Strong phase separation in a model of sedimenting lattices

Rangan Lahiri,[1,*] Mustansir Barma,[1,†] and Sriram Ramaswamy[2,‡]
[1]*Department of Theoretical Physics, Tata Institute of Fundamental Research, Homi Bhabha Road, Mumbai 400 005, India*
[2]*Centre for Condensed Matter Theory, Department of Physics, Indian Institute of Science, Bangalore 560 012, India*
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We study the steady state resulting from instabilities in crystals driven through a dissipative medium, for instance, a colloidal crystal which is steadily sedimenting through a viscous fluid. The problem involves two coupled fields, the density and the tilt; the latter describes the orientation of the mass tensor with respect to the driving field. We map the problem to a one-dimensional lattice model with two coupled species of spins evolving through conserved dynamics. In the steady state of this model each of the two species shows macroscopic phase separation. This phase separation is robust and survives at all temperatures or noise levels— hence the term strong phase separation. This sort of phase separation can be understood in terms of barriers to remixing which grow with system size and result in a logarithmically slow approach to the steady state. In a particular symmetric limit, it is shown that the condition of detailed balance holds with a Hamiltonian which has infinite-ranged interactions, even though the initial model has only local dynamics. The long-ranged character of the interactions is responsible for phase separation, and for the fact that it persists at all temperatures. Possible experimental tests of the phenomenon are discussed.

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I. INTRODUCTION

A. Background

Sedimentation—the settling of heavier particles in a lighter fluid—is a rich source of intriguing physics [1]. The steadily sedimenting state arises, of course, from a balance between gravity and viscosity. Viscous damping in this nonequilibrium steady state has important consequences: when a given particle is slowed down by the fluid, its momentum does not disappear, but produces disturbances in the fluid which affect the motion of other particles [2,3]. This makes sedimentation a challenging problem in the statistical physics of driven many-body systems.

In the general area of nonequilibrium steady states, much recent progress has come by stepping away from the difficulties of hydrodynamics and focusing instead on simple driven lattice-gas models [4]. In fact, intimate connections were discovered by two of the present authors (hereafter LR) [5] between these models and the physics of sedimenting crystalline suspensions (as well as a closely related problem, a flux-point lattice moving through a superconducting slab). The LR model was based on two crucial properties of collective settling discovered by Crowley [6] in his theoretical and experimental studies of hard spheres sedimenting in a viscous medium: (i) The *magnitude* of the local settling velocity of a region of the crystal depends on its *concentration*, i.e., on the particle number density in that region, and (ii) the *direction* of the local settling velocity depends on its *tilt*, that is, the orientation, relative to the applied force (gravity) of the principal axes of the local particle distribution. These effects, which also follow from symmetry arguments, were incorporated into a natural one-dimensional model for the coupled, stochastic, local spin-exchange dynamics of two sets of Ising variables, $\{\sigma_i\}$ with states denoted by $+$ and $-$ for the concentration relative to the mean, and $\{\tau_i\}$ with states denoted by / and \ for the tilt, on the sites i of a one-dimensional lattice. Analysis of this model leads to several interesting results, some published in [5] and some which we present here.

B. Update rules

Our results will be easier to understand after a quick summary of the update rules of the lattice model, which we turn to next. This will also serve to underline the simple nature and potentially wide applicability of the model. It is convenient to place the $\{\sigma_i\}$ and $\{\tau_i\}$ on two sublattices of our one-dimensional lattice; we label sites on the first sublattice by integers, and those on the other by half-integers. A configuration is then a string $\tau_{1/2}\sigma_1\tau_{3/2}\sigma_2\tau_{5/2}\sigma_3\tau_{7/2}\sigma_4\ldots$, say $/+\backslash-/-/+\backslash\ldots$. Using the above notation for the states of the two variables, and denoting the rate of an exchange process by W, the probabilities per unit time for the various possible exchanges can be represented succinctly by

$$W(+\backslash-\rightarrow-\backslash+)=D+a,$$

$$W(-\backslash+\rightarrow+\backslash-)=D-a,$$

$$W(-/+\rightarrow+/-)=D'+a',$$

$$W(+/-\rightarrow-/+)=D'-a',$$

$$W(/+\backslash\rightarrow\backslash+/)=E+b,$$

$$W(\backslash+/\rightarrow/+\backslash)=E-b,$$

$$W(\backslash-/\rightarrow/-\backslash)=E'+b',$$

$$W(/-\backslash\rightarrow\backslash-/)=E'-b', \tag{1}$$

*Deceased.
†Electronic address: barma@theory.tifr.res.in
‡Electronic address: sriram@physics.iisc.ernet.in

where the first line, for example, represents the rate of $+-$ going to $-+$ in the presence of a downtilt \, and so on. The quantities D,E,D',E' (all positive) and a,b,a',b' are all, in principle, independent parameters but we will argue below that the case of physical interest and relevance to the sedimentation and driven flux-lattice problems is $\mathrm{sgn}\, a = \mathrm{sgn}\, a'$, $\mathrm{sgn}\, b = \mathrm{sgn}\, b'$, and that the quantity which controls the qualitative behavior of the model is then $\alpha = \mathrm{sgn}(ab)$. We find two completely distinct kinds of behavior, depending on whether α is positive or negative. If $\alpha<0$, the steady state of the model is a mixture of pluses and minuses, and of uptilts and downtilts, which is statistically homogeneous on a coarse-grained level. If $\alpha>0$, such a state is *unstable* with respect to fluctuations which drive it to a strongly phase-separated state of a type defined and discussed below. We refer to the cases $\alpha<0$ and $\alpha>0$ as the stable and unstable LR (SLR and ULR) models, respectively.

C. Strong phase separation: Summary of results

The focus of this paper is the study of phase separation phenomena of a new and unusual sort, in the unstable LR (ULR) model of sedimenting colloidal crystals described above. Following the appearance of the LR model, the same type of phase separation was shown to occur [7] in a three-species permutation-symmetric model on a one-dimensional lattice with periodic boundary conditions, with dynamics which may be regarded as a simplified version of that in [5]. A further generalization which breaks the permutation symmetry between the three species was studied in [8]. The underlying mechanism of phase separation appears robust and simple enough that it might be worth looking for in other systems. Here is a summary of our results.

(1) In the present context, phase separation involves the spontaneous formation of macroscopic domains of + and − as well as / and \ in the ULR model [5] . This segregation is robust in that it survives at all temperatures T. Let us recall that most statistical systems which show phase separation at low T (or low noise level, in nonequilibrium cases [9,10]) lose this property at higher T or noise strengths. Certainly if one were to think in terms of energy and entropy, this would be the general expectation. Against this backdrop, a phase separation so robust as to persist at *all* finite T, and in a one-dimensional system at that, is quite unexpected. We suggest the name strong phase separation (SPS) for this unusual phenomenon.

The importance of SPS in the ULR model arises from the close relation of the latter to a physically realizable system of considerable current interest, namely, sedimenting colloidal crystals. Towards the end of this paper we suggest experiments which can be performed on fluidized beds of colloidal crystals to test some of the ideas presented in this work.

(2) The occurrence of SPS can be seen best in a certain limit in which the dynamics of the ULR model obeys the condition of detailed balance. In this limit, an energy function E can be constructed such that the steady-state probability of a configuration $\{\sigma_i,\tau_{i-1/2}\}$ is proportional to $\exp[-E(\{\sigma_i,\tau_{i-1/2}\}/T)]$. Although the dynamics is entirely local and involves rates of order unity, the emergent energy function E for the effective equilibrium theory involves interactions of unbounded range. As a result, E has a nonextensive (more properly, superextensive) character, which is how our model and those of [7,8] manage to get around the usual obstacles [11] to phase separation in one dimension. In our model, E has a simple interpretation: it is the energy of a collection of particles, viz. the σ_i's, in a potential landscape built from $\{\tau_{i-1/2}\}$. The superextensivity is then a consequence of having potential energy wells whose depths scale with the system size.

Thermodynamic properties can be calculated in the strongly phase-separated state. In particular, the width of the interfacial region is found to vanish as $T\to 0$ and diverge as $T\to\infty$.

(3) Strong phase separation is a robust phenomenon, and persists even when the condition of detailed balance does not hold. This can be seen through arguments [5] based purely on kinetics without recourse to an energy function: the transport of a + from one end of a $+++\ldots+$ domain to a point a distance n away requires a time which grows exponentially with n, as n moves against the tilt field would be required. Thus a macroscopically phase-separated state would be expected to survive infinitely long in the infinite size limit.

(4) Although phase separation is inevitable in the ULR model [5] and in the models of [7,8], the kinetics of domain growth is anomalously slow. The barriers that oppose the remixing of the macroscopically segregated state also inhibit the processes of diffusion that cause large domains to grow at the expense of smaller ones. These barriers, moreover, are produced by the dynamics of the model, not introduced *ex machina* in the form of quenched randomness. This results in intriguing *aging* effects: for instance, the growth of domains is logarithmic in time, as has been verified in numerical studies in [7]. Further, in the detailed-balance limit, the decrease of $E(\{\sigma_i,\tau_{i-1/2}\})$ is logarithmic in time as well. Thus, despite the *existence* of a thermodynamic equilibrium state in the detailed-balance limit, a system which starts from a random initial condition has an extraordinarily difficult time reaching it. Such a system is best thought of as perpetually evolving, never in a truly steady state, sinking slowly into progressively deeper minima, in a manner which recalls the glassy state of the model of [12].

(5) Arguments given in [5] already amounted to showing that SPS occurred in the ULR model. Specifically, it was shown there that the remixing of phase-separated domains would *always* be opposed by barriers whose height diverged with the system size. The simulation results of [5], however, were complicated by the presence of a repulsion between adjacent + sites, which modeled interactions between charged colloidal particles. Increasing this repulsion beyond a threshold value led, in the numerical studies of [5], to an apparent loss of phase separation. It is now clear, from the calculations reported in the present paper, that the observed remixing [5] was a finite-size effect.

D. Outline

The rest of this paper is organized as follows. In Sec. II, we review the derivation [5] of continuum equations of motion for a crystalline array moving through a dissipative medium, and show how, at the linearized level, they lead to either a new class of ''kinematic waves'' [13] or an instabil-

ity towards phase separation. Section II closes by presenting a simplified one-dimensional continuum model which retains all the essential features of the higher-dimensional problem. In Sec. III, we use arguments similar to those connecting the noisy Burgers equation to the driven diffusive lattice gas [4] to construct the LR lattice-gas model [5] whose long-wavelength limit has the relevant physics of the aforementioned one-dimensional continuum equations. We show, in a certain highly symmetric limit, that the unstable LR model has a detailed balance property. In this limit we demonstrate strong phase separation and calculate thermodynamic quantities. Further, we give arguments to show that SPS occurs in the entire parameter range of the ULR model. We argue that the coarsening of domains in the ULR model is ultraslow, with a characteristic length scale growing logarithmically in time. An analysis of a continuum model for SPS is the subject of Sec. IV. Section V summarizes our results and suggests experiments to test our predictions.

II. CONTINUUM DYNAMICAL MODEL FOR A MOVING CRYSTAL

A. Motivation

The LR lattice-gas model [5] arose as a simplified description of the dynamics of a crystal moving steadily through a dissipative medium. It is therefore useful to review the construction of the continuum equations of motion for such a system. There are at least two physical situations where this dynamical problem arises: (i) the steadily sedimenting colloidal crystal mentioned above; (ii) a flux-point lattice moving through a thin slab of type II superconductor under the action of the Lorentz force due to an applied current. In (ii), the dissipation comes both from the normal core of the vortices and from disturbances in the order-parameter and electromagnetic fields in the region around the vortices. There is, in principle, an important difference between the sedimentation and moving flux-lattice problems: in the former, the disturbances produced by the moving crystal are carried to arbitrarily large length scales by the long-ranged hydrodynamic interaction, while in the latter, both electromagnetic and order-parameter disturbances are screened and are thus limited to a finite range. A complete analysis of the sedimentation dynamics of a three-dimensional colloidal crystal thus requires the inclusion of the hydrodynamic velocity field as a dynamical variable. Instead, we consider an experimental geometry in which a thin slab of colloidal crystal (with interparticle spacing $\ell \gg$ particle size) is confined to a container with dimensions $L_x, L_z \gg L_y \sim \ell$ (gravity is along $-\hat{\mathbf{z}}$). The *local* hydrodynamics that leads to the configuration-dependent mobilities [6,5] is left unaffected by this, but the long-ranged hydrodynamic interaction is screened in the xz plane on scales $\gg L_y$ by the no-slip boundary condition at the walls. The model equations (4) in dimension $d=2$ apply to such a system.

B. Constructing the equations

Our construction of the equations of motion ignores inertial terms, which is justified both for the confined colloidal crystal and, except at very low temperatures [14], for the flux lattice. Rather than keeping track of individual particles, we work on scales $\gg \ell$, treating the colloidal crystal or flux lattice as a permeable elastic continuum whose distortions at point $\mathbf{r}$ and time t are described by the (Eulerian) displacement field $\mathbf{u}(\mathbf{r},t)$. In general, the equation of motion in the completely overdamped limit has the form velocity $=$mobility $\times$ force, i.e.,

$$\frac{\partial}{\partial t}\mathbf{u} = \boldsymbol{\mu}(\nabla \mathbf{u})(\mathbf{K}\nabla\nabla\mathbf{u} + \mathbf{F} + \mathbf{f}). \tag{2}$$

In Eq. (2), the first term in parentheses on the right-hand side represents elastic forces, governed by the elastic tensor $\mathbf{K}$, the second ($\mathbf{F}$) is the applied force (gravity for the colloidal crystal and the Lorentz force for the flux lattice), and $\mathbf{f}$ is a noise source of thermal and/or hydrodynamic origin. Note that in the absence of the driving force $\mathbf{F}$ the linearized dynamics of the displacement field in this overdamped system is purely *diffusive*: $\partial_t u \sim \nabla^2 u$, with the scale of the diffusivities set by the product of a mobility and an elastic constant. All the important physics in these equations, when the driving force is nonzero, lies in $\boldsymbol{\mu}$, the local mobility tensor, which we have allowed to depend on gradients of the local displacement field. The reason for this is as follows: The damping in the physical situations we have mentioned above arises from the interaction of the moving particles with the medium. A dynamical friction of this kind will, in general, depend on the local arrangement of particles [6,15]. Even for a perfect, undistorted lattice, the symmetry of the mobility tensor will thus reflect the symmetry of the underlying lattice. If the structure in a given region is distorted relative to the perfect lattice, the local mobility will depart from its ideal structure as well. Deviations of the structure from the perfect crystal are described by the full distortion tensor $\nabla\mathbf{u}$ [16] rather than its symmetric part, the strain, since we are not in a rotation-invariant situation. We further make the reasonable assumption that the mobility can be expanded in a power series in the distortion:

$$\boldsymbol{\mu}(\nabla u) = \boldsymbol{\mu}_0 + \mathbf{A}\nabla\mathbf{u} + O((\nabla\mathbf{u})^2), \tag{3}$$

where $\boldsymbol{\mu}_0$ is the mean macroscopic mobility of the undistorted crystal.

For a d-dimensional crystal driven steadily along the z direction, assuming isotropy in the $(d-1)$-dimensional ''$\perp$'' subspace normal to $\hat{\mathbf{z}}$, but *not* under $z \to -z$, Eqs. (2) and (3) lead directly to

$$\dot{\mathbf{u}}_\perp = \lambda_1 \partial_z \mathbf{u}_\perp + \lambda_2 \boldsymbol{\nabla}_\perp u_z + O(\boldsymbol{\nabla}\boldsymbol{\nabla} u) + O(\boldsymbol{\nabla} u \boldsymbol{\nabla} u) + \mathbf{f}_\perp , \tag{4a}$$

$$\dot{u}_z = \lambda_3 \boldsymbol{\nabla}_\perp . \mathbf{u}_\perp + \lambda_4 \partial_z u_z + O(\boldsymbol{\nabla}\boldsymbol{\nabla} u) + O(\boldsymbol{\nabla} u \boldsymbol{\nabla} u) + f_z , \tag{4b}$$

where the constant drift along z has been removed by shifting to the mean rest frame of the crystal. The terms that are manifestly most important at small wave numbers, at least within a linear description, are the linear, first-order space derivative terms. These terms arise from Eqs. (2) and (3) via the leading distortion dependence of the mobility tensor, multiplied by the driving force F. The coefficients λ_i [as well as those of the $O(\boldsymbol{\nabla} u \boldsymbol{\nabla} u)$ terms, as can be seen from Eq. (2)

and (3)] are thus proportional to F, and the corresponding terms are therefore present only in the driven state. At small enough wave numbers ($\lesssim F/K$ where F is the magnitude of the driving force density and K a typical elastic constant), these terms dominate the diffusive terms coming from the elasticity. The terms of this type in Eq. (4a) tell us that a tilt (a z derivative of a $\perp$ displacement or a $\perp$ derivative of a z displacement) leads to a lateral drift, and those in Eq. (4b) imply that the vertical settling speed depends on the compression (or dilation). Since the system is not invariant under rotations, there are no grounds for insisting that $\lambda_1=\lambda_2$ or $\lambda_3=\lambda_4$. $\mathbf{f}$ is a spatiotemporally white noise source containing the effects of thermal fluctuations as well as chaotic motion due to the hydrodynamic interaction [17,18]. The reader will note that the form of the diffusive second derivative terms and the distortion dependence of the mobility beyond linear order has been left rather general. This is because even for $d=2$, as can be seen by exhaustive listing, symmetry under $x\rightarrow -x, u_x\rightarrow -u_x$ permits, all told, in Eqs. (4a) and (4b), ten terms (this counting was wrong in [5]) bilinear in $\nabla\mathbf{u}$ and six linear second derivative terms, with as many independent coefficients. It is clearly difficult to make very useful general statements about a problem with so many phenomenological parameters so we restrict ourselves, in the next subsection, to a linearized description to lowest order in gradients. We will return to the effects of nonlinearities in later subsections.

C. Mode structure

If we retain only terms linear in the fields and work only to leading order in wave number, then the relation between frequency ω and wave vector $\mathbf{k}$ implied by Eq. (4) is

$$\omega=\frac{-1}{2}[(\lambda_1+\lambda_4)k_z\pm\sqrt{(\lambda_1-\lambda_4)^2k_z^2+4\lambda_2\lambda_3k_\perp^2}]. \quad (5)$$

The dispersion relation (5) has a wavelike character in all directions if $\lambda_2\lambda_3>0$. For $\lambda_2\lambda_3<0$, while it is still wavelike for $k_\perp=0$, it has a growing mode $\omega\propto -ik$ for $k_z\ll k_\perp$.

Linearly stable case—kinematic waves. The wavelike modes are the generalization, to the case of a moving lattice, of the kinematic waves which Lighthill and Whitham [13] discussed in the context of traffic flow and flood movements. The important difference in the present case is that the waves propagate not only along, but also transverse, to the direction of drift. Some remarks towards a more complete consideration of their dispersion relation, including the effects of nonlinearities, may be found in the context of a one-dimensional reduced model in [5].

Linearly unstable case—clumping. In the case $\lambda_2\lambda_3<0$, for wave vectors pointing outside a cone around the z axis, the system is linearly unstable, as already noted in [5]: small perturbations grow, with a growth rate which is *linear* in their wave number. Whereas the linearized treatment cannot give detailed information about the final state of the system, we expect the growing mode to appear as a clumping and tilting of the colloidal crystal, with material concentrated at the bottoms of the tilted regions. The wave vector of the inhomogeneity will be mainly normal to the sedimenting direction.

The remainder of this paper is directed towards a more detailed understanding of the statistical mechanics and dynamics of macroscopic clumping. Our studies are based mainly on the simplified one-dimensional lattice model of [5]. The construction of the lattice model is reviewed in Sec. III: its origins lie in a reduced, one-dimensional version of equations (4) which we now present.

D. A one-dimensional effective model

We saw above that the equations of motion for a moving lattice contained terms of a qualitatively new form, not present in the equations of a lattice at equilibrium. To linear order, these were the $\{\lambda_i\}$ terms in Eq. (4), which are proportional to the driving force, and of lower order in gradients than those arising from the elasticity of the crystal. The effects of the linear instability for $\lambda_2\lambda_3<0$ thus cannot be mitigated by including the diffusive terms arising from the linear elasticity. To see what final state, if any, emerges from the initial unstable growth in the case $\lambda_2\lambda_3<0$, we must go beyond a linear treatment. Even in the stable case $\lambda_2\lambda_3>0$, the combined effects of nonlinearities and noise could result in effective dispersion relations for long-wavelength modes which differ qualitatively in their form from those predicted by the linear theory. However, including nonlinearities, diffusion and noise, as we remarked in the previous subsection, introduce an enormous number of phenomenological parameters. We note instead that the important new physics of Eq. (4), namely, the wavelike (stable case) or growing modes (unstable case), arises from the coupling of the vertical and horizontal displacement fields, for excitations with wave-vector *transverse* to the direction of mean drift, while the modes with wave vectors along z play a relatively minor role. This suggests that much can be learned from a model in *one* space dimension, the x direction, corresponding to the $\perp$ direction of Eq. (4), but retaining a *two*-component displacement field $\mathbf{u}=(u_x,u_z)$. The symmetry $x\rightarrow -x, u_x\rightarrow -u_x$ then yields, to bilinear order in fields and leading orders in gradients, the equations of motion

$$\dot{u}_x=\lambda_2\partial_x u_z+\gamma_1\partial_x u_x\partial_x u_z+D_1\partial_x^2u_x+f_x, \quad (6a)$$

$$\dot{u}_z=\lambda_3\partial_x u_x+\gamma_2(\partial_x u_x)^2+\gamma_3(\partial_x u_z)^2+D_2\partial_x^2u_z+f_z, \quad (6b)$$

which have, in addition to the $\{\lambda_i\}$, three nonlinear coupling parameters $\{\gamma_i\}$ (also proportional to the driving force F), two diffusivities $\{D_i\}$, and Gaussian spatiotemporally white noise sources f_i, $i=x,z$, with zero mean, and variances N_x, N_z:

$$\langle f_i(0,0)f_j(x,t)\rangle=2N_i\delta_{ij}\delta(x)\,\delta(t). \quad (7)$$

If $\{\gamma_i\}$, $\{D_i\}$, and $\{f_i\}$ are set to zero, we recover the continuum limit of the equation derived by Crowley [6] for the dynamics of the small transverse and longitudinal displacements of a collection of hard spheres of radius a, prepared initially in a horizontal, one-dimensional periodic array with spacing d, settling vertically in a highly viscous fluid, with the hydrodynamic interaction cut off at the nearest neighbor scale. The correspondence is $\lambda_2=-\lambda_3=-(3/4)a/d$, in units of the Stokes settling speed of an isolated sphere. Crowley's

calculation can be extended beyond linear order to give $\{\gamma_i\}$, but the elastic forces and the thermal fluctuations that give the D_i's and f_i's are absent in his model. The diffusion and nonlinear terms in Eq. (6) are identical in structure to those in the Ertaş-Kardar (EK) models for the fluctuations of drifting lines [19,20], with u_x, u_z replaced by their variables $h_\perp, h_{||}$ in [19] or $R_\perp, R_{||}$ in [20]. The EK models, however, as a result of a larger symmetry [*independently* under (i) $x \to -x$ and (ii) $R_\perp \to -R_\perp$ or $h_\perp \to -h_\perp$] lack the linear first spatial derivative terms (the λ_i terms) of Eq. (6). Such linear terms can, however, be induced through the nonlinear terms, in [19,20] by constraining the ends of the line (polymer) to be at fixed mean separation normal to the drift direction, so that $\langle \partial R_\perp / \partial x \rangle \neq 0$. The related coupled-interface model of Barabási [21] has an $x \to -x$ symmetry and thus also lacks the λ_i terms of Eq. (6). These models are thus not relevant to the case of greatest interest to us here, namely, the unstable case $\lambda_2 \lambda_3 < 0$ of Eq. (6).

In the unstable case, within a linear treatment, the concentration $\partial_x u_x$ and the tilt $\partial_x u_z$ grow without bound [22]. Physically, since real colloidal crystals are made of impenetrable particles, and since the elasticity of the lattice will not tolerate arbitrarily large shear strains, the description implicit in Eq. (6) of small distortions about a perfect lattice must break down in conditions of unstable growth. It is best, therefore, to work from the outset with naturally bounded variables for the concentration and tilt. To this end, we first pass to a description in terms of the concentration fluctuation field

$$\sigma(x,t) = \frac{\partial u_x}{\partial x} \tag{8}$$

and the tilt field

$$\tau(x,t) = \frac{\partial u_z}{\partial x}. \tag{9}$$

Then Eq. (6) can be rewritten in the ''conservation-law'' form

$$\dot{\sigma} = \lambda_2 \partial_x \tau + \gamma_1 \partial_x(\sigma\tau) + D_1 \partial_x^2 \sigma + \partial_x f_x, \tag{10a}$$

$$\dot{\tau} = \lambda_3 \partial_x \sigma + \gamma_2 \partial_x(\sigma^2) + \gamma_3 \partial_x(\tau^2) + D_2 \partial_x^2 \tau + \partial_x f_z. \tag{10b}$$

As stated above, σ and τ should be bounded; what matters on large length scales is only whether the local concentration is large or small compared to the mean, and whether the local tilt is ''up'' or ''down.'' Accordingly, we construct a description in the next section in which the concentration and tilt fields of Eq. (10) are replaced by Ising variables evolving under a spin-exchange dynamics designed to mimic the most important aspects of Eq. (10). A continuum model which incorporates saturation is presented in Sec. IV.

III. STRONG PHASE SEPARATION IN A LATTICE MODEL

In this section, we introduce the notion of strong phase separation in connection with the LR lattice model, which describes two coupled species of spins on a lattice, with simple evolution rules which mimic the coupled dynamics of the density and tilt fields. This coupled-spin problem is too difficult to solve for the dynamics or, indeed, for the steady state for arbitrary values of parameters. However, for the symmetric case of half filling of both species, and a special relation between coupling constants, we show (Sec. III) that the condition of detailed balance is satisfied with respect to a Hamiltonian $\mathcal{H}$ with long-ranged interactions. In turn, this allows for a characterization of the steady state of the system. In Sec. III C, we show that at zero temperature T, the system exhibits phase separation. Moreover, we calculate thermodynamic properties and show that the phase separation survives at *all* finite temperatures, which is why we call this phenomenon SPS. The occurrence of SPS is linked to the long (actually infinite) range of the interactions in $\mathcal{H}$, which results in the energy being superextensive (proportional to L^2 rather than L). We emphasize that this happens although the underlying dynamical model is entirely local, with finite, bounded rates. In Sec. III D, we show that this unusually robust phase separation sets in anomalously slowly, with domain sizes growing as the logarithm of time. The survival of SPS away from the detailed-balance limit is discussed through a kinetic interpretation in Sec. III E.

A. The LR lattice model

From the study of driven diffusive systems, it is well known that hydrodynamic behavior can be recovered from the large-distance long-time behavior of simple lattice-gas models evolving by stochastic dynamics [23]. An example of such a model is the asymmetric exclusion process, in which particles on a lattice perform biased random walks subject to the constraint of no more than one particle per site; in the limit of large separations and time, density fluctuations are described by the Burgers equation with an additional noise term. An advantage of a lattice-gas description is that nonlinearities are incorporated implicitly in the nature of the variable—for instance, a (0,1)-valued occupation variable incorporates the effects of exclusion.

Are there simple lattice-gas models which capture the essential features of coupled density-tilt dynamics of the type discussed in the previous section? Any such lattice model must, of course, involve two sets of variables—say $\{\sigma_i\}$ and $\{\tau_i\}$—which are discrete versions of density and tilt fields and which evolve by rules which mimic the physics of sedimenting lattices. There are two crucial features of the σ-τ dynamics of Eq. (10): first, that both σ and τ fields are conserved so that their time derivatives involve the divergences of currents; and second that the local field which guides the σ current has a term which is proportional to τ, and *vice versa*. Accordingly, we define [5] a lattice model which incorporates just these effects. Consider a one-dimensional lattice made of two interpenetrating sublattices S ($i = 1,2,3,\ldots,N$) and T ($i = 1/2,3/2,\ldots,N-1/2$). Place Ising variables $\sigma_i = \pm 1$ at every site of S, and $\tau_{i+1/2} = \pm 1$ on every site of T. We take $\sigma_i = 1$ if there is a particle at site i, and $\sigma_i = -1$ if there is no particle, while $\tau_i = 1$ or -1 denotes the two possible values of the local tilt. The dynamics involves exchange of adjacent spins σ_i and σ_{i+1} at a rate which depends on the intervening spin $\tau_{i+1/2}$, while the rate of τ-spin exchanges depends on the intervening σ spin, i.e.,

we have Kawasaki [24] dynamics, with hopping rates which depend on the local value of the other species. The probability $P(C)$ that the system is in a configuration $C \equiv (\{\sigma_i\},\{\tau_{i-1/2}\})$ evolves through the master equation

$$\frac{dP(C)}{dt} = \sum_{\langle n,n+1\rangle} W(C_{n,n+1}\to C)P(C_{n,n+1}) - W(C\to C_{n,n+1})P(C). \tag{11}$$

Here $\langle n,n+1\rangle$ on the right hand side (with $n = 1/2,1,3/2,2,\ldots$) labels transitions which involve pairwise interchanges of neighboring σ's ($\sigma_i \leftrightarrow \sigma_{i+1}$) and τ's ($\tau_{i-1/2}\leftrightarrow\tau_{i+1/2}$), and configuration $C_{n,n+1}$ differs from C only through the interchange of spins on site n and $n+1$. The most general such model would involve the eight distinct transition rates listed in Eq. (1). For a left-right symmetric system, we have $D=D'$; $a=a'$; $\mathrm{sgn}(b)=\mathrm{sgn}(b')$: this defines the LR model [5].

In the interest of defining a *minimal version* of the LR model, we also impose the further restrictions $E=E'$, $b=b'$. The rates of the minimal model may be written compactly as

$$W(\sigma_i\leftrightarrow\sigma_{i+1};\tau_{i+1/2}) = D - \frac{a\tau_{i+1/2}}{2}(\sigma_i-\sigma_{i+1}),$$

$$W(\tau_{i-1/2}\leftrightarrow\tau_{i+1/2};\sigma_i) = E + \frac{b\sigma_i}{2}(\tau_{i-1/2}-\tau_{i+1/2}). \tag{12}$$

The evolution rules can be stated as follows: If a is positive, a particle tends to move downhill, and a hole uphill. If b is positive, a local peak ($\wedge$) tends to transform into a valley ($\vee$) if a particle resides on it, while local valleys tend to become peaks in the presence of holes. Changing the signs of a and b reverses these tendencies. As a result, the nature of the steady state is sensitive to the sign of $\alpha \equiv ab$. As we will see below, if α is positive, the exchanges of σ and τ spins in Eq. (12) act in concert to promote segregation of both species of spins, ultimately resulting in a phase-separated state. This is the unstable case of the LR model—the case of primary interest in this paper. By contrast if α is negative, ''easy'' σ and τ moves produce opposing tendencies, and hence result in a fluctuating but on-average spatially homogeneous state—the stable case of the LR model. The calculations of Crowley [6] for settling arrays of hydrodynamically interacting spheres and the discussion in [5] make it clear that for sedimenting colloidal crystals it is the ''unstable'' case that applies.

The other important parameters in the model are the magnetizations $M_\sigma \equiv \Sigma_i \sigma_i/N$, $M_\tau = \Sigma_i \tau_{i+1/2}/N$, both of which are conserved by the dynamics.

B. Symmetric case: Hamiltonian and detailed balance

We now consider the *symmetric case* of the LR model, which is defined by the vanishing of the magnetizations

$$M_\sigma = M_\tau = 0, \tag{13}$$

and the following relationship between coupling constants in Eq. (12):

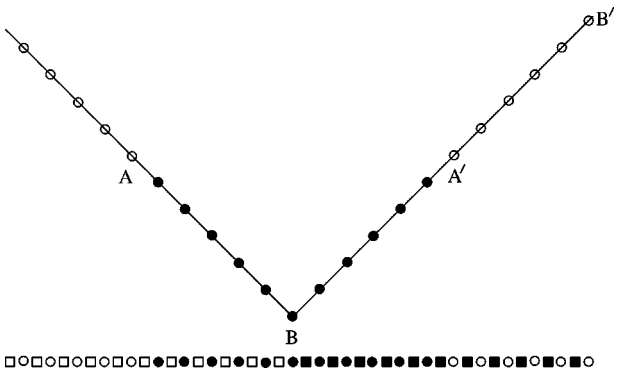

FIG. 1. The phase-separated state of the 1D lattice model at $T=0$ is shown. σ and τ variables are shown as circles and squares, respectively, with $\sigma,\tau=+1$ (-1) shown filled (empty). The configuration of the corresponding height model is also shown. Interfaces between $\sigma=+1$ and -1 are located at A and A', and those between $\tau=+1$ and -1 are at B and B'.

$$\frac{b}{E} = \frac{a}{D}. \tag{14}$$

Since $E,D>0$, it is clear that Eq. (14) is a special case of the *unstable* LR model. We show below that when conditions (13) and (14) are met, it is possible to find a Hamiltonian $\mathcal{H}$ such that the condition of detailed balance is satisfied with invariant measure $\exp(-\beta\mathcal{H})$.

Since the motion of σ particles is determined by the local tilt τ, we may think of the σ particles as moving in a potential landscape provided by the τ's (Fig. 1). With this in mind, we define the height at site k by

$$h_k\{\tau\} = \sum_{j=1}^{k} \tau_{j-1/2}. \tag{15}$$

With periodic boundary conditions ($\sigma_{N+i}=\sigma_i$; $\tau_{N+i-1/2} = \tau_{i-1/2}$), the zero-net-tilt condition $M_\tau = 0$ implies $h_{N+k} = h_k$. We associate a potential energy proportional to $h_k\sigma_k$ with site k, and write the Hamiltonian

$$\mathcal{H} = \epsilon \sum_{k=1}^{N} h_k\{\tau\}\sigma_k \tag{16}$$

to describe the total energy of the σ particles in the landscape derived from the τ particles.

In view of the symmetric role played by σ's and τ's in the symmetric model, we may equally ask for the potential energy of τ particles in the landscape provided by the σ particles. The corresponding Hamiltonian is then

$$\mathcal{G} = \epsilon \sum_{k=1}^{N} g_{k-1/2}\{\sigma\}\tau_{k-1/2} \tag{17}$$

where the height g is given by

$$g_{k+1/2}\{\sigma\} = \sum_{j=1}^{k} \sigma_j. \tag{18}$$

When the heights h_k and $g_{k-1/2}$ are written out in terms of τ_j's and σ_j's, respectively, the Hamiltonians $\mathcal{H}$ and $\mathcal{G}$ are seen to involve very nonlocal couplings:

$$\mathcal{H}=\epsilon\sum_{k=1}^{N}\sum_{j=1}^{k}\tau_{j-1/2}\sigma_k, \tag{19}$$

$$\mathcal{G}=\epsilon\sum_{k=2}^{N}\sum_{j=1}^{k-1}\sigma_j\tau_{k-1/2}. \tag{20}$$

We observe that

$$\mathcal{H}+\mathcal{G}=\epsilon M_\sigma M_\tau \tag{21}$$

and, since each of M_σ and M_τ vanishes in the symmetric case owing to the zero-tilt condition, we have $\mathcal{H}=-\mathcal{G}$. Thus the Hamiltonians corresponding to the two pictures, i.e., σ particles in a τ landscape or *vice versa* are completely equivalent. We will mostly use $\mathcal{H}$ for further work.

We now show that the steady state of the symmetric model defined by Eqs. (13) and (14) satisfies the condition of detailed balance and that the stationary measure is given by $e^{-\beta\mathcal{H}}$ where β is the inverse temperature T^{-1}, with $\beta\epsilon$ given by Eq. (27) below. To this end, let us ask for the changes in energy $\Delta E(\sigma_i\leftrightarrow\sigma_{i+1})$ of $\mathcal{H}$ when spins σ_i and σ_{i+1} are interchanged, and $\Delta E(\tau_{i-1/2}\leftrightarrow\tau_{i+1/2})$ in $\mathcal{H}$ when spins $\tau_{i-1/2}$ and $\tau_{i+1/2}$ are interchanged. For $i\neq N$, it is straightforward to see that

$$\Delta E(\sigma_i\leftrightarrow\sigma_{i+1})=\epsilon\tau_{i+1/2}(\sigma_i-\sigma_{i+1}), \tag{22}$$

$$\Delta E(\tau_{i-1}\leftrightarrow\tau_{i+1})=\epsilon\sigma_i(\tau_{i-1/2}-\tau_{i+1/2}). \tag{23}$$

In fact, Eqs. (22) and (23) are valid for $i=N$ as well, as can be verified on recalling that $\sigma_{N+1}=\sigma_1$, $\tau_{N+1/2}=\tau_{1/2}$ and using the zero-tilt conditions $M_\sigma=M_\tau=0$ while computing energy changes.

Consider the configuration $C_{\sigma_i,\sigma_{i+1}}$ obtained from a configuration C on exchanging two neighboring σ spins—an elementary kinetic move in the model. The condition of detailed balance is then

$$\frac{W(C\rightarrow C_{\sigma_i,\sigma_{i+1}})}{W(C_{\sigma_i,\sigma_{i+1}}\rightarrow C)}=\frac{\mu_{SS}(C_{\sigma_i,\sigma_{i+1}})}{\mu_{SS}(C)}, \tag{24}$$

where $\mu_{SS}(C)$ is the steady-state measure for configuration C. To verify that

$$\mu_{SS}(C)=e^{-\beta\mathcal{H}(C)}, \tag{25}$$

we use Eqs. (12) and (22) to obtain

$$\frac{D-aX_i}{D+aX_i}=e^{-2\beta\epsilon X_i} \tag{26}$$

where we have defined $X_i\equiv 1/2\,\tau_{i+1/2}(\sigma_i-\sigma_{i+1})$. Noting that $X_i=\pm 1$, we see that Eq. (26) is satisfied provided

$$\beta\epsilon=\frac{1}{2}\ln\left(\frac{D+a}{D-a}\right). \tag{27}$$

In order for the measure to be valid under interchanges of adjacent τ's ($\tau_{i-1/2}\leftrightarrow\tau_{i+1/2}$), similar reasoning leads to the condition

$$\beta\epsilon=\frac{1}{2}\ln\left(\frac{E+b}{E-b}\right). \tag{28}$$

In the symmetric case of the LR model, Eq. (14) holds, and so Eqs. (27) and (28) are consistent. Thus the condition of detailed balance holds with the equilibrium measure (25).

It is appropriate to recall that the three-species model of Evans *et al.* [7] also obeys the condition of detailed balance in the symmetric case. There too the Hamiltonian has infinite ranged interactions, but does not have as transparent an interpretation as Eq. (16).

C. Symmetric case: Thermodynamic properties and strong phase separation

Since the condition of detailed balance holds in the symmetric case of the minimal LR model, the steady state corresponds to the thermal equilibrium state with Hamiltonian $\mathcal{H}$. The thermodynamic properties of the system can be found, in principle, using equilibrium statistical mechanics. A calculation can be carried out in the grand canonical ensemble in the limit $N\rightarrow\infty$. The resulting state exhibits strong phase separation.

The Hamiltonian $\mathcal{H}$ [Eq. (16)] describes spins σ_k in a site-dependent magnetic field ϵh_k, which is itself a dynamical variable. Equivalently, in the lattice-gas description [associating an occupation variable $n_k=1/2(1+\sigma_k)$], it describes particles with a hard core constraint in a potential well of depth ϵh_k. The ground state of $\mathcal{H}$ is obtained by arranging the τ spins (which determine the heights h_k) so as to form as deep a potential well as possible, and then arranging the σ particles at the bottom of the well (Fig. 1). A spin configuration which corresponds to this choice is

$$\begin{aligned}\tau_{k-1/2}&=-1 \quad \text{for} \quad k=1,\ldots,N/2\\ &=1 \quad \text{for} \quad k=\frac{N}{2}+1,\ldots,N,\\ \sigma_k&=1 \quad \text{for} \quad k=N/4,\ldots,3N/4\\ &=-1 \quad \text{for} \quad k=1,\ldots,N/4-1\\ &\text{and} \quad k=3N/4+1,\ldots,N.\end{aligned} \tag{29}$$

Each spin species exhibits complete phase separation in this ground state. The ground state energy is straightforward to compute, and we find

$$E_G\simeq-\frac{\epsilon N^2}{8}. \tag{30}$$

Notice the quadratic dependence of E_G on N, which is an outcome of the infinite-ranged interactions in $\mathcal{H}$ [Eq. (30)]. As explained below, this unusual superextensive behavior of the energy is ultimately the feature responsible for the phenomenon of strong phase separation, namely the continued existence and stability of the phase-separated state at all finite temperatures.

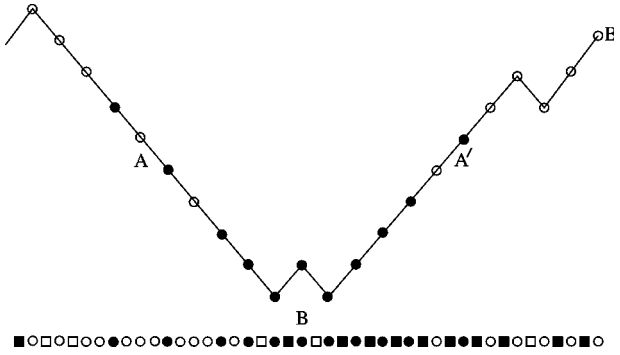

FIG. 2. Typical configurations of the unstable LR model at $T \neq 0$ are shown. The meaning of the symbols is as in Fig. 1. Phase separation persists, but there are particle-hole excitations of both species near the corresponding interfaces.

At $T=0$, phase separation is complete and there is a sharp boundary between regions of positive and negative spins of each species. Let A and A' be the locations of the $T=0$ interface between regions with $\sigma=1$ and $\sigma=-1$, and let B and B' be the locations of interfaces separating regions with $\tau=1$ and $\tau=-1$ (Fig. 1). The effect of raising the temperature to a finite value T is to smear out the interfacial zones around A, A', B, and B' (Fig. 2). To address this quantitatively, let us turn to the evaluation of thermodynamic properties.

The calculation can be carried out most easily in a grand ensemble in which the total magnetizations M_σ and M_τ are not held fixed. The corresponding grand partition function is

$$Z \equiv \sum_{\{\sigma\},\{\tau\}} e^{-\beta(\mathcal{H}-E_G)} = \sum_{\{\sigma\}\{\tau\}} e^{-\beta\epsilon \sum_k h_k(\sigma_k-\sigma_k^0)}, \quad (31)$$

where σ_k^0 denotes the value of σ_k in the ground state. The key observation that allows the calculation to be performed is that near the σ interfaces A and A', the field $h_k(\{\tau\})$ is essentially fixed at its $T=0$ value h_k^0; deviations are of order $\exp[-\beta\epsilon N/4]$ as explained below, and so are utterly negligible in the thermodynamic limit. Likewise, in the vicinity of the τ interfaces B and B', the σ spins are frozen to their $T=0$ values, and so $g_k(\{\sigma\})=g_k^0$. To proceed, let us divide the system into four equal parts R_A, R_B, $R_{A'}$, $R_{B'}$, where region R_A consists of the $N/4$ spins of each of the two species centered around A. Other regions are defined similarly, centered around B, A', and B'. Evidently, with negligible error we may set $h_k(\{\tau\})=h_k^0$ in regions R_A and $R_{A'}$, and set $g_k(\{\sigma\})=g_k^0$ in regions R_B and $R_{B'}$. The partition function Z can then be written as the product of four terms Z_A, Z_B, $Z_{A'}$, $Z_{B'}$, where, for instance,

$$Z_A = \sum_{\{\sigma\}} e^{-\beta\epsilon \sum_{k\in R_A} h_k^0(\sigma_k-\sigma_k^0)}, \quad (32)$$

$$Z_B = \sum_{\{\tau\}} e^{-\beta\epsilon \sum_{k\in R_B} g_{k+1/2}^0(\tau_{k+1/2}-\tau_{k+1/2}^0)}. \quad (33)$$

Each of these factorizes into single-site partition functions, and can be evaluated straightforwardly. Recalling that h_k^0 varies linearly with k near the $T=0$ interface location k_A, we find

$$Z_A = \prod_{k\in R_A} (1+e^{-2\beta\epsilon|k-k_A|}). \quad (34)$$

In the thermodynamic limit, we obtain

$$Z_A = K(e^{-2\beta\epsilon}), \quad (35)$$

where $K(y)\equiv\Pi_{k=-\infty}^{\infty}(1+y^{|k|})$ is a generating function that arises in the theory of partitions [25]. Evidently, each of Z_B, $Z_{A'}$, and $Z_{B'}$ equals the same quantity as well, so that $Z=\{K[\exp(-2\beta\epsilon)]\}^4$.

It is worth pausing to comment on the unusual size dependences of various quantities. The ground state energy E_G is proportional to N^2, a superextensive dependence. This has its origin in the infinite-ranged interactions in $\mathcal{H}$. Further, with energies measured from the ground state value, the partition function approaches an N-independent limit. Thus the total change in free energy and entropy away from $T=0$ remain finite in the thermodynamic limit, i.e., they are not extensive. This reflects the fact that the only effect of raising the temperature is to broaden the interfacial region between phases, which essentially affects only a finite number of sites.

In fact, an explicit calculation of the broadened interfacial profile can be carried out in the grand ensemble. For instance, near A we have

$$\langle\sigma_k\rangle = \tanh\beta\epsilon h_k^0, \quad (36)$$

where $h_k=(k-k_A)$. We see that $\langle\sigma_k\rangle$ deviates substantially from 1 only in a region where $|\beta\epsilon h_k| \lesssim 1$, or

$$|k-k_A| < T/\epsilon. \quad (37)$$

For sites k such that $|k-k_A| \gg T/\epsilon$, the deviation from ± 1 is $\approx 2\exp(-2\beta\epsilon|k-k_A|)$ which vanishes rapidly. We see that the primary effect of temperature is to smear out the interfaces. The formation of ''droplets'' far from the interfaces is prohibitively costly in energy, and hence the probability dies down exponentially. Recalling that the separation of the two $\sigma=1\to-1$ interfaces is $N/2$, in the thermodynamic limit $N\to\infty$, we see that only a vanishing fraction of spins (those close to the interfaces) deviate from values arbitrarily close to 1 and -1. In this sense, phase separation remains complete and cannot be effaced at any finite temperature T, i.e., we have strong phase separation.

These results obtained in the grand ensemble provide a qualitative, if not quantitative, guide to the thermodynamic properties of the system in which M_σ and M_τ are held fixed. The customary equivalence between ensembles is not obviously valid any longer, as particle-hole excitations are essentially confined to a finite region of width proportional to T, which does not increase as $N\to\infty$. Thus the difference between observables calculated in the two ensembles is expected to remain of order unity, and not die out in the $N\to\infty$ limit [26]. Interestingly, the calculation of the partition

function, though not the profile, has been carried out for the three-species model within a constant-species-number ensemble [7].

The stability of the strongly phase-separated state can also be understood in terms of kinetics. In the ground state arrangement of Fig. 1, each σ spin finds itself in a uniform field produced by the τ spins. Consider moving a spin over a macroscopic distance—say a $\sigma=+1$ spin from A' to A, via B. The movement from A' to B may be viewed as an activation process as the spin in question has to overcome a potential barrier of magnitude $\epsilon N/4$ to reach B; beyond that, in the region BA, the motion is ballistic as the τ-induced field helps it along. The rate-limiting step is thus the $A' \rightarrow B$ activation. At temperature T, the relevant time scale is of the order of $t_{CB} \sim \exp(\epsilon N/4T)$ which diverges rapidly as $N \rightarrow \infty$. Thus, in the thermodynamic limit, a rearrangement of the SPS state is not possible; the only effect of the temperature-assisted motion is to move a few $\sigma=1$ spins near the interface into the $\sigma=-1$ rich region and vice versa, but such penetration does not proceed far in view of the restoring fields. Defining the penetration depth Δk as that over which the activation time falls by a factor of $1/e$, we estimate $\Delta k = T/\epsilon$, in agreement with Eq. (37) which was based on the spatial decay of the interfacial profile.

D. Coarsening

Now imagine that Fig. 1 represented *half* the system, and that the other half was identical in structure. This would amount to a system that had phase-separated into *four* macroscopic domains, each of size $N/8$. For this state to proceed towards full phase separation, the two + domains, each at the bottom of a valley, must merge. The rate-limiting step can again be taken to be the movement of a + from the edge of an all $--$ region to the top of a hill, i.e., a distance $N/8$. Once this comes to pass, the two domains of length $N/4$ will rapidly merge to give one domain of length $N/2$. The time for this, which is the time for complete phase separation for a system of size N, can be seen from the argument in Sec. III C to scale as $\exp(\epsilon N/8T)$. This tells us that the characteristic domain size grows logarithmically in time, as stated in Sec. I.

The time required for the reverse process (from a two-domain to a four-domain state) scales as $\sim \exp[(N/4T)]$, which is overwhelmingly larger than the $4 \rightarrow 2$ coarsening time. This is true at all scales, and the transition from a $2n$-domain state to one with n domains is much more rapid than the reverse. Thus the transition from a statistically homogeneously mixed state to the equilibrium phase-separated state is irreversible, even though it occurs slowly.

The coarsening process was studied [7] both numerically in the 3-species model and within a mean-field approximation for a related ''toy'' model. The typical domain size was found to grow logarithmically in time. The arguments given above are consistent with this.

E. Nonsymmetric case

We now address the nature of the steady state for arbitrary values of M_σ and M_τ. Away from $M_\sigma = M_\tau = 0$ the problem is no longer described in terms of the equilibrium state of a long-ranged Hamiltonian; nevertheless we will argue below that the system continues to exhibit strong phase separation.

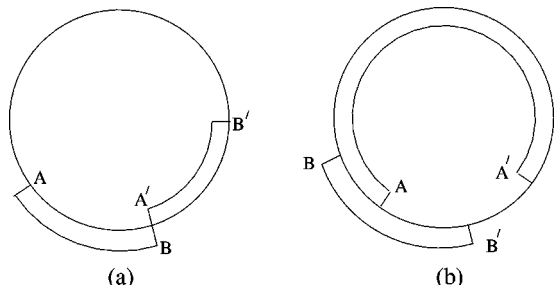

FIG. 3. Typical configurations are depicted away from the symmetric case in the limit of vanishing noise. (a) If the fraction of $\sigma=+1$ and $\tau=+1$ spins is low enough, interfaces A' and B coincide. (b) If the fraction of $\sigma=+1$ spins is high enough, the interface B' lies halfway between A and A'.

It is useful to define x and y as the density of up spins of the σ and τ types. We have $x = 1/2(1+M_\sigma/N)$ and $y = 1/2(1+M_\tau/N)$. If x and y are small enough that $2x+y<1$, the steady state is of the type shown in Fig. 3(a), with each of the σ and τ species showing phase separation, but with basically no spatial overlap of the $\sigma=1$ and $\tau=1$ regions. A useful way to characterize this state is through the sequence of interfaces, viz. $A \cdots B' \cdots BA' \cdots$, where $\cdots$ denotes a macroscopic stretch of the system. Here $A(B)$ separates an up-spin region of $\sigma(\tau)$ spins on the right, from the corresponding down-spin regions, while A' and B' separate the opposite regions. Trial states of the type $A \cdots B' \cdots B \cdots A' \cdots$ are seen to approach the nonoverlapping state on a time scale of order $t^\star$ where $\ln t^\star$ is less than but of the order of the smaller of $\epsilon Nx/T$ and $\epsilon Ny/T$. Once the nonoverlapping steady state has been reached, σ and τ spins can still be cycled around by activation processes across $A'A$ and $A'A$, respectively (Fig. 3), but such cycling around does not change the character of the state.

Now consider increasing y, keeping x fixed. The number of spins in the stretch between B' and A is $N(1-x-y)$, and once this drops below Nx, the predominant activation process occurs over this stretch. Thus the no-overlap state of Fig. 3(a) is unstable towards a state of the type shown in Fig. 3(b), once $2x+y$ exceeds unity. In this steady state, activation processes in a finite system lead to small currents of σ and τ spins, of magnitude

$$J_\sigma = a_1 \exp(-\epsilon \ell_{AB'}/T) - a_2 \exp(-\epsilon \ell_{A'B'}/T), \quad (38)$$

$$J_\tau = a_3 \exp(-\epsilon \ell_{BA}/T) - a_4 \exp(-\epsilon \ell_{B'A}/T), \quad (39)$$

where a_1, a_2, a_3, a_4 are prefactors of order unity and $\ell_{AB'}$ is the separation of interfaces A and B', and other ℓ's are defined similarly. Since the difference $\ell_{A'B'} - \ell_{AB'}$ is positive and grows proportionally to N, we may drop the second term on the right of Eq. (38). In steady state we must have $J_\sigma = J_\tau$, which then leads to $(a_1+a_4)\exp(-\epsilon \ell_{AB'}/T) \simeq a_3 \exp(-\epsilon \ell_{BA}/T)$ or

$$\ell_{AB'} = \ell_{BA} + \text{terms of order unity}. \quad (40)$$

Thus, A is very close to the halfway position in stretch BB'. The overlapping stretch BA' is a fraction $\delta = 1/2(2x+y$

-1) of the whole length. On setting $x=y=1/2$, we recover $\delta=1/4$ in agreement with the results of the equilibrium analysis of Sec. III C.

Analogously, keeping y fixed and increasing x we conclude that for $2y+x>1$, the steady state is $A\cdots B\cdots A'\cdots B'\cdots$ with B' between A' and A and an overlapping stretch of length $1/2N(2y+x-1)$. Finally, under $x\to(1-x)$, $y\to(1-y)$ we arrive at the condition for overlap of negative spins.

In short, strong phase separation persists even away from the symmetric point of the LR model. In general, two types of steady states, both phase separated, are possible as depicted in Fig. 3. In the overlapping case, there is generally a current in a finite system, but this vanishes exponentially with system size. While we have explored the effects of deviating from the symmetric case by moving away from the half-filling condition Eq. (13), without altering the condition (14) on the rates, another way to make the system nonsymmetric is to violate the latter condition. We have not explored this in detail, but expect that the phenomenon of SPS will persist in this case also so long as $ab>0$.

IV. DETAILED BALANCE AND STRONG PHASE SEPARATION IN THE CONTINUUM LIMIT

The continuum model of Sec. II, in the case $\lambda_2\lambda_3<0$ in Eq. (6), is linearly unstable. One way to deal with this instability is to resort, as we have done above, to a lattice model in which the variables are naturally bounded. An alternative way is to ask what nonlinear terms added to Eq. (6) for $\lambda_2\lambda_3<0$ would arrest the unstable growth [22]. To do this, we work in the detailed-balance limit of the lattice model, start with the Hamiltonian (16), and construct the corresponding continuum Ginzburg-Landau free-energy functional. We shall see below that this functional will give rise to dynamical equations with the same linear instability as in Eq. (6) with $\lambda_2\lambda_3<0$, but containing nonlinearities which prevent unbounded growth.

The derivation is straightforward, as the condition of detailed balance allows us to proceed as in any equilibrium statistical mechanics problem. The Ginzburg-Landau free-energy functional $F[\sigma,\tau]$ for our system, i.e., the effective Hamiltonian for a description in terms of the coarse-grained fields $\{\sigma(x),\tau(x)\}$ of Sec. II D, may be written as $U-TS$, where U is the energy (16) in the continuum limit, T the temperature, and S the entropy obtained by summing over all microscopic configurations $\{\sigma_i,\tau_i\}$ subject to a fixed coarse-grained configuration $\{\sigma(x),\tau(x)\}$. Since σ_i and τ_i are Ising variables, S can be found from a standard Bragg-Williams construction. Thus,

$$F[\sigma,\tau]=\epsilon\int_0^L dx\int_0^x dx'\,\sigma(x)\tau(x') + T\int_0^L dx\sum_{m=\tau(x),\sigma(x)}\left[\frac{1+m}{2}\ln\frac{1+m}{2}+\frac{1-m}{2}\ln\frac{1-m}{2}\right], \tag{41}$$

where x is measured in units of the lattice spacing and is hence dimensionless. The partition function is then $\int[d\sigma][d\tau]\exp(-F/T)$.

Then the usual, purely dissipative, *conserving* time-dependent Ginzburg Landau equations of motion generated by Eq. (41), i.e.,

$$\partial_t\sigma=\Lambda_\sigma\partial_x^2\frac{\delta F}{\delta\sigma}+\eta_\sigma \tag{42}$$

and likewise for τ, turn out to be precisely

$$\partial_t\sigma=\Lambda_\sigma(T\partial_x^2\tanh^{-1}\sigma+\epsilon\partial_x\tau)+\eta_\sigma;$$
$$\partial_t\tau=\Lambda_\tau(T\partial_x^2\tanh^{-1}\tau-\epsilon\partial_x\sigma)+\eta_\tau. \tag{43}$$

Here $\Lambda_\sigma,\Lambda_\tau$ are mobilities and η_σ,η_τ are noise sources with variances proportional to the corresponding mobilities. It is evident that Eqs. (43) and (10) are identical in the linearized limit, if we make the identification $\lambda_2=\Lambda_\sigma\epsilon$, $\lambda_3=-\Lambda_\tau\epsilon$. This corresponds to the linearly unstable limit of Eq. (6), in consonance with the fact that the detailed balance limit of the lattice model was derived in precisely that case.

We should thus be able to gain some insight into SPS by looking at the steady states of Eq. (43). The simplest of these are the zero current states, which satisfy

$$\partial_x P-\tanh Q=0,$$
$$\partial_x Q+\tanh P=0, \tag{44}$$

where

$$Q\equiv\tanh^{-1}\phi,\quad P\equiv\tanh^{-1}\psi. \tag{45}$$

The *spatial* development of P and Q with respect to x is like a Hamiltonian dynamics, conserving the ''energy''

$$E(P,Q)=\ln(\cosh P\cosh Q). \tag{46}$$

This leads to closed orbits in the $P-Q$ or $\psi-\phi$ plane, i.e., regions of large ψ and small ϕ followed by the opposite. These are spatially multidomain states which will not evolve further in the absence of noise.

V. SUMMARY AND DISCUSSION

A. Summary

In summary, we have constructed continuum and lattice models to describe the physics of steadily sedimenting colloidal crystals or, more generally, of a crystal driven through a dissipative medium. The models display two broadly distinct types of behavior, termed ''stable'' and ''unstable,'' depending on the sign of a parameter. We have concentrated on the unstable case and shown, through a mapping to a one-dimensional lattice model, that it *always* displays phase separation, a phenomenon which we call strong phase separation. This phase separation and the fact that it persists at all temperatures can be understood, in general, in terms of barriers to remixing which grow with system size. The barriers are erected by the system in the course of its evolution, and result in domain sizes growing as the logarithm of the time. In a particular limit, the detailed balance condition holds, allowing us to write the steady state distribution in the equilibrium form $\exp(-\beta\mathcal{H})$, and to calculate density profiles exactly. Here $\mathcal{H}$ involves long-ranged interactions even though the model has strictly local dynamics. This long-

ranged character of interactions in $\mathcal{H}$ is responsible for the phase separation in this one-dimensional system, and the fact that it persists at all temperatures.

B. Experimental tests

Finally, let us turn to the possibility of testing our results in experiments. We have demonstrated strong phase separation in a *one-dimensional* model system. It seems highly likely, therefore, that the same phenomenon will take place in the experimental systems which inspired our model, namely, steadily sedimenting crystalline suspensions in, for example, the two-dimensional geometry described in Sec. I. A good candidate system is a charge-stabilized crystalline array of polystyrene spheres with radius in the micron range. The lattice spacing of the crystal should be neither so large that hydrodynamic effects (proportional to the ratio of particle size to interparticle spacing) are negligible, nor so small that the flow is choked. This will ensure that appreciable hydrodynamic flow takes place between the spheres, giving rise to the strain-dependent mobilities [6] that are used in Eq. (4). If the system parameters are as in [27], the Reynolds number will be negligible, as required by our neglect of inertia, and the Peclet number large. Note that our model equations (4) were formulated to describe the nature of distortions about a single crystalline domain. In particular, the instability towards clumping takes place only on large enough length scales. In a polycrystalline sample, if the size of the crystallites is too small, terms from the elastic energy in Eq. (4) could dominate instead. In addition, it is important that the sedimentation be steady, a requirement best met by working in the fluidized-bed geometry in which the particles constituting the crystal are on average at rest in the laboratory frame of reference, and the fluid flows vertically upwards past them. We would recommend starting with the suspension in the fully sedimented state, and then switching on the upward flow. Observations in [28] suggest that strongly charge-stabilized crystalline suspensions appear stable whereas suspensions in a fluid state display the Crowley instability in a visible manner. We suspect that the instability is present even in the crystalline suspensions, but is masked either by finite crystallite size or by the logarithmically slow coarsening of domains. We predict that careful measurements of the time evolution of the static structure factor, using particle-imaging or ultrasmall-angle scattering techniques, should reveal a weak large-scale modulation of the particle concentration, with characteristic wave vector normal to the sedimentation direction and decreasing logarithmically in time.

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