Monte Carlo simulation of roughening at step-terraced surfaces

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Abstract. GaAs thermal smoothing at temperatures \( T \leq 650^\circ C \) under conditions close to equilibrium yields surfaces with atomically smooth terraces separated by steps of monatomic height. At higher temperatures \( T \geq 700^\circ C \), surface smoothing changes to roughening. In the present paper, thermal roughening of a step-terraced surface caused by atomic step flow around step pinning centers is studied using Monte-Carlo simulation. It is proved that the Schwöbel barrier is necessary for step bunching in the presence of step pinning centers. The lower limit of the Schwöbel barrier \( E_S = 0.4 \) eV is estimated for the GaAs(001) surface.

1. Introduction

Fundamental surface science [1-4], fabrication of nanostructures [5,6] and device applications [7] require high-quality, atomically flat crystal surfaces. Silicon surfaces with atomically smooth terraces separated by steps of monatomic height can be obtained by annealing in vacuum [8,9]. The application of vacuum annealing to GaAs and other III-V semiconductors is hindered due to the preferential evaporation of a more volatile V component. To avoid surface depletion with a more volatile component and possible kinetic instabilities, thermal smoothing should be made under conditions close to equilibrium, when neither sublimation nor growth occurs [10]. Ding et al. [11] proved that annealing MBE-grown GaAs(001) films at a sufficiently high arsenic vapor pressure and a moderate temperature yielded flat surfaces. Step-terraced surfaces were also obtained by annealing GaAs(001) substrates in MOCVD setups in a mixture of arsine and hydrogen [12]. An efficient and cost-effective technique for GaAs surface smoothing by annealing under conditions close to equilibrium between the surface and vapors of Ga and As was developed in [13,14]. This technique yields step-terraced GaAs(001) surfaces with atomically smooth terraces.

Increasing the annealing temperature speeds up surface mass transport and, thus, facilitates the smoothing process. However, at high temperatures surface smoothing changes to roughening [11]. Surface roughening, which consists of the formation of multilayer islands and pits, step bunches and destruction of step-terraced morphology, was also observed in [13] at high annealing temperatures \( T \geq 700^\circ C \). This roughening restricts the annealing temperature and, thus, the maximal initial surface roughness that can be smoothed at reasonable annealing times. Possible reasons for surface roughening were discussed in [15,16]. Since the disordered morphology differs qualitatively at a fixed temperature, depending on the annealing details, the experimental results [13] cannot be explained by the thermodynamic roughening transition. In [15,16], the formation of multilayer islands and pits under sublimation and growth is explained by atomic step flow around some surface spots at which the...
sublimation and growth are inhibited. In its turn, the instability of step bunching caused by step pinning at these spots should lead to the destruction of the step-terraced morphology. The goal of this paper is to clarify the “kinetic” surface roughening using Monte Carlo simulation and, in particular, to elucidate the influence of the Schwöbel barrier on surface roughening.

2. Method
Monte Carlo simulations of surface roughening were performed in the one-component model of the Kossel crystal. The model was extended to maintain equilibrium or a specified deviation from equilibrium between atomic fluxes from and to the surface. The model parameters used for the surface roughening simulation were obtained earlier from the description of the experimental GaAs smoothing kinetics by Monte Carlo simulation [17,18]. These parameters include the diffusion activation energy $E_d = 1.3 \pm 0.05 \text{ eV}$, the lateral bond energy $E_b = 0.32 \pm 0.02 \text{ eV}$ and the adatom desorption energy $E_{\text{des}} = 1.9 \pm 0.05 \text{ eV}$. The roughening simulations were performed for the terrace width of an order of magnitude smaller than in the experiment due to limited computational resources. The procedure of rescaling the value of $E_d$, which takes into account this difference, is described in [14].

3. Results and discussion
As discussed in [14,15], the formation of multilayer islands and pits can be explained by monatomic step flow around some surface spots at which the sublimation and growth are inhibited. To confirm this explanation, we performed Monte Carlo simulations of the interaction between moving atomic steps and a spot. Figure 1 shows the simulated step-flow sublimation (top row) and growth (bottom row) of a step-terraced surface with one spot in the center, at which sublimation and growth are artificially forbidden. Monatomic steps retract from the bottom towards the top under sublimation (figure 1(a)) and advance from the top towards the bottom under growth (figure 1(e)). After approaching the spots, the steps flow around the spots forming the inlets (figure 1(b,f)). With further step retraction (or advance), the inlet length $L$ increases (figure 1(c,g)) and, eventually, the steps detach from the spots (figure 1(d,h)). Each time a monatomic step passes through a spot, the spot height increases by one monolayer (ML) with respect to the decreased terrace level under sublimation, or decreases by 1 ML with respect to the increased terrace level under growth. Thus, the multilayer islands and pits are formed under sublimation and growth, respectively. The similarity between the growth and sublimation is seen from figure 1, except for the longer inlet in the case of growth.

![Figure 1](image-url)

**Figure 1.** Simulated step flow through a surface spot (white/black dot) during sublimation (a–d) and growth (e–h). Sublimation and growth on the spot are forbidden. Steps retract from the bottom towards the top (a–d) and advance from the top towards the bottom (e–h). $L$ denotes the inlet length before the step detaches from the spot.
To clarify the difference between sublimation-induced and growth-induced roughening, we simulated kinetic roughening at various sublimation and growth rates. Figure 2 shows the inlet length before the step detachment (i.e., the maximum inlet length) averaged over 10 step detachments. It is seen that increasing the sublimation and growth rates from $V = 10^{-4}$ ML/s to $V = 10^{-2}$ ML/s leads to decreased inlet length by a factor of ~4. This is produced by narrowing the inlets down to the spot lateral size while increasing the sublimation and growth rates. Narrow inlets readily break, so the step detaches earlier at a higher sublimation or growth rate. It is also seen in figure 2 that the growth yields inlets that are ~2 times longer as compared to the sublimation at the same rate. The qualitative explanation of the inlet length difference is as follows. Adatom mass transport plays the main role in the inlet formation because the vacancy concentration is 3-5 times lower. In particular, adatom mass transport along the step makes the inlet broader and, thus, prevents it from breaking. During the sublimation, the maximal inlet length is lower due to a lower adatom concentration, as compared with the growth.

![Figure 2](image)

**Figure 2.** Simulated maximum inlet length dependence on growth and sublimation rates. The lines are drawn to guide the eye.

To reproduce the rough morphology, which was observed experimentally in [14,15], we simulated the interaction between moving atomic steps and randomly arranged surface spots. The simulation, however, revealed that the formation of multilayer step bunches at surface spots is inhibited by atom exchange between the steps. When two steps approach each other, atoms detach from the upper step, travel across the terrace between the steps, and incorporate into the lower step. This results in effective step repulsion, which prevents step bunching. To reduce the mass transport across the steps, the Schwöbel barrier was introduced between the step and the upper terrace. To estimate the Schwöbel barrier value needed for step bunching at surface spots, we simulated roughening of a step-terraced surface with a relatively low terrace width (~5 nm). Figure 3 shows the simulated sublimation (top row) and growth (bottom row) of a step-terraced surface with the same surface spot positions. The Schwöbel barrier increases from left to right. No step bunches are seen on the surfaces with no barrier, although the step shape is distorted near the surface spots (figure 3(a,e)). The Schwöbel barrier $E_S = 0.2$ eV leads to the formation of 3–5 ML high step bunches, which are seen as a local deviation of the surface height from the mean plane (the brighter and darker areas in figure 3(b,f), correspondingly). The increase in the barrier up to $E_S = 0.4$ eV results in an increase in the height of step bunches up to 5–10 ML (figure 3(c,g)). It is seen that most of the steps stopped at surface spots and, thus, joined step bunches. A further increase in the barrier to $E_S = 0.6$ eV yields only subtle morphology changes (figure 3(d,h)). It should be noted that for step-terraced surfaces without surface spots, the Schwöbel barrier results in step bunching under sublimation and prevents step bunching under growth [10]. In contrast, in the presence of surface spots, the Schwöbel barrier yields step
bunching during both sublimation and growth. The reason is that here the step bunching is caused by step pinning, not by the Schwöbel barrier. The barrier reduces the step repulsion as described above. The lower limit of the Schwöbel barrier on the GaAs(001) surface is approximately $E_s = 0.4$ eV.

Figure 3. Simulated images of step-terraced surfaces with the same surface spot positions after sublimation (a–d) and growth (e–h) for different Schwöbel barrier values: a,e) no barrier; b,f) $E_s = 0.2$ eV; c,g) $E_s = 0.4$ eV; d,h) $E_s = 0.6$ eV.

4. Conclusions

The destruction of the GaAs(001) step-terraced surface morphology, which was observed experimentally at high temperatures due to deviations of annealing conditions towards growth or sublimation [15,16], was reproduced here using Monte Carlo simulations. For relatively broad terraces, the formation of multilayer islands and pits is caused by step flowing around some surface spots, at which the sublimation and growth are inhibited. The step bunching, which was observed for relatively narrow terraces, is caused by step pinning at these spots. The bunching is assisted by the Schwöbel barrier, which reduces the step repulsion.

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