Melting of Colloidal Crystal Films

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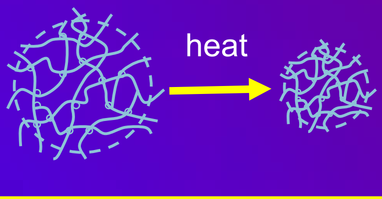
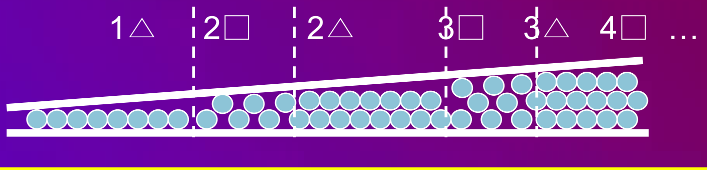
Two-dimensional and three-dimensional crystals have qualitatively different melting behaviors. As the crossover, thin-film melting is poorly understood. We studied thin-film melting with single-particle resolution for the first time by video-microscopy measurements on thermally sensitive colloidal crystals. Different melting behaviors were observed in three thickness regimes. (1) Thick film (>4 layers) melt heterogeneously from grain boundaries for polycrystals and from film-wall interfaces for single crystals; crystal-liquid coexistence regime decreases with film thickness and vanishes at 4 layers; (2) Thin-films (2 – 4 layers) melt homogenously in one step; (3) Monolayers melt homogeneously in two steps with an intermediate hexatic phase. A novel type of heterogeneous melting from dislocations was observed in 5- to12-layer films.

Experiment

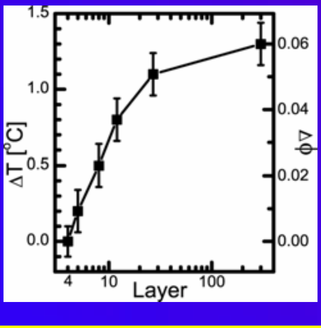
N-isopropyl acrylamide (NIPA) microgel sphere:



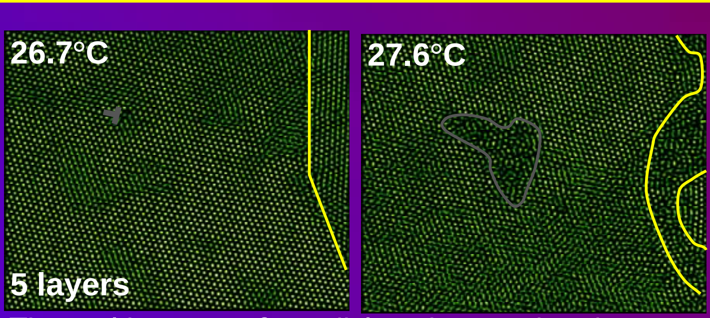
Phase diagram



Thick films (> 4 layers)



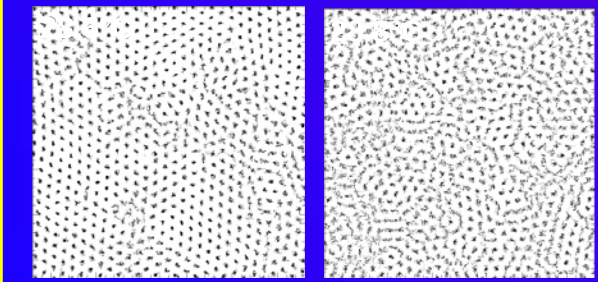
Liquid-crystal coexistence regime in temperature (ΔT) or volume fraction (ΔΦ) decreases with film thickness and vanishes at 4 layers.



The melting starts from dislocations and grain boundaries, and form liquid “lakes” and stripes

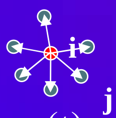
Thin films (2 – 4 layers)

Particle trajectories near the melting point



- Melting starts from both grain boundaries and inside domains.
- No large liquid and crystalline domains coexist

Orientational order parameter

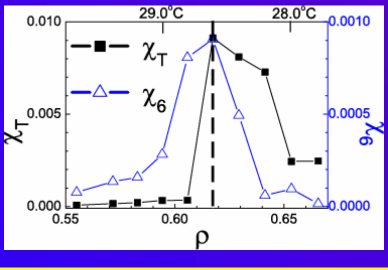


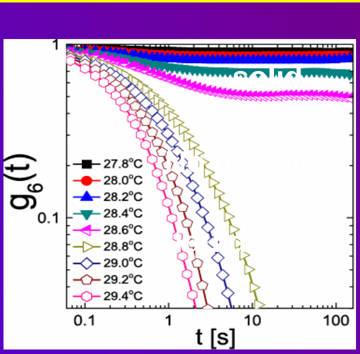
$$\psi_{6i} = \frac{1}{n_i}\sum_{j}^{n_i} e^{6i\theta_{ij}}$$

$$g(t) = \left\langle \psi^*(\tau)\psi(\tau+t) \right\rangle$$

$$\chi_{6,T} = L^2\left(\left\langle \left|\psi_{6,T}\right|^2 \right\rangle - \left\langle \left|\psi_{6,T}\right| \right\rangle^2\right)$$

susceptibility χ_6, χ_T





- $g_6(t)$:Constant curve – crystal
- $g_6(t)$: Exponential curve –liquid
- $g_6(t)$: No power-law curve – no hexatic phase.
- χ_6, χ_T diverge at the same point: ⇒ one step transition; no hexatic phase.

Monolayer χ_6, χ_T diverge at different point with power-law g6(t) ⇒ hexatic phase