

First-principles Calculation on the Core Structure of a Screw Dislocation in BCC Molybdenum

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Predicting atomistic properties of a dislocation is a first step toward an understanding of plastic behavior of materials, in particular BCC metals. The core structure of a screw dislocation in BCC molybdenum has been studied over the years using the first-principles and empirical methods, however, their conclusions vary due to the inefficiency of the methods. We executed the first-principles calculation based on the density functional method to determine the core structure of a screw dislocation of molybdenum on the Earth Simulator (ES) by monitoring the \mathbf{k} -point sampling and energy cutoff convergence. We have concluded that the core of a screw dislocation in BCC molybdenum has a 6-fold structure.

Keywords: first-principles calculation, screw dislocation, core structure, bcc, molybdenum

1. INTRODUCTION

Electronic and atomistic bonding states of materials are required to understand fundamental mechanical properties of nuclear materials, in particular, embrittlement and fracture due to irradiation. Hydrogen embrittlement in BCC metals is one of the important fundamental phenomena associated with the stress corrosion cracking observed in light-water nuclear reactors. Before studying the interaction of a hydrogen atom and a dislocation, it is first necessary to understand a behavior of a screw dislocation in BCC metals, in particular, the dislocation core structure.

Numerous first-principles and empirical atomistic simulation studies have been carried out to determine the core structure of a screw dislocation in BCC molybdenum. However, the results of the core structure differ depending on the simulation methods. Essentially, two core structures have been proposed for a screw dislocation in BCC molybdenum by various computational approaches [1]. The 3-fold structure (Fig. 1(a)) is obtained using the Finnis-Sinclair potential and the modified generalized pseudopotential theory (MGPT) potential, while the 6-fold structure (Fig. 1(b)) is obtained by the tight-binding and the density functional theory (DFT) methods. Although the DFT method gives the most accurate energy among these methods, its supercells are severely limited where the effects of core overlapping

and image stress can be significant. Moreover, \mathbf{k} -point samplings and energy cutoff convergence should be carefully monitored in calculating the BCC molybdenum dislocation core structure. These points have not been carefully considered in the previous DFT simulations.

There are essentially two factors to be considered in determining the core structure of a screw dislocation using the



Fig.1 (a) Differential displacement map of 3-fold structure

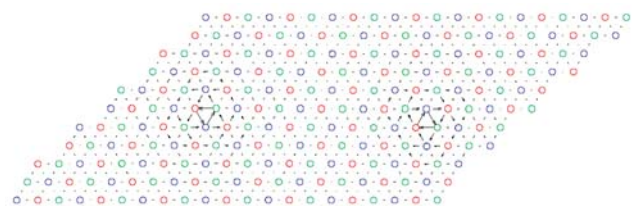


Fig.1 (b) Differential displacement map of 6-fold structure

Fig. 1 Two proposed structures of a screw dislocation core structures in BCC molybdenum

atomistic method. One is the boundary condition, and the other is the interatomic potential. Since a screw dislocation has a long-range strain field around the core, the simulation region should be taken as large as possible to reduce the effects of the boundaries. In order to take a large simulation region, an empirical potential, such as, the embedded atom method (EAM), for example, the Finnis-Sinclair potential, is introduced to reduce the computational burden of calculating the interatomic forces. However, the empirical potentials are basically constructed by fitting the parameters to the equilibrium states, and there is no guarantee for the empirical potential to describe the disordered core region accurately. On the other hand, although the first-principles calculation based on the density functional theory gives the most accurate results even for the core region, the computational region is limited to a very small area. Woodward et al. [2] proposed a boundary condition for the supercell containing one screw dislocation, in which atomic displacements due to a screw dislocation in the cell are allotted by the Green function method. Ju et al. [1] introduced a pair of screw dislocations in the supercell and proposed a shape of the supercell in such a way as to cancel the strains among supercells. They evaluated the accuracy of the core energy by changing the size of the supercell, and concluded that the supercell containing 231 atoms is good enough for the DFT calculation. In this paper, we perform the DFT calculations using the supercell containing 231 atoms.

2. COMPUTATIONAL METHOD

We have employed the VASP (Vienna ab-initio simulation package) code, which implements the density functional theory with the ultrasoft pseudopotential and the generalized gradient approximation (GGA) for the exchange correlation energy, for calculating accurately the total energy of the system. The original VASP code is parallelized, however, we found that the parallel speedup is not obtained using over 40 processors on the ES for this problem. In order to exploit the vector processors on the ES system, we have tuned this VASP code by replacing the one-dimensional FFT routine by the numerical library ASL/ES for science and engineering. We have also parallelized the code in terms of \mathbf{k} -points, since we concern here the effects of the \mathbf{k} -point sampling. We have achieved almost linear speedup using up to 80 processors, and found that the \mathbf{k} -point parallelization has almost no influence on the average vector length and vectorization ratio.

There are two conditions to be considered for the accurate calculation of the DFT method. One is the energy cutoff convergence for the plane wave expansion of the wavefunction, and the other is \mathbf{k} -point sampling in using the supercell. We have adopted the value of 233eV for the energy cutoff convergence, because this value is confirmed to be highly

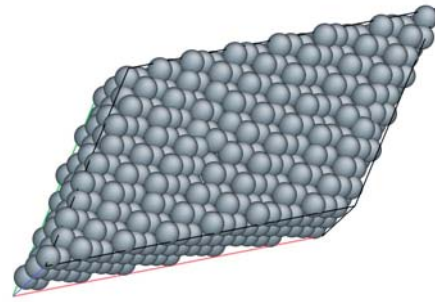


Fig. 2 The BCC Mo supercell consisting of 231 atoms

Table 1 The total energy for the model system with a pair of screw dislocations having 3-fold core structure as a function of \mathbf{k} -points.

\mathbf{k} -points	Total energy [eV]	Energy difference [eV]
1x1x20	-2493.89832301	0.158
2x2x40	-2494.05628398	0.0047
3x3x60	-2494.05157068	

accurate for the calculation of elastic constants [3], which tends to lead to erroneous results without a proper cutoff value. The energy cutoff convergence depends almost on the atom species and is taken to be large enough for the present lattice structure and boundary conditions. Here, we have performed a detailed study on the effects of \mathbf{k} -point samplings using the cutoff energy of 233eV and the supercell consisting of 231 atoms (Fig. 2) on the ES. We show in Table 1 the results of the relation between the total energy and the number of \mathbf{k} -points for the model system containing a pair of screw dislocations having 3-fold core structure. We found that the results with 2x2x40 \mathbf{k} -points have already converged to the energy difference of 4.7meV corresponding to 0.02meV per one atom.

3. RESULTS AND DISCUSSION

Firstly, we started with the initial supercell containing 231 atoms and a pair of screw dislocations with the 3-fold core, and relaxed this atomistic system using the \mathbf{k} -points of 1x1x20. We have found that this turns into a stable 6-fold core structure, and there exists no energy barrier between the two structures. This indicates that the 6-fold core structure is stable, and the 3-fold core structure is not even meta-stable. The energy difference between the 3-fold and the 6-fold core structures for the whole system is estimated to be 0.798eV, that is 0.399eV for each dislocation. Secondly, using the \mathbf{k} -points of 2x2x40, we have evaluated the forces on each atom and the total energy for the systems containing the initial 3-fold and the 6-fold core structures, respectively, keeping the positions of the atoms constant. As a result, in the case of the 3-fold core structure, the forces on each atom using the

2x2x40 \mathbf{k} -points correspond to those using 1x1x20 \mathbf{k} -points with the energy order of 0.01eV per each atom. The accuracy of the results for the case of the 6-fold core structure between the two cases of \mathbf{k} -points is within the energy of 0.01eV per each atom. On the basis of these results, we can conclude that the 6-fold core structure will be obtained by the relaxation process using the \mathbf{k} -points of 2x2x40. We have determined that the energy difference of the two systems is 0.813eV. The value obtained using the \mathbf{k} -points of 1x1x20 is found to be very close to the above value and well converged. With all the above results and analyses, we have concluded that the core of a screw dislocation in the BCC molybdenum has a 6-fold structure.

4. SUMMARY

We have implemented the VASP (Vienna ab-initio simulation package) code on the Earth Simulator by parallelization in terms of \mathbf{k} -points and achieved a good parallel performance. Using this first-principles method, we have simulated the core structure of a screw dislocation by carefully checking the \mathbf{k} -point sampling and the energy cutoff convergence for the plain wave expansion. We have concluded that core structure of a screw dislocation in BCC molybdenum is 6-fold.

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第一原理計算によるBCCモリブデン中らせん転位の芯構造の決定

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転位の原子レベルの性質を予測することは、物質の塑性変形挙動の機構を理解するために重要である。特に、BCC金属においてはらせん転位のパイエルズ応力が大きいこともあり、原子レベルからのその挙動の理解が求められている。BCC結晶構造をもつモリブデン金属においては、第一原理計算及び経験的手法を用いてらせん転位の芯の構造を決定する計算が数年来行われてきたが、計算手法が十分でないこともあり、手法により異なる結果が得られている。ここでは、密度汎関数法に基づく第一原理計算法を並列化して地球シミュレータ上に実装し、k点サンプリング及び波動関数の平面波展開におけるエネルギーカットオフ値に特に注意を払うことにより計算を行い、BCCモリブデン中のらせん転位の芯構造を決定した。その結果、BCCモリブデン中のらせん転位芯は6回軸構造を持っていることが分かった。

キーワード: 第一原理計算, らせん転位, 芯構造, 体心立方格子 (BCC), モリブデン