Quantum Science and Technology

PAPER

Interfacing a quantum dot with a spontaneous parametric down-conversion source

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Keywords: quantum interfaces, quantum dots, heralded single photons, Hong-Ou-Mandel interference

Abstract

Quantum networks require interfacing stationary and flying qubits. These flying qubits are usually nonclassical states of light. Here we consider two of the leading source technologies for nonclassical light, spontaneous parametric down-conversion and single semiconductor quantum dots. Down-conversion delivers high-grade entangled photon pairs, whereas quantum dots excel at producing single photons. We report on an experiment that joins these two technologies and investigates the conditions under which optimal interference between these dissimilar light sources may be achieved.

1. Introduction

In constructing quantum networks [1], light sources for long-distance quantum communications [2] play a pivotal role. Epitaxial semiconductor quantum dots are arguably the best developed single-photon source with many advantageous properties: they are mechanically and chemically stable, they don’t bleach and thus an individual quantum dot can be used for years. They offer well-understood and characterised energy levels that can be accessed optically and electrically, high quantum efficiency, short radiative lifetimes and low decoherence at moderate cooling to temperatures of a few Kelvins. With some engineering of the optical environment reasonably high collection efficiencies can be achieved [3, 4]. On the other hand, among sources of entangled photon pairs, it appears that spontaneous parametric down-conversion (SPDC), the conversion of a high-frequency photon to a pair of lower-frequency ones, is still the favourite. While SPDC suffers because it emits a random number of photon pairs and thus usually has to be operated at very low rates, SPDC is extremely tuneable in frequency [5] and can thus be matched to any quantum memory or stationary qubit. Therefore, we find it interesting to try and join these two technologies. Building on earlier results by Polyakov et al [6], we adapt the output of a bright SPDC source to the single photons emitted from an InAs quantum dot embedded in GaAs and demonstrate their Hong-Ou-Mandel interference [7]. This measurement forms the basis of linear optical Bell-state-analysis [8], quantum teleportation [9] and entanglement swapping [10]. These elementary operations feature in the conversion of stationary to flying qubits and in quantum repeater protocols.

Hong-Ou-Mandel interference is the coalescence of two indistinguishable photons into either output port of a beamsplitter. For complete indistinguishability in all photonic degrees of freedom (position-wavevector, time-energy, and polarisation) and a perfectly symmetric beamsplitter, two photons that arrive via the two input ports of a beamsplitter will always exit the beamsplitter together through a common output port and they coalesce. Thus, the detection rate of photons simultaneously exiting from the two different output ports (the so-called coincidence rate) will vanish. For two perfectly indistinguishable photons, there are two customary ways of establishing (partial) distinguishability. Either we, vary the polarisation of one photon or we apply a time delay. For delays much larger than the coherence time, the two photons become completely distinguishable and behave independently like classical, distinguishable particles. The coincidence rate will be exactly half of the incoming pair count rate, with the other half of the cases resulting in the two distinguishable photons randomly
The quantum dot

2. Quantum dot characteristics

The quantum dot (QD) sample we used in this experiment contained InAs QDs in GaAs embedded in a $4\lambda$ planar microcavity, where $\lambda$ is the quantum dot emission wavelength in the surrounding GaAs material. The microcavity consisted of 15.5 lower and ten upper DBR layer pairs of AlAs and GaAs, each layer being $\lambda/4$ thick. The sample was kept in a helium flow cryostat temperature stabilised to 5.0(3) K as measured at the cold finger. The excitation pulses were about 10 ps long and originated from a Ti:sapphire laser operating at a wavelength of 918.5 nm with a repetition rate of 82 MHz, which corresponds to a repetition period $T_\text{rep} = 12.2$ ns.

The photons emitted from a quantum dot show a different degree of indistinguishability depending on how the quantum dot is excited. In particular, resonant excitation to the biexciton enables higher indistinguishability than non-resonant pumping schemes [16]. This is due to two reasons: the resonant excitation creates the biexciton state jitter free [17] and the quantum dot environment does not suffer from electrostatic fluctuations present in the above-band excitation. The latter are caused by charge traps in the vicinity of the quantum dot, which are present even in the best man-made material. Unintentional dopants lead to deep levels that get randomly filled by the free carriers induced by a high-energy excitation laser and are randomly ionised again. Often this unwanted behaviour can be reduced by continuously applying a small amount of above-band light so as to keep these levels filled and thus reduce the fluctuation [18].

The two-photon resonant excitation method we used [19] coherently couples the ground state and the biexciton state via a two-photon resonance (see figure 1(a) for the energy schematics of a QD). The excitation laser wavelength lies about centred between the biexciton ($\lambda_{\text{XX}} = 919.6$ nm) and exciton ($\lambda_{\text{X}} = 917.7$ nm) luminescence wavelengths. Therefore, any scattered laser light may be rejected from the emitted luminescence photons by spectral filtering. Additionally, we used an orthogonal excitation geometry to further suppress the scattered laser light. This excitation method yields a high photon pair generation efficiency. It also enables the generation of time-bin entanglement [20].

In [16], we reported the indistinguishability of photons consecutively emitted by a quantum dot. The result, plotted in figure 1(b), shows an indistinguishability contrast of 0.39(2). This indistinguishability was measured using biexciton recombination photons. We showed in [16] that this result can be improved to 0.71(3) by active wavepacket shaping using a modulator. In the task at hand, we chose not to apply any active modulation because the SPDC pulses are very short in time and it is sufficient to employ temporal filtering of the detection events. Both types of filtering can improve the indistinguishability because the QD emission is generally not lifetime-limited, i.e., the QD emission is spectrally broadened, which leads to a coherence time $T_\text{c}$ that is shorter than twice the lifetime $T_1$.

It has been found [21] that the coherence time one extracts from HOM interference measurements between successive photons from a quantum dot is often longer than the one measured with a Michelson interferometer.
The likely reason for this is that any slow fluctuations, such as those of the charge environment, do not degrade the indistinguishability of successive photons too much if the environment is quasistatic between the two emission events that are only separated in time by a few nanoseconds. On the other hand, in a Michelson interferometer with a long and a short arm, any change in the central frequency (often called spectral diffusion) of the emitted photons will degrade the contrast of the averaged interference pattern and thus result in a shorter coherence time. This is consistent with our findings of [16], where we extracted a significantly longer coherence time from the HOM measurement than from the Michelson measurement. For the case at hand, the HOM interference between quantum dot and SPDC photons, we expect the effect to depend on the bandwidth of the SPDC. For extremely narrow filtering the drift of the central frequency should have a worse effect than for somewhat wider-band SPDC radiation. Nevertheless, as HOM interference is not intrinsically phase-sensitive, the effect of spectral diffusion will be less pronounced even though the apparent lengthening of the coherence time will still be observable.

### 3. SPDC characteristics

We built the spontaneous parametric down-conversion (SPDC) source in linear geometry. The nonlinear crystal we used was a 15 mm long type II periodically poled potassium titanyl phosphate (ppKTP) crystal, where the poling period (26.4 μm) was chosen for down-conversion of a single 460 nm photon into two 920 nm photons. By changing the temperature, it was possible to tune the down-converted photons to be non-degenerate. We exploited this tunability to match the central wavelength of the signal photons to the quantum dot biexciton photon wavelength of 919.6 nm. The SPDC source was pumped with the same pulsed Ti:sapphire laser as the quantum dot to generate photons synchronously. Since the SPDC source required a pump near 460 nm, we used second-harmonic generation in a beta barium borate (BBO) crystal to double the laser frequency.

The joint spectrum of the two SPDC photons in a pair is governed by energy conservation and phase matching. Energy conservation dictates that the frequencies of the signal and idler photons must sum to a frequency that lies within the pump laser spectral line. Phase matching is the approximate conservation of photon momentum and thus depends on the crystal refractive indices for the various involved modes. As a result, the joint spectrum is often highly anti-correlated with a broad distribution of frequencies for either photon individually, but a more or less narrow sinc-shaped bandwidth for the sum frequency [22]. The measured spectra of signal and idler of our SPDC source for the non-degenerate case together with the theory curves are shown in figure 2. Through the measurement of one photon, we can, in principle, determine the frequency of the other photon with high accuracy via the strong anti-correlation. In turn, this means that each photon’s individual state (traced over the partner photon’s frequencies) is in a mixed state and the photons have poor indistinguishability, leading to poor interference with independently created other photons. In our...
experiment, SPDC is used as a heralded single-photon source. While the signal arm of the SPDC source will interfere with the quantum dot photons, the partner SPDC photon (idler) serves as a trigger or herald for the presence of the other one. The heralding efficiency of the unfiltered source was 9.2%. For interference with independently created photons, narrow spectral filtering has to remove the frequency anti-correlation between the two SPDC photons. After filtering, the photon going to the beamsplitter will individually be in a pure state, fit for interference with the quantum dot photon.

To characterise the correlation properties of the down-conversion source itself, we performed a HOM interference measurement between the signal and the idler, with the temperature tuned to degeneracy of the source. Due to the narrow phase-matching function, even at degeneracy the signal and idler spectra are not identical, but the idler is wider than the signal, which can be seen (for the non-degenerate case) in figure 2. Thus, the indistinguishability of the unfiltered signal and idler photons was relatively poor, resulting in a visibility of only 39.9(4)% as shown in figure 3(a). Restricting the spectral bandwidth through a 30 GHz fibre Bragg grating (FBG) filter in one of the output ports of the beamsplitter strongly improved the indistinguishability and thus the HOM interference visibility to 96.3(2)% (see figure 3(b)). This happened with an FWHM pump pulse duration of approximately 10 ps, which is just a little shorter than the Fourier transform limit of 14.7 ps FWHM corresponding to the 30 GHz filter bandwidth. We include these results to demonstrate the effect of the FBG on the SPDC radiation which was used to generate one set of data quantum dot — SPDC HOM interfere reported below.

4. HOM Interference

To achieve high-quality interference between quantum dot emission and down-converted photons, their time/frequency shape is the crucial parameter. While position/wavevector, absolute time/central energy and polarisation can easily be adjusted or tuned, shaping the wavepacket or reducing dephasing is more difficult. The
The decay of an excited quantum dot state is predominantly radiative and spontaneous, resulting in an exponential time envelope of the emitted photons. The Fourier transform of this exponential decay function is a Lorentzian function in the spectrum. The SPDC photons’ individual spectra on the other hand are typically sinc shaped, as explained above. Besides the different spectral shapes of the photons from the QD and from SPDC, their bandwidth is also significantly different. While the QD emission has a natural width of about 1 GHz, the SPDC photons’ bandwidth is of the order of 100 GHz. To overcome this problem, we filtered the SPDC bandwidth down to 7.7 (1 GHz (FWHM), as estimated from a Michelson interference autocorrelation measurement. This was the practical technical limit of our filtering setup, but had the advantage of making active stabilisation of the center frequency obsolete. Unfortunately, this filtering reduced the (mean) heralding efficiency to 1.5%. As a filter, we used a pulse stretcher configuration of two diffraction gratings in a folded, space-saving 4-f configuration (see figure 4).

The entire setup is schematically depicted in figure 5. After the filter, the SPDC photons were delayed to match the arrival time of the quantum dot photons on the 50:50 beamsplitter by moving the element that coupled the SPDC signal photons to an optical fibre. This matching was only performed once and not changed thereafter. The Ti:sapphire laser was operated with the same parameters as described above. A beta barium borate (BBO) crystal frequency doubled a large part (≥300 mW) of the laser power to serve as a blue pump for the down-conversion process. This blue light at a wavelength of 459.25 nm was cleaned up spatially through a short piece of single-mode fibre and the remaining 4.5 mW were then focussed onto the ppKTP crystal. The quantum dot was excited with 1.2 mW of the 918.5 nm light as described in section 2. Single biexciton photons from the quantum dot and the signal photons from the SPDC source (both at 919.6 nm central wavelength) were sent simultaneously towards a 50:50 beamsplitter to observe the Hong-Ou-Mandel
interference. Single-photon counting avalanche photo-diodes $D_0$, $D_1$, and $D_2$ monitored the idler arm for heralding and the two output ports of the beamsplitter, respectively. A multi-channel event timer recorded the photon detection times with a resolution of 128 ps so that coincidence and time filtering could be implemented in software.

We considered only events where there was a herald photon detected at the heralding detector $D_0$ within the same repetition period of the laser, i.e. 'double' and 'triple' coincidences. We then calculated the time (micro-time) relative to the laser pulse (macro-time), which served as a synchronisation signal via a fast photodiode. This generated a conditioned list of heralded events, which could have detections at only one ('doubles') or both of the detectors $D_1$ and $D_2$ ('triples').

The distributions of micro-times (figure 6) for both detectors $D_1$ and $D_2$ clearly show that the timing and overall event counts are independent of polarisation. They also indicate that there are no significant difference between the distributions of 'doubles' and 'triples', however the responses of $D_1$ and $D_2$ are somewhat different, most likely due to the time discretization in conjunction with slightly different detector signal shape and temporal offset. The fact that the histograms in figures 6(c) and (d) exhibit reduced event counts for parallel polarisations as compared to the orthogonal case is caused by HOM interference; however, the contrast exhibited by the data here is not the contrast of the actual HOM interference because in this case, the second photon could be in any time bin, i.e. no narrow coincidence window is applied to the time difference between $D_1$ and $D_2$.

The heralded macro-time list defined its own ordered series of events from which we created a global pseudo-time axis, by assigning one laser repetition period $T_L$ to the (macro-)time difference between one heralding event sync laser pulse and the next, no matter how far apart they were in real time. We did this to be able to assess the correlation or delayed coincidence of independent events, which serves as a reference value for the HOM interference. To obtain the Hong-Ou-Mandel signals, we then took the differences between the pseudo times of $D_1$ and $D_2$, which of course are real times for actually coincident events, i.e. where both detectors click within the same laser repetition period as the heralding detector ('triples'). We recorded data for 5.5 h for each polarisation configuration, parallel and orthogonal. This actually happened in several blocks of time, between which various parameters of the laser, sources, and filters were checked and adjusted, if necessary.

The resulting correlation signals for orthogonal and parallel polarisations are shown in figure 7(a) on the global pseudo-time axis where they exhibit peaks at integer multiples of $T_L$. These side peaks turn out to be identically high for parallel and orthogonal polarisations within the Poissonian experimental uncertainty. Also, as expected, the central peak for orthogonally polarised photons is half as high as the side peaks because we are looking at a signal produced by two single-photon sources (one heralded) where even for fully distinguishable photons the probability to observe two photons simultaneously is only half of the probability of observing two...
photons at different times. This distinguishable central peak serves as the reference for the actual HOM measurement.

For further analysis, we focus on figure 7(b), which shows the central peaks on an enlarged time axis, again in blue for orthogonal and in red for parallel polarisation. The main result is that the data for parallel polarisation shows a substantially reduced and flattened peak compared to the orthogonal reference peak. Let us first discuss the features of the orthogonal polarisation case. We expect its shape to be governed by the comparably slow quantum dot radiative lifetime $T_1$, with additional broadening by the finite detector time jitter $T_D$, but presumably not significantly by the SPDC photon. The latter had a frequency bandwidth of 7.7 GHz (FWHM) corresponding to a pulse duration of about 95 ps (FWHM). We modelled the shape as a convolution of a double sided exponential with $T_3 = 328(7)$ ps, as obtained from a separate lifetime measurement and fitted the data with a Gaussian detector response $T_D = 240$ ps (FWHM) and a free amplitude parameter. The data appear to exhibit some small oscillations in the wings of the peak, which might stem from the SPDC temporal shape which is not purely Gaussian, but also oscillates weakly in the wings as a result from the spectral filtering with a slit. The green dashed lines in figure 7(b) are model curves with the same parameters as the fit curves but without the detector-induced broadening. For orthogonal polarisation this results in a pure two-sided exponential decay in time.

We used the same set of parameters to model the central peak of the HOM interference data for parallel polarisation shown in red in figure 7(b). The model function now has an interfering component due to the coherent part of the quantum dot photons for which we use the coherence time $T_2 = 216(3)$ ps. As mentioned above, this is likely underestimating the actual coherence, but given the detector broadening, we cannot make any more accurate statements. In addition, this value is equivalent to the one used in $[6]$ and thus helps the comparison. The interfering part results in a sharp dip at the center of the broader peak for the ideal case with no detector time jitter. Finally, in panel (c) we show a histogram of ‘triples’ where we selected data from only the first three time bins after the laser pulse (green shaded area in figures 6(c) and (d)).
Table 1. Comparison of our results with an earlier measurement using only a fibre Bragg grating for filtering instead of the pulse stretcher, and with work by Polyakov et al [6]. ‘Maximum theoretical coalescence’ here is defined as the highest possible result given the different photon wavepacket shape or spectrum, but not including any extra dephasing, impurity or other corrections to the Michelson interference coherence time.

|                      | Fibre Bragg grating | Pulse stretcher | [6] | Unit |
|----------------------|----------------------|-----------------|-----|------|
| Max. theoretical coalescence | 17                   | 36              | 67  | %    |
| Measured raw coalescence | 18(4)               | 39(4)           | 16(3)| %    |
| Time-selected coalescence | 35(9)               | 63(5)           | 61  | %    |
| Time window          | 384                  | 384             | 290 | 140  | ps   |
| Time selection efficiency | 13.3                 | 12.6            | 25  | 10   | %    |
| $2T_1/T_2$           | 3.0                  | 3.0             | 5.7 |      |
| $\Delta T_{\text{SPDC}}$ | 30                   | 7.7             | 0.9 | GHz  |
| $\Delta T_{\text{QD}}$ | 1.2                  | 1.2             | 1.1 | GHz  |

counts in the central peak for orthogonal polarisation and $A_{\parallel}$ the same for parallel polarisation we find $P_C = 0.39(4)$. Using the $T_1$ and $T_2$ values from above, we get a maximum possible value of $P_C^{\text{max}} = 0.36(3)$, which is obviously underestimated because of the underestimated value of $T_2$.

We can improve on the coalescence by additional time filtering in post-processing at the cost of further reduced efficiency. For this purpose, we limit the acceptable detection events to three 128 ps-wide time bins, i.e. 384 ps, for each detector using the earliest photons to arrive after a laser pulse for both detectors. The respective time bins are in the light green shaded area in figures 6(c) and (d) for the two detectors $D_1$ and $D_2$, respectively. The filtering window, however, is limited in its effective sharpness by the finite detector time jitter. Still, it will reject events that are far away from the zero time delay, which are for the quantum dot those that were emitted late and thus incoherently. Effectively, this filtering thus increases the indistinguishability. This procedure results in the histogram of detection time differences shown in figure 7, which happens to only have counts in four discrete detection time differences. While we could expect there to be up to seven different bins with nonzero counts, only four happen to contain any events. For this time-filtered data, the coalescence probability is 0.63(5). Investigating other filtering windows, we found that including only two more time bins brings us almost back to the unfiltered value of the coalescence probability. This postselection has the same effect as a temporal shaping of the QD photons, as presented in [16, 23]. In the case of our quantum dot-SPDC HOM interference, the temporal filtering discards photons that do not overlap in time, on top of removing incoherent photons from the QD luminescence.

5. Discussion and Outlook

To place these results in perspective, we compare them with an earlier measurement using the same setup but with a 30 GHz fibre-Bragg grating instead of the pulse stretcher, and to the results obtained in [6]. There the authors used an actively stabilised Fabry–Pérot cavity for filtering the SPDC photons and a similar quantum dot, but with quasi-resonant higher shell excitation, which introduces more time jitter into the state preparation of the QD. In table 1, we give a comparison of the most relevant parameters in the systems. The maximum theoretical coalescence value we calculated is the highest possible result given the different photon wavepacket shape or spectrum, but without including the extra dephasing of the quantum dot photons, without postselection, i.e., this would be the result for a lifetime-limited QD for the given filters, without temporal postselection. The temporal postselection can go over this value because only photons with a better temporal overlap are considered. In addition, the coherence times extracted from Michelson interferometer measurements underestimate the coalescence, so that even the unfiltered value can be slightly above.

Even with some time filtering via a narrow coincidence window, the coalescence stays far below unity. If we were to use our quantum dot and excitation method with bandwidth-matched Fabry–Perot filtering, we would expect a coalescence fraction of 86% even when taking the dephasing in the quantum dot into account. To get from this value to a high-fidelity quantum interface, several things need to improve. Most importantly, one needs to use quantum dots that emit indistinguishable photons, i.e. with high purity, which has recently been achieved for InAs/GaAs quantum dots [24, 25]. While we assume that appropriate filtering of SPDC will still be possible, this remains costly in terms of efficiency to match the usual quantum dot spectral bandwidth ($\lesssim 1$ GHz). An alternative approach is to pursue intracavity SPDC as discussed in the introduction, but we believe that a better option is to use even shorter lifetime quantum dots such as the GaAs/AlGaAs system, where quantum dots are grown by droplet epitaxy. Early results show radiative lifetimes of less than 250 ps [26, 27] and high indistinguishability without any filtering or shaping.
We are confident that these early experiments like [6] or ours help identify systematic and practical problems with optical quantum interfaces. They show us which improvements are most likely to solve the problems toward a future quantum internet. Single spins in quantum dots may well serve as stationary qubits at least for local storage in quantum repeater nodes and thus with this work we get closer to solving the problem of how to entangle remote quantum dots using high-quality entanglement from a spontaneous parametric down-conversion source.

Acknowledgements

GSS acknowledges support from the PFC@JQI and TH acknowledges support through a JQI postdoctoral fellowship and a DOC-fellowship of the Austrian Academy of Sciences. AP acknowledges funding for part of this work by the Austrian Science Fund (FWF), project no. V-375. This work was funded in part by the European Research Council (ERC), project ‘EnSeNa’ (257531), MP was supported in part by the Austrian Science Fund Doctoral Programme ‘Atoms, Light and Molecules’ project no. W-1259.

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