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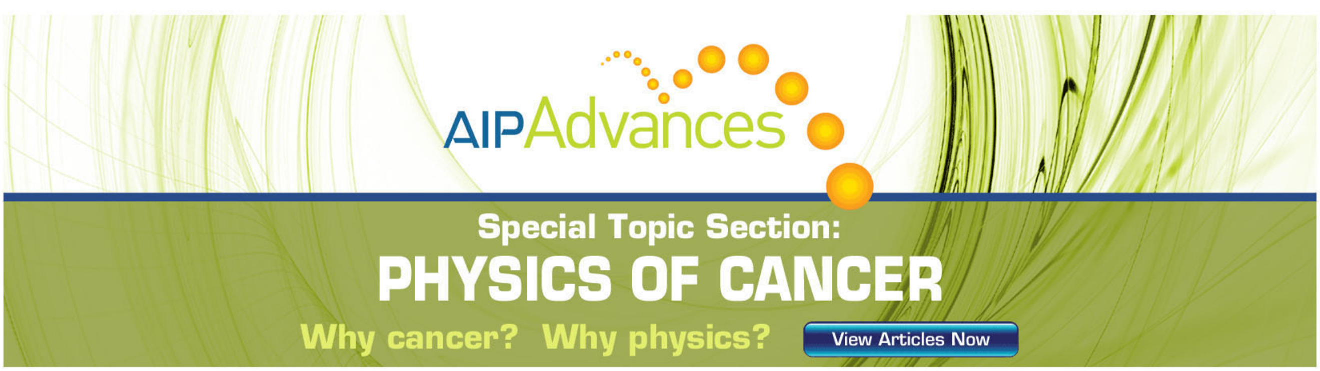

Cherenkov high-order harmonic generation by multistep cascading in $\chi^{(2)}$ nonlinear photonic crystal

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We propose a scheme for efficient Cherenkov high-order harmonic generation. Second to fifth order harmonic wave are observed in a single periodically poled ferroelectric crystal in our experiment. The noncollinear high-order harmonic generation is produced via enhanced Cherenkov second harmonic cascaded with successive multistep sum-frequency generation with simultaneously longitudinal phase-matching. The emission angle and power dependencies are analyzed in detail experimentally, which coincide with theoretical predictions. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4722931]

In the past decades, advances in multistep optical parametric processes based on periodically poled $\chi^{(2)}$ nonlinear crystals have motivated great interest in physics and applications.[1–3] As a method for high-order harmonic generation in single nonlinear media, multistep cascading represents a special type of second-order parametric process that involves several different second-order nonlinear interactions. In particular, much attention has been devoted to the investigation of various schemes of the cascaded Cherenkov-type parametric processes in $\chi^{(2)}$ nonlinear crystals,[4–11] where the phase velocity of radiation source v (i.e., nonlinear polarization) driven by the incident wave is faster than that of the harmonic wave v′. Experimental demonstration of the cascaded Cherenkov third harmonic generation (THG) in random quadratic media[12] and aperiodically poled $LiTaO_3$ waveguide[2] were reported recently, while more research effort is emphasized on the enhancement of such process with fundamental wave propagating along the domain walls.[13]

In previous Cherenkov-type parametric processes, what is worth highly noticing is that such Cherenkov emissions are remarkably enhanced by the existence of domain walls.[6,9,13,14] This enhancement provides a reliable guarantee of Cherenkov harmonics for further practical applications, for instance, domain structure characterization,[15] second-harmonic microscopy, and so on. Additionally, the significant enhancement of second harmonic generation (SHG) is supposed to support the generation of efficient high-order harmonics in multistep cascaded Cherenkov scheme, which have significant potential for optical waveforms synthesis[16] and extreme ultraviolet generation.[17]

In this work, we investigate the multistep cascaded Cherenkov second to fifth harmonic generation in periodically poled $LiNbO_3$ crystals (PPLN). By illuminating the PPLN along the domain walls (y-axis), we observed the cascaded multiple Cherenkov high-order harmonic generation, which is up to fifth-order, with simultaneously longitudinal phase-matching. The emission angle and conversion efficiency of each cascaded step via varying the wavelength of fundamental beam are analyzed experimentally, which are well agreeable with theoretical predictions.

The $\chi^{(2)}$ nonlinear crystal used in our experiment is fabricated via standard electric field poling of a Z-cut $LiNbO_3$ crystal at room temperature. The dimensions of the poled crystal are 15 mm (x) × 5 mm (y) × 0.5 mm (z), with the inversion periods of 30 μm (duty ratio of 1:1). The light source is an optical parametric amplifier (TOPAS, Coherent Inc.) generating 80 femtosecond pulses (1000 Hz rep. rate) at wavelength tunable from 280 nm to 2600 nm. The extraordinarily polarized fundamental wave propagates paralleled to the domain wall (y-axis) of the crystal and loosely focused by a 50.8 mm focal length lens, which creates a beam diameter of 100 μm. The emitted patterns are projected onto a screen located 5 cm behind the crystal (Fig. 1(a)).

Figure 1(b) displays the experimental image obtained with fundamental wavelength at 2050 nm. In this case, the recorded pattern consists of three visible symmetrical dot pairs with different frequencies and emission angles. From inner to outer, the symmetrical multicolor dot pairs are third, fourth, and fifth harmonic generation of fundamental beam, whose central wavelengths are 684 nm, 513 nm, and 410 nm, respectively. While the second harmonic wavelength is located in the near infrared field, it is invisible on the screen. However, it can be detected in spectrum (see Fig. 1(c)). Furthermore, we explored the polarization characteristics of the three neighboring spots on each side of the fundamental wave, which are all extraordinary-polarized. Considering that the incident beam is extraordinary polarized, we confirmed the interactions that are responsible for these harmonic spots as type 0 (e-ee) phase-matching.

We studied these nonlinear parametric interactions that involve several processes by employing the phase-matching conditions illustrated in Fig. 2. In the Cherenkov scheme, the general vector phase matching condition ($\mathbf{k}_2 = 2\mathbf{k}_1 + \Delta\mathbf{k}$) is

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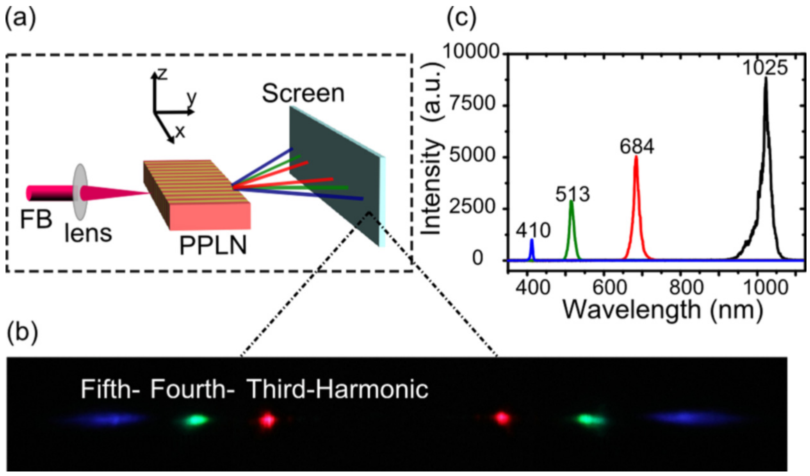

FIG. 1. (a) Schematic of the experimental setup with fundamental beam propagating along the domain wall. (b) Image of the spatial profiles of the Cherenkov harmonics at 2050 nm input wavelength. From the inside to the outside, the pair of harmonic wavelength are centered at 684 nm, 513 nm, and 410 nm, corresponding to the third, fourth, and fifth harmonic generation, respectively. (c) The spectrum of each order harmonic wave.

evolved to be only longitudinally fulfilled, which is $k_2 \cos\theta = k_1$. It is well established recently that the existence of domain wall prominently enhances the SHG (Refs. 9, 18, and 19), which would relatively promote overall conversion efficiency of the subsequent multistep processes. In this case, two parametric interactions, the SHG, and sum frequency generation process, result in a cascaded Cherenkov THG, which mimics the effect of strong third order nonlinearity despite of the small $\chi^{(3)}$ in congruent samples. Similarly, as long as the polarization phase velocity v exceeds the velocity of the harmonic radiation v′, namely Cherenkov radiation condition, it is no doubt that high-order cascading processes will consecutively come up. Given this prospect, parametric coupling between the cascaded Cherenkov THG and the incident wave (including second harmonic generation by the generated SHG waves) would naturally occur, which gives birth to a fourth harmonic generation. The fifth harmonic generation at 410 nm is also observed due to the cascaded fourth harmonic wave frequency mixing from the fundamental wave, or from wave frequency mixing by generated second- and third-order harmonic waves.

Giving the whole scene of the cascading processes, the corresponding phase matching condition for the multistep high-order harmonic generation can be described as

$$
\begin{aligned}
k_2 \cos\theta_2 &= 2k_1,\\
k_3 \cos\theta_3 &= k_2 \cos\theta_2 + k_1 = 3k_1,\\
k_4 \cos\theta_4 &= k_3 \cos\theta_3 + k_1 = 4k_1,\\
k_4 \cos\theta_4 &= 2k_2 \cos\theta_2 = 4k_1,\\
k_5 \cos\theta_5 &= k_4 \cos\theta_4 + k_1 = 5k_1,\\
k_5 \cos\theta_4 &= k_2 \cos\theta_2 + k_3 \cos\theta_3 = 5k_1.
\end{aligned}
\qquad (1)
$$

We attribute the frequency quadrupling and quintupling governed by single longitudinal phase-matching conditions of Eq. (1) to be Cherenkov-type fourth and fifth harmonic generation. From the phase-matching conditions, we can obtain that the internal emission angle of Cherenkov harmonic depends only on the ratio of refractive index of incident beam and its harmonic wave, which is defined as $\theta_i = \cos^{-1}(n_1/n_i)$, here $i = 2, 3, 4, 5$ denoting the ith Cherenkov harmonic generation. The measured values of θ_3, θ_4, and θ_5 are 32.6°, 46.1°, and 67.4°, respectively, which are in well agreement with the predictions of calculated ones.

When tuning the fundamental wavelength from 1600 nm to 2200 nm continuously, the evolution of output pattern recorded on the screen is shown in Fig. 3(a). The corresponding Cherenkov fourth harmonic generation is observed all the time with its color varying continuously from blue to green, whereas the fifth harmonic emerges only when the wavelength is above 2000 nm due to the absorption of lithium niobate crystal. Moreover, in Fig. 3(b), we presented the experimentally measured values of radiation angle for each visible harmonic wave. It is clear that θ_i appears to be monotonically decreasing with the growth of fundamental beam wavelength. The experimental data fit well with the theoretical curves. What is worth noting is the fact that the fifth harmonic signal (blue) has a large divergence angle in the far-field region, so there is an inevitable slight discrepancy due to the selection of a central location of the recorded spots. The dependence of domain period of PPLN is verified experimentally by using seven PPLN samples with different poling periods (5.24, 6.92, 8.1, 10.83, 13.86, 19.47, and 30 μm) (see Fig. 3(c)).

Furthermore, we set the sample on a stepping motor and used a microscope objective (50×) to scan the sample along x-axis with step length of 1 μm. In this manner, the beam, whose diameter is about 5 μm, was monitored at different locations throughout the PPLN crystal. The measured

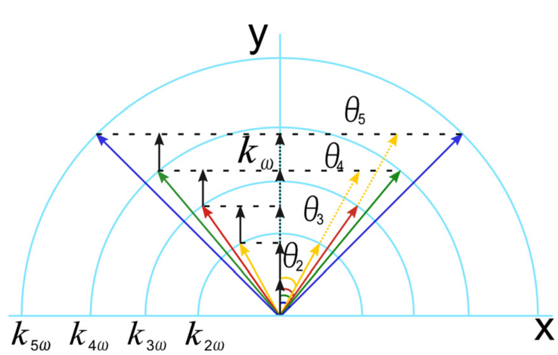

FIG. 2. The schematic diagram displaying the phase-matching geometries of multistep cascaded Cherenkov multiple-harmonic generation. The semicircles from inner side to outer side are corresponding to the second-, third-, fourth-, and fifth-harmonic wave vectors, respectively.

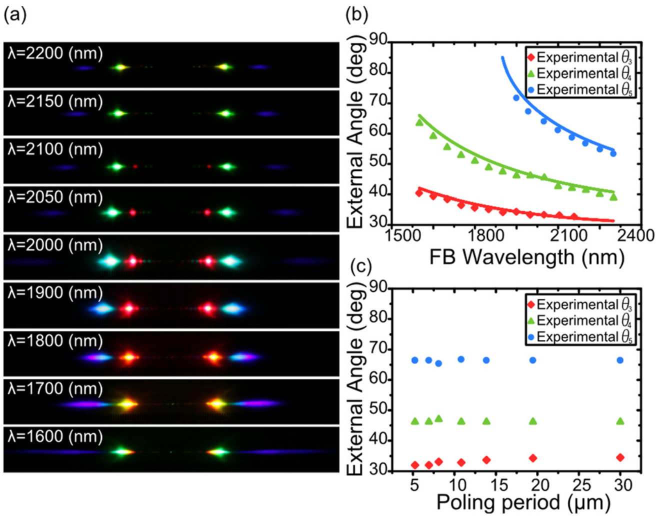

FIG. 3. (a) Evolution of recorded patterns varying with the fundamental wavelength. Plot of the experimental (signs) and calculated values (solid lines) of (b) the external radiate angles of Cherenkov third, fourth, and fifth harmonic generation and (c) Cherenkov high-order harmonic generation versus grating period (5.24, 6.92, 8.1, 10.83, 13.86, 19.47, and 30 μm).

intensity of Cherenkov fifth harmonic demonstrates position-dependence of the incident beam when the sample translates along X direction. According to Fig. 4(a), we can obtain the variation cycle is 15 μm, which is in coincidence with the period of domain wall. Namely, Cherenkov fifth harmonic only occurs when tightly focused fundamental beam overlaps the domain wall region, and the diagram of the process is schematically shown in Fig. 4(b).

As of the intensity of the converted harmonic wave, it is straightforward to derive that the intensity of ith order harmonic wave obeys the ith power law relation with that of input, i.e., $I_{i\omega} \propto d_{eff}^{2(i-1)} I_{\omega}^{i}$ (i = 2, 3, 4, and 5). We measured the conversion efficiency of the multiple harmonics as a function of the power of the fundamental beam. As shown in Figs. 4(c) and 4(d), the output power of the fourth and fifth harmonic grows gradually with the input power, and the curves are well fitted to be quartic and quintic relationship, which is consistent with the theoretical expectations. While the incident beam is e-polarized and the injecting power is 30 mW in our experiments, we get the power of the generated third, fourth, and fifth harmonic to be 2 mw, 0.8 mw, and 0.06 mw in one channel, which represents the conversion efficiency of about 6.7%, 2.7%, and 0.2%, respectively. With optimizing the emission properties of the harmonic signals, for instance, varying angle[20] or adjusting the position of fundamental beam,[9] one can promisingly improve the output power of the harmonics.

In conclusion, we propose a scheme for efficient high-order Cherenkov harmonic generation and experimentally observed the high-order harmonic generation (up to fifth order). We studied the unifying explanation of the phase matching geometry and also the angle-, power-, and position-dependence of the fundamental beam for the multiple harmonics. The relatively high efficiency of each order harmonic enables further purposeful manipulation, for example, Fourier synthesis of optical waveforms and emergence of attosecond pulses with wavelengths in the extreme ultraviolet.[17] Hence, this type of nonlinear responses provide a practical approach to generate multiple higher-order frequency simply via tuning the input wavelength in a single quadratic medium and thus will find widespread use in optical areas.

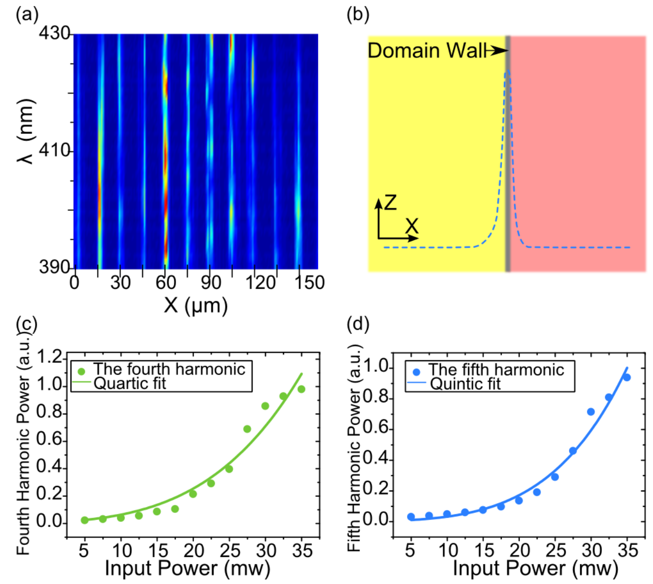

FIG. 4. (a) Measured intensity of the Cherenkov fifth harmonic vs. the lateral position of fundamental beam while the beam diameter is 5 μm. (b) Schematic of the fifth harmonic profile when tightly focused fundamental beam overlaps with a domain wall. Dependence of Cerenkov fourth (c) and fifth (d) harmonic power on input power at 2050 nm fundamental wavelength. Solid line represents quartic and quintic fit, while signs represent experimental data.

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