Scaling of the nucleation density for pulsed layer deposition

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We discuss the logarithmic scaling of the nucleation density for pulsed laser deposition, discovered recently by Hinnemann *et al.* [Phys. Rev. Lett. **87**, 135701 (2001)] in two dimensions. The logarithmic scaling is often observed in the upper critical dimension. We find that the nucleation density in one dimension also exhibits logarithmic scaling, implying that it is not a prerequiste for the upper critical dimension. The normalized island density also scales similarly both in one and two dimensions when plotted against the normalized coverage.

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In usual critical phenomena, many physical quantities are assumed to be generalized homogeneous functions satisfying

$$f(\lambda^{a_x}x,\lambda^{a_y}y)=\lambda f(x,y), \tag{1}$$

where $f(x,y)$ is an observable depending on parameters x and y. Then, the function $f(x,y)$ scaled by y^α can be expressed in terms of a single variable $z\equiv x/y^\beta$, i.e.,

$$f(x,y)=y^\alpha g(x/y^\beta), \tag{2}$$

where $\alpha=1/a_y$ and $\beta=a_x/a_y$ [1]. This implies that the scaled data of $f(x,y)$ by y^α plotted against a scaled variable z collapse onto a single curve, i.e., data scale with the scaling function $g(z)$.

Recently, it has been reported that the nucleation density created on a two-dimensional substrate by pulsed laser deposition (PLD) exhibits a scaling in an unusual way [2]. The PLD is an alternative technique of fabricating thin films [3,4], different from the molecular beam epitaxy (MBE) [5,6]. The main difference between PLD and MBE is that a particle beam deposits as a pulse of intensity I (atoms per second), i.e., many particles arrive on a substrate simultaneously in PLD, while deposition occurs with a steady-state flux F (atoms per second per unit area) in MBE. In both cases, adatoms diffuse onto a substrate with a diffusion constant D until they encounter another adatom, in which case they form a stable nucleus, or until they attach irreversibly to the edge of an existing island.

The nucleation density n, defined by the number of nucleation events per unit area accumulated with time up to coverage Θ $(=Ft)$, is a function of I and Θ in PLD, i.e., $n(I,\Theta)$. A similar but alternative quantity frequently studied in MBE is the island density $m(I,\Theta)$, defined by the number of islands per unit area. Apparently, the island density increases in the early-time regime, becomes nearly constant in the aggregation regime where adatoms aggregate to the existing islands, and eventually decreases in the coalescence regime in which two or more islands merge and form one larger island. The scaling of an island density in MBE is known to hold only in the limited range of coverage [7], with the standard power-law scalings similar to that described in Eq. (2). On the other hand, the nucleation density increases monotonically with time and saturates when the first monolayer is completed.

The nucleation density calculated up to a coverage of 1 ML was found to hold the scaling relation in a following way. Suppose $N(I,\Theta)$ is the normalized nucleation density by the density at $\Theta=1$ ML, i.e., $N(I,\Theta)=n(I,\Theta)/n(I,1)$. Then, the scaling relation

$$\ln N\sim(\ln I)g(\ln\Theta/\ln I) \tag{3}$$

holds with the piecewise scaling function $g(z)$, which is different from the usual power-law scaling function in Eq. (2). The "logarithmic" scaling relation similar to that in Eq. (3) is rarely observed in regular dimensions, but occasionally holds in the upper critical dimension, above which the local fluctuation of density can be neglected. If the scaling function in Eq. (3) is indeed the one that holds in the upper critical dimension, it would be expected that a different type of scaling relation may hold in a lower dimension.

Since the logarithmic scaling relation was found in two dimensions, one may regard the dimension 2 as the upper critical dimension for PLD. One can then expect the usual power-law scaling to hold in one dimension. It is the purpose of the present work to closely examine the scaling of the nucleation density in one dimension. The logarithmic scaling as well as the power-law scaling is investigated and the scaling of the data in two dimensions is also investigated in different respect.

The scaling relation in Eq. (3) can be explained as follows. From the definition of $N(I,\Theta)$, it is clear that $N(I,1)=1$ for all I. Thus, the data of $\ln N(I,\Theta)$ for various values of I fall on the same point and become 0 at the right end point of the data, i.e., at $\Theta=1$ ML. It was also found, by Monte Carlo simulation, that the data of $n(I,\Theta)$ at the minimal coverage of $\Theta=I$ (after deposition of the first pulse) exhibits a power-law behavior with the power of 1 when plotted against I, i.e., $n(I,I)\sim I$. Since the saturated nucleation density $n(I,1)$ scales as $n(I,1)\sim I^{2\nu}$, with $\nu=\frac{1}{4}$ for the compact islands [8] and $\nu\simeq0.28$ for the fractal islands [2], it is expected that the quantity $\ln N/\ln I$ plotted against the variable $\ln\Theta/\ln I$ yields the data at the minimum coverage, i.e., at the left end point of the data, collapsing onto the point $(1, 1-2\nu)$, irrespective of the values of I. The data at the right end point still collapse onto the same point, i.e., onto (0, 0). This prescription thus makes the data for $\ln N/\ln I$ on each end point collapse when plotted against $\ln\Theta/\ln I$. Then, the

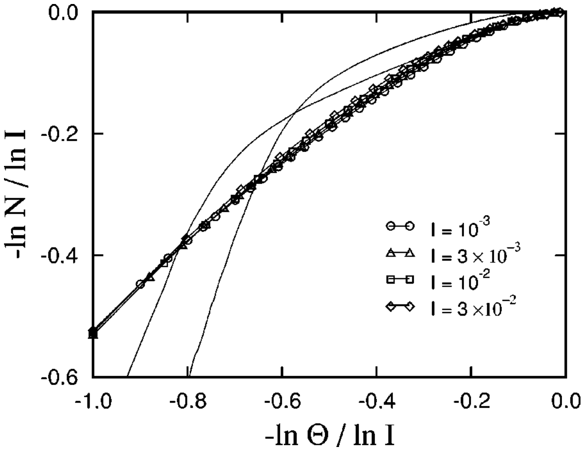

FIG. 1. The scaling function of Eq. (3) for the nucleation density for $D/F=10^{10}$ in one dimension. The solid lines are for $I=10^{-5}$ (right) and $I=10^{-4}$ (left), both of which are out of the PLD regime.

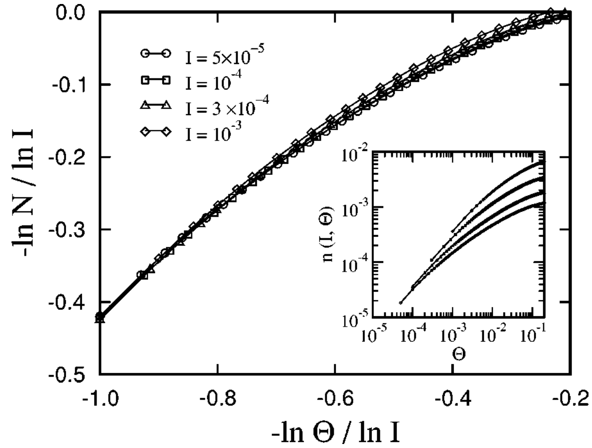

FIG. 2. The scaling function of Eq. (3) for the nucleation density, normalized by the density at $\Theta=0.2$ ML, for $D/F=\infty$ in two dimensions. The data in the inset are the unscaled data.

stretched data between the two end points for various values of I collapse onto the single curve.

The nucleation density was calculated in one dimension, essentially by the same technique as in Ref. [2] for various values of I and D/F, all for a system of size $L=10^5$ (in units of lattice constant). The summary of our findings is as follows. The data of $n(I,\Theta)$ for different values of D/F were not appreciably different, implying that $n(I,\Theta)$ is not a function of D/F, as long as D/F is not too small. The saturated nucleation density at $\Theta=1$ ML scaled as $n(I,1)\sim I^{2\nu}$ with $2\nu=0.4705$, which is slightly smaller than that in two dimensions. The nucleation density at the minimal coverage exhibited a power-law behavior $n(I,I)\sim I^{\mu}$ with $\mu=1.0015$ when plotted against Θ, similar to the case in two dimensions. These observations suggest that a similar logarithmic scaling might hold in one dimension as well.

Data for the nucleation density for $D/F=10^{10}$ in one dimension scaled by the same way as in two dimensions are plotted in Fig. 1. The solid lines are for $I=10^{-5}$ (right) and $I=10^{-4}$ (left), both of which are considered to be out of the PLD scaling region. (Note that for $I=10^{-5}$ a single atom arrives on the substrate during each pulse and the resulting nucleation density is expected to be similar to that for MBE.) Surprisingly, all data points fall on the same curve, implying that the nucleation density in one dimension scales in the same way as in two dimensions. In fact, the data in one dimension are not very different from those in two dimensions. This suggests that the logarithmic scaling might not be a sign of the upper critical dimension for the case of PLD. It is worth noting that the results in MBE regime cross the PLD data and become larger as the coverage increases. A similar observation in two dimensions confirms that the limiting scaling relation in the MBE regime presented in Ref. [2] does not appear to be correct.

The nucleation density increases in time until the first layer is completed. Since the nucleation events may occur frequently in PLD, it is expected that the second layer begins to grow before the first layer is completed. Therefore, $n(I,\Theta)$ increases even beyond $\Theta=1$ ML, though the rate of increase may be negligible. This implies that the coverage $\Theta=1$ ML may not be a special coverage in PLD. One may then raise a question of whether or not the nucleation density normalized by the density at an arbitrary coverage other than 1 ML still satisfies a similar scaling relation. Suppose that one calculates the nucleation density up to $\Theta=\Theta_{\max}$ <1 ML. Then, one can modify the normalized nucleation density as $N(I,\Theta)=n(I,\Theta)/n(I,\Theta_{\max})$. With the same prescription as described earlier, one can approximately make data on each end point fall on the same point. It is desirable to check whether or not the data in between also scale.

Results for $\Theta_{\max}=0.2$ ML in two dimensions are plotted in Fig. 2. Data for $I=5\times10^{-5}$, 10^{-4}, and 3×10^{-4} exhibit good collapsing, whereas those for $I=10^{-3}$ are slightly deviated from other sets, implying that the scaling region becomes narrower for this particular scaling. (It should be noted, however, that the data for $\Theta>\Theta_{\max}$ normalized in this way do not collapse, if plotted, because the right end points do not fall on the same point.) Similar data collapsing was also found in one dimension.

We have also calculated the island density $m(I,\Theta)$ for PLD. The island density increases monotonically up to the coverage Θ_c where islands begin to coalesce and decrease sharply beyond it. The unscaled data of $m(I,\Theta)$ are plotted in the inset of Fig. 3, with $\Theta_c=0.6$ marked by a dashed line. Now, suppose that we calculate the island density up to Θ_c. One can normalize the island density in a way similar to the nucleation density as $M(I,\Theta)=m(I,\Theta)/m(I,\Theta_c)$. It is interesting to investigate whether or not similar logarithmic scaling holds for $M(I,\Theta)$. The Monte Carlo data for different values of I scatter and the scaling relation does not hold (not shown). We, however, found that they fall on the same curve when plotted against the normalized coverage $\Lambda=\Theta/\Theta_c$, as we can see in Fig. 3. The quality of data collapsing is better than the quality for the nucleation density.

We have also examined the scaling for both the nucleation density and the island density in various other ways. When both D/F and I are not too small, most atoms either nucleate or aggregate to existing islands before the next pulse is deposited. Thus the data for both $n(\Theta,I)$ and $m(\Theta,I)$ do not depend on the values of D/F. The scaling relation in Eq. (3)

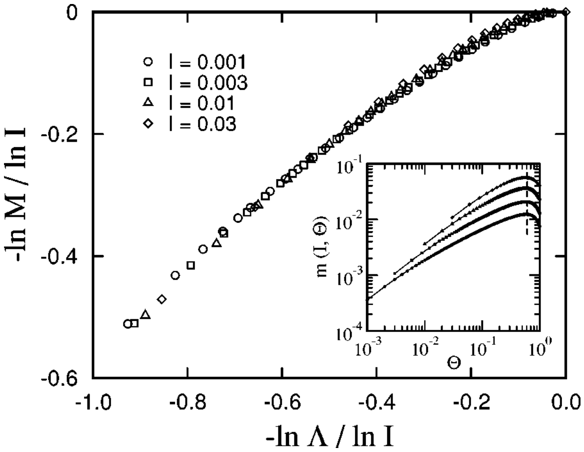

FIG. 3. The normalized island density plotted against the normalized coverage, calculated in one dimension for $D/F=10^{10}$ and various values of the pulse intensity. The data in the inset are the unscaled data, and the dashed line in it indicates the coverage for the maximal density.

holds in this ''PLD scaling regime.'' On the other hand, if I is sufficiently small, i.e., $I \ll I_c \sim (D/F)^{2\gamma-1}$ with $\gamma = 1/(4+d)$ [2,9], PLD displays essentially the same properties as MBE. The nucleation density is thus a function of three variables, i.e., $n(\Theta, I, D/F)$ in the ''crossover regime'' where $I \simeq I_c$ holds. The scaling of the crossover from PLD to MBE was discussed in Ref. [2] for a particular case of compact islands. The numerical investigation of such a scaling in a finite-size system is much complex and is beyond the scope of this work. We thus leave it for a future work. The data for the solid curves in Fig. 1 fall in this regime.

The scaling of the nucleation density in the PLD scaling regime was also closely examined in two extreme limits of the coverage. We found that, in the low-coverage regime, the scaling relation $n(I,\Theta) = I f_1(\Theta/I)$ appeared to hold within the limited range of Θ. In the high coverage regime, on the other hand, the nucleation density scaled as $n(I,\Theta) \sim I^{2\nu} f_2(\Theta)$. These scaling relations are not new and were already considered in deriving Eq. (3). We however were not able to make data for various values of I collapse onto a single curve over the entire range of coverage by a method other than the logarithmic scaling. Thus, Eq. (3) seems to be the only scaling function for both the nucleation density and the island density. Such a dimension-independent logarithmic scaling function is rare and unusual in the critical phenomena. The origin of such a logarithmic scaling is yet to be uncovered.

In summary, the scaling of the nucleation density and island density for PLD was examined in both one and two dimensions. It was found that data exhibited a logarithmic scaling in both dimensions, indicating that the logarithmic scaling is not a prerequisite for the upper critical dimension for PLD. The nucleation density normalized by the density at an arbitrary coverage was also found to collapse onto a single curve. The normalized island density by the maximal density also exhibited a similar logarithmic scaling when plotted against the normalized coverage. The judgment of whether it is indeed a scaling or simply an accidental coincidence of data points by a certain prescription should be left to the readers.

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