Structural and topological nature of plasticity in sheared granular materials

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Upon mechanical loading, granular materials yield and undergo plastic deformation. The nature of plastic deformation is essential for the development of the macroscopic constitutive models and the understanding of shear band formation. However, we still do not fully understand the microscopic nature of plastic deformation in disordered granular materials. Here we used synchrotron X-ray tomography technique to track the structural evolutions of three-dimensional granular materials under shear. We establish that highly distorted coplanar tetrahedra are the structural defects responsible for microscopic plasticity in disordered granular packings. The elementary plastic events occur through flip events which correspond to a neighbor switching process among these coplanar tetrahedra (or equivalently as the rotation motion of 4-ring disclinations). These events are discrete in space and possess specific orientations with the principal stress direction.

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Granular solids yield and flow upon applied stress\(^1,2\). So far, the flow behaviors of granular materials have mainly been treated macroscopically based on empirical constitutive laws\(^3-5\). More recent approaches treat granular materials within the category of amorphous solids and try to identify the microscopic plastic events to derive the macroscopic mechanical properties\(^6\). It is generally believed that microscopic plastic events in amorphous solids are induced by certain spatially isolated structural “defects” in the system, and the macroscopic yielding, avalanche and shear band formation are induced by their elastic interactions\(^6\). However, the exact nature of these “defects” remain elusive\(^6\) and it has been investigated based on free volume\(^19\), change of local topology\(^11\), energy landscape\(^12\), shear transformation zones (STZ)\(^13,14\), soft spots as determined by low-energy soft modes\(^15,16\), buckled force chains\(^17\), or defects of an amorphous order\(^18-21\). Experiments on two-dimensional (2D) soap bubble rafts have identified the elementary plastic event as T1 event which corresponds to two pairs of bubbles switching neighbors with each other\(^22-24\). Confocal microscopy experiments on three-dimensional (3D) colloidal systems have revealed the elementary plasticity events happening at shear transformation zones with a core radius around three particle diameters\(^25\).

However, the structural basis and topological pathways for these plasticity events have not been investigated in detail. Scattering techniques have also been used to probe local defects in granular systems\(^26\).

In the present study, we carry out quasi-static shear experiments on a 3D disordered granular system, and obtain its structural evolutions by synchrotron X-ray tomography technique (Methods). We find that, similar to T1 event in 2D, the elementary plastic events in 3D are flip events, which consist of two pairs of particles switching neighbors with each other at highly distorted coplanar tetrahedra (structural defects of polytetrahedral order) on Delaunay network. These flip events can equivalently be described as the rotation motions of 4-ring dissociation defects in the system and possess specific orientations with the principal stress direction. We therefore establish highly distorted coplanar tetrahedra as dissociation-like structural carriers of plasticity in disordered granular packings, and the flipping processes of them induce plastic deformations.

### Results

**Shear band formation.** Figure 1a is a schematic presentation of the shear cell used in our experiment\(^47\). Particles inside are monodisperse glass particles (Duke Scientific, \(d = 200 \pm 6 \mu m\)). The samples are prepared with different initial volume fractions \(\phi\) and thicknesses \(W\) (Supplementary Table 1) and are sheared in the \(z\)-direction. More details can be found in the Methods section. A tomography scan is carried out after each shear step during which the shear bracket moves up by about \(1/3d\). Particle trajectories in the imaging window can be traced during the entire shear process. We define \(\Delta \tau (i = x, y, z)\) as the displacement of a particle after a shear step. During the whole shear process, \(\Delta \tau_i\) is the dominant one among all three components. Figure 1b shows \(\Delta \tau_i\) at the beginning of the shear process. The particles with displacement \(\Delta \tau_i > 0.008d\) form a boundary inclined from the vertical direction. This is mainly due to a net positive \(\Delta \tau_i\) component when the system dilates at the beginning of the shear. Upon further shear, when the volume fraction \(\phi\) reaches a steady state value, a vertical shear band is formed (Fig. 1c) which can be easily seen either by the spatial distribution of particles with \(\Delta \tau_i > 0.008d\) (Fig. 1c) or the distribution of \(\langle \Delta \tau_i \rangle\), along \(x\)-direction (averaged for all particles located within a \(1d\)-thickness slice centered at different \(x\), Fig. 1d). From Fig. 1d, we estimate the shear band thickness to \(x = 1d\). The corresponding strain associated with each shear step can therefore be calculated as \(\gamma = \frac{\Delta \tau_i}{\Delta \tau_i(x=1d)}\), where \(\langle \Delta \tau_i \rangle_{x=1d}\) is the mean value of \(\Delta \tau_i\) for particles located around \(x = 1d\) after each shear step. It turns out \(\Delta \tau_i = 1.2 \pm 0.2\%\) during the whole-shear process (Supplementary Note 1). The cumulative strain is calculated by \(\gamma = \sum \Delta \tau\) and a total strain \(\gamma = 86\%\) is obtained consisting of 71 shear steps for all three samples measured. Despite different initial conditions, each sample reaches steady state forming shear band with similar width and \(\phi \sim 0.59\) after a cumulative critical strain \(\gamma \sim 40\%\) (Fig. 1e). Once steady state is reached, the shear band is in a flow state where the structure continuously relaxes (Supplementary Note 2).

**Non-affine displacement.** Next, we investigated microscopic plastic deformations by the particles’ non-affine displacements \(\delta r_i (i = x, y, z)\) which we define as \(\Delta \tau_i (i = x, y, z)\) of the particle minus the corresponding average \(\langle \Delta \tau_i \rangle (i = x, y, z)\) of all particles within its radial distance of \(2.5d\). It is worth noting that our results are not sensitive to this threshold value or to the particular way non-affine displacement is calculated\(^14,28\) (Supplementary Note 3). Figure 1f, g shows \(\delta r_i\) for particles at the beginning of the shear and when the system has reached steady state (only particles having \(|\delta r_i| > 0.008d\) are shown), respectively. Contrary to \(\Delta \tau_i\) which is dominated by \(\delta r_i\), \(\delta r_i\) have similar magnitudes along all three axes. At the beginning of the shear, there are more particles which have significant non-affine displacements \(|\delta r_i| > 0.008d\) but on average the absolute values are small. In contrast, at the steady state, \(|\delta r_i|\) have large absolute values within the shear band (Fig. 1g) and small values outside of it, which is consistent with the general belief that shear band consists of significant plastic activities\(^1\).

**Topology change of local structures.** To understand the structure and topology change upon shear, we partitioned the structural configurations of particles at different shear steps by Delaunay tessellation\(^20\). We use the parameter \(\delta = e_{\text{max}} - 1\) to characterize the shape of a tetrahedron, where \(e_{\text{max}}\) in units of mean particle diameter \(d\), is the length of the longest edge of the tetrahedron. A smaller value of \(\delta\) suggests that the tetrahedron is closer to a regular one. Upon shear, both structure and topology of the system can vary (Supplementary Note 4). Correspondingly, each tetrahedron can get distorted and eventually destroyed (its vertices no longer belong to the original tetrahedron) which leads to a local topology change. We term a tetrahedron unstable when it is destroyed after a shear step. However, for topological reasons a single tetrahedron cannot get destroyed on its own. In 2D, the topology change follows a specific pathway called T1 event which corresponds to a neighbor switching process\(^22\). It also corresponds to the destruction of two Delaunay triangles and the subsequent formation of two new ones\(^23\). In 3D, as shown in Fig. 2a, the topology change happens through pathways called flip events: In a 2–2 flip, two neighboring pairs of coplanar unstable tetrahedra form two new pairs of coplanar tetrahedra by exchanging their vertices, the 2–2 flip is equivalent to its counterpart T1 event in 2D\(^25\); additionally, a pair of unstable coplanar tetrahedra can also split into three coplanar tetrahedra, or vice versa, which is denoted as 2–3 (or 3–2) flip. The 2–3 (or 3–2) flip, despite its topological significance, could be considered to be only an intermediate step of 2–2 flip in our system, since a consecutive 2–3 and 3–2 flip will yield a 2–2 flip (Fig. 2a) and in reality they almost always happen successively. This is due to the fact that the transient structure is mechanically very unstable. In Fig. 2b, we also analyze the spatial distribution of flip events by showing the cluster size distribution of unstable tetrahedra in space (unstable tetrahedra which are face-adjacent to each other
are considered to belong to one cluster). From the distribution we can conclude that flip events are spatially localized since the cluster size is predominantly two (2–3 flip), three (3–2 flip) or four (2–2 flip), comprising only a single flip event. To analyze the occurrence probability of flip events, we calculated the flip frequency among all possible couples or triples in which we term any two coplanar tetrahedra as a couple and three coplanar tetrahedra as a triple. In a single shear step, only about 6% of couples or triples will flip and flips are more frequent in the shear band regime. Figure 2c shows the locations of the flipped couples and triples in the x–z plane within a 2d thickness (−1d<y<1d) after a shear step, overlaid with the corresponding non-affine displacement field at y = 0 when shear band is formed. The non-affine displacement field has been smoothed over a distance of two particle diameters. It is clear that correlation exists between the flip sites and cores of large non-affine displacement regions. Furthermore, we found that the orientational angles of flip events (Methods) are strongly correlated with the principal stress direction. Figure 2d shows the angular density distributions of the orientations of flipped tetrahedral groups with respect to the horizontal plane (x–y plane). It is clear that unstable couples and triples have preferred orientations before and after the flips. Specifically, the couples with an orientation around +45° are more likely to flip to form −45° couples or +45° triples, while the −45° triples are more likely to flip to form −45° couples. This is due to the fact that the particle distance tends to be compressed along the principal stress direction and expanded in the orthogonal direction which makes tetrahedral groups in specific orientations more vulnerable to flip instability. Once they flip, they have orientations which are difficult to flip again. We rule
depend on their shape and how their spatial proximity to other tetrahedra influence their non-affine behavior. We classify tetrahedra into three types based on their deformation by non-affine displacements: unstable tetrahedra, tetrahedra with none of their vertices involved in flip events (stable tetrahedra), and tetrahedra with some of their vertices involved in flip events (intermediate tetrahedra). We further investigate the connection between the shapes of the tetrahedra and flip events. From Fig. 3d, it is obvious that flip is much more likely among highly distorted tetrahedra which have δ > 0.245. And the more distorted, the more likely a tetrahedron will flip in the subsequent shear step. Stable tetrahedra are more likely to have smaller δ, i.e., more regular shape. Intermediate tetrahedra tend to have shapes between these two extremes. These results are reminiscent of previous findings in which a polytetrahedral glass order based on quasi-regular tetrahedra has been defined in granular packings based on δ > 0.245. It is interesting to see that the defective structure associated with this order plays a significant role in plasticity, similar to the role played by dislocations in crystals. As shown in the following, the analogy is much more profound as we find that the flip process of unstable tetrahedra is equivalent to the rotation motions of 4-ring disclinations which are topological defects associated with rotational degrees of freedom (Supplementary Table 2). Although the shape parameter δ does not influence the non-affine displacements directly (Fig. 3b), it nevertheless yields a facilitation mechanism for subsequent plastic activities, i.e., a tetrahedron has to be heavily distorted to undergo a flip event upon shear, and it is therefore more likely to have a new flip event earlier than a previous one since the tetrahedra on average are more distorted there (Supplementary Note 5). This facilitation effect is discernable within a diameter of 4d of the flipped site similar to the influence zone as observed in Fig. 3c, which is obtained by analyzing the spatial correlation of unstable tetrahedra and their neighbors, i.e., the mean square non-affine mobility μ = 1/4 \sum_{j=1}^{4} \delta r_j^2, i.e., the mean square non-affine displacements of the four vertices of the tetrahedron. As shown in Fig. 3b, μ is largest for unstable tetrahedra, smallest for stable tetrahedra, and has values in between for intermediate tetrahedra, in agreement with the result of Fig. 2c. It is also interesting to note that μ has a very weak dependency on the tetrahedral shape δ for stable tetrahedra. Since μ is directly related to the flip events and spatial proximity to them, we plot in Fig. 3c μ as a function of distance from unstable and stable tetrahedra. We recognize that unstable tetrahedra correspond to the cores of large plastic activities and the stable tetrahedra correspond to cores of much weaker but still finite plastic activities. The two curves roughly merge around r = 4d, which yields the range of influence zone. Next we investigate the connection between the shapes of the tetrahedra and flip events. From Fig. 3d, it is obvious that flip is much more likely among highly distorted tetrahedra which have δ > 0.245. And the more distorted, the more likely a tetrahedron will flip in the subsequent shear step. Stable tetrahedra are more likely to have smaller δ, i.e., more regular shape. Intermediate tetrahedra tend to have shapes between these two extremes. These results are reminiscent of previous findings in which a polytetrahedral glass order based on quasi-regular tetrahedra has been defined in granular packings based on δ > 0.245. It is interesting to see that the defective structure associated with this order plays a significant role in plasticity, similar to the role played by dislocations in crystals. As shown in the following, the analogy is much more profound as we find that the flip process of unstable tetrahedra is equivalent to the rotation motions of 4-ring disclinations which are topological defects associated with rotational degrees of freedom (Supplementary Table 2). Although the shape parameter δ does not influence the non-affine displacements directly (Fig. 3b), it nevertheless yields a facilitation mechanism for subsequent plastic activities, i.e., a tetrahedron has to be heavily distorted to undergo a flip event upon shear, and it is therefore more likely to have a new flip event earlier than a previous one since the tetrahedra on average are more distorted there (Supplementary Note 5). This facilitation effect is discernable within a diameter of 4d of the flipped site similar to the influence zone as observed in Fig. 3c, which is obtained by analyzing the spatial correlation of
flip events of two subsequent shear steps. The existence of facilitation mechanism thus indicates a spatial and temporal correlations between flip events. Recently, the collective behavior of microscopic plastic deformations in relation to macroscopic force fluctuations has been analyzed in sheared granular materials, and it is observed that a significant long-range strain correlation is directly related to the macroscopic avalanche behavior. It is therefore interesting to investigate in the future how the local facilitation mechanism as identified here is related to shear band formation and avalanches.

**Topological nature of plasticity.** It is well-known that for crystalline materials the carriers for microscopic plasticity are dislocations which are topological defects associated with translational degrees of freedom. For our system, we also characterized the topological nature of the flip events based on N-ring disclination structures. An N-ring structure on Delaunay network represents a tetrahedral group with one edge as a common axis and coplanar between neighboring members. A 5-ring structure is considered to be the disclination-free ground state structure, whereas other N-ring structures possess disclination defects, which are topological defects associated with rotational degrees of freedom. The N-ring concept was originally developed to describe the potential ideal glass state. Since 5-ring structures alone cannot tile space, the ideal glass structure is conjectured to possess evenly spaced 6-ring structures in a 5-ring structure background as introduced by Frank and Kasper. For low-density hard sphere systems, there should therefore exist many disclination defects. As shown in Fig. 4a, consistent with this picture, the 5-ring is most populous in the initial dense state, decreasing in amount after the shear, and the steady state has significantly more disclination defects. Interestingly, the only way an N-ring structure can transform into another one is through flips. It turns out that 2–2 flip is equivalent to a 4-ring structure rotating into another 4-ring structure (Fig. 2a) and 2–3/3–2 flips correspond to a 4-ring structure transforming into a 3-ring structure and vice versa. Since a 3-ring structure (even more distorted tetrahedra) is mechanically very unstable and therefore only exists transiently, plasticity in our system essentially happens through 2–2 flips or the rotation of 4-ring structures. We emphasize that a 2–2 flip is therefore the only pathway for N-ring structure to transform between each other, e.g., Fig. 4b shows how a 2–2 flip process can change a neighboring 5-ring into a 4-ring structure. We therefore establish close connections between a 2–2 flip, rotation of a 4-ring structure, and local plasticity.

**Discussion**

In conclusion, we find that elementary plastic events in sheared granular materials mainly happen through flip events of highly distorted coplanar tetrahedra of the Delaunay network. This result supports the concept that highly distorted coplanar tetrahedra are structural defects of disordered granular packings and carriers of microscopic plasticity. Since flip events can also be described as the rotation motions of 4-ring disclinations which are topological defects associated with rotational degrees of freedom, close analogies with dislocations in crystals can be drawn. We believe our results should not be considered as applicable to granular materials only, but also to atomic and molecular amorphous systems, despite the fact that granular materials are athermal and have friction. To understand why the presence of friction does not modify the overall picture, it is useful to compare the potential/free energy landscape of athermal glassy system with the one of a frictional granular system.
length scale of the size of the particles. However, because of friction, the former will remain very rugged even on much smaller scales; whereas, the landscape of an atomic-glass for-
mers is basically smooth for length scales below the size of atoms. This difference in the landscapes will lead to rather different behavior in their plastic behaviors. Despite the fact that the topological pathways to a local saddle point in a granular material and a thermal glass will be quite similar, the microscopic dyna-
mical and plastic behaviors of their two systems are quite dif-
ferent. For thermal glassy systems, the overcoming of the landscape barrier is related to thermal fluctuations and it will be
instantaneous. In granular materials, on the other hand, since the pathway can be stabilized by friction, it can freeze the motion on the topological pathway of a plastic deformation followed in thermal glassy systems. So in general we expect that the structural and topological characteristics of plastic deformations as observed in our system will remain also valid in thermal glassy systems, i.e., our results should be applicable to a wide range of amorphous materials, thus allowing to gain insight into mechanical properties of such materials.

Methods

Experimental details. A shear setup suitable for X-ray tomography study was built. As shown in Fig. 1a, the setup is a rectangular acrylic glass container with dimension of $8(L) \times 7(W) \times 32(H)$ mm$^3$. The shear is generated by a 2-mm-thick L-shaped bracket which can move in the vertical direction against a block of width $W$. The coordination axes are set so that the shearing direction is along $+z$- direction, opposite to gravity. The direction normal to the shear plane is the x-
direction with coordination zero at the interface between the bracket and the block. Two blocks with $W = 25d$ and $W = 15d$ were used to investigate the influence of the boundary on shear band formation. The bracket surface and the opposite surface of the container were roughened by glued glass particles. Before each shear sequence, the particles are slowly poured into the container up to a height around the middle of the sample. A complete tomography scan consists of 1500 projection images, which takes about 5 min. Through image processing and particle tracking algorithms, the centroids and trajectories of the all particles can be determined to be within an error less than 3.3 $\times 10^{-3}$d.

Fig. 4 Evolution and transformation of N-ring structures under shear. a Evolution of the fraction of N-ring structures upon shear. b A 5-ring structure with AB as common axis, evolves into a 4-ring structure as the couple ABCE/ABCD forms a new couple DEBA/DEBC through 2-2 flip as AC and DE switch neighbors

Data availability. The datasets generated during and/or analysed during the current study are available from the corresponding author on reasonable request.

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Y.W. designed the research. Y.C., J.L., B.K., C.X., Z.L., R.C., H.X., T.X., K.W., L.H., I.Z., and Y.W. performed the experiment. Y.C., J.L., W.K., and Y.W. analyzed the data and wrote the paper.

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