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Room temperature negative differential capacitance in self-assembled quantum dots

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Abstract
The negative differential capacitance (NDC) of Schottky diodes with layers of InAs quantum dots (QDs) has been clearly observed at room temperature. The frequency dependence of the NDC is investigated. The measured peak capacitances of NDC decay rapidly at the testing frequencies higher than a few kilohertz. A kinetic model considering the testing signal is proposed and the capture rates of QDs are extracted. The simulation result is quantitatively consistent with the experimental data when the charging effect in QDs is included.

In recent years the investigation of structures with self-assembled quantum dots (QDs) has drawn the increasing attention of researchers because of their potential application in nano-electronics. Researchers usually use optical methods to study the physical properties of QDs [1, 2], but electrical characterization like capacitance measurement is also essential for various potential applications. The charge accumulation in the QDs revealed specific features in capacitance–voltage (C–V) dependences [3–9]. Most of these reports presented the experimental results of the C–V dependences, and the parameters of QDs, such as concentration, energy levels and capture cross-sections, were determined accordingly. Models for calculating the capacitance dependence were also proposed for comparison with the experimental results. Recently, the negative differential capacitance (NDC) characteristic was observed [9, 10]. As stated in our previous report, the NDC could be caused by the fast charging–discharging process in the states of QDs and, more importantly, it is basically a zero-dimensional effect [10]. In this paper, we study the frequency dependence of NDC characteristics in the Schottky diodes with InAs QDs. Clear NDC behaviour is observed at room temperature. A small-signal model is also derived to explain the experimental data.

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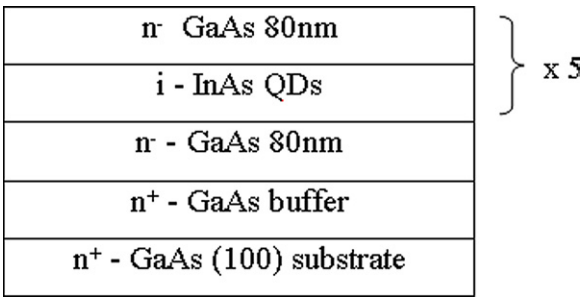

Figure 1. The grown sample structure with five layers of InAs QDs.

The sample was grown on n^+-GaAs (1 0 0) substrates by molecular beam epitaxy (MBE) using a Varian GEN II system equipped with an arsenic cracker cell. The detailed structure is shown in figure 1. The sample (LM3654) contains five layers of InAs QDs embedded in GaAs matrix. The spacer between the QDs layers is 80 nm lightly-doped ($N_D = 6.4 \times 10^{15}\,cm^{-3}$) GaAs. The InAs QDs were grown at 485 °C with an InAs growth rate of 0.05 ML s^{-1} and an arsenic (As_4) beam-equivalent-pressure (BEP) of 3×10^{-5} Torr. The area density of the QDs was about $1 \times 10^{11}\,cm^{-2}$ by using the atomic force microscope (AFM) measurement on a separate sample. Low temperature photoluminescence showed that the

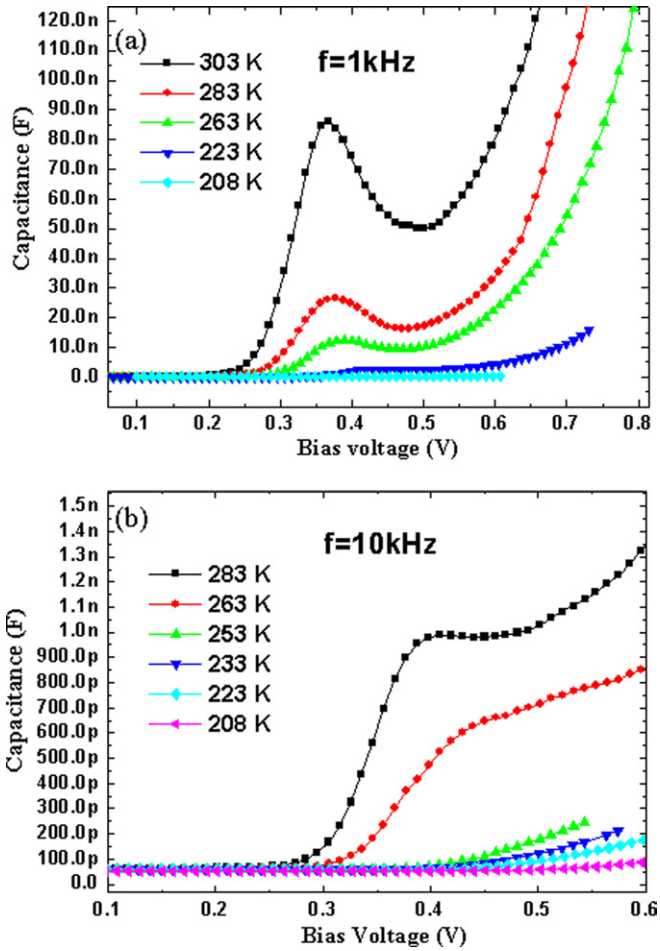

Figure 2. Temperature-dependent C–V curves measured with two testing frequencies, (a) 1 kHz and (b) 10 kHz.

ground-state transition energy was 1.23 eV with a full-width-half-maximum (FWHM) of 74 meV. The sample was then proceesed into 400 × 400 μm^2 Schottky diodes with Ti/Au (20 nm/100 nm) Schottky contacts.

The C–V measurement was carried out with an LCR meter (INSTEK LCR-819) in the frequency range from 400 Hz to 15 kHz at various temperatures. In figures 2(a) and (b), the C–V curves measured at 1 kHz and 10 kHz, respectively, are plotted. It is apparent that there is clear NDC behaviour around 0.4 V in almost all measured curves. The NDC phenomena become more significant as the temperatures go higher in both figures. This indicates that the charging–discharging time of QDs shortens as the temperature increases. Comparing the NDC peak values at the same temperature in figures 2(a) and (b), the difference is obvious. For example, at 283 K, the peak value of NDC at 1 kHz is around 25 nF but that at 10 kHz comes down to ∼1 nF. The frequency dependence can be seen more clearly in figure 3, where the C–V characteristics measured at 283 K in various testing frequencies are shown. Lower testing frequencies give higher NDC peak values, as expected. Roughly speaking, the charging–discharging time is in the order of 10^{-3} s because the peak values grow rapidly when the testing frequencies are lower than 2 kHz.

To understand the phenomena, we have to calculate the alternating current induced by the testing signal of capacitance.

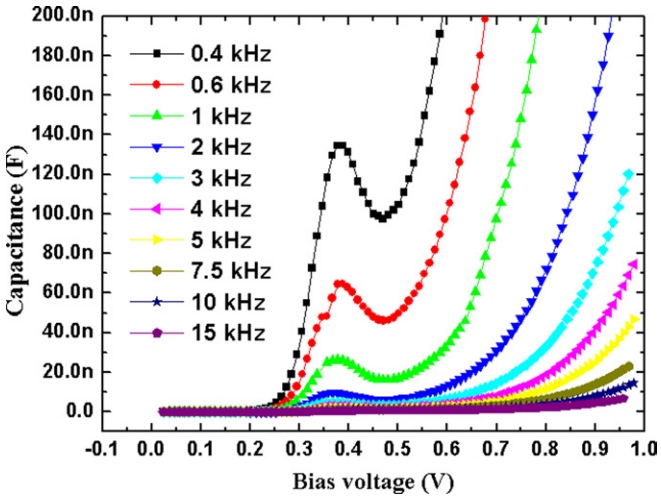

Figure 3. Frequency-dependent C–V curves measured at 283 K.

Based on the formulation in our previous letter [10], the present model further takes the testing signal into account. The band profile under consideration is sketched in figure 4. At first, without the alternating signal, the potential distribution is exactly the same as what we obtained previously [10].

$$\varphi(x) = -\frac{eN_{\mathrm{D}}}{2\varepsilon\varepsilon_0}(x-w)^2 + \left\{\begin{array}{l} 0,\ x > L \\ \dfrac{en_{\mathrm{d}}}{\varepsilon\varepsilon_0}(L-x),\ x < L \end{array}\right\}, \quad (1)$$

where the N_{D}, ε and L are the donor impurity concentration ($N_{\mathrm{D}} = 6.4 \times 10^{15}\,\mathrm{cm}^{-3}$), the dielectric constant of GaAs ($\varepsilon = 13.1$) and the distance between the layer of QDs and the Schottky contact ($L = 240\,\mathrm{nm}$), respectively. The electron concentration in QDs n_{d} and the depletion width w can be calculated by the following equations:

$$n_{\mathrm{d}} = N_{\mathrm{d}}\frac{2}{1+\exp\left(\frac{E_{\mathrm{F}}-E_1-e\varphi(L)}{kT}\right)}, \quad (2)$$

$$w^2 - \frac{2\varepsilon\varepsilon_0}{eN_{\mathrm{D}}}(\Phi_{\mathrm{B}}-V) - \frac{2N_{\mathrm{d}}}{N_{\mathrm{D}}} \times \frac{2}{1+\exp\left(\dfrac{E_{\mathrm{F}}-E_1-\frac{e^2N_{\mathrm{D}}}{2\varepsilon\varepsilon_0}(L-w)^2}{kT}\right)}L = 0, \quad (3)$$

where Φ_{B}, N_{d} and E_1 are the Schottky barrier height ($\Phi_{\mathrm{B}} = 0.81$ V), the QDs' area density ($N_{\mathrm{d}} = 1 \times 10^{11}\,\mathrm{cm}^{-2}$) and the QDs' ground-state energy below GaAs conductionband edge ($E_1 = 0.165$ eV), respectively. The solution of equation (3) can be obtained using the standard numerical method. In figure 5(a), we plotted the calculated band profile under various bias voltages. The effect of accumulated electrons in QDs becomes obvious when the bias voltage is higher than 0.4 V, as we can see the clear differential discontinuity of the band profile at a depth of 240 nm. The depletion width and carrier concentration in QDs are shown in figure 5(b). The depletion widths decrease as the bias voltages increase, as expected. However, the rate of decrease changes when the electrons occupation in QDs starts around 0.1 V (as shown by the other curve in figure 5(b)). The electrons in QDs have opposite

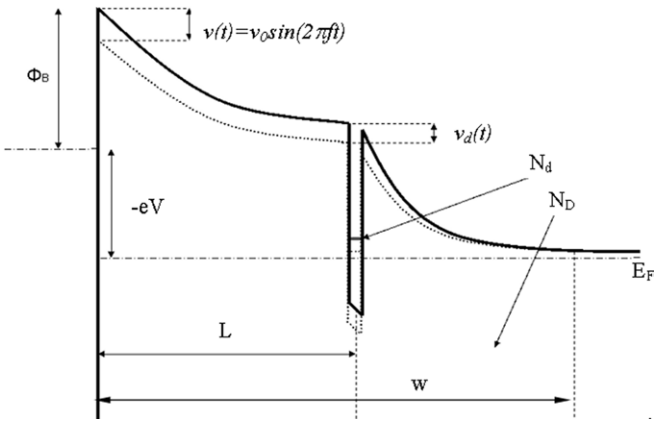

Figure 4. Band profile of a Schottky diode containing a single layer of QDs with/without the testing signal of capacitance.

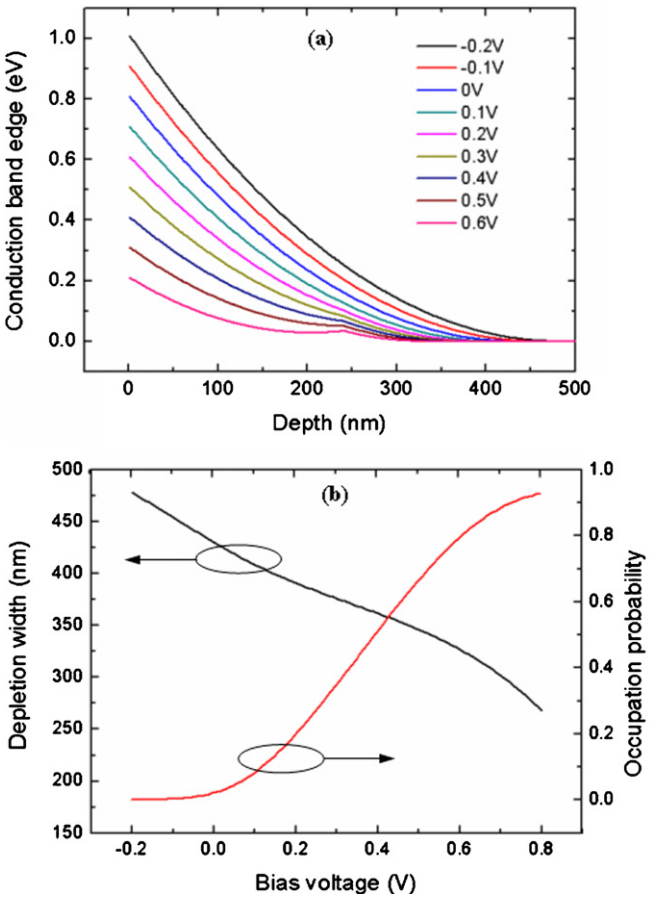

Figure 5. Simulation results of stationary solution: (*a*) the band profile under various bias voltages; (*b*) the depletion width and electron concentration in QDs versus the bias voltages.

charge polarity with the ionized impurities in the depletion region so the total effective charges decrease and then the depletion width shortens.

In the measurement of the capacitance of the diode, we have to apply a small ac signal upon the dc bias. When the testing sinusoidal signal of the capacitance is applied to the Schottky contact, we have the time-dependent voltage:

$$V = V_0 + v(t) = V_0 + v_0 \sin(2\pi f t), \tag{4}$$

where $v(t)$is the testing signal and f is its frequency. In the presence of the testing signal, the depletion width w and the electron concentration in QD n_d become time-dependent. Our task is to find out the current flow $I(t)$ responsible for the time-varying n_d and w:

$$\begin{aligned} I(t) &= \frac{\partial(\Delta q_\mathrm{d}(t))}{\partial t} + \frac{\partial(\Delta q_w(t))}{\partial t} \\ &= -e\frac{\partial(\Delta n_\mathrm{d}(t))}{\partial t} + eN_\mathrm{D}\frac{\partial(\Delta w(t))}{\partial t}. \end{aligned} \tag{5}$$

The first term accounts for the charging/discharging of QDs. The second one comes from the depletion width variation and can be evaluated with equation (1), provided that the amplitude of the testing signal is much smaller than the dc bias voltage ($V_0 \gg v_0$).

$$v_\mathrm{d}(t) = \frac{eN_\mathrm{D}}{\varepsilon\varepsilon_0}(w-L)\Delta w(t) = \left(1-\frac{L}{w}\right)\left(v(t) + \frac{eL}{\varepsilon\varepsilon_0}\Delta n_\mathrm{d}(t)\right), \tag{6}$$

where $v_\mathrm{d}(t)$ is the time-varying voltage at QDs depth L. At this stage, the only unknown factor to get the current $I(t)$ in equation (5) is $\Delta n_\mathrm{d}(t)$ because $\Delta w(t)$ can be calculated by equation (6). To obtain $\Delta n_\mathrm{d}(t)$, we consider the kinetic process of carrier capture/escape in QDs as follows [5, 11]:

$$\frac{\partial \Delta n_\mathrm{d}(t)}{\partial t} = \sigma_n \bar{v}_n n(t) - e_n n_\mathrm{d}(t). \tag{7}$$

The σ_n, $\bar{v}_n$ and e_n are the capture cross section area of QDs, the thermal velocity of electron in the conduction band and the emission rate of electron from QDs, respectively. The time-dependent functions $n(t)$ and $n_\mathrm{d}(t)$ are the free electron concentration near QDs and the number of electrons in QDs, which can be expressed as $n(t) = n_0 + \Delta n(t)$ and $n_\mathrm{d}(t) = n_{\mathrm{d}0} + \Delta n_\mathrm{d}(t)$, respectively. The emission rate can be easily estimated in view of which, without the testing signal, $\partial \Delta n_\mathrm{d}(t)/\partial t$ equals zero and so $e_n n_{\mathrm{d}0} = \sigma_n \bar{v}_n n_0 (N_\mathrm{d} - n_{\mathrm{d}0})$. Ignoring the higher order terms, equation (7) can be turned into the form

$$\frac{\partial \Delta n_\mathrm{d}(t)}{\partial t} = \sigma_n \bar{v}_n n_0 \left[\frac{(N_\mathrm{d} - n_{\mathrm{d}0})}{n_0}\Delta n(t) - \frac{N_\mathrm{d}}{n_{\mathrm{d}0}}\Delta n_\mathrm{d}(t)\right]. \tag{8}$$

The change in electron concentration in the conduction band, $\Delta n(t)$, can be approximated by $\Delta n(t) = n_0[\exp(ev_\mathrm{d}(t)/kT) - 1] \cong n_0 ev_\mathrm{d}(t)/kT$ if the amplitude of the testing signal is much smaller than kT/e. Putting this back into equation (8) and replacing $v_\mathrm{d}(t)$ by equation (6), we can solve the $\Delta n_\mathrm{d}(t)$ in terms of $v(t)$ with the form $\Delta n_\mathrm{d}(t) = \Delta n_{\mathrm{d}0}\exp(\mathrm{i}2\pi f t)$.

$$\Delta n_\mathrm{d}(t) = \frac{\varepsilon\varepsilon_0 f_0}{e\lambda}\frac{v(t)}{\mathrm{i}2\pi f + f_0\left(\frac{L}{\lambda} + \frac{N_\mathrm{d}}{n_{\mathrm{d}0}}\right)}. \tag{9}$$

The parameters are defined as $f_0 = \sigma_n \bar{v}_n n_0$ and $\lambda^{-1} = e^2 N_\mathrm{d}/kT\varepsilon\varepsilon_0(1 - (L/w))(1 - (n_{\mathrm{d}0}/N_\mathrm{d}))$. By putting equation (9) back into equation (5), and by using the definition in equation (10),

$$\frac{I(t)}{v(t)} = G(f) + \mathrm{i}2\pi f C(f) \tag{10}$$

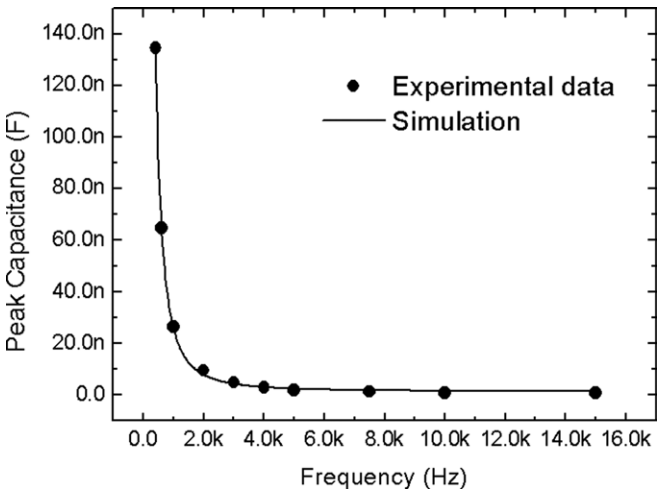

Figure 6. Measured and simulated frequency dependence of the peak capacitances around NDC.

the frequency-dependent capacitance can be obtained as follows:

$$C(f) = \frac{\varepsilon\varepsilon_0}{4\pi^2\lambda}\left(1 - \frac{L}{w}\right)\frac{f_0^2\left(\frac{L}{\lambda} + \frac{N_d}{n_{d0}}\right)}{f^2 + \frac{f_0^2}{4\pi^2}\left(\frac{L}{\lambda} + \frac{N_d}{n_{d0}}\right)^2} + \frac{\varepsilon\varepsilon_0}{w}. \quad (11)$$

The usefulness of equation (11) comes from the fact that the frequency dependence of the capacitance can be calculated once the stationary states solution is found. In figure 6, we have plotted the NDC peak capacitance versus the frequency of testing signal from the data in figure 3. Using the formula in equation (11), the fitted result is $C_{peak}(f) = 1.55 \times 10^{-9} + 0.0248/f^2 + (162.15)^2$. It is clear that the simulation curve is quite consistent with the experimental data. We can further get the value of f_0 from the denominator term in the fitted result if we know λ and N_d/n_{d0}. The latter is about 2 because the QDs states are half-filled when the capacitance reaches its peak value around the NDC [10]. The calculated depletion width without the testing signal is about 3.5×10^{-5} cm. Thus λ^{-1} is about $8.88 \times 10^3\,\mathrm{cm}^{-1}$. Accordingly, the obtained f_0 is 2.00×10^3 Hz. This value, corresponding to the capture rate of QDs ($f_0 = \sigma_n \bar{v}_n n_0$), is low but reasonable when we take the charging effect in QDs into account. That is, as the QD is occupied with one electron, the Coulomb repulsion builds up a potential barrier around the QD and lowers its capture rate of electrons. From the measured AFM image on a separate sample, the size of these QDs (σ_n) is about $10^{-12}\,\mathrm{cm}^2$. The thermal velocity of electrons ($\bar{v}_n$) at 283 K is $2.53 \times 10^7\,\mathrm{cm\,s}^{-1}$. The electron concentration in the conduction band (n_0) can be evaluated with

$$n_0 = N_C \exp\left(\frac{-\Delta E}{kT}\right). \quad (12)$$

N_C is the effective density of states in GaAs conduction band ($N_C = 4.7 \times 10^{17}\,\mathrm{cm}^{-3}$) and ΔE is the difference between the quasi-fermi level in QDs and the conduction band edge of GaAs [11]. Based on the fitted result of f_0, we can extract the ΔE of 0.549 eV. To consider the charging effect of QDs, we can approximate the QD as a disc to get its self-capacitance with $C_{QD} = 2\varepsilon\varepsilon_0\sqrt{\sigma_n}/\pi^{3/2}$, which is 4.17×10^{-19} F corresponding to a potential barrier of 0.384 eV due to its charging effect [12]. Therefore, the value of $\Delta E = 0.549$ eV is plausible if we take the ratio of conduction band discontinuity between the QDs states and the GaAs matrix to the GaAs bandgap difference as $\Delta E_c/\Delta E_g = 0.59$.

In conclusion, the NDC phenomenon in Schottky diodes with self-assembled QDs was observed at room temperature. The frequency dependence of the NDC behaviour was investigated. The extracted capture rate of the QDs is about 2 kHz. A small-signal model has been developed to explain the phenomenon. The simulation result is quite consistent with the experimental data. According to our analysis, the charging effect in these small-size QDs could play an important role in the capture process of electrons. Very recently, we learned that the frequency-dependent capture/escape process in QDs has also been observed with deep-level transient spectroscopy (DLTS) [13]. However, the behaviour was explained by the pure tunnelling effect. To clarify the connection and its physics, further studies on this issue are needed.

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