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Relaxation rate of conserved and nonconserved order parameters in replacive transitions

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We apply the kinetic Bragg-Williams model, introduced recently, to coherent precipitation of ordered phases. We show that the relaxation rate of the long-range order parameter (nonconserved) and of the concentration (conserved order parameter) are coupled not only by the free energy as usually assumed, but also by nondiagonal terms in the linear operator that connects the relaxation rate of both order parameters to the driving forces, i.e., the partial derivatives of the free-energy functional with respect to the order parameters. Moreover, we show that the relaxation of the nonconserved order parameter may occur by two routes, a homogeneous one (on site relaxation) and a heterogeneous one (migration of domain boundary). All such effects are ignored by existing theories.

I. INTRODUCTION

In the classical theory of the relaxation of order parameters,[1] one assumes that close to equilibrium, the rate of change of the order parameter (η) is proportional to the corresponding gain of free energy $\mathcal{F}(\eta)$:

$$\partial\eta/\partial t=-\mathcal{L}\delta\mathcal{F}/\delta\eta \ . \tag{1}$$

Such a model ("model A" in Ref. 1) has the virtue of driving the order parameter to its equilibrium value, η^* defined by

$$\left.\frac{\delta\mathcal{F}}{\delta\eta}\right|_{\eta^*}=0 \ . \tag{2}$$

If the order parameter is globally conserved, as is the case for the concentration field, $n(\mathbf{r})$,

$$\int n(\mathbf{r})d\mathbf{r}=N_B=\text{number of } B \text{ atoms in the alloy} \ , \tag{3}$$

the equilibrium concentration field $n^*(\mathbf{r})$ is such that $\mathcal{F}$ is a minimum under the constraint (3):

$$\left.\frac{\delta\mathcal{F}}{\delta n(\mathbf{r})}\right|_{n^*(\mathbf{r})}=\mu \ . \tag{4}$$

The "chemical potential" defined as the variation of the functional $\mathcal{F}$ with respect to the concentration field $n(\mathbf{r})$, $\delta\mathcal{F}/\delta n(\mathbf{r})$, is uniform at equilibrium and has the value μ defined by Eq. (3) applied to $n^*(\mathbf{r})$. It is then natural to assume that the rate of relaxation of $n(\mathbf{r})$ is such that it flattens out the inhomogeneity in the chemical potential; in a linear approximation ("model B" in Ref. 1):

$$\partial n/\partial t=-\nabla(-\mathcal{L}\nabla\delta\mathcal{F}/\delta n) \ . \tag{5}$$

Both $\mathcal{L}$'s in Eqs. (1) and (5) are usually assumed to be constants.

It should, however, be noticed that for nonconserved order parameter also, the chemical potential is uniform at equilibrium (and equal to zero). While Eq. (1) is certainly valid for a uniform system, it is not clear why Eq. (5) should not apply to *nonuniform nonconserved* order parameters as well.

The two types of order parameter may enter the free-energy functional:[2] Eqs. (1) and (5) are then assumed to govern the coupled relaxation of η and n ("model C" in Ref. 1). Such a process is of great interest in metallurgy where coherent precipitation of an ordered phase is a common practice for obtaining high-strength alloys at high temperature. The kinetics of this latter transformation is described by Eqs. (1) and (5), which are solved numerically with appropriate forms of the free-energy functional $\mathcal{F}$.[3-6]

As discussed in a previous work,[7] the weakness of the above formalism is to rely on a mobility coefficient, $\mathcal{L}$, with no connection to the underlying thermodynamics of the alloy. The purpose of this paper is to derive the expression for the relaxation rate of a nonuniform nonconserved order parameter in a binary alloy treated in the simplest mean-field approximation, the "kinetic Bragg-Williams" approximation introduced in Ref. 7. Such a model turned out to be useful in providing an expression for the mobility in the Cahn-Hilliard diffusion equation, as well as for the atomic transfer coefficient across coherent interfaces.[7,8]

The main result is that, in a nonuniform alloy, the evolutions of the long-range order parameter $\eta(\mathbf{r})$ and the concentration $n(\mathbf{r})$ fields (respectively the nonconserved and conserved order parameter) are *coupled* not only by the free-energy functional, as described by existing models,[2-6] but also by cross terms in the rate equations themselves. If $\boldsymbol{\psi}$ is the two-dimensional order parameter (η,n), we show that

$$\partial\psi/\partial t=\mathbb{M}\nabla_{\psi}\mathcal{F} \ , \tag{6}$$

where $\mathbb{M}$ is a *nondiagonal* operator. Equation (6) reduces to Eq. (1) for alloys with a uniform concentration and to Eq. (5) if the nonconserved order parameter relaxes much more rapidly than the concentration field.

The paper is organized as follows: we first introduce the "kinetic Bragg-Williams model," then derive Eq. (6) with an explicit form for the elements of the operator $\mathbb{M}$, namely the operators $\mathcal{M}^{\eta\eta},\mathcal{M}^{n\eta},\mathcal{M}^{\eta n},\mathcal{M}^{nn}$, and finally discuss some consequences of the latter expressions.

II. THE MODEL

In Ref. 7, we proposed a mean-field description of diffusion fully compatible with Cahn's thermodynamics of inhomogeneous systems. It is assumed that diffusion proceeds by direct exchanges between neighboring sites, that the activation energy for the process is a saddle-point energy minus the binding energy of the pair of exchanging atoms to its environment, and that the attempt frequency for atomic exchanges is independent of the alloy configuration. The configuration is defined by averaging the occupation number of lattice sites: the averaging can be performed among several identical systems, defining the concentration on each site, or among the sites of an atomic row, defining the concentration in the row, or among the sites of a plane normal to a given direction, defining the concentration in the plane.

To be more specific, we deal in the following with a binary alloy (A-B), and restrict to a one-dimensional problem, i.e., we consider a set of identical lattice planes labeled $1,\ldots,p,\ldots P$; each plane contains Ω atomic sites, sheared among two sublattices with a proportion ω_i of sites belonging to sublattice i ($i=1,2$). The configuration of the alloy is defined by the set of concentrations n_p^i of B atoms in plane p ($p=1$ to P) on sublattice i ($i=1$ or 2). The corresponding A concentration is $n_p'^i=1-n_p^i$. The configuration can alternatively be described by two other fields: (i) the overall concentration in plane p: $n_p=\omega_1 n_p^1+\omega_2 n_p^2$; (ii) the "degree of order" in plane p: $\eta_p=n_p^1-n_p^2$; in the disordered state, $n_p^1=n_p^2$ and $\eta_p=0$.

A free-energy functional $\mathcal{F}(\mathbf{B})=\Omega f(\mathbf{B})$ can be written where $\mathbf{B}$ is a $2P$ dimensional vector with components $(\Omega\omega_i n_p^i)$. It can be written in terms of the internal energy $\mathcal{E}(\mathbf{B})$ and an entropy $\mathcal{S}(\mathbf{B})$. $\mathcal{E}(\mathbf{B})=\Omega e(\mathbf{B})$ where the internal energy per site e writes

$$e(\mathbf{B})=\frac{1}{2}\sum_{p=1}^{P} e_p(\{n_p^i,n_p'^i\})+\text{surface term} \tag{7}$$

and $\mathcal{S}(\mathbf{B})=\Omega s(\mathbf{B})$, where the entropy per site s writes

$$s(\mathbf{B})=k_B\sum_{p=1}^{P} s_p(n_p^1,n_p'^1,n_p^2,n_p'^2)+\text{surface term} \tag{8a}$$

with

$$s_p=n_p^1\ln(n_p^1)+n_p'^1\ln(n_p'^1)+n_p^2\ln(n_p^2)+n_p'^2\ln(n_p'^2)\ . \tag{8b}$$

Depending on the range of interatomic interactions, e_p in Eq. (7) implies concentrations in planes $p\pm k$, $k=1$ or more.

For the sake of simplicity, we assume that the atomic exchanges occur among nearest-neighbor pairs of sites only, and that all such neighbors are either in the same plane or in nearest-neighbor planes. We introduce the partial coordination numbers z_{pq}^{ij} as the number of nearest neighbors a site (i,p), i.e., on sublattice i in plane p, has on sublattice j in plane q.

For describing the kinetics of such a model, we introduce the "forward" current of B atoms from one site (i,p) to one site (j,q), along one single link between such sites:

$$j_{pq}^{ij}=\nu n_p^i n_q'^j \exp-\beta\left[E_{\text{sp}}-\frac{\partial\mathcal{E}}{\Omega\omega_i\partial n_p^i}-\frac{\partial\mathcal{E}}{\Omega\omega_j\partial n_q'^j}\right]\ . \tag{9}$$

In Eq. (9), ν is the attempt frequency, β the inverse temperature ($1/k_BT$) and E_{sp} the saddle-point configuration energy (assumed to be a constant). The partial derivative of the internal energy with respect to n_p^i is to be taken keeping $n_p'^i$ constant. The *net* current of B atoms from site (i,p) to site (j,q) is the difference between the forward and backward current along the link $(i,p;j,q)$:

$$\mathcal{J}_{pq}^{ij}=j_{pq}^{ij}-j_{qp}^{ji}=f-b\ . \tag{10}$$

Since f and b are both positive, Eq. (10) can be symmetrized as

$$\mathcal{J}_{pq}^{ij}=\sqrt{fb}\times[\sqrt{f/b}-\sqrt{b/f}\,]\ . \tag{11}$$

Introducing expressions (9) and (10) into Eq. (11) yields

$$\mathcal{J}_{pq}^{ij}=\sqrt{fb}\times[\sinh\beta(\mu_p^i-\mu_q^j)]\ . \tag{12}$$

In Eq. (12), the local chemical potential μ_p^i has the following definition:

$$\mu_p^i=\frac{1}{\omega_i}\frac{de}{dn_p^i}+\beta^{-1}\ln\left[\frac{n_p^i}{n_p'^i}\right]\ . \tag{13}$$

μ_p^i is the derivative of the free energy of the alloy with respect to the B concentration on the sublattice i in plane p. Finally, the total flux of B atoms between sublattices i and j from plane p to plane q is given by the right-hand side of Eq. (12) times the number of appropriate links per site, L_{pq}^{ij}:

$$J_{pq}^{ij}=L_{pq}^{ij}\times\mathcal{J}_{pq}^{ij}\ . \tag{14}$$

For the sake of simplicity, we assume the structure to be centrosymmetric, i.e., $L_{pq}^{ij}=L_{pq}^{ji}$. When the alloy is in an equilibrium state, the net flux along any link is zero, i.e., the local chemical potential has a unique value on both sublattices in all planes. It is easily shown that this value is nothing but the chemical potential μ introduced in Eq. (4).

Close to equilibrium, μ_p^i depends both on i and p, but departs only slightly from μ; B fluxes take place according to Eq. (12) which can be expanded to first order in the deviation from μ:

$$J_{pq}^{ij}\approx m_{pq}^{ij}\cdot\beta(\mu_p^i-\mu_q^j)\ , \tag{15}$$

where the mobility coefficient is given by

$$\begin{aligned}m_{pq}^{ij}=\nu L_{pq}^{ij}\sqrt{n_p^i n_p'^i n_q^j n_q'^j}\\ \times\exp\frac{\beta}{2}\Bigg[-2E_{\text{sp}}+\frac{\partial\mathcal{E}}{\Omega\omega_i\partial n_p^i}+\frac{\partial\mathcal{E}}{\Omega\omega_j\partial n_q'^j}\\ +\frac{\partial\mathcal{E}}{\Omega\omega_i\partial n_q^j}+\frac{\partial\mathcal{E}}{\Omega\omega_j\partial n_p'^i}\Bigg]\ .\end{aligned} \tag{16a}$$

Since Eq. (15) results from a first-order expansion of Eq.

(12), the above mobility must be evaluated to zeroth order in the deviation from equilibrium: All *concentration dependent terms in Eq. (16) must be given their equilibrium value* ($\bar{n}$), so that, taking advantage of detailed balance, we get

$$m^{ij}_{pq} = \nu L^{ij}_{pq} \bar{n}^{i}_{p} \bar{n}'^{j}_{q} \exp\beta \left[-E_{\mathrm{sp}} + \frac{\partial \mathcal{E}}{\Omega \omega_i \partial n^i_p} + \frac{\partial \mathcal{E}}{\Omega \omega_j \partial n'^j_q} \right] . \tag{16b}$$

It is now a simple mater to establish Eq. (6): this is done in the next section.

III. THE COUPLED RELAXATION OF THE FIELDS OF CONCENTRATION AND OF DEGREE OF ORDER

The rate of change of the degree of order and of the overall concentration in plane p is obtained from the rates of change of the concentrations on each sublattice in the latter plane. The latter write

$$\frac{dn^i_p}{dt} = J^{ji}_{pp} + J^{ji}_{p-1p} + J^{ii}_{p-1p} - J^{ij}_{pp+1} - J^{ii}_{pp+1} . \tag{17}$$

Indeed,

$$\frac{dn_p}{dt} = \omega_1 \frac{dn^1_p}{dt} + \omega_2 \frac{dn^2_p}{dt} \quad \text{and} \quad \frac{d\eta_p}{dt} = \frac{dn^1_p}{dt} - \frac{dn^2_p}{dt} . \tag{18}$$

Simple but tedious algebra yields the following results:

$$\begin{aligned} \frac{d\eta_p}{dt} &= \mathcal{M}^{\eta\eta}_p \frac{df}{d\eta_{p-1}} + \mathcal{M}^{\eta n}_p \frac{df}{dn_{p-1}} , \\ \frac{dn_p}{dt} &= \mathcal{M}^{n\eta}_p \frac{df}{d\eta_{p-1}} + \mathcal{M}^{nn}_p \frac{df}{dn_{p-1}} , \end{aligned} \tag{19}$$

where the $\mathcal{M}^{\alpha\beta}_p$'s are linear operators with the following definition:

$$\begin{aligned} \mathcal{M}^{\eta\eta}_p &= \frac{1}{2\omega_1\omega_2} [-4m^{12}_{pp} + \mathbb{D}[(m^{11}_{p-1p} + m^{22}_{p-1p})\mathbb{D}\cdot] \\ &\qquad - \mathbb{S}[(m^{21}_{p-1p} + m^{12}_{p-1p})\mathbb{S}\cdot]] , \\ \mathcal{M}^{\eta n}_p &= \frac{1}{2} \mathbb{D}[(m^{11}_{p-1p} - m^{22}_{p-1p})\mathbb{D}\cdot] , \\ \mathcal{M}^{n\eta}_p &= \frac{1}{2\omega_1\omega_2} \mathbb{D}[[(m^{12}_{p-1p} + m^{11}_{p-1p}) \\ &\qquad - (m^{21}_{p-1p} + m^{22}_{p-1p})]\mathbb{D}\cdot] , \\ \mathcal{M}^{nn}_p &= \frac{1}{2} \mathbb{D}[[(m^{12}_{p-1p} + m^{11}_{p-1p}) + (m^{21}_{p-1p} + m^{22}_{p-1}p)]\mathbb{D}\cdot] , \end{aligned} \tag{20}$$

In Eqs. (20) the operators $\mathbb{D}\cdot$ and $\mathbb{S}\cdot$ have the following definition:

$$\mathbb{D}u_{p-1} = u_p - u_{p-1} \quad \text{and} \quad \mathbb{S}u_{p-1} = u_p + u_{p-1} . \tag{21}$$

It is interesting to notice that

$$\mathbb{D}[\mathbb{D}u_{p-1}] = u_{p-1} + u_{p+1} - 2u_p , \tag{22a}$$

and corresponds to the curvature of u at site p,

$$\mathbb{S}[\mathbb{D}u_{p-1}] = \mathbb{D}[\mathbb{S}u_{p-1}] = u_{p+1} - u_{p-1} , \tag{22b}$$

and corresponds to the derivative of u at site p, and

$$\mathbb{S}[\mathbb{S}u_{p-1}] = \mathbb{D}[\mathbb{D}u_{p-1}] + 4u_p . \tag{22c}$$

For deriving Eq. (20), the following relationships had to be used:

$$\begin{aligned} \frac{df}{d\eta_p} &= \omega_1\omega_2(\mu^1_p - \mu^2_p) , \\ \frac{df}{dn_p} &= \mu^1_p + \mu^2_p , \end{aligned} \tag{23}$$

and

$$a_\alpha u_\beta \mp a_\beta u_\alpha = \tfrac{1}{2}[(a_\alpha + a_\beta)(u_\alpha \mp \mu_\beta) + (a_\alpha - a_\beta)(u_\alpha \pm u_\beta)] .$$

Equations (19) and (20) with the mobilities given by Eq. (16) describe the coupled relaxation of the nonconserved and of the conserved order-parameter fields close to equilibrium; we state once more that *the mobilities are to be evaluated giving all quantities in Eq. (16) their value in the equilibrium state in the vicinity of which the relaxation is being described.* In the particular case where the equilibrium state is uniform (i.e., invariant by translation perpendicular to the plane p), the mobilities are independent of p and can be factorized in the operators $\mathcal{M}^{\alpha\beta}_p$ [Eqs. (20)].

IV. DISCUSSION

Several new interesting features are revealed by Eqs. (19) and (20).

(1) The relaxation of the fields of concentration and of degree of order in an alloy are coupled; indeed, the nondiagonal terms $\mathcal{M}^{\alpha\beta}_p$ ($\alpha \neq \beta$) of the operator $\mathbb{M}$ are nonzero in the general case. This coupling is ignored by the available theories of precipitation of ordered phases.[2–6] The reason for this omission is that the mobilities in the existing theories are introduced in an arbitrary way, the consistency of which, with the underlying thermodynamics, is not proven. In most theories, the mobilities are taken as constants.[1,9] The concentration and the degree of order are not the eigenmodes of the problem. Neither are the concentrations on each sublattice.

(2) Even if we ignore the nondiagonal element $\mathcal{M}^{\eta n}_p$, the relaxation of the degree of order (the nonconserved order parameter in our problem) does not obey Eq. (1): indeed $\mathcal{M}^{\eta\eta}_p$ is the sum of three terms [Eq. (20)], the last two of which contain a contribution of the *nonuniformity* in $df/d\eta_p$. In other words, if the system is not uniform, the nonconserved order parameter relaxes via two paths: homogeneous relaxation [properly described by Eq. (1)], which corresponds to intraplane exchanges between the sublattices, and migration of domain boundaries (described by the two last terms in $\mathcal{M}^{\eta\eta}_p$), which corresponds to interplane exchanges between the sublattices. This latter contribution is ignored in the classical treatments.[1]

(3) The relaxation of the conserved order parameter (the concentration field) is well described by an equation similar to Eq. (5). It is however also coupled to the relaxation of the nonconserved parameter by the operator $\mathcal{M}_p^{n\eta}$. One example of such a coupling is the change of concentration induced at an antiphase boundary by a wetting transition. This coupling is overlooked by classical treatments.

(4) It may be assumed that the nonconserved order parameter relaxes much more rapidly than the concentration field [the validity of this hypothesis might not be consistent with the actual values of the mobility coefficients in Eq. (20)]. Then $df/d\eta_p=0$, and we are left with the contribution of $\mathcal{M}_p^{\eta n}$ to the relaxation rate of η_p and of $\mathcal{M}_p^{nn}$ to that of n_p. We recover Eq. (5) for the relaxation of the conserved order parameter.

(5) If the equilibrium state towards which the system relaxes, is a fully disordered uniform solid solution, the concentrations on all the sublattices in all the planes are the same, so that $m_{pq}^{ij}=m_{pq}^{ji}$, but in the most general case $m_{pq}^{ii}\neq m_{pq}^{jj}$ for topological reasons. Under such circumstances, the elements of $\mathbb{M}$ are written as follows:

$$\begin{aligned}\mathcal{M}^{\eta\eta}&=\frac{1}{2\omega_1\omega_2}[-4(m_{pp}^{12}+m_{p-1p}^{12})\\&\qquad+(m_{p-1p}^{11}+m_{p-1p}^{22}-2m_{p-1p}^{12})\mathbb{D}^2\cdot]\,,\\\mathcal{M}^{\eta n}&=\frac{1}{2}(m_{p-1p}^{11}-m_{p-1p}^{22})\mathbb{D}^2\cdot\,,\\\mathcal{M}^{n\eta}&=\frac{1}{2\omega_1\omega_2}(m_{p-1p}^{11}-m_{p-1p}^{22})\mathbb{D}^2\cdot=\frac{1}{\omega_1\omega_2}\mathcal{M}^{\eta n}\,,\\\mathcal{M}^{nn}&=\frac{1}{2}(m_{p-1p}^{11}+m_{p-1p}^{22}+2m_{p-1p}^{12})\mathbb{D}^2\cdot\,.\end{aligned}\tag{24}$$

The transformation of Eq. (19) into a continuum theory is straightforward in this case:

$$\begin{aligned}\partial\eta(x,t)/\partial t&=-\mathcal{L}_0\delta\mathcal{F}/\delta\eta+\mathcal{L}_1\frac{\partial^2}{\partial x^2}(\delta\mathcal{F}/\delta\eta)\\&\qquad+\mathcal{L}_2\frac{\partial^2}{\partial x^2}(\delta\mathcal{F}/\delta n)\,,\\\partial n(x,t)/\partial t&=\mathcal{L}_3\frac{\partial^2}{\partial x^2}(\delta\mathcal{F}/\delta\eta)+\mathcal{L}_4\frac{\partial^2}{\partial x^2}(\delta\mathcal{F}/\delta n)\,.\end{aligned}\tag{25}$$

In a recent paper,[10] it was argued that the Laplacian terms in Eq. (25) are artificially introduced, that the $\mathcal{L}_0$ term is the only one to contribute to the relaxation rate of the nonconserved as well as of the conserved order parameter. The present model shows that this is not the case and moreover that Laplacian terms do also enter the relaxation rate of nonconserved order parameters.

Finally, it is worth mentioning that the model we used has been solved numerically prior to linearization, to study coherent ordered phase separation in binary alloys with the bcc structure, in particular in the presence of external forcing like shear under plastic deformation or ballistic mixing under irradiation.[11–13] It is worth using this technique to explore the range of validity of the linear approximation we used in the present work. Work is in progress in this direction.

CONCLUSION

We applied the kinetic Bragg-Williams model[7] to the coherent precipitation of ordered phases. We show that the relaxation rate of the long-range order parameter (nonconserved) and of the concentration (conserved order parameter) are coupled not only by the free energy as usually assumed,[1] but also by nondiagonal terms in the linear operator which connects the relaxation rate of both order parameters to the driving forces, i.e., the partial derivatives of the free-energy functional with respect to the order parameters. Moreover, we show that the relaxation of the nonconserved order parameter may occur by two routes, a homogeneous one (on site relaxation) and a heterogeneous one (migration of domain boundary). All such effects are ignored by current theories. It must be acknowledged that the "kinetic Bragg-Williams" approximation used in this paper is very crude indeed and excludes the possibility of exploring the effect of long-range fluctuations on the kinetics in the vicinity of critical points. Such questions could be addressed using a more sophisticated thermodynamical description (e.g., pair approximation).[14]

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