Spectral Analysis of Stress States in Atomistic Single Crystal Models

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Abstract
A molecular dynamics study of stress states in a single crystal is presented in the frequency domain. The material with finite and infinite boundaries is represented with atomistic representative volumes. Gold lattice is used for numerical illustrations. The effective stresses in the gold lattice are studied using equilibrium molecular dynamics (EMD) force potentials. The temperatures at which stress states are determined are reached by using a systematic heating and equilibrium procedures that produce a convergent energy state. Total mean energy and variance obtained at the equilibrium state is compared for different heating rates. The dynamics stress states are observed in the frequency domain and the effect of the model geometry and boundary conditions on the natural modes of vibration has been studied.

1. Introduction
Over the last decade, computer simulations based on the molecular dynamics (MD) are increasingly being used to study the mechanical behavior of nanostructures [1-3]. Empirical potentials like the embedded atom method (EAM) [4-5] are often used to determine the behavior of atomistic ensembles with the goal of extrapolating bulk material behavior from such results [6-15]. Basinski, Duesbery, and Taylor [16] proposed volumetric partition of the bulk, homogeneous stress tensor characterizes the stress in a small volume about an atom even in an inhomogeneously deformed atomic assemblage. The formalism developed by Hardy [17] use a finite-valued and finite-ranged localization function that contains atoms contributing to properties at the spatial point and show that the resulting expression for stress contains terms that theoretically remain constant for different size volumes. Zimmerman et al. [18] review Hardy’s formalism and present a computational comparison between Hardy’s expression for stress and local volume averages of the virial stress within a FCC crystal. Zhou [15] examined the virial stress and stated that it is not equivalent to the Cauchy stress in the continuum. But recently Subramaniyan et al. [19] reiterate that virial stress is equivalent to Cauchy stress. In this effort, elastic vibration behavior of single crystal materials is studied using equilibrium molecular dynamics. The spectral response of the viral stress is determined and compared to expected response of equivalent elastic continuum. Equilibrium temperatures are obtained by increasing the temperature in small increments over several simulation steps.

2. Atomistic model of single crystal material
The computational domain considered represents a gold lattice with a 4.08 Angstrom (Å) face centered cubic structure. The geometry modeled as shown in the Figure 1 with a, b and c as the side length. The model axes, x, y and z axes are oriented in the [100], [010] and [001] crystal axes, respectively. Both periodic boundary conditions representing infinite lengths and finite boundaries are considered. Table 1 shows the dimension of the models chosen and the notation used for denoting the boundary conditions used in the analysis. Finite boundaries (F) are modeled as traction free surfaces. Hence an IFI boundary as in Case 2 of Table 1, represents a thin plate (163.2 Å) with infinite boundaries. The material models typically consisted of 266,000 atoms.

Figure 1 Model and dimension parameters used in this analysis
The interatomic potential is modeled using the embedded atom method (EAM) developed by Daw and Baskes [4, 5]. The EAM potential consists of a many body term representing the interaction of an embedding atom core with the electronic charge density of the remaining atoms in the system and a pair wise term representing the electrostatic interactions between the atoms cores. The many body term is referred to as the embedding function. The total energy $E$ of a system of $N$ atoms can be written as

$$E_{\text{total}} = \sum_{i}^{N} \left( F_{i}(\hat{p}_{i}) + \frac{1}{2} \sum_{j \neq i}^{N} \phi_{ij}(r_{ij}) \right)$$  \hspace{1cm} (1)$$

where $\phi_{ij}$ is the pair potential function between atoms $i$ and $j$ and $r_{ij}$ is the distance between atom $i$ and $j$, while $F_{i}$, the embedding term, is the quantum mechanical energy involving the influence of electron density. For atomistic Gold, the parameters for Morse pair potential as per Voter and Chen [14] are $r_{c}(A)=5.5155, \beta_{M}(1/A)=3.6967, \alpha_{M}(1/A) = 1.8964, D_{M}(eV) = 0.6748, R_{M}(A)=2.5686$.

In molecular simulation studies, virial stress averaged over the total volume $\Omega$ is often found to be useful to study macroscopic material behavior to analyze stress distribution inside the system. The average virial stress is given as [15]

$$\sigma_{ij} = -\frac{1}{\Omega} \sum_{a=1}^{R} \left( m^{a} v^{a}_{i} v^{a}_{j} - \frac{1}{2} \sum_{\beta \neq \alpha} f^{a\beta}_{i} r^{a\beta}_{ij} \right)$$  \hspace{1cm} (2)$$

where, $N^{R}$ is the number of atoms in the region, $m^{a}$ is the mass, $v^{a}_{i}$ is the $i$-th component of the velocity of atom $a$, $f^{a\beta}_{i}$ is the $i$-th component of force between atom $\alpha$ and $\beta$; and $r^{a\beta}_{ij}$ is the $j$-th component of centre distance between atom $\alpha$ and $\beta$. It can be seen that that Eq. (2) represents average atomic stresses for the volume of the periodic box. Here, the first term is associated with the kinetic energy due to thermal vibration and the second term is related to change in potential energy due to applied deformation. Although the use of this measure to represent continuum stress state is question Zhao [14] and later studies by Subramanian and Sun [19] show that Eq.(2) represents the continuum stress when properly averaged over time and space as well as after removing any rigid body motion of the atomistic ensemble.

### 3. Temperature state determination

In order to determine the configuration of the ensemble and the initial vibrational states at a given temperature, a slow heat and dwell methodology is used. The heating and dwell rates are varied to obtain consistent and convergent ensemble energy and vibration states. Figure 2 shows five heating and dwell cycles in which an atomistic ensemble (Case-2) is heated from 0 K up to 250 K. For heating step used Nose/Hoover temperature thermostat and then it is equilibrated at this temperature for the time (dwell) by using NVT conditions. The process of heating and equilibration is repeated for every 50K rise in temperature; till it reaches an equilibrium temperature of 250K.

![Figure 2 Total energy profiles during temperature equilibrium process.](image)

The total energy during each equilibrium process is shown in the Figure 2. Although the mean equilibrium energy is convergent for various heating rates, the variance is considerably lower for slow heating rates. Total energy mean and standard deviation values at equilibrium condition at 250K are presented in the Table 2. In this study we use the results from the slowest heat-dwell rate in which the 250K is achieved over 5 steps and 7 nanoseconds of simulation time. The energy and stress states are determined at temperatures ranging from 250-1000K.

<table>
<thead>
<tr>
<th>Case</th>
<th>Boundary conditions</th>
<th>Dimensions</th>
<th>Numbers of atoms</th>
<th>Representation</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Infinite</td>
<td>Infinite</td>
<td>163.2</td>
<td>[I I I]</td>
</tr>
<tr>
<td>2</td>
<td>Infinite</td>
<td>Finite</td>
<td>163.2</td>
<td>[I F I]</td>
</tr>
<tr>
<td>3</td>
<td>Finite</td>
<td>Finite</td>
<td>163.2</td>
<td>[F F F]</td>
</tr>
</tbody>
</table>

Table 1 boundary conditions, dimensions and numbers of atoms of the gold lattice in x, y and z directions
4. Temperature-dependent spectral response

After the temperature equilibrium process step, the ensemble is further equilibrated under NVT conditions and the virial stress is computed at 0.01 ps time steps. The fundamental modes of vibration are determined using Fast Fourier Transformation (FFT) for 60000 samples. Frequency spectra of the hydrostatic stress component of the virial stress are plotted for equilibrium temperatures of 200K, 600K and 1000K as shown in Figure 3 for case-2. The effect of stiffness loss due to the temperature can be seen as the shift in the fundamental frequency.

5. Effect of model size and boundary conditions

The power spectrum and the phase plots are shown in Figure 4 for the atomistic ensembles defined in Table 2. Case I has infinite boundaries and vibration spectra show broadband behavior with no clear structural modes and representative of high frequency lattice vibration modes. For case 2 with [I F I] boundary conditions, the structural vibration modes are visible and are expected to correspond to transverse vibrations of the infinite plate. The frequency of the mode with peak power is seen around 90 GHz. This frequency is thickness and temperature dependent indicating that the relationship to the transverse vibrations of an equivalent continuum. In case of all side finite boundaries [F F F], the fundamental frequency is 82 GHz and indicates the unconstrained vibration of the continuum. The correlation of the elastic stiffness to the observed fundamental frequencies is considered elsewhere due to concerns on brevity of this paper.

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Table 2 Mean and standard deviation of total energy on equilibrium condition at 250 K

<table>
<thead>
<tr>
<th>Case</th>
<th>Mean (μ, fJ)</th>
<th>Standard Deviation (σ)</th>
</tr>
</thead>
<tbody>
<tr>
<td>a</td>
<td>-39.94</td>
<td>78.88</td>
</tr>
<tr>
<td>b</td>
<td>-39.94</td>
<td>52.68</td>
</tr>
<tr>
<td>c</td>
<td>-39.94</td>
<td>31.30</td>
</tr>
<tr>
<td>d</td>
<td>-39.94</td>
<td>19.43</td>
</tr>
<tr>
<td>e</td>
<td>-39.94</td>
<td>12.76</td>
</tr>
</tbody>
</table>

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Figure 3 Effect of temperature on the hydrostatic stress amplitude with [I F I] boundary condition

Figure 4 Spectrum response and phase plot of the hydrostatic stress in gold lattice [163.2x163.2x163.2] Å³ for (a) [I I I] (b) [I F I] and (C) [F F F] boundary conditions.
6. Viral stress distributions along the thickness

To examine the viral stress distribution along the thickness of the model, stress components are plotted by averaging the virial stresses at various regions of the model. Three different areas are identified in y direction as shown in Figure 5 and denoted as top, middle and bottom sections. In each of these sections, stress component in y direction \( (\sigma_{yy}) \) is plotted as shown in Figure 6. It shows that the amplitude of stress component \( (\sigma_{yy}) \) near the top and bottom boundaries are considerably lower indicating correlation with the traction-free boundary condition on the surface.

![Figure 5 Three different sections of the model](image)

![Figure 6 Frequency response of \( \sigma_{yy} \) in ( ______ ) upper and ( _ _ _ _ ) middle sections of model.](image)

7. Concluding remarks

This effort focuses on the study of dynamic stress and energy in the single crystal materials. We examined the gold lattice using EAM inter atomic potential and with different set of boundary conditions. The initial condition for the gold lattice is reached by heating the model by following five different equilibrium procedures. Individual stress components and hydrostatic stress states are examined in the frequency domain for three different cases. The fundamental frequencies excited correlate with the expected continuum behavior with appropriate temperature, length and boundary condition effects.

References