Kinetic Models of Thin Film Growth with Nonmonotonic Roughness Evolution

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Thin film deposition on weakly interacting substrates exhibits a unique growth mode characterized by initial formation of multilayer islands and rapidly increasing roughness, which reaches a maximum and subsequently decreases as the islands coalesce and the film returns to a smooth morphology. This rough-to-smooth growth mode was experimentally shown in CdTe films, perovskite films, and organic films with molecules of various geometries [1,2]. We first propose a geometrical model that captures the basic mechanisms of island growth, coalescence, and formation of a continuous film [2]. The model represents the morphological evolution of the rough-to-smooth mode and, with fine tuning of a small number of parameters, it provides an accurate description of the experimental data of ultrathin films (¡30 nm of thickness) of two organic molecules [effectively spherical buckminsterfullerene (C) and the disk-like 1,4,5,8,9,11-hexaazatriphenylenehexacarbonitrile (HATCN)]. This modeling can distinguish, for instance, systems in which Ehrlich-Schwoebel energy barriers are relevant (HATCN) or not (C) for the molecules to execute interlayer diffusion. Additionally, kinetic Monte Carlo simulations of a model with minimal ingredients demonstrate that this growth mode generally occurs for weakly interacting substrates, in which surface diffusion of molecules on the substrate is significantly faster than their diffusion on the upper film layers [2]. The simulation results that fit the experimental data provide estimates of activation energies for diffusion of the adsorbed molecules of C and HATCN. Both models highlight the generic nature of the phenomenon, independently of the details of the interactions and of the molecular flux. These findings reinforce the need of statistical physics approaches to understand materials growth and open up a path for experimental control of nanoscale film roughness. Along these lines, recent simulations show that a sudden increase of the growth temperature is more effective to fasten the film smoothening if that increase is implemented when the deposit has the maximal roughness.

References:

[1] T. B. T. To, R. Almeida, S. O. Ferreira, and F. D. A. Aarão Reis. Appl. Surf. Sci. 560, 149946 (2021).

[2] D. Lapkin, I. S. S. Carrasco, C. C. Luukkonen, O. Konovalov, A. Hinderhofer, F. Schreiber, F. D. A. Aarão Reis, and Martin Oettel. Phys. Rev. Lett. 136, 076202 (2026).