Quantum-state purity of heralded single photons produced from frequency-anticorrelated biphotons

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We analyze the quantum-state purity of heralded single photons produced from frequency-anticorrelated biphotons. We find that the quantum-state purity in the time-frequency domain depends strongly on the response time uncertainty of the trigger-photon detector that heralds the generation of its paired photon. If the trigger response time is much shorter than the two-photon coherence time, the time-frequency quantum-state purity of heralded single photons approaches unity and the heralded single photon is in a nearly pure state. If the trigger response time is much longer than the two-photon coherence time, the heralded photon is then projected onto a mixed state. Making use of the time-frequency entanglement, heralded single photons with a well-defined temporal wave function or a frequency superposition state can be produced and engineered. This time-frequency entanglement allows for shaping heralded single photons through nonlocal spectral modulation.

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I. INTRODUCTION

Correlated photon pairs can be used to generate heralded single photons: the detection of one photon heralds the presence of the remaining one and projects it onto a single-photon Fock state. Spontaneous parametric down conversion (SPDC, $\chi^{(2)}$ nonlinear process) [1,2] and spontaneous four-wave mixing (SFWM, $\chi^{(3)}$ nonlinear process) [3,4] have been two standard methods for producing paired photons. When driven by continuous-wave (CW) pump laser fields, the photon pairs generated from these parametric processes are time-frequency entangled because of the energy conservation raised from the time-translation symmetry, i.e., the sum of the frequencies of paired photons is fixed, while individual photons have their bandwidths. For a typical wide-band (>THz) SPDC source with such a frequency anticorrelation, its short coherence time cannot be resolved by a state-of-the-art single-photon detector, whose relative slow temporal response is modeled as tracing over the trigger photon and thus projects the heralded photon into a mixture of frequency modes [5,6]. Applying spectral filtering to obtain an approximate pure single-photon state results in a dramatically reduced photon rate by discarding photons outside of the filter frequency mode [7,8]. Currently, the widely adapted approach for obtaining a heralded pure single-photon state is to eliminate the entanglement and generate photon pairs with a factorable (frequency-uncorrelated) joint spectrum driven by pulsed lasers [5,6,9–15].

There has been considerable work done toward reducing the biphoton bandwidth and increasing the coherence time with cavity-enhanced SPDC [16–18] or cold-atom-based SFWM [4,19–21]. Most recently, the biphoton coherence time has been prolonged to several microseconds [22,23]. Heralded single photons from narrow-band biphotons can interact with atoms [24–26] and cavities [27,28] coherently. These recent experiments have strongly evidenced that the heralded single photons from narrow-band frequency-anticorrelated photon pairs have coherent wave packets, but their quantum-state purities have never been formally and theoretically justified. In this article, we analyze the quantum-state purity of heralded single photons produced from frequency-anticorrelated biphotons. We point out that an ideal heralding process with an instantaneous trigger-photon detection projects the remaining frequency-anticorrelated photon onto a pure quantum state, which is a superposition state of its frequency components. When the trigger-photon detection has a finite response time, it degrades the purity of the single photons. If the trigger response time is much longer than the two-photon coherence time, the heralded photon is projected onto a mixed state. If the trigger response time is much shorter than the two-photon coherence time, we find that the time-frequency quantum-state purity of heralded single photons approaches near unity without any need of spectral filtering.

II. THEORY: QUANTUM-STATE PURITY

Let us start with the biphoton state (in time-frequency space) of a frequency-anticorrelated photon pair [4],

$$|\Psi_{12}\rangle = \int d\Omega \Phi(\Omega) \hat{a}_2^\dagger(\omega_{20}-\Omega)\hat{a}_1^\dagger(\omega_{10}+\Omega)|0\rangle, \quad (1)$$

where $|0\rangle$ is the vacuum state, and ω_{10} and ω_{20} are the central angular frequencies of photons 1 and 2. $\hat{a}_1^\dagger$ and $\hat{a}_2^\dagger$ are the field creation operators. $\Phi(\Omega)$ is the two-photon joined spectrum function. The frequency entanglement of the biphoton state is a result of the energy conservation $\omega_1+\omega_2=\omega_{10}+\omega_{20}$. The perfect frequency anticorrelation is valid as long as the linewidths of the pump laser fields are much narrower than the bandwidth of the joint spectrum. The field operators in time domain can be expressed as

$$\hat{a}_i(t) = \frac{1}{\sqrt{2\pi}}\int d\omega \hat{a}_i(\omega) e^{-i\omega t}. \quad (2)$$

The field operators satisfy the commutation relations $[\hat{a}_i(\omega),\hat{a}_j^\dagger(\omega')]=\delta_{ij}\delta(\omega-\omega')$ and $[\hat{a}_i(t),\hat{a}_j^\dagger(t')]=\delta_{ij}\delta(t-t')$. In the state described in Eq. (1), the two photons are perfectly paired.

It can be shown that the biphoton state in Eq. (1) cannot be normalized. This is not surprising because in the CW operation with the time-translation symmetry, a photon pair can be generated at any time with equal probability density for $t\in(-\infty,+\infty)$. This can also be seen from the photon pair

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generation rate R and the individual single-photon rates R_i, which are equal and time invariant:

$$R = R_i = \langle \Psi_{12}|\hat{a}_i^\dagger(t)\hat{a}_i(t)|\Psi_{12}\rangle = \frac{1}{2\pi}\int d\Omega|\Phi(\Omega)|^2. \quad (3)$$

Detection of photon 2 at time t_2 reduces the two-photon state in Eq. (1) to the following state:

$$|\Psi_1\rangle_2 = \frac{1}{\sqrt{R}}\hat{a}_2(t_2)|\Psi_{12}\rangle, \quad (4)$$

which is the heralded single-photon state with the normalization factor $1/\sqrt{R}$. This heralded single-photon state is normalizable (${}_2\langle\Psi_1\|\Psi_1\rangle_2 = 1$) because when photon 2 is detected, photon 1 is "confined" within their relative correlation time duration. To prove this is a pure quantum state, we look at its density operator,

$$\hat{\rho}_{1|2} = |\Psi_1\rangle_{22}\langle\Psi_1|. \quad (5)$$

It is obvious that

$$\hat{\rho}_{1|2}^2 = \hat{\rho}_{1|2}, \quad (6)$$

which shows that the heralded single-photon state in Eq. (4) is indeed a pure quantum state. Therefore, we have proved that in the ideal heralding process described above, where the detection of the trigger photon 2 has an infinitely short response time and there is no time uncertainty in the heralding process, the projected single photon is in a pure state.

Now we turn to the real situation where the trigger-photon detector has a finite response time Δt: one does not know the exact time origin of the heralded single photon during this response time window. In this case, the density operator can be expressed as

$$\begin{aligned}\hat{\bar{\rho}}_{1|2} &= \frac{1}{\Delta t}\int_{-\Delta t/2}^{\Delta t/2} dt_2|\Psi_1\rangle_{22}\langle\Psi_1| \\ &= \frac{1}{R\Delta t}\int_{-\Delta t/2}^{\Delta t/2} dt_2\hat{a}_2(t_2)|\Psi_{12}\rangle\langle\Psi_{12}|\hat{a}_2^\dagger(t_2). \quad (7)\end{aligned}$$

Making use of Eq. (2), the density operator can be rewritten as

$$\begin{aligned}\hat{\bar{\rho}}_{1|2} = &\frac{1}{2\pi R}\int d\omega_2 d\omega_2' \mathrm{sinc}\left[\frac{(\omega_2-\omega_2')\Delta t}{2}\right] \\ &\times \hat{a}_2(\omega_2)|\Psi_{12}\rangle\langle\Psi_{12}|\hat{a}_2^\dagger(\omega_2'). \quad (8)\end{aligned}$$

We then get the density matrix element in the frequency domain,

$$\begin{aligned}\bar{\rho}_{1|2}(\omega_1,\omega_1') &= \langle 0|\hat{a}_1(\omega_1)\hat{\bar{\rho}}_{1|2}\hat{a}_1^\dagger(\omega_1')|0\rangle \\ &= \frac{1}{2\pi R}\mathrm{sinc}\left[\frac{(\omega_1-\omega_1')\Delta t}{2}\right] \\ &\quad\times \Phi(\omega_1-\omega_{10})\Phi^*(\omega_1'-\omega_{10}). \quad (9)\end{aligned}$$

The quantum-state purity of the heralded single photons can be computed from $\gamma = \mathrm{Tr}(\hat{\bar{\rho}}_{1|2}^2)$ [5,29]. With the frequency-entangled biphoton state in Eq. (1), we obtain the purity of its heralded single-photon state,

$$\begin{aligned}\gamma = &\frac{1}{(2\pi R)^2}\int d\Omega d\Omega' \mathrm{sinc}^2\left[\frac{(\Omega-\Omega')\Delta t}{2}\right] \\ &\times|\Phi(\Omega)\Phi(\Omega')|^2. \quad (10)\end{aligned}$$

Obviously, if the trigger-photon detection is instantaneously fast ($\Delta t \to 0$), the density operator is reduced to that of a pure state, $\hat{\bar{\rho}}_{1|2} \to \hat{\rho}_{1|2}(t_2 = 0)$, and we have a unity purity ($\gamma = 1$). This is consistent with our previously described ideal heralding process that projects the remaining photon onto a pure quantum state. If the trigger-photon detector is ultraslow ($\Delta t \to \infty$), $\mathrm{sinc}[\frac{(\omega_1-\omega_1')\Delta t}{2}] \to 2\pi\delta(\omega_1-\omega_1')/\Delta t$, then all nondiagonal density matrix elements in Eq. (9) vanish and the heralded single photon is in a completely mixed state.

For the case with a finite Δt, the quantum-state purity is characterized by Eq. (10). If the trigger-photon detection response time is much shorter than the two-photon coherence time (inverse of the bandwidth of the joint spectrum) such that $(\Omega - \Omega')\Delta t \ll 1$ holds within the bandwidth, we have $\gamma \sim 1$ and the heralded photon is in a nearly pure state.

III. NUMERICAL SIMULATION

To show how the purity of the heralded single-photon state is affected by the finite Δt, we consider two examples in the following.

Case 1: rectangular-shaped spectrum. In the first case, we consider transform-limited biphotons with maximum frequency entanglement within their angular-frequency bandwidth of $2\pi\Delta\nu$. In this case, the joint spectrum function $\Phi(\Omega) = \sqrt{R/\Delta\nu}$ is nonzero only within the bandwidth $\Omega \in [-\pi\Delta\nu,\pi\Delta\nu]$. The solid curve in Fig. 1 shows the numerical result of the purity as a function of the trigger response time. At $\Delta\nu\Delta t \leqslant 0.1$, the purity $\gamma \geqslant 0.99$. The purity decreases as we increase the response time, but not very much. When the response time approaches the two-photon coherence time

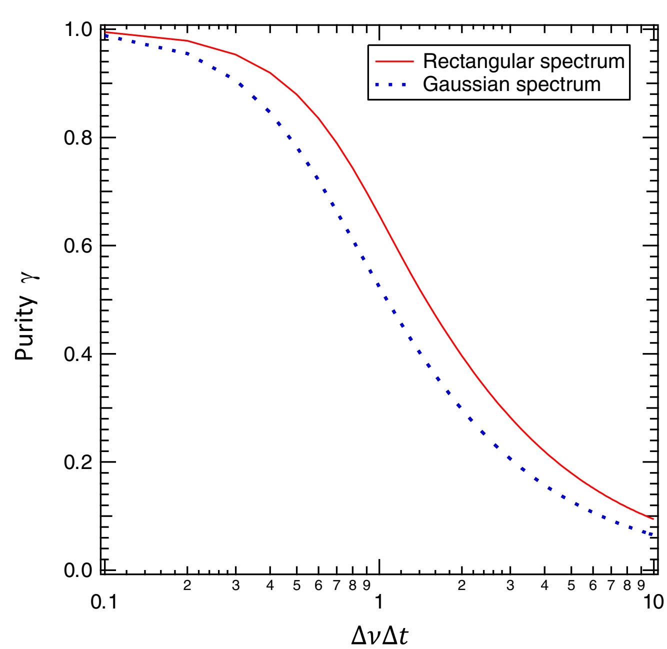

FIG. 1. (Color online) Quantum-state purity γ as a function of $\Delta\nu\Delta t$, i.e., the product of biphoton bandwidth ($\Delta\nu$) and the trigger-photon detection response time (Δt). The solid (red) curve is calculated from the rectangular-shaped spectrum and the dotted (blue) curve is calculated from the Gaussian-shaped spectrum with $\Delta\nu$ as the full bandwidth at half maximum.

($\Delta\nu\Delta t = 1$), the purity is still as high as 0.66. When we further increase the response time to make $\Delta\nu\Delta t = 10$, the purity drops down to 0.09.

Case 2: Gaussian-shaped spectrum. In this case, the joint spectrum function can be expressed as $\Phi(\Omega) = \sqrt{\frac{4R\sqrt{\pi \ln 2}}{2\pi\Delta\nu}} e^{-2\ln 2(\Omega/2\pi\Delta\nu)^2}$, where $2\pi\Delta\nu$ is the full width at half maximum of $|\Phi(\Omega)|^2$. The numerical result of the purity as a function of the trigger response time is plotted as the dotted curve in Fig. 1, which is comparable to that of Case 1 with a rectangular-shaped spectrum. At $\Delta\nu\Delta t = 0.1$, the purity $\gamma = 0.99$. At $\Delta\nu\Delta t = 1$, the purity becomes 0.52. The purity drops down to 0.07 when we further increase the response time to make $\Delta\nu\Delta t = 10$.

Experimentally, the quantum-state purity can be measured as the visibility of the Hong-Ou-Mandel interference [30] with two independent identical heralded single-photon sources [6,31]. In both of the cases discussed above, the quantum-state purity $\gamma > 0.98$ at $\Delta\nu\Delta t = 0.1$, and holds well above 0.90 for $\Delta\nu\Delta t < 0.3$, which is well beyond the classical limit of 0.5. We can treat the state of such a heralded photon ($\Delta\nu\Delta t \leqslant 0.1, \gamma > 0.98$) as a nearly pure single-photon state. For biphotons generated from SPDC or SFWM in solid-state materials without spectral filtering or cavity enhancement, their bandwidths are normally much wider than THz. As a result, it is impossible to herald pure single photons from such frequency-entangled photon pair source using a commercially available single-photon detector with a typical time resolution of about 1 ns. Even the state-of-the-art single-photon detector with an ultrahigh time resolution of about 10 ps is not fast enough. Therefore, disentangling the photon pairs into a factorable joint spectrum by shaping the pump field temporal modes has been considered as the only achievable method to produce heralded pure single-photon states from these wide-band sources without spectral filtering. This situation has been changed with the development of narrow-band biphoton generation from SFWM in cold atoms [4,19–22] and cavity-enhanced SPDC [16–18]. These biphotons, having bandwidths ranging from below 100 MHz down to sub-MHz and coherence time from 10 ns up to more than 1 μs, are ideal for generating heralded pure single-photon states using commercial detectors.

IV. TEMPORAL WAVE FUNCTION OF A HERALDED PHOTON

When the time origin is set by the detection of the trigger photon ($t_2 = 0$), the heralded photon has a well-defined temporal wave function. To illustrate this, we work at $\Delta\nu\Delta t \leqslant 0.1$ and thus it can be treated as a nearly ideal heralding process. From Eq. (4), we obtain the temporal wave function of the heralded single photon:

$$\begin{aligned}\psi_{1|2}(\tau) &= \langle 0|\hat{a}_1(\tau)|\Psi_1\rangle_2|_{t_2=0} \\ &= \frac{1}{2\pi\sqrt{R}}\int d\Omega\Phi(\Omega)e^{-i(\omega_{10}+\Omega)\tau} \\ &= \psi_0(\tau)e^{-i\omega_{10}\tau}, \qquad (11)\end{aligned}$$

where $\psi_0(\tau) = \frac{1}{2\pi\sqrt{R}}\int d\Omega\Phi(\Omega)e^{-i\Omega\tau}$ is the Fourier transform of the two-photon joint spectrum function. Equation (11) also clearly demonstrates that the quantum state of the heralded single photon is a coherent superposition of the frequency components in its spectrum, but not a mixed state. The time origin set by the trigger photon allows for shaping the heralded single photons with an electro-optic modulator [32]. The recent experiments of single photons coherently interacting with atoms [24–26] and cavities [27,28] have indicated that these heralded narrow-band single photons indeed have well-controlled coherent wave packets, but their quantum-state purities have never been theoretically justified.

Following Eqs. (4) and (11), we can produce many interesting heralded single-photon states by engineering the biphoton joint spectrum and frequency entanglement. For example, from a frequency-bin entangled two-photon state $|\omega_{10}+\delta\rangle|\omega_{20}\rangle + e^{i\theta}|\omega_{10}\rangle|\omega_{20}+\delta\rangle$, one can generate a heralded single-photon two-color qubit state $|\omega_{20}\rangle + e^{i\theta}|\omega_{20}+\delta\rangle$. This will certainly find important applications in quantum information processing and quantum communication.

As compared to the heralded photons generated from an frequency-uncorrelated source, heralding single photons from the frequency-entangled (anticorrelated) biphotons has two unique features and advantages. The first is that we can reverse the relative time by switching the trigger photon. In the above discussion, we take photon 2 as the trigger photon and obtain the heralded photon 1 with a temporal wave form $\psi_{1|2}(\tau) = \psi_0(\tau)e^{-i\omega_{10}\tau}$ given in Eq. (11). Now let us switch the trigger detection to photon 1 to heralding the presence of photon 2. We obtain the temporal wave function of the heralded photon 2 as $\psi_{2|1}(\tau) = \psi_0(-\tau)e^{-i\omega_{20}\tau}$, whose amplitude envelope is the time reversal of the heralded photon 1. This time-reversal feature, resulting directly from the frequency anticorrelation, has an important application. For example, when generated from an atomic multilevel system, the biphoton correlation can exhibit exponential decay wave forms because of the nature of spontaneous emission [21]. Depending on which photon is detected as the trigger, we can generate heralded photons with an exponential growth or decay wave form that are particularly useful for loading them into a cavity or interacting with atoms [26,27]. The second interesting feature is the nonlocal spectrum modulation [33–35], which allows for shaping the heralded photon by placing a spectrum modulator $M_2(\omega_2)$ on the trigger-photon path, as illustrated in Fig. 2. It can be derived that the temporal wave function of the heralded photon becomes

$$\psi_{1|2}(\tau) = \frac{1}{2\pi\sqrt{R}}\int d\Omega\Phi(\Omega)M_2(\omega_{20}-\Omega)e^{-i(\omega_{10}+\Omega)\tau}. \quad (12)$$

If photon 1 experiences any distortion caused by the propagation dispersion, it can also be nonlocally corrected by placing

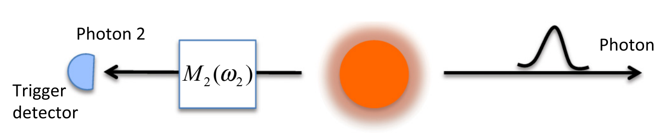

FIG. 2. (Color online) Nonlocal modulation. A spectrum modulator $M_2(\omega_2)$ is placed on the path of the trigger photon 2 to nonlocally shape the heralded photon 1.

a dispersion compensation element on the path of the trigger photon 2.

V. SUMMARY

In summary, we demonstrate that the detection of one photon from a frequency-entangled (frequency-anticorrelated) two-photon state, when the time origin established by the trigger-photon detection has uncertainty much shorter than the two-photon coherence time, projects the remaining photon onto a pure single-photon state. The physics behind such a heralding process can be pictured as the following: the trigger detection erases the frequency information of the trigger photon and, as a result, the heralded photon is in a superposition state of its frequency components. We provide a full theoretical treatment of the quantum-state purity of such heralded photons. We show that the quantum-state purity depends on the response time of the trigger-photon detector. A purity of higher than 0.98 can be achieved when the trigger-detection response time is shorter than one-tenth of the biphoton coherence time (inverse of the bandwidth). The purity becomes significantly degraded when the trigger-detection response time is longer than the biphoton coherence time. We also show that such a heralded narrow-band single photon has a well-defined coherent wave packet. The entanglement raised from the frequency anticorrelation allows for reversing the relative time by switching the trigger photon and shaping the heralded single photon through nonlocal spectral modulation.

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