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Thermally activated carrier transfer among CdTe/ZnTe self-organized quantum dots

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Carrier transfer among CdTe/ZnTe self-organized quantum dots (QDs) was studied using time-resolved photoluminescence (PL) measurements. The authors have confirmed that carriers in the high energy ground states of small QDs transfer to the lower-energy ground states of larger QDs even at 10 K. The energy dependence of PL decay time changes uniquely with increasing temperature. They have found that the change in the energy dependence of PL decay time can be explained by thermally activated carrier transfer. © *2006 American Institute of Physics*.
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CdTe/ZnTe self-organized quantum dots (QDs) are expected to be applicable for visible light-emitting diodes or visible laser diodes because they exhibit yellowish-green luminescence. However, self-organized QDs have a size-fluctuation problem that results in a spectrally broad luminescence. This broadening spoils the original sharp density of states of the QDs. To improve the luminescence intensity, information on carrier dynamics is very important. We have reported that carriers can laterally transfer by quantum mechanical tunneling among InAs self-organized QDs.[1] In closely aligned QDs, since the wave functions of carriers in a QD penetrate into adjacent dots, photoexcited carriers tunnel and relax into larger and energetically lower QDs. In high-density QDs, carrier hopping and thermal-carrier escape are also reported for II-VI QDs.[2–4] In this letter, we report a time-resolved photoluminescence study revealing the thermally activated carrier transfer among CdTe/ZnTe self-organized QDs.

CdTe/ZnTe QD sample was grown on semi-insulating (100)-oriented GaAs substrates by molecular beam epitaxy (MBE) and atomic layer epitaxy (ALE).[5] It consists of the following structures: a 100-nm-thick undoped ZnTe capping layer deposited by MBE, 3.5-monolayer (ML)-thick undoped CdTe QDs deposited by ALE, and a 900-nm-thick undoped ZnTe buffer layer deposited by MBE. The depositions of the ZnTe layer and the CdTe QDs were carried out at a substrate temperature of 320 °C. The source temperatures of the Cd and Te sources for the CdTe QDs were 195 and 280 °C, respectively, and those of the Zn and Te sources for the ZnTe buffer and capping layers were 280 and 300 °C, respectively. One cycle for the ALE growth was carried out using an optimum growth process, in which the Cd effusion cell was opened for 8 s, the growth was interrupted for 1 s, the Te effusion cell was opened for 8 s, and the growth was interrupted for 5 s. This growth-interruption process for the ALE growth was introduced to improve the film quality by stabilizing the positive and the negative ions on the surface. The deposition of the CdTe QDs was carried out at a pressure of approximately 2.8×10^{-8} Torr.

An atomic force microscopy image taken after the deposition of the 3.5-ML-thick CdTe on a ZnTe buffer layer indicated that the average diameter of the CdTe QDs is 47 nm with a standard deviation of 11%. The density of the CdTe QDs is 1.3×10^{10} cm^{-2}. The average distance between a dot and its nearest neighbor is approximately 80 nm.

Time-resolved photoluminescence (PL) measurements were performed using a femtosecond Ti:sapphire laser. The excitation laser pulses were frequency-doubled to the photon energy of 3.26 eV ($\lambda=380$ nm), which is above the band gap of ZnTe. The time evolution of PL was measured using a streak camera (Hamamatsu C4334-04) with a time resolution of 15 ps. Figure 1(a) shows the time-integrated PL spectra 4.5 ns after photoexcitation. The PL peak has a full width at half maximum of 46 meV at 10 and 50 K, and broadens to 59 meV at 100 K. These PL distributions result from the sum of the many ground states of the electrons and holes whose respective energies depend on individual dot sizes.

Figure 2 shows the time evolution of PL at the high-energy side ($E=2.213-2.229$ eV) and low-energy side ($E=2.152-2.167$ eV) of the 2.19 eV PL peak at 10 K. The PL of the high (low)-energy side corresponds to the photoemission between the electron and hole ground states of small (large) QDs. As shown in Fig. 2, the rise and decay times of the high-energy side are shorter than those of the low-energy side. The rise times from 10% to 90% are 74 and 90 ps for the high-energy and low-energy sides, respectively. The decay times obtained from a single-exponential fit are 178 and 307 ps for the high-energy and low-energy sides, respectively. These relatively long rise and decay times of the large QDs prove the presence of carrier transfer from the small QDs to the larger QDs even at 10 K.

Basically, the recombination lifetime in each quantum dot is not sensitive to 11% size fluctuation as we showed for InAs QDs.[1] In Fig. 1(b), we plotted the PL decay times obtained from a single exponential fit. At 10 and 50 K, the PL

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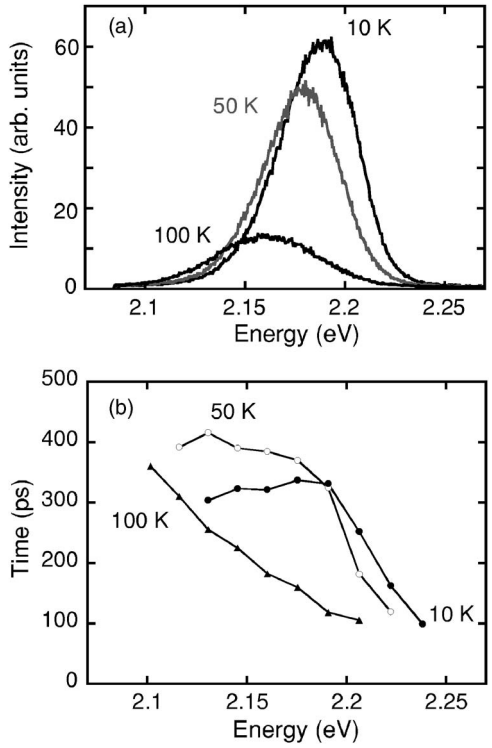

FIG. 1. (a) Time-integrated photoluminescence (PL) spectra during 4.5 ns after photoexcitation of CdTe/ZnTe QDs at 10, 50, and 100 K. (b) PL decay times evaluated by single-exponential fit.

decay time of the high-energy side becomes shorter for higher energy, although those of the low-energy side are almost constant. These uniform decay times of the low-energy side are a unique feature showing that the carriers do not transfer between these energy states. If carriers transfer between these energy states, the PL decay time would have an energy dependence. Therefore, we can conclude that the carriers transfer from the high-energy ground states which have the energy dependence in Fig. 1(b) to the low-energy ground states in the constant-decay-time region. Since only the high-energy side has the energy dependence at 10 and 50 K, this process is ascribed to a thermal process. At 50 K, the PL decay times of the constant-decay-time region become longer uniformly than those at 10 K. At 100 K, the constant-decay-time region disappears and all PL decay times become shorter for higher energy.

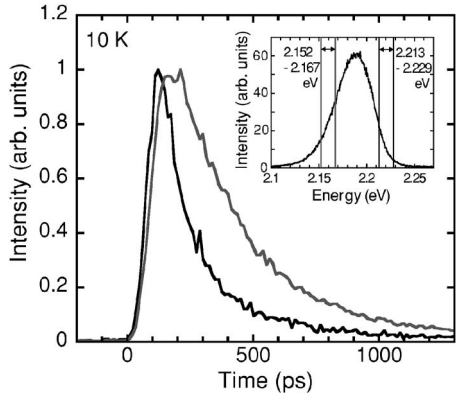

FIG. 2. Time evolutions of PL at the high-energy and low-energy sides of PL peak at 10 K. The black and gray curves represent the time evolutions at the high-energy (E=2.213–2.229 eV) and low-energy (E=2.152–2.167 eV) sides, respectively. The inset shows the corresponding PL spectrum.

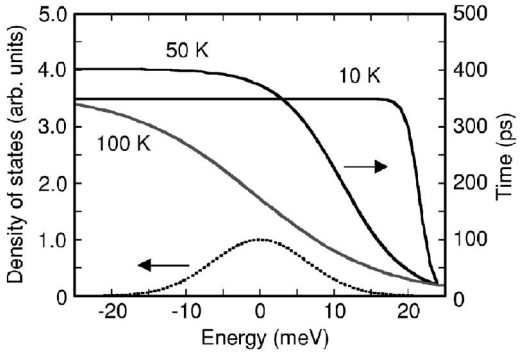

FIG. 3. Fitting results using rate equations assuming Gaussian-shaped density of states (dotted line) with a full width at half maximum of 15 meV. The recombination lifetimes τ_r are assumed to be 350, 400, and 350 ps at 10, 50, and 100 K, respectively.

To explain these interesting features, we propose a model consisting of thermal carrier escape and a redistribution process. The model assumes that the carriers escape thermally from dots to high-energy continuum states which spread spatially over all the QDs. The thermally excited carriers are presumed to move in the continuum states and relax into ground states in QDs. In this model, since the carrier escape is described by the Boltzmann distribution, $\exp(-(E_a-E_i)/k_BT)$, with an activation energy E_a, only the carriers in high-energy ground states can escape from the dots. Here, k_B and T are the Boltzmann factor and temperature, respectively. We assume that the carrier relaxation rate from the continuum states to the ground states in the QDs is proportional to the number of vacant ground states. In addition, we assume that the photoexcitation is very weak where the carrier occupation of each energy level is regarded to be very low. In this case, the rate equation for a quantum level E_i is written as

$$\frac{dN_i}{dt} = -\frac{N_i}{\tau_r} - \frac{N_i}{\tau_e} e^{-(E_a-E_i)/k_BT} + \frac{N_c}{\tau_d} D_i, \tag{1}$$

where τ_r, τ_e, τ_d, N_c, and D_i are the recombination lifetime, thermal escape time from the dots to the continuum states, relaxation time from the continuum states to the dot states, carrier population of the continuum states, and the density of the E_i state, respectively. The density of states is approximated to a Gaussian distribution with a full width at half maximum of 15 meV and is normalized to $\Sigma D_i=1$. For the initial condition, the carriers are assumed to be distributed over the energy levels in proportion to the Gaussian density of states. The candidates for the continuum states are the valence band states or a wetting layer's states. Thermally activated carrier transfer through a wetting layer was reported for InAs/GaAs QDs.[6] In CdTe/ZnTe QDs, the existence of a wetting layer has not been proven. Here, we take the valence band states as the continuum states because the valence band energy discontinuity, ΔE_v, is reported to be as small as 0.2 eV.[7] Since the observed inhomogeneous broadening of the spectrum of 46–50 meV consists of the distributions of electrons and holes, we assume that the energy distribution of hole states is 15 meV.

Figure 3 shows the fitting results assuming the activation energy of 25 meV. The parameters are τ_e=20 ps and τ_d =20 ps. Only the recombination lifetime τ_r is changed for

temperature. The τ_r values are 350, 400, and 350 ps at 10, 50, and 100 K, respectively. These curves explain well the experimental results, regardless of the simplicity of this model. With increasing temperature, the constant-decay-time region decreases because the number of energy states from which carriers can escape thermally increases. At 100 K, the carriers in all dots can escape to the continuum states.

From the application's point of view, the carrier escape from quantum dots is not preferable, because the luminescence efficiency of quantum dots decreases. To improve the luminescence intensity at higher temperature, we need further investigations about the carrier dynamics.

In summary, carrier transfer among CdTe/ZnTe self-organized QDs was studied using time-resolved photoluminescence measurements. We have confirmed that carriers in high-energy states transfer to lower-energy states even at 10 K. The energy dependence of the photoluminescence decay time changes uniquely at different temperatures. We have found that the change in the energy dependence of the PL decay time can be explained by thermally activated carrier transfer.

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