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Large-Scale Molecular Dynamics Simulations of Interstitial Defect Diffusion in Silicon

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ABSTRACT

The simulation of defect dynamics and evolution is a technologically relevant challenge for computational materials science. The diffusion of small defects in silicon unfolds as a sequence of structural transitions. The relative infrequency of transition events requires simulation over extremely long time scales. We simulate the diffusion of small interstitial clusters (I_1, I_2, I_3) for a range of temperatures using large-scale molecular dynamics (MD) simulations with a realistic tight-binding potential. A total of 0.25 μ sec of simulation time is accumulated for the study. A novel real-time multiresolution analysis (RTMRA) technique extracts stable structures directly from the dynamics without structural relaxation. The discovered structures are relaxed to confirm their stability.

INTRODUCTION

Transient enhanced diffusion of dopants in the presence of extended {311} defects in silicon is a limiting factor in the fabrication of shallow junction devices [1]. The growth of such extended defects involves the diffusion, capture and dissociation of silicon point defects [2-4]. The precise mechanism for nucleation of these defects from small interstitial clusters is less clear.

The dynamics of these defect proceed through thermally induced infrequent events, making direct molecular dynamics simulation difficult. The infrequency of events requires very long simulation times to accumulate statistically meaningful data. Furthermore, the important information in the simulation data (structures and transitions) is buried within the thermal noise in the system.

We avoid the use of *ab initio* MD due to its present limitation in accessible simulation time. As reported in Ref.[5], even the "fast" diffusive mechanisms in these systems occur on the picosecond time-scale, and one is fortunate to observe a few events during a single, costly simulation. As a consequence, the *ab initio* MD presently offers only a glimpse at the total picture of I_n cluster diffusion.

It is for this reason that we use tight-binding MD simulations which offer an increase in the accessible time-scale of two orders of magnitude. Our approach is to use tight-binding MD simulations to explore the diffusion of small interstitial clusters and validate the results with more accurate *ab initio* calculations.

METHOD

Tight-binding molecular dynamics

The diffusion of single-interstitial (I_1), di-interstitial (I_2) and tri-interstitial (I_3) clusters is studied using tight-binding molecular dynamics simulations as implemented within our OHMMS code[6]. Each I_n cluster is modeled using a 64+n atom supercell with a lattice constant of 10.864 Å and periodic boundary conditions. The silicon-silicon interaction is modeled with the tight-binding potential of Lenosky *et al.*[7]. Constant temperature runs are performed using a Langevin propagator with a 2 fs time-step. Defect diffusion is studied at temperatures of 800 K, 900 K, 1000 K and 1100 K. Total simulation time of 20 ns is accumulated at each temperature in 0.5 ns (262,144 MD step) runs. The initial configuration for each run is a compact I_n cluster in what is presumed to be the ground state configuration designated here as I_n^a. The system is thermalized for 1000 MD steps to eliminate transient effects.

The entire dynamics for each simulation is recorded for post-processing analysis using a wavelet-based real-time data compression scheme. The technique involves performing a time-domain wavelet transform of the time-series corresponding to each atomic coordinate. The algorithm used is more efficient than conventional fast wavelet transform techniques and designed for treating massive streaming datasets with minimal computational overhead. In order to implement this on-the-fly data compression scheme, the original MD code was retrofit with optimized library routines. The integration is seamless, involving only the replacement of previous "write statements" used to record the dynamics by more conventionally means.

Detection of stable structures

Stable structures are identified directly in the dynamics using real-time multiresolution analysis (RTMRA) techniques[8]. A modified wavelet transform is applied in the time-domain to detect temporal features of stability and transition in the system. The technique provides an analysis of the dynamics over multiple time scales. The computational efficiency of the algorithm allows its application during an ongoing simulation. This technique for directly detecting stable structures in the dynamics replaces the conventional technique of periodic structural relaxation. We find that the detected structures closely approximate the true local minimum configurations, and any subsequent verification of stability through structural relaxation is correspondingly accelerated. The advantages of the RTMRA techniques are two-fold. First, the computational cost involved with periodic relaxation of the system merely for the purpose of identifying the current state is all but eliminated in our approach. Second, the RTMRA technique imposes no specific time-scale on the detection of stability or transition. Instead, all time-scales are examined within the implicit dyadic hierarchy of the underlying wavelet basis.

The stability of discovered structures are verified by relaxing each structure within the same tight-binding Hamiltonian used in the MD simulation. These relaxations require only a modest effort since the detected configurations are very close to the relaxed local minimum energy configurations. Typical $|\Delta R|_{\max}$ and $|\Delta E_f|_{\max}$ for such relaxations are 0.1 Å and 0.2 eV, respectively. Finite-size effects are checked by repeating each relaxation

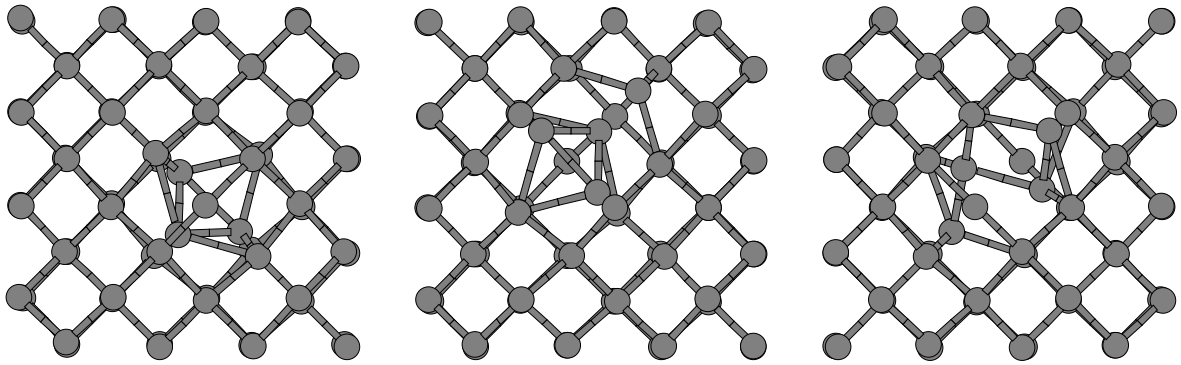

Figure 1. RTMRA detected I2 structures: (a) I_2^a, (b) I_2^{a*} and (c) I_2^b .

within a larger 216+n atom cell; these relaxations yield no significant change in the configuration or stability of the discovered structures.

RESULTS

Single-interstitial

From the I_1 diffusion constant extracted from the dynamics at each temperature we estimate an effective activation energy for the I_1 of 0.83 eV. Within the recorded dynamics from I_1 diffusion simulations we detect two stable structures: H and $\langle 110 \rangle$-split structures [7]. The dominant structure H is detected more frequently by a factor of 10, as measured in terms of relative percent-time detected.

Di-interstitial

From the I_2 diffusion constant extracted from the dynamics at each temperature we estimate an effective activation energy for the I_2 of 0.52 eV. Within the recorded dynamics from the I_2 diffusion simulations we detect the three structures shown in Fig. 1. The structures are relaxed to verify their stability. The dominant I_2^a structure may be identified with the previously reported minimum energy structure with C_{1h} symmetry[7]. The structure designated here as I_2^{a*} may be described as an I_2^a with a single additional atom displaced from a bulk lattice site. The I_2^{a*} has the same C_{1h} symmetry as the I_2^a. The structure designated here as I_2^b is similar, but not identical to the Z structure reported in Ref.[9]. The I_2^b has C_{2h} symmetry and is more extended than the I_2^a.

The detection of the I_2^a dominates the other two structures in relative percent-time detected by approximately a factor of 4, with the detection of the I_2^{a*} and I_2^b being roughly equal.

Tri-interstitial

From the I_3 diffusion constant extracted from the dynamics at each temperature we estimate an effective activation energy for the I_3 of 0.48 eV. When stable structures are

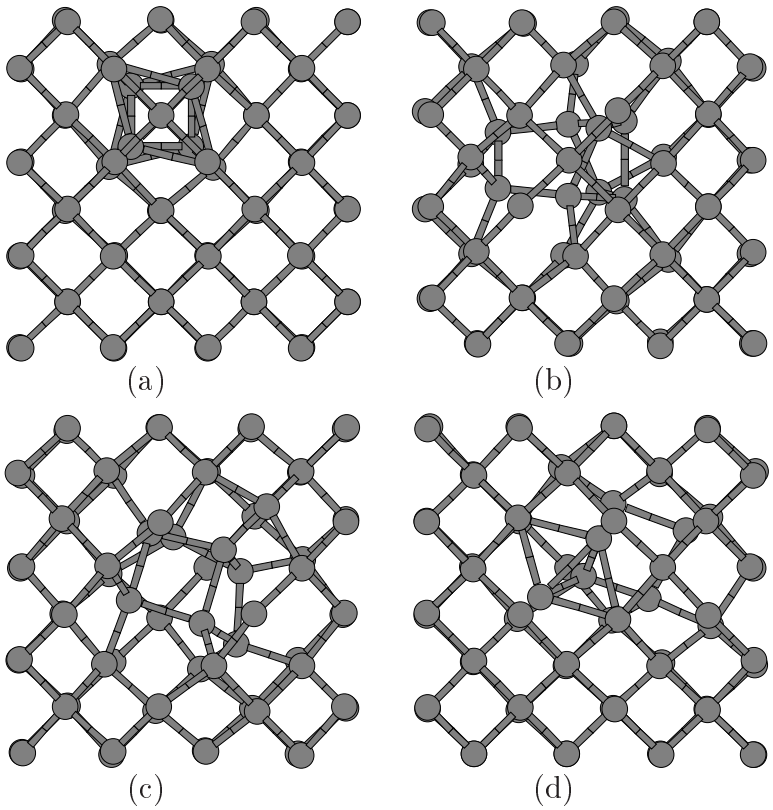

Figure 2. RTMRA detected I_3 structures: (a) I_3^a, (b) I_3^b, (c) I_3^c, and (d) I_3^d.

extracted from the dynamics, they are quite different from those observed for the I_1 and I_2. For the I_1 and I_2, the diffusion may be characterized by transitions between a few (two and three, respectively) compact, stable structures. For the I_3 we detect a rich and complex catalog of defect structures ranging from compact to extended, chain-like defects. Moreover, we identify $I_2 + I_1$ fragments from the temporary dissociation and re-formation of the I_3 as it diffuses. From the I_3 diffusion simulations we detect 10 unique structures. Each discovered structure is relaxed to confirm its stability.

The four detected structures corresponding to a stable I_3 are shown in Figure 2. The dominant structure designated here as I_3^a is identified as the Ref. [10] compact cluster with D_{2d} symmetry and thought to be the low energy ground state. The other discovered I_3 structures include an extended structure (I_3^c) with a relatively low formation energy within LDA only 0.21 eV higher than the ground state, and a compact structure (I_3^d) with a formation energy within LDA 0.51 eV higher than the ground state.

CONCLUSION

The use of tight-binding molecular dynamics simulations enables an extensive search for structures and transitions within the dynamics of small silicon self-interstitial cluster diffusion. Integrated real-time data compression allows the full dynamics to be recorded for detailed post-processing analysis. RTMRA techniques allow stable structures to be

detected directly in the dynamics without costly periodic structural relaxation. The detected structures are very close to the relaxed configurations. In the dynamics of single-, di- and tri-interstitials, this approach allowed the identification of new stable structures.

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