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Emergent Strain-Stiffening in Interlocked Granular Chains

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Granular-chain packings exhibit a striking emerging strain-stiffening behavior despite the individual looseness of their constitutive chains. Using indentation experiments on such assemblies, we measure an exponential increase of the collective resistance force F with the indentation depth z , and with the square root of the number $\mathcal{N}$ of beads per chain. These two observations are respectively reminiscent of the self-amplification of friction in a capstan or in interleaved assemblies of sheets, as well as the physics of polymers. The experimental data is well captured by a novel model based on these two ingredients. Specifically, the resistance force is found to vary according to the universal relation: $\log F \sim \mu\sqrt{\mathcal{N}}\Phi^{5/8}z/b$, where μ is the friction coefficient between two elementary beads, b is the size of such a bead, and Φ is the volume fraction of chains when semi-diluted in a surrounding medium of unconnected beads. Beyond the intrinsic similarity with the polymeric glass transition, this system allows to study friction in complex assemblies, with practical implications for the design of new materials, textile industry, and biology.

In Nature and architecture, a wide variety of stable structures are formed from dense assemblies of randomly-distributed objects [1, 2], as beautifully illustrated by the various shapes of bird nests [3]. The aggregates can be made of elementary components with arbitrarily complex and sometimes living shapes, as in the case of fire ants [4]. Most physical studies focus on simpler but highly anisotropic objects, *e.g.* rods, that ensure a solid-like collective behavior and thus the great stability of their assembly [5, 6], even under large compressive stresses. However, there is no steric constraint between the constitutive objects that could hinder their motion within the aggregate. The mechanical stability of rod aggregates is thus intimately related only to their packing [10] and the solid friction [11] at the contact points. Further works have been devoted to the mechanics of assemblies made of more complex objects, such as stars [7], or U- and Z-shaped particles [8, 9]. In those cases, in contrast to rods, it is considered that steric constraints are the key ingredient for the observed increase of rigidity.

Apart from using rods or rigid nonconvex objects, there is a third approach to produce cohesive assemblies from non-interacting elements: randomly mixing soft slender objects, such as fibers [12–14] or granular chains [15]. In the latter case, a strain-stiffening phenomenon was reported experimentally in the absence of dilution, and was qualitatively related to the formation of loops between chains [16]. However, a quantitative theoretical mechanism is still lacking to date, despite the fact that the jamming of these systems might have some fundamental connexions with the glass transition of polymeric materials. Regarding the influence of particle shape, the packing of rigid rods is drastically affected by

their aspect ratio. For very thin rods, packing fractions as low as 0.2 are reported and theoretically described through the random contact equation [5]. For granular chains, however, the change is less impressive, the lowest packing fractions ranging between 0.4 and 0.5 [15]. There is no theoretical model describing this effect, but we could infer that the reduced influence of the shape anisotropy might be related to the high flexibility of granular chains.

In this Letter, we investigate the effective mechanical rigidity of granular-chain assemblies from indentation experiments. Our observations show an exponential growth of the resistance force with the product of two factors: the indentation depth and the square root of the number of beads per chain. The first factor is reminiscent of the self-amplification of friction in a capstan [19], as well as in interleaved assemblies of sheets [20, 21], where the increase in resistance is created by, and proportional to, the force exerted by the operator. The second factor points towards the central role played by topological constraints, in a way that is similar to polymer physics [22]. Therefore, we propose a novel interlocking model based on those two ingredients, and confront it to the experimental data. We find very good agreement, including the extended situation of semi-dilute chains within a surrounding medium of unconnected beads.

Our experiments were conducted on large, monodisperse, assemblies of granular chains, each chain being made of $\mathcal{N}$ connected steel beads of diameter $b = 2$ mm, with $2 \leq \mathcal{N} \leq 50$. The random packing of the ensemble was achieved through sharp taps until a constant packing density was reached. Using a ProbeTack system, the force needed to indent the assembly in its cylindrical container was measured as a function of the indentation

depth z , with a cylindrical probe of contact area 1 cm^2 being displaced at constant velocity. Low velocities, in the $0.1 - 1\text{ mm} \cdot \text{s}^{-1}$ range, were used in order to ensure a quasistatic regime, and to avoid a velocity-dependent drag force [18]. To avoid the Janssen effect [17], we used floppy containers molded with soft crosslinked Sylgard. The system size ($> 10\text{ cm}$ in all directions) was much larger than the diameter of the indenter (1 cm). Semidilute systems were also prepared and studied by mixing monodisperse granular chains ($\mathcal{N} \geq 2$) with unconnected beads ($\mathcal{N} = 1$) of the same size, resulting in a volume fraction $\Phi = V_{\text{chains}}/(V_{\text{chains}} + V_{\text{beads}})$ of chains, where V_i indicates the volume of species i .

We first start with $\Phi = 1$, *i.e.* with pure granular-chain assemblies. We recall that rod ensembles initially packed in a container can either retain their shape or fully collapse when the container is removed, depending on the aspect ratio of the rods [6]. As qualitatively shown in Fig. 1(a), our granular-chain assemblies exhibit a similar behavior: when initially packed in a cylindrical container, short-chain assemblies collapse and form a conical pile, like bare sand, while long-chain assemblies are able to retain their cylindrical shape – despite their individual looseness.

To understand this striking emerging rigidity, we systematically study the influence of the indentation depth z and the number $\mathcal{N}$ of beads per chain, on the force needed to indent such a dense granular-chain assembly. The indentation process involves some local plastic-like structural reorganization [17] characterized by small force discontinuities. Figure 1(b) summarizes our raw results averaged over different realizations. First, the force needed to indent the system increases with the indentation depth z . For the well-known case of purely unconnected beads ($\mathcal{N} = 1$), we recover a nearly-linear force-displacement behavior, as usually observed for small indentation depths [23], *i.e.* before the intruder feels the bottom of the container [24]. More interestingly, for $\mathcal{N} \geq 2$, this increase is not compatible anymore with an effective Hookean behavior, as the slope (*i.e.* the stiffness) itself increases with z , thus confirming the strain-stiffening scenario previously reported [16]. Secondly, the force and stiffness both increase sharply with the number $\mathcal{N}$ of beads per chain, indicating the influence of chain connectivity in the problem.

Previous systems exhibiting a self-amplified friction mechanism have suggested that the resistance essentially grows exponentially with the relevant control parameter, *e.g.* the angle in a capstan [19], or the Hercules number [20, 21] in interleaved books. Inspired by this idea, we thus replot the $\mathcal{N} \geq 2$ data of Fig. 1(b) using a log-linear scale. The result is shown in the inset of Fig. 2, and we observe the data to be consistent with an exponential dependence of the force with the indentation depth. Since we operate in a quasistatic regime, there is no drag force, and the resistance to indenta-

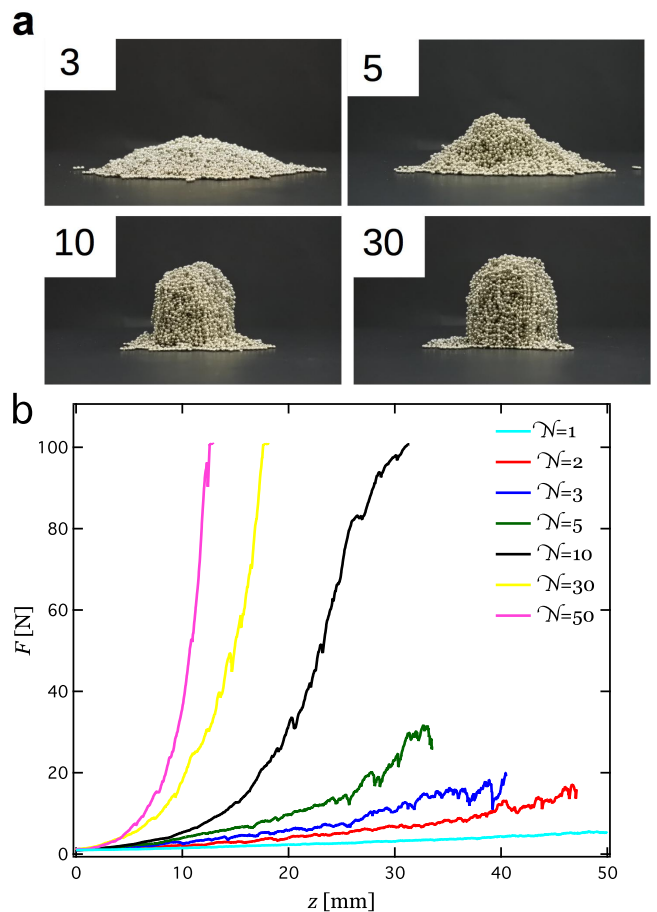


FIG. 1. (a) Snapshots showing the final states of dense ($\Phi = 1$) granular-chain assemblies after the removal of their cylindrical container, for various numbers $\mathcal{N}$ of beads per chain as indicated. (b) Indentation force F as a function of indentation depth z , for granular-chain assemblies with various numbers $\mathcal{N}$ of beads per chain as indicated. The cylindrical indenter has a 1 cm^2 base area and is displaced at constant speed. Each curve corresponds to an average over roughly 10 experiments.

tion should be only related to internal friction forces hindering the motion of the constitutive objects [18]. As a consequence, and in view of the measured exponential growth of the force, we postulate the existence of a first ingredient for the mechanism at stake in our granular-chain assemblies: a geometrically-induced self-amplification of friction. Furthermore, Fig. 2 shows the slope $d\log(F)/dz$ of the $\mathcal{N} \geq 2$ data as a function of the number $\mathcal{N}$ of beads per chain. The best power-law fit is good and gives $d\log(F)/dz \sim \mathcal{N}^{0.46}$ which is close to $d\log(F)/dz \sim \sqrt{\mathcal{N}}$. Interestingly, the latter exponent is reminiscent of the ideal random-walk statistics at play in a melt of polymer chains [22]. As a consequence, and in view of the fact that we are also dealing with flexible chains here, we postulate a second ingredient for

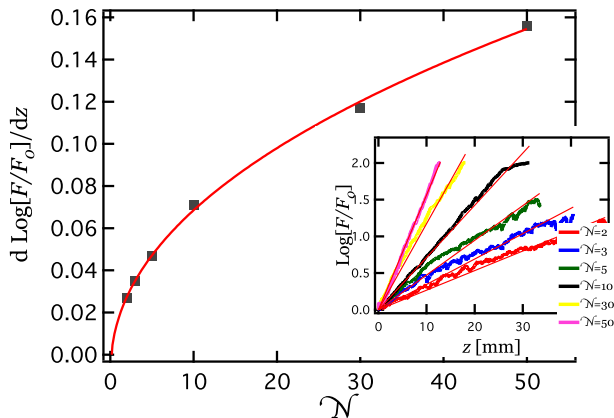


FIG. 2. Slope of the log-linear representation of the $\mathcal{N} \geq 2$ data of Fig. 1(b) (see current inset) as a function of the number $\mathcal{N}$ of beads per chain. The solid line indicates the best power-law fit $\simeq 0.03\mathcal{N}^{0.46}$. (Inset) Log-linear representation of the $\mathcal{N} \geq 2$ data of Fig. 1(b). The straight solid lines indicate linear fits in this representation.

the mechanism at stake in our granular-chain assemblies: the frictional resistance is essentially induced by contacts between chains. Note that for rod assemblies, where the components are non-flexible and with coordination numbers close to 10 [26], the average number of contacts per chain is quite different: it scales linearly with the rod length, as estimated by considering the number of rods sharing the excluded volume around a given rod [5].

Below, by combining the two above ingredients, we develop the proposed interlocking model. From the first ingredient on self-amplification of friction [19–21], one expects the increase dF of resistance force to be proportional to the friction coefficient μ , to the force F itself, and to the increase $\mathcal{M}dz/b$ in the number of locking points per chain when indenting further down by dz , with $\mathcal{M}$ being the number of such points in a slice of elementary depth b . Therefore:

$$\frac{dF}{dz} \sim \frac{\mathcal{M}\mu}{b} F. \quad (1)$$

$\mathcal{M}$ and $\mathcal{N}$ are now going to be connected by using the second argument based on the analogy with a polymeric system. It should be noted however that granular chains are not subjected to thermal noise, in contrast to polymer chains. The main problem is then to obtain reliable information on the average conformation adopted by granular chains, and on the number of contacts with other chains, for a system that is not controlled by the temperature. We here follow the suggestion of Edwards to keep a statistical approach in order to describe the structure of a granular state, even in the absence of thermal noise [25], which is justified by essentially replacing energy by volume in the microcanonical ensemble. It is thus assumed

that, in a random chain packing, the flexible chains do adopt an ideal coiled conformation characterized by their typical size $\sim \sqrt{\mathcal{N}}b$. We stress that self-avoiding effects indeed effectively cancel in polymer melts ($\Phi = 1$) [22], thus the Gaussian statistics invoked here. Since the assembly is dense, this result implies that the pervaded volume $\sim \mathcal{N}^{3/2}b^3$ of a single chain is in fact occupied by $\sim \sqrt{\mathcal{N}}$ chains. Assuming that only the interchain contacts generate interlocking, and that the chains are still short enough that two chains typically contact only once, we obtain:

$$\mathcal{M} \sim \sqrt{\mathcal{N}}. \quad (2)$$

Another way to get this relation is to consider that the release of $\mathcal{N}$ interlocking points over its size $\sqrt{\mathcal{N}}b$ are required to free a single chain, which yields $\mathcal{M}/b \sim \mathcal{N}/\sqrt{\mathcal{N}}b$.

Inserting Eq. (2) in the solution of Eq. (1) leads to:

$$\log\left(\frac{F}{F_0}\right) \sim \frac{\mu\sqrt{\mathcal{N}}z}{b}, \quad (3)$$

which is the observed form in Fig. 2, with $F_0 \approx 1$ N an arbitrary reference force. Since we remained at the level of scaling, we missed a prefactor in the right-hand side of Eq. (3). However, this prefactor can be estimated from the fit of Fig. 2, knowing μ and $b = 2$ mm. No specific cleaning procedure was applied here, in order to avoid any damage of the granular chains. Therefore, a thin layer of lubricant could likely remain between beads, and μ should then be considered close to 0.1, bringing the missing prefactor around 0.6.

We have thus demonstrated that our interlocking model describes well the $\mathcal{N} \geq 2$ data, in the case of pure chains ($\Phi = 1$). To avoid any coincidence, and validate the model further, we finally wish to test the polymer analogy by considering a semi-dilute regime of granular chains ($\mathcal{N} \geq 2$) mixed with unconnected beads ($\mathcal{N} = 1$). This situation corresponds to $\Phi^* < \Phi < 1$, where we introduced the critical dilution fraction $\Phi^* \sim \mathcal{N}b^3/R_F^3 \sim \mathcal{N}^{-4/5}$ at which dilute self-avoiding chains of size $R_F \sim \mathcal{N}^{3/5}b$ start to interpenetrate each other [22]. As shown in Fig. 3(a) in a log-linear representation, the measured indentation force F in such a semi-dilute case strongly increases with chain fraction Φ , while keeping the previous exponential behavior with indentation depth z . Similarly to Fig. 2, we then plot the corresponding slope $d\log(F)/dz$ as a function of Φ in Fig. 3(b), and find a best power-law fit of $d\log(F)/dz \sim \Phi^{0.69}$ that we would like to rationalize theoretically below.

First, for $\Phi > \Phi^*$ there is at least one interlocking point per chain and the self-amplification mechanism is maintained, so is Eq. (1). Therefore, our only task at this stage is to modify $\mathcal{M}$, and connect it to $\mathcal{N}$ and Φ . Essentially, the dilution of chains should reduce the number of interlocking points, from the previous melt-like

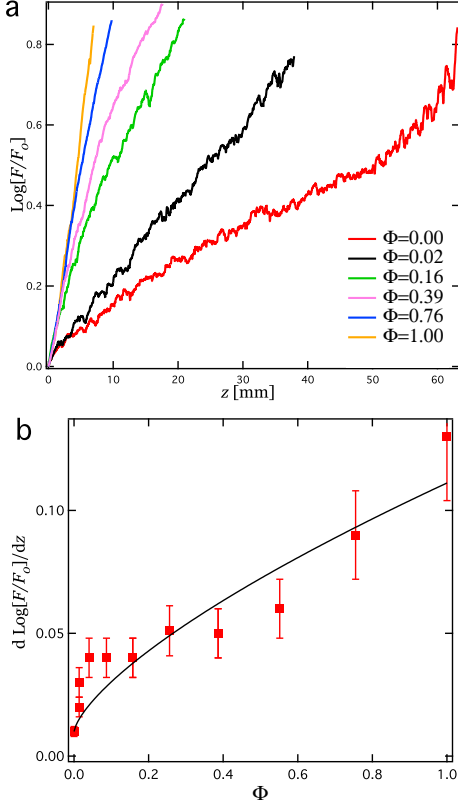


FIG. 3. (a) Log-linear representation of the indentation force F as a function of the indentation depth z , for granular-chain assemblies with $\mathcal{N} = 30$ beads per chain, semi-diluted in a medium of unconnected beads ($\mathcal{N} = 1$), with chain volume fraction Φ as indicated. The cylindrical indenter has a 1 cm^2 base area and is displaced at constant speed. Each curve corresponds to an average over 5-10 experiments. (b) Slope of the log-linear representation in (a) as a function of the chain volume fraction Φ . The solid line indicates the best power-law fit $\sim \Phi^{0.69}$, with an unspecified prefactor.

case $\mathcal{M} \sim \sqrt{\mathcal{N}}$ at $\Phi \sim 1$, down to $\mathcal{M} \sim 1$ at the critical dilution fraction $\Phi \sim \Phi^*$. According to the physics of semi-dilute polymer solutions in a good – athermal – solvent [22], each chain can be viewed as a succession of $\mathcal{N}/g$ independent units, the so-called blobs, of size ξ and containing a subset of g connected beads each. In such a renormalized picture, the whole solution can be viewed as a dense melt of blob chains, which implies $\Phi \sim gb^3/\xi^3$ as well as $\mathcal{M} \sim \sqrt{\mathcal{N}/g}$. In contrast, the portion of a chain inside a blob is in a purely dilute state (*i.e.* with only unconnected beads around) and thus has a self-avoiding conformation leading to $\xi \sim g^{3/5}b$. Combining those three relations, one gets:

$$\mathcal{M} \sim \sqrt{\mathcal{N}} \Phi^{5/8}, \quad (4)$$

that has the correct limiting values mentioned above, and that provides a theoretical exponent $5/8$ which differs by less than 10% from the measured value in Fig. 3(b).

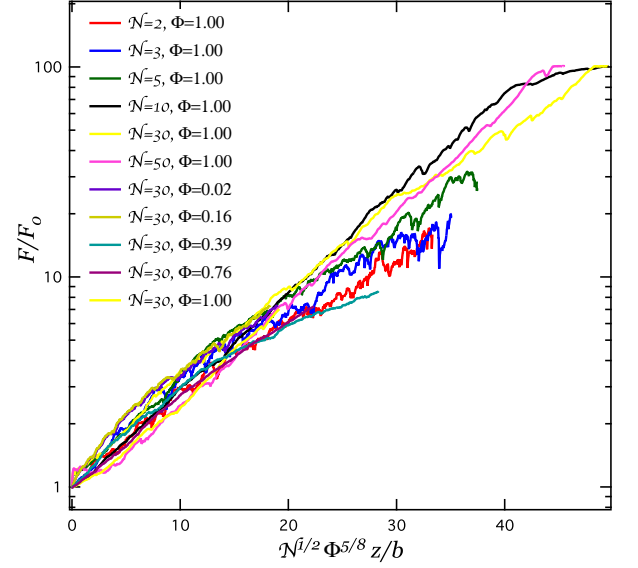


FIG. 4. Universal representation of all the dense and semi-dilute granular-chain indentation experimental data of Figs. 1(b), 2(inset), and 3(a), corresponding to $\mathcal{N} \geq 2$ and $\Phi > \Phi^* \sim \mathcal{N}^{-4/5}$, as predicted by Eq. (5). The ensemble corresponds to various numbers $\mathcal{N}$ of beads per chain and volume fractions Φ as indicated. Note that the dimensionless friction coefficient μ is not inserted in the x -axis as it is neither varied nor measured, and since there is anyway a missing prefactor in Eq. (5).

Finally, inserting Eq. (4) in the solution of Eq. (1) leads to the universal relation:

$$\log \left(\frac{F}{F_0} \right) \sim \frac{\mu \sqrt{\mathcal{N}} \Phi^{5/8} z}{b}, \quad (5)$$

that is valid for all Φ such that $\Phi^* < \Phi < 1$, and for which Eq. (3) is a limiting case when $\Phi \rightarrow 1$. The status of F_0 and the missing prefactor have already been discussed after Eq. (3) and are not modified here. Following the prediction of Eq. (5), all the experimental data of Figs. 1(b), 2(inset), and 3(a), corresponding to $\mathcal{N} \geq 2$ and $\Phi > \Phi^* \sim \mathcal{N}^{-4/5}$, should belong to a single master curve. The universal collapse indeed observed in Fig. 4 thus provides strong support for the proposed interlocking mechanism.

In conclusion, the emergent strain-stiffening behavior in granular-chain assemblies, as observed through indentation experiments, seems to originate from a self-amplification of friction due to polymer-like interlocking contacts. Interestingly, this system exhibits a novel self-amplification exponent ($1/2$, in chain length) with respect to the capstan (1 , in angle) [19] and interleaved-book (2 , in the number of pages) [20, 21] ones. It should be noticed however that, even if the model works well, it implicitly assumes long-enough chains for proper conformational statistics to be achieved. While we are confident

in the proposed explanation for long-enough chains, another mechanism might as well explain the observed mechanical behavior for the smallest-chain aggregates. Beyond the intrinsic polymer-mimetic feature of granular-chain assemblies, this study embodies a novel illustration of friction in complex assemblies, with practical implications for new materials, textiles [13, 14], and biology [27, 28].

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