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Correlating Polymer Crystals via Self-Induced Nucleation

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Crystallizable polymers often form multiple stacks of uniquely oriented lamellae, which have good registry despite being separated by amorphous fold surfaces. These correlations require multiple synchronized, yet unidentified, nucleation events. Here, we demonstrate that in thin films of isotactic polystyrene, the probability of generating correlated lamellae is controlled by the branched morphology of a single primary lamella. The nucleation density n_s of secondary lamellae is found to be dependent on the width of the branches of the primary lamella. This relation is independent of molecular weight, crystallization temperature, and film thickness. We propose a nucleation mechanism based on the insertion of polymers into a branched primary lamellar crystal. Even in single crystals, characterized by faceted structures with a well-defined envelope reflecting the underlying crystal unit cell, polymers are folded and thus in a meta-stable state. Annealing such meta-stable single crystals allowed to unveil the initial morphological framework of a dendritic single crystal, i.e. the initial stages of growth.