Sequential Path Entanglement for Quantum Metrology

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Path entanglement is a key resource for quantum metrology. Using path-entangled states, the standard quantum limit can be beaten, and the Heisenberg limit can be achieved. However, the preparation and detection of such states scales unfavourably with the number of photons. Here we introduce sequential path entanglement, in which photons are distributed across distinct time bins with arbitrary separation, as a resource for quantum metrology. We demonstrate a scheme for converting polarization Greenberger-Horne-Zeilinger entanglement into sequential path entanglement. We observe the same enhanced phase resolution expected for conventional path entanglement, independent of the delay between consecutive photons. Sequential path entanglement can be prepared comparably easily from polarization entanglement, can be detected without using photon-number-resolving detectors, and enables novel applications.
confirming the power of SPE states as a resource for quantum metrology. We generate our SPE states by feeding a polarization-entangled photon pair into a modified Mach-Zehnder interferometer (MZI). More generally, we establish that higher order SPE states could be deterministically produced from an initial resource of GHZ polarization-entangled states by means of optical switching.

**Results**

**Sequential path entanglement.** The archetypal path-entangled state is the N00N state \(|N0:0N\rangle = |N\rangle_a |0\rangle_b + e^{i\phi} |0\rangle_a |N\rangle_b\rangle \) (ignoring normalisation here, and below), in which N photons are superposed between all propagating in one arm, a, of an MZI, and all propagating in the other arm b (see Fig. 1 (a)), which accrues a phase rotation \(N\phi\), where \(\phi\) is the relative phase between the two arms of the MZI. The advantage of this path-entangled state for metrology can be understood using the concept of the photonic de Broglie wavelength \(\lambda/N\), with \(\lambda\) the single-photon wavelength.

The scheme that is most commonly used to produce approximate N00N states is shown in Fig. 1 (a). Two Fock states \(|N/2\rangle\) are made to interfere on the first beam splitter (BS) of an MZI. For \(N = 2\), Hong-Ou-Mandel (HOM) interference directly generates the required photon-bunched \(|20:02\rangle\) state. Unfortunately, for \(N > 2\), generating a genuine N00N state by HOM interference in this form no longer works: the resulting state is instead a superposition of \(|N0:0N\rangle\) with an undesired contribution \(|\Omega(N)\rangle\), which decreases the associated measurement precision. Nagata et al. prepared a 4-photon N00N state with \(|\Omega(4)\rangle \propto |22\rangle\), by interfering 2-photon Fock states. States of this form are known as Holland-Burnett states, and, while they are interesting for lossy phase estimation, they cannot achieve the Heisenberg limit. It has been proposed that \(|\Omega(N)\rangle\) can be suppressed by using feedforward, however, this proposal has not been implemented yet.

Multiphoton SPE states can be generated without suffering from the complications described above that make N00N states difficult to produce for \(N > 2\). We begin with a polarization-entangled GHZ state \(|\text{GHZ}(N)\rangle = |H\rangle_1 |H\rangle_2 \ldots |H\rangle_N + |V\rangle_1 |V\rangle_2 \ldots |V\rangle_N\rangle\) in which \(|H\rangle_i \ (|V\rangle_i\rangle\) denotes a horizontally (vertically) polarized photon in spatial mode \(i = 1, 2, \ldots, N\). The conversion of this GHZ state into an SPE state is schematically shown in Fig. 1 (b). Each photon from the GHZ state is delayed with respect to the next and then all are transferred into a single spatial mode. In our experiments we combine the photons probabilistically, using a beam splitter, but in general this step could be done deterministically by means of a fast optical switch, such as a Pockels cell. Finally, path entanglement is created by directing the photons one by one onto a polarizing beam splitter followed by a half-wave plate at one output mode, which applies the operation \(|H\rangle \rightarrow |1\rangle_a |0\rangle_b; |V\rangle \rightarrow |0\rangle_a |1\rangle_b\rangle\). This recipe allows the deterministic conversion of GHZ entanglement into SPE states within the MZI of the form

\[
|\text{SPE}(N)\rangle = \prod_{j=1}^{N} |1\rangle_{a,t_j} |0\rangle_{b,t_j} + \prod_{k=1}^{N} |0\rangle_{a,t_k} |1\rangle_{b,t_k}
\]

where now each temporal mode \(t_j\) contains a single path-entangled photon. Each of the \(N\) photons in mode \(b\) picks up the interferometer phase \(\phi\), and so this state also exhibits super-resolution, like the N00N state (see Methods). However, the time separation of the...
photons means that photon-number resolving detectors are not required to identify the N-photon events containing the enhanced metrological signal. In principle, 'on-off' detectors such as commercially-available avalanche photodetectors can be used, since there is only a single particle in each time bin. Additionally, if the MZI phase is varying in time — as would be typical in pump-probe type experiments — the Fourier components of the phase evolution can be accessed efficiently via the transmission statistics of SPE states by tuning the time delay $T = t_{j+1} - t_j$ into resonance with a component of interest (see Methods).

**Experimental setup.** Our experiment is shown schematically in Fig. 2. It contains two parts: the photon source (Fig. 2 (a)) which produces polarization-entangled photon pairs, and the modified MZI (Fig. 2 (b)). In the experiment, as shown in Fig. 2 (a), type-II spontaneous parametric down conversion (SPDC) is employed to generate a polarization-entangled state $|H\rangle_{P_1} |H\rangle_{P_2} + |V\rangle_{P_1} |V\rangle_{P_2}$ (see Methods). A translation stage with µm precision adds an adjustable delay $\Delta L_1$ into path $P_1$, and then $P_1$ and $P_2$ are combined at a 50:50 beam splitter BS1 and the resulting polarization-entangled state is coupled into an optical fibre in mode $P_3$ for delivery to the MZI. We note that BS1 passively combines the photons into the same spatial mode in a probabilistic way, which allows us to study several delay settings, including $\Delta L_1 = 0$. But for a sufficiently large delay $\Delta L_1 > \xi$, where $\xi$ is the single photon coherence length, the photons are well-separated and could be deterministically combined using a fast optical switch, allowing the efficient conversion of higher-photon-number GHZ states into SPE states. The state emerging from the fibre is converted into an SPE state of the form (2) at a polarizing beam splitter, and a µm-precision piezoelectric actuator mounted on a µm-precision translation stage is used to control the optical path length difference $\Delta L_2$ within the modified MZI.

To find the position $\Delta L_2 = 0$ we first maximise the visibility of single-photon interference in the MZI (see Fig. 2 (d)). For these measurements BS1 is replaced by a polarizer oriented at 45°, so that photons emitted from the source into path $P_1$ are delocalised at the polarizing beam splitter, producing a fringe pattern in the singles count rates recorded in the paths $P_7, P_8$, as $\Delta L_2$ is scanned (photons in path $P_2$ are simply dumped). The full width at half maximum of the pattern provides a measurement of the single-photon coherence length $\xi \approx 130\ \mu m$, consistent with the result deduced from the measured HOM interference dip shown in Fig. 2 (c) (see Methods). We then set $\Delta L_2 \approx 0$ and scan the piezoelectric actuator in steps of 10 nm to observe the fringe spacing, which matches the single photon de Broglie wavelength (simply the optical wavelength) $\lambda \approx 810\ nm$ (see Fig. 3 (a)).

Next, we verify phase super-resolution with a standard N00N state by setting $\Delta L_1 = 0$, so that both photons emitted from the source are temporally overlapped in the MZI. As shown in Fig. 2 (e), the coincidence count rate at detectors $D_1$ & $D_2$ exhibits an interference pattern with a broader envelope than the single photon interference pattern. This is due to spectral entanglement of the photon pairs emitted by our source, which introduces temporal correlations that are limited only by the coherence time of the downconversion pump laser, extending significantly beyond the single-photon coherence time (see Methods for details). We comment that to our knowledge this is the first time the extended interference pattern of a [20:02] state generated with a spectrally anti-correlated photon pair source has been fully mapped out. The fine-grained scan of the region around $\Delta L_2 = 0$ in Fig. 3 (b) shows the expected half-period fringes.

**Figure 2 | Schematic experimental setup and one- and two-photon interference.** (a) Preparation of coaxial polarization-entangled state. Polarization entangled photon pairs are generated by type II degenerate SPDC in a nonlinear crystal. BS1 serves as a probabilistic optical mixer coupling both entangled photons into path $P_3$. $\Delta L_1$ is tuned by scanning the position of a prism mounted on a translation stage. (b) A modified MZI. (c) The two-photon interference pattern measured in paths $P_1$ and $P_2$ exhibits a characteristic dip at $\Delta L_1$ is scanned, indicative of HOM interference at BS1. (d), (e) One- and two-photon interference patterns. BBO: β-barium-borate crystal; COMP: HWP and BBO (1 mm); POL: polarization plate; PIEZO: piezoelectric actuator.
associated with the reduced de Broglie wavelength of the \( |20:02\rangle\) state.

**Independence of the separation between photons.** To verify that SPE states have the same reduced de Broglie wavelength as the \( |N00N\rangle\) state, independent of the delay between successive photons entering the MZI, we carried out experiments for \( \Delta L_1 = 200 \) μm and 10,000 μm. The setting \( \Delta L_1 = 200 \) μm > \( \xi \) is larger than the single-photon coherence length but still less than the coherence length of the pump laser \( \xi_{\text{pump}} \approx 300 \) μm, while \( \Delta L_1 = 10,000 \) μm is significantly larger than \( \xi_{\text{pump}}\). Fig. 4 shows the interference patterns in the coincidence rates for detectors \( D_1, D_2 \) produced by scanning \( \Delta L_2 \). For each of the scans, the periodicity is always about 405 nm, half of the single-photon wavelength \( \lambda = 810 \) nm, which confirms that SPE states retain the phase super-resolution of a conventional \( |N00N\rangle\) state, even when consecutive photons are separated well beyond their individual coherence lengths. It is clear that the coincidence rates fall as \( \Delta L_1 \) is increased, but this simply represents a reduction in the SPE preparation efficiency. This is because for \( \Delta L_1 < \xi \), HOM interference at BS1 boosts the probability that photon pairs are coupled into our MZI, whereas for \( \Delta L_1 \geq 150 \) μm there is no longer HOM

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**Figure 3** | Characteristics of one- and two-photon interference. (a) Single-photon count rate in path \( P_7 \) (red solid circles) and at \( P_8 \) (blue solid circles) as the delay \( \Delta L_2 \) is varied. Both output modes of the interferometer are directed to similar detection set-ups, which provides a consistency check. (b) Two-photon coincidence rates at detectors \( D_1, D_2 \) (red solid circles) and at detectors \( D_3, D_4 \) (blue solid circles) versus \( \Delta L_2 \); events with a time separation within \( \Delta L_2/\xi \) are considered as coincident.

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**Figure 4** | Two-photon interference envelope and fringes versus interval \( \Delta L_2 \). Error bars are given by Poissonian statistics. (a), (b) \( \Delta L_1 = 0 \) μm. (c), (d) \( \Delta L_1 = 200 \) μm. (e), (f) \( \Delta L_1 = 10,000 \) μm.
interference. In addition, the fibre coupling efficiency drops for large $\Delta l_{1}$. In particular, the setting $\Delta l_{1} = 10,000 \mu m$ exceeds the Rayleigh range defined by our fibre, causing a significant drop in the coincidence rate for this measurement. However, the peak visibilities of the patterns in parts (b)-(f) of Fig. 4 remain high throughout the scan, at 0.98 $\pm$ 0.01, 0.95 $\pm$ 0.03 and 0.98 $\pm$ 0.03, respectively. From this we conclude that the quality of the bi-photon interference is independent of $\Delta l_{1}$, and the reduced photonic de Broglie wavelength is independent of the distance between photons in an SPE state.

Discussion

We have introduced SPE states as an alternative resource for quantum-enhanced optical interferometry. Conventional NOON states are difficult to produce, and photon-number-resolving detectors are required to extract a metrological signal. While multiplexed on-off detectors can be realized easily, they only count photon number approximately, and are therefore not suitable for metrological applications. By contrast, SPE states can be generated deterministically from GHZ states, which are easier to produce than NOON states. Photon-number-resolving detectors are also not needed, since photons can be detected selectively. In practice, the performance of photodetectors is critical to achieving a metrological advantage with NOON states, and this remains the case for SPE states also. However, realistic quantum interferometry could be realised using recently developed highly efficient, fast, detectors.

Many experiments have shown that two photons arriving simultaneously make a substantial difference in second harmonic generation, Kerr-like interactions and Rydberg atomic gases. This detrimental effect makes phase estimation modeling complicated. Furthermore, quantum metrology is often motivated by the need to probe refractive index variations in samples with extreme sensitivity to optical radiation. NOON states can excite 2-photon or multiphoton transitions in a sample that scatter light out of the interferometer, and such transitions can ionise quantum systems and damage biological molecules. The influence of multiple photons will become stronger if more photons are present at one time, as could be the case with high-$N$ NOON states, which are the ultimate goal of this avenue of research. By contrast even large-$N$ SPE states can excite only single-photon transitions because the constituent photons arrive one at a time.

It is also notable that SPE states can be used to extract the harmonic spectrum of a time-varying phase: if the consecutive time bins in an SPE state are separated by a constant delay $T$, the phase is sampled periodically, producing a signal proportional to the amplitude of the Fourier component of the dynamical phase at a frequency $f = 1/T$ (see Methods).

We experimentally confirmed that 2-photon SPE states posses the same phase super-resolution as 2-NOON states. Unlike schemes for achieving super-resolution via post-selection on classical light, this super-resolution is of quantum origin, and SPE states therefore represent a convenient and powerful platform for quantum-enhanced phase sensing via optical interferometry.

Methods

Preparation of polarization-entangled photonic source. As shown in Fig. 2 a CW semiconductor laser produces a blue beam (power 34.5 mW, waist 100 $\mu m$, central wavelength 405 nm) that pumps type-II degenerate SPDC in a 2 mm $\beta$-barium borate (BBO) crystal, producing pairs of photons with central wavelength 800 nm. The down-converted signal and idler photons have different propagation velocities and travel along different paths inside the crystal due to its birefringence. The resulting walk-off effects are compensated by a combination of a half-wave plate and a 1 mm BBO crystal in each arm. Photons are then coupled into single mode fibres and we prepare a polarization-entangled state $|H1\rangle |V2\rangle + |V1\rangle |H2\rangle$. The state is further transformed to be $|H1\rangle |V2\rangle + |V1\rangle |H2\rangle$, by placing a half-wave plate with 45° in mode $P_{2}$. The singles count rates measured directly in each of $P_{1}, P_{2}$ are about 80,000 per second, and the coincidence rate is about 20,000 per second. The visibilities for the polarization correlations are about 98% in the $|H\rangle |V\rangle$ basis and 93% for the $|+45\rangle|\pm45\rangle$ basis. This shows that we have produced a high-quality polarization-entangled photonic source.

Success rate for combination of photon pairs at BS1. When $\Delta l_{1} \approx \zeta$, the photons do not overlap at BS1 and each photon has the same probability to be transmitted or reflected. Thus, the photon state in $P_{1}$ can be a single-photon state $|H\rangle$ or $|V\rangle$ with probability 0.5 respectively, or the maximally polarization-entangled state with probability 0.25 (the rest of the time, $P_{2}$ contains no photons). When $\zeta > l_{c}$, HOM interference suppresses the single photon probabilities, boosting the probability for coupling both photons into $P_{3}$ up to 1/2 for $\Delta l_{1} = 0$. HOM interference with a visibility of 94.5 $\pm$ 0.4% is shown in Fig. 2 (c), obtained by scanning $\Delta l_{1}$ and measuring the coincidence rate at two output modes $P_{2}$ and $P_{3}$ (Fig. 2 (a)). The high visibility certifies good spatial and temporal overlap for the entangled photon pairs emitted by our source. A Gaussian fit to the HOM dip in Fig. 2 (c) yields the single-photon coherence length $\zeta \approx 126 \mu m$.

Single- and bi-photon coherence time. Here we briefly outline the origin of the different widths of the single and bi-photon interference patterns shown in parts Fig. 2 (d) and (e). We consider that our downconversion source creates photon pairs in the entangled state

$$|\psi\rangle = \frac{\zeta (\alpha \alpha')}{(\alpha + \alpha \alpha' + \alpha' \alpha)} \alpha d\alpha \alpha' d\alpha' ,$$

where $\zeta$ is the joint spectral amplitude for emitting signal and idler photons with frequencies $\omega + \omega_{0}, \omega' + \omega_{0}$, with $\omega_{0}$ the centre frequency of the downconversion. $\zeta$ generally extends over a broad bandwidth set by the phasematching conditions in the downconversion crystal, but energy conservation enforces tight frequency anti-correlations, with a correlation width equal to the pump laser bandwidth.

For the single photon interference measurement (Fig. 2 (d)) we dump photons $P_{2}$, leaving the photons in $P_{3}$ in the mixed state $\rho = \rho(\alpha \alpha') \alpha \alpha' d\alpha \alpha' \alpha d\alpha$ at the two output ports $P_{2}, P_{3}$ (postselected on no photon in the other port), and the single photon count probability is $P = \rho(\alpha \alpha') \alpha d\alpha \alpha'$, where $\rho(\alpha \alpha') = \rho(\alpha') \alpha d\alpha \alpha'$ is the joint spectral intensity. This exhibits fringes with period $\lambda = 2\pi c / \Delta \lambda_{2}$ and visibility given by $V(\Delta \lambda_{2}) = \sqrt{\rho(\alpha \alpha') \rho(\alpha' \alpha)} / \rho(\alpha \alpha')$, where $g(\omega) = F_{\rho(\alpha \alpha')}(\omega) / F_{\rho(\alpha \alpha')}$. The two-dimensional Fourier transform of the joint spectral intensity $|\psi(\alpha, \alpha')|^{2}$ the tight frequency anti-correlations between the photons produce strong temporal correlations such that the width of $\Delta \lambda_{2} / c$, $\Delta \lambda_{2}$ is proportional to the coherence length $\Delta \lambda_{1}$ of the pump laser.

Super-resolution with SPE states. To convert NOON states into SPE states we simply introduce a delay $\Delta l_{2}$ between photons, which for the bi-photon case corresponds to the transformation $|\psi(\alpha, \alpha')\rangle \rightarrow |\alpha \alpha'\rangle^{\Delta \lambda_{2} / c} / \sqrt{\Delta \lambda_{2}}$. However, as described above, the bi-photon interference visibility depends only on the joint spectral intensity, which is unaffected by this phase rotation. This is why our SPE state retains phase super-resolution for all values of $\Delta l_{2}$. For higher photon number states with $N > 2$ the visibility is determined by higher order correlation functions, but the coincidence rate is always invariant under phase rotations of the joint spectrum, so the metrological power of SPE states is independent of the delay between photons.

Extracting dynamical phase information with SPE metrology. An interesting feature of SPE states is that the photons sample the joint phase over a set of discrete times $\lambda_{i}$ meaning that the metrological signal is of the form $V \cos(\Phi)$, where $V$ is a visibility, and where $\Phi = \sum_{i=1}^{N} \phi(t_{i})$. Substituting in the Fourier relation $\phi(t) = (2\pi)^{-1/2} \int \phi(\omega) e^{i\omega t} d\omega$, where $\phi$ is the spectrum associated with the time-varying phase (notice that here $\omega$ is the Fourier conjugate of $t$, and not an optical frequency), we obtain

$$\Phi = \int \Delta \cos(\phi)d\omega = \Delta \cos(2\pi) - 2\pi \sin(\omega t_{i}/2) / \sin(\omega t_{i}/2),$$

where we assumed a constant inter-photon delay $t_{i} = t_{0} + (j - 1)T$ (in our experiment we had $T = \Delta l_{2} / c$). The function $\Delta$ comprises a series of spectral peaks of height $\Delta v = \Delta \lambda_{2} / c$. This allows an optimal choice of $\omega t_{i}$ leading to the most visible peaks.
of Dirac delta functions, so that the SPE signal can be written as
\[
\Phi(x) = \sum_{k=-\infty}^{\infty} e^{i\omega_0 k} \delta_x - \frac{T}{k} \delta_{x-k}.
\]

We would like to extract information about the Fourier components \( \Phi_k = \Phi(k \omega_0) \) of the phase; as an example we consider the quantity \( D_k = \Phi_k - \Phi_{k-1} \), where \( \tau = n_0 T = \frac{T}{2} \), calculated from two measurements with different values of the initial arrival time \( t_0 \). The zero-frequency component \( \Phi_0 \) is removed from \( D_k \), and we have
\[
D_k \propto \Re\{ \Phi_k \},
\]
where we have used the fact that \( \Phi_{-k} = \Phi_k^* \) (since \( \Phi \) is real), and where we also assumed that the phase variation is sufficiently narrowband that \( |\Phi_k| \ll |\Phi_{k-1}| \), which will hold for a wide class of smooth phase dynamics with compact temporal support of duration \( T \gg T \). This shows that just two measurements with SPE states provide access to the Fourier component of a dynamical phase at a frequency \( f = \omega_0 / 2\pi = 1/T \) fixed by the delay \( T \) between the probe photons. We note that the accuracy of this procedure is ultimately limited by the timing jitter of the nonclassical light sources that produce the photons in the SPE state, but parametric photon sources have demonstrated extremely low timing jitter\(^{16}\).

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