First-principles prediction of the structures, spectra, and phase diagrams of molecular crystals

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An *ab initio* computational prediction of the crystal structures and complete phase diagrams has long been a dream of chemists, physicists, and materials scientists. I present a general computational method that enables routine MP2 or CCSD calculations of the energies, enthalpies, or free energies of periodic and nonperiodic molecular crystals under high pressures and intermediate temperatures.

I will discuss application of this method, which is based on the embedded fragmentation, to the electronic and (anharmonic) vibrational structures of three-dimensional solid hydrogen fluoride, solid carbon dioxide, and proton-disordered ice Ich as well as proton-ordered ice VIII. I will address the crystal structures of these solids, their pressure dependence, their phase diagram and solid-solid phase transition, the assignments of infrared and Raman bands and their crystal-field splitting, the phonon dispersion and density of states as well as the interpretation of inelastic neutron scattering, and the pressure dependence of Fermi doublets in solid carbon dioxide as a spectroscopic geo-barometer.