# Growth models of pure supercooled materials

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For a pure material, the dynamics of the growth of one phase in a supercooled other phase for the case of a shallow temperature quench is traditionally understood via a kinetic thermal diffusion equation model or a quasistatic Laplace equation model, if order-parameter details can be neglected. In the quasistatic model, the interfacial boundary temperature $T_R$ is equal to the phase transition temperature $T_m$. In the kinetic model, however, growth is driven by a nonzero interfacial undercooling $T_m - T_R$. By assuming that the growth process occurs at small but finite, identical spatial steps, the growth laws for the cases of shallow and deep temperature quenches were derived analytically from the kinetic model in the limit of zero thermal diffusivity. For the case of a shallow temperature quench, it is shown that the apparent difference between the assumed interfacial boundary conditions of the quasistatic and the kinetic model does not exist.

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## I. INTRODUCTION

Understanding the phase-ordering of materials after a quench, i.e., the rapid change of an intensive thermodynamic variable of state, like temperature or pressure, is of fundamental scientific importance [1]. This is because phase-ordering is involved in the processing of a wide range of daily life materials, ranging from metal alloys, glasses, and polymers, to the crystallization of pharmaceutical organic compounds [1]. During a temperature-quench experiment, the temperature $T$ of a system is rapidly decreased across a transition temperature $T = T_m$ to a final temperature $T = T_o < T_m$. The stable low-temperature phase then grows isothermally at $T = T_o$ in the sea of the unstable high-temperature phase.

Nonequilibrium growth processes of this kind often result in the nontrivial observation of a variety of macroscopic patterns and scaling dynamics, in solids as well as liquid crystals [2–15]. Observed growth patterns include cubes [5], circular germs [6–8], bâtonnets [9,10], dendritic structures [2,3,11], and fractal structures [12–14]. For simple Euclidean patterns, the growth process is often characterized by a characteristic length $R$ of the growing germ as a function of time $t$. $R$ is usually chosen as the radius or diameter of a circular germ [6–8], or the long or short axis of a bâtonnet [9,10]. In the experiments, the growth dynamics can generally be described by a growth law $R \sim t^n$, where $n$ is the growth exponent. For a number of liquid crystalline systems, it was found that this exponent changes from $n = 1/2$ to 1 for increasing quench depth $T_m - T_o$ [6–10]. Exemplary experimental growth data of a liquid crystal material are presented in Fig. 1.

Existing theoretical treatments for the crystal growth dynamics of pure materials consider at least two aspects: (a) a temperature field $T$, and (b) a scalar order-parameter field $\phi$.

In pure order-parameter models [15], also known collectively as "phase-field" models, the temperature is assumed to be uniform at $T = T_o$ everywhere, and the time-dependent Ginzburg-Landau equation

$$\frac{\partial \phi}{\partial t} = -\frac{\delta F}{\delta \phi} \tag{1}$$

is employed to describe the dynamics of a nonconserved order parameter $\phi$. $F$ is a Landau free-energy functional given by

$$F(\phi) = \int \left[ \frac{1}{2}|\nabla \phi|^2 + V(\phi) \right] d^d x \tag{2}$$

for a $d$-dimensional system. In this expression, $V(\phi)$ is a double-well potential where the difference between its local minima drives the growth of one phase over the other. The interface between the phases is said to be "diffuse" because its spatial variation of $\phi$ is continuous. On the other hand, pure thermal diffusion models [1,16] consider only the diffusion equation

$$\frac{\partial T}{\partial t} = \kappa \nabla^2 T \tag{3}$$

for a nonuniform temperature field $T$, which arises from a continuous generation of transition enthalpy at the moving phase boundary ($\kappa$ is the thermal diffusivity). In contrast to pure order-parameter models, the interface between the phases is assumed to be sharp with a step change in $\phi$. Order-parameter details of the system are thus completely neglected. A third class of models takes into account both aspects of thermal diffusion and order-parameter evolution [17–19], giving a more general description of crystal growth. The focus of this study is on pure thermal diffusion models for circumstances where order-parameter details are not important.

This paper is organized as follows: In Secs. II and III, the quasistatic Laplace equation model and the kinetic diffusion equation models are analyzed. In Sec. IV, the $R \sim t^{1/2}$ and $R \sim t$ growth laws for the cases of shallow and deep temperature quenches, respectively, are derived analytically from the

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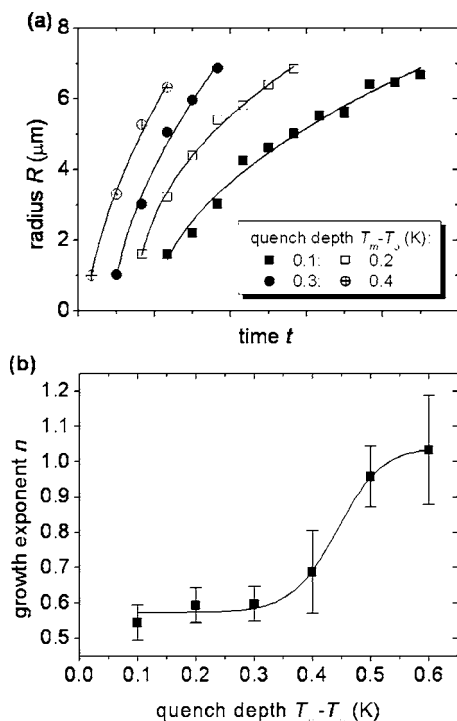
FIG. 1. Experimental data from Ref. [21] for the radial growth of the cholesteric phase of a liquid crystal mixture ZLI-5014-100: (a) Exemplary growth data $R(t)$ (radius of a circular germ) at varying quench depths. The time interval between successive data points is 1 s. At the smallest quench depth $T_m - T_o = 0.1$ K, there is considerable deceleration of the growth rate with $R \sim t^{1/2}$. As the quench depth increases, a constant growth rate with $R \sim t$ is approached. Each solid curve was obtained by fitting to a power law. (b) Growth exponent $n$ as a function of quench depth, showing an increase from $n = 1/2$ to 1 for increasing quench depth. The solid line is a guide to the eye.

kinetic model in the limit of zero thermal diffusivity. In the derivation of the $R \sim t^{1/2}$ growth law, the problem of the apparent difference between the assumed interfacial boundary conditions of the quasistatic and the kinetic model for the case of a shallow temperature quench is addressed.

## II. QUASISTATIC LAPLACE EQUATION MODEL

One model involves the Laplace equation for a quasistatic three-dimensional temperature field outside a growing sphere [1,20]. For shallow temperature quenches, the slowly growing sphere is regarded as a quasistationary object that generates transition enthalpy, slowly and continuously without any thermal turbulence. The temperature profile outside this heat source is thought to be approximately steady, i.e., $\partial T / \partial t \approx 0$. For any finite thermal diffusivity $\kappa$, this corresponds to the Laplace equation $\nabla^2 T = 0$. For a three-dimensional temperature field with radial symmetry, it is

$$\nabla^2 T = \frac{\partial^2 T}{\partial r^2} + \frac{2}{r} \frac{\partial T}{\partial r} = 0, \tag{4}$$
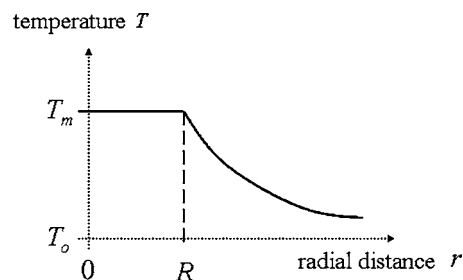
with the general solution



FIG. 2. Schematic diagram of the quasistatic Laplacian temperature profile for three-dimensional radial growth. At $r \leq R$, the sphere is heated up to the equilibrium temperature $T = T_m$. For $r > R$ outside the sphere, the temperature decays towards $T = T_o$ for increasing radial distance $r$.

$$T = K_o + \frac{K_1}{r}. \tag{5}$$

The constants $K_o$ and $K_1$ are determined by the following boundary conditions:

$$T = T_m \quad \text{at } r = R \tag{6}$$

and

$$T = T_o \quad \text{at } r \rightarrow \infty. \tag{7}$$

This gives

$$T = T_o + \frac{(T_m - T_o)}{r/R} \quad \text{for } R \leq r \leq \infty \tag{8}$$

and

$$\left. \frac{\partial T}{\partial r} \right|_{r=R_+} = - \frac{(T_m - T_o)}{R}, \tag{9}$$

where the subscript $r = R_+$ in Eq. (9) indicates that the temperature gradient is just outside the sphere, i.e., in the metastable medium. A schematic diagram of the temperature profile given by Eq. (8) is shown in Fig. 2. Note that the boundary condition of Eq. (6) is a local equilibrium approximation for sufficiently slow growth. Inside the sphere, the temperature distribution is uniform at $T = T_m$. The average temperature gradient at $r = R$ is therefore

$$\left. \frac{\partial T}{\partial r} \right|_{r=R} = - \frac{(T_m - T_o)}{2R} \sim \frac{1}{R}. \tag{10}$$

On assuming a heat-balance condition, i.e., the rate of heat production equals that of heat removal, the heat flux at the moving phase boundary is given by Fourier's law

$$J = \rho \ell \frac{dR}{dt} = \left. -\lambda \frac{\partial T}{\partial r} \right|_{r=R+} = \left. -2\lambda \frac{\partial T}{\partial r} \right|_{r=R}, \tag{11}$$

where $\lambda$ is the thermal conductivity, $\rho$ the density, and $\ell$ the latent heat, which are all assumed to be constant. Combining Eqs. (9) and (11) leads to

$$\frac{dR}{dt} = \frac{\lambda}{\rho\ell}\frac{(T_m - T_o)}{R},$$ (12)

which, upon integration, implies $R \sim t^{1/2}$.

Several imperfections of this model can be noticed. One of them is concerned with energy conservation. The total amount of transition enthalpy generated by the growing sphere is

$$E_{\text{total}} = \rho\ell\frac{4}{3}\pi R^3.$$ (13)

On the other hand, the Laplacian temperature distribution is

$$T = T_o + (T_m - T_o)\frac{R}{r},$$ (14)

which corresponds to an infinite amount of transition enthalpy

$$E_{\text{out}} = \rho c_p \int_R^\infty (T - T_o)r^2 dr \int_0^\pi \sin\theta d\theta \int_0^{2\pi} d\varphi$$

$$= 4\pi\rho c_p(T_m - T_o)R\int_R^\infty r dr \rightarrow \infty$$ (15)

outside the sphere ($c_p$ is the isobaric specific heat capacity). This implies $E_{\text{total}} \rightarrow \infty$, which contradicts Eq. (13) and violates the principle of energy conservation.

Another problem is that boundary conditions cannot be applied at $r \rightarrow \infty$ for lower-dimensional radial growth. For two- and one-dimensional systems, the radially symmetric Laplace equations are

$$\nabla^2 T = \frac{\partial^2 T}{\partial r^2} + \frac{1}{r}\frac{\partial T}{\partial r} = 0 \quad \text{(two-dimensional growth)}$$ (16)

and

$$\nabla^2 T = \frac{\partial^2 T}{\partial r^2} = 0 \quad \text{(one-dimensional growth)},$$ (17)

with the general solutions

$$T = K_o + K_1 \ln(r) \quad \text{(two-dimensional growth)}$$ (18)

and

$$T = K_o + K_1 r \quad \text{(one-dimensional growth)},$$ (19)

respectively. In these solutions, one cannot assume any boundary condition at $r \rightarrow \infty$, since $r \rightarrow \infty$ leads to $T \rightarrow \infty$.

A third concern is the local equilibrium approximation, $T = T_m$ at $r = R$. It is not clear why a finite growth rate $dR/dt$ can still be observed if the phases are approximately in equilibrium. Nevertheless, this model has captured the essential physics: At shallow temperature quenches, the growth rate is limited by an interfacial temperature gradient that scales with $1/R$.

### III. KINETIC DIFFUSION EQUATION MODEL

Another model, which can be viewed as "kinetic" in contrast to "quasistatic," adopts the full thermal diffusion equation and introduces the condition of "interfacial undercooling" at $r = R$ for radial growth in a generally $d$-dimensional system [16]. Advantages are that energy is automatically conserved and, unlike the Laplace equation, there is no problem of assigning boundary conditions for two- and one-dimensional systems. In this model, the interfacial boundary temperature at $r = R$ is $T = T_R < T_m$, so that the growth process occurs away from equilibrium. This is apparently different from the interfacial boundary condition assumed in the quasistatic Laplace equation model. The growth rate is then assumed to be increasing with the deviation from thermodynamic equilibrium at $r = R$, which increases with the interfacial undercooling $T_m - T_R$. Therefore it is

$$\frac{dR}{dt} = \gamma(T_m - T_R),$$ (20)

where a constant kinetic coefficient $\gamma$ is assumed for a first-order Taylor approximation. Because the moving phase boundary at $r = R$ generates transition enthalpy continuously, there is an additional source term $H$ in the diffusion equation for $r = R$:

$$\frac{dT_R}{dt} = \kappa\nabla^2 T|_{r=R} + H.$$ (21)

It can be shown that $H$ is proportional to the growth rate $dR/dt$ and thus depends on the interfacial undercooling $T_m - T_R$: For an infinitesimal growth step $dR$, the amount of enthalpy generated is

$$dH = \rho\ell A dR.$$ (22)

For three- and two-dimensional systems, $A$ is the area and length, respectively, of the interface between the phases. For one-dimensional systems, $A$ is dimensionless and equal to unity. If this phase boundary has a thickness of $\delta R \ll R$, it is

$$dH = \rho A \delta R c_p dT,$$ (23)

where $dT$ is the corresponding temperature rise at the interface $r = R$. Combining Eqs. (22) and (23) gives the source term in the diffusion equation as

$$H \equiv \frac{dT}{dt} = \frac{\ell}{\delta R c_p}\frac{dR}{dt}.$$ (24)

Substituting the model assumption of Eq. (20) into Eq. (24) gives

$$H = \beta(T_m - T_R),$$ (25)

where

$$\frac{\beta\delta R}{\gamma} = \frac{\ell}{c_p}.$$ (26)

### IV. ANALYTIC DERIVATIONS OF GROWTH LAWS

For the limiting case of $\kappa \rightarrow 0$, both the $R \sim t$ and $R \sim t^{1/2}$ growth laws can be derived analytically from the kinetic model by assuming that the growth process occurs at small but finite, identical spatial steps $\delta R$. At $\kappa \rightarrow 0$, Eqs. (20), (21), and (25) give
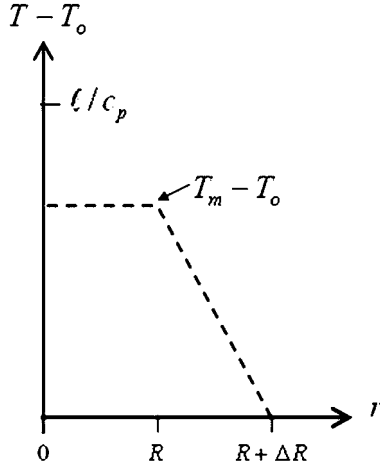
FIG. 3. Trapezoidal approximation of the temperature profile in the limit of zero thermal diffusivity for the case of a shallow temperature quench.

$$\frac{dT_R}{dt} \approx \beta(T_m - T_R) = \frac{\beta}{\gamma}\frac{dR}{dt}. \quad (27)$$

Upon integrating for a discrete growth step $\delta R$, one obtains the change in $T_R$ necessary for the completion of this growth step:

$$\delta T_R \approx \frac{\beta \delta R}{\gamma} = \frac{\ell}{c_p}, \quad (28)$$

where the relation with $\ell/c_p$ is given by Eq. (26).

In the case of *exactly* no thermal diffusion, growth of a step $\delta R$ cannot occur for $T_m - T_o < \ell/c_p$ because the maximum allowed change in $T_R$ is $T_m - T_o$, smaller than $\delta T_R \approx \ell/c_p$ as required [Eq. (28)]. In other words, the stepwise growth is

$$\delta R' = \frac{\gamma}{\beta}(T_m - T_o) < \delta R. \quad (29)$$

For $T_m - T_o > \ell/c_p$, each growth step $\delta R$ takes the same time interval

$$\delta t = \frac{1}{\beta} \ln\left[\frac{T_m - T_o}{(T_m - T_o) - (\ell/c_p)}\right] \quad (30)$$

to increase $T_R$ from $T_o$ to $T_o + \ell/c_p$, as obtained from the integration of Eq. (27). As a result, the growth rate is constant at

$$\frac{\delta R}{\delta t} = \beta / \ln\left[\frac{T_m - T_o}{(T_m - T_o) - (\ell/c_p)}\right], \quad (31)$$

and the growth is linear with $R \sim t$.

If *extremely slow* thermal diffusion is allowed for growth to occur at $T_m - T_o < \ell/c_p$, the temperature profile can be approximated as a trapezoid (Fig. 3) because the spatial spread of the generated transition enthalpy should be as small as possible. It then follows that the temperature gradient outside the growing germ is approximately uniform and everywhere inside the germ is heated up close to the equilibrium tem-

perature $T_m$. The temperature distribution outside the growing germ can therefore be written as

$$T(r) = T_m - \frac{(T_m - T_o)}{\Delta R}(r - R). \quad (32)$$

For three-dimensional growth, the conservation of energy implies

$$\rho c_p\left[\frac{\ell}{c_p} - (T_m - T_o)\right]\frac{4}{3}\pi R^3$$
$$= \rho c_p \int_R^{R+\Delta R}(T - T_o)r^2\,\delta r \int_0^\pi \sin\theta\,\delta\theta \int_0^{2\pi} \delta\varphi \quad (33)$$

for $\delta r = \delta R$ and $R, \Delta R \gg \delta R$. The left-hand side of the equation corresponds to the excess enthalpy $\ell - c_p(T_m - T_o)$ generated, while the right-hand side is the total enthalpy outside the germ. By writing Eq. (32) as

$$T - T_o = (T_m - T_o)\left[\left(1 + \frac{R}{\Delta R}\right) - \frac{r}{\Delta R}\right] \quad (34)$$

and substituting into the right-hand side of Eq. (33) for integration, one obtains

$$\left[\frac{\ell}{c_p(T_m - T_o)} - 1\right]\frac{R^3}{3} = \frac{1}{3}\left(1 + \frac{R}{\Delta R}\right)[(R+\Delta R)^3 - R^3]$$
$$- \frac{1}{4\Delta R}[(R+\Delta R)^4 - R^4], \quad (35)$$

where further rearrangement of terms gives

$$\frac{\ell}{c_p(T_m - T_o)} - 1 = \left(1 + \frac{R}{\Delta R}\right)\left[\left(1 + \frac{\Delta R}{R}\right)^3 - 1\right]$$
$$- \frac{3R}{4\Delta R}\left[\left(1 + \frac{\Delta R}{R}\right)^4 - 1\right]. \quad (36)$$

For $\Delta R \ll R$, the second- and high-power terms of $\Delta R/R$ can be neglected so that

$$\Delta R \approx \left[\frac{\ell}{c_p(T_m - T_o)} - 1\right]\frac{R'}{3}. \quad (37)$$

where $R' \equiv 2R$ Similar derivations for two- and one-dimensional systems lead to the following general result for a $d$-dimensional system with radial symmetry:

$$\Delta R \approx \left[\frac{\ell}{c_p(T_m - T_o)} - 1\right]\frac{R'}{d}, \quad (38)$$

which implies that the width of the temperature profile scales with the linear size $R$ of the growing germ.

From Eq. (38), the temperature gradient at $R < r < R + \Delta R$ is given by

$$\left.\frac{\partial T}{\partial r}\right|_{r=R+} \approx -\frac{T_m - T_o}{\Delta R} = -\frac{(T_m - T_o)d}{\left[\frac{\ell}{c_p(T_m - T_o)} - 1\right]}\frac{1}{R'} \quad (39)$$

for $d$-dimensional radial growth. For $0 < r < R$, the temperature gradient is negligible, due to the flat temperature profile

of $T \approx T_m$. It follows that, at $r=R$, the average temperature gradient and the spatial derivative of this gradient are given by

$$\left.\frac{\partial T}{\partial r}\right|_{r=R} \approx \frac{1}{2}\left.\frac{\partial T}{\partial r}\right|_{r=R+} \approx -\frac{1}{2}\frac{(T_m-T_o)d}{\left[\dfrac{\ell}{c_p(T_m-T_o)}-1\right]}\frac{1}{R'}, \tag{40}$$

and

$$\left.\frac{\partial^2 T}{\partial r^2}\right|_{r=R} \approx -\frac{1}{\delta R}\frac{(T_m-T_o)d}{\left[\dfrac{\ell}{c_p(T_m-T_o)}-1\right]}\frac{1}{R'}, \tag{41}$$

respectively. For $d$-dimensional radial growth, the Laplacian at $r=R$ is

$$\nabla^2 T|_{r=R} \approx \left.\frac{\partial^2 T}{\partial r^2}\right|_{r=R} + \frac{d-1}{R}\left.\frac{\partial T}{\partial r}\right|_{r=R}$$

$$\approx -\left[\frac{1}{\delta R}+\frac{(d-1)}{R'}\right]\frac{(T_m-T_o)d}{\left[\dfrac{\ell}{c_p(T_m-T_o)}-1\right]}\frac{1}{R'}. \tag{42}$$

For sufficiently large values of $R$, only the leading term of order $1/R$ has to be considered so that the Laplacian can be simplified to

$$\nabla^2 T|_{r=R} \approx -\frac{1}{\delta R}\frac{(T_m-T_o)d}{\left[\dfrac{\ell}{c_p(T_m-T_o)}-1\right]}\frac{1}{R'}. \tag{43}$$

That is, at $r=R$, both the temperature gradient and the Laplacian scale with $1/R$, according to Eqs. (39), (40), and (43).

Combining Eqs. (20), (21), and (25) for a finite diffusivity $\kappa$ gives

$$\frac{dR}{dt} = -\frac{\kappa\gamma}{\beta}\nabla^2 T|_{r=R} + \frac{\gamma}{\beta}\frac{dT_R}{dt}. \tag{44}$$

For a growth step of $\delta R$ that takes a time interval of $\delta t$ to complete, Eq. (44) implies

$$\frac{\delta R}{\delta t} = -\frac{\kappa\gamma}{\beta}\frac{\int \nabla^2 T|_{r=R}dt}{\delta t} + \frac{\gamma}{\beta}\frac{\delta T_R}{\delta t}. \tag{45}$$

Substituting Eq. (43) for the Laplacian of temperature $T$ into Eq. (45) gives

$$\frac{\delta R}{\delta t} \approx \frac{\kappa\gamma}{\beta\delta R}\frac{(T_m-T_o)d}{\left[\dfrac{\ell}{c_p(T_m-T_o)}-1\right]}\frac{1}{R'} + \frac{\gamma}{\beta}\frac{\delta T_R}{\delta t}. \tag{46}$$

For each growth step $\delta R$, the interfacial boundary temperature at $r=R$ is initially at $T_R=T_{Ri}$ and becomes $T_R \approx T_m$ after completion of the growth step. The initial value of $T_R$ is given by

$$T_{Ri} \approx T_m + \left.\frac{\partial T}{\partial r}\right|_{r=R}\delta R \tag{47}$$

for a first-order Taylor approximation, so that the change in $T_R$ for one growth step is

$$\delta T_R \approx T_m - T_{Ri} \approx -\left.\frac{\partial T}{\partial r}\right|_{r=R}\delta R. \tag{48}$$

This leads to

$$\frac{\delta T_R}{\delta t} \approx \frac{1}{2}\frac{(T_m-T_o)d}{\left[\dfrac{\ell}{c_p(T_m-T_o)}-1\right]}\frac{1}{R'}\frac{\delta R}{\delta t} \tag{49}$$

according to Eq. (40). Note that Eqs. (46) and (49) are a pair of equations for the two unknowns $\delta R/\delta t$ and $\delta T_R/\delta t$, which are therefore analytically solvable. By substituting Eq. (46) into Eq. (49), the following expression for $\delta T_R/\delta t$ is obtained:

$$\frac{\delta T_R}{\delta t} \approx \left\{1 - \frac{\gamma}{2\beta}\frac{(T_m-T_o)d}{\left[\dfrac{\ell}{c_p(T_m-T_o)}-1\right]}\frac{1}{R'}\right\}^{-1}$$

$$\times \frac{\kappa\gamma}{2\delta R\beta}\frac{(T_m-T_o)^2 d^2}{\left[\dfrac{\ell}{c_p(T_m-T_o)}-1\right]^2}\frac{1}{R'^2}$$

$$\approx \left\{1 + \frac{\gamma}{2\beta}\frac{(T_m-T_o)d}{\left[\dfrac{\ell}{c_p(T_m-T_o)}-1\right]}\frac{1}{R'}\right\}$$

$$\times \frac{\kappa\gamma}{2\delta R\beta}\frac{(T_m-T_o)^2 d^2}{\left[\dfrac{\ell}{c_p(T_m-T_o)}-1\right]^2}\frac{1}{R'^2}. \tag{50}$$

By only considering the leading term of order $1/R^2$, Eq. (50) reduces to

$$\frac{\delta T_R}{\delta t} \approx \frac{\kappa\gamma}{2\delta R\beta}\frac{(T_m-T_o)^2 d^2}{\left[\dfrac{\ell}{c_p(T_m-T_o)}-1\right]^2}\frac{1}{R'^2}; \tag{51}$$

but since $\delta T_R/\delta t$ is of the order of $1/R^2$, it can be neglected in Eq. (46) so that

$$\frac{\delta R}{\delta t} \approx \frac{\kappa\gamma}{\beta\delta R}\frac{(T_m-T_o)d}{\left[\dfrac{\ell}{c_p(T_m-T_o)}-1\right]}\frac{1}{R'}, \tag{52}$$

or equivalently,

$$\frac{\delta R}{\delta t} \approx -\frac{2\lambda}{\rho\ell}\left.\frac{\partial T}{\partial r}\right|_{r=R} = -\frac{\lambda}{\rho\ell}\left.\frac{\partial T}{\partial r}\right|_{r=R+}, \tag{53}$$

according to the definition of thermal diffusivity $\kappa \equiv \lambda/(\rho c_p)$, as well as Eqs. (26), (39), and (40). Note that Eq. (53) is just the heat-balance condition for diffusion-limited growth, imposed in the quasistatic Laplace equation model

[Eq. (11)]. Thus it can be concluded that the apparent difference between the assumed interfacial boundary conditions of the kinetic and the quasistatic model does not exist.

Since the growth process occurs at finite, identical growth steps $\delta R$, the value of $R$ in the $i$th growth step is given by

$$R_i = i \, \delta R. \tag{54}$$

Substituting this expression into Eq. (52) and using the relation in Eq. (26), one can write the following expression for the $i$th time step $\delta t_i$:

$$\delta t_i \approx \frac{2\ell(\delta R)^2}{\kappa c_p(T_m - T_o)d}\left[\frac{\ell}{c_p(T_m - T_o)} - 1\right]i. \tag{55}$$

In general, for the $m$th growth step where $m \gg 1$, it is $R = m\delta R$ following Eq. (54). With Eq. (55), the absolute time $t$ is given by

$$t = \sum_{i=1}^{m} \delta t_i \approx \frac{\ell(\delta R)^2}{\kappa c_p(T_m - T_o)d}\left[\frac{\ell}{c_p(T_m - T_o)} - 1\right]m^2$$

$$\approx \frac{(\ell/c_p)}{\kappa(T_m - T_o)d}\left[\frac{\ell}{c_p(T_m - T_o)} - 1\right]R^2, \tag{56}$$

where the approximation $m(m+1)/2 \approx m^2/2$ is used for

$m \gg 1$. Equation (56) implies $t \sim R^2$, or equivalently the $R \sim t^{1/2}$ growth law for the case of a shallow temperature quench, as observed in various experiments [5–10].

## V. CONCLUSIONS

In this study, the kinetic thermal diffusion equation model and the quasistatic Laplace equation model for the growth dynamics of pure supercooled materials were discussed and analyzed in detail. The $R \sim t^{1/2}$ and $R \sim t$ growth laws for the cases of shallow and deep temperature quenches, respectively, were derived analytically from the kinetic model in the limit of zero thermal diffusivity. For the case of a shallow temperature quench, it was shown that the apparent difference between the assumed interfacial boundary conditions of the quasistatic and the kinetic model does not exist.

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