Transactions of the Materials Research Society of Japan. Vol. 20

A NEW LATTICE MODEL FOR Si CRYSTAL GROWTH WITH ENERGY FUNCTION OF COORDINATION NUMBERS AND BOND ANGLES

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Abstract

A new model is proposed for Si crystallization from molten state, based on Monte Carlo (MC) simulation. In this model, atoms move only on BCC lattice sites, and the energy of atom depends on the numbers of the 1st and 2nd nearest neighbors and the bond angles. With using a parameter set that includes the bond angles or 2nd nearest neighbors in addition to the 1st nearest neigbhors, crystallization was observed with this model. The present model is qualitatively successful to represent condensed phases.

1. INTRODUCTION

It is necessary to clarify theoretically the process of Si single crystal growth from the melt in detail by numerical simulation because of highly advanced engineering requirement for Si devices from industries. A number of experiments and numerical simulations have been studied on crystallization, but there are only limited knowledge on them. However, there may be a relationship between the bulk defects and the unstable solidification process at the solid–liquid interface, it is difficult to observe dynamics of interfaces by experiments, and it is necessary to study them with simulations.

Several studied on melt and crystalline Si by Molecular Dynamics (MD) simulations with empirical interatomic potentials[1]–[5] and Car–Parrinello method[6] have already been reported. These studies are limited in time and it is necessary to simulate the phenomena for a large time scale by Monte Carlo (MC) in condensed phase.

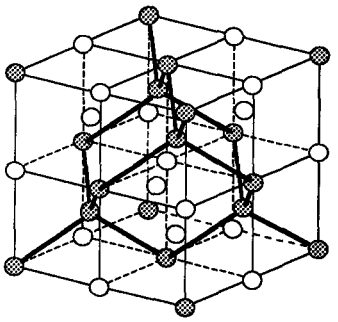
In this paper, a new model based on MC simulation on a BCC lattice which explicitly takes many–body interactions into account is proposed to describe dynamically this important process.

2. BCC LATTICE MODEL

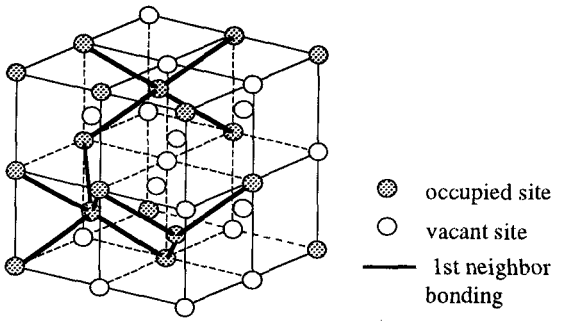
The present model is based on the idea that each atom moves only on the BCC lattice sites : 2×2×2 BCC cells are shown in Fig.1. Figure1(a) is the case where the atoms occupy these sites in order, and solid state having a diamond structure is realized. In contrast, Fig.1(b) shows a sample of liquid state, where the atoms occupy BCC sites randomly. In both cases the lattice occupation is set to be 50 %.

All the tetrahedrons consisting of 5 Si atoms centered at the body–center positions may be shifted along the X–axis (or Y–axis) by one BCC cell, or may be rotated 90° at each center around one of the Cartesian coordinates. Therefore, there are 4 equally stable diamond structures of Si crystal on the model lattice (Figs. 2 (a)–(d)).

Here MC simulations are carried out using the Metropolis method[7] with the 3–dimensional periodic boundary conditions. We start from random initial site occupations for which the temperature is fixed to be slightly lower than the melting point. The same model is also applied to examine the melting of a single crystal with slow



(a) Solid State



(b) Liquid State

Fig.1. Schematic diagrams of the BCC lattice model.

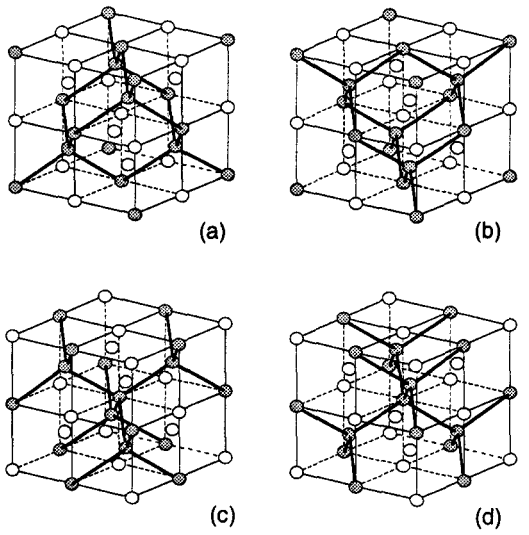


Fig.2.(a)–(d) Typical diagrams of the 4 equally stable diamond structures of Si crystal.

heating. The size of the system in this study varies from 8×8×8 diamond lattice sites (4,096 atoms) to 16×16×16 (32,768 atoms).

3. ENERGY PARAMETER SET

The energy of each atom is assumed to be determined by the numbers of the 1st nearest neighbors($0 \sim 8$) and the 2nd nearest neighbors($0 \sim 6$), and the bond angle between two nearest atoms ($70°, 109°$ and $180°$). The energy of the vacant lattice site is fixed to be 0.

3.1 Tetrahedron model

We first assumed that the energy of the atoms which construct the tetrahedral structure is lower than that of the other atoms and simply ignored the energy dependence on the 2nd nearest neighbors. In this case, 10,000 MC steps are necessary at least to get large single Si crystal which consumes atoms over 70 % of the system.

3.2 1st nearest neighbors only model

Next we simulated the crystallization process with the parameter set which takes account of the number of the 1st nearest neighbors. In this case, several structures have the most stable energy other than the diamond structures(Figs.3 (a)–(c) show those structures). It was impossible to obtain large crystalline Si by this model.

3.3 1st nearest neighbors and bond angles model

Lastly a parameter set that includes the bond angles in addition to the 1st nearest neighbors is applied.

One of the most satisfactory energy parameter sets obtained so far by the genetic algorithm[8] is shown in Fig.4(a) for the 1st nearest neighbors and Fig.4(b) for the bond angles.

This is the choice upon which all results reported in the remainder of this paper are based.

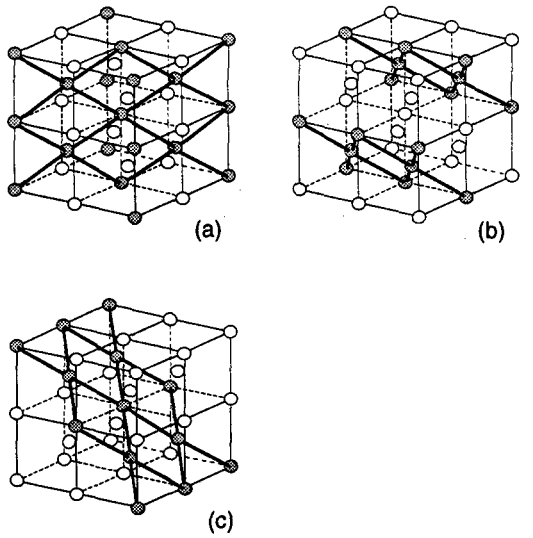
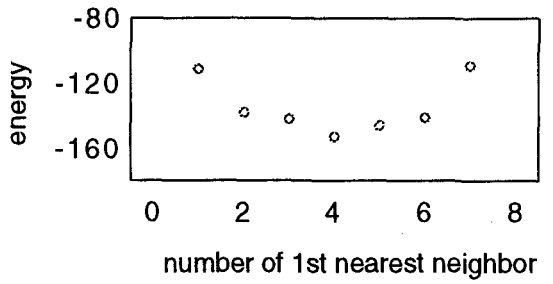
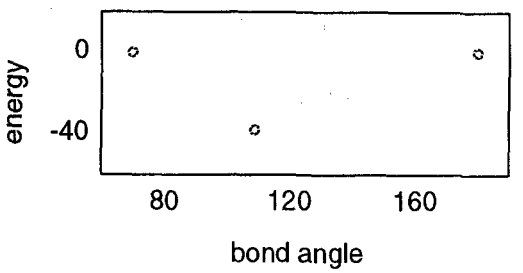


Fig.3.(a)–(c) Typical three diagrams of structures which have the most stable energy.



(a) Assumed relation between the number of the 1st nearest neighbors and energy.



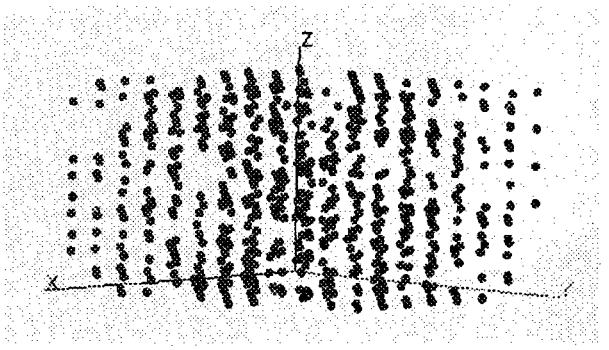
(b) Assumed relation between bond angle and energy.

Fig.4. Relation between structure and energy.

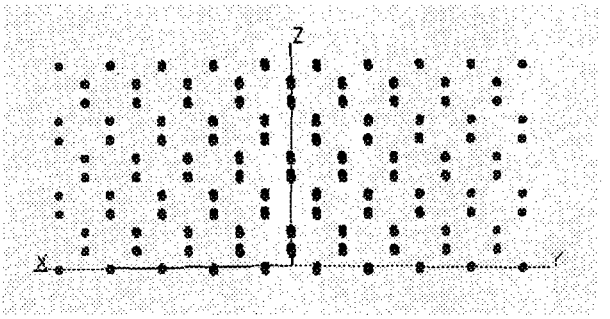
4. RESULTS AND DISCUSSION

4.1. Si crystal growth from melt

Figure5(a) shows an initial condition of simulation. The system is shown from the direction of $< 110 >$. This is a 50% filling random distribution of atoms on the BCC lattice. After 5,000 MC steps, we finally obtained a solid Si structure shown in Fig.5(b).



(a) Initial distribution of atoms on the BCC lattice.



(b) Distribution of atoms after 5,000 MC steps.

Fig.5 A sample of results of the present simulation.

Figure6 shows a sample of the results of the present simulation on the process of Si crystal growth. The number, $N = N_a + N_b + N_c + N_d$, represents the total number of atoms constructing the 4 different Si crystals. Since the temperature of the system is kept constant, there are many nuclei in the system. In the early stage of

simulation, many of them construct crystals with different orientations. Therefore, small crystals compete each other, yielding poly-crystals. However, for the large MC steps, crystal with one direction has precedently grown in this case.

In principle, the present model is able to describe the critical radius of the crystal, and the calculated parameter is expected to explain the effect of the interface energy.

4.2. Change of coordination number with temperature

Si is a 4 fold coordinated atom in the solid-state, while the liquid state includes 6 or more fold coordinated atoms for the stable coordination[9]. The coordination is defined by the number of the 1st and 2nd nearest neighbors. Figure7 shows a typical sample of time evolution of the coordination number with temperature. The simulation started from a single crystal with slow heating. It is confirmed that the 4 fold coordinated atoms mainly exist up to a temperature which is somewhat higher than the melting point. Higher-coordinated atoms appear as temperature rises.

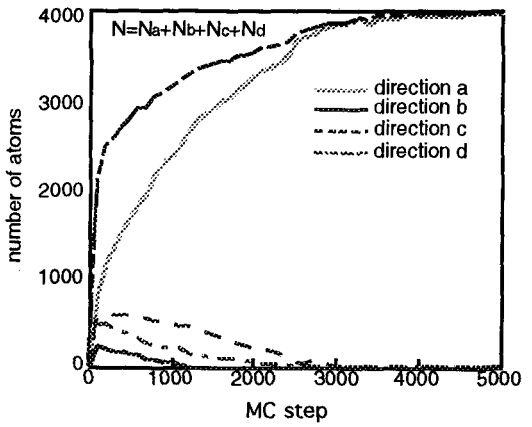


Fig.6. Number of atoms which construct tetrahedron as a function of MC step (N_a-N_d:Number of atoms grew in the 4 directions of Si).

This model seems to be qualitatively successful to represent condensed phases. To realize a large single crystal growth by simulation, the temperature gradient must be taken into account in the future.

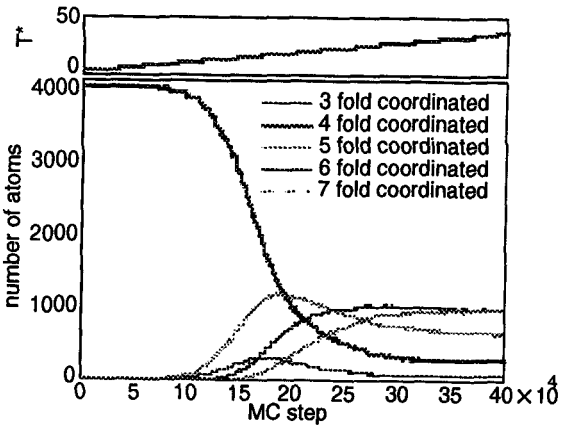


Fig.7. Time evolution of the coordination numbers with increasing temperature.

Acknowledgments

The authors are grateful to the Computer Science Group of the IMR, Tohoku University for their continuous support of the HITAC S-3800/380 supercomputing system.

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