Super-resolving phase measurements with a multi-photon entangled state

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Using a linear optical elements and post-selection, we construct an entangled polarization state of three photons in the same spatial mode. This state is analogous to a "photon-number path entangled state” and can be used for super-resolving interferometry. Measuring a birefringent phase shift, we demonstrate two- and three-fold improvements in phase resolution.

Interference phenomena are ubiquitous in physics, and often form the basis for the most demanding measurements. Obvious examples include Ramsey interferometry in atomic spectroscopy, x-ray diffraction in crystallography, and optical interferometry in gravitational-wave studies. It has been known for some time that entangled states can be used to perform super-sensitive measurements, for example in optical interferometry or atomic spectroscopy. The idea has been demonstrated for an entangled state of two photons, but for larger number of particles it is difficult to create the necessary multi-particle entangled states. Here we experimentally demonstrate a technique for producing a maximally-entangled 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. 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.

Our goal is to create the state

\[ |N :: 0\rangle_{a,b} = \frac{1}{\sqrt{2}} \left( |N,0\rangle_{a,b} + |0,N\rangle_{a,b} \right) \tag{1} \]

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 measurements, interferometry, and matter-wave gyroscopes. 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 \(H_{\text{ext}} = \varepsilon_{ab} b^\dagger b\) 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 \(|(1,0)_{a,b} + |0,1\rangle_{a,b}\rangle/\sqrt{2}\) and allow them to evolve to the state \(|\phi\rangle \equiv (|1,0\rangle_{a,b} + \exp[\ii \phi] |0,1\rangle_{a,b})/\sqrt{2}\), where \(\phi = \varepsilon_{ab} T/\hbar\). Measurements of \(A_1 = |0,1\rangle \langle 1,0| + |1,0\rangle \langle 0,1|\) on this ensemble give \(\langle A_1 \rangle = \cos \phi\) with shot-noise limited phase uncertainty, \(\Delta \phi = 1/\sqrt{N}\). In contrast, under the same Hamiltonian \(|N :: 0\rangle\) evolves to \(|(N,0) + \exp[iN\phi]\rangle \langle 0,N|)/\sqrt{2}\). If we measure the operator \(A_N \equiv |0,N\rangle \langle N,0| + |N,0\rangle \langle 0,N|\), we find \(\langle A_N \rangle = \cos (N\phi)\). The dependence on \(N\phi\) rather than \(\phi\) is phase super-resolution: one cycle of \(\langle A_N \rangle\) implies a smaller change of \(\phi\) (or \(\varepsilon_{ab}\)) than one cycle of \(\langle A_1 \rangle\). Phase super-sensitivity, a reduction of phase uncertainty, is also predicted. A number of schemes have been proposed to reach the so-called Heisenberg limit \(\Delta \phi = 1/N\). The simplest proposals would measure the operator \(A_N\). This can be implemented with coincidence measurements, as the probability of detecting all \(N\) quanta in a mode \((a+b)/\sqrt{2}\) is proportional to \(1 + \langle A_N \rangle\).

A related technique, quantum interferometric optical lithography, proposes using phase super-resolution to write features smaller than the single-photon diffraction limit. Here the modes \(a, b\) are spatial, with different propagation directions. A molecule exposed to both modes and capable of absorbing \(N\) photons would, in effect, perform the measurement of \(A_N\) as above, with \(N\)-fold super-resolution in position. Using coincidence detection in place of two-photon absorbers, this principle has been demonstrated for \(N = 2\) using down-converted pairs. In that experiment, two infrared photons showed the same (angular) resolution as the blue pump photon which generated them, a factor of two improvement over the resolution of a single infrared photon. The question remains as to whether resolution can be improved beyond that of the photons used to generate the entangled state. Here we answer that question in the affirmative by constructing a multi-particle state with greater phase resolution than any of its single-particle precursors. The technique could in principle be used to generate entangled states of arbitrarily large \(N\) with arbitrarily good resolution.

We prepare the state \(|3 :: 0\rangle_{a,b}\) where the modes a and b are the horizontal (H) and vertical (V) polarizations of a single spatial mode. The construction of the polarization state is based on earlier proposals to construct photon-number path entangled states. A similar technique for polarization has recently been independently proposed. The key to the construction is the fact that \(|3 :: 0\rangle_{a,b}\) when written in terms of creation operators \(a^\dagger_{a,b}\) and \(a^\dagger_{b,a}\) acting on the vacuum \(|0\rangle\), is

\[ |3 :: 0\rangle_{a,b} = (a^\dagger_a + a^\dagger_b)(a^\dagger_a + e^{i\chi} a^\dagger_b)(a^\dagger_a + e^{i2\chi} a^\dagger_b) |0\rangle \tag{2} \]

where \(\chi = 2\pi/3\) and normalization has been omitted. The terms in parentheses each create a particle, but in...
non-orthogonal states. If $a$ and $b$ are left and right circular polarization, these states describe linear polarizations rotated by $60^\circ$ from each other. Using post-selection, we can put one photon of each polarization into a single spatial mode and create $|3::0\rangle_{a,b}$.

We use two photons from pulsed parametric down-conversion plus one laser, or “local oscillator” (LO) photon. Pulses of 100 fs duration and 810 nm center wavelength are produced by a mode-locked Ti:sapphire laser and frequency-doubled giving 405 nm pulses with an average power of 50 mW. These are gently focused into a 0.5 mm thick beta-barium borate crystal aligned for Type-II parametric down-conversion in the “collapsed cone” geometry [24]. The down-converted (DC) photons thus produced are orthogonally polarized. A small part of the Ti:sapphire beam is split off and attenuated to contribute the LO photon. These three photons are transformed into the state $|3::0\rangle$ and detected by polarization-resolved three-fold coincidence detection. The transformation, shown in Figure 1 a), can be understood as a sequence of mode combinations. After the PBS, the DC photons are in the state $a_0^H a_1^V|0\rangle$, where subscripts indicate polarization. A half-wave plate rotates this to $a_{45H}^I a_{45V}^I|0\rangle$ where subscripts show linear polarizations measured from vertical. We perform the first post-selected non-unitary operation by passing the pair through three plates of BK7 glass near the Brewster angle. The six Brewster-angle interfaces act as a partial polarizer (PP) with transmission efficiencies of $T_H \approx 1$, $T_V = 1/3$. By post-selecting cases where no photons are reflected, we transform the state as (again without normalization)

\[
\begin{align*}
    a_{45H}^I a_{45V}^I|0\rangle &= (a_H^I + a_V^I)(a_H^I - a_V^I)|0\rangle \\
    &= (a_H^I + \frac{1}{\sqrt{3}}a_V^I)(a_H^I - \frac{1}{\sqrt{3}}a_V^I)|0\rangle \\
    &= a_{60H}^I a_{60V}^I|0\rangle .
\end{align*}
\]

This operation, putting orthogonally polarized photons into non-orthogonal modes, is non-unitary and requires post-selection.

The DC photons meet the LO photon at the last interface of the PP. This interface acts as a beamsplitter putting all three into the same spatial mode, conditioned on zero photons exiting by the “dark” port. Again, the operation is non-unitary and requires post-selection. The LO photon is vertically polarized and the state thus constructed is $a_0^V a_{60H}^I a_{60V}^I|0\rangle$. This state has six-fold rotational symmetry about the beam propagation direction, and thus can only contain the states |3,0⟩ $\pm$ |3,3⟩ $\pm$ |3,0⟩ $\pm$ |3,3⟩ $\pm$ |3,0⟩ $\pm$ |3,3⟩. Indeed, the probability of both post-selections succeeding is $\cos^4(\pi/12)/3^{1/6} \approx 72\%$.

Response to the phase shifter demonstrates phase super-resolution. Acting on a single photon, the quartz wedges shift by $\phi$ the phase of the $V$-polarization. Acting on three photons the phase shift is tripled: |3::0⟩ becomes $|3\phi⟩_{H,V}$ whereas |3::0⟩ $\pm$ $|3\phi⟩_{H,V}$ $\pm$ $\exp[3\phi]|3\phi⟩_{H,V}/\sqrt{2}$, where we have absorbed the negative sign above into the phase factor. The $3\phi$ behaviour can be seen in triples detection in the $\pm 45^\circ$ linear polarization basis. The rates for detection of |3::0⟩ $\pm$ $|3\phi⟩_{H,V}$ and |2,1⟩ $\pm$ $|3\phi⟩_{H,V}$ vary as $1 \pm \cos(3\phi)$, respectively. After passing through an 810 nm wavelength filter with a 10 nm passband, the photons enter the polarization analyzer, set to detect |3::0⟩ $\pm$ $|3\phi⟩_{H,V}$ or |2,1⟩ $\pm$ $|3\phi⟩_{H,V}$, as shown in Figure 1 b).

The use of down-converted pairs removes the need for detectors at the “dark” ports. Down-conversion very frequently produces more than two photons in a single pulse so we can infer with near-certainty the absence of photons in the “dark” port from the presence of both photons in the “bright” port. Using a weak coherent
state to supply the third photon, we can make small the probability that more than one LO photon was present in a triple detection event. Thus a single post-selection for three-fold coincidence at the detectors performs at once the post-selections for both non-unitary operations.

Figure 2 shows results for detection of multiple polarizations at the analyzer. Intensities of the DC and LO sources were adjusted such that singles detections are mostly produced by LO photons (about 10:1 ratio vs. DC singles), two-fold coincidences mostly by DC pairs (about 5:1 vs. LO accidentals), and three-fold coincidences principally by one LO photon plus one DC pair (about 2:1 vs. accidental triples contributions, below). Thus with a single scan of the phase shifter we can see qualitatively different behaviours for states of one, two, and three photons. Figure 2 c) clearly shows oscillation with $3\phi$, as predicted by theory. The resolution exceeds that achievable with any single photon in the experiment. The 405 nm photons would show oscillation with $2.1\phi$ due to their shorter wavelength and the somewhat larger birefringence of quartz at that wavelength. A cosine curve fitted to these data shows a visibility of 42±3%. This visibility is unambiguous evidence of indistinguishability and entanglement among the three photons. A non-entangled state of three distinguishable photons could also show three-fold coincidence oscillation at $3\phi$, but with a maximal visibility of 20%.

Figure 2 d) shows the same triples data after subtraction of background from accidental triples. In addition to the signal of interest from 1 DC pair + 1 LO photon, we also see events from 2 DC pairs, from 3 LO photons, and from 2 LO photons + 1 DC pair. We calculate these backgrounds from independent measurements of single and double detection rates for the DC and LO sources alone. Coincidence background is calculated by the statistics of uncorrelated sources using a time-window of 12.5 ns, the laser pulse period. Incoherence of the various contributions is ensured by sweeping the path length of the LO photon over ±2µm during acquisition. The calculated background has some variation with $\phi$, so it is important to note that it is qualitatively different than the observed $3\phi$ signal. Per 30 second interval, the accidental background contributes 22 ± 1 as a constant component, an average of 23±1 oscillating with $2\phi$, 4±1 with $1\phi$ and < 1 with $3\phi$. Here and elsewhere, uncertainty in the counting circuitry’s dead-time introduces a systematic error.

It is also possible to see $3\phi$ behaviour detecting a single polarization, as shown in Figure 3. This measurement corresponds to the original proposals for atomic spectroscopy and lithography. It gives a far weaker signal, in part because the maximum overlap of the state $|3::0\rangle_{H,V}$ with $|3,0\rangle_{±45^\circ}$ is smaller than with $|2,1\rangle_{±45^\circ}$. Also, the chance that all three photons go to distinct detectors (as needed for coincidence detection) is smaller.
for $|3, 0\rangle_{\pm 45^\circ}$. With these limitations, we are able to see the $3\phi$ behaviour, but only by subtracting a considerable coincidence background.

Using linear optical elements and post-selection, we have constructed a multi-particle entangled state useful for super-resolving phase measurements. The demonstrated resolution is not only better than for a single infrared photon, it is better than could be achieved with any single photon in the experiment, including the down-conversion pump photons. Given the difficulty of generating coherent short-wavelength sources, this is encouraging for the prospects of proposals such as quantum-interferometric optical lithography. Finally, we note that the construction, which proceeds from unentangled photons in distinct spatial modes to a maximally-entangled state in a single spatial mode, does not require prior entanglement of the component photons. As such, the method is well adapted for use with single-photon-on-demand sources [25, 26] which promise to be more efficient and scalable than down-conversion sources.

**ACKNOWLEDGEMENTS**

We thank K. Resch and J. O’Brien for helpful and stimulating discussions and J. Dowling for inspiration. This work was supported by the National Science and Engineering Research Council of Canada, Photonics Research Ontario, the Canadian Institute for Photonics Innovations and the DARPA-QuIST program (managed by AFOSR under agreement No. F49620-01-1-0468).

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