Unusual Scaling for Pulsed Laser Deposition

Berit Hinnemann,[1] Haye Hinrichsen,[2] and Dietrich E. Wolf[1]
[1]*Theoretische Physik, Fachbereich 10, Gerhard-Mercator-Universität Duisburg, 47048 Duisburg, Germany*
[2]*Theoretische Physik, Fachbereich 8, Universität GH Wuppertal, 42097 Wuppertal, Germany*
(Received 29 January 2001; published 7 September 2001)

We show that a simple model for pulsed laser deposition exhibits an unusual type of scaling behavior for the island density in the submonolayer regime. This quantity is studied as a function of pulse intensity and deposition time. We find a data collapse for the *ratios of the logarithms* of these quantities, whereas conventional scaling as observed in molecular beam epitaxy involves ratios of powers.

DOI: 10.1103/PhysRevLett.87.135701 PACS numbers: 64.60.Ht, 68.55.Ac, 81.15.Fg

Many systems in equilibrium and nonequilibrium statistical physics exhibit power-law scaling. This means that a system with an observable, M, depending, for example, on two parameters, z_1 and z_2, looks the same, if the units of M, z_1, and z_2 are rescaled by certain factors which are related to each other by power laws. Such a scaling transformation can be written as

$$z_1 \to \Lambda z_1, \qquad z_2 \to \Lambda^{\beta} z_2, \qquad M \to \Lambda^{\alpha} M\,, \tag{1}$$

where Λ is a scaling parameter and α, β are certain exponents. Equation (1) implies the scaling form,

$$M(z_1, z_2) = z_1^{\alpha} f(z_2/z_1^{\beta})\,, \tag{2}$$

where f is a scaling function depending on a scale-invariant argument. This type of scaling can be observed in a vast variety of applications, including equilibrium critical phenomena [1,2], growth processes [3–8], driven diffusive systems [9], as well as phase transitions far from equilibrium [10,11].

As an example for power-law scaling, which we are going to contrast with a different type of scaling in this paper, let us consider the following well-known simple model of molecular beam epitaxy (MBE): A particle beam deposits atoms onto a flat substrate at a flux F (atoms per unit area per unit time). The atoms diffuse on the substrate with a surface diffusion constant D until they meet another adatom, in which case they form a stable and immobile nucleus of a two-dimensional island on the surface, or until they attach irreversibly to the edge of an already existing island.

The observable examined in this paper is the time-dependent nucleation density, n, i.e., the number of nucleation events per unit area in the first layer integrated over time. Obviously, the nucleation density is a fundamental quantity characterizing the island morphology as it indicates how many islands are formed. By definition, the nucleation density increases monotonically with time and saturates when the first monolayer is completed.

In MBE, the two parameters D and F can be used to construct a characteristic length,

$$\ell_0 = (D/F)^{1/4}. \tag{3}$$

When the nucleation density reaches the value $1/\ell_0^2$, the rate of nucleation events decreases drastically since it becomes more likely that an adatom attaches to an already existing island instead of forming a new nucleus with another adatom. In terms of the coverage $\Theta = Ft$, i.e., the total number of deposited atoms per unit area, the time dependence of n is known to obey the scaling form,

$$n(\ell_0, \Theta) = \ell_0^{-2} f_1(\Theta \ell_0^2)\,. \tag{4}$$

As shown in Ref. [12], the scaling function f_1 behaves as

$$f_1(z) \propto \begin{cases} z^3 & \text{for } 0 \le z \ll 1 \\ z^{1/3} & \text{for } 1 \ll z \lesssim z_{\max}\,. \end{cases} \tag{5}$$

$z_{\max}$ is determined by the condition that the whole surface has been claimed by the islands, so that no further nucleation in the respective layer is possible [13]. Thus, MBE exhibits standard power-law scaling as described by Eqs. (1) and (2). However, in pulsed laser deposition (PLD), an alternative method of growing thin epitaxial films, we find that the nucleation density shows a fundamentally different type of scaling, which in MBE is realized only approximately.

In order to work out the difference most clearly, let us assume that the amplitudes of the asymptotic power laws in (5) are equal (which is approximately the case in MBE), so that one can get rid of them by dividing $n(\ell_0, \Theta)$ by the nucleation density at a particular coverage. In the following, we extrapolate the power law (5) beyond $z_{\max}$ to a full monolayer, $\Theta = 1/a^2$, and define

$$M(\ell_0, \Theta) = n(\ell_0, \Theta)/n(\ell_0, 1) = \ell_0^{-2/3} f_2(\Theta \ell_0^2), \tag{6}$$

where the lattice constant a has been set to unity. The scaling function f_2 is obtained from f_1 by replacing the proportionality in (5) by an equals sign. If we furthermore suppose that the asymptotic power laws in (5) would remain valid right to the crossover point at $\Theta \ell_0^2 = 1$ (which is certainly not the case), then the scaling function would have the additional symmetry,

$$f_2(z^{\lambda}) = f_2^{\lambda}(z)\,. \tag{7}$$

As a consequence, the system would not only be invariant under the scale transformation (1),

$$\ell_0 \to \Lambda \ell_0, \qquad \Theta \to \Lambda^{-2}\Theta, \qquad M \to \Lambda^{-2/3} M\,, \tag{8}$$

but also under

$$\ell_0 \to \ell_0^\lambda, \qquad \Theta \to \Theta^\lambda, \qquad M \to M^\lambda. \tag{9}$$

This is a scaling transformation for the *logarithms* with all critical exponents equal to 1. According to (2) this would imply

$$\ln M = (\ln \ell_0) g(\ln \Theta / \ln \ell_0). \tag{10}$$

with a piecewise linear scaling function $g(z)$ whose slopes are determined by the exponents in (5).

While in MBE this type of scaling holds only in an approximate sense, we are going to show that under certain conditions PLD indeed obeys the scaling form (10). PLD is a growth technique in which the target material is ablated by a pulsed laser and then deposited in pulses on a substrate surface, i.e., many particles arrive simultaneously at the surface [14]. Experimentally, each pulse has a length of about a few nanoseconds and the time between two pulses is of the order of seconds. It must be emphasized that the physical conditions of PLD are by far less well defined than for MBE. The particles deposited may be atoms, clusters, or even droplets. They may arrive with energies ranging from 0.1 to 1000 eV. Different theories are appropriate for various physical conditions. For example, in [15] the island statistics is studied for tin droplets deposited by PLD on a sapphire substrate, and power-law scaling is found. By contrast, our theory should apply for systems with two-dimensional islands and deposition at low energies.

In order to investigate the scaling behavior of PLD, we consider a variant of the model introduced in [16]. In contrast to that model, the duration of a pulse is assumed to be zero. The transient enhancement of the mobility of freshly deposited atoms is neglected. Our model is defined as a solid-on-solid growth model on a square lattice of $L \times L$ sites with integer heights representing the configuration of the adsorbed layer. The model is controlled by three parameters, namely, the intensity I of the pulses, the diffusion constant D, and the average flux density of incoming particles F. The dynamic rules are defined as follows. (i) In each pulse IL^2 atoms are instantaneously deposited at random positions on the surface. (ii) Between two pulses a time interval $\Delta t = I/F$ elapses, in which adatoms diffuse to neighboring sites with rate D. The Ehrlich-Schwoebel barrier is assumed to be zero, i.e., diffusion descending an edge of an island takes place at the same rate D. (iii) If two atoms at the same height occupy neighboring sites, they stick together irreversibly, forming the nucleus of a new island or leading to the growth of an already existing island. Note that due to the absence of edge diffusion the islands grow in a fractal manner before they coalesce.

If the intensity I is very low, PLD and MBE display essentially the same properties. However, if the intensity exceeds the average density of adatoms during a MBE process, we expect a crossover to a different type of behavior. This density is known to scale as $(D/F)^{2\gamma-1}$ [13], where $\gamma = 1/6$, if the islands are compact, and more generally $\gamma = 1/(4 + d_f)$, if they have the fractal dimension d_f. Thus, the crossover takes place at a critical intensity,

$$I_c = (D/F)^{2\gamma-1}. \tag{11}$$

The qualitative difference between PLD and MBE for $I > I_c$ is shown in Fig. 1. As can be seen, there are many more nucleations at an early stage, although the effective flux of incoming particles is the same in both cases.

In order to avoid the influence of the crossover at $I \approx I_c$, we restrict our PLD simulations to a particularly simple case, namely, to the limit of an infinite D/F, meaning that all adatoms nucleate or attach to an existing island before the next pulse arrives. In this limit $I_c = 0$, and the nucleation density again depends only on two variables, $n(I, \Theta)$.

Performing Monte Carlo simulations, we investigated the nucleation density for various intensities using a system size of 400×400. The measurement always takes place right before a new pulse is released. As can be seen in Fig. 2, $n(I, \Theta)$ increases with increasing intensity I. This is plausible since for a higher intensity more atoms arrive at the surface simultaneously so that more of them can meet and form new islands.

From the dashed line in Fig. 2, one sees that the nucleation densities after the first pulse, $n(I, I)$, lie on a straight line with slope 1,

$$n(I, I) = 0.35(\pm 0.01) I. \tag{12}$$

This observation can be explained as follows. After deposition of the first pulse, the adatoms diffuse until they meet and nucleate. Most of them will nucleate with another adatom and form an island consisting of two atoms. Thus, after completion of the nucleation process, the nucleation density would be $I/2$. In reality, however, some of the adatoms form bigger islands with three or more particles, but these processes lead only to a prefactor smaller than $1/2$ in (12).

At this point a remark on the system size is in order. Finite size effects have to be expected, if the pulse intensity is so small that the resulting island density is of the order

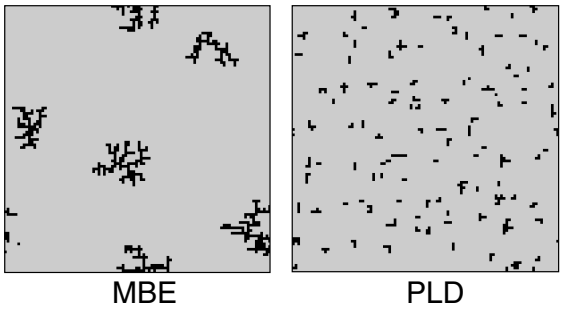

FIG. 1. Molecular beam epitaxy (left) compared to pulsed laser deposition (right) for $D/F = 10^8$ and $I = 0.01$. The figure shows typical configurations after deposition of 0.05 monolayers.

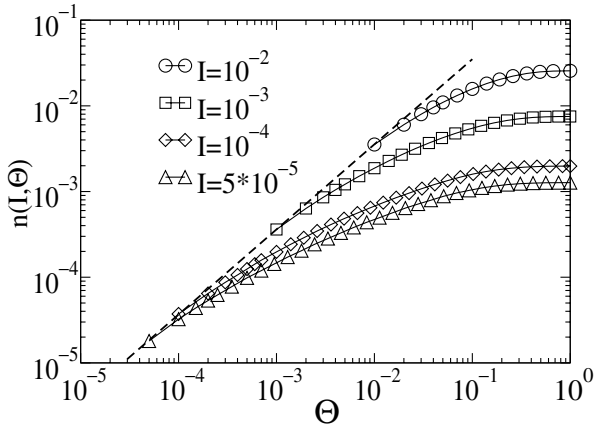

FIG. 2. The nucleation density versus time during the deposition of a monolayer. The dashed line has the slope 1.

L^{-2}. If finite size effects cannot be observed for the smallest pulse intensity I considered, one can be confident that also for larger intensities the results are accurate. For a system of size L^2, the minimal intensity which can be simulated is $I_{\min} = L^{-2}$, where one gets exactly one island in the system, independent of the coverage. Hence, for small intensities (12) cannot hold any more, as $n(I, I) \geq I_{\min}$. As we do not see any indication of the breakdown of (12) (our smallest intensity was 8 times larger than $I_{\min}$), finite size effects can be ruled out, which was also verified for other quantities [17].

Obviously, the usual scaling theory relying on power-law scaling fails to describe the data in Fig. 2. However, as shown in Fig. 3, it is possible to generate a data collapse by using the scaling form,

$$\ln M \simeq (\ln I) g(\ln\Theta / \ln I). \tag{13}$$

for the normalized nucleation density, $M(I, \Theta) = n(I, \Theta)/n(I, 1)$ [18].

The data collapse has several nontrivial implications. First, it is remarkable that the data points for the minimal

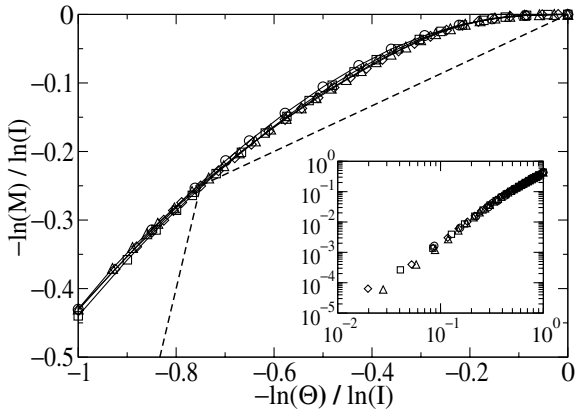

FIG. 3. Data collapse according to the scaling form (13). The inset shows a double-logarithmic plot of $\ln M/\ln I$ vs $\ln\Theta/\ln I$.

coverage $\Theta = I$ (i.e., after the first pulse) fall on top of each other in Fig. 3. This implies that

$$M(I, I) = n(I, I)/n(I, 1) = I^{\kappa}, \tag{14}$$

with an exponent $\kappa = 0.44 \pm 0.02$ and an amplitude equal to 1.

Combining (12) with (14), one concludes that also the maximum nucleation density $n(I, 1)$ must have a power-law dependence on the pulse intensity:

$$n(I, 1) \propto I^{2\nu}, \tag{15}$$

with $\nu = (1 - \kappa)/2 = 0.28 \pm 0.01$ and the same amplitude as in (12).

Equation (15) agrees with the power-law scaling of the maximum island density discussed by Jensen and Niemeyer [16], who give a slightly smaller exponent 1/4. This discrepancy can be explained by the fact that our islands are fractal. In [19] we showed that for finite D/F the island density, respectively, the nucleation density at a given coverage, obeys the following power-law scaling:

$$n(I, D/F, \Theta = 1) = (D/F)^{-2\gamma} f(I/I_c), \tag{16}$$

where I_c is given by (11). This equation describes the crossover from MBE-like behavior, $n \propto (D/F)^{-2\gamma}$, for $I \ll I_c$, to PLD-like behavior (15) for $I \gg I_c$. As in the PLD regime, the island density does not depend on D/F [16]; $I_c^{2\nu}$ must compensate the factor $(D/F)^{-2\gamma}$. Hence, $\nu = \gamma/(1 - 2\gamma) = 1/(2 + d_f)$. For compact islands ($d_f = 2$) this agrees with the value 1/4 given by Jensen and Niemeyer [16], while our ν value indicates a fractal dimension $d_f = 1.6 \pm 0.1$ in agreement with diffusion limited aggregation on a square lattice [20].

The second interesting observation concerns the crossover from PLD to MBE. As outlined before, the scaling form (13) for PLD should also hold for finite D/F provided that $I \geq I_c$. Therefore, this scaling form can be compared with the approximate scaling form for MBE (10) at the crossover point $I \simeq I_c$, where both scaling concepts should "intersect." For the sake of transparency, we give here only the derivations for the case of compact islands, $d_f = 2, \gamma = 1/6$, which is the most relevant one for comparison with experiments. Because of Eqs. (3) and (11), the crossover between the MBE and the PLD regime is characterized by the length scale $\ell_0 \sim I_c^{-3/8}$. Taking the logarithm of (10) and dividing by $\ln I_c = -(8/3)\ln\ell_0$, we obtain

$$\frac{\ln M(I_c, \Theta)}{\ln I_c} = \begin{cases} 3\frac{\ln\Theta}{\ln I_c} - 2 & \text{for } \frac{3}{4} \ll \frac{\ln\Theta}{\ln I_c} \\ \frac{\ln\Theta}{3\ln I_c} & \text{for } 0 \ll \frac{\ln\Theta}{\ln I_c} \ll \frac{3}{4}. \end{cases} \tag{17}$$

In the limit $I \simeq I_c \to 0$, the crossover between the two power laws becomes sharper so that (17) converges to a piecewise linear curve, which is shown in Fig. 3 as a dashed line. Surprisingly, the crossover point,

$$\ln\Theta_c/\ln I_c = 3/4, \qquad \ln M(I_c, \Theta_c)/\ln I_c = 1/4, \tag{18}$$

coincides within the numerical errors with the collapsed curves for PLD. This is plausible for the following reasons. First, the PLD curve must be an upper bound for the MBE curve, because the island density in PLD is always larger. Moreover, if the gap between the two extrapolated curves did not close at the crossover between PLD and MBE behavior, it would imply that there is an additional characteristic length in the system, for which we have no evidence. We did the calculation also for a general γ. The quality of the coincidence between the two curves at the crossover point does not significantly improve for this more accurate theory.

The third remarkable result is that the scaling function in Fig. 3 is approximately a power law,

$$g(z) \approx Az^{\beta}, \tag{19}$$

where $A = 1 - 2\nu$ due to (12), (14), and (15) and, hence, is determined by γ. The exponent β is also determined by γ, if one demands that the function (19) goes through the point (18) modified according to the actual value of γ. The result is a β value between 2.0 and 2.4. The log-log plot of the scaling function g is given in the inset of Fig. 3 and shows that the power law is only approximately fulfilled with an exponent of about 2 for small coverage and 2.4 for large coverage.

In summary, we have demonstrated that a simple model for pulsed laser deposition displays an unusual type of scaling behavior. In its most general setup, this kind of scaling behavior is observed in systems which are invariant under the transformation,

$$M \to M^{\lambda^{\alpha}}, \qquad z_i \to z_i^{\lambda^{\beta_i}}, \tag{20}$$

for arbitrary λ, where α and $\beta_1, \ldots, \beta_n$ are certain exponents. The corresponding scaling form reads

$$\ln M \sim (\ln z_1)^{\alpha} g\left(\frac{\ln z_2}{(\ln z_1)^{\beta_2}}, \ldots, \frac{\ln z_n}{(\ln z_1)^{\beta_n}}\right). \tag{21}$$

Performing numerical simulations, we have demonstrated that such a scaling form leads to a convincing data collapse for PLD in the limit of an infinite diffusion constant. In contrast to ordinary scaling functions, the function g is defined on a limited interval between two points.

The scaling forms (13) and (16) are limiting cases of a unified scaling theory for $n(I, D/F, \Theta)$ yet to be developed. Compatibility indicates that (13) is related to logarithmic corrections of (16), but we do not have a convincing physical explanation for them. Correspondingly, it is not yet clear to what extent the scaling function g is universal, i.e., independent of details of the dynamic rules of the model.

We thank the Deutsche Forschungsgemeinschaft for support within SFB 491.

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