Extreme events-driven glassy behaviour in granular media

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Abstract. – Motivated by recent experiments on the approach to jamming of a weakly forced granular medium using an immersed torsion oscillator (*Nature*, **413** (2001) 407), we propose a simple model which relates the microscopic dynamics to macroscopic rearrangements and accounts for the following experimental facts: 1) the control parameter is the spatial amplitude of the perturbation and not its reduced peak acceleration; 2) a Vogel-Fulcher-Tammann–like form for the relaxation time. The model draws a parallel between macroscopic rearrangements in the system and extreme events whose probability of occurrence (and thus the typical relaxation time) is estimated using extreme-value statistics. The range of validity of this description in terms of the control parameter is discussed as well as the existence of other regimes.

Introduction. – Granular-matter physics [1,2] is a very interesting laboratory for addressing open problems of non-equilibrium statistical mechanics, such as the nature of slow glassy dynamics [3] and jamming [4], the mechanisms of pattern formation, the physics of avalanche phenomena. In this context, recent experimental results have shown [5, 6] the intriguing analogy between the way a perturbed granular medium progresses towards complete rest by decreasing the amplitude of external taps, and the vitrification of glass-forming materials.

This slow glass-like granular behavior raises one of the main questions in the framework of granular matter, namely the link between the macroscopic response to an external perturbation and the microscopic (or mesoscopic) processes occurring at the scale of the single particle. Recently, it has been conjectured [7] that the occurrence probability of *rare events* might explain the origin of the slow dynamic behavior of granular matter. Such rare events arise when the largest externally induced vibration in the granular medium overcomes some suitable threshold, triggering in this way a macroscopic "fracture" of the granular solid, that is a grain rearrangement. In this paper we further develop this idea and provide a model close
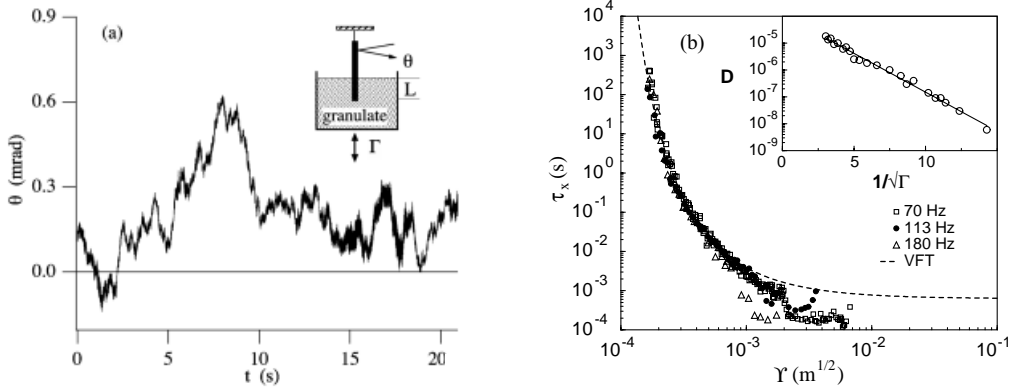
Fig. 1 – (a) Typical time series $\theta(t)$ obtained at $\Gamma = 0.056$ and $f_s = 113\,\text{Hz}$. Inset: sketch of the immersed torsion oscillator. (b) Relaxation time τ_x *vs.* Υ for various forcing frequencies f_s. Dashed line: VFT fit $\tau_x = A\exp[B/(\Upsilon - \Upsilon_0)^p]$ with $A = 6.5 \times 10^{-4}$, $B = 0.0072$, $p = 0.95$, $\Upsilon_0 = 1.37 \times 10^{-4}\,\text{m}^{1/2}$. Inset: diffusion coefficient (arbitrary units) D *vs.* $1/\sqrt{\Gamma}$, obtained directly from time series such as in (a).

to the intuitive picture given by experiments and which may be of interest for similar systems such as glasses or disordered systems [8–10].

Experimental results. – We first recall the basic experimental results, and fig. 1, which is in part adapted from previously published work [5], is used to introduce the method and summarize the basic data. The dynamic behavior of the perturbed granular medium is deduced from *noise measurements*. The granular noise, denoted $|\theta(f)|^2$, is obtained by detecting the irregular motion of a torsion oscillator (fig. 1a), deeply immersed into the perturbed granular material, and taking the squared amplitude of the Fourier transform of the observed data series. The granular medium is perturbed by shaking the container by well-isolated taps, or by continuous vibrations. An accelerometer on the container measures the intensity of the perturbation, quantified by the reduced peak acceleration $\Gamma = a_s\omega_s^2/g$, written in terms of the amplitude a_s and the frequency $f_s = \omega_s/2\pi$ of a sinusoidal shaking, and the acceleration of gravity g. Below the ideal fluidization limit at $\Gamma_f = 1$, and at low frequency (below the natural frequency of the oscillator), we observe $1/f^2$ noise spectra.

This low-frequency noise is the structural, or *configurational* noise. This is easily understood considering tapping experiments, where after each tap the granular system completely stops in a static configuration, which in turn determines an angular position of the immersed oscillator. A series of such taps drives the granular medium from one static (or jammed) configuration to another, and the corresponding series of static (or "low-frequency") angular positions $\theta(t)$ of the immersed oscillator probes this process, which displays $1/f^2$ noise. Before going further, we emphasize some experimental aspects: first, when continuous vibrations are used, the low-frequency spectrum is the same as the one seen in tapping experiments [5]. This gives us a very rapid and efficient method to measure the noise, and most of the data are obtained in this way. Second, our measurements are performed after the sample has been prepared in a reproducible state. In particular, after having been poured into the container, the granular medium is subject to very strong vibrations (large Γ) whose intensity is progressively reduced until reaching the value of Γ, where the measurements are performed. This discards the strong compaction effects occurring during the first run on a loose granular system, should

we have initiated the measurement directly at a low Γ after pouring. The procedure in fact brings the system directly into an almost stationary state which should correspond to the reversible curve described in [3].

In order to have a deeper insight into the fluctuations of the rotation angles, we have directly used the time series of $\theta(t)$ (one example is shown in fig. 1a) to compute the structure factor $S(\tau) = \langle(\theta(t+\tau) - \theta(t))^2\rangle$ for different values of Γ. The obtained linear dependence with τ allows us to determine a diffusion coefficient $D(\Gamma)$, which features an $\exp[-B'/\sqrt{\Gamma}]$ dependence at low Γ, as displayed in the inset of fig. 1b. Moreover, the $1/f^2$ noise resulting from the time series also gives important hints on the nature of the slow glass-like granular dynamics: since $|\theta(f)|^2$ is proportional to a diffusion coefficient, its inverse at a given frequency can be considered to be proportional to an intrinsic configurational relaxation time, τ_x, *i.e.*, $1/|\theta(f)|^2 = C\tau_x$. In order to obtain the constant of proportionality C, the noise data are compared with susceptibility measurements [7], since a peak in the loss factor (*i.e.* a peak in the tangent of the argument of the complex susceptibility) arises when $\omega_p\tau_x = 1$, where $f_p = \omega_p/2\pi$ is the frequency at which the susceptibility is measured. This gives approximately $C = 2\pi1500\,\mathrm{mrad}^{-2}\mathrm{s}$.

The following important points can be deduced from the experimental measurements. First, previous experiments [6] provide evidence for a parameter of the form $\Upsilon = b\sqrt{\Gamma}/\omega_\mathrm{s} = b\sqrt{a_\mathrm{s}/g}$, with b a constant. This key *control parameter* determines the "level" of the low-frequency noise, as shown in [6]. In other words, whatever amplitude a_s and frequency f_s of the perturbation is, provided $\Gamma < 1$, the noise only depends on Υ. (In fig. 1b we set $b = g^{1/2}$ and the control parameter is $\Upsilon = \sqrt{a_\mathrm{s}}$. Notice that, since we have not changed g, experiments only prove that the control parameter is proportional to the square root of the perturbation amplitude, *i.e.*, $\Upsilon \propto \sqrt{a_\mathrm{s}}$.) Another indication for a control parameter like Υ comes from the direct measurement of the diffusion coefficient (inset of fig. 1b) which shows an $\exp[-B'/\Upsilon]$ form, in agreement with the measurements of τ_x. The physical signification of this empirical control parameter will be discussed in detail below.

Summarizing, from noise measurements as a function of Γ at different shaking frequencies f_s, and having determined the relationship between $|\theta(f)|^2$ and τ_x from susceptibility measurements, we obtain the curves shown in fig. 1b, namely a plot of the configurational relaxation time τ_x as a function of the control parameter Υ. Secondly, the data for small Υ in fig. 1b have been fitted with the expression $\tau_x = A\exp[B/(\Upsilon - \Upsilon_0)^p]$, which gives in particular $p = 0.95$. Since the observed exponent is close to $p = 1$, we assume from now on that the configuration relaxation time is well described by the standard Vogel-Fulcher-Tammann (VFT) expression

$$\tau_x \simeq A\exp[B/(\Upsilon - \Upsilon_0)]. \tag{1}$$

In this VFT form, Υ is the empirical control parameter playing the role of temperature (which does not mean that it *is* a temperature!), and Υ_0 is the value of the control parameter where the configuration relaxation time scale diverges([1]).

Model. – Before going further, it is important to identify the granular regime we are addressing. It is evident from fig. 1b that Υ appears as the relevant control parameter only for external shaking with $\Gamma < 1$. In fact, the scaling described by eq. (1) ceases to hold for values of Υ which correspond, for each f_s, to values of Γ around unity. This is an important point

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([1])Actually, the data for τ_x can also be fitted by the simple exponential form $\tau_x \simeq A\exp[B/\Upsilon]$, as well as by the form $A\exp[B/(\sqrt{\Upsilon^2 - \Upsilon_0^2})]$, at least for $\Upsilon \gg \Upsilon_0$; we shall come back to this point on the discussion of the model. (An experimental estimate of Υ_0 gives $1.37 \times 10^{-4}\,\mathrm{m}^{1/2}$, which corresponds to a displacement of about $19\,\mathrm{nm}$. Notice that Υ_0 is very sensitive to the exponent p, and should be considered only as an order of magnitude.)

and the model is supposed to be valid for $\Gamma \leq 1$. In this regime no fluidization occurs and the granular medium can be considered in a *quasi-solid* phase, where geometric frustration plays a major role. During a periodic vertical shaking of amplitude a_s and frequency f_s, with $\Gamma < 1$, the granular medium moves as a whole, following the movements imposed to the container. At the same time, the torsion oscillator immersed in the medium is fixed in the laboratory reference frame, *i.e.*, it moves with respect to the medium with the same periodicity imposed by the shaking. It turns out that it is easier to explain what happens during the shaking if one inverts the point of view and considers the granular medium as globally immobile in the laboratory reference frame, while the torsion oscillator is subject to a vertical oscillatory forcing. Thus, at each period the torsion oscillator penetrates the granular medium for a length a_s and returns to the original position. In other words, one externally imposes a given displacement (or deformation) to the medium, while the stress exerted is not fixed.

What happens at the microscopic level? During the excursion of length a_s, the torsion oscillator will try to find its way inside the granular medium by advancing and possibly rotating of a certain angle in order to adapt to the reaction of the granular system. We have the following picture. For small excursions a_s (to be defined below) the medium stays in a given static (or jammed) configuration, and it is deformed either elastically or plastically. Eventually, the medium *fractures*, and jumps to another jammed configuration. Our immersed oscillator detects essentially each fracture event, and a time series represents a sequence of different jammed configurations. In contrast to an ordinary solid, for which a fracture is a unique fatal event, the granular solid is able to re-establish itself in a new jammed configuration and to support successive fractures.

Thus, we assume that the penetration/deformation process involves three distinct regimes: 1) An *elastic* regime for excursions a_s below a limit denoted a_s^0. For $a_s < a_s^0$ the system is able to absorb the imposed displacement elastically. (The experimental value of a_s^0 is material dependent and, as discussed below, we identify it with the displacement for which we observe the divergence at Υ_0, *i.e.*, $a_s^0 \approx 10^{-8}$ m.) In this regime the granular system responds as an elastic medium and the torsion oscillator returns to the original angular position at the end of the cycle. 2) A *plastic* regime, in which the imposed displacement is absorbed by the granular system by reorganizing the *internal stress network*, inducing irreversible rotations of the angular oscillator position, but without leaving the actual jammed configuration. 3) A *fracture* process in which the system is unable to further absorb the imposed displacement, and a *macroscopic grain rearrangement* (or internal avalanche) is required, resulting in a large jump of the torsion angle $\theta(t)$.

We now formalize this three-regime process. In the plastic phase, the torsion angle undergoes sudden (but very small) changes due to the reorganization of the internal stress distribution. In a first approximation, one can imagine that the variations of the torsion angle could be described by a discrete random walk, *i.e.* one has a Gaussian distribution for the torsion angles whose variance has to be computed by estimating the number of random walk steps the torsion oscillator undergoes for an excursion of length a_s. We shall see in the following how the hypothesis of a Gaussian distribution, partially supported by the results for the diffusion coefficient, is not crucial and can be relaxed without changing the main results.

Since the relaxation law (1) implies that below a certain value of a_s the relaxation time is always infinite, we can deduce the existence of an elastic threshold for a_s, *i.e.* a_s^0. This means that for $a_s < a_s^0$ no permanent deformation is produced in the system and the typical time for a macroscopic rearrangement is infinite, hence the existence of an *elastic phase*. For $a_s > a_s^0$ the reaction of the medium on the torsion oscillator induces irreversible rotations, hence the existence of a *plastic phase*. For these reasons a_s^0 can be considered as the length of the elementary step of the random walk.

Now, provided that $a_{\rm s} > a_{\rm s}^0$, the number of random walk steps n performed by the torsion oscillator for a displacement $a_{\rm s}$ will be proportional to $a_{\rm s} - a_{\rm s}^0$. If the elementary angle of rotation at each step is of the order of $\theta_{\rm el}$ we expect a Gaussian distribution for the rotation angles with variance $\sigma \propto \theta_{\rm el}\sqrt{n} \propto \theta_{\rm el}\sqrt{(a_{\rm s} - a_{\rm s}^0)/a_{\rm s}^0}$. This is a first indication that a parameter proportional to $\sqrt{a_{\rm s}} \propto \Upsilon$ (or $\sqrt{(a_{\rm s} - a_{\rm s}^0)/a_{\rm s}^0} \propto \sqrt{\Upsilon^2 - \Upsilon_0^2}$) could play the role of a control parameter, *i.e.* a role similar to a temperature in the sense that it determines the variance of the stress fluctuations induced indirectly by the advancing torsion oscillator.

Let us now try to explain the VFT behavior of the relaxation time. This time represents the typical time for the system to undergo a macroscopic grain rearrangement. The question can be then rephrased as follows: what is the probability that the torsion oscillator, in its angular random walk with variance proportional to $\theta_{\rm el}\sqrt{(a_{\rm s} - a_{\rm s}^0)/a_{\rm s}^0}$, will produce a macroscopic configurational change such that the system jumps to another static (jammed) configuration? Our hypothesis is that the fracture event takes places as an *extreme event* whose probability can be computed in the framework of the extreme order statistics [11]. Fluctuations in the torsion angle correspond to a stress redistribution and one expects larger angles to produce larger compression of the grain-chains involved. It is natural to expect that there will be some threshold value $\theta_{\rm f}$ for the fluctuating torsion angle above which the contact network will break and yield to a macroscopic rearrangement leading to a large jump for $\theta(t)$. We have then to look for the probability that, among the fluctuations of the torsion angle excited at each shaking (*i.e.*, at each penetration over $a_{\rm s}$), *the largest fluctuation will be larger than a given threshold* $\theta_{\rm f}$.

We define $Z_n = \max(X_1, X_2, \ldots, X_n)$, with $X_i = \theta_i/\sigma$. We search the probability that $\Pr[Z_n > x_{\rm f}] \equiv S_n(x_{\rm f})$, where $x_{\rm f} = \theta_{\rm f}/\sigma$ is the normalized angular threshold. The (cumulative) probability distribution is $\Pr[Z_n \leq x] \equiv H_n(x)$, and $S_n(x) = 1 - H_n(x)$. According to standard textbooks, for a normal parent probability density distribution, using the sequences given by $c_n = \sqrt{2\ln n} - (\ln \ln n + \ln 4\pi)/(2\sqrt{2\ln n})$ and $d_n = 1/\sqrt{2\ln n}$ [11], the probability distribution $H_n(c_n + d_n x)$ tends, as n increases, to a Gumbel probability distribution [12], *i.e.* $\lim_{n\to\infty} H_n(c_n + d_n x) = \exp[-\exp[-x]]$. For large n, the probability $S_n(x_{\rm f})$ is given by $S_n(x_{\rm f}) = 1 - \exp[-\exp[-(x_{\rm f} - c_n)/d_n]]$.

For $(x_{\rm f} - c_n)/d_n \gg 1$, the probability $S_n(x_{\rm f})$ can be approximated by an exponential function, *i.e.*, $S_n(x_{\rm f}) \approx \exp[-(x_{\rm f} - c_n)/d_n]$. Using the expressions for c_n and d_n given above, the above condition is verified if $n^2 \exp[\frac{-\theta_{\rm f}}{\sigma}\sqrt{2\ln n}] \ll \sqrt{4\pi \ln n}$. In our case $\sigma \propto \theta_{\rm el}\sqrt{(a_{\rm s} - a_{\rm s}^0)/a_{\rm s}^0}$ and the approximation is correct if $\theta_{\rm f}/\theta_{\rm el} \gg \sqrt{2n\ln n} - \frac{\sqrt{n}\ln\sqrt{4\pi\ln n}}{\sqrt{2\ln n}}$, which is a reasonable assumption given the experimental values of the parameters. Using the previous approximation the probability for a rare fracture event (corresponding to a configurational rearrangement) is

$$S_n(\theta_{\rm f}/\sigma) \approx \frac{n^2}{\sqrt{4\pi \ln n}} \exp\left[\frac{-\theta_{\rm f}}{\sigma}\sqrt{2\ln n}\right]. \qquad (2)$$

We expect that $\theta_{\rm f}$ will depend on the specific tribological properties of the grains, as well as on some geometrical properties of the system, such as the grain shape and size distribution. Thus, for a given granular system (*i.e.*, fixed n and $\theta_{\rm f}$) the fracture probability is determined only by σ. The inverse of the probability $S_n(\theta_{\rm f}/\sigma)$ determines the characteristic time for grain configuration rearrangements, *i.e.* the characteristic time of the macroscopic dynamics. One has then $S_n \propto \tau_x^{-1}$. Recalling that σ is directly related to the empirical control parameter $\Upsilon \propto \sqrt{a_{\rm s}}$, one finds

$$\tau_x \simeq A \exp\left[B/\sqrt{\Upsilon^2 - \Upsilon_0^2}\right], \qquad (3)$$

which, given the extremely small experimental value of a_s^0, is indistinguishable from (1) and also very close to an Arrhenius behavior. As already stressed, the experimental data do not allow to definitely discriminate between the three behaviors.

It is important to notice that in the present model the Gaussian distribution of the torsion angle in the plastic regime follows from the random-walk analysis. However, the precise form of the torsion angle distribution is irrelevant as long as it decays faster than any power law (see [10]). For instance for stretched exponential distributions as $\exp[-\alpha|\theta|^\beta]$, one would obtain again a Gumbel distribution for the largest fluctuation where the parameters c_n and d_n would be given, to logarithmic accuracy, by $c_n \simeq (2\ln n/\alpha)^{1/\beta}$ and $d_n = 1/c_n$. This leads to the conclusion that, independently of the precise form of the torsion angle distribution (*i.e.* under very mild assumptions on it) one gets the same result for the relaxation time provided the variance of the distribution is proportional to $\sqrt{(a_s - a_s^0)/a_s^0}$.

Conclusions. –   We have proposed a microscopic model which is able to explain some experimental results for the relaxation dynamics of a granular medium described [5,6]. The crucial hypothesis is that at low Γ ($\Gamma < \Gamma_f$, where $\Gamma_f \simeq 1$ in our experiments) the macroscopic dynamics is controlled by extreme events of the stress fluctuations in the system: if the variance of these stress fluctuations, driven by the imposed penetration of the oscillator over the distance a_s, is proportional to $\sqrt{(a_s - a_s^0)/a_s^0}$ (as it turns out if the microscopic torsion angle fluctuations are Gaussian), then a macroscopic change of $\theta(t)$ can occur only if such microscopic fluctuations overcome a threshold, *i.e.* if an extreme fracture event takes place. These simple ingredients immediately lead to an activated-like, VFT-like character of the relaxation time. The deviation of the relaxation behaviour from the Arrhenius law is explained by the existence of an *elastic threshold* a_s^0, which is material dependent: for $a_s < a_s^0$ the system can absorb the perturbation without any rearrangement.

For $\Gamma > \Gamma_f$ the situation is quite different: at each cycle most of the induced fluctuations in the system are large enough for a macroscopic rearrangement to occur, and the dynamics is not controlled any more by rare events. In this fluid-like phase, structural (configurational) changes are not rare events. Recently, Philippe and Bideau have identified, using a geometry very similar to the one of our experiments, a threshold value of the order of 1.2 above which the system is able to reach a stationary state with a density relaxation ruled by a stretched exponential law [13]. From this point of view, we speculate that the threshold value for fluidization could also mark the boundary between a glassy region (low Γ), where the relaxation is driven by extreme isolated events, and a quasi-liquid region (high Γ), where the system is able to reach a stationary state. It is important to stress that the threshold value for fluidization can depend on the geometry of the container. In particular, we expect the threshold value to increase a lot for highly confined geometries where the effect of the boundaries is strong. This could be the case for the Chicago experiments [3], where a narrow and tall container is used. If the effective threshold for this experiment was large (for instance $\Gamma_f \simeq 3$–4) this could explain why they observed a very slow relaxation even for values of Γ well above 1.

The present work represents only a first step in the direction of a better link between the microscopic dynamics of granular media and the macroscopic response to an external perturbation. From this point of view, various experiments can be thought of to clarify several points: the actual functional form for the relaxation time as well as the dependence of the elastic threshold a_s^0 on the material, the dependence of the control parameter on the acceleration of gravity g, the exploration of the fluid-like regime ($\Gamma > \Gamma_f$). Finally the experimental setup described in this paper could allow the measurement of some thermodynamical properties of granular media: in particular the existence of effective temperatures could be investigated by a suitable combination of susceptibility and noise measurements, in the spirit of recent

experiments on laponite [14]. Such experiments would open a way towards a comparison with recent theoretical predictions on a thermodynamical approach to granular matter [15, 16].

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