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Polymer-like Statistical Characterization of Two-Dimensional Granular Chains

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Statistical behaviors of packing collections of granular chains in a two-dimensional container have been investigated experimentally. On compaction from their own gravity, the longer chains pack into a structure with lower packing density due to the prevalence of backbone loops. The packing of chains can be considered as the jamming of the granular system. The structure factor of packing chains shows scaling behavior $g(q) \sim q^{-2}$ in good agreement with dense polymer solutions. In addition, we compute various probability distributions of distances and estimate three crucial contact exponents, finding that the scaling behavior from granular chains is in accord with the theoretical expectation of polymers. Finally, an orientational anti-correlation of granular chains is observed by bond-bond correlation function, which agrees with the results in two-dimensional model of compact polymers.

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I. INTRODUCTION

Static packing structure of granular materials remains a puzzle of fundamental interests studied by many theoretical [1] and experimental [2] works. Recently Zou *et al.* investigated the static packing structure of granular chains in a three-dimensional cylinder by x-ray tomography, and found that the low packing density of long chains originates from the jamming of myriad semi-rigid rings [3]. They suggested that the packing of granular chains showed a close similarity to that in glass transition of real polymers, that is, the glass transition in real polymers might parallel a jamming transition in granular chains. Thus, the research on the static packing of granular chains is probably helpful for understanding the structural properties of the compact polymer materials.

Inspired by the insights in Zou's findings, one naturally speculates that some characterizations of two-dimensional granular chains are supposed to be, at least to some extent, similar to polymers in two dimensions as well. It is well known that static scaling laws for two-dimensional polymers have been theoretically established [4, 5]. For example, the size $R(N)$ of dense polymers suggested by de Gennes can be scaled as $N \approx R^D(N)$ where $R^2(N)$ is the average square end-to-end reduced distance, D is the spatial dimension and N is the chain length, respectively. Furthermore, computer simulations provide a theoretical analysis of static and dynamic behaviors of two-dimensional polymers[6–10]. In spite of substantial theoretical progress achieved above, polymers confined to two dimensions are experimentally difficult to be realized [11]. However, we indeed know that static and dynamic structures of two-dimensional granular chains can be directly visualized[12–14]. It is therefore plausible that the experimental research on static behavior of two-dimensional granular chains should assist the progress in indirect validation of polymer theory if the analogy with two-dimensional polymers can be drawn.

In this report we initially establish a static granular chain system to create and observe a variety of conformations of compacted granular chains. After recording every conformation by a high-resolution digital camera, we compute the packing density of granular chains as a function of the chain length. By extracting ball-centre coordinates of every spherical grain on the chain precisely, we calculate the structure factor for different chain lengths, finding that the scaling exponent in agreement with the result of two-dimensional dense polymer solutions. We also calculate scaling probability distribution functions for long chains, and discuss the similarity of scale behavior between granular chains and polymers. Finally, the bond-bond correlation from long granular chains is obtained to show the absence of long-range orientational order.

II. EXPERIMENTAL SETUP

Our experimental establishment consists of granular chains confined in a two-dimensional rectangular glass container (300 mm long, 150 mm high, and 3.28 mm wide), shown as Fig.1 (a) and (b). The inner surface of the container is cleaned as smooth as possible to diminish the friction. The granular chains with free ends are composed of metal spheres and rods. The spheres with a diameter of 3.06 ± 0.15 mm connect to each other via a rigid rod with a diameter of 0.70 mm. The rod between two spheres can stretch out and draw back freely, and the maximum length of the rod connecting two neighboring spheres is about 1.28 mm as shown in Fig.1 (c). The spheres can freely rotate around rigid rods, and the maximum bone-flex angle between adjacent spheres is denoted by θ_{max}. For convenience, the amount of spheres required

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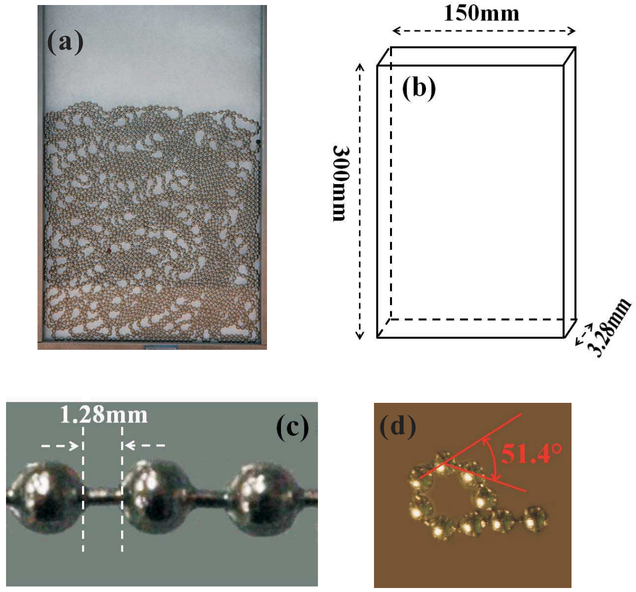

FIG. 1: (color online). (a) The photograph of a close-packed collection of a single long chain ($N = 2048$) confined in a two-dimensional container. The dyed spheres indicate both free ends. (b) Container dimensions. (c) Illustration of the length of the rod connecting two nearest spheres. (d) Minimum number of spheres required to form a loop on a semi-rigid chain, and the maximum bone-flex angle.

to form a smallest possible ring is used to characterize the stiffness of the granular chain instead of θ_{max}. In our experiment, the maximum bone-flex angle $\theta_{max} = 51.4^o$ and the minimum amount $\xi = 2\pi/\theta_{max} \approx 7$ shown as Fig.1 (d). To ensure sufficient number of spheres for data statistics, and leave space appropriate for spatial exchange of chains, chains are purposely filled to about 2/3 volume of the container [Fig.1 (a)]. Then we take advantage of a compaction method which has been successfully applied to the compaction of granular chains [14]. When the compaction is achieved, a high-resolution digital camera is used to take snapshots to capture static packing configuration of chains in the container. After repeating the compaction method many times, we record packing configurations, and from these photographs extract the coordinates of every ball center for following data statistics.

III. PACKING DENSITY AND STRUCTURE FACTOR

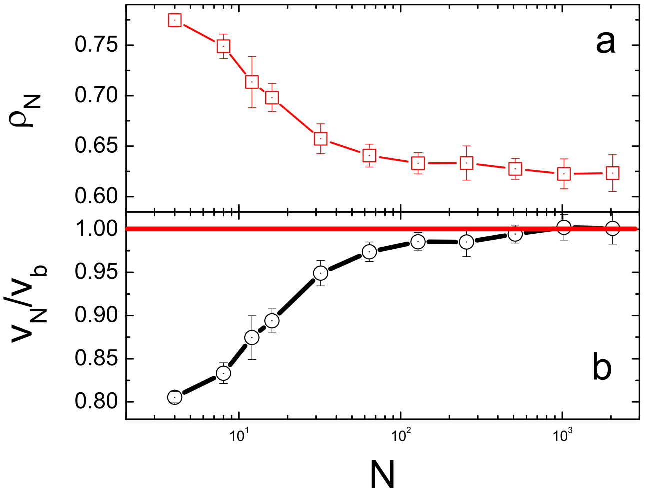

FIG. 2: (color online). (a) Packing density as a function of chain length N using a semi-logarithmic scale. Each point with error bar is the average of ten trials. (b) The plot of specific packing volume $V_N = 1/\rho_N$, normalized by the bulk ($N \to \infty$) value V_b, versus chain length N. The solid line indicates unity.

Here we define the static packing density as $\rho_N = A_0/A$, where A_0 is the area occupied by all chains in the container, and A is the total area including all chains and the voids among them. After every data point is averaged by 10 trails, the packing density is plotted as a function of chain length, that is, the number of spheres per chain, as shown in Fig. 2(a). We clearly observe that ρ_N declines monotonically with increasing N, from $\rho_N \approx 0.77$ for $N = 4$ to a saturation value $\rho_N \approx 0.62$ at large N on the characteristic curve. The plot is qualitatively similar to that of three-dimensional experiments in Ref.3, in which Zou *et al.* suggested that the glass transition temperature of polymers, T_g, and the specific packing volume, $V_N = 1/\rho_N$, of granular chains should be analogous quantities. The specific packing volume V_N can be described as the weighted sum of V_e and V_b, $V_N = V_b - 2l(V_b - V_e)/N$ [3], where V_b is the specific packing volume as $N \to \infty$, V_e is the contribution from the ends of chains, and $l \sim \xi$ is the correlation length of ends. The form of the specific packing volume is identical to the Flory law, commonly used to describe the dependence of the glass transition temperature on chain length and chain configuration, $T_g = T_{g,\infty} - K/N$, where K is a polymer-specific parameter. In the same way, we find that for the linear granular chains, the specific packing volume shown in Fig.2(b) can also be described by $V_N = V_b - 2l(V_b - V_e)/N$, where the best fit gives $l = \xi = 7$, $V_b = 1.60$, and $V_e = 1.49$. In the spirit of the jamming phase diagram[15] and the analogy proposed by Ref.3, we speculate that there might be a similar connection between polymers and macroscopic chains. In this sense, the specific packing volume of granular chains can presumably be analogous to glass transition temperature of polymers. Nevertheless, it is noteworthy that the observations do not prove that the polymer glass transition must be attributed to the jamming of chains.

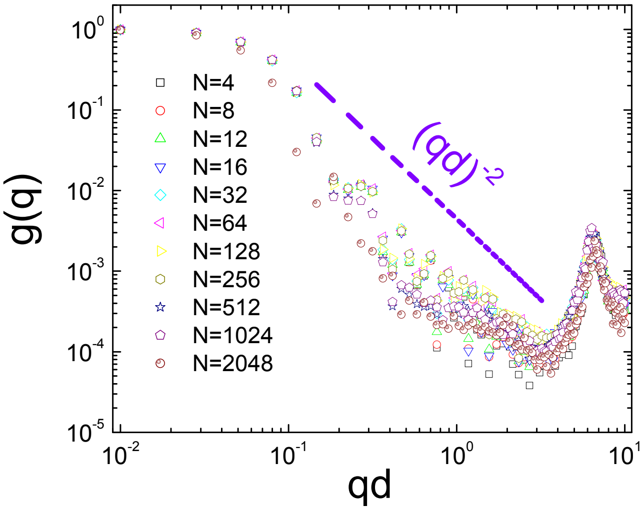

FIG. 3: (color online). The structure factor $g(q)$ of the chains obtained from the experiment as described in text, as a function of qd (d is the diameter of spheres on the chain) for different chain lengths. To characterize the decay in the intermediate regime of wave vector q, our results are compared to a power-law exponent -2, indicated by the blue dashed line to guide eyes. The peak near $qd = 2\pi$ corresponds to the average sphere spacing of chains.

In order to illustrate detailed packing structures of chains and over various length scales, we calculate the static structure factor defined as

$$g(q) =< g(\vec{q}) >= \frac{1}{M^2}\sum_{i=1}^{M}\sum_{j=1}^{M} < \exp[i\vec{q}\cdot(\vec{R}_i - \vec{R}_j)] > . \tag{1}$$

Here, $\vec{R}_i$ is the location of the ith ball and M is the number of balls in the system. We average $g(\vec{q})$ over all directions of the wave vector $\vec{q}$ and all conformations in the experiment to obtain $g(q)$. Fig.3 displays the structure factor $g(q)$ for the packings of granular chains from $N = 4$ to 2048. Usually, $g(q)$ for a linear polymer is expected to scale in the intermediate wave vector regime as $g(q) \sim q^{-1/\nu}$ [4]. Linear polymers in two dimensions of the dilute or semidilute solutions, or long granular chains confined within a flat container exhibits a fractal scaling behavior with the exponent $\nu = 3/4$ [5, 11, 12].

Here in Fig.3 the exponent $\nu = 1/2$, which indicates that our granular chains still maintain a relatively dense packing even though the avoided crossings are enough for long chains, is in agreement with the result of two-dimensional dense polymer solutions [4, 16].

IV. CONTACT EXPONENT

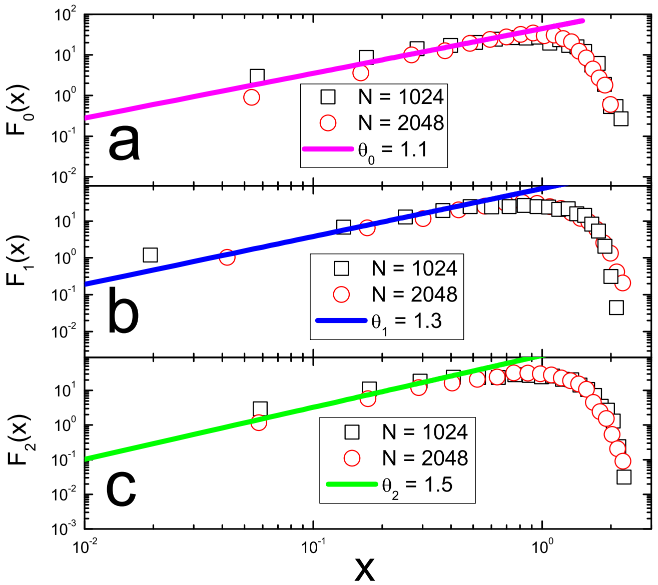

FIG. 4: (color online). Log-Log plot of the scaling probability distribution $F_i(x)$ for different chain lengths $N = 1024$ (open square) and $N = 2048$ (open circle) versus the scaling variable $x = |\vec{r}|/R_i$. The contact exponent θ_i is estimated by fitting the approximate scaling function x^{θ_i}, as shown by solid lines. All data come from 1000 original photographs.

In order to extract universal features of the granular chain structure, probability distribution function of the distance between two points which form a segment inside the same chain is always considered [4]. It is noteworthy that the distribution function in polymer science appears to be universal in D dimensions [17, 18] and can be described by [9]

$$G_i(\vec{r}) = R_i^{-D} F_i(x), \tag{2}$$

where $i = 0, 1, 2$, the scaling variable $x = |\vec{r}|/R_i$, $\vec{r}$ is the distance vector between two points, and R_i^2 is the second moment of the respective distribution. The scaling probability distribution functions $F_i(x)$, which is universal as well, for different chain length N and i are shown in Fig.4. For the case $i = 0$, $F_0(x)$ characterizes the end-to-end distance distribution function with the segment $S_0 = N$. When $i = 1$, $F_1(x)$ is the distribution function of the distance between two points with a segment $S_1 = N/2$; $i = 2$, $F_2(x)$ is the distribution of an inner segment space with $S_2 = N/4$. In Fig.4, $F_i(x)$ for long chain $N = 1024, 2048$ almost overlap completely in every panel. It is clearly shown that $F_i(x)$ exponentially decays for long distance $(x > 1)$. For short distance $(x < 1)$, the behavior of the universal function $F_i(x)$ in polymer is given by $F_i(x) \sim x^{\theta_i}$ for $x \to 0$ [17, 18], where θ_i is the contact exponent. In our experiment, fitting power functions are consequently used to produce final estimations $\theta_0 = 1.1$, $\theta_1 = 1.3$, and $\theta_2 = 1.5$ for $F_0(x)$, $F_1(x)$, and

$F_2(x)$, respectively. For three panels in Fig.4 we find that the scaling behavior measured from granular chains is in a good accord with the expectation of theory of polymers. In particular, in the region of $x << 1$, the similar scaling characteristics between granular chains and polymers further imply a possible unifying connection. However, it is important to point out that the contact exponents extracted from the power-law fitting disagree with the self-avoiding walk (SAW) model ($\theta_0^R = 11/24$, $\theta_1^R = 5/6$, and $\theta_2^R = 19/12$)[17] and the compact polymer model ($\theta_0^C = 3/8$, $\theta_1^C = 1/2$, and $\theta_2^C = 3/4$)[9]. The discrepancies between these exponents are serious enough to be noticed. Our estimate $\theta_0 = 1.1$ is larger than $\theta_0^R = 0.4583$ and $\theta_0^C = 0.375$, which indicates that the meeting possibility for two ends of compacted granular chains is less than two models. Additionally, the "limiting ring closure probability" exponent $\gamma_1 = (d + \theta_1)\nu = 1.65$ for a chain to form a "tadpole" ring is different from $\gamma_1^R = 17/8$ in SAW model[17]. The compacted granular chains thus cannot be exactly described by the SAW model and the compact polymer model. Nevertheless, the qualitative similarity of scaling probability distribution functions are clear enough to imply that simple macroscopic granular chains can capture some statistical characteristics of polymers.

V. BOND-BOND CORRELATION

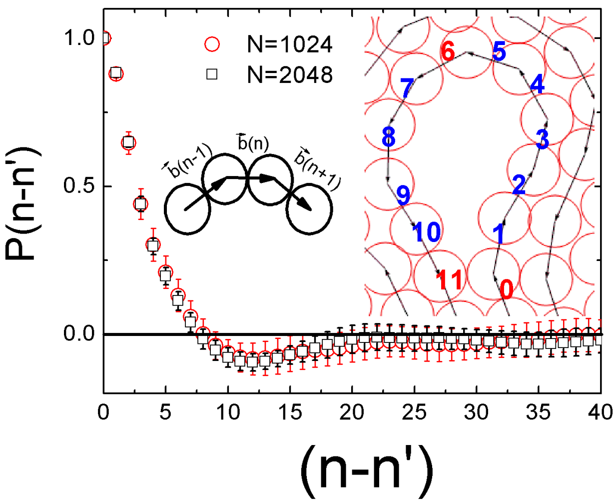

FIG. 5: (color online). The bond-bond correlation measured along a single long chain for $N = 1024$ (open square) and $N = 2048$ (open circle). The correlation function shows a maximum anti-correlation at $n - n' \approx 11$ due to the local bending of the chain. Inset: the spheres are arranged into a near-minimal loop.

We initially extract the coordinates of ball centers along a single chain in order. Using these data, we calculate bond-bond correlation function $P(n - n') =< \vec{b}(n) \cdot \vec{b}(n') >$, which is able to validate long-range orientational order in various system, where n and n' are sphere labels, and $\vec{b}(n)$ is the bond connecting sphere n and $n+1$, as indicated in the inset of Fig.5. $P(n-n')$ is shown in Fig.5 for long chains $N = 1024$ and $N = 2048$ respectively. Due to the local bending of the chain, the correlation function falls rapidly from unity, vanishing at $n - n' \approx 7$, and showing a maximum anti-correlation at $n - n' \approx 11$. This local backfolding of the chain can be directly visualized in the experiment, as illustrated in the inset of Fig.5. As $(n - n') >> 1$, the bond-bond correlation function will eventually return to zero. This result suggests that there is no long-range orientational order for static packing of long linear chains, which is consistent with the observation in the two-dimensional simulation of compact polymers [8].

VI. CONCLUSIONS

In summary, we experimentally investigate various static properties of granular chains in two dimensions, and our results exhibit important statistical information of chain system at macroscopic lengthscales. The long chains pack into a loose structure and do not show a long-range orientational order. The structure factor is calculated and the exponent $\nu = 1/2$ agrees with the result of dense polymer. The probability distribution function of distance satisfies a scaling form that is expected by the polymer theory. It is worth emphasizing that these polymer-like characterizations do not provide a conclusive proof where granular chains can be exactly analogous to molecular chains, that is, polymers in static statistics. These results imply that macroscopic granular chains are able to capture some essential aspects of real polymers. In some important respects such as the packing structure the similarity suggests that if the analog between the two-dimensional granular chains and polymers indeed exists, granular chains would be a helpful tool to understand the underlying physics which is difficult or inaccessible to the experiment of true polymer.

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