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**Engineering Molecular Order for Enhanced Stability of Organic Solar Cells** ANNE GUILBERT, Department of Chemical Engineering, Imperial College London, JENNY NELSON, Department of Physics, Imperial College London, JOAO CABRAL, Department of Chemical Engineering, Imperial College London — The microstructure of organic photovoltaics (OPV) is often unstable and very sensitive to processing parameters. It is known that fullerene crystallisation has a huge impact on device behaviour. However, the observation of different size and shape of fullerene crystals in different polymer matrices is not yet understood. We select as a model system amorphous regiorandom poly(3-hexylthiophene-2,5-diyl) (RRa-P3HT) and crystalline phenyl-C61-butyric acid methyl ester (PCBM). We study the nucleation and growth of PCBM crystals into RRa-P3HT matrix as a function of fullerene loading, solution concentration, film thickness, supercooling and surface energy (tuned by UV ozonolysis) using optical and atomic force microscopy, and DSC. We show that the shape of PCBM crystallites can be tuned from needles to spheroidal crystals by increasing undercooling. We argue that the different behaviour of polymer:PCBM blends can be rationalised in term of undercooling, viscosity and fragility of the composite. We finally evaluate the role of additives on the crystallisation of PCBM. By mapping the impact of the processing parameters in this model system, we establish a simple strategy towards controlling PCBM crystallisation at relevant lengthscales for OPV performance.

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