Shape Function of Directional Solidification Interface under Two-Dimensional Heat Conduction Condition

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Abstract: The interface shape function for the pure material directional solidification system with temperature disturbances on the heater and cooler is derived. It is approximately a fraction function including cosine terms in the denominator and the numerator. Calculation of the function shows the interface shape can respond to the temperature disturbance. When both the solidification rate V and the interface cooler distance α determined by the boundary temperatures are lower than critical value deteimined by a formula, the interface shape changes from sinusoidal wave to figures pattern with the increase of the rate or the distance. Once the rate or the distance reaches the critical value, the interface branches at the bottom of the grooves between figures and then the branches expand along the sidewall of the figures with further increase of the rate or the distance. According to that, we conclude that the sinusoidal interface shape assumption always used by the interface instability analyses is not always valid and the interface shape in Hele-Shaw solidification experiments in fact maybe is not planar but a cellular interface with quite small amplitude, and the role of temperature disturbance should be considered in experiments studying solidification interface stability.

Keyword: Directional solidification; Interface shape function; Temperature disturbance; Two-dimensional heat conduction

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1. Introduction

A large number of experiments have indicated that a solidification interface shows planar, cellular, dendritic, cellular and planar patterns in turn with increase of the solidification rate[1-3]. These patterns bear direct relationship with casting microstructures and determine the properties of a material[4], so the evolution of solidification interface shape has been of tremendous interest for both academic researches and industrial applications. In the past several decades, much work has been done to understand this phenomenon. However, they do not obtain the final knowledge even if an prevail accepted theory of it up to now since it is a complicated three dimensional Stefan problem[5] with obviously nonlinear effects and affected by multi-factors.

Survey the previous work, they almost focused on the solidification interface shape evolution of alloy especially binary alloys. Before the rising of simulation method for example phase filed algorithm, the experimental and theoretical methods were alternately employed. J. W. Rutter maybe firstly discovered cellular pattern during the directional solidification of 99.9860% purity "Chempur" tin melt[6]. This discover then excited the attention of W.A. Tiller who with his co-workers put forward the famous “Constitutional supercooling” theory to explain the emergence of the cellular pattern[7]. However, this theory just considered the effect of the temperature gradient and the solute redistribution on the interface shape but did not realize the role of the surface tension and the perturbation[8]. About ten years later, W. W. Mullins and R. F. Sekerka published a series of papers which introduced the basal theory, always called MS criterion, for analyzing solidification interface stability[9-13]. MS criterion points out that surface tension plays a positive role in stabilization of the planar interface, but negative temperature gradient and solute redistribution play negative roles. The latter has been clarified by the constitutional supercooling theory. The more important is that MS creterion foreshows a neutral instability of a planar interface when the amplitude of perturbation does not change in time that is to say a planar interface is instable just when it is disturbed by a perturbation with wavelength in a middle band. MS theory started the time of analyzing solidification interface instability using perturbation method with which many later scientists prompted the analysis of solidification interface instability to nonlinear models[14-23]. These nonlinear models mainly considered the nonlinear effect of the interface temperature determined by the Gibbs-Thomson relation through different mathematic methods or under different particular conditions. Their results made the neutral instability wavelength band accurater and discovered the nature of the branch and the selection of preferred wave number and pattern, which has been discussed by Davis in detail in his excellent monograph ‘theory of solidification’ [24].

Just like that Kerszberg have said ‘it is not an easy task to solve the basic equations the only way in which the difficult problem of directional solidification can be studied theoretically’[25] since explicit tracking of the free interface by accurately calculating the basic equations is quite difficult[26], the linear and nonlinear instability models rarely depicted a concrete interface shape beside supposed the interface is sinusoidal or sine series shape with infinitesimal amplitude. In many directional solidification experiments, the nearly sinusoidal interfaces were observed when the solidification rate or the temperature gradient was small. When the solidification rate or the temperature gradient was increased, the interface gradually changed into fingered cellular array; further increasing, dendritic structure formed. Moreover, the depth of the grooves increased with the increase of the solidification rate or the temperature gradient[27-30]. These patterns also were seen in the computer simulation through phase field models[31-35].

Phase field models has achieved considerable importance for simulating the temporal evolution of complex interface shapes associated with realistic features of the solidification [36]. However, its solution quality is significantly dependent on the prescribed interface thickness that is required to be sufficiently small for accurate simulations[37]. On the other hand, simulation can not directly show the mathematic relation between the interface shape and the solidification conditions. It is, therefore, yet human hope to accurately predict the evolution of solidification interface shape by solving the basical equations in theory so interpret the observations in experiments and simulation. It is should noted that M. Kerszberg has attempted that work in 1983[25]. He developed a complicated series function of interface shape based on the one-side assumption[14, 23] and numerically depicted the fingered pattern. Then also based on the one-side assumption, L.H Ungar and R.A Brown constructed a cosine series shape function of solidification interface[38], G.B McFadden and S.R Coriell[39] directly numerically simulated the field equations of a directional solidification system. They also predicted the cellular pattern. However like that Kerszberg has realized, the truth maybe drastically different from the results they obtained because the one-side assumption is too limited [25]. Later in 1989, D.A Kessler and H Levine solved the solute diffusion equation at constant solidification rate and fixed interface temperature gradients using Newton's method to solve a discretized version of the integro-differential equation for the solid liquid boundary. They successfully obtained cellular interface and shew the appearance of deep cell grooves with the increase of solidification rate[40]. However obviously, its solving conditions are not the solidification conditions actual solidification courses always meet.

It can be seen from above that even the solidification interface pattern of alloy has been studied on a large scale, it is still far from fully solving this problem. On the other hand, the study, especially experimental study, of directional solidification interface pattern of pure materials is nearly absente. However, in our opinion, the interface pattern and stability of pure materials are very important for understanding the interface pattern and stability of alloy since the heat diffusion is still a controlling mechanism of the solidification of alloy, the solution redistribution effects solidification still by effecting the temperature field, thus, the stability caused by the heat diffusion would still contribute to the total stability of interface of alloy, which could be seen partly from the experiment carried by X.W. Qian *etc,* where a growing crystalline dendrite would response to a brief localized heat pulse near its tip and the induced deformation could grow rapidly form unobserved small to sidebranch feature. S. Agarwal *etc* recently have pointed out that the depths of the phases and the width of the container influence the interface patterns of a pure material directional solidification system and there is a third critical stability point appearance at small disturbance wave number and independent on the surface tension[41], which means the instability of a pure material solidification interface is also very complicate. This paper aimed to explore if there exists a mathematic interface shape function when a pure material directional solidification system is disturbed by temperature perturbation on the finite away boundary wall, which is important for understanding the interface instability. Two reasons inspire us to chose that physical model: (1) The diffusion is nearly two dimensional when the interface is curved during the directional solidification most met in the previous Hele-Shaw experiments and in actual applications; (2) Pure material directional solidification system is also frequently applied in actual producing and maybe an ideal model for studying the solidification interface shape since the solving just needs to consider the heat diffusion the course easiest to be disturbed so arousing the interface instability in actual solidification. We firstly solve the two dimensional heat conduction equations without presupposing the form of the interface function and then analyze the interface shape evolution when the solidification conditions vary through numerical calculating the interface shape function with fractional form we derived.

2. Model description

The directional solidification model is shown in Fig.1, a non-planar solid liquid interface moves toward the liquid at a constant rate V. We supposed that 1) the material is pure; 2) the liquid and solid phases are isotropic and homogenous; 3) the physical characteristics of the liquid and solid phases are constant and independent of temperature; 4) the system is stationary; 5) the pressure is constant and equal to normal atmospheric pressure and 6) the crucible is adiabatic.

It is obvious that the heat will diffuse along X and Z direction because of the non-planar solid liquid interface in this model. We can describe this problem using the classical two-dimensional heat conduction equation in a rest frame fixed on the crucible[42]. Similar to that has been applied to analyze the mass diffusion in directional solidification by W. A. Tiller in the literature [7], if the solid-liquid interface is considered as the origin, and freezing is represented by moving the liquid distribution past it at the rate V, the differential equation (1) can be transformed as Eq.2 under the assumption four in the frame moving with the interface.

$$D\left(\frac{\partial^2 T}{\partial x^2}+\frac{\partial^2 T}{\partial z^2}\right)=\frac{\partial T}{\partial t} \tag{1}$$

$$D\left(\frac{\partial^2 T}{\partial x^2}+\frac{\partial^2 T}{\partial z^2}\right)+V\frac{\partial T}{\partial z}=0 \tag{2}$$

This differential equation can be solved through separation of variables[43]. Let $T(x, z)=X(x)Z(z)$ and take it into Eq.2, we have

$$\frac{D}{Z}\frac{d^2 Z}{dz^2}+\frac{V}{Z}\frac{dZ}{dz}=-\frac{D}{X}\frac{d^2 X}{dx^2} \tag{3}$$

Introduce a separation constant λ, the equations below are satisfied

$$\frac{D}{Z}\frac{d^2 Z}{dz^2}+\frac{V}{Z}\frac{dZ}{dz}=\lambda \tag{4}$$

$$\frac{D}{X}\frac{d^2 X}{dx^2}=-\lambda \tag{5}$$

According to the separation constant λ, Eq.2 has five solutions (see Appendix) and the solution corresponding to $\lambda=0$ and $\lambda>0$ as follow satisfy the boundary condition that the crucible is adiabatic.

$$T_S^0=a_S^0+b_S^0\exp\left(-\frac{V}{D_S}z\right) \qquad \lambda=0 \tag{6}$$

$$T_L^0=a_L^0+b_L^0\exp\left(-\frac{V}{D_L}z\right) \qquad \lambda=0 \tag{7}$$

$$\begin{gathered}T_S^k=c_S^k\left[a_S^k\exp\left(\frac{-V+\sqrt{V^2+4D_S\lambda_S^k}}{2D_S}z\right)+b_S^k\exp\left(\frac{-V-\sqrt{V^2+4D_S\lambda_S^k}}{2D_S}z\right)\right]\cos\left(\sqrt{\frac{\lambda_S^k}{D_S}}x\right)+T_S^{c,k}\\ \lambda_S^k=\left(\frac{k\pi}{h}\right)^2 D_S \quad k=1,2,3\ldots\end{gathered} \tag{8}$$

$$\begin{gathered}T_L^k=c_L^k\left[a_L^k\exp\left(\frac{-V+\sqrt{V^2+4D_L\lambda_L^k}}{2D_L}z\right)+b_L^k\exp\left(\frac{-V-\sqrt{V^2+4D_L\lambda_L^k}}{2D_L}z\right)\right]\cos\left(\sqrt{\frac{\lambda_L^k}{D_L}}x\right)+T_L^{c,k}\\ \lambda_L^k=\left(\frac{k\pi}{h}\right)^2 D_L \quad k=1,2,3\ldots\end{gathered} \tag{9}$$

where a, b, c, d and T^c are undetermined constants, D is thermal diffusivity . The subscript S and L denote the solid phase and the liquid phase. Thus, according to the theory of the solution of differential equations, we can get the solution of Eq.2 as follow, where the same order constants have been combined.

$$\begin{aligned}T_s&=T_S^c+b_S^0\exp\left(-m_S z\right)\\ &+\sum_{k=1}^{\infty}c_S^k\left[a_S^k\exp\left(\frac{-V+\sqrt{V^2+4D_S^2\left(\frac{k\pi}{h}\right)^2}}{2D_S}z\right)+b_S^k\exp\left(\frac{-V-\sqrt{V^2+4D_S^2\left(\frac{k\pi}{h}\right)^2}}{2D_S}z\right)\right]\cos\left(\frac{k\pi}{h}x\right)\end{aligned} \tag{10}$$

$$T_L = T_L^c + b_L^0 \exp\left(-\frac{V}{D_L} z\right) + \sum_{k=1}^{\infty} c_L^k \left[a_L^k \exp\left(\frac{-V + \sqrt{V^2 + 4D_L^2\left(\frac{k\pi}{h}\right)^2}}{2D_L} z \right) + b_L^k \exp\left(\frac{-V - \sqrt{V^2 + 4D_L^2\left(\frac{k\pi}{h}\right)^2}}{2D_L} z \right) \right] \cos\left(\frac{k\pi}{h} x\right) \tag{11}$$

From the past researches, we have known that the first part in Eq.10 and Eq.11 is right the solution of the temperature distribution of a directional solidification system with a planar solid liquid interface[11, 44] and the second part in the summation symbol expresses a disturbance [9, 43]. Adopting the infinite boundary condition that the disturbance is zero introduced in Ref [9], we can obtain

$$T_s = T_S^c + b_S^0 e^{-m_S z} + \sum_{k=1}^{\infty} A_S^k e^{-\omega_S z} \cos(\omega_k x) \tag{12}$$

$$T_L = T_L^c + b_L^0 e^{-m_L z} + \sum_{k=1}^{\infty} B_L^k e^{-\omega_L z} \cos(\omega_k x) \tag{13}$$

Where $T_S^c \equiv a_S^0 + \sum_{k=1}^{\infty} T_S^{c,k}$, $T_L^c \equiv a_L^0 + \sum_{k=1}^{\infty} T_L^{c,k}$, $A_S^k \equiv c_S^k a_S^k$, $B_L^k \equiv c_L^k b_L^k$, $m_S \equiv \frac{V}{D_S}$, $m_L \equiv \frac{V}{D_L}$, $\omega_S \equiv \frac{V - \sqrt{V^2 + 4D_S^2 \omega_k^2}}{2D_S}$, $\omega_L \equiv \frac{V + \sqrt{V^2 + 4D_L^2 \omega_k^2}}{2D_L}$ and $\omega_k = \frac{k\pi}{h}$. This result means that the disturbance parts of the temperature fields of a directional solidification under two dimensional heat diffusion condition are Fourier series. Thus, A_S^k and B_L^k in Eq.11 and Eq.12 can be determined using the Fourier series transformation of boundary conditions. For a actual directional solidification system, the temperature fields are configured by a cooler and a heater located from the interface finite distance. In this study, it is supposed that boundary condition corresponding to the cooler is $z=\alpha(x)$, $T=T_\alpha(x)$ and the boundary condition corresponding to the heater is $z=\beta(x)$, $T=T_\beta(x)$, where $T_\alpha(x)$ and $T_\beta(x)$ are functions which can be transformed as Fourier series. According to the Fourier series transformation[43], we have

$$A_S^k = \frac{1}{h} e^{\omega_S \alpha} \int_{-h}^{h} T_\alpha(\xi) \cos(\omega_k \xi) d\xi \tag{14}$$

$$B_L^k = \frac{1}{h} e^{\omega_L \beta} \int_{-h}^{h} T_\beta(\xi) \cos(\omega_k \xi) d\xi \tag{15}$$

Substitute them into Eq.12 and Eq.13, the temperature field in the solid phase and liquid phase can be obtained as follow.

$$T_S = T_S^c + \left[\frac{1}{h}\int_{-h}^{h} T_\alpha(\xi) d\xi - T_S^c\right] e^{m_S(\alpha - z)} + \sum_{k=1}^{\infty} \frac{1}{h} e^{\omega_S(\alpha - z)} \cos(\omega_k x) \int_{-h}^{h} T_\alpha(\xi) \cos(\omega_k \xi) d\xi \tag{16}$$

$$T_L = T_L^c + \left[\frac{1}{h}\int_{-h}^{h} T_\beta(\xi) d\xi - T_L^c\right] e^{m_L(\beta - z)} + \sum_{k=1}^{\infty} \frac{1}{h} e^{\omega_L(\beta - z)} \cos(\omega_k x) \int_{-h}^{h} T_\beta(\xi) \cos(\omega_k \xi) d\xi \tag{17}$$

At the interface $z=f(x)$, the energy conservation equation $(K_S \nabla T_S - K_L \nabla T_L)\cdot\vec{n} = (\Delta H + 2\kappa\gamma)\vec{V}\cdot\vec{n}$ is satisfied and the temperature is determined as $T_i(x) = T_M\left(1 + \frac{2\kappa\gamma}{\Delta H}\right) - \mu\vec{V}\cdot\vec{n}$ [24], where K_S and K_L are respectively the thermal conductivity coefficients of solid phase and liquid phase, $\vec{n}$ is the normal vector of interface, ΔH is latent heat of solidification, μ is the kinetic undercooled coefficient and κ is the mean curvature of interface,

$\kappa = \frac{1}{2} f_{xx} \left(1+f_x^2\right)^{-\frac{3}{2}}$. Thus we can obtain an equations set and the shape of interface can be obtained through solving it.

$$K_S \left\{ m_S \left[T_S^c - \frac{1}{h}\int_{-h}^{h} T_\alpha(\xi) d\xi \right] e^{m_S(\alpha-f)} - \sum_{k=1}^{\infty} \frac{\omega_S}{h} \cos(\omega_k x) e^{\omega_S(\alpha-f)} \int_{-h}^{h} T_\alpha(\xi)\cos(\omega_k \xi) d\xi + f_x \sum_{k=1}^{\infty} \frac{\omega_k}{h} \sin(\omega_k x) e^{\omega_S(\alpha-f)} \int_{-h}^{h} T_\alpha(\xi)\cos(\omega_k \xi) d\xi \right\} \tag{18.a}$$
$$-K_L \left\{ m_L \left[T_L^c - \frac{1}{h}\int_{-h}^{h} T_\beta(\xi) d\xi \right] e^{m_L(\beta-f)} - \sum_{k=1}^{\infty} \frac{\omega_L}{h} \cos(\omega_k x) e^{\omega_L(\beta-f)} \int_{-h}^{h} T_\beta(\xi)\cos(\omega_k \xi) d\xi + f_x \sum_{k=1}^{\infty} \frac{\omega_k}{h} \sin(\omega_k x) e^{\omega_L(\beta-f)} \int_{-h}^{h} T_\beta(\xi)\cos(\omega_k \xi) d\xi \right\}$$
$$= \left(\Delta H + \gamma \frac{f_{xx}}{\left(1+f_x^2\right)^{-3/2}} \right) V$$

$$T_{\mathrm{M}} \left(1 + \frac{\gamma}{\Delta H} \frac{f_{xx}}{\left(1+f_x^2\right)^{3/2}} \right) - \mu V = T_S^c + \left[\frac{1}{h}\int_{-h}^{h} T_\alpha(\xi) d\xi - T_S^c \right] e^{m_S(\alpha-f)} + \sum_{k=1}^{\infty} \frac{1}{h} e^{\omega_S(\alpha-f)} \cos(\omega_k x) \int_{-h}^{h} T_\alpha(\xi)\cos(\omega_k \xi) d\xi \tag{18.b}$$

$$T_{\mathrm{M}} \left(1 + \frac{\gamma}{\Delta H} \frac{f_{xx}}{\left(1+f_x^2\right)^{3/2}} \right) - \mu V = T_L^c + \left[\frac{1}{h}\int_{-h}^{h} T_\beta(\xi) d\xi - T_L^c \right] e^{m_L(\beta-f)} + \sum_{k=1}^{\infty} \frac{1}{h} e^{\omega_L(\beta-f)} \cos(\omega_k x) \int_{-h}^{h} T_\beta(\xi)\cos(\omega_k \xi) d\xi \tag{18.c}$$

3. Shape under *k*-wave disturbance

3.1 shape function

Let us firstly predict the interface shape when the cooler and heater are planar walls and their temperature is suffered wave disturbances with spatial angle velocity ω_k . This disturbance always appears and be used in theoretical analyses. Corresponding boundary condition is expressed as $z=\alpha$, $T_\alpha(x)=\psi+\delta_k\cos(\omega_k x)$ and $z=\beta$, $T_\beta(x)=\varphi+\zeta_k\cos(\omega_k x)$, where α, β, ψ, φ, δ_k and ζ_k are constant, $\delta_k\approx 0$ and $\zeta_k\approx 0$. Substituting $T_\alpha(x)$ and $T_\beta(x)$ into Eq.16 and Eq.17 we can obtain the corresponding temperature fileds from which the constant T_S^c and T_L^c can be deduced.

$$T_S = T_S^c + \left(\psi - T_S^c\right) e^{m_S(\alpha-z)} + \delta_k e^{\omega_S(\alpha-z)} \cos(\omega_k x) \tag{19}$$

$$T_L = T_L^c + \left(\varphi - T_L^c\right) e^{m_L(\beta-z)} + \zeta_k e^{\omega_L(\beta-z)} \cos(\omega_k x) \tag{20}$$

In a previous paper we have gave out the temperature field of a one-dimensional heat conduction directional solidification system with structure similar to Fig.1 but the cooler and heater are planar and with uniform temperature [44]. Thus, hen the amplitudes of harmonic wave disturbances, δ_k and ζ_k, are converged to zero under which $T_\alpha(x)=\psi$ and $T_\beta(x)=\varphi$, Eq.19 and Eq.20 should be equal to the temperature fields deduced in Ref [44].

$$T_S^c + \left(\psi - T_S^c\right) e^{m_S(\alpha-z)} = \frac{\left(T_{\mathrm{M}} - \psi\right) e^{-m_S z} + \psi - T_{\mathrm{M}} e^{-m_S \alpha}}{1 - e^{-m_S \alpha}} \tag{21}$$

$$T_L^c + \left(\varphi - T_L^c\right) e^{m_L(\beta-z)} = T_{\mathrm{M}} - \left[\frac{D_L}{K_L} \Delta H + \frac{K_S D_L}{K_L D_S} \frac{\left(T_{\mathrm{M}} - \psi\right)}{\left(1 - e^{-m_S \alpha}\right)} \right] \left(1 - e^{-m_L z}\right) \tag{22}$$

so

$$T_S^c = \frac{\psi - T_{\mathrm{M}} e^{-m_S \alpha}}{1 - e^{-m_S \alpha}} \tag{23}$$

$$T_L^c = T_M - \frac{D_L}{K_L} \Delta H - \frac{K_S D_L}{K_L D_S} \frac{T_M - \psi}{1 - e^{-m_S \alpha}} \tag{24}$$

$$\varphi = T_{\mathrm{M}} - \left[\frac{D_L}{K_L} \Delta H + \frac{K_S D_L}{K_L D_S} \frac{T_{\mathrm{M}} - \psi}{1 - e^{-m_S \alpha}} \right] \left(1 - e^{-m_L \beta}\right) \tag{25}$$

According to above equations and considering $T_S=T_L$ on the interface $z=f(x)$, we can get a express of f below from Eq.19 and 20, where we have used an approximation $e^y \approx 1+y$ when $y\approx 0$ for we believe that f is quite small.

$$f(x)=\frac{\left(\delta_k e^{\omega_S\alpha}-\zeta_k e^{\omega_L\beta}\right)\cos(\omega_k x)}{\frac{V}{K_L}\left[\frac{K_L-K_S}{D_S}\frac{T_\mathrm{M}-\psi}{1-e^{-m_s\alpha}}-\Delta H\right]+\left(\delta_k\omega_S e^{\omega_S\alpha}-\zeta_k\omega_L e^{\omega_L\beta}\right)\cos(\omega_k x)} \quad (26)$$

Now we check if Eq.18 can be satisfied when the shape of interface is Eq.26. Firstly we define $\delta_k e^{\omega_S\alpha}-\zeta_k e^{\omega_L\beta}\equiv\Theta_k$, $\delta_k\omega_S e^{\omega_S\alpha}-\zeta_k\omega_L e^{\omega_L\beta}\equiv\Omega_k$ and $\frac{V}{K_L}\left[\frac{K_L-K_S}{D_S}\frac{T_\mathrm{M}-\psi}{1-e^{-m_s\alpha}}-\Delta H\right]\equiv\Pi$, so

$$f(x)=\frac{\Theta_k\cos(\omega_k x)}{\Pi+\Omega_k\cos(\omega_k x)} \quad (27)$$

$$f_x=-\frac{\Pi\Theta_k\omega_k}{\left[\Pi+\Omega_k\cos(\omega_k x)\right]^2}\sin(\omega_k x) \quad (28)$$

$$f_{xx}=-\omega_k^2\frac{\Pi^2}{\left[\Pi+\Omega_k\cos(\omega_k x)\right]^2}\frac{\Theta_k\cos(\omega_k x)}{\Pi+\Omega_k\cos(\omega_k x)}-\omega_k^2\Pi\Theta_k\Omega_k\frac{1+\sin^2(\omega_k x)}{\left[\Pi+\Omega_k\cos(\omega_k x)\right]^3}\approx-\omega_k^2 f \quad (29)$$

Consequently, Eq.18 a) can be rewritten as follow under the approximation $e^y\approx 1+y$ when $y\approx 0$ and keeping the small terms up to the first order. This approximation is reasonable for we have supposed that the solidification system is stationary an assumption always is valid in an actual solidification system where $D_S\gg V$ and $D_L\gg V$ [9].

$$\left(K_S\delta_k\omega_S e^{\omega_S\alpha}-K_L\zeta_k\omega_L e^{\omega_L\beta}\right)\cos(\omega_k x)+\left[K_S m_S^2\frac{\psi-T_\mathrm{M}}{1-e^{-m_s\alpha}}-K_S m_L^2\frac{D_L}{D_S}\frac{\psi-T_\mathrm{M}}{1-e^{-m_s\alpha}}+m_L^2 D_L\Delta H-\omega_k^2\gamma V\right]f=0 \quad (30)$$

This equation could be established when

$$\frac{\zeta_k}{\delta_k}=\frac{K_S\omega_S}{K_L\omega_L}e^{\omega_S\alpha-\omega_L\beta} \quad (31)$$

$$\omega_k^2=\frac{V}{\gamma}\left[\frac{K_S}{D_S}\frac{\psi-T_\mathrm{M}}{1-e^{-m_s\alpha}}\left(\frac{1}{D_S}-\frac{1}{D_L}\right)+\frac{\Delta H}{D_L}\right] \quad (32)$$

Next, we check if Eq.18 b) can be satisfied. According to Eq.23 to Eq.30 and keeping the small terms up to the first order, Eq.18 b) can be approximated into the following equation.

$$\mu=\left\{\frac{T_\mathrm{M}-\psi}{1-e^{-m_S\alpha}}m_S\Theta_k-\Pi\delta_k e^{\omega_S\alpha}-\frac{\gamma}{\Delta H}T_\mathrm{M}\Theta_k{\omega_k}^2\right\}\frac{\Pi^2\cos(\omega_k x)}{\left[\Pi+\Omega_k\cos(\omega_k x)\right]^3 V} \quad (33)$$

Thus if only Eq.33 was established, the Eq. 18 b) can be satisfied. Under this condition, the Eq. 18 c) also can be satisfied since the f is obtain according to a equation group combined the Eq. 19 and the Eq.20 which are corresponding to the Eq.18 b) and c).

3.2 Simulation

During actual directional solidifications，the solidification course is controled by the cooler temperature and the heater temperature, so the solidification rate V is always varing with the interface advacing. Simulation of the interface shape evolution during a actual directional solidification under a k order harmonic temperature disturbance and using parameters ψ=243K, T_M=273K, h=10cm, D_S=200cm^2·s^{-1}, D_L=100cm^2·s^{-1}, K_S=2cal·K^{-1}·s^{-1}·cm^{-1}, K_L=1cal·K^{-1}·s^{-1}·cm^{-1} and ΔH=10000cal·cm^{-3} indicates the interface shape is near undeformed cosine wave when $|\alpha|$ is enough small. When decreasing α, three deforming behaviors gradually appear on the cosine wave: (1) the amplitude gradually increases; (2) the wave was gradually elongated toward the negative direction of z axis since its vales' amplitude increases much faster than its peaks' amplitudes, which cause the formation of deep grooves a characteristic morphology always was observed in the directional solidification experiments in a Hele-Shaw cell[27, 29, 45, 46]. In these experiments, the grooves always are so deep that their bottom can not be observed, this also is shown in Fig.2b where the depth of grooves are obviously larger than $|\alpha|$ so the bottoms would actually not exist when the distance of the average position of interface to the cooler is larger than a critical value; (3) the peaks of the wave gradually widen, contrary, the vales gradually narrow (Fig.2 α=-3cm to α=-12cm). As a result, the interface shape gradually becomes similar to the shape of cellular structure (Fig.2 α=-11.5cm to α=-12cm) [27, 29, 47]. Keep on decreasing α to a critical value, branch occurs, which cause the interface to be split into 7 bunches, one group of bunches is corresponding to the wave peaks and does not shift, another group is corresponding to the wave vales and shift large distance towards the positive direction of z axis (Fig.2c). Further decrease α, the branch point shift towards to the wave peaks bunches so that the wave peaks bunches become narrower and narrower and the width of the vales bunches increase, moreover, the tips of the bunches become blunter and blunter (Fig.2 α=-12.2cm to α=-14cm). When α decreases to an enough large value, the interface degenerates into nearly even.

The above results were obtained under varying interface advancing velocity V , a condition not applicable in many experimental directional solidification systems for detecting the interface instability where the interface advancing velocity V is always artificially kept constant during single experiment through pulling the solidification cell with the demand velocity[48]. Obviously, it is valuable to compare the results obtained through our model with the observations during those experimental directional solidification through they all are for alloys. Fig.3a shows that the amplitude of the interface will also increase with the interface advancing ahead (corresponding to decreasing α) when the interface advancing velocity is fixed. Also deep grooves gradually appear. This is in agreement with the observations in Ref [47] where the cell amplitude gradually increased and deep cell grooves gradually appeared accompanying with the extending of directional solidification time of the succinonitrile acetone alloy or pivalic acid ethanol system at fixed solidification rates. On the other hand, Fig.3a indicates the cell wavelength is decrease with the progress of solidification, this is consistent with the observation in Refs [27, 49] and is the famous wavelength selection problem about which there is a prevail answer that the cell wavelength decreases with the solidification rate[6, 29, 50]. Further study shows that results agree with the answer can be obtained through the Eq.32. We will detaily discuss it in another paper.

Comparing with the condition of Fig.3, we consider a group of Hele-Shaw solidification experiments with increased solidification rates. Fig.4 shows similar interface shape evolution with that shown in Fig.2 when the solidification rates was increased, which means if the interface is controlled to be located at a same position (represented by same α) for the group experiments by decreasing the temperature gradient so make the solidification rate for single experiment is increased one by one, it would be shown that the interface of higher rate experiment bears larger wave amplitude and smaller wavelength as well as smaller tip curvature. Fig.5 shows that surface tension plays an important role in determination the interface shape. The amplitude of the interface wave increases, the wave was gradually elongated toward the negative direction of z axis, the peaks of the wave gradually widen and the vales gradually narrow when minishing surface tension. This is similar to the evolution when decreasing the α.

4. Discussion

The results before have shown that there exists a shape function, even a approximate solution, for its interface when a pure material directional solidification system is disturbed by cosine-like temperature perturbations on the finite away boundary walls. However the function is not a simple sine or cosine function but is a fraction with both numerator and denominator including cosine function (see Eq.26). This function can be approximately simplified as simple cosine function so the interface bears a cosine-like shape when the distance the interface to the cooler α and the solidification rate V are small. With increase of $|\alpha|$ or V, the interface gradually change into finger shape with tip curvature gradually decrease. This finger interface and the cosine like interface here are both called cellular interface. When $|\alpha|$ or V increases to a critical value, the cellular interface would branch. After branch adjacent cells of the interface extend a part of their sides and form two branches which penetrate into melt. In theory, these branches obviously can not survive during a without undercooled solidification but may survive during a undercooled solidification. But unfortunately, as we know, they are never observed in experiments except the sidebranching at the cellular-dentrite transition[46]. Some differences are that the branch here firstly occurs at the bottoms of the grooves but the sidebranching of alloy solidification interface always firstly occurs at the sides of cell tips[46, 50] and that there are only two brunches of every adjacent cells, however there are always many sidebranches so form dentrite during the the sidebranching of alloy solidification interface[46, 50]. Fig.2 indicates $f(x)$ will became very large when the branch occurs, which means the denominator of Eq.26 will near zero when the branch occurs. Considering the Eq.31and the branch firstly occurs at the bottoms of the grooves, thus the critical value for the branch occurring can be obtained by

$$\frac{V}{K_L}\left(\frac{K_L-K_S}{D_S}\frac{T_{\mathrm{M}}-\psi}{1-e^{-m_S\alpha}}-\Delta H\right)+\delta_k\omega_S e^{\omega_S\alpha}\left(1-\frac{K_S}{K_L}\right)=0 \qquad (34)$$

Integrating $m_S\equiv\frac{V}{D_S}$ and $\omega_S\equiv\frac{V-\sqrt{V^2+4D_S^2\omega_k^2}}{2D_S}$, a critical line of V-α can be numerically calculated since it is impossible to obtain a explicit relation function between V and α through Eq.34. If we set the left hand of Eq.34 as a function $C(V, \alpha)$, then $C>0$ means the interface is cellular; $C\leq0$ means that interface branch occurs.

Actually, cellular shape is considered as a stable pattern of solidification interface through it has destroyed the MS stability [23], therefore, $C>0$ means the interface is stable and $C\leq0$ means the interface is instable. But a point should be noted is that this cellular-branch transition is not a instability dealt by the past instability analyses through perturbation method which answered a question that how the perturbation of a planar interface would evolve under an assumption that the solidification conditions, such as the temperature field and the concentration field, corresponding to the interface evolution could be automatically satisfied. An example process, temperature disturbances suddenly occur at the heater and the cooler of a Hele-Shaw system and then cause a infinitesimal deformation of the interface (this is possible according to out analysis front). For this process, the past instability analyses told us the deformation extent of the interface would , for alloy, become grater and grater if the solidification conditions locate in the instability region of the system for example the solidification rate exceeds a critical value, orelse the interface would return planar. Now, a question is that how the interface would evolve if the temperature disturbances were held at all times so the constrained temperature field in the system would not be that calculated for the interface evolution through the instability analysis, a condition would always exist in practice?

Our analyses front mean that for pure materials the interface could not return planar when temperature disturbances were held and if the solidification parameters (means V and α) lead $C>0$, the interface is cellular; if lead $C\leq0$, the interface would branch. Moreover, the interface shape is a kind of topological response to the temperature disturbances, their wavenumber is same but the temperature amplitude is magnified much by the interface response amplitude. Besides, a combination of solidification parameters before the temperature disturbance occuring just can response to a temperature disturbance component with wavenumber determined through the Eq.32, thus there exists wavenumber selection for a temperature disturbance which always can be expressed as a Fourier series in practice. As a consequence, it is not needed to analysis the interface shape under complex wave temperature disturbance.

Just as we have mentioned in section 1, a actual solidification is always directly controlled by temperature configuration, for a Hele-Shaw system adding a solidification rate controlling configuration. Therefore, the distance the interface to the cooler α should be corresponded to the directly controlled solidification parameters, which can be achieved through the Eq.25. Though a explicit function $\alpha=\alpha\,(V,\ \psi,\varphi)$ unfortunately can not be obtained by accurately solving Eq.25, we can know a trend that α decreases with the decrease of the heater temperature (notice α is negative) when the solidification rate and the cooler temperature are constant[44]. That means for a Hele-Shaw system the distance between interface and the cooler would increase with temperature gradient decrease.

5. Conclusion

It is shown that the directional solidification interface of a pure material bears periodic curve shape once the cooler and the heater temperature are disturbed by wave disturbance. The periodic curve is nearly sinusoidal wave when both the distance the interface to the cooler and the solidification rate are smaller than a certain value or the surface tension of the melt is very large. We can conclude that, therefore, the sinusoidal shape assumption of solidification interface always used by the infinitesimal perturbation analysis of interface instability is valid only when the solidification rate is very small, or the temperature gradient is very large, or the melt is of quite large surface tension. When the distance the interface to the cooler and the solidification rate increase, the interface is cellular shape with wavelength decreasing but grooves depth increasing, especially the depth could become very large. If they exceed certain critical values, the interface will branch.

These results imply that the interface shape in Hele-Shaw solidification experiments always used for studying alloy solidification interface stability in fact maybe is not planar but a cellular interface with quite small amplitude and the role of temperature disturbance should be considered since temperature disturbance is inevitable in actual experiments. If the temperature disturbance is overlooked, the interface instability threshold obtianed by experiment would be lower than that suggested by theory.

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APPENDIX

TABLE. Ⅰ Solutions of equation (2)

	λ	Solutions
Ⅰ	$(0, \infty)$	$T=\left[a\exp\left(\frac{-V+\sqrt{V^2+4D\lambda}}{2D}z\right)+b\exp\left(\frac{-V-\sqrt{V^2+4D\lambda}}{2D}z\right)\right]\left[m\cos\sqrt{\frac{\lambda}{D}}x+n\sin\sqrt{\frac{\lambda}{D}}x\right]+T^c$
Ⅱ	0	$T=\left[a+b\exp\left(-\frac{V}{D}z\right)\right]\left[m+nx\right]$
Ⅲ	$(-\frac{V^2}{4D}, 0)$	$T=\left[a\exp\left(\frac{-V+\sqrt{V^2+4D\lambda}}{2D}z\right)+b\exp\left(\frac{-V-\sqrt{V^2+4D\lambda}}{2D}z\right)\right]\left[m\exp\left(\sqrt{-\frac{\lambda}{D}}x\right)+n\exp\left(-\sqrt{-\frac{\lambda}{D}}x\right)\right]+T^c$
Ⅳ	$-\frac{V^2}{4D}$	$T=\exp\left(-\frac{V}{2D}z\right)\left[a+bz\right]\left[m\exp(\frac{V}{2D}x)+n\exp\left(-\frac{V}{2D}x\right)\right]+T^c$
Ⅴ	$(-\infty, -\frac{V^2}{4D})$	$T=\left[a\exp\left(-\frac{V}{2D}z\right)\cos\frac{\sqrt{-V^2-4D\lambda}}{2D}z+b\exp\left(-\frac{V}{2D}z\right)\sin\frac{\sqrt{-V^2-4D\lambda}}{2D}z\right]\bullet$ $\left[m\exp(\sqrt{-\frac{\lambda}{D}}x)+n\exp(-\sqrt{-\frac{\lambda}{D}}x)\right]+T^c$

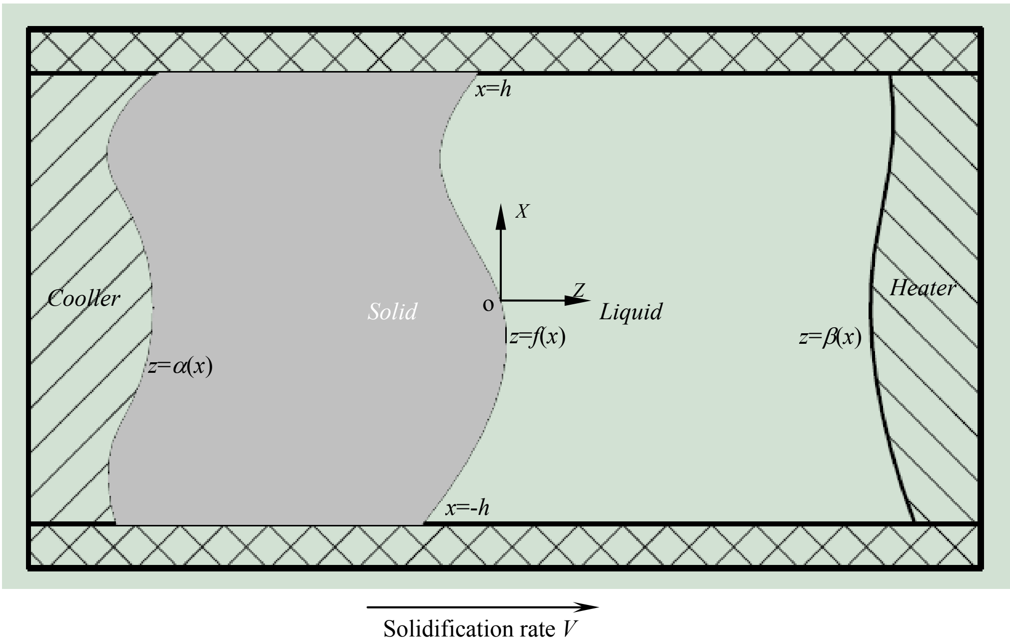

FIG.1 Directional solidification model with non-planar solid liquid interface

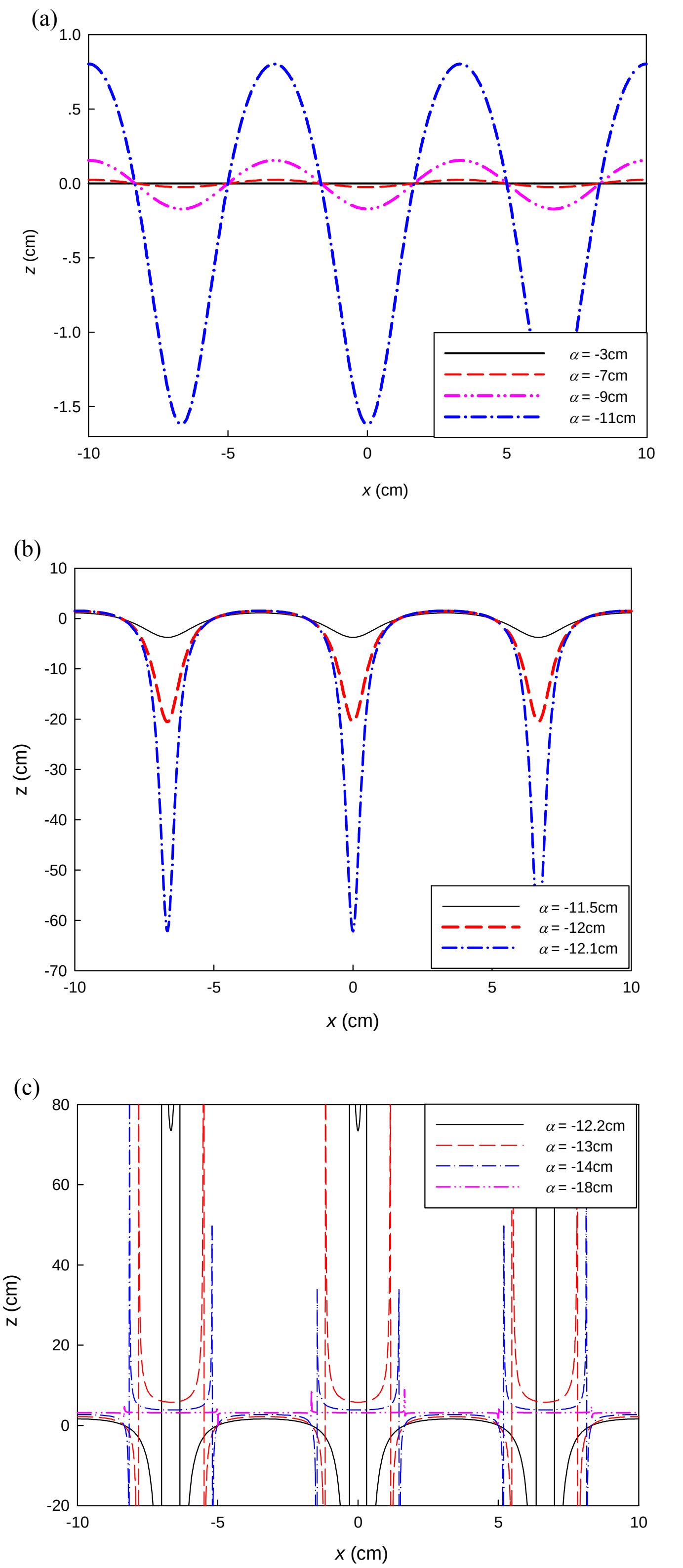

FIG.2 Evolution of interface shape with the increase of *a*. It was obtained through simulating the shape function, Eq.26, using parameters γ=0.001erg/cm^2, k=3 and δ_3=0.000001K.

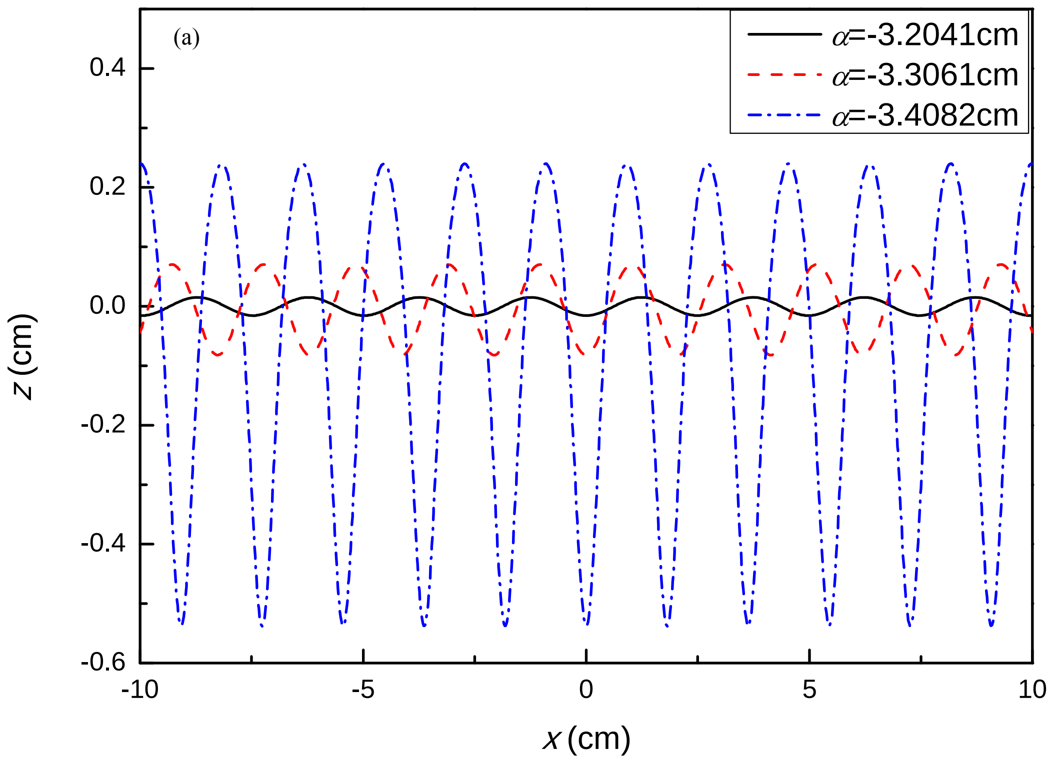

FIG.3 Evolution of the interface shape with increase of α at a fixed solidification rate. It was obtained using parameters δ_k=0.000001K, γ=0.001erg/cm^2 and V=0.001cm/s.

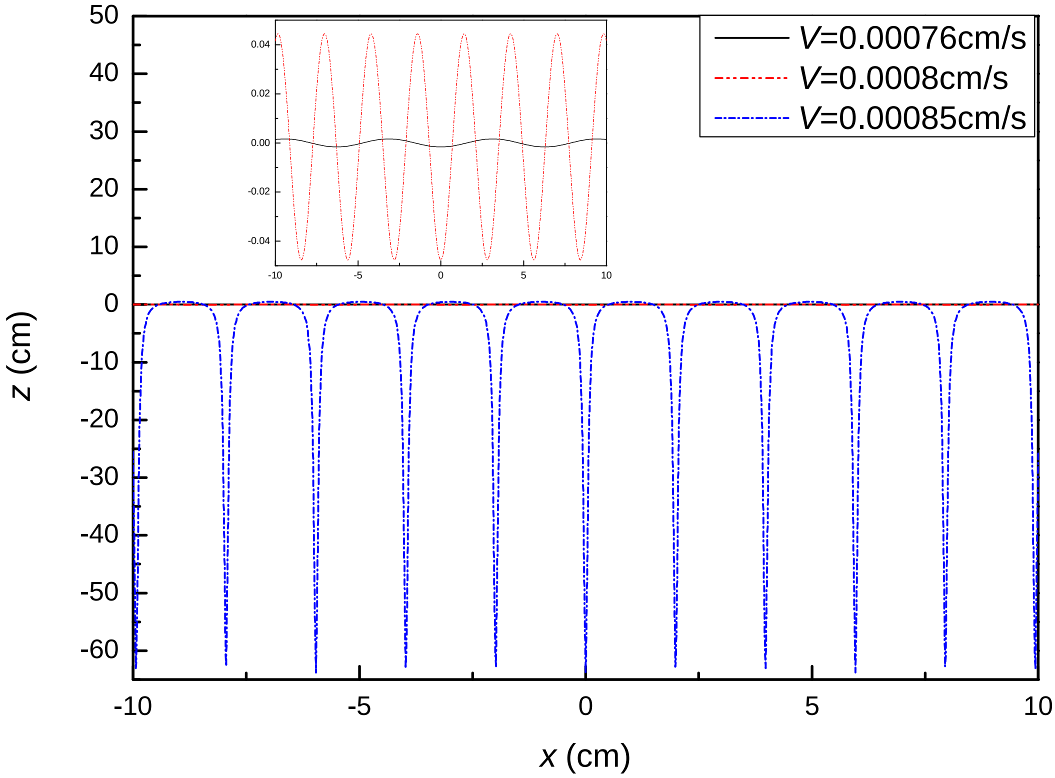

FIG.4 Evolution of the interface shape with increase of V. It was obtained using parameters α=-4cm, δ_k=0.000001K and γ=0.001erg/cm^2.

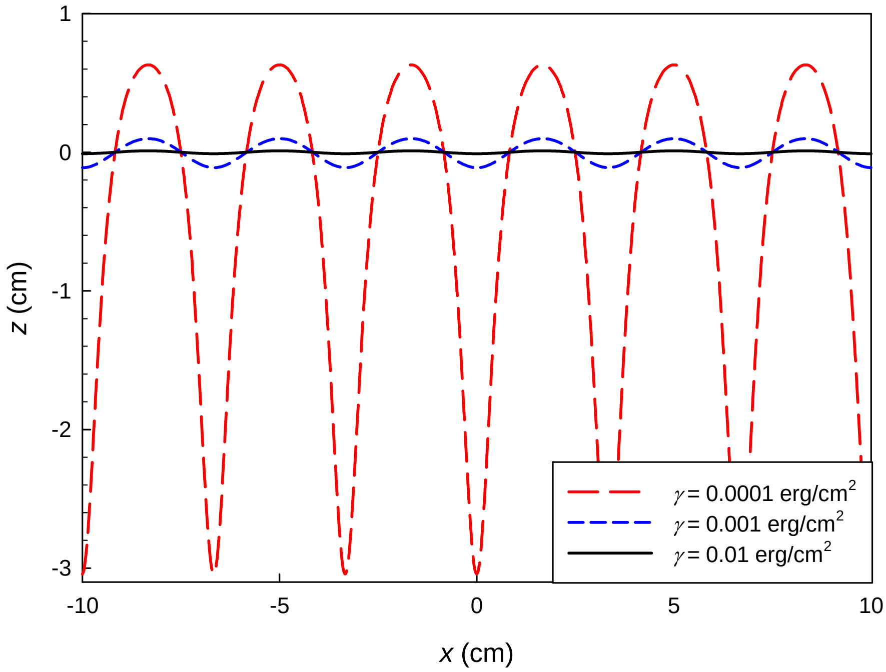

FIG.5 Evolution of the interface shape with increase of γ. It was obtained using parameters a=-5cm, k=6 and δ_6=0.000001K