Time-resolved and antibunching experiments on single quantum dots at 1300 nm

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The ability to manipulate a quantum state, such as the polarization of a single photon, leads to applications that are unachievable in the classical context. One such application is the transmission of unconditionally secure information using quantum key distribution (QKD). A crucial requirement for the implementation of QKD is the efficient generation of single photons in the telecom windows (1310 and 1550 nm), with a small jitter and narrow spectral width. Considerable effort is being invested in the development of suitable sources based on a variety of different systems (amongst others): atoms in high-Q cavities, single molecules, defects in diamond, and parametric down conversion. We investigate the optical properties of single semiconductor quantum dots (QDs). QDs that emit in the spectral range where silicon technology is used have been extensively studied and it has been demonstrated that they can be driven by an optical or electrical signal to generate single photons on request. However, the attenuation in optical fibers below 1270 nm restricts the potential use of these QDs in QKD applications. Working with QDs emitting in the telecom window presents several challenges: first the QD emission has to be redshifted while maintaining a low spatial density—a difficult combination of requirements for conventional epitaxial growth methods. Second, the single photon detection technology for the near infrared is still in its infancy, and noise levels, quantum efficiency, and temporal response are considerably poorer when compared to the single photon detection modules operating below 1000 nm. Suppression of multiphoton probability from QDs at 1300 nm has been demonstrated, but the relatively high density has not allowed a clear identification of the transitions in a single QD. Recently we have demonstrated a technique for achieving ultralow areal densities (1–2 dots/μm²) and large dot sizes for emission in the 1300 nm band. These QDs present several distinct features, as compared to widely studied short-wavelength QDs, such as larger confinement energy, higher strain fields, and consequently different electron-hole wave functions. Time-resolved studies on single exciton transitions not only reveal important information on the dynamics of the electron-hole relaxation into these new QDs, but provide essential data for the design of interferometric setups for QKD.

We have performed both time integrated and time-resolved spectroscopy on single exciton and biexciton transitions in quantum dots at ∼1300 nm. By embedding the QDs in a planar cavity and carefully tuning the performance of two InGaAs avalanche photodiodes (APD), we are able to measure the second order correlation function with integration times comparable to those made in the region of the spectrum where silicon technology is used, thus demonstrating suppression of multiphoton probability.

The low density InAs/GaAs QDs (2 dots/μm²) were grown by molecular beam epitaxy using an ultralow growth rate technique of which a detailed description can be found in Ref. 14. Apertures of 1 μm² in gold masks, realized by electron-beam lithography and lift-off, were used to spatially select single QDs. A pulsed diode laser emitting at 750 nm, with a maximum repetition rate of 80 MHz, was focused down to a 4 μm spot on the sample with a microscope objective (numerical aperture 0.5). The photoluminescence was collected with the same objective and dispersed into a 1 m focal length monochromator equipped with a cooled InGaAs photodiode array detector; the spectral resolution of the setup is better than 30 μeV (∼0.04 nm). The photoluminescence collected by the objective can also be coupled into a single mode fiber and used for time-resolved photoluminescence (PL) measurements or antibunching experiments in a fiber coupled Hanbury-Brown and Twiss (HBT) setup. The
HBT setup uses a fiber coupled bandpass filter [tunable between 1270 and 1310 nm with a full width at half maximum (FWHM) of 0.8 nm] for selecting the excitonic emission and a 1×2 splitter with a 50/50 ratio as a beam splitter (BS). Two InGaAs APDs (idQuantique) detectors operated in Geiger mode are used for single photon detection.

The spectroscopic signature of a single QD is shown in Fig. 1(a) as a function of pulsed excitation power at 80 MHz. At the lowest power density two peaks dominate the spectrum separated by 4.8 nm. The highest energy peak is attributed to the recombination of a positive trion: its large negative binding energy and its appearance at low power densities justifies the assignment.\(^{18,19}\) For higher pump powers new peaks start to appear on the low energy side of the line labeled X at 1293.5 nm. The peak assignment of exciton (X) and biexciton (BX) follows from the power dependence [Fig. 1(b)], as demonstrated previously\(^{14}\) for QDs on the same sample. The linewidths of X and BX are, respectively, 5.5 nm(43 μeV) and 0.8 nm(48 μeV), which are larger than the resolution of the setup. The Gaussian shape of the peaks suggests that the lines are broadened by the spectral jitter due to local electric fields originating from a random distribution of charges around the dot.\(^{20}\) The peaks labeled nX are attributed to the recombination of electron-hole pairs in the ground state of the QD in the presence of charges in the excited states: a comparison of these clean spectral lines with transitions of the same type in QDs emitting in the 900 nm region\(^{21}\) indicates the presence of a larger confinement energy in our QDs.

By coupling the QD photoluminescence into a single mode fiber and tuning the bandpass filter, we measured the lifetime of the exciton and biexciton, shown as continuous lines in Fig. 2(b), for the QD shown in Fig. 1. For these measurements the InGaAs APDs were biased above avalanche threshold for 100 ns at a repetition rate of 1 MHz with a dead time of 10 μs. In Fig. 2(a) we show the raw data from the exciton lifetime measurement superposed with a measurement of the same integration time, but with the pump laser switched off. When an avalanche is generated in the APD the biasing voltage is switched off and for the remaining duration of the gate the APD is disabled. As a consequence, a larger number of events are recorded at the beginning of the gate. This time-correlated single photon counting technique provides a dynamic range of 10\(^2\) on the signal from a single QD, much higher than up-conversion techniques and streak cameras at these wavelengths. The dashed lines in Fig. 2(b) show the least squares fits to the decaying part of the PL for the X and BX after dark noise subtraction. The least squares fits are calculated from the convolution between a two (one) term exponential decay function for the X (BX) and the setup response function (SRF). Due to the mismatch between the APDs spectral response and laser wavelength, the SRF was measured with a sample of GaNInAs quantum wells, emitting at 1300 nm at room temperature, with a lifetime previously measured to be 50 ps,\(^{22}\) which is well below the temporal resolution of the detector (600 ps). The error on the fits is estimated at ±0.2 ns. The delay in the start of the decay of the X emission compared to the BX is evidence of cascade emission and validates the peak assignment [inset of Fig. 2(b)]. The exciton lifetime is characterized by a double exponential decay of 1.1 and 8.6 ns: the fast component derives from the recombination of a bright exciton, while the slower decaying part suggests the presence of a dark exciton state that is repopulating the allowed exciton transition.\(^{20,23}\) A measurement of the temperature dependence of the exponential decay will be required to validate this conclusion. The BX decay time was measured to be 1.0 ns. The ratio of 1.1 between the exciton and biexciton lifetimes is consistent with previous studies on single QDs,\(^{24}\) however, the presence of a dark exciton state and the PL ratio BX/X greater than 1 suggests that the dark state is acting as a nonradiative decay channel depleting the exciton state.

A QD behaves as a nonclassical light source since successive photon emissions are separated by the same time needed for the ground state of the QD to be repopulated by just one electron-hole pair. To quantify the inability of the QD to emit two or more photons at the same time, we measured the second order correlation function for zero time delays. The PL of an X line from a QD is coupled into a single mode fiber and spectrally isolated, using a tunable bandpass filter, before entering a beam splitter (BS): 0.5% of the laser pulses produce a photon at the input of the BS. The calculated extraction efficiency from the microcavity into the objective numerical aperture is ≈10%: we attribute the remaining losses to nonradiative recombination in the dot, coupling losses in the fiber and absorption in the bandpass filter. An APD is placed in each arm of the BS and the output detection signals of the detectors are fed into a correlator that builds a histogram of the time differences between successive signals from each detector. The normalized histogram is
then related to the correlation function. An anticorrelation measurement is extremely sensitive to the signal to noise ratio (SNR), for example, the $g^{(2)}(0)$ of a perfect single photon source measured on noisy detectors, such that the SNR = 1, is equal to 0.75 instead of zero.\textsuperscript{25,26} Our single photon InGaAs APD detectors have at least three orders of magnitude higher noise counts when compared to the equivalent silicon modules. To obtain reasonable SNRs we operate our APDs in Geiger mode with a nominal gate of 2.8 ns which corresponds to an optical active window on the APD of about 300 ps. Although the APD is active for a time significantly smaller than the photon duration, the overall SNR is optimized in these conditions due to a dramatic decrease in the dark count rate. For a pump power of 600 pW and an integration time of 2 h, we recorded an average of 229±19 coincidences at $t \neq 0$, while the counts at $t=0$ represented only 0.38 of the normalized distribution (Fig. 3): this is strong evidence that on average the QD is emitting less than two photons per pulse. The number of events on the detectors were 340 and 376 s$^{-1}$, while the respective dark counts, measured with the laser switched off, were 33 and 25 s$^{-1}$. For a perfect single photon source these SNRs would produce a measured $g^{(2)}(0)=0.14$.\textsuperscript{26} This reduces the $g^{(2)}(0)$ of our single quantum dot source to 0.24. In practice this value should be lower since we underestimate the dark count rate: in the presence of the signal, a higher avalanche rate will increase the after pulse probability and thus the noise as compared to our estimate. The remaining correlation counts are attributed to stray light entering the setup and imperfect filtering. In Fig. 3 we show the dependence of the value of $g^{(2)}$ at $t=0$ as a function of pump power. The increase in the number of uncorrelated photons emitted by the QD is attributed to the emergence of the background which is present in the spectra around the excitation line (Fig. 1).\textsuperscript{27}

The low QD density, emission wavelength, narrow spectral width, low multiphoton probability, and high efficiency demonstrate that our QDs are suitable for QKD applications. Further improvement on the performance is required to achieve secret key exchange over long distances. The adoption of resolution limited optics designed to work at 1300 nm will increase the coupling efficiency in single mode fibers, while resonant pumping in the excited states of the QD and a bandpass filter designed to suite the narrow spectral width of our QDs will improve the suppression of multiphoton probability.

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