Diffusion-Induced Oscillations of Extended Defects

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From a simple model for the driven motion of a planar interface under the influence of a diffusion field we derive a damped nonlinear oscillator equation for the interface position. Inside an unstable regime, where the damping term is negative, we find limit-cycle solutions, describing an oscillatory propagation of the interface. In the case of a growing solidification front this offers a transparent scenario for the formation of solute bands in binary alloys and, taking into account the Mullins-Sekerka instability, of banded structures.

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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 are either of a phenomenological type, like those in Ref. [7], and partly in Refs. [1,2], or they rely on a Fokker-Planck [6] or a diffusion equation with nonequilibrium 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].

Because of 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

$$H = \frac{\gamma}{2}\int_{-\infty}^{+\infty} dz[C(z,t) - U(z - Z(t))]^2,$$
$$\partial_t Z = p\left(F - \frac{\delta H}{\delta Z}\right), \qquad \partial_t C = \partial_z^2 \frac{1}{\gamma}\frac{\delta H}{\delta C}, \tag{1}$$
$$F = F_P - m^2[Z(t) - v_P t]$$

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 γ 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 nonequilibrium 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 S, entering the parameter $m^2 \equiv A\xi S/T_M$, where, in physical units, ξ measures the width of the interface, visible in the profile $U(\zeta)$, T_M is the melting temperature of the pure solvent, and A is a numerical prefactor of order 1. Adopting from Ref. [10] typical values for S, T_M, ξ, 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

$$\begin{aligned}\frac{1}{p}\dot{Z}(t) &= F_P - m^2[Z(t) - v_P t] \\ &\quad - \gamma \int_{-\infty}^{+\infty} dz U'(z - Z(t))[C(z, t) \\ &\quad - U(z - Z(t))], \\ (\partial_t - \partial_z^2)C(z, t) &= -U''(z - Z(t)).\end{aligned} \tag{2}$$

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

$$\begin{aligned}\frac{1}{p}v_P &= F_P + G_P(v_P) - G_P(0), \\ G_P(v_P) &\equiv -\gamma \int_{-\infty}^{+\infty} d\zeta U'(\zeta) C_P(\zeta; v_P), \\ C_P(\zeta; v_P) &= \int_{-\infty}^{\zeta} d\zeta' U'(\zeta') \exp[v_P(\zeta' - \zeta)].\end{aligned} \tag{3}$$

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

$$C(z, t) = \int_{-\infty}^{t} dt' \int_{-\infty}^{+\infty} dz' \partial_{z'} \mathcal{G}(z - z', t - t') U'(z' - Z(t')), \tag{4}$$

involving the Green function

$$\mathcal{G}(z, t) = \int_{-\infty}^{+\infty} \frac{dk}{2\pi} \exp(-k^2 t + ikz). \tag{5}$$

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

$$\begin{aligned}&\mathcal{G}(\zeta - \zeta' + Z(t) - Z(t'), t - t') \\ &= \int_{-\infty}^{+\infty} \frac{dk}{2\pi} \exp[-k^2(t - t') + ik(\zeta - \zeta') + ik(t - t')v(t)] \\ &\quad \times \exp\Big[-ik \sum_{n \geq 2} \frac{(-1)^n}{n!} (t - t')^n \partial_t^{n-1} v(t)\Big],\end{aligned} \tag{6}$$

where $v(t) \equiv \dot{Z}(t) = v_P + \dot{h}(t)$.

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 $\propto \partial_t^{n-1} v(t)$. 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 quasistationary approximation, which is often used in phenomenological approaches. As we shall see, however, a proper understanding of oscillatory motions of the interface requires us to incorporate the term of order $n = 2$.

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

$$C(z, t) = C_P(\zeta; v) + \dot{v} \frac{1}{2} \frac{\partial^2}{\partial v^2} \frac{1}{v}[C_P(\zeta; v) + C_P(\zeta; 0)], \tag{7}$$

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

$$\begin{aligned}\frac{1}{p}v &= F_P - m^2(Z - Z_P) + G_P(v) - G_P(0) \\ &\quad + \dot{v} \frac{1}{2} \frac{\partial^2}{\partial v^2} \frac{1}{v}[G_P(v) + G_P(0)],\end{aligned} \tag{8}$$

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

$$M(\dot{h}(t))\ddot{h}(t) + R(\dot{h}(t)) + m^2 h(t) = 0, \tag{9}$$

where we have introduced the mass and friction functions

$$\begin{aligned}M(\dot{h}) &\equiv -\frac{1}{2} \frac{\partial^2}{\partial v_P^2} \Big[\frac{G_P(v_P + \dot{h}) + G_P(0)}{v_P + \dot{h}}\Big], \\ R(\dot{h}) &\equiv \frac{1}{p}\dot{h} - G_P(v_P + \dot{h}) + G_P(v_P).\end{aligned} \tag{10}$$

Equation (9) has the typical appearance of a nonlinear damped oscillator and represents one of the central results of the present Letter. We mention that, due to the singular dependence of $M(\dot{h})$ on $v_P + \dot{h}$, the differential equation (9) is only valid below the crossover line $m^2 \propto (v_P + \dot{h})^3$.

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 Eq. (3), yields

$$M(0)\ddot{h} + F_P'(v_P)\dot{h} + m^2 h = 0. \tag{11}$$

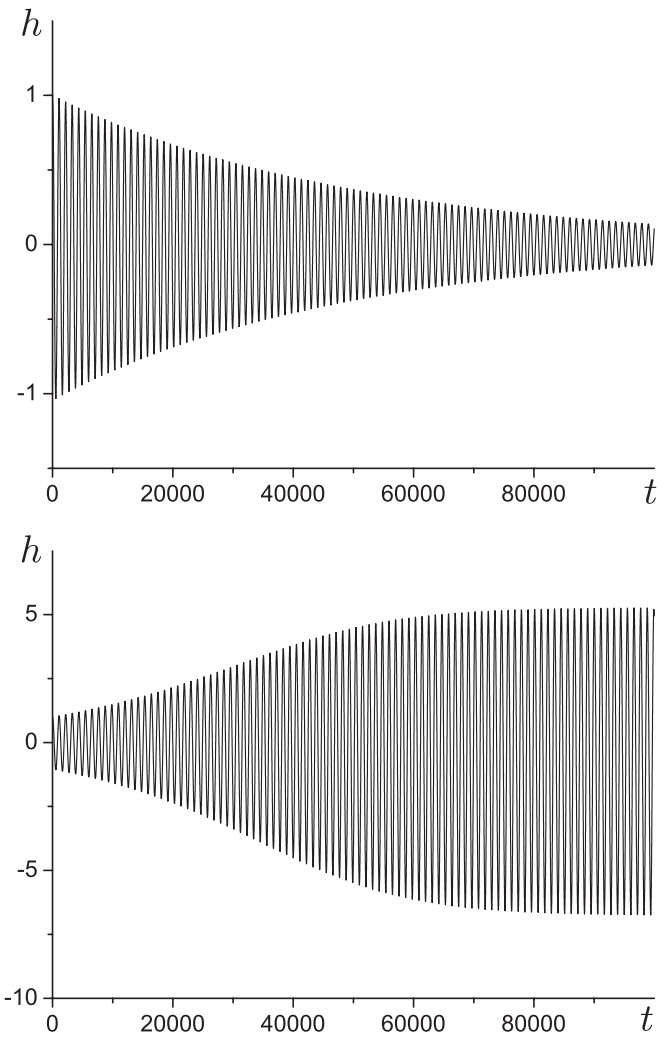

FIG. 1. Numerical solutions $h(t)$ for $\gamma = 0.01$, $p = 100$, $m = 0.003$, approaching the value $h = 0$ for $v_P = 0.522$, and a limit cycle for $v_P = 0.520$.

In this ordinary oscillator equation the friction coefficient can change sign at some critical velocity v_C, defined by $F'_P(v_C) = 1/p - G'_P(v_C) = 0$. A similar stability limit has been found by Cahn in grain-boundary motion [6].

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

$$U(\zeta) = \Theta(-\zeta)\exp\zeta + \Theta(\zeta)[2 - \exp(-\zeta)], \qquad (12)$$

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

$$G_P(v) = -\gamma\frac{v+2}{(v+1)^2}, \qquad (13)$$

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 $|v_P - v_C|/v_C \ll 1$ the envelopes of these curves can be calculated analytically

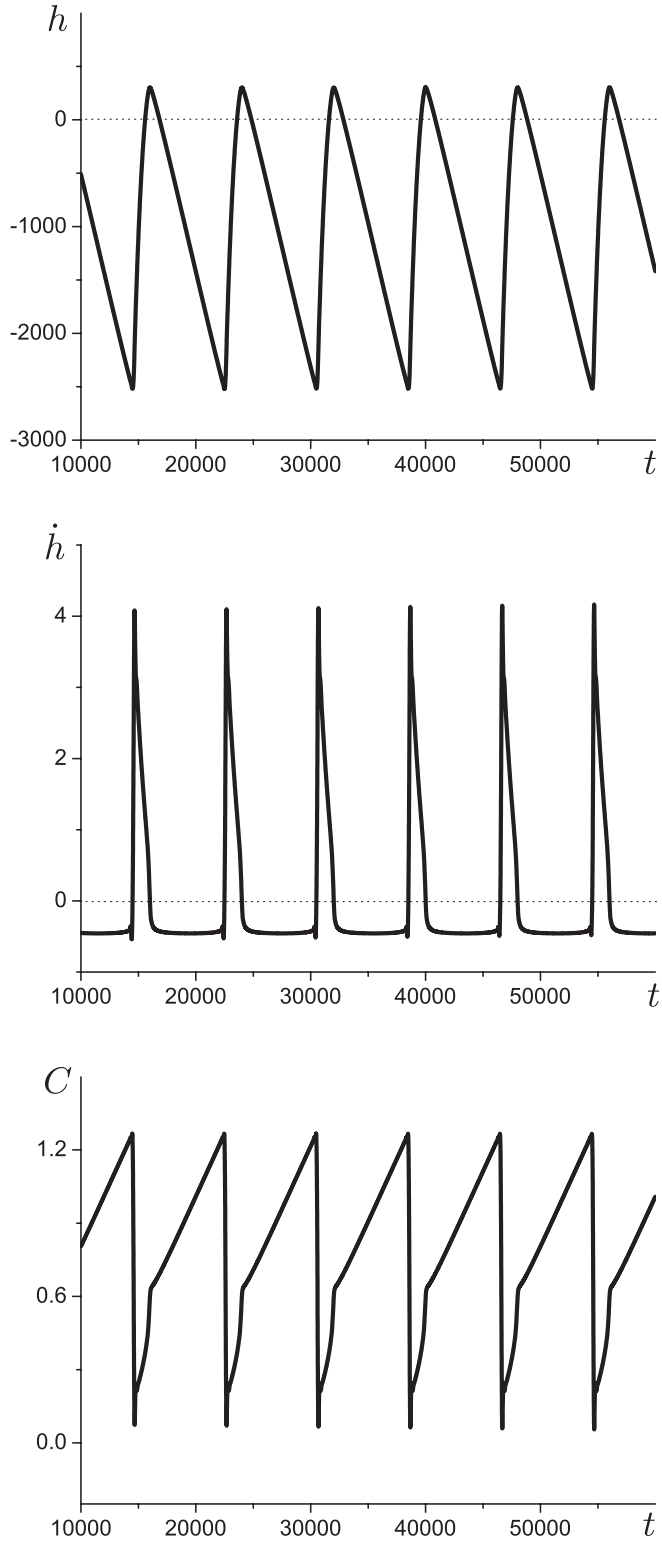

FIG. 2. Trajectories of $h(t)$, $\dot{h}(t)$, $C(Z(t), t)$ in the unstable regime for $\gamma = 0.02$, $p = 100$, $m = 0.003$, and $v_P = 0.5$.

by the Bogoliubov-Mitropolsky method [15]. To leading order one finds

$$h(t) = a(t)\cos\psi(t), \qquad (14)$$

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

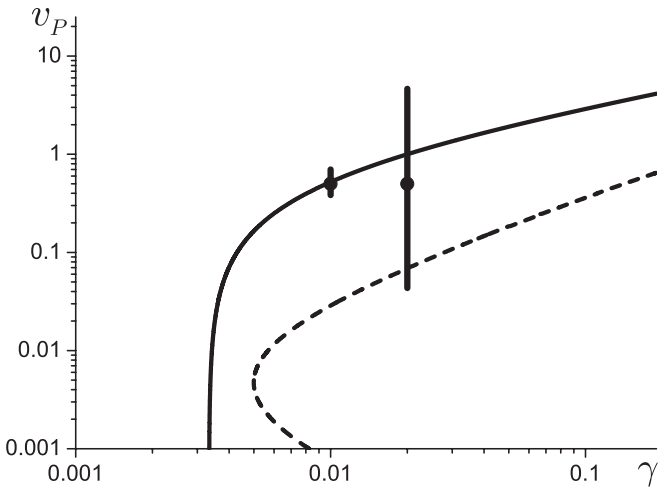

FIG. 3. Neutral-stability curves, enclosing the regions of the Cahn (solid line) and the Mullins-Sekerka instability (dashed line). The vertical lines are projections of limit cycles at $\gamma = 0.01$ and $\gamma = 0.02$, both at $p = 100$, $m = 0.003$, and $v_P = 0.5$.

$$\frac{da}{dt} = -\rho_1 a - \rho_3 a^3, \tag{15}$$

with $\rho_1 \equiv r_1(v_P - v_C)$, and the parameters r_1, ρ_3 fixed by the values of γ, p. The solution of Eq. (15) reads

$$a(t) = a_0\left[\left(1 + \frac{\rho_3}{\rho_1}a_0^2\right)\exp(2\rho_1 t) - \frac{\rho_3}{\rho_1}a_0^2\right]^{-1/2}, \tag{16}$$

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 $h(t)$, $\dot{h}(t)$, $C(Z(t), t)$ 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 $m^2 h(t)$. The pronounced oscillations of the solute concentration at the interface $C(Z(t), t)$ reflect the appearance of solute bands. From the curves in Fig. 2, which in part are remarkably close to the findings by Conti in Ref. [10], one concludes that the high- and low-concentration layers are connected by large-acceleration segments, explaining the sharpness of the interfaces between these layers.

In order to explore the possible formation of dendritic ripples, one has to consider perturbations of the form $h(\mathbf{x}, t) = \hat{h}(\mathbf{q}, \omega)\exp(i\mathbf{q}\cdot\mathbf{x} + \omega t)$ in a three-dimensional version of the model (1). In view of the almost stationary behavior of $\dot{h}(t)$ in Fig. 2 at low velocities, we choose, as an approximation, $\dot{Z}(t) = v_P$ as a reference velocity. Following Ref. [13], we then find the dispersion relation

$$\frac{\omega}{p} + q^2 + m^2 - v_P[G_P(v_P + \lambda) - G_P(v_P)] = \frac{\lambda^2 - q^2}{v_P + 2\lambda}[G_P(v_P + \lambda) + G_P(\lambda)], \tag{17}$$

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 the 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 nonplanar 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.

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