Broadband enhancement of Čerenkov second harmonic generation in a sunflower spiral nonlinear photonic crystal

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Abstract: We present an experimental study on the Čerenkov second harmonic emission in a novel sunflower spiral array of ferroelectric domains in LiNbO3 crystal. The spiral patterns offer a diffusive, circularly symmetric distribution of reciprocal lattice vectors, thereby enabling enhanced emission of the Čerenkov beam in a broad spectral range. Instead of the traditional electric field poling, the sunflower spiral patterns are fabricated here by using our pioneering method of ferroelectric domain engineering with ultrafast light. This all-optical method gives access to high quality domain structures with short periods, which is beneficial for efficient Čerenkov harmonic generation.

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References and links
**Introduction**

The nonlinear Čerenkov radiation refers to an interesting type of frequency conversion processes where the emission direction of the new waves are determined solely by the fulfillment of the longitudinal phase matching condition [1–3]. Taking the simplest process of second harmonic generation (SHG) as an example, the phase matching condition is written as $k_2 \cos \theta_c - 2k_1 = 0$, with $k_1, k_2$ denoting the magnitudes of the wave vectors of the fundamental and the second harmonic waves, respectively, and $\theta_c$ is the Čerenkov angle [Fig. 1(a)]. Efficiency of the Čerenkov harmonic emission critically depends on the spatial profile of quadratic nonlinearity ($\chi^{(2)}$) variation [4] and the spatial field distribution of interacting waves [5]. For instance, with a Gaussian-profile fundamental beam propagating in a two-dimensional nonlinear photonic crystal (with a circular motif of modulated $\chi^{(2)}$), a conical Čerenkov second harmonic is usually observed in far field [Fig. 1(b)] [6]. The nonlinear Čerenkov interaction has been shown to be attractive for applications in nonlinear optical microscopy [7,8], ultrashort pulses characterisation [9], high harmonic generations [10], and ferroelectric kinetics analysis [11].

![Phase matching diagram of the Čerenkov-type second harmonic generation.](image)

From the point of view of practical applications it is also desirable to enhance the efficiency of...
2. Structure design and fabrication

The sunflower patterns, also known as the golden-angle spirals, have been widely found in the arrangements of seeds, leaves, and stalks in pine cones, artichokes, nautilus, sunflowers and so on [19]. Such structures allow the most even distributions of seeds in the sunflower heads without overlapping. They are obtained by a simple generation rule, expressed in polar coordinates, first proposed by Vogel in order to approximate the seeds distribution in sunflower heads [20]

\[ r = b \sqrt{q}, \quad \theta = qa, \quad (1) \]

where \( b \) is a constant scaling factor, \( q = 0, 1, 2, \ldots \) are integers, and \( \alpha \approx 137.508^\circ \) is the so called golden angle that can be expressed as \( \alpha = 360^\circ / \varphi^2 \), where \( \varphi = (1 + \sqrt{5}) / 2 \approx 1.618 \) is the golden number. Fig. 2(a) shows an example of the sunflower structures with \( b = 1.45 \, \mu m \) and \( q \) being taken up to 2000. The golden spiral structure can be decomposed into an equal number of clockwise and counter-clockwise spiral families originating from its center. Additionally, since the golden angle is an irrationalumber, the structure lacks both translational and rotational symmetries. Accordingly, its spatial Fourier spectrum does not exhibit well-defined Bragg peaks, but show diffuse circular rings, as shown in Fig. 2(b). For comparison, Figs. 2(c) and 2(d) depict a periodic
circular domain structure and its Fourier spectrum, respectively. The narrow discrete features in the latter are clearly visible.

Fig. 2. (a) The sunflower spiral lattice created with $b=1.45 \, \mu m$ and $q=0, 1, \ldots, 2000$ in Eq. (1). (b) The Fourier space of the sunflower pattern, in which there are diffuse and rotational symmetric reciprocal lattice vectors for efficient nonlinear Čerenkov radiation. (c) The structure and (d) Fourier space of a periodic annular structure for comparison.

We fabricated the sunflower spiral nonlinear photonic crystals with the femtosecond laser poling technique [21,22]. The wafer of 400 $\mu m$ thick congruent LiNbO$_3$ was mounted on a high-precision translation stage that can move automatically in three dimensions. The laser beam with wavelength of 800 nm, pulse width of 180 fs, repetition rate of 80 MHz and pulse energy of about 5 nJ was delivered by MIRA 900 (Coherent). The beam was focused onto the crystal by a 40x (NA = 0.65) objective with beam waist of about 0.5 $\mu m$. For every X, Y position the sample was translated along the Z-direction such that focus of the beam moved from the crystal’s -Z to the +Z surface with an average velocity of 10 $\mu m$ s$^{-1}$ to complete a domain inversion. A shutter was used to block the beam when the crystal was positioned to the next domain inversion region. To avoid deleterious effect of spherical aberrations we restricted the domain reversal to relatively thin subsurface region of the crystal [23].

Fig. 3. (a-c) Three examples of the fabricated sunflower spiral nonlinear photonic structure in a congruent LiNbO$_3$ crystal, visualised by the Čerenkov second harmonic microscopy [7]. The structure parameters are as follows: structure (a): scaling factor $b=5 \, \mu m$, $q=0, 1, \ldots, 200$; (b): scaling factor $b=2 \, \mu m$, $q=0, 1, \ldots, 625$; (c): scaling factor $b=1.45 \, \mu m$, $q=0, 1, \ldots, 3086$. (d) 3D visualisation of the 30$\mu m$×30$\mu m$ section of the structure from graph (b). Radius of the inverted domains $r \approx 1 \, \mu m$ in all three structures.

Figure 3 depicts three fabricated sunflower domain patterns, obtained by the Čerenkov second-harmonic microscopy [7,8,24]. The average radius of the inverted domains is $r \approx 1 \, \mu m$ and their length (along Z) is about 25 $\mu m$. The structure shown in Fig. 3(c) with the smallest average period was used in the described below experiments. In this structure the distance between the neighboring inverted domains varies from 5 $\mu m$ to 2.5 $\mu m$. While all structures were relatively shallow, our further experimental results indicate that such a depth is already long enough to demonstrate their benefits, as the Čerenkov signal grows quickly with the propagation distance.

3. Čerenkov second harmonic generation: results and discussions

To demonstrate the nonlinear Čerenkov emission in the sunflower spiral structure we illuminated the sample [Fig. 2(c)] with a femtosecond laser beam with the wavelength tuneable from 1.2 to
1.6 µm (Coherent Chameleon Compact OPO). The normally incident beam (300 mW of power) was focused onto the sample by a NA=0.1 objective lens. Its diameter at the structure was 80 µm which was slightly less than the size of the whole sunflower spiral pattern. The emitted second harmonic was projected onto a screen and recorded by a CCD camera. The observed far-field harmonic pattern is shown in Fig. 4, where two rings are clearly visible. Here the external ring is the Čerenkov second harmonic (complying with the longitudinal phase matching condition) and the internal one is the first-order Raman Nath second harmonic that satisfies only the transverse phase matching condition, i.e. \( k_2 \sin \theta = G \), with \( \theta \) being the internal emission angle of the Raman-Nath second harmonic [25, 26]. Both rings exhibit asymmetric azimuthal intensity distribution, which results from the dependence of the effective nonlinear coefficient on both, polarization of the interacting beams and the emission direction within the crystal [25].

Because of short propagation length along Z-direction the overall power of the Čerenkov emission was rather week, around 1 µW. However, it is still evident that the Čerenkov emission peaks around roughly 1.3 µm of the fundamental beam. This is even better visible in Fig. 4(b) which depicts the wavelength tuning curve of the Čerenkov harmonic generation, i.e., the power of the emitted harmonic vs. fundamental wavelength, for constant input average power of 300 mW (corresponding to peak power of 25 kW). It is clear that the Čerenkov emission occurs in the whole wavelength range (1200–1600 nm) and that across 1220–1450 nm is significantly stronger. This broadband enhancement effect agrees with the diffusive Fourier spectrum of the sunflower spiral pattern. As we discussed above, to enhance the Čerenkov interaction, the transverse phase mismatch should be compensated by the appropriate reciprocal lattice vectors. Therefore, the strength of the Čerenkov signal is proportional to the square of the Fourier coefficient of the involved reciprocal lattice vectors. Using the material dispersion data of the congruently grown LiNbO₃ crystal [19], we find that the reciprocal lattice vectors in magnitudes 3.5 ≤ |G| ≤ 5.9 µm⁻¹ are mainly responsible for the transverse phase matching at the wavelengths 1200 –1600 nm, and those between 4.2 and 5.7 µm⁻¹ correspond to the stronger emissions. We plot the square of the modulus of the the Fourier coefficient of the sunflower pattern in Fig. 4(b) with solid line. A good agreement is obtained between the experiment and theory. The lower than expected experimentally observed contrast is mainly caused by the imperfection of the produced domain structures. It is known that in any poling processes the random period errors are unavoidable. Such random deviations from the designed period and the varied radius of individual inverted domains will broaden the wavelength tuning response and weaken its strength.

The resonant -like enhancement of the Čerenkov emission can be better understood by looking at the wavelength dependence of the emission angles of the Čerenkov and Raman-Nath second harmonics. This is depicted in Fig. 5(a). The experimental results are shown as points while
solid lines represent theoretical predictions. Both nonlinear processes exhibit opposite behaviour when input wavelength is varied. The angle of Raman Nath signal increases, but that of Čerenkov emission decreases. It is precisely at the wavelength when angular emission of both signals coincides when the resonant enhancement of the Čerenkov happens. For this wavelength the second harmonic generation process is phase matched in longitudinal and transverse directions, leading to the Bragg nonlinear emission. As Fig. 5(a) shows the Bragg emission takes place for $\lambda_0=2.24 \, \mu m$ which was unavailable in our experiment. Therefore the observed enhancement of the emission was caused by the second order Raman-Nath process. As it is evident in Fig. 5(a), the curve representing this process intersects with Čerenkov curve at 1.3 $\mu m$, which coincides with the experimental observation. However, since the second order effective nonlinear coefficient is three times less than the first order, the efficiency of the process is significantly (≈10 times) weaker. This is evident in Fig. 5(b) which shows the square of the modulus of the the Fourier coefficient in our sunflower spiral structure for broad spectral range of the fundamental beam. The first (highest) peak is responsible for the first order Raman-Nath SH emission. For input wavelengths available in our experiment, only the second (much weaker) peak contributed towards emission enhancement.

4. Conclusion

We employed the femtosecond laser poling technique to fabricate two dimensional sunflower spiral nonlinear photonic crystals in LiNbO$_3$ crystal. Taking advantage of the resulting diffusive Fourier space of the structure, we demonstrated enhanced broadband Čerenkov second harmonic emission in the fundamental wavelength range of 1220–1450 nm. Note that the sunflower spiral nonlinear photonic crystals can be used for many other devices such as the quasi-phase matched frequency convertors with broad angle and temperature acceptances. Our study indicates the femtosecond laser poling constitutes a powerful technique for fabricating nonlinear photonic crystals of complex spatial nonlinearity modulation, that cannot be easily accessed by traditional methods, thereby opening up the possibility to realize a wide range of new optical devices.

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