Tuning of electronic coupling between self-assembled quantum dots

S. I. Rybchenko and I. E. Itskevich[a)]
Department of Engineering, University of Hull, Kingston-upon-Hull, HU6 7RX, United Kingdom

M. S. Skolnick, J. Cahill, and A. I. Tartakovskii
Department of Physics and Astronomy, University of Sheffield, Sheffield, S3 7RH, United Kingdom

G. Hill and M. Hopkinson
Department of Electronic and Electrical Engineering, University of Sheffield, Sheffield, S1 3JD, United Kingdom

(Received 13 December 2004; accepted 2 June 2005; published online 11 July 2005)

Semiconductor self-assembled quantum dots (SAQDs) normally have zero-dimensional properties, but become coupled and acquire higher-dimensional character if the distance between the dots is small. Using photoluminescence spectroscopy under high hydrostatic pressure, we have obtained clear evidence for electronic coupling due to quantum-mechanical tunneling in stacks of InGaAs/GaAs SAQDs. We demonstrate that application of pressure allows controllable tuning and suppression of the electronic coupling. The effect originates from a pressure-induced increase in the effective mass of Γ-electrons and a related increase in the interdot-barrier height. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1995953]

Self-assembled quantum dots (SAQDs) are formed spontaneously when a layer of a semiconductor material such as In(Ga)As is deposited on a material with a differing lattice constant such as (Al)GaAs. Their small size (10–30 nm) results in fully quantized discrete spectra of strongly localized electrons and holes with large interlevel spacing. As a result, SAQDs provide a nearly ideal example of a zero-dimensional semiconductor system. In addition, their high optical efficiency due to the absence of internal defects provides enormous potential for device applications.[1,2]

If several layers of SAQDs are grown with spacer layer thicknesses less than ≈20 nm, the dots in successive layers are formed on top of the dots in the previous layer, forming vertical stacks in the growth direction.[3] Stacks of SAQDs are widely used, for example, as the active region of quantum-dot lasers.[4] If the interlayer distance is large, dots in a stack are isolated and zero-dimensional. However, if the distance is sufficiently small, the dots become coupled. This may happen because the wave functions of individual dots in a stack overlap due to quantum-mechanical tunneling, so that the dots are coupled electronically.[5] Another possible mechanism of coupling is by means of strain fields which build up with increasing number of SAQD layers. Coupling due to interdot Coulomb dipole-dipole interaction, followed by a transfer of electronic excitation[6] is also conceivable. Understanding the mechanisms of interdot coupling and their relative contribution is important from the point of view of fundamental properties of stacks of SAQDs, and is crucial when coupling is employed in device design.

In this letter, we report an experimental investigation of interdot coupling in stacks of SAQDs. The aim of the experiments was first, to distinguish between the coupling mechanisms and second, to modify the strength of the coupling in a controllable way. For this purpose, we have employed the technique of high quasihydrostatic pressure. Pressure provides a powerful tool to control the electronic properties of semiconductor systems since it induces a large perturbation of the band structure. At the same time, structural properties such as dot size and shape and the interdot distance are not changed. As a result, coupling due to strain fields or the Coulomb interaction is not expected to be sensitive to pressure.

On the contrary, electronic coupling between the dots is expected to be modified by pressure. There are two parameters that control the coupling of electron states of adjacent dots in a stack; namely, the thickness and the height of the potential barrier between the dots. (The height of the barrier is defined as the energy separation between a size-quantized level in the In(Ga)As dot and the conduction-band edge in the GaAs matrix.) The major effect of pressure on III-V semiconductors is to increase the band gap. For the GaAs direct band gap, the pressure coefficient is 11.6 meV/kbar.[7] We have shown previously that for SAQDs, the coefficient is about 20% smaller.[8,9] Such a difference is likely to originate from the strong nonparabolicity of the conduction band in III-V semiconductors.[10] Due to nonparabolicity, the increase in the band gap is followed by an increase in the effective mass of electrons in the Γ-valley of the Brillouin zone, which decreases the size-quantization energy of electrons in the dots and hence the pressure coefficient. As a result, application of pressure increases the barrier height, so that the barrier becomes less transparent to the electron wave functions. Therefore, high pressure provides an opportunity to tune electronic coupling between quantum dots.

Experimentally, coupling between zero-dimensional quantum dots can be revealed by means of photoluminescence (PL) spectroscopy under varying pumping intensity. Due to unavoidable variations in SAQD size and shape, the PL line from the ground state of the dots is inhomogeneously broadened. If the dots are isolated, all dots may be occupied with photoexcited electrons and holes and contribute to the emission with equal probability. In this case, increasing pumping should result in an increase in PL intensity, but the line shape and position should remain the same (as long as the excited states are not filled). On the contrary, if the dots are coupled, photoexcited electrons and holes relax to lower-

[a)]Electronic mail: i.itskevich@hull.ac.uk

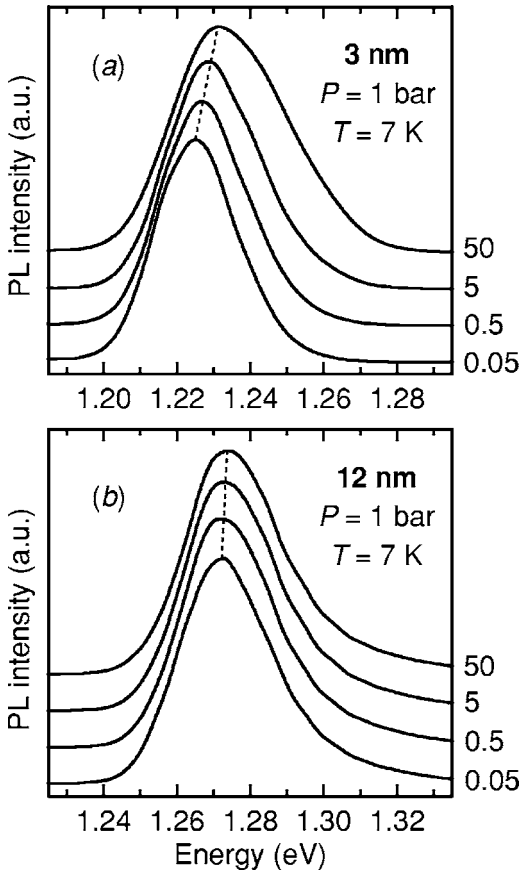

FIG. 1. PL spectra from SAQDs for various excitation power densities (indicated in W/cm^2 near the spectra) at ambient pressure: (a) 3 nm sample, (b) control sample. Spectra are normalized and offset for clarity. The lines which connect the peak maxima are guides to the eye.

energy delocalized states from which recombination occurs. Higher-energy states may be filled and contribute to PL when the pumping is increased. Therefore, in the case of coupled dots, a blueshift of the PL line with increasing pumping is expected.

A series of SAQD samples was grown by molecular-beam epitaxy on GaAs (001) substrates. Each sample incorporated ten sequential layers of $In_{0.5}Ga_{0.5}As$ quantum dots that were separated by GaAs barrier layers and formed vertical stacks. To form the SAQDs, 6-monolayer (ML) InGaAs layers (1.8 nm) were deposited at a growth rate of 0.18 ML/s without growth interruption. The growth temperature was 510 °C throughout. Investigations were performed on a sample with barrier-layer thickness of 3 nm, with a separate sample with barrier-layer thickness of 12 nm being used as a control. High pressure was applied in a diamond-anvil cell with Ar used as a pressure-transmitting medium. The sample in the cell was placed in a He cryostat with optical windows. PL spectra were excited by an Ar^+-ion laser, dispersed by a single-grating spectrometer and recorded by a Si CCD detector. Pressure values P were measured using both fluorescence from a small ruby crystal and emission from the GaAs exciton.

Figure 1 shows PL spectra from both the 3 nm sample and the control sample at atmospheric pressure. The spectra were recorded with varying excitation-power density W in the range below 50 W/cm^2, where emission from the SAQD ground states dominates the spectra. For the 3 nm sample, a significant blueshift of the line is observed with increase in

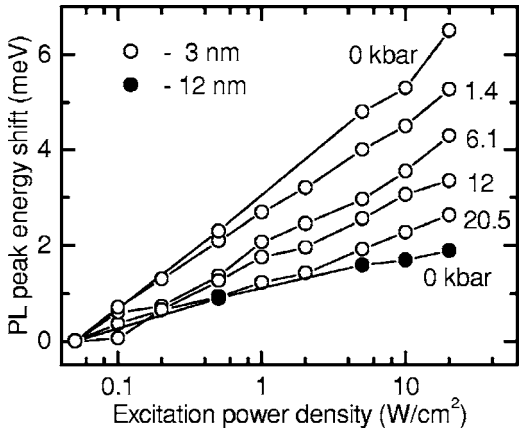

FIG. 2. Blueshift of the PL peak from SAQDs in the 3 nm sample (open circles) as a function of the excitation power density at various pressures, which are indicated in kbar. The lines are guides to the eye. Data for the control sample at the ambient pressure (solid circles) are also shown

W. By contrast, over the same range of W, the shift is negligibly small for the control sample. Note that within the range of W employed, excited states in the dots are not filled and there is only a minor change in the line shape. (Effects of filling of the excited states were observed in the PL spectra from both samples at higher values of W of 100–3000 W/cm^2. In this case, a much stronger blueshift is observed which is followed by a significant change in the line shape.)

Following the discussion above, the observed blueshift is indicative of delocalized states in the 3 nm sample, while in the control sample, the SAQDs are apparently isolated. However, the ambient-pressure data do not provide sufficient evidence to deduce the origin of the delocalization. Evidence for the nature of the delocalization was obtained from PL measurements under high pressure. PL spectra were recorded with varying W in the same range below 50 W/cm^2 at fixed values of P. We found that the blueshift of the PL line from the 3 nm sample is dramatically affected by pressure. Figure 2 shows the relative positions of the PL peak as a function of W for various pressures. With increasing pressure, the blue-

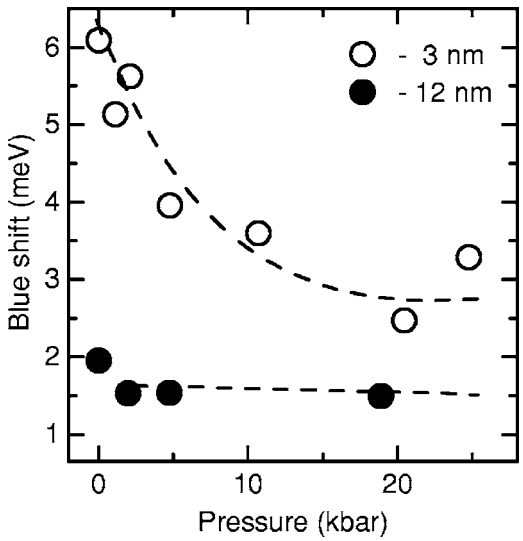

FIG. 3. Blueshift of the PL peak from SAQDs when the pumping density increases from 50 mW/cm^2 to 20 W/cm^2 as a function of pressure. Open and solid circles are for the 3 nm sample and the control sample (12-nm barriers), respectively. Dashed lines are guides to the eye.

shift of the peak gradually becomes weaker. For $P > 20$ kbar, the blueshift for the 3 nm sample becomes similar to that observed for the control sample at ambient pressure, the data for which are shown for comparison.

The evolution of the blueshift with pressure is summarised in Fig. 3. The figure shows the magnitude of the blue shift when W increases from 50 $\mathrm{mW/cm^2}$ to 20 $\mathrm{W/cm^2}$ as a function of pressure. It can be seen that for the 3 nm sample, the magnitude of the blueshift falls dramatically with pressure. This provides a possibility to deduce the mechanism giving rise to the delocalization. Mechanisms such as strain-induced coupling or Coulomb-interaction-induced coupling are not sensitive to hydrostatic pressure. Therefore, our results provide unambiguous evidence for electronic coupling between SAQDs, which occurs by means of tunneling of electron wave functions through the barriers between the dots in a stack.

Note that a small blueshift that is observed for the control sample does not depend on pressure and hence cannot be related to electronic coupling. This observation shows that the electronic coupling is suppressed at atmospheric pressure for SAQD stacks with 12-nm-thick barriers. The observed blueshift may be indicative of a relatively weak contribution from other mechanisms of coupling, which were discussed earlier. The same contribution may account for the nonzero blueshift in the studied sample for pressures $P > 20$ kbar.

Our results demonstrate that application of pressure allows tuning of the coupling strength in a controllable way. As was discussed in the introduction, the electronic coupling is suppressed by pressure because the tunneling barriers become higher and hence less transparent to the electron wavefunctions. A sufficiently high pressure leads to quenching of the coupling.

Our results thus agree qualitatively with our expectations. To test the conclusions more quantitatively, we approximated the stacks with a multiple-quantum-well potential in the growth direction. The energies of coupled ground states of the dots in the stack were obtained by solving the one-dimensional Schroedinger equation numerically. For stacks with parameters similar to the ones for the 3 nm barrier sample, the estimates give significantly (3–5 times) larger values for the coupling energies and hence the blueshift than those observed in experiment. However, it is important to note that in other calculations published so far for similar samples, coupling energies from several tens up to 100 meV have been predicted.[11–14] In addition, reported experimental data provide lower values in the range from several tens of meV[15] down to fractions of an meV.[16] These discrepancies demonstrate that the coupling strength is very sensitive to details of the sample structure. As an example, it is particularly sensitive to the dot height. Note that a coupling energy of less than 1 meV was reported for stacks of 6-nm-high dots (with the barrier height of 360 meV).[16] According to our estimates, variation in the dot height from 2 to 6 nm will result in a 5–6 times decrease in the coupling energies *for the same values of the barrier height and width*. (The effect happens because in taller dots, the electron wave function is more strongly localized.) This example illustrates one of the possible reasons for the relatively modest quantitative agreement between theory and experiment, even though the qualitative behavior is well understood. For a definitive conclusion, detailed theoretical investigations as well as full microscopic information on the structural parameters of the stacks are required.

To conclude, we have obtained experimental evidence for electronic coupling between self-assembled quantum dots in vertical stacks. The magnitude of the coupling can be tuned in a controllable way by applying high hydrostatic pressure. A sufficiently high pressure can quench the coupling.

This work was supported by the EPSRC and the Leverhulme Trust.

[1] D. Bimberg, M. Grundmann, and N. N. Ledentsov, *Quantum Dot Heterostructures* (Wiley, New York, 1999).
[2] *Single Quantum Dots. Fundamentals, Applications and New Concepts*, edited by P. Michler, in Topics in Applied Physics Vol. 90 (Springer, Berlin, 2003).
[3] R. Nötzel, Semicond. Sci. Technol. **11**, 1365 (1996).
[4] D. Bimberg, M. Grundmann, F. Heinrichsdorff, N. N. Ledentsov, V. M. Ustinov, A. E. Zhukov, A. R. Kovsh, M. V. Maximov, Y. M. Shernyakov, B. V. Volovik, A. F. Tsatsul'nikov, P. S. Kop'ev, and Z. I. Alferov, Thin Solid Films **367**, 235 (2000), and references therein.
[5] 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).
[6] C. R. Kagan, C. B. Murray, M. Nirmal, and M. G. Bawendi, Phys. Rev. Lett. **76**, 1517 (1996); S. A. Crooker, J. A. Hollingworth, S. Tretiak, and V. I. Klimov, *ibid.* **89**, 186802 (2002).
[7] P. Perlin, W. Trzeciakowski, E. Litwin-Staszewska, J. Muszalski, and M. Micovic, Semicond. Sci. Technol. **9**, 2239 (1994).
[8] I. E. Itskevich, M. Henini, H. A. Carmona, L. Eaves, P. C. Main, D. K. Maude, and J.-C. Portal, Appl. Phys. Lett. **70**, 505 (1997); I. E. Itskevich, S. G. Lyapin, I. A. Troyan, P. C. Klipstein, L. Eaves, P. C. Main, and M. Henini, Phys. Rev. B **58**, R4250 (1998).
[9] A. J. Williamson and A. Zunger, Phys. Rev. B **58**, 6724 (1998).
[10] G. Bastard, *Wave Mechanics Applied to Semiconductor Heterostructures* (Les Editions de Physique, Paris, 1988).
[11] C. Pryor, Phys. Rev. Lett. **80**, 3579 (1998).
[12] W. Sheng and J.-P. Leburton, Appl. Phys. Lett. **78**, 1258 (2001).
[13] B. Szafran, S. Bednarek, and J. Adamowski, Phys. Rev. B **64**, 125301 (2001).
[14] M. Korkusinski and P. Hawrilak, Phys. Rev. B **63**, 195311 (2001).
[15] M. Bayer, P. Hawrilak, K. Hinzer, S. Fafard, M. Korkusinski, Z. R. Wasilewski, O. Stern, and A. Forchel, Science **291**, 451 (2001).
[16] M. H. Son, J. H. Oh, D. Y. Jeong, D. Ahn, M. S. Sun, S. W. Hwang, J. E. Oh, and L. W. Engel, Appl. Phys. Lett. **82**, 1230 (2003).