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Computation of per atom strain in classical molecular dynamics simulations

The concept of local atomic-level stress is a powerful tool for describing several material phenomena and holds strong relevance for computational materials science. Its use extends beyond serving as a continuum interpreter of molecular simulations. For example, in metallic glasses, atomic-level stress has been recognized as a tool to characterize the structure, which is difficult to understand purely in terms of geometrical or topological descriptors. In other materials with no long-range order like Concentrated Solid Solution Alloys (CSAs), and amorphous polymers, the macro-scale mechanical properties typically have a direct correlation with their atomic structure due to the absence of grain boundaries and dislocations. Therefore, the atomic-level stress is a powerful tool in understanding diverse phenomena in disordered systems. The atomic-level stress is also very relevant in providing an understanding of the origin of mechanical failure at the atomic level in metals and metal alloys, as well as insight into the behavior of nanomaterials and biomolecules. However, for a complete continuum mechanical interpretation of classical atomistic simulations, it is necessary to compute the corresponding atomic level strains too. While there has expectedly been extensive research in defining and calculating atomic level stresses, research in defining and calculating atomic level strain is relatively scarce.

The most used method to compute local atomic level strains in classical atomistic simulations is by comparison of relative average atomic positions between a current and a reference configuration. Atomic level stress is zero only in a perfect crystal, and is referenced, by definition, to the stress-free state of an atom in a perfect crystal. However, the reference configuration chosen for atomic level strain is user-dependent and generally not the perfect crystal. Therefore, it does not have a one-to-one energetic correspondence to the atomic level stress. Direct application of local elastic constants to the lattice mismatch strain not referenced to the perfect crystal does not account for either structural inhomogeneity or thermal effects and yields highly erroneous results. Furthermore, due to the impossibility of physically applying strain to particular atoms via local displacements, the calculation of local atomic-level strain has been observed to be a very difficult theoretical problem.

In this work, we propose that the covariance of atomic positional fluctuations about equilibrium positions resulting from thermal vibration can be used to arrive at per-atom estimates for elastic components of both thermal strain as well as residual strain from structural inhomogeneity. For the example of a vacancy in an otherwise perfect crystal of fcc Aluminum, we demonstrate using molecular dynamics simulation that strain computed from lattice rearrangements with the reference configuration as the unrelaxed defective crystal results in strains too small in magnitude to cause the observed local residual stresses. We show that atomic positional covariance resulting from thermal vibrations can be used to estimate the elastic part of the total local atomic level strain.