Optically Induced Nonlinear Cubic Crystal System for 3D Quasi-Phase Matching

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Quasi-phase matching (QPM) is a technique in nonlinear optics for achieving efficient energy exchange among optical waves at different frequencies, by spatially modulating the quadratic nonlinearity ($\chi^{(2)}$) of the medium. To realize the full potential of QPM, 3D spatial modulation of $\chi^{(2)}$ is required. This has become experimentally feasible recently thanks to the invention of femtosecond laser-based nonlinearity engineering in ferroelectric crystals. Herein, the first experimental demonstration of QPM second harmonic generation (SHG) in a nonlinear cubic crystal system is presented, in which $\chi^{(2)}$ modulations form simple cubic, body-centered cubic, face-centered cubic, and diamond cubic lattices, respectively. The experimental results indicate that these nonlinear cubic structures share the same primary reciprocal lattice vectors (RLVs), but possess different Fourier coefficients (in conventional cells), leading to SHG with similar angular resonances but various intensity distributions in the far field. This work contributes to a comprehensive understanding of nonlinear optical processes in 3D periodic media, and thus sheds light on the development of high-performance QPM devices.

1. Introduction

It is well known that the phase matching condition plays a critical role in determining the efficiency of optical parametric processes in nonlinear optics. Due to material dispersion, the nonlinear polarization wave and emitted optical waves propagate with different phase velocities, leading to the optical power of the latter oscillating between zero and a rather small value. To solve this problem and ensure continuous power flow into the generated wave, one can utilize nonlinear interacting waves with orthogonal polarizations and take advantage of the birefringence of the medium to ensure that the nonlinear polarization and optical waves have the same phase velocity. Another solution is to use quasi-phase matching (QPM) which involves a spatial modulation of the second-order nonlinearity ($\chi^{(2)}$) of the medium to compensate for the phase mismatch with the reciprocal wave vectors of $\chi^{(2)}$ structures. The structures with modulated nonlinearity are called nonlinear photonic crystals (NPCs). Taking the simplest process of second harmonic generation (SHG) as an example, the QPM condition is written as $k_2−2k_1−G = 0$, where $k_1$ and $k_2$ are wave vectors of the fundamental and second harmonic (SH) waves, and $G$ is the reciprocal lattice vector (RLV) of the periodic $\chi^{(2)}$ structure. The QPM can be achieved among interacting waves with the same polarizations such that the largest nonlinear coefficient of the medium can be used. It can also eliminate the spatial walk-off effect to enhance the conversion efficiency by using long samples. Moreover, the QPM technique enables the study of new nonlinear optical effects, which cannot be obtained with the birefringence phase matching. For instance, 

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DOI: 10.1002/adpr.202100268
simultaneous $\chi^{(2)}$ modulation in transverse and longitudinal directions leads to nonlinear wave front shaping, where a fundamental Gaussian beam can be converted to, e.g., the second harmonic Airy, Hermite–Gaussian, or Laguerre–Gaussian waves.\cite{11–14}

Spatial modulations of $\chi^{(2)}$ without changing the linear optical properties of the medium can be realized in ferroelectric crystals employing the periodic poling technique. Ferroelectric crystals can be divided into domains, with each domain representing the volume of the medium having spontaneous polarization ($P_s$) pointing into the same direction. Applying an electric field exceeding the coercive field along the polar axis of the crystal, the direction of $P_s$ can be reversed. This process is known as electric field poling.\cite{15} With the ferroelectric domain inversion, the sign of the second-order nonlinear coefficient is reversed, but the refractive index of the crystal remains unchanged. The electric field poling has been commonly used to fabricate high-quality, large-area (in length of millimeters or centimeters) NPCs with 1D and 2D $\chi^{(2)}$ modulations. In fact, when the concept of NPC was proposed by Berger in 1998, the superiority of 3D $\chi^{(2)}$ modulations for nonlinear optical interactions and their control were drawn.\cite{6} It is obvious that only 3D structures can provide RLVs along arbitrary directions to utilize the full potentials of QPM. Despite theoretical studies on 3D NPC-based QPM effects,\cite{16–19} the fabrication of 3D QPM structures has been a challenge over the last two decades due to the lack of a technique capable of 3D $\chi^{(2)}$ engineering.

A breakthrough was achieved recently when tightly focused near-infrared femtosecond laser pulses were used to engineer the second-order nonlinearity $\chi^{(2)}$ of ferroelectric crystals.\cite{20–24} The ultrahigh power of the femtosecond pulses allows one to either revert the sign of $\chi^{(2)}$ ferroelectric domain inversion\cite{25–27} or reduce the amplitude of $\chi^{(2)}$ by material modifications\cite{28–30} in the focal volume of the laser beam. As the infrared pulses can be focused anywhere inside the transparent ferroelectric crystals, one may realize the 3D $\chi^{(2)}$ modulation. For instance, a 3D NPC with the tetragonal primitive structure was demonstrated by ferroelectric domain inversion in barium calcium titanate crystal, and its spatially abundant RLVs were used to control the emission angles of nonlinear Čerenkov radiation.\cite{20} Meanwhile, the tetragonal primitive $\chi^{(2)}$ structure was also realized by laser-induced $\chi^{(2)}$ reduction in LiNbO$_3$ crystal, and the SHG using different orders of RLVs was investigated.\cite{21}

Following these two pioneering works, 3D NPCs with specifically designed $\chi^{(2)}$ structures, such as the multilayered structures, periodic fork structures, and nonlinear volume holograms, have been fabricated.\cite{23,31–33} The research focus has been on the nonlinear beam shaping in 3D configuration, e.g., the conversion of a fundamental Gaussian beam to multiple SH beams with various shapes of wave front\cite{31} and the QPM SH vortex beam generation.\cite{32,33} While superiorities of 3D NPCs were well demonstrated in these previous works, the basic $\chi^{(2)}$ crystal systems with 3D Bravais lattices have not been systematically studied in the experiment. In fact, there are 14 Bravais lattices in 3D space. Each of them provides particular distribution of RLVs in Fourier space, and thus different QPM conditions could be accomplished. Among them, the cubic crystal system is one of the most common and simplest 3D structures, by which the 3D QPM characterizations (such as the dependences of conversion efficiency on the orders of QPM and duty cycles) can be easily verified. The 3D Bravais lattices also have a natural advantage in expanding the capabilities of NPCs for simultaneous QPMs in multiple directions, which may have important applications in optical information processing, laser displays, and communications. It has been theoretically predicted that 3D Bravais $\chi^{(2)}$ lattices possess more plentiful RLVs than 1D and 2D structures, which increases the flexibility in QPM interactions and enables more complex nonlinear processes.\cite{16,17} However, such predictions have not been experimentally verified yet.

This paper presents the first experimental fabrication of nonlinear cubic lattices by using the femtosecond laser-induced ferroelectric domain inversion technique, including simple cubic (SC), body-centered cubic (BCC), face-centered cubic (FCC), and diamond cubic (DC). The quasi-phase-matched SHG experiments are performed with these 3D $\chi^{(2)}$ lattices, respectively. The results show that the cubic NPCs process plenty of RLVs, enabling more than 20 channels of SHG with the incidence of a single fundamental beam. Depending on the associated Fourier coefficients and duty cycles of the $-\chi^{(2)}$ areas, certain high orders of QPM interactions are missing in the BCC, FCC, and DC structures. It is therefore necessary to obtain a full understanding of the QPM properties of nonlinear cubic lattices and select suitable structures according to practical demands.

2. QPM Properties of Nonlinear Cubic Crystal System: Theoretic Analysis

The cubic crystal system is one of the most common and simplest shapes found in crystals. It possesses the highest symmetrical characteristic in the seven crystal systems. The cubic NPCs refer to artificial microstructures, where the cubic lattice points are occupied by the areas with opposite signs of the second-order nonlinear coefficient, namely $-\chi^{(2)}$. Such structures can be mathematically treated as a convolution of the cubic lattice and a motif of $-\chi^{(2)}$. There are three Bravais lattices within the cubic crystal system: SC, BCC, and FCC. Accordingly, NPCs of SC, BCC, and FCC structures are investigated in this work. The nonlinear SC lattice is the simplest one, which consists of one lattice point on each corner of the cube. The conventional cell of the SC lattice is shown in Figure 1a, with the motifs taking spherical shapes as an example. The BCC-NPC has one lattice point in the center of the unit cell in addition to the eight corner points of SC, and the FCC-NPC has lattice points on the centers of the faces of the SC lattice. Their conventional cells are shown in Figure 1b,c, respectively. In addition, another kind of structure called DC lattice is generally classified into the cubic crystal system too, whose properties in linear optics have attracted great interest owing to the existence of complete bandgaps.\cite{34} Therefore, the DC-NPC is also included in this work. Its conventional cell is shown in Figure 1d, which can be regarded as a pair of intersecting FCC lattices, with each separated by one-fourth of the width of the unit cell in each dimension.

The QPM properties of the nonlinear cubic lattices are analyzed by considering the corresponding RLVs in Fourier space. Here, we use the conventional cells to calculate the Fourier spectrum of the cubic NPCs. In fact, both conventional and primitive
cells can be used, but the unit RLVs of the primitive cells are not orthogonal, making the analysis of the Fourier spectrum more complicated. It is worth noting that the conclusion that BCC and FCC lattices are real and reciprocal lattice of each other is established based on the primitive cell. By using the conventional cells, as shown in Figure 1a–d, the unit RLVs of the cubic NPCs can be written as, \( G_x = \frac{2\pi}{\Lambda} \hat{x}, G_y = \frac{2\pi}{\Lambda} \hat{y} \) and, \( G_z = \frac{2\pi}{\Lambda} \hat{z} \) in which \( \Lambda \) is the lattice period (Figure 1e). The available RLVs in the cubic

Figure 1. a–d) The conventional cells of the cubic NPCs with SC, BCC, FCC, and DC lattices, respectively. e) The primary RLVs of the cubic NPCs and the corresponding nonlinear Ewald sphere for 3D quasi-phase-matched SHG. The center of the nonlinear Ewald sphere is located \( 2k_1 \) away from the origin of the RLVs, and the radius of the sphere is \( k_2 \). f–i) The nonlinear optical microscopic images of the nonlinear cubic structures fabricated with the ultrafast laser engineering of ferroelectric domains in as-grown, z-cut Sr0.61Ba0.39Nb2O6 crystals. The insets give magnified views of the unit cells for each structure. These microscopic images were obtained with the Čerenkov second harmonic microscopy.
NPCs can be expressed by $G_{h,k,l} = hG_x + kG_y + lG_z$, where $h$, $k$, and $l$ are integers. To characterize the emission direction of the QPM SHG using RLV $G_{h,k,l}$, two emission angles are defined. The conical angle ($\alpha$) is the angle between the SH wave vector $(k_f)$ and the optical axis ($z$-axis) of the crystal, and the azimuthal angle ($\phi$) is the angle of the $k_f$ projection in the $x$-$y$ plane against the $y$-axis (see Figure 1e).

Assuming negligible depletion of the fundamental wave, the intensity of the QPM SHG ($I_{2\omega}$) follows the equation

$$I_{2\omega} \propto d_{\text{eff}}^2 |\langle h,k,l \rangle|^2 \sin^2 \left( \frac{\Delta kL}{2} \right)$$  \hspace{1cm} (1)

in which $d_{\text{eff}}$ is the effective nonlinear coefficient taking $d_{\text{eff}} = d_{11} \sin \alpha$ when the fundamental beam is $x$-polarized and propagates along the $z$-axis of the strontium barium niobate ($\text{Sr}_{0.61}\text{Ba}_{0.39}\text{Nb}_2\text{O}_6$, SBN) crystal used in this work (see Figure 1e); $G_{h,k,l}$ is the Fourier coefficient associated with the RLV $G_{h,k,l}$, $\Delta k = k_f - 2k - G_{h,k,l}$, $L$ is the propagation distance of the fundamental beam in crystal. While the RLVs of nonlinear cubic lattices can be expressed with the universal equation, their Fourier coefficients are different. Considering spherical motifs

$$g_{h,k,l}(\text{DC}) = g_{h,k,l}(\text{SC}) \left[ 1 + e^{i\pi(h+k+l)} + e^{i\pi(h+l)} \right] \left[ 1 + e^{i\pi(h+k+l)} \right] \left( D \leq \frac{3}{8} \right)$$  \hspace{1cm} (5)

The Fourier spectra of BCC-, FCC-, and DC-NPCs are similar to that of the SC-NPC, but their magnitudes are modulated by the structure factors, which are polynomials containing one (or several) exponential functions. These structure factors take the maximal values when all exponential functions in the polynomials are real and equal to 1. For instance, the maxima of structure factor in BCC structure occurs when the value of $h + k + l$ is even integer in Equation (3), and the structure factor of FCC-NPC takes the maxima when the values of $h + k$, $k + l$, and $h + l$ are all even integers ($h$, $k$, and $l$ are all even or all odd integers) in Equation (4). The detailed maximum conditions of Fourier coefficients for the BCC, FCC, and DC structures are listed in Table 1. With the same period and duty cycle, the maximal values of $|g_{h,k,l}(\text{BCC})|$ and $|g_{h,k,l}(\text{FCC})|$ are two and four times of $|g_{h,k,l}(\text{SC})|$, respectively, when $h$, $k$, and $l$ all take the same values.

The minimal Fourier coefficients of the nonlinear cubic structures can be zero, depending on both, the structure factor and the duty cycle. In Table 1, the conditions of zero Fourier coefficients related to the structure factors are listed. Taking the BCC-NPC as an example, $g_{h,k,l}(\text{BCC})$ is zero provided $h + k + l = 2n + 1$ $(n \in \mathbb{Z})$, i.e., when the sum of the indexes is an odd number. It means the SHG of QPM orders satisfying $h + k + l = 2n + 1$ $(n \in \mathbb{Z})$ will not be generated at all in BCC-NPC, no matter what the duty cycle is. In contrast, according to Equation (2), the Fourier coefficient of SC-NPC also depends on the duty cycle. For instance, when $D$ equals 0.41, $g_{1,1,1}(\text{SC})$ takes zero. It means no matter what the structure factors are, the SHG cannot be generated using $G_{1,1,1}$. This rule applies to all the other three cubic structures, according to Equation (3–5).

### 3. Results and Discussion

#### 3.1. Fabrication of Nonlinear Cubic Photonic Crystals

The direct femtosecond laser writing technique\(^{[20]}\) was used to fabricate the nonlinear cubic structures in a $z$-cut, as-grown SBN

| Fourier coefficient | Maximum condition | Maximum value | Minimum condition | Minimum value |
|---------------------|-------------------|---------------|-------------------|--------------|
| $g_{h,k,l}(\text{BCC})$ | $h + k + l = 2n$ $(n \in \mathbb{Z})$ | $2|g_{h,k,l}(\text{SC})|$ | $h + k + l = 2n + 1$ $(n \in \mathbb{Z})$ | 0 |
| $g_{h,k,l}(\text{FCC})$ | $h, k, l$ all even or all odd | $4|g_{h,k,l}(\text{SC})|$ | $h, k, l$ not all even or odd | 0 |
| $g_{h,k,l}(\text{DC})$ | $h, k, l$ all even and $h + k + l = 4n$ $(n \in \mathbb{Z})$ | $2|g_{h,k,l}(\text{SC})|$ | $h, k, l$ not all even or odd | 0 |
| | $h, k, l$ all odd | $4\sqrt{2}|g_{h,k,l}(\text{SC})|$ | $h, k, l$ all even and $h + k + l \neq 4n$ $(n \in \mathbb{Z})$ | 0 |
crystal via ferroelectric domain inversion. The SBN crystal has spontaneous ferroelectric domains of random sizes and distributions. But these random domains can only generate rather weak SHG,\textsuperscript{[35,36]} which can be ignored when compared with those generated by the optically induced periodic domain patterns. The laser pulses (wavelength 800 nm and pulse duration 180 fs) were tightly focused and then scanned in 3D to create a desired 3D pattern of inverted domains (see Experimental Section for details). The fabricated SC-, BCC-, FCC-, and DC-NPCs are visualized by the Čerenkov second harmonic microscopy\textsuperscript{[37]} and the results are shown in Figure 1f-i, respectively. The Čerenkov second harmonic signal gets much stronger where the second-order nonlinear coefficient undergoes spatial variations, so the bright regions in these graphs represent ferroelectric domain walls\textsuperscript{[37–40]}. To avoid the merging of neighboring domains, the periods of the 3D lattices are selected to be $\Lambda_{\text{SC}} = \Lambda_{\text{BCC}} = 20 \mu m$ and $\Lambda_{\text{FCC}} = \Lambda_{\text{DC}} = 35 \mu m$, respectively. As shown in Figure 1f-i, the shapes of laser-induced inverted domains are not perfectly spherical, but more like cones shrinking from the top (radius about 1.3 μm) to bottom (0.5 μm). The whole dimensions of the laser-processed NPCs are $200 \times 200 \times 140 \mu m^3 (x \times y \times z)$.

### 3.2. Quasi-Phase-Matched SHG Experiment

To experimentally characterize the QPM properties of the cubic NPCs, the SHG experiments were performed using a femtosecond laser source operating at 1560 nm. The fundamental beam was loosely focused and propagated along the $z$-axis of the SBN crystal to generate the SH waves using the nonlinear coefficient $d_{41}$. Detailed information on the experiment setup is given in the Experimental Section. The SH emissions distributed along a set of homocentric rings are observed in the experiment, and the recorded far-field images are shown in Figure 2a for the SC-, BCC-, FCC-, and DC-NPCs, respectively. The magnified visualizations of the SHG patterns in the first internal ring are given in Figure 2b–e.

To analyze the emission angles of the SHG, the principle of nonlinear Ewald construction is employed, i.e., the QPM SHG occurs along the directions where the end of an RLV is located on the Ewald sphere. The center of the Ewald sphere is located $2k_{1}$ away from the origin of the RLVs, and the radius of the sphere is $k_{2}$ (Figure 1e).\textsuperscript{[6]} The vectorial relation among $k_{2}$, $2k_{1}$, and $G$ can be divided into two scales along the transverse and longitudinal directions. The conical emission angle of SHG is determined by the longitudinal phase-matching condition, i.e., $\alpha = \cos^{-1}\left(\frac{2k_{1} + iG_{y}}{k_{2}}\right)$ (internal angle), whose fulfillment ensures the growth of the SH wave with interaction distance. It is obvious that the SHGs phase matched with those RLVs having the same index possess the same conical angle, and their projections on a screen in far field form the rings (Figure 2a). Taking the SHG in SC-NPC as an example, the emissions using RLVs with $l=1$ are arranged on a ring with the external conical angle of 14.82°, agreeing well with the predicted 14.30° ($\beta = \sin^{-1}(n_{r} \sin \alpha)$ in which $n_{r}$ is the refractive index of the SH wave\textsuperscript{[43]}). The bigger ring has an experimental conical angle of 28.27°, and the calculated is 28.79° with $l=0$.

The transverse component of the QPM condition can be written as $\sqrt{hG_{x}^2 + (kG_{y})^2} = k_2 \sin \alpha$. For the cubic NPCs in this work, it simplifies into $\sqrt{h^2 + k^2} = \frac{k_{2} \sin \alpha}{c_{\text{cr}}}$. And the azimuthal angle is $\phi = \tan^{-1}\left(h/k\right)$ as $G_{x} = G_{y}$. For a fixed conical angle $\alpha$, the subjects $h$ and $k$ of the RLVs that satisfy the full QPM condition follow the rule of $\sqrt{h^2 + k^2} = A$, and $A$ is a constant for a fixed conical angle $\alpha$. For example, for the perfectly quasi-phase-matched SHGs scattered on the first ring of the FCC structure ($l=1$), the theoretical value of $\sqrt{h^2 + k^2} = 6.333$. The RLVs $G_{5,4,1}$, $G_{5,4,1}$, $G_{5,4,1}$, and $G_{5,4,1}$ offer the value of 6.403, so the SHGs generated using these RLVs belong to the perfect QPM interaction. Aside from the perfect QPM, those that are very close to the perfect emission angles with concessional phase mismatch were also observed in our experiment. For instance,
the SHGs using RLVs $G_{234,1}$, $G_{344,1}$, $G_{344,7}$, and $G_{344,7}$ were all observed in the BCC-NPC ($\sqrt{h^2+k^2} = 5$, and phase mismatch $\Delta k = 0.419 \, \mu m^{-1}$). The values of experimentally measured and calculated external conical angles ($\beta$), azimuthal angles ($\varphi$), and the corresponding values of $\sqrt{h^2+k^2}$ are listed in Table 2 for the SHGs in SC, BCC, FCC, and DC-NPCs, respectively. Good agreement between theory and experiment has been achieved. The slight deviations may be caused by structure imperfections of the fabricated samples. Meanwhile, the limited number of QPM periods of the used sample broadens the Fourier spectrum in reciprocal space, and hence is also a reason for the discrepancy between experimental measurements and theoretical predictions. In addition, the inaccuracy of the Sellmeier equation used in theoretical calculations can also cause some deviations. More detailed information about the measurement and calculation of the SH emission angles is given in the Supporting Information (Table S1–S4).

In addition to the phase-matched SH spots distributed along the homocentric rings, there are also SH patterns at the central area, such as shown in Figure 2a,b. While these harmonics near the center suffer from phase mismatch in the longitudinal direction,[9,42] they are occasionally even brighter than those fully phase-matched. For example, as shown in Figure 2d, the SH using $G_{222,2}$ is stronger than the phase-matched using $G_{26,2}$. This is because, in addition to the phase-matching condition, other factors including Fourier coefficients and duty cycle affect the efficiency of SHG as well. In fact, the SH emission shown as spot #15 (Figure S3c, Supporting Information) features a small phase mismatch of 0.0594 $\mu m^{-1}$, corresponding to the coherent length of 53 $\mu m$. Considering the small phase mismatch and really short sample length (140 $\mu m$), the Fourier coefficients play a more obvious role such that SHG using $G_{222,2}$ was stronger in the experiment. We measured the strength of SHG formed using $G_{222,2}$ is nearly three times stronger than that using $G_{2,6,2}$, agreeing well with the calculated ratio of 2.6 with a duty cycle of 0.029. To obtain more efficient QPM interaction, one should utilize those RLVs with Fourier coefficient as large as possible. This can be achieved by using a bigger index $l$ to direct the SH closer to the central area, such that $h$ and $k$ take smaller values in favor of higher Fourier coefficients. In addition to the Fourier coefficients, one should also consider the effective nonlinearity to achieve a more efficient QPM interaction. In the geometry of interaction in this work, the effective nonlinear coefficient $d_{eff} = d_{13} \sin \alpha$ is zero for $\alpha = 0$. Therefore, the emission angle of the second harmonic should be designed giving consideration to both, the Fourier coefficients and the effective nonlinearity. The bigger SH rings ($l = 0$) in Figure 2a belong to the nonlinear Čerenkov radiation, with no participation of RLVs in the longitudinal direction.[43] The FCC and DC structures have additional SH rings with $l = 2$ because they have larger periods ($35 \, \mu m$) than the others ($20 \, \mu m$).

While the SH emissions from the four cubic NPCs all form concentric circles consisting of SH spots, the distributions and strengths of these spots depend on their Fourier coefficients. As we have mentioned in the previous section, the QPM interactions of certain orders cannot occur due to the zero Fourier coefficients in the BCC, FCC, and DC-NPCs. This is the case of RLVs, e.g., $G_{5,1,1}$ in the BCC, $G_{6,5,2}$ in the FCC, and $G_{8,8,2}$ in the DC structures, respectively. Giving more details with the DC structure as an instance, the missing order $G_{8,8,2}$ has a transverse phase mismatch of 0.002 $\mu m^{-1}$, even smaller than that of the observed fully-phase-matched using $G_{2,8,2}$ (0.055 $\mu m^{-1}$). However, the QPM order of $G_{8,8,2}$ was not observed at all due to its zero Fourier coefficients ($h = 3$, $k = 8$, and $l = 2$, satisfying the condition that $h$, $k$, and $l$ are neither all evens nor all odds according to Equation (5)).

Figure 3a–d shows the calculated second harmonic intensity distributions in the far field using Equation (1). It is clearly seen a very good agreement between the experimental (Figure 2) and calculation. For a better view, only SH rings of the first and second orders are displayed. The SH emissions on the same ring have the same conical angle $\alpha$, so they have the same effective nonlinear coefficient $d_{eff}$. In other words, the SH intensities are independent of the azimuthal angle $\varphi$ (Figure 2b–e and 3a–d).

As mentioned in Section 2, for the RLVs of the same order and the same period and duty cycles, the Fourier coefficient of the BCC-NPC is two times that of SC-NPC (Equation (1)–(3)). Accordingly, the intensity of the second harmonic generated in the BCC-NPC is four times that of the corresponding signal in the SC structure. (These two structures are compared here because they have the same periods in our experiment.) This relation has been experimentally verified. In Figure 3e, the measured intensity distributions of the SH spots corresponding to $G_{1,6,2}$ are shown for the SC- and BCC-NPCs, respectively. It is clearly seen that the intensity ratio is nearly 4. In these graphs, the angle $\theta$ is used to describe the angular width of the SH spots, and was measured via $\tan(\theta) = \Delta y/\Delta d$, with $\Delta y$ representing the variation of the SH beam size with respect to a change of observation distance $\Delta d$. Similarly, the FCC and DC samples have the same

| NPC | RLVs | $\beta$ | $\varphi$ | $\sqrt{h^2+k^2}$ | $\beta$ | $\varphi$ | $\sqrt{h^2+k^2}$ |
|-----|------|---------|---------|-----------------|---------|---------|-----------------|
| SC  | $G_{5,6,1}$ | 14.09° | 18.85°  | 6.325           | 14.30° | 18.43°  | 6.333           |
| BCC | $G_{6,4,1}$ | 14.53° | 51.53°  | 6.403           | 14.30° | 51.34°  | 6.333           |
| FCC | $G_{6,6,2}$ | 11.28° | 45.43°  | 8.485           | 10.99° | 45°     | 8.555           |
| DC  | $G_{2,8,2}$ | 10.23° | 14.68°  | 8.246           | 10.99° | 14.04°  | 8.555           |

Table 2. Experimental and theoretical values of emission angles of the 3D QPM interactions. In the experiment, the external conical angles $\beta$ and azimuthal angles $\varphi$ were measured, and the theoretical values are derived according to snell’s refraction. The refractive indices of fundamental and SH waves are available in the previous report.[41]
periods (35 μm), so the second harmonic signals are compared between these two structures. In Figure 3f, the intensity profiles of the SH spots using RLVs $G_{2,8,2}$ and $G_{11,13,1}$ are presented for the FCC- and DC-NPCs, respectively. The corresponding Fourier coefficients belong to the maxima ($G_{2,8,2}$) and the secondary maxima ($G_{11,13,1}$) of the DC structure (see Table 1). It is clearly seen the measured ratio of 4 and 2 between SHGs of the same orders from the DC and FCC structures, agreeing well with the predictions.

The measured dependences of the second harmonic powers on the power of the fundamental beam are shown in Figure 4a,b for the SC-, BCC-, FCC-, and DC-NPCs, respectively. Here, the SHGs of orders $G_{3,-6,1}$, $G_{2,8,2}$, and $G_{11,13,1}$ are presented as examples. The circles and squares in these graphs represent the experimental results, with the fitting curves showing quadratic relation. The influence of duty cycles on the SH intensities is theoretically studied, and the results are shown in Figure 4c (SH emissions using $G_{3,-6,1}$ in SC- and BCC-NPCs) and 4d (SH emissions using $G_{2,8,2}$ and $G_{11,13,1}$ in FCC- and DC-NPCs), respectively. It can be seen that the SH intensities oscillate with duty cycles, with zero intensities appearing at certain values. The duty cycles of the fabricated cubic NPCs are marked by the dashed vertical lines in the insets of Figure 4c,d. It can be seen that the experimental duty cycles are far away from the optimal values for all these 3D NPCs. To obtain more efficient SHG, one needs to increase the duty cycles in the experiment. This can be achieved either by increasing the size of inverted domains (e.g., writing several inverted domains that are close enough to merge into a big one) or by reducing the period of the cubic NPCs.

![Figure 3. The simulated SHG emissions from a) SC-, b) BCC-, c) FCC-, and d) DC-NPCs, respectively. e) The recorded intensity distribution of the SH signals produced by $G_{3,-6,1}$ in the SC- and BCC-NPCs. f) The comparison of the recorded SH signals produced by $G_{2,8,2}$ and $G_{11,13,1}$ in the FCC- and DC-NPCs. Here, $\theta$ represents the angular width of the SH spots.](image-url)
4. Conclusion

The 3D NPCs with cubic lattice systems (SC, BCC, FCC, and DC) have been fabricated using the femtosecond laser-induced ferroelectric domain inversion technique in z-cut Sr$_{0.61}$Ba$_{0.39}$Nb$_2$O$_6$ crystals. The quasi-phase-matched SHGs in such nonlinear cubic crystal systems were experimentally studied. It was found that some orders of second harmonic emission were missing from the BCC, FCC, and DC structures, resulting from zero Fourier coefficients of the associated RLVs or duty cycles. The SHG conversion efficiencies have also been investigated for the cubic NPCs, and the experimental results agree well with theoretical predictions. The 3D nonlinear cubic photonic crystals with abundant RLVs are promising for the construction of advanced nonlinear optical and photonic devices. While the experiments were carried out using SHG, the principles revealed in this work apply to other nonlinear processes such as sum/difference frequency mixing and spontaneous parametric down conversion. The conversion efficiencies may be remarkably enhanced, e.g., by optimizing the duty cycles of the cubic NPCs, thus allowing for their practical implementation.

5. Experimental Section

Fabrication of 3D Nonlinear Photonic Crystals: A femtosecond laser source (Coherent MIRA 900) operating at 800 nm, with a pulse duration of 180 fs and a repetition rate of 76 MHz, was employed in direct laser writing of ferroelectric domain patterns. The incident beam was x-polarized. The pulse energy of the incident beam was controlled by a half-wave plate and a polarizer. In the experiment, the utilized pulse energy was about 5.5–6 nJ. The incident beam was focused by a 50×/0.65 microscope objective. The diameter of the focused beam was about 1 μm. The z-cut SBN crystal with the dimensions of 5×5×1 μm$^3$ was fixed on a stage that could move along the perpendicular x, y, and z directions with the resolution of 100 nm. When the SBN crystal was illuminated by the focused laser for 0.5 s, the spontaneous ferroelectric domains aligned along the same direction in the focal volume of the laser pulses. After this, the laser focus was moved to the next position to repeat the ferroelectric domain inversion process and in this way, the 3D NPCs were obtained.

Quasi-Phase-Matched SHG Experiment: A fiber laser operating at the wavelength of 1560 nm, pulse width of 130 fs, and repetition rate of 80 MHz (Langyan Tech, ErFemto ProH) was utilized as the fundamental beam. This beam was x-polarized and propagated in the fabricated NPC along the z-axis. The fundamental light beam was focused by a 10× objective lens (NA = 0.25). The beamwidth was estimated to be 50 μm. The sample was mounted on the three-axis stage. The emitted SH waves were projected on a screen 1 cm away from the sample, with the fundamental wave being eliminated by a short-pass filter. The far-field SH intensity patterns were recorded by a charge-coupled device (CCD) camera.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.
Acknowledgements

This work was supported by the National Natural Science Foundation of China (Grant Nos. 61905124, 11974196, 61905125, 62090063, and 62090064), Natural Science Foundation of Zhejiang Province, China (Grant No. LY22F050009), Yongjiang Scholar Foundation of Ningbo, and K.C. Wong Magna Fund of Ningbo University. W.K. acknowledges the support from the Qatar National Research Fund (grant no. NPRP12S-0205-190047).

Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available in the supplementary material of this article.

Keywords

laser frequency conversion, nonlinear photonic crystals, quasi-phase matching, second harmonic generation

Received: August 29, 2021
Revised: December 14, 2021
Published online: January 18, 2022

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