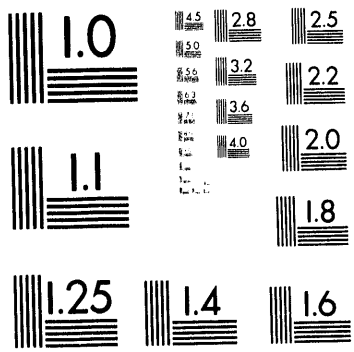
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MODELING DEFECT PRODUCTION IN HIGH ENERGY COLLISION CASCADES[†]

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ABSTRACT

A multi-model approach (MMA) to simulating defect production processes at the atomic scale is described that incorporates molecular dynamics (MD), binary collision approximation (BCA) calculations and stochastic annealing simulations. The central hypothesis of the MMA is that the simple, fast computer codes capable of simulating large numbers of high energy cascades (e.g., BCA codes) can be made to yield the correct defect configurations when their parameters are calibrated using the results of the more physically realistic MD simulations. The calibration procedure is investigated using results of MD simulations of 25 keV cascades in copper. The configurations of point defects are extracted from the MD cascade simulations at the end of the collisional phase, similar to the information obtained with a binary collision model. The MD collisional phase defect configurations are used as input to the ALSOME annealing simulation code, and values of the ALSOME quenching parameters are determined that yield the best fit to the post-quenching defect configurations of the MD simulations.

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MODELING DEFECT PRODUCTION IN HIGH ENERGY COLLISION CASCADES

Introduction

The processes of radiation damage, from the initial production of defects to the evolution of the microstructure, occur over a wide range of time and size scales. The initial displacement-producing collisions take place in the first few tenths of a picosecond, producing nanometer-scale damage regions. The thermal spike develops and dissipates within ten picoseconds, and defect interactions continue in the cascade region for tens of nanoseconds. Beyond this "primary damage state," long range migration of the mobile defects causes microstructure evolution on a scale well beyond the atomic level. A single theoretical model does not exist that can encompass all elements in this range and lead to a quantitative understanding of the damage process. A good description of the primary damage state at the atomic scale can be obtained, in principle, using molecular dynamics (MD) techniques. However, the present computational limits of MD effectively prevent it from being used for simulating large numbers of high energy cascades or for following the motion of defects by thermally activated diffusion. However, an atomic-level description of the primary damage state, through the short-term annealing stage, can be obtained using the proposed multi-model approach (MMA).

The central hypothesis of the MMA is that simple, fast computer codes capable of simulating large numbers of high energy cascades and containing the essence of the physical aspects being modeled, such as binary collision approximation

(BCA) calculations and stochastic annealing simulations (SAS), can be structured and calibrated to yield the correct defect configurations using the results of the computationally intense but more physically realistic MD simulations. The BCA code MARLOWE[1] simulates the collisional stage only, and it has been used to investigate the morphology of large numbers of high energy cascades[2]. The SAS code ALSOME simulates the diffusion and interaction of defects, treating the defects as interacting entities and ignoring the interactions of individual atoms. In earlier work, summarized in Ref. [3], the collisional stages of cascades were simulated with MARLOWE, and the defect distributions were followed through the short-term annealing stage with ALSOME. The considerable effects of the thermal spike were accounted for by using "quenching" values for the ALSOME parameters, determined empirically by requiring total and freely migrating defect yields to be similar to "measured" defect yields. Of course, defect production from individual cascades cannot be measured directly. Experimental defect yields, which are averages over time and volume, are inferred from measurements of diffusion-driven phenomena. These inferences are drawn on the basis of assumptions regarding defect production which may not necessarily be appropriate[4]. Although the freely migrating defect yields from individual cascades obtained in the early work compare favorably with recent MD simulations[5], the MMA will be more useful if it is based on a realistic description of the behavior of defects in individual cascades.

In the present work, MARLOWE and ALSOME are employed as in the earlier work, except that the quenching parameters are determined by comparison with MD results. The calibration is performed using results of MD simulations for

cascades in copper of 25 keV, which is near the threshold energy for subcascade formation. The configurations of point defects are extracted from output of the MOLDYCASK[6] MD cascade simulations at the end of the collisional stage, obtaining distributions of vacancies and displaced atoms similar to those obtained with the BCA code MARLOWE. The MD collisional stage defect configurations are used as input to ALSOME, and values of the ALSOME quenching parameters are determined that yield the best fit to the post-quenching defect configurations of the MD simulations. Thus, both the initial collisional stage configuration and the final configuration of the quenching stage come from the same MD cascade. These quenching parameter values are then used in ALSOME when applied to high energy MARLOWE BCA cascades.

Calculations

Defect Distributions in the Collisional Phase. For the quenching scheme to be useful, it is crucial that the collisional stage information extracted from MD output have vacancy-interstitial pair separations similar to those obtained in the BCA cascades. Collisional phase information was extracted from the output of the MOLDYCASK code for the two 25 kev cascades generated in copper at 10 K [7]. As in MARLOWE, the atoms in MOLDYCASK receiving kinetic energy greater than a selected threshold energy ECUT (4.8 eV) are identified and followed over many time steps until their energies fall below a cut-off value. Their initial lattice sites are considered to be vacant sites, and the positions of the atoms where their energies fall below the cut-off are taken as the locations of the displaced atoms. In MARLOWE the cut-off energy has the same value as the threshold energy ECUT, and each displaced atom is located at the

turning point of its last collision. In the MOLDYCASK output, each displaced atom identified is located at the position occupied during the last recorded time step in which it had energy greater than the cut-off energy. To maintain the same average vacancy-displaced atom pair separation distributions as MARLOWE, it was necessary to follow displaced atoms in the MOLDYCASK cascades until their kinetic energy dropped well below ECUT (2 eV). It should be noted that the collisional stage configurations from MOLDYCASK have somewhat fewer defect pairs, somewhat smaller volumes, and somewhat higher vacancy densities than the average values for 100 MARLOWE cascades, but the MOLDYCASK values are within one standard deviation of the MARLOWE distributions.

Calibrating ALSOME. ALSOME is based on a simple model of defect diffusion and interaction. Point defects and defect clusters are considered to be spherical and centered on lattice sites, with radii related to their cluster size. Mobile defects, including clusters of any size if desired, move one lattice site at a time in randomly chosen directions. The order in which the defects are chosen to move is determined by random selection, depending on the number of mobile defects and their assigned relative jump probabilities. Two defects interact when their separation falls within the assigned critical reaction distance for that interaction. The interaction product is placed at the target defect's lattice site. The adjustable parameters are the defect species (type and size) that are considered mobile (e.g., mono-, di- and tri-vacancies; mono-, di-, tri- and tetra-interstitials for normal annealing in this work), the relative jump probabilities for each of the mobile species, and the critical reaction distances. The annealing is modeled as a sequence

of events rather than as a function of elapsed real time, and it continues until no mobile defects remain in the volume being annealed, unless some other criterion is imposed. When simulating the short-term annealing in a single cascade region, the cascade is placed at the center of a volume that is somewhat larger than the cascade in all directions. During annealing, defects escaping this volume are no longer followed, and they are considered to be migrating defects.

ALSOME was designed to simulate normal, thermally activated diffusion, and it is most physically correct when applied to relatively dilute concentrations of defects. It is the aim of this work to determine if ALSOME can be effectively applied to the quenching stage of high energy cascades. To be sure, the actions of defects within a thermal spike (if they could be defined during the short-lived, extremely non-equilibrium excursion above the melting temperature) cannot be simulated using the assumptions of ALSOME. However, it may be possible to simulate the results of those actions, i.e., the distribution of remaining defects, by choosing ALSOME parameter values for the quenching stage that result in the required amount of clustering and annihilation in the cascade region. The anticipated success of this simple approach derives from the strong influence of the collisional stage defect configuration on the configuration after quenching.

The MD simulations show that during the quenching of a thermal spike, the vacancy distribution becomes concentrated toward the center of the cascade, and interstitial atoms are largely eliminated from the spike region. This is illustrated in Table 1, where the size of a defect distribution is represented

by the root mean square distance of defects from the centroid. The sizes of the vacancy and displaced atom distributions for the two 25 keV MOLDYCASK cascades are about the same after the collisional stage. After the quenching stage, the displaced atom distributions are about the same size as before, but the vacancy distributions are about half their previous size.

The collisional phase was extracted from the MOLDYCASK cascades in the form of a distribution of vacant sites and displaced atoms. There was no attempt to define stable defects or clusters of defects, since the cascade energy still resides in the significant kinetic energies of a relatively small number of atoms at that point. Following development and quenching of the thermal spike (at 11-12 ps, when the MOLDYCASK runs were terminated), point defects were identified as vacancies or self-interstitial atoms (SIAs) in terms of the occupation of Wigner-Seitz cells. As the list of point defects is read into ALSOME, defect clusters are immediately identified according to the clustering criteria represented by the critical reaction distances. Figure 1 shows the distribution of vacancy clusters at 12 ps, after the quenching stage (open bins). Both large clusters and single point defects are present after the quench. This is true in the SIA distribution as well.

The ALSOME quench was performed on the extracted collisional stage configurations in Monte Carlo fashion, running many times over different sets of random numbers with each set of trial parameter values. The criteria for a successful simulation of the quenching stage of the MOLDYCASK cascades are that 1) the correct number (about 60) of total point defect pairs should remain after the quench, 2) the vacancy distribution should collapse to about

half its collisional stage size, and 3) the cluster size distributions should be similar to those of the quenched configuration, especially in having large clusters as well as individual point defects.

Unfortunately, no set of parameter values was found that could simultaneously satisfy all three criteria. In particular, it was not possible to reduce the size of the vacancy distribution without eliminating the population of mobile vacancies through clustering. Subsequently, a revised set of criteria were adopted that involved comparing the results of short term annealing on the quenched cascades remaining at the end of the MOLDYCASK runs to the results after quenching and short term annealing of the collisional stage configurations extracted from MOLDYCASK. The criteria for selecting parameter values in this case are simply that the total number of remaining defect pairs should be the same, and the numbers of vacancies and SIAs escaping the cascade should be the same, respectively. The short-term annealing is carried out at an effective temperature and time such that no mobile defects remain in the cascade region.

In Table 1 the sizes of the vacancy and SIA distributions after quenching and short term annealing with ALSOME are listed. Compared with the MD standard, the interstitial distributions are slightly smaller, while the vacancy distributions are almost twice the size of the standard. The vacancy cluster size distribution after quenching is in Fig. 1, and the SIA cluster distribution after short term annealing, starting from both the quenched standard and the collisional stage, are shown in Fig. 2. After short term annealing the small mobile clusters are gone, either clustering, annihilating

or escaping. The large SIA clusters obtained from annealing the MD standard were not produced when starting from the collisional stage. This is also true for vacancy clusters.

A set of 100 cascades of 25 keV in copper was produced with MARLOWE and run through the ALSOME quenching and short term annealing. The average defect production from those cascades is listed in Table 2. The total number of defects at each stage agrees well with the MOLDYCASK cascades, but there are twice as many freely migrating vacancies and 50% more freely migrating SIAs on average resulting from the MARLOWE cascades. As described above, the collisional stage configurations of the two MOLDYCASK cascades are smaller and more compact than the average 25 keV MARLOWE cascade. The closer proximity of the defects in the compact MOLDYCASK cascades evidently results in the greater clustering observed.

Discussion

The ALSOME annealing code was unable to simulate all the features of the quenched MOLDYCASK cascade defect distributions using only the simple adjustment of its defect jumping and interaction parameters. It was, however, quite successful in simulating the proper yields of total and freely migrating defects after short term annealing.

ALSOME, as used, did not produce the compaction of the vacancy distribution during quenching that is observed in the MD results. This is understandable, since, after the collisional stage, a highly disordered (molten) region

develops in which the actions of point defects cannot be recognized. This thermal spike then rapidly quenches to the crystal temperature. To model that behavior in terms of only interacting defects is probably impossible. On the other hand, the quenched configuration exists only for a short time. Defect interactions continue to take place in the cascade region long after the spike has cooled.

The large clusters produced during the quenching stage in the MOLDYCASK cascades, and which continue to exist even after short term annealing, were not produced as frequently in quenching and short term annealing with ALSOME. This may be due primarily to the simplicity of the model used in ALSOME, in which all defect migration is by uncorrelated jumps. A region of high local defect density that collapses to a large cluster may form two or more smaller clusters under ALSOME, depending on the statistical variation of the uncorrelated random jumps, which would tend to favor the production of multiple clusters in such a case. With respect to defect production, this may not be a serious limitation. The same total number of defects is tied up in clusters, and two closely spaced medium-sized clusters may have about the same effect on subsequent defect disposition as a single large cluster.

An important cascade feature that ALSOME is presently incapable of simulating is the direct formation of both vacancy and interstitial loops in cascades[7]. The 25 keV cascades used in this work contain a loop of 14 vacancies in a single (111) plane and a loop of 17 SIAs, also on a single (111) plane.

Conclusions

The MMA, utilizing MOLDYCASK, MARLOWE and ALSOME, provides a feasible approach to modeling the production of defects in high energy cascades. Quenching parameter values were determined for ALSOME by calibration to results from 25 keV MD cascades in copper produced with MOLDYCASK. Cascades at the collisional stage produced using MARLOWE, then quenched and short term annealed using ALSOME, give defect yields consistent with cascades generated in MOLDYCASK MD simulations.

The ALSOME simulation in its simplest form cannot duplicate the compactness of the vacancy distribution or the large clusters or loops formed in the MD simulations. This limitation may be important when cascade and subcascade interactions or vacancy cluster dissociation are involved. However, present computing capabilities now allow the rules governing migration and interaction of defects in ALSOME to be much more complex. Continuing work will develop those rules from the actions of defects observed in detail in MD simulations.

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TABLE 1
Average Radius of Defect Distributions in 25 keV Cascades in Cu
(values in Angstrom units)

	VACANCY	INTERSTITIAL
COLLISIONAL STAGE		
CASCADE A	29	32
CASCADE B	28	31
AFTER QUENCHING*		
MOLDYCASK	19	40
MMA	33	38
AFTER SHORT-TERM ANNEALING*		
MOLDYCASK	13	32
MMA	26	28

* Average of Cascades A and B

TABLE 2
Defect Production in 25 keV Cascades in Cu

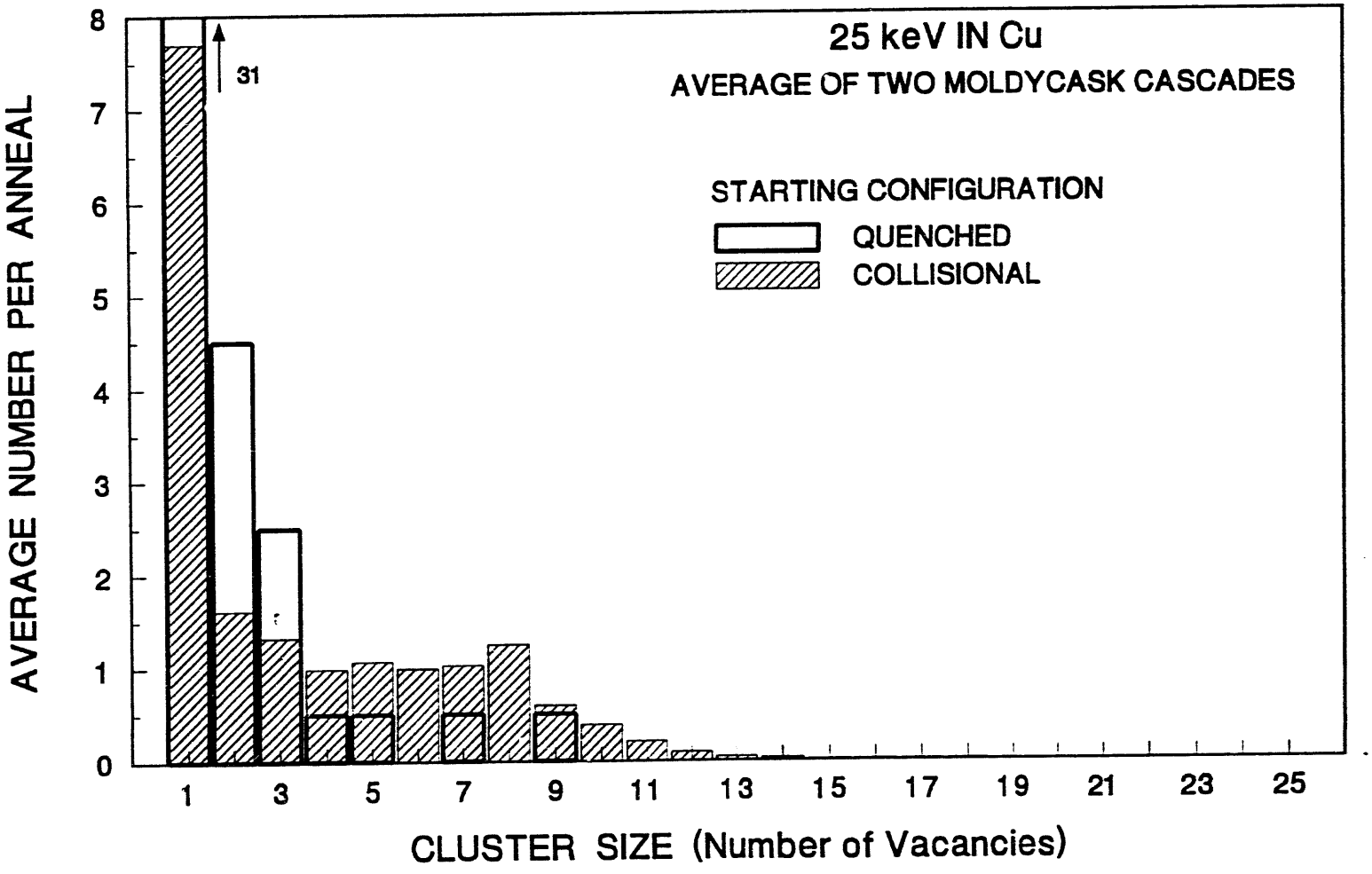
STARTING CONFIGURATION	TOTAL DEFECT PAIRS QUENCHED	ANNEALED	FREELY MIGRATING VACANCIES	SIAS
QUENCHING STAGE	60	49 ± 4	8 ± 4	9 ± 3
COLLISIONAL STAGE	61 ± 10	48 ± 7	8 ± 3	9 ± 4
MARLOWE, 100 CASCADES	65 ± 15	45 ± 11	16 ± 6	15 ± 5

FIGURE CAPTIONS

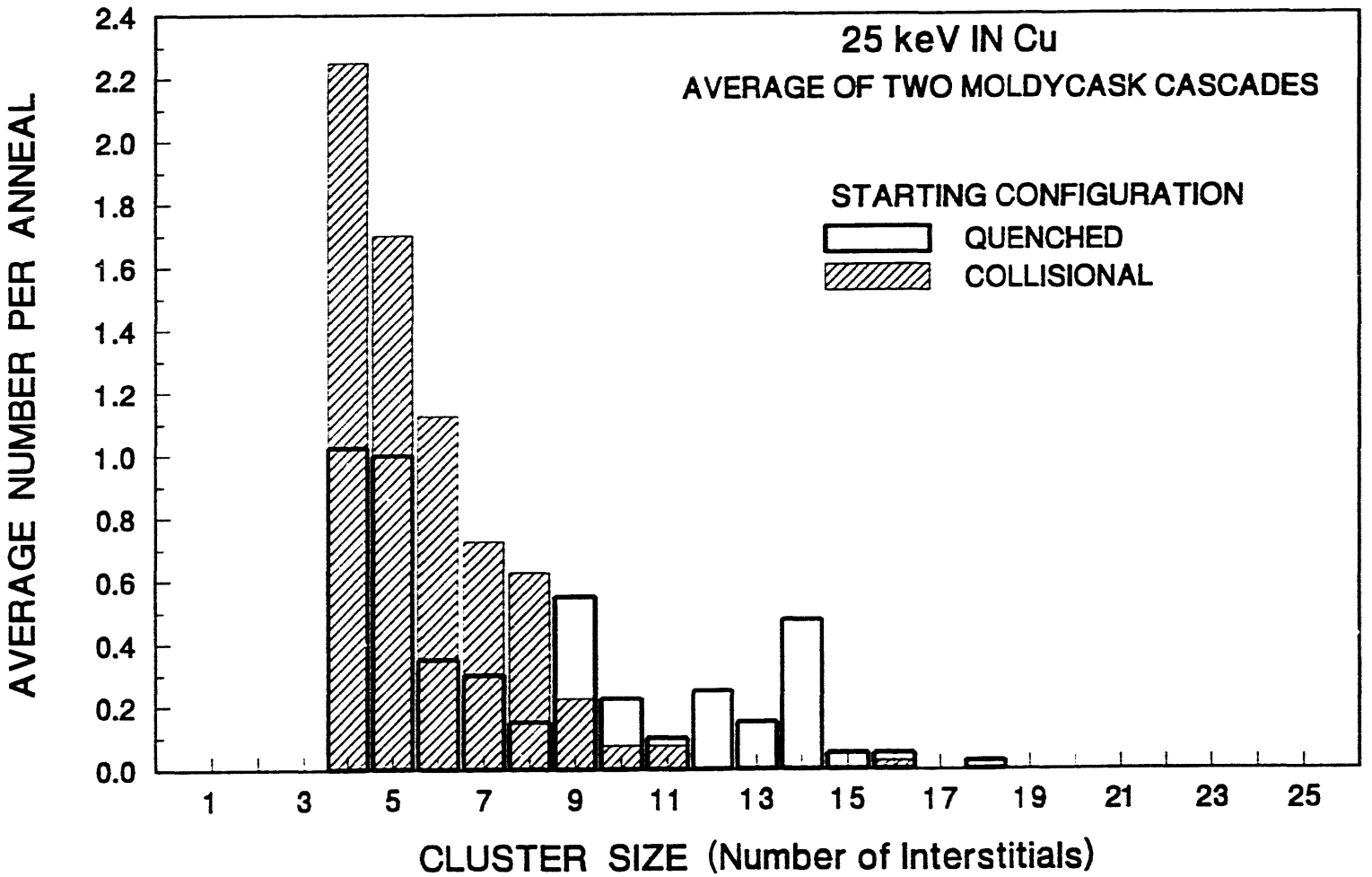
Figure 1. Vacancy cluster size distributions at the end of the thermal spike quenching stage for 25 keV cascades in copper. The open bars are for the quenched configuration directly from MOLDYCASK and the striped bars are for the quenched configuration as simulated with ALSOME, starting from the collisional stage in MOLDYCASK.

Figure 2. Interstitial cluster size distributions after short-term annealing for a time and temperature at which single vacancies diffuse from the cascade region, for 25 keV cascades in copper. The open bars are for the configuration when starting from the quenched cascades from MOLDYCASK and the striped bars are for the configuration as simulated with ALSOME, starting from the collisional stage in MOLDYCASK.

VACANCY CLUSTER SIZE DISTRIBUTIONS
AFTER QUENCHING STAGE



INTERSTITIAL CLUSTER SIZE DISTRIBUTIONS
AFTER SHORT-TERM ANNEALING



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