The statistics of intermittent jamming in shear-thickening suspensions

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Using a simple piezo sensor technique analogous to an old-fashioned record player needle, we measure the statistics of macroscopic intermittent forces during shear of concentrated colloidal suspensions. We find that the shear-thickening regime is characterised by intermittent jamming events whose magnitude distribution is exponential and whose distribution in time follows a power-law, demonstrating intriguing similarity to diverse other systems such as clogging in granular hoppers and pedestrian and animal flows, despite the obvious differences in materials and flow scenarios. The sum of event magnitudes per unit strain, E_j, has the same dependence on volume fraction as the time-averaged effective viscosity, demonstrating that the bulk system rheology in the shear-thickening regime is determined directly by the statistics of these intermittent jamming events. The sum E_j is independent of system size, however in larger systems fluctuations become more severe but less frequent, *i.e.* the way a fixed total 'jamming activity' E_j is shared out between statistical events depends on the size of the system. The observed fluctuations are related to dilation: when a closed cell (fixed total volume) is used, effective viscosity increases, fewer fluctuation events are observed, and at high enough volume fraction the confined system becomes completely jammed.

Concentrated colloidal and particulate suspensions can undergo discontinuous shear thickening (DST) when flowing, characterised by a dramatic increase in viscosity above a certain threshold of stress [1]. DST is thought to be closely related to flow-induced jamming, which can be defined as the conversion of a nominally liquid system into a solid by imposed stress [2, 3]. Jamming and DST can lead to processing complications, such as erratic flow patterns, density waves and fluctuating apparent viscosity and pressure, in a wide range of applications from pharmaceuticals, construction, coating, printing and painting to chemical processing, foods, cosmetics and mining. Unpredictable and often catastrophic consequences appear also across geology, environmental phenomena and biology, including earthquakes, landslides, blood flow interruption and pedestrian jamming. Shear thickening and jamming materials do, nevertheless, have useful applications. Strong increase in energy dissipation with shear makes them useful for applications such as enhanced protective clothing[4]. Controlled conversion between solid and liquid offers potential for 'smart' technology such as robot hands: in the liquid state the 'hand' can mold itself to an object of any shape, while in the jammed state lifting forces can be applied to move the object around[5] .

There remain significant questions about the mechanisms of DST and jamming. Previous work has explored key aspects including hydrodynamic clustering and lubrication [6, 7], frustrated dilatancy [8–10], the statistics of force chains and 'fragility'[11] and interparticle friction [12–14]. In this Letter we demonstrate, using a simple experimental technique, that flow of a concentrated suspension in the shear-thickening regime involves intermittent macroscopic force fluctuations and dilational events, and we investigate how the statistical characteristics of these events are determined by volume fraction, system size and the nature of geometrical boundaries. Our measured statistics of intermittent flow invite parallels with quite different systems such as clogging hopper flows of grains and jamming flows of animals and pedestrians [15]. Our results imply that the rheology of concentrated suspensions, *i.e.* how bulk quantities such as apparent viscosity and time-averaged shear rate depend on volume fraction, is determined in the thickening regime by the statistics of these intermittent jamming/dilation events, *i.e.* the dynamic formation of jammed networks and their subsequent breakup by dilatancy. While the instantaneous response of the shear-thickening material thus appears unpredictable and intermittent, statistically its behaviour remains well-defined.

We use concentrated suspensions of sterically-stabilised spherical polymethylmethacrylate (PMMA) colloids, suspended in a mixture of decalin and tetralin solvents. The particles have radius 488nm and a low size polydispersity of approximately 5%, as measured with dynamic light scattering. This widely-studied system is a good approximation to a hard-sphere suspension where the particles interact only through steric repulsion (at least until flow stresses are high enough to overcome this and friction becomes important [13, 14]). The rheology, shear thickening and jamming of the PMMA system have been closely examined in a range of previous studies [9, 14, 16]. To our knowledge how the statistics of intermittent forces relate to shear-thickening rheology has not been reported for such a simple shear-thickening system, although the role of fluctuations in shear thickening has been qualitatively reported on by various authors[2, 16–18].

Experiments are carried out in a simple home-built Couette device consisting of two concentric cylinders, with the inner cylinder rotating to shear a sample confined between the cylinders. (We have checked that Tay-

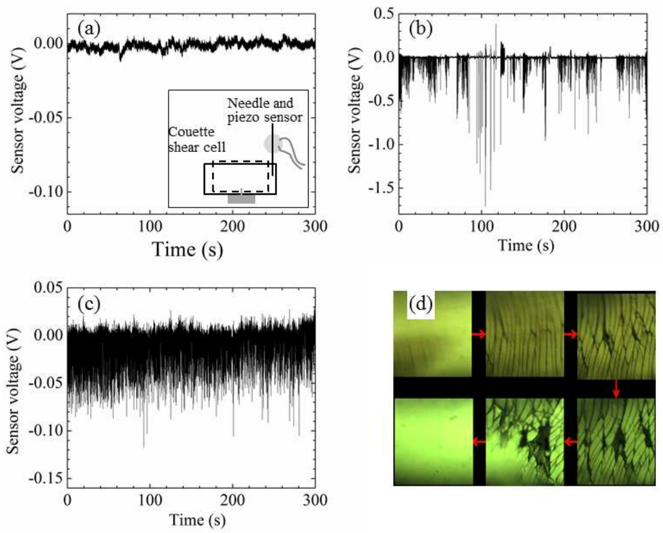

FIG. 1. Sensor voltage output during shear, for fixed stress and gap size 5mm. (a) $\Phi = 0.55$ (b) $\Phi = 0.56$ (c) $\Phi = 0.61$. (d) Time-sequence of images of top of sample (time increasing clockwise from top left image, time between images = 0.5s), showing appearance and disappearance of cracking at free surface. Inset to (a): schematic of experimental setup showing piezo needle sensor inserted into sample.

lor instabilities, which can occur with a rotating inner cylinder, are not observed at the shear rates and viscosities involved here.) Intercylinder gap widths studied include 2.5mm, 5mm and 10mm. The gap can either be open at the top (which is the case for most of the experiments described below) or tightly closed (see final discussion about the role of dilation). Rotation is provided by a small motor and rotation rate measured using a tachometer. While we do not directly measure torque or stress, using a calibrated motor with fluids of known viscosity we can define the relationship between applied motor voltage and shear rate to arrive at an estimate of the applied shear stress: as a test of precision and calibration we have compared viscosity measured with this home-made device with measurements using a commercial cone-plate rheometer and find agreement to within 5%. Our novel addition to the device, inspired by the old-fashioned vinyl record-player pickup, is a fine needle [inset to Fig 1(a)] inserted from the top of the gap into the bulk of the sheared suspension and connected to a piezo sensor. The sensor produces a voltage signal proportional to movement of the needle.

For steady shear of a simple fluid of non-jamming suspension (for example glycerol, water, or PMMA at low volume fraction Φ) the sensor shows low intensity noise [Fig 1(a)]. However, in jamming flows, we observe substantial intermittent peaks in the signal indicating macroscopic force-impulses in the sample that generate momentary deflection of the needle. Fig 1 shows example voltage traces over time during shear for various volume fractions at a fixed applied stress (motor supply voltage), with the top of the cell open to the atmosphere. (Measurements last up to a few minutes after loading, and repetitions at different times after loading show that there is no effect of solvent evaporation or particle sedimentation on this timescale.) We see a sudden onset of impulses from the needle device between $\Phi = 0.55$ and $\Phi = 0.56$. As Φ increases further the typical magnitude of the peaks decreases, but their frequency increases. Visual observations of the open top of the sample [Fig 1(d)] show 'cracking' patterns that dynamically appear and disappear that we intepret as signals of dilation of the particles into the liquid-air boundary. These visual events show qualitatively the same onset, severity and frequency dependence on Φ as the peaks measured through the needle sensor. Instantaneous shear rate shows similar temporal variation, although there is a well-defined average which is constant in time (see below). We propose that the intermittent deflections of the sensor correspond to dilation events in response to dynamic jam formation. By a 'jam' we mean a particle configuration where shear-induced forces can be transmitted macroscopically through the bulk of the sample via interparticle frictional contacts[2, 13, 14]. Imposed shear stress which, outside the thickening regime or in the absence of a jammed configuration, would generate laminar shear deformation of the sample, is instead transmitted to the boundaries. Dilation (macroscopic deformation) at the open boundary enables break up and deformation of the jam and hence subsequent flow. In this interpretation, our fluctuating voltage peaks, caused by macroscopic movement of the needle sensor, are the signal of these intermittent break-up (macroscopic deformation) events. The peak magnitude is indicative of the 'size' of the jam (or rather the instantaneous extent of deformation that occurs when it breaks up by dilation, see below), and its time indicates the moment when the jam collapses.

Fig 2 shows that peak magnitude is exponentially distributed. (Individual peaks are identified as signals above a noise-level threshold estimated from the signal in a non-jamming fluid: the statistical results are not sensitive to small changes in the chosen threshold.) If we interpret peak magnitude as proportional to a measure of jam 'size' this distribution indicates that there is a characteristic jam size which is constant over time at a given Φ and given stress. The Φ-dependence of the inverse of the characteristic peak magnitude (inset to Fig 2) demonstrates a lower critical value Φ_c for onset of jamming/dilation, and an upper limiting maximum Φ_j, with approximate scaling as a power of $(\Phi_j - \Phi)$. While there is insufficient data at higher Φ to make a fully quantitative fit of Φ_j, the data are consistent with a value around the random close packing limit $\Phi_j \approx 0.64$, where the inverse characteristic peak magnitude diverges or, equivalently, jam size

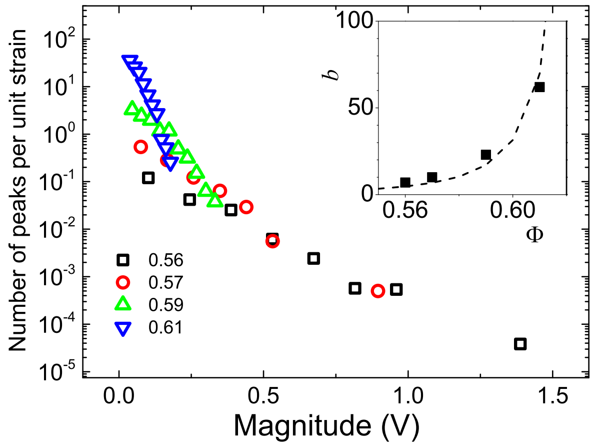

FIG. 2. Distribution of voltage peak magnitudes at fixed stress for different Φ. Inset: Φ-dependence of the inverse characteristic magnitude b from exponential fits to the distributions. The dashed line shows a function $(\Phi_j - \Phi)^{-\gamma}$ with $\Phi_j = 0.64$ and $\gamma = 2.75$.

shrinks to zero. Meanwhile decreasing Φ toward the critical value for onset of peaks, Φ_c, sees the characteristic jam size diverge. At first this behaviour seems counter-intuitive since jamming becomes more severe, not less, as Φ increases. However, as well as peak magnitude we must take into account the frequency of jamming events. As we show below, while jam size diverges toward Φ_c, jam probability shrinks to zero, meaning that jamming is no longer observed below Φ_c; conversely, while jam size shrinks toward Φ_j, jam probability diverges, meaning that at some Φ_j jamming is permanent. A useful measure of total 'jamming activity' may thus be formed from the product of jam size and jam probability, see below.

The 'fragile matter' model[11] and recent results on the role of interparticle friction in DST[13, 14] suggest a possible physical interpretation of this jam size/jam probability behaviour. Assume that some degree of local strain of the particle configuration in a given region is possible before this strain leads to a 'locked' network of direct frictional contacts (a jam), *i.e.* a network that then requires dilation to continue to deform[2]. The ensuing dilation event, by allowing separation of frictional contacts, releases this local strain as a sudden deformation producing a voltage peak with magnitude proportional to the released strain. It seems reasonable to expect the released strain and therefore the voltage peak magnitude to be proportional to the degree of local strain achieved prior to the jam. Decreasing Φ decreases the probability of frictional contacts so that more local strain is typically achievable before the locked network requires dilation, hence, as observed, dilation events generate larger peaks at lower Φ; but the decreasing probability of frictional contacts also means such events become increasingly rare.

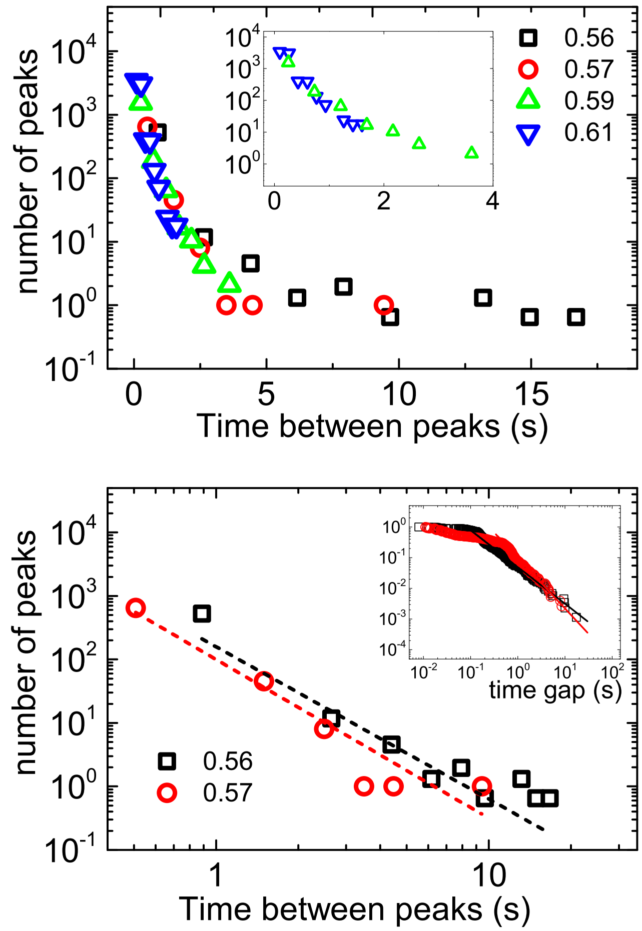

FIG. 3. Distribution of times between subsequent peaks for different Φ. Upper plot and inset shows shift toward exponential distribution for $\Phi = 0.59$ and $\Phi = 0.61$. Lower plot shows power law fits to data from $\Phi = 0.56$ and $\Phi = 0.57$. Inset to lower plot: cumulative probability $P(\Delta t \geq T)$ and power law fits.

The microscopic details of this picture remain speculative, of course: experiments that both resolve microscopic particle contacts and reveal large structural cooperative features on the fast timescale of intermittent jamming are challenging. Comparison of results with computer simulations is in progress.

Jam probabilities are explored further in Fig 3 where we show the distribution of times between consecutive events (voltage peaks) for a range of Φ. At intermediate Φ (0.56, 0.57) we observe an approximate power-law distribution with an exponent of around -2. If as above we interpret peaks as dilation events marking the collapse of a jam, and assume that the time between consecutive events is proportional to the lifetime of the jam whose collapse is indicated by the second event, then the power law distribution of jam lifetimes in Fig 3 invites comparison with similar distributions of 'clogging lifetimes' (periods when the flow outlet is clogged and flow is halted) in hopper flow [19] and even animal flow[15], despite the obvious differences in flow geometry and 'particle' characteristics.

At first sight such a comparison breaks down at higher Φ such as 0.61, where the temporal distribution is closer to exponential. However interpreting the time between peaks as the lifetime of a jam assumes that *only one jam happens at a time.* As Φ increases multiple jamming and collapsing events are likely to overlap in time, and no single lifetime distribution can be obtained from the time between peaks. (The approximately exponential distribution that we observe at higher Φ indicates that these multiple overlapping jams occur more or less independently of each other.) With a larger gap width between the concentric cylinders, reducing the frequency of jamming events at a given Φ [Fig 4(a)], the distribution of time between events for the higher Φ shifts to power-law, supporting our proposal that the underlying distribution of jam lifetimes is a power-law but is visible only for jam probability small enough that the detector 'sees' individual events that do not overlap in time.

That we observe typically larger magnitude but rarer events with increased gap size draws another interesting parallel with clogging flows in hoppers, pedestrian jamming *et.c.* In hoppers, a similar scale effect is observed: increasing outlet size leads to longer periods between formation of arches (jams), implying that, as might be expected, arch formation across a larger outlet is less probable. In our case, jam formation across a larger system similarly becomes less probable, allowing more time and thus more local strain buildup before formation of a jam, resulting in less frequent but larger dilation events. This has processing implications *e.g.* for materials transport: using a larger pipe may reduce the likelihood of jamming, but when jams do occur they will be more severe. In hopper flow, the 'severity' of jamming is measured via the number of particles that flow between jams, which is found to be exponentially distributed. This can be understood by considering a fixed probability of a given particle flowing without jamming: how many particles flow before an arch halts flow is then a simple statistical problem analogous to how many sequences of 'heads' of a given length occur before the first 'tail' when tossing a coin. By analogy, in continuous shear flow if there is a fixed probability of each 'incremental' unit of strain of the particle network leading to a jam, this would produce an exponential distribution of jam size (jams involving a given strain), as we observe.

As mentioned, a measure of total 'jamming activity' may be formed from the product of jam size and jam probability, or in practice simply by summing peak magnitudes over a fixed total strain. Fig 4(b) shows that this quantity, E_j, is *independent* of gap size at fixed stress and Φ, because while jams become rarer with increasing size, their magnitude also increases. The total jamming activity during a unit of strain is thus an intrinsic function of the fluid and the applied stress, independent of system size, while the statistical distribution of activity, *i.e.* how jam formation/breakup is shared out into a set of events over time and the individual magnitude of these events, is influenced by the system size and geometry.

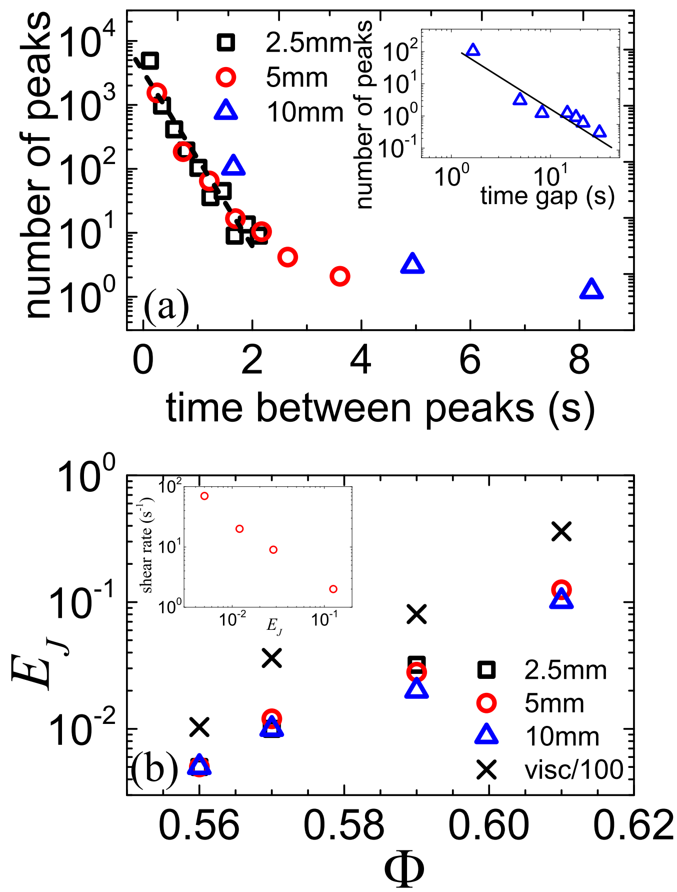

FIG. 4. (a) Distribution of times between peaks for $\Phi = 0.59$ at different gap sizes, demonstrating transition toward power-law behaviour (inset) as gap size increases and jam probability decreases. (b) Sum of peak magnitudes per unit strain, E_j, *vs* Φ showing independence from gap size. Crosses show the Φ-dependence of apparent viscosity calculated by dividing stress by measured shear rate (scaled by factor of 100 for clarity of comparison), demonstrating scaling with E_j. Inset: E_j *vs* shear rate, for gap size 5mm, showing inverse proportionality.

Fig 4(b) also shows that the Φ-dependence of E_j matches that of the apparent viscosity, obtained by dividing the (fixed) applied stress by the measured average shear rate at each Φ (the absolute value of viscosity is shown scaled by a constant factor for clarity of comparison in the figure). The inset to Fig 4(b) confirms this: the average shear rate at a given Φ is inversely proportional to E_j at that Φ. This link between the statistics of intermittent jamming/dilation and the 'averaged' bulk rheology implies that in shear thickening the reduction in shear rate, or equivalently the increase in apparent viscosity, as Φ increases, is directly related to the Φ-dependence of the statistics of jamming/dilation events.

Finally we briefly mention the role of dilation. Detailed study will be reported elsewhere, however it is useful to qualitatively compare behaviour of identical samples in the open cell and in the same geometry but with a tightly-fitting lid placed on the cell (incorporating a small hole for the needle sensor). With the closed system the sample cannot dilate, at least as long as cell walls are strong enough to resist imposed flow stress, and this results in a reduction in measurable fluctuations and a decrease in average shear rate (which means, at fixed stress, an increase in apparent viscosity). Dilation is frustrated, and more energy is dissipated by viscous and interparticle friction losses, *i.e.* the apparent viscosity increases. By $\Phi = 0.61$, at the limited stress we can apply through the motor, the system does not shear at all when dilation is disabled: the input stress is balanced against elastic response of the particles and boundary, and all input energy is stored elastically[2, 8].

To conclude, DST flow of a concentrated colloidal suspension is associated with intermittent macroscopic force fluctuations and dilation events, whose statistics indicate underlying universality of thickening and jamming phenomena across a wide range of quite different materials and flow scenarios. We suggest that the statistics can be understood qualitatively by considering the build-up of local strain until networks of frictional contacts form at the scale of the system that must subsequently dilate to allow flow. The dependence of total jamming 'activity' per unit strain on Φ mirrors the apparent viscosity: the growth of viscosity with Φ in the thickening regime is controlled by the statistics of intermittent jamming. However while average behaviour is intrinsic to the material, changing geometry and system size changes the scale and frequency of individual jamming events, with potential implications for applications. The key role of dilation as a response to jamming events further indicates the importance of boundaries and interparticle forces in jamming problems.

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