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Correlating TEM images of damage in irradiated materials to molecular dynamics simulations

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Abstract

TEM image simulations are used to couple the results from molecular dynamics (MD) simulations to experimental TEM images. In particular we apply this methodology to the study of defects produced during irradiation. MD simulations have shown that irradiation of FCC metals results in a population of vacancies and interstitials forming clusters. The limitation of these simulations is the short time scales available, on the order of 100 s of picoseconds. Extrapolation of the results from these short times to the time scales of the laboratory has been difficult. We address this problem by two methods: we perform TEM image simulations of MD simulations of cascades with an improved technique, to relate defects produced at short time scales with those observed experimentally at much longer time scales. On the other hand we perform in situ TEM experiments of Au irradiated at liquid-nitrogen temperatures, and study the evolution of the produced damage as the temperature is increased to room temperature. We find that some of the defects observed in the MD simulations at short time scales using the TEM image simulation technique have features that resemble those observed in laboratory TEM images of irradiated samples. In situ TEM shows that stacking fault tetrahedra are present at the lowest temperatures and are stable during annealing up to room temperature, while other defect clusters migrate one dimensionally above −100 °C. Results are presented here.
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1. Introduction

Molecular dynamics (MD) simulations have proven to be very useful in understanding the production and evolution of defects during irradiation [1]. However, connection to experimental observations has been difficult, mostly due to differences in time scales. While MD simulations can only achieve time scales on the order of 100 s of picoseconds to nanoseconds, laboratory time scales can be seconds to hours or years. On the other hand the images taken in the laboratory with transmission electron microscopy (TEM) and atomic positions calculated by MD need to be correlated, to ensure that those defects observed experimentally and those simulated correspond to the same object. TEM image simulations of atomic positions obtained by MD simulations provide a direct comparison to TEM experimental images [2,3].

Kiritani and co-workers have extensively studied the damage produced in Cu, Au and other metals during irradiation with high-energy neutrons [4]. They have demonstrated with these experiments that most of the defects produced are stacking fault tetrahedra (SFT) type and that some of the vacancy clusters appear forming groups, indicating that they were formed from a single collision cascade. However, it cannot be deduced experimentally whether SFTs are formed directly during the cooling of the cascade or formed by defect migration after the cool down. Concurrently, MD simulations of

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collision cascades in Cu have also been extensively studied during the last decade providing insight to the process of defect production during irradiation [1].

It should be noted that discrepancies still exist between TEM images, taken at laboratory time scales, and MD simulations. As an example, in the case of copper, experiments show that 90% of all defects produced during irradiation are SFT type [4,5]. Picosecond-scale MD simulations show the formation of both vacancy and interstitial type of defects during the cascade [1]. SFTs have been observed to form in collisional cascades [6,7], and the formation of SFTs from triangular platelets and vacancy clusters formed during collision cascades has been explained previously as due to a Silcox-Hirsch mechanism [8]. However, MD shows an efficiency of only 20% SFT formation versus 90% observed experimentally [6]. This discrepancy could be due, again, to the short times affordable in MD simulations or not enough statistics. The transport of the interstitial clusters to sinks is also still a matter of debate. MD simulations have shown the high mobility of the interstitial clusters [9,10], with migration energies on the order of 0.1 eV. The transport of these clusters to sinks will reduce the population of interstitial loops, resulting in a high concentration of SFTs at the experimental time scales. In order to understand the relationship between experiments and simulations it is necessary to identify those defects produced in the experiment and correlate them with the defects obtained in the MD simulations.

One focus of this work is, to address the correlation between TEM experimental images and MD simulations through TEM image simulations of MD simulations of collision cascades in Cu. The goal of these simulations is to identify defects with features similar to those observed experimentally at much longer time scales. If found we could then conclude that those defects have been formed during the picosecond time scale of the cascade. On the other hand, in order to understand defect evolution in time, we present an example of an experiment of irradiation of Au at liquid-nitrogen temperature. In situ TEM images are obtained as the temperature is increased in order to study the changes in defect population and character during annealing in an effort to understand the differences in time scales between experiments and simulations.

2. Relating TEM images and MD simulations through TEM image simulations

Fig. 1 shows in the top row the evolution of a collision cascade in copper generated by a 20 keV recoil. Kiritani's analysis of their experimental irradiation showed that the threshold energy for sub-cascade formation in Cu is around 10 keV [4], while our simulations seem to point towards a value between 25 and 30 keV. We therefore consider that 20 keV is a representative energy of the recoils produced during high-energy neutron irradiation. Several snapshots in time are shown. As explained by Diaz de la Rubia et al. in the late 1980s [11], there is a high temperature gradient during the collision cascade, with temperatures raising above the melting point during a few picoseconds that generates a large disordered region in the lattice, as shown in Fig. 1(b). Replacement collision sequences separate vacant sites from interstitial atoms. The cooling down of the cascade results in vacancies at the core of the collision cascade and interstitials in the surroundings. The number of defects produced with respect to the number predicted by the Norgett, Robinson and Torrens model (NRT) [12] has been studied previously by several authors [1,13,14], as well as the amount of clustering of vacancies and interstitials for many different metals.

In the particular case presented in Fig. 1, the total number of defects produced by this 20 keV recoil atom in Cu is 65 (32% efficiency with respect to the NRT modeling, considering a threshold displacement energy for Cu of 40 eV). 12% of the vacancies and 10% of interstitials are isolated. The largest vacancy cluster size is 50 vacancies while the largest interstitial size is 21 interstitials. For this calculation we have considered that two defects belong to the same cluster when the distance between them is smaller than the first nearest neighbors distance in Cu. The simulation was performed at 300 K using an EAM potential [15]. All interstitial clusters are glissile except one. This cluster, however, shows a rearrangement during the simulation. It rotates to align along a $\langle 1\,1\,0\rangle$ direction as can be observed it the last two frames of Fig. 1. All glissile clusters move along $\langle 1\,1\,0\rangle$ directions. Although the results presented here are of one single simulation, we have performed 12 independent 20 keV collision cascades in copper. Details of these simulations have been published elsewhere [16]. In general, all cascades present clustering of vacancies and interstitials with similar features as those shown here.

Identification of vacancies and interstitials shown in Fig. 1 was obtained by computing the number of atoms on each representative Wigner–Seitz cell. When there are no atoms within the cell a vacancy is identified while an interstitial corresponds to two atoms in a single cell. Using this method the distinction between vacancies and interstitials and their quantification is clear. However, it is not easy to identify the formation of 3D structures such as SFTs [7], since the result is a 3D cluster of vacancies, as also shown in Fig. 1. It is also important to point out that this analysis is done in a single configuration in time and at room temperature. Temperature fluctuation will also make the identification of 3D structures difficult, as explained below.

Fig. 1 shows at the bottom the HR-TEM image simulations along [0 0 1] of the collision cascade at

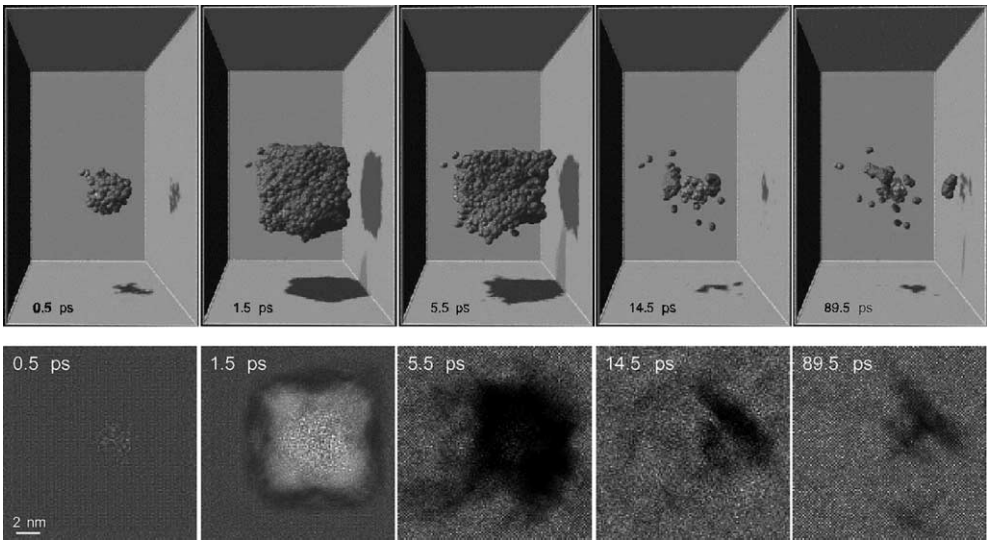

Fig. 1. Top row: Cu 20 keV cascade time evolution (MD). Dark symbols: interstitials, light symbols: vacancies. Bottom row: corresponding HR-TEM images.

different times. The acceleration voltage is 200 kV with Scherzer defocus and exposure time of 1 fs (MD time step). Noise in the images is due to temperature fluctuations and short exposure time. Nevertheless, these images show the production of a shock wave, due to the energetic recoil, at the peak of the thermal spike. The compressive wave presents symmetry related to the $\langle 1\,1\,0\rangle$ directions. The migration of a cluster is also observed between bottom Fig. 1(d) and (e). At the end of the simulation, 98 ps, two distinct clusters of defects are observed in the TEM image simulation. However, due to the low contrast and the graininess of the image, which basically results from thermal noise, it is difficult to determine the features of these defect clusters.

In order to reduce the noise in the image simulations we perform an average of the positions of all the atoms in the simulation during 1 ps and compute the TEM image of the averaged defect locations. It corresponds to an average of 1000 consecutive configurations. This mimics the exposure time of the experimental image acquisition. After performing this operation the contrast of the SFT is clearly seen, as shown in Fig. 2(b). As a comparison Fig. 2(c) shows an experimental TEM image of a number of SFTs produced in copper by irradiation with 590 MeV protons at room temperature and 0.01 dpa [5]. These experiments were compared to fission and fusion neutron irradiation, showing that at room temperature there are no significant differences in terms of defect concentration and type. For more information on experimental set-up see Ref. [5]. Fig. 2(a) shows the projection of the atom positions after doing the average in time and with the same tilt angle as in Fig. 2(b).

3. Evolution of defect population through irradiation and in situ annealing experiments

As explained in the example above, TEM image simulations can be used to compare more directly those atomic configurations obtained in MD simulations of collision cascades with those defects observed in TEM measurements. However, this comparison can only be done if we understand the time evolution of defects, since simulations and experiments are orders of magnitude different in time. A way of addressing this problem is through irradiation at low temperature, suppressing defect migration and identification of defect structures at these temperatures through TEM. Moreover, by in situ annealing of the irradiated material we can analyze defect evolution and compare the changes in defect type and population with time. We present here an example of such an experiment and the results obtained from this observation.

An in situ experiment was performed in pure polycrystalline Au by implanting W^+ ions at 164 keV to a dose of about 2×10^9 ions/cm^2 at liquid-nitrogen temperature. The implanted TEM sample was then cryo-transferred and observed in the transmission electron microscope. The microstructural evolution with temperature was recorded between liquid-nitrogen temperature and room temperature. It appears that SFTs are already present at the lowest temperatures. Fig. 3 shows clearly the contrast of an SFT observed at −140 °C in the TEM.

The temperature of the sample was increased by in situ heating, with increments of about 10 K every 15

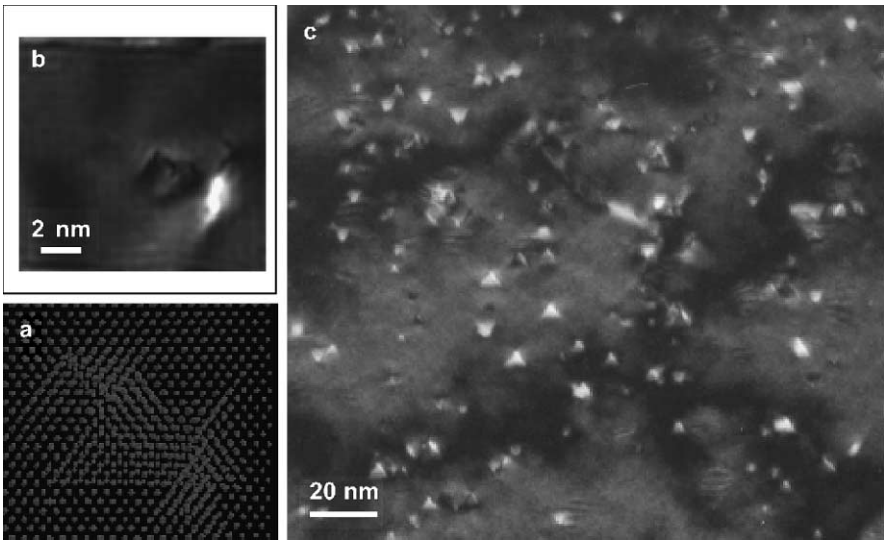

Fig. 2. (a) Atomic positions averaged over 1 ps of the defect configuration produced by a 20 keV cascade in Cu. (b) Corresponding weak beam TEM image g(6.1g) and (c) experimental weak beam TEM image g(6g) of Cu irradiated with 590 MeV protons at 0.01 dpa at room temperature.

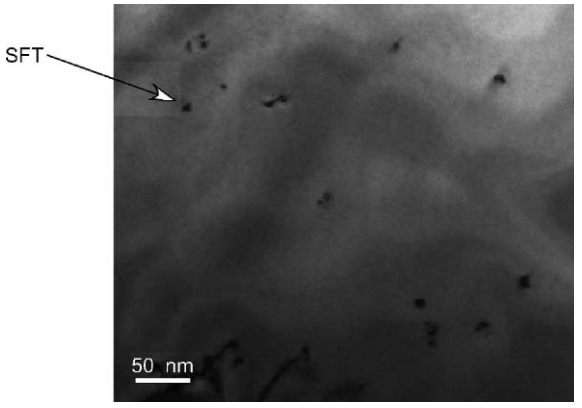

Fig. 3. Experimental TEM image of an SFT in Au at −140 °C produced under irradiation with 164 keV W.

min. We observe that the microstructure is stable up to ~−100 °C. Above this temperature, the peculiar 1D glide of dislocation loops emerging from displacement cascades is observed. Note that SFTs remain stable up to room temperature. Fig. 4 shows the presence of a cluster group, formed probably by a collision cascade, and similar to those shown previously by Kiritani [4] in neutron irradiated samples, and a loop in its proximity, marked in Fig. 4(a). After a few seconds at −100 °C we can observe how the loop migrates in one dimension away from the cluster group. No significant changes are observed at this temperature during the exposure time. This observation is in agreement with the existence of glissile loops formed during the collision cascades obtained from MD simulations in FCC materials such as Au [17] and the simulations in Cu shown above.

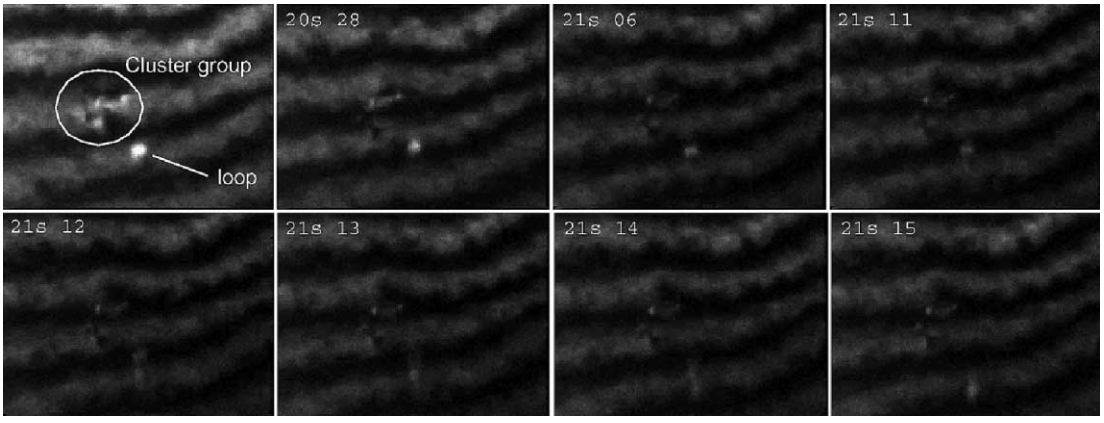

Fig. 4. Loop migration in 1-D. Loop produced during irradiation of Au with 164 keV W.

4. Conclusions

The TEM image simulation technique allows us to bridge the gap between the MD simulations and the experimental observations in the TEM. A new technique was developed in order to simulate the exposure time encountered in the photographic acquisition of the image in the TEM: atomic positions are averaged over 1 ps. By using this averaging technique the TEM image simulation of a Cu sample exposed to a displacement cascade clearly showed the features of an SFT as observed experimentally. This also implies that the microstructure derived from MD simulations, even at short times after the cooling of the collision cascade, is indeed matching the one observed experimentally at much longer times.

TEM observation of Au irradiated by $W^{=}$ at 164 keV and at liquid-nitrogen temperature show the presence of SFTs that do not evolve during in situ heating. This observation is again in agreement with the formation of these defects in the first few picoseconds of the collision cascade. In situ heating in the TEM of the implanted Au shows a stable microstructure up to −100 °C. Above this temperature, loops located close to cluster groups originating from collision cascades are observed to migrate in a 1D glide, in agreement with MD simulations.

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