COMPUTER SIMULATION OF NUCLEATION ON PATTERNED SURFACES

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ABSTRACT

We have studied qualitatively the effect of patterned substrate surface on adatom nucleation using Kinetic Monte Carlo simulations and solid-on-solid model. The inhomogeneity in the activation energies of adatom diffusion was incorporated into the simulations using two models: a) periodic variation of the adatom–substrate interaction energy as a function of the adatom lateral position and b) adding a direction dependent and periodically varying diffusion barrier energy to the adatom diffusion activation energy. The effect of the patterned surface was clearly manifested as a confinement of nucleated islands and, consequently, narrowing the island size distribution. The effect was strong in a narrow temperature range.

INTRODUCTION

Atomic or near-atomic scale structures are currently intensively studied because of their possible technological applications [1]. However, most applications require a large number of these structures with as little variation as possible in their shape and size.

One method to manufacture semiconductor quantum dots is the utilization of spontaneous self-organization of islands in heteroepitaxial thin film growth [1]. By growing several layers of these island ensembles on each other, the size distribution can be made narrower [2, 3]. This vertical correlation of quantum dots can in some cases be explained by the effect of strain caused by the underlying islands on the further growth of islands, i.e. nucleation and growth on a patterned surface.

In some heteroepitaxial systems the strain due to lattice mismatch is relieved by formation of domains that are separated by a regular dislocation network [4]. One example is 2 monolayers (ML) of Ag on Pt (111) [5]. Further deposition of atoms on this structure produces preferred nucleation in the centers of the domains and a narrow size distribution of adatom islands. In this case the confinement effect on the adatoms is caused by the repulsive adatom-dislocation interaction.

The objective of this study is to investigate qualitatively the effect of the spatial variation of the adatom diffusion activation energies on their nucleation. The models presented are chosen so that the above-mentioned cases may be investigated in more detail in the future.

COMPUTATIONAL DETAILS

The evolution of the surface was described by using the Kinetic Monte Carlo (KMC) simulation method [6] and cubic solid-on-solid (SOS) model. Deposition of adatoms randomly on the surface was performed at a constant deposition rate F (ML/s). The diffusion of each adatom was described by the hopping probability P

$$P = ke^{-E/k_{\mathrm{B}}T}, \tag{1}$$

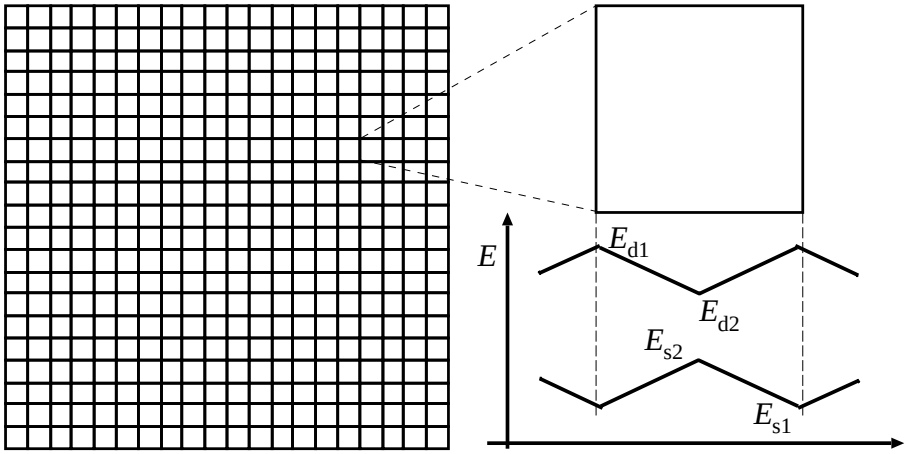

Figure 1: Domain structure of the substrate in the simulation model. Also shown are the variations of E_{s} and E_{d} inside a single domain.

where k is the prefactor ($k = k_{\mathrm{B}}T/h$) [7], E is the activation energy for diffusion, k_{B} is the Boltzmann constant and T is the temperature. The activation energy consists of the adatom-substrate interaction E_{s} and adatom-adatom interaction E_{n}:

$$E = E_{\mathrm{s}} + nE_{\mathrm{n}}, \tag{2}$$

where n is the number of lateral nearest neighbor of the adatom. The evolution of the system was followed using the N-fold method algorithm [6].

In order to investigate the effect of the patterns on the substrate the lattice was divided into square-shaped domains as shown in Figure 1. The patterned surface was incorporated to the nucleation model in two ways (see Figure 1):

1. Letting the parameter E_{s} vary piecewise linearly as a function of the position on the surface.

2. Introducing an additional diffusion barrier E_{d} for adatom jumps towards the nearest domain boundaries. This barrier depends also on position of the adatom inside the domain.

These are designated as models A and B, respectively.

In all the simulations presented in this work the lattice size was chosen to be 352×352 sites and the domain size was selected to be 22×22 sites. Periodic boundary conditions were applied in both lateral dimensions. The model parameters $E_{\mathrm{s}} = 0.75$ eV and $E_{\mathrm{n}} = 0.18$ eV were taken from Ref. [8]. The order of magnitude of the diffusion barrier E_{d} and variation of adatom-substrate interaction E_{s} inside a domain were based on the studies of Brune *et al.* [5], i.e. values (see Figure 1) $E_{\mathrm{d1}} = 0.02$ eV, $E_{\mathrm{d2}} = 0$ eV, $E_{\mathrm{s1}} = 0.65$ eV and $E_{\mathrm{s2}} = 0.85$ eV were used. It should be noted that there is no intention in this work to study quantitatively any particular system but to investigate the qualitative behavior of the models presented. The deposition flux F was chosen to be 0.0033 ML/s. The studied temperature range was determined by the behavior of the system. For $T = 360 \ldots 420$ K the effect of the patterns on nucleation was most pronounced.

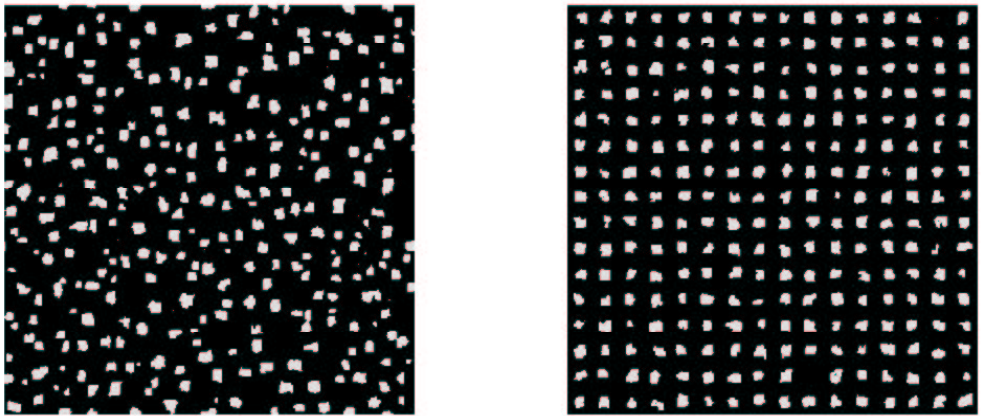

Figure 2: Examples of configurations generated by KMC simulations at the temperature of 390 K and at the adatoms coverage of 15 %. On the left is illustrated the results from simulations where the substrate was homogeneous. On the right is the lattice from simulations using model A for a patterned substrate. Dark areas designate the substrate and light areas the first layer of adatoms.

RESULTS AND DISCUSSION

All the results presented here are for systems simulated up to 15 % coverage of adatoms. At this coverage the islands are purely two dimensional. The results are averages of 50 simulation runs.

In Figure 2 are illustrated examples of surface structures for the homogeneous substrate and for the model A of a patterned substrate. Confinement of islands into the center areas of the domains is clearly seen for the patterned substrate. Some simulations were performed using a modification of the model B where the additional diffusion barrier due to dislocations was in effect only when the adatom was in the immediate vicinity of the dislocation. However, this model produced islands nucleation preferentially on the domain boundaries, not at the centers of the domains as has been observed experimentally for the Ag-Pt systems [5].

Island size distributions for all three cases at various temperatures are shown in Figure 3. The forms of the distributions can easily be understood [4]. At temperatures below 360 K all the distributions have similar forms. At these low temperatures the average diffusion length of adatoms is so short that the effect of inhomogeneity of the surface does not show up. At 360 K an additional maximum around $s = 70$ is observed. This is due to the fact that at these temperatures all the material deposited on a single domain is confined into a single island in this domain. For the coverage of 15 % and the domain of 22×22 lattice sites the size of this island is $s = 73$ particles. When the temperature is further increased the diffusion of adatoms across domain boundaries becomes possible resulting in coalescence of smaller islands into larger ones, thus leaving part of the domains empty.

The confining effect of the patterned surface can clearly be seen in Figure 4 where the average size of the islands $< s >$ and the standard deviation of the size distributions σ_s are plotted for the temperature range 320–440 K. There is a plateau in the curves of average island size around the temperature 390 K for both models A and B. The curves for the

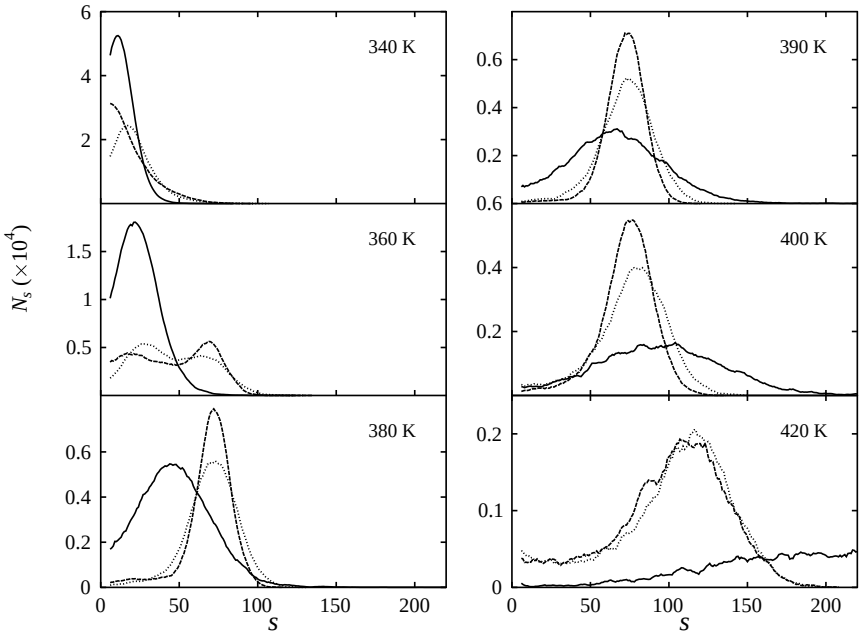

Figure 3: Island size distributions for 15 % coverage at different temperatures for the homogeneous substrate (solid line) and for the models A (dotted line) and B (dashed line). The horizontal axis is the number of particles in the island and the vertical axis is in units islands/lattice site.

standard deviation of the size distributions increase at low temperatures for all three cases. Around 360 K the curves for patterned surfaces decrease due to the confinement of islands inside domains. Although the average size curves for models A and B are similar for the temperature range 340–420 K the standard deviation curves differ for these two models. Moreover, the average island sizes of these models behave differently at low and high temperatures.

The effect of model parameters on results was studied by performing simulations using model A and varying the adatom-adatom interaction parameter E_{n}. The average island sizes and standard deviations of the island size distributions for these simulations are illustrated in Figure 5. A higher value of E_{n} results in a decrease in the confinement because the effect of the interaction between the adatom and substrate becomes smaller. The confinement also happens at higher temperatures when a larger value for parameter E_{n} is used. Also shown in Figure 5 are results of simulations using different strength of the patterns in the surface i.e. different values of adatom-substrate interaction energies E_{s1} and E_{s2}. Using a smaller E_{s1} and a larger E_{s2} makes the patterning of the surface stronger and increases the confinement. However, the curves for $< s >$ and σ_s remain unchanged at temperatures below 380 K. A similar effect of parameters E_{d1} and E_{d2} was found in the case of model B.

The parameters E_{s1}, E_{s2} and E_{n} have a different effect on the results. This suggests that it might be possible to extract these parameters from experimental data.

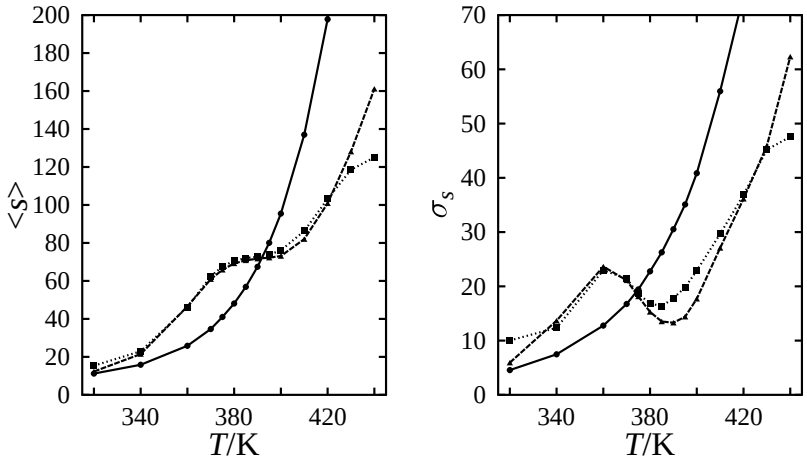

Figure 4: Average island size (on the left) and standard deviation of the island size distribution (on the right) for 15 % coverage at different temperatures for the homogeneous substrate (solid line) and for the models A (dotted line) and B (dashed line).

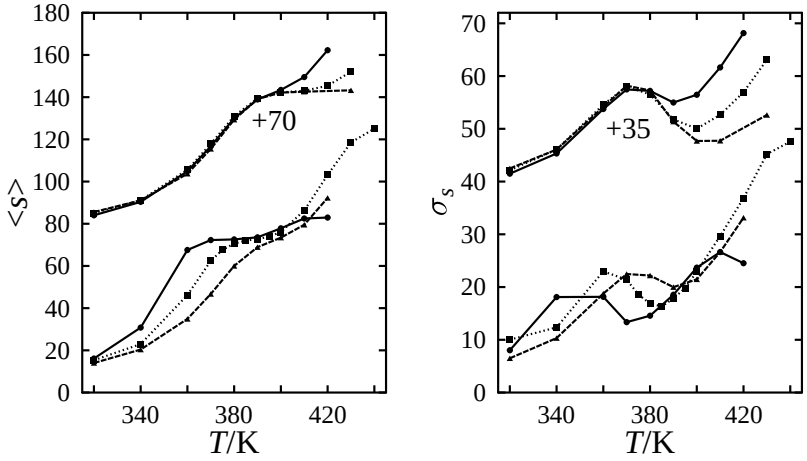

Figure 5: Average island size (on the left) and standard deviation of the island size distribution (on the right) for 15 % coverage at different temperatures for model A. The three lower curves correspond to simulations using parameter values $\{E_{\rm s1}, E_{\rm s2}, E_{\rm n}\} = \{0.65, 0.85, 0.14\}$ eV (solid line), $\{0.65, 0.85, 0.18\}$ eV (dotted line) and $\{0.65, 0.85, 0.22\}$ eV (dashed line). The three upper curves are the results of simulations using parameter values $\{E_{\rm s1}, E_{\rm s2}, E_{\rm n}\} = \{0.65, 0.85, 0.22\}$ eV (solid line), $\{0.60, 0.90, 0.22\}$ eV (dotted line) and $\{0.55, 0.95, 0.22\}$ eV (dashed line). Note that the upper curves are shifted vertically.

CONCLUSIONS

Periodic inhomogeneity in the activation energy of adatom diffusion can have a strong confining effect on adatom nucleation. The two models and parameter set studied in this work indicate that this confinement may happen in a narrow temperature range. The dependence of the results on model parameters suggests that information on their values could be extracted from experimental island size distributions; e.g. for the case of Ag-Pt system [5]. On the other hand, these parameter values could also be calculated by using realistic atomic level calculations.

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