From lattice-gas models to nonlinear diffusion models: A derivation of macroscopic equations and fluctuations

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We derive nonlinear diffusion equations and equations containing corrections due to fluctuations for a coarse-grained concentration field. To deal with diffusion coefficients with an explicit dependence on the concentration values, we generalize the Van Kampen method of expansion of the master equation to field variables. We apply these results to the derivation of equations of phase-separation dynamics and interfacial growth instabilities.

A rich variety of dynamic phenomena in pattern-formation theory[1-6] is described by phenomenological nonlinear partial differential equations. Nonlinear diffusion equations appear, for example, in interfacial growth instabilities,[1] spinodal decomposition in phase-separation dynamics,[2] and shock-wave propagation.[3] Other phenomena, such as the dynamics of fronts in liquid crystals,[4] flames and chemical reactions,[5] and convection in hydrodynamic flows[6] are described by reaction-diffusion or more complicated equations. The connection between a microscopic description of the systems and the corresponding macroscopic equations is a difficult task and has only been considered in a reduced number of simple situations.[7-10] This is the case of the derivation of the Cahn-Hilliard-Cook equation for spinodal decomposition[7] based on a coarse-grained procedure on a lattice-gas model. Other recent examples are the derivation of reaction-diffusion equations from lattice-gas models with the combination of conserved and nonconserved dynamics[8] and hydrodynamic equations from cellular automata.[10]

In the nonlinear diffusion equations, the diffusion coefficient is usually assumed to be a constant independent of the field variables. In the context of spinodal decomposition, it has been estimated[11] its expression is based on physical grounds, and it really depends on the field concentration variable. This explicit dependence might produce quite important changes on late-stage spinodal decomposition at low temperatures.[11] More recently, it has been pointed out[12,1] that the study of the effects of a uniform external field, such as gravity in spinodal decomposition[12] and interfacial growth instabilities,[1] may require the same type of generalization, giving rise to *modified* Cahn-Hilliard equations. The introduction of fluctuations in these models, by means of the standard coarse-grained derivations, presents a number of problems generated by the appearance of a multiplicative white-noise term in the resulting *modified* nonlinear Cahn-Hilliard-Cook equation.

In this Brief Report, we present a systematic method for deriving macroscopic nonlinear diffusion equations for coarse-grained fields and equations for the probability of fluctuations around the macroscopic evolution. The results are completely general, and they can be applied to situations that include field-dependent diffusion coefficients.

The standard phenomenological coarse-grained procedure[7] starts from a master equation governing the lattice-gas dynamics of interacting particles and derives a functional Fokker-Planck equation for the probability of a field concentration variable. To do so, the lattice is divided into cells of N sites and a coarse-grained concentration variable is defined for each cell. A main assumption in this derivation is that a master equation is also obeyed, at the coarse-grained level, by the probability of the configuration of cells. The corresponding Langevin equation could be immediately obtained from the Fokker-Planck equation. For example, in the case of a lattice-gas model with Kawasaki (conserved) dynamics, the result corresponds to the familiar B model of phase-separation dynamics,[2] also called the Cahn-Hilliard-Cook equation.

More recently, De Masi *et al.*[8] have addressed, from a more mathematical point of view, what conditions are required to obtain macroscopic equations from microscopic models. They have considered a lattice model in which two dynamics, Glauber (nonconserved) and Kawasaki, were competing. They have proved that if Kawasaki dynamics takes place in a microscopically faster scale than Glauber dynamics, a macroscopic reaction-diffusion equation is obeyed by the coarse-grained concentration field. Furthermore, the fluctuations of the concentration variable around the deterministic evolution have been obtained. They obey Gaussian statistics.

An interesting aspect of this derivation is that it exhibits an explicit implementation of the usual phenomenological assumptions at the level of the microscopic model. In this way, we can interpret the Kawasaki microscopic fast dynamics as inducing a local stirring, destroying correlations of spins inside the cells, and implying a situation of local equilibrium in the system. At the present moment, the rigorous method has not been applied to more general situations and, in particular, to the case for which only Kawasaki dynamics is considered, but a discussion on general aspects involved in this procedure has

been presented in Ref. 9.

A new situation appears when one tries to generalize the standard phenomenological coarse-grained procedure[7] to derive equations of models with field-dependent diffusion coefficients. In a direct application of this method, the resulting Fokker-Planck equation would give rise to a Langevin equation with a multiplicative white noise instead of the usual additive white noise. In this equation, the correct equilibrium solution, imposed by the fluctuation-dissipation theorem, would be ensured by the appearance of the diffusion coefficient in the fluctuating term. The interpretation of this multiplicative white-noise term requires a fixed prescription to resolve the Ito-Stratonovich dilemma. From both the Langevin and the equivalent Fokker-Planck equations it is possible to derive equations for dynamic properties, such as moments and correlation functions. In the resolution of these equations for the spinodal decomposition, the available theory[11] introduces an artificial dependence on the coarse-grained lattice constant. This is a simple way for avoiding the divergences induced by the special δ-correlated noise in this case of a nonlinear Langevin equation with additive noise. No renormalized equations have been obtained yet to avoid such a problem. In the case of nonlinear Langevin equations with multiplicative noise, it would not be surprising that problems of this type will increase. Here, we take a different approach.

One of our basic ingredients is the Van Kampen expansion of the master equation.[13] This method has been mainly used in situations without spatial inhomogeneities and here we generalize it to field variables. Our approach consists of two main steps. The first one is an expansion of the master equation for the probability of the cell configurations in powers of a small parameter that governs the size of the fluctuations. This parameter is the inverse of the number of spins in each cell, N^{-1}. The basic idea is that, in the limit of N going to infinity, no fluctuations would be present in the concentration of a cell. This would correspond to the deterministic macroscopic regime. In the next order of the expansion, we will obtain the first corrections due to fluctuations. The second step of our procedure deals with the particularities of the field variables. We have to introduce the macroscopic limit both in the spatial and temporal scales. Before implementing this step, we are at the level of the discrete cell space and we are describing the temporal scale of the individual occupancy cell dynamics. An interesting aspect of the method is that the same parameter N^{-1} controls the fluctuations in concentration and also the spatial and temporal rescaling. In this sense, we can talk about a unified macroscopic limit.

We start from a stochastic lattice-gas model of interacting particles. The dynamics of this model is given by a master equation for the probability $P(\{\sigma\},t)$ of a configuration $\{\sigma\}=\{\sigma_1,\sigma_2,\ldots\}$, where $\sigma_i=1,0$ indicates an occupied and an empty site of the lattice, respectively. Here, we will study Kawasaki dynamics, but the method could be, in principle, extended to Glauber dynamics or a combination of both. By using the standard coarse-grained procedure, one divides the lattice into regular cells of N sites and defines the concentration of the α cell by

$$C_\alpha = Nc_\alpha = \sum_{i,\epsilon\alpha} \sigma_i \ , \tag{1}$$

where C_α and c_α are "extensive" and "intensive" quantities, respectively. By considering a Markovian assumption, one can write a master equation for the probability $P(\{C\},t)$ of a configuration of cells, $\{C\}=\{C_1,C_2,\ldots\}$:

$$\partial_t P(\{C\},t) = \sum_\alpha \sum_i \sum_\epsilon [W(\{C\}^{\alpha i}\rightarrow\{C\})P(\{C\}^{\alpha i},t) - W(\{C\}\rightarrow\{C\}^{\alpha i})P(\{C\},t)] \ , \tag{2}$$

where the indexes α and i numerate the cells and their next neighbors respectively, ϵ is the concentration interchanged between two cells, and $W(\{C\}^{\alpha i}\rightarrow\{C\})$ is the transition probability between initial configuration $\{C\}^{\alpha i}=\{C_1,\ldots,C_\alpha-\epsilon,C_{\alpha+i}+\epsilon,\ldots\}$ and the final one $\{C\}=\{C_1,\ldots,C_\alpha,C_{\alpha+i},\ldots\}$. By assuming that the fluctuations are small, we express the intensive concentration c_α in the form

$$c_\alpha = c_\alpha + N^{-1/2}\xi_\alpha \ , \tag{3}$$

where c_α is the macroscopic concentration ($N\rightarrow\infty$) and ξ_α takes into account the corrections due to fluctuations.

In order to apply the expansion method, it is required that the transition probabilities have well-defined behavior for large N. Then it is natural to assume an expansion of W in powers of N^{-1} when W is expressed in terms of the intensive variable c. If one uses the fluctuation variable ξ instead of the variable c, from Eq. (3), one obtains, in general, an expansion of $W(\{C\}^{\alpha i}\rightarrow\{C\})\equiv\Phi((c+N^{-1/2}\xi)^{\alpha i};\alpha i,\epsilon)$ in powers of $N^{-1/2}$ as follows:

$$\Phi = T(N)(\Phi_0 + N^{-1/2}\Phi_{1/2} + N^{-1}\Phi_1 + \cdots) \ , \tag{4}$$

where $T(N)$ is a global factor that will be used to define a macroscopic time scale and Φ_j are general functions to be specified in each particular case. A similar expansion is valid for $W(\{C\}\rightarrow\{C\}^{\alpha i})\equiv\Phi((c+N^{-1/2}\xi);\alpha i,-\epsilon)$. Equations (3) and (4) are the basic ingredients of the expansion method. By taking into account that Eq. (3) implies that $P(\{c\},t)\equiv\Pi(\{\xi\},t)$ and substituting Eqs. (3) and (4) in Eq. (2), we obtain

$$\frac{\partial\Pi}{\partial t} - \sum_\beta N^{1/2}\frac{dc_\beta}{dt}\frac{\partial\Pi}{\partial\xi_\beta} = \frac{T(N)}{N}\sum_\alpha\sum_i D_\xi^{\alpha i}\left[N^{1/2}\Omega_{10}(\alpha i) + \sum_\beta\left[\frac{\partial}{\partial c_\beta}\Omega_{10}(\alpha i)\right]\xi_\beta + \Omega_{1\,1/2}(\alpha i) + \tfrac{1}{2}D_\xi^{\alpha i}\Omega_{20}(\alpha i)\right]\Pi \ , \tag{5}$$

where $D_{\xi}^{\alpha i}=(\partial/\partial\xi_{\alpha+i}-\partial/\partial\xi_{\alpha})$ and

$$\Omega_{mn}(\alpha i)=\sum_{\epsilon}\epsilon^{m}\Phi_{n}((c+N^{-1/2}\xi);\alpha i,\epsilon)|_{\xi=0} \tag{6}$$

are the moments of the transition probability (4). $\Omega_{mn}(\alpha i)$ could contain a dependence on $D_{\xi}^{\alpha i}$.

At this point, we start the second step of our procedure by taking the macroscopic limit in space and time. We use the rescaling of variables in a d-dimensional space given by

$$\tau=tT(N)/N, \quad x=x_{\alpha}/N^{1/d} . \tag{7}$$

The macroscopic time τ and space x could be interpreted as follows: in a τ interval many interchanges between different microscopic configurations are included to have a noticeable change at the macroscopic level; furthermore, in an x interval many cells are contained to ensure the spatial continuous limit. A similar rescaling was used in Ref. 9.

Now, we consider the different orders in the continuous version of the expansion (5). To the lowest order we get the macroscopic conservation equation for the concentration field

$$d_{\tau}c=a_{0}\nabla\Omega_{10}(c) , \tag{8}$$

where a_0 is the lattice spacing and the flux Ω_{10} is given by the first moment of the transition probability W. The next order gives a Fokker-Planck equation for the fluctuations

$$\frac{\partial\Pi}{\partial\tau}=-a_{0}\int dx\frac{\delta}{\delta\xi(x)}\nabla\left[\left[\frac{\partial}{\partial c}\Omega_{10}(c)\right]\xi(x)+\Omega_{1/2}(c)+\tfrac{1}{2}a_{0}^{1+d}\nabla\frac{\delta}{\delta\xi(x)}\Omega_{20}(c)\right]\Pi . \tag{9}$$

This is a linear equation with time- and space-dependent coefficients that evolve according to the macroscopic Eq. (8). Equations (8) and (9) are the main results of this paper.

Now, we present an example which is a prototype of field models in phase separation.[2] The transition probability is given by

$$W(\{C\}^{\alpha i}\rightarrow\{C\})=M(\{C\}^{\alpha i},\{C\})\exp(\tfrac{1}{2}\Delta F) , \tag{10}$$

where $\Delta F=F(\{C\}^{\alpha i})-F(\{C\})$ and $F(\{C\})=F_{\mathrm{GL}}(\{C\})+F_{h}(\{C\})$ is the Ginzburg-Landau functional free energy[7] plus any contribution from an external field. Furthermore, $M(\{C\}^{\alpha i},\{C\})$ has to be a symmetric function of its arguments to ensure that detailed balance holds. Here, we assume the simplest form $M(\{C\}^{\alpha i},\{C\})=m(\{C\}^{\alpha i})m(\{C\})$. It has been obtained[11] from a phenomenological theory that $m(\{c\})\equiv m(c)=[c(1-c)]^{1/2}$. For late-stage spinodal decomposition at low temperatures, the concentration values fluctuate approximately from 1 to 0 and then a reduction in the diffusion process could be present. In the literature it is usual to take $M=p(\epsilon)$, where $p(\epsilon)$ is a symmetric function of the interchanges. This last assumption implies a constant diffusion coefficient. From Eq. (10), one can obtain the corresponding equations (8) and (9) for this case.

In the standard phenomenological approach, and after the Fokker-Planck approximation is obtained, an expansion in powers of ϵ and a truncation to the second moment of $p(\epsilon)$ is done. To compare with these results, we make now the same type of approximation in the resulting equations. Then, we obtain

$$\frac{dc}{d\tau}=\tfrac{1}{2}\langle\epsilon^{2}\rangle a_{0}^{2+d}\nabla M_{0}(c)\nabla\frac{\delta F(c)}{\delta c} , \tag{11}$$

$$\frac{\partial\Pi}{\partial\tau}=-\tfrac{1}{2}a_{0}^{2+d}\langle\epsilon^{2}\rangle\times\int dx\frac{\delta}{\delta\xi(x)}\nabla\left[\frac{\partial}{\partial c}\left[M_{0}(c)\nabla\frac{\delta F(c)}{\delta c}\right]\xi(x)+M_{0}(c)\nabla\frac{\delta}{\delta\xi(x)}\right]\Pi , \tag{12}$$

where $\langle\epsilon^{2}\rangle$ is the second moment of $p(\epsilon)$. Equation (11) is a nonlinear diffusion equation for the macroscopic field c with a diffusion coefficient $M_0(c)=m^2(c)$. It was proposed by different authors.[11,12,1] Furthermore, our approach allows the study of fluctuations around the macroscopic evolution by means of the Fokker-Planck equation (12). This is a linear equation with time- and position-dependent coefficients and with additive white noise obeying a fluctuation-dissipation theorem with a diffusion coefficient $M_0(c(x,t))=M_0(x,t)$. The presence of an external field such as gravity is contained in Eqs. (11) and (12) thanks to the presence of $M_0(c(x,t))$, as can be seen by taking into account that the free energy of a uniform gravitational field is linear in the spatial coordinates. This contribution would be cancelled after two consecutive spatial derivatives if M_0 did not depend on c.

To conclude, we have presented a systematic method to derive macroscopic equations for field variables and equations for the fluctuations around the macroscopic solution. The treatment could be useful in situations for which small fluctuations are needed to initiate the evolution of the processes that later will evolve around the nonlinear macroscopic solutions. This is the case, for example, in spinodal decomposition for which the evolution from the initial unstable state is dominated by the macroscopic deterministic equation.[14] These equations could be applicable to describe late-stage spinodal decomposition and the effects of an external field in spinodal decomposition and interfacial growth phenomena. The treatment of nonlinear equations with multiplicative white noise that could be derived by the application of the standard coarse-grained procedure remains open. Work on this subject is in progress.

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