Identifying deformation mechanisms in molecular dynamics simulations of laser shocked matter

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ABSTRACT

In this paper we demonstrate a new post-processing technique that allows straightforward identification of deformation mechanisms in molecular dynamics simulations. We utilise reciprocal space methods by calculating a per-atom structure factor (PASF) to visualise changes in volume, orientation and structure, thus allowing unambiguous discrimination between key deformation/relaxation mechanisms such as uniaxial strain, twinning and structural phase transformations. The full 3-D PASF is reduced to a 2-D representation by taking only those points which lie on the surface of an ellipsoid passing through the nearest reciprocal lattice points. Projecting this 2-D representation onto the set of spherical harmonics allows for a numerical characterisation of the system state that easily captures various plastic deformation mechanisms that have been historically difficult to identify. The technique is used to successfully classify high temperature twinning rotations in shock compressed tantalum and to identify the α to ω phase transition in group-IV hcp metals.

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1. Introduction

Atomistic simulation through molecular dynamics (MD) has become an invaluable tool for modelling the response of material undergoing high rate deformation, such as that experienced during material irradiation with a high intensity optical laser [1–4]. Such conditions are of fundamental scientific interest and remain an active area of research due to their application in planetary science [5], materials synthesis [6] and within thermonuclear fusion research and the long-term goal of facilities such as the National Ignition Facility, USA [7,8]. The short timescales and high strain-rates present in these experiments make them particularly amenable to MD simulation, which has become an indispensable tool for investigating the richness of lattice level behaviours activated at these high temperature and pressure conditions, such as defect generation, twinning, dislocation motion and structural phase transitions [9–12].

Experiments in this field, routinely performed on state-of-the-art 4th generation light sources where the X-ray source is used as a probe, drive micron sized volumes of material to mega-bar conditions over picosecond timescales [13–15] and we are fast approaching a point where MD and free electron lasers (FELs) operate at commensurate scales. A recent experiment recorded in situ X-ray diffraction from Cu shocked to pressures of 100 GPa and demonstrated a remarkable agreement

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between MD and experiment, providing greater mechanistic understanding of the deformation mechanics present [13]. However, the analysis of large-scale MD simulations is non-trivial and real space methods currently have difficulty identifying some specific transitions of fundamental interest (α to ω phase transition, high temperature twinning, etc.).

Classical MD simulations typically keep track of just particle positions and velocities which then evolve according to a Hamiltonian and a prescribed interatomic potential. Hence, characteristic features of plastic deformation such as slip planes, Burgers vectors, lattice rotations, phase transformations or even unit cells must be computed *a posteriori*. Numerous real-space order parameters exist such as coordination number, common neighbour analysis [16] (CNA), centrosymmetry parameter [17] or bond angle analysis [18] which excel at determining structural changes such as solid–solid phase transitions and gives excellent spatial resolution, allowing parameters to be calculated on a per-atom basis. However, they work only for common structures and the inherently real space nature of these techniques mean their accuracy remains extremely sensitive to local disorder from high temperatures or large defect generation, both of which are present in laser shocked matter. A summary of existing analysis techniques can be found in the review article by A. Stukowski [19].

Here we introduce a framework based on the per-atom structure factor (PASF) concept developed by Higginbotham et al. [20]. This reciprocal space technique is easily able to identify lattice level changes such as uniaxial compression, crystallographic orientation or phase transformations in highly disordered or high temperature samples. This extension, achieved through projecting an ellipsoidal slice of reciprocal space onto the set of spherical harmonics, produces a numerical characterization of the system that can be used to colour or categorize the atoms to aid visualization, removing the need to know structures, strains or orientations *a-priori*.

The use of spherical harmonics in atomic structure identification is not new – see for example [21–23], or the section on Fourier descriptors in the work by Keys et al. [24], which matches Harmonic decompositions of real space density. The plethora of techniques available highlight the difficulties associated with accurate structure identification. Used in conjunction with the per-atom structure factor Harmonic decomposition provides benefits in the analysis of laser-shocked solids that has not been presented before. In contrast to previous work our method remains intentionally sensitive to lattice compression, achieved through taking a slice rather than a projection of reciprocal space, and to lattice rotations, allowing identification of twinning and twin fraction. This work provides a middle ground which alleviates issues with existing approaches; this is demonstrated in the much more complex environment of a shock, where effects due to disorder must be mitigated.

2. Per atom structure factor

The basis of this method is calculating the atomic structure factor, a technique analogous to taking the discrete Fourier transform of the atomic positions. The structure factor easily identifies periodicity in the crystal micro-structure and, in a solid with a single atomic species, is defined by,

$$S(\mathbf{k}) = \frac{1}{M} \left| \sum_{n=1}^{M} e^{i\mathbf{k}\cdot\mathbf{r_n}} \right|^2 \tag{1}$$

where $\mathbf{k}$ is a reciprocal space lattice vector and $\mathbf{r_n}$ are the atomic coordinates. The summation in equation (1) typically runs from $n = 1$ to M, where M is the total number of atoms in the simulation, hence the structure factor is by definition a non-local metric. For decades, calculation of the crystal structure factor has allowed comparison between experimentally recorded X-ray diffraction patterns and theoretically derived crystal micro-structures as, within the kinematic approximation, the intensity of an X-ray diffraction pattern is related to the modulus squared of the structure factor calculated over the X-ray interaction region [25].

In order to reduce the crystal structure factor to a per-atom quantity we reduce the region over which the summation in equation (1) runs to include only those atoms that lie within a predetermined radius around each target atom. This quantity we call the per-atom structure factor. A judicial choice of this radius is required, as too small a number will result in too few atoms being within the calculation sphere increasing the effect of noise and hiding any periodicity or symmetries present in the crystal. On the other hand, a larger radius will afford an increased reciprocal space resolution at the cost of real space resolution and thus could obscure features smaller than the chosen radius. In agreement with previous work we find a radius of 4 Å, typically producing a sphere containing between 10 and 20 atoms, to provide a good balance between these competing effects [20].

An outline of the general method is given in Fig. 1. The top row shows real space representation of a body-centred cubic (bcc) crystal structure along with three additional structures that could occur during prototypical high rate crystal deformation; these are a 20% uniaxial compression along z, a lattice rotation consistent with the {112}⟨111⟩ twin systems expected in bcc material and lastly a phase transformation into hexagonal close packed (hcp), e.g. the α to ϵ phase transition in iron. The middle row of Fig. 1 shows high resolution iso-surfaces of the PASF for each real space lattice, calculated with a reciprocal space resolution of 10^{-2} Å^{-1} and containing 10^6 k-points. From each 3-D PASF the deformation mechanism is readily apparent, as indicated by the arrows. However, there is a large computational cost with calculating such a number of reciprocal space values.

To reduce this computation cost without assuming *a-priori* knowledge of the system, here we instead calculate the value of the 3-D PASF on a ellipsoid slice through reciprocal space which passes through the nearest neighbour reciprocal lattice