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ANISOTROPIC DISPLACEMENT THRESHOLD ENERGIES
IN SILICON BY MOLECULAR DYNAMICS SIMULATIONS*

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LeAnn A. Miller,¹ David K. Brice,¹ Anil K. Prinja,² and S. Thomas Picraux¹

¹Sandia National Laboratories, Albuquerque, NM

²University of New Mexico, Department of Chemical and Nuclear Engineering, Albuquerque, NM

Abstract

A combination of molecular dynamics simulations and theoretical modeling was used to examine the orientation dependent threshold energies for displacement of silicon atoms from their lattice site due to energetic particle collisions. These results are important for a detailed understanding of both radiation effects in silicon devices and beam-enhanced stimulation of molecular beam epitaxial growth.

The molecular dynamics code developed for this study, which employs a Tersoff interaction potential, as well as the theoretical model that incorporates the symmetry of the crystal are described.

Bulk displacement threshold energies were determined by the molecular dynamics code for four directions through the open face in the $\langle 111 \rangle$. These values were then incorporated into the theoretical model for the average bulk displacement threshold energy. The average bulk displacement threshold energy was found to be 14.8 eV in 30° about $\langle 111 \rangle$ and 11.1 eV in 20° about $\langle 100 \rangle$.

Introduction

Models of radiation damage in crystalline materials typically assume a sharp energy threshold, E_d , for initiating atomic displacements from their ideal lattice sites [1]. The directional dependence of the displacement threshold energy is important in the development of a detailed understanding of both radiation effects in silicon devices [2] and beam-enhanced stimulation of molecular beam epitaxial growth [3]. To yield a better understanding of this phenomenon a combination of theoretical modeling and Molecular Dynamics (MD) simulations was used. This involved developing a MD code and a theoretical model of the displacement energy. Both of these tasks are discussed below as well as the application of the method to compare our results with previous experimental measurements and modeling [4].

Theoretical Model

Experimental data for silicon was compared with several simple models for the bulk displacement threshold energy, $E_d(\theta, \phi)$, in reference 2. The comparisons favored a model in which the average value of the bulk displacement threshold energy over a 30° cone of angles centered on $\langle 111 \rangle$, toward the open face of the

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basic tetrahedron (see Figure 1), was 22 eV. The average value of E_d over a 20° cone of angles centered on $\langle 100 \rangle$ was 26 eV, while displacements were not allowed outside these cones. We expand $E_d(\theta, \phi)$ as a function of the polar angle, θ , and azimuthal angle, ϕ , about these two crystallographic directions in order to take advantage of crystal symmetry in evaluating the average value, $\langle E_d \rangle$, for comparison with these previous studies.

For polar angle θ measured from the $\langle 111 \rangle$ direction, $E_d(\theta, \phi)$ may be expanded as

$$E_d(\theta, \phi) = \sum_{n=0}^{\infty} \sin^{3n}\theta \cos(3n\phi) \sum_{k=0}^{\infty} Q_{2k}^n \sin^{2k}\theta \quad (1)$$

where the Q_{2k}^n are expansion coefficients and the threefold symmetry in ϕ is explicit in the expression. Similar expansions may be written for other symmetry directions, in particular for $\langle 100 \rangle$, but we illustrate the procedure here only for the $\langle 111 \rangle$ axis. For values of θ near $\langle 111 \rangle$, equation 1 can be approximated by

$$E_d = Q_0^0 + Q_2^0 \sin^2\theta + Q_4^0 \sin^4\theta + Q_0^1 \sin^3\theta \cos(3\phi) \quad (2)$$

Figure 1. Basic tetrahedron of silicon. The central atom is directed through the open face indicated by the dashed lines.

where terms of order $\sin^3\theta$ and higher order have been neglected. Equation 2 provides an approximate expression for $E_d(\theta, \phi)$ accurate to order $\sin^4\theta$, the coefficients of which can be evaluated from our molecular dynamics calculations of E_d for four directions near $\langle 111 \rangle$. Averaging equation 2 over a cone of angular radius θ_m yields

$$\begin{aligned} \langle E_d \rangle = Q_0^\circ + \frac{Q_2^\circ}{3(1 - \cos\theta_m)} [2 - \cos\theta_m(\sin^2\theta_m + 2)] \\ + \frac{Q_4^\circ}{5(1 - \cos\theta_m)} \left[\frac{4}{3} (2 - \cos\theta_m(\sin^2\theta_m + 2)) - \sin^4\theta_m \cos\theta_m \right] \quad (3) \end{aligned}$$

Using $\theta_m = 30^\circ$ in equation 3 provides values for $\langle E_d \rangle$ which can be directly compared with the values from [2]. A similar procedure provides a MD value for $\langle E_d \rangle$ for the $\langle 100 \rangle$ direction.

Molecular Dynamics Code

As part of this study, a MD code was developed. The code includes a crystal generation routine in which a diamond crystal of any size is generated by simply inputting the desired number of cubic unit cells in each of the three directions. The crystal generated is an ideal crystal (i.e., all atoms are at their equilibrium site). The velocity of each atom is then determined by randomly sampling the Maxwell-Boltzmann distribution corresponding to the crystal temperature.

Following initialization of the atom positions and velocities, the actual integration of the equations of motion of the atoms begins. The Tersoff potential [4] was chosen for these simulations because it is relatively straight-forward to code, and parameters are available for both silicon and germanium.

Since each atom is affected only by those atoms that are within the range of the potential, a large list of "neighbors" is maintained for each atom and updated periodically throughout the simulation. This large list is reduced each time step to those atoms within the cutoff of the potential of the atom of concern during that time step. Only these atoms are used in the force calculation as a method of increasing computational speed.

The force on an atom is calculated by determining the potential energy of the crystal when the atom is in six different positions (positively and negatively displaced in x, y, and z). A finite difference of these values yields the gradient of the potential in each direction from which the force in each direction may be determined.

The integration procedure implemented uses information from two time steps and has the advantage that only one force evaluation is necessary per time step [5]. The formula is fourth order accurate in time step.

Periodic boundary conditions may be applied to any of the three directions. These conditions assume that the infinite crystal is a three-dimensional, periodic repetition of the computational cell.

The time step is allowed to vary to increase the speed of the simulation. An intermediate time step is inversely proportional to the maximum velocity. The new time step is a linear combination of the old and intermediate time steps. The advantage of this method is that longer time steps are taken as the maximum velocity decreases.

Calculations

The bulk displacement threshold energy was determined for four different directions through the open face in the $\langle 111 \rangle$. The four directions were $\langle 111 \rangle$, $\langle .7041 .7041 .0915 \rangle$, $\langle 100 \rangle$, and $\langle .9397 .2418 .2418 \rangle$. The calculations were performed with the above described code under the following conditions:

- 1) the temperature of the crystal was initially 0 K,
- 2) a single atom was given an initial velocity in the desired direction,
- 3) the crystal consisted of 608 atoms so that the temperature of the crystal, at thermal equilibrium, was approximately room temperature, and
- 4) periodic boundary conditions were applied in all three directions.

The energy given to the atom was varied until the bulk displacement threshold energy was determined. This process was repeated for each of the different directions.

Results

The results of the calculations for the bulk displacement threshold energies for the different directions are contained in Table I.

Table I. Bulk displacement threshold energies from molecular dynamics calculations. Values of θ and ϕ are for the $\langle 111 \rangle$ axis. Values for θ and ϕ in parenthesis are for the $\langle 100 \rangle$ axis.

<u>Direction $\langle hkl \rangle$</u>	<u>θ ($^\circ$)</u>	<u>ϕ ($^\circ$)</u>	<u>E_d (eV)</u>
$\langle 111 \rangle$	0. (54.74)	0. (90.)	11.4
$\langle .7041 .7041 .0915 \rangle$	30. (45.24)	0. (52.42)	18.3
$\langle 100 \rangle$	54.74 (0)	60. (0.)	10.1
$\langle .9397 .2418 .2418 \rangle$	57.14 (20)	35.97 (0.)	12.5

These bulk displacement threshold energies were used to determine the average bulk displacement threshold energy about the $\langle 111 \rangle$ and $\langle 100 \rangle$ for angles of 30° and 20° , respectively, using the theoretical model discussed above. This resulted in an average bulk displacement energy through the open face of 14.8 eV in the $\langle 111 \rangle$ and 11.1 eV in the $\langle 100 \rangle$.

The values of E_d appearing in Table I are accurate to 0.1 eV as determined by the MD calculations. Values of $\langle E_d \rangle$ obtained using the approximate equation 2 and the points in Table I are estimated to be accurate to approximately 15%.

Discussion and Conclusions

Comparison of the MD values for $\langle E_d \rangle$ with those obtained in [2] reveal that the present results are approximately a factor of 2 smaller (14.8 eV vs. 22 eV in the $\langle 111 \rangle$ and 11.1 eV vs. 26 eV in the $\langle 100 \rangle$). A simple explanation can account for this disparity. The experimental measurements used to evaluate $\langle E_d \rangle$ in [2] (carrier removal rates versus incident electron energy) were carried out at 290 K where the stable lattice damage in silicon is known to consist primarily of secondary defects such as di-vacancies [6]. Furthermore, the experiments in [6] suggest that di-vacancies form from agglomeration of single vacancies, rather than being produced directly by the incident irradiation. Thus, the stable damage is a result of single vacancy formation in which the vacancy-interstitial pair are initially sufficiently well separated that recombination is unlikely.

In contrast to the conditions of the experimental measurements, the criterion used in our calculations to signify lattice atom displacement were that the lattice atom should be removed from the tetrahedron of initial nearest neighbors and should come to thermal equilibrium with the surrounding lattice before recombination could occur. Thermal equilibrium generally occurred during the first 100 fs of our simulation, and calculations were generally continued to approximately 1 ps. The vacancy-interstitial pair were separated in all cases by less than a lattice spacing at the end of the calculation.

With this explanation our results are thus not directly comparable with the values obtained in [2]. We feel, however, that our results are the more fundamental since they indicate the true displacement thresholds for single vacancy formation. Furthermore, in applications to molecular beam epitaxial growth these close vacancy-interstitial pairs may exert a significant influence on the evolution of the morphology of the surface.

A better experimental test of our calculations would be a comparison with damage threshold measurements below 100 K where single vacancies are thermally stable. At present such measurements seem to be unavailable in the literature. While such comparisons would also be complicated by the mobility of the silicon interstitial at these temperatures [7], the values of $\langle E_d \rangle$ might yet be accessible to appropriate experimental measurement.

An alternate explanation for the difference between our results and those of [2] would call into question the adequacy of the Tersoff potential [4] for the present calculations. The potential, however, quite adequately accounts for the cohesive energy, the bulk moduli, and the lattice phonon spectrum. Adjustments to the potential to account for the factor of 2 difference in our E_d values without compromising these predictions seems unlikely. We thus reject this possibility.

A more dense selection of points at which E_d is evaluated is planned for further application of our MD code in order to refine our estimates of $\langle E_d \rangle$, and to provide a better approximation to the function $E_d(\theta, \phi)$. Additional estimates of the accuracy of our results awaits further low temperature experiments.

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