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APPLIED PHYSICS LETTERS VOLUME 84, NUMBER 16 19 APRIL 2004

Noncollinear type-II second-harmonic generation in a $Al_{(0.3)}Ga_{(0.7)}As/Al_2O_3$ one-dimensional photonic crystal

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(Received 27 October 2003; accepted 26 February 2004)

We demonstrate noncollinear type-II second-harmonic generation in one-dimensional photonic crystals. A 15-period $Al_{(0.3)}Ga_{(0.7)}As/Al_2O_3$ structure, 3.5 μm long, was designed, fabricated, and experimentally characterized. We measured an effective nonlinearity of (52±12) pm/V in perfect phase-matching conditions. [DOI: 10.1063/1.1713039]

Recently, interest in second-order nonlinear optical phenomena occurring in periodic structures[1–5] has grown considerably. These structures, usually referred to as photonic band gap (PBG) structures, may exhibit efficient parametric processes, such as second-harmonic generation (SHG) or frequency up- and down-conversion. In particular, during the last decade finite one-dimensional (1D), periodic or quasi-periodic multilayer stacks, the simplest types of PBG structures, have been widely investigated theoretically and experimentally.[6–12] The main advantage that these artificial structures offer is the ability to enhance the nonlinear response normally associated with bulk $\chi^{(2)}$ materials thanks to the localization of light at frequencies tuned at the photonic band edge, and the simultaneous availability of effective phase matching (PM) conditions.[6–9] In fact, the natural dispersion of ordinary materials can be compensated by introducing geometrical dispersion, which is brought about and controlled by properly juxtaposing a high and a low index material. This effect may provide enough anomalous dispersion across the photonic band gap to make exact PM conditions possible even for materials, such as III–V semiconductors, which cannot usually be phase-matched.[1,12]

In this letter we present experimental evidence of phase-matched, noncollinear, type-II SHG in a photonic crystal. We used a 1D structure composed of 15 $Al_{(0.3)}Ga_{(0.7)}As/Al_2O_3$ periods. The structure was designed according to a theoretical representation of the effective index of refraction[6] for a multilayer structure, such that multiple scattering events are taken into account to all orders, and the result is a bulk medium with an effective index of refraction n_{eff}. Using this approach, PM conditions can be found by applying the effective momentum conservation law in 1D structures $\mathbf{k}_{\mathrm{eff}}^{(2\omega,p)} - \mathbf{k}_{\mathrm{eff}}^{(\omega,p)} - \mathbf{k}_{\mathrm{eff}}^{(\omega,s)} = \mathbf{0}$ and projecting it onto two components, one along the plane of the layers, and the other one perpendicular to it:

$$2n_{\mathrm{eff}}^{(2\omega,p)} - n_{\mathrm{eff}}^{(\omega,p)} - n_{\mathrm{eff}}^{(\omega,s)} = 0, \quad (1a)$$

$$2\sin(\theta_{2\omega,p}) - \sin(\theta_{\omega,p}) - \sin(\theta_{\omega,s}) = 0, \quad (1b)$$

where θ corresponds to the external incidence angle formed by each $\mathbf{k}$ vector with the normal to the sample's surface; the labels p, s, ω, and 2ω refer to the fields' polarizations and frequencies. For a finite periodic structure, Eq. (1a) is automatically satisfied when the two fundamental fields are tuned at the band edge resonance, and the SH is tuned at the second peak near the second-order band gap.[6] Equation (1b) depends only on the incidence angles of the fields, as if the $\mathbf{k}$-vector components did not feel the refractive index discontinuity.

According to Eq. (1), exact, noncollinear PM for SHG of 1510 nm fundamental wavelength was found for a 15-period structure composed of $Al_{(0.3)}Ga_{(0.7)}As$ (160 nm)/Al_2O_3 (97 nm). Total sample thickness was 3.5 μm. A multilayer structure of AlGaAs/AlAs was first grown on a 500-μm-thick (100) GaAs substrate by solid-source molecular beam epitaxy. A 30 nm GaAs cap was grown on the top of the multilayer to protect the surface from subsequent oxidation. Successively, 20-μm-wide trenches were etched through the entire multilayer by reactive-ion etching in order to expose the AlAs layers for lateral oxidation. The AlAs layers were oxidized by heating the sample at 410 °C for 105 min in a H_2O atmosphere created by bubbling N_2 in a water bath at 85 °C.[13] The schematic layout of the multilayer structure and the real picture of the sample's top surface are shown in Fig. 1(a); its active area is formed by 100 μm broad stripes separated by 20 μm etched trenches.

A spectral analysis of the structure was performed with a micro-reflectometer connected to a Fourier-transform ana-

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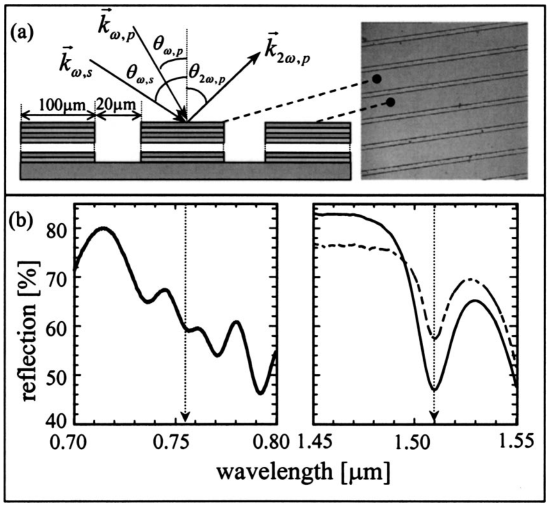

FIG. 1. (a) Layout of the multilayer structure (left view) and picture of the sample's top surface (right view). The largest stripes (100 μm) form the active area of the photonic crystal. (b) Experimental reflection spectral analysis. Left view, spectrum of p-polarized light around the SH wavelength for an incident angle of 42°; the second-order band gap for p polarization is the largest peak on the left-hand side. Right view, reflection spectra for p-polarized light incident at 30° (solid line) and for s-polarized light incident at 57° (dashed line). The vertical arrows indicate the tuning of the fundamental (1510 nm) and the SH (755 nm) wavelengths.

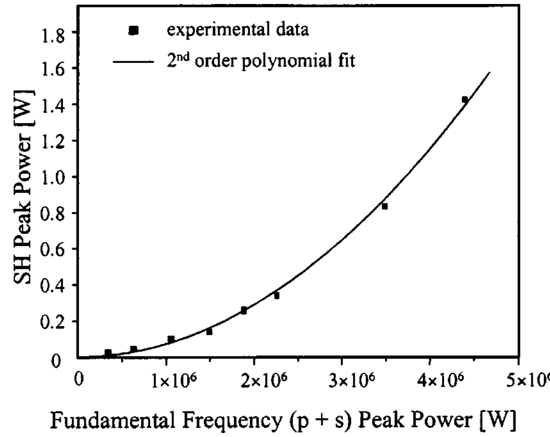

FIG. 3. Measured SH peak power vs total fundamental ($p+s$) beam power (dots). The solid line represents the second-order polynomial fit function needed to calculate the process efficiency.

lyzer. Reflection spectra were recorded at several incident angles, for both s and p polarizations. The spot-size was set to fit a single active stripe, a procedure that allowed checking the sample homogeneity by comparing different spectra from several stripes. Figure 1(b) depicts the experimental reflectivity spectra around the fundamental wavelength for p-polarized beam incident at an external angle of 30°, and for s-polarized beam incident at 57°, together with the "p" spectrum around the second-harmonic wavelength for an angle of 42°. For these angles, the two fundamental beams at 1510 nm are tuned to the first resonance peak closest to the band gap, while the p-polarized SH beam is tuned to the second peak close to the second-order band gap. As discussed earlier, this condition ensures noncollinear type-II phase matching at 1510 nm, as expected from the design.

The experimental SHG was obtained using the setup shown in Fig. 2. The laser system consisted of an amplified Ti-sapphire laser that pumped a parametric amplifier. The outgoing beam provided 2-ps-long pulses at 1 kHz repetition rate, with energy of about 25 μJ. A wave-plate rotated the polarization to obtain, after the polarizing beam splitter, two cross-polarized beams whose intensities were controlled by the variable attenuator. Both beams were focused on the sample by the same lens (focal length 500 mm) down to 180-μm-wide spot sizes. The two cross-polarized beams were sent to the sample with a relative angle of 27°. Their energies were varied up to a maximum of 4.5 μJ for each beam, corresponding to a peak intensity of approximately 9 GW/cm^2. In our conditions, the PBG acts like a resonant cavity; thus the SH is generated in both forward and backward directions. According to our theoretical model,[11] the conversion efficiency ($\eta = I_{SH}/I_{pump}$) in nondepleted pump regime can be written as

$$\eta^{(+,-)} = \frac{8\pi^2 |d_{eff}^{(+,-)}|^2 L^2 I_{pump}}{\epsilon_0 c \lambda^2 n_{eff}^{(\omega,p)} n_{eff}^{(\omega,s)} n_{eff}^{(2\omega,p)}}, \tag{2}$$

where λ is the pump wavelength in vacuum, $(+,-)$ stand for forward and backward directions, and $d_{eff}^{(+,-)}$ is the effective coupling coefficient containing the information on the fields overlap inside the structure, as described in Ref. 11. We note that Eq. (2) is formally equivalent to the equation that describes a phase-matched bulk medium, where the refractive index and the nonlinear coupling coefficient are replaced by effective quantities. Experimentally we were able to measure only the reflected SH because of the strong absorption present in the GaAs substrate at the SH wavelength. Figure 3 shows the measured reflected SH peak power versus the fundamental beam power. The experimental points are well fitted by a second-order polynomial function; this means that the process followed the theory of SHG in the nondepleted pump approximation. The measured conversion efficiency corresponding to the maximum fundamental beam intensity was $\eta_{PBG}^{(-)} = (5.6 \pm 0.1) \times 10^{-7}$. The effective nonlinearity shown by the sample was then calculated by comparing this result with the one obtained by a reference nonlinear crystal, operating in transmission, under the same conditions of incident angles and fundamental beam intensity. For this calibration we used a crystal known in the literature with the acronym DAST.[14] The crystal was 1.5 mm long, with

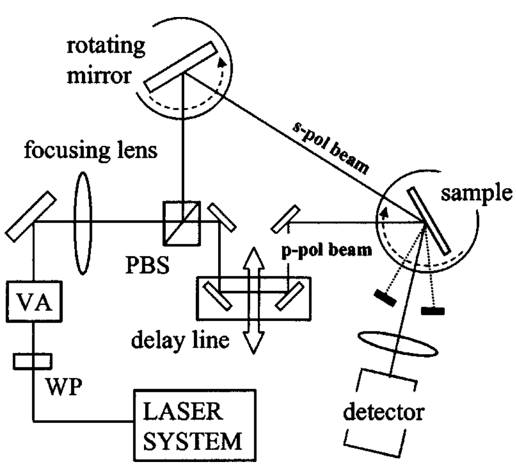

FIG. 2. Layout of the experimental setup. The SH signal was detected by a Si detector (response time about 2 ns) connected to an oscilloscope with 500 MHz bandwidth.

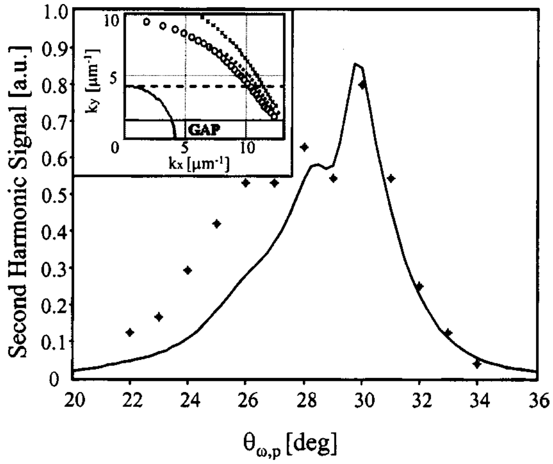

FIG. 4. Experimental (*) and theoretical (solid line) angular dependence of the generated p-polarized SH signal. The horizontal axis refers to the fundamental p-polarized beam incidence angle. The angle under which the SH is generated is varying fulfilling relation 2(b). Inset. Bloch equifrequency curves calculated at the fundamental (circles: p-pol, crosses: s-pol) and at p-pol second harmonic (dots, scaled by a 2 factor) frequencies. The solid-line circle represents the relation $k_x^2+k_y^2=\omega^2/c^2$ for the incident field. Photonic crystal modes can be excited with a k_y value between the dashed line and the gap area.

$d_{\rm eff}=(15\pm3)$ pm/V for SHG at 1530 nm.[15] The measured conversion efficiency was $\eta_D^{(+)}=(3.4\pm0.1)\times10^{-2}$. Substituting the experimental data and the refractive indices for DAST[14] and for the sample[6] in Eq. (2), we calculated by comparison:

$$d_{\rm eff}^{(\rm PBG-)}=\frac{d_{\rm eff}^{(D)}L_D}{L_{\rm PBG}}\sqrt{\frac{(n_{\rm eff}^{(\omega,p)}n_{\rm eff}^{(\omega,s)}n_{\rm eff}^{(2\omega,p)}\eta^{(-)})_{\rm PBG}}{(n^{(\omega,p)}n^{(\omega,s)}n^{(2\omega,p)}\eta^{(+)})_D}}. \quad (3)$$

Thus, the stratified structure showed an effective nonlinear coefficient $d_{\rm eff}^{(-)}=(52\pm12)$ pm/V.

Angular measurements of the SHG have been performed keeping the relative angle between the incident beams constant to 27°, and by varying the sample orientation. In Fig. 4 we show the generated power versus the angle between the sample normal and the fundamental p-polarized directions. The solid line represents the theoretical predictions:[11] our model developed for monochromatic plane waves is in qualitative agreement with the experimental data. We note that, in spite of what occurs in bulk media, the SH signal is generated in a broad angular range. The inset of Fig. 4 shows Bloch diagrams calculated for the fundamental and SH fields. Since the structure is embedded in air, there is a limit on the photonic-crystal modes that can be excited from outside. The dashed line sets the maximum value for the k_y component corresponding to a 90° external angle. PM conditions are almost fulfilled for any angle above the gap region. The presence of two peaks in the experimental angular emission is related to enhancement effects that are typical of structure of finite length. The main peak is obtained when the overlap between the two pump fields is maximized and perfect PM is achieved (30°). The second peak at 28° corresponds to band edge tuning for the SH. The strong SH field localization maintains the efficiency of the process, although the pump fields are slightly off resonance.

In conclusion, perfect PM conditions were achieved by using a material ($Al_{(0.3)}Ga_{(0.7)}As$) that cannot naturally be phase-matched because of its optical isotropy. We have provided experimental evidence of a relatively efficient, noncollinear, type-II SHG in one-dimensional photonic crystal 3.5 μm thick, i.e., more than twice the coherence length of bulk $Al_{(0.3)}Ga_{(0.7)}As$ (approximately 1.5 μm). The structure showed an effective nonlinearity $d_{\rm eff}^{(-)}=(52\pm12)$ pm/V.

The authors kindly acknowledge L. Andreani and M. Galli for measurements of the linear spectra. This work was performed in conjunction with the FemtoLab INFM network.

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