Diffusion and Mixing in Gravity-Driven Dense Granular Flows

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(Received 8 August 2003; published 27 April 2004)

We study the transport properties of particles draining from a silo using imaging and direct particle tracking. The particle displacements show a universal transition from superdiffusion to normal diffusion, as a function of the distance fallen, independent of the flow speed. In the superdiffusive (but sub-ballistic) regime, which occurs before a particle falls through its diameter, the displacements have fat-tailed and anisotropic distributions. In the diffusive regime, we observe very slow cage breaking and Péclet numbers of order 100, contrary to the only previous microscopic model (based on diffusing voids). Overall, our experiments show that diffusion and mixing are dominated by geometry, consistent with long-lasting contacts but not thermal collisions, as in normal fluids.

DOI: 10.1103/PhysRevLett.92.174301 PACS numbers: 45.70.Mg, 66.30.–h

Granular flow is an attractively simple and yet surprisingly complex subject [1]. Fast, dilute flows are known to obey classical hydrodynamics (with inelastic collisions), but slow, dense flows pose a considerable challenge to theorists, due to many-body interactions and nonthermal fluctuations. Beyond their fundamental scientific interest, such flows have important engineering applications [2], e.g., to new pebble-bed nuclear reactors [3], whose efficiency and safety depend on the degree of mixing in very slow granular drainage (<1 pebble/ min).

Although dense granular drainage is very familiar (e.g., sand in an hourglass), it is far from fully understood. Over the past 40 years, a number of theoretical approaches have been proposed for steady state flow [4–8]. Continuum approaches are based on the critical-state theory of soil mechanics and yield only mean velocity fields [7,8]. On the other hand, the diffusing void model [4,5] takes a particle approach, in which "voids" injected at the orifice cause drainage by diffusing upward and exchanging position with particles along the way. Averaging over the void trajectories yields the same continuum velocity field for particles as the "kinematic model" [6,8], which provides a reasonable fit to experimental data with only one fitting parameter (the diffusion length b) [5,6,9,10], although the void model on a regular lattice (as in Ref. [11]) underpredicts its value [12]. Remarkably, these models depend only on *geometry* and not on momentum, energy, etc.

In spite of the success of the kinematic model, however, its only microscopic basis, the void model, greatly overpredicts diffusion. To see this, consider the Péclet number, $\mathrm{Pe}_x = v_z d/D_x$, the dimensionless ratio of advection in uniform downward flow of speed v_z to diffusion with a horizontal diffusivity D_x at the scale of a particle diameter d. In the void model, when a particle falls a distance Δh, $\mathrm{Pe}_x = (\Delta h/\Delta t)d/(\langle \Delta x^2\rangle/2\Delta t) = \Delta h 2d/\langle \Delta x^2\rangle$ is of the order of 1 for any conceivable packing since $\Delta h \approx \Delta x \approx d$, and therefore it diffuses horizontally by roughly $\sqrt{\Delta h}$. This prediction is contradicted by everyday experience and our experiments below, which exhibit far less mixing. An attempt to resolve this paradox with a new model appears in a companion paper [12].

In this Letter we describe particle-tracking experiments on silo drainage using similar techniques as in a recent (lower-resolution) study of the velocity field [13]. We focus on the statistical evolution of particle displacements and topological "cages," which should aid in developing new microscopic models. Our data may also have implications for recent attempts to apply thermodynamic approaches from glassy dynamics to granular flows [14–16]. Although we do not define a "granular temperature," we observe the presumably related effect of varying the flow rate in all of our measurements.

Our experimental apparatus involves glass beads ($d = 3.0 \pm 0.1$ mm) in a quasi-two-dimensional silo

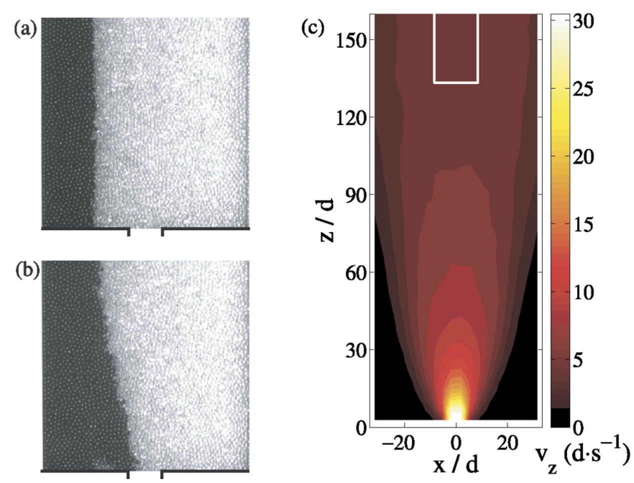

FIG. 1 (color online). An initially flat, off-center interface between two regions of differently colored beads (a) stretches and roughens after draining half of the silo (b), but little mixing is observed. (c) Contour plot of the average downward velocity field v_z with an orifice width, $W = 16$ mm $= 5.3d$. The white box indicates a region of nearly uniform flow where all subsequent measurements are made.

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($20.0 \times 90.0 \times 2.5$ cm $= 67 \times 300 \times 8.3d$). The particles are observed near the front wall made of transparent glass, where the slight polydispersity reduces the tendency for hexagonal packing. (As seen in Fig. 1, there is no long-range order, although the wall induces some short-range order that may affect our results.) We find that varying the thickness of the silo has insignificant effect on the diffusion properties and therefore we report our data for a single thickness. We track individual particles using a high-speed digital camera with a maximum resolution of 512×1280 pixels at 1000 frames/s. Particle positions are obtained to subpixel accuracy using a centroid technique ($\pm 0.003d$, $d = 15$ pixels).

Our first experiment provides a visual demonstration that particles mix much less than predicted by the void model. We load the silo with black and white (but otherwise identical) glass beads, forming two separate columns, as shown Fig. 1(a). From Fig. 1(b), where half of the particles have drained (in 30 s), it is clear that the black-white interface has not smeared significantly, although it has roughened. The small degree of mixing is consistent with the segregation of bidisperse beads in a similar apparatus [10].

The mean downward velocity field v_z is shown in Fig. 1(c) throughout the silo, as measured by direct particle tracking. A distributed filling procedure similar to that in Ref. [17] was used to add grains to the silo. Then the orifice was opened and steady state flow was allowed to develop before acquiring the data used for subsequent analysis. The flow speed is highest at the center line, with a maximum near the orifice, and decays to zero toward the sides. Fairly good agreement with the kinematic model is obtained with $b = 1.3d$.

In order to investigate particle dynamics in a simple setting, we focus on a small $17 \times 87d$ region of nearly uniform flow [the white box in Fig. 1(c)]. For the measurements below, we track about 1370 particles through this window for 4 s in steps of 1 ms. The average flow speed v_z, the only control parameter in this study, is varied by changing the width of the orifice W. The flow is fairly smooth for $W \geq 8$ mm (about $3d$) and nearly continuous for $W \geq 16$ mm. For simplicity, we vary W in the range $8 \leq W \leq 32$ mm, in increments of 4 mm, to avoid the complicated regime of intermittent flow [18]. This corresponds to $1.38 \leq v_z \leq 18.39d$/s, and data is consistent with $v_z \propto (W - d)^{1.5}$. We compile statistics from all tracked particles in six experiments per flow speed (except two for $W = 32$ mm).

From the positions of the particles sampled at 1 ms intervals, we calculate the horizontal and vertical displacements, Δx and Δz, relative to a frame moving with the mean speed of the flow. A typical trajectory computed in this way in Fig. 2(a) shows periods of small fluctuations with occasional, much larger steps. This suggests that the probability density functions (PDFs) for Δx and Δz (for $\Delta t = 1$ ms) should have fat tails compared to a Gaussian, which is confirmed in Figs. 2(b) and 2(c).

Fat-tailed PDFs have also been observed in colloidal glasses and attributed to cage breaking [19], but a special feature here is the asymmetry of the PDF for Δz in Fig. 2(c). Downward fluctuations ($\Delta z < 0$) are larger than upward ($\Delta z > 0$) and horizontal (Δx) fluctuations. We attribute this to the fact that particles are accelerated downward by gravity while being scattered in other directions by dissipative interactions with neighbors.

Looking again at Fig. 2(a), it seems that the large fluctuations in particle displacements would be reduced by coarse graining in time, perhaps enough to recover standard Gaussian statistics. Indeed, as shown in Fig. 2(d), the normalized kurtosis, $\kappa_x = \langle \Delta x^4 \rangle / 3\langle \Delta x^2 \rangle^2 - 1$, which measures how much the shape of the distribution of Δx differs from a Gaussian, decreases toward zero as $v_z \Delta t$ increases. (The data fluctuates somewhat

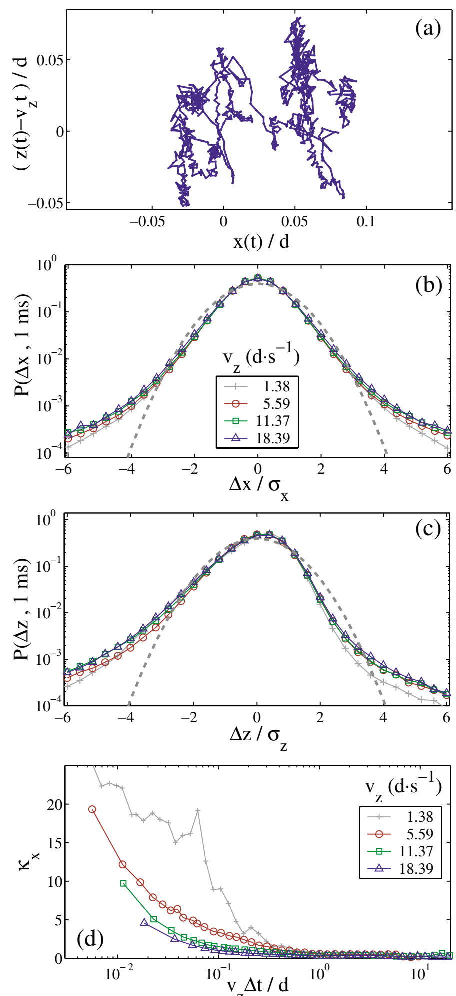

FIG. 2 (color online). (a) A typical particle trajectory sampled at 1 ms intervals in a frame moving with the average flow speed v_z. (b),(c) Normalized PDFs for the 1 ms particle displacements, Δx and Δz, for various flow speeds v_z compared to a standard Gaussian distribution (dotted line); standard deviations, σ_x and σ_z, are of order $10^{-3}d$. (d) The kurtosis of Δx versus the mean distance fallen, $v_z \Delta t$.

for $v_z = 1.38d/\mathrm{s}$, presumably due to intermittency.) This suggests a transition from super to normal diffusion.

As shown in Figs. 3(a) and 3(b), the scaling of the mean-square displacements does, in fact, change from superdiffusive, $\langle \Delta x^2 \rangle \propto \Delta t^{1.5}$ and $\langle \Delta z^2 \rangle \propto \Delta t^{1.6}$, to diffusive, $\langle \Delta x^2 \rangle \propto \langle \Delta z^2 \rangle \propto \Delta t$. The normal diffusion in long time scales is consistent with previous studies of dense drainage where particles were tracked with lower time resolution [20]. Curiously, the superdiffusion is slower than ballistic transport in fluids, $\langle \Delta x^2 \rangle \propto \langle \Delta z^2 \rangle \propto \Delta t^2$, which has been found in a recent indirect measurement of granular flow [21], albeit at the scale of surface roughness (<25 nm $\approx d/10\,000$).

Sub-ballistic scaling and non-Gaussian statistics at short times suggest that dense granular flows differ from classical fluids, as becomes more clear upon changing the flow rate. In a fluid, this causes a linear increase in Pe because the mean flow has no affect on molecular diffusion due to thermal fluctuations. Here, as shown in Figs. 3(c) and 3(d), the measured diffusion coefficients,

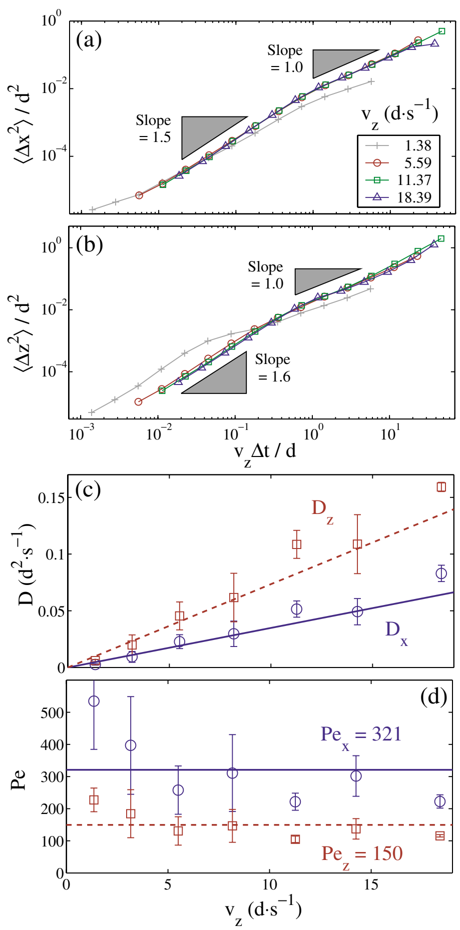

FIG. 3 (color online). (a),(b) Mean-squared horizontal ($\langle \Delta x^2 \rangle$) and vertical ($\langle \Delta z^2 \rangle$) displacements versus mean distance dropped, which collapse onto a single curve for different flow speeds v_z, except for the smallest where the flow is intermittent. (c),(d) Diffusion coefficients (D) and Péclet numbers (Pe) in the horizontal (x) and vertical (z) directions versus v_z.

e.g., $D_x = \lim_{\Delta t \to \infty} \langle \Delta x^2 \rangle / 2\Delta t$, are actually proportional to the flow speed (with $D_z \approx 2.1 D_x$, consistent with the discussion above), so the Péclet numbers, $\mathrm{Pe}_x = v_z d/D_x \approx 321$ and $\mathrm{Pe}_z = v_z d/D_x \approx 150$, are roughly constant. This suggests that diffusion and advection are caused by the same physical mechanism, such as a passing void. The measured Péclet numbers, however, are 2 orders of magnitude larger than predicted by the void model.

Since $D_x, D_z \propto v_z$, we plot the mean-square displacements versus the mean distance dropped in the laboratory frame, $v_z \Delta t$. Remarkably, as shown in Figs. 3(a) and 3(b), this collapses all of our data for different flow speeds onto a single curve, not only in the diffusive regime, but also in the superdiffusive regime. (The data for the smallest flow speed again differs somewhat.) A smooth crossover from super- to normal diffusion occurs after particles have fallen roughly one particle diameter.

Although advection dominates particle dynamics ($\mathrm{Pe} \gg 1$), diffusion causes a gradual rearrangement of the cage of nearest neighbors. To investigate this mixing directly, we measure the topological correlation function $C(\Delta t)$ defined as the fraction of nearest-neighbor pairs preserved from times t to $t + \Delta t$, averaged over all t. We chose the cutoff for a nearest neighbor, $1.5d$, as the first minimum of the radial distribution function (which yields coordinations near 0.59). As shown in Fig. 4(a), the data for $C(\Delta t)$ collapses when plotted versus $v_z \Delta t$, in the sense that no systematic dependence on v_z is observed (except perhaps for the smallest orifice widths), so in Fig. 4(b) we plot the average over all experiments in the continuous-flow regime ($W \geq 16$ mm or $v_z \geq 5.59d/\mathrm{s}$).

The cage correlation function in Fig. 4(b) exhibits a clear crossover, which closely parallels the crossovers for mean displacements in Figs. 3(a) and 3(b). In the superdiffusive regime, $C(\Delta t)$ decreases fairly quickly (with a decay length of roughly $20d$), but after falling more than one particle diameter the rate of decrease (neighbor loss) slows considerably. Since the topology remains more than 90% intact within the observation window, the precise form of the long-distance decay is uncertain, but a least-squares exponential fit, $\langle C \rangle \sim 0.976 \exp(-v_z \Delta t / 200d)$, yields a "cage breaking length" of $200d$. This gives direct evidence that the flow is characterized by long-lasting contacts, as is obvious to the naked eye. It also firmly rejects the void model because any void-particle exchange removes roughly half of the neighbors of a particle as it falls by only one diameter.

To counter the argument that a collisional regime may exist below the experimental resolution ($\Delta t \ll 1$ ms), we show that this is inconsistent with the fact that diffusion and mixing depend only on geometry (Figs. 3 and 4). In the standard model of a collisional gas, a particle dropping a distance L experiences an average of N collisions which must dissipate its gravitational potential energy: $mgL = (1/2)N(1 - e^2) m v_r^2$, where m is the mass, g the gravitational acceleration, e the restitution coefficient, and v_r the mean relative velocity. To be consistent with

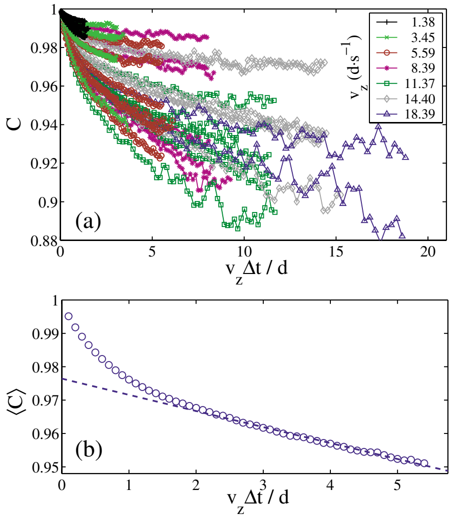

FIG. 4 (color online). (a) The topological cage correlation function $C(\Delta t)$ versus the mean distance fallen $v_z \Delta t$, and (b) the average over all experiments in the continuous-flow regime ($W \geq 16$ mm), compared with an exponential fit in the diffusive regime (dotted line).

our data, N should depend on L, but not the flow speed v_z. Although v_r is unknown, we can make two estimates—both of which lead to a contradiction. The first starts from the natural formula, $v_r^2 \approx (\langle \Delta x^2 \rangle + \langle \Delta z^2 \rangle)/\Delta t^2$ with fixed $\Delta t = 1$ ms, which suggests $v_r \propto v_z^{0.8}$ by looking at the initial slope of $\langle \Delta z^2 \rangle$ in Fig. 3(b). The second follows from direct measurements [21] of v_r yielding $v_r \propto v_z^{2/3}$. In either case, N would not be constant. [Note that $(1 - e^2)$ would typically correlate with v_r^2, so velocity-dependent restitution cannot compensate for the changes in v_r.]

More generally, in slow granular flows it seems that granular temperature may not be a useful concept. Figures 3 and 4 clearly show that fluctuations depend only on the distance fallen, and yet any notion of temperature should increase with the flow speed. The fact that the nearest-neighbor topology persists for distances comparable to the system size also seems to cast doubt on the assumption of ergodicity.

Instead, our experimental data suggests that cage rearrangements are caused by the relaxation of contact networks, as are believed to occur in Couette cells [22]. Such networks could absorb potential energy via rolling and sliding neighbors. The breaking of a contact could cause non-Gaussian fluctuations and small-scale superdiffusion among the particles in a network, while the gradual destruction of a network (and reformation of a new one) as a particle falls farther than its own diameter could cause the observed transition to normal diffusion. All of these effects are dominated by the geometry of random close packings, which is not fully understood, even without any dynamics [23].

In summary, we have experimentally investigated particle dynamics in dense granular flows—as they occur in silo drainage. Consistent with the void model, we observe diffusion after drainage by more than a particle diameter and Péclet numbers which are independent of the flow rate, suggesting that advection and diffusion have the same physical source. The Péclet numbers and cage-breaking lengths, however, are much larger than predicted, so the question of an appropriate microscopic model is left open.

We thank A. Samadani and D. Blair for helpful discussions. This work was supported by the U.S. Department of Energy (Grant No. DE-FG02-02ER25530), the Norbert Weiner Research Fund, the NEC Fund at MIT, and the National Science Foundation (Grant No. DMR-9983659) at Clark.

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