High-speed measurement of axial grain transport in a rotating drum

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Abstract. Granular mixtures segregate radially by size when tumbled in a partially filled horizontal drum on short timescales. The smaller component moves towards the axis of rotation and forms a buried core, which then forms undulations that grow into axial bands. Experimental and simulational studies have provided differing accounts of the type of transport taking place in radially segregated and self-diffusing grains. We investigate the bulk axial transport of small quantities of tracer particles travelling amongst glass spheres using non-invasive high-speed synchrotron x-ray particle tracking, which allows us to access timescales not explored previously. The mixtures used have varying propensity to axially segregate based on the glass sphere sizes when the tracers are present in larger proportions; we found that the single-particle dynamics of these mixtures when the tracer concentration is low do not appreciably depend on the relative particle sizes, implying that the potential for a mixture to axially segregate cannot be inferred from the microscopic dynamics of individual small particles. From the single-particle dynamics, we also found that while

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the standard measure of diffusive behaviour, the slope of the mean-squared displacement, is close to that expected from diffusive transport, more detailed analyses indicate anomalous transport.

Online supplementary data available from stacks.iop.org/NJP/13/105005/mmedia

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1. Introduction

A fascinating property of granular mixtures is their tendency to separate by size, or segregate, under a wide variety of flow conditions [1–4]. A popular example of this is the so-called ‘Brazil-nut effect’, which gets its name from the way the relatively large Brazil nuts are inevitably found at the top of a can of mixed nuts [2, 3]. Granular segregation is widely found in various natural phenomena as well as industrial processes; for example, the effect of segregation on avalanching flows has been proposed as a possible cause of sandstone or river bed stratification [4].

Possibly the most easily controlled, widely studied and least well-understood system is the segregation of grains in a partially filled, horizontal rotating drum ‘mixer’ [5–25]. A granular mixture composed of small and large grains will undergo radial segregation where the smaller particles migrate to the axis of rotation of the material in the drum after just a few rotations [6–8]. After hundreds of drum rotations axial bands can then form via a redistribution of grains within the radial core [7, 9]. The dynamics of grain transport along the radial core has a major influence on the axial segregation process, since axial bands can be considered to be undulations of the radial core which grow under repeated rotation [10, 11]; for segregation these undulations must then somehow be sustained against being mixed away by the random motion of the grains. Until now, most axial segregation models have generally assumed that the random motions of the grains along the axis of the drum imitate normal diffusive motion [10–13]. While theory and simulation derived from this standpoint have been able to reproduce some experimentally observed phenomena, such as axial band formation and oscillatory band dynamics, there have been experimental reports that have demonstrated anomalous diffusion under various conditions [15, 16]. In addition, the role of the radial core in the axial banding process is still unclear; hence, gaining an understanding of the axial transport process of the grains within and without the radial core is vital to develop a complete picture of the segregation process.

Quantifying axial transport processes is no simple task, as can be seen from various experimental studies of granular systems [14–17]. One such study investigated the mixing of bidisperse pharmaceutical spheres, which contain a liquid core of vitamin oil [14] using non-invasive MRI techniques. In these experiments the drum was rotated, stopped and inserted into the MRI bore to obtain concentration profiles of large particles evolving from a symmetrical...
two-band initial condition at discrete intervals spanning hundreds of rotations. It was observed that the concentration profiles were slightly asymmetrical; the small grains spread into the large grains more easily than the large grains spread over the small grains, and this behaviour was accounted for by using a diffusion model with a concentration-dependent diffusion coefficient. More recent MRI studies of water-immersed grains investigated the evolution of a three-band initial condition, where a narrow band or pulse of small glass spheres spread axially into surrounding larger glass spheres \[15\] along a radial core. It was found that the width of an evolving narrow pulse (or root-mean-squared displacement (MSD) of the grains) grows and the peak concentration decays as \( t^{1/2} \), as would be expected for a diffusive front-spreading process. In these experiments, the drum was also rotated, stopped and inserted into the MRI bore to obtain the order of ten fully resolved three-dimensional (3D) measurements over the course of hundreds of rotations. Fischer \textit{et al} \[15\] also reported that an evolving pulse of large grains spreading on the surface of small grains showed subdiffusive behaviour, where the pulse width grows and the peak concentration decays as \( t^\mu \) where \( \mu \sim 1/3 < 1/2 \). They associate this with a concentration-dependent diffusion process where the axial diffusion is weak since the grains slide down the front face of the mixture, and argue that at larger concentrations the large particles would collide more frequently with other large grains, increasing the stochastic axial motion resulting in Fickian diffusion.

Dynamic non-invasive imaging techniques that are used to measure the axial transport of grains while the drum is in motion have also yielded discrepant results. Khan and Morris \[16\], using optical transmission techniques, found that a narrow pulse of small opaque sand grains spread subdiffusively along the radial core into surrounding larger transluscent NaCl salt grains. This was interpreted to mean that axial bands are sustained against slower mixing processes than was previously supposed by continuum models \[11–13\], \[18–21\] or molecular dynamics simulations \[22–24\]. While the technique required that some assumptions had to be made about the subsurface structure of the radial core in order to obtain concentration measurements, surface measurements of dyed salt and glass grains mixing into otherwise identical salt and glass grains demonstrated similar behaviour. Positron emission particle tracking PEPT has also been used to non-invasively track the axial dispersion of irritated aspherical sand and titanium dioxide particles mixing with otherwise identical particles by tracking a radionuclide-labelled tracer particle in the sample via the triangulation of a number of detected annihilation events \[17\]. The findings of this study were consistent with diffusive behaviour, as axial displacement histograms of tracer displacements (measured once per drum rotation) were well described by Gaussian distributions.

It is still unclear how the grain properties (relative size differences, roughness, relative mass densities, asphericity, etc) affect the nature of both radial core and surface transport processes. As well, the relation of individual grain transport dynamics to axial segregation is not well understood. Here, we investigate the axial transport of grains in a rotating drum using high-speed synchrotron x-ray bulk 2D imaging of small tracer particles embedded in a matrix of typically larger particles. This technique allows imaging at high speeds relative to the rotation rate of the drum in order to non-invasively probe the grains’ microscopic motion; however, the maximum feasible concentration of tracer particles is insufficient for actual banding to occur. We use mixtures where, in larger proportions, the system is either known to dependably form axial bands or known to never form them, in the hope of relating the axial transport of individual grains to the tendency for axial banding. We found first that the individual particle dynamics does not markedly differ between the cases, implying that the macroscopic propensity...
of the system to form axial bands of small particles cannot be inferred from the microscopic motion of individual small particles alone. We also found that the typical quantity used to measure the diffusivity of transport, the MSD of individual tracers, is close to that expected from normal diffusion in all systems studied, although further analysis of the statistics of particle displacements indicates anomalous transport on short timescales.

2. Experimental details

Experiments were conducted in two stages. First, we investigated the ability of different mixtures of glass spheres and potassium iodide salt particles to mix or axially segregate by varying the glass sphere sizes; these experiments were conducted with 1/3 salt and 2/3 glass sphere mixtures using optical imaging, where the salt particles absorb more x-rays than glass spheres, making them ideal x-ray tracer particles for the single-particle tracking experiments. Once the segregation regimes of different mixtures were identified, we then performed high-speed synchrotron x-ray particle tracking using small quantities of tracers, as our ability to perform the measurements would be inhibited if they were present in large quantities.

The drum mixer used in these experiments consisted of a glass vial with a length $L$ of 110 mm and an inner diameter $D$ of 15.3 mm as sketched in figure 1. The drum was rotated about its long axis by a dc motor at a constant rotation rate of 0.41 rev. s$^{-1}$ where the flow was smoothly streaming. The typically larger grains were soda-lime glass spheres purchased from Whitehouse Scientific with diameters in the ranges of 0.212–0.250 mm, 0.360–0.420 mm and 0.500–0.600 mm and a mass density of 2.5 g cm$^{-3}$. The tracer grains used in all experiments were hand-ground irregularly shaped potassium iodide salt grains purchased from Sigma–Aldrich in pellet form, sieved to a size range of 0.200–0.250 mm with a mass density of 3.1 g cm$^{-3}$. Since the salt grains are slightly hygroscopic, they were placed in an oven at 80°C for a minimum of 30 min to remove excess moisture before loading into the drum. The cap of the drum was sealed with teflon tape to ensure an isolated environment, and no clumping of the salt grains was observed over the course of the experiments. The filled volume fraction of the drum was approximately 30% in all experiments.

Figures 2(a)–(c) show space–time diagrams of the mixing and segregation dynamics on the surface of mixtures of salt and glass spheres using optical visualization. Space–time diagrams

Figure 1. Sketch of the rotating drum setup. A drum of length $L$ and diameter $D$ rotates about its long axis at a constant rate, where the direction of rotation is indicated by the curved arrow. The Cartesian $x$-coordinate runs along the axis of the drum and the $y$-coordinate runs along the radius of the drum.
Figure 2. Space–time diagrams of the dynamics of granular mixtures in a rotating drum: (a) axial segregation does not occur in a mixture composed of 2/3 0.212–0.250 mm glass spheres and 1/3 small salt grains with 0.200–0.250 mm sizes. (b) Axial segregation does occur in a mixture composed of 2/3 0.355–0.420 mm glass spheres and 1/3 small salt grains with 0.200–0.250 mm sizes (see also supplementary movie M1 available from stacks.iop.org/NJP/13/105005/mmedia). (c) Axial segregation does not occur in a mixture composed of 2/3 0.500–0.600 mm glass spheres and 1/3 small salt grains with 0.200–0.250 mm sizes. Panels (d) and (e) show images of the final state of the experiment for the (a) 0.212–0.250 mm, (b) 0.355–0.420 mm and (c) 0.500–0.600 mm glass sphere and salt mixtures. These images correspond to the bottom row of the corresponding space–time diagrams.

were constructed by obtaining digital images of the rotating drum using a charge-coupled device (CCD) camera mounted perpendicular to the flowing surface of the grains at a rate of 2.05 frames s$^{-1}$ or 5 images for each drum revolution. A row of pixels running through the centre of the material in the drum (along the $x$, or axial direction) were obtained from each image, and averaged in groups of 5 in order to produce a slice of representative pixels at each
revolution of the drum. These rows were then stacked in time to produce a space–time diagram. The glass spheres were dyed black in order to enhance the optical contrast between the glass and salt grains, and the greyscale values are taken to be proportional to the concentration of black glass spheres and white salt grains on the surface of the flowing material in the drum. The imaging region was along the central 7 cm of the drum; the ends of the drum were not imaged.

From these space–time diagrams, we observe that axial segregation does not occur in mixtures of 1/3 salt and 2/3 glass spheres with diameters in the ranges of 0.212–0.255 mm and 0.500–0.600 mm shown in figures 2(a) and (c), respectively. The greyscale values in figure 2(a) are higher than those in figure 2(c) because the white salt grains are present at higher concentrations on the free surface as shown in images of the final state of the experimental runs, figures 2(d) and (f). A small number of salt grains were present on the free surface of the mixture in figure 2(f); however, the majority are present in a radial core as confirmed by excavating the material in the drum. We also observe that axial segregation does occur in mixtures of 1/3 salt and 2/3 glass spheres with diameters in the range of 0.355–0.420 mm as shown in figure 2(b) and supplementary movie M1 available from stacks.iop.org/NJP/13/105005/mmedia. An image of the segregation pattern is shown in 2(e). These measurements demonstrate that we can probe a variety of behaviour of 0.200–0.250 mm tracer particles embedded in mixtures of glass spheres with different sizes: mixing, radial segregation alone and radial segregation along with axial segregation. The x-ray single-particle tracking experiments described next were performed with these combinations of grain sizes, where approximately 500 salt tracer particles were used in these mixtures (which constitutes approximately one thousandth of the grain volume in the drum). As noted, this concentration is not sufficient to form a coherent core of small grains although it is possible to obtain good statistics of individual particle motion in order to probe the grains’ micro-environment.

Particle tracking experiments were performed at beamline ID15A at the European Synchrotron Radiation Facility (ESRF). The use of high-intensity synchrotron radiation to non-invasively penetrate our dense granular sample allowed us to follow the 2D motion of the tracer particles at high speeds, since the x-ray contrast between the glass spheres and the potassium iodide salt tracer particles was large enough to be distinguished. Salt grains were mixed with glass spheres in the rotating drum, and were imaged at the centre of the drum using a monochromatic beam with a photon energy of 52.5 keV, where this energy was chosen to optimize the combination of x-ray flux, x-ray transmission through the sample and the detector efficiency. The beam spot size was 8.5 × 8.5 mm². The imaging detector consisted of a 500 µm thick LuAG:Ce scintillator screen, which converts the x-rays passing through the sample into visible light. The light was then collected by a Carl Zeiss Macro Planar 100/2.8 objective with 1× magnification and the images were recorded at a rate of 250 frames s⁻¹ (i.e. a time interval of 0.004 s) using a Sarnoff CAM512 CCD camera with a 512 × 512 pixels² sensor and 18 µm pixel size. The total observation time for each run was 600 s. This imaging setup provided a bulk 2D projection image of the grains in the rotating drum system, where the camera is perpendicular to the rotation axis of the drum to facilitate observation of the axial displacements of the tracer particles. An example of a raw image obtained for this system is shown in figure 3(a)—of some 0.200–0.250 mm diameter potassium iodide salt grains embedded in 0.360–0.420 mm diameter glass spheres. The alternating dark and light horizontal bands in this image are due to the vertical beam profile generated by the high-energy x-ray undulator source. The salt tracers are more absorbing than the surrounding glass spheres, resulting in identifiable dark spots in the images.
Figure 3. (a) Raw x-ray image of 0.200–0.250 mm diameter potassium iodide salt grains embedded in 0.355–0.420 mm diameter glass spheres. The tracer particles appear as dark spots in the image, and the vertical intensity undulations are due to the vertical beam profile. (b) Processed image prior to application of a threshold to determine tracer particle position. See text for details of the image analysis steps.

The first step of the image analysis was to normalize the lateral intensity fluctuations of the x-ray radiation and to correct for pixel-dependent background artifacts. This was done by subtracting a dark field (an image with no beam) from all of the images including the flat field (an image with the beam but no sample) and then dividing all of the images by the flat field. The dark and flat field images were generated before each run, and were each produced by averaging 500 single images. Since the intensity variations, in particular the intensity undulations due to the beam’s profile, were not entirely corrected by this step, and because additional intensity variations were due to the sample’s non-constant cylindrical thickness, the images were further flattened; the average of 1500 images evenly spaced over the length of the run was obtained to wash out the intensity variations. To smooth out any remaining low-intensity regions due to the presence of the tracers, we additionally applied a circular averaging disc filter with a radius of 50 pixels to the averaged image. The raw data images were then divided by this averaged and filtered image. Finally, we applied a median filter with a 3 × 3 mask size to remove any salt-and-pepper noise from the images. An example of the output from these image processing steps is shown in figure 3(b).

As a result of this pre-processing, we were able to apply a constant grey level threshold over the entire image for the entire duration of a run where the darkest objects within the correct size range were identified as tracer particles. The particles’ centroid detections were performed on the resulting binary images with subpixel accuracy using the Matlab image processing toolbox. Finally, the positions of tracer particles in each image were tracked using a predictive algorithm [26] in order to determine the particles’ trajectories. Due to the occurrence of tracer particle overlap in the 2D projections of this 3D system, we terminated trajectories prior to the overlap in order to minimize errors in centroid detection. Some examples of tracer trajectories are shown in figure 4 and supplementary movie M2 available from stacks.iop.org/NJP/13/105005/mmedia, where the tracers embedded in 0.212–0.250 mm

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3. Results and discussion

The average MSD of tracer particle displacements obtained from particle tracking experiments in the axial or $x$-direction $\langle \Delta x^2(\tau) \rangle$ is shown in figure 5(a) for all three combinations of small and large particles mentioned above. We can see that the slopes of the MSD in all cases examined are well-described by a power-law time dependence

$$\langle \Delta x^2(\tau) \rangle \sim \tau^\alpha$$

where $\alpha < 1 \Rightarrow$ subdiffusion, $\alpha = 1 \Rightarrow$ diffusion, $\alpha > 1 \Rightarrow$ superdiffusion.

Least-squares fitting the logarithm of the MSDs against the logarithm of time yields slopes of $1.17 \pm 0.09$, $1.08 \pm 0.09$ and $1.10 \pm 0.04$ for tracers embedded in mixtures of glass spheres.
Figure 5. (a) Ensemble averaged MSD $\langle \Delta x(\tau)^2 \rangle$ of 0.200–0.250 mm tracer particles embedded in mixtures of glass spheres plotted against time on logarithmic axes. The fitted slopes of these curves (not shown) are 1.17, 1.08 and 1.10, respectively. (b) PDFs of tracer particle displacements $\Delta x(\tau)$ plotted logarithmically against linear displacement normalized by the mean tracer particle diameter $d_s$ for a displacement time $\tau$ of 0.004 s. These distributions have broader tails at large displacements than would be predicted by Fickian diffusion, where the expected form of the PDF would be a Gaussian curve, indicated by the grey dashed line. The tails of the distributions are well described by power-law fits, shown as red dashed lines. (c) PDFs of tracer particle displacements $\Delta x(\tau)$ embedded in 0.500–0.600 mm glass spheres for different displacement times $\tau$. The distributions approach a Gaussian (parabolic) shape for a large displacement time $\tau$. (d) Time dependence of the normalized kurtosis $\kappa$ of distributions generated by calculating the displacements for increasing wait times $\tau$. The colours indicate the different mixtures of glass spheres used, with diameters in the ranges of 0.212–0.250 mm (black squares), 0.355–0.420 mm (blue squares) and 0.500–0.600 mm (green squares).
with diameters in the ranges of 0.212–0.250 mm, 0.355–0.420 mm, and 0.500–0.600 mm, respectively (not shown). These slopes can certainly be interpreted as indicating superdiffusive behaviour; however, they are close enough to 1 that some may consider them to be consistent with normal diffusive (Brownian) behaviour. In order to see whether this is actually the case we subjected the data to further analysis, as discussed below.

The probability distribution function (PDF) of particle displacements $P(\Delta x(\tau))$ for $\tau = 0.004$ s is shown in figure 5(b), and we observe that these distributions have significantly heavier tails than a Gaussian distribution, shown as a dashed grey curve. We find that the tails of these distributions are well described by power-law fits, determined by least-squares linear fits to the base 10 logarithm of the tails, shown as red dashed lines. This indicates that the tracers take larger steps than would be expected for a Brownian transport process. Non-Gaussian displacement PDFs have been observed in the dynamics of colloidal glasses [27], dense granular materials under shear [28] and dense granular silo flows [29]. In the systems described in [27, 28], they are attributed to dynamical heterogeneities in the motion of the particles, where the narrow central region of the PDF corresponding to small displacements is attributed to caging of the particles, and the broad tails are attributed to cage-breaking events. In these studies, the non-Gaussianity of the tails depended on the time interval $\tau$ used to calculate the displacements, and the distributions of displacements approached a Gaussian as the time interval was increased. The displacement PDFs of tracer particles embedded in 0.500–0.600 mm glass spheres for different time intervals $\tau$ are shown in figure 5(c), and we similarly find that for a large time interval the distribution has a parabolic Gaussian-like form. We also observe that the PDFs we obtained are slightly asymmetric due to the presence of a small tilt in the setup; this is not expected to affect the functional form of the distribution [30].

In order to quantify the time dependence of the non-Gaussianity of our displacement PDFs, we calculated the normalized kurtosis, $\kappa(\tau) = \langle \Delta x(\tau)^4 \rangle / 3 \langle \Delta x(\tau)^2 \rangle^2 - 1$, for increasing $\tau$. The normalized kurtosis of a broad distribution has a large value, and is zero for a Gaussian distribution. We plotted $\kappa(\tau)$ against $\tau$ on logarithmic axes in figure 5(d), and we observe that while the normalized kurtosis approaches 0 at long times for all three systems, it does so without significant change in slope of the MSD demonstrated in some other systems. For example, in dense granular silo flows [29] and colloids approaching the glasses transition [27], the MSD slope evolves from having a slope close to 1 to a subdiffusive plateau back to having a slope close to 1; similarly, for dense granular materials under shear the MSD slope evolves from being subdiffusive with a slope of 0.5 at short times to diffusive with a slope of 1 at longer times [28]. In our system as well, the particle displacement distributions approach Gaussian statistics at longer time intervals, although in our case it involves a much gentler bending of the MSD curve towards 1.

The trajectories of representative tracer particles are shown in figure 4 for the three runs, and we speculate that the hallmarks of the dynamical heterogeneity found in glassy systems [27, 28], namely particle caging and cage-breaking, are not present in the traditional sense in our system due to the method in which the system is driven. We observe that the tracer particles take short axial steps as the grains are transported nearly as a solid body up the back of the rotating drum, and take larger axial steps as they are sheared by grains flowing down the front face of the material in the drum (see supplementary movie M2 available from stacks.iop.org/NJP/13/105005/mmedia). The range of axial step sizes available to the particles may also depend on the proximity to the free flowing surface. This manner of transport can account for broad displacement distributions at short timescales...
and Gaussian displacements on longer timescales, as the particles have undergone multiple rotations.

In order to examine more closely whether upward-moving and downward-moving particles make distinct contributions to the observed transport properties, we calculate the persistence and exchange time distributions of our tracer particle trajectories, an approach that has previously been used to illustrate the presence of dynamic heterogeneities in glassy systems [31–33]. The persistence time is defined as the time \( t_1 \) to traverse a distance \( \xi \), for a particle whose trajectory begins at time \( t = 0 \). The exchange times are then defined as the subsequent time intervals \( t_n - t_{n-1} \) (for all \( n > 1 \)) that it takes for the particle to travel additional distances of \( \xi \) in any direction [31]. For normally diffusive systems the persistence and exchange time distributions are identical, while it has been observed in dynamically heterogeneous systems that once a particle has jumped a cutoff distance \( \xi \) it is more likely to perform subsequent jumps; this has been attributed to the presence of spatially clustered particles with correlated dynamics [32, 33]. In figures 6(a)–(c), we show the persistence time and exchange time distributions for salt tracer particles mixing with glass spheres with diameters in the range of 0.355–0.420 mm. Distributions are shown separately for particles travelling upwards and flowing downwards in the drum; the cutoff distance is \( \xi = 0.213 \text{ mm} \), which corresponds to the mean small grain diameter. The distributions for tracer particles mixing with other glass sphere sizes are qualitatively similar to those presented.

We observe in figures 6(a) and (c) that the persistence and exchange time distributions for both entire trajectories and downwards movement show no obvious decoupling (i.e. the motion is uncorrelated); but the distributions for upward-moving particles (shown in figure 6(b)) are slightly decoupled. This indicates the presence of a correlated particle motion in the upward-moving layers, where once a particle has taken a jump of length \( \xi \), it is more likely to take another jump of equal length, with the effect of shifting the peak of the exchange time distribution to a shorter time relative to the peak of the persistence time distribution.

Having provided evidence that the tracer particles’ motion is correlated at distinct points in their trajectories, we also test for correlations at the ensemble level by considering whether the system as a whole satisfies the Markov property. This is the property that the probability of the system to make a transition to any possible state is only dependent upon its current state; such systems are commonly referred to as ‘memoryless’. For this reason we look at the conditional probabilities \( P_n(x(t+2n\tau) - x(t) | x(t+n\tau) - x(t)) \), the probability that a particle having experienced a displacement \( \Delta x \) in time \( n\tau \) will have a displacement of \( \Delta x' \) during the subsequent \( n\tau \) time interval. If the transport is a Markov-type process, then these conditional probabilities can be treated as state transition probabilities between displacement values [34].

Given the nature of our data, the most straightforward way of constructing our transition matrices is to first calculate and tabulate the joint probabilities \( P(A, B) \) for every pair of events \( A \) and \( B \) (here displacements in the first and second time intervals) and then convert them to conditional probabilities \( P(A|B) \) by normalizing the row sums to 1 [35], which then gives the row of the transition matrix governing transitions from the state corresponding to event \( B \). Figures 7(a) and (b) show joint probability distributions \( P_2 \) and \( P_4 \) for time delays of \( 2\tau \) and \( 4\tau \), \( \tau = 0.004 \text{ s} \). We first note that the joint probabilities are not symmetric, as would be expected for Gaussian distributions. They are instead noticeably elongated along the negative to positive diagonal, which indicates that particles moving in either the negative or the positive \( x \)-direction are more likely to continue in that direction. We can make this more rigorous by considering the
transition matrices $T_2$ and $T_4$ obtained from $P_2$ and $P_4$. The positive correlations in displacement values show up here as well (figures 7(c) and (d)), although they are stronger for $2\tau$ than for $4\tau$. For a memoryless stochastic process, iterative application of a transition matrix $T_n$ should give the transition probabilities for the time intervals $2n\tau$, $3n\tau$, . . . , i.e. $T_n^m = T_{mn}$. We can see from figure 7(e) that two iterations of our empirically derived transition matrix $T_2^2$ are not in any way close to the experimentally derived $4\tau$ transition values $T_4$ ($|T_2^2 - T_4|$ is shown explicitly in figure 7(f)), implying that the system is not Markov. We have confirmed that the same holds also for the other grain mixtures with respect to $2\tau \rightarrow 4\tau$, as well as for $\tau \rightarrow 2\tau$ and $4\tau \rightarrow 8\tau$.

4. Conclusions and outlook

Our results reveal that microscopic axial motion in this system is more complex than can be accounted for by diffusive models, as has been previously assumed. In particular, the particles
undergo larger displacements (broad-tailed PDFs) and possess correlations dependent upon physical position (up or down flows), while any accurate model of the microscopic motion of the axial transport of the grains on short timescales would have to be non-Markovian. One general question is: how can this be modelled on reasonable physical grounds to describe the phenomena observed here and by others in this system [15–17]?

One situation where non-Gaussian PDFs can arise is when an object experiences a nonlinear resistance to motion. For dry solids this has been demonstrated to arise from the presence of Coulombic dry friction [36], and it may account for the non-Gaussian velocity
distribution functions observed in granular gases [37, 38]. However, the functional forms for the PDFs that are predicted by this approach are exponential, while we have observed that our distributions have power-law tails in the early times of our experiment (cf figure 5(b)).

Another approach would be to apply the continuous time random walk model (CTRW) to this system. In this framework the observed motion is parameterized by both a jump length distribution function $\psi(x)$ and a waiting time distribution function $\phi(t)$. Depending on the respective distribution functions involved, CTRW models can describe straightforwardly diffusive transport, subdiffusion (when $\phi(t) \propto t^{-1-\alpha}$) or superdiffusion (when $\psi(x) \propto x^{-1-\beta}$). In particular, when both distributions are power laws the system could be said to feature competition between subdiffusive and superdiffusive aspects: for example, when we have superdiffusion on a subdiffusive medium. While such a system would certainly describe anomalous transport, a diffusive-looking MSD could result if $\beta$ was close to $2 \times \alpha$, since then we would have $\langle (\Delta x(t))^2 \rangle \propto t^{2\alpha/\beta} \approx t$ [34, 39]. With respect to the rotating drum system the trapping time distribution could come from time spent in a ‘passive layer’ moving in solid body rotation with the drum, and a jump length distribution could arise from transport in an ‘active layer’ being sheared by grains on the free surface flowing down the front face of the grains in the drum. This model could also account for the appearance of system memory, since the CTRW model can describe non-Markovian particle behaviour when the waiting time distribution is non-exponential [40].

We have observed qualitatively similar transport properties for small tracer particles in the three systems examined, although they would display different segregation behaviours if the concentrations of tracer particles were higher. The implication is that transport in the segregating regime is not fully determined by the transport properties of the individual elements of the system. As well, while particles undergo a transport process that superficially resembles Fickian diffusion since MSDs grow nearly linearly with time, this transport is not Fickian in all of the aspects examined here. It seems likely that the richness and complexity of the rotating drum system can only be fully understood in a framework that acknowledges that the particle transport can behave anomalously.

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