Multiscale strength (MS) models: their foundation, their successes, and their challenges

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Abstract. Multiscale strength (MS) models are constructed to capture a natural hierarchy in the deformation of metals such as V and Ta starting with atomic bonding and extending up through the mobility of individual dislocations, the evolution of dislocation networks and so on until the ultimate material response at the scale of an experiment. In practice, the hierarchy is described by quantum mechanics, molecular dynamics, dislocation dynamics, and so on, ultimately parameterizing a continuum constitutive model. We review the basic models and describe how they operate at extremely high pressures and strain rates, such as in Rayleigh-Taylor plastic flow experiments. The models use dislocation density as a state variable, and describe time-dependent, as well as rate-dependent, plasticity. They make interesting and testable predictions about transients in plastic flow. There are also clear challenges, however. The current MS models do not include a variety of mechanisms known to be important at low rates. Still, MS models provide compelling insight into plastic deformation of metals under extreme pressures and strain rates.

1. Introduction
The strength and flow stress of solid materials is related at the microscopic level to the motion of defects, especially dislocations, line defects in the crystal lattice. The amount of shear stress needed to plastically deform materials at macroscopic lengths is ultimately related to the strength of electronic bonds between atoms as those bonds bend and break during dislocation motion. It is not just the strength of those bonds, however. The population of dislocations and how they interact matters, too. Plasticity is a multiscale problem. Multiscale models are designed to describe those interactions across many scales to predict the macroscopic behavior of materials. For plasticity the model may be formulated in a hierarchical, information passing approach in which the relevant quantities characterizing plasticity at one scale are passed to the model at the next longer length (and time) scale.

Here we describe the application of multiscale modeling to the problem of the flow stress of a solid material under high pressure driven at high strain rate [1]. Due to the effect of inertial confinement, dynamic experiments are able to achieve high pressures, beyond the limits of confining materials like the diamonds in a diamond anvil cell. The tradeoff is that the experiment only lasts a brief period of time. Ramp compression provides a means to attain dynamically compressed states at Mbar pressures.
well below the melt curve, by minimizing shock heating. Techniques based on the Rayleigh-Taylor instability have been developed over the past few decades to infer the flow stress in ramp-compressed materials, and to test constitutive models against the observed materials behavior [2-7]. We also consider the response of dynamically loaded material at the level of unit cells in the lattice, as probed by in-situ x-ray diffraction [8-10].

2. The Formulation of Multiscale Strength Models

We begin with a review of how a multiscale strength model is constructed [11-13]. At the atomic level, bonds form between atoms. The behavior of the electrons in those bonds is described by quantum mechanics. Modern Density Functional Theory (DFT) codes solve the Kohn-Sham equations from first principles to predict the quantum mechanical total energy of a small representative portion of a crystal, and thereby capture the nature of the quantum mechanical bonds relevant to plasticity. With systems of up to about 1000 atoms, DFT can predict the energies of perfect and defective crystalline structures (vacancy and interstitial formation energies, dislocation line energies, twin boundary energies, gamma surfaces, etc.) and minimizing those energies leads to predictions of defect structures. Calculations across a series of deformations give elastic constants, the ideal shear strength or the Peierls barrier [11,14-15]. These calculations can be done at the density corresponding to ambient pressure, or they can be done for more compressed systems to give high pressure. Furthermore, finite temperature properties can be obtained with quantum molecular dynamics simulation in which the atoms move according to forces calculated with DFT.

DFT is limited to systems of ~1000 atoms due to computational cost. Classical molecular dynamics (MD) using interatomic potentials is able to treat systems that are still small, but considerably larger than those accessible to DFT. MD has been used to simulate dynamic processes in systems with many billions of atoms, roughly a cubic micron in size, for nanoseconds [16], although few-million-atom simulations are more typical. MD is well suited for calculating important processes in plasticity, such as the mobility of dislocations [17,13] and the growth of deformation twins [18]. In many cases this kind of MD simulation is done with an empirical potential, an interatomic force law that is fit to experimental data; however, it is also possible to construct the functional form of the potential and to calculate its parameters from first principles. One example is the Model Generalized Pseudopotential Theory (MGPT). The functional form is derived from a quantum mechanical empty-core pseudopotential tight-binding model [19], and the parameters are calculated using DFT [20]. This approach gives a quantum-based potential that provides the accuracy needed for dislocation simulations at a computational cost that is substantially less than DFT. The MGPT potentials for the body-centered cubic (bcc) metals Ta and V have been used for multiscale modeling [13].

A key piece of information for multiscale strength modeling is the mobility of a dislocation, a line defect in a crystal (see figure 1). The mobility determines the velocity at which a dislocation moves under a given resolved shear stress, at specified conditions of pressure, temperature and perhaps other non-zero stress components. The mobility depends on the character of the dislocation and its glide system. In bcc metals there is a high lattice resistance to the motion of screw dislocations. A certain amount of stress is needed to get a dislocation to move at zero temperature from one lattice site to the next, and for bcc metals that Peierls barrier is high and makes a significant contribution to the yield strength and flow stress of the material.

MD simulations can be conducted in which an edge or screw dislocation is introduced into an otherwise perfect crystal at a temperature and pressure of interest. After equilibration, a shear stress or shear strain can be applied. Following an initial transient, the dislocation will reach a steady state velocity. The mobility is determined by a series of those simulations, each determining one datum of velocity vs. shear stress [13]. In some cases the dislocation may cross-slip or climb. It may leave point defect debris [17], as shown in figure 2. MD is able to capture all of these effects.
At the next longer length scale dislocation dynamics is used. As materials strain harden, the dislocation density builds up and the dislocation-dislocation interactions become important. Since there may be many atoms in the regions between dislocation cores, an approach that tracks only dislocation core positions, not atomic positions, is more efficient. Dislocation dynamics tracks the core positions, calculates the dislocation stress fields and the forces they exert on dislocations, and moves the dislocations accordingly [12]. The motion of the dislocations is governed by the mobility law calculated in MD. Other model parameters such as the elastic constants are calculated with DFT. So this model, too, is ultimately grounded in first principles calculations rather than empiricism.

The final scale of the methodology is continuum plasticity. At this level the model provides a relationship between shear stress and strain rate at specified thermodynamic conditions (temperature and pressure). The model also tracks the dislocation density as a state variable. It plays a similar role to plastic strain in other models; however, unlike most formulations based on plastic strain, the dislocation density obeys a first order ordinary differential equation (ODE). As in the Gilman model [21], the ODE describes the multiplication of dislocations at low dislocation densities, and the increasing importance of annihilation at high dislocation densities so that the dislocation density tends to a rate-dependent saturation density. The equations of the continuum model, the Livermore Multiscale Strength (LMS) model [13] are shown in figure 3. This model is similar in form to a Mechanical Threshold Stress (MTS) model [22], but with the parameters calculated at lower length scales based on first principles within the hierarchical framework.

Recently a reformulation of the LMS model has been proposed [23]. The new model is no longer based only on first principles calculations and hierarchical information passing. It includes extensions to the model forms to give better behavior at low strain rates, and some of the parameters have been fit to agree with experimental data. The work also introduces a generalization of the LMS model that includes thermal softening through the shear modulus $G(P,T)$ [23], which makes a difference at higher temperatures such as in shock loading [10]. We use the original LMS model [13] in the discussion below since it has been used for the majority of the simulations of Rayleigh-Taylor strength experiments to date.
3. Plasticity in Ramp Compression

At quasi-static rates the yield strength and flow stress of a material can be measured with mechanical tests, which offer a fairly direct measure of the stress-strain behavior of a material. At Mbar pressures but low rates, diamond anvil cell techniques are available to characterize the strength of a material [24-25]. At high pressure and high rates different approaches are needed. A technique based on the Rayleigh-Taylor instability (RTI) has been developed to test constitutive models against the observed material behavior and to infer the flow stress in ramp-compressed materials, albeit in a model-dependent way [2-4,6]. The RTI is well known for fluid dynamics, where perturbations grow on the interface between a low-density material accelerating a high density material. If a layer of water were situated above a layer of air in a gravitational field, the surface of the water would be unstable and small perturbations on the surface would grow until ultimately spikes of water would fall down through the air and bubbles of air would rise through the water. Laser-driven ramp compression is able to generate sufficiently high accelerations for this phenomenon to occur for solids. The growth of the perturbations involves shear flow, so material strength slows the growth and the growth rate may be used to infer the flow stress of a metal at high pressure and high rates. Laser-driven RTI experiments have been carried out on vanadium [5-6] and tantalum [7], and the ripple growth measurements at a pressure of roughly 1 Mbar have been found to be in good agreement with the growth as calculated by continuum modeling of the experiments using the LMS models for those metals. The peak flow stress is found to be much higher than at ambient conditions: ~2.5 GPa in V [5] and ~6 GPa in Ta [7].

The ripple growth in these experiments takes place over a period of about 20 ns [5-7]. The time needed for the dislocation density to multiply to be roughly equal to the saturation density in the LMS model is on the order of a nanosecond, as explained below. So the plastic flow may be roughly considered to be saturated during the period of ripple growth. At very early times during the
compression, the dislocation density is not saturated, and it grows exponentially, but for the period of high RTI growth rate it is approximately saturated, somewhat under-saturated as the pressure increases and somewhat supersaturated as the pressure releases. To understand how the LMS model [13] behaves in this regime, it can be helpful to consider the saturated LMS model, setting the dislocation density to its saturation value: \( \rho = \rho_{sat}(\dot{\varepsilon}_p) \). With this approximation, the dislocation density evolution differential equation drops out. Since that ODE is the only time dependence in the LMS model, the simplified constitutive response gives a flow stress that only depends on the thermodynamic conditions (e.g., pressure and temperature) and the plastic strain rate [26]. The flow stress can be analyzed to see which parts of the model are most important: work hardening \( (M\dot{\varepsilon}) \), thermal activation \( (M\dot{\tau}_p\bar{\tau}_{therm}) \) or drag \( (M\dot{\tau}_p\bar{\tau}_{drag}) \). There is also a small athermal term which we will not consider. The results are shown in figure 4 for Ta at 1 Mbar and 1660 K, conditions characteristic of a shock to 1 Mbar followed by a ramp. Similar plots have been presented in reference [26] for adiabatic compression (430 K) and the RTI experiment (1270 K). In the shock-ramp case shown here, there is a cross-over from mobility dominated by thermal activation to mobility dominated by drag at a strain rate of 8.3x10^7/s. At fixed pressure this cross-over occurs at a higher strain rate at lower temperatures. Figure 5 is a plot of the cross-over strain-rate at temperatures on the adiabat, on the principal Hugoniot, and at intermediate temperatures. The 1 Mbar RTI experiment corresponds to 1 Mbar on the green curve. Using this analysis it is possible to determine which aspects of the LMS model are tested with the RTI experiments.

![Figure 4](image1.png)

**Figure 4.** Decomposition of the saturated LMS flow stress into contributions from thermal activation, drag and work hardening for Ta at 1 Mbar and 1660 K, conditions characteristic of a shock to 1 Mbar followed by a ramp at the indicated plastic strain rate. The approximate power law scaling of the different terms with plastic strain rate \( \dot{\varepsilon}_p \) is shown in the upper left.

![Figure 5](image2.png)

**Figure 5.** A plot of the strain rate at which the LMS mobility for Ta crosses over from the thermal activation regime to the drag regime as a function of pressure in four cases: on the room temperature adiabat (blue), on the principal Hugoniot (red), and at two intermediate temperatures between the adiabat to the Hugoniot. The 1 Mbar Ta RTI experiment is on the \((1/3)T_A + (2/3)T_H\) curve.

The three contributions to the saturated flow stress exhibit characteristic power-law scaling with the strain rate at high strain rates. Consider the work hardening term. Since the saturation density scales with strain rate as \( \rho_{sat} \propto (\dot{\varepsilon}_p)^n \), the work hardening goes like \( \dot{\varepsilon} \propto \sqrt{\rho_{sat} \propto \dot{\varepsilon}_p^{n/2}} \). Similarly, the
drag term at high strain rates goes like \( \tau_{\text{Drag}} \propto \dot{\varepsilon}_p / \rho_{\text{sat}} \propto (\dot{\varepsilon}_p)^{1-a} \), provided the velocities are non-relativistic \((v \ll c_0)\). The thermal activation term also shows a power-law behavior at high rates, with an exponent that is temperature dependent. The numerical values of the exponents are shown in figure 4. The different exponents lead to different terms becoming the most important in different regimes of strain rate. In figure 4, work hardening makes the largest contribution for \( \dot{\varepsilon}_p < 8.3 \times 10^7 / s \), transitioning to drag making the largest contribution for higher rates. In the 1 Mbar Ta RTI experiments most of the ripple growth happens at strain rates of a few times \(10^7/s\). While there are transients that are in the drag regime, in practice the experiments are sensitive to the work hardening, including the saturation dislocation density, and the thermal activation part of the mobility \([7,26]\).

4. Time-dependent Plasticity and 1D-3D Transition

If the dislocation density is not saturated, the LMS model is a model of time-dependent plasticity. The dislocation density evolves toward the target saturation density. This evolution sets a time scale for the model, and this time scale can have interesting implications for experiments, especially for laser-driven deformation experiments in which the time scale of the deformation and the resolution of the diagnostics are comparable to the time scale for plastic relaxation.

There are two time scales of particular interest associated with plastic relaxation in models like the Gilman model and the LMS model: the incubation time \( t_{\text{inc}} \) and the flow time \( t_{\text{flow}} \) \([27]\). These time scales describe how long the system takes to respond when the applied stress exceeds the work hardening and athermal contributions to the strength. For the purposes of this discussion we consider an instantaneous jump in applied shear stress, and how the system responds. The first time scale governs the rate of dislocation density build up (incubation), and the second time scale governs the rate at which shear stress decreases once the dislocation density has built up sufficiently to allow appreciable plastic flow. In principle both of these time scales are observable in experiment, although to date they have only been bounded \([9-10]\). They may also be studied in MD \([27-29]\) and DD.

A derivation of analytic formulas for \( t_{\text{inc}} \) and \( t_{\text{flow}} \) is given reference \([27]\). The model used there was a variant of the Gilman model, which is closely related to, but not identical to, the LMS model. It is useful to revisit that analysis for the LMS model. The incubation time is defined by the dislocation density evolution equation, equation (5) in reference \([27]\):

\[
\frac{d\rho}{dt} = t_{\text{inc}}(1 - \rho / \rho_{\text{sat}}) \rho.
\]

This equation holds approximately in the LMS model provided the dislocation density is approximately constant during incubation, \( v \equiv v_0 \). Then the incubation time is given by the constant \( t_{\text{inc}} = M / (\eta R v_0 b) \), as in equation (6) in reference \([27]\). \( v_0 \) is determined by the initial shear stress and the mobility. In the early stages of the response the dislocation density grows exponentially, \( \rho = \rho_0 e^{t/\tau_{\text{inc}}} \). In the LMS model the increasing dislocation density causes work hardening which slows down the dislocations and we have not derived analytic expressions for the late-time dislocation density that includes this effect, but comparisons made in reference \([27]\) show that for shock loading the initial exponential rise in the dislocation density is well described and the dislocation density rises to a peak value, for which an analytic estimate is given. A formula for \( t_{\text{flow}} \) has also been derived. The rate of change in the shear stress due to plastic relaxation is given by \( \dot{\sigma} = -2G \dot{\varepsilon}_p = -2G \eta b \rho v / M \). The mobility law can be inverted to express \( v \) in terms of \( \sigma \). If the dislocations are in the non-relativistic drag regime, the result is particularly simple: \( \sigma = -t_{\text{flow}}(\sigma - M \sigma_f) \), where \( \sigma_f = \beta b G \sqrt{\rho + \tau_0} + \chi_0 \chi R_v \), and \( t_{\text{flow}} = M^2 \chi_0 \tau_p / (2G \eta b \rho c_0) \) \([30]\). If the dislocation density and pressure are constant, the shear stress decreases exponentially toward a value of \( M \sigma_f \) with the time constant \( t_{\text{flow}} \), and roughly so even if \( \rho \) is only approximately constant. Of course, as the shear stress decreases, at some point the dislocation velocity leaves the drag regime and the approximations made for the simple formula break down. The
overall analysis does provide a good description of how time dependent plasticity operates in the 1D to 3D transition (the transition from an elastic uniaxial state to a plastic triaxial state) in shock loading in the case when the shock strength is below the threshold for homogeneous nucleation. The total relaxation time is given by $t_{\text{tot}} \approx t_{\text{inc}} \log \left( \frac{12 \rho_{\text{peak}}}{\rho_0} \right) + t_{\text{flow}}$ [27], which could be tested in in-situ diffraction experiments. This formula says that the dislocation density builds up exponentially from its initial value $\rho_0$ to its peak value $\rho_{\text{peak}}$ taking $t_{\text{inc}}$ for each e-fold increase. The initial dislocation density $\rho_0$ is a measurable property of the starting material. A formula for $\rho_{\text{peak}}$ is given in reference [27]. Intuitively, a measurement of $t_{\text{inc}}$ tests the dislocation multiplication rate $R$, and $t_{\text{flow}}$ tests the mobility and the peak dislocation density, which is sensitive to the saturation density. The relaxation time is predicted to be $\sim 3$ ns at 50 GPa, and greater at lower pressures. Above homogeneous nucleation threshold, the incubation time becomes a few picoseconds, reducing the predicted relaxation time to $\sim 1.5$ ns or less [27].

5. Additional challenges for multiscale strength modelling

The LMS model has agreed well with RTI experiments at high rates and high pressures. This agreement is remarkable for a model derived from first principles with essentially no empirically adjusted parameters. In constructing the model, rational choices have been made about what mechanisms are relevant at each scale and what functional forms describe those mechanisms. By its nature, multiscale modeling filters out irrelevant information at the lower length scales, but a mechanism that is largely irrelevant to the RTI experiments may be important to another application.

Going forward, a significant goal will be to understand the range of applicability of the model and what additions or modifications are needed to make it more robust. Some information about this is available already. At low rates, the dislocation mobility curves cannot be calculated by MD in the approach described here due to computational resource limitations [13]. Low rates require long times, and the cost of the simulation is proportional to the length of the simulation. A recent approach has been to create a model that effectively extends the LMS model to low rates by generalizing the form of the model somewhat and fitting to experimental data [23]. The model is not constructed from first principles, but it does match experimental data over a wide range of rates.

The form of the Ta LMS model is based on dislocation-mediated plastic flow. Deformation twinning has been observed in recovered Ta sample following dynamic loading [31-33], but the current MS models do not include twinning. Also, the model includes some dependence on the texture, the orientation distribution in a polycrystalline sample. It does not include mechanisms for the interaction of dislocations with the grain microstructure. The homogenization techniques for complex microstructures could be extended to include grain boundary motion, plastic rotation, and so on. At low rates bcc metals are known to exhibit non-associated flow [34] and other phenomena that would require an extension beyond a $J_2$ flow formulation. Fracture and failure are not part of the model, and a few of the vanadium RTI experiments exhibited very large ripple growth and failure (observed in the radiograph) that the LMS model did not capture. While there has been exploration of a multi-phase MS model [35], specifically for the subtle Martensitic transformation associated with the high-pressure rhombohedral phase of vanadium [36], this model of post-transformation plasticity has not been extended to other kinds of phase transformations. So there are several physical mechanisms known to be important under less extreme conditions that are not included in the model. Some dynamic data in Ta appear not to agree well with the MS model [37]. This may point to missing physics, perhaps already identified, perhaps something new; it remains to be seen.

6. Discussion

We have reviewed the application of multiscale modelling to laser-driven ramp compression experiments. We have described the formulation of a hierarchical multiscale model with particular emphasis on the LMS model. We have examined its behaviour in the context of saturated plastic flow,
largely the case in RTI experiments, and in the context of time dependent plastic relaxation, largely the case in in-situ x-ray diffraction shock experiments.

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