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Spectral neighbor analysis method for automated generation of quantum-accurate interatomic potentials

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ABSTRACT

We present a new interatomic potential for solids and liquids called Spectral Neighbor Analysis Potential (SNAP). The SNAP potential has a very general form and uses machine-learning techniques to reproduce the energies, forces, and stress tensors of a large set of small configurations of atoms, which are obtained using high-accuracy quantum electronic structure (QM) calculations. The local environment of each atom is characterized by a set of bispectrum components of the local neighbor density projected onto a basis of hyperspherical harmonics in four dimensions. The bispectrum components are the same bond-orientational order parameters employed by the GAP potential [1]. The SNAP potential, unlike GAP, assumes a linear relationship between atom energy and bispectrum components. The linear SNAP coefficients are determined using weighted least-squares linear regression against the full QM training set. This allows the SNAP potential to be fit in a robust, automated manner to large QM data sets using many bispectrum components. The calculation of the bispectrum components and the SNAP potential are implemented in the LAMMPS parallel molecular dynamics code. We demonstrate that a previously unnoticed symmetry property can be exploited to reduce the computational cost of the force calculations by more than one order of magnitude. We present results for a SNAP potential for tantalum, showing that it accurately reproduces a range of commonly calculated properties of both the crystalline solid and the liquid phases. In addition, unlike simpler existing potentials, SNAP correctly predicts the energy barrier for screw dislocation migration in BCC tantalum.

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1. Introduction

Classical molecular dynamics simulation (MD) is a powerful approach for describing the mechanical, chemical, and thermodynamic behavior of solid and fluid materials in a rigorous manner [2]. The material is modeled as a large collection

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of point masses (atoms) whose motion is tracked by integrating the classical equations of motion to obtain the positions and velocities of the atoms at a large number of timesteps. The forces on the atoms are specified by an interatomic potential that defines the potential energy of the system as a function of the atom positions. Typical interatomic potentials are computationally inexpensive and capture the basic physics of electron-mediated atomic interactions of important classes of materials, such as molecular liquids and crystalline metals. Efficient MD codes running on commodity workstations are commonly used to simulate systems with $N = 10^5$–10^6 atoms, the scale at which many interesting physical and chemical phenomena emerge. Quantum molecular dynamics (QMD) is a much more computationally intensive method for solving a similar physics problem [3]. Instead of assuming a fixed interatomic potential, the forces on atoms are obtained by explicitly solving the quantum electronic structure of the valence electrons at each timestep. Because MD potentials are short-ranged, the computational complexity of MD generally scales as $O(N)$, whereas QMD calculations require global self-consistent convergence of the electronic structure, whose computational cost is $O(N_e^\alpha)$, where $2 < \alpha < 3$ and N_e is the number of electrons. For the same reasons, MD is amenable to spatial decomposition on parallel computers, while QMD calculations allow only limited parallelism.

As a result, while high accuracy QMD simulations have supplanted MD in the range $N = 10$–100 atoms, QMD is still intractable for $N > 1000$, even using the largest supercomputers. Conversely, typical MD potentials often exhibit behavior that is inconsistent with QMD simulations. This has led to great interest in the development of MD potentials that match the QMD results for small systems, but can still be scaled to the interesting regime $N = 10^5$–10^6 atoms [1,4,5]. These quantum-accurate potentials require many more floating point operations per atom compared to conventional potentials, but they are still short-ranged. So the computational cost remains $O(N)$, but with a larger algorithm pre-factor.

In this paper, we present a new quantum-accurate potential called SNAP. It is designed to model the migration of screw dislocations in tantalum metal under shear loading, the fundamental process underlying plastic deformation in body-centered cubic metals. In the following section we explain the mathematical structure of the potential and the way in which we fit the potential parameters to a database of quantum electronic structure calculations. We follow that with a brief description of the implementation of the SNAP potential in the LAMMPS code. We demonstrate that a previously unnoticed symmetry property can be exploited to reduce the computational cost of the force calculations by more than one order of magnitude. We then present results for the SNAP potential that we have developed for tantalum. We find that this new potential accurately reproduces a range of properties of solid and liquid tantalum. Unlike simpler potentials, it correctly matches quantum MD results for the screw dislocation core structure and minimum energy pathway for displacement of this structure, properties that were not included in the training database.

2. Mathematical formulation

2.1. Bispectrum components

The quantum mechanical principle of near-sightedness tells us that the electron density at a point is only weakly affected by atoms that are not near. This provides support for the common assumption that the energy of a configuration of atoms is dominated by contributions from clusters of atoms that are near to each other. It is reasonable then to seek out descriptors of local structure and build energy models based on these descriptors. Typically, this is done by identifying geometrical structures, such as pair distances and bond angles, or chemical structures, such as bonds. Interatomic potentials based on these approaches often produce useful qualitative models for different types of materials, but it can be difficult or impossible to adjust these potentials to accurately reproduce known properties of specific materials. Recently, Bartók et al. have studied several infinite classes of descriptors that are related to the density of neighbors in the spherically symmetric space centered on one atom [1,6,7]. They demonstrated that by adding descriptors of successively higher order, it was possible to systematically reduce the mismatch between the potential and the target data. A variant of the GAP formalism has been successfully used to develop several new potentials for tungsten [8]. One of these descriptors, the bispectrum of the neighbor density mapped on to the 3-sphere, forms the basis for their Gaussian Approximation Potential (GAP) [1]. We also use the bispectrum as the basis for our SNAP potential. We derive this bispectrum below, closely following the notation of Ref. [7].

The density of neighbor atoms around a central atom i at location $\mathbf{r}$ can be considered as a sum of δ-functions located in a three-dimensional space:

$$\rho_i(\mathbf{r}) = \delta(\mathbf{r}) + \sum_{r_{ii'} < R_{cut}} f_c(r_{ii'}) w_{i'} \delta(\mathbf{r} - \mathbf{r}_{ii'}) \tag{1}$$

where $\mathbf{r}_{ii'}$ is the vector joining the position of the central atom i to neighbor atom i'. The $w_{i'}$ coefficients are dimensionless weights that are chosen to distinguish atoms of different types, while the central atom is arbitrarily assigned a unit weight. The sum is over all atoms i' within some cutoff distance R_{cut}. The switching function $f_c(r)$ ensures that the contribution of each neighbor atom goes smoothly to zero at R_{cut}. The angular part of this density function can be expanded in the familiar basis of spherical harmonic functions $Y_m^l(\theta, \phi)$, defined for $l = 0, 1, 2, \ldots$ and $m = -l, -l+1, \ldots, l-1, l$ [9]. The

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