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On the growth orientation of twin-related faceted dendrites

G.Y. Chen, C.W. Lan *

Department of Chemical Engineering, National Taiwan University, Taipei 10617, Taiwan

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ABSTRACT

The growth orientation of twin-related faceted silicon dendrites depends on the undercooling ΔT in the melt, which cannot be explained by the existing models. We propose a model to explain the growth behavior showing that the re-entrant corners would lead to the growth of ⟨112⟩ dendrites, while the ⟨110⟩ branch is due to continuous growth. Such a selection could be determined by the velocity ratio $V_{\text{re-entrant}}/V_{\text{ridge}}$ that is a function of ΔT. An analytical expression for this ratio is derived and compared to the experimental observations.

Dendritic growth behaviors of faceted materials such as Si and Ge have been investigated for nearly sixty years due to their unique crystal morphologies [1–21]. The surface of Si- or Ge-faceted dendrites is bounded by ⟨111⟩ habit planes. The growth orientation and morphologies of faceted dendrites differ under different undercoolings. For instance, faceted dendrites grow in ⟨112⟩, ⟨110⟩, or ⟨100⟩ in different undercooled Si [8,11–21] or Ge melts [1,2,4–6,9,10]. More specifically, faceted dendrites grow in the ⟨112⟩ direction at 10 K < ΔT (undercooling) <15 K, while the faceted dendrites have a stable growth orientation in the ⟨110⟩ direction at 25 K < ΔT (undercooling) <100 K in an undercooled Si melt [14]. In addition, more than two parallel twin planes can be found at the center of these dendrites [14,15]. For the region where ΔT > 100 K, the growth orientation becomes ⟨100⟩ and no twin plane exists in a faceted dendrite [14].

For the last six decades, several growth models of twined crystal [3,4,16,18,22–24] have been proposed. The Twin-Plane-Reentrant-Edge (TPRE) mechanism [3] explains the parallel twin structure accelerates the growth of twin-related dendrites, as illustrated in Fig. 1. The nucleation event at the type-1 corners leads to the formation of a new growth step, with the type-2 corners. According to the nucleation theory, the nucleation rates at both types of re-entrant corners are in the same order of magnitude. The ridges are allowed to grow because of the nucleation at type-2 corners. The crystal front is therefore back to its original symmetry, but has increased in length in the direction that is allowed to grow from the re-entrant corners, as shown in the last crystal of Fig. 2. The TPRE mechanism explains the six-fold ⟨112⟩ propagation, but the branched mechanism is missing. Later, in situ observations of twin related dendrites have been reported [15–21]; the dependence of growth direction on twin spacing and undercooling has also been studied [20]. Moreover, Fujiwara et al. [18] proposed two modified growth models, i.e., the F-models, to explain different growth behaviors of ⟨112⟩ and ⟨110⟩ dendrites. The F-models, similar to the TPRE mechanism but only considering nucleation from the type-1 corners, suggest that the type-1 corners gradually disappear and form in repetition during the growth. Although some of their experimental observations supported their theory, we doubt the correctness of the F-models because some of the intermediate morphologies in the models were never observed during experiments. Detailed discussion could be found elsewhere [25]. Most importantly, the F-models could not explain the transition of the branched directions.

According the branching mechanism in snow crystals [26], the step density at facet centers steadily increases due to the Berg effect. When the step density at some points on facets exceeds a critical value, the surface attachment kinetics at facet centers changes to from faceted growth to the growth on a rough surface. The branched growth occurs, and the new branches should remain the same symmetry as the original crystal front. We consider that the branching of Si dendrites is because of the increasing of the step density as well. But the breakdown position of dendrites should be further considered due to the complex growth mechanism. Recently, we have simulated Si-dendritic growth via the phase-field model [25]. Our work revealed that the growth rates at re-entrants ($V_{\text{re-entrant}}$) and ridges (V_{ridge}) affect the branched direction of Si-dendrites. When $V_{\text{re-entrant}} > V_{\text{ridge}}$, the crystal sprouts along ⟨112⟩; when $V_{\text{re-entrant}} \sim V_{\text{ridge}}$, the crystal sprouts along ⟨110⟩. However, the detailed physics of the transition from ⟨112⟩ to ⟨110⟩ is still missing.

Our primary goal of this work is to propose a growth model explaining the transition from ⟨112⟩ to ⟨110⟩. We present a possible physical interpretation as follows. The Si-dendritic growth should involve both the TPRE mechanism and facet kinetics. For lower

* Corresponding author.
E-mail address: cwlan@ntu.edu.tw (C.W. Lan).

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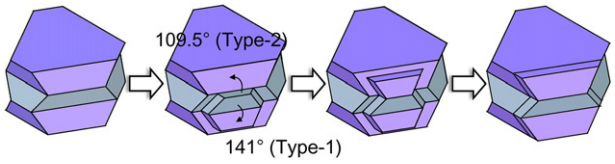

Fig. 1. Schematic of the TPRE mechanism [3]. The type-1 corners have an external angle of 141°, while the type-2 corners have an external angle of 109.5°.

undercooling, the Si-dendritic growth relies on the TPRE mechanism. If the lateral growth is much faster than the occurrence of the nucleation events, the step density is almost uniform across facets and the crystal would not breakdown into dendrites. Therefore, the lateral growth is considerable to the occurrence of the nucleation events when the branching occurs. If so, the growth velocity at ridges V_{ridge} is less than the growth velocity at re-entrants $V_{re\text{-}entrant}$. For the top layer of a twinned crystal, the growth velocity on one of the two ⟨112⟩ directions is $V_{re\text{-}entrant}$, while the other is V_{ridge}. If the velocity ratio $V_{re\text{-}entrant}/V_{ridge}$ is much greater than unity, the growth on the adjacent faces is unequal, resulting in ⟨112⟩ branching, as illustrated in Fig. 2(a). We also propose a plausible morphology of a twinned crystal with ⟨112⟩ branches as indicated in Fig. 2(a). In principle, the crystal could branch in all of the ⟨112⟩ directions if the ambient undercooling is sufficient [1]. However, in reality the branching might only occur in one or two of the ⟨112⟩ directions if one of the growth directions has a higher driving force [8,18]. This is particularly often when one branch consumes most of the undercooling.

For higher undercooling ($\Delta T > 25$ K), the nucleation occurs frequently on normal facets by two-dimensional (2D) nucleation. Also, the external angle of corners has little effects on the growth rate under this condition [27], which means that the growth of the ridges is no longer depending on the TPRE mechanism [14]. Then, $V_{re\text{-}entrant}$ is close to V_{ridge}, and the branching mechanism of the Si dendrite is similar to the one of snow. The step density at facet centers steadily increases during the crystal growth until reaching certain critical values, resulting in ⟨110⟩ branching, as illustrated in Fig. 2(b). One can expect that for extremely high undercooling, the growth eventually becomes the continuous growth. The branched direction alters to the direction of having the minimum interface stiffness, which is ⟨100⟩ for Si [14].

As compared to the F-models, the TPRE mechanism still plays an important role in our model. Another big difference between our model and the F-models is that the type-1 corner would not disappear at any moment of the growth. Our model explains the morphologies of Si dendrites. Moreover, we interpret the transition from ⟨112⟩ to ⟨110⟩ nicely, which is not clear from the description of the F-models. Unfortunately, there is no direct experimental evidence proving the correctness of our model. However, the experimental observations [20] have shown that the growth orientation of a dendrite is related to ΔT and twin spacing, d. As mentioned earlier, our model implies that ⟨112⟩ branching occurs when $V_{re\text{-}entrant}/V_{ridge} \gg 1$, and ⟨110⟩ branching occurs when $V_{re\text{-}entrant}/V_{ridge} \sim 1$. To further support our model, it is necessary to find the dependence of $V_{re\text{-}entrant}/V_{ridge}$ on ΔT and d.

We begin with an expression for facet growth developed by Obretenov et al. [28], which considers both the 2D nucleation and layer growth rates and has been compared to Monte Carlo computer simulations [29]:

$$V_{facet} = \frac{hJS}{1 + (J/v_l)^{2/3} S/\left(\beta b^{1/3}\right)} = \frac{h}{R_{facet}}, \tag{1}$$

where V_{facet} is the facet growth velocity, h is the monolayer height, J (m^{-2} s^{-1}) is the 2D nucleation rate which is proportional to $\exp[-\Delta G_i/(kT)]$, where ΔG_i is the formation energy of a nuclei, k is the Boltzmann constant, S (m^2) is the crystal face area, v_l is the lateral growth rate, β is a factor related to the geometric shape of the nuclei (π for circle and 4 for square), and b is a numerical coefficient near unity. The details of Eq. (1) can be found elsewhere [29]. We could also represent Eq. (1) by h/R_{facet}, where R_{facet} can be considered as the equivalent nucleation time or resistance for forming a facet. Eq. (1) should be able to describe the growth near re-entrants, and the expression for $V_{re\text{-}entrant}$ is thus similar:

$$V_{re-entrant} = \frac{hJ_{type-1}S}{1 + \left(J_{type-1}/v_l\right)^{2/3} S/\left(\beta b^{1/3}\right)} = \frac{h}{R_{type-1}}, \tag{2}$$

where $J_{type\text{-}1}$ is the 2D nucleation rate and $R_{type\text{-}1}$ is the equivalent nucleation time or resistance for a facet near the type-1 corners. For ridges, we assume that the growth follows the TPRE mechanism, which means the growth velocity of the ridges relies on the formation rate of the type-2 corners. To form a type-2 corner, it also needs to nucleate a facet on a type-1 corner first, followed by the lateral growth of the facet, until the gap between two twin planes is completely covered. Hence, the equivalent nucleation time for the TPRE mechanism R_{TPRE} equals to the sum of $R_{type\text{-}1}$, $R_{type\text{-}2}$ and R_l, where R_l is the time for covering the facet between the twin planes. Again, similar to $R_{type\text{-}1}$, $R_{type\text{-}2}$ can also be derived in a similar manner. Generally, the critical nucleus

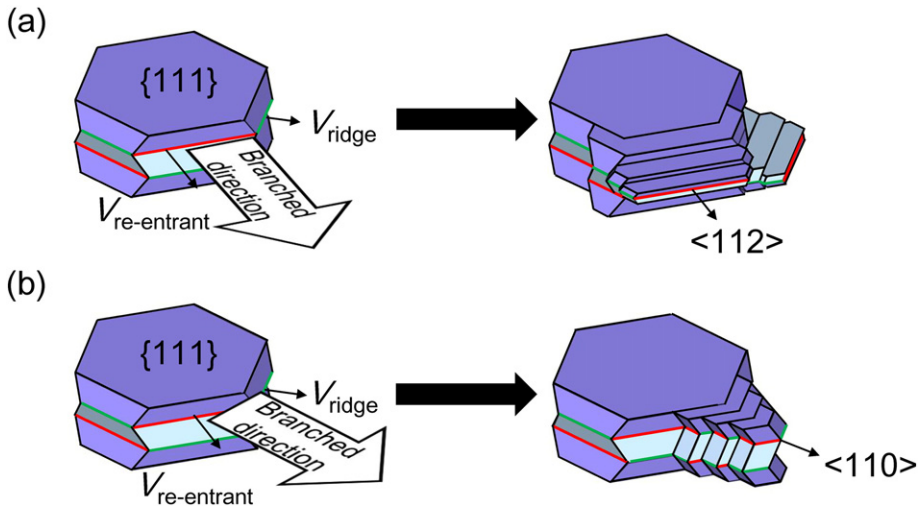

Fig. 2. (a) When the growth of ridges is mainly by the TPRE mechanism, the nucleation at re-entrants occurs frequently. Hence, $V_{re\text{-}entrant} > V_{ridge}$, resulting in ⟨112⟩ branching. (b) When the Si-dendritic growth is becoming continuous growth, the nucleation is allowed to occur at both re-entrants or ridges and $V_{re\text{-}entrant}$ becomes close to V_{ridge}, resulting in ⟨110⟩ branching.