Novel control strategies in molecular thin film deposition: from tailoring crystallinity and morphology to patterning of films

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Crystal structure and resulting electronic and optical properties of materials and thin films can increasingly be predicted by theory. However, despite advances in unraveling the mechanistic details of the thin film growth process, film properties such as morphology, grain size and defect density are rarely modelled before film deposition. To take our understanding beyond a recipe-based perspective to that of sound fundamental understanding we use real-time X-ray observation of growth processes (using growth oscillations and small angle scattering) to understand the atomic scale processes. We derive the energy barriers for diffusion and step edge crossing from X-ray data for examples of organic semiconductor thin films (C60 and PTCDI-C₈ molecules). Using this detailed understanding of growth processes we demonstrate different strategies to tailor film growth. We will demonstrate how fast temperature modulation instead of a constant substrate temperature can be used to tailor the nucleation phase of each successive monolayer to achieve smoother film growth. As another new control parameter in molecular film growth we show that light can be used to increase phase purity in organic thin films. We will further demonstrate how polarized light can be used for photo-alignment during molecular thin film growth by addressing specific anisotropic absorption bands, such as the lower Davydov component of tetracene films. The illumination allows us to achieve photo-alignment of the otherwise randomly oriented polycrystalline tetracene films and also enables patterning of films, e.g. for photonic meta-films or electronic applications.