Automatically tuned PAW pseudopotentials for accuracy and efficiency

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Pseudopotentials for electronic structure calculations using density functional theory (DFT) can lower computational expense compared to all-electron DFT. However, pseudopotentials that are accurate, efficient, and transferable are difficult to generate because these objectives can compete with each other. We have used a multi-objective genetic algorithm to optimize PAW pseudopotentials based on accuracy and efficiency. The parameters varied for optimization were cutoff radii and projector energies. In all optimizations, the computational work requirements of pseudopotentials are based on an estimate of floating point operations required to perform electronic structure calculations using those pseudopotentials. Pseudopotential accuracy was determined by calculating lattice constant and bulk modulus and comparing to the lattice constant and bulk modulus given by all-electron DFT. To encourage transferability, pseudopotentials were simultaneously optimized for different crystal systems. Additional pseudopotentials were optimized using an alternative force-based method where pseudopotential accuracy is determined by the interatomic forces in a perturbed crystal system. Motivations for our force-based approach are applications where atoms are not in a perfect-crystal environment, such as defects in lattices, material interfaces, and molecular dynamics.