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PHOTOLUMINESCENCE STUDY OF InAs/GaAs SELF-ORGANIZED QUANTUM DOTS WITH BIMODAL SIZE DISTRIBUTION*

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We have investigated the temperature dependence of the photoluminescence (PL) spectrum of self-organized InAs/GaAs quantum dots. A distinctive double-peak feature of the PL spectra from quantum dots has been observed, and a bimodal distribution of dot sizes has also been confirmed by scanning tunneling microscopy image for uncapped sample. The power-dependent PL study demonstrates that the distinctive PL emission peaks are associated with the ground-state emission of islands in different size branches. The temperature-dependent PL study shows that the PL quenching temperature for different dot families is different. Due to lacking of the couple between quantum dots, an unusual temperature dependence of the linewidth and peak energy of the dot ensemble photoluminescence has not been observed. In addition, we have tuned the emission wavelength of InAs QDs to 1.3 μm at room temperature.

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I. INTRODUCTION

The possibility of the creation of new electronic devices based on self-assembled quantum dots (QDs) has stimulated a great interest in both theoretical and experimental investigations of the optical and electronic properties of the QDs.[1−8] The unique electronic properties of QDs rely on the delta-function-like energy dependence of the density of states due to the quantum confinement of carriers in all three dimensions. The strong confinement potential in QDs results in a high intensity of photoluminescence (PL), which is a useful technique for determining transition energies between electron and hole levels in QDs. The effect of temperature on the optical properties of self-organized QDs is of great importance for their use in devices working at room temperature. The temperature-dependence of PL emissions has been a subject of extensive studies for clarifying the mechanism of PL quenching processes in a randomly distributed dot structure.[9−14] The unusual dependence on temperature of both the peak energy position and the peak energy width of the PL bands has been reported. The fast redshift of PL energy and the anomalous decrease of linewidth with increasing temperature have been attributed to the efficient relaxation process of carriers resulting from the spread and penetration of the carrier wave functions in coupled InAs QDs.

In this paper, we investigate the temperature dependence of the PL emission spectrum of self-organized InAs/GaAs QDs. A distinctive and novel double-peak feature of PL spectra from QDs has been observed. We use exciton-intensity-dependent PL spectra to demonstrate that these double emission peaks are associated with the ground-state emission from islands in different size branches. The scanning tunnel microscopy (STM) image for the uncapped sample also shows a bimodal distribution of dot sizes. Furthermore, the temperature-dependent PL spectra show that the PL quenching temperature for different dot families is different.

II. EXPERIMENTS

The samples studied here were grown in a molecular beam epitaxy (MBE) system (VG V80H MK II) equipped with an *in situ* reflection high-energy electron diffraction (RHEED) system and coupled to an ultrahigh vacuum (UHV) scanning probe microscopy (SPM) facility (Park Scientific Instrument) via a gate valve. The substrates are n^+-GaAs (001) (Si-doped). After growing a GaAs buffer (500 nm) at 600°C and at 0.1 nm/s growth rate, the growth temperature was lowered to 500°C, and then 2.5 ML InAs was deposited on the GaAs buffer at a growth rate of 0.1 ML/s. Si is continually supplied during the growth with a concentration of $2\times10^{18}\,\mathrm{cm}^{-3}$. The RHEED patterns were

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monitored throughout deposition and used to determine the growth mode transition, which is characterized by a sudden appearance of transition electron diffraction spots in the RHEED pattern. The critical thickness of QD nucleation is about 1.7 ML in our experiments. As soon as the InAs layer growth process was completed, the samples were cooled down rapidly in order to preserve the surface morphology as much as possible. they were then transferred to the adjoining UHV SPM chamber for detailed STM imaging.

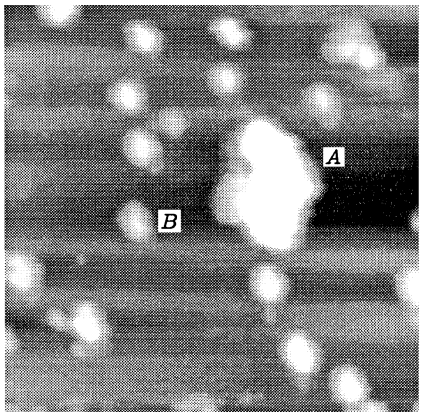
After STM imaging, the sample was transferred back to the MBE deposition chamber. Then the 50nm thick GaAs cap layer was deposited onto the sample at a growth temperature of 500°C. The purpose of using the same sample is to ensure that the results of STM and PL measurements arise from the same structure. Generally, samples are often grown with similar structures for SPM and other measurements. Then experimental results on different samples are corroborated with one another. This method is inappropriate because both the growth interruption and the slow quenching rates can change the surface morphology. Here, we use the same sample to avoid this problem.

PL measurements were performed in a variable-temperature(10–300K) closedcycle cryostat under the excitation of 632.8 nm line of a He-Ne laser. Signals from samples were detected by a cooled Ge detector.

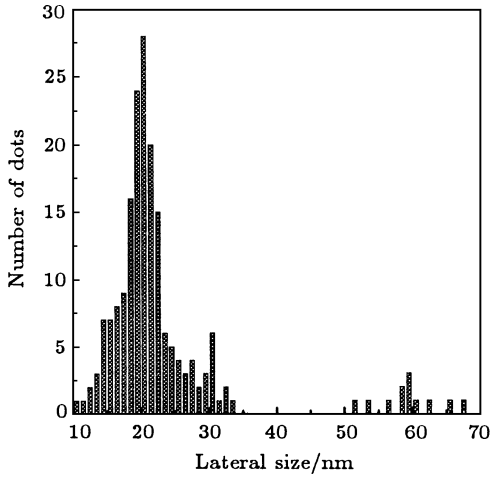
Ⅲ. RESULTS AND DISCUSSION

In Fig.1(a) we show the typical surface morphology of the uncapped sample with a nominal 2.5ML InAs by UHV-STM. The size of STM scans is 0.3μm$\times$ 0.3μm. A statistical lateral size distribution of QDs from the image has been obtained with the use of a public domain image processing software[15] (Scion Image Program). In Fig. 1 (b), we show the histogram of QDs with lateral size obtained from 1μm $\times$ 1μm STM images. There are two predominant families (A and B) of QDs. In family A with a density about 1.1×10^9 cm^{-2}, the average lateral size and height are approximately 60nm, and 9nm, respectively. In family B with a density about 1.9×10^{10} cm^{-2}, the average lateral size and height are approximately 22 nm, and 6 nm, respectively. The larger dots may result from small dots coalesced partly under our growth conditions. The unintentional annealing effect after depositing InAs for the STM measurement may be the cause that the InAs island bifurcates in our experiment.[16] By having two easily identifiable island size branches, the bifurcated island size sample overcomes the difficulty to associate island size with its physical properties in self-organized dots.

The PL spectra of the capped sample obtained with excitation intensity from 0.01 to 10 mW at a low temperature (10K) are shown in Fig.2. There are two peaks in all the PL spectra. The assignment of the two emission peaks to islands in different size branches is complicated due to the fact that multiple excited-state emission peaks are often observed on InAs/GaAs QD samples that emit in the same spectral range as our samples.[4] We have found that the PL intensities of the two peaks observed on our samples depend linearly on the excitation power density over three orders of magnitude, and that the PL intensity



(a) The size of STM scans is 0.3μm $\times$ 0.3μm



(b) Histogram of the lateral size is created from 1μm $\times$ 1μm STM images

Fig.1. STM image of the uncapped nominal 2.5 ML QD sample.

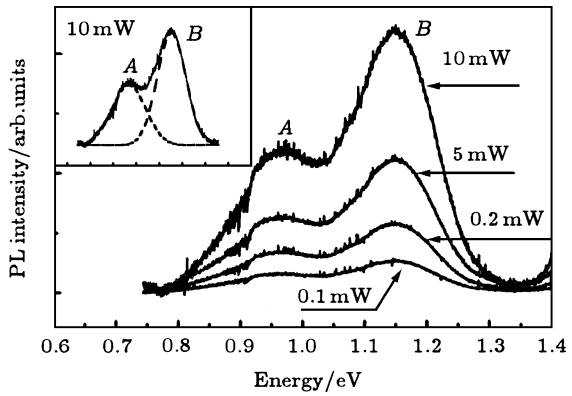


Fig.2. Photoluminescence spectra of the capped sample with different excitation power densities at 10K. The inset shows the Guassian fitting of each peak (A and B) with one predominant size in each dot family.

of the high-energy peak is stronger than that of the low-energy peak even at very low excitation intensity. The fact that two PL peaks can be seen at low exciton intensity suggests that they correspond to the ground-state transition from islands in different size branches. Furthermore, the intensity ratio of the ground-state emission from these two branches as a function of pumping power also shows that they are independent of each other. These results demonstrate that the high-energy peak cannot stem from the excited-state of InAs quantum dots.

The power-dependent PL study demonstrates that the double peaks are associated with the ground-state emission of islands in different size branches. The temperature dependence of the PL spectra further verifies this assignment. Figure 3 shows the PL spectra of the capped sample obtained with an excitation intensity of 10 mW at different temperatures. At low temperatures, there are distinctly two peaks in the PL spectra. Taking into account the quantum-size effect on the peak energy, we believe that the excitons localized in a family of smaller dots contribute to the high-energy peak, while the excitons localized in a family of larger dots contribute to the low-energy peak. The intensity of the high-energy peak is stronger than that of the low-energy peak, indicating that the density of smaller dots must be larger than that of larger dots. These results are in accordance with the STM image.

With the increase of temperature, the high-energy peak becomes weaker and weaker. They disappear at about 220K. However, the low-energy peak exists clearly up to 300K. It is commonly stated that the dot-luminescence intensity quenches with temperature due to carrier thermal escape from the dot ground states to the wetting layer and to the states of the matrix material, and/or nonradiative recombination centers. The larger dots have confined states at lower energies and the smaller dots at higher energies. With increasing temperature, excitons can overcome shallow energy minima by thermal activation and fall down into deep centers. Thus, the high-energy peak caused by the smaller dots will disappear at first.

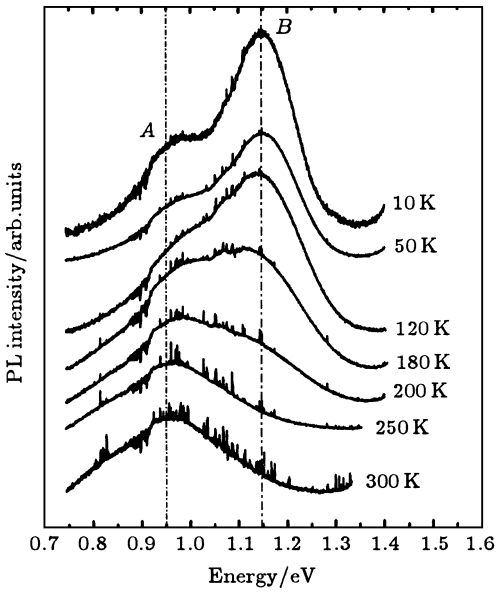


Fig.3. Photoluminescence spectra of the capped sample at different temperatures at exciton power of 10 mW.

At each temperature, every peak energy and the full width at half maximum (FWHM) can be fitted by single Gaussian shape, which suggests that one size predominates in each dot family. The energy position and the FWHM of each peak as functions of temperature are shown in Figs.4 and 5., respectively. The energy difference of two peaks is about 170 meV. It is caused by the difference of lateral size and height between two predominant families of QDs. In Refs. [3] and [7], detailed calculations of electronic structure of InAs/GaAs self-organized QDs were given, with which our experimental results are in good agreement. It should point out that the QD is a complicated strain system. The shape, size and distribution of QDs are quite different under different growth conditions. The theoretical calculation cannot give accurate results. In family B, the quantum dots have a lateral size distribution mainly from 15 to 25nm. The energy difference given by theoretical calculation[3,7] is about 120 meV, which approximately equals the FWHM that we obtained. In family A, the dots have both the large lateral size distribution (50–70nm) and height distribution (6–11nm), so peak A exhibits a larger FWHM

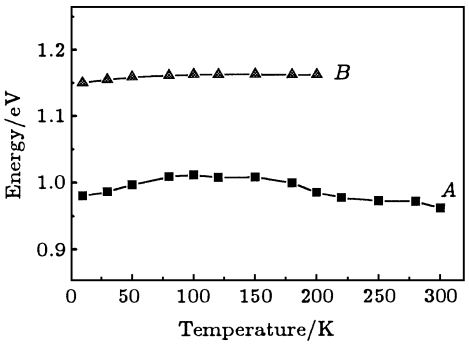


Fig.4. Temperature dependence of experimental peak energies from the PL spectra after deconvolution into Gaussian components for the capped sample.

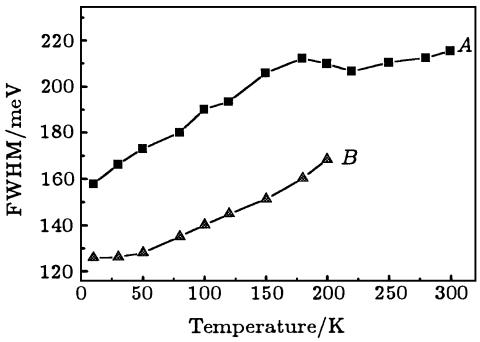


Fig.5. Temperature dependence of FWHM for each peak from the PL spectra after deconvolution into Gaussian components for the capped sample.

than Peak B. In addition,we notice that the high-energy peak exhibits a little blue shift and the low-energy peak exhibits first a blue shift and then red shift with increasing temperature. At low temperatures, the blue shift of peak position can be explained by the dislocalization of excitons and/or the band filling effect with increasing temperature. The red shift of peak position is caused by the shrinkage of bandgap of InAs with increasing temperature. Because the energy position of each peak is obtained by Gaussian fitting, the result would exist a certain error. The FWHM of both peaks increase with increasing temperature, which we will discuss in the following.

Our result here is in contrast with other experimental results, which show an unusual temperature dependence of the linewidth and peak energy of the dot ensemble photoluminescence.[9−14] In their reports, the fast redshift of PL energy and the anomalous decrease of linewidth with increasing temperature were attributed to the efficient relaxation process of carriers resulting from the spread and penetration of the carrier wave functions in coupled InAs QDs. At low temperatures, carriers are frozen randomly into the dot states and the PL spectrum reflects the absorption of the QD ensemble. With increasing temperature, carriers may be thermally activated outside the dot into the wetting layer and/or the GaAs barrier and then relax into a different dot. Carrier hopping between dots favors a drift of carriers toward the dots having a higher binding energy, and hence a lower energy emission, resulting in a narrowing of the PL spectrum followed by a luminescence peak redshift larger than that expected for the thermal shrinkage of the InAs band gap with increasing temperature. When the temperature is high enough, the electron-phonon scattering becomes dominant. The PL linewidth starts to increase with increasing temperature. These thermal effects are not expected in an ideal QD structure in which all the dots are decoupled from each other.

Now, we analyze the causes that make the differences between our results and others. From the STM image, we can see that the density of QDs (1.9×10^{10} cm^{-2}) is very low, which may be due to the high temperature growth and the unintentional post-growth annealing. The distances between vicinal QDs are mostly very large (more than 50 nm), which leads to the reduced tunneling between QDs. The efficient relaxation process of carriers resulting from the spread and penetration of the carrier wave functions in coupled InAs QDs has been cut off. Thus, we cannot observe the phenomenon of the narrowing of PL spectrum followed by a larger luminescence peak redshift due to carrier hopping between QDs. The FWHM of PL spectra increase with increasing temperature due to thermal distribution of carriers. It is shown that the tunneling between quantum dots plays a key role in unusual temperature dependence of QD photoluminescence.

In addition, the emission peak of PL spectra is located at 0.95 eV(1.3 μm) at room temperature (300 K) in Fig.3. So, the QDs offer the possibility for fabricating inexpensive 1.3 μm GaAs-based lasers and detectors suitable for low cost fiber interconnects. The unintentional annealing effect after depositing InAs for the STM measurement may lead to a long wavelength shift of the PL peak.

IV. CONCLUSION

In conclusion, we have presented the opical properties of self-organized InAs/GaAs QDs. A distinctive double-peak feature of the PL spectra from QDs has been observed, and a bimodal distribution of lateral sizes has also been confirmed by the STM image for the uncapped sample. The power-dependent PL

study demonstrates that the distinctive PL emission peaks are associated with the groundstate emission of islands in different size branches. Taking into account the quantum size effect on the peak energies, we ascertain that the high (low) energy peak results from the smaller (larger) dot family. The temperature-dependent PL study shows that the PL quenching temperature for different dot families is different. Due to lack of the couple among QDs, the unusual temperature dependence of the linewidth and peak energy of the dot ensemble photoluminescence has not been observed. In addition, we have tuned the emission wavelength of InAs QDs to 1.3 μm at room temperature.

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