

## Ginzburg-Landau theory for mass transport in a driven diffusive system

Hans C. Fogedby and Axel Svane

*Institute of Physics, University of Aarhus, 8000 Aarhus C, Denmark*

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A time-dependent Ginzburg-Landau theory is developed in order to characterize the results of a recent computer simulation on a lattice gas driven by an applied chemical potential gradient acting on the boundaries of the system. In accordance with the simulation results, the system shows a linear density profile above the critical temperature and phase separation below the critical temperature. It is furthermore shown that the driven system is equivalent to an equilibrium system in an applied linearly dependent chemical potential proportional to the current.

### I. INTRODUCTION

There is a current interest in phase transitions and critical phenomena of microscopic statistical-mechanical models driven in steady states far from thermodynamic equilibrium. In contrast to the situation for equilibrium phase transitions and critical phenomena where well-developed theoretical constructs are available, the present understanding of kinetic phase transitions is limited and mostly based on computer-simulation studies of simple microscopic models.

A particular microscopic model which has been studied extensively is the driven two-dimensional lattice gas with diffusive dynamics subject to an external uniform field.<sup>1-8</sup> Computer simulations have shown that this system undergoes a kinetic phase transition whose properties depend on the field strength. The character of the phase transition has also been elucidated using mean-field theories and renormalization-group calculations.<sup>4,6,8</sup>

Leaving aside the fact that kinetic phase transitions in driven systems are abundant in nature ranging from metallurgy to physics, chemistry, and biology, one of the important theoretical questions is, of course, whether kinetic phase transitions can be characterized by critical exponents and whether they allow for a classification in terms of universality classes similar to the scheme for equilibrium phase transitions.

Recently a model of mass transport in a two-dimensional lattice gas driven by an external chemical potential gradient was proposed.<sup>9</sup> Simulation studies show that this model also exhibits a kinetic phase transition whose properties depend on the chemical potential and the temperature. The model differs in important respects from the driven lattice gas in a field in that the driving force responsible for the flow only acts on the boundaries of the system. In the bulk region a Kawasaki-type Metropolis updating scheme<sup>10</sup> ensures local particle conservation as well as contact with a constant-temperature heat bath. Except for the mass current imposed at the edges of the system by means of a chemical potential in conjunction with a periodic boundary condition, there is no further coupling to the thermodynamic degrees of freedom.

In this paper we attempt to adapt a simple Ginzburg-Landau-type mean-field description together with a Langevin transport equation in order to describe some of

the features observed in the simulation study in Ref. 9. We are able to account for the linear density profile above the critical temperature and the phase separation below. We furthermore believe that the kinetic phase transition seen in the simulation is a particularly simple one since the thermodynamics and kinetics are equivalent to an equilibrium system in an applied linearly varying chemical potential.

### II. GINZBURG-LANDAU THEORY

We take as our starting point the usual Ginzburg-Landau form for the free energy,<sup>11</sup>

$$F = \int_{-L/2}^{L/2} dx \left[ \frac{1}{2} \left( \frac{dn(x)}{dx} \right)^2 + \frac{1}{2} r [n(x) - n_0]^2 + \frac{1}{4} u [n(x) - n_0]^4 \right]. \quad (1)$$

This form is appropriate as a coarse-grained description of a lattice-gas model with attractive interactions, corresponding to the ferromagnetic case in the Ising model analogue. We note that most of the simulations reported in Ref. 9 are for the repulsive lattice gas, however, as will become clear our main conclusion does not depend on the specific form of the free energy in Eq. (1) and we use for convenience the above generic form of  $F$ .

The order parameter  $n(x)$  is the spatially varying density and  $n_0$  the uniform average density. In order to model the simulations study in Ref. 9 as closely as possible we only assume a density variation in the  $x$  direction and consider a slab of thickness  $L$ .

#### A. Equilibrium properties

The equilibrium properties of the system described by  $F$  follow in the usual manner<sup>11</sup> by assuming  $u > 0$  and  $r \propto (T - T_c)$ , where  $T_c$  is the critical temperature, and minimizing  $F$  with respect to variations of  $n(x)$ . Above  $T_c$  the system has a single phase with density  $n_0$ . At  $T_c$  the system undergoes a second-order phase transition and can exhibit two possible uniform phases below  $T_c$  with densities

$$n_1 = n_0 + (-r/u)^{1/2}, \quad n_2 = n_0 - (-r/u)^{1/2}. \quad (2)$$

In an external field the system shows phase separation. Following Ref. 9 we impose opposite chemical potentials at the boundaries of the system, i.e.,  $\pm\mu$  at  $x = \mp L/2$ , corresponding to a symmetry-breaking term

$$-\mu(x)n(x) = \mu(\delta(x - L/2) - \delta(x + L/2))n(x) \quad (3)$$

in the free-energy density in Eq. (1).

Ignoring for the moment boundary layer and interface contributions to  $F$ , that is terms due to the presence of  $\frac{1}{2}(dn/dx)^2$  in Eq. (1), the chemical potential has no effect on the uniform phase  $n_0$  above  $T_c$ , whereas the system phase separates below  $T_c$  with a nonuniform density profile  $n(x) = n_2$  for  $x_0 < x < L/2$  and  $n(x) = n_1$  for  $-L/2 < x < x_0$ . In this approximation the position  $x_0$  of the interface is undetermined.

In order to correctly account for the boundary layer and interface profile we include the term  $\frac{1}{2}(dn/dx)^2$  in Eq. (1), and the minimum condition  $\delta F/\delta n = 0$  yields<sup>11</sup>

$$-\frac{d^2n(x)}{dx^2} + r[n(x) - n_0] + u[n(x) - n_0]^3 = -\mu(x). \quad (4)$$

The character of the solutions to Eq. (4) are well known (see, e.g., Ref. 12). Above  $T_c$  the uniform phase  $n_0$  is stable in the bulk region. At the boundaries the density  $n(x)$  adjusts to the chemical potentials and varies in an exponential fashion over a range given by the correlation length  $\xi = (1/|r|)^{1/2}$ . Below  $T_c$  the interface is smooth over a distance given by the correlation length  $\xi$ . In the bulk region the nonuniform density profile is symmetric about  $x = 0$  and is given by the kink solution<sup>12</sup>

$$n(x) = n_0 - (-r/u)^{1/2} \tanh(x/\xi\sqrt{2}). \quad (5)$$

At the boundaries the densities  $n_1$  and  $n_2$  are again adjusted to the chemical potential over a transition region of size  $\xi$ . This concludes our summary of the equilibrium properties of the model in Eq. (1) in a finite geometry with imposed chemical potentials pertinent to the simulation study in Ref. 9.

### B. Transport properties

The transport properties of the model in Eq. (1) are conveniently discussed by introducing the usual time-dependent Ginzburg-Landau description.<sup>11</sup> Considering only variations in the  $x$  direction and ignoring fluctuations due to the heat bath we have quite generally the conservation law

$$\frac{dn}{dt} = -\frac{dj}{dx}, \quad (6)$$

together with a model-dependent constitutive equation in terms of  $F$  for the current  $j$ ,

$$j(x) = -\Gamma \frac{d}{dx} \frac{\delta F}{\delta n(x)}, \quad (7)$$

$\Gamma$  being the damping coefficient. With a noise term included Eqs. (1), (6), and (7) constitute the so-called model  $B$  proposed in Ref. 13 in connection with dynamical critical phenomena, i.e., the case of a conserved order pa-

rameter. Here we use the equations in order to model the simulation study in Ref. 9.

Since the driving field (that is the chemical potentials) only acts at the boundaries of the system it is not included in Eqs. (6) and (7) which aim at describing the density profile in the bulk region. This situation is quite different from the driven lattice gas in an electric field  $E$ .<sup>1-3</sup> Here the field acts everywhere in the bulk region and is included in the simulations<sup>1-3</sup> by means of a biased  $E$ -dependent Metropolis updating scheme, and in the field theoretic analysis<sup>4,6</sup> by an additional current contribution in the transport equations (6) and (7).

Our analysis now proceeds in a very simple manner. Assuming a steady-state density profile Eq. (6) implies a constant current  $j_0$  throughout the system. Integrating Eq. (7) for a constant current  $j_0$ , inserting  $\delta F/\delta n$  extracted from Eq. (1), and assuming an inflection point in the density profile at  $x = 0$ , we obtain the mean-field equation describing the density profile in the driven system:

$$-\frac{d^2n}{dx^2} + r[n(x) - n_0] + u[n(x) - n_0]^3 = -\frac{j_0x}{\Gamma}. \quad (8)$$

The first thing we notice is that Eq. (8) also follows from the equilibrium condition  $\delta \bar{F}/\delta n = 0$  for the modified free energy

$$\bar{F} = \int_{-L/2}^{L/2} dx \left[ \frac{1}{2} \left( \frac{dn(x)}{dx} \right)^2 + \frac{1}{2} r[n(x) - n_0]^2 + \frac{1}{4} u[n(x) - n_0]^4 + \frac{j_0x}{\Gamma} n(x) \right], \quad (9)$$

and we conclude that as far as the density profile is concerned the driven system is equivalent to an equilibrium system with a linearly varying chemical potential  $-j_0x/\Gamma$ . This equivalence does not depend on the specific form of  $F$  but follows alone from Eqs. (6) and (7). The mass transport is essentially decoupled from the heat diffusion. Drawing the parallel to the simulation study in Ref. 9, the Kawasaki updating ensures local thermal equilibrium according to a free energy or Hamiltonian of the form in Eq. (9) and at the same time maintains the mass transport. The imposed mass current enters as a spatially varying chemical potential  $-j_0x/\Gamma$ . The above situation should apply to physical systems where the rate of mass diffusion is much slower than the rate of heat diffusion in such a way that the system rapidly attains thermal equilibrium. Matching the effective chemical potential in Eq. (9) to the applied chemical potential  $\mu$  we derive the relationship

$$j_0 = 2\mu\Gamma/L \quad (10)$$

for the current.

The discussion of the density profile  $n(x)$  now follows easily from Eq. (8). Ignoring for the moment rapid density variations, that is assuming  $d^2n/dx^2$  small, we obtain above  $T_c$  a density profile which, in the bulk region near  $x = 0$ , is linear about  $n_0$  with a slope

$$j_0/r\Gamma \propto \frac{1}{T - T_c}. \quad (11)$$

Below  $T_c$  a simple graphical analysis shows that the sys-

tem phase separates with densities  $n_1$  and  $n_2$  at  $x=0$ .

As in the discussion of the equilibrium properties in Sec. IIA the term  $d^2n/dx^2$  is necessary in order to correctly account for the detailed spatial variation of  $n(x)$ . The interface at  $x=0$  is continuous over a transition region of size

$$\xi = (1/|r|)^{1/2} \propto |T - T_c|^{-1/2} \quad (12)$$

and given by the kink solution in Eq. (5). Similarly, at the boundaries the density profile adjusts to the applied chemical potential over a range  $\xi$ . These findings are in qualitative agreement with the simulation study reported in Ref. 9. In Fig. 1 we have shown the density profiles above and below the critical temperature. By inserting Eq. (8) in Eq. (9) it is easily seen that the dashed curve corresponds to unstable and metastable solutions which are unphysical in the present context.

Finally we note that in the linear density regime the diffusion constant  $D$  inferred from the relationship  $j = -D dn/dx$  has the form

$$D = \Gamma |r| \propto |T - T_c|, \quad (13)$$

and hence varies linearly with  $|T - T_c|$ . This result is also in qualitative agreement with Ref. 9.

### III. SUMMARY AND CONCLUSIONS

We have developed a simple mean-field theory based on the time-dependent Ginzburg-Landau theory in order to capture some of the features observed in a recent simulation study of a lattice gas driven by a chemical potential gradient acting on the boundaries of the system.

In accordance with the simulation we find a linear density profile above the critical temperature and phase separation below the critical temperature. We furthermore show that the model is equivalent to an equilibrium system described by the same free energy  $F$  but with an applied linearly dependent chemical potential proportional

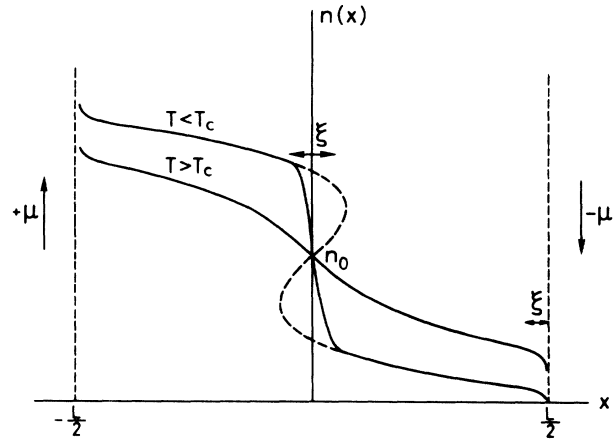


FIG. 1. The density profiles above and below the critical temperature. The linear regime is in the vicinity of  $x=0$ . The dashed curve indicates the unphysical region (arbitrary units).

to the current. This simple observation indicates that the phase transition reported in the simulation studies might be of a particular simple kind which falls in the universality class of the equivalent equilibrium transition.

We finally wish to emphasize the obvious limitations of the present approach. Since we adhere to a continuum formulation of the model and furthermore neglect fluctuation effects we are, of course, unable to account for features which are due to lattice effects such as a saturation current and percolation threshold effects, as reported in Ref. 9.

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