We present differential equation for evolution of interface based on continuous approximation of Temkin’s model.

We start from a free energy functional of type

\[
F[\phi] = \int a \nabla \phi \ln(a \nabla \phi) + \phi^2 f_S + (1 - \phi)^2 f_L + 2\phi(1 - \phi)f_{LS} d^3x
\]  

(1)

Where \(\phi\) is the order parameter equivalent to fraction of solid, and \(1 - \phi\) is the fraction of liquid in Temkin’s model of solidification, we can also assume that \(\phi\) represents fraction of phase A and \(1 - \phi\) fraction of phase B in an AB system. Consequently \(f_S, f_L\) and \(f_{LS}\) are free energies of pure solid, liquid and liquid-solid phases, respectively. In case of Temkin’s model \(a\) is vector representing crystal lattice parameter, but in above functional we can generalize this parameter to represents a characteristic length of growth not necessarily related to the lattice parameter. The lattice vector \(a\) can be some function of the order parameter which controls the evolution of the characteristic length and depend on additional space variables including Euler angles but this further complicates our model. By doing the variational derivative of Eq. (1) and defining

\[
\alpha = f_{LS} - f_L + f_S
\]

and

\[
\beta = f_{LS} - f_S
\]

we have:

\[
\frac{1}{a \nabla \phi} \times \left( a_x^2 \partial_x^2 + a_y^2 \partial_y^2 + a_z^2 \partial_z^2 + 2a_x^2 a_y^2 \partial_{xy} + 2a_x^2 a_z^2 \partial_{xz} + 2a_y^2 a_z^2 \partial_{yz} \right) \phi - \alpha \phi + \beta = \frac{\partial \phi}{\partial t}
\]  

(2)

Eq. (2) describes the general case of anisotropy growth of interface in three dimensional space. The parameters \(\alpha\) and \(\beta\) are defined as \(L/kT\) and \(\Delta \mu/kT\), respectively. The Jackson parameter \(\alpha\) is measure of surface energy and \(\beta\) is difference in chemical potential between two phases.

We restrict our considerations only on isotropic case where: \(a_x = a_y = a_z = a\) and one dimensional case reducing Eq.(2) to:

\[
\frac{d^2 \phi}{dx^2} \frac{\partial \phi}{\partial x} - \alpha \phi + \beta = \frac{\partial \phi}{\partial t}
\]  

(3)

We will now give some solutions of previous evolution equation.

I. EQUILIBRIUM SOLUTION

The solution of differential equation (3) for equilibrium conditions \(\beta = 0\) and \(\frac{\partial \phi}{\partial t} = 0\) is: \(\phi(x) = -\tanh(\alpha/2a(x - \delta))\) where \(\delta\) is an arbitrary constant representing current position of the interface. The thickness of the interface is given by:

\[
d = \frac{2a}{\alpha}
\]  

(4)

This result is in agreement with Temkin’s theory and it’s already given in. It predicts that interface thickness would depend on the parameter which Temkin defines as:

\[
\alpha = \frac{z \epsilon}{kT} = \frac{L}{kT}
\]  

(5)

Where \(z\) is number of nearest neighbors or coordination number and \(\epsilon = \epsilon_{sl} - \epsilon_{ss} + \epsilon_{ll}\) is binding energy of the interface. Interesting consequence of relation (5) is prediction of the thinner interface film for higher energy of binding when the thickness \(d\) is normalized with lattice constant \(a\). This can be compared with experimental facts for different materials.

Using Eq.(5) we can estimate the equilibrium thickness of the intergranular film for usual ceramic materials. If we take approximate values for ceramics materials, the equilibrium temperature of about 2200 K and corresponding value for latent heat \(L = 1.5 \times 10^{-20}\) J/atom we get the thickness of intergranular film which is in agreement with the experimental findings and previous theoretical work of Bobeth et al.
II. VECTOR CASE

Providing that we have slow growth of the crystal in x-direction we expect a slow change of \( a_x \) due to the small temperature gradient. Thus in the limiting case of the adiabatic change we can change \( a_x \) with the constant value of \( a \). In the general case \( a_x \) will change its value with the time \( a_x(t) \). We would like to describe the change of \( a_x \) as a function of time and the simplest model we can imagine is a harmonic oscillator. Then we can define \( a_x \) as:

\[
a_x = a(1 + \cos(\omega t))
\]

Where \( \omega \) is the mean angular velocity of all atoms at the interface. Solution of our new differential equation in the case of vanishing chemical potential \( \beta = 0 \) is:

\[
\phi(x, t) = -\tanh(m(x - \delta)) \times \left[ \frac{2am(\alpha^2 + \omega^2 + \alpha^2 \cos(\omega t) + \alpha \omega \sin(\omega t))}{\alpha(\alpha^2 + \omega^2)} \right]
\]

Where \( m \in N \) Eq. (7) comes from the solution by separation of variables. This is wave-like tangent hyperbolical solution contains two parts, first which is proportional to thickness of the interface and second is harmonic oscillations of atoms at the interface. The harmonic oscillations of the diffuse interface will be govern by ratio \( \alpha/\omega \).

III. EQUILIBRIUM SOLUTION IN 2 D CASE

In this section we map the solution of differential Eq.(2) in the case of cylindrical symmetry to solution of Burgers vector equation:

\[
\frac{\partial U}{\partial t} + U \nabla U = \nu \nabla^2 U
\]

This non-linear partial differential equation is described and solved in the detail in [7,9]. Main idea is to change the vector field \( U \) with the gradient of the scalar field \( \psi \) imposing \( \nabla \times U = 0 \) and \( \nabla \psi = U \). Modifying Eq. (8) we can simplify equation to:

\[
\frac{\partial \psi}{\partial t} + \left( \frac{\nabla \psi}{2} \right)^2 - \nu \nabla^2 \psi = E(t)
\]

Where \( E(t) \) is function of time only. Applying the same trick to our Eq. (2) we get simplified version:

\[
a \nabla^2 u - \frac{\alpha}{2} (\nabla u) + \beta \nabla u - \frac{\partial \phi}{\partial t} \nabla u = 0
\]

Where \( u = \nabla \phi \) and \( \nabla \times u = 0 \). For case of the cylindrical symmetry and case where lattice vector \( a \) is always parallel to normal of phase field, Eq. (10) can be reduced to:

\[
a \frac{\partial}{\partial r} \left( \frac{1}{r} \frac{\partial}{\partial r} (r \phi) \right) - \alpha \phi + \beta = \gamma \frac{\partial \phi}{\partial t}
\]

Equilibrium solution to Eq. (11) is given as:

\[
\phi(r) = -\frac{2a/\alpha}{(r - \delta) \ln(r - \delta)}
\]

As in the one dimensional case the thickness is characterized by \( 2a/\alpha \), but it has logarithmic divergence to infinity.

IV. DEPENDENCE OF THE INTERGRANULAR FILM THICKNESS ON THE MISFIT ANGLE AND GRAIN BOUNDARY ANGLE

The potential energy of grain boundary in Read-Shockley only depends on the dislocations in the crystal, produced by the mismatch of the crystal lattices. If the amorphous intergranular film is in the thermodynamic equilibrium with
crystalline solid, the necessary condition for thermodynamic equilibrium is the minimum of the free energy leading to conclusion that amorphous intergranular film has a lower free energy than grain boundary between two crystalline solids. If we take that $\varphi$ is angle of grain boundary and $\theta$ is misfit angle, then from Read-Shockley theory of the grain boundary dislocations in the crystals we can write potential energy of grain boundary as:

$$E = E_o \theta (A - \ln \theta)$$  \hspace{1cm} (13)

Necessary condition for existence of the amorphous intergranular layer is:

$$E \geq E_{amorph}$$  \hspace{1cm} (14)

Where:

- $E_o = Ga (\cos \varphi + \sin \varphi)/(4\pi (1 - \sigma))$
- $E_{amorph}$ - energy of an amorphous-crystal interface
- $A$ - function depending on $\varphi$
- $\theta$ - misfit angle

We assume that energy of amorphous layer is exactly equal to grain boundary energy of the Read-Schokley theory, which fulfill the necessary condition for thermodynamic equilibrium of amorphous film. In perfect solid the lattice vector $a$ is oriented in one particular direction, while in the diffuse interface the lattice vector has statistical distribution. This can be pictured as Ising XY-model where grain boundaries are regions in the plane where spins have different orientations and regions of the crystal are perfectly oriented.

The probability of an orientation of a lattice vector in the space angle is given by:

$$dW_o = \text{const} \ dK$$  \hspace{1cm} (15)

Or using the Boltzmann factor:

$$dW = \text{const} \ e^{E_o \theta (A - \ln \theta)/kT \sin \varphi \ d\varphi}$$  \hspace{1cm} (16)

$$(a_x) = \frac{\int a_x dW}{\int dW} = \frac{\int a \cos \varphi \exp \left(\frac{Ga (\cos \varphi + \sin \varphi)}{4\pi (1 - \sigma)} \theta (A - \ln \theta)/kT\right) \sin \varphi \ d\varphi}{\int \exp \left(\frac{Ga (\cos \varphi + \sin \varphi)}{4\pi (1 - \sigma)} \theta (A - \ln \theta)/kT\right) \sin \varphi \ d\varphi} = a \sqrt{2}/2$$

If we assume that $\cos \varphi \gg \sin \varphi$ which is true for small angles, the exponent in exponential function is simply $Ga (\cos \varphi + \sin \varphi) \approx Ga \cos \varphi$, changing the integration variable $x = a \cos \varphi G/4\pi (1 - \sigma) \theta (A - \ln \theta)$ and collecting all parameters in $\lambda = \frac{G}{4\pi (1 - \sigma)} \theta (A - \ln \theta)$ we have the following integral:

$$(a_x) = \frac{1}{\lambda} \int_0^{a_x} \frac{x e^x \ dx}{\int_0^{a_x} e^x \ dx} = \frac{1}{\lambda} \int_0^{a_x} \frac{x e^x \mid_0^{a_x} - e^x \mid_0^{a_x}}{e^x \mid_0^{a_x}} = \frac{e^{a \lambda/2} (a \lambda - 1) + e^{-a \lambda/2}}{2 \lambda \sinh(a \lambda/2)}$$

In most general cases $\lambda \approx 10^{26}$ is a big number for ceramic materials, so the value of $a_x$ can be calculated as $\lambda$ tends to infinity.

$$\lim_{\lambda \to \infty} \frac{e^{a \lambda/2} (a \lambda - 1) + e^{-a \lambda/2}}{2 \lambda \sinh(a \lambda/2)} = a$$  \hspace{1cm} (18)

Thus we see that in limes for small misfit angles and the condition that $\cos \varphi \gg \sin \varphi$ we have uniform thickness of the intergranular films. This is in good agreement with the experiments. Let’s calculate the special case where $\lambda = 0$, this the case when angle of misfit is $\theta = 0$.

$$\lim_{\lambda \to 0} \frac{e^{a \lambda/2} (a \lambda - 1) + e^{-a \lambda/2}}{2 \lambda \sinh(a \lambda/2)} = a/2$$  \hspace{1cm} (19)
Previous calculation can be used together with Eq. (I) to calculate the thickness $\langle d \rangle = \frac{2(\alpha)}{\alpha}$ of intergranular film in polycrystalline material. From this consideration we conclude that average thickness over wide range of crystallographic orientations is almost constant and it reduces to half this value only in the case where the misfit angle is zero.

V. KINETICS OF THE INTERGRANULAR FILM

The kinetics of interface in one dimensional case is already given by Mori et al. Solution of Eq. (2) is given as:

$$\tanh(v) = \frac{v}{(\beta/\alpha)v + 1}$$

(20)

Where $v = V \tau / a$ is non-dimensional velocity of the interface and $V$ is real velocity of the interface. Velocity diverges at point where $\beta \to \alpha$ this is non-physical solution but the validity of Time-Dependent-Ginsburg-Landau equation (TDGL) is questionable in the range of very high velocity of the interface. We can give several limiting cases for Eq. (20):

$V \gg a/\tau (\beta \rightarrow \alpha) :$

$$v \simeq -1/2 \ln \frac{1 - (\alpha/\beta)}{2}$$

$-V \gg a/\tau (\beta \rightarrow -\alpha) :$

$$v \simeq 1/2 \ln \frac{1 + (\alpha/\beta)}{2}$$

We conclude with application of evolution equation to phase separation in polymer mixtures. This is usually described with Flory-Huggins free energy given as:

$$\frac{F}{kT} = \phi_A \ln \phi_A + \phi_B \ln \phi_B + N\chi \phi_A \phi_B$$

(21)

Where $N$ is degree of polymerization, $\chi$ interaction parameter per monomer (Flory-Huggins parameter), $\phi_A$ and $\phi_B$ is probability of site in space is occupied with molecule of A or molecule of B species, respectively. Let’s take a characteristic length of polymer chain to be is given by $w = a\sqrt{N}$ where $a$ is size of monomer and $N$ characteristic loop size of random walk of polymer A protruding in space of polymer B. Then total energy coming from interaction between A and B segments of polymer is $L = \chi N kT \alpha = \frac{L}{\chi kT} = \chi N$. If we put this result inside Eq. (4) we have:

$$d = \frac{2w}{\alpha} = \frac{2a\sqrt{N_{loop}}}{\chi N_{loop}} = \frac{2a}{\chi \sqrt{N_{loop}}}$$

(22)

For equilibrium the interaction energy of monomer will be of order $kT$ thus: $\chi N_{loop} \approx 1$. Giving finally in equilibrium case:

$$d \approx \frac{2a}{\sqrt{\chi}}$$

(23)

Which is of course not a surprise as Temkin’s model is equivalent to Flory-Huggins theory of polymer mixtures.

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VI. APPENDIX

\[
\frac{\partial f}{\partial \phi} = \frac{\partial f}{\partial \phi} - \sum \frac{\partial}{\partial x_i} \left( \frac{\partial f}{\partial x_i} \right) \tag{24}
\]

\[
\frac{\partial f}{\partial \phi_x} = \frac{\partial}{\partial \phi_x} \left( (a_x \phi_x + a_y \phi_y + a_z \phi_z) \ln(a_x \phi_x + a_y \phi_y + a_z \phi_z) \right) \tag{25}
\]

Then

\[
\frac{\partial}{\partial x} \left( \frac{\partial f}{\partial \phi_x} \right) = \frac{\partial}{\partial x} \left( a_x \ln(a_x \phi_x + a_y \phi_y + a_z \phi_z) + a_x^2 \phi_x + a_y \phi_y + a_z \phi_z \right) \tag{26}
\]

Similarly, for components y and z we have:

\[
\frac{\partial}{\partial y} \left( \frac{\partial f}{\partial \phi_y} \right) = \frac{a_y}{a \nabla \phi} \left( a_x \phi_{xy} + a_y \phi_{yy} + a_z \phi_{zy} \right) \tag{27}
\]

\[
\frac{\partial}{\partial z} \left( \frac{\partial f}{\partial \phi_z} \right) = \frac{a_z}{a \nabla \phi} \left( a_x \phi_{xz} + a_y \phi_{yz} + a_z \phi_{zz} \right) \tag{28}
\]

Then the sum of these terms is:

\[
\frac{\partial}{\partial x} \left( \frac{\partial f}{\partial \phi_x} \right) + \frac{\partial}{\partial y} \left( \frac{\partial f}{\partial \phi_y} \right) + \frac{\partial}{\partial z} \left( \frac{\partial f}{\partial \phi_z} \right) = \frac{\partial^2 f}{\partial \phi_x^2} + \frac{\partial^2 f}{\partial \phi_y^2} + \frac{\partial^2 f}{\partial \phi_z^2} = \frac{1}{a \nabla \phi} \left( a_x^2 \phi_{xx} + a_y^2 \phi_{yy} + a_z^2 \phi_{zz} + 2a_x a_y \phi_{xy} + 2a_x a_z \phi_{xz} + 2a_y a_z \phi_{yz} \right) \tag{29}
\]
A. One-dimensional solution to evolution equation

We start from partial differential equation:

\[
a \frac{\partial^2 \phi}{\partial x^2} \frac{\partial \phi}{\partial x} - \alpha \phi + \beta = \tau \frac{\partial \phi}{\partial t}
\]  

(30)

with change of variables: \( u(\phi) = a \frac{\partial \phi}{\partial x} \) we have:

\[
\frac{\partial u(\phi)}{\partial \phi} = \frac{\partial}{\partial \phi} \left( a \frac{\partial \phi}{\partial x} \right) = a \frac{\partial^2 \phi}{\partial x^2} \frac{\partial \phi}{\partial x} = a \frac{\partial^2 \phi}{\partial \phi^2} \frac{\partial \phi}{\partial x}
\]  

(31)

\[
\frac{\partial \phi}{\partial t} = -V \frac{\partial \phi}{\partial x}
\]  

and we transform the equation in :

\[
\frac{\partial u}{\partial \phi} + vu - \alpha \phi + \beta = 0 \tag{32}
\]

\[
u(\phi) = Ce^{-\phi} - \frac{\alpha}{\beta} \frac{1}{\phi} - \frac{\beta}{\phi}
\]  

(33)

where

\[
C = \sqrt{\left( \frac{\alpha}{\beta} + \frac{\beta}{\phi} \right)^2 - \left( \frac{\alpha}{\beta} \right)^2}
\]  

(34)

Final solution is: \( \tanh \frac{v}{\beta/\alpha + 1} \) which for \( \beta = 0 \) gives \( \tanh v = -v \).

B. One-dimensional solution of evolution equation with harmonic oscillations

\[
\frac{\partial \phi}{\partial t} = \beta - \alpha \phi + a(1 + \cos(\omega t)) \frac{\partial^2 \phi}{\partial x^2} / \frac{\partial \phi}{\partial x}
\]

Let’s solve it by separation of variables:

\[
\phi(x, t) = X(x)T(t) \tag{35}
\]

Where \( X(x) \) is only function of \( x \) and \( T(t) \) is only function of \( t \). Then, derivatives are:

\[
\frac{\partial^2 \phi}{\partial x^2} = T(t) \frac{\partial^2 X}{\partial x^2} \tag{36}
\]

\[
\frac{\partial \phi}{\partial x} = T(t) \frac{\partial X}{\partial x} \tag{37}
\]

\[
\frac{\partial \phi}{\partial t} = \frac{\partial T}{\partial t} X(x) \tag{38}
\]

Substituting these derivatives in differential equation:

\[
a(1 + \cos(\omega t)) \frac{\partial^2 X}{\partial x^2} / \frac{\partial x}{\partial x} - \alpha XT + \beta = X \frac{\partial T}{\partial t} \tag{39}
\]

Dividing this differential equation with \( X \) we get:

\[
\frac{1}{X} a(1 + \cos(\omega t)) \frac{\partial^2 X}{\partial x^2} / \frac{\partial x}{\partial x} - \alpha T + \beta / X = \frac{\partial T}{\partial t} \tag{40}
\]

If search for solution of where there is no driving force for transition so we have \( \beta = 0 \):

\[
\frac{1}{X} \frac{\partial^2 X}{\partial x^2} / \frac{\partial x}{\partial x} - (\alpha T + \frac{\partial T}{\partial t}) \frac{1}{a(1 + \cos(\omega t))} = 0 \tag{41}
\]
This equation is separable, and we get two ordinary differential equations:

\[
\frac{d^2 X}{dx^2} - 2m \frac{dX}{dx} = 0
\]  
(42)

Where m is integer:

\[
\frac{dT}{dt} + \alpha T - 2ma(\cos(\omega t) + 1) = 0
\]  
(43)

The solution of second differential equation is simply:

\[
T(t) = C \exp(-\alpha t) + \frac{2am}{\alpha^2 + \omega^2 + \alpha^2 \cos(\omega t) + \alpha \omega \sin(\omega t)} \alpha(\alpha^2 + \omega^2)
\]  
(44)

The solution to first differential equation can be found by putting solution \( X(x) \) back in the equation:

\[
X(x) = -\tanh(m(x - \delta))
\]  
(45)

So final solution is:

\[
\phi(x,t) = -\tanh(m(x - \delta)) \left[ \frac{2am(\alpha^2 + \omega^2 + \alpha^2 \cos(\omega t) + \alpha \omega \sin(\omega t))}{\alpha(\alpha^2 + \omega^2)} \right]
\]  
(46)

C. Calculation of Read-Schokley integral

Calculation of the integral:

\[
\langle a_x \rangle = \frac{\int a_x dW}{\int dW} = \frac{\int \cos \phi \exp(Ga(\cos \phi + \sin \phi)/(4\pi(1 - \sigma))\theta(A - \ln \theta)/kT) \sin \phi d\phi}{\int \exp(Ga(\cos \phi + \sin \phi)/(4\pi(1 - \sigma))\theta(A - \ln \theta)/kT) \sin \phi d\phi}
\]  
(47)

We will evaluate this integral with method given in Massida. All constant factors can be extracted from previous integral and then we need to solve general integral of type:

\[
\int_0^{2\pi} e^{in\phi} \exp(\cos \phi + \sin \phi) d\phi
\]  
(48)

Using relation between exponential and Bessel function:

\[
\exp(\frac{z}{2}(t + \frac{1}{t})) = \sum_{k=\pm\infty} t^k I_k(z) \]  
(49)

\[
e^{\cos \phi} = \sum_k t^k I_k(1) = \sum_k e^{ik\phi} I_k(1)
\]  
(50)

\[
e^{\sin \phi} = \sum_h (-i)^h e^{ih\phi} I_h(1)
\]  
(51)

Thus integral can be written as:

\[
\int_0^{2\pi} e^{in\phi} \sum_h (-i)^h e^{ih\phi} I_h(1) \sum_k e^{ik\phi} I_k(1) d\theta
\]  
(52)

Using \( I_m(x) = I_m(x) \) we can transform integral to \( 2\pi \sum_k I_k(1)I_{k+1}(-i)^k \) using addition theorem for Bessel functions:

\[
\sum_n (-1)^n e^{in\phi} I_n(z)I_{n+\nu}(Z) = I_\nu(\omega)e^{i\nu\psi}
\]  
(53)
Where \( \omega = (z^2 + Z^2 - 2zZ\cos \phi)^{1/2} \) and \( \psi \) is defined by \( Z - z \cos \phi = \omega \cos \psi \) and \( z \sin \phi = \omega \sin \psi \). The addition theorem can then be applied to k summation in sum and we get solution as:

\[
2\pi \sum_k I_k(1)I_{k+1}(1)(-i)^k = I_1(\sqrt{2(1 - \cos \phi)})e^{i \arccos(\sqrt{2(1 - \cos \phi)})/2}
\]  

(54)

Final solution to integral is:

\[
\int_0^{2\pi} \cos(x) \exp[\cos(x) + \sin(x)] \, dx = 2\pi I_1(\sqrt{2})\frac{\sqrt{2}}{2}
\]  

(55)

Similarly for:

\[
\frac{1}{2} \int_0^{2\pi} \sin(2x) \exp[\cos(x) + \sin(x)] \, dx = I_v(\omega)e^{iv\phi} = I_2(\sqrt{2})2\pi
\]  

(56)

D. Two-dimensional solution of evolution equation

We start from partial differential equation:

\[
a \frac{\partial}{\partial r} \left( \frac{1}{r} \frac{\partial}{\partial r} (r \phi) \right) - \alpha \phi + \beta = \frac{\partial \phi}{\partial t} \tau
\]  

(57)

Using transformation from Nerney et al. we can write previous equation:

\[
\frac{d\theta}{d\phi} - \alpha \phi + \beta = -\nu \theta(\phi)
\]  

(58)

Where \( \nu = \frac{\nu}{\alpha} \) non-dimensional velocity, solution to equation is:

\[
\theta = Ce^{\nu \phi} + \alpha v \phi - \beta / \nu - \alpha / \nu^2
\]  

(59)

Where \( C = \sqrt{\left( \frac{\alpha}{\nu^2} + \frac{\beta}{\nu} \right) - \left( \frac{\alpha}{\nu} \right)^2} \). For equilibrium conditions is equation reduced to:

\[
a \frac{\partial}{\partial r} \left( \frac{1}{r} \frac{\partial}{\partial r} (r \phi) \right) - \frac{\partial \phi}{\partial r} \alpha \phi = 0
\]  

(60)

Solution of partial differential equation is:

\[
\frac{1}{r} \frac{\partial}{\partial r} (r \phi) = \frac{C}{\alpha} e^{\phi \alpha}
\]  

(61)

Determination of constant of integration C:

\[
\phi(r) = \begin{cases} 
1 \text{ when } r = 0, \\
-1 \text{ when } r = \infty
\end{cases}
\]  

(62)

Giving \( C = 0 \) and finally the solution to differential equation:

\[
\phi(r) = \frac{2a/\alpha}{r \ln r}
\]  

(63)