

Van der Waals Limit and Phase Separation in a Particle Model with Kawasaki Dynamics

Giambattista Giacomin¹

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A one-dimensional interacting particle system with a stochastic dynamics is studied in the local mean field limit, extending the results of Lebowitz, Orlandi, and Presutti to processes which satisfy detailed balance (with respect to Gibbs measures). The behavior of the system below the critical temperature and inside the unstable (spinodal) region is then investigated by means of computer simulations. The experiments clearly indicate the presence of phase separation and confirm the validity of some conjectures on the dynamics of the spinodal decomposition.

KEY WORDS: Kawasaki dynamics; Lebowitz–Penrose limit; spinodal decomposition.

1. THE PROBLEM

In this paper I extend the results proven in ref. 6 for a nonreversible particle model to its reversible version, introduced in Section 3 of ref. 6. This describes a jump process on a lattice gas with the exclusion condition [i.e., $\eta(x, t)$, the particle number at site x and time t , is 0 or 1]. The jump intensity from x to $x + b$ ($b = \pm 1$) is given by

$$c(x, x + b; \eta) = \frac{1}{2} [\eta(x)(1 - \eta(x + b))] \exp \left\{ -\frac{\beta}{2} [H_\gamma(\eta^{x, x+b}) - H_\gamma(\eta)] \right\} \quad (1.1)$$

We use the same notation as in ref. 6. $H_\gamma(\eta)$ is the Hamiltonian with pairwise interaction $V_\gamma(|x - y|)$ and V_γ is a Kac potential, $V_\gamma(|x - y|) = \gamma V(\gamma|x - y|)$. The choice (1.1) makes the evolution reversible with respect to the Gibbs measure with potential V_γ and inverse tem-

¹ Dipartimento di Fisica, Università di Pisa, Pisa, Italy.

perature $(k\beta)^{-1}$ (k is the Boltzmann constant). The process is a Kawasaki process with Hamiltonian H_γ . In the limit $\gamma \rightarrow 0$ Lebowitz and Penrose have proved^(7,8) that, if the integral of V is negative (we assume that V is regular and L^1), there is a phase transition, in the sense of the van der Waals Maxwell theory, namely that there is a critical value β_c , and for $\beta > \beta_c$ the infinite-volume free energy has a limit for $\gamma \rightarrow 0$ which is still convex, but not strictly convex. The phase transition region is the interval $[A, B]$, with A and B , respectively, given by $1/2 - u^*$ and $1/2 + u^*$ [u^* given by (4.4)], and the unstable region is $[a, b] \subset [A, B]$, with a and b equal to I_β^- and I_β^+ [I_β^\pm are given by (2.1e)]. I shall consider an initial measure μ_0^ε , which is a product measure whose average occupation numbers vary on a scale ε^{-1} , and a lattice Γ^ε of size $2\varepsilon^{-2}$, so that, in the limit $\varepsilon \rightarrow 0$, it is infinite also in macroscopic units (ε^{-1}). I will choose $\gamma = \varepsilon^{1-\alpha}$ so that the macroscopic scale ε^{-1} is much larger than γ^{-1} , the range of interaction.

In the next section I will state the theorems that prove that at time $O(\varepsilon^{-2})$ propagation of chaos holds and the average density converges to the nondegenerate equation found in ref. 6, if the initial density profile does not intersect the spinodal region. In Section 3, I sketch the proof of this theorem and in Section 4, I present some computer simulations on the behavior of a similar model in the spinodal region.

2. RIGOROUS RESULTS IN THE HYDRODYNAMIC LIMIT

We shall see that the system has a diffusive behavior described by the nonlinear partial differential equation

$$\frac{\partial \rho}{\partial t}(r, t) = \frac{\partial}{\partial r} \left[D_\beta(\rho(r, t)) \frac{\partial \rho}{\partial r}(r, t) \right] \tag{2.1a}$$

$$\rho(r, 0) = \rho_0(r) \in \mathcal{X}_\beta, \quad \forall r \in \mathbf{R} \tag{2.1b}$$

where

$$D_\beta(\rho) = \frac{1}{2} - \frac{\beta I}{2} \rho(1 - \rho) \tag{2.1c}$$

$$I = \int_0^\infty 2rV'(r) dr = -2 \int_0^\infty V(r) dr \tag{2.1d}$$

and

$$\mathcal{X}_\beta = \begin{cases} [0, I_\beta^-] \cup (I_\beta^+, 1] & \text{if } \beta \geq \beta_c = 4/I \\ \left[I_\beta^\pm = \frac{1}{2} \pm \frac{1}{2} \left(1 - \frac{4}{\beta I} \right)^{1/2} \right] & \\ [0, 1] & \text{if } \beta < \beta_c = 4/I \end{cases} \tag{2.1e}$$

To have a better insight into the meaning of this result in connection with equilibrium statistical mechanics, see Section 1 and Fig. 1 (Section 4) of this paper, and Section 3 of ref. 6. As in ref. 6, we assume that $\rho_0(r) \in C^\infty(\mathbf{R})$ with uniformly bounded derivatives and for each $\varepsilon > 0$ we consider the function $\tilde{\rho}_0^\varepsilon(r)$ ($|r| \leq \varepsilon^{-1}$) which is also C^∞ in $|r| \leq \varepsilon^{-1}$ with periodic boundary conditions having also uniformly bounded derivatives. We also require that $\tilde{\rho}_0^\varepsilon(r) = \rho_0(r)$ for all r such that $|r| \leq \varepsilon^{-1}$. We assume that the process starts from μ_0^ε , which is a product measure on $X = \{0, 1\}^{\Gamma^\varepsilon}$, $\Gamma^\varepsilon = [-\varepsilon^{-2}, \varepsilon^{-2}] \cap \mathbf{Z}$ with

$$\mu_0^\varepsilon(\{\eta(x) = 1\}) = \tilde{\rho}_0^\varepsilon(\varepsilon x) \tag{2.2}$$

We have the following:

Theorem 2.1 (Propagation of chaos). Given ρ_0 , $\tilde{\rho}_0^\varepsilon$, μ_0^ε , and $\gamma = \varepsilon^{1-\alpha}$ as before, there is α (sufficiently small) such that for all n and for all $\tau \in \mathbf{R}^+$

$$\lim_{\varepsilon \rightarrow 0} \sup_{x_1, \dots, x_n} \left| \mathbf{E}_{\mu_0^\varepsilon}^\varepsilon \left(\prod_{i=1}^n \eta(x_i, \varepsilon^{-2}\tau) - \prod_{i=1}^n \tilde{\rho}^\varepsilon(\varepsilon x_i, \tau) \right) \right| = 0 \tag{2.3}$$

the sup being taken over n different sites of Γ^ε , and $\tilde{\rho}^\varepsilon(\varepsilon x_i, \tau)$ is the solution of (2.1) with initial condition $\tilde{\rho}_0^\varepsilon(r)$ periodic over the whole \mathbf{R} and of period $2\varepsilon^{-1}$. Furthermore, $\tilde{\rho}_0^\varepsilon(r, \tau) \rightarrow \rho(r, t)$ uniformly on the compacts and faster than any power of ε .

For fixed $\delta \in (0, 1)$ we define the averages

$$M(x, t, \varepsilon^{-\delta}) \equiv \varepsilon^\delta \sum_{|y-x| \leq \varepsilon^{-\delta/2}} [\eta(y, t) - \rho(\varepsilon y, \varepsilon^2 t)]$$

We have:

Theorem 2.2 (With the same notation as in Theorem 2.1). For all $\delta \in (0, 1)$ there is $\xi > 0$ such that for all $\tau', \tau'' (0 < \tau' < \tau'')$ and for all $R \in \mathbf{R}^+$ we have

$$\lim_{\varepsilon \rightarrow 0} \mathbf{P}_{\mu_0^\varepsilon}^\varepsilon \left(\sup_{\tau' < \varepsilon^2 t < \tau''} \sup_{|x| \leq \varepsilon^{-1} R} |M(x, t, \varepsilon^{-\delta})| > \varepsilon^\xi \right) = 0 \tag{2.4}$$

and the convergence in (2.4) is faster than any power of ε .

The two theorems above extend the result in ref. 6 to the process generated by L_γ^β . The extension of the proofs presents a few nontrivial points, as briefly sketched below.

3. SKETCH OF THE PROOFS

In this section I lean heavily on refs. 2, 3, and 6. The proofs of Theorems 2.1 and 2.2 are based on *short-time estimates* on the functions

$$v_n(\underline{x}, t | \eta) = \mathbf{E}_\eta^\varepsilon \left(\prod_{i=1}^n (\eta(x_i, t) - \rho(x_i, t)) \right) \quad (3.1)$$

where $\mathbf{E}_\eta^\varepsilon$ denotes the expectation over the process generated by L_γ^β [given explicitly in (3.3)] and starting from η , $\underline{x} \in \mathcal{M}_n \equiv \{(x_1, \dots, x_n) \mid x_i \neq x_j \forall i \neq j\}$, and the density $\rho(x, t)$ is the solution of the discrete differential equation

$$\begin{aligned} \frac{\partial \rho}{\partial t}(x, t) &= \frac{1}{2} \Delta_1 \rho(x, t) - \frac{\beta}{4} \gamma^2 \nabla_1^+ \left\{ \rho(x-1, t) [1 - \rho(x, t)] \right. \\ &\quad \times \sum_r \rho(x-1+r, t) s(r) \tilde{V}'(r) - \rho(x, t) [1 - \rho(x-1, t)] \\ &\quad \left. \times \sum_r \rho(x-r, t) s(r) \tilde{V}'(r) \right\} \end{aligned} \quad (3.2)$$

with $\eta(x)$ as initial condition $\{(\nabla_1^\pm f(x) = \pm[f(x \pm 1) - f(x)])$ and $\Delta_1 = \nabla_1^+ \nabla_1^-\}$. The generator L_γ^β of the dynamics [see L_N defined in (2.6) of ref. 6 with $c(x, x+b; \eta)$ given by (1.1)] can be rewritten with little effort as

$$\begin{aligned} L_\gamma^\beta f(\eta) &= \frac{1}{2} \sum_x \sum_{b=\pm 1} \{ \eta(x) [1 - \eta(x+b)] \} \\ &\quad \times \exp \left\{ \frac{\beta}{2} \gamma^2 \sum_r \eta(x+br) s(r) \tilde{V}'(r) \right\} [f(\eta^{x, x+b}) - f(\eta)] \end{aligned} \quad (3.3a)$$

where

$$\tilde{V}'(r) = \begin{cases} \gamma^{-2} \nabla_1^+ V_\gamma(r')|_{r'=r-1} & \text{if } r > 1 \\ 0 & \text{if } r = 0, 1 \\ \gamma^{-2} \nabla_1^+ V_\gamma(r')|_{r'=-r} & \text{if } r < 0 \end{cases} \quad (3.3b)$$

and

$$s(r) = \begin{cases} -1 & \text{if } r \leq 0 \\ +1 & \text{if } r > 0 \end{cases}$$

Notice that in the limit $\varepsilon \rightarrow 0$ (hence $\gamma \rightarrow 0$) we have $\tilde{V}'(r) \rightarrow dV/dr'|_{r'=|r|}$.

If we choose a very small γ , we can expand L_γ^β in powers of γ up to the first order,

$$L_\gamma^\beta \approx L_0 + L_\gamma^{(1)} \tag{3.4a}$$

$$L_\gamma^{(1)} = \frac{1}{2} \sum_x \sum_{b=\pm 1} \eta(x) [1 - \eta(x+b)] \times \left\{ \frac{\beta}{2} \gamma^2 \sum_r V'(\gamma r) s(r) \eta(x+br) \right\} [f(\eta^{x,x+b}) - f(\eta)]$$

$$L_0 f(\eta) = \frac{1}{2} \sum_x [f(\eta^{x,x+1}) - f(\eta)] \tag{3.4b}$$

L_0 is the generator of the simple exclusion process.⁽⁹⁾ The case of a system with generator given by $L_0 + L_\gamma^{(1)}$ with the choice

$$V(r) = 2(r-1) \mathbf{1}_{\{r \leq 1\}} \tag{3.5}$$

is considered in ref. 6. We give the following:

Proposition. Given $\lambda > 2\alpha$, for all n there is c_n such that

$$\sup_{x \in \mathcal{M}_n} |v_n^\varepsilon(x, t | \eta)| \leq c_n t^{-n/8} \quad \forall \eta, \forall t \leq \varepsilon^{-2+\lambda} \tag{3.6}$$

The proof of this proposition is one of the central points in the proofs of both Theorems 2.1 and 2.2. From now on we will omit the dependence of the v -functions on η . We fix n and start from the formula

$$\begin{aligned} \frac{d}{dt} v_n(x, t) &= \mathbf{E}_\eta^\varepsilon \left\{ L_\gamma^\beta \prod_{i=1}^n [\eta(x_i, t) - \rho(x_i, t)] \right\} \\ &\quad - \sum_{i=1}^n v_{n-1}(x(i), t) \frac{\partial \rho}{\partial t}(x_i, t) \end{aligned} \tag{3.7}$$

$[x(i) \equiv (x_1, \dots, x_{i-1}, x_{i+1}, \dots, x_n) \in \mathcal{M}_{n-1}]$. If we define

$$\begin{aligned} L_\gamma^{(m^*)} f(\eta) &\equiv \frac{1}{2} \sum_x \sum_{b=\pm 1} \eta(x) [1 - \eta(x+b)] \\ &\quad \times \left\{ \sum_{n=2}^{m^*} \frac{1}{n!} \left(\frac{\beta \gamma^2}{2} \right)^n \left[\sum_r \eta(x+br) s(r) \tilde{V}'(r) \right]^n \right\} \\ &\quad \times [f(\eta^{x,x+b}) - f(\eta)] \end{aligned} \tag{3.8}$$

$[m^*$ will be chosen in (3.14)] and

$$R_\gamma^{(m^*)} = L_\gamma^\beta - L_0 - L_\gamma^{(1)} - L_\gamma^{(m^*)} \tag{3.9}$$

Using (3.7), we can write

$$\begin{aligned} \frac{d}{dt} v_n(\underline{x}, t) = & L_0 v_n(\underline{x}, t) + \Psi_1^{(n)}(\underline{x}, t) + \Psi_{m^*}^{(n)}(\underline{x}, t) + \mathbf{E}_\eta^\varepsilon \\ & \times \left\{ R_\gamma^{(m^*)} \prod_{i=1}^n [\eta(x_i, t) - \rho(x_i, t)] \right\} \end{aligned} \tag{3.10}$$

in which L_0 acts on \underline{x} and the Ψ 's are functions of v_m , with m varying in a subset that will be specified below. The first two terms in the right-hand side of (3.10) derive from the action of L_0 and $L_\gamma^{(1)}$ and from the last term in (3.7). As we have seen, they are obtained by expanding to first order the exponential in (3.3a) and they are the only terms present in the case of ref. 6. Furthermore,

$$\Psi_{m^*}^{(n)}(\underline{x}, t) = \mathbf{E}_\eta^\varepsilon \left\{ L_\gamma^{(m^*)} \prod_{i=1}^n [\eta(x_i, t) - \rho(x_i, t)] \right\} \tag{3.11}$$

We have

$$\left| \mathbf{E}_\eta^\varepsilon \left\{ R_\gamma^{(m^*)} \prod_{i=1}^n [\eta(x_i, t) - \rho(x_i, t)] \right\} \right| \leq c\gamma^{m^*+1} \tag{3.12}$$

By integrating (3.10), we have

$$|v_n(\underline{x}, t)| \leq \left| \int_0^t \mathbf{E}_x(\Psi_1^{(n)}(\underline{x}(t-s), s) + \Psi_{m^*}^{(n)}(\underline{x}(t-s), s)) ds \right| + c\gamma^{m^*+1}t \tag{3.13}$$

in which \mathbf{E}_x is the expectation taken over the simple exclusion process (for labeled particles, ref. 2). Because $t \leq \varepsilon^{-2+\lambda}$, by choosing

$$m^* > \left(\frac{n}{4} + 2\right) \left(\frac{1-\lambda/2}{1-\alpha}\right) - 1 \tag{3.14}$$

we have that

$$\gamma^{m^*+1}t \leq t^{-n/8} \tag{3.15}$$

and so we have reduced our task to the estimate of the first term on the right-hand side of (3.13). In order to obtain the bound (3.6), we iterate (3.13), by replacing the v_n -functions (contained in the Ψ 's) by the same

integral expression (3.13), m^* kept fixed during the iteration. After M iterations we obtain a sum of M -fold integrals of all the branches of the iteration.

Now we observe that $\Psi_1^{(n')}$ and $\Psi_{m^*}^{(n')}$ are functions of v_m respectively with $m \in \{\max(0, n' - 3), \dots, n' + 2\}$ and $m \in \{\max(0, n' - m^* - 2), \dots, n' + m^* + 1\}$: owing to the presence in the Ψ 's of $v_{n'}$ -functions with both $m \geq n'$ (birth terms) and $m < n'$ (death terms), after M iterations, while some branches have reached $v_0 \equiv 1$, some other branches still have correlation functions with strictly positive order. As in refs. 3 and 6, we need to choose M properly in order to obtain (3.6).

The idea of the proof is to show that the terms that we find are either those found in ref. 6 or smaller than those. As already mentioned, $\Psi_1^{(n')}$ collects all the terms present in ref. 6. The terms $\Psi_1^{(n')}$ can be written as the sum of two terms; one is just due to the simple exclusion and it survives even when there is no interaction ($V = 0$); the other one is the expectation of $A^{(n')}$, when $A^{(n')}$ is a linear combination of products of factors of the form $\tilde{\eta}(x_i, t) = \eta(x_i, t) - \rho(x_i, t)$. There are two important features in this structure; one is the presence of characteristic functions stating that some of the sites x_i and x_j appearing in the products are nearest neighbor sites; the other is that some of the terms have the structure of discrete derivatives. The extra terms have the structure

$$\left[\gamma^2 \sum_r \eta(x+r) s(r) \tilde{V}'(r) \right]^q A^{(n')} \tag{3.16}$$

where $1 \leq q \leq m^* - 1$ and

$$A^{(n')} = A^{(n')} + \sum_i \left[\frac{\partial \rho}{\partial t}(x_i, t) - \frac{1}{2} A_1 \rho(x_i, t) \right] \times \left\{ \prod_{j \neq i} [\eta(x_j, t) - \rho(x_j, t)] \right\} \tag{3.17}$$

We can write in (3.13) $\eta(y, t) = \tilde{\eta}(y, t) + \rho(y, t)$ and expand the q th power of the square bracket. The generic term in this expansion is

$$\gamma^{2q} \sum_{r_1, \dots, r_q} \tilde{\eta}(x+r_1, t) \cdots \tilde{\eta}(x+r_q, t) \rho(x+r_{q+1}, t) \cdots \rho(x+r_q, t) \times s(r_1) \tilde{V}'(r_1) \cdots s(r_q) \tilde{V}'(r_q) \tag{3.18}$$

When the arguments of the $\tilde{\eta}$'s are all distinct and different from those in $A^{(n')}$, then they produce an increase of the order of the correlation function; since there are essentially γ^{-1} terms in each sum, overall (3.18) produces

a factor γ^q and increases the order of the v -function. If there are coincidences, the order of the v -function is smaller than the previous one, but each unit it decreases, there is an extra γ factor, because the value of the corresponding site $x + r_i$ is constrained to be one of the other $x + r_j$, or one of those in $A^{(n')}$ (since the order of the v -function is finite, there are finitely many such choices). Therefore the contribution to (3.16) coming from the terms $A^{(n')}$ in $A^{(n')}$ is reduced to terms smaller than those present in the case of ref. 6 and with v -functions of not smaller degree.

The problem comes from the second term in (3.17), which was not present in ref. 6 and where one of the $\tilde{\eta}$ factors is missing. The terms (3.18) might not replace it, because there is a term in (3.18) with only ρ 's. But $q \geq 1$ and for $q=1$ this term is of the order of γ : by (3.2), $|\partial\rho/\partial t - (1/2) \cdot \Delta_1 \rho| = O(\gamma)$, so that we have a term γ^2 associated to the decrease by one unit of the order of the v -function. The time integral is bounded by $ct^2 < c\gamma^{-2}$ (because $\lambda > 2\alpha$), so that overall the loss of one unit in the order of the v -function does not contribute a converging factor up to the estimate (3.6). However, by a more careful analysis of (3.2), one can see that $|\partial\rho/\partial t - (1/2) \cdot \Delta_1 \rho| = O(\gamma)(1/\sqrt{t})$, so that the time integral is bounded by $c\sqrt{t} < c\gamma^{-1}$ and we indeed gain a factor $\gamma < t^{-1/8}$ which compensates the loss in the order of the v -function. Also for the other terms, similar estimates work and in this way we reduce the proof to that in ref. 6 and in turn to that in ref. 3.

Now observe that we prove (3.6) with the time derivative of ρ given by Eq. (3.2), i.e., for the first-order expansion in γ of the equation that we obtain assuming propagation of chaos: once we prove (3.6), the only difference between this case and that of ref. 6 is the generality in the choice of the potential V . So the other steps of the proof are the generalization of Sections 5–7 of ref. 6. In particular, we must repeat the parametrix method with Eq. (3.2). There is nothing drastically new, so we omit the details, for which we refer to ref. 5.

4. COMPUTER SIMULATIONS AND PHASE SEPARATION

For computer simulations it is more convenient to study another model whose relation with that introduced in Section 2 is discussed in ref. 6. This model assigns to every particle a velocity (± 1). The particle configurations are denoted by ξ , $\xi = \{\xi(x, \sigma): \xi(x, \sigma) \in \{0, 1\}, x \in \Gamma, \sigma(x) = \pm 1\}$. To compare the results in this model to that considered in the previous sections, we define the occupation number at x as $\xi(x) = [\xi(x, +1) + \xi(x, -1)]/2$. In every site of Γ (an interval of \mathbf{Z} with periodic boundary conditions) there are at most two particles (with different velocities). The dynamics is defined in two steps:

(1) *Velocity flips.* If there are two or no particles in the site x , then nothing happens. If there is one particle, its velocity σ' takes the values $+1$ or -1 with probability p and $1 - p$ (resp.) with p given by

$$p = \frac{1}{2} \left\{ 1 + \beta \gamma^2 \sum_{r=1}^{\lfloor 1/\gamma \rfloor} (\xi(x+r, t) - \xi(x-r, t)) \right\} \tag{4.1}$$

(2) *Advection.* Every particle at site x moves to $x + \sigma'(x)$.

From now on we choose γ such that γ^{-1} is an integer, $\beta > 2$, and initial density profiles have values in the interval $[I_\beta^-, I_\beta^+]$, cases which are not covered by Theorems 2.1 and 2.2.

To explain the computer simulations, we introduce the empirical densities

$$\bar{\rho}_l(x, t) = \frac{1}{2l+1} \sum_{i=-l}^{i=+l} \xi(x+i, t) \tag{4.2}$$

Typical lengths in the system are the lattice spacing (equal to 1) γ^{-1} , the interaction length, and ε^{-1} , the hydrodynamic spacing. We shall consider values of l either in the range $1 \ll l \ll \gamma^{-1}$ or $\gamma^{-1} \ll l \ll \varepsilon^{-1}$ and we shall see that some of the results change dramatically from one scale to the other.

On the other hand, if we reach a stationary or an almost stationary situation, we can observe the system by doing time averages

$$\bar{\rho}_L^T(x, t) = \frac{1}{T+1} \sum_{i=0}^T \xi(x, t+i) \tag{4.3}$$

at least for suitable T .

The purpose of our experiments is to investigate the following points:

- (1) Phase separation, growth of clusters, and their spatial patterns.
- (2) The values of the density of the clusters versus the temperature.
- (3) Hydrodynamic equations in the spinodal region.

(1) Computer simulations show (see, for instance, Fig. 2b or Fig. 6b) that the system separates into clusters with respectively high and low densities close to $1/2 \pm u^*$, where u^* is given by

$$\log \left(\frac{1 + 2u^*}{1 - 2u^*} \right) = 2\beta u^* \tag{4.4}$$

These values (we will call them, respectively, B and A) are exactly the densities of the two pure phases of the model in equilibrium at the critical

chemical potential. For further details we refer to Section 3 of ref. 6 and to Fig. 1. Recall that the hydrodynamics in the metastable region [$A \leq \rho \leq I_\beta^-$ ($=a$) and ($b=$) $I_\beta^+ \leq \rho \leq B$] is diffusive and that we are studying, by computer simulations, the unstable region of the model at equilibrium. We next examine the question of pattern selection when escaping from $\rho_0 \equiv 1/2$. We expect this solution to be unstable and, after a certain time, the system should glue together into clusters. There are some conjectures on this case. Assuming propagation of chaos, we see that the evolution of the average occupation numbers [call it $\rho(x, t)$] can be described by the equation

$$\frac{\partial \rho}{\partial t}(x, t) = \frac{1}{2} \mathcal{A}_1 \rho(x, t) - \beta \gamma^2 \nabla_1^- \left\{ \rho(x, t) [1 - \rho(x, t)] \sum_{r=1}^{1/\gamma} [\rho(x+r, t) - \rho(x+1-r, t)] \right\} \quad (4.5a)$$

In the limit $\gamma \rightarrow 0$ this equation is close to (3.2), with the choice (3.5). The eigenvalues of this equation, linearized around $\rho_0 = 1/2$, are

$$\lambda_\gamma(k) = -2 \sin^2\left(\frac{k}{2}\right) + \beta \gamma^2 \left[\sin^2\left(\frac{k}{2\gamma}\right) \right] \quad (4.5b)$$

First we consider the case $|I^-| = a\gamma^{-1}$ with $a > 1$ and such that $a\gamma^{-1}$ is an integer. In this case the wave numbers are

$$k_n = \frac{2\pi\gamma n}{a} \quad (n = 1, \dots, a\gamma^{-1}) \quad (4.6)$$

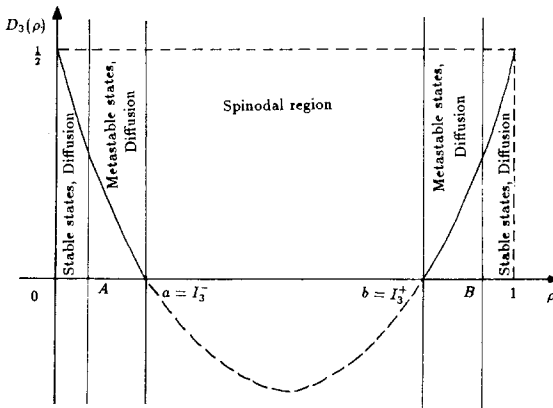


Fig. 1. Plot of the diffusion coefficient $D_\beta(\rho)$ with $\beta = 3 > \beta_c = 2$ ($I = 2$). The values a and b define the unstable (spinodal) region (see Section 3 of ref. 3) and they are equal, respectively, to I_3^- and I_3^+ . $A (= 1/2 - u^*)$ and $B (= 1/2 + u^*)$ are defined in (4.4) and I_β^\pm in (2.1e).

and we see that $\rho_0 \equiv 1/2$ is not linearly unstable for all values of $\beta > \beta_c = 2$. This is due to the finite size of the space (in the scale γ^{-1}): if we let $a \rightarrow \infty$, $\rho_0 \equiv 1/2$ is linearly unstable for all $\beta > \beta_c$. With the choice of $a = 4$ (and in the limit $\gamma \rightarrow \infty$) we have that for $\beta < \pi^2/4$ there is no positive eigenvalue. For $\pi^2/4 \leq \beta \leq \pi^2/2$ there is one positive eigenvalue and for $\beta > \pi^2/2$ there are two positive eigenvalues. On the other hand, for $\pi^2/4 \leq \beta < 3\pi^2/4$ the greatest eigenvalue is reached by the first harmonic [$n=1$ in (4.6)] and for $\beta > 3\pi^2/4$ is reached by the second harmonic ($n=2$). In this way we understand Figs. 2 and 3. We clearly see a sinusoidal shape, corresponding to the greatest eigenvalue, growing and then getting distorted when nonlinear effects become sensible, which agrees with the typical patterns of phase separation.⁽⁴⁾ Some theoretical considerations suggest that the time scale of escaping should be $\gamma^{-2} \log \gamma^{-1}$ and the simulations agree with this conjecture.

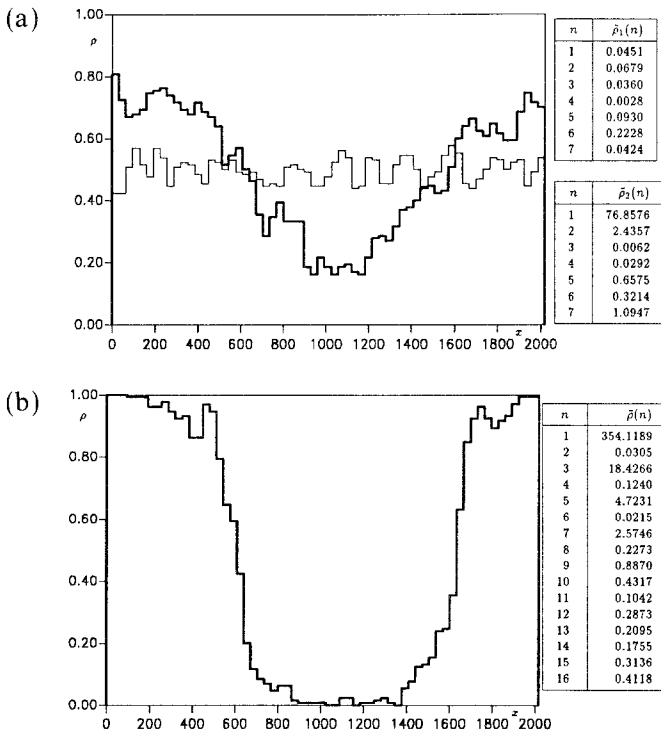


Fig. 2. (a) $|\Gamma| = 64 \cdot 32$, $\gamma^{-1} = 16 \cdot 32$ (so $|\Gamma| = 4\gamma^{-1}$), $\beta = 5$. We give the initial condition (graph 1: thin line) and the situation after $6 \cdot 10^5$ iterations (graph 2: thick line). The average is taken over a length $l = 32$. In the boxes we give also the Fourier transforms, $\hat{\rho}(n) = 4000 |\sum_{x=1}^{|\Gamma|} \rho(x) \exp(k_n x)|^2$, where k_n is given by (4.6) (b) As in part (a), but after $2 \cdot 10^6$ iterations.

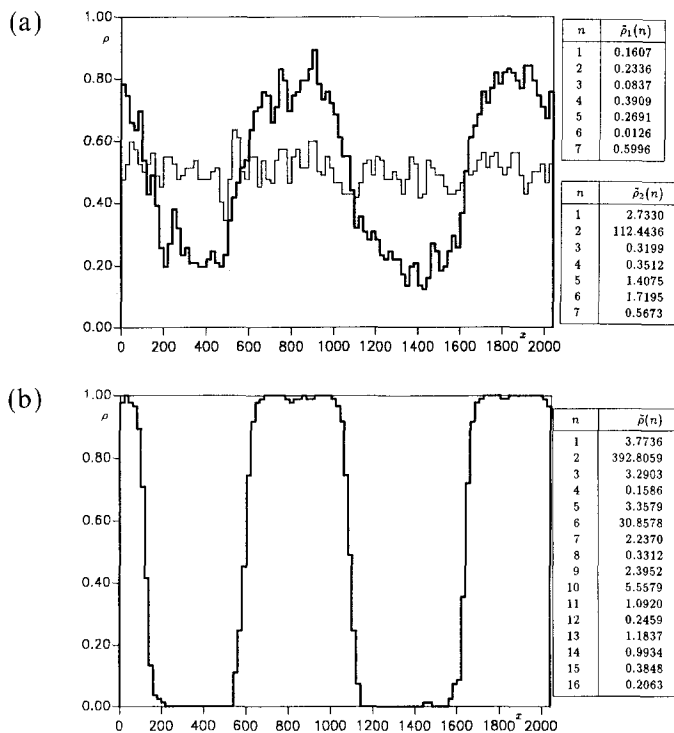


Fig. 3. (a) $|\Gamma| = 64 \cdot 32$, $\gamma^{-1} = 16 \cdot 32$ (so $|\Gamma| = 4\gamma^{-1}$), $\beta = 20$. We give the initial condition (graph 1: thin line) and the situation after $6 \cdot 10^5$ iterations (graph 2: thick line). The average is taken over a length $l = 20$. (b) As in part (a), but after $5 \cdot 10^6$ iterations.

On the other hand, if we consider the case of $|\Gamma| \gg \gamma^{-1}$ (as in the hydrodynamic regime) we easily see from (4.5b) that the greatest eigenvalue, for the values of β that are considered, corresponds to $k \cong \pi\gamma$. Since the escape is dominated by the wave number $\pi\gamma$, we expect again that the spatial scale of variation of the density after escape is γ^{-1} . In this case the spatial pattern seems to have a more complex evolution. The case with $\beta = 2.4$ and $|\Gamma| = 32\gamma^{-1}$ is listed in Fig. 5. In this particular case we see from (4.5) that the greatest eigenvalue has $n = 5$ and we see that the pattern of escape has a spatial structure with a periodicity given essentially by $n = 5$ (Fig. 5a). On longer times (Fig. 5b) some clusters have joined together and the new wave numbers of the system are around $n = 3$, i.e., they moved toward smaller k . A similar effect it is clear also in the case with $\beta = 32$ in Fig. 4. This seems to agree with the belief that actually the periodic stationary solutions are all unstable, except that with the lowest wave number.⁽⁴⁾

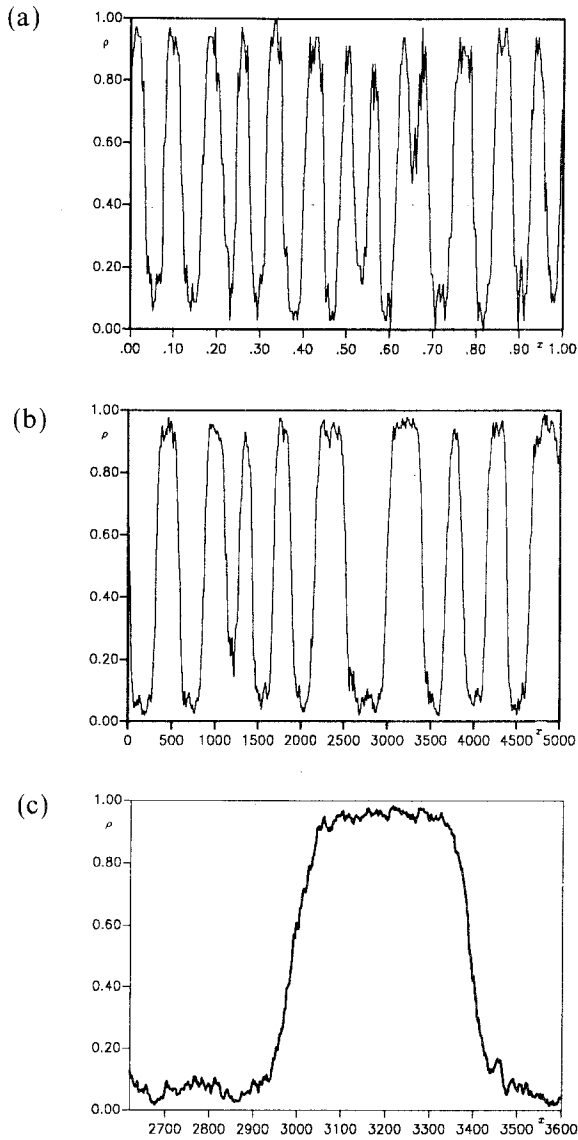


Fig. 4. (a) $|I| = 5000$, $\gamma^{-1} = 80$, $\beta = 3.2$. The initial condition (not given) is $\rho_0 \equiv 1/2$. The density is taken after $1.5 \cdot 10^5$ iterations. The average length is $l = 20$. Here $B = 0.95$ ($A = 0.05$), from (4.4), and $I_{\beta}^+ = 0.81$ ($I_{\beta}^- = 0.19$), from (2.1d). (b) As in part (a), but after $5 \cdot 10^6$ iterations. Time averaged with $T = 1000$ [see (4.3)]. (c) A particular case of part (b).

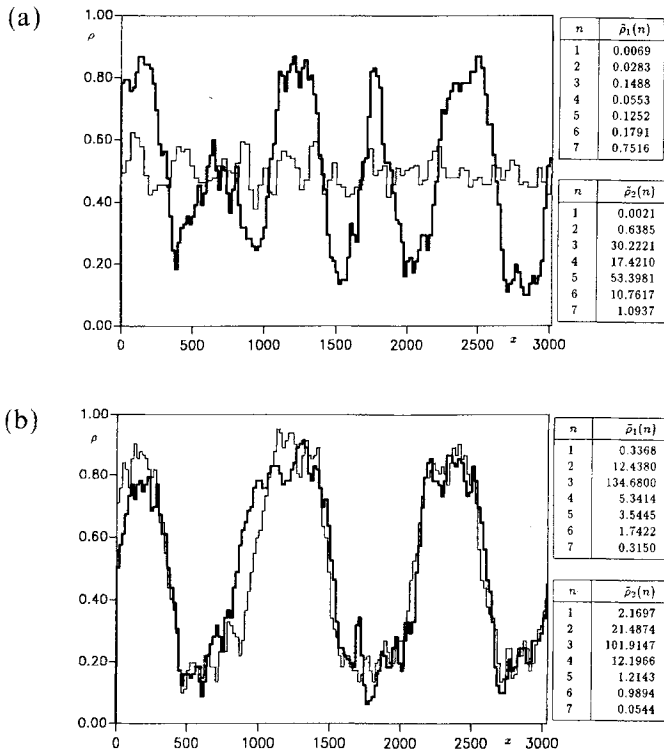


Fig. 5. (a) $|\Gamma| = 96 \cdot 32$, $\gamma^{-1} = 3 \cdot 32$, $\beta = 2.4$. We give the initial condition (graph 1: thin line) and the situation after $7 \cdot 10^5$ iterations (graph 2: thick line). The average is taken over a length $l = 20$. Here $B = 0.83$ ($A = 0.17$) and $I_\beta^+ = 0.70$ ($I_\beta^- = 0.30$). (b) As in part (a), but after $2 \cdot 10^6$ (graph 1: thin line) and $7 \cdot 10^6$ (graph 2: thick line).

(2) For values of $\beta \geq 5$, the interval of unstable densities is almost the whole interval $[0, 1]$, so that the densities in the clusters are forced to be very close to 0 and 1. For such a reason these experiments are not really decisive for concluding that the densities in the clusters are the same as those of the two pure phases. Figure 4 shows the case with $\beta = 3.2$: in this case the densities look the same as those of the pure phases. For $\beta = 2.4$ (see Fig. 5) the question is less clear. To have a better understanding of this problem we consider the equation derived from studying the system in the case $\alpha = 0$ (remember that $\gamma = e^{1-\alpha}$).

It can be shown (see section 3 of ref. 6) that, in this case, the behavior of the system is described, in the same sense as Theorems 2.1 and 2.2, by the integrodifferential equation (see Section 3 of ref. 6)

$$\frac{\partial \rho}{\partial t}(r, t) = \frac{\partial}{\partial r} \left\{ \frac{1}{2} \frac{\partial \rho}{\partial r}(r, t) - \beta \rho(r, t) [1 - \rho(r, t)] \right. \\ \left. \times \int_{-1}^{+1} s(r') \rho(r + r', t) dr' \right\} \quad (4.7)$$

[We restricted ourselves to the case of (3.5), but this result is true for arbitrary choices of the potential, with obvious modifications in (4.7).]

Dal Passo and De Mottoni⁽¹⁾ looked for stationary solutions of (4.7) in the set of monotonic functions such that

$$-\rho(-r) + \frac{1}{2} = -\frac{1}{2} + \rho(r) \quad (4.8)$$

and such that $\rho(r) - 1/2 > 0$ if $r > 0$. They proved existence and uniqueness of this solution if and only if Eq. (4.4) has a strictly positive solution. Furthermore, they proved that [call $\rho(r)$ the stationary solution]

$$\lim_{r \rightarrow \pm\infty} \rho(r) = \frac{1}{2} \pm u^* \quad (4.9)$$

[u^* given by (4.4)]. Nothing is known on the stability of this solution, but it should describe the temporal asymptotic behavior of the particle system (living in a space much greater than γ^{-1} : for example, γ^{-2}) starting from an initial condition given, for instance, by the step function that assumes values B for $r \geq 0$ and A for $r < 0$. It may be, however, that this solution does not describe the interface between the two phases in the case shown in this paper: the interfaces are too close and they can interact with each other.

To understand the origin of (4.9), we argue as follows. A stationary solution of (4.7) satisfies the equation

$$\frac{1}{2} \frac{\partial \rho}{\partial r} - \beta \rho(1 - \rho) \int_{-1}^{+1} s(r') \rho(r + r') dr' = C \quad (4.10a)$$

Owing to the symmetry $\rho(r) = 1 - \rho(-r)$

$$C = 0 \quad (4.10b)$$

By integration, (4.10) becomes

$$\int_0^R \frac{1}{2\beta\rho(1-\rho)} \frac{\partial \rho}{\partial r} dr = \int_0^R \int_{-1}^{+1} s(r') \rho(r + r') dr' \quad (4.11)$$

($R > 0$). By taking into account some cancellations on the right-hand side of (4.10a), we have

$$\begin{aligned} \frac{1}{2\beta} \log \left(\frac{\rho(R)}{1-\rho(R)} \right) = & - \int_0^{\min(1,R)} \int_{-1}^0 \rho(r+r') dr' dr \\ & + \int_{\max(R-1,0)}^R \int_0^1 \rho(r+r') dr' dr \end{aligned} \quad (4.12)$$

If we let $R \rightarrow +\infty$, we get the condition

$$\log \left(\frac{\rho(+\infty)}{1-\rho(+\infty)} \right) = 2\beta(\rho(+\infty) - 1/2)$$

which is (4.4). This result is easily extended to the case of a generic $V(r)$.

We can try to repeat the argument for the periodic stationary solutions of (4.7). Call L the period of a solution satisfying (4.8) and such that $\max_r \rho(r) = \rho(L/4) = 1/2 + u$. If there exists a solution in this class, then

$$\log \left(\frac{1+2u}{1-2u} \right) \leq 2\beta u \min((L/4), 1)$$

where the inequality is strict if we assume that the maximum is reached at an isolated point. Therefore we expect that if the system is close to one of these periodic solutions, then the densities of the clusters are, respectively, larger than A and smaller than B . The computer simulations do not give quantitative evidence on such questions.

(3) We finally consider the problem that remained unsolved in the first three sections (and in ref. 6): the hydrodynamics in the unstable region. We refer to Fig. 6. We consider a nonconstant initial profile, varying on the scale ε^{-1} (some 10^3), and with values in the spinodal region. We see that the system breaks up into clusters (Fig. 6b) of dimension $O(\gamma^{-1})$. To see this, we must take time averages [see (4.3)] or choose $l \ll \gamma^{-1}$ in (4.2). This linear dimension, in the hydrodynamic space scaling ($\gamma = \varepsilon^{1-\alpha}$), is infinitesimal because the hydrodynamic regime is determined by the space scale ε^{-1} . If we average over a distance much greater than the dimension of the clusters [in (4.2) take $l = \varepsilon^{-1+\alpha'}$ and $\alpha' > \alpha$, so $\gamma^{-1} \ll l \ll \varepsilon^{-1}$], we obtain a regular shape, reproducing identically the initial profile. As mentioned in ref. 6, results obtained for the Ginzburg–Landau model⁽¹¹⁾ and the Green–Kubo theory⁽¹⁰⁾ indicate that the diffusion coefficient should vanish in the phase transition region. This agrees with our experiments.

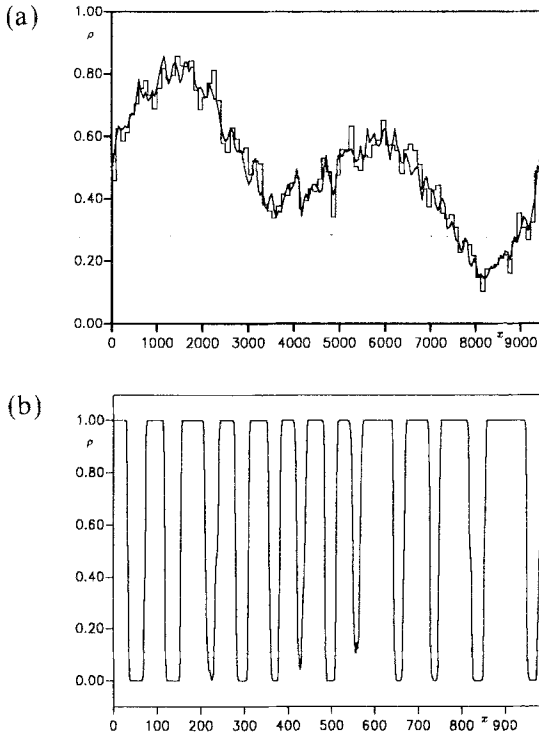


Fig. 6. (a) $|T| = 300 \cdot 32$, $\gamma^{-1} = 32$, $\beta = 20$. We give the initial condition (graph 1: histogram with average length $l = 50$) and the situation after $1 \cdot 10^6$ iterations (graph 2: continuous line with $l = 100$). (b) A particular of the situation in part (a) (graph 2) time averaged with $T = 100$ [see (4.3)].

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REFERENCES

1. R. Dal Passo and P. De Mottoni, Preprint, Rome (1991).
2. A. De Masi and E. Presutti, Lectures on the collective behavior of particle systems, CARR Reports in Mathematical Physics, no. 5/89 (November 1989); to be published as *Hydrodynamical Limits: Mathematical Methods* (Springer-Verlag, Berlin).

3. A. De Masi, E. Presutti, and E. Scaciatelli, The weakly asymmetric simple exclusion process, *Ann. Inst. H. Poincaré A* **25**:1–38 (1989).
4. P. C. Fife, Pattern dynamics for parabolic PDE's, Preprint; to appear in *Introduction to Dynamical Systems* (Proceedings Workshop at IMA) (Springer-Verlag, Berlin).
5. G. Giacomin, Dinamica stocastica di un gas di reticolo nel limite di campo medio locale, Tesi di laurea, Università di Pisa (1990).
6. J. L. Lebowitz, E. Orlandi, and E. Presutti, A particle model for spinodal decomposition, *J. Stat. Phys.* **63**:933–974 (1991).
7. J. L. Lebowitz and O. Penrose, Rigorous treatment of the van der Waals Maxwell theory of the liquid vapour transition, *J. Math. Phys.* **7**:98 (1966).
8. O. Penrose and J. L. Lebowitz, Rigorous treatment of metastable states in the van der Waals Maxwell theory, *J. Stat. Phys.* **3**:211–236 (1971).
9. T. M. Liggett, *Interacting Particle Systems* (Springer-Verlag, Berlin, 1985).
10. H. Spohn, *Large Scale Dynamics of Interacting Particles* (Springer-Verlag, Berlin, to appear).
11. F. Rezakhanlou, Hydrodynamic limit for a system with finite range interactions, *Commun. Math. Phys.* **129**:445–480 (1990).