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Free-energy functionals at the high-gradient limit

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It is shown that free-energy functionals have a unique infinite-gradient limit which assures a finite interaction energy. This limit is used to extrapolate the Ginzburg-Landau small-gradient theory. The resulting functionals allow the existence of cusped equilibria or equilibria with *sharp* interfaces. If perturbed, a sharp interface will not quench immediately, but rather dissolve within a finite time.

The essence of the present communication is as follows. We demonstrate that the finiteness of the interaction energy together with the requirement of well-behaved dynamics (in a sense to be clarified shortly) of a displaced system, suffice to determine *uniquely* the infinite-gradient limit of the free-energy functional. Interpolating between this limit [attained across a sharp interface(s)] and the classical, low-gradient counterpart yields a meaningful extension of the Ginzburg-Landau form for the free energy. The new form allows for equilibrium states with genuinely sharp interfaces. Alternatively, an out-of-equilibrium initial state endowed with sharp interfaces will not quench immediately. It will take a *finite* time to resolve it (see Fig. 1 for an example).

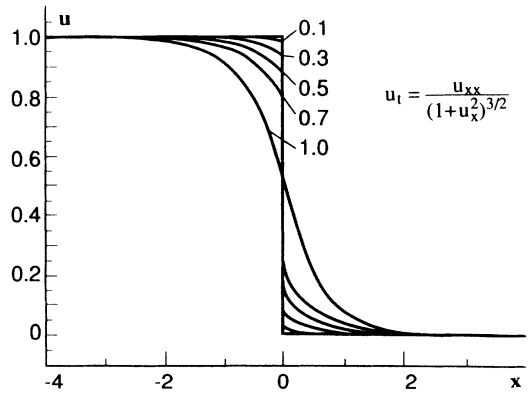

FIG. 1. Spatiotemporal evolution as given by Eq. (11b) of an initially sharp front with $p(s)=(1+s^2)^{1/2}-1$. Shown are profiles $u(x,t_i)$ at fixed times t_i. As explained in the text, the sharp front persists until the height of the discontinuity reaches zero.

We start with the free energy $F(u)$ in its classical form

$$F[u]=\int f(u)dx\,, \tag{1a}$$

$$f(u)=f_0(u)+\tfrac{1}{2}au_x^2,\ a>0\,; \tag{1b}$$

$f_0(u)$ is the energy density of the system in a local homogeneous equilibrium and its inhomogeneity, explicitly assumed to be small, it is measured via the gradients. This is the essence of the Ginzburg-Landau theory in its maximal, one-dimensional simplicity.[1] If large gradients occur, form (1b) cannot be correct. The split between the homogeneous and the interacting part is not guaranteed and certainly the interacting part cannot grow quadratically in gradients, as this must yield unbounded energy across a sharp interface or alternatively such interfaces must be excluded. The separation between the self-energy and the interacting part seems logically possible, only if the free energy, due to the interacting part, stays bound for arbitrarily large gradients. In other words, we can generalize (1b) to large gradients

$$f(u,u_x)=f_0(u)+P(u_x)\,, \tag{2}$$

only if $\int P(u_x)dx$ stays bound for arbitrary gradients. Taking that as the first prerequisite for representation (2) to make sense, what can one say about $P(u_x)$? The essence of the present communication is to show that when $s\to\infty$, $P(s)$ has a unique asymptotic characterization. This follows from the requirement that

$$P''(s)\geq 0 \text{ for all } s\,, \tag{3a}$$

i.e., $P(s)$ is convex and

$$0<sP'(s)-P(s)<\infty,\ \text{as } s\to\infty\,. \tag{3b}$$

Note that in the classical case $sP'-P=as^2/2$ [see Eqs.

(1b) and (2)]. Conditions (3) imply that

$$P(s) \sim P_0 s + P_1 s^{-\sigma} \text{ as } s \to \infty . \tag{4}$$

P_0, P_1 are constants and $\sigma > 0$.

To see how requirements (3) arise naturally, calculate the variation of F with respect to f given in Eq. (2). The equilibrium states are then given via the variation $\delta F/\delta u = 0$, or explicitly

$$Lu \equiv P''(u_x) u_{xx} - f_0'(u) = 0 . \tag{5}$$

Clearly, we need $P''(u_x)$ to always be non-negative. Otherwise equilibria calculated via (5) become unstable. This becomes self-evident if we look at

$$u_t = Lu , \tag{6}$$

which describes the dynamics of an out-of-equilibrium state. If $P''(u_x)$ changes sign, Eq. (6) describes the evolution of instability due to backward diffusion. Returning to Eq. (5), note that it has a first integral of motion given as

$$\{H\} - f_0(u) = E_0 , \tag{7a}$$

where

$$\{H\} \equiv u_x P'(u_x) - P(u_x) . \tag{7b}$$

Now the nature of equilibrium solutions crucially depends upon whether expression (7b) is bound as $|u_x| \to \infty$. If it is not, as in the case of the classical theory, all ensuing equilibria will be smooth functions of x. On the other hand, if the $\{H\}$ is bound from above, one can construct equilibria with cusps and sharp transitions. As an example, take $P(s) = (1+s^2)^{1/2} - 1$ which will be considered in more detail shortly. Not surprisingly the relation between P and $\{H\}$ is exactly that of a Lagrangian and Hamiltonian.

Now assume for a moment that only condition (3a) is satisfied. Then if $P(s)$ is strictly superlinear (though subquadratic), the interaction energy remains infinite across a sharp interface. It is only at the lowest limit admissible by (3a), as stated explicitly by (3b) [or asymptotically in (4)], that the total energy across a jump is finite and proportional to the height of the jump. It is exactly in this distinguished limit, if attained by P, that Eq. (7) predicts the possibility of new equilibria, precluded in the classical theory. Note that actually one can relax condition (4) on P, and allow the constant P_0 (and P_1 as well) to be some function of u, say $P_0 = J'(u)$. Then the energy content due to the jump becomes $J(u_2) - J(u_1)$, and assumption (2) on the separated form of free energy is not essential for the arguments presented here to hold. It is the asymptotic behavior of the gradients, as given by Eq. (4), that is essential to preserve the boundedness of the energy across a sharp interface.

Before we consider the dynamic consequences of condition (4) on P, we need to take one more step: to select P. The classical arguments provide its form for small gradients. With the results we have just obtained, we now have the other extreme—the large gradient limit. The simplest approach, due to lack of better knowledge of P is to interpolate between these two regimes. An attractive choice is to select

$$P(u_x) = (1+u_x^2)^{1/2} - 1 , \tag{8}$$

which affords the interpretation of P as an interfacial energy due to surface tension. In our approach the choice (8) does not play a fundamental role and an infinite number of other choices that satisfy condition (4) are possible. Relation (8) just seems to be the simplest one. Thus, inasmuch as different choices of P will result in a difference in intermediate wavelengths, since the upper and lower limits are fixed, the consequences due to these differences cannot be too large.

Note that forms such as (8) can incorporate exactly two pieces of information from the small gradient limit. That is, if in the expansion of $P(s)$ for small s,

$$P(s) = as^2/2 - bs^4/8 + \cdots , \tag{9}$$

we have access to both a and b, then rescaling s by $\beta = (|b/a|)^{1/2}$ and letting $u \to \beta\phi$ modifies (8) to make it coincide with (9) up to $O(s^6)$. The corresponding f will read

$$f(\phi) = f_0(\phi/\beta) + \alpha[(1+\phi_x^2)^{1/2} - 1] , \tag{10}$$

with $\alpha = a^2/b$. One might be tempted to say that the form

$$P(s) \approx \frac{a^2 s^2/2}{1+\beta^2 s^2/4}$$

should also do the trick; it saturates as $s \uparrow \infty$ and coincides with (9) in the small s limit. But it does not preserve convexity [condition (3a)] and must therefore be excluded.

Perhaps the most dramatic effect caused by the imposition of condition (4) is observed during a dynamic approach to equilibrium via (6). Focusing our interest on the interacting part of f only, we set $f_0 \equiv 0$ and rewrite Eq. (6) as

$$u_t = \frac{\partial}{\partial x} P'(u_x) . \tag{11a}$$

The choice of P given in Eq. (8) leads to

$$u_t = \frac{\partial}{\partial x}\left(\frac{u_x}{(1+u_x^2)^{1/2}}\right) . \tag{11b}$$

It is now natural to view $-P'(u_x)$ as a flux of u, which according to Eq. (4) is monotone, bound and saturates at the value P_0 as $|u_x|$ diverges. One thereby departs from Fick-type dependence as the gradients grow to the point of flux saturation. The finiteness of flux across a sharp interface signals that such a surface does not dissolve immediately but takes a finite time to do so. In fact, it was shown by us elsewhere[2] that given Eq. (11b) and initial conditions of the form

$$u(0,x) = \begin{cases} u_1 & \text{for } x < 0 \\ 0 & \text{for } x > 0 . \end{cases} \tag{12}$$

The initial evolution follows the self-similar pattern

$$u(t,x) = \begin{cases} u_1 - \sqrt{t} f(z) & \text{for } x < 0 \\ \sqrt{t} f(z) & \text{for } x > 0 , \end{cases} \tag{13}$$

where $z = x/\sqrt{t}$ and $f(z)$ is found via the solution of a

boundary value problem

$$2[P'(f_z)]_z = -zf_z + f \text{ for } 0 < z < \infty, \tag{14a}$$

with

$$f(z)\downarrow 0,\ f_z(z)\downarrow 0 \text{ as } z \to \infty, \tag{14b}$$

$$-f_z(z) \to \infty \text{ as } z \downarrow 0, \tag{14c}$$

and $f(0)$ playing the role of a nonlinear eigenvalue.

The similarity solution sustaining a sharp jump pertains until the height of the discontinuity $h(t) = u_1 - 2f(0)\sqrt{t}$ has decayed to zero (at $t = t_D \equiv [u_1/2f(0)]^2$). Thereafter the smooth solution pattern is very much like a classical one and is no longer given by the presented similarity solution. A numerical example of such a process is displayed in Fig. 1 ($u_1 = 1$). Conversely, it was also shown by us[2] that if an initial jump is not to be diffused immediately, P must be of the form (4). Recall that within the framework of classical diffusion a sharp front would smooth out immediately in order to avoid having infinite energy.

Using the choice in (8) we can further advance our knowledge of the equilibria. Using (7b)

$$\{H\} = 1 - \{1 + u_x^2\}^{-1/2}. \tag{15}$$

We rewrite (7a) as

$$\left[\frac{du}{dx}\right]^2 + V(u) = 0, \tag{16}$$

where

$$V(u) = 1 - \frac{1}{[E_1 - f_0(u)]^2} \equiv \frac{N(E_1,u)}{D(E_1,u)}, \tag{17}$$

and $E_1 = 1 - E_0$.

It is now patently clear that infinite gradients appear in a very natural way, if D has a root(s) between two consecutive roots of N. The roots of D, whether simple or double, occur at a finite x and correspond to a weak singularity; u remains finite. The corresponding equilibrium solution may be periodic with cusps or simply have kinks in a finite domain. To obtain a kink in an infinite domain, E_1 has been chosen so that $N(E_1,u)$ will have a double root. To be more specific, take

$$f_0(u) = -\beta u^2 + \gamma u^4. \tag{18}$$

Then the appropriate $V(u)$ reads

$$V(u) = -\gamma(u^2 - u_0^2)^2 \left[\frac{[2 - \gamma(u^2 - u_0^2)^2]}{[1 - \gamma(u^2 - u_0^2)^2]^2}\right], \tag{19}$$

where $u_0^2 = \beta/2\gamma$ and $E_1 = 1 - \beta^2/4\gamma$. $V(u)$ has double poles $u_{p\pm}^2$ and additional roots $u_{N\pm}^2$ located at

$$u_{p\pm}^2 = u_0^2 \pm 1\sqrt{\gamma},\ u_{N\pm}^2 = u_0^2 \pm \sqrt{2/\gamma}, \tag{20}$$

respectively. A typical graph of the effective potential $V(u)$ is shown in Fig. (2) where the domain relevant for a kink solution is $-u_0 \le u \le u_0$, with $\pm u_0$ being a double root of $V(u)$ corresponding to a point(s) at $\pm$ infinity.

Now if $E_1 > 0$, then $u_{p-}^2 < 0$ and $V(u)$ is finite in the domain of interest. Its graph [C_k in Fig. (2)] is a smooth function and so is the kink solution. This is essentially a classical kink.

If $E_1 \le 0$ then $u_{p-}^2 \ge 0$ and $V(u)\downarrow -\infty$ at $\pm u_{p-}$. The graph of V now has three branches: C_A, C_p, and C_B. A kink smooth everywhere is impossible. One possible solution is a partially smooth kink with a vertical jump in the profile connecting the two singular points $\pm u_p$ [$C_A \to$ jump $\to C_B$ in Fig. (2)]. Another possibility is a continuous solution that assumes vertical slopes at $\pm u_{p-}$ ($C_A \to C_p \to C_B$). The profile of this solution has an inflection point at $u = 0$. Therefore, if the analogy with fluid dynamics holds, such a profile should be linearly unstable. Besides, this solution is possible only for $V(0) < 0$ (the case displayed in Fig. 2). For $V(0) \ge 0$ only the discontinuous solution is admissible. Observe from Fig. 2 that as $|u_{p-}|$ decreases so does the height of the jump. When $|u_{p-}| = 0$, $E_1 = 0$, the C_p branch disappears and the singularities at $\pm u_{-p}$ coalesce. The kink now has a vertical slope at $u = 0$. Otherwise it is a "classical kink."

Though the stability of the kinks described is as yet unknown, the richness of the possible structures as compared with the classical theory is indeed notable. Besides kinks, from Fig. 2 one observes that the trajectory following $u_0 \to u_{p+} \to u_{N+} \to u_{p+} \to u_0$ corresponds to a stationary soliton. Its profile assumes a maximum at $u = u_{N+}$

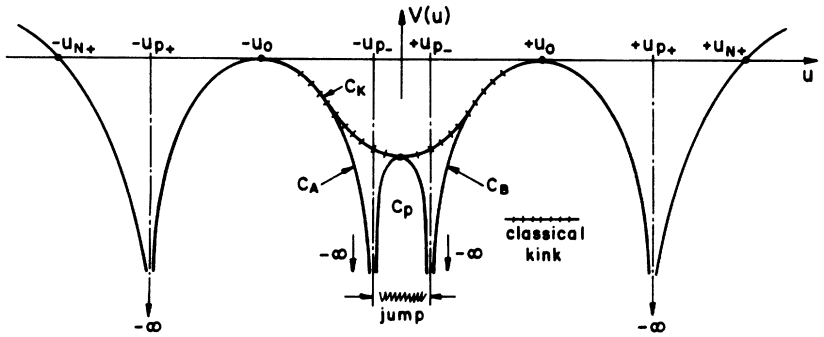

FIG. 2. Qualitative graph of the effective potential. Trajectory connecting $-u_0$ with $+u_0$ corresponds to a kink in an infinite domain. A continuous branch, C_k, yielding a classical kink is possible only for $E_1 > 0$. For $E_1 < 0$, a transit from branch C_A to C_B is via a jump. The corresponding part of the kink has a perfectly vertical profile.

and a vertical slope at two points where $u = u_p$.

Finally, consider the possibility of a nonclassical, ultimately sharp kink; a piecewise constant solution connecting the two equilibrium points $\pm u_0$ in (18), i.e.,

$$u = \begin{cases} +u_0, & x > x_0 \\ -u_0, & x < x_0 . \end{cases}$$

To this end we need $V(\pm u_0) = -\infty$. This is achieved simply by taking $E_1 = -\beta^2/4\gamma$. The location of x_0, the transit point between the two phases, is fixed by the constraint on the totality of the order parameter, i.e., $\int u\,dx = M$. Of course, one can construct an equilibrium with an arbitrary number of transitions between $\pm u_0$. As yet, the stability of such states is unknown, and we shall therefore not pursue this point any further.

A final note is now in order. In our recent work[3] we have treated a complementary case wherein the microscopic interaction was quadratic but the pitfalls of the continuum mean-field limit were avoided by preserving a trace of the mesoscopic scale. It was shown that the way to accomplish this is to replace the interacting part (u_x, u_x) with $(u_x, \mathcal{L}_A u_x)$, where $\mathcal{L}_A$ is a Lorentzian operator, isotropic for fluids and anisotropic if the interaction takes place on a lattice. Incorporating $\mathcal{L}_A$ into the free energy suffices to temper the growth of the gradients at infinity. In the present work, although we stay within the realm of continuum, we have addressed a more realistic problem because general interactions cannot typically be approximated as quadratic.

In summary, we have extended the Ginzburg-Landau free-energy functionals to include interaction due to high gradients. The requirement of finite interacting energy imposes a strict limit on the asymptotic growth of $P(u_x)$, the interaction energy density. Using this restriction, one can construct a useful interpolative approximation between the small gradient, classical limit, and the infinite gradient regime. We have described some dynamical and static consequences of a particular interaction form. The multidimensional extension should be considered as the next testing ground for the present approach.

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