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#### A Multiscale Micromophic Molecular Dynamics: Theory and Applications

by

Qi Tong

A dissertation submitted in partial satisfaction of the

requirements for the degree of

Doctor of Philosophy

in

Engineering–Civil and Environmental Engineering

in the

Graduate Division

of the

University of California, Berkeley

Committee in charge:

Professor Shaofan Li, Chair Professor Khalid Mosalam Professor Mark Asta

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### A Multiscale Micromophic Molecular Dynamics: Theory and Applications

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#### Abstract

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by

Qi Tong

#### Doctor of Philosophy in Engineering–Civil and Environmental Engineering

University of California, Berkeley

Professor Shaofan Li, Chair

Multiscale simulation is a long standing dream in computational physics and materials. The motivation is natural: each single-scale model has its deficiencies. For example, microscale models such as Molecular Dynamics are limited to size in space and time; macroscale models such as Finite Element Method find difficulty recovering some fundamental physical phenomena such as materials defects. Simulations across scales are challenging because quantities in different scales have distinct properties. Mechanism needs to be harnessed to translate the information. Cross-scale communication is a typical two-way message passing: bottom-up and top-down. Bottom-up approach is relatively straightforward, where statistical theory or homogenization is used to collect lower-scale information and interpret it in higher levels. On the other hand, top-down approach requires physical insights. Specifically, in a mechanical system, top-down message passing can be the response of the molecular system when macroscale boundary conditions such as distributed load are enforced.

In this work, we reveal an intrinsic multiscale structure in solid materials. A "supercell" is introduced as a cluster of particles. Compare with "material point" in continuum mechanics, the "supercell" has internal degrees of freedom, which makes it equivalent to molecular systems. By introducing different force fields, we derive the dynamical equations for the different scales in the structure. The systematic multiscale framework solves the issue of top-down message passing by including quantities from different scales and connecting them in a uniform dynamical framework. We discuss the technical aspects in implementing the theory, i.e. constraints of the variables, integrators and temperature control. Numerical example of phase transition are presented to validate the theory, including bulk Nickel lattice under displacement and traction boundary conditions and Nickel nanowire with traction.

Furthermore, based on the developed multiscale theory, we establish a computational model to achieve efficiency in realistic multiscale simulations. The model includes three parts: atomistic region, macro region and transition zone. Atomistic region is where physical details are desired and is simulated by Molecular Dynamics. Macro region only concerns macroscale deformable behaviors of solid materials, which can be calculated by various models depending on the problem of interests. We choose state-based peridynamics in this work as a demonstration. The essential part is the transition zone which is responsible for translating messages across different domains. The "supercell" developed in the previous theory is employed as a transition element to carry those different messages. With solid theoretic foundation, the cross-scale message translation is clearly characterized. We also construct a filter to solve the issue of high-frequency wave reflection. Examples of 1-D and 2-D wave propagations are presented to demonstrate the procedure of cross-scale transition and the effect of the filter. In memory of my father

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## Chapter 1

## Introduction

### **1.1** Motivation and challenges

Computer technology has been transforming almost all aspects of our lives. It is reshaping the way we think and we do things. Thank to the great invention, people are free from tedious and repeated works to increase productivity. In the fields of science and engineering, the advent of ever more powerful computers greatly facilitates the process of discovery and design. Traditional experiment-oriented scientific and engineering activities are usually expensive and sometimes extremely challenging, e.g. experiments in nanoscale structures of materials or large-scale atmospheric circulation. On the contrary, computer simulation is fast, cheaper, more efficient, informative, and flexible, which greatly expands the frontier of many disciplines including materials science, biology, chemistry, etc. Therefore, more and more scientific implementations are switching from experiments to computational simulations. The computer-aided simulation and design is realized due to the development and continuous update of different numerical methods. Among them, Finite Element Method(FEM)[2, 3, 4, 5, 6] and Finite Difference Methods(FDM)[7, 8] are proved to be successful in solving partial differential equations (PDE) of continuum-field models, such as problems in solid mechanics, fluid mechanics, electromagnetic field, etc. They are becoming indispensable and prevalent in modern industry.

However, PDE solvers find their limitations such as numerical singularity when handling problems with discontinuity and local concentration. Specifically, materials defects and flaws ranging from dislocation to crack lead to highly localized regions with large deformation and high stress concentration. Those defects strongly influence the macroscopic mechanical performance of the material, which is one of the major concerns in engineering. Calculating everything from beginning, i.e. by Molecular Dynamics(MD)[9, 10, 11, 12, 13], or, more fundamentally, ab initial[14, 15, 16, 17, 18], is a way to reproduce physical phenomena. However, by common sense, those tools are restricted to the scope of small scales in space and time. Today, the state-of-the-art exascale supercomputers are capable of handling a molecular system up to sub-millimeter with 110 billion  $(1.1 \times 10^{11})$  atoms [7]. However, the sizes of realistic engineering models are still far beyond their reach, not to mention the capacity of affordable common purpose computers. Such a size limit impedes the application of fundamental molecular simulation in real engineering. On the other hand, molecular simulation itself is painful for analyzing and extracting useful information out of large amount of atoms.

The limits in both categories of macroscopic field simulations(FEM,FDM, etc) and microscopic simulations(MD,ab initial,etc) motivated the long-standing dream of multiscale simulation. The strategy is to solve the problem with methods in different paradigms and operate them at different scales. With the help of coupling techniques, communications between scales are established. The possibility of such an operation is based on the physical nature of materials which manifests itself in multiscales of space and time. Materials defects and flaws as mentioned above are typical phenomena displaying multiscale nature. For exampe, microscopic dislocation largely determines the macroscopic strength of the material; highly localized region around the crack tip is characterized as bond breaking and strong discontinuity, but the region away from the crack tip may have only moderate deformation. Therefore, operating the model in different scales separately is feasible once suitable techniques are obtained to set up the communication.

Technically, cross-scale communication is one of the most challenging problems in computational physics and materials, since quantities in different scales have distinct properties. For example, force in microscale is in terms of point-wise interaction between two particles, while in macroscale, it has a continuous distributed form. Temperature and random velocity of particles are counterparts in macroscopic and microscopic scales, respectively. Therefore, an ideal multiscale method must have physically insightful messenger to translate different information. We briefly summarize and compare the basic quantities in different scales, which are presented in table1.1.

Quantities	Microscale properties	Macroscale properties
Thermodynamics	Random velocities	Temperature
Forces	Point-wise particle interaction	Continuum distributed load
Displacement	Particle displacements	Average displacements
Velocity	High and low frequencies	Only low frequency

Table 1.1: Comparison of quantities in macroscopic and microscopic scales

A complete cross-scale communication has two-direction information passage: bottom-up and top-down, as shown in Fig.1.1. The bottom-up approach is relatively straightforward and well studied, where information from microscale is properly collected and interpreted to describe macroscale phenomena. For example, macroscale displacement field is averaged or homogenized from atomistic displacements; stress and temperature can be calculated from atomistic forces and random velocities according to statistical mechanics[19, 20, 21, 22, 23].



velocities, etc.

temperature, etc.

Figure 1.1: Two directions of exchanges of information. Basic quantities in microscale are atomistic positions, velocities, pairwise forces, etc. In macroscale, quantities include stress, deformation, temperature, etc.

On the other hand, the top-down message-passing approach is more challenging and requires physical insight. Specifically, top-down approach may be illustrated as the response of a molecular system when macroscale boundary condition such as traction is enforced. This procedure is not trivial, because a single-scale model cannot describe both molecular system and macroscale boundary conditions. Macroscopic calculation usually employs empirical assumptions such as continuum deformation and constitutive law rather than deriving them from first principle. Those assumptions simplify the model and provide intuitive interpretation, which are adopted to solve problems in macroscale without connection to lower scales. As a matter of fact, the attempts to directly incorporate macroscale theories into atomistic scales would bring in unphysical results due to those artificial assumptions.

Motivated by above mentioned challenges in multiscale simulation, this work is aimed at solving multiscale problems in two stages. First stage, we construct a physical model with multiscale framework to build the connection between scales. For cross-scale communications, we focus on the top-down approach, e.g. characterize the motion of molecular systems when macroscale boundary conditions are enforced. We categorize microscale boundary conditions such as pointwise force or atomic displacement as "strong condition", and macroscale boundary conditions such as continuous distributed load as "weak condition". Apparently, "weak condition" can not be achieved by using molecular dynamics alone. Therefore, the multiscale framework incorporating macroscale quantities into molecular systems without empirical assumptions is necessary. Second stage, we further establish a computational paradigm based on the previous physical model. In this model, a body is separated into macroscale and microscale regions. By taking advantage of the multiscale framework, an adaptive multiscale element is introduced as a messenger to translate information between regions. We apply the model to solve realistic engineering problems, as the purpose of any multiscale model. But the clear top-down characterization makes the model more natural, smooth and theoretically solid to pass information and fill the gap between scales. Overview of the work is presented in section 1.3.

### 1.2 Background

Multiscale models can be categorized to two groups: hierarchical or concurrent. Hierarchical approach attempts to obtain information from lower scales and directly apply it to higher scales. It is a one direction message-passing procedure without interactive communication or feedback between different scales. Molecular Dynamics is essentially a hierarchical model, where interatomic potentials can be obtained from underlying quantum mechanical calculation such as the density functional theory. Similarly, in various coarse grain models[24, 25, 26], it is often the case that a subscale cluster is subjected to atomistic simulations to determine the nature of the intramolecular potentials. In concurrent methodology, on the other hand, different scales are strongly coupled and solved simultaneously. The interscale dependence is complex, and frequent feedback is desired to pass information between scales. The concurrent multiscale model usually does not require a priori knowledge of the macroscale physical variables, which is more suitable for solving complicated cross-scale problems such as inhomogeneous deformation and material defects. In this section, we briefly review several typical concurrent multiscale models, to identify their key features, find their connections and explain the things to be improved.

#### Quasicontinuum method

The quasicontinuum method[27, 28, 29, 30, 31, 32] was originally developed by Ellad B. Tadmor in his Ph.D.'s research at Brown university between 1992 and 1996 supervised by Michael Ortiz. Subsequently, several researchers made the contribution of enriching the method. The idea is to choose representative atoms instead of full atomistic calculation. The total energy of the system is based on the representative atoms with appropriate weighting and interpolation. Empirical atomistic potentials are then employed compare with constitute relation in continuum mechanics.

Denote the atomistic position  $\mathbf{r}_i$  in referential configuration as

$$\mathbf{r}_i = x_i \mathbf{e}_x + y_i \mathbf{e}_y + z_i \mathbf{e}_z \tag{1.1}$$

where  $\mathbf{e}_x$ ,  $\mathbf{e}_y$  and  $\mathbf{e}_z$  are the basis vectors,  $x_i$ ,  $y_i$ , and  $z_i$  are coordinates. Total energy of the

#### CHAPTER 1. INTRODUCTION

system can be written as the sum of all atoms,

$$E^{tot}(\mathbf{r}) = \sum_{i=1}^{N} E_i(\mathbf{r}_i) = \sum_{i=1}^{N} E_i\{\mathbf{r}_1, \mathbf{r}_2, ..., \mathbf{r}_N\}$$
(1.2)

where  $E_i$  is the energy associated with the i-th atom, which depends on relative positions of other atoms and is calculated by interatomic potential. When the system is subject to external force field, the total energy becomes

$$\Phi(\mathbf{r}) = E^{tot}(\mathbf{r}) - \sum_{i=1}^{N} \mathbf{f}_i \mathbf{r}_i$$
(1.3)

The second term on the right hand side is the work done by external forces  $\mathbf{f}_i$ .

Eqs.(1.3) is the energy calculation of full lattices at zero temperature. The displacements of atoms can be obtained by minimize the total energy. Note that the goal is not to seek absolute energy minimum state of the lattice but the physically relevant metastable configurations. Since full atomistic calculation is expensive, quasicontinuum method provides an alternative to count only a subset of representative atoms. The density of representative atoms can be chosen to adapt the smoothness of the region through mesh refinement procedure in finite element method. For example, a first order accuracy scheme sets the constraint for displacements of atoms as

$$\mathbf{u}^{h}(\mathbf{r}_{i}) = \sum_{j=1}^{N_{rep}} S_{j}(\mathbf{r}_{i})\mathbf{u}_{j}$$
(1.4)

where  $S_j$  is the shape function of the finite element space, and  $N_{rep}$  is the number of representative atoms,  $N_{rep} \ll N$ . The original approach of energy summation is a local model based on linear interpolation, i.e. Cauchy-Born rule. This approach is called element-based summation rule. Define deformation gradient  $\mathbf{F}$  through  $\mathbf{r}_i = \mathbf{F} \cdot \mathbf{R}_i$ , where  $\mathbf{r}_i$  and  $\mathbf{R}_i$  are lattice vectors in current and referential configurations, respectively. Thus, the stain energy density is a single variable function of  $\mathbf{F}$ , which is written as

$$W_{CB}(\mathbf{F}) = E_0(\mathbf{F})/\Omega_0 \tag{1.5}$$

where  $E_0(\mathbf{F})$  and  $\Omega_0$  are energy and volume of a unit cell with deformation  $\mathbf{F}$ , and the energy summation is

$$E^{tot} = \sum_{e=1}^{N_{element}} \Omega_e W_{CB}(\mathbf{F}_e)$$
(1.6)

Another summation rule is based on clusters of atoms. Each cluster is a group of atoms whose state and motion depend on the associated representative atom. The relation can be obtained by interpolation. The energy density is

$$E_j^{cluster} = \frac{1}{N_j} \sum_k E_k \tag{1.7}$$

where  $E_k$  is the atomistic energy and  $N_j$  is the number of atoms in j-th cluster. The total energy is given by

$$E^{tot} = \sum_{j} w_j E_j^{cluster} \tag{1.8}$$

with  $w_j$  the weight function.

Quasicontinuum method provides a doable way to reduced the work amount caused by full atomistic calculation. It finds successful applications in materials defects such as dislocation of single crystal. Nonetheless, several drawbacks are to be noticed. First, only the potential energy is taken into account, and velocity of atoms is not allowed, thus all simulations are in zero temperature. Since the method is devised to directly seek statically stable configurations without characterizing dynamics, the procedure of dynamical evolution of the system is not described, which is sometimes of great interests of audiences. Second, due to the mismatch of local and nonlocal regions, the so called "ghost force" is induced on the interface, which will produce unphysical consequences of the simulation results.

#### Coupled atomistic and discrete dislocation

The coupled atomistic and discrete dislocation(CADD)[33, 34, 35, 36] was proposed by L. E. Shilkrot, R. E. Miller, and W. A. Curtin at Brown university in 2002. CADD is specially designed for simulating dislocation. The model consists three parts: an full atomistic region, a continuum region including dislocations modeled as elastic line defects, and a hand-shaking region coupling the above two regions. The scheme replaces full atomistic degrees of freedom to coarse scale continuum displacements and dislocation lines in continuum region. The work load is largely reduced without loss of much accuracy. Models in atomistic and continuum region are general molecular dynamics and dislocation dynamics. The essential part is a passing methodology to aid dislocations moving across the interface back and forth.

In the boundary value problem, the system is divided to an atomistic region  $\Omega_A$  and a continuum region  $\Omega_C$ , with  $\mathbf{T} = \mathbf{T}_0$  on traction boundary  $\partial \Omega_T$  and  $\mathbf{u} = \mathbf{u}_0$  on displacement boundary  $\partial \Omega_u$ . The interface is denoted by  $\partial \Omega_I$ . Standard dislocation dynamics separate the continuum problem into two complementary problems: (1) an infinite elastic continuum induced by dislocations which is denoted by  $\sim$ . The infinite elastic field generates traction  $\tilde{\mathbf{T}}$  on  $\partial \Omega_T$ , dispacements  $\tilde{\mathbf{u}}$  and  $\tilde{\mathbf{u}}_I$  on  $\partial \Omega_u$  and  $\partial \Omega_I$ , respectively. (2) The original continuum region without dislocation. But the boundary conditions are changed to  $\hat{\mathbf{T}} = \mathbf{T}_0 - \tilde{\mathbf{T}}$ ,  $\hat{\mathbf{u}} = \mathbf{u}_0 - \tilde{\mathbf{u}}$  and  $\hat{\mathbf{u}}_I = \mathbf{u}_I - \tilde{\mathbf{u}}_I$ . With the decomposition, the total energy of the system is

$$E^{tot} = \frac{1}{2} \int_{\Omega_C} (\tilde{\boldsymbol{\sigma}} + \hat{\boldsymbol{\sigma}}) : (\tilde{\boldsymbol{\varepsilon}} + \hat{\boldsymbol{\varepsilon}}) dV - \int_{\partial\Omega_C} \mathbf{T}_0 \cdot \mathbf{u} dA + E^{atom}(\mathbf{r}_i) - \sum_i \mathbf{f}_i \cdot \mathbf{u}_i$$
(1.9)

The evolution of the system is then a problem of energy minimization. The construction of passing methodology consists two key steps. First, assume atomistic positions are linearly approach the configuration of the continuum field, i.e. Cauchy-Born deformation. Second,

a detecting and passing scheme should be introduced to translate the dislocation between atomistic and continuum regions. In CADD model, a "detection band" is defined in the atomistic region near the interface. The "detection band" is consists of triangular elements between atoms sitting on different slip planes. If a dislocation passes one of the planes, a plastic slip strain  $\mathbf{E}^p$  is produced. Since all possible slip systems of any given crystal structure are known, the associated  $\mathbf{E}^p$  can be calculated. When a specific dislocation occurs, the associated  $\mathbf{E}^p$  passes to the continuum field. Reversely, if the plastic strain from continuum region activates a slip system, the associated dislocation nucleates and propagates. Such mechanism also ensures the interplay of different dislocations.

CADD is useful and effective in simulating dislocation by bringing in basic quantities of characterizing the phenomenon, e.g. slip plane, Burgers vector, etc. Since this model is specially designed for dislocation, the application is restricted in this category. A priori knowledge of dislocation theory and crystal structure is necessary to ensure the successful practice of the method. Moreover, special treatment for detection and passage is required from time to time during the simulation, which makes the method not adaptive.

### Bridging scale method

The bridging scale method[37, 38, 39, 40, 41], originally developed by Wagner and Liu in 2003, is a systematic multiscale theory combining molecular dynamics and finite element method. The idea is separating the displacement to coarse and fine components. FEM exists everywhere and MD only presents in a small atomistic region. Therefore, FEM and MD models overlap in the atomistic region. The coarse/fine displacement transition is realized by a process of projection with minimizing least square error. An impedance force is also introduced to mitigate high-frequency wave reflection.

The decomposition of displacement field  $\mathbf{u}(\mathbf{r})$  is

$$\mathbf{u}(\mathbf{r}) = \bar{\mathbf{u}}(\mathbf{r}) + \mathbf{u}'(\mathbf{r}) \tag{1.10}$$

where  $\bar{\mathbf{u}}(\mathbf{r})$  is the coarse component and  $\mathbf{u}'(\mathbf{r})$  is the fine component. The coarse scale displacement is represented by basis functions introduced in finite element method through interpolation, which is

$$\bar{\mathbf{u}}(\mathbf{r}) = \mathbf{N}\mathbf{d} \tag{1.11}$$

where  $\mathbf{N}$  is the basis or shape function,  $\mathbf{d}$  is the nodal displacement. The fine scale displacement is defined as the part of total displacement  $\mathbf{q}$  that is not shown in the coarse scale part, it is calculated as

$$\mathbf{u}'(\mathbf{r}) = \mathbf{q} - \mathbf{P}\mathbf{q} \tag{1.12}$$

where  $\mathbf{P}$  is an operator used to minimize the difference of some quantities between coarse scale and total scale at atomic sites. Total displacement is the sum of above two components which are functions of two independent variables: finite element nodal displacement  $\mathbf{d}$  and total displacement  $\mathbf{q}$ ,

$$\mathbf{u}(\mathbf{r}) = \mathbf{N}\mathbf{d} + \mathbf{q} - \mathbf{P}\mathbf{q} \tag{1.13}$$

The Lagrangian of the system is given by

$$L(\mathbf{u}, \dot{\mathbf{u}}) = \frac{1}{2} \dot{\mathbf{u}}^T \mathbf{M} \mathbf{u} - U(\mathbf{u}) + \mathbf{f}_{ext}^T \mathbf{u}$$
(1.14)

where **M** is the mass matrix and  $\mathbf{f}_{ext}$  is the external force applied on the system. The dynamical governing equations can be derived through the standard procedure as

$$\frac{d}{dt}\left(\frac{\partial L}{\partial \dot{\boldsymbol{d}}}\right) - \frac{\partial L}{\partial \mathbf{d}} = 0, \qquad \frac{d}{dt}\left(\frac{\partial L}{\partial \dot{\boldsymbol{q}}}\right) - \frac{\partial L}{\partial \mathbf{q}} = 0 \qquad (1.15)$$

which give rise to

$$\mathbf{M}\ddot{\boldsymbol{d}} = \mathbf{N}^{T}(\mathbf{f}_{int} + \mathbf{f}_{ext})$$
(1.16)

$$\mathbf{M}\ddot{\boldsymbol{q}} = \mathbf{f}_{int} + \mathbf{f}_{ext} \tag{1.17}$$

The internal force  $\mathbf{f}_{int}$  is obtained from MD calculation. The Eqs.(??) represent coarse and fine scales evolution of the dynamical system. Wave reflection on the interface can be observed if the the equations used directly without special treatment. To solve this issue, an impedance force is introduced as

$$\mathbf{f}_{imp}(t) = \int_0^t \theta(t-\tau)(\mathbf{q}(\tau) - \bar{\boldsymbol{u}}(\tau))d\tau + \mathbf{R}(t)$$
(1.18)

where  $\theta(t-\tau)$  is a time history kernal and  $\mathbf{R}(t)$  is a random force.

This bridging scale method makes messages transit between different regions in a seamless fasion. By introducing impedance force, the reflection of high frequency waves is largely mitigated. Another advantage is due to the coarse and fine scales separation, which makes it possible to employ different time steps in different scales in compensation of length scale separation. The dynamical system also allows finite-temperature simulation.

#### Other concurrent methods

Several other concurrent models found their success in practice. Among them, the macroscopic, atomistic, ab initio dynamics(MAAD)[42, 43, 44] method is one of the earliest works, which spans three scales from quantum mechanics to continuum mechanics. It has been applied to solve dynamical fracture problem of silicon. On the other hand, the issue of numerical wave reflection at interscale boundary can be observed in this method. Concurrent atomistic continuum method(CAC)[45, 46, 47, 48] is a recently developed field theory connecting atomistic models with local properties and balance equations through a statistical procedure. Finite element approach is adopted in the model with fine and coarse mesh to achieve multiscale calculation. Different from continuum field theories where empirical constitutive relations are introduced, this model is derived in a statistical approach. Thus, shrinking mesh to molecular resolution will not significantly increase error as continuum field theories do. Perfectly matched multiscale simulation method[49, 50] introduces the perfectly matched layer into the bridging scale model to further solve the issue of wave reflection. These are notable multiscale models among others.

### **1.3** Overview of the Dissertation

As introduced in section 1.1, this work includes two parts: a physical model with multiscale theory and a computational model with application. In the first part, we propose a physical model characterizing top-down approach of communication, namely, the response of the associated molecular system when macroscale boundary conditions are applied. To reach the goal of complete top-down characterization, we construct a multiscale structure where the information is totally based on lower scale without any higher-level assumption. While at the same time, all desired quantities in different scales are included in the model with clear physical connection. In the second part, we employ the established multiscale theory to further construct a computational model for realistic multiscale practice. The top-down message passing model, together with bottom-up approaches, e.g. statistical mechanics, makes the transition of information in the computational model smooth without physical barrier. Examples for demonstrating both physical and computational model are to be presented. We briefly introduce the content of each chapter as follow,

Chapter 1 is the introduction of the dissertation starting from the motivation and challenges of multiscale simulations. The motivation of mutiscale simulation is natural: single scale models has their intrinsic limitations, e.g. traditional FEM finds its difficulty in simulating materials defects, and MD is limited by the size in space and time. Therefore, multiscale models are created to overcome issues in both category. The main challenges in multiscale simulation are summarized. Subsequently, four concurrent multiscale models are reviewed, including quasicontinuum method, coupled atmoistic and discrete dislocation, bridging scale method and concurrent atomistic continuum method. We introduced general ideas and formulas of each model, and discussed the advantages and aspects to be improved.

In Chapter 2, we shall propose a systematic physical model with multisale structure and dynamics. Following brief reviews of continuum mechanics and Parrinello-Rahman molecular dynamics, we reveal the fundamental multiscale structure and kinematics for deformable solids from molecular systems to macroscale continuum mechanics. The information is gathered from and equivalent to the original molecular system but we observe them on the pespectives of continuum mechanics. As in continuum mechanics, we focus the study on the object of "material point", whose motion includes rigid body translation, rotation and deformation. However, the proposed multiscale model recovers fully the atomistic resolution.

In our model, "material point" is a cluster of atoms and has internal degrees of freedom in addition to other properties in continuum mechanics. Globally, the "material point" may not be continuously connected to each other as assumed in continuum mechanics. Consequently, the kinematics is no more restricted in each single scale as continuous deformation or atomistic motion. We then characterize the force fields for the multiscale system. Different forms of forces can be found when we take into account different scales. For example, forces in molecular system are in terms of interactions among atoms and molecules. But in macroscale, forces are continuously represented as surface traction or body force. We intentionally put them together in a same framework and connect them with associated potential energy. After that, we describe the motion for the multiscale system by introducing driving force for each scale and deriving dynamic equations. With the proposed multiscale structure and force field, we are able to put quantities of different scales in a systematic dynamical framework, and bring various macroscale boundary conditions into molecular systems.

In Chapter 3, technical aspects of the multiscale theory are discussed. Introducing multiscale structure into the molecular system brings in redundant degrees of freedom, i.e. variables associated with centers of mass and deformation gradient. Thus, constraints must be enforced to guarantee the equivalence between the multiscale system and the original molecular system. The necessary constraints are presented in section 3.1, followed by discussions of time integration, including the requirements of the integrators and the numerical schemes. We derive the formulas for temperature control in the last section.

In Chapter 4, we present three numerical examples to validate and demonstrate the multiscale theory. First example is a test of macroscale displacement boundary condition. A bulk crystal is subject to phase transition from original face-center-cubic structure to hexagonalclose-packed structure under compressive displacement loading. Second example is the same model with traction boundary condition. Third example is a nanowire going through phase transition under traction boundary condition. Comparison with theoretical prediction and discussions of the results can be found subsequently.

In Chapter 5, a multiscale computational model is proposed. The model includes an atomistic domain and a macroscopic domain with an interface. The advantage of this model is the outcome of the multiscale molecular dynamics presented through chapters  $2\sim4$ . We import the "supercell" from the multiscale theory as a transition element or "messenger" to translate information between different domains. We call the "supercell" as adaptive multiscale element, because the capability of carrying both microscale information such as atomic force and macroscale information such as traction. The cross-scale transition is smooth due to the physical nature of the adaptive multiscale element. Molecular Dynamics is selected for simulating atomistic domain. In macroscopic domain, several models can be adopted based on the problem of interest. In this work, as a demonstration, we introduce the statebased peridynamics, which is a non-local theory suitable for simulating discontinuities and materials defects. To mitigate the typical issue of wave reflection in multiscale simulation, a filter is constructed to smooth out high-frequency wave components. Numerical scheme for implementing the computational model is shown in the last section.

In Chapter 6, we shall do benchmark tests of 1-D and 2-D wave propagations. The tests

are set up for validating the computational model in two perspectives. First, demonstrating the capacity of the adaptive element in message translation between atomistic and macroscopic domains. It will be seen that either 1-D or 2-D wave propagate through the interface smoothly. Second, verifying the effectiveness of the filter. We shall apply initial waves with low and high frequency components intentionally, and monitor the procedure of filtering when the waves pass the interface.

Chapter 7 summarizes the whole work with discussions. Outlook of future study is also presented.

## Chapter 2

## Theory of Multiscale Micromophic Molecular Dynamics

### 2.1 Basic Concepts of Continuum Mechanics

In this section, we shall briefly introduce the essential concepts in Continuum Mechanics, which are mostly relevant to our work. Therefore, this is not aimed at comprehensively review the theory. Detailed mathematical foundations and interpretations can be found in large amount of textbooks, e.g. [51, 52, 53, 54, 55, 56, 57, 58]

#### **Kinematics**

#### **Deformation Gradient**

Let a body  $\mathcal{B}$  be a collection of infinite number of material points, which occupies a region in 3-D space  $\mathbb{R}^3$ . This region is referred as *referential configuration* of  $\mathcal{B}$ . In a Cartesian coordinated system with basis  $\{E_1, E_2, E_3\}$ , position of any material point  $X \in \mathcal{B}$  can be represented by

$$\boldsymbol{X} = X_1 \boldsymbol{E}_1 + X_2 \boldsymbol{E}_2 + X_3 \boldsymbol{E}_3 = X_A \boldsymbol{E}_A \tag{2.1}$$

A deformation of  $\mathcal{B}$  occurs when each material point moves to a new position  $\boldsymbol{x}$  with mapping  $\boldsymbol{\varphi} : \boldsymbol{X} \to \boldsymbol{x}$ , which is denoted by  $\boldsymbol{x} = \boldsymbol{\varphi}(\boldsymbol{X})$ . The new body  $\boldsymbol{\varphi}(\boldsymbol{\mathcal{B}})$  is called *current configuration uration*. Representations of quantities in *referential configuration* and *current configuration* are called *material* (or Lagrangian) and *spacial* (or Eulerian) descriptions, respectively.

We require the *deformation* be continuous without singularity such as gap or overlap, thus the map is one-to-one. We choose a new coordinate system  $\{e_1, e_2, e_3\}$  which may be totally unrelated with  $\{E_A\}$ , then  $x \in \varphi(\mathcal{B})$  is

$$\boldsymbol{x} = x_1 \boldsymbol{e}_1 + x_2 \boldsymbol{e}_2 + x_3 \boldsymbol{e}_3 = x_i \boldsymbol{e}_i \tag{2.2}$$

The *displacement* field is defined as

$$\boldsymbol{u} = \boldsymbol{x}(\boldsymbol{X}) - \boldsymbol{X} \tag{2.3}$$

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*Deformation gradient* is an essential concept in finite deformation kinematics which totally describes the deformation state of a material point. It is an operator mapping two configurations, which is defined as

$$\boldsymbol{F} := \frac{\partial \boldsymbol{x}}{\partial \boldsymbol{X}} = \frac{\partial x_i}{\partial X_A} \boldsymbol{e}_i \otimes \boldsymbol{E}_A = F_{iA} \boldsymbol{e}_i \otimes \boldsymbol{E}_A$$
(2.4)

Since Deformation gradient F spans two coordinate systems  $\{E_A\}$  and  $\{e_i\}$  as in (2.4), we call it two point tensor. F as an operator maps infinite small line vectors dX in referential configuration to the counterpart dx in current configuration. The mapping procedure is

$$d\boldsymbol{x} = \boldsymbol{F} \cdot d\boldsymbol{X} \tag{2.5}$$

Note that the deformation gradient is a field variable depending on position vector of particles, which is F = F(X).

In terms of displacement field  $\boldsymbol{u}$ , we can write  $\boldsymbol{F}$  as

$$\boldsymbol{F} = \boldsymbol{I} + \boldsymbol{H} \tag{2.6}$$

where  $H := \nabla_X u$  is the so called *displacement gradient*.

#### **Strain Measures**

#### • Right Cauchy-Green strain tensor

Consider the length of a line vector defined as  $ds := \parallel d\boldsymbol{x} \parallel$  and  $dS := \parallel d\boldsymbol{X} \parallel$  in current and referential configurations, respectively. The relation is obtained by

$$(ds)^{2} = \parallel d\boldsymbol{x} \parallel^{2} = d\boldsymbol{x} \cdot d\boldsymbol{x} = \boldsymbol{F} d\boldsymbol{X} \cdot \boldsymbol{F} d\boldsymbol{X} = d\boldsymbol{X} \cdot \boldsymbol{F}^{T} \boldsymbol{F} \cdot d\boldsymbol{X} = d\boldsymbol{X} \cdot \boldsymbol{C} \cdot d\boldsymbol{X}$$
(2.7)

where C is called *right Cauchy-Green strain tensor*, which is defined as  $C := F^T F$ . Denoting the unit vector N := dX / || dX || = dX / dS, we have

$$ds = \sqrt{\mathbf{N} \cdot \mathbf{C} \cdot \mathbf{N}} dS \tag{2.8}$$

In components,  $C = C_{AB}E_A \otimes E_B$ . Since C is in terms of referential coordinate system, we call it *material tensor*.

Let  $d\mathbf{X}$  and  $d\mathbf{Y}$  be two vectors at point  $\mathbf{X}$ . The angle  $\Theta$  between these two vectors are

$$\cos\Theta = \frac{d\boldsymbol{X} \cdot d\boldsymbol{Y}}{\| \, d\boldsymbol{X} \, \| \| \, d\boldsymbol{Y} \, \|} = \boldsymbol{N} \cdot \boldsymbol{M}$$
(2.9)

where unit vector  $M = dY / \parallel dY \parallel$ . The associated angle  $\theta$  in current configuration is calculated as

$$\cos\theta = \frac{d\boldsymbol{x} \cdot d\boldsymbol{y}}{\parallel d\boldsymbol{x} \parallel \parallel d\boldsymbol{y} \parallel} = \frac{d\boldsymbol{X} \cdot \boldsymbol{C} \cdot d\boldsymbol{Y}}{\sqrt{d\boldsymbol{X} \cdot \boldsymbol{C} \cdot d\boldsymbol{X}} \sqrt{d\boldsymbol{Y} \cdot \boldsymbol{C} \cdot d\boldsymbol{Y}}} = \frac{\boldsymbol{N} \cdot \boldsymbol{C} \cdot \boldsymbol{M}}{\sqrt{\boldsymbol{N} \cdot \boldsymbol{C} \cdot \boldsymbol{N}} \sqrt{\boldsymbol{M} \cdot \boldsymbol{C} \cdot \boldsymbol{M}}}$$
(2.10)

• Left Cauchy-Green strain tensor

On the other hand, dS is calculated as

$$(dS)^{2} = \parallel d\boldsymbol{x} \parallel^{2} = d\boldsymbol{X} \cdot d\boldsymbol{X} = \boldsymbol{F}^{-1} d\boldsymbol{x} \cdot \boldsymbol{F}^{-1} d\boldsymbol{x} = d\boldsymbol{x} \cdot \boldsymbol{F}^{-T} \boldsymbol{F}^{-1} \cdot d\boldsymbol{x} = d\boldsymbol{x} \cdot \boldsymbol{b}^{-1} \cdot d\boldsymbol{x} \quad (2.11)$$

where  $\boldsymbol{b} := \boldsymbol{F} \boldsymbol{F}^T$  is called *left Cauchy-Green strain tensor*. The component form is given by  $\boldsymbol{b} = b_{ij} \boldsymbol{e}_i \otimes \boldsymbol{e}_j$ , which is expressed in current configuration. We call it *spatial tensor*. With the previous defined relations, we may calculate

$$dS = \sqrt{\boldsymbol{n} \cdot \boldsymbol{b}^{-1} \cdot \boldsymbol{n}} ds \qquad (2.12)$$

and

$$\cos\Theta = \frac{\boldsymbol{n} \cdot \boldsymbol{b}^{-1} \cdot \boldsymbol{m}}{\sqrt{\boldsymbol{n} \cdot \boldsymbol{b}^{-1} \cdot \boldsymbol{n}} \sqrt{\boldsymbol{m} \cdot \boldsymbol{b}^{-1} \cdot \boldsymbol{m}}}$$
(2.13)

where  $\boldsymbol{n} = d\boldsymbol{x} / \parallel d\boldsymbol{x} \parallel$  and  $\boldsymbol{m} = d\boldsymbol{y} / \parallel d\boldsymbol{y} \parallel$ .

• Green-Lagrange strain tensor

Introduce the change of length relative to original length dS as

$$\frac{(ds)^2 - (dS)^2}{(dS)^2} = \mathbf{N} \cdot \mathbf{C} \cdot \mathbf{N} - \mathbf{N} \cdot \mathbf{N} = \mathbf{N} \cdot (\mathbf{C} - \mathbf{I}) \cdot \mathbf{N} = 2\mathbf{N} \cdot \mathbf{E} \cdot \mathbf{N}$$
(2.14)

The symmetric tensor E is called *Green-Lagrange strain tensor*, which is defined as

$$\boldsymbol{E} := \frac{1}{2} (\boldsymbol{C} - \boldsymbol{I}) = \frac{1}{2} (\boldsymbol{F}^T \boldsymbol{F} - \boldsymbol{I})$$
(2.15)

Write E in terms of displacement field

$$\boldsymbol{E} = \frac{1}{2} \left[ (\boldsymbol{H} + \boldsymbol{I})^T (\boldsymbol{H} + \boldsymbol{I}) - \boldsymbol{I} \right] = \frac{1}{2} \left[ \boldsymbol{H} + \boldsymbol{H}^T + \boldsymbol{H}^T \boldsymbol{H} \right]$$
(2.16)

Linearize  $\boldsymbol{E}$ , we obtain a infinitesimal strain tensor

$$\boldsymbol{\varepsilon} = \frac{1}{2} \left[ \boldsymbol{H} + \boldsymbol{H}^T \right] \tag{2.17}$$

• Green-Almansi strain tensor

Similarly, the change of length relative to current length ds is calculated as

$$\frac{(ds)^2 - (dS)^2}{(ds)^2} = \boldsymbol{n} \cdot \boldsymbol{n} - \boldsymbol{n} \cdot \boldsymbol{b}^{-1} \cdot \boldsymbol{n} = \boldsymbol{n} \cdot (\boldsymbol{I} - \boldsymbol{b}^{-1}) \cdot \boldsymbol{n} = 2\boldsymbol{n} \cdot \boldsymbol{e} \cdot \boldsymbol{n}$$
(2.18)

The so called *Green-Almansi strain tensor* is defined as

$$\boldsymbol{e} := \frac{1}{2} (\boldsymbol{I} - \boldsymbol{b}^{-1}) = \frac{1}{2} \left[ \boldsymbol{I} - (\boldsymbol{F} \boldsymbol{F}^{T})^{-1} \right]$$
(2.19)

In terms of displacement field

$$\boldsymbol{e} = \frac{1}{2} \left[ \boldsymbol{h} + \boldsymbol{h}^T - \boldsymbol{h}^T \boldsymbol{h} \right]$$
(2.20)

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Introducing linearization gives  $d\mathbf{x} \approx d\mathbf{X}$  and removes the nonlinear term in the above equation. Therefore, we recover the infinitesimal strain tensor  $\boldsymbol{\varepsilon}$ .

#### Stretch and Rotation

As we know, deformation gradient F(X) carries all information of strain at a material point X. In fact, we can extract from F(X) not only information of strain, but the overall deformation including a rigid body rotation. Consider following right polar decomposition

$$\boldsymbol{F} = \mathbf{R}\mathbf{U} \tag{2.21}$$

where  $\mathbf{R}$  is a rotation and  $\mathbf{U}$  is positive definite. We call  $\mathbf{U}$  right stretch tensor and  $\mathbf{R}$  rotation tensor. Furthermore,  $\mathbf{U} = \sqrt{\mathbf{F}^T \mathbf{F}} = \sqrt{\mathbf{C}}$ . Therefore, Eq.(2.8) can be written as

$$ds = \sqrt{\mathbf{N} \cdot \mathbf{C} \cdot \mathbf{N}} dS = \sqrt{\mathbf{N} \cdot \mathbf{U}^2 \cdot \mathbf{N}} dS = \sqrt{\mathbf{U} \mathbf{N} \cdot \mathbf{U} \mathbf{N}} dS = \| \mathbf{U} \mathbf{N} \| dS \qquad (2.22)$$

which means the stretch of X is given by || UN ||. Similarly, Eq.(2.10) can be written as

$$\cos\theta = \frac{\boldsymbol{U}\boldsymbol{N}\cdot\boldsymbol{U}\boldsymbol{M}}{\|\|\boldsymbol{U}\boldsymbol{N}\|\|\|\|\boldsymbol{U}\boldsymbol{M}\|}$$
(2.23)

which gives the angle as between UN and UM after deformation.

The right polar decomposition can be understood by first apply a stretch to a material point, and then apply a rotation.

Similarly, consider left polar decomposition

$$\boldsymbol{F} = \boldsymbol{V}\boldsymbol{R} \tag{2.24}$$

in which  $\boldsymbol{V} = \boldsymbol{R} \boldsymbol{U} \boldsymbol{R}^T = \sqrt{\boldsymbol{F} \boldsymbol{F}^T} = \sqrt{\boldsymbol{b}}$ . Eqs.(2.12) and (2.13) are then written as

$$dS = \parallel \boldsymbol{V}^{-1}\boldsymbol{n} \parallel ds \tag{2.25}$$

and

$$\cos\theta = \frac{\boldsymbol{V}^{-1}\boldsymbol{n}\cdot\boldsymbol{V}^{-1}\boldsymbol{m}}{\parallel\boldsymbol{V}^{-1}\boldsymbol{n}\parallel\parallel\boldsymbol{V}^{-1}\boldsymbol{m}\parallel}$$
(2.26)

Therefore, the left polar decomposition can be understood as a manipulation of rotation followed by a stretch.

#### Motion

Consider the motion of a material point

$$\boldsymbol{x} = \boldsymbol{\varphi}(\boldsymbol{X}, t) \tag{2.27}$$

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We know that X is fixed, thus position x is a single-parameter variable depending on time t. Define the *material velocity* as

$$\boldsymbol{V} = \boldsymbol{V}(\boldsymbol{X}, t) := \frac{d\boldsymbol{\varphi}}{dt} = \dot{\boldsymbol{\varphi}}$$
(2.28)

which is a material time derivative. The material velocity describes the velocity of a material point with time. It is a tangent vector of the trajectory.

On the other hand, we can define spatial time derivative as

$$\boldsymbol{v} = \boldsymbol{v}(\boldsymbol{x}, t) = \boldsymbol{V}(\boldsymbol{\varphi}^{-1}(\boldsymbol{x}, t), t) = \boldsymbol{V} \circ \boldsymbol{\varphi}^{-1}$$
(2.29)

The spatial velocity v has the same value as V but express as a function of x. Physically, it represents the velocity of materials at a specific spatial point x. The symbol  $\circ$  represents the composition of mappings. Similarly, accelerations are defined as

$$\mathbf{A} = \frac{d^2 \boldsymbol{\varphi}}{dt^2} \quad and \quad \mathbf{a} = \mathbf{A} \circ \boldsymbol{\varphi}^{-1}$$
 (2.30)

Time derivatives of deformation gradient in material and spatial forms are

$$\dot{\boldsymbol{F}} = \frac{d}{dt} \left( \nabla_{\boldsymbol{X}} \boldsymbol{\varphi} \right) = \nabla_{\boldsymbol{X}} \boldsymbol{V} = \nabla_{\boldsymbol{x}} \boldsymbol{v} \cdot \boldsymbol{F}$$
(2.31)

where we can define the *velocity gradient* as

$$\boldsymbol{l} := \nabla_{\boldsymbol{x}} \boldsymbol{v} = \dot{\boldsymbol{F}} \boldsymbol{F}^{-1} \tag{2.32}$$

Furthermore, we decompose the velocity gradient to symmetric and antisymmetric terms, which is

$$\boldsymbol{l} = \boldsymbol{l}^s + \boldsymbol{l}^a = \boldsymbol{d} + \boldsymbol{w} \tag{2.33}$$

We call s the rate of deformation tensor and w the spin tensor.

#### **Stresses and Balance Principles**

#### **Conservation of Mass**

Consider a subregion  $\mathcal{P}$  in the body  $\mathcal{B}$  which is subjected to a deformation  $\varphi(\mathcal{B})$ . The density of at a material point in referential and deformed configurations are denoted by  $\rho_0(\mathbf{X})$  and  $\rho(\mathbf{X})$ , respectively. We may write the mass of the subregion  $\mathcal{P}$  as

$$m_{\mathcal{P}} = \int_{\mathcal{P}} \rho_0 dV = \int_{\boldsymbol{\varphi}(\mathcal{P})} \rho dv \tag{2.34}$$

Introducing the relation between volumes in referential and deformed configurations dv = JdV, where  $J = det \mathbf{F}$ , we have

$$\int_{\mathcal{P}} (\rho_0 - \rho J) dV = 0 \tag{2.35}$$

For all material parts in  $\mathcal{P}$ . The *localization theorem* says

$$\rho_0 = \rho J \tag{2.36}$$

for any material point in  $\mathcal{B}$ . We call it the local *conservation of mass equation*. To monitor the motion of the body, we consider

$$\dot{J} = \frac{d}{dt}(det \mathbf{F}) = J \mathbf{F}^{-T} : \dot{\mathbf{F}} = J \nabla_{\mathbf{x}} \cdot \mathbf{v}$$
(2.37)

and we arrive the following equation

$$\dot{\rho} + \rho \nabla_{\boldsymbol{x}} \cdot \boldsymbol{v} = 0 \tag{2.38}$$

which is a local form of transport theorem.

#### Cauchy Stress and Balance of Linear Momentum

The concept of stress can be derived from the notion of traction t, which is the applied force per unit area  $\Delta a$  with unit normal n in the limit of  $\Delta a \rightarrow 0$ ,

$$\boldsymbol{t} = \lim_{\Delta a \to 0} \frac{\Delta \boldsymbol{F}}{\Delta a} = \boldsymbol{\sigma} \cdot \boldsymbol{n}$$
(2.39)

The above definition is based on current configuration, which brings in the so called *Cauchy* stress  $\sigma$ . We will introduce several alternatives due to the requirement of energy conjugate. But among them, only Cauchy stress is physically meaningful.

The balance of linear momentum of the subregion  $\mathcal{P}_t$  in the current configuration says

$$\frac{d}{dt} \left[ \int_{\mathcal{P}_t} \rho \boldsymbol{v} dv \right] = \int_{\mathcal{P}_t} \rho \boldsymbol{b} dv + \int_{\partial \mathcal{P}_t} \boldsymbol{t} da$$
(2.40)

for the current density  $\rho$ , surface traction t, body force b and velocity v. Left hand side is the rate of change of linear momentum while right hand side is the total applied force. Utilizing divergence theorem and the definition of Cauchy stress, we arrive

$$\int_{\mathcal{P}_t} [\rho \boldsymbol{a} - \nabla_{\boldsymbol{x}} \cdot \boldsymbol{\sigma} - \rho \boldsymbol{b}] dv = 0$$
(2.41)

for any part  ${\mathcal P}$  which implies the local form

$$\nabla_{\boldsymbol{x}} \cdot \boldsymbol{\sigma} + \rho \boldsymbol{b} = \rho \boldsymbol{a} \tag{2.42}$$

Introducing the so called *first Piola-Kirchhoff stress tensor*  $\boldsymbol{P}$  as

$$\boldsymbol{P} := J\boldsymbol{\sigma}\boldsymbol{F}^T = \boldsymbol{\tau}\boldsymbol{F}^T \tag{2.43}$$

with  $\tau = J\sigma$  the Kirchhoff stress tensor, and employing the relation of area change, we obtain

$$tda = \sigma n da = P N dA = T dA \tag{2.44}$$

The balance of linear momentum can be written in reference configuration as

$$\int_{\mathcal{P}} [\rho_0 \boldsymbol{A} - \nabla_{\boldsymbol{X}} \cdot \boldsymbol{P} - \rho \boldsymbol{B}] dv = 0$$
(2.45)

where A and B are acceleration and body force evaluated in reference configuration. The local statement of the above equation is then

$$\nabla_{\boldsymbol{X}} \cdot \boldsymbol{P} + \rho \boldsymbol{B} = \rho_0 \boldsymbol{A} \tag{2.46}$$

#### **Power Conjugation**

The balance of linear momentum leads to the theorem of expanded work, which states

$$\int_{\partial \mathcal{P}_t} \boldsymbol{t} \cdot \boldsymbol{v} da + \int_{\mathcal{P}_t} \rho \boldsymbol{b} \cdot \boldsymbol{v} dv = \int_{\mathcal{P}_t} \boldsymbol{\sigma} : \boldsymbol{d} dv + \frac{dK}{dt}$$
(2.47)

In the above equation, left hand side is the external power; first term in right hand side is the stress power; last term is the rate of change of kinetic energy. The *strain rate tensor* d is given as

$$\boldsymbol{d} = sym(\nabla_{\boldsymbol{x}}\boldsymbol{v}) = \boldsymbol{F}^{-T} \dot{\boldsymbol{E}} \boldsymbol{F}^{-1}$$
(2.48)

The stress power can be expressed as alternative forms as

$$\int_{\varphi(\mathcal{P})} \boldsymbol{\sigma} : \boldsymbol{d}dv = \int_{\mathcal{P}} \boldsymbol{\tau} : \boldsymbol{d}dV$$
$$= \int_{\mathcal{P}} \boldsymbol{P} : \dot{\boldsymbol{F}}dV$$
$$= \int_{\mathcal{P}} \boldsymbol{S} : \boldsymbol{F}^{T}\dot{\boldsymbol{F}}dV$$
$$= \int_{\mathcal{P}} \boldsymbol{S} : \dot{\boldsymbol{E}}dV \qquad (2.49)$$

where the second Piola-Kirchhoff stress tensor is defined as

$$\boldsymbol{S} = \boldsymbol{F}^{-1} \boldsymbol{P} \tag{2.50}$$

The alternative expressions of stress power motivated the introduction of different stress measures, which are usually chosen for convenience of specific problems in practice.

### **Constitute Relations**

So far, we have introduced the kinematic quantities such as deformation gradient and strains, and kinetic quantities such as stresses with balance principles. Those quantities and relations are derived in the perspective of mathematics without introducing empirical assumptions. Therefore, they are hold for any circumstances at any time. However, based on common sense, mechanical performance depends on specific materials, i.e. different materials will result in distinct response under same conditions of external stimuli. Thus, constitutive relations connecting two sets of physical quantities of kinematics and kinetics are required to reflect the behaviors of various materials. Mathematically, we have 21 variables in a 3-D mechanical system while given 15 equations so far. 6 more equations regarding constitutive relation are to be added.

There are enormous constitutive theories developed to model different kinds of materials, e.g. elastic, plastic, viscoelastic, etc. Most of them are phenomenological, which is, depend on daily experience and observation. For example, stresses in a body is observed to depend on the history of the motion. If we assume the stress state of a material point depends on the motion of its neighbor points only, this material is termed as simple material. Experiments are usually implemented to obtain the parameters to fit the model. Another observation is the so called *material frame indifference*, which states that the response of materials is independent of the observer.

Due to the large amount of constitutive models for different materials, it is impossible to exhaust them. As a demonstration, we briefly introduce two simple models.

#### • Cauchy elastic materials

Generally, stress state and work done by stress depend on the deformation history. Specifically, if we assume that stress in a material depend on the current deformation only, this material is called *Cauchy elastic materials*. We represent the stress as

$$\boldsymbol{\sigma} = \boldsymbol{\sigma}(\boldsymbol{F}(\boldsymbol{X}, t), \boldsymbol{X}) \tag{2.51}$$

In some cases, we may be interested in Piola-Kirchhoff stress tensors, which are

$$\boldsymbol{P} = \boldsymbol{P}(\boldsymbol{F}(\boldsymbol{X}, t), \boldsymbol{X}) \quad and \quad \boldsymbol{S} = \boldsymbol{S}(\boldsymbol{F}(\boldsymbol{X}, t), \boldsymbol{X})$$
(2.52)

The second argument X is due to the inhomogeneity of the material, which is, the stressstrain relation or density changes from point to point. For homogeneous material, stress does not depend on X. A Cauchy elastic material is also called a simple material.

• Hyperelastic materials

Integrating Eq.(2.47) in the time range between  $t_0$  and  $t_1$  gives

$$W_{ext} = W_{stress} + K(t_1) - K(t_0)$$
(2.53)

If we define a closed process as  $\boldsymbol{x}(\boldsymbol{X}, t_0) = \boldsymbol{x}(\boldsymbol{X}, t_1)$  and  $\dot{\boldsymbol{x}}(\boldsymbol{X}, t_0) = \dot{\boldsymbol{x}}(\boldsymbol{X}, t_1)$ , we conclude from the above equation that  $W_{ext} = W_{stress}$  in a closed process. According to the second

law of thermodynamics,  $W_{ext} \ge 0$ . In the case of  $W_{ext} > 0$ , the process is said to be dissipative. Otherwise, the material is *hyperelastic*. We write the stress work in a closed process

$$W_{stress} = \int_{t_0}^{t_1} \int_{\mathcal{P}} \boldsymbol{P} : \dot{\boldsymbol{F}} dV dt = 0$$
(2.54)

for a subregion  $\mathcal{P}$ . The local form is

$$W_{stress} = \int_{t_0}^{t_1} \boldsymbol{P} : \dot{\boldsymbol{F}} dt = 0$$
(2.55)

The concept of *hyperelastic material* has an equivalent statement: the first Piola-Kirchhoff stress tensor is given by

$$\boldsymbol{P} = \frac{\partial W(\boldsymbol{F}, \boldsymbol{X})}{\partial \boldsymbol{F}}$$
(2.56)

where  $\boldsymbol{W}$  is a state function called *stored energy*.

### 2.2 Parrinello-Rahman Molecular Dynamics

As proposed by Parrinello and Rahman [59, 60, 61, 62], a MD cell is allowed to change its volume and shape, which is described by  $3\times 3$  matrix  $\boldsymbol{h}$  whose column vectors  $\boldsymbol{a}, \boldsymbol{b}, \boldsymbol{c}$  are three edges of the cell. The spatial positions of atoms  $\boldsymbol{r}_i, i = 1, 2, \cdots, N$  thus can be written in terms of  $\boldsymbol{h}$  and the local coordinates  $\boldsymbol{s}_i$ , which is given as,

$$\boldsymbol{r}_{i} = \boldsymbol{h}(t) \cdot \boldsymbol{s}_{i}(t) = \xi_{i} \boldsymbol{a} + \eta_{i} \boldsymbol{b} + \zeta_{i} \boldsymbol{c}$$

$$(2.57)$$

where i = 1, 2, ..., N is the index of the atoms, and  $\xi_i, \eta_i, \zeta_i$  are the projections of the local atom position vector  $s_i$  onto the representative MD cell edge vectors  $\boldsymbol{a}, \boldsymbol{b}$ , and  $\boldsymbol{c}$ . Note that the local atom coordinates are with respect to the center of mass of the MD cell, which makes the origin as the representative MD cell.

Since the center of the cell is the origin of the local coordinate  $s_i$ , we have

$$-0.5 \le \xi_i(t), \eta_i(t), \zeta_i(t) \le 0.5$$

One may view  $s_i$  as statistical coordinates or internal variables representing the pattern of atomic distribution. We may call the local space  $s_i \in \mathcal{B}_s$  as the (equilibrium) statistical configuration of the atomistic ensemble. Furthermore, both the cell size and local coordinates vary with time, thus each MD cell is a set with a controlled (fixed) number of atoms that have fixed mass, and it is neither a fixed spatial (Eulerian) space nor a fixed volume (Lagrangian) ensemble.

The original Lagrangian of the Parrinello-Rahman molecular dynamics for an isoenthalpicisobaric (NPH) ensemble is given as,

$$\mathcal{L} = \frac{1}{2} \sum_{i} m_{i} \dot{\boldsymbol{s}}_{i} \cdot \boldsymbol{G} \cdot \dot{\boldsymbol{s}}_{i} + \frac{1}{2} WTr(\boldsymbol{\dot{h}}^{T} \boldsymbol{\dot{h}}) - \frac{1}{2} \sum_{i} \sum_{j \neq i} V(r_{ij}) - p\Omega \qquad (2.58)$$

where p is the hydrostatic pressure results from the environment, i.e. the interaction from the atoms outside the cell;  $\boldsymbol{G} = \boldsymbol{h}^T \cdot \boldsymbol{h}$ ;  $\Omega$  is the current volume of the cell;  $m_i$  is the mass of the i-th atom, V is the interatomic potential;  $Tr(\cdot)$  is the trace operator, and W is a quantity with unit of mass  $\times$  length<sup>2</sup>, which was not thoroughly justified in the original PR-MD formulation.

The Euler-Lagrangian equations of the PR molecular dynamics are

$$\frac{d}{dt}\frac{\partial \mathcal{L}}{\partial \dot{\boldsymbol{h}}} - \frac{\partial \mathcal{L}}{\partial \boldsymbol{h}} = 0, \qquad (2.59)$$

$$\frac{d}{dt}\frac{\partial \mathcal{L}}{\partial \dot{\boldsymbol{s}}_i} - \frac{\partial \mathcal{L}}{\partial \boldsymbol{s}_i} = 0 \ . \tag{2.60}$$

Following the standard procedure, one may derive the equations of motion for each atom, which are given as follows,

$$\ddot{\boldsymbol{s}}_{i} = -\sum_{j \neq i} \left( \frac{V'(r_{ij})}{m_{i}r_{ij}} \right) (\boldsymbol{s}_{i} - \boldsymbol{s}_{j}) - \boldsymbol{G}^{-1} \cdot \dot{\boldsymbol{G}} \cdot \dot{\boldsymbol{s}}_{i}$$
(2.61)

$$W\ddot{\boldsymbol{h}} = -(\boldsymbol{\sigma}_{int} + p\boldsymbol{I}) \cdot \boldsymbol{h}^{-T}\Omega$$
(2.62)

In Eq. (2.62),  $\boldsymbol{\sigma}_{int}$  is the internal stress that is defined as,

$$\boldsymbol{\sigma}_{int} = \frac{1}{\Omega} \sum_{i} \left( -m_i \boldsymbol{v}_i \otimes \boldsymbol{v}_i + \sum_{j \neq i} V'(r_{ij}) \left( \frac{\boldsymbol{r}_{ij} \otimes \boldsymbol{r}_{ij}}{r_{ij}} \right) \right), \tag{2.63}$$

where  $\Omega$  is the volume of the MD cell in the current configuration, and the fine scale velocity  $\boldsymbol{v}_i$  is defined as

$$\boldsymbol{v}_i = \boldsymbol{h} \cdot \dot{\boldsymbol{s}}_i \tag{2.64}$$

The original PR-MD is actually a multiscale method, and it couples the mesoscale kinematic variable h with the microscale variables  $s_i$ . It is probably an earliest attempt to establish multiscale relationship between kinematic variables at different scales. To describe the mesoscale more naturally, Podio-Guidugli [63] suggested an alternative formulation of PR-MD based on the concept of continuum mechanics. The basic idea is using the well-defined deformation gradient F to replace the shape tensor of a MD unit cell, h, which is defined by

$$\boldsymbol{F} = \boldsymbol{h}(t) \cdot \boldsymbol{h}_0^{-1} \tag{2.65}$$

where  $h_0$  is a second order tensor that maps the MD statistical configuration (*s*-configuration) to a continuum referential configuration (*R*-configuration), i.e.  $R_i = h_0 s_i$ , so that we can use



Figure 2.1: Differential maps between different lattice configurations,  $\boldsymbol{h} = [\boldsymbol{a}(t), \boldsymbol{b}(t), \boldsymbol{c}(t)]$ .

the concept as well as the language of the well-known material-spatial configuration or the Lagrangian-Eulerian description in continuum mechanics in atomistic computations. The relationships among s-configuration, R-configuration, and r-configuration are illustrated in Fig.2.1.

In fact, the idea had been suggested in Parrinello and Rahman's original paper [59], and they suggested using  $h_0 = \langle h(t) \rangle$ , which indicated that they had vaguely anticipated a reference configuration thirty years ago. Therefore, in terms of the deformation gradient, we can write,

$$\boldsymbol{r}_i = \boldsymbol{h} \cdot \boldsymbol{s}_i = \boldsymbol{h} \cdot \boldsymbol{h}_0^{-1} \cdot \boldsymbol{R}_i = \boldsymbol{F} \boldsymbol{R}_i \quad and \quad \boldsymbol{R}_i = \boldsymbol{h}_0 \cdot \boldsymbol{s}_i$$

$$(2.66)$$

where  $\mathbf{r}_i$  are the local atom position (relative to the center of mass) in the current configuration, whereas  $\mathbf{R}_i$  are defined as the local atom position in the referential configuration.

In continuum mechanics, the referential configuration is often treated as the initial configuration, which means that the configuration of the system is in its initial state, or it is the frozen state of the system at initial time.

In an atomistic lattice system, it makes sense to specify the initial lattice configuration, i.e.  $\mathbf{h}_0 = \mathbf{h}(0)$  but it does not make sense to specific the initial atomic vibration state, because atoms are always oscillating in the lattice, and we cannot freeze atom vibrations if temperature is not zero. The atom vibration only stops when the temperature of the system reaches to absolute zero. Therefore, in *s*-configuration and *R*-configuration, both  $s_i(t)$  and  $\mathbf{R}_i(t)$  are function of time. In fact, one may want distinguish the time scale as well. For the time *t* in  $s_i(t)$  and  $\mathbf{R}_i(t)$ , it is a fine scale time, which has an atomistic time scale; whereas

the time variable t in h(t) has a much larger time scale, i.e. at mesoscale. By introducing the second Piola-Kirchhoff stress,

$$\boldsymbol{\mathcal{S}} = det(\boldsymbol{F})\boldsymbol{F}^{-1} \cdot \boldsymbol{\sigma}\boldsymbol{F}^{-T}, \qquad (2.67)$$

where  $\sigma$  is the external Cauchy stress, one may set forth an anisotropic Lagrangian as follows,

$$\mathcal{L} = \frac{1}{2} \sum_{i} m_{i} \dot{\boldsymbol{R}}_{i} \cdot \boldsymbol{C} \cdot \dot{\boldsymbol{R}}_{i} - \frac{1}{2} \sum_{i} \sum_{j \neq i} V(r_{ij}) + \boldsymbol{S} : \boldsymbol{E} \Omega_{0}$$
(2.68)

where  $C = F^T F$  is the right Cauchy-Green tensor, and  $E = \frac{1}{2}(C - I)$  is the Green-Lagrangian tensor, and  $\Omega_0$  is the referential volume. Thus the equation of motion

$$\ddot{\boldsymbol{R}}_{i} = -\sum_{j \neq i} \left( \frac{V'(r_{ij})}{m_{i}r_{ij}} \right) (\boldsymbol{R}_{i} - \boldsymbol{R}_{j}) - \boldsymbol{C}^{-1} \cdot \dot{\boldsymbol{C}} \cdot \dot{\boldsymbol{R}}_{i}$$
(2.69)

$$W\ddot{\boldsymbol{F}} = -\Omega_0 \boldsymbol{F}(\boldsymbol{S}_{int} - \boldsymbol{S}) . \qquad (2.70)$$

where  $\Omega_0$  is the volume of MD cell in the referential configuration, and

$$\boldsymbol{\mathcal{S}}_{int} := \frac{1}{\Omega_0} \sum_{i} \left( -m_i \dot{\boldsymbol{R}}_i \otimes \dot{\boldsymbol{R}}_i + \sum_{j \neq i} \frac{V'(r_{ij})}{r_{ij}} (\boldsymbol{R}_j - \boldsymbol{R}_i) \otimes (\boldsymbol{R}_j - \boldsymbol{R}_i) \right)$$
$$= \frac{1}{\Omega_0} \boldsymbol{h}_0 \cdot \sum_{i} \left( -m_i \dot{\boldsymbol{s}}_i \otimes \dot{\boldsymbol{s}}_i + \sum_{j \neq i} \frac{V'(r_{ij})}{r_{ij}} (\boldsymbol{s}_j - \boldsymbol{s}_i) \otimes (\boldsymbol{s}_j - \boldsymbol{s}_i) \right) \cdot \boldsymbol{h}_0^T \quad (2.71)$$

In above derivations, we can see that the approach of using deformation gradient to replace the shape tensor is more convenient to establish an anisotropic multiscale molecular dynamics. However, the modified APR-MD is still limited to a single representative volume element (RVE) cell under equilibrium condition due to its periodic boundary condition and constant stress condition. To overcome this limitation, we shall propose a complete multiscale micromorphic molecular dynamics that is applicable to arbitrary domain with arbitrary macroscale boundary conditions, i.e. it is a multiscale molecular dynamics that supports non-uniform stress field, and it can is subjected to general non-equilibrium conditions. In molecular dynamics, if spatial distribution of stress is non-uniform, the atomistic system is not in a mechanical equilibrium state.

### 2.3 Multiscale Structure and Kinematics

The basic thermodynamical ensemble unit in macroscale solid mechanics is a "material point" at which we can define stress and strain, and the material in a continuum body filled the entire domain of space it occupies. However materials in microscopic scale are basically atoms and molecules separated in space, and these basic units are approximated as rigid balls in


Figure 2.2: A molecular system is divided to several supercells. Each supercell is a "material point" standing on the level of macroscale continuum mechanics. The decomposition  $\mathbf{r}_i = \mathbf{r}_{\alpha} + \boldsymbol{\phi}_{\alpha} \cdot \mathbf{s}_i$  gives the atomic positions inside each cell. Boundary cells exposed to surface traction and displacement are marked in deep color, while light color is used for interior cells.

molecular dynamics. To find the connection between these two scales, a good starting point is introducing deformable "material point" into microscopic resolution.

As shown in Fig.2.2, a molecular system  $\Omega$  is subjected to macroscale boundary conditions as surface traction  $\bar{t}$  and boundary displacement  $\bar{u}$ . We divide the system into several supercells  $\Omega_{\alpha}$ . Each supercell is a cluster of atoms or molecules. This partition enables us to keep track the motion on the level of a supercell due to the relative motion of internal atoms as "deformation". The shapes of the supercells are arbitrary, so that they can be chosen to fit the geometry of surface domain. The size of cells, on the other hand, will be discussed later in the next chapter. In this section, we will discuss kinematics on both the perspectives of a single supercell and the global system.

We first put emphasis on the kinematic behavior of a generic  $\alpha$ -th cell. Throughout the dissertation, we will use Greek letters  $\alpha$  and  $\beta$  to index supercells, and lower-case letters i and j to index atoms. The motion of a single supercell can be described by **rigid body translation**, **rotation** and **stretch** which are at macroscale and mesoscale. For the precise definition of macroscale and mesoscale, we will discuss later in the motion of global system. In addition, there are **internal atomistic degrees of freedom** with microscopic resolution. The connection between these motions are found as

$$\boldsymbol{r}_{i}(t) = \boldsymbol{r}_{\alpha}(t) + \boldsymbol{\phi}_{\alpha}(t) \cdot \boldsymbol{s}_{i}(t)$$
(2.72)

where  $r_i$  is atomistic positions,  $r_{\alpha}$  is the center of mass of  $\alpha$ -th cell which is calculated as,

$$\boldsymbol{r}_{\alpha} = \sum_{i} m_{i} \boldsymbol{r}_{i} / \sum_{i} m_{i}$$
(2.73)

with  $m_i$  the mass of *i*-th atom in  $\alpha$ -th cell. The rigid body translation of a supercell can be casted into the motion of the center of mass  $\mathbf{r}_{\alpha}$ .  $\boldsymbol{\phi}_{\alpha}$  is the total deformation gradient of  $\alpha$ -th cell and is uniform throughout the cell. We use "total deformation" here to distinguish it from continuum deformation. Because the supercells are not necessary to be connected continuously and there can be gaps or overlaps among cells in atomistic resolution. We may decompose  $\boldsymbol{\phi}_{\alpha}$  by the polar decomposition theorem as,

$$\boldsymbol{\phi}_{\alpha} = \boldsymbol{R}_{\alpha} \cdot \boldsymbol{U}_{\alpha} = \boldsymbol{V}_{\alpha} \cdot \boldsymbol{R}_{\alpha} \tag{2.74}$$

 $U_{\alpha}$  and  $V_{\alpha}$  are right stretch tensor and left stretch tensor, and  $R_{\alpha}$  is the rotation tensor, which are referred from section 2.1. Thus, deformation gradient  $\phi_{\alpha}$  describes the rotation and stretch of the supercell.  $s_i$  is the independent local coordinate of *i*-th atom inside the supercell as internal degree of freedom which controls the pattern of atomistic distribution but does not influence the shape of the cell. The product  $\phi_{\alpha} \cdot s_i$  is interpreted as the relative position to the center of mass.

Fig.2.3 demonstrates the procedure of evolution for a generic supercell. (a) is the referential configuration of the cell with quantities marked by superscript "0". For convenience, we may define the initial cell at time t = 0 as in the referential configuration, namely,  $\mathbf{r}_{\alpha}^{0} = \mathbf{r}_{\alpha}(0)$ ,  $\boldsymbol{\phi}_{\alpha}^{0} = \boldsymbol{\phi}_{\alpha}(0)$  and  $\mathbf{s}_{i}^{0} = \mathbf{s}_{i}(0)$ . And initial deformation gradient  $\boldsymbol{\phi}_{\alpha}(0)$  can be chosen as identity tensor  $\mathbf{I}$ . Therefore,  $\mathbf{s}_{i}^{0}$  becomes the relative position in referential configuration as shown in Fig.2.3(a). Eq.(2.72) in referential configuration is then,

$$\boldsymbol{r}_{i}^{0} = \boldsymbol{r}_{\alpha}^{0} + \boldsymbol{\phi}_{\alpha}^{0} \cdot \boldsymbol{s}_{i}^{0} = \boldsymbol{r}_{\alpha}^{0} + \boldsymbol{s}_{i}^{0}$$

$$(2.75)$$

(c) is the current configuration of the cell described by Eq.(2.72). However, different from continuum mechanics, (c) and (a) are not directly linked by a deformation gradient. If we observe the particles inside the cell in (a) and (c) respectively, we may find the changes of not only the shape of the cell, but also the pattern of atomistic distribution. Thus, we introduce an intermediate configuration (b). In this configuration, the shape of the cell holds while  $s_i(t)$  changes with time. The step (a)~(b) modifies the internal distribution, but we cannot see it at the level of macroscale. Step from (b) to (c) is then, the standard procedure of macroscale deformation as in continuum mechanics. Note that the deformation is uniform for the whole cell. Also note that (b) configuration is "artificial" which is not really existed, and its time instance is overlapped with (c), which means (b) and (c) are at the same time but in different spaces. In summary,  $s_i(t)$  represents the internal degrees of freedom which controls the atomistic distribution, while  $\phi_{\alpha}(t)$  controls the deformation of the whole supercell.



Figure 2.3: Evolution of a general cell. Red circle is the center of mass of the cell. (a) is the cell in the referential (initial) configuration; (b) is the intermediate configuration where internal patterns of atomic distribution changes but the shape of the cell keeps fixed; (c) is the real physical configuration at current time t. From (b) to (c), the shape change of the cell is described by deformation gradient  $\phi_{\alpha}$ . (b) and (c) are at the same time instance but in different space.

Next, we discuss the motion of the global system, i.e. the relative displacements and deformations among cells. Since we did not enforce any continuum restriction on supercells, the motion is totally independent on each other. However, if we are interested in extracting or applying continuum deformation to the molecular system, we can further decompose  $\phi_{\alpha}(t)$ as,

$$\boldsymbol{\phi}_{\alpha}(t) = \boldsymbol{\chi}_{\alpha}(t) \cdot \boldsymbol{F}_{\alpha}(t) \tag{2.76}$$

where  $\mathbf{F}_{\alpha}$  describes the continuum deformation, and  $\boldsymbol{\chi}_{\alpha}$  is the independent cell rotation and distortion. With given boundary geometry, distribution of centers of mass and continuum restriction, deformation gradient  $\mathbf{F}_{\alpha}$  is uniquely determined. We write it as  $\mathbf{F}_{\alpha} = \mathbf{F}_{\alpha}(\{\mathbf{r}_{\beta}\})$ , where  $\{\mathbf{r}_{\beta}\}$  means the set of all centers of mass. We use  $\beta$  to distinguish from the generic cell  $\alpha$  under study. We categorize the independent deformation  $\boldsymbol{\chi}_{\alpha}$  as mesoscale motion.

Fig.2.4 schematically depicts the deformation of the system on macroscale and mesoscale level (only rigid body translation, rotation and stretch are showed in the figure, internal degrees of freedom are smoothed out). The total deformation gradient  $\phi_{\alpha}$  consists of a macro deformation  $F_{\alpha}$  and meso deformation  $\chi_{\alpha}$ . Configuration in Fig.2.4(a) is the undeformed



Figure 2.4: Macro and meso level deformation : (a)The original undeformed system that is divided into several supercells. The black circles represent the centers of mass; (b) The configuration undergoes macroscale deformation  $F_{\alpha}$ . Note the cells are connected to each other without gaps or overlaps in the framework of continuity, and this deformation is restricted by the relative positions of centers of mass from different cells; (c) The cells further undergoe mesoscale deformation  $\chi_{\alpha}$  around their own centers of mass separately without connection. The dashed parallelograms represent the corresponding configuration in (b)

system. From (a) to (b), with the mapping of  $\mathbf{F}_{\alpha}$ , the cells are deformed but still connected to each other without gaps or overlaps due to the continuum restriction on macroscale. From(b) to (c), the mesoscale deformation  $\chi_{\alpha}$  brings in further rotation and stretch to break the connection on macroscale level.

In summary, the proposed model has three scales. Macroscale or continuum scale describes motion of centers of mass  $r_{\alpha}$  and continuum deformation  $F_{\alpha}$ . They are physically the rigid body translation, continuous rotation and stretch. The macroscale kinematics may be described by using the language of continuum mechanics. Mesoscale deformation  $\chi_{\alpha}$  further reveals the independent distortion of the cells without relocating centers of mass. Microscale motion  $s_i$  is the internal DOF which describes the atomistic pattern or distribution. The combination of three scales fully recovers molecular systems without additional assumption. With the decomposition, higher scale quantities are embedded into the molecular system, which gives us insights on the multiscale structure and provides us physical principle to solve cross-scale problems. Total deformation  $\phi_{\alpha}(t)$  is enough for most applications unless we want to apply macroscale strain boundary condition. Usually, macroscale boundary conditions are in terms of traction or displacement. We shall discuss this later in this chapter.

### 2.4 Force Field and Potential Energy

Fundamentally, forces in a molecular system are interactions among discrete particles. With multiscale structure and kinematics, it is necessary to characterize the force field for supercells in other scales. As is shown in Fig.2.5, for a generic  $\alpha$ -th cell, the external force field acting on the surface of the cell can be distinguished to two different types. First part is interatomic forces from atoms outside a generic cell which is marked by blue arrows in Fig.2.5. This is the fundamental force field on microscale level. Besides interatomic forces, a supercell is sometimes exposed to external loads ideally applied at infinite distance. Loads in this circumstance are in macroscopic scale, and usually has the form of surface traction  $\bar{t}_{\alpha}$  and body force  $\bar{b}_{\alpha}$ .

For the  $\alpha$ -th cell, consider separating potential energy to internal part  $V_{\alpha}^{int}$  and external part  $V_{\alpha}^{ext}$ . We define,

$$V_{\alpha}^{int} = \frac{1}{2} \sum_{i,j \in \alpha} \varphi(r_{ij}) \tag{2.77}$$

where i and j are both indices of atoms inside  $\alpha$ -th cell,  $\varphi$  is the pair potential, and  $r_{ij}$  is the distance between *i*-th and *j*-th atoms. Note that there is a factor of 1/2 because each pair is doubly summated.

The external potential  $V_{\alpha}^{ext}$  is related to external force field which is contributed by microscale and macroscale sources. The interactions from surrounding particles give,

$$V_{\alpha}^{atom} = \sum_{i \in \alpha, j \notin \alpha} \varphi(r_{ij}) \tag{2.78}$$



Figure 2.5: A supercell is subject to microscale interatomic forces from other cells (blue arrow) and macroscale surface traction  $\bar{t}_{\alpha}$ 

Here i again is the index of atoms inside the cell, but j is the index of atoms in the neighbor cells. Abbreviation  $i \in \alpha$  means that the i-th atom belongs to the  $\alpha$ -th cell, and  $j \notin \alpha$ means that the j-th atom does not belong to the  $\alpha$ -th cell. There is no 1/2 factor because each pair is summated once. Both  $V_{\alpha}^{int}$  and  $V_{\alpha}^{atom}$  are in the scope of microscale and they have the same nature of particle interaction. For macroscale surface traction and body force, associated potential energy can be expressed as,

$$V_{\alpha}^{surf} = -S_{\alpha}^{0} \bar{\boldsymbol{t}}_{\alpha}^{0} \cdot \boldsymbol{r}_{\alpha}$$

$$(2.79)$$

$$V_{\alpha}^{body} = -\Omega_{\alpha}^{0} \bar{\boldsymbol{b}}_{\alpha}^{0} \cdot \boldsymbol{r}_{\alpha}$$

$$(2.80)$$

where  $S^0_{\alpha}$  is the surface area exposed to  $\bar{t}^0_{\alpha}$  in the referential configuration.  $\Omega^0_{\alpha}$  is the volume in referential configuration.  $\bar{b}^0_{\alpha}$  is the applied body force. We use referential quantities to represent dead load. In Eqs.(2.79) and (2.80), we apply forces at the center of mass by assuming that higher order terms such as stress couple are negligible. The total external potential for  $\alpha$ -th supercell reads,

$$V_{\alpha}^{ext} = V_{\alpha}^{atom} + V_{\alpha}^{surf} + V_{\alpha}^{body}$$

$$(2.81)$$

In some cases, we also want to determine stress distribution in simulation domain. If the stress is prescribed, we may introduce the work conjugation of stress and strain. For example,

under a prescribed first Piola-Kirchhoff (PK-I) stress  $\bar{\mathcal{P}}_{\alpha}^{ext}$  as defined in section 2.1, the work conjugation is the deformation gradient  $\phi_{\alpha}$ . We use total deformation instead of continuum deformation because the stress-strain conjugation is based on a single cell. Then the external potential energy is,

$$V_{\alpha}^{ext} = V_{\alpha}^{stress} = -\Omega_{\alpha}^{0} \bar{\boldsymbol{\mathcal{P}}}_{\alpha}^{ext} : \boldsymbol{\phi}_{\alpha}$$

$$(2.82)$$

Note that  $V_{\alpha}^{stress}$  is a joint effects of all potentials in Eq.(2.81). However, from solid mechanics, we know that stress state is a macroscale equilibrium concept, i.e., stress can induce deformation but not rigid body translation which requires net force. Thus, Eq.(2.82) is valid to derive motion for deformation  $\phi_{\alpha}$  but not for rigid body translation  $\mathbf{r}_{\alpha}$ . If we want to use Eq.(2.82) in the global system, we need to combine it with displacement loading for  $\mathbf{r}_{\alpha}$ . In general, we will use Eq(2.81).

Note that all macroscale external forces (traction, body force and PK-I stress) are in the referential configuration so that they are all dead load. In this way, we can obtain a simple potential energy expression as in Eqs. (2.79), (2.80) and (2.82). If the external forces are in current configuration and subjected to change with time, the potential energy must be expressed by integration.

### 2.5 Kinetic Energy and Statistical Assumptions

With decomposition in Eq.(2.72), we have the kinetic energy of the  $\alpha$ -th cell,

$$K_{\alpha} = = \frac{1}{2} \sum_{i} m_{i} \dot{\boldsymbol{r}}_{i} \cdot \dot{\boldsymbol{r}}_{i}$$

$$= \frac{1}{2} \sum_{i} m_{i} (\dot{\boldsymbol{r}}_{\alpha} + \dot{\boldsymbol{\phi}}_{\alpha} \cdot \boldsymbol{s}_{i} + \boldsymbol{\phi}_{\alpha} \cdot \dot{\boldsymbol{s}}_{i}) \cdot (\dot{\boldsymbol{r}}_{\alpha} + \dot{\boldsymbol{\phi}}_{\alpha} \cdot \boldsymbol{s}_{i} + \boldsymbol{\phi}_{\alpha} \cdot \dot{\boldsymbol{s}}_{i})$$

$$= \frac{1}{2} M_{\alpha} \dot{\boldsymbol{r}}_{\alpha} \cdot \dot{\boldsymbol{r}}_{\alpha} + \frac{1}{2} \dot{\boldsymbol{\phi}}_{\alpha}^{T} \dot{\boldsymbol{\phi}}_{\alpha} : \sum_{i} m_{i} \boldsymbol{s}_{i} \otimes \boldsymbol{s}_{i} + \frac{1}{2} C_{\alpha} : \sum_{i} m_{i} \dot{\boldsymbol{s}}_{i} \otimes \dot{\boldsymbol{s}}_{i}$$

$$+ \frac{1}{2} \dot{\boldsymbol{\phi}}_{\alpha}^{T} \boldsymbol{\phi}_{\alpha} : \sum_{i} m_{i} \boldsymbol{s}_{i} \otimes \dot{\boldsymbol{s}}_{i} + \frac{1}{2} \boldsymbol{\phi}_{\alpha}^{T} \dot{\boldsymbol{\phi}}_{\alpha} : \sum_{i} m_{i} \dot{\boldsymbol{s}}_{i} \otimes \boldsymbol{s}_{i} \qquad (2.83)$$

where  $M_{\alpha}$  is the mass of the whole cell;  $C_{\alpha} = \phi_{\alpha}^{T} \phi_{\alpha}$  is the right Cauchy-Green tensor as introduced in continuum mechanics. Consider representing contributions to kinetic energy by four parts: (1) Rigid body translation  $K_{\alpha}^{rigid}$ ; (2) Cell stretch and rotation  $K_{\alpha}^{cell}$ ; (3) Internal motion of atoms  $K_{\alpha}^{atom}$  and (4) Mix motion  $K_{\alpha}^{mix}$ . They are read from Eq.(2.83)

and rewritten as,

$$\begin{aligned}
K_{\alpha}^{rigid} &= \frac{1}{2} M_{\alpha} \dot{\boldsymbol{r}}_{\alpha} \cdot \dot{\boldsymbol{r}}_{\alpha} \\
K_{\alpha}^{cell} &= \frac{1}{2} \dot{\boldsymbol{\phi}}_{\alpha}^{T} \dot{\boldsymbol{\phi}}_{\alpha} : \sum_{i} m_{i} \boldsymbol{s}_{i} \otimes \boldsymbol{s}_{i} = \frac{1}{2} \dot{\boldsymbol{\phi}}_{\alpha}^{T} \dot{\boldsymbol{\phi}}_{\alpha} : \boldsymbol{J}_{\alpha} \\
K_{\alpha}^{atom} &= \frac{1}{2} \boldsymbol{C}_{\alpha} : \sum_{i} m_{i} \dot{\boldsymbol{s}}_{i} \otimes \dot{\boldsymbol{s}}_{i} \\
K_{\alpha}^{mix} &= \frac{1}{2} \dot{\boldsymbol{\phi}}_{\alpha}^{T} \boldsymbol{\phi}_{\alpha} : \sum_{i} m_{i} \boldsymbol{s}_{i} \otimes \dot{\boldsymbol{s}}_{i} + \frac{1}{2} \boldsymbol{\phi}_{\alpha}^{T} \dot{\boldsymbol{\phi}}_{\alpha} : \sum_{i} m_{i} \dot{\boldsymbol{s}}_{i} \otimes \boldsymbol{s}_{i}
\end{aligned}$$
(2.84)

where  $\boldsymbol{J}_{\alpha} = \sum_{i} m_{i} \boldsymbol{s}_{i} \otimes \boldsymbol{s}_{i}$  is defined as inertia tensor.

We next present two statistical assumptions which are originally suggested by Parrinello and Rahman [60, 59] but unexplained. We render brief physical interpretations and basic mathematical judgements.

#### **Assumption I**: Inertial tensor $J_{\alpha}$ can be approximated as a constant;

This assumption implies that the density distribution does not change significantly. For instance, if particles initially concentrated around the center of mass tend to move outward, the value of  $J_{\alpha}$  will increase. We demonstrate this point by choosing principal axes  $e_1$ ,  $e_2$  and  $e_3$ , and express  $J_{\alpha}$  as,

$$\boldsymbol{J}_{\alpha} = J_{11}\boldsymbol{e}_1 \otimes \boldsymbol{e}_1 + J_{22}\boldsymbol{e}_2 \otimes \boldsymbol{e}_2 + J_{33}\boldsymbol{e}_3 \otimes \boldsymbol{e}_3 \tag{2.85}$$

with  $J_{11} = \sum_{i} m_i s_{i1}^2$ ,  $J_{22} = \sum_{i} m_i s_{i2}^2$  and  $J_{33} = \sum_{i} m_i s_{i3}^2$ . We can see that these values are basically density distribution along each direction. The farther particles located away from the center of mass, the bigger  $J_{\alpha}$  is. For a homogenous cell, assumption I is always hold. It should be noted that in PR-MD,  $J_{\alpha}$  is a spherical tensor, namely,  $J_{11} = J_{22} = J_{33}$  and thus  $J_{\alpha} = WI$  with W a constant. This is because  $s_i$  space is homogenous and **cubic**, namely, the range of  $s_i$  on each direction is the same and normalized as [0, 1]. However, this is not the case in our cells.  $s_i$  in our model is a vector in real physical space which is not

From the above discussion, **Assumption I** can simplify the model and is best suit for a homogenous cell. We must carefully use it in applications for inhomogenous cases.

normalized. Therefore, arbitrary choosing of shape may result in polarization of  $J_{\alpha}$ .

**Assumption II**: The mixed kinetic energy  $K_{mix}$  is negligible compared to other kinetic energies.

Consider the following correlation function,

$$AC(\tau) = \langle s_i(t) \otimes s_i(t+\tau) \rangle = \sum_i m_i s_i(t) \otimes s_i(t+\tau)$$
(2.86)

where  $\tau$  is a infinite small time increment. Atomistic motion includes two parts: vibration around instant equilibrium positions with finite temperature and lattice displacement (polarization) under external force field. As shown in Fig.2.6, blue arrows represent lattice displacements, and atomistic vibration are modeled by springs connected between atoms. Atomistic vibration is much more rapid than lattice displacement. Therefore, in the neighborhood of time instant t, lattice displacement is negligible and only atomistic vibration is in scope. Statistically, for a cluster of particles vibrating around the equilibrium positions, we state without proof that the correlation function is approximately a constant of  $\tau$  in the neighborhood of t, thus,

$$\sum_{i} m_i \boldsymbol{s}_i(t) \otimes \boldsymbol{s}_i(t+\tau) \approx \sum_{i} m_i \boldsymbol{s}_i(t) \otimes \boldsymbol{s}_i(t)$$
(2.87)



Figure 2.6: Atomic vibration and polarization. Atoms are vibrating rapidly around their instant equilibrium lattice positions. This motion is modeled by springs and rigid balls. Under external force field, equilibrium positions changes slowly in the long term.

Thus, the derivation of correlation function is zero at  $\tau = 0$ , which is

$$\frac{d}{d\tau}\boldsymbol{A}C|_{\tau=0} = \sum_{i} m_{i}\boldsymbol{s}_{i}(t) \otimes \dot{\boldsymbol{s}}_{i}(t) = 0$$
(2.88)

Similarly,

$$\sum_{i} m_i \dot{\boldsymbol{s}}_i(t) \otimes \boldsymbol{s}_i(t) = 0 \tag{2.89}$$

Comparing with the Eq.(2.84), we have  $K_{\alpha}^{mix} = 0$ . Consequently, kinetic energy is decoupled and only separate terms  $K_{\alpha}^{rigid}$ ,  $K_{\alpha}^{cell}$  and  $K_{\alpha}^{atom}$  are left. This simplification makes the kinetic energy clearly associate with motions in different scales. We rewrite kinetic energy as,

> $K_{\alpha} = K_{\alpha}^{rigid} + K_{\alpha}^{cell} + K_{\alpha}^{atom}$ =  $\frac{1}{2}M_{\alpha}\dot{\boldsymbol{r}}_{\alpha} \cdot \dot{\boldsymbol{r}}_{\alpha} + \frac{1}{2}\dot{\boldsymbol{\phi}}_{\alpha}^{T}\dot{\boldsymbol{\phi}}_{\alpha} : \boldsymbol{J}_{\alpha} + \frac{1}{2}\boldsymbol{C}_{\alpha} : \sum_{i} m_{i}\dot{\boldsymbol{s}}_{i} \otimes \dot{\boldsymbol{s}}_{i}$  (2.90)

### 2.6 Dynamic Equation and Macroscale Boundary Conditions

In this section, we shall obtain equations of motion for independent variables  $\mathbf{r}_{\alpha}$ ,  $\boldsymbol{\phi}_{\alpha}$  and  $\mathbf{s}_{i}$  based on Lagrangian  $\mathcal{L}_{\alpha} = \mathcal{L}_{\alpha}(\mathbf{r}_{\alpha}, \boldsymbol{\phi}_{\alpha}, \mathbf{s}_{i})$ . If we introduce continuum deformation  $\mathbf{F}_{\alpha}$ ,  $\boldsymbol{\chi}_{\alpha}$  as a component of  $\boldsymbol{\phi}_{\alpha}$  can be chosen as an independent variable because  $\mathbf{F}_{\alpha}$  is totally determined by centers of mass  $\mathbf{r}_{\alpha}$ . We will show that the multiplicative decomposition  $\boldsymbol{\phi}_{\alpha} = \boldsymbol{\chi}_{\alpha} \cdot \mathbf{F}_{\alpha}$  does not influence the equation of motion for  $\mathbf{r}_{\alpha}$  even with the dependency of  $\mathbf{F}_{\alpha}$ . With the discussion in last two sections, we write the Lagrangian for  $\alpha$ -th cell as,

$$\mathcal{L}_{\alpha} = K_{\alpha} - V_{\alpha}$$

$$= K_{\alpha}^{rigid} + K_{\alpha}^{cell} + K_{\alpha}^{atom} - V_{\alpha}^{int} - V_{\alpha}^{ext}$$

$$= \frac{1}{2} M_{\alpha} \dot{\boldsymbol{r}}_{\alpha} \cdot \dot{\boldsymbol{r}}_{\alpha} + \frac{1}{2} \dot{\boldsymbol{\phi}}_{\alpha}^{T} \dot{\boldsymbol{\phi}}_{\alpha} : \boldsymbol{J}_{\alpha} + \frac{1}{2} \boldsymbol{C}_{\alpha} : \sum_{i} m_{i} \dot{\boldsymbol{s}}_{i} \otimes \dot{\boldsymbol{s}}_{i}$$

$$- \frac{1}{2} \sum_{i,j \in \alpha} \varphi(r_{ij}) - \sum_{i \in \alpha, j \notin \alpha} \varphi(r_{ij}) + S_{\alpha}^{0} \bar{\boldsymbol{t}}_{\alpha}^{0} \cdot \boldsymbol{r}_{\alpha} + \Omega_{\alpha}^{0} \bar{\boldsymbol{b}}_{\alpha}^{0} \cdot \boldsymbol{r}_{\alpha} \qquad (2.91)$$

The last two terms as external potential energy bring macroscale information into the molecular system. The standard procedure to derive equations of motion is stated as,

$$\frac{d}{dt}\frac{\partial \mathcal{L}_{\alpha}}{\partial \dot{\boldsymbol{r}}_{\alpha}} - \frac{\partial \mathcal{L}_{\alpha}}{\partial \boldsymbol{r}_{\alpha}} = 0$$
(2.92)

$$\frac{d}{dt}\frac{\partial \mathcal{L}_{\alpha}}{\partial \dot{\boldsymbol{\phi}}_{\alpha}} - \frac{\partial \mathcal{L}_{\alpha}}{\partial \boldsymbol{\phi}_{\alpha}} = 0$$
(2.93)

$$\frac{d}{dt}\frac{\partial \mathcal{L}_{\alpha}}{\partial \dot{\boldsymbol{s}}_{i}} - \frac{\partial \mathcal{L}_{\alpha}}{\partial \boldsymbol{s}_{i}} = 0 . \qquad (2.94)$$

#### Dynamical Equation of $r_{lpha}$

First, we consider the rigid body translation  $r_{\alpha}$ . We have,

$$\frac{d}{dt} \left( \frac{\partial \mathcal{L}_{\alpha}}{\partial \dot{\boldsymbol{r}}_{\alpha}} \right) = M_{\alpha} \ddot{\boldsymbol{r}}_{\alpha} \tag{2.95}$$

and

$$\frac{\partial \mathcal{L}_{\alpha}}{\partial \boldsymbol{r}_{\alpha}} = -\frac{1}{2} \sum_{i,j \in \alpha} \varphi'(r_{ij}) \frac{\boldsymbol{r}_{ij}}{r_{ij}} \cdot \frac{\partial \boldsymbol{r}_{ij}}{\partial \boldsymbol{r}_{\alpha}} - \sum_{i \in \alpha, j \notin \alpha} \varphi'(r_{ij}) \frac{\boldsymbol{r}_{ij}}{r_{ij}} \cdot \frac{\partial \boldsymbol{r}_{ij}}{\partial \boldsymbol{r}_{\alpha}} + S_{\alpha}^{0} \bar{\boldsymbol{t}}_{\alpha}^{0} + \Omega_{\alpha}^{0} \bar{\boldsymbol{b}}_{\alpha}^{0}$$

$$= \sum_{i \in \alpha, j \notin \alpha} \boldsymbol{f}_{ij} + S_{\alpha}^{0} \bar{\boldsymbol{t}}_{\alpha}^{0} + \Omega_{\alpha}^{0} \bar{\boldsymbol{b}}_{\alpha}^{0} \qquad (2.96)$$

by utilizing the relation

$$\frac{\partial \boldsymbol{r}_{ij}}{\partial \boldsymbol{r}_{\alpha}} = \begin{cases} -I & i \in \alpha, j \notin \alpha \\ \mathbf{0} & i, j \in \alpha \end{cases}$$
(2.97)

where I is the second order identity tensor.  $\varphi'(r_{ij})\hat{r}_{ij} = f_{ij}$ , where  $\hat{r}_{ij} = r_{ij}/r_{ij}$  is the unit vector point from i-th atom to j-th atom;  $r_{ij} = r_j - r_i$ ;  $r_{ij} = |r_{ij}|$ ;  $f_{ij}$  is the pair force acting on i-th atom by j-th atom. Note that the sign of  $f_{ij}$  is the same as  $\hat{r}_{ij}$  when  $r_{ij}$  is larger than equilibrium distance and opposite to  $\hat{r}_{ij}$  when  $r_{ij}$  is smaller than equilibrium distance. Thus, the dynamic equation for  $r_{\alpha}$  is,

$$M_{\alpha}\ddot{\boldsymbol{r}}_{\alpha} = \sum_{i \in \alpha, j \notin \alpha} \boldsymbol{f}_{ij} + S^{0}_{\alpha}\bar{\boldsymbol{t}}^{0}_{\alpha} + \Omega^{0}_{\alpha}\bar{\boldsymbol{b}}^{0}_{\alpha}$$
(2.98)

The driving force for rigid body translation  $r_{\alpha}$  consists of three parts: interaction from external particles, surface traction and body force.

We also want to learn the effect when introducing the decomposition  $\phi_{\alpha} = \chi_{\alpha} \cdot F_{\alpha}$  with  $F_{\alpha}$  depending on  $r_{\alpha}$ . In this case, the terms in dynamic equations are,

$$\frac{d}{dt} \left( \frac{\partial \mathcal{L}_{\alpha}}{\partial \dot{\boldsymbol{r}}_{\alpha}} \right) = \frac{d}{dt} \left( \frac{\partial \mathcal{L}_{\alpha}}{\partial \dot{\boldsymbol{r}}_{\alpha}} + \frac{\partial \mathcal{L}_{\alpha}}{\partial \dot{\boldsymbol{\phi}}_{\alpha}} \cdot \frac{\partial \dot{\boldsymbol{\phi}}_{\alpha}}{\partial \dot{\boldsymbol{r}}_{\alpha}} \right)$$
(2.99)

$$\frac{\partial \mathcal{L}_{\alpha}}{\partial \boldsymbol{r}_{\alpha}} = \frac{\partial \mathcal{L}_{\alpha}}{\partial \boldsymbol{r}_{\alpha}} + \frac{\partial \mathcal{L}_{\alpha}}{\partial \boldsymbol{\phi}_{\alpha}} \cdot \frac{\partial \boldsymbol{\phi}_{\alpha}}{\partial \boldsymbol{r}_{\alpha}} + \frac{\partial \mathcal{L}_{\alpha}}{\partial \dot{\boldsymbol{\phi}}_{\alpha}} \cdot \frac{\partial \dot{\boldsymbol{\phi}}_{\alpha}}{\partial \boldsymbol{r}_{\alpha}}$$
(2.100)

We will reach exactly the same equation as (2.98), which means that introducing continuum deformation does not influence rigid body translation. And continuum deformation is merely part of the independent motion of the whole system. Details of derivation is presented in Appendix A.

#### Dynamical Equation of $\phi_{\alpha}$

Then we derive the dynamic equation for  $\phi_{\alpha}$ . We have,

$$\frac{d}{dt} \left( \frac{\partial \mathcal{L}_{\alpha}}{\partial \dot{\boldsymbol{\phi}}_{\alpha}} \right) = \frac{d}{dt} \left( \dot{\boldsymbol{\phi}}_{\alpha} \cdot \boldsymbol{J}_{\alpha} \right) = \ddot{\boldsymbol{\phi}}_{\alpha} \cdot \boldsymbol{J}_{\alpha}$$
(2.101)

and

$$\frac{\partial \mathcal{L}_{\alpha}}{\partial \phi_{\alpha}} = \phi_{\alpha} \cdot \sum_{i} m_{i} \dot{\boldsymbol{s}}_{i} \otimes \dot{\boldsymbol{s}}_{i} - \frac{1}{2} \sum_{i,j \in \alpha} \varphi'(r_{ij}) \frac{\boldsymbol{r}_{ij}}{r_{ij}} \cdot \frac{\partial \boldsymbol{r}_{ij}}{\partial \phi_{\alpha}} - \sum_{i \in \alpha, j \notin \alpha} \varphi'(r_{ij}) \frac{\boldsymbol{r}_{ij}}{r_{ij}} \cdot \frac{\partial \boldsymbol{r}_{ij}}{\partial \phi_{\alpha}} \\
= \phi_{\alpha} \cdot \sum_{i} m_{i} \dot{\boldsymbol{s}}_{i} \otimes \dot{\boldsymbol{s}}_{i} - \frac{1}{2} \sum_{i,j \in \alpha} \boldsymbol{f}_{ij} \otimes \boldsymbol{s}_{ij} + \sum_{i \in \alpha, j \notin \alpha} \boldsymbol{f}_{ij} \otimes \boldsymbol{s}_{i} \qquad (2.102)$$

where  $\mathbf{s}_{ij} = \mathbf{s}_j - \mathbf{s}_i$ . Note the net forces  $S_{\alpha}^0 \overline{\mathbf{t}}_{\alpha}^0$  and  $\Omega_{\alpha}^0 \overline{\mathbf{b}}_{\alpha}^0$  do not influence the motion of  $\boldsymbol{\phi}_{\alpha}$ . If we define the internal and external first Piola-Kirchhoff(PK-I) stress as,

$$\boldsymbol{\mathcal{P}}_{\alpha}^{int} = \frac{1}{\Omega_{\alpha}^{0}} \left( \frac{1}{2} \sum_{i,j \in \alpha} \boldsymbol{f}_{ij} \otimes \boldsymbol{s}_{ij} - \boldsymbol{\phi}_{\alpha} \cdot \sum_{i \in \alpha} m_{i} \dot{\boldsymbol{s}}_{i} \otimes \dot{\boldsymbol{s}}_{i} \right)$$
(2.103)

$$\boldsymbol{\mathcal{P}}_{\alpha}^{ext} = \frac{1}{\Omega_{\alpha}^{0}} \sum_{i \in \alpha, j \notin \alpha} \boldsymbol{f}_{ij} \otimes \boldsymbol{s}_{i}$$
(2.104)

the dynamic equation for  $\phi_{\alpha}$  can be written as,

$$\ddot{\boldsymbol{\phi}}_{\alpha} \cdot \boldsymbol{J}_{\alpha} = \left(\boldsymbol{\mathcal{P}}_{\alpha}^{ext} - \boldsymbol{\mathcal{P}}_{\alpha}^{int}\right) \Omega_{\alpha}^{0}$$
(2.105)

 $\mathcal{P}_{\alpha}^{int}$  is the resistance stress compared with the driving stress  $\mathcal{P}_{\alpha}^{ext}$ , so it comes with a minus sign. When they reach a balance,  $\phi_{\alpha}$  is in equilibrium.

We know in continuum mechanics that PK-I stress is defined from Cauchy stress  $\sigma_{\alpha}$  as,

$$\boldsymbol{\mathcal{P}}_{\alpha} = det(\boldsymbol{\phi}_{\alpha})\boldsymbol{\sigma}_{\alpha} \cdot \boldsymbol{\phi}_{\alpha}^{-T}$$
(2.106)

Comparing with Eqs. (2.103) and (2.104), we find that,

$$\boldsymbol{\sigma}_{\alpha}^{int} = \frac{1}{det(\boldsymbol{\phi}_{\alpha})\Omega_{\alpha}^{0}} \left(\frac{1}{2}\sum_{i,j\in\alpha}\boldsymbol{f}_{ij}\otimes\boldsymbol{s}_{ij} - \boldsymbol{\phi}_{\alpha}\cdot\sum_{i\in\alpha}\boldsymbol{m}_{i}\dot{\boldsymbol{s}}_{i}\otimes\dot{\boldsymbol{s}}_{i}\right)\cdot\boldsymbol{\phi}_{\alpha}^{T}$$

$$= \frac{1}{\Omega_{\alpha}} \left(\frac{1}{2}\sum_{i,j\in\alpha}\boldsymbol{f}_{ij}\otimes\boldsymbol{r}_{ij} - \sum_{i\in\alpha}\boldsymbol{m}_{i}(\boldsymbol{\phi}_{\alpha}\cdot\dot{\boldsymbol{s}}_{i})\otimes(\boldsymbol{\phi}_{\alpha}\cdot\dot{\boldsymbol{s}}_{i})\right) \qquad (2.107)$$

$$\boldsymbol{\sigma}_{\alpha}^{ext} = \frac{1}{det(\boldsymbol{\phi}_{\alpha})\Omega_{\alpha}^{0}}\sum_{i\in\alpha,j\notin\alpha}\boldsymbol{f}_{ij}\otimes\boldsymbol{s}_{i}\cdot\boldsymbol{\phi}_{\alpha}^{T}$$

$$= \frac{1}{\Omega_{\alpha}}\sum_{i\in\alpha,j\notin\alpha}\boldsymbol{f}_{ij}\otimes\boldsymbol{r}_{i} \qquad (2.108)$$

which are internal and external Cauchy stresses.

Since  $\mathbf{r}_{\alpha}$ ,  $\phi_{\alpha}$  and  $\mathbf{s}_{i}$  are in different time scales, i.e.  $\Delta t_{\mathbf{r}_{\alpha}} > \Delta t_{\mathbf{\phi}_{\alpha}} \gg \Delta t_{\mathbf{s}_{i}}$ , we expect velocities  $\dot{\mathbf{r}}_{\alpha}$  and  $\dot{\phi}_{\alpha}$  are much smaller than  $\dot{\mathbf{s}}_{i}$ . Therefore, we have approximation  $\dot{\mathbf{r}}_{i} = \dot{\mathbf{r}}_{\alpha} + \dot{\phi}_{\alpha} \cdot \mathbf{s}_{i} + \phi_{\alpha} \cdot \dot{\mathbf{s}}_{i} = \phi_{\alpha} \cdot \dot{\mathbf{s}}_{i}$ . Eq. (2.107) can be written as,

$$\boldsymbol{\sigma}_{\alpha}^{int} = \frac{1}{\Omega_{\alpha}} \left( \frac{1}{2} \sum_{i,j \in \alpha} \boldsymbol{f}_{ij} \otimes \boldsymbol{r}_{ij} - \sum_{i \in \alpha} m_i \dot{\boldsymbol{r}}_i \otimes \dot{\boldsymbol{r}}_i \right)$$
(2.109)

which is the definition of Virial stress [64, 65], i.e.  $\boldsymbol{\sigma}_{\alpha}^{int} = \boldsymbol{\sigma}_{\alpha}^{Virial}$ . Apparently, microscale atomistic velocity plays a role in the macroscale stress. Extending the concept of supercell to a material point in continuum mechanics, we conclude that external stress from the environment provides driving force for stretch and rotation of the cell (material point), while the Virial stress as the internal stress resists the deformation. They will be balanced when the system in macroscale and mesoscale equilibrium. We can also interperate external stress as a pure mechanical stress and internal stress as a combination of mechanical and thermodynamical stress due to different physical origins.

#### Dynamical Equation of $s_i$

Next we derive equation of motion for  $s_i$ ,

$$\frac{d}{dt} \left( \frac{\partial \mathcal{L}_{\alpha}}{\partial \dot{\boldsymbol{s}}_{i}} \right) = \frac{d}{dt} \left( m_{i} \boldsymbol{C}_{\alpha} \cdot \dot{\boldsymbol{s}}_{i} \right) = m_{i} \dot{\boldsymbol{C}}_{\alpha} \cdot \dot{\boldsymbol{s}}_{i} + m_{i} \boldsymbol{C}_{\alpha} \cdot \ddot{\boldsymbol{s}}_{i}$$
(2.110)

and

$$\frac{\partial \mathcal{L}_{\alpha}}{\partial s_{i}} = -\sum_{j \in \alpha} \varphi'(r_{ij}) \frac{\boldsymbol{r}_{ij}}{r_{ij}} \cdot \frac{\partial \boldsymbol{r}_{ij}}{\partial s_{i}} - \sum_{j \notin \alpha} \varphi'(r_{ij}) \frac{\boldsymbol{r}_{ij}}{r_{ij}} \cdot \frac{\partial \boldsymbol{r}_{ij}}{\partial s_{i}}$$
$$= \sum_{j} \boldsymbol{f}_{ij} \cdot \boldsymbol{\phi}_{\alpha}$$
(2.111)

Note that in the above derivation, the 1/2 factor is gone because the i-th atom is summated twice in the internal potential energy term. And since internal and external parts have the same form, we simply combine them. And the dynamic equation is,

$$m_i \boldsymbol{C}_{\alpha} \cdot \ddot{\boldsymbol{s}}_i = \sum_j \boldsymbol{f}_{ij} \cdot \boldsymbol{\phi}_{\alpha} - m_i \dot{\boldsymbol{C}}_{\alpha} \cdot \dot{\boldsymbol{s}}_i$$
(2.112)

The first term on the right hand side is the driving force. The second term on the right hand side is associated with both macroscale velocity  $\dot{C}_{\alpha}$  and microscale velocity  $\dot{s}_i$ . Physically, this is a damping force which resists the motion of  $s_i$ , and the motion of the cell serves as a damping coefficient. If the system is in macro and meso scale equilibrium, namely,  $\dot{C}_{\alpha} = 0$ , this term is vanished.

So far, we have obtained dynamical Eqs. at different scales: (2.98), (2.105) and (2.112). In summary and for clarity, we rewrite them together as follow,

$$M_{\alpha}\ddot{\boldsymbol{r}}_{\alpha} = \sum_{i\in\alpha, j\notin\alpha} \boldsymbol{f}_{ij} + S^{0}_{\alpha}\bar{\boldsymbol{t}}^{0}_{\alpha} + \Omega^{0}_{\alpha}\bar{\boldsymbol{b}}^{0}_{\alpha}$$
(2.113)

$$\ddot{\boldsymbol{\phi}}_{\alpha} \cdot \boldsymbol{J}_{\alpha} = \left( \boldsymbol{\mathcal{P}}_{\alpha}^{ext} - \boldsymbol{\mathcal{P}}_{\alpha}^{int} \right) \Omega_{\alpha}^{0}$$
(2.114)

$$m_i \boldsymbol{C}_{\alpha} \cdot \ddot{\boldsymbol{s}}_i = \sum_j \boldsymbol{f}_{ij} \cdot \boldsymbol{\phi}_{\alpha} - m_i \dot{\boldsymbol{C}}_{\alpha} \cdot \dot{\boldsymbol{s}}_i$$
(2.115)

They represent rigid body translation, cell deformation and rotation, internal motion, respectively. The multiscale structure interperates the chaotic motion of molecular system in an organized atomistic-continuum viewpoint. As equations of motion, they have the same generalized form  $\tilde{M}\ddot{q} = \tilde{F}$  with generalized mass  $M_{\alpha}$ ,  $J_{\alpha}$  and  $m_i C_{\alpha}$ . The driving force for rigid body translation is the the net force acting on the cell. The motion of deformation and rotation is induced by external stress from the environment and resisted by internal Virial stress. And the internal motion is driven by interactions among particles, while damped due to the existence of cell motions.

#### Macroscale Boundary Conditions

To implement dynamical equations for a realistic multiscale system, we need proper boundary conditions. Applying macroscale boundary conditions in molecular systems as a top-down message passing is the main goal of introducing the multiscale model. There are four different kinds of macroscale boundary conditions:

- (I) Traction (force) boundary;
- (II) Displacement boundary;
- (III) Stress bounday;
- (IV) Strain boundary.

(I) and (II) are is relatively straightforward in practice. For example, we may directly apply surface traction  $\bar{t}^0_{\alpha}$  or body force  $\bar{b}^0_{\alpha}$  in equation (2.113). Conditions (III) and (IV), on the other hand, usually requires more care. We discuss stress and strain separately.

With stress boundary condition, boundary cells are controlled by desired stress states. Note that stress state is an equilibrium concept associated with cell deformation, while surface traction or body force are net forces to induce rigid body motion. In this case, we take advantage of external potential energy proposed in Eq. (2.82) and replace the  $V_{\alpha}^{ext}$  in Eq. (2.91) by  $V_{\alpha}^{stress}$ . After the same procedure of derivation, the dynamic Eq. (2.105) for  $\phi_{\alpha}$  becomes,

$$\ddot{\boldsymbol{\phi}}_{\alpha} \cdot \boldsymbol{J}_{\alpha} = \left( \bar{\boldsymbol{\mathcal{P}}}_{\alpha}^{ext} - \boldsymbol{\mathcal{P}}_{\alpha}^{int} \right) \Omega_{\alpha}^{0}$$
(2.116)

which has the same form as Eq. (2.105), but the external PK-I stress is replaced by the prescribed value  $\bar{\mathcal{P}}_{\alpha}^{ext}$ . Note that stress boundary alone is not enough to uniquely determine the evolution of the system, because we also need to specify rigid body motion. In this case, we may combine stress boundary with (I) or (II) or periodic boundary conditions .

To apply strain boundary, we may use the continuum deformation  $\boldsymbol{F}_{\alpha}$  instead of  $\boldsymbol{\phi}_{\alpha}$ . For example,  $\bar{\boldsymbol{E}}_{\alpha} = \frac{1}{2} \left( \bar{\boldsymbol{F}}_{\alpha}^{T} \bar{\boldsymbol{F}}_{\alpha} - 1 \right)$ , where  $\bar{\boldsymbol{E}}_{\alpha}$  is an applied Green-Lagrangian strain. Thus applying  $\bar{\boldsymbol{F}}_{\alpha}$  gives the desired Strain. We have learnt that  $\boldsymbol{F}_{\alpha}$  totally depends on the relative positions of centers of mass and boundary geometry, i.e.  $\bar{\boldsymbol{F}}_{\alpha} = \bar{\boldsymbol{F}}_{\alpha}(\{\bar{\boldsymbol{r}}_{\beta}\})$ . Therefore, strain boundary is finally equivalent to setting the relative positions of centers of mass. However, the relation between  $\boldsymbol{F}_{\alpha}$  and centers of mass is not a one-to-one map, because there can be

rigid body motions of the whole system (different from rigid body motion of a single cell). We still need to combine other boundary conditions.

In summary, stress and strain boundary conditions usually need to work together with other boundary conditions, otherwise the evolution of the system may not be unique. In general, we are more interested in traction and displacement boundary conditions in practice. Notice that in a micro mechanical system, macroscale boundary conditions may not be associated with a unique solution because of the possibility of phase transition. Different phase will result in distinct stress-strain relation.

### Chapter 3

## Technical Aspects of the Multiscale Theory

### 3.1 Constraints and Size Dependency

An issue that we have not discussed so far is the constraints of the variables. Recall that the physical meaning we discussed in last chapter: the center of mass  $\mathbf{r}_{\alpha}$ , the total deformation gradient  $\phi_{\alpha}$  and internal motion  $\mathbf{s}_i$ . We may specify them initially, but the physical interpretations are not guaranteed during the evolution. Actually the multiscale decomposition in Eq.(2.72) is non-unique. For example, at some certain time t, atomic position can be,

$$\mathbf{r}_{i}(t) = \mathbf{r}_{\alpha}(t) + \boldsymbol{\phi}_{\alpha}(t) \cdot \mathbf{s}_{i}(t) = \mathbf{r}_{\alpha}^{*}(t) + \boldsymbol{\phi}_{\alpha}^{*}(t) \cdot \mathbf{s}_{i}^{*}(t)$$
(3.1)

for different sets of  $\mathbf{r}_{\alpha}$ ,  $\boldsymbol{\phi}_{\alpha}$  and  $\mathbf{s}_{i}$ . The real position  $\mathbf{r}_{i}(t)$  is honestly calculated because we did not bring any additional assumption into the model, so that any comination should be equivalent to the original molecular system, i.e. another set  $\mathbf{r}_{\alpha}^{*}$ ,  $\boldsymbol{\phi}_{\alpha}^{*}$  and  $\mathbf{s}_{i}^{*}$  may give the same  $\mathbf{r}_{i}(t)$ . To avoid non-uniqueness, we shall enforce constraints on those variables.

The first constraint is that the center of mass must be concerved. With the definition of  $\mathbf{r}_{\alpha}$ , we have,

$$\mathbf{r}_{\alpha} = \frac{\sum_{i} m_{i} \mathbf{r}_{i}}{\sum_{i} m_{i}} = \frac{1}{M_{\alpha}} \left( \sum_{i} m_{i} \mathbf{r}_{\alpha} + \sum_{i} m_{i} \boldsymbol{\phi}_{\alpha} \cdot \mathbf{s}_{i} \right)$$
$$= \mathbf{r}_{\alpha} + \frac{\boldsymbol{\phi}_{\alpha}}{M_{\alpha}} \cdot \sum_{i} m_{i} \mathbf{s}_{i}$$
(3.2)

The condition to ensure Eq. (3.2) is,

$$\mathbf{G}_1 = \sum_i m_i \mathbf{s}_i(t) = 0 \tag{3.3}$$

Second, the shape change of the cell must be conserved, i.e.,  $\phi_{\alpha}$  always represents the real deformation gradient. The terminology "shape change" means the deformation of a cell

without considering internal motion as if the lattice pattern is smoothed out. We may check the configurations in Fig.3.1 where (a)-(b)-(c) is the correct mapping process. From (a) to (b), internal pattern of atomic distribution  $\mathbf{s}_i$  changes but the overall shape is still the same. With  $\boldsymbol{\phi}_{\alpha}$ , (b) is mapped to (c), which is merely the shape change. In another possible undesired path (a)-(d)-(c), where shape of  $\mathbf{s}_i^*$  space is different from that of initial  $\mathbf{s}_i^0$  space, the operator  $\boldsymbol{\phi}_{\alpha}^*$  is not the true deformation gradient for the cell anymore, which results in misinterpretation.



Figure 3.1: Possible misintepretation

To resolve this issue, we compare the configurations (b) and (d) in Fig.3.1, and introduce an operator  $\psi_{\alpha}$  which gives the mapping,

$$\mathbf{s}_i^* = \boldsymbol{\psi}_{\alpha} \cdot \mathbf{s}_i \tag{3.4}$$

then we have two equivalent paths from (b) to (c), i.e.  $\phi_{\alpha} = \phi_{\alpha}^* \cdot \psi_{\alpha}$ , and the mapping process of deformation can be further decomposed to,

$$\boldsymbol{\phi}_{\alpha} \cdot \mathbf{s}_{i} = \boldsymbol{\phi}_{\alpha}^{*} \cdot \boldsymbol{\psi}_{\alpha} \cdot \boldsymbol{\psi}_{\alpha}^{-1} \cdot \mathbf{s}_{i}^{*}$$
(3.5)

Thus, as long as we find the operator  $\psi_{\alpha}$ , we may pull  $\phi_{\alpha}^*$  back to the desired  $\phi_{\alpha}$ . As an example, we may keep track of three critical boundary particles  $\mathbf{s}_1$ ,  $\mathbf{s}_2$  and  $\mathbf{s}_3$  (e.g. particles around vertices) which are not in a same plane. We assume that the shape changes uniformly during the simulation as we shall discuss later in this section, and  $\mathbf{s}_1(t) \approx \mathbf{s}_1^0$ ,  $\mathbf{s}_2(t) \approx \mathbf{s}_2^0$ ,  $\mathbf{s}_3(t) \approx \mathbf{s}_3^0$  for those critical particles. Therefore,

$$[\mathbf{s}_1^* \ \mathbf{s}_2^* \ \mathbf{s}_3^*] = \boldsymbol{\psi}_{\alpha} \cdot [\mathbf{s}_1 \ \mathbf{s}_2 \ \mathbf{s}_3] \approx \boldsymbol{\psi}_{\alpha} \cdot [\mathbf{s}_1^0 \ \mathbf{s}_2^0 \ \mathbf{s}_3^0]$$
(3.6)

Since  $\mathbf{s}_1^0$ ,  $\mathbf{s}_2^0$  and  $\mathbf{s}_3^0$  are not in a same plane, we can reverse them to obtain  $\boldsymbol{\psi}_{\alpha}$ ,

$$\boldsymbol{\psi}_{\alpha} \approx [\mathbf{s}_{1}^{*} \ \mathbf{s}_{2}^{*} \ \mathbf{s}_{3}^{*}] \cdot [\mathbf{s}_{1}^{0} \ \mathbf{s}_{2}^{0} \ \mathbf{s}_{3}^{0}]^{-1}$$
(3.7)

To obtain more reliable result, we may choose a group of atoms around  $\mathbf{s}_1$ ,  $\mathbf{s}_2$  and  $\mathbf{s}_3$ , and calculate the average value. This criteria is satisfied for arbitrary shapes of cells. The constraint can be expressed as,

$$\mathbf{G}_{2} = [\bar{\mathbf{s}}_{1}^{*}(t) \ \bar{\mathbf{s}}_{2}^{*}(t) \ \bar{\mathbf{s}}_{3}^{*}(t)] - [\bar{\mathbf{s}}_{1}^{0} \ \bar{\mathbf{s}}_{2}^{0} \ \bar{\mathbf{s}}_{3}^{0}] = 0$$
(3.8)

Each quantity in the above equation is the average value of a cluster of atoms.

To enforce  $\mathbf{G}_1$ , we may employ traditional approaches such as Lagrangian multiplier method [66, 67, 68]. The extended Lagrangian for Eq. (2.91) is,

$$\mathcal{L}_{\alpha}^{*} = \mathcal{L}_{\alpha} - \boldsymbol{\lambda} \cdot \mathbf{G}_{1} \tag{3.9}$$

where Lagrangian multiplier  $\lambda$  is a vector. The constrained equation of motion for  $\mathbf{s}_i$  is,

$$\frac{d}{dt}\frac{\partial \mathcal{L}_{\alpha}^{*}}{\partial \dot{\mathbf{s}}_{i}} - \frac{\partial \mathcal{L}_{\alpha}^{*}}{\partial \mathbf{s}_{i}} = 0$$
(3.10)

Since we know that  $\partial \mathbf{G}_1 / \partial \mathbf{s}_i = m_i \mathbf{I}$ , then, from equation (3.10), we have

$$m_i \mathbf{C}_{\alpha} \cdot \ddot{\mathbf{s}}_i = \sum_j \mathbf{f}_{ij} \cdot \boldsymbol{\phi}_{\alpha} - m_i \dot{\mathbf{C}}_{\alpha} \cdot \dot{\mathbf{s}}_i - m_i \boldsymbol{\lambda}$$
(3.11)

By performing iterative schemes such as SHAKE algorithm [69, 70],  $\mathbf{G}_1$  can be restricted in a given tolerance. The procedure to enforce  $\mathbf{G}_2$  is similar.

On the other hand, if we exam the problem from a different perspective, we may have better choice to enforce the constraints. Note that the above constraints are in terms of  $\mathbf{s}_i$ , but the combination of  $\mathbf{r}_{\alpha}$ ,  $\boldsymbol{\phi}_{\alpha}$  and  $\mathbf{s}_i$  is  $\mathbf{r}_i$  which is not restricted. Therefore, we can convert the constraint conditions to  $\mathbf{r}_{\alpha}$  and  $\boldsymbol{\phi}_{\alpha}$ . The procedure is briefly stated as follow. Step 1, for conservation of the centers of mass, we first assemble real atomistic position  $\mathbf{r}_i$  by equation (3.1), then calculate the correct center of mass by  $\mathbf{r}_{\alpha} = \sum_i m_i \mathbf{r}_i / \sum_i m_i$ . Step 2, for conservation of deformation gradient, we calculate the operator  $\boldsymbol{\psi}_{\alpha}$  by Eq. (3.7), then perform the operation in (3.5) to obtain the desired deformation gradient  $\boldsymbol{\phi}_{\alpha}$ . We may perform step 1 and 2 once in every several time steps. This approach is more straightforward and efficient than Lagrangian Multiplier method.

Another issue is the size dependency of supercells. Intuitively, the size of cells cannot be too big or too small. If it is too small, statistical assumptions may not be guaranteed because certain amount of particles are required to ensure it statistically meaningful. So that is a lower bound of cell sizes. On the other hand, we use a unique deformation gradient to track the shapes of supercells. Therefore, cells cannot be too big to include non-unique distortion. That is the upper bound for cell sizes. Typically, the bound may range from a nanometer to hundred nanometers or even bigger, which depends on the problem of interest. We need to perform tests to study the influence of size when dealing with specific problems. For inhomogeneous models or problems including defects, the size should be relatively small. However, the chosen of the upper bound is not a strict requirement. In fact, even if the cell deformation is not unique due to the large sizes, the real atomistic position  $\mathbf{r}_i$  is not influenced as we discussed for constraints. Misinterpretation of deformation gradient  $\phi_{\alpha}$  will, at most, make the calculated stress and strain unreliable and the picture of  $\phi_{\alpha}$  ambiguous. For boundary conditions as traction or displacement, there is no such confusion because we can always keep track of the centers of mass for different cells, no matter how the shape distorts. In this case, we can neglect the physical picture of  $\phi_{\alpha}$ . The theory is still valid. Therefore, the multiscale structure is actually adaptive to inhomogeneity and defects even with big cells.

### **3.2** Time Integrators

Derived from fundamental molecular dynamics, the multiscale model has the same dynamical properties. Thus, we employ integrators that are popularly used in classical molecular dynamics which meet some certain criteria.

Molecular simulation usually involves large quantity of time steps to solve for a system with large quantity of particles. The goal is not to reflect the trajectory in a highly accurate manner although efficiency is important for an integrator as well as local accuracy. For a large chaotic molecular system, we put more emphasis on how well it preserves the statistical properties of the system, such as energy, momentum and symplecticity.

An symplectic integrator conserves the volume in phase space along the trajectory. That is an important property for Hamiltonian system to ensure the long-time stableness of energy. Figure 3.2 demonstrates the difference between symplectic and nonsymplectic algorithms. Assume the exact solution is on a circle with unit radius which is marked as red dashed lines. The real time evolution simulated by different integrators are in blue solid lines. The symplectic integrator in Fig.3.2(a) keeps long term stable for the shape even though there might be big local error. On the other hand, nonsymplectic integrator in Fig.3.2(b) may restrict the local error in a short time, but as time goes by, it tends to collapse the solution gradually. Thus, the first important requirement of integrators in molecular dynamical simulation is symplecticity.

Secondly, since force evaluation is the most time consuming part in molecular computation due to the double summation of large quantity of particles, a good integrator should involve as few force calculation as possible, preferably once in each step. Therefore, multistage methods such as Runge-Kutta method is not appropriate for molecular systems.

Widely used integrators in molecular dynamics include Velocity Verlet[71], which has second order accuracy, and Predictor-Corrector[72]. Velocity Verlet is symplectic, time reversible and straightforward in practice, which make it one of the most widely used algorithms in molecular dynamics. Predictor-Corrector is not unconditionally symplectic, but under



Figure 3.2: Demonstration of symplecticity. Red dashed lines represent exact solutions, and blue solid lines are results of simulations by different integrators. (a) Symplectic integrator has a long term stability even though large local errors can be found; (b)Nonsymplectic integrator may result in small local errors, but can collapse the solution in the long run.

certain conditions such as short time steps, it has property of conservation. The advantage is the order of accuracy can be controlled by choosing number of derivatives. Predictor-Corrector is a good compensation in some cases that are difficult for Velocity Verlet.

#### Velocity Verlet for Integrating $r_{\alpha}$

We employ Velocity Verlet[71] in calculating macroscale motion  $\mathbf{r}_{\alpha}$ . The order of updating time steps is as follows,

$$\mathbf{r}_{\alpha}^{n+1} = \mathbf{r}_{\alpha}^{n} + \dot{\mathbf{r}}_{\alpha}^{n} \Delta t_{\mathbf{r}_{\alpha}} + \frac{1}{2} \ddot{\mathbf{r}}_{\alpha}^{n} \Delta t_{\mathbf{r}_{\alpha}}^{2}$$
(3.12)

$$\rightarrow Evaluate \ \ddot{\mathbf{r}}^{n+1}_{\alpha}$$

$$\dot{\mathbf{r}}_{\alpha}^{n+1} = \dot{\mathbf{r}}_{\alpha}^{n} + \frac{1}{2} \left( \ddot{\mathbf{r}}_{\alpha}^{n} \Delta t_{\mathbf{r}_{\alpha}} + \ddot{\mathbf{r}}_{\alpha}^{n+1} \Delta t_{\mathbf{r}_{\alpha}} \right)$$
(3.13)

where  $\Delta t_{\mathbf{r}_{\alpha}}$  is the step size of coarse scale.

The intermediate step of evaluating acceleration is where we need to obtain information from  $\phi_{\alpha}$  and  $\mathbf{s}_i$  to assemble  $\mathbf{r}_i$  according to Eq. (2.113). We may choose serial or parallel algorithm for information exchange which will be presented in next section.

#### Predictor-Corrector for Integrating $\phi_{\alpha}$ and $s_i$

For  $\phi_{\alpha}$  and  $\mathbf{s}_i$ , as observed in equations (2.114)~(2.115), the accelerations depend on velocities which are not determined in the step of evaluation if we use Velocity Verlet. Therefore, six order Predictor-Corrector Algorithm[72] is selected for calculating the motions of  $\phi_{\alpha}$  and  $\mathbf{s}_i$ . Define scale time derivatives as,

$$\phi_{0} = \phi_{\alpha}; \quad \phi_{1} = h \frac{d\phi_{\alpha}}{dt}; \quad \phi_{2} = \frac{h^{2}}{2} \frac{d^{2}\phi_{\alpha}}{dt^{2}}; \quad \phi_{3} = \frac{h^{3}}{6} \frac{d^{3}\phi_{\alpha}}{dt^{3}}; \quad \phi_{4} = \frac{h^{4}}{24} \frac{d^{4}\phi_{\alpha}}{dt^{4}}; \quad \phi_{5} = \frac{h^{5}}{120} \frac{d^{5}\phi_{\alpha}}{dt^{5}}; \\ \mathbf{s}_{0} = \mathbf{s}_{i}; \quad \mathbf{s}_{1} = k \frac{d\mathbf{s}_{i}}{dt}; \quad \mathbf{s}_{2} = \frac{k^{2}}{2} \frac{d^{2}\mathbf{s}_{i}}{dt^{2}}; \quad \mathbf{s}_{3} = \frac{k^{3}}{6} \frac{d^{3}\mathbf{s}_{i}}{dt^{3}}; \quad \mathbf{s}_{4} = \frac{k^{4}}{24} \frac{d^{4}\mathbf{s}_{i}}{dt^{4}}; \quad \mathbf{s}_{5} = \frac{k^{5}}{120} \frac{d^{5}\mathbf{s}_{i}}{dt^{5}}; \quad (3.14)$$

where  $h = \Delta t_{\phi_{\alpha}}$ ,  $k = \Delta t_{s_i}$  are step sizes for  $\phi_{\alpha}$  and  $\mathbf{s}_i$  scales, respectively. Arrange them in matrix forms, i.e.  $\mathbf{\Phi} = [\phi_0 \ \phi_1 \ \phi_2 \ \phi_3 \ \phi_4 \ \phi_5]^T$ ,  $\mathbf{S} = [\mathbf{s}_0 \ \mathbf{s}_1 \ \mathbf{s}_2 \ \mathbf{s}_3 \ \mathbf{s}_4 \ \mathbf{s}_5]^T$ , then the predictors are,

$$\begin{aligned} \Phi_p^{n+1} &= \mathbf{C}_1 \cdot \Phi^n \\ \mathbf{S}_p^{n+1} &= \mathbf{C}_1 \cdot \mathbf{S}^n \end{aligned}$$
 (3.15)

where subscript "p" represents the predictor, superscript denotes time steps, and constant matrix  $\mathbf{C}_1$  is,

$$\mathbf{C}_{1} = \begin{bmatrix} 1 & 1 & 1 & 1 & 1 & 1 \\ 0 & 1 & 2 & 3 & 4 & 5 \\ 0 & 0 & 1 & 3 & 6 & 10 \\ 0 & 0 & 0 & 1 & 4 & 10 \\ 0 & 0 & 0 & 0 & 1 & 5 \\ 0 & 0 & 0 & 0 & 0 & 1 \end{bmatrix}$$
(3.16)

Evaluate new accelerations  $\ddot{\phi}_{\alpha}^{n+1}$  and  $\ddot{\mathbf{s}}_{i}^{n+1}$  based on predicted positions and velocities and equations (2.114) ~ (2.115). Calculate the error between new accelerations and quantities in (3.15), i.e.

$$\Delta \mathbf{a}_{\phi_{\alpha}} = (\phi_2)_p^{n+1} - \ddot{\phi}_{\alpha}^{n+1}$$
  
$$\Delta \mathbf{a}_{s_i} = (\mathbf{s}_2)_p^{n+1} - \ddot{\mathbf{s}}_{\alpha}^{n+1}$$
(3.17)

Then perform the Corrector step,

which are quantities to be used in n + 1 step. The constants  $C_2$  are

$$\mathbf{C}_2 = \begin{bmatrix} \frac{3}{16} & \frac{251}{360} & 1 & \frac{11}{18} & \frac{1}{6} & \frac{1}{60} \end{bmatrix}^T$$
(3.19)

### 3.3 Numerical Schemes

As observed in dynamics equations  $(2.113)\sim(2.115)$ , quantities  $\mathbf{r}_{\alpha}$ ,  $\boldsymbol{\phi}_{\alpha}$  and  $\mathbf{s}_{i}$  of multiple spacial and time scales in a same framework are strongly coupled and require intensive message exchange among them. A proper multiscale integration scheme is necessary to aid the communication. Step size of integration for each quantity should be chosen to reflect the time scale variation, i.e.  $\Delta t_{\mathbf{r}_{\alpha}} > \Delta t_{\boldsymbol{\phi}_{\alpha}} \gg \Delta t_{\mathbf{s}_{i}}$ . The procedure of information exchange among scales is similar to the scheme developed for fluid-structure interaction simulations in [73], where time step used in fluid is a small fraction of that in solid. We employ the following coupling algorithms in the multiscale integration with serial and parallel approaches, respectively.



Figure 3.3: Serial algorithm for time evolution.

The serial computation algorithm is shown in Fig.3.3.  $\mathbf{r}_{\alpha}$ ,  $\phi_{\alpha}$  and  $\mathbf{s}_{i}$  evolve with different time scales and step sizes. (1)~(4) and (5)~(8) are two subcycles in each updating loop. For subcycle (1)~(4), calculation of  $\phi_{\alpha}$  is advanced by  $\Delta t_{\phi_{\alpha}}$  for several substeps, and pass information back to macroscale for a single step  $\mathbf{r}_{\alpha}$  calculation. Same loop is implemented for subcycle (5)~(8), where  $\mathbf{s}_{i}$  is advanced for several substeps and pass information back for each  $\phi_{\alpha}$  step forward. In serial scheme, when one scale is evolving, quantities in other scales are standing still as "frozen". For parallel scheme,  $\mathbf{r}_{\alpha}$ ,  $\phi_{\alpha}$  and  $\mathbf{s}_{i}$  advance simultaneously as shown in Fig.3.4. The subcycles are observed in (1)~(3) and (4)~(6). On the common starting point of a substep in each scale, information is exchanged among the quantities at the same time instance without lead or lag. Usually, serial algorithm is preferred because parallel algorithm requires smaller step sizes in order to be numerically stable and sufficiently



Figure 3.4: Parallel algorithm

accurate[73]. But parallel algorithm can be naturally incorporated with parallel computing for multiple CPU cores without additional treatment, which is convenient in some cases.

### 3.4 Temperature Control

In this work, we focus the mechanical boundary conditions. All simulations are under controlled temperature, i.e. heat flow is not considered. We leave the heat conduction as a future work. The procedure of temperature control in the multiscale model is similar as classical Molecular Dynamics. A variety of thermostat are invented in classical Molecular Dynamics, including Anderson thermostat[74], Berendsen thermostat[75, 76], Nosé-Hoover thermostat[77, 78, 79], and Langevin thermostat[80, 81], etc. Here we employ the popular Nosé-Hoover thermostat in our model to control the microscale motion. We develop basic formulation in this section.

As stated by Nosé [77], a new variable w is introduced to exchange heat with the system by scaling velocities. w is interpreted as a time scaling variable as  $\Delta t = \Delta t'/w$ , where  $\Delta t$  is real time step and  $\Delta t'$  is virtual time step. Since higher-scale velocity varies much slower than microscale, as we discussed in last section, real velocity can be approximated as  $\dot{\mathbf{r}}_i = \boldsymbol{\phi}_{\alpha} \cdot \dot{\mathbf{s}}_i$ . Therefore, heat transfer primarily influence the velocity of microscale  $\dot{\mathbf{s}}_i$ . The extended Lagrangian is,

$$\mathcal{L}'_{\alpha} = \frac{1}{2} M_{\alpha} \dot{\boldsymbol{r}}_{\alpha} \cdot \dot{\boldsymbol{r}}_{\alpha} + \frac{1}{2} \dot{\boldsymbol{\phi}}_{\alpha}^{T} \dot{\boldsymbol{\phi}}_{\alpha} : \mathbf{J}_{\alpha} + \frac{1}{2} \mathbf{C}_{\alpha} : \sum_{i} m_{i} w^{2} \frac{d\mathbf{s}'_{i}}{dt'} \otimes \frac{d\mathbf{s}'_{i}}{dt'} - V_{\alpha} + \frac{Q}{2} \left(\frac{dw}{dt'}\right)^{2} - gk_{B}T lnw \qquad (3.20)$$

where Q is the effective mass of w, g is the number of total degrees of freedom,  $k_B$  is Boltzmann constant, T is the desired temperature. Last two terms are kinetic energy and potential energy for w. Primes denote the quantities and derivatives in virtual time axis. Dots and double dots represent the time derivatives in real axis. Equations of motion for  $\mathbf{r}_{\alpha}$ and  $\phi_{\alpha}$  are the same as in (2.113)~(2.114). Dynamic equations of  $\mathbf{s}'_i$  and w are derived in virtual time axis,

$$\mathbf{C}_{\alpha} \cdot \frac{d^2 \mathbf{s}'_i}{dt'^2} = \frac{1}{m_i w^2} \sum_j \mathbf{f}_{ij} \cdot \boldsymbol{\phi}_{\alpha} - \frac{d \mathbf{C}_{\alpha}}{dt'} \cdot \frac{d \mathbf{s}'_i}{dt'} - 2\frac{1}{w} \frac{d w}{dt'} \mathbf{C}_{\alpha} \cdot \frac{d \mathbf{s}'_i}{dt'}$$
(3.21)

$$Q\frac{d^2w}{dt'^2} = \mathbf{C}_{\alpha} : \sum_{i} m_i w \frac{d\mathbf{s}'_i}{dt'} \otimes \frac{d\mathbf{s}'_i}{dt'} - \frac{gk_BT}{w}$$
(3.22)

With relations  $\Delta t = \Delta t'/w$  and  $\mathbf{s}_i = \mathbf{s}'_i$ , we may convert the dynamic equations back to real time axis,

$$\mathbf{C}_{\alpha} \cdot \ddot{\mathbf{s}}_{i} = \frac{1}{m_{i}} \sum_{j} \mathbf{f}_{ij} \cdot \boldsymbol{\phi}_{\alpha} - \dot{\mathbf{C}}_{\alpha} \cdot \dot{\mathbf{s}}_{i} - 2\frac{\dot{w}}{w} \mathbf{C}_{\alpha} \cdot \dot{\mathbf{s}}_{i}$$
(3.23)

$$Q\ddot{w} = \mathbf{C}_{\alpha} : \sum_{i} m_{i} w \mathbf{\dot{s}}_{i} \otimes \mathbf{\dot{s}}_{i} - g k_{B} T w$$
 (3.24)

With Hoover's modification [79] by introducing a new variable  $\zeta$  as,

$$\zeta = \frac{dw}{dt'} = \frac{\dot{w}}{w}, \quad \dot{\zeta} = \frac{\ddot{w}}{w} \tag{3.25}$$

The real-time equations of motion become,

$$\mathbf{C}_{\alpha} \cdot \ddot{\mathbf{s}}_{i} = \frac{1}{m_{i}} \sum_{j} \mathbf{f}_{ij} \cdot \boldsymbol{\phi}_{\alpha} - \dot{\mathbf{C}}_{\alpha} \cdot \dot{\mathbf{s}}_{i} - 2\zeta \mathbf{C}_{\alpha} \cdot \dot{\mathbf{s}}_{i}$$
(3.26)

$$Q\dot{\zeta} = \mathbf{C}_{\alpha} : \sum_{i} m_{i} \dot{\mathbf{s}}_{i} \otimes \dot{\mathbf{s}}_{i} - gk_{B}T$$
(3.27)

Equations (3.26) and (3.27) are used in scaling the microscale quantity for temperature control.

### Chapter 4

### Validation of the Multiscale Theory

In this chapter, we present three examples of phase transition with displacement and traction boundary conditions, respectively. Dimensions of the models used in these examples are small, typically within the range of several nanometers, or tens of unit cells of crystal lattice. The purpose of these examples is to demonstrate the multiscale model by applying macroscale boundary conditions in molecular systems, rather than solving realistic large scale problems.

### 4.1 Bulk Lattice with Displacement Boundary Condition

The first example is devised to investigate the procedure of phase transit under displacement loading, e.g. uniaxial stretch boundary condition. The model is a finite size bulk Nickel with  $9 \times 9 \times 9$  unit cells of Face Centered Cubic (F.C.C.) lattice which is shown in Fig. 4.1(a). The bulk has in total 729 F.C.C. unit cells and 2916 atoms according to basic crystallography. The lattice constant is  $a_0 = 0.352$  nm at room temperature, with which the Nickel crystal is in a stress free state for infinite lattice. Atomic weight for Nickel is 58.69 u. The bulk metal is divided to  $3 \times 3 \times 3$  supercells, and each super cell has  $3 \times 3 \times 3$  unit cells. The separation ensures that at least one internal supercell is not exposed to boundaries. We may use this cell for benchmark test comparing with known results in infinite lattice. The interaction between atoms is modeled by Morse potential [82, 83]. It has the form of

$$\varphi(r) = D(e^{-2\alpha(r-r_0)} - 2e^{-\alpha(r-r_0)})$$
(4.1)

The pair force is given by

$$f(r) = -\frac{\partial\phi(r)}{\partial r} = 2D\alpha(-e^{-2\alpha(r-r_0)} + e^{-\alpha(r-r_0)})$$

$$\tag{4.2}$$

With the constants  $D = 3.5059 \times 10^{-20}$  J,  $\alpha = 8.766/a_0$  and  $r_0 = 0.71727 \text{\AA}$ . The parameters of the model and Morse potential is listed in Table4.1.



Figure 4.1: The model setup and the procedure of structure change under uniaxial displacement loading in [010] direction. The system consists of  $3 \times 3 \times 3$  supercells, and Each supercell consists of  $3 \times 3 \times 3$  unit cells. We use different color to distinguish supercells.

Material parameters		Morse parameters	
Material	Ni	D	$3.5059 \times 10^{-20} \text{ J}$
Initial structure	F.C.C.	α	$8.766/a_0$
Lattice constant	0.325  nm	$r_0$	$0.71727  m \AA$
Atomic weight	58.69 u	Step size	s
Model dimensions		$\Delta r_{lpha}$	$0.0015~\mathrm{ps}$
Supercell	$3 \times 3 \times 3$ unit cells	$\Delta_{\phi_{\alpha}}$	$0.0012~\mathrm{ps}$
System	$3 \times 3 \times 3$ supercells	$\Delta_{\boldsymbol{s}_i}$	$0.00015~\mathrm{ps}$

Table 4.1: Parameters of the bulk model and the Morse potential

During the entire simulation, the temperature is controlled around  $350^{\circ}K$  by using Nosé-Hoover thermostat on microscale motion as formulated in last chapter. Before loading, random perturbation of velocities were assigned to get the desired temperature initially. A 5000-steps run with free boundary is conducted to get the optimal initial configuration. Subsequently, we apply uniaxial compressive stretches incrementally on y direction which is [010] in F.C.C. lattice as shown in Fig. 4.1(a). The stretch is realized by moving close the centers of mass for top and bottom cells, and then allow a relaxation time. The compress-relax procedure gives a certain strain rate. Integration time steps used in simulation are 0.0015 ps, 0.0012 ps and 0.00015 ps for different scales. We relax the system for 2000 macro steps for each incremental stretch of 0.03.

Fig.4.1(b)~(d) show the snapshots of structural evolution under different stretches with lateral and top views. When the stretch is relatively small, e.g.  $\lambda = 0.92$ , the Ni bulk goes through simply elastic deformation. The pattern of particle distribution are still uniform. From both top view and lateral view in Fig.4.1(b), we do not observe relative displacements between any adjacent planes. When the stretch increases, e.g.  $\lambda = 0.88$ , some sort of interplanar slip is activated between the planes of {001} accompanied by the elastic in-plane deformation. As we observe from the top view in Fig.4.1(c), the shape is changed from square to parallelogram due to the slip between {001} planes. But the structure at this point is a little bit irregular and unstable. Further increase the stretch, e.g.  $\lambda = 0.77$ , the lateral planes {001} get more regular patterns as in planes {0001} of Hexagonal Closely Packed (H.C.P.) structure. The final configuration in Fig.4.1(e) with  $\lambda = 0.72$  is the new stable H.C.P. structure. The lattice constant turns into  $a_1 = 0.252$  nm which is approximately  $\sqrt{2}/2$  times  $a_0$  and is the same as atomic bond in original F.C.C. configuration.

We notice that the final H.C.P. configuration of the whole model is regular without boundary distortion, which is different from the results of traditional Molecular Dynamics. If the displacements are applied on boundary atoms as in Molecular Dynamics, the structural pattern near the boundary area will be irregular because those atoms are not free to search the

optimal positions. While prescribing the displacements on supercells relaxes the constraints for specific atoms. Average displacements are enforced as a form of overall rigid body translations for the prescribed cells, while the internal motions are still allowed. This example shows the advantage of the top-down multiscale model in the case of applying macroscale boundary conditions.

We studied the stress-strain relation as a benchmark test to compare with the theoretical prediction of infinite lattice as reported in [1]. For the model consist of  $3\times3\times3$  supercells, we expect the internal cell is more suitable to approximate the environment of the infinite lattice than boundary cells. We plotted the loading curve from the initial configuration to a stretch around  $\lambda = 0.7$ . As shown in Fig.4.2, the solid curve is the theoretical result, and the data collected from the internal cell during the simulation is marked by error bar. The data is obtained by averaging three calculations under similar situations to obtain more reliable results. Overall, the simulation agrees well with the theoretical result. However, in the range of [0.75 0.9], large deviation is observed between the curves. This is the transition area between two structures if we check the energy landscape in [1]. The structure is unstable in this range and an macroscale equilibrium is hard to reach. Therefore, the measure of stresses is meaningless. In ranges [0.9 1.0] and [0.7 0.75], the results have better agreements with theoretical result. Because the F.C.C. structure goes through elastic deformation in [0.9 1.0], and when a stable H.C.P. configuration reaches in the stretch range [0.7 0.75], the deformation is also elastic.

We plotted the stresses at  $\lambda_y = 0.73$  and  $\lambda_y = 0.97$  for different sizes of suppercells in Fig. 4.3. These two stretches are in elastic range of H.C.P. and F.C.C., separately. Thus they should be suitable to study the stress states. The size of the supercell ranges from  $2 \times 2 \times 2$  to  $7 \times 7 \times 7$  in unit cells. In Fig. 4.3, we observe no significant deviation when the supercell is larger than 3 unit-cell cubic. Sizes of supercells in this range should not influence the results. For problems such as elastic deformation and homogenous phase transition in this example, we expect to have satisfactory results up to much larger size of supercells.

### 4.2 Bulk Lattice with Traction Boundary Condition

As a second example, we studied the dynamic response and phase transition of the same Nickel bulk under a constant surface traction. The initial model is shown in Fig.4.6 (a). Same as before, the structure is Face Centered Cubic with  $3 \times 3 \times 3$  supercells, and each super cell has  $3 \times 3 \times 3$  unit cells. Total number of atoms is 2916. The weight of each Nickel atom is 58.69 u. The lattice constant a = 0.352 nm, and the volume of the bulk is about 31.8 nm<sup>3</sup>. The orientation of the faces are [100], [010] and [001], which correspond to x, y and z coordinates, separately. During the entire procedure, the temperature was controlled around 350 K. We use the same Morse potential and integration algorithm as in the displacement example. The integration time steps used in this simulation are 0.0015 ps, 0.0012 ps and 0.00015 ps for different scales.

At the beginning of the calculation, the bulk is relaxed in a stress free state for 5000 steps



Figure 4.2: Stress-strain relation under uniaxial displacement loading. The data is compared with the theoretical prediction by Milstein and Farber [1]

to obtain the energy-minimizing configuration. The volume change of the bulk is less than 1% compared to the initial model. We then apply a compressive traction  $\bar{\mathbf{t}} = 6$  GPa on the top and bottom surfaces of the bulk as shown in Fig.4.6 (a). The traction is a dead load throughout the simulation in [100] direction. Fig.4.6(b)~(d) show the snapshots of the loading history. At the beginning, i.e. t < 2 ps, the model is in a state of linear elastic deformation as observed in Fig.4.6(b), where particles are stretched uniformly, and the structure stays nearly in F.C.C. Subsequently, phase transformation initiates at the top and bottom surfaces where the tractions are applied, and propagates quickly to the center, as shown in Fig.4.6(c)-(d). After about t = 9 ps, the entire bulk turns into the H.C.P. phase. Based on morphology of the final equilibrium configuration shown in Fig.4.6(e), the original {001} planes in F.C.C. switch to {0001} closely packed planes in H.C.P. The phase transformation resulted in the new lattice constants a = 0.25 nm and c = 0.41 nm for the H.C.P. structure which is the same as in the example of displacement loading. And the structure is regular without boundary distortion.

An observation is the overall shape of the final configuration. Although the new configurations are standard H.C.P. structures for both displacement and traction loading, a slight difference between the shapes of the bulk can be found. In the example of displacement boundary condition, the final H.C.P. bulk is a regular parallelepiped as in Fig.4.1(c). When



Figure 4.3: Stress  $\sigma_{yy}$  for different sizes of supercells at stretches  $\lambda_y = 0.73$  and  $\lambda_y = 0.97$ . Horizontal lines are theoretical results in [1] as benchmarks.

the traction boundary condition is applied, the bulk turns out to be a zigzag shape which can be observed in Fig.4.6(e). This is a result of different orientations of interplanar slips. Theoretically, interplanar slip has no preference on directions. Either way is possible. In practice, many factors can contribute to the difference, e.g. initial imperfection of the geometry, potential energy, integration time steps, etc.

We also analyzed the traction-stretch relation for the model and plotted the result in Fig.4.8. As the compressive surface traction increases, the stretch decreases following the curve in Path I. At some certain point around 5 GPa, the curve jumps to another path which is marked by red circles. These two paths are almost straight which reflect the linear elastic deformation of the two distinct structures of F.C.C. and H.C.P. as we discussed previously. As we unload the traction gradually from the new H.C.P. structure(Path II), the curve goes straight up(Path III) without jumping back to the original F.C.C. Apparently the new H.C.P. bulk is stable under further loading/unloading. Different from displacement loading, the stress-strain curve with traction boundary has a break point because the model jumps over the unstable configurations. However, if displacement is controlled, the model still passes through unstable configurations between F.C.C. and H.C.P., but the measure of stress is meaningless. We can approximate the Young's modulus based on the curves in Fig.4.8.  $E_1$  is the Young's modulus for the initial F.C.C. structure on [010] direction, which is in the



Figure 4.4: Initial(a), final(e) configuration of the model when a constant surface traction of  $\bar{\mathbf{t}} = 5$  GPa is symmetrically applied on its top and bottom surfaces, and snapshots(b)~(d) of the propagation of phase transition. Different colors are used to distinguish different cells.

range of 150~220 GPa.  $E_2$  and  $E_3$  are measured for the H.C.P. structure on [1120] direction for loading and unloading procedures, respectively. They are in the range of 300~500 GPa.



Figure 4.5: Traction-stretch curve of the loading-unloading processes in equilibrium state.

### 4.3 Nanowire with Traction Boundary Condition

As a third example, we have studied dynamical responses and phase transition of a Nickel nanowire under constant traction with the proposed multiscale model. Mechanical behaviors of bulk Ni such as stress-strain relations[1], phase transition[59], etc. were extensively studied. Finite diameter nanowire, which is different from bulk metal, has mechanical properties largely depending on the surface-volume ratio, surface energy, crystallographic orientation, etc., and it has attracted much attention in the past decade, e.g. [84, 85, 86, 87, 88, 89]. The multiscale model enables us to describe the behavior of a nanowire when the macroscale traction is applied to the end boundary of the Nickel nanowire, which will be discussed subsequently.

The simulation model of the Nickel nanowire is divided into  $5 \times 1 \times 1$  supercells, and each supercell has  $5 \times 5 \times 5$  unit cells. As is shown in Fig.4.6 (a), different colors were used to distinguish the five supercells. The nanowire consists 2500 atoms in total. Atomic positions were generated as Face Centered Cubic structure according to metal crystallography. The



Figure 4.6: Initial(a), final(e) configuration of the nanowire when a constant surface traction of  $\bar{\mathbf{t}} = 1.66$  GPa is symmetrically applied on its both ends, and snapshots(b)~(d) of the propagation of phase transition. Gradient colors are used to distinguish cells.

lattice constant a = 0.352 nm which makes the length of the Ni nanowire 8.8 nm, and the cross section of the nanowire is  $1.58 \times 1.58 nm^2$ . The axial orientation of the wire is <100> along  $x_1(x)$ -coordinate, and lateral directions <010> and <001> correspond to  $x_2(y)$  and  $x_3(z)$  coordinates. Same as bulk lattice, the nanowire is modeled using the pairwise Morse potential with the parameters fitting to the lattice constant and elastic constants. During the entire procedure, the temperature is controlled around 350 K by standard thermostat technique in Molecular Dynamics. The integration time steps used in simulation are 0.0015 ps, 0.0012 ps and 0.00015 ps for each scale.

At the beginning of the calculation, the nanowire is relaxed in a stress free state for 5000 steps in order to reach a minimum energy configuration. The change of wire length and the change of the cross section area are less than 1% comparing to the initial configuration. We then apply compressive traction  $\bar{\mathbf{t}} = 1.66$  GPa on both ends of the nanowire as shown in Fig.4.6 (a). The traction remains constant throughout the simulation in <100> direction.

Fig.4.6(b)~(d) show a series snapshots of the loading history. At the beginning, when t < 3 ps, an elastic wave propagates through the nanowire, and the entire structure is in a state of linear elastic deformation with non-uniform stretches along the wire as observed in Fig.4.6(b). However, during this stage, the entire nanowire almost stays in F.C.C. phase. Subsequently, phase transformation initiates at both ends, and it quickly propagates to the center, as shown in Fig.4.6(c)-(d). After t = 10 ps, the entire nanowire turns into the Hexagonal closely Packed phase. Based on the morphology of the final equilibrium configuration shown in Fig.4.6(e), the original {001} planes in F.C.C. switches to {0001} closely packed planes in H.C.P. phase. The phase transformation results lattice constant changes, and the new lattice constants a = 0.249 nm and c = 0.41 nm indicate the formation of a H.C.P. structure. To this end, the length of the nanowire changes to 6.25 nm that is 0.71 times the original length. Note that these values are averaged throughout the entire nanowire.



Figure 4.7: Convergence of normalized length  $(L/L_0)$  with time. For small traction, i.e.  $\bar{t}_x = 0.33$  GPa and  $\bar{t}_x = 0.67$  GPa, the nanowire went through oscillation but still in elastic limit of F.C.C. For bigger traction from  $\bar{t}_x = 0.83$  GPa to  $\bar{t}_x = 1.0$  GPa, the normalized length gradually converged to about 0.7 and rest in that new H.C.P. configuration. For even bigger traction, the nanowire experienced long oscillation as for small traction after a rapid phase transition.

We have recorded the evolution history of the nanowire length in time under the surface traction of different magnitudes. We plot the curves of the stretch $(L/L_0)$  for several com-

pressive loads  $\bar{t}_x$  in Fig.4.7. For  $\bar{t}_x = 0.33$  GPa and  $\bar{t}_x = 0.67$  GPa, the stretch oscillates for about 50 ps then gradually reaches an equilibrium state around 1. The periods of vibration are about 11 ps and 12 ps separately. Since the length does not change significantly, the lattice structure is still F.C.C. with elastic deformation, and the vibration of the curve reflects the elastic wave propagation. We also observe that when  $\bar{t}_x = 1.66$  GPa, the curve has a similar dynamic response of vibration with a period about 5 ps. But the final stretch of the nanowire in static state is around 0.7. Compare to the result discussed above, this is the stretch ratio between H.C.P. and F.C.C. lattices. Before the elastic vibration in the new H.C.P. configuration, the stretch approaches quickly to the value about 0.7 within 10 ps. This is the time frame for phase transition. If we examine Fig.4.6, the loading history shown is in the range of  $0\sim10$  ps. The curves at  $\bar{t}_x = 0.83$  GPa  $\bar{t}_x = 0.91$  GPa and  $\bar{t}_x = 1.0$  GPa reveal a critical gradual damping process. When the stretch reaches 0.7, the nanowire simply stopped vibration. The speed of convergence increases as the traction increases. The range around the above tractions is the critical domain that can activate the phase transition.

We have also calculated the traction-stretch relation for the nanowire, and we plot the result in Fig.4.8. As the compressive surface traction increases, the stretch (normalized length) decreases following the curve in Path I, until a critical value is reached at about 0.8 GPa. The structure then jumps to another curve which is marked as Path II. These two paths reflect the linear elastic deformations of the two distinct structures of F.C.C. and H.C.P. within their own ranges, and they are separated at a critical point. As we unload the traction gradually from the new H.C.P. structure (Path II), we find that the curve goes straight up (Path III) without turning back to the original F.C.C. structure. Apparently the new structure is stable under further loading/unloading. The stress-strain curve of nanowire is similar to that of bulk lattice. But they have different critical values due to the size effect of nanowire.



Figure 4.8: Traction-length curve for the nanowire in equilibrium state. The loading and unloading are all along the axial direction of the nanowire.
## Chapter 5

## Coupling Molecular Dynamics and Peridynamics

In this chapter, we further establish a computational paradigm based on the previous physical multiscale theory. The computational model separates a body into atomistic and macroscale regions with an interface. An adaptive multiscale element is constructed as a messenger to translate information between regions. In the atomistic region, molecular dynamics is adopted to simulate the motion of particles. In the macroscopic region, we have several choices depending on the problem of interest. For example, finite element method can be used to model general continuous field, and meshfree methods[90, 91, 92, 93, 94] are suitable for discrete materials. In this work, we employ peridynamics[95, 96, 97, 98, 99, 100] because of the similar dynamical structure as molecular systems. The non-local balance law provides a seamless connection to atomistic region. To solve the common issue of wave reflection in multiscale simulations, we introduce a filter on the interface. The procedure of filtering high-frequency wave is straightforward and adaptive without sophisticated mathematical treatment. With the established framework of cross-scale communication in the multiscale theory, mechanical quantities in both regions can be described and translated honestly. Technically, time scale separation provides more choices to achieve efficiency in simulations.

## 5.1 Multiscale Domain Decomposition

The multiscale model consists three parts: atomistic region, macro region and a transition zone responsible for translating information between two regions, as shown in Fig5.1(a). The atomistic region is described by classical molecular dynamics. Interatomic potential is used to derive nonlocal forces between atoms or molecules. Macro region usually requires empirical constitutive relation. Many methods can be chosen in this region depending on the problem of interest. In this work, we adopt peridynamics to be consistent with atomistic region due to their similar non-local behaviors, and the constitutive relation is based on Cauchy-Born rule. The formulation will be briefly introduced subsequently.



Figure 5.1: (a) The multiscale model consists three part: atomistic region, macro region and transition zone. The essential part is the transition zone, which is served as a messenger to translate information from both regions. A filter is constructed near the interface to solve the issue of high-frequency wave reflection. (b) The adaptive multiscale element in the transition zone. This element is an assemble of atoms which has macroscale properties such as shape and average displacement while the atomistic resolution is retained. The element is capable of carrying and translating information from different scales.

The essential part of the multiscale model is the transition zone. To ensure a reliable passage of information between macro and atomistic regions, several conditions need to be taken into account. First, in the bottom-up procedure, atomistic information such as force and displacement should be sensed by transition zone and interpreted to macro domain. Specifically, macroscale only capture low-frequency waves with coarse resolution, and highfrequency atomistic vibration should be filtered out otherwise it will reflect back to the atomistic region. Second, in the top-down procedure, macroscale information such as stress, strain and displacement should also be properly interpreted to atomistic domain. The previous multiscale theory proposed in chapter 2 is employed here to characterize the top-down approach. The basic unit "supercell" can carry both atomistic and macroscale information, which makes it a good candidate of transition element. In this computational model, we rename it as "adaptive multiscale element". We briefly review the theory and the properties of "supercell" in the following subsection.

## 5.2 Adaptive Multiscale Element

This section is arranged as a review and summary of essential formulas and basic ideas presented in chapter 2. As shown in Fig5.1(a), the transition zone still has atomistic resolution. However, we divide those atoms into several supercells. Each supercell is viewed as a material point on macroscale which has shape and average displacement. In addition, each atom inside the supercell is free to move as internal degree of freedom. The atomistic position  $\mathbf{r}_i(t)$  at current time is proposed as,

 $\mathbf{r}_{i}(t) = \mathbf{r}_{\alpha}(t) + \boldsymbol{\phi}_{\alpha}(t) \cdot \mathbf{s}_{i}(t)$ (5.1)

where  $\mathbf{r}_{\alpha}$  is the center of mass of  $\alpha$ -th cell calculated as,

$$\mathbf{r}_{\alpha} = \sum_{i} m_{i} \mathbf{r}_{i} / \sum_{i} m_{i} \tag{5.2}$$

with  $m_i$  the mass of *i*-th atom in  $\alpha$ -th cell. The motion of the center of mass represents the rigid body translation of a supercell.  $\phi_{\alpha}$  is the total deformation gradient of  $\alpha$ -th cell and is uniform throughout the cell.  $\mathbf{s}_i$  is the internal degree of freedom which represents the atomistic distribution inside the cell.  $\phi_{\alpha} \cdot \mathbf{s}_i$  is the relative position comparing to the center of mass.  $\phi_{\alpha}$  can be further decomposed to

$$\boldsymbol{\phi}_{\alpha}(t) = \boldsymbol{\chi}_{\alpha}(t) \cdot \mathbf{F}_{\alpha}(t) \tag{5.3}$$

where  $\mathbf{F}_{\alpha}$  is related to macroscale continuum deformation and depending on the centers of mass among supercells.  $\chi_{\alpha}$  is an independent part of deformation including stretch and rotation. By introducing center of mass and deformation gradient, each supercell is endowed the properties of a material point in macroscale continuum mechanics. The internal degrees of freedom is then, enable the interaction between particles from atomistic domain.

The mechanical environment of a supercell is shown in Fig.5.1(b). Two different interfaces influence the motion of the cell. First part is the interaction of surrounding atoms which can be from atomistic domain or other supercells. Second part is the stress or displacement interface from macroscale domain. We characterize these two external forces by external potential energies as,

$$V_{\alpha}^{atom} = \sum_{i \in \alpha, j \notin \alpha} \varphi(r_{ij})$$
(5.4)

$$V_{\alpha}^{surf} = -S_{\alpha}^{0} \bar{\mathbf{t}}_{\alpha}^{0} \cdot \mathbf{r}_{\alpha}$$

$$(5.5)$$

where *i* and *j* index atoms from different cells;  $\varphi$  is the pair potential, and  $r_{ij}$  is the distance between *i*-th and *j*-th atoms.  $S^0_{\alpha}$  is the surface exposed to traction  $\bar{\mathbf{t}}^0_{\alpha}$ , which are both in the referential configuration. Thus the total effect is

$$V_{\alpha}^{ext} = V_{\alpha}^{atom} + V_{\alpha}^{surf} \tag{5.6}$$

On the other hand, the internal potential energy has similar form of  $V_{\alpha}^{atom}$  which is,

$$V_{\alpha}^{int} = \frac{1}{2} \sum_{i,j \in \alpha} \varphi(r_{ij}) \tag{5.7}$$

but here the indices i and j represent atoms in the same cell and there is an  $\frac{1}{2}$  factor. The kinetic energy is proposed as,

$$K_{\alpha} = \frac{1}{2} \sum_{i} m_{i} \dot{\mathbf{r}}_{i} \cdot \dot{\mathbf{r}}_{i}$$

$$= K_{\alpha}^{rigid} + K_{\alpha}^{cell} + K_{\alpha}^{atom}$$

$$= \frac{1}{2} M_{\alpha} \dot{\mathbf{r}}_{\alpha} \cdot \dot{\mathbf{r}}_{\alpha} + \frac{1}{2} \dot{\boldsymbol{\phi}}_{\alpha}^{T} \dot{\boldsymbol{\phi}}_{\alpha} : \mathbf{J}_{\alpha} + \frac{1}{2} \mathbf{C}_{\alpha} : \sum_{i} m_{i} \dot{\mathbf{s}}_{i} \otimes \dot{\mathbf{s}}_{i}$$
(5.8)

where  $M_{\alpha}$  is the mass of the whole cell;  $\mathbf{C}_{\alpha} = \boldsymbol{\phi}_{\alpha}^{T} \boldsymbol{\phi}_{\alpha}$  is the right Cauchy-Green tensor,  $\mathbf{J}_{\alpha} = \sum_{i} m_{i} \mathbf{s}_{i} \otimes \mathbf{s}_{i}$  is the inertia tensor which is approximately constant in time. Note that in the above equation, there should be an additional cross term originally which is negligible. The Lagrangian for  $\alpha$ -th cell can be written as,

$$\mathcal{L}_{\alpha} = K_{\alpha} - V_{\alpha} 
= K_{\alpha}^{rigid} + K_{\alpha}^{cell} + K_{\alpha}^{atom} - V_{\alpha}^{int} - V_{\alpha}^{ext} 
= \frac{1}{2} M_{\alpha} \dot{\boldsymbol{r}}_{\alpha} \cdot \dot{\boldsymbol{r}}_{\alpha} + \frac{1}{2} \dot{\boldsymbol{\phi}}_{\alpha}^{T} \dot{\boldsymbol{\phi}}_{\alpha} : \mathbf{J}_{\alpha} + \frac{1}{2} \mathbf{C}_{\alpha} : \sum_{i} m_{i} \dot{\boldsymbol{s}}_{i} \otimes \dot{\boldsymbol{s}}_{i} 
- \frac{1}{2} \sum_{i,j \in \alpha} \varphi(r_{ij}) - \sum_{i \in \alpha, j \notin \alpha} \varphi(r_{ij}) + S_{\alpha}^{0} \mathbf{\bar{t}}_{\alpha}^{0} \cdot \mathbf{r}_{\alpha}$$
(5.9)

which includes three independent variables  $\mathbf{r}_{\alpha}$ ,  $\boldsymbol{\phi}_{\alpha}$  and  $\mathbf{s}_{i}$  Through the standard procedure of derivation, the equations of motion for these variables are obtained as,

$$M_{\alpha}\ddot{\mathbf{r}}_{\alpha} = \sum_{i\in\alpha, j\notin\alpha} \mathbf{f}_{ij} + S^{0}_{\alpha}\bar{\mathbf{t}}^{0}_{\alpha}$$
(5.10)

$$\ddot{\boldsymbol{\phi}}_{\alpha} \cdot \mathbf{J}_{\alpha} = \left( \boldsymbol{\mathcal{P}}_{\alpha}^{ext} - \boldsymbol{\mathcal{P}}_{\alpha}^{int} \right) \Omega_{\alpha}^{0}$$
(5.11)

$$m_i \mathbf{C}_{\alpha} \cdot \ddot{\mathbf{s}}_i = \sum_j \mathbf{f}_{ij} \cdot \boldsymbol{\phi}_{\alpha} - m_i \dot{\mathbf{C}}_{\alpha} \cdot \dot{\mathbf{s}}_i$$
 (5.12)

where  $\mathbf{f}_{ij}$  is the interaction on i-th atom from j-th atom;  $\Omega^0_{\alpha}$  is the volume of the supercell in the referential configuration; and

$$\boldsymbol{\mathcal{P}}_{\alpha}^{int} = \frac{1}{\Omega_{\alpha}^{0}} \left( \frac{1}{2} \sum_{i,j \in \alpha} \mathbf{f}_{ij} \otimes \mathbf{s}_{ij} - \boldsymbol{\phi}_{\alpha} \cdot \sum_{i \in \alpha} m_{i} \dot{\mathbf{s}}_{i} \otimes \dot{\mathbf{s}}_{i} \right)$$
(5.13)

$$\boldsymbol{\mathcal{P}}_{\alpha}^{ext} = \frac{1}{\Omega_{\alpha}^{0}} \sum_{i \in \alpha, j \notin \alpha} \mathbf{f}_{ij} \otimes \mathbf{s}_{i}$$
(5.14)

are defined as internal and external first Piola-Kirchhoff(PK-I) stresses. The equations of motion have the general form of  $\tilde{M}\ddot{\mathbf{q}} = \mathbf{\tilde{F}}$ . The general masses are  $M_{\alpha}$ ,  $\mathbf{J}_{\alpha}$  and  $m_i \mathbf{C}_{\alpha}$  for three equations, respectively. The driving force for rigid body translation  $\mathbf{r}_{\alpha}$  consists two parts from atomistic interaction and macroscale surface traction. The deformation of the cell is driven by external PK-I stress while resisted by internal PK-I stress. Internal motion is induced by atomistic interactions and damped if macroscale velocity  $\dot{\mathbf{C}}_{\alpha}$  exists.

The adaptive nature of the supercell is due to the capability of carring information or boundary conditions from different scales. The atomistic interaction influences motions of all scales including rigid body translation, deformation and internal degrees of freedom as seen in Eqs.(5.10)~(5.12), while macroscale information such as traction and displacement can be readily applied to Eq.(5.10). Moreover, we can apply an equilibrium stress state on a supercell by replacing  $\mathcal{P}_{\alpha}^{ext}$  in Eq.(5.11) with a prescribed value  $\bar{\mathcal{P}}_{\alpha}^{ext}$ . Therefore, the adaptive multiscale element is reasonable to be selected as an messenger in transition zone for the multiscale model.

## 5.3 Theory of Peridynamics

Peridynamics [95, 96, 97, 98, 99, 100] is essentially a coarse grain model. Different from classic continuum mechanics, the interaction between material points is nonlocal. For example, a material point  $\mathbf{r}_{\alpha}$  interacts with neighboring particles  $\mathbf{r}_{\beta}$  within a range called horizon, which is a similar concept with cutoff range in molecular dynamics. Here we denote the material point of interest with  $\alpha$  to be consistent with center of mass in the adaptive multiscale element, and all other particles in the horizon with  $\beta$ . Superscript "0" is used to denote the quantities in referential configuration.  $\mathcal{H}_{\mathbf{r}_{\alpha}}$  is used to denote the horizon. Interactions from material points outside the horizon are set to zero. States of a material point are described by second order tensors, such as deformation state  $\mathbf{F}_{\alpha}(\mathbf{r}_{\alpha}^{0}, t)$  and force state  $\mathbf{T}_{\alpha}(\mathbf{r}_{\alpha}^{0}, t)$ . The operations of the above states on spatial vectors are,

$$\mathbf{r}_{\beta} - \mathbf{r}_{\alpha} = \mathbf{F}_{\alpha}(\mathbf{r}_{\alpha}^{0}, t) < \mathbf{r}_{\beta}^{0} - \mathbf{r}_{\alpha}^{0} >$$
(5.15)

$$\boldsymbol{t}_{\alpha}(\mathbf{r}_{\beta}^{0} - \mathbf{r}_{\alpha}^{0}, t) = \mathbf{T}_{\alpha}(\mathbf{r}_{\alpha}^{0}, t) < \mathbf{r}_{\beta}^{0} - \mathbf{r}_{\alpha}^{0} >$$
(5.16)

where  $\mathbf{t}_{\alpha}(\mathbf{r}_{\beta}^{0} - \mathbf{r}_{\alpha}^{0}, t)$  is the force density vector act on  $\alpha$ -th point. Balance of linear momentum of the material point  $\mathbf{r}_{\alpha}$  is

$$\rho_{\alpha}\ddot{\mathbf{r}}_{\alpha} = \mathbf{L}(\mathbf{r}_{\alpha}^{0}, t) + \rho_{\alpha}\mathbf{b}(\mathbf{r}_{\alpha}^{0})$$
(5.17)

where

$$\mathbf{L}(\mathbf{r}_{\alpha}^{0},t) = \int_{\mathcal{H}_{\mathbf{r}_{\alpha}}} (\mathbf{T}_{\alpha} < \mathbf{r}_{\beta}^{0} - \mathbf{r}_{\alpha}^{0} > -\mathbf{T}_{\beta} < \mathbf{r}_{\alpha}^{0} - \mathbf{r}_{\beta}^{0} >) dV_{\beta}$$
(5.18)

is the force vector acting on  $\alpha$ -th point by neighboring points  $\beta$ . The counterpart in classical continuum mechanics is  $\nabla_0 \cdot \mathbf{P}_{\alpha}$  which is a differential form.  $\mathbf{P}_{\alpha}$  is the PK-I stress in continuum mechanics. Technically, it is much easier to solve the integral form than differential form. Thus the main work left is to evaluate the force vector. In practice, since the domain is consist of discretized material points, the integral can be replaced by a summation,

$$\mathbf{L}(\mathbf{r}_{\alpha}^{0},t) = \sum_{\mathcal{H}_{\mathbf{r}_{\alpha}}} (\mathbf{T}_{\alpha} < \mathbf{r}_{\beta}^{0} - \mathbf{r}_{\alpha}^{0} > -\mathbf{T}_{\beta} < \mathbf{r}_{\alpha}^{0} - \mathbf{r}_{\beta}^{0} >) \Delta V_{\beta}$$
(5.19)

#### **Bond-Based Peridynamics**

If we assume the pair of force density vectors be parallel to each other and has same magnitude but opposite sign, this specific model is called *Bond-based Peridynamics*. We can write the force density vectors as linearly dependent on the unit vector along two material points, which are

$$\boldsymbol{t}_{\alpha}(\mathbf{r}_{\beta}^{0}-\mathbf{r}_{\alpha}^{0},t) = \mathbf{T}_{\alpha} < \mathbf{r}_{\beta}^{0}-\mathbf{r}_{\alpha}^{0} > = \frac{1}{2}C\frac{\mathbf{r}_{\beta}-\mathbf{r}_{\alpha}}{|\mathbf{r}_{\beta}-\mathbf{r}_{\alpha}|}$$
(5.20)

and

$$\boldsymbol{t}_{\beta}(\mathbf{r}_{\alpha}^{0}-\mathbf{r}_{\beta}^{0},t) = \mathbf{T}_{\beta} < \mathbf{r}_{\alpha}^{0}-\mathbf{r}_{\beta}^{0} > = \frac{1}{2}C\frac{\mathbf{r}_{\alpha}-\mathbf{r}_{\beta}}{|\mathbf{r}_{\alpha}-\mathbf{r}_{\beta}|}$$
(5.21)

where C is the parameter determined for specific material based on classic continuum theory. For example, for an isotropic material, C can be obtained by

$$C = \frac{18\kappa s}{\pi\delta} \tag{5.22}$$

where  $\kappa$  is the bulk modulus,  $\delta$  is the radius of the horizon and s is the stretch calculated by

$$s = \frac{|\mathbf{r}_{\beta} - \mathbf{r}_{\alpha}| - |\mathbf{r}_{\beta}^{0} - \mathbf{r}_{\alpha}^{0}|}{|\mathbf{r}_{\beta}^{0} - \mathbf{r}_{\alpha}^{0}|}$$
(5.23)

Since bond-based model has only one independent parameter, the application is restricted in isotropic materials with fixed Poisson's ratio. Therefore, the model cannot distinguish distortional and volumetric deformations. Moreover, it cannot describe plastic incompressibility.

#### **Ordinary State-Based Peridynamics**

Different from Bond-based Peridynamics, the Ordinary state-based Peridynamics uses different coefficient for each force density vector as

$$\boldsymbol{t}_{\alpha}(\mathbf{r}_{\beta}^{0}-\mathbf{r}_{\alpha}^{0},t) = \mathbf{T}_{\alpha} < \mathbf{r}_{\beta}^{0}-\mathbf{r}_{\alpha}^{0} > = \frac{1}{2}A\frac{\mathbf{r}_{\beta}-\mathbf{r}_{\alpha}}{|\mathbf{r}_{\beta}-\mathbf{r}_{\alpha}|}$$
(5.24)

and

$$\boldsymbol{t}_{\beta}(\mathbf{r}_{\alpha}^{0}-\mathbf{r}_{\beta}^{0},t) = \mathbf{T}_{\beta} < \mathbf{r}_{\alpha}^{0}-\mathbf{r}_{\beta}^{0} > = \frac{1}{2}B\frac{\mathbf{r}_{\alpha}-\mathbf{r}_{\beta}}{|\mathbf{r}_{\alpha}-\mathbf{r}_{\beta}|}$$
(5.25)

Two parameters make it possible to distinguish distortional and volumetric deformations as well as plastic incompressibility. The force density vectors can be calculated from strain energy density W as

$$\boldsymbol{t}_{\alpha}(\mathbf{r}_{\beta}^{0}-\mathbf{r}_{\alpha}^{0},t) = \frac{\partial W(\boldsymbol{r}_{\alpha}^{0})}{\partial(|\mathbf{r}_{\beta}-\mathbf{r}_{\alpha}|)} \frac{\mathbf{r}_{\beta}-\mathbf{r}_{\alpha}}{|\mathbf{r}_{\beta}-\mathbf{r}_{\alpha}|}$$
(5.26)

and

$$\boldsymbol{t}_{\beta}(\mathbf{r}_{\alpha}^{0}-\mathbf{r}_{\beta}^{0},t) == \frac{\partial W(\boldsymbol{r}_{\beta}^{0})}{\partial(|\mathbf{r}_{\alpha}-\mathbf{r}_{\beta}|)} \frac{\mathbf{r}_{\alpha}-\mathbf{r}_{\beta}}{|\mathbf{r}_{\alpha}-\mathbf{r}_{\beta}|}$$
(5.27)

Coefficients A and B are then determined by these relations.

#### **Nonordinary State-Based Peridynamics**

In the nonordinary state-based peridynamics, based on the energy equivalence [99],  $\mathbf{T}_{\alpha}$  can be calculated as

$$\mathbf{T}_{\alpha} < \mathbf{r}_{\beta}^{0} - \mathbf{r}_{\alpha}^{0} >= \omega(\mathbf{r}_{\beta}^{0} - \mathbf{r}_{\alpha}^{0})\mathbf{P}_{\alpha} \cdot \mathbf{K}_{\alpha}^{-1} \cdot (\mathbf{r}_{\beta}^{0} - \mathbf{r}_{\alpha}^{0})$$
(5.28)

where  $\omega(\mathbf{r}^0_{\beta} - \mathbf{r}^0_{\alpha})$  is the influence function of the bond  $\mathbf{r}^0_{\beta} - \mathbf{r}^0_{\alpha}$ , and

$$\mathbf{K}_{\alpha} = \sum_{\mathcal{H}_{\mathbf{r}_{\alpha}}} \omega(\mathbf{r}_{\beta}^{0} - \mathbf{r}_{\alpha}^{0})(\mathbf{r}_{\beta}^{0} - \mathbf{r}_{\alpha}^{0}) \otimes (\mathbf{r}_{\beta}^{0} - \mathbf{r}_{\alpha}^{0}) \Delta V_{\beta}$$
(5.29)

and deformation gradient can be calculated as

$$\mathbf{F}_{\alpha} = \left(\sum_{\mathcal{H}_{\mathbf{r}_{\alpha}}} \omega(\mathbf{r}_{\beta}^{0} - \mathbf{r}_{\alpha}^{0})(\mathbf{r}_{\beta} - \mathbf{r}_{\alpha}) \otimes (\mathbf{r}_{\beta}^{0} - \mathbf{r}_{\alpha}^{0}) \Delta V_{\beta}\right) \cdot \mathbf{K}_{\alpha}^{-1}$$
(5.30)

Based on deformation gradient, we can calculate the PK-I stress by introducing constitutive models from classical continuum mechanics.

#### **Damage Models**

#### • Damage based on bond stretch[96]

The constitutive relation of Bond-based Peridynamics is linear because the coefficient C depends on stretch s only. We assume the bond between two material points fails when the stretch exceeds a critical value  $s_0$ .  $s_0$  can be determined by calculating energy release rate  $G_0$ . The work required to break a single bond of linear elastic material is  $w_0 = C s_0^2 \xi/2$ , where  $\xi$  is the initial bond length given by  $\xi = |\mathbf{r}_{\beta}^0 - \mathbf{r}_{\alpha}^0|$ . Consider a planar fracture surface as shown in Fig.5.2. To break all bonds that connecting points in two halves of the fracture surface, the energy required per unit area is found to be

$$G_{0} = \int_{0}^{\delta} \int_{0}^{2\pi} \int_{z}^{\delta} \int_{0}^{\cos^{-1}z/\xi} w_{0}\xi^{2} \sin\phi d\phi d\xi d\theta dz$$
(5.31)

Evaluating the integral, we get

$$G_0 = \frac{\pi C s_0^2 \delta^5}{10} \tag{5.32}$$

and then

$$s_0 = \sqrt{\frac{10G_0}{\pi C \delta^5}} \tag{5.33}$$

Thus  $s_0$  is determined by specific energy release rate from the above equation.

• Damage based on strain state[101]

Two criteria of breaking bonds in state-based model are presented here. Consider the average Green-Lagrange strain tensor  $\mathbf{E}a = (\mathbf{E}(\mathbf{r}_{\alpha}) + \mathbf{E}(\mathbf{r}_{\alpha}))/2$ . The first criterion is related to the second invariant  $I'_2$  of deviatoric strain tensor  $\mathbf{E}a' = \mathbf{E}a - 1/3\mathbf{E}a_{\alpha\alpha}\mathbf{I}$ . The equivalent strain is obtained through

$$E_{eq} = \sqrt{\frac{4}{3}I_2'} = \sqrt{\frac{2}{3}\boldsymbol{E}\boldsymbol{a}:\boldsymbol{E}\boldsymbol{a}}$$
(5.34)

The critical value  $E_{eq}^{crit}$  can be obtained by experiments. Second, consider the averaged value of the volumetric strain

$$E_{vol} = I_1 + I_2 + I_3 = Ea_{11} + Ea_{22} + Ea_{33} + Ea_{11}Ea_{22} + Ea_{22}Ea_{33} + Ea_{33}Ea_{11} + Ea_{11}Ea_{22}Ea_{33}$$
(5.35)

The critical value  $E_{vol}^{crit}$  can also be obtained by experiments for specific materials.



Figure 5.2: Geometry for calculating critical stretch in bond-based model

#### **Cauchy-Born Stress Evaluation**

For PK-I stress used in non-ordinary state-based peridynamics, except classical constitutive relation, we may also derive it from atomistic potential with Cauchy-Born rule[102, 103]. In this case, each peridynamics point is associated with an atomistic unit cell (see Fig.5.3). The elastic energy density at each peridynamics point is

$$W = \frac{1}{\Delta V_{\alpha}} \sum_{k=1}^{n_b} \varphi_{r_k} \tag{5.36}$$

The calculation is inside an unit cell.  $\mathbf{k} = 1,...,n_b$ , where  $n_b$  is the number of bonds in a unit cell.  $\varphi(r_k)$  is the pair potential of k-th bond. Based on the Cauchy-Born rule, we can calculate the PK-I stress by taking derivative of elastic energy density with respect to deformation gradient,

$$\mathbf{P}_{\alpha} = \frac{\partial W}{\partial \mathbf{F}}|_{\mathbf{F}=\mathbf{F}_{\alpha}} = \frac{1}{\Delta V_{\alpha}} \sum_{k=1}^{n_{b}} \varphi'(r_{k}) \frac{\mathbf{r}_{k} \otimes \mathbf{R}_{k}}{r_{k}}$$
(5.37)

 $\mathbf{r}_k$  and  $\mathbf{R}_k$  are bond vectors in current and referential configurations, respectively. Cauchy-Born rule says  $\mathbf{r}_k = \mathbf{F}_{\alpha} \cdot \mathbf{R}_k$ . Therefore, if we have the deformation gradient of a material point, the above equation of PK-I stress is obtained from  $\mathbf{F}_{\alpha}$ .



Figure 5.3: The Cauchy-Born rule based multiscale peridynamics

## 5.4 Construction of the Filter

A common issue of multiscale methods is the reflection of high-frequancy waves on the interface. Due to the larger lattice spacing of the macroscale domain, high-frequency wave components cannot be recognized. Therefore, as a result of energy conservation, highfrequency signals will be reflected back to the atomistic domain. The issue of reflection is demonstrated in Fig.5.4. When the wave passes the interface, the low-frequency wave transits smoothly, but the reflection of high-frequency components can be observed. Note that high and low frequencies are relative concepts compared with lattice spacings of both atomistic and macro domain. Low frequency means the wavelength is longer than intrinsic lattice spacing of macro domain, while high frequency is where the wavelength in between the intrinsic lattice spacings of the domains.

To solve the issue of reflection, we introduce a filter in the transition zone near the surface of macro domain as previously demonstrated in Fig.5.1(a). The filter is similar as transition element in shape, which is also an assemble of atoms. The construction of the filter includes two steps as is shown in Fig.5.5. First, we allow all wave components enter the filter. In this step, the filter is basically the same as transition element, where atoms in the filter are free to move as any transition element. The motion is controlled by original dynamical Eqs.5.10~5.12. Second step is a process of homogenization. The atomistic positions are set to an average value. The equivalent manipulation is to replace  $\mathbf{s}_i$  with its initial value  $\mathbf{s}_i^0$ . Thus the homogenized atomistic position is

$$\mathbf{r}_i = \mathbf{r}_\alpha + \boldsymbol{\phi}_\alpha \cdot \mathbf{s}_i^0 \tag{5.38}$$



Figure 5.4: Illustration of wave reflection. Due to the larger lattice spacing, the macro region may not be able to recognize the high-frequency components. If the total energy is conserved without releasing, these high-frequency components will reflect back to the atomistic region.

The replacement is effective in Eqs. $5.10 \sim 5.11$ , but Eq.5.12 is eliminated. It is like the internal degrees of freedom are frozen and only macroscale motions are allowed, i.e. particles are adhered on the uniform deformation of the supercell. This is an energy release process. The interesting fact of the adaptive multiscale element is that we can lock any of the three scale variables to meet the need of different resolutions. The treatment of directly sweeping the high frequency components is brutal but effective. When the waves enter the filter, a "smooth" uniform displacement is taken to replace the waves. Theoretically, all high-frequency waves are expected to be cleaned while the low-frequency components are transmitted to the macro domain. We will demonstrate the process in the numerical examples.

## 5.5 Numerical Scheme

The transition zone is introduced to translate information between different regions. In a mechanical system, force(stress) and displacement(strain) are two basic sets of quantities. As we discussed in section 5.2, the adaptive multiscale element is capable of carrying all these information from both regions. Thus the message translation is seamless without unphysical consequences. We briefly state the two options of message passing on the interface as follow, 1. Traction-Force interchange. In this case, the transition zone is a separate part. The traction from macro domain acts on the transition elements, while at the same time the



Figure 5.5: Construction of the filter. Step I: The filter has the same function as transition element, where all wave components propagate into the the element. Step II: Homogenization. Displacements are averaged inside the filter. In practice, we set the internal variable  $\mathbf{s}_i$  to its initial value  $\mathbf{s}_i^0$ , which produces a uniform total deformation.

macro domain gets the reaction. These action and reaction has the form of macroscale traction, which is incorporated in Eq.(5.10). On the other side, atomistic domain interact with transition zone by atomistic forces. The procedure is concurrent in time, i.e. three domains evolve at the same time.

2. Displacement interchange. In this case, the transition zone can be treated as an overlap of both macro and atomistic domains. In MD updates, the transition zone is a part of atomistic domain. The updated centers of mass are then passed to the macro domain as displacement boundary condition. Similarly, the updated macroscale displacements are used for the transition zone as centers of mass, while the other two variables are still free of motion, as in Eqs.(5.11)~(5.12). Meanwhile, the atomistic region updates based on the new atomic positions in transition zone.

An appropriate integration scheme is necessary for updating the quantities and facilitate the exchange of information. The advantage of the multiscale structure is not only in space but also in time, as we discussed in chapter 3. An efficient strategy is choosing larger time steps for macroscale and smaller time steps for atomistic scale. There are two levels of computational cycles. The two domains are on the first level. Based on different approaches of force-traction or displacement interchange, parallel and serial algorithms can be used separately as presented in chapter 3. The second level is the three quantities of the adaptive multiscale element in transition zone, which are  $\mathbf{r}_{\alpha}$ ,  $\boldsymbol{\phi}_{\alpha}$  and  $\mathbf{s}_i$ . On the second level, both parallel and serial algorithm are applicable based on different conditions and requirements. Velocity verlet and predictor-corrector are chosen as integrators in the computational model. Velocity verlet is used in calculating Macro region, atomistic region and the centers of mass  $\mathbf{r}_{\alpha}$  of transition element. Predictor-corrector is used to calculate  $\boldsymbol{\phi}_{\alpha}$  and  $\mathbf{s}_i$  of transition element. As an example, the MD update is stated as follow,

$$\mathbf{r}_{i}^{n+1} = \mathbf{r}_{i}^{n} + \dot{\mathbf{r}}_{i}^{n} \Delta t_{\mathbf{r}_{i}} + \frac{1}{2} \ddot{\mathbf{r}}_{i}^{n} \Delta t_{\mathbf{r}_{i}}^{2}$$

$$(5.39)$$

$$\ddot{\mathbf{r}}_{i}^{n+1} = \mathbf{f}_{i}^{n+1}(\mathbf{r}_{i}^{n+1})/m_{i}$$
(5.40)

$$\dot{\mathbf{r}}_{i}^{n+1} = \dot{\mathbf{r}}_{i}^{n} + \frac{1}{2} \left( \ddot{\mathbf{r}}_{i}^{n} \Delta t_{\mathbf{r}_{i}} + \ddot{\mathbf{r}}_{i}^{n+1} \Delta t_{\mathbf{r}_{i}} \right)$$
(5.41)

In the second step, we need to evaluate force based on the new position  $\mathbf{r}_i^{n+1}$ . We organize the time integration procedure in the following table,

- Determine the specific problem of interest, e.g. wave or crack propagation, etc.
- Set up parameters of the system, e.g. material, model dimension, time steps, etc.
- Initialize the system: initial displacement, velocity, boundary condition, etc.
- Select the type of communication: force-traction/displacement.
- Update atomistic domain: Velocity-verlet for  $r_i$ .
- Update Transition zone:
- Velocity-verlet for  $\mathbf{r}_{\alpha}$  in Eq.(5.10);
- Predictor-corrector for  $\phi_{\alpha}$  and  $\mathbf{s}_i$  in Eqs.(5.11)~(5.12);
- Update macro domain: Velocity-verlet for  $\mathbf{r}_{\alpha}$  in Eq.(5.17).
- Exchange of information:
  - Force-traction: atomic forces  $\rightleftharpoons$  transition elements  $\rightleftharpoons$  macro traction
  - Displacement: atomic displacements  $\rightarrow$  transition zone  $\rightarrow$  macro displacements  $\rightarrow$  transition zone  $\rightarrow$  atomic displacements

## Chapter 6

## Benchmark Tests and Applications of the Coupling Model

In this chapter, we present two examples of 1-D and 2-D wave propagation to validate the adaptive multiscale model. Displacement transition is demonstrated in these examples. Pairwise Morse potential is used to model the atomistic interaction, which is

$$\varphi(r) = D(e^{-2\alpha(r-r_0)} - 2e^{-\alpha(r-r_0)})$$
(6.1)

The pair force is derived as

$$f(r) = -\frac{\partial\phi(r)}{\partial r} = 2D\alpha(e^{-2\alpha(r-r_0)} - e^{-\alpha(r-r_0)})$$
(6.2)

where the constants D = 0.0965 eV,  $\alpha = 2.71/\text{\AA}$  and  $r_0 = 2.878$  Å.

## 6.1 Benchmark Test: 1-D Wave Propagation

In the 1-D problem, Aluminum with an atomic weigh of 26.98 u is selected as the material under studying. The lattice space is  $a_0 = 2.878$  Å. Each adaptive multiscale element has 5 atoms, and each peridynamical material points is chosen the same size of the multisacle element. The atomistic domain consists of 475 atoms. 5 multiscale elements are located in the transition zone. A total 150 material points are assigned to the macro domain. The model is shown in Fig.6.1. An initial Gaussian displacement is applied as,

$$u(x,t=0) = \begin{cases} Ae^{-\frac{x^2}{2\sigma^2}} (1+b\cos(\frac{2\pi x}{H})) & x \le L_c \\ 0 & x > L_c \end{cases}$$
(6.3)

with A=0.01,  $\sigma = 200$ , b=0.08, and H= $\sigma/5$ , in angstrom. The initial displacement is truncated at  $L_c = 4\sigma$ . The exponential term in Eq.6.3 is the low-frequency wave which

is expected to pass through the transition zone to the macro region. The cosine term is the high-frequency component which should be appropriately filtered and the associated energy should be released. Time steps for atomistic and macro regions are  $\Delta t_a = 0.001$  and  $\Delta t_m = 0.006$ , respectively. For the transition elements, time steps are chosen as  $\Delta t_s = 0.001$ ,  $\Delta t_{\phi} = 0.002$  and  $\Delta t_r = 0.002$ . All units are in picosecond. The above selection of step sizes is just a demonstration. Practically, different combinations can be considered as long as they are fully tested to avoid lead or lag in time between different regions. During the updates, each scale or region includes several substeps. The number of substeps should be tested to ensure smooth transition.



Figure 6.1: 1-D model setup with an Gaussian initial displacement of magnitude 0.01. From left to right: atomistic domain ( $0 \le x < 1367$ ), transition zone( $1367 \le x < 1439$ ) and macro domain ( $1439 \le x < 3598$ ). All units are in angstrom.

Fig.6.2(a) and (b) show the procedure of wave propagation without filter in transition zone. When macroscale time steps are smaller than 2250, the whole wave is in the atomistic domain. We can clearly distinguish the low and high frequency of the wave. When it propagates to the macro domain, the low frequency wave is almost entirely transmitted. However, without filter, the high frequency wave is reflected back to the atomistic domain because macroscale cannot recognize this signal.

As we discussed in last section, constructing filter is simple and straightforward. In this example, we pick out 3 multiscale elements as filter. Fig.6.3 shows the transmission at

CHAPTER 6. BENCHMARK TESTS AND APPLICATIONS OF THE COUPLING MODEL



Figure 6.2: Displacement at (a)t=2250 and (b)t=5000 with units of macroscale step size. No filter is placed in the transition zone.

t=5000 when filter is turned on. Compare with Fig.6.2(b), reflection of high frequency waves is largely mitigated. We may increase the size of the filter to further tune up the result.

To better exam the passage of information in the procedure, we plot the normalized energy of each domain in macroscale time steps. Fig.6.4(a) shows the elastic energy of the atomistic domain with and without filter, respectively. When the macro time steps are between 2300 and 4300, the wave transits from atomistic to macro domain. Ideally, total energy in atomistic domain should be damped off after 4300 macro time steps, as observed in the energy curve with filter. However, when the filter is turned off, we find a small amount of residual energy which is not the case in reality. The filter is capable of getting rid of more than 90% residual energy. The comparison of energy transition of both domains is shown in Fig.6.4(b). Before and after the transition, energies are all constant with normalized value of 0 and 1.

## 6.2 Benchmark Test: 2-D Wave Propagation

In this example, a hexagonal close-packed (HCP) plane is employed as a 2-D model. The material is aluminum with an atomistic weigh of 26.98 u same as the 1-D model. The lattice constant is  $a_0 = 2.878$  Å. Same Morse potential is used for 2-D lattice. The shape of the multiscale elements in transition zone and material points in macro domain is shown in Fig.6.5. Each multiscale element includes 9 atoms. The distance from the center of each element to the nearest neighbor is  $3a_0$ . The entire model is a rectangle as shown in Fig.6.6.



Figure 6.3: Displacement at t=5000 with filter in the transition zone.



Figure 6.4: Evolution of energies in each region.



Figure 6.5: Shape of the multiscale elements in transition zone and material points in macro domain.

A atomistic domain is located at the center with  $150 \times 150$  atoms. The size is calculated as  $150 \times 75\sqrt{3}a_0^2$  according to the lattice pattern. The transition zone surrounding the atomistic domain has a thickness of 5 multiscale element, in which a 2-element thickness is assigned as filter. Outside the atomistic domain and the transition zone, a macro domain with  $160 \times 160$  material points except the central area, or a size of  $480 \times 240\sqrt{3}a_0^2$  is constructed. Similar as 1-D problem, an initial Gaussian wave in polar coordinate is applied as

$$u(r,t=0) = \begin{cases} Ae^{-\frac{r^2}{2\sigma^2}} (1+b\cos(\frac{2\pi r}{H})) & r \le L_c \\ 0 & r > L_c \end{cases}$$
(6.4)

Fig.6.7(a)~(f) shows the history of the wave propagate from atomistic domain into the macro domain. Fig.6.7(a) is the initial wave as mentioned above. Fig.6.7(b) is the snapshot when the whole wave is still inside the atomistic domain. Two separate rings are formed with higher magnitude of the inner ring and lower magnitude of the outer ring. Subsequently, the outer ring passes the interface where the multiscale elements are located to aid the transition. As shown in Fig.6.7(c), the interface does not cause any mismatch or discontinuity, and the outer ring is smoothly distributed on both sides of the interface. Fig.6.7(d) is the time instance when the inner ring spassing the interface. We can see that even the with higher magnitude and abrupt slope, the inner ring still passes under control. When the whole wave passes the interface, as is shown in Fig.6.7(e), minimum residual is observed in atomistic domain even sharp corners are existed on the interface. Note that the rings will be damped when they expand the radius due to the energy conservation. In Fig.6.7(e), the outer ring disappeared first. Subsequently, the inner ring is further propagating and damping in the macro domain.

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Figure 6.6: Initial model setup of the 2-D case. Inside is the atomistic region with rectangular shape. A larger macro region surrounds the atomistic region. Initial Gaussian wave is applied with a magnitude of 0.01.



Figure 6.7: History of wave propagation.

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# Chapter 7 Summary and Outlook

In this dissertation, we demonstrated a systematic multiscale mechanical theory for the purpose of bridging microscopic and macroscopic time and spacial scales. The multiscale model is based on the fundamental molecular system which conserves full atomistic degrees of freedom. While at the same time, we deal with the system by dividing atoms into several clusters. The advantage of this treatment is when atoms are grouped together, the clusters has properties such as deformation, global displacements and velocities due to the relative motion of atoms. Those new properties are actually descriptors of macroscale mechanical behaviors and are directly related to the concepts in continuum mechanics. For example, each cluster or "supercell" is similar to the counterpart of "material point" in continuum mechanics, with the properties of deformation, rotation and translation. On the other hand, atomistic motions are conserved in terms of internal degrees of freedom. Therefore, without extra approximation, the multiscale system is completely equivalent to the original molecular system. Based on this multiscale structure, we analyzed the force fields and the associated potential energies. The external force fields include microscale atomic interaction and macroscale distributed load directly act on the supercell. We derived the equations of motion for the system with quantities in different scales and discussed the capability of carrying macroscale mechanical boundary conditions. When we had characterized the properties and dynamics in the multiscale system, we were able to apply mechanical boundary conditions in both scales, e.g. apply marcoscale traction and observe the response of the molecular system in microscale. This top-down message passing expand the horizon of traditional molecular dynamics, where mechanical loads in terms of atomistic forces and displacements are adopted, but macroscale distributed loads can not be recognized. The top-down approach as well as established bottom-up statistical approach complete the cross-scale communication in multiscale simulations. The proposed model is inspired by Parrinello-Rahman molecular dynamics, where a representative volume element (RVE) with deformable shape allows constant stress control. However, we generalized the periodic boundary condition to arbitrary boundary conditions. Therefore, the new model is adaptive for finite-size problems with inhomogeneous domain, while the multiscale feature is conserved.

Since we introduced several macroscale quantities such as mass center and deformation

gradient into the molecular system, those degrees of freedom are redundant compare with the original system even though they are indispensable. To conserve the physical representations, necessary constraints were introduced and discussed. For the part of implementation, we employed several numerical schemes for time integration, e.g. predictor-corrector, velocity verlet, etc. And Nose-Hover thermostat is used to control temperature.

We found the applications of the multiscale theory in simulating stress-induced phase transition of Nickel FCC lattice. While traditional molecular dynamics has to apply forces on specific particles, our multiscale model is suitable in treating macroscale traction and displacement boundary. We studied different models such as bulk lattice and nanowire when they were exposed in traction and macroscale displacement boundary conditions. The original FCC lattice went through phase transition and turned into HCP lattice as expected. The stress-strain relation of displacement loading was compared with theoretical results and found satisfactory agreement. In traction loading curves, critical points with jumps were observed due to the unstableness of the intermediate structure, which were different from the continuous curve when displacement was controlled. Regular patterns throughout the whole domain after the transition shows the advantage of the multiscale model, since atomic distribution will be irregular around the boundary area when "strong conditions" are applied on specific atoms.

A computational framework coupling molecular dynamics and peridynamics was constructed to achieve efficiency. In this multiscale framework, we separate a body into macroscopic and atomistic regions with an interface as transition zone. Molecular dynamics is employed in atomistic simulation. Peridynamics is chosen for macroscopic domain due to the nonlocal nature of the theory. The previously developed multiscale theory is adopted in the transition zone. Since the "supercell" is capable of carrying information from both atomic and macroscale models, we repackage it as a transition element. Messages such as macroscopic distributed loads, atomic forces and displacements are then translated across regions with rigid theoretical fundation. We enriched the peridynamics theory by introducing the Cauchy-Born stress measurement, where interatomic potential is employed instead of artificial constitutive assumptions in macroscale models. Therefore, two regions are consistently linked with the same interatomic potentials. We constructed a filter near the interface to solve the common issue of wave reflection in multiscale simulations. Since the larger lattice spacing in macroscopic region cannot recognize short wavelengths, high-frequency wave components will be reflected back on the interface. The filter has an initial structure as the transition element. When the waves enter the filter, we force sweeping the signals with a homogenization procedure. The effect is influenced by the sizes of the element and time step chosen, so that they should be tested before practice. As a multiscale model, we need to wisely choose the numerical schemes to optimize the efficiency. Following the study in the previous multiscale theory, we choose velocity verlet and predictor-corrector algorithms to adapt scales or models with different properties. Sequential and parallel approaches are applied as different message-passing strategies, respectively. For stress-force interchange, all regions interact with each other and evolve concurrently. For displacement interchange, information passes from one region to the other and back.

We did benchmark tests to verify the multiscale computational model. Examples include 1-D and 2-D wave propagations. Aluminum was the test material. In the 1-D problem, a uniform lattice was separated into macroscopic and atomistic regions with a transition zone placed on the interface. An initial Gaussian wave was applied in the atomistic region and then propagated to the macroscopic region. We compared the results with and without the filter. High-frequency wave components were largely mitigated when the filter was turned on. We examined the energy profiles on both regions to further prove the effect of the filter and clear the procedure of transition. In the 2-D example, a closely packed plane was assigned as atomistic region which was surrounded by a larger macroscopic region. Same as the 1-D problem, the initial Gaussian wave in atomic region passed the interface smoothly and propagated to the macroscopic region.

This work provides the theoretical foundation for multiscale simulations and demonstrates a computational paradigm for engineering practice, which are expected inspire extensive applications in this field. The models are developed in the scope of mechanical systems, i.e. only force (traction)-displacement (deformation) responses are discussed. However, the idea can be extended to other systems. For example, we may consider heat flow in a molecular system when thermodynamical boundary condition is applied. In a more general scope, the philosophy of finding fundamental connection and translate properties between different models can be a inspiration for new discoveries in many other fields.

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## Appendix A

# Derivation of macroscale dynamic equation

In this Appendix, we present the derivation of macroscale dynamic equation with continuum deformation. We know that the continuum deformation gradient depends on the relative displacements of all centers of mass, which is,

$$\mathbf{F}_{\alpha} = \mathbf{F}_{\alpha}(\{\mathbf{r}_{\beta}\}) \tag{A.1}$$

where  $\{\mathbf{r}_{\beta}\}$  represent a set of all centers of mass. Then we have

$$\dot{\mathbf{F}}_{\alpha} = \mathbf{F}_{\alpha}(\{\mathbf{r}_{\beta}\}, \{\dot{\mathbf{r}}_{\beta}\}) \tag{A.2}$$

From equation (A.1), we also have,

$$\dot{\mathbf{F}}_{\alpha} = \sum_{\beta} \frac{\partial \mathbf{F}_{\alpha}}{\partial \mathbf{r}_{\beta}} \dot{\mathbf{r}}_{\beta} \tag{A.3}$$

therefore,

$$\frac{\partial \dot{\mathbf{F}}_{\alpha}}{\partial \dot{\mathbf{r}}_{\alpha}} = \frac{\partial \mathbf{F}_{\alpha}}{\partial \mathbf{r}_{\alpha}} \tag{A.4}$$

From equation (A.2), we can derive,

$$\ddot{\mathbf{F}}_{\alpha} = \sum_{\beta} \left[ \frac{\partial \dot{\mathbf{F}}_{\alpha}}{\partial \mathbf{r}_{\beta}} \dot{\mathbf{r}}_{\beta} + \frac{\partial \dot{\mathbf{F}}_{\alpha}}{\partial \dot{\mathbf{r}}_{\beta}} \ddot{\mathbf{r}}_{\beta} \right]$$
(A.5)

On the other hand, from Eq. (A.3), the relation is,

$$\ddot{\mathbf{F}}_{\alpha} = \sum_{\beta} \left[ \frac{d}{dt} \left( \frac{\partial \mathbf{F}_{\alpha}}{\partial \mathbf{r}_{\beta}} \right) \dot{\mathbf{r}}_{\beta} + \frac{\partial \mathbf{F}_{\alpha}}{\partial \mathbf{r}_{\beta}} \ddot{\mathbf{r}}_{\beta} \right]$$
(A.6)

Comparing Eqs. (A.5) and (A.6), and using relation (A.4), we get,

$$\frac{\partial \mathbf{F}_{\alpha}}{\partial \mathbf{r}_{\alpha}} = \frac{d}{dt} \left( \frac{\partial \mathbf{F}_{\alpha}}{\partial \mathbf{r}_{\beta}} \right) \tag{A.7}$$

Recall the total deformation  $\phi_{\alpha} = \chi_{\alpha} \cdot \mathbf{F}_{\alpha}$ , then the time derivative is,

$$\boldsymbol{\phi}_{\alpha} = \dot{\boldsymbol{\chi}}_{\alpha} \cdot \mathbf{F}_{\alpha} + \boldsymbol{\chi}_{\alpha} \cdot \mathbf{F}_{\alpha} \tag{A.8}$$

By knowing the fact that  $\chi_{\alpha}$  is an independent variable, and using relations (A.4) and (A.7), we have,

$$\frac{\partial \dot{\boldsymbol{\phi}}_{\alpha}}{\partial \dot{\mathbf{r}}_{\alpha}} = \boldsymbol{\chi}_{\alpha} \frac{\partial \dot{\mathbf{F}}_{\alpha}}{\partial \dot{\mathbf{r}}_{\alpha}} = \boldsymbol{\chi}_{\alpha} \frac{\partial \mathbf{F}_{\alpha}}{\partial \mathbf{r}_{\alpha}} = \frac{\partial \boldsymbol{\phi}_{\alpha}}{\partial \mathbf{r}_{\alpha}} \tag{A.9}$$

and,

$$\frac{\partial \dot{\boldsymbol{\phi}}_{\alpha}}{\partial \mathbf{r}_{\alpha}} = \boldsymbol{\chi}_{\alpha} \frac{\partial \dot{\mathbf{F}}_{\alpha}}{\partial \mathbf{r}_{\alpha}} + \dot{\boldsymbol{\chi}}_{\alpha} \frac{\partial \mathbf{F}_{\alpha}}{\partial \mathbf{r}_{\alpha}} \\
= \boldsymbol{\chi}_{\alpha} \frac{d}{dt} \left( \frac{\partial \mathbf{F}_{\alpha}}{\partial \mathbf{r}_{\beta}} \right) + \dot{\boldsymbol{\chi}}_{\alpha} \frac{\partial \mathbf{F}_{\alpha}}{\partial \mathbf{r}_{\alpha}} \\
= \frac{d}{dt} \left( \frac{\partial \boldsymbol{\phi}_{\alpha}}{\partial \mathbf{r}_{\alpha}} \right) = \frac{d}{dt} \left( \frac{\partial \dot{\boldsymbol{\phi}}_{\alpha}}{\partial \dot{\mathbf{r}}_{\alpha}} \right) \tag{A.10}$$

We rewrite Lagrangian here,

$$\mathcal{L}_{\alpha} = \frac{1}{2} M_{\alpha} \dot{\boldsymbol{r}}_{\alpha} \cdot \dot{\boldsymbol{r}}_{\alpha} + \frac{1}{2} \dot{\boldsymbol{\phi}}_{\alpha}^{T} \dot{\boldsymbol{\phi}}_{\alpha} : \mathbf{J}_{\alpha} + \frac{1}{2} \mathbf{C}_{\alpha} : \sum_{i} m_{i} \dot{\boldsymbol{s}}_{i} \otimes \dot{\boldsymbol{s}}_{i} - \frac{1}{2} \sum_{i,j \in \alpha} \varphi(r_{ij}) - \sum_{i \in \alpha, j \notin \alpha} \varphi(r_{ij}) + S_{\alpha}^{0} \bar{\mathbf{t}}_{\alpha} \cdot \mathbf{r}_{\alpha} + \Omega_{\alpha}^{0} \bar{\mathbf{b}}_{\alpha} \cdot \mathbf{r}_{\alpha}$$
(A.11)

We have

$$\frac{d}{dt} \left( \frac{\partial \mathcal{L}_{\alpha}}{\partial \dot{\mathbf{r}}_{\alpha}} \right) = \frac{d}{dt} \left( \frac{\partial \mathcal{L}_{\alpha}}{\partial \dot{\mathbf{r}}_{\alpha}} + \frac{\partial \mathcal{L}_{\alpha}}{\partial \dot{\phi}_{\alpha}} \cdot \frac{\partial \dot{\phi}_{\alpha}}{\partial \dot{\mathbf{r}}_{\alpha}} \right)$$

$$= M_{\alpha} \ddot{\mathbf{r}}_{\alpha} + \frac{d}{dt} \left( \frac{\partial \mathcal{L}_{\alpha}}{\partial \dot{\phi}_{\alpha}} \right) \cdot \frac{\partial \dot{\phi}_{\alpha}}{\partial \dot{\mathbf{r}}_{\alpha}} + \frac{\partial \mathcal{L}_{\alpha}}{\partial \dot{\phi}_{\alpha}} \cdot \frac{d}{dt} \left( \frac{\partial \dot{\phi}_{\alpha}}{\partial \dot{\mathbf{r}}_{\alpha}} \right) \quad (A.12)$$

and

$$\frac{\partial \mathcal{L}_{\alpha}}{\partial \mathbf{r}_{\alpha}} = \frac{\partial \mathcal{L}_{\alpha}}{\partial \mathbf{r}_{\alpha}} + \frac{\partial \mathcal{L}_{\alpha}}{\partial \phi_{\alpha}} \cdot \frac{\partial \phi_{\alpha}}{\partial \mathbf{r}_{\alpha}} + \frac{\partial \mathcal{L}_{\alpha}}{\partial \dot{\phi}_{\alpha}} \cdot \frac{\partial \dot{\phi}_{\alpha}}{\partial \mathbf{r}_{\alpha}} \\
= \sum_{i \in \alpha, j \notin \alpha} \mathbf{f}_{ij} + S_{\alpha}^{0} \bar{\mathbf{t}}_{\alpha} + \Omega_{\alpha}^{0} \bar{\mathbf{b}}_{\alpha} + \frac{\partial \mathcal{L}_{\alpha}}{\partial \phi_{\alpha}} \cdot \frac{\partial \phi_{\alpha}}{\partial \mathbf{r}_{\alpha}} + \frac{\partial \mathcal{L}_{\alpha}}{\partial \dot{\phi}_{\alpha}} \cdot \frac{\partial \dot{\phi}_{\alpha}}{\partial \mathbf{r}_{\alpha}} \quad (A.13)$$

The equations of motion for  $\mathbf{r}_{\alpha}$  and  $\boldsymbol{\phi}_{\alpha}$  are derived as,

$$\frac{d}{dt} \left( \frac{\partial \mathcal{L}_{\alpha}}{\partial \dot{\mathbf{r}}_{\alpha}} \right) = \frac{\partial \mathcal{L}_{\alpha}}{\partial \mathbf{r}_{\alpha}} \tag{A.14}$$

$$\frac{d}{dt} \left( \frac{\partial \mathcal{L}_{\alpha}}{\partial \dot{\phi}_{\alpha}} \right) = \frac{\partial \mathcal{L}_{\alpha}}{\partial \phi_{\alpha}} \tag{A.15}$$

Considering the relations in Eqs. (A.9), (A.10) and (A.15), last two terms in Eqs. (A.12) and (A.13) are cancelled when they are plugged into (A.14). Finally, we have,

$$M_{\alpha}\ddot{\mathbf{r}}_{\alpha} = \sum_{i \in \alpha, j \notin \alpha} \mathbf{f}_{ij} + S^{0}_{\alpha}\bar{\mathbf{t}}_{\alpha} + \Omega^{0}_{\alpha}\bar{\mathbf{b}}_{\alpha}$$
(A.16)

which is identical as equation (2.98).