

## Screened and Unscreened Phases in Sedimenting Suspensions

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A coarse-grained stochastic hydrodynamical description of velocity and concentration fluctuations in steadily sedimenting suspensions is constructed and analyzed using self-consistent and renormalization-group methods. We find a nonequilibrium phase transition from an “unscreened” phase in which we recover the Caffisch-Luke [Phys. Fluids **28**, 759 (1985)] divergence of the velocity variance to a “screened” phase where the fluctuations have a finite correlation length depending on the volume fraction  $\phi$  as  $\phi^{-1/3}$ , in agreement with Segrè *et al.* [Phys. Rev. Lett. **79**, 2574 (1997)] (if their observation of a  $\phi$ -independent diffusivity is used), and the velocity variance is *independent* of system size. [S0031-9007(98)08015-6]

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Sedimentation [1] is a rich and complex phenomenon in suspension science and a frontier problem in nonequilibrium statistical mechanics. The average sedimentation speed  $v_{\text{sed}}$  of solute particles drifting down in a solvent is determined by balancing the driving force (gravity) against the dissipative force (viscous drag). Giant non-thermal fluctuations in the velocity and concentration fields in a steadily settling suspension, observed even for non-Brownian systems, have been a puzzle for some years. Caffisch and Luke (CL) [2] showed, for steady sedimentation in a container of smallest linear dimension  $L$ , that the assumption of purely *random* local concentration fluctuations led to velocity fluctuations with a variance  $\langle v^2 \rangle \sim L$ . Most experiments, however, find *no* dependence of  $\langle v^2 \rangle$  on  $L$  [3–5], although Ladd’s simulations [6] and the data of Tory *et al.* [7] appear to be consistent with CL.

In this Letter we propose a resolution of this puzzle by means of a set of coarse-grained, fluctuating nonlinear hydrodynamic equations for the long-wavelength dynamics of concentration and velocity fluctuations in a suspension settling steadily in the  $-z$  direction, at vanishingly small Reynolds number. Our theory is similar in spirit to the Koch-Shaqfeh (KS) [8] “Debye-like” screening approach but differs in several important details and predictions.

The central conclusion of our study is that there are *two* qualitatively distinct nonequilibrium phases for a sedimenting suspension. In the “unscreened” phase  $\langle v^2 \rangle$  diverges as  $L$ , as in CL and, in addition, concentration fluctuations with wave vector  $\mathbf{k} = (\mathbf{k}_{\perp}, k_z)$  relax at a rate  $\propto k^{1/2}$ . The “screened” phase is characterized by a *correlation length*  $\xi$  similar to that predicted by KS such that  $\langle v^2 \rangle \sim L$  for  $L \ll \xi$  and  $\langle v^2 \rangle \sim \xi$  for  $L \gg \xi$ . Deep in the screened phase we predict  $\xi \sim \phi^{-1/3}$ , where  $\phi$  is the particle volume fraction. This is in agreement with the experiments of Segrè *et al.* [5], but not with KS [8]. The relaxation rate in the screened phase is *independent* of  $k$  for  $k_z = 0$  and  $\mathbf{k}_{\perp} \rightarrow \mathbf{0}$ . Detailed, experimentally

testable expressions for the structure factor and velocity correlations in the screened phase are presented after we outline our calculations. The two phases are separated in our “phase diagram” (Fig. 1) by a striking *continuous nonequilibrium phase transition* where  $\xi$  diverges at least as rapidly as  $(K - K_c)^{-1/3}$  as a control parameter  $K$  is decreased towards a critical value  $K_c$ .

The hydrodynamic equations we used to arrive at these results are

$$\frac{\partial c}{\partial t} + \mathbf{v} \cdot \nabla c = [D_{0\perp} \nabla_{\perp}^2 + D_{0z} \nabla_z^2]c + \nabla \cdot \mathbf{f}(\mathbf{r}, t) \quad (1)$$

and

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

where  $c(\mathbf{r}, t)$  and  $\mathbf{v}(\mathbf{r}, t)$  are the fluctuations about the mean concentration  $c_0$  and the mean sedimentation velocity  $-v_{\text{sed}} \hat{z}$ , respectively. We justify these equations briefly below; for a more detailed discussion we refer the reader to Ref. [9]. Equation (1) is the anisotropic randomly forced advection-diffusion equation with bare uniaxial diffusivities ( $D_{0z}, D_{0\perp}$ ) and a random stirring force  $\mathbf{f}(\mathbf{r}, t)$  [10]. The Stokes equation, Eq. (2), which expresses the balance between the driving by gravity and the dissipation by the viscosity  $\eta$ , describes how the concentration fluctuations produce velocity fluctuations. Here  $m_R g$  is the buoyancy-reduced weight of a particle, while the pressure field has been eliminated by imposing incompressibility via the transverse projection operator  $P_{ij} = \delta_{ij} - \nabla_i \nabla_j (\nabla^2)^{-1}$ .

Hydrodynamic equations such as Eqs. (1) and (2) arise from a coarse graining of the microscopic equations of motion. The latter, for the main case of interest here, viz., non-Brownian suspensions at zero Reynolds number, are the deterministic equations of Stokesian dynamics for  $N$  hydrodynamically coupled particles and are known to be chaotic [11]. The noise or random stirring current  $\mathbf{f}(\mathbf{r}, t)$

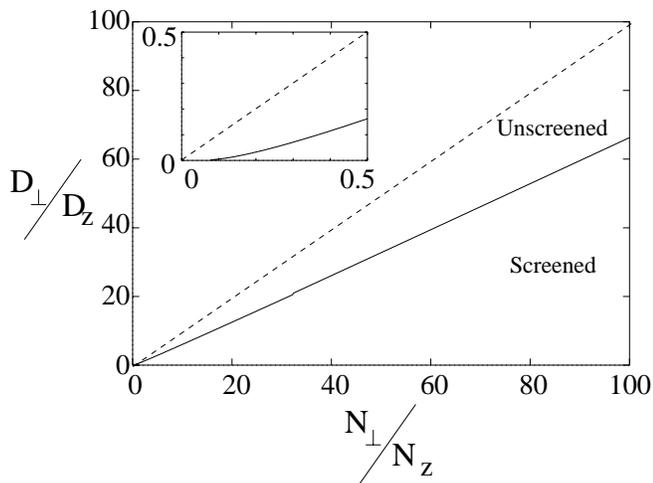


FIG. 1. Dynamical phase diagram for sedimentation. Below the solid line in the parameter space spanned by the anisotropy factors for the noise and diffusivities velocity and concentration fluctuations have a finite screening length in the limit of vanishing wave vector. This region is called screened above. In the upper region called unscreened the screening length becomes infinite. The dashed line represents the set of values for the anisotropy factors where the hydrodynamics obeys detailed balance. The inset shows the behavior of the phase boundary in the limit of large noise and diffusivity in the vertical direction as compared to the horizontal plane.

and the diffusivities in Eq. (1) represent a phenomenological description of the deterministic chaos at length scales below the coarse-graining length  $\ell$  (which must be large compared to the particle radius  $a$ ). We use these hydrodynamic equations to predict the velocity and concentration fluctuations at length scales large compared to  $\ell$  driven by the random stirring at short distances.

We assume, as is reasonable, that  $\mathbf{f}(\mathbf{r}, t)$  is Gaussian white noise with uniaxial symmetry:

$$\langle f_i(\mathbf{r}, t) f_j(\mathbf{r}', t') \rangle = 2c_0 N_0^{ij} \delta(\mathbf{r} - \mathbf{r}') \delta(t - t') \quad (3)$$

with an anisotropic noise amplitude  $N_0^{ij} = N_{0\perp} \delta_{ij}^\perp + N_{0z} \delta_{ij}^z$ , where  $\delta_{ij}^z$  and  $\delta_{ij}^\perp$  are the projectors along and normal to the  $z$  axis, respectively. Because of the non-equilibrium origin of the noise and diffusion constants, we may not [12] assume that  $N_{0\perp}/N_{0z} = D_{0\perp}/D_{0z}$  as would be true for the Langevin equation of a dilute suspension at thermal equilibrium. Note that no correlations have been fed in via the noise: any that emerge in the long-wavelength properties are a result of the interplay of advection and diffusion.

Let us now consider the nature of the spatiotemporal correlations implied by Eqs. (1) and (2). We will focus on the structure factor for concentration fluctuations

$$S(q) \equiv c_0^{-1} \int d^d r \langle c(\mathbf{0}) c(\mathbf{r}) \rangle e^{-i\mathbf{q}\cdot\mathbf{r}} \quad (4)$$

from which the velocity structure factor can be derived through Eq. (2). If we ignore the advective nonlinearity  $\mathbf{v} \cdot \nabla c$ , then  $S(\mathbf{q})$  can be computed by straightforward

Fourier transformation of Eq. (1), resulting in

$$S(\mathbf{q}) = S_0(\mathbf{q}) \equiv \frac{N_{0\perp} q_\perp^2 + N_{0z} q_z^2}{D_{0\perp} q_\perp^2 + D_{0z} q_z^2}. \quad (5)$$

Using Eq. (5) in Eq. (2) we can compute  $\langle v^2 \rangle$  as a function of the system size  $L$  with the result

$$\langle v^2 \rangle \sim \int_{q>1/L} \frac{S(\mathbf{q})}{q^4} \sim L. \quad (6)$$

In other words, neglecting large-scale advection by the velocity fluctuations leads to the CL [2] result.

To include the effect of the advective nonlinearity we have performed a self-consistent mode coupling calculation [13] on Eqs. (1)–(3). Our results can be expressed in terms of a *renormalized* relaxation rate

$$R(\mathbf{q}) = D_\perp(\mathbf{q}) q_\perp^2 + D_z(\mathbf{q}) q_z^2 + \Gamma(\mathbf{q}) \quad (7)$$

and a *renormalized* structure factor of the form

$$S(\mathbf{q}) = \frac{N_\perp(\mathbf{q}) q_\perp^2 + N_z(\mathbf{q}) q_z^2}{R(\mathbf{q})}. \quad (8)$$

The quantities  $D_{z,\perp}(\mathbf{q})$  and  $N_{z,\perp}(\mathbf{q})$  represent renormalized diffusivities and noise amplitudes [14]. But, most importantly, the advective nonlinearity to lowest-order perturbation theory leads to an additional term in the renormalization of the relaxation rate which is of the form  $\Gamma(\mathbf{q}) = \gamma(\mathbf{q}) q_\perp^2 / q^2$ . The singular form of this correction to the diffusivity is due to the long-ranged nature of the hydrodynamic interaction. Starting from the stochastic hydrodynamic equations, Equations (1)–(3), it turns out that the amplitude of this singular contribution becomes a constant,  $\lim_{q \rightarrow 0} \gamma(\mathbf{q}) \propto I(\beta_N, \beta_D)$ , which depends on the anisotropy ratios of the noise and diffusivity coefficients

$$\beta_N = \frac{N_\perp}{N_z}, \quad \text{and} \quad \beta_D = \frac{D_\perp}{D_z}. \quad (9)$$

In particular  $I(\beta_N, \beta_D)$  is proportional to  $\beta_N - \beta_D$  and consequently may change sign upon varying the noise and diffusivity ratios. We understand the above proportionality of  $I$  (setting  $D_z = D_\perp$  for simplicity) by looking at the effect of a noise-injected concentration fluctuation (NICF) on an imposed concentration inhomogeneity. If  $N_z \gg N_\perp$  [see Eq. (3)], the typical NICF varies predominantly along  $z$  and the induced  $z$  velocity alternates in sign mainly along  $z$  thus reinforcing the inhomogeneity. However, if  $N_\perp \gg N_z$ , the NICF and hence the sign of the resulting  $z$  velocity both vary mainly in the  $xy$  plane, so that the flow breaks up the inhomogeneity and thus enhances effective diffusion.

We restrict our attention to  $I(\beta_N, \beta_D) \geq 0$ , for which the model can be treated either within dynamic renormalization group theory or using self-consistency methods, although it is possible that the model itself is meaningful outside that regime.

We start our discussion at the borderline of stability,  $\beta_N = \beta_D$ . For these parameter values it can be shown that the fluctuating hydrodynamic equations describe a dynamics which obeys detailed balance [15]: the advective nonlinearity does not affect the equal-time correlations, and  $S(\mathbf{q})$  in particular is just the constant  $N_\perp/D_\perp$ . There are singularities in  $N_{\perp,z}$  and  $D_{\perp,z}$  which we discuss later.

For  $\beta_N \geq \beta_D$ , detailed balance is violated and a singular diffusion term  $\Gamma(\mathbf{q})$  is generated within perturbation theory. In order to analyze the dynamics in this regime we use one-loop self-consistent theory (mode coupling theory) and arrive at the expression

$$\Gamma(\mathbf{q}) = c_0 \left( \frac{m_R g}{\eta} \right)^2 \int_k \frac{q_i P_{iz}(\mathbf{k}) k_j P_{jz}(\mathbf{q})}{k^2 q^2} \times \frac{[S(\mathbf{q} - \mathbf{k}) - S(\mathbf{k})]}{R(\mathbf{k}) + R(\mathbf{q} - \mathbf{k})} \quad (10)$$

with  $R(\mathbf{q})$  given by (7), and similar self-consistent integral equations for  $D_\perp(\mathbf{q})$ ,  $D_z(\mathbf{q})$ ,  $N_\perp(\mathbf{q})$ , and  $N_z(\mathbf{q})$ . We find that there are two types of iteratively stable solutions to these coupled self-consistent equations: those with  $\gamma(q \rightarrow 0) > 0$ , which we obtain below the solid line in the phase diagram spanned by the two anisotropy ratios (screened phase in Fig. 1), and those with  $\gamma(q \rightarrow 0) = 0$ , which arises for values of the anisotropy parameters that lie above the solid line and below the dashed line of the same figure, i.e., in the unscreened phase.

*Screened phase.*—In the screened phase,  $\Gamma(\mathbf{q})$  is of the form  $\gamma q_\perp^2/q^2$  in the small  $q$  limit, with  $\gamma$  a finite constant. This implies that the structure factor at a small wave number becomes

$$S(\mathbf{q}) \simeq \frac{N_\perp q_\perp^2 + N_z q_z^2}{D_\perp q_\perp^2 + D_z q_z^2 + \gamma q_\perp^2/q^2} \quad (11)$$

with  $N_{\perp,z}$  and  $D_{\perp,z}$  constants. From Eq. (11) we can define a correlation length  $\xi \equiv (D_\perp/\gamma)^{1/2}$  such for  $q_\perp \gg 1/\xi$  the structure factor is not significantly affected by advection. On the other hand, for  $q_\perp \ll 1/\xi$  the in-plane structure factor reads  $S(\mathbf{q}_\perp, q_z = 0) \simeq (N_\perp/\gamma)q_\perp^2$ , while  $S(\mathbf{q}_\perp = 0, q_z) \simeq (N_z/D_z)$ . Physically, this means that at long wavelength advection strongly suppresses in-plane concentration fluctuations.

Using Eq. (11) in conjunction with Eq. (2), one finds that for length scales  $L$  less than  $\xi$ ,  $\langle v^2 \rangle \propto L$ , consistent with CL, while for  $L$  large compared to  $\xi$ ,  $\langle v^2 \rangle \propto \xi$ . Velocity fluctuations on length scales small compared to  $\xi$  are thus highly correlated while they become uncorrelated at larger length scales.

Deep inside the screened phase, i.e., for large  $\gamma$ , the renormalization of the diffusion and noise parameters is negligible and we can explicitly compute  $\gamma$ , and thus  $\xi$ , by inserting Eq. (8) in Eq. (10) using the bare values for the  $N$ 's and  $D$ 's. We find

$$\xi = 8 \left( \frac{m_R g}{\eta D} \right)^{-2/3} c_0^{-1/3} \left( 1 - \frac{2}{\beta_N} \right)^{-1/3}, \quad (12)$$

where for simplicity we have set  $D_{0\perp} = D_{0z} = D$ . According to Eq. (12), the correlation length increases as we decrease the  $\beta_N$  parameter (which could be done by increasing the *thermal* noise amplitude) and diverges at  $\beta_N = 2$ . Strictly speaking, as  $\beta_N \rightarrow 2$ , the diffusivity corrections are no longer negligible, and the actual divergence of  $\xi$  is probably stronger than (12) and occurs at a larger value of  $\beta_N$ . An explicit analytical (but lengthy) result for the correlation length  $\xi$  can be obtained throughout the screened phase as a function of both anisotropy parameters [9] and the phase boundary can also be computed. The phase boundary resulting from this result is shown in Fig. 1 as the solid line separating the screened from the unscreened phases.

*Unscreened phase.*—The hydrodynamic equations obey detailed balance [15] along the dashed line  $\beta_N = \beta_D$  in Fig. 1. As a consequence the ratio of noise to diffusivity can be identified as a direction-independent “noise temperature.” Furthermore, the structure factor  $S(\mathbf{q})$  becomes a constant  $D_\perp/N_\perp$  and we recover the CL result. In conjunction with an exponent identity resulting from Galilean invariance this is enough to determine the dynamic exponent exactly,  $z = d/2 - 1$ . This implies that the diffusivities and noise amplitudes scale as  $q^{-\epsilon/2} = q^{-3/2}$  for long wavelength. Even though there are now singular corrections to  $D_{z,\perp}(\mathbf{q})$  and  $N_{z,\perp}(\mathbf{q})$ , the anomalous  $\Gamma(\mathbf{q})$  term is zero. For parameter values in the regime between the dashed line (detailed balance line) and the solid line, which marks the location of the nonequilibrium phase transition, renormalization group methods may be used to determine the renormalization of the noise and diffusivity amplitudes. In view of the results from the above self-consistency calculation ( $\gamma = 0$  in the unscreened phase) and the exact results at the detailed balance line it is quite likely that the resulting renormalization group flow will tend towards a fixed point which obeys detailed balance. We leave the details of such an investigation for a future publication [9].

The analysis of our hydrodynamic equations thus confirms that screening can suppress the CL divergence of  $\langle v^2 \rangle$  with  $L$ , as argued by KS, while it allows for a second, unscreened phase. This result may help explain the conflicting results on  $\langle v^2 \rangle$  obtained by different workers [3–7]. The self-consistent structure factor Eq. (5) we obtained differs significantly from the one proposed by KS. Experimental tests will thus be of considerable importance. Measurements of  $S(\mathbf{q})$ , for example, by particle imaging velocimetry [16], would constitute the most direct test of the theory since our prediction that  $S(\mathbf{q}_\perp, q_z = 0) \propto q_\perp^2$  does not hold in the KS description. Detailed measurements of  $S(\mathbf{q})$  for sedimenting solutions are not yet available. However, Segrè *et al.* [5] do report that the size dependence of the amplitude  $\langle v^2 \rangle$  of the velocity fluctuations depends on a characteristic length scale  $\xi_S$  such that  $\langle v^2 \rangle \propto \xi_S$  for length scales  $L \gg \xi_S$  while for  $L \ll \xi_S$ ,  $\langle v^2 \rangle$  grows with  $L$ . They report that  $\xi_S \sim a\phi^{-1/3}$  with  $\phi$  the particle volume fraction.

Our correlation length  $\xi$ , in Eq. (12), has the same physical interpretation as  $\xi_S$ . Deep in the screened phase, i.e., for  $I(\beta_N, \beta_D) \gg 0$ ,  $\xi$  can be written as

$$\xi(\phi) \sim (m_{RG}/\eta D)^{-2/3} a \phi^{-1/3} I(\beta_N, \beta_D)^{-1/3}. \quad (13)$$

On scaling grounds, we expect that  $D \propto \delta v_{\text{rms}} \xi$  with  $\delta v_{\text{rms}}$  the root mean square of the velocity field fluctuations. Experimentally,  $\delta v_{\text{rms}} \xi$  is found to be independent of volume fraction  $\phi$ . In that case, Eq. (13) reproduces the experimentally observed volume-fraction dependence, in contrast to KS [8]. It should be noted that this volume fraction dependence of the correlation length implies that there is a fixed number of colloids within a correlation volume independent of volume fraction.

The observation of a transition from the screened to the unscreened phase would obviously be the most conclusive evidence supporting our theory, in particular, if the transition were accompanied by a divergence of the velocity fluctuation correlation length. Even in the absence of such direct evidence, the observation of screened behavior combined with our theory requires that the anisotropies in the noise and diffusivity lie in the lower region of our dynamical phase diagram, Fig. 1. A complete test of our theory thus requires measurement of the  $N$  and  $D$  parameters. These could be obtained from the measurement of the steady-state static structure factor  $S(\mathbf{q})$ , both along the  $z$  direction and in the  $x$ - $y$  plane, coupled with tracer diffusion measurements.

Finally, it would be interesting to vary the effective noise and diffusion constants in a controlled manner in an experiment. Despite the absence of a microscopic theory it is clear that at zero Reynolds number the parameter  $K$  can depend only on Peclet number  $Pe$  and particle shape [17]. Decreasing  $Pe$  (i.e., increasing the role of isotropic thermal diffusion) by reducing the mass-density difference of between particles and solvent is likely to drive the system into the unscreened phase. Thus by repeating the experiments of Segré *et al.* [5] with colloids that are more nearly density matched to the solvent one could test our prediction of a transition to an unscreened phase.

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