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AQCNES: A Quasi-Continuum Non-Equilibrium Solver

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Software

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Summary

The behavior of macroscopic structures is determined by fast atomic interactions at the nanoscales. Current atomic simulation techniques, such as molecular dynamics (MD), are limited to a millions of atoms and hence a few micrometers of domain length. Moreover, finite-temperature vibrational frequencies of around tens of terahertz restrict the time step of MD to femtoseconds, precluding the simulation of problems of engineering interest. Consequently, there has been a significant focus in recent decades on developing multiscale modeling techniques to extend atomistic accuracy to larger length scales and longer time frames. `AQCNES` is a scalable C++-based framework that integrates the spatial-upscaling technique of the quasicontinuum method with the statistical-mechanics-based temporal upscaling technique known as Gaussian phase packets. This enables computationally efficient and robust simulations of large atomistic ensembles at finite temperature.

Statement of need

`AQCNES` is a software that can predict long-term behavior of large atomic ensembles using spatio-temporal upscaling of classical atomistic calculations. It enables the calculation of material properties at finite (non-zero) temperature from atomic scales, offering promising applications in solid-state material science across scales. Current atomic simulation techniques describe the entire ensemble as a collection of particles, each having a position and velocity in the three-dimensional space. This fully refined spatial representation restricts the possible dimensions of the ensemble to microscopic scales. Moreover, case studies of material defects using atomistic simulations are often limited to Molecular Statics (MS) or use unrealistically high loading rates (Homer et al., 2022; Shenoy, 2005). This is unavoidable because finite-temperature MD simulations need long equilibration times and expensive post-processing techniques (Frenkel & Ladd, 1984) to extract relevant thermodynamic information. However, physically relevant material behavior is observed at finite temperature. Therefore, research in the past decades has focused on multiscale modeling techniques to advance atomistic simulations to larger length scales and longer time scales.

To bridge across length scales, `AQCNES` uses a spatial coarse-graining technique known as the quasicontinuum (QC) method (Ronald E. Miller & Tadmor, 2002; Tadmor, 1996). QC exploits the long-range order in crystalline materials to explicitly model a few representative atoms in the domain and uses techniques from continuum-level finite-element modeling to obtain all other atomic degrees of freedom as a function of those of the representative atoms. Fully atomistic resolution is retained in the vicinity of material defects where the long-range order is broken. A technique known as Gaussian Phase Packets (GPP) (Gupta et al., 2021) is used to upscale in time. Instead of the instantaneous phase-space coordinates (position and velocity), statistical averages and variances of these coordinates are tracked in GPP, thus separating the slow

mean atomic motion from fast atomic vibrations. The governing equations of those statistical parameters can be shown to yield configurations which minimize a thermalized Helmholtz free-energy at the temperature of interest. Thus a thermodynamically relaxed structure of the ensemble is obtained after a quasi-static minimization. Hence, this approach is more robust and computationally efficient (Saxena et al., 2022) than tracking individual atomic trajectories. Moreover, it makes the simulation of realistic and quasi-static loading scenarios possible, in contrast to the unrealistically high strain-rates accessible by MD simulations (Vu-Bac et al., 2014; Zhao et al., 2010).

Most of the research groups working in the broad field of upscaling atomistic simulations have their proprietary codes. Two available open-source codes are `QuasiContinuum` (Ron Earle Miller & Tadmor, 2012) based on the QC method and `MultiBench` (Ronald E. Miller & Tadmor, 2009), which is an implementation of fourteen popular spatial upscaling methods. However, both are limited to zero-temperature simulations for crystalline solids in two dimensions. The `MXE` package in `LAMMPS` (Mendez & Ponga, 2021) is an implementation of the temporal upscaling technique `max-ent` for a fully resolved atomic ensemble with no spatial coarse-graining. To the best of the authors' knowledge, there is no such open-source atomistic simulation software that combines the aforementioned spatial and temporal upscaling techniques. `AQCNES` is capable of simulating amorphous materials at atomistic resolution and crystalline materials with spatial coarse-graining, while seamlessly transitioning to atomistic resolution at both zero and non-zero temperatures. It can also simulate large atomic rearrangements induced by severe deformations in multi-resolution domains by using an updated Lagrangian formulation (Gupta et al., 2021). This formulation uses the relaxed state after every load step as the new reference configuration. An adaptive neighborhood calculation strategy (similar to Amelang (2016)) is adopted, where the neighborhoods are regenerated if the maximum relative displacement of a neighbor with respect to a sampling atom exceeds a given buffer radius. This unique combination of features positions `AQCNES` as a versatile and powerful tool in the realm of atomistic simulations.

Detailed documentation of the API can be found on the `AQCNES` website, and simple examples to get started can be found here. Importantly, the use of `AQCNES` is not limited to these examples and the ones listed in the next section.

Example applications

The following gives a summary of the mechanics and material science applications where `AQCNES` has already been used:

- **Surface Elasticity**: Surfaces in solids are the simplest extended defects and contribute to excess energy, leading to an inherent stress associated with them. Their presence also changes the elastic moduli of the solid as compared to those of the bulk solid. `AQCNES` was used by Saxena et al. (2022) to perform a detailed case study of three different crystallographically oriented surfaces for FCC and BCC metals as a function of temperature. The results were also compared against those obtained from state-of-the-art thermodynamic integration techniques (Freitas et al., 2016), suggesting a convincing accuracy. The comparison of computational times taken by MD and `AQCNES` (see Table 5 in Saxena et al. (2022)) shows that there is an approximately fifty-fold computational speedup and a reduction in the computational resources needed for the same simulation.

- **Grain Boundaries**: Grain boundaries (GBs) are regions of crystallographic mismatch between two differently oriented grains. They significantly influence the mechanical and thermal properties of polycrystalline materials. Hence, investigating GB properties via atomic simulations is of scientific interest in the material science community. `AQCNES` has been used to find relaxed energies of $[001]$ and $[011]$ symmetric-tilt GBs (Spínola et al., 2024) in copper as a function of temperature for a range of tilt angles. Different metastable states have been explored for each temperature and tilt angle. In addition, the lowest-energy metastable state was subjected to a quasi-static displacement-driven

shear to obtain the shear coupling factor of all grain boundaries. AQCNES could also identify the Helmholtz free energy of bicrystals, for which the standard thermodynamic integration techniques failed due to hops of the system from one metastable state to another.

- **Nanoindentation**: Nanoindentation is a widely used technique to probe the mechanical properties of materials and nanostructures. AQCNES has been used to simulate the three-dimensional thermo-mechanically-coupled nanoindentation of copper (Gupta et al., 2021). Two layers of spatial coarse graining were used to simulate a cube of side length 0.077µm with 0.2 million representative atoms. The complicated microstructure of prismatic dislocation loops below the nanoindenter could be observed in the finite-temperature simulations at 300 and 600 K. The temperature dependence of the critical indenter force before dislocation nucleation could also be captured.

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