Simplified Model for Monte Carlo Simulation of Point Defect Dynamics during Industrial Silicon Crystal Growth

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Abstract

A new Monte Carlo 2D model for vacancies and interstitials in a cubic crystal is proposed. These defects are modelled using the concept of the shifted lattice for interstitials in the base lattice and of covalent bonds. It is shown that at higher temperatures this model leads to the liquid structure of the material. Using Monte Carlo simulations the corresponding phase transition is investigated. The dependence of the phase transition temperature and latent heat from bond energies is shown. The generalisation of the 2D model for the 3D case (in mean field approximation) is proposed. The model also is used to investigate the kinetics of the agglomeration of vacancies (voids) at different crystal cooling rates.

Introduction

In order to model the formation, diffusion and annihilation of point defects during industrial crystal growth, it is necessary to use the microscopic theory of crystallization process. The difficulties to describe some important aspects of the crystallisation process with analytical models (e.g. nucleation, clustering, point defect development in grown crystal) and the rapidly growing computer resources determined that in the last decades the Monte Carlo simulations were chosen as the most powerful instrument for modelling of crystal growth processes. Between many works devoted in the last years to the Monte Carlo simulations we would like to mention first the article from Beatty and Jackson [1]. There the relationship between the silicon crystal growth rate and under cooling has been modelled for the (100) and (111) faces, and the results of the simulations compared with experimental observations. With the Monte Carlo simulation [2], Jackson investigates the normalized growth rates as a function of chemical potential difference for surfaces of various roughness. In [3] Jackson et al. report Ising model simulations with Monte Carlo method for the growth of alloys. E. van Veenendaal et al. in [4] study the kinetic roughening for the (100) surface of the simple cubic Kossel crystal taking into account the nearest-neighbour bonds only. In a similar way in [5] the interaction between dislocation growth, 2D-nucleation and misorientation step flow is investigated for a wide range of driving forces by means of Monte Carlo simulations for growth of the Kossel (100) surface. In [6] Veenendaal et al. analyse the asymmetry between growth and dissolution using Monte Carlo simulations of flat and vicinal {100} surfaces of Kossel crystal and the kinetic smoothing.

In this paper a 2D model of vacancies and interstitials in the cubic crystal is proposed. These defects are modelled using the concept of the shifted lattice for interstitials in the base lattice and of covalent bonds. It is shown that at higher temperatures this model leads to the liquid structure of the material. Using Monte Carlo simulations the corresponding phase transition is investigated. The dependence of the phase transition temperature and latent heat from bond energies is shown. The model also is used to investigate the kinetics of the
agglomeration of vacancies (voids) and agglomeration of interstitials. The generalization of the 2D model for the 3D case (quasi in mean field approximation) is proposed. The present stage of the work is devoted for studying of principal questions and we consider initially the cubic lattice. Nevertheless, the model and used methods can be further applied for real Si crystal with diamond lattice structure.

1. 2D Monte Carlo model of vacancies and interstitials in the cubic crystal

The experimental data show that there is a tendency for vacancies and interstitial defects to form clusters. It means that association of defects is energetically profitable. The proposed 2D model for cubic crystal assumes that the interstitial defects can form covalent bonds. In order to generalise the model, we assume that another sub-lattice can be formed in interstitial sites of cubic lattice (see. Fig. 1).

The atoms A form a main lattice, while atoms B can be considered as interstitial atoms. The energy \( e_{aa} < 0 \) denotes the energy of covalent bond, while \( e_{ab} \) is interaction (repulsion) energy between atoms belonging to the main and the sub lattice. As the energy of interstitial is positive, the energy \( e_{ab} > 0 \) is positive also. Nevertheless, the total energy of crystal lattice must be negative.

![Fig. 1. Principal scheme of the model for 2D case](image1)

![Fig. 2. Maximal change of energy for 2D model, \( \Delta E_{max} = E_1 - E_2 = 3e_{ab} - 4e_{aa} \)](image2)

![Fig. 3. Interaction between vacancies. The association of vacancies decreases the energy by: \( \Delta E = E_1 - E_2 = -e_{aa} > 0 \)](image3)

![Fig. 4. Interaction between interstitials. The energy decreases during association of interstitial atoms by: \( \Delta E = E_1 - E_2 = -e_{aa} > 0 \)](image4)
The 2D Monte-Carlo calculations have been performed to qualitatively and quantitatively appreciate the possible results. The transition probability from one state with energy $E_1$ to the state with energy $E_2$ satisfies the formulas (see also Fig. 2, 3, 4):

$$W_{1\rightarrow 2} = W_0 W_{\max} W_{1\rightarrow 2},$$

$$w_{1\rightarrow 2} = \frac{1}{W_{\max}} \exp\left(\frac{E_1 - E_2}{2k_B T}\right),$$

$$W_{\max} = \exp\left(-\frac{\Delta E_{\max}}{2k_B T}\right) = \exp\left(\frac{3\epsilon_{ab} - 4\epsilon_{aa} - 2\epsilon_{bb}}{2k_B T}\right),$$

where the multiplier $W_0$ determines the rate of the process and it can be included generally in time factor. The final stationary result in thermal equilibrium is independent from $W_0$. During the modelling of transient crystallisation process with real rate, the value of the multiplier is significant, because the relaxation time to thermodynamic equilibrium can be very large.

The quadratic lattice $N\times N$ with periodic boundary conditions has been chosen for modelling of 2D case. The random atom is chosen according to Metropolis algorithm. The chosen atom can move in any of possible directions according to stochastic description. The transition probability determines whether the transition occurs or not.

2. The generalization of the 2D model for the 3D case (mean field approximation)

![Fig. 5. Principal scheme for the 3D analysis](image)

![Fig. 6. The scheme of the actual layer for quasi 3D calculations](image)

In this section the most important mathematical relations for 3D case in mean field approximation are summarised. First, the following occupancy number are introduced: $n_a$ und $n_b$,

$$n_{a,b} = \frac{N_{a,b}}{N} = 0 \div 1$$

where $N_{a,b}$ is the total number of atoms in sub-lattices $a$ or $b$, $N$ – the total number of possible positions in one sub-lattice. Energies of interaction between actual layer and neighboured layer are: $\epsilon_a = 2\epsilon_{aa}n_a + 4\epsilon_{ab}n_b$ - for the $a$ sub-lattice; $\epsilon_b = 2\epsilon_{ba}n_a + 4\epsilon_{bb}n_b$ - for the $b$ sub-lattice. Change energy per one particle for transition from sub-lattice $a$ to $b$ is: $\epsilon_a - \epsilon_b = 2(n_a - n_b)(\epsilon_{aa} - 2\epsilon_{ab})$. E.g., for the maximal transition possibility follows than
\[ W_{\text{max}} = \exp \left( -\frac{\Delta F_{\text{max}}}{2k_B T} \right) = \exp \left( \frac{3\varepsilon_{ab} - 4\varepsilon_{aa} + 2(2\varepsilon_{ab} - \varepsilon_{aa})(n_a - n_b)}{2k_B T} \right) \]

Additional formulas used in Monte-Carlo calculations are (e.g., for averaged values after \( n \) iterations):

\[ A_n = \langle a \rangle = \frac{a_1 + a_2 + \ldots + a_n}{n}, \quad A_{n+1} = \frac{a_1 + a_2 + \ldots + a_{n+1}}{n + 1} = \frac{a_{n+1}}{n + 1} + \frac{n}{n + 1} A_{n+1}. \]

3. Results of Monte Carlo simulations for steady states and for phase transition points

This section presents some calculated steady state results with presented 2D model for investigation of phase transition point and for corresponding crystal lattice structures.

Fig. 7 shows an averaged potential energy \( E \) per atom in crystal and concentration \( n \) of atoms in basic lattice via temperature for two interactions energies cases. As it can be seen the chosen interaction energies are measured in K and have relatively low values, therefore the corresponding phase transition temperatures \( T_0 \) (temperatures at \( n=0.5 \)) are low also: for \( e_{aa} = -300K \) and \( e_{ab} =25K \) follows that \( T_0 =216K \); for \( e_{aa} = -300K \) and \( e_{ab} =50K \) - \( T_0=236K \). It is easy possible to scale these results to higher energies and temperatures because of \( T_0 \) depends linear on interaction energies.

From Fig. 7 follows that the system has a sharply defined phase transition point \( T_0 \) at \( n=0.5 \). If \( T>T_0 \) then both lattices are equally occupied (model of fluid), below the \( T_0 \) one lattice is dominating (solid structure). Fig. 8. illustrates the calculated crystal structure (steady state case) for various temperatures in case of \( e_{aa} =-300K \) and \( e_{ab} =50K \).

4. Influence of cooling dynamics on voids growth

The developed model is used to investigate the influence of cooling dynamics on voids growth. Fig. 9 presents an example of results for the following crystal energetic parameters: \( e_{aa} =-300K \) and \( e_{ab} =25K \). The cooling process is started at 210 K (i.e. beneath the
crystallisation temperature) and ended at 50 K. The start situation has only the point defects: vacancies and interstitials, but the vacancies are dominating so, that the total silicon atom concentration in basic lattice has the value 94%. The cooling rate is characterised with the number of millions of Monte Carlo iterations done for one temperature reduction step of 0.1 K. The lattices with 100*100, 150*150, 200*200 and 400*400 atoms were investigated.

The calculations have shown that there a temperature interval between 162 K and 98 K exists in which the intensive growth of voids can be observed. The Fig. 9 shows the state in system 400*400 at T=85.8K in case of low cooling rate (320 millions of iterations for one temperature step 0.1 K). On the left side the results of counted distribution of voids dimensions are shown. For the counting different methods were used, the most sensitive is the method “1D+” that shows that voids with dimension about 4 atomic distances have a maximum in the dimension distribution. The distribution shows also maxima with 8, 10 and 15 atomic distances.

Fig. 8. Calculated crystal structure (steady state case) for various temperatures in case of $e_{aa} =-300$K and $e_{ab} =50$K.
Fig. 9. The state in system 400*400 at T=85.8K in case of low cooling rate (320 millions of iterations for one temperature step 0.1 K).

Conclusions

With the proposed Monte Carlo 2D model for vacancies and interstitials in cubic crystals is shown that at higher temperatures this model leads to the liquid structure of the material. With this model the kinetics of the agglomeration of vacancies (voids growth) at different crystal cooling rates is investigated.

References


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