LETTER TO THE EDITOR

Interface dynamics under the elastic field

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Abstract. The interface equation of motion is derived in a system in which the local order parameter is coupled with the elastic strains. As a special case, the dynamics of spherical domains growing in a metastable matrix is studied. In a non-conserved system it is shown that larger domains grow dominantly around smaller domains when the shear modulus of the new phase is larger than that of the matrix, while the size difference of domains tends to diminish when they are softer than the matrix.

The elastic effect often plays a crucial role in the kinetics of a first-order phase transition. When a system is quenched below, but near, the transition point, the high-temperature phase becomes metastable. New domains of the low-temperature phase nucleate and grow at the expense of the metastable state. In many solid materials that undergo structural phase transition and phase separation, the two phases often exhibit different lattice structures. This causes an elastic strain in both the domains and the metastable matrix, so a long-range elastic interaction evolves between the domains. Here we consider the case in which the elastic strain around a domain is coherent [1].

The theoretical study of the elastic interaction in a system with two different lattice structures has a long history. Ardell *et al* [2] calculated the interaction energy between two inclusions in a linear elastic theory. A comprehensive study of structural transitions in which the order parameter is not conserved has been given by Khachaturyan [1]. In the phase separation problem in which the order parameter is a conserved quantity, the importance of the elastic effects on the kinetics was discussed by Cahn [3]. Recently Kawasaki and Enomoto [4] have investigated, in detail, the growth dynamics of spherical domains in Ostwald ripening based on the interaction obtained by Ardell *et al*. They have studied the time evolution of the domain radius distribution for small values of the volume fraction of the new phase. One of the conclusions is that when the matrix is harder than the precipitates, the domains cease to grow. This implies that the localized domains can be made to exist stably by the elastic interaction. (In a thermodynamical sense, this state is metastable. The uniform low-temperature state has a lower free energy.) It seems, however, that no dynamical theory of domain growth under the elastic field has been available for a non-conserved system.

In this letter, we derive the interface equation of motion under the elastic field for both conserved and non-conserved systems. As a particular example, we apply the equation of motion to the growth of spherical domains.

We take the following model equations. The local order parameter field $S(r, t)$ obeys

$$\partial S/\partial t = -|i\nabla|^a \, \delta H\{S, u\}/\delta S \tag{1}$$

where $a = 0$ for a non-conserved system, $a = 2$ for a conserved system and

$$H\{S, u\} = \int dr \left[\tfrac{1}{2}|\nabla S|^2 + W(S) - \lambda S^2(\nabla \cdot u) + \frac{K}{2}(\nabla \cdot u)^2 \right.$$

$$\left. + \frac{\mu}{4}\sum_{ij}\left(\frac{\partial u_j}{\partial x_i} + \frac{\partial u_i}{\partial x_j} - \tfrac{2}{3}\delta_{ij}(\nabla \cdot u)\right)^2 \right]. \tag{2}$$

The local displacement is denoted by $u(r, t)$ whose ith component is u_i. The last two terms with the elastic moduli K and μ are the elastic energies at small deviations. Here, for simplicity, we do not consider the elastic anisotropy. The coupling constant λ is assumed to be positive without loss of generality. The function $W(S)$ is an even function of S with the absolute minimum at $S = 0$ and two local minima at $S = \pm S_0$ where S_0 is positive and finite. An example is given by $W(S) = S^2(1 - bS^2 + S^4)$ with $b \leqslant 2$.

In order to take account of the difference in elastic properties between the high-temperature phase with $S = 0$ and the degenerate low-temperature phases with $S = \pm S_0$, we allow S-dependence of K and μ whose precise forms will be given below.

We assume mechanical equilibrium for $u(r, t)$:

$$\delta H/\delta u_j = 0. \tag{3}$$

The free energy functional (2) and its variants have been introduced and studied in various problems. The gas–liquid transition of hydrogen atoms absorbed in metals has been investigated on the basis of a model similar to (2) [5]. Recently, spinodal decomposition under the elastic interaction has been studied using (1) and (3) with (2) [6, 7]. In these cases, the order parameter S is a concentration and is a conserved quantity. The form of $W(S)$ and the coupling between S and $\nabla \cdot u$ utilized in [5] and [6] are slightly different from those in (2). A one-dimensional version of (2) has been introduced to obtain a stable localized domain (but with a singularity in the spatial variation of the order parameter) in a martensitic transition such as in TiNi [8]. The order parameter in this case is the amplitude of the displacement corresponding to the low-temperature structure and hence it is a non-conserved quantity. Finally, it is noted here that the elastic effect in a commensurate–incommensurate transition in ferroelectric materials [9] has been studied in the same spirit as the model (2). Application of the free energy functional, like (2), to both conserved and non-conserved systems has also been considered [10].

Our aim is to derive the interface equation of motion in the two-phase region from equations (1)–(3). Because of the degeneracy of the ordered phase, K and μ should be even functions of S. Here we put $K = K_0 + K_1 S^2$ and $\mu = \mu_0 + \mu_1 S^2$. First we solve equation (3) to eliminate u for given $S(r, t)$. This can be performed perturbatively for small values of K_1 and μ_1. Since the calculation is straightfoward [6], we write only the results here. We have $\nabla \cdot u$ up to the first order in μ_1 and $\partial u_j/\partial x_i$ up to the lowest order as follows

$$\nabla \cdot u = \frac{\lambda S(r)^2}{L + K_1 S(r)^2} + \frac{\mu_1}{L}\sum_{ij}\int dr' \int dr'' \, G(r, r')\partial'_i\partial'_j \left(S(r')^2 M_{ij}(r', r'')S(r'')^2\right) \tag{4}$$

$$\partial u_j/\partial x_i = -\lambda \partial_i \partial_j \int dr' \, G(r, r') \frac{S(r')^2}{L + K_1 S(r')^2} \tag{5}$$

where ∂_i' means $\partial/\partial x_i'$ and $L = K_0 + 4/3\,\mu_0$. The Green function $G(r, r')$ is defined by the relation

$$-\nabla^2 G(r, r') = \delta(r - r') \tag{6}$$

and

$$M_{ij}(r, r') = -(2\lambda/L)[\partial_i\partial_j G(r, r') + (\delta_{ij}/3)\delta(r - r')]. \tag{7}$$

The results (4) and (5) are essentially the same as those obtained by Onuki [6].

Substituting (4) and (5) into (1), and after some manipulation, we obtain a closed equation for S:

$$\partial S/\partial t = -|i\nabla|^a \, \delta F\{S\}/\delta S \tag{8}$$

where $F\{S\}$ is given up to the first order in K_1 and μ_1 by

$$F\{S\} = \int dr \left[\tfrac{1}{2}(\nabla S)^2 + W(S) - \frac{\lambda^2}{2L} S^4 \left(1 - \frac{K_1}{L} S^2\right) \right]$$

$$+ \frac{\mu_1}{4} \sum_{ij} \int dr \int dr' \int dr'' \, S(r)^2 (M_{ij}(r, r')S(r')^2)(M_{ij}(r', r'')S(r'')^2). \tag{9}$$

The last term is not symmetrized with respect to the arguments r, r' and r''. Notice that there are two contributions from the elastic field. One is the λ-coupling which is local and modifies the form of $W(S)$ effectively. When $S^2 K_1/L \ll 1$, as we have assumed, and the coupling constant λ is not extremely small, the potential $W(S)$ renormalized by the λ^2 terms in (9) takes a form such that the two local minima at $S = \pm S_0$ turn out to be stable absolute minima while the state with $S = 0$ becomes metastable. Hereafter we use the same notation $\pm S_0$ for the locations of the new minima. The other contribution arises from the S-dependence of the shear modulus, which gives rise to the long-range non-local interaction of $S(r)$ as the last term in (9). We note from (5) that if $S(r)^2 = S_0^2$, $\partial u_i/\partial x_j = 0$ for $i \neq j$ and the last term in (9) vanishes, as expected, in the uniform low-temperature state.

Now we consider the kinetics of domain growth in the low-temperature phase $S = \pm S_0$. Initially the system is supposed to be the metastable uniform state with $S = 0$. As the decay of the metastable state proceeds, domains emerge and grow. A sharp interface is constituted between the domains and the surrounding region. If we identify $S(r)^2$ with the local concentration $C(r)$, then equation (8) with (9) can be applied to spinodal decomposition in binary solids [5, 6].

Using the method developed previously [11], we readily obtain from (8) the interface equation of motion for a non-conserved case:

$$V(a) = H(a) + c + E(a) \tag{10}$$

where $V(a)$ is the normal component of the interface velocity at point a on the interface and $H(a)$ the mean curvature. The last term $E(a)$, which comes from the elastic interaction, is given by

$$E(a) = -\frac{\mu_1}{2\sigma}(S_0)^6 \sum_{ij} \int_D dr' \int_D dr'' \left[M_{ij}(r(a), r')M_{ij}(r', r'') \right.$$

$$\left. + \tfrac{1}{2} M_{ij}(r(a), r')M_{ij}(r(a), r'') \right] \tag{11}$$

where σ is the interfacial tension. The integrals with respect to r' and r'' run over all the domains. The constant c in equation (10) is proportional to the free energy difference between the metastable and the stable states, whose value is given if we specify the form of the renormalized potential. In the conserved case [11], the left-hand side of (10) is replaced by

$$(S_0^2/\sigma) \int \mathrm{d}a' \ G(r(a), r'(a')) V(a').$$

The constant c is, therefore, determined from the conservation of the total volume of the domains.

As an example, we apply equation (10) to spherical domains in three dimensions. First, note that since

$$\sum_{ij} M_{ij}(r, r') = 0$$

there is no self-interaction. The two-body interaction is obtained as follows. Suppose that there are two domains with radius R_1 at X_1 and R_2 at X_2. We assume that the distance $|X_1 - X_2|$ is large compared with R_1 and R_2 and that the positions X_1 and X_2 are independent of time. If $r(a)$ is the surface of the first sphere, the first term in (11) does not vanish when the integrals over r_1 and r_2 are inside the second and the first domains, respectively. The second term in (11) gives a finite contribution if the integrals are in the second sphere. Generalizing this to many domains, we obtain in a non-conserved system

$$\frac{\mathrm{d}R_i}{\mathrm{d}t} = -\frac{2}{R_i} + \frac{2}{R_c} - \sum_{i \neq j} \frac{\gamma}{|X_i - X_j|^6} R_j^3 (2R_i^3 + R_j^3) \tag{12}$$

where $\gamma = 2\mu_1 S_0^6 \lambda^2 / 3\sigma L^2$. We have introduced the critical radius R_c instead of the constant c. For a conserved case, the left-hand side should be replaced by

$$\frac{S_0^2}{\sigma} \left(\frac{R_i \, \mathrm{d}R_i}{\mathrm{d}t} + \sum_{i \neq j} |X_i - X_j|^{-1} R_j^2 \frac{\mathrm{d}R_j}{\mathrm{d}t} \right).$$

This together with (12) is consistent with the results obtained in [4] where the short-distance correction in the long-range interaction has been taken into account. It is noted here that a three-body interaction also arises from (11). However, we do not write it down since it does not dominate the binary interaction.

Equation (12) indicates that if γ (or μ_1) is positive—in other words, if the domains are harder than the matrix—the growth becomes slow as the domain radius increases. However, the details depend crucially on the radius of each domain. Let us focus our attention on two adjacent domains with the radius R_i and R_j. If $R_i > R_j$, equation (12) shows that the growth rate of the i-domain is larger than that of the j-domain. This implies that a larger domain grows more rapidly and eventually dominates the surrounding smaller domains. On the other hand, if the domains are softer ($\mu_1 < 0$), the growth is accelerated. A smaller domain grows rapidly compared with a larger one. Thus as the domains keep growing and before they merge, the radius distribution becomes narrower. When the order parameter is conserved, the domains cease to grow because of the conservation law, as was shown by Kawasaki and Enomoto [4].

The above property of domain growth can also be understood energetically. When μ_1 is positive, the elastic energy increases as the domain radius is increased, as is easily seen from the last term in (9). Note that M_{ij} is a positive definite quantity. Thus larger

domains grow rapidly while other domains remain small to suppress the increase in the elastic energy. It would be possible for the smaller domains to shrink and disappear, although this process is beyond the validity of the present perturbation theory. When μ_1 is negative, the energy decreases as the domains expand. Most favourable is the situation in which all domains grow with the same radius if they are equally spaced.

To conclude, we have derived the interface equation of motion under the long-range elastic interaction. Application to spherical domains shows that depending on the sign of μ_1, the growth kinetics exhibit drastically different behaviour. Any isolated localized domains cannot exist stably for a non-conserved system modelled by (1) in contrast to a conserved case.

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