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Ultrafast carrier capture in charged InAs quantum dots

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Abstract

We report theoretical and experimental results of our investigation on carrier capture and relaxation processes in undoped and modulation-doped InAs/GaAs self-assembled quantum dots (QDs). We find that carrier capture and relaxation in the ground state is faster in the modulation-doped quantum dots compared to the case in neutral dots at an excitation level as low as one electron–hole pair per dot. The ultrafast photoluminescence (PL) transient rise time observed in the charged dots is attributed to the relaxing of strained field induced by the presence of cold carriers in the dots. The Hamiltonian of electron's interaction with local vibrating field and carrier capture time are also calculated.
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1. Introduction

Quantum dots (QDs) are the subject of a rapidly developing area in semiconductor research. Many groups have reported the fabrication of InAs on GaAs by methods of self-organized growth. One of the prominent fabrication methods for QDs is the Stranski–Krastanov (S–K) process which uses the lattice mismatch between the substrate and the over layers [1–3]. Formation of ordered pyramidal-shaped QDs was observed on a residual two-dimensional layer above a nominal coverage of two monolayer (ML) for InAs on GaAs (001) substrates by Ruvimov and co-workers [4]. Segregation of InAs wetting layers (WLs) was investigated by Offermans et al. using cross-section scanning tunneling microscopy [5]. It was concluded that the formation of WLs is a delicate interplay between surface migration, strain-driven segregation, and the dissolution of QDs during growth. Calculations also indicated that the WL considerably affects both the single-particle energy and wave function [6]. Recently, a new self-organized growth method using droplet epitaxy was reported for direct formation of QD systems without WLs [7,8].

The study of carrier relaxation and capture in InAs/GaAs QDs [9–12] has attracted much attention due to their unique optical and electrical properties for potential device applications. It has been shown that the removal of WL does not affect the carrier capture and relaxation in QDs [13]. Recently, Yuan et al. [14,15] studied the carrier dynamics in InAs/GaAs QDs. Their observation on systematically longer PL risetimes in the higher excited states was interpreted in the frame work of sequential state filling, resulting from fast trapping and intra-dot relaxation. However, few experiments have examined carrier dynamics in charged QDs. In a recent work done by Gündoğdu et al. [16,17], carrier capture and relaxation to the ground state was found to be much faster in the highly charged dots compared to the neutral dots. Their results for p-doped QDs reveal a threefold decrease in the room temperature electron capture and

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relaxation time relative to corresponding undoped QDs. The enhancement of carrier capture and relaxation was attributed to the rapid electron–hole scattering involving the built-in carrier population.

In this paper, we investigate the carrier capture and relaxation process in undoped, lightly n-doped and p-doped InAs/GaAs QDs using time-resolved luminescence up-conversion spectroscopy techniques. We report observation of ultrafast carrier capture and relaxation in the charged QDs' ground states at very low doping concentrations and at low excitation levels. We have proposed a model for interpreting our experimental results. A comparison between experiments and calculations allows us to conclude that the presence of the few cold carriers has a significant influence on the capture and relaxation in the charged QDs.

2. Experiments and results

The InAs QD samples were grown by using a solid source molecular beam epitaxy (MBE) machine [18]. The growth procedure and condition are described as follows. After native oxide desorpted under As flux at 610 °C, a 2500 Å GaAs buffer layer was deposited at 570 °C to recover the substrate surface. 300 Å $Al_{0.3}Ga_{0.7}As$ and 1400 Å GaAs were deposited at a growth rate of 1 μm/h at the same temperature. Then, 100 Å GaAs was grown at a growth rate of 0.3 μm/h. In the meanwhile, the substrate temperature is lowered down (520 °C) for InAs deposition, and the desired As flux for QDs growth was achieved by adjusting the needle valve of the As cracker cell. Afterwards, 10 nm GaAs capped layer was deposited at the same temperature of QDs growth with a growth rate of 1 μm/h. Then the substrate temperature and As flux were raised to the original values for GaAs and $Al_{0.3}Ga_{0.7}As$ growth. Finally, a layer of uncapped QDs was grown with the same conditions, and then the substrate cooled down under As flux immediately. The n-doped (p-doped) samples contain a Si-delta (Be-delta) doping layer 2 nm below the QD layer with nominal densities of about $2 \times 10^{10}\ \mathrm{cm}^{-2}$. The average sheet density of the QDs is about $2 \times 10^{10}\ \mathrm{cm}^{-2}$. Images from atomic force microscopy (AFM) measurements reveal circular shaped QDs with an average dot size of approximately 20 nm in base width and a height of about 5 nm. Free carriers from the doped layer accumulated in the low energy states within the InAs QDs. In the lightly doped samples, most of the dots only contain a single electron or hole.

The measurements of carrier dynamics were performed by time-resolved photoluminescence with time-resolution of about 200 fs. The self-mode-locked Ti:sapphire laser was operated at 788 nm with spectral width of about 18 meV (full width at half maximum) to excited carriers in the GaAs barrier. According to the focus spot size and the absorption depth at the photoexcitation wavelength, the laser pumping power was adjusted to give injected carrier densities from $2 \times 10^{10}\ \mathrm{cm}^{-2}$ (low) to $5 \times 10^{11}\ \mathrm{cm}^{-2}$ (high), respectively. The accuracy of the excitation density is approximately a factor of two. The room temperature steady state PL spectra of the undoped QD sample taken at an excitation wavelength of 788 nm and intensity of $1 \times 10^3\ \mathrm{W/cm^2}$ are shown in Fig. 1. The spectral lines centered at 872 nm and 930 nm are attributed to the bandgap energies of the GaAs and WLs (as shown in Fig. 1(a)), respectively. Three spectrally well-separated PL lines $n = 1$ (ground state), $n = 2$ and $n = 3$ (excited states) at longer wavelengths (as shown in Fig. 1(b)) are due to electron–hole recombination between distinct QD states of the conduction and valence bands. Only the $n = 1$ peak is observed at low excitation intensity and it is assigned as the QD ground electron to ground hole transition. The center wavelengths of the spectral peak $n = 1$ to $n = 3$ are 1235 nm, 1160 nm and 1085 nm, respectively. The energy separations (also shown in Fig. 1(b)) between the QD confined states are 65 and 74 meV. In the PL studies on modulation-doped QDs, due to the lightly doping, we did not observe significant changes in the spectra lineshape or shift-

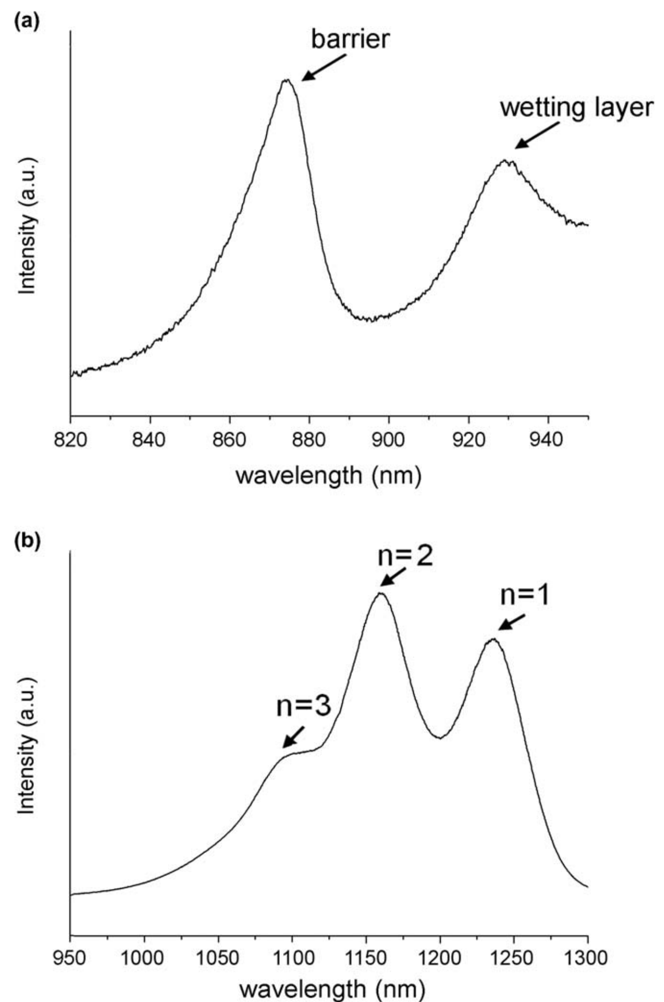

Fig. 1. Room temperature PL spectra of InAs/GaAs self-assembled QDs, displaying GaAs barrier, wetting layer and excited state radiative recombination in wavelength range from (a) 820 to 950 nm and (b) 950 to 1300 nm. Spectra were excited at 788 nm with a self-mode-locked Ti:sapphire laser.

ing of ground-state optical transition. Carrier capture and relaxation to the QD's ground level are examined as a function of excitation density and temperature by measuring PL rise times at the energy of QD ground state identified in the steady state PL spectra. The time evolution of the PL signal then follows from the analysis of the rate equations

$$I(t) \propto A * [\exp(-t/\tau_r) - \exp(-t/\tau_d)]/(\tau_r - \tau_d),$$

where τ_r and τ_d are the PL rise and decay time constants, respectively.

In Fig. 2 we show PL transients detected at the ground state of the undoped QDs for the first 20 ps at low temperature. Time-resolved PL measured at the same energy but at three different excitation levels (low, moderate and high) is displayed in parallel for comparison. The PL rise times of the ground state in the QDs accelerate as the excitation power increases and reach a value of less than 1 ps at a photoexcited carrier density $\geqslant 1 \times 10^{11}$ cm^{-2}. It is believed that the carrier density dependence of the ultrafast relaxation is due to Auger-like carrier–carrier scattering [9]. Experiments on our modulation-doped QDs allow the relaxation dynamics of electrons and holes to be investigated separately. At low temperature, low excitation densities, the photo-generated carriers do not significantly perturb the well-defined Fermi distribution of doped cold carriers. Therefore, the luminescence dynamics is dominated by the electron (hole) dynamics in p (n) modulation-doped QDs. For the modulation-doped samples with doping density of only 2×10^{10} cm^{-2}, only the lowest electron (n-doped QDs) or hole (p-doped QDs) level is occupied prior to the optical excitation. The initial transient

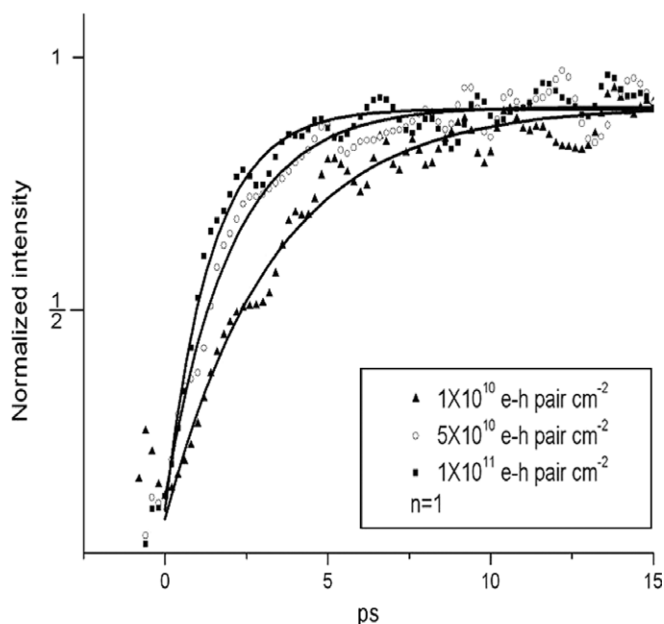

Fig. 2. Time-resolved photoluminescence intensity measured at the energy of the undoped QDs' ground states at three different excitation levels and at low temperature. The solid curves indicate fits of the PL rise to a single exponential.

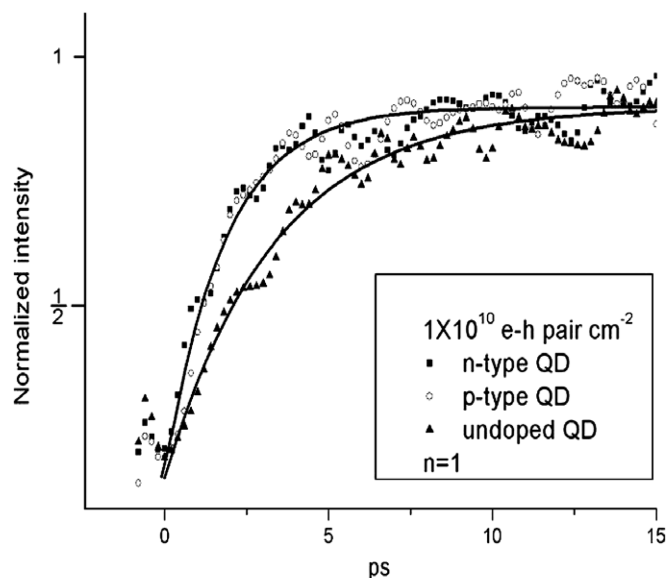

Fig. 3. Results of time-resolved PL experiments at 77 K for p-doped (circles), n-doped (squares) and undoped QDs (triangles).

PL at the ground-state optical transition in the modulation-doped QDs at low temperature is shown in Fig. 3. The results in Fig. 3 correspond to an optical excitation level of only one electron–hole pair per dot. Therefore, the major difference between the time-resolved PL experiments on the charged and uncharged QDs is the presence of small built-in electron (hole) population in the n-doped (p-doped) QDs prior to optical excitation. However, the fits of the PL transients in Fig. 3 indicate capture times of less then 2 ps for charged QDs which are more rapid than in the undoped QDs, regardless of the species of cold carrier involved. The time-resolved PL rise times also show no discernible temperature dependence for both the charged and uncharged QDs.

In contrast to the earlier report on carrier capture and relaxation in highly charged QDs [16,17], the total carrier densities in our experiments are only on the order of $\sim 2 \times 10^{10}$ cm^{-2}. According to the results reported in Ref. [19], for a plasma density of $\sim 10^{10}$ cm^{-2} and a quantum dot lateral size of 20 nm, the calculated Auger scattering rate was only on the order of $\sim 2 \times 10^{10}$ s^{-1}. Therefore, it is unlikely that the Coulomb scattering within the electron–hole plasma is responsible for the ultrafast carrier capture and relaxation observed in our charged QD experiments. The observation of no discernible temperature dependence on the PL transient rise time also indicates that phonon scattering was not responsible for the accelerated carrier capture observed in our experiments. In the following section, a theoretical model is proposed to interpret our experimental results.

3. Theoretical model

We model a circular shaped InAs QD with a base diameter of 20 nm and a height of 2 nm. In response to

the localized (doped) charges in the dots, a polarization field $\vec{P}(\vec{R}) = \varepsilon_0 \vec{\nabla}_R V(\vec{R})$ (where $V(\vec{R}) = e(1 - 1/\varepsilon_s)/4\pi\varepsilon_0 R$ and ε_s is the static dielectric constant of semiconductor) must be induced around the QDs. During the scattering, electron (or hole) must encounter two electric fields of opposite signs. Those fields are positive (negative) field of a bare hole (electron) confined within the dot and screening field of local polarizations around the dot. When the electric field inducing the strain around the dots suddenly disappears due to the capture of an electron or a hole into the QDs, local strain of crystal lattice must trigger vibrations of ions and bond electrons around the dot to relax the strain and to release the energy.

When a mobile electron was scattered on the polarization field produced by positively charged dots, by Fourier transforming this polarization field, the electron energy in the polarization field E_{kc} can be expressed by the sum of partial contributions from local strains with wave number q,

$$E_{\text{ev}} = -\frac{e^2(1 - 1/\varepsilon_s)}{2\pi^2\varepsilon_0} \cdot \int \frac{\sin(qR)}{qR} \mathrm{d}q. \tag{1}$$

Assuming that the system of ions and bond electrons is linear during interaction, the decay of locally induced vibrations is given by the Bessel function $J_0(\omega_{\text{m}} t)$, where ω_{m} is the maximum value of phonon frequency [20]. For simplicity we assume that polarization induces constant potential, $V(R_0)$, within the dot, i.e. the volume of the dot is never polarized, where R_0 is the average radius of quantum dot. Then for electron interacting with local vibrations triggered within the dot at the time t_0, the time-dependent Hamiltonian can be written as

$$H_{\text{ev}}(t - t_0) = -\frac{e^2(1 - 1/\varepsilon_s)}{2\pi^2\varepsilon_0} \cdot \int \frac{\sin(qR_0)}{qR_0} \cdot J_0[\omega_m(t - t_0) - qR_0(1 - R/R_0)]\mathrm{d}q, \tag{2}$$

where $qR_0(1 - R/R_0)/\omega_{\text{m}}$ is the time that vibration with wave number q takes to arrive to a point R within the dot. The Hamiltonian $H_{\text{ev}}(t - t_0)$ brings short-time perturbation to the electron scattering processes and it can result in the capture of photoexcited carriers into the QD's confined states. Due to low photoexcitation densities, number of excitons created in the sample was no more than one exciton per dot. Distance between QDs was far enough so that the Coulomb interaction was negligible. Therefore, the Coulomb interaction was not included in the Hamiltonian.

Denote n the density of mobile electrons and Ω the volume of quantum dot. In the frame of time-dependant perturbation theory, if the wave function of mobile states $|k\rangle$ is normalized to the flow of one electron per second, the rate $W = |\int \langle c|H_{\text{ev}}(t - t_0)|k\rangle \mathrm{d}t|^2 \sqrt{n\Omega}/\hbar^2 \Delta t$ of the variation-assisted transitions of electrons from mobile into confined electronic state $|c\rangle$ can be written as

$$W = \frac{e^4(1 - 1/\varepsilon_s)^2 n^{1/2}}{16\pi^2\varepsilon_0^2\hbar^2\omega_{\text{m}} R_0^{1/6}} F(\Delta t)|K(k)|^2, \tag{3}$$

where

$$F(\Delta t) = \frac{\omega_{\text{m}}}{\Delta t} \left| \int_0^{\Delta t} J_0(\omega_{\text{m}} t) \exp[\omega_{kc} t] \mathrm{d}t \right|^2, \tag{4}$$

$$K(k) = \frac{(12/\pi)^{1/4}}{\pi \cdot R_0^{3/2}} \int\int \frac{\sin(qR_0)}{q} \exp[\mathrm{i}\vec{k}\vec{R}] \times \exp\left[\mathrm{i}\frac{\omega_{kc}}{\omega_{\text{m}}} q(1 - r)\right] B\left[\Delta t - \frac{qR_0(1 - r)}{\omega_{\text{m}}}\right] \cdot \psi_c^* \mathrm{d}q\, \mathrm{d}R^3, \tag{5}$$

and

$$B[v] = \int_0^v J_0(\omega_{\text{m}} z) \exp(\mathrm{i}\omega_{kc} z)\mathrm{d}z \bigg/ \int_0^{\Delta t} J_0(\omega_{\text{m}} z) \exp[\mathrm{i}\omega_{kc} z]\mathrm{d}z. \tag{6}$$

Here $\vec{k}$ is the electron wave vector in mobile state, ψ_c^* is the electron wave function in confined state, $\hbar\omega_{kc}$ is the energy separation between confined and mobile electronic states in quantum dot and $r = R/R_0$. Vibrations around the quantum dot decay as cylindrical Bessel function $J_0(\omega_{\text{m}} t)$. By approximating the confined electronic states with the spherical Bessel function, $\Psi_c = \sqrt{2/3}\pi \cdot j_0(\pi \cdot r)/\sqrt{\Omega}$, Eq. (5) can be further reduced to

$$K(k) = \frac{4\sqrt[4]{3}}{\pi^{3/4}} \int_0^{\omega_{\text{m}}\Delta t} \mathrm{d}q \frac{\sin qR_0}{q} \times \int_0^1 \mathrm{d}r \exp\left(\frac{\mathrm{i}\omega_{kc} qR_0(1 - r)}{\omega_{\text{m}}}\right) \sin(r\pi) \frac{\sin(kR_0 r)}{kR_0}. \tag{7}$$

Therefore, the rate of carrier capture into the QDs $\tau = 1/W$ is calculated to be

$$\tau = \frac{16\pi^2\varepsilon_0^2\hbar m * (\omega_{kc}^2 - \omega_{\text{m}}^2)R_0^{5/2}}{e^4(1 - 1/\varepsilon_s)^2 n^{1/2}|K(k)|^2}. \tag{8}$$

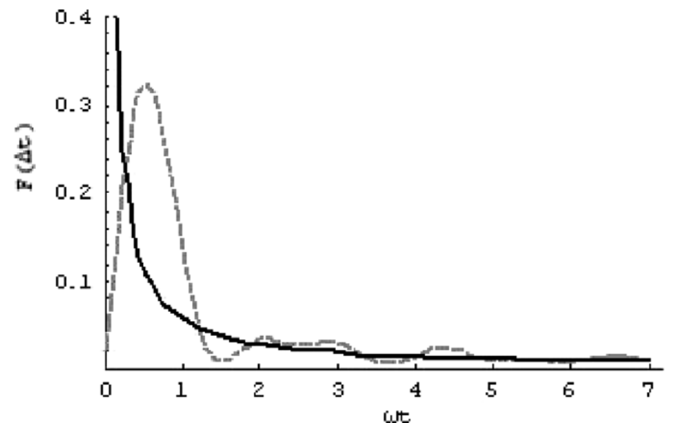

Fig. 4. Dependence of oscillation frequency $F(\Delta t)$ on $\omega_{\text{m}}\Delta t$ (dash curve) for $\omega_{kc}/\omega_{\text{m}} = 4.286$. When $\omega_{\text{m}}\Delta t > 1$, $F(\Delta t)$ can be reduced to $\frac{\omega_{\text{m}}}{(\omega_{kc}^2 - \omega_{\text{m}}^2)\Delta t}$ (solid line) for $\omega_{kc}/\omega_{\text{m}} = 4.286$.

The rate τ is mainly decided by the energy separation of confined states $\hbar\omega_{kc}$, the duration of perturbation Δt and the function $F(\Delta t)$. The dependence of $F(\Delta t)$ on $\omega_{\mathrm{m}}\Delta t$ is plotted in Fig. 4 for InAs/GaAs quantum dots with $\hbar\omega_{kc} = 150$ meV and $\hbar\omega_{\mathrm{m}} = 36$ meV. For the given parameters in our p-doped QD experiments, Eq. (8) gives a capture rate of $\tau = 1.7$ ps. This calculated value is in good agreement with our experimental results. In the case of holes been captured in an n-doped QD, the calculations should taking into account the warping of valence bands due to the quantum confinement and strain.

4. Conclusion

In conclusion, we have investigated carrier capture and relaxation in the ground state of undoped and modulation-doped InAs/GaAs QDs. We observe faster capture and relaxation processes in the charged QDs in comparing to the undoped dots even in the one electron–hole pair per dot regime. Our results suggest that, under low excitation intensity and low doping level, the relaxing of polarization field induced by the confined charge in the quantum dot is the dominant factor for the acceleration of carrier capture. Our calculations also show that carriers interact with decaying field results in ultrafast capture into confined electronic states. We have performed calculations reproducing the experimental conditions used in our experiments and a capture time of ~1.7 ps was obtained from our calculations.

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