

# Suspensions far from equilibrium

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A review is presented of recent experimental and theoretical work on the dynamics of suspensions of particles in viscous fluids, with emphasis on phenomena that should be of interest to experimenters and theoreticians working on the statistical mechanics of condensed matter. The article includes a broad introduction to the field, a list of references to important papers, and a technical discussion of some recent theoretical progress in which the author was involved.

## Equilibrium and nonequilibrium suspensions

SUSPENSIONS of particles in a fluid medium are all around us<sup>1,2</sup>. Examples include river water, smog, blood, many liquid foods, many medicines, cosmetics, paints, and so on. The particles, which we shall call the solute, are generally submicron to several microns in size, and the suspending fluid, which we shall call the solvent, is frequently less dense than the solute. The viscosity of the solvent could range from that of water (or air) to several thousands of times higher. The field of suspension science has distinguished origins: Einstein's<sup>3</sup> interest in Brownian motion as evidence for the existence of molecules led him to calculate the viscosity and diffusivity of a dilute suspension (before diversions such as relativity and quantum mechanics took him over completely); Smoluchowski's<sup>4,5</sup> studies of sedimentation and aggregation in colloids led to major advances in the theory of stochastic processes; somewhat more recently, the challenging many-body nature of the dynamics of suspensions was highlighted in the work of Batchelor<sup>6</sup>. Today, the study of the static and dynamic properties of suspensions from the point of view of statistical mechanics is a vital part of the growing field of soft condensed matter science.

In applications and in industrial processing, suspensions are usually subjected to strongly *nonequilibrium* conditions. By nonequilibrium I mean that the system in question is driven by an external agency which does work on it – stirring, pumping, agitation – which the system dissipates internally. The bulk of interesting and, by and large, incompletely understood phenomena in suspension science, and in the area of complex fluids in general, are also those that occur far from equilibrium. Problems in which I have been or am currently interested are: the

melting of colloidal crystals when they are sheared<sup>7–13</sup>; spontaneous segregation in sheared hard-sphere suspensions<sup>14</sup>; the collapse of elastic colloidal aggregates under gravity<sup>15</sup>, and its possible relation to the instability of sedimenting crystalline suspensions<sup>12,16,17</sup>; the enhancement of red-blood-cell sedimentation rates in the blood of a very sick person<sup>18</sup>; and the puzzle<sup>19–23</sup> of the statistics of velocity fluctuations in ultraslow fluidized beds. (References 24–31 should give the reader an idea of the range of this field.) None of the observations in the papers I have mentioned can be understood purely with the methods used to study hydrodynamic instabilities: they are fluctuation phenomena, and therefore belong in this special issue on nonequilibrium statistical physics.

A suspension can be out of equilibrium in a number of ways: in particular, it could be in a nonstationary state (in the process of settling or aggregating or crystallizing, for example), or it could be stuck in a metastable amorphous state<sup>32</sup> or it could be held, by the application of a driving force, in a time-independent but not time-reversal invariant state with characteristics different from the equilibrium state. In this article we shall mainly be concerned with these *nonequilibrium steady states*, characterized by a *constant mean throughput* of energy. These are to be contrasted with *thermal equilibrium* states which have a *constant mean budget* of energy, i.e. a temperature. A suspension of charged Brownian particles with precisely the same density as the solvent, such as are discussed in the review article by Sood<sup>33</sup> is the standard example of an *equilibrium* suspension. The two most common ways of driving a suspension out of equilibrium are shear<sup>34</sup>, wherein the solute and solvent are jointly subjected to a velocity gradient, and sedimentation or fluidization<sup>35</sup>, where the velocity of solute relative to solvent is on average nonzero and spatially uniform. This latter class of problems is very close to the currently rather active area of driven diffusive systems<sup>36</sup>, which has provided much insight into statistical physics far from equilibrium.

Accordingly, this review will focus largely on sedimentation and fluidized beds, although a brief summary of shear-flow problems with relevant references will be provided. Even with this restriction, the field is much too vast to allow anything like representative coverage, so my choice of topics will be dictated by familiarity, in the hope that the problems I highlight will attract the reader to the area. It is in that sense not a true review article, but an advertisement for a field and therefore includes as an

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integral part a reasonably large list of references. The aim is to give a general idea of the richness of phenomena in nonequilibrium suspensions, as well as a technical understanding of some of my theoretical work in this field. To theoreticians, especially in India, this article argues that the vast literature on suspension dynamics is a largely untapped lode of problems in the statistical physics of driven systems. Elegant models of the sort popular among practitioners of the more mathematical sort of statistical mechanics could acquire a greater meaning and relevance if born out of an attempt to understand phenomena in these systems (for an example of such an attempt, see the section on the stability of steadily drifting crystals). I hope in addition that this article persuades condensed-matter experimenters in India to look for problems in these and related soft-matter systems (including powders, on which I am not competent to write), which are as rich as traditional solid-state systems, without the complications of low temperatures, high vacuum, etc.

I shall work exclusively in the limit of slow motions through highly viscous fluids, i.e. the limit of *low Reynolds number*  $Re \equiv Ua/\nu$ , where  $U$  is a typical velocity,  $a$  the particle size, and the kinematic viscosity  $\nu \equiv \mu/\rho$ ,  $\mu$  and  $\rho$  being respectively the shear viscosity and mass density of the solvent.  $Re$ , which measures the relative importance of inertia and viscosity in a flow, can here be thought of simply as the fraction of a particle's own size that it moves if given an initial speed  $U$ , before viscosity brings it to a halt. For bacteria (sizes of order 1 to 10 microns) swimming at, say, 1 to 10 microns a second in an aqueous medium,  $Re \sim 10^{-6}$  to  $10^{-4}$ , and for polystyrene spheres (specific gravity 1.05, radius 1 to 10  $\mu\text{m}$ ) sedimenting in water,  $Re \sim 10^{-7}$  to  $10^{-4}$ . So we are amply justified in setting  $Re$  to zero. Recall that in the low  $Re$  limit, things move at a speed proportional to how hard they are pushed, and stop moving as soon as you stop pushing. While this corresponds very poorly to *our* experience of swimming, it is an accurate picture of how water feels to a bacterium or to a colloidal particle. A good general introduction to the subject of zero Reynolds-number flow is given in Happel and Brenner<sup>37</sup>, although standard fluid dynamics texts<sup>38,39</sup> also discuss it. A very detailed treatment of the dynamics of hydrodynamically interacting particles can be found in ref. 40.

The dimensionless control parameter relevant to zero Reynolds number suspensions is the Peclet number  $Pe \equiv Ua/D$  (ref. 1), where  $D = T/6\pi\mu a$  is the Stokes-Einstein diffusivity of the particle at temperature  $T$ . In a shear flow with velocity gradient  $\dot{\gamma}$ , clearly  $U \sim \dot{\gamma}a$ , so that  $Pe \sim \dot{\gamma}a^2/D$ , which is the ratio of a diffusion time to a shearing time. In sedimentation,  $Pe$  measures the relative importance of settling (gravitational forces) and diffusion (thermal fluctuations), and can be expressed as  $m_{Rg}a/T$ , where  $m_{Rg}$  is the buoyancy-reduced weight of the particle,  $g$  being the acceleration due to gravity. Since  $Pe \propto a^4$ , it is

clear that small changes in the particle radius make a large difference. For polystyrene spheres in water,  $Pe$  ranges from 0.5 to 5000 as the radius is varied from 1 to 10  $\mu\text{m}$ . Suspensions with  $Pe \gg 1$  are termed non-Brownian, since their behaviour is dominated by the gravity-induced drift and the resulting solvent flow, not by thermal fluctuations. This  $Pe = \infty$ ,  $Re = 0$  limit turns out to be a very interesting one.

The most trivial example of a nonequilibrium suspension is a single solute particle, heavier than its solvent, so that in the presence of gravity it settles to the bottom of the container. If we wish to study the *steady-state* properties of this settling process, we must make the particle settle forever. There are two ways of doing this: either use a very tall container and study the flow around the particle as it drifts past the middle, or arrange to be in the rest frame of the particle. The latter is realized in practice by imposing an *upward* flow so that the viscous drag on the particle balances its buoyancy-reduced weight, suspending the particle stably. The nature of the flow around such an isolated particle, without Brownian motion and in the limit of low velocity, was studied about a century and a half ago<sup>41</sup>. As soon as the number of particles is three or greater, the motion becomes random even in the absence of a thermal bath (this can be seen either in Stokesian dynamics simulations<sup>42</sup> or simply by using particles large enough that Brownian motion due to thermal fluctuations is negligible) because the flow produced by each particle disturbs the others, leading to chaos<sup>43</sup>. In the limit of a large number of particles, the problem can thus be treated only by the methods of statistical mechanics (suitably generalized to the highly nonequilibrium situations we have in mind). A collection of particles stabilized against sedimentation by an imposed upflow is called a *fluidized bed*.

It is useful here to distinguish two classes of non-equilibrium systems. In *thermal* driven systems the source of fluctuations is *temperature*, and the driving force (shear, for example) simply advects the fluctuations injected by the thermal bath. An example of this type would be a suspension of submicron particles, whose Brownian motion is substantial, subjected to a shear flow. *Non-thermal* driven systems are more profoundly non-equilibrium, in that the same agency is responsible for the driving force and the fluctuations. A sedimenting non-Brownian many-particle suspension is thus an excellent example of a non-thermal nonequilibrium system.

The structure of this article is as follows. In the next section I summarize our present state of understanding of the shear-induced melting of colloidal crystals. Then, I discuss the physics of steadily sedimenting colloidal crystals, including the construction of an appropriate driven diffusive model for which exact results for many quantities of physical interest can be obtained. The next section introduces the reader to the seemingly innocent problem of the steady sedimentation of hard spheres

interacting only via the hydrodynamics of the solvent. Puzzles are introduced and partly resolved. The last section is a summary.

### Shear-melting of colloidal crystals

It has been possible for some years now to synthesize spheres of polystyrene sulphonate and other polymers, with precisely controlled diameter in the micron and submicron range. These 'polyballs' have become the material of choice for systematic studies in colloid science<sup>33</sup>. In aqueous suspension, the sulphonate or similar acid end group undergoes ionization. The positive 'counterions' go into solution, leaving the polyballs with a charge of several hundred electrons. The counterions together with any ionic impurities partly screen the Coulomb repulsion between the polyballs, yielding, to a good approximation in many situations, a collection of polymer spheres interacting through a Yukawa potential  $\exp(-\kappa r)/r$  with a screening length  $\kappa^{-1}$  in the range of a micron, but controllable by the addition of ionic impurities (NaCl, HCl, etc.). Not surprisingly, when the concentration of particles is large enough that the mean interparticle spacing  $a_s \sim \kappa^{-1}$ , these systems order into crystalline arrays with lattice spacings  $\sim a_s$ , comparable to the wavelength of visible light. The transition between colloidal crystal and colloidal fluid is a perfect scale model of that seen in conventional simple atomic liquids<sup>33</sup>. It is first-order and the fluid phase at coexistence with the crystal has a structure factor whose height is about 2.8. The transition is best seen by varying not temperature but ionic strength. These *colloidal crystals*<sup>33</sup> are very weak indeed, with shear moduli of the order of a few tens of dyn/cm<sup>2</sup>. The reason for this is clear: the interaction energy between a pair of nearest neighbour particles is of order room temperature, but their separation is about 5000 Å. Thus on dimensional grounds the elastic moduli, which have units of energy per unit volume, should be scaled down from those of a conventional crystal such as copper by a factor of  $5000^{-3} \sim 10^{-11}$ . It is thus easy to subject a colloidal crystal to stresses which are as large as or larger than its shear modulus, allowing one to study a solid in an extremely nonlinear regime of deformation. One can in fact make a colloidal crystal *flow* if the applied stress exceeds a modest minimum value (the yield stress) required to overcome the restoring forces of the crystalline state.

When a colloidal crystal is driven into such a steadily flowing state, it displays at least two different kinds of behaviour, with a complex sequence of intermediate stages which appear to be crossovers rather than true nonequilibrium phase transitions, and whose nature depends strongly on whether the system is dilute and charge-stabilized or concentrated and hard-sphere-like<sup>7-9</sup>. I shall focus on the two main 'phases' seen in the

experiments, not the intermediate ones. At low shear-rates, it flows while retaining its crystalline order: crystalline planes slide over one another, each well-ordered but out of registry with its neighbours. At large enough shear-rates, all order is lost, through what appears to be a nonequilibrium phase transition from a flowing colloidal crystal to a flowing colloidal liquid. The shear-rate required to produce this transition depends on the ionic strength  $n_i$ , and appears to go to zero as  $n_i$  approaches the value corresponding to the melting transition of a colloidal crystal at *equilibrium*. This connection to the equilibrium liquid-solid transition prompted us to extend the classical theory<sup>44</sup> of this transition to include the effects of shear flow. I will not discuss our work on that problem here: the interested reader may read about them in Ramaswamy and Renn<sup>10</sup> and Lahiri and Ramaswamy<sup>11</sup>. The problem remains incompletely understood, and work on it especially in experiments and simulations continues<sup>13</sup>. I mention it here as an outstanding problem in nonequilibrium statistical physics on which I should be happy to see further progress.

### The stability of steadily drifting crystals

The crystalline suspensions of the earlier section are generally made of particles heavier than water. Left to themselves, they will settle slowly, giving slightly inhomogeneous, bottom-heavy crystals with unit cells shorter at the bottom of the container than at the top. To get a truly homogeneous crystal, one must counteract gravity. This is done, as remarked earlier, in the fluidized bed geometry, where the viscous drag of an imposed upflow balances the buoyant weight of the particles. As a result, one is in the rest frame of a steadily and perpetually sedimenting, spatially uniform crystalline suspension, whose steady-state statistical properties we can study. Although most crystalline suspensions are made of heavier-than-water particles, and therefore do sediment, there have been only a few studies<sup>45</sup> that focus on this aspect. Rather than summarizing the experiments, I shall let the reader read about them<sup>45</sup>. Our work was in fact inspired by attempts to understand drifting crystals in a different context, namely flux-lattice motion<sup>46</sup>, and the interest in crystalline fluidized beds arose when we chanced upon some papers by Crowley<sup>47</sup>, to whose work I shall return later in this review.

As with crystals at equilibrium, the first thing to understand was the response to weak, long-wavelength perturbation. For a crystal *at thermal equilibrium*, elastic theory<sup>48</sup> and broken-symmetry dynamics<sup>49,50</sup> provide, in principle, a complete answer. Our crystalline fluidized bed, however, is far from equilibrium, in a steady state in which the driving force of gravity is balanced by viscous dissipation. We must therefore simply guess the correct

form for the equations of motion, based on general symmetry arguments<sup>16</sup>. This general form should apply in principle to any lattice moving through a dissipative medium without static inhomogeneities. Apart from the steadily sedimenting colloidal crystal, another example is a flux-point lattice moving through a thin slab of ultraclean type II superconductor under the action of the Lorentz force due to an applied current<sup>51</sup>. We will restrict our attention to the former case alone in this review.

One technical note is in order before we construct the equations of motion. A complete analysis of the sedimentation dynamics of a three-dimensional crystalline suspension requires the inclusion of the hydrodynamic velocity field as a dynamical variable. This is because the momentum of a local disturbance in the crystalline fluidized bed cannot decay locally but is transferred to the nearby fluid and thence to more and more remote regions. We get around this difficulty by considering an experimental geometry in which a thin slab of crystalline suspension (particle size  $a \ll$  interparticle spacing  $\ell$ ) is confined in a container with dimensions  $L_x, L_z \gg L_y \sim \ell$  (gravity is along  $-\hat{z}$ ). The *local* hydrodynamics that (see below) leads to the configuration-dependent mobilities<sup>16,47</sup> 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, so that the velocity field of the fluid can be ignored.

Instead of keeping track of individual particles, we work on scales much larger than the lattice spacing  $\ell$ , treating the crystal as a permeable elastic continuum whose distortions at point  $\mathbf{r}$  and time  $t$  are described by the (Eulerian) displacement field  $\mathbf{u}(\mathbf{r}, t)$ . Ignoring inertia as argued above, the equation of motion must take the general form velocity = mobility  $\times$  force, i.e.

$$\frac{\partial}{\partial t} \mathbf{u} = \mathbf{M}(\nabla \mathbf{u}) (\mathbf{K} \nabla \nabla \mathbf{u} + \mathbf{F} + \mathbf{f}). \quad (1)$$

In eq. (1), 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 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 and novel physics in these equations, when the driving force is nonzero, lies in the local mobility tensor  $\mathbf{M}$ , 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 hydrodynamic interaction of the moving particles with the medium, and will in general depend on the local configuration of the particles. If the structure in a given region is distorted relative to the

perfect lattice, the local mobility will depart from its ideal-lattice value as well, through a dependence on the distortion tensor  $\nabla \mathbf{u}$ . Assuming, as is reasonable, that  $\mathbf{M}$  can be expanded in a power series in  $\nabla \mathbf{u}$ , eq. (1) leads to

$$\dot{u}_x = \lambda_1 \partial_z u_x + \lambda_2 \partial_x u_z + O(\nabla \nabla u) + O(\nabla u \nabla u) + f_x, \quad (2)$$

$$\dot{u}_z = \lambda_3 \partial_x u_x + \lambda_4 \partial_z u_z + O(\nabla \nabla u) + O(\nabla u \nabla u) + f_z, \quad (3)$$

where the nonlinear terms as well as those involving the coefficients  $\lambda_i$  are a consequence of the sensitivity of the mobility to local changes on the concentration and orientation, and are proportional to the driving force (gravity). They are thus absent in a crystal at equilibrium. The  $\{\lambda_i\}$  terms *dominate* the dynamics at sufficiently long wavelength for a driven crystal, since they are lower order in gradients than the  $O(\nabla \nabla u)$  elastic terms. It is immediately obvious that the sign of the product  $\alpha \equiv \lambda_2 \lambda_3$  decides the long-wavelength behaviour of the steadily moving crystal. If  $\alpha > 0$ , small disturbances of the sedimenting crystal should travel as waves with speed  $\sim \lambda_i$ , while if  $\alpha < 0$ , the system is linearly unstable: disturbances with wave vector  $\mathbf{k}$  with  $k_x \neq 0$  should *grow* at a rate  $\sim \sqrt{|\alpha|} k_x$ . Symmetry cannot tell us which of these happens: the sign of  $\alpha$  depends on the system in question, and is determined by a more microscopic calculation than those presented above.

This is where the work of Crowley<sup>47</sup> comes in. He studied the settling of carefully prepared ordered arrays of steel balls in turpentine oil, and found they were unstable. He also *calculated* the response of such arrays to small perturbations, taking into account their hydrodynamic interaction, and found that the theory said that they should indeed be unstable. Thus, one would expect crystalline fluidized beds to be linearly unstable (the analogous calculation for drifting flux lattices in type II superconductors finds linearly stable behaviour<sup>51</sup>) and is hence described by eqs (2) and (3) with  $\alpha < 0$ . However, Crowley's<sup>47</sup> arguments and experiments were for an array of particles merely *prepared* in the form of an ordered lattice. Unlike in the case of a charge-stabilized suspension, there were no forces that *favoured* such order at equilibrium in the first place. Our model eqs (2) and (3), however, contain such forces as well as nonlinearities and noise. While the elastic terms are of course subdominant in a linearized treatment at long wavelength to the  $\{\lambda_i\}$  terms, which are  $O(\nabla u)$ , we asked whether these terms, in combination with nonlinearities and noise could undo or limit the Crowley ( $\alpha < 0$ ) instability.

We answer this question not by studying eq. (3) with  $\alpha < 0$ , but instead by building a discrete Ising-like dynamical model embodying the essential physics of those

equations. The crucial features of the dynamics implied by eq. (3) (see Happel and Brenner<sup>37</sup> and Crowley<sup>47</sup>) are that a downtilt favours a drift of material to the right, an uptilt does the opposite, an excess concentration tends to sink, and a deficit in the concentration to float up. Let us implement this dynamics in a one-dimensional system with sites labelled by an integer  $i$ , and describe the state of the lattice of spheres in terms of an array of two types of two-state variables:  $\rho_i = \pm$ , which tells us if the region around site  $i$  is compressed (+) or dilated (-) relative to the mean, and  $\theta_i = \pm$  which tells us if the local tilt is up (+, which we will call '∕') or down (-, which we will denote '\'). Let us put the two types of variables on the odd and even sublattices. A valid configuration could then look like  $\rho_1\theta_1\rho_2\theta_2\rho_3\theta_3\rho_4\theta_4 = +\ \backslash\ +\ /-\ \backslash\ +\ \backslash\ -\ /-\ /$ . An undistorted lattice is then a statistically homogeneous admixture of + and - for both the variables (a 'paramagnet'). The time-evolution of the model is contained in a set of transition rates for passing from one configuration to another, as follows<sup>16,17,52</sup>:

$$\begin{aligned}
 W(+\ \backslash\ -\ \rightarrow\ -\ \backslash\ +) &= D + a \\
 W(-\ \backslash\ +\ \rightarrow\ +\ \backslash\ -) &= D - a \\
 W(-\ \backslash\ +\ \rightarrow\ +\ \backslash\ -) &= D' + a' \\
 W(+\ \backslash\ -\ \rightarrow\ -\ \backslash\ +) &= D' - a' \\
 W(/ + \ \backslash\ \rightarrow\ \backslash + /) &= E + b \\
 W(\ \backslash + / \rightarrow / + \ \backslash) &= E - b \\
 W(\ \backslash - / \rightarrow / - \ \backslash) &= E' + b' \\
 W(/ - \ \backslash \rightarrow \ \backslash - /) &= E' - b',
 \end{aligned}
 \tag{4}$$

where the first line, for example, represents the rate of + - going to - + in the presence of a downtilt \, and so on.  $D, E, D', E'$  (all positive) and  $a, b, a', b'$  are all in principle independent parameters, but it turns out to be sufficient on grounds of physical interest, relevance to the sedimentation problem, and simplicity to consider the case  $D = D', E = E', a = a', b = b'$ , with  $\gamma \equiv ab > 0$  corresponding to  $\alpha = \lambda_2\lambda_3 < 0$  in eqs (2) and (3).

Associating  $\rho_i$  with  $\partial_x u_x$  and  $\theta_i$  with  $\partial_x u_z$  we expect that the above stochastic dynamics yields, in the continuum limit, the same behaviour as a one-dimensional version of eqs (2) and (3) in which all  $z$  derivatives are dropped, and appropriate nonlinearities are included to ensure that the instability seen in the linear approximation is controlled. This mapping provides a convincing illustration of the close connection between quite down-to-earth problems in suspension science and the area of driven diffusive systems which is currently the subject of such intense study<sup>36</sup>.

Initial numerical studies<sup>16</sup> of eq. (4) suggested a tendency towards segregation into macroscopic domains of + and -, as well of / and \ with interfaces between + and - are shifted with respect to those between / and \ by a quarter of the system size. This arrangement is just such

as to make it practically impossible, given the dynamical rules eq. (4), for the domains to remix. The numerics seemed to suggest that enough interparticle repulsion or a high enough temperature could undo the phase separation, but this turned out to be a finite size effect. We have shown<sup>52</sup> that the model *always* phase-separates for  $\gamma > 0$ . More precisely, we have shown exactly, for the symmetric case  $\sum_i \rho_i = \sum_i \theta_i = 0$ , if  $\frac{b}{E} = \frac{a}{D}$ , that the steady state of our model obeys detailed balance, (i.e. acts like a thermal *equilibrium* system) with respect to the energy function  $\mathcal{H} = \epsilon \sum_{k=1}^N [\sum_{j=1}^k \theta_j] \rho_k$ . A little reflection will convince the reader that this is like the energy of particles (the  $\{\rho_i\}$ ) on a hill-and-valley landscape with a height profile whose local slope is  $\theta_i$ . Since the dynamics moves particles downhill, and causes occupied peaks of the landscape to turn into valleys, it is clear that the final state of the model will be one with a single valley, the bottom of which is full of pluses (and the upper half full of minuses). It is immediately clear that this phase separation is very robust, and will persist at any finite temperature (i.e. any finite value of the base rates  $D$  and  $E$ ). Figure 1 demonstrates this graphically.

Many properties of the model (eq. (4)) can be obtained exactly in the detailed-balance limit mentioned above. These include the prediction that the phase separation, while robust, is anomalously slow: domain sizes grow with time  $t$  as  $\log t$ . It is important to note that the behaviour obtained exactly for the above special values of parameters can be shown to apply much more generally. This phase separation, in terms applicable to a real crystalline suspension, leads to macroscopic particle-rich and particle-poor regions. In the middle of each such region, one expects a fracture separating regions of opposite tilt. There are preliminary reports of such behaviour in experiments<sup>53</sup>, and it is tempting to think that some recent observations<sup>15</sup> of the collapse of elastic aggregates in suspension are related to the above ideas, but this is mere speculation at this point. Detailed experiments on large, single-crystalline fluidized beds are needed to test the model.

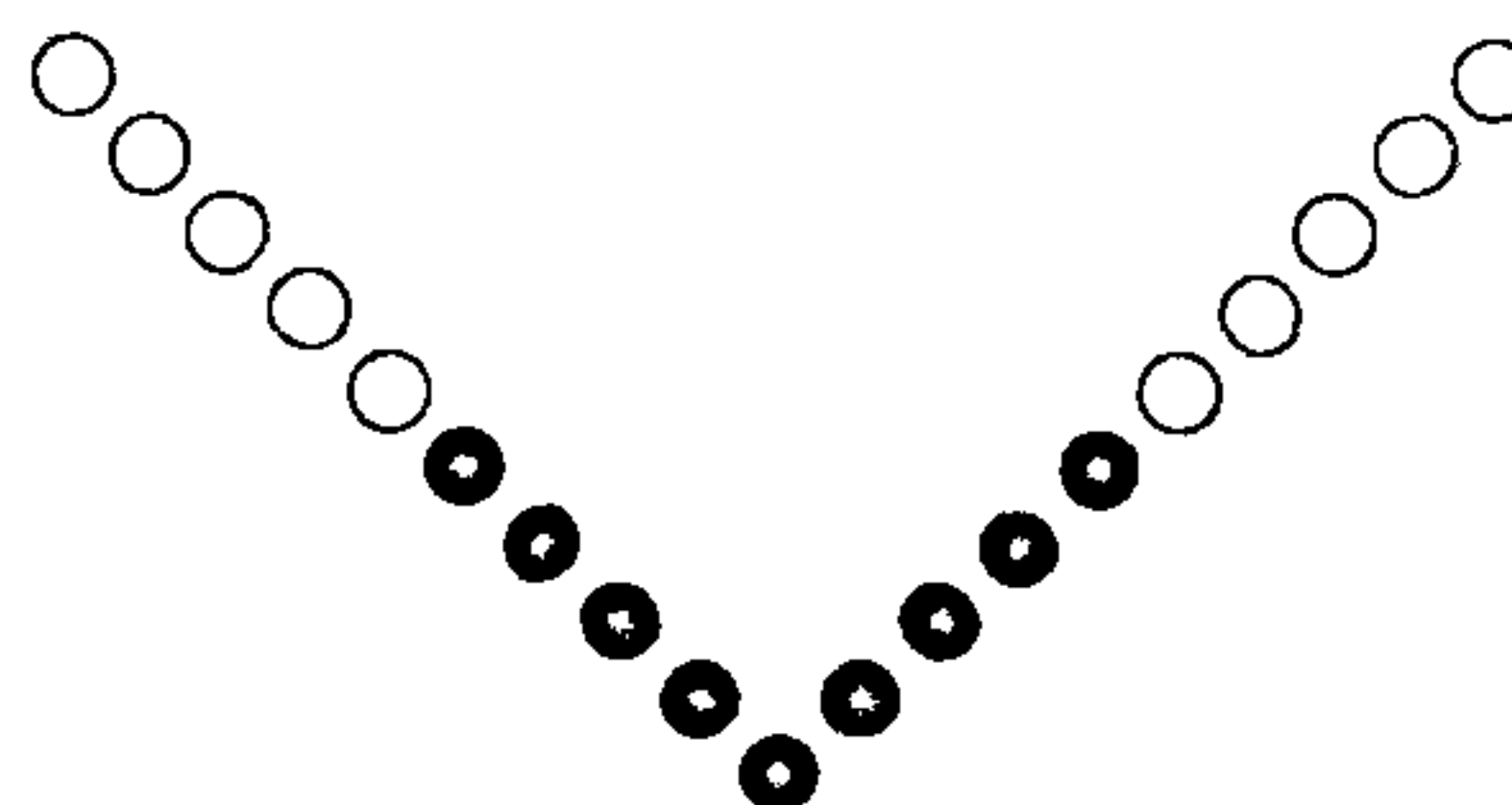


Figure 1. A schematic picture of the final state of phase separation of the one-dimensional model: The tilt field has been summed to give the height of the profile at each point, the filled circles denote pluses and the open circles minuses. It is clear that for the phase separation to undo itself a plus, for example, must climb a distance of order 1/4 of the system size. The time for this to happen diverges exponentially with system size.

## Velocity fluctuations in fluidized beds

Hard-sphere suspensions, in which the only interactions are hydrodynamic, are a subject of continuing interest to fluid dynamicists, as should be clear from a glance at current issues of fluid mechanics journals (or physics journals, for that matter). In practice, these suspensions are charge-stabilized, with ionic strength so large that electrostatics is screened within a tiny distance of the particle surface<sup>33</sup>, making the particles effectively hard spheres. Up to volume fractions of about 0.5, the particles in these suspensions have only short-ranged positional correlations, and move freely. They are thus *fluid* suspensions unlike the crystalline suspensions we discussed earlier. The suspensions of our interest are made of large particles (several microns in size), so that Brownian motion is unimportant, but (as discussed earlier) there is nonetheless a substantial random component to the motion if the system is sedimenting or sheared. Single-component suspensions are themselves the home of many mysteries, as the references in earlier sections will bear out, while two (or more) -component suspensions<sup>31,54</sup> are understood even less. I will focus on just one puzzle in monodisperse suspensions, and on what I believe is its resolution using methods imported from time-dependent statistical mechanics.

Until recently, theory and experiment disagreed rather seriously on the nature of velocity fluctuations in these suspensions in steady-state sedimentation. A simple theory<sup>19</sup> predicted that  $\sigma_v(L)$ , the standard deviation of the velocity of the particles sedimenting in a container of size  $L$ , should diverge as  $\sqrt{L}$ , while experiments<sup>20,21</sup> saw no such dependence (For a dissenting voice, see Tory *et al.*<sup>55</sup>). More precisely, Segrè *et al.*<sup>21</sup> see size dependence for  $L$  smaller than a 'screening length'  $\xi \sim 30$  interparticle spacings, but none for  $L > \xi$ . Further confusion is provided by direct numerical simulations which do seem to see the size-dependence<sup>56</sup>, although these contain about 30,000 particles, which, while that sounds large, means they are only about 30 particles on a side, which probably just means that these huge simulations have not crossed the scale  $\xi$  of Segrè *et al.*<sup>21</sup>!

It is important to note that experiments in this area are frequently done by direct imaging of the particles at each time (Particle Imaging Velocimetry or PIV, see Adrian<sup>57</sup>). Since motions are slow (velocities of a few  $\mu\text{m/s}$ ), this is relatively easy to do. Once the data on particle positions are stored, they can be analysed in detail on a computer and objects of interest such as correlation functions extracted. This is one of the nice things about the field of suspension science: problems of genuine interest to practitioners of statistical physics arise and can be studied in relatively inexpensive experiments, which measure quite directly the sorts of quantities a theoretician can calculate.

Our contribution to this issue<sup>22,23</sup> is to formulate the problem in the form of generalized Langevin equations,

and then use methods well-known in areas such as dynamical critical phenomena to construct a phase diagram for sedimenting suspensions. Our approach is close in spirit to that of Koch and Shaqfeh<sup>58</sup>, but differs in detail and predictive power. We find that there are two possible nonequilibrium 'phases', which we term 'screened' and 'unscreened', for a steadily sedimenting suspension. In the screened phase,  $\sigma_v(L)$  is independent of  $L$ , while in the unscreened phase, it diverges as in Caflisch and Luke<sup>19</sup>. The two phases are separated by a boundary which has the characteristics of a continuous phase transition, in that a certain correlation length diverges there in a power-law manner. Although the relation between the parameters in terms of which our phase diagram is drawn and conventional suspension properties is still somewhat uncertain, we believe that the screened and unscreened phases should occur respectively at large and small Peclet numbers. Properties such as particle shape (aspect ratio) and Reynolds number also probably play a role in determining where in our phase diagram a given system lies.

Let us begin by reconstructing the prediction of Caflisch and Luke<sup>19</sup>. Let  $\delta \mathbf{v}(\mathbf{r})$  and  $\delta c(\mathbf{r})$  be respectively the local velocity and concentration fluctuations about the mean in a steadily sedimenting suspension, each of whose particles has a buoyancy-reduced weight  $m_{RG}$ . Then, ignoring inertia, the balance between gravitational (acceleration  $g$ ) and viscous (viscosity  $\eta$ ) force densities is expressed in the relation  $\eta \nabla^2 \delta \mathbf{v} \sim m_{RG} \delta c(\mathbf{r}, t)$ . This implies that a concentration fluctuation at the origin leads to a velocity field  $\sim g/(\eta r)$  a distance  $r$  away. Now, if you *assume*, with Caflisch and Luke<sup>19</sup>, that the  $\delta c$ 's are *completely* random in space or have at best a finite range of correlations, the variance  $\langle |\delta \mathbf{v}|^2 \rangle$  due to all the concentration fluctuations is obtained by simply squaring and adding incoherently giving, for a container of finite volume  $L^3$ ,  $(g/\eta)^2 \int d^3r (1/r^2) \sim L$ . What this tells us, really, is that *either*  $\langle |\delta \mathbf{v}|^2 \rangle$  diverges *or* the concentration fluctuations are strongly *anticorrelated* at large length scales. What we should do, therefore, is not *assume* a spectrum of concentration fluctuations and *calculate* the velocity variance, but *calculate* both of them.

To this end, let me first summarize the construction of the equations of motion for the suspension. Our description is phenomenological, to precisely the same extent as a continuum Ginzburg-Landau model or its time-dependent analogue for an equilibrium phase transition problem such as phase separation in a binary fluid. Our construction is constrained by the following general principles, each of which plays an indispensable role: (i) We need to keep track only of the slowest variables in the problem. (ii) *Assuming* our suspension has not undergone a phase transition into a state where some invariance (translation, rotation) is spontaneously broken, the only slow variables are the local densities of conserved quantities. For an incompressible suspension, these are

just the particle concentration and the suspension momentum density (effectively, for a dilute suspension, the fluid velocity field). (iii) To get the long wavelength physics right, we can work at leading order in a gradient expansion. (iv) We must keep all terms *not explicitly forbidden* on grounds of symmetry, and impose *no* relations amongst the phenomenological parameters other than those *forced on us* by the symmetries of the problem. (v) Since the microscopic Stokesian dynamics shows chaotic behaviour and diffusion (see earlier sections), our coarse-grained model, since it is an effective description for the long-wavelength degrees of freedom, should contain *stochastic terms* (a direct effect of the eliminated fast degrees of freedom) as well as diffusive terms (an indirect effect) but with no special (fluctuation–dissipation) relation between them. Since only a limited range of modes (say, with wave numbers larger than a cutoff scale  $\Lambda$ , of order an inverse interparticle spacing) have been eliminated, the resulting noise can have correlations only on scales smaller than  $\Lambda^{-1}$ . As far as a description on scales  $\gg \Lambda^{-1}$  is concerned, the noise can be treated as spatially uncorrelated. This approach, we argue, should yield a complete, consistent description of the long-time, long-wavelength properties of the system in question. These premises accepted, one is led inevitably to a stochastic advection–diffusion equation

$$\frac{\partial \delta c}{\partial t} + \delta \mathbf{v} \cdot \nabla \delta c = [D_{\perp} \nabla_{\perp}^2 + D_z \nabla_z^2] \delta c + \nabla \cdot \mathbf{f}(\mathbf{r}, t), \quad (5)$$

for the concentration field  $\delta c$  and the Stokes equation

$$\eta \nabla^2 \delta v_i(\mathbf{r}, t) = m_R g P_{iz} \delta c(\mathbf{r}, t). \quad (6)$$

for the velocity field  $\delta v$ . Equation (6) simply says that a local concentration fluctuation, since the particles are heavier than the solvent, produces a local excess force density which is balanced by viscous damping. In eq. (5),  $\delta_{ij}^z$  and  $\delta_{ij}^{\perp}$  are respectively the projectors along and normal to the  $z$ -axis, which is the direction of sedimentation. Incompressibility ( $\nabla \cdot \delta \mathbf{v} = 0$ ) has been used to eliminate the pressure field in eq. (6) by means of the transverse projector  $P_{ij}$  (in Fourier space  $P_{ij}(\mathbf{q}) = \delta_{ij} - q_i q_j / q^2$ ). Equation (5) contains the advection of the concentration by the velocity, an anisotropic hydrodynamic diffusivity ( $D_{\perp}, D_z$ ), and a noise or random particle current  $\mathbf{f}(\mathbf{r}, t)$ . The last two are of course a consequence of the eliminated small-scale chaotic modes. The noise is taken, reasonably, to have Gaussian statistics with mean zero and covariance

$$\langle f_i(\mathbf{r}, t) f_j(\mathbf{r}', t') \rangle = c_0 (N_{\perp} \delta_j^{\perp} + N_z \delta_j^z) \delta(\mathbf{r} - \mathbf{r}') \delta(t - t'), \quad (7)$$

where  $c_0$  is the mean concentration. In an ideal non-Brownian system, the noise variances ( $N_{\perp}, N_z$ ) and the

diffusivities ( $D_{\perp}, D_z$ ) should, on dimensional grounds, scale (at fixed volume fraction) as the product of the Stokesian settling speed and the particle radius. However, no further relation between the noise and the diffusivities may be assumed, since this is a nonequilibrium system. In laboratory systems there will of course be in addition a thermal contribution to both noise and diffusivities. In either case, what matters, and indeed plays a crucial role in our nonequilibrium phase diagram, is that the parameter

$$K \equiv N_{\perp} D_z - D_{\perp} N_z \quad (8)$$

is in general nonzero. Since at equilibrium the ratio of noise strength to kinetic coefficient is a temperature,  $K$  measures the anisotropy of the effective ‘temperature’ for this driven system.

As a first step towards extracting the behaviour of correlation functions and hence the velocity variance, note that eqs (5) and (6) can be solved exactly if the nonlinear term in eq. (5) is ignored. This amounts to allowing concentration fluctuations to produce velocity fluctuations while forgetting that the latter must then advect the former. If we do this, then it is straightforward to see by Fourier-transforming eq. (5) that the static structure factor  $S(\mathbf{q}) \equiv c_0^{-1} \int_r e^{-i\mathbf{q}\cdot\mathbf{r}} \langle \delta c(0) \delta c(r) \rangle$  (where the angle brackets denote an average over the noise) for concentration fluctuations with wave vector  $\mathbf{q} = (\mathbf{q}_{\perp}, q_z)$  in a suspension with mean concentration  $c_0$  is

$$S(\mathbf{q}) = \frac{N_{\perp} q_{\perp}^2 + N_z q_z^2}{D_{\perp} q_{\perp}^2 + D_z q_z^2}, \quad (9)$$

and is hence *independent* of the *magnitude* of  $\mathbf{q}$ . In particular, it is therefore non-vanishing at small  $q$ . This can quickly be seen to imply, through eq. (6), that the velocity variance diverges exactly as in Caflisch and Luke<sup>19</sup>. In other words, Caflisch and Luke<sup>19</sup> fail to take into account the hydrodynamic interaction between concentration fluctuations. If nonlinearities are to change this, they must leave the noise relatively unaffected while causing the relaxation rate ( $D_{\perp} q_{\perp}^2 + D_z q_z^2$  in the linearized theory) to become nonzero at small  $q$ . This, if it happens, would be called ‘singular diffusion’ since diffusive relaxation rates normally vanish at small wave number. We shall see below that this does happen, in a substantial part of the parameter space of this problem.

More precisely, we have been able to show<sup>22,23</sup>, in a fairly technical and approximate ‘self-consistent’ calculation which I shall not present here, that the behaviour once the advection term is included depends on the temperature anisotropy parameter  $K$  defined in eq. (8). If  $K = 0$ , i.e. if the noise and the diffusivities happen to obey a fluctuation–dissipation relation, then the static structure factor is totally unaltered by the advective nonlinearity, and therefore the velocity variance diverges as in Caflisch

and Luke<sup>19</sup>. If  $K$  is larger than a critical value  $K_c$ , i.e. the fluctuations injected by the noise are substantially more abundant for wave vectors in the  $xy$  plane than for those along  $z$ , then there is a length scale  $\xi$  such that  $S(\mathbf{q}) \rightarrow 0$  for  $q\xi \ll 1$  with  $q_\perp \gg q_z$ . Thus the velocity variance is finite, and independent of system size  $L$  for  $L > \xi$ . We call this 'screening', and the regions  $K > K_c$  and  $K < K_c$  as the *screened* and *unscreened* phases, respectively. A schematic phase diagram is given in Figure 2. However,  $\xi$  diverges as  $K \rightarrow K_c$ . For  $K < K_c$ , according to preliminary calculations, the long-wavelength behaviour is the same as that at  $K = 0$ , although a detailed renormalization-group calculation to establish this is still in progress.

Let me try to provide a qualitative understanding of these results. The basic question is: how does an imposed long-wavelength inhomogeneity in the solute concentration decay in a steadily sedimenting suspension? It can, of course, always do so by hydrodynamic diffusion. In addition, it can scatter off the background of chaos-induced fluctuations, which I will call *noise-injected fluctuations* (NIF). This scattering is best thought of as the advection of the imposed inhomogeneity by the velocity field produced by the NIF. So consider two cases: (a) where the NIF has wave vector predominantly along  $z$ , and (b) where the wave vector is mainly in the  $xy$  plane. In (a), the induced flow has a  $z$ -velocity which alternates in sign as a function of  $z$ . The advection of the imposed inhomogeneity by this flow will *concentrate* it further, in general, thus enhancing the perturbation. In case (b), i.e. when the NIF has variation mainly along  $xy$ , the resulting  $z$ -velocity will alternate in sign along  $x$  and  $y$ , which will break up the inhomogeneity. Thus, a noise with Fourier components only with wave vector along  $z$  would give a negative contribution to the damping rate due to scattering, while one with Fourier components with wave vector only orthogonal to  $z$  would give a purely positive contribution. In general, it is thus clear that this mechanism gives a correction to the damping rate proportional to  $N_\perp - N_z$  (assuming, for simplicity, the same diffusivity in all directions). In addition, the long-ranged nature of the hydrodynamic interaction means that no matter how long-wavelength the NIF, it will produce macroscopic flows on scales comparable to its wavelength (and instantaneously, in the Stokesian approximation), hence the *singular diffusion*.

In addition, we predict the form of static and dynamic correlation functions of the concentration or the velocity fields in detail, in the screened and unscreened phases as well as at the transition between them<sup>22,23</sup>. Very careful experiments, in particular some light scattering measurements at very small angles, are currently underway to test our predictions.

We close this section by remarking that ours is not the only candidate theory of the statistics of fluctuations in zero Reynolds number fluidized beds. An earlier, nominally more microscopic approach<sup>58</sup> had some similar

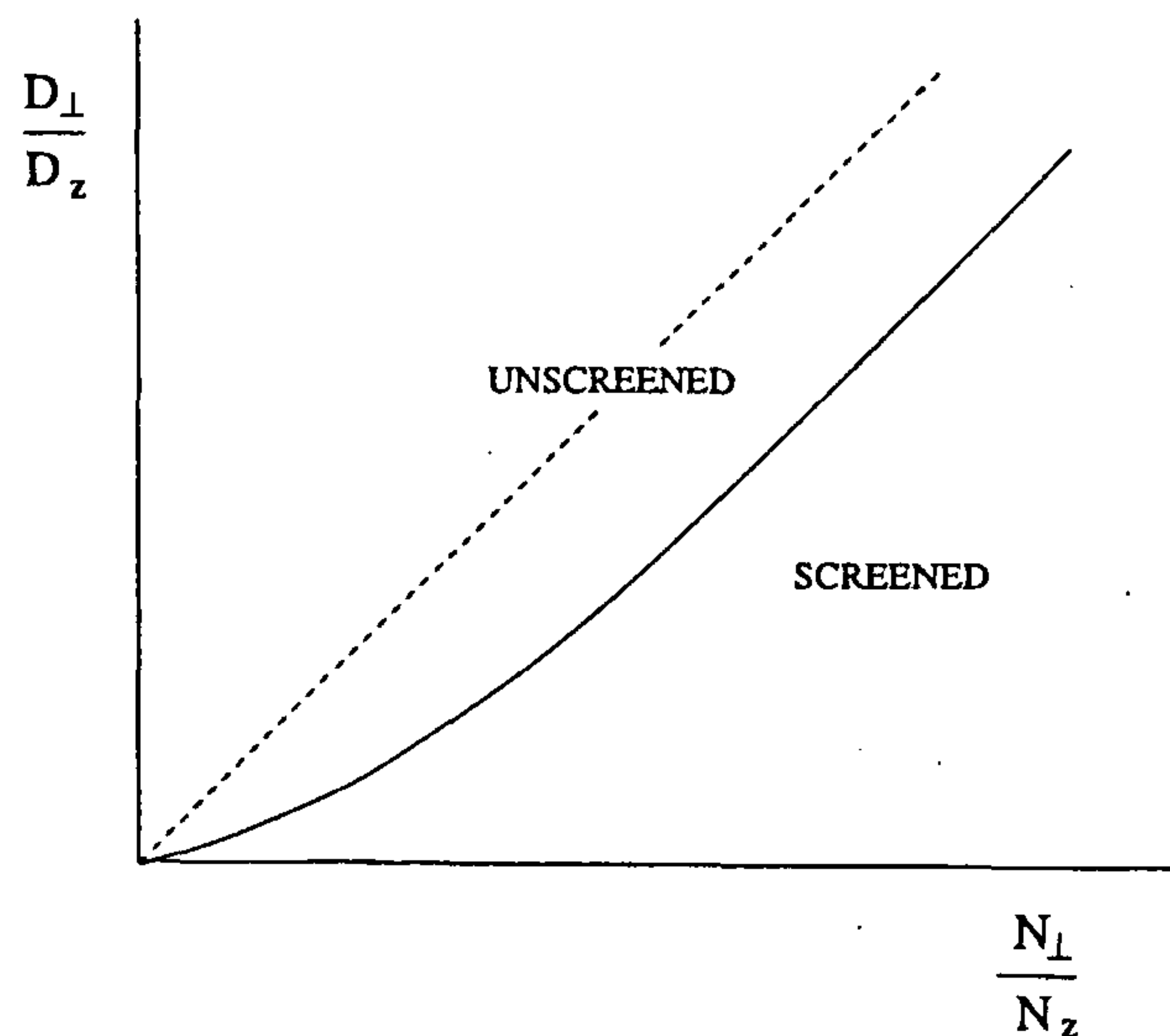


Figure 2. Schematic phase diagram for steadily sedimenting hard spheres (after Levine *et al.*<sup>22</sup>). On the dashed line and, we conjecture, near it, the structure factor is as in the linearized theory, and screening fails. The solid curve is the phase boundary between the screened and unscreened regions.

conclusions; we, however, disagree with that work in several details. There are some who criticize the experiments of Segrè<sup>21</sup> because they are done in narrow cells which could introduce finite size effects. There is also a very qualitative set of arguments<sup>59</sup> based on an analogy with high Prandtl number turbulence; it is unclear at this stage whether that work is a recasting of our theory of the screened phase or distinct from it.

## Conclusion

This review has tried to summarize experimental and theoretical work in the area of suspension hydrodynamics, from the point of view of a physicist interested in nonequilibrium statistical mechanics. The major aim has been to convince condensed matter physicists in India that this is a field which merits their attention. Apart from listing a large number of general references, I have tried to support my case by describing some problems on which I have worked, which have their origins squarely within suspension science, but whose solutions required all the machinery of statistical mechanics and used in a crucial way the fact that the systems concerned were not at thermal equilibrium. I hope this article will win some converts to this wonderful field.

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