Influence of Grain Boundary on Melting *

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The temperature behaviour of an Al bicrystal with surfaces consisting of (110) and (111) crystals is simulated using molecular dynamics. The result shows that the (110) crystal losses its crystalline order at 820 K, whereas the disorder does not propagate through the (111) crystal at this temperature. Instead, some disordered atoms are recrystallized into the (111) crystal and the initial grain boundary changes into a stable order–disorder interface. Thus, it was discovered that at a temperature near its melting point, the (111) crystal grew and obstructed the propagation of disorder. Such an obstruction is helpful for understanding melting.

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Understanding melting on a microscopic scale is a fundamental problem of condensed physics, which can be traced to Lindemann.[1] Some progress has been made in recent years. Now it is believed[2] that defects such as surfaces, grain boundaries (GB) and voids are the main sites initiating heterogeneous melting. For melting initiated by surfaces, it was demonstrated[2−5] that for fcc metals, the (110) surfaces premelt, the (100) surfaces seldom premelt and the (111) surfaces almost do not premelt. It is also known that different grain boundaries premelt at different temperatures.[6−9] That is to say, the structure is an important determinant to their melting process. Since in multicrystals, surfaces and grain boundaries coexist, it is important to know how they interact for both theoretical and practical reasons. The study of the interaction, as far as we know, has not been reported. In this letter, the interaction between surfaces and grain boundaries is investigated using molecular dynamics and the embedded-atom model potential.

Using the Voronoi method,[10] we constructed a bicrystal sample with a structure similar to those observed experimentally. The difference between the Voronoi bicrystal and a coincidence site lattice (CSL) bicrystal[11] is that in the Voronoi bicrystal, the crystalline orientations can be specified arbitrarily, which makes it apt to study the interaction between surfaces and grain boundary. This is the first time the Voronoi method has been used to construct a bicrystal. The crystallographic orientations in the Z-axis selected are [111] and [110] respectively, and thus the constructed sample is shown in Fig. 1. The lower crystal ($Z < 40$Å) of the sample has 18 (111) layers and the upper crystal ($Z > 40$Å) has 18 (110) layers perpendicular to the Z-axis. For clarity, the lower crystal is specified as the (111) crystal, while the upper one is specified as the (110) crystal. Period boundary conditions are applied only on the $X − Y$ plane and free boundary conditions are applied to the Z-axis, which

creates double-free surfaces perpendicular to the Z-axis. Thus a bicrystal with both (110) and (111) surfaces is constructed. With a time step of 2.11×10^{-15} s, the system is equilibrated at various temperatures using the velocity Verlet algorithm[12] and Nose–Hoover temperature control.[13] Forces among atoms are computed through the modified Johnson potential[14] for aluminium. When the temperature is changed, distances between atoms and the X and Y dimensions of the sample are adjusted according to the coefficient of thermal expansion determined by the independent bulk simulation at 1 atm.
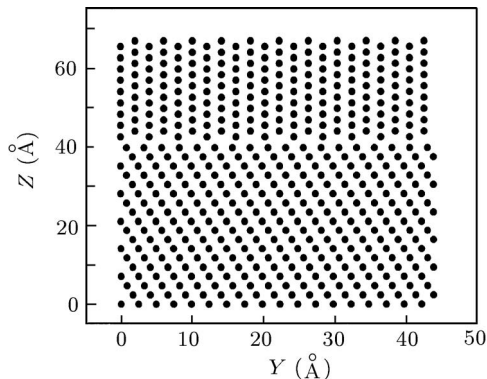

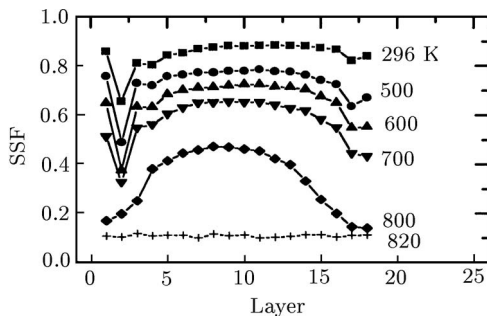Fig. 1. Initial bicrystal projected on the $Y − Z$ plane.

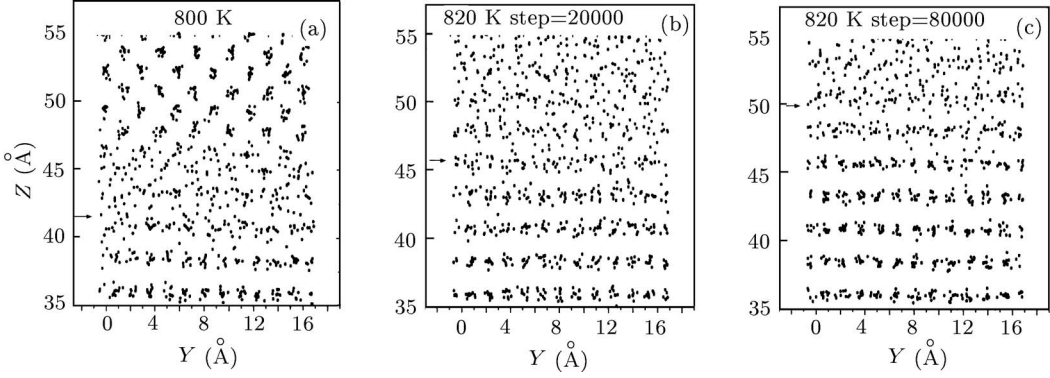
Fig. 2. Static structure factor of the (110) crystal.

Fig. 3. Part of the sample at 800 K (a) and 820 K after 20 000 steps (b) and 80 000 steps (c). The arrow points to the order−disorder interface.
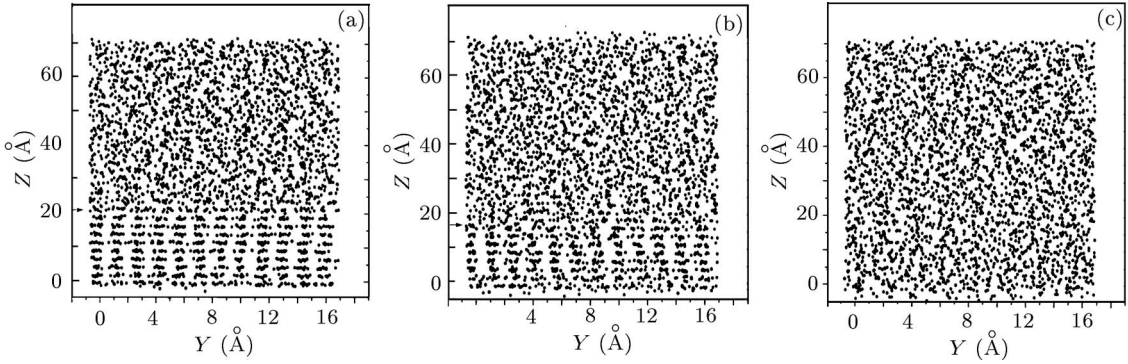

Fig. 4. Sample at 860 K: (a) at the 30 000th step; (b) at the 40 000th step; (c) at the 50 000th step. The arrows point to the order−disorder interface.

In order to characterize the structural change of the sample with temperature, static structure factors (SSF)[15] are also computed layer-by-layer at each temperature,

$$S(k) = \frac{1}{N_l} \left[\left(\sum_l \cos\left(\boldsymbol{k} \cdot \boldsymbol{q}_i\right) \right)^2 + \left(\sum_l \sin\left(\boldsymbol{k} \cdot \boldsymbol{q}_i\right) \right)^2 \right]^{1/2}$$

where N_l is the total number of atoms in the lth layer, $\boldsymbol{k}$ is the reciprocal lattice vector, i is the atomic number in the lth plane, and q_i are the coordinates of atom i. In the bicrystal, the reciprocal lattice vector in the (110) crystal is not the reciprocal lattice vector in the (111) crystal. Thus, two different wave vectors are required. We shall call these wave vectors $\boldsymbol{k}_1$ and $\boldsymbol{k}_2$, and the corresponding SSFs are $S(\boldsymbol{k}_1)$ and $S(\boldsymbol{k}_2)$. From our previous work,[6] we know that when SSF is smaller than 0.2, the corresponding layer lacks crystalline structure, while when it is larger than 0.5, the layer has definite crystalline structure.

Figure 2 shows $S(\boldsymbol{k}_1)$ of the (110) crystal at various temperatures. In Fig. 2, the first layer is the layer of the (110) crystal at the grain boundary, and the 18th layer is the (110) surface. It is shown that at temperatures below 600 K, the SSF of the 17th layer is lower than that of the surface layer, which is due to the "soft channel"[16] for thermal motion in the [110] direction. Attention should be paid to the second layer, whose SSF is lower than that of the 17th layer. This shows that the grain boundary makes the "soft channel" effect more evident. The mechanism for this is under further investigation. With the temperature increasing, the increased thermal motion makes the overall SSFs decrease. The SSFs of the surface and GB are lower than those of the internal layers, which is due to premelting. When the temperature increases to 800 K, the SSFs of the surface and GB are smaller than 0.2, which means their premelting is complete. A snapshot at this temperature [Fig. 3(a)] shows the premelting of the grain boundary. Then after 20 000 steps at 820 K, the (110) crystal lost its crystalline structure and premelted. Note that a single crystal with the (110) surfaces melts at 840 K, so the grain boundary makes (110) crystal melt at a relatively lower temperature.

Figure 3 shows the snapshots of the sample near the GB before and after the (110) crystal was completely premelted. Figure 3(a) indicates that besides the (110) crystal, in the (111) crystal, the atoms near the GB also disordered at 800 K, which is substantially

lower than the premelting temperature (900 K) of its (111) surface. Figure 3(b) shows that at 820 K, the (110) crystal disappeared before the 20 000th step and the initial GB became order−disorder interface. With some disordered atoms recrystallized into the (111) crystal, the order−disorder interface moved upward [as indicated by the arrow in Figs. 3(a) and 3(b)]. Thus the disorder did not propagate downward and its propagation was obstructed. After 80 000 steps, as the order−disorder interface held its position [as shown in Fig. 3(c)], the system with both solid and liquid kept balance.
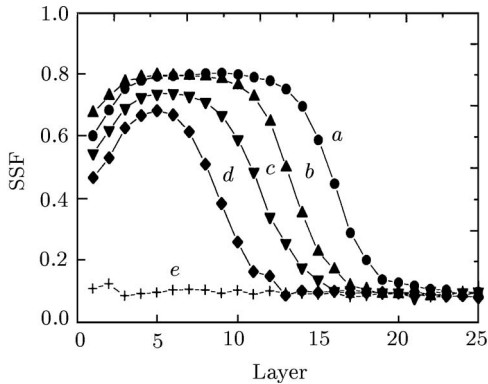

Fig. 5. Static structure factor of the sample at 860 K. The number represents the steps (×10 000) simulated.

Simulations show that when the sample was heated to 840 K, compared to Fig. 3(c), the sample changed slightly. When it was heated to 860 K, the order−disorder interface moved downward step-by-step. Figure 4 shows the sample at 860 K after 30 000 steps. It shows that the (111) surface did not change much until it melted. However, the disorder propagated from the top to the bottom (as shown in Fig. 4, the movement of the arrow) step-by-step and made the system melt completely. Figure 5 demonstrates quantitatively how the (111) crystal melted at a stable speed. Independent simulation shows that a crystal with only the (111) surface cannot initiate its melting at this temperature. Thus the melting of the (111) crystal in the bicrystal is completely determined by the propagation of disorder. If the initiating

procedure can be suppressed, a crystal can hold its crystalline structure.

In conclusion, the melting of the bicrystal does not proceed by the simultaneous melting of the two crystals. Instead, the (110) crystal premelted completely at 820 K and left the (111) crystal alone. The initial grain boundary changed into the order−disorder interface and subsequently the (111) crystal grew along the interface, thus obstructing the propagation of disorder and preventing the system from melting at this temperature. At 860 K, the interface could not suppress the propagation of disorder and the disorder initiated the melting of the (111) crystal. The grain boundary thus made the bicrystal melt at a temperature between the premelting temperature of the (110) crystal and that of the (111) crystal. We believe that a similar mechanism plays a role in the melting of multicrystals.

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