APPLIED PHYSICS LETTERS VOLUME 79, NUMBER 20 12 NOVEMBER 2001

Stable island arrays by height-constrained Stranski–Krastanov growth

J. Liang and Z. Suo[a]
Princeton Materials Institute and Department of Mechanical and Aerospace Engineering, Princeton University, New Jersey 08544

(Received 23 January 2001; accepted for publication 24 August 2001)

In the Stranski–Krastanov system, the lattice mismatch between the film and the substrate causes the film to break into islands. During annealing, both surface energy, and elastic energy drive the islands to coarsen: some islands enlarge and others shrink, keeping the total island volume constant. The islands produced this way are usually uneven in size and spacing. Motivated by several related studies, we suggest that stable, uniform islands should form when a stiff ceiling is placed at a small gap above the film. After contacting the ceiling, the islands are constrained to grow laterally and remain coherent with the substrate, preventing further stress relaxation. In fact, we show that the role of elasticity is reversed: with the ceiling, the total elastic energy stored in the system increases as the islands coarsen laterally. On the other hand, the total surface energy decreases as the islands coarsen. Consequently, the islands select an equilibrium size to minimize the combined elastic energy and surface energy. We estimate the equilibrium island size by analyzing an idealized model. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1415403]

The Stranski–Krastanov growth has been used to fabricate nanoscale semiconductor particles as quantum dots in optoelectronic devices.[1,2] A thin film of one crystal is deposited on a substrate of another crystal. The two crystals belong to the same symmetry group, have different lattice constants, and form a coherent interface. The lattice mismatch induces an elastic field. To lower the elastic energy, the film breaks into islands. The islands so grown are usually uneven in size and spatial arrangement. The nonuniformity causes a spread in the electronic energy levels, which limits potential devices. Uniformity improves to some extent when multiple stacks of islands are grown.[3–5] Despite intense efforts over the last decade, it remains a challenge to fabricate uniform quantum dots by the Stranski–Krastanov growth.

Because islands nucleate at random times and sites, the initial islands are uneven in size and spatial arrangement. On annealing, the islands coarsen: some islands grow and others shrink; over time, the mean island size increases, but the number of the islands decreases.[6,7] Random nucleation and coarsening are basic causes for island nonuniformity. In practice the two processes, deposition and annealing, can be concurrent. This letter focuses on annealing by itself. That is, after a certain amount of matter is deposited on the substrate, the flux is stopped, but atoms diffuse on the surface to transport matter among islands.

It is well known that the surface energy drives coarsening. Coarse islands reduce the collective surface area, and thereby reduce the total surface energy. It is less appreciated that, in the Stranski–Krastanov growth, elastic energy also drives coarsening. This is understood as follows. Let the average thickness of the deposited material be h. To simplify the argument, imagine that the islands are all identical and form a periodic lattice. Let the base diameter of each island be d, and the spacing between neighboring islands be l. When the islands coarsen, both d and l increase, but the islands are assumed to maintain a similar three dimensional shape and a similar periodic lattice. Volume conservation requires that $d^3=\beta l^2 h$, where β is a dimensionless number depending on the island shape and the type of the island lattice. Consequently, $l/h=\beta(l/d)^3$. When the islands coarsen, l/h increases, and so does the spacing-to-diameter ratio l/d. Both the film and the substrate are taken to be cubic crystals, with lattice constants a_f and a_s, respectively. The mismatch strain is $\varepsilon_M=(a_s-a_f)/a_s$. Because the total volume of the islands remains constant, we need only consider the average elastic energy (i.e., the total elastic energy stored in the system divided by the volume of the deposited matter). The average elastic energy takes the form

$$U_e=E\varepsilon_M^2 g(l/d), \tag{1}$$

where E is a representative elastic modulus, and $g(l/d)$ is a dimensionless function, which also depends on ratios of other elastic moduli to E. The elastic energy can be calculated from a unit cell containing a single coherent island on a substrate of the shape of a long prism, with zero displacement normal to the sides of the prism, as required by the periodic boundary conditions. For a fixed island diameter d, the larger the cell size l is, the more compliant the system is, and so can the more elastic energy be relaxed. Consequently, the function g decreases as l/d increases. This trend has been shown by the finite element calculation using an axisymmetric unit cell.[8] Combining the above considerations of volume conservation and elastic relaxation, we conclude that the total elastic energy decreases when the islands coarsen. Islands sometimes change shape during annealing, which does not appear to stop coarsening. The shape change is ignored in this letter. We also neglect energy change of other origins, such as surface stress and line tension, which may play roles when the islands are very small.[9]

Coarsening can be stopped by hindering stress relaxation, e.g., by constraining the height of the islands. We are unaware of any experimental demonstration of the height-constrained Stranski–Krastanov growth. Several related studies, however, have motivated this letter. Chou and

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[a] Electronic mail: suo@princeton.edu

Zhuang[10] deposited a thin polymer film on a substrate, and placed a stiff ceiling at a small gap above the film. Upon heating, the polymer flowed and self-assembled into a triangular lattice of islands, their height being constrained by the ceiling. Electrostatics was implicated for inducing the self-assembly. The gap between the ceiling and the substrate was of order 100 nm. The gap was controlled by the height of spacers, which could be made below 100 nm. In another area of research, it was observed that when a small amount of matter was deposited on a substrate, monolayer-thick islands might form, having a uniform size and ordering into a periodic lattice.[11–14] The phenomena were attributed to the competition between the elastic energy and the edge energy.[15,16]

We next describe the height-constrained Stranski–Krastanov growth, following the setup for polymers.[10] First deposit a crystalline film on a crystalline substrate. The two crystals have different lattice constants, but are coherent. The film need not be perfectly flat, h being the average thickness. Place a stiff ceiling at a spacing s from the substrate surface, e.g., by wafer bonding the ceiling to a spacer fabricated on the substrate. The ceiling can even be a functional part of the final device. To prevent impurities from being trapped in the structure, wafer bonding should be carried out in an ultrahigh vacuum. The film is then annealed to break into islands. After contacting the ceiling, the islands grow in two dimensions. The materials are so selected that, during annealing, atoms of the film diffuse on the surface, but atoms of the substrate and the ceiling remain immobile. The growing islands remain coherent with the substrate. It may be necessary that the ceiling be amorphous, or of the same crystal symmetry, so that the ceiling does not affect the crystalline perfection of the islands. From now on we will only consider the process after the islands contact the ceiling.

The ceiling reverses the role of elasticity: the elastic energy now drives the islands to refine, rather than coarsen. This trend is readily understood by considering two limits: pancakes and needles. When the islands are pancakes ($d/s \gg 1$), the stress state approaches that of the biaxial stress in the blanket film. When the islands are needles ($d/s \ll 1$), the stress in the system vanishes. One can also see readily that the total surface energy decreases when the islands coarsen. It is the competition between the refining driven by elastic energy and coarsening driven by surface energy that results in an equilibrium island size.

To estimate the equilibrium island size, we now analyze an axisymmetric model. The inset in Fig. 1 illustrates a circular disk of diameter d lies on an infinitely long cylinder of diameter l. They represent an island and a unit cell of the substrate. Cylinders do not fill space, unlike prisms. The elastic energy stored in an axisymmetric cell, however, should not differ too much from that in a prismatic cell, so long as the two cells have the same volume. Thus, l represents the effective spacing between the neighboring islands. The ratio $C = h/s$ sets the volume fraction of the matter in the gap between the substrate and the ceiling, and remains constant during annealing. Volume conservation requires that $d = l\sqrt{C}$. As islands coarsen the spacing-to-diameter ratio l/d remains constant, in contrast to the three dimensional islands considered above. The curved surface of the substrate has no radial displacement and shear stress. The ceiling exerts a capillary pressure on the top surface of the island. The capillary pressure is much smaller than the typical stress caused by the lattice mismatch, and should have a small effect on elastic energy, so that we take the top surface to be traction free. This letter does not consider the case where the island is also coherent with the ceiling, although the principle remains the same.

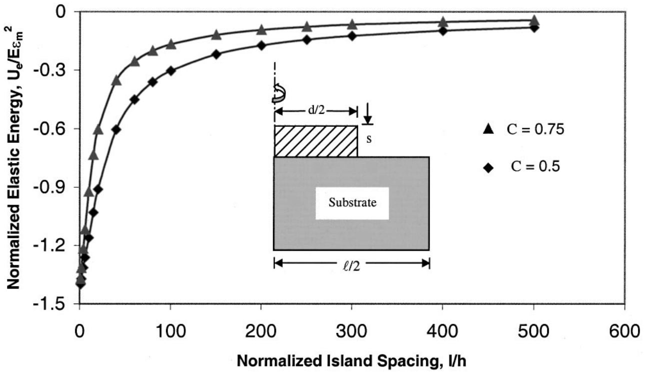

FIG. 1. Inset sketches the axisymmetric model. The island is represented by a circular disk, and the substrate by an infinitely long cylinder. The normalized elastic energy is plotted as functions of l/h at two values of the volume fraction. Both curves monotonically increase, and asymptote to zero as $l/h \rightarrow \infty$.

Assume that the film and the substrate have identical elastic constants and are isotropic, with Young's modulus E and Poisson's ratio ν. The average elastic energy now takes the form

$$U_e = E\varepsilon_M^2 f(C, l/h), \tag{2}$$

The dimensionless function f also depends on Poisson's ratio. We will take the blanket film as the energy reference state, so that $f(C,\infty) = 0$ for pancakes, and $f(C,0) = -1/(1-\nu)$ for needles. When the islands are between the two limits, the stress field cannot be determined analytically. We use the finite element package ABAQUS instead. The mismatch strain ε_M is represented by assigning different coefficients of thermal expansion to the two crystals, and then subjecting the system to a temperature rise. Poisson's ratio is assigned to be $\nu = 0.3$. Figure 1 shows the normalized elastic energy as a function of the normalized island spacing for $C = 0.5$ and $C = 0.75$. Indeed, the functions increase monotonically. Both approach the analytic value as $l/h \rightarrow 0$, and to zero as $l/h \rightarrow \infty$.

Next consider the change in the surface energy during annealing. In the idealized model, the edges of the islands are taken to be vertical. As the islands coarsen laterally, the volume of all islands remain constant, so that the area is constant for each of the following interfaces: islands–substrate, islands–ceiling, substrate–vacuum, and ceiling–vacuum. Consequently, the surface energies of these interfaces do not affect the energetics of the island coarsening. Wetting layers, of a few monolayer thickness, may cover the substrate–vacuum interface and the ceiling–vacuum interface. Because these interfaces have constant areas as the islands coarsen laterally, the presence of these wetting layers does not affect the energetics of coarsening. The change in the surface energy is the surface energy density of the island–vacuum interface, γ, times the total area of the island edge. The change in the surface energy per unit volume is

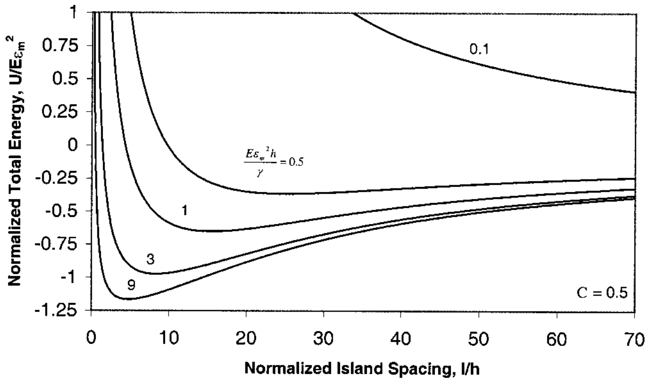

FIG. 2. Combined surface and elastic energy as a function of l/h. When the Griffith number is sufficiently large, the combined energy reaches a minimum, selecting an equilibrium island spacing.

$$U_s=\frac{4\gamma}{l\sqrt{C}}. \tag{3}$$

The surface energy decreases as the islands coarsen.

The combined surface energy and elastic energy, $U=U_s+U_e$, takes the dimensionless form

$$\frac{U}{E\varepsilon_M^2}=\frac{4}{(l/h)\Lambda\sqrt{C}}+f(C,l/h). \tag{4}$$

The Griffith number, $\Lambda=hE\varepsilon_M^2/\gamma$, measures the relative importance of the elastic energy and the surface energy.

Figure 2 plots the combined energy as a function of the normalized island spacing for $C=0.5$. When the Griffith number is small, the surface energy prevails, and the combined energy decreases monotonically as the island spacing increases, so that the islands coarsen indefinitely. When the Griffith number is sufficiently large, the combined energy reaches a minimum, selecting an equilibrium island size. Figure 3 plots the equilibrium island spacing, l_0, as a function of the Griffith number, at two values of the volume fraction. The equilibrium island diameter is given by $d_0=l_0\sqrt{C}$. The height-constrained growth offers is a means to tune the island size and spacing. The average film thickness h is controlled by the amount of deposited matter. The volume fraction C is controlled by the gap between the substrate and the ceiling. The Griffith number can be varied by varying the mismatch strain, or by varying the surface energy with a surfactant. For InAs on GaAs, taking $E=1.3\times10^{11}$ N/m^2, $\gamma=1$ N/m, $\varepsilon_M=7\times10^{-2}$, and $h=2\times10^{-9}$ m, we find that the Griffith number is $\Lambda=1.3$. For the volume fraction $C=0.75$, Fig. 3 gives the equilibrium island spacing $l_0\approx20$ nm. The equilibrium island diameter is $d_0\approx17$ nm, and the gap between the substrate and the ceiling is $s=2.67$ nm.

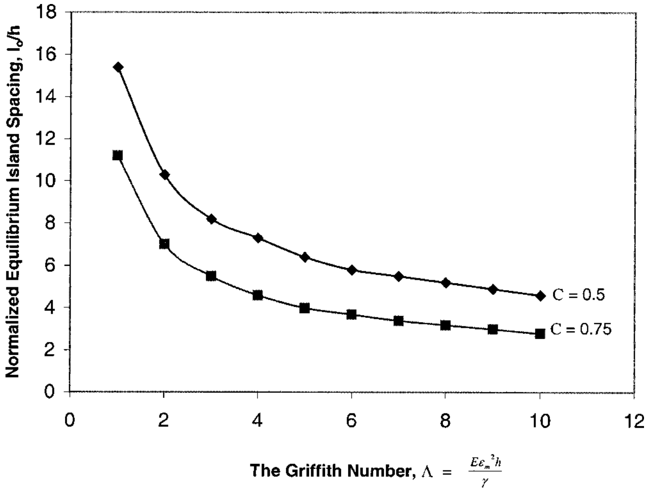

FIG. 3. Equilibrium island spacing as a function of the Griffith number for two values of the volume fraction.

In summary, we have described an approach to grow uniform islands, and analyzed an idealized model to help plan experiments. The model determines the equilibrium island size by minimizing the combined surface and elastic energy. Consequently, the islands so grown circumvent the randomness in nucleation and annealing. Although the model does not address the issue of spatial ordering, on the basis of the observations of monolayer-thick islands,[11–16] we expect that the elastic interaction will order the equal-sized islands into a periodic lattice. Furthermore, several intriguing possibilities demonstrated by Chou and Zhuang[10] for polymers have analogs for the Stranski–Krastanov systems. The ceiling can be prescribed with a coarse pattern of variable heights by using conventional lithography. Island arrays of different sizes may form on the same wafer. We have also developed a model for the electrostatics-driven self-assembly.[17] We hope that experiments will soon succeed in realizing the potential of this approach.

This work is supported by the National Science Foundation through Grant No. CMS-9820713.

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