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Effects of deposition angle in low-temperature metal (100) epitaxial growth

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The effects of oblique incidence on the surface roughness in low-temperature Cu/Cu(100) epitaxial growth are investigated via kinetic Monte Carlo simulations, which include the effects of shadowing as well as short-range and long-range attraction. While the effects of deposition angle are found to be relatively weak at 200 K, at a slightly lower temperature (160 K) both the surface roughness and the growth exponent β depend strongly on deposition angle. These results resolve a long-standing puzzle regarding the growth behavior of Cu/Cu(100) over this temperature range. Our results also demonstrate that, in general, the effects of deposition angle must be considered in low-temperature growth even for moderate deposition angles.

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Recently, there has been a great deal of progress in understanding the morphological evolution in epitaxial thin film growth (for a recent review, see Ref. 1), and a variety of effects and processes have been shown to play an important role. In addition to growth temperature and flux, these include the effects of crystal geometry,[2,3] as well as the Ehrlich-Schwoebel barrier to interlayer diffusion,[4] edge and corner diffusion,[5,6] and the attraction of depositing atoms to the substrate.[7,8] More recently, it has been found that, in the case of glancing incidence, the deposition angle can also play an important role.[9–14] While the effects of deposition angle have traditionally been assumed to be negligible for moderate deposition angles, they have not yet been fully investigated, especially at low temperatures. Understanding these effects is important, since they can have a strong effect on a variety of important properties including the surface morphology.

One case of particular interest is that of Cu/Cu(100) growth at low and intermediate temperatures, which was first studied by Ernst *et al.*[15] and, more recently, by Botez *et al.*[16] In both of these experiments, the growth of the surface roughness w corresponding to the rms height fluctuation was studied as a function of film thickness t along with the corresponding growth exponent β ($w \sim t^{\beta}$). In particular, as the growth temperature was increased from 160 to 200 K, Ernst *et al.* observed a significant increase in the surface roughness. The corresponding growth exponent β was also observed to increase dramatically from a value of approximately 0.26 at 160 K to approximately 0.56 at 200 K. In contrast, Botez *et al.* found very little difference between their results for the surface roughness at $T=200$ K ($\beta=0.54$) and $T=160$ K ($\beta=0.52$). Thus, while the results of Botez *et al.* and Ernst *et al.* are in agreement at 200 K, at $T=160$ K they disagree not only quantitatively but also qualitatively. This discrepancy between the experimental results for Cu/Cu(100) at 160 K has so far not been explained.[1] This puzzle is made more intriguing by the existence of good agreement between the experimental results of Ernst *et al.* and Botez *et al.* at $T=200$ K.

Here we demonstrate that, by taking into account the effects of deposition angle on the surface roughness, this puzzle can be resolved. In particular, we find that good agreement between our simulations and both experiments can be obtained if we assume that the experiments of Ernst *et al.*[15] correspond to normal or close-to-normal deposition, while the experiments of Botez *et al.*[16] correspond to off-normal deposition. Our results also demonstrate that, in general, the effects of deposition angle must be considered in low-temperature growth even for moderate deposition angles.

In order to simulate the effects of shadowing and attraction during deposition, as well as surface relaxation processes after deposition, we have carried out simulations using a hybrid model which combines molecular dynamics (MD) simulations to describe the deposition process, with kinetic Monte Carlo (KMC) simulations to describe surface relaxation. The activation barriers in our KMC model are based on a parametrization of the effective medium theory (EMT) barriers for Cu(100) calculated by Jacobsen.[17] We note that these barriers were originally used, along with the inclusion of "uphill funneling" due to short-range (SR) attraction of depositing atoms, to obtain good quantitative agreement with the observed roughening behavior in Cu/Cu(100) growth obtained by Ernst *et al.* at 160 K.[8] More recently, by slightly enhancing the rate of corner diffusion, it was shown[6] that this model can also explain the large roughening exponent ($\beta \simeq 0.5$) obtained by the same group at 200 K. While the detailed barriers have previously been described in detail elsewhere,[6,17] here we note that its main features are the existence of very fast edge diffusion, as well as an effective rate for dimer diffusion on a terrace which is the same as that for monomer diffusion. We also note that this model leads to parameters which are very similar to those recently obtained by Furman *et al.*[18] using an embedded atom method (EAM) potential, and which lead to excellent agreement with submonolayer experiments over the temperature range $T=180-300$ K.

As in several previous simulations of steering effects in Cu/Cu(100) growth,[8,12,13] a Lennard-Jones (LJ) copper potential[19] of the form $V_{\mathrm{LJ}}(r)=4\epsilon[(\sigma/r)^{12}-(\sigma/r)^{6}]$ (where $\epsilon=0.4093$ eV and $\sigma=2.3377$ Å) was used to take into account the SR interaction in our MD simulations of the deposition process. We note that, in simulations of adatom deposition near Cu(100) close-packed steps,[8] it was found that the results obtained using this potential (with cutoff distance 2σ) were essentially identical to those obtained using a more

sophisticated EAM Cu potential.[20] However, in order to include the effects of long-range (LR) interactions, we have also included a van der Waals attraction[9,21] for atoms which are farther than the cutoff distance for the short-range interaction.

For convenience, the LR interaction may be divided into two parts. The first part corresponds to the LR attraction between the depositing atom and the semi-infinite slab below the last completely filled layer of the substrate, and has the form $U_{\mathrm{LR}}(z)=-C_3/z^3$ (where z is the height of the depositing atom above this layer). The main effect of this interaction is to bend the path of the depositing atom as it approaches the substrate. The second part corresponds to pair interactions with the atoms above the last completely filled layer and has the form $V_{\mathrm{LR}}(r)=-C_6/r^6$, where r is the distance between the depositing atom and the substrate atom. At high angles, this interaction can lead to flux focusing toward protruding structures which can significantly enhance the surface roughness.[9] We note that the values of the constants C_3 and C_6 describing the long-range interaction are related [$C_3=2.1$ eV Å^3 and $C_6=(3a^3C_3/2\pi)\simeq 47.2$ eV Å^6, where a is the lattice constant of Cu], and were obtained in a previous calculation.[21] We also note that these values are significantly weaker than predicted by the $1/r^6$ "tail" of the LJ Cu potential. Therefore, to avoid a discontinuity we have used the following expression for the pair-potential in our simulations:

$$V_T(r)=[1-f(r)]V_{\mathrm{LJ}}(r)+f(r)V_{\mathrm{LR}}(r), \qquad (1)$$

where $f(r)=1/(1+e^{-(r-2\sigma)/R})$ is the sigmoid function centered on $r=2\sigma$ with width $R=0.125\sigma$. For comparison, we have also carried out calculations using the less accurate LJ Cu potential $V_{\mathrm{LJ}}(r)$ [corresponding to $f(r)=0$ in Eq. (1)] in which the LR interaction with atoms below the last completely filled layer was represented by the corresponding $1/z^3$ potential [$U_{\mathrm{LJ}}(z)=8\pi\epsilon\sigma^6/3a^3z^3$]. As expected, this leads to an enhancement of the effects reported here, i.e., to results that are equivalent to those obtained with the more accurate interaction for a deposition angle which is 2°–3° smaller.

In order to accurately simulate the effects of shadowing and steering during deposition, multiscale simulations[22] (in which both the depositing atom and the surrounding substrate atoms undergo molecular dynamics during each deposition) are required. Since a full MD simulation of each deposition event is prohibitive for the system sizes ($L=512$) and film thicknesses considered here, in most of our simulations a simplified method was used in which the substrate atoms were all held fixed at their lattice positions while a one-atom MD simulation of the trajectory of the depositing atom was carried out. In this case, the depositing atom was assumed to follow the trajectory determined by its interaction with the substrate until its distance to the closest substrate atom is equal to the nearest-neighbor distance. As in previous work,[2,3] it then "cascades" randomly via downward funneling[23,24] (DF) from a site corresponding to this atom, until it reaches a fourfold hollow site. This approach takes into account the bulk of the steering effects due to LR and SR attraction. However, for comparison we have also carried out fully multiscale simulations in which a full MD simulation was carried out for each deposition event for smaller system sizes for a few specific cases. In these simulations, velocity rescaling of atoms below the surface was used to maintain constant temperature. We note that, in a few percent of deposition events, the deposited atom does not find a fourfold hollow site on molecular dynamics time scales, and in this case DF was assumed. We believe that this is a reasonable approximation since the substrate temperature is not too low (for comparison, see for example, Ref. 25), while the relevant barriers are typically quite small, i.e., less than 0.2 eV.[24]

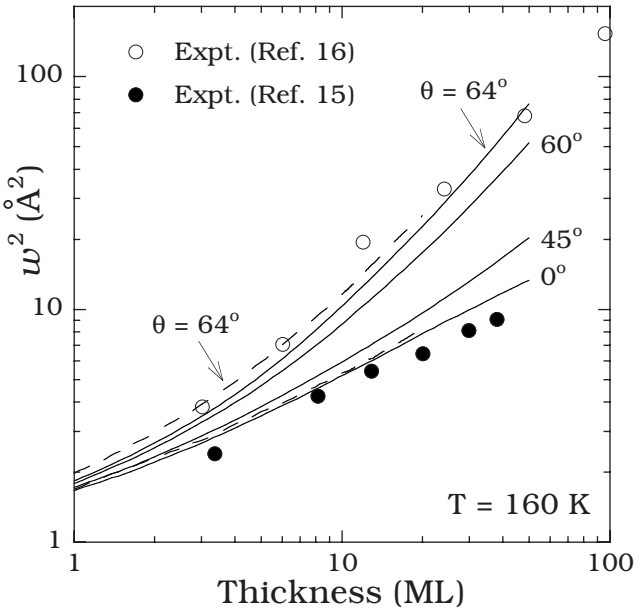

FIG. 1. Comparison of experimental results of Ernst *et al.* (Ref. 15) (filled circles) and Botez *et al.* (Ref. 16) (open circles) for Cu/Cu(100) growth at 160 K, along with full MD (dashed lines) and one-atom MD (solid lines) results for different angles of deposition. Thickness is indicated in monolayers (ML).

Since the results were found not to depend on the cutoff for larger values, a long-range cutoff $r_{\mathrm{cut}}=7\sigma$ was used for the pair interaction. As in previous work,[8] in our MD simulations the initial kinetic energy of the deposited atom corresponded to the average value $\bar{K}_i=2k_BT_m\simeq 0.20$ eV (where $T_m=1356$ K is the melting temperature of copper), while the starting point was randomly chosen a distance r_{cut} above the maximum film height. In the simulation results shown here the azimuthal angle was chosen such that the deposition direction was parallel to the close-packed step edge, i.e., along the [110] direction. However, we have also carried out simulations with other azimuthal angles, and while the surface morphology was found to depend on the azimuthal angle, only a negligible effect on the surface roughness was observed.

Figure 1 shows the experimental results of Ernst *et al.* (filled circles) and Botez *et al.* (open circles) for the surface roughness obtained in Cu/Cu(100) growth at 160 K. Also shown are our one-atom MD simulation results (solid lines) as well as our fully multiscale results (dashed lines) for dif-

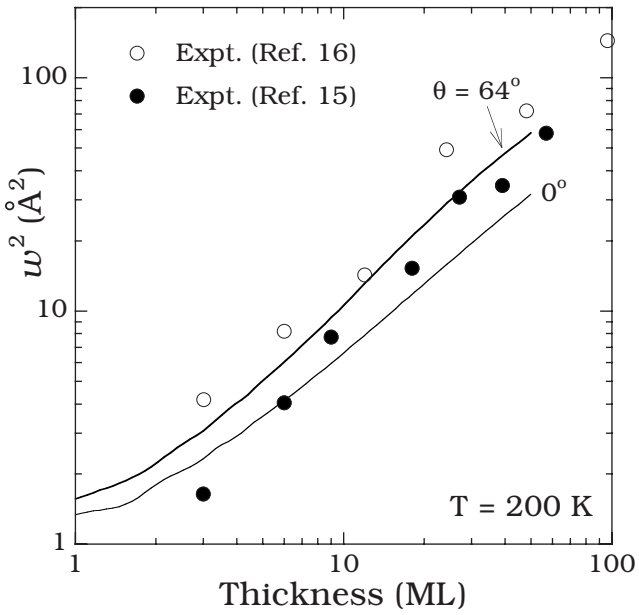

FIG. 2. Comparison of experimental results of Ernst *et al.* (Ref. 15) (filled circles) and Botez *et al.* (Ref. 16) (open circles) for Cu/Cu(100) growth at 200 K with one-atom MD simulation results (solid lines) corresponding to normal and off-normal deposition.

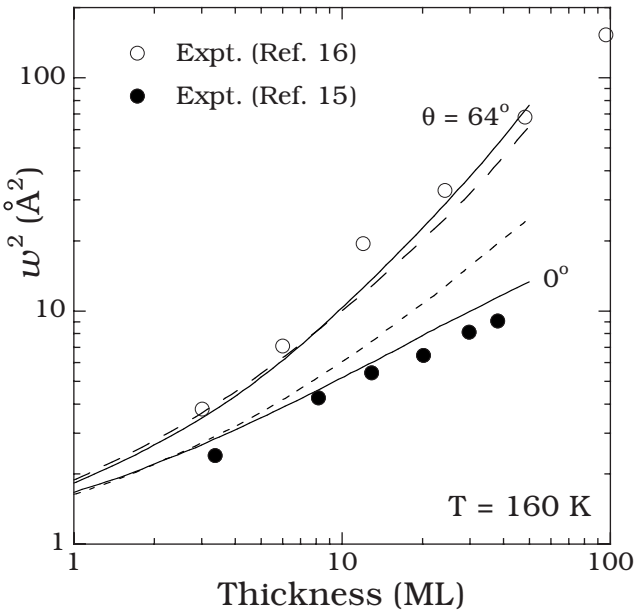

FIG. 3. Comparison of one-atom MD results at 160 K at normal and off-normal incidence (solid lines), with off-normal incidence simulation results (θ=64°) obtained without LR attraction (long dashes), as well as in the absence of both SR and LR attraction (short dashes). Experimental results from Refs. 15 and 16 are included for reference.

ferent deposition angles θ with respect to the substrate normal. As can be seen, at normal incidence our simulation results are in relatively good agreement with the experimental results obtained by Ernst *et al.* in Ref. 15, and are essentially the same as obtained in previous simulations of normal incidence growth at this temperature.[6,8] However, for deposition angles larger than 45°, both the growth exponent β and the surface roughness increase dramatically. In particular, for deposition angles close to 60° our simulation results are in good quantitative agreement with the experimental results of Botez *et al.* As discussed in more detail below, these results suggest that in these experiments the deposition angles were not negligible. They also indicate that, at low temperatures, the deposition angle can have a strong effect on the growth behavior even for moderate values, i.e., away from glancing incidence. We also note that our one-atom MD simulations slightly underestimate the effects of the SR interaction after collision with the substrate. In particular, the roughness obtained in our fully multiscale simulations is somewhat larger than in the case of one-atom MD, and the difference appears to increase with deposition angle.

Figure 2 shows the corresponding experimental results of Ernst *et al.*[15] and Botez *et al.*[16] at 200 K, along with our simulation results for the same range of deposition angles as in Fig. 1. As can be seen, at this temperature the differences between the experimental results are relatively small, although the roughness is slightly higher in the experiments of Botez *et al.* Similarly, our simulation results exhibit a relatively weak dependence on the deposition angle, although the roughness is somewhat higher for the case of off-normal deposition than at normal incidence. Thus, again there is reasonable agreement between our simulation results and the experimental results.

These results indicate that at the lower temperature (T =160 K) there is a strong dependence of the surface roughness on the deposition angle, while at higher temperature (T=200 K), the dependence is relatively weak. The significantly weaker dependence on deposition angle at 200 K may be explained by the fact that at this temperature, both the mound slope and step density are significantly smaller than at 160 K. This is consistent with the observation by Ernst *et al.* of (115) facets at 200 K and (113) facets at 160 K as well as with previous simulation results at normal incidence.[6] The decreased mound slope at 200 K reduces the effects of shadowing, while the lower step density minimizes the effects of steering due to SR attraction during deposition.

Figure 3 shows a comparison of our results for the surface roughness at 160 K (θ=64°) with the SR and LR interaction included, with the corresponding results obtained with SR but no LR attraction (long dashes), as well as in the absence of both SR and LR attraction, corresponding to ballistic deposition (short dashes). As can be seen, the LR attraction has a relatively weak effect on the surface roughness at this angle. This is consistent with the experimental results of van Dijken *et al.* at higher temperature,[9] and can be explained by the fact that, since the LR attraction is relatively weak, only for large glancing angles is the normal component of the depositing particle's velocity small enough to allow sufficient time for it to have an effect. In contrast, Fig. 3 indicates that for moderate deposition angles both shadowing and SR attraction play important roles in enhancing the surface roughness at low temperature.

We now compare the deposition angles used in the experiments with those in our simulations. In Ref. 15 the deposi-

tion geometry was described and indicates normal incidence, as assumed in previous simulations[6] as well as here. After the results presented here were obtained, an estimate of the deposition angle in the experiment of Ref. 16 was also carried out,[26] and is in good quantitative agreement with the values used here. Thus, the available evidence strongly supports our conclusion that the results of both experiments can be explained once the deposition angle is taken into account.

In conclusion, by taking into account the deposition angle along with the effects of shadowing and SR attraction, we have obtained good qualitative and semiquantitative agreement with the experimental results of Ernst *et al.*[15] and Botez *et al.*[16] at both 160 and 200 K. Our simulation results resolve the long-standing puzzle of a discrepancy between these experimental results at 160 K. Our results also demonstrate that the deposition angle can have a significant effect on the surface morphology at low temperatures, even for moderate deposition angles. In this connection, we note that low-temperature growth has recently been studied in a variety of other systems.[27–31] Our results indicate that, in general, the deposition angle should be considered along with the growth temperature in analyzing and/or interpreting these and similar experiments.

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