Quantitative complementarity of wave-particle duality

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To test the principle of complementarity and wave-particle duality quantitatively, we need a quantum composite system that can be controlled by experimental parameters. Here, we demonstrate that a double-path interferometer consisting of two parametric downconversion crystals seeded by coherent idler fields, where the generated coherent signal photons are used for quantum interference and the conjugate idler fields are used for which-path detectors with controllable fidelity, is useful for elucidating the quantitative complementarity. We show that the quantum source purity $\mu_s$ is tightly bounded by the entanglement $E$ between the quanta and the remaining degrees of freedom by the relation $\mu_s = \sqrt{1 - E^2}$, which is experimentally confirmed. We further prove that the experimental scheme using two stimulated parametric downconversion processes is an ideal tool for investigating and understanding wave-particle duality and Bohr’s complementarity quantitatively.

INTRODUCTION

Bohr’s principle of complementarity (1) was initially a qualitative statement about mutually exclusive but equally real properties of a single quantum object, such as photons, electrons, etc. (2). Later in 1979, Wooters and Zurek (3) proposed the wave-particle duality concept, as a quantitative version, i.e., experimentally testable complementarity relation, in its best-known representative. They formulated the total interference pattern of a photon within a double-slit experiment using information-theoretical approaches, which shows a monotonic change between wave-like behavior (sharpness of fringe pattern) and particle-like behavior (photon’s trajectory). After that, by considering an unbalanced two-beam interferometer (4), where two beams have unequal intensities, the concept of path predictability was introduced as a quantitative measure of the a priori which-path knowledge. This predictability limits the amount of visibility $V_0$ that can be achieved in an interferometer according to the complementarity inequality relation. The inequality becomes an equality if the quantum object can be described as a pure state, which is the case that the wave-particle duality becomes a quantitative statement. This relation has been generalized to the composite systems where which-way detectors are in place (5). Then, another property called path distinguishability $D$ (interference visibility $V$) takes the role of a priori path predictability $P$ (a priori visibility $V_0$) representing the particle (wave) nature of the composite quantum system as described in detail for double-path interferometers (6–8), multipath interferometers (9–11), and delayed choice quantum erasing schemes (12–17).

Recently, systematic approaches to establishing quantitative complementarity relations in various composite quantum systems have been reported (8, 18–21). In generalized complementarity relations, multipartite realities mutually exclude single-partite properties of the subsystems, and the complementarity was shown to be quantified by entanglement measures between subsystems (8). For example, the entanglement measure appears in the form of the concurrence for a bipartite system, which becomes an essential entry in the quantitative complementarity relation consisting of path predictability $P$, visibility $V$, and entanglement $E$ (19). The $E$ can be a measure of entanglement between the quanta and its internal states (22) or polarization states (23) or spin states (24) or which-path detector states (11). Here, the term “quanton” that was coined by Bunge (25) is used to refer to a generic quantum system without using words such as “particle” or “wave” (26). Although these quantitative complementarity relations proposed recently require an experimental system enabling one to measure not only the entanglement measure ($5, 8, 9$) but also the quantum coherence reflecting interference contrast or visibility (27, 28) of a specific composite system, a lack of composite system whose quantum states can be experimentally controlled makes it challenging to characterize wave-particle duality involving the well-known interference fringe visibility and path distinguishability. More specifically, an analytical model of the superposition state of the quanta as a subsystem of the composite system with well-defined which-path detectors or source impurity states is still missing for complete experimental verification of quantitative complementarity.

Here, we show that our quantum optical system (see Fig. 1 for a schematic representation of the setup) that was used to demonstrate frequency-comb single-photon interferometry is suitable for studying and confirming the quantitative complementarity relations reported recently by Qian and Agarwal (29). We used a pair of identical but otherwise independent nonlinear quantum sources, i.e., parametric downconversion (PDC) crystals, that are pumped by synchronized optical frequency-comb lasers with the same amplitudes. The PDC processes of the two nonlinear crystals were simultaneously stimulated by highly coherent lights with different but controllable amplitudes, which were generated from a single coherent laser with an extremely narrow bandwidth (see Materials and Methods). Using this pair of stimulated PDC quantum sources, we could generate a coherence-tunable superposition state of signal single photons and conjugate single photon–added coherent states (SPACS) (30) in an idler mode, where the idler photon states turned out to play the role of impurity states degrading the source purity of quanta, i.e., signal photons or which-path detectors (11) with adjustable fidelity $F$, if the outcome idler photons are experimentally measured. We further prove that the source purity, denoted as $\mu_s$, introduced by Qian and Agarwal (29) is related to the entanglement measure $E$ representing the quantum correlation between the quanta and path detectors as $\mu_s = V_1 - E^2$. Also, the interference visibility $V$ of the quanta, i.e., signal photons, is connected to the a priori visibility as $V = FV_0$, where the proportionality constant is the fidelity determined by the overlap of the entangled idler photon states that are not necessarily orthogonal to each other. We show that our

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double-path interferometer will be of use to control the coherence of single-photon states from a superstition of pure states with both maximum visibility and pure source character to a composite state with degraded source purity.

RESULTS

Double-path interferometer with two single-photon sources

The composite quantum system that will be referred to as the entangled nonlinear bi-photon source (ENBS) model is shown in Fig. 1. The ENBS consists of two spatially separated type 0 phase–matched spontaneous PDC (SPDC) crystals (Fig. 1), i.e., periodically poled lithium niobate crystals (PPLNs) (30), that are the entangled signal single-photon generation sources and the conjugate idler photons acting like which-path detectors (11). Hereafter, we shall refer to the signal photons as quantons because they are subject to a double-path interferometric detection in the ENBS (see the Supplementary Materials). The idler modes of two PPLNs are seeded by weak lasers in coherent states \(|\alpha_1\rangle\) and \(|\alpha_2\rangle\), which enables us to control the overlap of the two idler states precisely. The generated signal photons are used for double-path quantum interference experiments, whereas their conjugate idler photons in a SPACS provide the which-path information. By adjusting the seed beam amplitudes, we could control the overlap between the SPACS and the single-photon non-added coherent state (31, 32). The signal photons propagate in free space following precisely the same plane-wave spatial modes of the idler fields, which contrasts with the diffraction waves in typical double-slit experiments, which allows us to achieve almost perfect single-photon visibility (30). It should also be noted that unlike the previous cases using path detectors for extracting which-path information of the quanton, the single photons do not have to interact with external devices (5) on their paths from the corresponding source (PPLN) to a single-photon detector (PD) (Fig. 1).

In our ENBS, the energy and momentum conservation relations at two PPLNs are assumed to be matched perfectly between the pump, signal, and idler fields, i.e., \(\omega_p = \omega_s + \omega_i\). The seed beam frequency in an idler mode is also matched well within the emission spectrum of the idler fields. Let us consider the case that only one signal photon is generated from either PPLN1 or PPLN2 at a time, which is achievable by reducing the mean photon number of stimulating coherent seed laser. The conjugate idler photon can then be considered to be a which-path detector because a pair of signal and idler photons must be simultaneously generated by annihilation of a single pump photon via an SPDC process (see note S1) (33). We demonstrated that this double-path interferometer could generate a superposition state of signal single photons (see note S2 for a detailed description of the experimental setup). When a single quanton, signal single photon, emerges either one of the two PPLNs pumped by the same laser with equal intensities but with different coherent seed lasers with \(|\alpha_1| \neq |\alpha_2|\), the composite system can be represented by the following superposition state

\[
|\psi\rangle = c_1|\alpha_1\rangle|\alpha_2\rangle|1,0\rangle_s + c_2|\alpha_1\rangle|\alpha_2\rangle|0,1\rangle_s
\]  

(1)

where we used the following notation \(|p, q\rangle_s = |p\rangle_s|q\rangle_s\) for \((p, q) \in \{0, 1\} = \{\text{vacuum, single photon}\}\) for each single photon state. The normalization constants are given as \(c_j|\alpha_1, \alpha_2\rangle = \sqrt{1 + |\alpha_j|^2}/\sqrt{2 + |\alpha_1|^2 + |\alpha_2|^2}\), where \(\sum_{j=1}^2 |c_j|\alpha_1, \alpha_2\rangle|^2 = 1\). In Eq. 1, the idler states entangled with the signal states are

\[
|m_1\rangle = |\alpha_1, 1\rangle_{i1} |\alpha_2\rangle_{i2}
\]  

(2A)

and

\[
|m_2\rangle = |\alpha_1\rangle_{i1} |\alpha_2, 1\rangle_{i2}
\]  

(2B)

Here, \(\hat{a}^\dagger_j\) is the creation operator of the jth idler field, and \(|\alpha_j, 1\rangle_{i_j} = \frac{\hat{a}^\dagger_j|\alpha_j\rangle}{\sqrt{1 + |\alpha_j|^2}}\) is the SPACS of the jth idler field created by the stimulated PDC of PPLN_j (30). As can be seen in Eqs. 2A and 2B, \(|m_j\rangle\) is the product idler-state, where the jth idler field \(i_j\) is in the

![Fig. 1. Double-path single-photon interferometer with controllable source purity used in our ENBS model.](http://advances.sciencemag.org/)
SPACS $|\alpha_i,1\rangle_0$, while another idler field $|\alpha_s,1\rangle$ is in the incident coherent state $|\alpha_s,1\rangle_0$. In principle, the generation rate of the quantum composite state in Eq. 1 depends both on the pump and seed beam intensities, but here, we consider the case when the pump beam intensities are the same at the two PPLN crystals in the weak pump limit for simplicity (see note S2 for experimental parameters).

The signal single-photon states $|1,0\rangle_s$ and $|0,1\rangle_s$, which constitute the subsystem of quanta of the composite system, ENBS, form an orthonormal set. If the idler photons are detected with $D_A$ and $D_B$ in Fig. 1, then the two states $|m_1\rangle$ and $|m_2\rangle$ can be considered as which-path detector states (11). Such an entanglement between the quantum and which-path detector states is a fundamental requirement of the process of measurement as laid down by von Neumann (8). However, in our experiments, we did not have to measure the idler photons generated from either PPLN$_1$ or PPLN$_2$, since we measured the fringe visibility from the single-photon counts during a given measurement time. Therefore, the idler states $|m_1\rangle$ and $|m_2\rangle$ entangled with signal photon states $|1,0\rangle_s$ and $|0,1\rangle_s$, respectively, correspond to the states describing the remaining degrees of freedom other than the signal single-photon states.

**Superposition state of quanta pure states**

The superposition state described in Eq. 1 can be realized with the ENBS, as experimentally demonstrated in (30). Because of the entanglement between the signal and idler modes, which is the intrinsic property of PDC processes, the quanta, signal single photons, are not in a pure state. However, upon increasing the intensities of the two seed beams, each in an idler mode, the two idler states $|m_1\rangle$ and $|m_2\rangle$ become similar to each other and indistinguishable, i.e., $|\alpha_i\rangle \rightarrow |\alpha_s\rangle \rightarrow |\alpha_i\rangle$ as $|m_1\rangle \rightarrow |m_2\rangle$. Experimentally, as the average photon numbers of the two seed beams increase, it becomes impossible to identify whether a PDC-generated idler photon was produced by PPLN$_1$ or PPLN$_2$. In this limiting case, the superposition state in Eq. 1 reduces to

$$|\psi\rangle_0 = (c_1|\alpha_s,1\rangle + c_2|\alpha_i,1\rangle) |0,1\rangle_s.$$  

In this limit, the quantum, signal single photon state, becomes a superposition of pure states. After tracing over the idler states, we have that the reduced density operator of the quanton, which is spanned by the basis states $\{|1,0\rangle_s, |0,1\rangle_s\}$, is

$$\rho(\alpha_1, \alpha_2) = \begin{bmatrix} |c_1|^2 & c_1^*c_2 \\ c_1c_2^* & |c_2|^2 \end{bmatrix}$$  \(4\)

where, in the case of the ENBS

$$\rho_{jj} = \frac{1 + |\alpha_j|^2}{2 + |\alpha_1|^2 + |\alpha_2|^2}$$  \(5A\)

and

$$|\rho_{12}| = \sqrt{\frac{(1 + |\alpha_1|^2)(1 + |\alpha_2|^2)}{2 + |\alpha_1|^2 + |\alpha_2|^2}}$$  \(5B\)

For this pure state for the quanta, we find that the a priori predictability and the a priori visibility are given by, respectively

$$p^2 = 1 - \left(\sum_{i\neq j} \sqrt{\rho_{ii}\rho_{jj}}\right)^2 = 1 - 4\rho_{11}\rho_{22}$$  \(6A\)

and

$$V_0 = \sum_{i\neq j} \rho_{ij} = 2|\rho_{12}|$$  \(6B\)

They satisfy the wave-particle duality equality, $p^2 + V_0^2 = 1$, for a pure state, which has been observed by Greenberger and Yasin (4) and Wootters and Zurek (3).

In general, when the composite system is described by the superposition state in Eq. 1 instead of the pure state for the quanta in Eq. (3), $D$, a quantitative measure of the distinguishability of the ways or of the amount of which-way information that has become available, and $V$, the fringe visibility, are representative quantities reflecting the particle and wave natures of the quanta, where they are defined as

$$D^2 = 1 - \left(\sum_{i\neq j} \sqrt{\rho_{ii}\rho_{jj}}\right)^2 = 1 - 4\rho_{11}\rho_{22}F^2$$  \(7A\)

and

$$V = \sum_{i\neq j} \rho_{ij} |\langle m_i | m_j\rangle| = V_0 F$$  \(7B\)

Fig. 2. Numerical simulations of various components versus $|\alpha|$. Parameters, $D^2, P^2, E^2, V, F$, and $\mu_s^2$, appearing in wave-particle duality relation versus $|\alpha|$, the amplitude of seed beam with $|\alpha_1| = |\alpha_2| = |\alpha|$. (A) and $|\alpha_1| = |\alpha_2| = 2 |\alpha|$. (B). Colors associated with the measures are the same in (A) and (B).
Here, we introduce the fidelity $F = \langle \langle m_1 | m_2 \rangle \rangle$ that is the overlap of the two idler states entangled with $|1,0\rangle$ and $|0,1\rangle$. For the ENBS, the fidelity is the function of eigenvalues of the two coherent states describing the seed beams (30), i.e.

$$F = |\langle \langle \alpha_1 | \alpha_2 \rangle \rangle|^{\alpha_1 \alpha_2} = \frac{|\alpha_1| |\alpha_2|}{\sqrt{1 + |\alpha_1|^2 + |\alpha_2|^2}}$$

(8)

As demonstrated experimentally in (30), we could control the magnitudes of these two eigenvalues, $|\alpha_1|$ and $|\alpha_2|$, arbitrarily and independently to vary the quantum state from the superposition state of the composite system in Eq. 1 to the pure state of signal single-photons in Eq. 3.

**Second-order field correlation measurement with ENBS**

In Eqs. 6A, 6B, 7A, 7B, and 8, the definitions of a priori probability, interference visibility, and fidelity were provided. Here, we prove that they are consistent with the experimental observable that is the contrast of interference pattern obtained with our ENBS. To characterize the interference of signal photons, we use the approach in the Heisenberg picture as shown in (34, 35). The positive (negative)–frequency part of the signal electric field operator, $E^{(+)}_D (E^{(-)}_D)$, generated by crystals PPLN$_{1}$ and PPLN$_2$ and incident on PD in Fig. 1 via the symmetric beam splitter BS3, can be written as (35)

$$E^{(+)}_D = i \alpha_1 \hat{a}_1 e^{i\phi_1} + \alpha_2 \hat{a}_2$$

$$E^{(-)}_D = -i \alpha_1^* \hat{a}_1^* e^{-i\phi_1} + \alpha_2^* \hat{a}_2^*$$

(9A)

(9B)

where $\hat{a}_j$ is the annihilation operator for the signal photons emitted from PPLN$_j$, $\hat{a}_j^*$ is the creation operator for the conjugate idler photons, and constant $\alpha_j$ is proportional to the nonlinear susceptibility of PPLN$_j$ and the intensity of pump field incident on the PPLN$_j$. The factor $ie^{i\phi}$ with $\phi_j = k_j \Delta x_j$ in Eqs. 9A and 9B, where $k_j$ is the wave vector of the signal field, and $\Delta x_j$ is the path difference controlled by the mirror in Fig. 1, accounts for the relative phase change of the signal field to the detector via reflecting off the mirror.

Treating the PD as a perfectly efficient, fast, and broadband photodetector, we could show that the signal photon count rate $g^{(2)}_\alpha(0)$, i.e., second-order field correlation $R_D = \langle E^{(-)}_D E^{(+)}_D \rangle$ at the detector is proportional to the normal–ordered expectation value in (35, 36)

$$R_D = |v|^2 |\langle i_{10}^t i_{10}^t \rangle + \langle i_{20}^t i_{20}^t \rangle + |\alpha_1|^2 |\alpha_2|^2 + i e^{-i(\phi_1 + \phi_2)} |\alpha_1|^2 |\alpha_2|^2 + i e^{-i(\phi_1 + \phi_2)} |\alpha_1|^2 |\alpha_2|^2 + i e^{-i(\phi_1 + \phi_2)} |\alpha_1|^2 |\alpha_2|^2$$

(10)

where, for simplicity, we take $v_1 = |v| |e^{i\phi_1}|$ and $v_2 = |v| |e^{i\phi_2}|$, consistent with the equal powers of the pump fields incident on the PPLN$_1$ and PPLN$_2$ but with a phase change $\phi_j$ accrued by the mirror (see Fig. 1), i.e., $\phi_j = k_j \Delta x_j$. As shown in (30, 34, 35), we assume that the downconversion efficiency is small so that the higher–order terms, except for the lowest–order terms, in $v$ can be ignored. Note that, although $R_D$ is the normal–ordered correlation function ($E^{(-)}_D E^{(+)}_D$), it depends on the anti–normal–ordered idler–mode operators (Eq. 10). Therefore, the positive (negative)–frequency part of the signal electric field operator depends on the negative (positive)–frequency part of the idler field operator.

To account for the signal single-photon interference, we use $i_{10}^t = i_{10}^t + \alpha_1 e^{i\phi_1}$ and $i_{20}^t = i_{20}^t + \alpha_2$ in Eq. 10, where $i_{10}^t$ and $i_{20}^t$ are annihilation operators for the vacuum idler modes incident on the two PPLNs (35). Here, $\alpha_j$ is the complex amplitude describing the seed laser fields injected into the PPLN$_j$ with the phase factor $e^{i\phi_j}$ due to the presence of a mirror, i.e., $\phi_j = k_j \Delta x_j$. Note that there is no induced coherence without induced emission between the generated signal fields (36–39) because the vacuum fields at two crystals are associated with distinct modes in this parallel arrangement of two PPLNs. Therefore, $\langle i_{10}^t i_{10}^t \rangle = \langle i_{20}^t i_{20}^t \rangle = 0$, and from Eq. 10, we have

$$R_D = |v|^2 |\langle i_{10}^t i_{10}^t \rangle + \langle i_{20}^t i_{20}^t \rangle + |\alpha_1|^2 |\alpha_2|^2 + i e^{-i(\phi_1 + \phi_2)} |\alpha_1|^2 |\alpha_2|^2$$

(11)

where we used the boson commutation relation for the idler fields, i.e., $[\hat{a}_j^\dagger \hat{a}_j] = \delta_{j\mu}$, and $\Delta \theta = \phi_j + \phi_1 + \phi_2$. Now, from the definition of $V = \frac{R_D^{\max} - R_D^{\min}}{R_D^{\max} + R_D^{\min}}$ (40) and from Eq. 11, we have

$$V = \frac{2 |\alpha_1| |\alpha_2|^2}{2 |\alpha_1| |\alpha_2|^2} \equiv V_0 F$$

(12)
where \( R_p^{\text{max}} \) (or \( R_D^{\text{min}} \)) is the maximum (minimum) of the interference fringe when \( \Delta \theta \) is varied within the single-photon interferometer in Fig. 1. The second equality in Eq. 12 is validated by using the expressions for \( V_0 \) and \( F \) given in Eqs. 6B and 8, respectively. The relation \( V = V_0F \) indicates that the degradation of the source purity, i.e., reduction of fringe visibility, could result from a low fidelity. As an example, let us consider the particular case when \( | \alpha_1 | = | \alpha_2 | = | \alpha | \). The fringe visibility becomes identical to the fidelity \( F \), i.e.,

\[
V = F = \frac{| \alpha |^2}{1 + | \alpha |^2}.
\]

In this case, although the visibility can vary from 0 to 1, the a priori visibility is always unity, i.e., \( V_0 = 2 | \rho_{12} | = 1 \), regardless of the magnitude \( | \alpha | \). Thus, the fidelity can be considered to be an experimental parameter enabling to control the purity of the superposition state of quanta, single signal photons in the case of the ENBS, from \( V = V_0 \) (pure state) to \( V = 0 \) (ineffective state).

Hereafter, with the ENBS system described by Eqs. 1 and 3, we shall show that the wave-particle duality or triality relation for such a composite system can be quantitatively studied and that various relations among a priori predictability, a priori visibility, distinguishability \( D \), fringe visibility \( V \), entanglement \( E \), and source purity \( \mu \) can be expressed in closed forms.

### Complementarity from a source point of view

To make single-photon interferometry experimentally feasible, one could prepare two trapped two-level atoms, with one of the two being in an excited state. Then, the spontaneously emitted single photons can produce an interference fringe. Such a fluorescence single-photon interference from entangled two-level atoms was first experimentally realized by Eichmann et al. (41, 42) with a laser beam exciting one of the two trapped \(^{199}\text{Hg}^+ \) ions. Later, Araneda et al. (43) carried out the experiment with trapped \(^{138}\text{Ba}^+ \) ions. However, the observed visibility was found to be significantly smaller than unity. To explain the deviation, Qian and Agarwal (29) considered the active role of remaining degrees of freedom other than the superposition state of a singly excited two two-level atoms. They introduced the concept of source purity, denoted as \( \mu_s \), and found an interesting Pythagorean relation between wave-particle duality and source purity, i.e.

\[
P^2 + V^2 = \mu_s^2
\]

where the so-called source purity is defined as \( \mu_s = \sqrt{2 \text{Tr}[\rho_s^2] - 1} \) with \( \rho_s \), being the reduced density matrix obtained by tracing over the states representing the remaining degrees of freedom. Here, note that the distinguishability \( D \) is defined as \( | \rho_{11} - \rho_{22} | / (| \rho_{11} + \rho_{22} |) \) in (29) [see equation 7 in (29)] is identical to \( P \) in the present work. In their analysis, two entangled two-level atoms with one of them excited scatter a single photon at a time with equal probability into path 1 or 2 in the dual-path interferometer (29). The superposition state of the pure system can be written as \( | \psi_0 \rangle = c_a | e_A g_B \rangle + c_b | g_A e_B \rangle \), where \(| e \rangle \) (\(| g \rangle \)) is the excited (ground) state of atom \( A \) or \( B \). In practice, however, pure states \(| e_A g_B \rangle \) and \(| g_A e_B \rangle \) cannot be easily realized due to different states associated with all the other remaining degrees of freedom and due to the presence of external parties. They, thus, considered a composite state \(| \psi \rangle = c_a | e_A g_B \rangle | m \rangle + c_b | g_A e_B \rangle | n \rangle \), where \( | m \rangle \) and \( | n \rangle \) represent two sets of quantum states reflecting the entangled degrees of freedom associated with the two atoms and even any unspecified external fields (29). Although the state \(| \psi \rangle \) is similar to any other superposition states of various composite systems, e.g., ENBS and double-path interferometer with path detectors (11), Qian and Agarwal (29) could not or needed not to specify the unknown quantum states \( | j \rangle \) for \( j \in \{ m, n \} \), much like most previous works. where the entangled which-path detector states were not specified to establish various wave-particle duality or triality relations (11).

The relationship between wave-particle duality measures (\( P \) and \( V \)) and source impurity \( (\mu_s) \) given in Eq. 13 led to a new interpretation of the wave-particle duality because the source purity of quanta can limit the totality of complementarity between the wave-like and particle-like behaviors of quanta. There is an analogy between our ENBS and the dual-path interferometer with two trapped ions emitting a single photon (41–43). In the experiments by Eichmann et al. (41, 42) and Araneda et al. (43), two trapped ions, with one of them excited by pump radiation, are the sources generating a single photon. Similarly, the two PDC crystals pumped by a common laser are the sources producing signal-idler photon pairs in the ENBS, except that the ENBS measures the interference of signal photons not idler photons. Because of this close analogy between these two seemingly different experiments, we can use the concept of source purity for quantitatively analyzing the experimental results reported in (30).

From Eq. 13 and using the expressions for the predictability and visibility in Eqs. 6A, 6B, 7A, 7B, and 8, one can find

\[
\mu_s = \sqrt{1 - 4 \rho_{11} \rho_{22} (1 - F^2)}
\]

When the two seed beam intensities are the same, i.e., \( | \alpha | = | \alpha_1 | = | \alpha_2 | \), the source purity becomes identical to fidelity, \( \mu_s = F \), which means that the upper limit of wave-particle duality equality is limited by the source purity that is identical to detector fidelity in the case of the ENBS with \( | \alpha_1 | = | \alpha_2 | \).

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**Fig. 4.** Fringe visibility \( V \) and a priori visibility \( V_0 \) as functions of \( \gamma = | \alpha_2 | / | \alpha_1 | \) and \( | \alpha | = | \alpha_2 | \). Blue symbols are experimental data taken from our recent paper (30) (see fig. S3). Experimental data coincide with the visibility \( V \) of Eq. 7B, not a priori visibility \( V_0 \) across the whole ranges of \( \gamma \) and \( | \alpha | \). This plot validates our analysis of the ENBS experimental results in terms of the wave-particle duality and quantitative complementarity relations. \( V \) and \( V_0 \) although they both reflect the wave-like nature of the quantum, are different from each other in the regions of \( | \alpha | < 5 \) and \( \gamma < 0.5 \) but becomes identical when \( | \alpha | > 1 \) and \( \gamma = 1 \).
Complementarity from a path detector point of view

Recent studies have also focused on completing wave-particle duality relations using entanglement and polarization. Eberly and coworkers (23, 44) showed that, in the classical optics regime, polarization should be taken into consideration in the two-slit interference experiment and the triality relation among the predictability, interference visibility, and concurrence. De Zela (45) investigated the relationship between polarization indistinguishability and entanglement. More recently, Qureshi (11) considered an n-path interference system with path detectors, which can be represented as $\langle \psi | = \sum_{j=1}^{n} | \phi_j \rangle | d_j \rangle$ where $| \phi_j \rangle$ is the state corresponding to the quanta taking the $j$th path and $\{|d_j\rangle\}$ are certain normalized states of the path detector. Here, it should be emphasized that $\{|\phi_j\rangle\}$ form an orthonormal set of quanta states, but the detector states $\{|d_j\rangle\}$ are not necessarily orthogonal to one another. Qureshi (11) showed that the distinguishability $D$, the a priori predictability $P$, and the entanglement $E$ between the quanta and path detector satisfy the Pythagorean relation $D^2 = P^2 + E^2$, where the entanglement is defined as

$$E^2 = (\sum_{i\neq j} |\langle \phi_i | P_j \rangle|^2 - (\sum_{i\neq j} |\langle \phi_i | P_j \rangle | \langle d_i | d_j \rangle |)^2 = 4 P_{11} P_{22} (1 - F^2)$$

(15)

For the ENBS, this entanglement between signal and idler modes can be expressed in terms of $|\alpha_1\rangle$ and $|\alpha_2\rangle$ of the injected seed beams.

From the definition of the coherence $C$, a measure of coherence of quanta, of the composite system (28, 46), generalized duality equality for the $n$-path interferometer ($n \geq 2$) was proposed as $D^2 + C^2 = 1$ or equally $P^2 + E^2 + C^2 = 1$. Note that their coherence $C$ is, by definition, identical to the fringe visibility $V$ in the present work so that their duality and triality equalities can be rewritten as $D^2 + V^2 = 1$ or equally $P^2 + E^2 + V^2 = 1$ with the notations used here. Although the importance of this entanglement in the quantitative complementarity relation was theoretically clarified in the previous work (11), the most important question about how to realize such an interferometer with well-defined detector states experimentally has not been addressed.

If we take the idler states $|m_1\rangle$ and $|m_2\rangle$ entangled with the pure states of signal single-photon $|1,0\rangle_s$ and $|0,1\rangle_s$, respectively, as the probe reporting us the which-source or which-path information, i.e., which PPLN generates a single single-photon, much like the which-path detector states $|d_1\rangle$ and $|d_2\rangle$ in a double-path interferometer, we can use the same relations discussed above, i.e., $D^2 = P^2 + E^2$ and $P^2 + E^2 + V^2 = 1$. From these and Eqs. 14 and 15, we find that the source purity $\mu_s$ introduced by Qian and Agarwal is bounded by the quantum-path detector entanglement measure $E$ as

$$\mu_s = \sqrt{1 - E^2}$$

(16)

Equation 16 is another interesting relationship that has not been discussed before. The entanglement between the quanta and path detector states can play a role in degrading the purity of the source (signal single photon) state. In the case of the ENBS, the two idler states could be viewed as which-path detector states entangled with quanta states.

Inserting Eqs. 5A and 5B into Eqs. 6A, 6B, 8, 14, and 15, we obtained closed-form expressions of $P$, $\mu_s$, $V$, $F$, $E$, and $\mu_s$ in terms of two experimentally controllable eigenvalues $|\alpha_1|$, and $|\alpha_2|$ of the coherent seed beams (see Fig. 2 for the corresponding plots and note S2).

Within the feasibility window of ENBS experiments, it becomes possible to address various duality relations between distinguishability $D$, a priori predictability $P$, visibility $V$, and source purity $\mu_s$. First, let us consider the case that there is no way, in principle, to distinguish the signal photon emitted from which source, PPLN1 or PPLN2. This case corresponds to the limit that the fidelity $F$ equals one or to the limit that both $|\alpha_1| \approx 1$ and $|\alpha_2| \approx 1$. Then, the ENBS composite system’s quantum state can be written as Eq. 3, which means that there is no entanglement between the quanta (signal) and path detector (idler) states. In this case, distinguishability $D$ and a priori predictability $P$ becomes identical, i.e., $D = P$. They depend only on the probabilities $p_{11}$ and $p_{22}$ for paths 1 and 2, respectively, that are constrained by the normalization condition $p_{11} + p_{22} = 1$. In addition, the fringe visibility $V$ becomes identical to the a priori visibility $V_0$ of the superposition state of pure states $|1,0\rangle$, and $|0,1\rangle$, i.e., $V = V_0$.

Reducing the seed beam intensities at the two nonlinear crystals, we could experimentally control the fidelity to be in the range of $0 \leq F < 1$. The elements in Qureshi’s Pythagorean relation $D^2 = P^2 + E^2$ can be quantitatively controlled by adjusting the overlap of the path detector states or the path detector fidelity $F$. Also, fringe visibility $V$ and a priori visibility $V_0$ that are two measures of quanta’s wave nature can be tuned simultaneously by varying $|\alpha_1|$ and $|\alpha_2|$ due to the relation $F = FV_0$ with Eq. 8. If $|\alpha_1| = |\alpha_2| = |\alpha|$ (Fig. 2A), i.e., two seed beam intensities are identical, then $p_{11} = p_{22} = 1/2$ so that $P = 0$. In this case, one cannot predict whether the quanta will take either path 1 or 2 regardless of the magnitude of $|\alpha|$. Then, we...
have $D = E = \sqrt{1 - F^2}$, where $F = \frac{|\alpha|^2}{1 + |\alpha|^2}$. When $|\alpha| \gg 1$, we again reach the limit $F = 1$, i.e., $D = E = 0$ and $V = 1$, indicating that the path detector loses its role to distinguish which path the quanta takes because the coherent state $|\alpha\rangle$ and SPACS $|\alpha_1\rangle$ overlaps nearly perfectly. Because the quantum taking either one of the two paths is not entangled with the which-path detector, i.e., $E = 0$, the quantum propagates as a perfect coherent wave with a priori visibility $V_0 = 1$ and fringe visibility $V = 1$.

If $|\alpha_1| \neq |\alpha_2|$ (Fig. 2B), all the measures appear to be different and play their roles in the wave-particle duality or triality relation. Evidently, we find the relations such as $D^2 = P^2 + E^2$ and $P^2 + E^2 + V^2 = 1$ hold in the whole range of $|\alpha_j|$, demonstrating that all the measures can be precisely controlled by the experimental parameters $|\alpha_j|$. Reinterpretation of the experimental results in (30)

On the basis of the analysis in this paper, the set of experiments in (30) emit single photons with equal probability but with controllable fidelity $F$ or source purity $\mu_s = F = V$. Our ENBS system allows us, in principle, to cover the entire parameter ranges not only of the fidelity $F$ but also of the other wave-particle properties. To demonstrate the quantitative complementarity established by Qian and Agarwal (29) and us in this paper $P^2 + V^2 = \mu_s^2 = 1 - E^2$ experimentally, one needs to measure the visibility $V$ from the single-photon interference across the two-dimensional parameter space of $|\alpha_1|$ and $|\alpha_2|$. From Fig. 3 (A and B), the quantitative complementarity relation $P^2 + V^2 = \mu_s^2 = 1 - E^2$ indicates that the quantum object propagates through the double-path interferometer partly as particle-like measured by $P$ and partly as wave-like measured by $V$, where their totality is bounded by the source purity $\mu_s = \sqrt{1 - E^2}$ or equally by the entanglement. Figure 4 shows the visibility $V$ in Eq. 7B in the two-dimensional space of two experimental parameters of $\gamma = \frac{|\alpha_1|}{|\alpha_2|}$ and $|\alpha| = |\alpha_2|$, where the blue symbols are the experimental data taken from (30). Also shown in Fig. 4 is the a priori visibility $V_0$ as a function of $|\alpha|$ and $\gamma$ for comparison.

In our ENBS system (30), the stimulated downconversion rate can be controlled easily from the same order of SPDC to a much higher level than that of SPDC (34, 35). Furthermore, the source is free from the decoherence issue because the spectrum of the quantum is determined by coherent seed beams. By adjusting the seed beam photon numbers $|\alpha_j|^2$ while fixing the pump beam intensity $|\alpha|^2$, we have independent knobs to control both the particle and wave characters. These degrees of freedom in experiments enable one to enjoy additional flexibility and controllability of the quantum coherence (fidelity) of the signal photons and their emission rate with two independent (orthogonal) knobs $|\alpha_j|^2$ and $|\alpha|^2$. In short, we show that the wave-particle duality (triality) equality, i.e., quantitative complementarity, can be tested with our ENBS system, where the wave-like and particle-like behaviors of the quantum (signal photon) are tunable quantities through the experimentally adjustable path detector fidelity $F$ ranging from 0 to 1.

**DISCUSSION**

In summary, the wave-particle duality and the quantitative complementarity $P^2 + V^2 = \mu_s^2$ were analyzed and tested using our ENBS model, where the superposition states of the quanta (signal photons) are entangled with conjugate idler states in a controllable manner through the fidelity $F$. We find that the source purity $\mu_s$ depends on the entanglement $E$ by the following relation $\mu_s = \sqrt{1 - E^2}$. We showed that a priori predictability $P$, visibility $V$, and entanglement $E$ (thus, source purity $\mu_s$ and fidelity $F$ in our ENBS model) depend only on the seed beam photon numbers. This points to potential application in all-optical preparation of distant entangled state. Last, we anticipate that the interpretation based on the double-path interferometry experiments with ENBS will have fundamental implications for better understanding the principle of complementarity and the wave-particle duality relation quantitatively, leading to demystifying Feynman’s mystery for the double-slit experiment explanation based on the quantum mechanics (47, 48).

**MATERIALS AND METHODS**

A schematic diagram of the experimental setup used in (30) and in our ENBS system discussed in the results section of the main text is depicted in Fig. 5. Details of the experimental setup, not specified types of equipment, and the experimental parameters can be found elsewhere (30). However, a brief description of the setup is presented here. Pump laser is an optical frequency comb (pump comb) with a center wavelength of 530 nm, repetition rate of $f_{rep} = 250$ MHz, and carrier-envelope offset frequency $f_{CEO} = 20$ MHz, pulse width $\Delta t = 10$ ps, and optical spectral width of $\Delta \lambda = 3$ nm. Seed laser is a highly coherent continuous-wave laser at $\lambda_s = 1542$ nm and line width of an order of hertz (manufacturer specification). Two identical PPLN nonlinear crystals (a length of 7.9 mm, a poll period of 7.3 μm, and the phase-matching temperature of 121.5 °C) are used and phase-matched for type 0 SPDC process using pump photons at $\lambda_p = 530$ nm to generate identical pairs of signal photons at $\lambda_s = 807$ nm and idler photons at 1542 nm within optical spectral widths of $\Delta \lambda_s = \Delta \lambda_i = 5$ nm. After injecting the idler seed beam ($\lambda_i = 1542$ nm) into the two crystals, both optical spectral widths $\Delta \lambda_s$ and $\Delta \lambda_i$ reduce below the instrument resolution limit of 0.1 nm. To measure the single-photon count rate at the fixed pump and idler beam amplitudes and $\Delta \theta$ in Eq. 11 in the main text, the integration time $T_I$ of the photodetector is set to be $T_I = 10$ ms. The pump amplitude $v$ and idler amplitudes $\alpha_1$ and $\alpha_2$ are adjusted to make the signal photon counts per second less than $5 \times 10^6$, which means that at every 50 pump pulse, only one single-photon count is recorded on average. This experimental result confirms a deep single-photon generation regime. To scan the full range of coherence $C$ from 0 to 1, we needed to adjust $|\alpha_1|^2$ and $|\alpha_2|^2$ up to 100 for a fixed pump power of less than 10 mW. To determine the fringe visibility $V$ using the single-photon counting rate of Eq. 11, we vary $\Delta \theta = \theta_p + \theta_i + \theta_o$ over 2π by one of the three propagation length differences, i.e., $\Delta x_p$, $\Delta x_o$, or $\Delta x_i$ in Fig. 5, which was achieved by using one of the three different PZT (piezoelectric transducer)–mounted mirror mounts.

**SUPPLEMENTARY MATERIALS**

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/7/34/eabi9268/DC1

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