Vertically Aligned and Electronically Coupled Growth Induced InAs Islands in GaAs

G. S. Solomon,[1] J. A. Trezza,[1] A. F. Marshall,[2] and J. S. Harris, Jr.[1]
[1]*Solid State Laboratories, Stanford University, Stanford, California 94305-4055*
[2]*Center for Materials Research, Stanford University, Stanford, California 94305*
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Multilayer, vertically coupled, quantum dot structures are investigated using layers composed of InAs islands grown by molecular beam epitaxy in the Stranski-Krastanov growth mode. Single, 2, 5, and 10 InAs island layers are investigated in which the 40 Å high InAs islands are separated by 56 Å GaAs spacer layers. The InAs islands are vertically aligned in columns and are pseudomorphic. Between 1 and 10 layers of islands, 8 K photoluminescence shows a 25% reduction in PL linewidth, and a peak shift of 92 meV to lower energy, while transmission electron and atomic force microscopy show the island size in different layers remains constant. These effects are attributed to electronic coupling between islands in the columns, and a simple coupling model is used to simultaneously fit the spectral peak position shift and the linewidth changes.

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In some heteroepitaxially mismatched systems, strain-induced islanding in the Stranski-Krastanov (SK) growth mode can be used to make potentially useful heteroepitaxial islands. In the SK growth mode the mismatched epitaxy is initially accommodated by biaxial compression in a layer-by-layer growth region, traditionally called the wetting layer. After some thickness the strain energy increases and the development of heteroepitaxial islands becomes more energetically favorable than planar growth. Originally, island edges were presumed to be a nucleation source for dislocations. However, coherent, dislocation-free islands are typically observed [1] and theoretical studies [2,3] have shown that these islands are initially both partially relaxed and dislocation free. In the III-V semiconductor material system, the SK growth mode has been used to grow InAs and InGaAs islands on GaAs [4–6], and their size [7,8] and density [9] are well controlled if the InAs or InGaAs coverage is below an isolated island–coalesced island transition [7]. These islands have also been used as stressors [10] to create 3D confinement in lower quantum well regions. Growth-induced islanding may be a possible alternative to lithographically defined 0D quantum systems. Current lithography permits length scales on the order of 500 Å with low fill factors, while InAs islands 100 Å in diameter have been grown on GaAs [7,8] where the island packing fraction is approximately 80% of an ideal close-packed density [9].

There are additional limitations associated with lithographically defined 3D quantum dot arrays since lithography is an inherently 2D process. In this paper we report results of arrays of InAs islands in a matrix of GaAs which are vertically stacked, vertically aligned, and electronically coupled in the growth direction. Islands in different layers show a strong tendency to align vertically. Previously, the vertical alignment of 2 layers of islands has been observed in the degenerative islanding process in InGaAs quantum wells in GaAs, and was believed to be associated with dislocation generation [11]. Here, we report the vertical alignment of up to 10 islanding layers with no associated dislocation generation. These vertically aligned islands have been designed to be vertically connected and electronically coupled, since the 56 Å GaAs spacer layer is approximately equal to the InAs island height.

The epitaxial layers were deposited in a Varian Gen II molecular-beam epitaxy (MBE) system using As_2 (cracked As_4) as the arsenic source and a V/III beam equivalent pressure ratio of 9. The InAs island region and all subsequent depositions were conducted at 500 °C, as measured by the substrate thermocouple, and correspond to 457 °C using a more accurate optical technique [12]. Each InAs island layer is composed of the equivalent of 3 monolayers (ML) of planar InAs deposited at a rate of 0.19 μm/h. The GaAs growth rate was also 0.19 μm/h. The thickness of the GaAs spacer layer between InAs islanding layers is defined a the thickness between adjacent wetting layers, and not the thickness between islands. For the 56 Å GaAs spacer layer, the separation between islands in adjacent islanding layers is approximately 15 Å. Transmission electron microscopy (TEM) analysis was carried out in a Philips CM20 FEG microscope. Photoluminescence (PL) spectra were obtained using Ar^+ ion laser and samples were mounted in a circulation He cryostat maintained at 8 K.

In Fig. 1(a), a high resolution TEM image of 10 layers of InAs dots shows several vertically aligned InAs dot columns. The sample is tilted slightly off the zone axis to reduce strain contrast, so that the dark regions in the layered structure are predominately from the increased scattering of the heavier In atoms. The top InAs layer produces much larger, more rounded, and diffuse islands, which we believe are more heavily alloyed with GaAs than the InAs islands in the lower layers. The presence of a new InAs wetting layer close to the peak of the lower islands inhibits mixing of GaAs with the InAs islands. Atomic force microscopy (AFM) measurements

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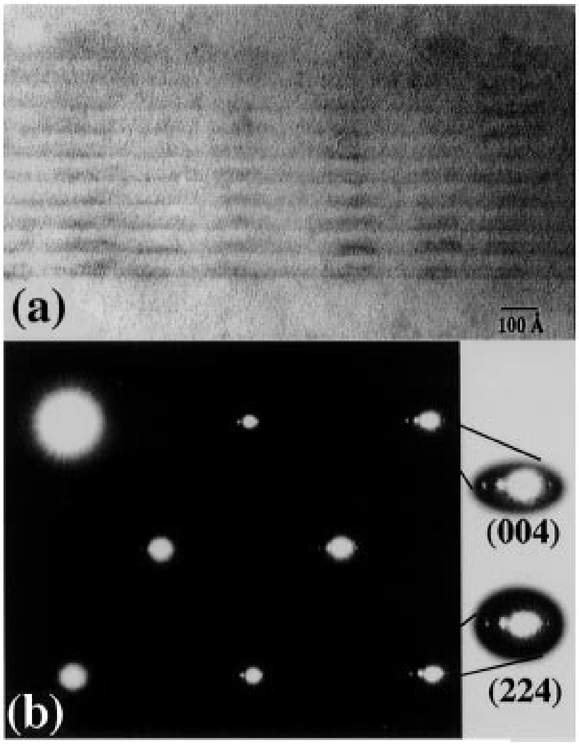

FIG. 1. (a) TEM image of several columns of vertically aligned InAs islands (dark regions). (b) Selected area diffraction pattern in TEM sample in (a) showing superlattice diffraction spots in the (001) growth direction.

indicate the islands are approximately 180 Å wide in the in-plane direction, which is confirmed in Fig. 1(a). AFM measurements also indicate that the in-plane island size does not change when islanding layers are added [13]. Although the small spacer thickness impedes vertical alloying with GaAs, the island shape remains consistent with the (113) sides observed by other workers [14]. Several TEM images made perpendicular to the zone axis to enhance the lattice contrast were investigated for dislocations in the layer island regions; none was observed. Dislocations at the upper interface between the InAs islands and the GaAs cap were investigated by tilting the sample up to 30° off the zone axis, and again no dislocations were observed.

A selected-area diffraction (SAD) pattern is shown in Fig. 1(b). Superlattice and InAs diffraction spots are present in all non-in-plane reciprocal lattice points. Of particular interest are the bright and faint reflections to the left of the GaAs reflections for non-in-plane reflections, for example, the (004) reflection. Our calculations indicate the brighter secondary spots are due to the superlattice unit cell and are determined from the average composition and lattice constant of the superlattice. The faint reflections farther to the left of the GaAs spot are due to InAs and a higher order superlattice reflection. In reciprocal space directions which are not the growth direction (001) or the in-plane direction (011), for example, the (224), the InAs reflections correspond to an in-plane InAs lattice constant that is pseudomorphically strained. Because the InAs islands form aligned columns, there are two separate diffracting regions that are structurally connected: the wetting layer regions without columns and the column regions. High resolution x-ray diffraction (HRXRD) in the [001] direction was used to confirm that the superlattice diffraction results from the pseudomorphically strained wetting layer region. Because of the high island density, if the islands were relaxed through a dislocation mechanism, this relaxation should extend to the wetting layer region, and this is not observed.

In Fig. 2 a high resolution TEM cross section highlights a column of InAs islands, clearly indicating the individual InAs islands and their vertical alignment. The 1.7 ML InAs wetting layer [8] is not observed in Fig. 2, possibly because of its thinness. The island height is approximately 40 Å, but is difficult to accurately determine since at the island peaks only a small number of InAs atoms remain in cross section to contribute to the contrast, and the observed cross section may not intersect the island centers. The lower island interface is flat and abrupt, indicating that GaAs fills in and smooths the islanding interface. As in Fig. 1, the island dimensions in the uppermost islanding layer are much larger and, from the contrast change, appear to be alloyed with GaAs. To

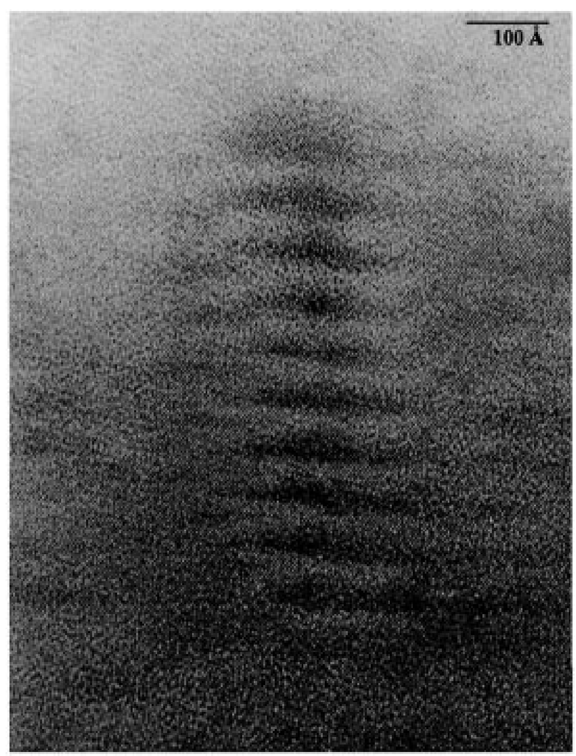

FIG. 2. TEM image showing one column of vertically aligned InAs islands. the Island height is approximately 40 Å and in the in-plane dimension is approximately 180 Å.

the right of the island column in Fig. 2 is a 4 or 5 layer column that does not continue through the upper layers. This decrease in island density after 5 layers is also observed in our AFM results [13].

We believe the local strain relaxation that occurs by islanding in one layer facilitates preferential island formation directly on top of islands in subsequent layers. The islanding process not only reduces the interface energy by limiting the contact of InAs with the GaAs underlayer, but allows the InAs free surface to partially relax. This partial relaxation is expected to be removed as the InAs island is covered by GaAs. However, if only a thin layer of GaAs is added to the islanding layer, the removal of the partial relaxation will not be complete and the strain field from the buried InAs island will extend to the surface leaving the GaAs locally strained and possibly distorted. In either case the region above a buried InAs island will act as a preferential nucleation site for further islands. We note that the InAs wetting layer and islands form a coupled system, yet SAD and XRD indicate that the wetting layer is unchanged by this preferential islanding and stacking process. The TEM and AFM observations that the island size does not change with additional island layering implies that the modification of the local strain state by the buried InAs island is not pronounced enough to measurably change the lattice mismatch.

Results from 8 K PL measurements are shown in Fig. 3. The broad luminescence of the single layer sample is a result of variations in the size of the InAs islands, in which each island contributes a narrow luminescence peak of varying position to create the broad Gaussian PL [15,16]. We observe for increasing layers of InAs islands that the spectral peak position shifts to lower energies, and the spectral linewidth decreases. In comparison to the single island layer, centered at 1.22 eV, the PL peak shifts 73 meV for 5 layers of islands and an additional 19 meV for 10 layers of islands. Since TEM and AFM show that the island size is constant, we attribute the PL peak position shifts to vertical coupling between InAs islands within the columns. There is a 25% reduction in the spectral linewidth between the case of a single islanding layer (72 meV), and the case of 10 islanding layers (54 meV). In all cases the spectral line shape is Gaussian.

The spectral peak shift and linewidth reduction in the single- and multi-islanding layers result from vertical coupling of islands in columns. The tunneling between vertical islands allows carriers to diffuse to the lowest energy dot in the column, resulting in a spectral peak shift to lower energy. However, since the number of dots contributing to the spectra is many orders of magnitude larger than the 10 dots in each column, this effect is not responsible for the spectra changes observed here. Vertical coupling also results in spectral peak shifts and linewidth changes by reducing the ground state confinement energy of each dot column through the formation of a miniband; the sum of the bonding and antibonding states from the overlap of the individual wave functions in a column. We approximate an individual dot as either a sphere or box with infinite potential, so that the linewidth of the ensemble is proportional to $E\sum_i(\Delta X_i/X)$, where ΔX_i and X_i are the variation of the size and the average size of an island in the ith direction, respectively. We then used the tight-binding formalism to model the coupling [17] and resulting energy. In this analysis we fit both the spectral peak position and spectral linewidth using a single coupling parameter (the same coupling parameter for both cases), and assume that conduction band coupling is dominant. The fit to the peak position and spectral linewidth is shown in Figs. 4(a) and

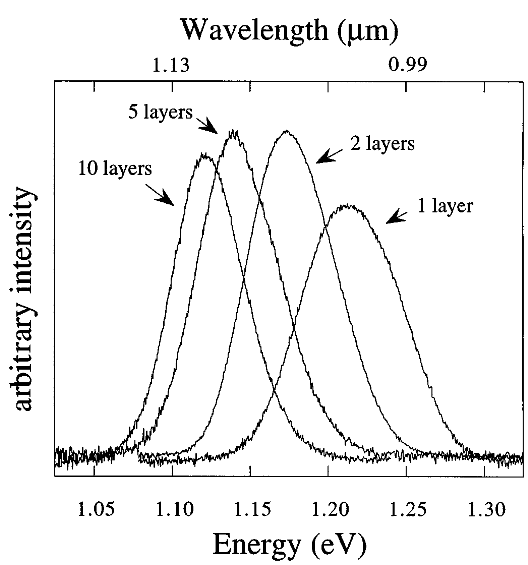

FIG. 3. 8 K photoluminescence of vertically stacked islanding layers showing the variation in spectral peak position and linewidth with the number of stacked islands.

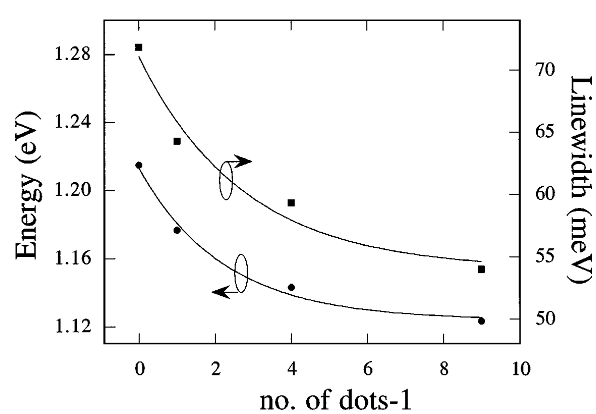

FIG. 4. Changes in spectral linewidth (a) and energy (b) are shown as a function of the number of vertical islanding layers. In each case, the exponential fit uses the same fitting parameter.

4(b). We have found that the peak shift and the reduction in linewidth between the 2, 5, and 10 layer samples can be fully attributed to the lower ground state energy due to coupling. The strongest coupling state results if all of the dots within a column have the same energy, and will produce the largest spectral peak shift. However, the strongest coupling state will not result in the largest reduction in the energy linewidth, since if all of the dots within a column have the same energy, the in-plane size variations will ensure a variation in the lowest miniband state in the ensemble of columns. The weak coupling state, where carriers tunnel through the vertical barriers in a dot column, produces small changes in spectral peak position and linewidth. These changes arise from the increased spatial extent of the dot wave functions and not from the strong overlap of coupled states. An interesting case is the intermediate coupling state, in which the vertical variation in the island size in a single column has the same distribution as the in-plane island size variation in an island layer. This coupling state produces the largest reduction in spectral linewidth since all of the dot columns now have the same size distribution. Since it has been shown that a significant element of the in-plane island size variation is due to small, position dependent flux nonuniformities across the growth surface [18], it is unlikely that the vertical distribution in island size can be made equal to the in-plane island distribution, and thus some significant inhomogeneous spectral broadening will always remain, as is the case here.

An additional source of the spectral redshift and linewidth reduction is that the islands may be increasing in size as the island layers increase or that the islands may be relaxing as the island layers increase, although we have not observed any island size increases with island layers in our TEM or AFM characterization. However, even if such changes were present and too small to observe, we still do not observe luminescence from the lower layers, with presumably smaller islands, in multilayered samples, and weak coupling (tunneling) from islands in a column to the lowest energy state in the column would have to be present. We have investigated this possibility by reducing the GaAs spacer between the InAs dot layers from 56 to 40 Å, where 40 Å is the average dot height. We observe an increased spectral peak shift in the 40 Å spacer layer case with respect to the 56 Å spacer case. This increased peak shift is consistent with coupling.

In summary, we have used a MBE growth-induced islanding process to construct vertically aligned InAs quantum dot structures in GaAs. We have shown that spectral peak shifts of up to 92 meV to lower energy and linewidth reductions of 25% result when 10 InAs island layers are formed with 56 Å GaAs spacer regions. It has been demonstrated that coupling between dots in the vertical columns is responsible for the energy shift and linewidth reduction.

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[1] D.J. Eaglesham and M. Cerullo, Phys. Rev. Lett. **64**, 1943 (1990).
[2] D. Vanderbilt and L.K. Wickham, Mater. Res. Soc. Symp. Proc. **202**, 555 (1991).
[3] B.J. Spencer, P.W. Voorhees, and S.H. Davis, Phys. Rev. Lett. **67**, 3696 (1991).
[4] M. Tabuchi, S. Noda, and A. Sasaki, in *Science and Technology of Mesoscopic Structures,* edited by S. Namba, C. Hamaguchi, and T. Ando (Springer-Verlag, Tokyo, 1992).
[5] D. Leonard, M. Krishnamurthy, C.M. Reaves, S.P. Denbaars, and P.M. Petroff, Appl. Phys. Lett. **63**, 3230 (1993).
[6] Qianghua Xie, P. Chen, A. Kalburge, T.R. Ramachandran, A. Nayfonov, A. Konkar, and A. Madhukar, J. Cryst. Growth **150**, 357 (1995).
[7] G.S. Solomon, J.A. Trezza, and J.S. Harris, Jr., Appl. Phys. Lett. **66**, 991 (1995).
[8] D. Leonard, K. Pond, and P.M. Petroff, Phys. Rev. B **50**, 11 687 (1994).
[9] G.S. Solomon, J.A. Trezza, and J.S. Harris, Jr., Appl. Phys. Lett. **66**, 3161 (1995).
[10] M. Sopanen and H. Lipsanen, Phys. Rev. B **51**, 13 868 (1995).
[11] J.Y. Yao, T.G. Andersson, and G.L. Dunlop, J. Appl. Phys. **69**, 2224 (1991).
[12] W.S. Lee, G.W. Yoffe, D.G. Schlom, and J.S. Harris, Jr., J. Cryst. Growth **111**, 131 (1991).
[13] G.S. Solomon, J.A. Trezza, and J.S. Harris, Jr., Vac. Sci. Technol. B (to be published).
[14] Y. Nabetani, T. Ishikawa, S. Noda, and A. Sasaki, J. Appl. Phys. **76**, 347 (1994).
[15] J.-Y. Marzin, J.-M. Gérard, A. Izraël, D. Barrier, and G. Bastard, Phys. Rev. Lett. **73**, 716 (1994).
[16] S. Fafard, D. Leonard, J.L. Merz, and P.M. Petroff, Appl. Phys. Lett. **65**, 1388 (1994).
[17] In the tight-binding model for a superlattice, $E = E_i + S_i + 2T_i \cos qd$, where E_i is the uncoupled energy in each dot, S_i is the energy change in each dot when an adjacent dot perturbates the barrier potential, and $T_i \cos qd$ is the wave-function overlap term. Here the bandwidth, $4T_i$, is approximated to increase as $e^{-c(d-1)}$, where c is a fitting parameter weighting the overlap and d is the number of coupled dots.
[18] J.M. Gęrard, J.B. Génin, J. Lefebvre, J.M. Moison, N. Lebouché, and F. Barthe, J. Cryst. Growth **150**, 351 (1995).