COUPLED ATOMISTIC-CONTINUUM ANALYSIS OF NANOWIRES AND NANOFILMS

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ABSTRACT

We present a formulation for coupling atomistic and continuum mechanical simulation methods for quasistatic analysis. The formulation assumes a finite element mesh covers all parts of the computational domain, while atomistic crystals are introduced only in regions of interest. Moreover, the formulation allows the geometry of the mesh and crystal to overlap arbitrarily. Our approach uses interpolation and projection operators to link the kinematics of each region, which result in a system potential energy from which we derive coupled equilibrium equations. A hyperelastic finite element formulation is used to compute the deformation of the defect-free continuum using the Cauchy-Born rule. A correction to the Cauchy-Born rule model is introduced in the overlap region to minimize fictitious boundary effects. Key features of our approach will be demonstrated with simulations in 1- and 2-dimensions.

1 INTRODUCTION

The rationale to construct and use a coupled atomistic-continuum simulation approach is straightforward to comprehend in this age of materials modeling. While continuum material models are evolving to include more physical bases, they can only reproduce anticipated deformation phenomena. In contrast, atomistic simulation, by its use of simple interatomic potentials, has the ability to display competing mechanisms of material deformation, such as fracture and dislocation nucleation and propagation, although limits of computational power prohibit analysis of micro-scale systems, even for large-scale, parallel calculations. Early work by Kohlhoff and collaborators [1] created a methodology that combines finite element (FE) analysis with atomistic modeling. More recently, several methods have been introduced, including the Quasicontinuum method by Tadmor, Ortiz and Phillips [2] , Coarse-Grained Molecular Dynamics by Rudd and Broughton [3], Molecular-Atomistic-Ab Initio Dynamics by Broughton, Abraham, Bernstein and Kaxiras [4], and the Bridging Scale method by Wagner and Liu [5]. These coupling methods have been used successfully to simulate phenomena such as crack-grain boundary interactions, dislocation nucleation from nanoindentation and the dynamic fracture of silicon. However, the weaknesses of these methods show that more consideration is needed in developing a coupled atomistic-continuum approach. Specifically, a rigorous methodology for partitioning potential energy between atomistic bonds and continuum strain energy within the overlapping regions needs to be developed.

In this paper, we describe a formulation for atomistic-to-finite element coupling for quasistatic simulation. The formulation covers all parts of the computational domain with a FE mesh, while introducing atomistic crystals only in regions of interest. The goal of the formulation is to allow the geometry of the mesh and crystal to overlap arbitrarily. A hyperelastic finite element formulation is used to compute the deformation of the defect-free continuum using the Cauchy-Born rule[6], supplemented with a correction used in the overlap region to minimize fictitious boundary effects. Key features of our approach will be demonstrated with simulations in 1- and 2-dimensions.

The coupled atomistic-continuum system is shown in Figure 1. A FE mesh covers all parts of the computational domain, while only limited regions of interest, such as crack tips or other defects, are also covered with an atomic crystal. Let the atomistic displacements in the system be written

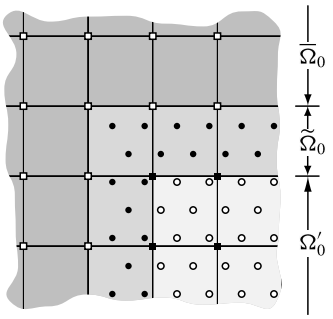


Figure 1: Patch of a coupled atomistic-continuum system. The set of FE nodes $\mathcal{N}$ is shown as open squares □. The set of nodes $\hat{\mathcal{N}}$ is shown as solid squares ■. The set of atoms $\mathcal{A}$ is shown as open circles ○, and the set of atoms $\hat{\mathcal{A}}$ is shown as solid circles ●.

as $\breve{\mathbf{Q}} = \left[\mathbf{q}^{(\alpha)}\right]^T$, where $\alpha \in \breve{\mathcal{A}}$, and $\breve{\mathcal{A}}$ is the set of all atoms. Let the nodal displacements be written as $\breve{\mathbf{U}} = \left[\mathbf{u}^{(a)}\right]^T$, where $a \in \breve{\mathcal{N}}$, and $\breve{\mathcal{N}}$ is the set of all FE nodes. Greek symbols denote atom indices, while lower case Roman symbols denote node indices. In order to satisfy continuity of the displacement field across the atomistic-continuum boundary, the motion of some of the atoms is prescribed by the continuum displacement field. For simplicity, these atoms are called "ghost atoms". This subset of atomistic displacements $\breve{\mathbf{Q}}$ will be denoted as $\hat{\mathbf{Q}} = \left[\mathbf{q}^{(\alpha)}\right]^T$, where $\alpha \in \hat{\mathcal{A}}$, while the complement which contains the unprescribed atomistic displacements (for free atoms) will be denoted as $\mathbf{Q} = \left[\mathbf{q}^{(\alpha)}\right]^T$, where $\alpha \in \mathcal{A}$, $\hat{\mathcal{A}} \cup \mathcal{A} = \breve{\mathcal{A}}$ and $\hat{\mathcal{A}} \cap \mathcal{A} = \emptyset$. Analogously, the motion of some FE nodes is prescribed by the underlying lattice. These displacements will be denoted as $\hat{\mathbf{U}} = \left[\mathbf{u}^{(a)}\right]^T$, where $a \in \hat{\mathcal{N}}$, and the unprescribed nodal displacements will be denoted as $\mathbf{U} = \left[\mathbf{u}^{(a)}\right]^T$, where $a \in \mathcal{N}$, $\hat{\mathcal{N}} \cup \mathcal{N} = \breve{\mathcal{N}}$, and $\hat{\mathcal{N}} \cap \mathcal{N} = \emptyset$. One can interpolate the continuum displacement field to the location of any atom as $\mathbf{u}(\mathbf{X}^{(\alpha)}) = \sum_{a \in \breve{\mathcal{N}}} N^{(a)}(\mathbf{X}^{(\alpha)})\, \mathbf{u}^{(a)}$, where $\mathbf{X}^{(\alpha)}$ is the undeformed position of atom α and $N^{(a)}$ is the FE shape function associated with node a. The FE shape functions typically have compact support, so the sum shown above involves only the nodes whose support includes $\mathbf{X}^{(\alpha)}$. Generally, one can consider the atomistic and continuum displacement fields to be related as

$$\begin{Bmatrix} \mathbf{Q} \\ \hat{\mathbf{Q}} \end{Bmatrix} = \mathbf{N} \begin{Bmatrix} \mathbf{U} \\ \hat{\mathbf{U}} \end{Bmatrix} + \begin{Bmatrix} \mathbf{Q}' \\ 0 \end{Bmatrix}, \tag{1}$$

where

$$\mathbf{N} = \begin{bmatrix} \mathbf{N}_{\mathbf{QU}} & \mathbf{N}_{\mathbf{Q}\hat{\mathbf{U}}} \\ \mathbf{N}_{\hat{\mathbf{Q}}\mathbf{U}} & \mathbf{N}_{\hat{\mathbf{Q}}\hat{\mathbf{U}}} \end{bmatrix}. \tag{2}$$

The sub-matricies of $\mathbf{N}$ contain shape functions as defined by the interpolation stated above. By definition, $\mathbf{N}_{\mathbf{QU}} = 0$ since $\mathbf{Q}$ and $\mathbf{U}$ are independent. In (1), $\mathbf{Q}'$ represents the portion of the atomic

displacements that cannot be represented on an (assumingly) coarser FE mesh through the shape functions $\mathbf{N}_{\mathbf{Q}\hat{\mathbf{U}}}$.

We can solve these coupled equations for the dependent quantities $\hat{\mathbf{Q}}$ and $\hat{\mathbf{U}}$ by minimizing the error $e \equiv \mathbf{Q}'^T\mathbf{Q}'$, as is done in [5]. Klein and Zimmerman[7] have shown that a moving least squares (MLS) interpolation can be substituted for the projection operation, yielding the solutions

$$\hat{\mathbf{U}} = \tilde{\mathbf{N}}_{\hat{\mathbf{U}}\mathbf{Q}}\mathbf{Q}, \tag{3}$$

and

$$\hat{\mathbf{Q}} = \mathbf{N}_{\hat{\mathbf{Q}}\mathbf{U}}\mathbf{U} + \mathbf{N}_{\hat{\mathbf{Q}}\hat{\mathbf{U}}}\tilde{\mathbf{N}}_{\hat{\mathbf{U}}\mathbf{Q}}\mathbf{Q}, \tag{4}$$

where $\tilde{\mathbf{N}}_{\hat{\mathbf{U}}\mathbf{Q}}$ is the matrix of shape functions derived from the MLS interpolation. Notice that the prescribed atomistic displacements depend on both the displacements of the free FE nodes and on the displacements of the free atoms through the projection of those displacements onto the FE mesh.

3 EQUILIBRIUM EQUATIONS

To solve for these unprescribed displacements $\mathbf{Q}$ and $\mathbf{U}$, we develop equilibrium equations derived by formulating the total potential energy of the entire coupled atomistic-continuum system. We express the potential energy of the coupled system as

$$\Pi(\mathbf{Q}, \mathbf{U}) = \Pi_{\mathbf{Q}}(\mathbf{Q}, \hat{\mathbf{Q}}(\mathbf{Q}, \mathbf{U})) + \Pi_{\mathbf{U}}(\mathbf{U}, \hat{\mathbf{U}}(\mathbf{Q})) - \mathbf{F}_{\mathbf{Q}} \cdot \mathbf{Q} - \mathbf{F}_{\mathbf{U}} \cdot \mathbf{U}, \tag{5}$$

where $\Pi_{\mathbf{Q}}$ represents the potential energy in the bonds of the crystal and depends only on $\mathbf{Q}$ and $\hat{\mathbf{Q}}$, $\Pi_{\mathbf{U}}$ is the strain energy density integrated over the continuum and depends only on $\mathbf{U}$ and $\hat{\mathbf{U}}$, and $\mathbf{F}_{\mathbf{Q}}$ and $\mathbf{F}_{\mathbf{U}}$ are external forces acting on the atoms and FE nodes, respectively. The equations of static equilibrium are derived from the total potential and when combined with (3) and (4) can be expressed as

$$\mathbf{R}_{\mathbf{Q}} = \frac{\partial \Pi_{\mathbf{Q}}}{\partial \mathbf{Q}} + \left[\frac{\partial \Pi_{\mathbf{U}}}{\partial \hat{\mathbf{U}}} + \frac{\partial \Pi_{\mathbf{Q}}}{\partial \hat{\mathbf{Q}}}\mathbf{N}_{\hat{\mathbf{Q}}\hat{\mathbf{U}}}\right]\tilde{\mathbf{N}}_{\hat{\mathbf{U}}\mathbf{Q}} - \mathbf{F}_{\mathbf{Q}} = 0, \tag{6}$$

$$\mathbf{R}_{\mathbf{U}} = \frac{\partial \Pi_{\mathbf{U}}}{\partial \mathbf{U}} + \frac{\partial \Pi_{\mathbf{Q}}}{\partial \hat{\mathbf{Q}}}\mathbf{N}_{\hat{\mathbf{Q}}\mathbf{U}} - \mathbf{F}_{\mathbf{U}} = 0, \tag{7}$$

4 CORRECTION TO THE CAUCHY-BORN RULE

The previous sections describe how atomistic and continuum degrees of freedom are coupled; however, the specific form of the total potential has not yet been given. Naturally, the atomistic contribution to the potential energy is computed from a sum of bond energies in the crystal. The continuum strain energy is computed using the Cauchy-Born rule, which accurately describes the long wavelength behavior of the lattice. One important detail is how one corrects for the overlap of the continuum and the underlying crystal. Within this overlapping region, the weighting of the contributions to potential energy from the bonds and finite elements needs to be determined such that the total energy for the coupled system is consistent with the result one would obtain from a full crystal, regardless of the location and orientation of the embedded crystals with respect to the overlaying FE mesh.

We determine immediately that the weighting of the bonds between free and ghost atoms must always be 1 to preserve the energy per atom among free atoms, while the weighting of contributions

from elements containing both active nodes and ghost atoms must be compensated to maintain the correct strain energy density. The approach for introducing weighting into the total potential follows directly from the Cauchy-Born rule. As presented in [7], $\Pi_{\mathbf{U}} = \int \Phi d\Omega$, where $\Phi = \Phi(\mathbf{C}, \mathbf{X})$ is the strain energy density. For a crystal subject to pair interactions, $\Phi(\mathbf{C}, \mathbf{X}) = \frac{1}{V_0} \sum_i^{n_b} \rho_{(i)}(\mathbf{X})\, \varphi\big(r_{(i)}\big)$, where φ is the interaction potential, $r_{(i)} = \sqrt{\mathbf{R}_{(i)} \cdot \mathbf{C} \cdot \mathbf{R}_{(i)}}$, $\mathbf{C} = \mathbf{F}^T\mathbf{F}$, $\mathbf{R}_{(i)}$ is the vector representing bond (i) in the undeformed configuration and $\mathbf{F}$ is the deformation gradient. The spatially varying bond density functions, $0 \leq \rho_{(i)}(\mathbf{X}) \leq 1$, are introduced because some of the energy for bonds in each orientation is represented in the coupled system by actual bonds between atoms present in system. In regions of the domain completely covered by the underlying crystal, $\rho_{(i)}(\mathbf{X}) = 0$ since all bonds are represented at the density of the crystal. Conversely, $\rho_{(i)}(\mathbf{X}) = 1$ over the parts of the domain without any underlying crystal since the Cauchy-Born strain energy density must account for all of the potential energy.

As shown in [7], the functions $\rho_{(i)}(\mathbf{X})$ are determined by enforcing a condition of homogeneous deformation given the appropriate boundary conditions. This is accomplished by minimizing the fictitious forces on the nodes that influence the overlap region within the coupled system. For a system governed by pair potentials, we seek to minimize the quantity

$$\min_{\rho_{(i)}} \left(\frac{1}{2} \sum_{a \in \widetilde{\Omega}_{0(i)}} \Big(\mathbf{R}_{(i)} \cdot \mathbf{f}_{(i)}^{(a)} \Big)^2 + \frac{1}{2} \kappa \int_{\widetilde{\Omega}_{0(i)}} \nabla \rho_{(i)} \cdot \nabla \rho_{(i)} d\Omega \right), \tag{8}$$

where

$$\mathbf{f}_{(i)}^{(a)} = \sum_{\beta \in \widetilde{\Omega}_{0(i)}^{(a)}} \mathbf{R}_{(i)}^{(\beta)} N^{(a)}\Big(\mathbf{X}^{(\beta)}\Big) + \frac{1}{V_0} [\mathbf{R} \otimes \mathbf{R}]_{(i)} \left[\int_{\overline{\Omega}_{0(i)}^{(a)}} \frac{\partial N^{(a)}}{\partial \mathbf{X}} d\Omega + \int_{\widetilde{\Omega}_{0(i)}^{(a)}} \rho_{(i)} \frac{\partial N^{(a)}}{\partial \mathbf{X}} d\Omega \right] \tag{9}$$

represents the force on node (a) due to bond (i), $\mathbf{R}_{(i)}^{(\beta)}$ is the bond vector of type (i) connecting ghost atom β to a free atom, and $\widetilde{\Omega}_{0(i)}^{(a)}$ and $\overline{\Omega}_{0(i)}^{(a)}$ are the elemental volumes that support node (a) in which $0 \leq \rho_{(i)} \leq 1$ and $\rho_{(i)} = 1$, respectively. A detailed derivation of (8) and (9) can be found in [7].

5 COUPLING EXAMPLES

5.1 One-dimensional example

Key features of the coupling formulation can be illustrated with a one-dimensional example. Consider the patch of a coupled system shown in Figure 2(a). The patch consists of the complete support of node a, which is comprised of two elements of dimension h, and a single pair bond of length R. The nodal shape function for this case is given by

$$N^{(a)}(X) = \begin{cases} 1 - \frac{X^{(a)} - X}{h} & X \in \widetilde{\Omega}_0, \\ 1 + \frac{X^{(a)} - X}{h} & X \in \overline{\Omega}_0, \\ 0 & \text{elsewhere.} \end{cases} \tag{10}$$

From (8), we find the optimal bond density must satisfy

$$\int_{\widetilde{\Omega}_0} \rho dX = h \left(1 - N^{(a)}(X^{(\beta)}) \right) \tag{11}$$

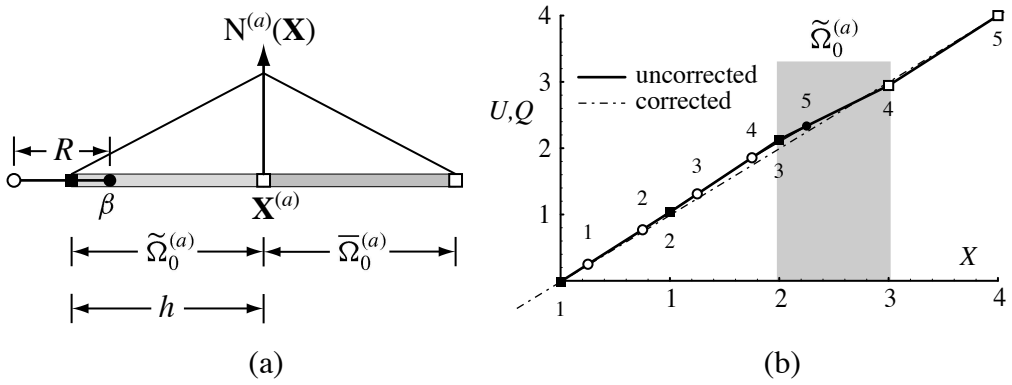


Figure 2: (a) Patch from a 1-dimensional coupled system. (b) Displacements of the 1-D coupled system for both the corrected and uncorrected bond density. Atom and node numbers are shown.

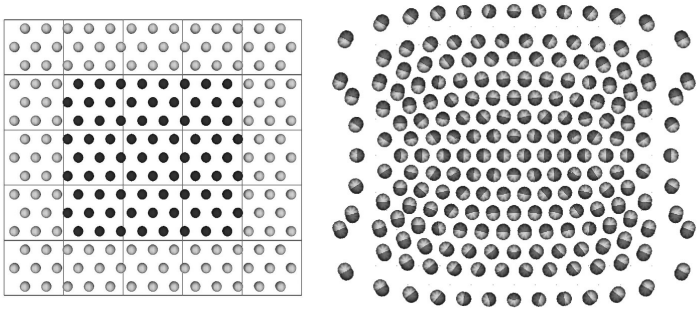
for $X^{(a)} - h \leq X^{(\beta)} \leq X^{(a)} - h + R$, which holds for $\rho = 1 - N^{(a)}(X^{(\beta)})$ over $\widetilde{\Omega}_0$. For this example, we see that the bond density simply compensates for the geometric overlap in the continuum and the underlying lattice. Figure 2(b) shows the displacements of a coupled system composed of five nodes and five atoms. The chain of atoms is bound by the quadratic potential $\varphi(r) = \frac{1}{2}k\,(r - R)^2$ acting between nearest neighbors with $R = \frac{1}{2}$. From the Cauchy-Born rule, the elements, with dimension $h = 1$, have an initial modulus $E = Rk$. In addition to the corrected solution that satisfies homogeneous deformation, $\rho(X) = \frac{3}{4}$ for $2 \leq X \leq 3$, Figure 2(b) also displays the solution obtained if we prescribe $\rho(X) = 1$ for the overlap element 3—4, *i.e.* no correction to the Cauchy-Born rule. This prescription produces inhomogeneous deformation and an increased stiffness in the overlap region.

5.2 Two-dimensional example

We have performed similar simulations for 2-dimensional systems. Figure 3(a) shows a system composed of a triangular lattice with free surfaces overlapping with a square FE mesh. This geometry is representative of the cross-section of a nanowire. For the coupled system, the atoms that lie within the outer layer of elements are free atoms while all other elements contain ghost atoms. When modeled with a 5^{th} nearest neighbor Lennard-Jones interaction, the system relaxes outward as shown in Figure 3(b). The coupled system (dark grey atoms) can be directly compared with a system simulated purely with atomistics, (light grey atoms). Agreement is very good, but not perfect due to the severe inhomogenous deformation at the corners and the range of the potential as compared with few number of real atom layers used in the coupled system.

6 CONCLUSION

We have presented the formulation for an atomistic-to-continuum coupling method for quasistatic analysis. Our approach is comprised of three components: kinematics, coupled equilibrium equations and a corrected Cauchy-Born rule. We have presented 1- and 2-dimensional examples that demonstrate the accuracy of the approach. We will apply this approach to 3-dimensional examples



(a) (b)

Figure 3: (a) A 2-D, triangular lattice with free surfaces composed of free (light grey) and ghost (dark grey) atoms. (b) The relaxed configuraton of (a) for the coupled system (dark grey) and a system treated purely with atomistics (light grey). Displacements are magnified by a factor of 200.

of nanowire and nanofilm relaxation and deformation, for which our generalized method is ideally applicable.

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