

De Broglie wavelength of a non-local four-photon state

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Superposition is one of the most distinctive features of quantum theory and has been demonstrated in numerous single-particle interference experiments^{1–4}. Quantum entanglement⁵, the coherent superposition of states in multi-particle systems, yields more complex phenomena^{6,7}. One important type of multi-particle experiment uses path-entangled number states, which exhibit pure higher-order interference and the potential for applications in metrology and imaging⁸; these include quantum interferometry and spectroscopy with phase sensitivity at the Heisenberg limit^{9–12}, or quantum lithography beyond the classical diffraction limit¹³. It has been generally understood¹⁴ that in optical implementations of such schemes, lower-order interference effects always decrease the overall performance at higher particle numbers. Such experiments have therefore been limited to two photons^{15–18}. Here we overcome this limitation, demonstrating a four-photon interferometer based on linear optics. We observe interference fringes with a periodicity of one-quarter of the single-photon wavelength, confirming the presence of a four-particle mode-entangled state. We anticipate that this scheme should be extendable to arbitrary photon numbers, holding promise for realizable applications with entanglement-enhanced performance.

To see the origin of multi-particle interference more clearly, consider first a simple analogue to Young's double-slit experiment, that is, a Mach–Zehnder (MZ) interferometer (Fig. 1a). There, single-photon interference occurs owing to the spatial separation of two modes of propagation a1 and b1 for a single particle entering the interferometer at the first beamsplitter. Variation of the path length induces a phase shift $\Delta\varphi$ and thus gives rise to detection probabilities $P_{a2} \propto 1 + \cos\Delta\varphi$ and $P_{b2} \propto 1 - \cos\Delta\varphi$ in each of the two output modes a2 and b2 behind the exit beamsplitter. Two-photon interference occurs when a1 and b1 are the modes of propagation for a state of two indistinguishable photons, that is, a biphoton state $|\Psi\rangle = \frac{1}{\sqrt{2}}(|2\rangle_{a1}|0\rangle_{b1} + e^{i2\Delta\varphi}|0\rangle_{a1}|2\rangle_{b1})$. It represents a path-entangled two-photon state, which exhibits pure two-particle interference at the output beamsplitter. The probability to find two photons in either mode a2 or b2 then oscillates with $P_{a2,a2} \propto 1 + \cos(2\Delta\varphi)$ and $P_{b2,b2} \propto 1 - \cos(2\Delta\varphi)$, respectively, while the single-photon detection probabilities P_{a2} and P_{b2} remain constant.

In the generalized case of an N -particle interferometer, the N photons will be in a superposition of being in either mode a1 or b1, resulting in

$$|\Psi\rangle = \frac{1}{\sqrt{2}}(|N\rangle_{a1}|0\rangle_{b1} + e^{iN\Delta\varphi}|0\rangle_{a1}|N\rangle_{b1}) \quad (1)$$

In other words, the paths are entangled in photon number. Here $|N\rangle_{a1}(|N\rangle_{b1})$ indicates the N -particle Fock state in spatial mode a1 (b1), respectively, and $N = 0$ represents an empty mode. The phase modulation $N\Delta\varphi$ increases linearly with the particle number N , which is the origin of all entanglement-enhanced interferometric schemes. In particular, the N -photon detection probability in each of the interferometer outputs would vary as $P_N \propto 1 \pm \cos(N\Delta\varphi)$. It

has therefore been suggested¹⁹ that an effective de Broglie wavelength λ/N could be attributed to the quantum state. This resembles the case of a heavy massive molecule⁴ consisting of N atoms; though here the particles are in no way bound to each other.

In order to benefit best from such entanglement-enhanced interferometric techniques, it is desirable to achieve experimentally a high photon number N for states of the form of equation (1). The special case of $N = 2$ was realized both in the original Young's double-slit geometry (by using collinear production of biphoton states via parametric down conversion²⁰), and in the Mach–Zehnder configuration (by using two-photon interference to suppress unwanted single-photon contributions^{15–18}). It is commonly believed that the realization for states with $N > 2$ requires the use of nonlinear gates²¹ or N additional 'ancilla' detectors with single-photon resolution²². Unfortunately, neither of these schemes is feasible with current technologies. We demonstrate how to overcome this limitation, giving a specific example of pure four-photon interferometry.

Our proposal is based on separating photon pairs into different pairs of modes, and using two-particle interferometry rather than distinguishing photon numbers or using nonlinear beamsplitters. To achieve this goal, we exploit type-II spontaneous parametric down-conversion (SPDC)²³. An ultraviolet (UV) pulse passes through a β -barium-borate crystal, probabilistically emitting pairs of energy-degenerate polarization-entangled photons into the spatial modes a1 and a2 (Fig. 1b). The UV pump beam is reflected back at a mirror, and can thus also emit entangled photon pairs into the spatial modes b1 and b2. The set-up is aligned to generate the following maximally entangled biphoton state

$$|\Phi^+\rangle = \frac{1}{\sqrt{2}}(|H\rangle_1|H\rangle_2 + |V\rangle_1|V\rangle_2) \quad (2)$$

for each of the pairs emitted into the pairs of modes a1–a2 or b1–b2. Here H (V) indicates horizontal (vertical) polarization of the photon.

We first consider the case where only one pair of entangled photons is emitted on a double pass of the UV pulse through the crystal. There are two probability amplitudes that will contribute to the emerging two-photon state; the pair is emitted either into the pair of modes a1–a2 or into the pair of modes b1–b2. We then coherently combine the two pairs of modes at the two polarizing beamsplitters (PBSs). As the PBS transmits horizontally polarized light and reflects vertically polarized light, conditional on detecting one photon in each of the output ports, for example, a3 and a4, the biphoton state will be

$$|\Phi\rangle_{a3a4} = \frac{1}{\sqrt{2}}(|H\rangle_{a3}|H\rangle_{a4} + e^{i2\Delta\varphi}|V\rangle_{a3}|V\rangle_{a4}) \quad (3)$$

where again $\Delta\varphi$ is the phase modulation of a single photon^{15,24}. The phase $\Delta\varphi$ can be modulated by changing the position of the pump mirror. Two-photon interference fringes may now be observed by performing a projection measurement in the modes a3 and a4 into the linear polarization basis $|\pm\rangle = (1/\sqrt{2})(|H\rangle \pm |V\rangle)$. Specifically, the probability of detecting a twofold coincidence $|+\rangle_{a3}|-\rangle_{a4}$ is proportional to $P_{a3,a4} \propto 1 - \cos(2\Delta\varphi)$. These correlations are already a signature of non-locality^{25,26} of the two-photon state (Fig. 2b).

We now explain how our scheme can be generalized to four photons and even higher photon numbers. Consider the case where four photons are emitted on a double pass of the pump beam through the crystal. There are two possibilities that will contribute to an overall four-photon state: first, a double-pair is emitted on either pass of the pump beam, that is, two photon pairs are emitted into a superposition of being in the same pair of modes a1–a2 or b1–b2, respectively, or, second, one pair of photons is emitted on each pass of the pump beam, that is, into each of the modes a1–a2 and b1–b2.

We first study the double-pair emission case. In our experiment, the spectral filtering in the photon detection is much narrower than the linewidth of the UV-pump, so the two pairs cannot be treated as independent but have to be described as one four-photon²⁷. In other words, the double-pair emission results in a four-photon propagating within the mode pair a1–a2 or b1–b2. A fourfold coincidence after the two PBS, that is, detection of a single photon in each of the output ports a3, a4, b3 and b4, will either result from a $|H\rangle_{a3}|H\rangle_{a4}|V\rangle_{b3}|V\rangle_{b4}$ contribution, if the double pair is in a1–a2, or from a $|V\rangle_{a3}|V\rangle_{a4}|H\rangle_{b3}|H\rangle_{b4}$ contribution, if the double pair is in b1–b2. Temporal overlapping of both pairs of modes at the two

PBS results in $|H\rangle_{a3}|H\rangle_{a4}|V\rangle_{b3}|V\rangle_{b4} + e^{i4\Delta\varphi}|V\rangle_{a3}|V\rangle_{a4}|H\rangle_{b3}|H\rangle_{b4}$, a coherent superposition of forward and backward emission at the same time, where all the four backward emitted photons are phase shifted by the pump mirror. Introducing a path difference induces a phase shift $\Delta\varphi$, and further performing a polarization measurement in the $|\pm\rangle$ basis to achieve indistinguishability results in interference fringes with one-quarter of the single-photon wavelength. Note again that it originates from an intrinsic four-photon effect and does not involve lower-order interference. We will now show how this four-photon effect was isolated in the experiment without unwanted lower-order interference.

With the same probability as the desired double-pair emission, one photon pair is emitted into each of the mode pairs a1–a2 and b1–b2 by one pulse. The resulting coherent superposition $e^{i2\Delta\varphi}(|H\rangle_{a3}|H\rangle_{a4}|H\rangle_{b3}|H\rangle_{b4} + |V\rangle_{a3}|V\rangle_{a4}|V\rangle_{b3}|V\rangle_{b4})$ has a fixed relative phase²⁸ which is independent of the position of the pump mirror. Also in contrast to the above four-photon emission, only two-photon interference contributes to these events. These contributions can be erased by performing a proper projection measurement of the four output modes a3, b3, a4 and b4 into the $|\pm\rangle$ bases; then the number of $|+\rangle$ projections is different from the number of $|-\rangle$ projections, say $|+\rangle_{a3}|-\rangle_{a4}|+\rangle_{b3}|+\rangle_{b4}$. The overall four-photon amplitude originating from one photon in each mode then becomes $e^{i2\Delta\varphi}(|+\rangle_{a3}|-\rangle_{a4}|+\rangle_{b3}|+\rangle_{b4} - |+\rangle_{a3}|-\rangle_{a4}|+\rangle_{b3}|+\rangle_{b4})$ and thus vanishes owing to destructive interference, a four-photon equivalent to a Hong-Ou-Mandel interference²⁹.

Figure 2 compares the resulting pure four-photon interference effect (Fig. 2c) with the well-known two-photon interference (Fig. 2b) and the single-photon interference (Fig. 2a) obtained with the same set-up. The data reveal a reduction of the oscillation wavelength, from 823 ± 46 nm for the single-photon case, via 395 ± 16 nm for the two-photon case, to 194 ± 9 nm for the four-photon case. The result demonstrates that one has to treat the four-photon state as one object of the form $|\psi\rangle = \frac{1}{\sqrt{2}}(|4\rangle_{a1,a2}|0\rangle_{b1,b2} + e^{i4\Delta\varphi}|0\rangle_{a1,a2}|4\rangle_{b1,b2})$, which is similar to a ‘NOON’-state³⁰. Our four-photon state has the additional interesting property that it is non-local, as it is a superposition of four photons either in mode a1 and a2 or b1 and b2.

Nevertheless there is room for an ambiguous interpretation of the observed four-photon oscillations via two-photon interference

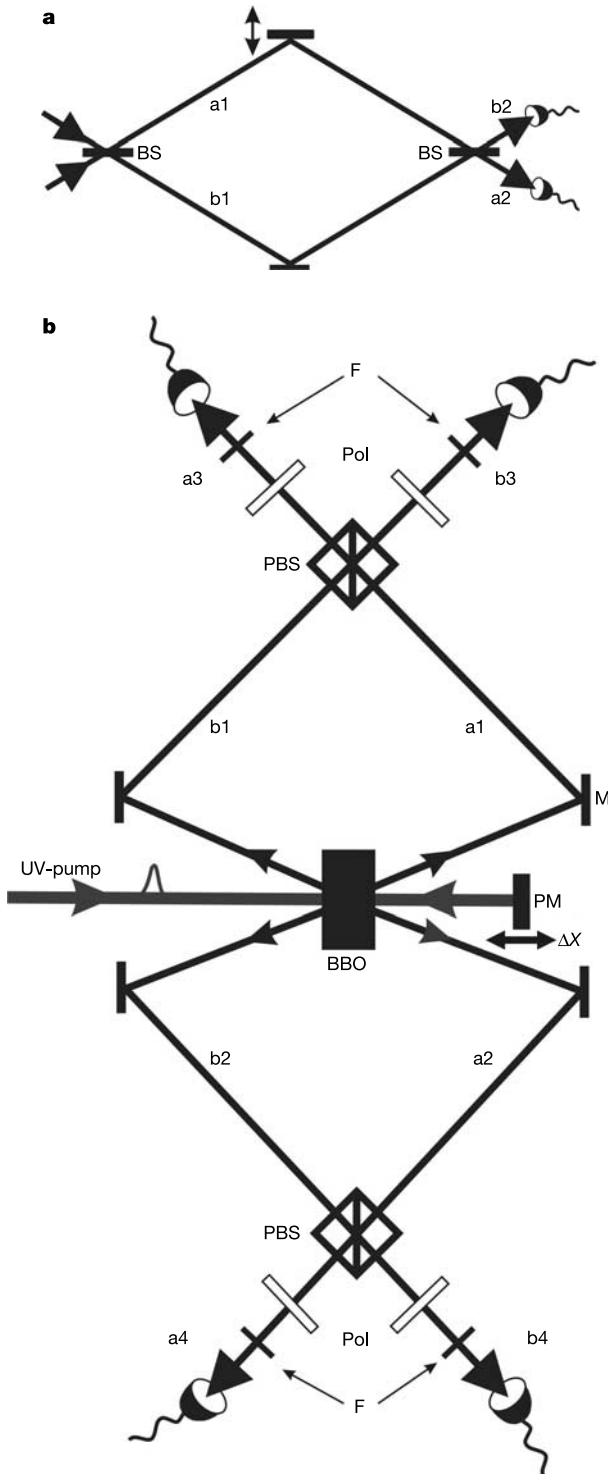


Figure 1 One-, two- and four-photon interferometry. **a**, In a two-mode Mach-Zehnder interferometer, the phase is changed by varying the path length via the position of a mirror. Single-photon interference occurs owing to the spatial separation of two possible modes of propagation a1 and b1 for a single particle entering the interferometer at the first beamsplitter (BS). Two-photon interference can be achieved when a1 and b1 are the two possible modes of propagation for a biphoton state. **b**, In our experiment, the required four-photon state is produced by type-II spontaneous parametric down-conversion (SPDC). A 200 fs pulse at a central UV wavelength of 395 nm and at a repetition rate of 76 MHz passes through a β -barium-borate (BBO) crystal, probabilistically emitting pairs of energy-degenerate polarization-entangled photons at 790 nm into the spatial modes a1 and a2. The UV pump beam is reflected back at a mirror, and might thus emit a second pair into the spatial modes b1 and b2. The probability of single-pair creation is of the order of p (in our set-up, $p \approx 10^{-2}-10^{-3}$), while the probability of creating two pairs is proportional to p^2 . Filters (F) of 3 nm bandwidth coupling into single-mode fibres in front of each detector enable good temporal and spatial overlap of the photon wavepackets at the polarizing beamsplitters (PBSs). The UV pump is reflected by the pump mirror PM, which is mounted on a computer-controlled translation stage. By scanning the position of PM with a step size of 1 μ m and performing fine adjustment of the position of M, we achieved the temporal overlap of modes a1 and b1, and of modes a2 and b2. An additional piezo translation stage is used to move the pump mirror PM and to perform a change of the phase between four photons emitted into modes a1 and a2 relative to the four photons emitted into b1 and b2. The detection of the spatially separated four-photon coincidences behind a 45° polarizer (Pol) while varying the position of PM leads to the observed interference fringes.

effects. However, this loophole can be definitely closed by the following experiment, in which we observe the desired four-photon interference without any signature of lower-order interference effects. If the assumption that the fourfold coincidence pattern of Fig. 2 could be reduced to two-photon interference were correct,

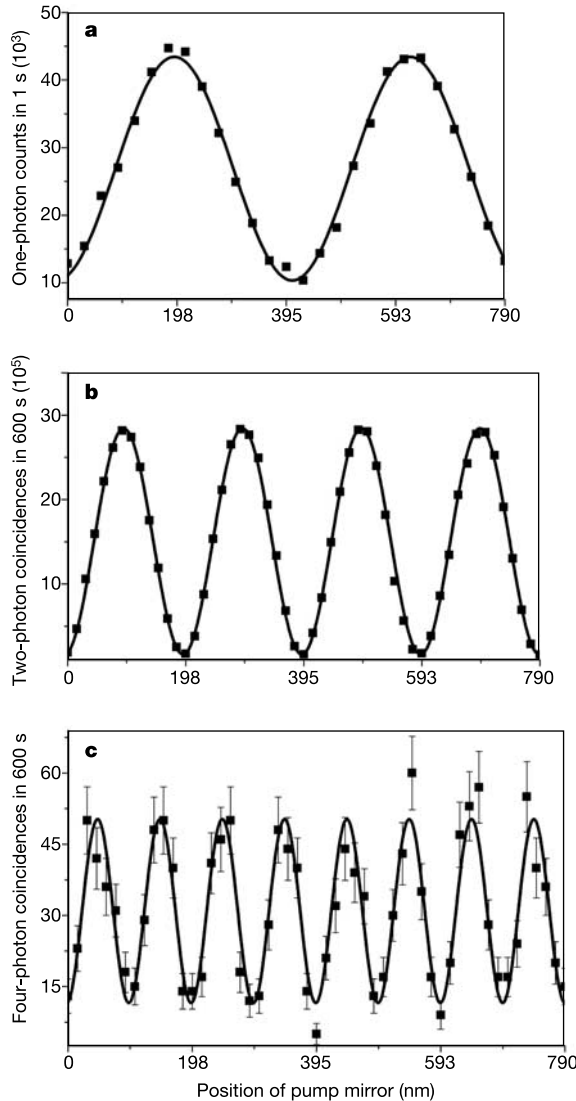


Figure 2 Experimental demonstration of pure one-, two- and four-photon interference. The two- and four-photon interference is recorded simultaneously, whereas for the one-photon interferometry the pulsed laser has been switched from mode-locking to continuous-wave (c.w.) mode. **a**, The single photon rate in mode a3 after performing a projection measurement in the linear polarization basis $|\pm\rangle = (1/\sqrt{2})(|H\rangle \pm |V\rangle)$. For this interference pattern, the pump laser is used in the c.w. mode at 790 nm (instead of the mode-locked frequency-doubled mode at 395 nm). A Mach-Zehnder configuration for modes a1–b1 arises for light scattered from the BBO crystal when passing through the crystal. By moving the pump mirror, interference fringes appear for single photons with a central wavelength of 790 nm, which corresponds to the down-converted photons. Note that, owing to the back reflection of the pump beam, the change in the optical path is twice as large as in the position of the pump mirror. **b**, The two-photon coincidence rate corresponding to the detection in mode a3 and a4 after projecting onto $|\pm\rangle_{a3}|\pm\rangle_{a4}$. **c**, A demonstration of how performing a projection onto $|\pm\rangle_{a3}|\pm\rangle_{a4}|\pm\rangle_{b3}|\pm\rangle_{b4}$ results in pure four-photon interference owing to projection onto the (non-local) path-entangled four-photon state $|\psi\rangle = \frac{1}{\sqrt{2}}(|4\rangle_{a1,a2}|0\rangle_{b1,b2} + e^{i4\Delta\varphi}|0\rangle_{a1,a2}|4\rangle_{b1,b2})$. The visibility of the observed four-photon oscillations, defined as $V = (I_{\max} - I_{\min}) / (I_{\max} + I_{\min})$, is approximately 61%. The error bars in Figs 2 and 3 are defined as the square root of the observed fourfold coincidence.

then a suppression of the two-photon amplitudes should also lead to a suppression of the fourfold oscillation. The experimental data clearly rules out this scenario (Fig. 3). In this experiment, the initial states are prepared such that a forward emitted pair is in state $|\Phi^+\rangle$ as before, while a backward emitted pair is in the state $|\Psi^-\rangle = \frac{1}{\sqrt{2}}(|H\rangle_{b1}|V\rangle_{b2} - |V\rangle_{b1}|H\rangle_{b2})$. A superposition of these states can never result in any two-photon interference owing to the bilateral parity check²⁴ performed by the two PBSs: for the $|\Phi^+\rangle$ -Bell state both photons are always transmitted or reflected, whereas for the $|\Psi^-\rangle$ -Bell state one photon is always transmitted and one is reflected, such that no interfering amplitudes for twofold detection events can build up. Only a double-pair emission on each side, where a four-photon is emitted either forwards or backwards, contributes to the four-photon state after the PBS and gives rise to pure four-photon interference. In both cases of a “ $|\pm\rangle_{a3}|\pm\rangle_{a4}|\pm\rangle_{b3}|\pm\rangle_{b4}$ ”-projection or a “ $|\pm\rangle_{a3}|\pm\rangle_{a4}|\pm\rangle_{b3}|\pm\rangle_{b4}$ ”-projection, all possible two-photon coincidence detections do not show any oscillations (Fig. 3a), while the four-photon coincidence rate oscillates with $P_{a3,a4,b3,b4} \propto 1 - \cos(4\Delta\varphi)$ or $P_{a3,a4,b3,b4} \propto 1 + \cos(4\Delta\varphi)$, respectively, that is, a fourfold reduction in wavelength (Fig. 3b). This complete absence of two-photon interference effects underlines the true four-photon character of the observed interference. Note that this is in contrast to the first case, in which a projection onto $|\pm\rangle_{a3}|\pm\rangle_{a4}|\pm\rangle_{b3}|\pm\rangle_{b4}$ would not eliminate the two-photon interference and thus does not

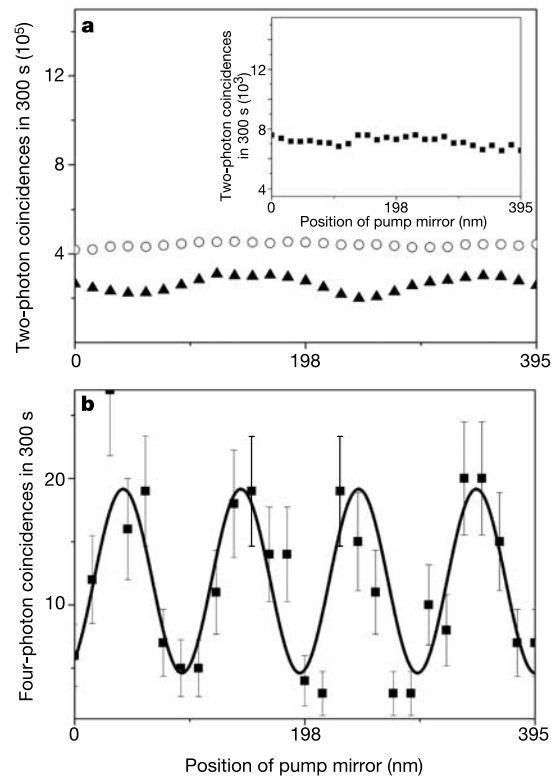


Figure 3 Pure four-photon interference without any two-photon interference. This can be observed when different entangled input states are used in forward and backward emission, specifically $|\Phi^+\rangle_{a1,a2}$ and $|\Psi^-\rangle_{b1,b2}$. Owing to the bilateral parity action of the PBS, only a four-photon emitted in a superposition of either forward or backward direction can contribute to the overall state. **a**, No interference is obtained from any possible twofold coincidence measurement $P_{a3,a4}$ (open circles), $P_{a4,b3}$ (filled triangles) and $P_{a4,b4}$ (inset). **b**, In the case of a $|\pm\rangle_{a3}|\pm\rangle_{a4}|\pm\rangle_{b3}|\pm\rangle_{b4}$ -projection, pure four-photon oscillations can be observed with detection probability $P_{a3,a4,b3,b4} \propto 1 - \cos(4\Delta\varphi)$. The corresponding measured visibilities of the two-photon curves are approximately 4% for $P_{a3,a4}$, 19% for $P_{a4,b3}$ and 7% for $P_{a4,b4}$, which is clearly below the four-photon fringe visibility of approximately 61%.

result in the desired $\lambda/4$ de Broglie wave effect³¹. In principle, this can be extended to higher-order interference effects because, obviously, when more than two double-pairs are emitted from the crystal the suppression of all lower-order interferences can be achieved by a proper projection analogous to the $N = 4$ case.

The method that we use here allows the generation of four-photon states and their subsequent utilization in pure four-particle interferometry. The result clearly confirms the theoretical expectation that the de Broglie wavelength of a four-photon state is one-quarter that of a single photon, thus leading to the general rule $\lambda(N) = \lambda(1)/N$. This overcomes the resolution limit of state-of-the-art two-particle interferometry, and opens new possibilities for multi-particle interference in fundamental quantum experiments and in applications such as quantum metrology. It is important to note that, in principle, this scheme can be extended to higher particle numbers if more spatial modes are involved. The actual limitation due to low count rates may eventually be overcome with the next generation of entangled photon sources and detectors. □

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Super-resolving phase measurements with a multiphoton entangled state

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Interference phenomena are ubiquitous in physics, often forming the basis of demanding measurements. Examples include Ramsey interferometry in atomic spectroscopy, X-ray diffraction in crystallography and optical interferometry in gravitational-wave studies^{1,2}. It has been known for some time that the quantum property of entanglement can be exploited to perform super-sensitive measurements, for example in optical interferometry or atomic spectroscopy^{3–7}. The idea has been demonstrated for an entangled state of two photons⁸, but for larger numbers of particles it is difficult to create the necessary multiparticle entangled states^{9–11}. Here we demonstrate experimentally a technique for producing a maximally entangled three-photon state from initially non-entangled photons. The method can in principle be applied to generate states of arbitrary photon number, giving arbitrarily large improvement in measurement resolution^{12–15}. The method of state construction requires non-unitary operations, which we perform using post-selected linear-optics techniques similar to those used for linear-optics quantum computing^{16–20}.

Our goal is to create the state

$$|N :: 0\rangle_{a,b} \equiv \frac{1}{\sqrt{2}} (|N, 0\rangle_{a,b} + |0, N\rangle_{a,b}) \quad (1)$$

which describes two modes a, b in a superposition of distinct Fock states $|n_a = N, n_b = 0\rangle$ and $|n_a = 0, n_b = N\rangle$. This state figures in several metrology proposals, including atomic frequency measurements⁴, interferometry^{3,6,7}, and matter-wave gyroscopes⁵. In these proposals the particles occupying the modes are atoms or photons.

The advantage for spectroscopy can be seen in this idealization: we wish to measure a level splitting $H_{\text{ext}} = \epsilon_{ba} b^\dagger b$ between modes b and a using a fixed number of particles N in a fixed time T . We could prepare N copies of the single-particle state $(|1, 0\rangle_{a,b} + |0, 1\rangle_{a,b})/\sqrt{2}$ and allow them to evolve to the state $|\phi\rangle \equiv (|1, 0\rangle_{a,b} + \exp[i\phi]|0, 1\rangle_{a,b})/\sqrt{2}$, where $\phi = \epsilon_{ba} T/\hbar$. Measurements of $A_1 \equiv |0, 1\rangle\langle 1, 0| + |1, 0\rangle\langle 0, 1|$ on this ensemble give $\langle A_1 \rangle = \cos(\phi)$ with phase uncertainty at the shot-noise limit, $\Delta\phi = 1/\sqrt{N}$. In contrast, under the same hamiltonian $|N :: 0\rangle$ evolves to $(|N, 0\rangle + \exp[iN\phi]|0, N\rangle)/\sqrt{2}$. If we measure the operator $A_N \equiv$