Convergence of local atomistic stress based on periodic lattice

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Local stress in an atomic system, which provides an average stress measurement within a spatial volume containing a collection of atoms, is essential for determining the mechanical properties of a nanoscale structure as well as developing a proper multiscale modeling technique. Theoretically, the smaller averaging volume where a local stress can converge, the closer this atomistic stress definition can approach the ideal continuum stress. As a result, the more accurate stress concentration can be evaluated for the inhomogeneous case. With reference to the previous studies focusing on the spherical averaging volume, dependent on the type of crystals, the convergent radius of the virial stress or Hardy stress usually spans the size of several lattice constants. In this paper, we find that, once the averaging volume is periodic, the convergence of the virial stress and Hardy stress can be accomplished within one single lattice, which is much smaller than what is required by other non-periodic volumes such as a sphere. In the final section, a cracked sodium chloride crystal is considered to demonstrate that the crack opening stress described by periodic lattices captures the stress concentration near the crack tip.

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1. Introduction

Continuum theory has demonstrated its value in modeling the mechanical properties of solids and structures. However, as the dimension is downsized to the atomistic scale, continuum stress and atomistic stress may not match each other due to the discrete nature of the atomistic structure. This brings up a question about how we can interpret results from molecular simulations in terms of continuum mechanics. One possible solution to this problem is homogenizing a group of atoms as a material point, similar to the micromechanics in composites materials (Sun and Vaidya, 1996). Since this method has not been standardized, the stress value may depend on the choice of the homogenization technique used. This issue becomes even more pronounced when the size of geometric discontinuity is also reduced to the nanometer range.

In order to connect a phenomenon at nanoscale to the macroscopic continuum variable, there is a need to develop a physically reasonable homogenization technique for obtaining the local stress information in atomistic systems.

In addition, the local atomistic stress definition is emphasized due to the emergence of multiscale modeling. Several different methods, i.e., quasicontinuum (Tadmor et al., 1996), bridging scale method (Wagner and Liu, 2003), bridging domain (Xiao and Belytschko, 2004), have been proposed to extend the length and time scale of molecular simulations. A comprehensive review of multiscale modeling techniques can be found in the work of Miller and Tadmor (2009). Since these multiscale modeling methods involve two or more levels of simulation techniques such as molecular dynamics (MD) and the continuum finite element method (FEM), it is of practical interest to understand how to define the concept of stress in atomistic systems and ensure smooth communication between different scale domains.

The development of stress definitions can be traced back to the 19th century. Clausius (1870) and Maxwell (1870) developed the virial theorem to measure the internal stress of a fixed volume containing the interacted atoms at finite temperatures ($T > 0$ K). Cheung and Yip (1991) revised the virial theorem to calculate the momentum flux and force across an area and claimed their definition can present the correct stress distribution near the free surface. Zhou (2003) pointed out the virial stress including the kinetic contribution is not equivalent to Cauchy stress in continuum. However, Subramaniyan and Sun (2008) performed a thermal stress analysis to demonstrate that both kinetic and potential parts in the virial theorem are essential to interpret the virial stress as Cauchy stress in MD simulations.

Irving and Kirkwood (1950) considered mass, momentum and energy conservations in terms of the statistical ensemble to derive alternative stress formalism. To simplify a series expansion of Dirac delta functions in Irving and Kirkwood’s work, Hardy (1982) and Murdoch (1983) proposed the localization function to represent the probability density distribution. As a result, the discrete
variables such as the atomistic mass, velocity, and total energy per atom, can be mapped into the continuous fields as density, momentum and energy density, respectively. Drawing on the concept of localization function, Ulz and Moran (2012) utilized Gaussian mixture modeling to measure the similarity of atoms and decide if atoms belong to the same space-averaging volume.

Recently, with the focus on perfect crystals under uniform loading, Zimmerman et al. (2004), Branicio and Srolovitz (2009), and Admal and Tadmor (2010) studied the size of a sphere that is necessary for the local virial stress or Hardy stress to converge to the continuum stress. If a single-atom crystal is considered, the convergent radii of the virial stress and Hardy stress cross 4 and 2 times the size of the lattice constant, respectively (Zimmerman et al., 2004; Admal and Tadmor, 2010). As for the multi-atom crystal, the advantage of Hardy’s definition diminishes and both stress definitions require a radius of 5 times the size of the lattice constant to achieve convergence (Branicio and Srolovitz, 2009). The main purpose of this paper is to demonstrate that if the spatial averaging volume is periodic, the convergence of the virial stress and Hardy stress can be accomplished within a single lattice distance. To demonstrate the superiority of the periodic averaging volume, we consider a central crack embedded in a sodium chloride crystal. With comparison to continuum finite element solutions, the crack opening stress from a series of periodic lattices captures more detailed stress concentration than what is presented by other non-periodic averaging volumes.

2. Definition of stresses in atomic systems

For a volume V with center of mass setting in position \( \mathbf{x} \) and moving with velocity \( \mathbf{v} \), the equation of motion of each atom inside is given by

\[
\sum_{i=1}^{N} \mathbf{F}^{\text{int}}_i + \mathbf{F}^{\text{ext}}_i = M_i \ddot{\mathbf{r}}_i
\]

where \( i = 1, 2, 3 \) represent the x, y, or z direction. \( \mathbf{F}^{\text{int}}_i \) is the interaction of atom \( j \) due to atom \( i \), \( \mathbf{b}^i \) is the external force outside the volume, and \( M_i \) is the mass of atom \( i \) as a function of position \( \mathbf{r} \) and velocity \( \dot{\mathbf{r}}_i \), i.e., \( \mathbf{v}^i = \mathbf{v} + \dot{\mathbf{r}}_i \), of atom \( x \) with respect to the center of mass follow the relations as

\[
\sum_{x} \dot{r}_i^x = 0, \quad \sum_{x} \dot{v}_i^x = 0
\]

The summation of tensor product between Eq. (1) and \( (\mathbf{x}^x - \mathbf{x}^b) \), where the atomistic position \( \mathbf{x}^x = \mathbf{x} + \mathbf{r}^x \) and \( \mathbf{x}^b \) is an arbitrary fixed point in space

\[
\sum_{x} \sum_{x \neq b} (\mathbf{x}^x - \mathbf{x}^b) \mathbf{F}^{\text{int}}_i + \sum_{x} (\mathbf{x}^x - \mathbf{x}^b) \mathbf{b}_i^x = \sum_{x} (\mathbf{x}^x - \mathbf{x}^b) M_i (\dot{v}_i^x + \dot{r}_i^x)
\]

Through Newton’s third law, \( \mathbf{F}^{\text{int}}_i = -\mathbf{F}^{\text{ext}}_i \), and the integration by parts, Eq. (3) can be rearranged as

\[
\sum_{x} \sum_{x \neq b} (\mathbf{x}^x - \mathbf{x}^b) \mathbf{b}_i^x = \frac{1}{2} \sum_{x} \sum_{x \neq b} (\mathbf{x}^x - \mathbf{x}^b) \mathbf{F}^{\text{int}}_i
\]

With consideration of Eq. (2) and neglect of pure rigid body terms without contributing deformation, Eq. (4) is simplified as

\[
\sum_{x} (\mathbf{x}^x - \mathbf{x}^b) \mathbf{b}_i^x = -\frac{1}{2} \sum_{x} \sum_{x \neq b} (\mathbf{x}^x - \mathbf{x}^b) \mathbf{F}^{\text{int}}_i - M_i \dot{v}_i^x \dot{r}_i^x + \frac{d}{dt} \int_{\mathcal{V}} M_i^x \mathbf{r}_i^x \mathbf{r}_i^x dV
\]

The last term in Eq. (5) is eliminated if the duration of time average is taken sufficiently long (Murdoch, 2007). In the continuum viewpoint, the moment produced by the traction vector \( \mathbf{t} \) with respect to the distance between a fixed point \( x^a \) and a material point position \( x \) is

\[
\int (\mathbf{x} - \mathbf{x}^a) \mathbf{F} \mathbf{d}S = \int (\mathbf{x} - \mathbf{x}^a) \mathbf{\sigma} \mathbf{d}V = \int \frac{\partial}{\partial \mathbf{x}} (\mathbf{x} - \mathbf{x}^a) \mathbf{\sigma} dV
\]

through the use of the divergence theorem and equilibrium condition. By assuming that the external force terms in the discrete and continuum viewpoints, i.e., Eqs. (5) and (6), are equivalent, we can define the virial stress as

\[
\mathbf{\sigma}_v = -\frac{1}{2} \int_{\mathcal{V}} \left( \mathbf{M}^a (v^a_i - v^a_j)(v^a_i - v^a_j) + \frac{1}{2} \sum_{x} (\mathbf{x}^x - \mathbf{x}^b) \mathbf{F}^{\text{int}, x} \right)
\]

Though only valid for pair potentials, the virial stress definition in Eq. (7) can be generalized for multi-body potentials. As Eq. (7) indicates, the information of calculating the virial stress, e.g., the atomistic velocity and the interaction force between pairs of atoms, is already known through the numerical integration in MD simulations. This straightforward implementation makes the virial theorem the most widely used atomistic stress definition. It is noted that Eq. (7) is derived with \( V \) as the total volume of a simulation box. In order to deal with the stress concentration resulting from the crystal imperfection, the local virial stress is calculated based on the average over a small group of atoms inside the box.

Introducing a localization function \( \psi \), Hardy (1982) transformed the discrete atomistic mass, velocity, and total energy of an atom [potential energy + kinetic energy] into the continuous density \( \rho \), momentum \( \mathbf{p} \), and energy density \( E \) fields at a material point \( \mathbf{x} \)

\[
\rho(\mathbf{x}, t) = \sum_x M^x \psi(r^x)
\]

\[
p_i(\mathbf{x}, t) = \sum_x M^x \dot{v}_i^x \psi(r^x)
\]

\[
E(\mathbf{x}, t) = \sum_x \left( \frac{1}{2} M^x (v^x_i)^2 + U^x \right) \psi(r^x)
\]

where \( r^x = |\mathbf{x}^x - \mathbf{x}| \) is the distance between atom position \( \mathbf{x}^x \) and material point \( \mathbf{x} \). The localization function has the following properties:

1. \( \psi \) has the unit of inverse volume as Eq. (8) implies.
2. \( \psi \) is a normalized function, i.e., \( \int \psi \mathbf{d}V = 1 \).
3. \( \psi \) is peaked at the material point \( \mathbf{x} \) and decays smoothly to zero along the boundary of averaging volume.

In the current study, we consider two kinds of localization functions. One is a step function remaining constant within the averaging volume. Another is the 3rd order polynomial used for the spherical or cylindrical volume

\[
\psi(\mathbf{r}^x) \big|_{\mathbf{r}^x = \mathbf{R}} = 1 - 3 \left( \frac{\mathbf{r}^x}{\mathbf{R}} \right)^2 + 2 \left( \frac{\mathbf{r}^x}{\mathbf{R}} \right)^3
\]

where \( \mathbf{R} \) is the radius of the averaging volume. It is noted that Eq. (11) and its first derivatives are equal to zero along \( r^x = R \). For the cubic volume discussed in the following sections, the isotropic averaging, the same weight for atoms at the same distance from material point, can be maintained by choosing a step function as the localization function. For a more detailed discussion about the anisotropic averaging resulted from the use of non-spherical volumes, the reader is referred to the work of Murdoch and Bedeaux.
On the basis of Eqs. (8) and (9), the material velocity is defined as

\[ v_i(x, t) = \frac{\sum M_i v_i(r)}{\sum M_i} \]  

(12)

Substituting Eqs. (8)–(10) and (12) into the relations of mass, momentum, and energy conservation, Hardy stress is expressed as

\[ \sigma_{ij} = -\sum a_i \left( M_i v_i^2 - v_i \right)(v_i^j - v_j^i) + \frac{1}{2} \sum b_{ij} \left( x_i^j - x_j^i \right) F_{ij} \]  

(13)

Eq. (13) has been shown to be valid for the multi-body potentials (Chen, 2006). The relative velocity between the atomistic velocity \( v \) and material velocity \( \bar{v} \) is considered in the kinetic term, and \( B_{ij} \) represents the weight of the bond length between atom \( a \) and atom \( b \).

\[ B_{ij} = \int_0^1 \psi(\bar{v}^\beta + v^\beta) d\lambda \]  

(14)

where \( r^\beta \) is the interatomic distance and \( r^\beta \) represents the distance between atom \( \beta \) and material point \( x \).

### 3. Local stress calculation in perfect crystals

For a perfect crystal subjected to uniform loadings, three kinds of stress definitions including local atomistic stress (either in virial or Hardy’s theorem), global stress (the whole volume-averaged virial stress), and continuum stress, are supposed to be identical. Therefore, this homogeneous case can provide a reference point for determining the size of the averaging volume necessary for the local atomistic stress to match the continuum stress. Here, we compare the results of spherical and cubic volumes to realize how the different shaped volumes influence the convergence of the virial stresses and Hardy stress.

In the present study, we consider two crystals: sodium chloride (NaCl) and Nickel (Ni) described by the pair (Catlow et al., 1977) and EAM potentials (Foiles et al., 1986), respectively. Both NaCl and Ni crystals, arranged in cubic orientations with \( x = [100] \), \( y = [010] \) and \( z = [001] \), contain 50 lattices in three directions. The MD simulations are performed using the package LAMMPS (Plimpton, 1995) with a time step of 1 fs. Under periodic boundary conditions in both \( x \) and \( z \) directions, the tensile loading is applied by moving the two outmost layers along the \( y \) direction under the canonical (NVT) ensemble. For all the specimens in this paper, the displacement increment \( \Delta u_y = 0.02 \text{ Å} \) is applied within 1 ps, followed by relaxation (fixing the positions of the outmost layers) for 2 ps and data collection (time averaging) for another 2 ps. Thus, the engineering strain is defined as

\[ \varepsilon_{yy} = \frac{u_y}{L_0} \]  

(15)

where \( u_y \) is the total applied displacement and \( L_0 \) is the height after initial relaxation. The following sections discuss the variation of local virial stress and Hardy stress against the increasing size of averaging volume under \( \varepsilon_{yy} = 0.01 \).

#### 3.1. Sodium chloride (NaCl)

Focusing on the spherical volume of radius \( R \), i.e., \( V = \frac{4}{3} \pi R^3 \), Fig. 1 shows with the increasing radius \( R \) (normalized by the lattice constant of NaCl at the temperature of 1 K, \( a = 2.786 \text{ Å} \)), the local stress \( \sigma_{yy} \) calculated either by the virial or Hardy’s theorem initially oscillates drastically and then approaches the continuum (global) stress. The local virial stress needs to have a volume of radius at least 5 times larger than the size of the lattice constant to reach the converged value. On the other hand, the Hardy stress shows dependence on the choice of localization functions due to different results from two types of localization functions. The convergence of the step function is very slow and the fluctuation is still noticeable at \( R = 10a \). Meanwhile, the 3rd order polynomial enables the Hardy stress to converge once \( R > 3a \). This verifies the statement in previous studies (Zimmerman et al., 2004; Admal and Tadmor, 2010) that the Hardy stress with high order localization functions performs a quicker convergence than the local virial stress.

Regarding the crystal structure of NaCl, a simple symmetric cubic lattice where the sodium and chloride ions are arranged alternately in three-dimensional space, we assert a cubic volume could be a more suitable candidate than a sphere. By varying the half-length \( R \) of a cube with volume \( V = (2R)^3 \), although the fluctuation of the local virial stress in Fig. 2 is larger than that in Fig. 1, the local virial stress can reach exact value if the dimension of the cubic volume \( R = 1a, 2a, 3a \ldots \) is selected. It is noted that all these cubic volumes that make the local virial stress converge are periodic in the NaCl crystal. We also find the Hardy stress under the step function leads to the same phenomenon as the local virial stress at \( R = 1a, 2a, 3a \ldots \). Therefore, with the periodic averaging volume, both the local virial and Hardy stress can achieve convergence within one lattice, which is much smaller than what is required by the spherical volume.

Periodicity of NaCl crystals can be further discussed by focusing on a single Na\(^+\)–Cl\(^-\) pair. The per-atom stress (Eq. (7) without volume average) of Na\(^+\) and Cl\(^-\) atoms (or ions) are around \( \Sigma_{Na^+} \approx 22 \times 10^3 \text{ MPa \ A}^3 \) and \( \Sigma_{Cl^-} \approx 1.8 \times 10^3 \text{ MPa \ A}^3 \) with fluctuation. For simplicity without loss of generality, the fluctuation is neglected in this uniform loading case at low temperature. The continuum stress \( \sigma_{yy} = 561.39 \text{ MPa} \) is approached by taking the sum of per-atom stress values within a single Na\(^+\)–Cl\(^-\) pair divided by its periodic volume \( 2a^3 \). Since it is the minimum periodic volume found in NaCl crystals, the other periodic volumes are supposed to be the multiple of \( 2a^3 \). If the periodicity of lattices is set up based on the red square in Fig. 3 (\( R = 0.5a \)), all the eight atoms are located at corners, representing each atom occupies one-eighth portion. The local virial stress \( \sigma_{yy} \) within this periodic volume can be calculated as

\[ \sigma_{yy} = \frac{1}{a^3} \left( \frac{1}{8} \times 4 \times \Sigma_{Na^+} + \frac{1}{8} \times 4 \times \Sigma_{Cl^-} \right) = \frac{1}{2a^3} \left( \Sigma_{Na^+} + \Sigma_{Cl^-} \right) \]  

(16)

![Fig. 1. Local stress averaged within the spherical volume of increasing radius R in NaCl subjected to applied strain \( \varepsilon_{yy} = 0.01 \).](image-url)
values averaged over different sizes of periodic volumes, e.g., for 
periodic volume \( a^3 \) (the lattice constant \( a = 3.517 \) Å at the temperature of 1 K), the convergent radii of 
non-periodic stresses are smaller than those of the corresponding local-
ization functions in NaCl crystals (see Fig. 1).

Because an FCC crystal is not arranged in simple cubic lattices,
Fig. 5 shows that the local virial stress and Hardy stress averaged 
with the cubic volume cannot reach the exact value at 
\( R = 1a, 2a, 3a, \) etc. Even though the normal stress varies regularly 
with the increase of the half-length \( R, \) a periodic volume in the 
FCC crystal is unlikely to be found by simply taking a cubic volume. 
Following the same procedure used in NaCl crystals, the contin-
um stress \( \sigma_{yy} = 2310.1 \) MPa) can be obtained by the per-atom 
stress at \( \sigma_{yy} = 0.01. \)

3.2. Nickel

Based on the Nickel atoms grouped together in the FCC crystal 
(the lattice constant \( a = 3.517 \) Å at the temperature of 1 K), the 
comparison between the local virial and Hardy stress averaged 
over spherical volumes is demonstrated in Fig. 4. The superiority 
of the Hardy stress in the single-atom system is even more prom-
inent than in the multi-component atomistic system since the con-
vergent radii of \( R = 2a \) for the 3rd order polynomial and \( R = 4a \) for 
the step function are smaller than those of the corresponding local-
ization functions in NaCl crystals (see Fig. 1).

Focusing on the virial theorem, the local stress within a perfect 
NaCl crystal at room temperature, 300 K, is estimated by the 
square-shaped and cylindrical volumes, respectively. These two 
volumes span over the thickness of the NaCl system consisting of 
10 unit cells in the z direction. Regarding the lattice constant 
expanded from 2.786 to 2.814 Å as the temperature increases from 
1 to 300 K, Fig. 6 demonstrates that the idea of periodic volume 
is still applicable at elevated temperature since the local virial 
stress averaged over the square-shaped volume achieves the global 
stress at \( R = 1a, 2a, 3a \) and so on. On the other hand, similar to the 
result at the temperature of 1 K, the convergent radius of non-peri-
odic (cylindrical) volume is about 5 times the size of the lattice 
constant.

4. Crack opening stress in NaCl crystals

One purpose of developing the local atomistic stress is to study stress 
concentration (inhomogeneity) in crystals. In this study, the 
non-uniform stress distribution is induced by embedding a crack in 
the NaCl crystals. As Fig. 7 shows, a central crack along thexz plane is 
created by cutting 6 or 30 (denoted as 6p or 30p model, respectively) 
interaction pairs. The specimen size is \( 417.9 \times 563.4 \times 27.9 \) Å

Fig. 2. Local stress averaged within the cubic volume of increasing half-length \( R \) in 
NaCl subjected to applied strain \( \varepsilon_{yy} = 0.01. \)

which is the same result from a single Na\(^+\)–Cl\(^-\) pair over its periodic 
volume \( 2a^3. \) As indicated by green dots in Fig. 2, the \( \sigma_{yy} \) values averaged 
over different sizes of periodic volumes, e.g., for \( R = 1.5a \) (blue 

cube in Fig. 3), 8 atoms inside the volume (entire portion), 24 

atoms located on the surface (half portion), 24 atoms along the 
intersection between two planes (quarter portion), and 8 corner 

atoms (one-eighth portion), are consistent with the continuum (global) 
stress.

3.3. NaCl at elevated temperature

Focusing on the virial theorem, the local stress within a perfect 
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Fig. 3. Periodic volume to calculate the local stress in NaCl. (For interpretation of 
the references to color in this figure legend, the reader is referred to the web version 
of this article.)

Fig. 4. Local stress averaged within the spherical volume of increasing radius \( R \) in 
Ni subjected to applied strain \( \varepsilon_{yy} = 0.01. \)
containing 150, 200, and 10 lattices in the \( x \), \( y \), and \( z \) directions, respectively. There have been some studies calculating the crack-tip stress field in the MD simulation by use of the virial or Hardy stress definition (Cleri et al., 1998; Mattoni et al., 2004; Buehler et al., 2006; Tsai et al., 2010). Some researchers (Yamakov et al., 2006; Krull and Yuan, 2011; Paliwal and Cherkaoui, 2013) further employed the concept of local atomistic stress to extract the traction–separation relation in molecular models. In this study, focusing on the failure conditions of brittle solids, we investigate the influence of stress averaging techniques on the near-tip stress distribution and related fracture toughness in terms of the critical stress intensity factor.

4.1. Global stress

Applying displacement loading (\( y \) direction) and periodic boundary condition (\( x \) and \( z \) direction), the global stress versus engineering strain curves of a perfect crystal and two cracked specimens are demonstrated in Fig. 8. The global stress of the perfect crystal is equivalent to the uniform continuum stress and thus the theoretic strength of NaCl is determined as \( \sigma_{th} = 3.43 \text{ GPa} \). As for the cracked specimens, since the continuum stress is non-uniform, the global stress can only provide the information of an entire cracked specimen, which indicates the failure condition corresponding to the maximum global stress, i.e., \( \varepsilon_{cr} = 0.039621 \) for the 6p-type model and \( \varepsilon_{cr} = 0.020638 \) for the 30p-type model.

4.2. Stress-averaging volumes

To describe the crack opening stress by the local atomistic stress definitions, the blot dots in Fig. 9(a) (the midpoints of each lattice along the crack line) represent the centers of averaging volumes or the material points in Hardy’s definition. In addition, Fig. 9(a) shows that the initial crack tip is assumed to be at the location of the first \( \text{Na}^+–\text{Cl}^- \) pair without removing the interaction, and the lattices are numbered according to their positions from the initial crack tip.

Following the square-shaped and cylindrical volumes used in Section 3.3 (perfect crystal at elevated temperature), Fig. 10 illustrates that the crack opening stress calculated by the square-shaped volume with volume \( V = 5.57 \times 5.57 \times 27.86 \text{ Å}^3 \) (\( 2a \times 2a \times 10a \), \( a = 2.786 \text{ at the temperature of 1 K} \)). Although this square-shaped volume spans over the entire thickness, it still
maintains the periodicity in space. In contrast, the cylindrical volume with the axis lying along the z direction is treated as the non-periodic volume. There are two radii considered in the cylindrical volume, 8.36 Å (3a) and 13.93 Å (5a), which correspond to the convergent radius of the Hardy and local virial stress in Fig. 1, respectively. For simplicity, the sizes and shapes of averaging volumes do not alter with the deformed crystal under small deformation assumption.

4.3. Crack opening (local) stress distributions

Take a look at the specimen with the larger crack size (30p-type model) first. While the global stress of the cracked specimen reaches the maximum (the failure condition \( \varepsilon_{cr} = 0.020638 \) in Fig. 8), Fig. 11(a) (with numbers to mark the positions of lattices in Fig. 9) shows the crack opening stress distributions based on the virial theorem. The result of the square-shaped volume shows the lattice that gives rise to the maximum normal stress is the second lattice ahead of the initial crack tip and this maximum value is close to the theoretic strength \( \sigma_{cr} = 3.43 \) GPa of perfect NaCl crystals. This provides the evidence that the maximum stress that a lattice can sustain is equivalent to the theoretic strength of the perfect crystal. Thus, instead of considering the critical stress intensity factor based on stress singularity from continuum fracture mechanics, it is natural to use the maximum stress as the fracture criterion in molecular models. Besides, according to our previous study (Cheng and Sun, 2014), the reason why the failure of a cracked specimen is not related to the maximum normal stress of the first lattice but the second one is because of the artificial cutting bond to initiate a central crack.

The continuum crack opening stress is determined by the two-dimensional plane strain FEM model. To maintain the equivalence between the MD and FEM model, except for the periodic boundary condition in the transverse direction and the identical magnitude of applied displacement, one consideration is that the crack tip no longer stays at the initial position but moves toward a lattice constant as shown in Fig. 9(b). We have this consideration because the crack tip position is assumed to follow the lattice with the maximum normal stress. To better explain, the distance from the present crack tip to the center of each lattice along the crack line, which is employed as the horizontal axis in Fig. 11, is also indicated in Fig. 9(b). Another consideration is that the material properties are not constant but decreasing along with the increasing applied strain due to the nonlinearity of the potential function, shown as the stress–strain curve of the perfect crystal in Fig. 8. To deal with this situation, in the stress–strain curve of the perfect crystal, the secant modulus can be defined as the slope of the secant straight line from the origin to the global stress corresponding to the level of applied strain. It turns out that the secant moduli are \( C_{11} = 52.82 \) GPa and \( C_{12} = C_{44} = 12.64 \) GPa (due to the pair potential) at \( \varepsilon_{yy} = 0.020638 \).

Based on the secant moduli and the crack length = 91.94 Å (initial crack length plus two sizes of lattice constant), the FEM solution for the critical strain \( \varepsilon_{cr} = 0.020638 \) is presented as the black line in Fig. 11. The virial stress averaged over the square-shaped volume closely matches with the FEM solution except for the lattice closest to the crack tip (the second lattice) where the stress approaches infinity according to continuum fracture mechanics. This explains that the atomistic stress under the minimum periodic volume provides the discrete information to calculate “point” continuum stress. As for the cylindrical volume with the radius 8.36 Å (3a) where the local virial stress does not converge (see Fig. 1), the corresponding crack opening virial stress deviates significantly from other distributions. Once the radius of the cylindrical volume is increased to 13.93 Å (5a), even though the performance of the virial stress is improved (the consistency with the FEM solution for the distance from the present crack tip larger than 16a), the near-tip stress distribution does not match with the FEM solution or the local stress averaged over the square-shaped volume.

As for Hardy’s definition, the crack opening stress distributions calculated by either the combination of the square-shaped volume
is the crack opening stress under failure condition and \( \sigma_{th} = 47.19 \) GPa, \( \sigma_0 = 12.25 \) GPa, crack length = 25.07 Å), in contrast to the more agreeable results of virial stress over square-shaped volume and Hardy stress with the smaller cylindrical localization volume \( (R = 3a) \). This illustrates that even with the use of Hardy's definition, the smaller averaging volume is still required to capture the actual near-tip stress distribution as crack length (stress concentration) decreases.

4.4. Critical stress intensity factor

The significance of the stress-averaging volume size can be further acquired by estimating the critical stress intensity factor \( K_{ic} \) defined as

\[
K_{ic} = \lim_{x \to 0} \sigma_{yy} \sqrt{2\pi x}
\]

where \( \sigma_{yy} \) is the crack opening stress under failure condition and \( x \) is the distance from the present crack tip. Given the distance from the crack tip position in Fig. 9(b), we multiply the crack opening stress distributions in Figs. 11 and 12 by \((2\pi x)^{0.5}\) to extract the stress intensity factor \( K_{ic} \). These results are plotted in Fig. 13(a) and (b) where numbers (2–6) represent the positions of lattices in Fig. 9(b). In Fig. 13(a) of the 30-type model, while the \( \sigma_{yy}(2\pi x)^{0.5} \) distributions in Hardy's definition approach the FEM solution after the fourth lattice from the initial crack tip, the matched virial result averaged by the minimum periodic volume starts from the third lattice. These two locations provide the starting points to project stress intensity factors shown as dash lines in Fig. 13. The critical stress intensity factor is estimated as the intersection of the projection line and the \( y \)-axis. The related values against different stress averaging techniques can be found in Table 1. It is noted that \( K_{ic} \) with step function or cylindrical volume with 3rd order polynomial are shown in Fig. 11(b). In comparison to the continuum FEM solution, the Hardy stress with the square-shaped volume and step function demonstrates the same accuracy as the local virial stress averaged over the same volume in Fig. 11(a) except for the lower peak stress. Such lower value of the maximum stress indicates when the extremely large stress gradient happens in the lattice, the local virial stress leads to a more precise result than the Hardy stress based on the step function. The result from the cylindrical volume with radius 8.36 Å \( (3a) \) in Fig. 11(b) is better fitted into the FEM solution than the corresponding virial stress since the Hardy stress reaches the converged value for \( R > 3a \). It is important to note that the Hardy stress, averaged over the cylindrical volume with radius 13.93 Å \( (5a) \) where both stress definitions converge (see Fig. 1), still performs better than the corresponding virial stress due to the high order localization function to decrease the contribution from the high stress level in the near-tip region.

Focusing on the specimen with smaller crack length (6p-type model), Fig. 12 shows the near-tip stress distributions determined by the virial stress (averaged by square-shaped volume in Fig. 10) and the Hardy stress over cylindrical volumes with 3rd order localization function. Under the failure condition of \( \varepsilon_{fr} = 0.039621 \), the peak value of local virial stress (at the second lattice from the initial crack tip) approaches the theoretic strength of perfect NaCl crystals \( \sigma_{fr} = 4.34 \) GPa. This is the same phenomenon happening in the 30p-type model as shown in Fig. 11(a). Therefore, we can say as the crack length decreases, the degree of stress concentration decreases.
the larger cylindrical volume ($R = 3a$) because its volume size is much larger than the square-shaped volume composed by periodic lattices.

5. Conclusions

The evidence from our study showed that if both the local virial stress and Hardy stress are averaged over a periodic volume, the convergence can be accomplished within one single lattice. To verify stress concentration preserved by this stress-averaging technique, the NaCl crystals with two sizes of crack lengths were utilized to show that the crack opening stress distributions, determined by the virial and Hardy stress with periodic volumes, are consistent with the continuum FEM solution. By contrast, under the spherical or cylindrical (non-periodic) averaging volume, even though Hardy’s definition with the 3rd order localization function performs a quicker convergence than the corresponding virial theorem, the spherical radius at which the Hardy stress converges still spans the size of several lattice constants. Therefore, the non-periodic volume loses the resolution to reflect the actual stress distribution around the crack tip because of the larger size to converge. The situation may deteriorate when the crack length is downsized, which produces an inaccurate stress intensity factor compared with results of the virial stress based on periodic volumes and FEM solutions.

It is also noted that the original idea of the virial and Hardy averages is to deal with the general cases, i.e., gas, liquid and amorphous materials, which requires the capability of placing a material point arbitrarily for stress-averaging techniques. The periodic lattice built on knowing the atomistic structure in advance limits the choice of material points. However, on the basis of fracture criteria, since the local virial stress over the square-shaped volume demonstrates the validity of the maximum normal stress criterion by presenting the equivalence between the peak stress and theoric strength, and estimates the critical stress intensity factor accurately if continuum fracture mechanics is considered, the periodic lattice is an appropriate stress averaging technique to describe the failure mechanism in crystal structures. How to include the large deformation using the averaging volume mapped by distorted crystals and extend the present study from single-crystal to nano-crystalline materials will be a subject of future study.

### References

Clausius, R., 1876. On a mechanical theorem applicable to heat. Phil. Mag. 40, 122–127.

### Table 1

Comparison of critical stress intensity factors obtained by different atomistic stress averaging techniques and FEM.

<table>
<thead>
<tr>
<th>Model</th>
<th>Virial periodic volume</th>
<th>Hardy cylinder ($R = 3a$)</th>
<th>Hardy cylinder ($R = 5a$)</th>
<th>FEM</th>
</tr>
</thead>
<tbody>
<tr>
<td>30p</td>
<td>$12.7253 \times 10^4$</td>
<td>$11.9409 \times 10^4$</td>
<td>$11.8587 \times 10^4$</td>
<td>$12.4224 \times 10^4$</td>
</tr>
<tr>
<td>6p</td>
<td>$11.8348 \times 10^4$</td>
<td>$11.8729 \times 10^4$</td>
<td>$10.0274 \times 10^4$</td>
<td>$11.4576 \times 10^4$</td>
</tr>
</tbody>
</table>


Maxwell, J.C., 1870. On reciprocal figures, frames and diagrams of forces. Trans. R. Soc. Edinburgh XXVI, 1–43.


