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Coulomb effect inhibiting spontaneous emission in charged quantum dot

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We investigate the emission dynamics of InAs/GaAs quantum dots (QDs) coupled to an InGaAs quantum well in a tunnel injection scheme by means of time-resolved photoluminescence. Under high-power excitation we observe a redshift in the QD emission of the order of 20 meV. The optical transition intensity shows a complex evolution, where an initial plateau phase is followed by an increase in intensity before a single-exponential decay. We attribute this behavior to the Coulomb interactions between the carriers in a charged QD and corroborate the experimental results with both a rate equation model and self-consistent eight-band $\mathbf{k}\cdot\mathbf{p}$ calculations. © *2010 American Institute of Physics*. [doi:10.1063/1.3484143]

Semiconductor quantum dots (QDs) have established themselves as an important new technology in a wide variety of applications, from semiconductor lasers and amplifiers for optoelectronics, to single photon sources and spintronics for quantum computing and cryptography.[1] In particular, the carrier dynamics in these structures is a topic of extensive study due to their significance for device operation. Several theoretical studies have demonstrated the importance of Coulomb effects in QD devices,[2] from the formation of exciton states[3] to their effect on gain spectra.[4] Consequently, as the number of carriers increases, the many-body interactions between the various states leads to complicated carrier dynamics, and can significantly influence the device behavior at high power. In the typical InAs/GaAs QD structures, the large difference in electron and hole effective masses means there are far more hole confined levels within the dot than there are electron levels. Subsequently, under high excitation, a fully filled dot contains many more holes than electrons, with the excess generated electrons occupying bulk states in the surrounding matrix or wetting layer (WL). Clearly, the Coulomb effects in such a situation will have important consequences for the device dynamics.

In this work, we report a strong suppression of the spontaneous emission in an InAs/GaAs QD ensemble caused by Coulomb interactions by investigating a tunnel injection structure under high-power excitation using time-resolved photoluminescence (TRPL). The structure, grown by molecular beam epitaxy, consists of high-quality InAs/GaAs QDs coupled to a 15 nm $In_{0.15}Ga_{0.85}As$ quantum well (QW) through a 4 nm GaAs spacer. The purpose of the well is to create an efficient carrier capture and transfer mechanism, where the electrons and holes can be easily confined by the well and then tunnel through a thin barrier into the dots, thereby increasing both the injection efficiency[5,6] and the modulation bandwidth.[7] A reference structure, containing only the QDs is also investigated. The TRPL experiment was performed at 7 K by exciting the structures using a 780 nm, 300 fs mode-locked Ti:sapphire laser, and by detecting the optical response with a streak camera system. The large incident photon density ($\sim 5\times 10^4$ photons/dot/pulse) creates a much greater number of photogenerated carriers than the number of available states in the dot.

The results are presented in Fig. 1, where we observe an enhancement of the emission from the excited states (ESs) in comparison to the reference structure. This indicates injection of carriers from the well to the dots via the WL, as in a tunnel injection scheme.[8] We can also observe that the spectra undergo several stages in the evolution of the emission dynamics. Initially after the excitation pulse all the radiative recombination channels in the QDs are redshifted with respect to their "neutral" emission wavelengths. The magnitude of this shift for the ground state (GS) is around 5 meV

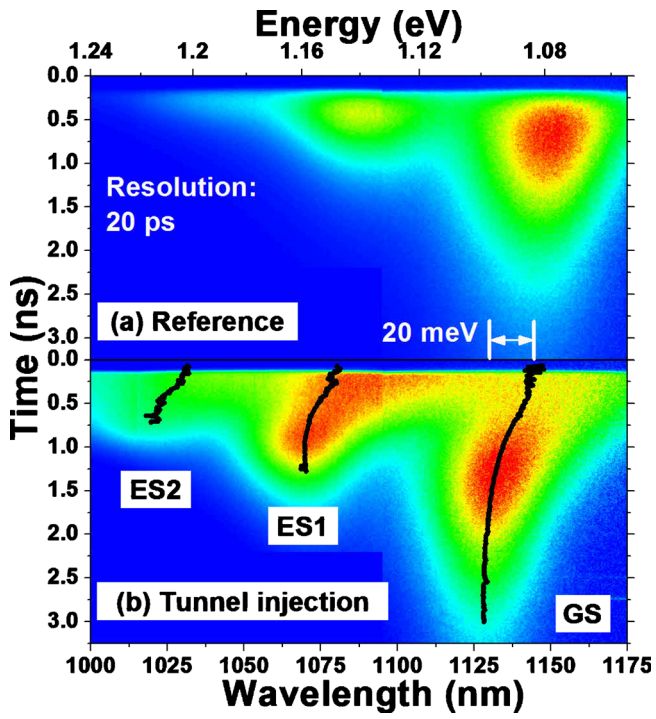


FIG. 1. (Color online) Streak images of (a) InAs/GaAs QDs and (b) QDs with a QW in a tunnel injection scheme.

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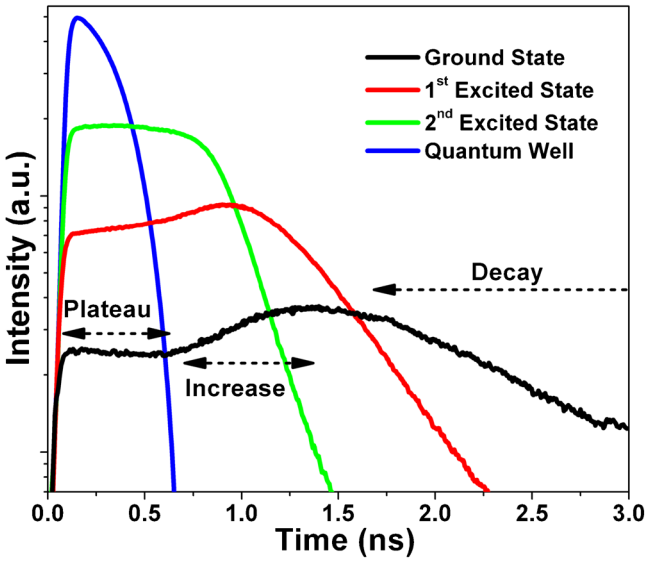


FIG. 2. (Color online) Decay traces of the QD and the injector QW states in the tunnel injection structure.

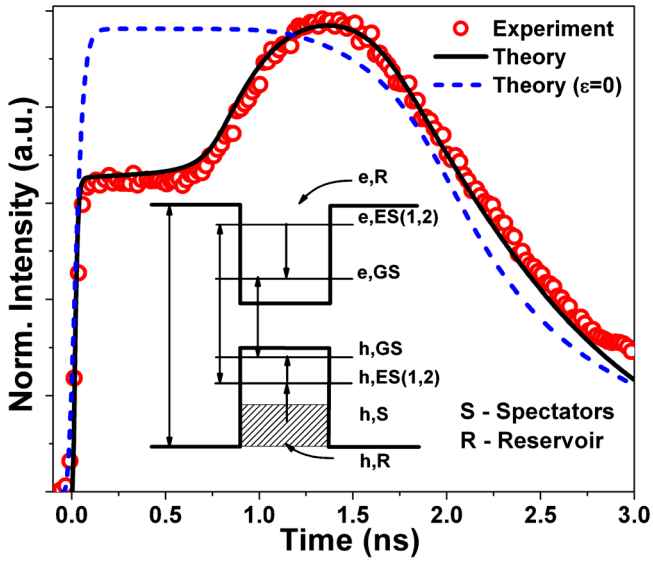


FIG. 3. (Color online) Comparison between the GS intensity and theoretical rate equation calculations in the tunnel injection structure. Dashed line represents the results of the simulation for $\varepsilon=0$.

for the reference structure, while in the tunnel injection structure the magnitude is significantly larger, around 20 meV. The temporal evolution of the emission intensity maxima, as determined by fitting a tri-Gaussian function and indicated in Fig. 1(b) as the black lines, shows that the GS gradually reverts back to its "neutral" transition energy in about 1.3 ns, after which a single-exponential decay occurs. The ES emission is also significantly shifted and for very high charge densities it overlaps with the GS emission, making them difficult to separate. Tracing the emission intensities along the peaks as a function of time, we observe unusual features, depicted in Fig. 2. The GS intensity remains constant for about 450 ps after the excitation pulse, and then it gradually increases for the following 800 ps before decaying exponentially. The first ES behaves in a similar manner with the plateau phase lasting for about 300 ps and the increase for the next 500 ps. The second ES however shows only the constant intensity before the single-exponential decay. The emission energy shift and intensity modification processes are mutually coupled; when the transition exhibits a normal decay phase, no further changes to the peak position occur, as observed under the low-power measurements.[9]

Time-resolved experiments performed on similar tunnel injection structures under low-excitation density (three to four orders of magnitude lower than used in our experiment) showed only an increase in the emission intensity before the exponential decay[10,11] and were interpreted as a slow transfer time between the well and the dots. In our case we observe that the initial rise time of all states is comparable to the temporal resolution of the experiment, which clearly indicates that the transfer between the QW and the QDs is of the order of tens of picoseconds, consistent with Ref. 12 and references therein.

It is consequently clear that, in order for the increase in the emission following the plateau period to occur, it is necessary for there to be a charge-density-dependent change in the recombination rates (i.e., due to Coulomb interactions). We have included this in a rate equation model as follows. We assume that there is a large number of levels in the valence band which do not participate in the radiative recombination but exert a Coulomb influence on the emission probability for the other states. We account for these states collectively using a "spectator level" occupation, $N_{h,S}$, and introduce a parameter ε, which represents the reduction in the spontaneous recombination rate due to the occupancy of the spectator level. The equations are of the following form:

$$\dot{n}_{e(h),\mathrm{GS}} = -\alpha_{\mathrm{GS}}(1-\varepsilon N_{h,S})n_{e,\mathrm{GS}}n_{h,\mathrm{GS}} + \gamma_{e(h),\mathrm{ES1}\to\mathrm{GS}}n_{e(h),\mathrm{ES1}}(1-n_{e(h),\mathrm{GS}}),$$

$$\dot{N}_{h,S} = -\gamma_{h,S\to\mathrm{ES2}}N_{h,S}(1-n_{h,\mathrm{ES2}}) + \gamma_{h,R\to S}N_{h,R}(1-N_{h,S}),$$

$$\dot{N}_{e(h),R} = -\alpha_R N_{e,R}N_{h,R} - \gamma_{e(h),R\to\mathrm{ES2}(S)}N_{e(h),R}(1-n_{e(h),\mathrm{ES2}(S)}),$$

where $n_{e(h),i}$ and $N_{e(h),i}$ represent the time-dependent electron (hole) population of the state i, α_i represents the spontaneous recombination rate of the state i, and $\gamma_{e(h),i\to j}$ characterizes the relaxation rate between constituent states. For convenience, we only explicitly present here the equations for the GS, spectator level (N_S) and the reservoir (N_R). In actuality, we model the system with two ESs, as depicted schematically in the inset of Fig. 3, with similar equations constructed for $n_{e(h),\mathrm{ES1}}$ and $n_{e(h),\mathrm{ES2}}$.

Figure 3 depicts the comparison between the experimental and the calculated evolution of the GS emission intensity. By taking typical values for carrier transfer times (~10 ps) and optical transition timescales (~1 ns) we can obtain an excellent fit. The magnitude of the Coulomb inhibition is calculated to be $\varepsilon=0.156$. When this parameter is set to zero, we only observe the saturation of the emission and no subsequent increase in the intensity (dashed line in Fig. 3). We note that in the model there is no physical meaning applied to the reservoir.

In order to investigate the microscopic origin of this change in transition probability, we have employed theoretical calculations based on a self-consistent 8-band $\mathbf{k}\cdot\mathbf{p}$ model according to the method in Ref. 13 and references therein.

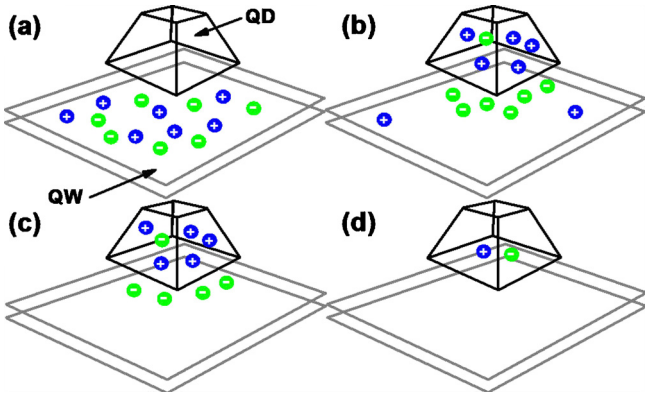


FIG. 4. (Color online) Schematic description of the Coulomb interactions in the tunnel injection structure.

The calculations show that for low charge densities the emission energy and optical matrix element of transitions inside the QD remain largely unchanged. However, when all the electron states in the dot are filled, the increasingly large repulsive forces between the holes confined to the QD inflate their wave functions, while at the same time the electron wave functions undergo a compression. These changes in confinement have different magnitudes, due the different effective masses of the electron and hole, giving rise to a redshift. They also lead to a reduction in the wave function overlap, which for a charge density of 100 particles/dot is equal to 7% for the GS. When all available hole states are filled, the system will have reached an equilibrium and no further modifications will be possible, which would account for the observed emission plateau.

The inclusion of the QW below the QD layer enhances the features via an effect similar to the quantum-confined Stark effect. The whole process is depicted schematically in Fig. 4. The large excitation density creates an abundance of electron-hole pairs, which are captured by the reservoir [Fig. 4(a)] and then quickly transferred to the dot. Due to efficient tunneling processes and to the difference in the carrier effective masses, we achieve a spatial separation of charge, where the majority of the holes are confined to the dot, while the excess electrons remain in the well. Those electrons are attracted to the positively charged dots. This creates a dipole and gives rise to an external (with respect to the dots) field, resulting in lower emission energy. The QD is in equilibrium since all carriers lost due to radiative recombination are replenished from the QW, hence the emission intensity remains constant [Fig. 4(b)]. The reduction in the well population, via tunneling as well as radiative processes, results in a decrease in the external influence, i.e., the emission energy experiences a blueshift. Once the QW is devoid of holes, the QD starts to discharge and, because of the decreasing Coulomb interactions, the emission intensity increases and the wavelength shift is waning [Fig. 4(c)]. Finally, the states of the neutral dot undergo a normal decay [Fig. 4(d)].

In conclusion, we have studied the emission dynamics of InAs/GaAs QDs in a tunnel injection structure under high-power excitation. We have shown that there is an anomalous regime, where the spectrum is redshifted and the emission intensity, following a plateau period, increases before decaying exponentially. This is contrary to a simple explanation of level saturation and long carrier transfer times as reported previously in Refs. 10 and 11, and requires a change in the transition probability. We have utilized a rate equation model and eight-band $\mathbf{k}\cdot\mathbf{p}$ calculations to establish that the reason behind the observed phenomena is a Coulomb interaction between the carriers in a charged QD system. This shows the paramount importance of the band-gap engineering of the tunnel injection structures, where the carrier densities inside the dot are enhanced by an efficient tunneling from the adjacent well and can cause a degradation of device performance. In order to minimize the impact of the Coulomb interactions one needs to limit the number of excess hole levels inside the dot, e.g., by growing a well with a deeper hole confinement potential.

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