Carrier storage and capture dynamics in quantum-dot heterostructures

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Photoluminescence and time-resolved photoluminescence measurements of charge tunable quantum-dot heterostructures reveal that by appropriate biasing of the device, about 90% of photogenerated holes can be stored at an interface near to the nanostructures and subsequently transferred into the nanostructures in a controlled fashion. The capture dynamics are sensitive to the form of the valence band potential in the layer that caps the Stranski–Krastanow dots. The dependence of the capture rate on applied electric field suggests that the valence band confinement potential is ''soft'' in the capping layer, with a spatial extent of around 14 nm. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1577830]

One of the most attractive features of Stranski–Krastanow (SK) quantum dots (QDs) compared with other varieties is the ease with which they can be incorporated into heterostructures to produce lasers, photodetectors, and memory storage devices.[1] More recently, the potential of QD heterostructures for use in spintronic and quantum information-processing applications has been identified.[2] For all such applications, detailed understanding of the dynamics of injected carriers is essential.

The electron and hole ionization energies of the nanostructures are crucial parameters in many device applications, and have been studied in InAs/GaAs SK QDs both by theoretical methods,[3,4] and experimentally by transient capacitance[5–8] and photocurrent spectroscopy[9] techniques. Pettersson *et al.*[10] have measured Poole–Frenkel-style emission from InAs/InP SK dots whereby a ''soft'' confining potential causes the ionization energy to reduce as the applied electric field is increased.[11] Although some evidence of Poole–Frenkel barrier lowering exists in InAs/GaAs SK dots,[12] the effect has yet to be discussed in this system.

We report measurements on InAs/GaAs SK dots in charge-tunable heterostructures grown by molecular-beam epitaxy, which demonstrate storage of photogenerated holes at a heterointerface near the SK layer. The capture rate of these holes into the nanostructures can be controlled by biasing the device, and offers a means of probing the form of the valence band potential in the dot capping layer. We show from the electric field dependence of the hole capture rate that Poole–Frenkel-style barrier lowering is present.

A schematic of the charge-tunable heterostructures is shown in Fig. 1(a). The SK layer is grown onto a 25-nm GaAs spacer layer which separates it from an n^+ contact region. The nanostructures are capped with 30 nm of GaAs, followed by an AlAs/GaAs superlattice that blocks carrier injection from the NiCr Schottky contact. The interface at which photogenerated holes collect is between the capping layer and the superlattice and is labeled ''A'' in the figure.

Data from two samples are included in this report. Sample 1 contains a layer of quantum rings[13] ($\lambda_{PL}=950$ nm), and Sample 2 contains a layer of dots ($\lambda_{PL}=1150$ nm). In each case the surface density of the nanostructures is

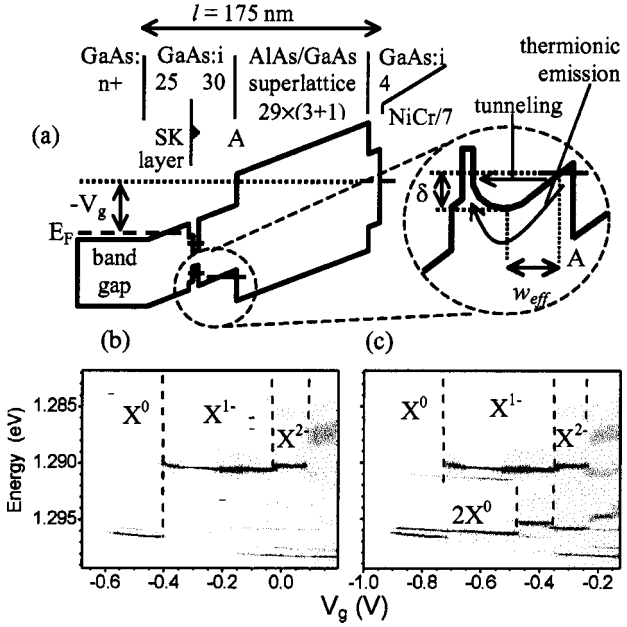

FIG. 1. (a) Schematic potential profile of the charge-tunable heterostructure with layer thicknesses in nanometers. The close-up illustrates how holes stored at interface A can be captured into the nanostructures by tunneling or thermionic emission. The valence band potential barrier reaches a maximum height δ a distance w_{eff} from the mean position of the holes at A. (b) and (c), Grayscale plots of single ring PL vs bias from Sample 1 at 4.2 K, under excitation power of (b) 0.2 kW cm^{-2} and (c) 1.0 kW cm^{-2}. Initial exciton states X^n and $2X^n$ attributed to the most prominent PL lines are labeled. The shift towards reverse bias in plot (b) is a space-charge effect due to holes at interface A in (a).

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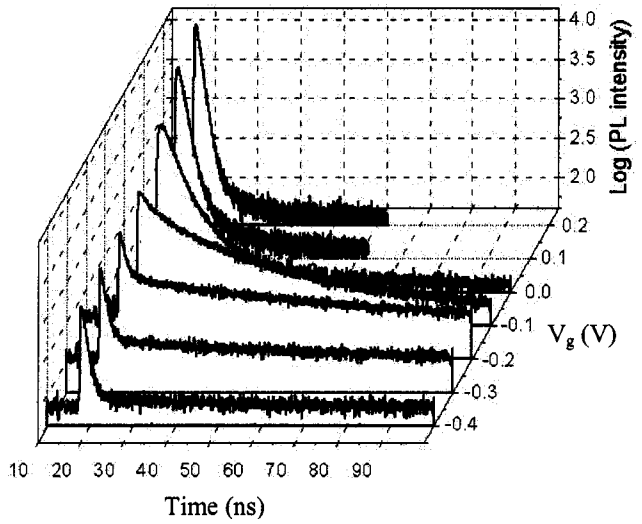

FIG. 2. TRPL from the QD ground state of sample 2 measured as a function of bias at 20 K.

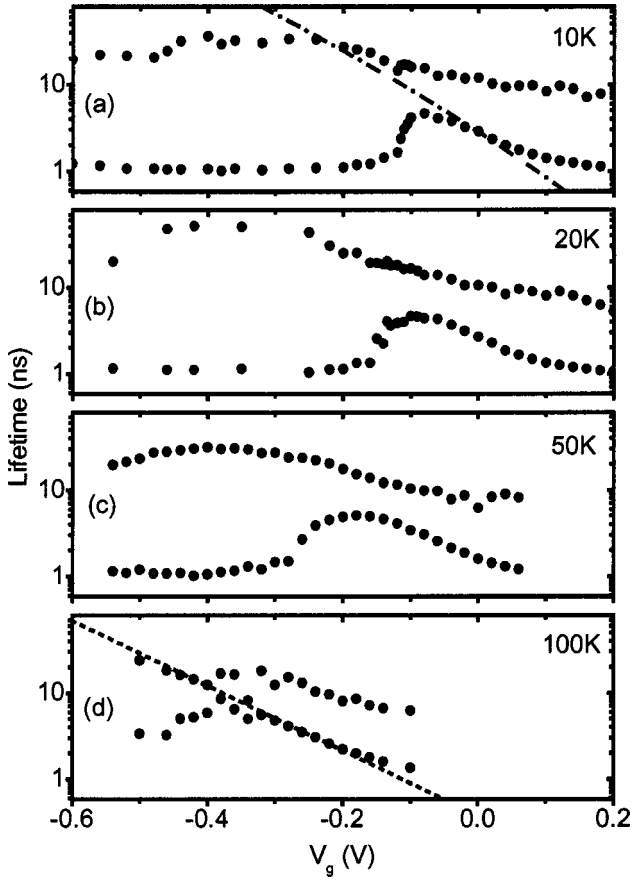

FIG. 3. Fitted TRPL decay lifetimes for sample 2, at temperatures of (a) 10 K, (b) 20 K (c) 50 K, and (d) 100 K. Also shown are bias-dependent lifetimes calculated for the capture process illustrated in Fig. 1, in the limits in which tunneling (plot a) and thermionic emission (plot d) dominate.

about 10^{10} cm^{-2}. By applying a gate voltage V_g across the device, we control the electric field experienced by the nanostructures, and their electron occupation.[14]

Figures 1(b) and 1(c) show single-dot photoluminescence (PL) from Sample 1 excited using 1 and 5 μW (respectively) of 850-nm laser light focused to a 800-nm-diameter spot. In each plot, we observe abrupt jumps in the luminescence energy as electrons are loaded into the nanostructure one by one, and different exciton complexes X^{n-} formed,[14] where n indicates the excess number of electrons. The increase in excitation power has two principal effects: the emergence of extra ''biexciton'' lines labeled $2X^{n-}$, and a clear shift of the entire plot towards reverse bias.

The bias shift between Figs. 1(b) and 1(c) is found to be linear with excitation power, with a coefficient of about -0.16 V cm^2 W^{-1}. A simple one-dimensional Poisson model shows that it can be accounted for by a hole population at interface A of about 4×10^{10} cm$^{-2}/\mu$W of excitation power. In comparison, the average hole density in the rings is estimated [by the approximately equal strength of the X^0 and $2X^0$ lines in Fig. 1(c)] to be 3×10^9 cm$^{-2}/\mu$W of excitation; a factor of 10 lower.

The bias axis of Fig. 1(c) is neither compressed nor elongated compared with Fig. 1(b), indicating that hole generation at A is independent of the applied electric field. We also observe little difference in the bias shift relative to the biexciton intensity at excitation wavelengths of 850 and 633 nm, below and above the bulk GaAs band gap, respectively. These observations are surprising, as they imply that carriers are generated directly in the capping layer even with 850 nm excitation. This combination of circumstances is only possible if the band gap in the GaAs capping layer immediately adjacent to the nanostructures is somewhat lower than that of bulk GaAs, which could be a result of tensile strain passed on from the SK layer,[15] or of indium migration during deposition.

Figure 2 shows how the capture of holes from interface A into the nanostructures in Sample 2 manifests itself in the time-resolved PL (TRPL) data at gate voltages between -0.4 and 0.2 V.[16] At $V_g=-0.4$ V, a single PL decay component with a lifetime of about 1 ns dominates, characteristic of TRPL for QDs when carriers are captured rapidly ($\tau_{cap}\approx50$ ps) from the wetting layer.[17] As the bias is increased, a second decay component with a much longer lifetime emerges. We attribute this component to the onset of capture of holes from A into the dots, as the height of the valence band potential barrier separating them [labeled δ in the close-up in Fig. 1(a)] is reduced. With further bias increase, the second decay component becomes faster and stronger until it first swamps the recombination-limited component, and then becomes recombination-limited itself as the capture lifetime of holes from A drops below 1 ns. The number of counts in the TRPL data at $V_g=0.2$ V is about ten times that recorded at $V_g=-0.4$ V over the same integration time, in agreement with the ratio between hole populations calculated using the single ring bias shift.

The temperature dependence of the bias-dependent TRPL data is revealing as to the nature of the capture process. Data recorded at 10, 20, 50, and 100 K were fitted using an analytic function applicable to the weak excitation limit,[17] and two decay lifetimes obtained for each data set (Fig. 3).[18] Note that the lifetimes in Fig. 3(b) were fitted to a superset of the raw data shown in Fig. 2. The key feature of interest is a peak in the shorter lifetime as the holes captured from interface A become the dominant component.

Little difference is observed between the data in Figs. 3(a) and 3(b), indicating that tunneling through the δ barrier is the dominant mechanism for hole capture into the dots at

the lowest temperatures. In Figs. 3(c) and 3(d), the features shift towards reverse bias, indicating that the capture rate at a given bias increases with increased temperature, and therefore that thermionic emission over the δ barrier is dominant in this temperature range. We can model the bias dependence of the capture lifetime in these two limiting cases. In the thermionic limit,

$$\frac{\partial \ln(\tau)}{\partial V_g} = \frac{w_{\text{eff}}}{lkT}, \tag{1}$$

where $w_{\text{eff}} = \delta/\xi$ is the effective thickness of the 30-nm GaAs capping layer, ξ is the electric field, and l is the device thickness. w_{eff} is less than the capping layer thickness due to the non-negligible extent of the QD confining potential and the hole wave function at the interface [see Fig. 1(a) close-up]. In the tunneling limit, we use the Wentzel–Kramers–Brillouin approximation,[19] to obtain

$$\ln(\tau) = \frac{4}{3\hbar}\sqrt{\frac{2m^* e w_{\text{eff}}^3 (V_b - V_g)}{l}} + \text{const}, \tag{2}$$

where m^* is the hole effective mass, and V_b is the Schottky barrier height ($\approx$0.6 V).

The best fit to the 100-K data is shown as a dashed line in Fig. 3(d), and is obtained by setting $w_{\text{eff}} = 14$ nm in Eq. (1). The same value of w_{eff}, along with $m^* = 0.4 m_e$, inserted into Eq. (2) provides a good fit to the low-temperature data, and is shown as a dash-dot line in Fig. 3(a). This low value of w_{eff} provides further evidence that the nanostructure confinement potential reaches well into the capping layer. The extent of the hole wavefunction at interface A is estimated to be $\sim$2 nm, implying that the band gap in the capping layer remains less than the GaAs bulk value up to $(30-14-2=)$ 14 nm away from the SK layer. TRPL data from sample 1, not shown, reveal very similar features to those highlighted in Figs. 2 and 3.

In summary, we have used single dot photoluminescence and ensemble time-resolved photoluminescence to demonstrate bias-controlled hole storage and capture in charge-tunable heterostructures. Measuring the capture lifetime as functions of bias and temperature suggest that a Poole–Frenkel-style barrier lowering occurs in the capping layer, which will considerably affect the ionization energies of such nanostructures.

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