Abstract:
We have achieved real-time direct-imaging of epitaxy in thin colloidal films, allowing us to observe in unprecedented detail fundamental mechanisms governing epitaxial growth that cannot be resolved in conventional atomic epitaxy. Our novel experimental techniques enabled us to discover a new fundamental kinetic mechanism that determines the two dimensional and three dimensional structures of growing atomic, nanoparticle, and colloidal crystalline films. This kinetic mechanism suggests new routes towards controlling film morphology during epitaxy.

Summary:
In recent years of colloidal crystallization studies, microfabricated templates have been successfully used to template the growth of sedimented colloidal particles into self-assembled structures [1]. This technique allows for accelerated colloidal crystallization, which has been exploited to study the effects of various boundary conditions, including stretched templates [2] and surfaces with embedded grain boundaries [3]. In our current research project, we fabricate polymethylmethacrylate (PMMA) templates on silica glass substrates. A suspension of 1.3 micron diameter charge stabilized polystyrene colloidal particles are sedimented onto the patterned surface, whose 1.3 micron diameter holes trap a monolayer of particles, forcing them to assume the underlying symmetry (Figure 1). The growth, of a single layer directly above the trapped layer is studied using an inverted microscope. The dynamics of crystallization of this single layer of colloidal particles can then be directly observed with single particle resolution in real time. Further analysis using optical tweezers that manipulated individual particles allowed us to measure the step edge barrier by recording how long it took for particles to move off crystal islands. We showed that Brownian motion rather than energetics was primarily responsible for the step edge barrier (Figure 2) [5].

The fabrication process utilizes electron beam lithography to pattern periodic arrays of 1.3 micron holes in a 500 nm thick layer of PMMA. Two specific underlying array symmetries are created on the templates to probe the dynamics of the colloidal particles. A square lattice of micron-size holes may be used to grow a face-centered cubic crystal along the (100) face—by tuning the lattice constant of the array, we can probe both commensurate and incommensurate lattice matches. Alternatively, we pattern triangular lattices to explore the (111) face. In combination with a temperature dependent depletion induced attractive interaction [4], we are able to observe the dynamics of both island growth and melting. An initial layer of colloidal particles, which form within the template holes, self-assemble due to the attractive interaction which occurs between both individual particles.
and individual particles with the substrate. Island growth occurs atop the initial layer of colloidal particles with the same symmetry as the underlying lattice.

We are now moving on to pursue two new projects. First we are performing similar experiments on stretched templates to explore processes relevant for heteroepitaxy. Second we are exploring the melting dynamics of crystalline Islands. Small changes in temperature cause the depletion induced interaction to weaken, leading the islands to melt (Figure 3). The rate of melting is found to vary dramatically depending on the underlying lattice symmetry (Figure 4).

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