Tunable Microphase Segregation of Gradient Copolymers: Ordering in Materials with Sinusoidal Composition Profiles

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— Gradient copolymers are a class of polymers that exhibit a gradual change in composition along the entire chain from mostly A-monomer to mostly B-monomer. Theoretical work has predicted that gradient copolymers organize into sinusoidal composition profiles rather than the step-like profiles seen for block copolymers. Here, small-angle x-ray scattering and rheological studies were performed to investigate the impact of gradient design and comonomer choice on this unique ordering. Samples showed a variety of non-terminal behaviors consistent with their chain architecture relative to block copolymers, indicating highly tunable microphase segregation. Scattering results also demonstrated that a range of ordering was attained, with higher order peaks visible in more microphase-segregated samples. In addition, it was demonstrated for the first time that application of high amplitude oscillatory shear induced domain shear alignment in a manner similar to block copolymers, even though gradient copolymers do not possess distinct domain boundaries.