Visualizing Kinetic Pathways of Homogeneous Nucleation in Colloidal Crystallization

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When a system undergoes a transition from a liquid to a solid phase, it passes through multiple intermediate structures before reaching the final state. However, our knowledge on the exact pathways of this process is limited, mainly due to the difficulty of realizing direct observations. Here, we experimentally study the evolution of symmetry and density for various colloidal systems during liquid-to-solid phase transitions, and visualize kinetic pathways with single-particle resolution. We observe the formation of relatively-ordered precursor structures with different symmetries, which then convert into metastable solids. During this conversion, two major cross-symmetry pathways always occur, regardless of the final state and the interaction potential. In addition, we find a broad decoupling of density variation and symmetry development, and discover that nucleation rarely starts from the densest regions. These findings hold for all our samples, suggesting the possibility of finding a unified picture for the complex crystallization kinetics in colloidal systems.

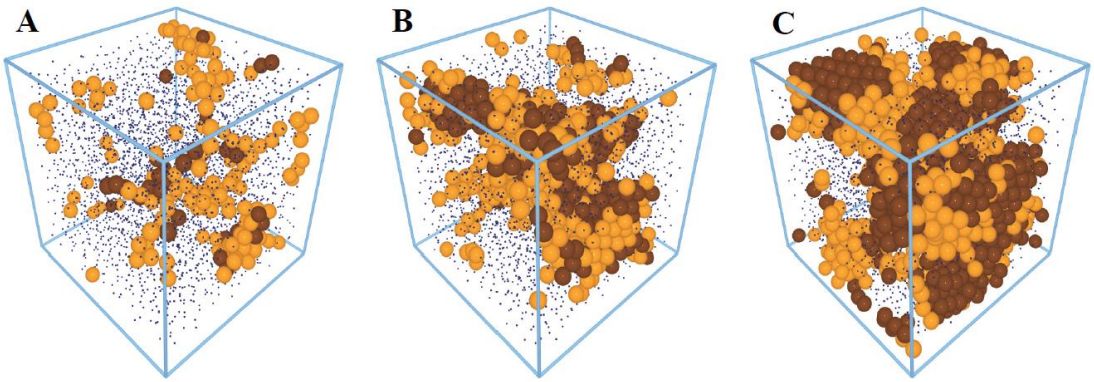

Figure 1. Precursor mediated crystallization process. The dark-brown spheres represent nuclei particles while the light-brown ones indicate relatively-ordered liquid precursors. (A) At the beginning of nucleation. (B)Around critical nuclei size. (C) Post critical growth.

References: