Diffusion-Induced Oscillations of Extended Defects

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PACS numbers: 68.35.Dv, 81.10.Aj, 05.70.Np

The interaction of propagating extended defects with a diffusion field frequently leads to oscillations or jerky motions of the defects. A prime example of such an effect is the oscillation of a solidification front, induced by the diffusion of the solute component in a dilute binary alloy, which is growing in the setup of directional solidification. In a large number of metallic materials this leads to the formation of banded structures [1], reflecting a periodic array of layers with high and low solute concentrations where the former ones show a dendritic microstructure. The appearance of similar banding effects has recently been discussed [2] in rapid solidification of colloids.

Layered structures are also generated by the oscillatory nucleation of a solid phase under the action of a diffusion field [3]. A related phenomenon is the oscillatory zoning, observed in solid solutions [4] and in natural minerals [5]. Another notable scenario is that of diffusion-controlled jerky motions of a driven grain boundary [6]. A similar behavior of dislocations in metallic alloys leads to the Portevin-Le Chatelier effect [7], denoting the appearance of jerky plastic deformations. We, finally, mention the oscillatory motion of a crack tip, which is coated by the nucleus of a new phase [8], replacing the attached cloud of a diffusion field.

Theoretical discussions of such effects either are of a phenomenological type, like those in Ref. [7], and partly in Refs. [1] and [2], or they rely on a Fokker-Planck [6], or a diffusion equation with non-equilibrium boundary conditions [9]. In all approaches the source of oscillatory defect motions is identified as an unstable regime where a reduction of the driving force leads to an increase of the defect velocity. Additional information is provided by simulations, based on phase-field models for directional solidification [10] and for nucleation [3] processes.

In the present Letter we introduce an extremely simple but powerful model for the diffusion-induced oscillatory motion of a planar interface, using the language adapted to the directional solidification of a dilute binary alloy. A major advantage of our approach is that it allows a transparent and, to a large extend, analytical evaluation. This includes a readjustment of the stability analysis by Merchant and Davis [11] who discovered an oscillatory instability, similar to that, discussed earlier by Coriell and Sekerka [12]. Also included is a clarifying analysis of the so far barely understood low-velocity sections of the cyclic trajectories, identified by Carrard et al. [1], and by Karma and Sarkissian [9]. The limit-cycle behavior, describing the oscillations of the interface deep inside the unstable regime, is, finally, in remarkable agreement with the simulation results by Conti [10].

Due to the restriction to a planar interface, our model is a one-dimensional version of the capillary-wave model, derived in Ref. [13] from a phase-field model. It is given in dimensionless form by the set of equations

$$H = \frac{\gamma}{2} \int_{-\infty}^{+\infty} dz [C(z,t) - U(z - Z(t))]^2,$$

$$\partial_t Z = p \left( F - \frac{\delta H}{\delta Z} \right), \quad \partial_t C = \partial_z^2 \frac{1}{\gamma} \frac{\delta H}{\delta C},$$

$$F = F_p - m^2 [Z(t) - v_p t]$$

for the interface position $Z(t)$, and for the excess solute concentration $C(z,t)$ relative to its value $C_S \equiv 1$ in the solid phase. The parameter $\gamma$ measures the miscibility gap $\Delta C = C_L - C_S$ where $C_L$ is the solute concentration in the liquid phase, and $p$ measures the mobility of the interface. From the equilibrium condition $\delta H/\delta C = 0$ it follows that $U(z - Z)$ is the equilibrium-concentration
profile at some fixed temperature $T_S$. This profile reveals
the smooth solid-liquid transition region of the original
phase-field model, and is regarded as an input quantity
of the model \([1]\). It effectively comprises non-equilibrium
effects of sharp-interface descriptions, which are crucial
for the behavior in the rapid-growth regime, including
the solute-trapping effect \([14]\).

The driving force $F$ includes two quantities, appearing
in the simplest scenario of directional solidification. One
of them is a constant temperature gradient, the other is
a model pre-factor of order one. Adopting
from Ref. \([10]\) typical values for $S,T,M$, the parameter
$m^2$ is of order $10^{-5}$. An independent second quantity is
the velocity $v_p$, applied in pulling the growing crystal in
opposite direction to the temperature gradient. The local
temperature $T_P$ at the steady-state position $Z(t) = v_P t$
finally, determines the fragment $F_P \equiv A(T_S - T_P)/T_M$
of the driving force $F$.

The resulting equations for $Z(t)$ and $C(z,t)$ read

\[
\begin{align*}
\frac{1}{p} \dot{Z}(t) &= F_P - m^2 [Z(t) - v_P t] \\
&- \gamma \int_{-\infty}^{+\infty} dz U'(z - Z(t))[C(z,t) - U(z - Z(t))] ,
\end{align*}
\]

(2)

For steady-state boundary conditions $C(z = \pm \infty) = 0$, they have the stationary solutions $Z(t) = v_P t$, and
$C(z,t) = C_P(z - v_P t; v_P)$, resulting in the relations

\[
\begin{align*}
\frac{1}{p} v_P &= F_P + G_P(v_P) - G_P(0) , \\
G_P(v_P) &= -\gamma \int_{-\infty}^{+\infty} d\zeta U'(\zeta) C_P(\zeta; v_P) , \\
C_P(\zeta; v_P) &= \int_{-\infty}^{\zeta} d\zeta' U'(\zeta') \exp[v_P(\zeta' - \zeta)] .
\end{align*}
\]

(3)

We are primarily interested in the late-stage behavior
of non-stationary solutions $Z(t)$, and, therefore, look for
a solution $C(z,t)$ of the last equation in Eqs. (2), obeying
the boundary condition $C(z, -\infty) = 0$. This leads to the
expression

\[
C(z,t) = \int_{-\infty}^{t} dt' \int_{-\infty}^{+\infty} dz' \partial_{z'} G(z - z', t - t') \cdot U'(z' - Z(t')) ,
\]

(4)

involving the Green function

\[
G(z,t) = \int_{-\infty}^{+\infty} \frac{dk}{2\pi} \exp(-k^2 t + i k z) .
\]

(5)

After the substitutions $\zeta = z - Z(t), \zeta' = z' - Z(t')$, and
expansion of $Z(t')$ around $Z(t)$, we obtain

\[
G(\zeta - \zeta' + Z(t) - Z(t'), t - t') =
\]

(6)

\[
\int_{-\infty}^{+\infty} \frac{dk}{2\pi} \exp[-k^2(t - t') + ik(\zeta - \zeta') + ik(t - t')v(t)]
\cdot \exp\left[ -ik \sum_{n \geq 2} \frac{(-1)^n}{n!} (t - t')^n \partial_{\zeta}^{n-1} v(t) \right]
\]

where $v(t) \equiv Z'(t) = v_P + \dot{h}(t)$.

If one temporarily applies the scaling transformations
$h \to m^{-2}h, \partial_t \to m^2 \partial_t$, one observes that, whereas $v(t)$
remains unchanged, a factor $m^{2n-2}$ is attached to the
contributions $\partial_{\zeta}^{n-1} v(t)$. Therefore, with increasing $n$
these terms are progressively negligible in Eq. (6) due to
the smallness of $m^2$. Neglecting all terms of order $n \geq 2$
we encounter the quasi-stationary approximation, which
is often used in phenomenological approaches. As we
shall see, however, a proper understanding of oscillatory
motions of the interface requires to incorporate the term
of order $n = 2$.

Evaluation of Eqs. (4)-(6) then leads to the expression

\[
C(z,t) = C_P(\zeta; v) + v \cdot \frac{1}{2} \partial_{\zeta}^2 \left[ C_P(\zeta; v) + C_P(\zeta; 0) \right]
\]

(7)

with $C_P(\zeta; v_P)$ determined by the last line in Eqs. (3).
Insertion of this result into the first equation in Eqs. (2)
yields the relation

\[
\frac{1}{p} v = F_P - m^2(Z - Z_P)
\]

(8)

\[
+ G_P(v) - G_P(0) + v \cdot \frac{1}{2} \partial_{\zeta}^2 \left[ G_P(v) + G_P(0) \right]
\]

with $G_P(v)$ following from the second line in Eqs. (3).
For $v = v_P$ the result \([8]\) consistently reduces to the first
equation in Eqs. (3). Subtracting the latter from Eq. (8),
we, finally, find for the displacement $\dot{h}(t) \equiv Z(t) - Z_P(t)$
the differential equation

\[
M(\dot{h}(t)) \ddot{h}(t) + R(\dot{h}(t)) + m^2 \dot{h}(t) = 0
\]

(9)

where we have introduced the mass and friction functions

\[
M(\dot{h}) \equiv -\frac{1}{2} \frac{\partial^2}{\partial v^2} \left[ G_P(v_P + \dot{h}) + G_P(0) \right] v_P + \dot{h}
\]

\[
R(\dot{h}) \equiv \frac{1}{p} \dot{h} - G_P(v_P + \dot{h}) + G_P(v_P) .
\]

(10)

Eq. (9) has the typical appearance of a nonlinear damped
oscillator, and represents one of the central results of the
Then, solving the integrals in Eqs. (3), one finds

\[ m(9) \text{ is only valid below the crossover line} \]

\[ v \]

and a limit cycle for \( m = 0 \) dependence of \( M \).

We mention that, due to the singular definitions (10) and the first line in Eqs. (3), yields

\[ F \]

can change sign at some critical velocity \( v \).

In this ordinary oscillator equation the friction coefficient \( h \) for \( \gamma = 0.01, \rho = 100, \]

\[ m = 0.003, \text{approaching the value} \]

\[ h = 0 \]

for \( v_p = 0.522, \]

and a limit cycle for \( v_p = 0.520. \)

In order to check the stability of the obvious solution \( h(t) = 0 \), we linearize Eq. (9) in \( h(t) \), which, due to the definitions (10) and the first line in Eqs. (3), yields

\[ M(0) \dot{h} + F_p(v_p) \dot{h} + m^2 h = 0 . \]

In order to check the stability of the obvious solution \( h(t) = 0 \), we linearize Eq. (9) in \( h(t) \), which, due to the definitions (10) and the first line in Eqs. (3), yields

\[ M(0) \dot{h} + F_p(v_p) \dot{h} + m^2 h = 0 . \]

which determines all terms in the oscillator equation (9).

The resulting numerical solutions for \( h(t) \) above and below the Cahn threshold \( v_C \) are shown in Fig. 1 where the approach to a limit cycle in the unstable regime is clearly visible. For small distances \( v_p - v_C \) \( \ll 1 \) and a limit cycle for \( h(t) = a(t) \cos \psi(t) \)

\[ h(t) = a(t) \cos \psi(t) \]

with \( \psi(t) \) a rapidly oscillating phase, and \( a(t) \) is an amplitude, obeying the differential equation

\[ \frac{d\rho}{dt} = -\rho_1 a - \rho_3 a^3 , \]

which for \( \rho_1 > 0 \) and \( \rho_1 < 0 \) describes the envelopes in Fig. 1. The asymptotic value of the limit-cycle amplitude shows the critical behavior \( a(\infty) = \sqrt{-\rho_1/\rho_3} \).

The numerically obtained limit-cycle trajectories of the quantities \( h(t), h(t), C(Z(t), t) \) deep inside the unstable regime are displayed in Fig. 2. They also inform on the local temperature at the oscillating interface, since this is measured by the quantity \( m^2 h(t) \).

For quantitative discussions of the behavior of \( h(t) \) we now adopt the specific model

\[ U(\zeta) = \Theta(-\zeta) \exp \zeta + \Theta(\zeta)[2 - \exp(-\zeta)] , \]

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derived in Ref. [13] from a double-parabola phase-field model. Then, solving the integrals in Eqs. (3), one finds

\[ G_p(v) = -\gamma \frac{v^2}{(v^2 + 1)^2} , \]

\[ G_p(v) = -\gamma \frac{v^2}{(v^2 + 1)^2} , \]

\[ \frac{\lambda^2 - q^2}{v_p + 2\lambda}[G_p(v_p + \lambda) + G_p(\lambda)] , \]

\[ \frac{\lambda^2 - q^2}{v_p + 2\lambda}[G_p(v_p + \lambda) + G_p(\lambda)] , \]

FIG. 1: Numerical solutions \( h(t) \) for \( \gamma = 0.01, \rho = 100, \]

\[ m = 0.003, \text{approaching the value} \]

\[ h = 0 \]

for \( v_p = 0.522, \]

and a limit cycle for \( v_p = 0.520. \)
with \( \lambda \equiv -(v_P/2) + \sqrt{(v_P/2)^2 + \omega + q^2} \) where the term \( m^2 \) is the only new element. The wave-number threshold \( q_c \) for the Mullins-Sekerka instability [16] is determined by the relations \( \omega_1(q_c) = \omega_1'(q_c) = 0 \). By elimination of \( q_c \) from these equations one generates the neutral-stability boundary of the instability in form of a function \( v_P(\gamma) \), with a parametric dependence on \( m \).

In Fig. 3 the projection of the limit cycle, belonging to Fig. 2, enters the Mullins-Sekerka unstable regime at low velocities where the interface develops a dendritic microstructure, a typical feature of banded structures in metallic alloys. The other small cycle in Fig. 3 generates layers of precipitation-free periodic solute concentrations.

The most obvious generalization of our procedure is to explore the formation of non-planar layering effects, which also is a field for experimental investigations. A typical example of such an effect is the oscillatory growth of a spherical nucleus, which has been discussed on the basis of a phase-field model in Ref. [3], and which we are going to reconsider within our approach.

A. L. Korzhenevskii wants to express his gratitude to the University of Düsseldorf for its warm hospitality. This work has been supported by the DFG under BA 944/3-3, and by the RFBR under N10-02-91332.

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