Computer simulation studies of high-energy collision cascades *

Mark T. Robinson

Solid State Division, Oak Ridge National Laboratory, P.O. Box 2008, Oak Ridge, TN 37831-6032, USA

A modified binary-collision-approximation algorithm allowing the proper order of the collisions in time was used to study cascades in Cu and Au at primary kinetic energies up to 100 keV. Nonlinearities were approximated by letting already-stopped cascade atoms become targets in later collisions, using an improved method of locating potential targets to extend the calculations to energies much higher than heretofore. Beside the effect of the approximate nonlinearity, the effect of thermal disorder in the targets was examined. Target redisplacements reduce the damage in Cu by 3% at most, but in Au they reduce it by amounts up to 20% at 100 keV. Thermal disorder is also important: by disrupting crystal effects, the damage is reduced significantly.

1. Introduction

Computer simulation in the binary collision approximation has long been used to study the development of collision cascades in crystals. Previous work with the MARLOWE program [1] showed how such collisions may be ordered properly in time and how nonlinear effects in cascade development may be approximated by letting already-stopped cascade atoms become targets in later collisions, but the work was limited to initial kinetic energies in the low kilovolt region by a need for very frequent searches of the entire list of atoms in the developing cascades. Because of this limitation, the effects of nonlinearities, though clearly evident, were modest and there were incentives for further study. This paper sketches recent changes in the MARLOWE model, including a new algorithm for locating potential targets and other refinements, allowing calculations to be extended to initial kinetic energies of 100 keV or more in both Cu and Au.

2. The computational model

Most aspects of the MARLOWE model are described in detail elsewhere [1–4]. Potential nonlattice (stopped cascade atom) targets used to be found by scanning a table of cascade particles exhaustively at every collision: since the table length and the number of times it was scanned were each roughly proportional to the initial kinetic energy E_0, the time for this task

varied as E_0^2 and dominated the computation above a few keV. A better procedure uses the fact that a nearby lattice site is associated with each atom in a MARLOWE cascade. A so-called "hashing" algorithm is used to find such sites in the same way used for vacant ones: the lattice vector to a site defines a location in a table. If the site is vacant or associated with a stopped nonlattice atom, a pointer to the site or atom occupies the place indicated; otherwise, it is marked as empty. Separate tables are maintained for vacancies and atoms. With large enough tables and with provisions for conflicts between different sites for places in them, most searching is eliminated, the program execution time is proportional to E_0, and high-energy cascade calculations are not inhibited.

Projectiles in MARLOWE are followed as long as their kinetic energies exceed a value E_c. To be displaced, lattice targets must surmount a binding energy E_b if the projectile continues moving, or E_b' if it stops, and still have kinetic energy $> E_c$. Originally, a nonlattice target receiving kinetic energy in a collision was always followed and no binding energy was used. To be redisplaced in the revised model, a nonlattice target must surmount an energy E_b'' and still have kinetic energy $> E_c$. If redisplacements do not occur, encounters with nonlattice targets are classed as subthreshold events: some atoms once regarded as redisplaced [1] are now in this group. The residual kinetic energy of a nonlattice target when it stops is ignored in determining its new kinetic energy.

Records are now kept of the times at which redisplaced atoms initially stop as well as of the times at which atoms in motion in a cascade. Records are kept of the energy transferred in redisplacement events and in subthreshold encounters with nonlattice atoms. For the

present work, a special analysis evaluated projectile kinetic energy spectra at intervals during cascade development.

Cascades were generated in Cu and Au for E_0 from 0.1 to 100 keV. Up to 20 keV, samples of 1000 cascades were generated; 500 were evaluated at 50 keV and 250 at 100 keV. Quantities averaged over sets of cascades are enclosed in $\langle \ \rangle$. In each case, $E_b' = E_b'' = 0.1E_b$, $E_b = E_c$, and $E_c = 3.50$ eV in Cu and 3.81 eV in Au. The fcc lattice constant a_0 was 0.3615 nm in Cu and 0.4078 nm in Au. The Molière potential was used with screening lengths of 7.38 pm in Cu and 7.50 pm in Au. Collisions with impact parameters $< 0.62a_0$ were evaluated. "Local" inelastic energy losses [3] were scaled to the LSS model [5]. Thermal disorder in the targets was described by the Debye model [6] with $\theta_D = 315$ K in Cu and 175 K in Au. Calculations with redisplacements were made for static targets and for temperatures of 0, 300, and 1000 K. Static calculations were also made without redisplacements. The collisions were time-ordered with a time interval of 1 fs. Calculations were carried out on a Data General AViiON™ 300 workstation and required up to several hours per case.

3. The temporal development of cascades

The initial kinetic energy of the primary recoil is dissipated partly in exciting target electrons, partly in subthreshold encounters in which the target atoms receive kinetic energy $< E_c$, and partly in encounters

producing new or redisplaced recoils. Fig. 1a shows $\langle E_k(t) \rangle$ for 20 keV cascades in Au, where $E_k(t)$ is the kinetic energy of the moving recoils at time t. When redisplacements are allowed in a static target, $\langle E_k(t) \rangle$ is unaffected below about 0.18 ps, but then falls faster than otherwise. In both static calculations, there is a pronounced tail extending to long times, which represents the slow decrease of kinetic energy along linear collision sequences (LCSs). If thermal displacements are added, long LCSs do not occur, eliminating the tail. Moreover, energy dissipation from the cascade is increased at all times. Only the 1000 K results are shown in fig. 1a; those for other temperatures fall between this line and the static case.

Figs. 1b–1d show recoil energy spectra in static calculations without redisplacements at three times. The spectra for other cases are similar. It is convenient to plot $f(E, t) = E^2 p(E, t)$ against $1/E$, where $p(E, t) dE$ is the number of recoils at time t with kinetic energy between E and $E + dE$. In the hard-core approximation in structureless media, $f(E, t)$ is a constant [7]. Early on, most recoils have high kinetic energies and there is a peak in the spectrum which moves to lower energies and spreads out as time advances. At long times, the spectrum approaches hard-core behavior at low recoil energies. The spectrum in fig. 1d has two noteworthy features. One is a small peak at the high-energy end of the spectrum which persists for a rather long time even with thermal disorder: it represents channeled atoms which lag behind most other recoils in slowing down. The other is a small peak near 7 eV, marked by an arrow, which also persists for a
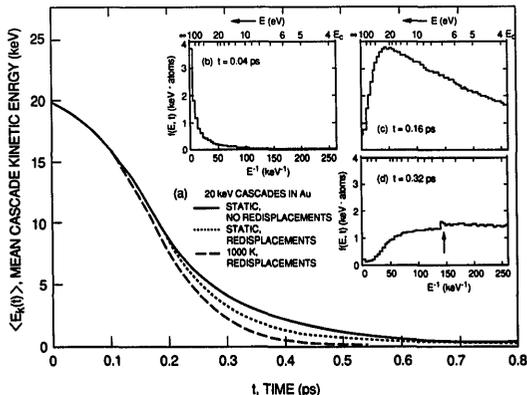
Fig. 1. (a) The mean cascade kinetic energy, $\langle E_k(t) \rangle$, as a function of time for 20 keV cascades in Au. (b)–(d) The kinetic energy spectra of recoils in static Au without redisplacements at 0.04, 0.16, and 0.32 ps.

long period: it eventually dissipates slowly, but is absent when thermal disorder is included. It represents long LCSs, along which energy dissipation is rather slow. At 7 eV, Au atoms require about 0.11 ps to move a nearest-neighbor distance, accounting for the slow dissipation.

Fig. 2a shows $\langle N(t) \rangle$, the mean number of atoms in motion in the same 20 keV cascades in Au. With redisplacements allowed, $\langle N(t) \rangle$ is reduced and falls more rapidly at longer times than otherwise. This behavior reflects the added loss of energy from the cascade because of the encounters of recoils with nonlattice target atoms. In the static calculations, a persistent tail due to LCSs is evident. If thermal disorder is added, $\langle N(t) \rangle$ drops further at long times, removing the tail almost entirely, but the number of moving atoms increases at short times. This is a result of the reduction of blocking effects by thermal disorder: many encounters in which moderate amounts of energy are transferred replace a smaller number of very hard encounters. Fig. 2b shows histograms of the times at which recoils stop which are later redisplaced and of the times at which they are redisplaced. The centroids of these distributions are about 70 fs apart, but distributions of the delay times show that half the atoms are redisplaced < 60 fs after they stop, many of them almost immediately, while a few are delayed for > 300 fs. Distributions of the delay times are independent of initial kinetic energy and are unaffected by thermal disorder. It should be noted that the bulk of the redisplacements occur while the largest numbers of recoils are in motion.

Cascades in Au show similar behavior at other energies, with the effects of redisplacements becoming greater at higher energies. In Cu, on the other hand, the effects are much smaller. Both results are consistent with theoretical expectations [8] that cascade nonlinearities are more important at high energies and in high-Z targets.

4. Overall cascade properties

At the end of a MARLOWE computation, the defects produced are arranged in Frenkel pairs and sorted into classes [1,2]. Only so-called distant pairs are regarded as permanent displacements: pairs of other classes are unstable and annihilate promptly. The number of pairs may be discussed in terms of the familiar "damage energy" model of radiation damage [9]: the damage energy $\hat{E}$ is E_0 less the energy lost in electron excitation. The number of Frenkel pairs produced in a cascade is $\nu = \hat{E}/Q$, $\hat{E} > Q$, a displacement energy. In the well-known model of Kinchin and Pease [10], for example, $Q = 2E_d$, with E_d the displacement threshold, about 29 and 43 eV for Cu and Au, respectively [11]. Fig. 3 shows the effective displacement energy $Q = \langle \hat{E} \rangle / \langle \nu \rangle$, as a function of E_0, derived from calculations in both Cu and Au. The MARLOWE results for both metals are roughly what is expected from the Kinchin–Pease model, namely ~ 58 eV for Cu and ~ 86 eV for Au. It must be emphasized that the only explicit displacement threshold MARLOWE is $E_c + E_b$, 7.02 eV for Cu and 7.62 eV for Au.
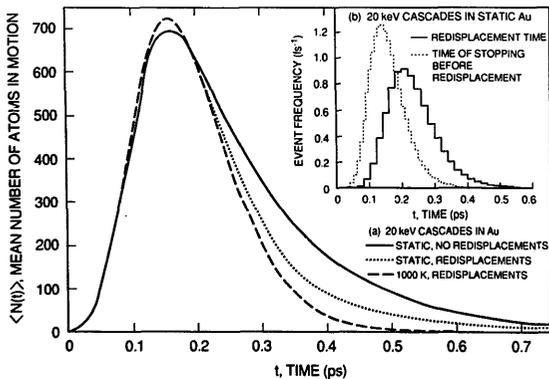
Fig. 2. (a) The mean number of atoms in motion as a function of time for 20 keV cascades in Au. (b) The times of occurrence of redisplacements in 20 keV cascades in static Au and the times at which recoils stop which are to be redisplaced later.
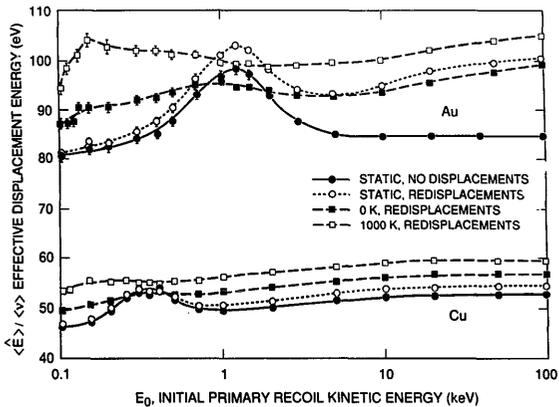
Fig. 3. The effective displacement energy, $\langle \hat{E} \rangle / \langle \nu \rangle$, as a function of the initial kinetic energy for cascades in Cu and Au.

In both metals, static calculations without redisplacements show rather constant values of Q except in regions near 0.35 keV in Cu and 1.25 keV in Au, where Q is significantly higher (that is, $\langle \nu \rangle$ is significantly lower) than at other energies. When redisplacements are added to the static model, Q increases slightly in Cu and substantially in Au, but the regions of maximum Q persist. When thermal disorder is introduced, however, the maxima are removed, showing them to be associated with lattice effects, mainly the formation of long LCSs. In fact, defining "long" LCSs to be those with three or more members, only above about 0.25 keV in Cu and about 0.7 keV in Au does essentially

every cascade have at least one such sequence. In the regions of the maxima, the LCSs are more efficient in carrying kinetic energy out of the cascade than they are at other energies. At lower energies, the number of LCSs is reduced; at higher energies, their lengths are limited by defocusing. This behavior is not affected by the redisplacement of cascade atoms.

The much greater importance of redisplacements in Au is evident in fig. 3: Q increases by $\sim 16\%$ at 10 keV and $\sim 19\%$ at 100 keV, while in Cu it increases by only $\sim 3\%$ at each energy. Thermal disorder further increases Q in both metals; again the effects are much smaller in Cu than in Au. The 1000 K data from Au
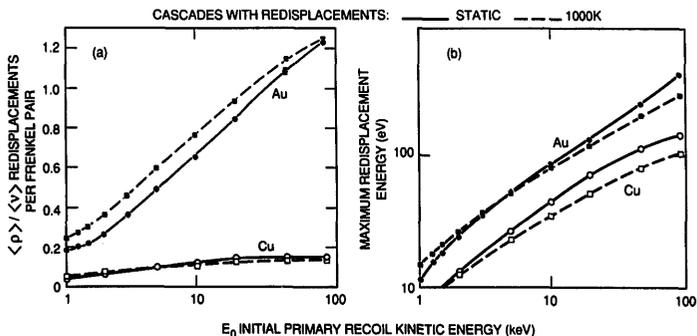
Fig. 4. (a) The number of redisplacements per Frenkel pair as a function of the initial kinetic energy for cascades in Cu and Au. (b) The maximum redisplacement energies in the same cascades.

VI. CASCADES, DAMAGE, RANGES

show some evidence of the proximity of the threshold near 0.1 keV; similar behavior may be developing in the 0 K data in the same region. No such behavior is seen in Cu, where Q is well below 100 eV. Another way of looking at the importance of redisplacements in cascade development is given in fig. 4a, which shows the ratio of $\langle \rho \rangle$, the mean number of redisplacements per cascade, to $\langle \nu \rangle$. In Cu, the ratio is small (~ 0.1) and essentially independent of E_0. In Au, on the other hand, it is larger and increases almost linearly with E_0. Above about 30 keV, there are more redisplacements than displacements in Au.

Fig. 4b shows the mean maximum kinetic energy at which redisplacements occur in cascades in both metals. In all cases, this quantity rises with energy, varying roughly as $E^{1/2}$. The spectrum of initial redisplacement energies is strongly peaked near E_c. Although there are occasional redisplaced atoms which carry 100 eV or more, they are quite rare, especially in Cu, and do not influence overall cascade properties significantly. Finally, there seem not to be any "local" structural differences in cascades caused by the redisplacement process. That is, the distribution of Frenkel-pair separation distances was not altered when redisplacements were included.

A few calculations were made to test the sensitivity of the results to the value of E_c. It is easily deduced in the hard-core approximation that the number of stopped cascade atoms and the number of low-energy recoils are both inversely proportional to E_c, so the frequency of encounters between them may be expected to vary as E_c^{-2} and this rule proves to describe high-energy cascades in Au fairly well. Both the number of redisplacements and the number of encounters with nonlattice targets show this behavior.

5. Discussion

Encounters between recoil atoms and nonlattice target atoms influence cascade development partly through their role in energy dissipation. The differences between the static calculations with and without redisplacements can be analyzed in this manner: the damage energy is modified by excluding energy transferred to nonlattice targets and a modified effective threshold is obtained. This agrees well with the value obtained without the nonlinearity in Cu. The agreement for Au is less good, suggesting that other factors are also significant. One of these factors is the cascade volume, of which one measure is the total number of lattice sites visited by the recoils. The cascade volume decreases when redisplacements are allowed, but in Cu by only about 5% at 100 keV. In Au, on the other

hand, the volume decreases by about 13% at 100 keV. The smaller cascade volume increases the concentration of nonlattice targets and, therefore, the number of redisplacements

The picture which emerges from these calculations is the following. As a collision cascade develops, stopped atoms accumulate until there are enough of them to supply targets for still-moving recoils. During the time when the number of recoils is the largest, collisions with nonlattice targets lead to redisplacements. The smallness of the delay between the stopping of a recoil and its redisplacement indicates that there is a very high density of recoils during this phase, but further study is required to analyze this situation in more detail. As E_0 rises, the number of potential nonlattice targets rises also, as does the number of moving recoils in each energy interval. The rate of such encounters depends on the product of these densities. Most encounters with nonlattice targets involve only low-energy recoils, but the highest energy at which such events occur rises slowly with E_0.

Finally, it must be noted that at the highest energies in Au, the number of encounters between recoils and nonlattice targets is great enough to suggest that further nonlinearities, involving collisions between low-energy moving recoils really need to be included in the model to obtain a more complete picture.

References

[1] M.T. Robinson, Nucl. Instr. and Meth. B48 (1990) 408.
[2] M.T. Robinson, Phys. Rev. B40 (1989) 10717.
[3] O.S. Oen and M.T. Robinson, Nucl. Instr. and Meth. 132 (1976) 647.
[4] M.T. Robinson, User's Guide to MARLOWE (Version 13), unpublished; this version of the program will be available from the Radiation Shielding Information Center, Oak Ridge National Laboratory, P.O. Box 2008, Oak Ridge, TN 37831-6362, USA, in early 1992.
[5] J. Lindhard, M. Scharff and H.E. Schiøtt, K. Dan. Vidensk. Selsk. Mat. Fys. Medd. 33 (14) (1963).
[6] M. Blackman, in: Handbuch der Physik, vol. 7, part 1 (Springer, Berlin, 1955) p. 192.
[7] M.T. Robinson, Philos. Mag. 12 (1965) 145.
[8] See, e.g.: H.H. Andersen, Nucl. Instr. and Meth. B18 (1987) 321.
[9] See, e.g.: M.J. Norgett, M.T. Robinson and I.M. Torrens, Nucl. Eng. Des. 33 (1975) 50.
[10] G.H. Kinchin and R.S. Pease, Rep. Prog. Phys. 18 (1955) 1.
[11] P. Lucasson, in: Fundamental Aspects of Radiation Damage in Metals, vol. 1, eds. M.T. Robinson and F.W. Young Jr. (U.S. Energy Research and Development Administration Report CONF-751006, 1975) p. 42.