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Kinetic roughening in polymer thin film growth

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Abstract

Kinetic roughening is revisited in the context of polymer thin film growth by vapor deposition using the simplest toy model of chemical vapor deposition polymerization (CVDP) growth, which was employed in [J. Stat. Mech. (2009) P02031]. As the ratio of monomer diffusion rate D to the deposition flux F of monomers ($G = D/F$) increases, dynamic scaling of the CVDP growth is investigated in (1+1) and (2+1) dimensions. Measuring the surface width (height fluctuations) and the q-th order moments of height-height correlation function, it is observed that anomalous scaling behavior and multifractality exist. In order to speculate the origin of such anomalies, the following two scenarios are suggested and tested: One is that the cosine flux of incident monomers (random angle deposition) is essential in anomalous kinetic roughening phenomena and the other is that the multi-affine structure of CVDP growing surfaces is attributed to the non-local shadowing effect caused by the cosine flux of incident monomers, which yields the power-law distribution of steps. Based on the comparison of the ballistic deposition model with some modification of noise, namely the power-law distribution of noise, it is numerically confirmed that two scenarios turn to be true. This implies that dynamic scaling of the CVDP growth depends not only on what value is used for the ratio of monomer diffusion rate to deposition flux (G) but also on what kind of intrinsic and extrinsic noise exists.

Keywords: kinetic roughening, thin film growth, chemical vapor deposition polymerization, anomalous scaling, multifractality, cosine flux, shadowing effect, ballistic deposition, noise
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1. Introduction

In the growth of thin films and multilayers, kinetic roughening and dynamic scaling have been of considerable interest [1, 2] by the demand for smooth or regularly structured surfaces and interfaces for miniaturized functional films in science and technology. There are lots of theoretical and experimental studies available for the growth of metal/semiconductor films by molecular beam epitaxy (MBE), which reveal the occurrence of scaling exponents corresponding to a few universality classes with minimal toy models and reasonable theories. While polymer thin films and growing mechanisms become technologically important in many aspects, such as molecular devices and microelectronic interconnects [3], only few systematic studies are available as pioneering works [4, 5, 6]. Chemical vapor deposition polymerization (CVDP) is one of the popular and useful techniques for polymer thin film growth since it well describes the process of coating with poly (p-xylylene)(PPX), also known by the trade name Parylene [7].

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The growth mechanism of CVDP is quite distinguishable from that of the ordinary physical vapor deposition (PVD), in the sense that a deposited monomer cannot be stable until it finds either an active end of the already formed polymers or another monomer to be an oligomer. While the ratio of monomer diffusion rate (coefficient) D to the deposition flux F of incident monomer depends on temperature in the corresponding experimental setup, the basic mechanism of CVPD are more or less universal as follows: A hot monomer in the gas phase deposits by a random angle (kind of random walk) on the cold substrate, reacts to form a high molecular weight as an oligomer (the basic unit of polymer), and becomes a part of the polymer consisting of the body (bulk) part and two active ends (boundaries). In order to mimic the CVDP growth process, some modification is needed in the well-known MBE-type growth model, so-called the ballistic deposition (BD) model [1, 8]. Kinetic roughening in (d+1) dimensions, where d is the substrate dimension. The main purpose of this paper is to provide a guideline for the CVDP growth model study as well as some explanation of recent experimental data from the growth of polymer thin films in (2+1) dimensions.

This paper is organized as follows: Section 2 describes the minimal toy model of CVDP growth in $d = 1, 2$ and define major physical quantities to investigate kinetic roughening and dynamic scaling. In section 3, updated numerical results are presented as well as the brief review of the earlier model studies in (1+1) dimensions. Finally, the physical origin of anomalous scaling behaviors and multifractality is discussed, compared with the modification of the BD model in section 4 with closing remarks and outlook for (2+1) dimensional CVDP growth in both model studies and experiments.

2. Model

A simple toy for CVDP thin film growth was first proposed by Bowie and Zhao [9] in a (1+1)-dimensional lattice with L sites, where periodic boundary conditions are employed in a spatial direction, x. Later, the coalescence process of polymers was added to the original model in [10]. During the CVDP process, each monomer transports in the vacuum, which is very similar to the conventional PVD/MBE growth process for the metal/semiconductor thin film growth [1]. One big difference between two processes occurs in the nucleation and cluster/polymer propagation after the hot monomer is condensed either on the cold substrate or on the film surface. While in the PVD/MBE process each monomer is stable once it attaches to the nearest neighbors of any nucleated sites, so that the films get dense and compact as monomer diffusion increases, in the CVPD process it becomes stable only either when it reaches one of two active ends of a polymer chain or when it meets another monomer, so that the film gets rough as the monomer-diffusion rate increases since it occurs along polymer bodies. Moreover, surface diffusion, edge diffusion, step barrier effect are relevant to the PVD/MBE case, but intermolecular interaction and chain relaxation are more relevant to in the CVDP case besides monomer diffusion. Summing up major and minor differences, some distinct dynamic behaviors must exist in the CVDP film morphology.

Dynamic rules and updates

For simplicity, the chain relaxation of polymers is omitted and only the following five processes are taken as studied in [10] (see also Figure 1).

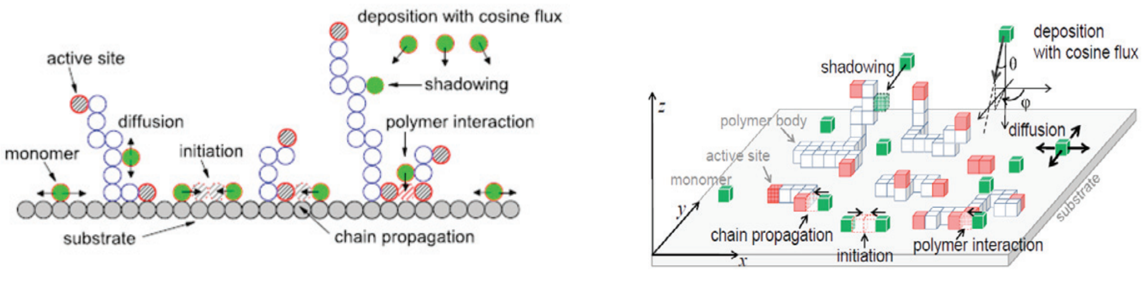

Figure 1: Five dynamic rules are illustrated as solid/green symbols (○ in the left panel for $d = 1$ and □ in the right panel for $d = 2$) for monomers, open symbols with thin lines for polymer bodies, and patterned/red symbols with thick lines for active ends.

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