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Photoluminescence study of carrier transfer among vertically aligned double-stacked InAs/GaAs quantum dot layers

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Photoluminescence (PL) properties of self-organized quantum dots (QDs) in a vertically aligned double-layer InAs/GaAs QD structure are studied as a function of temperature from 10 to 290 K. The QDs in a sample with a 1.8 ML InAs seed layer and a second 2.4 ML InAs layer are found to self-organize in pairs of unequal sized QDs with clearly discernible ground-states transition energy. The unusual temperature behavior of the PL for such asymmetrical QD pairs provides clear evidence for carrier transfer from smaller to larger QDs by means of a nonresonant multiphonon-assisted tunneling process in the case of interlayer transfer and through carrier thermal emission and recapture within one layer. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1510157]

The effect of temperature on both energy relaxation and carrier transfer mechanisms in semiconductor quantum dots (QDs) has been a subject of extensive investigations.[1–6] In general, it is reported that, as expected, the QD photoluminescence (PL) intensity decreases with increasing temperature due to carrier escape from the dot.[3] However, these investigations also report, rather unexpectingly, a redshift of the PL peak position and a decreasing PL linewidth with increasing temperature.[1,4–6] This unusual and interesting behavior has been explained by enhanced carrier relaxation between QDs due to several reasons including carrier thermionic emission,[3] carrier transport through the wetting layer (WL),[2] and tunneling mechanisms.[1] The effect of temperature can get even more interesting when the QD array exhibits a size distribution that shows more than one maximum, e.g., a bi- *or* multimodal QD size distribution either within one layer[7–10] or across multiple layers.[11,12] Despite such interesting possibilities and significant potential applications a complete picture of the energy and carrier transfer in such multimodal systems is still not available.[7,10]

In this letter we present a detailed study of the carrier transfer between two InAs QD families with different size distribution but separated from each other by a thin layer of GaAs. The particular QD system under investigation is a vertically aligned double-layer InAs/GaAs QD structure with different sized QDs in the first layer compared to the second layer.

Our samples were fabricated using a solid-source molecular beam epitaxy chamber coupled to an ultrahigh vacuum scanning tunneling microscope (STM). The growth structure consists of two InAs layers containing QDs, which was repeated eight times, in a GaAs matrix. All samples were grown on GaAs (100) substrates, followed by a 0.5 μm GaAs buffer layer and 10 min annealing at 580 °C to provide a nearly defect free atomically flat surface. The first QD layer was then added by depositing 1.8 ML of InAs with a growth rate of 0.1 ML/s, an As_4 partial pressure of 8×10^{-6} Torr, and a substrate temperature of 500 °C. This was followed with 16 nm of GaAs deposited on top of the first QD layer while the growth temperature was changed from 500 to 520 °C. The second QD layer was then added by depositing 2.4 ML of InAs. The resulting samples are vertically correlated double-layer QD structures with different QD sizes in each layer.[13,14] The substrate temperature was then reduced from 520 to 500 °C during a 40 nm GaAs growth, which was used to separate the pair of QD layers from seven additional pairs.[15] As seen by STM, the dot density in the bottom layer of the pair is about 4.5×10^{10} cm^{-2} while the density in the second layer is about 2.5×10^{10} cm^{-2}. Meanwhile, the top islands are nearly double the size of the bottom islands due to the additional deposition and higher growth temperature.

The PL was excited by the 514.5 nm line of a continuous wave Ar^+ laser. We applied excitation densities in the range 0.01–20 W/cm^2. The samples were mounted in a close-cycle cryostat, which allows measurements in the temperature range from 10 to 300 K. The PL signal was detected with a LN_2 cooled Ge photodiode using phase-sensitive detection.

Figure 1 shows the low-temperature PL spectrum from the 8× double-layer sample A. Also shown in the same figure are the PL spectra of two reference samples B and C containing multiple layers of only one of the two InAs QD double-layers, i.e., either sample B (2.4 ML) or sample C (1.8 ML). For samples B and C, the main PL peak can be fitted by a single Gaussian, indicating that the observed dot formation has only one dominant size. Sample B shows a single PL peak at an energy of 1.16 eV with the full width at half maximum (FWHM) of 50 meV while for sample C, the PL peak is at 1.27 eV with FWHM ~120 meV. These data are in agreement with expected values for the given growth conditions.[7]

The PL spectrum from the double-layer stacked InAs/GaAs QDs (sample A) shows a pronounced double-peak structure. This can be attributed to the total contribution in PL signal from QD ensembles of both layers. Indeed, a line

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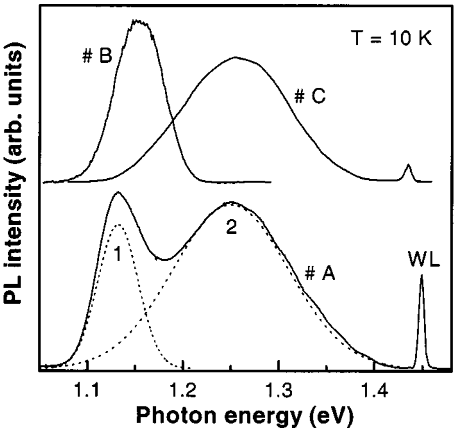

FIG. 1. Low temperature PL spectra for 1.8/2.4 ML double-layer sample A and single layer reference samples B (2.4 ML) and C (1.8 ML). A line shape analysis of the spectra proves that the QD PL signal from sample A is a convolution of two Gaussian-shaped peaks as shown by dotted lines.

shape analysis shows that the PL signal of sample A is well reproduced by a convolution of two Gaussian-shaped peaks and a third contribution arising from the WL. The higher-energy peak 2 (see Fig. 1) is correlated to the PL spectrum from sample C and the lower-energy peak 1 to sample B, despite the fact that it is narrower and redshifted (~30 meV). Hence, it is reasonable to assume that the peak at higher energy originates from the dots in the seed layer (1.8 ML) while the low-energy peak originates from dots in the second layer (2.4 ML).

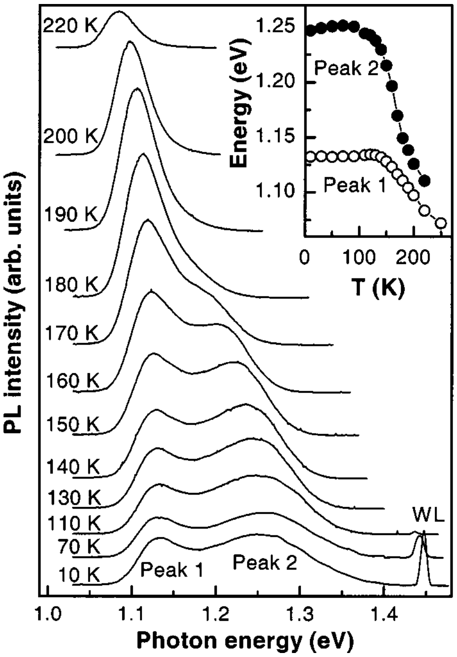

FIG. 2. PL spectra of sample A at different temperatures. The inset shows the temperature dependence of the two PL peaks energies.

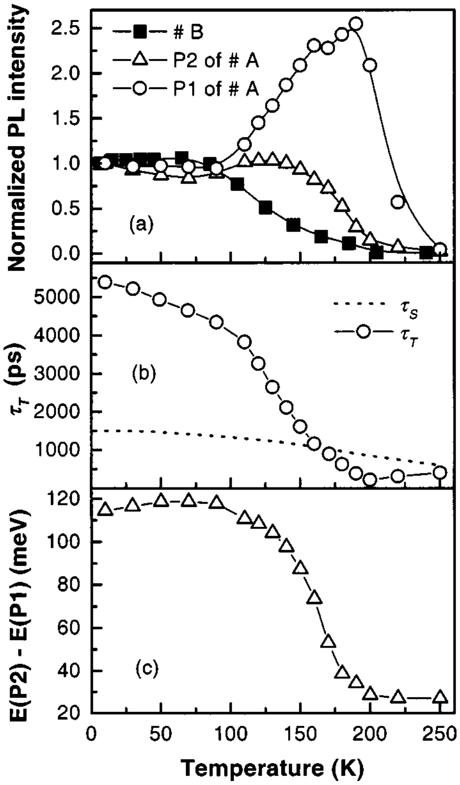

FIG. 3. Temperature dependence (a) of the normalized integrated PL intensity of sample B as well as peaks 1 and 2 from sample A; (b) estimated from Ref. 8 radiative ground state lifetime τ_S of seed layer and interlayer carrier transfer time τ_T; (c) energy difference between PL peak positions from QDs of the seed and second layers.

Figure 2 plots the temperature dependence of the PL emission from sample A at an excitation intensity of 1 W/cm^2. There are clearly two different temperature-dependent regions. At low temperatures (10–80 K), the spectra are quite stable and exhibit no significant changes. Such behavior of PL spectra is characteristic for excitonic recombination.[1,7] Dramatic changes in the PL spectrum occur, however, in the temperature range of 100–200 K. Beginning at around 110 K peak 2 (small dots) experiences a strong redshift of about 2 meV/K (see inset of Fig. 2). Simultaneously with the shift, a slight increase of the integrated PL intensity in the temperature interval of 110–160 K was observed with a subsequent decrease as the temperature is raised to 250 K. Such a strong redshift of the PL signal in an InAs/GaAs QD system has not been observed before. The slight increase of peak 2 in the range of 100–150 K is a real effect. It is due to a transfer of carriers with a short lifetime in small dots of the seed layer to states with a long lifetime in large dots of the seed layer,[8] resulting in a higher equilibrium density of excited QDs. Over the same temperature range the behavior of peak 1 (large dots) is basically different. The spectral redshift starting at $T\sim150$ K is much smaller than for peak 2 while the integrated PL intensity begins to grow dramatically with increasing temperature above 120 K. As shown in Fig. 3(a), peak 1 reaches a maximum at 190–200 K and then drops rapidly between 200 and 250 K.

We propose an explanation of the observed temperature behavior based on the interlayer carrier transfer between dots by means of nonresonant multiphonon-assisted tunneling processes (NPTP). These processes are ineffective at low temperature but become efficient at temperatures higher than 100 K. At the same time, based on previous investigations of stacked QDs,[16] we believe that the quality and thickness of the GaAs barrier layers limits the role of thermionic emission in explaining our observations of the double-layer QD structure in the 100–200 K range. Figure 4 gives a schematic conduction-band energy diagram for the double-layer structure of sample A for the case of low and high temperatures,

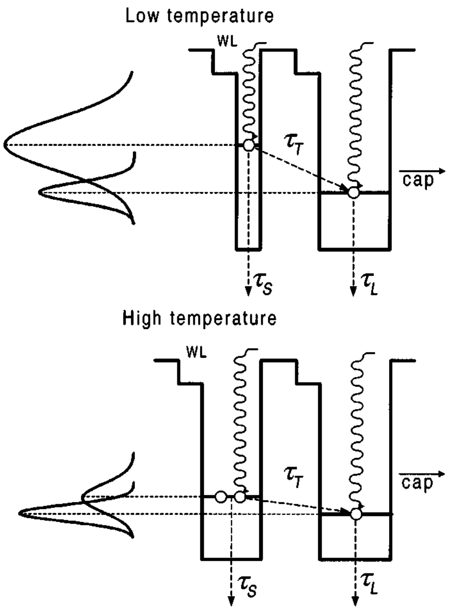

FIG. 4. Schematic conduction-band energy diagrams for the double-layer QD structure (sample A) in case of low and high temperatures.

respectively. According to previous investigations,[5,8,11] one can suggest a time hierarchy of carrier relaxation processes. The fastest times are for intralayer and intradot carrier relaxation τ_I, followed by interlayer carrier transfer time τ_T, and finally the radiative ground state lifetime of seed layer dots τ_S and second layer dots τ_L. For low temperatures a ground-state lifetime (τ_S) of ~1500 ps has been determined for the 1.8 ML single layer.[8] Following Ref. 11, the yield (η_T) of excitation transfer from small dots of the seed layer to large dots of the second layer may be determined by $\eta_T = \tau_S/(\tau_S + \tau_T)$ and estimated from the ratio of the integrated PL intensity of peaks 1 and 2 (Fig. 2). For $T=10$ K the estimated τ_T is ~6000 ps. This large value of τ_T in comparison with ground-state lifetime τ_S is in a good agreement with results in Ref. 11. Figure 3(b) depicts τ_T derived using η_T and utilizing the $\tau_S(T)$ estimated from Ref. 8. One can see that τ_T sharply decreases starting at 100 K and becomes smaller than the ground-state lifetime τ_S at about 150 K indicating that the process of interlayer carrier transfer begins to play a more essential role and becomes dominant at $T \geqslant 160$ K. It should be noted, that the estimated temperature dependence of τ_T gives only an upper limit, since we did not take into account nonradiative recombination channels, which can be activated at high temperatures.

The physical reason for a greater interlayer carrier transfer at temperatures higher than 120 K is that NPTP is becoming increasingly more resonant. Indeed, at low temperature [Fig. 3(c)] the average energy difference ΔE between ground states of dots from the seed and second layers is about 120 meV. Without going into the nature of actual transfer mechanism (which is not clear yet), one can emphasize that at least four LO phonons ($E_{\mathrm{LO,InAs}} \sim 29$ meV)[15] are required. With increasing temperature, however, the carrier thermal energy in the ground states of the small QDs becomes comparable to the exciton binding energy, so that the exciton dissociates into free carriers into the WL and GaAs barrier and then relaxes into a different dot. Carrier hopping between dots favors a transfer of carriers to the dots having a higher binding energy, and hence, a lower emission energy, resulting in a narrowing and spectral redshift of the PL spectrum. Thus, with elevating temperature carrier redistribution takes place and for high temperatures most carriers are in the ground states of the largest dots (see Fig. 4). For example, in the temperature range of 180–220 K [Fig. 3(c)], ΔE is only ~30 meV, a value which is close to the LO phonon energy. In this case, the yield of NPTP must be significantly higher.

In conclusion, we have uncovered a more complete picture of the energy and carrier transfer in multimodal and high-density QD systems, which will provide measures for greater control of PL behaviors.

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