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1 **Scaling law for growth of misoriented equiaxed Al-Cu dendrites: A phase-field study**
2 **with in situ experiment validation**

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6

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10

11 **Abstract**

12 We consider the effect of a mutual misorientation between two interacting equiaxed dendrites
13 in polycrystalline materials on the scaling law of growth from undercooled melt. This effect is
14 investigated by three-dimensional quantitative phase-field simulations of Al-Cu alloy
15 solidification in thin samples. It has been found that the equiaxed dendritic growth kinetics
16 changes due to the reduced solute interaction when considering mutual grain misorientation,
17 and both the characteristic growth rate and primary dendritic arm length can be approximately
18 correlated linearly to the misorientation of two dendrites. The scaling law, which originally
19 uses the solute composition and the distance between two nuclei as physical parameters to
20 describe the dynamics of primary dendritic arms, is then adapted with consideration of the
21 misorientation. Compared with the previous scaling law that only concerns face-to-face
22 growth of two dendrites, the predictions of the newly proposed scaling law taking the grain
23 orientation in account are in better agreement with the experimental data.

24 **Keywords** : Solidification, Equiaxed dendrite, Al-Cu alloys, Growth dynamics, Phase-field
25 method

26

27 **1. Introduction**

28 Microstructure formed during solidification strongly determines performance of materials,
29 such as mechanical properties and corrosion resistance [1], [2]. The dendrite is one of the
30 most common solidification microstructures, and the equiaxed grain is usually preferred in
31 practice to columnar grain, in order to obtain homogeneous and isotropic mechanical

32 properties in industrial metallic manufacture like aeronautics and automotive. Over the last
33 few decades, many experimental studies have focused on the equiaxed solidification process,
34 and it has been revealed that two growth kinetic stages are successively experienced by the
35 equiaxed grain after nucleation, namely the free growth and impinged growth regimes [3], [4].
36 At the beginning of the equiaxed grain growth in the early stages of solidification, a dendritic
37 grain is isolated from its neighbors, thus leading to an ever-increasing growth rate with
38 increasing undercooling. Whereafter, due to the solute rejection into liquid around the
39 dendrite during the liquid to solid transformation, a solute diffusion layer progressively builds
40 up ahead of the solid–liquid interface. After a while, the solute layer overlaps with that of
41 neighboring grains, which makes the dendrite growth slow down and even almost stop when
42 the solute interaction between neighboring grains becomes strong enough. These two
43 processes are responsible for the formation and evolution of the grain structure in most
44 polycrystalline materials.

45 In addition to experimental studies, several numerical methods have been proposed to
46 investigate the equiaxed dendrite growth during alloy solidification, as well as the underlying
47 physical mechanisms [1], [2]. The quantitative phase-field (PF) approach, based on the thin-
48 interface limit [5] and solute anti-trapping current [6], [7], has been recognized as the most
49 powerful tool to simulate dendrite growth because it avoids the explicit tracking of the
50 complicated solid–liquid interface [8], [9], [10]. Moreover, large-scale quantitative PF
51 simulations have been realized by means of the increasingly developed advanced numerical
52 algorithms, such as adaptive mesh refinement/coarsening and GPU parallel computing, which
53 also provides convenient tools to study solidification [11], [12], [13], [14]. Using the
54 quantitative PF method combined with in situ and real-time observation of solidification
55 experiments by synchrotron X-ray radiography, many works have been carried out to
56 investigate the growth and interaction of equiaxed dendrite during solidification [15], [16],
57 [17], [18], and a scaling law has been proposed to describe the dynamics of dendritic primary
58 arms, with the solute composition and the average distance between two nuclei as physical
59 parameters, which is given as [16], [17]

$$60 \quad \frac{L_m}{L} = \Lambda \quad (1)$$

61 where the characteristic primary dendritic arm length is $L_m = V_m \Delta t_m$, V_m is the maximum
62 growth rate of the primary arm tip, $\Delta t_m = t_m - t_m'$ with t_m and t_m' respectively corresponding to
63 V_m and $V_m' = V_m / 2$, $2L$ is the initial distance between two nuclei, and Λ is a constant which is

64 independent of the alloy composition. It has been indicated that $\Lambda = 0.537$ is a reasonable
65 value for equiaxed Al-Cu dendrites in thin samples, which has been validated by in situ and
66 real-time observed solidification experiments [16] and then revised by incorporating
67 thermodynamic effects [17]. However, the scaling law proposed in these two studies [16],
68 [17] only focused on the simple ideal case, in which the primary dendrite arms of two grains
69 grew in exactly opposite directions. However, the grain orientation in most of polycrystalline
70 materials is random. Therefore, one can suspect that the value of constant Λ and the scaling
71 law should also be related to the misorientation between primary dendrite arms of two
72 neighboring grains.

73 In this paper, the effect of misorientation between two grain primary arms on the scaling law
74 (1) is studied by using quantitative 3D PF simulations, accelerated by the fast simulation
75 schemes proposed in our previous work [14], including the nonlinear preconditioning of the
76 diffusive interface model, the front-tracking method to capture the orientation of grains, and
77 the parallel adaptive finite element method. The growth kinetics of the primary arm of an
78 equiaxed dendrite is quantitatively discussed. A novel scaling law that takes the
79 crystallographic orientation into consideration is proposed and then validated via the in situ
80 and real-time observation of solidification of Al-Cu alloys by synchrotron X-ray radiography.

81 **2. Model description and numerical implementations**

82 **2.1. Phase-field model**

83 The quantitative PF model presented in Ref. [7] is adopted to simulate isothermal
84 solidification of binary alloys under the continuous cooling condition. The quantitative
85 capability of this model to reproduce time evolution of microstructure during alloy
86 solidification has been demonstrated in many previous studies [16], [19], [20], [21]. The
87 governing equations of the phase field ϕ and the rescaled solute concentration U are
88 respectively given as

$$\left(1 + \frac{R_c t}{|m|C_0}\right) \tau(\mathbf{n}) \frac{\partial \phi}{\partial t} = \nabla \cdot \left[W(\mathbf{n})^2 \nabla \phi \right] + \sum_{i=x,y,z} \partial_i \left[|\nabla \phi|^2 W(\mathbf{n}) \frac{\partial W(\mathbf{n})}{\partial (\partial_i \phi)} \right] \quad (2)$$

$$+ \phi - \phi^3 - \lambda \left[U - \frac{R_c t}{(1-k)|m|C_0} \right] (1 - \phi^2)^2$$

$$\frac{(1+k)-(1-k)\phi}{2} \frac{\partial U}{\partial t} = \nabla \cdot \left\{ D \frac{1-\phi}{2} \nabla U + \frac{W_0 [1+(1-k)U]}{2\sqrt{2}} \frac{\partial \phi}{\partial t} \frac{\nabla \phi}{|\nabla \phi|} \right\} \quad (3)$$

$$+ \frac{1+(1-k)U}{2} \frac{\partial \phi}{\partial t}$$

with

$$U = \frac{2C/C_0 - [(1+k) - (1-k)\phi]}{(1-k)[(1+k) - (1-k)\phi]} \quad (4)$$

89

90 where C is the local solute concentration, C_0 is the initial concentration, k is the solute
 91 partition coefficient, R_c is the cooling rate, and m is the liquidus slope. The solid and liquid
 92 phases are represented by $\phi = +1$ and $\phi = -1$, respectively.

93 The spatial length and time are rescaled respectively by the interface width W_0 and relaxation
 94 time τ_0 , and therefore the dimensionless value for the solute diffusion coefficient in liquid is

$$\tilde{D} = \frac{D\tau_0}{W_0^2} = a_1 a_2 \frac{W_0}{d_0} \quad (5)$$

95

96 where $d_0 = \Gamma/[mC_0(k-1)]$ is the chemical capillary length with Γ the Gibbs-Thomson
 97 coefficient, $a_1 =$ and $a_2 = 47/75$ [7]. The coupling coefficient λ in Eq. (2) is taken as
 98 $\lambda = a_1 W_0/d_0$. The standard form of the fourfold symmetry of the surface energy anisotropy is
 99 considered, and thus $W(\mathbf{n}) = W_0 a_s(\mathbf{n})$ and $\tau(\mathbf{n}) = \tau_0 a_s(\mathbf{n})^2$ where

$$a_s(\mathbf{n}) = 1 - 3\varepsilon_4 + 4\varepsilon_4 \sum_{i=x,y,z} \frac{(\partial_i \phi)^4}{|\nabla \phi|^4} \quad (6)$$

100

101 In order to accelerate large-scale quantitative PF simulations, a nonlinear preconditioning [22]
 102 of the phase field ϕ is employed to allow coarser grids to be used in the interface layer. The
 103 transformed phase field ψ is given as

$$\psi = \tanh\left(\frac{\phi}{\sqrt{2}}\right) \quad (7)$$

104

105 And hence the standard PF model (Eqs. (2), (3)) is transformed to the nonlinear
 106 preconditioned formula, as follows:

$$\left(1 + \frac{R_c t}{|m|C_0}\right) \tau_0 a_s^2(\mathbf{n}) \frac{\partial \psi}{\partial t} = W_0^2 a_s^2(\mathbf{n}) \left[\nabla^2 \psi - \sqrt{2} \tanh\left(\frac{\psi}{\sqrt{2}}\right) |\nabla \psi|^2 \right] \quad (8)$$

$$+ \sum_{i=x,y,z} W_0^2 \partial_i \left[|\nabla \psi|^2 a_s(\mathbf{n}) \frac{\partial a_s(\mathbf{n})}{\partial (\partial_i \psi)} \right] + \sqrt{2} \tanh\left(\frac{\psi}{\sqrt{2}}\right) - \sqrt{2} \lambda \left[1 - \tanh^2\left(\frac{\psi}{\sqrt{2}}\right) \right] \left[U - \frac{R_c t}{(1-k)|m|C_0} \right]$$

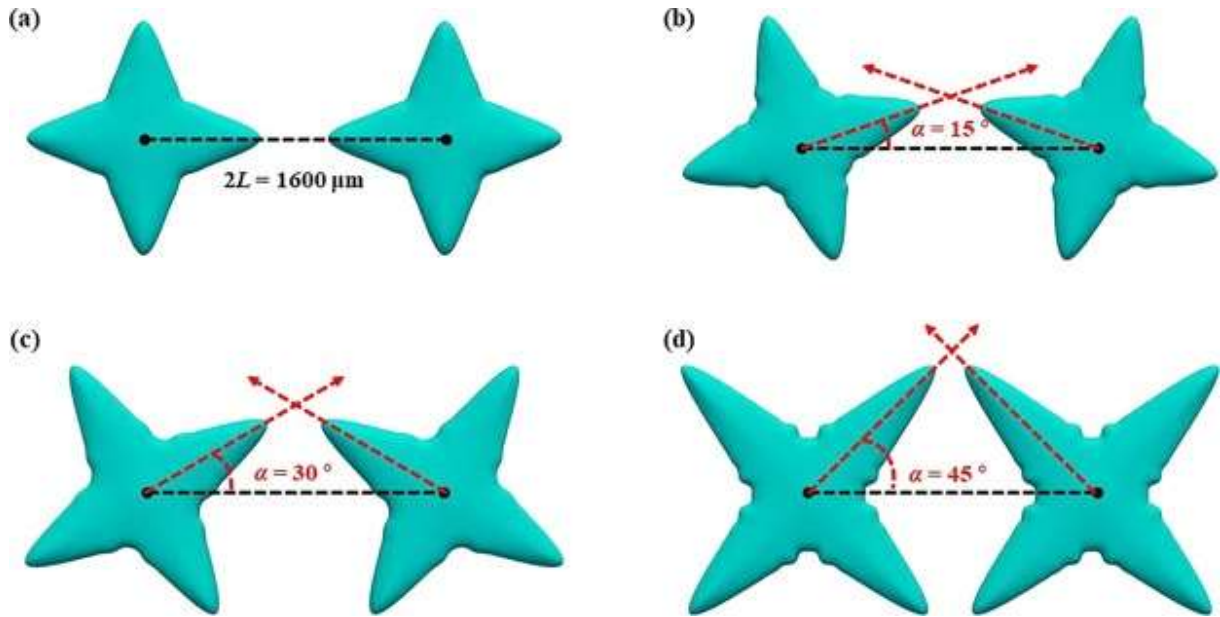
$$\begin{aligned} & \frac{(1+k)-(1-k) \tanh\left(\frac{\psi}{\sqrt{2}}\right)}{2} \frac{\partial U}{\partial t} \\ & = \nabla \cdot \left\{ D \frac{1-\tanh\left(\frac{\psi}{\sqrt{2}}\right)}{2} \nabla U + \frac{1}{2} W_0 [1 + (1-k)U] \frac{1-\tanh^2\left(\frac{\psi}{\sqrt{2}}\right)}{2} \frac{\partial \psi}{\partial t} \frac{\nabla \psi}{|\nabla \psi|} \right\} \\ & \quad + \frac{1+(1-k)U}{2} \frac{1-\tanh^2\left(\frac{\psi}{\sqrt{2}}\right)}{\sqrt{2}} \frac{\partial \psi}{\partial t} . \end{aligned} \quad (9)$$

107

108 The numerical accuracy and efficiency of the preconditioned PF formula has been examined
 109 and validated [14]. The detailed numerical and material parameters can be referred to our
 110 previous studies [15], but here C_0 is set to be 1 wt% to result in a larger capillary length.
 111 Large-scale simulations can be therefore realized quantitatively in a relatively wide range of
 112 grain misorientation. In addition, for consistency of conclusions, the values of several material
 113 parameters, such as the solute diffusion coefficient in liquid, the slope of liquidus, the
 114 anisotropy strength of interfacial energy and the Gibbs-Thomson coefficient are the same as
 115 those in the recent literature [16], [17].

116 2.2. Convergence test

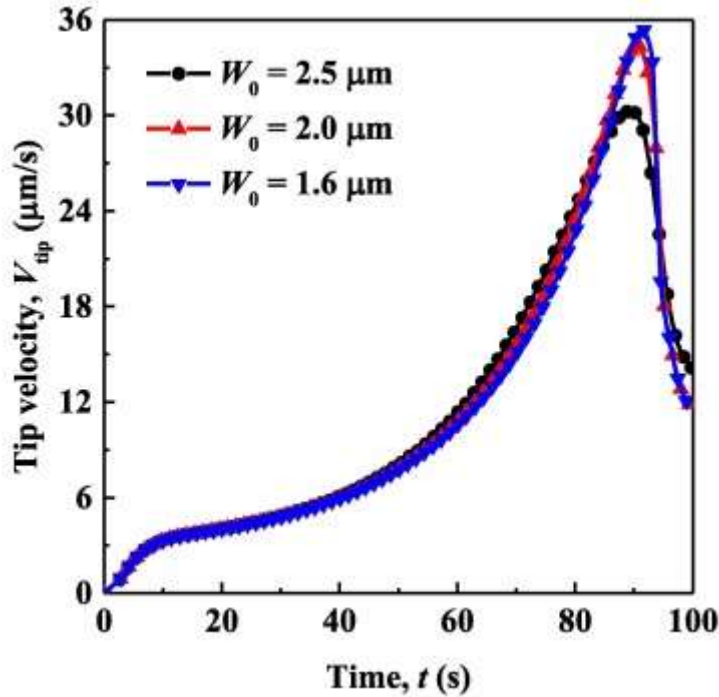
117 To achieve quantitative 3D PF simulations, the interface width, W_0 , must be carefully
 118 determined to obtain a reasonable compromise between the computational accuracy and
 119 efficiency. The numerical convergence test is carried out for the simulation with
 120 $\Delta\theta = 2\alpha = 90^\circ$ and the distance between two nuclei is $2L = 1600 \mu\text{m}$, for which the dendrites
 121 grow in the fastest rate. $\Delta\theta$ is the angle between the direction of primary dendrite arm growth
 122 of two grains in the thin sample, and α is the misorientation. The two grains are placed in the
 123 same xOy plane, so that the misorientation is preset by rotating the dendrites around the z -
 124 axis, and the specific configuration of two mutually misoriented interacting dendrites can be
 125 seen in Fig. 1. It is assumed that both grains nucleate simultaneously and therefore the grain
 126 morphologies are the same.



127

128 *Fig. 1. Configuration and morphology of the two interacting equiaxed Al-1.0 wt% Cu*
 129 *dendrites at t_m with different misorientations and $L = 800 \mu\text{m}$. (a) $\alpha = 0^\circ$; (b) $\alpha = 15^\circ$; (c)*
 130 *$\alpha = 30^\circ$; (d) $\alpha = 45^\circ$. The red dashed arrows represent the growth directions of primary arm*
 131 *tips. (For interpretation of the references to colour in this figure legend, the reader is referred*
 132 *to the web version of this article.)*

133 The simulated tip velocities of the primary dendrite arm with different interface width in the
 134 numerical convergence test are plotted in Fig. 2. As expected, a good convergence is observed
 135 as W_0 is reduced. However, the computing amount will also increase significantly with W_0
 136 decreasing, which means that we need to enlarge W_0 to reach accessible computing time. The
 137 maximum tip velocities at the end of the free growth stage, V_m , are 30.2, 34.3 and 35.4 $\mu\text{m/s}$,
 138 respectively for the cases with $W_0 = 2.5, 2.0$ and $1.6 \mu\text{m}$. The relative deviation of V_m for the
 139 cases with $W_0 = 2.0$ and $1.6 \mu\text{m}$ is only about 3 %, so it is reasonable to make a choice of
 140 $W_0 = 1.6 \mu\text{m}$ for all the 3D PF simulations in the present work.



141

142 *Fig. 2. Tip velocities of the equiaxed Al-1.0 wt% Cu dendrites with $\alpha = 45^\circ$ and the initial*
 143 *distance between two nuclei being $2L = 1600 \mu\text{m}$.*

144 **2.3. Numerical implementations**

145 The parallel adaptive finite element method, with multiple processors using distributed
 146 memory based on the MPI protocol, is employed to solve the nonlinearly preconditioned PF
 147 model. Numerical implementations are realized by the open-source deal.II library [23], and
 148 the adaptive mesh refinement/coarsening is achieved through a simple but efficient approach
 149 [14]. The front-tracking method [14] is used to capture grain orientations to improve the
 150 computing efficiency dramatically. The computational domain size is $4L \times 4L \times 200 \mu\text{m}$
 151 ($L = 600, 700, \text{ and } 800 \mu\text{m}$), and its thickness is consistent with that of the samples observed
 152 in situ and real-time by synchrotron X-ray radiography in the solidification experiments [16].
 153 At the beginning of simulations, the center-to-center distance between the two nuclei is set to
 154 be $2L$ and the two grains nucleate simultaneously with the misorientation being in the range of
 155 $0-45^\circ$. In simulations, the larger the angle, the larger the maximum growth rate (at the end of
 156 the free-growth stage), and consequently the longer the calculation time is. All the simulations
 157 were carried out on a supercomputer with an Intel® Xeon E5-2683 v4 CPU (2.10 GHz and 32
 158 cores) and 256 GB memory at each computing node. For the 3D simulation with the largest
 159 misorientation ($\alpha = 45^\circ$) and the largest computational domain size ($L = 800 \mu\text{m}$), it took
 160 about 35 h using 192 cores to finish the computation, when the dendritic growth came into the

161 impinged-growth regime in which the growth rate of the primary dendrite arm tip sharply
162 decreased.

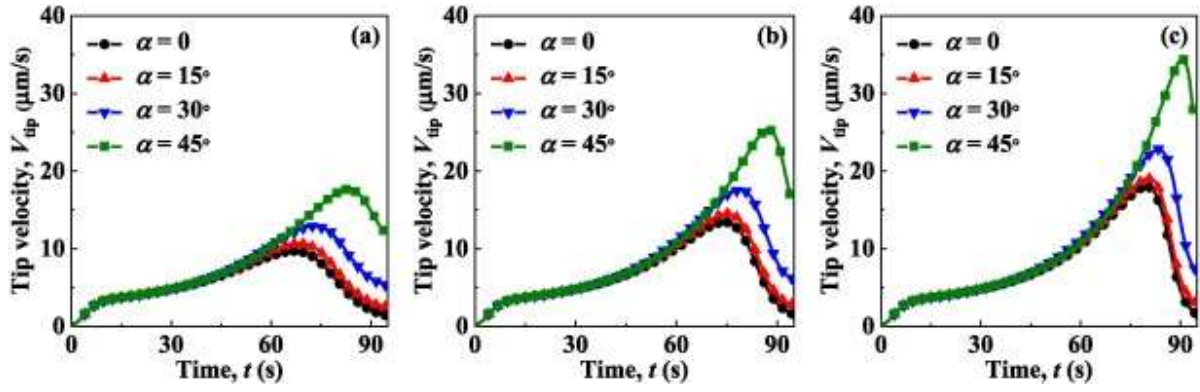
163 **3. Results and discussions**

164 **3.1. Dendrite growth kinetics**

165 The equiaxed dendritic morphologies of two grains with various misorientations at time t_m
166 when the tip growth rate reaches its maximum are shown in Fig. 1. It is visible that the length
167 of the primary dendritic arm increases with increasing the grain misorientation owing to the
168 reduced solute interaction. The reason for the reduced solute interaction can be attributed to
169 the fact that the solute boundary layer overlapping is delayed when misorientation is larger.
170 Therefore, it seems natural to conclude that there are significant differences in dendrite
171 growth kinetics with various misorientations, resulting in that the effect of the dendrite
172 orientation must be considered in the scaling law (1) to give a more general description of
173 equiaxed dendrite growth in polycrystalline materials.

174 In order to compare the dendrite growth kinetics in detail, the growth velocities of the primary
175 dendrite arm tips (marked by the red arrows as shown in Fig. 1) are measured, and the results
176 are plotted in Fig. 3. Without misorientation ($\alpha = 0$), the maximum tip growth velocity $V_{m,0}$ as
177 well as the characteristic length of the primary dendrite arm $L_{m,0}$ are both given in Table 1.
178 The results of the present work are in quite good agreement with the reference values [16],
179 [17]. The relative error due to the measurement time interval or calculation method is less
180 than 10 %, both in terms of the tip growth velocity and the characteristic primary arm length.
181 And this also adequately confirms that the simulated results in the present work are
182 sufficiently reliable to study the effect of mutual misorientation of two interacting equiaxed
183 dendrites on the scaling law.

184



185

186 *Fig. 3. Growth kinetics of the equiaxed Al-1.0 wt% Cu dendrites for different grain*
 187 *misorientations and computational domain sizes. (a) $L = 600 \mu\text{m}$; (b) $L = 700 \mu\text{m}$; (c)*
 188 *$L = 800 \mu\text{m}$.*

L (μm)	$V_{m,0}$ ($\mu\text{m/s}$)	Refs. [16], [17]	$L_{m,0}$ (μm)	Refs. [16], [17]
600	9.710	9.360	345.1	322.2
700	13.44	12.74	385.7	375.9
800	18.02	16.64	418.3	429.

189 *Table 1. Comparison of the values of V_m and L_m with $\alpha = 0$ and $L = 600, 700,$ and $800 \mu\text{m}$,*
 190 *between the present work ($W_0 = 1.6 \mu\text{m}$) and prediction by scaling laws in references [16],*
 191 *[17].*

192

193 Moreover, as can be seen in Fig. 3, the tip growth velocity at the same time (or undercooling)
 194 increases gradually with misorientation, which is due to the decreasing solute interaction
 195 between neighboring grains. To be specific, the ratio between the maximum tip velocity and
 196 the value with $\alpha = 0$ could be expressed as a power function of the misorientation, namely

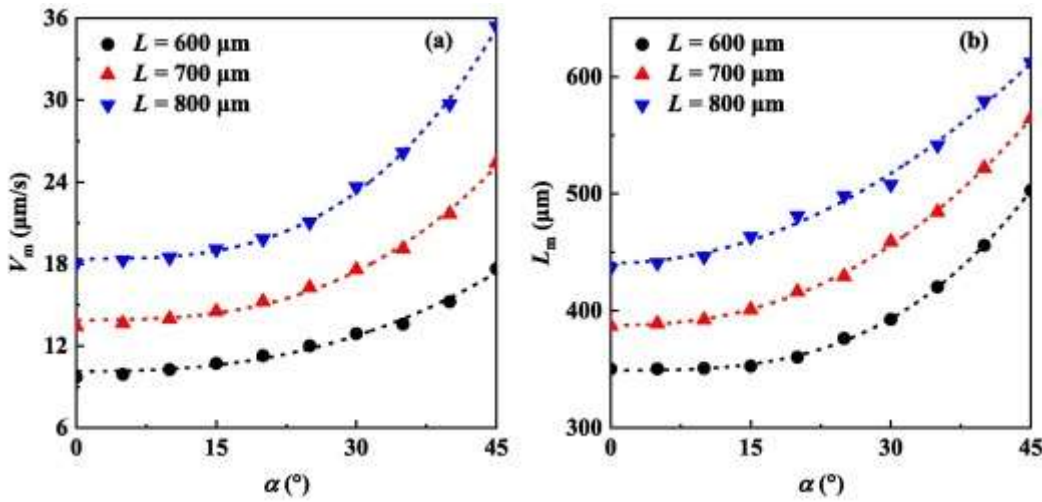
$$\frac{V_m}{V_{m,0}} = 1 + a\alpha^b \quad (10)$$

197

198 with a and b being two positive fitting parameters. The values of $V_m/V_{m,0}$ are shown in Fig.
 199 4(a), from which it is obvious that $V_m/V_{m,0}$ evolves with α as a power function. The power
 200 correlation with the misorientation is also valid for the ratio of the characteristic primary arm
 201 length, $L_m/L_{m,0}$, which could be written as

$$\frac{L_m}{L_{m,0}} = 1 + a' \alpha^{b'} \quad (11)$$

202
 203 with a' and b' being two other positive fitting parameters. The power relationship between the
 204 characteristic primary arm length and misorientation could be confirmed further from Fig.
 205 4(b). In general, a clear conclusion which can be drawn from Fig. 4 is that, the solute
 206 interaction between the primary dendrite arm of two neighboring grains will be changed when
 207 different grain orientations are taken in account, which leads to the change of dendritic growth
 208 kinetics. Consequently, the scaling law proposed for $\alpha = 0$ [16], [17] where the grain
 209 misorientation was ignored should be necessarily modified to adapt to the present more
 210 common situation ($\alpha \neq 0$).



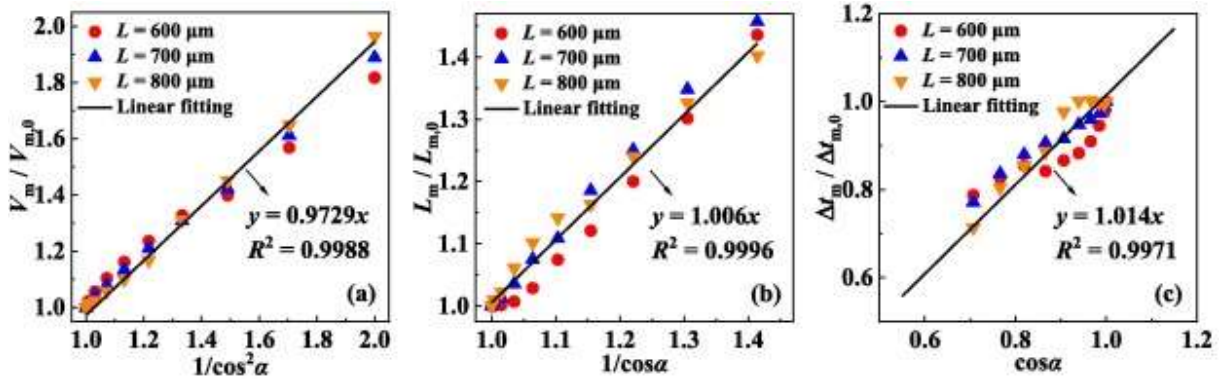
211
 212 *Fig. 4. Plotting of (a) V_m and (b) L_m with misorientations α . The data points are from the*
 213 *corresponding 3D PF simulations, while the dashed curves are the power law fitting.*

214 3.2. Scaling law

215 The previous section reveals that both the maximum tip growth velocity V_m and the
 216 characteristic primary arm length L_m are correlated to the misorientation α , as a power
 217 function. However, the power relationship is not a general law, since in practice the values of
 218 the two fitting parameters in Eq. (10) and Eq. (11) vary with the distance between the two
 219 dendrites. What is interesting, though, is that we can find a more general correlation between
 220 $V_m/V_{m,0}$ or $L_m/L_{m,0}$ and α . According to Fig. 5(a) and Fig. 5(b), there is a quite good linear
 221 relationship between $V_m/V_{m,0}$ and $1/\cos^2\alpha$, as well as between $L_m/L_{m,0}$ and $1/\cos\alpha$. Based on

222 this, it is easy to infer that the ratio of $\Delta t_m/\Delta t_{m,0}$ also varies with $\cos\alpha$ in an approximate linear
 223 manner, which has been validated in Fig. 5(c).

224



225

226 Fig. 5. Variation of (a) V_m , (b) L_m and (c) Δt_m with the grain misorientation α .

227 For the ratio of $V_m/V_{m,0}$, it could be expressed as $V_m/V_{m,0} = p/\cos^2\alpha$, and the value of the fitting
 228 parameter $p \approx 0.9729$ is approximately equal to 1. For the ratio of $L_m/L_{m,0}$, it could be
 229 expressed as $L_m/L_{m,0} = q/\cos\alpha$, and the values of the fitting parameter $q \approx 1.006$ is also
 230 approximately equal to 1. The rounding of fitting parameters ensures that the values of V_m and
 231 L_m exactly reduce to $V_{m,0}$ and $L_{m,0}$ when the misorientation is vanishing ($\alpha = 0$). Therefore,
 232 both $V_m/V_{m,0}$ and $L_m/L_{m,0}$ can be written as simple functions of α as

$$\frac{V_m}{V_{m,0}} = \frac{1}{\cos^2 \alpha} \quad (12)$$

$$\frac{L_m}{L_{m,0}} = \frac{1}{\cos \alpha} \quad (13)$$

233

234 and then the ratio of $\Delta t_m/\Delta t_{m,0}$ can be derived as

$$\frac{\Delta t_m}{\Delta t_{m,0}} = \cos \alpha \quad (14)$$

235

236 The dynamics scaling considered in the present work focuses on the time interval when the
 237 dendrite tip crosses over from a free-growth regime to a poisoned-growth one. Therefore, one
 238 can reasonably expect that the scaling in this time interval is not governed by the late solute
 239 interaction between dendrites. For the moment, we can thus stick to the assumption that
 240 geometric factors are dominant, which is consistent with Eq. (13). Indeed, for $\alpha = 0$ the
 241 scaling law $L_m = \Lambda L$ was recently justified on the sole basis of geometric arguments [24]. The
 242 Eq. (12) is essentially based on the solute conservation at the interface of the dendritic arm

243 tip, i. e., $V_i(1 - k)C_{L,i} = -D_L|\nabla C_{L,i}|$ where V_i is the propagation of tip interface, $C_{L,i}$ is tip
 244 solute concentration, and $|\nabla C_{L,i}|$ is gradient of tip concentration. Moreover, the solute in liquid
 245 approximately satisfies the exponential decay function, namely $C(\mathbf{r}) = C_0 +$
 246 $(C_{L,i} - C_0)\exp[-2(\mathbf{r} - \mathbf{r}_0)/L_s]$ with \mathbf{r}_0 the position of the tip interface and L_s the width of solute
 247 boundary layer. Combining the two relationships above, we can infer that $V_m \propto$
 248 $(1 - C_0/C_{L,i}) \cdot (1/L_s)$, and both terms in the product increase as the space of free growth
 249 increases. Therefore, it is natural that V_m is proportional to L_m to the second power as
 250 $V_m \propto L_m^2 \propto (1/\cos\alpha)^2$.

251 According to the expressions given in Refs. [16], [17], the final scaling law considering the
 252 grain misorientation can be expressed as

$$V_m = V_{m,0} \frac{1}{\cos^2 \alpha} \approx 2.6 \times 10^{-5} \frac{L^2}{C_0^{2/3}} \frac{1}{\cos^2 \alpha} \quad (15)$$

$$L_m = L_{m,0} \frac{1}{\cos \alpha} = \Lambda L \frac{1}{\cos \alpha} \quad (16)$$

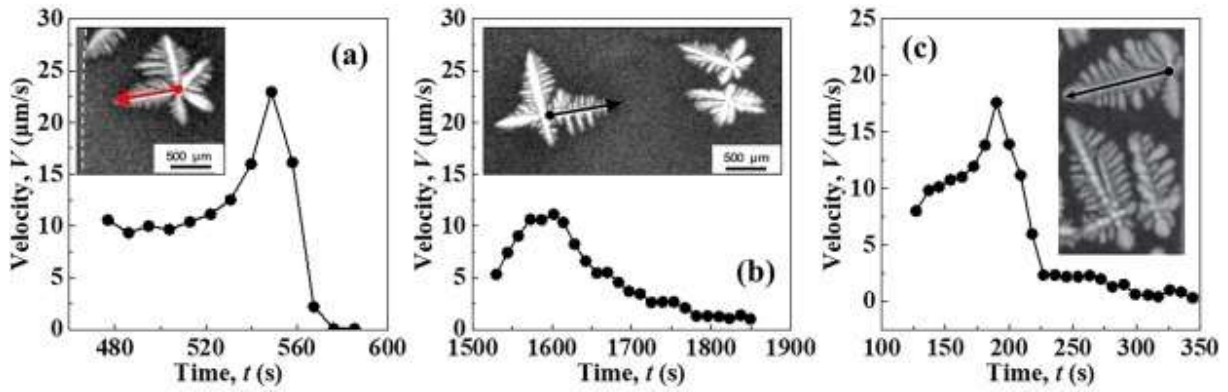
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254 where the value of the constant Λ was revealed to be 0.546 for the first time [16], and then re-
 255 calibrated to 0.537 through analysis of additional PF simulations considering a curved
 256 thermodynamic phase diagram and spatiotemporal noise [17].

257

258 3.3. Comparison with experimental data

259 In order to validate the scaling law considering the grain misorientation, the length of the
 260 characteristic primary dendritic arm predictions by Eq. (13) are compared to the experimental
 261 data, which was measured from the in situ and real-time observation of the of Al-Cu alloys
 262 solidification experiments by synchrotron X-ray radiography [15], [16]. A detailed description
 263 of the solidification experiment and in situ and real-time observation is given in [16]. Here we
 264 still chose the two dendrites from [16] and one from [15]. For the first dendrite shown in Fig.
 265 6(a), since there is no neighbor grains around the selected arm, the misorientation is measured
 266 according to the angle between the selected arm and the horizontal direction, which has a
 267 value of $\alpha = 9.20^\circ$. For the other two dendrites with neighbor grains in Fig. 6(b, c), the
 268 misorientations are 9.98° and 51.7° , respectively, which are measured according to Fig. 1.



269

270 *Fig. 6. Tip growth rates of the three dendrites in the solidification experiments of Al-Cu alloys*
 271 *by isothermal cooling at 0.5 K/min. (a) and (b) are from [16] and (c) is from [15].*

272

273 The experimental data and the predictions by the scaling law proposed for $\alpha = 0$ [16], [17] and
 274 for $\alpha \neq 0$ (present work) are given in Table 2. The characteristic primary arm length, L_m , is
 275 calculated as the product of the maximum tip growth velocity, V_m , and the time interval
 276 ($\Delta t_m = t_m - t_m'$) between the time when the tip growth velocity reaches V_m and $V_m / 2$. For the
 277 first grain with $\alpha = 9.20^\circ$, the measured characteristic primary length, L_m , is 575 μm . The
 278 value predicted for $\alpha = 0$ [16], [17] is 563 μm , while the value predicted for $\alpha = 9.20^\circ$ by Eq.
 279 (16) is 571 μm , which is in a better agreement with the experimental data. For the second
 280 grain with $\alpha = 9.98^\circ$, the measured characteristic primary length, L_m , is 760 μm , which is quite
 281 different from the predicted results of the two scaling laws, regardless of the misorientation is
 282 taken into account or not. The reason for this large discrepancy ($\sim 15\%$) is that the two grains
 283 nucleated at different instants, thus leading to that the second grain had already been growing
 284 for some time when its neighbors nucleated. As mentioned earlier, in the present PF
 285 simulations and the derivation of the scaling law, the difference in nucleation time between
 286 neighboring grains is not considered, which could explain why the predicted values are
 287 smaller than the experimental data. For the third grain with a larger misorientation
 288 ($\alpha = 51.7^\circ$), both lower values predicted by PF simulations compared to experimental data
 289 could be attributed to the time lag between grain nucleations. However, the result predicted in
 290 this work ($L_m = 805 \mu\text{m}$) is much better than that predicted by the scaling law without
 291 misorientation ($L_m = 499 \mu\text{m}$), which suggests that the scaling law considering grain
 292 orientation proposed in the present work can indeed predict values more consistent with the
 293 experimental data.

Experimental data L_m value predicted by Eq. (16)

L μm	α degree	L_m μm	$\alpha = 0$ μm	$\alpha \neq 0$ μm
1050	9.20	575	563	571
1200	9.98	760	643	654
930	51.7	1036	499	805

294 *Table 2. Comparison of the values of the characteristic primary arm length between the*
 295 *experiment and predictions by scaling laws.*

296 **4. Conclusions and outlooks**

297 In the present work, the effect of the grain orientation on the scaling law of equiaxed dendrite
 298 growth of Al-Cu alloys [16], [17] was investigated by quantitative 3D PF simulations. It was
 299 revealed that for random grain orientations, which is the case in polycrystalline materials, the
 300 solute interaction between two neighboring grains was generally reduced and the grain growth
 301 kinetics changed accordingly. To be more specific, the duration of the free-growth period of
 302 the equiaxed dendritic growth was shown to be longer with the increase of the misorientation,
 303 so that the maximum dendritic tip growth rate V_m and the characteristic primary dendritic arm
 304 length L_m also increase. Therefore, a good correlation of V_m or L_m with grain misorientation
 305 was obtained using a simple power function.

306 Furthermore, approximate linear relationships between $V_m/V_{m,0}$ or $L_m/L_{m,0}$ and α was
 307 established to get a more general, concise, and applicable scaling law. Then we compared the
 308 predicted values based on the new scaling law with those given by the previous relationship
 309 without considering the grain orientation, as well as the experimental data measured from in
 310 situ and real-time observation of Al-Cu alloys solidification by synchrotron X-ray
 311 radiography. Despite large discrepancy between the predicted values and the experimental
 312 data, most likely due to the different nucleation times of the interacted grains, the new scaling
 313 law proposed in the present work could still give predictions which are in much better
 314 agreement with the experimental results than the original scaling law for $\alpha = 0$. In the future
 315 work, other complex factors in the actual polycrystalline solidification will be considered,
 316 such as the nucleation time difference as just mentioned, the cooling rate, and the liquid
 317 natural convection arising from the uneven distribution of solute, and even the case where two
 318 grains are not mirror symmetric. Then a more general scaling law to be applied in complex
 319 solidification conditions is likely to be established. To further validate the scaling law

320 proposed in the present work, it would also be interesting to perform solidification
321 experiments in which grain nucleations are simultaneously provoked in the sample.

322

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