Migration of a bubble in front of a directionally solidified interface

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(Received 24 August 2006; published 30 April 2007)

We examine the quasistatic thermocapillary migration of a gas bubble normal to a solidified interface. The analysis accounts for the deformation of the solid-liquid interface caused by the bubble's presence. An expression for the distance between the bubble surface and the crystal-melt interface is derived, and used to quantify the dependence of the bubble's migration velocity on the processing variables and material properties of the directional solidification process.

DOI: 10.1103/PhysRevE.75.042602 PACS number(s): 81.10.−h, 64.70.Dv, 44.10.+i, 68.08.−p

I. INTRODUCTION

A gas bubble will migrate in the direction of decreasing interfacial tension, which is usually in the direction of increasing temperature, when it is immersed in an unbounded fluid medium that is subjected to a temperature gradient $G=|dT/dz|$. This thermocapillary phenomenon was put forth by Young *et al.* [1] and tested experimentally in a microgravity environment by Thompson *et al.* [2]. For a nondeformable gas bubble, the thermal conductance and viscosity coefficients of which are assumed nil, the expression for the migration velocity in a weightless setting is given by

$$u_\infty = \frac{aG\sigma_T}{2\mu}, \tag{1}$$

where a is the bubble's radius, μ is the fluid's dynamic viscosity, and $\sigma_T=-\partial\gamma/\partial T$ represents the dependence of the surface tension coefficient, $\gamma>0$, on the temperature field T. Meyyappan *et al.* [3] have conducted a theoretical study of bubble migration in a semi-infinite fluid domain in order to isolate the boundary effects. They have demonstrated that if the bubble, the motion of which is normal to a planar surface, is at three or more bubbles radii from the bounding wall, then it migrates as if it is isolated. To quantify the role of the boundary, they introduce an interaction parameter Ω, which represents the ratio of the migration velocity of an isolated bubble, u_∞, to that of the same bubble migrating normal to the planar surface. They isolate the dependence of Ω on the bubble-wall separation. They find that the wall effect becomes significant only when the bubble is in close contact with the bounding wall, with the migration velocity decreasing as the bubble-wall separation diminishes.

A frequently encountered problem in materials processing from the melt is the formation of air bubbles and their subsequent interaction with an advancing crystal-melt interface. These bubbles form due to the presence of dissolved air in the melt. The difference in the air solubility in the liquid and solid phases causes the bubbles to be rejected by the solidified interface. The bubbles then detach and recede from the interface. It is well known that the presence of a bubble in the melt ahead of a solidifying interface leads to the onset of local interfacial deformations [4–7]. The interfacial deflection is due to the modification of the thermal gradient in the melt between the solid front and the bubble. Owing to the insulating character of the bubble, the thermal gradient in the gap region is increased. If we assume that the crystal-melt interface has zero thickness and that its profile coincides with the liquid's melting point isotherm T_m, then the increase in the thermal gradient is associated with the bulging out of that part of the crystal-melt interface that is behind the bubble [7]. Therefore, the analysis of the thermocapillary migration of a bubble in a melt that is undergoing directional solidification becomes difficult due to both the movement and deformation of the crystal-melt interface, the profile of which is also an unknown to be determined. The techniques of bispherical coordinates or the method of images, which have been successful in tackling analytically the problems involving planar walls, seem to be of little help in this situation.

In this Brief Report, we follow closely the studies by Young *et al.* [1] and Meyyappan *et al.* [3] to quantify the effect of solidification and crystal surface deformation on the bubble's thermocapillary migration velocity. These studies consider the limits of vanishing Marangoni and Reynolds numbers, so that only heat transfer by conduction is accounted for both in the liquid phase and in the gas inside the bubble. The velocity field satisfies the creeping flow equations. The only coupling between the temperature and velocity fields takes place through the balance of the shear stresses at the bubble's surface. This balance equates the shear stress to the change of surface tension caused by variations of the temperature at the bubble's surface. The surface tension is assumed to vary linearly with temperature. Furthermore, we assume that the time scale due to the interface growth rate a/V, where V is the solid front's velocity, is large in comparison to the scale of the melt motion, a/u_∞. Thus, the motion of the crystal-melt interface is neglected in comparison with that of the bubble and, consequently, so is the induced shrinkage flow.

II. GAP THICKNESS

Suppose that an axisymmetric bubble of radius a is located in front of a crystal-melt interface that has emerged from the directional solidification of a pure substance. The bubble is assumed to be nondeformable and of noncolloidal size. We suppose that the bubble's center is at some distance $H_0=(a+h_\infty)$ from the planar portion of the crystal-melt inter-

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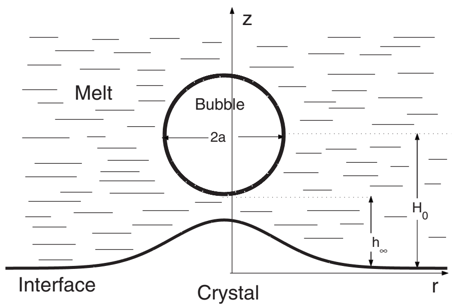

FIG. 1. Sketch of an axisymmetric bubble of radius a in front of a deformable crystal-melt interface (solid line). The portion of the solidified interface behind the bubble has bulged out into the melt. The distance from the bubble's lowest point to the planar part of the crystal surface is denoted by h_∞, i.e., the bubble-crystal front separation is equal to h_∞ if the crystal-melt interface is a nondeformable rigid wall.

face, with h_∞ being the distance from the bubble's lowest point to the planar crystal interface; $0 < h_\infty < a$ but not small enough that the intermolecular forces become active. Then the position of the bubble's surface is described by

$$(z - H_0)^2 + r^2 = a^2, \tag{2}$$

where r and z are the radial and vertical coordinates, respectively. The physical situation is depicted schematically in Fig. 1. The crystal-melt interface is assumed to be moving in the positive vertical direction with velocity V, while the bubble will migrate with velocity V_m upon its detachment from the interface. On assuming that $V \ll V_m$, and that the resulting Marangoni number $aV_m/D \ll 1$, with D being the melt's thermal diffusion coefficient, the convective effects in the conservation of energy equations can be neglected, and, to leading order in the Marangoni number, the thermal fields are described solely by the heat diffusion equations for the temperatures $T(r,z)$, $T_s(r,z)$, and $T_b(r,z)$ in the melt, crystal, and bubble, respectively. These equations are then solved subject to boundary and far field conditions. They consist of the continuity of temperatures and of the heat flux at the bubble's surface, namely,

$$T = T_b \quad \text{and} \quad \frac{\partial T}{\partial r} = 0. \tag{3}$$

Far away from the solid front, the thermal gradient satisfies $\partial T/\partial z \to G$ as $z \to \infty$. Following Ref. [8], if we suppose that the pure substance solidifies with a sharp interface, i.e., an interface of zero thickness, then the profile of the crystal-melt interface conforms to the isotherm corresponding to the thermodynamic melting point T_m, or in case of a deformed isotherm to the equilibrium interface temperature T_{eq}, and separates the solid phase ($T < T_{eq}$) from the liquid phase ($T > T_{eq}$). Far away from the particle, $(r^2+z^2) \to \infty$, the isotherms are not disturbed by the bubble and remain horizontal. (Note that the profile of the crystal's surface is also planar in the absence of bubble.) For mathematical convenience, we suppose that the T_m isotherm coincides with the r axis, i.e., $z=0$ as $r \to \infty$,

$$\lim_{r\to\infty} T(r,0) = T_m. \tag{4}$$

The temperature field in the liquid phase is then given by [9]

$$T(r,z) = T_m + Gz + \frac{Ga^3(z - H_0)}{2[(z - H_0)^2 + r^2]^{3/2}}. \tag{5}$$

Upon evaluating Eq. (5) at $z=0$ we find

$$T_m = T(r,0) + \Delta T_B, \tag{6}$$

where

$$\Delta T_B = \frac{Ga^3 H_0}{2(H_0^2 + r^2)^{3/2}}. \tag{7}$$

Equations (6) and (7) reveal the following. If the bubble were absent then the crystal-melt interface, the shape of which conforms to the planar T_m isotherm, coincides with the r axis. With the presence of the bubble in front of the crystal-melt interface, however, Eq. (6) can be interpreted as an equation for the crystal-melt interface temperature, $T(r,0)$, that has been modified by the amount ΔT_B. Therefore, the bubble's effect is akin to a bubble-induced undercooling. The interface temperature is reduced by an amount ΔT_B from the melting point and consequently the interface deforms by bulging out. The modification of the interfacial temperature and associated interface distortions from the planar morphology are related. Indeed, in the absence of kinetic undercooling, the change of the interface temperature from the melting point that invariably accompanies a curved interface is given by the Gibbs-Thomson equation [10],

$$T(r,0) = T_m - \frac{T_m \sigma_{sl}}{L}\mathcal{K}, \tag{8}$$

where σ_{sl} is the solid-liquid surface energy, L is the latent heat of fusion per unit volume, and $\mathcal{K}$ is the interface mean curvature taken here to be positive when the center of curvature lies in the solid side of the crystal-melt interface. Equations (5)–(8) then yield an expression for the interface curvature, namely,

$$\mathcal{K} = \frac{Ga^3 L H_0}{2T_m \sigma_{sl}(H_0^2 + r^2)^{3/2}}. \tag{9}$$

Note that Eq. (8) neglects the undercooling term due to the hydrodynamic pressure in the gap $g(r)$, namely, $\Delta T_{HP} = -(6\mu V_m T_m/L)\int_0^a g^{-3}(r) r\, dr \ll \Delta T_B$, and we also neglect the change in the thermal conductivity of the melt upon solidification. By considering small interfacial distortions, i.e., $|d\eta/dr| \ll 1$, the shape of the crystal-melt interface profile $\eta(r)$ is described by the differential equation

$$-\mathcal{K} \approx \frac{d^2\eta}{dr^2} = -\frac{LGa^3 H_0}{2T_m \sigma_{sl}(H_0{}^2 + r^2)^{3/2}}. \tag{10}$$

The integration of Eq. (10) subject to the requirement that the profile is symmetric, i.e., $d\eta/dr(0)=0$, yields

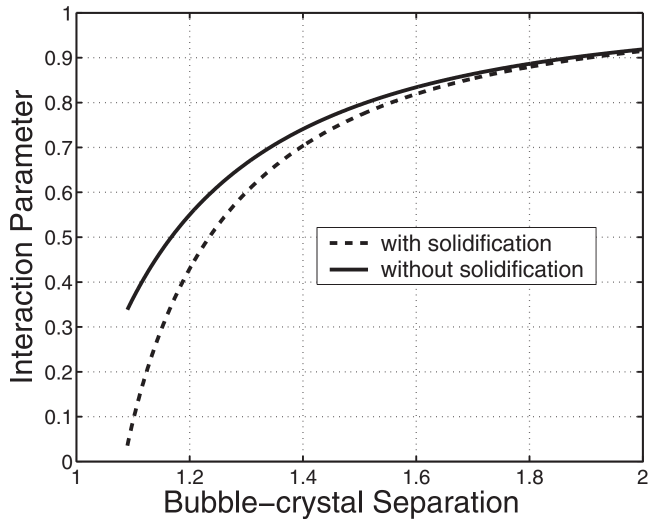

FIG. 2. Plot of the dimensionless interaction parameter Ω [Eq. (14)] versus the dimensionless bubble-crystal surface separation S for the two cases (a) without solidification, i.e., planar rigid wall (solid line) and (b) with solidification, i.e., deformable and rigid crystal-melt interface (dashed line).

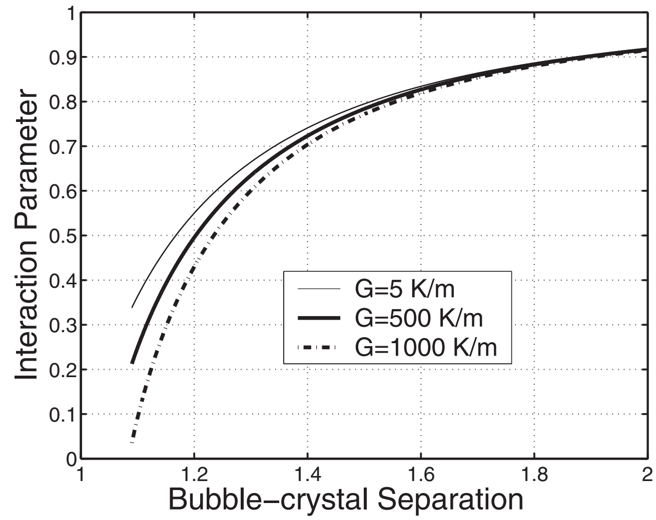

FIG. 3. Plot of the dimensionless interaction parameter Ω [Eq. (14)] as a function of the dimensionless bubble-crystal surface separation S for three distinct values for the imposed thermal gradient G=5 (thin solid line), G=500 (thick solid line), and 1000 K/m (dash-dotted line) for a bubbles radius a=10 μm.

$$\eta(r) = -\frac{LGa^3}{2T_m\sigma_{sl}H_0}\sqrt{H_0{}^2 + r^2} + C. \tag{11}$$

The constant of integration, C, is determined by using the fact that the crystal-melt interface decays to the planar position, $\eta=0$, outside the lubrication region. For instance, if we impose that $\eta \to 0$ as $r \to a$ then we have

$$\eta(r) = -\frac{LGa^3}{2T_m\sigma_{sl}H_0}\left(\sqrt{H_0{}^2 + r^2} - \sqrt{H_0{}^2 + a^2}\right), \tag{12}$$

and it follows that the interface deformation modifies the width of the gap region at the origin from h_∞ to

$$g_0 = h_\infty - \eta(0) = h_\infty - \frac{LGa^3[\sqrt{(a+h_\infty)^2 + a^2} - (a+h_\infty)]}{2T_m\sigma_{sl}(a+h_\infty)}, \tag{13}$$

with the understanding that the validity of Eq. (13) is limited to those parameters for which g_0 is a positive quantity and $g_0 > 100$ Å so that the intermolecular forces are not active [4].

III. MIGRATION VELOCITY

The results reported in [3] are utilized to evaluate the influence of the solidification process on the quasistatic thermocapillary migration of a gas bubble normal to a deformable crystal-melt interface. As shown in the previous section, the interaction between the solidified interface and the gas bubble leads to a decrease in the width of the gap that separates them. The calculations carried out in [3] are repeated here with their bubble-wall separation H replaced by the separation distance between the bubble and the crystal-melt interface, $S=(a+g_0)/a$. Thus, as the bubble approaches the crystal-melt interface, instead of interacting with a planar wall at a distance h_∞, it interacts with a solidified interface whose distance from the bubble is g_0 given by Eq. (13). Thus, when $g_0=h_\infty$ the solidification effect is assumed absent and we retrieve the case of bubble-wall interaction, i.e., $H=S$. Following the calculations in [3] the interaction parameter

$$\Omega(g_0) = \frac{u}{u_\infty}, \tag{14}$$

where u is the thermocapillary migration velocity of the bubble, is evaluated numerically using Eqs. (26)–(28) from Ref. [3]. We refer the reader to Ref. [3] for more details as these equations are too lengthy to reproduce here. These equations describe the approach of a nondeformable bubble normal to a rigid and planar wall in the limit of vanishing Reynolds and Marangoni numbers. The thermal field satisfies the heat conduction equation with continuity of temperature and heat flux at the bubble's boundary. The convective effects are negligible. The velocity field satisfies the creeping flow equations with impenetrability condition at the bubble's surface and vanishing velocity at the wall. The balance of tangential stresses at the bubble's surface yields one of the conditions for the velocity. This condition relates the velocity to the temperature via the dependence of the surface tension on temperature. Therefore, the thermal problem is solved independently of the hydrodynamic problem. The velocity field, however, is coupled to the thermal field through the balance of tangential stresses at the bubble's surface. Consequently, the assumptions made here in the setting up of the solidification problem are consistent with those made by Meyyappan *et al.* [3].

Using typical experimental parameters from experiments [11], we have plotted in Fig. 2 the interaction parameter Ω versus the bubble-crystal-melt interface separation S for a bubble of radius $a=10\ \mu\text{m}$, an imposed thermal gradient $G=500\ \text{K/m}$, solid-liquid surface energy $\sigma_{sl}=0.03\ \text{N/m}$, and latent heat of fusion per unit volume $L=4.6\times10^{7}\ \text{J/m}^{3}$. Figure 2 shows that the solidification process and corresponding crystal-melt interface deflection act to decrease the value of the migration velocity; with the largest decrease occurring at the small values of S. For $S\geq 2$ ($g_0\geq a$), however, the effect of the solidified interface is similar to that of a rigid wall. The influence of the processing variables, such as the thermal gradient G and bubble's radius a, is depicted in Fig. 3. As expected, the decrease of g_0 with G implies a decrease in the value of the migration velocity at higher thermal gradients.

In summary, then, the thermocapillary migration velocity of a nondeformable gas bubble normal to a deformable crystal-melt interface is lower than that of the same bubble approaching a rigid planar wall. The lowering of the bubble's velocity is attributed to the bulging out of the portion of the crystal-melt interface behind the bubble and the subsequent decrease in the bubble-crystal-melt interface separation.

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