**Effect of Collective Dynamics and Anharmonicity on Entropy in Heterogenous Catalysis: Building the Case for Advanced Molecular Simulations**

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TWe present a perspective on where anharmonicity should be expected, where it has been observed from a theoretical perspective, and the methods currently employed to address it. We concentrate on three types of systems where we have observed major, non-negligible anharmonic effects: (1) supported nanoparticles, where the migration of metal atoms, complexes, and entire clusters exhibit anharmonic behavior in their dynamic motion; (2) porous solids, where confinement effects distort potential energy surfaces and hinder molecular motions, resulting in large entropic terms; and (3) solid/liquid interfaces, where interactions between solvent molecules and adsorbed species can result in large solvent organization free energy and unique reactivity.

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[3] Collinge, G., *et al* Effect of collective dynamics and anharmonicity on entropy in heterogenous catalysis: Building the case for advanced molecular simulations. *ACS Catalysis*, *10*(16), 9236-9260 (2020).