Abstract:

Using optical stepper lithography, we create two-dimensional (2D) Brownian systems of microscopic, shape-designed platelets, which are dispersed in an aqueous surfactant solution and move just above a flat glass wall as a consequence of random thermal excitations. Depending on the specific shape, very slow 2D osmotic compression of a monolayer, composed of a single type of shape, from the dilute limit can lead to a wide variety of outcomes: highly ordered crystallites, liquid crystals, and disordered glasses. Optical video microscopy experiments reveal interesting phenomena in these systems that had not been previously predicted, such as short-range local chiral symmetry breaking in a triatic liquid crystal phase of equilateral triangles and long-range chiral symmetry breaking in a rhombic crystal phase of 72-deg rhombs. By considering simple cage models, we show that both forms of entropic chiral symmetry breaking are direct consequences of entropy maximization, when considering both translational and rotational accessible microstates. In addition, we have demonstrated that other shapes, such as Penrose kites and alternatively certain annular sector particles (i.e. “C”-shapes), form disordered systems through different mechanisms. For Penrose kites, the disorder is a consequence of an extreme diversity of incommensurate and topologically different local polymorphic configurations. By contrast, for annular sector particles, the disorder arises as a consequence of lock-and-key dimerization reactions that create a racemic mixture of chiral dimers that cannot phase separate because of crowding. Overall, our results show that the statistical mechanics of multi-body systems of hard shapes, even in the absence of significant attractive interactions and when slowly crowded in a near-equilibrium manner, can lead to a rich range of collective behavior that depends on the interplay of entropy and geometry.