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Hidden resonant excitation of photoluminescence in bilayer arrays of InAs/GaAs quantum dots

Yu. I. Mazur, Z. M. Wang, G. J. Salamo, and Min Xiao[a)]
Department of Physics, University of Arkansas, Fayetteville, Arkansas 72701

G. G. Tarasov, Z. Ya. Zhuchenko, and W. T. Masselink
Institut für Physik, Humboldt-Universität zu Berlin, Invalidenstrasse 110, 10115 Berlin, Germany

H. Kissel
Ferdinand-Braun-Institut für Höchstfrequenztechnik, Albert-Einstein-Strasse 11, D-12489 Berlin, Germany

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Photoluminescence (PL) of self-organized quantum dots (QDs) in bilayer InAs/GaAs structures is studied with a fixed seed layer and spacer, but variable second-layer coverage. Careful line shape analysis reveals modulation in the high-energy tail of the seed-layer PL spectrum. The oscillation-like behavior is reproducible with variations in both the temperature and optical excitation energy. These oscillations are attributed to carrier relaxation through inelastic phonon scattering from the wetting layer to the QD excited states. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1606109]

Carrier relaxation, including tunneling through potential barriers, represents one of the most fundamental processes described by quantum kinetics. Recently, tunneling between layers of semiconductor quantum dots (QDs) has attracted attention in view of the development of the growth capability to realize vertically aligned or stacked QD geometries.[1–4] It is also of interest because a good understanding of the different excitation and decay channels of excited states in multilayer QD structures will have important consequences for their potential application as emitter or detector arrays.

For stacked QD structures, the coupling strength between QD layers is controlled by systematic variation of the barrier thickness between layers and/or control over the dot size or composition from one layer to the next. For example, if the barrier is thick enough, the electronic levels in the layers of dots will have carrier recombination in each layer separately. However, if the barrier is reduced the carrier wave function in each dot in one layer can spread beyond the barrier into an adjacent dot in another layer, and as such coupling is established between the levels of QDs in bilayer structures. In this case carriers can be photogenerated in a given well and tunneling to an adjacent vertically aligned dot can occur if the tunneling time is less than the recombination time. In similar manner, by varying the QD size or composition, the energy separation between the two tunneling QD layers can be used to control the tunneling probability.

While due attention has been directed toward the coupling between closely stacked QD layers,[5–9] the role of the corresponding wetting layer (WL) in such vertically aligned structures has been relatively less explored. In order to distinguish different contributions to the relaxation of photoexcited carriers we consider a structure composed of two layers of weakly coupled InAs/GaAs QDs. This geometry allows us to discriminate between the coupling QDs in adjacent layers and the coupling between QDs in the seed layer and the WL associated with QDs of the adjacent or second layer.

The samples were grown using a solid-source molecular beam epitaxy chamber coupled to an ultrahigh vacuum scanning tunneling microscope (STM). The structures consist of two InAs layers. Each sample was grown on a GaAs (001) substrate, 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 seed QD layer is then grown by depositing 1.8 monolayer (ML) of InAs at a growth rate of 0.1 ML/s, As_4 partial pressure of 8×10^{-6} Torr, and substrate temperature of 500 °C. This is followed by 50 ML GaAs deposited on top of the seed QD layer. The second QD layer was then added. The InAs deposition coverage in the second layer was varied from 1.8 to 2.7 ML for different samples used in our measurements. Each sample for optical study was finally capped with a 150 ML GaAs layer. The samples were structurally characterized by plane-view STM and cross-sectional transmission electron microscopy (XTEM). The photoluminescence (PL) was studied in temperature range of 10–300 K using the 514.5 nm line of an Ar^+ laser for GaAs excitation, a Ti–sapphire laser for WL excitation, as well as a HeNe laser for intermediate energy excitation range, thus spanning excitation densities from 0.1 to 20 W/cm^2.

Figure 1(a) shows a typical XTEM image of the sample with 1.8 and 2.4 ML depositions for the seed layer and the second layer, respectively. Statistical analyses of XTEM images show the resulting sample to be a weakly vertically correlated (~50% for 50 ML GaAs spacer thickness) double-layer QD structure, designed with a significantly different average QD size in the seed layer compared to that in the second layer. The QDs in the second layer are nearly twice the size of those in the seed layer due to additional deposition, as well as to the influence of the strain field from the seed layer.[3,10] The STM statistical analysis [Fig. 1(b)] indicates a QD size distribution of 4±1.5 nm for the height, 20±3 nm for the width, and a dot density of about 4.5×10^{10} cm^{-2} in the seed layer. The dot density in the second layer is variable over the range of $2.5–4\times10^{10}$ cm^{-2},[10] depending on the InAs coverage.

[a)] Electronic mail: mxiao@mail.uark.edu

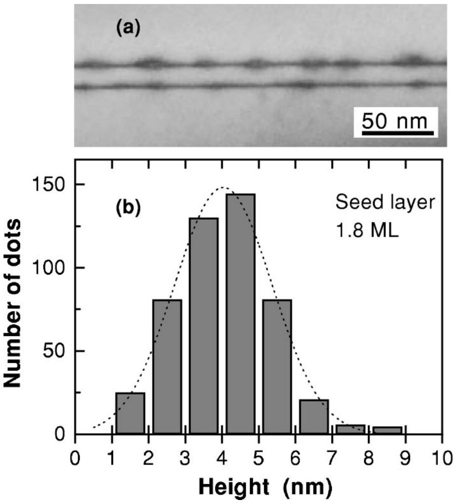

FIG. 1. (a) XTEM image of the bilayer sample with 1.8/2.4 ML InAs deposition on the seed and second layers, respectively. (b) The height distribution of InAs islands in the single-layer sample with a 1.8 ML InAs deposition.

Figure 2 depicts PL spectra of the sample with a second-layer coverage (D_2) of 2.7 ML measured at different temperatures from 10 up to 160 K. Two characteristic peaks centered at 1.07 and at 1.21 eV give evidence of two different size QD arrays, i.e., the larger base QDs in the second layer and the smaller base QDs in the seed layer. The temperature variation of the peak magnitudes is consistent with a model for QDs that are vertically aligned between the two layers that allows only a small amount of carrier transfer from the QDs in the seed layer to the QD ground states in the second layer.[10] Close examination of the data reveals slight modulation of the high-energy tail in the PL spectrum. The shape of this tail cannot be fitted to a Gaussian profile. In order to show the deviations from the Gaussian distribution more clearly, the difference spectra are also plotted in Fig. 2. The difference spectra indicate a strong correlation with the spectra of the second derivative for the measured PL. The energy separations between the adjacent peaks in the difference spectrum are notably periodic and coincide with the energies of longitudinal optical (LO) phonons in a strained InAs layer having $h\omega_{\mathrm{LO}} \approx 30$ meV. Such a characteristic difference spectrum was consistently observed for all samples used in this study.

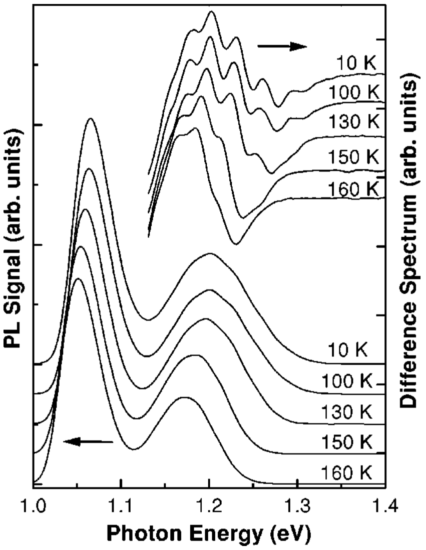

FIG. 2. PL and PL difference spectra measured at different temperatures of the sample with 1.8/2.7 ML InAs deposition on the seed and second layers, respectively.

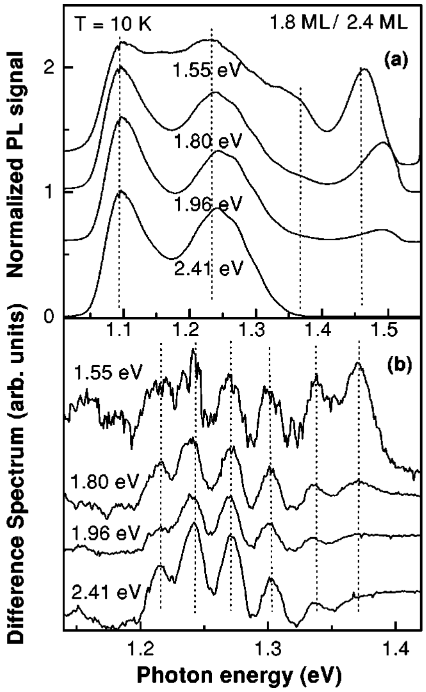

FIG. 3. (a) PL and (b) PL difference spectra of the sample with 1.8/2.4 ML InAs deposition on the seed and second layers, respectively, measured at different excitation wavelengths.

Figure 3(a) plots PL spectra of a sample with $D_2 = 2.4$ ML measured at different excitation wavelengths and at low temperature of $T = 10$ K. The substantial dependence of the PL spectral shape on the excitation energy is clearly seen. The spectrum, a result of deep excitation in the GaAs substrate, resembles the spectrum in Fig. 2. When the excitation energy approaches the band edge of GaAs, additional peaks arise in the region expected from absorption due to the WL. The broad PL feature that appears within 1.42–1.48 eV is in the typical WL spectral range for single layer samples,[10] and is due to the unresolved emission from both the seed and the second InAs WLs.[3] Interestingly, the PL feature at 1.34–1.39 eV also appears in the spectrum and can be assigned to the recombination of nonequilibrium carriers localized in two-dimensional (In,Ga)As islands that act together as the second WL and are only several ML thick.[11] These islands originate from well width fluctuations in the thin WL. Such two-dimensional islands predominantly appear in the second layer because of the rougher GaAs surface (grown at 500 °C) on which the second InAs layer is grown. This is in contrast to the flat GaAs surface (grown at 580 °C) used for InAs seed layer growth. This explanation is consistent with the absence of a peak at 1.34–1.39 eV as well as the observation of a sharp peak, rather than the broad peak, in the 1.42–1.48 eV range, in the PL spectrum of the 1.8 ML single layer reference sample.[10] Even with this broad range of tuning of the excitation energy, the difference spectra [in Fig. 3(b)] retain their shapes. The oscillation period again corresponds to the known LO phonon energy in InAs. Finally, Fig. 4 shows the PL spectrum and the difference spectrum of the sample with $D_2 = 1.8$ ML, i.e., the InAs coverage for both layers in this bilayer structure is basically the same. While the absence of

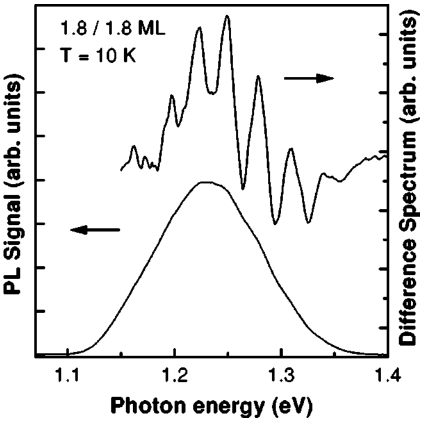

FIG. 4. PL and PL difference spectra of the sample with equal InAs deposition (1.8 ML) on seed and second layers.

a noticeable redshift of the PL spectrum does give evidence of weak vertical coupling between QDs, the difference spectrum, in each case, reproduces the same oscillating character.

There are several possible reasons for the observed oscillating behavior in the difference spectra of these samples that should be explored. Following Fafard *et al.*[2] such behavior could be due to the symmetric and antisymmetric states arising from wave function coupling between vertically coupled QD pairs. However, this mechanism has to be ruled out, because the predicted level splitting for the structure with such a thick spacer layer (50 ML) is much smaller than the energy spacing measured experimentally. Additional possibilities include that, the observed oscillations might be related to the excited states of QDs[6] in the seed layer or to the energy level clusters[7] arising from the removal of the orbital degeneracy of the bound states in the QDs. However, since these energy level clusters are not evenly spaced and the excitation intensity used in our PL experiments was not enough to substantially populate excited states, we conclude that neither the complicated structures of the QD's excited states nor quantum-size effects can give a regular oscillatory behavior in the PL difference spectrum as observed in our samples described here.

On the other hand, our data give strong evidence of InAs LO-phonon participation in the relaxation of excited carriers in the QD seed layer.[12,13] We associate the observed oscillations in the difference spectra with resonant decay of an excited level in the WL of the second layer with simultaneous excitation of a QD in the seed layer and generation of several LO phonons. The energy position of nth peak maximum $h\nu_n^{\max}$ in the difference spectrum would then be determined to be $h\nu_n^{\max}=h\nu_{\mathrm{WL}}-nh\nu_{\text{LO-phonon}}$, where n is an integer. Thus, basically one could expect the manifestation of peaks, separated by the InAs LO-phonon energy, in the difference spectrum all across the QD PL broadened peak of the seed layer. Our assumption that the WL responsible for resonant excitation of QDs in the seed layer is related to the second QD layer is based on the following arguments: First, modulation of the high-energy PL tail was not observed in the single layer structures. Second, the QDs in the seed layer definitely couple the states in the second layer in spite of the weakness of this coupling. The temperature behavior of the PL spectra (Fig. 2) proves such coupling.[10] Third, the energy differences between the WL states of the second QD layer and the QD states of the seed layer allow multiphonon relaxation with a minimal number of actual phonons, i.e., in the lowest order of perturbation theory.

A recent study of the magneto-optical properties of stacked self-assembled InAs QDs[14] supports the feasibility of this mechanism. In that study, it was shown that the increase in integrated intensity of the QD PL contribution in a magnetic field is completely compensated for by the decrease in integrated intensity of the GaAs and WL PL bands. In addition, systematic temperature-dependent measurements of PLE spectra from self-assembled InAs/GaAs QDs[15] also support our explanation by presenting evidence for the interaction of discrete QD excited states with a quasicontinuum of states that spread just below the WL absorption edge. That is, similar resonant coupling between discrete states of QDs and the quasicontinuum states of WL, namely, Fano resonance, supports the mechanism that we used to explain the oscillations observed in the bilayer QD PL spectra. Hence, the quasicontinuum provides an effective channel for carrier transfer from the WL states into QD excited states with the emission of multiple InAs LO phonons.

In conclusion we have detected an oscillating contribution in the high-energy tail of the QD PL spectrum from the seed layer in a bilayer array of InAs/GaAs QDs. The oscillation contribution is explained in terms of coupling between QDs in the seed layer and the WL associated with the second QD layer. This coupling leads to transfer of excitation from the WL to excitation of the seed QDs and excitation of InAs LO phonons. This resonant effect provides hidden resonant excitation of PL in the bilayer InGa/GaAs QD system.

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[1] G. S. Solomon, J. A. Trezza, A. F. Marshall, and J. S. Harris, Jr., Phys. Rev. Lett. **76**, 952 (1996).

[2] S. Fafard, M. Spanner, J. P. McCaffrey, and Z. R. Wasilewski, Appl. Phys. Lett. **76**, 2268 (2000).

[3] R. Heitz, I. Mukhametzhanov, P. Chen, and A. Madhukar, Phys. Rev. B **58**, R10151 (1998).

[4] M. A. Migliorato, L. R. Wilson, D. J. Mowbray, M. S. Skolnick, M. Al-Khafaji, A. G. Cullis, and M. Hopkinson, J. Appl. Phys. **90**, 6374 (2001).

[5] N. Perret, D. Morris, L. Franchomme-Fosse, R. Cote, S. Fafard, V. Aimez, and J. Beauvais, Phys. Rev. B **62**, 5092 (2000).

[6] R. Heitz, O. Stier, I. Mukhametzhanov, A. Madhukar, and D. Bimberg, Phys. Rev. B **62**, 11017 (2000).

[7] M. Bissiri, G. Baldassarri, H. Högerstral, M. Capizzi, P. Frigeri, and S. Franchi, Phys. Rev. B **64**, 245337 (2001).

[8] H. Jiang and J. Singh, IEEE J. Quantum Electron. **34**, 1188 (1998).

[9] L.-W. Wang, J. Kim, and A. Zunger, Phys. Rev. B **59**, 5678 (1999).

[10] Yu. I. Mazur, X. Wang, Z. M. Wang, G. J. Salamo, M. Xiao, and H. Kissel, Appl. Phys. Lett. **81**, 2469 (2002).

[11] A. F. Tsatsul'nikov, N. N. Ledentsov, M. V. Maksimov, A. Yu. Egorov, A. E. Zhukov, S. S. Ruvimov, V. M. Ustinov, V. V. Komin, I. V. Kochnev, P. S. Kop'ev, and Zh. I. Alferov, Semiconductors **30**, 938 (1996).

[12] C. Guasch, C. M. Sotomayor Torres, N. N. Ledentsov, D. Bimberg, V. M. Ustinov, and P. S. Kop'ev, Superlattices Microstruct. **12**, 509 (1997).

[13] M. J. Steer, D. J. Mowbray, W. R. Tribe, M. S. Skolnick, D. M. D. Sturge, M. Hopkinson, A. G. Cullis, C. R. Whitehouse, and R. Murray, Phys. Rev. B **54**, 17738 (1996).

[14] J. C. Gonzalez, F. Plentz, N. Köche, W. N. Rodrigues, M. V. B. Moreira, A. G. de Oliveira, M. I. N. da Silva, and M. S. Andrade, J. Appl. Phys. **89**, 3760 (2001).

[15] C. Kammerer, G. Cassabois, C. Voisin, C. Delalande, Ph. Roussignol, A. Lemaître, and J. M. Gérard, Phys. Rev. B **65**, 033313 (2002).