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Atomic-level Stress Calculation and Continuum-Molecular System Equivalence

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Abstract: An atomistic level stress tensor is defined with physical clarity, based on the SPH method. This stress tensor rigorously satisfies the conservation of linear momentum, and is appropriate for both homogeneous and inhomogeneous deformations. The formulation is easier to implement than other stress tensors that have been widely used in atomistic analysis, and is validated by numerical examples. The present formulation is very robust and accurate, and will play an important role in the multiscale simulation, and in molecular dynamics. An equivalent continuum is also defined for the molecular dynamics system, based on the developed definition of atomistic stress and in conjunction with the SPH technique. The process is simple and easy to implement, and the fields are with high-order continuity. This equivalent continuum maintains the physical attributes of the atomistic system. This development provides a systematic approach to the continuum analysis of the discrete atomic systems.

keyword: Atomistic analysis, stress, SPH, continuum.

1 Introduction

The macroscopic behavior of solids is widely studied from a microscopic level, using the viewpoints of atomistic mechanics [Askar (1985), Bardenhagen and Triantafyllidis (1994)]. To bridge the atomistic mechanics and the continuum mechanics, it is important to know the relationships between the microscopic quantities of atoms, and the macroscopic quantities of continua. Atomic-level stress calculation plays a very important role in comparisons of continuum predictions with atomistic simulations, and it allows the intensity and nature of internal interactions in the discrete particle systems to be measured. The atomistic stress can be employed to interpret the results of atomistic simulation in light of continuum mechanical calculations, which have been used in molecular dynamics simulations of solids in a variety of ways, such as characterization of defects, the determination of elastic constants, and the study of the local elastic properties of carbon nanotubes [Chandra, Namilae, and Shet (2004)]. There are different ways to calculate stress in atomistic simulations. Pioneering work has been done in this field by Born and Huang (1954) who used an elastic energy approach to evaluate the stress in lattices by means of the Cauchy-Born hypothesis for homogeneous deformation.

Another widely used stress measure at the atomic scale is the virial stress, which is based on a generalization of the virial theorem of Clausius (1870) for gas pressure. This quantity includes two parts, and can be expressed as:

$$\boldsymbol{\sigma}^{virial}(\mathbf{r}) = \frac{1}{\Omega}\sum_i \left[-m_i \dot{\mathbf{u}}_i \otimes \dot{\mathbf{u}}_i + \frac{1}{2}\sum_{j\neq i} \mathbf{r}_{ij} \otimes \mathbf{f}_{ij} \right] \tag{1}$$

Here i and j are the atomic indices. The summation is over all the atoms occupying the total volume Ω. m_i is the mass of atom i, $\mathbf{u}_i$ is the displacement vector of atom i relative to a reference position, $\dot{\mathbf{u}}_i = d\mathbf{u}_i/dt$ denotes the material time derivative of $\mathbf{u}_i$, $\mathbf{r}_{ij} = \mathbf{r}_j - \mathbf{r}_i$, and $\otimes$ represents the tensor product of two vectors. $\mathbf{f}_{ij}$ is the interatomic force applied on atom i by atom j,

$$\mathbf{f}_{ij} = \frac{\partial \phi(r_{ij})}{\partial r_{ij}} \frac{\mathbf{r}_{ij}}{r_{ij}} \tag{2}$$

where $r_{ij} = \left\| \mathbf{r}_{ij} \right\|$, $\phi(r_{ij})$ is the energy of the atomic ensemble. It is noted that this stress formulation is strictly valid only when a homogeneous stress state exists in the entire volume. The first term on the right-hand side of equation (1) is the kinetic-energy term, which accounts for mass transport across a fixed spatial surface. The second term comes from interatomic interactions. Based on the virial stress, some other formulations of stress in

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molecular dynamics such as BDT stress [Basinski, Duesbery, and Taylor (1971)], Lutsko stress [Lutsko (1988); Cormier, Rickman, and Delph (2001)], and mechanical stress [Cheung and Yip (1991)] are proposed. However, as pointed by Zhou (2003):

The virial, BDT, Lutsko and mechanical stresses, are not the Cauchy stresses or any other form of mechanical stresses. It must be clearly stated that the virial theorem for gas pressure is totally correct in the statistical sense. However, generalizing it to claim that mechanical stress also depends on mass transfer as well as internal interatomic force is unjustifiable and incorrect. The virial stress as defined in equation (1) has the geometric interpretation of being a measure for the momentum change in a fixed spatial region. This interpretation does not assign any physical significance to the virial stress as a possible measure of mechanical interaction. Stress is a measure of the effect of pure force on momentum change associated with a fixed amount of mass (not change in momentum contained in a spatial region). The "virial stress" is defined using a spatial cut which is fixed in space, and is related to the statistical average of the external forces between the system and a rigid non-deforming container. If the virial stress is treated as a measure of mechanical force, the balance of momentum would be violated.

However, if the kinetic-energy term in these expressions is thrown off, they reduce to Cauchy stress with a physical meaning. In this paper, we will not consider the kinetic-energy term in the formulations of BDT and Lutsko stresses. BDT stress is put forward by Basinski, Duesbery, and Taylor (1971), and is based on a volumetric partition of the homogeneous deformed bulk by extending the virial stress to one atomic volume. BDT stress is defined as:

$$\boldsymbol{\sigma}^{BDT}(\mathbf{r}) = \frac{1}{2\Omega^i} \sum_{j \neq i} \mathbf{r}_{ij} \otimes \mathbf{f}_{ij} \tag{3}$$

where Ω^i is a small volume around an atom i. Theoretically, the above definitions are valid only for a homogeneous system. In section 2, we will prove that the BDT stress in eq. (3) is equivalent to the Cauchy-Born hypothesis for homogeneous deformation. The total volume and the volume of a single atom are required in the calculation of virial and BDT stresses. The local stress proposed by Lutsko (1988) and extended by Cormier, Rickman, and Delph (2001) is based on the local stress tensor of statistical mechanics. The Lutsko stress can be expressed as

$$\boldsymbol{\sigma}^{Lutsko}(\mathbf{r}) = \frac{1}{2\Omega^{Avg}} \sum_i \sum_{j \neq i} \mathbf{r}_{ij} \otimes \mathbf{f}_{ij} l_{ij} \tag{4}$$

where Ω^{Avg} is the averaging volume, l_{ij} $(0 \leq l_{ij} \leq 1)$ denotes the fraction of the length of the $i-j$ bond lying inside the same averaging volume. For a homogeneously deformed system, $\boldsymbol{\sigma}^{Lutsko}$ approaches $\boldsymbol{\sigma}^{BDT}$ for large averaging volumes. Lutsko stress has been used to evaluate local elastic properties of grain boundaries in metals. Lutsko stress assumes that the stress state is homogenous in the averaging volume. The mechanical stress advanced by Cheung and Yip (1991) is calculated as the sum of the time rate of the change of the momentum flux and the forces divided by area across the particular surface of interest. The researchers interested in surface problems proposed the atomic stress based on force balance and the interplanar interaction, instead of the interatomic interaction [Machova (2001)].

Two years back, Atluri (2002) pointed out that in a multiscale modeling, the forces on particles are simply different: those on an atomic particle arise due to atomic interactions, while those on a continuum particle arise due to the divergence of the stress-state around the particle, as pointed out by Navier. Thus, in any continuum-molecular dynamics equivalence, the atomic forces in MD should be made equivalent to the divergence of the stress-field in a continuum. Thus, in this paper, a new atomistic stress formulation is proposed with physical clarity, *which is appropriate for both homogeneous and inhomogeneous deformations*. In this paper, the formulation of the atomistic stress is derived directly based on the physics. Since the stress is a continuum concept, at first, the discrete atomistic force-field is smoothed by using the Smoothed Particle Hydrodynamics technique [Lucy (1977)]. Then, *by analyzing the force-state of an infinitesimal parallelepiped at point* $\mathbf{r}$*, a relationship between the stress and the atomistic force can be developed*. This formulation is useful and convenient for computational applications, and satisfies the conservation of linear momentum. The results are compared with the BDT and Lutsko stress. Based on the new formulation of the atomistic stress, an equivalent continuum for molecular system is defined, which conserves the momentum and mass of the discrete atomic system. The smoothed parti-

cle hydrodynamics (SPH) technique is employed to make the discrete atomic system to be an equivalent continuum system.

Smoothed Particle Hydrodynamics (SPH) has been introduced by Lucy (1977) to study self-gravitating fluids. The idea of the method is to consider the fluid as an ensemble of (smooth) particles. Each particle has a kernel which represents its mass distribution, and carries information on the average values of dynamical and thermodynamical quantities, as well as on their gradients. After then, it has become a widely used tool in astrophysics [Monaghan (1992)]. In astrophysics, the system is discrete. By means of a localized kernel function, a local continuous field is generated to avoid singularity. SPH provides a systematic method to obtain these quantities based on a "smoothed" estimate over neighboring particles. The idea for estimating the quantities using a kernel is at the heart of the SPH technique. We can employ this idea to obtain any physical quantity of a particle using a kernel-weighted estimate over neighboring particles. A primary reason for the popularity of SPH, despite its shortcomings, is its overall simplicity and ease of use. Due to its distinct advantage, the SPH method was widely adopted as one of the efficient computational techniques to solve applied mechanics problems.

2 The derivation of the stress tensor

As we discussed in the previous section, there are lots of atomistic stress tensors in volume-averaged form. Here, we will give an atomistic tensor in a nonvolume-averaged form with physical clarity.

Consider a discrete atomic system, wherein the interatomic force on atom i is $\mathbf{f}_i$. As well known, the concept of stress is a continuum concept. Hence, to derive the atomistic stress, at first, we should make the discrete atomic system to be an equivalent continuum system. The idea of the SPH method is employed here to smooth the discrete atomistic force field. SPH is very popular in astrophysics, where the real physical system is discrete. In order to avoid singularity, a local continuous field is generated by introducing a localized kernel function, which can serve as a smoothing interpolation field. The physical meaning of the kernel function can be interpreted as the probability of a particle's position, as in a probabilistic method. The SPH is only a smoothing technique. In this paper, we will smooth the discrete atomic force field at first. In this case, the force per unit volume, i.e. the force density $\mathbf{g}(\mathbf{r})$, can be obtained by means of the SPH, as

$$\mathbf{g}(\mathbf{r}) = \sum_i \mathbf{f}_i w(\mathbf{r} - \mathbf{r}_i, h) \tag{5}$$

where $\mathbf{f}_i$ is the force on atom i, $w(\mathbf{x}, h)$ is the smooth kernel function, and the summation is over all the particles. Notice that we do not have to divide by volume, because the kernel is normalized to unite volume [$w(\mathbf{x}, h)$ has the units of inverse volume, as in eq. (6)], the division by volume is effectively incorporate into $w(\mathbf{x}, h)$. A common choice for a kernel is a Gaussian, namely

$$w(\mathbf{x}, h) = \frac{1}{\left(\sqrt{\pi}h\right)^d} \exp\left(-\frac{\mathbf{x}^2}{h^2}\right) \tag{6}$$

where d is the number of spatial dimensions in the problem, and h is the smoothing length. The kernel is normalized such that its integral is unity, i.e.

$$\int w(\mathbf{x}, h)\, d\mathbf{x} = 1 \tag{7}$$

where the integration is over all the space. In general, the kernel function has to be a compact-supported positive function, and its integral is unity. Moreover, as $h \to 0$, the kernel function should approach to $\delta(\mathbf{x})$. It is important to realize that although the summations are formally over all the particles, only a small number actually contribute, because $w(\mathbf{x}, h)$ can be chosen so that it falls off rapidly for appropriate h. Other commonly used compact-supported kernel functions include the cubic spline and the quartic spline [Atluri (2004), Atluri and Shen (2002)], in these cases, the smoothing length becomes the radius of the compact support.

Now, in the equivalent continuum system, we consider an infinitesimal parallelepiped at point $\mathbf{r}$ with surfaces parallel to the coordinate planes (as shown in Fig. 1). In the infinitesimal volume, the volume of the infinitesimal parallelepiped is $dv = dx_1 dx_2 dx_3$, and the Cauchy stress at point $\mathbf{r}$ is $\boldsymbol{\sigma}(\mathbf{r})$, then, the resultant forces at point $\mathbf{r}$ in x_i direction are: $\sigma_{ji,j} dv$. On the other hand, from eq. (5), we can get the force density at point $\mathbf{r}$, $\mathbf{g}(\mathbf{r})$, of the equivalent continuum system. Hence, the resultant forces at point $\mathbf{r}$ should be equal to $\mathbf{g}(\mathbf{r})dv$. Thus, we have the following

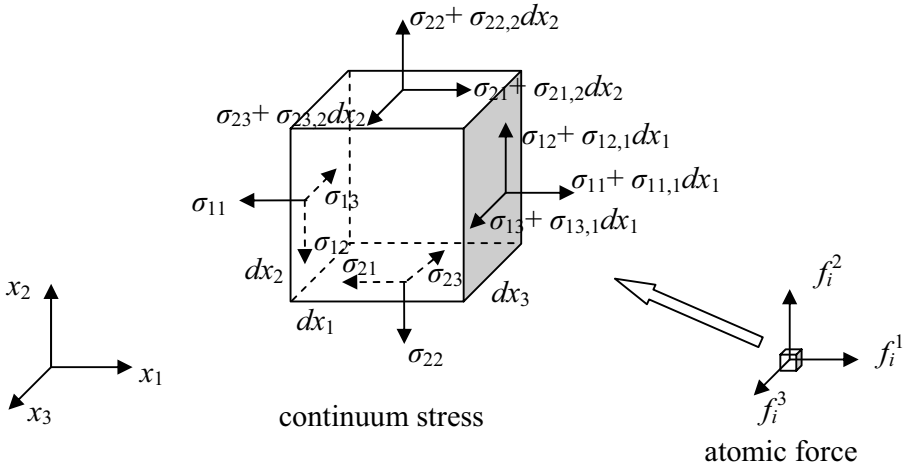

Figure 1 : The definition of the stress tensor

equation to relate the equivalent-continuum stress and the atomic force,

$$div\boldsymbol{\sigma}(\mathbf{r})\,dv = \mathbf{g}(\mathbf{r})\,dv \tag{8}$$

Then, we have

$$div\boldsymbol{\sigma}(\mathbf{r}) = \sum_i \mathbf{f}_i w(\mathbf{r}-\mathbf{r}_i, h) \tag{9}$$

Defining the Fourier transform of a function $F(\mathbf{r})$ as

$$\hat{F}(\mathbf{s}) = \int_V F(\mathbf{r})\, e^{\mathrm{i}\mathbf{s}\cdot\mathbf{r}} dV \tag{10}$$

and the inverse transform as

$$F(\mathbf{r}) = \frac{1}{(2\pi)^3}\int_{V^s} \hat{F}(\mathbf{s})\, e^{-\mathrm{i}\mathbf{s}\cdot\mathbf{r}} dV^s \tag{11}$$

where V^s is the transformed space. By using the Fourier transformation, equation (9) can be written as

$$\begin{aligned}
\mathrm{i}\mathbf{s}\cdot\hat{\boldsymbol{\sigma}}(\mathbf{s}) &= -\sum_i \mathbf{f}_i \hat{w}(\mathbf{s})\, e^{\mathrm{i}\mathbf{s}\cdot\mathbf{r}_i} \\
&= -\sum_i \sum_{j\neq i} \mathbf{f}_{ij} e^{\mathrm{i}\mathbf{s}\cdot\mathbf{r}_i} \hat{w}(\mathbf{s}) \\
&= -\frac{1}{2}\sum_i \sum_{j\neq i} \left[\mathbf{f}_{ij} e^{\mathrm{i}\mathbf{s}\cdot\mathbf{r}_i} + \mathbf{f}_{ji} e^{\mathrm{i}\mathbf{s}\cdot\mathbf{r}_j}\right] \hat{w}(\mathbf{s}) \\
&= \mathrm{i}\mathbf{s}\cdot\frac{1}{2}\sum_i \sum_{j\neq i} \mathbf{r}_{ij}\otimes\mathbf{f}_{ij}\frac{e^{\mathrm{i}\mathbf{s}\cdot\mathbf{r}_i} - e^{\mathrm{i}\mathbf{s}\cdot\mathbf{r}_j}}{\mathrm{i}\mathbf{s}\cdot\mathbf{r}_{ji}}\hat{w}(\mathbf{s})
\end{aligned} \tag{12}$$

Here, we use the following equation to deriving eq. (12),

$$\begin{aligned}
&\int_V [\nabla\cdot\boldsymbol{\sigma}(\mathbf{r})] e^{\mathrm{i}\mathbf{s}\cdot\mathbf{r}} dV \\
&= \int_V \left[\nabla\cdot\left(e^{\mathrm{i}\mathbf{s}\cdot\mathbf{r}}\boldsymbol{\sigma}\right) - \nabla\left(e^{\mathrm{i}\mathbf{s}\cdot\mathbf{r}}\right)\cdot\boldsymbol{\sigma}\right] dV \\
&= \int_V \left[\nabla\cdot\left(e^{\mathrm{i}\mathbf{s}\cdot\mathbf{r}}\boldsymbol{\sigma}\right) - \mathrm{i}\mathbf{s}\cdot\left(e^{\mathrm{i}\mathbf{s}\cdot\mathbf{r}}\boldsymbol{\sigma}\right)\right] dV \\
&= \int_\Gamma \mathbf{n}\cdot\left(e^{\mathrm{i}\mathbf{s}\cdot\mathbf{r}}\boldsymbol{\sigma}\right) d\Gamma - \mathrm{i}\mathbf{s}\cdot\int_V e^{\mathrm{i}\mathbf{s}\cdot\mathbf{r}}\boldsymbol{\sigma}(\mathbf{r}) dV \\
&= -\mathrm{i}\mathbf{s}\cdot\hat{\boldsymbol{\sigma}}(\mathbf{s})
\end{aligned}$$

Noting that

$$\frac{e^{\mathbf{i}\mathbf{s}\cdot\mathbf{r}_i}-e^{\mathbf{i}\mathbf{s}\cdot\mathbf{r}_j}}{\mathbf{i}\mathbf{s}\cdot\mathbf{r}_{ji}}=e^{\mathbf{i}\mathbf{s}\cdot\mathbf{r}_j}\int_0^1 e^{(\mathbf{i}\mathbf{s}\cdot\mathbf{r}_{ji})c}dc \tag{13}$$

By means of eq. (13) and carrying out the inverse transform of eq. (12), the Cauchy stress of the atomic level can be obtained as

$$\begin{aligned}\boldsymbol{\sigma}(\mathbf{r}) &= \frac{1}{2}\sum_i\sum_{j\neq i}\mathbf{r}_{ij}\otimes\mathbf{f}_{ij}\\ &\left\{\frac{1}{(2\pi)^3}\int_{V^s}e^{\mathbf{i}\mathbf{s}\cdot\mathbf{r}_j}\int_0^1 e^{(\mathbf{i}\mathbf{s}\cdot\mathbf{r}_{ji})c}dc\hat{w}(\mathbf{s})e^{-\mathbf{i}\mathbf{s}\cdot\mathbf{r}}dV^s\right\}\\ &= \frac{1}{2}\sum_i\sum_{j\neq i}\mathbf{r}_{ij}\otimes\mathbf{f}_{ij}\\ &\left\{\int_0^1\left[\frac{1}{(2\pi)^3}\int_{V^s}\hat{w}(\mathbf{s})e^{\mathbf{i}\mathbf{s}\cdot(\mathbf{r}_{ji}c+\mathbf{r}_j-\mathbf{r})}dV^s\right]dc\right\}\\ &= \frac{1}{2}\sum_i\sum_{j\neq i}\mathbf{r}_{ij}\otimes\mathbf{f}_{ij}\int_0^1 w[\mathbf{r}-(\mathbf{r}_{ji}c+\mathbf{r}_j)]dc\end{aligned} \tag{14}$$

As $h\to 0$, the kernel function $w(\mathbf{x},h)$ should approach to $\delta(\mathbf{x})$, and eq. (14) will be reduced to

$$\boldsymbol{\sigma}(\mathbf{r})=\frac{1}{2}\sum_i\sum_{j\neq i}\mathbf{r}_{ij}\otimes\mathbf{f}_{ij}\delta[\mathbf{r}-(\mathbf{r}_{ji}k+\mathbf{r}_j)] \tag{15}$$

where $0\leq k\leq 1$. $\delta[\mathbf{r}-(\mathbf{r}_{ji}k+\mathbf{r}_j)]$ is singular along the line segment between $\mathbf{r}_i$ and $\mathbf{r}_j$, and is zero elsewhere. This singular expression can be used to obtain the average stress over any region of an atomistic ensemble, *i.e.* the Lutsko stress. For a region with volume Ω^{Avg}, by integrating eq. (15), and divided by Ω^{Avg}, eq. (4) is derived. Moreover, if one thinks that the singular atomic stress exists only at atomic positions, the physically significant interpretation of eq. (15) in the context of discrete atomic system is

$$\boldsymbol{\sigma}(\mathbf{r})=\frac{1}{2}\sum_i\sum_{j\neq i}\mathbf{r}_{ij}\otimes\mathbf{f}_{ij}\delta(\mathbf{r}-\mathbf{r}_i) \tag{16}$$

This singular expression can be used to obtain the average stress over any region of an atomistic ensemble. For a region with volume Ω^i around atom i at the current configuration, the average stress can be derived as the BDT stress eq. (3). Equations (15) and (16) are useless for computational applications, but the coarse-grained average stress can be defined by integrating equations (15) and (16) over an appropriate averaging volume; while the stress formulation (14) is useful for computational applications. The virial, BDT and Lutsko stresses smear the effect of inhomogeneities due to volume averaging. The calculation of the Cauchy stress in eq. (14) does not involve *ad hoc* specification of a relevant volume, while the evaluation of the BDT stress (or the Cauchy-Born rule) for any set of atoms requires the identification of a proper volume whose extent is not always obvious: in atomic ensembles with irregular atom arrangement, the identification of this volume can be ambiguous.

For the finite deformation, we can also derive the first Piola-Kirchhoff stress $\mathbf{T}$, based on the initial configuration. We denote the initial position of the atom i as $\mathbf{R}_i$, then, in initial configuration, eq. (9) can be rewritten as

$$div\mathbf{T}(\mathbf{R})=\sum_i\mathbf{f}_i w(\mathbf{R}-\mathbf{R}_i,h) \tag{17}$$

Similarly, the first Piola-Kirchhoff stress tensor can be obtained as

$$\mathbf{T}(\mathbf{R})=\frac{1}{2}\sum_i\sum_{j\neq i}\mathbf{R}_{ij}\otimes\mathbf{f}_{ij}\int_0^1 w[\mathbf{R}-(\mathbf{R}_{ji}c+\mathbf{R}_j)]dc \tag{18}$$

As $h\to 0$, eq. (18) becomes

$$\mathbf{T}(\mathbf{R})=\frac{1}{2}\sum_i\sum_{j\neq i}\mathbf{R}_{ij}\otimes\mathbf{f}_{ij}\delta[\mathbf{R}-(\mathbf{R}_{ji}c+\mathbf{R}_j)] \tag{19}$$

with $0\leq c\leq 1$. For the discrete atomic system, the physically significant solution of eq. (19) is

$$\mathbf{T}(\mathbf{R})=\frac{1}{2}\sum_i\sum_{j\neq i}\mathbf{R}_{ij}\otimes\mathbf{f}_{ij}\delta(\mathbf{R}-\mathbf{R}_i) \tag{20}$$

This singular expression can be used to obtain the average stress over any region of an atomistic ensemble. For a region with volume Ω_0^i around atom i at initial configuration, the average stress is

$$\overline{\mathbf{T}}(\mathbf{R}) = \frac{1}{2\Omega_0^i} \sum_i \sum_{j \neq i} \mathbf{R}_{ij} \otimes \mathbf{f}_{ij} \tag{21}$$

Recognizing the relationship between stress, deformation and internal strain energy, Born and Huang (1954) used the Cauchy-Born hypothesis to evaluate the stress in lattices for homogeneous deformation. In that method, the gradient of the deformation $\mathbf{F}$ is defined as

$$\mathbf{r}_{ij} = \mathbf{F}\mathbf{R}_{ij} \tag{22}$$

The strain density is W/Ω_0^i, where W is the interatomic potential of the atomic volume Ω_0^i. Then, the stress can be derived as

$$\begin{aligned} \overline{\mathbf{T}}(\mathbf{R}) &= \frac{1}{\Omega_0^i} \frac{\partial W}{\partial \mathbf{F}} \\ &= \frac{1}{2\Omega_0^i} \sum_i \sum_{j \neq i} \frac{\partial W}{\partial \mathbf{r}_{ij}} \frac{\partial \mathbf{r}_{ij}}{\partial \mathbf{F}} \\ &= \frac{1}{2\Omega_0^i} \sum_i \sum_{j \neq i} \mathbf{R}_{ij} \otimes \mathbf{f}_{ij} \end{aligned} \tag{23}$$

Here, we employed $\partial \mathbf{r}_{ij}/\partial \mathbf{F} = \mathbf{R}_{ij}$ in derivation of eq. (23). From the finite deformation theory, we have

$$\boldsymbol{\sigma} = J\mathbf{F}\mathbf{T} \tag{24}$$

where J is the Jacobian. Hence, by means of eq. (24), from eq. (23), we can get the Cauchy stress

$$\begin{aligned} \overline{\boldsymbol{\sigma}}(\mathbf{r}) &= J\mathbf{F}\overline{\mathbf{T}}(\mathbf{R}) \\ &= \frac{J}{2\Omega_0^i} \sum_i \sum_{j \neq i} \mathbf{F}\mathbf{R}_{ij} \otimes \mathbf{f}_{ij} \\ &= \frac{1}{2\Omega^i} \sum_i \sum_{j \neq i} \mathbf{r}_{ij} \otimes \mathbf{f}_{ij} \end{aligned} \tag{25}$$

If Ω^i is taken to be the total volume of the system, this equation becomes the virial stress; if Ω^i is taken to be a small volume around an atom i, this equation becomes the BDT stress. Eq. (25) confirms that the BDT stress [or the second term of virial stress (1)] is identical to that based on the Cauchy-Born hypothesis, in homogeneous deformation. This also implies that the kinetic-energy term of eq. (1) should not be included in the expression of the atomic stress. However, there are a lot of limits in the Cauchy-Born hypothesis [Atluri (2004), Shen and Atluri (2004a, b)], while there are no limits on the formulation in (14).

It is important to point out that the derivation here is also appropriate for system with body forces, which can result from non-local effects of atoms or agents external to the system under consideration. The result would be the same, which will be shown in the next section.

3 Equivalent continuum for atomic system

It is important to know the relationships between the microscopic quantities of atoms, and the macroscopic quantities of continua, for nanoscale characterizations of material behavior. Molecular dynamics and continuum mechanics are on the opposite ends of the temporal and spatial scale spectrum, and consist of highly developed and reliable modeling methods. Continuum mechanics methods predict the macroscopic mechanical behavior of materials idealized as continuous media, based on known constitutive relationships of the bulk material, while molecular dynamic models predict molecular properties based on known quantum interactions. Each has its own advantages and limitations. Continuum analyses are appropriate only for a large enough system. Alternative to continuum analysis, the atomistic modeling and simulation calculates individual atoms explicitly, and follows them during their dynamic evolution. However, both continuum mechanics and molecular dynamics obey the same fundamental laws, including Newton's laws of motion and conservation of mass. These fundamental laws provide a bridge to link continuum mechanics and molecular dynamics. If a continuum is equivalent to a MD system, in addition that it contains the same amount of mass as the particle system, the Newton's laws of motion of the continuum system must be derived from the Newton's laws of motion of its corresponding MD system, vice versa, i.e. they are equivalent. The equivalent continuum development offers a high degree of fidelity to the discrete description.

An equivalent continuum is constructed by using the principle of the virtual work, and in conjunction with finite element interpolations in Zhou (2003), Zhou and

McDowell (2002); However, their formulations are very complicated. Their development is computationally intensive due to the construction of the finite elements. The determined fields are piecewise continuous, even for homogenous deformation. Moreover, the defined continuum deformation fields lack the consistency with the continuum differential requirement about the strain-displacement relations. In this section, an equivalent continuum is defined for molecular dynamics (MD) particle systems, based on the definition of atomistic stress (14) and in conjunction with the SPH technique. This process is simple and easy to implement, and the fields are with high-order continuity.

For the MD system, the Newton's laws of motion for each atom *i*, can be written as

$$\mathbf{f}_i = m_i \ddot{\mathbf{u}}_i \tag{26}$$

The force on atom *i* due to atoms or agents that are external to the system under consideration is denoted as $\mathbf{f}_i^b$, the total force on atom *i* is

$$\mathbf{f}_i = \sum_{j \neq i} \mathbf{f}_{ij} + \mathbf{f}_i^b = \mathbf{f}_i^s + \mathbf{f}_i^b \tag{27}$$

It is noted that $\mathbf{f}_i^b$ also includes the non-local interactions.

Let $\mathbf{b}$ denote the density of the continuum body force, and let ρ represent the density of the continuum mass. In this analysis, all the quantities are evaluated on the current configuration. Similar to the analysis in section 2, the resultant forces at point $\mathbf{r}$ in the equivalent continuum system are: $div\boldsymbol{\sigma} + \mathbf{b}$ then we have

$$\begin{aligned} div\boldsymbol{\sigma} + \mathbf{b} &= \sum_i \mathbf{f}_i w(\mathbf{r} - \mathbf{r}_i, h) \\ &= \sum_i \mathbf{f}_i^s w(\mathbf{r} - \mathbf{r}_i, h) + \sum_i \mathbf{f}_i^b w(\mathbf{r} - \mathbf{r}_i, h) \end{aligned} \tag{28}$$

Thus, the density of the continuum body force can be obtained as

$$\mathbf{b}(\mathbf{r}) = \sum_i \mathbf{f}_i^b w(\mathbf{r} - \mathbf{r}_i, h) \tag{29}$$

The associations of internal forces to the internal stress only, and the external force to the body force only, are strictly required by the balance of momentum. And the stress is still expressed as in eq. (14). The density of the continuum mass can be expressed as

$$\rho(\mathbf{r}) = \sum_i m_i w(\mathbf{r} - \mathbf{r}_i, h) \tag{30}$$

Integrating eq. (30), and by means of eq. (7), we can confirm the conservation of the mass, *i.e.* $\int \rho(\mathbf{r})\, d\mathbf{r} = \sum_i m_i$.

Substituting eq. (26) into eq. (28), the following equation can be derived

$$div\boldsymbol{\sigma}(\mathbf{r}) + \mathbf{b}(\mathbf{r}) = \sum_i m_i \ddot{\mathbf{u}}_i w(\mathbf{r} - \mathbf{r}_i, h) \tag{31}$$

On the other hand, the Newton's laws of motion for the equivalent continuum are

$$div\boldsymbol{\sigma}(\mathbf{r}) + \mathbf{b}(\mathbf{r}) = \rho(\mathbf{r})\, \ddot{\mathbf{u}}(\mathbf{r}) \tag{32}$$

Then, the acceleration field $\ddot{\mathbf{u}}$ of the equivalent continuum can be obtained as

$$\ddot{\mathbf{u}}(\mathbf{r}) = \frac{\sum_i m_i \ddot{\mathbf{u}}_i w(\mathbf{r} - \mathbf{r}_i, h)}{\rho(\mathbf{r})} = \frac{\sum_i m_i \ddot{\mathbf{u}}_i w(\mathbf{r} - \mathbf{r}_i, h)}{\sum_i m_i w(\mathbf{r} - \mathbf{r}_i, h)} \tag{33}$$

Thus, the equivalent continuum is constructed from the discrete MD system, which preserves the momentum, and conserves the mass. Moreover, from the Newton's laws of motion for the equivalent continuum, eq. (32), the Newton's laws of motion for each atom *i*, eq. (26), can also be derived.

A reinterpretation of the discrete atomistic force and deformation is reflected by the continuum field defined here, which maintains the physical effects of the atomistic system. This development provides a systematic approach to the continuum analysis of the discrete atomic system. It can also be applied to multiscale modeling of material behavior which combines both MD and continuum descriptions in the development of constitutive relations at different scales.

4 Numerical examples

At first, we apply the stress formulation (14) to the case of a homogeneously deformed cubic, crystalline solid. Similar to Cormier, Rickman, and Delph (2001), we considered a collection of 2048 atoms, initially on the sites of a face centered cubic (fcc) lattice and confined to a periodic cubic simulation cell at temperature T=0. Employed is a modified (truncated) Lennard-Jones potential [Broughton and Gilmer (1983)],

$$\phi(r) = \begin{cases} 4\varepsilon\left[\left(\frac{\sigma}{r}\right)^{12}-\left(\frac{\sigma}{r}\right)^{6}\right]+C_1,\ r\le 2.3\sigma \\ C_2\left(\frac{\sigma}{r}\right)^{12}+C_3\left(\frac{\sigma}{r}\right)^{6}+C_4\left(\frac{r}{\sigma}\right)^{6}+C_5,\ 2.3\sigma<r<2.5\sigma \\ 0,\ r\ge 2.5\sigma \end{cases} \tag{34}$$

with

$$C_1 = 0.016132\varepsilon,\ C_2 = 3.1366\times 10^3\varepsilon,\ C_3 = -68.069\varepsilon,$$
$$C_4 = -0.083312\varepsilon,\ C_5 = 0.74689\varepsilon$$

where ε and σ are the energy and length parameters, respectively (not to be confused with the stress and strain). The perfect fcc crystal has a lattice parameter of $a_0 = 1.550512\sigma$ and the corresponding energy per atom 7.45ε. The deformation can be imposed by simply changing the lattice parameter. The elastic constants can be deduced from the quadratic dependence of energy on strain, by using the Cauchy-Born hypothesis. We consider the simple case of uniform applied deformations, with corresponding strain tensor components $\varepsilon_{11} = \varepsilon_{22} = \varepsilon_{33} = 0.002$, the remaining components are 0. In this case, the BDT stress (or the stress from Cauchy-Born hypothesis) is equal to the bulk stress. In the calculations, the kernel function $w(\mathbf{x},h)$ is truncated, by a truncated radius r_w, beyond which it falls off rapidly to zero for appropriate h, as

$$w(\mathbf{x},h) = \begin{cases} \frac{1}{\left(\sqrt{\pi}h\right)^d}\exp\left(-\frac{\mathbf{x}^2}{h^2}\right),\ |\mathbf{x}|\le r_w \\ 0,\ |\mathbf{x}|>r_w \end{cases} \tag{35}$$

Thus, the kernel function becomes compact-supported. The studies of SPH show that $h = 0.4r_w$ or so give very good results. Our numerical tests also confirm this statement. Fig.2 shows the effects of the ratio h/r_w on the stress value, the stress values of the formulation (14) are normalized by the bulk stress, which is equal to the BDT stress (or the stress from Cauchy-Born hypothesis) here. It can be seen that a larger r_w is needed for $h = 0.2r_w$, and the error is a little bigger for $h = 0.5r_w$. Hence, in this paper, we take $h = 0.4r_w$ for all the calculations. For the purpose of comparison, we also compute the Lutsko stress. For Lutsko stress, the radius of the spherical averaging volume Ω^{Avg} is taken to be r_w.

Fig. 3 shows the magnitude of the stress σ_{11} at an observation point versus the radius r_w, where the stresses are normalized by the bulk stress, which is equal to the BDT stress. The results of the present formulation are almost overlapped with the bulk stress value, while the Lutsko stress value oscillates around the bulk stress value. The Lutsko stress value converges to the bulk stress value for sufficiently large radii. However, the larger radius is computational costly. Hence, in the following calculations, we choose the radius $r_w = 1.5a_0$, at which the Lutsko stress value agrees with the bulk stress value vary well in Fig. 3. Fig. 4 depicts the normalized stress σ_{11} at different observation points along the closed-packed [110] direction. This figure also shows the results of the present formulation agree with the bulk stress value very well, and the Lutsko stress value oscillates around the bulk stress value. From these figures, we can find that the present stress formulation (14) is very robust and accurate, while the accuracy of Lutsko stress depends on the radius of the spherical averaging volume and the position of the observation point.

Now, we apply the stress formulation (14) to the case of inhomogeneous deformation. A "big" atom replaces the atom at the origin of the previous fcc crystalline solid. The inhomogeneous elastic fields will arise due to the presence of the "big" atom. For the "big" atom, the energy and length parameters ε^b and σ^b are assumed to be $\varepsilon^b = 1.2\varepsilon$ and $\sigma^b = 1.2\sigma$, respectively. We employed the modified Newton-Raphson method to solve equations: $\mathbf{f}_i(\mathbf{r}) = \mathbf{0}$, to determine the equilibrium configuration (i.e. the minimum energy configuration) for this defected structure. This problem is similar to the inclusion problem in a continuum. As is well known, the continuum elastic field of an inclusion at the origin falls off with distance r from the origin as $1/r^2$, and the corresponding elastic stress field falls off with dis-

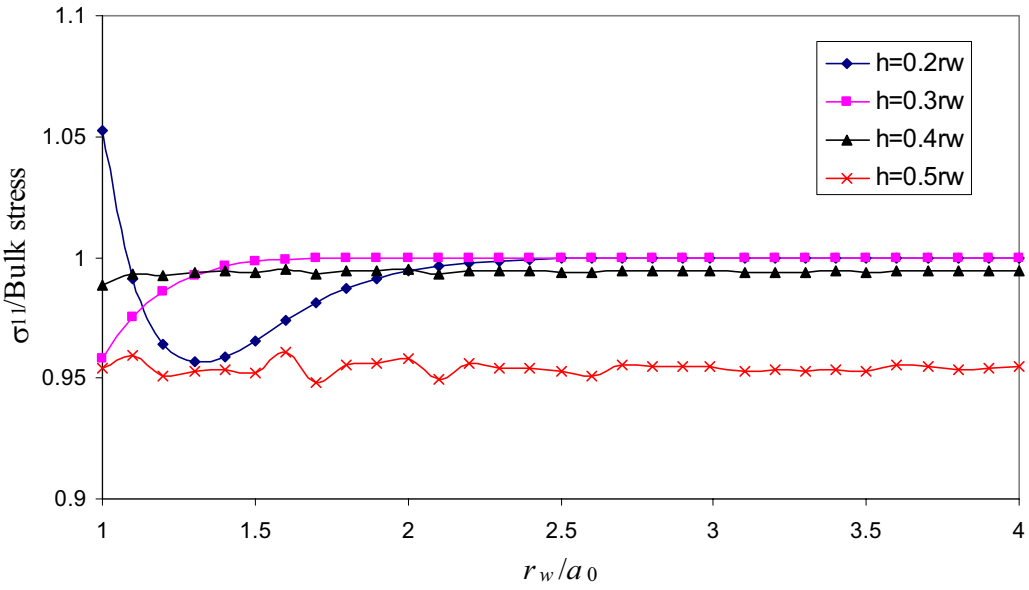

Figure 2 : The effect of the ratio h/r_w on stress value for uniform deformation

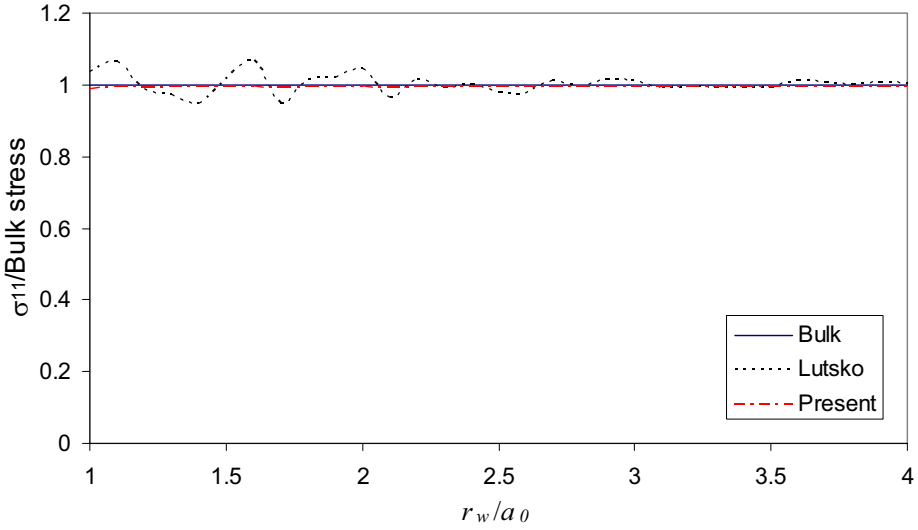

Figure 3 : The effect of the radius r_w on stress value for uniform deformation

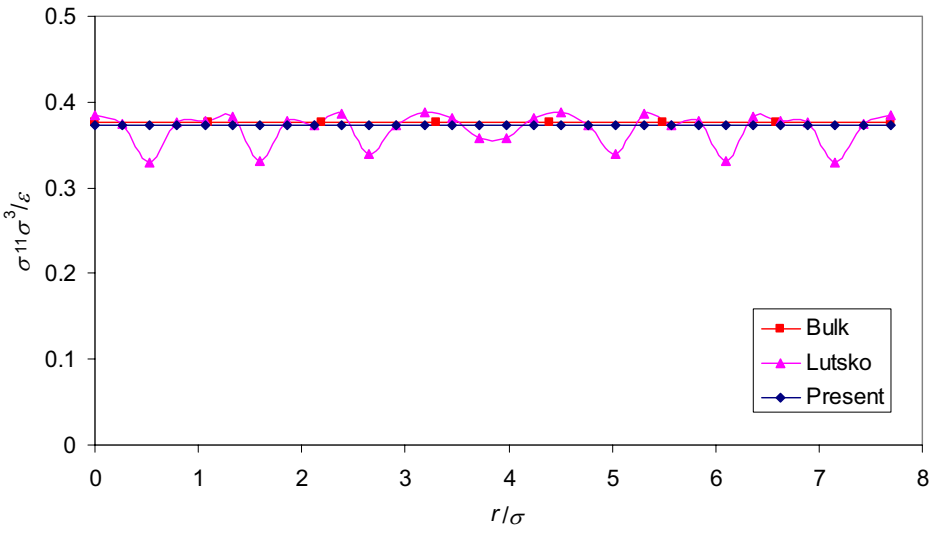

Figure 4 : The stress values at different position along [110] direction

tance r from the origin as $1/r^3$ [Li, Shen, Han and Atluri (2003)]. Fig. 5 depicts the radial component of the discrete atomic displacement field in the close-packed [110] direction versus $(\sigma/r)^2$. We can find that the atomic displacement demonstrates the expected $1/r^2$ behavior when $(\sigma/r)^2 < 0.0924$, *i.e.* $r/\sigma > 1.5\cdot\sqrt{2}a_0/\sigma = 3.288$ (i.e., beyond 1.5 times the lattice parameters from the origin). This is because the cut-off radius of the Lennard-Jones potential for the "big" atom is $r_{cut} = 2.5\sigma^b = 3\sigma$, and 3.288σ is the nearest atomic site to r_{cut}, the boundary of the inclusion should be around this value.

Figs. 6, 7 and 8, respectively, show the variation of the normalized stress components σ_{11}, σ_{12} and σ_{33} in the [110] direction versus $(\sigma/r)^3$. In these figures, the continuum solutions are plotted only for $r/\sigma > 3.288$ (*i.e.* $(\sigma/r)^3 < 0.0028$) due to the reason that we just discussed in the previous paragraph. The continuum solution is obtained by an approximate method. The displacement gradient is approximated form Fig. 5 at first, then using the elastic constants of the homogenous perfect fcc crystalline solid [Cormier, Rickman, and Delph (2001)] and the elastic constitutive relationship (Hook's Law), the continuum elastic stress can be determined. For the purpose of comparison, we also plot here the corresponding values of the Lutsko and BDT stresses. For the BDT stress, we only calculate the values at the atomic sites. Both of the Lutsko and BDT stress values are far away from the continuum solution. It can be seen that the stress values of the present formulation (14) agree with the continuum results very well beyond 2 times the lattice parameters from the origin (the "big atom): $(\sigma/r)^3 \le 0.0017$ (i.e., $r/\sigma > 2\cdot\sqrt{2}a_0/\sigma = 4.385$), which fall off with distance r from the origin as $1/r^3$. In range of 1.5-2 times the lattice parameters (i.e. $0.0017 < (\sigma/r)^3 < 0.0028$), the stress values of the present formulation (14) deviate a little from the $(\sigma/r)^3$ behavior. The deviations are due to the deviation of the elastic constants from those of the perfect fcc crystalline in this range [actually, in this range, the displacement shown in Fig. 5 already deviates from the $(\sigma/r)^2$ behavior]. In the range of 0-1.5 times the lattice parameters, comparison with continuum theory is impossible, since the elastic constants cannot be well defined. However, the trend of the stress value of the present formulation (14) in this range is consistent with that of the displacements shown in Fig. 5. The results of the Lutsko stress show bad shapes, perhaps a larger averaging volume is required to improve its performance, even for homogeneous deformations. The requirement of a very large averaging volume inherently decreases its use for computational applications. Since the BDT stress is strictly valid only for homogeneous deformations, it is not surprising that it should not perform

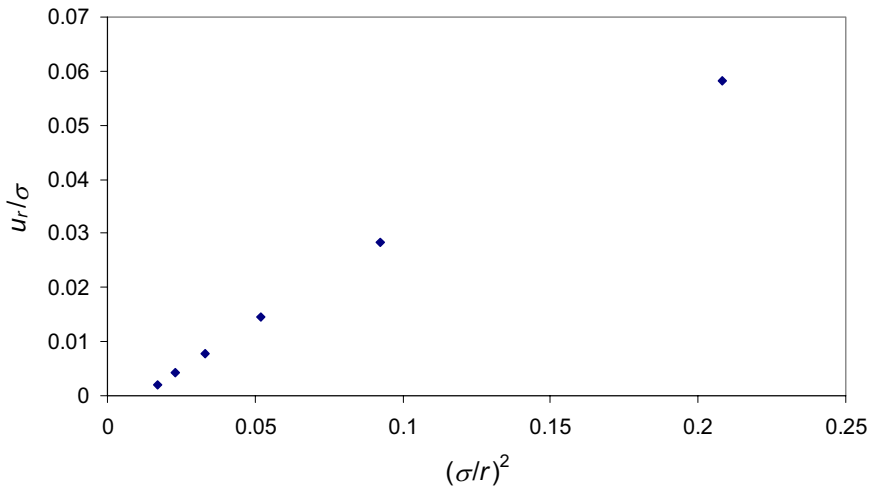

Figure 5 : Radial component of the displacement field in [110] direction

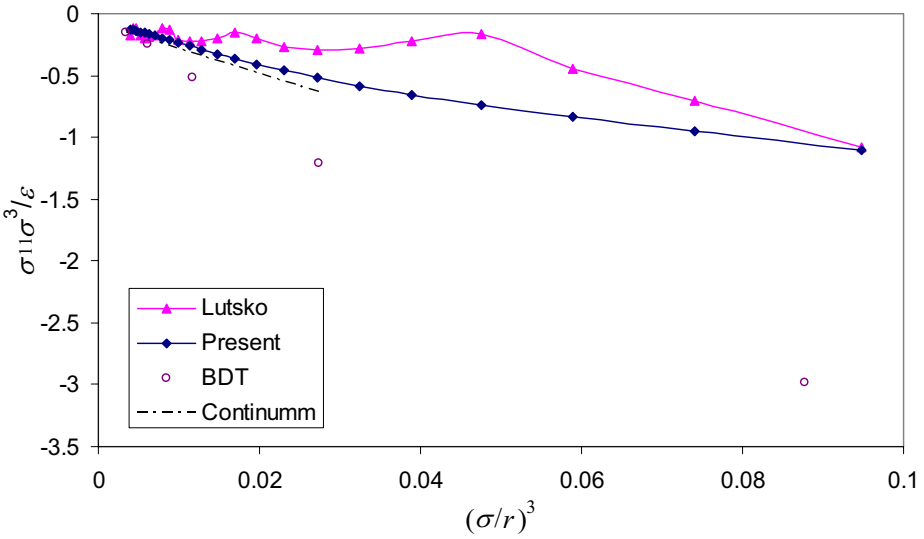

Figure 6 : σ_{11} values for inclusion in [110] direction

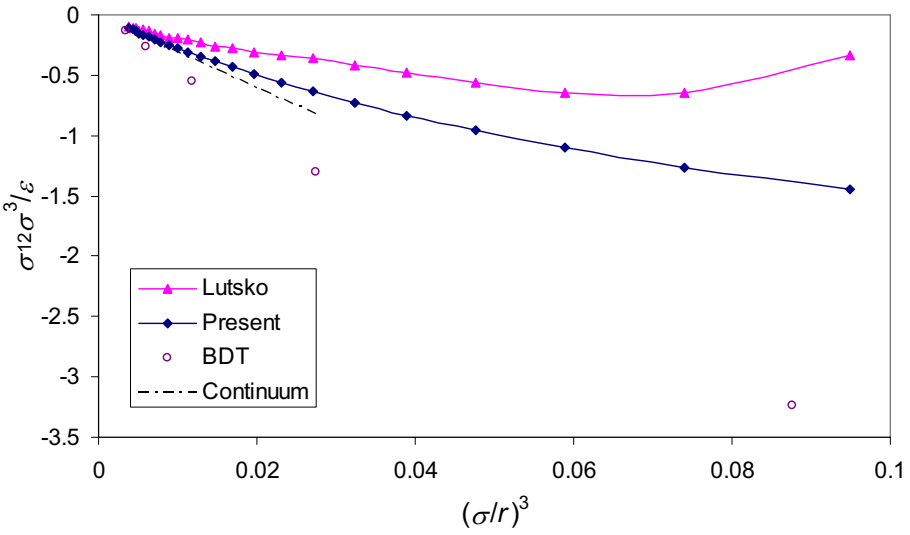

Figure 7 : σ_{12} values for inclusion in [110] direction

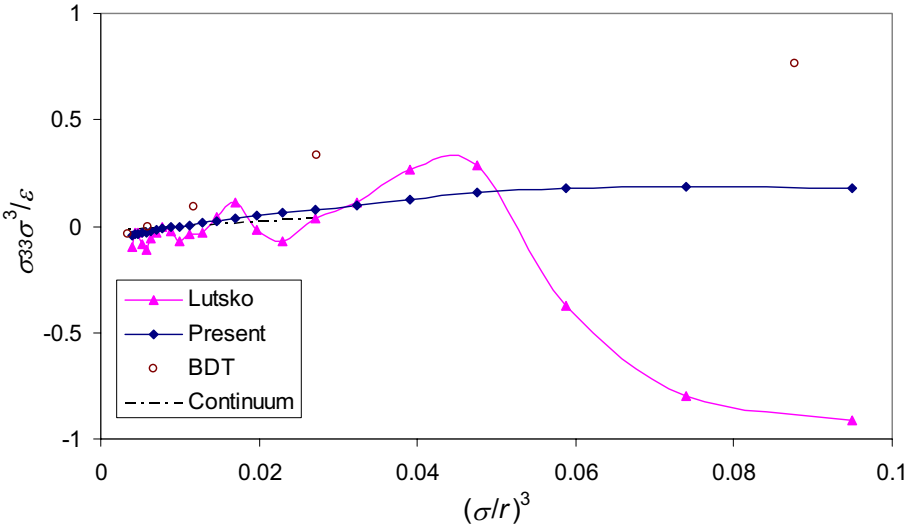

Figure 8 : σ_{33} values for inclusion in [110] direction

well in this case.

5 Conclusions

An atomistic level stress tensor is proposed with physical clarity, based on the SPH method. This stress tensor rigorously satisfies the conservation of linear momentum, and is appropriate for both homogeneous and inhomogeneous deformations. It is in a nonvolume-average form, and thus does not involve *ad hoc* specification of a relevant volume, whose extent is not always obvious: in atomic ensembles with irregular atom arrangement, the identification of this volume can be ambiguous. In contrast to this developed atomistic level stress tensor, other widely used stress tensors in atomistic analysis are in a volume-average form, and do not satisfy the conservation of linear momentum. The formulation is easy to implement, and is validated for both the homogeneous deformation, as well as defected crystalline solids. The numerical results show that the present formulation is very robust and accurate, and is superior to BDT and Lutsko stress formulations. Our numerical results also confirm that the BDT stress tensor is only appropriate for the homogeneously deformed system. The averaging volume and the location affect the Lutsko stress very much. A large averaging volume is required to get a stable value of the Lutsko stress, even for homogeneous deformations.

An equivalent continuum is also defined for molecular dynamics system, based on the developed definition of atomistic stress and in conjunction with the SPH technique. The process is simple and easy to implement, and the fields are with high-order continuity. This equivalent continuum is a reinterpretation of the discrete atomistic force and deformation fields of the MD system. This development provides a systematic approach to the continuum analysis of the discrete atomic system. It can also be applied to multiscale modeling of material behavior which combines both the MD and continuum descriptions in the development of constitutive relations at different scales.

The atomistic stress tensor derived in this paper will play an important role in the multiscale simulation [Srivastava, Atluri (2002a, b); Garikipati (2002); Ghoniem, Cho (2002)] and in molecular dynamics. The idea cannot be limited to mechanical properties, for it can be easily applied to multiscale modeling, directly linking the electronic structure level to the continuum level. A multiscale simulation based on this stress formulation is presented in another companion paper. It is also noted that the use of the SPH approximation is not central to the idea of continuum-stress presented here. Alternate approximation will be discussed elsewhere.

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References

Atluri, S. N. (2002): Seminars at the University of Florida, Virginia Tech, and the Office of Naval Research.

Atluri, S. N. (2004): *The meshless method (MLPG) for domain & bie discretizations*. Tech Science Press, USA.

Atluri, S. N.; Shen, S. (2002): The meshless local Petrov-Galerkin (MLPG) method: A simple & less-costly alternative to the finite element and boundary element method. *CMES: Computer Modeling in Engineering & Sciences* 3 (1): 11-52.

Askar, A (1985): *Lattice dynamical foundations of continuum theories*. World Scientific, Singapore.

Bardenhagen, S.; Triantafyllidis, N. (1994): Derivation of high order gradient continuum theories in 2, 3-D non-linear elasticity from periodic lattice models. *J. Mech. Phys. Solids* 42: 111-139.

Basinski, Z. S.; Duesberry, M.S.; Taylor, R. (1971): Influence of shear stress on screw dislocations in a model sodium lattice. *Canadian J. of Phys.* 49: 2160.

Born, M.; Huang, K. (1954): *Dynamical Theory of Crystal Lattices*. Oxford, Clarendon.

Broughton, J. Q.; Gilmer, G. H. (1983): Molecular dynamics investigation of the crystal-fluid interface. I. Bulk properties. *J. Chem. Phys.* 79: 5095-5104.

Chandra, N.; Namilae, S.; Shet, C. (2004): Local elastic properties of carbon nanotubes in the presence of Stone-Wales defects. *Phys. Rev. B* 69: 094101.

Cheung, K. S.; Yip, S. (1991): Atomic-level stress in an inhomogeneous system. *J. Appl. Phys.* 70: 5688-5690.

Clausius, R. (1870): On a mechanical theory applicable

to heat. *Phil. Mag.* 40: 122-127.

Cormier, J.; Rickman, J. M.; Delph, T. J. (2001): Stress calculation in atomistic simulations of perfect and imperfect solids. *J. Appl. Phys.* 89: 99-104.

Garikipati, K. (2002): A variational multiscale method to embed micromechanical surface laws in the macromechanical continuum formulation. *CMES: Computer Modeling in Engineering &Sciences* 3(2): 175-184.

Ghoniem, N.M.; Cho, K. (2002): The emerging role of multiscale modeling in nano and micro-mechanics of materials. *CMES: Computer Modeling in Engineering & Sciences* 3 (2): 147-173.

Li, Q.; Shen, S.; Han, Z-D.; Atluri, S. N. (2003): Application of Meshless Local Petrov-Galerkin (MLPG) to Problems with Singularities, and Material Discontinuities, in 3-D Elasticity, *CMES: Computer Modeling in Engineering & Sciences* 4(5): 567-581.

Lucy, L. B. (1977): a numerical approach to the testing of the fassion hypothesis. *Astrophysical Journal* 82: 1013.

Lutsko, J. F. (1988): Stress and elastic constants in anisotropic solids: molecular dynamics techniques. *J. Appl. Phys.* 64: 1152-1154.

Monaghan, J. J. (1992): Smoothed particle hydrodynamics. *Annu. Rev. Astron. Astrophys.* 30: 543-574.

Machova, A. (2001): Stress calculations on the atomistic level. *Modeling Simul. Mater. Eng.* 9: 327-337.

Shen, S.; Atluri, S. N. (2004a): Multiscale simulation based on the meshless local Petrov-Galerkin (MLPG) method. *CMES: Computer Modeling in Engineering & Sciences* 5(3): 235-255.

Shen, S.; Atluri, S. N. (2004b): Computational nanomechanics and multiscale simulation. *CMC: Computers, Materials, & Continuua* 1(1): 59-90.

Srivastava, D.; Atluri, S. N. (2002a): Computational Nanotechnology: A Current Perspective, *CMES: Computer Modeling in Engineering & Sciences* 3(5): 531-538

Srivastava, D.; Atluri, S. N. (2002b): Computational Nanotechnology, Special Issue, *CMES: Computer Modeling in Engineering & Sciences* 3(5).

Zhou, M. (2003): A new look at the atomic level virial stress: on continuum-molecular system equivalence. *Proc. R. Soc. Lond. A* 459: 2347-2392.

Zhou, M; McDowell, D. L. (2002): Equivalent continuum for dynamically deforming atomistic particle systems. *Philosophical Magazine A* 82: 2547-2574.