Storage of electrons and holes in self-assembled InAs quantum dots

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We report spectroscopic measurements of charge-tunable quantum dots. The samples contain vertically aligned double dots which we can fill with electrons from a back contact. We show how we can also accumulate holes in the dots by illuminating the samples with below band gap radiation when a large negative bias is applied. We argue that this is possible through a large disparity in the electron and hole tunneling times. Interband spectroscopy reveals a strong reduction in the quantization energy for the dots in the second layer. © *1999 American Institute of Physics.* [S0003-6951(99)00813-X]

Self-assembled quantum dots have already received much attention for possible applications in laser diodes. However, it might also be possible to use the large confinement potentials of self-assembled quantum dots to store charge and therefore to function as memory devices. We have already shown how an ensemble of dots can be loaded controllably with electrons.[1] This is achieved by embedding the dots in a field-effect structure, so that the dots' charge can be set simply by an external voltage. In fact, the dots are sufficiently homogeneous that Coulomb blockade can be observed on a large ensemble of dots.[2,3] Two groups[4,5] have recently created metastable populations of carriers in InAs quantum dots by positioning the dots close to a two-dimensional electron gas and by illuminating. The existence of the carriers was inferred indirectly by monitoring the transport properties of the two-dimensional electron gas. We report here how we can store holes in a double quantum dot structure, verifying their existence by spectroscopy measurements on the dots themselves.

It is known that dots grown in separate but closely spaced layers ($<$20 nm) are vertically correlated.[6,7] We have embedded a double dot sequence in a field-effect structure and find that we can accumulate holes in the dots in the second layer by applying a negative bias and illuminating with near infrared radiation. Both the present structure and the structure used in Ref. 4 can be considered as primitive optical memory devices. A light pulse creates spatially localized positive charge. At a later time, a light pulse can be reemitted by injecting electrons which recombine with the stored holes. This emission has not yet been demonstrated experimentally, however.

The InAs dots were grown on GaAs in the Stranski-Krastanov mode, with dot densities in the range 10^9-10^{10} cm^{-2}.[8] Two dot layers were grown, separated by 20 nm of GaAs, such that the reflection high energy electron diffraction (RHEED) patterns were as similar as possible. Transmission electron microscopy (TEM) characterization has verified that the dots in the second layer are vertically aligned with dots in the first. The double dot structure is embedded between a back contact (n^+ GaAs) and a gate on the sample surface.[9] Electrons are able to tunnel from the back contact into the dots, but a GaAs/AlAs superlattice between the dots and the gate prohibits transport to the surface. The first dot layer is 250 Å away from the back contact, and the distance between back contact and surface is 1750 Å. A sketch of the structure is shown in the inset to Fig. 1. At large and negative gate voltages, the dots in both layers are unoccupied. On applying more positive voltages, the dots in the first layer load with electrons, and at higher voltages still, the dots in the second layer also load. We can monitor this by measuring the capacitance between back contact and gate, as shown in Fig. 1 for the sample at 4.2 K. We observe Coulomb blockade for tunneling into the first state of the dots in the first layer (the s_1 state) around -1 V, then a broad feature corresponding to tunneling into the excited state of the same dots (the p_1 state). The dots can store a total of six electrons each and so for the present structure, the

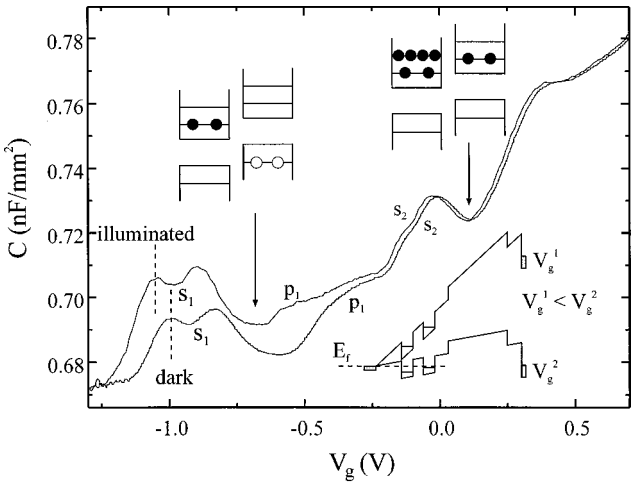

FIG. 1. The inset illustrates the band profile of the device; the main figure shows capacitance against gate voltage for the double dot sample, both with and without illumination, taken at 4.2 K. The illumination was broadband for photon energies $<$1.4 eV with a total power of $\sim\mu$W. The icons show the occupation states at two voltages with illumination.

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dots in the first layer load almost completely before the dots in the second layer start to fill. At ~ -0.1 V, Coulomb blockade for tunneling into the first state of the dots in the second layer (the s_2 state) can be just resolved.

In order to investigate the effects of light on the sample, we positioned an optical fiber above the gate (1.5 mm diameter) on the sample surface. We focused the broadband output of a Fourier transform spectrometer with halogen lamp source into the fiber. A room temperature GaAs filter was placed in the beam so that the light impinging on the sample has an energy well below the low temperature GaAs band gap. Electron-hole pairs are then excited both in the dots and in the wetting layers. It should be understood that the light represents an extremely small perturbation as the total power arriving at the sample is only $\sim\mu$W from which only a small fraction is absorbed.

Figure 1 also contains the capacitance trace with illumination of the entire gate area. It can be seen that the peaks corresponding to tunneling of electrons into the first dot layer are shifted to lower voltages. Our interpretation is that the dots in the second layer are occupied by holes. The presence of this positive charge allows electron tunneling into the dots in the first layer to occur at a more negative voltage. From the shift in voltage we can estimate how many holes we have on average per dot. We estimate the Coulomb interaction between an electron in a dot in the first layer and a hole in a dot in the second layer by assuming point charges separated by 20 nm, including the effects of the image charges in the back contact. The gate voltage is converted into an energy simply with the lever arm.[2,3] We find 2.1 ± 0.2 holes per dot. At higher gate voltage, the light has no significant influence on the capacitance. The voltage at which the two curves in Fig. 1 come together is exactly the voltage at which the dots in the second layer begin to fill with electrons. The explanation is that the electrons tunnel into the dots in the second layer and compensate the holes, so removing the influence of the light.

We have used transmission spectroscopy to confirm that the valence band *s* states in the dots in the second layer are occupied with holes at large negative V_g. Plotted in Fig. 2 are the transmission spectra at various gate voltages. In each case we have taken a reference spectrum at a large and positive voltage when the dots are fully occupied and all the dot transitions are blocked through the Pauli principle.[10] If the valence band *s* states of the dots in the second layer are occupied each with two holes then one would expect that transitions from this state are prohibited. This is exactly what we observe. The lowest energy feature in Fig. 2 corresponds to a transition between a hole *s* state and an electron *s* state (labeled as $s-s$) yet it disappears at -0.4 V when only the dots in the first layer are fully occupied. It represents therefore a transition in the first dot layer, s_1-s_1, i.e., there is no contribution from the transition in the second layer, s_2-s_2. Furthermore, the integrated intensity is proportional to the dot density;[10] the density we infer from the absorption at, say, -1.3 V is 0.8×10^{10} cm^{-2} which is approximately the density of the first dot layer, 1.2×10^{10} cm^{-2} which we deduce from the capacitance curve in Fig. 1. Both these facts imply that the s_2-s_2 transition is prohibited for $V_g<-0.2$ V. Interestingly, around $V_g=-0.21$ V a new absorption feature appears. This is the s_2-s_2 transition, allowed in a small range of gate voltage. We have argued that for $V_g<-0.2$ V the s_2-s_2 transition is blocked by the holes. For $V_g>0$ V, the s_2 states are filled with electrons which also block the transition. However, in between the dots are filled with on average one electron and so the transition is partially allowed. It is at present unclear why the s_2-s_2 (and also the p_2-p_2) absorption is considerably weaker than the s_1-s_1 absorption.

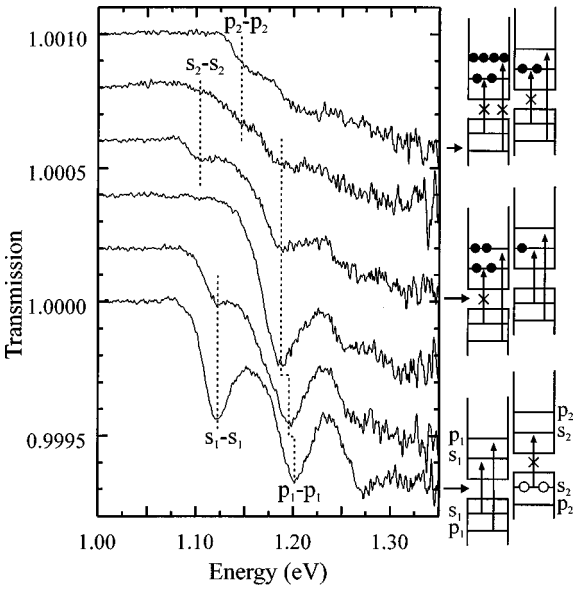

FIG. 2. Transmission against photon energy at gate voltages -1.3, -0.95, -0.63, -0.21, -0.10, and 0.11 V and at 4.2 K. The curves are offset from 1 for clarity.

At large and negative V_g single layer dot samples grown under identical conditions show very weak photoluminescence.[11] The electron-hole pairs are ionized by the electric field before recombination can take place. This mechanism is important for electrons in the double dot layer samples measured here. The tunneling time for the holes out of the dots, however, can be expected to be much larger because the effective mass is a factor of ~5 larger. This large lifetime enables us to accumulate holes despite the tiny absorption of the sample (see Fig. 2) and the weak light source. In principle, holes could also accumulate in a sample with a single dot layer, but this does not occur with our sample parameters. This may be because of a sufficiently large overlap of the hole wave function with the back contact wave functions, enabling the holes to recombine with electrons in the back contact. In fact, we have recently performed similar experiments on InAs/InP dots where single layer samples do illustrate the effects associated with hole accumulation.[12]

According to this picture, holes accumulate and a steady state is reached where the generation rate of holes is equal to the tunneling rate. To estimate the tunneling rate, we integrate the relative transmission at -1.3 V weighted with the intensity distribution from our source to give a very rough estimate of the tunneling time of ~1 s. Consistent with this is an estimation of the tunneling time from the Wentzel–Kramers–Brillouin (WKB) approximation. For realistic hole barrier heights of 0.15–0.2 eV we obtain tunneling times between 1 ms and 100 s (taking the effective mass to be $0.34m_o$). This illustrates the enormous sensitivity of the tun-

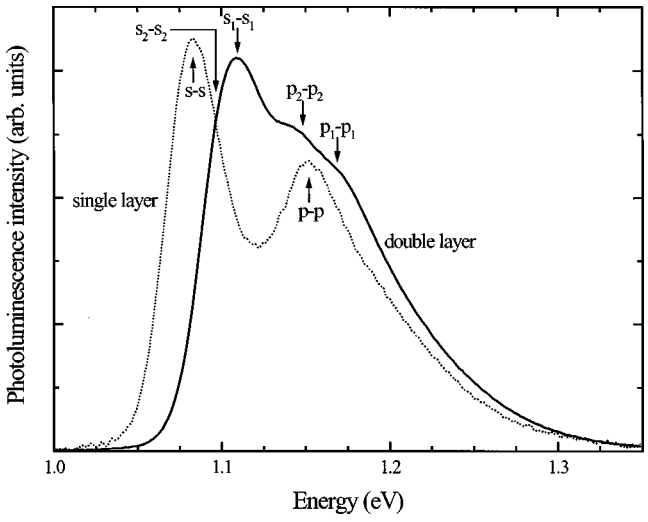

FIG. 3. Photoluminescence, excited with $\sim 8\ \mathrm{W\,cm^{-2}}$ of 514 nm laser radiation, showing the $s-s$ and $p-p$ transitions from the first and from the second dot layers. The sample was at 4.2 K. For comparison, the photoluminescence of a single layer sample is also shown (dotted line).

neling time on the barrier height, and it is clearly necessary to measure the tunneling time directly.

At $V_g \sim 0.1$ V, the first layer dots are fully occupied with electrons so that all interband absorption from the first layer is blocked, whereas the dots in the second layer are only partially occupied. This enables us to pick out the p_2-p_2 transition (see Fig. 2), which appears at a lower energy than p_1-p_1. The photoluminescence spectrum of Fig. 3 shows a split $p-p$ transition, supporting this assignment.[13] The separation between the $p-p$ and $s-s$ energies is essentially the sum of the electron and hole confinement energies. The electron confinement energies have been measured separately with far infrared spectroscopy;[9] using these values we can construct the band diagram as shown in Fig. 4. The vertical quantization energies are approximately the same for the two layers, yet the lateral quantization energies are significantly smaller in the second dot layer. Assuming that the In composition is similar for the two layers, the implication is that the dots in the second layer are significantly larger in the plane than those in the first. Microscopy measurements would appear to support this statement.[6]

In conclusion, we report a series of experiments on charge-tunable, vertically coherent InAs quantum dots. We can load the dots with electrons by applying a voltage between a back contact and gate electrode. We show how we

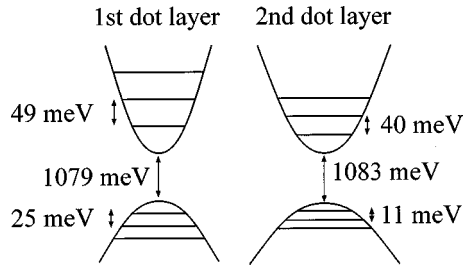

FIG. 4. A band diagram for the double dot structure deduced from the transmission experiments.

can also accumulate a steady state population of holes at large and negative bias with below band gap illumination despite the dots' very small absorption coefficient. Interband spectroscopy shows how the quantization energy in the second layer of dots is smaller than in the first layer.

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