Vertically Self-Organized InAs Quantum Box Islands on GaAs(100)

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Coherent InAs islands separated by GaAs spacer layers are shown to exhibit self-organized growth along the vertical (i.e., growth) direction. The driving force for such vertically self-organized growth is shown to be the interacting strain fields induced by the islands which give rise to a preferred direction for In migration. A model analysis accounting for the mechanochemical surface diffusion gives an island average size and average separation dependent characteristic spacer layer thickness z_0 below which a vertically self-organized growth occurs.

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The stress on a crystal surface can provide a natural driving force for nanostructure formation via growth on *nonplanar* patterned substrates and three-dimensional (3D) island formation in lattice mismatched growth on *planar* low index substrates such as for InAs on GaAs or GeSi on Si [1]. Coherent InAs island formation on GaAs(100) [2,3] at the earliest (~2 monolayer) stage of deposition provides an *in situ* growth approach to fabrication of optically efficient 3D island quantum boxes [4–6], thus avoiding the potential damage and contamination arising from *ex situ* pixellation techniques. The formation of the islands has been referred to as a "self-assembled" growth [4,5], although no model or mechanism for the same was offered. Achieving regular in-plane spatial distribution and sufficiently uniform island sizes remain challenging issues. We have recently proposed that the interaction between the evolving strain fields induced in the substrate by the evolving islands provides a mechanism and hence a tendency for intraplanar island size equalization [1]. However, given the random initiation of islands, a certain degree of spatial randomness sets in before the island induced strain fields begin to interact, thus introducing spatial and island size nonuniformity. We have also shown [7] that the 3D islands induce strain fields in the protective cap layer grown on top with a cap layer thickness dependent range (l_s) that is as long as ~400 Å. To shed light on the potential role of the evolving and interacting strain fields as a source of driving force for self-assembled growth, we have carried out systematic experiments to examine the role of the island-induced strain fields via deposition of multiple sets of islands separated by spacer layers of varying thicknesses. In this Letter, we present the first evidence for vertical self-organization (i.e., a vertically correlated arrangement) of coherent InAs quantum box islands separated by GaAs spacer layers along the growth direction. A phenomenological model that incorporates mechanochemical diffusion is introduced and shown to give a characteristic spacer layer thickness below which island formation is vertically self-organized, consistent with the experiments.

Details of sample growth via molecular beam epitaxy (MBE) are discussed elsewhere [3]. Two monolayers (MLs) of InAs were deposited on GaAs (100) substrates at 500°C, 6×10^{-6} torr As_4 pressure, and a growth rate of 0.25 ML/s. Then GaAs spacer layers with thin (3 ML) AlAs marker layers after 10 and 20 ML GaAs were deposited at 480 °C, 3×10^{-6} torr As_4 pressure, and a growth rate (for both GaAs and AlAs) of 0.25 ML/s. This combination of 2 ML InAs and GaAs spacer layer was repeated one or more times to provide two or more sets of islands. Samples were also grown with the GaAs spacer layers grown via migration enhanced epitaxy (MEE) as earlier work [6] on single set of islands has shown that the photoluminescence (PL) integrated intensity is then an order of magnitude higher. The reflection high energy electron diffraction (RHEED) pattern turned from spotty to streaky (though slightly nonuniform) again upon ~20 ML GaAs spacer growth, suggesting that the surface after GaAs spacer growth was well recovered. The subsequent growth of 2 ML InAs on such a GaAs surface does not show any evident difference from the growth of the first set of InAs islands. An *in situ, ultra high vacuum* (UHV) atomic force microscope (AFM), UHV interconnected to the MBE growth chamber, was employed for measurements on InAs islands grown under identical conditions. An average island density of $\sim 350/\mu m^2$, average height of 35 Å, and an average lateral size of ~170 Å were found. The volume of a typical InAs island is estimated to be $\sim 2 \times 10^5$ Å^3, equivalent to that of a sphere with radius of ~37 Å. The samples were examined in an Akashi-002B transmission electron microscope (TEM).

Figure 1(a) shows a representative [011] cross-sectional TEM (XTEM) picture of a sample with two sets of InAs islands separated by a MBE grown GaAs spacer of 46 ML. From the strain contrast of the islands in the two sets, each island in the second set is seen to be located on top of another in the first set, indicating a strong one-to-one vertical correlation between the two sets. A detailed statistical analysis on the sample shows that the pairing probability of islands in the two sets, measured from XTEM projected images, is 0.885 ± 0.032. As the GaAs spacer thickness increases to 92 ML [Fig. 1(b)],

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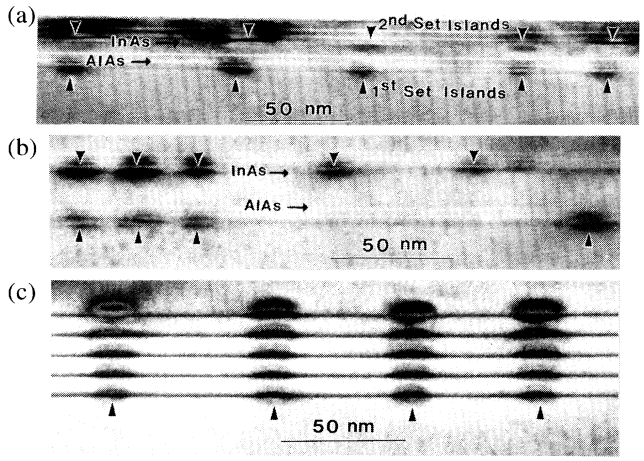
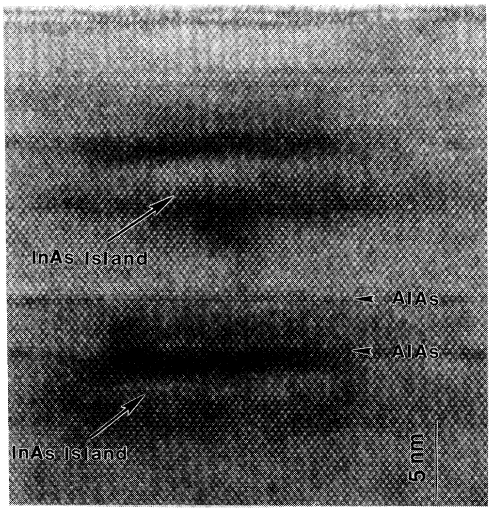


FIG. 1. Typical $g = (400)$ bright field TEM pictures taken along [011] azimuth for the samples with two sets of islands separated by (a) 46 and (b) 92 ML spacer layers, respectively. Arrows point to the island positions indicated by the strain contrast. (c) A typical $g = (200)$ dark field TEM picture for a sample with five sets of islands separated by 36 ML spacer layers. Note the vertical collinearity of the islands in five sets.

the pairing probability decreases to 0.492 ± 0.04. For a sample with five sets of islands and 36 ML thick spacers grown at 400 °C using MEE [Fig. 1(c)], the probability of island pairing to the set just below is maintained at a level of ~0.95 for all adjacent island sets. Figure 2 is a typical high resolution lattice image taken at the [011] azimuth from the sample with the 46 ML spacer and shows the spacer layer to be atomically flat and the islands to be coherent and on top of one another. Figure 3 summarizes the pairing probabilities (squares) obtained as a function



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of the spacer thickness (z_s) from the [011] and [01$\bar{1}$] XTEM azimuths. Three typical regimes can be seen: (1) for small z_s (<36 ML), P is greater than 95%, indicating a nearly completely correlated behavior, (2) a regime of gradual decrease in P, and (3) for large z_s, P saturating at a value corresponding to random overlapping of islands. The filled circles are the calculated results obtained from a phenomenological growth model that accounts for mechanochemical diffusion during island formation and shows that the island vertical self-organization is a consequence of the strain fields created by the lower set of islands. This is discussed next.

The role of strain fields and the ensuing mechanochemical processes have historically been faced in discussing such classic problems as grain growth and precipitation of misfitting metallic clusters in a matrix [8,9]. We have suitably adapted and generalized these classic formulations to provide what, to our knowledge, is the first analytical description of correlated island formation under strain fields during growth. In our case, the kinetic processes giving the vertically self-organized growth behavior are depicted in Fig. 4. Islands in the first set produce a tensile stress in the GaAs above the islands [7] (region I), whereas region II having little or no stress may exist depending upon the average separation l between the first set of islands and the GaAs spacer layer thickness de-

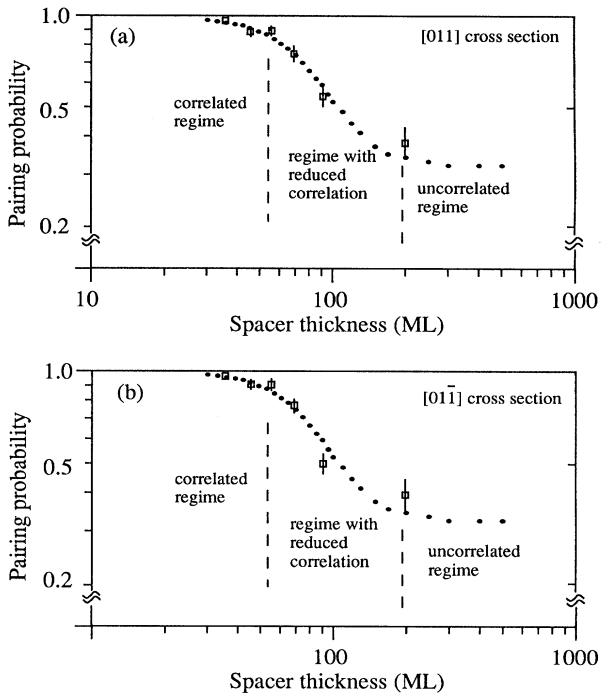

FIG. 3. Experimentally observed pairing probabilities (open squares) as a function of the spacer thickness z_s shown on a log-log plot for samples with two sets of islands (a) [011] cross section, and (b) [011] cross section. The filled circles show least-squares fit of expression (5a) to the data using input parameters $d = 170$ Å and $l = 535$ Å.

pendent range of the surface strain fields, l_s. The In atoms impinging in region I would then be driven by the strain field to accumulate on top of the lower islands, where they can also achieve the lower thermodynamic state energetically due to lower lattice mismatch of InAs with the GaAs in tension. On the other hand, the In atoms impinging in region II may initiate formation of islands in region II, an undesirable feature for achieving the most efficient vertical ordering. The most effective way to suppress island formation in region II is to maintain $2l_s \geq l$ by appropriate choice of the spacer layer thickness, as is the case in Figs. 1(a) and 1(c). To describe the formation of the second set of islands under the mechanochemical diffusion, we consider the surface chemical potential for InAs which may be written as [8,10].

$$\mu(x, y, z = z_s) = \mu_0 + \frac{\Omega}{2E}\sigma_\tau^2 + \gamma\Omega\kappa - \Omega\sigma_{nn}, \tag{1}$$

where μ_0 is the surface chemical potential for an unstressed InAs surface. The second term is from the tangential stress (σ_τ) contribution, the third term comes from the contribution of surface curvature which may be ignored in the present case since the surface is nearly flat, and the fourth term comes from the stress normal to the surface which may also be ignored since no evidence is found for atoms being driven out of the plane. Ω is the atomic volume of InAs. For simplicity, we consider a (1 + 1) dimension case in which the growth direction is along the z axis and the one-dimensional substrate is along the x axis (Fig. 4). The strain driven directed adatom migration is taken to generate effectively a net probability of In adatoms being sucked into the regions $-d/2 < x < d/2$ on top of the islands through the boundaries $x = \pm d/2$. This net probability K, with a dimension of velocity and containing the strength of directional migration, can be expressed as

$$K = \frac{D}{k_B T}\frac{\mu(l_s, z_s) - \mu(0, z_s)}{l_s}, \tag{2}$$

where D is the surface diffusion coefficient of the In adatoms, k_B is the Boltzmann constant, T is the growth temperature and, $[\mu(l_s, z_s) - \mu(0, z_s)]/l_s$ represents the average driving force. Note that D/k_BT is the mobility of the In adatoms. In the case of the present experiments, l (~535 Å) is less than $2l_s$ (~800 Å) so that we replace l_s in Eq. (2) by $l/2$. Considering an average lifetime (τ) for incorporation of In adatoms into the lattice in the region outside the islands (in the case of MBE, the reevaporation of In adatoms is negligible at the growth conditions employed), and adapting the general solution from Schwoebel [11], the adatom concentration at the boundaries $x = \pm d/2$ and the material M_1 arriving in the region $-d/2 < x < d/2$ in unit time, are, respectively, given by

$$n(\pm d/2) = F \times L_D \frac{\sinh Q}{K\cosh Q + V_D \sinh Q}, \tag{3a}$$

$$M_1 = Fd + Kn(d/2) + Kn(-d/2), \tag{3b}$$

where F is the In flux, $L_D = (D\tau)^{1/2}$ is the diffusion length, $V_D = L_D/\tau$, and $Q = (l - d)/2L_D$. We define the island pairing probability as the ratio $P = M_1/M_T$, where $M_T Fl$ is the total material delivered in unit time within length l. To obtain K using Eqs. (1) and (2), we estimate the radial dependence of the island-induced strain field in the GaAs spacer by the strain caused by spherical islands of the same volume [12]. This assumes that an anisotropy in the strain field at the GaAs spacer surface, and hence far from the islands, that may arise from the unknown detailed shape of the islands is small. The strain in an infinite GaAs matrix is therefore a pure shear strain (i.e., $\varepsilon_{rr} + \varepsilon_{\theta\theta} + \varepsilon_{\varphi\varphi} = 0$) without dilation [12]. The $\varepsilon_{xx}^{\mathrm{GaAs}}(x, y = 0, z_s)$ strain component on the surface ($z = z_s$), with the additional correction from an image island due to the existence of a free surface, is then given by [13]

$$\varepsilon_{xx}^{\mathrm{GaAs}}(x, y = 0, z_s) = 2A\varepsilon_0\frac{r_0^3}{r^3} = 2A\varepsilon_0\frac{r_0^3}{(x^2 + z_s^2)^{3/2}},$$

$$A = \frac{3B_{\mathrm{InAs}}}{3B_{\mathrm{InAs}} + 2E_{\mathrm{GaAs}}/(1 + \nu_{\mathrm{GaAs}})}, \tag{4}$$

where r_0 is the radius of the equivalent spherical island, r is the distance from the center of the island, B_{InAs} is the bulk modulus of InAs, E_{GaAs} (ν_{GaAs}) is Young's modulus (Poisson's ratio) of GaAs, and ε_0(~7%) is the natural lattice mismatch between GaAs and InAs. Considering only the dominant strain term $\varepsilon_{xx}^{\mathrm{GaAs}}(x, z_s) = \varepsilon_0 - \varepsilon_{xx}^{\mathrm{GaAs}}(x, z_s)$, and combining formulas (1)–(4), the pairing probability and a characteristic length z_0 are derived as [14]

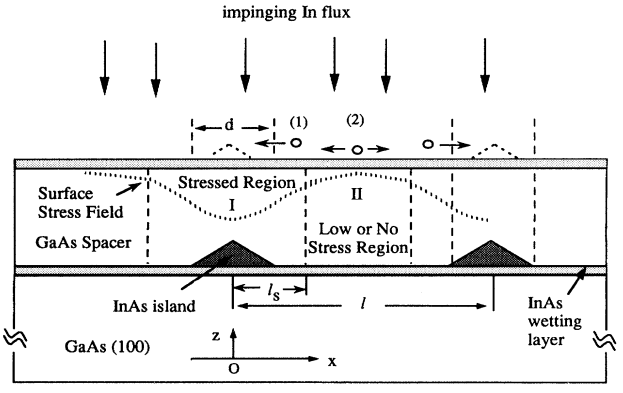

FIG. 4. A schematic representation showing the two major processes for the In adatom migration on the stressed surface: (1) directional diffusion under mechanochemical potential gradient contributing towards vertical self-organization and (2) largely symmetric thermal migration in regions from the islands contributing to initiation of new islands not vertically aligned with islands below.

$$P = \frac{d}{l} + \left(1 - \frac{d}{l}\right)\frac{1}{Q}\,\frac{\sinh Q}{\cosh Q + (z_s/z_0)^3\{(l^2/4z_s^2 + 1)^{3/2}/[(l^2/4z_s^2 + 1)^{3/2} - 1]\}\sinh Q}, \tag{5a}$$

$$z_0 = r_0\left(\frac{8L_D}{l}\,\frac{W}{k_BT}A\right)^{1/3}, \qquad W = \frac{\Omega_{\mathrm{InAs}}}{2E_{\mathrm{InAs}}}(C_{11}^{\mathrm{InAs}}\varepsilon_0)^2. \tag{5b}$$

Utilizing the definition of Q and Eq. 5(b), we replace Q in Eq. 5(a) in terms of z_0. Then Eq. 5(a) involves the quantities d, l, W, A, and z_0. For d and l we use the values determined via the AFM measurements and W (0.0557 eV) and A (0.572) are calculated using the bulk elastic constant of InAs and GaAs. Then treating the only remaining unknown z_0 in Eq. 5(a) as a parameter, the data of Figs. 3(a) and 3(b) are found to be simultaneously fitted for $z_0 = 35.6$ ML. From this fitted value of z_0 we extract, using Eq. 5(b), a value of ~ 0.28 μm for L_D, which is reasonable and consistent with the few attempts to determine In diffusion length during InAs growth on GaAs. For large z_s, Eq. (5a) correctly gives P as d/l representing a random overlapping of the islands. We note as a general feature that, with increasing diffusion length L_D, strain strength W, and the island size r_0, and with a decrease in interisland distance in the first set, the range for correlation (z_0) increases.

In summary, we have demonstrated a vertically self-organized growth of InAs quantum box islands. A phenomenological model based upon the nature of island-induced strain fields and their impact on the directed nature of mechanochemical diffusion is proposed and analyzed. It is shown to consistently account for the observations, thus providing the first clear evidence that the island-induced evolving strain fields provide the driving force for self-assembly or self-organization in island systems. The island vertical pairing probability P as a function of the spacer thickness shows three typical regimes manifesting the decaying nature of the strain fields. A characteristic length z_0 for vertically self-organized growth to occur is inferred from this model and is found to be consistent with the experimental data. The strong vertically self-organized islands open a new front for studying the novel properties of nanostructures, such as the coupled InAs quantum boxes. Work along this line is underway.

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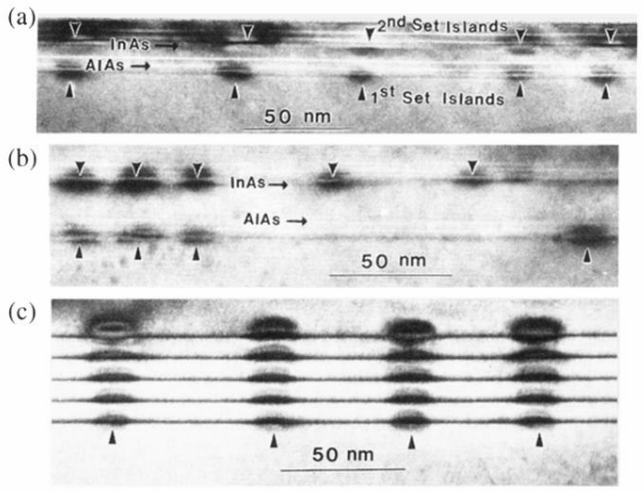
[14] Since $r_0/z < 0.5$, terms of order higher than $(r_0/z)^3$ have been dropped in obtaining μ from Eq. (4).



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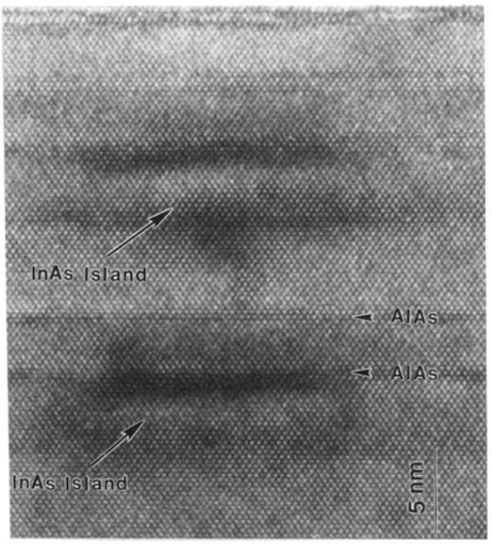

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