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Effects of edge dislocations on interstitial helium and helium cluster behavior in α-Fe

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ABSTRACT

The properties of interstitial He in the vicinity of an edge dislocation were studied using molecular dynamics (MD) simulation. The distribution of the binding energy of a single interstitial He to the dislocation with and without a jog is calculated. The results show that the distribution of the binding energy is governed by the elastic interaction between the interstitial He and the dislocation. The interstitial He is strongly attracted to the dislocation in the tensile region of the dislocation. The jog acts as a stronger sink to absorb interstitial He. The binding energy to the jog is even larger than that of the dislocation. A small He cluster (composed of three interstitial He atoms) was trapped by the dislocation core in the form of a chain along the dislocation line. The dislocation changes the migration behavior of the He cluster, and provides a pipe for the small cluster to exhibit one-dimensional motion. The diffusion of the He cluster in the dislocation is faster than in the defect-free iron, where the He cluster migrates three-dimensionally (3D). If the dislocation is decorated by a jog, the small cluster sinks deep into the jog. The jog prevents the He cluster from escaping.

1. Introduction

In a fission or fusion reactor, a high rate of helium is introduced into materials by direct implantation or indirect ways such as nuclear transmutation reactions. Because of its extremely low solubility in materials, helium precipitates into bubbles by absorbing vacancies, which deteriorate the mechanical properties of materials [1–5]. The interaction between He and irradiation-induced defects, especially vacancies, has been investigated [6,7]. A molecular dynamics study [8] has shown that a single interstitial He atom can easily migrate through a perfect α-iron lattice owing to its small migration energy of 0.08 eV. As a result, it is highly likely for helium to be trapped by sinks, such as vacancy clusters, interstitial clusters, dislocations and grain boundaries (GBs), and aggregate into bubbles even at low temperature. Thus, it is necessary to study the behavior of interstitial He in these sinks because the interaction of helium with these defects strongly affects the microstructural evolution [9], and hence the mechanical properties. For example, the behavior of helium in the presence of a vacancy cluster [10] or interstitial cluster [11] has been reported in recent years. The results showed that the SIA cluster migration slows down because of helium participation [11]. In particular, helium aggregation at GBs leads to dramatic embrittlement at high temperature, even for low densities. This triggered research using MD simulation: the resultant atomistic calculations demonstrated that He atoms were strongly bound to GBs [12] and that GBs provided a rapid diffusion path for He atoms [13].

At low temperatures, helium also induces irradiation hardening [14] and degradation of material lifetime [15], which was deduced from the reaction between helium and dislocations: the He cluster constitutes an obstacle, impeding dislocation motion. In addition, dislocations act as a pipe, promoting helium desorption. Thus, it is important to study He behavior in the presence of dislocations from the atomistic viewpoint. MD calculations [16] showed that the interstitial helium is strongly trapped in the tensile field of the dislocation core. In fact, the results reflect the elastic interaction between the dislocation and interstitial helium. In the present study, we first systematically determined the binding energy map of the interstitial helium in relation to an edge dislocation with and without a jog. We then explored the transport properties of a small He cluster in a dislocation using MD simulation.

2. Simulation method

Molecular statistics (MS) and molecular dynamics (MD) were employed to calculate the binding energy of a single interstitial He to an edge dislocation, and the diffusivity of He in α-iron, respectively. The *x*, *y* and *z* axes of the simulated crystal were oriented along the [1 1 1], [2 $\bar{1}$ $\bar{1}$] and [0 1 1] directions, respectively, as

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illustrated in Fig. 1. Fig. 1 schematically shows an $1/2[111](0\bar{1}1)$ edge dislocation in a simulation cell. The simulation size was set to be $30\sqrt{3}a_0 \times 10\sqrt{6}a_0 \times 20\sqrt{2}a_0$ along these three directions, where a_0 is lattice constant of α-Fe. An $1/2\langle 111\rangle\{110\}$ edge dislocation was created by the method used in [17]: periodic boundary conditions were employed in the x and y directions, which corresponded to the direction of the Burgers vector, b, and the line direction of the dislocation, respectively. Free conditions were used along the z axis, which was different from [17]. This was because we wished to focus primarily on the static behavior of the dislocation, rather than the dislocation motion. In the latter case, an external shear is needed to sustain dislocation motion.

The interatomic potentials describing the Fe–Fe, the Fe–He and the He–He interactions were determined by Ackland et al. [18], Wilson and Johnson [19], and Beck [20], respectively. Using this set of potentials, the formation energies of a SIA, an interstitial He, a substitutional He and a vacancy were calculated to be 4.88, 5.29, 3.24 and 1.70 eV, respective, which are in good agreement with previous results [8,16,21].

The binding energy of the He atom to the dislocation is defined as the difference of the formation energy of a He atom in a perfect Fe lattice and the formation energy of a He atom located in octahedral position in a 'dislocated' lattice [16]. Recently, *ab initio* calculations [22] indicated that the most stable configuration of an interstitial He is tetrahedral. This contradicts the results of our present calculations obtained using the above pair potentials, which show that the interstitial He in the octahedral site has lower formation energy. The physical reason for this discrepancy is the change in the magnetic moment of neighboring Fe atoms produced by the He defect. In order to accurately describe the Fe–He potential, the many-body interaction between He and Fe, rather than the pair potential, should be considered. Ref. [10] uses this as a basis for fitting a new Fe–He potential, in which the calculated formation energy of the interstitial He at the tetrahedral and octahedral sites approached the *ab initio* values. However, the formation energy of He at the octahedral site is still slightly larger than that at the tetrahedral site. To our knowledge, there are at present no Fe–He potentials that reproduce the *ab initio* results. Besides, the present Fe–He pair potential yields a He behavior similar to that predicted by the *ab initio* calculations. Thus, we have adopted Wilson's He–Fe potential in the present study to calculate the binding energy of the interstitial He located initially at the octahedral site, instead of the tetrahedral site. We believe that the map of binding energy to the dislocation is qualitatively the same regardless of whether the He atom is at the octahedral or tetrahedral site.

Diffusion of a small He cluster consisting of three He atoms in the presence of a dislocation was calculated at 600 K. The diffusion coefficient, D, of the He cluster was determined from the square displacement of the center mass of the He cluster (CMHC): $D = \frac{R^2}{2nt}$, where R^2 is the squared displacement (SD), n is the dimensionality of the system, and t is the simulation time. The method has been described in detail in Refs. [23,24].

3. Results and discussion

3.1. The map of the binding energy of an interstitial He to a dislocation

Fig. 2 is the binding energy distribution of an interstitial helium at the octahedral sites in the $(2\bar{1}\bar{1})$ plane, where the dislocation core is around the point (0,0.1) and the dislocation line is perpen-

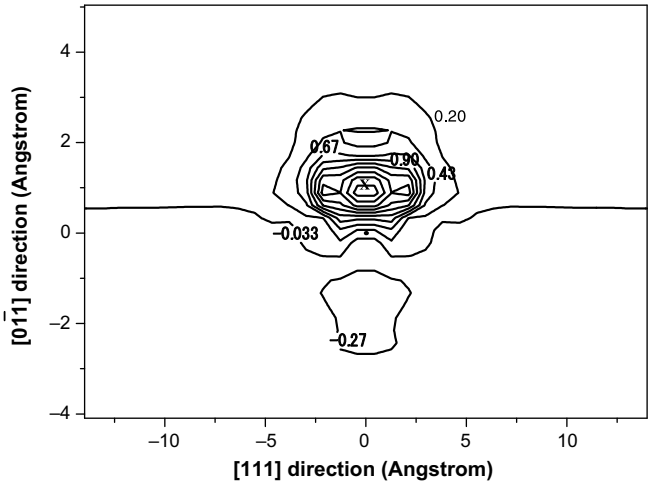

Fig. 2. Binding energy map of an interstitial He to a dislocation in the $(2\bar{1}\bar{1})$ plane. The separation between contours is about 0.23 eV and the position denoted by the cross star is the maximum binding energy (2.26 eV).

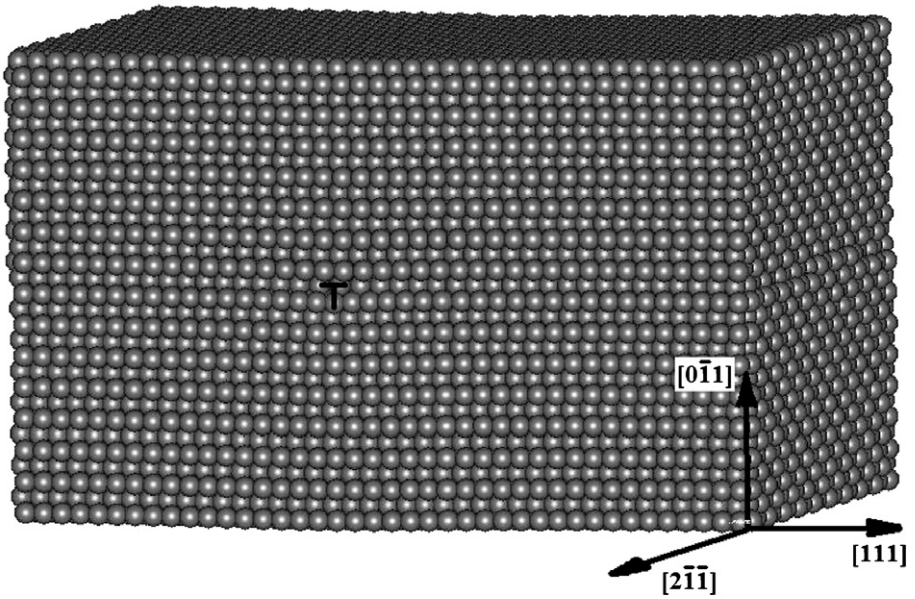

Fig. 1. Schematic of a $1/2[111]$ $(0\bar{1}1)$edge dislocation in a simulation cell.

dicular to the page. Although the binding energy is a discrete function of the octahedral position, its distribution was represented by the contour lines around the dislocation line, similar to the stress field of the dislocation. Thus, we plotted the binding energy as a successive function of the octahedral position in Fig. 2. Clearly, there is an attractive region with positive binding energy above the glide plane (the area above zero along the $[0\bar{1}1]$ direction). The maximum energy is 2.26 eV in the center of this region. That is, the He atom can be trapped deeply in the dislocation up to a high temperature of, say, around 900 K if the He resides in the center of the attractive region [16]. However, the binding energy declines rapidly. About 0.5 nm away from the center along [1 1 1] direction, the binding energy has decreased to 0.2 eV. For the binding energy of 0.2 eV, the temperature required to dissociate a He atom from the dislocation is about 77 K, which is temperature of liquid nitrogen and is quite low compared with 900 K. The repulsive region is below the glide plane, where the minimum energy is −0.38 eV at about the point $(0, -0.11)$ in Fig. 2. The contour of the binding energy almost corresponds to the strain field interaction between the interstitial He and the dislocation: the strain field of the He is spherical and compressive. Therefore, the interstitial He should be attractive in the tensile field of the dislocation and repulsive in the compressive field. This is confirmed by the contour distribution: the attractive region with positive binding energy is located in the tensile field of the dislocation, and the repulsive region with negative energy, in the compressive field. It is worth noting that when the He goes into the center of the attractive region, its initial octahedral configuration is transformed and rearranged along the $\langle 1\,1\,1\rangle$ direction of the Burgers vector, together with the neighboring Fe atoms. The configuration of the He rearrangement is shown in Fig. 3. Around the dislocation core is presumably a large strain field along the Burgers vector on the tensile side. Therefore, the configuration of the He preferentially transforms in order to reduce the tensile strain of the dislocation along the [1 1 1] direction.

Generally, dislocations in real specimens contain jogs, which are known to absorb interstitials leading to dislocation climb. What is the structure of the interstitial He with the dislocation decorated by a jog? We thus introduced a jog. The jog was formed by artificially removing two atomic layers in the extra atomic plane ([1 1 1] plane) of an edge dislocation along the dislocation line for the region of $y > 0$. Static lattice relaxation was carried out until a minimum energy of the system was obtained. The boundary condition used here is the same as that mentioned in the Section 2. Fig. 4 shows the structure of a jog where the 1/2[1 1 1] jog is in the $[2\,\bar{1}\,\bar{1}]$ plane (denoted A plan) with a step of 2 atomic layers and the line is along the $[0\,\bar{1}\,1]$ direction. The binding energy was calculated by scanning each octahedral interstitial position on the $(0\,\bar{1}\,1)$ plan (B plan), which intersects with the jog as shown in Fig. 4. Fig. 5 illustrates the binding energy distribution of a He atom to the dislocation with a jog. We can see that the jog was located in the region denoted by dash line. In Fig. 5, the maximum energy just in the jog is 2.87 eV, larger than the binding energy of the He to the dislocation (2.26 eV), indicating that the jog acts as a stronger sink than even a pure dislocation. This is due to the much stronger tensile strain field induced by the jog. Also, we can discern the

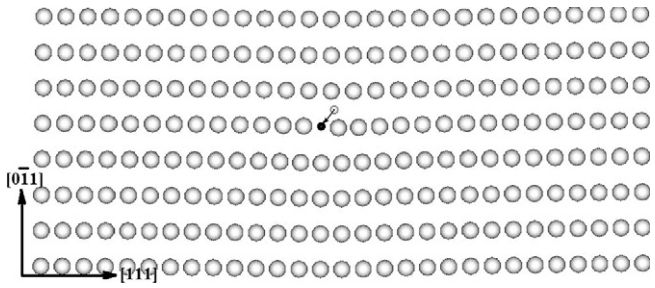

Fig. 3. Rearrangement of an interstitial He atom in the dislocation. Large white circles are Fe atoms. Small white circle is the initial octahedral position of a He atom, and small black circle is the final position of a He atom after relaxation. An arrow connects the initial and final position of a He atom.

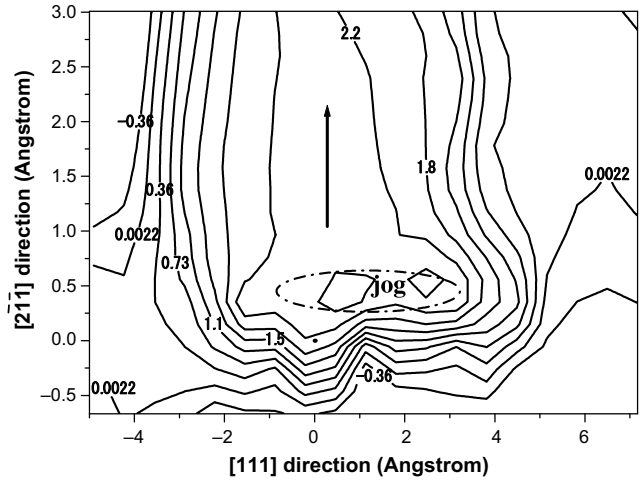

Fig. 5. Binding energy map of an interstitial He to a dislocation with a 1/2 [1 1 1] jog in the $[0\,\bar{1}\,1]$ plane. The dashed line shows the jog position and an arrow denotes the dislocation pipe.

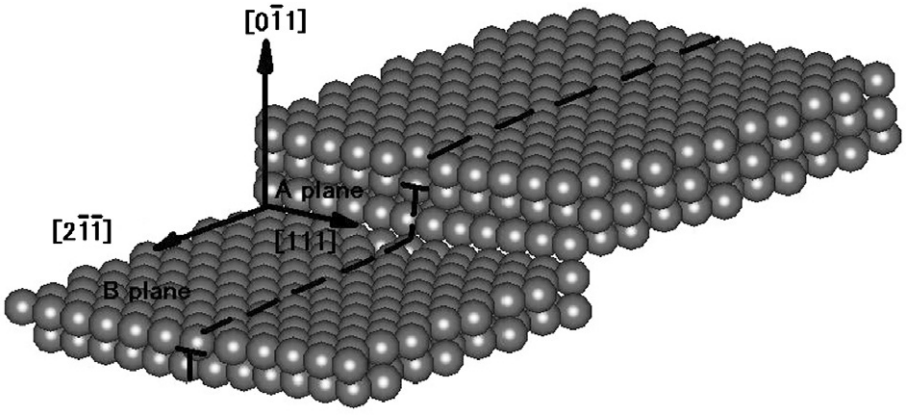

Fig. 4. Schematic of a 1/2 [1 1 1] jog. Dashed line represents the dislocation line.

dislocation pipe near the center of the x-axis along the $[2\bar{1}\bar{1}]$ direction, denoted by an arrow in Fig. 5. In this pipe, the binding energy is above 2.0 eV.

3.2. Helium cluster migration in the dislocation

Heinisch [16] found that the interstitial He tends to migrate into the core from the tensile side of the dislocation. In the core, the interstitial He has a high binding energy, as shown in Fig. 2. The interstitial He are thus restrained in the core. Not surprisingly, a single interstitial He migrates along the dislocation line, without migrating away from the dislocation [16]. In a fusion reactor, a large amount of helium is generated. The interstitial He may accumulate into clusters in the vicinity of dislocations. An interesting question is: What is the migration mechanism of a He cluster migrating along a dislocation?

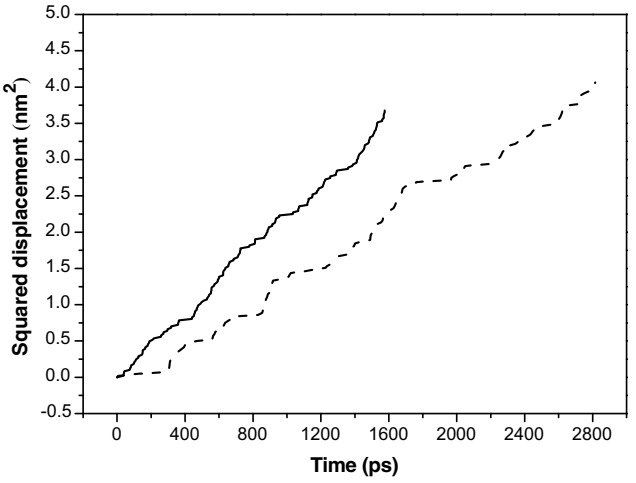

Fig. 6. SD of the He cluster as a function of time at 600 K. The solid line is the SD of the 3-He chain along the dislocation line, while the dashed line is that of the 3-He triangle in dislocation-free iron.

Three interstitial He atoms were randomly placed within each other's interaction range in the vicinity of a dislocation. After relaxation, the interstitial He spontaneously align in a linear chain confined to the dislocation core. The nearest-neighbor distance between the He atoms is about 0.17 nm at 0 K. Fig. 6 is the SD of the mass center of the chain as a function of time at 600 K (solid line). We also plotted the SD of a 3-He cluster in dislocation-free iron (dashed line). The slope of the solid line is higher than that of the dashed line. This means that the 3-He chain migrates faster in the dislocation than the 3-He cluster in the dislocation-free iron. It is interesting to note that the He cluster moves along the dislocation line in a worm-like fashion. Snapshots are shown in Fig. 7, where the dashed line represents the dislocation line. It can be seen that the He cluster stretches its chain in the $(0\bar{1}1)$ plane at the initial time. At about 6.8 ps, the chain shrinks, like a bow, in which the center He atom leaves the $(0\bar{1}1)$ plane and rises a little. After about 0.3 ps (i.e. at about 7.1 ps), the chain again stretched its body in the $(0\bar{1}1)$ plane and its mass center goes ahead about 0.866 a_0. In the next walking step, the same motion fashion is repeated as seen in Fig. 7 (the snapshots at 9.6 ps and 10 ps). Our detailed analysis revealed that the motion along the dislocation line, i.e., the $[2\bar{1}\bar{1}]$ direction, was faster than that along the other two orthogonal directions, as shown in Fig. 8(a). This demonstrates that the 3-He chain mainly exhibits 1D motion. Although there are small discrepancy in the preferential interstitial He site between *ab initio* calculation [22,25] and the calculation using the present He–Fe pair potential, both of the two models showed that a He substitution has the smallest formation energy compared with interstitial He (including octahedral and tetrahedral). This may be due to the largest free volume of the substitution site. For the edge dislocation core, the free volume is also large compared with

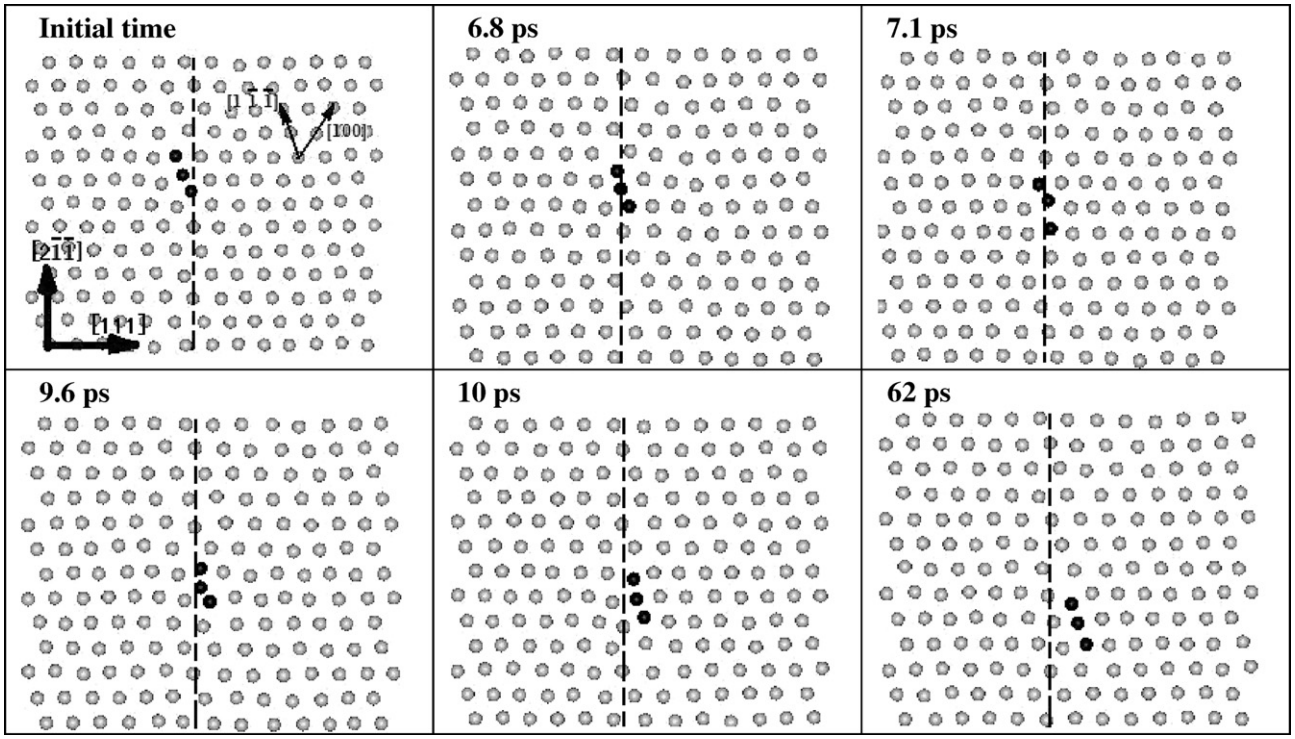

Fig. 7. Snapshots of the migration of a 3-He chain in a dislocation core. The black circles are interstitial He and grey circles are Fe atoms. The dashed line represents the dislocation line, which lies in the same position in all the snapshots.

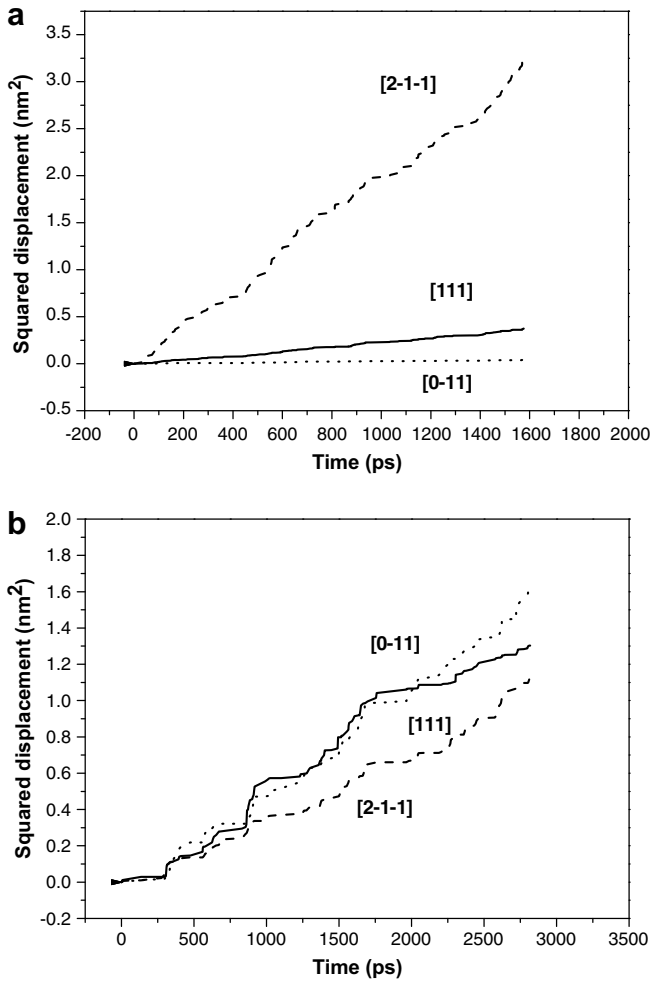

Fig. 8. SD along different directions at 600 K: (a) the 3-He chain in a dislocation (b) the 3-He cluster in defect-free iron. The solid, dashed, and dotted lines represent, respectively, the SD along the [1 1 1], $[2\,\bar{1}\,\bar{1}]$, and $[0\,\bar{1}\,1]$ directions.

interstitial sites. Moreover, the migration energy of interstitial He calculated by the two models is close [25]. Therefore, migration behavior of the 3-He chain may be quantitatively similar when using the *ab initio* calculation.

Fig. 8(b) shows the components of the SD along the [1 1 1], $[2\,\bar{1}\,\bar{1}]$and $[0\,1\,\bar{1}]$ directions for diffusion of the 3-He cluster in bulk Fe. In contrast to the 3-He chain in the dislocation, the He cluster exhibits 3D diffusion behavior. The diffusion mechanism is also different. The 3-He cluster in the iron forms a triangle with each side measuring about 0.17 nm. The formation energy of this configuration is 13.56 eV. Snapshots are shown in Fig. 9. When the triangle jumps, it first ejects the nearest-neighbor Fe atom, forming a self-interstitial (SIA). The snapshots at the time of 4 ps and 5 ps present the evolution of the SIA, where the open circle is the SIA. Then, the triangle takes the neighboring lattice position. The SIA generated may jump far from the triangle cluster, which is even beyond the scope of the frame at 15 ps. A few picoseconds later, it returns. At about 25 ps, the SIA reappears from the other side of the triangle. Finally, the SIA shares the lattice position with the triangle cluster at 36 ps. During this process, the triangle did not dissociate into single interstitial He. The interstitial He jumped together and generated the SIA, which resulted in a higher energy barrier than in the case of the migration of the 3-He chain in the dislocation. In the latter case, the 3-He chain has lower formation energy (7.87 eV). In addition, the chain need not eject any Fe atoms to generate a SIA during the diffusion. Interestingly, a single interstitial He has lower migration energy (0.08 eV) in the defect-free iron [8] than in the dislocation (0.4–0.5 eV) [16], which differs from the case of the small He cluster. This could be due to the different migration mechanism, namely, the cooperative migration of the clusters. It is interesting to study the migration of larger He clusters in the dislocation in the next work.

In our simulation, the squared displacement (R^2) was calculated by decomposing the He cluster trajectory into a sequence of jumps with the same length of jump vectors (about 0.866 a_0, the nearest-neighbor distance of bcc Fe). And then, R^2 can be obtained by the following equation [23]:

$$\langle R^2(t)\rangle = \frac{1}{N_{tr}}\sum_{i=1}^{N_{tr}}\left[r_i(t) - r_i^0\right]^2$$

where r^0 and $r_i(t)$ are initial and instantaneous position of atom i and N_{tr} is the number of tracer atoms in the simulated crystallite. Here, we assumed that the He cluster jump is independent event and random walk. To accurately calculate the diffusion coefficient,

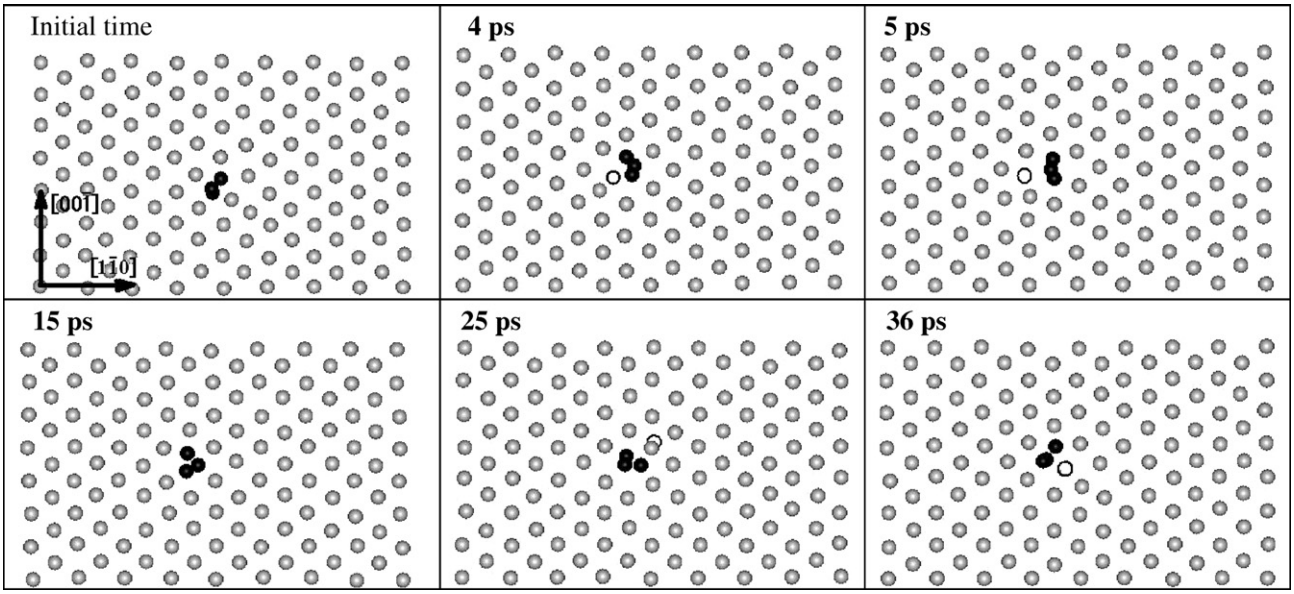

Fig. 9. Snapshots of 3-He cluster migration in defect-free iron. The black, grey and the open circles are interstitial He, Fe lattice atoms and Fe interstitials, respectively.

our further study will involve correlation analysis, and corresponding statistics will be done. In any case, we believe that the dislocation provides a fast diffusion pipe for the small He cluster, compared with the diffusion in defect-free crystal. Previous experimental research [26] also supports our simulation result very well. This experiment observed that He bubbles walk in the form of Brownian type in the matrix. When an edge dislocation is present, He bubbles easy move along the dislocation.

When the three interstitial He atoms were placed in the vicinity of a jog, the number of He cluster jumps was too small to satisfy the statistical requirement within the simulation time of 2 ns. Thus, the jog, as a strong sink, contributes to He accumulation, but not He diffusion. In addition, He atoms may impede the dislocation climb, and the dislocation may be pinned by He cluster when He atoms aggregate in the jog. The relative research requires further investigation.

4. Conclusion

The behavior of interstitial He in the presence of an edge dislocation was studied using the MD method. The binding energy of the interstitial He to the dislocation reflected the elastic interaction between the two. The interstitial He accumulate in the tensile side of the dislocation and are bound to the dislocation core. Thus, the 3-He cluster becomes aligned in the linear chain, parallel to the dislocation line, and confined to the core. This configuration contributes to the He chains' 1D migration behavior along the dislocation line. The dislocation also provides a fast diffusion pipe for the transportation of small He clusters. This differs from the 3D motion of the 3-He cluster in defect-free iron, in which the optimal configuration of the 3-He cluster is a triangle.

Acknowledgments

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