Nonlinear measures for characterizing rough surface morphologies

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We develop an approach for characterizing the morphology of rough surfaces based on the analysis of the scaling properties of contour loops, i.e., loops of constant height. Given a height profile of the surface we perform independent measurements of the fractal dimension of contour loops, and the exponent that characterizes their size distribution. Scaling formulas are derived, and used to relate these two geometrical exponents to the roughness exponent of a self-affine surface, thus providing independent measurements of this important quantity. Furthermore, we define the scale-dependent curvature, and demonstrate that by measuring its third moment departures from Gaussian behavior can be ascertained. These nonlinear measures are used to characterize the morphology of computer generated Gaussian rough surfaces, surfaces obtained in numerical simulations of a simple growth model, and surfaces observed by scanning-tunneling microscopes. For experimentally realized surfaces the self-affine scaling is cut off by a correlation length, and we generalize our theory of contour loops to take this into account.

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1. INTRODUCTION

Random surfaces are widely used in the physical sciences to model phenomena ranging from the extremely small (quantum gravity) to the very large (Earth’s relief). They describe crack fronts in materials science [1], ripple-wave turbulence [2], passive tracers in two-dimensional fluid flows [3,4], cloud parameters [5,6], and shapes of stomatolites (conjectured fossil ancestors of ancient bacteria) [7], to mention but a few recent examples. Our focus in this paper is on the morphology of deposited metal films, which develop random self-affine surfaces quite different non..equilibrium growth conditions, as indicated by theoretical, numerical, and experimental results over the past decade [8,9].

Surface configurations are parametrized by a two-dimensional field h(x) which represents the height of the surface above a reference plane (x). Theoretical dynamics of a growing surface are described by a continuum (Langevin) equation giving dh(x)/dt as a sum of a Gaussian white noise term, to mimic the random deposition of atoms and a polynomial of various gradients of h(x), to model relaxation processes on a coarse grained scale. The nonequilibrium growth behavior is due to the interplay of the deposition and relaxation terms.

One would guess that even a snapshot of the morphology should carry evidence of the nonequilibrium, nonlinear growth process which produced it, and should measure

from surfaces produced in equilibrium or by a linear process, even if they share the same scaling exponents. This motivates a search for roughness measures independent of the growth model (for fixed roughness height), which might identify important distinctions between different surface models that have similar spatial power spectra. Such measures, although motivated here in the context of self-affine or multifractal surfaces, should be handy even for surfaces showing no self-affine regime. They can quantify features of morphology which are presently characterized by eye, which should permit a more systematic comparison between observations and models than at present. One can imagine that, armed with two or three kinds of roughness measures tuned to different qualitative aspects of the surfaces morphology, one could construct empirical “phase diagrams” in this two- or three-dimensional parameter space, e.g., mapping our domains in the parameter space that correspond to various growth conditions.

Given the surface h(x) parametrized by an array of heights—obtained, e.g., from a simulation or a scanning-tunneling-microscope (STM) experiment [10]—we ask in what different ways can the surfacе morphology be characterized? In general, one requires more than one characterization to confirm a match between experimental and simulation data, or to convincingly verify self-affineness. For the applied problems of growing flat surfaces (e.g., for semiconductor devices) or regularly modulated ones (e.g., for nanofabrication arrays of quantum dots), it is also desirable to develop independent measures that quantify different aspects of a rough surface’s geometry. In this paper, we propose two categories of measure for characterizing spatial correlations of rough surfaces. These measures are usable on any kind of rough model, and require no dynamical information, so they should be useful in analyzing not only experiments on solid films, but also the diverse phenomena mentioned above.

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for small values of $|q|$. In the case of a surface with a finite correlation length, $S(q)$ crosses over to a constant value for $|q|<l/H$; this situation is discussed in more detail in Sec. IV.

Clearly, $S(q)$ does not uniquely characterize a self-affine ensemble of surfaces. For example, it is invariant under changes in the dimension by linear addition of Fourier components—we do this in Sec. V—yet the real growth process is typically non-linear, and the surface is non-Gaussian. Indeed, confirmation of the scaling given by Eqs. (2.2) or (2.7) in experiments cannot be interpreted as conclusive evidence for a self-affine geometry, that is a property of the whole ensemble, and so requires proper scaling of all moments and correlations, not just the second moment.

**Quadratic roughness measures**

The height-correlation function [Eq. (2.2)], is the most standard measure in theoretical discussions, in that “roughness” is defined by the divergence of this function as its argument $r$ approaches infinity. (Nonmonotonic behavior of this function has also been observed to measure the characteristic spatial scale of mounds or other patterns in non-self-affine surfaces.)

On the other hand, the Fourier power spectrum [Eq. (2.4)] is central in theoretical derivations but rarely used in experimental analysis except for Ref. (14). This seems to be the best quadratic measure, in that it most cleanly separates the contributions from fluctuations on different length scales, and it shows the sharpest knee (on a log-log plot) where self-affine scaling is cut off.

Another quadratic measure is the total variance of $h(x)$ in a box of size $b$, as a function of $b$ [15,16],

$$\langle (h(x) - \bar{h})^2 \rangle, \tag{2.8}$$

where $\bar{h} = \langle h(x) \rangle$, and $\langle \cdots \rangle$ means the spatial average is only taken over a square of side $b$ centered on $x_0$. This variance should be averaged over different choices of $x_0$.

**C. Fractal properties**

Self-affine surfaces are fractals only in a generalized sense, since the horizontal direction rescales differently from the vertical direction. On the other hand, the level set of such a surface (defined as its intersection with a horizontal plane) is a fractal object [17], see Fig. 1 below. Different planes of intersection give statistically equivalent level sets, since the height fluctuations of a rough surface are unbounded. Level sets consist of contour loops which are the connected components. We expect these to be fractal as well, with a fractal dimension smaller than the dimension of the whole level set, which is simply the union of all contour loops of the same height. Furthermore, contour loops come in all sizes limited only by the system size, and an exponent can be defined that characterizes their size distribution. Since contour loops are defined for clustering their geometrical exponents are analogous to those defined for percolation clusters.

These measures are associated with geometrical exponents that characterize contour loops on self-affine rough surfaces: the loop correlation exponent, the fractal dimension of a loop, and the length distribution exponent.

**A. Scale-dependent curvature**

The obvious real-space-based nontrivial generalization of the height-correlation function is

$$\langle [h(x+r) - h(x)]^2 \rangle, \tag{3.1}$$

however, this is identically zero on an isotropic surface (and whenever the function is even). To escape this problem, we observe that $h(x+r) - h(x)$ is a sort of first difference at scale $r$, and replace it by a sort of second difference. That is, we define the curvature at a scale $b$ as

$$C_b(x) = \sum_{0<m_2 b^2} [h(x+b_m e_2) - h(x)], \tag{3.2}$$

where the offset directions $e_2$ are a fixed set of vectors summing to zero. In our numerical implementation of this measure, where $x$ is a square lattice, we choose four such offsets related by $90^\circ$ rotations, pointing either along the [10] or [01]-type directions. These two sets of offsets should give equivalent results (for the same $h$), provided the surface is statistically invariant under rotations in the reference plane. We then define curvature moments for integer powers $q$.

The first moment of $C_1$ is manifestly zero; the second moment is linearly related to the height-correlation function:

$$\langle C_1(x)^2 \rangle = \sum_{0<m_2 b^2} D_b(b_m) - \frac{1}{2} \sum_{m_1,m_2} [D_b(b_{m_1} - e_2)], \tag{3.3}$$

where $D_b(b_m) = \frac{1}{2}[h(x+b_m e_2) + h(x+b_m e_2) - h(x+b_m e_2)]$.

This is a natural generalization of the height-correlation function using an absolute value to avoid the trivial cancellation in Eq. (3.1). D. Sarma and co-workers [23,21] used Eq. (3.6) to test for multifractality behavior (whereby the Fig power of the $q$ moment scales with exponent depending on $q$, unlike the simpler self-affine case). For odd $q$, Eq. (3.6) is insensitive to the up-down symmetry (or nonsensical anyhow. Our “curvature” seems to be the simplest function that detects the skew locally.

**B. Fractal dimension of contour loops**

For the remainder of this section, we must define the loop ensemble. Consider a contour plot of a rough surface with a finite spacing $b$ between heights of successive level sets. We take it to be an arbitrary constant much smaller than the typical (rms) fluctuation of $h(x)$. The value of $\Delta$ does not affect our exponents, and we need to consider it explicitly only in the arguments of Sec. IVa; in other places we may implicitly scale height $h(x)$ such that $\Delta = 1$. In STM images of rough metal surfaces $\Delta$ is usually the height of a single step on the surface.

The contour plot closed of cross-intersecting lines in the plane that connect points of equal height, we call contour loops (see Fig. 1). Every random-surface configuration maps to a configuration of contour-loops; when the probability weights of the respective configurations are taken into account, this defines a mapping of the random-surface ensemble to the contour-loop ensemble. The contour loop ensemble arising from self-affine random surfaces (we shall argue) is self-similar; the loops are connected clusters that can be studied using scaling, just as (critical) percolation clusters have been analyzed in previous work [24].

Asymptotically, any contour loop in the loop ensemble we define a loop length $l$ and a loop radius $R$. In all the examples we study the heights are defined on an L x L square lattice with lattice constant $a$. The loop length is measured with a ruler of length $a$ while the loop radius (really a diameter) is defined as the side of the smallest box that completely covers the loop; see Fig. 18.
In the loop ensemble we define a joint distribution \( \tilde{n}(x, R) \) (independent of the contour spacing \( \Delta \)) such that the number of loops with length in \( (s, s + ds) \) and radius in \( (R, R + dR) \), per unit area, is

\[
\Delta^{-1} \tilde{n}(x, R) dR.
\]

The factor \( \Delta^{-1} \) has the obvious significance that if one halves the contour spacing, one has twice as many contours.

Assuming that the loop ensemble is scale invariant, we expect that \( \tilde{n}(x, R) \) has a scaling form

\[
\tilde{n}(x, R) \sim R^{-\eta} f(x/R). \quad (3.8)
\]

Here \( D_f \) is the fractal dimension, and \( \eta \) is simply related to the length distribution exponent \( \tau \), which we define in Sec. III C.

In practice, the exponent \( D_f \) is measured by the scaling relation

\[
\langle x \rangle(R) \sim R^{\eta(D_f-1)/\tau}, \quad (3.9)
\]

where

\[
\langle x \rangle(R) = \frac{\int x \tilde{n}(x, R) ds}{\int \tilde{n}(x, R) ds} \quad (3.10)
\]

is the average loop length for loops with radius \( R \). The scaling in Eq. (3.9) follows immediately from the assumed scaling form in Eq. (3.8).

The dimension defined in Eq. (3.9) is really the scaling dimension of the loop length, i.e., it defines the relation between larger and smaller loops in the distribution. On the other hand, the proper fractal dimension (either the Hausdorff dimension \( D_H \) or the self-similarity dimension) refers to the relation between bigger and smaller pieces of the same loop. Thus \( D_f \) is defined by \( s = a^{-\eta} \), i.e., how the loop length scales with the ruler size. When the contour-loop distribution is self similar (as we shall assume), the two kinds of dimensions are equivalent.

C. Loop length distribution exponent

We define the loop number density \( \tilde{P}(s) \) so that \( \Delta^{-1} \tilde{P}(s) ds \) is the total number of loops, per unit area (measured in sites), with lengths in \( (s, s + ds) \); a related distribution of loop lengths, \( \tilde{P}(s) \), is defined such that \( \Delta^{-1} \tilde{P}(s) ds \) is the number of loops passing through a fixed point (say the origin) with lengths in the range \( (s, s + ds) \). In lattice models (including our numerical examples in Secs. V, VI, and VII), \( s \) is an integer and \( \tilde{P}(s) \) is essentially the probability that the loop has length \( s \).

From comparison to Eq. (3.7) it is obvious that

\[
\tilde{P}(s) = \frac{\int n(x, R) ds}{\tilde{n}(x, R)}, \quad (3.11)
\]

Since the total number of sites along a loop is equal to its length \( s \), we have

\[
P(\text{loop}) = \int_{s=1}^{\infty} \tilde{P}(s) ds = 1. \quad (3.12)
\]

the additional factor of \( s \) is because each site can be the origin of only one definition of \( \tilde{P}(s) \).

Assuming that the loop ensemble is scale invariant we can define the length distribution exponent \( \tau \) by

\[
P(R) \sim R^{1-\tau} \tilde{P}(s) \sim R^{1-\tau} \quad (3.13)
\]

This is to hold for large contour loops, i.e., those of radius much bigger than the microscopic scale \( s \). Indeed, inserting Eq. (3.8) into Eq. (3.11) gives Eq. (3.13), with

\[
y = s + 1/2 D_f \quad (3.14)
\]

On the other hand, we could also define \( \tilde{n}(x, R) \) such that

\[
\Delta^{-1} \tilde{n}(R) dR = s \quad (3.15)
\]

Doing the integral and then eliminating \( y \) using Eq. (3.14) gives

\[
\tilde{n}(x, R) \sim R^{\frac{1}{D_f}} \quad (3.16)
\]

We would have obtained the same result more quickly and (more dubiously) had we assumed a strict relationship between radius and length, \( s \rightarrow R \rightarrow R^{2/5} \), rather than write Eq. (3.8).

D. Loop correlation function

The loop correlation function \( G(\mathbf{r}) \) measures the probability that two points separated by \( \mathbf{r} \) lie on the same contour loop. This correlation function is nonlocal, for the connectedness of the two points depends on every site on the portion of loop between them. This loop correlation function should be distinguished from the level-set correlation function which simply measures the probability that two points separated by \( \mathbf{r} \) are at the same height. For the loop correlation function to be well defined the contour lines are considered to be of finite width given by the microscopic scale \( s \). Due to rotational symmetry of the loop ensemble, \( G(\mathbf{r}) \) depends on \( r = ||\mathbf{r}|| \) only, and for large separations (\( r > a \)) we expect it to fall off as a power law:

\[
G(r) \sim r^{-\delta} \quad (4.1)
\]

This scaling property of the contour ensemble justifies the power law dependence of \( G(\mathbf{r}) \) on \( r \) and \( P(\text{loop}) \) on \( x \), in Eqs. (3.17) and (3.13) respectively.

In writing Eq. (4.1), we made a nontrivial hypothesis that the correlation of the height function obtained by coarse graining a given realization of \( \tilde{h}(\mathbf{x}) \) is statistically the same as the coarse-grained version of the contours of \( \tilde{h}(\mathbf{x}) \). We know of no coarse-graining procedure for the height function which assures that the contours will stay the same. It will happen that, near a saddle point of \( \tilde{h}(\mathbf{x}) \), two loops (both of height \( h_{\text{peak}} \)) approach closely, but the coarse-graining shifts the height of the saddle point across \( h_{\text{peak}} \) so that the coarse-grained versions of the loops coalesce into one loop. Whether this phenomenon makes a relevant contribution to our scaling relations depends on the frequency of close approaches [25].

To determine the scaling of \( \tilde{n}(R) \), first apply the rescaling (2.1) to each configuration of \( \tilde{h}(\mathbf{x}) \); this maps the contour ensemble to a new contour ensemble with a rescaled contour interval \( \Delta_{\text{new}} = \Delta/2^{D_f} \). The total number of contours with radii in the large (\( R, R + dR \)) in the scaled relation is \( \tilde{L}^{D_f} \Delta_s^{-D_f} \tilde{n}(R) dR \), by our definition in Sec. III C. Since the contours are mapped one-to-one [according to the hypothesis implicit in Eq. (4.1)], we can equate this with the number of new contours in a box of side \( L/2 \) and of radius in \((R/2, R + R/2)\), which is \((L/2)^2 \Delta_s^{-D_f} \tilde{n}(R/2) dR/2\). On the other hand, by self-affineness the new height ensemble is statistically identical to the original one (for large \( L \)); this also holds for the new contour ensemble, thus \( \tilde{n}(R) \sim \tilde{n}(R/2) \). We obtain \( \tilde{n}(R/2) = b^{-2} \tilde{n}(R) \), which implies the scaling

\[
\tilde{n}(R) \sim R^{-3+\alpha} \quad (4.2)
\]

Equating Eqs. (4.2) and (3.16) leads to the first scaling relation (called “hyperscaling”)

\[
D_f (1-\tau) = 1 - \alpha. \quad (4.3)
\]

This scaling relation was derived previously by Huber et al. [26] in a slightly different context, and in a somewhat different form by Iuchi and Kalia [27]. Unlike the usual hyperscaling relation for percolation clusters which can be derived from the assumption that the number of clusters does not grow with scale of observation, here that number grows as a power with exponent \( \alpha \).
due to Mandelbrot [17]. The important difference is that Eq. (4.10) gives the fractal dimension $D$ of the level set of a random self-affine surface, and not the fractal dimension of a single contour loop. We emphasize this point because there has been some confusion in the literature where the two dimensions have been equated.

Observe [39] considered the same loop ensemble, but mostly focused their attention on the "islands" contained in the loops rather than the contours; their \( \gamma \) exponent (which we call \( \tau_{\gamma} \)) refers to the distribution of island sizes. They derived a formula \( \tau_{\gamma} = 2 - a^{2} \) (in our notation). It is easy to show that \( 2\tau_{\gamma} - \beta = -D + 1 \), the \( \gamma = 2 \) here is the fractal dimension of these islands [30]; upon inserting this conversion, their formula turns out to say $D = 2 - \alpha$, which is the same as our Eq. (4.10).

C. Loop correlation exponent

Now we turn our attention to the contour correlation exponent, and we conjecture that

$$\gamma = 1/2$$

is superuniversal in that it is independent of \( \alpha \). In the case of a \( \alpha = 0 \) Gaussian surface, we know \( \gamma = 1/2 \) exactly for a solvable statistical-mechanics model of contour loops, equivalent to the critical (\( D/2 \)) loop model on the honeycomb lattice [31]. Details are in Appendix A. By invoking universality this is valid for all logistically rough random Gaussian surfaces.

The exact value of \( \gamma \) can also be determined for \( \alpha = 1 \). That is, the fractal dimension \( (Df) \) of a contour loop must satisfy $D = 2 - \alpha$ since it is a subset of the level set, which has dimension $D = 2 - \alpha + 1$ [Eq. (4.11)]. On the other hand, \( Df = 1 \), since a loop has topological dimension 1. From these inequalities we conclude that for \( \alpha = 1 \) the fractal dimension of a contour loop is $D = 1$. This in turn leads to \( \gamma = 1/2 \), from Eq. (4.9).

The validity of conjecture (4.12) for general \( \alpha \) has been checked, to date, only through the numerical simulations reported in Sec. V and in numerical simulations of Zeng et al. [32].

Since \( \gamma = 1/2 \) for \( \alpha = 0 \) and 1, a proof of the monotonicity of \( \gamma \) with \( \alpha \) would suffice to establish the conjecture. Even that is very difficult owing to the nonlocal definition of the loop correlation function.

D. Combined scaling relations

Equipped with the (superuniversal) conjectured value of the loop exponent \( \gamma = 1/2 \), and the scaling relations, (4.9) and (4.10), we find the following formulas for the geometrical exponents of contour loops of a self-affine surface with roughness exponent \( \alpha \):

These relations form the basis of the contour loop analysis of rough surfaces, which implement in the following sections.

Our formula for $D_{f}$ differs from the one proposed by Ishchenko [33].

The first scaling relation is reminiscent of the relation

$$D = 2 - \alpha$$

$$\gamma = 1/2$$

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These relations form the basis of the contour loop analysis of rough surfaces, which implement in the following sections.
R for random Gaussian surfaces with \( a = 0.4, 0.8 \) from bottom to top; system size \( L = 512 \) and \( 10^3 \) loops were collected. The "direct" \( D_r \) data in Table I are obtained by linear least-squares fits to such plots in the scaling regime, which is roughly \( 10^{-5} < R < 100 \).

C. Loop measurements

The primary motivation for our Gaussian surface simulations was an initial test of the scaling predictions for the contour loop exponents from Sec. IV. A contour plot of a sample surface configuration for \( a = 0.4 \) is shown in Fig. 1. (A similar plot for \( a = 0.0 \) was published in Ref. [19].)

1. Measurement procedure

In a single run, which would typically take 10 min on a Sun Sparc workstation, 25 surfaces of specified roughness \( a \) were generated. For each surface typically 400 points were chosen at random, and through each point a contour loop was constructed using the loop finding algorithm as explained in Appendix C. While each loop was being traced along the loop we used to evaluate the loop correlation function \( G(r) \). For each contour loop its radius and length were measured and used to determine the length distribution of contour loops \( P(x) \), and the average loop length \( \langle x \rangle \) as a function of the loop radius \( R \).

2. Results

In order to measure the geometrical exponents \( D_r \), \( \tau \), and \( x_1 \), we plotted the data for system size \( L = 512 \) on a log-log graph and performed least-squares linear fits. Data were selected for fitting from the range in which a well developed power law was observed; see Figs. 4, 5, and 6. The results are given in Table I. We find excellent agreement between the predictions of the scaling theory and the measured geometrical exponents. In particular, note that the simulations confirm the super-universal nature of the loop correlation exponent \( x_1 \).

The loop correlation function \( G(r) \) has a size dependence which biases a direct fit to the exponent \( x_1 \); finite-size scaling (see below) partially overcomes this systematic error. Our theory (Sec. IV C) indicates that \( G(r) \) has a universal exponent \( 2x_1/\tau \); in fact, as shown in Fig. 6, \( G(r) \) itself appears practically independent of \( a \). Closer examination reveals that the coefficient in \( G(r) \sim 1/\nu \) decreases slightly as \( a \) grows. Furthermore, the fitted values of \( 2x_1 \) (see Table I) decrease a bit with \( a \), which we attribute to the systematic error just mentioned, combined with the small \( a \) dependence of the shape of the "knee" in the finite-size behavior of Fig. 8. There is no indication in the extracted \( 2x_1 \) values of any nonmonotonic dependence on \( a \); as shown in Sec. IV C, monotonicity of \( x(a) \) is sufficient to prove \( 2x_1 = \text{constant} \), independent of \( a \).

A better measure of the geometrical exponents was obtained from a finite-size scaling analysis of the data. Using the scaling forms in Eq. (1.5), we produced data collapses ("scaling plots") Sample data for the \( a = 0.4 \) case are given in Figs. 7(a) and 8(a); the data collapse is shown in Figs. 7(b) and 8(b). From the loop-size distribution plots like Fig. 7(b) we extracted both the exponents \( \nu \) and \( ? \). Similarly, we did not carry out finite-size scaling of the \( x \) versus \( R \) plots such as Fig. 4, since there was no obvious change in the slope as a function of \( R/L \). The geometrical exponents giving the best data collapses are reported in the "FSS" columns of Table I (see Fig. 9). The reported uncertainties were estimated by the interval over which changes in the exponent value did not visibly worsen the data collapse.

D. Curvature measurements

We measured moments \( \langle C^n \rangle \) \((n = 2, 3, 4) \) of the scale-dependent curvature, as defined in Sec. III A, for Gaussian surfaces generated by the Fourier method described above. These data (in Fig. 3) are a kind of check on the Fourier method, since the mean over an infinite number of samples can be computed analytically.

Self-affine scaling is evident on the log-log plot of the even moments in Fig. 3 (upper plot). The roughness exponent \( 2a \) is obtained as the slope of a straight-line fit to the \( C_2 \) plot. Ideally, the slopes of the \( C_2 \) and \( C_4 \) log-log plots should be \( 2a \) and \( 4a \), with exactly the input \( a \) values used in constructing the random surfaces. This is spoiled somewhat in practice due to discrete-lattice effects for \( a > 3 \) and by finite-size effects when \( h > 4 \). Furthermore, \( \langle C_4 \rangle/(\langle C_2 \rangle)^2 \) should be exactly 3 for every \( b \) value, even those for which the power-law dependence on \( h \) fails; this is true for any Gaussian random variable. Indeed, the measured ratio is close to 3. The third moment of \( C_3 \) is shown in Fig. 3. Independent of \( a \), \( \langle C_3 \rangle \) is roughly zero, as expected for a random Gaussian surface which possesses a \( h \rightarrow -h \) symmetry (i.e., the valley bottoms and the hill tops are equivalent for a Gaussian surface).

Note in Table I how the finite-size scaling exponents agree better with the scaling theory of Sec. IV than the exponents obtained from "direct" fitting of the data to power laws. The discrepancy becomes more obvious at larger values of \( a \). We infer from this that Gaussian surfaces with a large value of the roughness have more pronounced finite-size effects which lead to an overestimate of \( D_r \) and \( \tau \). This is of relevance to experimental data where the system size is typically not a tunable parameter, and the geometrical exponents are necessarily measured using the direct-fit method.

3. Relation to a previous simulation

Numerical measurements of the fractal dimension of contour loops have been done by Avellaneda et al. [37]. They found \( D_r = 1.28 \pm 0.015 \) for an \( a = 0.5 \) surface, which is close to the predicted value \( D_r = 1.25 \), from Eq. (4.13). They also measured the combination \( D_r (2 - \nu) \) which describes the scaling of the probability that a loop passing through a fixed point has a radius larger than \( \rho \), with \( \rho \). We evaluate this quantity by integrating \( n(x, R) = \mathcal{L}(x, R) \), from Eq. (3.8), over all \( x \) and for \( R > \rho \). The numerical result they quote, \( D_r (2 - \nu) = 0.21 \pm 0.017 \), is in fair agreement with our prediction \( D_r = 1.28 \pm 0.015 \) for \( a = 0.5 \), which follows from Eq. (4.13).

Wagner et al. [36] simulated a form of invasion percolation where the threshold pressures have the form of a self-affine surface. Hence the perimeters of the invaded clusters are the same as the contour lines of the surface. They claimed that the perimeter dimension is "consistent" with...
TABLE I  Geometrical exponents $\gamma$, $\beta$, and $\tau$ for loops on Gaussian surfaces with various roughness exponents $\alpha$. Columns marked “direct” are from direct fits to a power law of the data from system size $L=512$, inferred from $D_L$, and $r=2$ from plots such as Figs. 6, 4, and 5. Columns marked “FSS” were fitted to finite-size scaling plots like Figs. 8(b) and 7(b). According to our conjecture, the “theory” value of $\gamma$ is 1, independent of $\alpha$, and this is supported by the measurements here. Notice a slight systematic deviation of the run expectations from theory when $\alpha=0.5$, which we attribute to more severe finite size effects in those cases. The “theory” formulas for $D_L$ and $r=2$ are in Eq. (4.13).

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<td>0.98(2)</td>
<td>1.33(2)</td>
<td>1.32(3)</td>
<td>1.3</td>
<td>0.24(1)</td>
<td>0.225(5)</td>
</tr>
<tr>
<td>0.6</td>
<td>1.00(1)</td>
<td>0.97(2)</td>
<td>1.23(3)</td>
<td>1.19(3)</td>
<td>1.2</td>
<td>0.18(1)</td>
<td>0.165(5)</td>
</tr>
<tr>
<td>0.8</td>
<td>0.97(2)</td>
<td>0.97(2)</td>
<td>1.15(1)</td>
<td>1.11(2)</td>
<td>1.1</td>
<td>0.12(1)</td>
<td>0.112(0)</td>
</tr>
<tr>
<td>1.0</td>
<td>0.95(1)</td>
<td>0.96(2)</td>
<td>1.06(2)</td>
<td>1.04(3)</td>
<td>1.0</td>
<td>0.08(2)</td>
<td>0.022(0)</td>
</tr>
</tbody>
</table>

Ishchenko’s formula [Eq. (4.14)], but did not quote an error; perhaps their precision was such that the prediction of Eq. (4.14) could not have been distinguished numerically from the one we believe to be correct [Eq. (4.13)].

Reference [36] also mentioned measuring a behavior $\tau = \gamma$ with $\gamma = 0.9$ for the correlation between successive filled sites. If this were simply a correlation of two randomly chosen points along a perimeter, it would be identical to our loop correlation functions $G(r)$ or $G_s(r)$ defined in Eqs. (3.17) or (4.4); in fact, the filling process would appear to depend on correlations of the surface gradient, and might have a somewhat different exponent.

D. Surfaces with a finite correlation length

To test the percolation analysis of self-affine rough surfaces with a cutoff, as derived in Appendix B and summarized in Sec. IV, we performed curvature and loop measurements on Gaussian surfaces with a correlation length $\xi_c$. The correlation length is incorporated into the Fourier method of generating Gaussian surfaces by changing the variance of $R(q)$ in Eq. (5.1) to

$$\langle |\Delta q|^2 \rangle = \frac{|q|^{2+1+\alpha}}{(\pi \xi_c^2)^{2+1+\alpha}} \text{ for } |q| > \xi_c. \quad (5.2)$$

The effects of the cutoff are summarized in Fig. 10, which should be compared to the theoretical prediction of Fig. 2. The curvature and loop data shown in the figure are for system size $L=512$.

The second moment of the curvature displays self-affine scaling with roughness $\alpha=0.4$ up to a length scale set by $\xi_c$, and beyond this scale it levels off; see Fig. 10(a). We checked that the third moment of the curvature vanishes, as expected since the height fluctuations are still Gaussian, while the fourth moment follows affine scaling up to roughly the same correlation length as the second moment. The loop measures exhibit distinct crossover behavior, as seen in figures Figs. 10(b)–10(d). For loops whose radius is smaller than the correlation length, which is here $\xi_c=20$, we find values of the geometrical exponents consistent with those extracted previously for $\alpha=0.4$ random Gaussian surfaces.

For loops whose linear size exceeds the cutoff, a scaling consistent with the percolation analysis is found. The actual numerical values extracted by fitting the 20-8=200 data to a power law are somewhat larger than expected $[2\xi_c=1.46(6), D_L=1.76(1), \text{and } \tau=2.63(1)]$, which we attribute to finite-size and/or crossover effects. To check this we also simulated a Gaussian surface with completely uncorrelated heights, i.e., with $\xi=1$ and system size $L=512$, for which we find (by the direct-fit method)

$$2\xi_c=2.16(3), D_L=1.70(2), \tau=2.56(5)H, \quad (5.3)$$

in good agreement with Eq. (4.16).

VI. SIMULATION: NONEQUILIBRIUM GROWTH MODEL

In this section, the linear and nonlinear roughness measures of Sec. III, which in Sec. V were tested on artificial Gaussian random surfaces, are now applied to growth-roughened surfaces produced by a simple random deposition model, the well-known “single-step model.” Our results are in support of the view that the single-step model produces self-affine morphologies.

A. Single-step model

We implemented the “single-step model” [38–40] in $d = 2+1$ dimensions [41]. More details on this model are found in Sec. IIIE of Ref. [38], or Sec. II.A of Ref. [40]. There is one control parameter $p_s$. The allowed configurations are just those of the body centered solid-on-solid (BCSOS) model; each site of a square lattice has an integer-valued height and neighboring heights must differ by $\pm 1$. The Monte Carlo rule is that in each time step a deposition event occurs with probability $p_s$, or an evaporation event (inverse of a deposition event) occurs with probability $1-p_s$; once it is decided which type of event occurs, a site is picked at random among those sites at which that event is allowed [42].

We begin by an overview of the theoretical expectations. Up-down symmetry switches $p_s \rightarrow 1-p_s$; thus we need only report data for $0 < p_s < 0.5$. The case $p_s=0.5$ is special as the dynamics satisfies detailed balance. This should produce an equilibrium-rough surface, namely, the BCSOS model with all configurations weighted equally [43]. This interface, at long wavelengths, is described by the Gaussian model of Sec. V with $\alpha=0$ (Edwards-Wilkinson behavior).

On the other hand, the growth model for $p_s \neq 1/2$ is believed to asymptotically belong to the Kardar-Parisi-Zhang (KPZ) universality class [38,40]. It has been proposed that $\alpha=0.4$ exactly for the $(2+1)$-dimensional KPZ model [44], however, finite-size effects, small simulations, and naive fits systematically underestimate it as $\alpha \approx 0.38$ [45,46]. The KPZ behavior should be clearcut when $p_s$ is close to 1, but otherwise a crossover from initially Gaussian to asymptotic KPZ behavior is expected, which will be slow (as a function of time or system size) if $p_s$ is close to 1/2.

It turns out, in our numerical results (below), that $p_s = 0.5$ indeed shows Gaussian behavior and $p_s=0$ shows KPZ-like behavior, but $\alpha=0.3$ consistently resembles $\alpha=0.5$, at the sizes we could simulate (i.e., up to $L=128$). We attribute this to the above-mentioned crossover from initial Gaussian behavior.

B. Simulations

Starting from a flat surface, we ran the simulation (for systems of $128 \times 128$ sites) for 2000 Monte Carlo steps (MCS) per site to equilibrate ($p_s \neq 0.5$ “equilibrated” really means “reaches the steady-state ensemble”) and then took data for a period of 1200 MCS/site; one such run took 10–15 hours of CPU time on a RISC-6000 workstation. The standard-length runs (for size $L=128$) appeared to be insufficiently equilibrated for $p_s=0.1$, since they failed to collapse on finite-size-scaling plots with smaller systems. Therefore we performed one run for $L=128$, $p_s=0.1$ with 12000 MCS/site for data collection; this is the run reported in our results. In all other cases, we believe the run time was adequate, since much shorter runs showed no gross differences. We performed about four runs for each value $p_s \equiv 0.1, 0.3, 0.5$, verifying the symmetry $p_s \rightarrow 1-p_s$. [All measures are the same, apart from a change in sign of $\langle \delta h^2 \rangle$.] Only one of the $4$ runs was selected to be timed and plotted here; the data sets presented, e.g., $p_s=0.1$ are actually $p_s=0.9$ in some cases. Once every 100 MCS/site (during the data-collecting portion of a run), we performed a measurement step on the
are, respectively, we implicitly assumed and values from \(D_f\) is equivalent to having \(s_a\) satisfy Eq. (4.8) with \(s_a = 1/2\), which can be verified explicitly from the entries in Table II.

**VII. ANALYSIS OF EXPERIMENTAL DATA**

In this section we test our nonlinear measures against experimental scanning tunneling microscopy data sets. Rough metal surfaces grown under several different conditions are believed to develop a morphology with self-affine scaling, but only up to a time-dependent correlation length \(\xi(t)\) as discussed in Sec. IV E.

The most detailed analysis was done for the vapor deposited Ag surface on a quartz substrate of Palasantzas and Krim [47]. We obtained a 400x400 height array corresponding to a STM image of a 702-nm-thick Ag surface, and performed curvature and loop measurements. Note that all the results quoted below are from a single height profile. All in-plane lengths will be measured in units of the grid of this data, which is 1.625 nm. We also report briefly (Sec. VII C) a less thorough analysis of an STM data set from a different, but still self-affine, growth regime showing KPZ scaling.

**A. Quadratic measures and curvature moments**

Palasantzas and Krim [47] originally evaluated the roughness exponent

\[
\alpha = 0.82(5),
\]

from a fit to the standard (quadratic) height correlation function \(D_2(r)\) defined in Eq. (2.2). That correlation is similar to our second curvature moment \((C_2)\). This quantity shows a power law dependence on the scale \(b\), up to a correlation length which was estimated to be \(\xi = 25(5)\) (see Fig. 15a). A linear least-squares fit of the data with \(b < \xi\) gave \(2\alpha = 1.7(1)\), agreeing (as expected) with Eq. (7.1).

The third moment \((C_3)\) [Fig. 16(b)], shows distinct non-Gaussian behavior, as expected for nonequilibrium growth. It reaches a maximum at length scale \(b = 23\) which correlates well with \(\xi\). This indicates a morphology consisting of grains of typical size \(\xi\) that are rounded at the top. Such a morphology is clearly seen in three-dimensional renderings of the STM data in Ref. [47], or the gray-scale image in Ref. [48].

**Kleban’s nonlinear measure**

Recently, Kleban et al. defined a nonquadratic roughness measure rather different from any of those mentioned in Sec. III. First, for every scale \(b\) they constructed a smoothed version \(H_{s}(r)\) of the height function, as the average of \(H(s)\) for \(s\) in a \(b \times b\) square centered at \(s\) (alternatively by convolving with a Gaussian weight function of width \(\sigma\) ). Then they calculated the histogram \(P(H_s(x))\) of \(H_s(x)\) values for \(x\) ranging over the entire sample, and the skew moment of this

\[
\alpha = 0.82(5).
\]

**TABLE II**

<table>
<thead>
<tr>
<th>(p_c)</th>
<th>(C_2)</th>
<th>(\langle h^4 \rangle)</th>
<th>Direct</th>
<th>(D_{Ss})</th>
<th>(f)</th>
<th>Direct</th>
<th>(D_f)</th>
<th>(s_{\alpha})</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0.33(2)</td>
<td>0.33(1)</td>
<td>1.38(1)</td>
<td>1.35(2)</td>
<td>0.30(4)</td>
<td>0.30(1)</td>
<td>2.04(1)</td>
<td>0.77(3)</td>
</tr>
<tr>
<td>0.3</td>
<td>0.19(4)</td>
<td>0.09(2)</td>
<td>1.47(1)</td>
<td>1.46(2)</td>
<td>0.08(4)</td>
<td>0.38(1)</td>
<td>0.35(1)</td>
<td>0.08(5)</td>
</tr>
<tr>
<td>0.5</td>
<td>0.15(5)</td>
<td>0.06(1)</td>
<td>1.51(2)</td>
<td>1.50(2)</td>
<td>0.06(4)</td>
<td>0.40(2)</td>
<td>0.36(2)</td>
<td>0.13(5)</td>
</tr>
</tbody>
</table>

**FIG. 12** Second moment (a) and third moment (b) of the scale-dependent curvature \(C_s(x)\), for surfaces from the single-step model with \(p_c = 0.1 (\bigcirc)\), \(p_c = 0.3 (\bigcirc)\), and \(p_c = 0.5 (\bigstar)\). In (a), the \(p_c = 0.3\) and 0.5 data are consistent with \((C_2) = 0\), while the \(p_c = 0.1\) data show a strong (and non-Gaussian) breaking of up-down symmetry.

**FIG. 13** Average loop length \(\langle x \rangle\) as a function of the loop radius \(R\) for \(p_c = 0.1 (\bigcirc)\), \(p_c = 0.3 (\bigcirc)\), and \(p_c = 0.5 (\bigstar)\), in the single-step model. Note that the \(p_c = 0.3\) and 0.5 data are almost indistinguishable.
consistent with a KPZ-like ~50.4. That is, the loop data show a limited, P_s(2<0.22(2). After scaling regime yield D_f ~5 for the loop correlation function (~4.13) we calculate ~7.1. In Fig. 16 the large value of ~5, i.e., for the raw data. That is rather mysterious, since the surface certainly lacks up-down symmetry: it consists mostly of deep, narrow crevasses and rounded hills. Each crevasse would contribute to a tall on tail on the h=0 side of the distribution, while each hill contributes a peak and a sudden drop to zero on the h>0 side. However, the fluctuation in height from hilltop to hilltop smears out that sharp feature, making a spuriously symmetric distribution. As pointed out in Ref. [48], their smoothing of h(x) eliminates the deep crevasses so the smoothed surface H_s(x) does have a skewed height distribution (with skewness dependent on the observation scale b). This is consistent with our own conclusion that the height fluctuations are non-Gaussian.

B. Contour-loop analysis

We perform loop measurements and check whether the different scaling relations derived in Sec. IV are satisfied. The moment the scale-dependent curvature is useful as an independent measurement of the roughness and to assess the Gaussianity of the height fluctuations.

Using the loop algorithm (Appendix C), we measure the loop radii and corresponding loop lengths for 1000 contour loops constructed through randomly chosen points on the surface; however, we did not compute the loop correlation function. These loop measures support the scenario that the surface is self-affine up to a correlation length ~75. The average loop length is plotted against the loop radius in Fig. 16. We see a decade of power law scaling of the length with the radius, and from a linear least-squares fit of the data to a line, for 5<~<50, we find D_f = 1.06(2) (7.2) for the fractal dimension of contour loops. Using the formula for D_j (~Eq. [4.13]), we calculate ~7.2/4, in good agreement with the reported value (~Eq. [7.1]). In Fig. 16 the dashed line corresponds to the percolation value D_j = 1.75; we see that loops at scales much larger than ¸ show scaling consistent with this value.

Finally, the number of loops whose length exceeds s, P_s(¸), is plotted in Fig. 17. The data roughly show two scaling regimes with different exponents, before they are cut off by the system size. The knee occurs at loop lengths ~70 which, from Fig. 16, corresponds to a loop radius of ~20 or so; this again is comparable to the length scale ~ for loops whose length is in the interval (10,30) we extract the exponent ~2 = 6.6% (7.3)

while larger loops exhibit scaling consistent with the percolation value (indicated by the dashed line). Using Eq. (4.13) we find ~0.85(1), again in good agreement with the self-affine exponent reported by Palasantzas and Krim. Furthermore, inserting our results (7.2) and (7.3) into the scaling relation (4.8), we obtain ~0.51(1) (7.4) in agreement with our fundamental conjecture (~Eq. 14.12).

As also noted at the end of Sec. V, in Eq. (7.4) is mathematically equivalent to the already noted agreement of the ~ exponents obtained from D_j and from ~ using Eq. (4.13.) To summarize, the two loop measures we evaluated, as well as the moments of the scale-dependent curvature, all indicate self-affine scaling with a roughness exponent ~ ~0.85, up to a length scale ~ ~5. Beyond this scale the height fluctuations appear to be uncorrelated.

C. Other data sets

The large value of ~ found for the silver-on-quartz STM data of Palasantzas and Krim is indicative of molecular-beam-epitaxy-type growth, which has surface diffusion as a dominant relaxation process. This motivates the study of other data sets which might correspond to different universality classes of growth. For example, the KPZ equation describes growth dominated by desorption and/or vacancy formation, both of which are relaxation processes that do not conserve particle number [8,9].

Loop measurements were carried out previously on gold electrodeposits by Gomez-Rodriguez et al. [49]. These authors suggested the fractional dimension of loop contours as a useful measure for characterizing the surface morphology. While we lack a clean analysis in this field, we observe that the ratio of the fractal dimension to be D_j ~ 1.5 and 1.3, respectively. Now using Eq. (4.13) we calculate the roughness in these two regimes to be ~ ~0.6 and 0.4. The first is expected for Edwards-Wilkinson-type growth (~ ~0), while the second is in good agreement with the Kardar-Parisi-Zhang value ~ ~0.38 from most fits) or 0.40 (possibly exact) [45,46].

Cuskot et al. [50] studied the surface morphology of Ni films vapor deposited on a quartz substrate. (They were interested mainly in the effects of subsequent ion sputtering on the film.) We obtained a STM image of the as-grown Ni surface (before any sputtering) in the form of a 256x256 height array, and computed some of the contour-loop measures for it from a collection of 10000 loops. The results are consistent with a (KPZ-like) self-affine morphology with a roughness of ~ ~0.4. That is, the loop data show a limited range of scaling for loop radii 10<~<30. Direct fits to a straight line of the log-log plots of (s) vs (r) and P_s(r) in the scaling regime yield D_j = 1.34(4) and ~ ~2.2(2). After inverting the formulas for D_j (a) and (r) (a) in Eq. (4.13) we obtain the estimates ~ ~0.38(6) and 0.44(6), respectively.

VIII. DISCUSSION

Here we summarize our main results, compare and critique previously introduced measures of surface roughness, and describe some open problems and interesting directions.

A. Summary of results

We introduced (in Sec. III) measures for characterizing the spatial correlations of rough interfaces. Their common property is that they are not linearly related to the structure function of the height. First we introduced the scale-dependent curvature. Its third moment is an indicator of the skewness of the height distribution, and thus is a good criterion for whether or not a surface's height fluctuations are Gaussian. Our chief focus, though, was on the ensemble of contour loops as a means of characterizing surfaces. For a rough self-affine surface, the loop ensemble is critical, and we introduced three kinds of geometrical exponents associated with ~: for the loop correlation function (probability density) ~, that two points are on the same contour of loops, the fractal dimension of a contour loop, D_j; and ~ associated with the length distribution of loops. In particular, we conjectured a supersymmetry value ~ ~1 (see Sec. IV.C) which has been confirmed so far numerically (e.g., in Table I), but not analytically. The loop exponents satisfy scaling relations (derived in Sec. IV), and, granting the conjecture, their values are completely determined by the affine (roughness) exponent ~.

Next we showed how numerical values of the geometrical exponents can be extracted in practice from height data obtained from simulations or experiments. We first did this in Sec. V for artificial Gaussian surfaces (believed to be self-affine) and in Sec. VI for surfaces obtained from simulations of the single-step (growth) model (believed to be self-affine); this served as a test of the reliability of our scaling relations. Then in Sec. VII we processed an experimental data set—an STM image of a growth roughened silver film [47]—in the same fashion. The results here also confirmed the scaling relations which in this case adds to the evidence of the self-affine nature of the height fluctuations. The third moment of the scale-dependent curvature confirmed that the height fluctuations are non-Gaussian, while the contour-loop fractal dimension and size distribution indicated self-affine scaling with ~ ~0.85.

Experimental data often exhibit self-affine scaling up to a correlation length ~(~). We argued (in Sec. IV.E) that the loop exponents, for loops whose linear size exceeds the correlation length, are determined by exactly known percolation exponents. The crossover between the self-affine and percolative regime was visible (with a consistent value) in every kind of measure on the experimental data in Sec. VII—the same was true for Gaussian random surfaces with an artificial length scale cutoff (Sec. V.D). The numerical values of

FIG. 15. Second moment (~) and third moment (~) of the scale-dependent curvature, as evaluated for a 702-nm-thick Ag film grown on quartz, from the STM data of Palasantzas and Krim (~Ref. [47]).

FIG. 16. Mean contour length (~) as a function of radius ~, for the Ag film of Ref. [47]. Here 1000 contour loops were collected from the STM data of Ref. [47]. The solid line is the least-squares best fit for radius 2<~<125; its slope is the estimated fractal dimension D_j. The slope of the dashed line is equal to the bulk dimension of critical percolation clusters.

FIG. 17. Cumulative distribution of contour loop lengths from STM data, for the Ag film of Ref. [47]. The solid line is the result of a linear fit to the data in the affine-scaling regime. The slope of the dashed line corresponds to the exponent ~ ~2 in the percolation regime.

FIG. 18. Cross-sectional transmission electron microscope (TEM) images of the Ni surface of Ref. [50], showing (a) the as-grown surface, (b) the sputtered surface, and (c) the sputtered surface with a ~0.45% Ag film.
Nonlinear Measures for Characterizing Rough . . .
KONDEV, HENLEY, AND SALINAS

TABLE III. Roughness measures.

<table>
<thead>
<tr>
<th>Quantity</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>((h(x) - \langle h \rangle)_s^2)</td>
<td>variance in (h) on (s) patch</td>
</tr>
<tr>
<td>(D_s(h(x)))</td>
<td>height correlation</td>
</tr>
<tr>
<td>(\langle h(x) \rangle^{(2/3)})</td>
<td>Fourier spectrum</td>
</tr>
<tr>
<td>(C^2_s(h(x)))</td>
<td>&quot;b-dependent curvature&quot; variance</td>
</tr>
<tr>
<td>(\langle h(x) \rangle_s)</td>
<td>skew moment in (h) on (s) patch</td>
</tr>
<tr>
<td>(q)</td>
<td>q-multifluid height correlation</td>
</tr>
<tr>
<td>(\langle h(x) \rangle_s^q)</td>
<td>skew moment of (b)-smoothed height</td>
</tr>
<tr>
<td>(C^3_s(h(x)))</td>
<td>&quot;curvatures&quot; skew moment</td>
</tr>
<tr>
<td>(C^4_s(h(x)))</td>
<td>&quot;curvatures&quot; quartic moment</td>
</tr>
</tbody>
</table>

Cubic and other nonquadratic measures

1. Quadratic roughness measures

The most familiar measures are quadratic of which the first three were summarized in Sec. II B. Besides three well-known quadratic measures, we include a fourth which has not been previously applied: the variance of scale-dependent curvature, \(C^2_s(h(x))\), which we introduced in Eq. (3.3). (Of course, the ensemble expectation cannot depend on \(x\).) Its behavior is very similar to that of the height-difference correlation \(D_s(h(x))\); for \(C^2_s(h(x))\) is of interest mainly for comparison with the higher moments of \(h(x)\). In practical applications the Fourier spectrum is probably the best of these.

A key fact about the quadratic measures is that, given the computer algorithm for one of them, one can compute the complete function for any other one as a linear transform (correlation with some kernel). This property is not true for higher moments. Notice also that the quadratic measures are invariant under \(h(x) \rightarrow -h(x)\), and so cannot possibly characterize the breaking of up-down symmetry in the growth process. Nor can they identify deviations from Gaussianity, since one can produce a Gaussian ensemble (as in Sec. V A) with any given Fourier spectrum.

2. Nonquadratic roughness measures

Essentially all of these have been developed by analogy with quadratic measures, simply replacing the second power by a higher power. Our curvature-moment function seems to be the first generalization of the height-difference function that captures the up-down asymmetry. A simple generalization of the b-box variance is the b-box \(q\)-moment \((\langle h(x) \rangle - \tilde{h}(x))^q\) of \(q\)-moment \(\langle h(x) \rangle_s\). The loop correlation function captures the up-down asymmetry, when scaled by \(\langle h(x) \rangle_s - \tilde{h}(x)\), defines a scale-dependent, dimensionless skewness that measures the deviation from Gaussianity. This appears to be a simple and attractive measure, but we know of no nonquadratic \(C^2_s(h(x))\) that measures the deviation from Gaussianity. This fact is not the case for experimental data where the system size is typically not a tunable parameter.

The loop correlation function is too tedious to compute, and since its exponent \(2\chi_s\) is superuniversal it does not yield an estimate of \(\tilde{h}(x)\). Nevertheless \(\langle h(x) \rangle_s\) is a useful check of the relations to date; our curvature moment \(C^2_s(h(x))\) is similar in spirit, but probably not linearly related. We evaluated the quartic curvature moment \(C^4_s(h(x))\), but these data were less useful than our other measures: they do not reveal the non-Gaussian nature as strikingly as \(C^2_s(h(x))\) does. The dimensionless ratio \(C^4_s(h(x))/C^2_s(h(x))^2\) is in the Gaussian case, but may not differ much in a non-Gaussian ensemble. Furthermore, the roughness exponent was fitted less precisely from \(C^2_s(h(x))\) than from any other measure, probably due to the sensitivity of higher moments to rare events. The analysis in Ref. [48], summarized in Sec. VI B.4, appears to be the first application of a scale-dependent roughness measure to characterizing an up-down asymmetry. However, we believe a local roughness measure such as the box-skewness (or better) our mean cubed height convex gives a more meaningful characterization. In a sense, the smoothed-height skewness is the opposite of the local measures since it includes the fluctuations from all length scales larger than \(b\), while the local measures include the fluctuations from scales comparable to or smaller than \(b\); only the latter would be expected to scale as \(\sim b^\alpha\).

3. Loop measures

The other nonquadratic roughness measures, of course, are the loop measures defined in Sec. III. The length and connectedness of a loop, manifestly, depend on the heights \(h(x)\) in a highly nonlinear fashion, and one might expect the loop exponents to be independent of the roughness exponent \(\alpha\); the loop properties might have distinguished between different universality classes of growth which happen to have similar average values. From this viewpoint, we see much less reason to distinguish between loops of different sizes. The loop exponent \(\delta\) is of interest mainly for comparison with the higher moments of \(h(x)\) and nonlinear systems.

The other nonquadratic roughness measures include the loop exponents for measuring the size distribution exponent \(\gamma\); the loop plots show a weaker change of slope at this crossover than the Fourier spectrum does. It seems worthwhile nevertheless to compute loop measures. In a sense they depend on higher order correlation functions of the heights, then the agreement between the \(\alpha\) values extracted from loops and from other measures is an additional, stringent test of self-affineness. Also we empirically observe that loop measures, and in particular the average loop length as a function of loop radius, are very self-averaging, and measurement of \(\alpha\) from them produces numbers that are consistent with either the real-space or Fourier-space methods. Finally, although the loop exponents are the same for different universality classes with the same \(\alpha\), we do expect different loop characteristics.

For computer generated height data the loop-size distribution is, perhaps, the single most valuable plot, because two different exponents can be obtained from scaling plots such as Fig. 7. This is not the case for experimental data where the system size is typically not a tunable parameter.

The loop correlation function is too tedious to compute, and since its exponent \(2\chi_s\) is superuniversal it does not yield an estimate of \(\tilde{h}(x)\). Nevertheless \(\langle h(x) \rangle_s\) is a useful check of the relations to date; our curvature moment \(C^2_s(h(x))\) is similar in spirit, but probably not linearly related. We evaluated the quartic curvature moment \(C^4_s(h(x))\), but these data were less useful than our other measures: they do not reveal the non-Gaussian nature as strikingly as \(C^2_s(h(x))\) does. The dimensionless ratio \(C^4_s(h(x))/C^2_s(h(x))^2\) is in the Gaussian case, but may not differ much in a non-Gaussian ensemble. Furthermore, the roughness exponent was fitted less precisely from \(C^2_s(h(x))\) than from any other measure, probably due to the sensitivity of higher moments to rare events. The analysis in Ref. [48], summarized in Sec. VI B.4, appears to be the first application of a scale-dependent roughness measure to characterizing an up-down asymmetry. However, we believe a local roughness measure such as the box-skewness (or better) our mean cubed height convex gives a more meaningful characterization. In a sense, the smoothed-height skewness is the opposite of the local measures since it includes the fluctuations from all length scales larger than \(b\), while the local measures include the fluctuations from scales comparable to or smaller than \(b\); only the latter would be expected to scale as \(\sim b^\alpha\).

4. New experimental techniques which provide complete real-space images of the fluctuating quantity of interest (rather than the statistics of the system at the point only a few points), have been developed in every physical science. Consequently, measures which usefully exploit this wealth of information will gain in importance. In turn, the ability to measure new (and nonuniversal) correlations may inspire new theories that can predict the correlation behavior.

1. Turbulence

Fluid dynamics is a good example of the interplay just mentioned between theory and experiment: formerly two (sometimes more) point correlations were measured by hot-wire probes, and the same correlations were the objects of the Green’s function method. As full images become available of the velocity field, many new measures are computed in order to capture more of the available information.

Indeed, the measures introduced here might be adapted to a geometrical description of the advections of passive tracers by turbulent flows seem to be especially promising problem. There is already considerable interest in the characterization of the fractal measures of the contours of (say) constant tracer concentration [54, 53, 54]. To maintain the analogy of a surface’s symmetry under global shifts of the height, one should modify the lognormal distribution and use contours spaced equally on the logarithmic scale.) Measurements of the fractal dimension of isocontour lines of a passive tracer advected by a magnetically driven, turbulent, two-dimensional flow were reported by Cardosa et al. [4]. They found \(D_1 = 1.55\), which, assuming the concentration field is self-affine, yields a roughness exponent \(\alpha = 3 - 2D_1 = 0.31\). Indeed, \(q = 0.3(0.3)\) was measured by the authors, by applying the \(q = 1\) multifracture correlation measure [entry 6 in Table III]. We therefore infer that their measurements are consistent with a self-affine morphology for the concentration field. Details of the complete loop and curvatures analysis of this data set will be reported in a separate paper [56].

2. Other dimensions

In this connection, it is interesting to consider the generalization \(h(x)\) on spatial dimension \(d\) on \(x\) or 2. With each higher dimensionality there is greater richness in distinct geometrical measures that can be defined for iso-surfaces. For a hypersurface in \(d+1\) dimensions—like the concentration function in three-dimensional passive advection—the level set may be multiply connected and even knotted. Nevertheless the size distribution exponent \(r\) of the fractal dimension \(D_r\) and \(x_i\) of the connectedness correlations, can be generalized directly. But we see much less reason to expect a superuniversal connectedness correlation exponent in dimensions higher than \(d = 2 + 1\).
ACKNOWLEDGMENTS

where \( N_l \) is the number of loops in \( y \) (which is the same as the number of loops in \( y' \)), and the 2 appears as a result of summing over the two possible orientations for each loop in \( y' \).

By mapping the \((2,1)\) loop model to the four-state ferromagnetic Potts model on the triangular lattice, Nienhuis or perimeter function

\[
F(r) = \text{const} \int d^4 \nabla \phi(x),
\]

which by equirapidity implies Eq. (2.7) with \( a=0 \), so indeed the surface is self-affine. This appendix only summarizes arguments made previously in Refs. [19, 57], and [58].

Consider a statistical model with microscopic heights \( z_j \) defined on a triangular lattice (\( \mathbb{J} \)), such that \( z_j \) changes by 0 or ±1 between nearest neighboring sites. The partition function of the model is

\[
Z = \sum_{\mathcal{J}} \prod_j w(z_j - z_{\mathcal{J}}),
\]

where \( w(0)=1 \) and \( w(\pm 1)=K \); the sum goes over all microscopic height configurations unrelated by a global height shift.

A contour-loop configuration \( \gamma \) is specified by drawing closed (periodic boundary conditions ensure that all contour lines are closed), oriented, nonintersecting loops along the bonds of the dual honeycomb lattice, which separate sites that differ in height by ±1 (the sign determines the loops orientation). In terms of the loops, the partition function is

\[
Z = \sum_{\gamma} \prod_j K^{n_j},
\]

where \( K \) is the fugacity of an occupied bond (i.e., one covered by a loop), and \( n_j \) is the number of occupied bonds in \( y' \).

This model is equivalent to the \((2,1)\) loop model introduced by Nienhuis [31]. This is seen by rewriting the partition function in terms of nonoriented contour configurations.

\[
Z = \sum_{\gamma} K^{L}\gamma^{2}\lambda^{|L|},
\]

where \( L \) is the number of loops in \( y \), the number of loops in \( y' \), and the 2 appears as a result of summing over the two possible orientations for each loop in \( y' \).

In the percolation regime, evidently, the statistical properties of the contours of a particular level set depend on the chosen level \( h \). (This was impossible in the self-affine regime, since in that case the fluctuations of \( h \) were unbounded.) But we have previously studied the union of all contours with different \( h \), corresponding to all values of \( p(h) \) from 0 to 1. That is, indeed, the ensemble sampled by our computer codes (see Sec. V C). We will now derive the exponents \( \gamma_{r} \) and \( \gamma_{s} \) of this ensemble, defined analogously to \( r \) and \( s \) in Eqs. (3.13) and (3.17). Most of the loops at a large enough scale \( R \) come from levels sets at height \( h \equiv \langle p(h) \rangle R \), rather than from the exponential tails of the distribution (B4) for the other \( h \) values. Thus these obey the percolation scaling and have all fractal dimension \( D_{F} = D_{S} \). This behavior is illustrated in Fig. 2(c).

Now, in the percolation regime, \( P(\gamma) \) as defined in Sec. III C, is just proportional to the integral of \( P_{\gamma}(x)p(h) \) over \( h \). A weighting factor \( |d\phi|/\phi \) should be included as the contours are equally spaced, and \( p(h) \) is normalized to unity. Since the large contours come from \( p > p_c \), just that part of the distribution matters. Inserting Eq. (B4) into Eq. (B6), one obtains

\[
P(x) = \frac{dp_x}{\sqrt{1-x \gamma_{s}}} A(\const) \gamma_{r}^{p-\gamma_{r}^{-1}},
\]

with

\[
\gamma_{r} = \gamma_{l} + \gamma_{s} = 1.2, \quad \gamma_{s} = 1.2/3.17.
\]

Finally, given Eq. (B6), the simplest route to the loop (connectedness) correlation exponent is to use the exponent relation (4.8), this gives

\[
D_s = 7/4
\]

is known exactly [34].

The perimeter loops for percolation at \( p_c \) also satisfy a hyperscaling relation analogous to Eq. (4.3), with \( a \) replaced by zero. That is, the largest cluster (or perimeter) diameter inside a box of side \( l \) is least \( \sim l^{-1} \). From this follows a relation for \( \gamma_{r} \):

\[
\gamma_{r} = 1 + 2D_{F} = 15/7.
\]

When \( p>p_{c} \), the cluster (and perimeter) ensemble scaling is cut off at the percolation correlation length \( \xi_{p_c} \), which diverges near \( p_{c} \) as

\[
\xi_{p} = v_{p} \xi_{p_c}^{g_{p}},
\]

where \( v_{p} \) is the usual percolation-correlation length exponent, and \( g_{p} = 4/3 \) is known exactly [62, 34]. In this case, the loop distribution is

\[
P_{\gamma}(x)p \sim x^{-(\gamma_{1} + \gamma_{s})} A(\const) \gamma_{r}^{p-\gamma_{r}^{-1}},
\]

This behavior is illustrated in Fig. 2(c).

In particular, the \((2,1)\) loop model simply as one of many possible lattice discretizations of a logarithmically rough (\( a=0 \)) self-affine surface, then the exponent \( \gamma_{l}/2 \) should necessarily appear in other lattice models that map to rough surfaces. Indeed the same value of this exponent follows also from the exact solution of the \((2,1)\) loop model on the square lattice [59], and the \( n=2 \) fully packed loop model on the honeycomb lattice [60].

APPENDIX B: PERCOLATION SCALING OF CONTOURS FOR UNCORRELATED HEIGHTS

This appendix derives the scaling behavior of the loop ensemble when the random heights \( h(x) \) have a finite variance and [beyond a correlation length \( \xi \)] are uncorrelated, this describes early stages of growth, as in Sec. IV E.

To model the contour loops at length scales greater than \( \xi \), first coarse grain the system into boxes of side \( \xi \). The average height \( h \) in each box is an independent random variable parameterized by \( p(h) \), the probability that \( h \sim h \). Defining all the contours \( 0 \leq \xi < L \) as \( \Gamma \), the union of \( \Gamma \) reproduces the (uncorrelated) percolation clusters for occupancy \( p(h) \). Then every contour of constant height \( h \) simply permutes the orientation of such a cluster. This mapping is well known from Ref. [51], and is widely applied in the theory of the \( \phi^4 \) model (33, 31).

The percolation clusters— as well as their perimeters—are self-similar only at \( p=\mu_c \), the percolation threshold; we will first discuss their (known) loop exponents. The behavior when \( p>\mu_c \), can easily be derived from well-known percolation scaling relations. The final step will be to integrate these results over \( p \), since the loop ensemble we simulate actually corresponds to the union of perimeter ensembles for all \( p \).

Contour loops and critical percolation

Fixing \( p=p_{c} \), a moment, the perimeter loop ensemble may be characterized by exponents \( D_{s} \) and \( \gamma_{s} \), with definitions analogous to Eqs. (3.9) and (3.13) for \( D_{s} \) and \( \gamma_{s} \). (The subscript ‘h’ stands for ‘hull,’ as the perimeter is often called.)

The fractional dimension

\[
\gamma = 4.8
\]

FIG. 18. Construction of contour loops of a random surface on a lattice. Heights \( \Delta \) are indicated by numbers in the cells; \( h_{max} \) is the height of the level set through the chosen point (filled circle), while \( h_{min} \) is the height of the ‘saddle point’ (unfilled circle). Our definitions of the diameter \( R \) and loop length \( s \) are indicated. The solid arrow connects points on the same loop, and thus contributes to the loop correlation function \( G(r) \). The dashed arrow does not contribute: it connects points of the same level set, but they are on disconnected loops.

\[
2\pi s = 4 - 2D_{s} \Delta = 5/4\Delta.
\]

Equation (B7) could alternatively be reached by first noting that the corresponding exponent is \( 1/2 \) for the percolation hull ensemble at \( p_{c} \), and then averaging the loop connectedness correlation function analogous to Eq. (B5).

APPENDIX C: LOOP FINDING ALGORITHM

Given a square lattice \( L \times L \) on which the heights \( h \) are defined, and a point \( x_0 \) on the dual lattice \( L^* \), the task of the loop finding algorithm is to construct a contour loop of the surface which passes through the point \( x_0 \). The contour loop grows along the bonds of \( L^* \) that cuts those bonds of \( L \) that have vertices with heights lying above and below the contour height (Fig. 18). To implement this idea we first define the level height \( h_{max} \), which is the average of the four heights around the plaquette centered at \( x_0 \). Second, we assign to all sites of \( L \) or \( -L \) signs according to whether they are above or below the chosen level \( h_{max} \). Now, starting from \( x_0 \), we form the contour loop by drawing links on the dual lattice which cross the bonds of \( L \) connecting \( + \) and \( - \) sites. This is repeated until the walk returns to the starting point \( x_0 \); the finite extent of the lattice \( L \) is dealt with by implementing periodic boundary conditions.

Special care must be taken whenever a ‘saddle-point’ plaquette is reached, that is one where the sites of the lattice are assigned \( + \) or \( - \) signs cyclically around the plaquette. In this case four links meet at the point in the center and we must resolve the connectivity there by an additional rule, so as to convert this pattern into two \( 90^\circ \) turns that are not quite touching. One natural condition on the rule is that it should be reversible, that is one should find the same loop whether one starts traversing it clockwise or counterclockwise. A sec-
and condition is that it ought to be invariant under reflecting all heights by $h(x) \rightarrow -h(x)$. A physically sensible rule which satisfies both conditions makes use of the average height $h_{\text{avg}}$ of the four heights around the saddle-point plaquette. If $h_{\text{avg}} > h_{\text{max}}$, we view the center of the plaquette as being lower than the level of the contour loop and the connectivity is resolved by having the $+z$ sites inside the $90^\circ$ turns. In the opposite case, $h_{\text{avg}} < h_{\text{max}}$, the $+z$ sites lie outside the $90^\circ$ turns, see Fig. 18. (The agreement of loop data from the single-step model with parameters $p_x$ and $1 - p_x$, as explained in Sec. VI was a valuable check of the up-down symmetry of our loop-finding algorithm.)

Once a contour loop through $x_0$ has been constructed its length and radius are recorded, assuming that the loop is topologically trivial. (Due to periodic boundary conditions, loops with nonzero winding numbers are possible, and these we discard.) The contour loop length $r$ is equal to the number of steps made during the loop construction, while the radius $R$ is the size of the largest square which covers the loop, i.e., the maximum displacement in the $x$ or $y$ direction. In this case topologically trivial loops also contribute to the correlation function $G(r)$; for every point on the loop that is a distance $r \times (i+1)h(i)$ from the starting point $x_0$, the array element $G(l)$ is increased by 1. We define $G(r)$ for our simulations as $r G(r)/2$, which asymptotically is normalized to $G(r)$ in Sec. III C.

So far we have assumed that the height variables are real and the condition $h_{\text{avg}} < h_{\text{max}}$ is almost never fulfilled. This is not the case for interfaces which arise from discrete growth simulations like the one presented in Sec. VI, where the height variable takes on integer values. In this case the resolution of the connectivity should be completely random, but we must ensure that we use the same choice if the loop returns to the same plaquette. The simplest way to do this, which is what we have implemented, is to take the original integer heights and ‘divide’ them—add small amounts of random, uncorrelated Gaussian noise to all $h(i)$. This will also solve the problem of choosing $h_{\text{avg}}$; it would be meaningless for any two heights to precisely coincide, although this will happen occasionally as the price of roundoff error. When this does happen, we start over by choosing a new initial site $x_0$.

[10] Height configurations have also been derived (for biased interfaces) from dark-field transmission-electron-microscope images: see X. Chen and J. M. Gibson, Phys. Rev. Lett. 81, 4919 (1998).
[11] Our treatment is limited to a single-valued height field, thus coarse graining of $h(x)$ is mandatory when overhangs are present at atomic length scales, and we do not consider the case when overhangs persist to large scales.