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Packing Frustration and its Effects on Phase Transitions in Block Copolymer Films[1]

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Packing frustration in ordered asymmetric block copolymers arises from the non-uniform stretching of chains required to fill the Wigner-Seitz unit cells of the ordered structure. While its effects are clearly important for block copolymers in the bulk, these effects may be even more important for thin block copolymer films. This is because the Wigner-Seitz unit cells of surface block copolymer domains differ significantly for those of the bulk, giving rise to a more non-uniform chain stretching in the surface layers. Packing frustration plays an important role in the phase stability of multilayer films of spherical block copolymers that show transitions from hexagonal close packed (HCP) to face-centered orthorhombic (FCO) to nearly BCC as the number n of layers increases. Additions of short corona block homopolymers reduce the larger packing frustration of the HCP surface and bulk layers by segregating to the regions of the Wigner-Seitz cell where stretching would be most extreme. These additions increase the n at which a transition from HCP to FCO occurs. The order-disorder temperature of spherical and cylindrical block copolymers also decreases on going from a bilayer to a monolayer. A monolayer of cylinders has a square Wigner-Seitz cell whereas in the bulk, the cell is hexagonal; a monolayer of spheres has a Wigner-Seitz cell that is a hexagonal prism whereas the cells of the bulk BCC, FCO and HCP are significantly more uniform. Finally packing frustration is probably also partly responsible the very different cylinder to sphere order-order transition temperatures observed for diblock and triblock copolymers in thin films relative to bulk samples. Results from scanning force microscopy, transmission electron microscopy and grazing incidence transmission electron microscopy will be used to illustrate these effects experimentally and, where possible, these will be compared to predictions of self-consistent field theory simulations.

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