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The photoluminescence decay time of self-assembled InAs quantum dots covered by InGaAs layers

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Abstract
The temperature dependence of the time-resolved photoluminescence (PL) of self-assembled InAs quantum dots (QDs) with InGaAs covering layers was investigated. The PL decay time increases with temperature from 50 to 170 K, and then decreases as the temperature increases further above 170 K. A model based on the phonon-assisted transition between the QD ground state and the continuum state is used to explain the temperature dependence of the PL decay time. This result suggests that the continuum states are important in the carrier capture in self-assembled InAs QDs.

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

Self-assembled quantum dots (QDs) grown by the Stranski–Krastanov method provide peculiar optical properties such as high quantum yield, tunability, and thermal stability due to the δ-like density of states and strong quantum confinement. Because of these optical properties, the self-assembled QDs have led to considerable application success, in optoelectronic devices such as optical switches, light-emitting diodes, and lasers [1–4]. Self-assembled InAs/GaAs QDs on GaAs substrate are known to emit a photoluminescence (PL) peak with a wavelength of typically around 1050 nm [5]. The development of GaAs-based materials that emit at the telecommunication wavelengths of around 1.3 and 1.55 μm has attracted considerable interest. Recently many techniques have been proposed for extending the emission wavelength of InAs/GaAs QDs to 1.3 μm, such as cycled submonolayer InAs/GaAs deposition [6] and overgrowth of InAs/GaAs QDs with an InGaAs thin layer [7, 8]. The latter approach results in a redshift of the QD emission due to a reduction of the residual compressive strain, the strain-driven decomposition of the InGaAs layer, and the lowering of the lateral confinement (barrier lowering) [9–13]. Although a 1.3 μm InAs/GaAs laser with low threshold current density has been achieved, few investigations have been performed on the PL decay time of the InAs/GaAs QDs covered by InGaAs layers. The PL decay time of the InAs/GaAs QDs not only offers a convenient way to clarify their structures, but also provides useful information for extending their applications to optical devices. For example, it provides important information for improving the quantum efficiency of emission and for designing such optoelectronic devices as high speed laser sources and photodetectors.

A concept that is associated with continuum states in the self-assembled QDs has recently been proposed to account for some unexpected phenomena in PL [14–18]. Toda *et al* first suggested a 2D-like continuous state of a wetting layer to explain the broadening of transition lines in the single-dot

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[6] Author to whom any correspondence should be addressed.

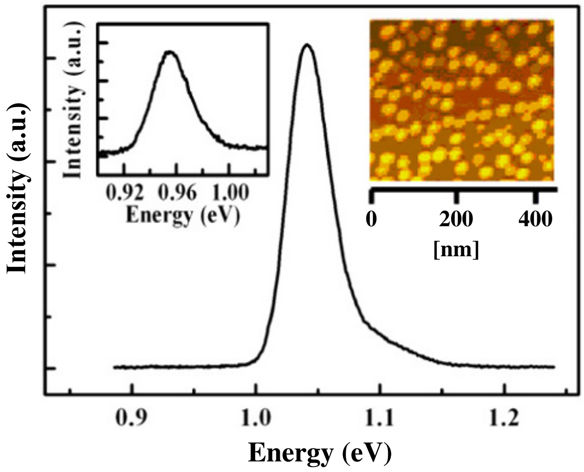

Figure 1. PL spectrum of the InAs QDs with the InGaAs layers at 20 K. The right inset shows an atomic force microscopy (AFM) image of uncapped QDs. The left inset displays the room temperature PL spectrum of the InAs QDs with the InGaAs layers.

photoluminescence excitation (PLE) [14]. Vasanelli *et al* indicated that QDs present an intrinsic deep continuous density of electron–hole states, and that these states lead to the crossed transitions between the bound QD state and delocalized states [15]. Urbaszek *et al* observed a drastic increase of the ground state transition for doubly charged excitons from 4 to 30 K, and ascribed this phenomenon to the coupling of the p-like excited state to the low energy tail of continuum states [16]. Bogaart *et al* demonstrated that carrier relaxation from the barrier layer toward the ground state is through continuum states, initially bypassing the excited states, followed by a single LO phonon emission [17].

In this report, the temperature dependence of the time-resolved PL properties of the self-assembled InAs QDs covered by InGaAs layers was investigated. The dependence of PL decay time on temperature is explained by a model based on the transition that involves the QD ground state and the continuum states through phonon-assisted interaction. We suggest that the continuum states in self-assembled InAs QDs play an important role in the carrier capture processes.

The structures investigated in this work were grown by solid source molecular beam epitaxy (SS MBE) on n$^+$-GaAs(100) substrates in a Riber Epineat machine. The structure of the InAs/GaAs QDs consists of a $Al_{0.3}Ga_{0.7}As$ bottom cladding layer, ten stacks of an InAs/InGaAs/GaAs QDs active region with a 30 nm thick GaAs barrier layer, a 30 nm thick AlAs top cladding layer, and a 10 nm thick GaAs cap layer. QDs were formed by the deposition of 2.6 MLs of InAs at a substrate temperature of 485 °C, and then covering with a 5 nm thick $In_{0.15}Ga_{0.85}As$ quantum well and a 5 nm thick GaAs barrier layer at the same temperature while the remaining layers were grown at 600 °C. The growth rate was 0.086 ML s^{-1} for InAs in QD formation, obtaining from the change of a streaky to a spotty reflection high energy electron diffraction (RHEED) pattern. The samples were mounted in a cold-finger cryostat and the sample temperature can be tuned between 10 and 300 K. Time integrated PL and PL decay measurements were made using pulsed diode lasers operating at the wavelength of 635 nm. The diode laser produces light pulses with about 40 ps duration and a repetition rate of 1 MHz. The collected luminescence was detected with a high speed photomultiplier tube (PMT). PL decay signals were measured using the technique of time-correlated single-photon counting (TCSPC) by a PC plug-in time-correlated counting card. The overall temporal resolution is about 250 ps.

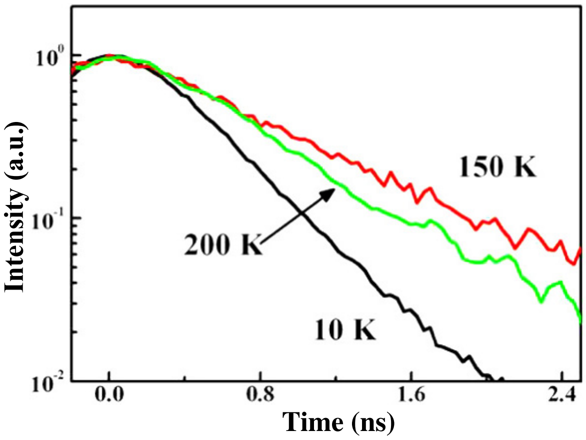

Figure 2. PL decay profiles of the InAs QDs with the InGaAs layers at various temperatures.

The right inset in figure 1 shows a plan-view atomic force microscopy (AFM) image of uncapped QDs. The average diameter of the dots is about 18 nm, revealing the high material quality and QD uniformity of the sample investigated. Figure 1 displays the 20 K PL spectra of InAs QDs with InGaAs layers. The energy of the main peak is centred at 1.038 eV, which is assigned to the ground state transition. The high energy shoulder is attributed to the recombination from the first excited state transition. The room temperature PL spectrum of this sample is displayed in the left inset of figure 1, exhibiting an emission peak wavelength (energy) near 1.3 μm (0.954 eV). Figure 2 displays the PL decay detected at the maxima of the ground state transition at different temperatures. All of the decay profiles in figure 2 reveal mono-exponential decay. The PL decay profiles of QDs were fitted to a single-exponential function of the form $A_0 e^{-\frac{t}{\tau_0}}$ to extract the decay time τ_0. The open circles in figure 3 display that the effect of temperature on $\tau_0 \cdot \tau_0$ increases with temperature from 50 to 170 K, and then decreases as temperature increases further above. Such behaviour has been observed experimentally in previous studies [19, 20] and is explained by a coupled rate-equation model, which includes the effects of carrier redistribution and retrapping at high temperatures [19]. In the following paragraph we will explain the observed temperature dependence of τ_0 using a model associated with the continuum states.

In order to demonstrate the existence of continuum states in our self-assembled QDs, we performed PLE measurements. Figure 4 shows the PLE spectrum of the ground state transition, displayed with respect to the excess excitation energy ($\Delta E = E_{\mathrm{exc}} - E_{\mathrm{det}}$). Starting at around $\Delta E = \sim$100 meV, an absorption background was gradually increased up to the wetting layer (WL) band edge. This absorption background originates from the low energy tail of the continuum states associated with WLs [16]. The continuum states can couple with the excited states of QDs, producing the

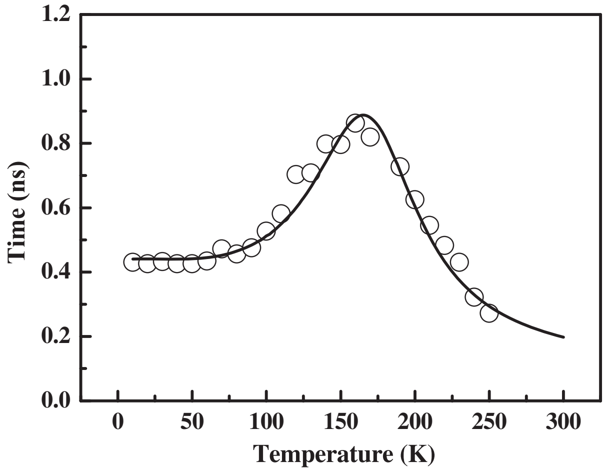

Figure 3. Temperature dependence of the PL decay times (τ_0) in InAs QDs with the InGaAs layers (open circles). The solid line shows the theoretical fit based on a model that incorporates the QD ground state and the continuum states through a phonon-assisted interaction.

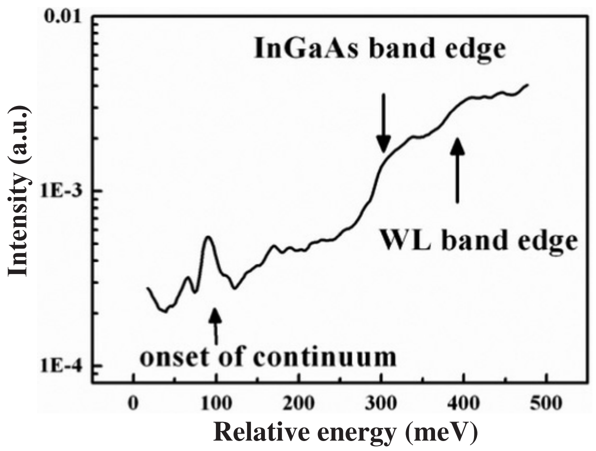

Figure 4. PLE spectrum of InAs QDs detected at the ground state transition at 20 K.

bound-to-bound, the continuum-to-continuum and the bound-to-continuum (continuum-to-bound) transitions [15]. The absorption resonances in the ΔE range between 50 and 300 meV are attributed to the transitions from bound states and delocalized states. The peaks positioned at $\Delta E = 305$ and 405 meV are assigned to absorption of the InGaAs and WL, respectively, as marked by the arrows in figure 4.

As shown in figure 5, this study proposes that an electron–hole pair in the ground QD state is scattered into the continuum states through a phonon-mediated interaction. This transition is based on the fact that any carrier transfer through the continuum states requires coupling—either Coulomb or phonon coupling—between the confined QD state and the continuum states [15]. The phonons provide additional energy for the phonon-assisted transition during scattering, increasing the probability of a carrier capture to continuum states. Therefore, the carrier is mostly captured by the continuum states, not the first excited state in QDs. Once the carriers have emitted to the continuum states, the carrier can either re-emit phonon back into the QD ground state or be scattered into the band edge of the wetting layers, where carriers are lost irreversibly.

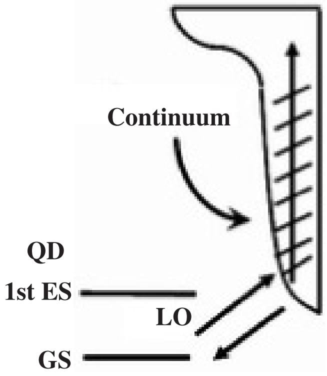

Figure 5. A schematic description of the model for carrier capture, involving the QD ground state and the continuum states in a phonon-assisted interaction.

According to the above model, the temperature dependence of τ_0 is analysed quantitatively. For a fixed temperature, the carrier density N in the ground state of QDs is given by a simple rate equation:

$$\frac{\mathrm{d}N}{\mathrm{d}t} = -\frac{N}{\tau_\mathrm{r}} - \frac{N}{\tau_\mathrm{op}} + \frac{\gamma(T)N}{\tau_\mathrm{op}}, \tag{1}$$

where τ_r is the radiative lifetime; τ_op is the scattering lifetime of the phonon-assisted interaction, and $\gamma(T)$ describes the thermally activated emission from the continuum background to the WL band edge, where carriers are consumed and lost irreversibly. The phonon scattering rate $1/\tau_\mathrm{op}$ is determined by

$$\frac{1}{\tau_\mathrm{op}} \propto [N_\mathrm{LO}(T)]^n, \tag{2}$$

with

$$N_\mathrm{LO}(T) = \frac{1}{\exp\left(\frac{E_\mathrm{LO}}{kT}\right) - 1}, \tag{3}$$

where $N_\mathrm{LO}(T)$ is the Bose–Einstein distribution function for LO phonons; E_LO is the LO phonon energy; k is the Boltzmann constant, and n is the number of phonons involved in the transition. $\gamma(T)$ can be described by

$$\gamma(T) = \frac{q}{1 + p \times \exp\left(\frac{-E}{kT}\right)}, \tag{4}$$

where E is the activation energy and the coefficients p and q measure the strength of the carrier-escape mechanism. The activation energy E is determined from the temperature dependence of PL intensity. The open circles in figure 6 show the integrated PL intensities as a function of the inverse of temperature. We extract the corresponding activation energy by using the following equation:

$$A(T) = A_0/[1 + C\exp(-E/kT)], \tag{5}$$

where A_0 is a constant; E is the thermal activation energy, and C is a fitting constant. The thermal activation energy for QDs is fitted as 118 meV. Equations (1)–(4) are used to fit the experimental temperature dependence of τ_0. For $n = 1$, $E_\mathrm{LO} = 32$ meV [21, 22] and $E = 118$ meV, the solid line in

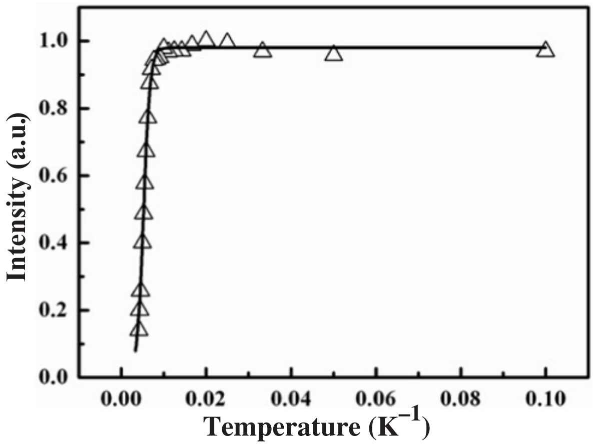

Figure 6. Temperature dependence of integrated PL intensities of the InAs QDs with the InGaAs layers. The solid lines are results calculated using equation (5), from which the thermal activation energies are determined.

figure 3 gives the theoretical fit, in close agreement with the experimental results. The good agreement indicates that the coupling of the continuum states with the ground state through the phonon-assisted interaction has an important role in the capture of carriers in InAs QDs.

In summary, the temperature dependent time-resolved PL of self-assembled InAs QDs with InGaAs layers is investigated. The PL decay time increases at temperatures from 50 to 170 K, and then decreases as temperature increases further above 170 K. This behaviour is well explained by a model that is based on a transition that involves the QD ground state and the continuum states via the phonon-assisted interaction, revealing that the continuum states in self-assembled InAs QDs play an important role in the carrier capture processes. This work is expected to be helpful in clarifying the carrier capture mechanism of self-assembled InAs QDs and may thus be useful in the further development of optoelectronic applications.

Acknowledgments

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