

Luminescence quenching in InAs quantum dots

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We report how photoluminescence from self-assembled InAs quantum dots depend on pumping power and vertical electric field. The InAs dots, which are embedded in a capacitor-like structure, act as efficient trapping centers for excitons. At a high enough electric field, however, the photoexcited electrons tunnel out of the dots fast enough to quench the emission. For samples with two adjacent layers of vertically aligned dots, we find that the threshold voltage for quenching depends very strongly on the optical pumping power. In total contrast to this, we find no comparable effect for samples grown with a single layer of dots. We explain this in terms of efficient storage of electrons and holes in the double-layer samples. © 2001 American Institute of Physics.

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Self-assembled InAs quantum structures such as dots¹ and rings² have been the focus of considerable recent interest. Their remarkable homogeneity in size and composition over macroscopic scales³ has triggered numerous transport⁴ and optical measurements^{5,6} which, although averaged over millions of dots, do not mask completely the dots' atom-like properties. Application in laser diodes and other optoelectronic devices is anticipated whereas more recent developments show the potential for memory systems.⁷⁻⁹ The charge captured by the quantum structures can be controlled with the electric field in a metal-insulator-semiconductor device. The stored electrons strongly affect the optical properties. For instance, complete occupation of an electron state inhibits the transition to that state through the Pauli principle.⁵ In addition, storage of excitons in InAs double-layer structures⁸ and optical writing and reading of information with a storage time of a few seconds⁹ have been demonstrated. This opens a path towards a new class of optical memories. In this work, we focus on luminescence quenching in InAs quantum structures. Our main goals are to understand the underlying mechanisms and to determine the possible operating range of a storage device based on InAs dots.

We investigated InAs self-assembled quantum dots and rings grown by molecular beam epitaxy in the Stranski-Krastanov mode. Quantum rings are obtained from dots by introducing an interruption in the growth after capping the dots with 1 nm of GaAs.^{2,10} The dots, with densities between 1 billion and 10 billion per cm², are sandwiched between a n^+ -doped GaAs substrate (the backcontact) and a semitransparent NiCr metal gate on the sample surface. A voltage U between the front and backgates controls the electron occupation of the dots or rings. Samples 1 and 2 host a single layer of self-assembled rings and dots respectively, located at

$b = 25$ nm from the backcontact. Sample 3 has two adjacent layers of dots which are located at $b_1 = 25$ and $b_2 = 45$ nm from the backcontact. In all three samples, the distance between the front and the backgate is $d = 175$ nm and a GaAs/AlAs superlattice located between the sample surface and the dots prevents carrier leakage to the NiCr gate. We focused the output of a HeNe laser ($\lambda = 633$ nm) onto the sample surface in a $1 \mu\text{m}$ tight spot in order to excite photoluminescence (PL). We detected the integrated PL with a Ge photodiode placed directly behind the sample using the sample substrate as a filter for the excitation source. Backscattered PL was also collected and spectrally resolved with a 0.25 m focal length grating spectrometer. We used the differential capacitance as a function of U (i.e., C - V profile) to monitor the charging of the dots and rings.^{4,6} All measurements were taken at 4.2 K.

Figure 1 shows the C - V profile of sample 1. Charging of the rings with 1, 2, and 3 electrons is observed around -0.2 , -0.05 , and $+0.05$ V, respectively, followed by a large increase of the capacitance due to the filling of the two-dimensional states of the thin InAs wetting layer. This behavior demonstrates that for $U < -0.4$ V the islands are fully depleted. Figure 1(b) shows the integrated PL from the rings as a function of U . For $U < -0.4$ V, the PL is completely quenched. The quenching of the PL observed here has also been observed in quantum dots¹¹ and can be understood with the help of the inset to Fig. 1(a). At large and negative bias voltages, the electrons of the photoexcited electron-hole pairs can tunnel out of the dots before there is an appreciable chance of recombination occurring. As U is made less negative, the electron tunneling time increases exponentially as a result of the increased barrier thickness. When the tunneling time exceeds the exciton lifetime, PL emission is no longer suppressed. From the integrated PL [Fig. 1(b)] we determine the threshold bias of the PL quenching. The threshold is

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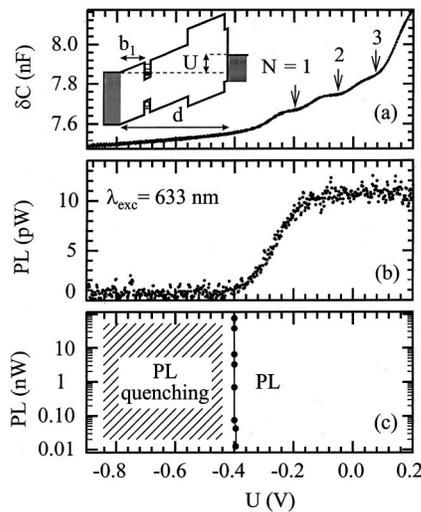


FIG. 1. Inset: energy diagram of a single-layer ring sample where $b_1=25$ nm and $d=175$ nm. The differential capacitance (a), and integrated PL emission (b) as a function of the gate voltage U . The threshold voltage at which the PL is quenched is shown (c) as a function of the illumination power (proportional to the PL emission).

defined to be the voltage where the gradient at the point is located at -0.150 V just slightly more negative than the capacitance peak. We conclude that in the voltage range located between this threshold and the capacitance peak, the excitons are neutral. Above the peak, the excitons are charged. We measured the threshold voltage over four decades of pump power. While the total PL increased linearly with excitation power, Fig. 1(c) shows that the threshold voltage has no dependence on the pump power. This indicates that the local electric field experienced by the rings does not depend on the excitation power. In other words, there is no measurable buildup of an internal electric dipole due to the spatial separation of the photoinduced electron and holes in sample 1. A very similar behavior was found in PL from sample 2.

The results presented so far were collected on ensembles of quantum structures and the inhomogeneities give rise to the broad onset of PL with gate voltage [Fig. 1(b)]. One may wonder how abrupt the transition is for a single quantum island. To answer this question, we examined the emission from a single ring. Although we inevitably detect PL from about 100 rings in our setup, we can isolate a single ring spectrally in the low energy tail of the ensemble emission.¹² Figure 2 shows PL spectra of a single ring as a function of gate voltage illustrating how the PL is quenched in about 20 mV in comparison with 200 mV for the ensemble. For the single ring, the PL intensity near quenching is an exponential function of the gate voltage as shown in Fig. 2. This is consistent with the interpretation of electron tunneling out of the dot through a triangular barrier potential whose width depends linearly on the gate voltage. We have modeled the system within the Wentzel–Kramers–Brillouin approximation and find that the threshold voltage is compatible with very reasonable parameters, for instance a radiative lifetime of about 1 ns and an electron barrier height of a few hundred milli-electron-volts. However, in the absence of a detailed model and also exact information on either the radiative lifetime or the barrier height it is not possible to make a more

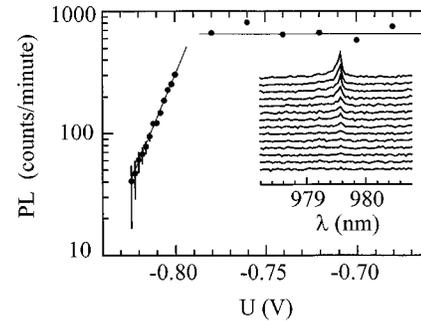


FIG. 2. PL emission of a single quantum ring embedded in a single-layer sample as a function of U . The exponential decrease of the PL intensity (quenching) with negative U is consistent with tunneling of electrons out of the rings.

definitive statement. Similar PL measurements were performed on 24 different individual rings. The threshold of quenching was found to increase linearly with the PL emission energy of the ground state neutral exciton from -0.8 V for 1.26 eV to -0.3 V for 1.36 eV. This is due to the size fluctuation of the confining potential in the quantum ring ensemble. In all measured cases the threshold of quenching is found lower than the first capacitance peak.

The behavior of sample 3, the double-layer structure, is in sharp contrast with that of samples 1 and 2, the single-layer structures. We find that the threshold voltage U_{th} separating the quenched and nonquenched regimes is very dependent on the pump power, shifting to more negative voltages with increasing power (Fig. 3). As for samples 1 and 2 the PL is linearly related to the pump power, but in this case, U_{th} is a logarithmic function of the PL intensity over four decades before a different behavior sets in at high powers, as can be seen in Fig. 3(c). Our interpretation is that the photo-generated electrons and holes thermalize in the lower ($b_1 = 25$ nm) and upper dot ($b_2 = 45$ nm) layers, respectively, by tunneling through the GaAs barrier between them. The carriers are stored in vertically aligned dots, forming a dipole that opposes the applied electric field. With additional stored

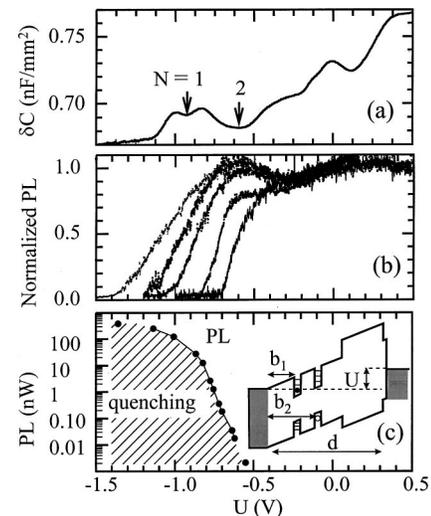


FIG. 3. Inset: energy diagram of a double-layer dot sample $b_1=25$ nm, $b_2=45$ nm, and $d=175$ nm. The differential capacitance (a), and integrated PL emission (b) as a function of the gate voltage U . The illumination voltage of the PL quenching is shown (c) as a function of the illumination power (proportional to the PL emission).

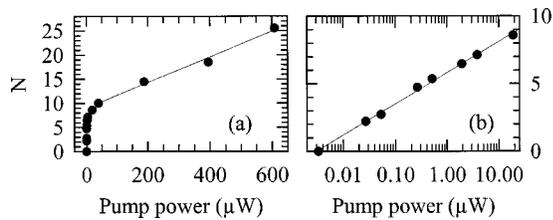


FIG. 4. Number of average elementary charges per dipole as a function of the pumping power (a). The low power part of the data shown in the left panel is plotted on a log scale (b).

carriers, a large negative bias is necessary to reduce the width of the electron tunnel barrier to the back gate in order to quench the PL.

In contrast to samples 1 and 2, the threshold of quenching for sample 3 appears at a more positive voltage than the first capacitance peak. The data at lowest illumination power for instance shows that quenching is possible even though two electrons are present in the lower dot layer. This supports the earlier interpretation that the photogenerated holes tunnel rapidly away from the lower dot layer into the upper layer. The illumination induced shift ΔU in the threshold voltage corresponds to the local dipole field in the barrier region between the dot layers $E = \Delta U/d$. This dipole field is also given by $E = Q_1/(C_1 b_1) + Q_2/(C_2 b_2)$, where C_1 (C_2) is the capacitance between the lower (upper) dot layer and the backgate, and Q_1 (Q_2) is the photostored charge in the lower (upper) dot layer. Equating both expressions for E we obtain the total number of elementary charges per dipole $N = (\Delta U/d)/[\epsilon \epsilon_0/en_s]$, where $n_s \approx 1.2 \times 10^{10}/\text{cm}^2$ is the areal density of dot pairs estimated from capacitance measurements, $\epsilon \epsilon_0 = 12\epsilon_0$ is the local static dielectric constant, and e is the elementary charge. The geometrical information concerning the dots conveniently cancels out of the expression for N .

Figure 4 shows N as a function of the illumination power. The fact that N depends on the excitation power shows that for each power the system reaches a steady state between hole generation and hole leakage. We observe a logarithmic behavior of N on the excitation power [Fig. 4(b)] at low powers over four decades, suggesting that hole leakage occurs through tunneling. Our interpretation is that the holes tunnel out of the dots in the upper dot layer towards the blocking barrier through a quasitriangular potential [see Fig. 1(a)]. At the threshold voltage, the electric field between the dots is always the same. But the threshold voltage becomes more negative with increasing power because of the stored charge's dipole field. This means that the hole tunneling barrier for leakage out of the dots, which is predominantly determined by U alone, decreases linearly with power, causing the hole tunneling time to decrease exponentially. At the highest powers, Fig. 4(a) shows that N has a different dependence on the power. This behavior is not yet fully understood. We speculate that it is related to the filling of the two-dimensional states associated with the upper dot wetting layer once all the dots within an exciton diffusion length of the excitation are full.

A clear result from these experiments is that the double-layer dot samples are much better at storing photoexcited dipoles than their single layered counterpart. Efficient storage may not be possible in a dot in the single-layer samples because of a slight overlap of the wave function of a back-contact electron with the dot. However, it might be possible even in the single layer structures for photogenerated holes to tunnel out of the dots and to be stored at the beginning of the blocking GaAs/AlAs superlattice barrier [inset to Fig. 1(a)]. If this were the case then we would expect a dipole field to buildup opposing the applied field. The data show that this contribution to the dipole field is vanishingly small. The difference with storage in the dots is that there is no atom-like confinement to localize the holes. The holes are allowed to spread laterally under the entire NiCr front gate (about 1 mm^2) and as a consequence the resulting dipole strength is greatly diluted. Conversely, in the case of efficient storage in the dots, the holes and electrons can only travel by about the exciton diffusion length (typically $1 \mu\text{m}$) before become trapped in a dot. This explains the extraordinary sensitivity of the PL quenching voltage on the illumination power for our double-dot samples. In essence, a double-layer sample behaves as a ‘‘photographic plate’’ because storage occurs at the same location as excitation. In principle, it would be possible to pursue this to a level of a single dot by employing an optical instrument with a super-resolution not limited by carrier diffusion, for instance a near-field technique for both excitation and collection.

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