Density fluctuations in granular piles traversing the glass transition: A grain-scale characterization via the internal energy

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The transition into a glassy state of the ensemble of static, mechanically stable configurations of a tapped granular pile is explored using extensive molecular dynamics simulations. We show that different horizontal subregions (“layers”) along the height of the pile traverse this transition in a similar manner but at distinct tap intensities. We supplement the conventional approach based purely on properties of the static configurations with investigations of the grain-scale dynamics by which the tap energy is transmitted throughout the pile. We find that the effective energy that particles dissipate is a function of each particle’s location in the pile and, moreover, that its value plays a distinctive role in the transformation between configurations. This internal energy provides a “temperature-like” parameter that allows us to align the transition into the glassy state for all layers, as well as different annealing schedules, at a critical value.

INTRODUCTION

Granular materials are ubiquitous in nature and have always fascinated scientists, Coulomb and Reynolds among them, due to their somewhat counterintuitive behavior (1). They have been considered as an additional state of matter in its own right (2) and although they are the second-most common form by which mankind handles materials (behind fluids), the way in which mechanical perturbations define its macroscopic properties are still not well understood. One of their key characteristic is that they tend to be at rest: Any input of energy provided to the system, whether by tapping, shearing, or tilting, is eventually dissipated through frictional contacts and collisions among the grains. Specifically, it requires such an external input of energy to be able to modify its configuration. Thus, the way in which the system behaves (fluid, solid, and gas) is strongly determined by the way this external energy is dispensed and the microscopic processes leading to its dissipation. Attributes such as friction, density, the granular contact network, container geometry, etc., will likely play a role in this process.

Although their propensity to stay at rest marks these materials as “athermal,” any boundary making it distinct from other, thermal states of matter are progressively weakening. For example, we have recently described the transition of a tapped granular pile into a glassy state (3) familiar from polymers, complex fluids, and frustrated magnets (4–9). For these disordered thermal materials, relaxation times increase for many orders of magnitude (and possibly even diverge) over a small range of temperatures (5), beyond which the systems remains out of equilibrium for any practical purpose. Whether the transition was approached gradually or via a hard-quench in the intensity of taps, we encountered the same phenomenology in the granular pile as is observed for other glass-forming materials. Although the distribution of fluctuations elicited by a perturbation within a granular medium may have profound differences to conventional thermal noise (10), as long as sizable fluctuations exist to activate events, glassy relaxation appears to be universal (11, 12).

In 1989, Edwards and Oakeshott (13) proposed the possibility of using the formalism of statistical mechanics to describe the properties of static granular materials. His theory considered as the statistical ensemble the set of static, mechanically stable configurations, which the system acquires after having dissipated the kinetic energy received by repeated perturbation. It set a milestone in the study of granular materials, as it hinted at some order behind the disorder. Since then, many studies (10, 14–22) have addressed these questions.

One of these studies is the well-known Chicago experiment (23), which presented a simple perturbation protocol able to create such a collection of states. This protocol consists of the repeated application of discrete “taps” to a granular pile confined inside a container, and the collection of static configurations obtained after the system has dissipated the injected energy. For taps with low (fixed) intensities, the system shows a logarithmic increase in density, or packing fraction $\phi$, with the number of applied taps (3, 10, 24, 25). However, executing a series of stepped annealing protocols, both for increasing and decreasing intensities of the perturbation, the system undergoes a fast “irreversible” transient of low densities and reaches a so-called “reversible regime.” There, the density of the static configurations becomes a function only of the intensity of the tap applied. The collection of static configurations at a given tap intensity serves as a setting to test Edwards’ hypothesis.

Using a protocol similar to stepwise annealing used in (23), but decreasing the tap intensity (represented conventionally by its reduced acceleration $\Gamma$) continually between taps at various variation rates ($\Gamma$), we have shown (3) that the packing fraction $\phi$ as a function of $\Gamma$ behaves in a manner resembling the glass transition found in thermal materials (5, 8, 26, 27). That is, at high $\Gamma$, $\phi(\Gamma)$ evolves independent of $\Gamma$, while for lower intensities, it splits into a separate branch for each $\Gamma$, reaching higher densities for lower $\Gamma$.

Here, we explore the origin of that transition in greater detail, accounting for the heterogeneous response of the system at different heights of the pile to the same tap (10, 28, 29), as imposed by gravity. As different layers attain different densities along the same protocol and enter a glassy state at different tap intensities $\Gamma$, we find that a...
As can be seen from Fig. 1A, the energy imparted by the tap travels along the height of the pile neither in an instantaneous nor homogeneous fashion. Instead, a complex process of energy transfer and dissipation ensues under the influence of gravity that we intend to study in more detail below. It is therefore not unexpected that the granular density of static configurations is often measured over narrow layers of constant height (23, 28, 29). To measure the packing fraction \( \phi \), we divide the entire system into 15 cylindrical sub-regions (“layers”), as schematized in Fig. 1B, stacked along the height of the pile.

In our simulations, we start from a high tap intensity and implement two distinct annealing protocols of decreasing intensity, one continuous and the other stepwise. For the first protocol, three different continuous rates of change were used. The packing fraction \( \phi \) of each layer is measured after each tap. To measure also the density fluctuations \( \Delta \phi \), corresponding to the “stationary state” at a given tap intensity, a stepped protocol inspired by (23) is performed. In this protocol, we apply a series of taps at each intensity \( \Gamma \) before decreasing it by decrementing the tap amplitude by an amount \( \Delta \phi \). Last, to verify the stationarity and reversibility of the produced states, we repeated the stepwise protocol in the reversed direction, i.e., for increasing intensities.

RESULTS AND DISCUSSION

Local density and its fluctuations

Figure 2 (A to C) shows \( \phi \) as a function of \( \Gamma \) for three \( \Gamma \) under continuous annealing for each one of the three layers located at the low, middle, and top of the pile marked in Fig. 1B. As previously reported (3), within each fixed layer, it can be seen that the data obtained at higher intensities \( \Gamma \) vary together, irrespective of \( \Gamma \), consistent with equilibrium behavior. However, for lower intensities, they split off into separate branches for different \( \Gamma \). As a consequence, the final density achieved for \( \Gamma \to 0 \) becomes a function of the protocol. Remarkably, the regime of \( \Gamma \) where this split occurs itself depends on the layer, suggesting that each strata in the pile transitions into a glassy state at a different tap intensity. Variations in the state between different parts of the pile due to gravity under the same perturbation have been noted previously (29). As we will show below,

![Fig. 1. Evolution of a single tap. (A)](image)

(A) Snapshots of the cylindrical silo illustrating the pile dynamics during a single tap. At time \( t = 0 \), the tap is applied (left). The purple line below the snapshots shows the schematic of the corresponding perturbation. The kinetic energy injected traverses the pile (from left to right) until it is fully dissipated through collisions and friction. The color scale reflects the instantaneous speed of each particle, increasing from blue over white to red. (B) A schematic of the 15 disk-like subregions (layers) used to measure the local packing fraction, with three representative layers (for low, middle, and top) highlighted for future reference.
these variations can be accounted for by the difference in the effective energy acting in each region.

Also shown in Fig. 2 (A to C) is the density as a function of $\Gamma$ obtained in the stepwise protocol. As many taps are spent at fixed values of $\Gamma$ (and many more overall compared to any of the continuous protocols), the densities for this protocol are well converged and provide an upper bound on the faster-moving, continuous protocols. Starting at high $\Gamma$, crosses mark the decreasing protocol, while open circles correspond to the same protocol but reversed after the decreasing protocol is completed at $\Gamma \approx 0.37$. Both sets of data demonstrate that there is only a minute aging effect on its reversibility that can be ignored for our purposes here.

Focusing on the corresponding density fluctuations $\Delta \phi$ obtained in the stepwise protocol, as shown in Fig. 2 (D to F), we find that irrespective of the direction of the protocol, the fluctuations exhibit identical features as a function of the tap intensity (with small quantitative differences in strength). It can be observed that $\Delta \phi$ as a function of $\Gamma$ remains almost constant for high intensities until it sharply peaks for lower $\Gamma$ values, before vanishing as the intensity further decreases. This single-peaked behavior for each layer is consistent with previously reported results (18, 28). However, we note that the right slope of each peak can be associated with the density transitioning into a glassy state, as it corresponds to the regime of intensities (highlighted by a blue stripe) where the splitting takes place in Fig. 2 (A to C) for the respective layer. This behavior is independent of the direction of the protocol, showing that it is a function of the tap intensity and not a result of a residual transient.

Thus, although the rise in fluctuations and the onset of glassy behavior are aligned for each layer, neither the intensity $\Gamma$ of the macroscopic perturbation nor the respective densities, as would be expected from Edwards’ hypothesis, allow aligning the data for all layers simultaneously.

**Effective energy of grains**

To align the onset of glassy behavior between different layers, we now examine in more detail the dynamic process by which each tap distributes energy throughout the system. It permits us to identify an effective energy $E_{\text{eff}}$ that quantitatively collapses the transition to glassy behavior for the different layers of the system. In particular, we find a specific threshold value $E_c$ for which particles with $E_{\text{eff}} < E_c$ always assume glassy arrangements, apparently. To this end, we now look at the grain-scale dynamics of the energy transmission and dissipation during the tap.

Figure 3A shows the kinetic energy $ke$ per particle [in units of $mgd$] as a function of time during the dynamics. In this example, we used an intermediate tap intensity of $\Gamma \approx 3.65$ and averaged over particles belonging to the three representative layers along the pile. Each exhibits an initial peak, whose value is consistent with $(A\omega)^2/(2gd) \approx 3.65$, the kinetic energy of a particle moving with the maximum speed reached by the perturbation. After this initial peak and a brief period of energy transfer between layers, each layer follows a “free-fall” behavior, decreasing its velocity until near zero before starting accelerating downward and reaching a last peak. It is the value of the last peak in $ke$ that interests us, as this is the kinetic energy acting in each region.
The low-energy regime most relevant to the glassy behavior, where departure from an entire pile to behave as a homogeneous solid. The inset shows the enlargement of this behavior is corroborated by the transfer, gain or loss, in the total (kinetic and potential) energy per unit time, shown in Fig. 3B. Here, as enlarged in the inset, we note the initial upward acceleration of particles due to the tap, followed by a coherent transfer of energy between layers from the bottom to the top, an effect similar to the “stacked balls demonstration” in introductory physics. The ballistic free-fall behavior leaves the total energy unchanged, which ends with a sequence of layer-by-layer crashes, progressively shorter and more intense, which dissipates the remaining kinetic energy we marked above as $E_{\text{eff}}$.

Figure 3C shows $E_{\text{eff}}$ as a function of $\Gamma$ for the same three layers represented in Fig. 3A. Open symbols correspond to the continuous protocol following $\Gamma_3$, while full symbols correspond to the $E_{\text{eff}}$ obtained through the stepwise protocol. It can be seen that $E_{\text{eff}}$ is largely independent on the protocol followed. A black dashed line represents $(A\omega)^2/(2gd)$, corresponding to the kinetic energy (in units of million gallons per day) that a particle would acquire by moving with the maximum speed reached by the perturbation. The inset in the same figure shows a close-up of the main figure for lower values of $\Gamma$. The “kink” corresponds to the intensity at which particles in a given layer stop separating and the system moves as a solid. For example, for the bottom layers, this happens at $\Gamma \approx 1$, as expected for a solid without elastic interactions.

### Data collapse as a function of $E_{\text{eff}}$

In the following, we use $E_{\text{eff}}$ to collapse our data. To this end, we first consider the stepwise protocol. Figure 4 (A and B) shows $\phi$ and $\Delta \phi$, respectively, for this protocol as a function of $\Gamma$, as in Fig. 2 but for all layers simultaneously. Layers 2, 3, 8, and 14 are highlighted by color. From Fig. 4B, we notice that the sharply peaked form of $\Delta \phi$ as a function of $\Gamma$ is present in all the individual layers but occurs, however, at different $\Gamma$ values, as pointed out in Fig. 2. Hence, when measured for the entire pile, these individual peaks in $\Delta \phi$ add up to form a “trough” as a function of $\Gamma$, as shown in the inset of Fig. 4B. This behavior is consistent with previously reported results (17, 31).

Figure 4 (C and D) shows $\phi$ and $\Delta \phi$ as a function of $E_{\text{eff}}$, respectively. Figure 4D shows an almost perfect alignment of the peaks of $\Delta \phi$ as a function of $E_{\text{eff}}$, supporting the hypothesis that $E_{\text{eff}}$ is a physically relevant parameter to characterize the state of the system. In particular, we mark $E_{\text{eff}} = E_{\text{c}} \approx 2$ as the effective energy where all fluctuations are simultaneously peaked as the “transition energy.” From Fig. 4C, it can be seen that this new parameter ($E_{\text{eff}}$) also aligns the inflection points for the densities $\phi$ in each layer in the same energy regime as the right slopes of $\Delta \phi$.

However, a better collapse for the density in Fig. 4C is hindered by behavior resembling a “finite-size” effect for higher energies, where the data splay out systematically with the height of the layer. Although this residual effect is minor, we attempt to collapse these data further by rescaling with height $z$, defining the new scaling variable

$$E' = (E_{\text{eff}} - E_{\text{c}})/z \quad (1)$$

This approach is entirely speculative and violates our desire to avoid system-specific parameters to unify the description. Figure 4 (E and F) shows $\phi$ and $\Delta \phi$ as a function of $E'$, respectively. While the collapse of the density $\phi$ in Fig. 4E improves somewhat, the density fluctuations $\Delta \phi$ become progressively distorted in width for $z \to 0$, see Fig. 4F.

While this result appears to support the existence of a geometric constraint on the dynamics depending on the column height,
following the proposal in (28, 32), this dependence could be explained using arguments based on the local expansion between particles mid-flight: From a configuration that would expand homogeneously, particles acquire a speed proportional to their relative height in the pile. Alternatively, this effect could also be associated with the contact network that particles form during flight, as has been discussed before (20). There it is shown that the existence of persistent contacts (i.e., contacts that are never broken) during a perturbation is a source of memory in the system. These possible dependencies will be investigated in more detail in future work.

Instead, we summarize all of our data for $\phi$ in Fig. 5. It demonstrates that there is a common transition into a glassy state occurring in the region of energies corresponding to the right slope of the peak in $\Delta\phi$ versus $E_{\text{eff}}$ (highlighted in light red). Above it, the scaling in Eq. 1 collapses the data for the stepwise and all continuous protocols in all layers simultaneously, whereas for $E_{\text{eff}} < E_c$, the data are splitting into separate branches for different annealing rates $\Gamma$ and between layers. Thus, the critical value $E_c$ provides a good prediction for the effective energy of particles at the glass transition. In contrast, the inset in Fig. 5 illustrates the breadth over which these transitions spread out for $\Gamma$ as the controlling parameter, reinforcing the value of our new approach.

In summary, we have shown that the density fluctuations, on approaching the glass transition from higher energies, rise to a sharp
peak for each layer of the pile, before vanishing as the perturbation intensity decreases. By analyzing the grain-scale dynamics of the kinetic energy transfer and dissipation during the perturbation process, we have been able to define an effective kinetic energy that allows quantifying the energy of the transition in a unified manner for all layers and intensities. This is a groundbreaking statement, further justified by our ongoing studies on alternate settings discussed elsewhere, as prior experiments and simulations on granular piles were able to agree on qualitative results but struggle to achieve any quantitative comparison within the geometries and parameters they used.

According to Fig. 4, the effective energy merely allows aligning the data but does not achieve by itself a satisfactory collapse of $\phi$, hinting at other relevant quantities affecting the transition internally, which we are currently investigating. Although the ad hoc rescaling with the height $z$ reduces the apparent height dependence and produces a rather satisfactory collapse of the density data in Fig. 4E (see also Fig. 5), it unnaturally broadened the density fluctuations for $z \rightarrow 0$ in Fig. 4F. Nevertheless, it provides a direction for further investigation into the role of other grain-scale, dynamic parameters, such as the network of granular contacts, or the dependence of the dissipated energy on the mean-free path (due to local expansion and frustration) among the grains. To conclude, we remark that it has been shown that $\phi$ by itself is insufficient to completely characterize the (static, mechanically stable) configurations of a granular system $\left(14, 18, 19, 28, 33, 34\right)$. For this reason, further work will also have to address the behavior under the scaling provided by $E_{\text{eff}}$ for other macroscopic quantities, such as the stress tensor of the system.

**MATERIALS AND METHODS**

Figure 1 illustrates the setup for the granular pile used in our MD simulations. It consists of a cylindrical silo of diameter $D = 2.4$ mm with 60,000 spherical grains of slightly bi-dispersed diameters $\left(1 \text{ to } 1.02 \text{ mm in equal number}\right)$, to reduce crystallization. The height of the granular pile is 12 cm, within a silo whose top (at 60 cm) was chosen high enough to ensure that the grains never interact with it. Within the LIGGGHTS (30) open-source software implementation, we also set a friction coefficient of $\mu = 0.5$, a young modulus of $Y = 10^8 \text{ Pa}$, a restitution coefficient of $\epsilon = 0.5$, a Poisson’s ratio of $\nu = 0.3$, and a density of $\rho = 2500 \text{ kg/m}^3$ for our grains. The initial condition for the pile is obtained by simply pouring the grains into the container.

The tap consists of a half sine-wave $A \sin (\omega t)$ with constant frequency $\omega = \left(2\pi/0.047\right) \text{ Hz}$, with the amplitude $A$ as the control parameter, applied to the silo by the movement of the entire container (both bottom and side walls). The dimensionless acceleration $\Gamma = A \omega^2 / g$, with the gravity acceleration $g$, is used to represent the tap intensity. Numerically, we consider that the system is static when the kinetic energy of the whole pile drops below a threshold of $10^{-1} \text{ J}$.

**Measuring local density of static configurations**

To measure the packing fraction $\phi$, we divide the entire system into 15 cylindrical subregions (layers), as schematized in Fig. 1B, stacked along the height of the pile from $z = 0.006875$ to $z = 0.11$ m. To reduce boundary effects, particles closer than 2 mm from the silo lateral walls, $\approx 7$ mm from the silo bottom, and particles on the surface ($z > 0.11$ m) are disregarded. Each layer defined in this way contains $\approx 2500$ particles.

Three different continuous rates of change were used: $\Gamma_1 = 0.00002 \text{ m/s}^2 / \text{g}$ per tap, $\Gamma_2 = \Gamma_1/4$, and $\Gamma_3 = \Gamma_1/8$. The packing fraction $\phi$ of each layer is measured after each tap. To this end, a Voronoi tessellation (35) of the whole system is performed, and the local density of each particle is obtained by dividing its volume by its corresponding Voronoi volume. The densities of those particles whose centers are in the subregion of interest are averaged to obtain the packing fraction $\phi$ of the corresponding region.

For the stepwise protocol, we decrement the tap intensity by $\Delta A = 0.00004 \text{ m}$ for the first seven steps and $\Delta A = 0.00002 \text{ m}$ for the remaining ones. For this protocol, the selection of the number of taps applied at each given $A$ needed to satisfy the condition of the system reaching the stationary state at that $A$ and to provide enough statistics for an accurate calculation of the density fluctuations. On the basis of preliminary inspection of our data, we found that 500 taps were sufficient. To avoid the transient regime between consecutive intensities, the first 250 taps at each $A$ are disregarded. Although this number is overly cautious at high tap intensities, it ensures that we only average over stationary states as the system evolves through its glassy phase. For the remaining 250 taps, we average over $\phi$, and $\Delta \phi$ is calculated as the SD of the mean.

**Measuring internal energies during the dynamics**

To measure the internal energy for each layer of the pile during the dynamics ensuing from the perturbation, we label particles with respect to the layers they reside in for the static configuration before the tap. We then track positions and velocities of all those particles during the dynamic process illustrated by Fig. 1A to calculate the average kinetic and gravitational potential energies per particle for each layer as a function of time. From the decrease in mechanical energy, we can deduce the dissipation particles from the given layer have experienced during the process.

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