Modeling Radiation Damage in Semiconductor SiC

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The steepest-entropy-ascent quantum thermodynamic (SEAQT) framework is used to investigate the kinetics of radiation damage to a silicon carbide (SiC) semiconductor. The energy eigenstructure is developed by the Replica-Exchange Wang-Landau (REWL) algorithm that determines the density-of-states of a discrete system. The REWL method is able to provide the various energies of a 3C-SiC cubic crystal system comprised of 1000 atoms. Scaling up to larger systems is in progress. Currently, the radiation damage mechanisms include vacancies and interstitial atoms ($\langle 100 \rangle$ dumbbells). The model predicts the rate with which clusters form during primary damage from irradiation. Using the energy eigenstructure provided by the REWL algorithm, the SEAQT equation of motion determines the unique non-equilibrium path taken by the system, providing insights into its microstructural evolution due to radiation damage.

I. INTRODUCTION

Silicon carbide (SiC) is a wide bandgap (WBG) semiconductor that offers desirable properties including high electrical conductance, high thermal stability, and high-power switching performance [1–3]. It is known to have many different crystal structures or polytypes (hexagonal, cubic, and rhombohedral) that differ from each other in the stacking sequence of a closed-packed plane. Each structure has somewhat different properties [4]. Although a candidate material for semiconductors and widely used in automotive applications, it is susceptible to solar radiation damage in space or avionic devices. There have also been numerous studies on its radiation resistance as a cladding material for the nuclear industry [5] . Under irradiation, point defects accumulate into defect clusters that ultimately lead to failure [6]. Many studies of WBG semiconductors [7, 8] utilize information on SiC under nuclear radiation as a baseline for possible damage mechanisms involved with solar radiation. Because of the covalent nature of SiC, the damage mechanisms differ quite significantly from metals under irradiation and are less prevalent. Noticeable radiation effects in SiC are the formation of anti-site defects, which entail a Si atom occupying a C atom site and vice versa. Also, single interstitials occupying tetahedral sites, along with dumbbells in the $\langle 100 \rangle$ orientation can form.

It is known that SiC devices have high resistance to total dose radiation [8] but are more affected by transient radiation effects, also known as single-event effects (SEE) that occur in space. The SEE are caused by high-energy protons or heavy ions in space, whereas neutrons are the main cause of radiation damage in nuclear reactors [2]. Simulating SEE by experimental techniques is costly and difficult, and there is not yet much modeling of solar radiation effects on SiC. In terms of numerical simulations, β-SiC (or the 3C-SiC polytype) is modeled with classical molecular dynamics (MD) for primary damage from radiation exposure [6, 9–11], which is a good technique for understanding the damage mechanisms by solar radiation.

Conventional computational modeling, however, has limitations that include small length and time scales and an assumption that the material is near equilibrium. With irradiation, non-equilibrium kinetic phenomena can be computationally burdensome when they rely on transport equations (diffusion, heat, or mass fluxes). Utilizing an alternative approach called SEAQT, there is no need to assume linear transport laws or near-equilibrium. SEAQT can predict a thermodynamically unique kinetic path from an initial non-equilibrium state to stable equilibrium based on the steepest-entropy-ascent (or maximal entropy production) principle at each instant of time. Its equation of motion predicts the probability distributions of each state in time and is, thus able to track the changing extensive property values of energy and entropy [12]. In this study, the rates of defect accumulation predicted with SEAQT equation of motion are compared with existing MD studies on 3C-SiC for $\langle 100 \rangle$ dumbbells. Plans for including other defects (anti-sites and single interstitials) are discussed.

II. METHOD

In order to determine the kinetics of radiation damage using the SEAQT equation of motion, construction of the energy eigenstructure is needed. For the purposes of this paper ony, a small demonstration system is used that consists of β-SiC (3C-SiC) with 1000 atoms in a $5 \times 5 \times 5$ unit cell with dimensions of $21.8 \times 21.8 \times 21.8$ Å. On-going work includes much larger systems using a combination of the Replica-Exchange Wang-Landau (REWL) algorithm [13, 14] coupled to LAMMPS [15] and neural networks. For the present system, only the former (i.e., REWL/LAMMPS) is needed to find all pos-

sible energy levels, e_k, and their degeneracies, g_k, from the ground state to a Monte Carlo-determined maximum energy. The energy levels and their statistically determined degeneracies constitute the density of states used by the SEAQT equation of motion. Associated with each energy eigenlevel is a set of microstructural configurations arising from dumbbell and vacancy locations in the LAMMPS simulation block. The REWL is a parallelization of the original the Wang-Landau algorithm [16–18], which is a stochastic non-Markovian Monte Carlo approach. The combination of REWL and LAMMPS allows the program to read in numerous interatomic energy potentials from NIST database files. As in the case here for 3C-SiC, a Tersoff potential is used for the Hamiltonian expressed as

$$e_k = \frac{1}{2}\sum_i \sum_j V_{ij} \tag{1}$$

where V_{ij} represents the two-body interactions between three bodies and the summations are over all the neighbors within a cutoff distance.

To model radiation damage with REWL, a Potts model is employed to characterize states corresponding to SiC defects, such as the $\langle 100 \rangle$ dumbbell orientation for C^+-C, C^+-Si, Si^+-C, and Si^+-Si [6], where the superscript + is the interstitial atom. For each Monte Carlo trial move, atoms are randomly chosen to form a dumbbell and the energy is computed and stored to generate the set of e_k for possible configurations of the simulation block and their respective degeneracies, g_k.

A. SEAQT Equation of Motion

The energy eigenstructure developed by the REWL method can be used to solve the SEAQT equation of motion for a simple quantum system [19, 20], namely,

$$\frac{d\hat{\rho}}{dt} = -\frac{i}{\hbar}[\hat{H}, \hat{\rho}] + \frac{1}{\tau(\hat{\rho})}\hat{D}(\hat{\rho}) \tag{2}$$

where $[\cdot, \cdot]$ is the commutator; $\hat{\rho}$ the density operator, which for classical systems reduces to a probability distribution; t the time; i the imaginary unit; $\hbar$ Planck's modified constant; $\hat{H}$ the Hamiltonian operator expressed classically in non-operator form by Eq. (1); $\hat{D}$ the dissipation operator that describes the non-linear dynamics of irreversible state evolution; and τ the relaxation parameter.

For an isolated classical system, no quantum correlations are present and, thus, $\hat{\rho}$ and $\hat{H}$ commute so that the equation of motion can be expressed when the only generators of the motion are $\hat{H}$ and the identity operator $\hat{I}$ as [12, 21, 22]

$$\frac{dp_j}{dt} = \frac{1}{\tau}\frac{\begin{vmatrix} s_j & p_j & e_j p_j \\ \langle s \rangle & 1 & \langle e \rangle \\ \langle es \rangle & \langle e \rangle & \langle e^2 \rangle \end{vmatrix}}{\begin{vmatrix} 1 & \langle e \rangle \\ \langle e \rangle & \langle e^2 \rangle \end{vmatrix}} \tag{3}$$

where $\langle \cdot \rangle$ represents an expectation value and the system energy, $\langle e \rangle$, and entropy, $\langle s \rangle$, are given by

$$\langle e \rangle = \sum_k e_k p_k \tag{4}$$

$$\langle s \rangle = \sum_k p_k s_k = -\sum_k p_k \ln \frac{p_k}{g_k} \tag{5}$$

$$\langle e^2 \rangle = \sum_k e_k^2 p_k \tag{6}$$

$$\langle es \rangle = -\sum_k p_k e_k \ln \frac{p_k}{g_k} \tag{7}$$

Here, p_j, e_k, and g_k are the occupation probability, energy eigenvalue, and degeneracy of the j^{th} eigenlevel, respectively.

Now, for a system interacting with a thermal reservoir, the equation of motion is written for an isolated composite of the system and reservoir with the system represented by subsystem A and the reservoir by subsystem B. In this case, there are three generators of the motion, namely, the $\hat{H}$ of the composite system and the identity operator, $\hat{I}^A$, of subsystem A and that, $\hat{I}^B$, of subsystem B. Thus,

$$\frac{dp_k^A}{dt} = \frac{1}{\tau}\frac{\begin{vmatrix} s_k^A p_k^A & p_k^A & 0 & e_k^A p_k^A \\ \langle s \rangle^A & 1 & 0 & \langle e \rangle^A \\ \langle s \rangle^B & 0 & 1 & \langle e \rangle^B \\ \langle es \rangle & \langle e \rangle^A & \langle e \rangle^B & \langle e^2 \rangle \end{vmatrix}}{\begin{vmatrix} 1 & 0 & \langle e \rangle^A \\ 0 & 1 & \langle e \rangle^B \\ \langle e \rangle^A & \langle e \rangle^B & \langle e^2 \rangle \end{vmatrix}} \tag{8}$$

Expanding the determinants, the equation of motion reduces to

$$\frac{dp_k^A}{dt} = \frac{1}{\tau} p_k [(s_k^A - \langle s \rangle^A) - (e_k^A - \langle e \rangle^A)\frac{C_3}{C_1}] \tag{9}$$

where

$$\langle e \rangle^A = \sum_j e_k^A p_k^A \tag{10}$$

$$\langle s \rangle^A = \sum_k s_k^A p_k^A = -\sum_k p_k^A \ln \frac{p_k^A}{g_k^A} \tag{11}$$

and the cofactors C_1 and C_3

$$C_1 = \begin{vmatrix} 1 & 0 & \langle e \rangle^A \\ 0 & 1 & \langle e \rangle^B \\ \langle e \rangle^A & \langle e \rangle^B & \langle e^2 \rangle \end{vmatrix} \tag{12}$$

$$C_3 = \begin{vmatrix} \langle s \rangle^A & 1 & 0 \\ \langle s \rangle^B & 0 & 1 \\ \langle es \rangle & \langle e \rangle^A & \langle e \rangle^B \end{vmatrix} \tag{13}$$

Now, using the hypoequilibrium concept developed by Li and von Spakovsky [22], each non-equilibrium state of the composite system is represented by a 2^{nd}-order hypoequilibrium state by factoring the overall state space of the composite system into two subspaces A and B. As a result, the ratio C_3/C_1 is the non-equilibrium intensive property, β, that accounts for the non-equilibrium energy and entropy fluctuations of subsystem A. At stable equilibrium when subsystem A is in mutual stable equilibrium with subsystem B (the reservoir R), β is inversely proportional to the temperature and $\beta = \beta^R$. Eq. (9) is is then rewritten as

$$\frac{dp_k^A}{dt} = \frac{1}{\tau} p_k [(s_k^A - \langle s \rangle^A) - (e_k^A - \langle e \rangle^A)\beta^R] \tag{14}$$

where $\beta^R = \frac{1}{k_B T^R}$, k_B is Boltzmann's constant, and T^R is the temperature of the reservoir [22].

In order to solve the equation of motion, an initial state represented by a non-equilibrium probability distribution is needed. This is found from the following perturbation function, which requires the probabilities p_k^{pe} and p_k^{se} from a partially canonical (pe) and a canonical (se) distribution function:

$$p_k{}^{init} = \lambda p_k{}^{pe}(e_k, \delta_k) + (1-\lambda) p_k{}^{se}(e_k) \tag{15}$$

Here, λ is a number between 0 and 1, and the closer to 1 the further it is from stable equilibrium. The probabilities corresponding to stable equilibrium can be calculated from the canonical distribution given by

$$p_k{}^{se} = \frac{g_k exp(-\beta^{se} e_k)}{\sum_k g_k exp(-\beta^{se} e_k)} \tag{16}$$

where β^{se} is proportional to the reciprocal of the stable equilibrium temperature T^{se}. The partially canonical probabilities, on the other hand, are found from

$$p_k{}^{pe} = \frac{\delta_k g_k exp(-\beta^{pe} e_k)}{\sum_k \delta_k g_k exp(-\beta^{pe} e_k)} \tag{17}$$

where β^{pe} is an unknown intensive property of the partially canonical state, and $\boldsymbol{\delta} = \{\delta_j\}$ is a vector of 0 and 1 values corresponding to whether or not a given energy eigenlevel of the partially canonical state is unoccupied or occupied, respectively. The unknowns for this last equation are all the p_k^{pe} as well as β^{pe} and, thus, an additional equation is needed in order to resolve this system of equations. This is given by energy of the partially canonical state expressed as

$$\langle e \rangle^{pe} = \sum_k p_k{}^{pe} e_k \tag{18}$$

where consistent with the choice of β^{se} made for the stable equilibrium state, $\langle e \rangle^{pe}$ must equal the energy, $\langle e \rangle^{se}$, given by

$$\langle e \rangle^{se} = \sum_k p_k{}^{se} e_k \tag{19}$$

For the radiation process, the equation of motion must be written for the SiC system interacting between a high-temperature and a low-temperature reservoir since the desired final state for the SiC system is not stable equilibrium but steady state instead. In this case, the equation of motion is written for an isolated composite system consisting of three subsystems A, B, and C with the latter two representing the two reservoirs. The generators of the motion now are $\hat{H}$ for the composite system; the identity operators, $\hat{I}^A$, $\hat{I}^B$, and $\hat{I}^C$, for the the three subsystems; and the particle number operator, $\hat{n}^A$, for subsystem A. The latter is added to account for the changing microstructure resulting from the defect clustering due to radiation damage. The equation of motion is then written as

$$\frac{dp_k^A}{dt} = \frac{1}{\tau} \frac{\begin{vmatrix} -p_k^A \ln \frac{p_k^A}{g_k^A} & p_k^A & 0 & 0 & p_k^A (n_k^A) & p_k^A e_k^A \\ \langle s \rangle^A & 1 & 0 & 0 & \langle n \rangle^A & \langle e \rangle^A \\ \langle s \rangle^B & 0 & P^B & 0 & 0 & \langle e \rangle^B \\ \langle s \rangle^C & 0 & 0 & P^C & 0 & \langle e \rangle^C \\ \langle ns \rangle^A & \langle n \rangle^A & 0 & 0 & \langle n^2 \rangle^A & \langle en \rangle^A \\ \langle es \rangle & \langle e \rangle^A & \langle e \rangle^B & \langle e \rangle^C & \langle en \rangle^A & \langle e^2 \rangle \end{vmatrix}}{\begin{vmatrix} 1 & 0 & 0 & \langle n \rangle^A & \langle e \rangle^A \\ 0 & P^B & 0 & 0 & \langle e \rangle^B \\ 0 & 0 & P^C & 0 & \langle e \rangle^C \\ \langle n \rangle^A & 0 & 0 & \langle n^2 \rangle^A & \langle en \rangle^A \\ \langle e \rangle^A & \langle e \rangle^B & \langle e \rangle^C & \langle en \rangle^A & \langle e^2 \rangle \end{vmatrix}} \tag{20}$$

where $n_k^A = n_k^{A,cl} + n_k^{A,ncl}$. This can then be reduced and re-cast in terms of the grand potential such that

$$\frac{dp_k^A}{dt} = \frac{\beta}{\tau} \left(p_k^A \langle \Phi \rangle^A - p_k^A \Phi_k^A \right) \tag{21}$$

where the grand potential is

$$\Phi_k^a = e_i^a - \beta^{-1} s_i^a - \beta^{-1} \gamma \, n_i^a \tag{22}$$

The final form shown equation 21 can simply be run on Matlab and obtain the necessary data for steady-state radiation damage.

III. RESULTS AND DISCUSSION

The energy eigenstructure obtained from REWL has approximately 234 energy eigenlevels, representing all possible configurations from the ⟨100⟩ dumbbells throughout the simulation box. The number of dumbbells range from 1 to 5 and provide details of scattering at lower energy levels and clustering near higher energy eigenlevels. These clustered defects can be a sink for other defects or dumbbells to cause migration towards the cluser during irradiation and after prolonged radiation damage, limiting their duration as a semiconductor. In Fig. 1 the y-axis labeled as $ln(g(e_k))$ is the logarithm of the degeneracy of each energy eigenlevel in the x-axis labeled as e_k.

Currently in this preliminary work, the energy eigenstructure only consists of the ⟨100⟩ dumbbells and does not include the other defects, so the higher energy range is not fulfilled just yet, hence not a complete parabolic curve. The lower energy levels have more scattering due to incomplete convergence during the REWL algorithm, where more testing is needed. Also, microstructural descriptors are necessary to detail the evolution during irradiation and can be taken as an arithmetic average for each energy eigenlevel e_k, where they are included in the Eq. 21 as $n_k^{A,cl}$ and $n_k^{A,ncl}$, where the data is obtained from the REWL simulation.

IV. CONCLUSION AND FUTURE WORK

In this work, the REWL algorithm gave the energy eigenstructure of many possible configurations representing radiation damage from solar flares. The microstructural evolution can then be determined by the SEAQT equation of motion to understand the effect of solar radiation on WBG semiconductors. Future work entails scaling the simulation size higher to account for realistic defect concentrations by using a neural network machine learning model. Also, applying the derived SEAQT equation of motion to determine steady non-equilibrium for the larger simulation size of 3C-SiC. Although the ⟨100⟩ dumbbells are the most favorable defect, including the anti-site and single interstitials for future modeling purposes will be assessed as well.

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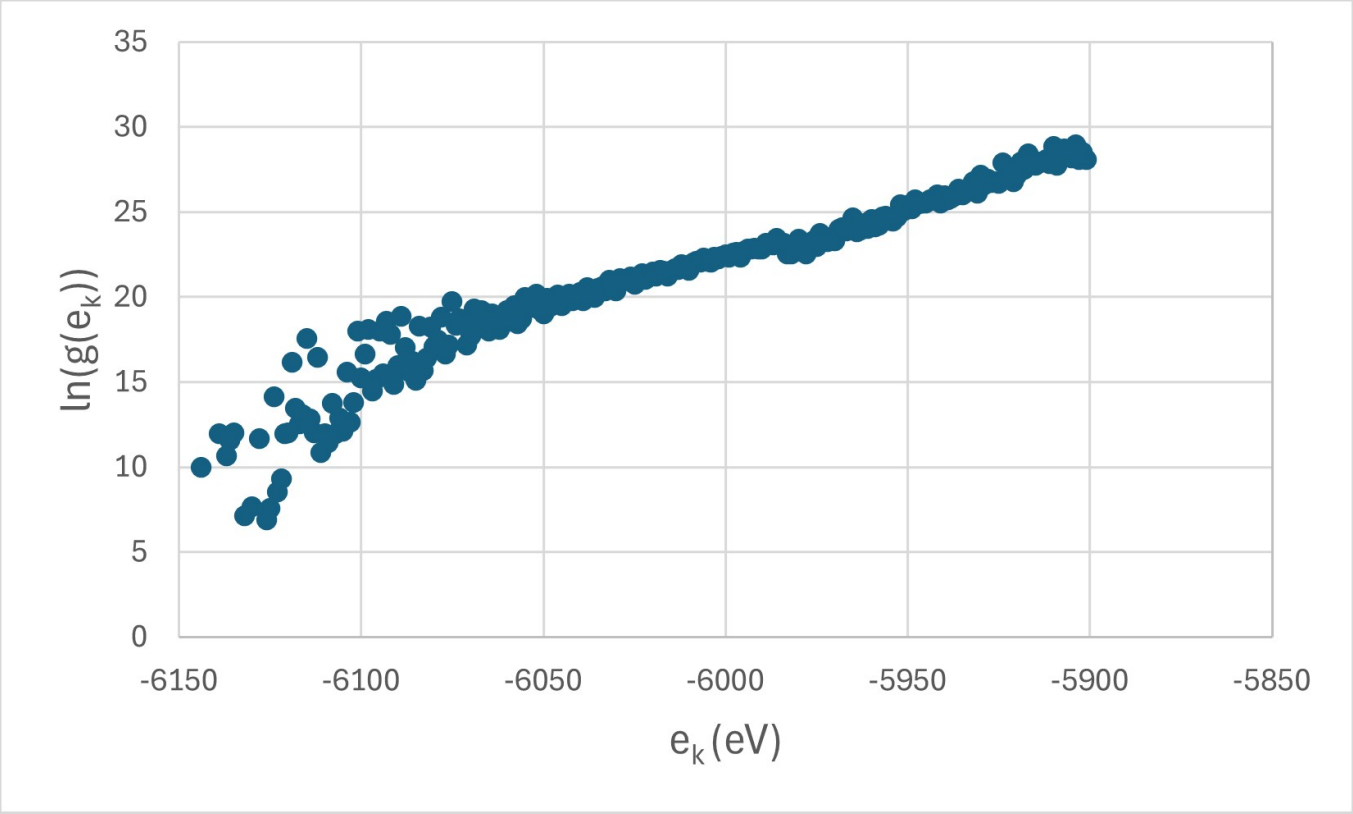

FIG. 1. Density of States of SiC

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