Cross-Scale Molecular Dynamics Simulations of Single Crystal Plasticity

A. Stukowski, L. A. Zepeda-Ruiz, T. Oppelstrup, V. V. Bulatov

published in

NIC Symposium 2018
K. Binder, M. Müller, A. Trautmann (Editors)

Forschungszentrum Jülich GmbH,
John von Neumann Institute for Computing (NIC),
Schriften des Forschungszentrums Jülich, NIC Series, Vol. 49,
ISBN 978-3-95806-285-6, pp. 255.
http://hdl.handle.net/2128/17544

Cross-Scale Molecular Dynamics Simulations of Single Crystal Plasticity

Alexander Stukowski[1], Luis A. Zepeda-Ruiz[2], Tomas Oppelstrup[2], and Vasily V. Bulatov[2]

[1] Technische Universität Darmstadt, Darmstadt, Germany
E-mail: stukowski@mm.tu-darmstadt.de

[2] Lawrence Livermore National Laboratory, Livermore, California, USA
E-mail: {zepedaruiz1, oppelstrup2, bulatov1}@llnl.gov

Strength and plasticity properties of a metal are typically defined by dislocations – line defects in the crystal lattice whose motion results in atomic slippage along lattice planes. It is of considerable fundamental and practical interest to identify and quantify conditions at which dislocation-mediated plasticity reaches its limits and to understand what happens to the metal beyond any such limit. Resolving every "jiggle and wiggle" of atomic motion, molecular dynamics (MD) simulations are faithful to every possible mechanism of material response making them uniquely suited for probing conditions when dislocation motion begins to be superseded by some other mechanism of crystal plasticity. Here, we present the first fully dynamic atomistic simulations of bulk single crystal plasticity in tantalum metal predicting that, on reaching certain limiting conditions of straining, dislocations alone can no longer relieve mechanical loads and another mechanism, deformation twinning, takes over as the dominant mode of dynamic response. Below this limit, however, the metal attains a path-independent steady state of plastic flow in which both the flow stress and the dislocation density remain constant indefinitely.

1 Introduction

Plasticity – permanent change of material shape – results from dislocation lines sweeping through the lattice planes, each line propagating a tiny quantum of atomic displacement (the Burgers vector) and thus relieving elastic lattice strain[1]. The strength of a ductile metal is typically defined by the magnitude of mechanical stress needed to push dislocations through the lattice. When dislocation motion alone is insufficient to relieve lattice strain, metals respond to continued straining in a more spectacular manner, e.g. twinning or fracture. Our key objective here is to answer the following question: Under what conditions of straining does dislocations innate ability to relieve stress become overwhelmed? Our large-scale simulations addressing this question were partly performed on the JUQUEEN supercomputer at Forschungszentrum Jülich and have recently been published in the journal *Nature*[2]. In the following we provide an overview of the employed simulation approach and give a brief summary of our central findings.

Our question posed above appears to be squarely within the purview of Dislocation Dynamics (DD), a method developed specifically for simulation of dislocation-based plasticity[3]. However, application of DD for our stated purpose here is problematic due to the method's own limitations, key of which is that DD models do not include any other mechanisms of inelastic response alternative to dislocation motion, e.g. fracture or twinning. Because a DD model cannot decide when any such behaviours become active, it does not know when dislocation plasticity reaches its limits. That is why here we employ Molecular Dynamics (MD) simulations to probe the limits of metal plasticity. Our MD simulations

incorporate every "jiggle and wiggle"[4] of atomic dynamics and are thus faithful to every possible mechanism of dynamic response.

MD simulations of single crystal plasticity are computationally challenging and, to the best of our knowledge, have not been carried out before in part due to the severe length and time scale limits that the less expensive DD method was developed to overcome. Another serious challenge is that MD simulations generate enormous amounts of data: an MD simulation of the kind reported here generates petabytes of trajectory data in just one day on a large supercomputer. To be useable, this transient MD data must be compressed to size and recast into a form that a human can grasp and comprehend. Here we rely on recently developed methods of *in situ computational microscopy* to identify and precisely characterise extended defects in crystals[5,6] and to reduce the amount of data to deal with by several orders of magnitude.

2 Simulation Setup and Data Analysis Methods

In this work we focus on the dynamic response of tantalum single crystal to compressive straining along the [001] crystal axis. In our MD simulations, the metal is subjected to simple and fully controlled straining conditions in which pressure, temperature and (true) straining rate remain constant. Most simulations were performed using the open-source code LAMMPS[7] on a rectangular fragment of tantalum crystal with dimensions $L_x \times L_y \times L_z = 128\,a_0 \times 256\,a_0 \times 512\,a_0$ oriented along the principal [100], [010], [001] axes of the cubic lattice, respectively (here $a_0 = 0.33$ nm is the lattice constant of the body-centred cubic lattice of tantalum). The EAM interatomic potential for Ta developed by Li *et al.*[8] was used. The number of atoms in the simulation box was approximately 33 million and 3d periodic boundary conditions were applied to embed the box seamlessly into an infinite crystal. In most of the simulations the crystal was compressed at a constant "true" rate $\dot{\epsilon}_T = \frac{1}{L_z}\frac{\mathrm{d}L_z}{\mathrm{d}t}$ along its initially longest dimension L_z while the two lateral dimensions L_x and L_y were left to dynamically expand to maintain xx and yy components of the mechanical stress near zero. Most straining simulations were continued until the "longest" dimension L_z was compressed to 1/4 of its initial size by which time L_x and L_y almost doubled thus keeping volume and pressure nearly constant throughout the simulation. Temperature was also maintained near 300 K using a Langevin thermostat. Everywhere in the text by "stress" we mean the uniaxial compressive stress σ_{zz} that develops in the model crystal in response to straining along its [001] z-axis.

In addition to standard local atom filtering techniques, we employ the Dislocation Extraction Algorithm[9] (DXA) and a Grain Segmentation Algorithm (GSA) to reveal structural defects in simulated crystals and to reduce the amount of data to deal with by orders of magnitude. The DXA analysis algorithm, developed by one of the authors, accurately traces and indexes the dislocation lines in the atomistic crystal and builds a concise network representation of the defect microstructure. The newly developed GSA algorithm automatically partitions the atomistic crystal into regions of similar crystal orientation (grains) and generates a geometric representation of the interfaces delineating the grains. This conversion from the fully atomistic model to a highly reduced description of the defect microstructure is performed at regular time intervals during the MD simulation and enables us to follow the motion and reactions of dislocations, the onset of twinning and other important events in arbitrary degree of detail[10].

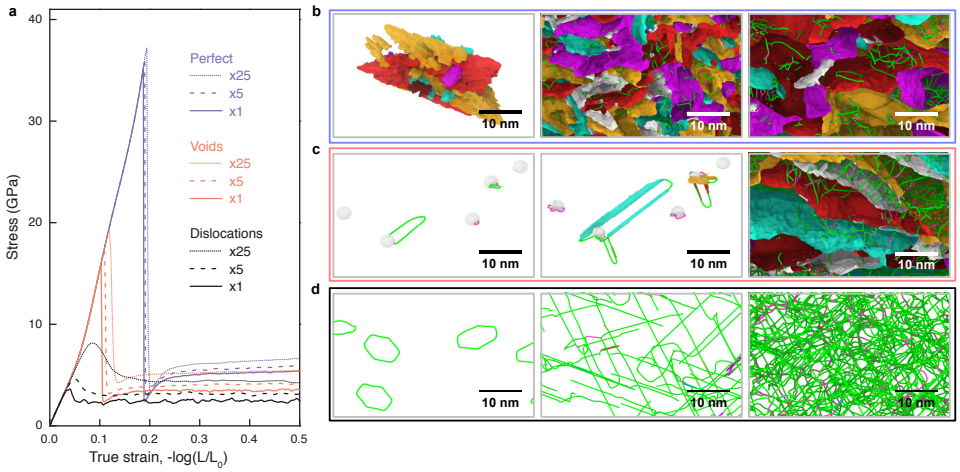

Figure 1. **Response to compression as a function of initial defect content.** (a), Stress as a function of strain. Blue, red and black curves were computed for the initially perfect crystal, crystal with voids and crystal with dislocations, respectively. Solid, dashed and dotted lines correspond to straining rates $\times 1$, $\times 5$ and $\times 25$, respectively. (b) From left to right: co-nucleation of embryonic twins, twin propagation and twin growth in the initially perfect crystal. (c) Dislocation nucleation at voids, nucleation of twins on stretched dislocations and twin growth in the crystal with voids. (d) Initial dislocation loops, loop extension and impingement and formation of a dense dislocation network. Dislocations appear as green and magenta lines. The twins appear as hollow volumes bounded by interfaces coloured gray, red, yellow, magenta or cyan to distinguish the parent crystal and four rotational twin variants.

3 The Important Role of Initial Defects

First, we compare the response of the crystal to compression starting from three different initial configurations: (A) a defect-free perfect crystal, (B) the same crystal with 24 randomly placed voids[11] and (C) 24 dislocation loops placed into the same locations as voids in B. Shown in Fig. 1 are the stress-strain curves computed from these three starting configurations under three different straining rates: $\dot{\epsilon}_T = 1.109 \cdot 10^7$ 1/s (our "base" rate denoted hereafter as $\times 1$), $\dot{\epsilon}_T = 5.545 \cdot 10^7$ 1/s ($\times 5$) and $\dot{\epsilon}_T = 2.773 \cdot 10^8$ 1/s ($\times 25$). Response of the three crystals to straining is markedly different. Crystals A and B give in (yield) to strain abruptly after reaching peak stresses of 36-37 GPa (crystal A) and 16-19 GPa (crystal B) followed by sharp drops in stress, whereas crystal C yields gradually along smooth stress-strain curves passing over peak stresses that are both much lower and distinctly rate-dependent.

Analysis of crystal configurations attained in these simulations reveals that crystals A and B yield by twinning – a sudden strain-induced reorientation of the crystal lattice within bounded volumes of the material[12]. In contrast, crystal C does not twin but yields exclusively through the motion and multiplication of the initial dislocations forming dense line networks during and soon after yielding. This comparison shows that, by depriving the metal of initial dislocations in crystals A and B, not only is the yield strength overpredicted by a large factor, but the metal yields through a mechanism qualitatively different from crystal C in which dislocations are introduced from the beginning. As is well

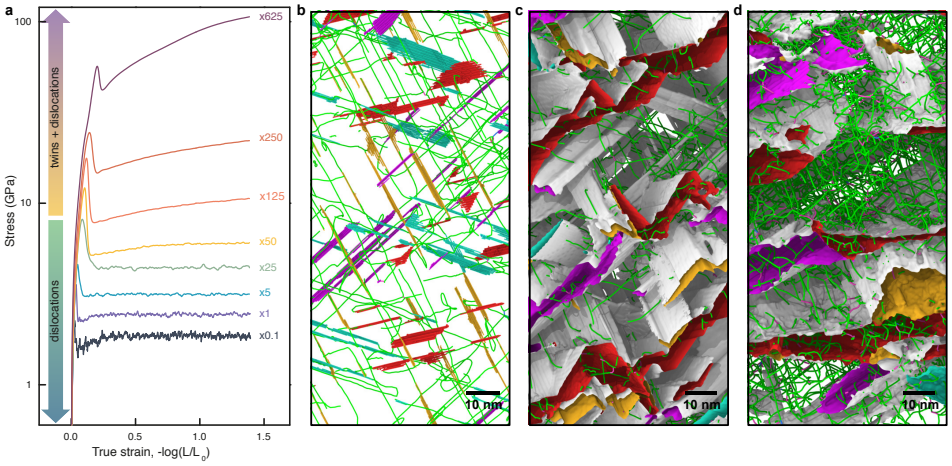

Figure 2. **Response to compression as a function of straining rate.** (a) Logarithm of stress as a function of true strain computed in MD simulations of specimen compression at different straining rates at temperature 300 K. (b) A snapshot taken immediately past yield showing embryonic twins in the simulation performed at rate $\times 50$. (c) A later snapshot from the same simulation showing twin propagation. (d) A still later snapshot in which the twins have grown to span the simulation volume. Colouring of dislocation lines and twin interfaces is the same as in Fig. 1. Compression rates are shown next to the corresponding stress-strain curves in matching colours.

known, multiplication of pre-existing dislocations entails much lower stress than dislocation nucleation[13]. As no ductile metal has ever been obtained dislocation-free, stress-strain behaviours observed in crystals A and B may be considered unrealistic. In the following, we thus confine ourselves to crystal models with pre-existing dislocations.

4 Determining the Limits of Dislocation Plasticity

We are interested here in determining the straining conditions at which dislocations ability to relieve stress becomes overwhelmed. First, we examine the dependence of stress response on the straining rate. As shown on Fig. 2, flow stress appears to eventually saturate at all straining rates equal or below $\times 25$, whereas at the higher rates flow stress continues to rise through the end of compression straining. Analysis of atomic snapshots reveals that at rates $> \times 25$ the crystal twins profusely (Fig. 2, right), whereas no twinning is observed at the lower rates.

In order to more accurately determine the conditions defining the transition from dislocation plasticity to twinning, we intend to continue by ramping the straining rate up gradually, either continuously or in steps, thus giving dislocations ample (ideally infinite) time to multiply and to avoid the stress overshoots observed at initial yielding. Using our grain segmentation algorithm we detected that, irrespective of the details of straining trajectory (constant rate, continuously increasing rate, or rate jump) twinning is first triggered whenever the stress developing in response to compressive straining reaches or exceeds 8 GPa. Using rate ramping as slow as we could afford, the highest rate the dislocations were found to sustain without triggering twinning is approximately $8 \cdot 10^8$ 1/s, at which

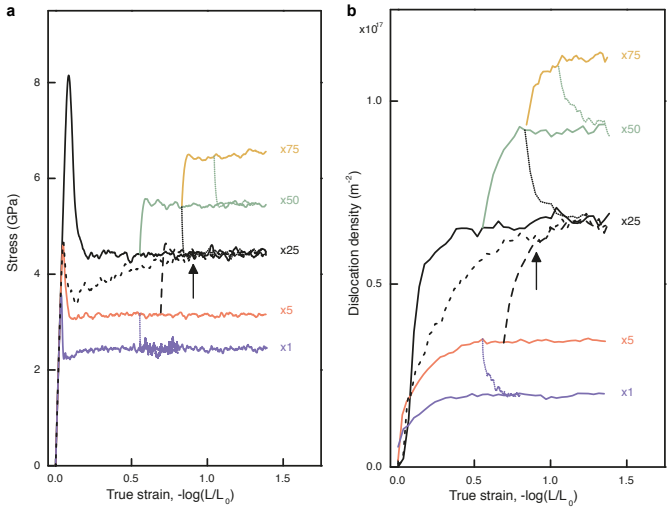

Figure 3. **Attainment of a path-independent flow state.** (a) Stress-strain response computed in MD simulations along four different types of straining trajectories at T=300 K: solid lines correspond to compression at constant rates, dotted lines show stress relaxations after jumps to lower rates, the long-dash line shows stress relaxation after a jump to a higher rate and the short-dash line corresponds to straining rate continuously increasing from zero to $\times 25$ and then held constant beginning from strain marked by the vertical arrow. (b) Dislocation density as a function of strain computed along the same straining trajectories. Line colours and types match the stress-strain trajectories depicted in (a).

point the stress approaches its trigger level of 8 GPa. Thus, the twinning transition is most succinctly defined by this twinning threshold stress.

Our simulations furthermore predict that the twinning threshold stress is nearly temperature-independent in tantalum, whereas the maximum sustainable rate sharply decreases with decreasing temperature. This is fully consistent with experimental observations of twinning under low straining rates at cryogenic temperatures and with the well-established fact that stress required to move screw dislocations at a given velocity in tantalum and other BCC metals increases sharply with decreasing temperature. Our simulations reveal a tight interplay between dislocation plasticity and twinning. On one hand, dislocation motion relieves stress and prevents the onset of twinning On the other hand, when dislocations are unable to keep up with rising stress, they instigate twinning and serve as preferential sites for heterogeneous nucleation of twins via the mechanism proposed by Sleeswyk[14,15].

5 Stationary State of Plastic Flow

We now turn to our simulated straining trajectories in which stress stays below the threshold and twinning is *not* triggered. It turns out that any such trajectory leads to an ultimate steady state of material flow that is then maintained indefinitely for as long as straining conditions (rate, temperature, straining axis) remain unchanged. Fig. 3 presents substantial evidence that, no matter how the ultimate straining conditions are attained –whether by

straining at a constant rate throughout, or in a sequence of rate jumps or under continuously varying rate– the state attained by the material in the end is manifestly path-independent. For example, the four different types of straining trajectories leading to the same limiting rate $\times 25$ clearly converge to the same stress (black lines in Fig. 3a) and dislocation density (black lines in Fig. 3b) that only depends on the final straining rate. In this state, while fully retaining its crystal lattice and remaining strong and elastically stiff, the bcc metal responds to straining just like a perfectly viscous fluid.

Additional evidence for the attainment of a stationary flow state is the behaviour of dislocation line density ρ during straining. Defined as the total length of dislocation lines in a unit volume, ρ is seen to attain a stationary value of its own, even if it takes markedly longer time to settle compared to the flow stress (Fig. 3b). Furthermore, just like the flow stress, the steady value of the dislocation density is path-independent and is the same, within statistical errors, no matter how the rate was attained. In the steady state, dislocation multiplication is exactly balanced by dislocation annihilation. We regard the observed flow state as a distinctive stationary state maintained at a constant rate of energy dissipation equal to the product of the flow stress and the straining rate. In this stationary open system, the supplied mechanical energy is converted through dislocation multiplication, annihilation and motion into heat and eventually collected and removed by the thermostat. In a further confirmation of its stationary nature, we have demonstrated that the flow stress remains unchanged over arbitrarily large compressive strains in a "metal kneading" simulation (see Ref. 2).

6 Discussion

Fully dynamic atomistic simulations of the kind reported here have long been thought impossible, necessitating the development over the last 30 years of the special method of discrete dislocation dynamics[16,17,3]. However, although greatly expanded due to a steady growth in massively parallel computing, the still present length- and time-scale bounds of MD simulations limit us to probing material dynamics under high straining rates. Although high, straining rates and dislocation densities encountered in our simulations are entirely common to high-rate shock and shockless compression experiments[18,19]. Such dynamic experiments, however, involve complex and largely unknown straining trajectories that are far from stationary[20,21] and which can only be indirectly inferred from continuum plasticity simulations.

Critical comparison of our simulation data to the literature suggests that, understandable quantitative differences notwithstanding, the basic physics of metal plasticity remains the same across the entire range of straining rates – from quasi-static to laser-driven shocks – for as long as dislocation motion dominates the straining response. Perhaps the most pertinent is an earlier experimental observation of saturated steady flow in single crystal molybdenum (a close cousin of tantalum) compressed along its 001 axis at rate 10^{-3} 1/s. In these experiments much care was taken to maintain straining conditions stationary over very large strains[22].

Our MD simulations suggest that dislocations behave the same as expected at lower rates: (1) they move in a stop-and-go fashion remaining relatively motionless against obstacles (junctions and self-pinning cusps) for periods of time intermittent with bursts of faster motion; (2) the distribution of dislocation line orientations is non-uniform and dom-

inated by line segments nearly parallel to their Burgers vectors (screws), (3) average dislocation velocities range between 1 and 60 m/s, placing the dominant screw dislocations squarely into the temperature- and stress-activated motion regime. Aside from understandable quantitative differences, we believe the mechanisms of dislocation-mediated plasticity in tantalum remain the same across the entire range ($\sim$ 13 decades) of straining rates accessible experimentally. If so, then simulation of the kind reported here present a distinct and exciting opportunity to examine and understand fundamental mechanisms underlying metal plasticity in arbitrary degree of detail. The presented simulations are *cross-scale* rather than multi-scale, i.e. simultaneously large enough to represent macroscopic crystal plasticity and yet fully resolved, tracing every detail of atomic motion.

Acknowledgements

The authors gratefully acknowledge the Gauss Centre for Supercomputing e.V. (`www.gauss-centre.eu`) for funding this project by providing computing time through the John von Neumann Institute for Computing (NIC) on the GCS Supercomputer JUQUEEN at Jülich Supercomputing Centre (JSC).

References

1. V. V. Bulatov and W. Cai, *Computer Simulations of Dislocations*, Oxford University Press, 2006.
2. L. A. Zepeda-Ruiz, A. Stukowski, T. Oppelstrup, and V. V. Bulatov, *Probing the limits of metal plasticity with molecular dynamics simulations*, Nature **550**, 492–495, 2017.
3. A. Arsenlis, W. Cai, M. Tang, M. Rhee, T. Oppelstrup, G. Hommes, T. G. Pierce, and V. V. Bulatov, *Enabling strain hardening simulations with dislocation dynamics*, Modelling and Simulation in Materials Science and Engineering **15(6)**, 553, 2007.
4. R. Feynman, Lectures on physics **1**, 3–6, 1963.
5. A. Stukowski, *Computational analysis methods in atomistic modeling of crystals*, Jom **66(3)**, 399–407, 2014.
6. A. Stukowski, V. V. Bulatov, and A. Arsenlis, *Automated identification and indexing of dislocations in crystal interfaces*, Modelling and Simulation in Materials Science and Engineering **20(8)**, 085007, 2012.
7. S. Plimpton, *Fast parallel algorithms for short-range molecular dynamics*, Journal of computational physics **117(1)**, 1–19, 1995.
8. Y. Li, D. J. Siegel, J. B. Adams, and X.-Y. Liu, *Embedded-atom-method tantalum potential developed by the force-matching method*, Physical Review B **67(12)**, 125101, 2003.
9. A. Stukowski and K. Albe, *Extracting dislocations and non-dislocation crystal defects from atomistic simulation data*, Modelling and Simulation in Materials Science and Engineering **18(8)**, 085001, 2010.
10. A. Stukowski, *Visualization and analysis of atomistic simulation data with ovito–the open visualization tool*, Modelling and Simulation in Materials Science and Engineering **18(1)**, 015012, 2009.

11. D. Tramontina, P. Erhart, T. Germann, J. Hawreliak, A. Higginbotham, N. Park, R. Ravelo, A. Stukowski, M. Suggit, Y. Tang, et al., *Molecular dynamics simulations of shock-induced plasticity in tantalum*, High Energy Density Physics **10**, 9–15, 2014.
12. K. G. Hoge and A. K. Mukherjee, *The temperature and strain rate dependence of the flow stress of tantalum*, Journal of Materials Science **12(8)**, 1666–1672, 1977.
13. F. C. Frank and W. T. Read Jr., *Multiplication processes for slow moving dislocations*, Physical Review **79(4)**, 722, 1950.
14. A. W. Sleeswyk, $1/2 <$ *111*$>$ *screw dislocations and the nucleation of* $\{112\}<$ *111*$>$ *twins in the bcc lattice*, Philosophical Magazine **8(93)**, 1467–1486, 1963.
15. J. Marian, W. Cai, and V. V. Bulatov, *Dynamic transitions from smooth to rough to twinning in dislocation motion*, Nature materials **3(3)**, 158, 2004.
16. N. Ghoniem, S.-H. Tong, and L. Z. Sun, *Parametric dislocation dynamics: a thermodynamics-based approach to investigations of mesoscopic plastic deformation*, Physical Review B **61(2)**, 913, 2000.
17. D. Weygand, L. H. Friedman, E. Van der Giessen, and A. Needleman, *Aspects of boundary-value problem solutions with three-dimensional dislocation dynamics*, Modelling and Simulation in Materials Science and Engineering **10(4)**, 437, 2002.
18. S. Nemat-Nasser, J. B. Isaacs, and M. Liu, *Microstructure of high-strain, high-strain-rate deformed tantalum*, Acta Materialia **46(4)**, 1307–1325, 1998.
19. L. L. Hsiung, *Shock-induced phase transformation in tantalum*, Journal of Physics: Condensed Matter **22(38)**, 385702, 2010.
20. B. W. Reed, J. Reed Patterson, D. C. Swift, J. S. Stolken, R. W. Minich, and M. Kumar, *A unified approach for extracting strength information from nonsimple compression waves. part ii. experiment and comparison with simulation*, Journal of Applied Physics **110(11)**, 113506, 2011.
21. C.-H. Lu, E. N. Hahn, B. A. Remington, B. R. Maddox, E. M. Bringa, and M. A. Meyers, *Phase transformation in tantalum under extreme laser deformation*, Scientific reports, **5**, 2015.
22. F. M. A. Carpay, G. Y. Chin, S. Mahajan, and J. J. Rubin, *Constrained deformation of molybdenum single crystals*, Acta Metallurgica **23(12)**, 1473–1478, 1975.