Journal of Korean Vacuum Science & Technology; JKVST
Vol. 4, No. 1, March 2000, pp. 27~32

Phonon bottleneck effects of InAs quantum dots

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(Received March 24, 2000)

We have studied the carrier relaxation of InAs/GaAs modulation-doped quantum dots depending on the excitation wavelength and modulation-doping concentration by using the time-ressolved spectroscopy. At the excitation below GaAs barrier band gap, the relaxation processes become very slow, implying to observe the phonon bottleneck effects. On the other hand, at the excitation far above GaAs band gap, phonon bottleneck effects are broken down due to Auger processes. Increasing modulation-doping concentration, the relaxation times, by virtue of Coulomb scattering between electrons in GaAs doped layer and carriers in InAs quantum dots, are observed to become fast.

I. Introduction

In recent years, the optical properties of self-assembled quantum dots (SADs) which are formed by the Stranski-Krastanow growth mode have been extensively investigated [1-5]. It is expected that the SADs have the strongly enhanced oscillator strengths due to the three dimensional quantum confinement and a freedom of extrinsic fabrication defects compared with the quantum dots (QDs) produced by complicated processing techniques.

The device application of SADs such as lasers [6-8], modulators [9], and detectors [10] has been attempted. Several groups have demonstrated that SAD laser would have lower threshold currents and better temperature stability compared with quantum well (QW) laser. In QDs, however, the hot carrier relaxation has been theoretically expected to be reduced by the discrete energy levels (phonon bottleneck effect), which is not due to extrinsic fabrication defects but due to inevitable intrinsic effects [11,12]. The performance of optical device like QD laser might be limited by intrinsically low luminescence efficiency caused by phonon bottleneck effect.

On the other hand, the alternative relaxation mechanisms which make carriers efficiently relax toward the ground state have been proposed by several groups [13-15]. Bockelmann and Egeler [13] calculated the Coulomb scattering between an electron-hole plasma in two-dimensional region and a single electron in the zero-dimensional (0D) state, where a scattering event involved a transition between two different 0D states and and excitation of the 2D electron-hole plasma as Auger processes. The resulting transition rates were obtained to be of the order of 10^{12} s^{-1} for a plasma density of 10^{15} m^{-2} in InGaAs/InP QDs. Inoshita and Sakaki[14] investigated the electron relaxation in QDs with particaular emphasis on the role of the multiphonon processes. The relaxation time contributed by multiphonon (LO±LA phonons) processes was calculated to be of the order of subnanosecond around the longitudinal-optical phonon energy.

Several experimental papers [16-21] were investigated on the relaxation processes in QDs. To our knowledge, no definitive observation of phonon bottleneck effect in QDs has been reported. In this letter, we have investigated the relaxation processes in self-assembled InAs/GaAs modulation-doped QDs (MDQDs) using time-resolved spectroscopy. As measuring the decay times of the excited states depending on various excitation energies and modulation-doping concentrations, we report the observation and breakdown of phonon bottleneck effects in MDQDs.

II. Experiments

The MDQD samples grown by MBE growth tech-

nique were formed as the following sequence [22]. An undoped 0.5 mm GaAs buffer layer was initially grown on the semi-insulating GaAs substrate. InAs QDs were grown during the deposition time corresponding to 2.5 monolayer (ML) thickness, and covered with 10 nm GaAs layer and 50 nm GaAs:Si doped layer, where the growth rate and growth temperature of InAs QDs are 0.086 ML/sec and 420°C, respectively. The Si concentration of the doped layer was varied with 1×10^{17}, 5×10^{17}, and 1×10^{18} cm^{-3}. The formation of the QDs in our samples was confirmed by the transmission electron microscope (TEM) measurements. We established that the average diameter and density of the QDs are about 16 nm and 1.3×10^{11} cm^{-2}, respectively.

The samples were mounted on a closed-cycle liquid-helium cryostat for low-temperature PL and TRPL measurements. As an excitation sources were employed a picosecond dye laser synchronously pumped by a mode-locked Ar-ion laser which has about 2 ps pulse width at 3.8 MHz repetition rate and a femtosecond Ti:Sapphire laser pumped by cw Ar-ion laser which has about 100 fs pulse width at 82 MHz repetition rate for PL and TRPL measurements. The PL and TRPL signals were dispersed by a 1-m monochromator and detected by a cooled Ge diode and photomultiplier tube (Hamamatsu, R406). The carrier lifetimes of MDQDs were measured with a time-correlated single photon counting system, which gives us about 100 ps time resolution after deconvolution.

III. Results and discussion

Fig. 1 shows the PL spectra of MDQD with modulation-doping concentration of 1×10^{17} cm^{-3} by using a 600 nm picosecond dye laser (the pulse width of 5 ps) and a 830 nm femtosecond Ti:Saphhire laser (the pulse width of 100 fs) as an excitation source. To minimize the thermalization effect at high-intensity excitation, a pulse laser instead of a continuous wave laser was used as an excitation source. In the case of 600 nm excitation, the ground-state and excited-state emissions in MDQDs are seen around 1.245 eV and 1.330 eV, respectively. Our previous report [22,23] have identified the origin of the each emission by virtue of the time-resolved spectroscopy. Decreasing the excitation intensity, the peak low energy and

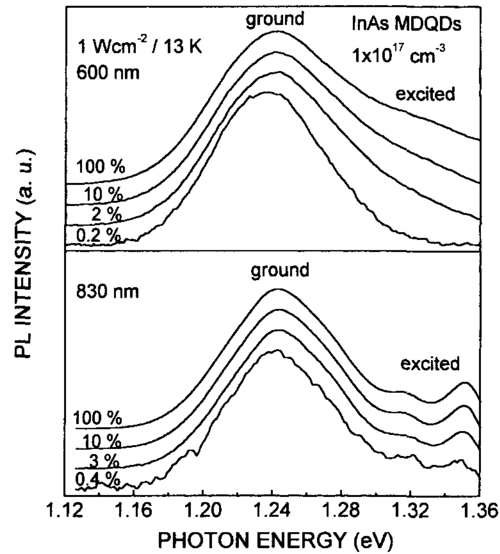

Fig. 1. The PL spectra of 1×10^{17} cm^{-3} MDQDs depending on the excitation intensities at 13 K by using a 600 nm picosecond dye laser (upper) and a 830 nm femtosecond Ti:Sapphire laser (lower).

excited-state emission rapidly disappears. On the other hand, when the sample is excited by 830 nm source, we can see a little shift of the ground-state emission peak at low intensity as well as a smaller full width at half maximum (FWHM) compared with the groun-state emission with 600 nm excitation source. While in 600 nm excitation, the electron-hole pairs are generated to GaAs barrier, those generated to InAs QDs in 830 nm excitation. The interaction between photo-generated carriers in GaAs barrier and InAs QDs vanishes in 800 nm excitation and thermalization and band renormalization effects are reduced [22,24], resulting in a little shift and the small FWHM of the ground-state emission in MDQDs. The excited-state transition in 830 nm excitation are very different from that in 600 nm excitation. First of all, PL spectra of the excited-state transition is shown to be split into two emission peaks at 1.318 eV and 1.352 eV. Secondly, the excited-state emission keep at the lowest excitation intensity. This is a main topic in this paper. We will discuss in detail with the time-resolved PL spectra later. To make clear the reasion that the excited-state transition is split into two emission peak, we carried out cw pump-probe measurements (Fig. 2(a)) the PL measurements depending on various excitation

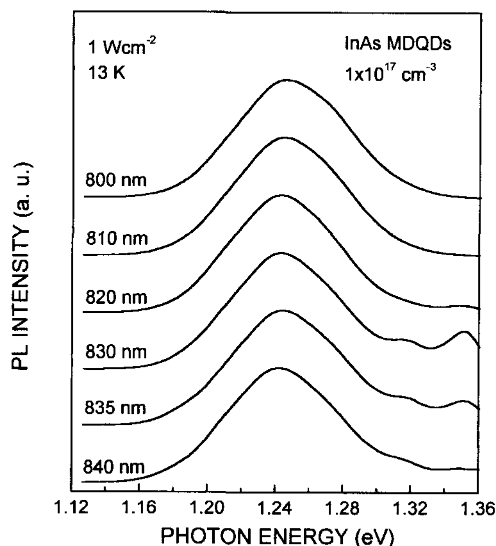

Fig. 2. (a) The cw pump-probe spectra of 1×10^{17} cm^{-3} MDQDs by using cw Ar^+ laser (pump) and tungsten-halogen lamp (probe) at 13 K. (b) The PL spectra of 1×10^{17} cm^{-3} MDQDs depending on the excitation wavelengths at 13 K by using the femtosecond Ti:Sapphire laser.

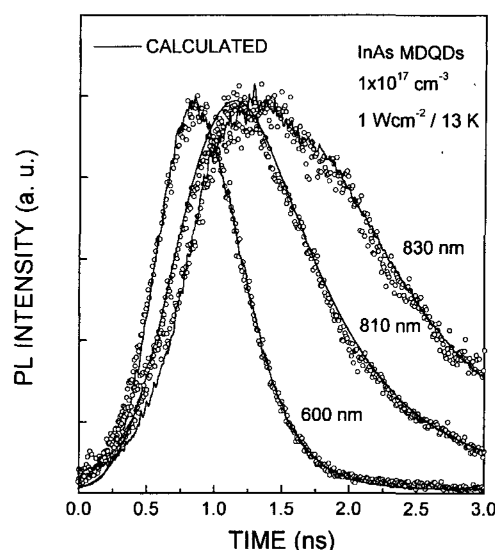

Fig. 3. The transient profiles of the excited-state PL spectra in 1×10^{17} cm^{-3} MDQDs depending on the excitation wavelengths. The open circle and solid line are the experimental data and calculation with deconvolution method, respectively.

wavelengths (Fig. 2(b)). As show in Fig. 2(a), the induced absorption spectra are obseved from about 825 nm, attributed to the band gap renormalization of GaAs barrier due to electron-hole pairs generated by pump beam (514.5 nm). From the PL spectra as a function of excitation wavelength in Fig. 2(b), it is observed that the splitting emissions of excited states appear at 820 nm excitation and have maximum intensity at 830 nm excitation, and are reduced again from 835 nm excitation This results from the resonant properties of excited-states for 830 nm excitation. It is possible to expain as the followings: (1) Before 830 nm excitation, the electron excited state is not resonant but very close to GaAs conduction band tail because of band bending effect due to modulation doping. (2) After 830 nm excitation, the electron excited state is resonant to GaAs conduction band tail, because the 830 nm pulse line has a FWHM of 20 nm in spectral region, and band gap renormalization a little affect on GaAs barrier, resulting from a few photo-created electron-hole pairs in GaAs barrier. (3) The electrons that exist in GaAs band tail are coupled with those of excited-states in QDs. The degenerated excited states in QDs are split by coupling interaction, and resonant tunneling of electrons from GaAs band tail to InAs QDs increase luminescence efficiency of excited states. For the excitation below or above 830 nm, there disappears the resonance between electrons in GaAs band tail and excited state of InAs QDs, because of no renormalization (below) and large renomalization (above).

Fig. 3 shows the time profiles of the excited-state PL spectra as a function of excitation wavelength. The open circles and solid lines indicate the experimental data and calculation, respectively. The calculation is given by deconvolution technech,

$$I'(t)=\int_{-\infty}^{\infty}P(t-\tau)I(\tau)d\tau \quad (1)$$

where $I'(t)$ is the detected signal, $P(t)$ the response function of the instrument, and $I(t)$ the luminescence signal. By an exponential decay law, $I(t)$ is given by

$$I(t)=\frac{A}{\tau_l}e^{-t/\tau_l}-\frac{B}{\tau_r}e^{-t/\tau_r} \quad (2)$$

where τ_l and τ_r are the lifetime and rise time, respectively. Substituting Eq. (2) for Eq. (1), the calculated curve and the parameters (A, B, τ_l, and τ_r) are obtained by the best numerical fit.

In Fig. 3, the rise time of the excited states in

MDQDs is observed to be shorter at 600 nm excitation than at 810 nm or 830 nm excitation, contrary to expectation that at the higher energy is excited the sample, the longer is the rise time. This means that the relaxation of the photo-generated carriers from the GaAs continuum band to the excited states in InAs QDs is faster than that from the states in InAs wetting layer to the excited states in InAs QDs. The life time also increase at the longer wavelength excitation, consistent with the PL spectra depending on the excitation intensity for the 600 nm and 830 nm excitation shown in Fig. 1.

Fig. 4 shows the lifetimes of the excited states in MDQDs depending on the excitation wavelength for the modulation-doping concentrations of 1×10^{17} and $1\times10^{18}\,cm^{-3}$. As shown in lifetime profiles of Fig. 3, for both of samples, the lifetime at the 830 nm excitation is twice or three times as long as that at the 600 nm excitation, where the PL spectra of the $1\times10^{18}\,cm^{-3}$ MDQDs is similar to those in Fig. 1. For the $1\times10^{17}\,cm^{-3}$ MDQDs, the life time is obtained to be about 800 ps with the 830 nm excitation. The lifetime (τ_l) of the excited state is determined by

$$\tau_l=(\tau_r^{-1} + \tau_d^{-1})^{-1} \quad (3)$$

where τ_r isthe relaxation time to the ground state and τ_d the decay time of the excited state caused by the radiative and nonradiative processes. Under the assumption that the decay time of the excited state emission is similar to that (840 ps) [23] of the ground state [17], the relaxation time from the excited state to the ground state is obtained to be about 17 ns. This result is in good agreement with the theoretical estimation that LA phonon limited electron relaxation times increase to tens of nanoseconds below a critical 3D confinement [11,12]. As a result, it is an evidence of observation of phonon bottleneck effect.

In the case of the 600 nm excitation, the relaxation time to ground state is obtained to be about 300 ps through the same calculation above. The value of this relaxation time is smaller than that for the 830 nm excitation by two orders of magnitude. The electron-hole pairs are generated in GaAs continuum band for the 600 nm excitation different from a little generation of them for 830 nm excitation.

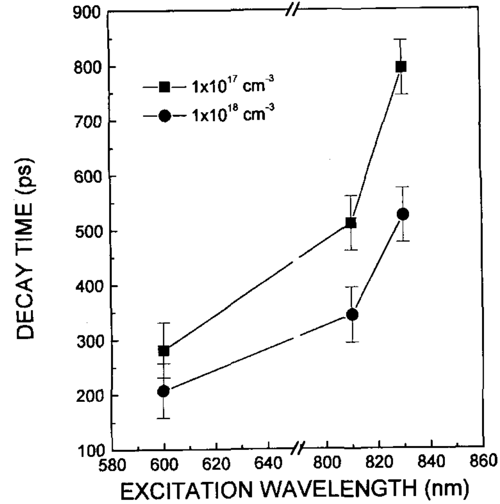

Fig. 4. The excited-state lifetimes of $1\times10^{17}\,cm^{-3}$ (square) and $1\times10^{18}\,cm^{-3}$ (circle) MDQDs as a function of excitation wavelength. The solid lines are a guide to the eye.

Auger processes which involve a transition between the excited and ground states in InAs QDs and a deexcitation of the electron-hole pairs in GaAs continuum band [13] are possible to play an important part in relaxation processes to ground state. The concentration of 600 nm photo-generated electron-hole pairs is obtained to be in range from 10 [11] to 10 [12] cm^{-2}. By the calculation of Auger process in Ref. 13, the relaxation time of the sample corresponding to the concentration of our photo-generated carriers is given to tens of picosecond. This is faster than our experimental results by an order of magnitude. We guess that this difference results that the calculation of Ref. 13, however, considered only electron Auger processes. Thus, our results are demonstrated to first observe a breakdown of phonon bottleneck effects expected sysmetically.

The lifetimes of both of samples at 810 nm excitation are slower (faster) than those at 600 nm (830 nm) excitation in Fig. 4. It is supposed that the electron-hole pair densities in GaAs continuum band at the 810 nm excitation are less than that at 600 nm excitation because the capture time into the InAs QDs at the 830 nm excitation is faster compared with at the 600 nm excitation. Therefore, the probobility of the Coulomb interaction between the electron-hole pairs in GaAs continuum band and carriers in InAs QDs would be reduced.

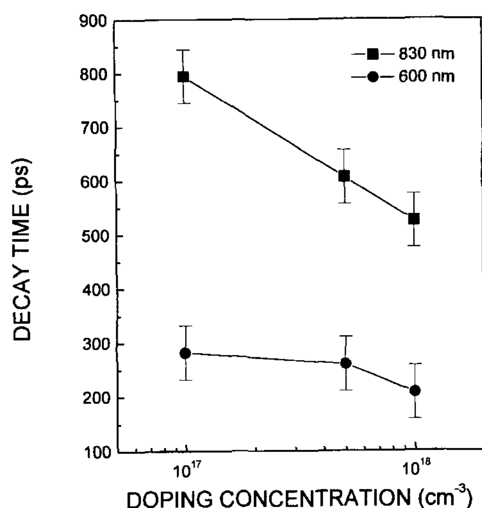

Fig. 5. The excited-state lifetimes of MDQDs at the 830 nm (square) and 600 nm (circle) excitation as a function of the modulation-doping concentration. The solid lines are a guide to the eye.

Fig. 5 shows the lifetimes of the excited states in MDQDs depending on the modulation-doping concentrations for the 600 nm and 800 nm excitations. For the 600 nm excitation, there is no variation of the lifetimes as a function of modulation-doping concentration within the experiments errors (±50 ps). For the 830 nm excitations, however, increasing the modulation-doping concentration, the lifetimes become fast. While 600 nm laser line creates electron-hole pairs in GaAs continuum band as many as the modulation-doping concentration, 830 nm laser line creats a little electron-hole pairs. As a results, the Coulomb interaction between the electrons in GaAs modulation-doping layer and the carriers in InAs QDs can be explained to make the relaxation to ground states be fast, when the samples are excited by the 830 nm laser line.

IV. Conclusion

We have investigated the carrier relaxation of InAs/GaAs MDQDs by using time-resolved spectroscopy. The transient profiles of the excited-state PL spectra in MDQDs show that the relaxation process become very slow to the relaxation time of about 17 ns. This is in good agreement with the theoretical relaxation times limited by LA phonon in QDs, implying to observe the phonon bottleneck effects. At 600 nm excitation, however, the relaxation process is shown to be very fast to about 300 ps. This results that the Auger processes, the Coulomb scattering between photo-generated electron-hole pairs in GaAs continuum band and carriers in InAs QDs, would break down the phonon bottleneck effect. The transient profiles of the excited-state PL spectra depending on modulation-doped concentrations, show that the Coulomb scattering between electrons in modulation-doped layer and carrier in InAs QDs plays an important role in the relaxation process at the excitation below the GaAs barrier band gap.

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