

Nonlinear diffraction in orientation-patterned semiconductors

Pawel Karpinski,^{1,2,*} Xin Chen,¹ Vladlen Shvedov,¹ Cyril Hnatovsky,¹
Arnaud Grisard,³ Eric Lallier,³ Barry Luther-Davies,¹ Wieslaw
Krolkowski,^{1,4} and Yan Sheng¹

¹Laser Physics Centre, Research School of Physics and Engineering, The Australian National University, Canberra, ACT 0200, Australia

²Wroclaw University of Technology, Wybrzeze Wyspianskiego, Wroclaw, Poland

³Thales Research and Technology, 1 Avenue Augustin Fresnel, 91767 Palaiseau Cedex, France

⁴Texas A & M University at Qatar, Doha, Qatar

*pawel.karpinski@anu.edu.au

Abstract: This work represents experimental demonstration of nonlinear diffraction in an orientation-patterned semiconducting material. By employing a new transverse geometry of interaction, three types of second-order nonlinear diffraction have been identified according to different configurations of quasi-phase matching conditions. Specifically, nonlinear Čerenkov diffraction is defined by the longitudinal quasi-phase matching condition, nonlinear Raman-Nath diffraction satisfies only the transverse quasi-phase matching condition, and nonlinear Bragg diffraction fulfils the full vectorial quasi-phase matching conditions. The study extends the concept of transverse nonlinear parametric interaction toward infrared frequency conversion in semiconductors. It also offers an effective nondestructive method to visualise and diagnose variations of second-order nonlinear coefficients inside semiconductors.

© 2015 Optical Society of America

OCIS codes: (250.4390) Nonlinear optics, integrated optics; (160.6000) Semiconductor materials; (190.5970) Semiconductor nonlinear optics including MQW; (190.2620) Harmonic generation and mixing; (190.1900) Diagnostic applications of nonlinear optics.

References and links

1. M. M. Choy and R. L. Byer, "Accurate second-order susceptibility measurements of visible and infrared nonlinear crystals," *Phys. Rev. B* **14**, 1693–1706 (1976).
2. P. S. Kuo, J. Bravo-Abad, and G. S. Solomon, "Second-harmonic generation using $\bar{4}$ -quasi-phases-matching in a GaAs whispering-gallery-mode microcavity," *Nat. Commun.* **5**, 3109 (2014).
3. A. Grisard, E. Lallier, and B. Gérard, "Quasi-phase-matched gallium arsenide for versatile mid-infrared frequency conversion," *Opt. Mater. Express* **2**, 1020–1025 (2012).
4. K. Suto, T. Sasaki, T. Tanabe, K. Saito, J. Nishizawa, and M. Ito, "GaP THz wave generator and THz spectrometer using Cr:Forsterite lasers," *Rev. Sci. Instrum.* **76**, 123109 (2005).
5. K. De Greve, L. Yu, P. L. McMahon, J. S. Pelc, C. M. Natarajan, N. Y. Kim, E. Abe, S. Maier, C. Schneider, M. Kamp, S. Hoeffling, R. H. Hadfield, A. Forchel, M. M. Fejer, and Y. Yamamoto, "Quantum-dot spin-photon entanglement via frequency downconversion to telecom wavelength," *Nature* **491**, 421–426 (2012).
6. O. Wada, "Femtosecond all-optical devices for ultrafast communication and signal processing," *New J. Phys.* **6**, 183 (2004).
7. M. Nikodem, D. Weidmann, and G. Wysocki, "Chirped laser dispersion spectroscopy with harmonic detection of molecular spectra," *App. Phys. B* **109**, 477–483 (2012).
8. A. Helmy, P. Abolghasem, J. Stewart Aitchison, B. Bijlani, J. Han, B. Holmes, D. Hutchings, U. Younis, and S. Wagner, "Recent advances in phase matching of second-order nonlinearities in monolithic semiconductor waveguides," *Laser Photon. Rev.* **5**, 272–286 (2011).

9. R. Lodenkamper, M. L. Bortz, M. M. Fejer, K. Bacher, and J. S. Harris, "Surface-emitting second-harmonic generation in a semiconductor vertical resonator," *Opt. Lett.* **18**, 1798–1800 (1993).
10. J. Armstrong, N. Bloembergen, J. Ducuing, and P. Pershan, "Interactions between light waves in a nonlinear dielectric," *Phys. Rev.* **127**, 1918–1939 (1962).
11. M. Fejer, G. Magel, D. H. Jundt, and R. Byer, "Quasi-phase-matched second harmonic generation: tuning and tolerances," *IEEE J. Quant. Electron.* **28**, 2631–2654 (1992).
12. T. Skauli, K. L. Vodopyanov, T. J. Pinguet, A. Schober, O. Levi, L. A. Eyres, M. M. Fejer, J. S. Harris, B. Gerard, L. Becouarn, E. Lallier, and G. Arisholm, "Measurement of the nonlinear coefficient of orientation-patterned GaAs and demonstration of highly efficient second-harmonic generation," *Opt. Lett.* **27**, 628–630 (2002).
13. L. Becouarn, E. Lallier, M. Brevignon, and J. Lehoux, "Cascaded second-harmonic and sum-frequency generation of a CO₂ laser by use of a single quasi-phase-matched GaAs crystal," *Opt. Lett.* **23**, 1508–1510 (1998).
14. G. Bloom, A. Grisard, E. Lallier, C. Larat, M. Carras, and X. Marcadet, "Optical parametric amplification of a distributed-feedback quantum-cascade laser in orientation-patterned GaAs," *Opt. Lett.* **35**, 505–507 (2010).
15. P. S. Kuo, K. L. Vodopyanov, M. M. Fejer, D. M. Simanovskii, X. Yu, J. S. Harris, D. Bliss, and D. Weyburne, "Optical parametric generation of a mid-infrared continuum in orientation-patterned GaAs," *Opt. Lett.* **31**, 71–73 (2006).
16. J. Schaer, K. Vodopyanov, P. Kuo, M. Fejer, X. Yu, A. Lin, J. S. Harris, D. Bliss, C. Lynch, V. Kozlov, and W. Hurlbut, "Terahertz sources based on intracavity parametric down-conversion in quasi-phase-matched gallium arsenide," *IEEE J. Select. Topics Quantum Electron.* **14**, 354–362 (2008).
17. I. Freund, "Nonlinear diffraction," *Phys. Rev. Lett.* **21**, 1404–1406 (1968).
18. V. Berger, "Nonlinear photonic crystals," *Phys. Rev. Lett.* **81**, 4136–4139 (1998).
19. S. M. Saltiel, D. N. Neshev, R. Fischer, W. Krolikowski, A. Arie, and Y. S. Kivshar, "Generation of second-harmonic conical waves via nonlinear Bragg diffraction," *Phys. Rev. Lett.* **100**, 103902 (2008).
20. S. M. Saltiel, D. N. Neshev, W. Krolikowski, A. Arie, O. Bang, and Y. S. Kivshar, "Multiorder nonlinear diffraction in frequency doubling processes," *Opt. Lett.* **34**, 848–850 (2009).
21. A. Shapira and A. Arie, "Phase-matched nonlinear diffraction," *Opt. Lett.* **36**, 1933–1935 (2011).
22. W. Wang, Y. Sheng, Y. Kong, A. Arie, and W. Krolikowski, "Multiple Čerenkov second-harmonic waves in a two-dimensional nonlinear photonic structure," *Opt. Lett.* **35**, 3790–3792 (2010).
23. N. An, H. Ren, Y. Zheng, X. Deng, and X. Chen, "Čerenkov high-order harmonic generation by multistep cascading in $\chi^{(2)}$ nonlinear photonic crystal," *Appl. Phys. Lett.* **100**, 221103 (2012).
24. T. Ellenbogen, N. Voloch-Bloch, A. Ganany-Padowicz, and A. Arie, "Nonlinear generation and manipulation of Airy beams," *Nature Photon.* **9**, 395–398 (2009).
25. E. Megidish, A. Halevy, H. S. Eisenberg, A. Ganany-Padowicz, N. Habshoosh, and A. Arie, "Compact 2D nonlinear photonic crystal source of beamlike path entangled photons," *Opt. Express* **21**, 6689–6696 (2013).
26. Y. Sheng, A. Best, H.-J. Butt, W. Krolikowski, A. Arie, and K. Koynov, "Three-dimensional ferroelectric domain visualization by Čerenkov-type second harmonic generation," *Opt. Express* **18**, 16539–16545 (2010).
27. A. Fragemann, V. Pasiskevicius, and F. Laurell, "Second-order nonlinearities in the domain walls of periodically poled KTiOPO₄," *Appl. Phys. Lett.* **85**, 375–377 (2004).
28. X. Deng and X. Chen, "Domain wall characterization in ferroelectrics by using localized nonlinearities," *Opt. Express* **18**, 15597–15602 (2010).
29. D. Faye, A. Grisard, E. Lallier, B. Gérard, M. Avella, and J. Jimenez, "Distribution of point defects in orientation-patterned GaAs crystals: A cathodoluminescence study," *Appl. Phys. Lett.* **93**, 151115 (2008).
30. D. Faye, E. Lallier, A. Grisard, and B. Gérard, "Thick low-loss Orientation-Patterned Gallium Arsenide (OP-GaAs) samples for Mid-Infrared Laser Sources," *Proc. SPIE* **6740** (2007).
31. L. A. Eyres, P. J. Tourreau, T. J. Pinguet, C. B. Ebert, J. S. Harris, M. M. Fejer, L. Becouarn, B. Gerard, and E. Lallier, "All-epitaxial fabrication of thick, orientation-patterned GaAs films for nonlinear optical frequency conversion," *Appl. Phys. Lett.* **79**, 904–906 (2001).
32. E. Gil-Lafon, J. Napierala, D. Castelluci, A. Pimpinelli, R. Cadoret, and B. Gérard, "Selective growth of GaAs by HVPE: keys for accurate control of the growth morphologies," *J. Cryst. Growth* **222**, 482–496 (2001).
33. Y. Yu, X. Gai, P. Ma, D.-Y. Choi, Z. Yang, R. Wang, S. Debbarma, S. Madden, and B. Luther-Davies, "A broadband, quasi-continuous, mid-infrared supercontinuum generated in a chalcogenide glass waveguide," *Laser Photon. Rev.* **8**, 792–798 (2014).
34. Y. Sheng, Q. Kong, W. Wang, K. Kalinowski, and W. Krolikowski, "Theoretical investigations of nonlinear Raman-Nath diffraction in the frequency doubling process," *J. Phys. B: At. Mol. Opt. Phys.* **45**, 055401 (2012).
35. Y. Sheng, Q. Kong, V. Roppo, K. Kalinowski, Q. Wang, C. Cojocar, and W. Krolikowski, "Theoretical study of Čerenkov-type second-harmonic generation in periodically poled ferroelectric crystals," *J. Opt. Soc. Am. B* **29**, 312–318 (2012).
36. M. Bass, C. DeCusatis, J. Enoch, V. Lakshminarayanan, G. Li, C. MacDonald, V. Mahajan, and E. Van Stryland, *Optical Properties of Materials, Nonlinear Optics, Quantum Optics*, vol. 4 of *Handbook of Optics* (McGraw-Hill Education, 2009), 3rd ed.
37. Y. Sheng, V. Roppo, K. Kalinowski, and W. Krolikowski, "Role of a localized modulation of $\chi^{(2)}$ in Čerenkov second-harmonic generation in nonlinear bulk medium," *Opt. Lett.* **37**, 3864–3866 (2012).

1. Introduction

Over the past decade, significant progress has been achieved in functionalizing III-V semiconductors to emit, manage, and detect light for laser and photonic applications. Some III-V compounds, notably gallium arsenide (GaAs), gallium phosphide (GaP) and indium phosphide (InP), exhibit outstanding quadratic nonlinear optical properties due to their noncentrosymmetric zinc-blende (cubic) structure. The second-order nonlinear coefficients of GaAs $d_{14} = d_{25} = d_{36} = 150$ pm/V [1] are much greater than those of widely used nonlinear ferroelectric crystals, such as lithium niobate ($d_{33} = 27$ pm/V) and potassium titanium oxide phosphate ($d_{31} = 16.9$ pm/V). The ability of III-V semiconductors to efficiently mediate energy exchange between the interacting light waves makes them very attractive for parametric frequency conversion [2, 3]; terahertz generation [4]; quantum optical applications [5]; and ultrafast all-optical signal processing [6]. GaAs-based devices also benefit from the broad infrared transparency window of GaAs, which spans from 0.9 to 17 μm [1], and are, therefore, important for military and sensing applications [7]. Moreover, combining the second-order nonlinear effects with the existing mature GaAs-based platform significantly facilitates the integration of laser sources with modulators, amplifiers and photonic bandgap structures inside all-on-chip optoelectronic devices.

GaAs has a high quadratic nonlinearity, but lacks optical birefringence. As a result, phase matching, i.e. synchronisation of phases of the interacting waves to efficiently exchange energy, cannot be achieved by traditional birefringence-based techniques. Many different approaches have been proposed to overcome this limitation [8,9]. The most effective one seems to be quasi-phase matching (QPM) [10, 11] realised by periodic modulation of the second-order nonlinearity in orientation-patterned GaAs (OP-GaAs) crystals [3]. Spatial engineering of the orientation of an OP-GaAs crystal during its growth makes neighbouring regions have reversed crystallographic orientation and consequently the opposite sign of the second-order nonlinear coefficient $\chi^{(2)}$. It is noteworthy that different crystal orientations do not affect linear optical properties of the material.

So far, QPM in OP-GaAs has been employed to demonstrate a number of second-order nonlinear optical effects, including second harmonic generation (SHG) [2, 12]; cascaded third harmonic generation [13]; optical parametric amplification [14]; mid-infrared continuum generation [15]; and terahertz generation based on intracavity parametric down-conversion [16].

All these applications, however, rely on *collinear* light interaction, during which the participating waves propagate in the direction along which the $\chi^{(2)}$ nonlinearity is modulated, as shown in Fig. 1(a). The periodic $\chi^{(2)}$ modulation is required to ensure QPM of the interacting waves via the introduction of a set of reciprocal lattice vectors, $\vec{G}_m$. In the simplest case of collinear SHG, the QPM condition can be written as a scalar equation $k_2 - 2k_1 - G_m = 0$, where $k_1 = |\vec{k}_1|$ and $k_2 = |\vec{k}_2|$ are the absolute values of the wave vectors of the fundamental and second harmonic (SH) waves, respectively. The absolute value of a reciprocal lattice vector $G_m = |\vec{G}_m|$ is given by $G_m = 2m\pi/\Lambda$, where Λ is the period of the $\chi^{(2)}$ modulation and m is an integer.

Here, we demonstrate that second-order nonlinear interaction in orientation-patterned semiconductors can also be realised in a novel *noncollinear* (*transverse*) geometry. This phenomenon is analogous to the well-known linear diffraction of waves on a dielectric grating and is hence known as nonlinear diffraction (NLD) [17–21]. An example of NLD is a noncollinear SHG. In this scheme, the incident fundamental wave propagates in the direction normal to that along which the $\chi^{(2)}$ nonlinearity is modulated, and a diffraction-like pattern of SH light is generated. As shown in Figs. 1(b)–1(d), three cases of noncollinear SHG can be distinguished. The first is Bragg SHG (BSHG) [17], which satisfies the full vectorial QPM condition

$\vec{k}_2 - 2\vec{k}_1 - \vec{G}_m = 0$ [see Fig. 1(b)]. The other two cases can be described by a partial QPM condition $\vec{k}_2 - 2\vec{k}_1 - \vec{G}_m - \Delta\vec{k}_{L(T)} = 0$ [19–21], where $\Delta\vec{k}_{L(T)}$ are the phase mismatches in the longitudinal and transverse directions, respectively. When $\Delta\vec{k}_T = 0$, the transverse QPM condition $k_2 \sin \theta + G_m = 0$ (θ is the nonlinear diffraction angle) is satisfied and Raman-Nath SHG (R-NSHG) can be observed even with a longitudinal phase mismatch $\Delta\vec{k}_L$ [see Fig. 1(c)]. In a similar way, Čerenkov SHG (CSHG) becomes observable when the longitudinal QPM condition is fulfilled, i.e. $k_2 \cos \theta - 2k_1 = 0$ [see Fig. 1(d)].

Such noncollinear interactions are of great interest because of their potential applications in all-optical signal processing. Firstly, they enable the generated wave to be emitted in a desired spatial direction, thus eliminating the necessity of filtering out the incident wave. Moreover, by combining different periodic structures it is possible to take advantage of different phase matching conditions responsible for different processes and realise, for instance, multiple frequency generation [22], higher order effects, such as third or fourth harmonic generation, as a result of cascading two lower order processes [23]. Furthermore, such transverse emission allows for the control of the transverse spatial distribution of the generated waves. For example, second harmonic generation in a form of Bessel or Airy beams have been demonstrated experimentally by illuminating a nonlinear crystal with a fundamental Gaussian beam [19, 24]. Transverse frequency conversion has been also shown to be a convenient method to create entangled photons [25].

Moreover, as we demonstrate below the CSHG also occurs at a single boundary between oppositely oriented regions of OP-GaAs, which is similar to what occurs on the boundaries of ferroelectric domains [26–28]. This effect enables three-dimensional (3D) visualisation of domain structure of the orientation-patterned semiconductors. Unlike traditional diagnostic techniques based on scanning electron microscopy or optical microscopy, which require cutting, polishing and etching of the samples, the Čerenkov-based technique is unique as it is nondestructive and enables one to characterise the QPM structure everywhere in the bulk of the sample.

2. Experiment and results

2.1. The orientation-patterned GaAs sample

We used an OP-GaAs crystal grown by Hydride Vapour Phase Epitaxy (HVPE) [29, 30], the method of choice for achieving fast growth rates with excellent selectivity so as to produce thick OP-GaAs structures for QPM frequency conversions. The epitaxial growth on orientation-patterned semiconductor crystals requires templates with modulated crystalline orientation, which constitute the seeds for the epitaxial regrowth. In a compound III-V semiconductor with the zinc-blende structure, such as GaAs, the reverse orientation is produced by exchanging atoms between the two sublattices, i.e. Ga and As, which is equivalent to a reversal of the III-V bond stacking. The first stage of the sample fabrication is the template preparation. During this process two GaAs wafers with the [001] and [00-1] crystallographic orientations are firstly bonded and then the [00-1] side is lapped until only a thin layer of the [00-1] GaAs remains on the [001] wafer. After that the patterned template is etched to reveal the orientation-patterned gratings, whose period and duty cycle are defined by photolithography [30].

The second stage of the fabrication consists of the regrowth on the OP-GaAs template to obtain a thick OP-GaAs crystal required for bulk optical pumping. In order to optimise QPM the crystallographic orientations must be preserved all along the growth process and the duty cycle of the structure must be kept equal to that defined on the template. Atmospheric pressure HVPE [31] allows one to grow hundreds of micrometres of a high quality GaAs layer. The HVPE growth is mainly limited by the adsorption of the gaseous molecules onto the surface, decomposition of the adspecies and desorption processes. HVPE growth is highly orientation-

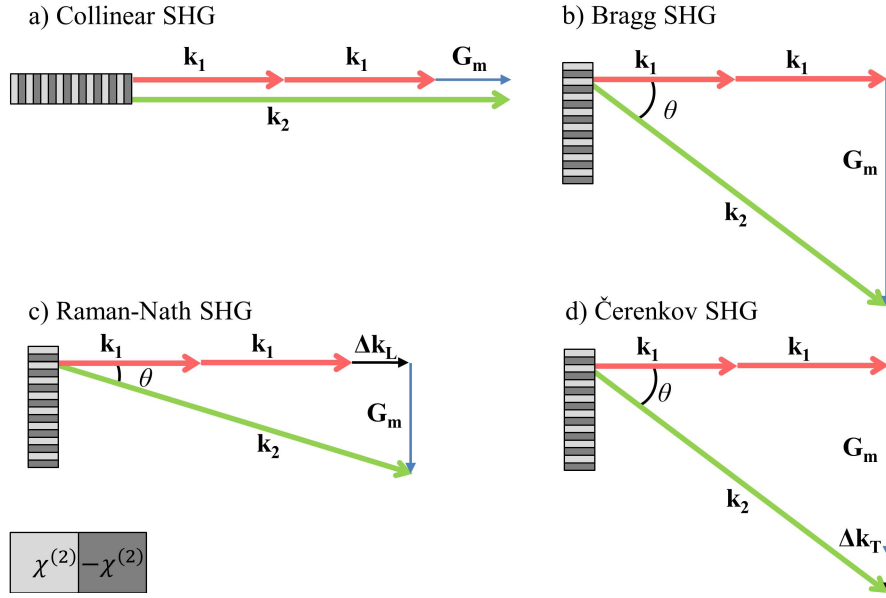


Fig. 1. Schematics of different types of SHG in OP-GaAs. (a) Traditional collinear SHG, (b) Bragg SHG satisfies the full vectorial quasi-phase matching condition, (c) Raman-Nath SHG satisfies only the transverse quasi-phase matching condition, and (d) Čerenkov SHG is defined by the longitudinal quasi-phase matching condition. $\vec{k}_1$ and $\vec{k}_2$ are the wave vectors of the fundamental and second harmonic waves, respectively, $\vec{G}_m$ is a reciprocal lattice vector and $\Delta k_{L(T)}$ are the longitudinal and transverse phase mismatches, respectively.

selective as it depends on the intrinsic growth anisotropy of the crystal, which can be controlled by the growth temperature and the precursor gas composition [32].

An optical microscopy image of a chemically etched surface of the OP-GaAs crystal used in our studies is shown in Fig. 2. A periodic modulation of the crystal orientation with a period $\Lambda = 64 \mu\text{m}$ and a 50% duty cycle is clearly visible. The thicknesses of the template and OP-GaAs layer were $300 \mu\text{m}$ and $500 \mu\text{m}$, respectively. Before the nonlinear diffraction experiment, both top and bottom surfaces of the OP-GaAs crystal have been polished in order to avoid any effects from linear diffractions.

2.2. Nonlinear diffraction from multiple domains in OP-GaAs

The experimental setup is shown in Fig. 3(a). As a light source we used an infrared (IR) femtosecond optical parametric amplifier (OPA) delivering 320 fs, 10 nJ pulses at a 21 MHz repetition rate. The central wavelength of the pulses was tunable from 2.9 to $4.0 \mu\text{m}$ [33]. The OP-GaAs sample was illuminated along the domain walls separating regions with positive and negative $\chi^{(2)}$ with loosely focused pulses from the OPA. The focal spot inside the sample was approximately $300 \mu\text{m}$ in diameter and was large enough to cover 10 domain walls. The emitted SH signal was projected onto a ground silicon wafer which acted as a scattering screen and visualised with an InGaAs camera.

During the NLD experiment we tuned the fundamental wavelength λ from 3.1 to $3.3 \mu\text{m}$ and recorded a NLD pattern at each wavelength. On the screen in Fig. 3(a) we show a NLD pattern produced at a fundamental wavelength of $3.3 \mu\text{m}$, which is representative of the whole frequency range investigated. The pattern consisted of (i) central diffraction spots, grouped

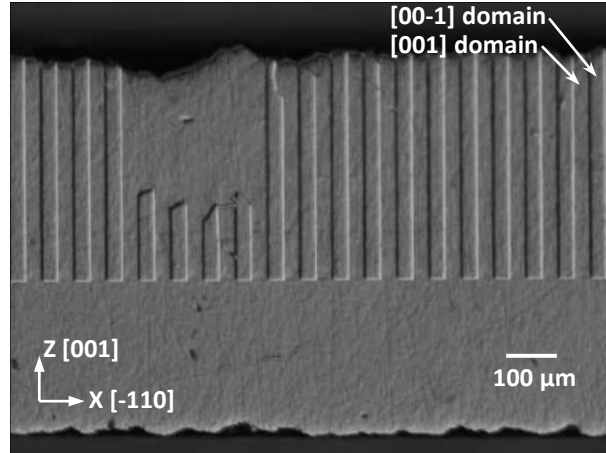


Fig. 2. Cross-sections of a 500 μm thick GaAs film grown on a OP-GaAs template of period $\Lambda = 64 \mu\text{m}$.

around the fundamental beam position, and (ii) peripheral diffraction spots, located relatively far away from the central spots on both sides of the diffraction pattern (left and right spots on the screen in Fig. 3(a)). The central diffraction spots included a collinear SH spot, which was observable only when the fundamental wave was focused at the OP-GaAs surface, and a pair of NLD spots, which were symmetric with respect to the collinear spot. The measured external diffraction angle of the two NLD spots was $\pm 1.5^\circ$. Figure 3(b) shows the normalised intensity of the peripheral NLD spots as a function of the fundamental wavelength λ and diffraction angle θ .

To explain the NLD pattern, we consider the intensity of SHG from a periodic $\chi^{(2)}$ structure, which can be written as [34, 35]:

$$I_{2\omega} \sim I_\omega^2 \times S_L^2 \times S_T^2, \quad (1)$$

where I_ω is the intensity of the fundamental beam inside the material and $S_{L(T)}$ are functions describing the longitudinal and transverse QPM conditions, respectively. The longitudinal QPM condition is satisfied when the function $S_L = \text{sinc} \left[z \left((k_2 \cos \theta)^2 - (2k_1)^2 \right) / 2 \right]$ approaches its maximum. In the above expressions, $\text{sinc}(x) = \sin(x)/x$ and z is the interaction length. The SHG angle inside the sample satisfying this QPM condition is the Čerenkov angle θ_C :

$$\theta_C = \cos^{-1} \frac{n_1}{n_2}, \quad (2)$$

where n_1 and n_2 are the refractive indices of the medium at the fundamental and SH wavelengths, respectively.

As in the previous case, the transverse QPM condition is defined by the maximum of the function $S_T = \sum_{m=0, \pm 1, \pm 2, \dots} g_m e^{-w^2(k_2 \sin \theta + G_m)^2 / 8}$, where w is the width of the fundamental beam and g_m are Fourier coefficients, which depend on the duty cycle of the $\chi^{(2)}$ nonlinear grating [34, 35]. The transverse QPM condition defines Raman-Nath angles $\theta_{RN, m}$ inside the sample:

$$\theta_{RN, m} = \sin^{-1} \frac{m\lambda}{2n_2\Lambda}. \quad (3)$$

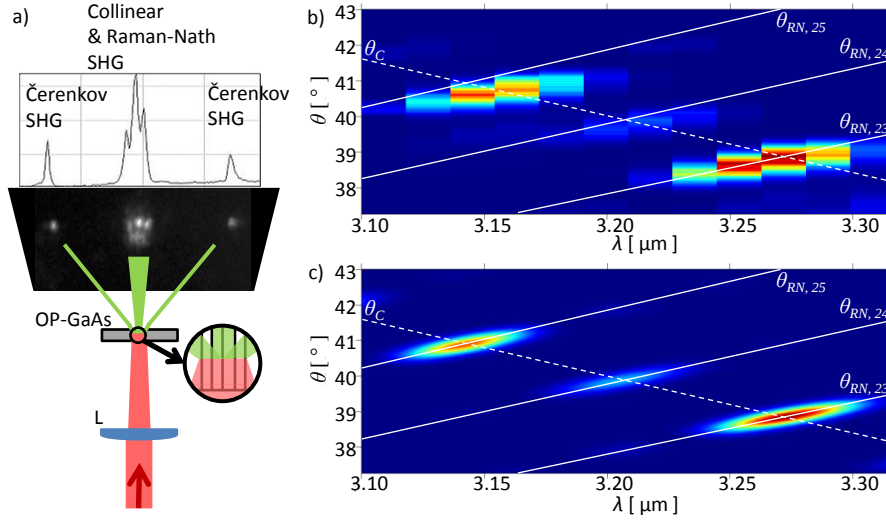


Fig. 3. (a) The experimental setup for observation of NLD. The incident laser beam is loosely focused with a lens (L , $f = 150$ mm) onto the OP-GaAs sample along the domain walls, which separate positive and negative $\chi^{(2)}$ domains. The closeup in the circle shows the focal spot, which covers multiple periods of the OP-GaAs. On the screen is shown the recorded nonlinear diffraction pattern at a fundamental wavelength of $3.3 \mu\text{m}$ together with the corresponding intensity profile. The experimentally measured (b) and modelled (c) intensity of the emitted SH signal as a function of the fundamental wavelength λ and diffraction angle θ outside the sample. In both cases the focal spot covers 10 domain walls. The solid lines represent Raman-Nath angles $\theta_{RN, m}$ for $m = 23, 24$ and 25 and the dashed lines represent the Čerenkov angle θ_C as a function of the fundamental wavelength λ .

Using Eq. (3) we calculated $\theta_{RN, 1} = 1.5^\circ$ for the first order of nonlinear R-NSHG outside the OP-GaAs with a period $\Lambda = 64 \mu\text{m}$, in agreement with the measured diffraction angle of the pair of SH spots in the central group. The refractive index of GaAs was calculated using the Sellmeier formula [36]. The intensities of the peripheral SH spots were calculated using Eq. (1) and in Fig. 3(c) the intensity of the peripheral diffraction spot $I_{2\omega}(\lambda, \theta)$ is presented as a function of the fundamental wavelength and diffraction angle outside the sample. In Figs. 3(b) and 3(c), the theoretical plots of the wavelength dependance of the Čerenkov angle θ_C (dashed lines) and Raman-Nath angles $\theta_{RN, m}$ (solid lines) outside the sample are produced using Eqs. (2) and (3), respectively.

It can be seen that the experimental results are in good agreement with the calculations. Importantly, the SH intensity gets much stronger when the Čerenkov angle θ_C becomes equal to a certain order of Raman-Nath angle $\theta_{RN, m}$. In fact, this is exactly the case of BSHG, when both the longitudinal and transverse QPM conditions are satisfied simultaneously. It can also be seen that the odd orders ($m = 23, 25$) of nonlinear diffraction are much more pronounced than the even order ($m = 24$). This is the direct consequence of an almost 50% duty cycle of the $\chi^{(2)}$ nonlinearity modulation, in which the regions with positive and negative nonlinear response have almost equal lengths [19, 34, 35].

2.3. Čerenkov SH emission from a single domain wall in OP-GaAs

Here, we investigate SHG in OP-GaAs induced by a tightly focused fundamental beam, i.e. the beam whose width is much smaller than the period of the $\chi^{(2)}$ nonlinearity modulation. In

this situation, the fundamental beam illuminates either a homogeneous region of the sample or the wall separating oppositely oriented domains. As we have shown recently [37], in this geometry only the Čerenkov SH signal is generated, which occurs when the fundamental beam illuminates a domain wall. No R-NSHG can be observed as it requires the participation of multiple grating periods.

The experimental setup is shown in Fig. 4(a). The sample was mounted on a XYZ-translation stage and could be scanned with a 60 nm resolution. We used the same OPA source as in Section 2.2. The fundamental beam ($\lambda = 3.5 \mu\text{m}$) was chopped at 1 kHz and focused inside the sample using a molded chalcogenide lens with an effective $NA = 0.3$. The SH signal was collected with an IR-corrected objective with $NA = 0.65$. A spatial filter was used to block the surface-induced collinear SHG to ensure that only the Čerenkov SH signal was focused onto the InGaAs photodiode and processed using a lock-in amplifier.

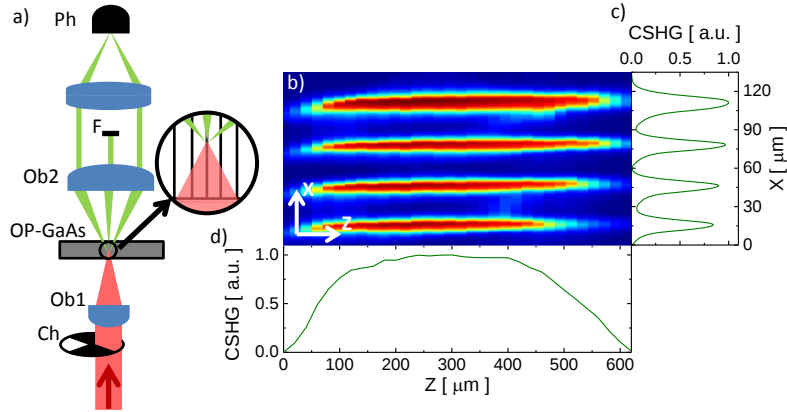


Fig. 4. (a) The experimental setup for observation of CSHG using a tightly focused fundamental beam: Ch - chopper, Ob1 - $NA = 0.3$ objective, Ob2 - $NA = 0.65$ objective, F - spatial filter, Ph - InGaAs photodiode. The inset shows the focusing configuration inside the OP-GaAs: the focal spot is much smaller than the period of the $\chi^{(2)}$ nonlinearity modulation. (b) The intensity of the emitted Čerenkov signal recorded by translating the sample in the XZ plane. (c) and (d) show the Čerenkov intensity profiles along the X and Z axes, respectively.

In this configuration we measured the CSHG intensity as a function of the focal spot position inside the sample. Scanning at a constant velocity proceeded first along the X direction. The lock-in signal from the detector was triggered by the stage controller to synchronise the data acquisition process with the motion of the stage. In this way, we could achieve a $0.5 \mu\text{m}$ resolution along the X axis. Scans along the Y and Z axes were performed at 2 and $20 \mu\text{m}$ resolution, respectively.

The resolution of the domain wall image is determined by both the resolution of the scanning system and the size of the OPA focus inside the sample. The focal radius w_0 of an incoming fundamental beam with a Gaussian intensity profile is given by $w_0 = \lambda / \pi NA$ and in our case is estimated at $3.7 \mu\text{m}$ in the lateral, i.e. XY, plane. In the longitudinal, i.e. Z, direction the size of the focus is determined by the Rayleigh length $z_R = (\pi n_1 w_0^2) / \lambda \approx 41 \mu\text{m}$. In addition, the resolution is also affected by spherical aberration caused by the high refractive index mismatch between the ambient air and the GaAs sample at both the fundamental and SH wavelengths.

Figure 4(b) shows a two-dimensional intensity pattern of the Čerenkov SH signal in the XZ plane. The colour change from blue to red reflects variation of the Čerenkov SH intensity from low to high. It can be seen that CSHG is very sensitive to the position of the focal spot with

respect to the domain walls. The maxima of the SH signal coincide with the locations of the domain walls in the OP-GaAs sample.

In Fig. 4(c), we plot a typical CSHG intensity profile along the X axis. The separation between the maxima is $32\ \mu\text{m}$, which is half of the $\chi^{(2)}$ nonlinearity modulation period Λ . This result confirms the 50% duty cycle. Figure 4(d) displays the CSHG intensity profile in the whole scanned region along the Z direction. One can see that the strength of the CSHG remains almost constant inside the sample and decreases gradually at the ends of the sample. This is because the effective interaction length decreases when the focus of the fundamental beam moves out of the sample.

2.4. Three-dimensional visualisation of orientation-patterned GaAs

The sensitivity of CSHG to the presence of domain walls can be used to provide a contrast in scanning microscopy to visualise the domain structure of OP-GaAs in 3D. It should be stressed that, the demonstrated CSHG-based scanning microscopy technique is totally non-destructive as it does not require any cutting or etching to reveal the bulk domain structure. In Fig. 5, we present a 3D image of the domain pattern inside the OP-GaAs sample. The image was created by stacking a number of XY scans recorded at different depths Z. The box in the Fig. 5 represents the whole scanning volume $200 \times 200 \times 600\ \mu\text{m}^3$. It is clear that our CSHG-based microscopy technique allows us to not only reveal the nonlinear grating parameters such as the period and duty cycle, but also to determine the quality of orientation patterning in the bulk of a sample. For example, in Fig. 5 we show that the domain structure is nonuniform in the scanned region. In particular, the effect of merging of two neighbouring domains, which were initially separated, is evident.

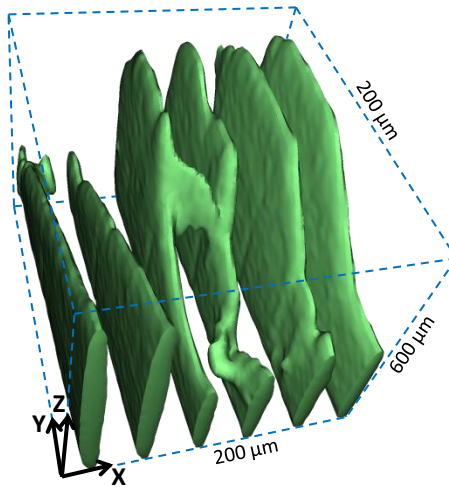


Fig. 5. A three-dimensional image of the OP-GaAs structure, obtained by scanning the fundamental beam inside the sample and recording the strength of the corresponding nonlinear Čerenkov diffraction at each position. The size of the box is $200 \times 200 \times 600\ \mu\text{m}^3$.

3. Conclusions

We have presented the nonlinear diffraction in a OP-GaAs crystal. In particular, we have shown nonlinear Raman-Nath and Čerenkov second harmonic emission from a periodically modulated

$\chi^{(2)}$ nonlinearity. We explain this effect by employing the concepts of longitudinal and transverse quasi-phase matching and show that the intensity of nonlinear diffraction is maximum, i.e. Bragg second harmonic generation, when these two conditions are satisfied simultaneously.

We have also shown that Čerenkov second harmonic emission in the sample can be used to visualise the 3D structure of the $\chi^{(2)}$ nonlinearity modulation. Unlike traditional characterisation methods based on scanning electron microscopy or optical microscopy, which require cutting, polishing and etching of the samples, our technique is nondestructive and enables one to visualise the QPM structure everywhere in the bulk of the sample. This constitutes a unique nondestructive diagnostic tool for characterisation of orientation-patterned semiconductors and, potentially, any variations of the second-order nonlinearities in semiconducting materials.

Acknowledgments

The authors thank the Australian Research Council for financial support. P. Karpinski thanks the Polish Ministry of Science and Higher Education for "Mobility Plus" scholarship.