Nonlinear Cherenkov Radiation in an Anomalous Dispersive Medium

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(Received 20 December 2011; published 29 May 2012)

By using ultrathin nonlinear media, the phase velocity of nonlinear polarization waves can be continuously adjusted. We realized nonlinear Cherenkov radiation (NCR) in an anomalous dispersion medium experimentally, which breaks the minimum speed limit of NCR. This modified nonlinear Cherenkov radiation has two new special states with respect to its prior, i.e., the degenerate state and extreme state. Also, periodically arranged ultrathin nonlinear media can enhance NCR and realize the nonlinear Smith-Purcell effect.

DOI: 10.1103/PhysRevLett.108.223901 PACS numbers: 42.65.Ky, 42.70.Mp, 68.35.Dv

Oscillators driven by a charged particle moving faster than the phase velocity of light in a medium can emit coherent light, known as Cherenkov radiation (CR)[1]. Conventionally, CR possesses three key characteristics: (1) a strict velocity threshold, (2) a forward-propagating energy, and (3) a forward-pointing conical wave front. However, these characteristics can be modified due to local structures. For example, in the Smith-Purcell effect [2], one can lower the velocity threshold by changing the metal grating's periodicities. In other cases: negative-index medium [3] and photonic crystal, unusual CRs have also been predicted to flow backward, i.e., opposite to the particle velocity. Moreover, photonic crystal structures enable a backward-pointing radiation cone [4]. These modifications allow potential applications like detector or counter, better light sources for frequencies that are difficult to access.

Nonlinearity is a universal phenomenon and most physics effects have their nonlinear counterparts, such as nonlinear Bragg diffraction [5], nonlinear Raman-Nath diffraction [6], and nonlinear Talbot effect [7]. In nonlinear optics, nonlinear Cherenkov radiation (NCR) has also been demonstrated [8], i.e., when light propagates in a nonlinear optical crystal, it can stimulate a spatially extended collection of dipoles through $\chi^{(2)}$, resulting in radiating coherent harmonics in the Cherenkov direction. Since then, NCR has been applied to ultrashort pulse characterization [9,10], domain wall imaging technique [11,12], and third harmonic generation [13,14]. While NCR still possesses the same characteristics as CR: the basic characteristic is that the phase velocity of the fundamental beam should exceed the harmonic radiation's phase velocity. In other words, NCR only occurs in a normal-dispersive medium, which means the first order derivative of wave vector with respect of frequency is positive. However, how NCR behaves under anomalous dispersion remains unclear.

In this work, we experimentally demonstrated the NCR process in an anomalous dispersive medium without phase velocity threshold, due to the modulation by nonlinear material with special geometry—ultrathin nonlinear planar material in this case. We also show a potential existence of transversely propagating energy and degenerated wave front. Periodic structures play a key role in modulating CR and NCR processes in previous studies. The presence of Bloch waves in photonic crystals make CR substantially different from that in one uniform medium, lowering the phase velocity threshold [4]. For NCR, domain reversal in nonlinear photonic crystals can add π phase jump to polarization wave, and thus affect the polarization wave speed [8]. Here we introduce one simpler method to tune phase velocity of nonlinear polarization wave from the velocity of incident light to infinity, by confining nonlinear polarization wave around one ultrathin nonlinear material, which has the same refractive index as the surrounding but high nonlinear coefficient.

For simplicity, we consider the fundamental wave as a plane one. In bulk nonlinear material, light can spread freely, and nonlinear polarization is described as

$$P = 2\varepsilon_0 d_{\mathrm{eff}} E_1^2 \exp[-i(2\vec{k}\cdot\vec{r} - 2\omega t)], \tag{1}$$

where d_{eff} is the effective nonlinear coefficient, E_1, k, and ω are the amplitude, wave vector, and frequency of incident light, respectively. But in the case of Fig. 1(a), fundamental beam propagates at an angle γ with respect to an ultrathin material, the dimension of which is smaller than the beam waist, then the nonlinear polarization wave will only propagate along the ultrathin material, and Eq. (1) will turn into a one-dimensional form:

$$P = 2\varepsilon_0 d_{\mathrm{eff}} E_1^2 \exp[-i(2kr\cos\gamma - 2\omega t)]. \tag{2}$$

In the direction along the ultrathin material, $k_{\mathrm{np}} = 2k\cos\gamma$, so the phase velocity of polarization will be $v_{\mathrm{np}} = v/\cos\gamma$, and the NCR phase matching condition will vary with v_{np},

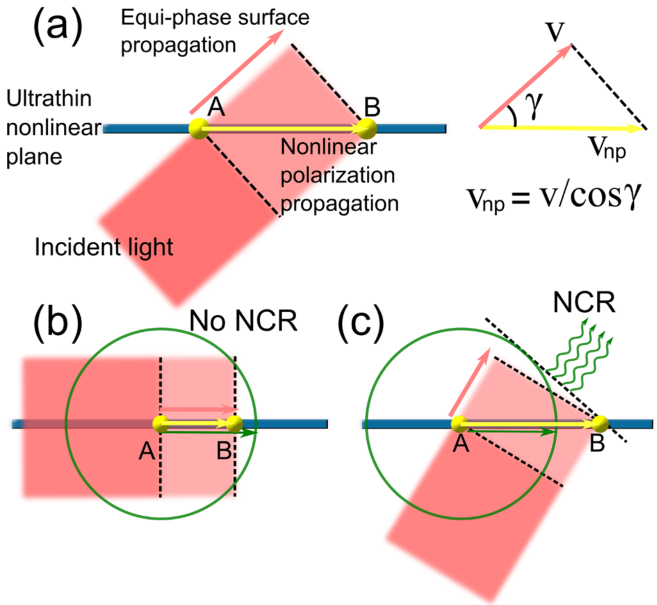

FIG. 1 (color online). Red along the incident arrow denotes the fundamental wave, the yellow arrow along the ultrathin plane denotes the polarization wave, and the green arrow in (b) and (c) denotes the harmonic wave. (a) Modulating of polarization wave velocity by the ultrathin nonlinear plane; (b) The fundamental wave cannot exceed the harmonics phase velocity in an anomalous dispersion medium; (c) Modulation of the polarization wave breaks the velocity threshold.

$$\cos\theta_c = \frac{v_2}{v_{\mathrm{np}}} = \frac{v_2 \cos\gamma}{v} = \frac{n_1 \cos\gamma}{n_2}, \tag{3}$$

where θ_c is the Cherenkov angle, subscript 1, 2 and np mark the fundamental and second harmonic and the nonlinear polarization waves, and n denotes the refractive index of the medium.

With proper γ, there is no more minimum phase velocity limit for NCR, so it can even exist in anomalously dispersive medium. From the perspective of Huygens' Principle, it is much easier to understand. As illustrated in Fig. 1(b), the velocity of the polarization wave is slower than that of the harmonics in the anomalously dispersive medium, if incident light is parallel to the plane, i.e., $\gamma = 0°$, harmonics generated at point A and point B cannot constructively interfere. Changing the incident angle γ until it satisfies $\cos\gamma \leq n_2/n_1$, then the harmonics from point A and B will interfere constructively at Cherenkov angle and thus NCR is generated [Fig. 1(c)].

There are two great obstacles in demonstrating NCR experimentally in anomalously dispersive plane medium. First, anomalously dispersive medium always has a strong absorption band [15], then the fundamental beam may be totally absorbed before NCR is generated. Second, it is hard to fabricate ultrathin nonlinear material less than several micrometers by mechanical methods. Fortunately, NCR is a nonlinear process associated with the polarization properties of light. Utilizing crystal birefringence, fundamental and harmonic beams with different polarization can effectively simulate an anomalous-dispersion-like environment in some normally dispersive medium. Taking the oo-e type NCR at the fundamental wavelength of 1064 nm as an example, the refractive index of e-polarized harmonic is lower than the index of o-polarized fundamental in a 5 mol% MgO: $LiNbO_3$ crystal at room temperature Fig. 2(a), which is anomalously dispersive in effect. To overcome the second obstacle, one method is coating a nonlinear material layer on a substrate, but here we choose another fascinating ultrathin material, the ferroelectric domain wall [16]. Ferroelectric domains refer to regions polarized in one of the built-in spontaneous polarization directions. Domain can be inversed at room temperature by electrical poling technique [17]; in this process, domain walls separating the two domains with $+P$ and $-P$ are formed. Figure 2(b) shows the scanning electron microscope (SEM) image of a domain wall in our sample. Ideal ferroelectric domain walls are well accepted to be only several lattice units wide [12,18]. Many reports indicate that walls show enhanced optical nonlinearity, which can generate stronger NCR than single-domain area [19–21].

We used two z-cut (z axis is the crystals' polar axis) samples of 5 mol% MgO:$LiNbO_3$ in the size of

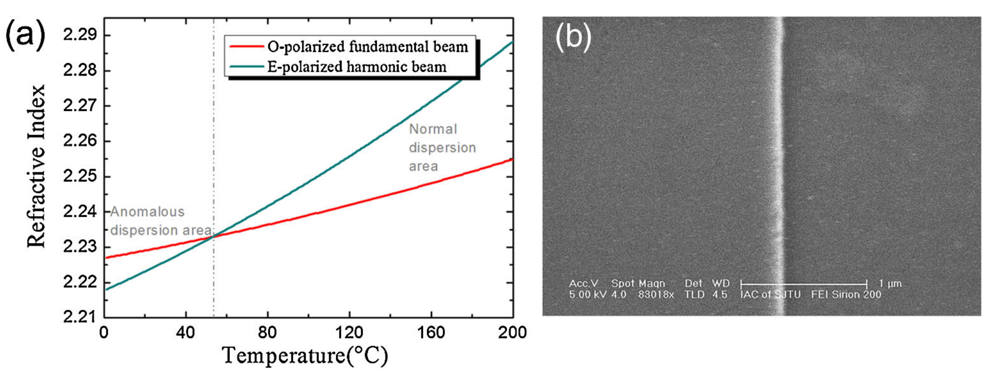

FIG. 2 (color online). (a) Theoretical refractive index in 5 mol% MgO: $LiNbO_3$ crystal; (b) SEM image of domain wall in the sample

5 cm × 1 cm × 0.2 cm ($z \times y \times z$). The first sample was a single-domain crystal used to check the validity of material's anomalous dispersion. In the second one, a domain wall along the y-z plane was fabricated by large scale electrical poling technique. The fundamental beam was derived from a mode-locked Nd:YAG laser which generated 40 ps pulses centered at a wavelength of 1064 nm, a pulse energy of 0.4 mJ and a beam waist radius of 20 μm was employed. It was loosely focused into the sample ($f = 10$ cm) along the y-axis.

The result of the single-domain crystal is shown in Fig. 3(a). When the fundamental beam was e-polarized, an e-polarized nonlinear Cherenkov ring was clearly observed. But when we tuned the fundamental beam to be o-polarized, the Cherenkov ring disappeared, and there was only the phase-mismatched collinear 2nd harmonic in the middle. This verifies that NCR did not exist in the anomalous dispersive bulk material. Then we attenuated beam intensity until NCR could be observed only when the e-polarized fundamental beam focused in the domain wall. This time, the NCR was no longer a ring, but degenerated into two spots [12]. Figure 3(b) shows the oo-e type NCR pattern of single wall with the increasing of incident angle γ. Still, when fundamental beam was o-polarized

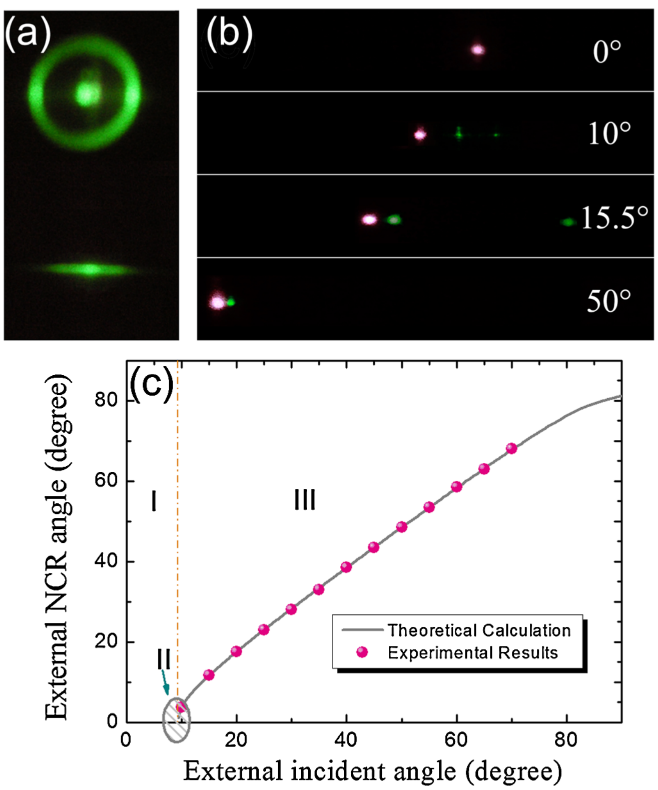

FIG. 3 (color online). (a) NCR pattern of e-polarized and o-polarized incident light in single-domain crystal; (b) NCR pattern of o-polarized incident light at single wall, the incident angle is marked on the right; (c) Relationship between NCR angle and incidence angle in single domain wall experiment.

and propagated along the domain wall, NCR was not observed, and it appeared until $\gamma > 9.8°$. The emergence angle of the NCR pair can be clearly measured in Fig. 3(c), which was consistent with Eq. (3).

There are four stages with different γ: I $0° \leq \gamma < \cos^{-1}(n_2/n_1)$; II $\gamma = \cos^{-1}(n_2/n_1)$; III $\cos^{-1}(n_2/n_1) < \gamma < 90°$; and IV $\gamma = 90°$ [Fig. 3(c)]. In stage I, the phase velocity of the nonlinear polarization does not exceed that of the harmonic beam, while in stage III, the Cherenkov condition is fulfilled. Stage II is a special case that the Cherenkov pair degenerates to a forward-propagating wave front. The nonlinear polarization propagating along the plane has the same phase velocity as the harmonic wave, so it becomes a collinear phase matching process. However, degenerated NCR was not observed in our experiment, because the domain wall is a very narrow transitional area from negative domain to the positive domain, it also can be divided into two parts with the nonlinear coefficient of opposite sign, and harmonic waves from positive and negative parts destructively interfere in the far field along the direction of the plane. In stage IV, $\gamma = 90°$ is another extreme case, where the phase velocity of polarization wave becomes infinite according to $v_{\rm np} = v/\cos\gamma$. It also can be explained in the view of Huygens' Principle. Since all the nonlinear oscillators in the plane are stimulated by the same equiphase surface of fundamental beam, the generated harmonic waves are naturally in phase, which will not propagate forward but transversely to the nonlinear polarization wave. The experimental results in Fig. 3(b) and 3(c) indicate the trend to this extreme case. If the domain wall is changed into an ultrathin column anomalously dispersive nonlinear material, such as nonlinear nanowire material, it will extend 2-D cases to 3-D cases as shown in Fig. 4. Materials with negative refractive index also give a possibility of backward Cherenkov cone and energy propagation by reversed Snell's Law, which may extend research on this new nonlinear phenomenon.

Smith-Purcell effect is a special kind of Cherenkov radiation that can be enhanced in certain directions at

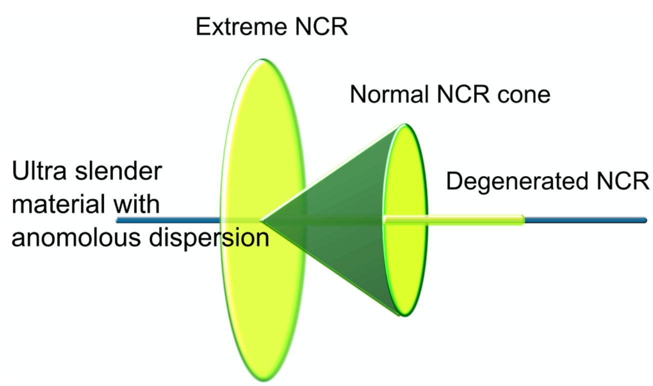

FIG. 4 (color online). Potential 3-D cases in an ultrathin column anomalously dispersive nonlinear material.

certain wavelengths through periodic structure, which is without minimum particle velocity limit. Its radiation direction θ satisfies

$$d\left(\frac{1}{\beta} - n\cos\theta\right) = m\lambda \qquad m = 0, 1, 2\ldots, \tag{4}$$

where d is the period of grating, $\beta = v/c$, v is particle's velocity, λ is the wavelength of the emitting light, and n is its refractive index. Similarly, NCR generated from adjacent planes also can superpose coherently if their phase difference is multiple times of 2π, which is determined by

$$\frac{d}{\sin\gamma}(n_1 - n_2\cos(\gamma - \theta)) = m\lambda_2 \qquad m = 0, 1, 2\ldots \tag{5}$$

λ_2 is the wavelength of the emitting light, which is 2nd harmonic, γ and θ are the incident and Cherenkov angle, respectively, and γ-θ is the actual radiation direction. As we can see, Eq. (5) nearly has the same form as Eq. (4), so this enhanced NCR can be considered as nonlinear Smith-Purcell effect.

Experimentally, we used the periodically domain wall series to verify this, because the electrical poling technique has been very mature, and the period can be fabricated as small as several micrometers with good duty cycle. We employed a 200 mW continuous laser beam at a wavelength of 1064 nm, and the sample was 5%/mol MgO: PPLN (periodically poled $MgO:LiNbO_3$) in the period of 30 μm. The room temperature was kept on 22.5 °C, because the matching incident angle γ was sensitive to the temperature. The inset of Fig. 5(a) shows the comparison of theoretical and experimental NCR intensity of different incident angles. As expected, NCR's intensity was greatly increased when the incident angle satisfied Eq. (5), where γ was determined by θ according to Eq. (3). We also simulated the NCR intensity changing with fundamental wavelength λ_1 and incident angle γ, taking the period $d =$ 30 μm [Fig. 5(a)]. If the incident light is broad bandwidth [22], it will stimulate nonlinear polarizations at different frequencies simultaneously, and the periodically placed planes enhance NCR of different frequencies in different directions. Temperature can affect the refractive index, and change the conditions of the nonlinear Smith-Purcell effect. We measured the incident angle of the 2nd order nonlinear Smith-Purcell peak ($m = 2$) of a sample in the period of 6.95 μm, by changing the temperature from anomalously dispersive region to normally dispersive region. As shown in Fig. 5(b), this enhanced peak exists through the whole regions from anomalous dispersion to normal dispersion, which gives good demonstration that a nonlinear Smith-Purcell effect possesses similar characteristics as its linear counterpart. A nonlinear Smith-Purcell effect exists through the whole transparent window of the material, no matter the dispersive property, which strongly extends its potential applications in optical detecting, harmonic generation, as well as in photonics studies.

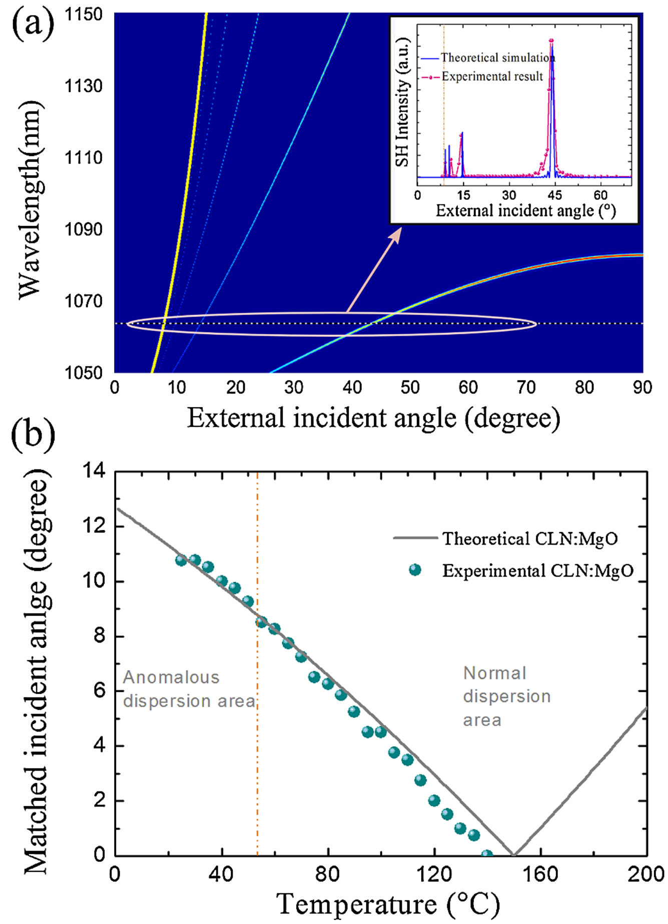

FIG. 5 (color online). (a) The incident angle satisfied the condition of the nonlinear Smith-Purcell effect in corresponding to different fundamental wavelength, the leftmost yellow line denotes the degenerated NCR; the inset is experimental results using continuous laser of 1064 nm. (b) Nonlinear Smith-Purcell effect achieved in different dispersion area, by adjusting the temperature. Sample: 5 mol% MgO:PPLN.

In conclusion, ultrathin nonlinear material can accelerate the phase velocity of nonlinear polarization, making NCR available in anomalous dispersive nonlinear medium. The extreme cases imply essential physical connections between NCR and common collinear harmonic generation processes, which deserves more systematical investigation. The nonlinear Smith-Purcell effect is discussed in periodically placed ultrathin nonlinear medium planes, enabling enhanced NCR peaks in special directions by CW laser source.

This research was supported by the National Natural Science Foundation of China (Grant No. 61125503, 61078009, 11121504), the National Basic Research Program 973 of China (Grant No. 2011CB808101), the Foundation for Development of Science and Technology of Shanghai (Grant No. 11XD1402600) and the Open Fund of the State Key Laboratory of High Field Laser Physics.

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