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Highly-Ordered Monolayers in a Hurry: Non-Equilibrium Pathways to High-Quality Superlattices TERRY BIGIONI, University of Chicago, TOAN NGUYEN, University of Chicago, ERIC CORWIN, University of Chicago, THOMAS WITTEN, University of Chicago, HEINRICH JAEGER, University of Chicago, XIAO-MIN LIN, Argonne National Laboratory — The formation of 2D superlattices has long been thought to occur at the end of evaporation when particle-particle, -solvent and -substrate interactions can induce spinodal phase separations. Surprisingly, recent experiments have shown that it is indeed possible to create highly-ordered nanocrystal monolayers by non-equilibrium processes such as drop drying. In these cases the monolayer self-assembly occurs on the liquid-air interface. We unravel details of the self-assembly mechanism by tracking the monolayer formation through its entire evolution using a combination of direct optical microscopy and transmission electron microscopy. Drop drying itself is a highly non-equilibrium phenomenon that is affected by solvent evaporation, mass transport inside the drop, drop geometry, environmental conditions, and other factors. Despite this complexity, we observed that highly-ordered monolayers form according to three different robust growth laws: linear, quadratic, and exponential in time. We delineate a phase diagram based on a simple geometric model that can be used to better understand the self-assembly of nanocrystal thin films from solution.

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