Non-equilibrium phase transition between bulk structures in ballistic-diffusive stochastic models of thin film growth

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The modelling of thin solid films growth by different deposition techniques has been a very active research field in the last decades driven by the variety and increasing relevance of its technological applications to fields such as microelectronics, optics, chemistry or biology[1 2]. From a theoretical perspective, thin film growth dynamics exhibits very interesting and challenging behaviors which are related to key topics on nonlinear and statistical physics, like critical phenomena and universality. Not surprisingly, the development of film growth modelling has run linked to the advances on the study of far-from-equilibrium growth processes, fractal growth and interface physics[3 4].

The simplest atomistic stochastic models for thin film formation adopt drastic approximations to the deposition and growth processes in order to achieve computational efficiency. These approximations include the deposition of the species on the substrate via ballistic trajectories and adatom interactions limited to first nearest neighbors. Despite — or arguably owing to — these approximations, simple atomistic models have proved a suitable tool for studying different aspects of thin solid films growth regimes and morphologies and the growth parameters that determine them. In particular, special attention has been paid to the possible universality of the kinetic roughening of the growing film surfaces and the existence of non-equilibrium phase transitions between different surface growth regimes[5]. Instead much less attention has been paid to the behavior of film bulk properties and its interplay with surface evolution[6], probably by assuming that bulk properties highly depend on the system characteristics and require details which simple atomistic models do not include. However, one of the more universal behaviors of film growth shown theoretically and experimentally is the existence of diverse characteristic mesoscopic morphologies, with different surface and bulk properties, as a consequence of the competition between the spatially disordered deposition of particles on the growing film surface and the ordering effect of activated particle mobility processes. These characteristic morphologies have been summarized on successive qualitative empirical models known as *Structure Zone Models* (SZM). The first structure zone model, introduced by Movchan and Demchishin for physical vapor deposition under simple experimental conditions[7], included three characteristic morphology zones obtained by varying the main deposition parameters: the deposition rate and the substrate temperature. Progressively, new structure zones have been incorporated into the original model as different experimental parameters have been introduced. Statistics of the microstructural arrangements of particles are used to characterize the film morphologies and their correspondence with experimental structure zones, treating the dependence of the diverse film morphologies with deposition parameters as a physical or structural transition[8 9 10].

In this context, we have developed an atomistic stochastic model with ballistic deposition and thermally activated surface diffusion for study the transition between the growth regimes corresponding to the zones I and II of the simplest SZM for thin films growth from physical vapor deposition. The model incorporates a novel approach to the microstructure representation which can reproduce the simple lattice microstructures used frequently among the literature, either in (1+1) and (2+1) dimensions, as well as more complex microstructures which can develop lattice frustration and texture competition effects. The results obtained with our model provide the first evidence that the transition between the structure zones I and II can be considered as a true non-equilibrium phase transition between a porous, low density bulk morphology with self-affine surface and a compact, high density bulk morphology with a columnar shape surface.

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