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A molecular dynamics study of nucleation of dislocation in growth of silicon from melt

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ABSTRACT

Molecular dynamics (MD) simulations of growths of crystalline silicon from the melt along ⟨100⟩, ⟨110⟩ and ⟨112⟩ directions have been carried out. Tersoff potential is employed for computing atomic interaction. The results show that in the growths along ⟨100⟩, ⟨110⟩, no dislocation was formed, while in the growth along ⟨112⟩, more than one dislocations formed. The dislocation formed is identified as a partial dislocation lying on an {111} plane, with segments of 30° partial dislocation, 90° partial dislocations and some kinks. The dislocation nucleates stochastically at the crystal/melt interface, where atoms interact with each other to form at least two units of stable periodic non-six-member-ring groups. Once formed, it extends with the crystal growth, with its two ends attached to the crystal/melt interface. The ⟨112⟩ growth distinguished from the ⟨100⟩ and ⟨110⟩ growths by its prone-to-{111}-faceting and twinning, which may be the cause of the significantly higher probability of dislocation nucleation in the ⟨112⟩ growth.

1. Introduction

It is well known that dislocations are detrimental to photovoltaic performance of crystal silicon [1,2]. In ⟨100⟩ growth of monocrystalline silicon, formation of dislocations can be avoided, and the so called dislocation-free monocrystalline silicon can be now be commercially produced. However, in the current major low cost solar cell material, multicrystalline silicon, large amounts of dislocations commonly exist, and they are believed to be the major reason for their being inferior to monocrystalline in photovoltaic performance. Ryningen et al. [3] have found that clusters of dislocations can be formed in growth of multicrystalline silicon, with the size of the cluster, i.e., the number of dislocations in the cluster, increase with the growth. They have also found that the cluster originates from a primary dislocation. As to how the primary dislocation nucleates, or what the conditions for the nucleation are, no information is available yet.

Such information cannot be obtained through physical observations because the dislocation nucleation occurs in nanometer scale and in pico-second time-span, at high temperature. Molecular dynamics (MD) simulations can track the trajectory of each atom in a system, and is therefore able to observe the process of dislocation nucleation at atomistic scale. Lots of researchers have done MD simulations to investigate the mechanisms of dislocation nucleation in crystalline silicon. In order to drive the formation of dislocation, surface defects were pre-set on the simulated silicon crystals and external stresses were applied, and their structural responses were examined [4–6]. Nucleation of different types of dislocations, including full and partial dislocations, from the surface defects has been observed. It has been commonly found that these dislocations nucleate by gliding along {111} planes due to the exterior conditions, i.e., the stresses or the surface defects. Occasionally, dislocations were observed forming along {110} plane in simulated nano-sized silicon [7]. These studies have obtained important information about the mechanism and condition of nucleation of dislocation in solid silicon, and demonstrate the effectiveness of MD simulations for such studies. However, none of the reported MD studies of dislocations in silicon address their nucleation in growth of silicon crystal from melt, though formation of other defects such as point defects [8,9] and twin boundaries [10] in the growth have been addressed. In the present work, we carried out MD simulations of the crystal growth of silicon from its melt, along three basic crystallographic directions, ⟨100⟩, ⟨110⟩ and ⟨112⟩ respectively, examine statistics of dislocation

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formation in the growth along the different direction, and trace back the origin and nucleation of a typical observed dislocation.

2. Models and methods

Fig. 1 shows the initial set up of the molecular dynamics simulation cell of silicon crystal growth. The middle part of the cell with light gray colored atoms represents the crystal "seeds", the left and right parts with dark (red) colored disordered-atoms represent the silicon melt, which were obtained through a simulation of melting silicon crystal. Periodic boundary conditions are applied in each dimension of the cell. The growth direction is set as *X* direction of the coordinate system, as shown in Fig. 1. Three different simulation cells, with *X*, *Y* and *Z*-axis being [100], [010] and [001] of the crystal seed part for the ⟨100⟩ growth, being $[1\bar{1}0]$, $[11\bar{2}]$ and [111] of the crystal seed part for the ⟨110⟩ growth, and being $[11\bar{2}]$, [111] and $[1\bar{1}0]$ of the crystal seed part for the ⟨112⟩ growth, are set accordingly. Their initial melt parts are all set as 12 nm long each side along the *X* axis. The cell sizes were chosen on basis of trials with approximately 50% larger sizes. If the larger ones produced the similar results, the original cell sizes were then regarded as out of non-physical constrained range and were thus acceptable for the concerned study.

The inter-atomic forces are calculated with Tersoff potential [11], which has been shown to be appropriate for modeling silicon in transition from melt to crystal [12], and has been successfully used in simulation studies of defects formation in silicon growth from melt [8,9]. The system is relaxed for 10^4 time steps first to let the total energy of the system to drop to and relatively stabilized at a lower level, and is then let to start the simulation of crystal growth in a NPT ensemble. A constant pressure of 1 atm is kept by Anderson algorithm [13]. A constant temperature at an undercooling of 300 K is kept by Nose–Hoover algorithm [14]. The length of each time step is 10^{-3} ps.

The reason for choosing an unrealistically great undercool is to allow a fast enough crystal growth, so that the simulation runs can be completed in feasible lengths of time. Similar compromises are often adopted in MD simulations, assuming the great acceleration would not change the nature and mechanisms of the concerned process. For the present study, indeed it is assumed that mechanisms for formation of dislocations, and qualitative dependence of the formation probability on growth directions, would not be altered by accelerating the crystal growth from melt, though higher general probability of dislocation formation in a greatly faster growth is expected from practical experiences of crystal growth.

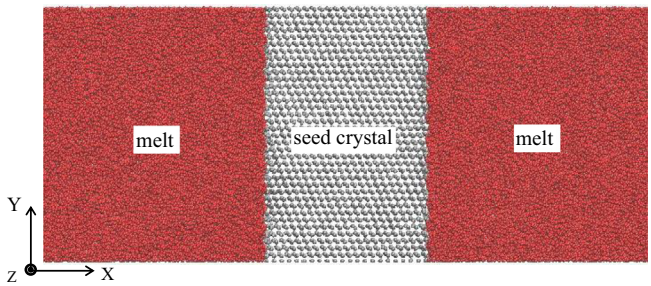

Fig. 1. The initial setup of the simulation cell for growth of silicon crystal from melt, along with the coordinate system. (For interpretation of the references to color in this figure, the reader is referred to the web version of this article.)

3. Results and discussion

Fig. 2 shows the snapshots of the systems in the simulated crystal growths at 1000 ps after starts of the simulation runs. Only the right halves of their XY projections are shown. As can be seen, no sign of defect is present in the crystals grown along ⟨100⟩ or ⟨110⟩, while in the crystal grown along ⟨112⟩, explicit disorders in the atomistic arrays appear, along with a feature of zigzag interface and twinning. The disordered region is located at the border between the normal crystal and the twin, which should be a core of a partial dislocation according to the known theory of dislocations. Observations from more simulation runs are summarized in Table 1. It confirms that, in the simulated growth along ⟨100⟩ or ⟨110⟩, the crystals do not form dislocations, while in the growth along ⟨112⟩, dislocation form at quite a high probability. In each of the three simulation runs of the ⟨112⟩ growth, a similar dislocation and the interface features as shown in Fig. 2c was found to appear before 400 ps of the simulated growth.

Orientations of the major parts of the zigzag interfaces appeared in the ⟨112⟩ growth are found to be uniquely fixed – they keep perpendicular to the *XY* plane $(1\bar{1}0)$, and incline at an angle of about 19.5° from $(11\bar{2})$ plane. A simple calculation confirms that they are $(11\bar{1})$ planes. We therefore suggest that the {111} facets growth occurs in ⟨112⟩ growth of crystalline silicon from melt. Such phenomenon has been observed in other MD simulations of ⟨112⟩ growth [10], and in an experiment, though the widths of the facets are much greater (~10 μm) [15]. In the experiment, ⟨001⟩ and ⟨110⟩ growths also appeared to evolve into {111}-faceted growths, after certain lengths of growth time. The authors suggested that the {111}-faceting is universal for growth of silicon from melt if only some perturbation to flatness of the crystal/melt interface takes place. The reason for such faceting is that the {111} surfaces have the minimum surface energy. In the present simulations of growths of very limited lengths of time, no profound faceting appeared in the ⟨100⟩ and ⟨110⟩ growths, after the same length of growth time. This at least suggests that ⟨100⟩ and ⟨110⟩ growths are relatively much less prone to {111}-faceting as compared to ⟨112⟩ growth.

Fig. 3 exposes a $(11\bar{1})$ facet plane across the disorder zones in the crystal formed in the simulated $[11\bar{2}]$ growth, by removing the atoms above the plane. The atoms are colored in their potential energies, with lighter and warmer colors representing higher potential energies. It is not surprising to see that the atoms in melt appear to be of higher potential energy. While interestingly, a line of half-loop of atoms with lighter and warmer color appeared on the crystalline part of the plane. The features of higher potential energy and the lining distribution suggest that this is a dislocation line, or more strictly, these atoms are of the core of a dislocation line. This dislocation line is lying on the exposed $(11\bar{1})$ plane, with both sides ending downward at the solid–liquid interface, forming a half-loop. Fig. 3b shows the projection of the top two atomic layers of the exposed $(11\bar{1})$ plane in Fig. 3a. The Si–Si bonds connecting the nearest neighbored atoms are displayed to help visual comprehension of the structure. Typical hexagon six-member rings of {111} planes are observed, except at the dislocation core region, where periodic series of non-six-member-ring groups appeared, e.g., the 558-558-558-…series along the horizontal segments of the dislocation, where the number 5 or 8 represents a five-member ring or an eight-member ring respectively. The line connecting them generally matches well the half-loop observed in Fig. 3a. Bennetto et al. [16] have proved by multiple approaches of energy minimization that the core structure of one type of partial dislocation is a periodic series of one five-member ring and a seven-member ring, 57-57-57-..., rather than a series of distorted six-member ring, 6-6-6-6-…. Supported by both the above observations and this proved core structure, it is postulated that