Quantum statistics of polariton parametric interactions

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Using a high-quality GaAs planar microcavity, we optically generate polariton pairs, and verify their correlations by means of time-resolved single-photon detection. We find that correlations between the different modes are consistently lower than identical mode correlations, which is attributed to the presence of uncorrelated background. We discuss a model to quantify the effects of such a background on the observed correlations. Using spectral and temporal filtering, the background can be suppressed and a change in photon statistics towards non-classical correlations is observed. These results improve our understanding of the statistics of polariton-polariton scattering and background mechanisms, and pave the way to the generation of entangled polariton pairs.

I. INTRODUCTION

While photons are the most obvious carriers for long-distance communication, using them in a quantum optical context is challenging. In order to realize strongly correlated photon pairs, two-qubit gates, quantum non-demolition measurements, and other quantum protocols, strong interactions between particles are required, but for photons interactions are extremely weak in vacuum. However, through mixing photons with interacting matter quantum particles one can obtain both strong interactions and long-distance information transport by tuning the interaction strength at will. Several theoretical works have shown that there are a multitude of schemes to create entangled states using microcavity polaritons, which are coherent superpositions of photons and quantum well (QW) excitons, ranging from polarization or energy-time entanglement to hyper-entanglement and cluster states raising interest in the quantum optics community. On the one hand, the photonic polariton component provides a direct interface to conventional optical methods for creation, information encoding, and detection of quantum states. On the other hand, their excitonic component provides strong interactions, much larger than in conventional non-linear optical crystals, which generate correlations. As the main interface to this system, external photons are coherently converted into polaritons in the microcavity and vice versa, conserving momentum, polarization and energy. Because of their interactions, polaritons can scatter with each other, changing their momentum, polarization or energy states. The polaritons are converted back into photons with a rate given by the photon fraction in the polariton divided by the microcavity emission lifetime. Because of this property one can regard polaritons as strongly interacting photons.

In fact, according to recent theoretical predictions, in properly designed samples, the interaction can be sizable (larger than the polariton linewidth) even at the single-photon level, which has interesting implications in quantum optics. However, until very recently, there has been no unambiguous demonstration of a single-polariton level non-linearity, and the values that can be reached in these confined systems are still to be explored.

Polariton scattering can be understood as a four-wave mixing process in non-linear optics: two pump polaritons are annihilated, and the signal/idler polaritons are created. The excited polaritons convert into photons on a picosecond timescale. The degenerate pump case, when using two identical pump polaritons, as shown in figure 1, is analog to the spontaneous parametric down-conversion process (SPDC). In the high excitation regime, the pump has an average polariton number much larger than one and can thus be treated as classical field, with an interaction Hamiltonian of polariton-polariton scattering.
The article is organized as follows. We begin by introducing Gaussian theory to compute correlation functions and witnesses in section II. Before applying the outlined theory to the experimental results in section IV, we discuss the experimental setup and some characteristics of the polariton source, in section III and we conclude the paper by identifying paths to increasing the quantumness of the emission in section V.

II. GAUSSIAN THEORY OF QUANTUM COHERENCE

In order to be able to produce entangled states from any pair process, it is necessary for the pairs to exhibit quantum correlations. We can measure these correlations using photon counting in a Hanbury Brown-Twiss (HBT) arrangement [26] (see figure 1). To make quantitative predictions about the magnitude of the measured correlations we employ a model that can be used to compute the underlying correlation functions and include the effect of uncorrelated background light superposed with the light from the polariton source.

Figure 2. Normalized triple coincidence rates \( n_0 = N^{(3)}_{\text{SSI}}(t - t_{S1}, t - t_{S2}, t)/N^{(3)}_{\text{SSI}}(t, \infty, t) \) (top row) and heralded coherence \( g_H^{(2)}(t - t_{S1}, t - t_{S2}, t) \) (bottom row) using the quantum correlators from equation (3) (left column) and the classical correlators (right column), with \( A_p = 0.003 \) and \( T_c = 10 \text{ ps} \).

We start by assuming that the underlying quantum state produced by our interaction process is a zero-mean Gaussian bi-photon state, which is fully characterized by utilizing non-classicality witnesses. In our measurements we find a clear link between background processes and the observed statistics on the boundary between quantum and classical physics.
only two non-zero temporal correlations, given by [27]:

\[ N_{SS}(t_1, t_2) = N_{II}(t_1, t_2) = A_p \exp\left(-\frac{(t_1 - t_2)^2}{2T_c^2}\right), \]

\[ N_{SI}(t_1, t_2) = \sqrt{\frac{A_p}{2\pi \epsilon}} e^{i\omega p t} \exp\left(-\frac{(t_1 - t_2)^2}{T_c^2}\right), \quad (3) \]

where \( N_{SS} \) and \( N_{II} \) are the auto-correlation in the signal and idler mode fields and \( N_{SI} \) is the cross-correlation between the two fields at a certain pump amplitude \( A_p \), pump frequency \( \omega_p \), and coherence time \( T_c \). Using Wick’s theorem, the correlations above can be used to construct higher-order coherences \( N^{(n)}_{AB} \), where A and B are arbitrary combinations of modes above. Since we perform photon counting measurements we are particularly interested in intensity correlations, which are second-order correlations \((n = 2)\) and in particular in the degree of second-order coherence for all combinations of modes in the HBT setup [28, 29]. For example, in the case of the cross-correlations, the coincidence rate between the signal and idler modes is given by

\[ N_{SI}^{(2)}(t_1, t_2) = |N_{SI}(t_1, t_2)|^2 + N_{SS}(t_1, t_2)N_{II}(t_1, t_2). \quad (4) \]

The second-order coherence is then given by

\[ g_{SI}^{(2)}(t_1, t_2) = \frac{N_{SI}^{(2)}(t_1, t_2)}{N_{SS}(t_1, t_1)N_{II}(t_2, t_2)} \quad (5) \]

\[ = 1 + \frac{|N_{SI}^{(2)}(t_1, t_2)|^2}{N_{SS}(t_1, t_1)N_{II}(t_2, t_2)}. \]

Second-order self-coherences \( g_{SS}^{(2)} \) and \( g_{II}^{(2)} \) and the two second-order self-coherences \( g_{SS}^{(2)}, g_{II}^{(2)} \) are computed analogously. For classical states the three coherence functions are bounded by a generalization of the Cauchy-Schwartz inequality, which can be derived from first principles (see for example [30] for further details) and is given by:

\[ g_{SS}^{(2)} g_{II}^{(2)} \geq g_{SI}^{(2)} g_{SI}^{(2)}. \quad (6) \]

The non-classicality of a weakly pumped SPDC-type source can be accessed via the heralded second-order coherence, where the signal mode is split by a 50:50 beam splitter with both arms detected by separate detectors. In this case the Hanbury-Brown Twiss (HBT) measurement is conditioned on the occurrence of an idler event and the corresponding signal photon has two pathways to reach a detector. In the ideal case this leads to sub-Poissonian anti-bunching [32]. Using the correlations given in equation (3), we can derive first the triple-coincidence rate and then the heralded coherence:

\[ N_{SSI}^{(3)}(t_{s1}, t_{s2}, t_1) = [2\Re \{N_{SS}(t_{s1}, t_{s2})N_{SI}(t_1, t_{s2})^*N_{SI}(t_{s1}, t_1)\} + \]

\[ + N_{SS}(t_{s1}, t_{s1}) |N_{SI}(t_{s2}, t_1)|^2 + N_{SS}(t_{s2}, t_{s2}) |N_{SI}(t_{s1}, t_1)|^2 \]

\[ + N_{II}(t_1, t_1) \left(|N_{SS}(t_{s1}, t_{s2})|^2 + N_{SS}(t_{s1}, t_{s1})N_{SS}(t_{s2}, t_{s2})\right) N_{II}(t_1, t_1), \quad (7) \]

\[ g_{HI}^{(2)}(t_{s1}, t_{s2}, t_1) = \frac{\langle \hat{a}^\dagger_1(t_1)\hat{a}^\dagger_{s1}(t_{s1})\hat{a}^\dagger_{s2}(t_{s2})\hat{a}_{s2}(t_{s2})\hat{a}_{s1}(t_{s1})\hat{a}_{s1}(t_{s2})\rangle \langle \hat{a}^\dagger_1(t_1)\hat{a}^\dagger_{s1}(t_{s1})\hat{a}_{s2}(t_{s2})\hat{a}_{s1}(t_{s1})\rangle}{\langle \hat{a}^\dagger_1(t_1)\hat{a}^\dagger_{s1}(t_{s1})\rangle \langle \hat{a}^\dagger_{s1}(t_{s1})\rangle \langle \hat{a}^\dagger_{s2}(t_{s2})\rangle \langle \hat{a}_{s2}(t_{s2})\hat{a}_{s1}(t_{s1})\rangle} = \frac{N_{SSI}^{(3)}(t_{s1}, t_{s2}, t_1)}{N_{SI}^{(2)}(t_{s1}, t_1)N_{SI}^{(2)}(t_{s2}, t_1)}, \quad (8) \]

where \( N^* \) denotes the complex conjugate of the correlations and \( g_{HI}^{(2)} \) conditions the second-order self-coherence on the measurement of an idler photon if the system were in a pure single biphoton state, \( N_{SSI}^{(3)} \) would be zero. One property common to all quantities defined above is that they only depend on time differences and the most interesting effects occur when all arrival times are close together.

The arrival-time dependence of the triples and the corresponding \( g_{HI}^{(2)} \) for both the low pump power quantum regime and the high pump power classical regime is given in figure 2. While the theory above assumes a continuous-wave pump, our measurements were performed in the pulsed regime, where the basic structure above is still valid, but only accessible in discrete form (when integrating over the detected pulses using finite integration windows). In our measurements the pulse repetition period \( T_{rep} \) is much larger than the lifetimes of the states \( (T_{rep} \gg T_c) \). Thus we can still use the results from above to characterize the behavior at zero time delay and asymptotically from the temporal distribution shown in figure 2 at large time delays in any direction. In order to capture the change from classical to quantum correla-
tions in this discrete framework we introduce the ratio $n_0$ between the value of the central peak $N_{SSI}(0,0,0)$ normalized to the asymptotic value $N_{SSI}(0, \infty, 0)$. For purely parametric light this ratio approaches different constant values, depending on the pump regime, as given by

$$n_0 = \frac{N_{SSI}(0,0,0)}{N_{SSI}(0,T_{\text{rep}},0)} \approx \frac{4\sqrt{2/\pi} + 2A_pT_c}{\sqrt{2/\pi} + A_pT_c} \approx \begin{cases} 4, & A_p \ll 1 \\ 2, & A_p \gg 1 \end{cases}. \tag{9}$$

For large pump amplitudes ($A_p \gg 1$) the value of $n_0$ is identical to the maximum value obtainable for a classical single-mode thermal state split into a “signal” and an “idler” mode. In the case of the classical parametric amplifier introduced in [27] the fields are composed of zero-mean jointly Gaussian random processes, where signal and idler photons are drawn from a bivariate Gaussian distribution. For this case we obtain the maximum value given by:

$$n_0^{\text{class}} = \frac{6A_p^3}{2A_p^2(1 + 2 \exp -\frac{T_{\text{rep}}}{T_c})} \approx 3, \tag{10}$$

independent of the pump amplitude.

In the pulsed case, the relevant temporal structure of the correlations can be captured with the time arguments discretized to multiples of the pulse repetition period,

$$n_{ij} = \frac{N_{SSI}(iT_{\text{rep}},jT_{\text{rep}},0)}{N_{SSI}(0,T_{\text{rep}},0)},$$

for $i, j \in \{-1, 0, 1\}$ and $T_{\text{rep}} \gg T_c$. The resulting $n_{ij}$ is shown in figure 3 for different pump amplitudes. For high pump powers the normalized triples converge to the values for purely classical thermal light as is detailed in figure 4.

In the framework established so far, a value $n_0 > 3$ would indicate non-classical behavior, however, care must be taken because the underlying theory is not general, since we assume particular statistical processes. In order to quantify the quantumness of our source in a general way we are going to analyze the measured photon emission probabilities, as outlined in the following section. In a real experimental setting the fluorescence from other excitations of the system or host material may cause strong background contributions in both the signal and the idler mode. Given that we know the nature of the background we can directly incorporate background light in the above model, as we will show in the next section.

A. Correlations of mixed light

In the following discussion we consider the case of a uniform background emission from a thermal source superimposed on the parametric emission. The main processes leading to background emission are all incoherent, because there is at least one scattering partner with random phase involved (phonons or excitons from the high k-vector reservoir). A coherent contribution could come from resonant Rayleigh scattering [33], but we strongly reduce it by spectral, spatial and polarization filtering (see, section §III) and can therefore neglect it. We therefore consider the dephasing to be fast enough to act as incoherent background light on the experimentally relevant time scales. In order to model this behavior we assume that an operator $\hat{F}_{\text{th}}$ creates additional photons in the same states as above leading to an effective photon annihilation operator $\hat{a}_{\text{tot}} = \alpha \hat{a} + \sqrt{1 - \alpha^2} \hat{F}_{\text{th}}$, where $\hat{F}_{\text{th}}$ obeys single-mode thermal state statistics with coherence.
the higher order correlations. The results from these calculations are similar to what we discussed above, where a gradual increase of the background contribution results in less pronounced anti-bunching and eventually bunching. The exact crossover depends on the driving strength of the parametric amplifier.

Figure 4. Theoretical pump power dependence of $n_{ij}$, as labeled, without background $\alpha = 1$ (dashed lines) and with $\alpha = 1/3$ (solid lines), having two thirds of the total counts from thermal background. The arrows indicate the asymptotic values towards the curves are converging towards.

length $T_{th}$ as defined by the following correlators:

$$N_{th}(t_1, t_2) = A_{th} e^{-i\omega_p t_1} \exp \left( -\frac{(t_1 - t_2)^2}{2T_{th}^2} \right),$$

$$N_{SI,th}(t_1, t_2) = 0.$$  

The cross-correlations are zero because $\langle F_i F_j \rangle_{th} = \langle F_i^\dagger F_j^\dagger \rangle_{th} = 0$. If we set all mixed terms like $\langle \hat{a}^\dagger \hat{F}_th \rangle$ to zero, corresponding to independent photon sources, the total two-time correlators are given by:

$$N_{SS, tot}(t_1, t_2) = N_{SS}(t_1, t_2) + N_{SS,th}(t_1, t_2),$$

$$N_{SI, tot}(t_1, t_2) = N_{SI}(t_1, t_2). \tag{11}$$

The contribution of each term depends on the contribution of parametric emission to the total emission given by the ratio $\alpha = A_p/(A_{th} + A_p)$. We can now use these correlators in the same way as in equation (3) and compute the higher order correlators. The results from these calculations are similar to what we discussed above, where $\alpha$ has a similar effect on the correlations as an increase of pump intensity - the higher the background contribution the lower the value of $n_0$ becomes until it approaches a value of three. We furthermore find that as the background becomes dominant, the power dependence of $n_{ij}$ becomes flatter, because of the power-independent cross-correlations of the thermal state. The conditional coherence for mixed light behaves in a similar way, where a gradual increase of the background contribution results in less pronounced anti-bunching and eventually bunching. The exact crossover depends on the driving strength of the parametric amplifier.

Figure 5. Calculated coincidence window dependence of the conditional coherence function $g_{th, in}$ for mixed light with different $\alpha$. The parameters are: the detector time resolution $T_{s1} = T_{s2} = T_1 = 35$ ps (FWHM), $T_{th}=100$ ps (FWHM), $T_c = 10$ ps (FWHM), and $A_p = 0.001 s^{-1}$.

**B. Including the temporal instrument response function**

In order to include the effect of the finite temporal response of the single photon detectors we convolve all the above quantities with the instrument response function of our detection system. As shown in figure [18] our best detectors exhibit an almost ideal Gaussian response we can therefore use convolutions with Gaussians $G(t, \tau)$ with a temporal width $\tau$. For example, the triple coincidence rate can be computed by the following expression [29]:

$$N_{SSI}^{IRF}(t_1, t_2, t_3) = \int dt_{S1} \int dt_{S2} \int dt_{1} G(t_{S1} - t_1, \tau_{S1})G(t_{S2} - t_2, \tau_{S2})G(t_1 - t_3, \tau_1).$$

We can now compute the number of triple coincidence events within a given coincidence window $[t - \tau, t + \tau]$

$$N_T(t_1, t_2) = \frac{1}{4\tau} \int_{-\tau}^{\tau} dt_1 \int_{-\tau}^{\tau} dt_2 N_{SSI}^{IRF}(t_1, t_2, 0),$$

where we have set $t_3 = 0$ for simplicity. All other quanti-
ties can be computed analogously and treated in the same way as described above. In order to illustrate the effect of the finite instrument response and the coincidence window we show in figure 3 the conditional coherence function \( \rho_{nm} \) of a mixture of parametric and thermal light after convolution and integration. It is evident that a coincidence window much smaller than the detector time resolution does not have any effect, whereas larger coincidence windows, greater than the coherence time, result in the signal becoming increasingly uncorrelated.

C. Nonclassicality characterization using witnesses

A state is nonclassical if it cannot be expressed by any statistical mixture of coherent states \([34]\). A quantity for characterizing the nonclassicality of single-photon states, as introduced in \([35]\), is defined in terms of photon emission probabilities \( P_0 = |\langle 0 | \rho | 0 \rangle|, P_1 = |\langle 1 | \rho | 1 \rangle| \) and \( P_{2^+} = 1 - P_0 - P_1 \) of a system described by the density matrix \( \rho \). Based on this, two witnesses are defined \([35]\):

\[
P_{2^+} < \frac{1}{2} P_1^2, P_{2^+} < \frac{2}{3} P_1^3, \tag{12}
\]

where the first inequality defines the non-classicality (NC) witness. If the measured probabilities fulfill the inequality, the underlying photon state is nonclassical. The second inequality is even more restrictive and defines the upper bound for the underlying quantum state to be additionally quantum non-Gaussian. A state is quantum non-Gaussian, if its Wigner function representation exhibits negativity. This means that the non-classicality witness can always certify that a given state is nonclassical, but false negatives are possible when \( P_{2^+} \gtrsim P_1 \) \([34]\), for example when detecting two-photon Fock-states.

We can compute the probabilities directly from the photon detection, as we will show later in this section.

For better illustration of the meaning of the above inequalities, we take three different states: a squeezed state from a parametric down conversion source, a single-mode thermal state and a coherent state from an ideal laser. The multiple versus single photon probability of the different states are shown in figure 6. The probabilities only depend on the mean photon number \( \mu \). In the regime of a weakly pumped source emitting low photon numbers \( \mu \ll 1 \), \( P_{2^+} < \frac{1}{2} P_1^2 \) is satisfied only by the squeezed state, while the coherent state of the laser marks exactly the boundary between classical and nonclassical (as given by equation (12)) and the thermal state is always above this boundary, see figure 6. The squeezed vacuum state above converges to the quantum non-Gaussian boundary for very small photon numbers, but still remains Gaussian. In the case of high average photon numbers the criterion does not allow the distinction between quantum and classical states.

The probabilities \( P_1 \) and \( P_{2^+} \) can be estimated directly from the measured detector counts in a coincidence-measurement, see figure 6 and \([36]\) for more details. Important for the derivation of the probabilities is only the total transmittance \( T \) (including losses and imbalance and relative detector efficiencies) of the last beam splitter in figure 7 splitting up the signal mode. The estimation of \( P_1 \) depends on the effective imbalance of the two detection channels \( T/(1-T) \) and if we assume, for example, that \( T > 1/2 \), we can give an upper bound on the transmittance, where \( T \leq T_c \) from the measured count rates \([36]\):

\[
T_c = \frac{N_{S1,1}}{N_{S1,1} + N_{S2,1}},
\]

where \( N_{Sn,1} \) are the double coincidence counts between the signal channels and the idler channel for \( n = 1, 2 \). As derived in \([36]\), we can estimate \( P_1 \) by the experimentally measurable quantity

\[
P_1^c = \frac{N_{S1,1} + N_{S2,1}}{N_I} - \frac{N_{S1,S2,1}(1 - T_c)^2 + T_c^2}{2N_I T_c(1 - T_c)},
\]

where \( P_1^c \leq P_1 \) with an error on the order of \( P_3 \) \([36]\) and \( N_{S1,S2,1} \) and \( N_I \) are the triples and idler-trigger counts, respectively. The probability to detect the vacuum state is given by:

\[
P_0^c = 1 - \frac{N_{S1,1} + N_{S2,1} + N_{S1,S2,1}}{N_I}.
\]

Putting all of the above together we can calculate the probability to detect two and more photons:

\[
P_{2^+} = \frac{N_{S1,1}^2 + N_{S1,S2,1}(2N_{S1,1} + N_{S2,1})N_{S2,1}}{2N_I N_{S1,1} N_{S2,1}}.
\]
We will use these expressions in section IV to characterize the light emitted from our polariton source.

In order to quantify the convex distance to non-classical states, we follow Ref. [36] in defining a witness function given by:

\[ W(a, r) \equiv ap_0(\mu) + p_1(\mu), \]

where \( a \) is a free parameter for optimization and the probabilities \( p_{0,1}(\mu) \) are computed from an ideal coherent state (Poisson distribution) with mean photon number \( \mu \) and we assume that \( p_0 + p_1 \leq 1 \) (low excitation regime). The convex distance from the experimentally determined probabilities is given by:

\[ \Delta W_f(a) = aP_0^\text{opt} + P_1^\text{opt} - W(a, \mu_{\text{opt}}), \]

where \( \mu_{\text{opt}} = 1 - a \) is the mean photon number maximizing \( W \). In order to obtain an upper bound for the distance we maximize \( \Delta W_f \) with respect to \( a \), i.e:

\[ \Delta W \equiv \max(\Delta W_f(a)). \]

Further details can be found in Ref. [36] and in the discussion of the experimental data in section IV.

III. EXPERIMENTAL DETAILS

The investigated sample is an epitaxially grown semiconductor structure that contains a \( \lambda \) cavity with a single 25 nm wide GaAs quantum well (QW) located at the anti node of the cavity field. Details about the sample can be found in [37]. The measured polariton linewidths are on the order of 120 µeV and the Rabi splitting is 3.6 meV [38]. The corresponding polariton lifetimes are on the order of 10 ps [33]. In contrast to most GaAs-based polariton samples reported in the literature, in our case, the unusually wide QW leads to three visible polariton branches, because the light hole-heavy hole splitting is only 4 meV, and thus, both the light hole and the heavy hole exciton are efficiently coupled to the cavity. However, we always work on the lowest branch, and the middle and upper polaritons can safely be neglected.

The experimental setup is shown in figure 7. The sample is held in a closed-cycle cryostat at temperatures in the range of 15-20 K, and is excited resonantly on the lowest branch by short (3-20 ps) laser pulses focused onto the sample by an aspheric lens (Edmund Optics #67-257), \( L_1 \), of focal length \( f_1 = 9 \) mm. We use a second lens \( L_2 \) at the distance \( f_1 + f_2 \) to focus the beam into the back-focal plane of the first lens. The excitation angle is set by a tilted mirror located in the back-focal plane of \( L_2 \).

The luminescence is collected through \( L_1 \), and passed through an optical relay consisting of two 500-mm focal length lenses, \( L_2 \), and \( L_3 \), respectively. The distance between \( L_2 \) and \( L_3 \) is given by \( f_2 + f_3 \) and produces an image of the back-focal plane of \( L_1 \) at distance \( f_3 \) from \( L_3 \), which are the two Fourier planes \( FP \). In these planes the signal and idler modes are spatially selected by two multi-mode fibers with 62 µm core diameter. The fibers are attached to 2-axis translation stages, and thus, arbitrary position in the momentum plane, and thus signal and idler modes can be chosen.

In order to select the real-space region from which the emission is collected, we spatially filter the emitted light by a pinhole \( PH \) in the common focal plane of the lenses \( L_2 \) and \( L_3 \), which is where the real-space image is formed. Spectral filtering is achieved by means of two interference filters, \( LF \), of full width at half maximum of 0.25 nm and a central frequency around the lower polariton resonance (\( \approx 817 \) nm). The filters are mounted on rotation stages to provide angle tuning in a range of about 6 nm. Since we turn the filters only by a few degrees around the center, there are only negligible changes in transmission line width and changes in polarization are adjusted using fiber polarization controllers. The light filtered in this way is split once more by a polarization independent 50:50 beam splitter \( BS \) in the case of the signal mode, collected through fibre collimators, matching the fibers, and then fed into the detectors. The detector signals are analyzed by a HydraHarp time correlator from PicoQuant. For the measurements in section IV we used avalanche photodiodes with a combined time resolution (detectors and counter electronics) of roughly 45 ps FWHM, while we used superconducting nanowire detectors with 35 ps FWHM for all other measurements. Further details about the detectors can be found in section V.
We measured correlations between two points in the far-field, i.e., at two defined polariton momenta, as shown on the left hand side of figure 8, which was acquired by a CCD camera inserted in one of the Fourier planes, FP, in figure 7, two pump polaritons scatter into a lower and a higher momentum state on a figure-eight shaped pattern given by energy-momentum conservation on the lower polariton branch, see also the data in Ref. [39, 40]. In what follows, we designate the low momentum state as signal (red), and the high momentum state as idler (green). The pump polariton direction has been blocked at the back-focal plane of $L_1$ to avoid overexposure. Typical focal spot sizes, measured in the real-space plane between $L_2$ and $L_3$ are about 80 $\mu$m (including the pinhole diameter), which leads to a mode size of about 0.1 $\mu$m$^{-1}$, and thus, the image in figure 8 contains approximately 5000 (70 by 70) modes. As mentioned earlier, the fibers are of diameter 62 $\mu$m. Taking the magnification of our setup into account, the fibre diameter corresponds to approximately half of a spatial mode size in the far-field.

The excitation wavelength is 815.6 nm and the pump power was set at $W = 100\mu$W pump power with the position of the pump (blue), signal (red) and idler (green). The shadow is from the small wrench that blocks the reflected pump light in the back-focal plane of $L_1$. Figure 9 is a typical correlation histograms, where the signal mode was set at $k_S = (-2.34, 1.6)\, \mu m^{-1}$ and idler mode at $k_I = (-3.5, -0.84)\, \mu m^{-1}$. The cross-correlation is shown at the top and auto-correlation of the signal mode at the bottom. The excitation wavelength is 815.6 nm and the pump power is 32 $\mu$W.

The cross-correlations above do not exist for modes that do not fulfill the phase matching condition. For the measurement on the right hand side of figure 8 we kept the momentum of the idler mode fixed and scanned with the signal fibre over a momentum range of about 0.7 × 0.7 $\mu$m$^{-2}$, as indicated by the small black square in the far-field image (8 left). At each fibre position, we plot the value of the correlations. The cross-correlation on the top right hand side of figure 8 (signal-idler) reduces to one at about 0.2 $\mu$m$^{-1}$ away from the phase matching in any direction, which is equivalent to a Fourier plane shift of four times the fibre diameter. Given that both

![Figure 8](image_url)

Figure 8. Left: Measured logarithmic far-field emission intensity (in arbitrary units) using 100 $\mu$W pump power with the position of the pump (blue), signal (red) and idler (green). The shadow is from the small wrench that blocks the reflected pump light in the back-focal plane of $L_1$. Right: auto-correlations of the signal beam at the red circle in the left graph (bottom) and cross-correlations between signal (red) and idler (green) beams (top) as a function of the pick-up momentum of the signal mode. The scan range is denoted by the small square around the signal (red) mode.

![Figure 9](image_url)

Figure 9. Typical correlation histograms, where the signal mode was set at $k_S = (-2.34, 1.6)\, \mu m^{-1}$ and idler mode at $k_I = (-3.5, -0.84)\, \mu m^{-1}$. The cross-correlation is shown at the top and auto-correlation of the signal mode at the bottom. The excitation wavelength is 815.6 nm and the pump power is 32 $\mu$W.

IV. RESULTS

A. General results and first observations

A typical correlation histogram, between the signal-signal and signal-idler modes is shown in figure 9. The histograms contain a train of pulses of nearly constant height, except for the one at zero delay, which is a sign of correlation. The pulses are separated by the repetition rate of the excitation laser. As a measure of correlations, we use the ratio of the area of the peak at zero time relative to the area of the peaks at ±13 ns, which corresponds to the second-order coherence (equation (5)) in the limit of low efficiencies [41] (we measured total efficiencies of roughly 0.4 % for all channels). The peak areas are computed by integration over a time window as wide as the repetition period (in this case 13 ns). A value of one corresponds to coherent light, and it is evident from the figure that we observe bunching at zero delay.
the mode size and the fiber diameter correspond to about 0.1 µm⁻¹, a simple convolution would lead to an apparent momentum range of about 0.15 µm⁻¹, slightly smaller than what we measure. We note that similar results were obtained by reversing the roles, fixing the signal momentum, and scanning with the idler, and also when we measured the auto-correlation (signal-signal) by setting both fibers to either the signal momenta, and scanning with one of them, as shown on the right hand side bottom panel of figure 8. In the case of auto-correlations we obtain a measurement of the mode size in k-space, whereas the cross-correlation measurement is sensitive to both the phase-matching condition and the mode size.

It is also clear from figure 9 that the auto-correlation is higher than the cross-correlation. The figure does not display the idler-idler histogram, which is very similar to the signal-signal histogram, but with lower count rates, because the idler mode is more excitonic and therefore, dimmer. It turns out that for almost all excitation powers the auto-correlation is larger than the cross-correlation, as demonstrated in figure 10. Only at low pump powers the correlations are similar in magnitude. Similar results were reported for 1D microcavities by Ardizzone et al. [12]. Also shown in figure 10 is a set of the corresponding emission intensity at the signal momentum as a function of excitation power. First, the intensity increases linearly, and at a certain threshold power it becomes quadratic, indicating that polaritons responsible for the emission are generated by the interaction Hamiltonian in equation (1). The intensity at the idler position (not shown) behaves in the same way with the same threshold, except that its value is reduced due to the lower photon content.

Figure 10 also demonstrates that the auto-correlation is higher than one only, when the pair intensity starts to overcome the uncorrelated background. We should mention here that the maximum of the correlations, which strongly depends on the exact location on the sample, always occurs at a somewhat higher excitation power than the threshold power between the linear and quadratic dependencies. As seen in figure 10 for this particular position, the threshold position is at around 60 µW, while the correlations reach their maximum at around 90 µW. A similar behavior was found in [23, 24, 43]. What is also clear from this plot, is that the bunching has a rather sharp threshold, above which it increases nearly linearly till it reaches a maximum, and then drops slowly. As the pump intensity increases, the polariton dispersion is renormalized because of the density-dependent repulsive interactions between polaritons [13]. In order to avoid spurious effects related to this renormalisation, we always re-align the fibers in such a way that the bunching was highest. The slow drop in the correlations can be attributed to the presence of more than one polariton in the cavity and the creation of multiple pairs [14]. Similar conclusions were drawn in [43].

The low pump power regime is dominated by background from bound state excitons [40] that trap the initially generated free polaritons until all impurities in the excitation region are saturated, which occurs approximately at 50 µW pump power as can be seen in figure 11. Unfortunately the energy and momentum range of these bound states has a large overlap with the experimentally accessible range for signal and idler polaritons [40] and it is therefore not possible to simply filter out all the background light. We, however, expect the signal-to-background ratio to improve if we additionally apply filtering in the time domain, because the radiative lifetime of the bound states is more than a factor of 5 longer than the polariton lifetime [40]. We analyze two and three photon measurements using the highest time resolution we could achieve in more detail in the following section.

B. High time resolution coincidence measurements

From the results in the previous section we can deduce that the optimal pump power lies somewhere below 80 µW and possibly even significantly lower when reducing the background further through temporal filtering. The goal is to find a setting where we can reduce the power as much as possible while still retaining some parametric signal above the background. We then record coincidence events with the HBT setup described above, but replace the APD with superconducting nanowire single photon detectors (SNSPDs) in order to achieve a higher total time resolution of ≈ 35 ps per channel. For the data presented in this section we use a bin width of 4 ps and again a pulsed laser as pump.

In the following analysis we use, unless otherwise specified, a dataset obtained with the following settings. We have set a pump power of 32 µW at 1.5204 eV and excite with $k_p = 1.34 \text{µm}^{-1}$. Through fitting a three oscillator

![Figure 10. Luminescence intensity of the signal of figure 8 (left axis, solid red circles), and signal-signal/signal-idler correlations (right axis, open blue squares/solid green triangles) as a function of the excitation power. The excitation wavelength is 815.8 nm. Uncertainties for the correlations, are in the 0.02-0.05 range, while for the intensity, they are smaller than the symbols.](image)
Figure 11. Temporal width of the background extracted when fitting the central peak of the histograms as in figure 9 with double-Gaussian distributions, for both the auto- and cross-correlations (as labeled). The underlying data is the same as for figure 10. For the fit we assume that the parametrically scattered polaritons have a constant width given by two-times the time resolution of the detection system, in this case we have $\sigma_{\text{pol}} = 2\tau = 324\text{ ps}$.

model [39] we find the photon-exciton detuning to be $\Delta C - \chi(0) = -2.2\text{ meV}$ and the heavy hole exciton energy is $E_{\text{HH}} = 1.52169\text{ eV}$.

Shown in figure 12 are the recorded triple coincidences as a function of the relative times between each of the two signal modes and the idler mode. Plotted are, on a $3 \times 3$-grid, only the central $\pm 80\text{ ps}$ around the pulse maxima (each cell in the blue grid) for integer multiples of $T_{\text{rep}} = 13.1\text{ ns}$ (cell spacing), corresponding to the pulse repetition period of the pump laser. Because of the short lifetime of the polariton states ($T_c < 10\text{ ps}$) the different regions in the $3 \times 3$-grid satisfy the condition $T_{\text{rep}} \gg T_c$ well.

Since we know that the background states have lifetimes $T_{\text{th}} \approx 100\text{ ps}$, much longer than the polariton states, we filter coincidences around the center of each pulse with varying coincidence windows $\tau_w$ in order to reduce the background contribution. To capture the influence of the different channels we additionally analyze the measured coincidences for all three permutations of channels assignments, i.e.:

$$n_{ij}^{(\text{SS})} = \frac{N_{\text{SSI}}(0, T_{\text{rep}}, 0)}{N_{\text{SSI}}(0, T_{\text{rep}}, 0)},$$

$$n_{ij}^{(\text{SI})} = \frac{N_{\text{SSI}}(T_{\text{rep}}, 0, 0)}{N_{\text{SSI}}(T_{\text{rep}}, 0, 0)},$$

producing delays along different physical channels. Shown in figure 14 is the dependence of $n_{ij}^{(k)}$ on the coincidence window width $\tau_w$. In figure 13 we compare coarse grained correlation histograms from the dataset used for figure 12 with a different dataset with a higher background contribution. Because we permute all channels the overall symmetry changes the “ridge”, namely where $t_{\text{S1}} = t_{\text{S2}}$, (see figure 3) from diagonal (like in figure 12) to along one of the axes. The first dataset (top row) shows the expected classical signature quite clearly, while the second one shows a more pronounced central peak with the components on the $t_{\text{S1}} = t_{\text{S2}}$-diagonal reaching below one, which approaches the non-classical distribution in figure 3. We expect the central peak to have the same height for all permutations, which is also observed apart from small variations.

Since the height of the central peak is theoretically unbounded in the case of the parametric amplifier source, while it remains constant for a classical source, we expect it to rise with decreasing $\tau_w$ as soon as enough of the long-lifetime thermal light is removed. We can see this rise in triples quite clearly as well as it starting to settle below $\approx 35\text{ ps}$, which corresponds to the time resolution of our setup. In addition, we observe that for the lowest filter values, $n_0$ reaches values above three indicating a departure from the classical parametric regime and the onset of the quantum regime, as discussed in section II.

In order to gain further insight we can additionally evaluate the data from coincidences between the combination of only two detectors (of the same measurement) in a similar way (as described in section IV A) to obtain the degree of second-order coherence along the different directions:

$$g^{(2)}_{AB}(0) = \frac{N_{AB}(0)}{N_{AB}(T_{\text{rep}})}.$$

The denominator in this case is given by the coincidence histogram delayed by (multiples of) $T_{\text{rep}}$. This gives either the second-order auto-correlation function $g^{(2)}_{\text{SS}}$, if the double coincidences are between the two “signal”-channels, or cross-correlation functions $g^{(2)}_{\text{SI}}$ otherwise.
Figure 13. Measured $n^{(k)}$ for two different datasets (rows) for $\tau_W = 12 \text{ ps}$ and binned into a single bin per pulse. From left to right: time delays $t_{S1}, t_{S2},$ and $t_I$ is kept at zero delay, respectively, while the other two channels are delayed. Note that the normalization is done by averaging over the nearest neighbor of the off-diagonal peaks. The bottom row shows the same dataset as figure 12 whereas the top row shows a similar dataset, at a different sample position, with more background contribution at a pump power of $37 \mu \text{W}$. All color bars span the same linear range from 0 to 4.

Figure 14. Coincidence window dependence of the normalized triples $n_0$ (equation 9) for the three different channel permutations. The dashed line indicates the maximum value reachable using the (classical) high pump limit (equation 10).

We have not measured the corresponding idler auto-correlation $g_{II}^{(2)}$ for this particular dataset, but we found from several other measurements that it takes on values similar to $g_{SS}^{(2)}$. The dependence of these quantities on $\tau_W$ is shown in figure 15. We can again see a clear dependence on the coincidence window and the cross-correlations reaching values $> 2$. Assuming that $g_{II}^{(2)} \approx g_{SS}^{(2)}$, which we found in an independent measurement, we estimate the Cauchy-inequality (equation 6) and find it to be violated for all cases at low coincidence window widths.

From the cross-correlation value we can also estimate the value of the heralded coherence function at zero delays (see 29):

$$g_H (0) = \frac{2}{g_{SI}^{(2)}} \left( 2 - \frac{1}{g_{SI}^{(2)}} \right),$$

(16)

where we take the geometric mean of the two cross-correlation combinations $g_{SI}^{(2)}$, often referred to as coincidence-to-accidentals ratio (CAR), as input, see figure 16. However, due to the still weak cross-correlations, the heralded coherence does not reach values below one, which is another requirement for a quantum source. In order to reach a truly non-classical value a CAR greater than $2 + \sqrt{2} \approx 3.4$ would be required.

Another interesting observation is that the auto-correlations reach values close to two, which means that the underlying effective mode number collected by the multi-mode fibers is close to one [44] and the background processes have to be emitting into the same (spatial) mode with thermal statistics.

We can now combine both double and triple coincidences to compute the heralded coherence (equation 8), again by computing the probability for multi-photon clicks by taking the central peak in figure 13 and normalizing it to the off-diagonal ($N_{XXX}^0$) peaks, i.e.
Figure 15. Coincidence window dependence of all three two-detector correlations for different reference channels, as labeled. From left to right: $t_{S1}$, $t_{S2}$ and $t_1$ is set as reference, respectively. The right-hand axis (purple) shows the corresponding $\Delta = \left( g_{S1S2}^{(2)}(0) \right)^2 - g_{S1I}^{(2)}(0)g_{S2I}^{(2)}(0)$, which below zero (dashed lines), violates the Cauchy-Schwarz inequality.

\[
g_{H,I}^{(2)} \approx \frac{N_{S1S2}(0)}{N_{S1S2}(0)N_{S1I}(0)N_{S2I}(0)},
\]

where we took the idler channel as the reference and restrict ourselves to that case in the following discussion.

The measured coincidence window dependence of $g_{H,I}^{(2)}$ is shown in figure 16. Interestingly, the conditional coherence does not exhibit the pronounced dependence on window size we found when looking at the coincidences alone because both the three-fold and two-fold coincidences change in the same way and the effect partially cancels. The temporal distribution of the more non-classical dataset can be seen in figure 16 (bottom). We can see a clear anti-bunching dip in the central peak when going along the diagonal values, however, the value of the central peak does not drop below one. The minimal value for the central peak in the case of the parametric amplifier in the classical regime is 1.5. In our measurements we reach values as low as $\approx 1.36(3) \cdot$ more than four standard deviation below 1.5. This is consistent with the bunching observed in the cross-correlations, where we find with equation (16) $g_{SI}^{(2)} \approx 2.31$. Even though there is clear anti-bunching, we want to point out that the obtained values correspond to super-Poissonian statistics and are still far above the non-classical limit.

For the final analysis we want to use the non-classical character witness defined in section II C and compare the single photon and multiphoton probabilities of our source. We compute the probabilities, as defined in section II C, as a function of the coincidence window width and plot them in the same way as in figure 6, resulting in the plot shown in figure 17. Interestingly for this measure the coincidence filtering seems to have a much greater effect than for the conditional coherence as can be seen in the right-hand figure in figure 17. By comparing the calculated probabilities to ideal states we can see that the underlying quantum state always lies somewhere in-between a classical thermal state and the coherent state and is therefore classical.

While filtering reduces the distance to states with non-classical character $\Delta W$ (given by equation (14)) significantly, we find it to be still approximately 2.6 standard deviations away from the boundary to the non-classical region at the smallest coincidence window, which is roughly what we expect from the value of the conditional coherence.

V. POSSIBLE IMPROVEMENTS AND CONCLUSIONS

The main contribution to the bunching at zero delay comes from the thermal background, which is difficult to remove. In order to significantly reduce the overlap with the parametrically scattered polariton states of the state space, a stronger Rabi splitting with simultaneously narrower linewidths could help. Based on our structure this would require an increase of the splitting by at least 1 meV ($\Delta E_{RS} = 3.5$ meV [37]), while still retaining the lowest possible free exciton linewidth. This could be achieved by using two 25 nm GaAs quantum wells in the cavity.

Another limiting factor in our measurements is the time resolution of the the detection system, which is still roughly a factor of 3 greater than the lifetime of the polariton states. If we assume that all detectors have a Gaussian response function we can extrapolate the increase in coherence we got between measurements performed with avalanche photo diodes with 45 ps FWHM (figure 10) and the SNSPD detectors. We find that a detection system with a total timing jitter of 10 ps FWHM could almost double the measured cross-correlation value giving rise to non-classical anti-bunching (using equation (16) we get $g_{H}(0) \approx 0.88$). Reaching such low jitter values might be possible in the near future [35]. A higher detector time resolution would at the same time also help separating the signal from the background in
time, which may yield a small additional increase in cross-correlations.

A different approach would be to confine polaritons in all three dimensions and use interference effects in coupled photonic dots to create entanglement. Quantum confinement additionally gives another handle to control the energy of the polariton states and at the same time, slightly increases the interaction strength. Very recently the successful generation of non-classical polariton states in a single photonic dot was reported. In conclusion, we have demonstrated that correlated photon pairs can be generated by means of polariton parametric scattering in planar microcavities. In particular, we have shown that the signal-signal, and idler-idler correlations are consistently higher than the signal-idler correlations. This finding is in stark contrast with the predictions of models based on simple four-wave mixing processes. We can explain the differences to the theoretical models by including uncorrelated background light. We attribute this background light to donors resonantly absorbing and re-emitting free excitons (polaritons) in the same phase-space region as the modes of the polariton parametric amplifier. We measured that with appropriate spatiotemporal selection at very low pump powers the photon statistics changes significantly. Fluctuations in photon numbers increase in both the triples and doubles measurements, anti-bunching gets more pronounced and the distance to quantum non-Gaussian emission characteristic is decreased significantly, but unfortunately, the values still stay in the classical regime. We consider different improvements that could reduce the background further and find that an increase in time resolution of the detection system would suffice to observe non-classical photon statistics.

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APPENDIX A

Table 1 provides the raw data extracted directly from a 15 hours time-tag measurement. This data can be used to reproduce the traces in figures 15 and 16, and we used $N_0 = \frac{N_{1S1}^0N_{1S2}^0}{N_{S1}^0N_{S2}^0}$:

| $\tau W$ | $N_0$  | $N_{S1}^0$ | $N_{S2}^0$ | $N_{S1S2}^0$ | $\Delta W$ | $\delta (\Delta W)$ | $g_1(0)$ |
|----------|--------|------------|------------|--------------|-----------|-------------------|---------|
| 4        | 380986080 | 304359    | 57152     | 67            | -2.1e-7   | 8.1e-8           | 1.465   |
| 12       | 379223482 | 888105   | 168648    | 540           | -1.42e-6  | 2.28e-7          | 1.302   |
| 20       | 416809617 | 1411497  | 279937    | 1333          | -3.68e-6  | 3.25e-7          | 1.444   |
| 28       | 424958538 | 1850823  | 358853    | 2245          | -5.8e-6   | 4.1e-7           | 1.423   |
| 36       | 427447870 | 2194373  | 43013     | 3155          | -7.99e-6  | 4.8e-7           | 1.413   |
| 44       | 428759185 | 2447406  | 484692    | 3910          | -9.55e-6  | 5.29e-7          | 1.396   |
| 52       | 440205249 | 2622812  | 524654    | 4513          | -0.00001121 | 5.5e-7       | 1.425   |
| 59       | 440384359 | 2738787  | 551939    | 4925          | -0.00001122 | 5.64e-7      | 1.436   |
| 68       | 450183896 | 2812835  | 570090    | 5189          | -0.00001124 | 5.72e-7      | 1.437   |
| 76       | 455177765 | 2859698  | 581303    | 5375          | -0.00001132 | 5.75e-7      | 1.451   |
| 84       | 453838553 | 2859698  | 581303    | 5375          | -0.00001134 | 5.82e-7      | 1.444   |
| 92       | 454860004 | 2908194  | 592454    | 5549          | -0.00001136 | 5.83e-7      | 1.444   |
| 100      | 456618971 | 2921304  | 595115    | 5592          | -0.00001137 | 5.83e-7      | 1.448   |
| 108      | 457247663 | 2931057  | 596773    | 5615          | -0.00001137 | 5.84e-7      | 1.447   |
| 116      | 458948286 | 2938524  | 597919    | 5632          | -0.00001138 | 5.83e-7      | 1.445   |
| 124      | 459435903 | 2944550  | 598754    | 5647          | -0.00001138 | 5.83e-7      | 1.445   |
| 132      | 458480851 | 2949705  | 599453    | 5663          | -0.00001138 | 5.86e-7      | 1.447   |
| 140      | 458979902 | 2953990  | 600035    | 5677          | -0.00001139 | 5.86e-7      | 1.449   |
| 148      | 459510005 | 2957740  | 600526    | 5679          | -0.00001139 | 5.86e-7      | 1.448   |
| 156      | 460134970 | 2961157  | 600946    | 5684          | -0.00001139 | 5.85e-7      | 1.449   |
| 796      | 470727911 | 3032347  | 610362    | 5803          | -0.00001406 | 5.82e-7      | 1.453   |
| 1596     | 475778960 | 3067924  | 615553    | 5848          | -0.00001401 | 5.78e-7      | 1.452   |
| 3996     | 487707513 | 3126042  | 624531    | 5931          | -0.00001407 | 5.7e-7       | 1.46    |

Table I. Experimental raw data used for computing the conditional coherence and non-classical character witnesses.

APPENDIX B

In this paper we show data from two different detector types, avalanche photodiodes from Micro-Photon Devices (PDM Series) and superconducting nanowire detectors from Single Quantum. The corresponding time responses of one of the detectors is shown in figure 18. The time response of the SNSPDs is almost ideally Gaussian and significantly narrower.
Figure 18. Total time response of two different detectors, as labeled. Both responses include the time response of the counting system.