COMPUTING DIFFUSION COEFFICIENTS OF INTRINSIC POINT DEFECTS IN CRYSTALLINE SILICON

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1 Introduction

The quality of crystalline silicon highly influences the quality of semiconductor devices fabricated with it. Grown-in defects, such as octahedral voids or networks of large dislocation loops can be detrimental to the functionality of devices. Both type of defects result from the interaction of intrinsic point defects, vacancies and self-interstitials during growth and subsequent annealing of the crystal. In order to qualitatively describe the formation of microdefects in crystalline silicon, a detailed understanding of intrinsic point defects is necessary.

Modeling of defect dynamics in silicon crystals during growth requires the description of physical phenomena on different length and time scales. Continuum balance equations are used to describe the distribution, transport and kinetic interactions of point defects, either vacancies or self-interstitials, throughout the crystal as a function of the local temperature.[9] These equations contain highly temperature-dependent material properties, which describe atomistic events, such as the diffusion of a self-interstitial through the silicon lattice or the recombination of a vacancy with a self-interstitial. The absence of direct experimental measurements of intrinsic point defect properties at high temperatures make it necessary to compute these properties from atomistic simulations based on empirical or semiempirical atomistic models. Here, the microscopic quality of the crystal is given by the intrinsic point defect concentration. In order to understand the dynamics of these defects large scale molecular dynamics simulations based on the potentials of Stillinger-Weber and Tersoff are performed and the temperature dependence of the diffusion coefficients for self-interstitials and vacancies is computed.

2 Computational Method

Simple pair potentials like the Lennard-Jones interaction are not sufficient for the simulation of covalent systems. Here, Stillinger and Weber[4] and Tersoff[6–8] proposed new empirical potentials consisting of two- and three-body terms. Both potentials stabilize the diamond structure at low pressure. Stillinger and Weber fitted the incorporated parameters to give good agreement with experimental data for the melting point and the liquid structure. The parameters of the Tersoff potential are fitted to correctly reproduce cohesive energies and the elastic properties of silicon. We use both potentials for molecular dynamics in the (NVT)-ensemble to compute the diffusion coefficients of self-interstitials and vacancies.

Macroscopic values in the canonical ensemble can be measured by averaging the microscopic values with the Boltzmann factor. In this sense the diffusion coefficient is directly related to the spatial motion of the particles. Einstein first derived that the diffusion coefficient $D_{I,V}$ can be written as

$$D_{I,V} = \frac{1}{6}\frac{\partial}{\partial t}\langle(\vec{x}_{I,V}(t_0) - \vec{x}_{I,V}(t))^2\rangle, \qquad (1)$$

where $\vec{x}_{I,V}(t)$ is the position of a particle (interstitial (I) or vacancy (V)) at time t and $\langle.\rangle$ denotes averaging with the probability function. Since we cannot compute the average without knowing the probability function, we use the alternative formulation

$$D_{I,V} = \lim_{t\to\infty}\frac{1}{6(t-t_0)}\left(\vec{x}_{I,V}(t_0) - \vec{x}_{I,V}(t)\right)^2 \qquad (2)$$

and approximate this by

$$D_{I,V} \approx \frac{1}{6(t-t_0)}\left(\vec{x}_{I,V}(t_0) - \vec{x}_{I,V}(t)\right)^2. \qquad (3)$$

For t large enough this gives a good approximation to the diffusion coefficient.

The procedure is now as follows: Starting from an initial configuration of a perfect crystal with one self-interstitial or vacancy, we perform a molecular dynamics run of several nanoseconds at a given temperature in a periodic box. The coupling to a heat bath is simulated by the Nosé thermostat.[2] The position of the single point defect is tracked over time. Its identification is achieved by checking the local environment of each particle for variations from the perfect tetrahedral structure. The resulting trajectory of the point defect is afterwards corrected with respect to the periodic boundary conditions and smoothed by a low-pass filter to remove local fluctuations, see Fig.1 (left). From this data the function $r_{I,V}(t) = (\vec{x}_{I,V}(t_0) - \vec{x}_{I,V}(t))^2$ is computed, Fig.1 (right). Linear regression results in an approximation of the diffusion coefficient at this temperature according to equation (3). As one can see in Fig.1 (right), large fluctuations of $r(t)$ imply the necessity for molecular dynamic runs with long time intervals in order to minimize the error. The choice of the simulation parameters is therefore a compromise between large system sizes to reduce finite size effects and an acceptable computation time for the necessary physical simulation time.

3 Numerical experiments

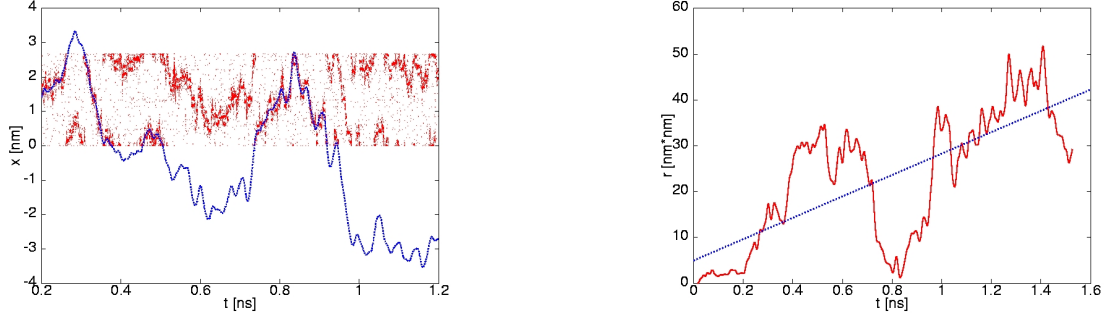We decided to use a system size of 1000 particles in a periodic box of length 2.67nm, which is larger than that

Fig. 1: Left: The x-coordinate of the trajectory (points) of a self-interstitial and the x-coordinate of the periodically corrected trajectory (dashed line). Right: Function $r_I(t)$ (solid line) and resulting linear fit (dashed line).
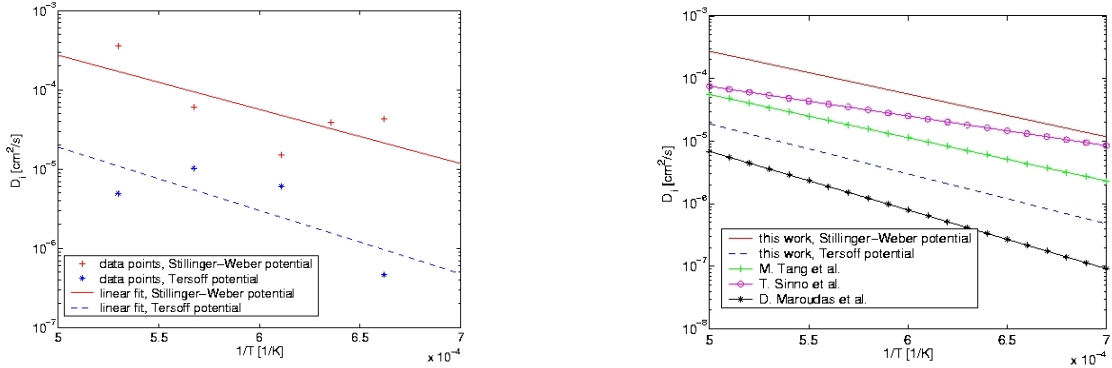

Fig. 2: Left: Diffusion coefficients for interstitials computed with Stillinger-Weber and Tersoff potential and linear fits. Right: Functions $D_I(T)$, comparison of our results with the results of M. Tang[5], T. Sinno[3] and D. Maroudas[1].

used in most of the literature.[1,3,5] This leads to a density of 2.44g/cm^3 conforming to the specifications given in.[4]
The resulting diffusion coefficients for a self-interstitial at several temperatures computed by the method described above with either the Stillinger-Weber or the Tersoff potential are shown in Fig.2 (left). Since the temperature dependence of the diffusion coefficient is expected to show an Arrhenius behavior

$$D_{I,V} = D^0_{I,V} e^{-E^m_{I,V}/k_B T} \qquad (4)$$

with migration energy $E^m_{I,V}$ and diffusivity prefactor $D^0_{I,V}$, we fit the logarithm of these data points to a straight line, see Fig.2 (left). The results for a self-interstitial are

Stillinger-Weber: $\quad E^m_I = 1.35\text{eV} \quad D^0_I = 0.72\text{cm}^2/\text{s}$
Tersoff: $\quad E^m_I = 1.58\text{eV} \quad D^0_I = 0.19\text{cm}^2/\text{s}$.

As shown in Fig.2 (right), the values are in good agreement with the ones found in the literature.[1,3,5] Such data can then be used as input parameters for the macroscopic equations.

4 Concluding remarks

The temperature dependence of the diffusion coefficient for a point defect in crystalline silicon computed by means of molecular dynamics with two empirical potentials is in agreement with the results found in the literature. Due to the small number of data points so far, more simulations have to be performed to stabilize the results, whereas the

uncertainty of each data point itself can only be minimized by increasing the simulation time. The resulting more accurate data can be used to transfer the point defect dynamics on the atomistic level to the macroscopic process simulation of silicon crystal growth. This can result in an optimization of this process and the ability to produce crystals of improved quality.

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