行政院國家科學委員會專題研究計畫 成果報告

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行政院國家科學委員會補助專題研究計畫成果報告

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量子點負微分電容特性研究
“Study on Negative Differential Capacitance of Quantum Dots”

計畫編號：NSC97-2221-E-009-161
執行期間：97 年 08 月 01 日 至 98 年 07 月 31 日
主持人：林聖迪 交通大學電子工程系副教授

一、中文摘要

研究零維系統的量子能態的主要方法是光學特性量測，光學方法的重要性在於它們在量子點的光電元件應用；可是，在僅使用一種載子的元件中(如量子點紅外線偵測器)，因為在量子點(quantum dot, QDs)中的載子-載子交互作用，量子點的特性與光學量測並不相同；要研究僅有電子或電洞的量子點性質，我們轉而使用電性量測的方法；在此為期一年的計畫中，我們使用包含量子點之 Schottky 二極體進行電容-電壓分析，基於負微分電阻的觀察所建立的模型，我們進行了 InAs/GaAs 量子點之溫度及頻率相關的捕捉/逃脫特性研究；藉由變頻變溫量測與我們所建立的物理模型，發現量子點的載子充放電時間約在微秒(ms)等級。

I. Abstract

The frequency dependence of negative differential capacitance (NDC) in Schottky diodes with InAs quantum dots (QDs) is studied. The measured peak capacitances of NDC decay rapidly as the testing frequencies are higher than a few kHz. 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.

二、計畫緣由與目的 (II. Motivation and goal)

In these years the investigation of structures with self-assembled quantum dots (QDs) draws the increasing attention of researchers because of their potential application on 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. Recently, 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 to compare with the experimental results. More interestingly, the negative differential capacitance (NDC) characteristic was observed at low temperatures [9]. The NDC behavior was confirmed in our previous work and a model considering charge distribution in the device was also proposed [10]. The simulation results showed that the NDC is caused by the fast charging-discharging process in the states of QDs. It is not difficult to guess that the NDC behavior would strongly depend on the period of capacitance testing signal, particularly when the period is comparable with the charging-discharging time. In this report, the frequency dependence of C-V characteristics is investigated. A small signal model is also derived to explain the experimental results.

三、研究方法及成果 (III. Method and result)

The sample was grown on n$^+$-GaAs (100) substrates by molecular beam epitaxy (MBE) using a Varian GEN II system equipped with an arsenic cracker cell. The detailed structure can be found in our previous letter [10]. Briefly, the sample (LM3654) contains 5 layers InAs QDs embedded in GaAs matrix. The spacer between QDs layers is 80nm lightly-doped (N_D=6.4x10^{15}cm^{-3}) GaAs. The InAs QDs were grown at about 485^{o}C with the InAs growth rate of 0.05ML/s and the arsenic (As_4) beam-equivalent-pressure (BEP) of 3x10^{-5}torr. The area density of the QDs was about 1x10^{11}cm^{-2} by using the atomic force microscope (AFM) measurement. Low temperature photoluminescence showed that the ground state transition energy was 1.23eV with full-width-half-maximum (FWHM) of 74meV.

The sample was then proceeded into 400x400μm^2 Schottky diodes with Ti/Au (20nm/100nm) Schottky contacts.

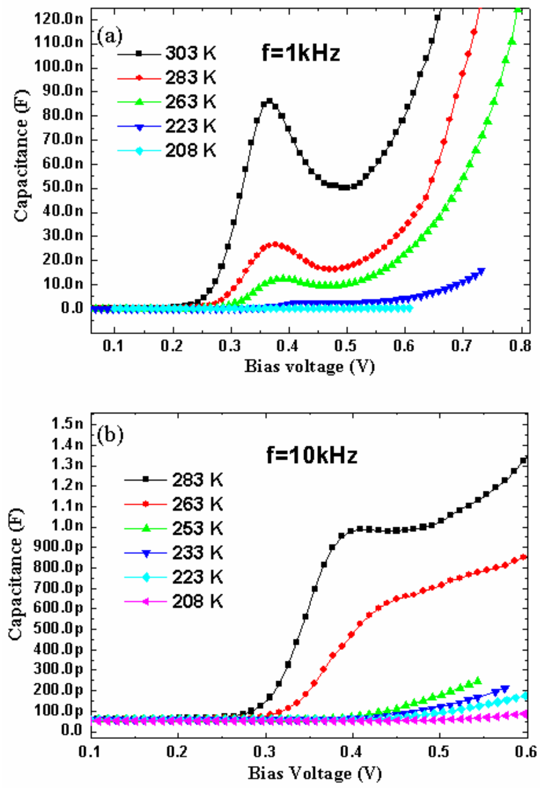

Fig.1 Temperature-dependent C-V curves measured with two testing frequencies, (a) 1kHz and (b) 10kHz.

The C-V measurement was carried out with an LCR-meter (INSTEK LCR-819) in the frequency range from 400Hz to 15kHz at various temperatures. The capacitance-voltage characteristics were obtained with a small AC signal (20mV in amplitude) over DC bias voltages. In the Fig.1a and Fig.1b, the C-V curves measured at 1kHz and 10kHz are plotted, respectively. It is apparent that the NDC behavior happening around 0.4V becomes more significant at higher temperatures in both figures. This tells us that the charging-discharging time of quantum dots shortens as the temperature increases. Comparing the NDC peak values at the same temperature in the Fig.1a and Fig.1b, the difference is obvious. For example, at 283K, the peak value of NDC at 1kHz is around 25nF but that at 10kHz comes down to ~1nF. The frequency dependence can be seen more clearly in the Fig.2, where the C-V characteristics measured at 283K 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 2kHz.

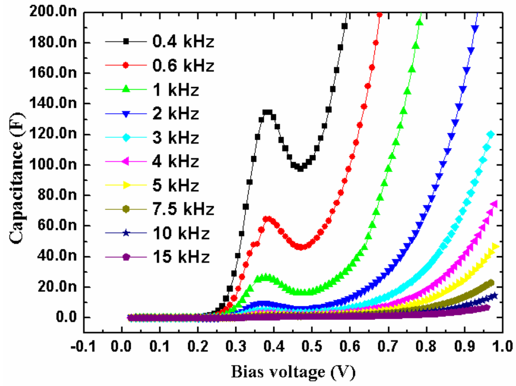

Fig.2 Frequency-dependent C-V curves measured at 283K.

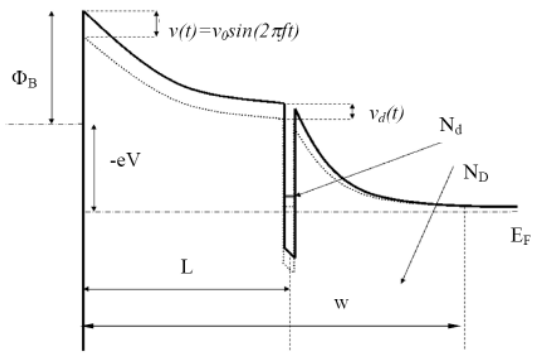

Fig.3 Band profile of a Schottky diode containing a single layer of QDs with/without the testing signal of capacitance.

To understand the phenomena, we have to calculate the alternating current induced by the testing signal of capacitance. Based on the calculation in our previous letter [10], the present model takes the testing signal into account. The band profile under consideration is sketched in the Fig.3. Here, to simplify the calculation, only a single layer of QDs is considered and its carrier concentration is taken as a delta-function due to QDs' nano-scale size. At first, without the alternating signal, the potential distribution is exactly the same as what we obtained previously [10].

$$\varphi(x) = -\frac{eN_D}{2\varepsilon\varepsilon_0}(x-w)^2 + \left\{ \begin{array}{l} 0, x > L \\ \frac{en_d}{\varepsilon\varepsilon_0}(L - x), x < L \end{array} \right\} \qquad (1)$$

where the N_D, ε and L are the donor impurity concentration (N_D=6.4x10^{15}cm^{-3}), the dielectric constant of GaAs (ε=13.1) and the distance between the layer of QDs and the Schottky contact (L = 8x10^{-6}cm), respectively. The electron concentration in QDs n_d and the depletion width w can be calculated by the following equations.

$$n_d = N_d \frac{2}{1+\exp(\frac{E_F - E_1 - e\varphi(L)}{kT})}, \tag{2}$$

$$w^2 - \frac{2\varepsilon\varepsilon_0}{eN_D}(\Phi_B - V) - \frac{2N_d}{N_D}\frac{2}{1+\exp(\frac{E_F - E_1 - \frac{e^2 N_D}{2\varepsilon\varepsilon_0}(L - w)^2}{kT})}L = 0 \tag{3}$$

where N_d and E_1 are the area density (N_d =1x10^{11}cm^{-2}) and the QDs' ground-state energy below GaAs conduction-band edge, respectively. When the testing sinusoidal signal of 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 the $v(t)$ is the testing signal and f is its frequency. The depletion width w and the electron concentration in QD n_d become time-dependent in the presence of the testing signal. Our task is to find out the current flow $I(t)$ responsible for the time-varying n_d and w.

$$I(t) = \frac{\partial(\Delta q_d(t))}{\partial t} + \frac{\partial(\Delta q_w(t))}{\partial t} = -e\frac{\partial(\Delta n_d(t))}{\partial t} + eN_D\frac{\partial(\Delta w(t))}{\partial t} \tag{5}$$

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

$$v_d(t) = \frac{eN_D}{\varepsilon\varepsilon_0}(w - L)\Delta w(t) = (1 - \frac{L}{w})(v(t) + \frac{eL}{\varepsilon\varepsilon_0}\Delta n_d(t)) \tag{6}$$

where the $v_d(t)$ is the time-varying voltage at QDs depth L. The only unknown factor to get the current $I(t)$ in the eq.(5) is $\Delta n_d(t)$ now because the $\Delta w(t)$ can be calculated by the eq.(6). To obtain the $\Delta n_d(t)$, we have to consider the kinetic process of carrier capture/escape in QDs as follows.

$$\frac{\partial \Delta n_d(t)}{\partial t} = \sigma_n \bar{v}_n n(t)[N_d - n_d(t)] - e_n n_d(t) \tag{7}$$

The σ_n, $\bar{v}_n$, $n(t)$ and e_n are the capture cross section area of QDs, the thermal velocity of electron in the conduction band, the electron concentration near QDs in the conduction band and the emission rate of electron from QDs, respectively [5, 11]. The number of electrons in QDs is the time-dependent $n_d(t)$. The emission rate can be easily estimated in view of that, without the testing signal, the $\frac{\partial \Delta n_d(t)}{\partial t}$ equals to zero and so $e_n n_d(t) = \sigma_n \bar{v}_n n(t)[N_d - n_d(t)]$. The eq.(7) can be turned into the form, by using $n_d(t) = n_{d0} + \Delta n_d(t)$, $n(t) = n_0 + \Delta n(t)$ and by ignoring the higher order terms,

$$\frac{\partial \Delta n_d(t)}{\partial t} = \sigma_n \bar{v}_n n_0 \left[\frac{(N_d - n_{d0})}{n_0}\Delta n(t) - \frac{N_d}{n_{d0}}\Delta n_d(t)\right] \tag{8}$$

The change of electron concentration in the conduction band, $\Delta n(t)$, because of the testing signal $v(t)$, can be approximated by $\Delta n(t) = n_0[\exp(ev_d(t)/kT) - 1] \cong -n_0 e v_d(t)/kT$ if the amplitude of testing signal is much smaller than *kT/e*. Put this back to the eq.(8) and replace the $v_d(t)$ by the eq.(6), we can solve the $\Delta n_d(t)$ in terms of $v(t)$ with the assumption of $\Delta n_d(t) = \Delta n_{d0}\exp(i2\pi f t)$. According to the definition

$$\frac{I(t)}{v(t)} = G(f) + i2\pi f C(f) \tag{9}$$

, we can finally get the frequency-dependent capacitance from the eq.(5).

$$C(f) = \frac{\varepsilon\varepsilon_0}{4\pi^2\lambda}\left(1 - \frac{L}{w}\right)\frac{f_0^2\left(\frac{L}{\lambda} + \frac{N_d}{n_{d0}}\right)}{f^2 + \frac{f_0^2}{4\pi^2}\left(\frac{L}{\lambda} + \frac{N_d}{n_{d0}}\right)^2} + \frac{\varepsilon\varepsilon_0}{w} \tag{10}$$

The parameters are defined as follows, $f_0 = \sigma_n \bar{v}_n n_0$ and $\lambda^{-1} = \frac{e^2 N_d}{kT\varepsilon\varepsilon_0}\left(1 - \frac{L}{w}\right)\left(1 - \frac{n_{d0}}{N_d}\right)$.

The usefulness of the eq.(10) comes from that the frequency-dependence of capacitance can be calculated once the stationary states solution is found [10]. To compare the measured data with the derived formula in the eq.(10), we took the values of peak capacitance near the NDC (around 0.3-0.4V) in the Fig.2 and plotted the NDC peak capacitance v.s. the frequency of testing signal. The data and the fitting curve are shown in the Fig.4. Using the formula in the eq.(10), the curve is $C_{peak}(f) = 1.55\times10^{-9} + \frac{0.0248}{f^2 + (162.15)^2}$. It is clear that the simulation curve is consistent well with the experimental data. We can also get the value of f_0 from the denominator term in the fitted result if we know λ and N_d/n_{d0}. The later one 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 2.3×10^{-5} cm thus λ^{-1} is about 1.84×10^{4} cm^{-1}. Accordingly, the obtained f_0 is 1.57×10^{3} Hz. The 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 down its capture rate of electron. From the measured AFM image, the size of these QDs (σ_n) is about 10^{-12}cm^2. The thermal velocity of electrons ($\bar{v}_n$) at 283K is 2.53×10^{7} cm/sec. The electron concentration in the conduction band (n_0) can be evaluated with

$$n_0 = N_C \exp\left(\frac{-\Delta E}{kT}\right) \qquad (11)$$

N_C is the effective density of states in GaAs conduction band (N_C = 4.7×10^{17} cm^{-3}) and the 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 0.555eV. To consider the charging effect of QDs, we can approximate the QD as a disk 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 0.384eV due to its charging effect [12]. Therefore, the value of ΔE =0.555eV 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 $\Delta E_c / \Delta E_g = 0.61$.

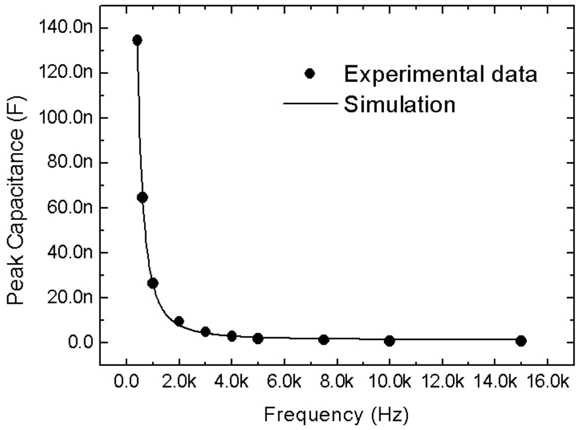

Fig.4 Measured and simulated frequency dependence of the peak capacitances around NDC.

四、結論 (IV. Conclusion)

In conclusion, the frequency dependence of the NDC behavior in a Schottky diode with the layers of QDs has been investigated. A small-signal model has been developed to explain the phenomena. The simulation result is well 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. The capture rate of the self-assembled QDs, which is an important parameter in unipolar QDs devices like quantum-dots infrared photodetector [12], can be extracted from our experimental data with our model. Our results also indicate that the potential distribution in the Schottky diode can be modified by the captured electrons in QDs. Due to the slow emission/capture rate in these small QDs even at room temperature, applications like QDs memory devices are also promising [13].

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