Dynamics of a pulsed single photon source

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Abstract. We propose and demonstrate a method for an independent verification of a degree of single photon purity and coherence applicable for all single-photon emitters used in pulsed mode. Using two-time second-order correlation measurements, we reconstruct the dynamics of the nonclassical photon wavepacket. This reveals the temporal evolution of the photon field during the excitation-relaxation cycle of an emitter. The technique allows for the simultaneous measurement of multiphoton content and decoherence. Here we applied this technique to characterize a nonclassical state produced from a single InAs quantum dot (QD). We experimentally observe variations in the degree of multiphoton content and coherence of the wavepacket. A rate equation is introduced to explain multiphoton variations of our source and is found to describe the observed two-dimensional second-order correlation function accurately.

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INTRODUCTION

Single photon generation is of a paramount importance for the numerous experiments in quantum optics. These experiments range from a development of quantum information protocols \cite{1, 2, 3, 4, 5} to answering most fundamental questions of nature, in particular, Bell-like tests \cite{6, 7, 8, 9}. First and foremost, a nonclassical state that approximates a single photon state for an experiment must be duly characterized. A characterization of a single-photon-like state normally includes a test for a multi-photon component \cite{10, 11, 12}, and a test of indistinguishability of subsequent photons \cite{13}. Some foundational experiments and most of scalable quantum information protocols rely on discrete (pulsed) single photon states. Single-photon purity and coherence are generally treated statically in experiments, but depending on the excitation and the environment, in many systems these properties will be dynamic. A proper description (for instance, reconstruction of a wavefunction) of pulsed states requires a separate consideration of time within the emission cycle. Therefore, it is imperative that verification of pulsed non-classical states to include temporal effects, to observe changes in single photon purity and/or indistinguishability with time.

There are quite a few physical systems that can produce single photon states. They can be divided into two main categories: heralded sources (such as these based on parametric down-conversion or four-wave mixing), and single quantum emitter systems (such as an atom, ion, quantum dot (QD), or a color center). These physical systems are governed by different laws of physics, but in most cases properties of their non-classical output state would depend on where in the emission cycle a photon is detected. We il-
Illustrate this technique using a semiconductor QD with pulsed excitation. The technique allows for the independent characterization of single-photon purity and coherence during the excitation-relaxation cycle of the QD. We observe that photons detected early in the cycle have reduced single-photon purity and coherence compared to those detected later. We show instances when a traditional characterization implies that a source produces single-photon-like states, while our time-dependent method shows regions in an emission cycle with an unacceptable multiphoton content. An exciton recapture model describes single-photon purity dynamics.

**METHOD**

A characterization of a single-photon emitter normally includes measuring a second-order correlation function in two settings. First, a Hanbury Brown and Twiss (HBT) measurement is used to check for multiphoton content (or purity). In this measurement an input field is sent to a 50/50 beam splitter, with two click/no click detectors: one at each output port. A correlation function between photoelectronic detections of the two detectors is measured. Second, indistinguishability (coherence properties) are assessed with the help of Hong Ou and Mandel (HOM) interferometer. In this case, the two subsequent outputs from a source are overlapped on a 50/50 beamsplitter. The detection system (the two detectors: one at each output of a beamsplitter) and the data collection is identical to that of an HBT measurement. In an HOM experiment, a second-order correlation function retains the effect of second-order interference between the two input fields, providing a good measure of coherence, and single photon purity of both fields. In typical second-order correlation measurements (i.e. HBT and HOM measurements), a stationary emission process is implied, and thus a second-order correlation function \( g^{(2)} \) can only depend on \( \tau \), where \( \tau \) is a time difference between clicks on the two detectors. For non-stationary processes, this treatment obscures the dynamics of emission, because measured values are integrated over the whole excitation-relaxation cycle. Thus, a two-time correlation function \( g^{(2)}(t_1,t_2) \) should be used, where \( t_{1,2} \) are the times between a start of each trial (here, an excitation pulse) and detections on detector 1 and 2, respectively. A resulting 2-dimensional histogram reveals a dependence of a parameter in question on time within the cycle. This information can in general be used to build an underlying physical model of the interactions within a non-classical emitter or between a classical emitter and its immediate surroundings.

In our contribution, we illustrate the above method with the example of optically pumped single QD. It turns out that the observed \( g^{(2)}(t_1,t_2) \) functions strongly depend on times \( t_{1,2} \), as opposed to just depending on \( \tau = t_2 - t_1 \). This dependence is different for different excitation wavelengths. In addition to measuring the time-dependent characteristics of the QD output, we find an appropriate physical model that explains the temporal effects seen. Certain excitation wavelengths populate one or more reservoirs with carriers; for example, the host semiconductor’s conduction and valence bands and/or wetting layer states. Over time these carriers either get captured in the QD or decay by other channels. Thus a single quantum dot can produce more than photon per cycle due to re-excitation from a reservoir. As a result, a clear time-dependent signature of photon purity arises.
EXPERIMENTAL SETUP

The sample with strain-induced InAs QDs is maintained at 5 K in a cryostat and excited by a mode-locked Ti:sapphire laser with a repetition rate of 76.1 MHz (period $= 13.14$ ns) and 8 ps pulse duration. The emission from the QD is collected by a fiber-coupled objective lens and sent to the HBT/HOM measurement device shown in Fig. 1. To measure the time-dependence of the multiphoton emission probability (an HBT mode) we remove the first beamsplitter. With both beamsplitters in place, one arm of the interferometer is exactly three periods ($3 \times 13.14$ ns) longer than the other so the emissions from two separate excitation pulses meet simultaneously at the final beamsplitter. The outputs of the beamsplitter are each sent to a single-photon avalanche detector (SPAD). A time-correlated single-photon counting system records the time differences, $t_1$ and $t_2$, between each SPAD detection, and a nearest synchronous signal from the mode-locked laser. From this raw data we construct the two-time second-order correlation function, $g^{(2)}(t_1, t_2)$.

We recorded $g^{(2)}$ data in an HBT configuration at 755 nm which is above the GaAs band-gap, and two quasi-resonant wavelengths, 893 nm and 904.1 nm. A quasi-resonant peak at 893 nm sits on a broad absorption background of a wetting layer. A peak at 904.1 is isolated from that absorption background. Above-band excitation at 755 nm will excite carriers which rapidly decay to the band edge energy whence one or more is then captured by the QD. Quasi-resonant excitation nominally excites an exciton directly into the corresponding QD energy level, meaning that no additional carriers should be excited and multiphoton emission due to recapture is eliminated. However, at 893 nm multiple carriers from the wetting layer will also be excited. Therefore, one may expect somewhat different scenarios of interactions between a single quantum emitter (a QD) and its immediate vicinity, depending on an excitation wavelength.

![FIGURE 1. Schematic of the experimental setup. The first beamsplitter (marked A) can be removed to obtain an HBT configuration. With the first beamsplitter in place, an HOM measurement takes place.](image-url)
EXPERIMENTAL RESULTS AND DISCUSSION

Single photon purity

Figures 2(a)-(c) show the measurement results of autocorrelation function $g^{(2)}_{HBT}(t_1, t_2)$ in an HBT setup for the three excitation wavelengths 755 nm, 893 nm, and 904.1 nm, respectively. Figure 2(f) plots a cut through a $g^{(2)}_{HBT}$ surface along $t_1 + t_2 = 1.6$ ns diagonal. The experimental results clearly show a dependence of multi-photon content on when during an excitation - emission cycle a photodetection occurred. For excitations at 755 and 893 nm (Fig. 2(a)-(b)) note that the value of $g^{(2)}_{HBT}(t_1, t_2)$ is greater when either $t_1 \approx t_2 \approx 0$ or both, i.e. when the QD has emitted a first photon early in the cycle. This is because for a short time after the excitation pulse there are still free charges (either in the bulk of the semiconductor or in the wetting layer) that can excite the QD for the second time. This allows subsequent emission of more than one photon from a single cycle. The longer the QD holds on to the first photon, the less likely it is that another exciton will be captured.

Much of the behavior of $g^{(2)}_{HBT}(t_1, t_2)$ in Fig. 3 is related to the occupation and decay dynamics of various non-QD states under different pumping conditions. Different excitation wavelengths populate reservoirs in the GaAs and InGaAs wetting layer with
carriers differently. Over time these carriers either are captured in the QD or decay by other channels such as recombination through bulk exciton states or wetting layer states. Upon excitation, a band with a large density of states (the conduction band for 755 nm excitation and the wetting layer for 893 nm excitation) is filled with a certain density of electrons, leaving an equal density of holes behind. Over time these carriers either get captured by the QD or decay by other channels, such as recombination through the bulk exciton state or the wetting layer. For simplicity we only consider the lowest exciton energy level in the QD and assume that the decay of carriers in the band is unaffected by the presence of the QD.

The equations for diagonal density matrix elements derived from the master equation for such an incoherently pumped system are

\[ \frac{d}{dt} \rho_{11}(t) = f(t)\rho_{00}(t) - \Gamma_1 \rho_{11}(t) \]

\[ \frac{d}{dt} \rho_{00}(t) = -f(t)\rho_{00}(t) + \Gamma_1 \rho_{11}(t) \]

where \( f(t) \) is capture rate of carriers into the QD from the reservoirs, \( \Gamma_1 \) is the emission rate for the ground state exciton. See [14] for a similar set of evolution equations including the biexciton state. Note that off-diagonal elements \( \rho_{01}(t) \) and \( \rho_{10}(t) \) are not relevant for this calculation.

Carrier population in reservoirs decays exponentially, at different rates for each reservoir. The resulting expression for the capture rate is

\[ f(t) = \sum_i \tilde{n}_i \exp(-\gamma_i t) \quad (1) \]

where \( \tilde{n}_i \) is the product of the capture rate for a single carrier and the initial number of carriers in the \( i \)th reservoir, and \( \gamma_i \) is the recombination rate of carriers in the \( i \)th reservoir. There may be several distinct decay paths with different values of \( \tilde{n}_i \) and \( \gamma_i \), such as the bulk exciton, defects in the vicinity of the QD, or the wetting layer. The above master equations with a reservoir description of the form 1 allow calculating \( S^{(2)}_{\text{HBT}}(t_1, t_2) \) theoretically. Figs. 2 (d)-(e) show the results of such modeling for the excitations at 755 and 893 nm respectively with \( \Gamma_1 \approx 1.5\text{GHz} \), obtained independently from streak camera measurements, and with \( \gamma_i, \tilde{n}_i \) values obtained from fitting the experimental data and averaged over the detector jitter to match the experimental conditions. A rectangular shape of the valley with low \( S^{(2)}_{\text{HBT}}(t_1, t_2) \) values for 893 excitation occurs because there is only one reservoir with a relatively fast decay (on the order of 5 GHz). In contrast, an oval shape of the similar valley for 755 excitation is due to contributions from two reservoirs: a fast-decaying reservoir with a strong coupling constant and a slow-decaying reservoir with small coupling. A comparison of data to a model shows that main features of dynamics seen experimentally are captured. It follows from this model that in the limit when the capture rate is large compared to the emission rate, the source is in general not anti-bunched: it approaches Poissonian statistics. In contrast, when the capture rate is small compared to the emission rate, the source becomes substantially anti-bunched. In Figs. 2 (d)-(e) at short time periods the capture rates are similar to
the emission rate, therefore the values of $g^{(2)}_{\text{HBT}}(t_1,t_2)$ are large. At long time periods, the capture rate is much less than the emission rate, therefore $g^{(2)}_{\text{HBT}}(t_1,t_2)$ approaches zero. In previous work where only a one-dimensional $g^{(2)}_{\text{HBT}}(\tau)$ is considered for a pulsed source, the dynamics, especially the central valley, are lost due to time averaging. These pump-dependent features are clearly identifiable in our experiment, although some fast dynamics are obscured by the jitter of the detectors.

We conclude that according to the model above, it is expected that any single-emitter-based single photon source with a reservoir-like excitation will produce a field with statistics that approaches poissonian early in the excitation-relaxation cycle, regardless of the excitation mode. One way to avoid this effect is to excite a single emitter directly, resonantly or quasi-resonantly, as is done in our experiment, Fig. 2(c). We see that such excitation results in a relatively flat and low $g^{(2)}_{\text{HBT}}(t_1,t_2)$ because an exciton is excited directly into the QD with no possibility to populate reservoirs.

### Single photon indistinguishability

To address the coherence properties of the field emitted by a nonclassical source we use a two-time resolved HOM cross-correlation measurement. In this measurement two photons are incident on the beamsplitter: one from each of the two inputs. If these photons are indistinguishable, then there will be a two-photon Fock state at one output and no photons at the other. This bunching process has been called coalescence [15, 16, 17]. A measurement procedure yields a second-order correlation function $g^{(2)}_{\text{HOM}}(t_1,t_2)$, that includes the effects of coalescence and multiphoton emission from both input sources. Because we are interested in coalescence properties only, we need to separate the two

\[ g^{(2)}_{\text{HOM}}(t_1,t_2) \]
contributions. An expression for the coalescence can be derived from the definition of the HOM cross-correlation for the special case of identical emitters:

\[ C(t_1, t_2) = 1 + g^{(2)}_{\text{HBT}}(t_1, t_2) - 2g^{(2)}_{\text{HOM}}(t_1, t_2). \]

Thus, the statistical effects of multi-photon emission are removed from the HOM data by means of an independent \( g^{(2)}_{\text{HBT}}(t_1, t_2) \) measurement. The coalescence measures the degree of indistinguishability of the emission from separate excitations, conditional on detecting one photon at time \( t_1 \) and another at time \( t_2 \) after the laser pulse. Therefore the coalescence can be viewed as a measure of the indistinguishability of emission from consecutive excitations, regardless of the photon statistics, and \( C(t_1, t_2) \) has a range \( 0 \leq C(t_1, t_2) \leq 1 \): 0 for distinguishable photons, and 1 for indistinguishable ones.

Figure 3 shows \( C(t_1, t_2) \) for excitation at 755 nm. As expected for a state with decoherence, the highest coalescence probability is a ridge along \( t_1 = t_2 \). \( C(t_1, t_2) \) decreases monotonically away from \( t_1 = t_2 \), with the width of the ridge proportional to the photons’ coherence time. With our detectors, \( C(t_1, t_2) \) gets averaged over the jitter time of the detectors and the maximum value becomes proportional to the width of the ridge. The coalescence values along \( t_1 = t_2 \) are projected onto the rear planes of the figure, and we see that \( C(t_1, t_2) \) is smaller for small \( t_1 \) and \( t_2 \) then for later times. This shows that during the transient time after the initial pump pulse the indistinguishability of the state is significantly reduced.

This experimental result above demonstrates the time-dependent coherence loss in an emitter. In QDs it is associated in part with incoherent excitation first predicted by Kiraz et al. [18]. It was pointed out that for high QD decay rates, indistinguishability will be degraded when using incoherent pumping because the jitter in the capture time becomes comparable to the lifetime. However, here we observe an additional dynamical effect: though the lifetime is long compared to the excitation jitter, for measurement times comparable to the jitter there is an additional dynamical reduction in the indistinguishability. In addition to jitter, the dynamical reduction could also be due to extra dephasing from non-equilibrium carrier population around the QD.

**CONCLUSIONS**

We presented a method for characterization of pulsed single-photon sources that reveal additional information about the physical interactions within a source and/or between a source and its surroundings. We applied this method to a single QD emitter, and characterized its non-classical dynamics. We show that both auto-correlation and cross-correlation data have temporal structure which depends on the pumping method used. In several pumping schemes, the dynamics of auto-correlation are consistent with a multi-carrier capture model. We show situations where typical time-difference measures indicate a single-photon character in the emission statistics while the full dynamics indicate temporal regions where it is not. This result not only bears on optically excited emitters, but also electrically excited single emitters because such processes are based on probabilistic capture from a reservoir. The dynamics of the cross correlation is
also affected by the pulsed incoherent pumping, where a temporal dependence in the indistinguishability is observed, beyond the traditional decoherence behavior.

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REFERENCES