RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 58, NUMBER 16 15 OCTOBER 1998-II

Scanning transmission-electron microscopy study of InAs/GaAs quantum dots

P. D. Siverns, S. Malik, G. McPherson, D. Childs, C. Roberts, R. Murray,* and B. A. Joyce
Interdisciplinary Research Centre for Semiconductor Materials, Imperial College, London SW7 2BZ, United Kingdom

H. Davock
Materials Department, University of Liverpool, Liverpool L69 3BX, United Kingdom
(Received 14 May 1998)

We present results obtained from a scanning transmission-electron microscopy study of InAs/GaAs quantum dot (QD) layers. It is shown that the QD's are embedded within an $In_xGa_{1-x}As$ confining layer following overgrowth with GaAs. Using energy dispersive x-ray analysis (EDX) the QD dimensions can be measured with reasonable accuracy and are not affected by strain contrast. In QD bilayers where the dots are uncorrelated along the growth direction, a comparison of the indium EDX signals from the confining layer and a dot allow us to estimate the compositions of these regions as $In_{0.07}Ga_{0.93}As$ and $In_{0.31}Ga_{0.69}As$, respectively.
[S0163-1829(98)50636-4]

Self-organized III-V semiconductor quantum dots (QD's) have been extensively studied over the last few years because they promise improved laser operation,[1] but these improvements have not yet been unequivocally demonstrated.[2–4] One possible reason for this is the size distribution of the three-dimensional (3D) islands (roughly 10%), which may negate the advantages of a true zero-dimensional density of states. The size distribution can be seen directly in scanning tunneling microscopy[5] (STM) and atomic force microscopy[6] (AFM) of uncapped QD's and indirectly in the large inhomogeneous linewidths evident in photoluminescence measurements of capped dots.[7] In principle, cross-section transmission-electron microscopy (TEM) can give structural information on capped QD's, but in practice TEM images are formed by a convolution of strain contrast with atomic number (Z) dependent contrast that leads to overestimates of the dot dimensions and, despite a large body of information on QD's, we are still unable to answer some basic questions concerning their size, shape, and composition. Yet these measurements are crucial if modeling of QD structures is to reach the same level of maturity that exists for quantum well structures. In this paper we present scanning transmission-electron microscopy (STEM) measurements on QD layers that resolve some of these problems. Using the high spatial resolution and compositional sensitivity available with energy dispersive x-ray analysis[8] (EDX) we are able to make detailed estimates of the size and shape of the QD's and avoid the problems associated with strain contrast. In addition these results show that overgrown QD's do not conform to the conventional Stranski-Krastanow picture of a 3D island on top of a 2D wetting layer (WL) but the dots lie within an $In_xGa_{1-x}As$ confining layer (CL) that has the same thickness as the dot height. The EDX measurements also allow us to deduce the compositions of the QD's and the CL.

The samples were grown by solid source molecular beam epitaxy on GaAs semi-insulating (001) substrates using As_4. The V III ratio was 14 and the InAs growth rate was 0.25 μmh^{-1}. The GaAs buffer layer was grown at 580 °C and then the substrate temperature reduced to 520 °C for growth of two monolayers (2 ML) of InAs that form the 3D islands. The dots were then overgrown at a rate of 1 μmh^{-1} with 200 Å of GaAs at 520 °C and finally capped with 300 Å of GaAs grown at 580 °C. Both single and bilayer QD samples were grown to investigate the effects of vertical stacking of the dots. The separation of the bilayers was varied from a nominal 50 to 300 Å.

Figure 1(a) shows a scanning transmission micrograph of a bilayer sample showing two well-defined correlated QD layers with a nominal 150 Å spacing. From the contrast, the islands are estimated to have a lateral dimension of 400 Å, a height of 115 Å, and an average lateral separation of 600 Å. However, such images are subject to both atomic number (Z)-contrast and strain contrast that may exist within the GaAs as well as the 3D islands, leading to overestimates of the dot size. Figure 1(b) shows an EDX In K_α linescan (excited by a 10-Å-wide 100-kV beam with 10-Å steps) along the growth direction through the dots. The height of the peaks indicates that the In content of the dots is the same while the peak separation of 126 Å is in reasonable agreement with the intended layer separation. Taking the full

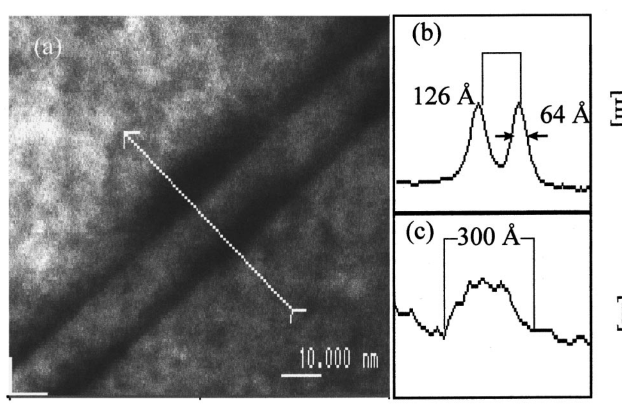

FIG. 1. (a) STEM image of an InAs/GaAs QD bilayer showing a pair of correlated dots separated by 150 Å. Also shown are EDX linescans (of length 640 Å) taken along (b), and perpendicular (c), to the growth direction, which indicate that the dot dimensions are approximately 64 Å high and 300 Å across.

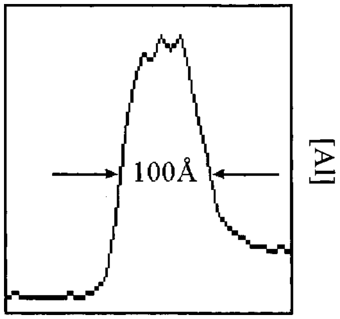

FIG. 2. EDX linescan taken across a GaAs/AlAs/$Al_{0.2}Ga_{0.8}As$ heterojunction. The width of the AlAs layer was intended (and confirmed from the STEM image) to be 100 Å, in good agreement with the FWHM of the EDX peak.

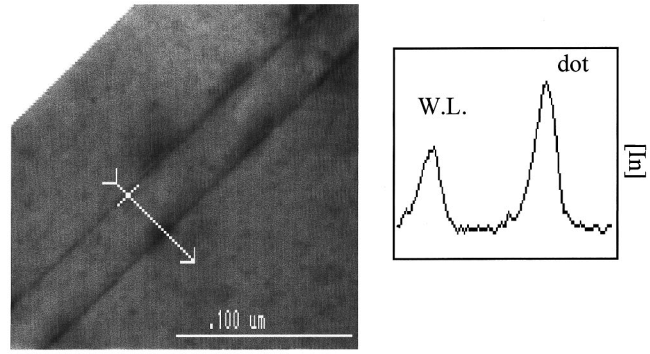

FIG. 3. STEM image of a InAs/GaAs QD bilayer with separation 300 Å. In this case the dots are not vertically correlated. The right-hand part of the figure shows the EDX linescan (length 640 Å) along the growth direction through the CL of the upper dot layer and a dot in the lower layer. The FWHM of the peaks indicate that the thickness of the CL is close to the height of the dot.

width at half maximum (FWHM) of the peaks as an estimate of the dot height we obtain a value of 64 Å. The dot lateral dimension was estimated to be 300 Å by performing a linescan *along* the direction of the WL [Fig. 1(c)]. The EDX measurements are always smaller than those estimated from the TEM contrast but are considered to be more accurate because they are not subject to strain effects. It is also evident from Fig. 1(b) that there is indium present in the region between the QD layers. This has important consequences for electronic coupling between dot layers and will be the subject of another paper. The dimensions obtained for these capped dots are considerably smaller than those deduced from STM measurements that we have made on a series of uncapped samples grown at different substrate temperatures.[5] From these measurements we estimate the dot height to be ≈85 Å and the diameter to be close to 600 Å. This indicates that there may be a significant change in the dot size when overgrown with GaAs. Xie, Chen, and Madhukar[9] have reported transfer of indium into the adjoining barrier regions during deposition of the GaAs cap and Garcia *et al.*[10] have reported reductions in island heights of up to 50% during overgrowth.

The accuracy of the EDX measurements will, of course, depend on the resolution of the technique. To determine this we have performed linescans across a GaAs/AlAs/$Al_{0.2}Ga_{0.7}As$ heterostructure as shown in Fig. 2. The (Al,Ga)As system was chosen because the interfaces are thought to be abrupt (within 2 ML) and the system unstrained. The linescan detecting the Al K_α emission clearly shows the two interfaces and easily distinguishes the different (Al,Ga)As layers from the GaAs. The AlAs thickness of 100 ± 14 Å, deduced from the FWHM of the [Al] signal, is in excellent agreement with the intended thickness and that measured from the contrast (strain is absent in this system). A value for the ultimate resolution of the EDX technique can, in principle, be obtained by performing linescans on increasingly narrow AlAs layers but our preliminary measurements indicate that the resolution is close to 20 Å and significantly smaller than the size of the QD's. In addition, the *L* and *K* lines for In, Ga, and Al are sufficiently close that in these thin samples thickness corrections are unnecessary.

The role of the WL in self-assembled QD's has not been completely elucidated but it is thought that it acts as an efficient source of photogenerated carriers that are then transferred to the dots.[11] However, the conventional view based on the Stranski-Krastanow growth mode is that the WL is a very thin (≈1.7 ML) InAs layer. Figure 3 shows a micrograph and EDX linescan obtained from a bilayer sample where the separation of the dot layers is about 300 Å. In such samples the dots have a roughly 50% probability of vertical stacking[12] and for the case shown here the electron beam traverses through a dot in one layer and the "WL" in the adjacent layer. The EDX trace shows that the peak width (FWHM) in both cases is approximately the same, indicating that the dots lie within a 2D CL that has the same thickness as the dot height. A similar conclusion was reached in recent papers by Woggon *et al.*[13] on the basis of TEM results using a strain/atomic number contrast deconvolution routine and Wu *et al.*[14] using cross-sectional STM. Note also that the indium signal in this sample drops to the background level between the layers so the dots are less likely to be electronically coupled. The EDX measurements also indicate that the indium content of the dot and CL are within a factor of two but this excludes differences in the scattering volume of the dot and CL shown schematically in Fig. 4. Assuming that only a single dot is probed, the indium signals shown in Fig. 3 arise in one case (the left-hand peak) from the CL and in the other from a dot embedded in the CL. Taking the dot diameter to be *D* and the sample thickness to be *L* (see Fig. 4), the indium signal from the CL peak only is

$$[\mathrm{In}]_{\mathrm{CL}} \propto LAy,$$

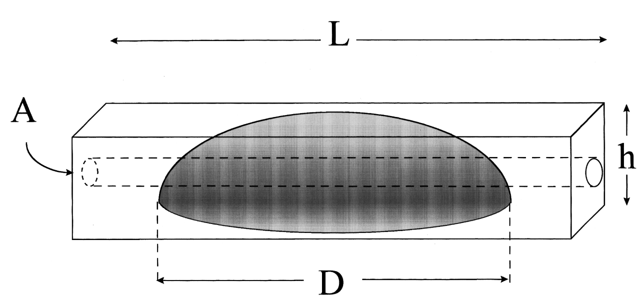

FIG. 4. Schematic of part of the STEM sample (thickness *L*) showing the volume occupied by the lens-shaped dot embedded in a CL of thickness *h*. The volume probed by the EDX beam is indicated by the dotted cylinder of cross section *A*.

where A is the area of the probing electron beam and y is the indium fraction of the CL alloy. For the second (dot plus CL) peak

$$[\mathrm{In}]_{\mathrm{dot+CL}} \propto DAx + (L-D)Ay,$$

where x is the indium fraction of the dot alloy. From Fig. 3 we have

$$[\mathrm{In}]_{\mathrm{dot}} = 2[\mathrm{In}]_{\mathrm{CL}},$$

which leads to

$$x = (1 + L/D)y. \qquad (1)$$

We can obtain another expression involving x and y by equating the total number of In atoms deposited (2 ML) with the number contained in the dots (density$=\rho$ cm^{-2}) and within the CL. The dots are assumed to be lens shaped with a base diameter D and height h and have a volume

$$V = \frac{\pi h}{24}(4h^2 + 3D^2).$$

The total volume occupied by dots (per cm^2) is $V\times\rho$ while for the CL it is $(h - V\times\rho)$. Assuming that the concentration of group III atoms is the same throughout the sample we can write

$$\text{deposited volume InAs (2 ML)} = V\rho x + (h - V\rho)y. \qquad (2)$$

Combining this with Eq. (1) yields

$$\left(\frac{V\rho L}{D} + h\right)y = \text{deposited volume InAs (2 ML)}. \qquad (3)$$

Values for h (64 Å) and D (300 Å) are taken from the EDX measurements. The dot density was determined to be 3×10^{10} cm^{-2} from AFM measurements made on a different sample grown under identical conditions and L is estimated from electron energy loss spectra measurements to be close to 1000 Å, which justifies our earlier assumption that only one dot is probed within the sample thickness. This gives the In fraction of the CL as $y=0.07$ and the dot as $x=0.31$. This contradicts the conventional view of the WL as a 1.7-ML-thick InAs layer and our results indicate that the WL and the QD's alloy with GaAs during the capping process to produce a 2D CL that has a thickness close to 60 Å. Assuming the In to be uniformly distributed within the CL would result in an upper limit for y close to 0.1. The value of x for the dot composition is at first glance surprisingly small. However, using the expression for the low-temperature band gap of strained $In_xGa_{1-x}As$ quantum wells reported by Arent *et al.*[15]

$$E_g = 1.519 - 0.945x + 0.19x^2 + 0.019x^3,$$

with $x=0.31$ gives $E_g = 1.23$ eV. Allowing for confinement effects [~60 meV (Ref. 16)] and assuming an exciton binding energy of 20 meV yields a value of the band gap of 1.27 eV, in good agreement with our photoluminescence value of 1.2 eV.[17] Rosenauer *et al.*[18] have estimated the indium composition of *uncapped* InAs QD's by measuring the local lattice constants using TEM and deduced a value of $x=0.6$, showing that alloying of the dot with the GaAs substrate occurs before capping in agreement with STM results.[5] It is clear from the measurements presented here and from the results obtained by other groups that the interfaces between the dots and the GaAs matrix are not abrupt and the QD's represent a compositional modulation within a 2D $In_xGa_{1-x}As$ CL rather than a spatial modulation normal to a WL as has already been suggested for QD's grown by atomic layer epitaxy.[19] This also means that the confining potential will be slowly varying, making it difficult to model, particularly since photoluminescence excitation measurements fail to provide information on the energies of the excited states of the dots.[17] Although there are potential errors inherent in our EDX measurements, for instance we cannot be certain of the true dot shape (the dots may be more cylindrical), this would involve only minor changes to our estimates of x and y.

Finally, we wish to make some remarks concerning the interaction between the WL and the QD's during capping with GaAs. The QD dimensions, extrapolated from our STM measurements, are estimated to be ≈85 Å high and ≈600 Å in diameter prior to capping. It is well known that In segregates readily at these temperatures[20] in quantum wells and we might therefore expect a grading of the dot/GaAs interface during capping with GaAs and an increase in the size of the dots. However, our EDX measurements show that the overgrown dots are smaller by a factor of about two in agreement with AFM studies on partially covered dots.[10] In order to understand this difference we propose that material removed from the dots alloys with the GaAs capping layer between the dots thus explaining the width of the CL. In fact we have observed complete dissolution of QD's by STM when the capping layer is deposited slowly, indicating that the dots are far from inert during the capping process.[5,9,10]

We are pleased to acknowledge many useful discussions with Professor P. J. Goodhew and Dr. P. Maksym.

*Electronic address: r.murray@ic.ac.uk

[1] Y. Arakawa and H. Sakaki, Appl. Phys. Lett. **40**, 939 (1982).

[2] K. Kamath, P. Battacharya, T. Sosnoswki, T. Norris, and J. Phillips, Electron. Lett. **32**, 1374 (1996).

[3] O. G. Schmidt, N. Kirstaedter, N. N. Ledentsov, M.-H. Mao, D. Bimberg, V. M. Ustinov, A. Y. Egorov, E. Zhukov, M. V. Maximov, P. S. Kop'ev, and Z. I. Alferov, Electron. Lett. **32**, 1302 (1996).

[4] H. Shoji, Y. Nakata, K. Mukai, Y. Sugiyama, M. Sugawara, N. Yokoyama, and H. Ishakawa, Appl. Phys. Lett. **71**, 193 (1997).

[5] B. A. Joyce, J. L. Sudijono, J. G. Belk, H. Yamaguchi, X. M. Zhang, H. T. Dobbs, A. Zangwill, D. D. Vvedensky, and T. S. Jones, Jpn. J. Appl. Phys., Part 1 **36**, 4111 (1997).

[6] J. M. Moison, F. Houzay, F. Barthe, L. Leprince, E. André, and O. Vatel, Appl. Phys. Lett. **64**, 196 (1994).

[7] L. Goldstein, F. Glas, J. Y. Marzin, M. N. Charasse, and G. Le Roux, Appl. Phys. Lett. **47**, 1099 (1985).

[8] U. Bangert, A. J. Harvey, S. Gardelis, R. J. Keyse, C. Dieker, and A. Hartmann, Inst. Phys. Conf. Ser. **146**, 61 (1995); **146**, 379 (1995).

[9] Q. Xie, P. Chen, and A. Madhukar, Appl. Phys. Lett. **65**, 2051 (1994).

[10] J. M. Garcia, G. Medeiros-Ribeiro, K. Schmidt, T. Ngo, J. L. Feng, A. Lorke, J. Kotthaus, and P. M. Petroff, Appl. Phys. Lett. **71**, 2014 (1997).
[11] H. Yu, S. Lycett, C. Roberts, and R. Murray, Appl. Phys. Lett. **69**, 4087 (1996).
[12] Q. Xie, A. Madhukar, P. Chen, and N. P. Kobayashi, Phys. Rev. Lett. **75**, 2542 (1995).
[13] U. Woggon, W. Langbein, J. M. Hvam, A. Rosenauer, T. Remmele, and D. Gerthson, Appl. Phys. Lett. **71**, 377 (1997).
[14] W. Wu, J. R. Tucker, G. S. Solomon, and J. S. Harris, Jr., Appl. Phys. Lett. **71**, 1083 (1997).
[15] D. J. Arent, C. Van Hoof, G. Borghs, and H. P. Meier, in *Condensed Systems of Low Dimensionality*, Vol. 253 of *NATO Advanced Study Institute, Series B: Physics*, edited by J. L. Beeby (Plenum, New York, 1991) p. 547.
[16] H. Drexler, D. Leonard, W. Hansen, J. P. Kotthaus, and P. M. Petroff, Phys. Rev. Lett. **73**, 2252 (1994).
[17] S. Malik, C. Roberts, R. Murray, and M. Pate, Appl. Phys. Lett. **71**, 1987 (1997).
[18] A. Rosenauer, U. Fischer, D. Gerthsen, and A. Förster, Appl. Phys. Lett. **71**, 3868 (1997).
[19] K. Mukai, N. Ohtsuka, M. Sugawara, and S. Yamazaki, Jpn. J. Appl. Phys., Part 2 **33**, L1710 (1994).
[20] H. Yu, C. Roberts, and R. Murray, Appl. Phys. Lett. **66**, 2253 (1995).