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August 3, 2011

APS Shock Compression of Condensed Matter Conference
Chicago, IL, United States
June 26, 2011 through July 1, 2011

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THEORY AND SIMULATION OF 1D TO 3D PLASTIC RELAXATION IN TANTALUM

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Abstract. In plane shockwaves the uniaxial strain rate can greatly exceed the rate at which dislocation flow can relax the concomitant shear stress. The result is an overdriven plastic state in which the compression is 1D uniaxial initially and only after a period of time does the lattice relax to a more 3D compressed state due to plastic flow. Here we use an analytic calculation based on a generalization of the Gilman model of flow involving dislocation evolution to predict the phases of plastic relaxation and to derive an analytic estimate of the relaxation time, including a decomposition into incubation and flow times, suitable for comparison with *in-situ* x-ray diffraction. We use molecular dynamics (MD) to study the threshold for homogeneous nucleation both in shock compression of single crystal Ta $\langle 100 \rangle$. We find that shock heating on the Hugoniot substantially lowers the threshold pressure for homogeneous nucleation.

Keywords: Plasticity, Molecular Dynamics, Shock Wave, Plastic Relaxation

PACS: 62.20.F-, 62.50.-p, 62.50.Ef

INTRODUCTION

The advent of *in-situ* x-ray diffraction in material dynamics experiments has provided new insight into the processes of plasticity, as well as phase transformation and other processes that affect the crystalline lattice. In-situ x-ray diffraction was first used to study plasticity in gas gun experiments [1], and was subsequently extended to laser-driven experiments [2–4]. Diffraction from single crystals has shown that the material that is initially compressed uniaxially relaxes to a more equiaxed state over a time period that depends on the material. Loveridge-Smith, et al. [2] have shown that copper relaxes to an approximately 3D state in less than a nanosecond, whereas silicon has not relaxed after a nanosecond.

Like silicon, body-centered cubic (bcc) materials have high Peierls barriers, and there is interest in determining the plastic relaxation time for these materials. The interest in bcc plastic relaxation times has other motivations as well. Recently, a technique for

measuring the strength of metals at high pressure has been developed on fusion-class lasers. The inferred strength of the bcc metal vanadium [5] has been found to be in good agreement with predictions of a multiscale strength model for vanadium developed from first principles [6, 7]. Experiments and analysis that determine the mechanisms of dislocation flow independently help reduce the non-uniqueness and gain more confidence in the model.

Here we show explicitly how plastic relaxation times can be used to constrain parameters of models of plastic flow that explicitly evolve dislocation density, ρ , including the Gilman model [8] and the multiscale strength (MS) model [7]. The analysis assumes conventional plastic flow, in which dislocations flow in response to shear stress, undergoing multiplication during flow. When the shear stress is sufficiently high, ρ can jump rapidly due to homogeneous nucleation. We use molecular dynamics (MD) to investigate the threshold for homogeneous nucleation in a shocked tantalum crystal.

METHODS

The rate of plastic relaxation following a strong shock can be calculated theoretically within various models. We focus on two. The first is a continuum analysis based on a generalization of the Gilman model for the dislocation kinetics. The second is an atomistic analysis using MD to investigate plastic relaxation due to homogeneous nucleation.

The dislocation-based continuum model we use includes the motion and multiplication of dislocations under applied shear stress. The constitutive model consists of four equations:

$$\dot{\rho} = R(1 - \rho/\rho_{sat}) \dot{\epsilon}^P \quad (1)$$

$$\dot{\epsilon}^P = \eta \rho b v / M \quad (2)$$

$$v = b \sigma / (MB) \quad (3)$$

$$\rho_{sat} = \rho_{sat0} \left(\frac{\dot{\epsilon}^P}{\dot{\epsilon}_0} \right)^n \quad (4)$$

where ρ is the dislocation density, ρ_{sat} is the asymptotic saturation density, R is a dislocation multiplication rate, v is the dislocation velocity, b is the Burgers vector, σ is the shear stress, and $\dot{\epsilon}^P$ is the plastic strain rate. Dots indicate time derivatives at fixed Lagrangian position. Equation (1) is a reformulation of the Gilman model for dislocation evolution [8]. Equation (2) is the Orowan Law [9], which kinematically links the plastic strain rate to the dislocation flow. Equation (3) is the dislocation mobility law. Since the shear stress is high, at least initially, we assume that the dislocations are in the phonon drag regime. B is the phonon drag coefficient [9]. Equation (4) approximates the saturation dislocation density as a power law in the plastic strain rate with parameters ρ_{sat0} and n , following Barton, et al. [7]. We are going to interpret the dislocation velocity as the mean velocity of dislocations, and have thus included a Taylor factor M and an Orowan factor η to average over the glide systems in the grains of a polycrystalline metal. For simplicity, we assume that the strength of the crystal is dominated by the lattice resistance, and thus neglect the work hardening contribution. Its effect will be considered elsewhere. The state variable ρ must be initialized with the dislocation density in the metal prior to deformation, ρ_0 . With suitable parameters, this model is an approximation to the MS model [7].

We also use molecular dynamics simulation of the compression of the bcc lattice to understand plas-

tic relaxation at stresses sufficiently high that defects nucleate homogeneously from the lattice. MD integrates Newton's equation $F = ma$ for a large set of atoms. We calculate the forces from a quantum-based many-body potential designed for high pressure: the Model Generalized Pseudopotential Theory (MGPT) potential [10]. The MGPT potential for Ta contains many-body bond-bending and bond-stretching forces, and has been shown to agree well with first principles calculations of elastic constants and defect energies at pressures up to and beyond the shock melting pressure of 3 Mbar. We use the dd-cMD code [11, 12] and rapidly compress the finite-temperature lattice uniaxially along the Rayleigh line to the Hugoniot state. The simulation is held at that volume and internal energy, and the ensuing stresses are output. We also output the various structural information for each atom in order to verify whether defects have nucleated or not [13].

RESULTS

Relaxation of stresses below the threshold for homogeneous nucleation involves incubation and flow of dislocations. In this case we apply the dislocation-based continuum model described above to a material point near the drive surface. Similar considerations apply to other locations, but not considered here. Initially the dislocation density may be too low to permit an appreciable dislocation flow since the plastic flow described by Orowan's Eq. (2) contains the mobile dislocation density as a factor. The result is an *incubation period* with exponentially increasing dislocation density due to dislocation multiplication but little relaxation of the shear stress. The Gilman model for dislocation evolution may be rewritten assuming constant σ as

$$\dot{\rho} = \tau_{inc}^{-1} (1 - \rho/\rho_{sat}) \rho \quad (5)$$

$$\tau_{inc} = \frac{M}{\eta R v_0 b} = \frac{M^2 B}{\eta R b^2} \sigma_0^{-1} \quad (6)$$

where v_0 is the dislocation velocity at the initial shear stress, σ_0 . If $\rho \ll \rho_{sat}$, the solution of this equation is exponential growth of the dislocation density with time constant τ_{inc} : $\rho \approx \rho_0 e^{t/\tau_{inc}}$. Here we have assumed that the stress has not relaxed appreciably so that $v \approx v_0$. The solutions will be discussed in more generality elsewhere. At long times the dislocation density asymptotes to ρ_{sat} .

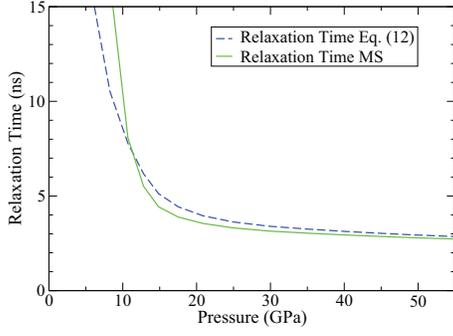


FIGURE 1. (Color online) Plastic relaxation time as estimated from Eq. (12) and calculated using the multiscale strength model [7]. The two agree to about 10% above the pressure of 10 GPa.

The asymptotic value of the saturation density depends on the strain rate through Eq. (4). Substituting this power law into $\rho = \rho_{sat}$ along with the mobility law (3) results in an upper bound (supremum) estimate for the saturation density:

$$\rho_{sup} = \left[\rho_{s0} \left(\frac{\eta b^2 \sigma_0}{M^2 B \xi_0} \right)^n \right]^{1/(1-n)} \quad (7)$$

$$\rho_{peak} \approx \rho_{sup}/4, \quad (8)$$

where ρ_{peak} is an estimate of the peak ρ . As the dislocation density builds up toward this value, the plastic strain rate rises from $\rho_0 b^2 \sigma_0 / (M^2 B)$ toward

$$\dot{\epsilon}_{max}^p = \rho_{peak} \eta b^2 \sigma_0 / (M^2 B), \quad (9)$$

and then decreasing as the shear stress relaxes.

After the incubation period the relaxation enters the *flow period*. Here the shear stress relaxation rate is proportional to the plastic strain rate: $\dot{\sigma} = -2G\dot{\epsilon}^p$, where G is the shear modulus. Using this relationship, Orowan's equation may be rewritten as

$$\dot{\sigma} = -\tau_{flow}^{-1} \sigma \quad (10)$$

$$\tau_{flow} \approx \frac{M^2 B}{2G(P) \eta \rho_{peak} b^2} \quad (11)$$

where we have approximated ρ as the constant ρ_{peak} . This expression then gives the time constant τ_{flow} for the exponential decrease in stress with plastic flow.

Putting this all together, we arrive at an estimate for the total plastic relaxation time:

$$\tau_{tot} \approx \tau_{inc} \log(12\rho_{peak}/\rho_0) + \tau_{flow} \quad (12)$$

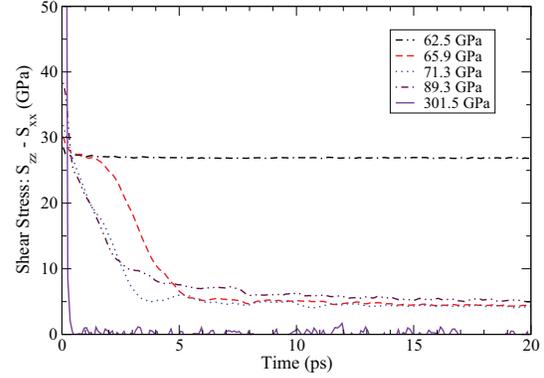


FIGURE 2. (Color online) Shear stress as a function of time following the $\langle 100 \rangle$ shock in Ta as calculated with MD. The response is elastic at $P = 62.5$ GPa. The next 3 curves show rapid relaxation to a non-zero strength due to homogeneous nucleation of defects. An example of shock melting at 301.5 GPa is shown for comparison.

This relaxation time depends logarithmically on the initial dislocation density. It increases by τ_{inc} (6) for each e-fold that the dislocation has to increase to saturate and then an additional flow time constant τ_{flow} (11) for the stress to relax.

The constitutive model used here is an approximation to the MS model [7] at high stresses. We can use the MS model parameters to get quantitative estimates of the relaxation time (the phonon drag coefficient is $B = \xi_0 b \tau_p / c_0$). The results are shown in Fig. 1. Relaxation times for $P > 10$ GPa are less than 8 ns (for $10 < P < 60$ GPa, τ_{inc} is 0.26 to 0.75 ns and the incubation period is 1.6 to 3.5 ns). The analytic formulas derived here agree well with relaxation times we have calculated using the MS model.

Next we turn to homogeneous nucleation. At high shear stresses new mechanisms can become active as the lattice approaches a mechanical instability. Dislocation shear loops and other defects can nucleate directly from the bulk lattice, as seen in MD [14], but with little experimental evidence thus far. Analysis of this process to date is based on finding a negative stress-strain coefficient [15] or an imaginary phonon [16] as calculated at zero temperature. Here we simulate $\langle 100 \rangle$ shock compression of a perfect single crystal of Ta, including the considerable increase in temperature due to dissipation in strong shocks.

In the plot in Fig. 2 we show the evolution of shear stress in the time after shock compression

for five different pressures (negative mean stresses): $P = 62.5, 65.9, 71.3, 89.3$ and 301.5 GPa. The upper curve is an elastic response for the ~ 50 ps duration of the simulation with no appreciable relaxation. The 65.9 GPa curve shows a metastable state for ~ 2 ps prior to homogeneous nucleation and relaxation. The 71.3 and 76.9 GPa curves show rapid relaxation with no detectable metastable state, relaxing to a final stress of ~ 5 GPa (a lower bound for the strength). For the fifth curve P is above shock melting, leading to rapid relaxation and no residual shear stress. For $P \geq 65.9$ GPa, relaxation happened in less than 5 ps and for $P \geq 71.3$ GPa less than 2 ps. The threshold for homogeneous nucleation in the shock, ~ 65 GPa, is considerably lower than in isothermal compression at room temperature, ~ 85 GPa. The state following homogeneous nucleation on the Hugoniot is very disordered and quite different than that in 300 K isothermal compression where shear loops and twins are observed with orientation analysis [13].

DISCUSSION

We have developed a theoretical model of plastic relaxation from 1D to 3D compression due to plastic flow. We have derived an analytic estimate of the relaxation time, including incubation and flow times. This model is closely related to the recent MS model [7], and using the parameters from that model we have predicted ns relaxation times. Conversely, the formulas have shown explicitly what parameters of the model can be tested by diffraction experiments measuring relaxation times.

We have also used MD to go beyond conventional plasticity and study the threshold for homogeneous nucleation of dislocations and other defects in shock compression of single crystal Ta. We have found that shock heating on the Hugoniot substantially lowers the threshold for homogeneous nucleation and shortens the relaxation time from ns to ps. The plastic state might be related to recently proposed phenomena in high temperature Ta: Bingham plasticity [17] and ω phase [18]. The MD simulations predict a threshold for homogeneous nucleation easily within the range of laser-driven shock diffraction experiments on Ta and point to the need to extend conventional plasticity models to include homogeneous nucleation.

ACKNOWLEDGMENTS

We acknowledge discussions with N. R. Barton, J. N. Glosli, R. Minich and R. Smith. We thank J. A. Moriarty for the MGPT Ta potential and J. N. Glosli, D. F. Richards, and F. H. Streitz for the ddcMD code. We appreciate support from the LDRD program at Lawrence Livermore National Laboratory (LLNL) under project 09-SI-010 and supercomputer time from the Computing Grand Challenge program at LLNL. This work was performed under the auspices of the U.S. Dept. of Energy (DOE) by LLNL under Contract DE-AC52-07NA27344.

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