

Understanding Torque-Generating Forces Enabling Crystal Growth by Oriented Attachment

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Interfacial forces between crystals that depend on their mutual crystallographic alignment enable diverse multiscale phenomena such as crystal growth by oriented attachment, Schiller layer formation, clay swelling, adhesion and friction anisotropy, and grain boundary structuring in polycrystals. While the attraction or repulsion of two crystallites is based on a set of forces that are generally well understood, aspects of these forces that are sensitive to lattice alignment have been more difficult to probe. For example, in oriented attachment, where crystal growth entails self-assembly from nanocrystals, the existence of torque-generating forces that align approaching particles to enable adhesion have been implied more often than proven.

Conceptually, the types of interfacial forces that can be sensitive to relative orientation include Coulombic, van der Waals (vdW), solvation, and ion correlation forces. The theoretical underpinnings of these anisotropic forces are well established, but techniques that can isolate and measure their magnitudes for a given pair of interacting oriented crystal faces have generally been limited to use of macroscopic yet atomically flat single crystals (e.g., mica). Lattice-specific adhesion anisotropy between faces of microscopic crystals has been demonstrated, but a detailed interpretation of such data is difficult owing to the complex interplay of electrostatic, hydration, and vdW interactions giving rise to the observed total force.

When lattice polarizability is anisotropic, the vdW dispersion attraction can, in principle, contribute to this direction dependence. We report measurement of this attraction between rutile nanocrystals, as a function of their mutual orientation and surface hydration extent. At tens of nanometers of separation, the attraction is weak and shows no dependence on azimuthal alignment or surface hydration. At separations of approximately one hydration layer, the attraction is strongly dependent on azimuthal alignment and systematically decreases as intervening water density increases. Measured forces closely agree with predictions from Lifshitz theory and show that dispersion forces can generate a torque between particles interacting in solution and between grains in materials.