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Non-universal mound formation in non-equilibrium surface growth

S. Das Sarma, P. Punyindu *, Z. Toroczkai

Department of Physics, University of Maryland, College Park, MD 20742-4111, USA

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Abstract

We demonstrate, using well-established non-equilibrium limited-mobility solid-on-solid growth models, that mound formation in the dynamical surface growth morphology does not necessarily imply the existence of a surface edge diffusion bias ('the Schwoebel barrier'). We find mounded morphologies in several non-equilibrium growth models that incorporate no Schwoebel barrier. Our numerical results indicate that mounded morphologies in non-equilibrium surface growth may arise from a number of distinct physical mechanisms, with the Schwoebel instability being one of them. © 2000 Elsevier Science B.V. All rights reserved.

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In vacuum deposition growth of thin films or epitaxial layers, e.g. molecular beam epitaxy (MBE), it is common [1–10] to find mound formation in the evolving dynamical surface growth morphology. Although the details of the mounded morphology could differ considerably depending on the systems and growth conditions, the basic mounding phenomenon in surface growth has been reported in a large number of recent experimental publications [1–10]. The typical experiment [1–10] monitors vacuum deposition growth on substrates using scanning tunneling microscope (STM) and/or atomic force microscope (AFM) spectroscopies. Growth mounds are observed under typical MBE-type growth conditions, and the resultant mounded morphology is statistically analyzed by studying the dynamical surface height $h(\boldsymbol{r}, t)$ as a function of the position $\boldsymbol{r}$ on the surface and growth time t. Much attention has focused on this ubiquitous phenomenon of mounding and the associated pattern formation during non-equilibrium surface growth for reasons of possible technological interest (e.g. the possibility of producing controlled nanoscale thin film or interface patterns) and fundamental interest (e.g. understanding non-equilibrium growth and pattern formation).

The theoretical interpretation of the mounding phenomenon has often been based [1–10] on the step-edge diffusion bias [11] or the so-called Schwoebel barrier [12,13] effect (also known as the Ehrlich–Schwoebel [12,13], or ES, barrier). The basic idea of the ES barrier-induced mounding (often referred to as an instability) is simple: the

* Corresponding author. Fax: +1-301-314-9465.
E-mail address: patcha@physics.umd.edu (P. Punyindu)

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ES effect produces an additional energy barrier for diffusing adatoms on terraces from coming 'down' toward the substrate, thus probabilistically inhibiting attachment of atoms to lower or down-steps and enhancing their attachment to upper or up-steps; the result is therefore mound formation because deposited atoms cannot come down from upper to lower terraces and so three-dimensional mounds or pyramids result as atoms are deposited on the top of already existing terraces.

The physical picture underlying mounded growth under an ES barrier is manifestly obvious, and clearly the existence of an ES barrier is a *sufficient condition* [11] for mound formation in non-equilibrium surface growth. Our interest, in this paper, is to discuss the *necessary* condition for mound formation in non-equilibrium surface growth morphology — more precisely, we want to ask the inverse question, namely whether the observation of mound formation requires the existence of an ES barrier. Through concrete examples we demonstrate that the mound formation in non-equilibrium surface growth morphology does *not* necessarily imply the existence of an ES barrier, and we contend that the recent experimental observations of mound formation in non-equilibrium surface growth morphology should not be taken as definitive evidence in favor of an ES barrier-induced universal mechanism for pattern formation in surface growth. Mound formation in non-equilibrium surface growth is a non-universal phenomenon, and could have very different underlying causes in different systems and situations, with the Schwoebel instability being one particular mechanism (among many) for the mounded morphology.

Before presenting our results we point out that the possible non-universality in surface growth mound formation (i.e. mounds do not necessarily imply an ES barrier) has recently been mentioned in at least two experimental publications [14,15], where it was emphasized that the mounded patterns seen on Si [14], GaAs [15], and InP [15] surfaces during MBE growth were not consistent with the phenomenology of a Schwoebel instability. In two other recent experimental publications [16,17] mound formation during semiconductor surface growth (Ge, GaAs) was carefully analyzed using the prevailing Schwoebel instability phenomenology with a conclusion not very dissimilar from that in Ref. [15]. In particular, the ES barrier-based analyses of the experimental data in both Refs. [16,17] produced rather weak Schwoebel effects in both experiments, leading to the conclusion in both experiments that the Schwoebel instability in all likelihood is playing a small to negligible role in the observed mound formations in Refs. [16,17]. Very recently, experimental observations of striking mound formation [18,19] in Au and MgO vapor deposition growth have been interpreted without invoking any ES barrier effect. Thus, the observed mound formation in the non-equilibrium surface growth in Refs. [14–19] is interpreted essentially without invoking any key role being played by an ES barrier, whereas the mounded growth morphologies in Refs. [1–10] have mostly been interpreted as arising essentially due to a Schwoebel instability. Thus the inevitable conclusion from recent experimental observations [14–19] is that the mound formation in surface growth does not necessarily arise from the universal mechanism of a Schwoebel instability [1–10], but may be caused by different non-universal mechanisms in different experimental situations. The purpose of the current article is to explore this non-universality in the mound formation in some detail using simple solid-on-solid (SOS) growth models where the kinetic mechanisms leading to the mounded morphologies are explicitly obvious, and therefore compelling conclusions can be drawn about the precise physical mechanism producing the mounds. A direct comparison between experimental results and our rather simplistic limited mobility non-equilibrium SOS models, however, is unwarranted due to the extreme simplicity in the growth and diffusion rules in our models — our models do serve the purpose of explicitly demonstrating the fact that mounded morphologies can arise without any ES barriers whatsoever.

There have been two proposed mechanisms in the literature that lead to mounding without any explicit ES barrier. One of them invokes [20] a preferential attachment to up-steps compared with down-steps (the so-called 'step–adatom' attraction), which, in effect, is equivalent to having an ES barrier because the attachment probability to

down-steps is lower than that to up-steps exactly as it is in the regular ES barrier case [11–13] — we therefore do not distinguish it from the ES barrier mechanism, and, in fact, within the simple growth models we study, these two *energetic* mechanisms are physically and mathematically indistinguishable. The second mounding alternative [21,22], which is a purely topologic–kinetic effect, is the so-called *edge-diffusion*-induced mounding, where diffusion of adatoms around cluster edges is shown to lead to mound formation during non-equilibrium surface growth even in the absence of any finite ES barrier. One of the concrete examples we discuss below, the spectacular pyramidal pattern formation (Fig. 3c) in the $2+1$ dimensional d noise reduced Wolf–Villain (WV) model [23], arises from such a non-equilibrium edge diffusion effect (perhaps in a somewhat unexpected context). We also demonstrate, using the WV model and the Das Sarma–Tamborenea (DT) model [24–27], that mound formation during non-equilibrium surface growth is, in fact, almost a generic feature of *limited mobility* SOS discrete growth models [23–31], which typically have comparatively large values of the roughness exponent α [28–31] characterizing the growth morphology. We find that a large roughness exponent coupled with atomistic SOS growth almost invariably leads to visually mounded growth morphology. Below we demonstrate that mound formation in surface morphology arising from this generic 'large α' effect (without any explicit ES barrier) is often qualitatively virtually indistinguishable from that in growth under an ES barrier. Mound formation in the presence of strong edge diffusion [21,22] (as in the $d=2+1$ WV model in Fig. 3) is, on the other hand, morphologically quite distinct from the ES barrier- or the large α-induced mound formation.

Our results are based on the extensively studied [28–31] limited mobility SOS non-equilibrium WV [23] and DT [24–27] growth models. Both models have been widely studied [28–31] in the context of kinetic surface roughening in non-equilibrium SOS epitaxial growth — the interest in and the importance of these models lie in the fact that these were the first concrete physically motivated growth models falling outside the well-known Edwards–Wilkinson–Kardar–Parisi–Zhang [28–31] generic universality class in kinetic surface roughening. Both models involve random deposition of atoms on a square lattice singular substrate (with a growth rate of 1 layer/s, where the growth rate defines the unit of time) under the SOS constraint with no evaporation or desorption. An incident atom can diffuse instantaneously before incorporation if it satisfies certain diffusion rules, which differ slightly in the two models. In the WV model the incident atom can diffuse within a diffusion length l (which is taken to be one with the lattice constant being chosen as the length unit, i.e. only nearest-neighbor diffusion, in all the results shown in this paper — larger values of l do not change our conclusions) in order to maximize its local coordination number or equivalently the number of nearest-neighbor bonds it forms with other atoms. (If there are several possible final sites satisfying the maximum coordination condition equivalently then the incident atom chooses one of those sites with equal random probability and if no other site increases the local coordination compared with the incident site then the atom stays at the incident site.) The DT model is similar to the WV model except for two crucial differences: (1) only incident atoms with no lateral bonds (i.e. with the local coordination number of one — a nearest-neighbor bond to the atom below is necessary to satisfy the SOS constraint) are allowed to diffuse (all other deposited atoms, with one or more lateral bonds, are incorporated into the growing film at their incident sites); (2) the incident atoms move only to *increase* their local coordination number (and not to maximize it as in the WV model) — all possible incorporation sites with finite lateral local coordination numbers are accepted with random equal probability. Although these two differences between the DT and the WV model have turned out to be crucial in distinguishing their asymptotic universality class, the two models exhibit very similar growth behavior for a long transient pre-asymptotic regime. It is easy to incorporate [32] an ES barrier in the DT (or WV) model by introducing differential probabilities P_u and P_l for adatom attachment to an upper and a lower step respectively — the original DT model [24–27] has $P_u=P_l$, and an ES barrier can be

explicitly incorporated [32] in the model by having $P_1 < P_u \leq 1$. We call this situation [32] the DT-ES model (we use $P_u = 1$ throughout with no loss of generality). We also note, as mentioned above, that within the DT-ES model the ES barrier [11–13] ($P_1 < P_u$) and the step–adatom attraction [20] ($P_u > P_1$) are manifestly equivalent, and we therefore do not consider them as separate mechanisms. We note also that in some of our simulations below we have used the noise reduction technique [32,33], which has previously been successful in limited mobility growth models in reducing the strong stochastic noise effect through an effective coarse-graining procedure. All three models described above are studied in both one-dimensional substrate ($d = 1+1$) and two-dimensional substrate ($d = 2+1$) systems with periodic boundary conditions being used in all simulations. Detailed descriptions of DT and WV models are available in the literature [23–33].

In Figs. 1 and 2 we present our $d = 1+1$ growth simulations, which demonstrate the point we want to make in this paper. We show in Fig. 1 the simulated growth morphologies at three different times for four different situations, two of which (Fig. 1a and b) have finite ES barriers and the other two (Fig. 1c and d) do not. The important point we wish to emphasize is that, while the four morphologies and their dynamical evolutions shown in Fig. 1 are quite distinct in their details, they all share one crucial common feature: they all indicate mound formation, although the details of the mounded morphologies and the controlling length scales are obviously quite different in the different cases. Just the mere observation of mounded morphology, which is present in Fig. 1c and d, thus does not necessarily imply the existence of an ES barrier. To further quantify the mounding apparent in the simulated morphologies of Fig. 1 we show in Fig. 2 the calculated height–height correlation function, $H(r) \approx \langle h(\boldsymbol{x}) h(\boldsymbol{r}+\boldsymbol{x}) \rangle_x$, along the surface for two different times. All the calculated $H(r)$ show noisy oscillations as a function of r, which implies mound formation (corresponding to the noisy mounded morphologies of Fig. 1). It is indeed true that the presence of considerable stochastic noise associated with the deposition process in the DT and WV models make the $H(r)$-oscillations quite noisy, but the important feature to note in Fig. 2 is that the qualitative oscillatory nature of $H(r)$ in situations with or without an ES barrier is essentially the same. Thus, the mound formation, although noisy, is qualitatively similar with or without an ES barrier in Fig. 1 and 2. We have explicitly verified that such growth mounds [or equivalently $H(r)$ oscillations] are completely

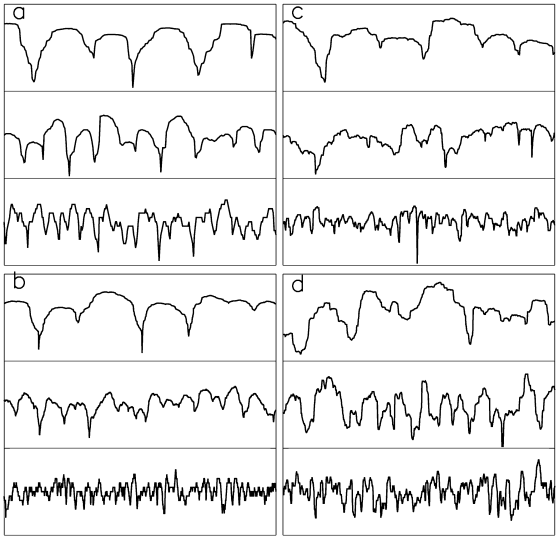


Fig. 1. Dynamical morphologies at 10^2, 10^4 and 10^6 monolayers (ML) for (a) DT-ES with $P_1 = 0.5$, $P_u = 1$; (b) DT-ES with $P_1 = 0.9$, $P_u = 1$; (c) DT; and (d) WV models.

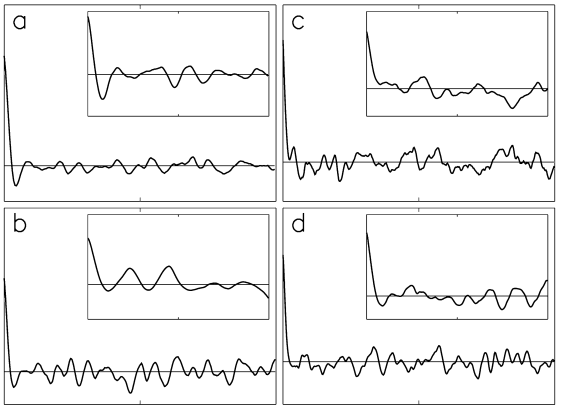


Fig. 2. The height–height correlation function $H(r)$ at 10^2 ML (main plots) and 10^4 ML (insets) corresponding respectively to the morphologies in Fig. 1.

absent in the growth models [28–31] that correspond to the generic Edwards–Wilkinson–Kardar–Parisi–Zhang universality class, and arise only in the DT and WV limited mobility growth models which are known to have large value of the roughness exponent α arising from (linear or non-linear) surface diffusion processes. In fact, the effective α in the DT and WV models is essentially [23–31] unity (in $d=1+1$), which is the same as what one expects in a naive theoretical description of growth under the ES barrier (although the underlying growth mechanisms are completely different in the two situations). We believe that any surface growth involving a 'large' roughness exponent ($0.5<\alpha\leqslant 1$) will invariably show 'mounded' morphology independent of whether there is an ES barrier in the system or not. We contend that this effectively large α is the physical origin for the mounded morphology in semiconductor MBE growth where one expects the surface-diffusion-driven linear or non-linear conserved fourth-order (in contrast to the generic second-order) dynamical growth universality [28–31] class to apply which has the asymptotic exponent: $\alpha(d=1+1)\approx 1$; $\alpha(d=2+1)\approx 0.67$ (nonlinear) and 1 (linear). One recent experimental paper [15], which reports the observation of mounded GaAs and InP growth with $\alpha\approx 0.5$–0.6, has explicitly made this case, and other recent reported mound formations [16,17] in semiconductor MBE growth are also consistent with our contention that mounds may arise from a large effective roughness exponent rather than a Schwoebel instability. Two very recent experimental publications [18,19] have reached the same conclusion in non-semiconductor MBE growth studies — in these recent publications [18,19] spectacular mounded surface growth morphologies have been interpreted on the basis of the fourth-order conserved growth equations [23–33]. The crucial message of our simulated $d=1+1$ growth morphologies in Figs. 1 and 2 is the fact that the mound formation with (Figs. 1a and b and 2a and b) and without ES barrier (Figs. 1c and d and 2c and d) are qualitatively similar, and therefore the mere observation of a mounded morphology does not necessarily imply a Schwoebel instability.

Finally, in Figs. 3 and 4 we present our results for the physically more relevant $d=2+1$ non-equilibrium surface growth. In Fig. 3a–c we show the growth morphologies for the DT-ES, DT, and the noise-reduced WV model respectively, whereas in Fig. 4 we show the scaled height–height correlation function for the mounded morphologies

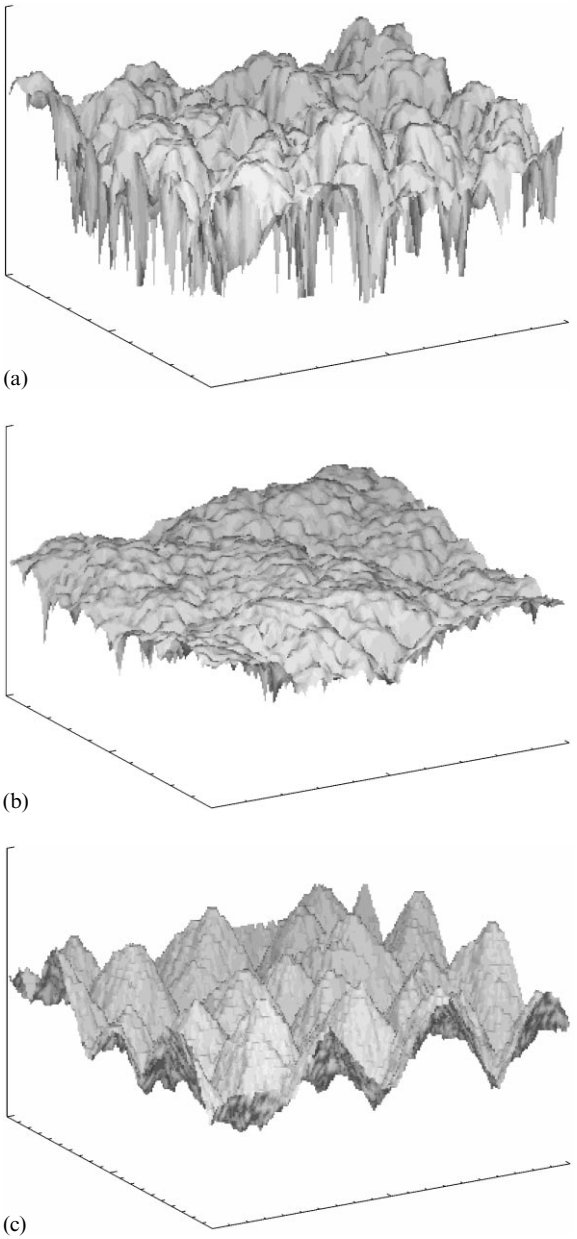


Fig. 3. Morphologies from the (a) DT-ES with $P_l=0.5$, $P_u=1$; (b) DT; and (c) noise-reduced WV models.

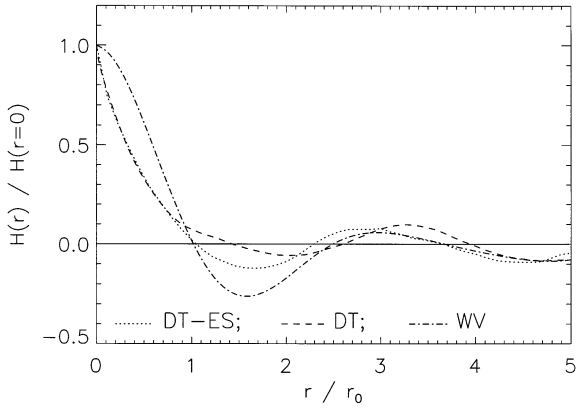


Fig. 4. The scaled $H(r)$ correlation functions corresponding to the morphologies shown in Fig. 3 (r_o: mound radius).

depicted in Fig. 3. It is apparent that all three models (one with an ES barrier and the other two without) have qualitatively similar oscillations in $H(r)$ indicating mounded growth, and the differences in the mounding between the growth models are purely quantitative. Again, the important point is that mounded morphologies with and without ES barriers manifest similar oscillating behavior in $H(r)$, indicating that such an oscillatory height–height correlation function by itself does not establish a Schwoebel instability. Thus we come to the same conclusion: mound formation, by itself, does not imply the existence of an ES barrier; the details of the morphology obviously will depend on the existence (or not) of an ES barrier. We note that the effective values of the roughness exponent are very similar in Fig. 3a and b, i.e. with and without an ES barrier, both being approximately $\alpha \approx 0.5$ (far below the asymptotic value $\alpha \approx 1$ expected in the ES barrier growth — we have verified that this asymptotic $\alpha \approx 1$ is achieved in our simulations at an astronomically long time of 10^9 layers). The most astonishing result we show in Fig. 3 is the spectacular pyramidal mound formation in the $d=2+1$ noise-reduced WV model (without any ES barrier), which has not previously been reported in the literature. The strikingly regular pyramidal pattern formation (Fig. 3c) in our noise-reduced WV model in fact has a magic slope and strong coarsening behavior. The pattern is very reminiscent of the theoretical growth model studied earlier in Refs. [34,35] in the context of non-equilibrium growth under an ES barrier, where very similar patterns with slope selection were proposed as a generic scenario for growth under a Schwoebel instability. In our case of the noise-reduced $d=2+1$ WV model of Fig. 3c there is no ES barrier, but there is strong cluster-edge diffusion. This strong edge diffusion (which obviously cannot happen in $1+1$-dimensional growth) arises in the WV model (but not in the DT model) from the hopping of adatoms which have finite lateral nearest-neighbor bonds (and are therefore the edge atoms in a cluster). This edge diffusion (discussed in entirely different contexts in Refs. [21,22]) leads to an 'uphill' surface current in the [111] direction, which leads to the formation of the slope-selected pyramidal patterned growth morphology. Although noise reduction enhances the edge current strengthening the pattern formation (the uphill current is extremely weak in the ordinary WV model due to the strong suppression by the deposition shot noise), our results of Fig. 3 establish compellingly that the WV model in $d=2+1$ is, in fact, unstable (uphill current) in contrast to the situation in $d=1+1$. Thus, the WV model belongs to totally different universality classes in $d=1+1$ and $2+1$ dimensions! We mention that in (unphysical) higher (e.g. $d=3+1$, $4+1$, etc.) dimensions the WV model would be even more unstable, forming even stronger mounds, since the edge diffusion effects will increase substantially in higher dimensions due to the possibility of many more configurations of nearest-neighbor bonding. Such unstable growth in a high-dimensional ($d>2+1$) WV model has previously been reported [36] in the literature without any physical explanation. We have therefore provided the explanation for the long-standing puzzle of an instability in the high-dimensional ($d>2+1$) WV model simulations that were reported [36] in the literature some years ago. More details on this phenomenon will be published elsewhere [37].

In conclusion, we have shown, through concrete examples, that while a Schwoebel instability is certainly sufficient to cause mounded surface growth morphology, the reverse is not true: an ES barrier is by no means necessary to produce mounds, and mound formation in non-equilibrium

surface growth morphology does not necessarily imply the existence of a Schwoebel instability. In particular, we show that a large roughness exponent (without any ES barrier), as in the fourth-order conserved growth universality class [23–33], produces mounded growth morphologies that are indistinguishable from the ES barrier effect. Any experimentally observed mounded morphology therefore requires careful and detailed quantitative analyses [15–19] to determine the physical mechanism (e.g. ES barrier, edge diffusion, large roughness exponent without any ES barrier) underlying its cause — in particular, the existence of a mounded growth morphology by itself may not imply the existence of any significant Schwoebel barrier.

Acknowledgements

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