Optimal detrended fluctuation analysis as a tool for the determination of the roughness exponent of the mounded surfaces

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We present an optimal detrended fluctuation analysis (DFA) and applied it to evaluate the local roughness exponent in non-equilibrium surface growth models with mounded morphology. Our method consists in analyzing the height fluctuations computing the shortest distance of each point of the profile to a detrending curve that fits the surface within the investigated interval. We compare the optimal DFA (ODFA) with both the standard DFA and nondetrended analysis. We validate the ODFA method considering a one-dimensional model in the Kardar-Parisi-Zhang universality class starting from a mounded initial condition. We applied the methods to the Clarke-Vvedensky (CV) model in 2+1 dimensions with thermally activated surface diffusion and absence of step barriers. It is expected that this model belongs to the nonlinear Molecular Beam Epitaxy (nMBE) universality class. However, an explicit observation of the roughness exponent in agreement with the nMBE class was still missing. The effective roughness exponent obtained with ODFA agrees with the value expected for nMBE class whereas using the other methods it does not. We also characterized the transient anomalous scaling of the CV model and obtained that the corresponding exponent is in agreement with the value reported for other nMBE models with weaker corrections to the scaling.

I. INTRODUCTION

Molecular beam epitaxy (MBE) is a fundamental technique suited to the production of layered materials driven by vapor deposition \textsuperscript{11}. In particular, crystal quality requires that each layer is formed before the next one in nonequilibrium surface conditions \textsuperscript{11,12}, which is achieved at high adatom mobility. At sufficiently high temperatures, the resulting surfaces can be smooth with global roughness no larger than a few nanometers corresponding to one or two atomic layers.

At moderate temperatures, the interface exhibits kinetic roughening \textsuperscript{2,3}. If the growth is ruled by surface diffusion, it is expected that the dynamics in the hydrodynamic limit is described by the non-linear stochastic equation \textsuperscript{3}

$$\frac{\partial \eta}{\partial t} = \eta \nabla^2 + \nu \nabla^4 + \lambda \nabla^2 (\nabla \eta)^2 + \eta,$$ \quad (1)

where \(\eta(r,t)\) is the height at position \(r\) and time \(t\) measured perpendicularly to a \(d\)-dimensional substrate, \(\eta(r,t)\) is a Gaussian, nonconservative noise given by \(\langle \eta \rangle = 0\) and \(\langle \eta(r,t)\eta(r',t') \rangle = \delta^{d}(r - r')\delta(t - t')\), while \(\nu\) and \(\lambda\) are constants. This equation was independently proposed by Villain \textsuperscript{4} and by Lai and Das Sarma \textsuperscript{5}, being also known as the Villain-Lai-Das Sarma (VLDS) equation and it is a standard model of the nonlinear molecular beam epitaxy (nMBE) universality class. This equation has been investigated in the framework of renormalization group \textsuperscript{8,10} and many features observed in kinetic Monte Carlo simulations have been elucidated.

MBE is often modeled by microscopic transition rules built to capture the atomistic mechanisms. The basic approach is to use stochastic transition rates for atomistic processes such as deposition and thermally activated adatom hopping \textsuperscript{11,12}. A fundamental example in this class is the Clarke-Vvedensky (CV) model \textsuperscript{13}, in which deposition occurs at a constant and uniform rate and the adatom diffusion rate is given by an Arrhenius law in the form \(D = \nu_0 \exp(-E/k_B T)\) where \(\nu_0\) is an attempt frequency, \(k_B\) the Boltzmann constant, and \(E\) is an energy barrier for the hopping of an adatom with near-estimate neighbors (bonds). The activation barrier includes the contribution of the substrate \((E_S)\) and bonds in the same layer \((E_N)\) assuming the form \(E = E_S + nE_N\). Renormalization studies \textsuperscript{8,10} point out that the CV model belongs to the nMBE class. The presence of step barriers \textsuperscript{1} in the CV model, in which the diffusion between different atomic layers is depleted, asymptotically leads to mounded morphologies with non self-affine structure \textsuperscript{12,14}.

Some investigations of the dynamic scaling of the surface roughness in CV-type models have been reported \textsuperscript{15,19}. The interface fluctuation within a window of length \(l\) (hereafter called quadratic local roughness) is defined as

$$\omega^2(l, t) = \langle h^2 \rangle_i - \langle h \rangle_i^2,$$ \quad (2)

where \(\langle \cdot \rangle_i\) means averages over the window \(i\). The quadratic local interface roughness \(\omega^2(l, t)\) is defined as the average of \(\omega^2\) over different windows and samples. In self-affine dynamical scaling, the local roughness increases as \(\omega \sim t^\alpha\) for \(t \ll l^z\) and saturates as \(\omega \sim l^\nu\).
for \( t \gg L \), with \( z = \alpha/\beta \), and these scaling exponents are called growth (\( \beta \)), roughness (\( \alpha \)) and dynamic (\( z \)) exponents\(^3\), respectively. If \( l \) is chosen to be the system size \( L \), Eq. (2) yields the squared global roughness. Former works on the CV model suggest temperature dependent exponents and anomalous scaling of the surface roughness\(^{15,17}\). Recently, the local roughness of the CV model in \( d = 2 \) has been analyzed and a transient anomalous (non self-affine) scaling has been found\(^{19}\). As a consequence, nMBE asymptotical exponents with large corrections to the scaling were conjectured for the nMBE models\(^{20}\). Consequently, nMBE asymptotical exponents with large corrections to the scaling were conjectured for the nMBE models\(^{20}\).

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\[
\xi_{0,m} \sim t^{1/z_c}
\]

where \( z_c \) is the coarsening exponent that, in case of self-affine growth, corresponds to the dynamical exponent defined previously.

Under the hypothesis of normal (non anomalous\(^{19,28,29}\)) scaling, the local roughness obeys the Family-Vicsek ansatz\(^{30}\) given by

\[
\omega(l, t) \sim t^{\beta} F\left( \frac{l}{l_1/z} \right),
\]

where \( F \) scales as \( F(x) \sim x^\alpha \) for \( x \ll 1 \) and \( F(x) = \text{const} \) for \( x \gg 1 \), leading to \( \omega \sim t^\beta \) for \( t \ll l^c \) and \( \omega \sim t^{\ell^c} \) for \( t \gg L \). For anomalous scaling\(^{31}\), the local roughness follows the modified ansatz

\[
\omega(l, t) \sim t^{\beta} F_{\text{ano}}\left( \frac{l}{l_1/z} \right),
\]

where \( F_{\text{ano}}(x) \sim x^{\alpha_{\text{loc}}} \) if \( x \ll 1 \) and \( F_{\text{ano}}(x) = \text{const} \) for \( x \gg 1 \). Note that if \( \alpha \neq \alpha_{\text{loc}} \), one has \( \omega(l, t) \sim l^{\alpha_{\text{loc}}-\kappa} \), where \( \kappa = (\alpha - \alpha_{\text{loc}})/z \), for short scales. Therefore, for anomalous scaling, the amplitude of \( \omega \) vs \( l \) scales as \( t^\kappa \), where \( \kappa \) is the anomaly exponent\(^{19,28,29}\).

II. METHODS

A. Dynamical scaling

We will consider the surface evolution in the growth regime where the global roughness scale as \( W = \omega(L, t) \sim t^{\beta} \). A characteristic surface length can be extracted from the autocorrelation function defined as

\[
\Gamma(r, t) = \langle \tilde{h}(r_0 + r, t) \tilde{h}(r, t) \rangle,
\]

where \( \tilde{h} = h - \bar{h} \), \( \bar{h} \) is the mean height of profile, and averages in Eq. (3) are performed over different reference positions \( r_0 \), different orientations and independent samples. Surface growth under nonequilibrium conditions may present mounded morphologies\(^{2,12,27}\). In mounded surfaces, the characteristic lateral surface length can be estimated as the first zero \( (\xi_0) \) or the first minimum \( (\xi_m) \) of autocorrelation function\(^{14}\). Those lengths are expected to scale as

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where \( F_{\text{ano}}(x) \sim x^{\alpha_{\text{loc}}} \) if \( x \ll 1 \) and \( F_{\text{ano}}(x) = \text{const} \) for \( x \gg 1 \). Note that if \( \alpha \neq \alpha_{\text{loc}} \), one has \( \omega(l, t) \sim l^{\alpha_{\text{loc}}-\kappa} \), where \( \kappa = (\alpha - \alpha_{\text{loc}})/z \), for short scales. Therefore, for anomalous scaling, the amplitude of \( \omega \) vs \( l \) scales as \( t^\kappa \), where \( \kappa \) is the anomaly exponent\(^{19,28,29}\).

B. Optimal DFA

Let us consider the standard DFA method using a \( n \)th order polynomial to detrend the surface\(^{24}\), called here of DFA\(_n\). For sake of simplicity, we consider one-dimensional cross sections for two-dimensional surfaces. The interface fluctuation within a window \( i \) of size \( l \) in DFA\(_n\) is defined as

\[
\omega_i^{(n)} = \langle (\delta_i^{(n)})^2 \rangle_i^{1/2}
\]
corresponds exactly to DFA\(_0\) but for higher orders they are different. In particular, we can easily verify that \(\delta^{(1)} = [h(x) - G_i] \cos(\theta_i)\) is the minimal distance to the detrending curve. Figure 1 shows a schematic representation for DFA\(_1\) and ODFA\(_1\). The variable \(\theta_i\) corresponds to the angle between substrate and the local trend \(\theta_i = \arctan(A_i^{(1)})\). Observe that for \(l \gg 1\), both DFA and ODFA correspond to the global roughness given by Eq. (2) with \(l = L\), since the whole surface is not trended. Figure 2 shows the probability that a window of size \(l\) has slope \(\theta\) in a simulation of the CV model (see Sec. IV for simulation details). The distributions are Gaussian and converge to Dirac delta functions centered at \(\theta = 0\) as \(l\) increases. In higher order ODFA, the minimal distance can be computed numerically using root finding algorithms as the bisection method [32] used in the present work.

III. VALIDATING THE ODFA METHOD

In order to validate the ODFA method and compare it with DFA, we consider the deposition on an initially mounded one-dimensional surface shown in Fig. 3(a). The surface evolves according to the etching model rules [20] in \(d = 1\) that belongs to the KPZ universality class [25]. This model has a large roughness and also strong corrections to the scaling [33, 34], characteristics that make it suitable to perform the tests. The model rules are as follows: at each time step, a site \(x\) is randomly chosen and all nearest-neighbors of \(x\) that obey \(h(y) < h(x)\) are increased as \(h(y) \rightarrow h(x)\) and subsequently \(h(x) \rightarrow h(x) + 1\). The surface preserves its initially mounded structure (\(\xi_0 \approx 1000\)) in the whole interval of simulation considered, as shown by the correlation function in the inset of Fig. 3(a).

Figure 3(b) compares the effective exponent analyses for the local interface roughness using non detrended analysis (DFA\(_0\)), DFA and ODFA up to second order. The effective roughness exponent \(\alpha_{\text{eff}}\) is defined as \(\alpha_{\text{eff}} \equiv d \ln(\omega^{(n)})/d \ln(l)\). DFA\(_0\) leads to a large exponent, typical of mounded surfaces because it is dominated by the long wavelength scales and does not capture the local fluctuations. The DFA\(_1\) analysis provides a plateau at \(\alpha_{\text{eff}} = 0.44\) below the exactly known value \(\alpha_{\text{KPZ}} = 1/2\) [25]. Using DFA\(_2\), we observe an increasing of the plateau but no significant improvement of the exponent value is found. For ODFA method however, we observe a plateau at \(\alpha = 0.49\) very close to the KPZ exponent 1/2 with a larger plateau for ODFA\(_2\). We stress that, in scales larger than the average mound length, the values of the effective roughness exponent reflect the geometrical aspects of the mounded surface, i.e. \(\alpha_{\text{eff}} > 0.5\) is expected.
IV. RESULTS FOR CV MODEL IN $d=2$

We performed simulations of the CV model in a simple cubic lattice, with an initially flat substrate at $h = 0$ of lateral size $L$. Periodic boundaries conditions along the substrate directions are considered. Deposition occurs with a flux normal to the substrate of $F$ atoms per site per unit of time under a solid-on-solid condition. The ratio

$$R = \frac{D_0}{F},$$

in which $D = D_0 \epsilon^a$ is the hopping rate if an adatom has $a$ lateral neighbors, is a control parameter of the model. Here, $D_0 = \nu_0 \exp (-E_N/k_B T)$ is the hopping rate of an adatom with no lateral bond, and $\epsilon = \exp (-E_N/k_B T)$. One time unit corresponds to the deposition of $L^2$ adatoms, fixing $F = 1$ without loss of generality. In a deposition event, the atom is adsorbed on the top of the previously deposited adatom or the substrate site to grant the solid-on-solid condition. For the same reason, only adatoms at the top of the columns are mobile and perform random hopping towards the top of their nearest neighbor sites.

![FIG. 3. (Color online) (a) Mounded initial condition and the (shifted) profile after a deposition time $t = 10^3$ using the Etching model. The corresponding correlation functions are shown in the inset. (b) Effective local roughness exponent as a function of the window size for a deposition time $t = 10^3$ using different methods. The horizontal dashed line indicates the value of the KPZ roughness exponent $\alpha_{KPZ} = 1/2$.](image)

![FIG. 4. (Color online) (a) Typical morphology of a surface generated with the CV model, at deposition time $t = 10^5$, showing the formation of mounds. (b) Scaled autocorrelation function $\Gamma(r, t)/\Gamma(0)$ versus distance $r$. The inset shows the time evolution of the characteristic length given by the minimum of the autocorrelation function. The dashed line has a slope 0.30.](image)
the correlation length in this regime.

FIG. 5. (Color online) (a) Local roughness using different methods as a function of the window size for different times. (b) Effective local roughness exponent $[\alpha_{\text{eff}} \equiv d\ln(\omega^{(t)})/d\ln(l)]$ analysis for different methods for a deposition time $t = 10^6$. The horizontal dashed line indicates the value of the nMBE roughness exponent $\alpha_{\text{nMBE}} = 2/3$. The inset shows the time evolution of the global roughness obtained with ODFA1 and the dashed line has slope 0.20.

Figure 5(a) compares the local roughnesses using DFA and ODFA methods at different times. In Fig. 5(b), the effective roughness exponent is shown as a function of $l$ for $t = 10^6$. The analysis for ODFA1 provides a plateau for $\alpha_{\text{eff}} \approx 2/3$ for the range $25 \leq l \leq 60$. A larger plateau is observed for ODFA2. We can see, for the time intervals considered, that DFA up second order provides estimates of the roughness exponent bellow the value expected for the nMBE class. Only the sizes of the plateaus are increased for DFA2 similarly to the behavior observed for the etching model in Fig. 3.

From Fig. 4(b), we obtain that the average size of the mounds at $t = 10^6$ is $\xi_m \approx 75$. Therefore, ODFA methods indicate that nMBE roughness exponent can be extracted from the CV model considering fluctuations with optimal (minimal) distance to the local trending within scales up to the same order of the mound size. This result is particularly useful and raises the possibility of measuring the roughness exponent for experimentally accessible times. The inset of Fig. 5(b) shows the global roughness against time computed using ODFA1 (similar curves were obtained for DFA and ODFA2) for $l = L$. The results provide $\omega \sim t^{0.2}$ for $t \gtrsim 10^3$, which is fully consistent with the nMBE growth exponent.

Now, we address the transient anomalous scaling of the CV model. In self-affine (non anomalous) scaling, the local roughness at very short scales (of order of just a few lattice spaces) approaches a finite value at long times following a power-law correction given by

$$\omega^{(t)} = C_1 + C_2 t^{-y},$$

where $C_1$ and $C_2$ are constants. This is the same behavior of the average squared local slope $\langle (\nabla h)^2 \rangle$ [20]. For the Edwards-Wilkinson (EW) [25] and KPZ [25] equations, the values of the exponent $y$ were computed analytically as $y_{\text{EW}} = d/2$ and $y_{\text{KPZ}} = 2(1-\alpha)/z$, respectively [36][37]. In Ref. [19], it was shown that the effective anomaly exponent in the range $0.08 \leq \kappa \leq 0.23$ observed for the CV model is due to a transient effect since the local roughness for a very small scale approaches a constant value according Eq. (11) with $y = 0.09$.

The local roughness for a window of size $l = 5$ is shown as a function of $t^{-y}$ in Fig. 6 for DFA0, DFA1 and ODFA1, considering $y = 0.09, 0.12$ and 0.23, respectively, showing that the convergence to a constant value is quicker for ODFA1. This is an additional evidence that CV model exhibits asymptotically normal scaling, corroborating conjecture of Ref. [20] where other models of the nMBE class were investigated. It is worth noticing that the exponent $y = 0.23$ found with ODFA1 is the same found in the conserved restricted solid-on-solid (CRSOS) model [20], where scaling corrections are weaker.

Finally, we notice that ODFA$$_n$$, with $n \geq 1$, also works for self-affine surfaces and improved results were obtained as compared with DFA$$_0$$. However, no significant differences were observed as compared with DFA$$_n$$. Therefore, we propose that using ODFA in experimental and computational procedures is indicated since it is equivalent
to standard DFA in self-affine growth but captures better the nature of the fluctuations of mounded surfaces.

V. CONCLUSIONS

In this work, we investigated a detrended fluctuation analysis, in which the height fluctuations are taken with respect to the optimal (minimal) distance from the detrending curve. This modification was observed to be irrelevant for the determination of the roughness exponents of purely self-affine surfaces but it matters for systems with transient mounded behavior, which encompass models belonging to the important universality class of non-linear molecular beam epitaxy. The method was validated and compared with standard DFA analysis using a one-dimensional growth model with a well-known roughness exponent.

We applied the method to the Clarke-Vvedensky model where deposition competes with thermally activated surface diffusion producing interfaces with rough mounds. Since this model possesses the central mechanisms of the nMBE class, one expects that it exhibits the nMBE exponents. We compared the present method with non detrended and standard DFA analyses for the CV model at low temperature and long times. A roughness exponent in agreement with the nMBE universality class ($\alpha_{\text{nMBE}} = 2/3$) was observed only for ODFA. We also investigated the transient anomalous scaling, in which the local roughness within small windows converges to a constant value with a power law correction in time, Eq. (11), and found that the ODFA method yields the same exponent $y = 0.23$ observed for other nMBE models with weaker corrections to the scaling $[20]$. Since this exponent is universal for other universality classes, namely EW and KPZ $[39, 37]$, our results point that the same holds for the nMBE class.

The reason why ODFA method is more efficient than DFA is intuitively simple since the natural distance to a deterministic referential is the minimal one as illustrated in Fig. [1]. The larger distances to the detrending curve in DFA introduces subleading corrections that are relevant in experimentally and computationally accessible times and sizes, for which the surfaces actually does not reach its asymptotic dynamical behavior.

We expect that our results will be of relevance for experimental analyses, in which mounded morphologies are commonly observed $[1]$ and the resolution of the universality class is challenging. As a perspective, it would be interesting to consider the role played by the intrinsic smoothening of the surfaces obtained by the widely used scanning probe microscopy techniques that can markedly interfere in the roughness exponent determination $[38]$ or even indicate a misleading universality class $[39]$.

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