Conditional generation of *N*-photon entangled states of light

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We propose a scheme for conditional generation of two-mode *N*-photon path-entangled states of traveling light field. These states may find applications in quantum optical lithography and they may be used to improve the sensitivity of interferometric measurements. Our method requires only single-photon sources, linear optics (beam splitters and phase shifters), and photodetectors with single-photon sensitivity.

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Quantum entanglement represents one of the most remarkable and intriguing features of the quantum mechanics. Recently, entanglement has been identified as a fundamental resource necessary for quantum information processing [1] such as quantum teleportation [2–5] and quantum computing [6]. The entangled states may also help to improve the sensitivity of interferometric measurements [7–10] and they form a key ingredient of quantum optical lithography [11–13], which employs *N*-photon entangled states to fabricate patterns on lithographic substrate with resolution $\lambda/(2N)$, where λ is the optical wavelength.

In view of these potential applications, it is highly desirable to build a source of *N*-photon path-entangled states of traveling light field,

$$|\psi_N\rangle=\sum_{k=0}^{N} c_k|k,N-k\rangle. \tag{1}$$

Here $|k,N-k\rangle$ denotes the usual Fock state with k photons in mode a and $N-k$ photons in mode b. Of particular interest could be the entangled state

$$|\psi_N^0\rangle=\frac{1}{\sqrt{2}}(|N,0\rangle+|0,N\rangle). \tag{2}$$

In the Schrödinger picture, $|\psi_N^0\rangle$ evolves in time according to $|\psi_N^0(t)\rangle=\exp(-iN\omega t)|\psi_N^0(0)\rangle$, where $\omega=2\pi c/\lambda$. We can interpret Eq. (2) as a state of a quasiparticle with energy $N\hbar\omega$ and effective de-Broglie wavelength $\lambda_{\rm eff}=\lambda/N$. This specific feature of $|\psi_N^0\rangle$ is the origin of the improvement of the resolution in quantum optical lithography [11–13].

For $N=2$, the state (2) can be generated by feeding two ports of a balanced beam splitter with single-photon Fock states, e.g., signal and idler photons generated by means of spontaneous parametric downconversion [14]. For $N>2$, however, single-photon sources and linear optics are not sufficient for deterministic preparation of state (2), and Kerr or other nonlinear media are required. Unfortunately, sufficiently strong nonlinear interactions between single traveling photons are not currently available.

Nevertheless, one may avoid the necessity of nonlinear interactions. In a recent paper, Lee *et al.* [15] showed that the states (2) can be prepared probabilistically using only Fock-state sources, linear optical elements and single-photon counting detectors. Lee *et al.* provided schemes for $N=3$ and $N=4$ but were not able to extend them to higher N. In the present paper we design a scheme for conditional generation of *arbitrary* entangled *N*-photon states (1) for *any N*. We first present a generic scheme and then, as an application, we shall consider preparation of the state (2).

The quantum-state preparation schemes, whose success is conditioned on the results of quantum measurements, have attracted considerable amount of attention recently. Schemes for probabilistic preparation of Fock states [16,17], arbitrary superpositions of Fock states of single-mode field [18,19], and Schrödinger cat states [20,21], have been found. Experimental conditional preparation of the single-photon Fock state with negative Wigner function has been reported [22]. In cavity QED, the state (1) can be generated by injection of a sequence of N suitably prepared three-level Λ-type atoms into a two-mode resonator [23]. In that scheme, one detects whether the atom leaving the resonator is in excited or ground state and the desired state (1) is prepared only if all atoms are in a ground state. Here, we design a scheme for generation of two-mode entangled states of *traveling* light field.

For our purposes it is convenient to express the target state (1) in terms of bosonic creation operators $a^\dagger$ and $b^\dagger$ acting on two-mode vacuum state,

$$|\psi_N\rangle=\sum_{k=0}^{N} d_k a^{\dagger k} b^{\dagger N-k}|0,0\rangle, \tag{3}$$

where $d_k=c_k/\sqrt{k!(N-k)!}$. The polynomial on the right-hand side of Eq. (3) can be factorized into a product of N terms linear in creation operators,

$$|\psi_N\rangle=\frac{1}{\sqrt{\mathcal{N}}}\prod_{k=1}^{N}(\cos\theta_k a^\dagger - e^{i\phi_k}\sin\theta_k b^\dagger)|0,0\rangle, \tag{4}$$

where $\mathcal{N}$ is a normalization factor and $z_k=e^{i\phi_k}\tan\theta_k$ are complex roots of the polynomial $\Sigma_{k=0}^{n} d_k z^k$. The factorization (4) suggests that we can prepare the state $|\psi_N\rangle$ from the vacuum state $|0,0\rangle$ by applying N times a nonunitary transformation

$$|\psi_k\rangle=(\cos\theta_k a^\dagger - e^{i\phi_k}\sin\theta_k b^\dagger)|\psi_{k-1}\rangle, \tag{5}$$

starting with $|\psi_0\rangle=|0,0\rangle$. We shall show that the transformation (5) can be implemented probabilistically using only

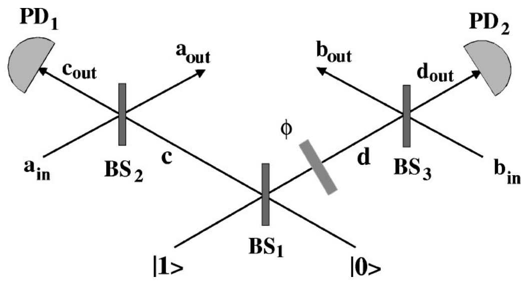

FIG. 1. Setup for probabilistic implementation of the transformation (5) consists of single photon source, three beam splitters BS_1, BS_2, BS_3, phase shifter ϕ, and two photodetectors PD_1 and PD_2.

single-photon sources, linear optical elements, and photodetectors, provided that $|\psi_k\rangle$ is an eigenstate of the operator of total number of photons $n_{ab}=a^\dagger a+b^\dagger b$,

$$n_{ab}|\psi_k\rangle=k|\psi_k\rangle. \tag{6}$$

The setup under consideration is shown in Fig. 1. In addition to modes a and b, that initially contain the state $|\psi_{k-1}\rangle$, we also need two auxiliary modes c and d, initially in a single-photon Fock state $|1,0\rangle_{cd}$. With the help of the beam splitter BS_1 with transmittance $\sin^2\theta_k$ and a suitable phase shifter we transform this state to

$$|\varphi_k\rangle_{cd}=\cos\theta_k|1,0\rangle_{cd}-e^{i\phi_k}\sin\theta_k|0,1\rangle_{cd}. \tag{7}$$

The next step consists of mixing the mode a with c at a beam splitter BS_2 while the mode b is mixed with d at BS_3. The beam splitters BS_2 and BS_3 are identical. The corresponding unitary transformation describing the operation of BS_2 and BS_3 can be thus parametrized by a single real number κ,

$$U=\exp(\kappa a^\dagger c-\kappa a c^\dagger)\exp(\kappa b^\dagger d-\kappa b d^\dagger). \tag{8}$$

The photodetectors PD_1 and PD_2 measure number of photons in the output modes $c_{\rm out}$ and $d_{\rm out}$. The transformation (5) is performed if and only if both detectors do not detect any photons. This means that PD_j need not resolve between single- and two-photon states, they should only distinguish vacuum state from any Fock state with nonzero number of photons. Efficient avalanche photodiods are suitable for this purpose [24].

If no photons are registered in output modes $c_{\rm out}$ and $d_{\rm out}$, then the photon contained in the input state $|\varphi_k\rangle_{cd}$ has been added to modes a or b. This intuitively explains the principle of operation of the scheme shown in Fig. 1. In order to provide a rigorous mathematical treatment, we rewrite the unitary transformation (8) in a disentangled form [25],

$$U=e^{-Kac^\dagger}e^{-Kbd^\dagger}(\cos\kappa)^{n_{ab}-n_{cd}}e^{Ka^\dagger c}e^{Kb^\dagger d}, \tag{9}$$

where $K=\tan\kappa$ and $n_{cd}=c^\dagger c+d^\dagger d$. The conditionally generated state $|\psi_k\rangle$ in the output modes $a_{\rm out}$ and $b_{\rm out}$ can be obtained from the transformed input state $U|\psi_{k-1}\rangle_{ab}|\varphi_k\rangle_{cd}$ by applying a projection operator

$$\Pi=\mathbb{1}_{ab}\otimes|0,0\rangle_{cd}\langle 0,0|, \tag{10}$$

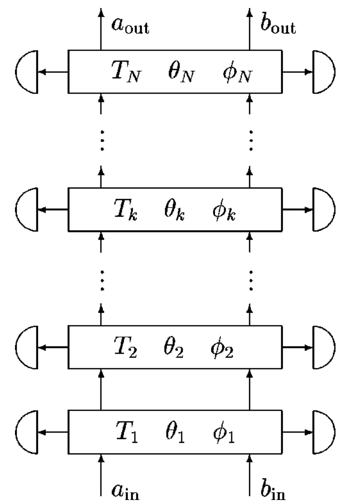

FIG. 2. Setup for probabilistic generation of N-photon path-entangled states of light. The device consists of N basic building blocks, each block represents the scheme shown in Fig. 1 and is characterized by three parameters $T_k=\cos^2\kappa_k$, θ_k, and ϕ_k.

which describes the conditioning on no photons present in the modes $c_{\rm out}$ and $d_{\rm out}$. Here $\mathbb{1}_{ab}$ is an identity operator acting on Hilbert space of modes a and b. Thus we can write

$$|\psi_k\rangle_{ab}|0,0\rangle_{cd}=\Pi U|\psi_{k-1}\rangle_{ab}|\varphi_k\rangle_{cd}. \tag{11}$$

We insert the factorized form (9) of the operator U into Eq. (11) and make use of the vacuum stability condition ${}_{cd}\langle 0,0|c^\dagger={}_{cd}\langle 0,0|d^\dagger=0$ to simplify Eq. (11) as follows:

$$|\psi_k\rangle_{ab}|0,0\rangle_{cd}=\Pi(\cos\kappa)^{n_{ab}}e^{Ka^\dagger c}e^{Kb^\dagger d}|\psi_{k-1}\rangle_{ab}|\varphi_k\rangle_{cd}. \tag{12}$$

Now we expand the two exponentials in Taylor series. Since the state $|\varphi_k\rangle_{cd}$ contains only a single photon, we have to keep only terms up to linear in annihilation operators c and d,

$$\exp(Ka^\dagger c+Kb^\dagger d)\rightarrow 1+Ka^\dagger c+Kb^\dagger d. \tag{13}$$

Inserting Eq. (13) back into Eq. (12) and taking into account that ${}_{cd}\langle 0,0|\varphi_k\rangle_{cd}=0$ we obtain

$$|\psi_k\rangle_{ab}=q_k(\cos\theta_k a^\dagger-e^{i\phi_k}\sin\theta_k b^\dagger)|\psi_{k-1}\rangle_{ab}, \tag{14}$$

where

$$q_k=(\cos\kappa)^{k-1}\sin\kappa \tag{15}$$

and we have used that the state $|\psi_{k-1}\rangle$ is an eigenstate of the total photon number operator n_{ab}. The desired N-photon entangled state (1) can be generated if we repeatedly N times apply the basic transformation (5), as illustrated in Fig. 2. There are altogether $2N$ detectors. If none of them detects any photon, then the state $|\psi_N\rangle$ is generated at the output. We assume that κ may be different for each basic building block, hence we have $3N$ parameters κ_k, θ_k and ϕ_k ($k=1,\ldots,N$) characterizing the setup shown in Fig. 2. The unnormalized conditionally generated output state reads

$$|\psi_N\rangle=\prod_{k=1}^{N} q_k(\cos\theta_k a^{\dagger}-e^{i\phi_k}\sin\theta_k b^{\dagger})|0,0\rangle. \quad (16)$$

The probability P_N of generation of the state $|\psi_N\rangle$, i.e., the yield of our scheme, can be obtained as a norm of the output state (16),

$$P_N=\mathcal{N}\prod_{k=1}^{N} q_k^2. \quad (17)$$

We can maximize the probability P_N by maximizing independently each term q_k^2. It is convenient to introduce a transmittance $T_k=\cos^2\kappa_k$. Thus we have

$$q_k^2=T_k^{k-1}(1-T_k) \quad (18)$$

and the optimal T_k maximizing q_k^2 reads

$$T_k=\frac{k-1}{k}. \quad (19)$$

Notice that the optimum beam splitter transmittance does not depend on the state that we want to generate. On inserting $q_k^2=(k-1)^{k-1}/k^k$ into Eq. (17) we obtain the optimum probability of generation

$$P_N=\mathcal{N}N^{-N}. \quad (20)$$

The normalization factor $\mathcal{N}$ has been introduced in Eq. (4).

As an example of application of our generic method, we shall consider generation of the entangled state (2). It is easy to see that this state may be written as

$$|\psi_N^0\rangle=\frac{1}{\sqrt{2}\sqrt{N!}}\prod_{k=1}^{N}(a^{\dagger}-e^{i\phi_k}b^{\dagger})|0,0\rangle, \quad (21)$$

where $\phi_k=(2k+1)\pi/N$. Upon comparing Eqs. (21) and (4) we find that $\theta_k=\pi/4$. After some algebra one obtains the optimum probability of generation

$$P_N=(N-1)!(2N)^{1-N}. \quad (22)$$

With the help of Stirling's formula we find that for large N we may approximate Eq. (22) as $P_N\approx 2\sqrt{2\pi N}(2e)^{-N}$. The yield decays exponentially with the number of photons N.

If N is even, then we can simplify our scheme and reduce the number of necessary elements by a factor of two. We write the state (2) as

$$|\psi_N^0\rangle=\frac{1}{\sqrt{2}\sqrt{N!}}\prod_{k=1}^{N/2}(a^{\dagger 2}-e^{2i\phi_k}b^{\dagger 2})|0,0\rangle. \quad (23)$$

We can generate this state if we perform $N/2$ times the transformation

$$|\psi_k\rangle=(a^{\dagger 2}-e^{2i\phi_k}b^{\dagger 2})|\psi_{k-1}\rangle, \quad (24)$$

which can be conditionally implemented with only a slight modification of the scheme shown in Fig. 1. Instead of the vacuum state $|0\rangle$, we send a single-photon Fock state $|1\rangle$ into the right input port of BS_1. After mixing on balanced beam splitter BS_1 [$\theta_k=\pi/4$] and passing through the phase shifter, the state $|\varphi_k\rangle_{cd}$ of the modes c and d reads

$$|\varphi_k\rangle_{cd}=\frac{1}{\sqrt{2}}(|2,0\rangle_{cd}-e^{2i\phi_k}|0,2\rangle_{cd}). \quad (25)$$

Similarly as before, we take the condition of detecting no photons in the output modes c_{out} and d_{out}. In this way we add two photons to the modes a or b at each step. After $N/2$ steps we thus end up with N-photon entangled state.

The calculations of the conditionally generated output state closely follow those presented above. Since the state $|\varphi_k\rangle_{cd}$ now contains two photons, we must keep quadratic terms in the expansion (13),

$$e^{Ka^{\dagger}c}e^{Kb^{\dagger}d}\to 1+K(a^{\dagger}c+b^{\dagger}d)+\frac{K^2}{2}(a^{\dagger 2}c^2+2a^{\dagger}b^{\dagger}cd+b^{\dagger 2}d^2). \quad (26)$$

Assuming that the state $|\psi_k\rangle$ is an eigenstate of total number of photons, $n_{ab}|\psi_k\rangle=2k|\psi_k\rangle$, we find that

$$|\psi_k\rangle=\frac{1}{2}(\cos\kappa)^{2k}\tan^2\kappa(a^{\dagger 2}-e^{2i\phi_k}b^{\dagger 2})|\psi_{k-1}\rangle. \quad (27)$$

The optimal transmittance of the beam splitters BS_2 and BS_3 in kth basic block is again given by Eq. (19). The probability of generation of the state (2) (i.e., the yield) reads

$$P_N'=2(N-1)!N^{1-N}. \quad (28)$$

A comparison of Eqs. (28) and (22) immediately reveals that $P_N'=2^N P_N$. The scheme where we add two photons in a single step is much more efficient, because the number of necessary measurements is halved. To be specific, for $N=4$ we have $P_4=3/256$ and $P_4'=3/16$. Lee *et al.* [15] designed schemes for generation of the state $(|4,0\rangle+|0,4\rangle)/\sqrt{2}$ with yield 3/64 and our second method improves on this result by a factor of 4.

On the way towards experimental implementation of the scheme proposed in the present paper, several obstacles have to be overcome. The main obstacle is that one needs a controlled source of single-photon Fock states. Currently available triggered single-photon sources operate by means of fluorescence from a single molecule [26] or a single quantum dot [28,27] and they exhibit very good performance. However, in our scheme we need a synchronized arrival of N single photons into N input ports of N beam splitters, which will be experimentally challenging.

Instead of those novel sources of single photons one could utilize photon pairs generated by means of spontaneous parametric downconversion (PDC). Entangled pairs of downconverted signal and idler photons have been used in many recent fundamental experiments in quantum optics and quantum information such as quantum teleportation [3], entanglement swapping [29] and measurement of Bell inequali-

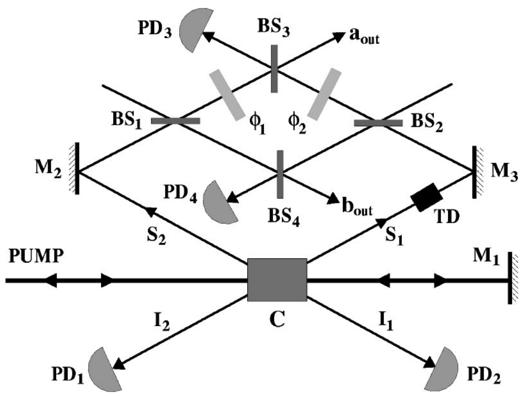

FIG. 3. Setup for generation of arbitrary two-photon entangled state: C, nonlinear crystal; M, mirrors; PD, photodetectors; TD, time delay that compensates for time interval between generation of the first and second photon pair; BS_1 and BS_2, beam splitters with mixing angles θ_1 and θ_2; BS_3 and BS_4, balanced (50:50) beam splitters; ϕ_1 and ϕ_2, phase shifters.

ties [30] and the experimental technology is well mastered. Indeed, the first experiment demonstrating the principle of quantum lithography was based on pairs of downconverted photons [13]. Note, that for our present purposes we do not require polarization-entangled photons, we only need correlated photon pairs.

With the help of pair of correlated photons one can generate any two-photon path-entangled state as follows. One has to build the interferometer according to the Figs. 1 and 2 with proper parameters (phase shifts and splitting ratios) and then feed two ports of that interferometer with signal and idler photons, respectively. With certain probability, the desired two-photon state is prepared at the two output ports. Due to its relative simplicity, this setup has several drawbacks. First, since the downconversion event is inherently random, we are not aware of the presence of the output entangled state until we detect the output photons. Second, the effective de-Broglie wavelength of the generated two-photon entangled state is the same as that of the pump beam whose photons split into signal and idler ones, hence one does not reach better spatial resolution than that achievable with the original pump beam.

A more sophisticated setup that avoids these problems is shown in Fig. 3. The proposed experimental arrangement is similar to that used in the recent entanglement-swapping experiments by Jennewein *et al.* [29]. The pump beam passes twice through the nonlinear crystal C and, with certain probability, two pairs of photons are emitted. When both the detectors PD_1 and PD_2 detect idler photons, then we know that a signal photon is present in each of the arms S_1 and S_2 [22,31]. These two signal photons then enter an interferometer consisting of four beam splitters BS_j and two phase shifters ϕ_k. If the two photodetectors PD_3 and PD_4 do not detect any photon, then the two-photon entangled state is prepared at the output modes a_{out} and b_{out}. From the results of the measurements of the four detectors PD_j we can say whether the desired entangled state has been prepared without the necessity to measure it. Moreover, the wavelengths of the signal and idler photons may be different (nondegenerate PDC), the only condition stemming from energy conservation is that the sum of signal and idler frequencies must be equal to frequency of pump beam. In terms of wavelengths we have

$$\frac{1}{\lambda_S}+\frac{1}{\lambda_I}=\frac{1}{\lambda_P},$$

hence the effective de-Broglie wavelength $\lambda_{\text{eff}}=\lambda_S/2$ of the two-photon state may be significantly smaller than the pump wavelength λ_P, provided that the idler wavelength is large enough. In quantum lithography, we would thus achieve a real improvement of the spatial resolution in comparison to using directly the pump beam.

Imagine that we would measure the output signal photons in modes a_{out} and b_{out} together with the idler ones. In this case, the setup shown in Fig. 3 can be seen as a typical four-photon coincidence experiment. The main technical obstacle in this type of experiment would be the rather low probability of spontaneous parametric downconversion. In the pulsed regime of operation the probability of generation of a single pair is typically 10^{-4} per pulse. An advantage of this low probability is that the higher photon contributions are negligible and do not spoil the generated state. This is important because the currently employed detectors with single-photon sensitivity are not able to distinguish one and more photons (see, however, Refs. [36,37]).

It should be stressed here that the four-photon coincidence experiments have been already successfully carried out in the case of entanglement swapping [29] and measurement of Bell-type inequalities for spin-1 systems [32,33]. In the latter case the observed four-photon joint count rate was about 5 counts per sec, which is high enough for collecting a sufficient amount of experimental data in a relatively short time. A very promising technology in this context is the generation of correlated pairs by means of spontaneous PDC in nonlinear optical waveguides [34,35]. Waveguides integrated on a periodically poled substrate allow for very efficient quasiphase matching [34] and the guided light is spatially confined to a small region. These factors contribute to much higher conversion efficiency than what is achievable in the experiments with bulk crystals. For instance, Tanzilli *et al.* [34] reported the conversion rate of 10^{-6} pairs per pump photon, which is four orders of magnitude higher than that typically obtained with bulk crystals. The whole experimental arrangement shown in Fig. 3 could be, in principle, built from linear and nonlinear optical waveguides and couplers on a single substrate. Such an integrated optical implementation would be very compact and could exhibit superior interferometric stability.

In view of these recent improvements we can expect that in a near future the experiments with three photon pairs will be feasible which would enable generation of three-photon entangled states in a setup similar to that shown in Fig. 3. If the probability of generation of a photon pair per pulse would be 10^{-2} then for pulse repetition rate 80 MHz [32,33] one would generate simultaneously three pairs 80 times per sec, while the probability of simultaneous generation of four pairs would be still small enough, hence the four-photon part of the generated entangled state would be negligible.

Another obstacle stems from the less-than-unit efficiencies of the single-photon detectors. Note that the currently achievable detection efficiency is about 88% [36,37]. In case of detectors PD_1 and PD_2 in Fig. 3 that should indicate the presence of idler photons, a low efficiency will only reduce the rate of generation of the entangled state. However, in our scheme we mainly need detectors to verify that no photon leaked into output auxiliary modes (all detectors in Figs. 1 and 2 and PD_3 and PD_4 in Fig. 3). If these detectors are imperfect, then the quality of the output state will be degraded and it will become a mixed state described by some density matrix ρ_{ab} [15]. However, in some applications, such as quantum lithography, this problem may be circumvented because these detectors are actually not necessary. If we do not carry out any measurements on the auxiliary modes and always accept the state prepared in modes $a_{\rm out}$ and $b_{\rm out}$, then this mixed output state can be expressed as

$$\rho_{ab}=P_N|\psi_N\rangle\langle\psi_N|+(1-P_N)\tilde{\rho}_{ab}, \tag{29}$$

where the density matrix $\tilde{\rho}_{ab}$ represents the output state when one or more photons leak into the output auxiliary modes. This implies that the operator $\tilde{\rho}_{ab}$ is supported on Hilbert space of Fock states $|k,M-k\rangle$ with $M\leqslant N-1$.

Now consider the quantum lithography. If the lithographic process is based on the *N*-photon absorption, then the absorption rate at the imaging surface will be proportional to the expectation value of normally ordered operator [11]

$$\delta=\frac{e^{\dagger N}e^{N}}{N!}, \tag{30}$$

where $e=a+b$ is the effective positive-frequency field operator. It follows that the medium will respond only to the *N*-photon part of the output state ρ_{ab} and we have

$$\mathrm{Tr}\rho_{ab}\delta=P_N\langle\psi_N|\delta|\psi_N\rangle, \tag{31}$$

which is essentially the same result as for an ideal pure output state $|\psi_N\rangle\langle\psi_N|$. The rate is only reduced by the factor P_N representing the yield of our scheme.

In summary, we have designed a universal scheme for conditional generation of an arbitrary *N*-photon path-entangled quantum state of traveling light field. The necessary resources comprise single-photon sources, beam splitters, phase shifters, and photodetectors with single-photon sensitivity. However, in certain applications, when one wishes to measure or utilize the *N*-photon coincidence rates, the conditioning is not necessary, because the desired *N*-photon part of the output state is selected automatically. The proposed method should be experimentally feasible at least for modest photon numbers $N=2$ and $N=3$, when one could utilize single photons conditionally obtained from downconverted photon pairs by detection of idler photons.

Note added in Proof: After this work was completed I learned that Zou *et al.* independently proposed an alternative scheme for conditional generation of *N*-photon entangled states [38].

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