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Irreversible island nucleation and growth with anomalous diffusion in $d > 2$

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HIGHLIGHTS

- Island growth with anomalous diffusion is studied in $d = 2, 3$, and 4 dimensions.
- Both the case of subdiffusion and superdiffusion are studied.
- Good agreement is obtained with a recently developed general rate-equation theory.
- Results are presented for the scaled capture-number distribution in $d = 2$.
- Results for the scaled island-size distribution in $d = 2$ are also presented.

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ABSTRACT

Recently, there has been significant interest in the effects of anomalous diffusion on island nucleation and growth. Of particular interest are the exponents χ and χ_1 which describe the dependence of the island and monomer density on deposition rate as well as the dependence of these exponents on the anomalous diffusion exponent μ and critical island size. While most simulations have been focused on growth on a 2D and/or quasi-1D substrate, here we present simulation results for the irreversible growth of ramified islands in three-dimensions ($d = 3$) for both the case of subdiffusion ($\mu < 1$) and superdiffusion ($1 < \mu \leq 2$). Good agreement is found in both cases with a recently developed theory (Amar and Semaan, 2016) which takes into account the critical island-size i, island fractal dimension d_f, substrate dimension d, and diffusion exponent μ. In addition, we confirm that in this case the critical value of μ is given by the general prediction $\mu_c = 2/d = 2/3$. We also present results for the irreversible growth of point-islands in $d = 3$ and $d = 4$ for both monomer subdiffusion and superdiffusion, and good agreement with RE predictions is also obtained. In addition, our results confirm that for point-islands with $d \geq 3$ one has $\mu_c = 1$ rather than $2/d$. Results for the scaling of the capture-number distribution (CND), island-size distribution (ISD), and average capture number for the case of irreversible growth with monomer superdiffusion in $d = 2$ are also presented. Surprisingly, we find that both the scaled ISD and CND depend very weakly on the monomer diffusion exponent μ. These results indicate that – in contrast to the scaling of the average capture number which depends on the monomer diffusion exponent μ – both the scaled ISD and CND are primarily determined by the capture-zone distribution, which depends primarily on the "history" of the nucleation process rather than the detailed mechanisms for monomer diffusion.

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1. Introduction

Recently, there has been significant interest in the effects of anomalous monomer diffusion [1–14] including its effects on submonolayer island growth [15–20]. This interest has been partially stimulated by recent experiments [16–18] in which values of the exponent χ which describes the dependence of the peak island density N_{pk} on deposition rate F (e.g. $N_{pk} \sim F^{\chi}$) were obtained which were significantly larger than 1. This result is in contrast to the standard rate-equation (RE) theory prediction [21,22] that in the case of deposition on a 2D substrate with ordinary monomer diffusion – and assuming the existence of a critical cluster size i such that clusters larger than i are stable while clusters of size i and below are unstable – one has $\chi = i/(i+2)$.

While this result applies in the case of ordinary diffusion such that the dependence of the mean-square monomer displacement on time t satisfies $\langle r^2(t)\rangle \sim t^{\mu}$ with $\mu = 1$, it does not apply in the case of anomalous diffusion. In particular, in Ref. [17] it was suggested that the large value of the exponent χ found in the case of submonolayer growth of para-hexaphenyl [16] on amorphous mica may be explained by the existence of transient hyperthermal behavior which leads to ballistic monomer diffusion ($\mu = 2$). Similar results, e.g. $\chi > 1$, have also been obtained in the case of submonolayer growth of pentacene on amorphous mica [18]. In addition, in Ref. [19] a RE approach was used to show that for the case of compact islands on a 2D substrate, ballistic diffusion implies that $\chi = 2i/(i+3)$.

In order to obtain a better understanding of the effects of anomalous diffusion on island nucleation and growth, a rate-equation (RE) theory has recently been developed [20] which leads to general expressions for the exponent χ as a function of the critical island size i, substrate dimension d, island fractal dimension d_f, and diffusion exponent μ, where $0 \le \mu \le 2$. General expressions were also obtained [20] for the exponent χ_1 which describes the dependence of the monomer density N_1 on deposition rate at fixed coverage θ (e.g. $N_1(\theta; F) \sim F^{\chi_1}$) in the pre-coalescence or aggregation regime in which the island density remains constant. Here θ is an effective coverage which is equal to the fraction of the convex envelope surrounding stable islands, which may be approximated as $\theta \simeq N(\frac{\phi}{N})^{d/d_f}$ where N is the stable island density and the dose $\phi = Ft$ corresponds to the equivalent coverage if all deposited particles are placed on the d-dimensional substrate.

General expressions were also obtained [20] for the exponents χ' and χ_1' which describe the deposition-rate dependence of the island and monomer densities at fixed dose $\phi = Ft$, e.g. $N(\phi; F) \sim F^{\chi'}$ and $N_1(\phi; F) \sim F^{\chi_1'}$. We note that ϕ is equal to the coverage θ if the island fractal dimension d_f is equal to the substrate dimension d. In addition, two distinct cases were identified – one corresponding to $\mu < \mu_c$ and the other corresponding to $\mu > \mu_c$ – where $\mu_c = 2/d$ for finite d_f as well as for point-islands with $d \le 2$, while $\mu_c = 1$ for point-islands with $d \ge 3$. However, these theoretical predictions have only been tested in the case of growth on a 2D substrate ($d = 2$) [20,23] and or quasi-1D substrate ($d = 1$) [24].

Here we consider the effects of anomalous diffusion on irreversible island nucleation in higher dimensions, e.g. $d = 3$ and $d = 4$ and compare with the predictions of Ref. [20]. We note that the case of irreversible nucleation in $d = 3$ with normal diffusion has been previously studied in Ref. [25]. In particular, results are presented for both ramified islands ($d_f \simeq 2.5$) and point-islands ($d_f = \infty$) in $d = 3$ for the case of both monomer subdiffusion ($\mu < 1$) and superdiffusion ($1 < \mu \le 1.5$). In general, excellent agreement is found between our simulation results for the exponents $\chi(\mu)$, $\chi'(\mu)$ and $\chi_1'(\mu)$ in $d = 3$ and the RE predictions. We also find good agreement for the case of point-islands in $d = 4$ between our simulation results for the exponent $\chi'(\mu)$ and RE predictions for the case of subdiffusion ($\mu < 1$). Our results also confirm that for islands with finite d_f the critical value of μ is given by the general prediction $\mu_c = 2/d$, e.g. $\mu_c = 2/3$ for $d = 3$, while for point-islands $\mu_c = 1$.

In addition to these results for $d \ge 3$ - since previous work for the case of substrate dimension $d = 2$ with anomalous monomer diffusion has focused almost exclusively on the average island and monomer densities — we also present results for the dependence of the scaled island-size and capture-number distributions on the monomer diffusion exponent μ for the case of superdiffusion with irreversible growth in $d = 2$. Somewhat surprisingly, we find that in this case the scaled capture number distribution (CND) does not depend on μ. This result also explains the relatively weak dependence of the scaled island-size distribution (ISD) on the value of μ found in our simulations in the case of irreversible growth. It also indicates that – in contrast to the scaling of the average capture number which depends on the monomer diffusion exponent μ – the capture number distribution is primarily determined by the capture-zone distribution [26–32], which depends primarily on the "history" of the nucleation process and not on the detailed mechanisms for monomer diffusion. We also present results for the scaling of the average capture number σ_{av} at fixed coverage as a function of island density for the case of ballistic diffusion ($\mu = 2$) which directly confirm the assumptions made in the analyses of Ref. [19] and Ref. [20].

This paper is organized as follows. In Section 2 we first briefly review the RE theory, while in Section 3 we discuss the details of our simulations. We then present our simulation results in Section 4 and compare with the corresponding RE theory predictions. Finally, in Section 5, we present our conclusions and discuss possible future work.

2. Rate-equation theory

By combining the steady-state assumption that in the aggregation regime the rate of monomer deposition is balanced by the rate of island attachment with an expression for the average monomer lifetime as a function of the island density N and diffusion exponent μ it was found [20] that the average capture number σ_{av} at fixed coverage θ (dose ϕ) is proportional to