A First-principles Investigation of Superprotonic Activity in CsHSO$_4$

BRANDON WOOD, MIT, NICOLA MARZARI — With the recent push to realize the hydrogen economy, there has been a surge of interest in viable solid-state materials for use as fuel cell electrolytes. Among the candidates are various derivatives of the anhydrous superprotonic conductor CsHSO$_4$, which exhibits reasonably high ionic conductivity at relevant operating temperatures while remaining electronically insulating. However, despite being widely characterized experimentally, a truly atomistic picture of the diffusion pathways and mechanisms in the material is missing. To this end, we have characterized this material with extensive first-principles static and dynamical calculations on 112-atom supercells of CsHSO$_4$. We isolate the dominant atomistic mechanisms involved in the superprotonic behavior and discuss the effect of correlated diffusive events in enhancing proton transport. We also offer a detailed description of the dynamics of the hydrogen bond network topology as the diffusing protons propagate through the lattice structure. The role of our findings in understanding superionic behavior are discussed.