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CALCULATION OF DIFFRACTION PATTERNS ASSOCIATED WITH ELECTRON IRRADIATION INDUCED AMORPHIZATION OF CuTi.

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ABSTRACT

A new approach that uses the multislice method in conjunction with molecular dynamics simulations to study electron irradiation induced amorphisation is presented. Diffraction patterns were calculated for CuTi and found to be more sensitive than the pair correlation function to the structural changes preceding amorphisation. The results from this approach and from a study of long range order are presented.

INTRODUCTION

Crystalline materials have been rendered amorphous by a variety of solid-state techniques such as irradiation by energetic particles, hydrogenation, mechanical alloying and annealing of multilayer films [1]. Because of its simple polymorphous nature, the amorphization of intermetallic compounds by high energy electron beams has been extensively studied [2-8]. The damage produced by electrons is homogeneous and consists of two defect components; namely, isolated interstitials and vacancies (i.e. Frenkel pairs) and antisite defects (i.e. chemical disorder).

One highly controversial aspect of the transformation concerns the relative importance of Frenkel defects versus chemical disorder in inducing amorphization. This question was recently addressed by Sabochick and Lam [9], using the molecular dynamics (MD) simulation technique. The crystal-to-glass transformation of a model intermetallic compound was monitored via the changes in the calculated pair correlation function caused by the introduction of Frenkel pairs or by chemical disorder introduced by random atom pair interchanges. Although very informative, the calculated pair correlation functions are difficult to compare directly with experiments, in particular with the commonly observed, but poorly understood changes in single crystal diffraction patterns that often precede the onset of amorphization [10].

In order to bring the MD results in closer contact with experiments, the multislice method was used to calculate the single crystal diffraction patterns and high resolution lattice images of the atomic defect structures obtained from MD simulations.

THE MOLECULAR DYNAMICS SIMULATION

The simulation technique used by Sabochick and Lam [9] was an isothermal isobaric molecular dynamics scheme using a modified version of the computer code DYNAMO [11]. The simulation cell consisted of 288 atoms each of copper and titanium arranged in a B11 lattice as shown in Figure 1. The system was maintained at 160K and zero pressure. The interatomic potentials for Cu, Ti and CuTi were developed using the approach of Oh and Johnson [12] based on the embedded-atom method [13].

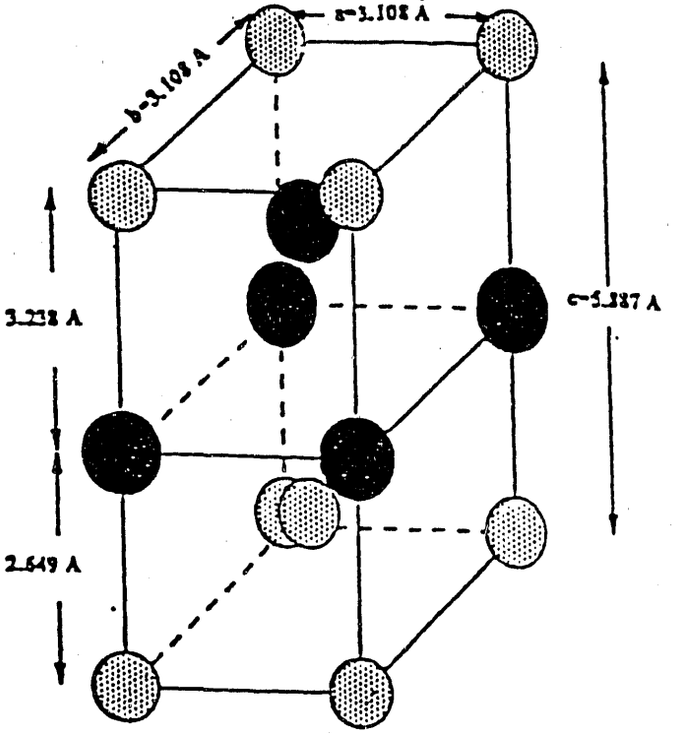
A perfect lattice was initially equilibrated for 500 time steps ($dt = 2 \times 10^{-15}$ s). Every 20 time

steps a randomly chosen pair of copper and titanium atoms were interchanged to investigate the effect of disordering on the structure of the system. The system configuration was periodically saved during the disordering process. The configurations were subsequently allowed to evolve sufficiently long for the system volume and energy to attain equilibrium.

The effect of Frenkel pair generation was simulated by removing an atom at random and inserting it into a random interstitial site. The interstitial atom was inserted at least 1.5 Å from any other atom and the system was allowed to relax. As before the system configurations were periodically saved and equilibriated.

Figure 2 shows the dependence of the system volume and energy on the number of atom interchanges and the number of Frenkel pairs introduced. Sabochick and Lam [9] found that the maximum volume expansion was 2% for the case of Frenkel pairs and 1.7% for atom interchanges. The increase in energy of the system was 0.06 eV/atom for Frenkel pairs as opposed to 0.04 eV/atom for the atom switches.

Figure 3 shows the pair correlation functions (PCF) of the system obtained by Sabochick and



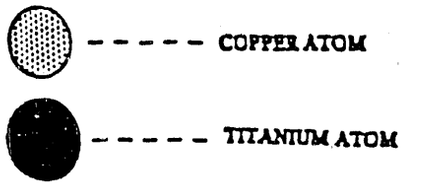


Fig. 1. Crystal structure of CuTi.

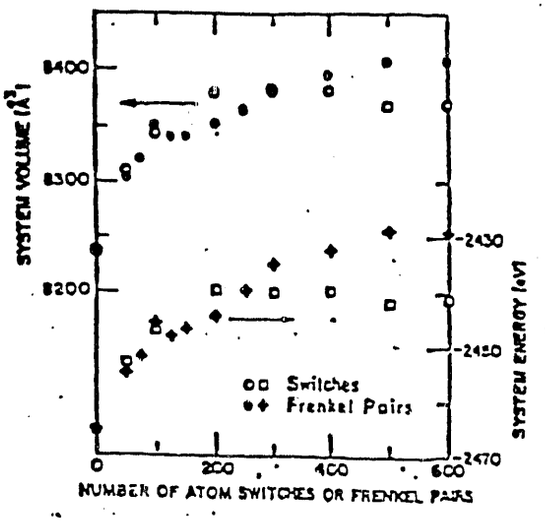


Fig. 2. Volume and energy of the system as a function of the number of switches and the number of Frenkel pairs introduced.

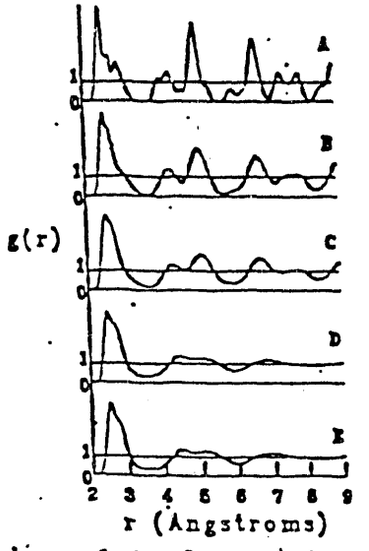


Fig. 3. Pair correlation functions for the CuTi system at 160 K. (a). Perfect lattice. (b) After 600 switches. (c) After 250 Frenkel pairs (d) After 400 Frenkel pairs and (e) quenched liquid.

Lam [9] at various stages during the simulation. After 400 or more Frenkel pairs are introducedthe curve shows features similar to that of the quenched liquid. On the contrary, the PCF after 600 atomic switches shows crystalline features. Based on the above evidence Sabochick and Lam [9] have concluded that chemical disorder by itself cannot cause amorphization in CuTi.

THE SCOPE OF THE PRESENT WORK

The PCF obtained from the above simulation offered information on whether the system went amorphous or not. But it could not offer accurate information on the changes preceding amorphization. The extent of disorder produced by the recombination of Frenkel pairs was also not known. The use of multislice in conjunction with MD makes possible for the first time direct comparisons between experimental results and the results of MD simulations.

The multislice calculation was performed using the system configurations obtained by Sabochick and Lam [9]. The details of the multislice method can be found in Cowley and Moodie [14], Goodman and Moodie [15] and Cowley [16]. The multislice simulation was performed using the Northwestern University Multislice and Imaging System (NUMIS). Diffraction patterns and lattice images were simulated for various configurations obtained using molecular dynamics.

The atomic configurations were also analysed to obtain the changes in long range chemical order during the molecular dynamics simulations. From Figure 1 it can be seen that the structure of CuTi consists of alternating double layers of Cu and Ti perpendicular to the 'c' axis. The atomic positions were sorted to obtain layers of atoms perpendicular to the c axis. In the case of Frenkel pair introduction for 300 or more Frenkel pairs the topological disorder was so great that separation into discrete layers was not possible.

Using this technique it was possible to identify 2 sets of layers which would have been composed respectively of only Cu atoms and only Ti atoms in a perfectly ordered crystal. A Ti atom in a 'Cu layer' or a Cu atom in a 'Ti layer' would be at a wrong site. The long range order parameter (LROP) was defined as the difference between the fraction of atoms on the right sites and that on the wrong sites.

RESULTS

The multislice simulation was performed with the electron beam along the 'c' axis of the CuTi structure (Figure 1). The changes in the diffraction pattern of CuTi as randomly chosen Cu and Ti atoms are switched are shown in Figure 4. There is some streaking of the diffraction spots for 150 or more switches. After 600 switches the diffraction pattern shows spots. On the other hand Figure 5 shows that after 75 or more Frenkel pairs are introduced there is considerable streaking of the diffraction spots. An amorphous halo coexists with diffraction spots at 300 Frenkel pairs. After400 Frenkel pairs are introduced the spots disappear and the diffraction pattern shows only an amorphous halo.

Figure 6 shows the changes in the simulated lattice images as random switches are made. Crystalline contrast is seen after 600 switches. As the number of switches increases some lattice distortion results. This can be inferred from the distortions in the lattice image.

Figure 7 documents the changes in the image for the case of Frenkel pairs. Lattice distortion is significant for more than 75 Frenkel pairs. The contrast begins to show amorphous features at 300 Frenkel pairs. Beyond 400 Frenkel pairs the image is characteristic of an amorphous material.

Figure 8 shows the changes in the long range order parameter for the case of switches and for Frenkel pairs. The LROP is almost the same after about 150 Frenkel pairs and about 125 switches. From Figure 2 it can be seen that the increase in energy for these 2 cases is almost identical. The energy difference between the onset of amorphisation (assuming it to be at about 300 Frenkel pairs) and the near complete destruction of long range order (600 switches) is of the

order of 5 eV. By making a reasonable estimate of about 3 eV for the energy of formation of a

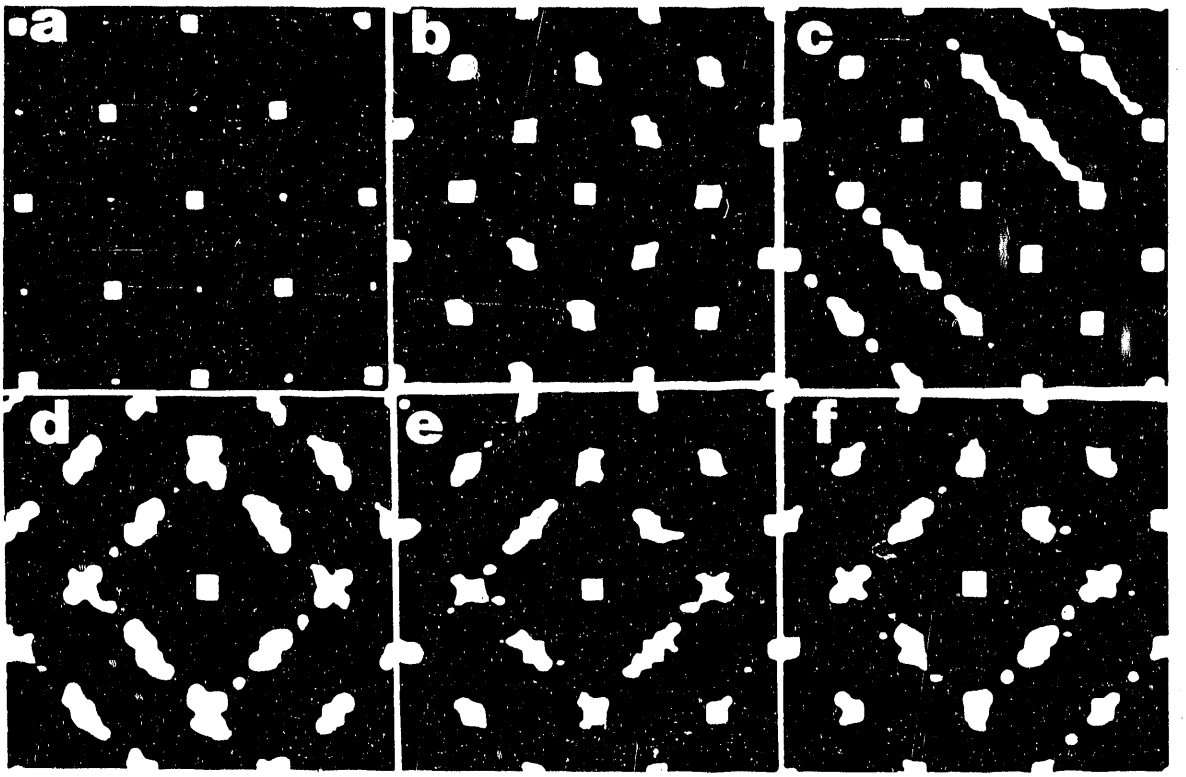


Fig. 4. The changes in the simulated diffraction pattern of CuTi following random switches. (a) Perfect lattice. After (b) 75 switches. (c) 150 switches. (d) 300 switches (e) 400 switches. and (f) 600 switches.

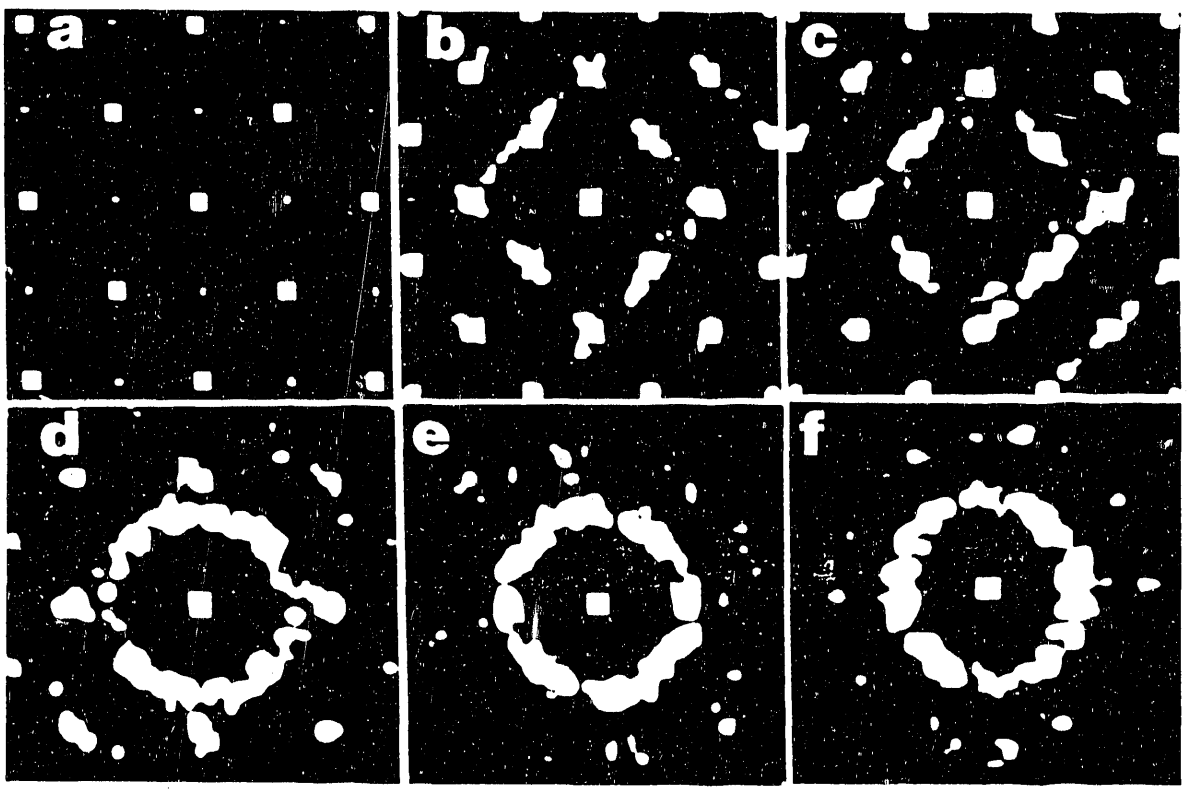


Fig. 5. The changes in the simulated diffraction pattern of CuTi following random introduction of Frenkel pairs. (a) Perfect lattice. After (b) 75 pairs. (c) 150 pairs. (d) 300 pairs (e) 400 pairs. and (f) 600 pairs.

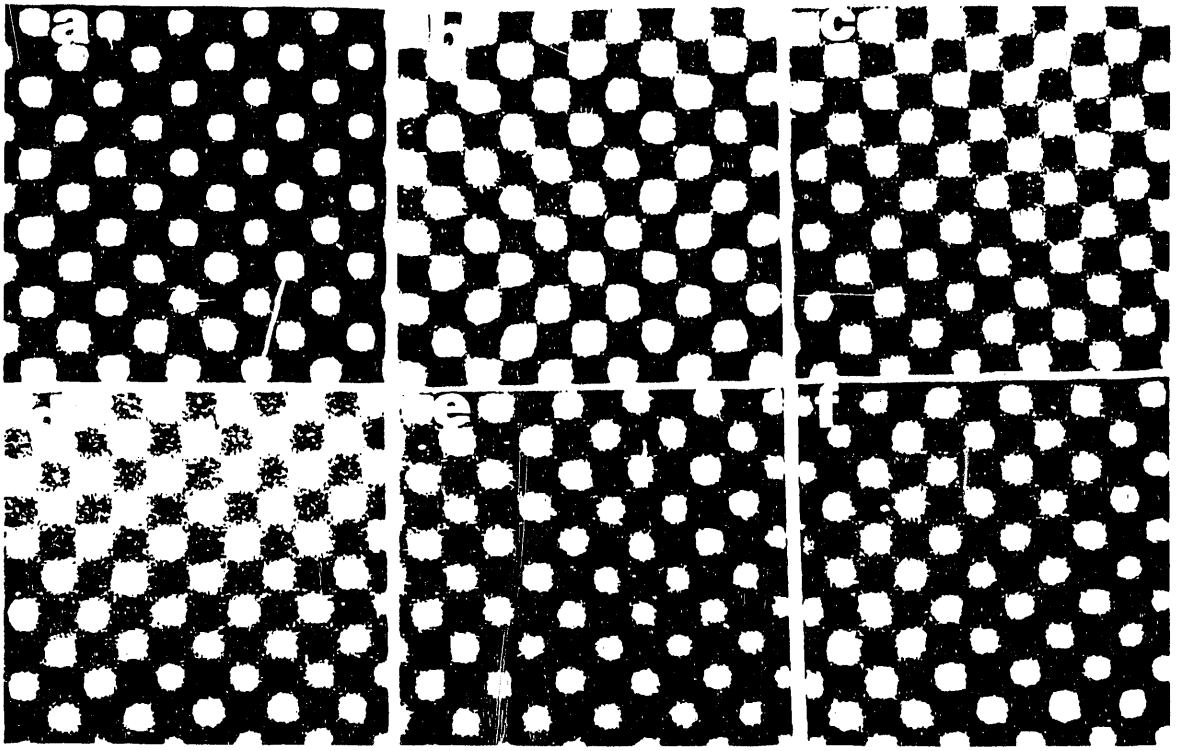


Fig. 6. The changes in the simulated lattice images of CuTi following random switches. (a) Perfect lattice. After (b) 75 switches. (c) 150 switches. (d) 300 switches (e) 400 switches. and (f) 600 switches.

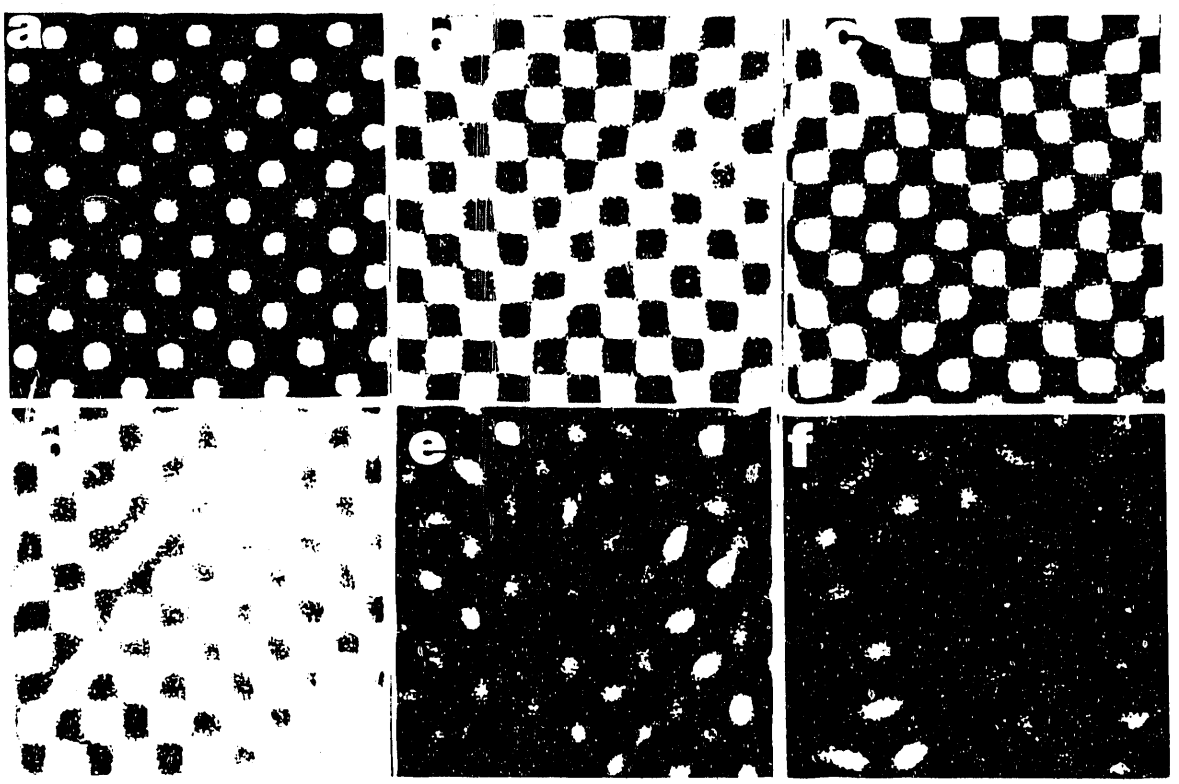


Fig. 7. The changes in the simulated lattice images of CuTi following random introduction of Frenkel pairs. (a) Perfect lattice. After (b) 75 pairs. (c) 150 pairs. (d) 300 pairs (e) 400 pairs. and (f) 600 pairs.

Frenkel pair it can be seen that the survival of atmost 2 Frenkel pairs is sufficient to provide the extra energy required to amorphize the material.

DISCUSSION OF THE RESULTS

In view of the evidence from the lattice images the streaking of the diffraction spots can be related to lattice distortion. It is quite clear that the system remains crystalline after 600 switches. The atom switches merely lead to lattice distortion. On the other hand, the system begins to go amorphous when about 300 Frenkel pairs are introduced. Beyond 400 Frenkel pairs the system is completely amorphous. These results are consistent with the pair correlation functions obtained by Sabochick and Lam [9]. However, the diffraction patterns and lattice images indicate that the present approach is much more sensitive to the changes preceding amorphization than a calculation of the PCF alone.

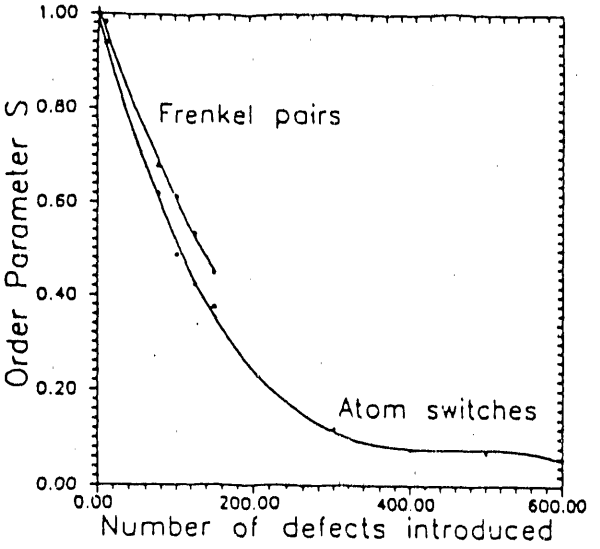


Fig. 8. The effect of switches and Frenkel pair introductions on the long range order of CuTi

The fact that the introduction of Frenkel pairs renders the system amorphous is consistent with the fact that CuTi is amorphized by electron irradiation. The results from the study of long range order (LRO) indicate that the destruction of LRO contributes significantly to the driving force for electron irradiation induced amorphisation of CuTi. However, it is not sufficient by itself to render the material amorphous. The survival of $\approx 5 \times 10^{-3}$ Frenkel pairs is essential to complement the contribution from disorder and drive the material amorphous.

CONCLUSIONS

A new approach that uses the multislice method in conjunction with molecular dynamics simulations has been applied to study the amorphization of CuTi. The calculated diffraction patterns have been found to be more sensitive than the pair correlation function to the structural changes that take place prior to amorphization. Evidence from this approach and from a study of long range order indicates that while chemical disorder plays an important role in the amorphization of CuTi, Frenkel pairs are essential for this process to occur.

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