Energy Transfer within Ultralow Density Twin InAs Quantum Dots Grown by Droplet Epitaxy

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ABSTRACT Ultralow density (~10^6/cm^2) of twin InAs quantum dot (QD) hybrid structure was grown by a droplet epitaxy technique. The photoluminescence (PL) from ensemble and individual twin InAs QD structures showed a bimodal behavior and an energy transfer between the well-separated (~190 nm) twin QDs, which was supposedly due to the special wetting ring that built the channel for exciton transfer. This research demonstrates a novel approach to fabricate lateral InAs QD pairs as the candidate for a laterally coupled QD molecule.

KEYWORDS: twin InAs quantum dots · quantum dot molecule · photoluminescence · energy transfer · coupling

Self-assembled semiconductor quantum dots (QDs) are the subject of increasing interest due to their potential in developing a new generation of optoelectronic devices.[1−5] As an example, a group of closely spaced QDs can act as a "QD molecule" (QDM), which is interesting, both as a new playground for studying interacting electronic systems and as a building block to perform complex quantum computing operations.[6−9] The simplest QDM is composed of two interacting QDs. A well-developed technique to fabricate such QDM is the epitaxial growth of a vertically aligned QD pair,[10−12] in which the coupling between two QDs can be tuned by the spacer thickness and external fields.[13,14] Another approach to obtain QDMs is to fabricate a laterally coupled QD pair. Since self-assembled QDs are typically characterized by a flat geometry, the nature of the lateral coupling may differ appreciably from that of vertical coupling, rendering the study of laterally coupled QDMs of fundamental interest.[15,16] However, the stochastic nature of self-assembly growth results in the random distribution of InAs QDs on GaAs surfaces, presenting an obstacle for fabrication of laterally coupled QDMs. Only recently, Schmidt *et al.* invented a special fabrication technique allowing control of InAs lateral QDMs on a GaAs surface decorated with nanoholes.[17] Also, Wang *et al.* and Kuguchi *et al.* obtained GaAs lateral QD pairs with the help of droplet epitaxy growth.[18,19] In this paper, ultralow density (~10^6 cm^{-2}) lateral QD pair hybrid structures, consisting of twin InAs QDs, were developed based on the droplet epitaxy technique. The optical properties of twin InAs QDs were investigated by photoluminescence (PL) measurements. The results showed an energy transfer between the twin QDs, which support the twin InAs QD hybrid structure as a candidate for laterally coupled QDM.

RESULTS AND DISCUSSION

The twin InAs QD hybrid structures were grown on semi-insulating GaAs (100) substrate by a solid source molecular beam epitaxy (MBE) system. After oxide desorption and the growth of a 0.5 μm GaAs buffer layer at 600 °C, the substrate was cooled to 350 °C and the As valve was fully closed before the substrate stabilized at the desired temperature. Once the growth temperature was reached, an indium flux equivalent to form one monolayer (ML) InAs at 0.1 ML/s was supplied to the GaAs surface to form indium droplets. In order to crystallize the indium droplets into InAs nanocrystals, the sample was kept at 350 °C for the first annealing of 120 s with the As valve 5% open (arsenic beam equivalent pressure ~ 0.8×10^{-6} Torr). Then with the As valve fully opened (arsenic beam equivalent pressure ~ 7.0×10^{-6} Torr), the sample endured the second annealing of 120 s. After that, the achieved QD morphology was quenched and atomic force microscopy (AFM) characterization was carried out on the sample in air. To grow the sample for PL measurement, after the second annealing, the sample was capped with 10 nm GaAs at

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Received for review April 16, 2008 and accepted October 01, 2008.

Published online October 14, 2008.
10.1021/nn800224p CCC: $40.75

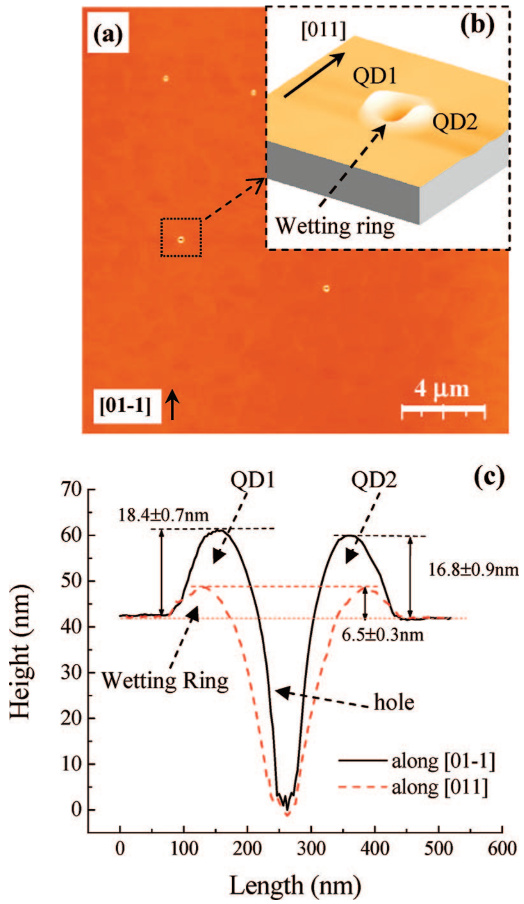

Figure 1. (a) 20 μm × 20 μm AFM image of the twin InAs QDs sample; (b) magnified three-dimensional AFM image showing a typical twin InAs QD structure given by the dashed line rectangle in panel (a); (c) line profiles along [011] and [01−1] directions of the twin InAs QDs structure given in panel (b).

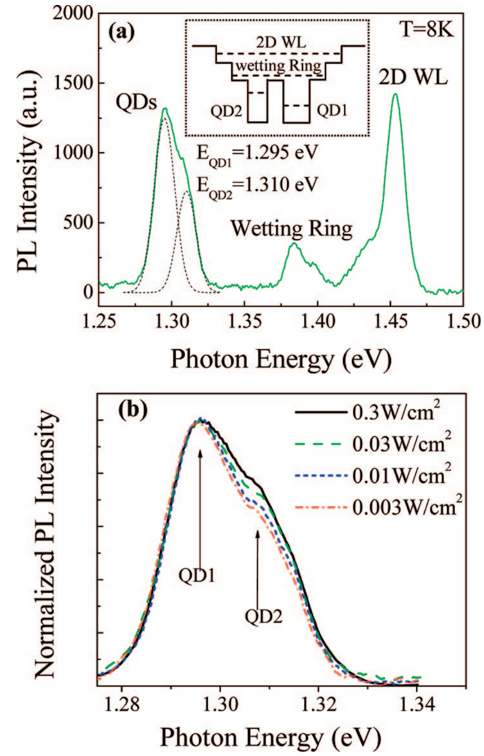

Figure 2. (a) PL spectrum of the twin InAs QD ensembles measured at 8 K using a YAG laser (532 nm) with an excitation intensity of 0.1 W/cm^2; the inset gives a conduction band schematic corresponding to the twin InAs QDs. (b) Normalized QD PL spectra as a function of excitation laser intensity.

350 °C and followed by another 100 nm GaAs cap layer at 600 °C. AFM image showed a smooth sample surface after this capping growth.

Figure 1a shows a 20 μm × 20 μm AFM image of the twin InAs QDs. The density of the QD structures was ~10^6 cm^{-2}, 2 orders of magnitude lower than previously reported low density InAs QDs achieved by droplet epitaxy on nanoholes.[20] In order to clearly see the morphologic features, Figure 1b presents a magnified AFM image of a single hybrid structure, and Figure 1c gives the line profiles of this structure along the [011] and [01−1] directions. It can be seen that, after the growth, every indium droplet turned into a hybrid structure with twin QDs sitting on a wetting ring base of a height of 6.5 ± 0.3nm. The separation between the tips of the twin QDs was about 190 nm. The twin QDs were about 16.8 ± 0.9 and 18.4 ± 0.7 nm above the sample surface. Meanwhile, each hybrid structure also had a hole in the center as deep as 40 nm. The formation of such deep holes during the droplet epitaxy growth has been attributed to the nanodrill effect under indium-rich conditions.[21] Therefore, the growth dynamic was complicated, and it combined several processes including indium droplet ripening, As desorption, indium droplet crystallization, InAs diffusion, and indium and gallium intermixing. We suppose that the formation of twin InAs QDs was mainly due to the anisotropic migration of indium atoms along the [011] and [01−1] directions during the crystallization of the indium droplet.[21−23] The resulting hybrid structure of twin InAs QDs is substantially interesting, which provides a new method to obtain an ultralow density lateral QD pair for the studying InAs QDM.

Figure 2a shows a PL spectrum of the twin InAs QD ensembles measured at 8 K using a YAG laser (532 nm) with an excitation intensity of 0.1 W/cm^2; a conduction band schematic corresponding to the twin InAs QDs is also given in the inset. Three different peaks could be identified in the PL spectrum. The peak at 1.45 eV originated from the two-dimensional InAs wetting layer (2D WL) on the sample surface, and its relatively strong intensity was due to the ultralow density of the QDs. The peak around 1.38 eV was attributed to the emission from the wetting ring shown in Figure 1, and it may be regarded as the one-dimensional wetting layer (1D WL) of the InAs QDs. The emission around 1.29 eV was from the twin InAs QDs. The very high signal-to-noise ratio at such low excitation intensity showed that the twin InAs QDs were rather good quality nanocrystals. Clearly, the twin QDs have the PL emission at a wavelength shorter than typical self-assembled InAs QDs grown by MBE (~1.1 μm). This is likely due to the strong Ga, In intermixing during the droplet epitaxy growth of the twin QD hybrid structures.[20,24] Meanwhile, as shown by the Gaussian fitting results, the emission from the twin InAs QDs exhibited a bimodal distribution behavior. The energy separation between the bimodal PL peaks was about 15 meV, which is much less than the typical energy separation between the ground state and the excited states of InAs QDs.[25] Therefore, we believe the bimodal PL was not due to the excited states of the InAs QDs but from bimodal QD size distribution. The AFM shown in Figure 1 proved that the twin InAs QD hybrid structures had anisotropic properties and the heights of the twin InAs QDs in each hybrid structure were not identical. As a result, the InAs QD ensemble had bimodal behavior in their height distribution and subsequently their PL spectrum. The conduction band schematic given in the inset of Figure 1a indicated that the wetting ring reduced the height of the potential barrier between the twin InAs QDs, so the carrier may have more possibility of transfer between the twin QD. While the QD PL spectra were investigated with variation of the excitation laser intensity, Figure 2b gives the normalized QD PL spectra in low pump regime to avoid an excited state effect. Interestingly, we noticed a clear increase of PL with pump intensity at the high energy side of the PL band corresponding to small QDs. This can be regarded as a result in terms of energy transfer processes between the bimodal of QDs with different size.[26,27]

In micro-PL measurements of a single structure of twin InAs QDs, the sample was excited at 4 K using a Ti:sapphire laser emitting at 720 nm. The laser beam was focused onto the sample by a lens with a focus spot of ~30 μm diameter. The PL was collected by a 50× objective lens and recorded with a CCD camera. Due to the extremely low QD density, no shadow mask or patterning was required for spectroscopy of single twin QDs. Shown in Figure 3a is a PL image of the sample. This image revealed bright spots, and each spot originated from the individual structure of the twin InAs QDs. The number density of the twin InAs QDs observed from Figure 3a was ~10^6 cm^{-2}, which was consistent with the result obtained from the AFM image in Figure 1a. One twin InAs QD, as shown in Figure 3b, was randomly selected to study the micro-PL spectroscopy. Its PL image showed clearly the GaAs substrate PL at 1.52 eV, the 2D wetting layer signal at 1.45 eV, and the wetting ring emission at about 1.37 eV, as presented in Figure 3c. The wetting ring emission was localized only in the area of the twin InAs QD structures. However, the PL signal from the 2D wetting layer covered almost the whole sample surface, which indicated that the 2D wetting layer was likely formed due to InAs diffusion from the droplets during the crystallization and annealing process. Also, as shown in Figure 3d,e, two groups of sharp luminescent lines appeared at approximately

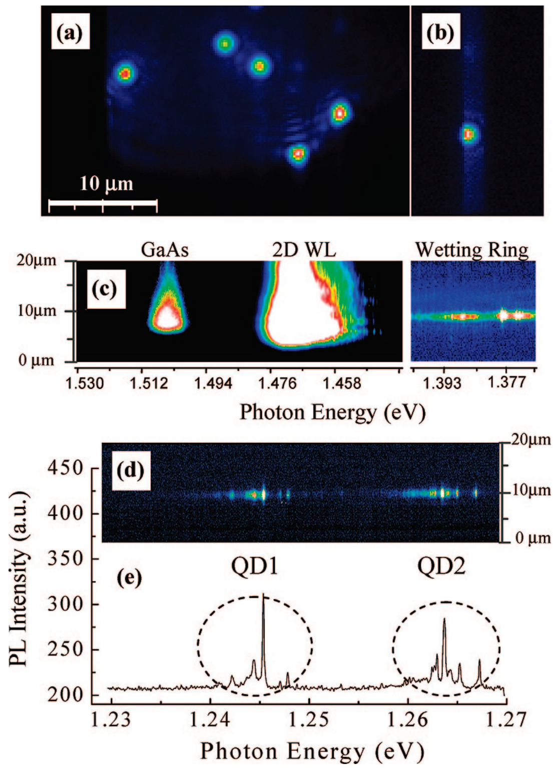

Figure 3. (a) PL image of the twin InAs QD sample, in which each bright spot came from a single structure of the twin InAs QDs; (b) one representative twin InAs QD was randomly picked up for the study of micro-PL spectroscopy; (a) and (b) were obtained by using a 900 nm long pass filter so that only the optical emissions from QDs can be seen. The *x*-axis/*y*-axis corresponds to the spatial positions cut/along the spectrometer slit. (c) PL images from the GaAs substrate, 2D WL, and 1D wetting ring; (d) PL image of the representative twin InAs QDs; in (c) and (d), the *y*-axis corresponds to the spatial position along the spectrometer slit, while the *x*-axis denotes photon energy after the PL signal was dispersed by the spectrometer. (e) PL spectrum extracted from panel (d) for the representative twin InAs QDs.

1.246 and 1.263 eV, respectively. These were attributed to the twin QDs in the investigated individual hybrid structure. The energy separation between these two groups of luminescent lines was ~17 meV, which agreed with the result obtained from Figure 2a. These PL features were found to be similar for other twin InAs QDs.

Then PL spectra obtained at 4 K from the representative twin InAs QDs are presented in Figure 4a for varying excitation intensity. As the excitation intensity increased, the spectra showed two sets of peaks, corresponding to the emission of two InAs QDs, highlighting the consistency of the observed behavior. For QD1, the sharp peak at 1246 meV can be attributed to ground state exciton X1 recombination. As evidenced by its excitation intensity dependence, at very low pump intensity, the X1 peak is dominant. With a concomitant rise of the pump intensity, some other peaks appeared, which have been documented due to multiexciton and charged exciton recombination.[28] The similar behavior was observed in QD2, and the sharp peak

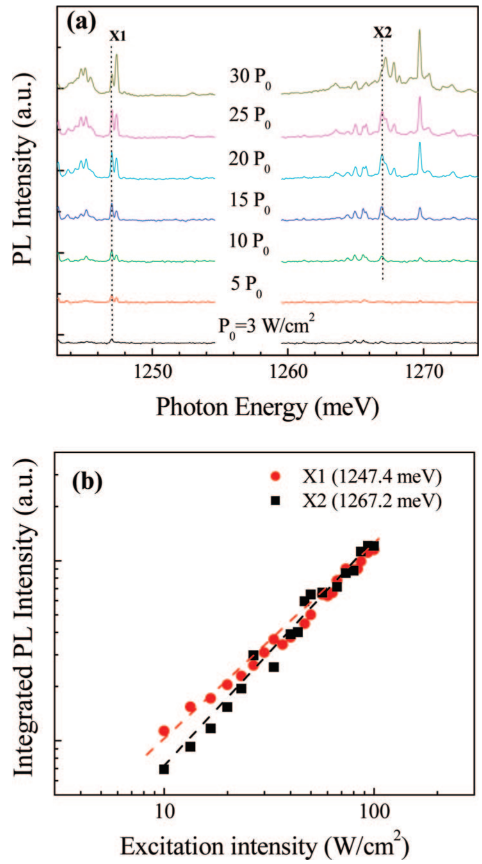

Figure 4. (a) Micro-PL spectra from the representative twin InAs QDs obtained at 4 K with different excitation intensity; (b) the integrated intensities of exciton (X1, X2) PL peaks of the representative twin QDs as a function of excitation intensity.

at 1269 meV can be attributed to the ground state exciton X2 recombination. The integrated intensities of exciton (X1, X2) peaks of the twin QDs are plotted in Figure 4b as a function of excitation intensity. At low excitation intensity, approximately linear dependence was observed for X1 and X2 peaks.[29,30]

One interesting feature observed in Figure 4a is that the sharp PL peaks from the bigger QD1 appeared with smaller excitation intensity than those from the smaller QD2. This filling effect was further confirmed while several other twin InAs QDs were randomly selected for micro-PL measurement. As the focus point of the excitation laser was about several tens of micrometers in diameter, which was much bigger than the diameter of the twin InAs QDs, the twin InAs QDs received identical excitation laser intensity and inhomogeneous excitation effect was excluded. Meanwhile, at the low temperature of 4 K, the thermal energy was too small to activate and transfer the carrier from small QDs into large QDs. Generally, for two isolated QDs at 4 K, the excitons occupy the QDs statistically and the exciton PL should have the same dependence on the laser excitation intensity. Therefore, the observations in Figure 2b and Figure 4a indicated that a carrier transfer or a certain cross-talk existed between the twin InAs QDs.

A number of possibilities exist to interpret this cross-talk phenomenon. First, it is possible that the observed filling effect in Figure 4a between the twin QDs is due to spectral variation in the carrier relaxation from the wetting ring into the QDs. If this was the case, however, one would expect that the PL intensity ratio of the two QDs does not change with the variation of the pump intensity before their PL emissions reach saturation. Second, the two most common mechanisms for energy transfer between two neighbor QDs is the short-range carrier tunneling due to quantum coupling and the long-range transfer of exciton energy due to the Förster coupling.[31−33] However, from the AFM shown in Figure 1, the center-to-center separation between the twin QDs was measured to be about 190 nm. For such a big separation, experimental and theoretical investigations have proved that both mechanisms for direct exciton transfer between the twin QDs are neglected because they require a tunneling/transfer time much longer than the exciton lifetime.[34,35] Therefore, we conclude that the nature of the lateral cross-talk of our twin InAs QD structures differs appreciably from the coupling of laterally/vertically aligned QDM. While a more systematic study of the carrier dynamic properties is out of the scope of this work, we speculate here that the cross-talk between the twin QDs is likely due to the special twin InAs QD hybrid structures with a wetting ring. The wetting ring may couple (quantum or dipole−dipole) with the twin QDs, respectively, and the subbands in the wetting ring build a channel for the carrier transfer between the twin QDs,[32] which provides the twin InAs QD hybrid structures with the potential as a laterally coupled QDM for information communication. These two QDs and the associated 1D wetting ring and 2D wetting layers compose a unique system for studying such processes as electron flow and thermal activation that could only be achieved on the ensemble level previously. For example, the filling effect through the 1D wetting layer has been achieved in QD-chain structures that consist of smaller and larger QDs associated with 1D wetting wires on the ensemble level.[36]

CONCLUSION

In summary, an ultralow density (~10^6/cm^2) QD structure was obtained by a droplet epitaxy technique. The AFM investigation indicated that, after the growth, each indium droplet formed a hybrid structure with twin InAs QDs sitting on a wetting ring. The ensemble PL showed a bimodal characteristic due to the anisotropic properties of hybrid structure and consequently the bimodal distribution of QD height. Remarkably, energy transfer between the bimodal of QDs was observed. The micro-PL investigation also indicated the existence of a filling effect between the two InAs QDs in an individual hybrid structure. These observations demonstrate that there is lateral cross-talk in each twin

QD. We suppose it is due to the special geometrical properties of the hybrid structure and the wetting ring forming a channel for exciton transfer, which provided this hybrid structure with potential as lateral-coupled QDM for information communication.

Acknowledgment. The authors gratefully acknowledge support of this work by NSF (DMR-0520550) and NSF (DMR-0210383 and DMR-0606485).

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