Atomistic submodel implementation and application within microstructure analysis by molecular dynamics

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ABSTRACT

Classical continuum mechanics faces substantial difficulties for adequately describe stress and strain distributions around microstructural material discontinuities such as crystallographic defects, voids, and grain boundaries. One way to improve the microstructural model is the development of the atomistic submodel that provides a minimal increase in the amount of atomistic data, but provides more accurate stress predictions without time consuming calculation of full atomistic model. In the proposed approach continuum model, discretized by finite elements, provides a displacement field for atoms on the edge of the simulation cell of the the atomistic submodel driven by molecular dynamics. The final result is the utilization of the best from both worlds, calculation speed of the constinuum mechanics using finite elements method and informing it with relevant material properties inferred from atomistic simulations by using molecular dynamics where it is the necessary.

1 INTRODUCTION

One way to improve the continuum mechanics (CM) material model is the development of the atomistic, *ab initio*, atomistic submodel (AS) for atomic dynamics that underline material strength under the temperature and stress loading. Molecular dynamics (MD) is a prominent theoretical tool of choice used to investigate responses of different materials at the nanoscale. With the advent of access to the high-speed computer throughout recent decades, there has been a substantial advance in the atomistic simulation applications. However, as soon as one tries to tackle with a vast quantity of information that arises at that scale, even tiny macro scale piece of real material, usually force us to use multiscale (MS) approach. Coupling of the two models poses significant challenges as described in [1], [2]. In the essence difficulty reconciling the CM and MD approach is caused by the very different nature of the underlining theories. Discrete chunky nature of atomistic reality features phenomena that are very hard to connect with some CM features. For example, the crack tip propagation is a process that simultaneously involves many different scales at ones. This fact of the crack propagation brings us to the high interest in the development of MS models tuned up for a specific purpose. Many examples of a MS approach aiming to couple atomistic and continuum models have been proposed, among many, in the following reviews [2-6].

One route to go around that problem is a combination of the standard finite elements method (FEM) techniques and the MD approach. Microstructural objects like defects, micro porosities, impurities, or misorientation in crystalline systems very often cause long-range effects, far beyond immediate atomistic neighborhood where they are [7]. From the physical point of view, we also do not need an

immense quantity of information that is provided by MD simulation. Important types of nanoscale defects in material typically involve many atoms and are identified as critical individual objects that describe material properties. Atomistic to CM models coupling methods are used to mutually exchange of information between atomistic and continuum systems. If this coupling is a successful one can describe nanosized effects in great detail within the unified model.

2 MULTISCALE MODELING

2.1 Molecular dynamics

Modeling of materials by MD starts with a description of the atom dynamics within MD where they are material points without internal degrees of freedom. By this approach, we are left with a system of multiple point particles which position and velocities can be calculated using Newton's laws of motion. However apparent simplicity of general idea hides complexities and nonlinearity of the coupled partial differential equations that are second-order and nonlinear. Also of the complexity of the system of equations are even further scaled up with the sheer number of atoms and dimensionality of the problem. Therefore the system is far from trivial to solve and even more difficult to accurately interpret from the macroscopic point of view. The central question is how to reconcile immense complexity and data redundancy given by atomistic degrees of freedom with standard CM physical quantities. For a recent review, see an in-depth discussion of the subject provided by Admal and Tadmor [8].

For atomistic simulation, we use field *de facto* standard software suite known as LAMMPS (stands for **L**arge-scale **A**tomic/**M**olecular **M**assively **P**arallel **S**imulator) as described in [9]. LAMMPS is a classical MD simulation code that is capable to effectively solve the system with millions of Newtonian equations for different macro phases of matter including solid material. To be predictive, it needs information about particles interactions and external forces. A significant effort of the community in recent decades resulted in many realistic potentials that can be used in modeling tasks. One of the prominent projects along those lines that we use here is OpenKIM framework [10-11] that gives us reproducible and reliable realistic interaction potential input for atomistic simulation.

2.2 Atomistic to continuum coupling

MS modeling incorporates forming of a material model that spans over several orders of magnitude in the time and length scale. Usually, MS methods are used to connect the fine scale of atoms and more coarse scale of CM. By this coupling, one can, in the same model, study nanostructure in the localized region of particular interest where dynamics of an individual atom is relevant and use computation less expensive CM in the area, where deformation is more homogenous and smooth. Usualy the atomistic model described by using MD is surrounded by finite element (FE) mesh. Model space is therefore divided into two parts: one dominated by discrete atomistic features like chemical bonding and the rest of the area where computationally less demanding CM approach is applied. Depending on the type of transitions between two regions, different computational strategies are developed. Frequently used methods are *Quasicontinuum* (QC) method developed by Tadmor, Ortiz, and Phillips [12] and *Bridging Domain* (BD) developed by Belytschko and Xiao [13]. In QC coupling of the atomistic and continuum model is achieved by refining FE mesh in the area of interest until one node corresponds to one atom. In BD to achieve a smooth transition from atomistic into the CM so-called handshake region is introduced, where atomistic and continuum models are simultaneously present.

The first problem to be solved is the identification of the parts of the model space where MD calculation is to be applied. This way, atomistic submodel can used only around the areas where

continuum models are about to fail. Significant problem visible in such a composite model is the correspondence of the continuum and atomistic quantities like stress, strain and displacement. In our approach, we have chosen a path inspired by already developed MS methods and couple the AS by imposing displacement boundary conditions. Simulations run separately on different scales and by using FEM we have implied that one FE contains many atoms. This is similar to QC and that the displacement of atoms follows the movement of FE edges (in accordance with Cauchy-Born rule [14]). Since the goal is to get the entire stress field, there has been no necessity to impose any kind of model overlapping since beyond the stress concentration area there is an agreement of stress field of FEM and MD.

2.3 Atomistic stress analysis

Here we briefly outline the procedure for atomistic stress calculation. For the AS we go along the same lines as in [7] for microscopic stress calculation, which can be broken down into four steps:

1) define a representative set of points in space,
2) choose one point and identify all atoms within the averaging radius r_{ave} around that point,
3) add up all stress contributions per chosen atoms (equation (1)),
4) divide the obtained cumulative stress with the total volume (area in our 2D case) summing the volumes per identified atoms (equation (2)).

For calculating stress per atom we use standard expression available in LAMMPS:

$$\tilde{\sigma}_{ij} = -\left[m v_i v_j + \frac{1}{2}\sum_{\kappa=1}^{N_p}\left(r_i^{\kappa,1} f_j^{\kappa,1} + r_i^{\kappa,2} f_j^{\kappa,2} \right) + \frac{1}{3}\sum_{\zeta=1}^{N_q}\left(r_i^{\zeta,1} f_j^{\zeta,1} + r_i^{\zeta,2} f_j^{\zeta,2} + r_i^{\zeta,3} f_j^{\zeta,3} \right) + \ldots \right] \tag{1}$$

where interaction potential takes care of atoms present within the defined cutoff range that contributes to the stress.

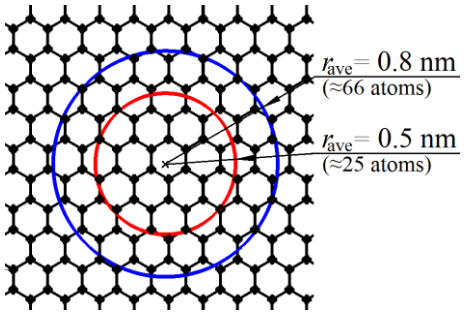

Figure 1, Determination of the averaging radius (all atoms within the circle are taken into account for the stress averaging procedure) [7]

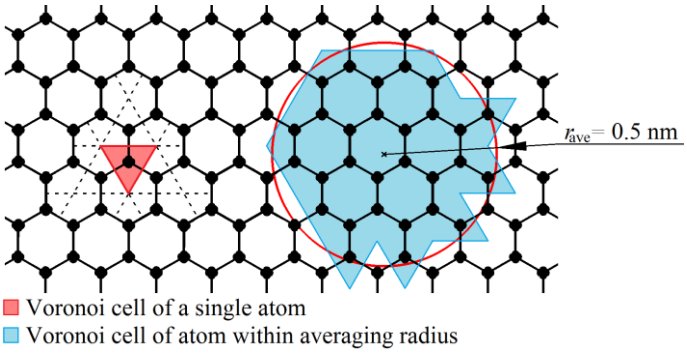

Figure 2, Representation of the Voronoi tessellation used for volume per atom determination [7]

Once the stress for each atom in a given region is calculated, we obtain spatially averaged stress for this region using the standard expression for N atoms:

$$\boldsymbol{\sigma}(r) = \frac{\sum_{\alpha=1}^{N} \tilde{\boldsymbol{\sigma}}^{\alpha}}{\sum_{\alpha=1}^{N} \Omega^{\alpha}} \tag{2}$$

N is the number of particles within the area we examine, while Ω^{α} is volume associated with the particular atom obtained using Voronoi tessellation. The process is shown in Figure 1 and Figure 2.

In addition to spatial stress also averaging include time averaging by conducting stress averaging procedure on snapshots of atomistic configuration every 1 ps, so local thermal excitement of atoms is washed out which result in the correct stress field. Time averaging is a simple mean value of averaged stress for the same point in space. In both cases (whole model and AS) configuration snapshots are taken after the system thermalizes for at least 10 ps.

3 MODELS

3.1 1D system

For the demonstration purposes, we outline the main idea in the case of a 1D system, as shown in Figure 3. To be as simple as possible but keep the main features, we analyze the standard mechanical problem of truss fixed at one end and loaded with some force on the other one. When translated into the atomistic picture, we get a chain of atoms connected with nonlinear elastic springs. As is well known realistic atomistic interaction like here, Lennard-Jones contains nonlocal contributions coming from distant atoms. Despite rapid decay of the force magnitude with distance and screening effects, this introduces a noticeable difference in calculated response to the external loading.

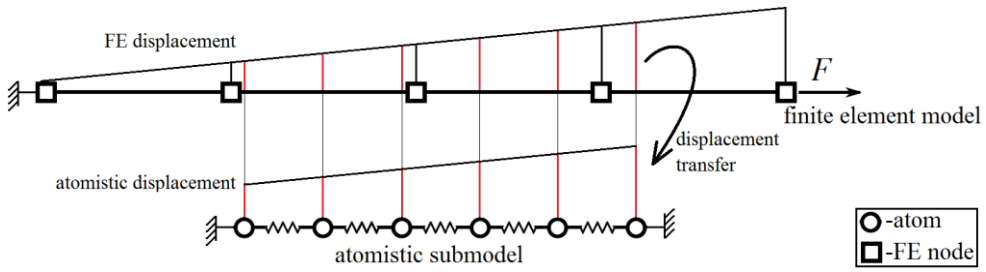

Figure 3, Atomistic submodel within full model space

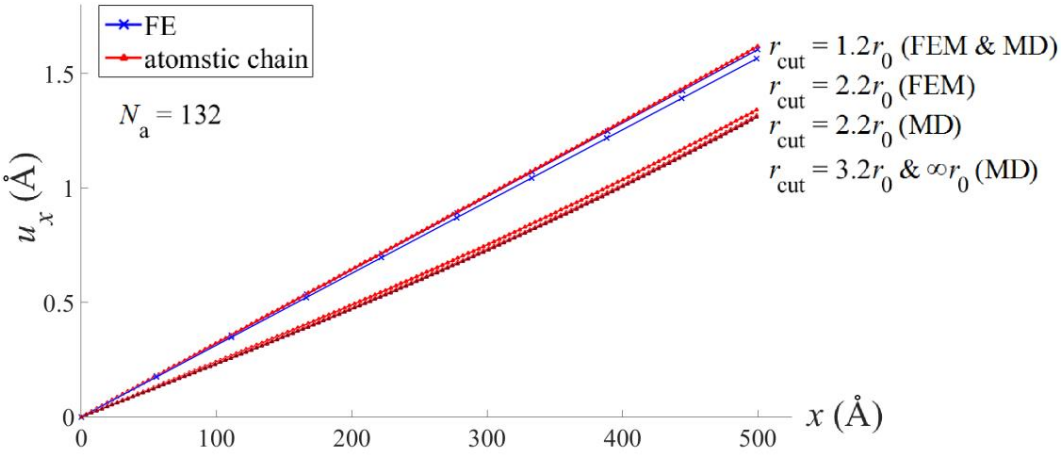

Figure 4, Comparison of atomistic chain analysis by FEM and MD for different cutoff radii

In Figure 4 we show the main results in the 1D case with the dependency of the displacement from the equilibrium position and coordinate along the full chain. The main message to be drawn from this is that we see a noticeable difference between MD and CM results. This difference gives a clear indication of how nonlinear and long-range interaction contribution influence results even in elastic regime. Namely, for CM and MD results to agree it is not enough to stay in elastic regime during loading but also cut off radii r_{cut} has to be small including just nearest-neighbor interatomic interactions. So this result demonstrates clearly that physical modeling in regions of high-stress

concentration even in topologically trivial 1D case calls for caution in the proper account for relevant physical quantities.

3.2 2D system

Our strategy following these results from the 1D model is to devise a combined model taking the CM results far from the hole and impose deformation as boundary conditions to the AS. This limited region is identified based on atomistic to CM stress ratio as criteria following the prescription given in [7]. Basic configuration in a 2D system that is the main objective of the present paper is shown in Figure 5. One of the results that we have obtained is that it is unimportant what is the shape of the submodel region as long as we are far away from the high values of stress gradients. We have chosen the square because of computational simplicity. According to the external loading, we first calculate deformation within the CM framework by standard FE discretization in the system.

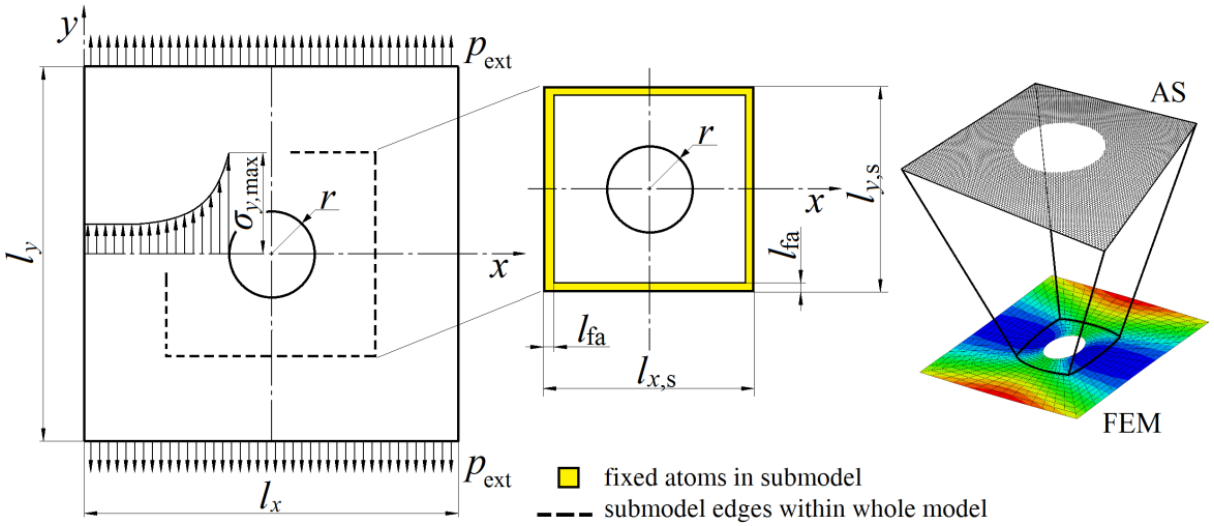

Figure 5, The geometry of the 2D sheet with the position of a circular hole

Table 1, Model parameters follow labeling symbols in Figure 5

	full model	submodel
sheet size (before loading)	l_x = 168.4 nm l_y = 169.9 nm	$l_{x,s}$ = 39,86 nm $l_{y,s}$ = 40,91 nm
the thickness of fixed atoms in the atomistic submodel	/	l_{fa} = 2 nm
hole radius	r = 9 nm	
imposed deformation	ε_y = 0.025	/
2D elasticity modulus	3300 GPa/Å	/
Poisson ratio	0.16	/
temperature	T = 300 K	
initial thermalization time	t_{therm} = 10 ps	
stretching time	t_{load} = 100 ps	/
equilibration time	t_{eq} = 10 ps	t_{eq} = 30 ps
number of atoms	1,116,371	53,611
averaging radius	r_{ave} = 2 nm	
the average number of atoms within an average radius	$N_{ave} \approx 473$	
number of time frames for averaging	10	
the span between time frames	1 ps	
calculation time	660.42 h	5.17 h

In the region depicted in Figure 5 by yellow color (narrow square strip) we impose deformation on atoms as obtained from FE calculation. Using MD simulation, we let the subsystem equilibrate and calculate stress distribution using equations (1) and (2). For relevant simulation parameters, see Table 1. For atomistic simulation, we have used AIREBO (**A**daptive **I**ntermolecular **R**eactive **E**mpirical **B**ond **O**rder potential) potential, developed by Stuart et al. [15], from OpenKIM repository for carbon giving us account for graphene sheet with a topology defined in Figure 5. AIREBO potential is an upgrade to REBO (**R**eactive **E**mpirical **B**ond **O**rder potential) initial developed by Brenner et al. [16]. Using such a realistic description of interaction gives us the confidence to faithfully reproduce experimentally relevant elastic properties and strength, as presented in [17]. In the case of full atomistic model calculations are conducted using thermostating procedure in the isobaric-isothermal (NpT) ensemble at a zero pressure and temperature of 300 K (provided by LAMMPS thermostating facility) with periodic boundary conditions were used along x and y axsis with imposed deformation in with imposed deformation in y direction.

First, we solve the CM problem using standard FEM procedures for linear static analysis and elements are first order four nodes 2D FEs. After FEM analysis, deformation is imposed faraway from hole (yellow square edge of the AS) to the atomistic system. Boundary conditions for AS are fixed edge atoms. Bulk atoms are subjected to the Canonical ensemble and left to thermalize for 30 ps.

4 RESULTS

Figure 6 is the result of the AS output. Stress averaging has been performed spatially using the averaging radius r_{ave} of 2 nm. Since boundary atoms of the AS are fixed and unaffected by relaxation, their contribution to the stress distribution is not affected by thermalization of the system and therefore should not be taken into account. The results of stress averaging on the AS are shown in Figure 6 with leaving out the parts of the model space with unmovable atoms.

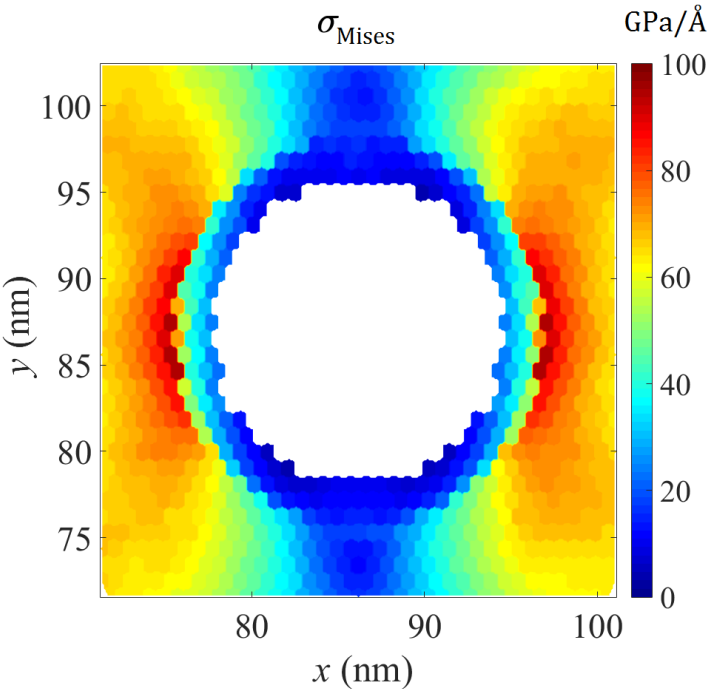

Figure 6, Spatially averaged atomistic stress distribution within AS

The stress distribution calculated using the atomistic model by the spatial and time averaging procedure is used to inform the full model in the concurrent region. Figure 7 shows stress distribution

through the model space with CM stress far away from the hole, and atomistic averaged stress around the opening. To visualize the final results more clearly in Figure 7, we show a series of the stress distribution results from the CM model on the left figure and averaged atomistic on the right one. In the Figure 7 right, we have inserted instead of CM results values obtained from AS. Connecting criteria is that the displacement of the atoms and CM media agree in the joint region as inspired by the Cauchy-Born approximation. This way, we have obtained a composite multiscale solution which combines the calculation speed of CM with accuracy and realistic properties of MD. With the natural implementation of boundary effects around the hole and with a substantial reduction of computer processing time.

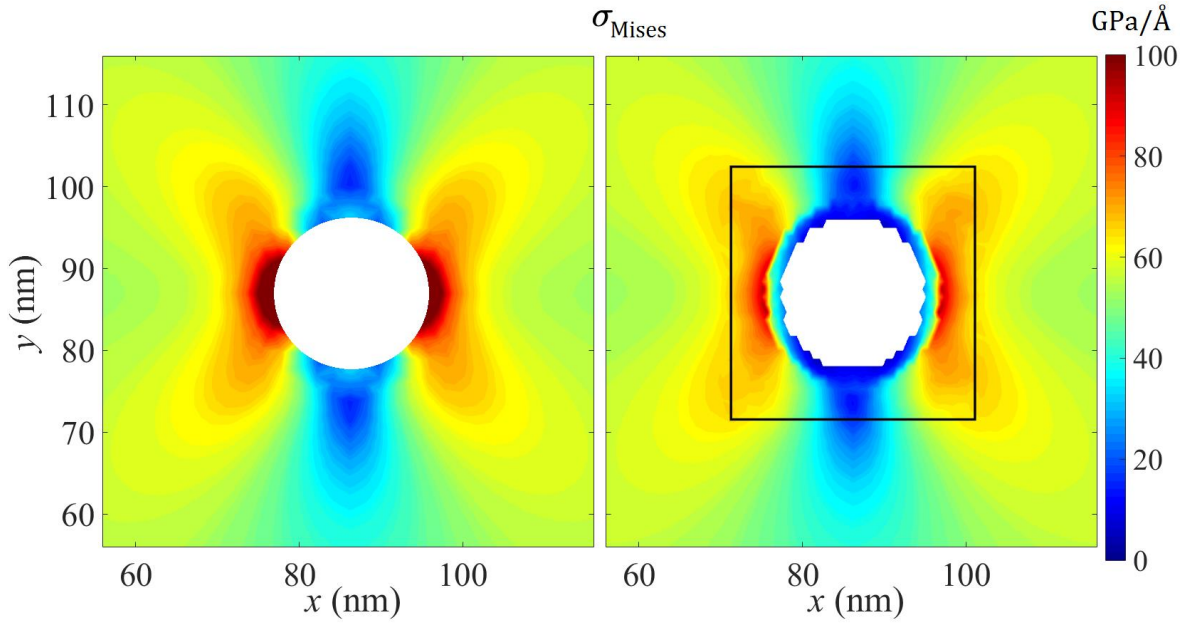

Figure 7, Stress distribution around the hole as seen from the full model only by FEM (left) and with submodel correction (right),

5 CONCLUSION

In this paper, we report on the simple yet effective way to unite CM and atomistic approach for the case of 1D and 2D system under external loading. First, we have shown a simple 1D academic example that examined key differences between atomistic and CM models and possible difficulties we might face in connecting the two approaches. In the second part, we show how we can effectively deal with the 2D problem of a graphene sheet with a circular hole under the uniaxial tension. We present an effective procedure of how one can use atomistic MD simulation in regions where the CM model is about to fail without proper account for near the edge stress distribution. Precise calculations of the stress distribution were atomistic discrete nature of matter plays a decisive role is of great importance with nanosized structures similar to the ones presented here. We have also outlined the main difficulties in developing proper physical MS connections of the CM system with the AS.

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