Solitonic Migration and Collisions of Self-Interstitial Defects in BCC Iron

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Self-interstitial atoms in bcc iron display unusual migration behaviors; strong anisotropy toward a (111) direction with occasional rotation to an equivalent direction as well as retracing the same way as it has come, and also ultra-high mobility when they are clustered. These singularities cannot be explained by simple interstitial or interstitialcy diffusion mechanisms. However, some of them will be well accounted if the SIA could behave as a soliton, which makes three-dimensional movements in appearance, but essentially a serial combination of one-dimensional migration. Indeed, a crowdion, one of isomeric configurations of the self-interstitial atom, has an atomic arrangement very similar to the one-dimensional dislocation core structure, whose migration kinetics has been well modelled by a one-dimensional soliton equation. Here we report a decisive observation that both a single and colliding two crowdions really behave as solitons in iron crystals using molecular dynamics simulations. In addition, we ascertain that the present results are attributed to the intrinsic nature of the crowdon where an overall potential felt by atoms therein is very shallow and periodical along the migration direction. [doi:10.2320/matertrans.47.2658]

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1. Introduction

Recently, molecular-dynamics (MD) simulations have revealed unusual behaviors of irradiation-induced self-interstitial atoms (SIAs); strong anisotropy along a specific crystallographic direction as well as ultra-high mobility when they are clustered. An SIA in bcc-iron has two allotropic configurations; one is stable being composed of two dumbbell atoms splitting over a lattice point with their bonding axis laid on a (110) direction, and the other is metastable being at an excited state with its bonding axis rotated from the (110) direction to a (111) direction according to the Fermi-Dirac distribution function. The latter one, called crowdon, has a little higher formation energy by about 0.2 eV than the stable one and has high mobility along the (111) direction along which atoms are most densely packed.

Theoretical treatment for the dynamical properties of an SIA has firstly appeared in the dislocation model by Frenkel and Kontrova (FK) who assumed a one-dimensional atomic chain composed of atoms connected by springs and an extra atom. Their formulation (FK-equation) was developed into more sophisticated form by Frank and van der Merwe and then applied to the internal friction of actual materials by Seeger and Schiller. Lately the FK-equation was, at a more sophisticated form by Frank and van der Merwe, and Kontrova (FK) who assumed a one-dimensional atomic chain, one of soliton equations. So far, a large number of studies in relation to the theory of FK has been performed on model lattices. Despite the many theoretical studies, there have been still few reports whether an SIA itself behaves as a soliton or not. In the present work, on the assumption that the dislocation core structure is similar to the arrangement of atoms around a self-interstitial atom, we aim at obtaining the evidence that a crowdion, having the most densely arranged atomic configuration of the SIA, really plays as a soliton using realistic MD simulations. We also aim at explicating the origin of the unusual migrational behavior of SIAs in irradiated crystals.

2. Modeling for One-Dimensional Soliton Equations

We carried out a calculation described below in order to assure whether the interatomic potential, which we are going to apply to the three-dimensional MD simulations for the dynamical process of crowdions, can reproduce the same atomic structure of a static crowdion as calculated from the one-dimensional FK-equation. We can estimate the values for parameters appeared in the FK-equation that gives the total energy of the one-dimensional dislocation system,

\[ V = \frac{\alpha}{2} \sum_{n=0}^{N-1} (r_{n+1} - r_n - b)^2 + \frac{w}{2} \sum_{n=0}^{N-1} \left[ 1 - \cos \left( \frac{2\pi r_n}{a} \right) \right], \]  

where \( a \) is the period of a sinusoidal potential on the atomic chain, \( b \) is the equilibrium distance between the atoms, \( w \) is the potential depth, \( r_n \) is the position of the \( n \)-th atom, and \( \alpha \) is the force constant. The positions of SIAs in the atomic chain are obtained by differentiating the FK-equation,

\[ \frac{\partial V}{\partial \zeta_n} = -2l_0^2 w(\zeta_n + 2\zeta_n^2 + 2 \zeta_n) + \pi w \sin(2\pi \zeta_n) = 0, \]  

where the parameters in eq. (1) are converted as \( \zeta_n = r_n/a, l_0^2 = \alpha a^2/(2w) \). In order to evaluate the values of these parameters, we performed two molecular-dynamical calculations to obtain the changes in the total energies; \( \Delta E_c \) for the case when one of the crowdion atoms is coercively pushed by \( \Delta \tau \) along the \( (111) \) axis allowing no relaxation for the rest of atoms. The many-body interatomic potential we applied to the present calculations is based on the embedded-atom-method theory by Daw and Baskes, and the formulations for potential of bcc iron were constructed by Haftel et al. in such a way that the cohesive energy, elastic properties, the vacancy formation energy, and the surface reconstruction of the real crystal are reproduced. The procedures for the present calculations are as follows. Every sample with three dimensional periodic boundaries is pre-annealed for equilibration at temperatures below 1 K and at pressures within \( \pm 10^6 \) Pa setting the time...
method similar to that applied to the evaluation of parameters

Three-Dimensional Dynamical Properties of Crowdions

Fig. 1 Atom positions on a crowdion axis; (a) minimum, (b) maximum energy configurations by MD, (c) those obtained from the FK equation, and (d) the equilibrium lattice point of a perfect crystal. \( r/d \) means atom position scaled by the equilibrium atom spacing of a perfect crystal. The arrows show the center of symmetry.

The change in the energy \( \Delta E_k \) is sinusoidal and is fitted to \( w/[2(1 - \cos(2\pi \Delta r/a))] \) for very small values of \( \Delta r \). The parameters are calculated to be \( a = b/8 \) and \( w = 7.70 \times 10^{-4} \) eV. It is notable that the value of \( w \) is very small compared to the migration energy (0.33 eV) of SIAs in bcc iron due to an ordinary interstitialcy diffusion mechanism, which indicates that the crowdion atoms feel extremely low potential barriers. A detailed investigation of the \( \langle 111 \rangle \) interstitial configuration, which is performed by another MD simulation, has revealed that there are two isomorphs; one has a minimum-energy configuration with a three-fold symmetry on the \( \langle 111 \rangle \) crowdion axis (see Fig. 1(a)), and the other has a maximum-energy configuration with both a three-fold and a two-fold symmetries (see Fig. 1(b)).

The energy difference between the above two configurations, to which the periodicity of the weak potential is originated, is just equal to the height of the periodic potential. In a perfect lattice an atom has eight nearest neighbors (A\(_1\) to A\(_8\)), and, for small values of \( \Delta r' \), the gain in the elastic energy is approximately fitted to

\[
\Delta E_k = \sum_{i=1}^{8} \frac{1}{2} \alpha(\Delta a_{Ai} - b)^2 = \frac{4}{3} \alpha(\Delta r')^2, \tag{3}
\]

where \( \alpha \) is evaluated to be 5.8 eV/b\(^2\). Now, because all the parameters \( a, w, \alpha \) are evaluated, Equation (2) can give the equilibrium atom positions. As shown in Fig. 1(c), the calculated atom positions are in fairly good agreement with those obtained from the MD simulation, indicating that the one-dimensional FK-equation also gives precise atom positions of a crowdion in a realistic three-dimensional crystal lattice. Figure 1(d) shows, for a comparison, the atom positions for a perfect lattice. The present result suggests that the dynamical behavior of a crowdion has solitonic feature indeed.

3. Three-Dimensional Dynamical Properties of Crowdions as Solitons

In order to investigate the migrational properties of a single crowdion, a crystal of bcc iron is constructed using the method similar to that applied to the evaluation of parameters in eq. (2). A crowdion is preceidently introduced into a crystal with the size of 18 \( \times \) 22 \( \times \) 26 in units of lattice parameter. The rectangular simulation box is specially contrived to avoid the repeated movement of a crowdion on a fixed path, which takes place when the crystal has cuboidal shape. After sufficiently long equilibration time of \( 3 \times 10^8 \Delta t \) \( (\Delta t = 10^{-5} \) r, in this case), during which both the average temperature and pressure of the crystal are controlled to be below 10\(^{-3} \) K and 10\(^3 \) Pa respectively, one of crowdion atoms is knocked-on at a velocity of \( v_k \) with a kinetic energy of \( E_k (= m v_k^2/2, m \) is the mass of an iron atom) toward a direction of the crowdion axis. In case \( E_k \) is low enough, only small oscillation of the atoms is observed, whereas, in case \( E_k \) exceeds a critical value \( (E_c) \), the crowdion is observed to steadily propagate along the \( \langle 111 \rangle \) direction. In Fig. 2 the initial propagation velocity \( (v_0) \), which is defined as the tangent of the propagation distance versus elapsed time immediately after the knock-on, is plotted against the knock-on velocity \( (v_k) \). The critical values for the occurrence of the steady propagation are \( E_k^c = 6.0 \times 10^{-3} \) eV and \( v_k^c = 7.5 \times 10^3 \) m/s. Remembering that a crowdion is composed of about 8 atoms, it will be reasonable to consider that the critical knock-on energy is simply the sum of the weak potential (w). As can be seen in Fig. 2, there are two characteristic regions for the initial propagation velocity of a crowdion; (I) the value of \( v_0 \) asymptotically approaches \( v_k \), and (II) the value of \( v_0 \) is almost equal to \( v_k \). We found that a crowdion in the region I stops its movement after some running time, whereas a crowdion in the region II never dump nor stop. For instance, when the initial propagation velocity is \( 1.4 \times 10^2 \) m/s (region I), the ultimate propagation distance is over \( 10^3 b \). Due to a large CPU-time consumption, we couldn’t have measured the ultimate propagation distances in case the initial propagation velocity exceeded the above instance. Figure 2 also indicates that the effective mass of a crowdion depends on the knock-on velocity so far as the initial
propagation velocity is low. The present observations suggest that so far as the propagation energy is higher than a critical value, the energy is localized to only a small region around the crowdion. This means that a crowdion in bcc iron really behaves as a soliton migrating without dissipation.

Now we show the results for collisions between two crowdions which are initially put apart some distance on the same \( h_{111} \) atomic row. This time, the crystal with the size of \( 26 \times 30 \times 34 \) in units of lattice parameter is used. Investigated are three instances where (1) crowdions have the same but opposite velocity, i.e., head-on collisions, (2) a moving crowdion collides with a stopping one, and (3) crowdions with different propagation velocity make a rear-end and head-on collisions.

In case the crowdions make head-on collisions, three different results are observed depending on the magnitude of the velocity (see Fig. 3); (a) for low velocity \( (v_0 = 3.3 \times 10^2 \text{ m/s}) \) one crowdion is stopped from propagation and the other is bounded with a little higher velocity than that before collision, (b) for medium velocity \( (v_0 = 4.9 \times 10^2 \text{ m/s}) \) a semi-elastic collision takes place, and then one crowdion gains a little higher propagation velocity and and the other loses a little, (c) for high velocity \( (v_0 = 7.5 \times 10^2 \text{ m/s}) \) an elastic collision takes place.

In case a moving crowdion collides with a stopping one, three different results similar to the above head-on collisions are observed depending on the magnitude of the velocity also; (d) for low velocity \( (v_0 = 5.7 \times 10^2 \text{ m/s}) \) the stopping crowdion keeps stopping and the moving one is bounded with nearly the same velocity as before collision, (e) for medium velocity \( (v_0 = 7.4 \times 10^2 \text{ m/s}) \) the stopping crowdion is pushed ahead and the moving one is bounded with a little lower velocity than that before collision, (f) for high velocity \( (v_0 = 1.1 \times 10^3 \text{ m/s}) \) there an elastic collision takes place.

As shown in Figs. 3(a)–(f) the total kinetic energies are virtually conserved during the collisions, giving no excess energies to the atoms surrounding the crowdions, and consequently no gain in the average temperature of the system. The non-elastic collisions (Fig. 3(a,b,d,e)) are possibly caused by the transference of small vibrational energies among the atoms located between the two crowdions.

In cases of different velocities for both a head-on and rear-end colliding crowdions, propagation velocities are completely exchanged after the collision. These are shown in Fig. 4(a) for the case of head-on collision where initial propagation velocities are \( 3.3 \times 10^2 \text{ m/s} \) and \( 4.9 \times 10^2 \text{ m/s} \), and in Fig. 4(b) for the case of rear-end collision where initial
propagation velocities are $3.3 \times 10^2$ and $7.5 \times 10^2$ m/s. These results bear close resemblance to an analytical solution, which is modeled for the collision of two solitonic waves with different amplitudes and speeds, indicating an evidence that not only a single crowdion but also a couple of crowdions really behave as solitons.

Summing up, the present results show that in case at least one of the colliding crowdions has sufficiently high propagation velocity, the interactions between the crowdions are elastic. On the contrary, collisions are no more elastic for crowdions with low velocities. It is very interesting that an attractive interaction has worked between the two approaching crowdions when they are within a critical distance. This phenomenon is more pronounced on the case for lower propagation velocities as displayed in Figs. 3(a,d) and 4(b).

We find that a migrating SIA in bcc iron frequently changes its direction and occasionally traces back on the same way as it has come. This is shown in Fig. 5, which is obtained by annealing an iron crystal containing an SIA at 300 K under another MD simulation. In this figure, the trajectory is obtained by sampling the position of SIA at every 1 ps. Symbols $S$ and $E$ in Fig. 5 mean the starting and ending positions, and green lines indicate that the SIA lies in the state of $h_{110}i$ dumbbell configuration and the red ones in the state of crowdion configuration. The arrows indicate the retracement of the SIA. It is notable that the trajectory has a strong anisotropy along $h_{111}i$ directions and that the SIA makes long jumps when it lies in the state of a crowdion.

Although these singularities cannot be explained by simple interstitial or interstitialcy diffusion mechanisms, they can be well accounted if the SIA could behave as a soliton making three-dimensional movements in appearance, but essentially a serial combination of one-dimensional migration. Here we remember that among the colliding two crowdions, one of the crowdions is in some cases bounded to its initial route after the collision (Fig. 3). Although the detailed mechanism is not given here, the present retracing phenomenon is associated with the results of colliding crowdions; the retracement is suggested to be caused essentially by an interaction between a moving crowdion and lattice phonons, which casually give rise to a locally crowdion-like dense arrangement of atoms having vibrational energies (estimated to be $h\nu \sim 10^{-2}$ eV for $\nu \sim 8$ THz, the highest phonon frequency of iron) comparable to the kinetic energy of a migrating crowdion ($E_k > 6.0 \times 10^{-3}$ eV). A careful inspection may point out that the retracing phenomenon of an SIA has been already visualized in figures presented in literatures, but unfortunately it is a little hard to recognize it distinctly.

So far, studies on solitons in real materials have been confined to those concerning wave mechanics such as optics and plasma dynamics. To the author’s knowledge, the present work firstly visualized that a certain defect structure also plays solitonic behavior during its migration in real crystals.

4. Conclusion

Crowdions in bcc iron are ascertained to behave as solitons through investigations of their migrational and colliding sequences at very low temperatures, using molecular dynamics simulations. It is also shown that the solitonic features of a crowdion can well account for the migrational singularities of self-interstitial atoms at high temperatures.
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REFERENCES