Three-dimensional kinetic Monte Carlo simulation of prepatterned quantum-dot island growth

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A special prepatterning method is proposed for spatially ordered self-organizing quantum dots on anisotropic semiconductor substrates. Using three-dimensional kinetic Monte Carlo simulations, atoms are deposited with varying intermediate interruption times. We demonstrate the effect of interruption time and long-range anisotropic strain energy on island size uniformity and lateral alignment. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2812572]

Self-organized semiconductor quantum dots (QDs) have attracted large interest due to their improved[1] and unique[2,3] physical properties that may be exploited in devices such as lasers, detectors, and memories.[4–6] Those grown through the Stranski-Krastanov mode are promising candidates for use in quantum devices because of their defect-free properties and ease of fabrication.[7] However, physical properties are sensitive to the ordering on the surface. Thus, controlling lateral spacing and size distribution of the islands would enable improved device design through indirect control of correlation effects and improved vertical alignment of stacked layers.[8]

Various methods have been proposed recently for patterning the substrate so that uniform lateral ordering and size distribution can be achieved. For instance, surface modification techniques, such as photolithography or nanoindentation, have been used to pattern the substrate[9] where the modifications (using individual atoms, small clusters, or generating mesas or holes) can act as nucleation sites and areas of preferential island formations.[10,11] However, the dots from lithographic techniques are often too large and not of sufficient spatial density for device application.[12,13] The most promising quantum structures so far have been fabricated using techniques based on direct crystal growth.[14] Strain-relief patterns are created spontaneously when a material is deposited on a substrate with a different lattice constant.[15,16] It is also possible to confine growth by using templates or nanocavity arrays.[17,18] A brief review of various prepatterning methods can also be found in Kiravittaya *et al.*[19]

Spatial ordering of QD islands can also be improved by employing interlayer strains through vertical stacking[20] or by using high index substrates.[21,22] It is particularly interesting that uniform spatial distributions of QD chains can also be obtained by adjusting the deposit material coverage and growth interruptions.[23]

In this letter, we present computer simulations based on a three-dimensional (3D) kinetic Monte Carlo (KMC) method for QD island self-organization on prepatterned substrates where the pattern control is achieved by adjusting the growth interruption time. With consideration of the long-range elastic strain energy[24] and under fixed growth parameters, spatially ordered QD island patterns are computationally predicted. Furthermore, correlation between the island pattern and substrate anisotropy (due to different crystalline orientations) is observed.[25,26]

Our 3D layer-by-layer KMC growth model is developed from the two-dimensional (2D) (x,y)-plane growth model.[27] The 2D hopping probability of an atom from one lattice site to a nearest or next nearest neighbor site in the (x,y) plane is still governed by the Arrhenius law enhanced by the long-range strain energy field.[28,29]

$$p = \nu_0 \exp\left(-\frac{E_s + E_n - E_{\rm str}(x,y)}{k_B T}\right),$$

where ν_0 is the attempt frequency ($=10^{13}\ {\rm s}^{-1}$), T the temperature, k_B the Boltzmann constant, E_s and E_n are the binding energies to the surface and to the neighboring atoms, respectively, and $E_{\rm str}(x,y)$, which is a function of the plane coordinates (x,y), is the energy correction from the long-range strain field due to the lattice mismatch between the substrate and the deposited material. Furthermore, in order to simulate the 3D adatom diffusion, the surface binding energy is modified to include the effect of the surface geometry.[30] We remark that our model is multiscale based where the continuum long-range strain is considered through the strain energy function in the atomistic KMC simulation. Computer modeling of QD island growth over prepatterned substrates employing pure continuum mechanics was also reported recently.[18,31]

In simulating the effect of the growth interruption on the QD island distribution, three cases are studied. In case 1, we deposit 1.6 ML InAs atoms to the GaAs substrate and after that, we give the system an interruption time of 250 s (Fig. 1). In case 2, after depositing 0.3 ML InAs atoms to the GaAs substrate, we interrupt growth for 50 s (Fig. 2) after which InAs atoms are deposited until the total coverage reaches 1.6 ML. Finally, we let the system self-assemble for 200 s (Fig. 3), which gives a total interruption time of 250 s.

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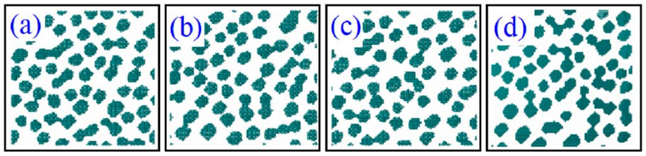

FIG. 1. (Color online) InAs islands on a 100×100 grid substrate with 1.6 ML coverage (top-down plan view). (a) Isotropic substrate, (b) GaAs (001), (c) GaAs (111), and (d) GaAs (113). Fixed growth conditions $T=800$ K, flux rate $F=0.1$ ML/s, and interruption time $t_i=250$ s.

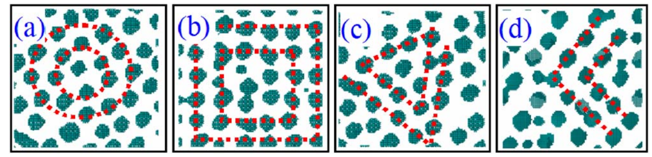

FIG. 3. (Color online) InAs islands on a 100×100 grid substrate with a total coverage of 1.6 ML coverage, grown from the island pattern in Fig. 2 (top-down plan view). Fixed growth conditions $T=800$ K, $F=0.1$ ML/s, and $t_i=200$ s after the total coverage of 1.6 ML. Therefore, the total interruption time is still 250 s.

In case 3, deposition of the InAs atoms to the GaAs substrate is interrupted for 50 s at the end of graduated coverages of 0.3, 0.6, 0.9, 1.2, and 1.6 ML (Fig. 4). Therefore, the total interruption time is still 250 s. To study the correlation between the QD island pattern and the substrate anisotropy, four different substrates are selected: isotropic, GaAs (001), GaAs (111), and GaAs (113). We also remark that the InAs and GaAs atomic species are indistinguishable in our 3D KMC, but the different lattice constants and material properties between them are utilized in the long-range strain energy calculation.[30]

Figure 1 shows, for case 1, the QD islands distributed on a 100×100 grid over the isotropic substrate and the GaAs substrate with different crystalline orientations. The 1.6 ML was deposited once without intermediate interruption. After the total deposition, the system is annealed for an interruption time $t_i=250$ s at $T=800$ K and a flux rate of $F=0.1$ ML/s. It is clearly observed from Fig. 1 that, even though the long-range strain energy is included in the growth, the spatial pattern remains irregular.

Similarly, Fig. 2 shows the distribution on a 100×100 grid over the isotropic substrate and the GaAs substrate having varying crystalline orientations. In contrast to Fig. 1, here only 0.3 ML coverage is deposited initially and the process is then interrupted for 50 s for annealing. By depositing a smaller amount of the atoms on the substrate, well organized patterns can be achieved, with the latter being able to be used as prepattern for further deposition, as we will show below for cases 2 and 3.

Figure 3 shows, for case 2, the island distribution on a 100×100 grid over the isotropic substrate and the GaAs substrate with different crystalline orientations. Here, the prepattern obtained in Fig. 2 is utilized for continuous deposition. In other words, after deposition of 0.3 ML followed by 50 s interruption, the remaining 1.3 ML was deposited and the system is then annealed for 200 s so that the total interruption time is still 250 s. Comparing Fig. 3 to Fig. 1, it is observed that introducing an intermediate interruption time during growth can help us to achieve better lateral alignment. This conclusion is similar to the recent experimental observation,[23] and it is also consistent with other previous experimental and simulation results. It has been demonstrated that a growth interruption has a smoothing effect on crystal surface,[32] and that after the growth interruption, the system exhibits an ordered pattern.[33] Therefore, it could be possible to control the QD island pattern by adjusting the growth interruption time. In contrast to the vertical stacking technique[20] for improved surface order, this method can be applied to a single growth layer.

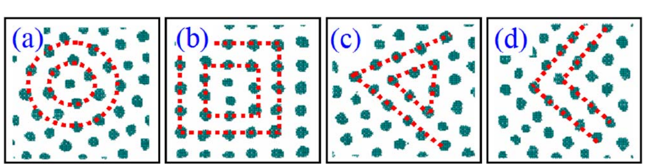

FIG. 2. (Color online) InAs islands on a 100×100 grid substrate with 0.3 ML coverage using the same substrates as in Fig. 1 (top-down plan view). Fixed growth conditions $T=800$ K, $F=0.1$ ML/s, and $t_i=50$ s. This QD pattern is used for continuous deposition of atoms in cases 2 and 3.

We show in Fig. 4 the spatial distribution of the self-organized QD islands on different substrates for case 3 (bottom row for the top-down plan view). Shown in the top row is also the strain energy distribution on the surface of the substrate. The 3D KMC simulation is also on the 100×100 grid at $T=800$ K and $F=0.1$ ML/s, as in cases 1 and 2. However, the deposition is interrupted for 50 s at the end of coverages of 0.3, 0.6, 0.9, 1.2, and 1.6 ML, keeping the total interruption time at 250 s. Comparing the spatial distribution in Fig. 4 with those in Figs. 1 and 3, we observe that improved spatial ordering can be achieved with increasing intermediate interruption steps. Comparing the strain energy distribution in the top row with the growth patterns in the bottom row in Fig. 4, we also notice a clear correlation between the growth pattern and substrate anisotropy (or orientation). This result is consistent with other recent reports.[20,27]

The proposed prepatterning approach cannot only improve the spatial ordering of the QD islands but also make the island size more uniform on a single growth layer. This is shown in Fig. 5 where the histograms for the number of islands versus island size are presented. The first, second, third, and fourth rows are, respectively, the results for the growth over the isotropic, GaAs (001), GaAs (111), and GaAs (113) substrates, while the left, middle, and right columns are, respectively, for cases 1, 2, and 3. Comparing the

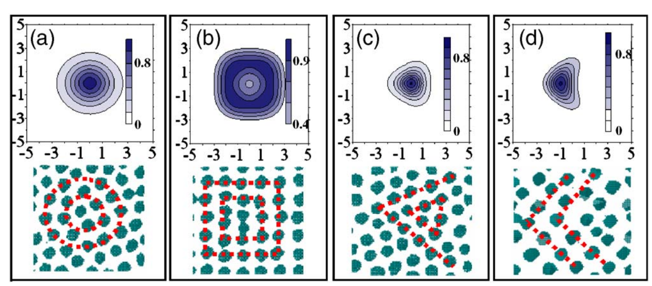

FIG. 4. (Color online) InAs islands on a 100×100 grid substrate with a total coverage of 1.6 ML using same the substrates as in Fig. 1. Top and bottom rows are, respectively, the strain energy distribution on the substrate and top-down plan view. Fixed growth conditions $T=800$ K and $F=0.1$ ML/s. Deposition is interrupted at $t_i=50$ s at the end of coverages of 0.3, 0.6, 0.9, 1.2, and 1.6 ML with total interruption time of 250 s.

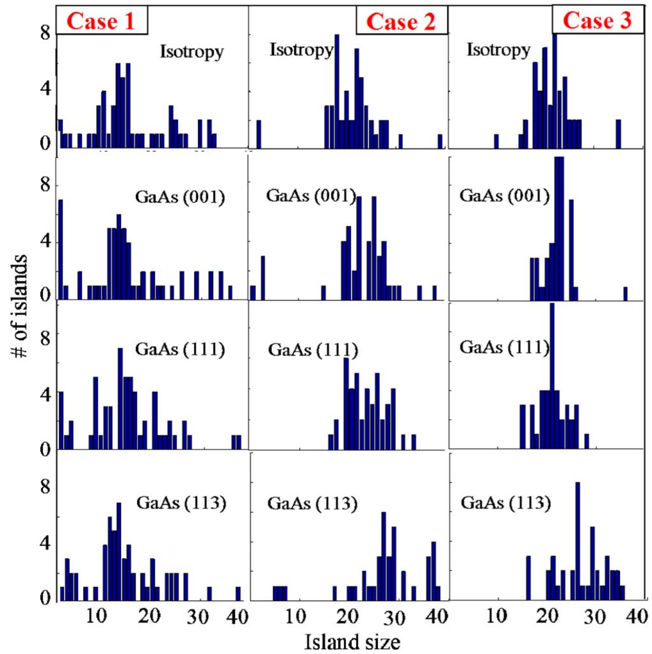

FIG. 5. (Color online) Histograms of number of QD islands vs island size for InAs QD growth of cases 1 to 3 (columns left, middle, and right) on the isotropic, GaAs (001), GaAs (111), and GaAs (113) substrates (rows from top to bottom). The growth is on a 100×100 grid over the substrate with a total coverage of 1.6 ML. Fixed growth conditions $T=800$ K and $F=0.1$ ML/s. Case 1 is for without intermediate interruption; case 2 is for an intermediate interruption of 50 s after 0.3ML coverage; and case 3 is for intermediate interruption time of 50 s at the end of coverages of 0.3, 0.6, 0.9, 1.2, and 1.6 ML.

results column by column, we observe a pronounced island size normalization effect by adding the intermediate interruption time.

Using a recently proposed 3D KMC algorithm for QD self-assembled growth, we simulated the effect of prepatterned substrates on lateral alignment on a single layer, where the prepattern template is created by interrupting the growth process and permitting intermediate annealing. By introducing interruption times, islands with greater size and shape uniformity and better alignment can be achieved. The correlation between the pattern and substrate anisotropy (orientation) was also observed. Our results suggest that in MBE growth of QD islands, intermediate interruption times can be introduced. This experimentally specifiable parameter could enable an additional means of controlling 3D QD island array alignment and uniformity.

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