Multiple shape light sources generated in LiNbO$_3$ nonlinear photonic crystals with Sierpinski fractal superlattices

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Abstract

LiNbO$_3$ nonlinear photonic crystals with two kinds of Sierpinski fractal superlattice structures, in which their domain walls are circle and square cylindrical surfaces for the second order, respectively, are successfully fabricated. Quasi-phase matching and nonlinear Čerenkov harmonics under pulse laser beams are measured. The spot- and line-shaped light sources are realized by quasi-phase matching harmonics. Ten kinds of spot-shaped outputs can be accomplished, in which the quasi-phase matching harmonic conversion efficiency for the fundamental wavelength 1.352 µm reaches 27%. A green line-shaped light source with expansion angle of about 3° is observed, which originates from the coexistence of imperfect collinear and non-collinear quasi-phase matching processes. In addition, the ring-shaped light sources of continuous wavebands from 425 nm to 675 nm are obtained by nonlinear Čerenkov harmonics. The optical properties of quasi-phase matching and Čerenkov harmonics in two kinds of crystals are highly similar to each other.

Keywords: nonlinear optical crystals, frequency conversion, quasi-phase matching, nonlinear Čerenkov harmonics

1. Introduction

Nonlinear photonic crystal (NPC) is a kind of processed ferroelectric crystal, in which partial domains are reversed by applying high voltage electric field. Over the past few decades, much attention has been focused on several aspects of NPCs; for example, the type of ferroelectric crystals, the structure of domain distribution and their optical properties [1–18]. Traditionally, researchers select LiTaO$_3$ (LT), LiNbO$_3$ (LN), KTP or strontium barium niobate (SBN), etc, as the crystal raw materials. The structures mainly relate to the one- or two-dimensional, periodic, quasi-periodic, random and super quasi-phase matching (QPM) superlattice structures. The unique optical properties include QPM harmonics, Talbot effects, Čerenkov radiation (CR) and so on.

LiTaO$_3$ nonlinear photonic crystal with a one-dimensional Fibonacci series was fabricated; from this, the red, green and blue visible lights were obtained. Recently, a new scheme was presented to directly produce fundamental, second- and third-harmonic three-color continuous-variable entangled beams [1, 2]. In NPC of strontium tetraborate (SBO), non-collinear random quasi-phase-matched frequency doubling was used for background-free autocorrelation measurements of ultrashort femtosecond pulses in a wide spectral range, and conversion of the femtosecond supercontinuum to the ultraviolet range from 260 to 305 nm was obtained [4, 5]. The conical emission of Čerenkov-type second- and third-
harmonic generations were investigated experimentally and theoretically in NPCs of 2D-χ(2) distribution [7, 11]. In addition, nonlinear Čerenkov radiation was realized experimentally in an anomalous dispersive medium, which breaks the minimum speed of radiation, i.e. the realization of the Smith–Purcell effect [12]. Moreover, a second-harmonic vortex beam generated from a Gaussian pump beam was demonstrated in twisted nonlinear photonic crystals, and a new configuration for engineering various orbital angular momentum (OAM) was studied [13]. Recently, several bandwidths of non-collinear phase-matching second harmonic generation (SHG), with angle, temperature and wavelength about 0.51°, 4.1 °C and 6 nm, respectively, were generated by sum-frequency of the incident and reflected wave on the inner surface of a MgO:LiNbO3 crystal [15]. A highly integrated two-dimensional aperiodic LN NPC was built to demonstrate a compact, high-peak-power intracavity sum-frequency generator (ISFG) radiating at orange 593.5 nm [16].

LiNbO3 ferroelectric crystal has unique characters, such as the fact that it is not liable to deliquesce and its wide transparent range (from 0.3 μm to 5.2 μm), so we select LiNbO3 as a candidate for NPC. Fractal structure has the unique characteristic of self-similarity, which means that the original pattern can be repeated after exaggerating its part integer times. Then in the reciprocal space of fractal structure, the reciprocal vectors (RVs) have the relationship of integer times each other. We choose the two-dimensional LiNbO3 nonlinear photonic crystals with Sierpinski fractal superlattice structures as the experimental samples. Their typical optical properties of QPM and CR harmonics are described in detail. Multiple light sources with single spot, series of spots, line or ring shape can be realized in two-dimensional LiNbO3 NPCs with one kind of Sierpinski superlattice structure. Differently shaped light sources may be needed in nonlinear optics, waveguide detection, domain detection, nano- or micro-structure formation and industrial laser fabrication.

2. Sample fabrication

The conventional Sierpinski fractal structure can be formed by the following steps. Firstly, divide a big square into nine small squares uniformly; secondly, delete the center small square to obtain a one-order Sierpinski fractal structure; thirdly, divide each of the eight left small squares into nine small squares uniformly; fourthly, delete the center smaller square to obtain a two-order Sierpinski fractal structure. Then by repeating these steps, one can get an ideal Sierpinski fractal structure with different orders. Considering the fabrication of the mask and nonlinear photonic crystal, we adopt the four-order Sierpinski fractal structure in our experiments. In nearly all two-dimensional NPCs, the reverse domain walls show up as circle cylindrical surfaces. In order to explore the effects of different domain walls, some modifications are made in the conventional structure. We substitute some of the deleted circles for the deleted squares, where the side lengths of the squares are the same to the diameters of the circles. Then, we set the four-order Sierpinski fractal structure as a unit and repeat the unit to two-dimensional plane, in which the interval between the neighbor units is the distance between the above two neighbor minimum circles (i.e. the fourth order), i.e. 13.64 μm. The ratio of the distance between the two neighbor maximum and minimum circles is 27.

We successfully fabricate the nonlinear photonic crystals by applying the external high-voltage electric fields to z-cut LiNbO3 crystals. Their length, width and thickness are about 8, 8, and 0.5 mm, respectively. In figure 1, +z surface graphs of the LiNbO3 NPCs with two kinds of four-order Sierpinski fractal structures are shown. Figure 1(a) shows that all of the deleted parts are circles, and figure 1(b) shows that the deleted parts are circles except the second order. It is clear that the poled patterns are uniform in the whole plane. In figure 1(a), the diameters of the four-order circles are 107, 33, 12, and 5 μm, respectively. In figure 1(b), the diameters of the three-order circles are 110, 12, and 5 μm, and the side length of the square is about 37 μm.

![Figure 1](image_url)
The diffraction phenomena of these two kinds of Sierpinski fractal structures are experimentally investigated under He–Ne laser beams. The beams are incident perpendicularly to the two-dimensional fractal structure planes, i.e. parallel to the z-axis. Figures 2(a) and (b) display their diffraction patterns. Firstly, the above-described self-similarity is clear. There are always two weaker diffraction spots between two neighboring stronger diffraction spots. In other words, it has many more reciprocal vectors than the two-dimensional square periodic structures in the reciprocal space [6]. In addition, diffraction spots do not concentrate along the vertical and horizontal directions like two-dimensional H-shaped fractal structures [19]. This is crucial for the following optical properties. Secondly, it can be drawn that the positions of the strong diffraction spots in the two different structures are the same as each other, other than some weak diffraction spots shown in the background. This can be attributed to the fact that there are plenty of same minimum circles in the real spaces of the two different structures. In addition, it is evident that a circle envelope is shown in the first structure, in which all of the reverse domains in real space are circles. Correspondingly, a square envelope is clearly shown in the second structure, which means that square components affect the diffraction intensity in reciprocal space. Our results agree very well with the previous results [20].

3. Experiments

As is well known, harmonics can be obtained by quasi-phase matching technique in nonlinear photonic crystal [21]. For example, for a QPM second-harmonic generation, the laws of energy and momentum conservation need to be satisfied simultaneously. Energy conservation means that $2\omega_1 = \omega_2$, where $\omega_1$ and $\omega_2$ are the frequencies of input fundamental and output second-harmonic laser beams, respectively. Momentum conservation means that $2k_1 + G = k_2$, where $k_1$ and $k_2$ are the wave vectors of input fundamental and generated second-harmonic laser beams, respectively, and $G$ is the reciprocal vector. If the wave vectors of input fundamental laser beams and the adopted reciprocal vectors distribute along the same direction, it is termed a collinear quasi-phase matching (CQPM) process. On the other hand, if there is a certain angle between input wave vectors and reciprocal vectors, it is termed a non-collinear quasi-phase matching (NCQPM) process. In the following text, section 3.1 mainly relates to CQPM, and section 3.2 mainly relates to NCQPM. In these two sections, the fundamental laser beams are perpendicularly incident to the crystal polished-side surfaces along the x- or y-axis of the crystal, and their polarization is parallel to the z-axis of the crystal, i.e. nonlinear coefficient $d_{33}$ is adopted. It is noted that all of our experiments are done at room temperature.

Thus, we investigate the Čerenkov radiation in nonlinear photonic crystal with Sierpinski fractal structures. Originally, the concept of Čerenkov radiation relates to charged particle with high speed. When the particle travels faster than light in the medium, the conical radiation diverging from the traveling direction of the particle will generate what is called Čerenkov radiation. Later, this concept is introduced to the field of optics, in which the input light is used as a substitute for the original particle, and CR is the harmonic of input light [7, 17, 18]. The Čerenkov radiation angle inside the crystal is represented by $\theta = \arccos (v_2/v_1)$, in which $v_2$ and $v_1$ mean the phase velocity of the Čerenkov harmonic radiation and the incident light, respectively. In the following, section 3.3 relates to Čerenkov radiation. In this section, the crystals are laid vertically, so that the fundamental laser beams are perpendicularly incident to the crystal polished surfaces along the z-axis of the crystal, in which nonlinear coefficients $d_{32}$ and $d_{22}$ are adopted.

3.1. Spot-shaped light sources

The input fundamental laser beams are generated from 355 nm pumped optical parametric oscillator (OPO). The pulse duration and the repetition frequency are about 9 ns and 10 Hz, respectively. The input laser beams are linearly
polarized propagation. The focal length of the lens is 20 cm. Sample 1 is a piece of LiNbO$_3$ NPC with Sierpinski fractal superlattice structure, in which all four orders are circles, as shown in figure 1(a). Sample 2 is a piece of NPC, in which three orders are circles, as shown in figure 1(b).

Firstly, we study the CQPM second-harmonic processes in sample 1. The effective red spot-shaped harmonic at wavelength 675.05 nm is generated in figure 3(a). Figure 3(b) shows the CQPM SHG conversion efficiency versus the input power under the input fundamental wavelength 1350.1 nm. The higher the input power is, the higher the output power is. When the input power is 3.73 mW, the output power of the harmonic is 328 $\mu$W. Considering the transmittance of the filter and reflection of the polished surfaces, the conversion efficiency is about 13%. It can be seen that the conversion efficiency tends to be stable, which originates from the saturation. In addition, in order to detect the monochromaticity of the generated spot-shaped light sources, we measure the conversion efficiencies at different harmonic wavelengths around 675.05 nm, as shown in figure 3(c). The circle dots represent the experimental values, and the line is the fit result. The conversion efficiency decreases sharply when the wavelength diverges from 675.05 nm. The bandwidth is less than 1 nm. It can be concluded that the monochromaticity of the harmonic is very good, so it is perfect as a spot-shaped light source.

It is noted that pairs of blue and red harmonic spots at symmetric positions about the input laser direction are observed simultaneously in figure 3(a). When the distance between the crystal and screen is 30 cm, the interval between a pair of red second harmonics is 5.4 cm. And the interval between a pair of blue third-harmonics is 8.4 cm. Series of symmetric spot-shaped light sources are induced by NCQPM harmonic processes [22].

Comparatively, we investigate the optical properties of sample 2 under similar conditions. At input fundamental wavelength 1351 nm, a red second-harmonic spot is obtained at the center by CQPM processes in figure 4(a). Figure 4(b) shows the conversion efficiency of 675.5 harmonics as a function of input fundamental laser power. The conversion efficiency increases gradually by increasing the input power of laser beams. It can reach 27.4% when input power is 2.2 mW. In addition, figure 4(c) shows that the conversion efficiency varies with the generated second-harmonic wavelength. The solid dots and line are experimental and theoretical results, respectively. The conversion efficiency can reach a maximum of about 26% at harmonic wavelength 675.27 nm. When the conversion efficiency is half maximum, the variable harmonic wavelength band is about 0.5 nm. It is concluded that the spot-shaped light source has a good quality with narrow bandwidth.

At symmetric positions around the center of 675.5 nm red harmonic spot, the distances between each pair of symmetric harmonic spots are 3.6, 5.5, 8.6 and 9.6 cm, respectively, in figure 4(a). The distances for the central two pairs are approximately equal to those of sample 1. Based on the similar harmonic wavelengths and symmetric NCQPM outputs, it can be concluded that two kinds of Sierpinski fractal structures have similar reciprocal vectors but different intensities of some diffraction spots.
We compare the diffraction patterns of the two samples shown in figure 2 with those of the two-dimensional square periodic superlattice structure, in which its period is 13.64 μm. It can be seen that the distance represented by black arrows in figure 2 is equal to the basic reciprocal lattice vector \( a = 2\pi / 13.64 \), i.e. \( a = 0.461 \) μm\(^{-1}\). According to momentum conservation in the QPM process and the dispersion equation of LiNbO\(_3\) [23], one can know this reciprocal vector is theoretically fit for the generation of CQPM harmonics of the input fundamental wavelength 1351 nm, which agrees very well with the above experimental values, i.e. 1350.1 nm in sample 1 and 1351 nm in sample 2.

Further, we measure the generation of the spot-shaped light sources in sample 1 when the incident fundamental laser beams are picosecond pulses, as seen in figure 5. The input laser beams are generated from 355 nm pumped optical parametric amplification (OPA). The generated wavelengths can be tuned in the range of 710 and 2300 nm. The pulse duration and the repetition frequency are 21 ps and 1 kHz, respectively. The focus of the lens in the experimental setup is 20 cm. As shown in figures 5(a)–(g), seven colors of spot-shaped harmonic light outputs are observed under different second-harmonic wavelengths, including 592, 577.5, 564.5, 552.5, 532, 506.5 and 502 nm. The reciprocal vectors adopted in generating the harmonic at 532 nm is double that of the harmonic at 532 nm, i.e. \( 2a = 0.922 \) μm\(^{-1}\).

In figure 2, we select the above basic reciprocal vector \( a \) between two neighbouring strong diffraction spot as the standard length. There are some weak diffraction spots between these diffraction spots, in which the distances between neighbouring weak diffraction spots can be \( a/3 \) or \( a/9 \) due to the self-similarity of fractal structures. This displays exactly the advantage of the Sierpinski fractal superlattices over the periodic superlattices [6]. Plenty of diffraction spots will provide various reciprocal vectors in the reciprocal spaces, as shown in table 1. In figure 6, the collinear QPM second-harmonic processes are illustrated at fundamental wavelengths 1129 nm and 1064 nm, in which the reciprocal vectors \( (1 + 2/3)a \) and \( 2a \) are adopted. By using the QPM concept, the reciprocal vectors and the corresponding input fundamental wavelengths are calculated theoretically. For the above experimental CQPM harmonics 592, 577.5, 564.5, 552.5, 532, 506.5 and 502 nm, the fundamental wavelengths are 1184, 1155, 1129, 1105, 1064, 1013, and 1004 nm, respectively. The adopted reciprocal vectors are the linear combinations of \( a, a/3 \) and \( a/9 \), i.e. \( (1 + 1/3 + 1/9)a \), \( (1 + 1/3 + 2/9)a \), \( (1 + 2/3 + 1/9)a \), \( 2a \), \( (2 + 1/3)a \), and \( (2 + 1/3 + 1/9)a \), respectively. Correspondingly, the theoretical fundamental wavelengths are 1184.9, 1155.7, 1129.5, 1105.9, 1064.8, 1014.6 and 1000 nm. These results agree very well with the experimental results. The difference is about 1 nm,
except in the case of the experimental fundamental wavelength 1004 nm, where it is 4 nm.

Additionally, in figure 5(h), a series of NCQPM spot-shaped light sources can be generated when the input laser is 992 nm; in this series, four spots in the central section are of approximately the same intensity. The second- and third-harmonic generations 866 nm (infrared) and 577 nm (yellow) at input wavelength 1731 nm can be also detected, as shown in figure 5(i). By calculation, we can know that harmonics 866 nm and 577 nm originate from the direct CQPM harmonic processes. The reciprocal vectors used are 0.264 μm⁻¹ and 0.922 μm⁻¹, respectively, and most probably result from the reciprocal vector combinations \( \frac{1}{3} + \frac{2}{9} a \) (i.e. 0.256 μm⁻¹) and 2a. This three-color (fundamental, second- and third-harmonic waves) coexistence at the same position is beneficial to the quantum entanglement [2, 24]. Moreover, pairs of light spots at wavelengths 659 nm (red) and 340 nm (ultraviolet) can be detected at the symmetric positions, which could be attributed to NCQPM nonlinear processes. All of these phenomena can also be obtained in sample 2.

### 3.2. Line-shaped light sources

For sample 2, by using the reciprocal vector 2a, we can realize the CQPM second-harmonic outputs of input fundamental laser 1064 nm under nanosecond pulses. When the input fundamental wavelength decreases to 1063.34 nm, the line-shaped harmonics are observed, as shown in figure 7(a). When the input power is 3.27 mW, the second-harmonic power is 365 μW. The length of the line-shaped light source is about 1.1 cm when the distance between the NPC and the screen is 30 cm. This means that the divergence angles along the input laser beams in the external and internal sections of the NPC are 2.1° and 0.9°, respectively. With the input laser wavelength further decreasing to 1063.06 nm, the length of the line-shaped light output becomes 1.5 cm in figure 7(b). The external angle of the whole line-shaped output can reach 2.86°. In order to confirm the cause of line-shaped light outputs, we continuously tune the fundamental wavelength. When the wavelength is 1062.7 nm, the spot- and line-shaped lights coexist, as shown in figure 7(c). When the wavelength decreases by 0.79 nm, the two symmetric strong spot outputs become clear, as in figure 7(d), and obviously originate from NCQPM second-harmonics. Then the distance between two spot-shaped lights becomes 2.3 cm and the external angle is about 4.38°. It has been shown that during the changing from perfect CQPM to perfect NCQPM harmonic processes, the adopted reciprocal vectors prefer the closest distributed transversely reciprocal vectors, in which the transversely reciprocal vectors are perpendicular to the input laser beam [22]. By calculation, the adopted transversely reciprocal vectors in perfect NCQPM harmonic generation in figure 7(d) are about 0.4533 μm⁻¹, which is approximately equal to the basic reciprocal vector \( a = 0.461 \text{ μm}^{-1} \).

In the quasi-phase matching technique, the QPM process by using the basic reciprocal vector \( a \) is termed a first-order CQPM process; for example, at the fundamental wavelength of about 1.3501 or 1.351 μm in figure 7(a) or figure 4(a). The QPM process using the combined reciprocal vectors 2a is termed a second-order CQPM process; for example, at the fundamental wavelength 1.064 μm in figure 5(e). It is noted that when the wavelength decreases from the above wavelength by 1.3501 or 1.351 μm, there is no obvious line-shaped light source generation as that from wavelength 1.064 μm by the same transverse reciprocal vector, whose direction is orthogonal to the input fundamental laser and whose length is \( a \). We think this can be explained as follows. When input fundamental laser wavelengths decrease from the wavelength suitable for CQPM processes, NCQPM processes, in which the closest effective transverse reciprocal vectors are adopted, will occur first. For the same transverse reciprocal vectors in the NCQPM process, because of the larger divergence angles of the harmonics from input fundamental beams on the basis of longer fundamental wavelengths or shorter wave vectors, a series of spot-shaped light outputs are more likely to generate. Correspondingly, because of the smaller divergence angle of the harmonics from input fundamental beams on the basis of shorter fundamental wavelengths or longer wave vectors, line-shaped light outputs are more likely to generate. For comparison, two NCQPM processes are illustrated together in figure 8. For the internal divergence angle, \( \theta = \frac{Gd}{2k_1} \), in which \( G = a = 0.461 \text{ μm}^{-1} \). For NCQPM harmonics of input
wavelength about 1.3505 μm, \(2k_i = 19.9 \mu m^{-1}\), then \(\theta_1 = 1.32^\circ\). For NCQPM harmonics of input wavelength about 1.064 μm, \(2k_i = 25.5 \mu m^{-1}\), then \(\theta_2 = 1.03^\circ\). Therefore, for the same transverse reciprocal vector, the longer is the input fundamental wave vector, and the smaller is the diverge angle, making the line-shaped light outputs easier to realize by the coexistence of imperfect CQPM and NCQPM processes. In other words, the line-shaped light source originates from the process varying from CQPM to NCQPM.

Moreover, this line-shaped output may be partially attributed to an abundance of weak diffraction spots (shown in the ellipses in figure 2(b)) between the neighboring strong diffraction spots (the distance is \(a\)) along the direction perpendicular to the input laser beam. These weak diffraction spots derive from the self-similarity of fractal superlattice structures and cannot exist in the periodic structures. It is noted that the intensity along the whole line-shaped light pattern is approximately the same, so it can be used as a good light source in micro/nano optics. All of these above results may provide a new way to generate line-shaped light sources.

Similarly, the line-shaped light sources can also be realized in sample 1. However, the obvious line-shaped light output under picosecond laser beams has not yet been observed. Therefore, the output harmonics may be affected greatly by pulse duration, beam divergence angle or peak power density.

### 3.3. Ring-shaped light sources

Besides QPM harmonics, we investigate Čerenkov second-harmonic radiation in two-dimensional LiNbO₃ NPCs with Sierpinski fractal superlattices. The LiNbO₃ NPCs, in which three-order structures are circles with diameter 109, 11 and 3.5 μm, respectively, and the side length of the second-order square is about 35 μm, are termed sample 3. The LiNbO₃

| Reciprocal Vector Combinations | Reciprocal Vector Values (μm⁻¹) | Theoretically | Experimentally |
|--------------------------------|---------------------------------|---------------|---------------|
| \((1 + 1/3 + 1/9)a\)          | 0.6654                          | 1184.9        | 1184          |
| \((1 + 1/3 + 2/9)a\)          | 0.7166                          | 1155.7        | 1155          |
| \((1 + 2/3)a\)                | 0.7677                          | 1129.5        | 1129          |
| \((1 + 2/3 + 1/9)a\)          | 0.8189                          | 1105.9        | 1105          |
| \(2a\)                        | 0.9212                          | 1064.8        | 1064          |
| \((2 + 1/3)a\)                | 1.0748                          | 1014.6        | 1013          |
| \((2 + 1/3 + 1/9)a\)          | 1.126                           | 1000          | 1004          |
 NPCs, in which four-order structures are circles with diameters 110, 34, 12.5, and 5.5 µm, respectively, are termed sample 4. The $xoy$ surfaces of the NPCs in samples 3 and 4 are polished. The input fundamental laser beams originate from the above picosecond OPA. In the following experiments, the laser beams are incident along the $z$-axis of the NPCs. The experimental setup is illustrated in figure 9. The input laser beams are loosely focused on the LiNbO$_3$ NPC by a lens with focal length of about 20 cm. The distance between NPC and screen is denoted by $d$, and $\beta$ is the external conical angle of Čerenkov second-harmonic radiation. The ring-shaped second harmonics based on conical CR can always be observed on the screen while tuning the wavelength from 850 to 1350 nm. In the following, we will describe them in detail.

Firstly, the Čerenkov radiations of sample 3 are measured in the following. The distance $d$ is 2.6 cm. The experimental image on the screen is recorded with a common camera. Figures 10(a)–(g) shows the Čerenkov second-harmonic radiations at wavelengths 500, 525, 550, 575, 600, 625 and 650 nm under multiple input laser beams, between which the wavelength interval is 25 nm. In each image, there are six arcs forming a ring-shaped light output at the following positions, which diverge the angles of $0^\circ$, $30^\circ$ and $60^\circ$ from the horizontal line. This sixfold output can be attributed to the hexagonal shape of the reverse domains in LN NPC [6, 22]. It can be seen that the ring-shaped Čerenkov radiations are two concentric ring structures (external and internal), in which two rings are close to each other. Then the average diameter of the radiation rings is obtained by measuring the distance between the middle points of two rings on the left side and on the right side along the horizontal line. The average conical angle as a function of input fundamental wavelength is given in figure 11. The squares correspond to the externally measured values outside the NPC. It can be seen that the conical angles of Čerenkov radiation decrease almost linearly with the input wavelengths increasing. Noted that the Čerenkov second-harmonic radiation pattern will rotate simultaneously when the NPC is rotated. For instance, the ring-shaped radiation pattern will rotate $180^\circ$ when the LiNbO$_3$ NPC is rotated $180^\circ$ around the $x$-, $y$- or $z$-axis. For this point, it can be used to determine the $x$- or $y$-axis of the crystal. The above phenomena are also observed in sample 4.

Moreover, in order to deeply understand the Čerenkov radiation, we describe some parameters of double rings in detail. Figure 10(h) shows the Čerenkov second-harmonic radiations of 1164 nm lasers in sample 4, in which there are six arcs at the following positions, which diverge the angles of $0^\circ$, $30^\circ$ and $60^\circ$ from the vertical line. This difference from sample 3 can be explained by the fact that there is a $90^\circ$ rotation difference around the $x$- or $y$-axis between samples 3 and 4. This agrees with the above description that the rotation of the radiation pattern will occur with the rotation of NPC.

Other than the above external conical angle $\beta$ outside the crystal, there is the internal conical angle of Čerenkov radiation inside the crystal $\theta$. Because LN is a negative single-axis birefringent crystal, both the external and internal angles relate to two kinds of output conical harmonics, which are ordinary ($o$) and extraordinary ($e$) lights forming the external and internal double ring pattern [17]. As for the definition, the internal conical angles inside the crystal are denoted by $\theta_o$ for ordinary light and $\theta_e$ for extraordinary light:

$$\theta_o = \cos^{-1}\left(\frac{n_1}{n_o}\right)$$

$$\theta_e = \cos^{-1}\left(\frac{n_1}{n_e(\theta_e)}\right) = \cos^{-1}\left(\sqrt{\frac{n_1^2 n_o^2 + n_1^2 n_e^2 - n_1^2 n_2^2}{2 n_1^2 n_o^2 n_e^2}}\right)$$

in which $n_1$ is the refraction index of the incident laser and ordinary polarized, $n_o$ and $n_e$ are the indexes of refraction of the Čerenkov external and internal harmonic radiations. $\theta_o$ is the angle between extraordinary wave vectors and optical $z$-axis of the LiNbO$_3$ crystal, and $n_e(\theta_e)$ is the index of refraction of the extraordinary wave propagating at an angle $\theta_e$ with respect to the optic axis due to the anisotropic crystal. Then on the basis of the law of refraction, the external conical angles outside the crystal are denoted by $\beta_o$ for ordinary light and $\beta_e$ for extraordinary light:

$$\beta_o = \sin^{-1}(n_o \sin \theta_o),$$

$$\beta_e = \sin^{-1}(n_e \sin \theta_e).$$
We are concerned about the conical angles of CRs under different input laser beams, so we make a specific measurement on sample 4. The distance $d$ is 1.3 cm.

Figures 12(a) and (b) correspond to internal and external rings, respectively. As shown in figure 12(a), stars represent the external angle obtained by measuring the radius of the internal ring on the screen. By the law of refraction, the internal angles symbolized by circles can be calculated by the measured external angle. Comparatively, the external and internal angles of the internal ring under different input wavelengths can also be calculated by the above theoretical equation, corresponding to the lines in figure 12(a). It can be concluded that the experimental results agree very well with the theoretical results. For instance, when the input wavelength is 0.95 $\mu$m, the external angles and internal angles experimentally are 45° and 17.5°, and theoretically 44.5° and
17.4°, respectively. Moreover, the external and internal angles of the external rings are given in figure 12(b). Taken as a whole, the distributions of the various angles in figure 12(b) are similar to those in figure 12(a). In addition, from the upper or lower parts in figures 12(a) and (b), it is drawn that the interval between the two rings is very little. In other words, the two rings are very close to each other. For instance, when the input wavelength is 1.3 μm, the external angles of internal and external rings are 33.2° and 34.7°, and the internal angles of internal and external rings are 13.9° and 14.5°, respectively. Finally, when incident laser wavelength is 1.05 μm, the external angles of the internal and external rings are 40° and 42.7°, respectively. They approximately agree with the results of 38° and 40° under 1.053 μm in LN crystals in [17].

4. Conclusion

In conclusion, two-dimensional LiNbO3 nonlinear photonic crystals with two kinds of Sierpinski fractal superlattices are successfully fabricated by applying high-voltage electric pulses. These structures have distinct advantages over H-shaped fractal and square periodic structures. The optical properties in the two kinds of LN NPCs with different Sierpinski superlattice structures are similar to each other. The single and multiple spot-shaped and line-shaped light outputs are obtained by quasi-phase matching harmonics. Ten kinds of spot-shaped outputs can be accomplished due to plenty of reciprocal vectors, in which the QPM harmonic conversion efficiency of fundamental wavelength 1.352 μm reaches 27%. Also, green line-shaped light sources with an expansion angle of about 3° are observed, which originates from the coexistence of imperfect collinear and non-collinear quasi-phase matching processes. In addition, for Čerenkov radiation, ring-shaped light sources of continuous wavebands in the harmonic wavelength range of 425 nm to 675 nm can be realized. Differently shaped light sources generated in NPCs with only one kind of superlattice can be useful in nonlinear optics, nano- or micro-structure formation and laser fabrication.

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References

[1] Zhu S N, Zhu Y Y and Ming N B 1997 Quasi-phase-matched third-harmonic generation in a quasi-periodic optical superlattice Science 278 843

[2] Yu Y B, Wang H J, Xiao M and Zhu S N 2011 Directly produced three-color entanglement by quasi-phase-matched third-harmonic generation Opt. Express 19 13949

[3] Berger V 1998 Nonlinear photonic crystals Phys. Rev. Lett. 81 4136

[4] Aleksandrovsky A S, Vyunixhev A M, Zaitsev A I, Pospelov G I and Slabko V V 2011 Diagnostics of fs pulses by noncollinear random quasi-phase-matched frequency doubling Appl. Phys. Lett. 99 211105

[5] Aleksandrovsky A S, Vyunixhev A M, Zaitsev A I and Slabko V V 2013 Random quasi-phase-matched nonlinear optical conversion of supercontinuum to the ultraviolet Appl. Phys. Lett. 103 251104

[6] Ni P G, Ma B Q, Wang X H, Cheng B Y and Zhang D Z 2003 Second-harmonic generation in two-dimensional periodically poled lithium niobate using second-order quasi-phase matching Appl. Phys. Lett. 82 4230

[7] Sheng Y, Roppo V, Ren M L, Kalinowski K, Cojocaru C, Trull J, Li Z Y, Koynov K and Krolikowski W 2012 Multidirectional Čerenkov second harmonic generation in two-dimensional nonlinear photonic crystal Opt. Express 20 3948

[8] Zhao X H, Zheng Y L, Ren H J, An N and Chen X F 2014 Čerenkov second-harmonic Talbot effect in one-dimensional nonlinear photonic crystal Opt. Lett. 39 5885

[9] Ren M L, Ma D L and Li Z Y 2011 Experimental demonstration of super quasi-phase matching in nonlinear photonic crystal Opt. Lett. 36 3696

[10] Sheng Y, Ma D L and Ren M L 2012 Broadband cascading of second-order nonlinearity in randomized nonlinear photonic crystal J. Phys. D: Appl. Phys. 45 365105

[11] Ayoub M, Roedig P, Imbrock J and Denz C 2011 Cascaded Čerenkov third-harmonic generation in random quadratic media Appl. Phys. Lett. 99 241109

[12] Ren H J, Deng X W, Zheng Y L, An N and Chen X F 2012 Nonlinear Čerenkov radiation in an anomalous dispersive medium Phys. Rev. Lett. 108 223901

[13] Bloch N V, Shemer K, Shapiro A, Shiloh R, Juwiler I and Arie A 2012 Twisting light by nonlinear photonic crystals Phys. Rev. Lett. 108 233902

[14] Lin Y C, Su K W, Huang K F and Chen Y F 2014 Pattern formation of second harmonic conical waves in a nonlinear medium with extended defect structure Opt. Express 22 27859

[15] Wang X J, Ren H J, An N, Zhao X H, Zheng Y L and Chen X F 2014 Large acceptance of non-collinear phase-matching second harmonic generation on the surface of an anomalous-like bulk dispersion medium Opt. Express 22 28234

[16] Chou P Y, Chang W K, Chung H P and Chen Y H 2014 Two-dimensional aperiodic nonlinear photonic crystal in a dual-wavelength Nd:YVO4 laser for pulsed orange generation Opt. Express 22 28857

[17] Saltiel S M, Sheng Y, Voloch-Bloch N, Neshev D N, Krolikowski W, Arie A, Koynov K and Kivshar Yuri S 2009 Čerenkov-type second-harmonic generation in two-dimensional nonlinear photonic structures IEEE J. Quantum Electron. 45 1465

[18] Zhang Y, Gao Z D, Qi Z, Zhu S N and Ming N B 2008 Nonlinear Čerenkov radiation in nonlinear photonic crystal waveguides Phys. Rev. Lett. 100 163904

[19] Ma B Q, Ren M L, Ma D L and Li Z Y 2013 Multiple second-harmonic waves in a nonlinear photonic crystal with fractal structure Appl. Phys. B 111 183

[20] Aguirre Vélez C, Lehman M and Garavaglia M 2001 Two-dimensional fractal gratings with variable structure and their diffraction Optik 112 209
[21] Armstrong J A, Bloembergen N, Ducuing J and Pershan P S 1962 Interactions between light waves in a nonlinear dielectric Phys. Rev. 127 1918

[22] Ma B Q, Wang T, Ni P G, Cheng B Y and Zhang D Z 2004 High-order quasi-phase-matching harmonic generation in two-dimensional orthorhombic lattice Europhys. Lett. 68 804

[23] Edwards G J and Lawrence M 1984 A temperature-dependent dispersion equation for congruently grown lithium niobate Opt. Quantum Electron. 16 373

[24] Coelho A S, Barbosa F A S, Cassemiroy K N, Villar A S, Martinelli M and Nussenzveig P 2009 Three-color entanglement Science 326 823