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The effects of interfaces on radiation damage production in layered metal composites

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Abstract

Molecular dynamics computer simulations of 5 keV displacement cascades near cube-on-cube Cu–Ni interfaces have been performed. Both coherent interfaces (with large coherency stresses) and semi-coherent interfaces (with misfit dislocations in the interface) typical of nanolayer metal composites are considered. The primary damage state of cascades from 5 keV recoil atoms in the composite differs from that in cascades generated in pure Cu and Ni. In the semi-coherent case some defect clusters form directly on the misfit dislocations. Excluding these, the defect yields for both interface types are about two-thirds of the average yield of defects for cascades in the pure metals. In single cascades, as well as for overlapping multiple cascades, the numbers of surviving defects in the semi-coherent interface are significantly different in the Cu and Ni layers, but they are the same in Cu and Ni for the coherent case. Anti-site atomic mixing occurs near both types of interfaces, more so in the coherent case.

1. Introduction

Multilayered composites consisting of many alternating metal layers, each only nanometers thick, possess enormous strength, approaching theoretical limits. These materials also display unexpectedly high thermal and mechanical stability [1]. Their unique properties derive from the operation of deformation mechanisms that do not occur in conventional metallic materials and are a result of the large internal interfacial areas and high coherency strains of the nanolayered metals. Property tailoring can be achieved through control of the layer thickness and the lattice mismatch by careful selection of composite constituents. The enormous interface area to volume ratio of these materials may also positively affect their resistance to radiation damage, making them potentially useful materials for applications in fusion reactors.

As a first step in assessing the response of such materials to irradiation, we have begun studying the effects of typical interfaces on primary damage production. Atomic-scale molecular dynamics (MD) simulations of displacement cascades have been performed near bilayer interfaces of layered Cu and Ni, one of the systems for which nanolayer composites have been produced in the laboratory by physical vapor deposition techniques. Companion irradiation experiments on these samples are in progress.

These multilayer composites are typically made of alternating layers of two metals, each layer having the same uniform thickness. Samples have been made with layer thicknesses varying from 1 nm to 4 mm [1]. For very thin layers the interfaces are coherent, and each layer is in compression or tension as necessary to maintain coherency of the lattice parameter across the interface. At larger layer thicknesses, it is energetically favorable for misfit dislocations to form in the interface to reduce or eliminate the coherency stresses at some distance from the interface (on the order of half the misfit spacing), and the interfaces are referred to as 'semi-coherent'. When the two metals have the same crystal structure, e.g. face centered cubic for Cu and Ni,

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a cube-on-cube layering scheme results in the formation of misfit dislocations (Lomer–Cottrell $\langle 1\,1\,0\rangle\{1\,0\,0\}$ dislocations lying on the (1 0 0) Cu/Ni interface plane) in a regularly spaced network, the spacing of which depends on the mismatch of their lattice constants.

2. The model

We have studied the nature of cascade damage in both coherent and semi-coherent Cu–Ni bilayer models by simulating cascades from 5 keV recoil atoms originating at varying distances from the interface in both the Cu and Ni layers. Although 5 keV recoils are quite small compared to those from fusion neutrons, they are of sufficient energy to display the essential elements of cascade damage for this initial study, and they fit within a model of convenient size, about 110 000 atoms total. The model consists of a rectangular volume containing two rectangular sub-volumes of approximately equal numbers of Cu and Ni atoms, respectively, oriented with their respective (0 0 1) faces parallel and their crystallographic axes aligned. See Fig. 1. Periodic boundaries were applied in all three dimensions.

The issues and procedures involved with creating coherent and semi-coherent interfaces are discussed in earlier work by Hoagland et al. [2]. A brief description is given here. The coherent interface was formed by straining each of the two cube-on-cube layers such that the lattice spacing in the x–z plane throughout both layers was the same. This requires compressing the Cu layer (perfect lattice parameter 0.3615 nm) and expanding the Ni layer (perfect lattice parameter 0.352 nm) in directions parallel to the interface. To achieve the final configuration, the elastic properties of the two materials must be taken into account, as well as lattice contraction or expansion in the transverse (y) direction due to the Poisson effect. The composite lattice parameter is 0.3563 nm.

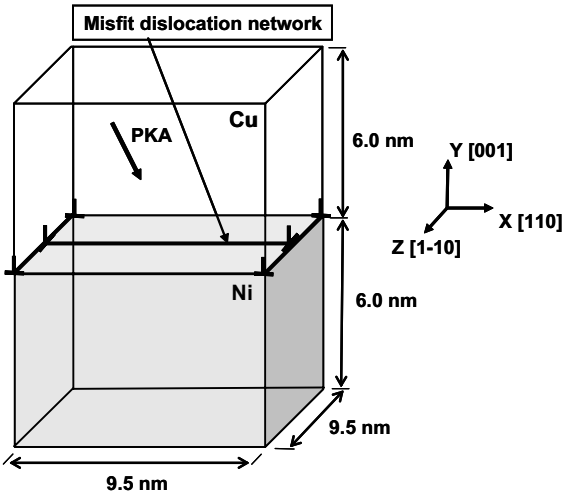

Fig. 1. The orientation and approximate dimensions of the computational model for the semi-coherent Cu/Ni model, which contains a network of misfit dislocations along $\langle 1\,1\,0\rangle$ directions in the [0 1 0] interface The coherent Cu/Ni model has the same orientation and dimensions as the semi-coherent model, but with no misfit dislocations. Coherency is maintained by applied stresses along the x and z directions.

The semi-coherent interface was formed by starting with perfect Cu and Ni layers and displacing all the atoms in the model according to the elastic field of an infinite network of perpendicular misfit dislocations lying in the interface. The model is then relaxed. The present semi-coherent model with periodic boundaries is oriented such that an $a/2\langle 1\,1\,0\rangle$ misfit dislocation along the x direction is centered in the (0 1 0) Cu/Ni interface, and two transverse $a/2\langle 1\,1\,0\rangle$ misfit dislocations along the z direction intersect it at the edges of the model cell. See Fig. 1. For Cu and Ni the calculated misfit dislocation spacing determined by the mismatch in the lattice parameters is 9.46 nm. The size of the model is approximately the same for the coherent and semi-coherent cases.

Interatomic potentials for Cu–Cu, Ni–Ni and Cu–Ni interactions were based on the embedded atom potentials of Voter and Chen [3], with the universal potential of Ziegler et al. [4] spline-fitted to the pair potential at small interatomic separations. In all cases the model was equilibrated for a simulated time of 10 ps at a temperature of 100 K prior to starting the cascade simulations.

In this initial study of cascade-induced displacement damage, a total of 12 cascades were simulated one at a time in each of the coherent and semi-coherent models. In each model six cascades were generated on each of the Ni and the Cu sides of the interface, with the 5 keV primary knock-on atoms (PKA) being initially aimed in various directions toward the interface. The distance of the PKA from the interface varied from about 0.2 to 4.4 nm, and in all cases at least some part of the initial collisional phase of the cascade (i.e., the thermal spike) occurred in both materials. In a separate study, the effects of multiple cascades in the same sample were also investigated. Up to four cascades were produced sequentially in the same model, and the damage was recorded as a function of 'dose.'

In the post-cascade analysis, vacancies and interstitials were identified in terms of the occupation of initial lattice sites, and atomic mixing due to cascades was determined by counting the number of atoms of type A in the material of type B. In this paper we refer to atoms of type A occupying a lattice site of type B, and vice versa, as 'anti-site' defects. In all cases the numbers of atoms and lattice sites of each type were conserved in the model. The defect configurations were analyzed in each case after the cascade had cooled sufficiently near

to 100 K, and such that the numbers of defects did not change, a total simulated time of about 15–20 ps.

3. Results

Considering all twelve cascades in the coherent interface model, the average number of defects produced per cascade was about the same in the Ni and Cu layers. See Table 1. The average numbers of residual vacancies, self interstitial atoms (SIA) and anti-site defects were about the same in the Cu layer as in the Ni layer. Considering the cascades individually, the PKAs furthest from the interface produced the greatest numbers of defects on their side of the interface, as expected. The numbers of anti-site defects in both layers were the same and about equal to the sum of the number of residual vacancies and SIA produced. The PKAs nearest the interface produced the greatest number of anti-site defects. Since the simulated times are too short for significant diffusional processes to be observed (the exception is the migration of crowdion clusters), the atomic mixing observed in the simulations is primarily due to ballistic effects. In this case we expect that the amount of mixing is not affected much by the solubility of one element in the other, but rather is determined to first-order by the PKA energy and location as well as the relative masses of the Cu and Ni atoms, which differ by only 8%.

In the semi-coherent model the defect production is somewhat different from the coherent model (Table 1). About twice as many total point defects are produced in the semi-coherent case as compared to the coherent case. Also, the relative numbers of vacancies and SIA in the Cu and Ni layers are different. There are about twice as many SIA as vacancies in Cu and about twice as many vacancies as SIA in the Ni. Also, there are about twice as many anti-site defects in Cu as in Ni. In all cases both the numbers of Cu and Ni atoms and the balance between vacant sites and interstitial atoms are conserved.

A striking feature of the cascade damage near the semi-coherent interface is the formation of stacking fault-like defects associated with the misfit dislocations.

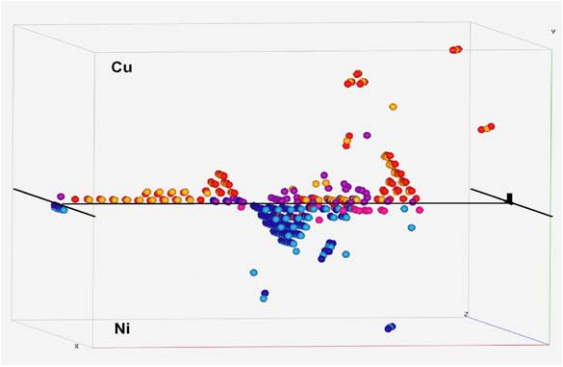

Fig. 2. Defects resulting from a 5 keV cascade in the semi-coherent Cu/Ni model. In the Cu layer vacant sites are yellow, displaced atoms are red, and a Ni atom in a Cu lattice site is purple. In the Ni layer vacant sites are light blue, displaced atoms are dark blue, and a Cu atom in a Ni lattice site is dark pink. Interstitials are depicted as two displaced atoms on either side of the vacant site they share. Longer strings of alternating vacant sites and displaced atoms along the z direction [1–7] in Cu are more highly strained single interstitials forming in tetrahedral shaped clusters along the misfit dislocation. In the Ni layer the triangular-shaped defect cluster attached to the misfit dislocation is a stacking fault 'pyramid' of vacancies.

See Fig. 2. These are most likely related to a property of the misfit dislocations in the Cu–Ni interface: the misfits are Lomer–Cottrell dislocations that dissociate along close packed {1 1 1} planes projecting into the Cu layer, and the extent of the dissociation increases under applied tensile stresses parallel to the interface. Moreover, given sufficient layer thickness and the appropriate applied stresses, the array of misfit dislocations dissociates into a three-dimensional array of stacking fault pyramids [2]. In our simulations, during the cascade cooling process, after most of the in-cascade recombination has occurred, isolated areas of stacking fault can be observed emanating into the Cu layer from various places along the misfit dislocation. They disappear and reappear with time as thermal equilibrium is approached. After complete cooling, some vacancy clusters reside at the misfit dislocation in stacking fault tetrahedra (SFT)-

Table 1
Defect production in Ni and Cu layers by 5 keV cascades near coherent and semi-coherent Cu/Ni interfaces

Model	Ni			Cu		
	V	SIA	Cu in Ni	V	SIA	Ni in Cu
Coherent	6	5	9	4	5	10
Semi-coherent	18	8	11	6	15	21
Semi-coherent (excluding defects on MFD)	9	4	–	5	2	–

The average numbers of vacancies (V), self-interstitial atoms (SIA) and anti-site defects for 12 cascades in each interfacial model are shown. Also shown are the numbers of defects not associated with the misfit dislocations (MFD).

like configurations. Clusters of SIA are also observed to form in pyramidal structures at the misfits, as depicted in Fig. 2. SFT-like defects associated with the misfit dislocations appear as part of the damage in nearly all the cascades in the semi-coherent case. It is interesting to note in Table 1 that the numbers of vacancies and SIA in the semi-coherent interface that are clearly not associated with the misfit dislocations as described above are about the same as the total numbers of defects produced by cascades in the coherent case.

In Table 2 we compare the defect production in the coherent and semi-coherent Cu–Ni bilayers with the numbers of defects produced by 5 keV cascades in perfect bulk Ni and Cu, respectively, as reported by Bacon et al. [5]. On average, about 35% fewer defects are produced in the coherent case than in the pure metals. On the other hand, in the semi-coherent case about 50% more defects are produced than in the pure metals, of which about half are those associated with the misfit dislocations. This comparison with cascades in pure metals should be considered with some caution because it results from a small number of highly variable cascades that were initiated in a variety of locations in the highly stressed, inhomogeneous Cu/Ni bilayer models.

The effects of multiple 5 keV cascades on defect production were also studied in both the coherent and semi-coherent models. Four cascades, starting from different locations, were introduced sequentially into the same model at intervals of 10–20 ps. This was repeated with different cascades four times, and the results were averaged for each type of interface. The results are displayed in Fig. 3. For the coherent interface (Fig. 3(a)) the numbers of vacancies and SIA increase fairly uniformly with dose in both the Cu and Ni. The numbers of anti-site defects increase with dose at a much higher rate than the numbers of vacancies and SIA in both the Cu and Ni, tending toward saturation at the higher doses. In the semi-coherent case (Fig. 3(b)) the numbers of anti-site defects increase faster with dose than the

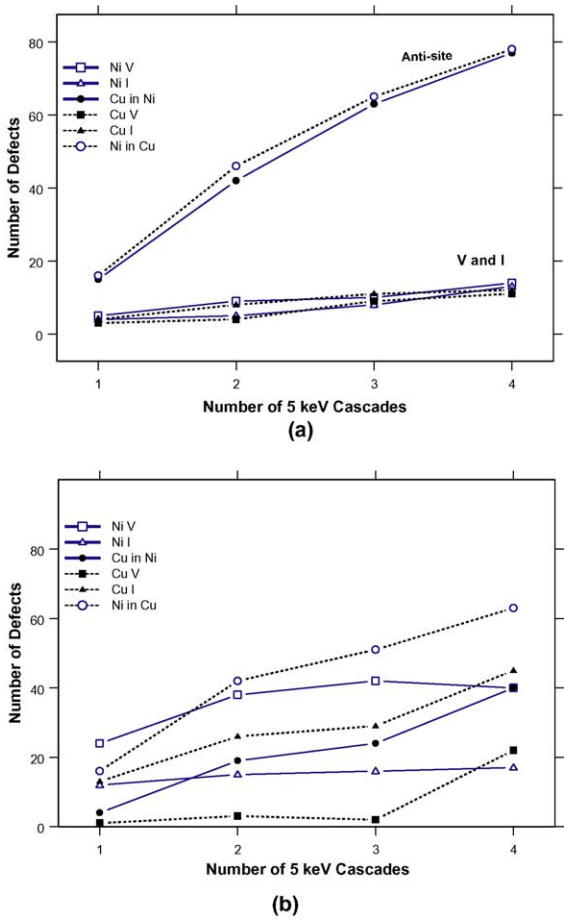

Fig. 3. The numbers of residual defects as a function of dose (the number of successive 5 keV cascades in the same sample) for: (a) the coherent model and (b) the semi-coherent model. The separate curves are for NiV, the numbers of vacancies in the Ni layer, NiI, the numbers of interstitials in Ni, Cu in Ni, the numbers of Cu atoms in Ni sites, and likewise for the defects in the Cu layer.

Table 2
Comparison of total defects from 5 keV cascades in layered Cu/Ni models with the average defect yields from 5 keV cascades in pure Cu and pure Ni [5]

Model	V	SIA	Total (V + SIA)	Total anti-site defects
Coherent	10	10	**20**	19
Semi-coherent	23	23	**46**	32
Semi-coherent (excluding defects on MFD)	14	6	20	
Pure	Cu	17	17	34
Pure	Ni	14	14	28
Average of pure Cu and Ni				**31**

The average numbers of vacancies (V), self-interstitial atoms (SIA) and total defects for 12 cascades in each interfacial model are shown.

vacancies and SIA, but at different rates in the two materials. As in the single cascades, the SIA in Cu and vacancies in Ni are present in greater concentration than the SIA in Ni and vacancies in Cu, and their numbers continue to increase at a somewhat higher rate with dose. There is some indication of saturation, or at least very slow growth, in the numbers of vacancies and SIA with dose, and overall the atomic mixing is less in the semi-coherent case.

4. Discussion and conclusions

There are clear differences in defect production for the coherent and semi-coherent interfaces, and in both cases the defect production differs from that in the respective perfect pure metals. In the coherent composite both sides accumulate damage of the same type at the same rate, but fewer vacancies and SIA are produced than in the pure metals. Anti-site defects are also produced in about equal numbers on both sides. Thus, in the coherent case both sides accumulate damage of the same type at the same rate, but fewer vacancies and SIA are produced than in the pure metals. (In regard to the comparison with pure metals, it should be noted that defect yields in MD simulations of pure metals have a standard deviation of about 25% of the mean value, while for our simulations in Cu/Ni bilayers the standard deviations are 50–100%.) In addition, atomic mixing is substantial, increasing with dose much faster than the numbers of vacancies and SIA. However, the mixing appears to saturate with dose. Since coherent interfaces are stable only for composites with very thin layers (less than about 5 nm in the Cu–Ni system), significant mixing of the Cu and Ni layers by prolonged irradiation could change the material properties significantly.

In the semi-coherent composite more defects are produced than in the coherent case, although about half of the defects in the semi-coherent case are directly formed at the interface and are associated with the misfit dislocation. It is not yet clear how the defects associated with the misfit dislocations will affect the subsequent behavior of the material. In the semi-coherent case the numbers and types of radiation-induced defects are different in Cu and Ni layers. It is premature to try to predict the overall effects of the asymmetry in defect production between the Cu and Ni layers on the properties of irradiated nanolayer Cu–Ni composites, especially when based solely on the properties of the primary damage state after 20 ps, since the migration energies and transport properties of defects in these systems is unknown.

The Cu–Ni composite system was chosen for these initial cascade studies because it is easily characterized and modeled, and the radiation damage phenomena can be compared with similar studies in the pure metals. Other nanolayered metal composites for which we have samples have interfaces with quite different characteristics. For example, Cu–Ag interfaces have a bigger difference in lattice mismatch than Cu–Ni, which creates more closely-spaced misfit dislocations [6], while layered fcc Cu and bcc Nb have entirely different types of interfaces [7]. Modeling radiation damage in these more complicated systems will undoubtedly reveal quite interesting effects.

Acknowledgements

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