Effects of Steering in Metal Epitaxial Growth

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ABSTRACT

Recent experiments on metal epitaxial growth indicate that at a high-angle of incidence the long-range attraction of the surface to incoming atoms can lead to steering effects near step-edges which significantly modify the growth and contribute to a growth instability. Here we present molecular dynamics simulation results obtained using embedded atom potentials for Cu/Cu(100) growth carried out in order to measure the strength of the steering effect due to short-range interactions. Our results indicate that even at normal incidence, the short-range attraction can lead to a significant uphill current which can significantly increase the selected mound angle and surface roughness. The results of kinetic Monte Carlo simulations of Cu/Cu(100) growth with steering effects included are also presented and compared with recent experiments.

Keywords: Epitaxy, molecular dynamics, metals, steering.

1 INTRODUCTION

Recently, there has been significant progress in understanding the fundamental mechanisms which control the surface morphology and structure in epitaxial growth [1]. These include an improved understanding of nucleation in submonolayer growth and its role in controlling the morphology in multilayer growth [2], of the role of the Ehrlich-Schwoebel barrier in mound-formation and unstable growth [3,4], and of the effects of surface currents and downward funneling in determining a selected mound angle. For example, the origin of the mound instability in homoepitaxial growth is now understood to be the existence of diffusion bias due, for example, to a barrier to interlayer diffusion at steps.

The general role of surface currents in controlling the symmetry, mound coarsening behavior and surface morphology has also been studied. In particular, it has recently been shown that both edge-diffusion and the short-range attraction of diffusing adatoms to ascending steps can lead to an uphill current and contribute to an instability [5,6]. In addition, one of us has recently shown [7] that the existence of corner diffusion can play a fundamental role in affecting the mound morphology and coarsening behavior in unstable growth. All of these studies highlight the importance of taking detailed atomic mechanisms into account in order to understand and predict the behavior on the nanoscale and above.

We note that in a number of kinetic Monte Carlo (KMC) simulations the standard assumption of downward funneling (DF) [8], in which atoms approaching the surface funnel downward to the nearest nearby epitaxial growth site, has been used. For example in the case of downward funneling on an fcc(100) surface, atoms are assumed to "cascade" downward to nearest-neighbor sites until a four-fold hollow site is found. This is assumed to be a reasonable approximation in the case of deposition at normal incidence except at very low temperatures when terrace diffusion is inactive and there is a high density of microprotrusions which may lead to restricted downward funneling [9]-[11].

However, recent work by van Dijken et al [12]-[14] indicates that at a high angle of incidence the long-range van der Waals attraction of the surface to incoming atoms can lead to 'steering' and shadowing effects which dramatically alter the surface morphology as well as the assumption of downward funneling. These effects can lead to a strong enhancement of the mound instability as well as facetting and ripple structures. Furthermore, we expect that even in the case of normal incidence, steering effects due to the short-range attraction of incoming atoms to nearby steps can lead to significant deviations from the DF assumption. In the presence of an Ehrlich-Schwoebel barrier, these effects can lead to a significant uphill current and thus play an important role in controlling the selected mound angle and surface morphology. For example, recent analytical calculations [5] have shown that, in the presence of a non-negligible Ehrlich-Schwoebel barrier, the surface current J(m) (where m is the local slope) and selected mound angle depend strongly on the 'bias' for atoms landing near a step.

Steering effects may also play a significant role at very low-temperature. For example, recent molecular dynamics simulations using embedded atom potentials by Montalenti and Voter [15] for normal-incidence growth of just a few monolayers of Ag/Ag(100) at zero K, indicate that steering effects due to short-range attraction can play a significant role. Therefore it is important to determine the effects of steering due to short-range attraction in epitaxial growth.

2 MOLECULAR DYNAMICS SIMULATIONS

In order to determine the effects of short-range steering at normal incidence we have carried out molecular dynamics simulations of adatom deposition at a single step on the Cu(100) surface using embedded-atom potentials for

copper. Simulations were carried out over a range of incident kinetic energies K_i (0.04 eV < K_i < 0.2 eV) corresponding to typical thermal deposition energies in order to study the dependence of the steering effect on the incident energy [16].

Figure 1 shows a typical trajectory (top and side views) of an atom approaching a (110) step on the Cu(100) surface with incident kinetic energy (K_i = 0.1 eV) from a position which is significantly beyond the step edge. As can be seen, due to steering effects the adatom is attracted to the top terrace rather than the lower terrace as would be expected in the case of downward funneling.

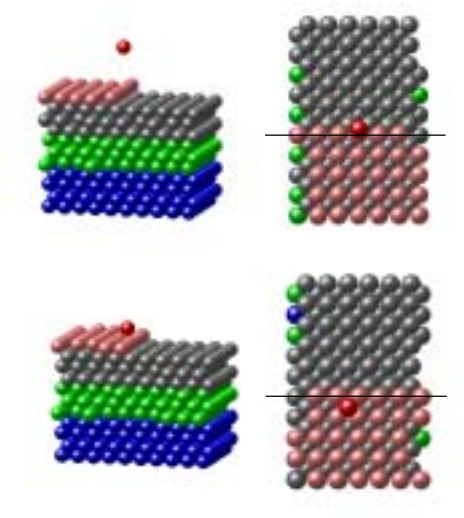

Figure 1: Sequence of snapshots (side and top views) of molecular dynamics simulations of deposition near a monatomic step showing effects of steering.

In order to quantify this effect we have measured the probability of 'upward funneling' at an edge by averaging over many possible deposition sites within a 'window' of width equal to the nearest-neighbor distance surrounding the step-edge. Our results indicate that over a range of typical values of the incident atom kinetic energy used in epitaxial growth, the 'upward funneling' probability (P_{up}) at a single-layer step is significantly higher than the value (P_{up} = 1/2) expected in the absence of attraction. As already noted, analytical calculations [6] demonstrate that in the presence of a non-negligible Ehrlich-Schwoebel barrier, this implies a significant uphill current. Thus we expect the effects of steering due to short-range interactions to play a significant role in epitaxial growth at normal incidence.

3 KINETIC MONTE CARLO SIMULATIONS

In order to demonstrate the effects of short-range attraction on the surface morphology in multilayer epitaxial growth, we have carried out kinetic Monte Carlo (KMC) simulations for Cu/Cu(100) growth at 160 K. Our KMC simulations were carried out using energy barriers which were calculated using effective medium theory (EMT). One of the main features of the EMT calculations is that the rates of monomer and dimer diffusion are essentially the same [17] while the barriers for edge-diffusion are significantly lower. A simple parametrisation of the energy barrier for an adatom to hop on a flat (100) surface based on the occupation of its nearest and next-nearest neighbors was found to give a good representation of the derived configuration-dependent EMT barriers. By comparing with experimental results for the submonolayer island density at 213 K [18] the appropriate prefactors for monomer and dimer diffusion were determined. A further comparison with experimental results by Zuo et al [19] for the island density as a function of temperature also gave good quantitative agreement. Recent results obtained by Furman et al [20] using a similar but slightly more elaborate parametrisation to describe submonolayer Cu/Cu(001) growth yield similar rates for adatom and dimer diffusion over the same range of temperatures.

Using these parameters, we have carried out kinetic Monte Carlo simulations of multilayer growth in order to compare with experimental results of Ernst et al [21] for the surface roughness as a function of coverage for Cu/Cu(100) growth at 160 K. Using the usual downward funneling (DF) the kinetic Monte Carlo results for the surface width at 160 K are significantly below the experimental results.

In order to see if inclusion of steering effects due to short-range attraction can lead to a noticeable difference in the simulations, we have also carried out kinetic Monte Carlo simulations using a modified downward funneling in which atoms which land near the edge of a surface step are attracted preferentially to the upper terrace. Our results indicate that inclusion of such a bias leads to significantly better agreement with experiment, thus confirming that steering effects have a significant impact on the surface morphology and roughness even in the case of normal incidence.

4 MULTISCALE SIMULATIONS

While the KMC simulation results described above provide a good approximation of the effects of steering due to short-range attraction on epitaxial growth, a much more accurate description may be obtained by carrying out multiscale simulations in which molecular dynamics is used during the deposition process while kinetic Monte Carlo (KMC) is used to describe the diffusion of adatoms once they have landed on the surface. We are currently carrying out such simulations using a hybrid MD/KMC code in order to compare with our KMC results.

5 CONCLUSIONS

Our molecular dynamics and KMC simulations demonstrate that steering effects due to short-range attraction can play an important role in determining the surface morphology in multilayer metal epitaxial growth. We are currently studying the effects of both long-range attraction due to van der Waals interactions and short-range attraction, as well as shadowing on the surface morphology at off-normal incidence.

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