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Electronic structure of self-assembled InAs quantum dots in GaAs matrix

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Capacitance-voltage characteristics have been measured at various frequencies and temperatures for structures containing a sheet of self-assembled InAs quantum dots in both n-GaAs and p-GaAs matrices. Analysis of the capacitance-voltage characteristics shows that the deposition of 1.7 ML of InAs forms quantum dots with electron levels 80 meV below the bottom of the GaAs conduction band and two heavy-hole levels at 100 and 170 meV above the top of the GaAs valence band. The carrier energy levels agree very well with the recombination energies obtained from photoluminescence spectra. © 1998 American Institute of Physics. [S0003-6951(98)01034-1]

In recent years there has been great interest in the properties of heterostructures containing self-assembled quantum dots (QDs).¹⁻⁸ While photoluminescence (PL) spectroscopy is useful for determining transition energies between electron and hole levels in QDs, it gives little information on the position of the quantum levels relative to the band structure of the host matrix. Capacitance-voltage (C-V) spectroscopy may be used to investigate independently the electronic structure of QDs. $^{6-10}$ Using either n- or p-type matrices it is possible to study separately the electron and hole levels in the QDs. In this letter we report combined PL and C-Vstudies of both n- and p-type structures containing an array of self-assembled InAs QDs in a GaAs matrix. We compare our results with recent experimental and theoretical works.

Both samples, *n*- and *p*-type, are based on a type I InAs-GaAs heterostructure grown by molecular beam epitaxy (MBE) on n^+ and semi-insulating GaAs substrates, respectively. The growth temperature was 580 °C, except during the growth of InAs when the temperature was lowered to 480 °C. The growth rates were 1 ML/s for GaAs and 0.066 ML/s for InAs with background arsenic pressure of 1.2 $\times 10^{-5}$ Torr. The average thickness of InAs deposited was 1.7 ML. The QDs were sandwiched between a 0.5-\mu m-thick GaAs cap and a 1-µm-thick GaAs buffer layer which were uniformly doped in the n-type structure (p-type structure) with Si (Be) at a level of $2 \times 10^{16} \text{ cm}^{-3}$ ($5 \times 10^{16} \text{ cm}^{-3}$) except for 10-nm-thick undoped spacers on each side of the QD layer. Au-Cr Schottky barriers were deposited on top of the n-type structure through a shadow mask with holes of 200 μ m diameter. The growth of the p-type structure was terminated with a 0.5- μ m-thick n^+ -GaAs layer to form an asymmetric p-n junction. Mesa structures with diameter of 100 μ m were etched to a depth of 2 μ m. Finally, ohmic contacts were made to both n^+ and p^+ layers.

The C-V characteristics of the devices were measured over a frequency range of 10 kHz to 1 MHz using an HP4275A LCR meter with a 10 mV measuring signal ($V_{\rm osc}$). PL measurements were performed on the as-grown material at 10 K. The optical excitation was provided by the 514.5 nm line of an Ar⁺ laser and the luminescence was dispersed by a 3/4 m monochromator and detected by a cooled Ge diode detector in standard lock-in mode.

Plan-view transmission electron microscopy (TEM) reveals that for both types of sample there is a characteristic size of 10 nm for the QDs which produces a strong contrast with sheet concentration $N_{\rm OD} = (5 \pm 2) \times 10^{10} \ {\rm cm}^{-2}$. Smaller and/or flatter dots, having a weaker black-white contrast (due to a much smaller strain field), have also been observed.³ We considered these dots to be too small to form localized states.

Figures 1(a) and 1(b) show the PL spectra at high excitation power for the n- and p-type samples, respectively. Each spectrum was fitted with two Gaussians. The groundstate emission [peak 1 in Figs. 1(a) and 1(b)] for the *n*-type sample is about 40 meV higher than for the p-type sample. The slight energy difference in the PL peaks for the two samples may be due to the additional annealing required for

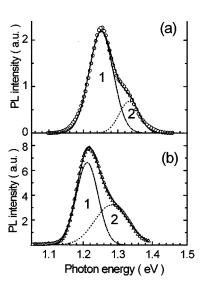


FIG. 1. PL spectra of (a) n-type and (b) p-type QD structures measured at T=10 K for high excitation density of 900 W/cm². The Gaussian fitting parameters of PL spectra are: (a) energy peak $E_{\text{max}}1(2)=1.252$ (1.333) eV, standard deviation $\Delta E1(2) = 0.070 (0.057)$ eV for the *n*-type sample; (b) energy peak $E_{\text{max}}1(2)=1.211$ (1.282) eV, standard deviation $\Delta E1(2)$ = 0.063 (0.098) eV for the *p*-type sample.

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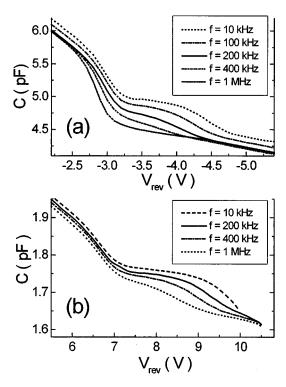


FIG. 2. Frequency dependence of the C-V characteristics of (a) n-type and (b) p-type QD structures measured at T=50 K and T=100 K, respectively.

the growth of the 0.5- μ m-thick n^+ layer (30 min at 580 °C) on the p-type structure. ¹¹ The high energy component of the spectra [peak 2 in Figs. 1(a), 1(b)] disappears at low excitation power (<100 W/cm²), indicating that it is due to the recombination of ground-state electrons with holes in the first excited state. ³⁻⁵ The energy separation of the ground and the first excited hole state is similar (\sim 70 meV) for p-and n-type devices. Theory predicts nearly 100 meV (Ref. 4) and 40 meV (Ref. 5) for QDs having the same transition energy and characteristic size as ours.

Figures 2(a) and 2(b) show the C-V characteristics of the n-type (p-type) device for various measurement frequencies at 50 K (100 K). The step in the C-V characteristic is related to the discharging of the QDs. $^{8-10}$ According to a quasistatic model based on the solution of Poisson's equation, $^{8-10}$ the width of a plateau in C-V depends on the steady-state occupation of the electron (hole) levels in the QDs. For the n-type structure, the charge in the QDs is determined by the sheet concentration $N_{\rm QD}n$ of QDs and the relative positions of the electron level in the QDs (E_e) and chemical potential (μ) in the GaAs matrix. $^{8-10}$ We use a Gaussian distribution with standard deviation ΔE to describe the spread of QD energy levels, and hence the density of electron states in the QD sheet may be written as 9

$$n_e = \int \frac{2N_{\text{QD}n}}{\sqrt{(\pi/2)\Delta E}} \exp\left(\frac{-2(E - E_e)^2}{(\Delta E)^2}\right) f(\mu, T) dE, \quad (1)$$

where $f(\mu, T)$ is the Fermi–Dirac function.

We include the spin degeneracy factor of 2 and neglect the charging energy (\sim 10 meV) required to place a second electron on a QD because this is much less than the spread of electron energies. ΔE is taken from the fits to the PL spectra. $N_{\rm QD}$ $_{n}$ $_{n$

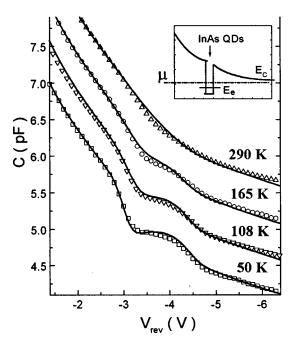


FIG. 3. C-V characteristics of the *n*-type QD structure at different temperatures: experimental data (open symbols) and model simulations (solid lines) with $\Delta E_c = 70$ meV, $N_{\rm QDn} = 3 \times 10^{10}$ cm⁻² and electron confinement energy $E_c = 80$ meV. The frequency is f = 10 kHz for T = 50 K and f = 1 MHz for T > 50 K (the curves are offset by successive 0.5 pF steps for clarity). The insert shows the conduction band diagram of the structure. The horizontal line represents the electron subband energy level in the QDs.

croscope (TEM) studies is an upper limit to the density of active dots since coulombic repulsion effects would tend to reduce the effective value of $N_{\rm OD}$.

Solving Poisson's equation for different values of the reverse bias $V_{\rm rev}$, we obtain the charge in the structure per unit area. 8 The calculated capacitance is derived from C $=\Delta Q/\Delta V$, based on "quasistatic" conditions, i.e., the temporal charge variation ΔQ caused by the increment of the reverse bias ΔV is neglected. However, in practice a small ac signal $V_{\rm osc}$, frequency f, is applied in addition to the dc reverse bias $V_{\rm rev}$. $V_{\rm osc}$ causes a modulation of the charge both at the edge of the space charge region (dQ_{3D}) and at the point where the Fermi level crosses the electron level in the QDs (dQ_{ad}) . Since escape of electrons from the QDs is a slower process than capture,³ to reach equilibrium between the QD layer and adjacent GaAs layers, the thermionic emission rate of electrons (e_n) from the QDs must be much higher than $2\pi f$. The thermionic emission rate depends exponentially both on the temperature and the energy of the QD electron levels. As temperature decreases, e_n becomes lower than the $2\pi f$, i.e., carriers freeze onto the QD levels¹⁰ and the plateau associated with QD discharge is suppressed [Figs. 2(a) and 2(b)]. For our devices in the frequency range 10 kHz-1 MHz, this effect occurs around 50 K for the ntype sample [Fig. 2(a)] and 100 K for the p-type sample [Fig. 2(b)]. This means that the hole states in the InAs QDs are deeper than the electron states with respect to the corresponding band edge of the GaAs matrix.

C-V characteristics recorded at different temperatures are displayed in Figs. 3 and 4, together with theoretical fits. In all cases f was low enough for the system to reach equilibrium.

ensity determined directly from transmission electron mi-By modeling⁸⁻¹⁰ the measured temperature-dependent Downloaded 05 Sep 2003 to 141.108.20.53. Redistribution subject to AIP license or copyright, see http://ojps.aip.org/aplo/aplcr.jsp

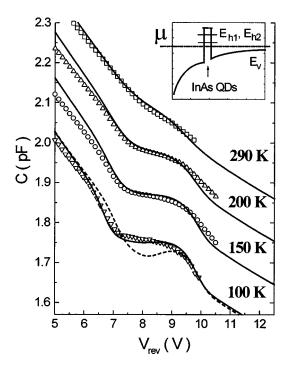


FIG. 4. C-V characteristics of the p-type QD structure at different temperatures: experimental data (open symbols) and model simulations (solid lines) with $\Delta E_{h1} = 60$ meV, $\Delta E_{h2} = 100$ meV, and $N_{\rm QDp} = 4 \times 10^{10}$ cm $^{-2}$. The hole confining energies are $E_{h1} = 170$ meV and $E_{h2} = 100$ meV. The dashed line shows a simulation of the 100 K C-V with a single hole level at $E_{h1} = 170$ meV. The frequency is f = 10 kHz for T = 100 K and f = 1 MHz for T > 100 K (the curves are offset by successive 0.1 pF steps for clarity). The insert shows the valence band diagram of the structure. Two horizontal lines represent the hole subband energy levels in the QDs.

C-V, we find that the electron states in the QDs are centred at E_e = 80 ± 10 meV below the GaAs conduction band (Fig. 3).

The analysis of C-V for the p-type QD structures indicates that there are two filled hole states in InAs QDs: at $E_{h1} = 170 \pm 10 \text{ meV}$ and $E_{h2} = 100 \pm 10 \text{ meV}$ above the top of the GaAs valence band (Fig. 4). For the hole excited state, we used a degeneracy factor of 4 in Eq. (1). Also shown for comparison in Fig. 4 is a simulation of the 100 K C-V with a single hole level at $E_{h1} = 170$ meV. The difference between the two hole states of 70 meV coincides with that between the ground- and excited-state PL peaks. Note that the sum of the ground-state hole energy ($E_{h1} = 170 \text{ meV}$), the electron energy ($E_e = 80 \text{ meV}$) and the ground-state recombination energy $(E_{1peak}=1250 \text{ meV})$ gives a total of 1500 ±20 meV, which is close to the GaAs band gap at 4 K (1519 meV). The small difference may be due to the exciton binding energy which is $\sim 20 \text{ meV.}^4$ The fitted values of the dot densities are respectively $N_{\rm QDn} = 3 \times 10^{10} \ {\rm cm}^{-2}$ and $N_{\rm QDp}$ $=4\times10^{10}$ cm⁻² and agree very well with the values determined by TEM [$(5\pm2)\times10^{10}$ cm⁻²].

In addition, recent midinfrared absorption measurements on InAs/GaAs QD structures showed an electron confinement energy E_e equal to 190 meV and a separation between the ground and the first excited states of the holes equal to 115 meV.¹²

In conclusion, we have investigated both n-GaAs and p-GaAs structures containing self-assembled InAs QDs using the C-V technique, PL spectroscopy, and TEM. Whereas in other recent work, ⁶ the electron binding energy was reported to be larger than that for holes, for our QDs we find the opposite behavior. Our values are more consistent with recent theoretical estimations of the binding energies, ^{4,5} but the difference between our values and those of Refs. 6 and 12 may point to a strong sensitivity of the binding energy on growth conditions which may affect both morphology and composition of the self-organized QDs.

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