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Abstract

Using molecular dynamics simulations with recent interatomic potentials developed for Fe, we have studied the defects in thin films of pure bcc Fe induced by the displacement cascade produced by Fe atoms of 50, 100, and 150 keV impinging under a channeling incident angle of 6º to a [001] direction.

The thin films have a thickness between 40 and 100 nm, to reproduce the thickness of the samples used in transmission electron microscope in-situ measurements during irradiation. In the simulations we focus mostly on the effect of channeling and free surfaces on damage production. The results are compared to bulk cascades. The comparison shows that the primary damage in thin films of pure Fe is quite different from that originated in the volume of the material. The presence of near surfaces can lead to a variety of events that do not occur in bulk collisional cascades, such as the production of craters and the glide of self-interstitial defects to the surface. Additionally, in the range of energies and the incident angle *used, channeling is a predominant effect that significantly reduces damage compared to bulk cascades.*

Keywords: Fe, molecular dynamics, fusion, channeling, ion implantation, surface damage

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1. Introduction

One of the main challenges facing the use of nuclear fusion as a future energy source is related to the reactor materials. In the future fusion reactors, a large amount of 14 MeV neutrons will be produced, which will deteriorate the reactor vessel as they continuously collide with the first wall or plasma facing components. In order to have long-lived nuclear reactors that permit fusion to be an economically competitive energy source, it is necessary to understand radiation damage in the harshest conditions expected for these materials. However, experiments with high energy neutrons are very expensive and there are only a few facilities around the world where they can be performed. In addition, neutrons produce radioactive isotopes in the irradiated material, which complicates its analysis. For these reasons, ion irradiation appears to be an excellent tool for understanding defect production in materials. However, the correlation to neutron irradiation induced damage is not trivial. Therefore, substantial effort is devoted to modeling these effects, mainly using molecular dynamics, and to model-oriented experiments using ion implantation. This would increase our knowledge of the correlation to neutron irradiation and, eventually, it will expand our capacity to predict these effects.

Experiments are currently being performed in JANNuS, France, where while a target is being irradiated with Fe ions it is observed *in-situ* in a transmission electron microscope (TEM). For these experiments to be performed the samples can only be a few tens of nanometers thick to be transparent to electrons. There are many studies of ion irradiation in alpha-Fe using molecular dynamics simulations (see reference [1] for a review), but the majority of these calculations are performed in bulk specimens in view of reproducing neutron damage in reactor materials. The conditions in an ion irradiation experiment are significantly different. On the one hand, for low ion irradiation energies, the damage will occur near the surface. On the other hand, in TEM in situ irradiation, the effect of surfaces cannot be neglected because the specimen thickness is between 40 and 200 nm. The influence of surfaces on the primary damage was studied in detail during the 1990s using molecular dynamics simulations for low energies in f.c.c. materials [2-4]. However, the only studies in Fe including surfaces are those from references [5, 6]. Those calculations were performed for very low ion energies (10-20 keV) and using interatomic potentials that are now known to provide the wrong self-interstitial configuration. Nonetheless, the influence of the surface in terms of damage production and defect distribution was already shown.

Our objective is to further study how damage is produced in these thin films and how it relates to bulk specimens. In this article we study, using molecular dynamics simulations with recent interatomic potentials, the very first stages of the damage produced by Fe ions with energies between 50 and 150 keV in thin films of pure alpha-Fe under channeling conditions.

2. Method

Calculations were performed using the molecular dynamics code MDCASK, developed at Lawrence Livermore National Laboratory , with the interatomic potential of Dudarev and Derlet [7] for alpha-Fe. This interatomic potential was developed to include the magnetic character of Fe by adding a new term in the embedding function of the potential [7]. For short range interactions the potential is connected to the Universal potential as described in [8]. Displacement cascades were simulated injecting an external Fe atom, or primary knock-on atom (pka), in the top free surface of a thin film with a [001] normal. The energy of the incident Fe atom is 50, 100, and 150 keV. The Fe atom incident angle was tilted 6º from normal incidence, lower than the critical angle for channeling which, according to the Lindhard expression [9] is 21º, 15º, and 12º for the 50, 100, and 150 keV respectively. Cell dimensions are 40 nm x 40 nm x 40 nm (140 x 140 x 140 lattice parameters) with a total of 5,154,801 atoms. We used the Lindhard model to include a friction force proportional to the velocity for all atoms with a kinetic energy greater than 5 eV to mimic the inelastic energy losses produced by collisions with the electrons. Periodic boundary conditions were applied to directions perpendicular to that of the incident ion to emulate an infinite system, whereas in the direction of the incident ion free surfaces are considered. The conditions used are intended to reproduce the experiments already published by Yao et al. [10].

The simulations were run for the duration needed for the event to be completed and the number of defects stabilized, which occurs in less than 15 picoseconds in most cases. The temperature was kept near 0K to minimize thermal atomic vibrations and thus facilitate the identification of defects. Before the recoil atom was started, the cell was equilibrated for 1 ps to the desired temperature of the simulation. A thermal bath was imposed to dissipate the energy deposited by the ion in the solid by rescaling the velocity of two atom layers situated at the end of the [100] and [010] directions.

In order to identify the defects, vacancies and interstitials, we used Wigner-Seitz cells centered in each (perfect) lattice position so that an empty cell corresponds to a vacant and a double occupied cell corresponds to an interstitial defect. Variability was introduced to obtain statistical results by changing the value for the impact angle orientation from 0º to 180º for up to a total of 20 cases for the 50 and 100 keV pka, and 30 cases for the 150 keV pka.

3. Results and Discussion

Results are presented here and compared to the results by C. Björkas for bulk cascades with energies of 50 keV and 100 keV [11].

Figure 1 represents the final number of defects (vacancies and interstitials) that results for the series of 20 simulations of irradiation of thin films with a pka of 100 keV. It appears that the dispersion in the number of defects is much wider in cascades affected by free surfaces than in cascades of the same energy produced in the bulk of the material [11]. In the latter case, the mean value of Frenkel pairs produced is 159 with a standard deviation of 7 [11]. In our case, we observe, on the one hand, that the average number of vacancies does not match that of interstitials, with 52 vacancies and 34 self-interstitials. On the other hand, the standard deviation for vacancies is 17 and for interstitials is 11. This is in agreement with the results of Stoller et al. [5, 6] that show both a higher production of vacancies than self-interstitials as well as larger standard deviations in their results for surface damage. It is interesting to note that recent simulations of cascades close to grain boundaries by Bai et al. also show a larger production of vacancies than interstitials close to the grain boundary [12]. In that case, with the use of temperature accelerated dynamics (TAD), the authors show how at later times self-interstitials trapped at grain boundaries can annihilate those remaining vacancies in the bulk.

The higher number of vacancies is mainly the result of the migration of interstitials to the surface. Differences in the energy spatial deposition account for the dispersion in the results, with some cases in which the energy is deposited very close to the surface, creating surface damage [2], and other cases in which damage is deeper, being more similar to the results obtained in bulk cascades.

Figure 2 shows three snapshots of the damage produced by a 150 keV Fe ion in a 40 nm thick Fe sample. Dark dots show the locations of self-interstitials ions while light dots are the locations of vacancies. Figure 2(a) shows the distribution of defects at 0.06 ps, before the ion reached the back surface. Figure 2(b) shows the damage at 0.5 ps which corresponds to the time of maximum number of displacements in the lattice, that is the peak of the collision cascade. Much of the damage is recovered during the next stage, reaching an almost constant number of defects. The final distribution at 10 ps is shown in Figure 2(c). Note that the damage produced in the top and bottom surfaces is mostly vacancies since the interstitials are ejected to the surface and stay as adatoms (not shown in the figure), again because of the creation of surface damage. It should be noted that in this case the pka traveled through the sample and escaped through the bottom surface, producing little damage, which shows that the impact angle used (6º) is within channeling conditions. Therefore the damage produced consists of Frenkel pairs and small vacancy and self-interstitial clusters. In all cases studied in these conditions there was transmission of the pka. We have analyzed the damage produced as a function of the distance to the front and back surfaces. For the case of Figure 2(c), the total number of vacancies produced in this cascade is 137 while the total number of self-interstitials produced is 95. The total number of vacancies at a distance of 3 nm from either the back or the front surface is 56, while only 6 interstitials are found at this distance. Below 3 nm we then find 89 self-interstitials and 81 vacancies, that is, similar values for the two types of defects, getting closer to bulk calculations.

Comparing the different energies studied, we observe some trends. The maximum production of defects occurs when events very near the surface take place, leading to the formation of surface damage either at the top or the bottom surface. On the other hand, in the cases with the minimum number of defects, events very near the surface are not important and the damage created is more similar to that produced in bulk material.

The results for our simulations in thin films and in bulk cascades [11] are summarized in Table 1. Figure 3 graphically represents these values. Unlike the results of Stoller et al. [5, 6], the average number of defects is significantly lower in our simulations than in the case of bulk cascades. This is due to the channeling conditions used in the calculations, which allow the ion to travel through the whole sample thickness without depositing all its energy in the target. The total energy deposited by the ion can be calculated from the difference between the initial energy and the energy of the ion after it crosses the film. On average, the percentage of the energy deposited for the 50keV, 100keV, and 150keV cascades is 44%, 32%, and 29% respectively. However, even under these conditions, where little damage should be produced, there are cases where a high number of vacancies is created, as can be seen in Figure 1. These are related to cases where surface damage is formed in the front or back surfaces, or in both.

4. Conclusions

In this paper we have studied damage produced by Fe ions with energies between 50 and 150 keV in thin films of pure bcc Fe using molecular dynamics simulations under channeling conditions. The results are also compared with those obtained in bulk cascades. Conclusions can be summarized as follows:

1. Unlike cascades simulated in the bulk that produce the same number of vacancies and interstitials, cascades in thin films simulated by external ion irradiation produce more vacancies than interstitials, even under channeling conditions such as those studied here.
2. Dispersion in the number of defects in ion irradiated thin films is greater than in simulations in bulk materials due to the variety of events that can occur because of the surface influence, and this dispersion increases with the pka energy.

These conclusions show that to be able to reproduce and understand ion irradiation experiments in thin films analyzed by TEM, a detailed description of the primary damage is needed. This implies the inclusion of surfaces in the simulations because the damage produced is completely different from that originated in bulk materials. The time scale in MD simulations is too short to be able to make a direct comparison with TEM experimental measurements. Kinetic Monte Carlo (kMC) simulations using the information obtained from these MD calculations could be used to make such comparison. If the initial damage distribution has an impact on microstructure evolution under irradiation, as shown in previous works, the concentration of defects with dose as well as the cluster size distribution obtained by kMC should be different when using bulk cascades or surface cascades. These results could then be contrasted with the experimentally measured values.

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Table 1 Average number of stable defects and their standard deviation produced in thin films and bulk cascades.

Energy (keV)	**Thin films vacancies**	**Standard deviation**	**Thin films interstitials**	**Standard deviation**	**Bulk[a] Frenkel pairs**	**Standard deviation**
50	47	8	32	7	91	5
100	52	17	34	11	159	7
150	73	28	46	14	-	-

[a] Reference 11

M.J. Aliaga, One column.

Figure captions

Fig. 1 Defect distribution for a series of 20 simulations of cascades in thin films produced by irradiation with a 100 keV Fe atom.

Fig. 2 (Color online) Three snapshots of the time evolution of the damage produced by a 150 keV Fe ion in a 40 nm thick Fe sample. Light dots are vacancies and dark dots are self-interstitials. a) 0.06 ps after the initiation of the recoil, b) 0.5 ps, and c) 10 ps.

Fig. 3 (Color online) Average number of defects produced in thin films (vacancies and interstitials) compared to bulk cascades (Frenkel pairs) [11].

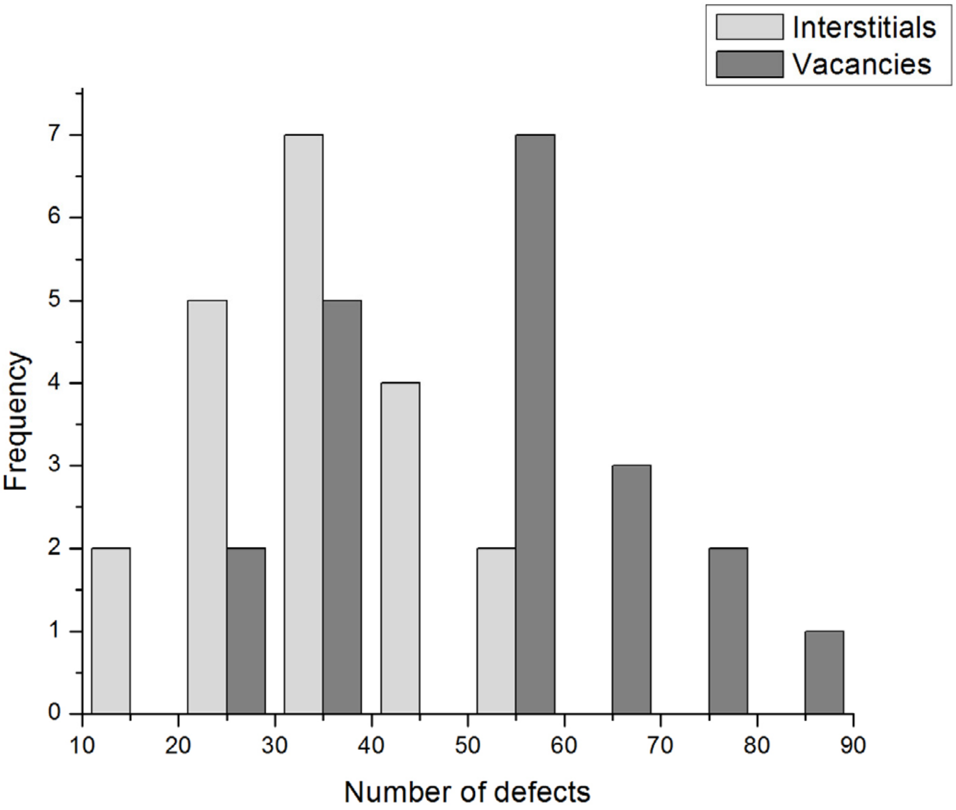
Interstitials
Vacancies
Frequency
0
1
2
3
4
5
6
7
10
20
30
40
50
60
70
80
90
Number of defects

Figure 1

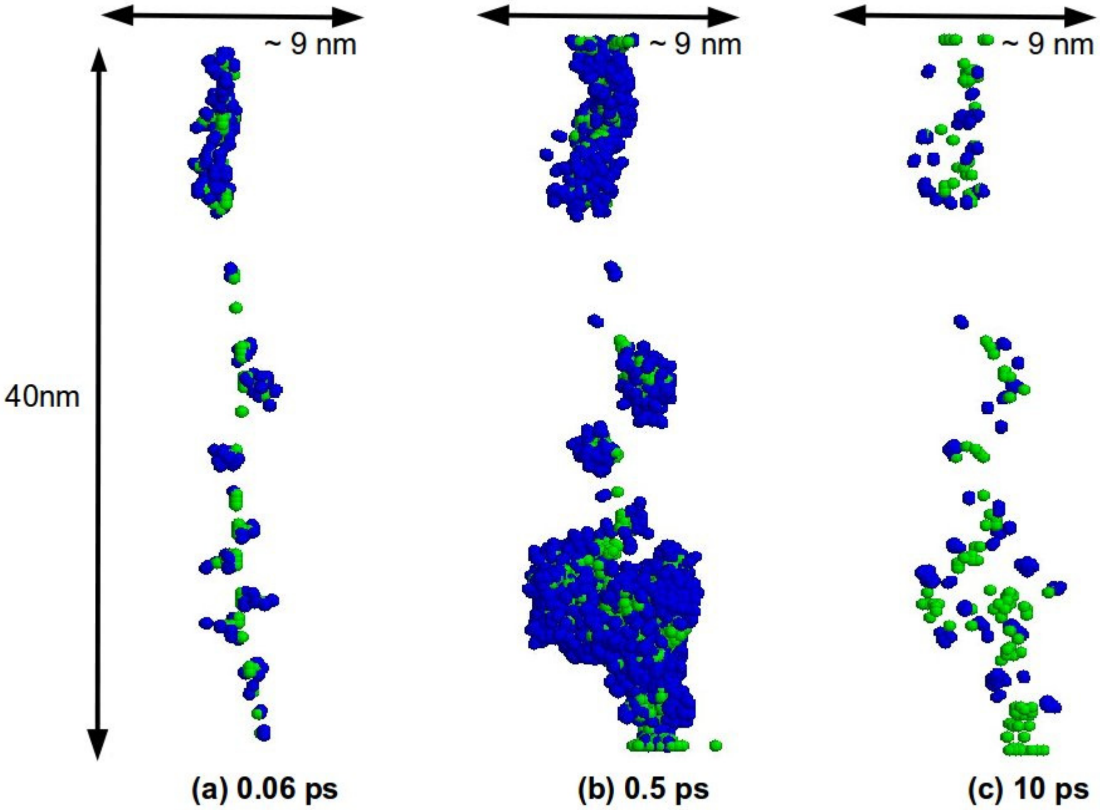

Figure 2

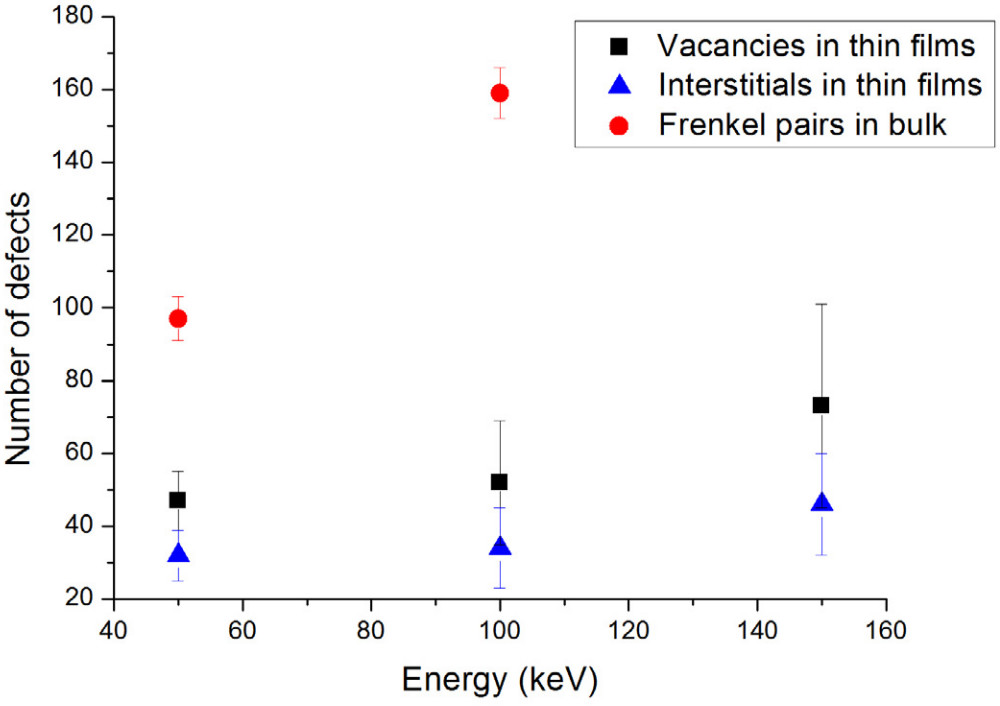
Number of defects
180
160
140
120
100
80
60
40
20
40
60
80
100
120
140
160
Energy (keV)
■ Vacancies in thin films
▲ Interstitials in thin films
● Frenkel pairs in bulk

Figure 3