Ab-Initio Calculations of Materials Properties

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Introduction

Predicting and understanding the properties and behavior of real materials systems is of great importance both from technological and academic points of view. The theoretical problems associated with these systems are, of course, quite complex. However, we are currently at the forefront of beginning to overcome many of these problems. Our research is devoted to creating a realistic microscopic quantum mechanical description of the properties of real material systems. In the past, theoretical attempts to deduce microscopic electronic and geometric structure have been generally based on optimizing a geometry to fit known experimental data. Our approach is more fundamental: predicting geometric, electronic, and dynamical structure, *ab-initio* — that is, given *only* the atomic numbers of the constituent atoms as experimental input. Briefly, our method makes it possible to accurately and efficiently calculate the total energy of a solid by the use of density functional theory, pseudopotential theory and a conjugate gradients iterative minimization technique for relaxing the electronic and nuclear coordinates. *Ab-initio* investigations are invaluable because they make possible theoretical calculations or simulations that can stand on their own. They may complement experimental observations but need not be guided by experimental *interpretations*. Our objective is to obtain a fundamental, microscopic understanding of various physical and chemical phenomena of real materials systems.

Shock Waves in Solid Materials

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Study of the propagation of shock waves in condensed matter has led to new discoveries ranging from new metastable states of carbon[1] to the metallic conductivity of hydrogen in Jupiter[2, 3] to the possibility of delivery of life to planets via meteorite impact.[4] Shock waves are currently the only practical way to simultaneously probe the high temperature and high pressure behavior of matter, but progress in understanding the microscopic details of shocked materials has been extremely difficult. Highly non-equilibrium regions may exist that give rise to the formation of unexpected metastable states of matter and determine the structure, instabilities, and time evolution of the shock wave.[5-8] Some progress in understanding these microscopic details can be made through molecular dynamics simulations.[9-12] The popular nonequilibrium molecular dynamics (NEMD) approach to atomistic simulations of shock compression involves creating a shock on one edge of a large system and allowing it to propagate until it reaches the other side. The computational work required by NEMD scales at least quadratically in the evolution time because larger systems are needed for longer simulations. When quantum mechanical methods with poor scaling of computational effort with system size are employed, this approach to shock

simulations rapidly becomes impossible. Another approach that utilizes a computational cell moving at the shock speed has the same drawbacks.[13] This work presents a method which circumvents these difficulties by requiring simulation only of a small part of the entire system. The effects of the shock wave passing over this small piece of the system are simulated by dynamically regulating the applied stress which is obtained from continuum theory description of the shock wave structure. Because the size of the molecular dynamics system is independent of the simulation time in this approach, the computational work required to simulate a shocked system is nearly linear in the simulation time, circumventing the scaling problems of NEMD. Molecular dynamics simulations have been performed that utilize a shock Hugoniot-based thermodynamic constraint for the temperature at fixed volume.[14] This approach is a thermodynamic one for a single shock wave and fails to capture the spontaneous formation of multiple shock waves and dynamical effects like long-lived metastable phases, elastic-plastic phase transitions and chemical reactions, which are ubiquitous in shocked condensed matter. The new method outlined in this work is a completely general method for the dynamical simulation of shock waves that solves these problems. It enables the *dynamical* simulation of shock waves in systems that have material instabilities which lead to the formation of multiple shock waves and chemical reactions that can change the speed of shock propagation with time. It is a tractable method that requires no *a priori* knowledge of the system.

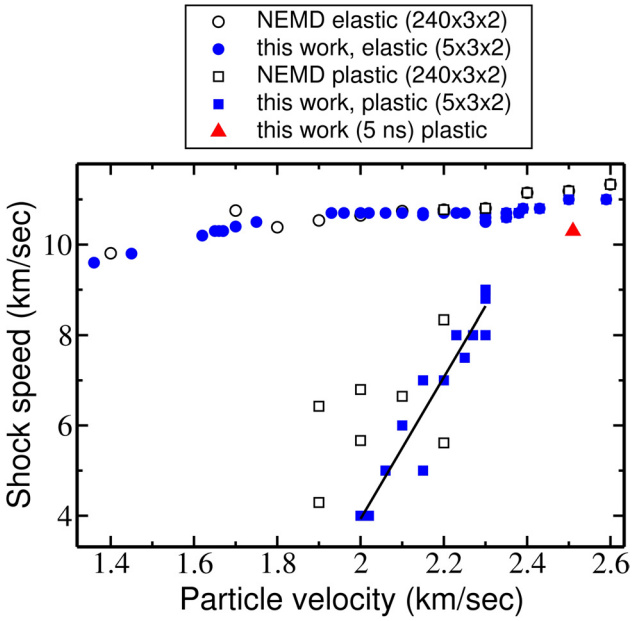

Figure 1. Comparison of calculated Hugoniots for the NEMD approach and the method presented in this work for roughly 10 ps runs. Note the ability to utilize much smaller computational cell sizes with the new method. Also included is one data point for a 5 ns simulation using our new method which would be prohibitive with NEMD, requiring a factor of 10^5 increase in computational effort.

As an illustrative example, we apply the new method to an elastic-plastic transition in a model potential for silicon. Figure 1 shows calculations of shock speed as a function of particle velocity (local speed of the material in the laboratory frame) for shock waves propagating in the [011] direction in Silicon described by the Stillinger-Weber potential.[15] Data calculated using the NEMD method are compared with results of the new method presented in this work. NEMD simulations were done with a computational cell of size 240 *x* 3 *x* 2 unit cells (5760 atoms) for a

duration of about 10-20ps. Simulations with the new method were done with a computational cell size of 5 *x* 3 *x* 2 unit cells (120 atoms). Both simulations were started at 300K and zero stress. Since the NEMD simulations were limited to the 10ps timescale by computational cost, simulations with the new method were performed to calculate the Hugoniot on this 10ps timescale for comparison. The final particle velocity in these simulations was taken to be a point of steady state after a few ps.

Figure 1 indicates a single shock wave exists below 1.9 km/sec particle velocity. Above this particle velocity, an elastic shock wave precedes a slower moving shock characterized by plastic deformation. Agreement between the two methods is good for all regions except for the plastic wave speed for particle velocities less than 2.1 km/sec. The wide range of values for the plastic wave speeds in NEMD simulations in this regime is due to lack of adequate simulation time to reach a steady state plastic wave speed. Plastic deformation occurs slowly in NEMD simulations in this regime, while it tends to occur all at once in the relatively small simulation cell used for the new method. Therefore the new method tends to reach the final particle velocity more quickly than the NEMD simulations. Better agreement on short timescales in this regime can be obtained by conducting longer NEMD simulations and by using a larger simulation cell for the new method simulations. One of the primary advantages of using the method outlined in this work is the ability to simulate for much longer times than is possible with NEMD. As an example, Figure 1 shows the result of a 5 ns simulation performed along a Rayleigh line corresponding to a shock speed of 10.3 km/sec. The uniaxially compressed elastic state required 5 ns to undergo plastic deformation. The difference in particle velocity between the 10 ps and 5 ns simulations at this shock speed is 0.8 km/sec, indicating the importance of the proper treatment of dynamical effects. This simulation done with NEMD would require *more* than 5 ns simulation time. For an $O(N)$ method of force evaluation, the computational cost of this simulation with the NEMD method would be at least 10^5 times greater, and therefore not tractable.

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