Are Steadily Moving Crystals Unstable?

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We study the dynamics of small fluctuations about the uniform state of a crystal moving through a dissipative medium, e.g., a sedimenting colloidal crystal or a moving flux lattice, using a set of continuum equations for the displacement fields, and a one-dimensional driven lattice-gas model for the coupled concentration and tilt fields. For the colloidal crystal we predict a continuous nonequilibrium phase transition to a clumped state above a critical Péclet number. [S0031-9007(97)03739-3]

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What is the response of a crystalline lattice to a small, long-wavelength disturbance? Elastic theory [1] and its extension to time-dependent phenomena [2] provide a complete answer to this question for a system *in thermal equilibrium.* In this Letter we ask and answer the same question for a lattice being *driven through a dissipative medium* by a constant external force. This important nonequilibrium steady state arises, for example, in the steady sedimentation [3,4] of a colloidal crystal [5] and in the motion of a depinned flux lattice in a current-carrying superconductor. Using symmetry arguments we construct continuum and lattice-gas models for the dynamics of small distortions about the uniform state of such a steadily moving lattice. The effects that we discuss arise from the dependence of the mobility of a given region on the local strain of the crystal. Our most striking result is that the dominant linear response at long wavelengths is proportional to the *driving speed* of the lattice, not to its elastic constants, and that this response can lead to a nonequilibrium phase transition.

Before presenting our results in more detail let us recall an important early study. Crowley [6] carried out experiments (on steel balls dropped gently into turpentine oil) and theory (calculating the hydrodynamic interactions between the spheres) to show that a regular horizontal array of sedimenting spheres was *linearly unstable* to clumping and buckling. Elastic forces, Brownian motion, and nonlinearities, all of which can resist this instability, were not considered in [6]. Since experiments on model colloidal systems are most conveniently performed by varying interaction strength rather than temperature [5], the appropriate Péclet number for this problem is the ratio $\mathrm{Pe} = \Delta\rho a g/G$ of gravitational to *elastic* rather than Brownian stresses. Here $\Delta\rho$ is the difference between the mass densities of particles and solvent, a the particle radius, g the acceleration due to gravity, and G is a typical elastic modulus of the suspension. A sedimenting colloidal crystal, according to [6], is therefore unstable in the $\mathrm{Pe} = \infty$ limit. Hence the question posed in our title: does the instability persist at finite Pe?

Our answer to this question is in two parts: (i) analytical results from a system of coupled nonlinear stochastic partial differential equations for displacement fields, and (ii) numerical studies of an equivalent driven lattice-gas model [7] describing the coupled dynamics of concentration and tilt fields. Our simulations of the lattice model are done in the parameter range where there is a local tendency to undergo the Crowley [6] instability. We find that the system is nonetheless *stable* with respect to clumping up to a critical Pe at which a *continuous nonequilibrium phase transition* to a clumped state takes place. For the corresponding parameter range the continuum model, neglecting nonlinearities and noise, is unstable without threshold, i.e., at $\mathrm{Pe} = 0$. In view of the results of the lattice model, we expect that nonlinearities and fluctuations shift the critical Pe for the onset of clumping in the continuum model to a nonzero value.

We first derive the continuum model and perform a mean-field analysis, then show how the lattice-gas model follows naturally from it. Ignoring inertia altogether, which is justified for the experimental geometry we wish to consider [8], the displacement field $\mathbf{u}(\mathbf{r}, t)$ of a d-dimensional lattice moving through a frictional medium with a mobility which depends on the local strain obeys

$$\dot{\mathbf{u}} = \boldsymbol{\mu}(\nabla\mathbf{u})\,(\mathbf{D}\nabla\nabla\mathbf{u} + \mathbf{F} + \boldsymbol{\zeta})\,, \tag{1}$$

where the first term on the right represents, through the tensor $\mathbf{D}$, the elastic restoring forces, the second is the driving force, and the third is a random force of thermal or possibly hydrodynamic origin [9]. $\boldsymbol{\mu}$ is the mobility tensor which, in the single particle limit for colloids in a solvent with viscosity η, approaches the isotropic Stokes's law value $\frac{1}{6\pi\eta a}$. We expand $\boldsymbol{\mu}$ in powers of $\nabla\mathbf{u}$,

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

where $\boldsymbol{\mu}_0$ is the mean macroscopic mobility of the undistorted crystal. For steady sedimentation along z, assuming isotropy in the $d-1$ transverse ($\perp$) dimensions but *not* under $z \to -z$, (1) and (2) lead directly to

$$\dot{\mathbf{u}}_\perp = \lambda_1\partial_z\mathbf{u}_\perp + \lambda_2\nabla_\perp u_z + \mathcal{O}(\nabla\nabla u) + \mathcal{O}(\nabla u\nabla u) + \mathbf{f}_\perp\,, \tag{3a}$$

$$\dot{u}_z = \lambda_3\nabla_\perp\cdot\mathbf{u}_\perp + \lambda_4\partial_z u_z + \mathcal{O}(\nabla\nabla u) + \mathcal{O}(\nabla u\nabla u) + f_z\,, \tag{3b}$$

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where the constant drift along z has been removed by comoving with the crystal [10]. Here $\mathbf{f}$ is a spatiotemporally white noise source [11] and λ_i are phenomenological coefficients whose origin, in the case of a colloidal crystal, lies in the hydrodynamic interaction between the particles [6,8,12]. We explain the physical content of the terms in (3) below. First note that linearizing and Fourier transforming in space and time yields modes with frequencies of the form $\omega = \pm\sqrt{\lambda_2\lambda_3}\, q - iDq^2$ for $q_z = 0, q_\perp \to 0$. If $\lambda_2\lambda_3 > 0$, this leads to wavelike excitations at small wave number q, which are not the usual shear waves of a crystal at equilibrium. The latter have been turned already into diffusive modes by the frictional dynamics adopted in (3). The speed of these waves is determined by the strain dependence of the mobility, and the damping by the tensor $\mathbf{D}$ which is a ratio of elastic constants to friction coefficients.

When $\lambda_2\lambda_3 < 0$, the dispersion relation at small q becomes $\omega \sim \pm iq$ so that the model is linearly unstable with growth rate $\propto q$. Thus there exist long-wavelength distortions of the perfect lattice which grow exponentially in time within the linear theory. We do not know the sign of $\lambda_2\lambda_3$ for moving flux lattices, but for colloidal crystals it is negative, making them linearly unstable. This is because hydrodynamic interactions cause denser regions in the suspension to sink faster, and tilted regions to move laterally in a way so as to cause an instability [6]. Note that in (3) the *linear* elasticity of the crystal enters at second order in wave number and can thus not alter our conclusions about linear stability at long wavelengths. For $q > q_* \sim \sqrt{\lambda_2\lambda_3}/D$, elastic forces suppress the linear instability. Small crystals are thus linearly stable.

To go beyond this linear analysis is daunting: even in $d = 2$, symmetry permits nine terms bilinear in $\nabla\mathbf{u}$ and six linear second derivative terms. Remarkably, all the essential physics is retained in a greatly simplified version in *one* space dimension. Consider a two-dimensional crystal described by a two-component displacement field (u_x, u_z), with the sedimentation direction z averaged out so that *only x derivatives are included*. The equations of motion, retaining the lowest order nonlinearities and gradients, read

$$\dot{u_x} = \lambda_2\partial_x u_z + \gamma_1\partial_x u_x\partial_x u_z + D_1\partial_x^2 u_x + f_x\,, \tag{4a}$$

$$\dot{u_z} = \lambda_3\partial_x u_x + \gamma_2(\partial_x u_x)^2 + \gamma_3(\partial_x u_z)^2 + D_2\partial_x^2 u_z + f_z\,. \tag{4b}$$

The physics of each of the terms in (3) or (4) is reasonably clear. The first two terms on the right of (4a) say that a tilt (i.e., $\partial_x u_z$) produces a lateral drift. The first two terms on the right of (4b) contain the concentration dependence, and the third (a Burgers/Kardar-Parisi-Zhang(KPZ)-like term [13]) the tilt dependence, of the vertical sedimentation speed. Note that the γ_i terms can be seen as arising from the dependence of the λ_is on concentration and tilt. The second derivative terms in both equations arise simply from elastic forces, and f_x and f_z are spatiotemporally white noises. The symmetry of (4) is under the *joint* inversion $x \to -x$, $u_x \to -u_x$.

For $\gamma_1/2\gamma_2 = \lambda_2/\lambda_3$, $u_x \to u_x - \lambda_2 x/\gamma_1$ eliminates the λ_2 and λ_3 terms from (4), reducing it to the model of Ertaş and Kardar (EK) [14] in their limit $\lambda_\perp = \lambda_\parallel$, with its higher symmetry ($x \to -x$), albeit in a state of nonzero mean $\partial_x u_x$. The fluctuation-dissipation theorem, Galilean invariance, and Cole-Hopf properties that arise in [14] for special parameter values thus obtain here as well. If in addition $\gamma_1 = 2\gamma_3$ and $D_1 = D_2$, the equations decouple in terms of transformed variables $\phi_\pm = u_x \pm \sqrt{\lambda_2/\lambda_3}\, u_z$ into two equations: $\dot{\phi}_\pm = \pm\sqrt{\lambda_2\lambda_3}\,\partial_x\phi_\pm + D_1\partial_x^2\phi_\pm \pm \sqrt{\lambda_2/\lambda_3}\,\gamma_2(\partial_x\phi_\pm)^2 + f_\pm$, a pair of independent KPZ [13] equations with oppositely directed kinematic wave [15] terms, and nonlinear couplings with opposite signs. Clearly, for these parameter values, the stable driven crystal should exhibit KPZ exponents in its correlation functions. The relevance of perturbations about the highly symmetric EK limit, as well as the statics and dynamics of the "stable" case $\lambda_2\lambda_3 > 0$ will be studied in later work. In the present paper we focus on $\lambda_2\lambda_3 < 0$.

Let us first look for steady-state solutions to (4) in the absence of noise, in terms of $\rho = \partial_x u_x$ (the local concentration fluctuation) and $\theta = \partial_x u_z$ (the local up or down tilt). If we restrict ourselves for simplicity to spatially uniform states with left-right symmetry (so that $\theta = 0$, and the net currents of ρ and θ are zero), we are left with only two possibilities: $\rho = \theta = 0$ or $\rho = -\lambda_3/\gamma_2$, $\theta = 0$. In the vicinity of $r \equiv \lambda_2\lambda_3 = 0$, the former is stable for $r > 0$, the latter for $r < 0$. This exchange of stabilities leads to a continuous onset of the $\rho \neq 0$ state, $\rho \sim |r|^\beta$ with $\beta = 1$. Similar analysis [16] gives a correlation length diverging as $|r|^{-\nu}$ with $\nu = 1/2$.

Instead of attempting a perturbative treatment of the effect of nonlinearities and fluctuations on the above mean-field picture, we replace the continuous variable x by a discrete index i, and $\partial u_x/\partial x$ by $\rho_i = u_x(i+1) - u_x(i)$ [similarly $\theta_i = u_z(i+1) - u_z(i)$], with ρ_i and θ_i restricted to ± 1. Such an approach [7] has proved very successful for simulating the KPZ equation. The "paramagnetic" phase of these Ising variables corresponds to the the undistorted crystal, and the "ferromagnetic" phase represents a macroscopically clumped and tilted state, in terms of suitable order parameters which we define below. The best way to visualize the discrete model is to think of two sublattices: a typical configuration can then be described by a sequence of spins $\rho_1\theta_1\rho_2\theta_2\rho_3\theta_3\ldots$. The dynamics of the spins is constructed by analogy with lattice models [7] for the KPZ equation, retaining the essential features of (4), viz., conservation of θ and ρ, stochasticity, lack of up-down symmetry, and the bias provided by each species on the motion of the other. The two approaches should yield identical long-distance properties.

Let us denote the states of ρ_i by "+," "−" and those of θ_i by "/" (up tilt) and "\" (down tilt). In the update

rule corresponding to the linearly unstable case of (4), the rates for the following exchanges are enhanced relative to the corresponding reverse rates: $+\backslash- \rightarrow -\backslash+$, $-/+ \rightarrow +/-$, $/ + \backslash \rightarrow \backslash + /$, and $\backslash - / \rightarrow / - \backslash$ Since we are modeling charge-stabilized suspensions, it is useful to introduce a repulsion between regions of high density in the form of an enhanced probability for $+ + -$ or $- + +$ to go to $+ - +$. Combining all of the above, we get the following exchange probabilities for adjacent pairs of concentration and tilt:

$$P_{\rho_i,\rho_{i+1}} = D_\rho - \epsilon_\rho \theta_i \rho_i + \alpha\{(1+\rho_i)(1+\rho_{i-1}) + (1+\rho_{i+1})(1+\rho_{i+2})\}, \quad (5a)$$

$$P_{\theta_i,\theta_{i+1}} = D_\theta + \epsilon_\theta \rho_{i+1}\theta_i + g_1\rho_{i+1} + g_2\theta_i, \quad (5b)$$

where D_ρ and D_θ are related to the elastic constants, ϵ_ρ, ϵ_θ, g_1, and g_2 to the γ_i and λ_i in (4), and α is the repulsion [17]. Note that decreasing α reduces the stiffness of the system, thus increasing the effective Péclet number. The last two terms in (5b) arise because of the lack of up-down symmetry. Our results in this paper are for $\epsilon_\rho\epsilon_\theta > 0$, corresponding to $\lambda_2\lambda_3 < 0$ in (4).

The mean value of both ρ and θ are expected to be zero in the experimental system. We worked, therefore, at zero total "magnetization" for both fields and studied the model starting from random initial conditions, evolving it according to the above update rules for various system sizes N. Periodic boundary conditions were used for all the runs. For high values of repulsion the spin configurations continued to be homogeneous under time evolution. When the repulsion was small or absent, there was a phase separation into regions of high and low concentration and of up and down tilt, separated by interfaces. Thus the lattice seems to be stable for strong repulsion, but undergoes Crowley's clumping instability [6] for weak repulsion. The same behavior, qualitatively, is observed as D_ρ or D_θ are increased keeping other parameters fixed [16].

To describe the ordered phase in this model with conserved dynamics, we use two essentially equivalent order parameters: $\Phi_\rho = \sqrt{-\frac{1}{N}\sum_i \rho_i\rho_{i+N/2}}$ (similarly Φ_θ for tilt), which measure how anticorrelated the spins are across half the system size; and $(|\Psi_\rho|, |\Psi_\theta|)$, the moduli of the Fourier amplitudes of the spin fields at the smallest nonzero wave vector $k_1 = \frac{2\pi}{N}$ [18] (the amplitude at $k = 0$ is zero). Figure 1 shows that the order parameter Φ_ρ is appreciable for small repulsion and decreases rapidly to a value consistent with zero for sufficiently large repulsion α. Moreover, Φ_ρ *increases* with N for α small and *decreases* [16] roughly as $1/\sqrt{N}$ for α large. There must thus be a continuous nonequilibrium phase transition at α around 0.05, although to pin down the critical value of α would require careful finite-size scaling.

We now present an independent check that the observed phase separation is not merely the result of transients. A truly phase-separated state in a system of length N should have barriers to remixing which grow as N^ζ for some power ζ. The lifetime of such a state would then go as $\exp(N^\zeta)$. To look for such barriers, we define a lifetime $\tau(N)$ to be the mean time of first passage of the order parameter $|\Psi_\rho|$ from a value $b_2|\Psi_\rho|_{\max}$ to $b_1|\Psi_\rho|_{\max}$, where $|\Psi_\rho|_{\max}$ is the maximum value of $|\Psi_\rho|$ over the runs. b_1 and b_2 are numbers *independent* of N, chosen to get good statistics; we took $b_2 = 0.8, b_1 = 0.5$.

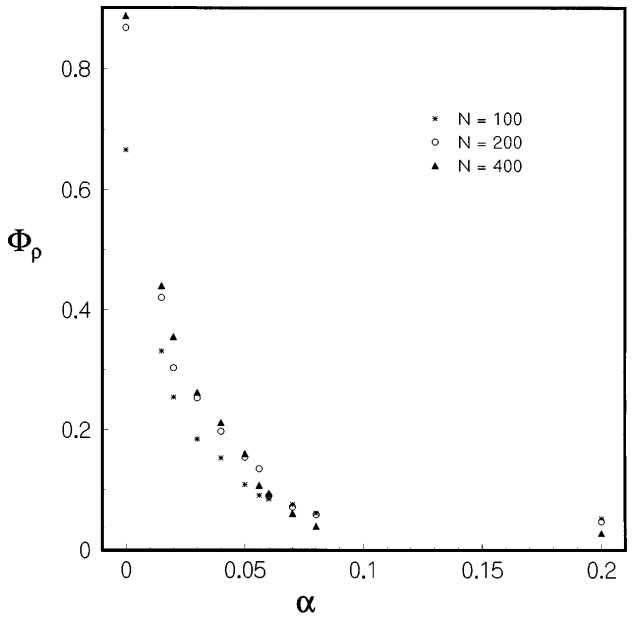

FIG. 1. Order parameter (Φ_ρ) as a function of repulsion strength α for various system sizes N. Increasing α is like decreasing Péclet number.

Figure 2 shows that $\tau(N)$ is roughly exponential in N for $\alpha = 0$ and distinctly faster than a power law, presumably a stretched exponential, for $\alpha = 0.015$. This is strong evidence [19] for a true clumped phase at weak repulsion.

To see why phase separation can occur in this one-dimensional model one has to look at the positions of the concentration and tilt domains. We find in our simulations that the system goes into a steady state in which the domains are staggered with respect to each other by an approximate distance $N/4$. This happens in such a way that a concentration interface $+ + + - - -$ lives in a region crowded with up tilts / which inhibit the exchange of a pair $+-$. The dissolution of the interface by interdiffusion of $+$ and $-$ thus requires uphill motion over a nonzero fraction of N.

Since our simple one-dimensional model undergoes a clumping transition, it is reasonable to expect that a real charge-stabilized colloidal crystal in a fluidized bed [4,8] will do so as well. The repulsion between polyballs may be decreased by adding salt to the fluid, which should lead to an observable clumping transition at ionic strengths much lower than those required to produce melting or aggregation at equilibrium. The clumping will manifest itself as a breakup of the crystal into smaller crystallites (since the crystal is stable at small enough system size),

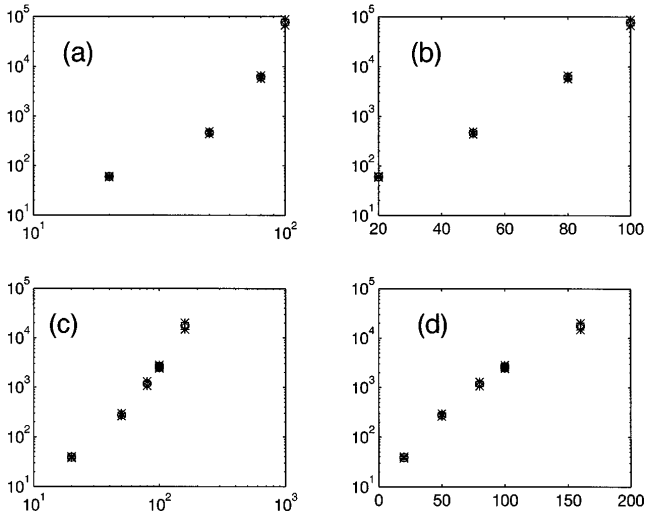

FIG. 2. Lifetime of the ordered phase $\tau(N)$, from the concentration order parameter $|\Psi_\rho|$. $\alpha = 0$: (a) log-log and (b) semi-log plot. $\alpha = 0.015$: (c) log-log and (d) semi-log plot. Similar results hold for the tilt order parameter [16]. Note the upward curvature of the log-log plots in either case, indicating lifetimes growing faster than any power of N. The downward curvature in (d) indicates that $\tau(N)$ is slower than e^N for $\alpha = 0.015$. The error in τ is determined by allowing the system to run for several lifetimes and statistically estimating the standard deviation.

separated by regions of strong upward fluid flow [20]. A detailed analysis of this dynamics would require the inclusion of the hydrodynamic flow.

In summary, we have demonstrated that the long-wavelength dynamics of a crystal moving steadily through a dissipative medium is qualitatively different from its equilibrium counterpart. In particular, we have shown that a natural driven lattice-gas model for this system shows a dramatic nonequilibrium phase transition to a clumped state, and we urge experimenters to test our predictions.

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