Nonlinear Photonic Quasiperiodic Spiral

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The design of nonlinear photonic Vogel’s spiral based on quasicrystal theory was demonstrated. Two main parameters of Vogel’s spiral were arranged to obtain multi-reciprocal circles. Typical structure was fabricated by near infrared femtosecond laser poling technique, forming nonlinear photonic structure, and multiple ring-like nonlinear Raman-Nath second harmonic generation were realized and analyzed in detail. The structure for cascaded third harmonic generation process was predicted. The results could help deepen the understanding of Vogel’s spiral and quasicrystal and pave the way for the combination of quasicrystal theory with more aperiodic structures.

**Keywords**: Nonlinear photonic quasicrystal; Second harmonic generation; Vogel’s spiral; Nonlinear Raman-Nath diffraction; Femtosecond laser poling.

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1. Introduction

Nonlinear photonic crystals (NPCs) generally refer to materials with periodic second-order nonlinearity $\chi^{(2)}$ and homogeneous refractive index, and they provide a flexible method to manipulate nonlinear optical parametric processes [1, 2]. The famous quasi phase matching condition could be satisfied to reach efficient optical frequency conversion and generate entangled photons via spontaneous optical frequency down conversion. Meanwhile, transverse reciprocal lattice vectors may modulate the phase of harmonic waves, achieving nonlinear wavefront shaping [3-9].

Instead of periodic $\chi^{(2)}$ gratings, many nonlinear photonic crystals with non-strictly periodic structures, such as Fibonacci and chirped lattices, have been reported widely for broadband or cascaded phase matching processes [10-12]. These structures can provide multiple components in Fourier space to satisfy several phase matching conditions at the same time. Among the design method of aperiodic NPC structures, the construction of nonlinear photonic quasicrystal is an active strategy to obtain the desired reciprocal lattice vectors for the required phase matching process at any spatial direction [13]. Quasicrystals are short-range disordered, but long-range ordered structures. In 1984, they were discovered by D. Shechtman et al. in an Al-Mn alloy with extremely rapid cooling, which has a fivefold axis of rotational symmetry, and is not possible in crystallography [14]. In 2005, nonlinear photonic quasicrystals were first proposed by applying dual-grid quasicrystal construction [13]. In the past decades, quasi phase matching with multi-fundamental wavelengths, multi-directions, and other functional nonlinear optical devices based on nonlinear photonic quasicrystals have been investigated popularly [15-24].

Vogel’s spiral is a kind of famous aperiodic structure with centrosymmetric pattern in Fourier space [25]. The arrangements of seeds in Vogel’s spiral could be expressed by the following equations:

\begin{equation}
\begin{cases}
    r = b \sqrt{n} \\
    \theta = n \alpha
\end{cases}
\end{equation}

where $r$ and $\theta$ are the coordinates in polar coordinate system, $b$ is the parameter describing density of the seeds, and $n=0,1,2,3\cdots$ and $\alpha$ give the azimuthal offset between two adjacent seeds. When $\alpha$ is the golden-angle ($\varphi = (1 + \sqrt{5})/2, \alpha = 2\pi / \varphi^2 \approx 2.4$), the structure evolves into the famous sunflower lattice, also named as golden-angle spiral, exhibiting an inner sharp ring with outer weak rings in Fourier spatial spectrum [26]. On the basis of this unique property, the sunflower-structured photonic crystals, photonic crystal fibers and nanoparticles have shown characteristic performances in complete photonic bandgap, birefringence, optical orbital angular momentum control, wave focusing, and mode localization [27-31]. In nonlinear optics, the sharp ring-like peak in Fourier spectrum means possibly efficient phase matching process. Thus, the golden-angle spiral structured nonlinear photonic crystals were proven to have prominent effect on the enhancement of broadband Čerenkov second harmonic generation [32]. This is the sole application example of Vogel’s spiral in nonlinear optics with the generation of an annular second harmonic signal. However, its application on multichannel second harmonic wave production is unknown, so the application of Vogel’s spiral structured nonlinear photonic crystals on other fields, such as generation of entangled photon pairs and quantum communication, is restricted.

In this paper, the quasicrystal theory and Vogel’s spiral were combined to obtain structures with strong multi-rings in Fourier spatial spectrum. Typical spiral was fabricated by near-infrared femtosecond laser poling forming
nonlinear photonic quasiperiodic spirals, and second harmonic generation via nonlinear Raman–Nath diffraction was captured and discussed. Furthermore, a nonlinear photonic quasiperiodic spiral aiming at cascaded third harmonic generation was designed. This work may provide new ideas for the design of hybrid nonlinear photonic structures.

2. Structure design and fabrication
In accordance with the Vogel's spiral expressed by Eq. (1), as a universal rule, when α is an irrational, the spiral is approximately homogeneous and isotropy. Hence, diffuse rings constitute the Fourier spectrum with the innermost one being far stronger than the outer ones. When the isotropy is destroyed, such as α is a rational number, radial arms form in Vogel's spiral and the Fourier spectrum shows several scattered rings instead of a single strong ring [33]. The isotropy of golden-angle spiral must be destroyed on purpose to obtain a spiral with two or more desired sharp rings near the center in Fourier spectrum. For this aim, the concept of quasicrystal could be borrowed.

Fig. 1 (a) Golden-angle spiral with \( b=1.70 \mu m \) and a total of 3000 points. (b) Nonlinear photonic quasiperiodic spiral structure based on rearrangement of \( \alpha \), the two construction parameters with \( \alpha_1 \) being the golden angle and \( \alpha_2=1.57 \), \( b=1.70 \mu m \). (c) Nonlinear photonic quasiperiodic spiral structure based on rearrangement of \( b \), the two construction parameters with \( b_1=1.50 \mu m \) and \( b_2=2.00 \mu m \). (d)-(f) Fourier spatial spectra of (a)-(c).

Fig. 2 (a) Golden-angle spiral with \( b=1.70 \mu m \) [an element of quasiperiodic spiral shown in Fig. 1(b)], (b) Vogel's spiral with \( \alpha =1.57 \) and \( b=1.70 \mu m \) [the other element of quasiperiodic spiral shown in Fig. 1(b)], (c) golden-angle spiral with \( b=1.50 \mu m \) [an element of quasiperiodic spiral shown in Fig. 1(c)], (d) golden-angle spiral with \( b=2.00 \mu m \) [the other element of quasiperiodic spiral shown in Fig. 1(c)]. (e)-(h), Fourier spatial spectra of (a)-(d).

The cut-and-project method for quasicrystal construction is employed [34]. First, the quasicrystal is constructed by two angles, with \( \alpha_1 \) being the golden angle and \( \alpha_2=1.57 \). A series of quasi-periodic angles with approximate radian values of 0, 1.57, 3.97, 6.37... could be obtained. In the new spiral, the \( q+1 \)th value from the smallest to the largest is set as \( \theta(q+1) \) with \( q=0,1,2,3... \). The spiral function is as follows:
where \(a_1\) and \(a_2\) are the construction parameters for \(\theta (q+1)\).

The quasiperiodic spiral expressed by Eq. (2) with \(b=1.70\ \mu m\) is shown by Fig. 1(b). This spiral has two elements: golden-angle spiral with \(b=1.70\ \mu m\) [Fig. 2(a)] and Vogel's spiral with \(a=a2\) and \(b=1.70\ \mu m\) [Fig. 2(b)]. The distance between the adjacent seeds in Fig. 1(b) is no longer invariable. The Fourier spectrum of Fig. 1(b) is shown in Fig. 1(e). Two sharp rings near the central region emerge. Then, the quasicrystal constructed by two distances was considered as follows: \(b_1=1.50\ \mu m\) and \(b_2=2.00\ \mu m\). Similar to that constructed by two angles, a quasi-period arranged distance values could be provided by the cut-and-project method, and the \(q+1\)th value from the smallest to the largest is set as \(b(q+1)\) with \(q=0,1,2,3,..\) The spiral function is as follows:

\[
\begin{align*}
    r &= b(q + 1) \\
    \theta &= q\alpha
\end{align*}
\]

where \(b_1\) and \(b_2\) are the construction parameters for \(b(q+1)\).

The typical spiral pattern with \(a\) being the golden-angle spiral are shown in Fig. 1(c), and its two elements are exhibited by Figs. 2(c) (golden-angle spiral, \(b=b_1\)) and (d) (golden-angle spiral, \(b=b_2\)). The corresponding Fourier spatial spectrum of Fig. 1(e) is exhibited in Fig. 1(f). Two sharp rings can be found, with another external one with weaker strength. These results indicate that the nonuniformity of \(a\) and \(b\) could be utilized for multi-ring-like peaks in Fourier space.

For nonlinear photonic quasiperiodic spirals, the radius of rings in Fourier spatial spectrum indicates the magnitude of the reciprocal lattice vector, which is pivotal in quasi phase matching. So, a definite relationship between the ring radius and elements of the quasiperiodic spiral is essential. As shown in the quasiperiodic spiral in Fig. 1(b), the radii of the two rings in its Fourier spectrum [Fig. 1(e)] were calculated to be 1.29 and 2.21 \(\mu m\). The Fourier spectra in Figs. 2(a) and (b) are shown by Fig. 2(e) and (g). In Fig. 2(e), a sharp ring with a radius of 2.04 \(\mu m\) emerges, which is separated from the two in Fig. 1(e). As shown in Fig. 2(g), no available ring-like reciprocal lattice vectors are present due to the formation of radial arms in Fig. 2(b). The quasiperiodic spiral expressed by Eq. (2) with different construction parameters was studied, and the ring radius in Fourier spatial spectrum can be hardly predicted. In other words, the numerical relationship between the radius of the two rings in Fig. 1(e) with \(a_1\) and \(a_2\) is uncertain, which is not friendly for the design of desired centrosymmetric ring-like reciprocal lattice vectors by quasiperiodic spiral following Eq. (2).

Two elements of the quasiperiodic spiral shown in Fig. 1(e) are demonstrated by Figs. 2(c) and (d), with Figs. 2(g) and (h) exhibiting their Fourier spectra. On the basis of the basic characteristic of golden-angle spiral, a sharp ring near the center is located in both of the two patterns. Their radii were calculated to be 1.75 [Fig. 2(g)] and 2.28 \(\mu m\) [Fig. 2(h)]. In Fig. 1(e), the rearranged \(b_1\) and \(b_2\) do not broke the isotropy of the spiral but only change the average distance of the adjacent seeds from a constant to two constants. So, the radii of the innermost and middle rings in Fig. 1(f) were calculated to be the same with the ones shown in Figs. 2(f) and (h). In other words, as long as two golden-angle spirals with ring-like reciprocal lattice vectors of given magnitude are designed, the quasiperiodic spiral simultaneously providing the two annular vectors could be obtained by the combination of the two golden-angle spirals following Eq. (3). For deep understanding of this method, the relation between the radius of ring in Fourier spectrum with \(b\) in Eq. (1) can be considered. In golden-angle spiral, the radius of ring is inversely proportional to the average particle spacing of the structure, which is determined by \(b\) [33]. When \(b\) is a quasi-periodic arrangement of \(b_1\) and \(b_2\) [Fig. 1(e)], its manifestation in Fourier spatial spectrum [Fig. 1(f)] should be corresponding with spirals with the two parameters severally. In addition to the two inner rings, a new ring is present, with the radius of 2.90 \(\mu m\). This ring comes from the increased disorder, similar to the rings in Fig. 1(e) by rearrangement of \(a\). The magnitude of this new ring is also unpredictable.

The spiral shown in Fig. 1(e) was employed for experimental investigation. The structure was machined out in an x-cut strontium barium niobate ferroelectric crystal (Sr0.61Ba0.39NbO3, SBN) by using infrared femtosecond laser poling at room temperature [35]. The size of the crystal is 5 mm \(\times\) 5 mm \(\times\) 1 mm, with the x-surfaces being polished for femtosecond laser processing and nonlinear detection. The SBN crystal was mounted on a translatable stage that can be moved along x-, y- and z-directions with a resolution of \(~100\) nm. The laser used in processing of this structure is a Ti:sapphire laser system (Chameleon Ultra II, Coherent) operated at 750 nm with a pulse width of 141 fs and a repetition frequency of 80 MHz. The processing light polarized along the z-axis of the crystal was focused by an objective lens (50 \(\times\), NA = 0.65) with a beam diameter of 1.2 \(\pm\) 0.2 mm and incidents normally into the crystal cross the x-surface. This laser focus was about 90 \(\mu m\) below the x-surface of the crystal and a halogen lamp is used to illuminate the crystal. When processing the structure, the laser power was regulated using a half-wave plate and polarizer, and the working pulse energy was regulated to 1.5–4 nJ during processing. The laser was chopped using an automatic shutter (SH05, Thorlabs). Each point was flashed two times with the laser to obtain domain inversion effectively [36].

3. Results and Discussion

Second harmonic generation based on the fabricated nonlinear photonic quasiperiodic spiral was carried out with a broadband femtosecond laser source (Chameleon Compact OPO, 1000–1600 nm) as the fundamental light.
The beam was focused by a 10× microscopic objective (NA = 0.3) and incident perpendicular with the crystal surface. The generated second harmonic signal was projected onto the light screen at far field, and then the diffraction pattern of the second harmonic wave was photographed using a charge-coupled device (CCD) camera. Different wavelengths (1480–1580 nm) were used for the observation of the structure. The diffraction patterns produced using different wavelengths were similar. Typical patterns obtained with the fundamental wavelengths at 1480 nm are shown by Fig. 3(a), with three distinct rings present in this image. The color in this figure is artificial for enhanced visualization.

![Fig. 3 (a) Typical second harmonic pattern generated by the fabricated nonlinear photonic quasiperiodic spiral. Three ring-like patterns are modulated by the designed structure via nonlinear Raman–Nath diffraction. The crosswise line that lies in the middle is produced by spontaneous domains. (b) Geometrical relation of the phase matching condition of nonlinear Raman–Nath diffraction. (c) Variation tendency of external angle relying on fundamental wavelength.](image)

The three ring-like second harmonic waves are produced by nonlinear Raman–Nath diffraction [37]. The phase matching condition could be expressed as follows:

$$ k_2 \sin \theta_m - G_m = 0 \ .$$  \hspace{1cm} (4)

The geometrical diagram is exhibited in Fig. 3(b), where $k_1$ and $k_2$ represent the wave vectors of the fundamental and second harmonic waves; $G_m$ is the reciprocal lattice vector; $\theta_m$ is the corresponding internal divergence angles of $k_2$ relative with $k_1$; $\Delta k_m$ is the longitudinal phase mismatch, and $m = 1$, 2, and 3 denote the innermost, middle, and outermost ring, respectively. The three reciprocal lattice vectors provided by the processed structure are $G_1 = 1.75 \ \mu m^{-1}$, $G_2 = 2.28 \ \mu m^{-1}$, and $G_3 = 2.90 \ \mu m^{-1}$. The longitudinal phase mismatches of the three ring-like second harmonic waves are $\Delta k_1 = 0.37 \ \mu m^{-1}$, $\Delta k_2 = 0.31 \ \mu m^{-1}$, and $\Delta k_3 = 0.23 \ \mu m^{-1}$. The proportion of vectors in Fig. 3(b) was exaggerated for enhanced visualization. External divergence angles $\beta_m$ could be obtained by the internal angles $\theta_m$, Snell’s law, and the Sellmeier equation of SBN crystal [38]. The measured results are shown in Table I. They fit well with the calculated angles. In the as-grown x-cut SBN crystal, spontaneous domains along the $z$-direction providing random reciprocal lattice vectors in the $x$-$y$ plane can be obtained. They can produce random quasi phase-matched second harmonic generation. The transverse bright streaks in Fig. 3(a) are second harmonic signals modulated by the spontaneous domains [39]. The theoretical external angles of the second harmonic rings that depend on fundamental wavelength are shown in Fig. 3(c). As phase mismatch exists in nonlinear Raman-Nath diffraction, the external angles change slowly with the variation of wavelength.

<table>
<thead>
<tr>
<th>$\beta_1$</th>
<th>$\beta_2$</th>
<th>$\beta_3$</th>
</tr>
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<tbody>
<tr>
<td>Theoretical angle (°)</td>
<td>11.89</td>
<td>15.89</td>
</tr>
<tr>
<td>Experimental angle (°)</td>
<td>11.55</td>
<td>15.54</td>
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</tbody>
</table>

The relative intensities of the three rings in Fig. 3(a) are shown by the red line in Fig. 4(a). The intensities of second harmonic waves produced by nonlinear Raman–Nath diffraction can be simulated by considering phase mismatch and Fourier components following the equation

$$ I_2 \propto c_m^2 \sin^2 \left( \frac{\Delta k_m}{2} \right) ,$$

(5)

where $I_2$ is the intensity of second harmonic beam, and $c_m$ is the Fourier coefficient. The modulation by random reciprocal lattice vectors is ignored in calculation. The theoretical second harmonic intensities related to the external diffraction angles are shown by the blue curve in Fig. 4(a), which fit well with the experimental results. The slight errors may be caused by the Sellmeier equation and the component of SBN crystal, as the refractive indexes relate closely with the ratio of Sr and Ba in SBN.
The demonstrated nonlinear photonic quasiperiodic spiral by Eq. (3) has extensive effect in complicated optical frequency conversion processes such as cascaded third harmonic generation and spontaneous frequency down conversion. With third harmonic generation as an example, appropriate ring-like reciprocal lattice vectors could realize frequency doubling and sum frequency generation processes simultaneously. The fundamental wavelength is assumed to be 1560 nm and perpendicular to the spiral, and the spiral was fabricated in x-cut SBN crystal. Nonlinear Bragg diffraction should be realized without phase mismatch to obtain a fine conversion efficiency. The phase matching diagram is shown in Fig. 4(b) on the basis of phase matching condition as follows:

\[
\begin{align*}
2k_1 + G_{SH} &= k_2 \\
k_2 + k_1 + G_{TH} &= k_3,
\end{align*}
\]

where \(k_1\), \(k_2\), and \(k_3\) are the wave vectors of the fundamental, second harmonics and third harmonic waves, respectively, and \(G_{SH}\) and \(G_{TH}\) are the transverse reciprocal lattice vectors for second and third harmonic generation, respectively. In Fig. 4(b), \(\theta_{SH}\) and \(\theta_{TH}\) are internal divergence angles of \(k_2\) and \(k_3\) relative with \(k_1\). Taking fundamental wavelength and Sellmeier equation of SBN into consideration, \(G_{SH}\) and \(G_{TH}\) should be 3.76 and 7.96 \(\mu m^{-1}\), respectively, and \(\theta_{SH}\) and \(\theta_{TH}\) should be 11.95° and 16.43°, respectively, with the corresponding external angles being 27.79° and 41.23°, respectively, to achieve the geometrical relation in Eq. (6).

The two parameters were calculated to be \(b_1 = 0.93\) and \(b_2 = 0.44\) \(\mu m\) for the construction of quasiperiodic spiral by Eq. (3) to proceed with the third harmonic generation via nonlinear Bragg diffraction on the basis of Eq. (6). In the spiral shown by Fig. 1(c), the closest distance between two seeds is 1.15 \(\mu m\), which almost reached the size limitation in femtosecond laser induced domain inversion. The closest distance between two seeds in spiral for third harmonic generation is 0.30 \(\mu m\), which is difficult to fabricate. So, the third harmonic generation process was exhibited in theory, and the calculated pattern containing the inside second harmonic ring and the outside third harmonic ring is shown in Fig. 4(c). In fact, an outermost ring is still present in the Fourier spatial spectrum of the designed spiral for third harmonic generation with the radius of 12.16 \(\mu m^{-1}\). When the fundamental wavelength is 1560 nm, this ring may also modulate harmonic waves via nonlinear Raman–Nath diffraction with the existence of phase mismatch. However, the efficiency should be far below the third harmonic generation via nonlinear Bragg diffraction. So, the effect of the outermost ring is ignored in Fig 4(c). Furthermore, if other wavelength components could be added in the incident fundamental wave, additional phase matching condition could be satisfied. For example, by utilizing the collinear second harmonic wave which is modulated by the random domains such the central bright spot in Fig. 3(b), the outermost ring could generate another third harmonic wave when a light source of 1372 nm is coupled into the incident beam.

4. Conclusion

In summary, nonlinear photonic quasiperiodic spiral was designed based on the basis of the combination of Vogel’s spiral and quasicrystal theory. Two basic parameters of Vogel’s spiral, \(a\) and \(b\), were rearranged to form a quasiperiodic spiral, which can provide multiple ring-like peaks in Fourier spatial spectrum. The predictability of ring radius was discussed, and the quasiperiodic spiral constructed by rearrangement of \(b\) was employed for further study. Typical structure was fabricated by femtosecond laser-induced domain inversion in ferroelectric SBN crystal to produce a nonlinear photonic quasiperiodic spiral. Conical second harmonic generation via nonlinear Raman–Nath diffraction was captured. The characteristics of the second harmonic patterns were measured, and they fitted well with the theoretical prediction. The nonlinear photonic quasiperiodic spiral for cascaded third harmonic generation was investigated in...
theory. The results may help for the extension of quasiperiodic theory and pave the way for the design of hybrid nonlinear photonic quasiperiodic spirals.

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