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Tunable statistics of multicolor photons emitted from semiconducting quantum dots

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Abstract

We report the intensity auto- and cross-correlation functions of two consecutively emitted photons from an optically excited single semiconductor quantum dot. We show that a quantum dot is not only a source of non-classically correlated monochromatic photons but is also a source of *multicolor* photons with excitation power dependent correlation properties. We found that the emitted photon statistics is evolving, as the excitation power is increased, from a sub-Poissonian statistics, where the photons are temporally antibunched, to a super-Poissonian one, where they are temporally bunched.

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1. Introduction

It has recently been demonstrated that under continuous wave (cw) excitation, a single quantum dot emits antibunched photons obeying a sub-Poissonian statistics [1], while under optical pulse excitation, they emit single photon per each excitation pulse [2–4]. Similar effects were previously observed in optical studies of the luminescence of single atoms and molecules [5,6].

In this work we report on measurements of temporal correlations among multicolor photons emitted from cw optically excited single semiconductor quantum dots (SCQD). We found a tunable intensity correlation among photons emitted at the same, as well as at different wavelengths due to the recombination of excitons from different collective quantum states. In addition, the temporal correlations among the photons emitted by the SCQD change dramatically with the excitation power. While observing single color photons, emitted from SCQD, strong characteristic antibunching correlations are observed at low power excitations. These correlations disappear with the increase of the excitation power and gradually transform into bunching correlations for yet higher excitation power. When observing a pair of two-color photons, emitted consecutively from the SCQD, a strong characteristic bunching

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correlations are observed for very low and very high power excitations. In the intermediate power excitations, one can observe the characteristic antibunching correlations. These observations demonstrate that a multiply populated quantum light source may emit bunched photons, obeying a super-Poisson statistics.

Using classical rate equations model, we quantitatively account for the experimentally measured distribution of the time interval, τ, between consecutively emitted photons. In particular, we explain the changes in the distribution under variable excitation powers, both for photons originating at the same spectral line and at two different spectral lines. We analytically solve a set of coupled rate equations [4,7] describing the conditional probability that a photon is emitted from a collective state of j electron–hole (e–h) pairs (i.e. the jth multiexciton) following a photon emission event from a collective state of $i>j$ e–h pairs.

2. Experimental

The SCQD sample was grown by molecular beam epitaxy of a strained epitaxial layer of InAs on (1 0 0) oriented GaAs substrate. Small islands of In(Ga)As connected by a very thin wetting layer are thus formed in the Stranski–Krastanov growth mode. The vertical and lateral dimensions of the InAs SCQDs were adjusted during growth by the partially covered island growth technique [8]. The sample was not rotated during the growth of the strained layer, resulting in the formation of a gradient in the quantum dots density. Thus, low density areas, in which the average distance between neighboring QDs is larger than our optical spatial resolution, could easily be found on the sample surface.

We use a diffraction limited low temperature confocal optical microscope [9] for the photoluminescence (PL) studies of the single SCQDs. In order to measure the temporal correlation between emitted photon pairs, we constructed a wavelength selective Hanbury–Brown and Twiss (HBT) [10] setup. The setup is described in detail in Ref. [11]. The setup allows us to correlate two photons emitted not only from the same spectral line ("auto-correlations"), but also from different spectral lines ("cross-correlations").

A Ti:Sa laser at photon energy of 1.6 eV excited the sample non-resonantly, populating the SCQDs via diffusion. An optically excited SCQD was located by scanning the sample surface while monitoring the resulted PL spectra using a 0.22 m monochromator followed by a liquid-nitrogen-cooled charge coupled device (CCD) array detector. Once a typical SCQD emission spectrum is observed the scan is terminated and the objective position is optimized above the SCQD.

3. Results and discussion

The PL emission spectra from a single quantum dot for increasing cw excitation powers are shown in Fig. 1. The measured spectra strongly depend on the excitation power because of the shell filling effect and the Coulomb interactions between the carriers [9,12,13]. For the lowest excitation power there is a non-zero probability for one e–h pair

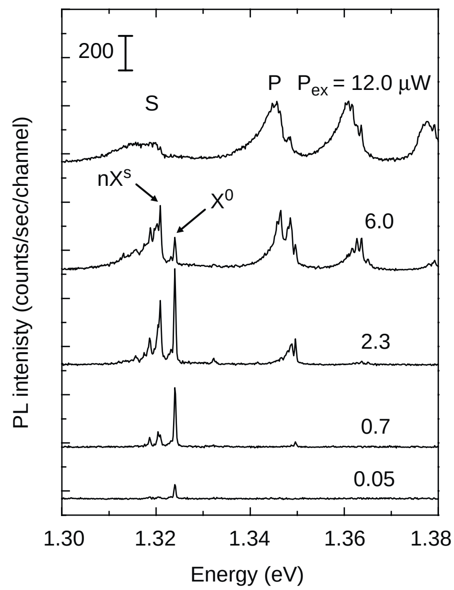

Fig. 1. PL spectra from a cw excited SCQD for various excitation power.

(exciton) to occupy the dot. The radiative recombination of this pair gives rise to a sharp line (denoted as X^0). As the excitation power is increased, the probability to have few e–h pairs (multiexciton) in the dot increases considerably. Since only two carriers with opposite spins can occupy each non-degenerate level, a second level is occupied when there are three or more e–h pairs in the SCQD. The radiative recombination of carriers occupying the 1st and 2nd energy levels, gives rise to the emission of photons in two groups, denoted S and P, respectively, in analogy with atomic shells. The Coulomb exchange interaction reduces the effective band gap of an optically excited SCQD (in a similar way to the well-known bulk phenomenon of band gap renormalization). This effect gives rise to red shifted PL lines [9,13] (denoted, for the S group, by nX^S), when $n>1$ e–h pairs are present in the SCQD during the recombination.

At yet higher excitation powers, the probability to find larger number of e–h pairs within the QD increases, and the probability to find the QD with a small number of e–h pairs decreases, as shown in Fig. 2. As a result, under increasing cw excitation power, the intensity of each emission line increases, reaches a maximum and then decreases as the excitation power is further increased.

Using our wavelength selective HBT setup, we have measured the intensity correlation function between photons emitted at various wavelengths,

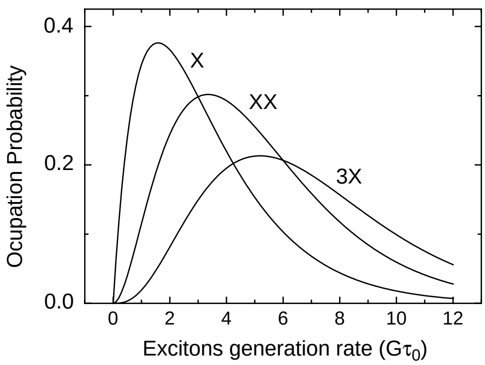

Fig. 2. Calculated occupation probability of single- (X), bi- (XX) and tri-exciton (3X) in photoexcited SCQD, as a function of the exciton generation rate.

due to the recombination of e–h pairs in the presence of various number of spectator pairs. The measured correlation function is expressed as

$$g_{ij}^{(2)}(\tau) = \langle I_i(t)I_j(t+\tau)\rangle / \langle I_i(t)\rangle\langle I_j(t)\rangle, \qquad (1)$$

where $I_k(t)$ is the emission intensity at a wavelength corresponding to the recombination of a kth multiexciton at time t. The function $g_{ij}^{(2)}(\tau)$ represents, therefore, the conditional probability that a photon from a recombination event which involves j e–h pairs will be emitted at time τ after such an emission, which involves i pairs, has previously occurred. Obviously, when the same spectral line is monitored on both channels, $(i=j)$, Eq. (1) simply turns into the second-order temporal coherence function. We note that completely uncorrelated photons have $g^{(2)}(\tau)=1$ (Poisson statistics), photons with a positive correlation ("bunched photons") have $g^{(2)}(\tau)>1$ (super-Poisson statistics), and photons with a negative correlation ("antibunched photons") have $g^{(2)}(\tau)<1$ (sub-Poisson statistics). It is very well established [14] that chaotic and thermal light sources are characterized by $g^{(2)}(0)>1$ and $g^{(2)}(\tau)<g^{(2)}(0)$, while quantum light sources are characterized by $g^{(2)}(0)<1$ and $g^{(2)}(\tau)>g^{(2)}(0)$.

The measured intensity auto-correlation function for the X^0 spectral line ($g_{11}^{(2)}(\tau)$) for an increasing excitation power is presented in Fig. 3a. The figure demonstrates that for a low excitation power the probability of simultaneous detection of two photons is zero (the photons are antibunched [1]). This can be readily understood intuitively, since the recombination of a single e–h pair, which results in the detection of a photon from the X^0 line, empties the SCQD. The probability to detect a subsequent photon immediately after the emission of a same-wavelength photon is then zero. The time it takes for the SCQD to repopulate and emit a second photon with the same wavelength depends on the excitation power and the e–h pair lifetime. Thus, as can be seen in Fig. 3a, this population regeneration time (the width of the antibunching notch) decreases as the excitation intensity is increased. For further increase in the excitation power, the population regeneration time continues to decrease, and as a result the measured $g_{11}^{(2)}(0)$, which is limited by the temporal resolution

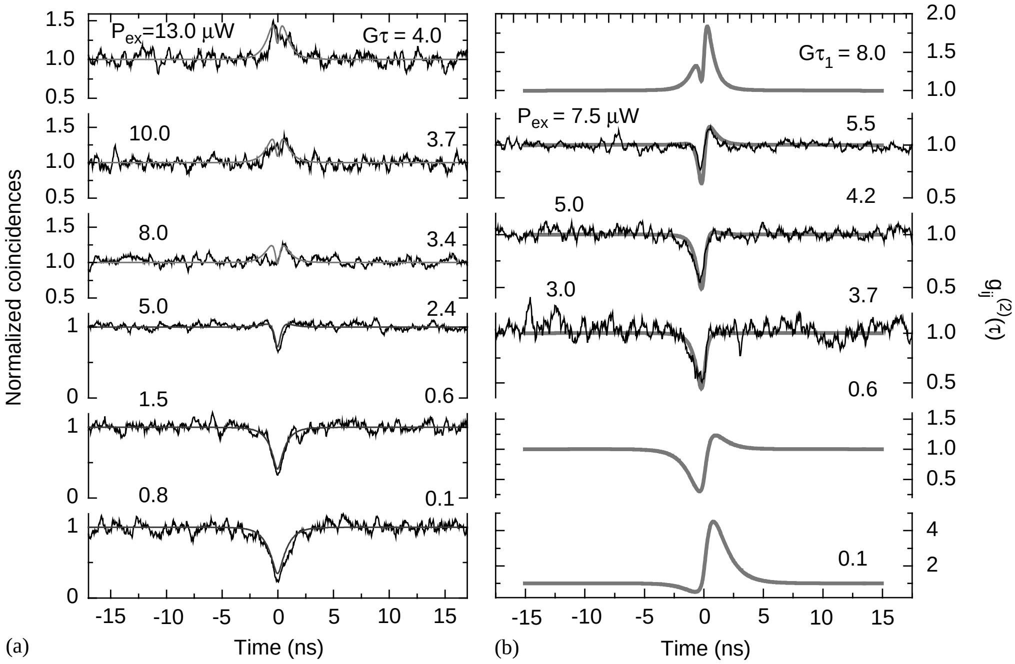

Fig. 3. (a) Measured (thick solid lines) and calculated (thin solid lines, see text) temporal intensity auto-correlation function of the X^0 spectral line for various excitation powers. (b) Measured and calculated temporal intensity cross-correlation function of the X^0 and nX^S spectral lines for various excitation powers.

of our setup, ceases to vanish. With yet further increase of the excitation power, as the X^0 line emission intensity decreases (see Fig. 1), the emitted photons appear to be *bunched*. This novel observation can intuitively be understood as follows. The X^0 line auto-correlation function, $g_{11}^{(2)}(\tau)$, gives the probability that the SCQD is occupied with a single e–h pair at time τ after the emission of previous X^0 photon, which actually left the SCQD empty. At high excitation powers the average number of e–h pairs occupying the dot in steady state is large. Therefore, the average probability for the SCQD to be occupied with only a single pair is small [4]. However, a short time after the SCQD was emptied from pairs (as evident by the detection of the earlier emitted X^0 photon), this probability increases, thus becoming larger than the average one.

From the above discussion it naturally follows that there are also strong temporal correlations between the emission of photons due to recombination of a given collective state of e–h pairs and that from any other collective state. These correlations depend on the dynamics of the SCQD population regeneration. In Fig. 3b we present the measured cross-correlation between the emitted photons from the nX^S line and the photons from the X^0 line for various excitation powers. While the auto-correlation function (Fig. 3a) is symmetric, the cross-correlation function is notably asymmetric. At very low excitation power, the average number of e–h pairs in the SCQD is less then one. A short time after the recombination of one pair, out of n ($n\approx 3$) pairs in the SCQD, has occurred, the probability that a single pair recombines is larger than the product of the average probabilities. Conversely, the probability for the opposite order of events is smaller than the product of the average probabilities. With increasing excitation power, the asymmetry shows that the elapsed time between the emission of an X^0 photon (after which the SCQD is empty) and the emission due to the recombination of one pair out of the collective n pair state, is larger than the reverse order of events. At high excitation power, the cross-correlation function even changes sign. This means that the probability that a single pair recombines in the SCQD, short time after the recombination of one pair out of n ($n \approx 3$) has occurred, is larger than the product of the average probabilities, while the probability for the opposite order of events is smaller than the product of the average probabilities. This peculiar behavior can also be understood intuitively. The average number of e–h pairs in the SCQD at this excitation power is larger than $n \approx 3$. Therefore, the probability that a single pair recombines within the SCQD short time after the detection of recombination from the n pair collective state is larger compared with that probability at long times (the product of average probabilities). While after the SCQD has been found empty, as evident by the detection of the X^0 photon, it takes time for the SCQD to repopulate. During that time no emission from the nX^S line is likely.

We quantitatively model the correlation properties of photons emitted from a single SCQD by the multiexcitonic coupled rate equations used earlier [4,7]. Assuming for simplicity that the radiative recombination of different multiexcitons result in different spectral lines [7], we can write the intensity function $I_i(t)$ in Eq. (1) as $I_i(t) = n_i(t)/\tau_i$, where n_i is the probability for the SCQD to be occupied by the ith multiexciton. The function $I_j(t+\tau)$ in Eq. (1) can now be calculated analytically for each τ, with the appropriate initial conditions. We choose initial conditions such that the radiative decay of the ith *multiexciton* at some time t results in a probability 1 for the SCQD to be populated by the $(i-1)$th multiexciton state at that time. In order to calculate the intensity correlation functions between the various SCQD emission lines, one has to know the multiexciton recombination rates τ_i. We directly measured τ_1 ($\tau_1 \approx 1$ nsec) and used a model to estimate the decay rates of higher multiexcitons [4,7,15]. We note that our solution to the rate equation model is quite robust and that its general behavior is not strongly dependent on the particular τ_i.

The calculated auto- and cross-correlation functions convoluted with the system impulse response function are presented by solid lines overlaid on the experimental data in Fig. 3. General good agreement with the experimentally measured data is obtained by slightly adjusting the exciton generation (G) within the experimental uncertainties, to best fit the experimental data. As can be seen, our calculated correlation functions describe quantitatively the following phenomena:

(a) The crossover from antibunching to bunching for the auto-correlations of monochromatic photons as the excitation power increases.
(b) The asymmetric antibunching–bunching behavior of the cross-correlation function between different wavelength photons and its evolution with excitation power. We note here that while it is easy to calculate the cross-correlation function at very low and very high excitation powers (see Fig. 3b), it is quite difficult to measure it in these extreme limits. In the low limit, the nX^S line is rather weak, whereas in the high limit, the X^0 line nearly vanishes (see Fig. 1).

In summary, we have demonstrated that the statistical properties of photons emitted from a cw excited single semiconductor quantum dot can be variably controlled by the external excitation intensity. For same wavelength photons the quantum dot acts as a quantum light source, although the emitted photons can be either bunched (i.e. super-Poissonian statistics) or antibunched (i.e. sub-Poissonian statistics) in time. We have also measured temporal correlations of photons of different wavelengths, and developed a semiclassical model which quantitatively accounts for the statistical properties observed at various excitation intensities.

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