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Noncollinear type-II second-harmonic generation in a Al\(_{(0.3)}\)Ga\(_{(0.7)}\)As/Al\(_2\)O\(_3\) one-dimensional photonic crystal

A. Bosco, M. Centini, L. Sciscione, C. Sibilia, \(^a\) E. Fazio, and M. Bertolotti
Istituto Nazionale di Fisica della Materia, INFM, at Dipartimento di Energetica—Università di Roma “La Sapienza,” Via A. Scarpa 16, I-00161 Roma, Italy

A. Fiore
Institute of Quantum Electronics and Photonics—École Polytechnique Fédérale de Lausanne, CH-1015 Lausanne, Switzerland and Institute of Photonics and Nanotechnology, CNR, via del Cineto Romano 42, 00156 Roma, Italy

A. Convertino and L. Cerri
Istituto per lo Studio dei Materiali Nanostrutturati, CNR, via Salaria Km. 29.300, 00016 Roma, Italy

M. Scalora
Weapons Sciences Directorate, AMRDEC-WS-ST, RD&EC, U.S. Army Missile Command, Building 7804, Redstone Arsenal, Alabama 35898-5000

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We demonstrate noncollinear type-II second-harmonic generation in one-dimensional photonic crystals. A 15-period Al\(_{(0.3)}\)Ga\(_{(0.7)}\)As/Al\(_2\)O\(_3\) structure, 3.5 \(\mu\)m long, was designed, fabricated, and experimentally characterized. We measured an effective nonlinearity of (52±12) pm/V in perfect phase-matching conditions. © 2004 American Institute of Physics.

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\(_2\)O\(_3\) 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 \(n_{\text{eff}}\). Using this approach, PM conditions can be found by applying the effective momentum conservation law in 1D structures \(\mathbf{k}_{\text{eff}}^{(2\omega,p)} - \mathbf{k}_{\text{eff}}^{(\omega,s)} = \mathbf{0}\) and projecting it onto two components, one along the plane of the layers, and the other one perpendicular to it:

\[
2 n_{\text{eff}}^{(2\omega,p)} - n_{\text{eff}}^{(\omega,p)} - n_{\text{eff}}^{(\omega,s)} = 0,
\]

\[
2 \sin(\theta_{2\omega,p}) - \sin(\theta_{\omega,p}) - \sin(\theta_{\omega,s}) = 0,
\]

where \(\theta\) corresponds to the external incidence angle formed by each \(\mathbf{k}\) vector with the normal to the sample’s surface; the labels \(p\), \(s\), \(\omega\), and \(2\omega\) 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\(_2\)O\(_3\) (97 nm). Total sample thickness was 3.5 \(\mu\)m. A multilayer structure of AlGaAs/AlAs was first grown on a 500-\(\mu\)m-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-\(\mu\)m-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\(_2\)O 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 \(\mu\)m broad stripes separated by 20 \(\mu\)m etched trenches.

A spectral analysis of the structure was performed with a micro-reflectometer connected to a Fourier-transform ana-
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 $\mu$J. 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 $\mu$J for each beam, corresponding to a peak intensity of approximately 9 GW/cm². 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, the conversion efficiency ($\eta = I_{\text{SH}}/I_{\text{pump}}$) in nondepleted pump regime can be written as

$$\eta^{(+,-)} = \frac{8\pi^2|d_{\text{eff}}^{(+,-)}|^2L^2I_{\text{pump}}}{\epsilon_0c\lambda^2|r_{\text{eff}}^{(0,p)}(u,p)|^2|r_{\text{eff}}^{(0,s)}(2u_s,p)|^2},$$

where $\lambda$ is the pump wavelength in vacuum, (+,−) stand for forward and backward directions, and $d_{\text{eff}}^{(+,-)}$ 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 $\eta_{\text{PBG}} = (5.6\pm0.1)\times10^{-7}$. 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. The crystal was 1.5 mm long, with...
The solid line represents the theoretical predictions: our model developed for monochromatic plane waves is in qualitative agreement with the experimental data. We note that, in comparison: the experimental data and the refractive indices for the stratified structure showed an effective nonlinearity $d_{\text{eff}}^\text{PBG}=52\pm12$ pm/V.

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 $\mu$m thick, i.e., more than twice the coherence length of bulk Al$_{(0.3)}$Ga$_{(0.7)}$As (approximately 1.5 $\mu$m). The structure showed an effective nonlinearity $d_{\text{eff}}^\text{PBG}=52\pm12$ pm/V.

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