Nonlinear Čerenkov Radiation in Nonlinear Photonic Crystal Waveguides

Y. Zhang, Z. D. Gao, Z. Qi, S. N. Zhu,* and N. B. Ming

National Laboratory of Solid State Microstructures and Department of Physics, Nanjing University, Nanjing, 210093, China

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We study nonlinear Čerenkov radiation generated from a nonlinear photonic crystal waveguide where the nonlinear susceptibility tensor is modulated by the ferroelectric domain. Nonlinear polarization driven by an incident light field may emit coherently harmonic waves at new frequencies along the direction of Čerenkov angles. Multiple radiation spots with different azimuth angles are simultaneously exhibited from such a hexagonally poled waveguide. A scattering involved nonlinear Čerenkov arc is also observed for the first time. Čerenkov radiation associated with quasi-phase matching leads to these novel nonlinear phenomena.

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A charged particle traveling faster than the speed of light can drive the medium to emit coherent light called Čerenkov radiation (CR) [1,2]. In such a process, the coherent radiation is observable at a conical wave front defined by the Čerenkov angle $\theta_c = \arccos(v'/v)$, where v is the speed of the moving charged particle and v' is the phase velocity of the radiation wave. CR occurs as $v > v'$. Light propagating in a nonlinear optical crystal or waveguide can also create such an emission via a second-order nonlinear process, such as Čerekov second harmonic generation (SHG) [3–8] and THz emission from optical beating [9–11]. Although closely resembling CR via relativistic charged particles, it has unique distinguishing features. Most importantly, it is a nonlinear optical process associated with the susceptibility tensor $\chi^{(2)}$ and the radiation source is not a point particle, but a spatially extended collection of dipoles driven by the incident light field, i.e., nonlinear polarization ($P_{\rm non}$). Therefore, we call that nonlinear Čerenkov radiation (NCR). However, the phase velocity of the source does need exceed the radiation velocity. Comparing with a conversional $\chi^{(2)}$ process, NCR now can be realized only by extremely short optical pulses [9,10] or in the waveguide [3,4], which results in a relaxation of phase-matching requirements. This is relatively simple if the incident light is only monochromatic—for example, Čerenkov SHG from a planar waveguide [3]. In this case, the radiation source (second-order polarization) and incident light (guide modes of the waveguide) have the same phase velocity v. In a normally dispersive medium, it is faster than v' of SH and the harmonic radiation emits into the substrate. Because the geometry is planar and $P_{\rm non}$ is coherent over the full width of the beam, i.e., over many wavelengths, NCR emerges as a well-collimated beam with the Čerenkov angle relative to the direction of $P_{\rm non}$.

We are concerned with two questions about NCR. Up to now, NCR has been usually generated from monochromatic light. If incident lights contain two or more than two frequency components, can they generate NCR? In principle the answer is yes, but it is not a simple accumulation of several Čerenkov SHG processes. For example, ω_1 and ω_2 for the simplest case $P_{\rm non}$ will include terms $2\omega_1$, $2\omega_2$, $\omega_1 + \omega_2$, and $\omega_1 - \omega_2$. Obviously, terms $2\omega_1$ and $2\omega_2$ can lead to Čerenkov SHG at frequency $2\omega_1$ and $2\omega_2$, as discussed above. If $\omega_1 \neq \omega_2$, phase velocity v_p of $P_{\rm non}$ for $\omega_3 = \omega_1 \pm \omega_2$ is equal to neither ω_1 nor ω_2. It is decided by

$$\frac{\omega_1}{v_1}\vec{x}_1 \pm \frac{\omega_2}{v_2}\vec{x}_2 = \frac{\omega_3}{v_p}\vec{x}_p, \tag{1}$$

where v_i is phase velocity and $\vec{x}_i$ the direction of $\vec{v}_i$ ($i = 1$, 2, p). In theory, $P_{\rm non}$ can emit NCR as long as v_p is faster than v', the phase velocity of ω_3, and Čerenkov radiation angle is defined by

$$\theta_c = \arccos(v'/v_p). \tag{2}$$

This process is more complicated but more general. From this point, we can analyze NCR with more frequency components. And if $\omega_1 = \omega_2$, it becomes Čerenkov SHG.

It is easy to obtain from Eq. (1) that in a homogenous medium, NCR for frequency up-conversion, such as the generation of $2\omega_1$, $2\omega_2$, $\omega_1 + \omega_2$, always satisfies $v_p > v'$ for normal dispersion, but does not for the process of $\omega_1 - \omega_2$ because $v_p < v'$ at the case and Eq. (2) is violated. NCR is intrinsically determined by the medium's dielectric constants (or refractive index) and their dispersion; therefore, the Čerenkov angle is fixed. Can we change NCR's behavior by another physics process or microstructure? Several recent studies showed these could occur with phonon assistance [12] or using photonic crystal [13]. We provide a new approach by introducing nonlinear photonic crystal [14]. Differing from that in a homogeneous medium, the behavior of radiation source $P_{\rm non}$ in a nonlinear photonic crystal can be changed by the modulation of $\chi^{(2)}$ in terms of quasi-phase matching (QPM) [15–17]. $P_{\rm non}$ will change its phase π from positive domain to negative domain as $\chi^{(2)}$ alters its sign from +1 to −1, which leads to an effective change in v_p along the propagating direction of $P_{\rm non}$ (Fig. 1) and, therefore, changes the behavior of

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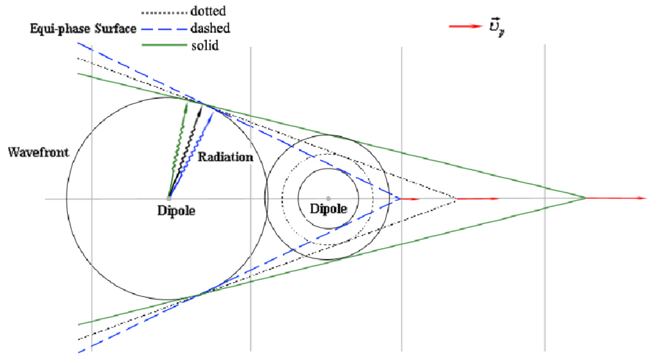

FIG. 1 (color online). A collection of dipoles ($P_{\rm non}$) driven by incident light emits coherent radiation. In nonlinear photonic crystal $P_{\rm non}$ will change its phase $+\pi$ (dashed) or $-\pi$ (solid) as $\chi^{(2)}$ alters its sign compared with that in homogenous material (dotted), which leads to an effective change in the phase velocity of $P_{\rm non}$ as well as Čerenkov angle.

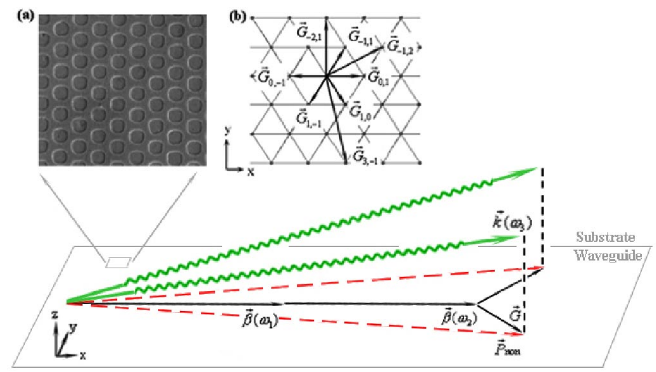

FIG. 2 (color online). Phase-matching process for NCR in nonlinear photonic crystal waveguide. Dashed lines represent polarization and curves are radiation waves. The insets are (a) domain structure and (b) reciprocal lattice of hexagonally poled $LiTaO_3$.

NCR, such as the threshold value and radiation angle, and even makes NCR appear in a frequency down-conversion process. These properties have not been studied in any detail until very recently.

In this Letter, we study NCR generated from a nonlinear photonic crystal waveguide both experimentally and theoretically. For simplicity, here the fundamental beam only contains two frequency components, ω_1 and ω_2. The nonlinear photonic crystal provides a set of collinear and noncollinear reciprocals $\vec{G}$ [14] in this NCR process. In comparison with Eq. (1), the corresponding expression for sum-frequency generation (SFG) ($\omega_1 \neq \omega_2$) and SHG ($\omega_1 = \omega_2$, $v_1 = v_2$) in such a structure is

$$\frac{\omega_1}{v_1}\vec{x}_1 + \frac{\omega_2}{v_2}\vec{x}_2 + \vec{G} = \frac{\omega_3}{v_p}\vec{x}_p. \qquad (3)$$

The equation extends NCR from the homogeneous medium to nonlinear photonic crystal, showing that Čerenkov radiation can be realized in the QPM scheme. In the experiment, both ω_1 and ω_2 are guide modes and $\vec{x}_1$ is collinear with $\vec{x}_2$. $\vec{x}_p$ and $\vec{v}_p$ will depend on the reciprocal $\vec{G}$. According to Eqs. (2) and (3), in wave vector space, QPM-NCR can be written as

$$|\vec{\beta}(\omega_1) + \vec{\beta}(\omega_2) + \vec{G}| = k(\omega_3)\cos\theta_c, \qquad (4)$$

where $\vec{\beta}(\omega_1)$ and $\vec{\beta}(\omega_2)$ are wave vectors of guided modes, $k(\omega_3) = \omega_3/v'$ is the magnitude of the radiation mode's wave vector in substrate. The schematically phase-matching geometry is shown in Fig. 2. The wave vector of $P_{\rm non}$ can make up a vector triangle together with $\vec{\beta}(\omega_1)$, $\vec{\beta}(\omega_2)$, and $\vec{G}$. When the Čerenkov condition [Eq. (2)] is satisfied, NCRs with frequency $\omega_3 = 2\omega_1, 2\omega_2, \omega_1 + \omega_2$ can be simultaneously achieved and, respectively, emit along different azimuth angles.

In the experiment, the nonlinear photonic crystal was a hexagonally poled $LiTaO_3$ by the field poling technique with a domain interval $a = 9.0$ μm and reversal factor of ~30% [Fig. 2(a)]. The structure provides reciprocals of sixfold symmetry [Fig. 2(b)], defined by $G_{m,n} = (4\pi/\sqrt{3}a)(\sqrt{m^2 + n^2 + m \cdot n})$ with m and n being indexes of reciprocals [14]. Then a planar waveguide was fabricated by a proton-exchange method. A laser-diode-pumped, Q-switched Nd:YAG dual-wavelength laser provided two z-polarized fundamental waves at 1.064 and 1.319 μm at the same time. The propagation constants of fundamental modes in the waveguide are $\beta(1.064\ \mu\text{m}) = 12.678\ \mu\text{m}^{-1}$ and $\beta(1.319\ \mu\text{m}) = 10.177\ \mu\text{m}^{-1}$. A screen was set at the position 15 cm away from the end face of the sample and no lens was used after the output end face. The operating temperature was kept at 25 °C.

As these two fundamental beams were collinearly coupled into the waveguide by a cylindrical lens transmitting along the x axis of the crystal, a beautifully colorful pattern like a "Christmas tree," which was composed of spots and lines with red (R), green (G), and yellow (Y), was projected on the screen behind the crystal [Fig. 3(a)]. In the picture, five vertical lines, three with green and two with red, at the bottom of the picture, compose the "stems" of "tree." They correspond to the guided-to-guided QPM-SHG processes [15], but are not perfectly phase matched at 25 °C (or else, they will be brighter than that in the picture). The "crown" of the tree consists of colorful spots with red, yellow, and green. They distribute in mirror symmetry as shown in Fig. 3(a). All the spots on the screen are Čerenkov radiation modes. They can be divided into groups and each contains three red, yellow, and green spots. The spots in a group associate the same $\vec{G}_{m,n}$ as indexed in Fig. 3(b). Red and green originate from SHG of two fundamental waves, respectively, and yellow spots come from their sum frequency. The brightest three spots locating in the center area of Fig. 3(a) are a direct Čerenkov SHG or SFG in which no reciprocal vector is involved. These spots could appear in a homogenous $LiTaO_3$ planar waveguide as well. The yellow one in the center is located

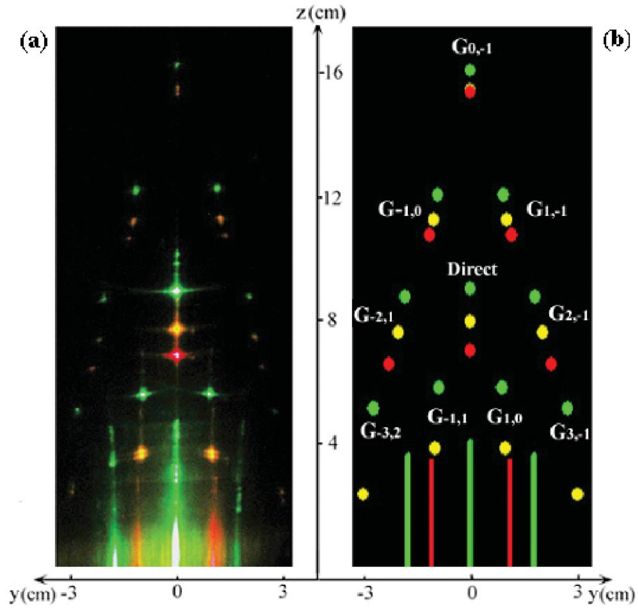

FIG. 3 (color). The pattern of NCR from fundamental waves of 1.064 and 1.319 μm projected on the screen behind the sample: (a) measured and (b) calculated.

at an angle of 27.4° with the waveguide, which is in good agreement with the calculated 27.5° from Eq. (4). Two weaker green spots, which present just above the brightest green spot shown in Fig. 3(a), are SH of high-order guide modes of 1.064 μm. The lower spots associate with forward $\vec{G}_{m,n}$ which retards the v_p of $P_{\rm non}$ wave, while the spots at the higher position involve backward $\vec{G}_{m,n}$ that accelerates v_p. Especially, three spots in that group locating on the top of crown correspond to $\vec{G}_{0,-1}$. It is antiparallel with the propagation direction of the fundamental waves. The exit angle of the yellow spot in this group is measured to be about 46.8°, which is much larger than that of direct Čerenkov SFG. In this case, the actual phase velocity v_p [$1.06v'$, calculated from Eq. (3)] goes faster than that ($1.02v'$) in the homogenous medium. The increase of v_p results in a larger Čerenkov angle. The result hints at the possibility of NCR in the frequency down-conversion process as long as actual v_p exceeds v' by introducing a backward reciprocal—for example, $v_p = 0.95v'$ in difference frequency generation with fundamental waves of 0.532 and 1.064 μm, which could be compensated using a reversed reciprocal.

In Fig. 3(a), we found a new phenomenon—an arc tangent with each Čerenkov spot, the brighter spot the brighter arc. According to analyses, these arcs are involved with elastic scattering of incident light in waveguide and emit in NCR by sum frequency of scattering and incident light. In a previous Letter [16], we reported conical SHG by conversional sum frequency of scattering and incident light in 2D nonlinear photonic crystal. In this experiment, such a sum-frequency process was achieved in Čerenkov

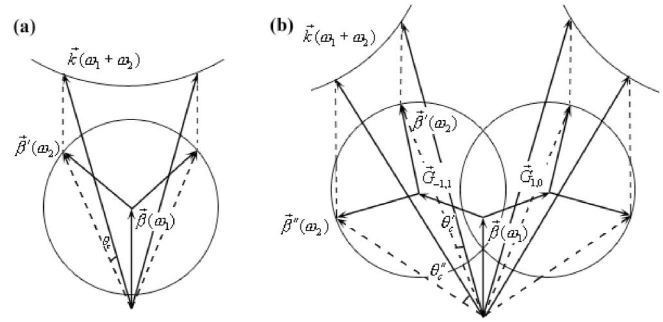

FIG. 4. Phase-matching processes for Čerenkov arcs of direct NCR (a) and QPM-NCR involving $\vec{G}_{-1,1}$ and $\vec{G}_{1,0}$ (b) with scattered $\vec{\beta}'(\omega_2)$.

radiation. In contrast to the bulk case, scattering light mainly exists in the x-y plane due to confinement of the waveguide. The Čerenkov condition may be understood in terms of Eq. (4), where one of two fundamental waves, such as $\vec{\beta}(\omega_2)$, is replaced by the scattering wave $\vec{\beta}'(\omega_2)$ that presents a continuously and symmetrically spatial distribution from $\vec{\beta}(\omega_2)$ [16]. The phase-matching condition for this configuration can be written as

$$|\vec{\beta}(\omega_1) + \vec{\beta}'(\omega_2) + \vec{G}_{m,n}| = k(\omega_3)\cos\theta_c. \qquad (5)$$

Phase-matching geometries are shown in Fig. 4, where Fig. 4 (a) corresponds to a direct NCR, and (b) correspond to QPM-NCRs being concerned with $\vec{G}_{-1,1}$ and $\vec{G}_{1,0}$, respectively. Obviously, one spot corresponds to one arc in SHG processes. If $\omega_1 \neq \omega_2$ in SFG processes, there will be two arcs with different curvatures to be tangent at the same spot because of $\vec{\beta}(\omega_1) + \vec{\beta}'(\omega_2) \neq \vec{\beta}'(\omega_1) + \vec{\beta}(\omega_2)$. However, the calculated result shows that the maximum interval for these two yellow arcs in our experiment is less than 0.1 mm, which is too small to be differentiated from the imaging on screen.

NCR can be analyzed by following the coupled-mode theory [7,8]. For the case of SHG, an approximately analytic expression of SH power can be deduced from the coupled wave equations by using a nondepletion approximation as below:

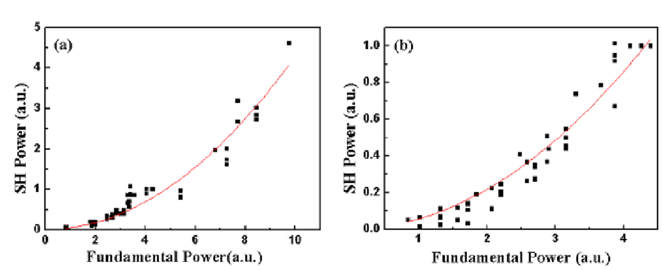

FIG. 5 (color online). Dependence of SH power on input power of fundamental 1.064 μm. (a) Direct Čerenkov SHG and (b) QPM Čerenkov SHG with $\vec{G}_{1,-1}$. Dots are measured data which are well fitted to be quadratic (line).

TABLE I. Measured and calculated relative intensity of Čerenkov SHG.

Intensity of SHG of 1.064 μm	Direct Čerenkov SHG	Čerenkov SHG with $\vec{G}_{1,0}$	Čerenkov SHG with $\vec{G}_{-1,1}$	Čerenkov SHG with $\vec{G}_{1,-1}$	Čerenkov SHG with $\vec{G}_{-1,0}$	Čerenkov SHG with $\vec{G}_{0,-1}$
Measured (a.u.)	1	0.41	0.42	0.17	0.17	0.10
Calculated (a.u.)	1 (Assumed)	0.43	0.43	0.16	0.16	0.08

$$P(2\omega) = 2l\pi[P(\omega)]^2|I_s + \alpha I_w|^2 \bar{d}_{33}^2 \frac{|2\frac{\omega}{v}\vec{x} + \vec{G}_{m,n}|}{\rho} \times \left[\frac{n_e(2\omega)}{n_o(2\omega)}\right]^2, \tag{6}$$

where $P(\omega)$ and $P(2\omega)$ are powers of the fundamental wave and SH wave, l is the waveguide length, I_s and I_w represent the overlap of electric fields between fundamental and SH waves in substrate and in waveguide, $\bar{d}_{33}$ is an effective nonlinear coefficient, $\alpha < 1$ is due to the reduction of $\bar{d}_{33}$ in the proton-exchanged waveguide, ρ is the wave number of radiation mode, $n_e(2\omega)$ and $n_o(2\omega)$ are the extraordinary and ordinary refractive index in the medium. For the details of deductions, one can refer to [7,8]. Figure 5 shows the dependence of Čerenkov SH power on the input of fundamental waves at 1.064 μm. The harmonic power increased proportionally with the square of the fundamental power, for both direct Čerenkov SHG and QPM Čerenkov SHG, which is consistent with theoretical analysis. Assuming that the intensity of direct Čerenkov radiation is 1, we calculated relative intensity of QPM Čerenkov SHG with different $\vec{G}_{m,n}$ at the same input power. The result is well consistent with the measured data, as shown in Table I. The coefficient α was fitted to be about 70% for our sample. It has been mentioned that Čerenkov radiation strongly depends on the discontinuity at the interfaces of waveguide [6]. The radiation in our experiment was weakened because of a graded index due to the annealing process in waveguide fabrication.

In general, the linear susceptibility tensor $\chi^{(1)}$ associates with the refractive index. It is a constant in a $\chi^{(2)}$ photonic crystal with inverse domain structure. $\chi^{(2)}$ photonic crystal is equivalent to a homogenous crystal in the linear optical regime. However, as incident light is strong enough to be able to drive an effective nonlinear polarization P_{non}, the situation will be greatly different. The modulation of $\chi^{(2)}$ can change the phase velocity of P_{non}. If $v_p = v'$, where v' is the phase velocity of free wave at the same frequency in the medium, a conventional nonlinear frequency conversion occurs, meanwhile, $\vec{v}'$ is collinear with $\vec{v}_p$. As $v_p > v'$, although phase matching cannot achieve collinearly, effective frequency conversion can still be achieved by the NCR. As $v_p < v'$, no matching geometry exists: neither QPM parametric process nor QPM-NCR process can be observed. Compared with the conventional QPM parametric process, QPM-NCR can extend the tolerance of phase matching from $v_p = v'$ to $v_p \geq v'$, and multiple NCR processes can exist simultaneously because the Čerenkov phase-matching condition is automatically satisfied. Because of the fact that there are multiple modulation periods with hexagonal symmetry for a 2D hexagonal structure and every modulated period corresponds to one phase velocity, it is easily understood why NCRs can be divided into a group with the index of reciprocal m, n. And also it exhibits more plentiful radiation patterns compared with a direct NCR. These have been verified from theory to experiment in this work. In this sense, the action of $\chi^{(2)}$ modulation can be understood as an effective approach to control the phase velocity v_p of P_{non} to achieve the required nonlinear parametric process. For example, the backward reciprocal can be used to accelerate v_p of P_{non}, which provided a possibility to realize QPM-NCR in a parametric down-conversion process. A probable application for this process is to generate multiple entangled photon pairs for potential quantum communication and computation networks.

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*To whom correspondence should be addressed.
zhusn@nju.edu.cn

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