SOLIDS AND LIQUIDS

Molecular Dynamics Simulation of the Migration of Tilt Grain Boundaries in Ni and Ni₃Al

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Abstract—The migration of the $\langle 100 \rangle$ and $\langle 111 \rangle$ tilt boundaries in Ni and the Ni₃Al intermetallic compound is studied by molecular dynamics simulation. The low-angle $\langle 100 \rangle$ boundaries in Ni and Ni₃Al are shown to migrate much more slowly (by approximately two times at a temperature of 1700 K) that the $\langle 111 \rangle$ boundaries, which is related to different migration mechanisms of the $\langle 100 \rangle$ and $\langle 111 \rangle$ boundaries. The migration of the $\langle 100 \rangle$ boundaries is found to occur by the splitting of paired grain-boundary dislocations followed by a change of partner dislocations, which proceeds via the slip of split dislocations. The $\langle 111 \rangle$ tilt boundaries move according to a combined action of the following two mechanisms: the mechanism described above and the mechanism consisting in joint slip of paired grain-boundary dislocations, which have common slip planes in contrast to the grain-boundary dislocations in $\langle 100 \rangle$ boundaries. Since the joint slip of paired dislocations has a relatively low activation energy, the mobility of the $\langle 111 \rangle$ tilt boundaries is higher than that of the $\langle 100 \rangle$ boundaries. Our molecular dynamics studies demonstrate that the migration rate of analogous boundaries in Ni₃Al is significantly lower than in Ni (by approximately three times at a temperature of 1700 K), which is partly caused by the additional energy consumed for the formation of a disordered region behind a migrating boundary in Ni₃Al. Because of a relatively low boundary mobility in Ni₃Al, the contribution of diffusion atomic displacements during boundary migration is higher as compared to Ni.

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1. INTRODUCTION

The migration of a grain boundary is its motion normal to the surface, and it plays a certain role in recrystallization, in many phase transformations. Although the problem of grain-boundary migration has long been studied, differences of opinions and unresolved problems related to a migration mechanism still exist. Low-angle tilt boundaries are considered to migrate via a combination of slip and climb of grain-boundary dislocations [1]. For example, the authors of [2, 3] concluded that the main mechanism of tilt boundary migration is the climb of grainboundary dislocations. However, the low-angle $\langle 111 \rangle$ tilt boundaries in fcc metals are known to have the highest mobility, and the migration rate of the $\langle 100 \rangle$ tilt boundaries is substantially lower despite the fact that the density of jogs in grain-boundary dislocations in the $\langle 100 \rangle$ boundaries is higher [1, 3–5]; that is, climb in them should be more intense than in the $\langle 111 \rangle$ boundaries. The cause of this radical difference between the mobilities of the $\langle 111 \rangle$ and $\langle 100 \rangle$ boundaries and their migration mechanism are still incompletely clear.

The migration of low-angle boundaries is known to be slower than that of high-angle boundaries [1, 6]. However, different opinions regarding the migration activation energy still exist. For example, the activation energy decreases almost monotonically with increasing misorientation angle in the low-angle boundary range [6, 7]. The results of experiments on the migration of tilt boundaries [3, 8] showed that lowangle boundaries with the same axis of misorientation have almost the same migration activation energy over a wide misorientation angle range, which indirectly points to the same elementary migration mechanism of these boundaries.

The migration of grain boundaries in ordered alloy and intermetallics is also poorly understood. One of the interesting and promising intermetallics is Ni₃Al, which has the L1₂-ordered fcc lattice. Among the row of similar ordered alloys, the Ni₃Al intermetallic compound has unique physical and mechanical properties, namely, a positive temperature dependence of the yield strength and a high thermal stability (order–disorder transition temperature of Ni₃Al is higher than its melting temperature) [9, 10]. Therefore, this interme-

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Fig. 1. Calculation block for simulating the $\langle 111 \rangle$ with disorientation angle $\theta = 30^{\circ}$ tilt boundary. The dark gray atoms at the edge of the calculation block were fixed during a computer experiment (rigid boundary conditions).

tallic compound is used in practice to produce structural superalloys.

The purpose of this work is to study the specific features and mechanism of migration of the $\langle 100 \rangle$ and $\langle 111 \rangle$ tilt boundaries in Ni and Ni₃Al using molecular dynamics simulation.

2. DESCRIPTION OF THE MODEL

For investigation, we used the technique of studying the migration of tilt grain boundaries proposed and developed in [1, 11]. A clearly certified boundary in the form of a loop or arch is created (Fig, 1, black dashed line). The tension of the boundary, which appears due to the tendency of the boundary to minimize its energy (as in the case of surface tension), causes the directional motion of the boundary toward a decrease in its area. The migration-induced force and the boundary migration rate are constant during almost the entire boundary motion and decrease gradually to the end of a computer experiment.

A similar model was used in [12–15] to perform molecular dynamics simulation of the migration of the triple junction of grain boundaries. Such a simulation in [13, 14] was performed with a two-dimensional (2D) model. It should be noted that the 2D and 3D models have radical differences in the mechanisms of grain-boundary migration. In the 2D model, grainboundary edge dislocations have no jogs periodically arranged along dislocation cores; such jogs are known to play an important role in grain-boundary processes, especially diffusion [16]. Therefore, we decided to create a 3D calculation block in a molecular dynamics model in the form of a plate 12 atomic planes thick (see Fig. 1). This thickness is sufficient for the appearance of the effects related to grain-boundary dislocation jogs. In the case of $\langle 111 \rangle$ tilt boundaries, the calculation block of nickel had a height of 18.0 nm, a width of 12.0 nm, and a thickness of 2.4 nm. These sizes for (100) boundaries were 18.2, 12.1, and 2.2 nm, respectively. The calculation blocks of Ni₃Al had slightly larger sizes because of the difference between the lattice parameters of Ni and Ni₃Al, and they contained approximately 50000 atoms. Infinite repetition of a structure was imitated along axis z; that is, periodic boundary conditions were imposed (see Fig. 1). Grain boundaries should be fixed at the edge of a calculation block, which implied the retained orientation of the crystal lattices of two different grains at the block boundary. Therefore, the block boundaries along axes x and y were rigidly fixed to detect the misorientation of the grains (Fig, 1, dark gray).

To describe the interatomic interactions in Ni and Ni₃Al, we used the many-body Cleri–Rosato tightbinding potentials [17]. Potentials of this type were repeatedly used in molecular dynamics models and were tested for a large number of characteristics [18, 19]. The experience of their application demonstrates that they can be used to describe the diverse properties of metals and alloys. The time step of integration in the molecular dynamics method was 2 fs. The temperature was specified through the initial atomic velocities according to the Maxwell–Boltzmann distribution, and the thermal expansion of the calculation blocks was taken into account. The Nose–Hoover thermostat was used to maintain a constant temperature level during simulation.

3. RESULTS AND DISCUSSION

Figure 2 shows the energies of the $\langle 100 \rangle$ and $\langle 111 \rangle$ tilt grain boundaries and their migration rates at a temperature of 1700 K versus the misorientation angle. Special and symmetric boundaries are beyond the scope of this work. The grain-boundary energy was calculated as the ratio of the difference between the energies of a calculation block with a boundary and the same number of atoms in an ideal crystal to the boundary area. Structure relaxation was carried out before energy calculation. It should be noted that the grain-boundary energy includes a certain error because of possible existence of various defects in a grain boundary. Apart from geometrically necessary defects (such as grain-boundary dislocations in lowangle boundaries), boundaries can contain equilibrium and nonequilibrium (excess) defects. This diversity of defects and the curvature of boundaries can introduce an error in determining the energy. In this work, we calculated the energies of three boundaries, the migration of which was studied with the developed model.



Fig. 2. (a) Energies of (\triangle) (100) and (\bigcirc) (111) tilt boundaries and (b) their migration rates at a temperature of 1700 K vs. misorientation angle θ in Ni.

The tension of grain boundaries is proportional to their energy. The energy and the tension increase with the misorientation angle (Fig. 2a). The energies of high-angle boundaries are approximately the same, which is thought to be characteristic of a wide class of high-angle boundaries and mixed-type boundaries; therefore, most angles between the boundaries in triple junctions are close to 120° [20, 21].

The migration rate of grain boundaries was measured at 1700 K. At this temperature (which is close to the melting temperature of nickel), the migration of boundaries with a misorientation angle higher than 10° has the rate that is high enough for it to be measured in a molecular dynamics model. The migration rate was approximately the same during simulation, which allowed us to simply determine it as the ratio of the displacement of the upper part of the boundary to the molecular dynamics simulation time (see Fig. 1).

Misorientation angle θ of boundaries was varied from 10° to 45° for the $\langle 100 \rangle$ boundaries and from 10° to 40° for the $\langle 111 \rangle$ boundaries. In the case of the $\langle 100 \rangle$ boundaries, 45° is the maximum misorientation angle. In the case of $\langle 111 \rangle$ boundaries, we took into account the fact that the $\langle 111 \rangle$ tilt grain boundaries with a misorientation angle of 38° have the maximum mobility [1, 4, 5]. As the misorientation angle increases, the migration rate of the boundaries increases, which is a well-known feature [1, 6].

Note that, at a misorientation angle higher than 25°, high-angle $\langle 100 \rangle$ and $\langle 111 \rangle$ boundaries move at approximately the same velocity (30–37 m/s at 1700 K), and the migration rates of low-angle $\langle 100 \rangle$ and $\langle 111 \rangle$ tilt boundaries differ substantially: the migration rate of the $\langle 100 \rangle$ low-angle boundaries is approximately half that of the $\langle 111 \rangle$ boundaries.

Figure 3 shows the atomic displacements during the migration of $\langle 100 \rangle$ and $\langle 111 \rangle$ boundaries with a misorientation angle of 20°. The displacements are

shown as the segments that connect initial and final atomic positions (only the displacements larger than 0.1 nm are shown). It is clearly visible that the atomic displacements during boundary migration are not chaotic, but their trajectories have a pronounced pattern, namely, a network with square meshes for the $\langle 100 \rangle$ boundaries and a network with hexagonal meshes for the $\langle 111 \rangle$ boundaries. Although the misorientation angle (20°) is rather high and belongs to highangle boundaries (i.e., the boundaries where individual grain-boundary dislocations are not distinguished), the atomic displacements during the migration of low-angle boundaries have the same form and differs only in a small mesh size, which decreases with increasing misorientation angle. An ordered network of atomic displacements can hardly be detected at angles higher than $25^{\circ}-30^{\circ}$.

To understand the mechanism of appearance of such networks during the migration of low-angle tilt boundaries, we consider dislocations in them. Edge grain-boundary dislocations differ from conventional intragranular dislocations. First, they are at least paired dislocations (Fig. 4). The atomic planes of both grains, i.e., the planes belonging to crystal lattices with different orientations, terminate at low-angle boundaries. From an energy standpoint, such terminated atomic half-planes belonging to different grains should form a common defect, which is represented by a grain-boundary dislocation, and some of such dislocations have a relatively large Burgers vector. Second, in contrast to conventional dislocations, grain-boundary dislocations have a high jog density, which depends on the orientation of a boundary plane and the direction of a misorientation axis. In our case, the first circumstance and the fact that such paired dislocations can split are important. For example, disloca-



Fig. 3. Atomic displacements during the migration of tilt boundaries (a) $\langle 100 \rangle$, $\theta = 20^{\circ}$ (within 540 ps) and (b) $\langle 111 \rangle$, $\theta = 20^{\circ}$ (within 300 ps) in Ni at a temperature of 1700 K. Displacements larger than 0.1 nm are shown.

tions in the $\langle 111 \rangle$ boundaries can split according to the reaction [16]

 $1/2[1\overline{1}0] \rightarrow 1/6[2\overline{1}\overline{1}] + 1/6[1\overline{2}1],$

in the $\langle 100 \rangle$ boundaries, they can split as

 $1[0\overline{1}0] \rightarrow 1/2[0\overline{1}1] + 1/2[0\overline{1}\overline{1}].$

When studying the dynamics of the atomic mechanism of migration of low-angle $\langle 100 \rangle$ and $\langle 111 \rangle$ boundaries, we found that grain-boundary paired dislocations split and changed their partner dislocations during boundary motion. As a result, zigzag atomic displacements appeared in, e.g., side boundaries (see Fig. 3). Here, split dislocations slipped and no climb was observed.

Low-angle boundaries moved via splitting and changing partner dislocations. As is seen in Fig. 3a, the atomic displacements according to this mechanism during the migration of low-angle $\langle 100 \rangle$ tilt boundaries form a network with square meshes. In the case of migration of $\langle 111 \rangle$ boundaries, this mechanism is supplemented with the mechanism of joint slip of paired grain-boundary dislocations (Fig. 4b. 2). In contrast to $\langle 100 \rangle$ boundary dislocations, paired $\langle 111 \rangle$ boundary dislocations have common slip planes, along which they can slip at a relatively low activation energy. The following combined action of both mechanisms was detected during the migration of $\langle 111 \rangle$ boundaries: joint slip of paired grain-boundary dislocations and their slip accompanied by a change of partner dislocations. During migration, symmetric sections formed in the grain toward which a boundary moved, and these sections rotated to take the structure of another grain. Therefore, the meshes of the atomic displacement network during the migration of $\langle 111 \rangle$ boundaries were hexagonal.

The migration of boundaries in Ni₃Al was studied similarly. Figure 5a shows the energies of $\langle 100 \rangle$ and $\langle 111 \rangle$ tilt boundaries as functions of the misorientation angle in Ni₃Al. It should be noted that the boundary energies are higher than those in Ni by approximately 1.5 times. This difference is mainly caused by the contribution of the disordering effect, i.e., an increase in the fractions of Ni–Ni bonds and, especially, Al–Al bonds (which have a lower energy as compared to the Ni–Al bond) during grain misorientation.

Figure 5b shows the boundary migration rate in Ni₃Al versus the misorientation angle. Despite a higher boundary energy and, hence, their tension force, the boundary migration rate in Ni₃Al turned out to be substantially lower than that in Ni, by approximately three times at 1700 K. This difference is likely to be caused by the additional energy consumed for the disordering and the rupture of Ni–Al bonds during the boundary motion in Ni₃Al. Figure 6a shows an example of the formation of a disordered region behind a migrating $\langle 111 \rangle$, $\theta = 20^{\circ}$ boundary. An order is not



Fig. 4. Scheme of the migration mechanism of $\langle 100 \rangle$ and $\langle 111 \rangle$ low-angle tilt boundaries in a monoatomic plane: (a) $\langle 100 \rangle$, $\theta = 10^{\circ}$ boundary, change of partner dislocations; (b) $\langle 111 \rangle$, $\theta = 7^{\circ}$ boundary ((1) change of partner dislocations, (2) joint slip of paired dislocations).

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Fig. 5. (a) Energies of $(\triangle) \langle 100 \rangle$ and $(\bullet) \langle 111 \rangle$ tilt boundaries and (b) their migration rates at a temperature of 1700 K vs. misorientation angle θ in Ni₃Al.

restored and ordering is much longer than the boundary migration.

As in the case of boundaries in Ni, the mobility of the low-angle (100) boundaries in Ni₃Al was approximately half that of the $\langle 111 \rangle$ boundaries (Fig. 5b). On the whole, the migration mechanism of low-angle boundaries in Ni₃Al was similar to that in Ni. One of the substantial differences was a higher contribution of diffusion atomic displacements during migration (because of a relatively low boundary migration rate in Ni₃Al). Figure 6b shows an example for the atomic displacements during pipe diffusion (diffusion along grain-boundary dislocation cores) in the course of the migration of a low-angle (100), $\theta = 10^{\circ}$ boundary. Typical atomic displacements near grain-boundary dislocations, the cores of which are perpendicular to the figure plane, are visible (radius of trajectories of these atomic displacements indicates the oscillation of dislocation cores).

4. CONCLUSIONS

Using molecular dynamics simulation, we studied the features and mechanism of migration of the $\langle 100 \rangle$ and $\langle 111 \rangle$ tilt boundaries in Ni and the Ni₃Al intermetallic compound and found the boundary energy and the boundary migration energy at a temperature of 1700 K as functions of the misorientation angle. The high-angle $\langle 100 \rangle$ and $\langle 111 \rangle$ boundaries were shown to move at approximately the same velocity at this temperature, and the migration rate of the low-angle $\langle 100 \rangle$ boundaries is approximately half that of the $\langle 111 \rangle$ boundaries. This difference was found to be caused by the difference between the migration mechanisms of the $\langle 100 \rangle$ and $\langle 111 \rangle$ boundaries. The $\langle 100 \rangle$ boundaries move via splitting paired grain-boundary dislocations followed by a change of partner dislocations. The change of partner dislocations was accompanied by the slip of split dislocations and no climb was detected. During the migration of low-angle $\langle 111 \rangle$ tilt boundaries, we observed a combined action of the following two mechanisms: the mechanism described above and the mechanism consisting in joint slip of paired grainboundary dislocations, which have common slip planes in contrast to the grain-boundary dislocations in $\langle 100 \rangle$ boundaries. Since the second mechanism has a relatively low activation energy, the $\langle 111 \rangle$ boundaries are much more mobile that the $\langle 100 \rangle$ boundaries.

Our molecular dynamics studies showed that the migration rate of analogous boundaries in the Ni₃Al intermetallic compound is significantly lower than that in Ni (by a factor of 3 at 1700 K), which is partly caused by the formation of a disordered region behind a migrating boundary in Ni₃Al. On the whole, the migration mechanism of low-angle boundaries in Ni₃Al is similar to that in Ni. One of the substantial



Fig. 6. Grain boundary migration in Ni₃Al: (a) formation of a disordered region behind a moving boundary ((1) initial boundary position, (2) current position) during the migration of the $\langle 111 \rangle$, $\theta = 20^{\circ}$ tilt boundary at a temperature of 1700 K within 1000 ps; (b) pipe self-diffusion along grain-boundary dislocation cores during the migration of the $\langle 100 \rangle$, $\theta = 10^{\circ}$ boundary within 4000 ps.

differences consisted in a higher contribution of diffusion atomic displacements during migration because of a relatively low boundary migration rate in Ni₃Al.

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