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Stochastic Self-Consistent Harmonic Approximation: Inverse Isotope Effect in Palladium Hydride superconductors

Palladium hydrides display the largest isotope effect anomaly known in literature. Replacement of hydrogen with the heavier isotopes leads to higher superconducting temperatures, a behavior inconsistent with harmonic theory.

Anharmonicity is so large that approaches treating it as perturbation are expect to fail. To solve this issue, we implement, within a first-principles approach, the self-consistent harmonic approximation (SCHA), to obtain the anharmonic free energy, the thermal expansion and the superconducting properties fully ab initio [1]. We solve the SCHA by a novel stochastic approach, where we replace the cumbersome calculations of anharmonic coefficients (required by previous implementations) by the evaluation of atomic forces on supercells with suitably chosen stochastic ionic configurations.

In palladium hydrides, we find that the phonon spectra are strongly renormalized by anharmonicity far beyond the perturbative regime. Superconductivity is phonon mediated, but the harmonic approximation largely overestimates the superconducting critical temperatures. The SCHA explains the inverse isotope effect, obtaining a -0.38 value for the isotope coefficient in good agreement with experiments, hydrogen anharmonicity being the main responsible for the isotope anomaly. Finally we note that, beside the present application, our stochastic implementation of the SCHA will allow us to investigate strongly anharmonic systems in the non-perturbative regime, such as other Hydrogen compounds, ferroelectrics, and charge-density wave systems.

References:

[1] Ion Errea, Matteo Calandra, Francesco Mauri,Phys. Rev. Lett. 111, 177002 (2013); Phys. Rev. B 89, 064302 (2014)