Probing the band structure of InAs/GaAs quantum dots by capacitance-voltage and photoluminescence spectroscopy

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The band structure of self-assembled InAs quantum dots, embedded in a GaAs matrix, is probed with capacitance-voltage spectroscopy and photoluminescence (PL) spectroscopy. The electron energy levels in the quantum dots with respect to the electron ground state of the wetting layer (WL) are determined from the capacitance-voltage measurements with a linear lever arm approximation. In the region where the linear lever arm approximation is not valid anymore (after the charging of the WL), the energetic distance from the electron ground state of the WL to the GaAs conduction band edge can be indirectly inferred from a numerical simulation of the conduction band under different gate voltages. In combination with PL measurements, the complete energy band diagram of the quantum dot sample is extracted. © *2008 American Institute of Physics*.
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Recently, much effort has been devoted to studying self-assembled nanostructures, such as quantum dots (QDs) and quantum wires, due to their potential application in devices.[1,2] For improved device performance, it is very important to know the electronic structure of the QDs and the energy band diagram of QD samples. Much attention has been paid to theoretically resolving the electronic structure of QDs.[3–5] Experimentally, optical and transport measurements can provide access to the band structure of QDs. Some experimental work has also been done to understand the electronic structure of QDs by using photoluminescence (PL) and PL excitation spectroscopy, photocurrent spectroscopy, capacitance-voltage (*C-V*) spectroscopy, absorption spectroscopy, and related techniques.[6–12] Regarding the use of *C-V* spectroscopy to determine absolute energy scales, it is a problem to determine the Schottky barrier formed on the sample surface, which varies from sample to sample. One way to extract the Schottky barrier is to fabricate several devices with different lever arms or to grow a reference sample in which no QD layer is present.[11,12] In this paper, we demonstrate that the electron energy levels of QDs with respect to the GaAs conduction band edge can be determined through *C-V* measurements in combination with a self-consistent numerical solution of the one dimensional (1D) Schrödinger/Poisson equations. With the additional information provided by PL measurements, the entire band structure of InAs QDs buried in a GaAs matrix can be obtained, taking into account the exciton binding energy of the interband transitions in QDs.

The sample was grown by solid-source molecular beam epitaxy on a semi-insulating (001) GaAs substrate. The active part of the sample was composed of 20 nm heavily Si-doped GaAs (back contact) layer, a 42.5 nm undoped GaAs tunneling barrier layer, 5 ML of InAs for the QD layer, 30 nm GaAs spacer, 54 periods of an AlAs (3 nm)/GaAs (1 nm) superlattice as a blocking layer, and a 10 nm GaAs cap. The n^+-doped GaAs back contact layer was contacted from the sample surface by a AuGe alloy annealed at $T=450°$. Subsequently, a NiCr Schottky gate electrode was prepared. The *C-V* characteristics of the sample were measured by superimposing a DC gate bias (V_g) with a small AC voltage (5 mV rms) of variable frequency (f). In the *C-V* measurements, a standard lock-in technique was used to measure the capacitance signal. Whenever the chemical potential of the back contact (Femi energy level) is in resonance with either the discrete energy levels in the InAs QDs or the InAs wetting layer (WL), or the two-dimensional electron gas (2DEG) formed at the interface between the intrinsic GaAs region and the AlAs/GaAs superlattice, resonant tunneling occurs and the capacitance signal shows characteristic peaks.[8–12] The PL measurements of the sample were carried out under excitation by the 532 nm line of a $Nd:YVO_4$ laser. The luminescence signal was collected by a charge coupled device camera and an InGaAs photodetector. All of the *C-V* and PL measurements were taken at 4.2 K

A schematic profile of the conduction band edge of the sample is shown in Fig. 1(a). The structure parameters l_1, l_2, and l_0 in Fig. 1(a) are the distance from the back contact to the QD layer, 2DEG, and sample surface, respectively. E_1 and E_2 in Fig. 1(a) represent the energetic distance from the Femi energy level to the GaAs conduction band edge at the position of the QD layer and the 2DEG, respectively.

A typical *C-V* spectrum of the sample taken at $f=83$ Hz and the second derivative of the *C-V* trace are shown in Fig. 1(b). Clearly, at least seven charging features can be observed. These charging peaks are related to: loading of the first and second electrons into the *s*-like states in the QDs at -0.62 V (s_1) and -0.43 V (s_2), loading of the third and sixth electrons into the *p*-like states of the QDs at -0.04 (p_1) and 0.4 (p_4), and loading of electrons into the *d*-like states of the QDs, WL and 2DEG, at 0.74 V (d_1), 0.9 V (WL), and 1.8 V (2DEG), respectively.[11–14]

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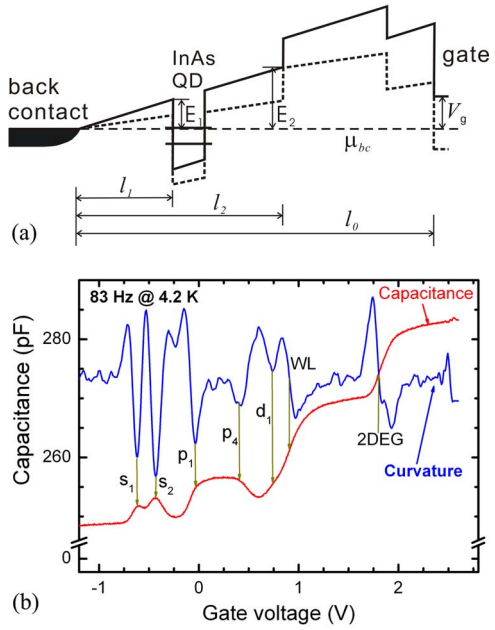

FIG. 1. (Color online) Schematic profile of the conduction band edge (a) and the typical *C-V* spectrum and its second derivative (b) of the sample. The parameters l_0, l_1, and l_2 are 298.5, 42.5, and 72.5 nm here, respectively. The arrows in (b) mark the positions of the charging events.

As for the low QD density ($\sim 8\times 10^9$ cm^{-2}) grown in the sample, it is reasonable to assume that the charge accumulated in the QDs is not sufficient to pin the conduction band. Therefore, the linear lever arm approximation ($\Delta E = e\Delta Vg/\lambda$, with the lever arm $\lambda = l_0/l_1$) can be used up to the onset of the charging of the InAs WL to convert gate voltages into charging energies.[8,14,15] By using this linear lever arm approximation, the energetic distance from the electron ground state of the InAs QDs to the electron ground state of the InAs WL can be extracted to be ≈216 meV. Moreover, from the charging voltage difference between s_1 and s_2, the electron Coulomb blockade energy E_{ss}^C can be determined to be 27 meV. For the energetic distance from s_1 to p_1, a value of 82 meV is found. Theoretically, the energetic difference between loading of the first and third electrons into the QD is estimated to $\Delta E_{(s_1\rightarrow p_1)} = \hbar\omega + \frac{5}{4}E_{ss}^C$.[14] By using the electron Coulomb blockade energy E_{ss}^C obtained above, the result for the quantization energy is $\hbar\omega \approx 48$ meV.

After the charging of WL, there is much accumulated charge in the WL, which pins the conduction band and makes the above linear lever arm approximation invalid. However, the effect of the accumulated charge in the WL on the conduction band of the sample can be simulated by using the free software "1D POISSON/SCHRÖDINGER."[16] In the simulation, a 1.2 nm $In_{0.26}Ga_{0.74}As$ layer was used to act as an InAs WL.[17] The Schottky barrier on the sample surface and the Si-doping concentration in the back contact are assumed to be 0.6 V and 1×10^{18} cm^{-3}, respectively. In order to see the onset of the charging of WL and 2DEG, a series of gate voltages (−0.6–1.4 V) is applied in the simulation. Figure 2 shows the conduction band and the carrier distribution in the sample, which are simulated with different gate voltages. It is observed that loading the electrons into WL and 2DEG occurs at 0.2 and 1.1 V, respectively. The simulated voltage difference (0.9 V) between the charging of the WL and the 2DEG is the same as that determined from the *C-V* measurements, which indicate that the 1.2 nm $In_{0.26}Ga_{0.74}As$ layer works well to simulate the InAs WL.

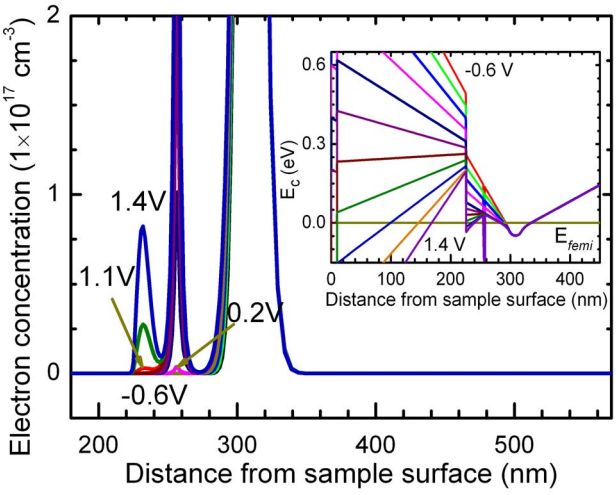

FIG. 2. (Color online) Calculated carrier distribution in the sample structure for different gate voltages. The inset shows the calculated conduction bands.

Figure 3 shows the values of E_1 and E_2, which are simulated under different gate voltages. It should be pointed out that the spatial position of the 2DEG is not exactly at the GaAs/AlGaAs interface but 6 nm removed, which can be seen from the spatial distribution of the carrier concentration in the conduction band simulated under higher gate voltages (1.1–1.4 V). When the gate voltage increases from −0.6 to 1.4 V, both E_1 and E_2 decrease at different rates in the different gate voltage regions (the rates of decrease in different gate voltage regions are determined by sample structure). In the range of −0.6 to 0.2 V, both E_1 and E_2 linearly decrease with increasing gate voltage, with slopes of −0.13 and −0.214 for E_1 and E_2, respectively. These values correspond with the geometric lever arms of l_1/l_0 (0.14) and l_2/l_0 (0.22) well, which indicate that the simulation works very well for the sample structure.[18]

As shown in Fig. 3, the linear lever arm approximation of $E_2/E_1 = l_2/l_1$ is valid up to the charging of the WL. When the ground state of the WL is in resonance with the Femi energy level, E_1 represents the energetic distance from the WL subband edge to the GaAs conduction band edge. However, when the gate voltage is further increased, the lever arm approximation ($E_2/E_1 = l_2/l_1$) cannot be used anymore, which can be seen from the small change of E_1 when the gate voltage increases from 0.2 to 1.1 V in the simulation. On the

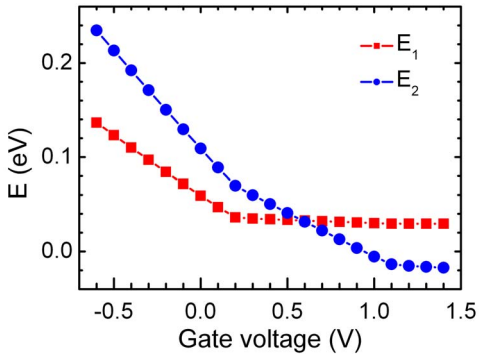

FIG. 3. (Color online) Energies E_1 and E_2 as indicated in Fig. 1(a), calculated for different gate voltages.

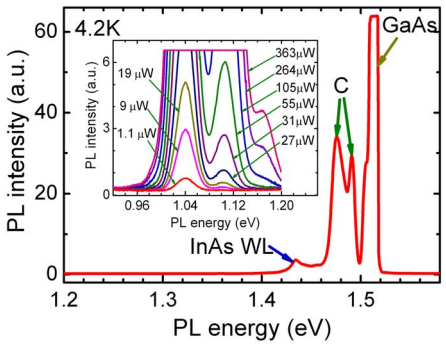

FIG. 4. (Color online) PL spectra of the sample taken at 4.2 K. The inset shows the excitation-power dependent PL spectra of the quantum dots.

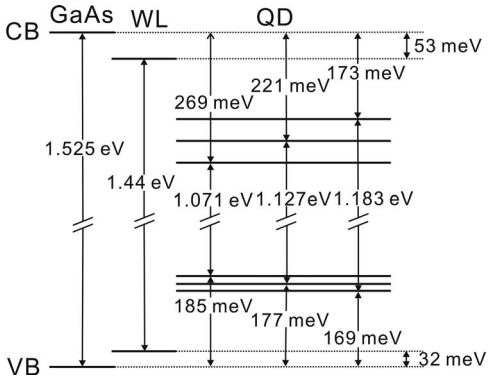

FIG. 5. Energy diagram of the sample determined from *C-V* and PL spectra.

other hand, it is observed that when the gate voltage increases from 0.2 to 1.1 V, E_2 also linearly decreases with increasing gate voltage. The slope of $E_2(V_g)$ in the range of 0.2–1.1 V is −0.1, which is also determined by sample structure, and can be used to derive E_2 when the WL is just charged in the *C-V* measurement $[E_2(\text{WL charging}) = (1.8-0.9)\times 0.1 = 90\ \text{meV}]$. Then, by using the lever arm $E_2/E_1 = l_2/l_1$, the energetic distance from the electron ground state of WL to GaAs conduction band edge in the *C-V* measurements can be extracted to be 53 meV with an assumption that the 2DEG is at the GaAs/AlGaAs interface.

Figure 4 shows the PL spectra of the sample taken at 4.2 K. Clearly, five emission peaks can be observed, which are centered at 1.041,1.107, 1.167, 1.43, and 1.52 eV, and are attributed to the interband transition of the ground state (*s* shell), the first (*p* shell) and second (*d* shell) excited states in QDs, and the interband transition in the InAs WL and the GaAs matrix, respectively. For interband transitions in QDs, it is important to take into account the exciton binding energy, which is of the same order as E_{ss}^c. By using the model described by Warburton *et al.*,[14] the exciton binding energies for the interband transition of the ground state, the first, and second excited states in the QDs are estimated to be 30, 20, and 16 meV, respectively, based on the E_{ss}^c and the $\hbar\omega$ values obtained above and the measured PL energies. Compared to those in QDs, the exciton binding energies for the interband transitions in the WL and the GaAs matrix are much smaller, namely, around 10 and 5 meV, respectively.[19] Combined to the electron energy levels of the QDs and the WL in conduction band determined above, the hole energy levels of the QDs and the WL in the valence band can also be inferred from the interband transition energies. This leads to the determination of the complete energy band diagram of the QD sample, which is shown in Fig. 5.

In conclusion, *C-V* and PL spectroscopy are demonstrated as a simple yet powerful method to establish the complete energy diagram of QDs. By using *C-V* measurements and a 1D Schröding/Poisson simulation, the conduction band energy levels of QDs can be determined. This, combined with PL measurements, makes it possible to construct the entire energy diagram of QDs, taking into account the exciton binding energy of the interband transitions in the QDs.

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