ARTICLES

Dispersion relation around the kink solution of the Cahn-Hilliard equation

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The dispersion relation of the long-wavelength fluctuations of an interface exhibited by the Cahn-Hilliard equation is studied analytically and numerically. The expected asymptotic dispersion relation $\omega \sim k^3$ is demonstrated. Further, using a well-defined microscopic length scale ξ, the dispersion relation is numerically found to have a nearly universal form $\omega/k^3=(1/\xi)\Omega(k\xi)$ for a wide variety of potentials.

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I. INTRODUCTION

The asymptotic-growth law of the average pattern size L in the spinodal decomposition process of binary alloys (critical quench case) seems to be given by $L \sim t^{1/3}$. Actual experimental results [1] and computational studies [2,3] both strongly suggest that this is the universal asymptotic relation. Analytic derivations for this law have been proposed, but unfortunately they are not well controlled mathematically [4].

In the present report, we give a well-controlled result about the dispersion relation of the waves on the interface in d-dimensional ($d \geq 2$) space, and then empirically demonstrate extensive universality found for this dispersion relation. We will demonstrate that the spectrum of the linearized Cahn-Hilliard operator around its kink solution has an eigenvalue $\omega=f(k)$ which is continuous with respect to k and $\lim_{k\to 0}\omega/k^3$ exists and is a nonzero constant. In this limit the essential spectrum of the linearized operator and the Nambu-Goldstone mode of the interface (i.e., the zero eigenvalue of the operator) are not separated. Thus some care is required in applying perturbation theory to the eigenvalue problem. We justify Jasnow and Zia's study [6] of the same problem which assumed that the essential spectrum is totally harmless.

Next, a numerical result for the dispersion relation for a wide range of $|\mathbf{k}|$ and for a wide variety of potentials is given with the aid of a formula for the eigenvalue which explicitly separates the $|\mathbf{k}|^3$ factor. Empirically, we find that there is a universal form to the dispersion relation independent of the potential, and that it is close to but distinct from the form found for an analytic solution to a special potential by Jasnow and Zia. We also compute for comparison the dispersion relation for a less singular potential which is also exact up to the determination of the root of a transcendental equation. The result is indistinguishable from the numerically obtained universal form.

If we may assume that there is only one representative length scale other than the thickness of the interface (i.e., the microscopic correlation length), then these results imply that the $t^{1/3}$ asymptotic law is exact, and that the universality in the dispersion relation implies the universality of the form factor stressed in [5].

In Sec. II, the long-wavelength asymptotic dispersion relation is demonstrated for the Cahn-Hilliard equation. Despite the essential spectrum of the linearized operator, perturbational approach is justified. In Sec. III, an extensive universality in the dispersion relation is demonstrated. Section IV is for concluding remarks.

II. LONG-WAVELENGTH DISPERSION RELATION

Perhaps the most appealing analytical argument for the $\frac{1}{3}$-power law growth is based on the kinetics of the interface [7]. The dimensional analysis of the equation of motion for the evolution of interfaces gives the $\frac{1}{3}$ power law [8]. However, no one has been able to give a mathematically well-controlled derivation of the interface equation from the Cahn-Hilliard equation [9]. The difficulties are well documented in [10]. If we may assume as in the dimensional analysis that there is only one relevant length scale as mentioned above, then the relation between the block copolymer equilibrium lamellar thickness and the spinodal asymptotic growth law can also be established [11]. Furthermore, if we assume the universality, then we can devise an exactly solvable model for the block copolymers, which implies the exact $\frac{1}{3}$ power law [12]. The same idea was also used in [6] to get a $|\mathbf{k}|^3$ dispersion exactly (under the above-mentioned implicit assumption).

We start with the following equation:

$$\frac{\partial\psi}{\partial t}=\Delta[-\psi+\psi^3-(\partial_z^2+\alpha\Delta_{\parallel})\psi]\ , \tag{2.1}$$

where z would become the coordinate perpendicular to the interface, $\Delta_{\parallel}$ is the Laplacian in the subspace perpendicular to the z axis, and $\alpha\in[0,1]$. When $\alpha=1$, the equation is the standard Cahn-Hilliard equation. The kink solution centered around the origin perpendicular to

the z axis is given by $\psi_0=\tanh(z/\sqrt{2})$. Setting $\psi=\psi_0+u_{\mathbf{k}}(z)\exp[i\mathbf{k}\cdot\mathbf{r}]$, where $\mathbf{r}$ denotes the coordinates perpendicular to the z axis, we linearize (2.1) around ψ_0,

$$\frac{\partial u_{\mathbf{k}}(z)}{\partial t}=-D_k L_k(\alpha)u_{\mathbf{k}}(z)\ , \tag{2.2}$$

where $D_k\equiv-\partial_z^2+k^2$ ($D_0\equiv-\partial_z^2$), $L_k(\alpha)\equiv L_0+\alpha k^2$, and $L_0=-\partial_z^2+f(z)$ with $f(z)=2-3/\cosh^2(z/\sqrt{2})$ [henceforth we write $|\mathbf{k}|$ as k; $\mathbf{k}$ is a $(d-1)$-dimensional vector]. The boundary condition is that $u_{\mathbf{k}}(z)$ and $\partial_z u_{\mathbf{k}}(z)$ both vanish at infinity. The spectrum of L_0 is well known [13]: it consists of two eigenvalues zero and $\frac{3}{2}$ and the remaining essential part well separated from zero. We denote the normalized eigenfunction of L_0 corresponding to zero by u_0. We consider, for simplicity, all the operators in the intersection of $\mathcal{L}_2$ and the domain of ∂_z^4 on the real axis (denoted by $\mathcal{D}$), which is dense in $\mathcal{L}_2$ (henceforth $\mathcal{L}_p$ implies the set of all the functions f on the real axis such that $|f|^p$ is integrable). Furthermore, we may regard all the operators closed or closable.

We wish to ultimately study the spectrum of the linearized Cahn-Hilliard equation,

$$\lambda_k u_k=D_k L_k(1)u_k\ , \tag{2.3}$$

particularly the eigenvalue branch connected to $\lambda_{k=0}=0$. We will henceforth use $\lambda\equiv\omega$ to describe the dispersion relation. However, one cannot simply assume that the above continuous branch exists or that even if it does, it is attainable as a perturbation of the operator D_0L_0. In fact, a difficulty does appear at $k=0$. Physically, the interface in the conserved system cannot move freely using the Goldstone mode as it could in the nonconserved countepart represented by the linearized operator L_k. Thus, at $k=0$, the system can only satisfy conservation if it "ripples" transversely to the interface. Such states would likely be members of the essential spectrum. At $k\neq0$, a perturbation local to the interface of a form like the Goldstone mode is possible, since conservation is taken care of by the fluctuation along the interface.

We consider the spectrum of $D_kL_k(\alpha)$. The existence of an eigenvalue might follow from that for $L_k(\alpha)$, or might be inferred from the variational formulation as done by Jasnow and Zia. However, notice that the variational formulation can be used to get the eigenvalue only when we know there is one. Before demonstrating the dispersion relation, we will show that the lower limit of the spectrum is indeed an eigenvalue.

Notice that $D_k u=0$ implies $u=0$ in our problem, so that $G_k\equiv D_k^{-1}$ is a strictly positive bounded self-adjoint operator as can be seen from the explicit form of its kernel as an integral operator,

$$\tilde{G}_k=\frac{1}{2k}e^{-k|z-z'|}\ . \tag{2.4}$$

The set of the eigenvalues of $D_kL_k(\alpha)$ and the set of λ which makes $\lambda G_k-L_k(\alpha)$ not one-to-one are obviously identical thanks to the uniqueness of the solution to $G_k u=0$. The essential spectrum σ_{ess} of $D_kL_k(\alpha)$ and the set S of λ which makes $\lambda G_k-L_k(\alpha)$ one-to-one but $[\lambda G_k-L_k(\alpha)]^{-1}$ unbounded are again the same. Since $\|[\lambda-D_kL_k(\alpha)]^{-1}\|\le\|G_k\|\,\|\lambda G_k-L_k(\alpha)\|$, $S\supset\sigma_{\text{ess}}$. Suppose $\mu\in S$, then there exists a sequence $\{v_n\}$ such that $\|v_n\|=1$ and $[\mu G_k-L_k(\alpha)]v_n=u_n$ converges to zero. This implies that $[\mu-D_kL_k(\alpha)]v_n$ also converges to zero. Since G_k and $\mu G_k-L_k(\alpha)$ are one-to-one, $[\mu-D_kL_k(\alpha)]$ must also be one-to-one. The existence of such a sequence implies that $[\mu-D_kL_k(\alpha)]^{-1}$ is not a bounded operator. Hence μ must be in σ_{ess}, so $S\subset\sigma_{\text{ess}}$.

Consider the following variational formula:

$$\mu=\inf_\psi\frac{\langle\psi,L_k(\alpha)\psi\rangle}{\langle\psi,G_k\psi\rangle}\ . \tag{2.5}$$

Since $L_k(\alpha)$ is nonnegative definite, and G_k is positive definite, $\mu\ge0$. For $\forall\epsilon>0$ there exists ψ such that

$$\mu+\epsilon>\frac{\langle\psi,L_k(\alpha)\psi\rangle}{\langle\psi,G_k\psi\rangle}\ . \tag{2.6}$$

If the dimension of the set of all such ψ is finite, then this μ must be an eigenvalue of $D_kL_k(\alpha)$ (at this point we cannot assert that it is the smallest one). If it is infinite, then μ must not be smaller than the lower limit of the essential spectrum of $D_kL_k(\alpha)$. [The proof is akin to that for theorem XIII.1 (min-max principle) in Reed and Simon [16], with the aid of the fact that $L_k-\mu G_k$ is self-adjoint and non-negative, so its square root exists.] Using $u_0(z)\propto\psi_0'(z)$ we may see for $k<1$ that $\mu\le k^3\langle u_0^2\rangle/\langle u_0(z),\int dz'\exp(|z-z'|)u_0(z')\rangle$.

The essential spectrum of $D_kL_k(\alpha)$ is identical to that of the operator $D_k(-\partial_z^2+2+\alpha k^2)$, since the coefficients to the differential operators have the same limit as $z\to\pm\infty$ (see proposition 26.2 in Collet and Eckmann [14]). Hence the essential spectrum of $D_kL_k(\alpha)$ lies outside the disk of radius $2k^2$ around the origin for any k and α (>0). Consequently, for sufficiently small $k>0$, μ obtained above cannot be the lower limit of the essential spectrum. Hence $D_kL_k(\alpha)$ has an eigenvalue, which is positive, and is bounded by μ, from above. In particular, it cannot be larger than of order k^3.

We have demonstrated that the eigenvalue problem

$$\lambda_k u=D_kL_k(\alpha)u \tag{2.7}$$

is meaningful. Using the fact that $L_k(\alpha)$ is self-adjoint, and $L_k(\alpha)u_0=\alpha k^2u_0$, we can solve (2.7) as

$$\lambda_k=2\alpha k^3\left[\frac{\int_{-\infty}^{+\infty}dz\,u_0(z)u_{k,\alpha}(z)}{\int_{-\infty}^{+\infty}dz\int_{-\infty}^{+\infty}dz'u_0(z)e^{-k|z-z'|}u_{k,\alpha}(z')}\right], \tag{2.8}$$

where $u_{k,\alpha}$ is the corresponding eigenfunction of $D_kL_k(\alpha)$. Incidentally, it should be noted that the contribution to the dispersion of the overall Laplacian (i.e., the Laplacian needed to impose the conservation law) in the Cahn-Hilliard equation is k and not k^2. Using the variational principle above, we can show that the eigenfunction of the lowest eigenvalue of $D_kL_k(\alpha)$, which is not self-adjoint, does not have any node; the proof is

analogous to that given in Courant-Hilbert [17] Chap. 6, Sec. 6.

We wish to demonstrate that actually $\lambda_{k,\alpha}$ depends on k continuously, and the $k\to 0$ limit of the square-bracketed quantity in (2.8) exists and equal to that value at $k=0$. We wish to use a perturbative approach to this end starting from the $k=0$ case. There is, however, one obstacle for our approach: zero is not only an eigenvalue but also the lower limit of the essential spectrum of D_0L_0. We can avoid this difficulty, noting the following. $L_0u=0$ implies $D_kL_0u=0$, so zero is an eigenvalue of the operator D_kL_0 in $\mathcal{D}$ for any k (and α). Thus zero is an isolated point spectrum of D_kL_0 for $k\neq 0$. We choose D_kL_0 as the unperturbed starting point.

We wish to get $D_kL_k(1)$ perturbatively from D_kL_0, but the *a priori* estimate of the perturbation we can get is of order k^2, so we cannot ignore the essential spectrum whose lower limit is $\sim 2k^2$. To avoid this difficult we decompose the perturbation into sufficiently small perturbations, so that we may use the stability theory of the spectrum with a simple *a priori* estimate of the magnitude of the perturbation. For some $\alpha=1/N$ we find the existence of an eigenvalue of $D_kL_k(1/N)$ proportional to k^3/N. Hence, for small k, the perturbed spectrum is still well inside a disk, of radius, say, k^2 and we may continue to perturbed the operator further. Our demonstration below of the dispersion relation is well controlled with the one assumption that no other point spectrum that we consider comes near the disk of radius k^2 centered at the origin for any $\alpha\in[0,1]$.

Define $L_k^{(1)}=L_k(1/N)$ and $L_k^{(n+1)}=L_k^{(n)}+k^2/N$. The perturbation

$$(D_kL_k^{(n+1)}-D_kL_k^{(n)})=\frac{k^2}{N}D_k \tag{2.9}$$

can be demonstrated to be $D_kL_k^{(n)}$ bounded, for any $N\geq n\geq 0$, that is $\|D_ku\|_p\leq b\|D_kL_k^{(n)}u\|_p+a\|u\|_p$. b is a constant freely adjustable to any value $b>0$ and a is a function of b. Fixing $b<1$, we may find a bound for the gap (Chap. IV, theorem 2.14 in Kato [15]), between $D_kL_k^{(n+1)}$ and $D_kL_k^{(n)}$, $\hat{\delta}(D_kL_k^{(n+1)},D_kL_k^{(n)})\leq(1-bk^2/N)^{-1}(a^2+b^2)^{1/2}k^2/N$, which is bounded by a positive number proportional to k^2/N. a and $b<1$ can be found and set independent of n, N, or k. Note that $L_k^{(0)}\equiv L_0$.

Let Γ be a circle of radius ck^2 centered about zero, where c is say $\frac{1}{2}$. The disk whose boundary given by this circle is called $\mathcal{U}$. Our standing hypothesis is that there is some neighborhood of the disk $\mathcal{U}$ into which no eigenvalue of the operator $D_kL_k(\alpha)$ moves from outside for any $\alpha\in(0,1]$. This appears to be true empirically. If this is not the case, we cannot successfully use the argument below without additional estimates on the behavior of the eigenvalue deep inside the essential spectrum.

For Γ and the operator $D_kL_k(\alpha)$ there is a $\delta(\alpha)$ (>0) for which any gap between this operator and its perturbed version smaller than $\delta(\alpha)$ ensures that the original spectrum and the perturbed spectrum are separated into like parts by Γ (Sec. IV, theorem 3.16 in Kato [15]). $\delta(\alpha)$ can be computed as $\min_{\zeta\in\Gamma}(\frac{1}{2})[1+\|R(\zeta)\|^2]^{-1/2}(1+k^2)^{-1}$, where $R(\zeta)$ is the resolvent $[D_kL_k(\alpha)-\zeta]^{-1}$. Since resolvent are continuous functions of closed operators (Chap. IV, theorem 3.15 in Kato [15]), $\delta(\alpha)$ is a continuous function of α. Suppose for given k, which is sufficiently small and fixed, $\delta(\alpha)>0$ for $\alpha<\alpha_0$, and $\delta(\alpha_0)=0$. Choose an arbitrary small positive number ϵ. There is δ (>0) such that $\delta(\alpha)>\delta$ for all non-negative $\alpha<\alpha_0-\epsilon$. Consider

$$\lambda_k^{(1)}u_k^{(1)}=D_kL_k^{(1)}u_k^{(1)}\ . \tag{2.10}$$

Using the previous obtained estimate for $\hat{\delta}(D_kL_k^{(1)},D_kL_0)\leq ck^2/N$, we set N large enough to satisfy the inequality $\hat{\delta}\leq\delta$. Fixing that N, we may now safely perturb D_kL_0 (Sec. IV, theorem 3.16 in Kato [15]). More importantly, by the same theorem, the eigenprojections $P^{(1)}$ to the eigenspace of $\lambda_k^{(1)}$ converges in the L_p norm $(p\geq 1)$ to P_0, which selects the eigenspace belonging to the zero eigenvalue of D_kL_0 as $N\to\infty$ or $k\to 0$ since the bound on the gap is proportional to k^2/N.

Corresponding to (2.8), we have

$$\lambda_k^{(1)}=\frac{2k^3}{N}\left[\frac{\int_{-\infty}^{+\infty}dz\,u_0(z)P^{(1)}u_0(z)}{\int_{-\infty}^{+\infty}dz\int_{-\infty}^{+\infty}dz'u_0(z)e^{-k|z-z'|}P^{(1)}u_0(z')}\right]. \tag{2.11}$$

We know that the square bracketed quantity is bounded from above by a number M independent of k for sufficiently small k, since the eigenvalue is the one bounded by the variational estimate for any α. Hence the perturbed eigenvalue is still well inside the disk $\mathcal{U}$.

The rest is taken care of by a mathematical induction. We consider

$$\lambda_k^{(n)}u_k^{(n)}=D_kL_k^nu_k^{(n)}\ . \tag{2.12}$$

For n we may assume that $\lambda_k^{(n)}=n\sqrt{2}k^3/3N+o(k^3)$. The gap [15] between $D_kL_k^{(n+1)}$ and $D_kL_k^{(n)}$ can be bounded identically to the $n=0$ case. The eigenvalue $\lambda_k^{(n)}$ we are interested in for $D_kL_k^{(n)}$ is of order k^3. Hence it is far away from the essential spectrum of $D_kL_k^n$. Since $L_k^{n+1}u_0=[(n+1)k^2/N]u_0$, we arrive at a formula analogous to (2.11),

$$\lambda_k^{(n+1)}=\frac{2(n+1)k^3}{N}\frac{\int_{-\infty}^{+\infty}dz\,u_0(z)P^{(n+1)}u_k^{(n)}(z)}{\int_{-\infty}^{+\infty}dz\int_{-\infty}^{+\infty}dz'u_0(z)e^{-k|z-z'|}P^{(n+1)}u_k^{(n)}(z')}\ , \tag{2.13}$$

where $P^{(n+1)}$ is the eigenprojection for $\lambda_k^{(n+1)}$ which converges in the $\mathcal{L}_p$ norm to $P^{(n)}$ in the k zero limit. The quantity in the square brackets is bounded from above as before, by a number M independent of k and n.

Up to $N'=[N(\alpha_0-\epsilon)]$ we can use the same estimates needed in the above stepwise procedure. In this way we demonstrate that the eigenvalue close to zero of $D_k L_k(\alpha)$ is $\sim k^3$ up to $\alpha_0-\epsilon$ for any ϵ (>0). That is, $\lim_{\alpha\to\alpha_0-0}\lambda_k$ is still bounded from above by $2Mk^3$. Hence the norm of the resolvent is bounded from above on Γ by a number M' of α up to α_0. However, $\delta(\alpha_0)=0$, so the resolvent cannot be bounded on Γ. This violates the continuity of the resolvent. Hence actually α_0 must be larger than 1. We may continue our argument up to $\alpha=1$ for small enough k.

In this way we arrive at

$$\lambda_k \equiv \lambda_k^{(N)} = 2k^3 \left[\frac{\int_{-\infty}^{+\infty} dz\, u_0^2(z)}{\left[\int_{-\infty}^{+\infty} dz\, u_0(z)\right]^2} + o(1) \right] . \tag{2.14}$$

Hence we may conclude that $\lambda_k \simeq \sqrt{2}k^3/3$ in the small k limit for the (normalized) Cahn-Hilliard equation. As is seen in the next section, the formula implies $\lambda_k \propto k^3/\xi$, where ξ is the correlation length (interface thickness) of the system as expected dimension analytically. Since ξ is for our problem a cutoff length scale, so the exponent 3 instead of 4 may be said to be due to the anomalous dimension [18].

Notice that the above argument does not use the detailed shape of the potential function in the standard Cahn-Hilliard equation. Of course, u_0 depends on the detail, but the existence of a finite limit of the square bracketed quantity in (2.13) in the $k\to 0$ limit does not.

III. EMPIRICAL UNIVERSALITY

We have established the form of the isolated point spectrum λ_k, for small k and the corresponding eigenfunction $u_k(z)$ in $\mathcal{L}_1\cap\mathcal{L}_2$ for sufficiently small k. The formula for the dispersion relation can be written as

$$\lambda_k = k^3 R(k) , \tag{3.1}$$

where $R(k)$ is defined by

$$R(k) = 2\frac{\int_{-\infty}^{+\infty} dz\, u_0(z)u_k(z)}{\int_{-\infty}^{+\infty} dz \int_{-\infty}^{+\infty} dz' u_0(z) e^{-k|z-z'|} u_k(z')} , \tag{3.2}$$

with $u_k(z)$ being the eigenfunction with the smallest modulus eigenvalue of $D_k L_k$. The formula is numerically very convenient. As noted at the end of Sec. II, the formula can be justified for any potential function (or the chemical potential formula μ) which gives physically realistic interface.

In order to investigate the dependence of the dispersion relation to the details of the free-energy functional, we study

$$\frac{\partial\psi}{\partial t} = \Delta[-D\Delta + \mu(\psi)] . \tag{3.3}$$

We can easily construct the linear operator corresponding to Eq. (2.2). The corresponding eigenvalue problem is

$$\lambda_k u_k = D_k L_k[\mu] u_k , \tag{3.4}$$

where $L_k[\mu] \equiv D(-\partial_z^2 + k^2) + \mu'(\psi_0)$, ψ_0 being the kink solution. Note that $u_0 \propto \psi_0'$.

One may numerically compute the eigenvalues and eigenfunctions for $D_k L_k[\mu]$ by discretizing the operators over a large, finite domain and solving the resulting matrix eigenproblem. In general, however, it is very difficult to see the small k behavior of the dispersion relation due to the k^3 behavior. Numerical inaccuracy will swamp any result at small k. However, to use (3.2), only modestly accurate eigenfunctions are necessary. In fact, most of the dispersion-relation behavior should arise from knowledge of $u_0(z)$ since we believe that $u_k(z) \approx u_0(z) + o(k)$ in norm, with $\|u_k\|_p = 1$ for any k. This is empirically checked below.

We numerically determine the eigenfunctions as functions of k for various systems with various local free energies. We graph in Fig. 1, for qualitative comparison, the various free energies we consider including the piecewise double parabolic potential used by Jasnow and Zia. It is easier to represent the chemical potential $\mu = \delta F/\delta\psi$. We list the various potentials (A)–(D) we will study, as well as the piecewise parabolic (PP) potential,

$$(A)\ \mu = -\psi + \psi^3 , \tag{3.5}$$

$$(B)\ \mu = -\psi + \psi^2 + \psi^3 , \tag{3.6}$$

$$(C)\ \mu = \begin{cases} -\psi & \text{if } |\psi| < 0.8 \\ 4\psi - \mathrm{sgn}(\psi)4 & \text{otherwise} , \end{cases} \tag{3.7}$$

$$(D)\ \mu = \begin{cases} -\psi & \text{if } |\psi| < 0.95 \\ 19\psi - \mathrm{sgn}(\psi)19 & \text{otherwise} , \end{cases} \tag{3.8}$$

$$(\mathrm{PP})\ \mu = \psi - \mathrm{sgn}(\psi) . \tag{3.9}$$

We compute results for the above with $D=1$. Additionally we compute the (C) potential with $D=0.5$ and 2. In Fig. 2, we show the various numerically determined kink

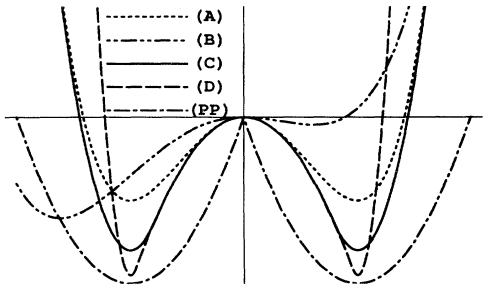

FIG. 1. A comparative plot of the different local free-energy functions used to generate the different potentials discussed. The line style of the curves are labeled with the letter code of the corresponding potential described in the text, where PP denotes the piecewise parabolic potential.

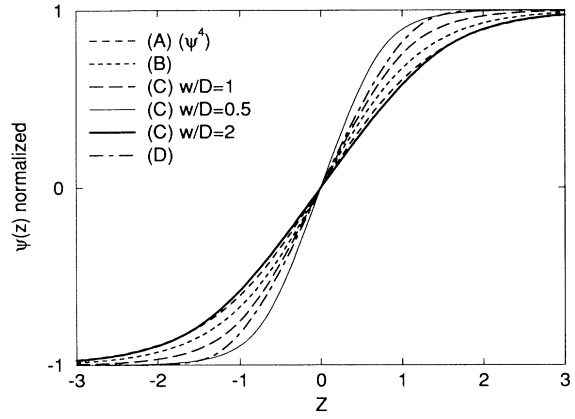

FIG. 2. Equilibrium wall profiles generated by the different potentials studied numerically. The line style of the curves are labeled with the letter code of the corresponding potential described in the text. The (*C*) potential was studied with three different *D*: 1, $\frac{1}{2}$, and 2. All the other potentials were studied with $D=1$.

or equilibrium wall solutions, normalized in the case of potential (*B*) so that ψ ranges from $+1$ to -1 for comparison. Note that although the (A)–(D) systems have identical initially unstable wavelengths, their wall widths vary over a wide range. We note in passing that the wall profiles nearly collapse onto each other by empirically rescaling the z coordinate by dividing by a $\xi_{\text{empirical}}$, the width of the wall from $\psi=\pm 0.9\psi_{\max}$.

We calculate the dispersion-relation factor $R(k)$ from the eigenfunctions for each potential. For the ψ^4 potential (*A*), we may calculate various quantities explicitly. The unnormalized $u_0=1-\tanh^2(z/\sqrt{2})$. Thus we may find $R(0)=\sqrt{2}/3$ as already mentioned.

We expect $u_k(z)=u_0(z)+o(k)$ and this appears to be confirmed for potential (*A*) from the numerically determined eigenfunctions by Fig. 3. In this figure, we compute the $\mathcal{L}_2$ distance of u_k from u_0 where the various u_k are normalized so that the $\mathcal{L}_2$ norm is a fixed constant. It is curious that the distance rises as k^2 until roughly $k\sim 0.3$. In Fig. 4, we see that the numerically determined eigenfunctions u_k vary somewhat over the range of $k\in[0,2]$. However, the form of the eigenfunction does not seem radically different.

We replot the eigenfunctions, which must be nonnegative as mentioned before, in Fig. 5 on a linear-log plot. While we dismiss the behavior beyond $|z|>8$, due to the finite size of the computational domain, we see that for small k, the eigenfunctions deviate from $u_0(z)$ in the large z tails. These tails rise until they appear to hit a maximum deviation at about $k\sim 0.4$ at which point the eigenfunctions appear to relax to a fixed high k form which is outlined by the $k=3.8$ eigenfunction. Thus this explains the odd behavior of the $\mathcal{L}_2$ distance. We note that the behavior of the eigenfunctions is consistent with the observation that the order parameter tends to overshoot the bulk equilibrium value in the region within a curved domain wall.

The general numerical procedure was to use an adaptive general purpose ordinary differential equation solver to get the kink solution. Boundary conditions were fixed to be of the bulk equilibrium values. About 1000 mesh points were used over a range of $z\in[-10,10]$. This solution was then interpolated onto a fixed mesh size array before used in a 384×384 matrix eigenvalue problem. The discretized Laplacian operator was of the $1:-2:1$ form. Although the computations were done on a finite domain, we found that cutting down the range of z to $z\in[-5,5]$ or using $\frac{1}{2}$ the number of grid points were generally unobservable.

In the large k limit, we see that $e^{-k|z-z'|}\sim 2k^{-1}\delta(z-z')$, giving $\lambda_k=k^4$. Since $u_0\in\mathcal{L}_2$, we may calculate the small k behavior of $R(k)$ by expanding $e^{-k|z-z'|}\sim 1-k|z-z'|\ldots$. Integrating numerically, we get

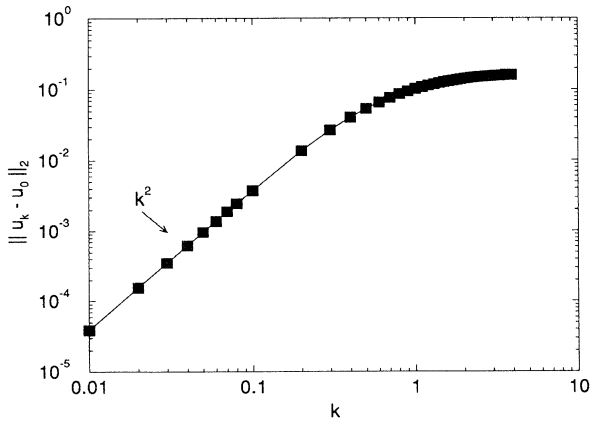

FIG. 3. Plot of the $\mathcal{L}_2$ distance between u_0 and u_k as determined numerically over a variety of k. Each of the u_k were normalized such that $\|u_k\|_2=c$, where c is a fixed common constant. Note that the norm decreases as $O(k^2)$.

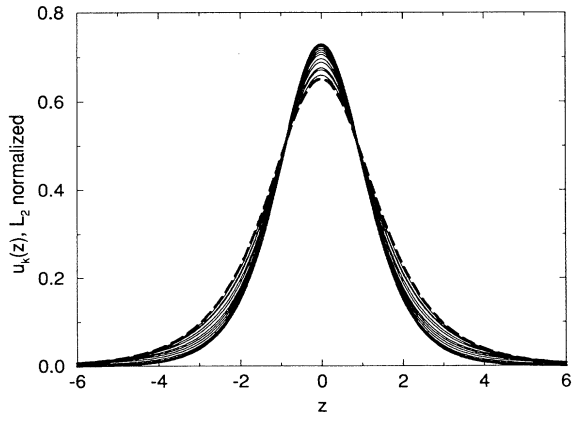

FIG. 4. A plot of numerically determined eigenfunctions for the ψ^4 potential (*A*), vs various k. The solid dark curve, which is the thinnest and tallest peak, is $k=0$. The dark dashed curve is for $k=3.8$ and represents the general form of eigenfunctions for large k. Each u_k is $\mathcal{L}_2$ normalized to a common constant.

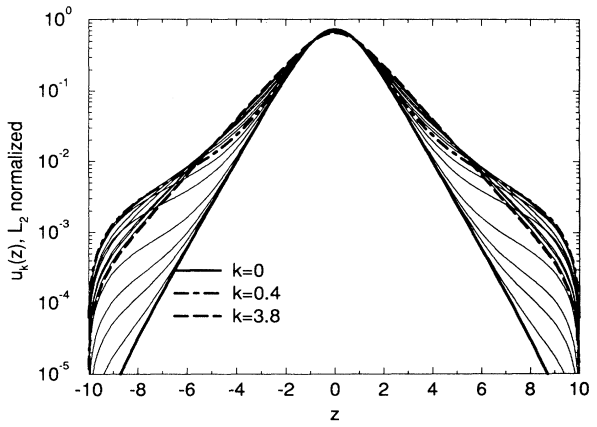

FIG. 5. A linear-logarithmic plot of numerically determined eigenfunctions for the potential (A) for various k. The dashed-dotted curve represents the eigenfunction for $k=0.4$. Note that it appears to be the maximally distorted eigenfunction. For $k>0.4$, the eigenfunctions appear to relax toward a high-k form typified by the $k=3.8$ eigenfunction which is plotted as a dashed curve.

$$\int_{-\infty}^{+\infty} dz \int_{-\infty}^{+\infty} dz' u_0(z)|z-z'|u_0(z') \simeq 11.2 \ , \tag{3.10}$$

so that $R(k) \approx \sqrt{2}/3(1+11.2k/8)$. Figure 6 shows $R(k)$ numerically calculated for the (A) or ψ^4 potential and the small k estimate. The small k estimate works well for $k<0.4$ which corresponds to the point where we saw that the eigenfunctions started to relax toward a high k form in Fig. 5.

We computed $R(k)$ for each system listed above. If we normalized each $R(k)$ by dividing by $R(0)$ and then rescaled k to $k\xi_{\text{empirical}}$, all the $R(k)$ curves nearly collapse. In fact, we may define a precise length scale as follows:

$$\xi = \frac{\left[\int_{-\infty}^{+\infty} dz\, u_0(z)\right]^2}{2\int_{-\infty}^{+\infty} dz\, u_0(z)^2} \ . \tag{3.11}$$

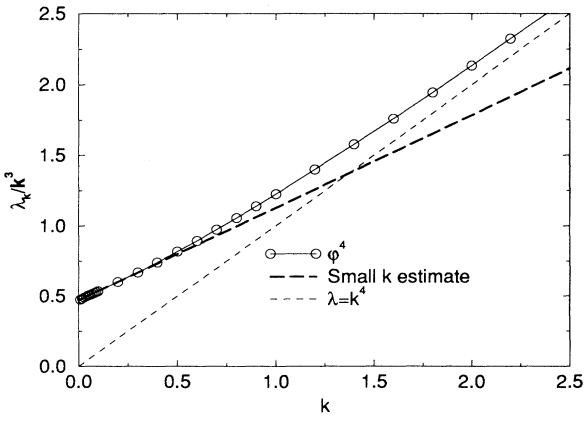

FIG. 6. A plot of the dispersion relation for the ψ^4 potential (A), vs a plot of the small k estimate to the dispersion relation and the large k behavior of $\lambda_k \simeq k^4$.

Recall that the we may define $u_0(z) \propto \partial_z \psi_0(z)$. We note that

$$\sigma = \int_{-\infty}^{+\infty} dz\, D[\partial_z \psi_0(z)]^2 \tag{3.12}$$

and

$$\Delta\psi = \int_{-\infty}^{+\infty} dz\, \partial_z \psi_0(z) \ , \tag{3.13}$$

where σ is the surface free energy (surface tension) of the planar interface and $\Delta\psi$ is the miscibility gap between the two bulk phases. Hence the length scale $\xi = \frac{1}{2}D(\Delta\psi)^2/\sigma$. This is easily computed solely from the equilibrium wall profile. If we use this ξ, by plotting $\xi R(k)$ vs $k\xi$, we find that all the dispersion relations collapse as shown in Fig. 7. Defining $\Omega(k\xi) \equiv \xi R(k)$, we find a nearly universal form $\omega/k^3 = \xi^{-1}\Omega(k\xi)$ as stated at the beginning.

Jasnow and Zia had computed the exactly solvable dispersion relation for a double parabolic local free energy. After scaling, this relation also nearly follows numerically determined relations. To see if this small difference was an artifact of our procedures, we calculated the dispersion relation analytically for potential (C). This is exact up to determining the roots of a transcendental equation numerically and is described in the Appendix. All the dispersion relations calculated numerically and exactly are shown in Fig. 7. The differences between the Jasnow and Zia dispersion relation and those for the potentials listed above persist, but are very small and at large k, and may be solely due to the singular nature of the double parabolic local free energy they used.

For small k, we note that one may write

$$(-\partial_z^2 + k^2)[-\partial_z^2 + k^2 + f(z)] = \Lambda\psi \tag{3.14}$$

by dividing through by k^4 and rescaling $zk^2 = z'$ and get

$$(-\partial_{z'}^2 + 1)\left[-\partial_{z'}^2 + 1 + \frac{1}{k^2} f(z'/k^2)\right]\psi = \Omega\psi \ . \tag{3.15}$$

However, $f(z)$ decays exponentially or faster to a con-

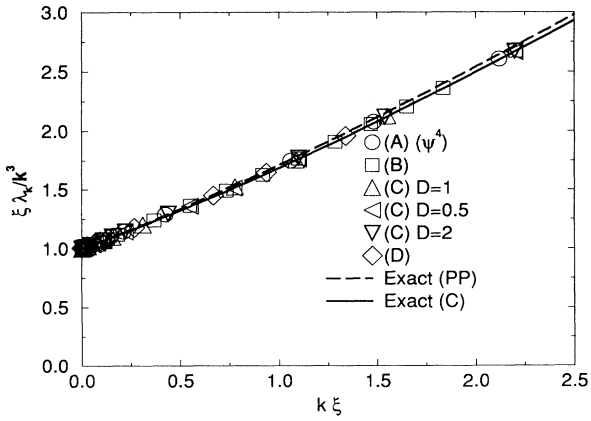

FIG. 7. A plot of the numerically calculated dispersion relations for the various potentials described in the text as well as the exact dispersion relation calculated by Jasnow and Zia for the double parabolic free energy and the exact dispersion relation for potential (C) with $D=1$.

stant, and in the limit $k \to 0$, one may reinterpret

$$\frac{1}{k^2} f(z'/k^2) \sim \frac{c}{k^2} - c'\delta(z') , \tag{3.16}$$

where $c = \lim_{z \to \infty} f(z)$ and $c' = \int_{-\infty}^{\infty} dz\,[f(z) - c]$. Thus all problems have the same small k structure for $k \ll 1/\xi$.

Hence, we suggest that there is a nearly universal form for the dispersion relation for any bistable potential when scaled by the above ξ. This dispersion relation provides us with a comparative crossover law for the long but pre-asymptotic growth of the length scale for computational simulations of spinodal decomposition. This crossover is relevant for computational simulations since it is difficult for the domain-size length scale $l(t)$ to be much greater than ξ. We will consider this further in an another paper.

IV. CONCLUDING REMARKS

We have demonstrated the asymptotic dispersion relation $\omega \sim k^3$ around the kink solution of the Cahn-Hilliard (or the conserved time-dependent Ginzburg-Landau equation), and numerically exhibited an extensive universality in this dispersion-relation independent of the free-energy functional describing the phase segregation.

The correction to the k^3 behavior in the universal dispersion curve (Fig. 7) starts immediately at $k = 0$. This explains why the true asymptotic power growth law $L \sim t^{1/3}$ is not available in computational studies. At best we can get $L \approx 5\xi \sim 10\xi$. However, the universal form factor can be well obtained before we reach the true asymptotic scaling law regime through a hardening procedure [3]. This is possibly due to the fact that the form factor is largely localized in a small region about the peak wave vector. The various structures which contribute to the form factor see approximately the same preasymptotic growth law, so that the global structure may not depend strongly on the nonasymptoticity of the growth law.

The speed of coarsening may be compared with the value of λ_k for $k\xi = 1$. From the universal curve we see that $\lambda_{1/\xi} \simeq 1.7/\xi^4$. This implies that so long as there is no numerical freezing, the potential which gives a steeper interface profile is advantageous. To avoid freezing, it appears we must have a potential whose unstable region is wide, that is, the ratio $\psi_{\rm eq}/\psi_{\rm sp}$ of the positive solution $\psi_{\rm eq}$ to $\mu(\psi) = 0$ (the equilibrium value of the order parameter) and that $\psi_{\rm sp}$ to $\mu'(\psi) = 0$ (the spinodal value) is as small as possible. A judicious choice of the potential can accelerate the simulation considerably. The result will be given elsewhere.

ACKNOWLEDGMENTS

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APPENDIX: EXACT DISPERSION RELATIONS

Jasnow and Zia [6] found an exact solution to the dispersion relation for the local free energy,

$$U(\psi) = \begin{cases} \frac{1}{2}\kappa(\phi - M)^2, & \phi > 0 \\ \frac{1}{2}\kappa(\phi + M)^2, & \phi < 0 . \end{cases} \tag{A1}$$

We briefly restate their solution to prepare for a more complicated, but smoother potential. The "potential" in the linearized equation is

$$f(z) = \kappa^2 - 2\kappa\delta(z) . \tag{A2}$$

They sought the solution to the adjoint problem, which is defined by

$$[D_k + f(z)] D_k \bar{\psi} = \Omega \bar{\psi} , \tag{A3}$$

where $\psi = D_k \bar{\psi}$.

The conditions on $\bar{\psi}$ are that the first and second derivatives are continuous at $z = 0$. Taking (A3) and integrating both sides from $+\epsilon$ to $-\epsilon$ and taking the limit $\epsilon \to 0$ they find a third condition

$$\partial_z^3 \bar{\psi} \big|_{-\epsilon}^{+\epsilon} = 2\kappa(D_k)\bar{\psi}(0) . \tag{A4}$$

They found it convenient to rewrite $\Gamma = q^4\Omega$ and $y = k^2/(2q^2)$. One may write down a solution to Eq. (A3) for $z > 0$ and $z < 0$ and match the solutions at $z = 0$ using the above three conditions. From symmetry, and the arbitrary normalization of $\bar{\psi}$, one can eliminate undetermined coefficients in favor of an equation for Ω,

$$\begin{aligned} 0 = {} & \Omega^4 - \Omega^3(2 + 2y - y^2) + \Omega^2(1 + 2y - 2y^2 - 2y^3) \\ & + \Omega(-2y^2 + 2y^3) + y^4 . \end{aligned} \tag{A5}$$

This represents the solution of Jasnow and Zia. The physically relevant root is graphed in Fig. 6.

Now let us turn to the construction of a solution for a less singular potential. We choose the following local free energy:

$$U(\phi) = \begin{cases} \frac{1}{2}\kappa_0(\phi - M)^2, & \phi > \phi_s \\ -\frac{1}{2}\kappa_i\phi^2 + C, & |\phi| \le \phi_s \\ \frac{1}{2}\kappa_0(\phi + M)^2, & \phi < -\phi_s , \end{cases} \tag{A6}$$

where C and ϕ_s are defined so that $U(\psi)$ is C^1. The corresponding chemical potential $\mu = dU/d\phi$ gives us ϕ_s by the continuity of μ,

$$\phi_s = \frac{M}{1 + \kappa_i^2/\kappa_0^2} . \tag{A7}$$

The $f(z)$ for the linearized equation is a square well of height κ_0^2 and depth κ_i^2 with walls at z_s. z_s is determined by the point where the equilibrium solution $\phi_0(z_s) = \phi_s$. One may construct the equilibrium solution from

$$-\partial_z^2\psi_0 + \mu(\psi_0) = 0 \tag{A8}$$

and find that $z_s = (1/\kappa_i)\tan^{-1}(1/\sqrt{\alpha})$ where $\alpha = \kappa_i^2/\kappa_0^2$.

One constructs the adjoint problem as before. Making

the substitution $\Gamma=q^4\Omega$, $y=\kappa_o^2/2q^2$ and we arrive at

$$(-\partial_{z'}^2+1+2y)(-\partial_{z'}^2+1)\bar{\psi}=\Omega\bar{\psi} \quad \text{for } |z'|>z_s\frac{\kappa_o}{\sqrt{2y}}$$
$$(-\partial_{z'}^2+1-\alpha 2y)(-\partial_{z'}^2+1)\bar{\psi}=\Omega\bar{\psi} \quad \text{for } |z'|<z_s\frac{\kappa_o}{\sqrt{2y}} . \tag{A9}$$

We will define $z_s'=z_s\kappa_o/\sqrt{2y}$. The explicit solution for which $\bar{\psi}\to 0$ as $z\to\pm\infty$ can be written as

$$\bar{\psi}=\exp\{-[(y+1)+(\Omega+y^2)^{1/2}]^{1/2}z'\}+B\exp\{-[(y+1)-(\Omega+y^2)^{1/2}]^{1/2}z'\} \tag{A10}$$

for $|z'|>z_s'$

$$\bar{\psi}=C\cosh\{-[(1-\alpha y)+(\Omega+y^2)^{1/2}]^{1/2}z'\}+D\cos\{-[-(1-\alpha y)+(\Omega+y^2)^{1/2}]^{1/2}z'\}+E$$

for $|z'|<z_s'$ The solution should satisfy continuity in $\bar{\psi}$, $\partial_{z'}\bar{\psi}$, and $\partial_z^2\bar{\psi}$. Applying the integration over $z_s-\epsilon$ to $z_s+\epsilon$, we see that $\partial_{z'}^3\bar{\psi}$ should also be continuous. However, we need one additional constraint. Upon examining (A10) one finds that there is a jump condition on $\partial_{z'}^4\bar{\psi}$ at $\pm z_s$ of the form

$$\partial_{z'}^4\bar{\psi}|_{z_s'-\epsilon}^{-z_s'+\epsilon}+2y(1+\alpha)(-\partial_{z'}^2+1)\bar{\psi}(z_s')=0 . \tag{A11}$$

Using this, one gets five transcendental equations with five underdetermined quantities. After tedious reduction, we arrive at a transcendental equation for Ω as a function of y. This is solved numerically for a given set of y values and plotted in Fig. 6 for $\kappa_i=1$ and $\kappa_o=2$. Note that M does not play a role in the dispersion relation.

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