Domain-shape-based modulation of Čerenkov second-harmonic generation in multidomain strontium barium niobate

Mousa Ayoub,* Philip Roedig, Jörg Imbrock, and Cornelia Denz

Institute of Applied Physics and Center for Nonlinear Science (CeNoS), Westfälische Wilhelms-Universität Münster, Corrensstrasse 2, 48149 Münster, Germany

**Corresponding author: ayoubm@uni-muenster.de*

Received September 2, 2011; revised October 3, 2011; accepted October 5, 2011; posted October 10, 2011 (Doc. ID 153749); published November 11, 2011

We study experimentally and numerically the second-harmonic Čerenkov emission with two different characteristic azimuthal intensity distributions in strontium barium niobate with a random structure of χ^2 nonlinearity. We monitor *in situ* the Čerenkov emission during domain switching and show that a change of domain size and shape results in a fourfold azimuthal modulation of the Čerenkov cone.

OCIS codes: 190.0190, 190.4410, 190.4975, 160.2260.

Čerenkov-type second-harmonic generation in ferroelectric crystals is one of the most interesting parametric processes in nonlinear optics [1–3]. This type of second-harmonic generation (SHG) was demonstrated in different bulk nonlinear media [1,2,4], waveguides, [3] as well as in crystals with a periodic, quasi-periodic, and random modulation of the $\chi^{(2)}$ nonlinearity [4–7]. In 2D nonlinear photonic structures where the $\chi^{(2)}$ nonlinearity is two-dimensionally modulated, the Čerenkov SHG is enhanced in comparison with bulk nonlinear crystals and emitted on a cone [6]. The cone angle is defined only by the longitudinal phase-matching condition $k^{(2\omega)}\cos(\theta) - 2k^{(\omega)} = 0$, where θ is the Čerenkov angle inside the crystal and $k^{(\omega)}$, $k^{(2\omega)}$ are the wave vectors of the fundamental and the second-harmonic beams, respectively [cf. Fig. 1(a)]. The angle θ does not depend on the $\chi^{(2)}$ pattern, whereas the azimuthally modulated SH intensity on the ring depends on the domain shape. For instance, in 2D $LiNbO_3$ and $LiTaO_3$ nonlinear photonic structures the Čerenkov ring is typically hexagonally modulated because of the hexagonal and triangular domain shape, respectively [7–9]. The polarization properties of the nonlinear $\chi^{(2)}$ tensor can lead to an additional modulation. As-grown or unpoled strontium barium niobate (SBN) crystals show a 2D disordered domain structure and the ferroelectric domains have the form of rods with a shape of fourfold symmetry, e.g., a square with rounded corners [10]. The reported domain width in SBN crystals ranges between a few nanometers and a few micrometers depending on the poling state [10–12]. Recently, the influence of the domain shape on the SH intensity distribution was theoretically analyzed for the case of SBN and it is expected that a domain shape with fourfold symmetry should lead to a fourfold azimuthally modulated Čerenkov ring [13]. However, up to now all reported Čerenkov SHG patterns in random SBN possess a homogeneous azimuthal intensity distribution [1,2,14,15].

In this Letter, we investigate both experimentally and numerically the effect of the domain shape in an SBN crystal on the modulation of the azimuthal intensity distribution of the Čerenkov ring. We monitor *in situ* the Čerenkov emission during domain switching and show that a change of domain size and shape results in a fourfold azimuthal modulation of the Čerenkov cone.

For Čerenkov SHG we use ultrashort laser pulses with a wavelength of $\lambda_{\text{input}} = 1200\,\text{nm}$, a pulse duration of about $\tau_p = 80\,\text{fs}$, pulse energies up to $100\,\mu\text{J}$, and a repetition rate of 1 kHz. Collimated Gaussian laser pulses with a beam diameter of about 1.5 mm are propagating along the c axis of the crystal. We use an $Sr_{0.61}Ba_{0.39}Nb_2O_6$ (SBN) crystal with the dimensions $6.6 \times 6.6 \times 1.6\,\text{mm}^3$. The large surfaces perpendicular to the c axis are polished to optical quality. In preparation, the crystal was first heated up above the Curie temperature $T_c \approx$ 70–80 °C and then cooled down without applying an electric field to produce an unpoled sample. In order to influence the domain size and form, the sample was subsequently poled and repoled stepwise at room temperature [12,16]. For this, an electric field was applied along the polar axis and increased in steps of 10 V/cm every 2 s until finally 7.5 kV/cm are reached. For visualizing the

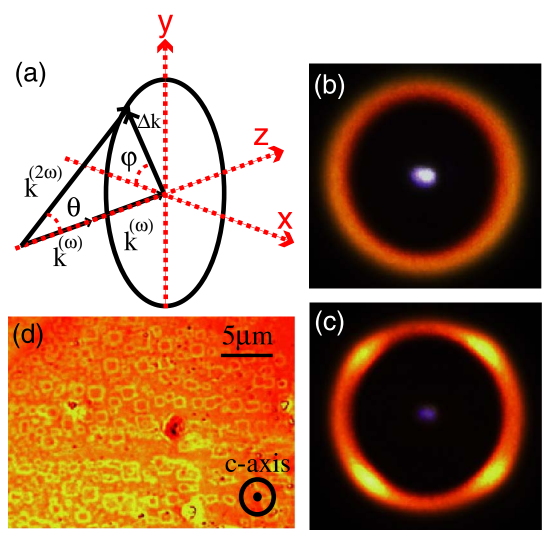

Fig. 1. (Color online) (a) Phase-matching condition for Čerenkov-type SHG. (b), (c) Images of two different SH patterns of $\lambda_{\text{input}} = 1200\,\text{nm}$ corresponding to the unpoled and repoled states, respectively. (d) Phase contrast picture of a repoled polydomain SBN crystal.

0146-9592/11/224371-03$15.00/0

evolution of Čerenkov emission during the domain switching process, we used transparent electrodes of indium tin oxide and projected the emitted SH signal onto a screen and recorded it using a CCD color camera.

Figures 1(b) and 1(c) show different Čerenkov emission at two different poling states. In the case of an unpoled sample, where the domain size is estimated between 200–400 nm [2,12], the emitted SH signal is weak and homogeneous [Fig. 1(b)]. As the size of the domains in an unpoled sample is too small to resolve them by phase contrast microscopy we cannot determine the real size and shape of the domains here. After the initially unpoled crystal was poled the cone is still observable but much weaker than in the unpoled case and the azimuthal distribution of the SH intensity is still homogeneous. During the following domain switching (repoling) the modulation on the cone begins to arise after exceeding the coercive field of about 3 kV/cm. The corresponding domain pattern after the second switching process is depicted in Fig. 1(d). The phase contrast image shows domains that have the shape of a square with rounded corners and a typical diameter of about 1.7 μm.

We have monitored *in situ* the Čerenkov emission during the domain switching process (see Media 1) and in Fig. 2 six experimental photographs, representing the second switching process, are depicted. The field is applied from zero field [Fig. 2(a)] to 7.5 kV/cm [Fig. 2(f)]. We want to point out that the total Čerenkov intensity has always its highest value nearly at the coercive field when the absolute value of spontaneous polarization is zero, i.e., when the number of domains pointing in the $+c$ direction is the same as the number of domains pointing in the $-c$ direction. No changes of the cone intensity or of the strength of the modulation were observed when further increasing the applied field. Further switching of the domains also does not add any significant changes to the form of the modulated cone. The pattern also rotates when rotating the crystal (see Media 2) supporting the fact that origin of the azimuthal intensity modulation is the domain shape.

In order to understand the physical origin of this phenomenon, we routinely use the Green's function approach [13,17]. In general, the second-harmonic E field from an arbitrarily shaped single nonlinear volume V_{NL} can be written as

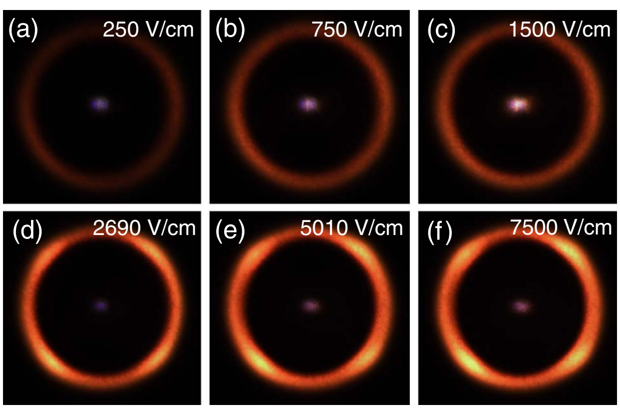

Fig. 2. (Color online) Čerenkov emission recorded during the switching process of the SBN sample (Media 1, Media 2), (a) at 0.25 kV/cm to (f) at 7.5 kV/cm.

$$\mathbf{E}^{(2\omega)}(\mathbf{r}) \propto \int_{V_{\mathrm{NL}}} d\mathbf{r}'\hat{\mathbf{G}}(\mathbf{r},\mathbf{r}')\mathbf{P}^{(2\omega)}(\mathbf{r}'), \tag{1}$$

where $\hat{\mathbf{G}}(\mathbf{r},\mathbf{r}')$ is the dyadic Green's function, and $\mathbf{P}^{(2\omega)}(\mathbf{r}')$ defines the nonlinear polarization for the SH signal, i.e., $P_i^{(2\omega)} \propto \sum_{j,k}\chi_{ijk}^{(2)}E_j^{(\omega)}E_k^{(\omega)}$. As pointed out, the nonlinear volume V_{NL} can take different geometrical shapes. The far-field intensity can then be easily calculated from $I^{(2\omega)}(\mathbf{r}) \propto |\mathbf{E}^{(2\omega)}(\mathbf{r})|^2$:

$$I^{(2\omega)}(\mathbf{r}) \propto \sin(\theta)^2(d_{\mathrm{eff}}(\theta)h)^2\mathrm{sinc}^2[\Delta k_z(\theta)h/2] \times\left[\iint_A \mathrm{d}x'\mathrm{d}y' \exp(-i\Delta\mathbf{k}_{xy}(\varphi,\theta)\mathbf{r}')\right]^2, \tag{2}$$

where $\Delta k_z(\theta)$ represents the phase-matching wave vector along the z axis, $\Delta\mathbf{k}_{xy}(\varphi,\theta)$ is the transverse phase-matching vector in the x–y plane, and d_{eff} is the effective second-order nonlinear coefficient. Because of the polarization properties of the nonlinear tensor in SBN, d_{eff} is independent of the azimuthal angle φ. Therefore, the observed modulation cannot be attributed to the polarization properties of d_{eff}. For the generality, the numerical simulations were done with a nonlinear medium V_{NL}, which has a 3D parallelepiped shape with a height h along the z axis, diameter d, and a cross section A. In the calculations, the cross section is described by a so-called *squircle* with a parameter s, which determines the squareness. For instance, at $s = 0$ the nonlinear medium has a cross section in the form of a circle, corresponding to a cylinder, and at $s = 1$ the cross section is a square. In case of an unpoled sample, Fig. 3 displays the SH signal of a domain illuminated along the c axis and defined by $d = 0.3\,\mu$m and $h = 7\,\mu$m. For this small domain diameter the azimuthal SH intensity distribution is always homogeneous and does not change when the squareness parameter s is varied. The calculated opening angle amounts to $\theta = 14.7°$, which fits well to the measured value of 14°. The calculations show for such small domains that the modulation on the cone should arise for input wavelengths shorter than about $\lambda_{\mathrm{input}} = 700$ nm. This cannot be verified experimentally because below

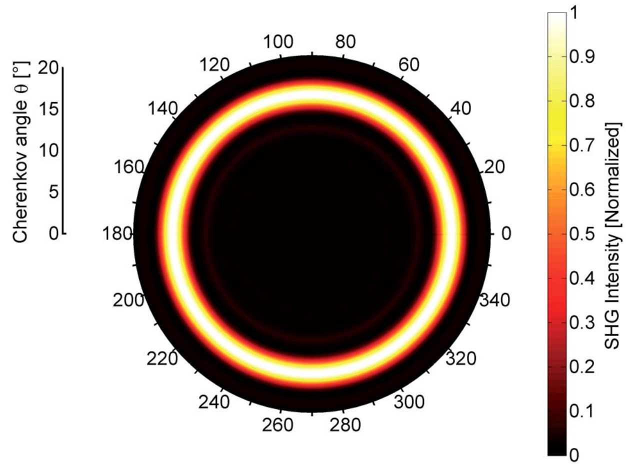

Fig. 3. (Color online) Numerical simulation of the azimuthal intensity distribution of the Čerenkov ring in the case of $d = 0.3\,\mu$m and $h = 7\,\mu$m, corresponding to the case of an unpoled sample.

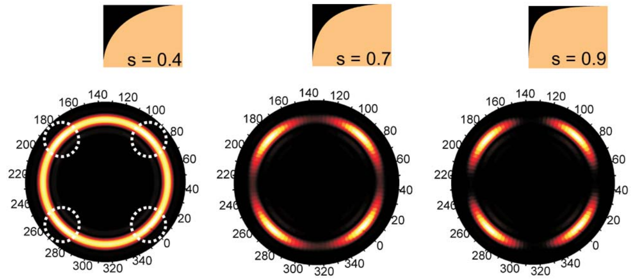

Fig. 4. (Color online) Numerical simulation of the azimuthal intensity distribution in dependence of the squareness parameter s for a domain width of $d = 1.7\,\mu$m. For $s = 0.4$ the weak modulation is marked with dashed circles.

400 nm Čerenkov SHG is totally internal reflected and also strongly absorbed. However, after switching and reswitching the polarization, as mentioned above, the modulation on the cone appears while the domains become larger. For this case, the domain size will be defined by the chosen size parameters $d = 1.7\,\mu$m and $h = 15\,\mu$m [cf. Fig. 1(d)].

The theoretical angular intensity distribution is exemplary depicted in Fig. 4, in which the effect of the squareness degree is clearly shown. Three different degrees were chosen to explain the effect. Because of the nature of this geometrical shape, the corners become rapidly sharper with increasing s. Moreover, for this size the calculation becomes very sensitive to the squareness degree and this clearly results in a modulation of the SH cone at $s = 0.7$ and 0.9, without any change in the opening angle.

The results depicted in Figs. 3 and 4 point out that the far-field intensity distribution from a single domain already reflects in a very good agreement the experimental patterns where hundreds of several randomly distributed domains are illuminated. However, the calculations were repeated for the case of a random distribution of 300 domains, whose size is normally distributed with different standard variance. As an example, for the unpoled case we performed the calculation with a mean value of $\langle d \rangle = 0.3\,\mu$m and a variance of $\sigma_d = 0.1\,\mu$m, and for the case of a repoled sample with $\langle d \rangle = 1.7\,\mu$m and $\sigma_d = 0.3\,\mu$m. The incoherent overlap of the generated SH signals due to the random distribution of the nonlinear domains does not add significant changes to the fourfold modulation on the SH ring.

In conclusion, we have studied the Čerenkov-type SHG in a multidomain SBN crystal with different poling states, which are defined by the domain sizes. We have observed that starting from a defined domain size a characteristic azimuthal modulation on the Čerenkov cone appears. Then we have numerically explained, in a very good agreement with the experimental observations, this remarkable phenomenon by analyzing the basic geometrical form of the individual ferroelectric domains in the far field, which can be influenced by the poling process. The advantage is that there is no need to pole the crystal artificially or completely to reform the shape of the individual domains.

References

1. A. R. Tunyagi, M. Ulex, and K. Betzler, Phys. Rev. Lett. **90**, 243901 (2003).
2. P. Molina, M. de la O Ramírez, and L. E. Bausá, Adv. Funct. Mater. **18**, 709 (2008).
3. Y. Zhang, X. P. Hu, G. Zhao, S. N. Zhu, and M. Xiao, J. Phys. D **42**, 215103 (2009).
4. V. Vaičaitis, Opt. Commun. **209**, 485 (2002).
5. K. A. Kuznetsov, G. K. Kitaeva, A. V. Shevlyuga, L. I. Ivleva, and T. R. Volk, JETP Lett. **87**, 98 (2008).
6. X. Deng, H. Ren, H. Lao, and X. Chen, J. Opt. Soc. Am. B **27**, 1475 (2010).
7. S. M. Saltiel, Y. Sheng, N. Voloch, D. N. Neshev, W. Krolikowski, A. Arie, K. Koynov, and Y. S. Kivshar, IEEE J. Quantum Electron. **45**, 1465 (2009).
8. A. Arie and N. Voloch, Laser Photon. Rev. **4**, 355 (2010).
9. Y. Sheng, S. M. Saltiel, W. Krolikowski, A. Arie, K. Koynov, and Y. S. Kivshar, Opt. Lett. **35**, 1317 (2010).
10. L. Tian, D. A. Scrymgeour, and V. Gopalan, J. Appl. Phys. **97**, 114111 (2005).
11. P. Lehnen, W. Kleemann, T. Woike, and R. Pankrath, Phys. Rev. B **64**, 224109 (2001).
12. M. Ayoub, J. Imbrock, and C. Denz, Opt. Express **19**, 11340 (2011).
13. J. Bravo-Abad, X. Vidal, J. L. D. Juarez, and J. Martorell, Opt. Express **18**, 14202 (2010).
14. R. Fischer, S. Saltiel, D. Neshev, W. Krolikowski, and Y. Kivshar, Appl. Phys. Lett. **89**, 191105 (2006).
15. U. Voelker, K. Dinges, U. Heine, and K. Betzler, Phys. Rev. B **81**, 214104 (2010).
16. U. Voelker, U. Heine, C. Gödecker, and K. Betzler, J. Appl. Phys. **102**, 114112 (2007).
17. J.-X. Cheng and X. S. Xie, J. Opt. Soc. Am. B **19**, 1604 (2002).