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Origin of Non-cubic Scaling Law in Disordered Granular Packing

Chengjie Xia¹, Jindong Li¹, Bingquan Kou¹, Yixin Cao¹, Zhifeng Li¹, Xianghui Xiao², Yanan Fu³, Tiqiao Xiao³, Liang Hong^{1,6}, Jie Zhang^{1,6}, Walter Kob⁴, and Yujie Wang^{1,5*}

¹*Department of Physics and Astronomy, Shanghai Jiao Tong University, 800 Dong Chuan Road, Shanghai 200240, China*

²*Advanced Photon Source, Argonne National Laboratory, 9700 South Cass Avenue, Illinois 60439, USA*

³*Shanghai Institute of Applied Physics, Chinese Academy of Sciences, Shanghai 201800, China*

⁴*Laboratoire Charles Coulomb, UMR 5521, University of Montpellier and CNRS, 34095 Montpellier, France*

⁵*Materials Genome Initiative Center, Shanghai Jiao Tong University, 800 Dong Chuan Road, Shanghai 200240, China*

⁶*Institute of Natural Sciences, Shanghai Jiao Tong University, Shanghai 200240, China*

Recent diffraction experiments on metallic glasses have unveiled an unexpected non-cubic scaling law between density and average interatomic distance, which lead to the speculations on the presence of fractal glass order. Using X-ray tomography we identify here a similar non-cubic scaling law in disordered granular packing of spherical particles. We find that the scaling law is directly related to the contact neighbors within first nearest neighbor shell, and therefore is closely connected to the phenomenon of jamming. The seemingly universal scaling exponent around 2.5 arises due to the isostatic condition with contact number around 6, and we argue that the exponent should not be universal.

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The origin of dynamic arrest and mechanical rigidity in amorphous materials remains one of the important unresolved questions in condensed matter physics [1-3]. Whether it has a structural origin or is just a dynamic phenomenon remains controversial [4,5]. For metallic glasses, it has long been speculated that dense local packing structures of short-range order serve as the building blocks in these systems [6]. However, how these local structures can be extended to medium or large scales remains at present a mystery due to the existence of geometric frustration or intrinsic chemical disorder [7-9]. Recently, it has been proposed that metallic glasses possess a medium range fractal order, which could rationalize the

31 commonly observed non-cubic scaling law between the position of the first diffraction peak and the bulk
32 density found in neutron and X-ray scattering experiments on these systems [10-13]. The first diffraction
33 peak position is usually associated with the largest inter-plane distance in crystals or the typical nearest
34 neighbor distance in liquids [14-16], and it shows in these systems a power law of exponent 3 as a
35 function of the bulk density since they are three-dimensional by nature. It is therefore surprising that the
36 scaling exponent obtained for metallic glasses under density change induced by either pressure or
37 composition tuning has instead values that are between 2.3 and 2.5 [10-13]. The origin of this anomalous
38 scaling law has been attributed to the presence of a regular or statistical fractal network formed by glass
39 order [6,10,12]. In this picture, the atoms move affinely relative to each other under deformation, and their
40 coherent scattering intensity yields the non-cubic law. However, real metallic glasses are in fact quite
41 compact, while a large-scale fractal structure has zero mass density. Therefore, in order for this picture to
42 be valid, one requires that a substantial amount of atoms exist within the fractal interstitials which do not
43 contribute coherently to the sharp scattering peaks [10]. Another possibility is that the fractal structure
44 only exists up to a finite length scale, above which the system is still homogeneous and three-dimensional
45 [6,12]. These explanations are appealing since they naturally refer to a fractal medium-range glass order,
46 such as percolating icosahedral structures, for metallic glasses, and therefore explain how glass order
47 extends in space. However, the interpretation of the existence of the non-cubic law based on the fractal
48 picture is not without controversy [17], and sometimes one also find deviations from the non-cubic law
49 [18,19].

50 In this work, we provide microscopic insight to this problem by studying the three-dimensional
51 packing of spherical granular particles, which is a prototypical hard-sphere glass former and has long been
52 considered as a structural model for metallic glasses [20-22]. We identify a non-cubic scaling law in our
53 system, and provide evidences that its origin is local, i.e., without resorting to any fractal structures.
54 Instead, it results from a complex structural evolution of the first-shell neighbors when the packing
55 fraction varies, controlled mainly by the contact neighbors as required by mechanical stability, and the
56 global behavior is a simple statistical average of the local ones. Therefore, such phenomenon is directly

57 related to jamming phenomenon and might be universal near the jamming criticality [23,24]. In the
 58 experiment, we used synchrotron X-ray CT techniques to obtain the packing structures of packing with a
 59 wide range of packing fractions Φ [25-34] (see Supplemental Material [35]). In the following, we use the
 60 average particle diameter as a unit of length.

61 The investigation of the non-cubic law can be carried out in both reciprocal and real space. First, we
 62 followed the previous scattering experiments on metallic glasses, and studied the structural factor of the
 63 packing to investigate the evolution of the peak positions versus Φ . The structure factor is calculated

64 according to $S(q) = \frac{1}{N} \left| \sum_j e^{-iq \cdot r_j} \right|^2$, where N is the number of particles in the probed volume, and shown

65 in Fig. 1(a). The position q_i of i th peak is obtained by fitting the peak to a Gaussian function. Previous

66 studies on metallic glasses report scaling behaviors of $\Phi \propto q_i^{D_q(i)}$, with a scaling exponent D_q varies

67 between 3 and 2.5 for the first and second peaks [12]. In our system we find however that, q_2 does not

68 change in the whole investigated Φ range, which corresponding to a very large $D_q(2)$, and the analysis

69 on q_1 yields a $D_q(1)$ which is clearly larger than 3 [Fig. 1(b, c)]. Since the interpretation of $S(q)$ is not

70 completely trivial, we have also calculated the pair correlation function $g(r)$ [Fig. 1(d)]. Similar to

71 $S(q)$, we obtain the peak position p_i of the i th peak of $g(r)$ by a Gaussian function fit, and

72 determine the scaling behavior $\Phi \propto p_i^{-D_p(i)}$. Note that $p_1 = 1$ for all values of Φ since the distances

73 between contact neighbors are always 1.0 which yields $D_p(1) = \infty$. $D_p(i)$ decreases from about 5.2 for

74 the second peak to 3.1 for the fourth peak, indicating a cross-over from an anomalous scaling (weak Φ -

75 dependence) at short distances to a normal Φ -dependence on larger length scales [Fig. 1(e-g)]. At first

76 sight the rather different behaviors in both reciprocal and real spaces with respect to the findings in

77 metallic glass systems look surprising. However, we notice that extracting the dimension of a fractal

78 structure from the position of the peak of either $S(q)$ or $g(r)$ is difficult because of the ambiguous and
 79 incomplete information they carry [36,37] (see Supplemental Material [35]).

80 To avoid such ambiguity and understand the essence of non-cubic law on the level of the particles,
 81 we develop a more suitable method to define the length scale associated with a fixed number of particle,
 82 and then to determine its scaling behavior with Φ . For this, we first sort for each particle its distances to
 83 all of its neighbors in ascending order, with the n th nearest distance being r_n , and then calculate the
 84 average neighbor distance of the nearest n neighbors as $R_n = \frac{1}{n} \sum_{i=1}^n r_i$. We find that for all n the average
 85 distance $\langle R_n \rangle$ follows a scaling relationship, i.e., $\Phi \propto \langle R_n \rangle^{-D_R(n)}$ [see Fig. 2(a) for $n = 13$], with an
 86 exponent $D_R(n)$ that shows a complex dependence on n i.e., on the length scale considered. In Fig.
 87 2(b) we plot $D_R(n)$ as a function of $\langle r_n \rangle$, where $\langle r_n \rangle$ is the average distance of the n th nearest
 88 neighbor, which grows for large n like $\langle r_n \rangle \propto n^{1/3}$. Surprisingly we find that $D_R(\langle r_n \rangle)$ shows an
 89 oscillatory behavior that is very similar to the one of $g(r)$, and reaches its minimum value
 90 $D_R(13) \approx 2.5$ at $\langle r_{13} \rangle \approx 1.37$, which is close to the location of the first valley in $g(r)$ normally
 91 considered to be first-shell boundary [Fig. 2(b)]. Thus we see that the scaling exponent of 2.5 found in a
 92 series of metallic glasses is reproduced here in our granular system as the minimal value of $D_R(n)$. The
 93 figure also shows that for large n , $D_R(n)$ converges towards the expected value of 3.0.

94 The similarity of $D_R(\langle r_n \rangle)$ and $g(r)$ suggests that there exists a close connection between the shell
 95 structure of granular packing and the unusual scaling behavior. To elucidate this better, we define
 96 $\langle R_{shell,N} \rangle$ as the average distance between the central particle and the particles in the N th shell, which
 97 are between the $(N-1)$ th and the N th valleys of $g(r)$, and determine how this distance depends on

98 Φ : $\Phi \propto \langle R_{shell,N} \rangle^{-D_{shell}(N)}$. Thus $R_{shell,N}$ is a coarse-grained quantity of r_n . We find that $D_{shell}(1) \approx 2.5$
99 and $D_{shell}(N)$ evolves towards 3.0 for large N [left inset of Fig. 2(b)], a result that agrees with
100 previous simulation works on metallic glasses in which the non-cubic scaling law are observed only up to
101 a finite length scale [12].

102 The oscillatory behavior of $D_R(n)$ shows that neighbors at different distances undergo non-uniform
103 displacements with respect to the central particle when Φ changes. We thus can single out their
104 contributions to $D_R(n)$ by investigating the behavior of the n th nearest neighbors individually, i.e., the
105 scaling relationship of $\Phi \propto \langle r_n \rangle^{-D_r(n)}$ [right inset of Fig 2(b)]. If all particles change their distances to the
106 central particle by the same rate when Φ varies, $D_r(n)$ should equal to 3 regardless of n , while
107 $D_r(n) < (>) 3$ corresponds to an average radial displacement larger (smaller) than a homogeneous one.
108 Within the first shell, we find that $D_r(n) > 3$ for $n \in [1,6]$ and $D_r(n) < 3$ for $n \in [7,13]$, and their
109 overall behavior gives rise to the 2.5 scaling law. This behavior gives us a first hint of how the non-cubic
110 law emerges, which results from the complex non-uniform structural evolution mainly within the first
111 shell as Φ changes.

112 To obtain a more specific understanding of the structural origin of the non-cubic law, we determine
113 the Φ -dependence of the local structure within the first shell. For this, we divide the neighbors in the first
114 shell of each particle into two groups. The nearest six ones, i.e., those with $D_r(n) > 3$, and the rest. This
115 classification basically corresponds to the division of quasi-contact and non-contact neighbors owing to
116 the isostatic requirement for mechanical stable granular packings. For each group, we calculate the radial
117 distribution function [Fig. 3(a, b)]. The probability distribution function (PDF) of neighbor-to-center
118 distance r for particles with $n \in [1,6]$ are basically independent of Φ , while the ones for particles with
119 $n \in [7,13]$ show a considerable shift of weight from large to small r as Φ increases. This observation

120 thus explains why for $n \in [1, 6]$ the exponent $D_r(n)$ is large, i.e., no Φ -dependence of $\langle r_n \rangle$, whereas
 121 for $n \in [7, 13]$ it is small, i.e., strong Φ -dependence of $\langle r_n \rangle$. We also calculate a three point correlation
 122 function that gives structural information not accessible from scattering experiments. For this, we measure
 123 the angle θ spanned by the central particle and any two of its neighbors. The distribution of θ for
 124 $n \in [1, 6]$ shows a peak at 60° that becomes sharper with increasing Φ [Fig. 3(c)], which suggests that
 125 these particles tend to aggregate to form regular triangles which can further lead to the formation of quasi-
 126 regular tetrahedral structures [33,34]. In contrast to this, the distribution of θ for $n \in [7, 13]$ does not
 127 show a significant change apart from a slight change in the peak positions [Fig. 3(d)]. The described
 128 complex non-affine structural evolution is consistent with the previous observation that the average shape
 129 of the Voronoi cells changes from being anisotropic to more isotropic as Φ increases [33,38,39]. It is this
 130 non-affine deformation which induces the deviation from a cubic law between the local packing fraction
 131 ϕ of the Voronoi cell defined by the first-shell neighbors and their average neighbor-to-center distance
 132 R_{13} . (We define ϕ as the ratio between the volumes of each particle and its Voronoi cell.) Together with
 133 the fact that the average ϕ is very close to the global Φ , the non-cubic law between Φ and $\langle R_{13} \rangle$
 134 naturally emerges. Thus above structural analysis supports the local explanation of the non-cubic law
 135 irrespective of structural information at medium or long-ranges.

136 To further justify this local explanation, we make a scatter plot of ϕ v.s. R_{13} , and fit the scatter plot
 137 using $\phi \propto R_{13}^{-d}$ to capture the average behavior (Fig. 4). The scaling exponent can essentially be
 138 evaluated by $d = \sigma[\log(\phi)] / \sigma[\log(R_{13})]$, where $\sigma(\cdot)$ represents the standard deviation.
 139 Interestingly, d shows an increasing trend from about 2.6 to 2.9 with decreasing Φ (inset of Fig. 4),
 140 which indicates that a local version of the same non-cubic law holds, suggesting that a low- Φ packing
 141 with more liquid-like structure, i.e., smaller contact numbers, has an exponent d closer to 3. This subtle

142 trend is hidden if one fits the global quantities Φ versus $\langle R_{13} \rangle$ to obtain a single $D_R(13)$. Furthermore,
 143 as shown in Fig. 4, the relationship between Φ and $\langle R_{13} \rangle$ is consistent with the overall local trends,
 144 suggesting that the global scaling law is simply an average manifestation of the local non-cubic law
 145 between ϕ and R_{13} with gradually varying d values.

146 In the following, we demonstrate that the exponent is closely related to the existence of contact
 147 neighbors as required by mechanical stability in granular packing [24,40,41], and is a phenomenon
 148 connected to jamming, instead of the fractal glass order as we set out to relate in the first place [34]. This
 149 finding is not totally surprising as we recall that even in the work which tried to relate the non-cubic
 150 exponent to a presumed fractal glass order in metallic glasses, the anomalous scaling is observed only far
 151 below the glass transition temperature, and the potential relationship to jamming is alluded [12].

152 To illustrate this point, we investigate the dependency of the non-cubic exponents on contact number.
 153 Two particles are considered to be in quasi-contact with each other if their surfaces are closer than a cut-
 154 off distance around 0.01 of the particle diameter [27,31]. We use *quasi-contact* to identify very close
 155 neighbors, which are not necessarily in actual geometric or mechanical contact. In Fig. 5(a), we group the
 156 particles based on their local quasi-contact number z . The conditional probability distribution of both ϕ
 157 and R_{13} shift for increasing z values. In each group of particles with fixed z , the correlation between ϕ
 158 and R_{13} can be described by $\phi \propto R_{13}^{-d_z}$, and d_z is again evaluated as the ratio between the standard
 159 variances of $\log(\phi)$ and $\log(R_{13})$ for particles with given z . As expected, d_z depends on z , and
 160 increases towards 3 for decreasing z . Furthermore, it's intriguing to notice that the relationships between
 161 d_z and z are almost identical for all packing with different Φ [Fig. 5(b)], further confirming that it is a
 162 local property. The universal behavior of d_z can therefore describe the Φ -dependence of d , even if d_z
 163 is a bit smaller than d . As we show in the Supplemental Material, this difference originates from the
 164 complex inter-dependency between ϕ , R_{13} and z .

165 In conclusion, we give a local explanation for the origin of a non-cubic law in granular hard-sphere
166 systems, and find it to be related to the phenomenon of jamming instead of a fractal glass structure.
167 Although we do observe in our system the non-cubic scaling laws, the exponents we extract for the peak
168 positions in $\mathcal{S}(q)$ and $g(r)$ do not match the ones found in metallic glasses. Thus, our work makes it
169 clear that the non-cubic law might not be universal for both granular and metallic glass systems. For
170 granular systems, the non-universal behavior is presumably due to the presence of friction, which moves
171 system away from the isostatic jamming point. For metallic glasses, since there must be other important
172 parameters (stiffness of potential, covalent bonding, *etc.*), which go beyond the hard-sphere picture and
173 thus will influence this exponent [22]. Also, the rather high temperature at which the scaling law is
174 normally probed in metallic glasses could also influence the exponent. Nevertheless, we believe that a
175 very similar physical mechanism is at work for both systems, since to the first approximation metallic
176 glasses can be described as hard-sphere systems. It is possible in the limit of the isostatic jamming point
177 (with contact number of 6), a universal scaling law of 2.5 indeed exists. This brings us the attention to
178 recent advances in the theory of hard-sphere glasses of a new type of glass transition, the Gardner
179 transition [24,42,43]. This transition happens by breaking the glass metabasins into subbasins by forming
180 a marginal glass. The length scale of this transition is close to that investigated in the current work. It is
181 therefore possible that the scaling exponent identified here is a new structural property of the marginal
182 glass phase or jamming transition, in addition to the cage order parameter or vibration motions normally
183 studied [44]. It is therefore interesting to probe this connection in the future.

184

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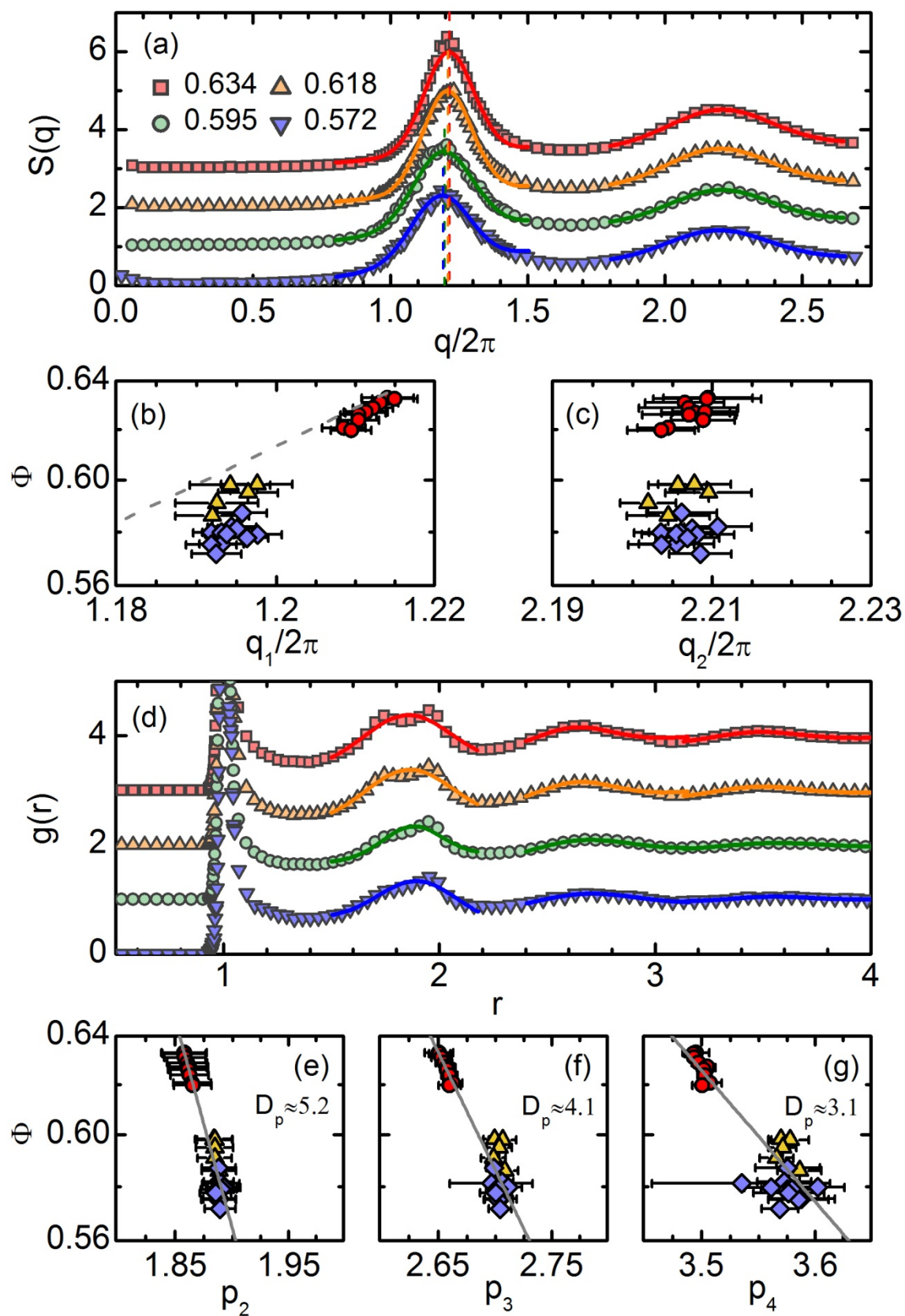
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193 * Corresponding author.

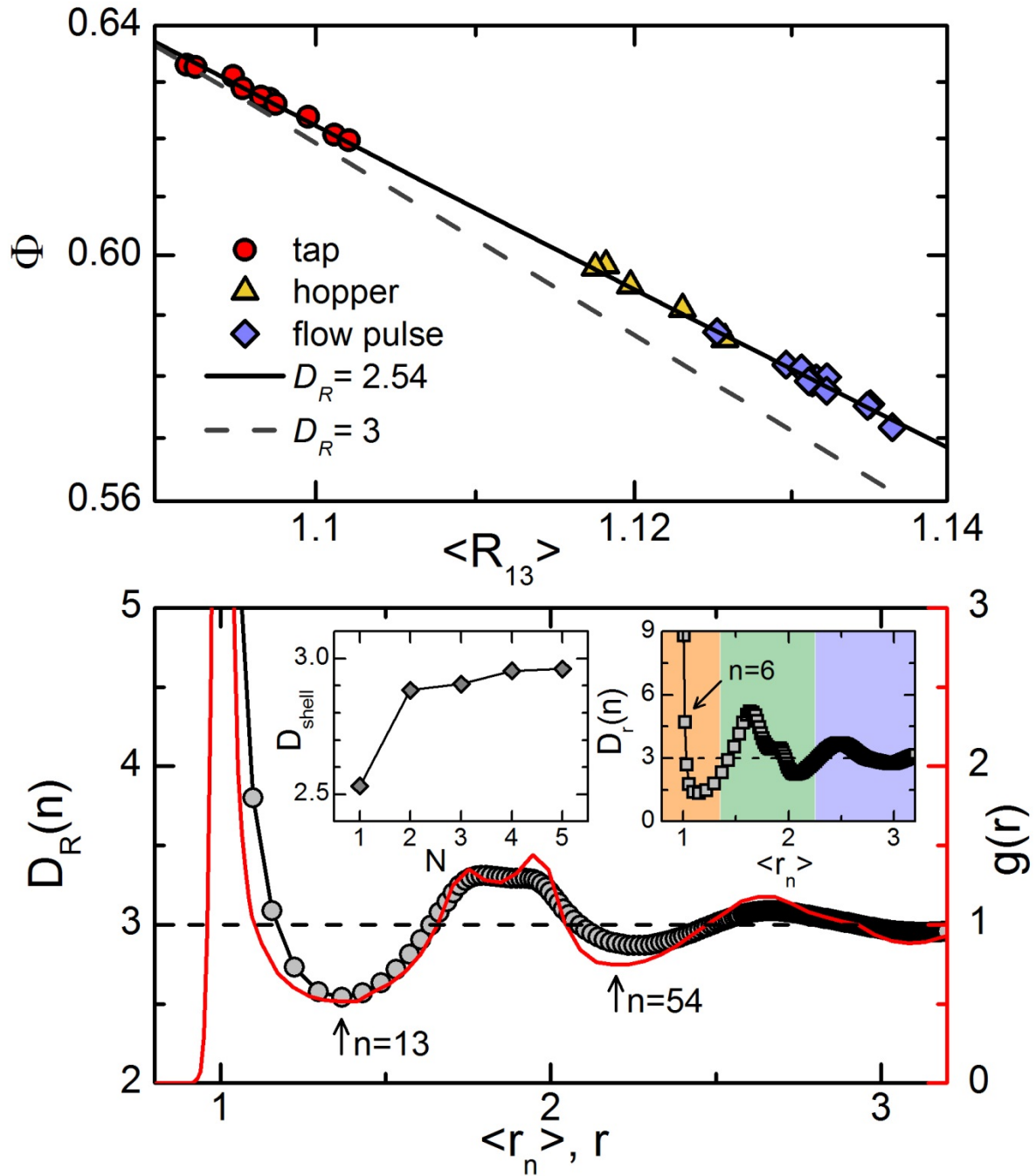
194 yujiewang@sjtu.edu.cn

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252 FIG. 1. (a) Structure factors and (d) pair correlation functions for four different packing fractions Φ (see
253 legend). The data are shifted vertically for clarity. Solid lines are Gaussian fits to the peaks. Dashed lines
254 mark the peak positions. (b, c) Peak positions of the first and second peak of $S(q)$ for all packing. The
255 size of the error-bar of Φ is smaller than the symbol size. The dashed line in (b) represents $\Phi \propto q_1^3$. (e-g)
256 Peak positions of the second to fourth peak of $g(r)$. The solid lines is the fit of the form $\Phi \propto p_i^{-D_p(i)}$,
257 with $D_p(2) = 5.2 \pm 0.5$, $D_p(3) = 4.1 \pm 0.5$ and $D_p(4) = 3.1 \pm 0.3$. Different symbols in (b, c, e-g)
258 represents three different packing preparation protocols as explained in Fig. 2(a).
259



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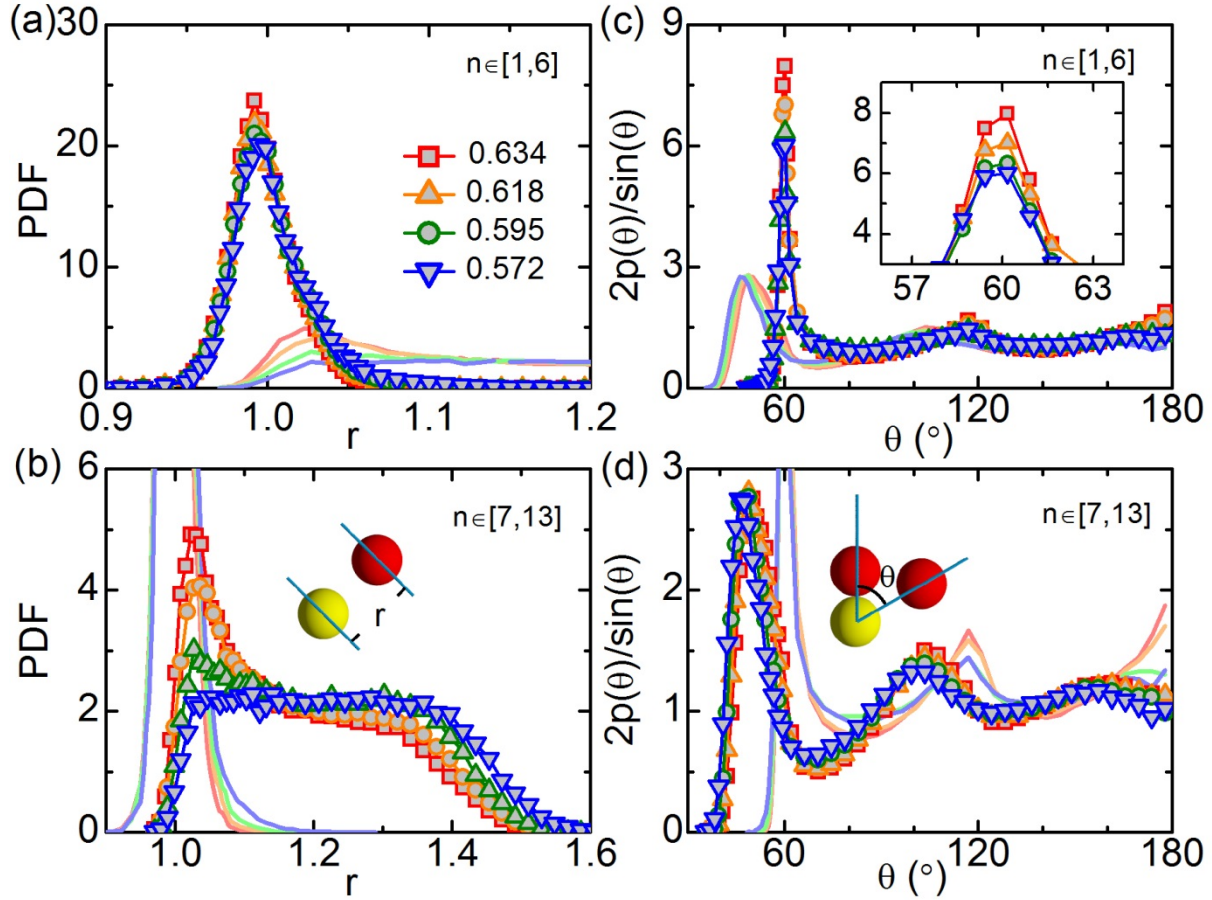
261 FIG. 2. (a) Φ versus $\langle R_{13} \rangle$. Different symbols represent different packing preparation protocols:

262 Tapping (circles), hopper (triangles) and flow pulse (diamonds). A clear non-cubic law can be identified

263 through the fit $\Phi \propto \langle R_{13} \rangle^{-D_R(13)}$ with $D_R(13) = 2.54 \pm 0.03$ (solid line) with a cubic law (dashed line)

264 for comparison. (b) $D_R(n)$ versus $\langle r_n \rangle$ (symbols, left axis) and $g(r)$ (line, right axis) for packing with

265 $\Phi=0.634$. The dashed line represents $D_R=3$. $n=13$ and $n=54$ are marked as the boundaries of
266 the first two shells. Left inset: $D_{shell}(N)$ versus shell number N . Right inset: $D_r(n)$ versus $\langle r_n \rangle$. The
267 dashed line represents $D_r=3$, and the background colors separate the different shells.
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270

271 FIG.3. PDFs of neighbor-to-center distances r (a, b) and angle θ (c, d) for particles with $n \in [1, 6]$ (a,

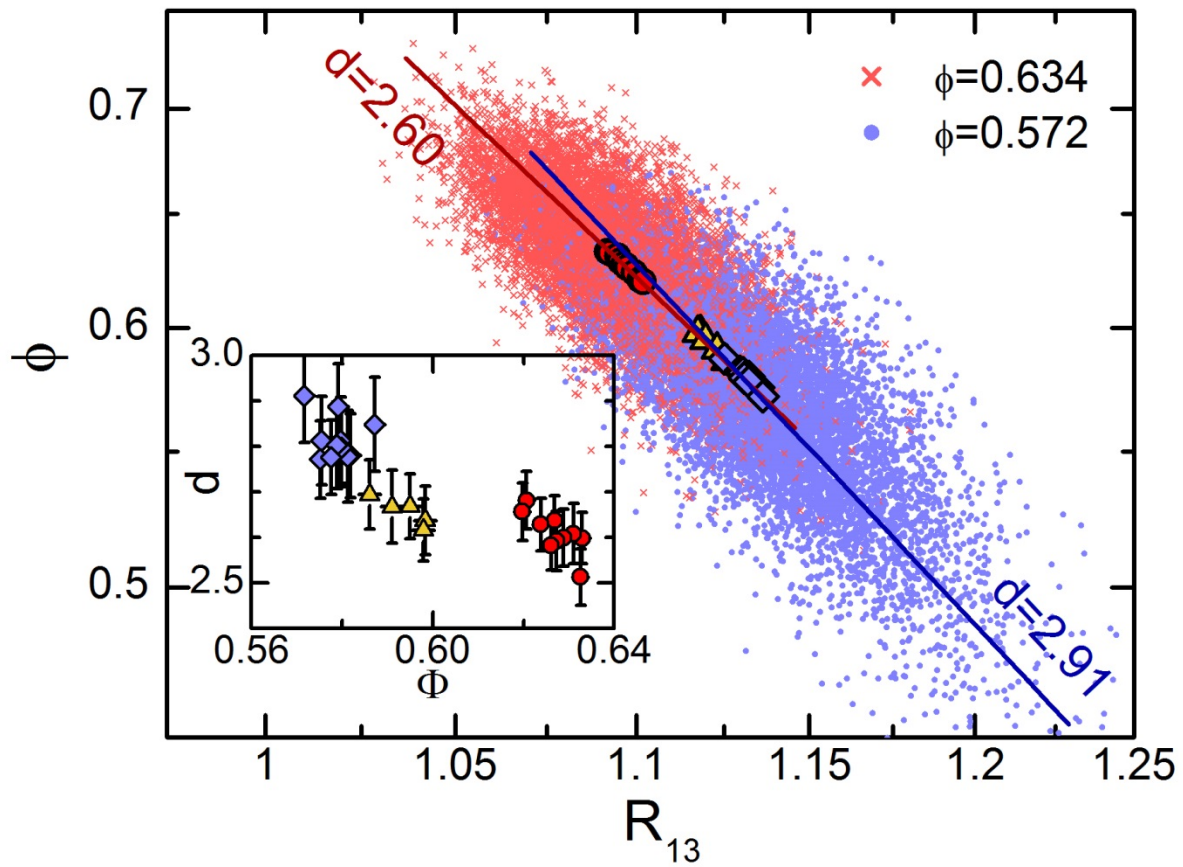
272 c) and $n \in [7, 13]$ (b, d) for four different packing fractions given in the legend. The inset in (c) is a

273 zoom onto the peak at 60° . In (c, d), $\sin(\theta)/2$ is a normalization factor. For the sake of comparison,

274 we plot in each panel the PDF for the other group of particles (solid lines). Panels (b) and (d) also show a

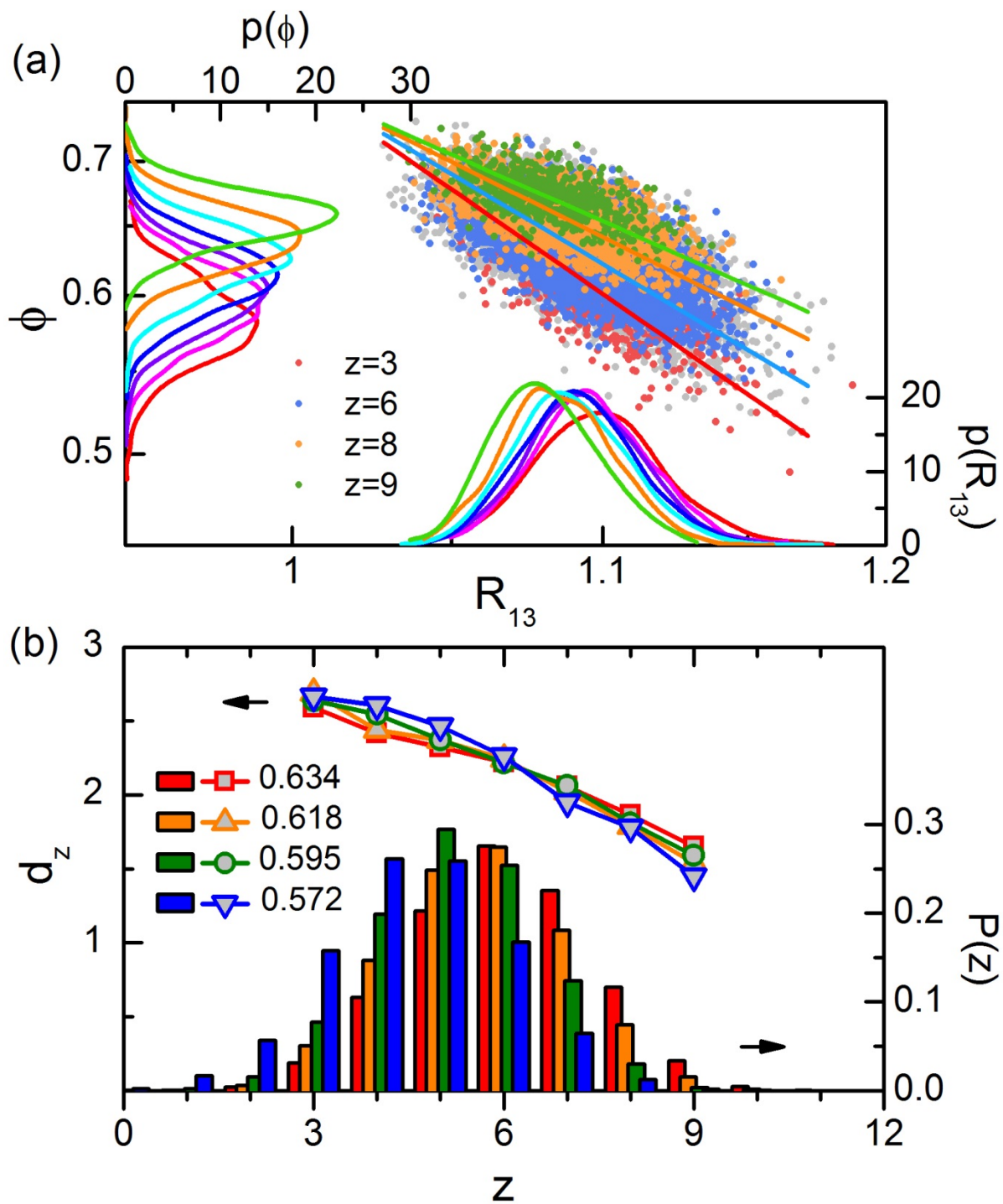
275 schematic picture of the definitions of r and θ .

276



277
 278 FIG.4. Scatter plot of ϕ and R_{13} for our densest (cross) and loosest packing (dot). The solid lines
 279 represent $\phi \propto R_{13}^{-d}$, with $d = 2.60 \pm 0.02$ for $\Phi = 0.634$ and $d = 2.91 \pm 0.03$ for $\Phi = 0.572$. The
 280 global average values of Φ and $\langle R_{13} \rangle$ are also shown. Inset: d as a function of Φ .

281



282
 283 FIG.5. (a) Scatter plot of ϕ and R_{13} for particles of packing with $\Phi = 0.634$ with different z . For
 284 clarity, only particles with $z = 3, 6, 8, 9$ are colored and the rest ones are plotted in gray dots. The solid

285 lines represent $\phi \propto R_{13}^{-d_z}$. Conditional PDFs of ϕ (top axis) and R_{13} (right axis) for different z are also
286 shown. (b) d_z as a function of z for four different packing (left axis), and the probability distribution of
287 z for the same four packing (right axis).