Electron-filling modulation reflectance in charged self-assembled $In_xGa_{1-x}As$ quantum dots

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We present some observations of electron-filling modulation reflectance in charged self-assembled $In_xGa_{1-x}As$ quantum dots. This electron-filling modulation reflectance is a different type of electroreflectance, which is based on the Pauli blocking of interband transitions in quantum dots. By adjusting the appropriate ac and dc reverse biases, electron filling in the quantum dots can be modulated. Experimentally determined interband transitions have been compared with those obtained from photoluminescence spectra. The good agreement between these results reveals that at least three quantum-confined electron states are contained in our quantum dots due to their electron-filling character. As the temperature is increased, the relative intensity of each state can directly reflect the electron populations of the quantum states. The technique developed here provides an efficient way to observe the interband transitions of quantum dots. [S0163-1829(99)51028-X]

Self-assembled quantum dots (QD's) have recently been of interest due to the application of this material in optoelectronic devices.[1,2] In particular, the δ-function-like density of states is expected to fabricate temperature independent and low threshold current density QD lasers.[3–5] Self-assembled QD's are grown by molecular beam epitaxy (MBE) in the Stranski-Krastanow (SK) mode,[6,7] which lays one semiconductor ($In_xGa_{1-x}As$) on top of another material (GaAs), whose lattice constant differs from that of the overlayer. Initially, a two-dimensional growth of $In_xGa_{1-x}As$ is laid down, until the film reaches a critical thickness, and three-dimensional QD's are formed on a residual two-dimensional wetting layer (WL). When the dot layer is capped by another semiconductor (GaAs), the structure exhibits rich electrical and optical properties.[8–13] Recently, the interband transitions of QD's have been studied by photoluminescence (PL),[14,15] calorimetric,[9] and transmission spectroscopies,[16] but the QD modulation reflectance spectrum is very weak so far, with only a few works being reported.[17] Because modulation reflectance is a powerful method for studying two-dimensional semiconductor structures, such as quantum wells, superlattices, and high electron mobility transistors, etc.,[18] it is expected to develop an efficient modulation mechanism for the investigation of zero-dimensional QD's. In this paper, we will present a different type of electroreflectance (ER), which is called electron-filling modulation reflectance (EFR), to study the interband transition of charged $In_xGa_{1-x}As$ QD's. The EFR mechanism is based on the Pauli blocking of QD interband transitions.[16] As the electron is occupied in a QD energy level, the interband transition for this level will be blocked. On the other hand, when the electron is evacuated from the level, the interband transition will be allowed. Modulating the electrons in and out of the level, by suitably adjusting the ac and dc bias, will induce a strong change in the reflectance (ΔR) which will enable us to observe the EFR signals ($\Delta R/R$).

The samples used in this study were grown by MBE on an n^+ (001) GaAs substrate. A 300-nm n^+-doped (Si, $5\times10^{18}\,cm^{-3}$) GaAs buffer layer was first grown on the substrate, followed by a 400-nm n-doped (Si, $5\times10^{16}\,cm^{-3}$) and p^+- (Be, $1\times10^{19}\,cm^{-3}$) doped GaAs layers. The self-assembled $In_{0.5}Ga_{0.5}As$ QD's were embedded in an n-type layer at 300 nm below the p^+-n interface. Transmission electron microscopy (TEM) was used to observe the QD's. Both plan and cross-sectional specimens were prepared by using conventional argon ion milling after mechanical thinning and polishing. TEM images showed that our QD's were lens shaped, 3-nm high in the growth direction, with a 15-nm average diameter and a dot density of $1\times10^{11}\,cm^{-2}$. In order to verify the QD's electron-filling character, we grew a referent sample for comparison. The referent sample had the same structure as the dot sample except for the absence of $In_{0.5}Ga_{0.5}As$ QD's and WL, i.e., a GaAs p^+-n structure.

The PL experiments were performed using the 514.5-nm line of an Ar^+ laser as an excitation source. The laser beam was focused on a spot about 100 μm in diameter. The luminescence was dispersed by a 0.5-m monochromator and detected by a cooled Ge detector or a Si photodiode. The EFR was performed by applying a dc reverse bias and ac modulation voltage between the front and back contact electrodes. The front contact was fabricated by evaporating gold film onto a p^+ cap surface through a mask with 1-mm-square apertures. The back contact was formed by the ohmic contact of an n^+ substrate to a copper plate. The ac modulation frequency was about 230 Hz. The EFR experiment was the same as the ER except that the ac and dc voltages were adjusted to move the electrons in and out of the QD's. As shown in Fig. 1(a), if a QD is located at the n-type flat band region, it is charged with electrons. As the reverse bias is applied, the electron will be evacuated from the dot. The modulation between a charged and empty dot will give the EFR spectrum. The modulation voltage in this case is defined as $\Delta V=|V_b|-|V_0|$, where V_b is the reverse bias and V_0 is a constant voltage that defines the initial QD charging state. From the capacitance-voltage (C-V) measurements, shown in Fig. 1(b), we find that the QD's are loaded with

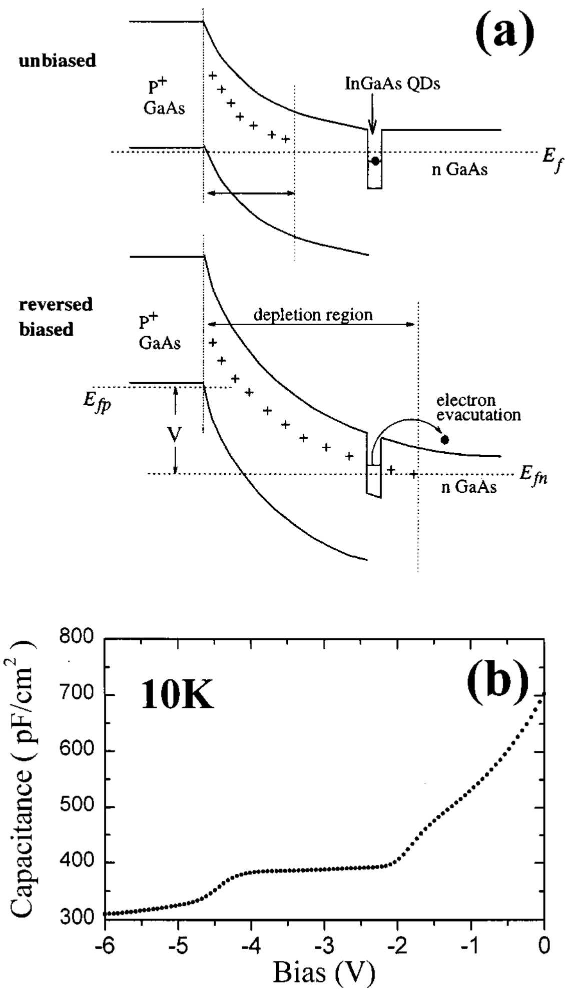

FIG. 1. (a) Schematic diagram of the electron-filling modulation mechanism for quantum dots. (b) Capacitance spectrum of dot samples measured at 10 K.

electrons at zero bias, and start to evacuate electrons at $V_b = -2$ V. Therefore, if we modulate the bias voltage between $V_0=0$ V and $|V_b| \geqslant 2$, the QD's EFR spectra may be observed.

Figure 2 shows the EFR for the QD and referent samples, at 10 K. These spectra were obtained by varying the bias voltage V_b from -1 to -4 V. Spectra for the two samples display similar oscillation signals at photon energies larger than 1.4 eV. These signals indicate the band gap transition of GaAs and their associated Franz-Keldysh oscillations[18] contributed by the electric field near the GaAs p^+-n interfaces. Below 1.4 eV, the QD sample exhibits additional structures for reverse bias $|V_b| \geqslant 2$ V. These extra structures occur when electrons move in and out of the QD. Therefore, they are referred to as signals arising from the QD's. Because the QD's EFR signal is proportional to the number of modulated electrons, its line shape will be a first derivative of the dielectric function with respect to the intensity (or oscillatory strength).[18,19] Fitting the QD line shape with a Gaussian (or Lorentzian) function, the fitting intensity will reflect the number of modulated electrons in the QD ensemble. As shown in Fig. 2, the increase in QD intensities for larger reverse biases indicates an increase in the modulated electrons in the QD ensemble. When we further increased the

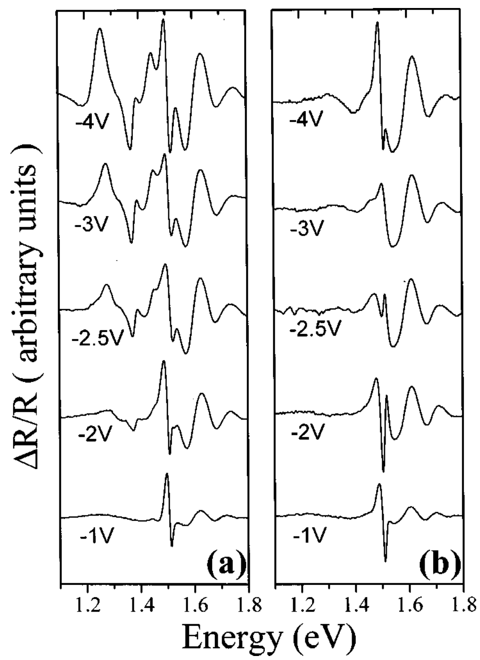

FIG. 2. EFR spectra of (a) QD and (b) referent samples at different reversed biases measured at 10 K.

reverse bias, a saturation of the QD intensity was found at $|V_b| > 4.6$ V. From the C-V measurement [Fig. 1(b)], this intensity saturation reveals that the electrons in our QD ensemble have all been modulated. Therefore, a further increase of reverse bias does not change the QD intensity. This behavior provides more evidence for the electron-filling character of the EFR experiments.

In Fig. 3(a), we compare the EFR spectra with the PL, for the QD sample, at a temperature of 10 K. The PL spectrum shows two peaks at energies between 1.2 eV and 1.4 eV, while the EFR spectrum displays three structures in this energy range. By deconvoluting the PL and EFR spectra with a Gaussian fit we find that the signals near 1.25 eV (E_0) and 1.29 eV (E_1) correspond to the QD's ground state and excited state interband transitions. The structure near 1.38 eV in the EFR is attributed to the interband transition of two-dimensional (2D) $In_{0.5}Ga_{0.5}As$ WL. The WL signal was not observed in the 10-K PL spectrum, but it did appear at higher temperatures ($T \geqslant 50$ K). Using the bias dependent EFR spectra at 10 K, a plot of E_0 and E_1 transition intensities at different biases is shown in Fig. 3(b). For the bias of $-2.8\,\text{V} < V_b \leqslant -2$ V, only the electrons occupied in the E_1 state are modulated and, as a consequence, only the E_1 transition was observed. When $V_b \leqslant -2.6$ V, the Fermi level (E_F) is well below the E_1 states in our QD ensemble, the electrons occupied in the E_1 states have all been modulated and the transition intensity would become saturated. Similarly, the E_0 intensity starts to become significant near $V_b = -2.6$ V and saturated when all the electrons are evacuated at $V_b > -4.6$ V. These results demonstrate that a selective modulation of a particular QD electron state can be achieved by suitably adjusting the bias voltage.

We also measured the QD's EFR at different temperatures, which are shown in Fig. 4. All of these spectra were measured with $V_0 = 0$ V and $V_b = -4.6$ V. Since the electrons in the QD's are fully depleted at $V_b = -4.6$ V, the

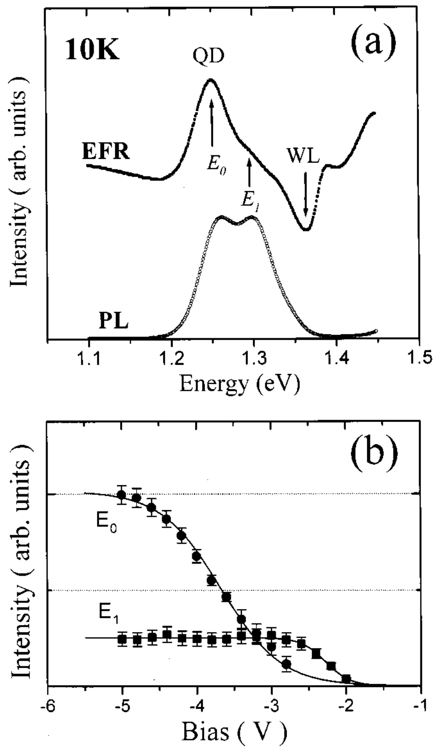

FIG. 3. (a) Comparison of EFR and PL spectra for a QD sample at 10 K. The EFR was measured at $V_0 = 0$ V and $V_b = -4.6$ V. (b) A plot of bias dependent transition intensities of the ground (E_0) and first excited (E_1) state. The lines are guides for the eye.

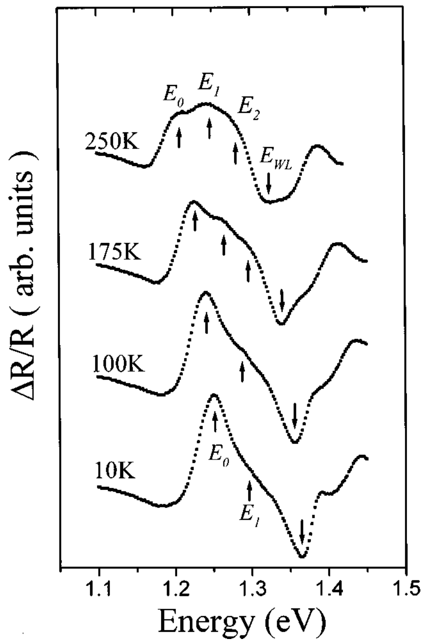

FIG. 4. The EFR spectra of a QD sample at different temperatures. The EFR were measured at $V_0 = 0$ V and $V_b = -4.6$ V.

relative intensity of the EFR directly reflects the electron occupation of each quantum state in the entire QD's ensemble under a zero bias. As indicated by arrows, in Fig. 4, the excited states (E_1 and E_2) become more clearly resolved as the temperature is increased. The most striking feature in Fig. 4 is the temperature dependence of the relative intensity for each quantum state. The ground state intensity is decreased, accomplishing an increase in excited states at higher temperatures. Since only the E_0 and E_1 transitions are observed at 10 K, the E_F is near the energy level of E_1. In other words, the twofold degenerate E_0 states are filled with two electrons and the E_1 states may be partially empty. As can be inferred in Fig. 3(b), the ratio of the saturated intensity of the E_1 and E_0 states is about $\frac{1}{4}$. It means that the average number of electrons in the QD's is about 2–3 electrons per dot. As the temperature is increased, the population of electrons in the E_0 states will be decreased, while the excited states will be thermally occupied due to the Fermi-Dirac distribution. Therefore, the temperature dependence of the EFR intensity reflects the thermal distribution of electron populations in QD's states. The electron filling in QD's states is determined by the density-of-state (DOS) and the Fermi-Dirac distribution function. It should be balanced by the positive charges in the space-charge-region (SCR) forming around the QD's plane, and also determined by the E_F and the doping concentration in the GaAs bulk. The theoretical description of the temperature dependent electron populations in EFR is similar to the calculation of electron distributions in QD's C-V experiments.[21] We assume the QD's electronic DOS as multiple Gaussian functions and the electronic confinement energy of each QD state was treated as a parameter. Once the electronic confinement energies were specified, the number of electrons accumulated in the QD layer can be uniquely determined from the E_F and conduction band bending due to the SCR forming around the QD's plane. We choose the QD's confinement energies so that, at 10 K, the average occupation is about 2–3 electrons per dot. The electron occupation in each state at different temperatures was then calculated and compared with the EFR intensities. From this calculation, we find that the calculated electron occupation of each state is consistent with our EFR intensities at different temperatures. It indicates that the EFR can be a direct tool for observing the temperature dependence of electron populations in QD's states.

We analyzed the transition energies of these EFR spectra with a Gaussian line shape. The fitting results for the interband transition energies are listed in Table I, and are compared with those obtained from the PL spectra. In the PL experiments, information is lacking about whether the transition occurs due to the same electron state to several hole states,[20] or just between different electron and hole states with the same quantum number.[8] Therefore, a different theoretical approach will lead to a conflicting assignment of these luminescence peaks. From Table I, good agreement is found between EFR and PL in the determination of QD and WL interband transitions. Due to the electron-filling character of the EFR spectrum, we can state that these optical transitions, as observed in our EFR and PL experiments, are contributed by different electron states. In other words, our QD's contain at least three quantum-confined electron states. These results are also consistent with electron-addition capacitance spectroscopy,[10] transmission spectroscopy,[16] and theoretical predictions.[8]

In conclusion, we have presented the observation of EFR in charged $In_{0.5}Ga_{0.5}As$ self-assembled QD's. EFR is a different type of ER, which is based on the Pauli-blocking mechanism for interband transitions. EFR experiments were performed by adjusting the modulation voltage to control

TABLE I. Comparison of the ground state (E_0), first excited state (E_1), and second excited state (E_2), QD's interband transition energies and the interband transition energies of WL (E_{WL}) between the EFR and PL.

Temperature	EFR (eV)				PL (eV)			
	E_0	E_1	E_2	E_{WL}	E_0	E_1	E_2	E_{WL}
10 K	1.247	1.289		1.369	1.253	1.300	1.338	
100 K	1.239	1.282		1.361	1.240	1.287	1.330	1.364
175 K	1.222	1.262	1.301	1.342	1.218	1.260	1.296	1.341
250 K	1.203	1.240	1.276	1.325	1.198	1.238	1.273	1.315

electron filling in the QD's. From these experiments, we found that the EFR signal was rather strong, comparable to the band gap signal of the GaAs buffer layer. Therefore, the EFR technique developed here provides a simple and direct method for studying the interband transition of charged QD's. Experimentally determined interband transitions have been compared to those obtained from the PL spectra. The good agreement between these two results indicates that at least three quantum-confined electron states are contained in our QD's, due to the electron-filling character of the EFR. From the temperature dependent relative intensity of each state, we found that the EFR can be a direct tool for observing the temperature dependence of electron populations in QD's states.

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