Memory of jamming – multiscale flow in soft and granular matter

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Soft, disordered, micro-structured materials are ubiquitous in nature and industry, and are different from ordinary fluids or solids, with unusual, interesting flow properties. The transition from fluid to solid – at the so-called jamming density – features a multitude of complex mechanisms, but there is no unified theoretical framework that explains them all. In this study, a simple yet quantitative and predictive model is presented, which allows for a changing jamming density, encompassing the memory of the deformation history and explaining a multitude of flow phenomena at and around jamming. The jamming density, introduced as a new state-variable, changes due to the deformation history and relates the system’s macroscopic response to its micro-structure. The packing efficiency can increase logarithmically slow under gentle “tapping” or repeated (isotropic) compression, leading to an increase of the jamming density. In contrast, shear deformations cause anisotropy, changing the packing efficiency exponentially fast with either dilatancy or compactancy. The memory of the system near jamming can be explained by a micro-statistical model that involves a multiscale, fractal energy landscape and links the microscopic particle picture to the macroscopic continuum description, providing a unified explanation for the qualitatively different flow-behavior for different deformation modes. To complement our work, a recipe to extract the history-dependent jamming density from experimentally accessible data is proposed, and alternative state-variables are compared. The proposed simple, usable macroscopic model, will help predicting and avoiding geophysical hazards, bring forward industrial process design and optimization, and understand and solve scientific challenges in fundamental research.

1. Introduction

Granular materials are a special case of soft-matter with micro-structure, as also foams, colloidal systems, glasses, or emulsions (Denisov et al. 2013; Trappe et al. 2001; Walker et al. 2015). Particles can flow through a hopper or an hour-glass when shaken, but jam (solidify) when the shaking stops (Wambaugh et al. 2007). These materials jam above a “certain” volume fraction, or jamming density, referred to as the “jamming point” (Bagnan et al. 2013; Bi et al. 2011; Cates et al. 1998; Coulais et al. 2014; Liu & Nagel 1998, 2010; Majmudar et al. 2007; O‘Hern et al. 2003; Otsuki & Hayakawa 2011; Pica Ciamarra & Coniglio 2009; Reichhardt & Reichhardt 2014; Silbert 2010; Silbert et al. 2006, 2009; Song et al. 2008; Torquato & Stillinger 2010; Van Hecke 2010; Walker et al. 2015; Wang et al. 2012; Zhang & Makse 2005), and become mechanically stable with finite bulk- and shear-moduli (Dagois-Bohy et al. 2012; Inagaki et al. 2011; Métayer et al. 2011; O‘Hern et al. 2003; Otsuki & Hayakawa 2011; Parisi & Zamponi 2010; Pica Ciamarra & Coniglio 2009; Zhang & Makse 2005). Notably, these systems “flow” even in the jammed state by

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reorganizations of their micro-structure (Farhadi & Behringer 2014; Saitoh et al. 2015).

Around the jamming transition, these systems display considerable inhomogeneity, such as reflected by over-population of weak/soft/slow mechanical oscillation modes (Silbert et al. 2006), force-networks (Radjai et al. 1998; Silbert 2010; Snoeijer et al. 2006), diverging correlation lengths and relaxation time-scales (Brown & Jaeger 2009; Lerner et al. 2012; O’Hern et al. 2003; Reichhardt & Reichhardt 2014; Wang et al. 2012), and some universal scaling behaviors (Otsuki & Hayakawa 2012). Related to jamming, but at all densities, other phenomena occur, like shear-strain localization (Otsuki & Hayakawa 2011; Peyneau & Roux 2008; Schall & van Hecke 2009; Singh et al. 2014; Van Hecke 2010), anisotropic evolution of structure and stress (Bi et al. 2011; Ciamarra et al. 2011; Imole et al. 2013, 2014; Kumar et al. 2014a; O’Hern et al. 2003; Peyneau & Roux 2008; Radjai et al. 1998; Schall & van Hecke 2009; Silbert et al. 2006; Singh et al. 2014; Snoeijer et al. 2006; Wang et al. 2012), and force chain inhomogeneity (Bi et al. 2011; Cates et al. 1998; Saitoh et al. 2015). To gain a better understanding of the jamming transition concept, one needs to consider both the structure (positions and contacts) and contact forces. Both of them illustrate and reflect the transition, e.g., with a strong force chain network percolating the full system and thus making unstable packings permanent, stable and rigid (Bi et al. 2011; Cates et al. 1998; Walker et al. 2014; Wang et al. 2013; Zhang et al. 2010b).

For many years, scientists and researchers have considered the jamming transition in granular materials to occur at a particular volume fraction, $J$ (Brown & Hawksley 1945). In contrast, over the last decade, numerous experiments and computer simulations have suggested the existence of a broad range of $J$, even for a given material. It was shown that the critical density for the jamming transition depends on the preparation protocol (Bandi et al. 2013; Charbonneau et al. 2012; Liu & Nagel 2010; Mari et al. 2009; O’Hern et al. 2001; Olsson & Teitel 2011, 2013; Otsuki & Hayakawa 2011, 2012; Ozawa et al. 2012; Reichhardt & Reichhardt 2014; Torquato & Stillinger 2010; Torquato et al. 2000), and that this state-variable can be used to describe and scale macroscopic properties of the system (Inagaki et al. 2011). For example, rheological studies have shown that $\phi$ decreases with increasing compression rate (Ashwin et al. 2013; Mari et al. 2009; Vågberg et al. 2011; Zhang & Makse 2005) (or with increasing growth rate of the particles), with the critical scaling by the distance from the jamming point ($\phi - \phi_J$) being universal and independent of $\phi_J$ (Charbonneau et al. 2012; Chaudhuri et al. 2010; Majmudar et al. 2007; Otsuki & Hayakawa 2012; Zhao et al. 2011) Recently, the notion of an a-thermal isotropic jamming “point” was challenged due to its protocol dependence, suggesting the extension of the jamming point, to become a $J$-segment (Ciamarra et al. 2010, 2011; Vågberg et al. 2011). Furthermore, it was shown experimentally, that for a tapped, unjammed frictional 2D systems, shear can jam the system (known as “shear jamming”), with force chain networks percolating throughout the system, making the assemblies jammed, rigid and stable (Bi et al. 2011; Farhadi & Behringer 2014; Grob et al. 2014; Ren et al. 2013; Wang et al. 2013; Zhang et al. 2010b), all highlighting a memory that makes the structure dependent on history $H$. But to the best of our knowledge, quantitative characterization of the varying/moving/changing transition points, based on $H$, remains a major open challenge.

Here, we consider frictionless sphere assemblies in a periodic system, which can help to elegantly probe the behavior of disordered bulk granular matter, allowing to focus on the structure (Walker et al. 2015), without being disturbed by other non-linearities (Bi et al. 2011; Farhadi & Behringer 2014; Hartley & Behringer 2003) (as e.g. friction, cohesion, walls, environmental fluids or non-linear interaction laws). For frictionless assemblies, it is often assumed that the influence of memory is of little importance, maybe even
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negligible. If one really looks close enough, however, its relevance becomes evident. We quantitatively explore its structural origin in systems where the re-arrangements of the micro-structure (contact network) are the only possible mechanisms leading to the range of jamming points, i.e. a variable state-variable jamming density.

In this study, we probe the jamming transition concept by two pure deformation modes: isotropic compression and deviatoric pure shear (volume conserving), which allow us to combine the J-segment concept with a history dependent jamming density. Assuming that all other deformations can be superimposed by these two pure modes, we coalesce the two concepts of isotropic and shear induced jamming, and provide the unified model picture, involving a multiscale, fractal-type energy landscape (Krzakala & Kurchan 2007; Liu & Nagel 2010; Möbius & Heussinger 2014; Xu et al. 2011); in general, deformation (or the preparation procedure) modify the landscape and its population; considering only changes of the population already allows to establish new configurations and to predict their evolution. The observations of different $\phi_J$ of a single material require an alternative interpretation of the classical “jamming diagram” (Liu & Nagel 1998).

Our results will provide a unified picture, including some answers to the open questions from literature: (i) What lies in between the jammed and flowing (un jammed) regime? – as posed by Ciamarra et al. (2010). (ii) Is there an absolute minimum jamming density? – as posed by Ciamarra et al. (2010). (iii) What protocols can generate jammed states? – as posed by Torquato et al. (2000). (iv) What happens to the jamming and shear jamming regime in 3D and is friction important to observe it? – as posed by Bi et al. (2011). Eventually, accepting the fact that the jamming density is changing with deformation history, significant improvement of continuum models is expected, not only for classical elasto-plastic or rheology models, but also, e.g., for anisotropic constitutive models (Imole et al. 2013; Kumar et al. 2014b; Rognon et al. 2008; Sun & Sundaresan 2011), GSH rate type models (Jiang & Liu 2008, 2015), Cosserat micro-polar or hypoplastic models (Göncü 2012; Mohan et al. 2002; Tejchman 2008) or continuum models with a length scale and non-locality (Henann & Kamrin 2013). For this purpose we provide a simple (usable) analytical macro/continuum model as generalization of continuum models by adding one isotropic state-variable. Only allowing $\phi_J(H)$ to be dependent on history $H$, as key modification, explains a multitude of reported observations and can be a significant step forward to solve real-world problems in e.g. electronic industry related novel materials, geophysics or mechanical engineering.

The paper continues with the simulation method in section 2, before the micromechanical particle- and contact-scale observations are presented in section 3, providing analytical (quantitative) constitutive expressions for the change of the jamming density with different modes of deformation. Section 4 is dedicated to a (qualitative) mesoscale stochastic model that explains the different (slow versus fast) change of $\phi_J(H)$ for different deformation modes (isotropic versus deviatoric/shear). A quantitative predictive macroscale model is presented in section 5 and verified by comparison with the microscale simulations, before an experimental validation procedure is discussed in section 6 and the paper is summarized and conclusions are given in section 7.

2. Simulation method

Discrete Element Method (DEM) simulations are used to model the deformation behavior of systems with $N = 9261$ soft frictionless spherical particles with average radius $r = 1$ [mm], density $\rho = 2000$ [kg/m$^3$], and a uniform polydispersity width $w = r_{\text{max}}/r_{\text{min}} = 3$, using the linear visco-elastic contact model in a 3D box with periodic boundaries (Kumar et al. 2014b). The particle stiffness is $k = 10^8$ [kg/s$^2$], contact viscos-
ity is $\gamma = 1 \text{ [kg/s]}$. A background dissipation force proportional to the moving velocity is added with $\gamma_b = 0.1 \text{ [kg/s]}$. The particle density is $\rho = 2000 \text{ [kg/m}^3\text{]}$. The smallest time of contact is $t_c = 0.2279 \text{ [\mu s]}$ for a collision between two smallest sized particles (Imole et al. 2013).

2.1. Preparation procedure and main experiments

For the preparation, the particles are generated with random velocities at volume (solid) fraction $\phi = 0.3$ and are isotropically compressed to $\phi_t = 0.64$, and later relaxed.

From a relaxed, unjammed, stress free initial state with volume fraction $\phi_t = 0.64 < \phi_J$, we compress it isotropically to a maximum volume fraction, $\phi^\text{max}_t$, and decompress back to $\phi_t$; during the later process $\phi_J$ is identified. This process is repeated over $M$ (100) cycles, which provides different isotropic jamming points $\phi_J = M \phi_{J,i}$, related with $\phi^\text{max}_i$ and $M$ (see section 3.1).

Several isotropic configurations $\phi$, such that $\phi_t < \phi < \frac{1}{M} \phi_{J,i}$ from the decompression branch are chosen as the initial configurations for shear experiments. We relax them and apply pure (volume conserving) shear with the strain-rate tensor $\dot{\varepsilon} = \dot{e}_{11} (1, 1, 0)$, for four cycles. The $x$ and $y$ walls move, while the $z$ wall is stationary. The strain rate of the (quasi-static) deformation is small, $\dot{e}_{11} t_c < 3 \times 10^{-6}$, to avoid transient behavior.

2.2. Macroscopic (tensorial) quantities

Here, we focus on defining averaged tensorial macroscopic quantities – including strain-, stress- and fabric (structure) tensors – that provide information about the state of the packing and reveal interesting bulk features.

From DEM simulations, one can measure the ‘static’ stress in the system (Christoffersen et al. 1981) as

$$\sigma = \frac{1}{V} \sum_{c \in V} I^c \otimes f^c,$$  \hspace{1cm} (2.1)

average over all the contacts in the volume $V$ of the dyadic products between the contact force $f^c$ and the branch vector $I^c$, where the contribution of the kinetic fluctuation energy has been neglected (Imole et al. 2013; Luding 2005). The dynamic component of the stress tensor is four orders of magnitude smaller than the former and hence its contribution is neglected. The isotropic component of the stress is the pressure $P = \text{tr}(\sigma)/3$.

In order to characterize the geometry/structure of the static aggregate at microscopic level, we will measure the fabric tensor, defined as

$$F = \frac{1}{V} \sum_{P \in V} V^P \sum_{c \in P} n^c \otimes n^c,$$ \hspace{1cm} (2.2)

where $V^P$ is the volume relative to particle $P$, which lies inside the averaging volume $V$, and $n^c$ is the normal unit branch-vector pointing from center of particle $P$ to contact $c$ (Kumar et al. 2013; Luding 2005; Zhang et al. 2010a). Isotropic part of fabric is $F_v = \text{tr}(F)$. The corrected coordination number (Bi et al. 2011; Imole et al. 2013) is $C^* = M_4/N_4$, where, $M_4$ is total contacts of the $N_4$ particles having at least 4 contacts, and the non-rattler fraction is $f_{NR} = N_4/N$. For any tensor $Q$, its deviatoric part can be defined as $Q_d = \text{sgn}(q_{yy} - q_{xx}) \sqrt{3q_{ij}q_{ij}/2}$, where $q_{ij}$ are the components of the deviator of $Q$, and the sign function accounts for the shear direction, in the system considered here, where a more general formulation is given by Kumar et al. (2014b). Both pressure $P$ and shear stress $\Gamma$ are non-dimensionalized by $2(\gamma)/k$ to give dimensionless pressure $p$ and shear stress $\tau$. 
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3. Micromechanical results

3.1. Isotropic deformation

In this section, we present a procedure to identify the jamming-points and their range. We also show the effect of cyclic over-compression to different target volume fractions and present a model that captures this phenomena.

3.1.1. Identification of the jamming point

When a sample is over-compressed isotropically, the loading and unloading paths are different in pressure $p$. This difference is most pronounced near the jamming point $\phi_J$, and for the first cycle. It brings up the first question of how to identify a jamming point, $\phi_J$. The unloading branch of a cyclic isotropic over-compression along volume fraction $\phi$ is well described by a linear relation in volumetric strain, with a tiny quadratic correction:

$$p = \frac{\phi C}{\phi_J} p_0 (-\varepsilon_v) [1 - \gamma_p (-\varepsilon_v)],$$

where $p_0$, $\gamma_p$, as presented in Table 1, and the jamming point $\phi_J$ are the fit parameters, and $-\varepsilon_v = \log(\phi/\phi_J)$ is the true or logarithmic volumetric strain of the system, defined relative to the reference where $p \to 0$, i.e. jamming volume fraction. $C$ is the ratio of total non-rattler contacts $M_4$ and total number of particles $N$, i.e., $C = M_4/N = (M_4/N_4) (N_4/N) = C^* f_{NR}$, with corrected coordination number $C^*$ and fraction of non-rattlers $f_{NR}$.

Using Eq. (3.1), we can extrapolate $p$ to zero, to get $\phi_J$. We apply the same procedure for different over-compressions, $\phi_i^{\text{max}}$, and many subsequent cycles $M$ to obtain $M \phi_{J,i}$, for which the results are discussed below. The material parameter $p_0$ is finite, almost constant, whereas $\gamma_p$ is small, sensitive to history and contributes mainly for large $-\varepsilon_v$. 

Figure 1. (a) Dimensionless pressure $p$ plotted against volume fraction $\phi$ and for an isotropic compression starting from $\phi_i = 0.64$ to $\phi_i^{\text{max}} = 0.68$ (green ‘$\uparrow$’) and decompression back to $\phi_i$ for $M = 1$, leading to $\phi_J(\phi_i^{\text{max}} = 0.68) = 0.66$ and $\phi_J(\phi_i^{\text{max}} = 0.82) = 0.6652$. The blue ‘$\downarrow$’ data points represent cyclic over-compression to $\phi_i^{\text{max}} = 0.82$ for $M = 100$, leading to $100 \phi_J(\phi_i^{\text{max}} = 0.82) = 0.6692$. The $M \phi_{J,i}$ are extracted using a fit to Eq. (3.1). The upward arrow indicates the loading path (small symbols) while the downward arrow indicates the unloading path (big symbols). The inset is the zoomed in regime near the jamming point, and lines are just connecting the datasets. (b) Scaled pressure $p^{\phi_J}/\phi C$ plotted against volumetric strain $-\varepsilon_v = \log(\phi/\phi_J)$ for the same simulations as (a). Lines represents the scaled pressure, when Eq. (3.1) is used, with different $\gamma_p = -0.1, 0.07$ and -0.01 for green, red and blue lines respectively. The inset is the zoomed in regime for small $-\varepsilon_v$. 

3. Micromechanical results
with values ranging around 0 ± 0.1; in particular, it is dependent on the over-compression $\phi_i^{\text{max}}$ (data not shown). Unless strictly mentioned, we shall be using the values of $p_0$ and $\gamma_p$ given in Table 1.

Fig. 1(a) shows the behavior of $p$ with $\phi$ during one full over-compression cycle to display the dependence of the jamming point on the maximum over-compression volume fraction and the number of cycles. With increasing over-compression amplitude, e.g. comparing $\phi_i^{\text{max}} = 0.68$ and $\phi_i^{\text{max}} = 0.82$, the jamming point, as realized after unloading, is increasing. Also, with each cycle, from $M = 1$ to $M = 100$, the jamming point moves to larger values. Note that the difference between the loading and the unloading curves becomes smaller for subsequent over-compressions. Fig. 1(b) shows the scaled pressure, i.e., $p$ normalized by $\phi C/\phi_J$, which removes its non-linear behavior. $p$ represents the average deformation (overlap) of the particles at a given volume fraction, proportional to the distance from the jamming point $\phi_J$. In the small strain region, for all over-compression amplitude and cycles, the datasets collapse on a line with slope $p_0 \approx 0.042$.

Only for very strong over-compression, $-\varepsilon_v > 0.1$, a small deviation (from linear) of the simulation data is observed due to the tiny quadratic correction in Eq. (3.1).

### 3.1.2. Isotropic cyclic over-compression

Many different isotropic jamming points can be found in real systems and – as shown here – also for the simplest model material in 3D. Fig. 2(a) shows the evolution of these extracted isotropic jamming points $M \phi_{J,i}$, which increase with increasing $M$ and with over-compression $\phi_i^{\text{max}}$: for subsequent cycles $M$ of over-compressions, the jamming density $M \phi_{J,i}$ grows slower and slower and is best captured by a Kohlrausch-Williams-Watts (KWW) stretched exponential relation:

$$M \phi_{J,i} := \phi_J(\phi_i^{\text{max}}, M) = \infty \phi_{J,i} - (\infty \phi_{J,i} - \phi_c) \exp \left[ - (M/\mu_i)^{\beta_i} \right],$$

with the three universal “material”-constants $\phi_c = 0.6567$ (section 3.2.2), $\mu_i = 1$, and $\beta_i = 0.3$, the lower limit of possible $\phi_J$’s, the relaxation (cycle) scale and the stretched exponent parameters, respectively. Only $\infty \phi_{J,i}$, the equilibrium (steady-state or shakedown (García-Rojo et al. 2005)) jamming point limit (extrapolated for $M \to \infty$), depends on the over-compressions $\phi_i^{\text{max}}$.

Very little over-compression, $\phi_i^{\text{max}} \gtrsim \phi_c$, does not lead to a significant increase in $\phi_{J,i}$, giving us information about the lower limits of the isotropic jamming points achievable by shear, which is the critical jamming point $\phi_c = 0.6567$. With each over-compression cycle, $M \phi_{J,i}$ increases, but for large $M$ it increases less and less. This is analogous to compaction by tapping, where the tapped density increases logarithmically slow with the number of taps. The limit value $\infty \phi_{J,i}$ with $\phi_i^{\text{max}}$ can be fitted with a simple power law

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<tr>
<th>Quantity</th>
<th>Isotropic</th>
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<tr>
<td>$p$</td>
<td>$p_0 = 0.042; \gamma_p = 0 \pm 0.1^*$</td>
<td>$p_0 = 0.042; \gamma_p = 0 \pm 0.1^*$</td>
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<td>$C^*$</td>
<td>$C_1 = 8.5 \pm 0.3^*; \theta = 0.58$</td>
<td>$C_1 = 8.5 \pm 0.3^*; \theta = 0.58$</td>
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<td>$f_{NR}$</td>
<td>$\phi_c = 0.13; \phi_v = 15$</td>
<td>$\phi_c = 0.16; \phi_v = 15$</td>
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Table 1. Parameters used in Eqs. (5.1), (5.2) and (5.3), where ‘*’ represents slightly different values than from Imole et al. (2013), modified slightly to have more simple numbers, without big deviation, and without loss of generality.
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relation:

$$\infty \phi_{J,i} = \phi_c + \alpha_{\text{max}} \left( \frac{\phi_{i}^{\text{max}}}{\phi_c} - 1 \right)^{\beta},$$  \hspace{1cm} (3.3)

where the fit works perfect for $\phi_c < \phi_{i}^{\text{max}} \leq 0.9$, with parameters $\phi_c = 0.6567$, $\alpha_{\text{max}} = 0.02 \pm 2\%$, and $\beta = 0.3$, while the few points for $\phi_{i}^{\text{max}} \sim \phi_c$ are not well captured. The relation between the limit-value $\infty \phi_{J,i}$ and $\infty \phi_{J,i}$ is derived using Eq. (3.2):

$$\infty \phi_{J,i} - \phi_c = \frac{1}{1 - e^{-1}} \approx 1.58 \left( \infty \phi_{J,i} - \phi_c \right),$$  \hspace{1cm} (3.4)

only by setting $M = 1$, as shown in Fig. 2(b), with perfect match. With other words, using a single over-compression, Eq. (3.4) allows to predict the limit value after first over-compression $\infty \phi_{J,i}$ (or subsequent over-compression cycles, using appropriate $M$).

Thus, the isotropic jamming point $\phi_J$ is not a unique point, not even for frictionless particle systems, and is dependent on the previous deformation history of the system (Ciamarra et al. 2010; Möbius & Heussinger 2014; O’Hern et al. 2002), e.g. over-compression or tapping/driving (data not shown). Both (isotropic) modes of deformation lead to more compact, better packed configurations (Bi et al. 2011; Rosato et al. 2010; Zhang et al. 2010b). Considering different system sizes, and different preparation procedures, we confirmed that the jamming regime is the same (within fluctuations) for all the cases considered (not shown). All our data so far, for the material used, are consistent with a unique limit density $\phi_c$ that is reached after large strain, very slow shear, in the limit of vanishing confining pressure. Unfortunately this limit is vaguely defined, since it is not directly accessible, but rather corresponds to a virtual stress-free state. The limit density is hard to determine experimentally and numerically as well. Reason is that any slow deformation (e.g. compression from below jamming) also leads to perturbations (like tapping leads to granular temperature): the stronger the system is perturbed, the better it will pack, so that usually $\phi_J > \phi_c$ is established. Repeated perturbations, lead to a slow stretched exponential approach to an upper-limit jamming density $\phi_J \rightarrow \phi_J^{\text{max}}$ that itself increases slowly with perturbation amplitude, see Fig. 2(b). The observation of different $\phi_J$ of a single material, was referred to as $J$-segment (Ciamarra et al. 2010; O’Hern et al. 2002), and requires an alternative interpretation of the classical “jamming diagram” (Bi et al. 2011; Grob et al. 2014; Liu & Nagel 1998), giving up the misconception of a single, constant jamming “point”. Note that the J-segment is not just due to fluctuations, but it is due to the deformation history, and with fluctuations superposed. The state-variable $\phi_J$ varies due to deformation, but possibly has a unique limit value that we denote for now as $\phi_c$. Jammed states below $\phi_c$ might be possible too, but require different protocols (Hopkins et al. 2013), or different materials, and are thus not addressed here. The concept of shear jammed states (Bi et al. 2011) below $\phi_J$, is discussed next.

3.2. Shear deformation

To study shear jamming, we choose several unjammed states with volume fractions $\phi$ below their jamming points $\infty \phi_{J,i}$, which were established after the first compression-decompression cycle, for different history, i.e., various previously applied over-compression to $\phi_{i}^{\text{max}}$. Each configuration is first relaxed and then subjected to four isochoric (volume conserving) pure shear cycles ((see section 2.1)).

3.2.1. Shear jamming below $\phi_{J}(H)$

We confirm shear jamming, e.g., by a transition in the coordination number $C^*$, from below to above its isostatic limit, $C^*_3 = 6$, for frictionless grains (Imole et al. 2013; Peyneau & Roux 2008; Snoeijer et al. 2006; Wang et al. 2012). This was consistently
Figure 2. (a) Evolution of isotropic jamming points $\phi^{M}_{J,i}$ after performing $M$ isotropic compression-decompression cycles up to different maximum volume fractions $\phi^{\text{max}}_i$, as given in the inset. With increasing $\phi^{\text{max}}_i$, the range of the established jamming points $\phi^{M}_{J,i} = \phi_{J}(M, \phi^{\text{max}}_i)$ increases. The minimum (lower bound) of all $\phi^{M}_{J,i}$ is defined as the critical jamming limit point, $\phi_c = 0.6567$. The solid lines through the data are universal fits to a stretched exponential (Andreotti et al. 2013; Knight et al. 1995; Richard et al. 2005; Rosato et al. 2010) with only one single variable parameter $\phi^{\text{max}}_i$, i.e., the upper limit jamming point for $M \to \infty$, which depends on $\phi^{\text{max}}_i$. (b) The first jamming point $1_{J,i}$ (blue ■) and after many over-compression $\infty_{J,i}$ (brown ●) are plotted against over-compression amplitude $\phi^{\text{max}}_i$. Solid lines represent Eqs. (3.3) for $1_{J,i}$ and (3.4) for $\infty_{J,i}$. The shaded region is the exploratory range of jamming points $\phi^{M}_{J,i}$, denoted as $J$-segment. The red base line indicates the critical jamming point $\phi_c$.

Figure 3. (a) Snapshots of unjammed, fragile and shear jammed states, when the force networks are percolated in none, one or two, and all the three directions, respectively. Only the largest force network, connecting strong forces, $f^2 \geq k(f)$, with $k \geq 2.2$ are shown for the three states for clarity, and hence the white spaces in the background. (b) Plot of $C^*$ and cluster sizes $\xi/L$ in the three directions for tension in $x-$ and compression in $y-$ directions against the non-rattler fraction $f_{\text{NR}}$, along the loading path for an isotropic unjammed initial state with volume fraction $\phi = 0.6584$ and $\phi_{J}(\phi^{\text{max}}_i = 0.82, M = 1) =: 1_{J,i} = 0.6652$. The upward arrow indicates the direction of loading shear strain.
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(independently) reconfirmed by using percolation analysis (Bi et al. 2011; Radjai et al. 1998), allowing us to distinguish the three different regimes namely, unjammed, fragile and shear jammed states during (and after) shear (Grob et al. 2014), as shown in Fig. 3(a). For this, first we study the percolation analysis, that allows to distinguish the three regimes namely, unjammed, fragile and shear jammed states during (and after) shear, as shown in Fig. 3(a). We study how the $k$–cluster, defined as the largest force network, connecting strong forces, $f \geq kf_{av}$ (Hidalgo et al. 2002; Smith et al. 2011), with $k = 2.2$, different from $k = 1$ for 2D frictional systems (Bi et al. 2011), percolates when the initially unjammed isotropic system is sheared. More quantitatively, for an exemplary volume fraction $\phi (\phi_{l}^{\text{max}} = 0.82, M = 1) = 0.6584$, very close to $\phi_c$, Fig. 3(b) shows that $f_{NR}$ increases from initially zero to large values well below unity due to the always existing rattlers. The compressive direction percolating network $\xi_y/L_y$ grows faster than the tension direction network $\xi_x/L_x$, while the network in the non-mobile direction, $\xi_z/L_z$, lies in between them. For $f_{NR} > 0.82 \pm 0.01$, we observe that the growing force network is percolated in all three directions (Fig. 3(a)), which is astonishingly similar to the value reported for the 2D systems (Bi et al. 2011). The jamming by shear of the material corresponds (independently) to the crossing of $C^*$ from the isostatic limit of $C_0^* = 6$, as presented in Fig. 3(b).

From this perspective, when an unjammed material is sheared at constant volume, and it jams after application of sufficient shear strain, clearly showing that the jamming point has moved to a lower value. Shearing the system also perturbs it, just like over-compression; however, in addition, finite shear strains enforce shape- and structure-changes and thus allow the system to explore new configurations; typically, the elevated jamming density $\phi_J$ of a previously compacted system will rapidly decrease and exponentially approach its lower-limit, the critical jamming point $\phi_c$, below which no shear jamming exists. Note that we do not exclude the possibility that jammed states below $\phi_c$ could be achieved by other, special, careful preparation procedures (Atkinson et al. 2014).

Next, we present the evolution of the strong force networks in each direction during cyclic shear, as shown in Fig. 4, for the same initial system. After the first loading, at reversal $f_{NR}$ drops below the 0.82 threshold, which indicates the breakage/disappearance of strong clusters, i.e. the system unjams. The new tension direction $\xi_y/L_y$ drops first with the network in the non-mobile directions, $\xi_z/L_z$, lying again in between the two mobile direction. With further applied strains, $f_{NR}$ increases and again, the cluster associated with the compression direction grows faster than in the tension direction. For $f_{NR}$ above the threshold, the cluster percolates the full system, leading to shear jammed states again. At each reversal, the strong force network breaks/fails in all directions, and the system gets “soft” or even unjams temporarily. However, the network is rapidly re-established in the perpendicular direction, i.e., the system jams and the strong, anisotropic force network again sustains the load. It is important to note that for some systems with volume fractions away from $\phi_c$ can resist shear strain reversal (data not shown).

3.2.2. Relaxation effects on shear jammed states

Here, we will discuss the system stability by looking at the macroscopic quantities in the saturation state (after large shear strain), by relaxing them sufficiently long to have non-fluctuating values in the microscopic and macroscopic quantities. Every shear cycle after defining e.g. the $y$--direction as the initial active loading direction, has two saturation states, one during loading and, after reversal, the other during unloading. In Fig. 5, we show values attained by the isotropic quantities pressure $p$, isotropic fabric $F_v$ and the deviatoric quantities shear stress $\tau$, shear stress ratio $\tau/p$, and deviatoric fabric
Figure 4. Cluster sizes, $f_{NR}$ (top panel), over three strain cycles bottom for $\phi = 0.6584$ and jamming point $\phi_J (\phi_{\text{max}}^{i} = 0.82, M = 1) =: ^{1}\phi_{J,i} = 0.6652$. Dashed horizontal black line represents transition from unjammed to shear jammed states. The cluster sizes are smoothed averages over two past and future snapshots.

$F_d$ for various $\phi$ given the same initial jamming point $\phi_J (\phi_{\text{max}}^{i} = 0.82, M = 1) =: ^{1}\phi_{J,i} = 0.6652$. Data are shown during cyclic shear as well as at the two relaxed saturation states (averaged over four cycles), leading to following observations:

(i) With increasing volume fraction, $p$, $F_v$ and $\tau$ increase, while a weak decreasing trend in stress ratio $\tau/p$ and deviatoric fabric $F_d$ is observed.

(ii) There is almost no difference in the relaxed states in isotropic quantities, $p$ and $F_v$ for the two directions, whereas it is symmetric about zero for deviatoric quantities, $\tau$, $\tau/p$, and $F_d$. The decrease in pressure during relaxation is associated with dissipation of kinetic energy and partial opening of the contacts to “dissipate” the related part of the contact potential energy. However, $F_v$ remains at its peak value during relaxation, showing that the contact structure is almost unchanged and the network remains stable during relaxation.

(iii) For small volume fractions, close to $\phi_c$, the system becomes strongly anisotropic in stress ratio $\tau/p$, and fabric $F_d$ rather quickly, during (slow) shear (envelope for low volume fractions in Figs. 5(d) and 5(e)), before it reaches the steady state (Walker et al. 2014).

(iv) It is easy to obtain the shear jamming point $\phi_c$ from the relaxed critical (steady) state pressure $p$, and shear stress $\tau$, by extrapolation to zero, as the envelope of relaxed data in Figs. 5(a) and 5(c).

We use the same methodology using Eq. (3.1), to extract the shear jamming point $\phi_c$. When the relaxed $p$ is normalized with the contact density $\phi C$, we obtain $\phi_c = 0.6567 \pm 0.0005$ by linear extrapolation. A similar value of $\phi_c$ is obtained from the ex-
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Figure 5. Scatter plots of isotropic quantities (a) pressure \( p \), (b) isotropic fabric \( F_v \) and deviatoric quantities (c) shear stress \( \tau \), (d) shear stress ratio \( \tau/p \), and (e) deviatoric fabric \( F_d \) for various \( \phi \) and jamming point \( \phi_J(\phi_i^{\max} = 0.82, M = 1) =: 1\phi_{J,i} = 0.6652 \). Black ‘x’ symbols represent the initial loading cycle, while the green ‘+’ and blue ‘*’ represent states attained for \( \phi < \phi_J \) and \( \phi > \phi_J \), respectively for the subsequent shear. Cyan ‘*’ and the brown ‘■’ are states chosen after large strain during loading and unloading shear respectively, and are relaxed. The red and purple lines indicate the critical jamming point \( \phi_c = 0.6567 \) and the jamming point \( 1\phi_{J,i} \) respectively.

3.3. Jamming phase diagram with history \( H \)

We propose a jamming phase diagram with shear strain, and present a new, quantitative history dependent model that explains jamming and shear jamming, but also predicts that shear jamming vanishes under some conditions, namely when the system is not tapped, tempered or over-compressed before shear is applied. Using \( \varepsilon_d \) and \( \phi \) as parameters, Fig. 6(a) shows that for one initial the history dependent jamming state at \( 1\phi_{J,i} \), there exist sheared states within the range \( \phi_i \leq \phi \leq \phi_J(\phi_i) \), which are isotropically unjammed. After small shear strain they become fragile, and for larger shear strain jam and remain jammed, i.e., eventually showing the critical state flow regime, where pressure, shear stress ratio and structural anisotropy have reached their saturation levels and forgotten their initial state (data not shown). The transition to fragile states is accompanied by partial percolation of the strong force network, while percolation in all directions indicates the shear jamming transition. Above jamming, the large fraction of non-rattlers provides a persistent mechanical stability to the structure, even after shear is stopped.

For \( \phi \) approaching \( \phi_c \), the required shear strain to jam \( \varepsilon_d^{SJ} \) increases, i.e., there exists a divergence “point” \( \phi_c \), where ‘infinite’ shear strain might jam the system, but below...
which no shear jamming was observed. The closer the (constant) volume fraction \( \phi \) is to the initial \( \phi_{ji} \), the smaller is \( \varepsilon_d^{SJ} \). States with \( \phi \geq \phi_{ji} \) are isotropically jammed already before shear is applied.

Based on the study of many systems, prepared via isotropic over-compression to a wide range of volume fractions \( \phi_{max} \geq \phi_c \), and subsequent shear deformation, Fig. 6(b) shows the strains required to jam these states by applying pure shear. A striking observation is that independent of the isotropic jamming point \( \phi_{ji} \), all curves approach a unique shear jamming point at \( \phi_c \sim 0.6567 \) (see section 3.2.2). When all the curves are scaled with their original isotropic jamming point \( \phi_{ji} \) as \( \varepsilon_d = (\phi - \phi_c) / (M \phi_{ji} - \phi_c) \) they collapse on a unique master curve

\[
\left( \frac{\varepsilon_d^{SJ}}{\varepsilon_d^0} \right)^\alpha = - \log \phi_{sc} = - \log \left( \frac{\phi - \phi_c}{M \phi_{ji} - \phi_c} \right),
\] (3.5)

shown in the inset of Fig. 6(b), with power \( \alpha = 1.37 \pm 0.01 \) and shear strain scale \( \varepsilon_d^0 = 0.102 \pm 0.001 \) as the fit parameters. Hence, if the initial jamming point \( M \phi_{ji} \) or \( \phi_{ji}(H) \) is known based on the past history of the sample, the shear jamming strain \( \varepsilon_d^{SJ} \) can be predicted.

From the measured shear jamming strain, Eq. (3.5), knowing the initial and the limit value of \( \phi_J \), we now postulate its evolution under isochoric pure shear strain:

\[
\phi_J(\varepsilon_d) = \phi_c + (\phi - \phi_c) \exp \left[ \left( \frac{\varepsilon_d^{SJ}}{\varepsilon_d^0} \right)^\alpha - \left( \frac{\varepsilon_d^0}{\varepsilon_d^0} \right)^\alpha \right].
\] (3.6)

Inserting, \( \varepsilon_d = 0 \), \( \varepsilon_d = \varepsilon_d^{SJ} \) and \( \varepsilon_d = \infty \) leads to \( \phi_J = M \phi_{ji} \), \( \phi_J = \phi \) and \( \phi_J = \phi_c \), respectively. This means the jamming point evolution due to shear strain \( \varepsilon_d \) is faster than exponential (since \( \alpha > 1 \)) decreasing to its lower limit \( \phi_c \). This is qualitatively different from the stretched exponential (slow) relaxation dynamics that leads to the increase of \( \phi_J \) due to over-compression or tapping, see Fig. 7(a) for both cases.
4. Mesoscale stochastic model for slow dynamics

The last challenge is to unify the observations in a model that accounts for the changes in the jamming densities for both isotropic and shear deformation modes. Over-compressing a soft granular assembly is analogous to tapping (Coulais et al. 2014; Rosato et al. 2010; Zhang et al. 2010b) more rigid ones, in so far that both methods lead to more compact packing structures, i.e., both represent isotropic perturbations. These changes are shown in Fig. 2(a), where the originally reported logarithmically slow dynamics for tapping (Andreotti et al. 2013; Knight et al. 1995) is very similar to our results that are also very slow, with a stretched exponential behavior; such slow relaxation dynamics can be explained by a simple Sinai-Diusion model of random walkers in a random, hierarchical, fractal-type free energy landscape (Luding et al. 2000; Richard et al. 2005) in the a-thermal limit, where the landscape does not change – for the sake of simplicity. The granular packing is represented in this picture by an ensemble of random walkers in (arbitrary) configuration space with (potential) energy according to the height of their position on the landscape. (Their average energy corresponds to the jamming density and a decrease in energy corresponds to an increase in $\phi_J(H)$, thus representing the “memory” and history dependence.) Perturbations, such as tapping with some amplitude (corresponding to “temperature”) allow the ensemble to find denser configurations, i.e., deeper valleys in the landscape, representing larger (jamming) densities (Mobius & Heussinger 2014; Reichhardt & Reichhardt 2014). Similarly, over-compression is squeezing the ensemble “down-hill”, also leading to an increase of $\phi_J$, as presented in Fig. 7(b). Larger amplitudes will allow the ensemble to overcome larger barriers and thus find even deeper valleys. Repetitions have a smaller chance to do so, which explains the slow dynamics in the hierarchical multiscale structure of the energy landscape.

In contrast to the isotropic perturbations, where the random walkers follow the “down-hill” trend, shear is anisotropic and thus pushing parts of the system “up-hill”. For example, under planar simple shear, one (eigen) direction is tensile (up) whereas another is compressive (down). If the ensemble is random, shear will only re-shuffle the population. But if the material was previously forced or relaxed towards the (local) landscape minima, shear can only lead to a net up-hill drift of the ensemble, i.e., to decreasing $\phi_J$, referred to as dilatancy. For ongoing perturbation, if volume is conserved, both coordination number and pressure slowly decrease (data not shown) whereas for fixed confining pressure the volume would decrease (compactancy). This process is much faster than relaxation, since it is driven by shear strain amplitude. For large enough strain the system will be sufficiently re-shuffled, randomized, or “re-juvenated” such that it can be close to its quenched, random state $\phi_c$.

5. Macroscopic constitutive model

In this section, we present the simplest model equations, as used for the predictions, involving a history dependent $\phi_J(H)$, as given by Eq. (3.2) for isotropic deformations and Eq. (3.6) for shear deformations. The only difference to Imole et al. (2013), where these relations are taken from, based on purely isotropic unloading, is a variable $\phi_J = \phi_J(H)$. 

Figure 7. Relaxation mechanisms and dynamics in an energy landscape due to memory effects. (a) Schematic evolution of the jamming points $\phi_J(H)$ due to history $H$. Solid lines represent many isotropic compression decompression cycles for three different $\phi_j^{\text{max}}$, leading to an increase in $\phi_J(H)$ by slow stretched exponential relaxation, see Eq. (3.2). Dashed lines represent the much faster decrease in $\phi_J(H)$ due to shear strain $\varepsilon_d$, using Eq. (3.6). (b) The sketch represents only a very small, exemplary part of the hierarchical, fractal-type energy landscape. The red horizontal line represents the (quenched) average, while the dotted horizontal line indicates the momentary average $\phi_J(H)$ (of the ensemble of states, where the population is represented by green circles). The blue solid arrows show (slow) relaxation due to perturbations, while the dashed arrows indicate (fast) re-arrangements (re-juvenation) due to finite shear strain. The green dots represent with their size the population after some relaxation, in contrast to a random, quenched population where all similar valleys would be equally populated (Xu et al. 2011).

5.1. Presentation and model calibration

5.1.1. During cyclic isotropic deformation

During (cyclic) isotropic deformation, the evolution equation for the corrected coordination number $C^*$ is:

$$C^* = C_0 + C_1 \left( \frac{\phi}{\phi_J(H)} - 1 \right)^\theta,$$

with $C_0 = 6$ for the frictionless case and parameters $C_1$ and $\theta$ are presented in Table 1. The fraction of non-rattlers $f_{NR}$ is given as:

$$f_{NR} = 1 - \varphi_c \exp \left[ -\varphi_v \left( \frac{\phi}{\phi_J(H)} - 1 \right) \right],$$

with parameters $\varphi_c$ and $\varphi_v$ presented in Table 1. We modify Eq. (3.1) for the evolution of $p$ together with the history dependent $\phi_J = \phi_J(H)$ so that,

$$p = \frac{\phi C}{\phi_J(H)} p_0 (-\varepsilon_v) [1 - \gamma_p (-\varepsilon_v)],$$

with parameters $p_0$ and $\gamma_p$ presented in Table 1, and the true or logarithmic volume change of the system is $-\varepsilon_v = \log(\phi/\phi_J(H))$, relative to the momentary jamming density. The non-corrected coordination number is $C = C^* f_{NR}$, as can be computed using Eqs. (5.1) and (5.2). The isotropic fabric $F_v$ is given by the relation $F_v = g_3 \phi C$, as taken from Imole et al. (2013), with $g_3 = 1.22$ for the polydispersity used in the present work. Also the parameters $C_1$, $\theta$ for $C^*$, $\varphi_c$, $\varphi_v$ for $f_{NR}$; and $p_0$, $\gamma_p$ for pressure $p$ are similar to Imole et al. (2013), with the second order correction parameter $\gamma_p$ most sensitive to
the details of previous deformations; however, not being very relevant since it is always a small correction due to the product $\gamma_p(-\varepsilon_d)$.

The above relations are used to predict the behavior of the isotropic quantities: dimensionless pressure $p$ and coordination number $C^*$, as shown in Fig. 9(a-b) during isotropic compression, as well as for the fraction of non-rattlers in Fig. 9(c) for cyclic shear, with corresponding parameters presented in Table 1. Note that during isotropic deformation, $\phi_J(H)$ was changed only during the compression branch, using Eq. (3.2) for fixed $M = 1$ using $\phi_J^\text{max}$ as variable, but is kept constant during unloading/expansion.

The above relations are used to predict the behavior of the isotropic quantities: dimensionless pressure $p$ and coordination number $C^*$, by only adding the history dependent jamming point $\phi_J(H)$ to constitutive model, as presented in section 5.2.

5.1.2. Cyclic (pure) shear deformation

During cyclic (pure) shear deformation, a simplified equation for the shear stress ratio $\tau/p$ is taken from Imole et al. (2013) as:

$$\tau/p = (\tau/p)^\text{max} - \left((\tau/p)^\text{max} - (\tau/p)^0\right) \exp[-\beta_s\varepsilon_d], \quad (5.4)$$

with $(\tau/p)^0$ and $(\tau/p)^\text{max}$ the initial and maximum (saturation) shear stress ratio, respectively, and $\beta_s$ its growth rate. Similarly, a simplified equation for the deviatoric fabric $F_d$ can be taken from Imole et al. (2013) as:

$$F_d = F_d^\text{max} - [F_d^\text{max} - F_d^0] \exp[-\beta_F\varepsilon_d], \quad (5.5)$$

with $F_d^0$ and $F_d^\text{max}$ the initial and maximum (saturation) values of the deviatoric fabric, respectively, and $\beta_F$ its growth rate. The four parameters $(\tau/p)^\text{max}$, $\beta_s$ for $\tau/p$ and $F_d^\text{max}$, $\beta_F$ for $F_d$ are dependent on the volume fraction $\phi$ and are well described by the general relation from Imole et al. (2013) as:

$$Q = Q_a + Q_c \exp \left[-\Psi \left(\frac{\phi}{\phi_J(H)} - 1\right)\right], \quad (5.6)$$

where $Q_a$, $Q_c$ and $\Psi$ are the fitting constants with values presented in Table 2.

For predictions during cyclic shear deformation, $\phi_J(H)$ was changed with applied shear strain $\varepsilon_d$ using Eq. (3.6). Furthermore, the jamming point is set to a larger value just after strain-reversal which is discussed next.

<table>
<thead>
<tr>
<th>Evolution parameters</th>
<th>$Q_a$</th>
<th>$Q_c$</th>
<th>$\Psi$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$(\tau/p)^\text{max}$</td>
<td>0.12</td>
<td>0.091</td>
<td>7.9</td>
</tr>
<tr>
<td>$\beta_s$</td>
<td>30</td>
<td>40</td>
<td>16</td>
</tr>
<tr>
<td>$F_d^\text{max}$</td>
<td>0</td>
<td>0.17</td>
<td>5.3</td>
</tr>
<tr>
<td>$\beta_F$</td>
<td>0</td>
<td>40</td>
<td>5.3</td>
</tr>
</tbody>
</table>

Table 2. Parameters for Eqs. (5.4) and (5.5) using Eq. (5.6), with slightly different values than from Imole et al. (2013), that are extracted using the similar procedure as in Imole et al. (2013), for states with volume fraction close to the jamming volume fraction.
Figure 8. Phase diagram showing the minimum reversal shear strain \( \varepsilon_{d,J,R}^{SJ,R} \) needed to jam the states below \( \phi_{cr} \), for states prepared from the first over-compression cycle with different \( \phi_i^{\text{max}} \), as given in the legend. The inset shows a collapse of the states using a similar scaled definition as Eq. (3.5) that includes the distance from both \( \phi_{cr} \) and critical jamming point \( \phi_c \), using Eq. (5.7).

5.1.3. Behavior of jamming point at strain reversal

As mentioned in section 3.2, there are some states below \( \phi_J \), where application of shear strain jams the systems. The densest of those can resist shear reversal, but below a certain \( \phi_{cr} \approx 0.662 < \phi_J \), shear reversal unjams the system again. With this information, we postulate the following:

(i) After the first large strain shear, the system should forget where it was isotropically compressed to before i.e., \( M_{\phi,J,1} \) is forgotten and \( \phi_J = \phi_c \) is realized.

(ii) There exists a volume fraction \( \phi_{cr} \), above which systems can just resist shear reversal and remain always jammed in both forward and reverse shear.

(iii) Below this \( \phi_{cr} \), reversal unjams the system. Therefore, more strain is needed to jam the system (when compared to the initial loading), first to forget its state before reversal, and then to re-jam it in opposite (perpendicular) direction. Hence, the strain necessary to jam in reversal direction should be higher than for the first shear cycle.

(iv) As we approach \( \phi_c \), the reverse strain needed to jam the system increases.

We use these ideas and measure the reversal shear strain \( \varepsilon_{d,J,R}^{SJ,R} \), needed to re-jam the states below \( \phi_{cr} \), as shown in Fig. 8. When they are scaled with \( \phi_{cr} \) as \( \phi_{sc} = (\phi - \phi_c) / (\phi_{cr} - \phi_c) \), they collapse on a unique master curve, very similar to Eq. (3.5):

\[
\left( \frac{\varepsilon_{d,J,R}^{\phi_{cr}}}{\varepsilon_{d,J,R}^{\phi_{cr}}} \right)^\alpha = -\log \phi_{sc} = -\log \left( \frac{\phi - \phi_c}{\phi_{cr} - \phi_c} \right),
\]

shown in the inset of Fig. 6(b), with the same power \( \alpha = 1.37 \pm 0.01 \) as Eq. (3.5). Fit parameter strain scale \( \varepsilon_{d,J,R}^{\phi_{cr}} = 0.17 \pm 0.002 > \varepsilon_d^0 = 0.102 \), is consistent with the above postulates (iii) and (iv).

The above relations are used to predict the isotropic and the deviatoric quantities, during cyclic shear deformation, as described next, with the additional rule that all the quantities attain value zero for \( \phi \leq \phi_J(H) \). Moreover, for any state with \( \phi \leq \phi_{cr} \), shear
Figure 9. Model prediction of cyclic loading: (a) Dimensionless pressure $p$ and (b) coordination number $C^*$ plotted against volume fraction $\phi$ for an isotropic compression starting from $\phi_i = 0.64$ to $\phi_i^{\text{max}} = 0.73$ (small symbols) and decompression (big symbols) back to $\phi_i$, with $\phi_{J;i} = 0.667$, for $M = 1$ (red ‘•’) and for $M = 300$ (blue ‘■’). (c) Deviatoric stress ratio $\tau/p$ and deviatoric fabric $F_d$, fraction of non-rattlers $f_{NR}$, coordination number $C^*$, pressure $p$ and history dependent jamming point $\phi_J(H)$ over three pure shear strain cycles (bottom panel) for $\phi = 0.6584$ and initial jamming point $\phi_J(\phi_{\text{max}} = 0.82, M = 1) = 0.6652$. Solid lines through the data are the model prediction, involving the history dependent jamming point $\phi_J(H)$, using Eq. (3.2) for isotropic deformation and Eq. (3.6) for shear deformation, and others. Dashed red lines in $f_{NR}$ and $C^*$ represent transition from unjammed to shear jammed states, whereas in $\phi_J(H)$ the red line indicates the critical jamming point $\phi_c$.

strain reversal moves the jamming point to $\phi_{cr}$, and the movement of jamming point follows Eq. (5.7).

Any other deformation mode, can be written of as a unique superposition of pure isotropic and pure shear deformation modes, and hence the combination of the above can be easily used to describe any general deformation, e.g. uniaxial cyclic compression (data not presented).

5.2. Prediction: minimal model

Finally, we test the proposed history dependent jamming point $\phi_J(H)$ model, by predicting $p$ and $C^*$, when a granular assembly is subjected to cyclic isotropic compression
to $\phi^\text{max}_i = 0.73$ for $M = 1$ and for $M = 300$ cycles, with $\sim \phi^\text{max}_i = 0.667$, as shown in Fig. 9(a-b). It is observed that using the history dependence of $\phi^\text{max}(H)$, the hysteretic behavior of the isotropic quantities, $p$ and $C^*$, is very well predicted, qualitatively similar to isotropic compression and decompression of real 2D frictional granular assemblies, as shown in by Bandi et al. (2013) and Reichhardt & Reichhardt (2014).

In Fig. 9(c), we show the evolution of the deviatoric quantities shear stress ratio $\tau/p$ and deviatoric fabric $F_d$, when a system with $\phi = 0.6584$, close to $\phi_c$, and initial jamming point $\phi(H)(0) = 0.6652$, is subjected to three shear cycles (lowest panel). The shear stress ratio $\tau/p$ is initially undefined, but soon establishes a maximum (not shown) and decays to its saturation level at large strain. After strain reversal, $\tau/p$ drops suddenly and attains the same saturation value, for each half-cycle, only with alternating sign. The behavior of the anisotropic fabric $F_d$ is similar to that of $\tau/p$. During the first loading cycle, the system is unjammed for some strain, and hence $F_d$ is zero in the model (observations in simulations can be non-zero, when the data correspond to only few contacts, mostly coming from rattlers). However, the growth/decay rate and the saturation values attained are different from those of $\tau/p$, implying a different, independent stress- and structure-evolution with strain – which is at the basis of recently proposed anisotropic constitutive models for quasi-static granular flow under various deformation modes (Imole et al. 2013). The simple model with $\phi^\text{max}(H)$, is able to predict quantitatively the behavior the $\tau/p$ and $F_d$ after the first loading path, and is qualitatively close to the cyclic shear behavior of real 2D frictional granular assemblies, as shown in Supplementary Fig. 7 by Bi et al. (2011).

At the same time, also the isotropic quantities are very well predicted by the model, using the simple equations from section 5.1, where only the jamming point is varying with shear strain, while all material parameters are kept constant. Some arbitrariness involves the sudden changes of $\phi^\text{max}$ at reversal, as discussed in section 5.1. Therefore, using a history dependent $\phi^\text{max}(H)$ gives hope to understand the hysteretic observations from realistic granular assemblies, and also provides a simple explanation of shear jamming. Modifications of continuum models like anisotropic models (Imole et al. 2013; Kumar et al. 2014b), or GSH type models (Jiang & Liu 2008, 2015), by including a variable $\phi^\text{max}$, can this way quantitatively explain various mechanisms around jamming.

6. Towards experimental validation

The purpose of this section is two-fold: First, we propose ways to (indirectly) measure the jamming density, since it is a virtual quantity that is hard to measure directly, just as the “virtual, stress-free reference state” in continuum mechanics which it resembles. Second, this way, we will introduce alternative state-variables, since by no means is the jamming density the only possibility.

Measuring $\phi^\text{max}$ from experiments

Here we show the procedure to extract the history dependent jamming point $\phi^\text{max}(H)$ from measurable quantities, indirectly obtained via Eqs. (5.1), (5.2), (5.3), and directly from Eq. (3.6). There are two reasons to do so: (i) the jamming point $\phi^\text{max}(H)$ is only accessible in the unloading limit $p \to 0$, which requires an experiment or a simulation to “measure” it (however, during this measurement, it might change again); (ii) deducing the jamming point from other quantities that are defined for an instantaneous snapshot/configuration for $p > 0$ allows to indirectly obtain it – if, as shown next, these indirect “measurements” are compatible/consistent: Showing the equivalence of all the different $\phi^\text{max}(H)$, proofs the consistency and completeness of the model and, even more important, provides a way to obtain $\phi^\text{max}(H)$ indirectly from experimentally accessible quantities.
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Figure 10. (a) Evolution of the history dependent jamming point $\phi_J(H)$ during isotropic over-compression to $\phi^i_{\text{max}} = 0.82$ for two cycles, calculated back from the measured quantities: coordination number $C^*$ (green) and pressure $p$ (red), using Eqs. (5.1) and (5.3) respectively. The ‘.’ and ‘■’ represent the first and second cycle respectively. Solid lines are the loading path while the dashed lines represent the unloading path for the corresponding cycle. Evolution of history dependent jamming point $\phi_J(H)$ using (b) coordination number $C^*$ and (c) pressure $p$ for three levels of over-compression $\phi^i_{\text{max}}$, as shown in the inset. Solid black line represents Eq. (3.2) with $M = 1$, and $\phi_{J,i}$ calculated using Eq. (3.3).

For isotropic compression

Fig. 10 shows the evolution of $\phi_J(H)$, measured from the two experimentally accessible quantities: coordination number $C^*$ and pressure $p$, using Eqs. (5.1) and (5.3) respectively for isotropic over-compression to $\phi^i_{\text{max}} = 0.82$ over two cycles. Following observations can be made: (i) $\phi_J$ for isotropic loading and unloading can be extracted from $C^*$ and $p$; (ii) it rapidly increases and then saturates during loading.; (iii) it mimics the fractal energy landscape model in Fig. 4 from Luding et al. (2000) very well; (iv) while it was assumed not to change for unloading, it even increases, which we attribute to the perturbations and fluctuations (granular temperature) induced during the quasi-static deformations; (v) the indirect $\phi_J$ are reproducible and follow the same master-curve for first over-compression as seen in Figs. 10, independent of the maximum – all following deformation is dependent on the previous maximum density.

For shear deformation

Fig. 11 shows the evolution of $\phi_J(H)$, measured from the two experimentally accessible quantities: coordination number $C^*$ and pressure $p$, using Eqs. (5.1) and (5.3) respectively during volume conserving shear with $\phi = 0.66$, and the initial jamming point $\phi_J(\phi^i_{\text{max}} = 0.82, M = 1) = \phi_{J,i} = 0.6652 > \phi$ and shows good agreement with the theoretical predictions using Eq. (3.6) after shear jamming. Thus the indirect measurements
Figure 11. Evolution of the history dependent jamming point $\phi_J(H)$ during pure shear, calculated back from the measured quantities: coordination number $C^*$, fraction of non-rattlers $f_{NR}$ and pressure $p$, using Eqs. (5.1), (5.2), (5.3) respectively, as marked with arrows. The volume fraction is constant, $\phi = 0.66$, and the initial jamming point $\phi_{J}(\phi_{J}^{\text{max}} = 0.82, M = 1) =: \phi_{J,i} = 0.6652$ is greater than $\phi$ (represented by horizontal cyan line). The solid black line represents Eq. (3.6), and the dashed vertical line indicates the shear strain needed to jam the system, $\varepsilon_{\delta J}^J$, from which on – for larger shear strain – the system is jammed.

of $\phi_J(H)$ can be applied if $\phi_J(H) < \phi$; the result deduced from pressure fits the best, i.e., it interpolates the two others and is smoother.

7. Summary, Discussion and Outlook

In summary, this study presents a quantitative, predictive macroscopic model that unifies a variety of phenomena at and around jamming, for quasi-static deformation modes. The most important ingredient is a scalar state-variable that responds very slow to (isotropic, perturbative) deformation, whereas it responds exponentially fast to finite shear deformation. This different response to fundamentally different modes of deformation (isotropic or shear) is (qualitatively) explained by a stochastic (mesoscale) model with fractal (multiscale) character. Discussing the equivalence of alternative state-variables and ways to experimentally measure the model parameters and apply it to other, more realistic materials, concludes the study.

7.1. Some questions answered

The questions posed in the introduction can now be answered: (i) The transition between the jammed and flowing (unjammed) regimes is controlled by a single new, isotropic, history dependent state-variable, the jamming density $\phi_J(H)$, with history $H$, which (ii) has a unique lower critical jamming density $\phi_c$; so that (iii) the history (protocol dependence) of jamming is completely encompassed by this new state-variable; and (iv) jamming, unjamming and shear jamming can all occur in 3D without any friction, only by reorganizations of the micro-structure.

7.2. Lower limit of jamming

The multiscale model framework implies now a minimum $\phi_c$ that represents the (critical) steady state for a given sample in the limit of vanishing confining stress, i.e., the lower limit of all jamming points. This is nothing but the lowest stable random density a
sheared system “locally” can reach due to continuously ongoing shear, for vanishing confining stress.

This lower limit is difficult to access in experiments and simulations, since every shear also perturbs the system leading at the same time to (slow) relaxation and thus a competing increase in \( \phi_J(H) \). However, it can be obtained from the (relaxed) steady state values of pressure, extrapolated to zero, i.e., from the envelope of pressure in Fig. 5. Note that special other deformation modes or careful preparation procedures e.g., energy minimization techniques (O’Hern et al. 2003; Torquato & Stillinger 2010) may lead to jammed states at even lower density than \( \phi_c \), from which shear would lead to an increase of the jamming point (a mechanism which we could not clearly identify from our frictionless simulations due to very long relaxation times near jamming for soft particles). This suggests future studies in the presence of friction so that one has wider range of jamming densities and lower density states might be much more stable as compared to the frictionless systems.

7.3. Shear jamming as consequence

Given an extremely simple model picture, starting from an isotropically unjammed system, shear jamming is not anymore a new effect, but is just due to the shift of the state-variable (jamming density) to lower values during shear. In other words, shear jamming occurs when the variable \( \phi_J(H) \) drops below the density \( \phi \) of the system.

Even though dilatancy is that what is typically expected under shear (of a consolidated packing), also compactancy is observed in some cases (Imole et al. 2013) and can be explained by our model. Given a certain preparation protocol, typically a jamming density \( \phi_J > \phi_c \) will be reached for a sample, since the critical limit \( \phi_c \) is very difficult to reach. When next a shear deformation is applied, it depends e.g., on the strain rate whether dilatancy or compaction will be observed: if the shear mode is “slower” than the preparation, or if \( \phi_J > \phi_c \), dilatancy is expected as a consequence of the rapidly decreasing \( \phi_J \) of the sample. In contrast, for a relatively “fast”, violent shear test (relative to the preparation procedure), compactancy also can be the result, due to an increase of \( \phi_J \) during shear.

7.4. Towards experimental validation

The history dependent jamming point \( \phi_J(H) \) is difficult to access directly, but can consistently be extracted from other, experimentally measurable quantities, e.g., pressure \( p \), coordination number \( C^* \) or fraction of non-rattlers \( f_{NR} \). We explain the methodology to extract \( \phi_J(H) \) experimentally, and confirm by indirect measurement, as detailed in section 6, that the jamming density is indeed increasing during isotropic deformation and decreasing during shear.

With other words, we do not claim that the jamming density is the only choice for the new state-variable that is needed. It can be replaced by other quantities as, e.g., the isotropic fabric, or the fraction of non-rattlers, or an empirical stress-free state that is extrapolated from pressure (which can be measured most easily).

7.5. Outlook

Experiments should be performed to calibrate our model for suspended soft spheres and real, frictional materials (Brodu et al. 2015; Brujić et al. 2007; Yu et al. 2014). Over-compression is possible for soft materials, but not expected to lead to considerable relaxation due to the small possible compressive strain for harder materials. However, tapping or small-amplitude shear can take the role of over-compression, also leading to
perturbations and increasing \( \phi_J \); in contrast, large-amplitude shear is decreasing \( \phi_J \) and can be calibrated indirectly from different isotropic quantities.

The accessible range of \( \phi_J - \phi_c \) is expected to much increase for more realistic systems, e.g., with friction, for non-spherical particle shapes or for cohesive powders.

The model presented implies that a superposition of the different deformation modes is possible or, with other words, that the responses are mostly decoupled. Even though this decoupling is consistent with most of our data (the responses to isotropic and deviatoric deformations are mostly independent and can be measured independently), this separation and superposition cannot be taken for granted for realistic granular and powder systems.

From the theoretical side, a measurement of the multiscale energy landscape, e.g. the valley width, depth/shapes and their probabilities (Xu et al. 2011) should be done to verify our model-picture, as it remains qualitative so far. Finally, applying our model to glassy dynamics, ageing and rejuvenation, and frequency dependent responses, encompassing also stretched exponential relaxation, see e.g. Lieou & Langer (2012), is another open challenge for future research. All this involves the temperature as a source of perturbations that affect the jamming point, and will thus also allow to understand more dynamic granular systems where the granular temperature is finite and not negligible as implied in most of this study.

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REFERENCES


Memory of jamming – multiscale flow in soft and granular matter


