Non-Gaussian Dynamics in Quasi-2D Noncolloidal Suspensions

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We study experimentally the hydrodynamic-interaction-controlled random walk (RW) of noncolloidal spheres in a quasi-2D fluidized suspension. This macroscopic "molecular" system is anisotropic. Particle trajectories are Brownian-like and, from time to time, fast upward correlated. The vertical RW is hyperdiffusive, but the horizontal one is Gaussian. Velocity fluctuation distributions vary from Gaussian to exponential as particle concentration C increases. The dynamics varies from liquidlike to gaslike as C decreases. These features are attributed to the formation of "transient channels" at higher C.

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Complex fluid flows are typically associated with inertial effects, as, for example, in turbulent and granular material flows. The trajectories in such flows are often chaotic, and can lead to anomalous random walks, such as the celebrated Levy walk [1]. Slow, viscous flows can also be complex, however, as a result of the structure in which the flows occur: porous media or suspensions are two such examples. In porous media, the flow reflects the frozen disorder of the pore space. In suspensions, fluid and particles interact in continuously changing geometrical configurations, as a result of their relative motion. In some way, suspensions can be viewed as fluctuating porous media, in which the fluid flow adapts to the instantaneous porous medium created by the particle configuration, and at the same time, induces a change of this configuration.

In recent numerical simulations [2,3] in 2D and 3D porous media, it was found that flows in low-porosity porous media form stationary, localized, channelized structures. These focused flow patterns result in long-tailed stretched exponential probability distribution functions (PDF) for the velocity fluctuations [2,4]. They were also experimentally confirmed using nuclear magnetics resonance [5,6]. In noncolloidal suspensions of monodisperse spherical particles in a viscous liquid, the particle velocity is determined by the instantaneous configuration of all other particles. As a result of the long-range hydrodynamic interaction, the spatial distribution of the particles is not stationary and the particles experience a permanent stochastic motion, or random walk (RW), the so-called hydrodynamic dispersion [7–11]. So far, theoretical efforts have addressed issues of the particles mean square velocity fluctuations [7,10]. However, there are no predictions on their PDFs.

The statistical description of complex flows has proved to be a powerful tool in understanding their main features. For example, the complexity of turbulent and granular material flows can be captured by analyzing the moments and correlation functions of the velocity fluctuations [12,13]. Likewise, various statistical approaches (determining mean velocity and mean square velocity fluctuations) have been applied for the understanding of porous media and suspensions [2,14,15]. In this Letter, we present experimental results for the PDFs of the velocity fluctuations and for the properties of the RW of monodisperse spherical particles in a quasi–2D suspension. For this purpose, we use a counterflow stabilized suspension [8], namely a liquid-solid-fluidized bed.

To follow the suspension dynamics and the particle trajectories reliably and extensively, we designed a transparent quasi-2D fluidized bed. The bed consists of a transparent vertical, Hele-Shaw cell, filled with spherical particles, just slightly smaller than the gap of the cell, which are kept suspended by an upward uniform flow of a viscous fluid. We record the trajectories and velocities of all the particles. Various particle concentrations, up to close packing, can be studied. Our data conclusively show the following: (i) the velocity fluctuation PDFs are stretched exponential, with concentration dependent exponents; (ii) the particle RW is anisotropic, namely Gaussian diffusive along the horizontal, but hyperdiffusive along the vertical direction; and (iii) the particle system exhibits a change from gaslike to liquidlike behavior as the concentration increases. The hyperdiffusion can be understood as a kind of Levy walk on nonstationary channelized structures. The results suggest an analogy between porous media and 2D suspension flows, which will be discussed below. More generally, our experiment can be viewed as a macroscopic "fluidlike" system, in which the "molecules" are the spherical particles, and where we can study the "fluid" at the scale of individual molecules, along with the resulting "pseudoturbulence" of particles in laminar flow.

Experimental.—The Hele-Shaw cell consists of two parallel glass plates of length $L = 80$ cm and width $W = 10$ cm, separated by a uniform spacer which ensures a constant gap of thickness $b = 2.0$ mm. The cell is held vertically along the larger dimension. The monodisperse spherical particles are roller bearings made of aluminum

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or brass (of relative density 2.7 or 8.7, respectively), with a diameter $2a = 1.50$ mm. The injected fluid is a water-glycerin mixture, with density 1.25 and kinematic viscosity ν ranging between 2.5 and 5 10^{-4} $\mathrm{m^2\,s^{-1}}$. At such conditions, the sedimentation velocity of a single sphere is $V_s \sim 1$ $\mathrm{cm\,s^{-1}}$, implying a small corresponding Reynolds number, $\mathrm{Re} = aV_s/\nu < 10^{-1}$. Hence, the flow is viscous and far away from any kind of inertial turbulence. Under the low Re number condition, the bed exhibits a stable uniform expansion [8,14]. As is well known, the bed expansion, and the resulting mean particle concentration, are controlled by the upward fluid velocity V_d. We note that in the present 2D fluidized bed, one can use the area fraction, C, as a measure of the particle concentration. In the experiments, this fraction was varied in the range between 8% and 76%, with maximum packing near $C \sim 80\%$, which is close to the maximum packing of well arranged disks [16].

During the process, the particles are in constant motion, participating from time to time in the formation of doublets, triplets, etc. To record particle trajectories and velocities, the entire bed is illuminated and a CCD videotape camera records the movements of each of the 2000 beads. Each image is digitized and the position of the illuminated centers of all particles is recorded using NIH software. By tracing each particle between consecutive frames, we can record trajectory and instantaneous velocities of all 2000 particles (Fig. 1), with an accuracy of 0.1 mm in position and 5% in velocity. Figure 1a shows a snapshot of the fluctuating structure, which consists partly of instantaneous swirls, analogous to those recently observed in sedimenting dilute suspension experiments [11]. One notes that the vectors of the larger velocities typically point upwards. Figure 1b shows two trajectories, corresponding to two different particles.

Velocity PDF.—Using our technique we measured all velocity components in the fluidized bed. As the particles are stationary on average, the velocities observed also represent velocity fluctuations. The mean square horizontal and vertical velocity components have the same value (within 5%), denoted u_o. Thus, the quasi-2D suspension reveals more isotropy than in 3D, where the ratio between vertical and horizontal velocity fluctuations has been reported to be larger than 2 [9,10]. Figure 2a shows in a log-linear plot the PDFs of the vertical velocity fluctuations. From our statistics (involving more than 5×10^5 values over 4 decades), the velocity PDFs can be fitted reasonably well with a stretched exponential (solid lines in Fig. 2a), namely

$$P(u/u_o) \sim \exp[-\beta(|u|/u_o)^{\xi(C)}], \qquad (1)$$

where the exponent $\xi(C)$ varies between 1 and 2 as C decreases in the range of Fig. 2b. We recall that the Gaussian value 2 corresponds to a Maxwell-Boltzmann distribution, as in the kinetic theory of a perfect gas. The PDFs of the horizontal velocity are left-right symmetric regardless of the concentration, and evolve from nearly Gaussian ($\xi = 2$) at low C to almost exponential ($\xi = 1$) at high C. At low concentrations, the PDFs of the vertical velocity tend to be also symmetric (up-down symmetric). When the concentration increases, however, the vertical PDFs become asymmetric: they are Gaussian for the downward velocity and stretched exponential

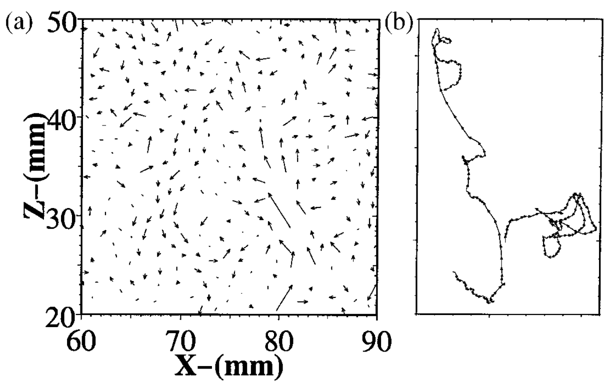

FIG. 1. (a) A snapshot of particle velocities. (b) Two different particle trajectories (scale in mm, $C = 54\%$).

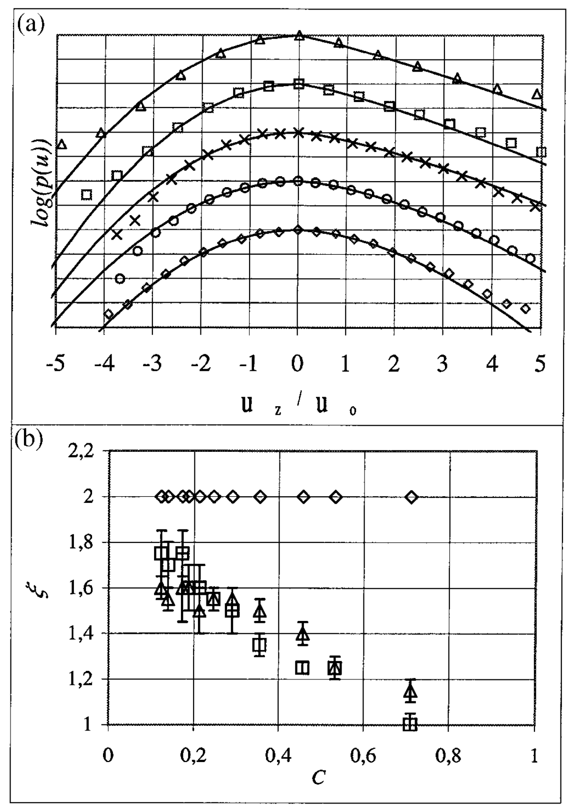

FIG. 2. (a) Probability distribution function of vertical velocity components at various concentrations [$C = 12(\diamond), 22(\circ), 36(\times), 54(\square), 70(\triangle)\%$] in a log-linear plot (grid space: one decade). The solid curves are best-fit stretched-exponentials with exponent ξ. (b) Concentration dependence of the exponent ξ [left and right velocity ($\triangle$), downward velocity exponent ($\diamond$), upward velocity exponent ($\square$)].

for the upward velocity with an exponent equal to the horizontal ones. At large concentrations, our suspension exhibits transient, continuously evolving structures. As shown in Fig. 1a, particles moving downwards have small velocity and organize into large density areas, analogous to clusters. Particles that move upwards are faster and flow along paths analogous to channels in porous media. Whereas the "slow particle" trajectories are Brownian-like (Fig. 1b, right), the "fast particles" exhibit correlated features (Fig. 1b, left). As the upward trajectories of the fast particles also include a horizontal component of motion, they increase the tails of the PDF of both the horizontal and upward velocity, making such PDFs stretched exponentials.

We note that stretched-exponential PDFs have been previously observed in both porous media and in granular flows. In numerical simulations of the fluid velocity fluctuations in porous media [2], lowering the porous medium porosity (which in our case is equivalent to increasing C) led to an increased focusing of the fluid along channellike structures. This focusing resulted in stretched-exponential PDFs. At larger porosity, however, the channelized structure increasingly weakened and the PDF tends to a Gaussian. Non-Gaussian PDFs have been also observed in simulations of inelastic granular media [13] and in experiments with granular monolayers [17]. They were associated with the continuous creation and destruction of clusters of particles. The nearly exponential tails of the PDFs obtained there were attributed to the dynamics dominated by particle-particle interaction [17].

Both analogies with porous media and with granular monolayers can be tentatively applied to our system. For example, the fast particles behave like particles moving from one cluster of slow particles to another one, as in granular flows. At the same time, the fast particles appear more localized than the slow ones, while their upward velocity reveals a strong upward localized fluid flow, as in porous media flows. These features support a tempting analogy between 2D suspensions and porous media: the fast particles could be seen as particles convected by the fast upward eroding flow which takes place in transient localized channels ("fractures"), whereas the slow particles represent the porous media backbone. However, contrary to the porous medium case, there is no sharp transition between the Gaussian and the stretched-exponential regimes (even in the flatness of the PDF [2]); see Fig. 2b.

Random walk.—From the recording of the position of each particle versus time, we can compute the horizontal and vertical mean square displacements. At short times, the mean square displacements in both directions are found to be proportional to t^2. This is the signature of a ballistic regime from which we can obtain the mean square velocity fluctuations: $\overline{V_x^2} \sim \overline{V_z^2} \sim u_0^2$. This regime is observed for all concentrations with no particular scaling for the value of the velocity fluctuations, which varies smoothly from $\sim 0.2 V_d$ at low C to $\sim 0.6 V_d$ at larger C for the two types of beads used (aluminum and brass). To track the diffusive behavior, we plot in Fig. 3 the diffusivity, namely the mean square displacement normalized by twice the time, for an intermediate concentration value ($C = 0.36$). This plot clearly demonstrates the anisotropy of the random walk. The horizontal direction diffusivity reaches a long time plateau which reflects the diffusive nature of this component of the random walk, namely

$$\lim_{t\to\infty} \overline{[x(t) - x(0)]^2} = 2D_x t\,. \tag{2}$$

This is true regardless of the concentration C. On the other hand, the diffusivity of the vertical component keeps on increasing with time, at large times, and up to a traveled distance comparable to the cell size. Here, the nature of the random walk along the vertical direction is hyperdiffusive [18,19] and can be tentatively described as a power law,

$$\lim_{t\to\infty} \overline{[z(t) - z(0)]^2} \sim t^{\alpha}, \quad \text{where } \alpha = 1.25 \pm 0.05\,. \tag{3}$$

This combined behavior is displayed in the trajectories of Fig. 1b. The maximum displacements of the two different particles are of the same order along the horizontal direction, but much different along the vertical one, where one is five times larger than the other. We can infer that the hyperdiffusive behavior is due to the presence of fast particle trajectories. Any given particle experiences at different times a sequence of fast and slow trajectories. This sequence is reminiscent of Levy walks, which also manifest a hyperdiffusive behavior [1]. Again, our quasi-2D suspension appears to compare better with flow in porous media, which can induce hyperdiffusion [18], than with 3D suspensions, which lead to normal diffusion [7,9,10]. It is likely that the particles in our quasi-2D suspension interact more strongly with the walls than with the other particles. As a consequence, they are less "mobile" than in 3D suspensions and constitute a kind of

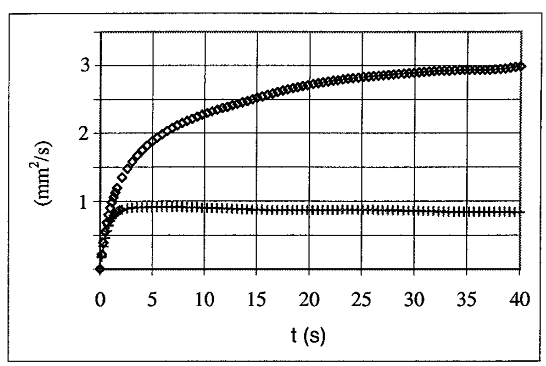

FIG. 3. Horizontal (bottom) and vertical (top) diffusivity (mean square displacements normalized by time) $\overline{[x(t) - x(0)]^2}/2t$ and $\overline{[z(t) - z(0)]^2}/2t$, as a function of time, for $C = 36\%$. The long-time horizontal random walk is diffusive, whereas the vertical one is hyperdiffusive.

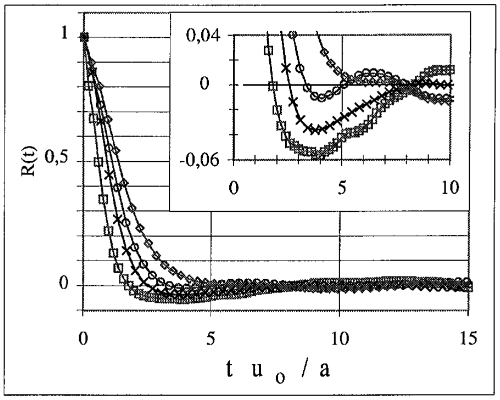

FIG. 4. Time dependence of the horizontal velocity autocorrelation functions at $C = 12(\diamond), 22(\circ), 36(\times), 54(\square)\%$. The inset shows an enlargement around the negative minimum. It reflects the signature of a liquidlike behavior, except at low concentrations, where it vanishes and where the quasi-2D suspension has a gaslike behavior.

porous media, as mentioned previously. Given that their velocity reflects the local velocity of the fluid, they should then exhibit characteristics similar to the fluid velocity in porous media.

The anisotropy of the 2D suspension is also evident in the velocity autocorrelation function (VAF) (which is the second derivative of the mean square displacement [20]: $R(t) = \overline{V(t) * V(0)} = (1/2)d^2\{\overline{[z(t) - z(0)]^2}\}/dt^2$). $R(t)$ computed from our velocity measurement is plotted in Fig. 4. The vertical VAF first decreases as an exponential decay and then breaks into a long time tail which does not reach zero in the range of accessible time (here limited by the cell boundary), as expected from the vertical RW. The horizontal VAF decreases rapidly, becomes negative, and then tends to zero from below at large times (Fig. 4). This negative minimum is usually identified as the signature of a liquid structure [20]. It is deeper at large concentrations and disappears at low ones (inset of Fig. 4). This is also consistent with the fact that at low concentrations the velocity PDFs tend to be Gaussian symmetric. These two remarks support the contention that our particle system exhibits a gas-like behavior at low concentrations. Note that 3D simulations of suspensions did not show such a gaslike limiting state [10].

In conclusion, we have measured the dynamics, namely the velocity PDF, the RW, and the VAF, of a quasi-2D suspension of monodisperse macroscopic particles. The particles execute a random walk, which is increasingly anisotropic as the concentration increases, with the horizontal component always being Gaussian. In the limit of small C, the behaviour of this macroscopic "molecular" system is isotropic and gaslike. As C increases, fast and slow trajectories along the vertical direction, analogous to a Levy walk, lead to stretched-exponential PDFs and hyperdiffusion. The vertical component PDFs are more asymmetric, as C increases. These features are attributed to dynamic localized channelized structures analogous to the static ones observed in low-porosity porous media.

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