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Surface growth by random deposition of rigid and wetting clusters

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ABSTRACT

Surface grown by the deposition of rigid and wetting clusters has been investigated using Monte Carlo simulations in 1 + 1 dimensions. Dynamic scaling exponents were determined using the time evolution of the roughness, the local width, the height–height correlation function, and the power spectrum. The values obtained for the roughness exponent clearly reflect the growth mechanism adopted for deposition. In the case of wetting clusters, the roughness exponent corresponds to that of random deposition, but a correlation appears for low window size, with a crossover that is related to the average cluster size and cluster size distribution. On the other hand, rigid cluster deposition belongs to the KPZ universality class. However, determined scaling exponents converge very slowly to those corresponding to KPZ.

1. Introduction

The kinetic roughening of thin-film growth fronts under non-equilibrium conditions has attracted considerable interest due to their wide application in critical components for electronic, magnetic, and optical devices [1]. The functionality of these films is determined by their physical structure, in particular the surface roughness and the grain size distribution in the case of polycrystalline films. A complete characterization of the surface and the microstructure can provide a deeper understanding of the processes that drive the physical evolution during film formation. In many cases, the dynamics of the growing film leads to spread correlations over the whole system and produce scale invariant surfaces that are described by the Family–Vicsek ansatz [2–5]. Depending on the growing mechanisms, the resulting surface evolution can be determined as belonging to distinct universality classes. Thus, measuring the set of scaling exponents for a particular system allows to associate it with some universality class and consequently with a dominant growing mechanism.

Random deposition of agglomerated particles (clusters) is one of the commonly used methods in the fabrication of nanostructured materials. Since clusters at the surface occupy more than one unit size, depending on the deposition mechanisms, a porous bulk can be generated. This property is desirable in manufacturing nanostructured materials for many applications, such as magnetic storage and solar cells [6,7]. In the past, a lot of work has been devoted to study the deposition of particles. However, less attention has been paid to the cluster incorporation process, but at least two different universality classes were reported [8–10].

In the present work, we report a dynamic scaling analysis of a surface formed by deposition of clusters onto a one-dimensional substrate. We studied two types of cluster incorporation mechanisms, one in which the clusters fall randomly and wet the surface copying its profile, and the resulting aggregate is not porous. Hereafter, we will refer to this model as wetting cluster deposition (WCD). The second model deals with rigid clusters that stick at the first point of contact and do not change their shape after their incorporation to the substrate, generating a porous deposit. Hereafter, we will refer to this model as rigid cluster deposition (RCD). We also studied the effects of the cluster size distribution on the scaling exponents.

Hellmut Haberland and co-workers have investigated the structure of thin films grown by energetic cluster impact deposition [11]. In this technique, ionized metal clusters are electrically accelerated onto the substrate. It has been observed that the final film morphology depends on the deposition parameters, such as the size and the incident energy of the clusters. In particular, the increase of the cluster impact energy leads to the formation of more compact films. They found a transition from a porous film with multiple voids to a dense film with a nearly bulk density as the incident energy is increased from 0.1 eV/atom to 10 eV/atom [12]. For low kinetic energies, clusters stick at the first point of contact, resembling a ballistic deposition. For high-impact energies, incident clusters lead to a redistribution of atoms in which the substrate atoms are involved, a smoother resulting surface, and then the Edwards–Wilkinson class is expected. At intermediate energies, impinging clusters wet the surface without affecting the substrate atoms, as seen in Ref. [12] for Mo clusters. In this case, we expect that this technique resembles the WCD. Softer metals are prone to wet the surface after landing. Indeed, in Refs. [13] and [14] it can be seen that Al and Cu clusters with very low kinetic energy incorporate to the surface without the generation of voids and no intermixing resembling the WCD.

To obtain the scaling exponents for the rigid cluster deposition (RCD), we calculated the interface width, $W(L,t)$ and also the local width (LW), the height–height correlation function (HHCF), and the power spectrum (PS). The local surface roughness $w(l,t)$, which

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represents height fluctuations in different length scales, shows a crossover related to the cluster lateral size, giving rise to a small-scale roughness exponent that differs from the global one [15,16]. Also, for distances smaller than the mean cluster size, we found different values for the roughness exponents, obtained from HHCF and LW, whereas for large distances, the exponents have the same values and are determined by the growth mechanism adopted for deposition, as reported in Refs. 9 and 10. For rigid clusters, the scaling exponents converge very slowly to those of KPZ universality class for very large system sizes [8], whereas for wetting cluster deposition (WCD), the scaling exponents correspond to those of random deposition but a correlation appears at small scale due to cluster finite size.

2. Dynamic scaling framework

The most obvious quantitative characteristic of a rough surface is the root-mean square of the height field, known as the surface width or roughness

$$W(L,t) = \sqrt{\frac{1}{L}\sum_{i=1}^{L}\left[h(i,t)-\bar{h}(t)\right]^2}, \tag{1}$$

where $h(i,t)$ is the surface height measured from the flat substrate of size L at the position i at the time t, and $\bar{h}(t)$ is the mean height of the interface at the same time.

Family and Vicsek proposed a scaling relation that connects the surface roughness with the linear size of the lattice and time [3]. This scaling relation, applicable to a large number of growth models, is written as

$$W(L,t) \sim L^{\alpha} f\left(t/L^{z}\right), \tag{2}$$

where the scaling function $f(u)$ is a function that behaves as u^{β} for $u << 1$ and as a constant for $u >> 1$. The parameters α and β are the roughness and growth exponents, respectively, and $z = \alpha/\beta$ is the dynamic scaling exponent. α and β constitute a pair of numbers that can be used to classify quantitatively the spatial and temporal scaling of growing surfaces and then to identify the growth process.

On the other hand, quantitative information about the height fluctuations and lateral correlation is given by the height–height correlation function HHCF

$$C(l,t) = \left\langle (h(x+l,t)-h(x,t))^2 \right\rangle^{1/2}. \tag{3}$$

$C(l)$ constitutes a quantitative description of how the heights at different points of the surface are correlated as a function of their separation. For a self-affine surface, $C(l,t)$ scales with l as

$$C(l,t) \approx l^{\alpha} f\left(t/l^{z}\right), \tag{4}$$

Another important quantity to characterize the surface growing process is the local interface width $w(l,t)$ defined as

$$w(l,t) = \left\langle \frac{1}{l}\left(h^2(x,t)-\langle h(x,t)\rangle\right)^2 \right\rangle^{1/2}. \tag{5}$$

It is well known that $w(l,t)$ scales as

$$\begin{aligned} &w(l,t) \sim t^{\beta}, \text{ for } t << l^{z}, \text{ and} \\ &w(l,t) \sim t^{\alpha}, \text{ for } t << l^{z}, \end{aligned} \tag{6}$$

where l is the window size.

Finally, other convenient way of summarizing data is the spectral power density or structure factor

$$S(k,t) = \langle h(k,t)h(-k,t)\rangle, \tag{7}$$

being $h(k,t)$ the kth Fourier mode of the surface height deviation around its spatial average for a given time t

$$h(k,t) = \frac{1}{L^{1/2}}\sum_{x}\left[h(x,t)-\bar{h}(t)\right]\exp(ikx). \tag{8}$$

The structure factor scales as

$$S(k,t) = k^{-(2\alpha+1)} g\left(t/k^{-z}\right). \tag{9}$$

For $u << 1$ $g(u) = u(2a+1)/z$ and for $u >> 1$ $g(u) =$ constant, and then

$$\begin{aligned} &S(k,t) \sim k^{-(2\alpha+1)} \quad u << 1 \\ &S(k,t) \sim t^{(2\alpha+1)/z} \quad u << 1 \end{aligned} \tag{10}$$

Surface structures that preserve a similar morphology upon a change of magnification are termed self-affine and obey the well-known Family–Vicsek (FV) scaling ansatz, which plays a central role in growth theories [2–4]. However, not all systems exhibit FV scaling. For instance, it has been reported that the formation of features during etching or film growth by grains leads to a more complex roughening process [16,17]. On the other hand, an evolving pattern can show different scaling at the global and at the local length scales. Thus, a common set of scaling parameters is no longer adequate to characterize the dynamic behavior at different scales and additional exponents are needed to fully characterize the observed growth [18–21].

3. Monte Carlo modeling

We performed simulations using the standard Monte Carlo method in 1D. The surface is represented by a one-dimensional vector where each element corresponds to the height at each site. To study the dynamic scaling exponents, we used clusters of size $N \times N$ (a square) and $1 \times N$ (a horizontal rod). The deposition process starts building a cluster and choosing a site at random (i) over de surface. We evaluated two types of aggregation mechanisms. One in which the arriving cluster can disassemble, and it changes its shape by copying the surface profile. All sites between i and $i + N$ grow the cluster original height, and the growth is conservative (WCD model). The second model corresponds to rigid clusters in which they land atop of the highest surface site between i and $i + N$, giving rise to a non-conservative growth. If the site j, with $i \le j \le i + N$, presents the local highest site $h(j)$, then the cluster incorporation at the surface, for a horizontal rod, takes place changing the heights of all sites, from i to $i + N$ to $h(j) + 1$ (RCD model). A similar process is used for the deposition of a cluster of size $N \times N$ but finally the heights of all sites from i to $i + N$ are changed to $h(j) + N$. Other processes, such as rearrangement of surface particles (diffusion) or detachment, are not allowed.

Fig. 1 depicts the studied models. In the RCD model, once a cluster is in contact with a particle of the substrate, it is incorporated without changing its shape. In Fig. 1a, two clusters consisting of four particles, of 2×2 and 1×4, are shown after arriving to the surface in their final position. Conversely, in the WCD model, arriving clusters are incorporated to the aggregate by wetting the surface. This means that the particles of the cluster can move down until making contact with a particle of the substrate. In Fig. 1b, the final aggregate morphology is depicted after the incorporation of two clusters as in Fig. 1a but using the rules of the WCD model.

The simulation starts from a flat substrate configuration and evolves with successive deposition of clusters until it approaches steady state. One Monte Carlo time corresponds to the deposition of one monolayer of particles. We checked that steady state was reached by assessing the evolution of the surface roughness. Monte Carlo simulations were