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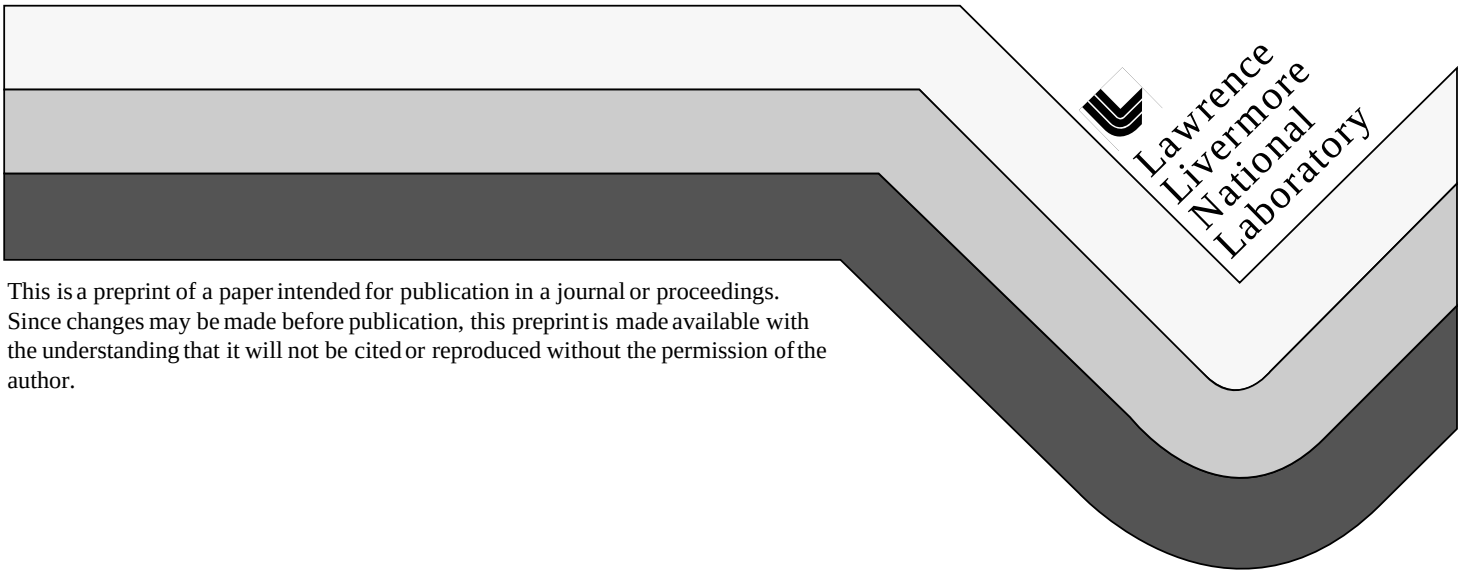

The DOE Accelerated Strategic Computing Initiative: Challenges and opportunities for predictive materials simulation capabilities

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Abstract

In response to the unprecedented national security challenges derived from the end of nuclear testing, the Defense Programs of the Department of Energy has developed a long-term strategic plan based on a vigorous Science-Based Stockpile Stewardship (SBSS) program. The main objective of the SBSS program is to ensure confidence in the performance, safety, and reliability of the stockpile on the basis of a fundamental science-based approach. A central element of this approach is the development of predictive, full-physics, full-scale computer simulation tools. As a critical component of the SBSS program, the Accelerated Strategic Computing Initiative (ASCI) was established to provide the required advances in computer platforms and to enable predictive, physics-based simulation technologies. Foremost among the key elements needed to develop predictive simulation capabilities, the development of improved physics-based materials models has been universally identified as one of the highest-priority, highest-leverage activity. We indicate some of the materials modeling issues of relevance to stockpile materials and illustrate how the ASCI program will enable the tools necessary to advance the state-of-the-art in the field of computational condensed matter and materials physics.

1. Background:

The Defense Programs (DP) office of the U.S. Department of Energy (DOE) has recently launched a series of new national security initiatives in response to the end of nuclear testing. Foremost among the drivers of these initiatives are the emerging and extraordinary technical challenges derived from President Clinton's 1993 decision to continue the U.S. moratorium on underground nuclear testing and to rapidly seek a Comprehensive Test Ban Treaty (CTBT). To support the President's decisions, DOE/DP has implemented a strategic plan – "DP-2010" – supporting a 15-year vision. Two critical elements of this strategic plan were the creation of a vigorous Science-Based Stockpile Stewardship (SBSS) program and the establishment of a steady-state weapon replacement capability (stockpile life extension program) to support reduced production demands and utilizing the technologies developed under the SBSS program.

Several key elements of the SBSS program utilize and critically depend on information and high-performance computing technologies. High-end computing is particularly important because advanced high-fidelity numerical simulations must ultimately replace the integrated "experiments" formerly performed in underground nuclear testing. Moreover, high-end computing is likely to be the single most important key technology in reducing production costs within the DP complex. Consequently, a cornerstone element of the DP-2010 strategic plan is to promptly shift from *empirical* nuclear-testing-based methodologies to *predictive* simulation-based methodologies for assessment, certification, and re-manufacture of nuclear weapons in order to maintain the performance, safety, and reliability of the U.S. nuclear weapon stockpile without testing. This is a challenge of unprecedented magnitude. For simulation capabilities to successfully fulfill this role will require an unprecedented increase in compute power – by as much as six to eight orders of magnitude – coupled with radical advances in the ability to simulate nuclear weapons as supported by application developments and computational performance.

The driving force for computing and simulation for nuclear weapons for the foreseeable future is the challenge of modeling weapon systems to a level commensurate with that formerly afforded by nuclear testing. The challenge is formidable because important phenomena in weapon systems are not fully understood from fundamental physical principles. Improved physics-based models will become necessary. In addition, computing power in excess of millions times more powerful than what exists today will be required to model important simulation sequences on the basis of empiricism-free, first-principles approaches based on improved sub-grid physics models.

2. The DOE Accelerated Strategic Computing Initiative:

In response to these formidable challenges, DOE/DP has developed an overarching strategy for national-scale high-performance computing. At the heart of this strategy is (1) an amplification of its existing Stockpile Computing program and (2) the launching of a new program – the Accelerated Strategic Computing Initiative (ASCI). The Stockpile Computing program embodies all the elements needed to maintain the current and anticipated DP high-end computing capabilities: computer operations, code development, code maintenance, and computer model development. On the other hand, ASCI is the strategic computing component of SBSS. The ultimate goal of ASCI is to provide the advanced computational and simulation capabilities needed to maintain a safe and reliable nuclear stockpile within the context of a zero-yield comprehensive test-ban treaty and the stockpile life extension program. Consequently, ASCI will advance the state-of-the-art in computational and simulation capabilities and ensure timely insertion of these leading-edge tools in current- and next-generation production codes. Foremost within the ASCI program is its vision of promptly shifting from a nuclear-test-based methodology to a computational-based methodology for issues of nuclear safety, reliability, performance, aging, and manufacturing. As a result, the ASCI objectives are mostly focused on the development of a new generation of codes which feature three-dimensional (3-D), high-fidelity modeling, "full-physics" sub-grid models, and full-scale systems. The timely insertion of ASCI-developed advanced capabilities is critical to ensure that DP retains access to the most comprehensive and up-to-date set of simulation capabilities in support of its SBSS missions.

A critical aspect of the ASCI application efforts is the development of truly predictive simulation capabilities. In the past, empirical physics models, normalized and adjusted on the basis of physics-integrated nuclear test data, were inserted in nuclear design codes, leading to limited predictive power. Presently, the absence of nuclear testing is driving the need to develop validated, predictive, first-principles physics models on the basis of theoretical and experimental activities addressing physics-sensitive weapons phenomena. Selected examples of physics-sensitive weapons phenomena include:

- three-dimensional, time-dependent phenomena,
- high explosive driven implosions,
- dynamic response of materials, particularly actinides, under extreme conditions: high temperatures, high pressures, and high-strain rates
- deuterium-tritium (DT) gas fusion ignition and burn,
- radiation transport and materials opacities, and
- instabilities and turbulent phenomena.

Computation-based "virtual testing" is the process by which these validated first-principles weapons physics models are integrated and coupled together

within full-physics, full-system applications capable of simulating nuclear performance in a truly predictive manner.

3. Materials theory, modeling and simulation

The development of improved physics-based materials models has been explicitly identified as one of the highest-priority, highest-leverage activities in the establishment of truly predictive simulation capabilities. Of particular importance is the prediction, on the basis of first-principles approaches, of the effects of aging- and manufacturing-related changes in materials properties on stockpile performance. Theoretically and computationally, such predictions are extraordinarily challenging and are at the very forefront of the field of theoretical and computational condensed matter and materials physics.

In what follows, we briefly describe selected areas of modeling and simulation of relevance to stockpile materials. These areas can be broadly classified in terms of their relevance to stockpile performance, stockpile aging, and stockpile manufacturing. In the present article, we will focus uniquely on issues germane to stockpile performance.

Of particular importance to the prediction of weapons performance is an accurate determination of the thermodynamical properties of materials, i.e. their equation-of-state (EOS), under a broad range of conditions of pressure, temperature, and density. In addition, since materials are subject to extreme strain and strain rates, the development of physics-based models to predict the high-pressure mechanical response (deformation and plastic flow, strength, spall, etc.) of materials is necessary. In terms of methodologies, thermodynamic properties are determined at the electronic/atomic level. On the other hand, mechanical properties are determined by the collective motion of defects at different length scales and their prediction requires a multi-length-scale approach.

We now briefly illustrate issues related to the prediction of the thermodynamic and high-pressure mechanical properties of stockpile materials. These include metals and alloys – with particular emphasis on actinides, hydrogen, and body-centered cubic transition metals – and high explosives and organics

4. Thermodynamic properties:

4.1 Actinides

Existing equation-of-state (EOS) theories break down in the δ phase of plutonium (Pu) where it is known that strong electron-electron correlation is inadequately treated by standard (mean field) *ab initio* electronic structure techniques, in contrast to the more weakly correlated behavior characteristic

of the α phase. Because of the electronic nature of the δ – α transition, it is believed that remnants of the dramatic (~ 20 %) volume change observed in the solid-solid transition will persist in pronounced modulations of the EOS surface well into the liquid, and will be critical to the calculation of the expansion EOS of Pu. Similar transitions occur in a number of lanthanide and actinide metals under pressure. Consequently, the development of advanced electronic structure methods beyond mean-field implementations is necessary for the treatment of strongly-correlated electron systems. A combination of recent effective Hamiltonian and quantum Monte Carlo (QMC) techniques developed specifically for strongly-correlated electrons systems has been developed at Lawrence Livermore National Laboratory (LLNL) to directly address this problem. In particular, this approach allows the determination of the distortions of the EOS surface as a function of temperature (the critical region), and affords an elucidation of the very nature of the electron-thermal EOS on either side of the critical region.

4.2 Hydrogen

Important aspects of the hydrogen EOS, especially molecular dissociation and electronic excitation, are poorly treated by *ab initio* self-consistent field methods based on density functional theory. However, such phenomena can be accurately treated by modern path-integral quantum Monte Carlo (QMC) techniques. Of particular interest are EOS regions of maximum uncertainty (approximately 1 – 5 Mbar on the Hugoniot) and isentropic compression to high densities from 0.5 – 3 Mbar in shocked states of D_2 and DT. In addition, the more empirical finite-temperature tight-binding molecular dynamics (TBMD) method has also been applied to this system. The matrix elements describing this model can be determined through extensive comparison with molecular and high-temperature limit properties. Calibrated against the QMC results, the TBMD approach is expected to provide a means of rapidly extending these results to a wide range of densities and temperatures.

4.3 High explosives and organics

Current production hydrocodes used in weapons simulation incorporate an EOS of high explosives (HE) that is empirically extracted from hydrodynamics experiments. Such empirical HE EOS's have limited predictive power because they follow only a single track in phase space. Explicit use of thermodynamics combined with kinetics is the next important step in performance prediction. Modern HE EOS thermochemical codes are based on the use of reliable statistical mechanical theories, molecular interaction potentials, and their ability to handle multi-phase fluid mixtures.

In addition to the EOS issues mentioned above, fundamental questions remain unresolved in HE detonation physics and chemistry, in part because of the difficulty of addressing very short-time-scale processes. A clear example illustrating this point is the role of carbon condensation in the

detonation process – carbon being a key component of conventional HE's (37 and 45 mole % for TATB and TNT respectively). As carbon condenses into nano-scale clusters, a pressure-induced phase transformation from graphite to diamond occurs. This phase transformation in carbon nanoclusters is empirically modeled within HE performance codes. For example, state-of-the-art thermochemical calculations of the detonation velocity for TNT differ from experimental measurements by as much as 5 % under some conditions. This disagreement is resolved in an empirical manner by postulating that the graphite – diamond transition is shifted to significantly higher pressures than the bulk equilibrium value. It is well established that free carbon forms clusters of nanoscale dimensions (~ 10's of A) during the detonation process, and it is believed that this phenomenon is at the origin of the observed discrepancy between the theoretical models and the experimental detonation velocity data.

Nanoscale materials exhibit properties that differ markedly from bulk systems in that they are composed of a limited number of atoms (~ 10^3 – 10^5 atoms) and surface energy considerations become increasingly important. It is anticipated that the graphite – diamond transition will occur at higher pressures with decreasing crystallite size, though this has never been established either theoretically or experimentally. Moreover, the kinetics of the transition are expected to change markedly as a function of pressure as well as crystallite size. Consequently, the prediction of pressure-induced structural phase transitions in carbon clusters which are important to the determination and modeling of HE performance.

5. High-pressure mechanical properties:

5.1 Multi-length scale materials modeling of plastic flow

The wide variety of materials models needed for the different computer-code simulations of weapons systems is overwhelming. Several of the modeling requirements can be fulfilled on the basis of descriptive models validated by laboratory test data. However, one specific modeling area where such descriptive materials models are qualitatively inadequate is the description of materials which undergo extreme plastic deformation under conditions of high pressure and high temperature. The predictive modeling of plastic deformation of materials under extreme conditions of pressure and temperature is difficult because laboratory testing is impractical or in some cases impossible. Incorporation of fundamental physics-derived laws and rules into practical continuum-based materials models, so that behavior can be extrapolated under extreme conditions of pressure and temperature, is currently the only way to develop truly predictive materials modeling capabilities and fulfill requirements for fully-integrated simulation codes. Current empirical materials models embody uncontrolled approximations

and, as a result, cannot be systematically improved on a firm scientific physical basis.

In response to the challenge of developing predictive models of the high-pressure properties of materials, LLNL has launched an integrated program to develop and implement methodologies to predict the mechanical response of stockpile metals on the basis of a physics-based multi-length-scale approach. Such an approach provides the framework to link theoretical approaches at the electronic, atomic, microscopic, mesoscopic, and continuum levels. Of particular importance is the establishment of rigorous theoretical links between quantum-based descriptions at the electronic and atomic levels and engineering continuum-based treatments at the macroscopic level through the identification of appropriate degrees of freedom and internal variables to describe the microstructural evolution which determines the materials mechanical response at high pressure.

Several metals and alloys of interest to DOE/DP exhibit the body-centered cubic (bcc) crystal structure. These include tantalum (Ta), molybdenum (Mo), vanadium (V) and iron (Fe). Moreover, Ta is currently used in "integrated" hydrodynamic experiments. The mechanical properties of bcc metals, especially the low temperature deformation behavior, exhibit several unique characteristics which distinguish them from that of other crystal structures like face-centered-cubic (fcc) and hexagonal-close-packed structure (hcp). These features include a rapid increase of the yield and flow stresses with decreasing temperature, a marked sensitivity of the stress to the imposed strain rate, and a tendency in many cases to brittle cleavage fracture at low temperatures. Intrinsically, the technical difficulty for modeling bcc transition metals (Ta, Mo, etc.) lies on its plastic anisotropy which exhibits strong dependence on temperature and orientation. Experimental evidence indicates that dislocations, especially screw dislocations, in bcc transition metals play a significant role in controlling the plastic deformation of bcc metals. Hence, the understanding of the unique properties of dislocation core and dislocation interactions in bcc metals is critical to enable the successful prediction of the plastic flow of bcc metals.

The metals Mo and Ta have been chosen as protopypical materials systems for their leverage in DOE/DP applications. Both Mo and Ta exhibit the bcc structure, which is representative of other materials of importance to DOE and the Department of Defense (DoD) (Fe, etc.). The technical program at LLNL includes the following activities:

- *Ab initio* electronic structure calculations of point defects and dislocations to determine energetic and fundamental length scales
- Atomic-level simulations of dislocations and dislocation interactions to derive rules and mechanisms for microstructure-level simulation

- Three-dimensional (3-D) dynamic dislocation simulation of microstructure and plasticity studies, mesoscale studies of polycrystalline grains to develop improved plasticity models, and
- Connection with mesoscale and continuum-based simulations based on improved anisotropic plasticity models.

5.2 Predictive materials failure models

Understanding and modeling materials failure during aging or dynamic shock loading is of critical importance to DP programs. In particular, when tensile shock waves overlap, the material experiences an extreme state of tension. Depending on the magnitude and duration of this state of tension, voids or cracks nucleate and grow. When the voids link up, the material fails. This process is known as spallation. Of particular interest is the development of models of materials failure during long-time aging and dynamic shock loading that correlate microstructural characterization with observed materials failure. Specific technical areas of interest include:

- Simulation of void nucleation processes and correlation to microstructural characterization, including grain distribution and texture, impurities, and pre-existing distribution and concentration of voids/vacancies.
- Models of void growth and linking in the presence of complex microstructure.
- Fundamental fracture precipitation and propagation models in metals and polymers which incorporate atomic-scale driving phenomena but address fracture at longer length scales (meso- and continuum-scale) and time scales.

Current materials models for spall and failure under shock loading are phenomenological in nature and do not incorporate materials microstructure or the effects of aging. An important goal of the ASCI program is to develop physics-based materials models for spall and ejecta through large-scale atomic-level simulations, thereby leading to an improved understanding of the fundamental processes that cause materials failure under shock loading conditions. *Ab initio* electronic structure and molecular dynamics (MD) methods can serve as the basis to study the effects of materials microstructure on dynamical phenomena. These include processes such as spall, yield strength, multi-phase EOS, ejecta, chunk/atomic mix, and turbulence. In particular, the development of predictive microscopic models for defects and defect-defect energetics is important. These defect models can subsequently form the basis to the development of mesoscopic models of materials microstructure and microstructure evolution. In support of this effort, large-scale atomic-scale MD simulations are being performed on scaleable multi-processor ASCI high-performance computing platforms to predict the effect of internal materials microstructures, impurities and surface roughness on determining the volume and nature of materials ejecta leading to mix.

The surge in high-performance computing capabilities afforded by the DOE ASCI program will enable routine atomic-level simulations containing 10^8 or more atoms. This capability can be used to perform large-scale MD simulations of shock-loading of grain boundary junctions, large voids, and rough surfaces. *Ab initio* electronic structure calculations are being performed to develop improved impurity potentials and interactions at grain boundaries. In addition, kinetic Monte Carlo (KMC) methods are used to study long time scales and impurity migration, thus forming the basis to the development of improved nucleation and growth model for voids.

6. Summary

The objective of the DOE Science-Based Stockpile Stewardship (SBSS) program is to ensure confidence in the performance, safety, and reliability of the U.S. nuclear stockpile without nuclear testing on the basis of a vigorous science-based approach. A critical element of this approach is the development of predictive, first-principles, full-physics computer simulation tools and advanced experimental facilities. In support of the SBSS program, the DOE has launched the Accelerated Strategic Computing Initiative (ASCI) to enable these computational developments and to promptly shift from an empirical test-based methodology to a predictive simulation-based approach. In particular, the development of advanced materials simulation capabilities to predict the effects of materials properties – as these properties change as a result of aging and/or re-manufacturing – on stockpile performance has explicitly been identified as one of the most critical component of the SBSS program. Consequently, the emerging SBSS program at the national laboratories presents both real opportunities and unprecedented challenges for solving important materials physics problems of national significance. A key element in the development of predictive materials simulation capabilities is the establishment of rigorous theoretical links between *ab initio* quantum-based descriptions at the electronic and atomic levels and engineering continuum-based treatments at the macroscopic scale. These links can be established through the identification of the appropriate degrees of freedom which determine the materials response. In this article, we indicated applications which illustrate the use of advanced materials simulation methods for the prediction of the thermodynamic and high-pressure mechanical properties of stockpile materials.

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