Transition Dynamics of Frictional Granular Clusters

Antoinette Tordesillas∗ and David M. Walker

Department of Mathematics and Statistics, University of Melbourne, Parkville, VIC 3010 Australia

Gary Froyland

School of Mathematics and Statistics, University of New South Wales, Sydney NSW 2052 Australia

Jie Zhang

Department of Physics, Shanghai Jiao Tong University, Shanghai 200240, P. R. China

Robert P. Behringer

Department of Physics, Duke University, Durham, NC 27708 USA

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Force chains, the primary load-bearing structures in dense granular materials, rearrange in response to applied stresses and strains. These self-organized grain columns rely on contacts from weakly stressed grains for lateral support to maintain and find new stable states. However, the dynamics associated with the regulation of the topology of contacts and strong-versus-weak forces through such contacts remains unclear. This study of local self-organization of frictional particles in a deforming dense granular material exploits a transition matrix to quantify preferred conformations and the most likely conformational transitions. It reveals favored cluster conformations reside in distinct stability states, reminiscent of “magic numbers” for molecular clusters. To support axial loads, force chains typically reside in more stable states of the stability landscape, preferring stabilizing truss-like, 3-cycle contact triangle topologies with neighboring grains. The most likely conformational transitions during force chain failure by buckling correspond to rearrangements among, or loss of, contacts which break the 3-cycles.

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I. INTRODUCTION

Modern materials science abounds with explorations into a diverse array of dissipative media (e.g. concentrated emulsions, granular materials, cellular materials, slurries, pastes, molecular glasses) [1–8]. In part, this broad appeal arises from their distinctive mechanical response to applied forces. On a macroscopic scale, such response often bears the hallmarks of complexity, including self-organization and emergent behavior (e.g. [2–5, 9–12]). By far one of the best examples of these patterns of behavior, common in both nature and technology, are those exhibited by dense granular materials under quasi-static loading conditions [10–19]. Here we study the underlying mesoscopic mechanisms for such behavior, paying particular attention to recurring ordered arrangements (i.e. structural motifs) and preferential rearrangements which are larger in length scale than those of the constituent grains. Unravelling the details of these mesoscopic structures and associated frictional rearrangements, and their impact on bulk mechanical response – particularly on the development of failure – presents a great scientific challenge [7, 18, 20, 21]. Indeed, although granular materials have stimulated intense research activity for decades [5–8], robust predictions of their mechanical behavior under load remain elusive [12, 22–26]. To date, a crucial missing element that limits prediction and control of dense granular behavior, particularly that of inelastic deformation and failure, is a precise understanding of the interplay between the dynamics of prevalent and persistent particle rearrangements – and the evolution to failure of key functional mesoscopic structures, epitomized by major load-bearing force chains [10, 14, 18, 26–32]. Knowledge of the dynamical rules governing these essential inelastic rearrangements, especially as they relate to the stability of and energy released from force chains, is crucial for reliable model predictions [11, 21]. Advances in this respect not only hold great promise in the control of granular behaviour in many practical settings (e.g. forecasting and mitigation of earthquakes and landslides) [17, 31, 32]; improvement of road and off-road transport infrastructure [33]; development of mining processes and technologies which minimize environmental impact and energy use [7, 8, 34] – but also the creation of novel materials with a level of functionality similar to, if not potentially superior than, those observed in nature (e.g. bio-inspired ceramics) [1].

Recent numerical and experimental developments have yielded insights into how the local structural behavior of columnar force chains leads to inelastic deformation and failure at the macroscopic scale [9–18, 21, 30, 31, 35–41]. As the material deforms in response to applied stresses, the constraints imposed by the local structure
influence the resultant kinematics of individual grains, both in these strong columns and their relatively weak confining neighbors. Experiments and discrete element (DEM) simulations have shown that conformational re-arrangements occur frequently, transitioning the system towards the various stable structural configurations accessible to the system (e.g. [11, 42, 43]). These dissipative rearrangements, resisted by interparticle contact friction and geometric particle interlocking, are strongly nonlinear and heterogeneous in space and time (e.g. [11–13, 15, 21, 44]). The most pronounced manifestation of these rearrangements occur during failure in the presence of shear bands (e.g. [13, 15, 17, 31, 41, 44]). A shear band is a region in the material in which deformation and particle rotations are concentrated. Once fully developed, shear bands split the granular body into parts which may move relative to each other in near-rigid body motion [9, 15, 44, 45]. Grain-scale information from non-invasive micromechanical experiments on sand and photoelastic disk assemblies suggests that force chains fail by buckling, and that their collective localized buckling is the mechanism underpinning shear band formation and, ultimately, macroscopic localized failure [13, 37, 44]. A comprehensive experimental and numerical investigation into force chain buckling in a series of monotonic and cyclic shear tests, for a range of granular materials corroborate this [11, 21, 30, 36–41]. Moreover, cooperative evolution among linear and cyclic building blocks of self-organization, i.e., force chains and surrounding minimal contact cycles, has been observed [11, 16, 39, 40]. How force chains regulate their contacts with respect to both topology and the forces they carry for a given loading condition is largely unknown. An overarching question is whether particles rearrange in response to the changing applied forces to provide optimum stability to these axially loaded columns of grains. Transition pathways between granular cluster conformations in a stability landscape would provide such information, but to date, these have not been established for any granular system. Here we report on a study of these transition pathways from the standpoint of cluster topology and structural stability. By probing relative probabilities of cluster conformations and conformational transitions we find that there are favored conformations residing in distinct structural stability states. Analysis of stability transitions and associated dynamical barriers elucidate the role of topology in the stability and failure of force chains by buckling.

The rest of this paper is arranged as follows. In Section II, we provide details of the experimental system, before discussing how the structural stability of cluster conformations of frictional particles are calculated from knowledge of contacts and contact forces. Next, we present an analysis in Section III of the dynamics of conformational transitions, modeled by a Markov transition matrix, before focussing on those specific to force chains in Section IV. In Section V, the structural stability landscape of cluster conformations is examined, with the transitions in conformational stabilities similarly quantified using a Markov transition matrix. The stability of force chains and the stability transitions specific to the failure of force chains by buckling is presented. We close the paper in Section VI with a discussion of the implications of the work for constitutive modelling of dense granular media, and highlight some common ground and potential synergies with studies of molecular complex systems.

II. MATERIAL AND METHODS

Experimental apparatus and procedure

We examine a bidisperse, densely-packed assembly of frictional circular disks subject to quasi-static cyclic shear under constant volume: see Fig. 1 top left for an image of the assembly with the stresses visualized from birefringence patterns in the photoelastic disks. The experiment involves 1568 bidisperse, photoelastic disks (particles), confined in a biaxial apparatus: ∼ 80% have diameter 0.74 cm, ∼ 20% have diameter 0.86 cm. The particles rest on a flat, smooth horizontal surface, and the walls of the biax move to create specific strains, here pure shear: compression in one direction, dilation in the other, with fixed system area/density. Photoelastic materials used here are birefringent under strain. The image of a photoelastic particle, placed between crossed polarizers, shows light and dark bands that are determined by the stresses within the disk (Fig. 1 top left). These stresses are in turn, fixed by the (vector) contact forces acting on the particle. We solve the inverse problem for each photoelastic disk image to obtain these contact forces [41, 46]. Separate images measure the position, displacement, rotation and contact topology of each particle at each state of the loading or deformation history. Using this information, we can calculate the structural stability for each particle at each strain state [11].

The initial state is specified by: square system boundaries, a (fixed) packing fraction of \( \phi = 0.795 \), and as nearly isotropic and stress-free conditions as possible. We then shear the system cyclically, first by compressing/expanding in the vertical/horizontal directions, keeping the area fixed. After reaching a maximum shear deformation, we reverse the shear strain, so that there is compression/expansion along the horizontal/vertical axes. We carry out reverse shear past the configuration of square boundaries, until the shear strain reaches a minimum negative value. We again reverse the shear strain, and we return the boundaries to a square. This completes one shear cycle. Starting from the final state of the first shear cycle, we then apply similar cyclic strains for a total of six shear cycles. Each step in this process consists of a small quasi-static increment of strain (0.3%), and after each small step, we freeze the boundaries and
FIG. 1: (Color online) Granular cluster conformations. Image of the assembly of photoelastic birefringent circular disks showing the force chains when sample is compressed vertically (top left). Subgraphs of the 28 conformations found for the experimental system throughout loading history (top right). Densities of conformations shown as solid line, and the most prevalent clusters classified into four almost-invariant sets distinguished by colour and symbol (bottom left). Representative members of three of these sets are drawn as clusters showing typical rearrangements are due to a single contact forming or breaking for the same number of disks or a new disk joins the cluster. (bottom right).

Acquire three images: one with polarizers, one without polarizers, and a third that allows us to characterize disk rotation. In the present discussion, we only examine the data for the first two shear cycles.

A shear band develops at certain stages of each shear cycle. The evolving mechanical response of the granular material is quantitatively characterized by analysis of the individual equilibrium states, each corresponding to a strain state in the loading history, and their sequential state-to-state transitions. For each equilibrium state, an unweighted and undirected graph models the particles as the nodes, and their contacts as the connecting links or edges between corresponding nodes [35, 39, 40]. Particle rearrangements are primarily responsible for the material’s deformation. These are reflected in the evolving complex network of contacts: old network connections break as contacts are lost, while new connections form as contacts are created.

Structural stabilities of cluster conformations

Frictional particles in a cluster interact at their contacts through: (i) an elastic normal force given by Hooke’s law (spring), (ii) an elastic-plastic tangential force given by Hooke’s law if elastic and Coulomb’s friction law if plastic (spring-slider combination), and (iii) an elastic-plastic contact moment analogous in form to the tangential force [11]. At each equilibrium state, we compute the stability of each cluster configuration, \( \lambda \), by considering these forces using structural mechanics techniques [30, 42, 47]. A contact through which elastic forces...
and moment act is termed “elastic”; otherwise it is “plastic”. It was proven [11] that two conservative systems $S_+$ and $S_-$, having only elastic contacts, can be constructed for any nonconservative (frictional) granular cluster so that their stabilities provide upper and lower bounds on the cluster stability. $S_+$ is constructed by replacing every plastic contact in the cluster with a resistive spring of stiffness equal to zero, hence removing any resistance at that contact: $\lambda^-$, the stability of $S_-$, bounds $\lambda$ from below. $S_-$ is constructed by replacing every plastic contact in the cluster with a spring of stiffness equal to $\gamma \lambda^+$, where $\gamma$ is the number of plastic contacts to total number of contacts in the cluster $[11]$. $\lambda$ has a dimensional unit of force per unit length, here $N/m$. Although the matrices $M_+$ and $M_-$ apply to the conservative systems, they each contain information on the frictional granular cluster: i.e., the number of contacts and their individual type, position and orientation, the material properties (i.e., spring stiffness and coefficient of friction), and the boundary/loading conditions. These are all directly measurable quantities in the experiment $[11, 41, 46]$.

### Markov transition matrix analysis

A Markov transition matrix $P$ is constructed by counting the observed transitions that each cluster conformation undergoes over time steps throughout experimental loading. The states of the transition matrix are either (i) the granular cluster conformation or (ii) the interval of stability in which a granular cluster’s structural stability sits. The matrix $P$ is normalized to be a row stochastic matrix, and the entries of the leading left eigenvector give an approximation to the invariant density of the assumed underlying stochastic process modeling the dynamics. The approximate invariant density and $P$ can be used to construct a time-reversible Markov matrix $R$ whose eigenspectrum, starting with the second largest eigenvalue, can be used to determine almost-invariant transition sets $[48]$. Gaps in the spectrum of real eigenvalues of $P$ indicate how many eigenvectors of $R$ to use in order to find a given number of almost-invariant transition sets. The eigenvalue separation can also be used to gauge whether the time step interval length is appropriate. A clustering algorithm is employed to determine the (transition state) membership of each almost-invariant set by minimizing a weighted cost function, where the weighting is closely related to the approximate invariant density of each state $[48]$.

### III. MESOSCALE CONFORMATIONS AND CONFORMATIONAL TRANSITIONS OF GRANULAR SELF-ORGANIZATION

In this study, we focus on that class of mesoscopic cluster conformations comprising a particle and its first ring of contacting neighbors (analogous to the first coordination shell in molecular clusters). This class comprises the smallest clusters for which emergent preferential orientations in contacts and contact forces can be comprehensively probed. Next we sort the clusters according to the network properties of degree (number of contacts of the central reference particle) and number of 3-cycles (triangles) within the cluster $[35, 39, 49]$. The physical and geometrical restrictions imposed by the particle size distribution and the system boundaries imply a finite number of distinct cluster conformations involving one node (particle) and its connected neighbors. We found 28 distinct cluster conformations after examining the first ring of contacting neighbors of each particle at every state throughout loading. Fig. 1 top right shows the subgraphs of the conformations, each labelled $x_j^k$: $x$ is the central particle degree, $i$ is the number of 3-cycles, $j$ is the number of shared edges among these 3-cycles.

The transitions between the 28 cluster conformations are modeled as a discrete time Markov process $[48]$ with one time step representing an interval of four quasi-static equilibrium states, i.e., the number of times a particle of a given conformation transforms to another conformation, possibly of the same topology, after four time steps (recall Materials and Methods for an explanation on how to select a suitable time step interval). The corresponding $28 \times 28$ transition matrix is analyzed to extract the most prevalent conformations and to identify sets of conformations which are almost-invariant under the dynamics, i.e., the most persistent sets of conformations $[48]$. In order of decreasing frequency, the most recurrent granular conformations are $3_0^0$, $2_0^0$, $4_0^0$, $4_1^0$ and $3_0^0$ (Fig. 1 bottom left). Almost-invariant transition sets of granular conformations are identified from the eigenvectors of the transition matrices. The number of sets are determined by the separation of the spectrum of eigenvalues using a clustering algorithm $[48]$; Fig. 1 bottom left shows four almost-invariant sets found using three eigenvectors. Conformations of similar degree typically lie in the same set, with the distinction between members of each set being mainly due to the rearrangements of the first ring neighbors (i.e., no loss or gain of contacts): see Fig. 1 bottom right. The top five conditional transition probabilities $p(x_j^k \rightarrow y_j^k)$, quantifying conformational transitions from $x_j^k$ to $y_j^k$ are: $p(2_0^0 \rightarrow 2_0^0) = 0.1771$, $p(3_0^0 \rightarrow 3_0^0) = 0.1696$, $p(2_0^0 \rightarrow 3_0^0) =$
IV. TOPOLOGY AND DYNAMICS OF FORCE CHAINS

The strength of a granular assembly under load, i.e., its resistance to particle rearrangements or deformation, comes from two sources. One is inherent (e.g., initial packing, particle shape and surface properties); the other is induced by the loading conditions, i.e., by self-organization. Recent evidence of cooperative behavior among emergent mesoscopic structures, such as the formation of truss-like 3-cycles around force chains, provides incisive clues for functional activity with governing design principles resembling those employed in architectural structures [11, 16, 39, 40].

Force chains are prone to fail by buckling [14, 27, 30, 37, 44]. To function effectively, these primary axial load-bearing columns of grains must be supported laterally through contacts with comparatively weak neighboring particles [30]. Past studies have revealed strong similarities in force chain behavior for a wide range of material properties and boundary conditions [11, 30, 38, 40, 41]. Here we find that, at any given equilibrium state, force chain particles make up from 10% to 60% of the total number of particles corresponding to unjammed and jammed states, respectively [11, 41]. We find these force chains reside in regions of relatively high connectivity, as evident in their higher average degree and clustering coefficient (a measure of the density of 3-cycle configurations in a network [16, 40]).

As in past studies, we find that force chains favor contacts with laterally supporting neighbors in 3-cycle, truss-like formations [11, 39], specifically, through conformation 4$_1^0$ (Figs. 2-3). Non-force chains frequently, though not exclusively, comprise particles in less connected conformations 2$_0^0$ and 3$_0^0$, which have no 3-cycles. A dual buckling resistance underpins this cooperative evolution between columnar force chains and truss-like 3-cycles: 3-cycles (i) "prop-up" the force chain to restore alignment (Figs. 2-3), and (ii) frustrate particle rotations integral to buckling [16, 39].

Force chains are not only primary load bearers, but are also major repositories of stored energy in the system [19, 27, 29, 38]. Consequently, their failure by buckling directly: (i) reduces the load the material can carry, and (ii) leads to dissipation following the collective release of stored energy accumulated at member contacts [30, 37, 38]. For instance, note the decrease in the brightness of the birefringence pattern in Fig. 2 as a force chain buckles. Load-carrying capacity and energy dissipation form the two halves of mathematical formulations that relate the deformation to the stress in a dissipative material (i.e., continuum models). To achieve reliable and robust predictions, such models must incorporate the kinematics of those rearrangement events which are chiefly responsible for energy dissipation (i.e., internal variables or plastic strains [20, 23, 26, 36]). Knowing what conformational transitions are likely to occur when a force chain buckles and their associated probabilities, forms an essential and heretofore missing element in continuum models [26, 30]. To date, the modeller’s only recourse has been to offer either an "educated guess" [26, 30] or empirical evolution laws for the plastic strain with no clear connection to the geometric or mechanical details associated with force chain buckling [20, 22, 29]. This study shows that, within a buckling force chain, the most likely conformational transitions tend to either preserve the topology or involve a transition from a more connected conformation (i.e., 3$_0^0$ or 4$_1^0$) to a less connected conformation (i.e., 3$_1^0$), as seen in Figs. 2-3. Conformational transitions from 4$_1^0$ to 3$_1^0$ involve loss of both a contact and supporting 3-cycle. Such transitions are not the most dominant within the assembly, indicating that buckling does not have to be ripe to induce formation of shear bands and/or global failure. This confirms past findings on the initiation of failure in dense systems from numerical and experimental data [11, 30, 36–38].

V. CONFORMATIONAL STABILITY, TRANSITIONS AND DYNAMICAL BARRIERS

With the recently developed techniques from structural mechanics [11] and the ability to measure contact forces [10, 41, 46], we can now compute the structural stability of the cluster conformation for each particle, at every equilibrium state of the loading history – and study their transition dynamics. The structural stability measure in Tordesillas et al. [11] depends on material properties, the contact forces, as well as the number, positions and directions of contacts within the cluster. From the frequency distribution of conformational stabilities, we find pronounced peaks that persist throughout the loading history (Fig. 4). That such persistent peaks emerge in the stability landscape is surprising, given the extreme heterogeneity in the factors that govern the evolution of stability of a particle relative to its first ring of contacting neighbors. The most dominant conformations and conformational transition sets (sets 1 – 4 in Fig. 1 bottom) mainly lie within the stability peaks evident from the well-resolved “bands” in Fig. 4. In particular, the conformations distinguish themselves by increasing stability, which correlates well with an increasing central particle degree: for example, conformations 3$_0^0$ and 3$_1^0$ mainly reside in a lower stability peak compared to 4$_1^0$ and 4$_2^0$. The peaks in the conformational frequency distribution and the stability landscape are reminiscent of so-called “magic numbers” encountered in molecular clusters, whose physical rearrangements also govern function
FIG. 2: (Color online) The network topology of contacts and contact forces for a representative force chain over ten consecutive stages of the loading history of the sample. This process is driven by cooperative interactions among structural building blocks. Columnar force chains bearing the majority of the load, rely on confining contacts with comparatively weak neighbors for lateral support. As the force chain buckles, some 3-cycles strengthen leading to 3-force cycles [39] which prop-up the buckling chain to restore alignment. Compare, for example, stage 1 with stages 6-7: note the increased birefringence brightness from the left (right) 3-cycle supporting the 2nd-3rd (1st, 5th-6th) particles from the bottom of the force chain identified in Fig. 3.

through ways in which topology and stability are strongly interrelated [50–52].

We next consider transition dynamics within the stability landscape (Fig. 4). Instead of directly describing transitions between the 28 conformations as a Markov chain, we partition the range of stability values (i.e., (0, 0.1190)) into ~ 500 contiguous intervals and model transitions between stability intervals as a Markov chain. To better resolve highly frequented stability values, we use an adaptive partition: that is, the frequency of a conformation possessing a stability value in each interval is roughly equal. We found distinct peaks in stability, Fig. 5 top right, that are consistent with those in Fig. 4. In particular, we find a surprisingly smooth build-up of probability density to each stability peak followed by a precipitous drop.

Sharp changes in the entries of the second largest eigenvector of the stability value transition matrix, as in Fig. 5 right, delineate dynamical barriers that are infrequently traversed [48]. These sharp changes partition the stability landscape into six major bands (Fig. 5 right): 1 : (0, 0.0315), 2 : (0.0315, 0.0397), 3 : (0.0397, 0.0472), 4 : (0.0472, 0.0597), 5 : (0.0597, 0.0630) and 6 : (0.0630, 0.1190). Transitions within each of these six bands are much more likely than transitions between different bands. This banding structure can also be seen in the block-like structure of the full stability transition matrix (Fig. 5 left).

By aggregating the fine stability intervals into the six large bands, we arrive at a $6 \times 6$ transition matrix describing band-to-band transitions. Typically 40 to 60% of stability transitions are within a band. However, we
FIG. 3: (Color online) The conformational transitions of the particles in the force chain in Fig. 2. Particles which change in conformation by the next stage, over stages 1-9, are coloured: yellow (blue) particles gain (lose) connectivity by changing contacts and/or 3-cycles. Together with Fig. 2, this figure highlights the dynamics of particles in force chains associated with the continual regulation of the topology of, and forces through, their contacts.

Also observe that inter-band stability transitions prefer to jump over the intervening stability band: that is, transitions between bands 1, 3, and 5 and between bands 2, 4, and 6 are favored (cf. the interlacing of low entries in the block matrix of Fig. 5 left and the dotted and solid demarcation of the bands in Fig. 5 right). Based on this observation, we can further aggregate by setting state 1 to be the union of bands 1, 3, and 5, and state 2 to be the union of bands 2, 4, and 6, to produce a $2 \times 2$ transition matrix $B = [0.9215 \ 0.0768; 0.1872 \ 0.8115]$ which effectively partitions the stability space into two almost-invariant stability regions, each consisting of three interlaced stability bands. That is, transitions within state 1 (stability bands 1, 3, 5) occur with probability 0.9215 and transitions within state 2 (stability bands 2, 4, 6) occur with probability 0.8115. On average 70.9% of clusters are in state 1 and 29.1% of clusters are in state 2. The diagonal dominance of $B$ further highlights the “jumping” between every second stability band. Some of the preferred conformations (e.g., $3_0^1$) appear in more than one stability band, thereby indicating that topology does not completely overshadow all other factors that influence stability.

Conformational stability transitions during force chain failure by buckling

So far we have discussed the dynamics of force chains, in particular, the continual loss and formation of supporting contacts as the chain responds to and resists an applied force, leading ultimately to buckling. We now focus on the stability transitions of force chain particles during this ultimate failure state of buckling – a mechanism implicated as the precursor for shear band formation and
FIG. 4: (Color online) Stability landscape. The landscape of stability during loading showing the most prevalent granular conformations found in the most resolved stability bands of the cyclic shear experiment.

global failure in dense materials [13, 17, 27, 31, 44]. Conformations of particles in columnar force chains typically possess higher stability (Fig. 6) and those in buckling force chains (BFC) undergo a similar process of jumping over one stability band (Fig. 6 inset). A relatively higher probability exists for BFC transitions from higher to lower stability states than vice versa. That some force chain particles see an increase in their stability during buckling, highlights the system’s continual regulation of the topology of contacts and forces to adapt and delay imminent collapse of these load-bearing structures (Figs. 2 & 3). This is entirely consistent with earlier findings on “3-force cycles”, i.e., 3-cycles whose contacts each bear above the global average force. These were found to emerge just at the time and location in the sample in greatest need of support: the onset of force chain buckling in what ends up being the shear band [39].

Past experiments and simulations have shown that the build-up of new chains and collapse by buckling of old force chains govern granular deformation in the presence of shear bands [10, 11, 13, 14, 21, 27, 30, 38, 44]. The “critical state regime” of soil mechanics, in which the material flows at near-steady shear stress and global volume, is believed to occur when these two processes balance each other [13, 43]. Focussing on the six stability bands, we find the following: the topology of the dominant cluster in the higher stability bands has greater or equal degree relative to the dominant cluster in the lower stability bands; different stability bands can have the same dominant cluster topology; conformations of force chain particles are among the dominant conformations within a stability band; the topology of buckled force
FIG. 5: (Color online) Favoured pathways and dynamical barriers in the stability landscape. Scaled entries of the Markov transition matrix reveal the block structure of the stability bands (left). The approximate invariant density of the stability transition matrix revealing the preferred stability peaks (top right). Sudden changes in the entries of the second largest eigenvector correspond to the transition matrix blocks and determine the stability band barriers indicated by alternating solid and dashed lines (bottom right).

TABLE I: Dominant cluster (DC) within stability bands and relationship to particles in force chains and buckled force chains. In bands 5 and 6, multiple clusters whose populations are on a par with each other dominate.

<table>
<thead>
<tr>
<th>Stability band</th>
<th>Dominant cluster (DC) in force chain</th>
<th>% of DC in force chain</th>
<th>% of buckled force chain with DC</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>$2_1^4$</td>
<td>40%</td>
<td>68%</td>
</tr>
<tr>
<td>2</td>
<td>$2_2^4$</td>
<td>43%</td>
<td>42%</td>
</tr>
<tr>
<td>3</td>
<td>$3_3^4$</td>
<td>65%</td>
<td>83%</td>
</tr>
<tr>
<td>4</td>
<td>$3_5^4$</td>
<td>65%</td>
<td>54%</td>
</tr>
<tr>
<td>5</td>
<td>$4_4^4, 4_5^4$</td>
<td>79%, 73%</td>
<td>54%, 37%</td>
</tr>
<tr>
<td>6</td>
<td>$4_6^4, 4_5^4, 5_3^4$</td>
<td>80%, 76%, 87%</td>
<td>31%, 16%, 18%</td>
</tr>
</tbody>
</table>

In summary, the mechanical response of a granular material under load – ultimately rooted in the physical motions or rearrangements of the particles – is systematically and comprehensively quantified here by a study of conformational and stability transition dynamics. We have examined the dynamics of that class of conformations consisting of a particle and its first ring of contacting neighbors in a quasi-statically deforming granular material. Relative probabilities of the 28 conformations found, and conformational transitions, have identified the prevalent and persistent rearrangements, including those in key functional structures – the primary load-bearing force chains – which govern the strength and load-carrying capacity of the material. We found self-organization towards favored equilibrium cluster conformations which, in turn, reside within favored stability states in a stability landscape with infrequently traversed dynamical barriers. The most likely conformational transitions during force chain failure by buckling, the mechanism believed to be responsible for global granular failure,
FIG. 6: (Color online) Force chains need lateral support for stability. Cumulative distribution function (CDF) of stability of force chain particle conformations (red square) and non-force chain conformations (blue triangle). Force chain particle conformations generally possess higher stability. (Inset) The six stability band transition probabilities of buckled force chain (BFC) particle clusters are typically within the same band (diagonally dominant) or to a lower stability band; probabilities below the diagonal are greater than those above the diagonal demonstrating that buckling is more likely to cause a transition from a higher stability state to a lower stability state.

correspond to rearrangements among, or loss of, contacts which break the stabilizing truss-like 3-cycles. We note that the patterns obtained from experimental observations reported here were also found, at least qualitatively, in a discrete element simulation of a two-dimensional polydisperse assembly under monotonic biaxial compression with constant confining pressure.

The granular cluster dynamics uncovered here provides direct structural and mechanical insights into the mechanisms that underpin the rheology of granular media. Conformational transition probabilities comprehensively quantify the relative contributions of the various rearrangements to deformation, including those directly responsible for energy dissipation and global failure of the material. This opens the door to robust and tractable predictive continuum models of granular deformation and failure (e.g., [21]). The patterns of self-organization are remarkably reminiscent of those seen in molecular clusters (e.g., [50, 51]). Anomalously high density stability states, populated by clusters with specific number of contacts in the first ring of neighbors, hint at the possible existence of “magic numbers” in granular systems. In contrast to molecular systems for which the complete tracking of molecular bond formation and breakage is as yet out of reach experimentally [50], granular clusters offer a unique paradigm for studies of emergence and self-organization through conformational transition dynamics: the formation and loss of contacts, as well as the force
between constituent particles can be completely tracked in real time – enabling the connections between topology, structural stability, dynamics and function to be directly unravelled. Recent studies on nanoscale viral assemblies which found that “concepts borrowed from macroscopic materials science are surprisingly relevant” [52] highlight further common ground and potential synergies in concepts and techniques for unraveling self-organization across a vast range of length scales in complex materials.

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