Exploring the Possibility of Deformation Twinning in Pure Aluminum

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Abstract Can aluminum generate deformation twins? Contradictories exist among the experimental results, theoretical predictions, and molecular dynamics simulations. Our first-principles calculations based on the full-potential linearly augmented plane wave method show that the bonding electron density of aluminum gathers at the tetrahedral interstices, and this specific electronic structure is in agreement with the experimental data from quantitative convergent-beam electron diffraction. In terms of this characteristic, the matching between the cohesive energy of local deformation twins with different thickness and global matrix can shed light on deformation twins in aluminum. This study may give a lane to insight into deformation twins in different metals.

KEY WORDS: Metals and alloys; Deformation twinning; First-principles; Density functional theory

1 Introduction

Deformation twinning (DT) is a basic deformation mechanism in many materials such as pure metals, steels, shape-memory alloys, rocks, ferroelectric, and ferromagnetic materials [1–5]. In general, a consensus is that the preference for DT in face-centered cubic (fcc) metals depends largely on their stacking fault energies (SFEs). For instance, coarse-grained Cu, Al, and Ni with medium-to-high SFE prefer to deform via dislocation slip at ambient temperature and low strain rate; while Ag and Au with low SFE can deform by DT [6, 7]. In addition, the addition of Al into Cu will make it easier to generate deformation twins in Cu–Al alloys owing to the decrease of SFE [8, 9].

Generally, SFE is regarded as a critical parameter that determines whether DT happens or not. In many fcc metals and alloys, there is a reasonable tendency to form deformation twins with the decrease of SFE. Among the previous studies, DT was observed in Ir, while DT was rarely reported in bulk Al samples, although its SFE is lower than Ir [10]. In addition, three criteria present the similar result that Al can hardly deform via DT. The first criterion was proposed a decade ago by Bernstein and Tadmor [11] to predict twinning tendency based on the heterogeneous dislocation nucleation from a crack tip and the generalized SFE curve. The second one was proposed by Asaro and Suresh [12] in terms of the inhomogeneous dislocation nucleation from a grain boundary and the generalized SFE curve. The third one was proposed by Cai et al. [13] to predict the twinning tendency in the framework of the generalized SFE curve. However, molecular dynamics (MD) simulations [14–16] demonstrate that DT can still occur in Al with a sufficiently small grain size.

In addition to the theoretical predictions and MD simulations, various experiments have been performed to investigate the DT mechanism in single-crystalline, coarse-

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grained, and nanocrystalline Al. Firstly, almost no deformation twins have been experimentally observed in single or coarse-grained pure Al [10], even under shock loading at very low temperatures [17]. Secondly, the occurrence of Shockley partial dislocations from a crack tip of Al has been regarded as an evidence of DT; however, more detailed evidences supporting this result have not been observed again. Thirdly, deformation twins have been observed in nanocrystalline Al after severe plastic deformation [18–22]. The last one is that microtwins have been observed in single-crystalline Al via equal-channel angular pressing (ECAP) with the perfect crystallographic orientation design [23]. In contrast to these observations, twinning and detwinning have been captured for polycrystalline Al with grain sizes from several micrometers to 200 μm in tensile experiment [24].

According to the results above, there are some mismatches among different experimental results, MD simulations, and different theoretical predictions for the generation of deformation twins in Al. Firstly, the three criteria present the similar prediction results that DT can hardly occur in Al [11–13], while MD simulations illustrate the presence of deformation twins in Al with a very small grain size [14–16]. Secondly, DT can occur in single-crystalline and nanocrystalline Al [18–22], whereas twinning and detwinning have been captured in coarse-grained Al [24]. It is obvious that those theoretical predictions and MD simulations are not in good agreement with the experimental results. These analyses above give rise to an interesting question whether Al can deform via DT or not. Is there a channel to understand and explicate the contradictions between the theoretical prediction and experimental results?

In view of those contradictions above, a computational model is proposed in the present study based on a fundamental hypothesis that a successive deformation twin is generated in Al matrix. And then the cohesive energy of this deformation twin with different thickness is calculated by first-principles method, which is based on the full-potential linearly augmented plane wave (FPLAPW) method incorporating local orbital in the framework of density functional theory (DFT) as packaged in WIEN2k code [25]. In succession, the difference between the cohesive energy of fcc Al and a successive deformation twin with different thickness in the range from 0.5 to 7 nm is responsible for whether Al can deform via DT or not. This method has been used to solve other problems previously [26, 27]. Finally, the particular electronic structure of fcc Al is that the bonding electron density locates at the tetrahedron interstices, which has been confirmed by the quantitative convergent-beam electron diffraction (QCBED) experiment [28].

2 Computation Methods

The present computational results are calculated by means of FPLAPW method incorporating local orbital within DFT which is packaged in WIEN2k code [25]. The generalized gradient approximation (GGA) of Perdew *et al.* [29] is used for exchange–correlation energy functional. And the muffin-tin radius of Al atom is 2.0 Bohr. The size of the basis set is given by the product $R_{\text{mt}} \times K_{\text{max}} = 8.0$, where K_{max} is the largest reciprocal space wave vector in the basis set and R_{mt} is the smallest muffin-tin radius of atom. The separation energy between inner electrons and valence electrons is -81.6 eV in the calculation of exchange–correlation energy. The energy accuracy is thus less than 0.00001 eV. 3000 k points can converge our total energy in the first Brillouin zone for all computations. The calculated bulk properties of $a = 4.038$ Å, $B = 72.6$ GPa, and $E_{\text{coh}} = 3.517$ eV (lattice constant, bulk modulus, and cohesive energy of Al, respectively) are consistent with the experimental results of $a = 4.05$ Å [30], $B = 72.2$ GPa [31], and $E_{\text{coh}} = 3.39$ eV [32].

3 Results and Discussion

The computational mode is illustrated in Fig. 1. The mode is built along x , y , and z directions, where x is equal to one $\frac{1}{2}\langle\bar{1}10\rangle$, y is equal to one $\frac{1}{6}\langle11\bar{2}\rangle$, and z is equal to $2h$ along the $\langle111\rangle$ direction. This mode shows successive twins with the thickness h , and...ABCACBA... layer-to-layer

Fig. 1 Computational mode of twinned Al with thickness h . The structure is composed of layers A, B, C, A, C, B, A, filled by the tetrahedral interstices that are colored with the light magenta and the octahedral interstices that are colored with the bright yellow, in which T4, T5, T6 and T7 are the representative tetrahedral interstice sites

structure, in which A represents a twin boundary (TB). The thickness h is a successive variable, which can adjust the thickness of deformation twins. A series of successive deformation twins are constructed, the cohesive energy of these models with different thickness is calculated by the first-principles method, and the computational results are presented in Fig. 2. It is clear that the cohesive energy of deformation twins gradually increases as h increases layer by layer. When h increases to about 3 nm, the cohesive energy converges at the about 3.5 eV, which is still apart from that of fcc Al [32]. Although the cohesive energy of the deformation twin converges at one equilibrium state when h is greater than 3 nm, it is not a stable structure in contrast to perfect fcc Al, which is just a metastable state. In view of this, even if DT can occur in single-crystal Al and deformation twins can generate layer by layer, no matter how thick it grows up, the cohesive energy of the local deformation twins cannot arrive at that of the global fcc Al. Therefore, the difference between the cohesive energy of local deformation twins and global fcc Al results in that the equilibrium state for deformation twins disappears through inverse twinning, i.e., detwinning, as observed in the previous study [24], unless the samples are Al film [18] and nanocrystalline Al [19–21].

Why the cohesive energy of deformation twins cannot approach to the value of perfect fcc Al with increasing h ? In view of the particular electronic structure of Al, this question should be well resolved. The deformation electron density $\Delta\rho$ is defined as below,

$$\Delta\rho = \rho_{\text{total}} - \rho_{\text{IAM}} \quad (1)$$

where ρ_{total} is the total electron density and ρ_{IAM} is the density associated with free, unbonded atoms, otherwise

known as the independent atom model (IAM) [33]. The iso-surface and contour of $\Delta\rho$ in fcc Al are illustrated in Fig. 3. It is obviously shown that the $\Delta\rho_{\text{tet}}$ (the deformation electron density at the centers of the tetrahedral interstices) is greater than the $\Delta\rho_{\text{oct}}$ (the deformation electron density at the centers of the octahedral interstices) and the positive $\Delta\rho$ nearly concentrates at the tetrahedral interstices, which is in good agreement with the experimental result by means of QCBED method [28].

Similarly, the deformation electron density $\Delta\rho$ of a deformation twin is illustrated in Fig. 4. It is found in Fig. 1 that the bridge between the tetrahedral interstices can merely rely on the atom (a point) or edge (line) between the nearest atoms and never depend on the lattice plane in the fcc Al [28]. However, this rule is broken when the DT occurs in fcc Al. The bridge between the tetrahedral interstices can also rely on lattice plane, such as a $\langle 111 \rangle$ plane, because TBs are generated in fcc matrix. In general, the relative positions between T6 (a tetrahedral interstice) and T7 (another tetrahedral interstice) are similar to those between T4 (a tetrahedral interstice) and T5 (a tetrahedral interstice) which are linked via an edge; however, after DT occurs, both T6 and T7 are linked via a lattice plane (TB plane). These changes directly result in the decrease in the distance between T6 and T7 from $a/2$ to $\sqrt{3}a/6$ (a is the lattice constant of fcc Al). Therefore, the original repulsive force between T6 and T7 is expressed as follows:

$$F_0 = k \frac{\Delta\rho_{T6} \times \Delta\rho_{T7}}{(a/2)^2}, \quad (2)$$

and the resultant repulsive force between T6 and T7 is given as:

Fig. 2 Comparison between the cohesive energy of twinned Al with different thickness and a perfect fcc Al: **a** original cohesive energy and **b** normalized cohesive energy

Fig. 3 **a** Iso-surface value $\Delta\rho$ with $0.0275 \text{ e}/\text{\AA}^3$ in the unit cell of Al, located at the tetrahedral interstice. **b** Contour of $\Delta\rho$ in $\{110\}$ plane-sectioned planes including the tetrahedral and octahedral interstices. The contours are the six intervals from -0.075 to $0.055 \text{ e}/\text{\AA}^3$

Fig. 4 Contour of iso-surfaces for **a** $\Delta\rho = 0.0284 \text{ e}/\text{\AA}^3$, **b** $\Delta\rho = 0.0231 \text{ e}/\text{\AA}^3$, and **c** $\Delta\rho = 0.0184 \text{ e}/\text{\AA}^3$, respectively. **d** Contour of $\Delta\rho$ in plane-sectioned plane including the tetrahedral and octahedral interstices. The contours are the six intervals from 0 to $0.04 \text{ e}/\text{\AA}^3$

$$F_1 = k \frac{\Delta\rho_{T6} \times \Delta\rho_{T7}}{(\sqrt{3}a/6)^2}, \quad (3)$$

where k is the Coulomb constant. Because $F_0 < F_1$, the cohesive energy of deformation twins is lower than that of fcc Al.

In Fig. 4a, there are two small bonding electron densities in the adjacent of TB plane. As the value of iso-surface gradually increases, these bonding electron densities fuse into one bonding electron density $\Delta\rho_1$ from Fig. 4b, and then $\Delta\rho_1$ gets bigger and another bonding electron density $\Delta\rho_2$ appears in the TB plane and near $\Delta\rho_1$ from Fig. 4c. $\Delta\rho_1$ is defined as double-core-pseudo-charge, and $\Delta\rho_2$ is also defined as extra-pseudo-charge. The repulsive force between double-core-pseudo-charge $\Delta\rho_1$ and extra-pseudo-

charge $\Delta\rho_2$ minifies the cohesive energy of deformation twins because these repulsive forces and TBs are symbiotic in fcc Al matrix.

Although the cohesive energy of local deformation twins is far smaller than that of the global fcc Al matrix, the cohesive energy of global matrix decreases when the grain size is smaller (the global matrix changes from single-crystalline to coarse-grained and nanocrystalline Al), because the volume fraction of atoms in the grain boundaries (GBs) gets larger and larger, and the energy of atoms in GB is higher than that of atoms in bulk sample. Why could twinning and detwinning be captured in the coarse-grained crystalline Al? Because the cohesive energy of the coarse-grained crystalline Al with grain sizes ranging from several micrometers to 200 μm is still smaller than that of

the deformation twins, the stability of local deformation twins can match that of global Al matrix [24]. As a result, it is not difficult to understand why deformation twins were observed in the nanocrystalline Al film with thickness of 200–400 nm [19–21], and a grain size of 10–35 nm [18]. The latest experimental result [34] presents that deformation twins are captured in single Al; however, the figure shows that the deformation twins are also embedded in polycrystalline Al, and the grain size just is large.

4 Conclusions

The matching of cohesive energy between the local deformation twins and the global matrix in Al should be responsible for the contradictories among the experimental results, theoretical predictions, and MD simulations. Firstly, when the cohesive energy of deformation twins is far smaller than that of the single- and coarse-grained crystalline Al, the deformation twins in the matrix should behave in detwinning synchronously. Secondly, when the cohesive energy of local deformation twins is close to that of global nanocrystalline Al, deformation twins can form in nanocrystalline Al film. In nature, the particular electronic structure of Al that bonding electron density gathers at the tetrahedral interstices underlays the inconsistency between the experimental results and theoretical predictions for the formation of deformation twins in Al.

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