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Phase field modeling of grain structure evolution during directional solidification of multi-crystalline silicon sheet

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ABSTRACT

Evolution of grain structures and grain boundaries (GBs), especially the coincident site lattice GBs, during directional solidification of multi-crystalline silicon sheet are simulated by using a phase field model for the first time. Since the coincident site lattice GBs having lower mobility, tend to follow their own crystallographic directions despite thermal gradients, the anisotropic energy and mobility of GBs are considered in the model. Three basic interactions of GBs during solidification are examined and they are consistent with experiments. The twinning process for new grain formation is further added in the simulation by considering twin nucleation. The effect of initial distribution of GB types and grain orientations is also investigated for the twinning frequency and the evolution of grain size and GB types.

1. Introduction

Grain boundaries (GBs) of multi-crystalline silicon (mc-Si), grown by directional solidification, play a crucial role on the electrical properties of solar cells. Controlling GBs is often necessary for improving the ingot quality. For example, the dendritic casting method [1] is to initiate many highly symmetric coincident site lattice (CSL) GBs, e.g., Σ3 GBs, through grain growth at high undercooling generated along the crucible bottom [1], and they are useful for the high lifetime in the grown ingot. In ribbon growth, the twin grains have better lifetime and their formation have also been discussed by Stockmeier et al. [2] based on thermodynamics arguments. Nevertheless, the detailed GB interactions and how to control them are still not quite clear. Recently, the ingot growth has made a significant progress by initiating uniform and small grains at the crucible bottom, i.e., the so-called high-performance mc-Si (HPMC) [3]. The large amount of non-Σ GBs, generated at the beginning of solidification, would relax the thermal stress and thus reduce the generation of the dislocations; they also terminate the propagation of the dislocations during ingot growth [3–5]. However, during ingot growth, more Σ3 GBs appear while the proportion of non-Σ GBs decreases, and the ingot quality near the top is thus deteriorated. Therefore, understanding the evolution of grain structures is very important to further control the directional solidification of Si ingots or ribbons.

GB evolution during directional solidification of silicon has been paid much attention in recent years. Fujiwara et al. [6] found that the low-energy grain, such as ⟨1 1 1⟩, tends to overgrow the others at low undercooling. Chen et al. [7] were the first to simulate this phenomenon by using a two-dimensional phase field model considering anisotropic interfacial energy and kinetic coefficient with some success. Duffar and Nadri [8] further described the twinning mechanism for the nucleation on the {1 1 1} facet plane near GBs. However, the critical undercooling proposed to form a twinned nucleus in their mechanism was 9 K or larger. Their model was recently modified by Lin and Lan [9], who considered the interactions between the nucleus and the neighbor grain at the groove. A more reasonable undercooling for the twin nucleation on the {1 1 1} facet plane was derived, i.e., less than 1 K. This value was more consistent with the experimental observation [10].

To confirm the twin formation, in-situ X-ray synchrotron imaging was used to investigate silicon crystal growth by Tandjaoui et al. [10]. They characterized the birth of new grains and showed that twins nucleated exactly on {1 1 1} facets at the GB grooves [11]. Moreover, Wong et al. [12] investigated the evolution of grain orientations and GBs by analyzing wafers at different heights from directional solidification ingots seeded with small and randomly oriented silicon beads. Indeed, twin nucleation was found at the tri-junctions. In addition to these research, there have been several works related to the interactions between $\Sigma 3^n$ GBs [13–16]. Recently, the interactions between $\Sigma 3^n$ and non-Σ GBs were further investigated by Lin et al. [17] for the directional solidification of mc-Si sheets at different speeds. At lower speeds, they found

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that Σ3 GBs decreased due to the interactions between Σ3 GBs and non-Σ GBs, while twinning was a key mechanism for the increase of Σ3 GBs at higher speeds. Their study using mc-Si sheets unfolded the detailed mechanisms of GB interactions in silicon. However, up till now these have not yet been simulated successfully.

Moreover, the anisotropic properties of GBs in silicon play a crucial role in the GB interactions, and thus in their evolution. The classic Read and Shockley dislocation model [18] describes the energy in low-angle non-Σ GBs. For high-angle GBs, the well-known Brandon criteria [19] are used to calculate the maximum deviation angle from CSL GBs, which have relatively lower energy comparing with other non-Σ GBs [20]. In addition, CSL GBs have a much lower mobility as well [21]. Therefore, several simulations for annealing have been reported by taking these anisotropic energy and mobility of GBs into account [22–26]. Two-dimensional (2D) grain growth during coarsening has also been simulated and discussed for the evolution of grain orientation distribution [24–26]. Nevertheless, the geometric relation and interactions between CSL GBs have not been applied to the simulation of mc-Si directional solidification. Furthermore, there were few studies on the simulation of twinning nucleation. Pohl et al. [27] found that a stable twin only existed at the three-phase boundary by using molecular dynamic simulation. Nadri et al. [28] also added twinning to their model to simulate the grain structure in mc-Si ingot by using a geometric model. The results looked close to the experiments, but the model did not consider GB types and their interactions.

In this paper, we simulate the grain structure and the evolution of GBs during directional solidification of mc-Si sheet by phase field modeling. The geometric and energetic properties of $\Sigma 3^n$ GBs are considered. In the next section, the phase field model used in the simulation is briefly described. Section 3 is devoted to results and discussion followed by the conclusion in Section 4.

2. Phase field modeling

The phase field model (PFM) used here is based on the thin-interface model proposed by Karma and Rappel [29]. The phase field variable ϕ is set to 1 in the solid, -1 in the melt, and 0 at the interface. To represent the model in dimensionless form, the length is rescaled by W_0, which characterizes the interface thickness, and the time t is rescaled by τ_0, which characterizes the atomic movement. The dimensionless variables are denoted by a superscript asterisk, unless otherwise stated. The dimensionless phase field equation could be written as follows:

$$\begin{aligned}\tau^*(\mathbf{n})\frac{\partial\phi}{\partial t^*} &= \nabla^*\cdot[W^*(\mathbf{n})^2\nabla^*\phi] + [\phi-\lambda_c u(1-\phi^2)](1-\phi^2)\\ &\quad - H\left(\frac{1+\phi}{2}\right)|\Delta\theta| + \partial_{x^*}\left[|\nabla^*\phi|^2 W^*(\mathbf{n})\frac{\partial W^*(\mathbf{n})}{\partial(\partial_{x^*}\phi)}\right]\\ &\quad + \partial_{y^*}\left[|\nabla^*\phi|^2 W^*(\mathbf{n})\frac{\partial W^*(\mathbf{n})}{\partial(\partial_{y^*}\phi)}\right], \end{aligned} \quad (1)$$

where $\tau^*(\mathbf{n}) = \left[a_s^2(\mathbf{n}) + \frac{\beta_0 D_m}{a_1 a_2 W_0} a_s(\mathbf{n}) a_k(\mathbf{n})\right]$ for the thin-interface model, where $\mathbf{n}$ is the normal unit vector at the interface and $a_s(\mathbf{n})$ is the anisotropy function for the interfacial free energy, β_0 is the kinetic coefficient, D_m is the mean thermal diffusivity, a_1 and a_2 are constants, and $a_k(\mathbf{n})$ is the anisotropy function for the kinetic coefficient. Moreover, $W*(\mathbf{n}) = a_s(\mathbf{n})$ and λ_c is a coupling constant between the phase field and the temperature field. In addition, u is the dimensionless temperature, i.e., $u = C_{p,l}(T - T_m)/\Delta H$, where T is temperature, $C_{p,l}$ is the specific heat of the liquid, and ΔH is the heat of fusion. H is a parameter related to grain boundary energy, and $|\Delta\theta|$ is the angular difference between two grain orientations, as the coupling between orientation and phase fields. The relationship between H, $|\Delta\theta|$ and grain boundary energy γ_{gb} can be derived based on the same procedure in [30]. The highly anisotropic interfacial free energy $a_s(\mathbf{n})$ and kinetic $a_k(\mathbf{n})$ functions are selected from the previous work for the facet formation of silicon [31], which showed that the wavelength of facets was affected by $a_s(\mathbf{n})$ and $a_k(\mathbf{n})$ influenced the facet tips.

To model polycrystalline materials, especially for describing the CSL GBs, the normal vector of each grain needs to be calculated individually and correctly. The concepts of orientation-field [32] and multi-phase-field models [33] are adopted. We introduce N crystalline variables φ_i to the orientation-field equation for specifying the grains, where $i = 1, N$. For the crystalline variables, $\varphi_i Z$ is set to 1 for grain i, and 0 for the others. This crystalline field resemble the concept of the orientation field. The real orientation of a grain relative to a reference frame, θ, is assigned to each grain when φ_i exceeds a threshold, e.g., 0.9 is used here. More detail derivation and procedure has been discussed in [34]. Then, the crystalline evolution equations could be derived as follows:

$$\frac{\partial\varphi_i}{\partial t^*} = M^*_{\varphi_i}\cdot\nabla^*\cdot\left[h\cdot\left(\frac{1+\phi}{2}\right)^{\alpha}\cdot\frac{\nabla^*\varphi_i}{\sum_{i=1}^{N}|\nabla^*\varphi_i|}\right], \quad i = 1,2,\ldots,N \quad (2)$$

where $M^*_{\varphi_i} = (1-(\frac{1+\phi}{2})^2)\cdot a_{\varphi_i}$ is the mobility of each grain. Moreover, a_{φ_i} is the anisotropic function for grain boundary mobility; h and α are the interpolation parameters, which are set to 5 and 100 in our simulation for eliminating the crystalline field diffusion into the liquid phase. Larger α makes the boundary layer of crystalline field smaller, and this makes the determination of the orientation easier.

For example, if we have four seeds, the initial profiles of the grain crystalline variables φ_i ($i = 1, 4$) can be set, as shown in Fig. 1(a). The crystalline variable φ_i is set to 1 for grain i, and 0 for the rest. The orientation field variable θ is assigned to each grain, as shown in Fig. 1(b). In this study, we take θ as the tilt angle from $\langle 0\,1\,1\rangle$. When θ is 0°, the x-axis and y-axis are $\langle 0\,\bar{1}\,1\rangle$ and $\langle 1\,0\,0\rangle$, respectively.

To simplify our calculation, the frozen temperature approximation (FTA) is also adopted. The dimensionless temperature distribution is given by:

$$u = G^*\cdot(y^* - V^*\cdot t^*) \quad (3)$$

where G^* and V^* are the dimensionless temperature gradient and the drift velocity. A positive temperature gradient (G) of 200 K/mm and the drift velocity (V) of 2 mm/min are used in our simulation, which are close to our previous experimental parameters [17]. The temperature profile is shown in Fig. 1(c). The above equations including one phase equation in Eq. (1) and N crystalline equations in Eq. (2) are solved by an adaptive finite volume method [35]. The interface thickness W_0 is taken to be 2.5 μm, and the corresponding τ_0 is 9.5×10^{-3} s. The domain size and an example of the corresponding adaptive mesh are shown in Fig. 1(d). Detail numerical implementation can be found elsewhere [35].

3. Results and discussion

3.1. GB properties

Typical CSL GBs for Si $\langle 0\,1\,1\rangle$ are Σ3, Σ9, and Σ27, which have relatively lower GB energy. The Read-Shockley model [18] is considered for small angle GBs. The GB energy is controlled by the parameter H and is extended from [25] as follows:

$$\begin{cases} H = 12.5\cdot\left(1-\ln\left(\frac{\Delta\theta}{15^\circ}\right)\right) & \text{for } \Delta\theta < 15^\circ \\ H = 12.5\cdot\left(\frac{15^\circ}{\Delta\theta}\right)\cdot(1-A\cdot\cos^n(\Delta\theta-\theta_{CSL})) & \text{for } \Delta\theta > 15^\circ \end{cases}, \quad (4)$$