Detection of hidden entanglement by photon anti-bunching

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Two-photon anti-bunching at a beamsplitter is only possible if the photons are entangled in one specific state, anti-symmetric in the modes. Thus, detection of anti-bunching is a direct indication of entanglement in a variable which might not be directly accessible in an experiment, since the total state of two photons must be symmetric. We demonstrate this explicitly quantitatively in the case of frequency entanglement of two photons with continuous frequency detunings. The principle of detecting hidden entanglement via anti-bunching at a beamsplitter may facilitate the observation of entanglement in other systems, like atomic ensembles or BECs. The analogue in fermionic systems would be to observe bunching.

The study of quantum interference of two particles has been one of the pillars of the rapidly evolving field of quantum information processing. One important milestone was the observation of a dip in the coincidence probability of two indistinguishable photons interfering on a beamsplitter (BS) in the famed Hong Ou Mandel (HOM) experiment [1]. Rather surprising was the discovery of anti-bunching of two photons at a BS [2]. This phenomenon arises when the photons have anti-symmetric polarization states. Therefore, since the total state must be symmetric, the spatial part of the state must also be anti-symmetric, leading to the anti-bunching phenomenon. Both anti-bunching and the HOM dip are now essential tools in experimental quantum information, e.g. for Bell state measurements [2], specifically in dense coding [3] and quantum teleportation [4] or in linear optical quantum computing, where the interaction of photons via the Bosonic commutation relation is used to realize effective photon-photon nonlinearities [5,6].

Ou and Mandel showed that interference can also be observed for photons with non-degenerate center frequencies; sinusoidal oscillations will arise in the coincidence dip of broadband photons generated in spontaneous parametric downconversion (SPDC) when detuned interference filters are placed behind the BS [7]. The interference of frequency non-degenerate photons was later treated in a number of theoretical [8] and experimental investigations [3, 10, 11, 12], which found similar results. However, in all these experiments, two essential facts were not emphasized; first, the fact that photon anti-bunching, i.e. the observation of a coincidence probability at the output of a BS above the random level, is a direct consequence of an antisymmetric part of the spectral amplitude of a two-particle state. This connection was made in a comprehensive theoretical paper by K. Wang [12]. The second is that this anti-bunching can be directly attributed to entanglement present in the system.

In this paper we experimentally study two-photon interference of continuously frequency-detuned photon pairs on a beamsplitter. In contrast to previous experiments [3, 9, 10, 11], our scheme does not require frequency selection by filtering or mapping of frequency-to discrete path-entanglement. Instead, the observed interference patterns originate from the whole frequency-correlated SPDC two-photon spectra. In addition to measuring HOM interference by introducing variable time delays between photons, we exploit the tunability of our photon source to demonstrate two-particle interference at fixed time delays but with continuous relative photon center-frequency detunings that are far larger than the narrow single-mode spectral bandwidths. This adds a new dimension to the phenomenon of two-particle interference which has not been studied so far. We show that the observed anti-bunching is a consequence of the antisymmetric spectral amplitudes of the two-particle states. The fact that the complete two-photon state has to follow Bose-Einstein statistics implies that anti-bunching is a direct evidence of frequency entanglement.

Experiment – Photon pairs with orthogonal polarizations are generated via type-II SPDC in a periodically poled KTiOPO$_4$ (PPKTP) crystal at the degenerate wavelength of 810 nm (Fig. 1). The temperature $T$ of the PPKTP is controlled by a thermo-electric element, which is stable to about 0.1˚C. As shown in [13], due to the periodic poling, the photon wavelength can be tuned over a wide range around degeneracy by a change in $T$. The photons of a pair are separated at a polarizing beamsplitter (PBS) and coupled into single-mode fibers before they interfere on a 50/50 fiber beam splitter. The events detected by avalanche photo diodes are analyzed in a coincidence circuit within a time window of 4.4 ns. Note that there are no bandpass filters in the setup, just a pair of long-pass color filters is needed to block the pump light. We first measured the interference of two photons with center frequencies $\omega_0^a$ and $\omega_0^b$ by scanning the relative optical delay $\tau$ between them. The coinci-
dence probabilities $p_c$, which are obtained by normalizing the coincidences (no background subtraction) to the rates observed outside the photon coherence lengths, are shown in Fig. 2a. At zero detuning $\omega_a^0 - \omega_b^0 = 0$, the interference pattern exhibits a distinct triangular dip which is characteristic for photons produced in Type-II SPDC [14, 15]. For increasing frequency detunings, oscillations arise in the triangular interference pattern. Eventually, as the detuning of the frequencies changes, the coincidence probability $p_c$ shows distinct harmonic features of up to $p_c^{\text{max}} = 0.593$ beyond the average value $p_c = 0.5$. In an additional measurement, we studied the two-photon interference in the frequency domain at zero time delay $\tau = 0$. First, the PPKTP was heated to $T = 90^\circ\text{C}$. The heating current was then switched off and we recorded coincidences while the crystal cooled down to 28 °C. This heating current was then switched off and we recorded coincidences (no background subtraction) to the rates observed outside the photon coherence lengths, are shown in Fig. 2a. At zero detuning $\omega_a^0 - \omega_b^0 = 0$, the interference pattern exhibits a distinct triangular dip which is characteristic for photons produced in Type-II SPDC [14, 15]. For increasing frequency detunings, oscillations arise in the triangular interference pattern. Eventually, as the detuning of the frequencies changes, the coincidence probability $p_c$ shows distinct harmonic features of up to $p_c^{\text{max}} = 0.593$ beyond the average value $p_c = 0.5$. In an additional measurement, we studied the two-photon interference in the frequency domain at zero time delay $\tau = 0$. First, the PPKTP was heated to $T = 90^\circ\text{C}$. The heating current was then switched off and we recorded coincidences while the crystal cooled down to 28 °C. This temperature range is equivalent to a frequency detuning of $\Delta \omega = 1.58 \text{THz}$, which corresponds to a relative detuning of $[-16.1, 26.7]$. The distinct interference pattern in Fig. 2b (crystal cooling curve in the inset) shows that $p_c$ periodically exceeds the random probability $p_c = 0.5$. Obviously, in both measurements, frequency-detuned photons partially anti-bunch at the BS.

Theory – We now consider the output of the photon-pair source in figure 1 theoretically. By first-order perturbation theory [14], we obtain the two-photon state produced by SPDC at collinear, type-II phase matching, where the fields $\omega_a$ and $\omega_b$ have orthogonal polarization:

$$
|\psi(\omega_a, \omega_b)\rangle = \int d\omega_a d\omega_b \delta(\omega_p - \omega_a - \omega_b) \times \text{sinc} \left( \frac{L \Delta k(T)}{2} \right) a_{a,H}^\dagger(\omega_a) a_{b,V}^\dagger(\omega_b) |0\rangle.
$$

(1)

Here, $\Delta k(T) = k_p(\omega_p, T) - k_a(\omega_a, T) - k_b(\omega_b, T) - \frac{2\pi}{\Lambda(T)}$ is the phase mismatch, a function of optical and thermal properties of the crystal. The energy uncertainty due to the finite interaction time in the crystal is negligible in regard to the phase matching, and is therefore represented by a $\delta$-function. The spectral amplitude $\text{sinc}(L \Delta k(T)/2)$ emanates from integration of the interacting fields over the finite crystal length $L$. We expand $\Delta k$ into a Taylor series around $(\omega_a^0, \omega_b^0)$, where $\omega_a^0$ and
\(\omega_0\) satisfy both energy conservation and phasematching conditions, \(\Delta k = -(\omega_\alpha - \omega_0(T))k'_0 - (\omega_\beta - \omega_0(T))k'_1\). We can then parameterize the state \(|\psi(\omega_\alpha, \omega_\beta)\rangle\) of Eq. 4 in terms of frequency differences \(\nu = \omega_\alpha - \omega_\beta\) and \(\mu = \omega_0(T) - \omega_0^0(T)\). The spectral amplitude term of \(|\psi(\mu, \nu)\rangle\) then reads:

\[
sinc\left(\frac{L\Delta k(T)}{2}\right) \rightarrow sinc\left(\frac{\nu - \mu(T)}{\zeta}\right)
\]

(2)

where \(\zeta = 4/(L(k'_0 - k'_1))\) is directly connected to the spectral single-photon bandwidth \(\Delta \omega\) via \(\zeta = 2\Delta \omega/\pi\).

The two created photons of Eq. 1 are combined on a 50/50 beamsplitter with a relative optical delay \(\tau\). The BS transforms modes \(a\) and \(b\) into the output modes \(a_1^\dagger(\omega) = e^{i\omega_{\tau}}/\sqrt{2}(a_0^\dagger(\omega) - ie^{-i\omega\tau}a_1^\dagger(\omega))\) and \(a_2^\dagger(\omega) = e^{i\omega_{\tau}}/\sqrt{2}(ie^{-i\omega\tau}a_0^\dagger(\omega) + a_1^\dagger(\omega))\). At the BS, the photons have identical polarization, so we can neglect the polarization part of the modes. By parametrization of the integral and some calculation (details in Appendix A), we obtain the coincidence detection probability \(p_c\) at the output of the BS:

\[
p_c(\tau, \mu) = \begin{cases} 
\frac{1}{2} & \text{for } |\tau| < \frac{\zeta}{2} \\
\frac{1}{2} & \text{otherwise} 
\end{cases}
\]

(3)

The two-dimensional interference pattern from Eq. 3 is depicted in a density plot in Fig. 3 as a function of the output of the BS:

\[
\text{FIG. 3: Theoretical coincidence probability as a function of optical delay } \tau \text{ and temperature } T \text{ (relative frequency detuning } \mu/\Delta \omega). \text{ We experimentally measured the coincidence probability by scanning } \tau \text{ for different frequency detunings (lines 1-5) and, with variable detuning, for a fixed optical delay } \tau = 0 \text{ (line a).}
\]

For the difference frequency dependence at zero delay \(\tau = 0\) (dotted line a in Fig. 3), we find:

\[
p_c(\mu) = \frac{1}{2} \left(1 - \frac{\mu}{\zeta}\right),
\]

(5)

For detuned frequencies, the coincidence probability exhibits the damped sinusoidal oscillations which can, in principle, be observed even for arbitrarily large detunings. The highest anti-bunching appears at \(p_c(0, \frac{2\mu}{\zeta}) = 0.609\), where the amount of anti-symmetry in the spectral amplitude (2) reaches a maximum.

Because the two-photon state in our experiment is not entangled in polarization, this anti-bunching is a direct evidence for entanglement in another degree of freedom - the frequency. To see that, remember that the bosonic nature of photons require the overall wave function of a photonic quantum state to be totally symmetric, i.e. the sign of the wavefunction has to remain unchanged under a parity operation. In consequence, two entangled photons, specifically those entangled in a singlet state, show perfect anti-bunching when they interfere at a beamsplitter [2]. This is because the polarization part of the two-photon wavefunction of the singlet state is totally anti-symmetric and in order for the total wavefunction to be symmetric, its spatial part has to be anti-symmetric too. Since this is only possible for photons in different spatial modes and since the beamsplitter (that acts on these modes) does not change the symmetry of the spatial part of the wavefunction, the particles can only emerge in two different output ports, i.e. they anti-bunch. More generally, anti-bunching is a sufficient (but not necessary) criterion for the presence of entanglement in at least one degree of freedom other than the spatial one. Since anti-bunching requires anti-symmetry in the
spatial part of the wavefunction, at least one other degree of freedom needs to contribute an anti-symmetric part to maintain the overall required symmetry. While entangled states can be both symmetric and anti-symmetric, no anti-symmetric state can be separable. It is straightforward to see that separable states can therefore never exhibit anti-bunching (see Appendix B), and that spatial anti-bunching can only occur if one other degree of freedom, in our case the frequency, is entangled. It is interesting to note that while the observation of anti-bunching is a signature of entanglement, the observation of bunching, i.e. the HOM-dip, is no signature of entanglement, as it might also occur for product states. We further remark that a similar phenomenon should arise for the case of two fermions incident on a beamsplitter, as for example in electron interferometry.\textsuperscript{16, 17} There, the total state must be anti-symmetric and bunching is a clear signature of entanglement.

**Conclusion** – We measured two-particle interference of photon pairs in time and, for the first time, with continuous frequency differences. We observed oscillating interference patterns even when the difference frequency of the photons is much larger than the single-photon bandwidth. This is of high interest for quantum information experiments that rely on single photons emitted by quantum dots, color centers in solids or atoms in cavities. Due to material properties, these photons will not have identical center frequencies.\textsuperscript{18} We showed that the perfect bunching of a symmetric polarization product state is destroyed by an increasing anti-symmetric contribution induced by frequency detuning and that the observation of anti-bunching at a beamsplitter is compelling evidence for the presence of frequency entanglement. The fact that entanglement of particles can be directly shown by partial anti-symmetrization of otherwise hidden degrees of freedom and subsequent anti-bunching on a BS may facilitate the observation of entanglement in systems such as electrons or BECs.\textsuperscript{19}

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**Appendix A** – We start with the SPDC state in Eq. that is transformed at the BS. Parametrization of the integral and projection onto output modes 1, 2 yields the probability amplitude $A(t; \tau)$ for a coincidence in 1 and 2:

$$A(t; \tau) = \frac{1}{2} \int d\nu \text{sinc} \left( \frac{\nu - \mu}{\zeta} \right) \left( e^{-i\nu t} - e^{i\nu(t+\tau)} \right),$$

(6)

where $t = (t_1 - t_2)/2$. This integral can be solved via a Fourier transform, and gives:

$$A(t; \tau) = \frac{1}{2} \left( e^{i\mu \frac{\tau}{2}} \left( \frac{\zeta}{2} \right) - e^{-i\mu(t+\tau)} \Pi \left( \frac{(t + \tau) \zeta}{2} \right) \right),$$

(7)

where $\Pi(x)$ is the rectangular function $\Pi(x) = 1$ if $|x| \leq 1/2$, and $\Pi(x) = 0$ elsewhere. Thus, the coincidence detection probability in the output modes is:

$$p_c(\tau) = A_0 \int dt |A(t; \tau)|^2 = \frac{A_0}{2} \left( \int dt \frac{\zeta}{2} \right) + \left( -Re \int dt e^{i\nu(2t+\tau)} \Pi \left( \frac{\zeta}{2} \right) \Pi \left( \frac{(t + \tau) \zeta}{2} \right) \right).$$

(8)

By evaluating the first integral of this expression, we find the normalization constant $A_0 = \frac{1}{2}$. Evaluation of Eq. 8 then leads to $p_c(\tau)$ in Eq. 8.

**Appendix B** – We now consider a separable state of two photon wave packets with identical polarization, produced by independent sources. A generalized, separable state is:

$$|\psi[f, g]\rangle = \int d\omega_1 d\omega_2 f(\omega_1)g(\omega_2) a^{\dagger}(\omega_1) b^{\dagger}(\omega_2)|0\rangle,$$

(9)

where $f(\omega_1)$ and $g(\omega_2)$ are properly normalized spectral amplitudes. Some calculation leads to:

$$p_c = \frac{1}{2} - \frac{1}{2} \left| \tilde{g} * \tilde{f}(\tau) \right|^2.$$

(10)

It is obvious that $p_c$ for separable states is always less than 1/2. For mixed states, one observes incoherent contributions in the form, from which no interference effects arise. Thus, also in the case of a mixed state, we expect $p_c < 1/2$.

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