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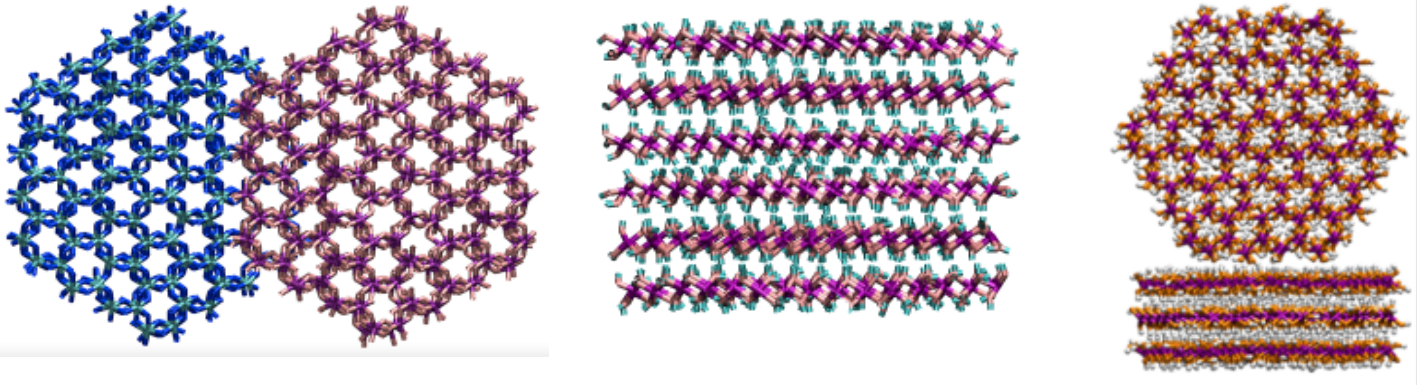
Sandia National Laboratories

GOLDSCHMIDT® Hawai'i 2022

Effect of Particle Misalignment on Energetic Pathways towards Oriented Attachment of Gibbsite Nanoparticles



Tuan Ho and Louise Criscenti

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U.S. DEPARTMENT OF ENERGY NNSA

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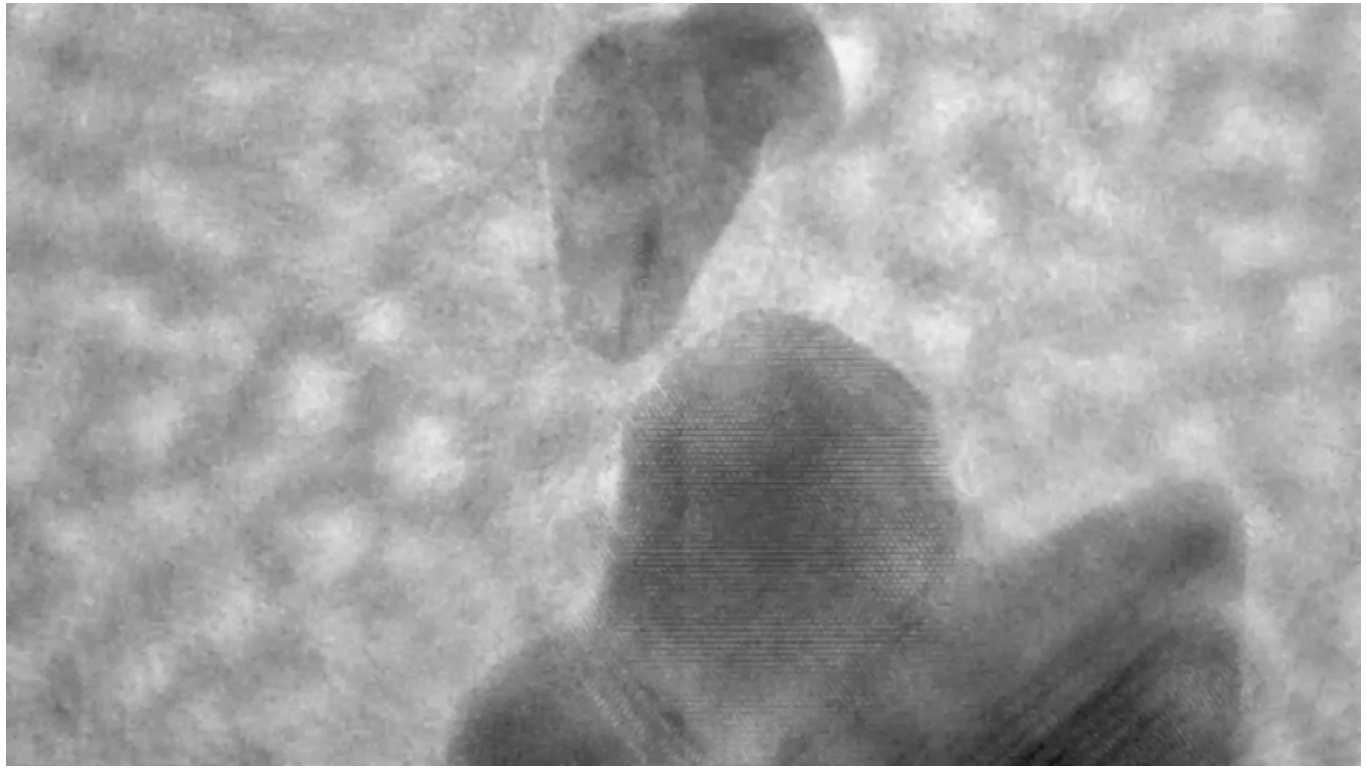
Introduction: particle aggregation and oriented attachment

- Nuclear waste management of Hanford and Savanah River nuclear waste tanks:

Gibbsite and boehmite particles form complex aggregates

- Oriented attachment is a special case of particle aggregation:

Crystalline particles assemble into a larger particle by attaching on specific crystal faces that are lattice-matched.



iron oxyhydroxide nanoparticles

(Li et al, Science 2012, 336, 1014)

Introduction: oriented attachment

- Thermodynamics: particle-particle, particle-solution, and solution-solution interactions

Kinetics: Brownian motions and experimental conditions (e.g., dehydration)

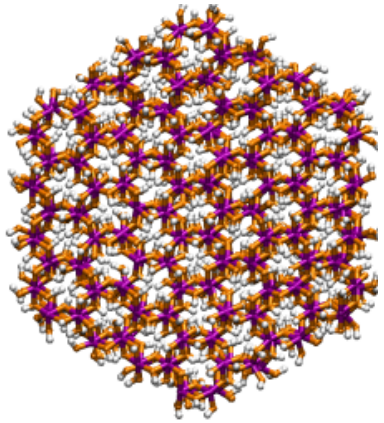
Particle motions: approaching, translating, rotating

Energy-structure relationship?

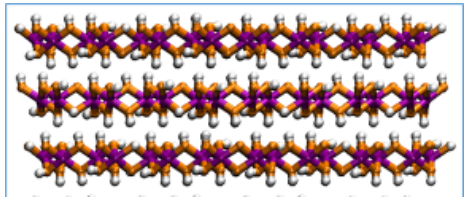
Gibbsite particle oriented attachment

Molecular dynamics simulations

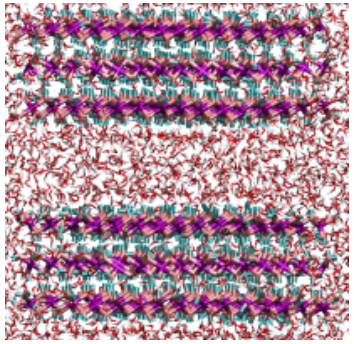
Top view



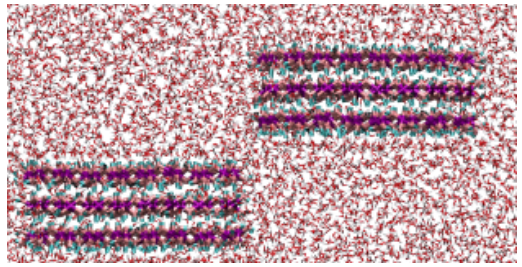
Side view



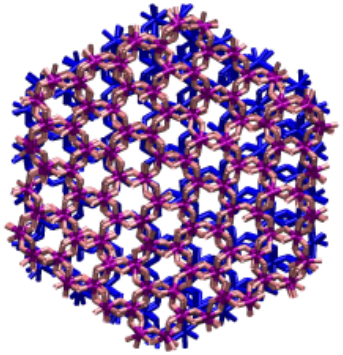
Approaching

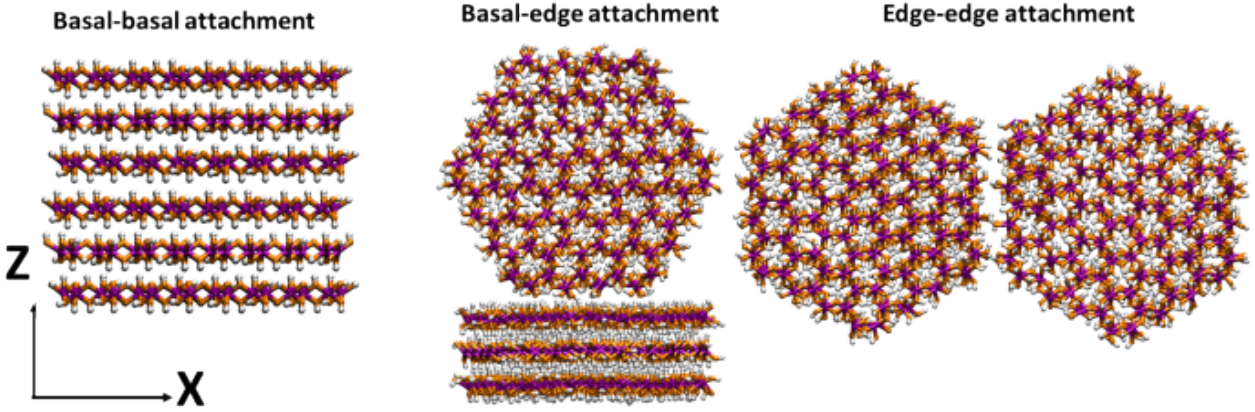


Sliding



Rotating

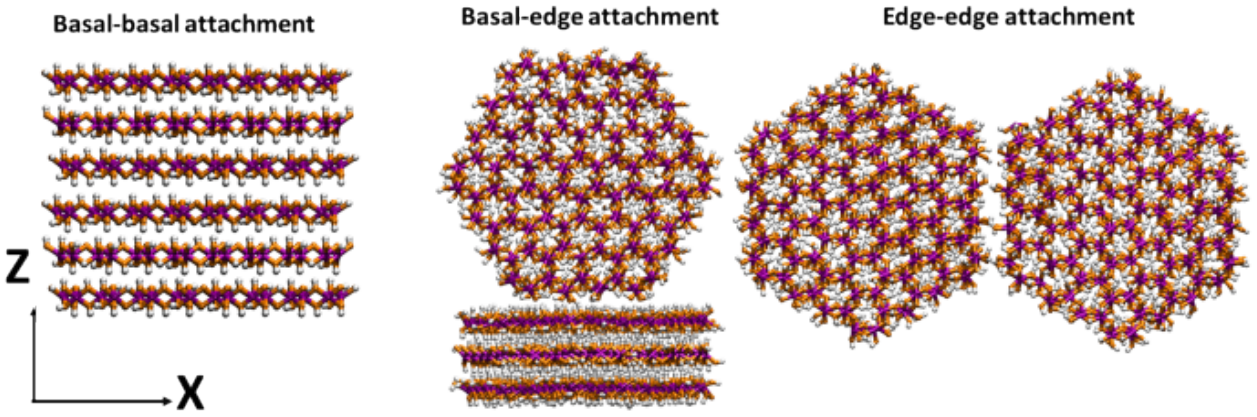


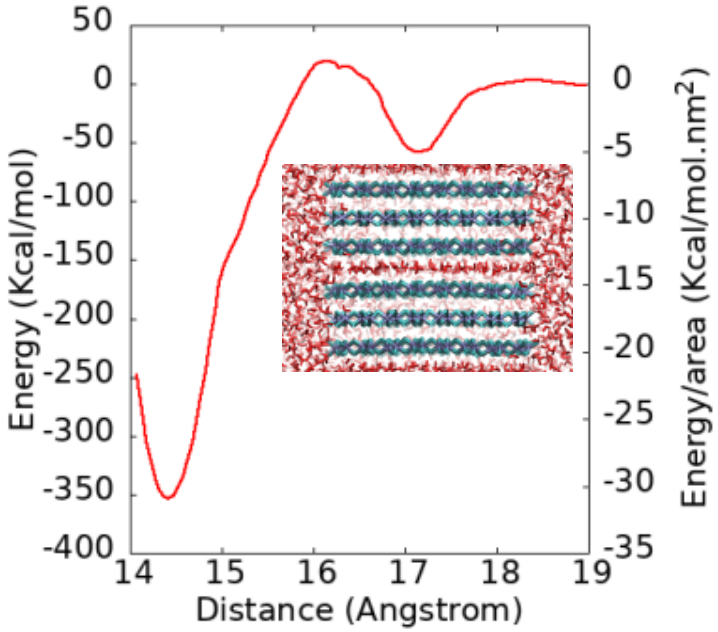


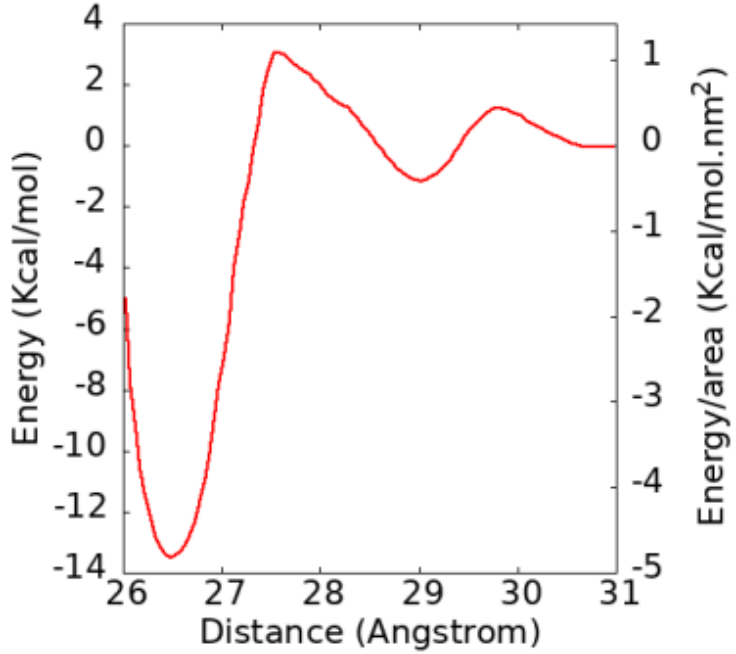
Basal-basal surfaces interactions

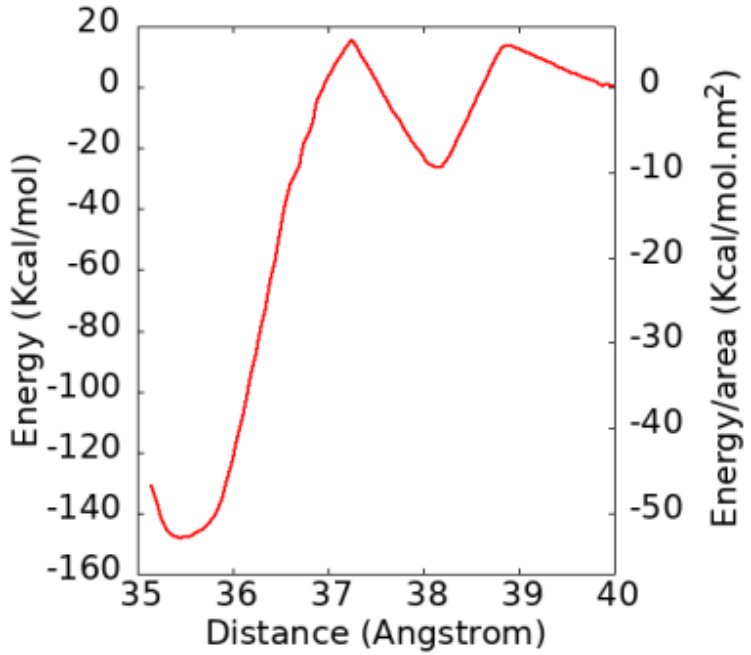
Energy-structure relationships:
Potential of mean force calculations
(very expensive)

Approaching motion





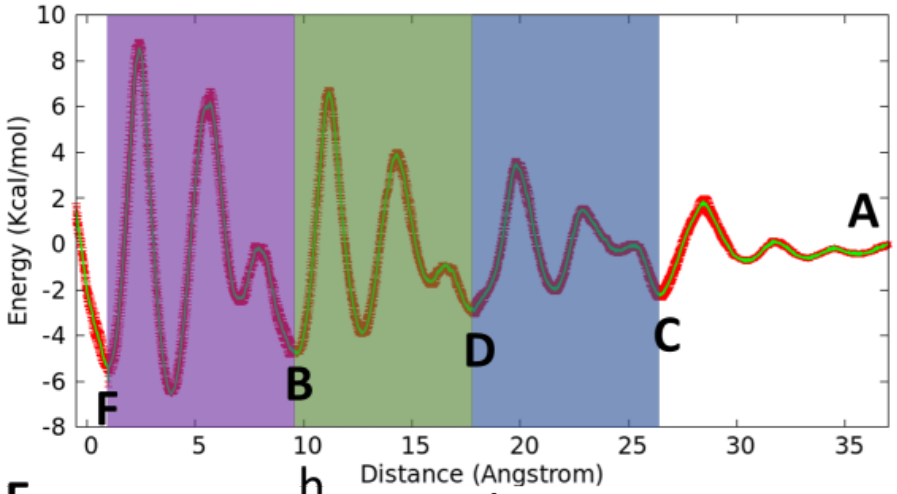


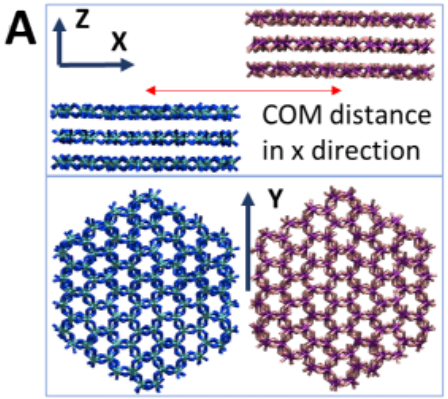


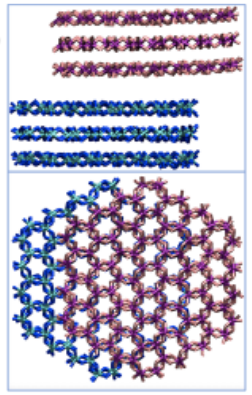
Per surface area: edge-edge attachment is more favorable
Large particle: basal-basal attachment is more favorable

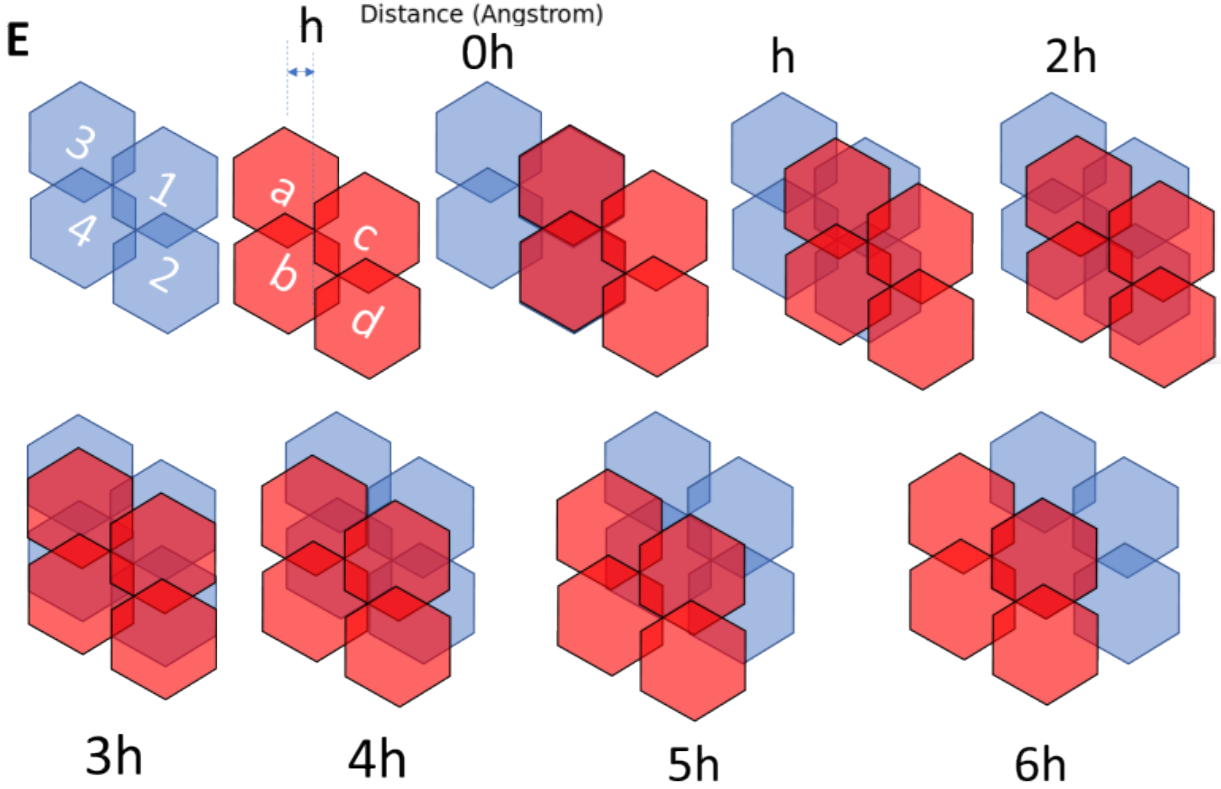
Journal of Colloid and Interface Science 600, 310-317

Sliding motion

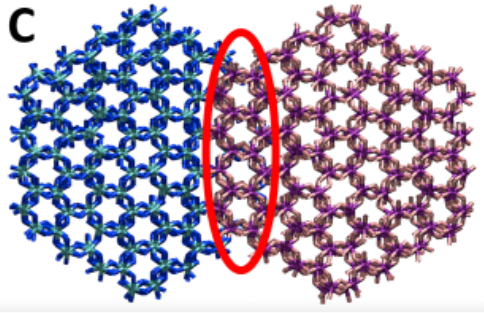


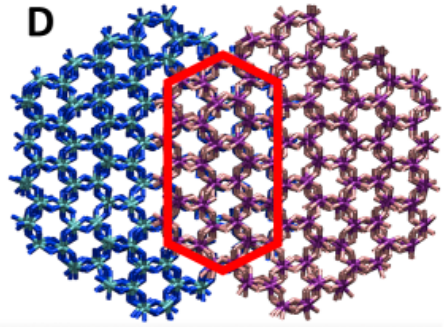


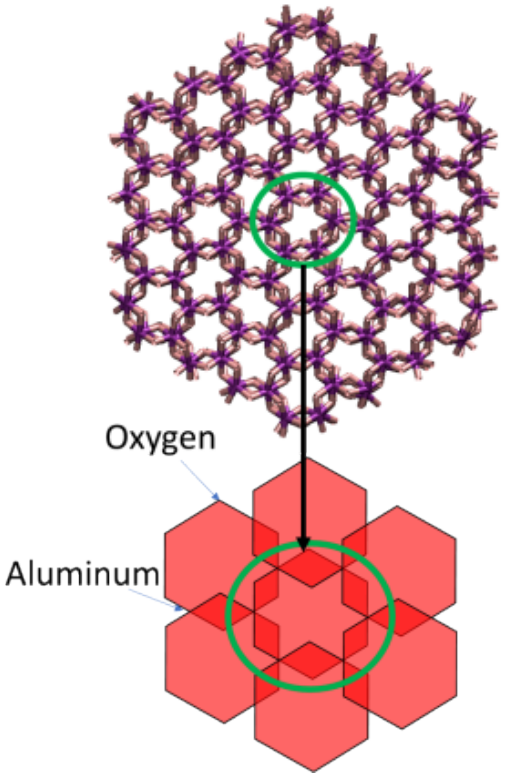




Lattice alignment



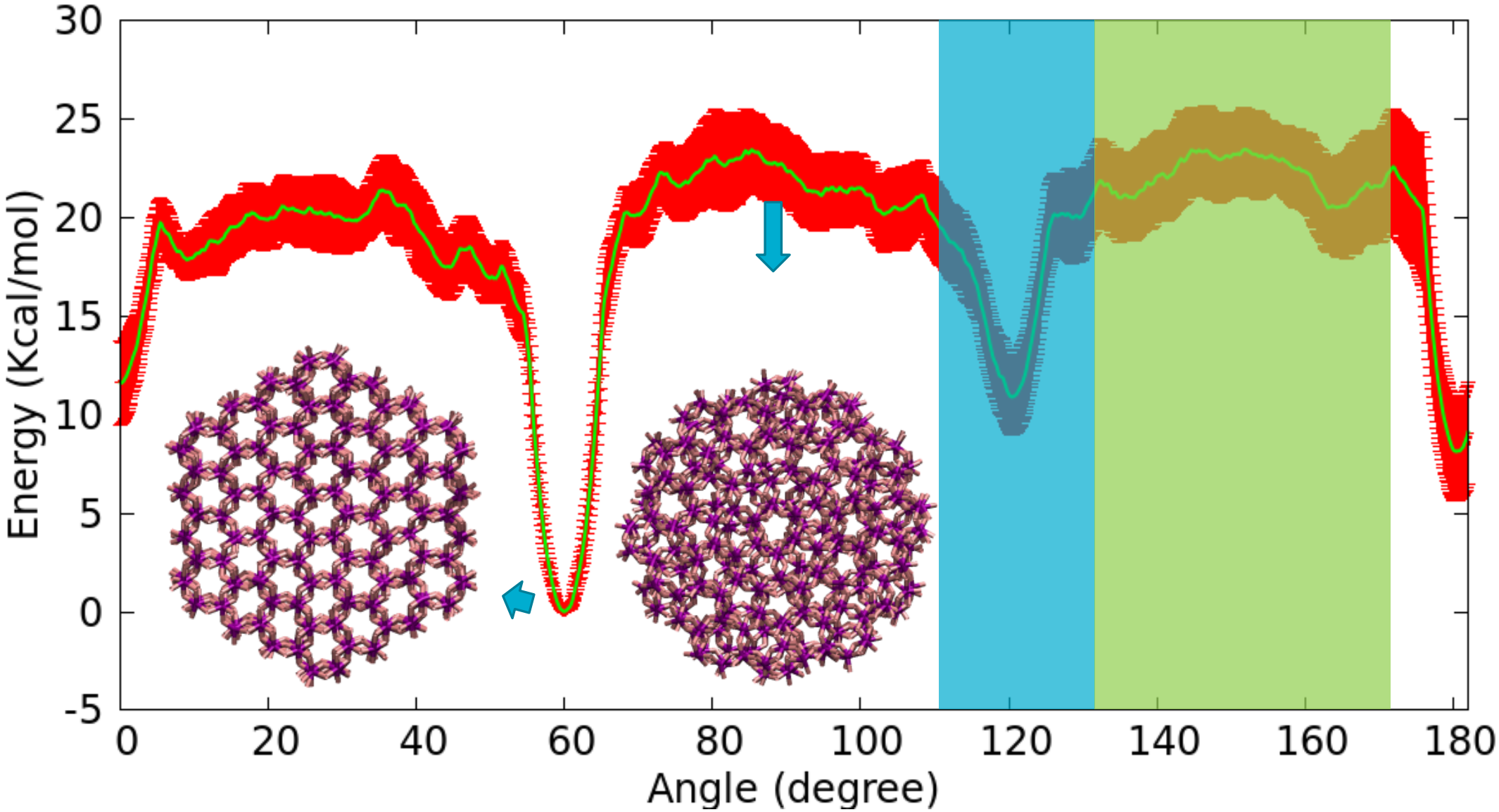




Importance of atom-by-atom mismatch

Ho et al., in preparation

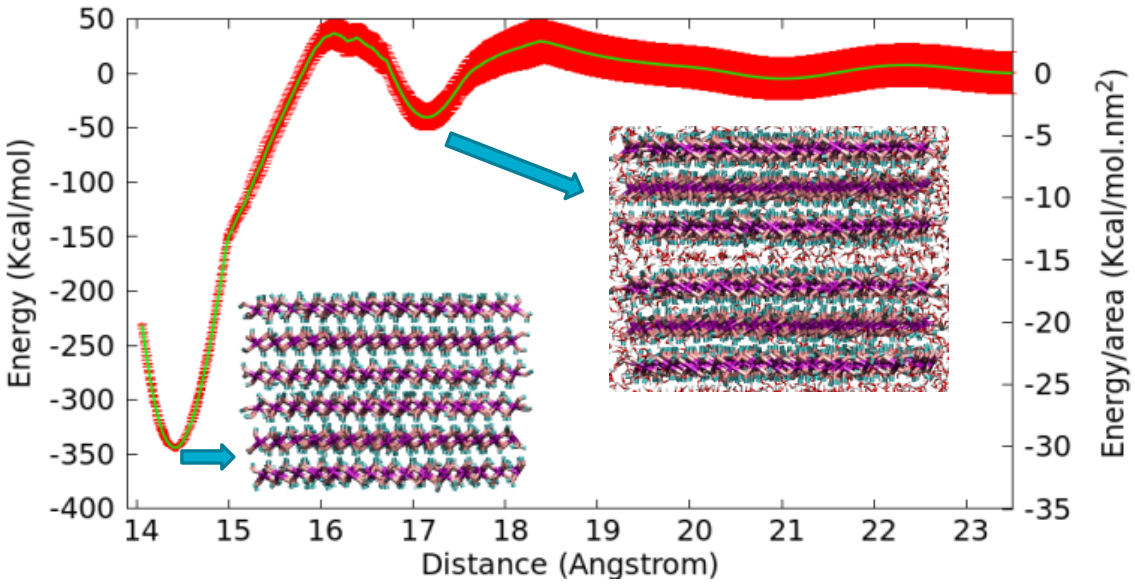
Rotating motion



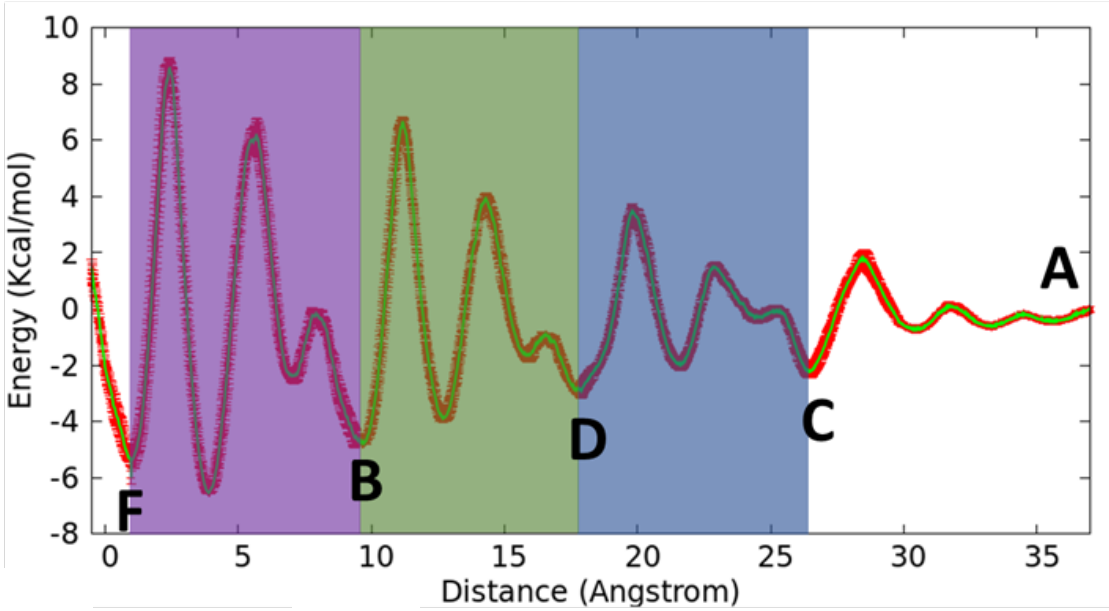
60º periodicity

Energy barriers

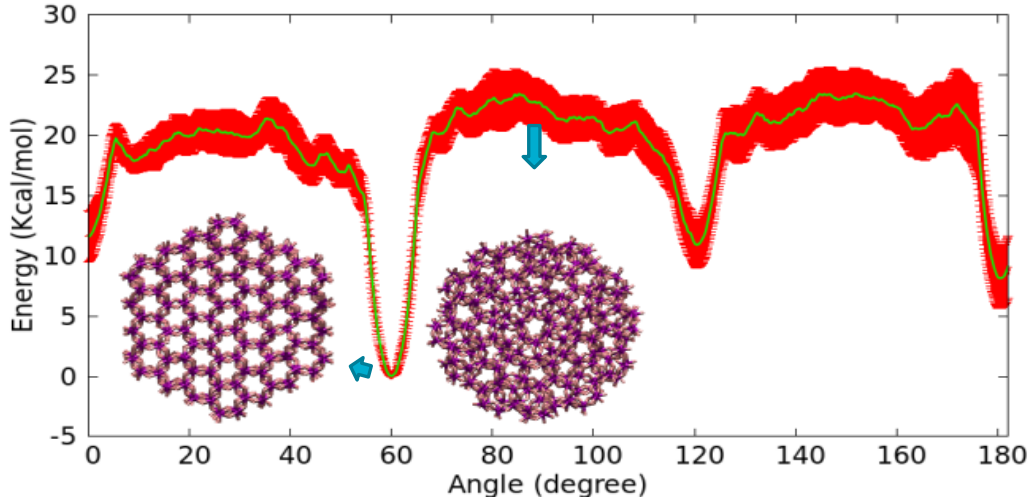
Approaching motion



Sliding motion



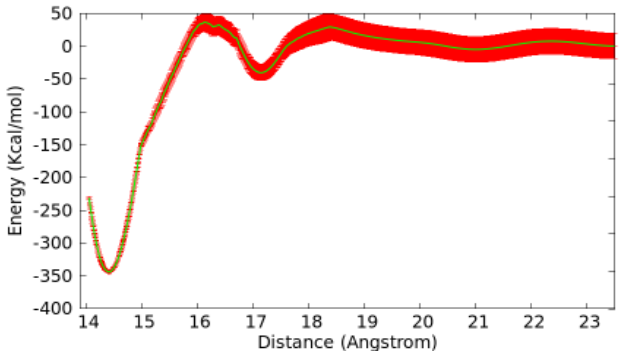
Rotating motion



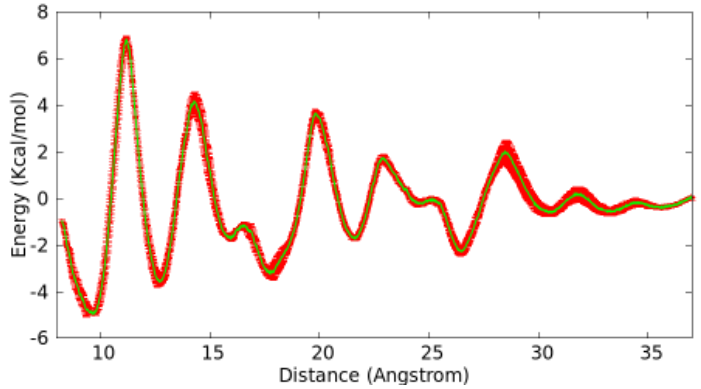
Approaching motion encounters the highest energy barrier

Roles of water

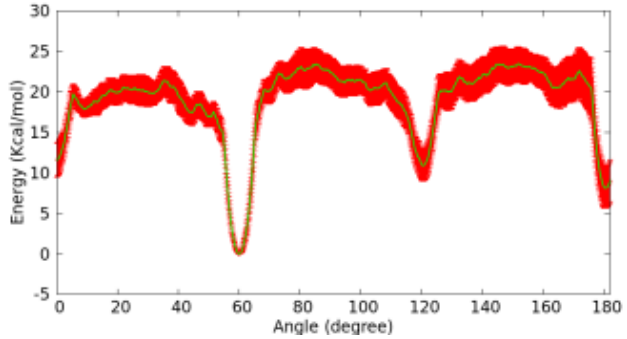
Interaction in water



Approaching motion

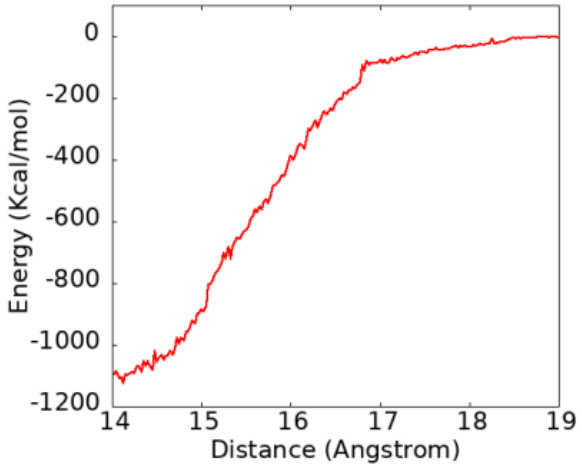


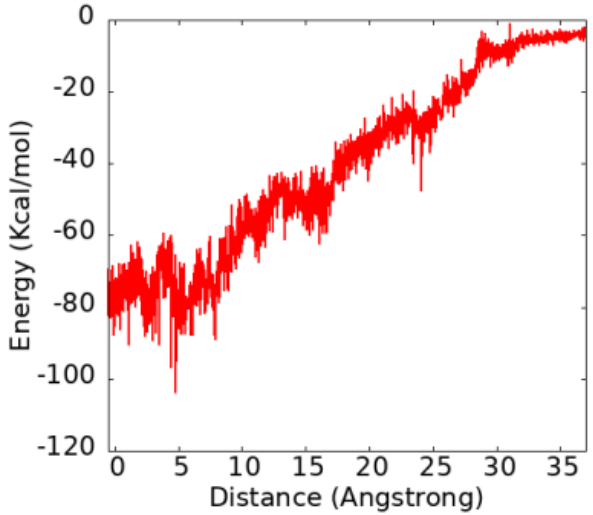
Sliding motion

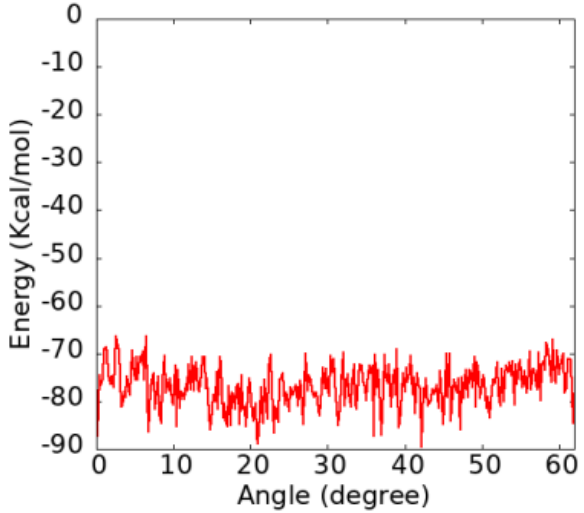


Rotating motion

Interaction in vacuum



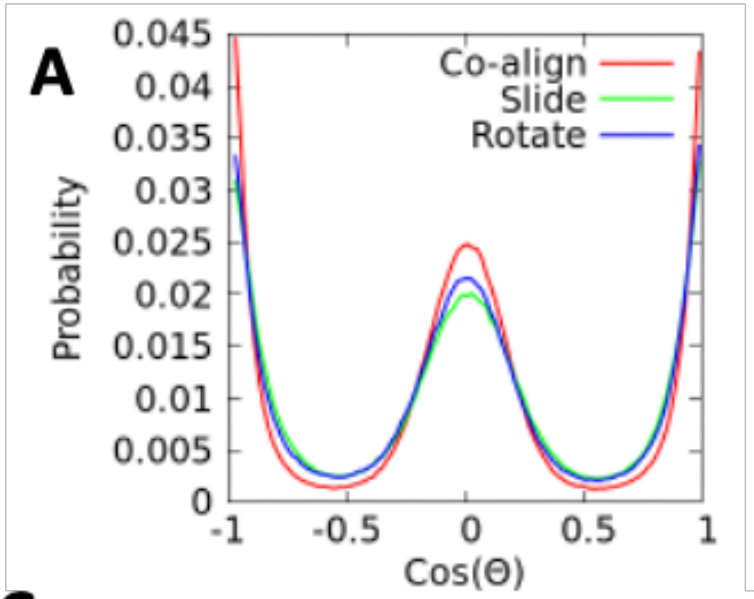


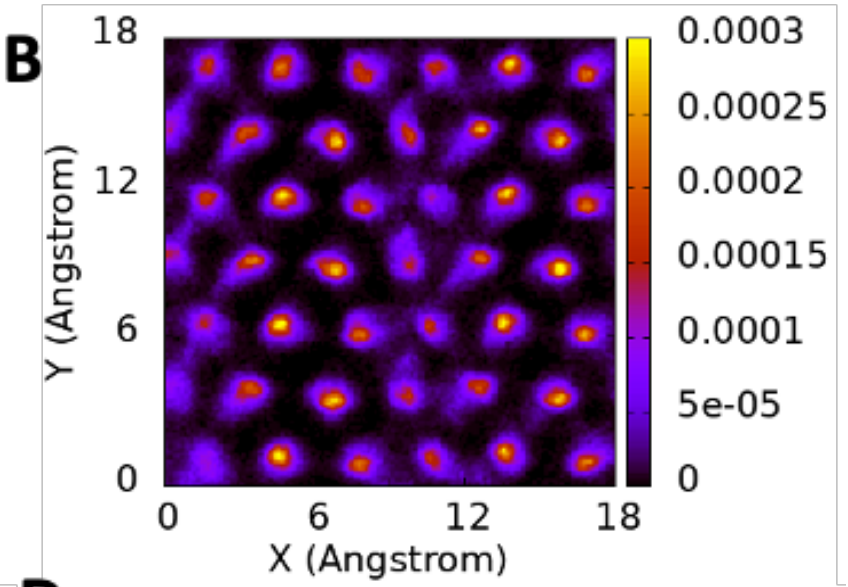


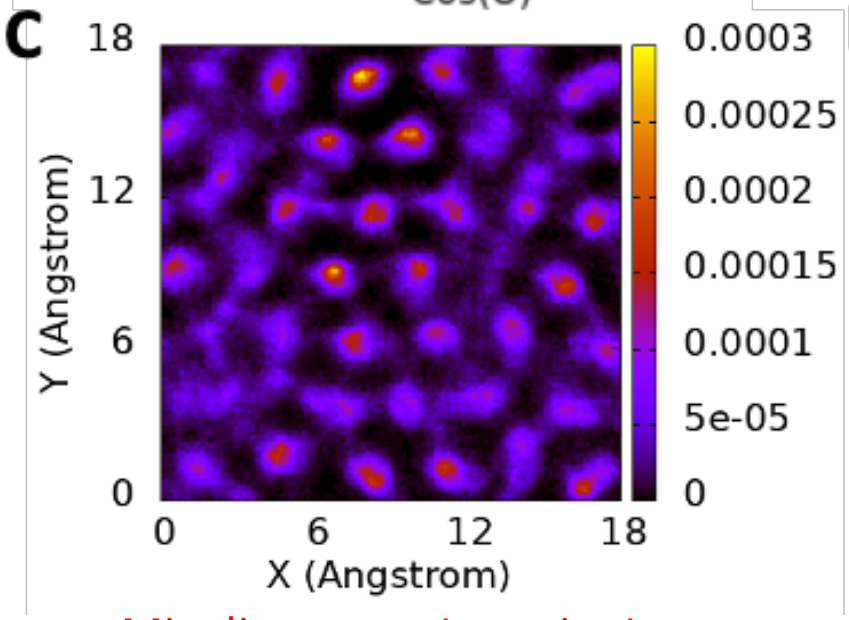
- Water controls the fluctuations in the PMF profiles for all three motions studied
- Water reduces the interaction between two particles
- However, particles still “feel” each other in water.

Water structure

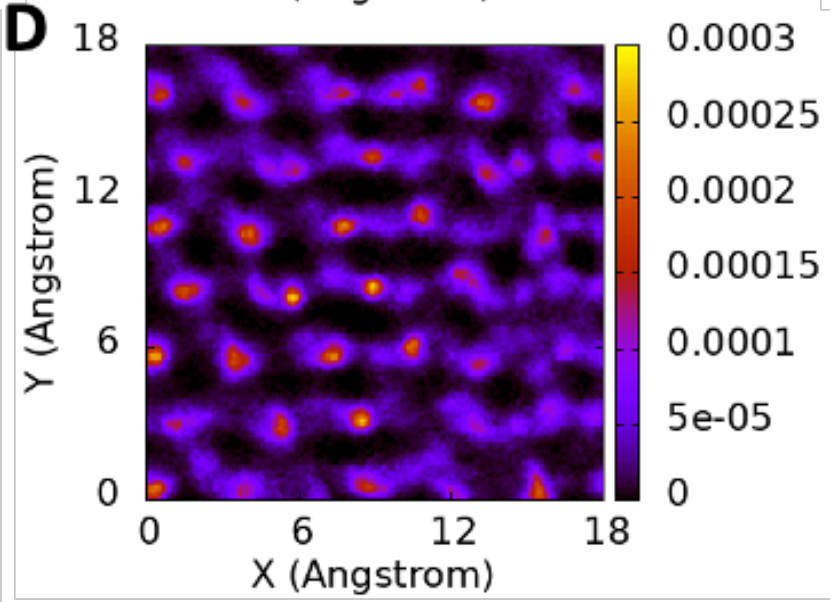
Perfect alignment







Misalignment (rotating)

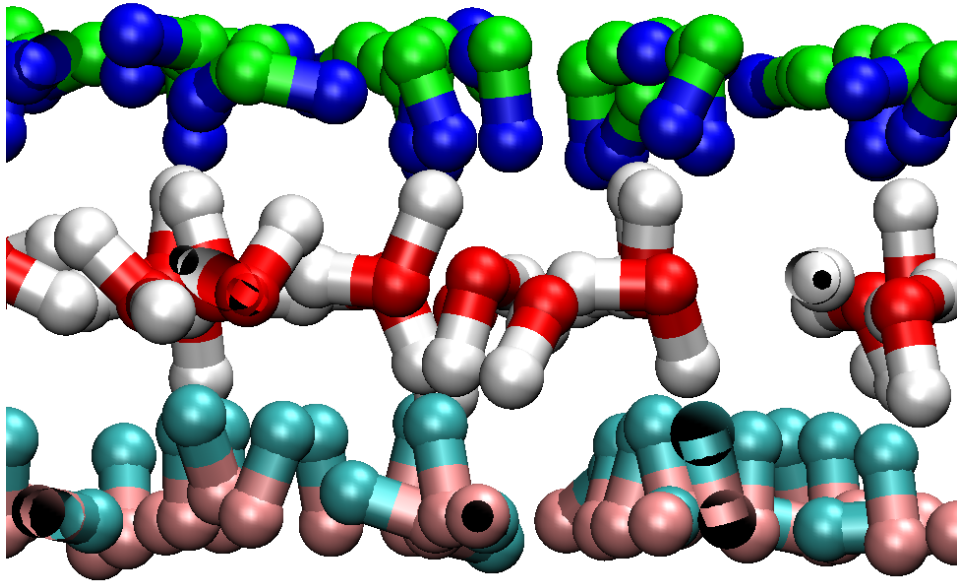


Misalignment (sliding)

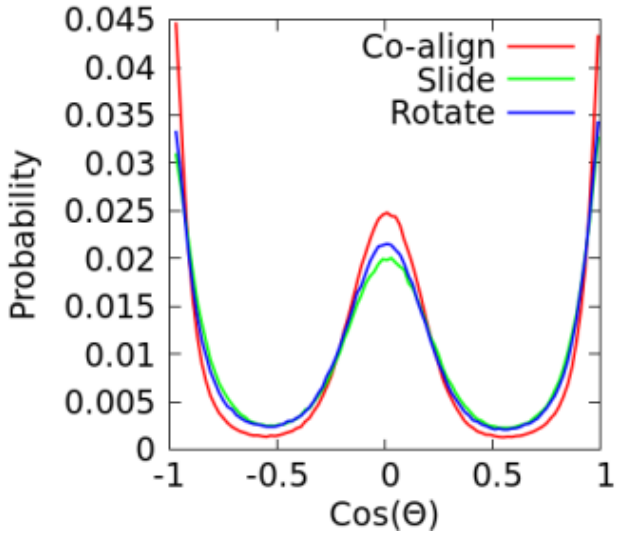
Water is less structured for the mis-aligned configurations, compared to the co-aligned configuration

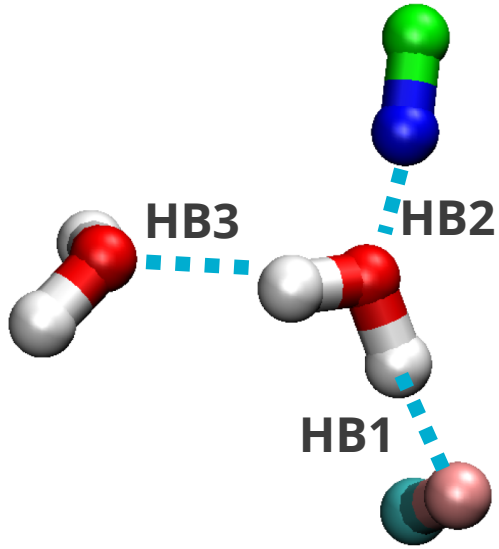
Hydrogen bond network

Surface OH



Surface OH



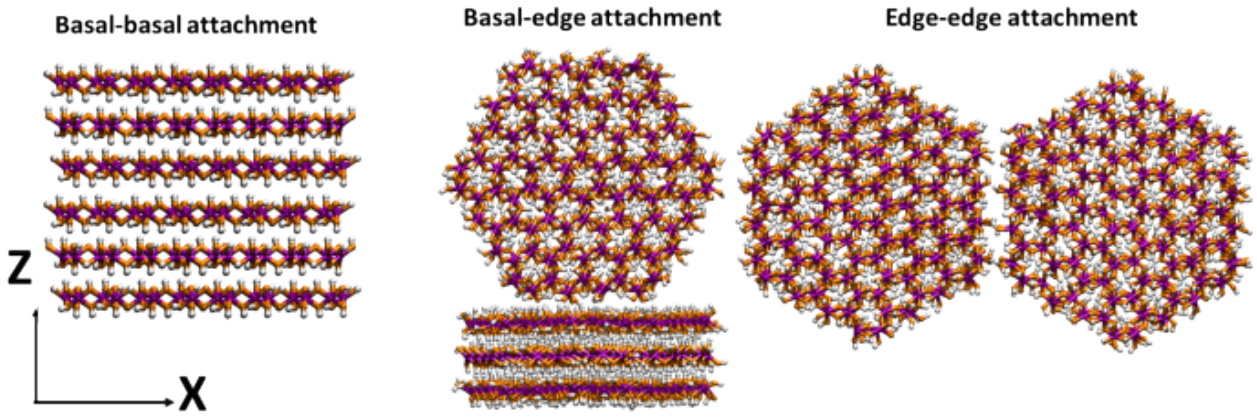


Coalignment: # water-water HB 1.86

Misalignment: # water-water HB 1.75

Conclusions

- Energy-structure relationships during particle motions: approaching, sliding, and rotating



Per surface area: edge-edge attachment is more favorable
Large particle: basal-basal attachment is more favorable

- Approaching motion encounters highest energy barrier
- Water properties and atom-by-atom mismatch control the energy-structure relationship during the motions

Thank you!

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