Fluctuations in domain growth: Ginzburg-Landau equations with multiplicative noise

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Ginzburg-Landau equations with multiplicative noise are considered, to study the effects of fluctuations in domain growth. The equations are derived from a coarse-grained methodology and expressions for the resulting concentration-dependent diffusion coefficients are proposed. The multiplicative noise gives contributions to the Cahn-Hilliard linear-stability analysis. In particular, it introduces a delay in the domain-growth dynamics.

I. INTRODUCTION

The study of fluctuations in spatially extended systems far from equilibrium constitutes an active field of research. Recent experiments indicate that noise seems to have an important role in dynamic processes such as pattern formation and growth. Examples of such effects have been observed in the generation of sidebranching in dendritic growth,[1] cells in Rayleigh-Benard convection[2] and William domains in the electrohydrodynamic instability of nematic liquid crystals.[3] For an overview of this topic see Ref. 4.

Some analytical work and computer simulations have been carried out to explain such phenomena.[5-9] However, many fundamental aspects remain to be clarified, for instance, the origin of the noise,[1,2] or the correct modeling of the coupling between the noise and the state of the system.[6,7] The simplest way to consider fluctuations is to add a thermal noise to the macroscopic equations. However, it has been observed that in some cases[2,5] there is a large discrepancy, as large as four orders of magnitude,[2] between the values of the intensity required to get a good agreement with experiments[1,2] and those corresponding to the assumption of thermal noise. The origin of the noise in these experiments is not known and a type of modeling of fluctuations may be needed to explain them. In other cases[6,7] the fluctuations are introduced in such a way that it is difficult to determine the form in which the noise is coupled to the state of the system. For example, small perturbations or random numbers are added to the values of the variables in the numerical integration of the corresponding macroscopic equations.[6,7]

A different type of stochastic model is required when the effects of the noise are considered to depend on the state of the system. In this situation, the noise appears in the equations multiplying a function of the relevant variables. This is known as multiplicative noise. Its effects on the system are, in general, more important than those induced by simple additive noise. The reason is that the existence of a coupling could induce amplification of the

stochastic effects. Multiplicative noise usually appears when noise of external origin is considered. In some experiments in liquid crystals,[3] this situation has been studied by deliberately superimposing a noise to the ac voltage. The results imply a strong effect on the response of the system, like changes in the threshold of the instability points.[3] The possibility of externally originated noise included in the apparatus in Rayleigh-Benard cell generation is an open question.[2] An internal noise could also appear in a multiplicative way. This is the situation that we will consider in this paper.

Theoretical studies of the effects of multiplicative noise have been carried out in simple models without spatial dependence,[10] but very little is known regarding spatially extended systems. First, some aspects need to be studied in detail, like the interpretation of the stochastic equations or the analytical and even the numerical treatment of the equations. To this end, in this paper we start by deriving a model with spatial dependence in which the noise of thermal origin appears in the equation in a multiplicative way. The resulting Ginzburg-Landau equations with multiplicative noise could be used to describe the temporal evolution of the concentrations of a system of two components, like a binary liquid or an alloy that could undergo phase separation.[11] In this situation, the system is suddenly quenched from a one-phase region inside its coexistence region. Then, the homogeneous phase becomes unstable and domains of the new stable phases start growing. This mechanism is called spinodal decomposition. In a previous paper,[12] the deterministic evolution of such a system was studied when a variable-dependent diffusion coefficient was taken into account. This assumption has been considered to model deep quenching[13,14] or to take into account the presence of an external field, like gravity.[15] In Sec. II we introduce fluctuations in this macroscopic model.[13-15] We find that the assumption of a concentration-dependent diffusion coefficient implies multiplicative thermal fluctuations. The field model that we derive is given by the following Ginzburg-Landau-type equation with multiplicative noise:

$$\frac{\partial c(\mathbf{r},\tau)}{\partial \tau} = \nabla M(c)\cdot\nabla\frac{\delta F[c]}{\delta c} - \frac{\beta^{-1}}{2}\nabla\cdot\left[\nabla\frac{\delta}{\delta c}\right]M(c)$$

$$+\nabla^i m(c)\xi^i(\mathbf{r},\tau) , \tag{1.1}$$

where $c(\mathbf{r},\tau)$ is the concentration variable, $F[c]$ is the Ginzburg-Landau free-energy functional, and $M(c)=m^2(c)$ is the concentration-dependent diffusion coefficient. The noise is a d-dimensional vector with a correlation:

$$\langle \xi^i(\mathbf{r},\tau)\xi^j(\mathbf{r}',\tau')\rangle = 2\beta^{-1}\delta_{i,j}\delta(\mathbf{r}-\mathbf{r}')\delta(\tau-\tau') . \tag{1.2}$$

β^{-1} is the intensity of the Gaussian white noise. A common assumption regarding the dependence of M on concentration has been obtained by phenomenological arguments.[13–15] That is

$$M(c)=1-ac^2 , \tag{1.3}$$

where a is a parameter related to temperature. For $a=0$ we obtain the usual model B of phase separation dynamics with additive noise.[11] For $a\neq0$, apart from the multiplicative term, we find a spurious term, the second term on the right-hand side of Eq. (1.1), of stochastic origin. This spurious term ensures the evolution of the system to the correct equilibrium solution.

In Sec. II we present a derivation of the model in two main steps. In the first, we obtain the formal Fokker-Planck and Langevin equations from a general master equation by means of a coarse-grained procedure. This method has been used to derive dynamic models of phase separation in the presence of additive fluctuations.[16] Here, we generalize the procedure to multiplicative noise. The second step of our derivation is required because the coarse-grained procedure does not give explicit expressions of the concentration-dependent diffusion coefficient. We propose a general expression for this magnitude at the level of the cell variables and discuss the continuous spatial limit. The model contains a characteristic mesoscopic length which gives a size of the region of interchange of matter between different cells at each time step. In Sec. III, as one of the relevant results of this paper, we show that both terms of stochastic origin give relevant contributions even in a linear-stability analysis, that is to the standard Cahn-Hilliard theory. Our theory predicts that the multiplicative noise induces a delay in the short term behavior of the domain-growth dynamics. In a following paper,[17] we present an algorithm for the numerical integration of general equations of the type of Eqs. (1.1)–(1.3). Results from this numerical integration are in good agreement with the linear analysis. In Sec. IV we give a summary of conclusions. In Appendix A, we derive the Fokker-Planck equation corresponding to the Langevin equations (1.1)–(1.3). In Appendix B, we present a derivation of Langevin equations with multiplicative noise for the case of a nonconserved order parameter.

II. DERIVATION OF THE MODEL

Following the standard coarse-grained procedure,[16,18] we start from a lattice-gas model of interacting particles.

The lattice is divided into regular cells of volume Δx^d containing N sites and the concentration of the binary mixture, c_α, at the cell α is defined by

$$c_\alpha = \frac{1}{N}\sum_{k\in\alpha}\sigma_k , \tag{2.1}$$

where $\sigma_k=1,-1$ indicates a site occupied by a particle A or B, respectively. Then, it is assumed that a Markovian master equation is obeyed by the probability $P(\{c\},t)$ of the configuration of cells, $\{c\}=\{c_1,c_2,\ldots\}$,

$$\partial_t P(\{c\},t)=\sum_{\alpha i\epsilon}[W(\{c\}^{\alpha i}\rightarrow\{c\})P(\{c\}^{\alpha i},t)$$

$$-W(\{c\}\rightarrow\{c\}^{\alpha i})P(\{c\},t)] , \tag{2.2}$$

where the indices α and i enumerate the cells and their nearest neighbors in the positive direction, respectively. $W(\{c\}^{\alpha i}\rightarrow\{c\})$ is the transition probability between the initial configuration

$$\{c\}^{\alpha i}=\{c_1,c_2,\ldots,c_\alpha-\epsilon,c_{\alpha+i}+\epsilon,\ldots\}$$

and the final one $\{c\}=\{c_1,c_2,\ldots,c_\alpha,c_{\alpha+i},\ldots\}$. ϵ is the concentration exchanged in an elementary step of the evolution. The model could be generalized to include long-range interactions.

We consider situations for which the system evolves to an equilibrium state given by a steady-state distribution $P_{st}(\{c\})$, which is proportional to the Boltzmann factor. Then, we write for the transition probability, W

$$W(\{c\}^{\alpha i}\rightarrow\{c\})=M(\{c\}^{\alpha i},\{c\})e^{\beta\Delta F/2} , \tag{2.3}$$

where $F(\{c\})$ is a coarse-grained free energy. The detailed balance condition is fulfilled provided M is symmetric by interchange of the initial and final states. In the usual derivation of the field model, with constant diffusion coefficient and additive noise, no dependence of M on the configurations is considered, and it is assumed that $M(\{c\}^{\alpha i},\{c\})=P(\epsilon)$, where $P(\epsilon)$ is a sharp function around $\epsilon=0$.[16,18]

Here, we present a generalization of this procedure that will give rise to a model with a variable-dependent diffusion coefficient and a multiplicative noise. By assuming that ϵ is a small quantity, we can expand the different terms of the right-hand side of Eq. (2.2) in power series of ϵ and we get to the lowest order:

$$\frac{\partial P(\{c\},t)}{\partial t}=\Gamma\sum_{\alpha i}K_{\alpha i}M_{\alpha i}[(K_{\alpha i}F)+\beta^{-1}K_{\alpha i}]P(\{c\},t) , \tag{2.4}$$

where $\Gamma=\langle\epsilon^2\rangle\beta/2$ and $\langle\epsilon^2\rangle$ is the second moment of $P(\epsilon)$. The operator $K_{\alpha i}$ is given by

$$K_{\alpha i}=\frac{\partial}{\partial c_{\alpha+i}}-\frac{\partial}{\partial c_\alpha} . \tag{2.5}$$

In Eq. (2.4), we have made use of the symmetry condition on

$$M_{\alpha i}=M(\{c\}^{\alpha i},\{c\})=M(\{c\},\{c\}^{\alpha i}) .$$

Equation (2.4) can be written in more usual form (see Ap-

pendix A for details):

$$\frac{\partial P}{\partial t} = -\Delta x^2 \Gamma \frac{\partial}{\partial c_\alpha} (\nabla_L^i)_{\alpha\beta} M_{\beta i} (\nabla_R^i)_{\beta\sigma} \left[\frac{\partial F}{\partial c_\sigma} + \beta^{-1} \frac{\partial}{\partial c_\sigma} \right] P ,$$

(2.6)

where ∇_L^i and ∇_R^i are the left and right discrete versions of the gradient operators:

$$(\nabla_R^i)_{\alpha\beta} = \frac{1}{\Delta x} (\delta_{\alpha+i\beta} - \delta_{\alpha\beta}) ,$$

$$(\nabla_L^i)_{\alpha\beta} = \frac{1}{\Delta x} (\delta_{\alpha\beta} - \delta_{\alpha-i\beta}) .$$

(2.7)

The operator $K_{\alpha i}$ has been expressed as

$$K_{\alpha i} = \Delta x (\nabla_R^i)_{\alpha\beta} \frac{\partial}{\partial c_\beta} ,$$

(2.8)

where summation over repeated indices is understood.

The Langevin equation, in the Stratonovich interpretation, associated with the Fokker-Planck equation (2.6) is given by

$$\dot{c}_\alpha = \Gamma \Delta x^2 (\nabla_L^i)_{\alpha\beta} M_{\beta i} (\nabla_R^i)_{\beta\sigma} \frac{\partial F}{\partial c_\sigma} - \frac{\Gamma}{2} \beta^{-1} \Delta x^2 (\nabla_L^i)_{\alpha\beta}$$

$$\times (\nabla_R^i)_{\beta\sigma} \frac{\partial M_{\beta i}}{\partial c_\sigma} + (\nabla_L^i)_{\alpha\beta} m_{\beta i} \xi_\beta^i(t) ,$$

(2.9)

where $\xi_\beta^i(t)$ is a Gaussian white noise of zero mean and correlation

$$\langle \xi_\alpha^i(t) \xi_\beta^j(t') \rangle = 2\Delta x^2 \Gamma \beta^{-1} \delta_{ij} \delta_{\alpha\beta} \delta(t - t') ,$$

(2.10)

and $m_{\beta i}$ is defined by

$$M_{\beta i}(\{c\}) = [m_{\beta i}(\{c\})]^2 .$$

(2.11)

The Langevin equation (2.9) can be shown to correspond to be Fokker-Planck equation (2.6) by deriving the latter from the former. In Appendix A we present the details of this derivation.

At this point, Eqs. (2.6) and (2.9) are formal and general equations in which the expressions of $M_{\alpha i}(\{c\})$ need to be specified for each model. In particular, explicit expressions of $M_{\alpha i}(\{c\})$ are required to perform numerical simulations. Furthermore, these equations are given in terms of the cell variables and we are also interested in finding the corresponding equations in the continuous spatial limit. Here, we make some assumptions about the form of $M_{\alpha i}(\{c\})$ and then we propose a family of models. In general, $M_{\alpha i}(\{c\})$ depends on the concentration values of all the lattice points in a given configuration. In the continuous spatial limit, this gives rise to a functional expression of $M[c]$. In order to obtain a local mobility function $M(c)$ like the one considered in the macroscopic model,[13-15] given by Eq. (1.3), we assume that the transition probability, Eq. (2.3), only involves exchanges of matter between nearest-neighbor cells α, $\alpha+i$ at each elementary step. Furthermore, we restrict ourselves to functions $M_{\alpha i}(\{c\})$ that only depend on the concentration values of the cells α, $\alpha+i$, and on a limited number,

n, of cells in the vicinity of α and $\alpha+i$:

$$M_{\alpha i} = \sum_\beta Q_{\alpha\beta}^i f(c_\beta) ,$$

(2.12)

where $f(c_\beta)$ is a function of only one variable c_β and the matrix elements $Q_{\alpha\beta}^i$ are different from zero only when the index β corresponds to α, $\alpha+i$, or the **n** cells in the vicinity of this couple. By taking into account the normalization condition:

$$\sum_\beta Q_{\alpha\beta}^i = 1 ,$$

(2.13)

we find that the continuous limit of $M_{\alpha i}(\{c\})$ is given by $M(c) = f(c)$. To simplify the model, we take that $Q_{\alpha,\alpha}^i = Q_{\alpha,\alpha+i}^i \equiv Q_0$. A characteristic mesoscopic length is present in the family of models described by Eq. (2.12). This length gives the size of the region which includes all cells which appear in the definition of $M_{\alpha i}(\{c\})$. The transition probability, Eq. (2.3), depends only on the concentration values of cells that are closer than a distance of the order of $n^{1/d}\Delta x$. To characterize the size of this region, we take into account that, from Eq. (2.13), Q_0 is of order n^{-1} and we define a new parameter R by

$$R = \Delta x Q_0^{-1/d} ,$$

(2.14)

which is precisely of the order of $n^{1/d}\Delta x$ and represents the mesoscopic length scale of the model.

An explicit example of $M_{\alpha i}(\{c\})$, Eq. (2.12), which corresponds to Eq. (1.3) in the continuous limit, is given by

$$M_{\alpha i}(\{c\}) = Q_0\{(1 - ac_\alpha^2) + (1 - ac_{\alpha+i}^2)\} + Q_0 \sum_\beta (1 - ac_\beta^2) ,$$

(2.15)

which depends on c_α, $c_{\alpha+i}$, and on all the nearest neighbors β of this couple.

The dynamics represented by Eqs. (2.4), (2.6), or (2.9) involves not only the function $M_{\alpha i}$ but also derivatives in terms of the operator $K_{\alpha i}$, Eq. (2.5), like $K_{\alpha i} M_{\alpha i}$. The result of the action of $K_{\alpha i}$ on the mobility of Eq. (2.12), or, in particular, on the example given by Eq. (2.15), could be written as

$$\frac{1}{\Delta x^{d+1}} K_{\alpha i} M_{\alpha i} = (\nabla_R^i)_{\alpha\beta} \frac{1}{\Delta x^d} \frac{\partial M_{\alpha i}}{\partial c_\beta}$$

$$= \frac{1}{R^d} (\nabla_R^i)_{\alpha\beta} \frac{df(c_\beta)}{dc_\beta} .$$

(2.16)

Now, the statistical properties of the mesoscopic model considered in this paragraph are completely specified by Eqs. (2.12) and (2.16) and the corresponding Fokker-Planck or Langevin, Eqs. (2.6) or (2.9), respectively.

At this point, we give a version of this mesoscopic model in the continuous spatial limit, as is given in Eqs. (1.1)-(1.3). Then, writing Eq. (2.16) in the continuous limit, we make the following identification:

$$\nabla \frac{\delta}{\delta c} M(c) = \frac{1}{R^d} \nabla \frac{df(c)}{dc} .$$

(2.17)

Then, from Eqs. (2.16) and (2.17) we find that the continuous limit of $K_{\alpha i}M_{\alpha i}$ could be written in terms of the gradient of the functional derivative:

$$\frac{1}{\Delta x^{d+1}}K_{\alpha i}M_{\alpha i} \rightarrow \nabla \frac{\delta}{\delta c}M(c) \ , \qquad (2.18)$$

where $M(c)$ is the continuous limit of Eq. (2.12), $M(c)=f(c)$. It is interesting to notice that Eq. (2.17) involves the mesoscopic parameter R for a complete specification of the model and it could be considered as a definition of the functional derivative of $M(c)$.[19]

Now, for the generic form of $M_{\alpha i}$, Eq. (2.12), we can write the Fokker-Planck equation (2.6) at the continuous limit:

$$\frac{\partial P}{\partial \tau} = -\int d\mathbf{r}\frac{\delta}{\delta c(\mathbf{r})}\nabla M\cdot\nabla\left[\frac{\delta F}{\delta c(\mathbf{r})}+\beta^{-1}\frac{\delta}{\delta c(\mathbf{r})}\right]P \ ,$$

$$(2.19)$$

where the new time scale is

$$\tau = t\Gamma\Delta x^{2+d} \ . \qquad (2.20)$$

Analogously, Eq. (1.1) is the continuous Langevin equation corresponding to the discrete Langevin equation (2.9).

From the Fokker-Planck or the Langevin, Eqs. (2.6) and (2.9), respectively, it is possible to derive the equation for the moments. For example, at the continuous limit the first moment obeys the following equation (see Appendix A):

$$\frac{\partial\langle c(\mathbf{r},\tau)\rangle}{\partial\tau} = \nabla\cdot\left\langle M\nabla\frac{\delta F}{\delta c}\right\rangle - \beta^{-1}\nabla\cdot\left\langle\nabla\frac{\delta}{\delta c}M\right\rangle \ , \qquad (2.21)$$

where the functional derivative included in the last term is evaluated in accordance with the prescription given by Eq. (2.17). The most interesting aspect of the Langevin equations (2.9)–(2.12), or equivalently (1.1)–(1.3), is the presence of multiplicative noise that gives rise to the second term of Eq. (2.21).

As the last remark of this section, it is worth sounding a note of caution regarding other Langevin equations that one could be tempted to propose to consider concentration-dependent diffusion coefficients. In principle, the statistical properties of the noise in a Langevin equation should be independent of the variable for the stochastic process techniques to be used safely. For example, this requirement is needed in a derivation of a Fokker-Planck equation from a Langevin equation, like that used in Appendix A. However, one could try to use these standard techniques in a formal manner and postulate a Langevin equation of the following type:

$$\dot{c}_\beta = \Delta x^2 \nabla_L^i M \nabla_R^i \frac{\partial F}{\partial c_\beta} + \eta_\beta(t) \ , \qquad (2.22)$$

where $\eta_\beta(t)$ is now a Gaussian white noise with a correlation:

$$\langle\eta_\beta(t)\eta_{\beta'}(t)\rangle = -2\beta^{-1}\Delta x^2\nabla_L^i M\nabla_R^i\delta_{\beta\beta'}\delta(t-t') \ . \qquad (2.23)$$

In this way, the noise would still be additive but the correlation would depend on the concentration variable.

Then this conjecture appears to avoid the problems introduced by a multiplicative noise, whereas, this is not, in fact, the case. In principle, a formal derivation of a Fokker-Planck equation from Eqs. (2.22) and (2.23) does not give rise to Eq. (2.6) if the Gaussian noise has zero mean. In fact, to reproduce the stationary equilibrium solution, one needs to introduce a spurious term in Eq. (2.22), which involves the presence of functional derivatives of $M(c)$. Then a similar treatment to that presented here would be required. Furthermore, from the practical point of view a numerical integration of the stochastic Eqs. (2.22) and (2.23) introduces additional problems due to the requirement of a prescription to simulate a Gaussian noise that depends on the variable.

III. CONTRIBUTIONS TO LINEAR-STABILITY ANALYSIS

A simple way to analyze some of the effects of the multiplicative noise is by means of a linear approximation on the equation of motion for the structure function. Certainly, this analysis is limited to short times after the quench, but it will give results which help us to understand the evolution of the long-wavelength instability.

The structure function $S(k,t)$ is defined as the Fourier transform of the correlation function

$$G(\mathbf{r},t) = \frac{1}{V}\int d\mathbf{r}'\langle c(\mathbf{r}+\mathbf{r}',t)c(\mathbf{r}',t)\rangle \ . \qquad (3.1)$$

By studying the behavior of $S(k,t)$ as a function of k, we can determine which modes grow or decay in the early stages of evolution. To do this explicitly, we choose a model which is described by a Ginzburg-Landau energy

$$F[c] = \frac{1}{2}\int d\mathbf{r}\left[-\frac{c^2}{2}+\frac{c^4}{4}+\frac{(\nabla c)^2}{2}\right] \qquad (3.2)$$

and by a mobility function given by Eq. (2.15), which corresponds, at the continuous limit, to the mobility of Eq. (1.3). The evolution of the correlation function can be obtained by the same method that was used in Appendix A for the equation of the first moment. By writing only the linear terms (that is, up to order c^2 in the equation for the correlation function) and performing the Fourier transform we have

$$\frac{d}{dt}S(k,t) = -k^2\left[k^2-1+\frac{4a\beta^{-1}}{R^d}\right]S(k,t)+2\beta^{-1}k^2$$

$$-2\beta^{-1}ak^2\frac{1}{(2\pi)^d}\int d\mathbf{q}\,S(q,t) \ . \qquad (3.3)$$

From this result one can conclude that, for the early stages of the evolution, those modes with $k<k_c=1-4a\beta^{-1}/R^d$ are unstable and grow with time. In contrast, the modes with $k>k_c$ relax. For $a=0$ (the case studied until now in the literature), $k_c=1$. Hence, the presence of the multiplicative noise reduces the domain of the unstable modes in the k space. This implies a delay in the domain-growth dynamics at an early stage. This is an explicit prediction of our theory that is confirmed by computer simulation.[17]

IV. CONCLUSIONS

In this paper we have derived Ginzburg-Landau equations for conserved and nonconserved order parameters with concentration-dependent mobility and multiplicative noise, which could be relevant in the context of phase-separation and domain-growth dynamics. These equations incorporate new terms of stochastic origin that give new contributions to the evolution of statistical properties. In particular, we have obtained new contributions to the Cahn-Hilliard theory.

Our derivations of the Fokker-Planck equations are based on coarse-grained procedures in a discrete lattice, and the corresponding Langevin equations are obtained by standard techniques of stochastic processes. A mesoscopic model, which contains a characteristic mesoscopic length, is introduced. This model is used in a numerical integration of the Ginzburg-Landau equations that is presented in a following paper. An expression for the mesoscopic model is also obtained in the continuous spatial limit. In this respect, we would like to remark that the dynamics depend on the characteristics of the particular mesoscopic model and it would be of interest to dedicate some effort to obtain such models from first principles.

APPENDIX A: CONSERVED ORDER PARAMETER

Here we present the mathematical details of the equivalence between the Fokker-Planck equation (2.6) and the Langevin equation (2.9). Although the derivation of the Fokker-Planck equation corresponding to a Langevin equation is standard, the case of the model derived in Sec. II presents special aspects owing to the presence of the gradient operators and the dependence of M on the concentrations of the whole configuration.

The first step is to write the Fokker-Planck equation (2.6) in a more familiar form:

$$\frac{\partial P(\{c\},t)}{\partial t} = -\Delta x^2 \Gamma \frac{\partial}{\partial c_\alpha}(\nabla_L^i)_{\alpha\beta}$$

$$\times \left[M_{\beta i}(\nabla_R^i)_{\beta\sigma}\frac{\partial F}{\partial c_\sigma} - \beta^{-1}m_{\beta i}(\nabla_R^i)_{\beta\sigma}\frac{\partial m_{\beta i}}{\partial c_\sigma} \right.$$

$$\left. + \beta^{-1}m_{\beta_i}(\nabla_R^i)_{\beta\sigma}\frac{\partial}{\partial c_\sigma}m_{\beta i} \right] P(\{c\},t) ,$$

(A1)

where the function $m(\{c\})$ was defined in Eq. (2.11). In Eq. (A1) we have made use of the property $(\nabla_R^i)_{\alpha\beta} = -(\nabla_L^i)_{\beta\alpha}$.

In this Fokker-Planck equation the first term is the deterministic drift, the second term is the "Stratonovich" spurious drift and the last is the Stratonovich-like diffusion term. From Eq. (A1) one can conjecture that the corresponding Langevin equation is given by Eq. (2.9). This is a stochastic differential equation in the Stratonovich interpretation, with a Gaussian white noise of zero mean and correlation given by Eq. (2.10).

Now, to prove that the Langevin equation (2.9) corresponds to the Fokker-Planck equation (2.6), we derive the latter from the former. According to van Kampen lemma,[20]

$$P(\{c\},t) \equiv \left\langle \prod_\alpha \delta[c_\alpha(t)-c_\alpha] \right\rangle \equiv \langle \rho(\{c\},t) \rangle ,$$

(A2)

and averaging the stochastic Liouville equation for $\rho(\{c\},t)$ we obtain

$$\frac{\partial P}{\partial t} = -\Delta x^2 \Gamma \frac{\partial}{\partial c_\alpha}(\nabla_L^i)_{\alpha\beta}$$

$$\times \left[M_{\beta i}(\nabla_R^i)_{\beta\sigma}\frac{\partial F}{\partial c_\sigma} - \beta^{-1}m_{\beta i}(\nabla_R^i)_{\beta\sigma}\frac{\partial m_{\beta i}}{\partial c_\sigma} \right] P$$

$$- \frac{\partial}{\partial c_\alpha}(\nabla_L^i)_{\alpha\beta}m_{\beta i}\langle \xi_\beta^i(t)\rho \rangle .$$

(A3)

Now, the average in Eq. (A3) is worked out with the aid of Novikov's theorem[21]

$$\langle \xi_\beta^i(t)\rho \rangle = -\Delta x^2 \beta^{-1}\Gamma \frac{\partial}{\partial c_\sigma}\left\langle \frac{\delta c_\sigma(t)}{\delta \xi_\beta^i(t')}\bigg|_{t'=t}\rho \right\rangle .$$

(A4)

The response function $\delta c/\delta\xi$ is obtained from Eq. (2.9):

$$\frac{\delta c_\sigma(t)}{\delta \xi_\beta^i(t')}\bigg|_{t'=t} = (\nabla_R^i)_{\sigma\beta}m_\beta .$$

(A5)

By substituting Eqs. (A4) and (A5) into Eq. (A3), we recover the Fokker-Planck equation (A1).

Both Eqs. (2.6) and (2.9) can be used indistinctly to derive the statistical properties of the system. As an example, the equation of the first moment is

$$\frac{\partial}{\partial t}\langle c_\mu(t) \rangle = \Delta x^2 \Gamma \left\langle (\nabla_L^i)_{\mu\beta}M_\beta(\nabla_R^i)_{\beta\sigma}\frac{\partial F}{\partial c_\sigma} \right\rangle$$

$$- \beta^{-1}\Gamma\Delta x^2 \left\langle (\nabla_L^i)_{\mu\beta}(\nabla_R^i)_{\beta\sigma}\frac{\partial M_\beta}{\partial c_\sigma} \right\rangle ,$$

(A6)

where the first term accounts for the mean of the drift and the second term contains the contributions of both the spurious term and the noise term of the Langevin, Eq. (2.9), which turn out to be identical. Equation (A6) reduces to Eq. (2.21) in the continuous limit.

APPENDIX B: NONCONSERVED ORDER PARAMETER

This case corresponds to the study of ferromagnetic systems in which the order parameter is not conserved. Now the change of concentration ϵ in a cell does not involve the neighboring cells. The transition probability, Eq. (2.3), has the same expression but the initial configuration is now

$$\{c\}_\alpha = \{c_1, c_2, \ldots, c_\alpha - \epsilon, \ldots\} \ .$$

We perform the same steps as in the former case, also reaching the Fokker-Planck equation (2.4) but with a different K_α operator, now given by

$$K_\alpha = -\frac{\partial}{\partial c_\alpha} \ . \tag{B1}$$

By taking this fact into account, we finally obtain

$$\frac{\partial P(\{c\}, t)}{\partial t} = \frac{\langle \epsilon^2 \rangle \beta}{2} \sum_\alpha \frac{\partial}{\partial c_\alpha} M_\alpha \left[\frac{\partial F}{\partial c_\alpha} + \beta^{-1} \frac{\partial}{\partial c_\alpha} \right]$$
$$\times P(\{c\}, t) \ . \tag{B2}$$

The continuous version of this equation is easily obtained:

$$\frac{\partial P}{\partial \tau} = \int d\mathbf{r} \frac{\delta}{\delta c} M(c) \left[\frac{\delta F}{\delta c} + \beta^{-1} \frac{\delta}{\delta c} \right] P \ , \tag{B3}$$

where we have defined a new time scale:

$$\tau = t \frac{\langle \epsilon^2 \rangle \beta \Delta V}{2} \ . \tag{B4}$$

From this last equation one can write the corresponding Langevin equation:

$$\frac{\partial c(\mathbf{r}, \tau)}{\partial \tau} = -M(c) \frac{\delta F}{\delta c} + \frac{\beta^{-1}}{2} \frac{\delta M(c)}{\delta c} + m(c) \xi(\mathbf{r}, t) \ ,$$
$$\tag{B5}$$

where $M(c) = m^2(c)$ is the mobility corresponding to this case. Actually there is no derivation of an explicit form for it. The noise is now a scalar with a correlation:

$$\langle \xi(\mathbf{r}, t) \xi(\mathbf{r}', t') \rangle = 2\beta^{-1} \delta(\mathbf{r} - \mathbf{r}') \delta(t - t') \ . \tag{B6}$$

A linear analysis of this case would give similar results to the conserved case, and the computer simulations can be implemented much more easily.[17]

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[19] The same identification given by Eq. (2.17) is applicable to more general forms of M and m, such as $M_{ai} = \sum_{\beta j} Q^{ij}_{\alpha\beta} f(c_\beta, c_{\beta+j})$ or $m_{ai} = \sum_\beta Q^i_{\alpha\beta} f(c_\beta)$.

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