Oral presentation | Numerical methods
Numerical methods-II
Tue. Jul 16, 2024 4:30 PM - 6:30 PM Room A

[6-A-02] Determining the Significance of Nonequilibrium Effects in Materials Using a Continuum-Atomistic Framework

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Keywords: multiscale simulation, molecular dynamics, finite elements

Twelfth International Conference on Computational Fluid Dynamics (ICCFD12), Kobe, Japan, July 14-19, 2024

ICCFD12-2024-xxxx

Modeling Nonequilibrium Effects in Fluids Using a Continuum-Atomistic Framework

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In the field of high-energy-density physics, understanding material behavior under non-equilibrium conditions is essential. Traditional hydrodynamic simulations, relying on predefined equations of state (EOS), often fail to capture the complex phenomena that occur far from equilibrium. This paper presents an alternative approach using a continuum-atomistic framework that concurrently couples macroscopic and microscopic length and time scales. By integrating the Finite Element Method (FEM) with Molecular Dynamics (MD) simulations, we investigate and enhance the predictive capabilities of material modeling, particularly under extreme conditions. We validate the approach by comparing to experimental and first-principle data, conduct a performance analysis and present an example of shock-driven flows in copper.

Introduction

To accurately predict fluid flows, understanding the behavior of materials over a wide range of conditions is essential. Particularly within high-energy-density physics, the deviation from equilibrium conditions largely reflects in the reaction of a material. These conditions often arise in extreme environments such as those found in inertial confinement fusion experiments (1). Traditionally, hydrodynamic simulations are used, but heavily rely on predefined equations of state (EOS) to capture the reaction of the underlying medium. These EOS describe the relationship between thermodynamic variables such as pressure, temperature, volume, and density. Their accuracy can be severely limited when materials are pushed far from their equilibrium states. This limitation arises as EOS are generally derived under the assumption of thermodynamic equilibrium.

The modeling of nonequilibrium behavior in materials is thus a major challenge in hydrodynamic simulations. The equation of state is coupled to the conservation equations for mass, momentum, and energy, and ensures that changes in density, pressure, and internal energy are consistently reflected across these equations. For example, a widely used equation of state in hydrodynamic simulations is the Mie-Grüneisen EOS (2) (3). Its parameters are derived from experimental measurements or computational simulations, thus restricting itself to near-equilibrium conditions. It therefore cannot adequately model nonequilibrium processes. Current state-of-the-art EOS designs counteract this by using the experimental and/or simulation results to choose functional forms for phase-dependent free energies (4). This allows for highly accurate multiphase EOS in a wider range, but still cannot guarantee accuracy when materials deviate significantly from the conditions under which their EOS was created.

It can be, however, a necessity to guarantee accuracy in these regimes. Nonequilibrium effects are often responsible for crucial processes such as energy dissipation and entropy production in turbulence (5), or phase transitions (6). In fact, nonequilibrium processes have been shown to lead to kinetic stabilization of metastable phases (7). These metastable phases can significantly influence transport coefficients in continuum mechanics. Understanding nonequilibrium effects is therefore essential to advance our description of the behavior of matter, and promises to greatly enhance the accuracy of computational fluid dynamics. Since analytical equations of state lack the atomic-level detail, alternative approaches for simulating materials under extreme conditions are needed.

One such approach is the equation-free method, which offers a powerful and flexible framework to bridge the gap between microscopic dynamics and macroscopic behavior without relying on explicit macroscopic equations (8) (9) (10) (11). By leveraging microscopic simulations, such as molecular dynamics or kinetic Monte Carlo methods, the equation-free approach informs the macroscopic state of a system. Thus, it bypasses the challenging or impractical derivation of an equation. The method is defined through iteratively *lifting* and *restricting* the system. The lifting operator translates macroscopic flow variables to microscopic states, and the restricting operator summarizes the microscopic results to update the macroscopic variables. Between the two operators, the system is evolved in the microscopic domain. Using these operators, a variety of sub-models can be assembled into a multi-scale model (12).

Therefore, the equation-free approach lifts the restrictions imposed onto an analytical equation of state. The responsibility to accurately model the material behavior is passed onto the microscopic dynamics. For nonequilibrium processes, especially for phase transitions, a molecular perspective has proven essential (6). Methods such as density functional theory (DFT) and quantum Monte Carlo (QMC) replicate material properties in detail by describing electronic structures. However, they require vast computational resources and are limited to small systems on short timescales. In contrast, molecular dynamics (MD) simulations can efficiently handle larger systems and longer timescales, a necessity when bridging to macroscopic domains. MD has also been shown to accurately capture complex phase transitions far from equilibrium (7). It is therefore the most performant option for the microscopic solver. By leveraging the strengths of MD, the equation-free approach ensures that the macroscopic predictions are based on atomistic interactions.

This paper presents the use of MD to capture nonequilibrium effects and inform a hydrodynamic simulation of its influence. We introduce a continuum-atomistic framework that concurrently couples macroscopic and microscopic simulations. Based on the equation-free approach, we bypass the formulation of an equation of state in hydrodynamic simulations. To achieve this, we introduce a series of operators to translate between the macroscopic and microscopic domain. We employ MD simulations to capture the atomistic behavior of the underlying material, and use the findings to update our macroscopic simulation. The concurrent continuum-atomistic framework contributes to more accurately simulate matter at extreme conditions, such as those found in inertial confinement fusion.

Methodology

The Macroscopic Model

The Euler equations provide a great case study for this research as they find a broad range of applications, such as vortex dynamics, wave formation and shock physics (13). Part of continuum mechanics, they encompass the equations for conservation of mass, momentum, and energy. A general treatment of the Euler equations is done using numerical schemes. Three main approaches are used in computational fluid dynamics: the finite difference method (FDM), the finite volume method (FVM), and the finite element method (FEM).

FDM's implementation is computationally efficiency and often straightforward, but it is challenged by unstructured grids and thus more complex geometries (13). FVM improves upon this limitation by using control volumes of various shapes. Nevertheless, it has significant computational overhead and a complex application of boundary conditions (14). FEM is highly accurate with unstructured grids and varying boundary conditions, and proves powerful for complex material properties and deformations. Mitigations to major weaknesses of FEM, such as its computational cost and complexity, are increasingly available (15) (16). Therefore, the finite element discretization scheme is suitable for a wide range of problems. This work uses the higher-order FEM Lagrangian hydrodynamics solver *Laghos* (16). *Laghos* is an open-source solver developed by the Lawrence Livermore National Laboratory (17).

The Microscopic Model

Atomistic simulations such as MD track individual atoms or molecules and their interactions for a deterministic evolution of the flow field by solving Newton's equations of motion. In MD simulations, the positions and velocities of atoms and molecules are updated over time by numerically integrating these equations. The forces that act on the particles result from interatomic potentials and external force fields. The simulation starts by initializing the positions and velocities of all particles. The force between each particle is then evaluated, and positions and velocities are updated. Various properties like temperature, pressure, and energy are obtained throughout the simulation by averaging the position and velocity of each particle (18). While it inherently captures the time and length scales of atomistic processes, computational advances have led to a bridge into experimentally observable time and length scales (19). MD thus provides a dynamic picture of molecular systems and enables the observation of diffusion, phase transitions, and other macroscopic properties in real time.

We use the Large-scale Atomic/Molecular Massively Parallel Simulator (*LAMMPS*) (20). It has a wide range of numerical integrators, interatomic potentials, and boundary conditions, and seamlessly scales from a single CPU to supercomputer architectures, which makes it particularly suitable for our approach. Despite these advances in microscopic modeling, it remains impractical to bridge molecular dynamics to hydrodynamic scales in both time and space. A coupled approach promises to maintain atomistic detail while simulating flow fields at the scales typically needed by engineering applications.

FEM-MD Coupling

To achieve this, many approaches have been developed. An excellent summary describing each in detail is provided by Lee & Basaran (21). Generally, these can be categorized into two main methods, shown in Figure 1. The handshake region approach combines MD and the finite element method by creating an overlapping region between the two domains. Within this region, MD particles and FEM nodes exactly overlap. This overlapping area typically spans a distance equal to the cutoff distance of the interaction potential used in the MD region, ensuring that all particles in the transition zone have a complete set of neighbors within their interaction range. The handshake region ensures consistent forces between both domains by considering the interaction with their neighbors through the MD interaction potential while also calculating nodal forces in the FEM mesh. Forces and displacements are thus consistently transferred between the MD and FEM regions. To conserve mass within the region, the mass of each node is set to the mass of the corresponding MD particle. Additionally, both the MD and FEM regions typically use the same numerical integration scheme. (22)

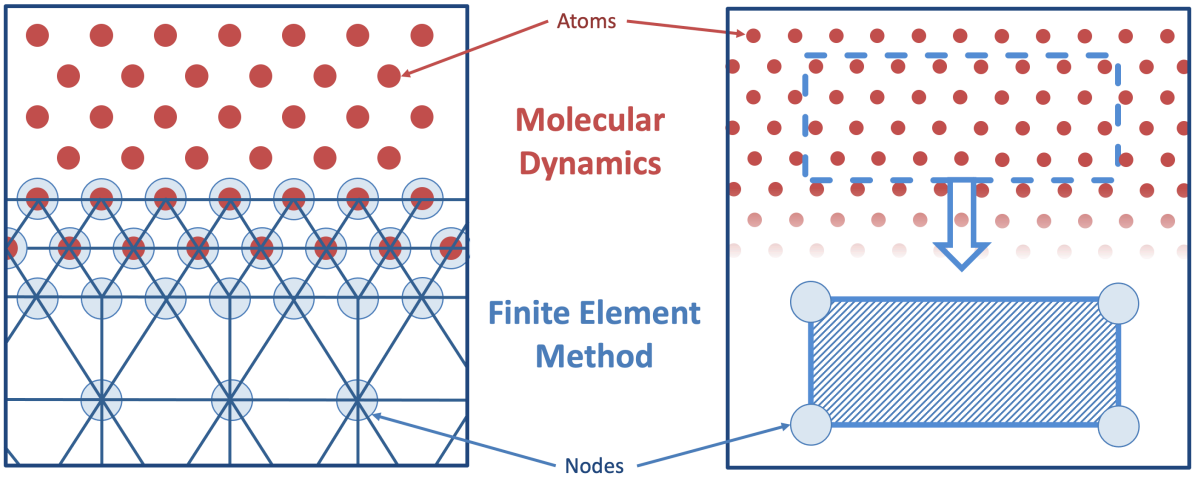

Figure 1: Two main FEM-MD coupling approaches exist. The handshake region (left) overlaps atomic particles and continuum nodes to pass information, while indirect approaches (right) use support simulations.

The advantage of a direct physical continuum-atomistic coupling in the handshake approach has proven successful in a wealth of cases (21) (22) (23) (24) (25). Its implementation is often straightforward, as the region is predefined in space. However, this introduces significant limitations. A fixed region where the coupling shall occur requires prior knowledge of areas of physical importance, which may not always be available. In addition, the overlap between particles and nodes constraints the FEM simulation to MD length and time scales. To assure numerical stability in FEM, hydrodynamic scales can only be recovered far from the handshake region. Furthermore, forces must be evaluated twice for both the MD domain as well as the FEM mesh for each point in space, introducing significant computational costs.

The indirect coupling approach provides a solution to many of these issues. One indirect coupling is the *atomistic finite element method* (AFEM). It uses MD as the driver code and employs the finite element method as a support simulation. By continually running an MD simulation, the AFEM captures atomic-level processes very well. It dramatically improves the accuracy of continuum FEM, as it accounts for multibody interactions. For cases where such detail is not required, it bridges into the continuum FEM domain through the translation of particles into nodes (26). This has the advantage of summarizing the actions of many particles into fewer nodes, which speeds up the computation time significantly. It also eliminates artificial interface errors often encountered in the handshake region. The coupling can also occur at any temporal and spatial point throughout the domain, lifting a critical restriction of the handshake region. The essence of this approach lies in the indirect connection between the two domains, and a variety of cases have shown the success of the indirect approach (26) (27) (28). However, given that MD is used as the driver code, it still limits all applications to near-atomistic scales.

Equation-Free Equation of State Coupling

We present a computational framework that bridges from atomistic to hydrodynamic scales by coupling MD to the finite element method. Part of the indirect approaches, we design FEM as the primary solver and invoke MD simulations when atomic-level detail is desired. The results are used to update material properties, such as stress, in the FEM domain. This effectively bypasses the equation of state evaluation. Figure 2 displays the main workflow of the approach.

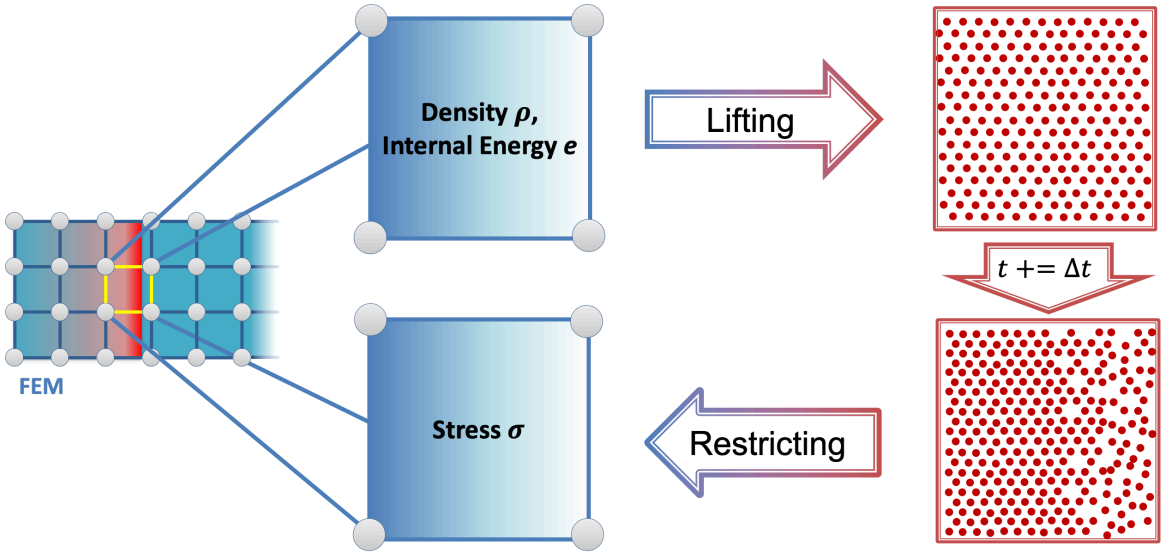

Figure 2: The equation-free EOS coupling uses the lifting operator to initialize a microscopic instance and the restricting operator to inform the macroscopic domain of material properties.

The method is comprised of two principal operators. The lifting operator translates macroscopic variables, such as density and internal energy, into microscopic counterparts like the number density and atom velocities. It is responsible for initializing the MD simulation with an atomic configuration that accurately reflects the macroscopic state. Any particle method is fully defined through position, velocity and interatomic potential, so the lifting operator ensures that all three are met accurately. The MD simulation can then be run. Upon

completion, the restricting operator summarizes the detailed microscopic information back into macroscopic flow variables used in FEM. In the case of hydrodynamic simulations, the material reaction is reflected in the stress tensor. The restricting operator updates the stress tensor based on the atomic-scale computes.

We implement the method in *Laghos*. Throughout every integration step, it calls a material model to obtain the stress tensor at current conditions. It is within this call that an instance of *LAMMPS* is created. The lifting operator initiates a microscopic simulation that runs concurrently, and the restricting operator returns the resulting stress tensor to *Laghos*.

Results

For validation purposes, an excellent material candidate is copper. Copper has well-documented experimental and theoretical data for a broad range of conditions and exhibits nonequilibrium effects under strong compression (7). We used the embedded atom method (EAM) interatomic potential created by Mishin et al. (29). Mishin's EAM potential has been widely adapted in the literature (7). As extreme conditions show large deviations in both pressure and temperature, our validation study must span these conditions. The calculation of the melting curve of copper ensures that a wide range of temperatures and pressures are tested, and supplies a quantitative measure to validate against. We obtain the melting curve using the Heat Until Melt (HUM) method, which gradually increases the temperature of a system until it transitions from a solid to a liquid phase as shown in Figure 3 for 100 GPa.

We compare the melting curve with experimental data of Tan et al. (30) and first-principles MD calculations of Baty et al. (31). Figure 4 shows that our automated MD method obtains general agreement. The quantum molecular dynamics data (blue) shows minimal deviations from our results at pressures below 50 GPa. More significant differences are observed at higher pressures, but the experimental data of Tan et al. (black) is matched very well in this domain. In addition, previous works have reported artifacts of the Mishin potential in the same range, indicating that the interatomic potential (rather than the method) is the cause of the discrepancies (32). Excellent agreement is once again observed at pressures above 200 GPa, with deviations of less than 3%.

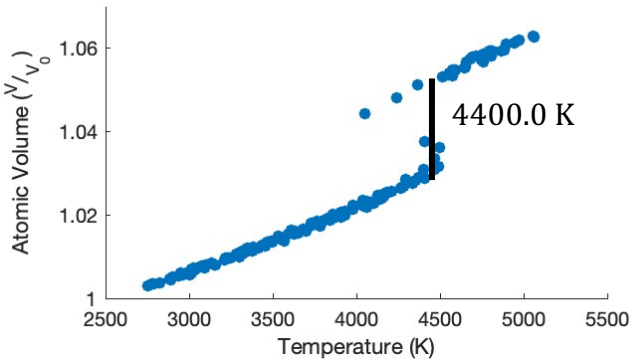

Figure 3: Using the HUM method, we found that a discontinuity in the atomic volume occurs at a temperature of approximately 4400 K, indicating that a phase transition likely occurs at this temperature.

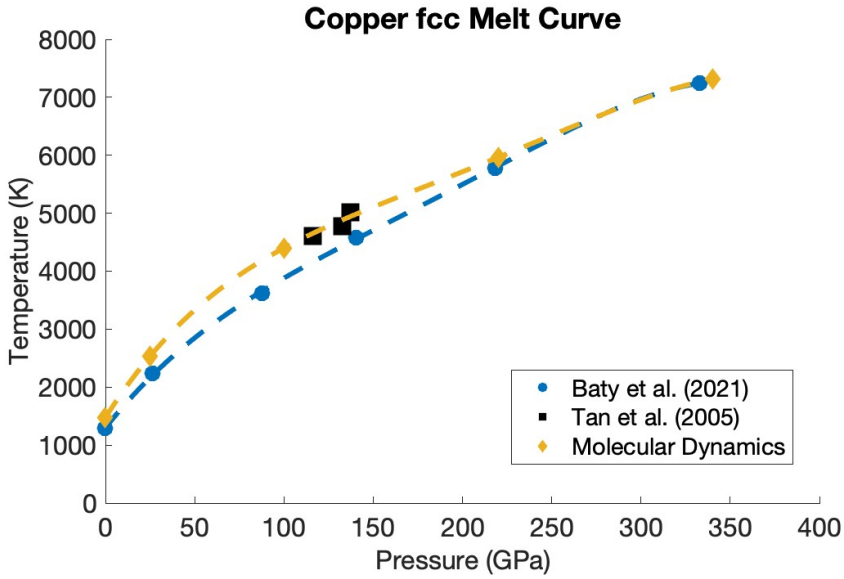

Figure 4: The melting curve of copper at various pressures is compared to experimental data (black) and first principles MD simulations (blue). General agreement is observed with slight deviations at lower pressures.

In addition to validity, a performant coupling is key for the practicality of our method. While the lifting and restricting operators perform algebraic calculations to set and retrieve data, the microscopic solver constitutes the majority of the workload. Efficiency in this step is crucial, and a determining factor of its performance is the number of atoms in the domain. To assist the effort of the MD simulation, we tested various system sizes to determine the minimum number of atoms necessary for accurate results.

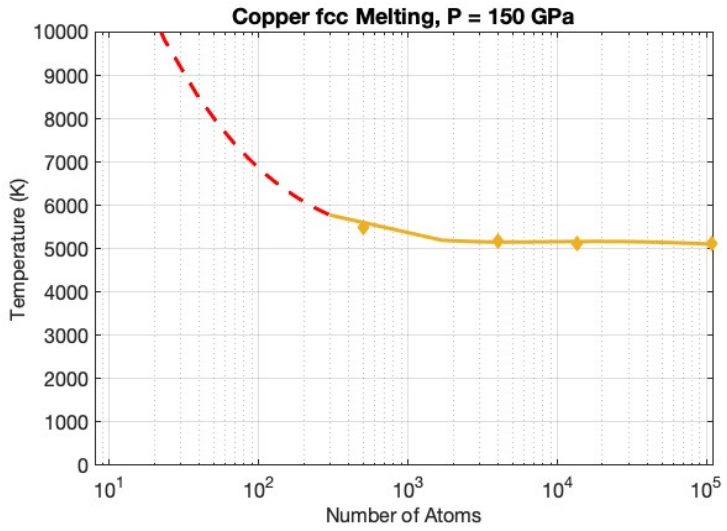

Figure 5: The system size analysis shows that fewer than 400 atoms fail to produce consistent results, 500 atoms show a 7.2% error, and systems with 5000 atoms or more achieve errors below 1%.

Figure 5 shows the melting temperature obtained at a pressure of 150 GPa for various system sizes. We observe unphysical behavior for systems smaller than 400 atoms. Strong fluctuations in the volume-temperature plots (such as Figure 3) fail to identify a melting point, deeming such small systems useless. Acceptable deviations are first observed for systems of 500 atoms, which show a 7.2% error. Error margins below 1% are observed for systems of 5000 atoms or more. The system's ability to capture the relevant physics highly depends on the physical behavior that is at play. It is also worth noting that the choice in system size should consider the availability of computational resources, which sees a direct correlation with accuracy in this case.

Shock Wave Simulation

We apply our method to simulate shock-driven flows, as they are characterized by large deviations from equilibrium. The simulation setup compares two scenarios: a FEM simulation with an analytical EOS and our FEM-MD concurrently coupled approach. Both cases use copper at an initial density of $\rho = 8.9 \frac{\mathrm{g}}{cm^3}$, an initial pressure gradient of $\Delta P = 170$ GPa and an internal energy of $e = 20$ kJ. Figure 6 shows the onset of the induced velocity field by the shock wave at $t = 1\ \mu s$. The top graph displays the use of an analytical EOS, and bottom graph shows the coupled FEM-MD method. In the analytical case, we observed a distinct onset of induced velocity and its evolution throughout the domain. This behavior is qualitatively matched by the FEM-MD approach. Quantitative agreement is seen in the shock wave's velocity magnitude, indicating that the FEM-MD approach, and especially the lifting and restricting operators, are functioning correctly. A difference in perturbation speed is observed, as the analytical case induces a higher velocity throughout the domain than the FEM-MD approach. This indicates a different sound speed prediction, a direct consequence of the equation of state. A possible cause of the deviation is that the copper parameters used in the analytical equation of state and the Mishin interatomic potential were obtained from different sources. To expect an exact agreement between the two methods requires an analytical equation of state derived from the same parameters as the interatomic potential. This is subject to future work.

Figure 6: A shock wave induces a velocity field. We analyzed its behavior using an analytical EOS (top) and MD-coupled approach (bottom).

Conclusion

We presented a novel framework for coupling the Finite Element Method with MD, enabling simulations that bridge between microscopic and macroscopic scales. The approach bypasses traditional equations of state, relying on atomistic simulations to describe material behavior. The automated coupling uses lifting and restricting operators that lift spatial and temporal scale limitations. The concurrent integration of MD into a hydrodynamic code is a major contribution to the field. Since particle simulations can readily be subject to large fluctuations

and errors if one chooses inaccurate procedures, such as thermostatting (33), a validation study is conducted. We then demonstrated the coupling using a shock-driven flow simulation. General agreement with an analytical EOS comparison suggests that our approach is a valuable addition to modeling techniques for non-equilibrium hydrodynamics. Its computational performance was optimized through a performance analysis, which suggests that microscopic systems should be of at least 500 atoms. While this research focuses on high-energy-density physics, the continuum-atomistic framework can be applied to a wide range of fields, including aerospace engineering, nanotechnology, and materials science.

Acknowledgements

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract No. DE-AC52-07NA27344 with release number LLNL-PROC-866098.

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