Tailoring Crystalline Order in Organic Thin-Film Transistors by Exploring Interactions at Interfaces

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Electrical properties of organic crystalline films are determined by their nano- and microstructure, with a high degree of order, short intermolecular distances and parallel planar molecular orientations encouraging a good overlap between the π-orbitals and a superior charge carrier mobility. However, achieving long-range order in the active semiconducting layers of organic electronic devices is challenging. Film formation using solution deposition methods is a complex process, determined by many processing factors which can affect molecular orientation. We show that the microstructure in organic thin-film transistors can be controlled by manipulating halogen-halogen interactions between the organic semiconductor and the self-assembled monolayers (SAMs) present at contact and dielectric surfaces. We selectively choose the surface treatments to introduce targeted interactions during deposition of the organic films, and to isolate key effects behind microstructure formation. We demonstrate that the halogen interactions can template and drive the self-assembly of the organic semiconductor molecules along specific growth fronts and the strength of this interaction governs molecular alignment within the organic film. As a result, we can precisely control the preferential molecular orientation on surfaces and tune the charge carrier mobility from $10^{-6}$ cm$^2$/Vs to 1 cm$^2$/Vs in the same material. The effect of SAM presence on thin-film transistor properties such as mobility, contact resistance, interfacial trap-density and threshold voltage will be discussed, and the results will be correlated with structural data obtained from X-ray diffraction studies.

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