Mound formation in nonequilibrium surface growth morphology does not necessarily imply a Schwoebel instability

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We demonstrate, using well-established nonequilibrium growth models, that mound formation in the dynamical surface growth morphology does not necessarily imply a surface edge diffusion bias ("the Schwoebel barrier") as has been almost universally accepted in the literature. We find mounded morphologies in several nonequilibrium growth models which incorporate no Schwoebel barrier. Our work should lead to a critical re-evaluation of recent experimental observations of mounded morphologies which have been theoretically interpreted in terms of Schwoebel barrier effects.

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In vacuum deposition growth of thin films or epitaxial layers (e.g. MBE) it is common 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. The typical experiment monitors vacuum deposition growth on substrates using STM and/or 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 as a function of the position on the surface and growth time. Much attention has focused on this ubiquitous phenomenon of mounding and the associated pattern formation during nonequilibrium 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 nonequilibrium growth and pattern formation).

The theoretical interpretation of the mounding phenomenon has been almost exclusively based on the step-edge diffusion bias or the so-called Schwoebel barrier effect (also known as the Ehrlich-Schwoebel, ES, barrier). The basic idea of the ES barrier-induced mounding (often referred to as an instability) is simple: The 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 for mound formation in nonequilibrium surface growth. Our interest in this Letter is to discuss the necessary condition for mound formation in nonequilibrium 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 as has been almost exclusively (and uncritically in our opinion) accepted in the recent literature. Through concrete examples we demonstrate rather compellingly that the mound formation in nonequilibrium surface growth morphology does not necessarily imply the existence of an ES barrier, and we contend (and results presented in this Letter establish) that the recent experimental observations of mound formation in nonequilibrium 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 nonequilibrium surface growth is a non-universal phenomenon, and could have very different underlying causes in different systems and situations.

Before presenting our results we point out that the possible nonuniversality 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 where it was emphasized that the mounded patterns seen on Si and GaAs, InP surfaces during MBE growth were not consistent with the phenomenology of a Schwoebel instability. These papers have, however, been essentially ignored in the literature, and the ES barrier-Schwoebel instability paradigm is by now so well-entrenched in the literature that the experimental observations of mound formation during nonequilibrium growth are often forced to conform to the ES barrier scenario even when the resultant data analyses lead to the inconsistent conclusion about the nonexistence of any ES barrier in the systems under study. There have been only two proposed mechanisms in the literature which lead to mounding without any explicit ES barrier: One of them invokes 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 — we therefore do not distinguish it from the ES barrier mechanism, and in fact, within the growth models we study, these two mechanisms are physically and mathematically indistinguishable. The second mounding alternative is the so-called edge diffusion induced mounding, where diffusion of adatoms around cluster edges is shown to lead to mound formation during nonequilibrium 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. 3(c)) in the 2+1 dimensional (d) noise reduced Wolf-Villain (WV) model, arises from such a nonequilibrium edge diffusion effect (perhaps in a somewhat unexpected context). We also demonstrate, using the WV model and the Das Sarma-Tamborenea (DT) model that mound formation during nonequilibrium surface growth is, in fact, almost a generic feature of limited mobility growth models, which typically have comparatively large values of the roughness exponent (α) characterizing the growth morphology. Below we demonstrate that mound formation in surface morphology arising from this generic “large α” effect (without any explicit ES barrier) is qualitatively virtually indistinguishable from that in growth under an ES barrier. Mound formation in the presence of strong edge diffusion (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 limited mobility nonequilibrium WV and DT growth models. Both models have been widely studied in the context of kinetic surface roughening in nonequilibrium solid-on-solid (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 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/sec. 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 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 has P_u = P_l, and an ES barrier is explicitly incorporated in the model by having P_l < P_u ≤ 1. We call this situation 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 (P_l < P_u) and the step-adatom attraction (P_u > P_l) 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 which have earlier been successful in limited mobility growth models in reducing the strong stochastic noise effect through an effective coarse-graining procedure.

In Fig. 1 and 2 we present our d=1+1 growth simulations, which demonstrate the point we want to make in this Letter. We show in Fig. 1 the simulated growth morphologies at three different times for four different situations, two of which (Fig. 1(a),(b)) have finite ES barriers and the other two (Fig. 1(c),(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 mounds and the controlling length scales are obviously quite different in the different cases. Just the mere observation of the first mound morphology, which is clearly present in Figs. 1(c),(d), thus does not necessarily imply the existence of an ES barrier. To further quantify the mound apparent in the simulated morphologies of Fig. 1 we show
in Fig. 2 the calculated height-height correlation function, \( H(r) \sim \langle h(x)h(r+x) \rangle^{1/2} \), along the surface for two different times.

All the calculated \( H(r) \) show clear oscillations as a function of \( r \), which by definition implies mound formation. It is indeed true that the presence of considerable stochastic noise associated with the deposition process in the DT, WV models make the \( H(r) \)-oscillations quite noisy, but there is no questioning the fact that oscillations are present in \( H(r) \) even when there is no explicit ES barrier present in the growth model (Figs. 2(c),(d)). We have explicitly verified that such growth mounds (or equivalently \( H(r) \) oscillations) are absent in the growth models [1] which correspond to the generic Edwards-Wilkinson-Kardar-Parisi-Zhang universality class, and arise only in the DT, WV limited mobility growth models which exhibit non-generic behavior with a large value of the roughness exponent \( \alpha \). In fact, the effective \( \alpha \) in the DT, WV models is essentially 0.5 unity, 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 < \( \alpha \) < 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 \( \alpha \) is the physical origin for mounded morphology in semiconductor MBE growth where one expects the surface diffusion driven linear or nonlinear conserved fourth order (in contrast to the generic second order) dynamical growth universality [11] class to apply which has the asymptotic exponent : \( \alpha (d=1+1) \approx 1 \); \( \alpha (d=2+1) \approx 0.67 \) (nonlinear), 1 (linear). One recent experimental paper [3], which reports the observation of mounded GaAs and InP growth with \( \alpha \approx 0.5 - 0.6 \), has explicitly made this case, and all the reported mound formations [3] in semiconductor MBE growth are consistent with our contention of the mounds arising from [as in our Fig. 1(c),(d)] a large effective roughness exponent rather than a Schwoebel instability. The calculated [4] ES barrier on semiconductor surfaces are invariably small, providing further support to our contention that mounding in semiconductor surface growth is not an ES barrier effect, but arises instead from the fourth order growth equations [4] [13] which have large roughness exponents. Two very recent experimental publications [13] have reached the same conclusion in non-semiconductor MBE growth studies — in these recent publications [13] spectacular mounded surface growth morphologies have been interpreted on the basis of the fourth order conserved growth equations [13].

Finally, in Fig. 3 and 4 we present our results for the physically more relevant \( d=2+1 \) nonequilibrium surface growth. In Fig. 3(a)-(c) we show the growth morphologies for the DT-ES, DT, and the noise-reduced WV model, respectively whereas in the main Fig. 3 we show the scaled height-height correlation function. 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 between the growth models are purely quantitative. 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. 3(a) and (b), 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).

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. We have therefore provided the explanation for the long-standing puzzle of an instability in high-dimensional (\( d > 2+1 \)) WV model simulations which were reported \cite{fig1} in the literature some years ago. More details on this phenomenon will be published elsewhere \cite{fig2}.

![Fig. 3](image)

**FIG. 3.** The scaled \( H(r) \) correlation functions corresponding to the morphologies shown in the insets. \( (r_0 \equiv \text{mound radius}) \). Insets: Morphologies from the (a) DT-ES with \( P_L = 0.5, \ P_u = 1 \); (b) DT; and (c) noise reduced WV models.

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). The strikingly regular pyramidal pattern formation (Fig. 3(c)) 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 ref. \cite{fig2} in the context of nonequilibrium 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. 3(c), there is no ES barrier, but there is strong cluster-edge diffusion as explained schematically in Fig. 4. 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 leads to an “uphill” surface current (discussed in entirely different contexts in \cite{fig2}), which leads to the formation of the slope-selected pyramidal patterned growth morphology. While 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!

In conclusion, we have shown through concrete examples that, while a Schwoebel instability is certainly sufficient to cause mounded surface growth morphology, the reverse (which has been almost universally assumed in the literature) is simply not true: an ES barrier is by no means necessary to produce mounds, and mound formation in nonequilibrium 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 \cite{fig2} produces mounded growth morphologies which are indistinguishable from the ES barrier effect.

![Fig. 4](image)

**FIG. 4.** Schematic illustration of the instability caused by the line tension in the step-edge. The view is from above, the shaded region is the terrace of higher elevation. Left panel shows a continuum picture while others represent the same on a lattice for different step orientations.

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