

Molecular dynamics study of the displacement threshold surfaces and the stability of Frenkel pairs in β -SiC *

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The displacement energy threshold surfaces of Si and C atoms in β -SiC are determined using the molecular dynamics (MD) technique. Average displacement energies of 92.6 and 16.3 eV are calculated for Si and C atoms, respectively. Phase space analysis is used to determine Frenkel pairs separations and to study the effects of the pka initial energy and the initial crystal temperature on the stability of defects produced, within the framework of the nonlinear dynamics theory of systems of many degrees of freedom. For Si pka a slight decrease in the displacement threshold energies is observed as the initial crystal temperature increases from 10 to 100 K. For C pka no significant changes in displacement energies are observed. Channeling of C atoms along the [110] direction is observed to be the only mechanism of producing Frenkel pairs of large separation.

1. Introduction

In a previous study [1], the dynamics of point defect production in β -SiC is studied using the molecular dynamics (MD) technique. A hybrid pair/three-body potential developed by Pearson et al. [2] is used to model interatomic forces. Tersoff [3] has extended his empirical form of potential, developed in refs. [4,5], to study equilibrium β -SiC crystal structure. A qualitative comparison between Pearson's potential and Tersoff's potential is introduced in ref. [1]. Concerning the dynamical effects involved in the process of defect production, Pearson's potential seems more attractive because it includes more parameters which characterize the different combinations of interacting atoms at the pair and triplet levels. It also predicts differences in equilibrium properties for the Si and C atoms in β -SiC, which is more realistic.

The aim of the present work is to complement the previous study [1] by fulfilling the following objectives: (1) to construct the displacement threshold surfaces for Si and C atoms in β -SiC, (2) to determine average displacement energies for both species, and (3) to examine the stability of Frenkel pairs produced in a typical cascade within the framework of the theory of nonlinear dynamics of systems with a large number of degrees of freedom.

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2. Theory

The potential energy function of a crystal consisting of N atoms, assumed by Pearson et al. [2], is written as:

$$\Phi = \frac{1}{2!} \sum_{i \neq j}^N \sum \phi^{(2)}(r_{ij}) + \frac{1}{3!} \sum_{i \neq j \neq k}^N \sum \sum \phi^{(3)}(r_{ij}, r_{ik}, r_{jk}), \quad (1)$$

where $\phi^{(2)}$ is the standard ($m = 12, n = 6$) Lennard-Jones pair potential and $\phi^{(3)}$ is the three-body interaction potential derived by Axillord and Teller [6], and given by:

$$\phi^{(3)} = Z \left(\frac{1 + 3 \cos \theta_i \cos \theta_j \cos \theta_k}{r_{ij}^3 r_{ik}^3 r_{jk}^3} \right). \quad (2)$$

The parameters of the potential for SiC are tabulated in ref. [1]. The values of the pair parameters depend on the pair combination. Also the triplet parameter, z , depends on the triplet combination (i.e., Si-Si-Si, Si-Si-C, Si-C-C, etc.) The interatomic forces are derived by taking the negative of the gradient of interatomic potential. It can be shown that the force acting on the particle i , in the direction α is given by:

$$F_{i\alpha} = - \sum_{j \neq i}^{N_0} \frac{d}{dr_{ij}} \phi^{(2)}(r_{ij}) a_{ij} \cdot \alpha + \sum_{j \neq k \neq i}^{N_0} \sum_{k \neq i}^{N_0-j} \left(- \frac{\partial \phi^{(3)}}{\partial r_{ij}} a_{ij} \cdot \alpha - \frac{\partial \phi^{(3)}}{\partial r_{ik}} a_{ik} \cdot \alpha \right), \quad (3)$$

where a_{ij} and a_{ik} are unit vectors along r_{ij} and r_{ik} . In deriving eq. (3), $\partial \phi^{(3)} / \partial r_{jk}$ is set equal to zero since a

differential change in the position vector of atom i does not result in any change in the vector r_{jk} , which is a result of the central nature of interatomic forces. The range of interaction is taken to be up to the third neighbors. In this case N_0 is 28. The equations of motion for atom i can be written as:

$$\begin{aligned} \dot{q}_i &= F_i, \\ \dot{r}_i &= q_i/m_i, \end{aligned} \quad (4)$$

where q stands for momentum, r for the position referred to a global coordinate system, F is the force, and m is the particle mass. Substituting eq. (3) in eq. (4) and decomposing the momentum and position vectors into three basic components in a global cartesian system, we obtain six first order ordinary differential equations for each particle. Thus, in terms of dynamical systems theory, we have a phase space of dimension $6N$, where N is the total number of particles in the system. The $6N$ variables constitute a vector field in phase space. Equilibrium positions of the particles correspond to the singularities of the vector field, such that $(dq_i/dt \text{ and } dr_i/dt = 0)$; $i = 1, 6N$. The stability of the system can be formally studied by solving a system of nonlinear differential equations of the form $dX/dt = f(X)$, where X is the vector field (position and velocity) and $f(X)$ is a nonlinear function of the vector X . If a Taylor expansion of the vector field is performed around equilibrium and only linear terms are retained, the resulting set of linear ordinary differential equations can be solved to study the stability of the system close to equilibrium. The eigenvalues of the resulting linearized system will determine the nature of stability close to equilibrium.

It has been previously [1] shown that carbon displacements occur in a cascade initiated by a Si pka of energy less or equal to the displacement threshold of the pka itself. The opposite is not true. However, in order to make a practical use of the phase space technique in analyzing atomic collision cascades as dynamical systems, the following simplifications are introduced: (1) For a C pka, a phase space of dimension six (6D phase space) is used to represent the system. These six dimensions comprise the vector field of the pka only. This is fairly valid since a C pka of energy around its displacement threshold does not cause significant perturbation of atoms in its close neighborhood. In other words, sustained phase orbits (dr/dt vs $r(t)$) are presumed for the rest of the particles in the system. (2) For a Si pka, a phase space of dimension $6(n + 1)$, where n is the number of associated C displacements, is required. The criterion for the occurrence of a C displacement is based on a minimum value of the change of the position vector of the C atom of concern, $|\Delta r| = |r - r_0|$, where r_0 is the initial position vector. Whenever the motion of an atom shows symmetry w r t two or three of the global

coordinate axes, the 6D phase space of that atom can be reduced to 4d or 2d space, respectively.

In section 4, phase portraits are presented for a few typical cases to illustrate how Frenkel pairs are produced, and to show the effects of the pka energy and the crystal temperature as most important bifurcation parameters. The CASCMD code [7] is used for computation purposes in the present study.

3. Selection of directions and displacement energy averaging methodology

Fig. 1 shows the basic structure of β -SiC. Although the β -SiC crystal as a whole exhibits cubic symmetry, each individual atom is a center of tetrahedral symmetry only. For a given atom there are two types of $[111]$ direction, or alternatively, along any $[111]$ direction there are two types of lattice sites A and B or Si and C. Displacement surfaces must be defined for each type. In case of a single specie covalent solid (e.g. Si) the displacement surfaces for both A and B atoms are mirror images of each other in the (111) plane which bisects a nearest neighbor bond [8]. However, in β -SiC the situation is different because A and B atoms are not alike.

It is interesting to note that (see fig. 1) the four vertices of the tetrahedron are $[111]$ type directions. The centers of the tetrahedral faces, previously mentioned as the $\langle 111 \rangle$ gaps [1], are also $[111]$ type directions. For the sake of clarity, the directions of the first neighbors are always denoted as the $[11\bar{1}]$ directions, while the $\langle 111 \rangle$ gap center directions are simply denoted as $[111]$ directions. This notation is valid for both kinds of atoms in β -SiC and will be used throughout the rest of this work. Since the four tetrahedral faces are equivalent in terms of lattice symmetry, it is sufficient to investigate the atomic displacement behavior over one tetrahedral face only. Moreover, because of the symmetry over each tetrahedral face, only the solid angle bounded by a tetrahedral face center $[111]$, a first

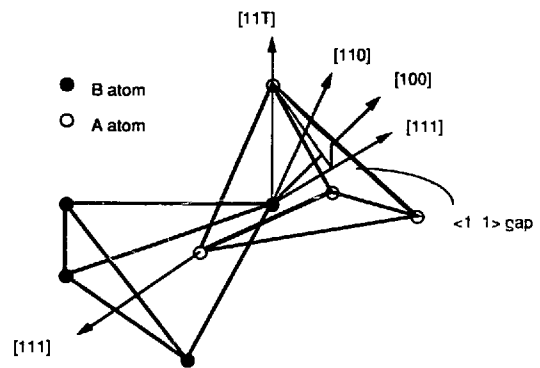


Fig 1

neighbor $[11\bar{1}]$ and a $[100]$ direction, is required for the analysis. The solid angle of concern is $\Omega_1 = \pi/6$. An average displacement energy, $\langle E_d \rangle$, can be defined as:

$$\langle E_d \rangle = \frac{1}{\Omega_1} \int_{\Omega_1} E_d(\Omega) \cdot d\Omega = \frac{6}{\pi} \sum_i E_{d_i} \Delta\Omega_i, \quad (5)$$

where the subscript i represents a given direction within Ω_1 .

4. Results and discussion

In fig 2 the directional dependence of the displacement threshold energies for Si and C atoms is shown. The atomic displacement threshold energies are determined to accuracies of ± 4 eV for silicon and to ± 1 eV for carbon. In general, a Si pka shows much higher displacement threshold energy along all directions. Average displacement threshold energies are found to be ~ 92.6 eV for silicon and ~ 16.3 eV for carbon. Considering the atomic mass differences as well, this implies high nonstoichiometry for large scale cascades in β -SiC with the majority of displacements being of carbon type. The differences in displacements threshold energies of Si and C atoms can be explained in terms of the interaction parameter z used in eq. (2). It is ~ 20 times higher for Si triplets, which means that the interaction energy and the interatomic forces on the Si atoms are higher by the same factor. This results in higher energy transfer rates from a fast moving Si atom (pka) to the atomic neighborhood implying much higher initial energy for a Si displacement event.

An important parameter of this problem is the number of stable carbon displacements, n , produced in a cascade initiated by a Si pka. The value of n varies dramatically, depending on the pka direction. At the displacement threshold of the Si pka, n takes on many values between two and twelve for different pka direc-

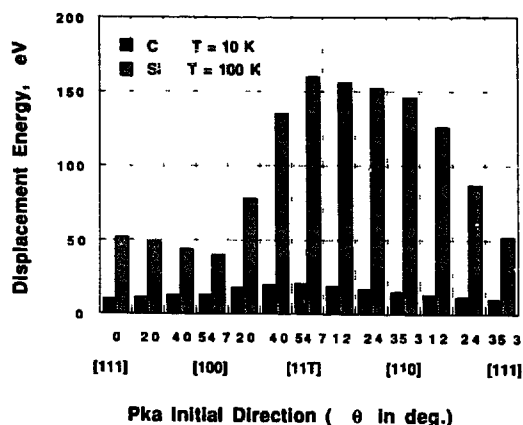


Fig 2

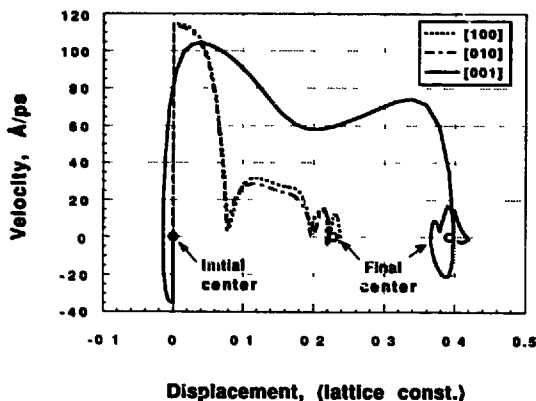


Fig 3.

tions. The lowest value occurs for pkas along the $[100]$ directions and the highest value occurs for pkas along the $[110]$ directions. However, this feature brings in the effects of crystal structure in calculating the damage function in SiC, at least in the sub-keV range of pka energy.

Displacements are represented by sustained orbits (centers) in phase space around singular points (oscillations around final equilibrium sites). The size and shape of the orbit depend on the momentum vector components of the interstitial atom as it first approaches that site. Fig 3 shows phase space portraits for a C pka of energy of 17 eV. The initial direction is 12° with the $[110]$ direction and 23.3° with the $[11\bar{1}]$ direction in the plane containing the two directions. The motion of the C atom is symmetric with respect to the global $[100]$ and $[010]$ directions. A Frenkel pair separation distance can be found directly from the phase space as the distance between stable limit cycles. For the atom of fig. 3, the Frenkel pair separation is $\sim 0.51a_0$. In fig 4 we compare the displacement be-

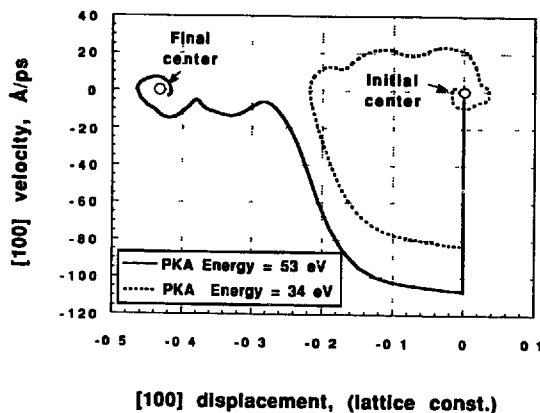


Fig 4.

havior of two Si pkas of energy 52 eV (superthreshold) and 34 eV (subthreshold) along the [111] direction by phase space illustration. The atomic motion in both cases is symmetric with respect to the three global coordinate axes: [100], [010] and [001] directions. It is shown that atomic displacement occurs at 52 eV, while at 34 eV the pka relaxes back towards the original equilibrium site.

The effect of the initial crystal temperature on the displacement behavior is also investigated. The displacement threshold energy along a particular direction undergoes a *slight* decrease as the temperature increases. As an example, a 5 eV decrease in the displacement threshold along the [110] direction is observed for Si pka between 10 and 100 K. Also the final cascade configuration is strongly dependent on the initial temperature in cascades initiated by Si pkas. No significant temperature effects are observed for C pkas.

Perhaps the most important result of the present study is the verification of the existence of channelling of energetic atoms. Channelling of C atoms is observed along the [110] direction, which results in stable Frenkel pairs of relatively long separation. The cross section of the channelling part is hexagonal (50% silicon, 50% carbon). According to our experience on cascade dynamics in β -SiC, channelling of C atoms along the [110] direction is the only atomic displacement mechanism that can produce stable Frenkel pairs of large separation. However, in terms of the Frenkel pair separation, channelling can be considered as the counterpart of the replacement collision sequences observed along the [110] direction in fcc copper [7]. Because a fast moving Si atom has no transfer its energy to the atomic neighborhood at significantly higher rates, channelling of this kind of atoms in SiC is not likely to occur since most of the atoms comprising the channelling path can be displaced

5. Conclusions

Conclusions of the present study are summarized as follows:

(1) Phase space analysis reveals that for pka energies above the threshold displacement value, the solution trajectory is a bifurcation of the dynamical system leading to a new stable limit cycle, while stable limit cycles exist for all atoms with energies below E_d .

(2) The average values of E_d are 16.3 ± 1 and 92.6 ± 4 eV for carbon and silicon, respectively.

(3) For Si pkas, a slight decrease in the displacement threshold energies is observed as the initial crystal temperatures increases from 10 to 100 K. No effects are observed in the case of C pkas.

(4) In most of the cases studied here, displaced atoms come to equilibrium within the same unit cell, i.e., the separation of a Frenkel pair is less than a lattice constant. However, channelling of C atoms is observed along the [110] direction, and it is the only mechanism observed to produce stable Frenkel pairs of relatively larger separation.

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