Vogel-Fulcher-Tammann–Type Diffusive Slowdown in Weakly Perturbed Granular Media

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We study the process by which a perturbed granular medium of millimetric-size particles reaches a "frozen" static configuration by decreasing perturbation intensity. The granular system is perturbed by isolated taps or by continuous vibrations. The granular process is observed by an immersed oscillator, which is used as a low-frequency "thermometer." A diffusive noise is seen until well below the fluidization limit, and the approach to the frozen configuration arises according to a modified Vogel-Fulcher-Tammann (VFT) behavior. As a function of an empirical control parameter with unit of time one recovers a standard VFT form.

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A granular system is a collection of macroscopic particles interacting solely via contact forces. In the absence of external perturbations, the system stays indefinitely in a static configuration, with all the particles at rest. External perturbations, e.g., isolated "taps," will move the system from one static configuration to another, and so on. This driven granular process is a very interesting problem of statistical mechanics of its own, but it can also be seen as a paradigm for a more general problem in solid state theory, namely, the nature of glass and the glass transition. With this in mind, in pioneering experiments by Knight *et al.* [1,2], isolated vertical taps were applied to a collection of millimetric-size glass beads, and the granular density after each tap observed. The time evolution of the compaction process was characterized by a slow dynamics, with the density still evolving after 10^5 taps. The phenomenology was indeed reminiscent of the behavior of glasses, and the problem has received growing attention [3]. In fact, analogies to the glass dynamics are observed in the behavior of a wide variety of macroscopic and microscopic physical systems, suggesting there might be a common underlying process known as "jamming" [4].

Here we report a study where a granular system is externally perturbed, either by isolated taps or by continuous vibrations, and the intensity of the perturbation is progressively reduced until the granular medium reaches a "frozen" static configuration, in which it will remain trapped. We characterize the statistical properties of the process by observing the irregular, Brownian-like deflection of a torsion oscillator immersed at some depth into the granular medium. The oscillator moves a step back or forth each time the grains change configuration. In the appropriate experimental conditions, operationally defined below, the oscillator can be seen as a "thermometer" sensing the configuration noise. The method provides evidence for diffusive slowdown and jamming [5], as seen in usual glass-forming liquids [6]. A control parameter with unit of time apparently plays a key role in the dynamics, since as a function of this parameter the slowdown is accounted for by a standard VFT form.

Our granular system is composed of glass beads of diameter 1.1 ± 0.05 mm. The granular material is contained in a metallic bucket of 150 mm height and 94 mm diameter, filled to a height h. The rotating probe of a torsion oscillator [5,7] (see Fig. 1b inset) is immersed at a depth L into the granular material. Except for the immersed probe, the oscillator is otherwise mechanically isolated from the container. The granular system can be perturbed using a vertical shaker. In a continuous vibration experiment, the shaker is driven by a sine wave at the frequency f_s, and a spectrum analyzer captures time sequences of the oscillator angular deflection, $\theta(t)$. In a tapping experiment, the container is submitted to isolated taps, separated by a time t_w, and where a single tap consists of one complete cycle of a f_s-sine wave (see Fig. 1a inset). A voltmeter is used to measure the sequences of the angular deflection, θ_n, measured about t_w s after each tap, with n the number of taps. The intensity of the perturbation is quantified by an accelerometer fixed on the container, which measures the reduced acceleration, Γ, related to the amplitude a_s and pulsation $\omega_s = 2\pi f_s$ of the sine wave by $\Gamma = a_s \omega_s^2/g$. For $\Gamma > 1$ single particles start to "fly," and the medium is fluidized. Notice that the grain size is large enough to discard interstitial gas effects [8] and moisture-induced effects [7]. The experiments are conducted in well compacted granular samples, submitted to a large number of taps at each Γ, and in practice further compaction is negligible [1,2].

We are interested in the statistical properties of the granular medium driven from a static configuration to the next. One expects that such properties can be extracted from a tapping sequence θ_n, each point being measured well after the end of the tap, when all the grains are at rest. When all motion has stopped, the oscillator is jammed in a given static configuration, and the power spectra, i.e., the squared amplitude of the Fourier transform of such tapping sequences, can be considered as the *configuration noise.* The results of such an experiment, with the probe deeply immersed (this guarantees that the oscillator does not disturb the intrinsic granular dynamics, as shown below), are reported in Fig. 1a, which shows the

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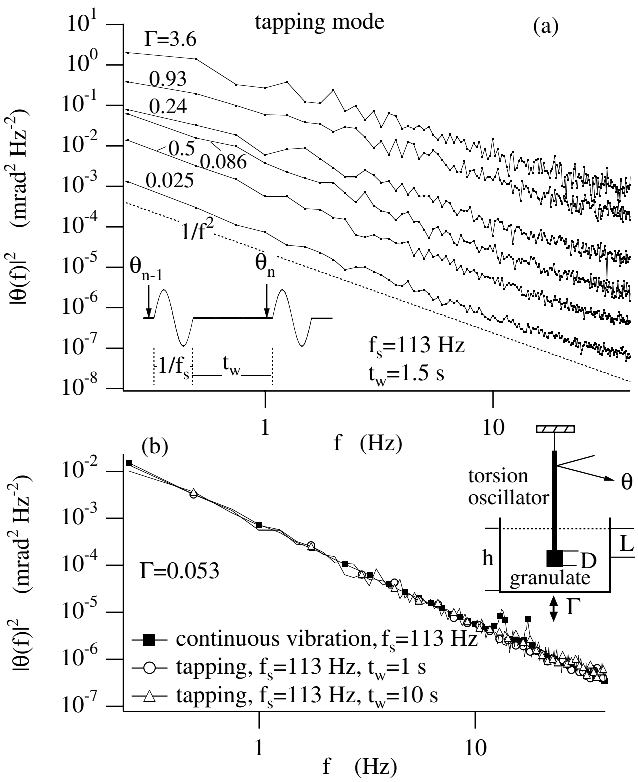

FIG. 1. (a) Power spectra obtained in the tapping mode, for different tapping intensity Γ. Here $h = 130$ mm, $D = 20.2$ mm, and $L = 65$ mm. Each curve is an average of about 10 sequences, 454 points long. The unit of time is defined as the point number times $1/133$ s (see text for details). Inset: Typical voltage fed to the shaker to produce taps. (b) Superposition of tapping and continuous vibration spectra with same Γ. (For clarity, only one symbol every four data values is marked. This is also the case in Fig. 2b, the Fig. 3 inset, and Fig. 4.) Inset: Sketch of the immersed torsion oscillator. The probes are either cylinders or cylinders with an enlarged terminal part, of length D. The whole cylinders, in the first case, or only the enlarged parts in the second case, are covered by a single layer of glass beads, glued on by an epoxy. The effective radius is 2.5 mm.

power spectra, denoted $|\theta(f)|^2$, for various tapping intensities Γ. Here we define the unit of time as $t = n/f_s$ and f is in Hz: this permits the confrontation with continuous vibration later. We observe a noise spectrum which is essentially $1/f^2$. Deviation from the $1/f^2$ dependence arises at large Γ; however, in the low Γ regime the $1/f^2$ dependence is well followed. A $1/f^2$ noise spectrum is evidence that the underlying statistical process is a diffusive, random walk. In other words, the configuration noise is a *diffusion noise*. By the Wiener-Khintchine theorem [9], for a $1/f^2$ noise the value of the noise at a fixed frequency is proportional to the diffusivity. If we assume the $1/f^2$ noise can be extrapolated to zero frequency [10], then the inverse diffusivity is a measure of an intrinsic configuration (structural) relaxation time. To understand the process by which the perturbed granular medium evolves toward its final frozen configuration under tapping of decreasing intensity, we must study how this relaxation time changes with Γ.

We need first to establish the appropriate experimental conditions under which the oscillator *senses without disturbing* the granular medium. For this we have used continuous vibrations, and we emphasize two points before proceeding further.

(1) The oscillator, and in general *any* massive object immersed in a granular medium [11], can induce extra perturbation in the medium by its inertia. In a tapping experiment, if the inertia acquired by the oscillator during the tap is large enough to overcome the resisting force in the granular medium, the oscillator will move after the end of the tap, by successive failure of the resisting grains' arrangement. Eventually, the oscillator comes to complete rest only well after the end of the tap, producing a sort of local "inertial heating effect." The same arises in a continuous vibration experiment and results in a viscouslike oscillation at about the natural frequency f_n. This is an unwelcome situation since such inertial effects will mask the intrinsic granular dynamics at very low Γ. However, since the pressure, thus the resisting force opposing the oscillator motion, increases with the depth, one expects that the inertial effect can be prevented by immersing the oscillator deeply into the granular medium.

Figure 2 shows that this is the case. Typical continuous vibration noise spectra are shown in Fig. 2a, over a large frequency range, for various depth of immersion L of the probe, and at a given Γ, well below 1. (For all L, there are peaks at f_s and high harmonics of f_s, which appear as soon as the probe "touches" the granular material. They are due to the nonlinear coupling of the oscillator to the external vibration via the granular medium. A discussion of this interesting phenomenon will be presented elsewhere.) The natural frequency, f_n, of the oscillator is 20 Hz in this case. For small L, say $L = 1$ to 6 mm in Fig. 2a, one observes a broad peak close to f_n, and a background increasing at low frequency. By increasing L beyond about $L_c \equiv L = 20$ mm in Fig. 2a, the noise tends to a *L-independent $1/f^2$ spectrum.* The broad peak close to f_n is the typical viscouslike inertial effect discussed above. The inertial effect disappears for large L, as expected. This behavior, with $L_c = L_c(\Gamma)$ depending on Γ, is seen until about $\Gamma \approx 1$. (Notice that L is still much smaller than the depth at which the pressure tends to a constant value in tall containers.) Figure 2b shows the noise level at 1 Hz, denoted $|\theta(1)|^2$, as a function of Γ, for various L. Here it is apparent that the noise level at $L > L_c(\Gamma)$ tends to an "envelope" (the dashed line in Fig. 2b). Data points on the envelope correspond to noise levels of spectra where viscouslike inertial effects have disappeared, as for $L > 20$ mm in Fig. 2a. The envelope is the same if different probe dimensions are used, or if the probe is immersed at different positions in the container, unless too close to the walls. Therefore, data points on the envelope represent L-independent $1/f^2$ spectra, and the envelope can

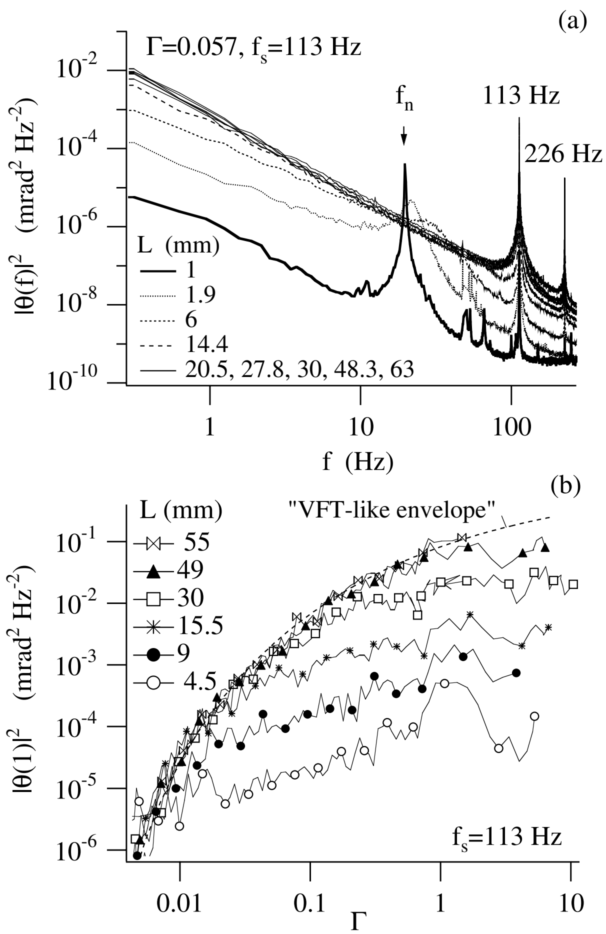

FIG. 2. (a) Power spectra obtained by continuous vibration, for different immersion depth L. Here a probe with $D = L$ is used, and $h = 96$ mm. Each curve is an average of 100 time sequences, 4 s long. (b) The noise spectra levels at 1 Hz, denoted $|\theta(1)|^2$, versus Γ for different L. The dashed line is a guide representing the "VFT envelope" (see text for details). Below $\Gamma = 1$ deviation from the envelope arises when inertial effects of the massive oscillator disturb the intrinsic granular dynamics.

be considered *intrinsic* to the granular medium. In other words, we have determined the conditions under which the "thermometer is in equilibrium" with the perturbed granular medium. The use of the word "equilibrium" is, of course, only pictorial, since energy is continuously dissipated.

(2) As one expects, we observe that at low frequency, roughly below f_n, the noise spectrum obtained by tapping *is the same noise* obtained by continuous vibration at the equal Γ. This is shown, e.g., in Fig. 1b, where a noise spectrum obtained by continuous vibration at 113 Hz and $\Gamma = 0.053$ is shown with two spectra obtained by tapping, with tap duration $1/113$ s and tap separation $t_w = 1$ s or $t_w = 10$ s, respectively. Defining the time scale for tapping as $t = n/113$ s, the three spectra overlap almost perfectly. This low-frequency overlapping is observed for all Γ, as can be seen in Fig. 3. (Figure 3 is discussed

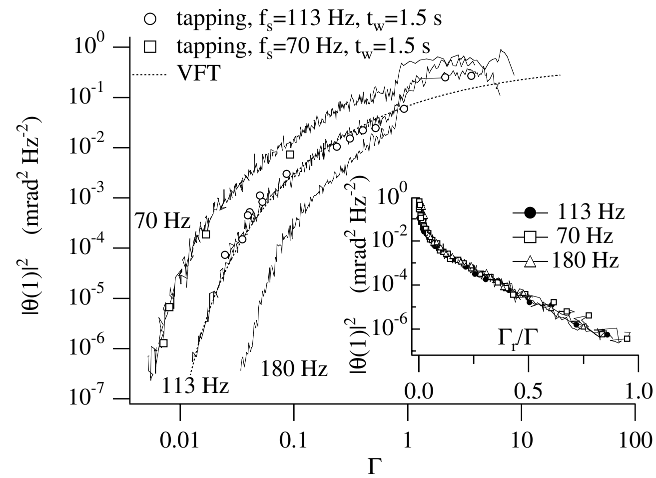

FIG. 3. The noise level $|\theta(1)|^2$ versus Γ for different f_s. Here $h = 130$ mm, $D = 20.2$ mm, and $L = 65$ mm. The continuous vibration curves (continuous lines) are composed of about 290 points, one point collected every 144 s. The same curves are obtained by reducing the rate $d\Gamma/dt$ by 2 orders of magnitude. Notice that the same curves are seen if probes with $D = 10.2$ mm, $D = 6$ mm, or $D = L$ are used. The data are collected by decreasing Γ, but the same curves are obtained by increasing Γ. The dashed line is a modified VFT fit (see text for details.) Inset: The noise level $|\theta(1)|^2$ versus Γ_r/Γ, with $\Gamma_r = 4 \times 10^{-7}$ mrad^2 Hz^{-2}.

in detail below.) Since the tapping spectra are measured when all motion has stopped after the tap, and the oscillator is jammed in a static configuration, we conclude that, in general, the continuous vibration spectra can be decomposed in a low-frequency *configurational component,* and

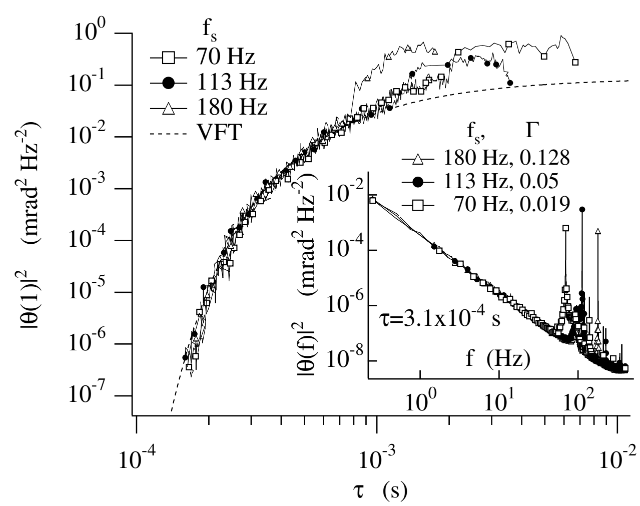

FIG. 4. The noise level $|\theta(1)|^2$ versus the parameter $\tau \equiv \Gamma^{1/2}/2\pi f_s$, for different f_s. The data are the same continuous vibration data of Fig. 3. The dashed line is a fit to a modified VFT form $|\theta(1)|^2 = C \exp[D(\tau - \tau_0)^p]$ (see text for details.). As a function of the control parameter τ the fit gives $p \approx -1$; i.e., one recovers a standard VFT form. Inset: Power spectra for different f_s but Γ selected such that the quantity τ is the same.

a high-frequency *viscouslike component* ascribed to inertial effects of the massive oscillator.

According to points (1) and (2), by immersing the oscillator at large depth and measuring the low-frequency noise of continuous vibration spectra, we now have a rapid experimental method to study the Γ dependence of the intrinsic noise level, i.e., a quantity proportional to the configuration relaxation time. Figure 3 shows the noise level at 1 Hz versus Γ, for different f_s. Here the filling level has been increased to $h = 130$ mm. Some points obtained by tapping are also shown. This figure reports our main findings: First, there is a feature at about $\Gamma \approx 1$ which apparently marks the fluidization transition. (The nature of this sharp feature is unknown at the moment.) Second, the noise level approaches zero following a critical behavior, namely, the previous envelope cannot be fitted by a power law. The inverse noise level, thus the intrinsic configuration relaxation time, diverges. The critical approach to zero can be described by a modified VFT form $|\theta(1)|^2 = A \exp[B(\Gamma - \Gamma_0)^p]$, e.g., for the 113 Hz curve, $A = 0.5$, $B = -2$, $\Gamma_0 = 0.005$, and $p = -0.4$, where Γ_0 is the perturbation intensity at which the noise level extrapolates to zero. This is strong evidence of a VFT-like diffusive slowdown [5] in the perturbed granular system by decreasing Γ. The same data are also shown in the Fig. 3 inset in a semilogarithmic plot of $|\theta(1)|^2$ versus Γ_r/Γ, where Γ_r is a reference vibration intensity at which $|\theta(1)|^2$ reaches a fixed small value. The inset shows that close to Γ_0 an Arrhenius behavior also fits the data. We underline that deviation from the Arrhenius behavior is small. Figure 3 provides evidence that the granular system evolves toward a frozen (glass) configuration *in a similar way* as the viscosity diverges, or the diffusivity approaches zero, in general glass-forming liquids [6] . On the basis of this analogy, we could say that below Γ_0 the perturbed granular medium is an "amorphous solid" or "glass," between Γ_0 and the fluidization limit $\Gamma_f \approx 1$ it is a "supercooled fluid," and finally it is a "fluid" above Γ_f.

Figure 3 also shows that the slowdown behavior, in particular Γ_0, depends on f_s, and confrontation with Fig. 2 shows that Γ_0 also depends on other external factors, such as the filling height h. Indeed, a single tap excites all sorts of "vibration modes." These vibration modes are very complicated and depend mostly on the tap duration $1/f_s$ and tap amplitude a_s, but also on the container form and material, on the total mass and filling height of the granular material, and possibly on the specific elastic and tribological state of the grains. These vibrations are expected to determine the details of the static configuration in which the grains will be trapped; hence they determine the $|\theta(1)|^2$ vs Γ "trajectory" toward the glass state [12].

To emphasize the f_s and a_s dependence of the slowdown behavior, Fig. 4 presents a "scaling" of the data of Fig. 3. We find that by plotting the data as a function of the scaled variable $\tau \equiv \Gamma^{1/2}/2\pi f_s$, all the curves overlap at low Γ. Moreover, fitting to a VFT-like form $|\theta(1)|^2 = C \exp[D(\tau - \tau_0)^p]$ gives now $C = 0.17$, $D = -0.003$, $\tau_0 = 5 \times 10^{-5}$, and $p = -0.92$, i.e., gives an exponent close to $p = -1$ as expected for a standard VFT form. The inset of Fig. 4 shows that, accordingly, the granular medium submitted to perturbations with different f_s, but with Γ selected such that τ is the same, have the same "configurational" $1/f^2$ noise spectrum. The empirical parameter τ has a simple meaning: $\tau = (a_s/g)^{1/2}$ is the time of flight of a body, initially at rest, falling for a distance a_s in the gravitational field. However, by no means is the relationship between a "granular temperature" and τ obvious [13]. Previous susceptibility measurements [14] have suggested that the parameter τ plays an important role, although its physical signification in the context of the dynamics and jamming of nonequilibrium driven systems remains to be elucidated.

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