



Twinnability of Al–Mg alloys: A first-principles interpretation

Dong-dong ZHAO¹, Ole Martin LØVVIK², Knut MARTHINSEN¹, Yan-jun LI¹

1. Department of Materials Science and Engineering,
Norwegian University of Science and Technology (NTNU), 7491 Trondheim, Norway;

2. SINTEF Materials and Chemistry, 0314 Oslo, Norway

Received 3 May 2016; accepted 18 October 2016

Abstract: Al–Mg alloys are considered to have potentials to form twins during deformation because Mg can reduce the intrinsic stacking fault energy γ_{ISFE} of Al. Nevertheless, twinning has rarely been found in Al–Mg alloys even subjected to various severe plastic deformation (SPD) techniques. In order to probe the twinning propensity of Al–Mg alloys, first-principles calculations were carried out to investigate the effects of Mg and vacancies on the generalized planar fault energy (GPFE) of Al. It is found that both Mg and vacancies exhibit a Suzuki segregation feature to the stacking fault, and have the influence of decreasing the γ_{ISFE} of Al. However, γ_{ISFE} does not decrease and the twinnability parameter τ_a of Al does not increase monotonically with increasing Mg concentration in the alloy. On the basis of τ_a evaluated from the calculated GPFE of Al–Mg alloys, we conclude that deformation twinning is difficult for Al–Mg alloys even with a high content of Mg. Besides, the decrease of γ_{ISFE} caused by the introduction of Mg and vacancies is supposed to have the effect of improving the work-hardening rate and facilitating the formation of band structures in Al–Mg alloys subjected to SPD.

Key words: generalized planar fault energy; intrinsic stacking fault energy; Suzuki segregation; Mg; vacancy; Al–Mg alloys

1 Introduction

Nano twin structures have received great interest since they are effective in improving material strength without sacrificing ductility [1]. In the past decades, massive efforts have been devoted to achieving deformation twinning in Al alloys, aiming to further increase their strength. However, it is found difficult to fabricate deformation twin structures in Al alloys even subjected to various severe plastic deformation (SPD) techniques, including equal channel angular pressing (ECAP) [2], high pressure torsion (HPT) [3], dynamic plastic deformation (DPD) [4], which is ascribed to the high intrinsic stacking fault energy γ_{ISFE} and high ratio of unstable twinning fault energy γ_{UTFE} to unstable stacking fault energy γ_{USFE} . Several works have suggested that Mg can decrease the γ_{ISFE} of Al and is thereby considered as a promising alloying element to increase the twinning tendency of Al. Nevertheless, it remains controversial whether Mg solutes can drastically decrease the γ_{ISFE} of Al. Therefore, further investigation is needed to clarify

the Mg concentration effect on γ_{ISFE} in Al–Mg alloys. Moreover, a large density of vacancies will be created during the process of SPD [3]. Higher Mg concentration in Al alloys subjected to SPD would facilitate a higher supersaturation of vacancies [5]. As an important category of defects, vacancies have long been believed to influence various materials properties since they participate in many crucial materials processes, i.e., diffusion, precipitation, and segregation. Up to now, rare focus has been put on the vacancy effect on generalized planar fault energy (GPFE) of Al and its alloys, except for LU and KAXIRAS [6], ASADI et al [7], both of which have predicted that vacancy can reduce the γ_{ISFE} of Al. Thus, the vacancy effect upon γ_{ISFE} in Al alloys processed by SPD needs to be considered.

GPFE curve has been proven to be effective in characterizing the deformation behavior of crystalline materials and has attracted extensive attention in recent years [8]. Generally, there are four typical energy extrema along the GPFE curve, i.e., unstable twinning fault energy γ_{UTFE} , intrinsic stacking fault energy γ_{ISFE} , unstable stacking fault energy γ_{USFE} and twinning fault

energy γ_{TFE} (two-layer micro twin), which is equivalently the extrinsic stacking fault energy γ_{ESFE} . GPFE curves can be used to investigate the nucleation of dislocations [9], twinning propensity [10], as well as the plastic deformation regimes in nano-crystalline (nc) materials [11], etc.

In the present work, a systematic first-principles study was initiated to investigate the impurity effect of Mg solutes and vacancy upon GPFE of Al, trying to probe the twinnability and the variation of γ_{ISFE} of Al–Mg alloys subjected to SPD.

2 Calculation

The calculation of GPFE in FCC metals involves the rigid displacement of half the crystal in reference to the other along the $\langle 11\bar{2} \rangle$ direction. A slab shear methodology was adopted to calculate the GPFE curves. As indicated in Fig. 1(a), vacuum spacing of 15 Å was utilized to prohibit the interactions of stacking fault. The first displacement operation with distance $a_0/\sqrt{6}$ was enforced for (111) planes numbered 1–6 to create a stacking fault. Afterwards, (111) planes numbered –1 to –5 were displaced in the opposite direction to form a two-layer micro-twin. The total energy of the intermediate configurations along the displacement path was calculated via selectively relaxing the atomic positions in the direction perpendicular to the stacking fault plane.

All the calculations in the present work were carried out using Vienna ab initio simulation package (VASP) [12,13]. The ion–electron interaction was described by the projector augmented wave (PAW) method and the exchange–correlation functional was

described by the generalized gradient approximation (GGA) [14]. A k -points sampling of $13 \times 7 \times 1$ for the Brillouin zone was performed using the Monkhorst–Pack scheme [15]. The plane wave cutoff energy was set to 350 eV, which was sufficient to converge the total energy to be within 1 meV/atom.

3 Results and discussion

3.1 Generalized planar fault energy (GPFE) curve of Al

Generally, there are three distinct deformation regimes in FCC metals, i.e., twinning (TW), full slip (FS), stacking fault (SF). Figure 2 shows the GPFE of Al, indicating the energy barriers for the activation of these three deformation regimes. The first energy extrema correspond to γ_{USFE} , which is the energy barrier for the emission of leading partial dislocations. The propagation of leading partial will produce a stacking fault, of which the energy is indicated as γ_{ISFE} in Fig. 2. A competition would persist between TW, FS and SF deformation regimes after the formation of stacking fault. As shown in Fig. 2, SF needs the highest energy penalty to occur and is rarely activated in Al. Along TW path, γ_{UTFE} is the energy barrier for nucleation of twin partial dislocation, an important parameter to evaluate twinning propensity. As shown in Table 1, Al possesses a high $\gamma_{\text{UTFE}}/\gamma_{\text{USFE}}$, and low twinnability parameter τ_a (defined by Eq. (1), larger than unity would signify twinning [16]), which accounts for the difficulty of activating twinning in Al.

$$\tau_a = \left[1.136 - 0.151 \frac{\gamma_{\text{ISFE}}}{\gamma_{\text{USFE}}} \right] \sqrt{\frac{\gamma_{\text{USFE}}}{\gamma_{\text{UTFE}}}} \quad (1)$$

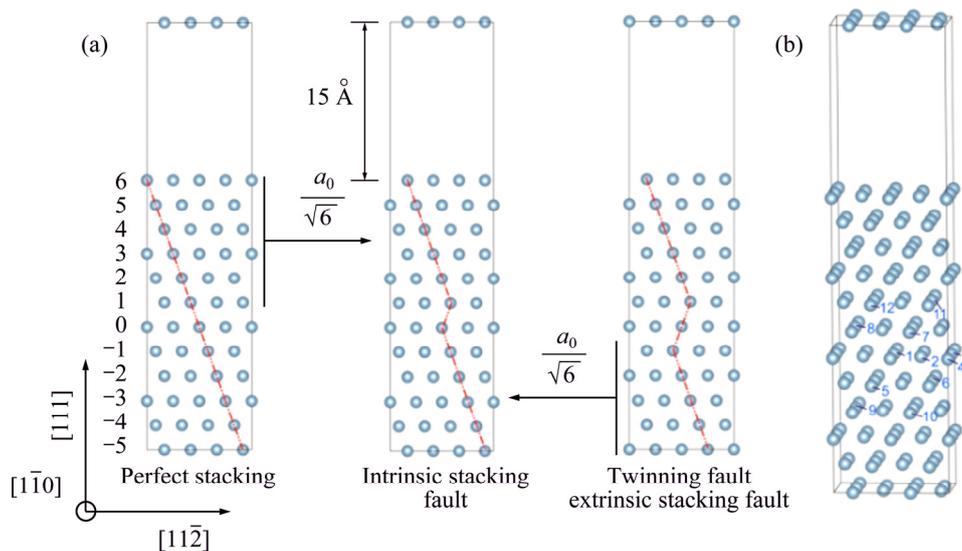


Fig. 1 Displacement path for calculation of GPFE potential substitutional positions for Mg (a) and vacancy to evaluate their effect upon GPFE of Al (b)

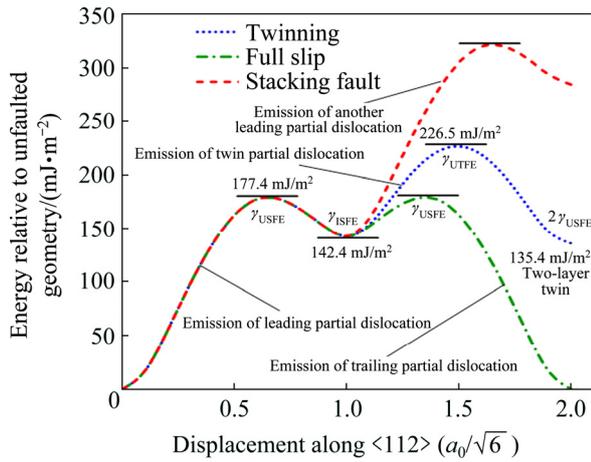


Fig. 2 Generalized planar fault energy (GPFE) curve of Al

3.2 Mono-Mg and mono-vacancy effect on GPFE curve of Al

Figure 3 shows the mono-Mg and mono-vacancy influenced GPFE of Al, where substitutional mono-Mg and mono-vacancy were introduced in the stacking fault plane. As can be seen in Fig. 3, both Mg and vacancy can decrease the γ_{ISFE} of Al, as will do for γ_{USFE} , γ_{UTFE} , and γ_{TFE} . However, mono-vacancy has a more severe decreasing effect on the GPFE curve of Al. For instance, mono-vacancy can reduce γ_{ISFE} of Al from 142.4 to 121.3 mJ/m^2 . Increased twinnability τ_a (cf. Table 1) attributed to Mg or vacancy introduction, compared to pure Al, indicates the twinnability-promoting role of Mg and vacancy.

3.3 Suzuki segregation of Mg and vacancy in Al

In order to investigate the Suzuki segregation of Mg and vacancy in Al, the interaction energy of Mg, vacancy with stacking fault were calculated based on the following equation:

$$E_{\text{int}}^n = (E_{\text{SF}}^{\text{Sol-n}} - E_{\text{PS}}^{\text{Sol-n}}) - (E_{\text{SF}} - E_{\text{PS}}) \quad (2)$$

where $E_{\text{SF}}^{\text{Sol-n}}$ and $E_{\text{PS}}^{\text{Sol-n}}$ are the corresponding total energies of stacking fault and perfect configurations, respectively, with Mg or vacancy residing at the n th layer (cf. Fig. 1). E_{SF} and E_{PS} are the total energies of stacking fault and perfect configurations without Mg or vacancy, respectively. As shown in Fig. 4, the negative E_{int}^n indicates that Mg or vacancy would like to segregate to the stacking fault of Al. The small energy barrier in layer 3 and -2 suggests the slight activated Suzuki segregation feature of Mg to intrinsic stacking fault of Al, which is distinct to the natural Suzuki segregation feature of vacancy, as shown in Fig. 4(b). It is worth noting that E_{int}^n is only noticeable at several atomic layers near the stacking fault region. Both Mg and vacancy exhibit an evidently activated Suzuki segregation character for extrinsic stacking fault in Al (cf. Figs. 4 (a) and (b)).

3.4 Complex Mg and vacancy effect on GPFE of Al

Figures 5(a) and (b) show the hybrid Mg and vacancy effect on the GPFE curves of Al. The detailed Mg and vacancy distributions in models S1–S7 can be referred to Fig. 1(b). The binding energy of Mg and vacancy was calculated to determine the energetically favorable distributions near the stacking fault region. As can be seen in Figs. 5(a) and (b), a different local distribution and local concentration of Mg and vacancy in the vicinity of stacking fault can produce different effects upon the GPFE of Al. All the models as presented in Fig. 5 have a decreased γ_{ISFE} compared with pure-Al (see detail in Table 1). Specifically, model S7 possesses the lowest γ_{ISFE} , i.e., 113.9 mJ/m^2 , which is probably attributed to the high vacancy density in the stacking fault region. As indicated in Figs. 5(a) and (b), a higher vacancy concentration in the vicinity of stacking fault

Table 1 Mg and vacancies effect on γ_{USFE} , γ_{ISFE} , γ_{UTFE} and γ_{TFE} of Al (The subscript number in the ‘‘Occupation’’ column corresponds to the atomic positions for Mg and vacancy substitutions as indicated in Fig. 1(b). The energies are in mJ/m^2)

System	Occupation	Mg content/%	γ_{USFE}	γ_{ISFE}	γ_{UTFE}	$2\gamma_{\text{TFE}}$	$\gamma_{\text{ISFE}}/\gamma_{\text{USFE}}$	τ_a (Twinnability)
Al ₉₆	–	–	177.4	142.4	226.5	135.4	0.803	0.898
^a Al	–	–	185	142	236	128	0.768	0.902
Al ₉₅ Mg ₁	Mg ₍₁₎	1.08	168.0	135.0	211.6	119.1	0.804	0.904
Al ₉₅ Va ₁	Va ₍₁₎	–	156.2	121.3	196.0	102.5	0.777	0.909
S1	Mg _(9,10,11,12) Va ₍₁₎	4.21	155.7	128.9	206.7	108.3	0.828	0.877
S2	Mg _(1,4) Va _(9,11)	2.13	158.5	131.4	204.1	113.9	0.829	0.891
S3	Mg _(1,4,10,12) Va _(9,11)	4.26	166.3	140.8	221.0	120.9	0.846	0.875
S4	Mg _(1,9,11) Va ₍₄₎	3.16	152.0	125.4	193.1	95.7	0.825	0.898
S5	Mg _(1,10,12) Va ₍₄₎	3.16	157.8	125.0	201.3	102.4	0.792	0.900
S6	Mg _(1,9,10,11,12) Va ₍₄₎	5.25	157.5	130.6	212.0	112.2	0.829	0.871
S7	Mg _(1,9,11) Va _(4,10,12)	3.26	138.0	113.9	195.2	105.3	0.825	0.850

^afrom Refs. [17,18]

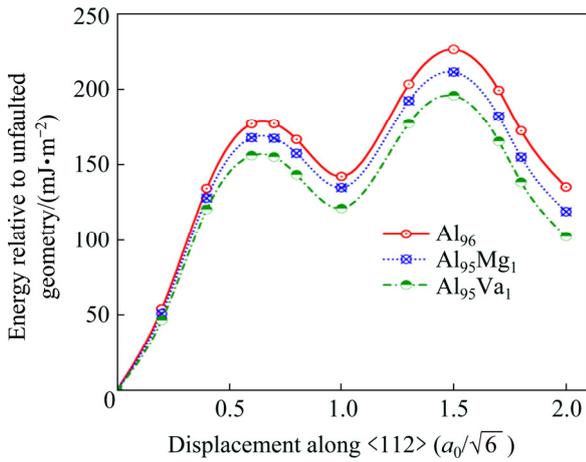


Fig. 3 Mono-Mg and mono-vacancy effect upon GPFE of Al (Mg and vacancy were placed in the stacking fault plane, with a planar content of 12.5%)

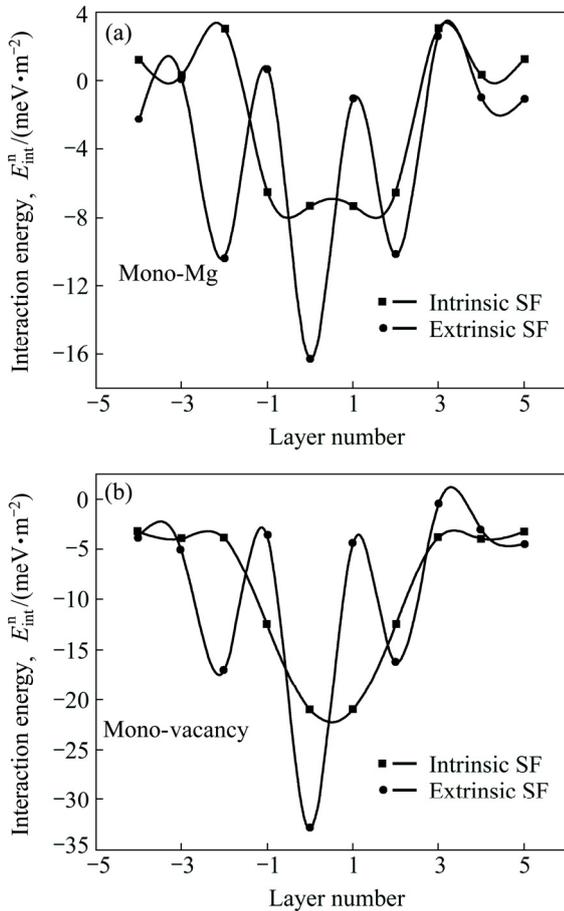


Fig. 4 Interaction energy of mono-Mg and stacking fault including intrinsic and extrinsic of Al at different layers (a), interaction energy of mono-vacancy and stacking fault including intrinsic and extrinsic of Al at different layers (b)

can produce a further reduced γ_{ISFE} . However, a detailed analysis of Table 1 implies that γ_{ISFE} does not decrease and the twinnability parameter τ_a does not increase monotonically with increasing Mg concentration in Al–Mg alloys. Note that all the models investigated in

the present work have a τ_a value being less than unity, indicating that deformation twinning is generally difficult for Al–Mg alloys even with a high content of Mg in the process of severe plastic deformation. More systematic analysis of the Mg and vacancy effect on the GPFE of Al can be found in Refs. [19,20].

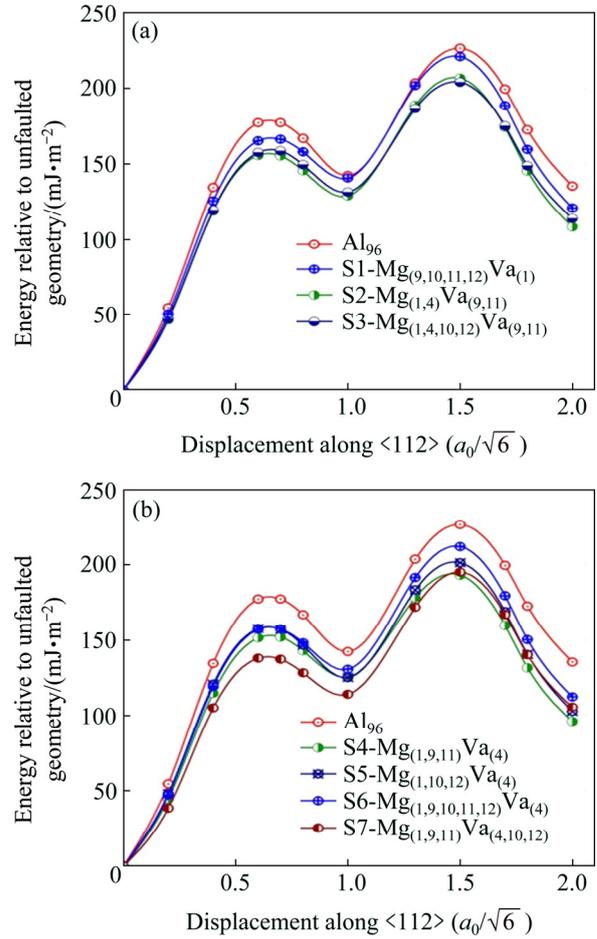


Fig. 5 Hybrid Mg and vacancy effect on GPFE curves of Al

3.5 Discussion

It is shown that Mg solutes, vacancy, as well as Mg-vacancy complexes can all decrease the γ_{ISFE} of Al. The reduction of γ_{ISFE} is expected to contribute to suppressing dynamic recovery in SPDed Al–Mg alloys. As is known that perfect dislocations turn to dissociate into partials and the split-up distance of a full dislocation is closely connected with γ_{ISFE} . High γ_{ISFE} materials usually correspond to a small split-up distance. Thus, a reduction of γ_{ISFE} would mean the increase of split-up distance, which would make it more difficult for the cross-slip of dislocations and thereby increase dynamic recovery suppression and work-hardening rate in plastic deformation. The decreasing effect upon γ_{ISFE} of Al attributed to Mg, vacancy, and Mg-vacancy complex can help contribute to suppress dislocation annihilation and increase work hardening rate in SPDed Al–Mg alloys.

Al belongs to high stacking fault energy materials,

of which dislocations have three dimensional mobility and feasible cross-slip. The massive cross-slip of dislocations would produce a wavy glide behavior of dislocations in the materials, which facilitates the formation of cell structure. Al–Mg alloys with high Mg concentration have been reported to exhibit planar glide behavior. As is discussed, Mg solutes, vacancy, as well as Mg-vacancy complexes can decrease the γ_{SFE} of Al and induce a more dissociate-up of perfect dislocations, which can therefore suppress cross-slip and increase the planar movement of dislocations. The planarity of dislocations would favor the formation of band structures in the plastic deformation. In other words, Mg and vacancy introduction can contribute to the formation of band structure in Al–Mg alloys subjected to severe plastic deformation.

4 Conclusions

1) First-principles calculations were initiated to investigate the effects of solute Mg and vacancy on the generalized planar fault energy (GPFE) of Al.

2) Both solute Mg atoms and vacancies were found to exhibit a Suzuki segregation feature to the stacking fault, and have a decreasing effect upon the γ_{SFE} of Al.

3) A monotonic decrease of γ_{SFE} and a continuous increase of twinnability parameter τ_a of Al with increasing Mg concentration were not observed.

4) The evaluated twinnability parameter τ_a calculated from the GPFE curves of Al–Mg alloys in the present work reveals that deformation twinning is difficult for Al–Mg alloys even with a high content of Mg in the process of severe plastic deformation.

5) The reduction of γ_{SFE} attributed to Mg and vacancy introduction is considered to contribute to the work-hardening rate and promote the formation of band structures in Al–Mg alloys subjected to severe plastic deformation.

References

- [1] LU K, LU L, SURESH S. Strengthening materials by engineering coherent internal boundaries at the nanoscale [J]. *Science*, 2009, 324: 349–352.
- [2] ZHA M, LI Y J, MATHIESEN R H, BJORGE R, ROVEN H J. Microstructure evolution and mechanical behavior of a binary Al–7Mg alloy processed by equal-channel angular pressing [J]. *Acta Mater*, 2015, 84: 42–54.
- [3] SAUVAGE X, ENIKEEV N, VALIEV R, NASEDKINA Y, MURASHKIN M. Atomic-scale analysis of the segregation and precipitation mechanisms in a severely deformed Al–Mg alloy [J]. *Acta Mater*, 2014, 72: 125–136.
- [4] JIN S B, ZHANG K, BJORGE R, TAO N R, MARTHINSEN K, LU K, LI Y J. Formation of incoherent deformation twin boundaries in a coarse-grained Al–7Mg alloy [J]. *Appl Phys Lett*, 2015, 107: 091901.
- [5] KUHLMANN-WILSDORF D. The connection between recovery and glide type in Aluminum and Al–Mg [J]. *Mater Sci Forum*, 2000, 331–337: 689–702.
- [6] LU G, KAXIRAS E. Can vacancies lubricate dislocation motion in aluminum? [J]. *Phys Rev Lett*, 2002, 89: 105501.
- [7] ASADI E, ZAEEM M A, MOITRA A, TSCHOPP M A. Effect of vacancy defects on generalized stacking fault energy of fcc metals [J]. *J Phys: Condens Matter*, 2014, 26: 115404.
- [8] ZHU Y T, LIAO X Z, WU X L. Deformation twinning in nanocrystalline materials [J]. *Prog Mater Sci*, 2012, 57: 1–62.
- [9] LU G, KIOUSSIS N, BULATOV V V, KAXIRAS E. Generalized-stacking-fault energy surface and dislocation properties of aluminum [J]. *Phys Rev B*, 2000, 62: 3099–3108.
- [10] KIBEY S, LIU J B, JOHNSON D D, SEHITOGLU H. Generalized planar fault energies and twinning in Cu–Al alloys [J]. *Appl Phys Lett*, 2006, 89: 191911.
- [11] SWYGENHOVEN H V, DERLET P M, FROSETH A G. Stacking fault energies and slip in nanocrystalline metals [J]. *Nature Mater*, 2004, 3: 399–403.
- [12] KRESSE G, FURTHMULLER J. Efficiency of ab-initio total energy calculations for metals and semiconductors using a plane-wave basis set [J]. *Comp Mater Sci*, 1996, 6: 15–50.
- [13] KRESSE G, FURTHMULLER J. Efficient iterative schemes for ab initio total-energy calculations using a plane-wave basis set [J]. *Phys Rev B*, 1996, 54: 11169–11186.
- [14] PERDEW J P, BURKE K, ERNZERHOF M. Generalized gradient approximation made simple [J]. *Phys Rev Lett*, 1996, 77: 3865–3868.
- [15] BLOCHL P E, JEPSEN O, ANDERSEN O K. Improved tetrahedron method for Brillouin zone integrations [J]. *Phys Rev B*, 1994, 49: 16223–16233.
- [16] HUNTER A, BEYERLEIN I J. Relationship between monolayer stacking faults and twins in nanocrystals [J]. *Acta Mater*, 2015, 88: 207–217.
- [17] LIU L L, WANG R, WU X Z, GAN L Y, WEI Q Y. Temperature effects on the generalized planar fault energies and twinnabilities of Al, Ni and Cu: First principles calculations [J]. *Comp Mater Sci*, 2014, 88: 124–130.
- [18] WU X Z, LIU L L, WANG R, LIU Q. The generalized planar fault energy, ductility, and twinnability of Al and Al–X (X=Sc, Y, Dy, Tb, Nd) at different temperatures: Al and Al–X (X=Sc, Y, Dy, Tb, Nd) at different temperatures: A first-principles study [J]. *Chin Phys B*, 2014, 23: 066104.
- [19] ZHAO D D, LOVVIK O M, MARTHINSEN K, Li Y J. Impurity effect of Mg on the generalized planar fault energy of Al [J]. *J Mater Sci*, 2016, 51: 6552–6568.
- [20] ZHAO D D, LOVVIK O M, MARTHINSEN K, Li Y J. Combined effect of Mg and vacancy on the generalized planar fault energy of Al [J]. *J Alloys Comp*, 2017, 690: 841–850.

Al-Mg 合金孪晶变形的第一性原理研究

赵冬冬¹, Ole Martin LØVVIK², Knut MARTHINSEN¹, 李彦军¹

1. Department of Materials Science and Engineering,

Norwegian University of Science and Technology (NTNU), 7491 Trondheim, Norway;

2. SINTEF Materials and Chemistry, 0314 Oslo, Norway

摘 要: 镁元素可以降低铝的本征层错能, 因而 Al-Mg 合金被认为具备孪晶变形的潜力。然而在多种大变形 Al-Mg 合金中很难发现变形孪晶。为了探究 Al-Mg 合金的孪晶变形潜能, 采用第一性原理计算研究镁和空位对铝广义层错能的影响。研究发现 Mg 和空位均具有层错 Suzuki 偏析特性, 并且会降低 Al 的本征层错能。但是随着镁含量的提高, 铝的本征层错能不会持续降低, 孪晶特性参数 τ_a 也不会持续升高。基于 Al-Mg 合金的孪晶特性参数 τ_a , 我们预测即使在高固溶镁含量下, Al-Mg 合金依然很难发生孪晶变形。镁和空位所引起的本征层错能的降低在一定程度上能够提高大变形 Al-Mg 合金的加工硬化速率并且促进变形带的形成。

关键词: 广义层错能; 本征层错能; Suzuki 偏析; 镁; 空位; Al-Mg 合金

(Edited by Yun-bin HE)