MOLECULAR DYNAMICS STUDIES ON THE LOCAL DISORDERING OF Σ3 AND Σ11 GRAIN BOUNDARIES IN ALUMINIUM BICRYSTAL*

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ABSTRACT: Molecular dynamics (MD) simulations using Morse interaction potential are performed in studies of [110] symmetrical tilt grain boundary (GB) structures with mis-orientation angles $50.5°(\Sigma 11)$, $129.5°(\Sigma 11)$, $70.5°(\Sigma 3)$ and $109.5°(\Sigma 3)$ at various tempratures. The GB structures are found to start local disordering at about $0.5T_m(T_m$ is the melting point of aluminium) for $50.5°(\Sigma 11)$, $0.32T_m$ for $129.5°(\Sigma 11)$ and $0.38T_m$ for $70.5°(\Sigma 3)$, respectively. These results agree with conclusions deduced from the anelastic measurements. But, for twin-boundary structure $109.5°(\Sigma 3)$, this disordering has not been found even when temperature increases up to $0.9T_m$.

KEY WORDS: molecular dynamics simulation, grain boundary disordering, aluminium bicrystal

I. INTRODUCTION

The stability of grain boundary (GB) structure at high temperatures has long been a problem in dispute. The main point is whether the grain boundary would change into a structure similar to that of the super-cooled liquid at temperatures much lower than the melting point T_m.

In respect of the experimental measurements, there are many different results by using electron microscope[1−4]. Glicksman and Vold[1] observed that the GB structures in bismuth bicrystals are replaced by a liquid-like layer at temperatures much lower than T_m. Erb and Gleiter[2] obtained that the GB structures in copper remain ordered at temperatures up to the melting point. Bulluffi et al.[3,4] found that the GB structures are not replaced by any liquid-layer and melting is not observed in aluminium even when the temperature rises to $0.999\% \ T_m$. Ke et al.[5,9] performed the anelastic measurements for the multi-crystal, bamboo-crystal and a series of aluminium bicrystals with different mis-orientation angles. They found that the physical quantities in anelastic relaxation have a sudden change at about $0.5T_m$ which appears to be caused by the behavior of GB[5−10].

In respect of the theoretical studies, Kikuchi and Cahn[11] used a two-dimensional lattice gas model for a symmetric $\Sigma 5$ tilt grain boundary and found that the boundary starts to be disordered upon heating to about $0.5T_m$. The degree of disorderness increases continuously with the increase of temperature until melting takes place at T_m. Ciccotti et al.[12] employed molecular dynamics and Lennard-Jones potential to simulate a symmetric $\Sigma 5$ tilt grain boundary and obtained results which are similar to those of Kikuchi and Cahn qualitatively. The difference between boundary structure and liquid structure disappears only at T_m[13]. Broughton and Gilmer[14] also found that GB melting can not occur under any specific temperature lower than T_m. Nguyen and Yip[15] found that the disordering can occur in the interfacial region at about 0.8 to $0.9T_m$.

On the basis of our anelastic experimental results[5−10], the present article attempts to clarify the above-metioned dispute through the molecular dynamics simulations with Morse interaction potential. In order to compare with our experimental results, the simulation systems are chosen as [110] symmetrical tilt grain boundaries of aluminium with mis-orientation angles 70.5°($\Sigma 3$), 109.5°($\Sigma 3$), 50.5°($\Sigma 11$) and 109.5°($\Sigma 11$) whose experimental results are reported in Refs.[7−10]. From the GB structures at different temperatures, it can be found whether the GB structures start to be disordered at the temperature T_0 which is much lower than T_m. The difference between two angles with a same Σ number is also investigated and compared with the anelastic measurement results.

II. PRINCIPLE AND PROCEDURE OF SIMULATIONS

2.1 Simulation Principles

The dynamic equations of N atoms with mass m are

$$F_i = m\frac{\mathrm{d}^2 r_i}{\mathrm{d}t^2} \qquad\qquad i = 1, 2, ..., N \qquad\qquad (1)$$

In Eq.(1), the r_i and F_i are the coordinate vector of i-th atom and the force exerted on it. F_i can be derived by the atomic interaction potential $U(r_{ij})$

$$F_i = -\sum_{j \neq i}^{N} \frac{\partial U(r_{ij})}{\partial r_{ij}} r_{ij}^0 \qquad\qquad i = 1, 2, ..., N \qquad\qquad (2)$$

r_{ij} and r_{ij}^0 are the distance and the unit displacement vector of i-th atom relative to j-th atom, respectively. The initial velocities of the atoms are given with a Maxwellian distribution corresponding to the given temperature. Equation (1) can be solved by using central difference method.

Morse potential satisfies the conditions based on Born's analysis of the crystal elasticity and can be used to explain the general behaviour of solid cohesion and chemisorption. It has also been used in many fields of solid state physics. The valence electronic structure of aluminium is $3s^2 3p^1$ and there is only one electron on its outer shell porbital. So, it can be approximately considered to have spherical symmetry. That is to say, the Morse pootential is a good aproximation for the atomic interaction potential of aluminium. Therefore, we use it in this work. The formula of Morse potential is

$$U(r_{ij}) = D_0\{\exp[-2\alpha(r_{ij} - r_0)] - 2\exp[-\alpha(r_{ij} - r_0)]\} \qquad\qquad (3)$$

For aluminium, the parameters D_0, r_0 and α in above equation are taken from Refs.[16–17], where, D_0=0.12 eV, r_0=2.86×10^{-10} m and α=2.35×10^{10} m^{-1}.

2.2 Simulation Models

According to coincidence site lattice (CSL) model[18], the GB structures in the simulations are chosen as Σ=11 and Σ=3 tilt boundaries in FCC aluminium bicrystals. The $\Sigma11$ boundaries are generated by a rotation of 50.5° and 129.5° about an axis along [110] and

(a) initial   (b) translated   (c) T=250K   (d) T=350K
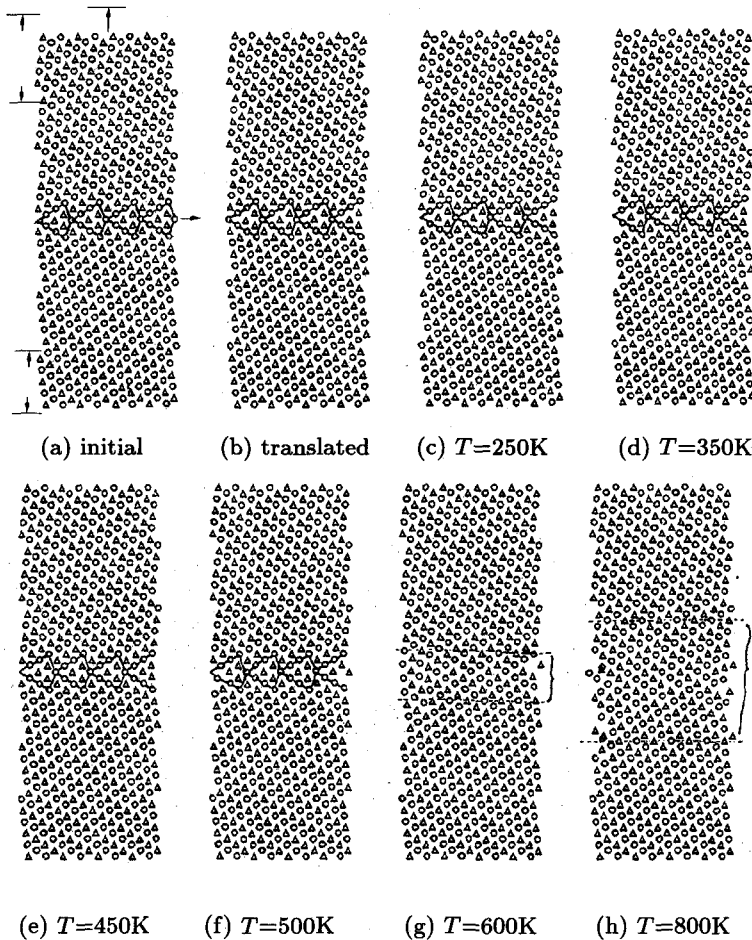
(e) T=450K   (f) T=500K   (g) T=600K   (h) T=800K

Fig.1   The initial, translated and relaxed structures at different temperatures for θ=50.5°($\Sigma11$)

shown in Figs.1(a) and 2(a), respectively. Similarly, the $\Sigma3$ boundaries are generated by a rotation of 70.5° and 109.5° about the same axis and shown in Figs.3(a) and 4(a), respectively. In these figures, Z axis is along [110], which is perpendicular to the paper, X axis is in the GB plane, and then, the Y axis is the normal line of GB plane. In the X and Z directions, periodic border conditions are taken into account according to their natural periods. In the X direction, 4 CSL periods are considered for 50.5°, 129.5° and 70.5°, but, 8 CSL periods are considered for 109.5°. In the Z direction, 4 atomic layers are taken. However, in the Y direction, a movable border condition is used. By this condition we mean

that, from the main block two Y-border regions are divided out. In these two regions, atoms do not move according to Eq.(1), but move with motions of the neighbor atoms in the rest of the block. The total numbers of atom for the four systems are 1040,1464,760 and 1056, respectively.

In the simulations, the unit of length is taken to be the cystal lattice constant, the unit of time is $r_0(m/D_0)^{1/2}= 4.26\times10^{-13}$ s, and the time step in the central difference method is taken as 1.7×10^{-14} s, which is approximately $1/25$ of the time unit.

In order to improve the efficiency of simulations, a two-stage procedure is used. Firstly, give the two crystals a relative solid translation and make the system to have a relative stable structure which is in a state of the lowest energy. The convergence criterion is that the variation of total energy of the system is smaller than 10^{-6}. Secondly, for each system, the atomic relaxation is carried out, i.e., the atoms move according to Eq.(1). It means, positions and velocities of the atoms are obtained by solving Eq.(1) step by step, in which the structure after translation is taken as the initial atomic configuration and the initial velocities of atoms are given with a Maxwellian distribution corresponding to the given temperature. The potential energy and kinetic energy can be obtained also. From these, the dynamic behavior of the system can be described completely. When the system is in equillibrium, the average total force on each particle is near zero, and each particle vibrates around its equillibrium position. From our simulation, it is seen that when the number of time step is over 3000, the average positions of the particles do not change any more. Therefore, the number of time step is taken as 4000 in our computation. In order to keep the temperature constant, the atomic velocities are adjusted corresponding to the given temperature after each 25 time steps.

III. RESULTS AND DISCUSSIONS

3.1 The Results from the Solid Translation

The structures after solid translation for the four systems are shown in Figs.1(b) to 4(b) and the values of the translation in three directions are summarized in Table 1.

Table 1
The number of atoms and the solid translations for each system

angle (°)	system	border	total number	X-translation	Y-translation	Z-translation
50.5	624	416	1040	-1.93×10^{-3}	-5.77×10^{-2}	-2.98×10^{-9}
129.5	904	560	1064	-0.29591	-0.39565	-3.92×10^{-10}
70.5	472	288	760	-0.28868	-0.10349	-5.70×10^{-10}
109.5	672	384	1056	-9.10×10^{-10}	-4.98×10^{-3}	-3.12×10^{-10}

From Table 1, it càn be found that the displacements in Z direction are small for all of the four systems because of the lattice periodicity in this direction. But, in the X and Y directions, the displacements are much larger and play important role in reducing the energy of the systems. Therefore, the extent of the difference between real structure and CSL structure can be shown from the value of the displacements. Smaller displacements mean that the system is nearer to the ideal CSL structure and has a larger stability. So, the structure of $109.5°$ is more stable than that of $70.5°$, and the structure of $50.5°$ is more stable than that of $129.5°$. From Table 1, it can be found that stabilities of the four systems are in the order: $109.5° \gg 50.5° > 70.5° > 129.5°$.

3.2 The Results from Atomic Relaxation

The atomic relaxed structures for the four systems at various temperatures, which belong to two Σ's with 4 angles, are shown in Figs.1 to 4.

Comparing Figs.1(c) to 1(h) with Fig.1(b), it can be found that between 250K to 450K the GB of $50.5°(\Sigma 11)$ is nearly the same as its translated structure. When temperature increases to 500K (approximately $0.5T_m$), the quadrilateral structure in GB has a little distortion. When temperature is higher than 600K, there is obvious change in GB region and the ordered structure of GB has large distortions. Some parts of the GB start to be disordered.

Comparing Figs.2(c) to 2(f) with Fig.2(b), it can be found that below 250K(Fig.2(c)) the GB of $129.5°(\Sigma 11)$ is nearly the same as its translated structure (Fig.2(b)). When temperature increases to 300K (approximately $0.32T_m$), the quadrilateral structure in GB has a little distortion. When temperature increases to 350K (Fig.2(e)), the ordered structure of
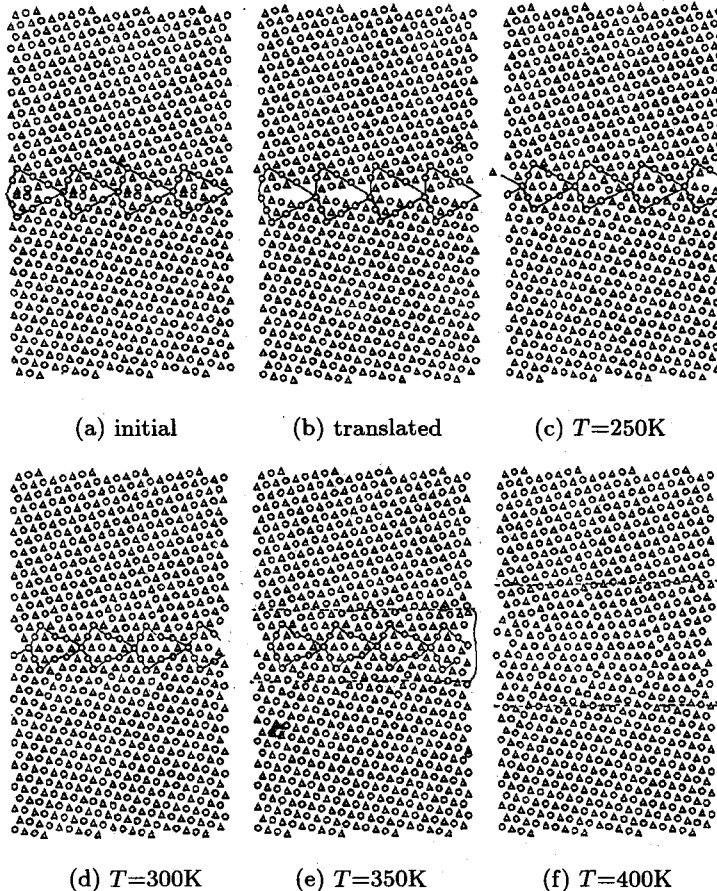

(a) initial   (b) translated   (c) $T=250K$

(d) $T=300K$ (e) $T=350K$ (f) $T=400K$

Fig.2   The initial, translated and relaxed structures at different temperatures for $\theta=129.5°(\Sigma 11)$.

GB has large distortion and starts to be disordered. When temperature is higher, the CSL structure of GB is damaged and an extended region in GB becomes disordering. All ordered structures of GB tend to be disordered with the temperature increase.
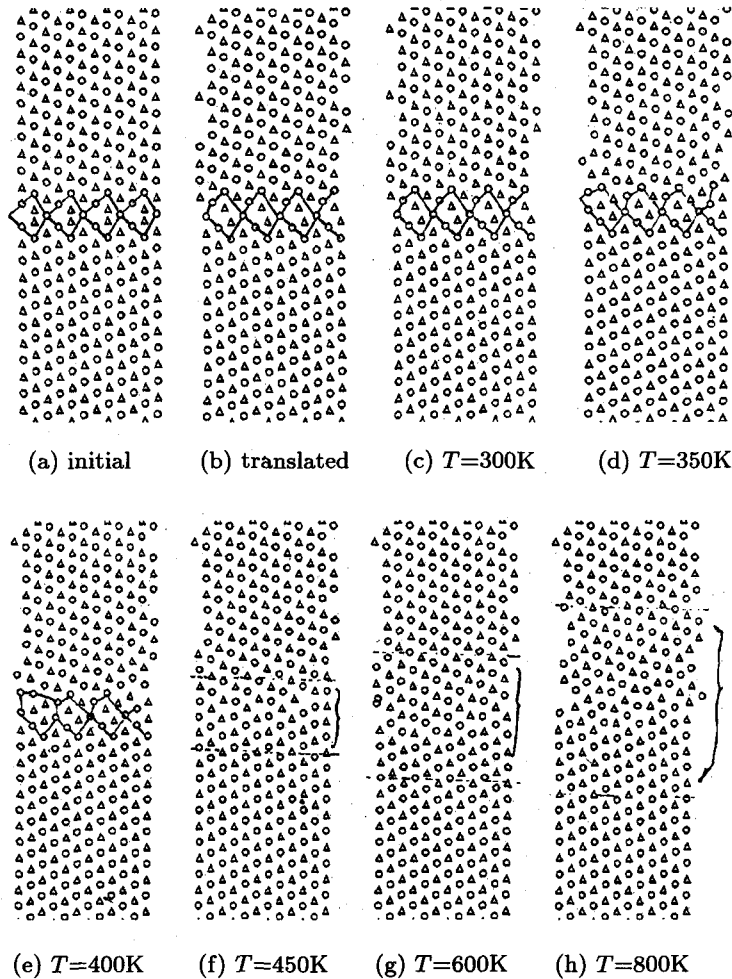
(a) initial   (b) translated   (c) $T=300$K   (d) $T=350$K

(e) $T=400$K   (f) $T=450$K   (g) $T=600$K   (h) $T=800$K

Fig.3   The initial, translated and relaxed structures at different temperatures for $\theta=70.5°(\Sigma3)$

In the same way, comparing Figs.3(c) to 3(h) with Fig.3(b), it can be found that below 300K (Fig.3(c)) the GB of 70.5°($\Sigma3$) is nearly the same as its translated structure (Fig.3(b)). When the temperature increases to 350K (approximately $0.38T_m$), the quadrilateral structure in GB has a little distortion. When temperature rises up to 400K (Figs.3(e)), there is an obvious change in GB region and some parts of the GB start to be disordered. When the temperature is higher, the structure of CSL is damaged and the disordered region around GB becomes wider. With further increase of temperature (up to 800K), this disordered region extends from GB to the single crystal regions.

Comparing Fig.4(c) with Fig.4(b), their structures are in the same without any difference even when temperature increases to 900K. In other words, there is no transformation temperature from order to disorder in this twin-crystal GB. The $\Sigma3(109.5°)$ bicrystal GB has special characteristics. Actually, its GB structure is a twin-lattice one. All atoms in GB are at the co-lattice points. The coordinate number in GB is the same as in grains and there is very little different coordinate distances between them. Therefore, the structure of

109.5°(Σ3) is the most stable structure among the four systems. It can be predicted that the transformation temperature in this GB is quite near to its melting point. Perhaps, when the temperature increases to T_m, the GB will melt into liquid together with the crystal regions.
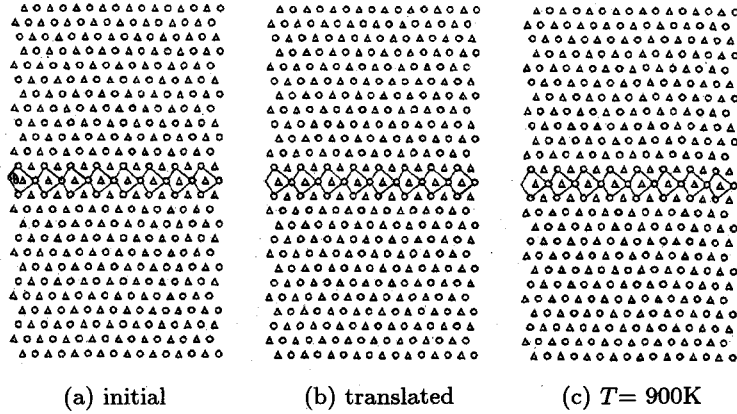

(a) initial   (b) translated   (c) $T = 900K$

Fig.4   The initial, translated and relaxed structures at different tempera-
        tures for $\theta = 109.5°(\Sigma 3)$

Comparing Fig.1 with Fig.2, it can be found that the transformation temperature from order to disorder in Σ11 GB with angle 129.5°$(0.32T_m)$ is much lower than that with angle 50.5°$(0.5T_m)$. Although the Σ number and rotation axis are the same in these two systems, but, the CSL period of GB with 129.5° is much longer than that with 50.5° and the atomic arrangement and the coordinate number in their GB zones are quite different from each other. The former one has higher GB energy and lower stability than the Σ11 with 50.5° does. So, it has a lower transformation temperature from order to disorder.

In order to compare with the experimental results of anelastic measurements, The simulation results are summarized in Table 2. The anelastic measurement results of T_0 are also listed in Table 2.

Table 2

Temperatures of GB disordering and the anelastic measurement results

Angle (°)	Σ	T_0 of GB disordering	Experimental results
50.5	11,	$0.50T_m$	$0.42T_m$ [9]
129.5	11	$0.32T_m$	$0.37T_m$ [10]
70.5	3	$0.38T_m$	$0.40T_m$ [9]
109.5	3	$\sim T_m$	——

From Table 2, it can be seen that the transformation temperatures of GB from order to disorder are in agreement with the sudden change temperature T_0 of the anelastic physical quantities qualitatively. That is to say, the temperature T_0 is related with the disordering in GB region, which depends upon the angles and the Σ numbers of GB. Therefore, GB plays an important role in bicrystal anelastic relaxation.

IV. CONCLUSIONS

1. MD simulation shows that the transformation temperatures from order to disorder are $0.50T_m$, $0.32T_m$ and $0.38T_m$ in the GB's 50.5°(Σ11), 129.5°(Σ11) and 70.5°(Σ3), respec-

tively, which are much lower than the melting point T_m. However, in the $109.5°(\Sigma 3)$ GB, such a transformation temperature is not found.

2. The stabilities of the four GB structures are in the order: $109.5° \gg 50.5° > 70.5° > 129.5°$.

3. The transformation temperatures from order to disorder in the GB $50.5°(\Sigma 11)$, $129.5°(\Sigma 11)$ and $70.5°(\Sigma 3)$ are qualitatively in agreement with the sudden change temperature T_0 of the anelastic physical quantities in anelastic relaxation measurements.

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