Connecting energetics to dynamics in particle growth by oriented attachment using real-time observations

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Abstract

The interplay between crystal and solvent structure, interparticle forces and ensemble particle response dynamics governs the process of crystallization by oriented attachment (OA), yet a quantitative understanding is lacking. Using as a model system ZnO, we combine *in situ* TEM observations of single particle and ensemble assembly dynamics with simulations of interparticle forces and responses to relate experimentally derived interparticle potentials to the underlying interactions. We show that OA is driven by forces and torques due to a combination of electrostatic ion-solvent correlations and dipolar interactions that act at separations well beyond 5 nm. Importantly, coalignment is achieved before particles reach separations at which strong attractions drive the final jump to contact. The observed barrier to attachment is negligible, while dissipative factors in the quasi-2D confinement of the TEM fluid cell lead to abnormal diffusivities with timescales for rotation much less than for translation, thus enabling OA to dominate.