

INFLUENCE OF PKA DIRECTION, FREE SURFACES, AND PRE-EXISTING DEFECTS ON CASCADE DAMAGE FORMATION—R. E. Stoller, S. G. Guiriec (Oak Ridge National Laboratory)

OBJECTIVE

The objective of this work is characterize primary damage formation in irradiated structural materials, including the impact of all the various irradiation, material, and experimental conditions that may influence defect formation.

SUMMARY

Primary cascade damage production in iron has been extensively investigated by molecular dynamics, and average defect production parameters, such as the total number of stable point defects, in-cascade defect clustering fractions, and in-cascade cluster size distributions have been derived. However, preliminary results indicated several factors could alter “normal” cascade evolution and lead to quite different defect production behavior. Further investigations of three such factors have been carried out: (1) primary knock-on atom (PKA) direction, (2) nearby free surfaces, and (3) pre-existing effects. Results of the investigation confirm these factors can significantly impact cascade damage formation. The effects include enhanced defect survival for PKA directions that lie in close-packed {110} planes, increased point defect clustering and larger defect clusters for cascades initiated near a surface, and reduced defect survival in simulation cells containing defects. The origin and implications of these effects are discussed relative to the interpretation of certain experimental observations and parameters used in other modeling studies.

PROGRESS AND STATUS

Introduction

The use of molecular dynamics (MD) simulations to investigate the evolution of atomic displacement cascades has provided a detailed understanding of primary radiation damage formation in irradiated materials [1-10]. In particular, increased computational capability has enabled this method to be used to obtain a statistically meaningful cascade database for iron [1,5]. Representative values have been derived for several measures of primary damage, including: the number of stable point defects (interstitials and vacancies) created, the number of these stable defects that cluster directly during the cascade event, and the size distribution of these in-cascade defect clusters. For convenience when comparing different materials and cascade conditions, the number of displacements obtained from the standard NRT model [11,12] is often used as a normalizing factor for the number of defects produced and in clusters. This convention is followed in the present work.

Although the iron cascade database in the literature is relatively large, a number of factors have been identified that can cause cascade evolution, and hence defect production, to deviate from the average behavior exhibited in the database. Three of these factors are: PKA direction [3,13], presence of a nearby surface [8,9,14], and pre-existing defects (such as cascade debris) in the simulation cell [15,16]. In some cases, the results just referenced exhibited rather significant differences from the typical MD cascade that is carried out in an atomic simulation cell that contains perfectly crystalline material. However, in other cases the results were either ambiguous, limited in their parameter range, or an insufficient number of simulations were done to establish statistical significance. Since displacement cascades are stochastic events, the quantitative impact of any cascade variable can only be determined by a systematic study with “enough” events to capture inherent statistical variations in their behavior [1,5]. The objective of the current investigation is to establish the degree to which these three factors influence cascade evolution and defect formation by carrying out additional simulations and extending the range of the previous work to higher PKA energies and temperatures. Since the methods and models used have been discussed in detail in previous publications, [1-5,13,14] only a brief description is included here.

Application of MD Simulation Method

MD simulations were carried out using the code MOLDY and a modified version of the Finnis-Sinclair potential [3,17,18]. Periodic boundary conditions were imposed on a constant pressure ensemble of atoms. The simulation cell size varied from 16,000 atoms for the low-energy investigations of PKA direction effects, to 250,000 atoms for the 10 and 20 keV simulations employed to evaluate the impact of free surfaces and pre-existing damage. The iron cascade database mentioned above served as the basis for comparison with the new simulations [1,5].

Results of MD Simulations

Influence of PKA Direction

A fairly dramatic effect of PKA direction was observed at low cascade energies during an earlier analysis of the complete cascade database [1, 13]. In order to avoid lattice effects such as channeling and directions with particularly low or high displacement thresholds, MD cascade simulations are typically carried out using a high index PKA direction. For example, most of the simulations in the database discussed in Ref. [1] were generated using a [135] PKA direction. A preliminary evaluation of PKA direction effects using 1 keV cascades indicated that mean values obtained with [135] PKA should be representative of the average behavior at this energy [4]. However, analysis of a large number of 300 eV simulations found that stable defect production was significantly above the average when the PKA direction lay in the close-packed {110} planes. For example, some cascades initiated with a [114] developed completely within a single plane. The planar channeling effect resulted in greater separation of vacancies and interstitials and therefore less in-cascade recombination.

It was expected that this effect would not persist to higher energies where the higher kinetic energies would tend to kick recoils out of the {110} plane. However, when the energy dependence of this enhanced defect formation was explored using simulations at 100K, and it was somewhat surprisingly found that the increased defect survival was detected at energies as high as 2 keV. The ratio of the defect survival using [114] to that using [135] was 1.59, 1.12, 1.28, 1.15, and 0.995 for cascade energies of 0.3, 0.5, 1.0, 2.0 and 5.0 keV, respectively [13]. In order to obtain further information on the potential for this phenomenon to influence defect production, additional simulations have been carried out at 600 and 900K to see if increased atomic thermal motion would act to minimize the planar channeling.

Some of the results of 600 and 900K simulations are shown in Figs. 1a and 1b, respectively. This figure compares stable defect formation as a function of energy for the [114] and [135] PKA directions, where the error bars in the figure are the calculated standard errors of the mean values. The error bars are somewhat larger than in the 100K comparisons shown in Refs. [1 and 13] because the number of cascades at each condition is smaller. However, it seems clear that there is no systematic deviation between the two PKA directions at either temperature. Even at 300 eV, where the effect was strongest in 100K cascades, the impact is small at 600K. Additional simulations are underway to improve the statistical comparison in Fig. 1, and to provide data for comparison at additional energies.

Influence of Free Surfaces

A previous publication describes the motivation for concern about the influence of a nearby free surface on cascade evolution, and results obtained from a series of 10 keV cascades were presented [14]. The primary interest is to provide an explanation for the lower yield of visible defects from MD cascades simulations as compared to the results of thin foil ion irradiations that are carried out in a transmission electron microscope. The 10 keV simulations exhibited a clear effect on both total stable defect production and in-cascade clustering for cascades initiated at or near a free surface [14]. However, the largest vacancy and interstitial clusters formed were still too small to be observed by TEM.

Therefore, additional simulations have been carried out using 20 keV cascades, an energy where point defect clustering is increased and larger clusters have been observed in the existing database. The new simulations were carried out using the same method discussed in Ref. [14]. A free surface was created by

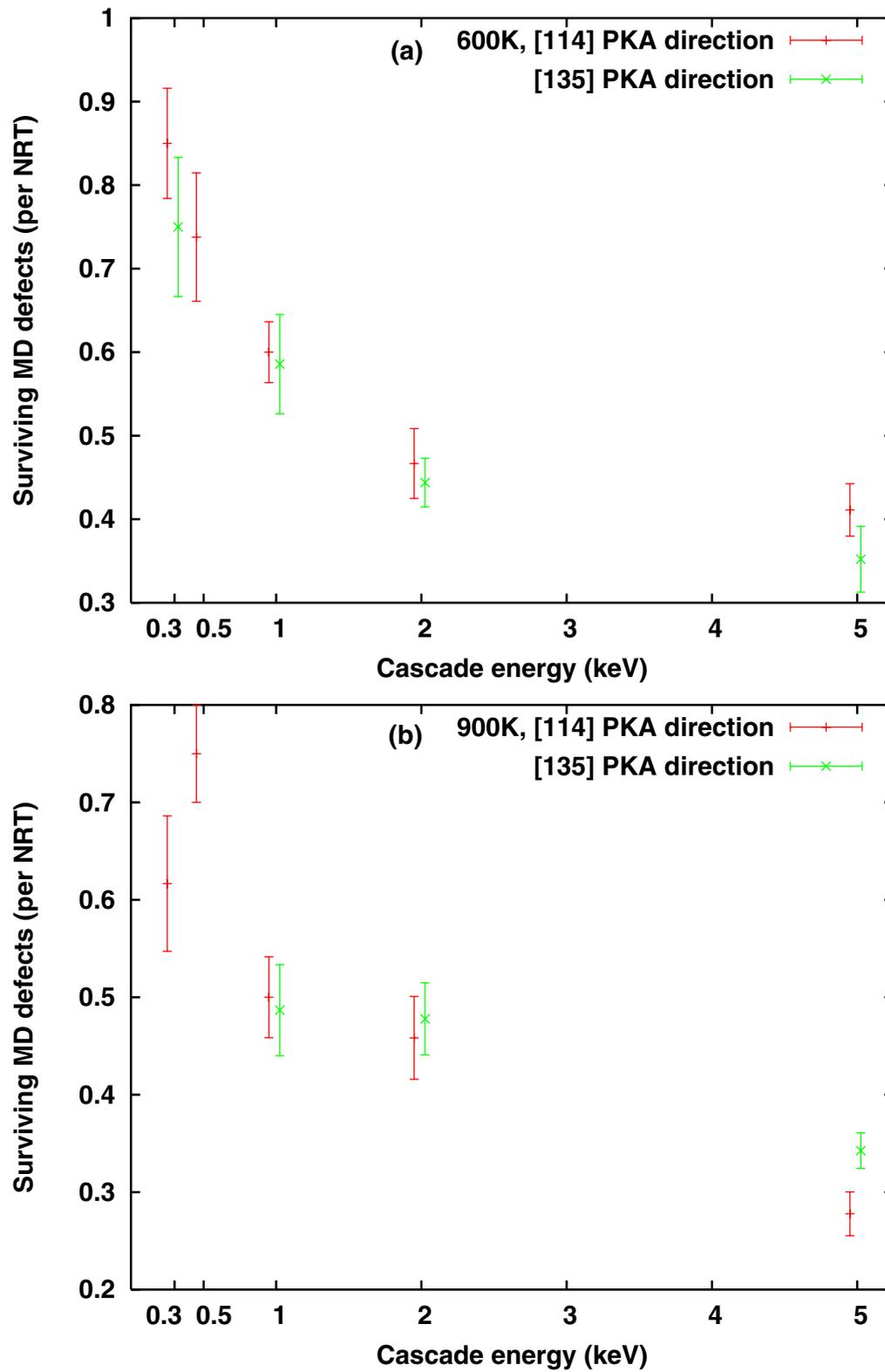


Fig. 1. Energy dependence of the PKA direction effect on stable point defect formation at (a) 600K and (b) 900K.

removing 5 layers of atoms from one surface of a $(50a_0)^3$ atom cell, containing 250,000 atom sites. Periodic boundary conditions are otherwise imposed. Eight simulations were carried out at 100K in which all the PKAs selected were surface atoms. Several PKA directions were used, with each of these directions slightly more than 10° off the [001] surface normal. The results of these simulations can be compared with the 10 “bulk” cascades conducted previously in which cascades were initiated near the simulation cell center.

The number of surviving point defects (normalized to NRT displacements) is shown in Fig. 2a for both the bulk and surface cascades, with error bars indicating the standard error of the mean. The results are similar to those observed at 10 keV. The average number of stable interstitials produced by the surface cascades is not significantly different than for the bulk cascades, with the mean value shifted slightly higher for the 20 keV surface cascades, whereas it was slightly lower for the 10 keV case. However, there is a substantial increase in the number of stable vacancies produced, and the change is clearly significant. As discussed previously, the number of surviving interstitials and vacancies is no longer equal for cascades initiated at the surface. Interstitials can be lost to the surface by two mechanisms; sputtering or the diffusion of interstitials and small glissile interstitial clusters to the surface. Reducing the number of interstitials leads to a greater number of vacancies surviving since less recombination can take place.

The results for in-cascade clustering shown in Fig. 2b are also similar to the 10 keV simulations, i.e. no significant change in the fraction of interstitials in clusters ($\sim 0.18/\text{NRT}$) while the in-cascade vacancy fraction (based on the 4th-nearest-neighbor criterion [4]) increased from $\sim 0.06/\text{NRT}$ to $0.25/\text{NRT}$. Moreover, the vacancy cluster size distribution changed dramatically, with larger clusters produced than were observed at 10 keV. The size distributions obtained for bulk and surface cascades are shown in Fig. 3a and b for vacancy and interstitial clusters, respectively. The largest vacancy cluster observed in the bulk cascades contained only six vacancies, while the surface cascades had clusters as large as 21 vacancies. This latter size is near the limit of visibility in TEM, with a diameter of almost 15 nm. The change in the interstitial cluster size distribution was less dramatic.

Influence of Pre-existing Damage

A cascade energy of 10 keV and a temperature of 100K was chosen for this study to expand the range of previous work carried out using 1 keV simulations in copper [15] and 0.40, 2.0, and 5.0 keV simulations in iron [16]. The 10 keV cascade energy is high enough for some in-cascade clustering to occur, is near the plateau region of the defect survival curve, and initiates a limited degree of subcascade formation. For these conditions, the database contains two independent sets of cascades, 7 in a 128k atom cell and 8 in a 250k atom cell that provide a basis of comparison. A cell size of 250k atoms was used for the cascade simulations with pre-existing damage.

Simulations were carried out to investigate three different types of pre-existing damage. The first involved the as-quenched debris from a 10 keV cascade in perfect crystal. A total of 30 vacancies and interstitials were present, including one di-interstitial and one 7-interstitial cluster. The second case was similar, but the vacancies were rearranged to obtain a 6-vacancy void and a 9-vacancy loop. Some additional interstitial clustering occurred when the modified configuration was equilibrated, so that it contained four di-, one tri-, and one 8-interstitial cluster. This starting configuration is shown in Fig. 4. The third configuration contained only a single 30-vacancy void. Eight simulations were carried out starting with different initial PKAs and a $\langle 135 \rangle$ PKA direction. The same set of PKAs were used for all three defect configurations, with the PKA location such that it was aimed at the center of the defect field.

The influence of pre-existing damage on the number of stable defects formed in 10 keV cascades is shown in Fig. 5a, where results from the three defect configurations are compared with those obtained in perfect crystal. Results for the fraction of interstitials in clusters are shown in Fig. 5b. The variation between two sets of perfect crystal simulations is provided for comparison. Both defect survival and interstitial clustering results are summarized in Table 1. As expected, substantial variation was observed between the different simulations for any given pre-existing defect configuration, but a significant reduction in the average defect formation was observed for the two configurations most typical of cascade debris. A slight increase (that

may not be statistically significant) in stable defect production was observed when an otherwise perfect crystal contained a small void. Only the second defect configuration (Fig. 4) lead to a significant change in interstitial clustering.

Table 1. Summary of defect production results from cascades with pre-existing damage

	survival fraction (per NRT)	standard error	interstitial cluster fraction (per NRT)	standard error
Perfect crystal (128k and 250k atoms)	0.336	0.0137	0.170	0.0155
Defective crystal				
30 i,v: cascade debris, with 1 di- and 1 7-interstitial cluster	0.260	0.0214	0.179	0.0119
30 i,v: cascade debris with 4 di-, 1 tri-, and 1 8-interstitial cluster; 6-vacancy void, 9-vacancy loop	0.279	0.0258	0.110	0.0191
30-vacancy void only	0.370	0.0288	0.190	0.0188

Discussion and Conclusions

A fairly complete picture of displacement cascade evolution in bulk, perfect crystal iron has been obtained through extensive simulation studies during the past several years. The work reported here is intended to illustrate the impact of three cascade conditions that deviate from the average behavior displayed in the overall cascade database. These are: PKA direction effects, the influence of free surfaces, and the impact of pre-existing defects or cascade debris. In each case, the results reported here extends the range of previous work by the current authors and others.

Higher cascade energies and temperatures have been used to investigate the trends obtained at 100 K and lower energies. The intent is to determine the overall significance of these effects to the average defect production parameters that are used in the damage source terms of kinetic microstructural models. The influence of a planar channeling effect was observed in low energy cascades (up to 2 keV) at 100K for PKA directions that lie in the close-packed {110} planes in bcc iron. This phenomenon has now been investigated in MD simulations up to 5 keV at temperatures up to 900K. Although dramatic differences are observed between cascades that exhibit planar channeling and “normal,” more isotropic cascades, the overall impact of PKA direction on average defect formation in any practical sense appears to be limited. The maximum energy where an effect was seen at 100K was 2 keV, and at 600 and 900K essentially no effect was observed even in 300 eV cascades.

In the case of surface-influenced defect formation, a sufficient number of 20 keV iron cascades were completed to statistically evaluate variations between these and bulk cascades. Relative to cascades initiated far from the free surface, stable vacancy defect production increased for surface-influenced cascades, while the number of surviving interstitials was nearly unchanged. The difference between the number of surviving vacancies and interstitials arises from sputtering and surface absorption of mobile interstitial defects. The fraction of vacancies contained in clusters increased substantially for cascades initiated at the surface, and larger vacancy clusters were formed. The impact on defect clustering increases with cascade energy, and are reaching the limit of TEM visibility in the 20 keV simulations reported here. These results have implications for interpreting experiments in which cascade defect production is observed *in-situ* using transmission electron microscopy

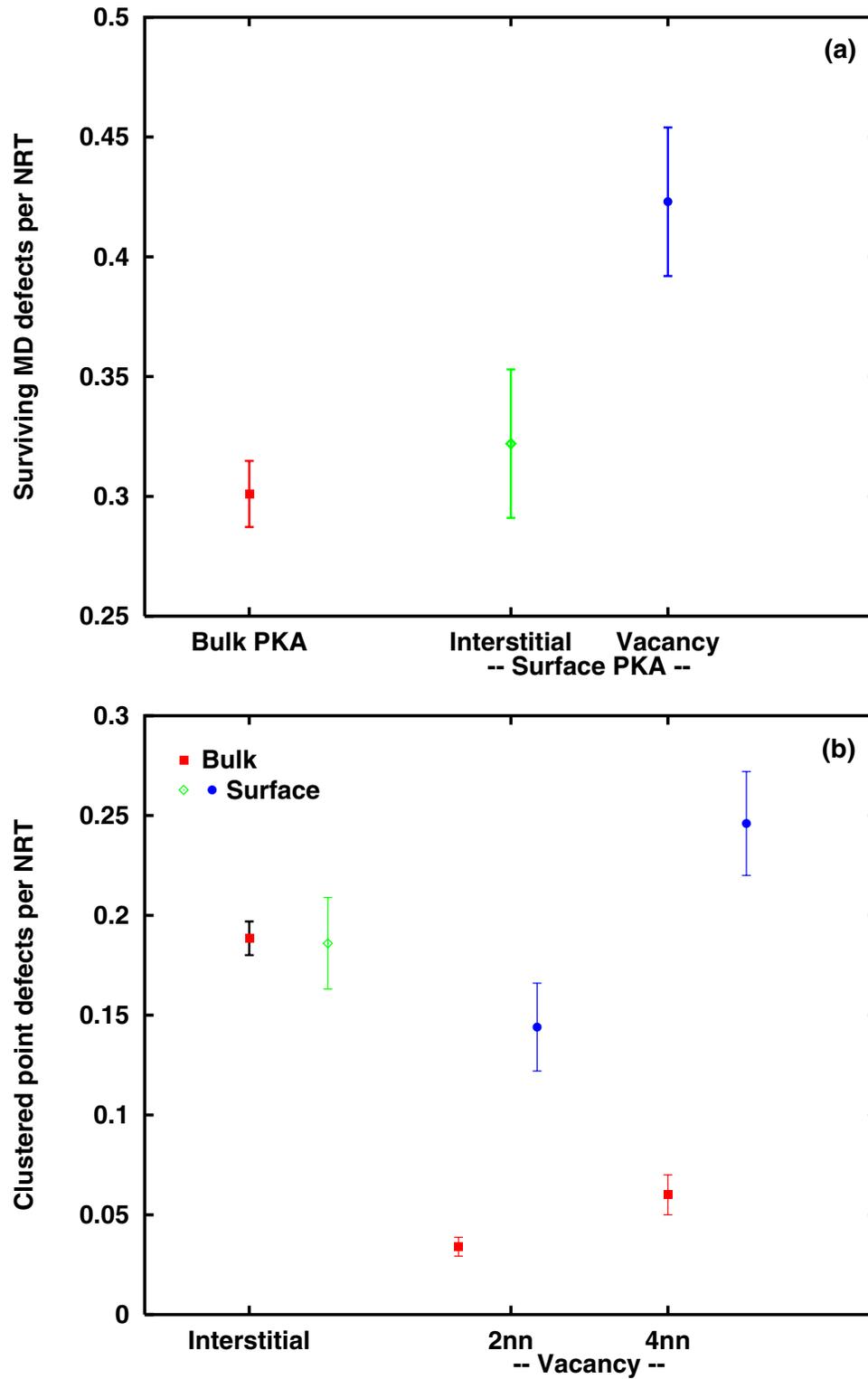


Fig. 2. Comparison of defect production parameters obtained in 20 keV cascades at 100K for cascades initiated by a PKA near the center (bulk) and at the free surface of the simulation cell: (a) average stable defect production (per NRT) and (b) clustered point defect fraction (per NRT).

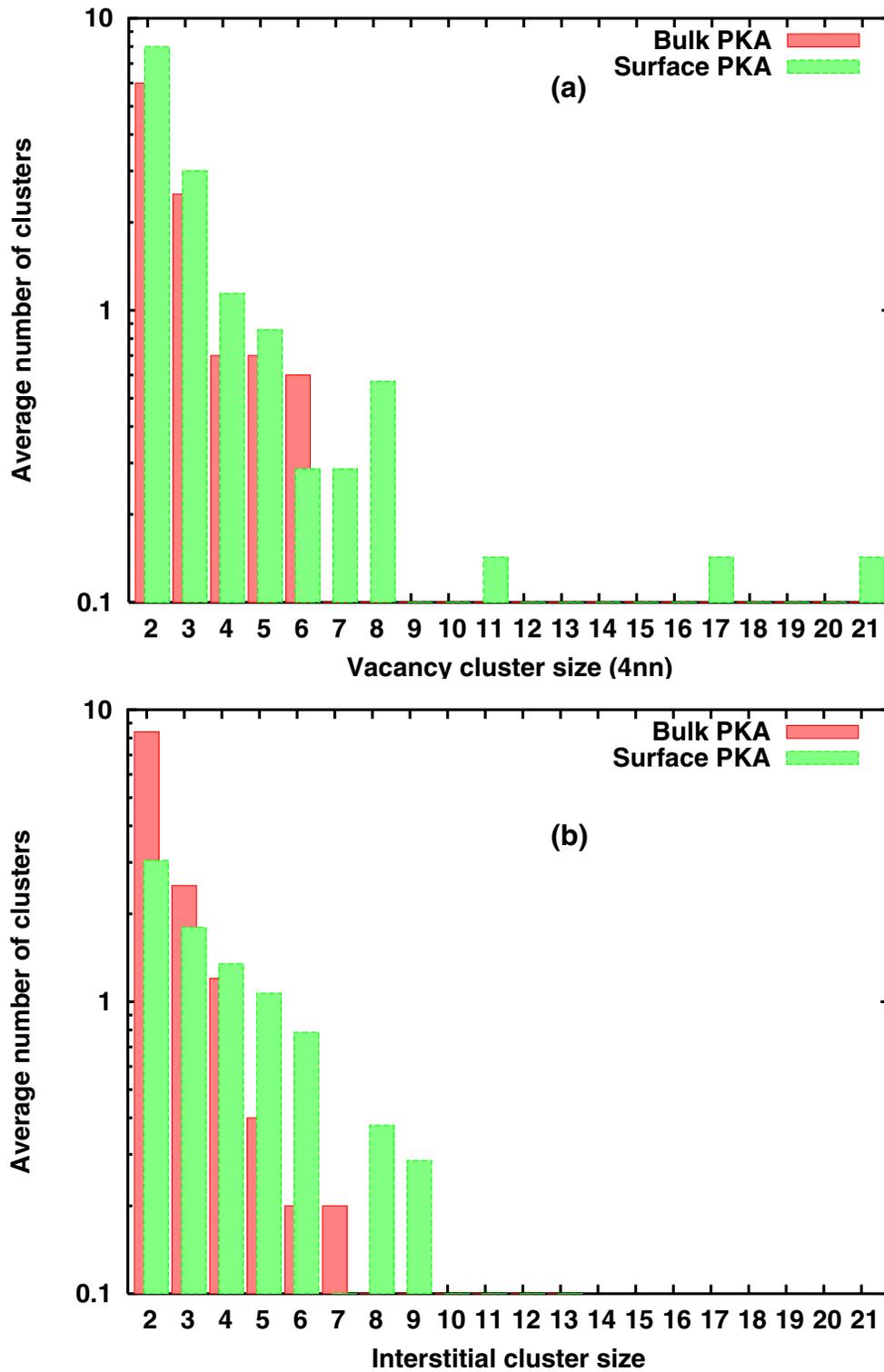


Fig. 3. Comparison of in-cascade point cluster size distributions in 20 keV, 100K cascades initiated by a PKA near the center (bulk) and at the free surface of the simulation cell: (a) vacancy clusters and (b) interstitial clusters.

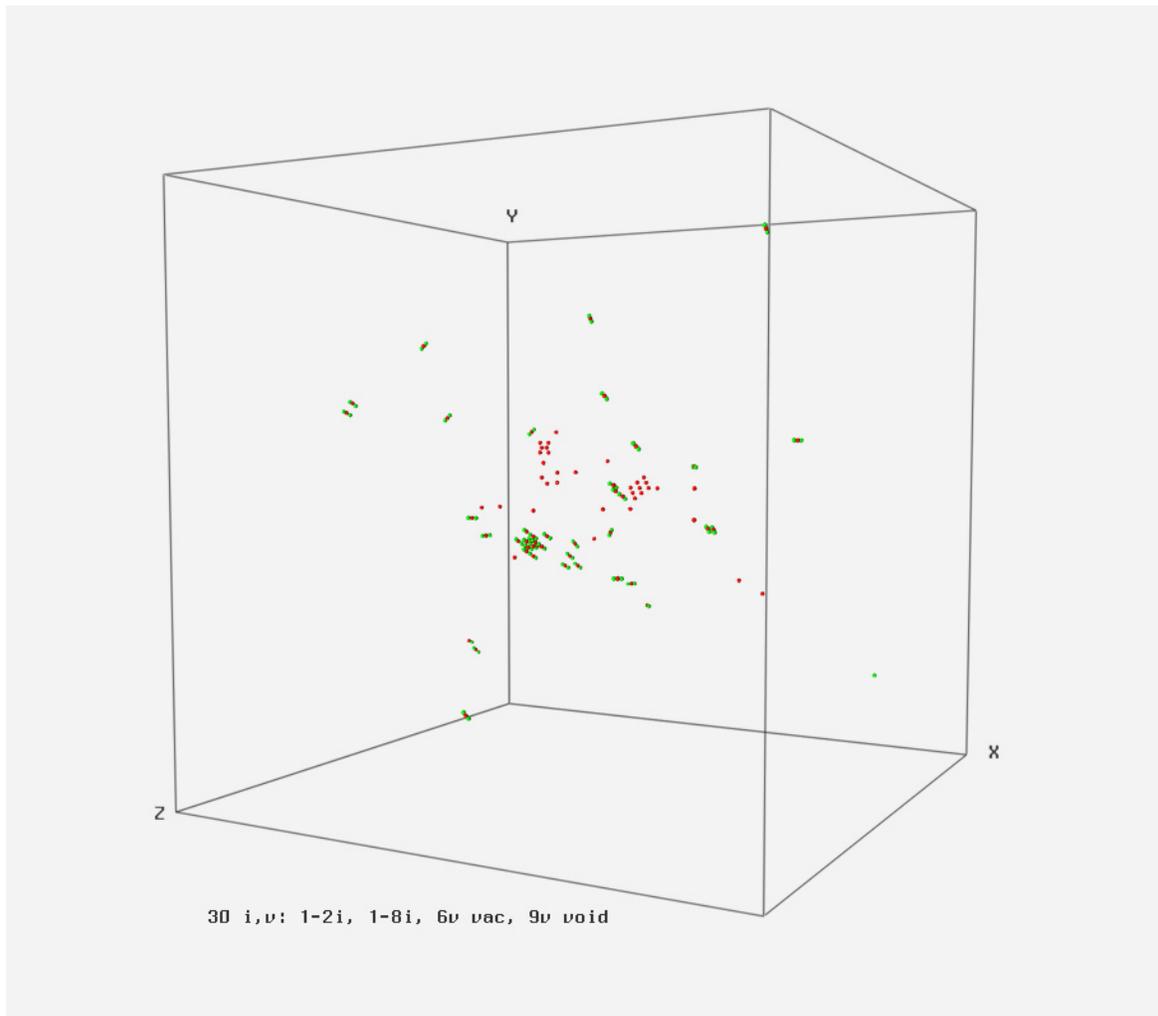


Fig. 4. Second pre-existing defect configuration for 10 keV displacement cascades (see text).

Although the approach in this investigation of pre-existing damage effects was slightly different, the results are generally consistent with the previous studies by English, et al. [15] and Gao, et al. [16]. In both cases, the authors observed substantial reductions in defect production when a cascade was initiated in material containing defects. The reductions in defect production observed in this study (Fig. 5 and Table 1) are somewhat smaller. This difference may partially be due to the higher cascade energy employed here (10 keV vs. 0.4 to 5 keV); but the statistical nature of cascade damage production is also a factor. A number of cascades were carried out in the work of Gao, et al., and the results were analyzed as a function of the distance between the center of mass (COM) of the new cascade and that of the pre-existing damage. A good correlation was found between this spacing and the number of defects produced, and their results provide a trend based on individual cascades as a function of COM distance. In the work reported here, the distance between the cascade origin and the pre-existing damage was nearly the same for all of the simulations. However, the morphology developed in each of the 8 cascades was quite different, so the COM spacings also varied. The average behavior for a fixed initial separation can not be directly compared to the earlier results. Because of the reduced defect survival observed in defective material, these results and the earlier work suggest that the possibility of developing a fluence-dependent cascade survival efficiency for use in kinetic radiation damage models should be investigated.

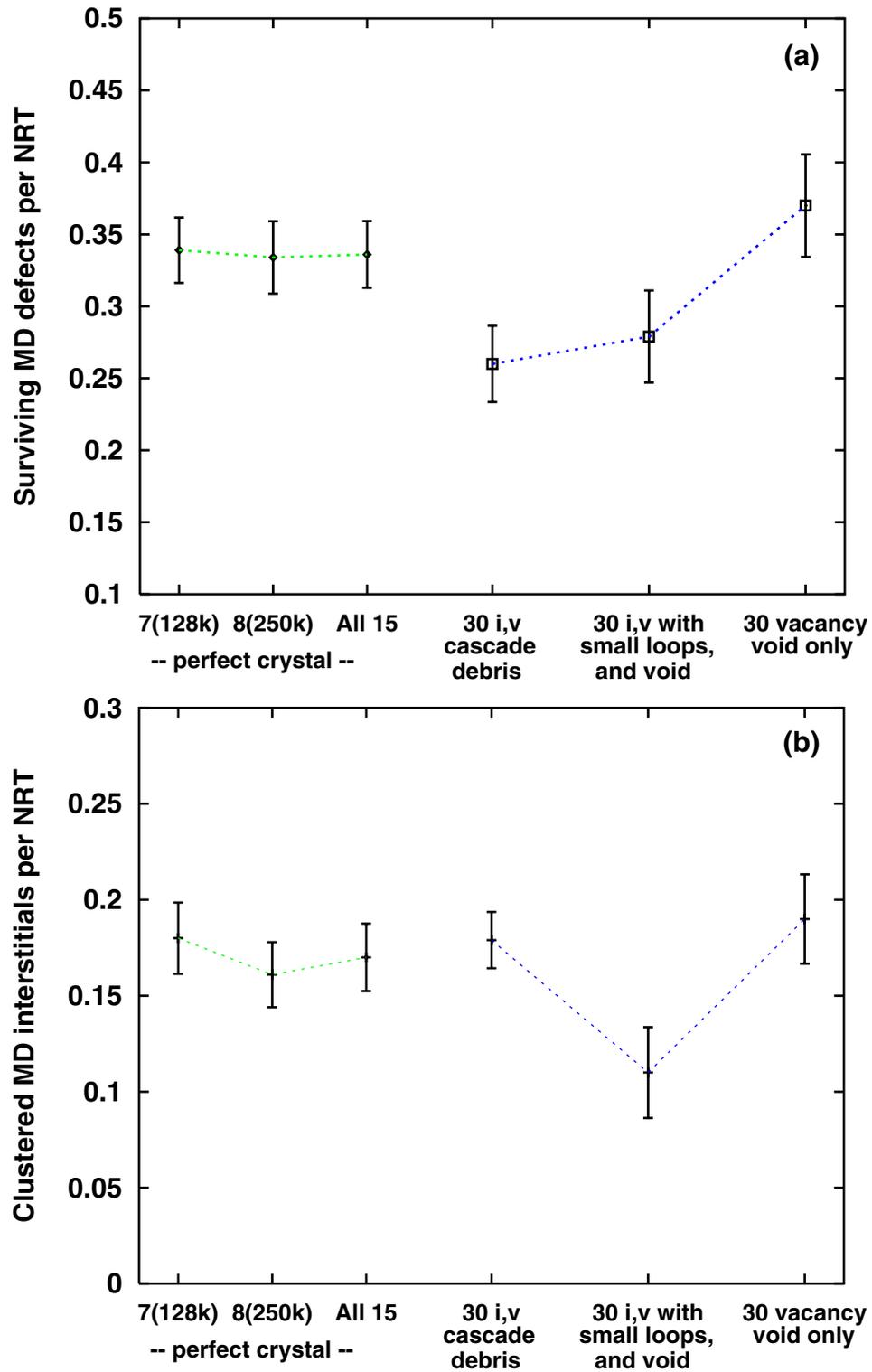


Fig. 5. Comparison of defect formation parameters in perfect and defective simulation cells: (a) total point survival and (b) interstitial clustering.

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References

- [1] R. E. Stoller and A. F. Calder, *J. Nucl. Mater.* 283-287 (2000) 746-752.
- [2] W. J. Phythian, R. E. Stoller, A. J. E. Foreman, A. F. Calder, and D. J. Bacon, *J. Nucl. Mater.* 223 (1995) 245.
- [3] A. F. Calder and D. J. Bacon, *J. Nucl. Mater.* 207 (1993) 25-45.
- [4] R. E. Stoller, G. R. Odette, and B. D. Wirth, *J. Nucl. Mater.* 251 (1997) 49-69.
- [5] R. E. Stoller, *J. Nucl. Mater.* 276 (2000) 22-32.
- [6] D. J. Bacon, F. Gao, and Y. N. Osetsky, *J. Nucl. Mater.* 276 (2000) 1-12.
- [7] M. J. Caturla, N. Soneda, E. Alonso, B. D. Wirth, T. Diaz de la Rubia, and J. M. Perlado, *J. Nucl. Mater.* 276 (2000) 13-21.
- [8] K. Nordlund and R. S. Averback, *J. Nucl. Mater.* 276 (2000) 194-201.
- [9] M. Ghaly and R. S. Averback, *Phys. Rev. Lett.* 72 (1994) 364.
- [10] K. Nordlund, J. Keinonen, M. Ghaly, and R. S. Averback, *Nature* 398 (1999) 48.
- [11] M. J. Norgett, M. T. Robinson, and I. M. Torrens, *Nucl. Engr. and Design* 33 (1975) 50-54.
- [12] ASTM E521, Standard Practice for Neutron Radiation Damage Simulation by Charged-Particle Irradiation, Annual Book of ASTM Standards, Vol. 12.02, American Society of Testing and Materials, West Conshohocken, PA.
- [13] R. E. Stoller, The Influence of PKA Direction on Displacement Cascade Evolution, Microstructural Processes in Irradiated Materials, Eds. G. E. Lucas, L. L. Snead, M. A. Kirk, and R. G. Elliman, Materials Research Society, Pittsburgh, PA, 650 (2001) 3.5.1-3.5.6.
- [14] R. E. Stoller, *J. Nucl. Mater.* 307-311 (2002) 935-940.
- [15] A. J. E. Foreman, W. J. Phythian, and C. A. English, private communication, unpublished.
- [16] F. Gao, D. J. Bacon, A. F. Calder, P. E. J. Flewitt, and T. A. Lewis, *J. Nucl. Mater.* 276 (1996) 47-56.
- [17] M. W. Finnis, MOLDY6-A Molecular Dynamics Program for Simulation of Pure Metals, AERE R-13182, UKAEA Harwell Laboratory, Harwell, UK, 1988.
- [18] M. W. Finnis and J. E. Sinclair, *Phil. Mag. A* 50 (1984) 45-55, and Erratum, *Phil. Mag. A* 53 (1986) 161.