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Phase Segregation Dynamics in Particle Systems with Long Range Interactions. I. Macroscopic Limits

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We present and discuss the derivation of a nonlinear nonlocal integrodifferential equation for the macroscopic time evolution of the conserved order parameter $\rho(r, t)$ of a binary alloy undergoing phase segregation. Our model is a d-dimensional lattice gas evolving via Kawasaki exchange dynamics, i.e., a (Poisson) nearest neighbor exchange process, reversible with respect to the Gibbs measure for a Hamiltonian which includes both short-range (local) and long-range (nonlocal) interactions. The nonlocal part is given by a pair potential $\gamma^d J(\gamma |x-y|)$, $\gamma > 0$, x and y in $\mathbb{Z}^d$, in the limit $\gamma \to 0$. The macroscopic evolution is observed on the spatial scale γ^{-1} and time scale γ^{-2}, i.e., the density $\rho(r, t)$ is the empirical average of the occupation numbers over a small macroscopic volume element centered at $r = \gamma x$. A rigorous derivation is presented in the case in which there is no local interaction. In a subsequent paper (Part II) we discuss the phase segregation phenomena in the model. In particular we argue that the phase boundary evolutions, arising as sharp interface limits of the family of equations derived in this paper, are the same as the ones obtained from the corresponding limits for the Cahn–Hilliard equation.

KEY WORDS: Interacting particle systems; Kac potential; hydrodynamic limit; phase segregation; spinodal decomposition.

1. INTRODUCTION

The process of phase segregation following a quench (sudden cooling) of a system from high temperature, where the system has a unique uniform

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equilibrium phase, into the miscibility gap where two (or more) phases can coexist is variously known as spinodal decomposition, nucleation, coarsening, etc.. It concerns the tendency of the different phases to segregate, creating larger and larger domains of approximately homogeneous single-phase regions. The problem is of great practical importance in the manufacturing of alloys, where the degree of segregation influences the properties of the material. The mathematical description of the time evolution of the local macroscopic order parameter in such systems, e.g., the difference in the concentration of A and B atoms in a binary A–B alloy, is commonly given by nonlinear fourth-order equations of the Cahn–Hilliard type.[4]

These equations appear to capture much of the phenomena. In particular, their numerical solutions show good agreement with experiments and with computer simulations of the Ising model (thought of as a binary alloy) evolving via Kawasaki exchange dynamics. This agreement relates both to the appearance and shape of segregated domains, which seem to exhibit a self-similar structure, and also to the quantitative behavior of the characteristic length describing these structures, which seems to grow like $t^{1/3}$, where t is the time. While this agreement is certainly satisfying, the original Cahn–Hilliard equation (CHE) or the modifications of it proposed so far[9] do not seem to arise as exact macroscopic description of microscopic models of interacting particles, such as the Ising model with Kawasaki dynamics (the CHE can, however, be derived from certain mesoscopic Ginzburg–Landau continuous-spin models[2]). This is unlike some other physically motivated equations, e.g., the *diffusion* equation and the *Boltzmann equation*, which can be derived from idealized microscopic models in suitable limits.[7,29] Such derivations are both of intrinsic interest and also indicate something about the range of applicability of the macroscopic equations. The latter might be particularly relevant for the CHE, where all that is known mathematically about the behavior of the solutions is restricted to the late stages of the coarsening process when the evolution is *assumed* to be dominated by the motion of sharp interfaces between well-formed domains of the pure phases. It is far from clear how much this singles out the CHE from other possible equations describing phase segregation.

In this paper we rigorously derive a macroscopic equation describing phase segregation in microscopic model systems with long-range interactions evolving according to stochastic Kawasaki dynamics with nearest neighbor exchanges. We will then, in Part II,[14] study the interface motion obtained from the derived macroscopic equation, in the *sharp interface limit*, by means of formal matched asymptotic expansions of the solution of the macroscopic evolution equation (see, e.g., refs. 3, 5, 18, and 24). By *sharp interface* we mean the limit in which the phase domains are very large compared to the size of the interfacial region, i.e., denoting by L the *typical* size of the domains, we look for results in the limit $L \to \infty$. The time will have to be properly scaled as well, typically as some integer power of L, according to the type of initial condition and the choice of the temperature. Our conclusion there is that, from the *sharp interface viewpoint*, the equation derived from a particle system and the Cahn–Hilliard equation are essentially equivalent.

The models we consider are dynamical versions of lattice gases interacting via long-range Kac potentials, also known as *local mean-field* interactions. The equilibrium properties of these systems are well known.[16,21,25] They provide microscopic models in which the van der Waals or mean-field description of phase transition phenomena, which is in good qualitative (or even quantitative) agreement with experiments away from the critical point, holds exactly. This includes metastability phenomena.[25] The corresponding dynamical models which we study here are sometimes called *local mean-field Kawasaki dynamics*. They can be described in words as follows: each particle hops (at a random time) from a site of the lattice $\mathbb{Z}^d$ to one of its $2d$ neighboring sites with a rate which depends on the particle configuration. These rates are chosen to satisfy detailed balance (reversibility) with respect to the Gibbs measure having the specified interaction between the particles.[29]

In the simplest model we consider here there is only a long-range (Kac-type) potential. We will also discuss, but not investigate in detail, the case in which additional short-range interactions are also present. Further work on the same model and related ones can be found in refs. 12, 20, and 32; these papers focus on the diffusive regime, i.e., on the region in which there is only one phase, but versions of the integral equation on which we are focusing are already present there. The case without a conservation law, *Glauber* dynamics, has received much attention (see ref. 6 and references therein).

The precise definition of the model is given in the next section. We show there that the evolution of the macroscopic density (in the simplest model) is given in terms of a second-order integrodifferential equation (2.16). We then show that this equation can be written in terms of the gradient flux associated with the classical local mean-field free-energy functional[21,25] and a density-dependent mobility. This allows us to make a direct connection between the properties of the solution of the derived evolution equation and the equilibrium phase diagram as well as with the solution of the CHE. In Section 3 we argue that the gradient structure for the macroscopic evolution law should hold generally in systems with long-range Kac potentials and arbitrary additional short-range interactions. In

Section 4 we make a remark on the case in which a weak external field is present. The proof for the simple case is given in Section 5.

2. THE PARTICLE MODEL AND ITS HYDRODYNAMIC LIMIT

The particles live on the d-dimensional lattice $\Lambda_\gamma = \{1, 2,..., [\gamma^{-1}]\}^d$, where $\gamma > 0$ is a small parameter and $[r]$ denotes the integer part of the real number r. We impose periodic boundary conditions on Λ_γ. Each site of Λ_γ is either occupied (1) or empty (0), hence a particle configuration is an element η of $\Omega_\gamma = \{0, 1\}^{\Lambda_\gamma}$ and the latter is endowed with the product topology. The dynamics is specified by giving an initial condition $\eta_0 \in \Omega_\gamma$, which may be random, i.e., a measure on the Borel sets of Ω_γ, and some (stochastic) evolution rules which will define η_t for any t positive. Our aim is to have a Markovian dynamics for which the Gibbs measure associated with a given Hamiltonian and a given total particle number is the unique reversible time-invariant measure at a fixed temperature. The Hamiltonian is a real-valued function defined on Ω_γ and we take it to be the sum of two terms

$$H = H_s + H_\gamma \tag{2.1}$$

with

$$H_s = -\tfrac{1}{2} \sum_{x, y \in \Lambda_\gamma} K(x-y)\, \eta(x)\, \eta(y) \tag{2.2}$$

$$H_\gamma = -\tfrac{1}{2} \sum_{x, y \in \Lambda_\gamma} \gamma^d J(\gamma(x-y))\, \eta(x)\, \eta(y) \tag{2.3}$$

in which J is a smooth function (C^∞) from the d-dimensional unit torus T^d to the real numbers such that $J(r) = J(-r)$, and $K(x) = 0$ if $|x| > R$ for some R independent of γ. The term in (2.3) will be called *nonlocal*, while the one in (2.2) will be called *local* or *short range*. The Gibbs measure with Hamiltonian H at the temperature $1/\beta$ ($\beta > 0$) and total number of particle $N \in \mathbb{Z}^+$ is defined as

$$\mu_\gamma^\beta(\eta) = \frac{\exp(-\beta H(\eta))}{Z_\gamma(N)} \tag{2.4}$$

where

$$Z_\gamma(N) = \sum_{\eta \in \Omega_\gamma^N} \exp(-\beta H(\eta)) \tag{2.5}$$

(2.4) is a probability measure over $\Omega_\gamma^N \equiv \{\eta \in \Omega_\gamma: \sum_{x \in \Lambda_\gamma} \eta(x) = N\}$. The stochastic process $\{\eta_t\}_{t \geqslant 0}$ is the Poisson jump process[22,29] generated by the operator

$$L_\gamma f(\eta) = \sum_{x,\, y \in \Lambda_\gamma} c_\gamma(x, y; \eta)[f(\eta^{x,y}) - f(\eta)] \tag{2.6}$$

where f is a real-valued (bounded) function on Ω_γ,

$$\eta^{x,y}(z) = \begin{cases} \eta(x) & \text{if} \quad z = y \\ \eta(y) & \text{if} \quad z = x \\ \eta(z) & \text{otherwise} \end{cases} \tag{2.7}$$

and

$$c_\gamma(x, y; \eta) = \Phi\{\beta[H(\eta^{x,y}) - H(\eta)]\} \tag{2.8}$$

if $|x - y| = 1$ and is zero otherwise. Here Φ: $\mathbb{R} \to \mathbb{R}^+$ is twice differentiable in a neighborhood of 0 and satisfies the detailed balance or reversibility condition,

$$\Phi(E) = \exp(-E)\, \Phi(-E) \tag{2.9}$$

for all $E \in \mathbb{R}$. We also assume $\Phi(0) = 1$: the case $\Phi(0) \in (0, \infty)$ can be recovered with a time change.

Loosely speaking, the process η_t can be described in the following way: if at time t the configuration is η_t, the probability that in the time interval $[t, t + \Delta t]$ the sites x, y ($|x - y| = 1$) exchange their occupation numbers is

$$c_\gamma(x, y; \eta_t)\, \Delta t + O((\Delta t)^2) \tag{2.10}$$

We note that if $\eta(x) = \eta(y)$, then an exchange between x and y does not modify the configuration η and it is thus possible to interpret the dynamics in terms of particles which attempt to jump from x to y, but the jump is performed only if the site y is empty: this type of dynamics is said to have an *exclusion* rule, that is, the particles have an on-site hard-core repulsion. A detailed construction of this process in terms of Poisson jump processes is given on p. 158 of ref. 29. The configuration space we are working on (Ω_γ) is finite and this avoids the difficulties connected to defining such a dynamics on an infinite state space (Chapter 1 of ref. 22). In Section 1.2 (Part II) of ref. 29 it is shown that for f and g, bounded functions on Ω_γ,

$$\int g(\eta)\, L_\gamma f(\eta)\, d\mu_\gamma^\beta(\eta) = \int f(\eta)\, L_\gamma\, g(\eta)\, d\mu_\gamma^\beta(\eta) \tag{2.11}$$

This property is called *reversibility* and it is a direct consequence of (2.9). In particular, (2.11) implies that

$$\int L_\gamma f(\eta)\, d\mu_\gamma^\beta(\eta) = 0 \tag{2.12}$$

We will use the following notation: the generic initial condition is a probability measure μ on Ω_γ. The law of the process $\{\eta_t\}_{t\geqslant 0}$ with initial condition μ will be denoted by P_γ^μ (E_γ^μ for the expectation). The process P_γ^μ is linked to the semigroup generated by L_γ via the formula $E_\gamma^\mu(f(\eta_t)) = \int (\exp(L_\gamma t) f)(\eta)\, \mu(d\eta)$. Equation (2.12) implies that for any $t \geqslant 0$ and any f bounded

$$\frac{d}{dt} E_\gamma^{\mu_\gamma^\beta}(f(\eta_t)) = 0 \tag{2.13}$$

which means that μ_γ^β is invariant under the process generated by L_γ.

The Hydrodynamic Limit

We are interested in initial states μ_γ such that, when $\gamma \to 0$, μ_γ *resembles* more and more a profile ρ_0, where ρ_0 is a measurable function from the d-dimensional unit terms T^d to $[0, 1]$, stretched by γ^{-1}. More precisely, we say that $\{\mu_\gamma\}_{\gamma>0}$ is an initial condition associated with ρ_0 if, for any continuous function ϕ from T^d to $\mathbb{R}$ and every $\delta > 0$,

$$\lim_{\gamma\to 0} \mu_\gamma \left(\left| \gamma^d \sum_{x\in T^d} \phi(\gamma x)\, \eta(x) - \int_{T^d} \phi(r)\, \rho_0(r)\, dr \right| > \delta \right) = 0 \tag{2.14}$$

The condition (2.14) is clearly satisfied if μ_γ is such that $\langle \eta(x) \rangle = \int_{\Omega_\gamma} \eta(x)\, d\mu_\gamma(\eta) = \rho_0(\gamma x)$ for all x in Λ_γ and the occupation numbers of the sites are independent. On the other hand, the initial condition concentrated on the chessboard configuration [$\eta(x) = 1$ if the sum of the components of x is even, $\eta(x) = 0$ otherwise] is also obviously associated with $\rho_0 \equiv 1/2$ and many other examples can be easily constructed. Loosely speaking, when we say that the particle system has a hydrodynamic limit, we mean that (2.14) holds also at later times if we replace ρ_0 by the solution of a suitable hydrodynamic evolution equation (in our case we will have an integrodifferential equation) with initial condition ρ_0. In Section 4 we prove the following theorem:

Theorem 1. The hydrodynamic limit without short-range interactions. Set $K(x) = 0$ so that the Hamiltonian coincides with H_γ. Let μ_γ be an initial condition associated with $\rho_0 \in C^2$. Then, for any t positive, $\delta > 0$, and any continuous function ϕ from T^d to $\mathbb{R}$

$$\lim_{\gamma \to 0} P_\gamma^{\mu_\gamma}\left(\left|\gamma^d \sum_{x \in T^d} \phi(\gamma x)\, \eta_{t\gamma^{-2}}(x) - \int_{T^d} \phi(r)\, \rho(r, t)\, dr\right| > \delta\right) = 0 \tag{2.15}$$

when $\rho(r, t)$, $r \in T^d$, and $t \in [0, \infty)$, is the unique classical solution of the equation

$$\begin{cases} \partial_t \rho(r, t) = \nabla \cdot [\nabla \rho(r, t) - \beta \rho(r, t)(1 - \rho(r, t)) \int_{T^d} \nabla J(r - r')\, \rho(r')\, dr'] \\ \rho(r, 0) = \rho_0(r) \end{cases} \tag{2.16}$$

It is now an observation, which at first sight appears surprising, that (2.16) can be rewritten in the form

$$\frac{\partial \rho(r, t)}{\partial t} = \nabla \cdot \left[\sigma^0(\rho)\left(\nabla \frac{\delta}{\delta \rho} \mathscr{F}^0\right)\right] \tag{2.17}$$

where

$$\mathscr{F}^0(\rho) = -\frac{1}{\beta} \int_{T^d} s(\rho(r))\, dr - \frac{1}{2} \iint_{T^d \times T^d} J(r - r')\, \rho(r)\, \rho(r')\, dr\, dr' \tag{2.18}$$

with

$$s(\rho) = -\rho \log \rho - (1 - \rho) \log(1 - \rho) \tag{2.19}$$

and

$$\sigma^0(\rho) \equiv \beta \rho (1 - \rho) \tag{2.20}$$

Rewriting (2.16) in the form (2.17), $\mathscr{F}^0$ is recognized as the free energy functional and $\sigma_0(\rho)$ as the mobility of our model without the long-range interactions. This allows us to connect our equation with that of the CHE. Before doing that we rewrite $\mathscr{F}^0(\rho)$, up to an irrelevant additive constant, in the form

$$\mathscr{F}^0(\rho) = \int_{T^d} f_c^0(\rho(r))\, dr + \tfrac{1}{4} \int_{T^d \times T^d} J(r - r')[\rho(r) - \rho(r')]^2\, dr\, dr' \tag{2.21}$$

where

$$f_c^0(\rho) = -\frac{\hat{J}(0)}{2}\left(\rho - \frac{1}{2}\right)^2 + \frac{1}{\beta}(\rho \log \rho + (1 - \rho) \log(1 - \rho)) \tag{2.22}$$

and $\hat{J}(0) = \int J(r)\, dr$.

Note that if ρ is constant, the second term in (2.21) vanishes, so that f_c^0 is the *constrained equilibrium* free energy density of a homogeneous system.[21,25] In fact $f_c^0(\rho)$ is the correct equilibrium free energy density as long as $\beta \leqslant \beta_c = 1/T_c = 4/\hat{J}(0)$. If $\beta > \beta_c$, $f_c^0(\rho)$ has a double minimum at $\rho = \rho_\beta^\pm$, the two nontrivial solutions of $\log(\rho/(1-\rho)) = \beta\hat{J}(0)(\rho - 1/2)$, and the correct free energy is then obtained by the double tangent construction. A heuristic derivation of (2.16) and an explanation of (2.17) is given in Section 3, where the generalization to the case $K \neq 0$ is considered. Equation (2.17) has the same structure as the CHE, whose various forms correspond to different $\sigma^0(\rho)$ and $f_c^0(\rho)$ chosen by different authors.[4,5,18,26] What is common to the different CHEs in the literature is that the second term on the right side of (2.21) is of the form $\frac{1}{2}\zeta \int (\nabla\rho)^2\, dr$, where $\zeta > 0$ is related to the surface tension. This can be thought of as expanding the term in (2.21) and keeping only some terms, which is reasonable when the scale on which ρ varies is large compared to γ^{-1}. We discuss the relationship between the solutions of (2.16) and the CHE in ref. 14; see also ref. 13.

3. THE GENERAL CASE: $K \neq 0$

We start by giving a heuristic explanation of the result in Theorem 1, which is in fact a sketch of its proof. We are for the moment still in the case $K = 0$: if $\beta = 0$, the particle system reduces to the symmetric *simple exclusion process* (SEP), i.e., all the particles are performing exchanges with rate one, so their only interaction is given by the exclusion rule. As it is straightforward to verify, the Bernoulli measures μ_ρ with uniform density $\rho \in [0, 1]$, under which the random variables $\{\eta(x)\}_{x \in \Lambda_\gamma}$ are independent and $\int \eta(0)\, d\mu_\rho(\eta) = \rho$, are invariant for the SEP dynamics and the hydrodynamic limit for the SEP is simply given by the heat equation (see, e.g., ref. 17), as in (2.16) with $\beta = 0$. Moreover, we observe that in the case $\beta \geqslant 0$,

$$\begin{aligned} H(\eta^{x,x+e}) - H(\eta) &= \gamma(\eta(x+e) - \eta(x)) \left[\gamma^d \sum_z \eta(z)(e \cdot \nabla J)(\gamma(x-z)) \right] + O(\gamma^2) \\ &= O(\gamma) \end{aligned} \tag{3.1}$$

for all $x \in \Lambda_\gamma$ and e a unit vector in $\mathbb{Z}^d$ [(3.1) is derived in Lemma 2, Section 5], so that

$$\begin{aligned} c_\gamma(x, x+e; \eta) &= 1 - \frac{\beta}{2} [H(\eta^{x,x+e}) - H(\eta)] + O(\gamma^2) \\ &= 1 + O(\gamma) \end{aligned} \tag{3.2}$$

where we used (3.1) and the fact that (2.9) and $\Phi(0) = 1$ imply $\Phi'(0) = -1/2$.

Formula (3.2) indicates clearly that the dynamics with $\beta > 0$ is a weak perturbation of the $\beta = 0$ dynamics. In particular, the dominant SEP dynamics will enforce at time $t\gamma^{-2}$ $(t > 0)$ *local equilibrium* with respect to its invariant measure, i.e., the state of the system will locally (on spatial scales shorter that γ^{-1}) be very close to the Bernoulli measure μ_ρ and ρ will vary on the macroscopic scale γ^{-1}. The perturbation term in (3.1) generates a force term given by the negative gradient of the energy density at r,

$$F(r) \equiv \nabla \left(\int J(r - r')\, \rho(r')\, dr' \right) \tag{3.3}$$

This gives an extra contribution to the macroscopic current equal to this force times the mobility $\sigma^0 = \beta\rho(1 - \rho)$. In this expression β measures the intensity of the bias in the exchange rates and $\rho(1 - \rho)$ gives the rate at which the exchanges actually take place when a particular site is chosen at random, since the system is locally described by the Bernoulli measure μ_ρ. This explains the form of (2.16).

That (2.16) can be transformed into (2.17) is clearly due to the fact that $s(\rho)$ in (2.19) satisfies the relation

$$-\frac{1}{\beta} s''(\rho)\, \sigma^0(\rho) = 1 \tag{3.4}$$

for all $\rho \in (0, 1)$. This is no accident, but, as will now show, (3.4) follows from the *Einstein relation* between fluxes and forces for this system.

This will become clearer if we consider the general case $K \neq 0$. We call the system with only the local Hamiltonian H_s the *reference* system. The equilibrium free energy $f^s_{\mathrm{eq}}(\rho)$ associated with this reference system at an average density $\rho \in [0, 1]$, which depends also on β, is uniquely defined in *the thermodynamic limit.*[27,28] We shall further assume that $f^s_{\mathrm{eq}}(\rho)$ is strictly convex and real analytic, which implies that there is no phase transition for the equilibrium reference system associated with H_s at the temperature $1/\beta$. Moreover, we are assuming that for each $\rho \in [0, 1]$ there exists a unique, translation-invariant and ergodic, infinite-volume limit Gibbs state μ^s_ρ such that $\int \eta(0)\, d\mu^s_\rho(\eta) = \rho$. All these properties, which are to a certain extent equivalent, are known to hold if β is sufficiently small.[27,28]

We now claim that the correct macroscopic evolution law for the case $K \neq 0$ should be

$$\frac{\partial \rho}{\partial t} = \nabla \cdot \left[\sigma_s \nabla \left(\frac{\delta \mathscr{F}}{\delta \rho} \right) \right] \tag{3.5}$$

where $\sigma_s(\rho)$ is the mobility of the reference system,

$$\mathscr{F}(\rho)=\int f_c(\rho(r))\,dr+\tfrac{1}{4}\iint J(r-r')[\rho(r)-\rho(r')]^2\,dr\,dr' \tag{3.6}$$

and we have defined the constrained equilibrium free energy density (in analogy with ref. 21) as

$$f_c(\rho)=-\frac{\hat{J}(0)}{2}\left(\rho-\frac{1}{2}\right)^2+f^s_{\mathrm{eq}}(\rho) \tag{3.7}$$

which is clearly a straightforward generalization of (2.17).

The definition of σ_s for a system with short-range interactions in the one-phase system can be found in Part II of ref. 29, formulas (2.27), (2.71), and (2.72) [in our notation formulas (2.27) and (2.71) must be multiplied by β]. It is given by a Green–Kubo formula and is related to the diffusion coefficient of the reference system $D_s(\rho)$ by the relation

$$\sigma_s=\chi_s D_s \tag{3.8}$$

where $\chi_s(\rho)$ is the inverse of the derivative of the chemical potential, i.e., the compressibility of our reference system is defined as

$$\chi_s\equiv\frac{1}{f^{s\prime\prime}_{\mathrm{eq}}(\rho)}=\beta\sum_{x\in\mathbb{Z}^d}\left(\int\eta(x)\,\eta(0)\,d\mu^s_\rho(\eta)-\rho^2\right) \tag{3.9}$$

While a complete proof of (3.7) for general short-range interactions is lacking, some particular cases can be handled.[1] In order to justify (3.5) on a heuristic level we will draw an analogy with Section 3.4 of Part II of ref. 29. There a linear response argument is developed for the system with Hamiltonian

$$H_V(\eta)=H_s(\eta)+\sum_{x\in\Lambda_\gamma}V(\varepsilon x)\,\eta(x) \tag{3.10}$$

where $V(r)$ is a smooth function from T^d to $\mathbb{R}$ and $\varepsilon>0$ is a small parameter. This is the Hamiltonian of a weakly driven lattice gas: the dynamics is defined by the Poisson rates $c_{\varepsilon,V}(x,y;\eta)=\Phi(\beta\Delta_{x,y}H_V(\eta))$ for $|x-y|=1$ [and $c_{\varepsilon,V}(x,y;\eta)=0$ otherwise]. Hence we have

$$\begin{aligned}c_{\varepsilon,V}&(x,x+e;\eta)\\&=\Phi(\beta\Delta_{x,x+e}H_s(\eta))-\beta\varepsilon\Phi'(\beta\Delta_{x,x+e}H_s(\eta))\\&\quad\times(\eta(x+e)-\eta(x))(e\cdot\nabla V)(\varepsilon x)+O(\varepsilon^2)\end{aligned} \tag{3.11}$$

In the hydrodynamic scaling limit, with r scaled as εx, $\varepsilon \to 0$, external force will vanish like ε. Hence it can be argued (see the macroscopic continuity equation for the density $\partial\rho/\partial t = -\nabla \cdot J$), that the current J should be the sum of a diffusive term, unaffected by the weak external force, and a drift term proportional to $F = -\nabla V(r)$,

$$J = -D_s(\rho)\,\nabla\rho + \sigma(\rho)\,F \tag{3.12}$$

with $\sigma(\rho)$ a mobility to be determined. To obtain σ we observe that the Gibbs measure μ_V^β associated with $H_V(\eta)$ at the temperature $1/\beta$ is a stationary state of our system. This implies that $\partial\rho(r,t)/\partial t = 0$ whenever $\rho(r,0) = \rho_{\text{eq}}(r)$ is the density obtained from μ_V^β. In the limit $\varepsilon \to 0$, ρ_{eq} is the density which minimizes the functional[21]

$$\mathscr{F}_V(\rho) = \int \left[f_{\text{eq}}^s(\rho(r)) + V(r)\,\rho(r) \right] dr \tag{3.13}$$

with a total mass constraint. In particular, at the minimum, $\delta\mathscr{F}_V/\delta\rho =$ const. But, according to (3.12), $\rho(r,t)$ satisfies the equation

$$\frac{\partial\rho}{\partial t} = \nabla \cdot \left[D_s(\rho)\,\nabla\rho + \sigma(\rho)\,\nabla V \right] \tag{3.14}$$

so the requirement that $\partial\rho/\partial t = 0$ at $\rho(r) = \rho_{\text{eq}}(r)$ then requires that $\sigma = D_s\chi$. This gives the described Einstein relation, showing that $\sigma = \sigma_s$.

In the model we are considering $\varepsilon = \gamma$ and the external potential V is replaced by an internally generated one, $-\gamma^d \sum_{y \in \Lambda_\gamma} J(\gamma(x-y))\,\eta(y)$. Taking now the hydrodynamic scaling limit, with the scaling parameter ε set equal to γ and letting $\gamma \to 0$, the previous analyses should remain valid with $V(r) \to -\int J(r-r')\,\rho(r')\,dr'$ in (3.14), which then gives (3.5).

We note that if we look at a hydrodynamic space scale ε^{-1} which is much larger than γ^{-1}, say $\varepsilon = \gamma^\delta$, $\delta > 1$, then we get a purely diffusive equation for $\rho(r,t)$. This is obtained from (3.5) by dropping the second term in (3.6). This equation holds rigorously *outside* the spinodal region, where the new diffusion constant becomes negative.[20,12]

4. A REMARK ON THE EFFECT OF AN EXTERNAL DRIVING FORCE

As one may expect, domain-coarsening phenomena become richer and more complicated in the presence of an external driving force, and some important points are still controversial (see, e.g., refs. 19 and 33). One of the interesting problems is understanding the scaling behavior of the cluster size; different behaviors are expected depending on whether the characteristic length is measured in the direction perpendicular or parallel to the field.

Here we simply observe that it is straightforward to extend the results of this paper to the case of a system on which a weak external force is acting. This system has a Hamiltonian

$$H(\eta)+\sum_{x\in\Lambda_\gamma} V(\gamma x)\,\eta(x) \tag{4.1}$$

where V is a smooth function from T^d to $\mathbb{R}$, as in Section 3. Remember that $H=H_s+H_\gamma$ [formula (2.2)]. The discussion in Section 3 easily gives the following guess for the hydrodynamic limit of this system:

$$\frac{\partial\rho}{\partial t}=\nabla\cdot\left[\sigma\nabla\left(\frac{\delta\mathscr{F}}{\delta\rho}+V\right)\right] \tag{4.2}$$

Alternatively, one can redefine $\mathscr{F}(\rho)$ as $\mathscr{F}(\rho)-\int V(r)\,\rho(r)\,dr$ and the evolution equation would still be (3.5). We observe that the proofs in Section 4 directly extend to this case if $K=0$.

The situation is similar in the case of an external force which is not the gradient of a potential V, e.g., a constant force $E\in\mathbb{R}^d$. In this case the rates of the process would be

$$\Phi(\beta(H(\eta^{x,y})-H(\eta)-\gamma E\cdot(x-y)(\eta(x)-\eta(y))) \tag{4.3}$$

if $|x-y|=1$ and zero otherwise and the macroscopic equation is

$$\frac{\partial\rho}{\partial t}=\nabla\cdot\left[\sigma\nabla\left(\frac{\delta\mathscr{F}}{\delta\rho}\right)\right]+\nabla\cdot(E\sigma) \tag{4.4}$$

The term added to (3.5) to obtain in (4.4) is exactly the term which in ref. 33 is added to the CH equation to adapt it to a situation in which an external field is present. Problems and limitations to the use of macroscopic models like (4.4) are discussed in ref. 19: (4.4) in fact does not seem to be suitable to describe systems with strong [$O(1)$ and not, like here, $O(\gamma)$] external fields.

5. THE HYDRODYNAMIC LIMIT OF THE PARTICLE SYSTEM: PROOFS

The proof of Theorem 1 is an immediate consequence of Theorem 2 and Proposition 1 given below.

Some Preliminary Definitions. In this section we will denote by $e \in \mathbb{Z}^d$ a unit lattice vector. By Σ_e we will mean the sum over all $e_j \in \mathbb{Z}^d$, $(e_j)_i = \delta_{j,i}$ $(i, j \in \{1, ..., d\})$. Let us recall the Poisson rates

$$c_\gamma(x, x+e; \eta) = \Phi(\beta \Delta_{x, x+e} H_\gamma(\eta)), \qquad x \in \Lambda_\gamma, \quad \eta \in \Omega_\gamma \tag{5.1}$$

where we used the notation

$$\Delta_{x, y} H_\gamma(\eta) = H_\gamma(\eta^{x, y}) - H_\gamma(\eta)$$

for $x, y \in \Lambda_\gamma$. We recall that $\Phi \in C^2$ satisfies (2.9), $\Phi(0) = 1$, and that $c_\gamma(x, y; \eta) = 0$ if $|x - y| \neq 1$. In the proofs we shall denote by $c_\gamma^0(x, y; \eta)$ the rates in the case in which $J \equiv 0$, that is,

$$c_\gamma^0(x, x+e; \eta) = 1, \qquad x \in \Lambda_\gamma \tag{5.2}$$

This process is called *the simple exclusion process*: the process with $J \neq 0$ will be treated as a perturbation of the simple exclusion process. Moreover, we will denote by $c_\gamma^\rho(x, y; \eta)$ the rates when $\Phi(E) = \exp(-E/2)$ $(E \in \mathbb{R})$. The law of the simple exclusion process with initial condition μ is denoted by $\mathbf{P}_\gamma^{0,\mu}$ $(\mathbf{E}_\gamma^{0,\mu})$, while the law of the process with rates $c_\gamma^\rho(x, y; \eta)$ is denoted by $\mathbf{P}_\gamma^{\rho,\mu}$ $(\mathbf{E}_\gamma^{\rho,\mu})$.

Let M_1 be the set of measurable functions ρ: $T^d \to [0, 1]$. The set M_1 is equipped with the weak topology induced by duality by $C(T^d)$, the continuous real-valued functions on T^d, according to

$$\langle \rho, G \rangle = \int_{T^d} G(r)\, \rho(r)\, dr$$

where $\rho \in M_1$ and $G \in C(T^d)$. Given $\gamma > 0$, we define the empirical measure of the process at r as

$$\rho_\gamma(r; \eta) = \sum_{x \in \Lambda_\gamma} \eta(x)\, \chi_{\prod_{i=1}^d [x_i \gamma, (x_i+1)\gamma]}(r) \tag{5.3}$$

where χ_A denotes the characteristic function of the set $A \subset T^d$. The empirical average of a function f: $\Omega_\gamma \to \mathbb{R}$ over a ball of radius $R > 0$ is defined as

$$\mathrm{Av}_{R,x} f = \frac{1}{|B(R) \cap \mathbb{Z}^d|} \sum_{y \in B(R) \cap \mathbb{Z}^d} f(\tau_{x+y} \eta) \tag{5.4}$$

where $B(R)=\{r: |r|\leqslant R\}$, $x\in\Lambda_\gamma$, and $\tau_z: \Omega_\gamma\to\Omega_\gamma$ is defined by $(\tau_z\eta)(x)=\eta(x+z)$. For $\alpha\in[0,1]$, let ν_α^{B} be the Bernoulli measure over Ω_γ such that $\nu_\alpha^{\mathrm{B}}(\eta(x))=\alpha$ for all $x\in\Lambda_\gamma$. We set

$$\tilde{f}(\alpha)=\nu_\alpha^{\mathrm{B}}(f) \tag{5.5}$$

Given $\eta\in D([0,\infty),\Omega_\gamma)$, the space of right continuous functions from $[0,\infty)$ to Ω_γ with limit from the left, equipped with the Skorohod topology, for every $t\in\mathbb{R}^+$ and $r\in T^d$ we define

$$\rho_\gamma(r,t)=\rho_\gamma(r;\eta_{\gamma^{-2}t}) \tag{5.6}$$

We will adopt the notation $\rho_\gamma=\rho_\gamma(\cdot,\cdot)$ and when we want to keep the time fixed we will write $\rho_{\gamma,t}$, which stands for $\rho_\gamma(\cdot;\eta_{\gamma^{-2}t})$. So $\rho_{\gamma,t}\in M_1$ and $\rho_\gamma\in D([0,\infty);M_1)$.

With this notation the family of measures $\{\mu_\gamma\}_{\gamma>0}$ is an initial condition associated with ρ_0 if for any $\delta>0$ and any $\phi\in C(T^d)$ we have that

$$\lim_{\gamma\to 0}\mathbf{E}_\gamma^{\mu_\gamma}(|\langle\rho_{\gamma,0},\phi\rangle-\langle\rho_0,\phi\rangle|>\delta)=0 \tag{5.7}$$

[compare with (2.14)].

A function $\rho\in C([0,\infty),M_1)$ is a weak solution of (2.16) if for all $t\in\mathbb{R}^+$ and all $\phi\in C^{2,1}(T^d\times[0,\infty))$, ρ satisfies

$$\begin{aligned}&\int_{T^d}\rho(r,t)\,\phi(r,t)\,dr-\int_{T^d}\rho_0(r)\,\phi(r,0)\,dr\\&\quad=\iint_{Q_t}\rho(r,s)\,\partial_s\phi(r,s)\,dr\,ds+\iint_{Q_t}\rho(r,s)\,\Delta\phi(r,s)\,dr\,ds\\&\qquad+\beta\iint_{Q_t}\rho(r,s)(1-\rho(r,s))(\nabla J*\rho)(r,s)\,\nabla\phi(r,s)\,dr\,ds\end{aligned} \tag{5.8}$$

in which $Q_t=T^d\times[0,t)$.

Finally, for $f\in C^k$ $(k\in\mathbb{Z}^+)$ we set

$$\|f\|_{C^k}=\|f\|_{C^0}+\sum_{i_1,\dots,i_d}\|\partial_{i_1}\cdots\partial_{i_d}f\|_{C^0}$$

($i_1,\dots,i_d\in\mathbb{Z}^+$ and $|i_1+\cdots+i_d|=k$), where $\|\cdot\|_{C^0}$ is the sup-norm. Here $\|\cdot\|_p$ will denote the L^p-norm.

Theorem 2. For any $t>0$

$$\lim_{\gamma\to 0} \mathbf{P}_{\gamma}^{\mu_\gamma}(|\langle \rho_{\gamma,t}, \phi\rangle - \langle \rho_t, \phi\rangle| > \delta) = 0 \tag{5.9}$$

where ρ_t is the unique solution of (5.8).

Remark. A by-product of the proof of Theorem 2 is that the random variable $\rho_\gamma \in D([0,\infty); M_1)$ converges weakly, as γ approaches zero, to the deterministic trajectory $\rho \in C([0,\infty); M_1)$, unique solution of (5.8).

Proof of Theorem 2. For all $\varepsilon>0$, $f\colon \Omega_\gamma \to \mathbb{R}$, and $\eta \in \Omega_\gamma$ set

$$R_{\varepsilon,\gamma}(f;\eta) = \sum_x |\mathrm{Av}_{\varepsilon\gamma^{-1},x}(f;\eta) - \tilde{f}(\mathrm{Av}_{\varepsilon\gamma^{-1},x}(\pi_0;\eta))| \tag{5.10}$$

in which $\pi_0\colon \Omega_\gamma \to \{0,1\}$ is defined as $\pi_0(\eta) = \eta(0)$.

Let us recall the following result, which is a straightforward extension to any dimension of Proposition 2.1 of ref. 17.

Lemma 1. Given a cylindrical[3] function f, $t \in \mathbb{R}^+$, and $\delta>0$, we have

$$\limsup_{\varepsilon\to 0}\ \limsup_{\gamma\to 0} \gamma^d \log \mathbf{P}_{\gamma}^{0,\nu_{1/2}^{\mathrm{B}}}\left(\gamma^d \int_0^{\gamma^{-2}t} R_{\varepsilon,\gamma}(f;\eta_s)\,ds \geqslant \delta\right) = -\infty \tag{5.11}$$

We have to extend Lemma 1 to the case of $J\neq 0$ and general initial condition. We are not going to extend the lemma to the general case, but only to $\mathbf{P}_{\gamma}^{\rho,\mu_\gamma}$: in the general case we will obtain a weaker, but sufficient, estimate via a relative entropy argument. In order to extend Lemma 1, it is sufficient to show that there is a constant c such that

$$\gamma^d \log\left(\frac{d\mathbf{P}_{\gamma}^{\rho,\mu_\gamma}}{d\mathbf{P}_{\gamma}^{0,\nu_{1/2}^{\mathrm{B}}}}(\{\eta_t\}_{t\in[0,\gamma^{-2}\tau]})\right) \leqslant c \tag{5.12}$$

for all $\eta \in D([0,\gamma^{-2}\tau]; \Omega_\gamma)$, since (5.11) and (5.12) easily imply

$$\limsup_{\varepsilon\to 0}\ \limsup_{\gamma\to 0} \gamma^d \log \mathbf{P}_{\gamma}^{\rho,\mu_\gamma}\left(\gamma^d \int_0^{\gamma^{-2}t} R_{\varepsilon,\gamma}(f,\eta_s) \geqslant \delta\right) = -\infty \tag{5.13}$$

[3] A function f on $\{0,+1\}^{\mathbb{Z}^d}$ is said to be cylindrical if there exists a finite set $A\subset \mathbb{Z}^d$ such that $f(\eta) = f(\eta')$ whenever $\eta(x) = \eta'(x)$ for all $x\in A$. Hence any cylindrical function has an obvious restriction to Ω_γ, provided that γ is small enough (i.e., if $A\subset \Lambda_\gamma$). The f which appears in formula (5.11) is precisely this restriction.

under the same conditions stated in Lemma 1. The bound (5.13) will be obtained via direct evaluation of the Radon–Nikodym (RN) derivative, whose explicit expression is

$$\log\left(\frac{d\mathbf{P}_\gamma^{p,\mu_\gamma}}{d\mathbf{P}_\gamma^{0,\nu_{1/2}^{\mathrm{B}}}}\left(\{\eta_t\}_{t\in[0,\gamma^{-2}\tau]}\right)\right)$$

$$=\log\left(\frac{d\mu_\gamma}{d\nu_{1/2}^{\mathrm{B}}}(\eta_0)\right) \tag{5.14}$$

$$-\int_0^{\gamma^{-2}\tau}\sum_{x,e}\left[c_\gamma^p(x,x+e;\eta_t)-c_\gamma^0(x,x+e;\eta_t)\right]dt \tag{5.15}$$

$$+\sum_{x,e}\int_0^{\gamma^{-2}\tau}\log\left(\frac{c_\gamma^p(x,x+e;\eta_{t^-})}{c_\gamma^0(x,x+e;\eta_{t^-})}\right)d\mathscr{T}_t^{x,x+e} \tag{5.16}$$

in which $\mathscr{T}_t^{x,x+e}$ is the Poisson process that counts the exchanges of occupation number between x to $x+e$ in the time interval $[0,t]$. We will bound the three terms of the RN derivative [(5.14)–(5.16)] separately. Let us observe that for (5.14), since $(d\mu_\gamma/d\nu_{1/2}^{\mathrm{B}})(\eta)\leqslant 2^{\gamma^{-d}}$, the desired estimate is immediately obtained.

Now and later we will make use of the following lemma.

Lemma 2. For $x\in T^d$ and $\eta\in\Omega_\gamma$, let

$$h_\gamma^{x,e}(\eta)=\Delta_{x,x+e}H_\gamma(\eta)-\gamma(\eta(x+e)-\eta(x))\times\left[\gamma^d\sum_z\eta(z)(e\cdot\nabla J)(\gamma(x-z))\right] \tag{5.17}$$

There exists a constant c_1 such that for every $x\in\Lambda_\gamma$, every $\eta\in\Omega_\gamma$, and every e

$$|h_\gamma^{x,e}(\eta)|\leqslant c_1\gamma^2 \tag{5.18}$$

In particular, there is a constant c_2 such that for all $x\in\Lambda_\gamma$ and all $\eta\in\Omega_\gamma$

$$|\Delta_{x,x+e}H_\gamma(\eta)|\leqslant c_2\gamma \tag{5.19}$$

Remark. An immediate consequence of Lemma 2 is that the rates are bounded: for any J, β, and Φ, there exists γ_0 such that

$$\sup_{\gamma\in(0,\gamma_0)}\ \sup_{\eta,x,y} c_\gamma(x,y;\eta)\leqslant 2 \tag{5.20}$$

Proof of Lemma 2. By definition,

$$-2\Delta_{x,y}H_\gamma(\eta)=\sum_{z,z'}[\eta^{x,y}(z)\,\eta^{x,y}(z')-\eta(z)\,\eta(z')]\,\gamma^d J((z-z')\,\gamma) \tag{5.21}$$

Add and subtract in the square brackets $\eta^{x,y}(z)\,\eta(z')$ and use the symmetry $J(r)=J(-r)$ to get

$$\begin{aligned}
&-2\Delta_{x,y}H_\gamma(\eta)\\
&\quad=(\eta(y)-\eta(x))\sum_z(\eta^{x,y}(z)+\eta(z))\,\gamma^d[J(\gamma(z-x))-J(\gamma(z-y))]\\
&\quad=-2(\eta(y)-\eta(x))\sum_z\eta(z)\,\gamma^d[J(\gamma(y-z))-J(\gamma(x-z))] &(5.22)\\
&\qquad+2(\eta(y)-\eta(x))^2\,\gamma^d[J(0)-J(\gamma(x-y))] &(5.23)
\end{aligned}$$

and if $|x-y|=1$, the modulus of the term (5.23) can be bounded by $2\,\|J\|_{C_1}\gamma^{d+1}$ and we can substitute the discrete gradient in (5.22) with $(x-y)\cdot\nabla$ and the error will be bounded by $\|J\|_{C^2}\gamma^2$, so that (5.18) is proven with $c_1=2\,\|J\|_{C_1}+\|J\|_{C^2}$. Formula (5.19) follows immediately from (5.17) and (5.18). This ends the proof of Lemma 2. ∎

Let us go on with the proof of Theorem 2. The term in (5.15) can be written as

$$\int_0^{\gamma^{-2}\tau}\sum_{x,e}\eta_t(x)(1-\eta_t(x+e))[c_\gamma^p(x,x+e;\eta_t)-1]\,dt \tag{5.24}$$

By Lemma 2 we obtain that (5.24) is equal to

$$\begin{aligned}
&\int_0^{\gamma^{-2}\tau}\frac{1}{2}\sum_{x,e}\Bigg\{\eta_t(x)(1-\eta_t(x+e))\\
&\quad\times\left[\exp\left(\frac{\beta\gamma}{2}\gamma^d\sum_z\eta_t(z)(e\cdot\nabla J)(\gamma(z-x))+\frac{\beta}{2}h_\gamma^{x,e}(\eta_t)\right)-1\right]\\
&\quad+\eta_t(x+e)(1-\eta_t(x))\\
&\quad\times\Bigg[\exp\Bigg(-\frac{\beta}{2}\gamma^{d+1}\sum_z\eta_t(z)(e\cdot\nabla J)(\gamma(z-x-e))\\
&\quad+\frac{\beta}{2}h_\gamma^{x+e,-e}(\eta_t)\Bigg)-1\Bigg]\Bigg\}\,dt
\end{aligned} \tag{5.25}$$

Set $g_\gamma(x;\eta)=(\beta/2)(\gamma^d \sum_z \eta(z)(e\cdot\nabla J)(\gamma(z-x))$. The modulus of (5.25) can be bounded by

$$\left|\int_0^{\gamma^{-2}\tau} \tfrac{1}{2}\sum_{x,e} \{-\eta_t(x)\,\eta_t(x+e)\right.$$

$$\times[\exp(\gamma g_\gamma(x;\eta_t))+\exp(-\gamma g_\gamma(x+e;\eta_t))-2]$$

$$\left.+\eta_t(x)[\exp(\gamma g_\gamma(x;\eta_t))+\exp(-\gamma g_\gamma(x;\eta_t))]\}\,dt\right|$$

$$+2(c_1\gamma^2)(\tau\gamma^{-2})(d\gamma^{-d})\leqslant c\gamma^{-d}$$

in which we used (5.20), (5.18), (5.19), the fact that $|g_\gamma(x;\eta)-g_\gamma(x+e;\eta)| \leqslant (\beta/2)\,\|J\|_{C^2}\,\gamma$, and $c=c(d,\tau,\beta,\|J\|_{C^2})<\infty$. The third and last piece (5.16) in the RN derivative is treated as follows. Given a sample path $\{\eta_t\colon t\in[0,\gamma^{-2}\tau]\}$, call $n=\sum_{x,e}\mathscr{T}^{x,x+e}_{\gamma^{-2}\tau}$ the total number of exchanges in the interval $[0,\gamma^{-2}\tau]$, which is finite for every $\eta_\cdot\in D([0,\gamma^{-2}\tau],M_1)$. The ith jump ($i=1,\ldots,n$) happens at $t=t^i$ and it moves a particle from x^i to x^i+e^i. The last term in the RN derivative is equal to

$$\sum_{x,e}\int_0^{\gamma^{-2}\tau}\log\left(\frac{c^p_\gamma(x,x+e;\eta_{t^-})}{c^0_\gamma(x,x+e;\eta_{t^-})}\right)d\mathscr{T}^{x,x+e}_t$$

$$=\sum_{i=1}^{n}(\Delta_{x^i,x^i+e^i}H_\gamma)(\eta_{t_i^-})$$

$$=H_\gamma(\eta_{t\gamma^{-2}})-H_\gamma(\eta_0) \tag{5.26}$$

which is easily bounded by $2\max_\eta|H_\gamma(\eta)|\leqslant(\|J\|_{C^0})\,\gamma^{-d}$ and so the proof of (5.12) [and so of (5.13)] is complete.

Clearly (5.13) implies that for ε sufficiently small

$$\lim_{\gamma\to 0}\mathbf{P}^{p,\mu_\gamma}_\gamma\left(\gamma^d\int_0^{\gamma^{-2}t}R_{\varepsilon,\gamma}(f,\eta_s)\geqslant\delta\right)=0 \tag{5.27}$$

The aim is to obtain a similar inequality for $\mathbf{P}^{\mu_\gamma}_\gamma$. To this purpose let us introduce the relative entropy between $\mathbf{P}^{\mu_\gamma}_\gamma$ and $\mathbf{P}^{p,\mu_\gamma}_\gamma$ on the time interval $[0,\gamma^{-2}\tau]$:

$$H^\gamma_\tau(\mathbf{P}^{\mu_\gamma}_\gamma\,|\,\mathbf{P}^{p,\mu_\gamma}_\gamma)=\mathbf{E}^{\mu_\gamma}_\gamma\left[\log\left(\frac{d\mathbf{P}^{\mu_\gamma}_\gamma}{d\mathbf{P}^{p,\mu_\gamma}_\gamma}(\{\eta_t\}_{t\in[0,\gamma^{-2}\tau]})\right)\right] \tag{5.28}$$

and we will evaluate (5.28) directly by writing the RN derivative explicitly. The obtained expression is absolutely analogous to (5.15) and (5.16) (now the expressions $c_\gamma - c_\gamma^p$ and c_γ / c_γ^p replace $c_\gamma^p - c_\gamma^0$ and c_γ^p / c_γ^0, respectively) and there is no term (5.14), since the initial condition is the same. Observe that by differentiating (2.9) we obtain

$$\Phi'(E) = -\exp(-E)\,\Phi(-E) - \Phi'(E)\exp(-E) \tag{5.29}$$

and so $\Phi'(0) = -\Phi(0)/2 = -1/2$. This implies that there exists a constant c such that

$$|c_\gamma(x, y; \eta) - c_\gamma^p(x, y; \eta)| \leqslant c\gamma^2$$

$$|\log(c_\gamma(x, y; \eta)/c_\gamma^p(x, y; \eta))| \leqslant c\gamma^2$$

Hence

$$H_\tau^\gamma(\mathbf{P}_\gamma^{\mu_\gamma} \,|\, \mathbf{P}_\gamma^{p,\mu_\gamma}) \leqslant (d\gamma^{-d})(\gamma^{-2}\tau)(c\gamma^2) + (c\gamma^2)\,\mathbf{E}_\gamma^{\mu_\gamma}(n) \leqslant c'\gamma^{-d} \tag{5.30}$$

in which n is the total number of Poisson exchanges in $[0, \gamma^{-2}\tau]$. The bound in (5.30) follows directly from (5.20). By applying a well-known entropy inequality [see, e.g., ref. 31, formula (2.18)] to our setup we obtain

$$\begin{aligned} &\mathbf{P}_\gamma^{\mu_\gamma}\left(\gamma^d \int_0^{\gamma^{-2}t} R_{\varepsilon,\gamma}(f, \eta_s) \geqslant \delta\right) \\ &\leqslant \frac{\log 2 + H_\tau^\gamma(\mathbf{P}_\gamma^{\mu_\gamma} \,|\, \mathbf{P}_\gamma^{p,\mu_\gamma})}{\log[1 + 1/\mathbf{P}_\gamma^{p,\mu_\gamma}(\gamma^d \int_0^{\gamma^{-2}t} R_{\varepsilon,\gamma}(f, \eta_s) \geqslant \delta)]} \end{aligned} \tag{5.31}$$

From (5.13), (5.30), and (5.31) it directly follows that

$$\lim_{\varepsilon \to 0}\ \limsup_{\gamma \to 0}\ \mathbf{P}_\gamma^{\mu_\gamma}\left(\gamma^d \int_0^{\gamma^{-2}t} R_{\varepsilon,\gamma}(f, \eta_s) \geqslant \delta\right) = 0 \tag{5.32}$$

Let $\phi \in C^{2,1}(T^d \times [0, \infty))$: we will use the notation $\phi_t(\cdot) = \phi(\cdot, t)$. Define

$$\langle \rho_{\gamma,t}, \phi_t \rangle_\gamma = \gamma^d \sum_x \eta_{\gamma^{-2}t}(x)\, \phi(\gamma x, t) \tag{5.33}$$

and observe that for any $t > 0$ there exists $c = c(\phi)$ such that

$$|\langle \rho_{\gamma,t}, \phi_t \rangle_\gamma - \langle \rho_{\gamma,t}, \phi_t \rangle| \leqslant c\gamma \tag{5.34}$$

which makes the two quantities appearing in the left-hand side of (5.34) interchangeable for our purposes. Let us define

$$\Gamma_{1,\gamma}(t) = \gamma^{-2} L_\gamma(\langle \rho_{\gamma,t}, \phi_t \rangle_\gamma) + \partial_t(\langle \rho_{\gamma,t}, \phi_t \rangle_\gamma) \tag{5.35}$$

where by partial derivative in t we mean the derivative with respect to the time dependence of ϕ, and

$$\Gamma_{2,\gamma}(t) = \gamma^{-2} L_\gamma[(\langle \rho_{\gamma,t}, \phi_t \rangle_\gamma)^2] - 2(\langle \rho_{\gamma,t}, \phi_t \rangle_\gamma)\, L_\gamma(\langle \rho_{\gamma,t}, \phi_t \rangle_\gamma) \tag{5.36}$$

It is well known (see, for instance, ref. 7) that

$$M_\gamma(\tau; \phi) \equiv \langle \rho_{\gamma,\tau}, \phi_\tau \rangle_\gamma - \langle \rho_{\gamma,0}, \phi_0 \rangle_\gamma - \int_0^\tau \Gamma_{1,\gamma}(t)\, dt \tag{5.37}$$

and

$$N_\gamma(\tau; \phi) \equiv (M_\gamma(\tau; \phi))^2 - \int_0^\tau \Gamma_{2,\gamma}(t)\, dt \tag{5.38}$$

are $\mathbf{P}_\gamma^{\mu_\gamma}$-martingales with respect to the filtration generated by the family of random variables $\{\eta_\tau\}_{\tau \in \mathbb{R}^+}$. Moreover, they vanish at $\tau = 0$.

Let us start by computing $\Gamma_{1,\gamma}$ and $\Gamma_{2,\gamma}$. By using the definition of the generator, we obtain

$$\begin{aligned} \Gamma_{1,\gamma}(t) = {} & \gamma^{-2}\gamma^d \sum_{x,e} c_\gamma(x, x+e; \eta_{\gamma^{-2}t}) \\ & \times [(\phi(\gamma x, t) - \phi(\gamma(x+e), t)(\eta_{\gamma^{-2}t}(x+e) - \eta_{\gamma^{-2}t}(x))] \\ & + \gamma^d \sum_x \partial_t \phi(\gamma x, t)\, \eta_{\gamma^{-2}t}(x) \end{aligned} \tag{5.39}$$

and the first term on the right-hand side of (5.39) can be rewritten as

$$\begin{aligned} & \gamma^d \sum_x (\Delta\phi)(\gamma x, t)\, \eta_{\gamma^{-2}t}(x) \\ & \quad + \gamma^d \sum_{x,e} \eta_{\gamma^{-2}t}(x)(1 - \eta_{\gamma^{-2}t}(x+e)) \\ & \quad \times \left[\frac{\beta\gamma^d}{2} \sum_z \eta_{\gamma^{-2}t}(z)(e \cdot \nabla J)(\gamma(x-z)) \right] (e \cdot \nabla\phi)(\gamma x, t) \end{aligned} \tag{5.40}$$

apart from corrections that are uniformly vanishing as γ goes to zero, and so $|\Gamma_{1,\gamma}(t)|$ is uniformly bounded [this follows from the observation following (5.29) that $\Phi'(0) = -1/2$]. Moreover,

$$\Gamma_{2,\gamma}(t) = \gamma^{-2} \sum_{x,e} c_\gamma(x, x+e; \eta_{\gamma^{-2}t})$$

$$\times \left[\gamma^d \sum_z \eta_{\gamma^{-2}t}^{x,x+e}(z)\, \phi(z,t) - \gamma^d \sum_z \eta_{\gamma^{-2}t}(z)\, \phi(z,t) \right]^2$$

$$= \gamma^{-2}\gamma^{2d} \sum_{x,e} c_\gamma(x, x+e; \eta_{\gamma^{-2}t})[\phi(\gamma(x+e), t) - \phi(\gamma x, t)]^2 \quad (5.41)$$

and from (5.41) we obtain

$$|\Gamma_{2,\gamma}(t)| \leqslant 2\, \|\phi\|_{C^1}\, \gamma^d \quad (5.42)$$

The fact that $|\langle \rho_{\gamma,t}, \phi_t \rangle|$, $|\Gamma_{1,\gamma}(t)|$, and $|\Gamma_{2,\gamma}(t)|$ are bounded uniformly in $t \in [0, \tau]$ ($\tau \in \mathbb{R}^+$), $\eta \in D([0,\tau]; \Omega_\gamma)$, and $\gamma > 0$ immediately implies that the family of random variables $\{\rho_\gamma\}_{\gamma>0}$ is (sequentially) relatively compact in $D([0,\infty); M_1)$ (see, for instance, refs. 17 and 15). Moreover, every limit point ρ of $\{\rho_\gamma\}_{\gamma>0}$ lies in $C([0,\infty); M_1)$, which follows directly from the fact that with $\mathbf{P}_\gamma^{\mu_\gamma}$-probability one the $\sup_t |\sum_{x,e} (\mathcal{T}_t^{x,x+e} - \mathcal{T}_{t^-}^{x,x+e})| = 1$ and so the quantity in (5.33) can have jumps of size at most $\|\phi\|_{C^0}\,\gamma$, which vanish in the limit (see ref. 7, Theorem 2.7.8). We are then left with the task of identifying the limit.

Let us observe that (5.42) and Doob's martingale inequality imply that for every $\delta_1 > 0$, $\phi \in C^{2,1}$, and $\tau \in \mathbb{R}^+$

$$\lim_{\gamma \to 0} \mathbf{P}_\gamma^{\mu_\gamma}\Big(\sup_{t \in [0,\tau]} |M_\gamma(t;\phi)| > \delta_1 \Big) = 0 \quad (5.43)$$

Observe that, because of the smoothness of ϕ and J, for every $\delta_2 > 0$, there is an $\varepsilon_0 > 0$ such that for all $\varepsilon \in (0, \varepsilon_0]$ the following holds:

$$\bigg| \beta \int_0^\tau \gamma^d \sum_{x,e} \pi_x(\eta_t)\, \pi_{x+e}(\eta_t) (\rho_{\gamma,t} * e \cdot \nabla J)(\gamma x)(e \cdot \nabla\phi)(\gamma x, t)\, dt$$

$$- \int_0^\tau \gamma^d \sum_{x,e} \mathrm{Av}_{\varepsilon^{-1}\gamma, x}(\pi_0 \pi_e, \eta_t)\, \beta(\rho_{\gamma,t} * e \cdot \nabla J)(\gamma x)(e \cdot \nabla\phi)\, dt \bigg| \leqslant \delta_2$$

and by (5.32) we obtain that for every $\delta > 0$

$$\lim_{\varepsilon \to 0} \limsup_{\gamma \to 0} \mathbf{P}_{\gamma}^{\mu_\gamma} \left(\left| \beta \int_0^\tau \gamma^d \sum_{x,e} \mathrm{Av}_{\varepsilon^{-1}\gamma,x}(\pi_0 \pi_e, \eta_t)(\rho_{\gamma,t} * e \cdot \nabla J) \right.\right.$$
$$\times (\gamma x)(e \cdot \nabla \phi)(\gamma x, t)\, dt$$
$$- \int_0^\tau \gamma^d \sum_{x,e} (\mathrm{Av}_{\varepsilon^{-1}\gamma,x}(\pi_0, \eta_t))^2$$
$$\left.\left. \times [\beta(\rho_{\gamma,t} * e \cdot \nabla J)](e \cdot \nabla \phi)(\gamma x, t)\, dt \right| > \delta \right)$$
$$\leqslant \lim_{\varepsilon \to 0} \limsup_{\gamma \to 0} \mathbf{P}_{\gamma}^{\mu_\gamma} \left(\beta \, \|\nabla \phi\|_{C^0} \, \|J\|_{C^1} \right.$$
$$\left. \times \int_0^\tau \gamma^d \sum_{x,e} |\mathrm{Av}_{\varepsilon^{-1}\gamma,x}(\pi_0 \pi_e, \eta_t) - (\mathrm{Av}_{\varepsilon^{-1}\gamma,x}(\pi_0, \eta_t))^2| \, dt > \delta \right) = 0 \quad (5.44)$$

Take the limit in γ along a convergent subsequence to obtain from (5.37), (5.40), (5.43), and (5.44) that for every $\delta > 0$ with $\mathbf{P}_{\gamma}^{\mu_\gamma}$-probability going to 1 as ε goes to zero, the limit point ρ satisfies the inequality

$$\left| \int_{T^d} \rho(r, \tau)\, \phi(r, \tau)\, dr - \int_{T^d} \rho(r, 0)\, \phi_0(r)\, dr \right.$$
$$- \iint_{Q_\tau} \rho(r, t)(\partial_t \phi + \Delta \phi)\, dr\, dt$$
$$\left. - \iint_{Q_\tau} \nabla \phi(r, t)(\rho - (\alpha_\varepsilon * \rho)^2)[\beta(\rho * \nabla J)]\, dr\, dt \right| < \delta$$

in which

$$\alpha_\varepsilon = (2\varepsilon)^{-d} \chi_{[-\varepsilon, +\varepsilon]^d}$$

Let ε go to zero and by the arbitrariness of δ we obtain that ρ is a weak solution of (2.16). To complete the proof of Proposition 1, we have to show that there is only one weak solution.

Let us then consider ρ_1 and ρ_2 weak solutions of (2.16) with the same initial datum. Set $w = \rho_1 - \rho_2$. For each $(r_0, t_0) \in Q_\tau$ and each $\varepsilon > 0$ let us define

$$\phi_{r_0, t_0, \varepsilon}(r, t) = \theta(t_0 + \varepsilon - t, r - r_0) \quad (5.45)$$

in which $\theta(r, t) = \sum_{x \in \mathbb{Z}^d} \mathscr{G}_t(r - x)$ is the periodic heat kernel, which solves $\partial_t \theta = \Delta\theta$ on $T^d \times (0, \infty)$. So the test function $\phi_{r_0, t_0, \varepsilon} \in C^\infty(Q_{t_0})$ solves

$$\partial_t \phi_{r_0, t_0, \varepsilon}(r, t) + \Delta\phi_{r_0, t_0, \varepsilon}(r, t) = 0 \tag{5.46}$$

for all $(r, t) \in Q_{t_0}$. By using (5.46), we obtain $[\sigma(\rho) = \beta\rho(1 - \rho)]$

$$\begin{aligned}\int_{T^d} &\phi_{r_0, t_0, \varepsilon}(r, t_0)\, w(r, t_0)\, dr \\ &= \iint_{Q_{t_0}} \nabla\phi_{r_0, t_0, \varepsilon}(r, t)[(\nabla J * w)(r, t)\, \sigma(\rho_1(r, t)) \\ &\qquad + (\nabla J * \rho_2)(r, t)\, \sigma'(\tilde\rho)\, w(r, t)]\, dr\, dt\end{aligned}$$

in which $\tilde\rho \in [(\rho_1 \wedge \rho_2) \vee 0, (\rho_1 \vee \rho_2) \wedge 1]$. Since $\sigma(\rho_1) \in [0, \beta/4]$ and $|\sigma'(\tilde\rho)| \leqslant \beta$, we obtain that

$$\begin{aligned}\left|\int_{T^d} \phi_{r_0, t_0, \varepsilon}(r, t_0)\, w(r, t_0)\, dr\right| \\ \leqslant c[\beta\, \|\nabla J\|_1] \operatorname*{ess\,sup}_{(r,t) \in Q_{t_0}} |w(r, t)| \iint_{Q_{t_0}} |\nabla\phi_{r_0, t_0, \varepsilon}(r, t)|\, dr\, dt\end{aligned} \tag{5.47}$$

The integral term on the right-hand side of (5.47) is bounded by $c(\sqrt{t_0 + \varepsilon} - \sqrt{\varepsilon})$, where c is a constant depending only on the dimension d. By observing that $\phi_{r_0, t_0, \varepsilon}(\cdot, t_0)$ is an approximate identity in ε, we obtain that there exists a constant C (which depends only on the L^1 norm of ∇J and on d) such that for almost every $(r_0, t_0) \in Q_\tau$ we have that

$$|w(r_0, t_0)| \leqslant C\sqrt{t_0} \operatorname*{ess\,sup}_{(r,t) \in Q_{t_0}} |w(r, t)|$$

and so

$$\operatorname*{ess\,sup}_{(r,t) \in Q_\tau} |w(r, t)| \leqslant C\sqrt{\tau} \operatorname*{ess\,sup}_{(r,t) \in Q_\tau} |w(r, t)|$$

Choosing τ such that $C\sqrt{\tau} < 1$ implies local uniqueness in time. But C depends only on J and d, so the global uniqueness follows by a bootstrap argument. ∎

Proposition 1. If $\rho_0 \in C^2(T^d)$, then the solution ρ of (5.8) can be chosen in $C^{2,1}(T^d \times \mathbb{R}^+)$.

Proof. Given a solution of (5.8), define

$$F(r, t) = (\nabla J * \rho_t)(r) \tag{5.48}$$

so $F \in C^{\infty,0}(T^d \times \mathbb{R}^+; \mathbb{R}^d)$. Observe that this means that ρ solves in the weak sense analogous to (5.8) the equation

$$\partial_t \rho = \nabla \cdot \{\nabla \rho - \beta \rho(1-\rho) F\} \tag{5.49}$$

with initial condition $\rho_0 \in C^2$. But (5.49) is a nondegenerate parabolic equation which has a classical solution (Chapter 7, Section 4 of ref. 11), besides having a unique weak solution by the same argument used to prove uniqueness in the proof of Theorem 2. ∎

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