Semiconductor Quantum Dot: A Quantum Light Source of Multicolor Photons with Tunable Statistics

D. V. Regelman, U. Mizrahi, D. Gershoni, and E. Ehrenfreund
Solid State Institute and Physics Department, Technion–Israel Institute of Technology, Haifa 32000, Israel

W. V. Schoenfeld and P. M. Petroff
Materials Department, University of California, Santa Barbara, California 93106
(Received 11 July 2001; published 30 November 2001)

We investigate the intensity correlation properties of single photons emitted from an optically excited single semiconductor quantum dot. The second order temporal coherence function of the photons emitted at various wavelengths is measured as a function of the excitation power. We show experimentally and theoretically that a quantum dot is not only a source of nonclassically correlated monochromatic photons but is also a source of *multicolor* photons with tunable correlation properties. We found that the emitted photon statistics can be varied by the excitation rate from a sub-Poissonian one, where the photons are temporally antibunched, to super-Poissonian, where they are temporally bunched.

DOI: 10.1103/PhysRevLett.87.257401 PACS numbers: 78.67.Hc, 42.50.Dv, 78.55.Cr, 85.35.Be

Semiconductor quantum dots have been extensively investigated recently as potential, technology-compatible quantum light emitters [1–3]. Such emitters are important for possible future quantum computing [4] and cryptography [5]. It has been recently demonstrated that under continuous wave (cw) excitation, a single quantum dot emits antibunched photons obeying a sub-Poissonian statistics [6], while under optical pulse excitation, they emit a single photon per each excitation pulse [2,3,7]. Similar effects were previously observed also in optical studies of the fluorescence from single atoms and molecules [8,9].

In this Letter, we report on measurements of temporal correlations among multicolor photons emitted from cw optically excited single semiconductor quantum dots (SCQD). We show that there is a tunable intensity correlation among photons emitted at the same and at different wavelengths due to the recombination of excitons from different collaborative quantum states. We show that the temporal correlations among the photons emitted by the SCQD change dramatically with the excitation power. While strong characteristic antibunching correlations are observed for low power excitations, these correlations disappear with the increase in the excitation power and gradually transform into bunching correlations for yet higher excitation power. These observations demonstrate that a multiply populated quantum light source may emit bunched photons, obeying super-Poisson statistics.

We quantitatively account for the experimentally measured distribution of the time interval, τ, between consecutively emitted photons. Specifically, we explain the changes in the distribution under variable excitation powers, both for photons originating from the same spectral line, as well as for photons from two different spectral lines. We do that by analytically solving a set of coupled rate equations [7,10] describing the conditional probability that a photon is emitted from a collective state of j confined 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.

The SCQD sample was grown by molecular beam epitaxy of a strained epitaxial layer of InAs on a (100) 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 [11]. The sample was not rotated during the growth of the strained layer; therefore a gradient in the QDs density was formed and 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 [12] 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) [13] setup (see Fig. 2b inset) including a beam splitter which divides the PL emission from the SCQD into two equal beams. Each beam is then dispersed by a 0.22 m monochromator (MC1 and MC2) and focused into a thermoelectrically cooled, avalanche silicon photodiode (D1 and D2). Each time a photon is detected in one of the detectors, an electronic pulse is produced and transmitted to a time to analog converter. The temporal difference between a pair of pulses is translated into an output pulse of proportional voltage while a multichannel analyzer sorts the pulses and produces histograms of the number of counts vs the time between pairs of detected photons. The temporal resolution of our system is $\simeq$ 250 ps. By tuning the monochromators we can correlate two photons emitted not only from the same spectral line (autocorrelations), but also from different spectral lines (cross correlations).

We used a Ti:Sa laser at 1.6 eV, in order to nonresonantly excite carriers in the sample. The SCQDs are thus

populated by diffusion of the photogenerated carriers into them. We locate an optically excited SCQD 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 array detector. Once a typical SCQD emission spectrum is observed the scan is terminated and the objective position is optimized above the SCQD. 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 [12,14,15]. For the lowest excitation power there is a finite probability for one *e-h* pair (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 find few *e-h* pairs in the dot increases significantly. Since only two carriers with opposite spins can occupy each nondegenerate level, a second level is occupied when there are three or more excitons in the SCQD. The radiative recombination of carriers occupying the first and second 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) and gives rise to redshifted PL lines [12,15] (denoted by nX^S), when $n > 1$ spectator *e-h* pairs are present in the SCQD during the recombination.

At yet higher excitation powers, the probability to find a higher 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 a result, the intensity of any emission line under cw excitation increases, reaches a maximum, and then decreases as the excitation power is further increased.

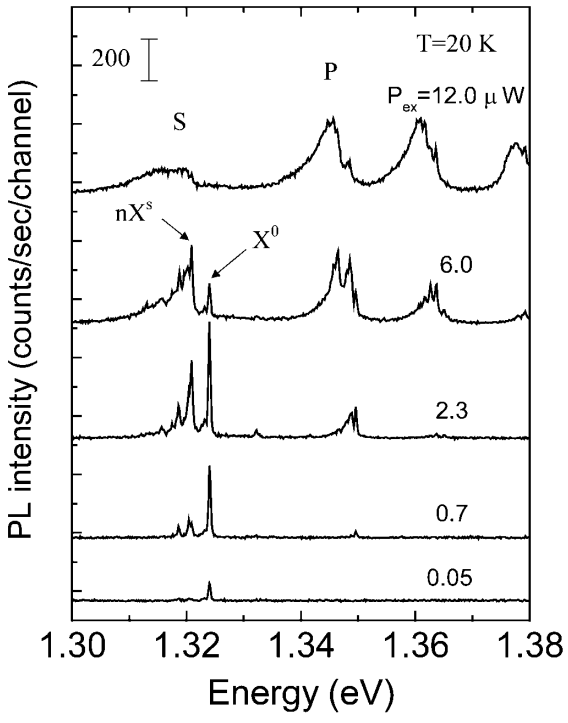

FIG. 1. PL spectra from a cw excited SCQD for various excitation powers.

By using our wavelength selective HBT setup, we measure the intensity correlation function between photons emitted at various wavelengths, due to the recombination of *e-h* pairs in the presence of a variable 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 recombination events 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 positive correlation (bunched photons) have $g^{(2)}(\tau) > 1$ (super-Poisson statistics), and photons with negative correlation (antibunched photons) have $g^{(2)}(\tau) < 1$ (sub-Poisson statistics). It is very well established [16] 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 autocorrelation function for the X^0 spectral line [$g_{11}^{(2)}(\tau)$] for increasing excitation powers is presented in Fig. 2a. The figure demonstrates that for low excitation power the probability of simultaneous detection of two photons is zero (the photons are antibunched [6]). 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 another photon immediately after the detection of the first one is then zero, since the time it takes for the SCQD to repopulate and emit another photon depends on the excitation power and the *e-h* pair lifetime. Thus, as can be seen in Fig. 2a, this population regeneration time (the width of the antibunching notch) decreases as we increase the excitation intensity. For a further increase in the excitation power, the population regeneration time continues to decrease, as a result the measured $g_{11}^{(2)}(0)$, which is limited by the temporal resolution of our setup, ceases to vanish. With yet a further increase of the excitation power, as the X^0 line emission intensity decreases (see Fig. 1a) the emitted photons appear to be *bunched*. This novel observation can intuitively be understood as follows. The X^0 line autocorrelation function $g_{11}^{(2)}(\tau)$ reflects the probability to find the SCQD occupied with a single *e-h* pair at time τ after the emission of the previous X^0 photon, which actually left the SCQD empty. At high

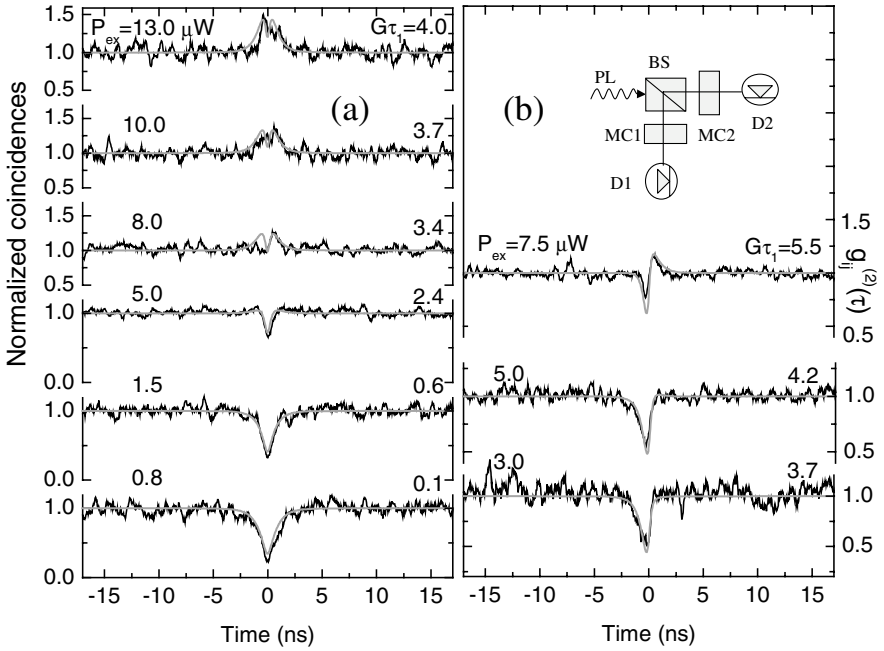

FIG. 2. (a) Measured (thick solid lines) and calculated (thin solid lines; see text) temporal intensity autocorrelation function of the X^0 spectral line for various excitation powers. (b) Measured and calculated temporal intensity cross-correlation function of the X^0 and nX^S spectral lines for various excitation powers. Inset: Schematic description of the modified HBT setup.

excitation powers the average number of e-h pairs occupying the dot in steady state is large. Therefore, the average probability to find the SCQD occupied with only a single pair is small [7]. However, a short time after the SCQD was emptied from pairs (as evident by the detection of the first X^0 photon), this probability increases, becoming thus 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 the 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. 2b 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 autocorrelation function (Fig. 2a) is symmetric, the cross-correlation function is very asymmetric. At low excitation power this asymmetry reflects the fact that the elapsed time between the emission of an X^0 photon (after which the SCQD is empty) and the emission due to recombination of one pair out of the collective n pair state is larger than the reverse order of events. At higher 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 [7,10]. We denote by n_i the probability to find the SCQD occupied with the ith multiexciton and by a vector $\vec{n}(t) = [n_0(t), n_1(t), \ldots, n_N(t)]$ a given multiexcitonic state of the SCQD at time t. Here $n_0(t)$ represents the probability to find the SCQD empty and N is the highest multiexciton with nonvanishing probability. At nonresonant cw excitation the set of coupled rate equations which describes the temporal evolution of the SCQD population [10] can be expressed using a time independent matrix Ψ:

$$\frac{d\vec{n}(t)}{dt} = \Psi\vec{n}(t) = \begin{pmatrix} -(G + \frac{1}{\tau_0}) & \frac{1}{\tau_1} & & & \\ G & -(G + \frac{1}{\tau_1}) & \frac{1}{\tau_2} & & \\ & G & -(G + \frac{1}{\tau_2}) & \frac{1}{\tau_3} & \\ & & & \cdots & \\ & & & G & -\frac{1}{\tau_N} \end{pmatrix} \begin{pmatrix} n_0(t) \\ n_1(t) \\ n_2(t) \\ \cdots \\ n_N(t) \end{pmatrix}. \quad (2)$$

Here τ_i are the ith multiexciton decay times ($\tau_0 = \infty$) and G is the cw e-h pairs photogeneration rate. The steady state solution to Eq. (2) $\Psi\vec{n}(t) = 0$ is given by [7]

$$n_i^{ss} = n_0^{ss} G^i \prod_{j=1}^{i} \tau_j, \quad n_0^{ss} = \left(1 + \sum_{i=1}^{N} G^i \prod_{j=1}^{i} \tau_j\right)^{-1}. \tag{3}$$

A general solution for Eq. (2) is given by [17]

$$\vec{n}(t) = \sum_{k=1}^{N} c_k \vec{\xi}_k e^{r_k t}, \tag{4}$$

where $\vec{\xi}_k$ is the eigenvector corresponding to the eigenvalue r_k of the matrix Ψ, and the constant coefficients c_k are determined by the initial conditions of the problem. Note that because $\det(\Psi) = 0$, $r = 0$ is always one of its eigenvalues. Thus for $t \to \infty$ the solution $\vec{n}(t)$ is given by the steady state solutions Eq. (3). This means that $\langle\vec{n}(t)\rangle = \vec{n}^{ss}$.

Assuming for simplicity that the decay of different multiexcitons results in different spectral lines [10], we have $I_i(t) = n_i(t)/\tau_i$. The function $I_j(t + \tau)$ in Eq. (1) can now be expressed in terms of $n_j^i(\tau)$ which is calculated analytically for each τ by solving Eq. (2) with the appropriate initial conditions: $n_k(t) = \delta_{(i-1),k}$. These initial conditions reflect the important fact that a radiative decay of the ith multiexciton at some time t results in a probability 1 to find the SCQD populated by the $(i - 1)$th multiexciton state at that time. Since $I_j(t + \tau)$ is therefore independent of t, the correlation function $g_{ij}^{(2)}(\tau)$ can be now expressed as

$$g_{ij}^{(2)}(\tau > 0) = n_j^i(\tau)/n_j^{ss}, \qquad g_{ij}^{(2)}(\tau < 0) = g_{ji}^{(2)}(-\tau). \tag{5}$$

For low enough excitation power, when the probability of the dot to be occupied by more than one exciton is negligibly small, we find for the autocorrelation function a relatively simple formula: $g^{(2)}(\tau) = 1 - \exp[-\tau(1/\tau_1 + G)]$ provided that $N = 1$. It is important to note here that it is incorrect to use this last formula for higher excitation levels, where $N > 1$ [6]. In order to calculate the intensity correlation functions between the various SCQD emission lines, one has to know the multiexcitonic 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 [7,10,18]. We note that our solution to Eq. (2) is quite robust and that its general behavior is not strongly dependent on the specific 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. 2. General good agreement with the experimentally measured data is obtained, by slightly adjusting G within the experimental uncertainties to best fit the experimental data. As can be seen our calculated correlation functions describe quantitatively the following phenomena: (i) the crossover from antibunching to bunching for the autocorrelations of monochromatic photons as the excitation power increases and (ii) the asymmetric antibunching-bunching behavior of the cross-correlation function between different wavelength photons and its evolution with excitation power.

In conclusion, 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 color 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 for the first time temporal correlations between photons of different colors and developed a semiclassical model which quantitatively accounts for our observations.

The work was supported by the Israel Science Foundation and by the Israel–U.S. Binational Science Foundation.

[1] P. Michler *et al.,* Nature (London) **406**, 968 (2000).
[2] C. Santori *et al.,* Phys. Rev. Lett. **86**, 1502 (2001).
[3] P. Michler *et al.,* Science **290**, 2282 (2000).
[4] D. Loss and D. P. DiVincenzo, Phys. Rev. A **57**, 120 (1998).
[5] C. H. Bennett and G. Brassard, in *Proceedings of the IEEE International Conference on Computers, Systems, and Signal Processing, Bangalore, India, 1984* (IEEE, New York, 1984), p. 175.
[6] C. Becher *et al.,* Phys. Rev. B **63**, 121312 (2001).
[7] E. Dekel *et al.,* Phys. Rev. B **62**, 11 038 (2000).
[8] B. Lounis and W. E. Moerner, Nature (London) **407**, 491 (2000).
[9] C. Brunel *et al.,* Phys. Rev. Lett. **83**, 2722 (1999).
[10] E. Dekel *et al.,* Phys. Rev. B **61**, 11 009 (2000).
[11] J. M. Garcia *et al.,* Appl. Phys. Lett. **72**, 3172 (1998).
[12] E. Dekel *et al.,* Phys. Rev. Lett. **80**, 4991 (1998).
[13] R. Hanbury-Brown and R. Q. Twiss, Nature (London) **178**, 1447 (1956).
[14] L. Landin *et al.,* Science **280**, 262 (1998).
[15] M. Bayer *et al.,* Nature (London) **405**, 923 (2000); R. J. Warburton *et al.,* Nature (London) **405**, 926 (2000).
[16] R. Loudon, *The Quantum Theory of Light* (Oxford University Press, Oxford, New York, 2000), 3rd ed.
[17] W. E. Boyce and R. C. DiPrima, *Elementary Differential Equations and Boundary Value Problems* (John Wiley, New York, 1969).
[18] E. Dekel *et al.,* Solid State Commun. **117**, 395 (2001).