Tuning the optical performance of surface quantum dots in InGaAs/GaAs hybrid structures

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Abstract: GaAs spacer thicknesses are varied to tune the coupling between InGaAs surface quantum dots (QDs) and multilayers of buried QDs. Temperature and excitation intensity dependence of the photoluminescence together with time resolved photoluminescence reveal that coupling between layers of QDs and consequently the optical properties of both the surface and the buried QDs significantly depend on the GaAs spacer. This work provides an experimental method to tune and control the optical performance of surface QDs.

OCIS codes: (160.6000) Semiconductors, including MQW; (300.6470) Spectroscopy, semiconductors

References and links

1. D. Bimberg, M. Grundmann, and N. N. Ledentsov, *Quantum Dot Heterostructures*, (John Wiley & Sons, New York/Chichester, 1998).
2. M. Bayer, P. Hawrylak, K. Hinzer, S. Fafard, M. Korkusinski, Z. R. Wasilewski, O. Stern, and A. Forchel, "Coupling and Entangling of Quantum States in QuantumDot Molecules," Science **291**, 451 (2001).
3. S. S. Li, J. B. Xia, J. L. Liu, F. H. Yang, Z. C. Niu, S. L. Feng, and H. Z. Zheng, "InAs/GaAs single-electron quantum dot qubit," J. Appl. Phys. **90**, 6151 (2001).
4. P. Bhattacharya, S. Ghosh, and A. D. Stiff-Roberts, "Quantum dot opto-electronic devices," Annu. Rev. Mater. Sci. **34**, 1 (2004).
5. N. T. Yeh, T. E. Nee, J. I. Chyi, T. M. Hsu, and C. C. Huang, "Matrix dependence of strain-induced wavelength shift in self-assembled InAs quantum-dot heterostructures," Appl. Phys. Lett. **76**, 1567 (2000).
6. F. Ferdos, S. M. Wang, Y. Q. Wei, A. Larsson, M. Sadeghi, and Q. X. Zhao, "Influence of a thin GaAs cap layer on structural and optical properties of InAs quantum dots," Appl. Phys. Lett. **81**, 1195 (2002).
7. Y. Nabetani, T. Matsumoto, G. Sasikala, and I. Suemune, "Theory of strain states in InAs quantum dots and dependence on their capping layers," J. Appl. Phys. **98**, 063502, 2005.
8. H. Saito, K. Nishi, and S. Sugou, "Influence of GaAs capping on the optical properties of InGaAs/GaAs surface quantum dots with 1.5 μm emission," Appl. Phys. Lett. **73**, 2742 (1998).
9. J. Z. Wang, Z. Yang, C. L. Yang, and Z.G. Wang, "Photoluminescence of InAs quantum dots grown on GaAs surface," Appl. Phys. Lett. **77**, 2837 (2000).
10. Z. F. Wei, S. J. Xu, R. F. Duan, Q. Li, J. Wang, Y. P. Zheng, and H. C. Liu, "Thermal quenching of luminescence from buried and surface InGaAs self-assembled quantum dots with high sheet density," J. Appl. Phys. **98**, 084305 (2005).
11. Z. L. Miao, Y. W. Zhang, S. J. Chua, Y. H. Chy, P. Chen, and S. Tripathy, "Optical properties of InAs/GaAs surface quantum dots," Appl. Phys. Lett. **86**, 031914 (2005).
12. C. Y. Zhang, H. C. Yeh, M. T. Kuroki, and T. H. Wang, "Single-quantum-dot-based DNA nanosensor," Nat. Mater. **4**, 826 (2005).
13. W. Cai, D. W. Shin, K. Chen, O. CHeysens, Q. Z. Cao, S. X. Wang, S. S. Gambhir, and X. Y. Chen, "Peptide-labeled near-infrared quantum dots for Imaging tumor vasculature in living subjects," Nano Lett. **6**, 669 (2006).
14. K. Adlkofer, E. F. Duijs, F. Findeis, M. Bichler, A. Zrenner, E. Sackmann, G. Abstreiter, and M. Tanaka, "Enhancement of photoluminescence from near-surface quantum dots by suppression of surface state density," Phys. Chem. Chem. Phys. **4**, 785 (2002).
15. E. F. Duijs, F. Findeis, R. A. Deutschmann, M. Bichler, A. Zrenner, G. Abstreiter, K. Adlkofer, M. Tanaka, and E. Sackmann, "Influence of Thiol coupling on photoluminescence of near surface InAs quantum dots," Phys. Stat. Sol. (b) **224**, 871 (2001).
16. B. L. Liang, Zh. M. Wang, Yu. I. Mazur, G. J. Salamo, E. A. Decuir Jr., and M. O. Manasreh, "Correlation between surface and buried InAs quantum dots," Appl. Phys. Lett. **89**, 043125 (2006).

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17. B. L. Liang, Zh. M. Wang, Yu. I. Mazur, G. J. Salamo, "Photoluminescence of surface InAs quantum dots stacking on multilayer buried quantum dots," Appl. Phys. Lett. **89**, 243124 (2006).
18. Zh. M. Wang, Yu. I. Mazur, Sh. Seydmohamadi, G. J. Salamo, and H. Kissel, "Photoluminescence linewidths from multiple layers of laterally self-ordered InGaAs quantum dots," Appl. Phys. Lett. **87**, 213105 (2005).
19. P. Hove, B. Abbey, E. C. Le Ru, R. Murray, and T. S. Jones, "Strain-interactions between InAs/GaAs quantum dots layers," Thin solid Films. **464-465**, 225 (2004).
20. V. Talalaev, J. Tomm, N. Zakharov, P. Werner, B. Novikov, and A. Tonkikh, "Transient spectroscopy of InAs quantum dot molecules," Appl. Phys. Lett. **85**, 284 (2004).
21. Yu. I. Mazur, Z. M. Wang, G. G. Tarasov, M. Xiao, G. J. Salamo, J. W. Tomm, V. Talalaev, and H. Kissel, "Interdot carrier transfer in asymmetric bilayer InAs/GaAs quantum dot structures," Appl. Phys. Lett. **86**, 063102 (2005).
22. R. Heitz, I. Mukhametzhanov, P. Chen, and A. Madhukar, "Excitation transfer in self-organized asymmetric quantum dot pair," Phys. Rev. B. **58**, R10151 (1998).

1. Introduction

During the past decade self-assembled semiconductor quantum dots (QDs) have attracted extensive research interest [1-3]. They have been successfully used in lasers, detectors and many other device structures [4-7], holding the promise for the next generation of optoelectronic devices due to their inherent quantum confinement. Generally, semiconductor QDs are buried (buried quantum dots or BQDs) in a material with a larger bandgap, confining the wave function in all dimensions. When the QDs are directly exposed to air (surface quantum dots or SQDs) [8-11], their optical and electronic behavior becomes very sensitive to fluctuations in the surface potential. Such surface-sensitive QD structures are expected to play an important role in future sensor applications [12]. For example, water-soluble colloidal QDs have received much attention as biological markers and sensors [13]. In(Ga)As SQDs could also be prospective candidates for sensing biological information in solution instead of surface bonded colloidal QDs [14-16]. However, reports indicate that In(Ga)As SQDs exhibit much weaker and broader emission at significantly lower energy than BQDs [10-11]. It is therefore necessary to improve the optical performance of SQDs for use in such devices. Recently, we studied the optical properties of SQDs through a series of samples with uncapped InAs QDs stacked on a variable number of BQD layers. We observed that the optical performance of InAs SQDs was improved as the number of stacked BQD layers increased due to the strong carrier transfer from BQDs to SQDs [16-17]. It is well known that carrier transfer between layers of stacking QDs depends on the spacer thickness. However, a picture of the carrier transfer between the SQDs and BQDs is still lacking. In this work we report on a hybrid QD structure composed of $In_{0.4}Ga_{0.6}As$ SQDs and multilayers of $In_{0.4}Ga_{0.6}As$ BQDs with spacer thicknesses between 30ML to 120ML. Temperature and excitation intensity dependence of the PL together with the PL temporal decay reveal that different GaAs spacer thicknesses will change the transfer rate of carriers between the layers of QDs and consequently the optical performance of the SQDs.

2. Experiments

The samples were grown on GaAs (311)B substrates, for enhanced QD uniformity, by solid-source molecular beam epitaxy (MBE) [18]. After oxide desorption and growth of a 0.5 μm GaAs buffer, 12 monolayers (MLs) of $In_{0.4}Ga_{0.6}As$ were deposited at a substrate temperature of 530 °C and then capped by a GaAs spacer. This growth was repeated to form the 14-layers of BQDs. A fifteenth layer of 12 ML of $In_{0.4}Ga_{0.6}As$ formed the SQDs with no capping. The growth rates of GaAs and InAs are 1 ML/s and 0.35 ML/s, respectively. Three samples with spacer thicknesses of 30 ML, 60 ML, and 120 ML were grown and compared for this work. For all of these samples the dots feel the strain transferred from the buried layers of QDs, which leads to vertical alignment [18-19]. However, the QDs do undergo a strong variation of quantum coupling between them as the GaAs spacer increases from 30 ML to 120 ML. This

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permits study of the influence of GaAs spacer thickness on the tunneling of electrons and consequently the optical performance of the stacked heterostructure.

For continuous-wave PL measurements, samples were mounted on the cold finger of a close-cycled cryostat. A frequency doubled YAG laser operated at 532 nm was employed as the excitation source. The PL signal was dispersed by a spectrometer and then detected by a CCD detector. For time-resolved PL measurements, the excitation source was switched to a mode-locked Ti:sapphire laser operated at 750 nm and the PL was detected by a Hamamatsu C5680 Streak Camera.

3. Results and discussion

Figure 1 shows the AFM images of the SQDs. For the 30 ML spacer sample, the number density of the SQDs is estimated to be ~ 7.9×10^{9} cm^{-2}. The SQDs are elongate with a length of (116.5±10.0) nm, a width of (81.8±8.6) nm and a height of (17.9±2.9) nm. For the 60 ML spacer sample, the number density of the SQDs is estimated to be ~7.6×10^{9} cm^{-2}, which is almost identical to the 30 ML spacer sample. The SQDs are elongated with a length of (113.5±8.3) nm, a width of (81.3±8.6) nm and a height of (19.5±2.3) nm. For the 120 ML spacer sample, again the SQDs have a similar number density at ~ 7.7×10^{9} cm^{-2}. But, the SQDs are more round with a diameter of (80.6±8.1) nm and a height of (17.0±1.5) nm. So the shape and size of SQD depend on the spacer thickness, which is related to the anisotropic adatom migration and relieved strain-field on the sample surface [19].

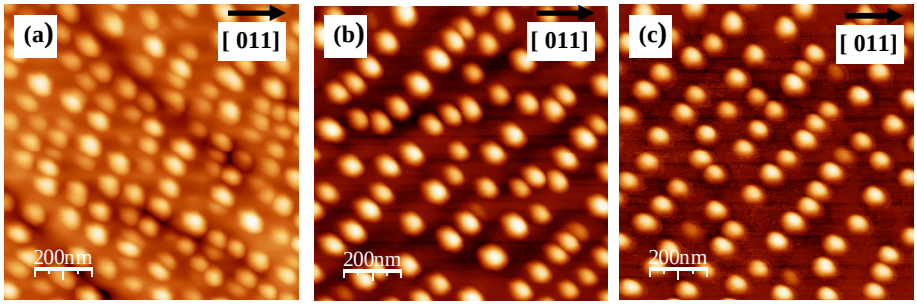

Fig. 1. 1μm×1μm AFM image of the SQDs for the sample with (a) 30 ML spacer, (b) 60 ML spacer, and (c) 120 ML spacer.

Low temperature (10K) PL spectra at a laser excitation intensity of 3 W/cm^2 are plotted in Fig. 2. We see distinct peaks which can be identified as being from the SQDs and the BQDs in each of the three samples [16]. The high energy peak in each case is expected to be from the BQDs due to added compressive strain and narrowing of the confinement potential during the growth of the capping layers [8]. With the exception of a drop in absolute intensity for the thinnest spacer, the BQD peak is very similar in each of the samples. The SQD peak, however, exhibits a dramatic change in both intensity and position by varying the thickness of the spacer layer. For example, a significant shift, ~169 meV, towards lower energy is observed for the SQD PL with larger spacer thicknesses. We note here that the intensity, relative to the BQDs, being equal, and the linewidth of the SQDs, ~100 meV, for the 30 ML sample is among the best reported in the literature [8-11]. The narrow PL linewidth of the SQDs is likely due to the smooth sample surface and the homogenous QDs grown on GaAs (311)B [18]. Also, the SQD peak has a 250~450 meV red-shift with respect to that of the BQDs, which is expected due to the strain change before and after the growth of the GaAs capping layer [8].

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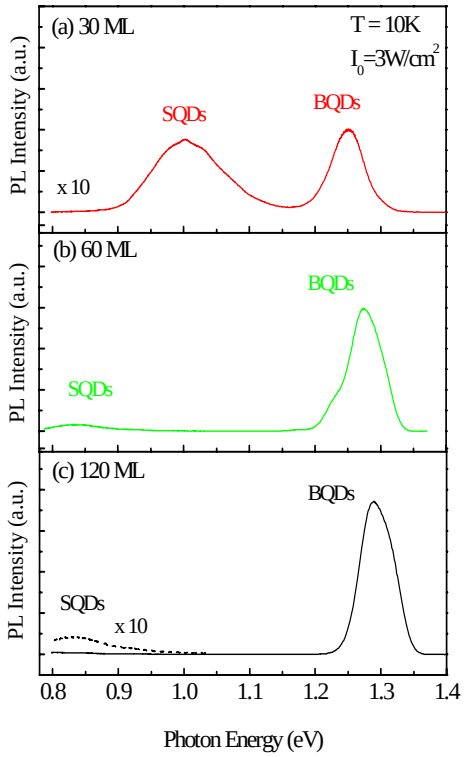

Fig. 2. Low temperature (10K) PL spectrum of the sample with (a) 30 ML spacer, (b) 60 ML spacer, and (c) 120 ML spacer.

The observed variation of the SQD PL emission in Fig. 2 is understood due to the difference in coupling between layers of QDs for the different samples. It is well documented that for these types of samples the QDs will vertically self-align due to the strain field transmitted through the GaAs spacer layers. For the sample with 30 ML GaAs spacers, there should be efficient carrier transfer between dots in different layers [20]. The multiple layers of BQDs can act as a carrier reservoir for the SQDs. This would enhance the PL intensity from SQDs at the expense of the PL intensity of BQDs, as we see in Fig. 2. However, the increased amplitude of the PL intensity from the SQDs is much less than the PL intensity loss from the BQDs. We attribute this to the effect of surface states [17]. The quantum coupling between layers of QDs decreases exponentially with increasing GaAs spacer thickness [21-22]. This is evident in comparing the PL from the 30ML spacer sample with that from the 60 ML spacer sample where there is nearly an order of magnitude reduction in intensity. The quantum coupling is further reduced between layers of QDs for the 120 ML spacer sample such that the carrier transfer to the SQDs vanishes. In this case, the BQDs and the SQDs each have an optical response similar to what would be observed if they were independent, as in previous reports [8-11].

We also investigated the PL as a function of laser excitation intensity to examine the change in coupling between the SQDs and BQDs as the spacer thickness was varied. Fig. 3 shows the evolution of the PL integrated intensity as the excitation power is increased. All samples exhibit a nearly linear behavior at low to moderate powers indicating mostly single exciton recombination. For the 120 ML spacer sample, we assert that there is no coupling between layers of QDs, because the PL of the BQDs responds linearly to excitation power over large ranges of intensities like a typical multilayer QD sample. This is due to the increase

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in the number of dots in multilayers over a single layer increasing dramatically the power required to reach the biexciton regime. The 60 ML spacer sample, however, appears to exhibit weak coupling between the layers of QDs. The variation of the BQDs PL integrated intensity is only slightly different from that of BQDs with 120 ML spacers. However, for this sample, we see that the emission from the SQDs and the BQDs have a similar, nearly linear, dependence on the excitation intensity. This similarity between the SQD and BQD power dependence is more pronounced in the 30 ML spacer sample where there is strong coupling between layers of QDs, so the photo-excited carriers can easily transfer from the BQDs to the SQDs.

It is evident, here, that the BQDs are acting as a reservoir of carriers for the SQDs. Modeling this as a three level system, i.e., BQDs, SQDs, and vacuum, and applying simple rate equations reproduces the observed co-linear behavior of the power dependence if we assume that the BQDs capture the photo-excited carriers and the SQDs receive carriers through tunneling only.

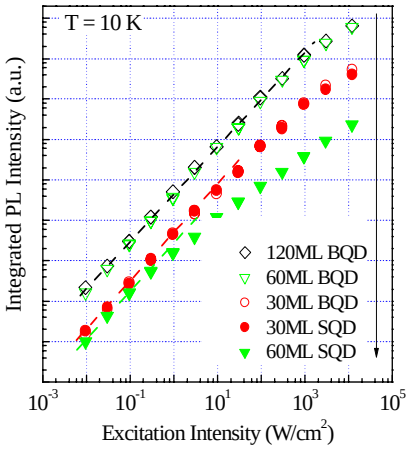

Fig. 3. Integrated PL intensities as a function of the excitation intensity

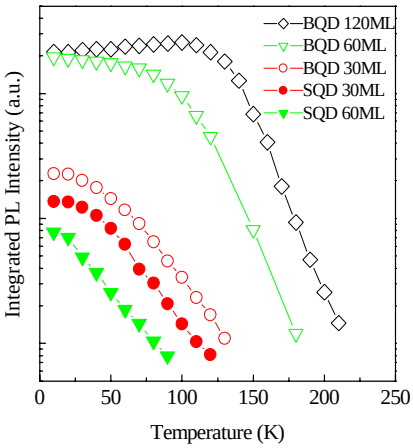

Fig. 4. Integrated PL intensities as a function of the temperature

Figure 4 adds to the physical picture with the integrated PL intensity as a function of temperature. For the 120 ML spacer sample, the integrated BQD PL intensity remains nearly constant up to 140K. This is a typical feature for multi-layers of BQDs and demonstrates the ~12 meV exciton binding energy in these dots. For the 60 ML spacer sample, due to carrier transfer from the BQDs, the BQD PL emission begins to quench as early as ~ 90 K. The SQD PL emission rapidly decreases as the temperature increases and vanishes at ~ 90 K. For the 30 ML spacer sample, due to the strong carrier transfer, it is expected that the SQD PL peak and BQD PL peak would have totally different temperature dependences than in the other samples. As shown by Fig. 4, the integrated PL intensity of SQDs begins to rapidly decrease at a lower temperature ~ 30 K. This is due to a faster thermal quenching of the luminescence from the SQDs [10]. Remarkably, the integrated PL intensity of BQDs has a temperature dependence similar with and correlates to that of the SQDs. This is another indication that carrier transfer from the BQDs strongly influences the PL from SQDs. It is also worthy to note that as shown in Fig. 4, due to the stronger carrier transfer, the quenching of the SQD PL of 30 ML sample is extended to a higher temperature as compared with that of the 60ML sample.

The effect of spacer thicknesses is further demonstrated by time-resolved PL measurements. Since the PL band of the SQDs is beyond the response region of our streak camera, the BQD PL signals are measured at the ground state transition for all samples to

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provide evidence of carrier transfer. As shown in Fig. 5, the BQD PL decay time (450 ps) from 30 ML spacer sample is very fast. In comparison with that, the BQD PL decay time from the 60 ML (1100 ps) and 120ML (1150 ps) spacer samples are very long and similar to the radiative lifetime of isolated layers of QDs. Therefore, in the 30 ML sample, carrier transfer from the BQDs is in strong competition with their radiative recombination. As was found in Fig. 2(a), this transfer of carriers out of the BQDs corresponds to an increase in PL intensity of the SQDs. This reinforces the claim that the BQDs are acting as a source of carriers for recombination in the SQDs.

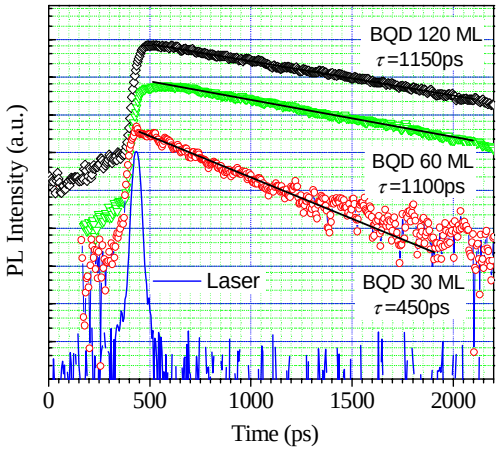

Fig. 5. Time-resolved PL of BQDs measured at ground state transition

4. Conclusion

In conclusion, we varied GaAs spacer thicknesses to tune the coupling between $In_{0.4}Ga_{0.6}As$ SQDs and a multilayer stack of BQDs. For the sample with a 30 ML GaAs spacer between layers of QDs, there is found to be efficient carrier transfer towards the SQDs due to the strong quantum coupling between layers. As a result of this carrier transfer the BQDs and the SQDs are found to have correlated optical behavior. When the spacer is increased to 60 ML, the coupling between the SQDs and the multi-layers of BQDs becomes very weak and appears to be completely turned off for the 120 ML thick spacers. In this case, the BQDs and the SQDs each have an optical behavior as would be observed if they were independent. So the carrier transfer efficiency and consequently the optical behavior of the SQDs as well as the BQDs is strongly dependent on the GaAs spacer thickness, which provides an experimental method to tune and control the performance of the surface QDs.

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