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# AQCNES: A Quasi-Continuum Non-Equilibrium Solver

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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. Existing techniques, such as the quasicontinuum (QC) method, are restricted to spatial upscaling at zero temperature, while temporal upscaling methods like the maximum entropy (max-ent) approach are constrained to fully resolved atomistic simulations at finite temperature.

The software introduced here, `AQCNES`, is a 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. Message Passing Interface (MPI) is employed to enable massive parallelism, which enhances the scalability of the software. This enables computationally efficient and robust simulations of large atomistic ensembles at finite temperature.

## Statement of need

Commonly used atomic simulation techniques describe the entire ensemble as a collection of particles, each having a position and velocity in three-dimensional space. This fully refined spatial representation in state space 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 (Miller & Tadmor, 2009; Wernik & Meguid, 2009).

Most research groups working in the broad field of upscaling atomistic simulations have their proprietary codes. Two available open-source codes are `QuasiContinuum` (Miller & Tadmor, 2012) based on the quasicontinuum (QC) method and `MultiBench` (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 a software that can predict long-term behavior of large atomic ensembles using the 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. In the present implementation, full atomistic resolution is needed for regions of local disorder as well as for amorphous materials, although extension of the QC for amorphous systems have been proposed (Ghareeb & Elbanna, 2020) and can be considered as a possible extension. Hence, AQCNES is capable of simulating both crystalline and amorphous materials in a temporally upscaled fashion at zero and non-zero temperature, while the spatial upscaling capability is limited to crystalline systems. 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 Tembhekar et al. (2017)) 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.

## Functionality

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 $\boldsymbol{q}$ and momentum $\boldsymbol{p}$ ), statistical averages $\{\bar{\boldsymbol{p}}, \bar{\boldsymbol{q}}\}$ and variances $\{\Sigma^{(\boldsymbol{p},\boldsymbol{p})}, \Sigma^{(\boldsymbol{p},\boldsymbol{q})}, \Sigma^{(\boldsymbol{q},\boldsymbol{q})}\}$ of these coordinates are tracked for the entire ensemble in GPP, thus separating the slow mean atomic motion from fast atomic vibrations. The covariance matrices represented above are fully populated for the most general case. However, in order to make the system of equations more tractable, interatomic correlations are assumed to be zero. Further, intra-atomic covariance matrices are assumed to be spherical Gaussian clouds. This leads to

$$\Sigma_i^{(\boldsymbol{p}_i,\boldsymbol{p}_i)} = \Omega_i \mathbf{I}, \qquad \Sigma_i^{(\boldsymbol{p}_i,\boldsymbol{q}_i)} = \beta_i \mathbf{I}, \qquad \Sigma_i^{(\boldsymbol{q}_i,\boldsymbol{q}_i)} = \Sigma_i \mathbf{I}, \tag{1}$$

where subscript $i$ runs over all atoms in the ensemble and $\mathbf{I}$ is an identity matrix in 3D space. Hence, the set of variables $\{\bar{\boldsymbol{p}}_i, \bar{\boldsymbol{q}}_i, \Omega_i, \beta_i, \Sigma_i\}$ is solved for every atomic site. It can be shown that in the quasistatic limit, mean momenta $\bar{\boldsymbol{p}}_i$ and thermal momenta $\beta_i$ vanish for every atom. The mean positions $\{\bar{\boldsymbol{q}}_i : i = 1, \dots, N\}$ and position variances $\{\Sigma_i : i = 1, \dots, N\}$ are obtained using the following equilibrium conditions (Gupta et al., 2021):

$$\begin{aligned} &\langle \boldsymbol{F}_i \rangle = \boldsymbol{0} \qquad \text{and} \\ &\frac{\Omega_i}{m_i} + \frac{\langle \boldsymbol{F}_i(\boldsymbol{q}) \cdot (\boldsymbol{q} - \bar{\boldsymbol{q}}) \rangle}{3} = 0, \end{aligned} \tag{2}$$

where $\boldsymbol{F}_i$ denotes the net force acting on atom $i$ having mass $m_i$. Infomation about momentum variances $\{\Omega_i : i = 1, \dots, N\}$ is obtained from the thermodynamic process which brings the system to equilibrium. The governing equations (2) of these statistical parameters can be shown to yield configurations which minimize a thermalized Helmholtz free-energy at the temperature of interest. The reader is referred to Gupta et al. (2021) for further details. AQCNES uses FIRE (Bitzek et al., 2006) to relax the system of equations (2) and thus obtain a thermodynamically relaxed structure of the ensemble after a quasi-static minimization. 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 quasistatic loading scenarios possible, in contrast to the unrealistically high strain rates accessible by MD simulations (Vu-Bac et al., 2014; Zhao et al., 2010).

| | No. of atoms | No. of procs. | No. of iterations | Simulation time (min) |
|---|---|---|---|---|
| AQCNES | 2624 | 25 | 767 | 9.79 |
| LAMMPS | 16400 | 128 | $4.8 \cdot 10^7$ | 422 |

**Table 1:** Simulation times for a surface free energy computation for the (001) surface in Fe at 300 K with AQCNES and thermodynamic integration in LAMMPS (adapted from Saxena et al. (2022)).

For spatial coarse-graining, AQCNES uses QC (Miller & Tadmor, 2002; Tadmor, 1996). QC exploits the long-range order in crystalline materials to explicitly model $N^h \ll N$ representative(*rep-*) 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. The mean position $\bar{\boldsymbol{q}}_i$ and position variance $\Sigma_i$ of an atom $i$ are obtained by the following interpolation:

$$\bar{\boldsymbol{q}}_i = \sum_{a=1}^{N^h} N_a(\bar{\boldsymbol{X}}_i)\bar{\boldsymbol{q}}_a, \qquad \Sigma_i = \sum_{a=1}^{N^h} N_a(\bar{\boldsymbol{X}}_i)\Sigma_a, \tag{3}$$

where $N_a(\bar{\boldsymbol{X}}_i)$ is the shape function for repatom $a$ evaluated at the position of atom $i$ in the reference configuration. This reduces the number of degrees of freedom to solve from $4N$ to $4N^h$ for finite-temperature simulations. For energy and force calculation in every minimisation iteration, a set of $N^s \ll N$ sampling atoms is selected. The approximate Hamiltonian of the entire system can then be written as:

$$\mathcal{H} \approx \sum_{\alpha=1}^{N^s} w_\alpha \mathcal{H}_\alpha, \tag{4}$$

where $w_\alpha$ are the sampling atom weights and $\mathcal{H}_\alpha$ is the energy of the $\alpha$th sampling atom. A detailed explanation of optimally choosing sampling atom locations and weights can be found in Amelang et al. (2015).

## 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 differently 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), showing convincing accuracy. The comparison of computational times taken by MD and AQCNES (see Table 1) shows that there is an approximately fifty-fold computational speedup and a significant reduction in the computational resources needed for computing the excess surface free energy for an ensemble at finite temperature. Figure 1 shows the surface free energy, stresses, and elastic constants for the (001), (011), and (111) surfaces in BCC iron computed using AQCNES and their comparison with MD simulations and *ab-initio* results by Schönecker et al. (2015). The embedded atom method (EAM) potential developed by Chamati et al. (2006) was used for AQCNES and MD simulations.

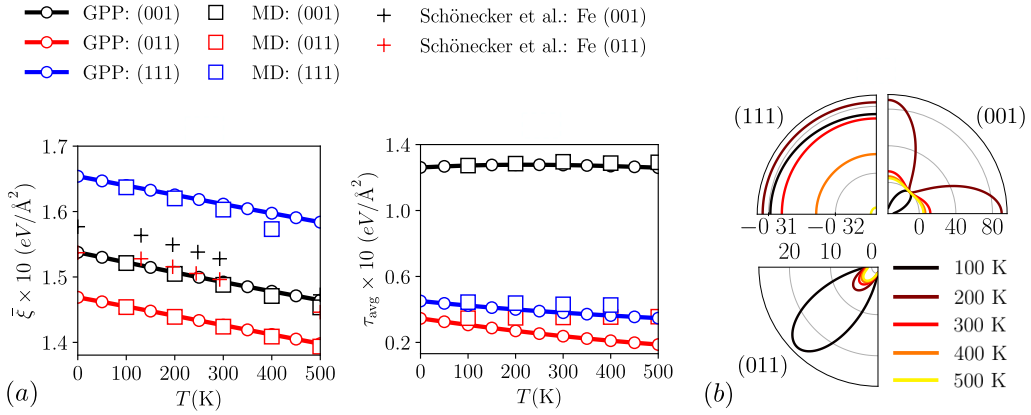


**Figure 1:** (a) Surface free energy density $(\xi)$ and average surface stress $(\tau_{\text{avg}})$ vs. temperature $(T)$ and (b) polar compliance plots (in $\text{Å}^2/eV$) for the (001), (011), and (111) surfaces in BCC iron (adapted from Saxena et al. (2022)).

- **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 in copper as a function of temperature for a range of tilt angles (Spínola et al., 2024). Different metastable states have been explored for each temperature and tilt angle. In addition, the lowest-energy metastable state was subjected to a quasistatic 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. Figure 2 summarizes the excess grain boundary free energy density and the shear coupling factors for the lowest energy metastable state of $[001]$ tilt axis grain boundaries in copper. The EAM potential developed by Mishin et al. (2001) was used for these simulations.

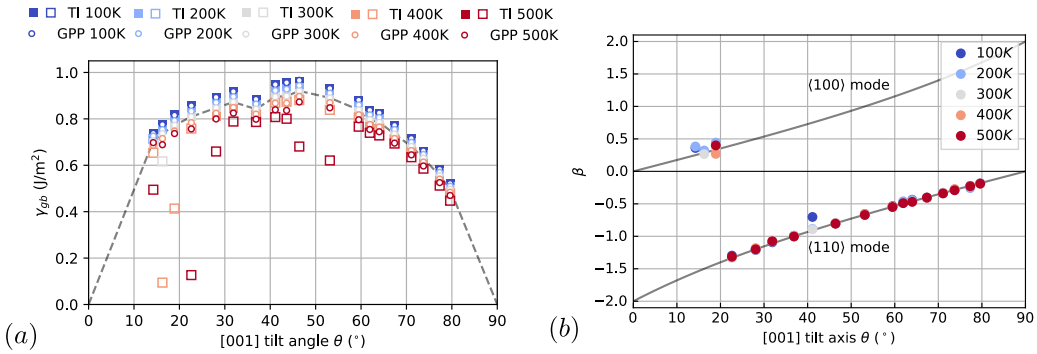


**Figure 2:** (a) Excess grain boundary free energy density $(\gamma_{gb})$ and (b) shear coupling factor $(\beta)$ vs. tilt angle for the $[001]$ symmetric tilt grain boundaries in copper at different temperatures (adapted from Spínola et al. (2024)).

- **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 $0.077\mu$m 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. Figure 3 shows the microstructure below the indenter and the indenter force for different temperatures.

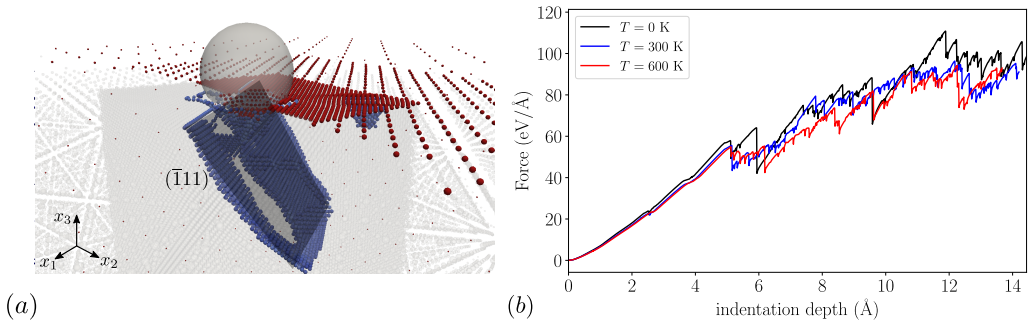


**Figure 3:** (a) Dislocation loops generated as a result of nanoindentation beneath the spherical indenter at a depth of 1 nm. (b) Indenter force vs. depth for isothermal nanoindentation at different temperatures (adapted from Gupta et al. (2021)).

## Dependencies and API documentation

The project uses CMake as its build system generator. The following third party libraries are required and located using CMake's `find_package`.

- Boost (components: program_options, mpi, serialization): version 1.67
- Eigen: version 3.4
- MPI: version 3.1
- PETSc: version 3.15
- CGAL: version 5.4
- PnetCDF: Version 1.12
- nlohmannjson: Version 3.10
- Scotch/ParMETIS
- qcmesh: Version 1.0
- VTK: Version 9.3

The dependency versions mentioned above are not strictly the minimum versions required, but the ones which have been tested to work well. The code quality of the project is analyzed in a CI pipeline which runs inside the development docker container and on the ETH HPC cluster Euler. The pipeline covers the following checks:

- Building the docker container and publishing it to `registry.gitlab.ethz.ch`.
- Building documentation and publishing it to `qc.mm.ethz.ch`.
- Code compilation and running unit/integration tests.
- Presence of a license header for each source file.
- Consistency of code formatting using clang-format for C++ and ruff for Python.
- Static code analysis (linter) using clang-tidy for C++ and ruff for Python.

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 this contribution.

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## References

Amelang, J. S., Venturini, G. N., & Kochmann, D. M. (2015). Summation rules for a fully nonlocal energy-based quasicontinuum method. *Journal of the Mechanics and Physics of Solids*, *82*, 378–413. https://doi.org/10.1016/j.jmps.2015.03.007

Bitzek, E., Koskinen, P., Gähler, F., Moseler, M., & Gumbsch, P. (2006). Structural relaxation made simple. *Physical Review Letters*, *97*(17), 170201. https://doi.org/10.1103/PhysRevLett.97.170201

Chamati, H., Papanicolaou, N., Mishin, Y., & Papaconstantopoulos, D. (2006). Embedded-atom potential for fe and its application to self-diffusion on fe (1 0 0). *Surface Science*, *600*(9), 1793–1803. https://doi.org/10.1016/j.susc.2006.02.010

Freitas, R., Asta, M., & De Koning, M. (2016). Nonequilibrium free-energy calculation of solids using LAMMPS. *Computational Materials Science*, *112*, 333–341. https://doi.org/10.1016/j.commatsci.2015.10.050

Frenkel, D., & Ladd, A. J. (1984). New monte carlo method to compute the free energy of arbitrary solids. Application to the fcc and hcp phases of hard spheres. *The Journal of Chemical Physics*, *81*(7), 3188–3193. https://doi.org/10.1063/1.448024

Ghareeb, A., & Elbanna, A. (2020). An adaptive quasicontinuum approach for modeling fracture in networked materials: Application to modeling of polymer networks. *Journal of the Mechanics and Physics of Solids*, *137*, 103819. https://doi.org/10.1016/j.jmps.2019.103819

Gupta, P., Ortiz, M., & Kochmann, D. M. (2021). Nonequilibrium thermomechanics of gaussian phase packet crystals: Application to the quasistatic quasicontinuum method. *Journal of the Mechanics and Physics of Solids*, *153*, 104495. https://doi.org/10.1016/j.jmps.2021.104495

Homer, E. R., Hart, G. L., Owens, C. B., Hensley, D. M., Spendlove, J. C., & Serafin, L. H. (2022). Examination of computed aluminum grain boundary structures and energies that span the 5D space of crystallographic character. *Acta Materialia*, *234*, 118006. https://doi.org/10.1016/j.actamat.2022.118006

Mendez, J. P., & Ponga, M. (2021). MXE: A package for simulating long-term diffusive mass transport phenomena in nanoscale systems. *Computer Physics Communications*, *260*, 107315. https://doi.org/10.1016/j.cpc.2020.107315

Miller, & Tadmor, E. B. (2002). The quasicontinuum method: Overview, applications and current directions. *Journal of Computer-Aided Materials Design*, *9*(3), 203–239. https://doi.org/10.1023/A:1026098010127

Miller, & Tadmor, E. B. (2009). A unified framework and performance benchmark of fourteen multiscale atomistic/continuum coupling methods. *Modelling and Simulation in Materials Science and Engineering*, *17*(5), 053001. https://doi.org/10.1088/0965-0393/17/5/053001

Miller, & Tadmor, E. B. (2012). *Quasicontinuum method*. https://nanohub.org/resources/qcmethod

Mishin, Y., Mehl, M., Papaconstantopoulos, D., Voter, A., & Kress, J. (2001). Structural stability and lattice defects in copper: Ab initio, tight-binding, and embedded-atom calculations. *Physical Review B*, *63*(22), 224106. https://doi.org/10.1103/PhysRevB.63.

224106

Saxena, S., Spinola, M., Gupta, P., & Kochmann, D. M. (2022). A fast atomistic approach to finite-temperature surface elasticity of crystalline solids. *Computational Materials Science*, *211*, 111511. https://doi.org/10.1016/j.commatsci.2022.111511

Schönecker, S., Li, X., Johansson, B., Kwon, S. K., & Vitos, L. (2015). Thermal surface free energy and stress of iron. *Scientific Reports*, *5*(1), 1–7. https://doi.org/10.1038/srep14860

Shenoy, V. B. (2005). Atomistic calculations of elastic properties of metallic fcc crystal surfaces. *Physical Review B*, *71*(9), 094104. https://doi.org/10.1103/PhysRevB.71.094104

Spínola, M., Saxena, S., Gupta, P., Runnels, B., & Kochmann, D. M. (2024). Finite-temperature grain boundary properties from quasistatic atomistics. *Computational Materials Science*, *244*, 113270. https://doi.org/10.1016/j.commatsci.2024.113270

Tadmor, E. B. (1996). *The quasicontinuum method. Modeling microstructure on multiple length scales: A mixed continuum and atomistics approach*. Brown University.

Tembhekar, I., Amelang, J. S., Munk, L., & Kochmann, D. M. (2017). Automatic adaptivity in the fully nonlocal quasicontinuum method for coarse-grained atomistic simulations. *International Journal for Numerical Methods in Engineering*, *110*(9), 878–900. https://doi.org/10.1002/nme.5438

Vu-Bac, N., Lahmer, T., Keitel, H., Zhao, J., Zhuang, X., & Rabczuk, T. (2014). Stochastic predictions of bulk properties of amorphous polyethylene based on molecular dynamics simulations. *Mechanics of Materials*, *68*, 70–84. https://doi.org/10.1016/j.mechmat.2013.07.021

Wernik, J., & Meguid, S. A. (2009). Coupling atomistics and continuum in solids: Status, prospects, and challenges. *International Journal of Mechanics and Materials in Design*, *5*, 79–110. https://doi.org/10.1007/s10999-008-9087-x

Zhao, J., Nagao, S., & Zhang, Z. (2010). Thermomechanical properties dependence on chain length in bulk polyethylene: Coarse-grained molecular dynamics simulations. *Journal of Materials Research*, *25*, 537–544. https://doi.org/10.1557/JMR.2010.0061