Role of indistinguishability and entanglement in Hong-Ou-Mandel interference and finite-bandwidth effects of frequency-entangled photons

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(Received 22 February 2022; accepted 10 February 2023; published 9 March 2023)

We investigate the relation between indistinguishability and quantum entanglement in Hong-Ou-Mandel interference experiments theoretically and relate these quantum mechanical principles to the theorem of entanglement monogamy. Employing Glauber's theory of quantum coherence we compute the detection statistics in HOM interference of frequency-entangled photons and find an additional term in the coincidence detection probability, which is related to the spectral indistinguishability of the considered photons that arises from finite-bandwidth effects and therefore is relevant in the limit of low-frequency separations or large single-photon bandwidths. Compared to previous work in that context we treat all photonic degrees of freedom on equal footing.

DOI: 10.1103/PhysRevA.107.032205

I. INTRODUCTION

Indistinguishability of physical systems is one of the important defining features of quantum systems without classical analogs. For example, the exchange interaction in metals, which arises as a consequence of the indistinguishability between the electrons in the conduction band, can change the magnetic properties of the system completely [1]. In the case of bosons in particular, indistinguishability leads to intriguing phenomena, such as Bose-Einstein condensation or *photon bunching* within interference experiments. The latter was witnessed in the pioneering Hong-Ou-Mandel (HOM) experiment [2], and today it is one of the celebrated results of quantum optics that the phenomenon of photon bunching can be used to measure time intervals in a sub-picosecond regime. This accuracy can even be enhanced in the presence of spectral entanglement [3], giving rise to the yet more counterintuitive and interesting phenomenon of *photon antibunching*, which is very uncharacteristic for photons as these naturally follow bosonic statistics. Needless to say, quantum entanglement is one of the most important physical phenomena which cannot be explained by means of a classical theory, and moreover, its existence has wide consequences in our interpretation of the nonlocal behavior of matter and the probabalistic character of nature. Since neither photon bunching nor photon antibunching can be explained without considering the particle character of photons, and since their occurrence is ultimately related to nonclassical features such as indistinguishability and entanglement, which are heralded in a particular way in HOM interference, these experiments today constitute one of the most important methods to test and study quantum mechanical principles. It is, therefore, of fundamental interest to precisely understand the role of indistinguishability and entanglement on the detection statistics of HOM experiments, and the reader is referred to Ref. [4] and references therein for an extensive overview on recent progress and advances in the field and to Refs. [5,6] for a modern mathematical formulation of the phenomenon in the broader context of multiparticle interference.

In addition, the opportunity to provide highly accurate time measurements with HOM interference adds to the practical appeal of these experiments. Only recently it was demonstrated that HOM interference can be used to enhance the accuracy of clock synchronization [7,8], a feature that was already predicted theoretically [9]. Therefore, HOM-like schemes are also candidates for space-based implementations, such as the Global Positioning System (GPS), the installation of a global time standard, or, in a broader context, high-precision metrology in general.

Recently, the possibility to resolve time intervals below $(100\,\mathrm{THz})^{-1}$ (i.e., in the inverse optical regime) with HOM interference of frequency-entangled photons was recognized to bare the potential of conducting fundamental tests of physics by addressing the interplay between gravity and quantum mechanics [10–12].

In the present paper we employ Glauber's theory of optical coherence [13] and extend previous work on multiphoton quantum interference [14] to incorporate all photonic degrees of freedom on equal footing. As a result, we obtain a convenient formalism, which on the one hand enables us to compute the detection statistics of multiphoton interference experiments from a general wave function and on the other hand heralds the role of quantum mechanical principles such as indistinguishability and quantum entanglement in a particular way.

We apply our formalism to compute the detection statistics of HOM interference with frequency-entangled photons [15], and we find an additional term in the corresponding interference pattern of coincidence detection compared to established results [3,15,16] and provide a physical interpretation for this term.

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By contrasting the interference behavior of spectrally indistinguishable frequency-entangled photons with the one of spectrally distinguishable frequency-detuned photons, we draw conclusions on the relation between entanglement and indistinguishability in HOM interference and relate these concepts via the theorem of *entanglement monogamy*.

This work is organized as follows. In Sec. II we revise the basics of HOM interference, and we present our formalism and apply it to frequency-entangled photons. In Sec. III we relate the previously obtained results to former work and provide physical interpretation. Sections V and IV are dedicated to concluding our work and providing an outlook.

II. HONG-OU-MANDEL INTERFERENCE

Sections II A and II B of this paper are dedicated to developing the theoretical formalism to characterize HOM interference based on Glauber's theory of coherence. In Sec. II C we show one of the main results of the present paper that is the HOM interference pattern of frequency-entangled photons, where we find an additional term related to the finite bandwidth of the employed photons, which was not mentioned in the literature [3,15,16] before.

A. Two-photon states

The theory for describing N-photon states has been extensively developed [17], and we leave details to the interested reader. A general two-photon state in the context of HOM interference is given by [14]

$$|\psi(\tau_1,\tau_2)\rangle = \mathcal{N}_\psi \int d\boldsymbol{\omega}_1 d\boldsymbol{\omega}_2\, \Phi(\boldsymbol{\omega}_1,\boldsymbol{\omega}_2) e^{i\omega_1\tau_1} e^{i\omega_2\tau_2} \hat{a}^\dagger_{\boldsymbol{\omega}_1} \hat{a}^\dagger_{\boldsymbol{\omega}_2} |0\rangle\,, \tag{1}$$

where the bold $\boldsymbol{\omega}$ denotes the set of parameters that characterize all degrees of freedom (DOFs) of a single photon, e.g., its frequency, orbital momentum, polarization, spatial mode, and so forth. In the case that a discrete DOF is considered (for instance, the polarization DOF) the integral in Eq. (1) has to be evaluated as a sum. We call $\Phi(\boldsymbol{\omega}_1,\boldsymbol{\omega}_2)$ the two-photon wave function. The slim ω denotes the photon frequency, which we treat separately from the other photonic DOFs $\boldsymbol{\sigma}$. Thus, we can write all photonic DOFs as $\boldsymbol{\omega} = \{\omega, \boldsymbol{\sigma}\}$. Depending on the context we alternatively denote the photonic wave function as $\Phi(\boldsymbol{\omega}_1,\boldsymbol{\omega}_2) = \Phi(\omega_1,\boldsymbol{\sigma}_1,\omega_2,\boldsymbol{\sigma}_2) = \Phi_{\boldsymbol{\sigma}_1\boldsymbol{\sigma}_2}(\omega_1,\omega_2)$.

The operators $\hat{a}_{\boldsymbol{\omega}}$ and $\hat{a}^\dagger_{\boldsymbol{\omega}}$ are bosonic annihilation and creation operators, respectively, which satisfy the canonical commutator relations $[\hat{a}_{\boldsymbol{\omega}}, \hat{a}^\dagger_{\boldsymbol{\omega}'}] = \delta_{\boldsymbol{\omega}\boldsymbol{\omega}'}$, while all other commutators vanish. Here $\delta_{\boldsymbol{\omega}\boldsymbol{\omega}'}$ is the multidimensional delta function of the considered photonic DOFs. For each continuous DOF (for instance, the frequency) $\delta_{\boldsymbol{\omega}\boldsymbol{\omega}'}$ contains a Dirac δ distribution and for each discrete DOF it contains a Kronecker symbol as a factor. For instance, if we consider only the photon's polarization $P = H$ and V [horizontal (H) and vertical (V) polarization] and the photon's frequency ω (i.e., $\boldsymbol{\omega} = \{\omega, P\}$), the delta function reads $\delta_{\boldsymbol{\omega}\boldsymbol{\omega}'} = \delta(\omega-\omega')\delta_{PP'}$.

Expression (1) depends on two times, τ_1 and τ_2, that describe optical delays that can be applied independently to wave packets in HOM interferometry [14] (see Fig. 1). From an experimental point of view, these delays are typically

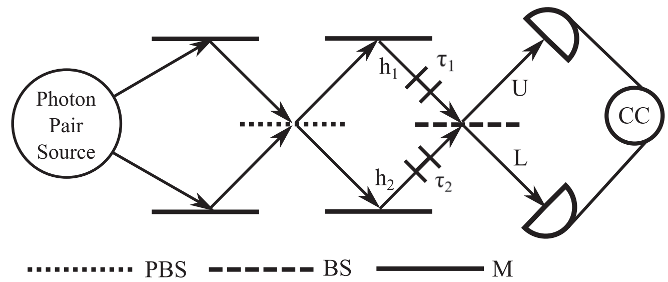

FIG. 1. Hong-Ou-Mandel (HOM) experiment with frequency-entangled photons. PBS, polarizing beam splitter; BS, beam splitter; M, mirror; CC, coincidence count logic.

realized via a variation of the optical path of the respective photons. In practice, this can be achieved, for instance, by a variation of the refractive index of the respective transmission path. The normalization constant $\mathcal{N}_\psi$ has to be chosen in order to fulfill $\langle\psi|\psi\rangle = 1$. However, the procedure that is described in the following to compute the detection statistics of HOM interference is independent of the normalization constant as we show further below. For this reason, we omit it for the rest of this work.

B. Photon detection

Photon detection is described within Glauber's quantum theory of coherence [13]. The electric field operators are defined as

$$\hat{E}^{(+)}_{\boldsymbol{\sigma}}(t) = i\int d\omega\, \mathcal{E}_\omega e^{-i\omega t} \hat{a}_{\boldsymbol{\omega}}, \tag{2a}$$

$$\hat{E}^{(-)}_{\boldsymbol{\sigma}}(t) = [\hat{E}^{(+)}_{\boldsymbol{\sigma}}(t)]^\dagger, \tag{2b}$$

where $\mathcal{E}_\omega = \sqrt{[\hbar\omega/(4\pi\epsilon_0 c)]}$ is the frequency-dependent electric field per photon. The electric field operators are characterized by the parameter set $\boldsymbol{\sigma}$.

Analogously we may define creation (annihilation) operators, which create (annihilate) a photon at time t, which are characterized by a parameter set $\boldsymbol{\sigma}$ as

$$\hat{a}_{\boldsymbol{\sigma}}(t) = \int d\omega e^{-i\omega t} \hat{a}_{\boldsymbol{\omega}}, \tag{3a}$$

$$\hat{a}^\dagger_{\boldsymbol{\sigma}}(t) = \int d\omega e^{+i\omega t} \hat{a}^\dagger_{\boldsymbol{\omega}}. \tag{3b}$$

The temporal creation (annihilation) operators (3) are basically Fourier transforms with respect to the frequency DOF of the spectral creation (annihilation) operators $\hat{a}^\dagger_{\omega\boldsymbol{\sigma}}$ ($\hat{a}_{\omega\boldsymbol{\sigma}}$). Note that for the remaining DOFs, $\boldsymbol{\sigma}$ are not integrated out in Eq. (3).

In Glauber's theory of optical coherence [13], the expectation value of a joint detection of two electric field quanta characterized by $\boldsymbol{\sigma}_1$ and $\boldsymbol{\sigma}_2$ at times t_1 and t_2 in the quantum state (1) reads

$$\Gamma_{\boldsymbol{\sigma}_1\boldsymbol{\sigma}_2}(t_1,t_2) = \langle\psi|\hat{E}^{(-)}_{\boldsymbol{\sigma}_1}(t_1)\hat{E}^{(-)}_{\boldsymbol{\sigma}_2}(t_2)\hat{E}^{(+)}_{\boldsymbol{\sigma}_2}(t_2)\hat{E}^{(+)}_{\boldsymbol{\sigma}_1}(t_1)|\psi\rangle\,, \tag{4}$$

and the probability $p_{\boldsymbol{\sigma}_1\boldsymbol{\sigma}_2}(t_1,t_2,\tau_1,\tau_2)$ of a joint detection of two photons characterized by $\boldsymbol{\sigma}_1$ and $\boldsymbol{\sigma}_2$ at times t_1 and t_2 is proportional to $\langle\psi(\tau_1,\tau_2)|\hat{a}^\dagger_{\boldsymbol{\sigma}_1}(t_1)\hat{a}^\dagger_{\boldsymbol{\sigma}_2}(t_2)\hat{a}_{\boldsymbol{\sigma}_2}(t_2)\hat{a}_{\boldsymbol{\sigma}_1}(t_1)|\psi(\tau_1,\tau_2)\rangle \equiv \langle\varphi|\varphi\rangle$, where $|\varphi\rangle := \hat{a}_{\sigma_2}(t_2)\hat{a}_{\sigma_1}(t_1)\,|\psi(\tau_1,\tau_2)\rangle$. Using Eqs. (1) and (3), together with some algebra, we find that the detection probability $p_{\sigma_1\sigma_2}(t_1,t_2,\tau_1,\tau_2) \propto \langle\varphi|\varphi\rangle$ reads

$$p_{\sigma_1\sigma_2}(t_1,t_2,\tau_1,\tau_2) \propto |\tilde{\Phi}_{\sigma_1\sigma_2}(t_1-\tau_1,t_2-\tau_2) + \tilde{\Phi}_{\sigma_2\sigma_1}(t_2-\tau_1,t_1-\tau_2)|^2, \quad (5)$$

where we have introduced the Fourier transform of the photonic wave function

$$\tilde{\Phi}_{\sigma_1\sigma_2}(T_1,T_2) = \int d\omega_1 d\omega_2\, \Phi_{\sigma_1\sigma_2}(\omega_1,\omega_2)e^{-i\omega_1T_1}e^{-i\omega_2T_2}. \quad (6)$$

The probability $P_{\sigma_1\sigma_2}(\tau_1,\tau_2)$ to detect two photons irrespectively of the instance of time, when they are detected, is proportional to

$$\tilde{P}_{\sigma_1\sigma_2}(\tau_1,\tau_2) := \int dt_1dt_2\, p_{\sigma_1\sigma_2}(t_1,t_2,\tau_1,\tau_2). \quad (7)$$

We employ the convolution theorem from Fourier analysis and obtain

$$\begin{aligned}\tilde{P}_{\sigma_1\sigma_2}(\tau_1,\tau_2) = &\int d\omega_1 d\omega_2\, [|\Phi_{\sigma_1\sigma_2}(\omega_1,\omega_2)|^2 \\ &+ |\Phi_{\sigma_2\sigma_1}(\omega_2,\omega_1)|^2] \\ &+ 2\mathrm{Re}\Bigg\{\int d\omega_1 d\omega_2 \Phi_{\sigma_1\sigma_2}(\omega_1,\omega_2) \\ &\times \Phi^*_{\sigma_2\sigma_1}(\omega_2,\omega_1)e^{-i(\omega_1-\omega_2)(\tau_1-\tau_2)}\Bigg\}, \end{aligned} \quad (8)$$

which can be further simplified to the compact form

$$\tilde{P}_{\sigma_1\sigma_2}(\tau_1,\tau_2) = \int d\omega_1 d\omega_2 \left|\mathcal{S}\left[\Phi^{(\tau_1,\tau_2)}_{\sigma_1\sigma_2}(\omega_1,\omega_2)\right]\right|^2, \quad (9)$$

where $\Phi^{(\tau_1,\tau_2)}_{\sigma_1\sigma_2}(\omega_1,\omega_2) = \Phi_{\sigma_1\sigma_2}(\omega_1,\omega_2)e^{i\omega_1\tau_1}e^{i\omega_2\tau_2}$ is the time-dependent photonic wave function, which depends on the optical delays $\tau_{1,2}$, and $\mathcal{S}$ is the symmetrization operator, which acts as $\mathcal{S}[\Phi^{(\tau_1,\tau_2)}_{\sigma_1\sigma_2}(\omega_1,\omega_2)] = \Phi^{(\tau_1,\tau_2)}_{\sigma_1\sigma_2}(\omega_1,\omega_2) + \Phi^{(\tau_1,\tau_2)}_{\sigma_2\sigma_1}(\omega_2,\omega_1)$. Finally, to obtain the normalized probabilities to detect one photon characterized by $\boldsymbol{\sigma}_1$ and another characterized by $\boldsymbol{\sigma}_2$, we have to compute

$$P_{\sigma_1\sigma_2}(\tau_1,\tau_2) = \frac{\tilde{P}_{\sigma_1\sigma_2}(\tau_1,\tau_2)}{\int d\boldsymbol{\sigma}_1 d\boldsymbol{\sigma}_2\, \tilde{P}_{\sigma_1\sigma_2}(\tau_1,\tau_2)}. \quad (10)$$

From the last expression it is seen that the normalization constant $\mathcal{N}_\psi$ from Eq. (1) cancels out in the calculation because its square appears in the nominator and the denominator of Eq. (10).

Note the symmetries of the probabilities $P_{\sigma_1\sigma_2}(\tau_1,\tau_2) = P_{\sigma_2\sigma_1}(\tau_1,\tau_2) = P_{\sigma_1\sigma_2}(\tau_2,\tau_1)$ and that the probabilities are always a function of the difference $\Delta\tau := \tau_1 - \tau_2$ as can be seen from Eq. (8).

Equation (9) is a central result of the paper as we can use it to easily compute the joint detection statistics of HOM interference for any given two-photon wave function $\Phi_{\sigma_1\sigma_2}(\omega_1,\omega_2)$, and it moreover highlights the interpretation of the absolute square value of the (time-dependent) photonic wave function as being the probability for certain detection events. Moreover, Eq. (9) clearly shows that only the symmetrized part of the photonic wave function is of physical relevance, which can be already inferred from Eq. (1), since it can be written solely in terms of the symmetrized time-dependent photonic wave function by use of the canonical commutator relations.

Apart from that, Eq. (8) emphasizes the role of indistinguishability between the two involved photons in HOM interference. The first two terms of Eq. (8) do not depend on the delays, in contrast to the last third term, the interference term. This term can be interpreted as the overlap of the joint photonic wave function at the interfering beam splitter (BS) with itself under an exchange of the function arguments, i.e., $\boldsymbol{\omega}_1 \leftrightarrow \boldsymbol{\omega}_2$, i.e., under particle exchange, and thus can serve as a measure of indistinguishability. If this overlap vanishes, the particles are considered as entirely distinguishable and HOM interference does not occur, and we show this in some concrete examples in the later sections.

Moreover, in the absence of entanglement, which is distinguished by the factorization of the two-photon wave function into two single-photon wave functions [18] [i.e., $\Phi_{\sigma_1\sigma_2}(\omega_1,\omega_2) = \Phi_{\sigma_1}(\omega_1)\Phi_{\sigma_2}(\omega_2)$], the interference term of Eq. (8) results in a positive value for $\boldsymbol{\sigma}_1 = \boldsymbol{\sigma}_2$. This increased detection probability for equally behaved ($\boldsymbol{\sigma}_1 = \boldsymbol{\sigma}_2$) photons implies that the factorization of the joint photonic wave function leads to the exclusive occurrence of photon bunching and thus certifies the occurrence of photon antibunching as a sufficient validation for the presence of entanglement, which was already recognized before in Ref. [19] and was also used in Ref. [20].

Furthermore, we want to add that using the same techniques as in the derivation of Eq. (9) we obtain for the single-particle detection statistics which is characterized by the probability $P_{\sigma}(\tau_1,\tau_2) = \int dt\, \langle\psi(\tau_1,\tau_2)|\hat{a}^\dagger_{\sigma}(t)\hat{a}_{\sigma}(t)|\psi(\tau_1,\tau_2)\rangle$ to detect a single photon in state $\boldsymbol{\sigma}$ the intuitive result

$$P_{\sigma} = \sum_{\bar{\sigma}} \frac{1}{2}(P_{\sigma\bar{\sigma}} + P_{\bar{\sigma}\sigma}) = \sum_{\bar{\sigma}} P_{\sigma\bar{\sigma}} \quad (11)$$

for all τ_1 and τ_2, which we omitted as function arguments in Eq. (11).

Finally, we want to remark that the presented formalism here naturally extends to multiphoton interference experiments, where more than two photons are involved (see, for instance, Ref. [21]). In these cases an analog result to Eq. (9) is obtained for the multiphoton detection statistics where one has to symmetrize over the various DOFs of the involved photons. This also includes the special case of single-photon interference experiments like the Mach-Zehnder interferometer. Furthermore, also extending our formalism to mixed states is straightforward. However, we leave the concrete documentation of the extension of our formalism to mixed states for future work.

C. Frequency-entangled photons

The generation of frequency-entangled photons and their subsequent measurement within a HOM interference experiment is shown in Fig. 1; this was first demonstrated in Ref. [3] and was recently analyzed in more detail in Ref. [15]. For this, one first generates two polarization-entangled and frequency-detuned photons which originate from a spontaneously down-converting periodically poled $KTiOPO_4$ crystal (ppKTP crystal). These photons render (up to normalization) the state

$$|\psi_{\text{f.d.}}\rangle = \int d\omega_1 d\omega_2\, \phi_{\text{f.d.}}(\omega_1,\omega_2) e^{i\omega_1\tau_1} e^{i\omega_2\tau_2} \times \left(\hat{a}^\dagger_{Hh_1\omega_1}\hat{a}^\dagger_{Hh_2\omega_2} + \hat{a}^\dagger_{Vh_1\omega_1}\hat{a}^\dagger_{Vh_2\omega_2}\right)|0\rangle\,, \tag{12}$$

where the spectral wave function in this case reads

$$\phi_{\text{f.d.}}(\omega_1,\omega_2) = \delta(\omega_p-\omega_1-\omega_2)\,\text{sinc}\left(\frac{\omega_1-\omega_2-\mu}{\xi}\right). \tag{13}$$

The subscript f.d. means frequency detuned, ω_p is the pump frequency of the down-converting process, μ is the frequency separation or detuning of the photons (which can be adjusted by tuning the ppKTP crystal's temperature), and ξ is the single-photon bandwidth.

The spectral wave function (13) is not square integrable due to the occurrence of the delta distribution $\delta(\omega_p-\omega_1-\omega_2)$. This constitutes a problem in the evaluation of Eq. (8) [or rather Eq. (9)] due to the occurrence of squared delta functions $\delta^2(\omega_p-\omega_1-\omega_2)$ under the integrals to evaluate. One approach to treat this problem stems from scattering theory. There, one uses the identity $\delta(\omega)=\lim_{T\to\infty}\int_{-T}^{T}dt\,\exp(i\omega t)$ to rewrite squares of delta distributions as $\delta^2(\omega)=T\delta(\omega)$, where T is the time over which the various detection events in an experiment are integrated. To obtain particle fluxes instead of particle numbers one has to divide expectation values by T. Thus, particle fluxes remain finite, also in the limit $T\to\infty$. The probability of a certain detection event is then recovered through division of the corresponding particle flux of the respective detection event by the sum of particle fluxes of all possible detection events.

After generation, the two polarization-entangled frequency-detuned photons interfere on a polarizing beam splitter (PBS), a process that transfers polarization entanglement onto the frequency DOF. The resulting (un-normalized) state after the PBS therefore reads

$$|\psi_{\text{f.e.}}\rangle = \int d\omega_1 d\omega_2\, \phi_{\text{f.d.}}(\omega_1,\omega_2) \times \left(\hat{a}^\dagger_{Dh_1\omega_1}\hat{a}^\dagger_{Dh_2\omega_2} e^{i\omega_1\tau_1}e^{i\omega_2\tau_2} + e^{i\varphi}\hat{a}^\dagger_{Dh_1\omega_2}\hat{a}^\dagger_{Dh_2\omega_1} e^{i\omega_2\tau_1}e^{i\omega_1\tau_2}\right)|0\rangle\,, \tag{14}$$

where the subscript f.e. means frequency entangled and the operator $\hat{a}^\dagger_{Ds\omega}=(\hat{a}^\dagger_{Hs\omega}+\hat{a}^\dagger_{Vs\omega})/\sqrt{2}$ creates a photon in the diagonal polarization state with frequency ω in the spatial mode s. Diagonal polarization of both photons has been achieved by a postselective measurement in Ref. [15] (not shown in Fig. 1). The polarization state of the photons is neither manipulated nor filtered or measured after the generation of the frequency-entangled photons in the experiment [15]. This is why we may discard it from here on, i.e., apart from the frequency we have only the spatial mode as the only left photonic DOF. Thus, we have $\boldsymbol{\sigma}=s\in\{U,L\}$.

We also included the additional parameter φ in the state (14). This parameter controls the symmetry of the photonic wave function in the frequency DOF. The value $\varphi=0$ corresponds to a symmetric spectral wave function, while $\varphi=\pi$ corresponds to an antisymmetric spectral wave function as can be seen in Eq. (16).

We can rewrite the state (14) as

$$|\psi_{\text{f.e.}}\rangle = \int d\omega_1 d\omega_2\, \phi^{\varphi}_{\text{f.e.}}(\omega_1,\omega_2) e^{i\omega_1\tau_1} e^{i\omega_2\tau_2} \times \hat{a}^\dagger_{h_1\omega_1}\hat{a}^\dagger_{h_2\omega_2}|0\rangle\,, \tag{15}$$

where we have defined the frequency-entangled spectral wave function

$$\phi^{\varphi}_{\text{f.e.}}(\omega_1,\omega_2) = \phi_{\text{f.d.}}(\omega_1,\omega_2) + e^{i\varphi}\phi_{\text{f.d.}}(\omega_2,\omega_1). \tag{16}$$

The state (15) then passes through the 50 : 50 BS and it transforms into

$$|\psi_{\text{f.e.}}\rangle = \int d\omega_1 d\omega_2\, \phi^{\varphi}_{\text{f.e.}}(\omega_1,\omega_2) e^{i\omega_1\tau_1} e^{i\omega_2\tau_2} \times \left(\hat{a}^\dagger_{U\omega_1}\hat{a}^\dagger_{U\omega_2}e^{i\theta} - \hat{a}^\dagger_{L\omega_1}\hat{a}^\dagger_{L\omega_2}e^{-i\theta} + \hat{a}^\dagger_{U\omega_1}\hat{a}^\dagger_{L\omega_2} - \hat{a}^\dagger_{L\omega_1}\hat{a}^\dagger_{U\omega_2}\right)|0\rangle\,. \tag{17}$$

From this we can read off the photonic wave function after the beam splitter $\Phi_{\boldsymbol{\sigma}_1\boldsymbol{\sigma}_2}(\omega_1,\omega_2)=\Phi_{s_1s_2}(\omega_1,\omega_2)$, which is characterized by four scalar functions in ω_1 and ω_2 (since s_1 and s_2 respectively can take two values, U and L). We can arrange the wave function in a 2×2 matrix:

$$\Phi_{s_1s_2}(\omega_1,\omega_2) = \phi^{\varphi}_{\text{f.e.}}(\omega_1,\omega_2)\begin{pmatrix} e^{i\theta} & +1 \\ -1 & -e^{-i\theta}\end{pmatrix}. \tag{18}$$

The row and column numbering of Eq. (18) is U, V.

We now employ Eqs. (18), (9), and (10) to compute the joint detection probabilities $\boldsymbol{P}_{s_1s_2}(\tau_1,\tau_2)$, and we find

$$\boldsymbol{P}_{s_1s_2}(\tau_1,\tau_2) = \tfrac{1}{4}[\boldsymbol{R} + d(\tau_1,\tau_2)\boldsymbol{C}], \tag{19}$$

where the matrices $\boldsymbol{R}$ and $\boldsymbol{C}$ are given in the (s_1,s_2) basis by

$$\boldsymbol{R} = \begin{pmatrix} 1 & 1 \\ 1 & 1\end{pmatrix},\quad \boldsymbol{C} = \begin{pmatrix} 1 & -1 \\ -1 & 1\end{pmatrix}, \tag{20}$$

while the functional dependence on the delays τ_1 and τ_2 appears only in the function $d(\tau_1,\tau_2)$, which we specify immediately.

Note that we have chosen the bold font for $\boldsymbol{P}_{s_1s_2}(\tau_1,\tau_2)$ in Eq. (19) to emphasize the fact that it has to be read as a matrix. The different entries are equal to the probabilities of the various detection events. The row and column numbering of Eq. (19) is the same as in Eq. (18). For instance, the entry of row 2 and column 2 in Eq. (19) means that the probability of detecting both photons at detector L (see Fig. 1) is equal to $\boldsymbol{P}_{LL}(\tau_1,\tau_2)=1/4[1+d(\tau_1,\tau_2)]$.

The probability $P^{\text{c}}(\tau_1,\tau_2) := \boldsymbol{P}_{UL}+\boldsymbol{P}_{LU}$ for a coincidence measurement (i.e., to detect one photon at one detector U or L and the other photon at the other detector L or U) is given by

$$P^{\text{c}}(\tau_1,\tau_2) = \tfrac{1}{2}[1-d(\tau_1,\tau_2)], \tag{21}$$

with

$$d(\tau_1,\tau_2) = \frac{R^{\varphi}_{\mu\xi}(\tau_1,\tau_2) + S_{\mu\xi}(\tau_1,\tau_2)}{1+\cos(\varphi)\text{sinc}(2\mu/\xi)}, \tag{22}$$

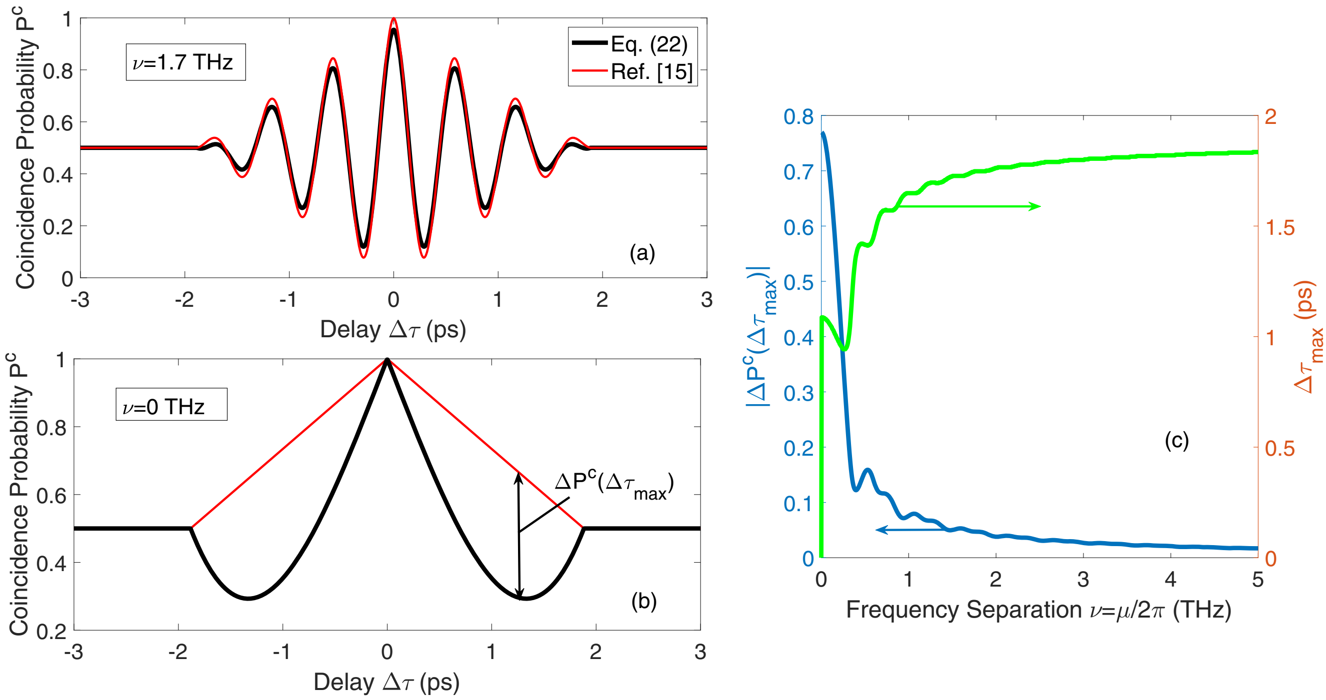

FIG. 2. Hong-Ou-Mandel interference experiment with frequency-entangled photons [see Eq. (16) for joint spectral profile]. Coincidence probability for a frequency separation of (a) $\nu = \mu/2\pi = 1.7$ THz and (b) $\nu = \mu/2\pi = 0$ THz. Note that our correction leads to a qualitative different behavior of the coincidence probability. The red curve in panel (b) predicts photon antibunching ($P^c > 0.5$) for all delays, where the black curve predicts photon bunching ($P^c < 0.5$) for some delays around $\Delta\tau \approx \pm 1.5$ ps. In panel (c) the blue curve shows the absolute value of the maximum discrepancy $|\Delta P^c(\Delta\tau_{max})|$ in the coincidence probability between our result [Eq. (22)] and the result that was obtained in Ref. [15] in dependence of the frequency separation $\nu = \mu/2\pi$. The green curve shows the delay $\Delta\tau_{max}$ at which this discrepancy is maximized. The single-photon bandwidth $\xi = 4/\tau_c = 1.356$ THz was taken from Ref. [15], where the coherence time from the mentioned reference was approximately $\tau_c \approx 0.3/0.885$ THz. The delay is $\Delta\tau = \tau_1 - \tau_2$. All computations were done for $\varphi = \pi$ in our evaluation of Eq. (22).

where we have introduced the functions

$$R^{\varphi}_{\mu\xi}(\tau_1, \tau_2) = \cos[\mu(\tau_1 - \tau_2) - \varphi]\,\mathrm{tri}\left(\frac{\xi(\tau_1 - \tau_2)}{2}\right), \quad (23a)$$

$$S_{\mu\xi}(\tau_1, \tau_2) = \frac{\sin\left[\frac{2\mu}{\xi}\mathrm{tri}\left(\frac{\xi(\tau_1-\tau_2)}{2}\right)\right]}{\frac{2\mu}{\xi}}, \quad (23b)$$

and

$$\mathrm{tri}(x) = \begin{cases} 1 - |x| & \text{if } |x| \leqslant 1 \\ 0 & \text{if } |x| > 1 \end{cases} \quad (24)$$

is the *triangular function*.

III. DISCUSSION

A. Relation to previous experiments

Our result (22) for the HOM effect extends the scope of application of the one obtained in previous work [15]. There, the expression $d(\tau_1, \tau_2) = R^{\varphi}_{\mu\xi}(\tau_1, \tau_2)$ was obtained, which coincides with our result in the limit $\mu/\xi \gg 1$ since $\lim_{\mu_\xi \to \infty} S_{\mu\xi}(\tau_1, \tau_2) = 0$. Let us call $d_0(\tau_1, \tau_2) := d(\tau_1, \tau_2)|_{\mu/\xi \to \infty}$. Since the previous experiment [15] was carried out in a parameter regime where one has $\mu/\xi > 10$, the discrepancy with our result was not measured. A qualitative difference between our result and the aforementioned result in the literature would have been revealed if the authors had driven the ppKTP crystal at lower temperatures, but they started their measurement series from the lowest crystal temperature of $T = 33.7\,^\circ$C, which corresponds to a frequency separation of $\nu = \mu/2\pi = 1.7$ THz [see Fig. 2(a)]. Nevertheless, also at this frequency separation a slight difference between our result and the one mentioned before can be seen. Our result (22) predicts slightly lower interference fringes. This tendency was already seen in the original investigation of frequency-entangled photons by Ou and Mandel [3], where this lower fringe visibility was attributed to an imperfect alignment of the measurement apparatus. However, we suggest that this discrepancy might originate, at least in part, due to our correction term $\Delta P^c(\tau_1, \tau_2) = d_0(\tau_1, \tau_2) - d(\tau_1, \tau_2)$. In the original derivation [15] this lower fringe visibility was explained by imperfect frequency entanglement of the photons.

A clear discrepancy between the approximation formula from the result found in the literature [15] and our result for the coincidence probability (22) would have been observed if the ppKTP crystal would have been driven only $10\,^\circ$C lower at a room temperature of about $T \approx 20\,^\circ$C, which would correspond to a frequency separation of $\nu = \mu/2\pi \approx 0.265$ THz. This would lead to a discrepancy of the coincidence probability to our result of $|\Delta P^c(\Delta\tau_{max})| \approx 0.2$ at a delay of $\Delta\tau_{max} \approx 1$ ps as seen in Fig. 2(c).

It is also worth noting that in the parameter regime of low-frequency separations, the additional term $S_{\mu\xi}(\tau_1, \tau_2)$ predicts physics qualitatively different than those previously obtained via the approximation formula [15], as can be seen in Figs. 2(b) and 2(c). For a delay of $\Delta\tau_{\max} \approx \pm 1.5$ ps, our result shows the occurrence of photon bunching (i.e., $P^c < 0.5$) where the approximation formula $d_0(\tau_1, \tau_2) = R^{\varphi}_{\mu\Delta}(\tau_1, \tau_2)$ here predicts photon antibunching (i.e., $P^c > 0.5$).

B. Physical interpretation

The additional term $\Delta P^c(\tau_1, \tau_2) \propto S_{\mu\xi}(\tau_1, \tau_2)$, which was not yet documented in the literature [15,16], has a concrete physical meaning as it quantifies the contribution of the spectral indistinguishability related to finite-bandwidth effects to the coincidence interference pattern, which becomes more dominant for lower values of μ/ξ and, in particular, in the limit $\mu \to 0$ of vanishingly small frequency separations.

To see this, it is illusive to repeat the calculation of the coincidence detection probability with the spectrum (13) of frequency-detuned photons in place of the spectrum (16) of frequency-entangled photons. This yields

$$P^c(\tau_1, \tau_2) = \tfrac{1}{2}[1 - S_{\mu\xi}(\tau_1, \tau_2)], \tag{25}$$

which precisely coincides with the corresponding result of coincidence detection probability of frequency-detuned photons found in the literature [20]. Contrasting this result with the interference pattern (22) of frequency-entangled photons shows up the meaning of spectral distinguishability in HOM interference and moreover relates this to the presence of entanglement between the spectral DOF and other DOFs of the considered photons, as we explain further below.

One can regard the spectral wave function (13) of frequency-detuned photons in the $\omega_1\omega_2$ plane in the limit $\mu \gg \xi$ as a distribution peaked around the point $[(\omega_p + \mu)/2, (\omega_p - \mu)/2]$, meaning one photon is emitted at frequency $\omega_1 \approx (\omega_p + \mu)/2$ and the other at frequency $\omega_2 \approx (\omega_p - \mu)/2$. As can be seen in state (12), the photon of frequency ω_1 is emitted in mode h_1 and the photon of frequency ω_2 is emitted in mode h_2, meaning that here we face a state in which the spatial modes of the photons are entangled with their frequencies. This is why the photons of state (12) are spectrally distinguishable from each other. The spectral distinguishability increases with increasing values of μ/ξ, i.e., with increasing entanglement between the spectral and the spatial DOF of each photon. This suppresses the tendency of frequency-detuned photons to bunch or antibunch [15], which is reflected in the vanishing of the interference term $S_{\mu\xi}(\tau_1, \tau_2)$ in Eq. (25) for increasing values of μ/ξ.

This emphasizes the role of distinguishability in the context of second quantization, in which no particle can be addressed individually by means of a "label" of the particle. This is the reason why it is often stated that in the second quantization formalism the involved particles are always considered as being indistinguishable. However, the second quantization formalism also carries a notion of distinguishability of particles, which is not encoded in the particle "labels" but in their properties (e.g., DOFs), and the interested reader is referred to Refs. [22,23] for the original and more detailed discussion on the topic, which was summarized in Ref. [24].

In the case of two completely indistinguishable photons of two frequencies, ω_1 and ω_2, it is impossible for an experimenter to detect (i.e., to address) a photon of a certain frequency, say ω_1. However, because the property of the photon's frequency (i.e., the photon's frequency DOF) is entangled with its transmission path (i.e., its spatial DOF) in state (12), the experimenter can simply place a detector in the transmission path h_1 to detect and address the photon of frequency ω_1 with certainty (in the limit $\mu/\xi \to \infty$).

In contrast to this, the spectrum (16) of frequency-entangled photons is invariant against the exchange of the function arguments ω_1 and ω_2 for $\varphi = 0$. Operationally, this means that an experimenter is unable to detect and address a photon of a certain frequency, $\omega_1 \approx (\omega_p + \mu)/2$ or $\omega_2 \approx (\omega_p - \mu)/2$ thorough *any* measurement. This is because [in contrast to the state (12)] the frequency of the photons is not entangled with their spatial modes (or other DOFs). The only information which is known with certainty is that at whatever frequency ω_1 or ω_2 one photon of the state (15) is detected, the other photon is detected at the other frequency ω_2 or ω_1. In other words the two frequencies at which the two photons are detected in state (15) are known, while the frequency of each individual photon is completely unknown, which is the essence of (spectral) entanglement [25].

This in turn shows up the relation between distinguishability and entanglement in second quantization. The impossibility to address a certain DOF of single particles (for instance, the frequency of a single photon) through *any* measurement corresponds to the indistinguishability of the considered particles with respect to this DOF, which is closely related to the presence of entanglement of the considered particles with respect to this DOF. However, one should note that the indistinguishability of two particles with respect to a certain property (DOF) is not equivalent but rather is a necessary condition for the presence of entanglement in the considered DOF. This means that two photons can be completely indistinguishable in their frequency DOF but nevertheless frequency unentangled (like it is the case with photons from the original HOM experiment [2]). However, the amount of frequency entanglement in a two-photon state is limited by the spectral indistinguishability of the considered photons.

This can be also seen as a consequence of the theorem of *entanglement monogamy*, which states that any pair of physical systems that are maximally entangled with each other cannot be entangled with any other physical system and that any entanglement with another, third, physical system comes at the cost of shrinking the amount of entanglement between the first two systems. This is why the interference term $S_{\mu\xi}(\tau_1, \tau_2)$ of the coincidence detection probability (25) of frequency-detuned photons vanishes in the limit $\mu \to \infty$, because with growing frequency separation μ the photon frequencies get stronger and stronger entangled with their spatial DOF (i.e., their transmission path), thereby becoming spectrally more and more distinguishable and thus imposing an upper limit on the exploitable frequency entanglement certified by the occurrence of photon antibunching [19].

C. Further mathematical considerations

Finally, we want to provide some mathematical arguments why the interference pattern $d_0(\tau_1, \tau_2)$, commonly used in the literature, cannot characterize the coincidence detection statistics of frequency-entangled photons in its whole generality. This can be best seen in the limit $\mu \to 0$. In this limit the spectrum of frequency-detunded photons (13) is invariant under the exchange of the function arguments [i.e., $\phi_{\text{f.d.}}(\omega_1, \omega_2) = \phi_{\text{f.d.}}(\omega_2, \omega_1)$] and thus coincides [apart from a prefactor $(1 + e^{i\varphi})$] with the spectrum of frequency-entangled photons (16). Thus, in the limit $\mu \to 0$ also the corresponding interference patterns of frequency-detuned and frequency-entangled photons should coincide and, in particular, be independent from the phase φ (because it only enters the calculations as a prefactor of the photonic wave function). Note that $\varphi = \pi$ [the case shown in Fig. 2(b)] is a special case which we discuss further below separately; i.e., we first consider the case $\varphi \neq \pi$. Indeed our results (21) and (25) are identical for $\varphi \neq \pi$ and $\mu \to 0$, and both result in $\lim_{\mu\to 0,\varphi\neq\pi} d(\tau_1, \tau_2) = \text{tri}(\xi\Delta\tau/2)$, where $d_0(\tau_1, \tau_2)$ does not reproduce the interference pattern of frequency-detuned photons (25) in the limit $\mu \to 0$ (except for $\varphi = 0$). For $\varphi = \pi/2$ and $\mu \to 0$ the approximation formula even predicts the absence of interference, i.e., $d_0(\tau_1, \tau_2) = 0$. It is interesting to see that for the special case $\varphi = \pi$ our result for the interference terms in the limit $\mu \to 0$ is $\lim_{\mu\to 0,\varphi=\pi} d(\tau_1, \tau_2) = -\Theta(\xi|\Delta\tau|/2)[(\xi|\Delta\tau|)^3 - 6\xi|\Delta\tau| + 4]/4$, which is shown in Fig. 2(b), where $\Theta(x)$ is the Heaviside step function. Despite $\varphi = \pi$ being an interesting mathematical special case, in the limit $\mu \to 0$ this special case is physically unstable, since any deviation from the value $\varphi = \pi$ forces the interference term to collapse to the triangular function. Moreover, the special case $\varphi = \pi$ is unphysical in the limit $\mu \to 0$ when one considers frequency-entangled photons, since in this case the corresponding spectral wave function of frequency-entangled photons (16) vanishes. However, we wanted to show this case here [see Fig. 2(b)].

IV. OUTLOOK

There are several possible extensions to this work. First of all, we think that our formalism can be easily extended to a much broader class of multiphoton interference experiments [21] and moreover also to atom interference experiments [26]. The analysis of many-particle interference with bosons, fermions, or both would be covered by our formalism by a generalized symmetrization operator $\mathcal{S}$ in Eq. (9), accounting for the respective parity of the wave function of the quantum system under consideration, and similar results in this direction have already been reported in combinatorial approaches in Refs. [27,28]. We think that these considerations can be translated into our formalism and vice versa. However, showing a strict mathematical analogy between these different formalisms remains a subject of future work.

Moreover, a general theoretical framework for the entanglement analysis in the second quantization formalism, which was recognized [29] early to be fundamentally different from the conventional entanglement analysis of distinguishable particles [30], would be desirable. However, despite tremendous progress [24,31,32] it is still a subject of ongoing research how to translate key concepts from standard quantum mechanics such as the execution of partial traces [33–35] and mixed states [32,36], various entanglement criteria [18,37–39], or the separability problem in general [40,41] to second quantization.

Furthermore, we want to emphasize that HOM experiments are ruled by the spectral properties of the employed light sources which are sensitive to one of the trademark predictions from general relativity, namely, the redshift on photons propagating through curved space-time. Moreover, as constituting highly accurate entangled photonic clocks, the study of frequency-entangled photons in HOM interference in a relativistic setting is of great interest. We intend to cover these aspects in a follow-up work.

Finally, we want to point out that the considerations of the present work might be connected to recent experimental advances in the study of the relation between entanglement and indistinguishability [42–46]. For instance, the authors of Ref. [42] conducted a direct measurement of the particle exchange phase of fermions, bosons, and anyons (particles with a mixed wave function parity) and showed that this can be useful for quantum-enhanced phase estimation. As frequency-entangled photons can reveal bosonic, fermionic, or anyonic behavior represented through the occurrence of photon bunching, photon antibunching, or a mixture of both in HOM experiments, they could be interesting to be considered as resources for similar studies addressing the physics of the wave-function parity. Other experiments recently demonstrated that the indistinguishability of photons can be used as a resource for various elementary processes in quantum technological applications, such as coherence generation [43], quantum teleportation [44], and remote entanglement distribution in quantum networks [45,46]. All these works suggest that the degree of coherence and, in particular, entanglement correlations are closely related to the indistinguishability of the involved photons. The theoretical relation studied in this work, between indistinguishability and quantum entanglement via the theorem of entanglement monogamy (for photons in HOM interference), might be used as an estimator for the exploitable resources in quantum technological applications due to the indistinguishability of the involved particles and could possibly constitute a step towards a better understanding and control of these quantum qualities in future experiments.

V. CONCLUSIONS

Indistinguishability and entanglement are two of the most important genuine quantum mechanical principles without classical analogs. Both these principles play a crucial role in HOM interference experiments which cannot be explained without considering the particle character of single photons which by itself can only be explained within an entirely quantum mechanical theory of electromagnetism. Therefore, HOM experiments are ideal candidates to study the quantum nature of photons and, moreover, the relation between indistinguishability and quantum entanglement.

Using Glauber's theory of optical coherence we developed a formalism to predict the detection statistics of HOM interference experiments in a systematic way where we treated all photonic DOFs on equal footing under the inclusion of entanglement correlations. This formalism naturally extends also to more complicated experimental setups and multiparticle quantum states.

We analyzed the role of indistinguishability and entanglement in HOM interference in the example of two fundamental two-photon sources: Sources which produce spectrally distinguishable frequency-detuned photons [20] and sources which produce spectrally indistinguishable frequency-entangled photons [15].

The comparison between these sources showed that the amount of entanglement which can be exploited in HOM interference through the occurrence of photon antibunching is limited by the spectral indistinguishability of the employed photons, which by itself is limited by the amount of entanglement between the spectral DOF of the photons with other DOFs, for instance, the spatial one. Therefore, we could relate the relation between indistinguishability and entanglement in HOM interference to the theorem of entanglement monogamy.

Apart from that, with our formalism we found an additional term in the interference pattern of frequency-entangled photons. Because frequency-entangled photons seem to be one of the most promising candidates to test fundamental aspects of physics and, moreover, because they can be used as a resource in quantum technological applications related to high-precision metrology, it is important to accurately characterize their spectral properties, also in the regime of low-frequency separations.

ACKNOWLEDGMENTS

We gratefully acknowledge Dennis Rätzel, David Edward Bruschi, and Andreas Wolfgang Schell for fruitful discussions on the topic. R.B. was funded by the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) under Germany's Excellence Strategy–EXC-2123 QuantumFrontiers, Grant No. 390837967. We further gratefully acknowledge support through the TerraQ initiative from the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation), Project-ID 434617780–SFB 1464, and the Research Training Group 1620 "Models of Gravity".

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