Interface dynamics in a mean-field lattice gas model: Dendritic growth

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Abstract

We study the dynamics of a lattice gas model by means of a mean-field kinetic equation. Phase separation and dendritic growth are observed. We calculate the orientation-dependent surface tension and show that our model is consistent with the standard theory of dendritic growth. Finally, interface kinetics are investigated.

1 Introduction

Pattern formation in systems out of equilibrium is an old subject. Classical fields of study are phase separation dynamics (spinodal decomposition) [1, 2] and dendritic growth [3]. The former occurs when a system is quenched in an unstable state: fluctuations are strongly amplified, and a complicated pattern of interwoven domains develops, which then coarsens. The latter appears during the growth of a stable phase into a metastable one, for example during crystallization from a supersaturated solid solution. Both processes are technologically interesting in the field of metallurgy, because they affect the microstructure of materials. In spite of great theoretical advances in recent years, many questions remain open.

The classical phenomenological approach is the use of continuum models. The evolution is decribed by partial differential equations: the Cahn-Hilliard equation [4] for spinodal decomposition, and diffusion equations with suitable conservation laws and a Gibbs-Thomson boundary condition at the (sharp) interface for dendritic growth [3]. In statistical physics, it is highly interesting to find simple models showing the same behavior, and to recover the macroscopic equations in the hydrodynamic limit. For example it is known [2] that the kinetic Ising model with spin-flip (exchange) dynamics reproduces the phase separation dynamics of a system with a non-conserved (conserved) scalar order parameter in the large time limit.

For dendritic growth, the situation is less clear. In contrast to phase separation, where the late stage evolution is driven only by surface energy, there is a thermodynamic force associated to the conversion of the metastable into the stable phase (in the above example, this force is proportional to the supersaturation). This leads to a continuos amplification of noise. We encounter the well-known Mullins-Sekerka instability [5], which leads to formation of fingers by growth of the long-wavelength perturbations of a smooth surface, the short-wavelength cutoff being provided by the surface tension. In the presence of anisotropy, dendrites develop in some stable growth directions, which are given by the maxima of the surface tension. For vanishing anisotropy, on the other hand, we encounter the "seaweed" type of growth [6].

A paradigm for microscopic growth models is diffusion-limited aggregation (DLA) [7]. It is known that it produces irregular shapes, which means that it can be no model for dendritic growth if we interpret the walkers as individual atoms (it may be a large-scale

representation for isotropic aggregates, with the size of one walker being the typical diameter of a finger). This is due to the strong noise and the absence of a local equilibrium condition at the interface: a candidate for a microscopic dendritic growth model should present a well-defined anisotropic surface tension and a Gibbs-Thomson boundary condition at the interface. Progress in this direction has been made by the introduction of noise-reduced DLA and by models which incorporate curvature effects [8]. But in all the cases we are aware of, the desired effect is achieved by some algorithmic prescription, and there is no precise relation between the model parameters and the surface tension.

We want to present a model which is based on a stochastic lattice gas, constituting a very simple model of a binary alloy. We establish its kinetic equation by a mean-field method recently developed by one of the authors and subsequently applied to an order-disorder transition [9]. This equation of motion takes the form of a generalized Cahn-Hilliard equation on the lattice. We can show that this model has a Gibbs-Thomson boundary condition and we compute the surface tension. It presents an anisotropy which is related to the lattice structure and the microscopic interactions. Simulations show indeed the formation of nice "snowflakes". Our theory allows to understand the selection of growth directions in terms of the microscopic interactions of the model.

A second point of interest are interface kinetics. Usually, the static Gibbs-Thomson boundary condition is supplemented with a kinetic term expressing the departure from local equilibrium due to the advance of the interface. For small interface velocities it is linear in the velocity, and the knowledge of the orientation-dependent proportionality constant completely determines the growth process. Our model shows the same behavior, and we investigate it by numerical simulations and analytically, following ideas of Langer and Sekerka [10]. This method can be used to calculate the interface mobility. In addition, we find the effect of solute trapping, which is well documented in rapid solidification experiments.

Our model, which is isothermal, could describe the formation of alloy dendrites, where heat diffusion can often be neglected and the diffusion of solute is the rate-limiting process. It bears some similarities to the phase-field models developed in recent years [11]. But in our case, there is only one dynamic field (the concentration). This avoids the splitting between phase- and diffusion field and the introduction of sometimes arbitrary coupling terms. Our equation incorporates naturally diffusion and aggregation.

After a presentation of the model, we will sketch the calculation of the surface tension and of the kinetic terms; more details can be found in [12].

2 Lattice gas model and kinetic equation

Consider a square lattice in two dimensions (coordination number $z = 4$). $n_i \in \{0, 1\}$ is the occupation number at site i. Thus we exclude multiple occupation. The Hamiltonian of a configuration $\{n_i\}$ is given by

$$H = -\epsilon \sum_{\langle i,j \rangle} n_i n_j - \mu_0 \sum_{i=1}^{N} n_i, \tag{1}$$

where the interaction energy ϵ is a positive constant, μ_0 is an external chemical potential, the first sum goes over all nearest neighbor pairs, and N is the number of lattice sites. This is equivalent to the Ising model, and also to a simple binary (AB) alloy. Then, particles correspond to atoms of type A, holes to atoms of type B.

We assume that particles can move only via nearest-neighbor-hops (exchange dynamics). The system is in contact with a heat bath: temperature is constant, energy is not conserved.

Particles can be introduced or taken out only at the boundaries. To define the jump probabilities, we imagine that the lattice points are located at the equilibrium positions of the atoms in a crystal structure. Then an atom is trapped at its site in a potential well. If all the saddle points are at the same energy, the depth of the potential well is determined by the binding energy of the atom. We assume the jump probabilities to follow an Arrhenius law: the transition rate from site i to site k is given by

$$w_{i\to k}(\{n\}) = w_0 \exp\left(-\frac{\epsilon}{k_B T}\sum_a n_{i+a}\right). \tag{2}$$

w_0 is an attempt frequency which sets the overall time scale and will be set to unity in the following, k_B is the Boltzmann constant, and T is temperature. a denotes a lattice vector, and summation over a means summation over all bonds connected to i. This defines a stochastic process, which clearly includes DLA in the limit $T \to 0$. Let's remark that this expression differs from the usual Monte Carlo rule, which uses the energy difference between initial and final state. This introduces an asymmetry between the phases which is absent in the usual Ising model with exchange dynamics: here, the diffusion is slower in the dense phase.

We now write a local master equation. The local occupation probabilities $p_i = \langle n_i \rangle$ (the average is temporal) obey a conservation law:

$$\frac{\partial p_i}{\partial t} = -\sum_a j_{i,i+a}, \tag{3}$$

where the current in the bond ik is defined by

$$j_{i,k} = \langle n_i(1-n_k)w_{i\to k}(\{n\}) - n_k(1-n_i)w_{k\to i}(\{n\})\rangle. \tag{4}$$

The prefactors ensure that the start site is occupied and the target site is empty. In the simplest mean-field approximation, we now replace everywhere the operators by their mean values, that is n_i by p_i. The stochastic process is changed into a deterministic set of coupled nonlinear differential equations. Clearly, this completely alters its properties; in particular, we do not recover DLA in the low-temperature limit. But as usual we can hope to get a first qualitative insight, especially near the critical temperature, by this simple treatment. As we will see, already the mean-field equations are complicated and have interesting properties. Possible improvements could be obtained using more sophisticated mean-field-like theories like the path-probability method or dynamical density functionals [13]. This is currently under study.

Making some algebraic rearrangements (see [9] for details), we can rewrite the conservation equation in the following form:

$$\frac{\partial p_i}{\partial t} = \sum_a M_{i,i+a}(\mu_{i+a} - \mu_i). \tag{5}$$

μ_i is a local chemical potential, deriving from a free energy F:

$$\mu_i = \frac{\partial F}{\partial p_i} = -\epsilon\sum_a (p_{i+a} - p_i) - z\epsilon(p_i - 1/2) + k_B T \log\frac{p_i}{1-p_i}. \tag{6}$$

The first term is a discretized Laplacian, the other terms are local; the free energy is given by

$$F(\{p\}) = \sum_{k=1}^{N}\left(\frac{\epsilon}{4}\sum_a (p_{k+a} - p_k)^2 + f(p_k)\right) \tag{7}$$