Versatile correlated frozen phonons from universal MLIPs for (S)TEM simulations

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Harmonic phonon modes are not only critical for understanding the thermal properties of materials, but also desirable for accurate transmission electron microscopy simulations within the widely-used frozen-phonon approach. Although uncorrelated random displacements are typically sufficient for modeling diffuse backgrounds [1], the true correlated phonons may be of relevance for techniques including quantitative electron diffraction and electron ptychography [2].Although ab initio methods are routinely applied to calculate phonon dispersions for small and often non-orthogonal unit cells, (scanning) transmission electron microscopy ((S)TEM) scattering simulations typically require atomic displacements for potentially quite large orthogonal supercells. Further, since both the technical and computational effort of accurately simulating phonons from first principles is often greater than that of scattering, this severely limits their accessibility and adoption by the electron microscopy community.This challenge can be overcome with the help of force-constant potentials [3], which allow phonon properties learned for small cells to be extrapolated to arbitrary supercells. In this way, correlated phonon displacements for a given specimen model can be easily generated – including quantum-mechanical zero-point motion that is increasingly important as cryogenic instrumentation becomes more widespread. To further enhance the universality of this approach, the required forces can be efficiently calculated using recently developed foundational machine-learning interatomic potential (MLIPs), which have very recently become sufficiently accurate to model harmonic phonons for many materials across the periodic table [4].In this contribution, we establish a fully Python-based high-performance open-source workflow for including ab initio frozen phonons in electron scattering simulations using the abTEM package [5]. As demonstrations of our versatile approach, we quantify the effect of phonon correlations for selected-area and convergent-beam electron diffraction of $SrTiO_3$, and accurately model phonon-dependent electron scattering in silicon to show how correlated thermal motion between neighboring atoms preserves coherence even at elevated temperatures. Experimentally, this enables atomic-scale double-slit interferometry with a focused electron probe transmitting through a thin silicon crystal [6].

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