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Crystalline self-stratification in polymer thin films

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The orientation of molecules within thin films is of critical importance to the emerging field of organic electronics. Particularly in the case of solution processable polymers and small molecules, where alkyl side chains, included for solubility, impede conduction along that molecular direction, understanding and controlling the molecular orientation both at surfaces and in the bulk of thin films is increasingly important to further increase electronic performance. Grazing Incidence Wide Angle X-ray Scattering has been widely used to look at the orientation of crystallites within films, but a capability which has not been widely used is its potential to characterize the depth within films at which different kinds of molecular stacking occur. Using very fine control over the angle of incidence of the X-ray beam, we observe a distinct segregation of edge-on crystallinity in a film of the polymer PNDI-SVS which otherwise stacks in a highly face-on orientation. Using simulations of the X-ray Electric Field Intensity within the film, the angular variation of scattering intensity can be matched, resulting in the conclusion that the surface region extends 9 nm into the 72 nm film.

During the spin-coating deposition process, a face-on orientation is initially observed, likely the result of preaggregation in solution in combination with a relatively fast-drying solvent. The stratified morphology is produced by annealing the film for a brief time, while upon further annealing, the bulk of the film eventually reorients to become edge-on, suggesting that the stratification is a non-equilibrium, kinetically-trapped state. With brief annealing, only the surface region of the film has time to reorient to the ultimately lower energy edge-on orientation. The time and temperature of this reorganization can reveal the difference in energetics at different depths within the film, illustrating how grazing incidence scattering can open up the possibility of examining thin films in novel and important ways.

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