Steering effects on growth instability during step-flow growth of Cu on Cu(1,1,17)

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A kinetic Monte Carlo simulation in conjunction with a molecular dynamics simulation is utilized to study the effect of the steered deposition on the growth of Cu on Cu(1,1,17). It is found that the deposition flux becomes inhomogeneous in the step train direction and that the inhomogeneity depends on the deposition angle when the deposition is made along that direction. The steering effect is found to always increase the growth instability with respect to the case of homogeneous deposition. Further, the growth instability depends on the deposition angle and direction, showing a minimum at a certain deposition angle off-normal to the (001) terrace, and shows a strong correlation with the inhomogeneous deposition flux. The increase of the growth instability is ascribed to the strengthened step Ehrlich-Schwoebel barrier effects, which are caused by the enhanced deposition flux near the descending step edge due to the steering effect.

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I. INTRODUCTION

In the growth of thin films on a vicinal surface of high areal step density, there is a net current of deposit particles toward the ascending step edge due to the Ehrlich-Schwoebel barrier at the descending step edge. Such transport of deposit atoms to the ascending step increases the possibility of step-flow growth and stabilizes the growth on vicinal surfaces against step bunching.[1,2] Moreover, such asymmetric flow of deposit atoms provides the possibility of forming a structure along the step edge and has been a subject of numerous studies of the growth of one-dimensional systems.[3] Even in the thin-film growth on a vicinal surface, however, the meandering instability develops along step edges.[4–7] Possible sources for this instability[2] have been suggested to be the asymmetric adatom diffusion due to the step Ehrlich-Schwoebel barrier[4,7] and kinetically limited diffusion of the adatoms due to the kink Ehrlich-Schwoebel barrier.[8,9]

In addition to the kinetic effects mentioned above, the deposition process, one of the most recently recognized dynamic processes, has been found to affect the thin-film growth.[10–12] That is, the interaction between a deposit atom and the atomic structure on the surface modifies the trajectory of the deposit atom, called the steering effect, and causes the inhomogeneous distribution of adatoms affecting the growth of thin films. Adjacent to the edge of the islands or steps, the steering effect is conspicuous due to the rapid variation of the interaction potential. Thus, the steering effect should be more influential on the deposition in the vicinal surface having a high areal step density than on a singular surface.

The purpose of the present study is to explore the role of steered deposition on the thin-film growth on a vicinal surface, which has been ignored in most of the previous simulation or theoretical studies (see Ref. 2 for a review). Specifically, we study the correlation between the deposition flux distribution and growth instability on the vicinal surface, varying the deposition angle and direction. We also search for any possibility to overcome such kinetic growth instability by adjusting the dynamic variables involved in the deposition process. We have chosen to study the growth of Cu on Cu(1,1,17), because Cu(1,1,17) shows no surface reconstruction and has been a subject of many experimental[6,7] and theoretical studies,[9,13] allowing us to compare our results with pre-existing ones. The present study utilizes a computer simulation combining a molecular dynamics (MD) simulation for the dynamics of deposit atoms with a kinetic Monte Carlo (KMC) simulation for the growth of adatoms on the surface.

We find that the steering-induced enhancement in the deposition flux near a descending step edge is a critical factor affecting the growth instability on a vicinal surface. The inhomogeneity of the deposition flux depends on the deposition angle, and a deposition angle that gives the minimum growth instability is found. Nevertheless, the steering effect always increases the growth instability regardless of the deposition angle, with respect to the case in which the steering effect is absent.

II. SIMULATION METHOD

KMC simulation is adopted to simulate the whole process of thin-film growth. In conventional KMC simulation where the steering effect is neglected, the deposition event is carried out by placing an atom on an arbitrary site available on the surface. To simulate the deposition process as in the experiment, we incorporate MD into KMC simulation: Whenever a deposition event is selected in the otherwise conventional KMC,[12] MD is employed to simulate the trajectories of the depositing atoms in detail.

In the MD simulation, a Lennard-Jones potential $U(r)=4D[(\sigma/r)^{12}-(\sigma/r)^{6}]$ is used for the pair interaction between a deposit atom and an atom on the surface, with $D=0.4093$ eV and $\sigma=2.338$ Å. These values of D and σ are adopted from Dijken *et al.*[10] and Sanders *et al.*[14]

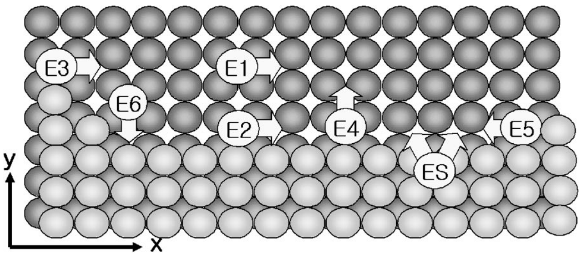

FIG. 1. Illustration of some diffusion processes taken into account in the present simulation.

The initial kinetic energy of the deposit atom is set to 0.15 eV, corresponding to the melting temperature of Cu. Newton's equation of motion is solved using the Verlet algorithm. The atom approaches the substrate by MD, and is then positioned to the nearest fourfold hollow site from the terminal position. The transient mobility is not included in the present study. That is, the deposit atoms are assumed to be in equilibrium with the substrate right after the deposition.

In the KMC simulation, a lattice gas model is adopted, which allows jump diffusion for adatom motion on the fcc lattice. The possibility of each jump diffusion is calculated from the corresponding hopping rate $\nu = \nu_0 \exp^{-\beta E}$, with attempt frequency $\nu_0 = 3.6 \times 10^{12}/\text{s}$. The definitions of the most relevant diffusion processes in the present simulation are illustrated in Fig. 1. Listed in Table I are the values of the diffusion barriers (E_i) that are adopted from the values used by Rusanen[9] and Merikoski[13] in a growth simulation of Cu on Cu(1,1,17) and those obtained by Furman[15] from a simulation study for thin-film growth on Cu(001).

We take the diffusion barrier controlling diffusion along the step edge (E_2) to be 0.38 eV, which is larger than those found in the literature (about 0.25–0.26 eV).[16] On the vicinal surface where the step density is very high, the most frequent and thus the most time-consuming process is the diffusion along the step edges. If E_2 is taken to be below 0.3 eV, the simulation time becomes untolerably long. Hence, we perform the growth simulation with $E_2 = 0.38$ eV to secure the statistical reliability of the simulation by repeating simulations enough to have converging results. For the justification of the use of such large E_2, we examine how the kinetic variables affect the steering effect on growth instability by performing simulations while varying some of the most influential diffusion barriers, such as E_2, E_3, and E_5, and find that the kinetic effects do not obliterate the steering effect. The large values of E_2 adopted in the present simulation seem only to reduce the kinetic relaxation of the steering effect.[17]

TABLE I. Diffusion barriers and parameters used in KMC. The same notation is used for each diffusion process as in Fig. 1.

Diffusion type	Diffusion barrier (eV)
E1	0.42
E2	0.38
E3	0.51
E4	0.68
E5	0.59
E6	0.18
ES	E1+0.1
Jump frequency (ν_0)	3.6×10^{12}
Deposition rate (F_0)	0.003 ML/s

Cu(1,1,17) surface has a (001) terrace of 8.5 atomic width between two steps of an atomic height in [−1,1,0] direction. In the following, the x axis is along the step edge, as shown in Fig. 1, and the y axis is along the step-train direction. The simulation box has 12 terraces with a step-edge length of 800 a_0, where a_0 is the surface lattice constant of Cu(001): 2.55 Å. Periodic boundary conditions are adopted in both x and y directions.

III. RESULTS AND DISCUSSION

As a preliminary investigation of the steering effect on thin-film growth, the deposition flux distributions or deposition probabilities are examined for various deposition angles by MD. The deposition angle is measured from the normal to the (0,0,1) terrace to [−110] direction (y axis). The positive deposition angle is for the deposition direction from the upper terrace to the lower one along the y axis, as shown in Fig. 2(b), and the negative angle is for the opposite direction, as shown in Fig. 2(a). The trajectories of the deposit atoms in Figs. 2(a) and 2(b) show the steering effect, where bending of trajectories of the incident atoms, most notably near steps, occurs due to the interaction between the deposit atom and substrate atoms. Figures 2(c)–2(g) show the deposition flux distributions normalized to homogeneous flux for various deposition angles. The deposition flux, shown with solid circles in Figs. 2(c)–2(g), increases near the step, while that on terrace decreases compared with the homogeneous flux.[18] It is important, however, to note that this enhanced flux near the step is not due solely to the steering effect. The deposition flux distribution in Fig. 2(h) is for deposition with no steering effect considered, and still shows relatively high flux near the steps. This is because there are only two adsorption sites available in 2.5 a_0 distance from each step edge along the y axis, while one adsorption site is available in each 1.0 a_0 distance on the terrace. The deposition flux after subtracting this purely geometrical contribution is shown with open circles in Figs. 2(c)–2(g), and shows the enhanced deposition flux near the steps to be due purely to the steering effect.

For deposition angles closer to the grazing angle (that is, angles of larger magnitude), the deposition flux becomes more inhomogeneous or more enhanced near the steps, as can be seen by comparing Figs. 2(c) and 2(d) with Figs. 2(e) and 2(f), respectively. As the deposition angle becomes larger, so does the flight time of depositing atoms, during which their trajectories and in turn, the deposition fluxes are apt to be more disturbed by the inhomogeneous substrate potential. It is also interesting to note the

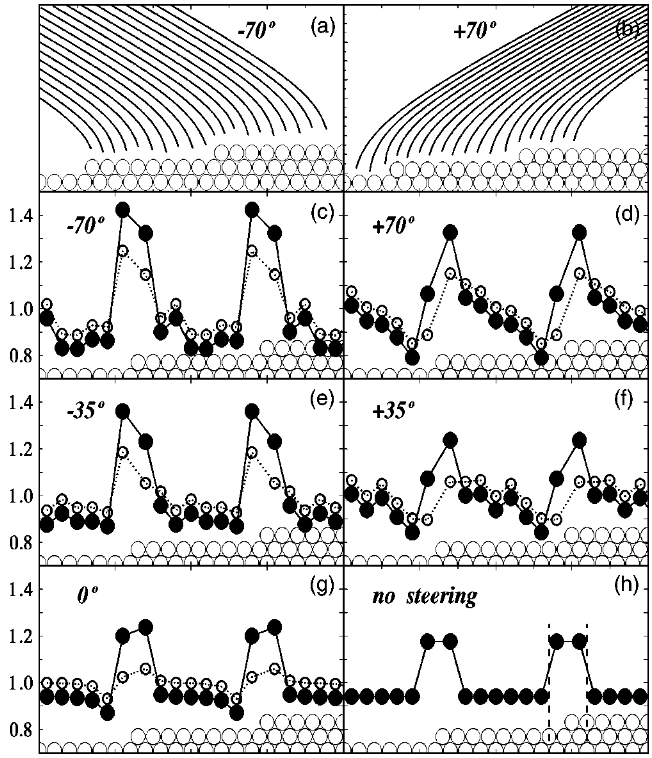

FIG. 2. Trajectory of deposit atoms and normalized deposition flux. (a) Trajectory of deposit atoms at deposition angles of (a) −70° and (b) +70°. Steered deposition fluxes at deposition angles of (c) −70°, (d) +70°, (e) −35°, (f) +35°, (g) 0°, and (h) deposition without the steering effect. Normalization is made with respect to homogeneous flux. Solid circle: Deposition flux. Open circle: Deposition flux after subtracting the enhancement due to the purely geometrical contribution.

difference between the flux profiles at positive deposition angles [Figs. 2(d) and 2(f)] and those at negative deposition angles [Figs. 2(c) and 2(e)]. In the negative angles, the deposition flux at the ascending step edge is larger than that at the descending step edge, and vice versa. This may be explained by the fact that at positive deposition angles, the shadowing effect[12] diminishes the probability for deposit atoms to sit on the sites next to the ascending step edge, while no such shadowing is expected for negative deposition angles.

The effect of the steering-induced inhomogeneous deposition flux on thin-film growth on a vicinal surface is studied by KMC utilizing MD for each deposition event. During the growth, the substrate temperature is set to 204 K. Snapshots of a simulated system are shown in Figs. 3(a) and 3(b). Figure 3(c) shows the evolution of a step as the coverage increases. We observe that the average position of the step edge proceeds 8.5 a_0 for each monolayer (ML) deposition, indicating step-flow growth. However, the lateral roughness increases, and the coherence between adjacent step edges develops to form finger-like structures [Fig. 3(c)] as the coverage increases. Each "finger" shows a ledge envelope along [100] and [0,−1,0] directions, as observed for both experimental studies[7] and the simulation results by Rusanen *et al.*[9]

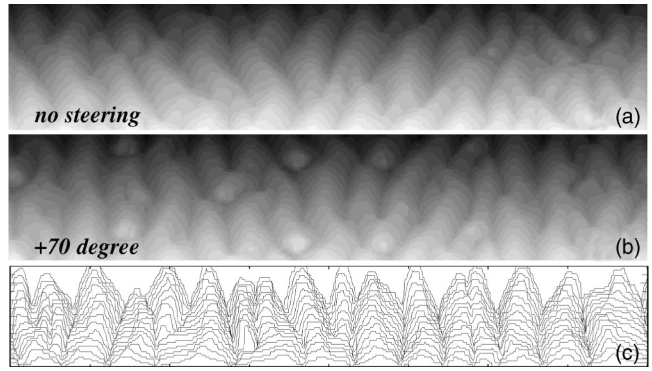

FIG. 3. Snapshot images of the 5 ML Cu grown on Cu(1,1,17) at 240 K. (a) Deposition with no steering effect. (b) Steered deposition at 70°. The size of (a) and (b) is $(800\times 150)\ a_0^2$. (c) Evolution of a step edge with increasing coverage. Successive curves show the development of step edges at Cu coverages below 5 ML with an increment of 1/3 ML. The size of (c) is $(800\times 54)\ a_0^2$. a_0 is the surface lattice constant of Cu(001): 2.55 Å.

For a quantitative understanding of the growth instability on a vicinal surface, the lateral roughness and the finger width taken as a measure of lateral coarseness are calculated. We define the lateral height $h(x)$ as the distance from a position x at a pristine step edge to the growth front in the direction normal to the step edge (that is, in the y direction), and the lateral roughness as $w(x)\equiv\sqrt{\langle h(x)^2-\langle h(x)\rangle^2\rangle}$. The lateral coarseness is calculated from the average separation between fingers within heights $h_{avg}\pm 5a_0$, where h_{avg} is the average lateral height of each step. As a measure for the growth instability, we take the aspect ratio of the lateral roughness to finger width (lateral coarseness). For an ideal step-flow growth or a stable growth, the aspect ratio should be very small.

In Fig. 4(a), the lateral roughness increases monotonically as coverage increases. At the maximum coverage of the present simulation (5 ML), the roughness is about 7 a_0, indicating a very rough step edge. The roughness shows a distinct dependence on the deposition angle. In the inset of Fig. 4(a), shown is the lateral roughness as a function of deposition angle after depositing 5 ML. The roughness is minimum at deposition angles at 0°. As the deposition angle becomes larger, so does the roughness. In addition to the deposition angle, the roughness depends also on the direction of deposition. When the deposition is made facing an ascending step edge or at a negative angle, the roughness of the film is small compared with that grown at the same magnitude of deposition angle, but in the opposite direction facing a descending step edge.

The development of lateral coarseness with increasing coverage was estimated by the finger width. In Fig. 4(b) and its inset, the finger width monotonically decreases as coverage increases, and also shows a definite dependence on both deposition angle and direction. The simulated finger width after 5 ML of deposition is around 55 a_0, which is in good agreement with the experimental one, 50 a_0.[5] The finger width shows a maximum at −35° and decreases to a minimum at +70°. Most notable is that the lateral roughness and coarseness have a close correlation in their dependence on

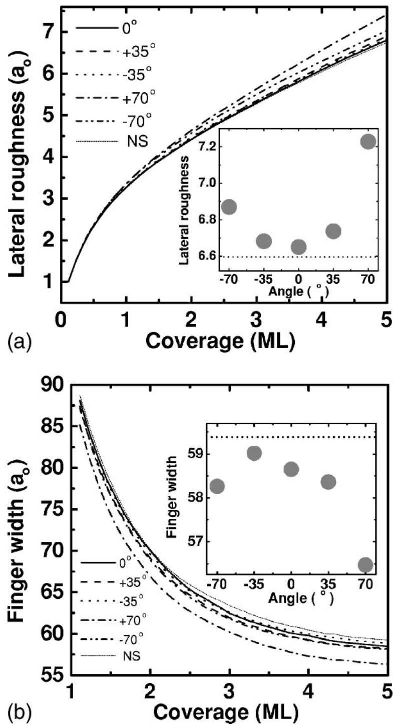

FIG. 4. (a) Lateral roughness and (b) finger width (lateral coarseness) as function of coverage in the growth of Cu on Cu(1,1,17) at 240 K. Refer main text for the definitions of lateral roughness and finger width. Insets: (a) lateral roughness and (b) finger width as a function of deposition angle after depositing 5 ML. The dotted lines in the figures (NS) and insets are the results of growth without considering the steering effect.

the deposition angle; deposition at angles between −35 and 0° shows the most stable step-flow growth with the minimum roughness and maximum finger width or the minimum aspect ratio, while deposition at +70° shows the opposite behavior—the most unstable growth with the maximum roughness and the minimum finger width, or the maximum aspect ratio.

The aforementioned angular dependence of the growth instability should have originated from the dynamic effect of the deposition process, the steering effect, since all the kinetic variables are identical for each deposition at various angles. A direct result of the steering effect is the inhomogeneous deposition flux. Hence, we investigate the correlation between deposition flux distribution and growth stability: The atoms deposited near the ascending step edge are expected to reproduce the step edge by directly adhering to the sites near the step edge, and should not be the main source of steering-induced growth instability. However, the atoms near the descending step edge would diffuse across the terrace before reaching the ascending step edge due to the step Erlich-Schwoebel barrier. During such terrace diffusion, the atoms redistribute themselves to feed and form laterally inhomogeneous structures, thus being a source of meandering instability.[4] Indeed, we find the predicted correlation between the growth instability and the enhanced deposition flux near descending step edge; in Fig. 5, the deposition flux averaged over the three sites adjacent to the descending step edge is

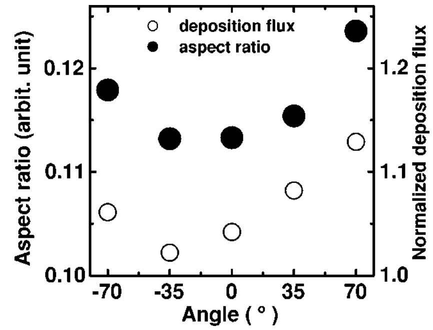

FIG. 5. The aspect ratio of lateral roughness to finger width (solid circle) and the normalized deposition flux averaged over three adsorption sites next to the descending step edge (open circle) are plotted as a function of deposition angle. Normalization is made with respect to the homogeneous flux.

well matched with the aspect ratio for varying deposition angles. As the average deposition flux near the descending step edge is more enhanced, the mean travel length of deposit atoms to the ascending step edge should become longer, and the growth becomes more unstable, giving a larger aspect ratio.

For a possible origin of growth instability on a vicinal surface, two pictures have been proposed based on kinetics of adatoms; one attributes the instability to the step Erlich-Schwoebel barrier effect (SESE)[4,7] and the other to the kink Erlich-Schwoebel barrier effect (KESE).[8,9] SESE affects the motion and redistribution of deposited atoms on terrace, which should be directly dependent on the deposition flux distribution. KESE, however, governs the motion of atoms along step edges, and is not directly affected by the initial deposition flux. The intimate correlation between deposition flux near the descending step edge and growth instability shown in Fig. 5 indicates that the steering-induced deposition flux enhancement near the descending step edge strengthens the role of SESE on growth instability.

In Figs. 4(a) and 4(b), the steered growth always shows larger roughness and smaller coarseness, regardless of the deposition angle, than does the growth neglecting the steering effect (dotted curves). That is, the steering effect always increases the growth instability. Such behavior is expected from the relatively small flux enhancement near the descending step edge for steering-free deposition as shown in Fig. 2, consistent with the aforementioned explanation. Although the steering effect is inevitable for vapor deposition for thin-film growth, the existence of a deposition angle producing the minimum growth instability (Fig. 5) suggests that the optimizaton of the deposition angle should be a prerequisite for the most stable growth of thin films on a vicinal surface.

IV. SUMMARY AND CONCLUSION

KMC simulation in conjunction with MD simulation is performed to study the steering effect, in which the trajectory of each deposit atom is affected by interactions with the substrate, in relation to the growth of Cu on Cu(1,1,17). It is found that the steered deposition flux becomes inhomogeneous and that the inhomogeneity depends on the deposition angle and direction. The deposition flux enhancement near descending step edges is found to be the most critical factor for the increase of growth instability due to the steering effect. The mechanism of such a steering-induced increase of growth instability is discussed in detail. In the present simulation, we also find a deposition angle that produces minimum growth instability and show that the optimization of the deposition angle should be desirable for the most stable thin-film growth on a vicinal surface.

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[18] The flux distribution is determined by the shape of potential formed by substrate atoms near the surface, and is strongly dependent on the terrace width, especially for vicinal surfaces with a narrow terrace, like the present one.