Far-from-equilibrium Statistical Mechanics of Epitaxial Growth on (110) Crystal Surfaces

Leonardo Golubovic

Department of Physics, West Virginia University, Morgantown, WV 26506-6315

Abstract. We theoretically discuss the far-from-equilibrium interface morphologies occurring in the multilayer homoepitaxial growth and erosion on (110) crystal surfaces. Experimentally, these surfaces exhibit a multitude of interesting non-equilibrium interfacial structures, such as the rippled one-dimensional periodic states that are not present in the homoepitaxial growth and erosion on the high symmetry (100) and (111) crystal surfaces. Within a unified phenomenological model, we reveal and elucidate this multitude of novel surface morphologies on (110) surfaces as well as the transitions between them. By analytic arguments and numerical simulations, we address experimentally observed transitions between two types of rippled states on (110) surfaces. We discuss several intermediary interface states intervening, via consecutive transitions, between the two rippled states. One of them is the Rhomboidal Pyramid State, theoretically predicted by Golubovic, Levandovsky, and Moldovan [Phys. Rev. Lett. 89, 266104 (2002); Phys. Rev. E74, 61601(2006)] and subsequently seen, by de Mongeot and coworkers, in the epitaxial erosion of Cu(110) and Rh(110) surfaces [A. Molle et al., Phys. Rev. Lett. 93, 256103 (2004), and A. Molle et al., Phys. Rev. B 73, 155418 (2006)]. In addition, we find a number of interesting intermediary states having structural properties somewhere between those of rippled and pyramidal states. Prominent among them are the Rectangular Rippled states of long roof-like objects (huts) recently seen on Ag(110) surface. We also predict the existence of a striking interfacial structure that carries non-zero, persistent surface currents. Periodic vortex lattice formed in this so called Buckled Rippled interface state is a far-from-equilibrium relative of the self organized convective flow patterns in hydrodynamic systems. We discuss the coarsening growth of the multitude of the interfacial states on (110) crystal surfaces.

Mutilayer epitaxial growth and erosion of crystal surfaces often exhibits the formation of fascinating surface nanostructures, [1-10]. At their origin is the classical Ehrilich-Schwoebel-Villain instability, [5,6]. Thus, on high symmetry (100) and (111) surfaces, pyramidal structures are frequently seen in the homoepitaxial deposition growth and erosion, which is essentially a deposition of surface vacancies by ion beams, [7]. These pyramids grow in time via far-from-equilibrium coarsening processes studied in numerous experiments and simulations on high symmetry crystal surfaces, such as the square symmetry (100) surfaces and the hexagonal symmetry (111) surfaces, [1-4,7,8,12]. However, much less is known about related growth phenomena on low symmetry surfaces. Thus, the far-from-equilibrium interfacial structures formed in the epitaxial growth and erosion on rather typical rectangular symmetry (110) crystal surfaces have attracted attention only recently, [9,10]. Rather than pyramids, Rippled one-dimensionally (nearly) periodic structures are more commonly seen on these surfaces, such as Fe(110) [11], Ag(110) [9,10], Cu(110) and Rh(110) [13]. There are two types of these Rippled states, with their wave-vectors oriented along the two perpendicular principal axes of the (110) surfaces. In addition to the Rippled states, intriguing intermediary states of interface have been recently revealed in the "90-degrees ripple rotation" transitions between the two types of Rippled states on Ag (110) surfaces [9,10], and, more recently, on Cu(110) and Rh(110) surfaces [13]. These intermediary states are believed to have a pyramidal character. Exotic pyramidal structures have been seen also on Al(110), having the form of self-assembled “huts”, i.e., roof-like pyramids [14].

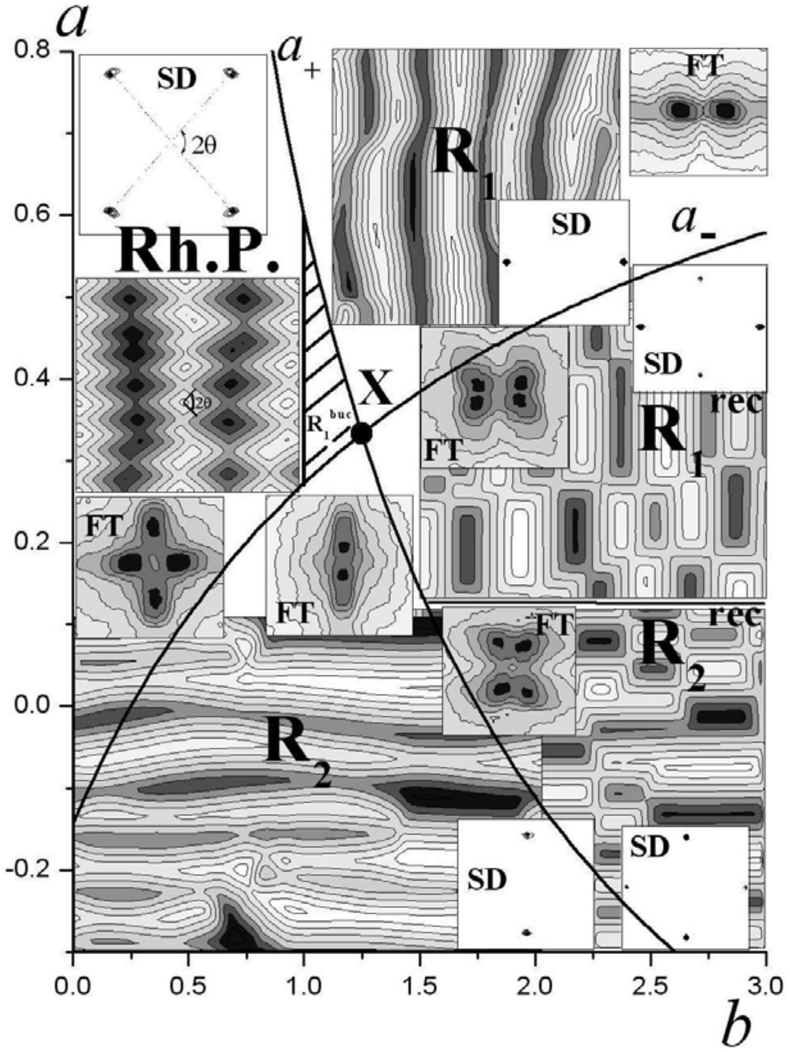

FIGURE 1. Our unified far-from-equilibrium phase diagram for the epitaxial growth and erosion of (110) crystal surfaces: interface contour plots from our simulations, Fourier Transforms (FT), i.e., near in-phase diffraction patterns, and interface slope distributions (SD), i.e., out-of-phase diffraction patterns of various interfacial states: the Rhomboidal Pyramid State (RhP), two ordinary Rippled states R_1 and R_2 and two Rectangular Rippled states $R_1^{(rec)}$ and $R_2^{(rec)}$. The Buckled Rippled State is in the hatched domain.

Within a unified interface dynamics model [2,3], here we discuss these far-from-equilibrium phenomena occurring in the multilayer epitaxial growth and erosion on (110) crystal surfaces. We expose generic multitude of novel interfacial states on (110) surfaces as well as the far-from-equilibrium phase transitions between these states. By analytic arguments and numerical simulations, we obtain the generic non-equilibrium phase diagram of (110) surfaces; see Fig. 1. It reproduces experimentally observed transitions between the two aforementioned types of Rippled states. The model predicts a number of intermediary interface states intervening, via consecutive transitions, between the two Rippled states. One of them is the Rhomboidal Pyramid State *RhP* (Fig. 2(b)) that has been theoretically predicted by us in [2] and subsequently seen by de Mongeot and coworkers in the epitaxial erosion of Cu(110) and Rh(110) surfaces [13].

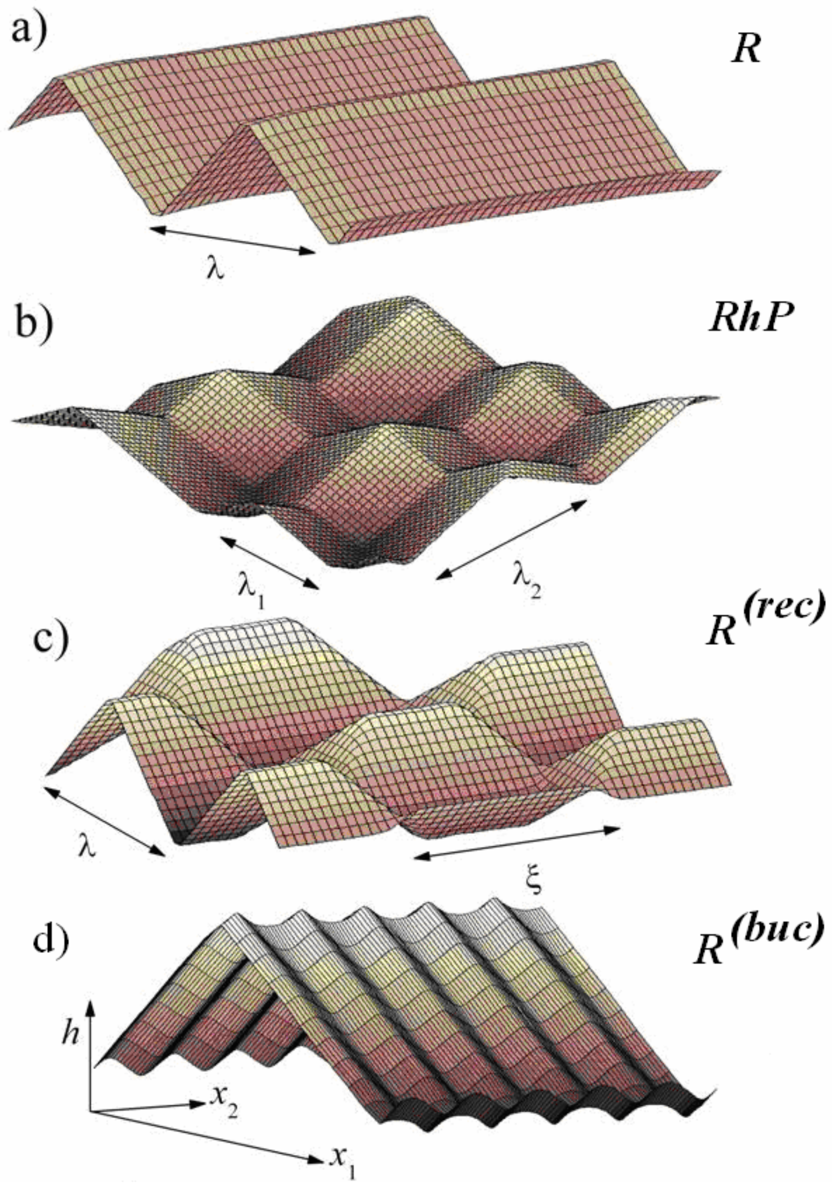

FIGURE 2. Predicted interfacial morphologies in the epitaxial growth and erosion of (110) surfaces: (a) Ordinary Rippled R state, (b) Rhomboidal Pyramid RhP state, (c) Rectangular Rippled $R^{(rec)}$ state, (d) Buckled Rippled $R^{(buc)}$ state.

In addition, our model yields a number of other experimentally interesting intermediary states having structural properties between those of rippled and pyramidal states; see Fig. 2. Among them are two Rectangular Rippled $R^{(rec)}$ states of long roof like objects (huts) seen in Figs. 1 and 2. These states, theoretically anticipated by us in [2,3], represent the long time scale form of the aforementioned roof-like pyramid (huts) state seen to emerge in the epitaxial growth on Al(110) [14]. In fact, our basic Rectangular Rippled structure [the checkerboard arrangements of huts and pits (inverted huts)] has been clearly seen in the ion erosion experiments on Ag(110) [see Ref. 10 and Fig. 4(d) therein]. We predict that there are two types of the Rectangular Rippled states, with their wave-vectors oriented along the two perpendicular principal axes of the (110) surfaces; see Fig. 1. To elucidate the transition between these states, we generalize the classical Gibbs phase coexistence rule to the situations involving far-from-equilibrium phase transitions, in the *absence* of an effective free energy governing dynamics. Our theory [2,3] has revealed also the exotic Buckled Rippled $R^{(buc)}$ interface state; see Fig. 2. Unlike the usual interfacial states (see, e.g., Refs. [1], [2], [5], and therein), the interfaces in the Buckled Rippled state exhibit non-vanishing, persistent surface currents forming a periodic convection like pattern of vortices. This interface state is a far-from-equilibrium relative of the Rayleigh-Benard's and other self organized convective flow patterns occurring in hydrodynamic systems [15]. Our model can be used to explore the dynamics of various interfacial states on (110) surfaces [2,3]. In particular, we find that the coarsening, i.e., the growth of the spatial period of the Rippled states on (110) surfaces is mediated by ensembles of climbing dislocations destroying perfect periodicity of these states.

ACKNOWLEDGMENTS

The author is grateful to Dr. Mirjana Golubovic for lifetime inspiration, and to Drs. Artem Levandovsky and Dorel Moldovan for collaborating in this research. This research was supported in part by DAAD through the grant No. A/02/15054 sec.315.

REFERENCES

1. D. Moldovan and L. Golubovic, *Phys. Rev.* E **61**, 6190 (2000).
2. L. Golubovic, A. Levandovsky, and D. Moldovan, *Phys. Rev. Lett.* **89**, 266104 (2002).
3. A. Levandovsky, L. Golubovic, and D. Moldovan, *Phys. Rev.* E **74**, 61601 (2006). See, also, A. Levandovsky, L. Golubovic, and D. Moldovan, in "Modeling of Morphological Evolution at Surfaces and Interfaces", edited by J. Evans, C. Orme, M. Asta, and Z. Zhang, *Mater. Res. Soc. Symp. Proc.* Vol. **859E**, article JJ7.7 (© 2005 Materials Research Society JJ7.7.1).
4. A. Levandovsky and L. Golubovic, *Phys. Rev.* B **69** R, 241402 (2004).
5. See, J. Krug, *Adv. in Phys.* **46**, 139 (1997), and *Physica* A **313**, 47 (2002).
6. J. Villain, *J. Phys. I (France)* **1**, 19 (1991).
7. T. Michely, et al., *Phys. Rev. Lett.* **86**, 2589 (2001); J.-K. Zuo and J. F. Wendelken, *Phys. Rev. Lett* **78**, 2791 (1997).
8. J. Amar, *Phys. Rev.* **B** 60, R 11 317 (1999).
9. F. Bautier de Mongeot, G. Constantini, C. Boragno, and U. Valbusa, *Phys. Rev. Lett*. **84**, 2445 (2000).
10. G. Constantini, S. Rusponi, F. Bautier de Mongeot, C. Boragno, and U. Valbusa, *J. Phys.: Cond. Matter* **13**, 5875 (2001).
11. M. Albrecht, H. Fritzsche and U. Gradmann, *Surf. Sci*. **294**, 1 (1993).
12. K. J. Caspersen, et al., *Phys. Rev.* B **65**, 193407 (2002).
13. A. Molle, F. Bautier de Mongeot, A. Molinari, F. Xiaerding, C. Boragno, and U. Valbusa, *Phys. Rev. Lett.* **93**, 256103 (2004), and A. Molle, F. Bautier de Mongeot, A. Molinari, C. Boragno, and U. Valbusa, *Phys.Rev.* **B** 73, 155418 (2006). See, also, F. Bautier de Mongeot, A. Toma, A. Molle, S. Lizzit, L. Petaccia, and A. Baraldi, *Phys. Rev. Lett.* **97**, 056103 (2006).
14. F. Bautier de Mongeot, et al., *Phys. Rev. Lett.* **91**, 016102 (2003).
15. See, e.g., H. Haken, "Synegetics" (Springer-Verlag, 1978).