Fig. 1 inset) shows indeed there is a slight dip in the transmission curve center around the second stop band (660–700 nm). This result shows the existence of the second stop gap, although slight disorder and absorption in our sample smear out this effect. The rapid drop in transmission around 500 nm in the experimental curve is due to strong absorption near the ZnSe band gap ($E_g \sim 460$ nm), which we did not take into account in the simulation.

An optical parametric amplifier tunable from 0.5 to 1.9 μm with pulse duration of ~ 70 ps was used to generate the fundamental wave for the SHG measurements. The excitation intensity is controlled by variable neutral density filter and a spectrometer coupled to a CCD detects the SHG signal. Figure 2 shows the forward and backward SH intensity as a function of wavelengths at excitation intensity of $200 \text{ MW}/\text{cm}^2$. A sharp increase in the magnitude of the forward SH signal when the pumping laser wavelength is about 1413 nm is shown in Fig. 2. This result is consistent with the theoretical simulation presented in Fig. 1, which shows that the dramatic enhancement of the SHG in this PC as a result of phase matching and high DOM at the photonic band edge. Only one enhanced SHG peak appear at the long wavelength edge of the photonic band clearly show that this SHG enhancement is not simple due to cavity effect where the increase in density of modes near the photonic band edges enhance the SHG. In this simple cavity case, we would expect to see two SHG peaks at the long and short wavelength edges of the photonic stop band [see DOM simulation in Fig. 1(b)]. Therefore, our results clearly demonstrated that the SHG enhancement is due to both good phase matching condition and field localization near the photonic band edge. The backward SH signal peaked at a similar wavelength position but the signal intensity is about five times smaller than the forward SH signal. Our simulation also indicates that the backward SH is about six times smaller than the forward SH signal, again in good agreement between the theory and experiment for the backward SHG signal. To further prove that the enhanced SHG effect is indeed the result of a 1D PC structure, we compared the SH signals of the present PC sample to a multilayer film of the same ZnSe/ZnMgS material composition but nonperiodic structure (100 random lay-

FIG. 3. The measured signal intensities of the SH with 50 layers PC (solid line) and the SH of 100 layers of nonperiodic structure (dashed line) at the same incident laser intensity.

ers) as shown in Fig. 3. The transmission curve of the random sample does not show any Bragg stop band proving that the ZnSe/ZnMgS layers are nonperiodic. The SH signal of the periodic structure is almost two orders of magnitude larger than the nonperiodic one (note that the total thickness of the nonperiodic sample is twice as thick as the 1D PC sample also), which shows the SHG enhancement is indeed related to the periodic structure of the sample.

Figure 4 shows the theoretically calculated and experimentally measured SHG conversion efficiency as a function of wavelengths. The SHG data is converted to conversion efficiency by using a calibrated energy meter and taking into account the wavelength dependence of the filter, spectrometer grating and CCD. Experimental excitation intensity of 200 MW/cm² and $\chi^{(2)} = 1 \times 10^{-7}$ esu for ZnSe were used for the simulation. Reasonably good agreement between experiment and theory for the SHG peak positions, the excitation wavelength response, and absolute magnitude of the conversion efficiency is obtained; despite the fact that the actual sample is slightly aperiodic and various losses such as scattering and/or absorption were not taken into account in the simulation. Figure 4 inset shows the SH intensity as a func-

FIG. 4. The experimentally measured and theoretically calculated SHG conversion efficiency versus wavelengths at excitation intensity of 200 MW/cm². The inset is the plot of the square root (Sqrt) of the SH intensity vs the input laser energy, and a straight line dependence is obtained.

tion of fundamental pump beam energy and the expected quadratic behavior was observed. The maximum SHG conversion efficiency of 1.5×10^{-5} at $\lambda_{SH} \approx 707$ nm was obtained at a pumping intensity of 200 MW/cm² as shown in Fig. 4. However, when the incident laser intensity was increased to ~ 3 GW/cm², a maximum conversion efficiency of 8×10^{-3} (i.e., 0.8%) was obtained, to the best of our knowledge, this is the highest conversion efficiency obtained directly from PC of only a few micrometers length.

In conclusion, we have observed the enhancement of nearly phase-matched SHG in 1D multilayer semiconductor photonic band gap structure. This SHG enhancement near the photonic band edge is due to simultaneous availability of the high DOM of fundamental and SH waves and the exact phase matching condition. The theoretical simulation of the spectral response and SHG conversion efficiency is in good agreement with the experimental results. SHG conversion efficiency up to 0.8% from only a few micrometers semiconductor PC was demonstrated. With improvement in growth technique to produce better periodic PC structure resulting in more exact phase matching and higher DOM, much higher SHG conversion efficiency than presently achieved can be expected. Overall, our results exhibit a good prospect for PC in nonlinear optics applications in the visible spectral region.

This work is partially supported by the Research Grant Council of Hong Kong (grant Nos. HKUST6063/02P and 605804).

- ¹E. Yablonovitch, Phys. Rev. Lett. **58**, 2059 (1987); S. John, *ibid.* **58**, 2486 (1987).
- ²J. D. Joannopoulos and P. R. Villeneuve, Nature (London) **386**, 143 (1997).
- ³*Nonlinear Photonic Crystals*, edited by R. E. Slusher and B. J. Eggleton (Springer, Berlin, 2003).
- ⁴See, for example, Y. R. Shen, *The Principles of Nonlinear Optics* (Wiley, New York, 1984).
- ⁵H. Cheng and P. B. Miller, Phys. Rev. **134**, A683 (1964).
- ⁶J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan, Phys. Rev. **127**, 1918 (1962); P. A. Franken and J. F. Ward, Rev. Mod. Phys. **35**, 23 (1963).
- ⁷M. M. Fejer, G. A. Magel, D. H. Jundt, and R. L. Byer, IEEE J. Quantum Electron. **28**, 2631 (1992).
- ⁸N. Bloembergen and A. J. Sievers, Appl. Phys. Lett. **17**, 483 (1970); J. P. Van der Ziel and M. Ilegem, *ibid.* **28**, 437 (1976).
- ⁹*Handbook of Lasers*, edited by R. J. Pressley (Chemical Rubber Co., Cleveland, Ohio, 1971).
- ¹⁰Y. Dumeige, P. Vidakovic, S. Sauvage, I. Sagnes, J. A. Levenson, C. Sibilia, M. Centini, G. D. Aguanno, and M. Scalora, Appl. Phys. Lett. **78**, 3021 (2001).
- ¹¹M. Scalora, M. J. Bloemer, A. S. Manka, J. P. Dowling, C. M. Bowden, R. Viswanathan, and J. W. Haus, Phys. Rev. A **56**, 3166 (1997).
- ¹²M. 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).
- ¹³M. Centini, M. Scalora, C. Sibilia, G. D'Aguanno, M. Bertolotti, M. J. Bloemer, and C. M. Bowden, J. Opt. A, Pure Appl. Opt. **2**, 121 (2000).
- ¹⁴Y. Dumeige, I. Sagnes, P. Monnier, P. Vidakovic, I. Abram, C. Mériadec, and A. Levenson, Phys. Rev. Lett. **89**, 043901 (2002).
- ¹⁵A. V. Balakin, V. A. Bushuev, N. I. Koroteev, B. I. Mantsyzov, I. A. Ozheredov, A. P. Shkurinov, D. Boucher, and P. Masselin, Opt. Lett. **24**, 793 (1999).
- ¹⁶T. V. Dolgova, A. I. Maidikovski, M. G. Martemyanov, A. A. Fedyanin, O. A. Aktsipetrov, G. Marowsky, V. A. Yakovlev, G. Mattei, N. Ohta, and S. Nakabayashi, J. Opt. Soc. Am. B **19**, 2129 (2002).
- ¹⁷D. S. Bethune, J. Opt. Soc. Am. B **6**, 910 (1989).
- ¹⁸N. Hashizume, M. Ohashi, T. Kondo, and R. Ito, J. Opt. Soc. Am. B **12**, 1894 (1995); P. Xei and Z. Q. Zhang, Phys. Rev. A **69**, 053806 (2004).
- ¹⁹H. H. Li, J. Phys. Chem. Ref. Data **13**, 103 (1984).