Enhancement of visible second-harmonic generation in epitaxial GaN-based two-dimensional photonic crystal structures

Gabriele Vecchi a)
INFM and Dipartimento di Fisica “A. Volta,” Università degli Studi di Pavia, Via Bassi 6, I-27100 Pavia, Italy

Jérémie Torres, Dominique Coquillat, and Marine Le Vassor d’Yerville
GES-UMR 5650, CNRS-Université Montpellier II, 34095 Montpellier Cedex 5, France

Andrea Marco Malvezzi
INFM and Dipartimento di Elettronica, Università degli Studi di Pavia, Via Ferrata 1, I-27100 Pavia, Italy

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Second-harmonic (SH) radiation generated in reflection is measured from the surface of a two-dimensional triangular photonic crystal in a GaN layer. A very large SH enhancement is observed when the incident radiation is resonant with a leaky photonic mode. The potential of second-harmonic generation as a tool for photonic band mapping is also envisaged. The extended transparency window of III-nitride wide band gap semiconductors coupled with large nonlinearities is an appealing feature pointing toward light control and manipulation in photonic structures. © 2004 American Institute of Physics. [DOI: 10.1063/1.1649800]

The nonlinear properties of the wide-band gap III–V material systems like gallium nitride (GaN), aluminum nitride (AlN) have been recently investigated1,2 for their potential optoelectronic application, such as optical wavelength conversion, in the visible and near-UV. In particular, GaN-based photonic band gap materials are of special interest for the wide transparency range, which makes possible nonlinear frequency generation and manipulation from the IR up to UV.3

In this letter we present detailed second-harmonic generation (SHG) studies in GaN two-dimensional photonic crystals, aimed at obtaining enhanced conversion efficiency by coupling the pump [and possibly the second-harmonic (SH)] fields with the photonic structure.4–6 We have chosen to probe the photonic crystal (PhC) slab by injecting pump radiation in the quasi-guided modes propagating out of the waveguide plane. Therefore the whole energy–momentum space above the light line can be explored by collecting the reflected SH generated by incoming pump pulses.

The structure investigated consists of a 260-nm-thick GaN waveguide epitaxially grown on a sapphire substrate. The photonic pattern is a triangular lattice of circular holes characterized by an average air-filling factor estimated at 22% and lattice constant \( a = 500 \) nm.7

Variable-angle linear reflectance and transmittance measurements8,9 yield the dispersion of photonic bands in the 0.45–0.75 \( a/\lambda \) normalized frequency range for both s- and p-polarized incident light along the two high symmetry directions, as shown in Fig. 1. The dispersion of photonic modes calculated (open circles) by using the scattering matrix method9 matches well with the experimental results (closed circles). This agreement represents a firm starting point for an accurate search for resonant conditions in the nonlinear investigations.

The source of excitation for SHG measurements is a Ti:Al\(_2\)O\(_3\) laser system providing pulses \( \approx 80 \) fs in duration with an 80 MHz repetition rate and a tunability range between 700 and 1000 nm. The average power on sample can reach 200 mW. Pulses are focused down to 30–50 \( \mu \)m in diameter onto the sample after spectral filtering and intensity and polarization control. The SH radiation is collected by an

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4Electronic mail: vecchi@fisicavolta.unipv.it

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FIG. 1. Photonic band dispersion diagram above the light lines shown along the directions of symmetry \( \Gamma \rightarrow K \) and \( \Gamma \rightarrow M \) for the s- (a) and p- (b) incident polarization. Closed (open) circles denote experimental (calculated) data. The modes are labeled according to the \( \Gamma \rightarrow K \) direction for both s and p polarization. Labeling along the \( \Gamma \rightarrow M \) direction refers to the possibility of different polarization state for the same mode (see Ref. 7).
optical fiber (1 mm core diameter) and delivered in front of a cooled photomultiplier after passing through color filters which reject the pump radiation. The geometry of the apparatus involves the angle of incidence \( \theta \) of the pump radiation, the angle \( \phi \) between the plane of incidence and the \( \Gamma - M \) direction of the photonic crystal and the polar diffraction angle \( \theta' \) measured from the normal to the sample. In the experimental setup\(^{10}\) all these angles are variable via remotely controlled motorized actuators. The detection of the SH signal is performed using a light chopper which modulates the pump radiation. The photomultiplier signal is then recorded by an averaging oscilloscope so that the amplitude of the signal with respect to background is evaluated.

The SH signals detected show a very wide amplitude variation as a function of geometry and pump wavelength. In particular, very sharp and pronounced peaks are observed whenever resonant conditions are approached between laser pump and photonic band energies. In Fig. 2(a) [2(b)] we report measured SH signals reflected from the PhC and the unpatterned GaN slab surfaces as a function of incident angle \( \theta \) (azimuthal angle \( \phi \)), for \( p \)-polarized pump beam at \( \lambda = 892\) nm (\( \omega a/2\pi c = 0.56 \)). The curve in Fig. 2(a) is obtained at \( \phi = 24^\circ \), close to the \( \Gamma - M \) direction of the photonic lattice. Closed (open) squares denote the SH field generated by the PhC (unpatterned GaN). The SH field generated from the PhC has a very high intensity at \( \theta = 14^\circ \) (note the vertical log scale). Identification of this pronounced peak appears somewhat troublesome, due to overcrowding of photonic bands near the \( \Gamma \) point. After combined analysis of linear and nonlinear measurements, we can attribute the peak at \( \theta \approx 14^\circ \) to the coupling between incident radiation and the \( 3p \) mode, while the second peak visible at around \( 52^\circ \) comes from the \( 4s \) mode.

The azimuthal scan is instead performed at \( \theta = 13^\circ \), i.e., close to the first peak described above, and it shows that the SH maxima come at \( \phi = 24^\circ \) and \( \phi = 36^\circ \), near the \( \Gamma - M \) direction. With the azimuthal measurement reported in Fig. 2(b) we scan the whole Brillouin zone with fixed energy and fixed in-plane wave vector component. As the triangular lattice is symmetrical with respect to the \( \Gamma - M \) direction (\( \phi = 30^\circ \)), the observed peak also appears with \( 60^\circ \) periodicity in all the reciprocal space. The appearance of a double peak instead of a single one means that the band is cut twice around the \( \Gamma - M \) symmetry for that selected energy. A smaller peak also appears along the \( \Gamma - K \) direction. It corresponds to the coupling between the \( p \)-polarized pump beam and the \( 2p \) photonic mode. The main result from Fig. 2(b) is the increase in SH intensity generated by the PhC with respect to the one generated by an unpatterned GaN layer under the same conditions. From the vertical logarithmic scale we argue that the SH field generated in reflection by the PhC is up to \( 2700 \) times larger. The interaction between the incident light and the photonic structure is expected to reach its maximum near the \( \Gamma \) point, due to the low group velocity of the quasiguided mode close to the center of first Brillouin zone. The observed extremely strong enhancement of SH intensity, which resulted in blue radiation spots visible on paper to the naked eye, is experimental evidence of the statement above.

In Fig. 3 we plot the SH reflected signals as a function of azimuthal angle, for \( s \) polarization and for three different choices of \( (\theta, \lambda) \). These measurements were meant to follow the band dispersion behavior across the Brillouin zone.\(^{11}\) In particular this was done for the \( 4s \) mode by the peaks that we observe symmetrically positioned with respect to the \( \Gamma - K \) high symmetry direction. Each \( (\theta, \lambda) \) set was selected from the experimental linear band dispersion to peak exactly along the \( \Gamma - K \) azimuthal direction, \( (\phi = 0^\circ) \). The appearance of double SH peaks therefore can reveal either an uncertainty over the chosen pump wavelength or a discrepancy between the experimental points in Fig. 1 and true photonic band position in the energy–momentum space. In the scans performed at \( 820 \) and at \( 807 \) nm the weaker peaks symmetrically emerging in an intermediate azimuthal position come from the coupling to the \( 4p \) photonic mode.

The observed PhC-enhanced SH overall conversion efficiency is even higher if we remember that also diffracted SH beams are present. Due to geometrical grating conditions a single in-plane diffracted order was just accessible in our case, with a SH intensity even exceeding the reflected one.
Thus, large PhC diffraction efficiencies appear as an interesting feature still under investigation.

In conclusion, we have presented a nonlinear method to map the dispersion band diagram of PhC structures. The attractive feature is that the nonlinear coupling between pump beam and quasiguided modes usually appears in the form of SH peaks. This technique can validate and also improve the precision of standard linear measurements. The main result of this study, however, is that a SH resonance has been measured 2700 times larger than the corresponding signal from the unpatterned GaN layer. By comparison between this result and that obtained in Ref. 12 from a one-dimensional GaN-based PhC, where the same light confinement mechanism gives an enhancement factor of 350, the experimental path toward amplified SH field up to 6 orders of magnitude seems to be open.

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