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Self-interstitial structures in body-centred-cubic W studied by molecular dynamics simulation

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Abstract. This paper reports a simulation study of the detailed structure of an interstitial in a body-centred-cubic (bcc) metal, tungsten (W), using molecular dynamics and the embedded-atom method. Several distinct configurations can be discerned, including the well-known split-interstitial ⟨011⟩ dumb-bell. The ⟨111⟩ crowdion is also observed, as is a set of similar, one-dimensional, defect configurations. These are grouped into a proposed classification scheme, in which the traditional crowdion is revealed as part of a whole family. The defect energies of the various configurations are computed, and comparisons are made with bcc iron (Fe) results. The ⟨011⟩ dumb-bell is the energetically most favourable configuration in W. It is found to have a radius of approximately one W lattice constant, 3.16 Å.

1. Introduction

In order to control and design desired properties of modern materials, a thorough understanding of the material and the manufacturing process is needed. With methods like molecular beam epitaxy (MBE) that allow control of the growth process on the scale of an atomic monolayer, there is a need for knowledge on an atomistic level. Of special interest is the knowledge of defect structures and behaviour, as the concentration and type of defects present in a material has a strong impact on its properties, like electrical conductivity and mechanical strength. For vacancies and vacancy agglomerates, positron annihilation spectroscopy [1–4] (PAS) can be used to gather such information. This has been done for metal carbides and nitrides [2–4]. For monovacancies, PAS has a lower detection limit of about one tenth of a part per million (0.1 ppm); the sensitivity increases with increased vacancy cluster size. The lifetime of the trapped positrons mainly provides information about the size and density of defects. For interstitials, some information can be gained by the use of electron diffraction [5] and x-ray diffraction, but detailed knowledge is readily provided by atomistic simulations such as molecular dynamics (MD). MD was thus recently used to study divacancy and di-interstitial pairs in Si and the configurations obtained were supported by high-resolution electron microscopy [6]. MD has furthermore been used to study e.g. the impact of ions, with a kinetic energy in the keV range, on metals and metallic bi-layers [7], to compute energy accommodation and point defect generation in metallic superlattices [8], as well as to study many other phenomena [9–13]. The results of such studies rely heavily on the use of a

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realistic interaction potential which includes many-body effects, such as the embedded-atom method (EAM) described by Daw and Baskes [14]. To date, most MD studies on point defect arrangements in the class of bcc metals have concerned Fe and Mo [15, 16]. This paper reports a study of the detailed structure of a self-interstitial atom (SIA) in a bcc metal, tungsten (W), using MD and the EAM.

2. Methodology

The simulation reported in this paper has been carried out by molecular dynamics, with the interaction between atoms given within the EAM [14] framework, using the form of the EAM given by Johnson and Oh [17]. The number of particles is constant, and the volume is kept constant during the simulation, as is the total energy, even though the amounts of kinetic and potential energy might vary. Thus, the ensemble used is the *EVN*-ensemble. The time step is dynamically adjusted to optimize speed while conserving the energy. Other details on the implementation can be found in previous work by the authors [8].

The structures studied in this work were prepared in the following manner. First, a bulk portion of the material was generated, with the bulk simulated by the standard method of applying periodic boundary conditions in all three directions. The simulation cell was then adjusted to achieve a zero mean pressure at the temperature 0 K. The defect was introduced by adding an extra atom close to the centre of the simulation cell, in the middle between two atoms at their ideal lattice points, on the $\langle 111 \rangle$ diagonal. The temperature was then set to 2000 K after which the structure was allowed to equilibrate for 1ps simulated time to accommodate the defect. A further 500 ps were then simulated and analysed. The analysis was carried out by visually inspecting the sequence of configurations to identify the meta-stable configurations. The simulation contained 3457 atoms arranged in a cube with a side of 12 unit cells.

The specific energies of the various defects were computed using a fully relaxed cell. The defect configuration was fixed and the lattice around it relaxed, and then cooled to 0 K, keeping the pressure in the cell at zero. The energy of the configuration was then computed and compared to the base configuration at 0 K, without the defect, also at zero pressure. The stability of the observed configurations was tested by quenching them and then allowing the lattice to relax.

The vibration amplitudes at the simulation temperature of 2000 K were determined from the standard deviation of atoms that were not part of the defect configuration during the simulation.

3. Results

3.1. The dumb-bell

The interstitial is accommodated into the lattice through small distortions. There exist several distinct configurations in which the interstitial stays long enough to be considered at least meta-stable. The first of those is the split-interstitial SIA dumb-bell (referred to further on simply as the dumb-bell), in which two atoms basically share one lattice site. In this configuration, the interstitial can be said to be contained in an {011} plane (see figure 1) if the lattice distortions around the defect's core are considered. The defect is, in this sense, two dimensional. The thermal motions of the atoms are such that a {011} plane has a width (two times the standard deviation of the thermal motions) of 5.2% of the nearest-neighbour distance (8.6% of the distance between two adjacent {011} planes), i.e. less than 0.3 Å. Taking the thermal motions of atoms at ideal lattice positions into account, the distortion of the lattice around the defect is

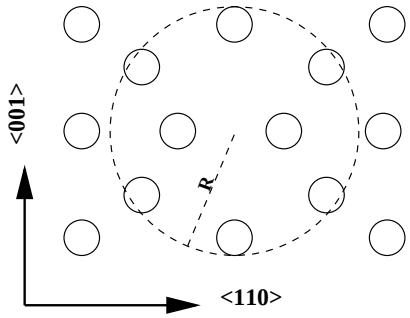

Figure 1. A schematic picture of the dumb-bell configuration. The lattice distortions are contained in the {011} plane, giving the defect a two-dimensional structure. The dashed circle with the radius R, 3.16 Å, is a suggested limit to the extent of the defect.

well contained in the {011} plane, and limited to ± one bcc unit cell in the {011} plane. In W, this means that the 'radius' of the defect within the {011} plane is approximately 3.16 Å. Stoller *et al* [18] have reported in a MD study a dumb-bell configuration along the ⟨111⟩ direction in bcc Fe. Such a configuration was, however, not observed in this study.

3.2. The crowdions

While the dumb-bell can basically be said to consist of two atoms sharing one lattice position, the crowdion is traditionally said to consist of three atoms sharing two lattice positions. This was observed in the present simulation. In addition, a set of configurations where 4, 5, 6 and 7 atoms share 3, 4, 5 and 6 lattice positions, respectively, was observed. These configurations all lie along a ⟨111⟩ direction and are thus similar to each other. The ⟨111⟩ crowdion has therefore in this work been promoted to a whole group of configurations, with the notation $C_{\alpha,\beta}$, where α is the number of atoms and β the number of lattice sites that the atoms share. While the dumb-bell can be considered two dimensional, the crowdion can be considered one dimensional; the distortions induced in the lattice outside the ⟨111⟩ line on which the atoms lie were less than 10% of the width of an {011} plane. The $C_{3/2}$, $C_{4/3}$ and $C_{7/6}$, crowdions are shown schematically (excluding thermal vibrations) in figure 2. In this scheme, the ⟨111⟩ dumb-bell of Stoller *et al* can be classified in a natural way as $C_{2/1}$, since it shares the one-dimensional quality with the other crowdions.

3.3. Defect configuration energies

The defect configuration energies for observed defects, as well as those for the unobserved configuration of $C_{2/1}$, were computed. These energies are presented in table 1, together with values for bcc Fe [18]. Clearly, the ⟨011⟩ dumb-bell has a significantly lower configuration energy than the crowdions. If the lattice is quenched and then allowed to relax, the crowdions collapse into the ⟨011⟩ dumb-bell configuration. Also, for bcc Mo, the ⟨011⟩ dumb-bell has the lowest configuration energy [15]. For the crowdions, the configuration energy has a shallow minimum. The variation in energy between different defects is much larger in W than in Fe, as is to be expected since W is a much stiffer material. Also presented in table 1 is the relative number of times that the interstitial was observed to be in the various defect configurations. The interstitial was found in the ⟨011⟩ dumb-bell configuration for more than half of the simulation which is consistent with the dumb-bell having the lowest configuration energy. However, the interstitial was found in the $C_{3/2}$ and $C_{4/3}$ crowdion configurations close to one third and one

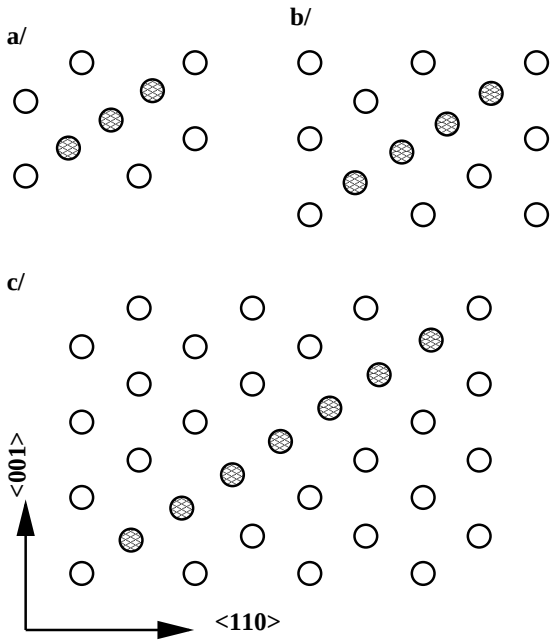

Figure 2. Schematic pictures of some of the crowdions. (a) The traditional crowdion, where three atoms share two lattice positions; (b) the $C_{4/3}$ crowdion; (c) the $C_{7/6}$ crowdion. The interstitial is distributed along a ⟨111⟩ direction (on the grey atoms), with no visible distortions around the line. This gives the defect a one-dimensional quality.

Table 1. The defect configurations and their configuration energies compared to the case of a lattice without defects. The comparison is done at 0 K and zero pressure. $C_{2/1}$ was not observed in the simulations but is included for comparison with the value for bcc Fe. Nor was the $C_{8/7}$ observed but it has been included to establish the trend. The 'occurrence' is the number of times that the defect configuration was observed in the simulation at 2000 K, not the relative time spent in that particular configuration.

Configuration	Occurrence in %	Energy for W (eV)	Energy for bcc Fe (eV)
⟨011⟩ dumb-bell	58.0	1.8	4.76 [18]
$C_{2/1}$	0	8.6	4.87 [18]
$C_{3/2}$	32.1	5.4	4.91 [18]
$C_{4/3}$	8.9	3.5	
$C_{5/4}$	0.6	2.8	
$C_{6/5}$	0.2	2.7	
$C_{7/6}$	0.2	3.0	
$C_{8/7}$	0	3.4	

tenth of the number of times respectively and only at about one per cent of the number of times in any of the larger crowdions. This apparently contradicts the assertion that the crowdion configuration energy was found to have a shallow minimum for the $C_{5/4}$–$C_{6/4}$ crowdions. The measure used—the number of times that the configuration occurs, rather than the time spent in the specific configuration—makes it difficult to evaluate this discrepancy. The evaluation is made even more difficult by the temperature of the simulation, which might also influence the specific details.

4. Discussion and forward look

Our simulation shows that the ⟨011⟩ dumb-bell is the most stable interstitial configuration in W. It also reveals a class of less stable configurations, the ⟨111⟩ crowdions. These configurations are important in themselves since they can be frozen into the lattice if the atoms around the interstitial are not given enough time to rearrange themselves to form the ⟨011⟩ dumb-bell configuration when the structure is cooled. However, they also play a very important part when the interstitial eventually diffuses in the lattice. Preliminary studies shows that the dumb-bell does not move by itself. Instead it transforms into one of the $C_{3/2}$ crowdions which resides in the same {110} plane. The crowdions can transform into each other and/or move along their direction. However, it is only the $C_{3/2}$ crowdion that can transform into the dumb-bell configuration.

This paper has mainly been concerned with the lattice geometry around the interstitial. However, in order to be able to understand how an interstitial recombines with vacancies, forms conglomerates, aggregates at grain boundaries or inner surfaces etc, it is not enough to know the defect structures—one has to know how the interstitial moves in a structure. Future studies should thus be more concerned with how the interstitial moves in the lattice through successive transformations between the dumb-bell and various crowdion defect configurations. For example, Stoller *et al* [18] reported a very high apparent mobility of the $C_{3/2}$ crowdion for Fe. The exact dynamics of those mechanisms should be revealed. The overall interstitial diffusion rate as well as the rate of transition between the various defect configurations should be obtained.

5. Conclusions

It has been shown that EAM MD simulations can be used to study different kinds of defects, e.g. the 2-D dumb-bell and the 1-D crowdions of different sizes, in the bcc metal W. The traditional crowdion, where three atoms share two lattice sites, is found to be part of a family of crowdions. Among the energies of these, there is a minimum for a certain length of the crowdion. Our simulations show that the SIA ⟨011⟩ dumb-bell is the energetically most favourable defect configuration. In this sense, W shows the same behaviour as other bcc metals, e.g. Mo and bcc Fe [15, 18].

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References

[1] Elddrup M and Singh B N 1997 *J. Nucl. Mater.* **251** 132
[2] Puska M J, Sob M, Brauer G and Korhonen T 1994 *Phys. Rev.* B **49** 10 947
[3] Brauer G, Sob M and Puska M J 1992 *Mater. Sci. Forum* **105–110** 611
[4] Brauer G, Anwand W, Nicht E, Coleman P G, Knights A P, Schut H, Kogel G and Wagner N 1995 *J. Phys.: Condens. Matter* **7** 9091
[5] Larsson A K, Withers R L and Stenberg L 1996 *J. Solid State Chem.* **127** 222
[6] Motooka T 1998 *Mater. Sci. Eng.* A **253** 42
[7] Gades H and Urbassek H M 1995 *Phys. Rev.* B **51** 14 559
[8] Carlberg M H, Münger E P and Chirita V 1996 *Phys. Rev.* B 2217

[9] Moško M and Cambel V 1994 *Phys. Rev.* B **50** 8864
[10] Haberland H, Insepov Z, Karrais M, Mall M, Moseler M and Thurner Y 1993 *Nucl. Instrum. Methods* B **80+81** 1320
[11] Fang C C, Prasad V and Jones F 1993 *J. Vac. Sci. Technol.* A **11** 2778
[12] Alfonso D R and Ulloa S E 1993 *Phys. Rev.* B **48** 12 235
[13] Seifert G and Schmidt R 1993 *New J. Chem.* **16** 1145
[14] Daw M and Baskes M I 1984 *Phys. Rev.* B **29** 6443
[15] Wei-Xu and Moriarty J A 1996 *Phys. Rev.* B **54** 6941
[16] Wirth B D, Odette G R, Maroudas D and Lucas G E 1997 *J. Nucl. Mater.* **244** 185
[17] Johnson R A and Oh D J 1989 *J. Mater. Res.* **4** 1195,
[18] Stoller R E, Odette G R and Wirth B D 1997 *J. Nucl. Mater.* **251** 49