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June 2nd, 2021, 17:00-18:00 CET

Investigating crystallization pathways by liquid phase TEM

By Jim De Yoreo

Abstract:
The emergence of order in materials systems ranging from simple salts to complex supramolecular arrays has long been viewed through the lens of classical nucleation theory in which monomeric building blocks assemble into ordered structures identical to that of the bulk through inherent thermal fluctuations that overcome a free energy barrier.1 However, recent observations have revealed a rich set of hierarchical pathways involving higher-order species ranging from multi-ion clusters to dense liquid droplets to transient amorphous or crystalline phases.2 Moreover, both intrinsic factors, such as supersaturation and temperature, and extrinsic factors, including added electrolytes and organics, lead to divergent pathways in a single system. Identifying these pathways and determining the thermodynamic and/or kinetic reasons why they occur is a difficult challenge due to the transient, nanoscopic nature of the intermediates. Due to its unique combination of spatial and temporal resolution, liquid phase TEM has provided new insights into crystallization processes.3 After setting the context for understanding hierarchical nucleation pathways, I will describe results of recent liquid phase TEM studies on iron4 and zinc5 oxides and on calcium carbonate6-8 aimed at understanding crystallization pathways, including the impact of additives that act either as surface-bound ligands or dopants. The results provide insights into crystallization via dense liquid and amorphous precursors, oriented attachment of nanocrystals, and mesocrystal formation via interface-driven secondary nucleation and attachment. More broadly, these results highlight the opportunity that liquid phase TEM provides for deciphering underlying mechanisms of crystallization.

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