Atomistic simulation of the fcc–hcp transition in single-crystal Al under uniaxial loading

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\textbf{Abstract.} The dynamic behavior of the single-crystal Al under [001] uniaxial strain is simulated by classic molecular dynamics. The fcc–hcp structural transition is successfully observed when the loading pressure reaches about 90 GPa, and the reverse transition is also found with hysteresis. The mechanism and morphology evolution of both the forward and backward transitions are analyzed in detail. It is found in the process of the structural transition that the (010)\textsubscript{fcc} or (100)\textsubscript{fcc} planes transit into (0001)\textsubscript{hcp} planes, and the twins of the hcp phase along the (112)-plane appear, whose boundaries finally become along the (110)-plane. Besides, we find the twinning (along the (110)\textsubscript{fcc} planes) in the hcp phase prior to the back transition (hcp–fcc). Our simulations show the coexistence of fcc and hcp phases over a wide range of pressures, and finally, the phase transition is evaluated by using the radial distribution functions.
1. Introduction

The behavior of aluminum (which is one of the most important metals) under high pressure has been the subject of many theoretical and experimental studies. On the theoretical side, the fcc $\rightarrow$ hcp phase transition of Al has consistently been predicted with increasing pressure $[1]$–$[6]$. However, different techniques yield different transition pressures. For example, Moriarty and McMahan $[1]$ studied phase stability in Al by two methods: the generalized pseudo potential technique (GPT) that predicted the fcc $\rightarrow$ hcp phase transition at 360 GPa and the all-electron linear muffin-tin-orbital method (LMTO) that predicted the transition pressure at 120 GPa. In the meantime, the critical pressure was reported from 175 to 220 GPa by some other first-principles total-energy calculation techniques $[2]$–$[4]$. On the experimental side, Greene et al $[7]$ performed a high-pressure experiment in fcc Al by using a diamond-anvil-cell, where they did not find any structural transformation up to 219 GPa. But from measurements up to $P = 330$ GPa of the static equation, the fcc $\rightarrow$ hcp transition of Al was found by Akahama et al at 217 ± 10 GPa in 2006 $[8]$. This experiment also reported the coexistence of fcc and hcp phases over a wide range of pressures in the transition. Obviously, it is very interesting to know how the phase transition takes place.

In recent years, molecular dynamics (MD) simulations have been widely used in the micromechanism study for many dynamic phenomena. Owing to the rapid development of computer capability and the great predominance to detect atomic-scale details, the phase transition, twinning, dislocation emitting and so on are all reproduced successfully by computer $[10]$–$[13]$. Also, many MD simulations were applied to the research on the deformation of Al. For example, Yamokov et al $[11, 12]$ simulated the dislocation process in the deformation of nanocrystalline Al successfully. And the occurrence of deformation twinning in Al with a sufficient small grain size has been suggested by recent MD simulations $[13]$. Thus, the structural transition in Al may be simulated by MD simulation, which has not been reported.

In the present work, we perform the MD simulation of the behavior of single-crystal Al by uniaxial strain, finding the fcc $\rightarrow$ hcp phase transition and its back process. The simulations show that the phase transition takes place at about 90 GPa, corresponding to the reduced volume of 0.68 $V_0$, and the reverse transformation at about 71 GPa. On the other hand, we show the formation process of the hcp twin structures whose boundaries are studied along the (110)-plane in detail. The radial distribution functions (RDFs) are evaluated and used to discuss the forward and backward transitions. This paper is organized as follows. In section 2, we briefly explain the
method used in our simulation. In section 3, we give the results and discussion, including the pressure and temperature effects, the phase transformation mechanism and the micro-process of the nucleation and growth of a new phase; finally, the transitions are evaluated and discussed by RDFs.

2. Method and simulations

It is well known that the potential between the atoms plays an important role in MD simulations. Here, we adopt the embedded-atom method (EAM) according to Yamakov et al [12] to describe the interatomic force, which has already been widely used for deformation research of the crystal Al [11, 12]. We construct a single-crystal Al using crystallography. The sample consists of 256 000 atoms, with lengths $L_x = 80a$, $L_y = 40a$ and $L_z = 80a$, at a temperature of 300 K; the $L_x$, $L_y$ and $L_z$ axes present the [100]-, [010]- and [001]-directions, respectively; and periodic boundary conditions are employed in all three directions. Before loading, the speed calibration method is used to control the original temperature of the sample at 300 K. The pressure was manipulated at zero through modulating the lattice constant, and in this work, the lattice constant of the sample is $a = 0.4053$ nm. Loading style is adiabatic high-strain-rate loading along the [001] crystal orientation, i.e. shorten the [001]-direction lattice constant in every step. The loading/unloading rate in this work is described as the loading/unloading strain divided by the loading/unloading time. We use the Verlet algorithm [15] to integrate the equation of motion where the time step is 0.001 ps and the total simulation time is up to 100 ps. The simulation of the corresponding unloading process is done by extending the [001]-direction lattice constant to $L_z$ at the same rate of straining as loading.

It should be pointed that the EAM potential we used is indeed a little higher than experiments for the extreme pressure (see figure 1). In this work, we focus on how the transition takes place, and obtain some informative results. Of course, this potential can only give a qualitative result, for the transition pressure is distinctly lower than experiments. In addition, the effects of simulated size and boundary conditions are not discussed in the present work. We believe the transition process is not changed due to the limitation of size and boundary conditions.

3. Results and discussion

3.1. Pressure and temperature effects

The pressure changes both in the loading process and in the unloading process are shown in figure 1 by black and green lines, respectively. There, $\varepsilon = (V_0 - V)/V_0$ and $V_0$ denotes the initial volume of the sample. Comparing our simulation data of $P-\varepsilon$ with some data of the experiment and theoretical calculation, we found that the EAM potential we used can well describe the compressing process of Al. Figure 2 shows the change in temperature with strain $\varepsilon$. It is clear that the relationships between $P$ and $\varepsilon$ and between $T$ and $\varepsilon$ are near-linear dependent at the beginning stage of the loading process; when the strain $\varepsilon$ approaches 0.32, the near-linear dependence will be interrupted; the mutation points appear in the curves of $P-\varepsilon$ and $T-\varepsilon$, as shown in figures 1 and 2. In subsequent analysis, we can clearly see that the mutation points ($P_{s1}$, $T_{s1}$) are induced only by the fcc $\rightarrow$ hcp phase transition, not by plastic deformation based on the dislocation emitting. Temperature experiences an abrupt enhancement when the strain $\varepsilon$. 

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Figure 1. Pressure–strain ($\varepsilon$) of Al in experiments, theories and simulations. The yellow line corresponds to a form fitted by Greene et al [7] to their experimental values. The blue line corresponds to the experimental data by Akahama et al [8]. The red line corresponds to the calculated EOS using the empirical potential functions proposed by Cleri and Rosato [16]. The black line and the green line correspond to the loading and unloading processes from our simulations. Points $P_{s1}$ (0.32, 90 GPa) and $P_{s3}$ (0.26, 71 GPa) indicate the beginning of phase transition in the loading and unloading processes, respectively.

Figure 2. Temperature as a function of strain $\varepsilon$ in loading and unloading processes. Points $T_{s1}$ (0.32, 445 K) in the loading process and $T_{s3}$ (0.26, 600 K) in the unloading process indicate the beginning of forward and backward phase transitions, respectively.

exceeds 0.32, for the release of the hidden calorific of the phase transition. The critical pressure of the fcc $\rightarrow$ hcp transition of Al in the high loading rate is about 90 GPa (labeled as $P_{s1}$), which is lower than the results of first-principle total-energy calculation.

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Fig. 3. hcp mass fraction $\lambda_1$ in the loading process and fcc mass fraction $\lambda_2$ in the unloading process as a function of strain $\varepsilon$. The value of $\lambda_1$ increases to nearly 0.8 at the end of the phase transition. In the unloading process, the nucleation of the fcc phase occurs at $\varepsilon = 0.26$. The strains $\varepsilon$ for different points in the unloading process are (a) $\varepsilon = 0.38$, (b) $\varepsilon = 0.34$, (c) $\varepsilon = 0.28$, (d) $\varepsilon = 0.26$, (e) $\varepsilon = 0.24$ and (f) $\varepsilon = 0$.

Fig. 4. Snapshots of the sample loaded with different strains in the [001]-direction: (a) $\varepsilon = 0.08$, (b) $\varepsilon = 0.32$, (c) $\varepsilon = 0.328$, (d) $\varepsilon = 0.36$, (e) $\varepsilon = 0.38$ and (f) $\varepsilon = 0.4$. The fcc phase, grain boundaries and hcp phase are shown by red atoms, purple atoms and green atoms, respectively, and the blue atoms present the atoms that are under the middle state in the phase transition process. The sample transforms into hcp structure when the strain overstep is 0.32; the hcp twins along the (110)-plane are found at last.
Figure 5. Cross sections of the sample corresponding to figure 4. Twins are the primary structure in the sample at the end of the loading process; twin boundaries (purple atoms) and fcc stacking faults (red atoms) exist in the sample with other types of structure. The color criteria are the same as in figure 4.

In the unloading process, there are two mutation points in the $P-\varepsilon$ curve labeled as $P_s^2$ and $P_s^3$ in green line and there are two mutation points in the $T-\varepsilon$ curve labeled as $T_s^2$ and $T_s^3$, as shown in figures 1 and 2, respectively, which are different from the loading process. In the following analysis, we will see that the reversal phase transition occurs at about 71 GPa, corresponding to a temperature of about 600 K in the unloading process. We found that the mutation points $P_s^2$ in the $P-\varepsilon$ curve and $T_s^2$ in the $T-\varepsilon$ curve are not caused by the phase transition. This result can be explained by analysis of the mechanism of phase transition in section 3.2.

3.2. hcp nucleation and growth

The structural change of the sample can be judged from the pair analysis [19], which can distinguish the fcc phase, hcp phase and grain boundaries. Figure 3 displays the hcp mass fraction $\lambda_1-\varepsilon$ in the loading process and the fcc mass fraction $\lambda_2-\varepsilon$ in the unloading process.

In the loading process, as shown by figure 3, there is nearly no hcp phase in the sample when the strain $\varepsilon$ is below 0.32; the hcp mass fraction $\lambda_1$ begins to increase at strain $\varepsilon = 0.32$ and abruptly reaches 0.8 at strain $\varepsilon = 0.36$; then it maintains almost this value until the end of the loading process. In order to find how the hcp phase nucleates and grows, we perform the microstructure of the sample under different strains $\varepsilon$ in the loading process. Figure 4 shows microscopic views of the sample under different strains, and the corresponding cross-sections ((001)-plane) of the sample in figure 4 are shown in figure 5, where the fcc phase, grain boundaries and hcp phase are shown by red, purple and yellow atoms, respectively, and the blue
Figure 6. Snapshots of the atoms in the (010) plane (panels (a)–(c)) and the (100) plane (panels (d)–(f)), which come from the MD simulations; the strains in the snapshots are (a) 0, (b) 0.32, (c) 0.40, (d) 0, (e) 0.32 and (f) 0.40. The changes of location of atoms from 1 to 6 show that the (010) plane and the (100) plane in the fcc phase transit to the (0001) plane in the hcp phase.

atoms in figures 4(b) and 5(d) present the atoms under the critical state in the phase transition process. The initial nucleation of the hcp phase (denoted by the green atoms) for the sample can be clearly seen from figures 4(b) and 5(b); here the strain $\varepsilon = 0.32$, the critical stress of the phase transition of Al in the loading process. For the strain $\varepsilon$ rising from 0.32 to 0.36, the hcp phase grows rapidly and becomes the dominant structure in the sample. It is worth noting that the hcp phase mainly exists in twins and twin boundaries in the (110)-plane and the (112)-plane, which are labeled in figure 4(c), and a part of the atoms form twin boundaries and fcc stacking faults. But the twins in the (112)-plane are unstable structures, which will disappear with increasing of the strain $\varepsilon$ as shown in figures 4(d) and 5(d). At the end of the loading process, the sample mainly exists in hcp twins and twin boundaries in the (110)-plane. Other atoms exist in the form of fcc stacking faults mainly in the (010)-plane and the (100)-plane. We can see from figure 3 that the hcp mass fraction $\lambda_1$ is about 0.8 at the end of the loading process; there are about 20% atoms in the form of fcc stacking faults and twin boundaries. Thus, the fcc and hcp structures coexist over a wide range of pressures above the critical pressure of the phase transition, which is similar to what was reported in [5].

To further investigate the dynamic properties of the fcc $\rightarrow$ hcp phase transition, we perform evolution of the atoms in two areas of the sample, which are labeled as A and B in figure 4(f) in the loading process. The character of phase transition in area A and B is shown in figures 6–8. We found that the process of fcc $\rightarrow$ hcp phase transition can be realized by two virtual steps. The first step can be seen from figure 6, which shows the snapshots of the atoms in the (010) and (100) planes from the initial fcc structure to the final hcp structure from the MD simulations,
Figure 7. Comparison of the atoms motion to the initial position in the (001) plane in the [010]-direction (a, b) and the [100]-direction (c, d), where $L_x : L_y = 6 : 1$ (a, b) and $L_x : L_y = 1 : 6$ (c, d); the red, green and purple atoms represent atoms at strain $\varepsilon = 0$, 0.32 and 0.4, respectively. Atoms in the [010]-direction begin to slip along the [100]- and [010]-directions at strain $\varepsilon = 0.32$. In the meantime, atoms in the [100]-direction begin to slip along the [010]- and [010]-directions.

in which the strains are (a) 0, (b) 0.32, (c) 0.40 (d) 0, (e) 0.32 and (f) 0.40. We can see from figures 6(a) and (d) that the atoms are in a regular configuration in the fcc phase at the initial stage; when the strain reaches 0.32, the atoms along the [001]-direction in the (010) and (100) planes are compressed to form a hexagon as shown by atoms 1–6; thus the phase transition completes its first step, in which the (010) and (100) planes transit to the hcp (0001) plane in areas A and B, respectively. The second step of the phase transition contains a slip procedure as shown in figure 7, which performs the moving of the atoms in the (001)-plane relative to the initial location at the strain $\varepsilon = 0.32$ and $\varepsilon = 0.4$ in areas A (a, b) and B (c, d), respectively; the atoms at strain $\varepsilon = 0$, 0.32 and 0.4 are shown by red, green and purple atoms, respectively. When strain $\varepsilon$ reaches 0.32, the atoms in the (001)-plane in the [010]-direction begin to slip along the [100]- and [010]-directions in area A; in the meantime, atoms in the [100]-direction begin to slip along the [010]- and [001]-directions in area B; thus the phase transition begins its second step. The slip procedure does not stop until the strain $\varepsilon$ reaches 0.40; then the second step finishes, and the fcc phase transits to the hcp phase. In order to see the character of phase transition more intuitively, we select some atoms from areas A and B and track their traces in our simulation. Figure 8 shows snapshots of the selected atoms at different strains $\varepsilon$; the two steps of fcc $\rightarrow$ hcp phase transition are illustrated clearly. Firstly, when the strain $\varepsilon$ reaches 0.32, the atoms in the (010) (figure 8(c)) and (100) planes (figure 8(g)) are compressed to form hexagons as shown by atoms from 1 to 6; then the fcc (010) and (100) planes transit to the hcp (0001)-plane, and the phase transition finishes its first step; secondly, when the strain $\varepsilon = 0.32$, atoms 8–11 begin to slip along the [010]- and [010]-directions, as shown in figures 8(c, d) and (g, h), respectively, and this finally results in the structure of the hcp phase.

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Figure 8. The traces of the atoms that come from the MD simulations show the dynamic phase transformation course from the fcc to the hcp structure. The strains in the snapshots are (a) 0, (b) 0.30, (c) 0.32, (d) 0.40, (e) 0, (f) 0.30, (g) 0.32 and (h) 0.40. The phase transformation mechanism is realized by two-step process; in (a)–(d) it contains a compression in the [001]-direction; and a slip in the [100]-direction; in (e)–(h) it contains a compression in the [001]-direction and a slip in the [010]-direction. Panels (a), (b), (e) and (f) show compression of the lattice in the [001]-direction, and panels (c), (d), (g) and (h) show the slip course.

The evolution of the structure of the sample in the unloading process is shown in figures 9 and 11. Figure 9 shows snapshots of sample under different strains in the unloading process, and the corresponding cross sections ((001)-plane) of the sample are shown in figure 10, respectively. The strains \( \varepsilon \) of the sample in (a–f) in figures 9 and 10 correspond to the strains \( \varepsilon \) of labeled points in (a–f) in figure 3. When the strain \( \varepsilon \) reaches 0.34 (corresponding to the pressure of 115 GPa), one can see that mutation points \( P_{2s} \) and \( T_{2s} \) appear in the \( P-\varepsilon \) (figure 1) and \( T-\varepsilon \) (figure 2) curves, but these are brought out not by the nucleation of the fcc phase but by the appearance of a new structure. We can see from figures 9(b) and 10(b) the new structure; twins in the (112)-plane that emerged in the loading process present at strain \( \varepsilon = 0.34 \), which is the mechanism that causes mutation points \( P_{2s} \) and \( T_{2s} \). In the meantime, the amount of atoms existing in the structure of fcc stacking faults (red atoms) increases as strain \( \varepsilon \) decreases from 0.34 to 0.28 as can be seen from figures 9(b, c) and 10(b, c); thus \( \lambda_2 \) begins to increase at strain \( \varepsilon = 0.34 \) and reaches about 0.2 at strain \( \varepsilon = 0.28 \), but the hcp phase is still the main structure in

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Figure 9. Snapshots of the sample in the unloading process. The strains $\varepsilon$ for different images are (a) $\varepsilon = 0.38$, (b) $\varepsilon = 0.34$, (c) $\varepsilon = 0.28$, (d) $\varepsilon = 0.26$, (e) $\varepsilon = 0.24$ and (f) $\varepsilon = 0$. The unstable structure ((112)-plane) presents itself at about $\varepsilon = 0.34$. The fcc structure nucleates and grows at about $\varepsilon = 0.26$. The sample mainly exists in fcc structure at the end of the unloading process. The color criteria are the same as in figure 4.

3.3. RDF analysis

To further confirm the occurrence of a phase transition, we also calculate the RDFs by using MD simulation. Figure 11 displays the RDFs of the sample at different strains $\varepsilon$ in the loading process (left) and the unloading process (right), where the strain $\varepsilon$ changes from 0 to 0.4. The red curve in figure 11 panel (left) presents an initial perfect fcc structure. Obviously, the three peaks that are marked by the circles present the typical fcc structure in the range of $r \leq 0.55$ nm, and all peaks experience an extending process because of the temperature effect. In the loading process, all the first peaks in the curves shift to the left slightly because the lattice constant in the [001]-direction is compressed continually during the loading process. When the strain $\varepsilon$ is above
Figure 10. Cross sections of the sample in the unloading process (a–f). Cross sections ((001) plane) of the samples in figure 9(a)–(f), respectively. The color criteria are the same as in figure 4.

0.16, the lattice constant in the [001]-direction is compressed to a certain extent; thus the first peak shows a distinct split. However, when strain $\varepsilon$ is above 0.32, the RDFs undergo prominent changes: the two split peaks merge into one peak, and a few new peaks that present the hcp phase appear; the sample transforms into hcp structure. In the unloading process, the RDFs undergo an opposite change relative to the loading process. In the initial stage ($\varepsilon = 0$), the RDF presents hcp structure mainly. When the strain $\varepsilon = 0.26$, the peaks in the curve become inconspicuous because voluminous atoms in the sample are undergoing a redistribution process, and the fcc phase begins nucleation. When strain $\varepsilon = 0.24$, it can be found that the peaks of the RDF curve appear again and mainly present an fcc structure. However, like the loading process, the first peak shows a distinct split because the lattice constant in the [001]-direction is compressed to a certain extent, and the split will disappear in the subsequent unloading process. At the end of the unloading process ($\varepsilon = 0$), the peaks of the curve present a typical fcc phase, and the sample mainly exists in the fcc phase.

4. Conclusion

Using the MD method, we have successfully simulated the process of fcc $\rightarrow$ hcp phase transition in single-crystal Al under uniaxial pressure with high-strain-rate loading in the [001]-direction. The corresponding unloading process at the same rate is simulated.

In the loading process, the critical pressure of the fcc $\rightarrow$ hcp phase transition of Al is about 90 GPa and the reduced volume is about 0.68 $V_0$. After the phase transition, about 80% atoms exist in twins in the hcp phase and twin boundaries in the (110)-plane; other atoms exist in fcc
Figure 11. RDFs of the sample under different strains $\varepsilon$ in the loading process (left) and the unloading process (right). In the loading process, for the case of $\varepsilon = 0$ (initial structure), there are three peaks in the range of $r \leq 0.55$ nm, similar to the perfect fcc structure.

stacking faults in the (100) and (010) planes. The fcc and hcp structures coexist over a wide range of pressures when the pressure is above the critical pressure of the phase transition.

The phase transformation mechanism can be realized by a two-step process. Firstly, the fcc (010)-plane and the (100)-plane transit to the hcp (0001) plane; secondly, the atoms in the area where the fcc (010)-plane transits to the hcp (0001)-plane slip along the [100]- and [010]-directions, and atoms in the area where the fcc (100)-plane transits to the hcp (0001)-plane slip along the [010]- and [001]-directions and this finally results in the hcp phase.

For the unloading process, the pressure of reverse phase transition is about 71 GPa. At the end of the unloading process, the fcc phase is the main structure in the sample, but the regular configuration of the atoms is destroyed.

All the results reported here are based on the Ercolessi and Adams EAM potential; the results are informative, but further study on the quality of this potential is needed. Also, the hcp–fcc transition is still lacking sufficient experimental evidence in the unloading process.

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