# Multiphoton path-entanglement generation by concurrent parametric down-conversion in a single $\chi^{(2)}$ nonlinear photonic crystal

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We propose a two-dimensional $\chi^{(2)}$ nonlinear photonic crystal capable of satisfying quasi-phase-matching conditions for two or three parametric down-conversions simultaneously. Seeded by a two-mode coherent state, this crystal can produce two-, three-, four-, or five-photon path-entangled states, provided the corresponding number photons are produced, i.e., in a postselection-free way. In particular, up to five-photon NOON state can be generated, which enables phase supersensitive measurements at the Heisenberg limit. The concurrent multiple quasi-phase-matchings provided by the nonlinear photonic crystal open up a way to integrated quantum light sources.

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## I. INTRODUCTION

Photonic entangled states, especially multiphoton entangled states, play a key role in photonic quantum technologies [1]. Particularly useful states are maximally multiphoton path-entangled states (called NOON states),

$$|N :: 0\rangle = (|N,0\rangle + |0,N\rangle)/\sqrt{2}, \tag{1}$$

which mean $N$ indistinguishable photons in an equal superposition of being all together in either path mode. The NOON states can be used in the precision phase measurement achieving the ultimate Heisenberg limit [2,3] and have potential applications in super-resolution lithography exceeding the Rayleigh diffraction limit [4]. More generally, two-mode $N+M$ entangled states

$$|N :: M\rangle = (|N,M\rangle + |M,N\rangle)/\sqrt{2} \tag{2}$$

are superior to the NOON states in the presence of loss [5], although they cannot reach the Heisenberg limit. Consequently, a practical method for generating $|N :: M\rangle$ states is demanding for their realistic applications.

Experimentally, the two-photon NOON state can be easily generated by Hong-Ou-Mandel interference [6]. However, producing NOON states with $N > 2$ is not straightforward. A variety of theoretical schemes [7] based on single photons or Fock states, linear optical elements, and detectors have been proposed for generating large NOON states via measurement-induced nonlinear interactions [8]. However, due to limitations of sources or detectors, these schemes cannot be easily realized at present and only three-photon NOON states [9,10] and four-photon "NOON-like" states [11] were obtained. Several experiments depending on the state projection measurement succeeded in observing three- [12], four- [13], and six-photon NOON states [14]. However, these NOON states, in a postselected way, cannot work if the two path modes are mixed into one. This drawback limits many applications, for example, in quantum lithography. Thus postselection-free [9,10] or heralded [15] NOON states are more applicable.

Another efficient way to create NOON states is by suppressing the unwanted terms via interference of the parametric down-conversion (PDC) light and the coherent light [16,17]. A recent experiment [18] yielded up to five-photon NOON states with an optimal theoretical fidelity between 92% and 94.3% for any large photon number NOON states [17]. In this paper we design a compact postselection-free multiphoton path-entanglement source by utilizing multiple quasi-phase-matching (QPM) PDC processes in a single $\chi^{(2)}$ nonlinear photonic crystal (NPC) seeded by a two-mode coherent state. The interference effect gives rise to possible productions of two-, three-, four-, or five-photon $|N :: M\rangle$ states. As in the schemes of Refs. [16–18], our scheme does not waste any $(N+M)$th-order photon, i.e., all the $(N+M)$-photon events contribute to the measurable $(N+M)$-photon interference, and this advantage is crucial to supersensitive phase measurement [13,14,19]. The advantage of our scheme lies in the concurrent multiple quasi-phase-matchings in the NPC, which opens up a way to integrated quantum light sources.

The remainder of the paper is arranged as follows. In Sec. II we give a description of the $\chi^{(2)}$ NPCs in our scheme and analyze the possible QPM PDC processes. In Sec. III we present the scheme for generating path-entangled states via the specific NPCs. Section IV contains a discussion and we summarize in Sec. V.

## II. DESCRIPTION OF THE $\chi^{(2)}$ NPC STRUCTURES IN OUR SCHEME

The NPC is specified as the two-dimensional (2D) QPM material in which the second-order nonlinear susceptibility $\chi^{(2)}$ is spatially modulated. Providing flexible multiple-beam or multiple-frequency nonlinear processes [20–24], the NPC attracts great interest since proposed by Berger in 1998 [25]. Here we design a 2D NPC structure that can satisfy two or three concurrent QPM conditions. Figure 1(a) shows a 2D $\chi^{(2)}$ NPC, where circularly inverted domains (with $-\chi^{(2)}$) distribute rectangularly on a $\chi^{(2)}$ background. Its reciprocal lattice is depicted in Fig. 1(b) and the reciprocal vectors $\vec{G}_{m,n}$

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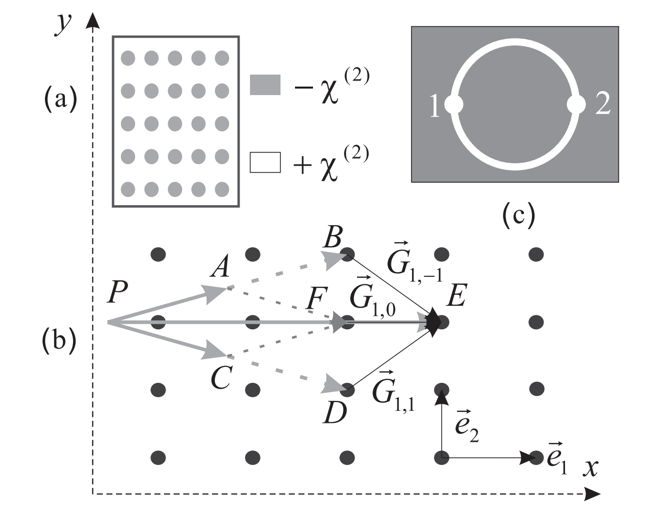


FIG. 1. (a) Schematic of a 2D $\chi^{(2)}$ NPC with a rectangular inverted domain structure. (b) Reciprocal lattice of the crystal. Thick gray arrows represent pump, signal, and idler wave vectors. Thin black arrows represent reciprocal vectors of the crystal. (c) Transverse pattern of the parametric light in the Fourier plane.

are

$$\vec{G}_{m,n} = m\vec{e}_1 + n\vec{e}_2, \quad |\vec{e}_1| = \frac{2\pi}{\Lambda_x}, \quad |\vec{e}_2| = \frac{2\pi}{\Lambda_y}, \tag{3}$$

where $\Lambda_x$ ($\Lambda_y$) is the period along the $x$ ($y$) axis of the $\chi^{(2)}$ modulation in the crystal and the subscripts $m$ and $n$ are integers. For the case of infinitely large crystal size, the dependence of the nonlinear susceptibility $\chi^{(2)}$ on the 2D spatial coordinate $\vec{r} = (x,y)$ can be written as a Fourier series [25]

$$\chi^{(2)}(\vec{r}) = \sum_{m,n} \mathcal{G}_{m,n} e^{-i\vec{G}_{m,n}\cdot\vec{r}}, \tag{4}$$

where $\mathcal{G}_{m,n}$ are the Fourier coefficients.

### A. Parametric down-conversion in a $\chi^{(2)}$ NPC

Considering a classical pump wave illuminating the NPC with a volume of $V$, we can write the interaction Hamiltonian of the PDC process as (for details see Ref. [26])

$$\hat{H}_I(t) = \varepsilon_0 \int_V d^3\vec{r}\, \chi^{(2)}(\vec{r}) \hat{E}_p^{(+)}(\vec{r},t) \hat{E}_s^{(-)}(\vec{r},t) \hat{E}_i^{(-)}(\vec{r},t) + \text{H.c.}, \tag{5}$$

where H.c. denotes the Hermitian conjugate part and $\hat{E}_s^{(-)}$ and $\hat{E}_i^{(-)}$ are the field operators of the signal and idler, respectively, expressed as

$$\hat{E}_j^{(-)}(\vec{r},t) = \int d^3\vec{k}_j E_j^*(\omega_j) e^{-i(\vec{k}_j\cdot\vec{r}-\omega_j t)} \hat{a}_j^\dagger(\omega_j,\vec{k}_j), \tag{6}$$

where $E_j = i\sqrt{\hbar\omega_j/16\pi^3\varepsilon_0 n^2(\omega_j)}$, with $j = s,i$. The complex amplitude of the pump $E_p^{(+)}$ has the form

$$E_p^{(+)}(\vec{r},t) = \int d^3\vec{k}_p E_p(\omega_p) e^{i(\vec{k}_p\cdot\vec{r}-\omega_p t)}. \tag{7}$$

Not that here we consider type-0 PDC, i.e., the signal, idler, and pump light are all $e$-polarized and hence we omit the polarization terms in $\hat{E}_j$ and $E_p$.

The evolution of the PDC process can be described in the interaction picture by the unitary operator

$$\hat{U}(t,t') = \exp\left[\frac{1}{i\hbar}\int_{t'}^{t} \hat{H}_I(\tau)d\tau\right] \tag{8}$$

and the state evolution from time $t'$ to $t$ is expressed by

$$|\psi(t)\rangle = \hat{U}(t,t')|\psi(t')\rangle. \tag{9}$$

Considering the steady-state output and that the interaction volume approaches the infinite, we may write the state evolution operator as

$$\hat{U} = \exp\left[\sum_{m,n}\chi_{m,n}\int d\omega_p \int d\omega_s \int d\omega_i \delta(\omega_s+\omega_i-\omega_p) \times \delta^{(3)}(\vec{k}_s+\vec{k}_i+\vec{G}_{m,n}-\vec{k}_s)\hat{a}_s^\dagger(\omega_s,\vec{k}_s)\hat{a}_i^\dagger(\omega_i,\vec{k}_i) - \text{H.c.}\right], \tag{10}$$

where we put $\mathcal{G}_{m,n}E_p(\omega_p)E_s^*(\omega_s)E_i^*(\omega_i)$ and all the extra constants into a $\chi$ function and take it out of the integral as it is a slowly varying function.

We can see that multiple PDC processes are possibly involved and the absolute square of $\chi_{m,n}$ determines the intensity of an individual parametric light. In principle, there is an infinite number of possible reciprocal vectors contributing to the processes; however, in general, for high values of $m$ and $n$, the corresponding $\mathcal{G}_{m,n}$ magnitudes become negligible.

### B. A NPC structure capable of three concurrent PDCs

We first consider the pump wave vector in the $x$ direction, as shown in Fig. 1(b). In this case we consider three most prominent reciprocal vectors $\vec{G}_{1,0}$, $\vec{G}_{1,1}$, and $\vec{G}_{1,-1}$. We choose the right wavelength ($|k_s| = |k_i|$) and temperature so as to satisfy three type-0 phase-matching conditions simultaneously,

$$\vec{k}_s^{\overrightarrow{PA}} + \vec{k}_i^{\overrightarrow{PA}} + \vec{G}_{1,-1}^{\overrightarrow{BE}} - \vec{k}_p^{\overrightarrow{PE}} = 0, \tag{11}$$

$$\vec{k}_s^{\overrightarrow{PC}} + \vec{k}_i^{\overrightarrow{PC}} + \vec{G}_{1,1}^{\overrightarrow{DE}} - \vec{k}_p^{\overrightarrow{PE}} = 0, \tag{12}$$

$$\vec{k}_s^{\overrightarrow{PA}} + \vec{k}_i^{\overrightarrow{PC}} + \vec{G}_{1,0}^{\overrightarrow{FE}} - \vec{k}_p^{\overrightarrow{PE}} = 0, \tag{13}$$

with the superscripts representing the vectors shown in Fig. 1(b). The above three conditions can be easily obtained by the geometry calculations via the triangles $PBE$, $PDE$, and $PAF$, respectively. We can infer the transverse distribution of the photon pair as shown in Fig. 1(c). More explicitly, the signal and idler photons satisfying Eqs. (11) and (12) are emitted to positions 1 and 2, respectively, in a collinear and beamlike way. The emitted signal and idler photons satisfying Eq. (13) transmit noncollinearly as a cone.

If using single-mode fiber collections at positions 1 and 2, in the case of single-frequency approximation guaranteed by narrow-band interferometer filters, we can write the operator

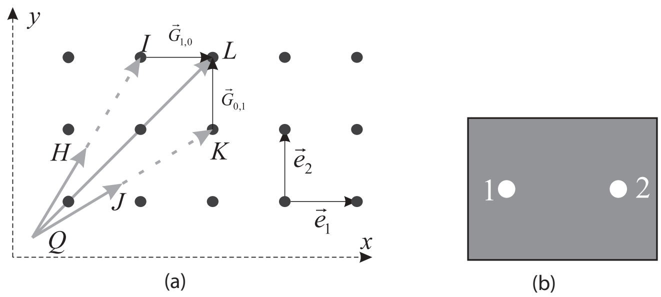


FIG. 2. (a) Schematic of the reciprocal lattice of a 2D $\chi^{(2)}$ NPC with a square inverted domain structure. Thick gray arrows represent pump, signal, and idler wave vectors. Thin black arrows represent reciprocal vectors of the crystal. (b) Transverse pattern of the parametric light in the Fourier plane.

of Eq. (10) in the single-mode form

$$\hat{U} = \exp\left(\chi_{1,-1}\eta_1\hat{a}_1^{\dagger 2} + \chi_{1,1}\eta_2\hat{a}_2^{\dagger 2} + \chi_{1,0}\eta_3\hat{a}_1^{\dagger}\hat{a}_2^{\dagger} - \text{H.c.}\right), \tag{14}$$

where $\eta_j$, with $j = 1,2,3$, is the fiber collection efficiency for the three PDC processes and we assume $\eta_1 = \eta_2$. For a symmetry crystal, $\chi_{1,-1} = \chi_{1,1}$ is guaranteed. Then if we let $\chi = \chi_{1,1}\eta_1$ and $r = \chi_{1,0}\eta_3/\chi$, we obtain

$$\hat{U} = \exp\left[\chi\left(\hat{a}_1^{\dagger 2} + \hat{a}_2^{\dagger 2} + r\hat{a}_1^{\dagger}\hat{a}_2^{\dagger}\right) - \text{H.c.}\right]. \tag{15}$$

In principle, $r$ can be any positive real number. It depends on the duty cycle, the ratio of $\Lambda_x$ to $\Lambda_y$, and fiber collection efficiencies. Moreover, a $\pi$ shift to $\hat{a}_1^{\dagger}$ or $\hat{a}_2^{\dagger}$ can make $r$ become a negative real number.

### C. A NPC structure capable of two beamlike PDCs

To realize $r = 0$ we can utilize a 2D $\chi^{(2)}$ NPC with a square inverted domain structure, whose reciprocal lattice is depicted in Fig. 2(a) and signal and idler photons are generated collinearly but noncollinearly with the pump as shown in Fig. 2(b). The reciprocal vectors $\vec{G}_{m,n}$ are

$$\vec{G}_{m,n} = m\vec{e}_1 + n\vec{e}_2, \quad |\vec{e}_1| = |\vec{e}_2| = \frac{2\pi}{\Lambda}, \tag{16}$$

where $\Lambda$ is the poling period. The pump wave vector is in the diagonal direction and again we choose right wavelength ($|k_s| = |k_i|$) and temperature so as to satisfy the following two type-0 phase-matching conditions simultaneously:

$$\vec{k}_s^{\overrightarrow{QH}} + \vec{k}_i^{\overrightarrow{QH}} + \vec{G}_{1,0}^{\overrightarrow{IL}} - \vec{k}_p^{\overrightarrow{QL}} = 0, \tag{17}$$

$$\vec{k}_s^{\overrightarrow{QJ}} + \vec{k}_i^{\overrightarrow{QJ}} + \vec{G}_{0,1}^{\overrightarrow{KL}} - \vec{k}_p^{\overrightarrow{QL}} = 0. \tag{18}$$

Through analogous calculations, we can write the single-mode state evolution operator as

$$\hat{U} = \exp\left[\chi'\left(\hat{a}_1^{\dagger 2} + \hat{a}_2^{\dagger 2}\right) - \text{H.c.}\right], \tag{19}$$

where $\chi' = \chi_{1,0}\eta_1 = \chi_{0,1}\eta_2$.

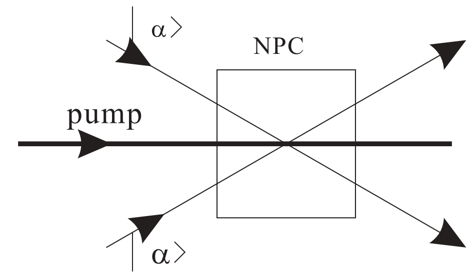


FIG. 3. Scheme for path-entanglement generation from a single NPC.

## III. SCHEME FOR GENERATING MULTIPHOTON PATH ENTANGLEMENT

Up to now we have constructed a special PDC operator given by Eq. (15) with $r$ being any real number in principle. To generate the path-entangled states $|N :: M\rangle$ we consider a PDC seeded by a two-mode coherent state as shown in Fig. 3, in which the coherent state is injected along the output path modes 1 and 2, respectively. Then the output state can be written as

$$|\Psi\rangle = \exp\left[\chi\left(\hat{a}_1^{\dagger 2} + \hat{a}_2^{\dagger 2} + r\hat{a}_1^{\dagger}\hat{a}_2^{\dagger}\right) - \text{H.c.}\right]|\alpha\rangle_1|\alpha\rangle_2. \tag{20}$$

The analytic photon number distribution of the output state can be obtained via that of the squeezed coherent state [27] and the squeezed number state [28]. However, in realistic quantum photonic experiments, PDC works in the weak interaction regime ($|\chi| \ll 1$), so in the following we write the photon number distribution in an approximate but much simpler way.

### A. Two-photon NOON state generation

Considering PDC in a weak interaction regime ($|\chi| \ll 1$) and a weak coherent state ($|\alpha|^2 \ll 1$), we expand the output state given by Eq. (20) up to a two-photon term as

$$|\Psi_2\rangle \approx |0\rangle + \alpha(|1,0\rangle + |0,1\rangle) + (\alpha^2 + r\chi)|1,1\rangle + (\alpha^2 + 2\chi)|2 :: 0\rangle + \cdots, \tag{21}$$

where we neglect higher-order terms. We can see that if we let the amplitude of state $|1,1\rangle$ be zero, i.e., $\alpha^2 + r\chi = 0$, the two-photon term of the output state will be the two-photon NOON state $|2 :: 0\rangle$. A straightforward solution is $\alpha = 0$, $r = 0$. The resulting output state is $|\Psi_2\rangle \approx |0\rangle + 2\chi|2 :: 0\rangle + \cdots$. The generation probability is the absolute square of the amplitude of the state $|2 :: 0\rangle$, i.e., $4|\chi|^2$. The generation probability of the other states in the following can be calculated in the same way and thus we will not give them for simplicity.

### B. Generation of three-photon path-entangled states

To find the condition for generating the three-photon path-entangled state we expand Eq. (20) up to three-photon term and obtain

$$|\Psi_3\rangle \approx \cdots + C_{30}|3 :: 0\rangle + C_{21}|2 :: 1\rangle + \cdots, \tag{22}$$

where we neglect lower- and higher-order terms. The coefficients are $C_{30} = \alpha(\alpha^2 + 6\chi)/\sqrt{3}$ and $C_{21} = \alpha(\alpha^2 + 2\chi r + 2\chi)$. Therefore, to generate the three-photon NOON state

$|3::0\rangle$ we need $C_{21}=0$, a possible solution of which is $r=0$, $\chi=-\alpha^2/2$, which induces $C_{30}=-2\alpha^3/\sqrt{3}$. The state $|2::1\rangle$ can be generated via the condition of $C_{30}=0$, namely, $\chi=-\alpha^2/6$, and then $C_{21}=(2-r)\alpha^3/3$.

### C. Generation of four-photon path-entangled states

To study the four-photon path-entangled states generation we expand Eq. (20) up to a four-photon term

$$|\Psi_4\rangle \approx \cdots + C_{40}|4::0\rangle + C_{31}|3::1\rangle + C_{22}|2,2\rangle + \cdots, \tag{23}$$

where $C_{40}=\sqrt{3}(\alpha^4+12\alpha^2\chi+12\chi^2)/6$, $C_{31}=(\alpha^4+3\alpha^2\chi r+6\alpha^2\chi+6\chi^2 r)/\sqrt{3}$, and $C_{22}=(\alpha^4+4\alpha^2\chi r+4\alpha^2\chi+2\chi^2r^2+4\chi^2)/2$. Therefore, the four-photon NOON state $|4::0\rangle$ can be produced in the case that $C_{31}=C_{22}=0$ and $C_{40}\neq 0$, from which we find $\chi=(1/\sqrt{3}-1/2)\alpha^2\approx 0.0774\alpha^2$, $r=-2(\sqrt{3}+1)\approx -5.46$, or $\chi=-(1/\sqrt{3}+1/2)\alpha^2\approx -1.08\alpha^2$, $r=2(\sqrt{3}-1)\approx 1.46$. Both solutions result in $C_{40}=\alpha^4/\sqrt{3}\approx 0.577\alpha^4$.

To generate the state $|3::1\rangle$ we require $C_{40}=C_{22}=0$ and $C_{31}\neq 0$. The solution is $\chi=(1/\sqrt{6}-1/2)\alpha^2\approx -0.0918\alpha^2$, $r=2(5+2\sqrt{6})\approx 19.8$, or $\chi=-(1/\sqrt{6}+1/2)\alpha^2\approx -0.908\alpha^2$, $r=2(5-2\sqrt{6})\approx 0.202$, and the resulting $C_{31}=4\alpha^4/\sqrt{3}\approx 2.31\alpha^4$.

### D. Generation of five-photon path-entangled states

By expanding Eq. (20) up to a five-photon term we have

$$|\Psi_5\rangle \approx \cdots + C_{50}|5::0\rangle + C_{41}|4::1\rangle + C_{32}|3::2\rangle + \cdots, \tag{24}$$

where $C_{50}=\sqrt{15}\alpha(\alpha^4+20\alpha^2\chi+60\chi^2)/30$, $C_{41}=\sqrt{3}\alpha(\alpha^4+4\alpha^2\chi r+12\alpha^2\chi+24\chi^2 r+12\chi^2)/6$, and $C_{32}=\alpha(\alpha^4+6\alpha^2\chi r+8\alpha^2\chi+6\chi^2r^2+12\chi^2 r+12\chi^2)/\sqrt{6}$. Following the same procedure as above, we can get the five-photon NOON state $|5::0\rangle$ by setting $C_{41}=C_{32}=0$ and $C_{50}\neq 0$. The solution is found to be $\chi=(\sqrt{2}-1)\alpha^2/6\approx 0.069\alpha^2$, $r=-2(\sqrt{2}+1)\approx -4.828$, or $\chi=-(\sqrt{2}+1)\alpha^2/6\approx -0.402\alpha^2$, $r=2(\sqrt{2}-1)\approx 0.828$, and in these cases we have $C_{50}=4\alpha^5/(3\sqrt{15})\approx 0.344\alpha^5$.

For the state $|4::1\rangle$ we find that when $\chi=(\sqrt{10}-5)\alpha^2/30\approx -0.0613\alpha^2$, $r=2(3+\sqrt{10})\approx 12.3$, or $\chi=-(\sqrt{10}+5)\alpha^2/30\approx -0.272\alpha^2$, $r=2(3-\sqrt{10})\approx -0.325$, we have $C_{50}=C_{32}=0$ and $C_{41}=-4\sqrt{3}\alpha^5/15\approx -0.462\alpha^5$. Note that the state $|3::2\rangle$ cannot be obtained in this scheme.

## IV. DISCUSSION

To show the experimental possibility of making NPCs required, we consider the fabrication of a periodically poled $LiTaO_3$ crystal. The designed NPC works for 397.5 nm $\to$ 795 nm + 795 nm. For rectangle NPC, which serves for three concurrent PDCs, we design $\Lambda_x=\Lambda_y=8.302\ \mu$m. For two concurrent PDCs, we design $\Lambda_x=\Lambda_y=5.870\ \mu$m. The working temperature is 190 °C. Here the third-order reciprocal vectors are utilized to ensure the homogeneous $\chi^{(2)}$ modulation in experiment. To get the settings of $r$, as we have shown, we can adjust the duty cycle, the ratio of $\Lambda_x$ to $\Lambda_y$, and fiber collection efficiencies. From the different spatial distributions of three concurrent PDC processes shown in Fig. 1(c) we can infer $\eta_3 \ll \eta_1$, so the solution with smaller $|r|$ is preferred in experiment. For a fixed crystal, the ratio of $\chi$ to $\alpha^2$ can be easily tuned by changing the intensity of the pump and coherent light.

## V. CONCLUSION

We have presented a scheme for generating two-, three-, four-, or five-photon path-entangled states $|N::M\rangle$, which may have applications in high-precision phase measurement and super-resolution lithography. In particular, up to five-photon NOON states can be produced. The key element of our scheme is the special NPC designed to satisfy QPM conditions for two or three PDCs simultaneously. The success of the $N$-photon entangled state generation is certified as long as we "know" $N$ photons are produced, for instance, by the final $N$-photon detections or $N$-photon absorbers. This feature, namely, a postselection-free way, is crucial in many applications where postselected entanglement sources fail. Furthermore, the integration of multiple PDCs in a single crystal makes our entangled photon source very compact. Finally, we should note that to generate more-than-five-photon path-entangled states we need the interference at beam splitters with such NPCs cascaded. Although more complicated, we believe the generation of larger photon number path-entangled states is possible with future technologies of engineering QPM structures and manufacturing waveguide devices.

## ACKNOWLEDGMENTS

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