

The role of ferroelectric domain wall in nonlinear Cerenkov frequency up-conversion in 1D nonlinear crystal

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We reveal a variety of nonlinear Cerenkov radiation (NCR) patterns that occur in a single photonic crystal modulated by domain walls, which manifest themselves as normal, degenerated, and anomalous-dispersion-like NCR type sum-frequency generation. The phase-matching geometries of the evolution of Cerenkov radiation varying with the dispersion relationship among the interaction waves are exploited, respectively.

Keywords: Nonlinear Cerenkov radiation; domain wall; sum-frequency.

1. Introduction

In recent years, there has been increasing attention given to ferroelectric domain walls and lots of brand new features are reported¹⁻⁹ which are quite different from those in bulk ferroelectric materials. Among these, the existence of domain wall could enhance^{10,11} and modulate¹² certain category of nonlinear effect, for instance, nonlinear Cerenkov radiation (NCR), which has been intensely studied. Oscillators driven by charged particles travelling faster than the phase velocity of light in the medium can emit coherent electromagnetic waves, which is known as Cerenkov radiation.^{13,14} Similarly, NCR emerges in nonlinear optics when the phase velocity of polarization wave exceeds that of the harmonic generation.¹⁵⁻¹⁷ Owing to

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the domain wall, input light with even moderate intensity could access nonlinear processes which can barely be observed in the bulk medium. In this case, the polarization wave is confined along domain wall direction, which could be noncollinear with fundamental wave when there is oblique incidence.

As long as the nonlinear polarization wave could only transmit along the domain wall, the phase velocity of polarization wave is modulated by a factor of γ , where γ is the angle between the plane of domain walls and the incident wave in the crystal. Previous studies have revealed that with proper γ , the phase velocity of polarization is accelerated enough to break through the ‘prohibited threshold’ of NCR in media of anomalous dispersion media.^{12,18} Stimulated by this result, intensive works about NCR in anomalous media have been done both in the periodically poled nonlinear crystal and also on the crystal surface,^{19,20} which are of great potential for practical applications.

So far, previous studies were focused on the performances of the essential role that domain wall played in the NCR involving either normal or anomalous dispersion situation. Here in this paper, by exploiting the dispersion relationship between incidences and harmonics, we demonstrate a generic successive picture of Cerenkov sum-frequency generation (CSFG), from normal dispersive to anomalous dispersive cases in a single nonlinear crystal.

2. Phase-Matching Condition

The intrinsic condition for NCR is the nonlinear polarization wave driven by incident field which has larger phase velocity than that of harmonic wave in the media. For normal incidence second harmonic generation, in other words, the nonlinear polarization propagating along the domain wall, the Cerenkov condition is defined as

$$\cos \theta = \frac{v_2}{v_1} = \frac{2k_1}{k_2} = \frac{n_1}{n_2}, \quad (1)$$

where v_1, v_2, k_1, k_2 , and n_1, n_2 denote the phase velocities, wave vector, and refractive indices of the fundamental and harmonic waves, respectively (see Fig. 1(a)). Considering the oblique incidence, the phase velocity of polarization wave v_{np} no longer equals the fundamental one v_1 , which is modulated by the factor of $\cos \gamma$. Since the nonlinear polarization wave is noncollinear with the fundamental wave, the phase velocity of nonlinear polarization wave is determined by the phase velocity of fundamental wave and the angle between them, which can be written as $v_{\text{np}} = v_1 / \cos \gamma$. It is worth noticing that the phase velocity of nonlinear polarization wave could be enlarged continuously by increasing the incident angle γ . The NCR phase-matching condition Eq. (1) will also vary with v_{np} as follows:

$$\cos \theta = \frac{v_2}{v_{\text{np}}} = \frac{v_2 \cos \gamma}{v_1} = \frac{n_1 \cos \gamma}{n_2}. \quad (2)$$

Obviously, Eq. (2) always holds for normal dispersion media. In the case of anomalous dispersion, by deliberately adjusting the incident angle to accelerate the

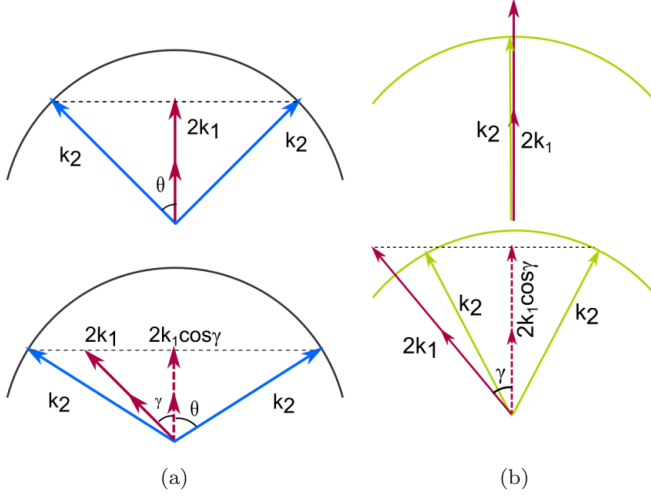


Fig. 1. The phase-matching geometries of incidences and Cerenkov harmonic generation in (a) normal and (b) anomalous dispersion media with normal and oblique incidences, respectively. k_1, k_2 instead of the wave vectors of the fundamental and harmonic waves, respectively. In both situation, the NCR is only determined by the longitudinal phase-matching conditions.

phase velocity of nonlinear polarization wave, one can realize the NCR as long as the phase-matching condition $v_1 \geq v_2 \cos \gamma$ is satisfied. The critical NCR phase-matching condition is defined as $\cos \theta = 1$, while the incident angle γ satisfies $\cos \gamma = n_2/n_1$. Beyond this point, NCR will be generated in the anomalous dispersion media (see Fig. 1(b)).

In the case of sum-frequency generation (SFG), analogy to Cerenkov second harmonic generation, it is required that the phase velocity of the sum-frequency polarization wave exceeds that of harmonics. When there exists domain wall, the modulated phase velocity of the sum-frequency polarization wave could be expressed as

$$v_p = \frac{\omega_3}{\left(\frac{\omega_1}{v_1} + \frac{\omega_2}{v_2}\right) \cos \gamma}, \quad (3)$$

where $\omega_1, \omega_2, \omega_3$ denote incidences and sum frequency, v_1, v_2 denote the phase velocities of incidences, respectively. By adjusting the incident angle, the phase velocity of sum-frequency nonlinear polarization could be accelerated and even exceeds that of the harmonic in anomalous dispersion environment resulting in nonlinear CSFG finally. The modulated CSFG angle with respect to domain wall is expressed as

$$\cos \theta = \frac{(|\vec{k}_1| + |\vec{k}_2|) \cos \gamma}{|\vec{k}_3|}, \quad (4)$$

where k_1, k_2 , and k_3 are the wave vectors of fundamental waves and the harmonic wave, respectively.

3. Dispersion Relationships

Previous researches showed that by utilizing the birefringence of nonlinear crystal, one can mimic anomalous dispersion environment in normal dispersion media.^{9,12,21} Here in SFG vision, we perform three kinds of wave vector relationships between interaction waves with different polarizations. The incidences F1 and F2 are settled at 800 nm and tunable from 1000 nm to 1600 nm, respectively, whose polarization could be adjusted flexibly. Figure 2 demonstrates the dispersion relationship of interaction waves varying with incident wavelength with eo-e, oe-e and oo-e phase-matching conditions, respectively.

In the first two situations (see Figs. 2(a) and 2(b)), which satisfy type II phase-matching condition, it is clear that the wave vectors of incident waves are always smaller than that of the harmonics, which obey normal dispersion circumstances. Previous studies have been intensely focused on several types of NCR in normal

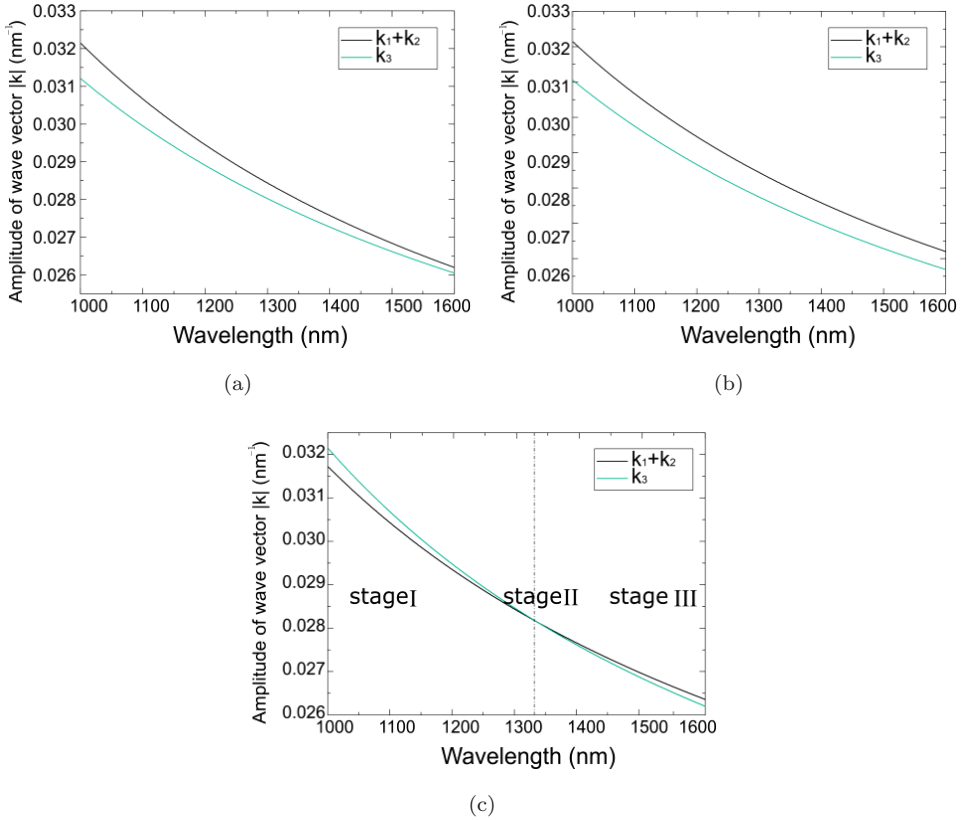


Fig. 2. (a) The dispersion relationship of interaction waves when F1 is extraordinary polarization and F2 is ordinary polarization. (b) The dispersion relationship of interaction waves when F1 is ordinary polarization and F2 is extraordinary polarization. (c) The dispersion relationship of interaction waves when F1 and F2 are both ordinary polarization.

dispersion media, which is not the key point in this work. In the third one (see Fig. 2(c)), the dispersion relationship underlies the fundamental waves and harmonics varying with the incident wavelengths under oo-e phase-matching condition. There are three cases of the wave vector relationship among the interaction waves: (I) $k_1 + k_2 < k_3$, (II) $k_1 + k_2 = k_3$, (III) $k_1 + k_2 > k_3$. Case I shows the normal dispersion circumstance where the CSFG condition is naturally satisfied. In Case II, the nonlinear Cerenkov degenerates into a forward-pointing wavefront, thus the wave vectors of fundamental and harmonic waves are parallel to each other. As regards Case III, the curves of wave vectors of incidences exceed that of the harmonic, which indicates such phase-matching pattern obeys anomalous dispersion relationship. Since the phase velocity of sum-frequency nonlinear polarization does not exceed that of the harmonic wave, one can expect that there would be no NCR generation in the normal incidence configuration.

4. Experiments

In our experiments, the two incident waves F1 and F2 are generated from the OPA (TOPAS, Coherent Inc.), with the residual pump (F1) centered at 800 nm and the signal (F2) tunable from 1000 nm to 1600 nm. The incidences are set both ordinarily polarized, synchronized and loosely focused into the sample along y -axis (domain walls). A periodically poled 5 mol.% MgO:LiNbO₃ crystal with dimensions of 15 mm (x) \times 5 mm (y) \times 0.5 mm (z) and period of 30 μ m is used in our experiments.

The screen is set at 5 cm behind the sample to record experimental patterns. There are three typical patterns as predicated while varying the incidence F2 from 1000 nm to 1600 nm, which are demonstrated in Figs. 3(a), 3(b), and 3(c), respectively. The first picture illustrates the CSFG and Cerenkov second harmonic generation while the incidences F1 and F2 are centered at 800/1250 nm, respectively. The outer pair of harmonic spots is the Cerenkov frequency doubling of incident wave F1, whose central wavelength is 400 nm and the external angle is 33.1°. When the incident wavelengths reach Case II, the momentum conservation law between the fundamental waves and generated harmonic waves are fulfilled in the collinear arrangement (see Fig. 2(e)). The collinear phase-matching between fundamental and sum-frequency wave is thus fulfilled which would maximize the conversion efficiency with the measured conversion efficiency up to 10.2%. This circumstance distinguishes from the collimated SFG in bulk media in that the confinement effect of domain walls greatly enhances the nonlinear polarization and improves the sum-frequency intensity.

In Case III, the same as theoretical predication, there is no NCR generation in the normal incidence configuration. By rotating the incident angle, the NCR can even exist in such anomalously dispersive medium while the modulated Cerenkov condition (Eq. (4)) is satisfied. Figure 3(f) illustrates the phase-matching geometry in such scheme. Distinguishing from those in normal dispersion, the pair of CSFG spots appears on the same side of incidences. The Cerenkov angle in anomalous

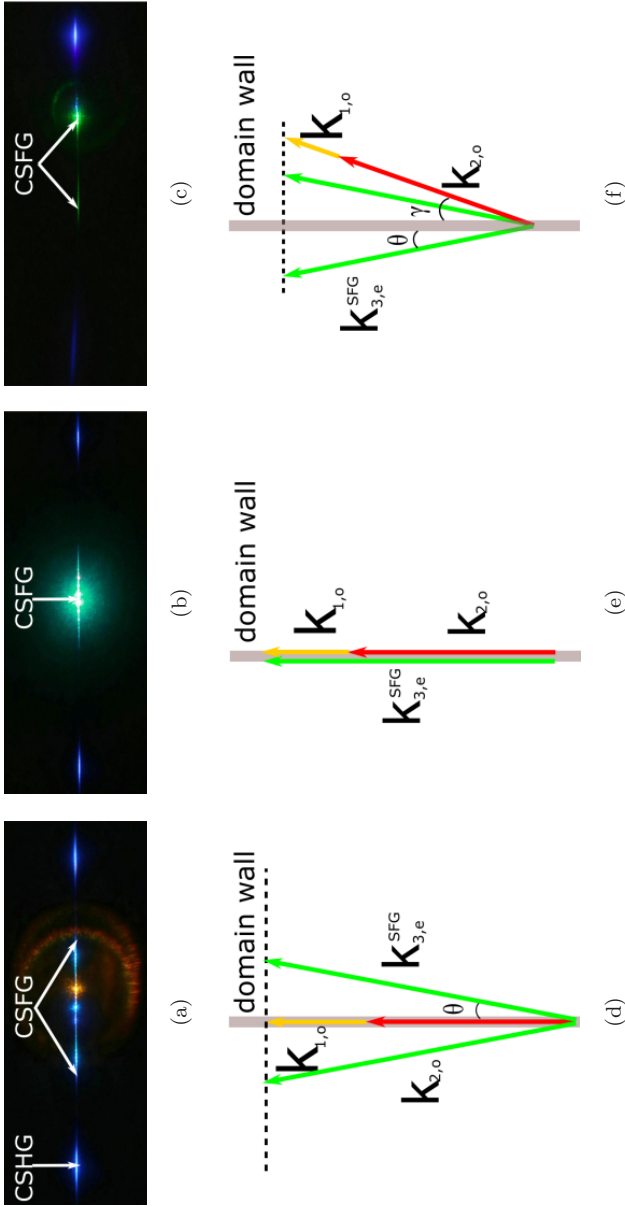


Fig. 3. (a)–(c) Illustrations of the typical patterns with the wavelength of incidence F2 at 1250 nm, 1340 nm and 1450 nm, respectively. (d)–(f) Schematics of the phase-match conditions of the CSFG in normal dispersion stage, degenerated stage, and anomalous dispersion stage, respectively.

dispersion situation strictly obeys Eq. (4). Overall, a successive scheme of normal, degenerated, and anomalous-dispersion-like CSFG can be realized in one single nonlinear crystal only by adjusting the incident wavelength.

5. Conclusions

In conclusion, a generic successive picture of CSFG, from normally dispersive to anomalously dispersive scheme, has been realized in one nonlinear crystal. Analysis on the three stages indicates that the Čerenkov phase-matching geometries vary with the dispersion condition. It is noteworthy that the degenerated Čerenkov generation localized in the domain wall region is achieved with forward-pointing wavefront, which shows the similarities and differences of essential physical features with common collinear up-conversion processes and deserves further investigation.

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