Symmetric tilt boundaries in body-centered cubic tantalum

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A B S T R A C T
Grain boundaries can play a significant role in the mechanical response of materials. Atomistic simulations are used to investigate 79 coincidence site lattice grain boundary structures and energies in tantalum, a model body-centered cubic transition metal. Quasi-symmetric Σ3, Σ5, Σ7, Σ13, and Σ27 boundaries are observed, of which Σ3 and Σ7 also exist as traditional mirror-symmetry conserving boundary structures. These results are supported by previous observations of similar phenomena in other bcc transition metal Σ5 boundaries. Metastable low energy Σ3 boundary structures in tantalum could influence the formation and stability of deformation twins and abnormal growth grain favoring Σ3 boundaries.

Grain boundaries play a critical role in the determination of mechanical, chemical, and thermal properties of polycrystalline materials. Specifically, the internal structure and energy can strongly determine grain boundary (GB) stability and influence the deformation response by affecting dislocation nucleation, dislocation motion, grain boundary sliding, diffusion, and radiation damage processes [1–7]. The distribution and character of grain boundaries are critical in controlling the strength of metals [8], especially tantalum [9–11]. The nature of grain boundaries is inherently complex because their energy is dependent on their character, which depends on five degrees of freedom. To these, one could add translation, which also changes the nature of the boundaries and has been shown to be particularly important for bcc metals [12]. A number of analytical approaches have been developed to treat GB structures: coincidence site lattice (CSL), displacement shift complete, and other topological treatments e.g., [13]. To investigate the dependence of deformation mechanisms on GB structural details, we first must understand the structural and energetic landscape of GBs as in recent surveys of face-centered cubic (fcc) [14] and some body-centered cubic (bcc) [15] materials.

Supporting these surveys, there have been numerous reports using both simulations and experiments to understand the GB structure and energies of fcc materials [14,16,17]. These studies have included both symmetric and asymmetric tilt boundaries on (111), (100), (110) and (113) planes [14,18]. In contrast, only a handful of studies exist on bcc symmetric and asymmetric tilt boundaries on (111), (100), (110) and energies of fcc materials [14,16,17]. These studies have included both simulations and experiments to understand the GB structure and character, which depends on character, which is inherently complex because their energy is dependent on their character, which depends on five degrees of freedom. To these, one could add translation, which also changes the nature of the boundaries and has been shown to be particularly important for bcc metals [12].

In the present work, we investigate GB structures in bcc tantalum and their associated energies using molecular dynamics (MD). Seventy-nine grain boundaries of varying tilt axis and misorientation (as described by the CSL model) are evaluated. The accompanying ‘Data in Brief’ details complete tilt axis (abc), grain boundary normal (hkl), CSL Σ index, misorientation, and conversion of these values into appropriate simulation basis for four tilt axes: (001), (011), (111), and (112). We use the MD code LAMMPS [24] to generate and relax bicrystal structures based on the scheme of Tschopp et al. [18,25,26]. Briefly, within a fully periodic system, one crystal is sequentially shifted with respect to another along its γ-surface; atoms that exceed overlap criteria are selectively removed; and the boundary is relaxed in the GB normal direction. The grain boundary energy (GBE) is calculated by evaluating the excess energy of the system per grain boundary area, taking into account that each simulation contains two parallel grain boundaries. The embedded atom model (EAM) potential developed by Ravelo et al. [27] is principally employed, but the importance of the Σ3 coherent twin boundary to deformation behavior warranted a “quantum accurate” investigation using a spectral neighbor analysis potential (SNAP) developed by Thompson et al. [28]. Depending on the boundary orientations, between hundreds and thousands of possible atomic arrangements containing up to fifty thousand atoms were sampled to produce each minimum energy configuration, thus neither density functional theory (DFT) nor extended use of SNAP is practical for the present study.
Fig. 1 shows the calculated GBEs as a function of misorientation angle alongside previous results for other bcc elements. GB structures presented in subsequent figures are indicated by vertical dashed lines. Based on calculations of surface energies for bcc transition metals, the energy of tantalum interfaces are bracketed between iron and molybdenum [29]. We note that it is to be expected that GBEs calculated in this work will be slightly lower than those calculated by tight binding or ab-initio methods [17,30].

Fig. 1. Grain boundary energy as a function of misorientation for four tilt axis. GBEs for Ta are shown as black asterisks (GBE values can be found in the supplemental material). Also provided is relevant data for bcc Fe (blue) and bcc Mo (red) from empirical potentials (Wolf [12,19], Morita and Nakashima [31], and Tschopp et al. [6]) illustrating similar trends.

Fig. 2. Configurations of $\Sigma 3$ boundaries colored by eV/atom shown for two projections. In each projection the GB normal is horizontal. (Left) EAM symmetric twin boundary. (Middle) EAM quasi-symmetric boundary with broken mirror symmetry in both (110) and (111) projections. (Right) SNAP quasi-symmetric boundary.
Two GB structures for the Σ3 twin boundary are obtained using EAM: a structure with a well-defined mirror plane, Σ3 in Fig. 2a, and an alternate structure with a broken mirror plane, alternate-Σ3 (alt-Σ3) in Fig. 2b. The alt-Σ3 boundary is quasi-symmetric, i.e. grain normals are equal and opposite in sign, but crystallographic mirror symmetry across the GB plane is broken [32]. The alt-Σ3 structure can be obtained by shearing the coherent Σ3 boundary in the boundary plane. As a result, the boundary takes a zig-zag configuration (light blue atoms) which decreases its energy. In fact, the alt-Σ3 has the lowest observed energy of 278 mJ/m² in comparison to 293 mJ/m² for the Σ3 boundary. In order to verify the unique low energy structure of the alt-Σ3 structure we also employed the SNAP. Fig. 2c shows the SNAP predicted alt-Σ3 which also has a structure and energy similar to the one predicted by EAM, indicating that the alt-Σ3 structure is not a potential-dependent artificial GB structure.

Higher energy Σ3 boundaries (1298 mJ/m²) are also identified when the GB plane is changed to (111) from (112) (tilt: <011>). Ab-initio calculations of the low energy Σ3 boundary in tungsten [33] and iron [34] only evidence fully symmetric structures, but such symmetry was reinforced by small system size in combination with a priori atom displacements and minimizations. Broken and/or conserved mirror symmetry ultimately serves as a primary indication of atomic accuracy in bcc transition metal potentials owing to the strong influence of non-spherically symmetric d-orbitals.

Similarly, the Σ5 boundary (Fig. 3) structure breaks mirror symmetry; this contrasts the early work of Ochs et al. [35], which shows, using simulations, that the Ta Σ5 GB has full mirror symmetry. The Σ5 is composed of “B” structural units analogous to those identified in Fe [6]. The presently calculated GBE of 1318 mJ/m² is markedly less than the previously predicted 1544 mJ/m² [35]. More recent calculations using model generalized pseudopotential theory (MGPT) do predict a break in mirror symmetry for this boundary [22]. Ab-initio work by Ochs et al. [35] does identify other bcc transition metal elements (Ta, W, and Nb) that break mirror symmetry. For Nb and Mo, there exist experimental high-resolution transmission electron microscopy (HRTEM) evidence for both structures with conserved and broken symmetry [36, 37]. The relative shift across the boundary identified here for Ta is 0.81 Å (as measured by the vertical displacement between opposing red atoms in Fig. 2b) as compared to ~0.78 Å for Mo. Relative to the respective lattice parameter (3.304 Å for Ta and 3.147 Å for Mo) the displacements are within ~1% of one another. If instead the shift is measured as the distance between atomic planes projected across the boundary, the shift is 0.55 Å, showing superb agreement to the experimentally observed shift of ~0.55 Å in Ta [22] (the MGPT value is not explicitly stated [22]).

Other boundaries corresponding to various energy minimas in Fig. 1 or comparable structures within the literature were also explored. Fig. 4 shows selected boundaries for each of the tilt axes. Here, the grain boundary units of Σ11 and Σ13 are expressly identified for comparison with other bcc metals. The structure of the Σ13(001) boundary is analogous to the HRTEM observations of boundaries in Mo by Morita and Nakashima [31] and the Σ11(011) structure agrees well with the density functional theory (DFT) calculations of Σ11 boundary in iron [34]. A large majority of the (112) tilt axis boundaries exhibit large (periodic) boundary unit cells such as that shown for the Σ11(112). There exist multiple other predicted boundary structures that break mirror symmetry such as the alt-Σ27, alt-Σ17 and alt-Σ13(111). Notably, the Σ7 boundary also exhibits both a symmetric and quasi-symmetric structure of nearly equivalent energies, further suggesting that the phenomenon of symmetry breaking is prevalent for many boundaries in bcc transition metals.

The classic processing–structure–properties–performance material relationship underscores the need to identify grain boundary structures and their energies. Here we show a large number of grain boundary energies for tantalum, as a function of tilt axis and misorientation, in order to inform future studies such as those investigating abnormal grain growth [10], heterogeneous deformation of poly and nanocrystals.
[9,39], and deformation twinning [11]. Specifically, deformation twinning involves the nucleation of Σ3 coherent twin boundaries through the movement of twinning dislocations. It is shown here that there is a decrease in energy associated with the formation of quasi-symmetric boundaries. In many cases, twin boundaries are not fully coherent and are likely combinations of symmetric, asymmetric, and quasi-symmetric components. The metastability of the quasi-symmetric alt-Σ3 boundaries in bcc tantalum, identified using both EAM and SNAP interatomic potentials, may play a governing role in determining the active mechanism for plastic deformation via the twinning vs. slip transition. Previous experimental evidence has shown quasi-symmetric Σ5 GBs boundaries in bcc Nb, Mo, and Ta; in the present work we identify symmetry breaking Σ3, Σ5, and Σ7 GBs (among many others) in Ta. These quasi-symmetric structures appear to be unique to bcc transition metals and are not explicitly reported for fcc GB structures.

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References


Fig. 4. Assorted low energy grain boundary structures for each of the four tilt axis with equivalent coloring scheme as Fig. 3. From left to right the boundaries are: Σ13(001), Σ11(011), Σ27(011), Σ7(111), alt-Σ7(111), Σ13(311), Σ11(131).