Quantum enhanced multiple-phase estimation with multi-mode NOON states

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Quantum metrology can achieve enhanced sensitivity for estimating unknown parameters beyond the standard quantum limit. Recently, multiple-phase estimation exploiting quantum resources has attracted intensive interest for its applications in quantum imaging and sensor networks. For multiple-phase estimation, the amount of enhanced sensitivity is dependent on quantum probe states, and multi-mode NOON states are known to be a key resource for this. However, its experimental demonstration has been missing so far since generating such states is highly challenging. Here, we report generation of multi-mode NOON states and experimental demonstration of quantum enhanced multiple-phase estimation using the multi-mode NOON states. In particular, we show that the quantum Cramer-Rao bound can be saturated using our two-photon four-mode NOON state and measurement scheme using a 4 × 4 multi-mode beam splitter. Our multiple-phase estimation strategy provides a faithful platform to investigate multiple parameter estimation scenarios.
Quantum metrology has attracted intensive interest in recent years, as it allows us to estimate an unknown parameter with enhanced sensitivity over classical approaches. Developments are now directed towards various applications such as microcopy\(^1\)–\(^3\), biological imaging\(^4\)–\(^9\), and sensor network\(^10\)–\(^13\). In such practical applications, quantum metrology is naturally extended to multiple parameter scenarios, since the number of parameters that affect a physical process is generally more than one. For estimating multiple parameters, simultaneous estimation is desirable as it can achieve higher precision than an individual estimation of each parameter using the same amount of resources\(^14\)–\(^16\). Furthermore, if a physical system of interest has time dynamics, estimation of multiple parameters has to be done simultaneously. However, unlike single parameter estimation, it is non-trivial to optimize multiple parameter estimation to achieve the maximum sensitivity. Various strategies have been thus proposed to enhance the sensitivity by optimizing either the probe states or the measurement scheme\(^14\)–\(^22\). For example, quantum strategies exploiting various quantum probe states such as Greenberger–Horne–Zeilinger states\(^23\), single-photon Fock states\(^24\), squeezed states\(^25\)–\(^28\), Holland–Barnett states\(^29\)–\(^31\), and N00N states\(^32\)–\(^33\) have been extensively studied.

In particular, N00N states have been outstanding to investigate the fundamental quantum limit of quantum metrology given by the Heisenberg uncertainty principle with a fixed number of particles\(^34\). In a single-phase estimation scheme, N00N states can saturate the Heisenberg limit thanks to its largest number variance between the two modes\(^34\). Enhanced sensitivity beyond the standard quantum limit has been experimentally demonstrated with N00N states\(^35\)–\(^36\). Recently, the concept of N00N states has been extended to generalized multi-mode N00N states to investigate the quantum enhancement in multiple-phase estimation\(^14\).

Moreover, for multiple-phase estimation with limited resources, it has been known that multi-mode N00N states allow achieving the enhanced sensitivity outperforming the other quantum probe states or classical strategies for a multi-mode interferometer\(^14\)–\(^15\)–\(^17\)–\(^32\). However, since generation of such multi-mode quantum probe states is challenging, experimental demonstrations of multiple parameter quantum metrology have been limited to utilizing quantum states other than multi-mode N00N states\(^12\)–\(^15\)–\(^23\)–\(^24\)–\(^27\)–\(^29\).

In this work, we experimentally demonstrate quantum enhanced multiple-phase estimation using a multi-mode N00N state with photon number \(N = 2\) and mode number \(m = 4\). To this end, we propose a scheme for generating multi-mode N00N states. At first, we observe that the measured interference fringes exhibit phase super-resolution as a function of phase differences \(\phi_i\) (\(i = 1, 2, 3\)) of the generated 4-mode 2002 state with a measurement scheme using a \(4 \times 4\) multi-mode beam splitter, so-called a quarter\(^37\)–\(^38\). Then, we demonstrate quantum enhanced phase sensitivity of the 4-mode 2002 state by analyzing the Cramer–Rao bound (CRB) and the quantum Cramer–Rao bound (QCRB)\(^14\)–\(^16\)–\(^17\)–\(^39\). Moreover, we show that the measured sensitivity is better than a coherent state \(|\alpha\rangle\), which is a classical probe state, as well as another quantum probe state prepared by single-photon Fock states\(^24\). Our results can motivate an investigation into the quantum strategies using multi-mode and multi-particle entanglement to develop the quantum enhanced multiple parameter metrology.

**Results**

**Multiple-phase estimation scenario.** Let us begin by introducing the system model for multiple-phase estimation schemes with multi-mode N00N states defined as

\[
|\psi_m^N\rangle = \frac{1}{\sqrt{m}} \left(|N0\cdots0\rangle + |0N0\cdots0\rangle + \cdots + |0\cdots0N\rangle\right),
\]

where \(N\) is the number of photons, and they are distributed along \(m\) modes\(^34\)–\(^33\). The multi-mode N00N state is a coherent superposition of all possibilities where \(N\) photons in one mode and none in any of the other \(m - 1\) modes\(^34\). Then, we theoretically analyze the sensing scheme of \(d = m - 1\) unknown multiple phases and the fixed photon number \(N = 2\) with different probe states. A quantum probe state undergoes individual phase shifts \(\phi = [\phi_1, \phi_2, \ldots, \phi_d]\) and the phase shifted state is combined using an \(m \times m\) multi-mode beam splitter and then detected by photon number-resolving detectors (PNRDs) at each mode. Here, the goal is to minimize the total uncertainty of the phase estimation governed by the CRB and the QCRB. The lower bound of the sum of the variance of each phase estimation given by the CRB and the QCRB is\(^14\)–\(^16\)–\(^17\)–\(^39\)

\[
\frac{1}{m} \sum_{i=1}^{d} |\Delta \phi_i|^2 \geq \frac{\text{Tr} \left[ F^{-1}_C(\phi) \right]}{\mu} = \frac{\text{Tr} \left[ F^{-1}_Q(\phi) \right]}{\mu},
\]

where \(F_C(\phi)\) is the classical Fisher information matrix (CFIM), \(F_Q(\phi)\) is the quantum Fisher information matrix (QFIM), and \(\mu\) is the number of measurements. Note that \(F^{-1}\) refers to the inverse of the Fisher information matrix \(F\). The first and second inequalities are direct consequences of the CRB and the QCRB, respectively. Note that in our experiments, the CRB can always be saturated, asymptotically in \(\mu\), by using maximum likelihood estimator, and the mean of phases are expected to converge to the true values\(^17\)–\(^39\).

We theoretically analyze the CRB and the QCRB of \(d\) multiple-phase estimation scheme with two different probe states. The first probe state we consider is a classical state prepared by injecting a coherent state \(|\alpha\rangle\) with an average photon number \(N = 2\) into one of the input ports of an \(m \times m\) multi-mode beam splitter (Fig. 1a), and the other probe state is the two-photon \(m\)-mode N00N state (Fig. 1b). The measurement scheme is identical for two probe states. Each mode of the probe states is combined at another \(m \times m\) multi-mode beam splitter after undergoing phase shifts, and then measured by PNRDs. We provide the CRB and the QCRB values of the total variance \(\sum_i |\Delta \phi_i|^2\) for two probe states of coherent states and multi-mode N00N states with various number of phases \(d\) in Fig. 1c. Note that multi-mode N00N states always have lower phase uncertainty than coherent states, i.e., classical probe states.

**Generation of the 4-mode 2002 state.** The conceptual diagram of our proposed scheme for preparing the multi-mode N00N state with \(N = 2\) and \(m = 4\) is shown in Fig. 2a. The generation process of 4-mode 2002 state \(|\psi_4^2\rangle\) is the following:

\[
|\Phi^+\rangle = \frac{1}{\sqrt{2}} \left(|H_a|^2|V_b\rangle + |V_a|^2|H_b\rangle\right)
\]

\[
\text{BS} \frac{1}{2} \left[ |2H_a^00_0\rangle + |2V_a^00_0\rangle + |2V_a^10_0\rangle + |2H_a^10_0\rangle \right] \rightarrow \text{PBS, HWP} \frac{1}{2} \left[ |2H_a0_0^00_0\rangle + |2V_a0_0\rangle + |2V_a0_1\rangle + |2H_a0_1\rangle \right].
\]

Here, \(|\Phi^+\rangle = \frac{1}{\sqrt{2}} (|1_H^0\rangle + |1_V^0\rangle)\) is the triplet Bell state where, for example, \(|1_H^0\rangle\) denotes the horizontally polarized single-photon state in the mode \(a_0\). Our scheme can generate \(|\psi_4^2\rangle\) with a unity conversion probability from the pre-selected...
Then, in order to directly confirm the generation of $|\psi_4^\pm\rangle$, we measure two-photon probability distributions for the output states when we use all of the input modes $a_0$, $a_1$, $a_2$, and $a_3$ of $|\psi_4^\pm\rangle$ while we vary one of the phase encoding $\phi_i$. The experimental results of the interference fringes from all of the modes are shown in Fig. 3d–f, and they are compared with the theoretical calculations. We obtain the theoretical predictions of $C_{\theta_0,\theta_i}$, $C_{\theta_0,\theta_i}$, $C_{\theta_0,\theta_i}$, and $C_{\theta_0,\theta_i}$ based on the ideal $|\psi_4^\pm\rangle$ and the experimentally reconstructed quarter transition matrix. Then we experimentally measured post-selected coincidence counts of $C_i$ for $i = b_0 b_0, b_0 b_1, b_0 b_2$ and $b_0 b_3$. See Supplementary Note 1 for the detailed information on a quarter transition matrix and theoretical calculations. The results of Fig. 3d–f shows that the experimentally obtained interference fringes without normalization are very well-matched to the theoretical calculations, thus we can confirm that the prepared input state is $|\psi_4^\pm\rangle$.

**Experimental multiple-phase estimation.** Then, we investigate the sensitivity bound of multiple-phase estimation using our prepared $|\psi_4^\pm\rangle$ as a probe state. At first, we theoretically calculate the CRB with an ideal $|\psi_4^\pm\rangle$ and an ideal quarter when the CRB saturates the QCRB. We obtain the theoretical two-photon detection probability set of $|P_i(\phi)|$ ($i = 0, 1, \ldots, 9$) with a set of projectors $\{\Pi_i\} = \{\langle 2000 | (2000), | 0200 | (0200), | 0020 | (0020), | 0002 | (0002), | 1100 | (1100), | 0110 | (0110), | 0011 | (0011), | 0101 | (0101), | 1010 | (1010), | 1101 | (1101), | 0110 | (0110) \}$ satisfying the normalizing condition $\sum_j |P_i(\phi)| = 1$. Note that $P_0 = P_3 = P_5$, $P_4 = P_7$, and $P_2 = P_9$ for all $\phi$ (See Methods). Then CFIM is given by

$$F_{\text{CFM}}(k) = \sum_i \frac{1}{P_i} \frac{\partial P_i(\phi)}{\partial \phi_i} \left( \frac{\partial P_i(\phi)}{\partial \phi_k} \right),$$

where $j$ and $k$ can be 1, 2, and 3. The minimum value of the CRB is obtained to be $\text{Tr}[F_{\text{CFM}}(\phi)] = 1.5$ where $\phi_i \approx n/2$, and $\phi_3 = 0$, and it saturates the QCRB $= 1.5$. In order to experimentally estimate the CRB, we obtained interference fringes by scanning $\phi_i$ near the point where we expected both the CRB and the QCRB to be saturated ($\phi_1 \approx -0.07\pi$ and $\phi_3 \approx 0.52\pi$) for the prepared $|\psi_4^\pm\rangle$ probe state (See Methods). Two-photon detection probabilities $P_{m_0 b_0}^m$, $P_{m_1 b_1}^m$, $P_{m_2 b_2}^m$, $P_{m_3 b_3}^m$ and $P_{m b_i}^m$ are then obtained from the measured post-selected coincidence counts with $\mu \approx 8, 144, C_{\theta_0,\theta_i}$, ($i = b_0 b_0, b_0 b_1, b_0 b_2$, and $b_0 b_3$), which were appropriately normalized assuming the following relations $P_{\theta_0 b_0}^m = P_{\theta_1 b_1}^m = P_{\theta_2 b_2}^m = P_{\theta_3 b_3}^m$, $P_{\theta_0 b_0}^m = P_{\theta_1 b_1}^m = P_{\theta_2 b_2}^m = P_{\theta_3 b_3}^m$, $P_{\theta_0 b_0}^m = P_{\theta_1 b_1}^m = P_{\theta_2 b_2}^m = P_{\theta_3 b_3}^m$. Note that the assumed relations are always satisfied for $P_i(\phi)$ with an ideal quarter and the experimentally reconstructed quarter transition matrix is close to an ideal quarter. Then obtained $P_{\theta_i}^m(\phi)$ are functions of unknown phases $\phi$ and used to calculate the derivatives in Eq. (4). The detailed relation between $P_i^m(\phi)$ and $P_m b_i$ are provided in Supplementary Note 3.

Experimentally obtained two-photon detection probabilities are shown in Fig. 4a where our experimental data are well-matched to our fitting function $F_{\text{CFM}}^m(\phi)$, which are obtained from $|\psi_4^\pm\rangle_{\text{exp}}$ and $U_{\text{exp}}$ (see “Methods”). The CFIM can be obtained by using $P_{\theta_i}^m(\phi)$, and the diagonal terms of the CFIM at various $\phi_i$ are plotted in Fig. 4b (see “Methods” for the detailed information). Note that the maximum values of all diagonal terms of the CFIM are 3 at $\phi_i \approx 0.57\pi$ with an ideal $|\psi_4^\pm\rangle$ state and an ideal quarter. Then we numerically find the minimum CRB of $\text{Tr}[F_{\text{CFM}}^m(\phi)^{-1}] = 1.85 \pm 0.01$ when $\phi_1 \approx 0.47\pi$, $\phi_2 = -0.07\pi$, and $\phi_3 = 0.52\pi$ from

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**Fig. 1 Quantum enhanced multiple-phase estimation scheme.** The goal is to estimate the $d$ multiple phases with phase shifts $\phi_0, \ldots, \phi_d$ while minimizing the total uncertainty of phase estimation governed by the CRB and the QCRB. To estimate the multiple phases, coherent states with $N = 2$ (a) and $(d + 1)$-mode NOON states with $N = 2$ (b) are used for probe states. The total variance of multiple-phase estimation obtained by the CRB and the QCRB as a function of the number of unknown phases $d$ with two probe states. It clearly shows that multi-mode NOON states always have better sensitivity compared to coherent states.

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As a first step to verify the generation of $|\psi_4^\pm\rangle$, we investigate the coherence among all four modes of $|\psi_4^\pm\rangle$ by observing the interference fringes between the reference mode ($a_0$) and one of the other modes ($a_i$). We measure the two-photon coincidence counts on $b_0$ and $b_0$, $C_{\theta_0}^{00\theta_0}$, of two input modes while we block the other two input modes. The phase shift $\phi_i$ is realized by adjusting the optic axis angle $\theta_i$ of the half waveplate (HWP) located between two quarter waveplates (QWP). Note that $\phi_i = 2\theta_i$. As shown in Fig. 3a–c, results of $C_{\theta_0}^{00\theta_0}$ clearly reveal two times faster sinusoidal modulations compared to the single-photon input case due to the $\lambda/2$ photonic de Broglie wavelength of two-photon NOON states $n^{1/2}$.
\( P_{\text{QCRB}}(\varphi) \), which is clearly smaller than the CRB of the coherent state \( \text{Tr}(\{ F_{\text{coh}} \})^{-1}(\varphi) \) with the same average photon number \( \overline{N} = 2 \) as shown in Fig. 4c (see Supplementary Note 3 for the detailed calculation on estimating the CRB and results with respect to \( \varphi_2 \) and \( \varphi_3 \)). Note that the ideal CRB value can achieve 1.5 to saturate the QCRB with an ideal \( |\psi_2^\text{coh} \rangle \) state and an ideal quarter at \( \varphi_1 \approx 0.5\pi, \varphi_2 = 0, \) and \( \varphi_3 = 0.5\pi \).

In our experiment, experimental errors are mainly from non-unity visibility of the observed interference, a phase fluctuation in each arm of an interferometer, a normalization assumption due to lack of superconducting nanowire single-photon detector (SNSPD) channels, and the fact that the quarter \( U_q^{\exp} \) used in our experiment is slightly different from an ideal quarter \( U_q^\text{ideal} \), see Methods for comparison between \( U_q^{\exp} \) and \( U_q^\text{ideal} \).
probabilities $P_b$.

Corresponding orthogonal term of CFIM obtained from the measured standard deviation and shaded areas correspond to the one standard error bars for all $\beta$ happen at slightly different probe states. One can notice it from Fig. 4a that $P_0^m$, $P_1^m$, and $P_2^m$ do not have their minimum (maximum) value at $\phi_1 \approx 0.5\pi$. This is the reason why the minimum value of the CRB is not obtained at $\phi_1 \approx 0.5\pi$ but $\phi_1 \approx 0.47\pi$.

Furthermore, we emphasize that the enhanced sensitivity can be obtained by the prepared $|\psi_F^m\rangle$ probe state compared to other quantum probe states using single-photon Fock states $|\psi_F^m\rangle$ proposed in ref. 24, where $|1100\rangle$ state is used as an input state instead of $|\alpha\rangle$ in Fig. 1a (detailed calculations are provided in Supplementary Note 2). Moreover, we theoretically compare the sensitivity bounds between our multi-mode N00N state and an amplitude-unbalanced multi-mode N00N state $|\psi_u\rangle$, which is proposed in refs. 14,15,39. The amplitude-unbalanced multi-mode N00N state has the form of $|\psi_u\rangle = |\alpha(N0\cdots0) + \beta(0N0\cdots0) + \cdots + |0\cdots0N\rangle$ with $\alpha^2 + \beta^2 = 1$ and $\beta = 1/\sqrt{d + \sqrt{d}}$. $|\psi_u\rangle$ is known to have the minimum CRB among the multi-mode N00N states14,15,39. In a measurement scheme using a quarter and PNDRs, the QCRB and the CRB values of $|\psi_u\rangle$ with $N = 2$ and $d = 3$ are theoretically calculated to be 1.4 and 1.62, respectively.

Here, we find that even though $|\psi_u\rangle$ has the lower CRB of 1.4 than 1.5 of our $|\psi_2\rangle$, $|\psi_4\rangle$ can provide a better sensitivity (smaller CRB) 1.5 than 1.62 of $|\psi_u\rangle$ with a realistic measurement scheme using a quarter. Note that an optimal measurement saturating the QCRB may not be experimentally feasible14,17.

In Table 1, we summarize the ideal QCRB and CRB values for various probe states as well as the CRB values obtained from our experimental results. We emphasize that the experimentally obtained CRB value of $1.85 \pm 0.01$ provides a better sensitivity than the ideal CRB values of 3 for $|\alpha\rangle$ and 2.44 for $|\psi_F^m\rangle$ using an ideal quarter, respectively. All error bars represent one standard deviation and shaded areas correspond to the one standard deviation from uncertainty of the fitting parameter.

### Table 1 CRB and QCRB for total variances of $\sum|\Delta \phi|^2$ with various probe states.

| Probe state | $U$ | QCRB | CRB |
|-------------|-----|------|-----|
| $|\alpha\rangle$ ($N = 2$) | $U_0$ | 3 | 3 |
| $|\psi_{\text{Fock}}\rangle_{24}$ | $U_1$ | 2.33 | 2.44 |
| $|\psi_2\rangle_{14}$ | $U_2$ | 1.4 | 1.62 |
| $|\psi_4\rangle_{\text{exp}}$ | $U_{\text{exp}}$ | 1.5 | 1.5 |
| $|\psi_{\text{Fock}}\rangle_{\text{exp}}$ | $U_{\text{exp}}$ | $1.54 \pm 0.01$ | $1.85 \pm 0.01$ |

Discussion

In conclusion, we propose a scheme for generating a multi-mode N00N state and experimentally demonstrate that the prepared quantum probe state is the 4-mode 2002 state by observing various interference fringes shown in Fig. 3 using a quarter and photon number resolving detection using post-selective pseudo-PNDRs. Moreover, we exploit the prepared 4-mode 2002 state as a quantum probe state for simultaneously estimating three phases of a 4-mode interferometer with quantum enhanced sensitivity. Then, we confirm that the CRB obtained by our 4-mode 2002 state and measurement scheme can saturate the QCRB. Our results provide a practical platform to investigate intriguing issues in the field of quantum multiple parameter metrology. At first, we emphasize that our scheme can be extended to generation of higher-mode N00N states. For instance, one can exploit a multiple-path Sagnac interferometer44 to increase the number of higher-mode qubits from 4 to 4$n$, i.e., $|\psi_{\text{Sagnac}}\rangle$, where $n$ is the number of Sagnac interferometers, and then one can estimate up to 4$n$ – 1 phases simultaneously. Note that another scheme for generating multi-mode N00N states with $N \geq 2$ has been theoretically proposed33. However, experimental demonstration seems to be challenging within current technology since it requires extremely strong nonlinearity or deterministic generation of multi-photon states.

Another interesting future direction would be finding a realistic
measurement scheme for minimizing the CRB. In general, the CRB obtained by measurement scheme using a balanced multimode beam splitter cannot saturate the QCRB, see Fig. 1c with $d = 2, 4, 5$. Since the optimal measurement saturating the QCRB involves complex multi-photon states, it may not be experimentally feasible\cite{14,17}. Hence, finding an experimentally realistic measurement scheme minimizing the CRB is essential for practical applications. Our results have direct applications for the quantum enhanced phase object imaging requiring a low photon flux\cite{45,46}. In addition, our results can pave the way for demonstrating distributed quantum enhanced multiple-phase estimation by increasing the number of phases in local interferometers consisting of distributed quantum sensors\cite{12,23,27}.

**Methods**

4-mode 2002 state preparation. We used a CW single frequency laser operating at a central wavelength of 780 nm. The polarization of pump laser is set to $|L\rangle = (|H\rangle + |V\rangle)/\sqrt{2}$ polarization. The 10 mm-thick type-II periodically poled KTiOPO4 (PPKTP) crystal with 46.15 $\mu$m poling period is located at a center of the Sagnac interferometer, which consists of a dual wavelength polarizing beam splitter (PBS), a dual wavelength HWP whose optic axis is oriented at 45°, and two dual wavelength mirrors as shown in Fig. 2c\cite{33}. Here, dual wavelength optical components are designed for working at both 780 and 1560 nm photons. The horizontal (vertical) polarization component of the pump laser is transmitted (reflected) at dual wavelength PBS. The vertically polarized pump laser is changed to the horizontal polarization after passing through the SNSPD whose detection efficiency of $\Phi_{\text{det}}$ is 37% with our SNSPD whose detection efficiency of $\Phi_{\text{det}}$ is 37%. The experimentally obtained CRB is 37% with our SNSPD whose detection efficiency of $\Phi_{\text{det}}$ is 37%.

Two photons prepared in $|\Phi^+\rangle$ state simultaneously entered at both input ports after the initial state $|\Phi^+\rangle = |0\rangle_1 |1\rangle_2 + |1\rangle_1 |0\rangle_2$. Our results have direct applications for the quantum enhanced phase object imaging requiring a low photon flux\cite{45,46}. In addition, our results can pave the way for demonstrating distributed quantum enhanced multiple-phase estimation by increasing the number of phases in local interferometers consisting of distributed quantum sensors\cite{12,23,27}.

**Theoretical analysis of $|\psi_{\text{out}}\rangle$.** Our probe state is the 4-mode 2002 state of the form

$$|\psi_{\text{in}}\rangle = \frac{1}{2\sqrt{2}} (|a_0^1 a_2^1 + a_1^1 a_2^1 + a_2^1 a_2^1 + a_2^1 a_2^1\rangle 0),$$

where $\{a_i^j\}$ is a creation operator, which creates a single-photon in the input mode $a_i$ of a quarter. A quarter has four input modes ($a_0, a_1, a_2, a_3$) and four output modes ($b_0, b_1, b_2, b_3$), respectively, as shown in Fig. 2c. The unitary matrices for the phase encoding $U_\phi$ and the ideal quarter transformation $U_\chi$ are given by\cite{15,18}:

$$U_\phi = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & e^{i\phi} & 0 & 0 \\ 0 & 0 & e^{i\phi} & 0 \\ 0 & 0 & 0 & e^{i\phi} \end{pmatrix},$$

$$U_\chi = \frac{1}{2} \begin{pmatrix} 1 & 1 & 1 & 1 \\ 1 & 1 & -1 & -1 \\ 1 & -1 & -1 & -1 \\ 1 & -1 & -1 & -1 \end{pmatrix},$$

respectively. After the initial state $|\psi_{\text{in}}\rangle$ undergoes the phase encoding and the quarter transformation, then the output state $|\psi_{\text{out}}\rangle = U_\phi U_\chi |\psi_{\text{in}}\rangle$ becomes

$$|\psi_{\text{out}}\rangle = \sum_{i=0}^{3} c_i |0000\rangle + 2 c_i |0020\rangle + c_i |0002\rangle + c_i |1100\rangle + 2 c_i |0011\rangle + c_i |0110\rangle + c_i |0101\rangle + c_i |1001\rangle + c_i |1101\rangle + c_i |0110\rangle,$$

with $c_0 = c_1 = c_2 = c_3 = c_4 = c_5 = c_6 = c_7$ and $c_8 = c_9$. Then, one can obtain the two-photon detection probability of $P(\Phi) = |\langle \psi_{\text{in}} |\langle \psi_{\text{in}} \rangle |^2$ (l = 0, 1, ..., 9), and it satisfies the normalizing condition $\sum_{l=0}^{9} P(\Phi) = 1$. $P(\Phi)$ is a function of $\Phi$ and used for theoretical calculations in Fig. 3. See Supplementary Note 3 for the detailed information on $P(\Phi) = |\langle \psi_{\text{in}} |\langle \psi_{\text{in}} \rangle |^2$.

**Analysis considering experimental errors**. In order to include the errors caused by our experimental imperfection, we consider the generation of 4-mode 2002 state from an imperfect Bell state of $|\Phi_{\text{exp}}^+\rangle$, which is given by\cite{9}:

$$|\Phi_{\text{exp}}^+\rangle = \frac{1}{\sqrt{2}} (|\Phi_{\text{in}}^+\rangle + |\Phi_{\text{in}}^-\rangle) = \frac{1}{\sqrt{2}} \left( \begin{pmatrix} 1 \\ 1 \\ 0 \\ 0 \end{pmatrix} + \begin{pmatrix} 1 \\ -1 \\ 0 \\ 0 \end{pmatrix} \right),$$

where $\alpha_{\text{in}}^1 (\alpha_{\text{in}}^2)$ creates a photon at a horizontal (vertical) polarization state with different $\alpha_{\text{in}}$. Then, experimental two-photon detection probabilities are obtained as $P_{\text{exp}}(\Phi)$, which is given for fitting curves in Fig. 4a. See Supplementary Note 3 for the detailed calculations on our error analysis.

**Quantum Fisher information matrix and CRB.** The QFIM is given by

$$F_{\Phi}(\delta\Phi) = 4 \text{Re} \left( \langle \delta\varphi | \langle \psi_{\text{in}} | \psi_{\text{in}} \rangle | \delta\varphi \rangle \langle \psi_{\text{in}} | \psi_{\text{in}} \rangle | \delta\varphi \rangle \right).$$

By calculating $F_{\Phi}$, one can obtain the QCRB from $\text{Tr}[F_{\Phi}^{-1}]$. For a $|\Phi_{\text{exp}}^+\rangle$ probe state, the QCRB is calculated to be $\text{Tr}[F_{\Phi}^{-1}] = 1.5$, and the minimum value of the CRB is obtained at $\varphi_1 = 0.3$, $\varphi_2 = -0.5$, and $\varphi_3 = 0.5$. Then, the CRB has the same value with $\text{Tr}[F_{\Phi}^{-1}] = 1.5$. The ideal $F_{\Phi}$ and $F_{\phi}$ matrices are obtained to be

$$F_{\Phi} = \begin{pmatrix} 3 & -1 & -1 \\ -1 & 3 & -1 \\ -1 & -1 & 3 \end{pmatrix},$$

$$F_{\phi} = \begin{pmatrix} 2.33 & -0.63 & -0.93 \\ -0.63 & 2.70 & -1.09 \\ -0.93 & -1.09 & 2.66 \end{pmatrix},$$

where $\varphi_1 = 0.3$, $\varphi_2 = -0.5$, and $\varphi_3 = 0.5$. The experimentally obtained $F_{\Phi}^{\text{exp}}$ is given as below:

$$F_{\Phi}^{\text{exp}} = \begin{pmatrix} 2.33 & -0.63 & -0.93 \\ -0.63 & 2.70 & -1.09 \\ -0.93 & -1.09 & 2.66 \end{pmatrix}.$$
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Author contributions

H.-T.L initiated and led the project. The experimental scheme was designed by S.H., Y.-W.C., H.-T.L., and performed by S.H. Theoretical calculations were carried out by S.H., J.R., S.-W. L., and H.-T.L. S.H. and H.-T.L. analyzed the experimental data. H.-T.L. and S.-W.H. helped to analyze the data and discuss the results. All authors contributed to discussions and writing the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

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