PHYSICAL REVIEW B, VOLUME 63, 121403(R)

Ripple formation and rotation in the growth of Ag/Ag(110): A microscopic view

F. Hontinfinde[1] and R. Ferrando[2,*]
[1]*The Abdus Salam ICTP, P.O Box 586, 34100, Trieste, Italy*
[2]*INFM and CFSBT/CNR, Dipartimento di Fisica dell'Università di Genova, via Dodecaneso 33, 16146 Genova, Italy*
(Received 3 January 2001; published 5 March 2001)

The recently observed formation of cross-channel ripples in the growth of Ag/Ag(110) at low temperatures and their rotation at higher temperatures are studied by kinetic Monte Carlo simulations. The key microscopic mechanisms of both ripple formation and rotation are singled out.

DOI: 10.1103/PhysRevB.63.121403 PACS number(s): 68.35.Fx, 66.30.Fq, 81.15.Hi

Upon the deposition of several atomic layers, even initially flat crystal surfaces can begin to develop spontaneous kinetic patterning, with the formation of regular arrays of three-dimensional mounds. Mound formation can take place by different mechanisms, like Ehrlich-Schwoebel (ES) barriers, adatom-step attraction, fast atomic motion along step edges, and others.[1] On symmetric surfaces, these mounds take a pyramidlike shape, which follows the substrate symmetry [square for Cu/Cu(001) and Fe/Fe(001), symmetry for Pt/Pt(111) (Ref. 2)]. Very recently, also multilayer growth on the initially flat anisotropic Ag(110) surface has been experimentally investigated.[3] The Ag(110) surface is of rectangular symmetry, with channels along the $[1\bar{1}0]$ direction (see Fig. 1); the channels are separated by close-packed atomic rows. On this surface, diffusion of single adatoms is easy along the in-channel (IC) direction, while the cross-channel (CC) motion (along the [001] direction) takes place by exchange and is more difficult.[4,5] Surprisingly, instead of the formation of anisotropic mounds[6] only, a much richer phenomenology has been observed.[3] In fact, at low temperatures T (around 130 K), a ripplelike instability is produced, with ripples oriented along the CC direction. If T is raised to about 160 K, an intermediate regime, with mounds elongated in the two directions, is found, and finally, around 200 K, ripples appear again, but rotated by 90°, so that they become oriented along the IC direction.

In this Rapid Communication we study the microscopic mechanisms of ripple formation and rotation in the growth of Ag(110) by realistic kinetic Monte Carlo (KMC) simulations. The mechanisms of the mounding instability have been investigated in several papers (see, for example Ref. 1); on the contrary, much less is known about the mechanisms leading to the organization of mounds into ripples and to their rotation with increasing T. To our knowledge, what follows is the first microscopic study of the latter issues. In the following, we show that our model reproduces quantitatively the experimental observations of Ref. 3, giving a complete microscopic explanation, which involves one more key ingredient besides the rotation of surface currents[3] and which is closely related to island shapes in the submonolayer regime.[8,3,9] In fact, here we demonstrate that, even though the mounding instability is due to ES barriers hindering interlayer mobility, the organization of mounds into CC ripples at low T (around 130 K) is due to the stability of the weak CC bonds for adatoms in nearby channels and to the anisotropy of diffusion of isolated adatoms. Then we show that CC ripples disappear at intermediate T (around 150 K) because there the weak CC bonds are easily broken and that, up to this T, neither CC diffusion nor interlayer mobility play an important role, so that the surface current still flows in the in-channel direction. This is a crucial point because it demonstrates that ripple formation and rotation during growth are qualitatively different from mounding: the latter can be explained in terms of surface currents caused by adatom diffusion, while the former are deeply connected also to the properties of the bonds in the different directions. Finally, we confirm that, at high T (around 200 K), the formation of IC ripples requires CC both intralayer diffusion and IC interlayer mobility.[3] However, these ingredients are not sufficient: also CC bond breaking is crucial. Strong CC bonds would inhibit IC ripple formation. Our results demonstrate that the anisotropy of bonds is extremely important for the morphology of surface instabilities in multilayer deposition, besides determining the submonolayer island shapes.[10,8,11,12]

The most important elementary diffusion processes in our model are summarized in Fig. 1. We consider both intralayer and interlayer processes. The most important intralayer process is the IC diffusion of isolated adatoms (process *a* in Fig. 1), with a barrier of 0.28 eV (as calculated in Refs. 5 and 7 by many-body tight-binding potentials); on the other hand, CC diffusion (*b*) costs more [0.38 eV (Refs. 5 and 7)]. The other intralayer processes are modeled by the *anisotropic bond-breaking model* (ABBM),[8,12] where the breaking of the

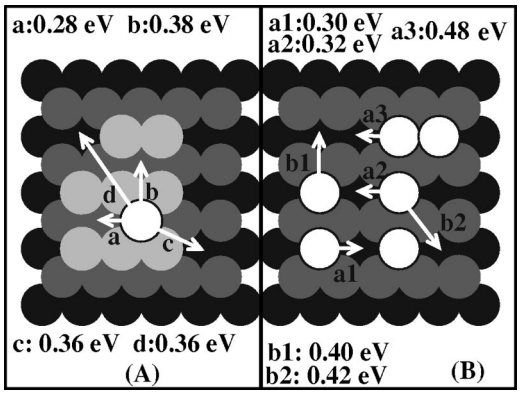

FIG. 1. Diffusion processes with their barriers. (A) Diffusion processes for isolated adatoms, both intralayer and interlayer. (B) Intralayer diffusion processes for atoms with neighbors; in the simulation we let also interlayer processes for atoms with neighbors (not shown in the figure).

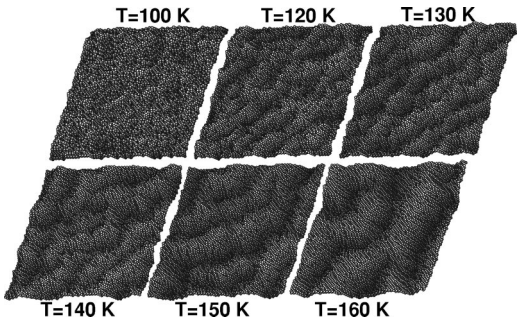

FIG. 2. Snapshots at different temperatures after deposition of 30 monolayers. The in-channel (IC) and cross-channel (CC) directions are along the short and long diagonals, respectively. At 130 K, CC ripples are evident; these are transformed into anisotropic mounds at 150–160 K.

strong IC bonds costs 0.18 eV, while the breaking of the weak CC bonds costs 0.02 eV, so that, for example, process $a3$ (which is IC diffusion with the breaking of one IC and one CC bond) has a barrier of $0.28+0.18+0.02=0.48$ eV. We stress that IC bonds are practically never broken at the temperatures we shall consider in the following. The ABBM reproduces quantitatively the intralayer diffusion barriers as calculated by many-body potentials; a complete discussion of intralayer mobility is given in Ref. 12. Concerning interlayer processes, IC interlayer mobility is relatively easy (process c) with an additional ES barrier of 0.08 eV, whereas CC interlayer mobility is possible only at kinks [process d (Ref. 7)], being extremely difficult along straight steps.[7] We take in the following a common prefactor of 10^{12} s^{-1}. Adatoms are deposited randomly on the surface. If atoms land on growth sites (fourfold hollows), they stick; otherwise, they cascade[13] and search for the closest growth site with lower height.

Let us first consider the low and intermediate T ranges ($100<T<160$ K). Our results are well summarized by the snapshots in Fig. 2, taken after the deposition of 30 monolayers at a flux of 1 monolayer (ML) per minute,[3] the same as in experiments. In Fig. 2, IC and CC directions are along the short and long diagonals, respectively. The size is of about 30 nm and 40 nm along the two directions; this is not far from the size of experimental terraces[3] and it can accommodate several ripple wavelengths at low T. At 100 K no evidence of mounding is found: none of the processes of Fig. 1 is active. The situation changes at $120<T<140$ K. There, well-defined ripples with CC orientation are formed, in agreement with the experiments. This finding is confirmed by the analysis of the height-height correlation function $G(x,y,t)$ at fixed time t (see Fig. 3; x and y are the IC and CC directions, respectively). G is defined by $G(x,y,t)=g(x,y,t)/g(0,0,t)$, where g is given by

$$g(x,y,t)=\langle[h(x,y,t)-\overline{h(t)}][h(0,0,t)-\overline{h(t)}]\rangle, \qquad (1)$$

$h(x,y,t)$ is the surface height at (x,y), and $\overline{h(t)}$ is the average surface height. In the following, t is measured in ML. At 100 K and $t=30$ ML, both $G(x,0,t)$ and $G(0,y,t)$ show a monotonic decay. On the contrary, at $120<T<140$ K,

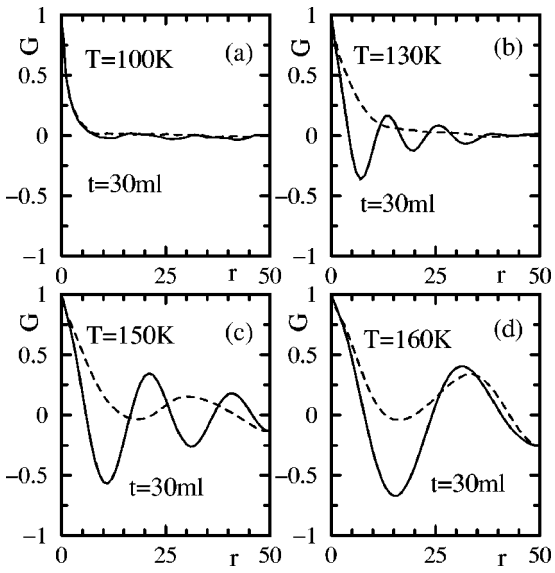

FIG. 3. Height-height correlation $G(x,0,t)$ (along the IC direction, solid lines) and $G(0,y,t)$ (CC direction, dashed lines) in different conditions for the full model. $r=x$ or y is measured in the number of atomic rows or columns. Oscillations in $G(x,0,t)$ together with a monotonic decay of $G(0,y,t)$ indicate the existence of CC ripples.

$G(x,0,t)$ presents a damped oscillation, indicating the onset of a periodicity along the IC direction, which is perpendicular to the ripple orientation, whereas in the other direction the monotonic decay is again found. From the distance between subsequent maxima, we find a ripple wavelength λ_x of 4.0 nm at 130 K, quite close to the experimental value of 4.6 nm. If T is raised further to 150 K, a periodicity arises also in $G(0,y,t)$ and anisotropic mounds are formed instead of CC ripples. λ_x increases with T and is of 6 nm at 150 K, 9 nm at 160 K, and 14.4 nm at 170 K, again in good quantitative agreement with the experiments [see the open circles in Fig. 3(b) of Ref. 3]. Also for the ripple slope $\alpha(x,t)$ we obtain very good agreement with experiments. We estimate α as follows. First we calculate the ripple lateral size $\xi(x,t)$ from $G(\xi(x,t)/2,0,t)=1/2$ (Ref. 14); then α is given by $\alpha(x,t)=\tan^{-1}[2w/\xi(x,t)]$, where w is the interface width. We find, for $t=30$ ML, $\alpha=20.6°$ at 120 K, 19.9° at 130 K, 17.1° at 140 K, 15.4° at 150 K, and 13° at 160 K.

What are the microscopic mechanisms of CC ripple formation at low T? How does their transformation into mounds at intermediate T take place? In our KMC simulations we can answer these questions by selectively either suppressing or enhancing some microscopic processes, and then looking at the effects on the growth morphology.

First, we analyze ripple formation around 130 K, where interlayer mobility is not active due to the ES barriers; this should cause a mounding instability.[1] But something more is needed to organize mounds into CC ripples. If we eliminate CC bonds, so that both $a1$ and $a2$ have the same barrier as a (see Fig. 1), mounds are already formed at low T instead of CC ripples [see G in Fig. 4(a); both $G(x,0,t)$ and $G(0,y,t)$ are already developing a damped oscillation at 130 K, as at 150 K when CC bonds are taken into account]. Therefore,

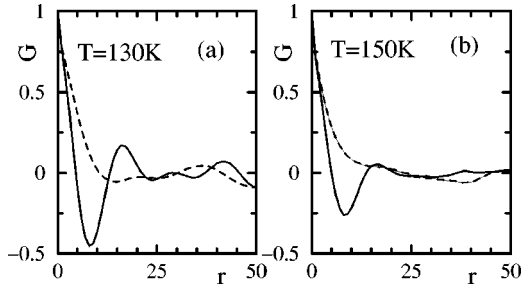

FIG. 4. G at low and intermediate T when some processes are either enhanced or suppressed; symbols as in Fig. 3. (a) Results at 30 ML and $T=130$ K when CC bonds are eliminated; both $G(x,0,t)$ and $G(0,y,t)$ are developing oscillations, corresponding to the formation of anisotropic mounds. (b) Strong CC bonds are assumed; ripples are still present at intermediate T.

CC bonds are important for the formation of well-defined CC ripples because these bonds are needed to correlate atoms in different channels; without them, mounds do not merge coherently into ripples. Even if they are weak, these bonds are rather stable up to 140 K. Also the anisotropy of diffusion is important. We have verified that a more isotropic diffusion give less well-defined CC ripples. The preferential IC diffusion causes the formation of islands which are mainly oriented along the CC direction already in the submonolayer regime.[12]

Now we look for the microscopic processes causing the disappearance of the ripples at intermediate T. One may be tempted to attribute this to the onset of intralayer CC diffusion (process b) or of interlayer IC mobility (process c),[3] but this is not the case. In fact, both b and c are not frequent up to 160 K, and, suppressing them, we have not noticed any qualitative change in the results of Figs. 2 and 3. Therefore another mechanism is responsible for the disappearance of ripples, and it is again related to the CC bonds. In fact, if we assume that CC bonds are as strong as IC bonds (0.18 eV instead of 0.02 eV), so that they cannot be broken, CC ripples do form at low T, but they do not disappear at intermediate T [see Fig. 4(b), where a periodicity is clearly seen only in $G(x,0,t)$, and compare it to Fig. 3(c)]. Therefore, the disappearance of CC ripples is related to the breaking of CC bonds. This shows that, at low and intermediate T, weak CC bonds and preferential IC diffusion, together with the absence of interlayer mobility, are the key ingredients for the explanation of the growth morphologies.

Let us now turn to the analysis of the high-T regime (close to 200 K), where the experiments[3] show that ripples rotate to the IC direction. In this regime, KMC simulations become more and more time consuming, because of the increased number of diffusion moves in between deposition events, so that the study by the model we used at low T becomes cumbersome. Because of that, we have employed a simplified model, in which we assume a simple cubic geometry with the same anisotropic barriers (see Fig. 1), without cascades. This simplified model reproduces the qualitative features of the full fcc model at low and intermediate T, but gives an enhanced roughness of the surface at low T. As an example, we consider $T=210$ K as in experiments.[3] At this

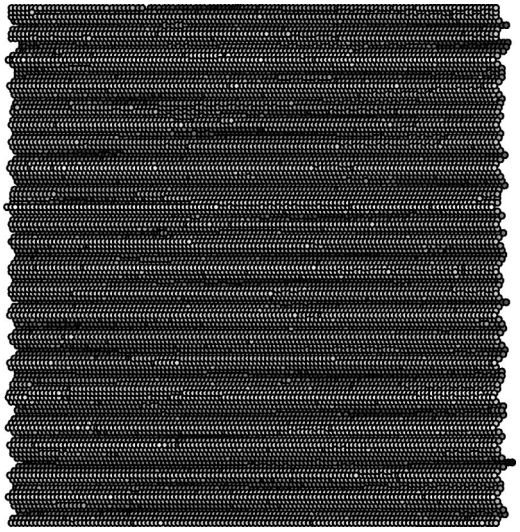

FIG. 5. Snapshot after 30 ML at 210 K with the simplified model (see text). In-channel ripples are evident

T, CC bonds are negligible, while IC bonds are still stable. In Fig. 5 (where the IC direction is horizontal) we show a snapshot of the surface after deposition at the same flux and coverage as in Fig. 2. Clearly, well-defined ripples are now formed along the IC direction, again in agreement with experiments,[3] as can also be seen in Fig. 6(a). At high T, if one suppresses CC diffusion, IC ripples do not form at 210 K [see Fig. 6(b)]. Moreover, we have checked that, suppressing completely interlayer mobility, well-defined IC ripples are not formed. This demonstrates that the above processes must be active to complete ripple rotation, in agreement with the qualitative explanation in Ref. 3. However, these processes are not sufficient, because also CC bond breaking is needed: in fact we have verified that strong CC bonds inhibit IC ripple formation even at high T. Concerning the quantitative agreement at high T, we find that our simulations form the IC ripples in the same T range as in the experiments,[3] but λ_y is shorter by more than a factor of 2 at 210 K. Here λ_y increases with T, in agreement with experiments, but remains in comparison too short. However, one cannot expect quantitative accuracy for the simplified model, so that the search of the accuracy at high T is left for future work. What is

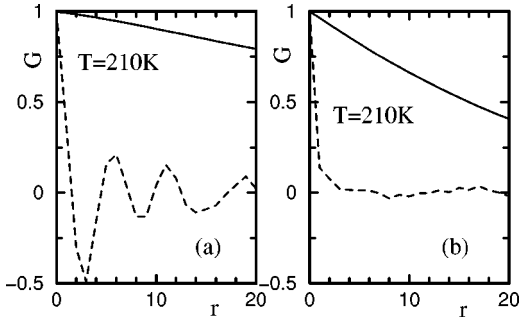

FIG. 6. G at 210 K; symbols as in Fig. 3. (a) Results at the conditions of Fig. 5, with clear oscillations is $G(0,y,t)$ and a long-distance decay of $G(x,0,t)$. (b) CC diffusion is suppressed; the decay of $G(0,y,t)$ indicates that IC ripples are not formed.

important is that the general ripple behavior is reproduced, in the right T range, even with such a simplification.

In conclusion, we have proposed a model for Ag/Ag(110) growth which reproduces quantitatively the experimental results at low and intermediate temperatures (up to 160 K) and, in a simplified version, gives a general semiquantitative description up to 210 K. This model has allowed a complete study of the microscopic mechanisms of ripple formation and rotation in order to answer to the following questions: Why do ripples form on this surface? And why do the ripples rotate? In particular, we have demonstrated that at low T, the key ingredients for cross-channel ripple formation are the existence of weak CC bonds, together with a preferential in-channel diffusion; the role of CC bonds is crucial to correlate structures along the CC direction, determining the growth of ripples instead of simple mounds. When T is raised to the intermediate regime, these ripples transform into mounds because of the breaking of such bonds, while CC diffusion and IC interlayer processes are not important for this transformation. On the other hand, both intralayer CC and interlayer IC diffusion are needed to complete the high-T rotation towards the IC direction. Our results demonstrate the essential role of the anisotropy of bonds to reproduce the morphology of surface instabilities as they develop with temperature.

ACKNOWLEDGMENTS

We acknowledge useful discussions with F. Buatier, G. Costantini, C. Boragno, and U. Valbusa, and thank A. Pimpinelli and Z. Zhang for a careful reading of the manuscript. R.F. acknowledges financial support from the Italian MURST under Project No. 9902112831; F.H. thanks the ICTP, Trieste, Italy and the Swedish International Development Agency (SIDA) for financial support. F.H. is on leave from IMSP/FAST, Université Nationale du Benin, Porto-Novo, Benin.

*Corresponding author. Electronic address: ferrando@fisica.unige.it

[1] J. Villain, J. Phys. I **1**, 19 (1991); J.G. Amar and F. Family, Phys. Rev. Lett. **77**, 4584 (1996); M.V. Ramana Murty and B.H. Cooper, *ibid.* **83**, 352 (1999); S. Das Sarma, S. Punyindu, and Z. Toroczkai, Surf. Sci. **457**, L369 (2000).

[2] J.K. Zuo and J.F. Wendelken, Phys. Rev. Lett. **78**, 2791 (1997); J.A. Stroscio *et al.*, *ibid.* **75**, 4246 (1995); K. Thurmer, R. Koch, M. Weber, and K.H. Rieder, *ibid.* **75**, 1767 (1995); M. Bott, T. Michely, and G. Comsa, Surf. Sci. **272**, 161 (1992).

[3] F. Buatier de Mongeot, G. Costantini, C. Boragno, and U. Valbusa, Phys. Rev. Lett. **84**, 2445 (2000).

[4] C.J. Liu, J.M. Cohen, J.B. Adams, and A.F. Voter, Surf. Sci. **253**, 334 (1991).

[5] R. Ferrando, Phys. Rev. Lett. **76**, 4195 (1996).

[6] M.D. Johnson, C. Orme, A.W. Hunt, D. Graff, J. Sudijono, L.M. Sander and B.G. Orr, Phys. Rev. Lett. **72**, 116 (1994).

[7] F. Hontinfinde, R. Ferrando, and A.C. Levi, Surf. Sci. **366**, 306 (1996).

[8] R. Ferrando, F. Hontinfinde, and A.C. Levi, Phys. Rev. B **56**, 4406 (1997).

[9] F. Buatier de Mongeot, G. Costantini, C. Boragno, and U. Valbusa (unpublished).

[10] Y.-W. Mo, B.S. Swartzentruber, R. Kariotis, M.B. Webb, and M.G. Lagally, Phys. Rev. Lett. **63**, 2393 (1989).

[11] Y. Li, M.C. Bartelt, J.W. Evans, N. Waelchli, E. Kampshoff, and K. Kern, Phys. Rev. B **56**, 12 539 (1997).

[12] C. Mottet, R. Ferrando, F. Hontinfinde, and A.C. Levi, Surf. Sci. **417**, 220 (1998).

[13] M.C. Bartelt and J.W. Evans, Phys. Rev. Lett. **75**, 4250 (1995); J.G. Amar and F. Family, Phys. Rev. B **54**, 4742 (1996).

[14] M. Rost, P. Smilauer, and J. Krug, Surf. Sci. **369**, 393 (1996); J. Krug and F. Hontinfinde (unpublished).