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Strained growth in surfactant-mediated heteroepitaxy

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Abstract

It was shown recently that in the submonolayer regime the shape of islands of exchanged adatoms can be changed substantially by temperature, deposition flux, or coverage in the surfactant-mediated heteroepitaxial growth systems. The shape transition from fractal islands to compact ones can be induced by decreasing temperature or increasing flux. This shape transition is completely contrary to the classic diffusion-limited aggregate (DLA) theory, but can be explained in the frame of a reaction-limited aggregate (RLA) model, in which a stable island consists of the exchanged (or dead) adatoms only, an adatom must overcome a large energy barrier to become the seed of a stable island, or overcome another little smaller barrier to join an existing island. We propose that the strain due to the mismatch in the heteroepitaxy plays the key role in the coverage-induced shape change. Our simulation shows that the strain always makes the islands more compact. With the strain taken into account, there is indeed an island shape change to more compact islands. Applied to the growth of Ge on the Si(1 1 1) substrate pre-covered by a monolayer of Pb, the coverage-induced shape change of Ge islands can be explained.

Keywords: Epitaxy; Growth; Semiconductor; Surfactant; Pattern formation

1. Introduction

The early stage, or submonolayer regime, of epitaxial growth is essential to the whole growth and the resultant film quality as well. For a direct growth of type-B adatoms on a substrate of type-A atoms, it has been established firmly that the diffusion-limited aggregate (DLA) theory [1–9] satisfactorily describes submonolayer growth and pattern formation. The original model of this theory was proposed by Witten and Sander [1]. An impinging adatom hits and then sticks to the existing island without any relaxation. This model produces an ideal fractal island. But for real growth systems there is always some relaxation after the 'hit-and-stick' process. Therefore, fractal islands are formed usually at low temperature or high flux; and compact islands are favoured at high temperature or low flux because of relaxation of adatoms along the island edges [2–9].

But a completely contrary growth phenomenon was observed in a surfactant-mediated heteroepitaxial growth of Ge on Si(1 1 1) substrate pre-covered by a monolayer of Pb atoms as the surfactant [10,11]. Fractal Ge islands are formed at

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high temperature or low flux; compact Ge islands are formed at low temperature or high flux. In addition, they observed a coverage-induced shape change of Ge islands: Ge islands are more compact at higher coverage. These cannot be explained in the framework of the classic standard DLA theory. We proposed a simple model [12,13] to explain the experimental growth phenomenon contrary to the DLA theory. It was noted that the nucleation and growth of adatoms islands in the surfactant-mediated growth are very different from those in the DLA growth systems in that stable islands consist of only *dead adatoms*, i.e., immobile adatoms which have already exchanged positions with the surfactant atoms beneath them. For the position exchange an *active* adatom must overcome an energy barrier which is much larger than the diffusion barrier. Therefore, the nucleation and growth in the surfactant-mediated growth are exchange-limiting processes, rather than the diffusion-limiting process in direct B-on-A growth of the DLA type. The shape transition, induced by temperature and flux [10,11], of stable Ge islands were explained by emphasizing two exchange processes: seed exchange and enhanced exchange, besides the quick diffusion of adatoms on the surfactant terrace [12,13].

In this paper, we shall further explore the effect of the strain due to the mismatch of adatoms with the substrate on the island growth. Especially, the possible island shape evolution with increasing coverage is discussed. We parameterize the strain in terms of existing first-principle calculation results of strained systems. A coverage-induced shape change can, indeed, be achieved in an appropriate parameter regime after the strain is taken into account.

We start with a square lattice of an ideally flat substrate of material A, covered with a complete surfactant layer of material S. Atoms of type B are deposited onto the surfactant layer at a given deposition rate, F. We take into account only three elementary rate processes [12,13]: diffusion of a B-type atom on the surfactant terrace; seed exchange of B-type adatom with the surfactant atom beneath it; and the enhanced exchange of subsequent B atoms to join the existing dead atoms. We denote the activation barriers of these three processes by V_d, V_s, and V_e, respectively, and the corresponding rates by R_d, R_s, and R_e, with $R = v\exp(-V/kT)$. The three barriers satisfy the inequality chain $V_d \ll V_e < V_s$. V_d is the smallest barrier because adatom diffusion is often significantly enhanced due to the passivation of the surface by the surfactant layer. V_s is the largest barrier, making it the rate-limiting process for eventual formation of a stable island. The last inequality reflects the fact that the last process is enhanced by its neighbouring exchanged dead atoms. Usually there are two kinds of strain: on-island strain and terrace strain defined as follows [14]. The on-island strain mainly affects the binding energy between the atoms within the island and between the atoms in the island and other atoms stuck at the island edge. The terrace strain mainly affects the diffusion barrier of a mobile adatom on the terrace. Because here we are interested in the key effect of the strain on the islands and submonolayer pattern formation of the stable islands, we shall take the on-island strain into account only.

Our kinetic Monte Carlo (KMC) simulations are performed on a 200×200 square lattice. For the activated diffusion processes, we use $V_d = 0.57\,\text{eV}$, $V_s = 0.90\,\text{eV}$, and $V_e = 0.78\,\text{eV}$ in order to achieve quite large islands at quite low coverage. The strain of an island of adatoms should generally change the energy barriers for the exchange of adatoms stuck at the island, and for the diffusion of its neighbouring adatoms on the surfactant terrace, as considered in KMC simulations of other growth systems [15]. But in order to capture the key point of the surfactant-mediated growth in a simple model, we take into account the strain effect on the exchange barriers only. This is reasonable because the nucleation and growth in this case is achieved through the exchange processes, being different from the DLA case. The strain energy can, in principle, be calculated as a function of position from elastic continuum theory [16]. Here, we use a simplified model which approximates the strain energy by a power-law dependence upon the number of island atoms. We assume that a stable Ge island of N_{is} dead atoms contributes a strain energy pN_{is}^q at its edge, where p is a constant coefficient (dimension: energy) and q is the power-law exponent, and

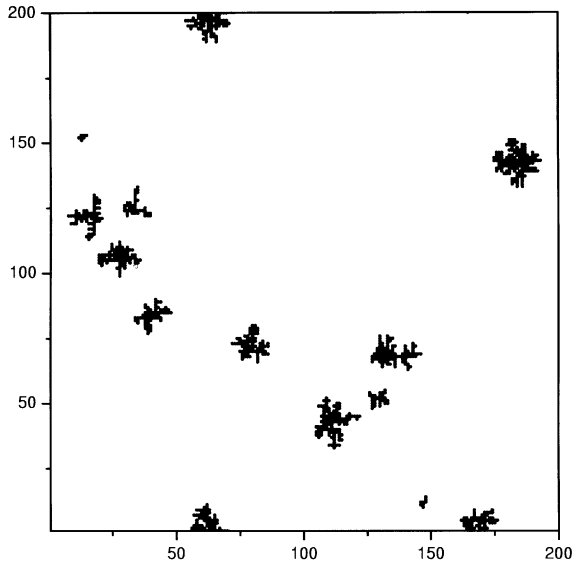

Fig. 1. The KMC result with the parameters: $T = 310$ K, $F = 0.004$ ML/s, $\theta = 0.025$ ML, and $p = 0.0$ eV.

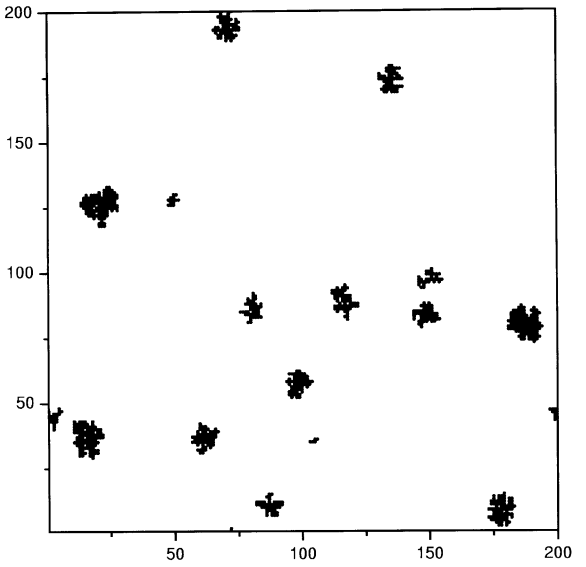

Fig. 2. The KMC result with the parameters: $T = 310$ K, $F = 0.004$ ML/s, $\theta = 0.025$ ML, and $p = 0.012$ eV.

p and q are determined by comparing the simulation results with real experiments. In our simulations we vary the temperature, flux, and coverage in a wide range of parameters in order to find an optimum regime in which we can reproduce the shape transition induced by temperature and flux, and then can achieve more compact islands at higher coverage.

Fig. 1 is a typical KMC result at temperature $T = 310$ K, the flux $F = 0.004$ ML/s, and coverage $\theta = 0.025$ ML. Here no strain is taken into account, $p = 0$. It is clear that all islands are fractal. Fig. 2 shows the effect of the strain on the island shape. All parameters are kept the same as those for Fig. 1 except the strain. Here the strain parameters are set to be $p = 0.012$ eV and $q = 1.67$. The islands are much more compact when compared with Fig. 1. This means that the strain makes the islands more compact. Fig. 3 shows the KMC result at $\theta = 0.07$ ML with other parameters remaining the same as those of Fig. 2. The islands are much more compact than those in Fig. 2. We should compare it with Fig. 2 to clarify the effect of increasing coverage on the island shape. There is indeed a coverage-induced change of the island shape. To clarify the role the strain plays, we perform a further KMC

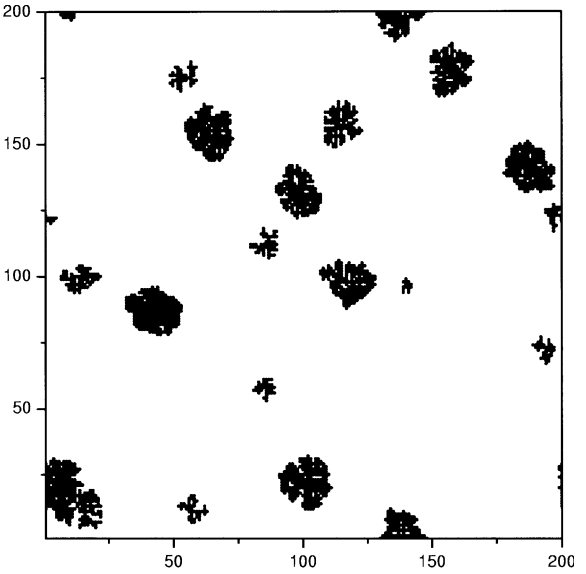

Fig. 3. The KMC result with the parameters: $T = 310$ K, $F = 0.004$ ML/s, $\theta = 0.07$ ML, and $p = 0.012$ eV.

by keeping all parameters unchanged except for the strain. Fig. 4 shows the KMC result when the strain is set to be zero but other parameters are the same as those for Fig. 3. The islands in Fig. 4 are again

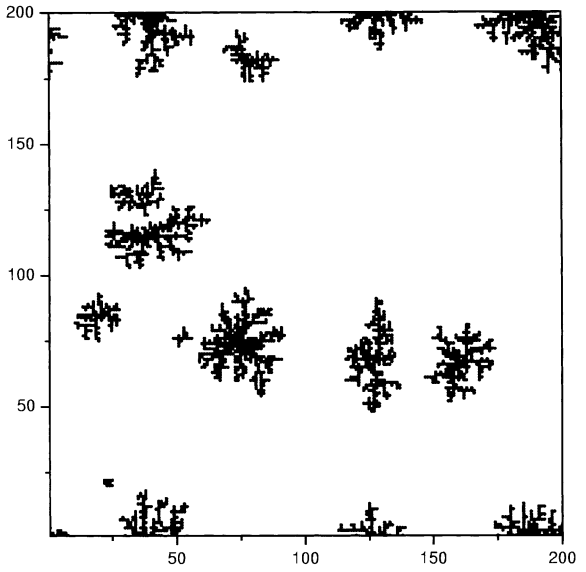

Fig. 4. The KMC result with the parameters: $T = 310$ K, $F = 0.004$ ML/s, $\theta = 0.07$ ML, and $p = 0.0$ eV.

ideally fractal. Therefore, it can be concluded that the strain plays the essential role in the coverage-induced change of the island shape in the surfactant-mediated heteroepitaxial growth. There would not be any coverage-induced change of island shape without the strain.

With the barrier set $(V_d, V_s, V_e) = (0.57, 0.90, 0.78)$ eV and the strain, we not only reproduce the previous shape transition induced by temperature and flux [12,13], but also obtain the island shape change induced by coverage. The strain is essential for the coverage-induced shape change. We have taken into account the effect of the on-island strain only. The terrace strain also tends to enhance the coverage-induced change of the island shape. But it is less important, and will be taken into account in further quantitative work [14]. Here, we have calculated the on-island strain in a simple way, which is accurate enough to obtain the qualitative key result. We also extend our KMC simulations to wider regimes in the parameter space of (V_d, V_s, V_e) and the strain. The above key results persist in a quite large parameter regime. Applied to the experimental observations by Hwang et al. [10,11], our model including the strain can naturally explain the coverage-induced change of the Ge island shape besides the shape transition induced by temperature and flux.

2. Conclusions

In summary, we have explored the possibility that island shape is induced by coverage. We propose that the strain due to the mismatch in the heteroepitaxy plays the key role in the coverage-induced shape change. Our simulation shows that the strain always makes the islands more compact. With the strain taken into account, there is indeed an island shape change to more compact islands at higher coverage. Applied to the growth of Ge on the Si(1 1 1) substrate pre-covered by a monolayer of Pb, the coverage-induced shape change of Ge islands can be explained naturally.

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