Supplementary Materials

A. Senarmont Compensation Method

By simply introducing a QWP after the target sample with an optical axis parallel to the polarizer, we can introduce the so-called Senarmont compensation method into our system. In the following calculation, we will demonstrate the advantages of the Senarmont method by comparing the Fisher information of the quantum polarization measurement system with or without the QWP compensator.

First, let's consider the Fisher information without the compensator (i.e., Polarizer-Sample-Analyzer, PSA configuration). With an input state $\left|\phi^{+}\right\rangle = 1/\sqrt{2}\left(\left|HV\right\rangle + \left|VH\right\rangle\right)$, there is only the sample in the measurement arm that changes the polarization state before the polarization analysis part. Therefore, the output state is calculated to be $\left|\phi^{+}_{sample}\right\rangle = \left(I \otimes S\right)\left|\phi^{+}\right\rangle$, where I and S stand for the Jones matrices of the unitary matrix and an unknown retarder in the control and measurement arms, respectively. After passing through the polarization analysis part, the photon state would collapse to a mixed state $\rho = P_a\left|11\right\rangle\left\langle 11\right| + P_b\left|10\right\rangle\left\langle 10\right| + P_c\left|01\right\rangle\left\langle 01\right| + P_d\left|00\right\rangle\left\langle 00\right|$. In between, P_a, P_b, P_c and P_d stand for all the possible results including obtaining one photon in each detector, obtaining one photon only in control/measurement path, and no photon detected, respectively. With such a probability distribution, we can calculate the Fisher information for the sample's retardance δ:

$$F_{q|\delta} = \sum_{m=a,b,c,d} \frac{1}{P_m}\left(\frac{\partial P_m}{\partial \delta}\right)^2 = \frac{\sin^2 a'\sin^2 b\sin^2\delta}{1-\left(\cos a'\cos b - \cos\delta\sin a'\sin b\right)^2}. \tag{S.1}$$

Here $a' = 4h_1 + 2\theta$ and $b = 4h_2 - 2\theta$ stand for twice the angle difference between the measurement bases in the two arms and the sample's optical axis. This result shows a limitation of the PSA configuration. When the phase retardance of the sample is $k\pi(k\in\mathbb{Z})$ (e.g. an HWP), the Fisher information is fixed to zero, which means poor measurement accuracy under arbitrary measurement bases.

As a comparison, after we introduce the compensator to form the Polarizer-Sample-Compensator-Analyzer (PSCA) configuration, the polarization state before the analysis part is $\left|\phi^{+}_{sample}{}'\right\rangle = \left(I \otimes \left(U_{QWP}S\right)\right)\left|\phi^{+}\right\rangle$, and the corresponding Fisher information turns out to be:

$$F_{q|\delta}{}' = \frac{\sin^2\left(4h_1+2\theta\right)\left[\cos\delta\sin 4h_2 - \cos 4h_2\sin\delta\sin 2\theta\right]^2}{1-\left[\cos 4h_2\left(\cos 4h_1\cos^2\frac{\delta}{2} + \cos\left(4h_1+4\theta\right)\sin^2\frac{\delta}{2}\right) + \sin 4h_2\sin\left(4h_1+2\theta\right)\sin\delta\right]^2}. \tag{S.2}$$

Different from the PSA polarimeter, Eq. (S.2) shows that we can obtain maximum value of 1 for any unknown phase retardance δ by flexibly selecting the measurement bases h_1 and h_2. This is an advantage of the Senarmont compensation method.

B. Photon source evaluation

The performance of the entangled photon source is evaluated by temporally removing the sample from the measurement arm. Two-photon polarization interference curves are collected under H-basis and D-basis. The raw interference visibilities (V=(Cmax-Cmin)/(Cmax+Cmin)) are calculated to be 99.75% ± 0.01% and 99.12% ± 0.01% for H- and D-basis, respectively, well above the 71% indicated by the Bell inequality. In addition, the CHSH inequality is measured to quantify the entanglement quality. The S-parameter is calculated to be 2.754 ± 0.006191 in 10 s, violating the CHSH inequality by 122 standard deviations. With a 2mW pump input, the maximum coincidence counts can achieve 2×10^4 cps, with total collecting efficiencies of the control and measurement arms to be 13.68% and 14.23%, respectively.

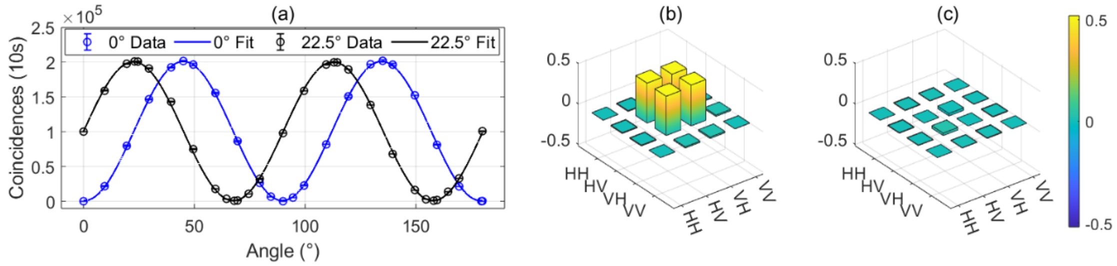

Fig.S1 Entangled photon source properties.

(a) two-photon polarization interference curve; **(b)&(c)** real and imaginary part of reconstructed density matrix.

C. Calculation Process of Improved Four-step Phase-shifting Method

Here we show the improved four-step phase-shifting method combined with Eq. (1). When the HWP in the control arm is fixed at 45° while the compensated QWP is fixed at 0° (H-basis), the output photon numbers under four degrees are:

$$IH_{1(h_m=0^\circ)}=\frac{1}{8}\left(3+\cos4\theta+2\cos\delta\sin^2 2\theta\right),\quad IH_{2(h_m=22.5^\circ)}=\frac{1}{4}\left(1+\sin\delta\sin2\theta\right),$$

$$IH_{3(h_m=45^\circ)}=2\cos^2\theta\sin^2\frac{\delta}{2}\sin^2\theta,\quad IH_{4(h_m=62.5^\circ)}=\frac{1}{4}\left(1-\sin\delta\sin2\theta\right). \tag{S.3}$$

Therefore, we have:

$$AH=\frac{IH_1-IH_3}{IH_1+IH_3}=\cos^2\frac{\delta}{2}+\cos4\theta\sin^2\frac{\delta}{2},$$

$$BH=\frac{IH_2-IH_4}{IH_2+IH_4}=\sin\delta\sin2\theta. \tag{S.4}$$

The corresponding phase retardance and optical axis can be derived from the above parameters:

$$\delta_H=\arccos\left[\frac{1-BH^2-AH}{AH-1}\right],\quad \theta_H=\frac{1}{2}\arcsin\left(\pm\sqrt{\frac{(AH-1)^2}{2-2AH-BH^2}}\right). \tag{S.5}$$

Note that the measurable phase retardance range is confined to $[0,\pi]$, we could decide the sign before the square root in Eq. (S.5) based on the sign of BH in the Eq. (S.4). In this situation, we have $\theta_H\in[-\pi/4,\pi/4]$.

Similarly, when the HWP in the control arm is fixed at 22.5° while the compensator at 45° (D-basis), the output photon numbers under four degrees are:

$$ID_{1(h_m=0^\circ)}=\frac{1}{4}\left(1+\sin\delta\cos2\theta\right),\quad ID_{2(h_m=22.5^\circ)}=\frac{1}{4}\left(1+\cos\delta\cos^2 2\theta+\sin^2 2\theta\right),$$

$$ID_{3(h_m=45^\circ)}=\frac{1}{4}\left(1-\sin\delta\cos2\theta\right),\quad ID_{4(h_m=62.5^\circ)}=\frac{1}{2}\cos^2 2\theta\sin^2\frac{\delta}{2}. \tag{S.6}$$

Therefore, we have:

$$AD=\frac{ID_1-ID_3}{ID_1+ID_3}=\sin\delta\cos 2\theta,$$

$$BD=\frac{ID_2-ID_4}{ID_2+ID_4}=\cos^2\frac{\delta}{2}-\cos 4\theta\sin^2\frac{\delta}{2}. \tag{S.7}$$

The corresponding phase retardance and optical axis can be derived from the above parameters:

$$\delta_D=\arccos\left[\frac{1-AD^2-BD}{BD-1}\right],\ \theta_D=\frac{1}{2}\arccos\left(\pm\sqrt{\frac{(BD-1)^2}{2-2BD-AD^2}}\right). \tag{S.8}$$

By combining Eq. (S.4) and (S.7), the sign of $\sin 2\theta$ and $\cos 2\theta$ can be determined. Therefore, the measurable range of the optical axis angle θ is extended to $[0,\pi]$. Besides, the phase retardance is calculated from:

$$\delta=\arccos\left[AH+BD-1\right]. \tag{S.9}$$

D. System calibration

Based on the above entangled-photon source, we measure a commercial QWP (808 nm, MFOPT Ltd.) to calibrate the angle of each polarization-correlated optical component and test the measurement accuracy of the system. The measurement accuracy refers to the difference between the measured image and the standard reference image. The standard reference image of the commercial QWP is derived from a commercial automatic polarimeter (NMV-3000S, Suzhou PTC Optical Instrument) with a retardation measurement accuracy of ±1nm.

After testing, the measurement results of the commercial QWP, including the retardance and the optical axis angle over the whole surface, are shown in Fig.S2.

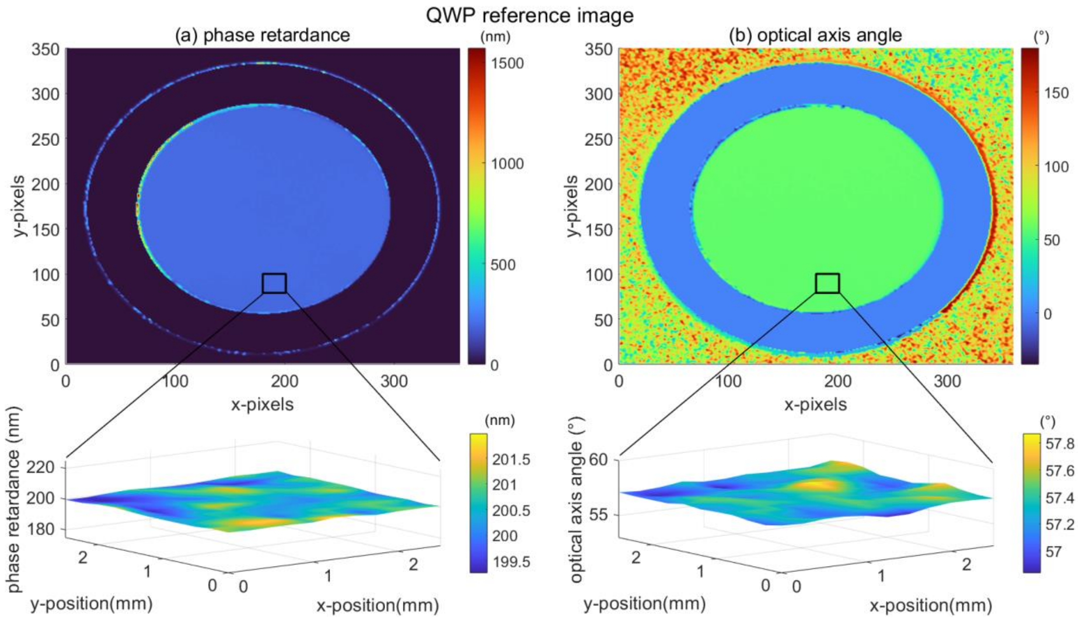

Fig. S2 Reference measurement results of a tested QWP. (a)&(b) Phase retardance and Optical axis angle of the entire surface of the QWP with the inserts show the 2.5 mm × 2.5 mm region stressed by the black rectangle.

In Fig. S2, the bottom two insects are the parts that stressed in the upper figures by the black rectangles. They cover an area of 2.5 mm × 2.5 mm with total 11 × 11 pixels, which is accord with our quantum imaging system. The average retardance value in this area is 200.51±0.12 nm, while the average optical axis angle is 57.29±0.05°. These values will be used as the standard reference values in the following analysis, since the commercial instrument using strong light incidence offers sufficient measurement accuracy.

The results obtained from measuring the same QWP using a quantum system are shown in the Fig. S3 of the revised version, with 11 × 11 pixels also taken in a 2.5 mm × 2.5 mm area. The average measurement results for retardance and angle are 202.88±0.51 nm and 56.9±0.06°, respectively, with estimated deviations from the standard reference values of 1.18% and 0.69%.

To further quantify the accuracy of the system, we compared the measurement results of the quantum measurement system with those of the reference system pixel-by-pixel using image quality assessment metrics. In between, the simplest and most widely used one is the Root Mean Square Error (RMSE), which calculates the square intensity differences between the estimated image and the reference image for each pixel. The calculation formula is:

$$\mathrm{RMSE}=\sqrt{\frac{1}{\mathrm{mn}}\sum_{i=1}^{m}\sum_{j=1}^{n}\left(I(i,j)-K(i,j)\right)^{2}}\ . \tag{S.10}$$

where m and n are the number of rows and columns of the image, while $I(i,j)$ and $K(i,j)$ are the intensity values of the reference image and the estimated image at the position (i,j), respectively. By substituting the reference values from Fig.S2 and the measured values from Fig.S3 into this formula, we obtain an RMSE value of 3.1003±0.3896 nm for the retardance and an RMSE value of 0.4992±0.1402° for the angle measurement.

This comparison demonstrates that the measurement accuracy of the experimental retardation can reach the nanometer level, and the angle can be accurately measured within 1°.

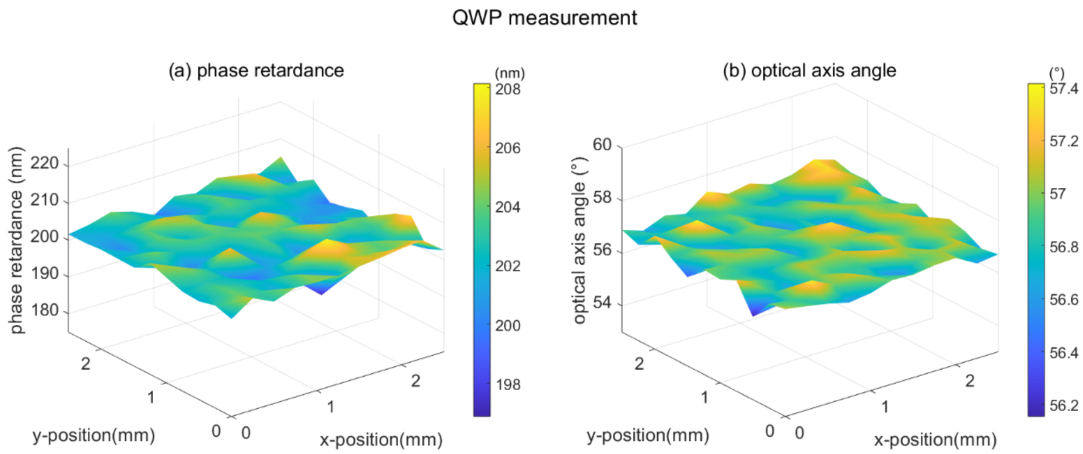

Fig.S3 Measurement result of a standard QWP. **(a)** phase retardance and **(b)** optical axis angle