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Photo-capacitance response of internal tunnelling coupling in quantum-dot-imbedded heterostructures under selective photo-excitation

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Abstract
Under selective photo-excitation, the capacitance response of internal tunnelling coupling in quantum-dots-imbedded heterostructures is studied to clarify the electronic states and the number densities of electrons filling in the quantum dots (QDs). The random nature for both optical transitions and the filling in a QD assembly makes highly resolved capacitance peaks appear in the C–V characteristic after turning off the photo-excitation.

(Some figures in this article are in colour only in the electronic version)

In recent years, with continuously increasing interest in both optical and electronic properties of self-assembled quantum dots (SAQDs), various experimental techniques, such as optical spectroscopy [1–6], tunnelling spectroscopy [7–10] and capacitance spectroscopy [11–14], have been employed to characterize the electronic states of SAQDs. Due to the charging or discharging of the SAQD energy level in SAQD-imbedded heterostructures, a pronounced capacitance plateau usually shows up in the C–V characteristics [5, 12, 13]. To clarify the level structures of SAQDs, however, a sophisticated calculation involving some self-consistent solution of the Poisson equation seems to be unavoidable as a result of the space-charge effect in the structure [15, 16]. On the other hand, the electron tunnelling between the three dimensional (3D) emitter and the SAQD plane, imbedded in a metal–insulator–semiconductor field effect transistor (MESFET) or in a single tunnelling barrier or in the central quantum well (QW) of a double-barrier-resonant-tunnelling diode (DBRTD), has revealed valuable knowledge about the atom-like shell structures, the lifting of the orbit and spin degeneracy and even the two-dimensional (2D) spatial images of the probability density of a single electron in an SAQD [7–10].

In the present work, we study the specific capacitance response induced by internal tunnelling occurring in an SAQD-imbedded heterostructure under selective photo-excitation. Using the relations between the bias of the capacitance peak and the energy position of the electronic state which participates in the tunnelling process, the level structures in the QW or QDs can be extracted. In addition, the number densities of electrons in the QDs can also be derived from the photo-capacitance spectra.

Samples (A and B) used were grown by molecular beam epitaxy in the following growth sequence. For sample A, a 500 nm n$^+$-GaAs buffer layer, doped by 1×10^{18} cm^{-3} Si, was first deposited on an n$^+$-GaAs substrate, and then covered by a 10 nm-thick GaAs spacer. The successive growth of a three-barrier-two-well heterostructure consists of a 10 nm-thick AlAs barrier, a $In_{0.17}Ga_{0.83}As$ QW composed by 1 nm GaAs/5 nm $In_{0.17}Ga_{0.83}As$/1 nm GaAs, a 3 nm-thick AlAs barrier, a 6 nm-thick GaAs QW and a 10 nm-thick AlAs barrier. On the top, a 10 nm-thick GaAs spacer and a 100 nm-thick n$^+$-GaAs cap layer were grown. Sample B has the same layer structure as sample A, except that a 6 ML $In_{0.5}Ga_{0.5}As$ SAQD layer is inserted in the middle of the second GaAs QW. The conduction band diagrams of sample A and B at the bias of zero are shown in the insets of figures 2 and 3 respectively. The sample was patterned into a mesa of 370 μm × 660 μm. For the back contact, indium was deposited and alloyed. The top ohmic contact was made by evaporating and alloying AuGaNi/Au with an optical window of 200 μm × 200 μm. Photoluminescence (PL) spectra were measured with an optical excitation of the 488 nm line of an Ar$^+$ laser, and the PL signals were detected by a cooled InGaAs detector. An HP4284 LCR meter (with modulation amplitude of 5 mV and modulation frequency of 10 kHz) was used to measure the photo-capacitance spectra under the illumination from a wavelength-tunable Ti–sapphire laser.

Figure 1 shows the PL spectra measured in sample A (panel (a)) and sample B (panel (b)) at 12 K. In addition to the peak at 1.51 eV from bulk GaAs, sample A exhibits two sharp peaks at 1.425 and 1.61 eV, which can be apparently assigned to the e1–h1 transitions of $In_{0.17}Ga_{0.83}As$ QW and GaAs QW, respectively. For sample B, the PL peak from the $In_{0.17}Ga_{0.83}As$ QW still appears at 1.425 eV, and a new broad peak at 1.35 eV, showing up the depletion of the 1.61 eV PL of GaAs QW, stems unambiguously from the photo-emission of the QD assembly in the GaAs QW.

In figure 2, the photo-capacitance curves of the sample A are measured as a function of applied bias under the selective excitation of 1.33, 1.36, 1.39, 1.42, 1.44 and 1.61 eV at 12 K. The bottom trace is the C–V characteristic in the absence of light. A weak bump near +2 V may come from the possible tunnelling coupling between the GaAs QW and the n$^+$-GaAs top contact as the ground level E_{a1} of the GaAs QW (figure 2) is brought into alignment with the Fermi sea.

Upon turning on the laser light, the background capacitance in the positive bias region is continuously increased; it does not matter that the photon energy is lower or higher than the energy gap (1.51 eV) of GaAs. Since our n$^+$-GaAs substrate does show defect-related photoluminescence around 1.17 and 1.37 eV at 12 K (not shown), the enhanced background capacitance can be attributed to the photo-capacitance response of the n$^+$-GaAs substrate. Very intriguing is the appearance of two capacitance peaks at the biases of 1.26 and –0.80 V when the GaAs QW is resonantly excited by 1.61 eV laser light. Obviously, the photo-excited electrons in the ground state E_{a1} of the GaAs QW make the internal tunnelling coupling between the subbands in the $In_{0.17}Ga_{0.83}As$ QW (E_{b1}, E_{b2}) and the GaAs QW (E_{a1}) effective, leading to two sharp capacitance peaks at 1.26 and −0.80 V respectively. To verify this point, the energy levels in the GaAs QW and the $In_{0.17}Ga_{0.83}As$ QW are calculated in the absence of space charges within the framework of the envelope wavefunction approximation. The parameters used are listed in table 1. When the conduction band minimum of GaAs is taken as the energy

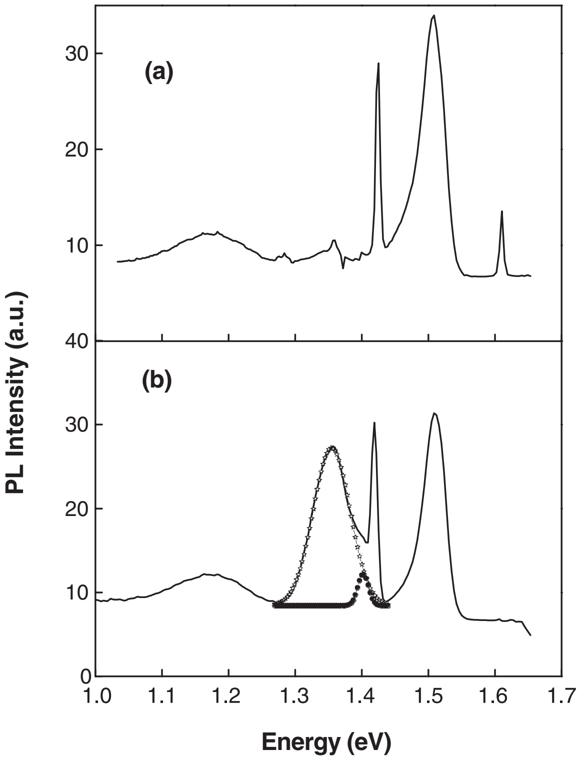

Figure 1. PL spectra of sample A (a) and sample B (b) at 12 K under the excitation of a 488.0 nm laser. Lines with stars and solid circles are the results after taking Gauss fit to the PL of SAQDs.

Table 1. Parameters used in calculation.

E_g(GaAs)	1.519 eV
E_g(AlAs)	3.099 eV[a]
E_g($In_{0.17}Ga_{0.83}As$)	1.261 eV[b]
ΔE_c(GaAs/AlAs)	1 eV
ΔE_v(GaAs/$In_{0.17}Ga_{0.83}As$)	0.02 eV[c]
m_e^*(GaAs)	$0.067m_0$
m_h^*(GaAs)	$0.45m_0$
m_e^*($In_{0.17}Ga_{0.83}As$)	$0.056m_0$[d]
m_h^*($In_{0.17}Ga_{0.83}As$)	$0.44m_0$

[a] Reference [19].
[b] Reference [20].
[c] Reference [21].
[d] Reference [22].

reference, the calculated values for E_{a1}, E_{b1} and E_{b2} are equal to 80, −106 and 284 meV, respectively. We also simultaneously obtain the energy levels of heavy holes in GaAs and $In_{0.17}Ga_{0.83}As$ QWs in order to make the interband transition energies in accordance with the measured values of 1.61 and 1.42 eV (figure 1). In reality, an energy level like E_{b1} below the structure's Fermi level, E_F, has to be filled to some extent in a thermal equilibrium condition. In other words, E_{b1} is going to pin at E_F, resulting in the lift-up of the whole set of E_{b1}, E_{b2} and E_{a1} to the new values of 0, 390, and 155 meV, respectively (see the inset of figure 2). Here,

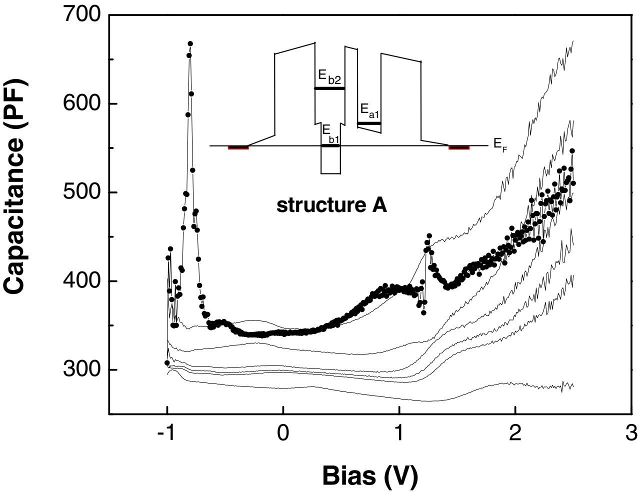

Figure 2. C–V curves of structure A. The trace on the bottom is measured in the absence of photo-excitation. The trace with solid circles is under the excitation of 1.61 eV. Others are under photo-excitation with photon energy of 1.33, 1.36, 1.39, 1.42, and 1.44 eV from the bottom up, respectively. The inset is a diagram of the conduction band structure with Fermi level pinning at E_{b1}.

E_{a1} is lifted by a potential energy increment of 106 meV × γ. Here γ is a leverage factor equal to 23 nm/32.5 nm. Based on Gauss's law, it can be easily proven that as long as the filling of E_{b1} is not larger than 1×10^{11} cm^{-2}, the following equation

$$(E_{\mathrm{b2}} - E_{\mathrm{a1}})/(E_{\mathrm{a1}} - E_{\mathrm{b1}}) = |V_{+}/V_{-}| \tag{1}$$

should be approximately satisfied. V_{+}, V_{-} are the bias positions for two capacitance peaks, and are equal to +1.26 and −0.80 V, respectively. The measured value, $|V_{+}/V_{-}|$, of 1.58 is close to the calculated ratio 1.52 of $(E_{\mathrm{b2}} - E_{\mathrm{a1}})/(E_{\mathrm{a1}} - E_{\mathrm{b1}})$. The above agreement gives evidence that the capacitance peak at the negative (positive) bias is a fingerprint of the internal resonant tunnelling between the ground state (first excited state) in the $\mathrm{In_{0.17}Ga_{0.83}As}$ QW and the ground state in the GaAs QW.

The photo-capacitance traces of sample B, selectively excited by photon energies of 1.33, 1.39, 1.42 and 1.44 eV at 12 K, are plotted in figure 3. In the absence of photo-excitation, the bottom trace is of no specific feature except for a broad bump in the bias range from 1.3 to 2.0 V. The latter may have a similar origin as in sample A, and arise from the weak tunnelling coupling between the GaAs QW and the n^{+}-GaAs top contact region at large positive bias. Upon illuminating the sample by the laser lights, two broad peaks show up at −0.32 and 0.71 V. Since the photon energies of the photo-excitation in use are already above the low energy tail of the SAQDs' photoluminescence (see figure 1(b)), for the same reason as mentioned for sample A, it is natural to attribute these two broad peaks to the internal tunnelling coupling between the ground states (the first excited states) of the SAQDs, E_{a1} (E_{a2}), and the ground state, E_{b1}, in the $\mathrm{In_{0.17}Ga_{0.83}As}$ QW. As also shown in figure 3, the higher the photon energy of the excitation, the stronger the capacitance peaks are as a result of the greater portion of the QD assembly being populated. According to the similar argument used in sample A, E_{a1} should also be pinned at the structure's Fermi level E_{F} (lying close to the GaAs band-edge), changing the band-edge profile into the one schematically drawn in the inset of figure 3.

As mentioned, since the photo-generated electrons in SAQDs are the only carrier source that enables an internal tunnelling process to occur between the $\mathrm{In_{0.5}Ga_{0.5}As}$ QD assembly and

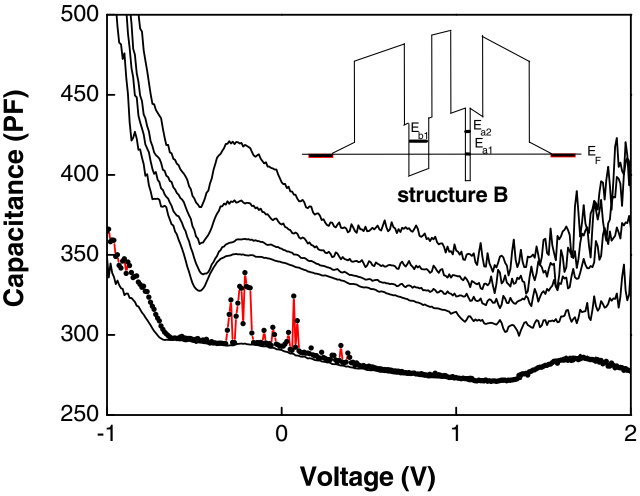

Figure 3. C–V curves of structure B. The trace on the bottom is measured in the absence of photo-excitation. The trace with solid circles is the spectra measured after the laser is turned off. Others are under different laser energies of 1.33, 1.39, 1.42 and 1.44 eV, respectively. The inset is a diagram of the conduction band structure with Fermi level pinning at E_{a1}.

the $In_{0.17}Ga_{0.83}As$ QW, we have proven in our previous work [17] that such a photo-capacitance response can be used to directly extract the averaged electron number density of the SAQDs in a form of

$$\frac{\mathrm{d}N_t}{\mathrm{d}V_t} = \frac{1}{\mathrm{e}}\frac{C_t^2 \Delta C(S)}{SC_0^2 - \Delta C(S)(C_t - C_0)} \qquad \text{(in the unit of cm}^{-2}\text{ meV}^{-1}\text{).}$$

Here, $C_0 = \varepsilon_0\varepsilon_r/d$ (d is the thickness of the undoped region), $C_t = \varepsilon_0\varepsilon_r/d_t$ (d_t is the distance across which the internal tunnelling takes place), and S is the area of the sample. $\Delta C(S)$ is the increment of the total capacitance under the excitation which can be well extracted from the experimental data in figure 3. As a result, the calculated maximum values of the densities of electrons in the first excited and ground states are 8.4×10^9 and 2.2×10^9 cm^{-2} mV^{-1} respectively, which are of the order of the density of self-assembled QDs in general. In addition, we shall demonstrate that one is also able to estimate the energy splitting, $\Delta E_a = E_{a2} - E_{a1}$, between the first and ground states of SAQDs from the available experimental data. First, from the PL spectra one can obtain the emission energies for electrons to escape from the energy levels in QWs or SAQDs. As seen in figure 1, the energy difference ΔE_{PL}(QD) between the bulk GaAs's and the $In_{0.5}Ga_{0.5}As$ QDs' transitions is about 160 meV, while the energy difference ΔE_{PL}(QW) between the bulk GaAs's and the $In_{0.17}Ga_{0.83}As$ QWs' transitions is equal to 85 meV. ΔE_{PL} is partitioned into the emission energy ΔE_e for the electrons and the emission energy ΔE_h for the holes according to a ratio of 2:1 [5, 18]. As a result, $\Delta E_e(\mathrm{QD}) = 107$ meV, $and \Delta E_e(\mathrm{QW}) = 56.7$ meV. The potential energy at the $In_{0.5}Ga_{0.5}As$ SAQD layer is lifted by 107 meV in order to make E_{a1} in alignment with the structure's E_F. Then, the potential energy at the $In_{0.17}Ga_{0.83}As$ QW layer is lifted up to 76.2 meV by considering the leverage factor of 23.5 nm/33 nm. The actual energy difference $E_{b1} - E_{a1}$ is given by 76.2 meV − 56.7 meV = 19.5 meV. Similar to equation (1), one has the relation of

$$(E_{a2} - E_{b1})/(E_{b1} - E_{a1}) = |V_-/V_+| \qquad (2)$$

where $V_- = -0.32$ V, $V_+ = 0.71$ V, and $E_{a2} - E_{b1} = 8.8$ meV. Finally, $E_{a2} - E_{a1} = 28.3$ meV. On the other hand, the PL energy difference between the first excited and ground states transitions can also be obtained to be 48 meV after taking a multiple Gauss fit to the PL spectrum of the SAQDs as shown in figure 1(b). Using the same partition ratio of 2:1, one has the energy splitting of $\Delta E_{a2a1} = 47$ meV $\cdot$ $(2/3) = 31.3$ meV. It is clear that the result of $E_{a2} - E_{a1}$, derived from the photo-capacitance, is consistent with that from PL spectra. As a matter of fact, such consistency does not very much depend on the partition ratio chosen in the evaluation.

Now, we turn to the capacitance trace, as indicated by the line with solid circles in figure 3, which was measured immediately after the laser light was turned off. It is very striking that many sharp peaks appear mainly in the bias range, where the electron tunnelling occurs between the first excited states of the SAQDs and the ground state of the $In_{0.17}Ga_{0.83}As$ QW. During the different run of turning on and turning off the laser light, such sharp peaks show up rather often, but their peak positions are not reproducible. As clarified previously, the SAQDs are already partially filled with electrons in the absence of illumination. It is well known that the energy levels as well as their filling in SAQDs are random in nature due to the fluctuation in size, shape and strain of the QDs. On the other hand, the photo-excitation and the subsequent capture/recombination of photo-excited carriers all take place statistically.

Based on the above argument, we propose the following conjecture. Upon turning off the photo-excitation, the photo-excited holes in the QD assembly should be exhausted by electrons very quickly. However, very few QDs might still have their first excited level filled by redundant electrons. Obviously, their number is very small. Accordingly, it is these few QDs with electrons filling in the first excited states that give rise to the occurrence of highly resolved capacitance peaks, no matter that the area of the optical window on our sample is as large as 200 μm × 200 μm.

In conclusion, the specific capacitance response induced by the internal tunnelling in three-barrier-two-well heterostructures (with and without SAQDs imbedded) has been verified to be useful in extracting the level structures for both QWs and QDs as well as the averaged electron number density of filled QWs and QDs levels. Because of the random natures for both optical transitions and the filling in a QD assembly, highly-resolved capacitance peaks have been observed after turning off the photo-excitation since only very few tunnelling events are initiated from a small number of QDs with their first excited states also filled.

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References

[1] Raymond S, Fafard S, Poole P J, Wojs A, Hawrylak P, Charbonneau S, Leonard D, Leon R, Petroff P M and Merz J L 1996 *Phys. Rev.* B **54** 11548

[2] Schmidt K H, Medeiros-Ribeiro G and Petroff P M 1998 *Phys. Rev.* B **58** 3597

[3] Landin L, Miller M S, Pistol M-E, Pryor C E and Samuelson L 1998 *Science* **280** 262

[4] Warburton R J, Schäflein C, Half D, Bickel F, Lorke A, Karral K, Garcla J M, Schoenfeld W and Petroff P M 2000 *Nature* **405** 926

[5] Chang W-H, Hsa T M, Yeh N T and Chyi J-I 2000 *Phys. Rev.* B **62** 13040

[6] Fry P W, Itskevich I E, Mowbray D J, Skolnick M S, Finley J J, Barker J A, O'Reilly E P, Wilson L R, Larkin I A, Maksym P A, Hopkinson M, Al-Khafaji M, David J P R, Cullis A G, Hill G and Clark J C 2000 *Phys. Rev. Lett.* **84** 733

[7] Itskevich I E, Ihn T, Thornton A, Henini M, Foster T J, Moriarty P, Nogaret A, Beton P H, Eaves L and Main P C 1996-I *Phys. Rev.* B **54** 16401

[8] Narihiro M, Yusa G, Nakamura Y, Noda T and Sakaki H 1997 *Appl. Phys. Lett.* **70** 105

[9] Main P C, Thornton A S G, Hill R J A, Stoddart S T, Ihn T, Eaves L, Benedict K A and Henini M 2000 *Phys. Rev. Lett.* **84** 729

[10] Vdovin E E, Levin A, Patanè A, Eaves L, Main P C, Khanin Yu N, Dubrovskii Yu V, Henini M and Hill G 2000 *Science* **290** 122

[11] Miller B T, Hansen W, Manus S, Luyken R J, Lorke A and Kotthaus J P 1997 *Phys. Rev.* B **56** 6764

[12] Kapteyn C M A, Lion M, Heitz R, Bimberg D, Brunkov P N, Volovik B V, Konnikov S G, Kovsh A R and Ustinov V M 2000 *Appl. Phys. Lett.* **76** 1573

[13] Ibanez J, Leon R, Vu D T, Chaparro S, Johnson S R, Navarro C and Zhang Y H 2001 *Appl. Phys. Lett.* **79** 2013

[14] Medeiros-Ribeiro G, Pinheiro M V B, Pimentel V L and Marega E 2002 *Appl. Phys. Lett* **80** 4229

[15] Wetzler R, Wacker A, Schöll E, Kapteyn C M A, Heitz R and Bimberg D 2000 *Appl. Phys. Lett.* **77** 1671

[16] Brunkov P N, Polimeni A, Stoddart S T, Henini M, Eaves L and Main P C 1998 *Appl. Phys. Lett.* **73** 1092

[17] Li G R, Zheng H Z, Yang F H and Hu C Y 2003 *Semicond. Sci. Technol.* **18** 760

[18] Schmidt K H, Medeiros-Ribeiro G, Oestreich M, Petroff P M and Döhler G H 1996 *Phys. Rev.* B **54** 11346

[19] Basio C, Staehli J L, Guzzi M, Burri G and Logan R A 1988 *Phys. Rev.* B **38** 3263

[20] Gilperez J M and Sanchez-Dehesa J 1994 *J. Appl. Phys.* **76** 5931

[21] Arent D J 1990 *Phys. Rev.* B **41** 9843

[22] Berolo O, Woolley J C and Van Vechten J A 1973 *Phys. Rev.* B **8** 3794