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Collisions and intermittency in granular flow

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Abstract. We review recent and ongoing experiments in which grain dynamics are probed by correlations in intensity fluctuations for light multiply scattered from bulk samples under flow. This includes application of standard diffusing-wave spectroscopy (DWS), when the driving force is strong and the resulting flow is smooth and continuous, as well as use of higher-order temporal correlation functions, when the driving force is gentle and the resulting flow is an intermittent series of avalanches.

1. Introduction

When subjected to small forces, or no forces at all, the grains within a bulk granular material remain at rest. Thermal forces plus small external forces are not sufficient to overcome gravity and permit grains move relative to one another. Static sand is thus jammed, stuck forever in a random disordered configuration determined by its most recent flow history [1]. When subjected to sufficiently large forces, the grains can rearrange and the bulk material can flow and deform at roughly constant density. This is reminiscent of ordinary liquids, except that the source of dissipation is inelastic grain–grain collisions that operate even in the absence of bulk shear flow. When subjected to moderate forces, the flow can be intermittent—alternately static and fluidized as in an avalanche. This range of behaviour is known to anyone who has poured a bucket of sand or a box of cereal, or has drunk from a glass with too much ice and too little water.

A key experimental challenge is how to characterize the full range of grain dynamics encountered during flows. This includes both the single-grain dynamics in the fully and intermittently fluidized regimes, as well as the collective on/off avalanche behaviour in the latter. Recently we have shown how to deduce single-grain dynamics in the fully fluidized state using diffusing-wave spectroscopy (DWS) [2, 3], a dynamic light scattering technique for cases of strong multiple scattering [4–6]. We observe that grains fly randomly and ballistically between successive collisions, and that many collisions can be required for a grain to break out of its cage of nearest neighbours. This permits direct contact with kinetic theories of granular hydrodynamics, see e.g. [7–9], for which the square of the random velocity scale (the 'granular temperature') is a crucial phenomenological ingredient. The point to be especially emphasized here is that neither these theories, nor the standard DWS approach, are valid when the forcing is moderate and the flow is intermittent; new theoretical and experimental methods are needed. Below I first review our previous work [2, 3], then I discuss ongoing experiments with higher-order correlation functions [10] regarding the latter.

2. Continuous flow

Popular methods of fluidizing sand include rotating a drum, tilting a pile and shaking a heap, all with great vigour. These excite complex flows that are spatially or temporally heterogeneous, preventing straightforward application of DWS. To provide more uniform forcing, and hence more uniform flows, we have employed two different methods. The first is a tall rectangular chute, where a reservoir at the top permits long runs, and a sieve at the bottom controls the average downward flow rate [2]. The second is a gas-fluidized bed, where an upflow of gas fluidizes sand in a tall column of constant cross-section [3].

Grain dynamics in the fully fluidized state may be probed by DWS as follows. Coherent laser light (i.e. with a delta-function power spectrum) is directed toward the sample. It reflects strongly from the grain–air interfaces and thus performs a random walk within the sample; this gives sand its familiar opaque appearance. If the grains are moving, then the light is Doppler shifted and the power spectrum of the emerging multiply scattered light is accordingly broadened. From the shape of the spectrum, one may deduce the nature of the motion. Since the motion is slow compared to the speed of light, a beating method must be employed. Namely, one measures the intensity autocorrelation function via photon counting and digital signal processing methods. If the detected photon paths are numerous *and* uncorrelated, then the central limit theorem implies that the total electric field is a Gaussian random variable. Hence the measured intensity autocorrelation function can be factorized, giving the well known Siegert relation, $g^{(2)}(\tau) \equiv \langle I(0)I(\tau)\rangle/\langle I\rangle^2 = 1 + \beta/\gamma(\tau)|^2$. Here, $\beta < 1$ depends on the detector area and $\gamma(\tau)$ is the normalized electric field autocorrelation function, which is the Fourier transform of the power spectrum. For the usual case where all detected photons experience single scattering by wavevector $\boldsymbol{q}$, we have $|\gamma(\tau)| = \langle \exp\{-\mathrm{i}\boldsymbol{q}\cdot[\boldsymbol{r}(t+\tau) - \boldsymbol{r}(t)]\}\rangle$, which reduces to

$$|\gamma(\tau)| = \exp[-q^2\langle\Delta r^2(\tau)\rangle/6] \tag{1}$$

if the distribution of scattering site displacements in time τ is Gaussian, as is usually the case [11]. For the case of strong multiple scattering (DWS), we instead have

$$|\gamma(\tau)| = \int P(s)\exp\left[-\frac{s}{3l^*}k^2\langle\Delta r^2(\tau)\rangle\right]\mathrm{d}s \tag{2}$$

an average over the photon path length distribution $P(s)$, which may be found by assuming photons diffuse and execute random walks of stepsize l^*. For both single scattering and DWS, one thus measures the intensity autocorrelation and ultimately deduces the average mean-squared displacement of the scattering sites as a function of delay time τ.

The main features of the single-grain dynamics, $\langle\Delta r^2(\tau)\rangle$ against τ, for 100 and 200 μm diameter glass spheres in the rectangular chute are similar to those for a glassy liquid [2]. At the shortest times, $\langle\Delta r^2(\tau)\rangle$ grows as τ^2, indicating random ballistic motion—consistent with the notion of a granular temperature. It then abruptly rolls over into a plateau that only slightly increases over a few decades in delay time—consistent with repeated collisions with a nearly fixed cage of nearest neighbours. At the latest times, the DWS results head smoothly toward macroscopic video observations of diffusive motion, providing an important check. A more crucial check is that the DWS signal is strongly dependent upon illumination/sample/detection geometry, through $P(s)$ in equation (2), but the extracted single-grain dynamics *are not*.

The scales for ballistic dynamics and collisions deduced from $\langle\Delta r^2(\tau)\rangle$ data are as follows. The granular temperature speed is the same order of magnitude as the average hydrodynamics flow speed, as expected since the flow itself generates the fluctuations, but scales as a power law $\delta v \sim \langle v\rangle^{2/3}$; the exponent is not understood. For the gas-fluidized bed [3], we find ballistic speeds nearly identical to those obtained by acoustic shot noise [12, 13]. The collision times and distances, where $\langle \Delta r^2(\tau) \rangle$ bends from τ^2 towards a plateau, are in the ranges τ_c: 10^{-5}–10^{-4} s and l_c: 10–100 nm respectively. It may, at first glance, be surprising that slow dense flows can generate such rapid collisions at lengths much smaller than the grain diameter. Since such behaviour is not accessible by video or other imaging methods, and is sufficiently nonintuitive, it may be tempting to dismiss our results. Besides the tremendous redundancy and self-consistency in our data set, and the agreement with acoustic shot noise results for δv already noted, we add that the collision scales are consistent with simple order-of-magnitude estimates based on the nature of the driving force, namely gravity. Given a characteristic flow speed of $\langle v \rangle = 1$ mm s^{-1}, we immediately obtain a characteristic time scale $\langle v \rangle / g = 10^{-4}$ s and a length scale $\langle v \rangle^2 / 2g = 50$ nm, which are right on the mark. A slightly more sophisticated version of this argument is to equate the energy per grain per unit time supplied by gravity, $mg\langle v \rangle$, with that dissipated due to collisions, $\sim m\delta v^2(1-\varepsilon^2)/\tau_c$, where $\varepsilon \sim 0.9$ is the grain–grain restitution coefficient. Another order-of-magnitude calculation is that the particle Reynolds number, inertia/drag $\sim (m\delta v^2/D)/(\eta_{air}\delta v D)$, is much greater than one, consistent with a collisional regime. Finally, our results have been largely reproduced by a recent simulation of our experiment [14]. Altogether, there is a substantial preponderance of evidence supporting our interpretation.

3. Intermittent flow

Consider now what happens when the sieve size is progressively decreased in our chute experiments. Both the average flow speed $\langle v \rangle$ and fluctuation speed δv decrease, but, owing to the 2/3 power law, the relative size of the fluctuations increases. Also the duration of the plateau increases as the grains spend more and more time stuck in a cage of nearest neighbours. Eventually, on both counts, the system jams up and stops flowing smoothly. In fact, there is a lower limit to the values of $\langle v \rangle$ that may be realized. This is highly reminiscent of 'ticking' in hourglasses [15], and has analogues with avalanche-like behaviour for other means of forcing. Such on/off intermittency is highly correlated, and invalidates crucial assumptions underlying both the analysis of DWS experiments and the continuum PDEs of kinetic granular hydrodynamic theories. The latter may well apply *during* the flows, and cellular automata exhibiting avalanches of various size distributions, depending on the rules, may well apply *in between* the flows, but neither theoretical approach can capture the full range of behaviour. New theoretical and experimental tools are needed to understand the regime of intermittent flows.

Experimentally, we have embarked on a programme to exploit higher-order intensity correlation functions as a means of characterizing both single-grain as well as collective on/off avalanching dynamics: $g^{(n)}(\tau_1, \ldots, \tau_{n-1}) = \langle I(0)I(\tau_1)\ldots I(\tau_{n-1})\rangle / \langle I \rangle^n$. The underlying idea is that, since the total detected field is no longer Gaussian, there is extra information contained in the $g^{(n)}$. By contrast, for the more usual case that the total detected field $\sum E$ is Gaussian, the higher orders decompose into sums of products of two-point correlations, as we showed in [10]:

$$g^{(2)}(\tau_1) = 1 + \beta|\gamma_{01}|^2 \qquad \text{(the usual Siegert relation)} \tag{3a}$$

$$g^{(3)}(\tau_1, \tau_2) = 1 + \beta(|\gamma_{01}|^2 + |\gamma_{12}|^2 + |\gamma_{20}|^2) + 2\beta^2 \,\mathrm{Re}\,(\gamma_{01}\gamma_{12}\gamma_{20}) \tag{3b}$$

$$\begin{aligned} g^{(4)}(\tau_1, \tau_2, \tau_3) = {} & 1 + \beta(|\gamma_{01}|^2 + |\gamma_{02}|^2 + |\gamma_{03}|^2 + |\gamma_{12}|^2 + |\gamma_{13}|^2 + |\gamma_{23}|^2) \\ & + \beta^2(|\gamma_{01}|^2|\gamma_{23}|^2 + |\gamma_{02}|^2|\gamma_{13}|^2 + |\gamma_{03}|^2|\gamma_{12}|^2) \\ & + 2\beta^2 \,\mathrm{Re}\,(\gamma_{01}\gamma_{12}\gamma_{20} + \gamma_{01}\gamma_{13}\gamma_{30} + \gamma_{02}\gamma_{23}\gamma_{30} + \gamma_{12}\gamma_{23}\gamma_{31}) \\ & + 2\beta^2 \,\mathrm{Re}\,(\gamma_{01}\gamma_{12}\gamma_{23}\gamma_{30} + \gamma_{02}\gamma_{23}\gamma_{31}\gamma_{10} + \gamma_{02}\gamma_{21}\gamma_{13}\gamma_{31}). \end{aligned} \tag{3c}$$

The time intervals at which the field autocorrelation is to be evaluated are denoted by subscripts. Data for the special slices $g^{(3)}(T, \tau)$ and $g^{(4)}(T, \tau, \tau + T)$ against τ are obtained *in real time* by passing the photon count signal through a digital delay line of controllable length T, multiplying with the undelayed signal and feeding the product into a standard digital auto- or cross-correlator. By comparing data with predictions for $g^{(3)}$ and $g^{(4)}$ generated from $g^{(2)}$ data using equations (3), we can determine whether the scattering process is Gaussian or not. If so, then scattering site dynamics can be extracted using equations (1) or (2); if not, then a different analysis is required according to the nature of the non-Gaussianity. In [10], we demonstrated this for Gaussian scattering and a variety of non-Gaussian processes including laser drift and temporal incoherence, number fluctuations, a stray heterodyne component, nonergodic dynamics and on/off intermittency.

For our current on/off intermittency experiments we have developed and tested an analysis procedure for extracting useful information from the measured $g^{(n)}$. Ingredients include the normalized field autocorrelation function $\gamma(\tau)$ for the on state, from which single-grain dynamics may be extracted using equations (1) or (2); the probability $P_0(\tau)$ to be in the same off state after a time interval τ; and the interrelated switching probabilities $P_{ij}(\tau)$ to be in state j after a time interval τ from being in state i. Note that $P_0(\tau)$ decays monotonically from 1 to 0, and that $P_{ij}(\tau)$ decay from either 1 or 0 to the probability f_j to be found in state j; subscripts 0/1 denote off/on. For example, one could almost guess our result

$$g^{(2)}(\tau) = 1 + \beta[f_1 P_{11}(\tau)|\gamma(\tau)|^2 + f_0 P_0(\tau)] \tag{4}$$

which replaces the usual Siegert relation, equation (3*a*); we also have such predictions for temporal intensity correlations up to fourth order. At short τ, the decay is due primarily to the fast single-grain dynamics with the slow switching functions remaining constant; at long τ, following a plateau, the final decay is entirely due to the switching functions. Note that the normalized second intensity moment is $g^{(2)}(0) = 1 + \beta$, as for Gaussian processes. Intermittency is thus different from number fluctuations, where the intensity distribution is strongly affected by occasional intense bursts. Here, correlations—not lack of sufficiently great numbers—prevent application of the central limit theorem.

To confirm our analysis procedures for on/off intermittency, we have performed 'toy' experiments in which the on/off switching is imposed by periodic rotation of an auger plunged into a tube filled with sand. In this case $P_0(t)$ decays linearly from one at $t = 0$ to zero at $t = T_{off}$, the duration of the off cycle; the P_{ij} are periodic tent-shaped functions. Indeed, we are able to reconstruct $g^{(3)}$ and $g^{(4)}$ data in terms of these expected switching functions plus $\gamma(\tau)$ data obtained during continuous rotation, absolutely with no fitting parameters.

As of this writing only preliminary data are available from one 'real' experiment: surface flow along a heap between clear plates onto the top of which grains are added at a controlled rate. For fast rates, the flow is smooth and continuous, and the $g^{(n)}$ are Gaussian with a single decay time scale apparently indicative of the shear flow within the layer. For slow rates, below about eight grains per second per unit width, the flow becomes intermittent, and the $g^{(n)}$ become non-Gaussian. It appears that the on dynamics $\gamma(\tau)$ are independent of flow rate, equal to that found for the slowest continuous flow; the corresponding depth of the flowing surface layer is ten grain diameters deep. The P_0 in this regime are nearly exponential, and scale with the flow rate. This is indicative of a Poisson switching process, as in random telegraph noise. If the switching process were like that for self-organized criticality, the P_0 would instead decay as a negative power of time. The P_{ij} have not yet been extracted from $g^{(4)}$ data, as their contribution is 24 times smaller than P_0 and sufficiently good statistics have not yet been obtained. This should not pose a serious problem, in that the P_{ij} are solely responsible for the periodicity already clearly seen in $g^{(4)}$ data for the auger experiments.

4. Conclusion

The phenomenon of on/off flow intermittency is generic to any granular system subjected to gentle forcing. Long quiescent times of no motion are punctuated by sudden avalanche-like flows in which energy is dissipated by inelastic grain–grain collisions at high frequency. The collective nature of the switching, as well as the vast separation in time scales separating collision and switching events, pose a severe challenge to both theory and experiment. The continuum partial-differential equations of kinetic theories for granular hydrodynamics cannot capture intermittency, and the cellular automata of avalanche models cannot capture the fundamental source of dissipation in grain–grain collisions. Of course neither approach can be used to study the crossover from intermittent to smooth continuous flow with increased forcing rate. Experimentally, as discussed here, we can now realize such studies with the advent of higher-order temporal intensity correlation functions for multiply scattered light. We have previously shown how to apply conventional DWS to extract single-grain dynamics for continuous flows [2, 3]; now, we can additionally extract probability functions that quantitatively describe the on/off switching statistics. The status of our programme is as follows. We have designed, built and tested hardware for real-time measurement of higher-order correlation functions. We have derived, and experimentally verified, the predictions in equations (3) for the usual case of a Gaussian scattering process; we have also shown how deviations arise in a characteristic manner for several non-Gaussian processes. This is all described in [10]. In addition we have developed a theory for analysing correlation data in terms of switching functions, and have thoroughly demonstrated its validity with 'toy' experiments in which intermittency is imposed in a known way by periodic rotation of an auger. Preliminary data have been obtained and tentatively analysed for a 'real' experiment in which intermittency arises naturally by the steady sprinkling of grains onto an existing pile. Thanks to this investment, we are thus now poised to collect good statistics on the single-grain dynamics and the on/off switching statistics for a variety of important granular flow situations. Special interest is in the nature of the transitions from complete fluidization (simple liquid) to intermittent fluidization (correlated/glassy liquid) to fully jammed (static solid) behaviour as the driving rate is systematically lowered.

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