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DYNAMICS OF AN INTERFACE COUPLED TO A DIFFUSIVE BULK MODE

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We consider the dynamics of an interface in the two-phase region of two component systems where the order parameter is coupled to a non-ordering, diffusive bulk mode. Equations of motion for the soft modes, appropriate for low temperatures, are presented. Studying the dispersion relations in the linear regime, we find $\omega \propto q^2$ or q^3, depending on a certain symmetry between the phases.

In a two component system displaying phase co-existence at temperatures far below criticality ($T << T_c$, e.g., water and vapor at room temperature), the most prominent fluctuation is that associated with the interface separating the phases. Though it seems to have a life of its own, the interface is, in fact, a collective mode, deriving its properties from those of the bulk.

Since there are obviously much fewer interfacial degrees of freedom than the bulk ones, we could first ask how the former are picked out and then investigate the effects of the latter. The answer to the first question lies in the recognition that all fluctuations, except interfacial ones, are massive for $T<T_c$. Therefore, all, except the interface, are suppressed in the $T\to 0$ limit. The effects of these massive modes may be absorbed into an effective hamiltonian ($\mathscr{H}_{\text{eff}}$) for the interface. In principle, $\mathscr{H}_{\text{eff}}$ can be derived from the microscopic hamiltonian for the entire system, by 'summing out' the massive modes.

For example, one could start with the Lenard-Jones potential between molecules and try to arrive at an effective hamiltonian for the interface between a liquid and its vapor. Similarly, beginning with an Ising model with ferromagnetic nearest neighbor coupling, one might attempt to take the $T\to 0$ limit of a system in the two phase region and, in principle, come up with an $\mathscr{H}_{\text{eff}}$ for the in-

terface between the + and − phases. In practice, however, a crucial intermediate step is taken. By coarse graining, one identifies an order parameter and its associated Landau-Ginzburg hamiltonian. From this starting point, it is feasible to derive an effective interface hamiltonian, which consists of a leading term with a universal, geometric form[1,2]

$$\mathscr{H}_{\text{eff}} = \sigma \cdot (\text{area of interface}). \tag{1}$$

Here, σ is the surface tension, the only parameter that depends on the details of bulk properties. The word 'leading' indicates that there are higher order universal, geometric terms[3,2] and that Eq. (1) has limited validity. To be specific, it should be a good approximation in the hydrodynamic regime, where the correlation length ξ is short compared to the wavelength λ of the fluctuations. Their ratio, which is basically a typical curvature times ξ, supplies us with the small parameter in our systematics. Perhaps because of its universality or perhaps because of its contents being intuitively clear, the form (1) has been used long before the development of microscopic models of interfaces.

Along these lines, we could ask how interfacial dynamics is governed by the dynamics of the bulk. Since there is a variety of dynamical models all leading to the same statics, it is not clear that there would be 'universal equations of motion' (one for each model) in much the same way that (1) is independent of the bulk details. Indeed, the symmetries responsible for the universal form (1) are not the only ones in dynamics. In the study of critical dynamics, for example, there are many models[4] with identical static exponents but distinct dynamic ones. We will concentrate on models *A*, *B* and *C* in Ref. 4.

For two of these dynamic models (*A* and *B*), distinct equations of motion for the interface were derived[5–9] by taking the limit T and $\xi/\lambda \to 0$. For the simplest case of pure relaxational dynamics (*A*), the critical dynamic exponent was found by renormalization group methods, near one (bulk) dimensions.[6] In this letter, we report studies on a third model (*C*), where ϕ, the non-conserved order parameter is coupled to m, a conserved, non-ordering field (e.g., energy density). The equations of motion for the bulk are

$$\frac{\partial \phi}{\partial t} = -\Gamma \frac{\delta \mathscr{H}}{\delta \phi} \text{ and } \frac{\partial m}{\partial t} = \Lambda \nabla^2 \frac{\delta \mathscr{H}}{\delta m}, \tag{2}$$

where $\mathscr{H}(\phi, m)$ is the Landau-Ginzburg hamiltonian

$$\mathscr{H}[\phi, m] = \int \mathrm{d}^d\mathbf{x} \{\tfrac{1}{2}(\nabla\phi)^2 + U(\phi) + \tfrac{1}{2}[m - W(\phi)]^2\} \tag{3}$$

with U representing a potential energy for ϕ and W the coupling between ϕ and m. No special form of these potentials are assumed, except that, to describe two phase co-existence, U should have two degenerate minima. Also, in (2), we have

assumed constant Onsager coefficients Γ and Λ. In general, both would depend on ϕ and m. In a subsequent paper, we will explore the consequences of such dependence. To study stochastic (thermal) fluctuations, we must add Langevin noise terms and specify their distributions.

Following the approach of statics and other dynamical models, we consider first the classical configuration describing phase co-existence at $T < T_c$, i.e., $\phi = \phi_c(\mathbf{x})$ and $m = m_c(\mathbf{x}) \equiv W(\phi_c)$. As usual, $\phi_c(\mathbf{x}) = \phi_c(u)$ is a function of only one of the d coordinates, varying from one minima of U to the other as u varies from $-\infty$ to $+\infty$. A flat interface has a constant normal, $\mathbf{n}$, so that $u = \mathbf{x}\cdot\mathbf{n}$ is just one of the d Cartesian coordinates. For low temperatures, $\phi_c' \equiv d\phi_c/du$ is expected to be sharply peaked at the position of the interface, chosen to be $u=0$. Due to universality,[2] the precise form of ϕ_c is not important, while only $\sigma \equiv (\phi_c', \phi_c') \equiv \int(\phi_c')^2\, du$ will be relevant.

To study the dynamics near such an equilibrium configuration, we extract, from the fluctuations of ϕ, the Goldstone mode, i.e., perturbations of the interface from flatness. It is best taken into account by the method of collective coordinates.[10,11] The simplest way to implement this[2] is to regard u as a normal coordinate which depends on the interface configuration. Connected to this are the $d-1$ other coordinates, denoted by $\mathbf{v}$, which label points *along* the interface. For nearly planar configurations, it is common to let $\{\mathbf{v}\}$ be points on the flat equilibrium interface.

All other fluctuations of ϕ will be denoted by $\{\eta\}$. To ensure that they are orthogonal to the collective coordinate, $(\phi_c', \eta) = 0$ is imposed.[1] The fluctuations of the diffusive field are taken into account by $\chi \equiv m - W$, with $\chi_c = 0$.

Proceeding to the low temperature and long wavelengths limit, we expect $\{\eta\}$ to be massive (hard) and 'frozen', so that they can be dropped from the equations of motion. In other words, we will concentrate on the low frequency behavior ($\omega \to 0$) of the system. An inextricable coupling between interfacial and diffusive degrees of freedom is derived. Since the diffusive modes are always soft, while the interface mode is soft by virtue of being Goldstone, we should not be surprised by their coupling.

Since these modes couple, an essential complication, absent from the two models considered previously, arises. Our equations will couple fields depending on both $\mathbf{v}$ and $\mathbf{x}$, which are variables in $d-1$ and d dimensions, respectively. Like the previous models, the equations here are both non-linear[5,7] and non-local,[8,9] typical features due to a spontaneously broken symmetry and the coupling of interfacial and bulk fluctuations. The results are

$$\partial_t u = \Gamma\kappa - (\Gamma/\sigma)\,(m_c', \chi) \qquad \text{and} \tag{4a}$$

$$(1 + W'\,QGQW')\,\partial_t\chi + m_c'\,\partial_t u = \Lambda\nabla^2\chi\,. \tag{4b}$$

We define and comment on various quantities in these equations:

(a) $\partial_t u$ is the (normal) velocity of the interface.

(b) κ is the mean curvature.

(c) (m_{c_x}', χ) is a projection of χ onto a component 'enslaved' to the interface, since m_c' is the spatial derivative of m_c and so, localized there. In the absence of ϕ-m coupling, this is zero, so that (4a) reduces to the Allen-Cahn equation.[5]

(d) $W' \equiv \partial W/\partial \phi_c$, to be regarded as a function of **x**.

(e) Q is an operator that projects out the Goldstone mode, i.e., $QF \equiv F - (\phi_c', F)\,\phi_c'/\sigma$.

(f) QGQ is the inverse of the operator $-\nabla^2 + \partial^2 U/\partial \phi_c^2$, restricted to the subspace of the massive modes. So, it is non-singular. This is the source of non-locality, arising from eliminating the massive modes. We neglected ω compared to these masses, since we are interested in the $\omega \to 0$ limit. Note that $\partial_t u$, κ and (m_c', χ) are functions of **v**, 'living' on the interface. In contrast, χ depends on **x** which involves the bulk.

In the classic Stefan problem[11] and within the context of diffusion-limited crystal growth,[12] equations similar to (4a,b) appear. Significant differences exist, however. First, for their specific problems, the choice $W = \phi$ is natural. Unfortunately, this automatically excludes other possibilities (the symmetric, $\delta m_c = 0$, case below). Second, their main concern is interfaces moving with a steady velocity and its origin, while our interest is behavior of fluctuations near a flat equilibrium interface. It is possible that, in a co-moving frame, our equations can be applied to that problem. Third, in their study, the effect of the hard modes are completely ignored. Finally, we will also consider noise.

To see how the Langevin noises and their distributions enter into our equations, it is convenient to write $\mathcal{J}$, the (effective) dynamic functional. Indeed, to arrive at the deterministic part, we started with the dynamic functional for the entire system.[13] Expanding in the vicinity of ϕ_c and m_c, we identify the hard and soft modes and 'sum over' the hard ones. The result is

$$\mathcal{J} = \sigma \int \sqrt{g}\,\mathrm{dv}\,\mathrm{d}t\,\{-\Gamma \tilde{f}^2 + \tilde{f}\,[\partial_t u - \Gamma\kappa + (\Gamma/\sigma)\,(m_c', \chi)]\}$$

$$+ \int \mathrm{dx}\,\mathrm{d}t\,\{\Lambda \tilde{m} \nabla^2 \tilde{m} + \tilde{m}[(1 + W'QGQW')\,\partial_t \chi - \Lambda \nabla^2 \chi - m_c' \partial_t u]\}\,, \tag{5}$$

where $\tilde{\phi}$ and $\tilde{m}$ are the Martin-Siggia-Rose response fields[14] associated with the bulk, while $\tilde{f} \equiv (\phi_c, \tilde{\phi})/\sigma$ is the projection onto the interface mode. Note that $\sqrt{g}\,\mathrm{dv}$ is the invariant area element. Apart from displaying the noise terms and their distributions, $\mathcal{J}$ is particularly well suited for demonstrating the underlying (Galilean) symmetry in the system, spontaneously broken by the interface and re-

alized non-linearly in our equations. Respecting this symmetry is not merely an academic interest. A non-linear theory is generally not renormalizable without some symmetry that is typically well camouflaged.

While the geometric properties of the interface are manifest in our equations, Eqs. (4a,b) are hardly transparent displays of the dynamics. To remedy this situation and to appreciate some of the physics hidden in these equations, we consider the linearized version, valid for an almost planar interface. From here, we may derive 'dispersion' relations, $\omega_i(q)$, which govern the rate of dissipation of small amplitude disturbances with wave vector q. The subscript i labels various eigenmodes. Since we retain only the soft modes to arrive at (4a,b), all the ω's will vanish with q. In particular, far from the interface, we expect to find the pure diffusion modes, with $\omega \propto q^2$. However, our main interest here is the small q behavior of a mode involving the interface. To our surprise, we find its ω to follow either q^2 or q^3, depending on the *symmetry in the bulk mode under an exchange of the phases.* To see this behavior, we will take a route slightly different from the one described above. The advantage of this approach lies in its simplicity. The disadvantage is that it is too crude to yield $\omega(q)$ for all $q<<1/\xi$. Nevertheless, it is sufficient to predict the different powers, the existence of a crossover and the symmetry controlling such effects. In a later paper, we will present $\omega(q)$ in full.

Since we are interested only in linear response, the relevant equations could also be derived from (2) directly.[15,16] Of course, the massive modes will still be present in this approach. However, for the Goldstone mode, a variational principle may be used to estimate ω for small q. The previous result,[15] $\omega \propto q^2$, was obtained under some unnecessarily restrictive assumptions. Relaxing these but following the same route, we arrive at

$$-i\omega \simeq 2\Lambda\sigma q^3/[\delta m_c^2 + O(q)], \tag{6}$$

where $\delta m_c \equiv m_c(\infty) - m_c(-\infty)$, which can be non-zero provided there is some asymmetry between the two phases. Within the context of our models, an asymmetry could arise from an asymmetric U, or W, or both. However, we see that the precise details are not important and only δm_c is relevant. Indeed, we can conceive $\delta m_c = 0$ due to some conspiracy between an asymmetry in U and the one in W. This type of 'universality' is on the same footing as that associated with σ in the static $\mathscr{H}_{\text{eff}}$.

In the long wavelength limit, Eq. (6) reduces to $\omega \propto q^3$, unless $\delta m_c = 0$, when it gives $\omega \propto q^2$. Indeed, the assumption made previously[15] amounts to $\delta m_c = 0$. The coefficient of q in the $O(q)$ term, though not reliable in this approach, remains positive even when the system is symmetric. Thus, it provides a lengthscale, λ_c, for crossover behavior. Examining (6), we see that $\lambda_c \propto \delta m_c^{-2}$.

How can one best understand the importance of an asymmetry in m_c for such

drastically different behaviors? We appeal first to a similar difference between the interface dynamics in models A and B, namely, q^2 and q^3, respectively. The key to the higher power, and therefore slower relaxation, lies in the conservation of the order parameter. A localized 'bump' on an interface cannot relax by changing the associated ϕ locally. The bulk must be involved, slowing the process down further. Armed with this precedent, we can draw an intuitive picture for model C, in which there is a conserved field, even though the order parameter is not. Now, the interface is associated with both, through m_c and ϕ_c. However, if m_c is symmetric, then its conservation law does not 'enslave' the bulk to the interface. To explicate this point, consider a finite system, with a flat interface dividing equal fractions of the two phases. If the dynamics is of the model A type, the interface can be displaced as a whole while the fractions of the phases will be altered. In model B dynamics, this displacement is impossible due to the conservation law. For our case, ϕ is not conserved. But in the bulk, m_c must assume the values appropriate for each phase. If these are the same ($\delta m_c = 0$), the interface can be displaced, without disturbing the total m in the system. If $\delta m_c \neq 0$, such a displacement would clearly violate the conservation law.

Note that an asymmetry is necessary for $\omega \propto q^3$ in model B also, in the sense that $\delta\phi_c$ must be non-zero. However, we cannot even consider the possibility of its being zero, since we are dealing with systems composed of two *distinct phases.* Thus, a symmetric U (e.g., the usual ϕ^4 theory, the Ising model, etc.) provides us with an asymmetric ϕ_c. It is natural, in such models and in appropriate physical systems, to assume a symmetric W as well, so that $\delta m_c = 0$. So, although $\delta m_c = 0$ is not 'generic', physical symmetries at the microscopic level (e.g., a uniaxial antiferromagnet) may force it on us. For these, we should expect $\omega \propto q^2$ always. However, for many systems, e.g., liquid-gas and binary alloys, we should anticipate $\omega \propto q^3$ generically. In particular, $W = \phi$ for the Stefan and crystal growth problems,[11,12] so that $m_c = \phi_c$ and δm_c can never be zero.

Before discussing possible experimental support for (6), let us summarize our understanding. There are two relevant lengths in our systems: ξ and λ_c ($\propto \delta m_c^{-2}$). Both depend on T and diverge at T_c. So, in the critical regime, where $1/q$ is small compared to both, we should not expect to see the effects of λ_c. Instead, we expect only $\omega \propto q^2$, the usual critical behavior of model C (at this level where noise and fluctuations are not yet included). However, in the $T \to 0$ limit, ξ should vanish while λ_c should remain positive. Now, there are two regimes, $q\lambda_c > 1$ and $q\lambda_c < 1$, in which we predict $\omega \propto q^2$ and $\omega \propto q^3$, respectively. Like σ, λ_c does not depend on the details of (2), but only on a specific combination of ϕ_c', m_c and the transport coefficients. We will give the precise formula for λ_c in a later publication.

Though we are not aware of an experiment which verifies (6) directly, we may point to some indirect evidence. In the study of dynamics of first order transitions,[17] two types of asymptotic growth laws predominate. Associated with the average size (L) of a growing domain as a function of time t, these laws

state:[5,18] $L \propto t^{1/2}$ and $L \propto t^{1/3}$. They apply to systems in which the order parameter is *not* conserved and conserved, respectively (i.e., models *A* and *B*). Although there is no rigorous connection between a growth law and the dispersion relation, there are arguments[19] for an intimate connection. Coming from linear stability analysis about a planar interface, both express a relationship between a time and a length. Based on this argument, we expect the growth of large clusters in systems with model *C* dynamics to obey a $t^{1/2}$ or a $t^{1/3}$ law, depending on the symmetry found above.

Such dynamical behavior of tricritical systems is supposedly well described by model *C*. From theoretical analysis,[20] a $t^{1/3}$ growth law was predicted. In a recent numerical study,[21] the $t^{1/3}$ law was observed. Both appear to start with a completely *symmetric* $\mathscr{K}$. There is no cause for alarm, however, since the relevant co-existence here is between an ordered ($\phi \neq 0$) and a disordered ($\phi = 0$) phase, leading to $\delta m_c \neq 0$. Our prediction is that, if an asymmetry is introduced into $\mathscr{K}$ and 'tuned' appropriately to give $\delta m_c = 0$, the growth will obey a $t^{1/2}$ law. It would be interesting to check this in numerical studies, though it would be difficult to arrange such conspiracies in physical systems. For experimental verification of our theory, it is far better to measure directly decay rates of nearly planar interfaces in systems with natural symmetries and compare them with rates in asymmetric systems.

In this brief letter, we presented the equations of motion for an interface, coupled to a bulk diffusive mode. A variational estimate shows that the dispersion relation for the linearized equations, in the long wavelength regime, displays very different behavior, depending on a certain symmetry between the phases. We argue on physical grounds that this should be expected. Using the variational method is facile but may be unreliable. To be certain of its prediction, one must carry out a full analysis. In another publication, we will discuss in detail several topics: inclusion of field dependent transport coefficients, effects of summation over the hard modes, realization of the full Galilean symmetry, linearized equations of motion and derivation of the complete dispersion relation associated with the lowest mode.

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