A Mathematical Homogenization Perspective of Virial Stress

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Abstract

A continuum stress measure is derived from molecular dynamics equations using a Generalized Mathematical Homogenization (GMH) theory. GMH consists of solving a coupled fine scale (atomistic unit cell) problem and a coarse scale (continuum) problem. The fine scale problem derived can be interpreted as a molecular statics (at $0^0 K$) problem, where the coarse scale problem derived is a constitutive law-free continuum equation, which calculates the Cauchy stress directly from atomistics. The continuum stress derived is compared to various versions of the virial stress formula.

1 Introduction

An important issue in bridging atomistic and continuum scales is the calculation of continuum fields from atomistics. The local virial stress $\mathbf{\Pi}_i$ is commonly used to find the local or atomic level stress in molecular dynamics computations. It was developed by Clausius [1] and Maxwell [2] and gives the local stress around an atom i

$$\mathbf{\Pi}_i = \frac{1}{\Theta_i}\left(-m_i \dot{\mathbf{u}}_i \otimes \dot{\mathbf{u}}_i + \frac{1}{2}\sum_{j\neq i} \mathbf{x}_{ij} \otimes \mathbf{f}_{ij}\right) \tag{1}$$

where m_i is the mass of atom i, $\mathbf{x}_i$ its position, $\mathbf{u}_i$ its displacement; $\dot{\mathbf{u}}_i = d\mathbf{u}_i / dt$ the velocity ; $\mathbf{x}_{ij} = \mathbf{x}_j - \mathbf{x}_i$; $\otimes$ denotes the tensor or dyadic product of two vectors; Θ_i is the volume around atom i and $\mathbf{f}_{ij}$ designates the interatomic force between atoms i and j computed as

$$\mathbf{f}_{ij} = \frac{\partial \Phi\left(x_{ij}\right)}{\partial x_{ij}} \frac{\mathbf{x}_{ij}}{x_{ij}} \tag{2}$$

where $x_{ij} = \left|\mathbf{x}_{ij}\right|$ is the distance between atoms i and j; Φ is the interatomic potential or the energy of the atomic ensemble; $\mathbf{x}_{ij} / x_{ij}$ is the unit vector in the direction of $\mathbf{x}_{ij}$. In Eq. (2) for simplicity we considered the pairwise interatomic potential, which may be inadequate for solids.

The first term in Eq. (1) reflects the notion that motion of atoms across a fixed surface applies pressure. The second term arises from interatomic forces. The first (kinetic) term is small compared to the second (mechanical) for solids but is dominant for gases. The local version of the virial stress has been shown to be erroneous in several cases. For instance, Cheung and Yip [3] have shown that for crystals with free surface the normal component of the local virial stress does not vanish for atoms at the outer surface. To circumvent some of the deficiencies found in local or point wise stress values, Cormier *et al.* [4] based on the work of Lutsko [5] introduced the average virial stress. The average virial stress, $\mathbf{\Pi}$, in a representative volume Θ containing a number of atoms is given by

$$\mathbf{\Pi} = \frac{1}{\Theta}\sum_{\mathbf{x}_i \in \Theta}\left(\Theta_i \mathbf{\Pi}_i\right) = \frac{1}{\Theta}\sum_{\mathbf{x}_i \in \Theta}\left(-m_i \dot{\mathbf{u}}_i \otimes \dot{\mathbf{u}}_i + \frac{1}{2}\sum_{j \neq i}\mathbf{x}_{ij} \otimes \mathbf{f}_{ij}\right) \tag{3}$$

Some authors argued that only the fluctuation part of the atomistic velocity should be used in the virial formulas (1) and (3). This version has been suggested by Irving and Kirkwood [6] and Hardy [7] among others. It modifies the expression of the average virial stress as

$$\hat{\mathbf{\Pi}} = \frac{1}{\Theta}\sum_{\mathbf{x}_i \in \Theta}\left(-m_i\left(\dot{\mathbf{u}}_i - \dot{\bar{\mathbf{u}}}\right) \otimes \left(\dot{\mathbf{u}}_i - \dot{\bar{\mathbf{u}}}\right) + \frac{1}{2}\sum_{j \neq i}\mathbf{x}_{ij} \otimes \mathbf{f}_{ij}\right) \tag{4}$$

where $\dot{\bar{\mathbf{u}}}$ is the local average velocity in Θ.

Recently Zhou [8] and Zhou and McDowell [9] have cast some doubt on the validity of the first, dynamical, term in the average virial formulas (3),(4) since the desired continuum Cauchy stress is supposed to represent mechanical forces only. Based on the conservation principles they suggested that the Cauchy stress $\boldsymbol{\sigma}$ be equal to the mechanical term in the average virial formula

$$\boldsymbol{\sigma} = \frac{1}{2\Theta}\sum_{\mathbf{x}_i \in \Theta}\sum_{j \neq i}\mathbf{x}_{ij} \otimes \mathbf{f}_{ij} \tag{5}$$

It is important to note that many authors including Srolovitz *et al.* [10], Horstemeyer and Baskes [11] and Alber *et al.* [12] among others adopted (5) as a measure of the continuum stress.

In this paper, we derive the Cauchy stress expression directly from the molecular dynamics equations using the Generalized Mathematical Homogenization (GMH) theory developed in Section 3. The theory derives a coupled atomistic-continuum problem where the fine scale (atomistic) can be interpreted as a molecular statics (at $0^0 K$) problem, while the coarse scale is a constitutive law-free continuum equation, which gives the overall or Cauchy stress directly from atomistics. This multiscale approach is closely related to the Heterogeneous Multiscale Method (HMM) introduced by E and Enquist [13]. The main difference between the GMH and HMM is that the coarse and fine scale problems in GMH are derived directly from atomistics without making any *a priori* assumption about the mathematical structure of these equations.

2 Governing Equations

2.1 Molecular dynamics equations of motion

We consider a periodic atomistic system composed of N atoms. The initial position of atom i in the reference configuration is denoted by $\mathbf{X}_i$, $i=1,2,\cdots,N$. The displacement of atom i with respect to the reference position is designated as $\mathbf{u}_i$. Upon deformation, the new position of atom i is $\mathbf{x}_i$ given by

$$\mathbf{x}_i = \mathbf{X}_i + \mathbf{u}_i, \quad \mathbf{u}_i = \mathbf{u}_i(\mathbf{X}_i, t) \tag{6}$$

or in spatial description as

$$\mathbf{X}_i = \mathbf{x}_i - \mathbf{u}_i, \quad \mathbf{u}_i = \mathbf{u}_i(\mathbf{x}_i, t) \tag{7}$$

The vector separating two atoms i and j in the reference configuration is given by

$$\mathbf{X}_{ij} = \mathbf{X}_j - \mathbf{X}_i \tag{8}$$

The corresponding vector separating the two atoms in the deformed configuration is

$$\mathbf{x}_{ij} = \mathbf{x}_j - \mathbf{x}_i \tag{9}$$

Substituting Eq. (7) into (9) yields

$$\mathbf{x}_{ij} = \mathbf{X}_j - \mathbf{X}_i + \mathbf{u}_j - \mathbf{u}_i = \mathbf{X}_{ij} + \mathbf{u}_j - \mathbf{u}_i \tag{10}$$

Hereafter the Roman subscripts i and j are reserved for atom labels and will not be subject to the summation convention. Spatial directions, for which summation convention over repeated indices is enforced, will be denoted by Greek subscripts.

For simplicity, we focus our attention on the pairwise potentials. However, the formulation can be extended to the three-body potentials and the EAM (Embedded-atom method) potentials [14]. We refer to [15, 16] for additional references. For pairwise potentials, the interaction between atoms i and j is depicted by $\Phi(x_{ij})$. The interatomic force $\mathbf{f}_{ij}$ applied on atom i by atom j is evaluated based on Eq. (2).

The equation of motion for atom i can be written as

$$m_i \ddot{\mathbf{u}}_i = \sum_{j(\neq i)} \mathbf{f}_{ij}(\mathbf{x}_{ij}) \tag{11}$$

$\dot{\mathbf{u}}_i = d\mathbf{u}_i / dt$ represents the material time derivative of $\mathbf{u}_i$. The label j denotes the neighboring atoms which interact with atom i, such that $\left|\mathbf{x}_j - \mathbf{x}_i\right| < r_c$, with r_c being the cutoff radius. For stability issues related to the selection of the cutoff radius we refer to [17]. For simplicity, the initial external forces are absent.

Due to periodic atomic structure, the mass of the atom m_i and the interatomic forces $\mathbf{f}_{ij}$ are assumed to be periodic. Attention is restricted to the case where the wavelength of the traveling signal λ is much larger than the size of the unit cell l, i.e., $\varepsilon = l/\lambda \ll 1$.

2.2 Multiple spatial scales and rescaling of the MD equations

Due to the rapidly varying interatomic potentials, two distinct spatial coordinates are employed to describe the heterogeneity at the atomistic level: (i) the coarse scale denoted by $\mathbf{x}$, at which the atomistic features are invisible, and (ii) the atomistic scale or fine scale, denoted by $\mathbf{y}$. The two scales are related by

$$\mathbf{y} = \mathbf{x}/\varepsilon \qquad 0 < \varepsilon \ll 1 \tag{12}$$

The corresponding scales in the reference configuration are denoted by $\mathbf{X}$ and $\mathbf{Y}$, respectively and are related by $\mathbf{Y} = \mathbf{X}/\varepsilon$. The resulting displacement field and its derivatives are functions of $\mathbf{x}, \mathbf{y}$ and t. Solution dependence on the fast time scale for finite temperature applications is discussed in [18].

Prior to carrying out the multiple scale asymptotic analysis it is necessary to rescale the molecular dynamics (MD) Eq. (11). We start by considering continuum equations of motion $\rho\ddot{\mathbf{u}}(\mathbf{x})-\nabla\cdot\boldsymbol{\sigma}=0$, where ρ is the mass density; $\boldsymbol{\sigma}$ the Cauchy stress tensor and $\nabla\cdot\boldsymbol{\sigma}=\mathrm{div}\boldsymbol{\sigma}$ denoting the divergence of the Cauchy stress tensor. For homogeneous media, stress derivatives are of order one, whereas for heterogeneous systems, certain components of stresses may be discontinuous, and therefore stress derivatives could be of $O(\varepsilon^{-1})$. Assuming $\rho\sim O(1)$ and the characteristic size of the unit cell $l\sim O(\varepsilon)$ then the volume of the unit cell $\Theta\sim O(\varepsilon^3)$ and $m_i\sim\rho\Theta\sim O(\varepsilon^3)$. For more details on rescaling, we refer to [21]. Dividing Eq. (11) by the volume of the unit cell, yields

$$k_1\rho\ddot{\mathbf{u}}_i(\mathbf{x}_i,\mathbf{y}_i,t)=\frac{1}{k_2\varepsilon^3}\sum_{j(\neq i)}\mathbf{f}_{ij}(\mathbf{x}_{ij}) \tag{13}$$

where k_1 and k_2 are *O(1)* constants. Comparing Eq. (13) to the continuum equation of motion it follows that

$$\mathbf{f}_{ij}(\mathbf{x}_{ij})\sim O(\varepsilon^2) \tag{14}$$

To this end, we introduce the following $O(1)$ quantities:

$$\bar{m}=m/\varepsilon^3\sim O(1),\qquad \bar{\mathbf{f}}_{ij}(\mathbf{x}_{ij})=\mathbf{f}_{ij}(\mathbf{x}_{ij})/\varepsilon^2\sim O(1) \tag{15}$$

Due to periodicity of masses, we have $\bar{m}=\bar{m}(\mathbf{y})$. The rescaled MD Eq. (11) can be expressed as:

$$\bar{m}_i(\mathbf{y})\ddot{\mathbf{u}}_i(\mathbf{x}_i,\mathbf{y}_i,t)=\frac{1}{\varepsilon}\sum_{j(\neq i)}\bar{\mathbf{f}}_{ij}(\mathbf{x}_{ij}) \tag{16}$$

where $\mathbf{u}_i(\mathbf{x}_i,\mathbf{y}_i,t)\sim O(1)$ in the stretched coordinate system $\mathbf{y}$.

3 Generalized Mathematical Homogenization

3.1 Multiple-scale asymptotic analysis

We assume that the coarse scale coordinate $\mathbf{x}$ takes continuous series of values and displacements $\mathbf{u}_i(\mathbf{x}_i,\mathbf{y}_i,t)$ are continuous and differentiable in $\mathbf{x}$, while the fine scale

coordinate $\mathbf{y}$ is discrete. We denote the displacement at atom i by $\mathbf{u}(\mathbf{x},\mathbf{y}_i,t)$ with $\mathbf{x}=\mathbf{x}_i$. The displacements of the neighboring atoms $\mathbf{u}_j(\mathbf{x}_j,\mathbf{y}_j,t)$ are expanded using Taylor series around the point $\mathbf{x}$

$$\mathbf{u}_j=\mathbf{u}_j(\mathbf{x}_j,\mathbf{y}_j,t)$$

$$=\mathbf{u}(\mathbf{x},\mathbf{y}_j,t)+\nabla_{\mathbf{x}}\mathbf{u}(\mathbf{x},\mathbf{y}_j,t)\cdot(\mathbf{x}_j-\mathbf{x}_i)+\frac{1}{2}\nabla_{\mathbf{x}}\nabla_{\mathbf{x}}\mathbf{u}(\mathbf{x},\mathbf{y}_j,t):((\mathbf{x}_j-\mathbf{x}_i)\otimes(\mathbf{x}_j-\mathbf{x}_i))+\cdots \quad (17)$$

where the dot denotes contraction and $\otimes$ designates dyadic or tensor product. In the indicial notation, the components of the gradient of the displacement field $\mathbf{u}$ with respect to the deformed coarse scale configuration $\mathbf{x}$ are given as

$$[\nabla_{\mathbf{x}}\mathbf{u}(\mathbf{x},\mathbf{y}_j,t)]_{\alpha\beta}=\frac{\partial u_\alpha(\mathbf{x},\mathbf{y}_j,t)}{\partial x_\beta} \quad (18)$$

and subsequently, Eq. (17) can be rewritten as

$$u_{j\alpha}=u_{j\alpha}(\mathbf{x}_j,\mathbf{y}_j,t)=u_\alpha(\mathbf{x},\mathbf{y}_j,t)+\frac{\partial u_\alpha(\mathbf{x},\mathbf{y}_j,t)}{\partial x_\beta}(x_{j\beta}-x_{i\beta})+\frac{1}{2}\frac{\partial^2 u_\alpha(\mathbf{x},\mathbf{y}_j,t)}{\partial x_\beta\partial x_\gamma}(x_{j\beta}-x_{i\beta})(x_{j\gamma}-x_{i\gamma})+\cdots \quad (19)$$

From Eq. (17) we have

$$\mathbf{u}_j-\mathbf{u}_i=\mathbf{u}_j(\mathbf{x}_j,\mathbf{y}_j,t)-\mathbf{u}_i(\mathbf{x}_i,\mathbf{y}_i,t)$$

$$=\mathbf{u}(\mathbf{x},\mathbf{y}_j,t)-\mathbf{u}(\mathbf{x},\mathbf{y}_i,t)+\nabla_{\mathbf{x}}\mathbf{u}(\mathbf{x},\mathbf{y}_j,t)\cdot(\mathbf{x}_j-\mathbf{x}_i)+$$

$$\frac{1}{2}\nabla_{\mathbf{x}}\nabla_{\mathbf{x}}\mathbf{u}(\mathbf{x},\mathbf{y}_j,t):((\mathbf{x}_j-\mathbf{x}_i)\otimes(\mathbf{x}_j-\mathbf{x}_i))+\cdots \quad (20)$$

Since the coarse and fine scales coordinates are related by Eq. (12), we have

$$\mathbf{x}_{ij}=\mathbf{x}_j-\mathbf{x}_i=\varepsilon(\mathbf{y}_j-\mathbf{y}_i)=\varepsilon\mathbf{y}_{ij} \quad (21)$$

Inserting Eq. (21) into (20) yields

$$\mathbf{u}_j - \mathbf{u}_i = \mathbf{u}_j(\mathbf{x}_j, \mathbf{y}_j, t) - \mathbf{u}_i(\mathbf{x}_i, \mathbf{y}_i, t)$$

$$= \mathbf{u}(\mathbf{x}, \mathbf{y}_j, t) - \mathbf{u}(\mathbf{x}, \mathbf{y}_i, t) + \varepsilon \nabla_{\mathbf{x}} \mathbf{u}(\mathbf{x}, \mathbf{y}_j, t) \cdot \mathbf{y}_{ij} + \varepsilon^2 \frac{1}{2} \nabla_{\mathbf{x}} \nabla_{\mathbf{x}} \mathbf{u}(\mathbf{x}, \mathbf{y}_j, t) : (\mathbf{y}_{ij} \otimes \mathbf{y}_{ij}) + \cdots \quad (22)$$

A multiple scale asymptotic expansion is employed to approximate the displacement as:

$$\mathbf{u}(\mathbf{x}, \mathbf{y}, t) = \mathbf{u}^0(\mathbf{x}, t) + \varepsilon \mathbf{u}^1(\mathbf{x}, \mathbf{y}, t) + \cdots \quad (23)$$

where the leading order term $\mathbf{u}^0$ is assumed to be independent of the fine scale coordinate $\mathbf{y}$; In Eq. (23) only the first two terms in the asymptotic expansion are considered whereas the remaining higher order terms are neglected.

Substituting the asymptotic expansion (23) into Eq. (22) yields

$$\mathbf{u}_j - \mathbf{u}_i = \varepsilon[\mathbf{u}^1(\mathbf{x}, \mathbf{y}_j) - \mathbf{u}^1(\mathbf{x}, \mathbf{y}_i) + \nabla_{\mathbf{x}} \mathbf{u}^0(\mathbf{x}) \cdot \mathbf{y}_{ij}] +$$

$$\varepsilon^2 [\nabla_{\mathbf{x}} \mathbf{u}^1(\mathbf{x}, \mathbf{y}_j) \cdot \mathbf{y}_{ij} + \frac{1}{2} \nabla_{\mathbf{x}} \nabla_{\mathbf{x}} \mathbf{u}^0(\mathbf{x}) : (\mathbf{y}_{ij} \otimes \mathbf{y}_{ij})] + \cdots \quad (24)$$

where for notation brevity we omit the dependence of displacements on time t.

The position vector separating two atoms i and j in the reference configuration expressed in the stretched coordinate $\mathbf{y}$ is denoted by

$$\mathbf{Y}_{ij} = \mathbf{Y}_j - \mathbf{Y}_i = (\mathbf{X}_j - \mathbf{X}_i)/\varepsilon = \mathbf{X}_{ij}/\varepsilon \quad (25)$$

where $\mathbf{Y}_{ij} \sim O(1)$.

Substituting Eqs. (24) and (25) into (10) yields

$$\mathbf{x}_{ij} = \mathbf{X}_{ij} + \mathbf{u}_j - \mathbf{u}_i = \varepsilon \boldsymbol{\phi}_{ij}(\mathbf{x}, \mathbf{y}) + \varepsilon^2 \boldsymbol{\psi}_{ij}(\mathbf{x}, \mathbf{y}) + \cdots \quad (26)$$

where

$$\phi_{ij}(\mathbf{x}, \mathbf{y}) = \mathbf{Y}_{ij} + \mathbf{u}^1(\mathbf{x}, \mathbf{y}_j) - \mathbf{u}^1(\mathbf{x}, \mathbf{y}_i) + \nabla_{\mathbf{x}} \mathbf{u}^0(\mathbf{x}) \cdot \mathbf{y}_{ij} \quad (27)$$

$$\psi_{ij}(\mathbf{x}, \mathbf{y}) = \nabla_{\mathbf{x}} \mathbf{u}^1(\mathbf{x}, \mathbf{y}_j) \cdot \mathbf{y}_{ij} + \frac{1}{2} \nabla_{\mathbf{x}} \nabla_{\mathbf{x}} \mathbf{u}^0(\mathbf{x}) : (\mathbf{y}_{ij} \otimes \mathbf{y}_{ij}) \quad (28)$$

Since $\boldsymbol{\phi}_{ij}(\mathbf{x},\mathbf{y}) \sim O(1)$ and $\boldsymbol{\psi}_{ij}(\mathbf{x},\mathbf{y}) \sim O(1)$, we have

$$\frac{\left\|\varepsilon^2 \boldsymbol{\psi}_{ij}(\mathbf{x},\mathbf{y})\right\|}{\left\|\varepsilon \boldsymbol{\phi}_{ij}(\mathbf{x},\mathbf{y})\right\|} \sim O(\varepsilon) \tag{29}$$

where $\|\cdot\|$ denotes the vector norm. Due to (29) the normalized interatomic force can be expanded around $\varepsilon\boldsymbol{\phi}_{ij}$ as

$$\begin{aligned}\overline{\mathbf{f}}_{ij}(\mathbf{x}_{ij}) &= \overline{\mathbf{f}}_{ij}(\varepsilon\boldsymbol{\phi}_{ij} + \varepsilon^2\boldsymbol{\psi}_{ij} + \cdots) \\ &= \overline{\mathbf{f}}_{ij}(\hat{\boldsymbol{\phi}}_{ij}) + \frac{\partial \overline{\mathbf{f}}_{ij}}{\partial \hat{\boldsymbol{\phi}}_{ij}} \cdot (\varepsilon^2\boldsymbol{\psi}_{ij} + \cdots) + o(\varepsilon^2\boldsymbol{\psi}_{ij} + \cdots)\end{aligned} \tag{30}$$

where

$$\hat{\boldsymbol{\phi}}_{ij}(\mathbf{x},\mathbf{y}) = \varepsilon\boldsymbol{\phi}_{ij}(\mathbf{x},\mathbf{y}) \tag{31}$$

We further define the normalized gradient $\overline{\mathbf{f}}'_{ij} \sim O(1)$ as

$$\overline{\mathbf{f}}'_{ij} = \frac{\partial \overline{\mathbf{f}}_{ij}}{\partial \boldsymbol{\phi}_{ij}} = \varepsilon \frac{\partial \overline{\mathbf{f}}_{ij}}{\partial \hat{\boldsymbol{\phi}}_{ij}} \tag{32}$$

Equation (30) can be rearranged as

$$\overline{\mathbf{f}}_{ij}(\mathbf{x}_{ij}) = \overline{\mathbf{f}}_{ij}(\hat{\boldsymbol{\phi}}_{ij}) + \overline{\mathbf{f}}'_{ij}(\hat{\boldsymbol{\phi}}_{ij}) \cdot (\varepsilon\boldsymbol{\psi}_{ij} + \cdots) + o(\varepsilon^2\boldsymbol{\psi}_{ij} + \cdots) \tag{33}$$

Substituting the asymptotic expansion (23) and Eq. (33) into the rescaled molecular dynamics equations of motion (16) yields

$$\bar{m}_i(\mathbf{y})[\ddot{\mathbf{u}}^0(\mathbf{x},t) + \varepsilon\ddot{\mathbf{u}}^1(\mathbf{x},\mathbf{y}_i,t) + \cdots] = \frac{1}{\varepsilon}\sum_{j(\neq i)}[\overline{\mathbf{f}}_{ij}(\hat{\phi}_{ij}) + \overline{\mathbf{f}}'_{ij}(\hat{\boldsymbol{\phi}}_{ij}) \cdot (\varepsilon\boldsymbol{\psi}_{ij} + \cdots) + o(\varepsilon^2\boldsymbol{\psi}_{ij} + \cdots)] \tag{34}$$

Collecting terms of equal power of ε, gives the equilibrium equations at different orders starting at $O(\varepsilon^{-1})$:

$$O(\varepsilon^{-1}): \quad \sum_{j(\neq i)} \overline{\mathbf{f}}_{ij}(\hat{\boldsymbol{\phi}}_{ij}) = 0 \tag{35}$$

$$O(\varepsilon^0): \quad \bar{m}_i(\mathbf{y})\ddot{\mathbf{u}}^0(\mathbf{x},t) = \sum_{j(\neq i)} [\bar{\mathbf{f}}'_{ij}(\hat{\boldsymbol{\phi}}_{ij}) \cdot \boldsymbol{\psi}_{ij}] \tag{36}$$

3.2 The atomistic unit cell problem

Consider the $O(\varepsilon^{-1})$ equilibrium equation (35) first. Substituting the normalized interatomic force vector (15) into Eq. (35) yields

$$\sum_{j(\neq i)} \mathbf{f}_{ij}(\hat{\boldsymbol{\phi}}_{ij}) = 0 \qquad \forall i \tag{37}$$

From Eq. (27) we have

$$\hat{\boldsymbol{\phi}}_{ij} = \varepsilon\boldsymbol{\phi}_{ij}(\mathbf{x},\mathbf{y}) = \varepsilon[\mathbf{Y}_{ij} + \mathbf{u}^1(\mathbf{x},\mathbf{y}_j) - \mathbf{u}^1(\mathbf{x},\mathbf{y}_i) + \nabla_{\mathbf{x}}\mathbf{u}^0(\mathbf{x}) \cdot \mathbf{y}_{ij}]$$

$$= \mathbf{X}_{ij} + \hat{\mathbf{u}}^1(\mathbf{x},\mathbf{y}_j) - \hat{\mathbf{u}}^1(\mathbf{x},\mathbf{y}_i) + \nabla_{\mathbf{x}}\mathbf{u}^0(\mathbf{x}) \cdot \mathbf{x}_{ij} \tag{38}$$

where

$$\hat{\mathbf{u}}^1(\mathbf{x},\mathbf{y}) = \varepsilon\mathbf{u}^1(\mathbf{x},\mathbf{y}) \tag{39}$$

From Eqs. (9), (21), (26) and (31), we have

$$\varepsilon\mathbf{y}_{ij} = \mathbf{x}_{ij} \cong \varepsilon\boldsymbol{\phi}_{ij}(\mathbf{x},\mathbf{y}) = \hat{\boldsymbol{\phi}}_{ij}(\mathbf{x},\mathbf{y}) \tag{40}$$

Inserting Eq. (40) into (38) yields

$$(\mathbf{1} - \nabla_{\mathbf{x}}\mathbf{u}^0(\mathbf{x})) \cdot \hat{\boldsymbol{\phi}}_{ij}(\mathbf{x},\mathbf{y}) = \mathbf{X}_{ij} + \hat{\mathbf{u}}^1(\mathbf{x},\mathbf{y}_j) - \hat{\mathbf{u}}^1(\mathbf{x},\mathbf{y}_i) \tag{41}$$

where $\mathbf{1}$ is the second order identity tensor. Defining the inverse of the coarse scale deformation gradient as $\left(\mathbf{F}^0\right)^{-1} = \mathbf{1} - \boldsymbol{\nabla}_{\mathbf{x}}\mathbf{u}^0(\mathbf{x})$, it follows that

$$\hat{\boldsymbol{\phi}}_{ij}(\mathbf{x},\mathbf{y}) = \mathbf{F}^0(\mathbf{x}) \cdot (\mathbf{X}_{ij} + \hat{\mathbf{u}}^1(\mathbf{x},\mathbf{y}_j) - \hat{\mathbf{u}}^1(\mathbf{x},\mathbf{y}_i)) \tag{42}$$

Inserting Eq. (42) into (37) yields the atomistic unit cell problem

$$\boxed{\sum_{j(\neq i)} \mathbf{f}_{ij}\left(\mathbf{F}^0(\mathbf{x}) \cdot \left[\mathbf{X}_{ij} + \hat{\mathbf{u}}^1(\mathbf{x},\mathbf{y}_j) - \hat{\mathbf{u}}^1(\mathbf{x},\mathbf{y}_i)\right]\right) = 0 \qquad \forall i} \tag{43}$$

Motion of atoms is governed by two sources: (i) the uniform coarse scale deformation gradient, $\mathbf{F}^0$, and (ii) the fine scale correction $\mathbf{u}^1(\mathbf{x},\mathbf{y})$ induced by the heterogeneity of the atomistic structure. This fine-scale contribution $\mathbf{u}^1$ can be interpreted as a correction to the classical Cauchy-Born rule. Eq. (43) reflects the fact (*c.f.* Born & Huang [19]) that when a macroscopically uniform strain is applied on the atomistic cell, the deformation field is generally nonuniform, i.e., an internal relaxation occurs and the corresponding inhomogeneous atomic displacements are determined by the equilibrium condition of each atom in the cell [12].

3.3 The coarse scale equations of motion

We proceed by considering the $O(\varepsilon^0)$ equilibrium equation (36). Substituting the normalized quantities (15) into (36) yields

$$m_i(\mathbf{y})\ddot{\mathbf{u}}^0(\mathbf{x},t) = \varepsilon \sum_{j(\neq i)} [\mathbf{f}'_{ij}(\hat{\boldsymbol{\phi}}_{ij}) \cdot \boldsymbol{\psi}_{ij}] \quad \forall i \tag{44}$$

Summing Eq. (44) for all atoms in the unit cell and dividing the resulting equation by the volume of the unit cell, Θ, yields

$$\frac{1}{\Theta}\sum_i m_i(\mathbf{y})\ddot{\mathbf{u}}^0(\mathbf{x},t) = \frac{\varepsilon}{\Theta}\sum_i \sum_{j(\neq i)} [\mathbf{f}'_{ij}(\hat{\boldsymbol{\phi}}_{ij}) \cdot \boldsymbol{\psi}_{ij}] \tag{45}$$

Since the above summation is performed over the unit cell, the coarse scale field $\mathbf{u}^0(\mathbf{x},t)$ is taken as constant for all the atoms in the unit cell. Recalling the definition of mass density as

$$\rho = \frac{1}{\Theta}\sum_i m_i(\mathbf{y}) \tag{46}$$

then Eq. (45) can be written as

$$\rho\ddot{\mathbf{u}}^0(\mathbf{x},t) = \frac{\varepsilon}{\Theta}\sum_i \sum_{j(\neq i)} [\mathbf{f}'_{ij}(\hat{\boldsymbol{\phi}}_{ij}) \cdot \boldsymbol{\psi}_{ij}] \tag{47}$$

Further exploiting the chain rule yields

$$\nabla_{\mathbf{x}}\mathbf{f}_{ij} = \frac{\partial \mathbf{f}_{ij}}{\partial \mathbf{x}} = \frac{\partial \mathbf{f}_{ij}}{\partial \boldsymbol{\phi}_{ij}} \cdot \frac{\partial \boldsymbol{\phi}_{ij}}{\partial \mathbf{x}} = \mathbf{f}'_{ij} \cdot \nabla_{\mathbf{x}}\boldsymbol{\phi}_{ij} \tag{48}$$

From Eq. (38), we have

$$\begin{aligned}\nabla_{\mathbf{x}}\boldsymbol{\phi}_{ij} &= \nabla_{\mathbf{x}}\left[\mathbf{Y}_{ij} + \mathbf{u}^1(\mathbf{x},\mathbf{y}_j) - \mathbf{u}^1(\mathbf{x},\mathbf{y}_i) + \nabla_{\mathbf{x}}\mathbf{u}^0(\mathbf{x})\cdot\mathbf{y}_{ij}\right] \\ &= \nabla_{\mathbf{x}}\mathbf{u}^1(\mathbf{x},\mathbf{y}_j) - \nabla_{\mathbf{x}}\mathbf{u}^1(\mathbf{x},\mathbf{y}_i) + \nabla_{\mathbf{x}}\nabla_{\mathbf{x}}\mathbf{u}^0(\mathbf{x})\cdot\mathbf{y}_{ij}\end{aligned} \tag{49}$$

Substituting Eq. (49) into (48) yields

$$\mathbf{f}'_{ij}\cdot[\nabla_{\mathbf{x}}\mathbf{u}^1(\mathbf{x},\mathbf{y}_j) - \nabla_{\mathbf{x}}\mathbf{u}^1(\mathbf{x},\mathbf{y}_i) + \nabla_{\mathbf{x}}\nabla_{\mathbf{x}}\mathbf{u}^0(\mathbf{x})\cdot\mathbf{y}_{ij}] = \nabla_{\mathbf{x}}\mathbf{f}_{ij} \tag{50}$$

From Eq. (28), we have

$$\begin{aligned}\mathbf{f}'_{ij}\cdot\boldsymbol{\psi}_{ij} &= \mathbf{f}'_{ij}\cdot[\nabla_{\mathbf{x}}\mathbf{u}^1(\mathbf{x},\mathbf{y}_j)\cdot\mathbf{y}_{ij} + \frac{1}{2}\nabla_{\mathbf{x}}\nabla_{\mathbf{x}}\mathbf{u}^0(\mathbf{x}) : (\mathbf{y}_{ij}\otimes\mathbf{y}_{ij})] \\ &= \mathbf{f}'_{ij}\cdot[\nabla_{\mathbf{x}}\mathbf{u}^1(\mathbf{x},\mathbf{y}_j) + (\frac{1}{2}\nabla_{\mathbf{x}}\nabla_{\mathbf{x}}\mathbf{u}^0(\mathbf{x})\cdot\mathbf{y}_{ij})]\cdot\mathbf{y}_{ij} \\ &= \frac{1}{2}\mathbf{f}'_{ij}\cdot[2\nabla_{\mathbf{x}}\mathbf{u}^1(\mathbf{x},\mathbf{y}_j) + (\nabla_{\mathbf{x}}\nabla_{\mathbf{x}}\mathbf{u}^0(\mathbf{x})\cdot\mathbf{y}_{ij})]\cdot\mathbf{y}_{ij} \\ &= \frac{1}{2}\mathbf{f}'_{ij}\cdot[\nabla_{\mathbf{x}}\mathbf{u}^1(\mathbf{x},\mathbf{y}_j) - \nabla_{\mathbf{x}}\mathbf{u}^1(\mathbf{x},\mathbf{y}_i) + (\nabla_{\mathbf{x}}\nabla_{\mathbf{x}}\mathbf{u}^0(\mathbf{x})\cdot\mathbf{y}_{ij})]\cdot\mathbf{y}_{ij} \\ &\quad + \frac{1}{2}\mathbf{f}'_{ij}\cdot[\nabla_{\mathbf{x}}\mathbf{u}^1(\mathbf{x},\mathbf{y}_j) + \nabla_{\mathbf{x}}\mathbf{u}^1(\mathbf{x},\mathbf{y}_i)]\cdot\mathbf{y}_{ij}\end{aligned} \tag{51}$$

In view of Eq. (50), Eq. (51) can be written as

$$\mathbf{f}'_{ij}\cdot\boldsymbol{\psi}_{ij} = \frac{1}{2}\nabla_{\mathbf{x}}\mathbf{f}_{ij}\cdot\mathbf{y}_{ij} + \frac{1}{2}\mathbf{f}'_{ij}\cdot[\nabla_{\mathbf{x}}(\mathbf{u}^1(\mathbf{x},\mathbf{y}_i) + \mathbf{u}^1(\mathbf{x},\mathbf{y}_j))]\cdot\mathbf{y}_{ij} \tag{52}$$

To proceed with the derivation, we recall the identity

$$\nabla_{\mathbf{x}}\cdot(\mathbf{v}\otimes\mathbf{w}) = \mathbf{v}\nabla_{\mathbf{x}}\cdot\mathbf{w} + \nabla_{\mathbf{x}}\mathbf{v}\cdot\mathbf{w} \tag{53}$$

where **v** and **w** are vectors, and for the special case of **w** being independent of **x** we have

$$\nabla_{\mathbf{x}} \cdot (\mathbf{v} \otimes \mathbf{w}) = \nabla_{\mathbf{x}} \mathbf{v} \cdot \mathbf{w} \tag{54}$$

Based on Eq. (54), Eq. (52) can be written as

$$\mathbf{f}'_{ij} \cdot \boldsymbol{\psi}_{ij} = \frac{1}{2} \nabla_{\mathbf{x}} \cdot [\mathbf{f}_{ij} \otimes \mathbf{y}_{ij}] + \frac{1}{2} \mathbf{f}'_{ij} \cdot \nabla_{\mathbf{x}} \cdot [(\mathbf{u}^1(\mathbf{x}, \mathbf{y}_i) + \mathbf{u}^1(\mathbf{x}, \mathbf{y}_j)) \otimes \mathbf{y}_{ij}] \tag{55}$$

Substituting Eq. (55) into Eq. (47) yields

$$\rho \ddot{\mathbf{u}}^0(\mathbf{x}, t) = \frac{1}{2\Theta} \sum_i \sum_{j(\neq i)} \{\nabla_{\mathbf{x}} \cdot [\mathbf{f}_{ij} \otimes \mathbf{x}_{ij}]\} +$$

$$\frac{1}{2\Theta} \sum_i \sum_{j(\neq i)} \{\mathbf{f}'_{ij} \cdot \nabla_{\mathbf{x}} \cdot \left[\left(\mathbf{u}^1(\mathbf{x}, \mathbf{y}_i) + \mathbf{u}^1(\mathbf{x}, \mathbf{y}_j) \right) \otimes \mathbf{x}_{ij} \right]\} \tag{56}$$

In the remainder of this section we show that the second term in Eq. (56) vanishes for a periodic atomistic medium. We start by recalling

$$\begin{aligned} \mathbf{x}_{ji} &= \mathbf{x}_i - \mathbf{x}_j = \mathbf{X}_i - \mathbf{X}_j + \mathbf{u}_i - \mathbf{u}_j = -\mathbf{x}_{ij} \\ &= \varepsilon \boldsymbol{\phi}_{ji}(\mathbf{x}, \mathbf{y}) + \varepsilon^2 \boldsymbol{\psi}_{ji}(\mathbf{x}, \mathbf{y}) + \cdots \end{aligned} \tag{57}$$

where

$$\boldsymbol{\phi}_{ji}(\mathbf{x}, \mathbf{y}) = -\boldsymbol{\phi}_{ij}(\mathbf{x}, \mathbf{y}) = \mathbf{Y}_{ji} + \mathbf{u}^1(\mathbf{x}, \mathbf{y}_i) - \mathbf{u}^1(\mathbf{x}, \mathbf{y}_j) + \nabla_{\mathbf{x}} \mathbf{u}^0(\mathbf{x}) \cdot \mathbf{y}_{ji} \tag{58}$$

$$\boldsymbol{\psi}_{ji}(\mathbf{x}, \mathbf{y}) = -\boldsymbol{\psi}_{ij}(\mathbf{x}, \mathbf{y}) = \nabla_{\mathbf{x}} \mathbf{u}^1(\mathbf{x}, \mathbf{y}_j) \cdot \mathbf{y}_{ji} - \frac{1}{2} \nabla_{\mathbf{x}} \nabla_{\mathbf{x}} \mathbf{u}^0(\mathbf{x}) : (\mathbf{y}_{ji} \otimes \mathbf{y}_{ji}) \tag{59}$$

The Newton's third law requires

$$\mathbf{f}_{ij} = -\mathbf{f}_{ji} \tag{60}$$

From Eqs. (58) and (60) we have the following relation

$$\mathbf{f}'_{ij} = \frac{\partial \mathbf{f}_{ij}}{\partial \boldsymbol{\phi}_{ij}} = -\frac{\partial \mathbf{f}_{ji}}{\partial \boldsymbol{\phi}_{ij}} = -\frac{\partial \mathbf{f}_{ji}}{\partial (-\boldsymbol{\phi}_{ji})} = \frac{\partial \mathbf{f}_{ji}}{\partial \boldsymbol{\phi}_{ji}} \equiv \mathbf{f}'_{ji} \tag{61}$$

Let the interacting neighbor atoms of atom i be denoted as $n_1, n_2, \cdots, n_p, \cdots, n_k$ where k is the number of the interacting atoms such that $\left|\mathbf{x}_{n_p} - \mathbf{x}_i\right| < r_c$, $p = 1, 2, \cdots, k$. The second term in Eq. (56) can be expanded as

$$\begin{aligned}
&\sum_i \sum_{j(\neq i)} \{\mathbf{f}'_{ij} \cdot \nabla_{\mathbf{x}} \cdot [(\mathbf{u}^1(\mathbf{x}, \mathbf{y}_i) + \mathbf{u}^1(\mathbf{x}, \mathbf{y}_j)) \otimes (\mathbf{x}_j - \mathbf{x}_i)]\} \\
&\quad = \sum_i \{\mathbf{f}'_{in_1} \cdot \nabla_{\mathbf{x}} \cdot [(\mathbf{u}^1(\mathbf{x}, \mathbf{y}_i) + \mathbf{u}^1(\mathbf{x}, \mathbf{y}_{n_1})) \otimes (\mathbf{x}_{n_1} - \mathbf{x}_i)] \\
&\quad + \mathbf{f}'_{in_2} \cdot \nabla_{\mathbf{x}} \cdot [(\mathbf{u}^1(\mathbf{x}, \mathbf{y}_i) + \mathbf{u}^1(\mathbf{x}, \mathbf{y}_{n_2})) \otimes (\mathbf{x}_{n_2} - \mathbf{x}_i)] + \\
&\cdots + \mathbf{f}'_{in_p} \cdot \nabla_{\mathbf{x}} \cdot [(\mathbf{u}^1(\mathbf{x}, \mathbf{y}_i) + \mathbf{u}^1(\mathbf{x}, \mathbf{y}_{n_p})) \otimes (\mathbf{x}_{n_p} - \mathbf{x}_i)] + \\
&\cdots + \mathbf{f}'_{in_k} \cdot \nabla_{\mathbf{x}} \cdot [(\mathbf{u}^1(\mathbf{x}, \mathbf{y}_i) + \mathbf{u}^1(\mathbf{x}, \mathbf{y}_{n_k})) \otimes (\mathbf{x}_{n_k} - \mathbf{x}_i)]\}
\end{aligned} \tag{62}$$

The summation in (62) is carried out over all atoms in the unit cell. First, we consider the case that both atom i and any of its interacting neighboring atoms n_p $(p = 1, 2, \cdots, k)$ are in the unit cell. The summation for each interacting atom pair given by

$$\begin{aligned}
&\mathbf{f}'_{in_p} \cdot \nabla_{\mathbf{x}} \cdot [(\mathbf{u}^1(\mathbf{x}, \mathbf{y}_i) + \mathbf{u}^1(\mathbf{x}, \mathbf{y}_{n_p})) \otimes (\mathbf{x}_{n_p} - \mathbf{x}_i)] + \\
&\mathbf{f}'_{n_p i} \cdot \nabla_{\mathbf{x}} \cdot [(\mathbf{u}^1(\mathbf{x}, \mathbf{y}_{n_p}) + \mathbf{u}^1(\mathbf{x}, \mathbf{y}_i)) \otimes (\mathbf{x}_i - \mathbf{x}_{n_p})] = 0 \quad (p = 1, 2, \cdots, k)
\end{aligned} \tag{63}$$

vanishes due to Eq. (61).

If any of the interacting neighboring atom n_p lies outside the unit cell, by periodicity, the displacement and force vector of atom n_p take the same value as the corresponding atom in the unit cell and thus summation (63) holds. More details are provided in the Appendix for a model problem of an atomistic chain with a second nearest neighbor interaction.

In view of Eqs.(62) and (63) we have

$$\sum_i \sum_{j(\neq i)} \{\mathbf{f}'_{ij} \cdot \boldsymbol{\nabla}_{\mathbf{x}} \cdot [(\mathbf{u}^1(\mathbf{x}, \mathbf{y}_i) + \mathbf{u}^1(\mathbf{x}, \mathbf{y}_j)) \otimes \mathbf{x}_{ij}]\} = \mathbf{0} \tag{64}$$

Substituting Eq. (64) into (56) yields

$$\rho \ddot{\mathbf{u}}^0(\mathbf{x}, t) = \boldsymbol{\nabla}_{\mathbf{x}} \cdot \left\{ \frac{1}{2\Theta} \sum_i \sum_{j(\neq i)} [\mathbf{f}_{ij} \otimes \mathbf{x}_{ij}] \right\} \tag{65}$$

where $\mathbf{x}_{ij}$ is evaluated based on Eq. (42)

$$\mathbf{x}_{ij} \cong \varepsilon\phi_{ij}(\mathbf{x},\mathbf{y}) = \mathbf{F}^0 \cdot \left(\mathbf{X}_{ij} + \hat{\mathbf{u}}^1(\mathbf{x},\mathbf{y}_j) - \hat{\mathbf{u}}^1(\mathbf{x},\mathbf{y}_i)\right) \tag{66}$$

The coarse scale governing equations of motion can be written as

$$\boxed{\begin{aligned} &\rho\ddot{\mathbf{u}}^0(\mathbf{x},t) - \nabla_{\mathbf{x}} \cdot \boldsymbol{\sigma} = \mathbf{0} \\ &\boldsymbol{\sigma} = \frac{1}{2\Theta}\sum_i \sum_{j(\neq i)} \mathbf{f}_{ij} \otimes \mathbf{x}_{ij} \end{aligned}} \tag{67}$$

where $\boldsymbol{\sigma}$ in Eq. (67) coincides with the Cauchy stress. By comparing Eq. (67)*b* and Eq. (4) it can be seen that Cauchy stress is identical to the mechanical term in the virial stress formula. Note that symmetry of Cauchy stress follows from the balance of the angular momentum and therefore the order of the tensor product between $\mathbf{f}_{ij}$ and $\mathbf{x}_{ij}$ is interchangeable. We will refer to (67)*b* as the modified virial formula.

4 The two-scale information-passing algorithm and verification

The two-scale problem described by Eqs. (43) and (67) can be solved as follows:

(i) Solve the coarse scale problem (Eq. (67)*a* with appropriate initial and boundary conditions) using finite element semidiscretization in space and explicit time integration and calculate the coarse scale deformation gradient $\mathbf{F}^0(\mathbf{x},t)$

(ii) For every Gauss point in the coarse scale, solve the unit cell problem for $\hat{\mathbf{u}}^1(\mathbf{x},\mathbf{y}_i)$ using Eq. (43) and calculate the Cauchy stress by Eq. (67)*b*. Go to (i).

4.1 Formulation of the model problem

In this Section, we detail the formulation, implementation and verification of the generalized mathematical homogenization approach for the model problem of the atomic chain illustrated in Figure 1.

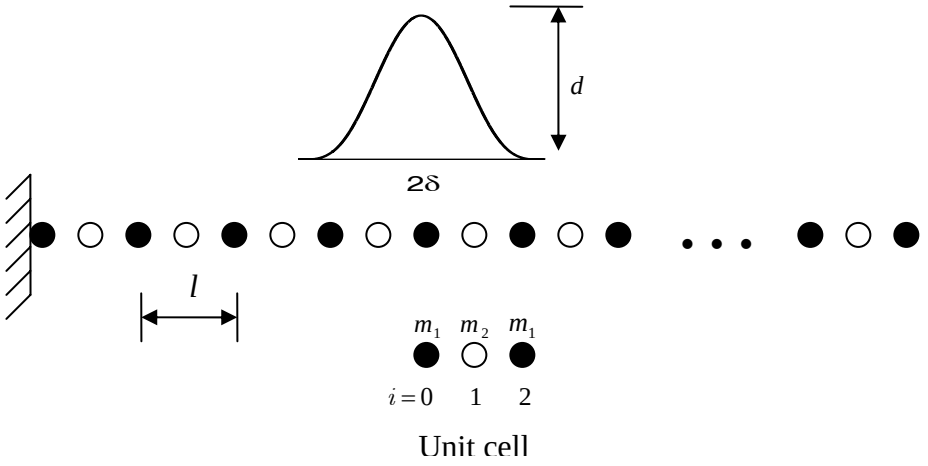

Figure 1: An atomic chain with the three-atom unit cell

We suppose that the atoms are initially equally spaced with spacing a and every atom interacts only with its nearest neighbors. The interatomic potentials are assumed to take the form of the Lennard-Jones potential. The interatomic potential between the first and the second atom in the unit cell is Φ_1 and that between the second and the third atom is Φ_2 given by

$$\Phi_1(r) = 4\varepsilon_1[(\frac{\sigma}{r})^{12} - (\frac{\sigma}{r})^6], \qquad \Phi_2(r) = 4\varepsilon_2[(\frac{\sigma}{r})^{12} - (\frac{\sigma}{r})^6] \tag{68}$$

where ε_1 and ε_2 are characteristic energy scales of the interaction and σ the characteristic length scale of the interaction. We assume that the initial configuration of the atomic chain is in equilibrium without external forces so that $a = 2^{1/6}\sigma$.

The interatomic forces are evaluated as

$$f_{01} = \frac{d\Phi_1}{dr} = \frac{24\varepsilon_1}{\sigma}[(\frac{\sigma}{r})^7 - 2(\frac{\sigma}{r})^{13}], \qquad f_{12} = \frac{d\Phi_1}{dr} = \frac{24\varepsilon_2}{\sigma}[(\frac{\sigma}{r})^7 - 2(\frac{\sigma}{r})^{13}] \tag{69}$$

The atomistic unit cell problem (43) reduces to

$$f_{i,i+1}(F^0(a + \hat{u}^1(x, y_{i+1}) - \hat{u}^1(x, y_i))) -$$

$$f_{i-1,i}(F^0(a + \hat{u}^1(x, y_i) - \hat{u}^1(x, y_{i-1}))) = 0 \qquad \forall i = 0,1,2 \tag{70}$$

Eq. (70) provides three equations for $i = 0,1$ and 2 . Due to periodicity

$$f_{-1,0} = f_{12}, \quad f_{23} = f_{01}, \quad \hat{u}^1(x, y_{-1}) = \hat{u}^1(x, y_1) = \hat{u}^1(x, y_3), \quad \hat{u}^1(x, y_2) = \hat{u}^1(x, y_0) \tag{71}$$

only one of the equations is independent, given by

$$f_{01}(F^0(a + \hat{u}^1(x, y_1) - \hat{u}^1(x, y_0))) -$$

$$f_{12}(F^0(a + \hat{u}^1(x, y_0) - \hat{u}^1(x, y_1))) = 0 \tag{72}$$

There are two unknowns in Eq. (72). To solve for Eq. (72), we specify periodic boundary conditions

$$\hat{u}^1(x, y_0) = \hat{u}^1(x, y_2) = 0 \tag{73}$$

Inserting Eq. (73) into (72) yields the nonlinear unit cell problem

$$f_{01}(F^0(a + \hat{u}^1(x, y_1))) - f_{12}(F^0(a - \hat{u}^1(x, y_1))) = 0 \tag{74}$$

The constitutive law-free coarse scale equation of motion (67) becomes

$$\rho \ddot{u}^0(x, t) - \sigma_x = 0 \tag{75}$$

with the initial and boundary conditions:

$$u^0(x, 0) = p(x), \qquad \dot{u}^0(x, 0) = q(x) \tag{76}$$

$$u^0(0, t) = 0, \qquad \sigma(L, t) = 0 \tag{77}$$

where L is the length of the atomic chain. Assuming a unit cross-sectional area for the atomistic chain the mass density is given by

$$\rho = \frac{1}{l} \sum_{i=1}^{2} m_i(y) = (m_1 + m_2) / l \tag{78}$$

and the Cauchy stress is

$$\sigma = \frac{1}{2l} \sum_{i=1}^{2} \sum_{j(\neq i)} f_{ij} x_{ij} = \frac{1}{2l} \sum_{i=1}^{2} [f_{i,i+1} x_{i,i+1} + f_{i,i-1} x_{i,i-1}] = \frac{1}{2l} \sum_{i=1}^{2} [f_{i,i+1} x_{i,i+1} + f_{i-1,i} x_{i-1,i}] \tag{79}$$

with

$$x_{i,i+1} = F^0[a + \hat{u}^1(x, y_{i+1}) - \hat{u}^1(x, y_i)]$$

$$x_{i-1,i} = F^0[a + \hat{u}^1(x, y_i) - \hat{u}^1(x, y_{i-1})] \tag{80}$$

In view of the periodicity conditions

$$f_{01} = f_{23}\,, \qquad r_{01} = r_{23} \tag{81}$$

the Cauchy stress reduces to

$$\sigma = f_{01}\,\frac{x_{01}}{l} + f_{12}\,\frac{x_{12}}{l} \tag{82}$$

4.2 The two-scale algorithm

The coarse scale equation (75) depends on the fine scale variable $\hat{u}^1$, which is determined by the nonlinear unit cell problem (74) subjected to the periodic boundary conditions (73). The nonlinear unit cell problem in turn depends on the coarse scale deformation gradient F^0, which is obtained from the constitutive law-free coarse scale equation of motion. The coarse scale equation of motion (75) and the nonlinear unit cell problem (74) are coupled and have to be solved concurrently.

The two-scale algorithm summarized in this Section is based on the finite element semidiscretization in space and explicit time integration for advancing the coarse scale problem and the Newton's method for solving a sequence of nonlinear molecular statics problems. Alternatively, fixed-point iteration methods discussed in [17] can be employed to improve robustness but at the expense of super-linear rate of convergence.

Finite element semidiscretization of the weak form of the coarse scale equation yields the semi-discrete equations of motion

$$\mathbf{M}\ddot{\mathbf{d}}(t) + \mathbf{f}^{in}(\mathbf{d}(t)) = \mathbf{0} \tag{83}$$

with initial conditions

$$\mathbf{d}(0) = \mathbf{d}_0\,, \qquad \dot{\mathbf{d}}(0) = \dot{\mathbf{d}}_0 \tag{84}$$

where $\mathbf{d}(t)$ is the vector of nodal displacements; $\mathbf{M}$ and $\mathbf{f}^{in}(\mathbf{d}(t))$ are the usual definitions of mass matrix and internal force vector. Eq. (83) is integrated using explicit time integration. For details, we refer to [20].

The nonlinear unit cell problem (74)

$$g(z) = f_{01}(F^0(a+z)) - f_{12}(F^0(a-z)) = 0 \tag{85}$$

is solved using Newton-like method with prescribed F^0 and $z = \hat{u}^1(x, y_1)$.

At each time step, the nonlinear unit cell problem (85) is first solved for $\hat{u}^1(x, y_1)$, then the Cauchy stress σ and the internal force vector $\mathbf{f}^{in}(\mathbf{d}_n)$ are evaluated.

4.3 Verification

We consider an atomistic chain consisting of 601 atoms schematically depicted in Figure 1. The fixed-free boundary conditions are assumed. The chain is subjected to a bell-shaped initial displacement pulse with width 2δ centered at the midpoint of the atomic chain. The atomistic chain possesses a periodic structure with the unit cell of length *l* composed of three atoms with masses m_1 and m_2. The inter-atomic potentials are Lennard-Jones potential given in Eq. (68). Material parameters are: $m_2 / m_1 = 2$ and $\varepsilon_2 / \varepsilon_1 = 3$. The ratio between the pulse width and the size of the unit cell is set to $\eta = 2\delta / l = 80$ in order to minimize the effect of dispersion, which cannot be captured by the O(1) theory developed in the manuscript. We refer to [21] for a methodology aimed at resolving dispersion effects.

The time step for the integration of the coarse-scale equation of motion is chosen approximately as the time for the wave to transverse a unit cell:

$$\Delta t_c = l / c\,, \qquad c = \sqrt{E / \rho} \tag{86}$$

where E is the homogenized property evaluated based on the quadratic approximation of the interatomic potentials

$$E = \frac{k_1 k_2 l}{k_1 + k_2}\,, \qquad k_1 = \frac{36 \times 4^{1/3} \varepsilon_1}{\sigma^2}, \qquad k_2 = \frac{36 \times 4^{1/3} \varepsilon_2}{\sigma^2} \tag{87}$$

The choice of the time step Δt_m for the molecular dynamics simulation is dictated by the natural fine scale time scale [22] evaluated as

$$\tau = \sigma\sqrt{m/\varepsilon} \tag{88}$$

where ε, σ and m are the characteristic energy and length scales of the interatomic potential and the characteristic mass of the atom, respectively. Choosing Δt_m of the 1-2% percent larger than τ generally leads to satisfactory energy conservation [22].

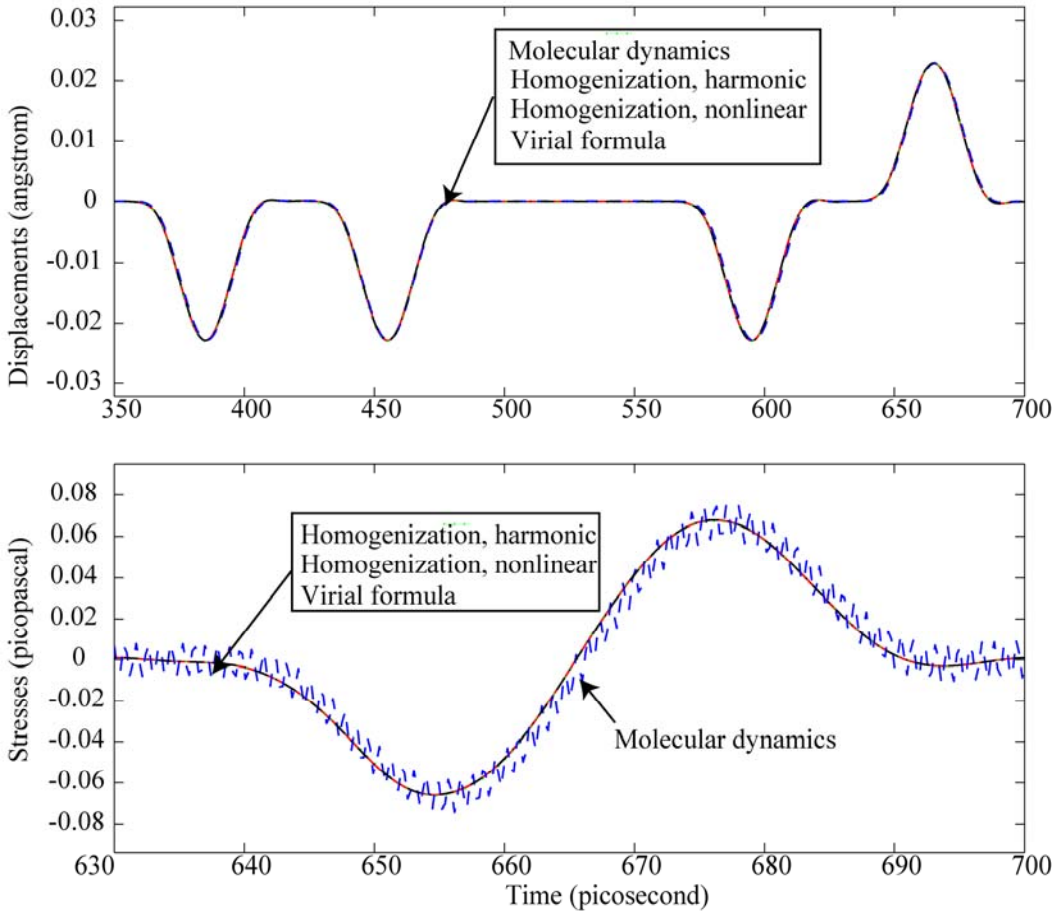

Figure 2: Displacements and stresses $(d/l = 0.006)$

In our numerical investigation, the ratio between the time steps of the coarse-scale equation of motion and the molecular dynamics simulation is $r = \Delta t_c / \Delta t_m = 3$. In general, however, when a unit cell contains thousands of atoms, this ratio would be substantially higher.

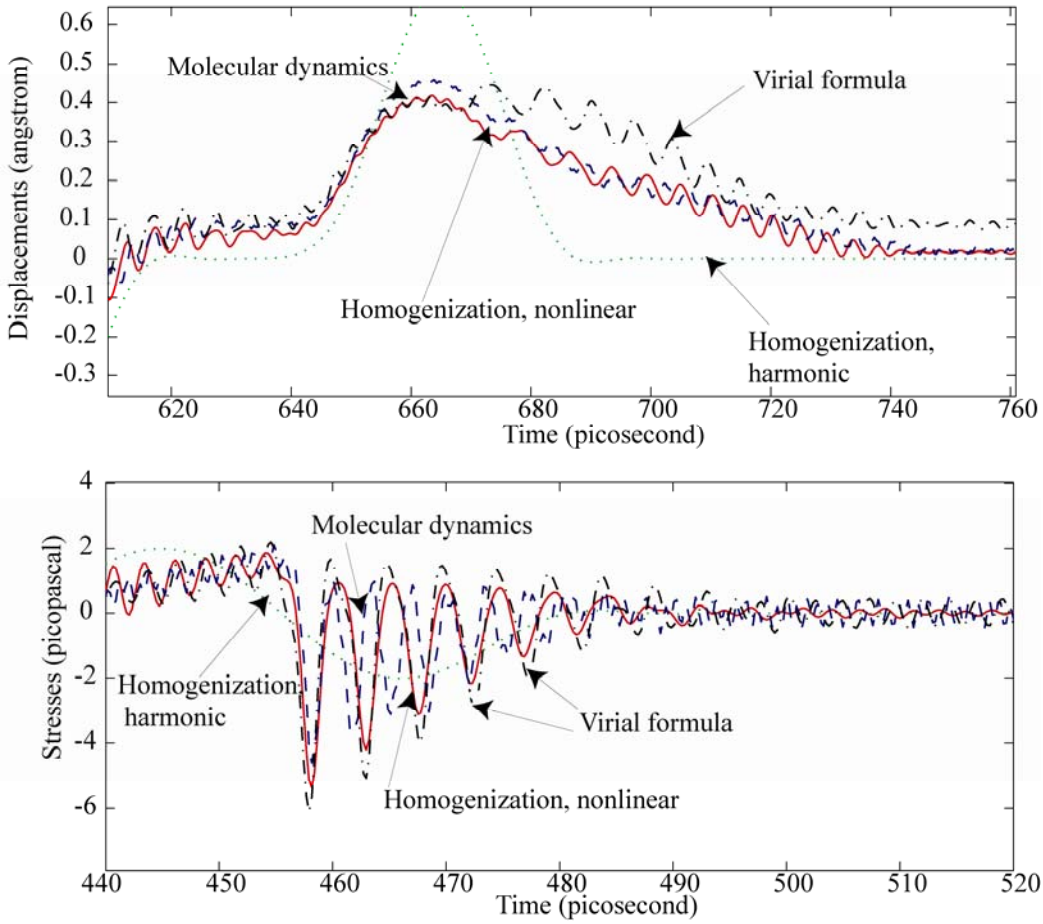

Figure 3: Displacements and stresses $(d/l = 0.18)$

The displacements at atom 401, the Cauchy and virial stresses in the unit cell containing atom 401 are plotted in Figures 2 and Figure 3. In Figure 2, the amplitude of the pulse is relatively small compared with the size of the unit cell $(d/l = 0.006)$ giving rise to linear wave propagation, whereas in Figure 3 the amplitude of the pulse is comparable to the size of the unit cell $(d/l = 0.18)$ resulting in nonlinear wave propagation. We compare four methods: the proposed nonlinear GMH, homogenization technique employing harmonic approximation of the interatomic potentials, the homogenization technique with the coarse-scale stress evaluated according to the virial formula Eq. (3) and the reference solution based on molecular dynamics.

It can be observed from Figure 2 and Figure 3 that for relatively small excitations, the harmonic approximation provides a satisfactory solution, while in the nonlinear regime,

the responses predicted by the harmonic approximation is inadequate. On the other hand, the response predicted by the nonlinear GMH compares favorably with the molecular dynamics simulation. The response predicted by the virial formula (Eq.(3)) shows a considerable deviation from the molecular dynamics solution.

For the model problem considered, the deviation from the average velocity in the unit cell has been found to be small compared to the average velocity. Thus, the version of the virial formula given in Eq. (4) provides comparable results to the GMH approach developed in this manuscript.

5 Appendix

Consider the 1D atomistic unit cell depicted in Figure 4 consisting of $n+1$ atoms. The linear mass density is

$$\rho = (m_1 + m_2 + m_3 + \cdots + m_n)/l \tag{89}$$

where by periodicity $m_0 = m_n$.

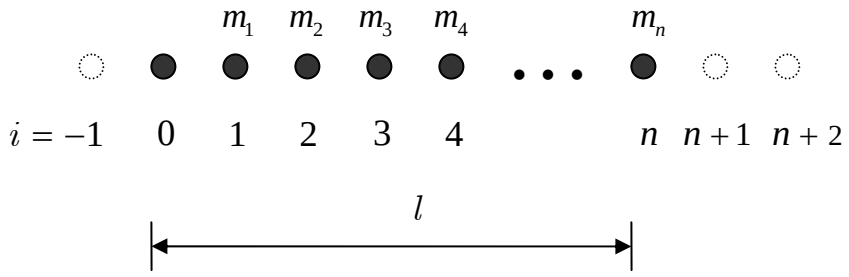

Figure 4: Atomistic unit cell

Assuming the second nearest neighbor interaction, the summation in Eq. (62) is expanded as

$$\sum_i \sum_{j(\neq i)} \{\mathbf{f}'_{ij} \cdot \nabla_{\mathbf{x}} \cdot [(\mathbf{u}^1(\mathbf{x}, \mathbf{y}_i) + \mathbf{u}^1(\mathbf{x}, \mathbf{y}_j)) \otimes (\mathbf{x}_j - \mathbf{x}_i)]\} = \sum_i \sum_{j(\neq i)} \{ f'_{ij}[u^1_x(x, y_i) + u^1_x(x, y_j)](x_j - x_i)\}$$

$$= \sum_{i=1}^{n} \{ f'_{i(i-2)}[u^1_x(x, y_i) + u^1_x(x, y_{i-2})](x_{i-2} - x_i) + f'_{i(i-1)}[u^1_x(x, y_i) + u^1_x(x, y_{i-1})](x_{i-1} - x_i) +$$

$$f'_{i(i+1)}[u^1_x(x, y_i) + u^1_x(x, y_{i+1})](x_{i+1} - x_i) + f'_{i(i+2)}[u^1_x(x, y_i) + u^1_x(x, y_{i+2})](x_{i+2} - x_i)\}$$

$$
\begin{aligned}
&= f'_{1(-1)}[u^1_x(x,y_1)+u^1_x(x,y_{-1})](x_{-1}-x_1)+\\
&f'_{10}[u^1_x(x,y_1)+u^1_x(x,y_0)](x_0-x_1)+ f'_{12}[u^1_x(x,y_1)+u^1_x(x,y_2)](x_2-x_1)+\\
&f'_{13}[u^1_x(x,y_1)+u^1_x(x,y_3)](x_3-x_1)+ f'_{20}[u^1_x(x,y_2)+u^1_x(x,y_0)](x_0-x_2)+\\
&f'_{21}[u^1_x(x,y_2)+u^1_x(x,y_1)](x_1-x_2)+ f'_{23}[u^1_x(x,y_2)+u^1_x(x,y_3)](x_3-x_2)+\\
&f'_{24}[u^1_x(x,y_2)+u^1_x(x,y_4)](x_4-x_2)+ f'_{31}[u^1_x(x,y_3)+u^1_x(x,y_1)](x_1-x_3)+\\
&f'_{32}[u^1_x(x,y_3)+u^1_x(x,y_2)](x_2-x_3)+ f'_{34}[u^1_x(x,y_3)+u^1_x(x,y_4)](x_4-x_3)+\\
&f'_{35}[u^1_x(x,y_3)+u^1_x(x,y_5)](x_5-x_3)+ f'_{42}[u^1_x(x,y_4)+u^1_x(x,y_2)](x_2-x_4)+\\
&f'_{43}[u^1_x(x,y_4)+u^1_x(x,y_3)](x_3-x_4)+ f'_{45}[u^1_x(x,y_4)+u^1_x(x,y_5)](x_5-x_4)+\\
&f'_{46}[u^1_x(x,y_4)+u^1_x(x,y_6)](x_6-x_4)+\cdots+\\
&f'_{(n-1)(n-3)}[u^1_x(x,y_{n-1})+u^1_x(x,y_{n-3})](x_{n-3}-x_{n-1})+ f'_{(n-1)(n-2)}[u^1_x(x,y_{n-1})+u^1_x(x,y_{n-2})](x_{n-2}-x_{n-1})+\\
&f'_{(n-1)n}[u^1_x(x,y_{n-1})+u^1_x(x,y_n)](x_n-x_{n-1})+ f'_{(n-1)(n+1)}[u^1_x(x,y_{n-1})+u^1_x(x,y_{n+1})](x_{n+1}-x_{n-1})+\\
&f'_{n(n-2)}[u^1_x(x,y_n)+u^1_x(x,y_{n-2})](x_{n-2}-x_n)+ f'_{n(n-1)}[u^1_x(x,y_n)+u^1_x(x,y_{n-1})](x_{n-1}-x_n)+\\
&f'_{n(n+1)}[u^1_x(x,y_n)+u^1_x(x,y_{n+1})](x_{n+1}-x_n)+ f'_{n(n+2)}[u^1_x(x,y_n)+u^1_x(x,y_{n+2})](x_{n+2}-x_n)
\end{aligned}
$$

$$
\begin{aligned}
&= f'_{1(-1)}[u^1_x(x,y_1)+u^1_x(x,y_{-1})](x_{-1}-x_1)+\\
&f'_{10}[u^1_x(x,y_1)+u^1_x(x,y_0)](x_0-x_1)+ f'_{20}[u^1_x(x,y_2)+u^1_x(x,y_0)](x_0-x_2)+\\
&f'_{(n-1)(n+1)}[u^1_x(x,y_{n-1})+u^1_x(x,y_{n+1})](x_{n+1}-x_{n-1})+ f'_{n(n+1)}[u^1_x(x,y_n)+u^1_x(x,y_{n+1})](x_{n+1}-x_n)+\\
&f'_{n(n+2)}[u^1_x(x,y_n)+u^1_x(x,y_{n+2})](x_{n+2}-x_n)=0 \qquad (90)
\end{aligned}
$$

The equality (90) is obtained due to the periodicity

$$f'_{(n-1)(n+1)} = f'_{(-1)1}, \qquad f'_{n(n+1)} = f'_{01}, \qquad (91)$$

$$f'_{n(n+2)} = f'_{02}, \quad x_{n-1} = x_{-1}, \quad x_n = x_0, \quad x_{n+1} = x_1, \quad x_{n+2} = x_2. \qquad (92)$$

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