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Local stress calculation in simulations of multicomponent systems

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ABSTRACT

The virial and Hardy methods provide accurate local stresses for single component materials such as monatomic metals. In contrast to the elemental material case, both methods provide poor estimates of the local stress for multicomponent materials. Using binary materials such as CaO, SiC and AlN and homogeneous strain, we demonstrate that there are several sources for the slow convergence of the virial and Hardy local stresses to the bulk values. Different approaches such as enforced stoichiometry, atomic localization functions and the atomic voronoi volume are used to improve the convergence and increase the spatial resolution of the local stress. The virial method with enforced stoichiometry and atomic voronoi volumes is the most accurate, giving exact stress values by the first atomic shell. In the general case, not assuming stoichiometry, the virial method with localization functions converge to 93% of the bulk value by the third atomic shell. This work may be particularly useful for the real-time description of stresses in simulations of shock waves and deformation dynamics.

1. Introduction

The local stress field quantifies the microscopic deformation state in solids. In atomistic simulations, the local stress field can be used to locate individual defects and characterize their structure and dynamics under arbitrary loading conditions. In molecular dynamics (MD) simulations, local stresses have been useful in the analysis of a wide range of phenomena such as film growth [1], shock loading [2–4], dislocation dynamics [5,6], and semiconductor nanopixel relaxation [7,8].

The most common definition of stress used with MD simulations originates from the Virial theorem developed by Clausius [9] and Maxwell [10,11]. Because of its ease of implementation and relatively low computational demands, the virial stress can be computed directly in the main loop in an MD simulation code using the interaction forces between pairs of atoms that are already calculated. Because of its computational convenience, the most widely used definition of local stress is the atomic virial, which is calculated at every atomic position and averaged to give the system or mean stress. Although the virial stress expression can be shown to be rigorous only as an ensemble average over an entire system in thermodynamic equilibrium, the atomic virial has been widely used as a measure of local stress, even in systems far from equilibrium. Nevertheless, the virial stress has been shown to lead to erroneous results in some special cases, such as close to a free surface [12,13].

Instead of starting from the definition of an ensemble average stress, Hardy derived an expression for the local stress, as well as for the local heat flux, from the definition of local mass and momentum density and the continuity equations for

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mass, momentum, and energy conservation [14]. The expression for the local stress resembles the Lutsko expression [15] and also that derived by Irving and Kirkwood [16]. However, it requires that densities and fluxes exactly satisfy local, continuum conservation laws. One of the advantages of the Hardy approach is the arbitrary choice of localization function and average volume to fine tune the resolution of the calculated quantities [17]. The Hardy and virial stresses have been compared for single component systems, subject to deformation and finite temperature, and the Hardy stresses were shown to converge quicker to values expected from continuum theory [18]. The original Hardy formulation assumed only two-body interatomic potentials but recently it was demonstrated by Delph [19] and Chen [20] to also be valid for arbitrary multibody interatomic potentials, thereby extending its applicability to any translational and rotational invariant interatomic potential.

The accuracy of local stress expressions was analyzed for single component systems. However, we find no thorough examinations of their applicability to multi-component systems. While ensemble averages of the stress are independent of the number of atomic components, the local stress can suffer from stoichiometric unbalance in multiple atom unit cells in small average volumes. Therefore, it is important to examine the convergence of the calculation of local stress for multicomponent systems in order to quantify their accuracy and spatial resolution.

In this paper, we discuss the calculation of the local stress for multicomponent systems based on the virial and Hardy methods. As there is no uniqueness in the calculation of local stresses, we quantify the accuracy of these approaches for different systems at different strain loads. In particular, we examine the binary systems CaO, SiC and AlN, subject to homogeneous strains to determine their convergence and computational convenience. Details of the definitions of local stress are presented in Section 2. Details of the calculation of local stress for multicomponent systems, its convergence and computational convenience are discussed in Section 3. The calculation of local density is discussed in Section 4. Finally, we discuss alternative approaches for determining the atomic-level stress in Section 5 and summarize the work in the last section of the paper.

2. Definition of local stresses

The virial stress [9–11] for a system with N atoms and total volume Ω is given by the relation

$$\Pi^{\alpha\beta} = -\frac{1}{\Omega}\sum_{i=1}^{N}\left[\frac{1}{2}\sum_{\substack{j=1\\ j\neq i}}^{N} f_{ij}^{\alpha} r_{ij}^{\beta} + m_i v_i^{\alpha} v_i^{\beta}\right], \tag{1}$$

where $\alpha, \beta = x, y, z$ indicate directions in an orthogonal, laboratory frame, m_i and v_i^{α} are the mass and the α component of the velocity of atom i. r_{ij}^{β} is the β component of the vector connecting atoms i and j, and f_{ij}^{α} is the α component of the force on atom i due to atom j. As is common in MD simulations, this expression can be rewritten as

$$\Pi^{\alpha\beta} = \frac{1}{\Omega}\sum_{i=1}^{N} \Omega_i^a \pi_i^{\alpha\beta}, \tag{2}$$

with

$$\pi_i^{\alpha\beta} = -\frac{1}{\Omega_i^a}\left[\frac{1}{2}\sum_{\substack{j=1\\ j\neq i}}^{N} f_{ij}^{\alpha} r_{ij}^{\beta} + m_i v_i^{\alpha} v_i^{\beta}\right], \tag{3}$$

where $\pi_i^{\alpha\beta}$ and Ω_i^a are the atomic virial stress and the atomic volume associated with atom i. As the fluctuations of the atomic virial stress can be large, the atomic virial stress is often averaged over a volume containing several atoms. This volume can be centered at any given $\mathbf{R}$ position, including the atomic positions, and the local virial stress is calculated using the expression

$$\pi^{\alpha\beta}(\mathbf{R}) = -\frac{1}{\Omega^c}\sum_{\substack{i=1\\ i\in\Omega^c}}^{N}\left[\frac{1}{2}\sum_{\substack{j=1\\ j\neq i}}^{N} f_{ij}^{\alpha} r_{ij}^{\beta} + m_i v_i^{\alpha} v_i^{\beta}\right], \tag{4}$$

where $\Omega^c = \frac{4}{3}\pi r_c^3$ is the spherical volume of radius r_c centered at $\mathbf{R}$.

In the Hardy formulation [14,17], the properties of each atom are spread out in space by use of a localization function $\psi(\mathbf{r}_i - \mathbf{R})$. This function should satisfy the following conditions:

1. non-negative and smooth;
2. peaked at $\mathbf{r}_i = \mathbf{R}$ and decays to zero at $|\mathbf{r}_i - \mathbf{R}| = r_c$;
3. normalized such that $\int \psi d^3\mathbf{r} = 1$.

Hardy [14] defined the mass density and momentum density using this localization function as: