Master equation for the probability distribution functions of forces in soft particle packings† Kuniyasu Saitoh,* Vanessa Magnanimo, and Stefan Luding

Employing molecular dynamics simulations of jammed soft particles, we study microscopic responses of force-chain networks to quasi-static isotropic (de)compressions. We show that not only contacts but also interparticle gaps between the nearest neighbors must be considered for the stochastic evolution of the probability distribution functions (PDFs) of forces, where the mutual exchange of contacts and interparticle gaps, i.e. opening and closing contacts, are also crucial to the incremental system behaviors. By numerically determining the transition rates for all changes of contacts and gaps, we formulate a Master equation for the PDFs of forces, where the insight one gets from the transition rates is striking: The mean change of forces reflects non-affine system response, while their fluctuations obey uncorrelated Gaussian statistics. In contrast, interparticle gaps are reacting mostly affine in average, but imply multi-scale correlations according to a wider stable distribution function.

Quasi-static deformations of soft particles, e.g. glasses, colloids, emulsions, foams, and granular materials, have been widely investigated because of their significant importance in industry and science. Moreover, many challenges of describing their macroscopic behaviors still remain due to disordered configurations, complex dynamics, etc.† At the microscopic scale, mechanical responses of soft particle packings are probed as a reconstruction of force-chain networks, where complicated non-affine displacements of particles cause the “recombination” of force-chains, i.e. opening and closing contacts. Once a macroscopic quantity is defined as a statistical average in force-chains, e.g. the stress tensor, elastic moduli, etc, its non-trivial response to quasi-static deformations (i.e. non-affine response) is governed by the change of the probability distribution function (PDF) of forces. Therefore, the PDFs in soft particle packings have practical importance so that a lot of theoretical studies (e.g. based on the stress ensemble,† force network ensemble, entropy maximization,† and so on) have been devoted to determine their functional forms observed in experiments. In general, the PDFs are asymmetric and cannot be described by conventional distribution functions. Moreover, there is still much debate about their tails, as well as their shapes for small forces.

In this study, we propose a new method for describing the evolution of the PDFs of forces under quasi-static deformations. Employing the Delaunay triangulation (DT) for two-dimensional packings (see Fig. 1(a)), we generalize the “overlap” between particles (i and j) connected by a Delaunay edge as

\[ x_{ij} \equiv R_i + R_j - D_{ij}, \]

where \( R_i + R_j \) and \( D_{ij} \) are the sum of radii and the Delaunay edge length, respectively, so that not only contacts \( (x_{ij} > 0) \), but also interparticle gaps or virtual contacts \( (x_{ij} < 0) \) can be included in force-chain networks.† We then apply quasi-static isotropic (de)compressions to the packings, where the area fraction, \( \phi \), increases (or decreases) by \( \delta \phi \) and the PDF of generalized overlaps, Eq. (1), captures the statistics of contacts and virtual contacts after opening or closing contacts. Our main result is that we numerically calibrate a Master equation for the PDFs of generalized overlaps, where transition rates of generalized overlaps are symmetric and can be described by conventional distribution functions. In addition, we find that the transition rates depend on both an applied strain step, \( \delta \phi \), and the distance from jamming point, \( \phi - \phi_j \), through only one scaling parameter, \( \gamma \equiv \delta \phi / (\phi - \phi_j) \), where \( \phi_j \) is the area fraction at jamming. The Master equation is able to describe all features of the PDFs, e.g. their changes during compressions and discontinuous “jumps”, i.e. restructuring around zero-overlaps, which had been observed in a previous study.

The application perspective of our method is that it allows us to compute the local energy density given by the second moment of particle overlaps as a statistical approach to large scale problems. The hydrostatic pressure and bulk modulus can be deduced from the first and second derivatives of the energy density, respectively, where the derivatives are defined by the Master equation (see the ESI ‡).

As method, we use molecular dynamics (MD) simulations

† Electronic Supplementary Information (ESI) available: [details of any supplementary information available should be included here]. See DOI: 10.1039/b000000x/‡ Since the DT is unique for each packing, virtual contacts are uniquely determined, where the total number of contacts and virtual contacts is a conserved quantity which is independent of the area fraction. We have not observed any flips of the Delaunay edges if \( \gamma \leq 10^{-3} \), and the number of flipped edges are less than 1% at most for \( \gamma \sim 10 \).
of two-dimensional frictionless soft particles. The normal force between particles in contact (i and j) is given by \( f_{ij} = k x_{ij} - \eta v_{ij} (x_{ij} > 0) \) with a spring constant, \( k \), viscosity coefficient, \( \eta \), and relative speed in the normal direction, \( v_{ij} \). A global damping force, \( \mathbf{f}_d = -\eta \mathbf{v}_p \), proportional to the particle’s velocity, \( \mathbf{v}_p \), is also introduced to enhance the relaxation, where the particles lose their kinetic energy by means of inelastic contacts and global damping. We randomly distribute a 50:50 binary mixture of \( N \) particles with two kinds of radii, \( R_i > R_j \) (\( R_i/R_j = 1.4 \)), in a square periodic box, where no particle touches others. We then rescale every radius to make mechanically stable particle packings (our method is similar to the one used in Ref. 22). In our simulations, distances from jamming are determined by the known scaling of averaged overlap \( \bar{x} = A(\phi - \phi_J) \). From our 10 samples of \( N = 8192 \) particles, we estimate \( \phi_J = 0.8458 \pm 10^{-4} \) with a critical amplitude, \( A = (0.31 \pm 0.01) \bar{x} \), where \( \bar{x} \) is the mean diameter in a packing closest to the jamming point, \( \phi - \phi_J = 1.2 \times 10^{-5} \). We also prepared 10 samples for small systems (\( N = 512, 2048 \)) and 2 samples for the largest one (\( N = 32768 \)), while we only report the results of \( N = 8192 \) since none of the results depends on system size.}

We apply an isotropic compression to the packings by multiplying every radius by \( \sqrt{1 + \delta \phi/\phi} \), where the area fraction increases from \( \phi \) to \( \phi + \delta \phi \). At the same time, all the generalized overlaps, \( x_{ij} \), change to \( x_{ij}^{\text{affine}} = x_{ij} + (D_{ij}/\phi) \delta \phi \). However, the particles are randomly arranged and their force balance is broken by compression so that the system is allowed to relax to a new mechanically stable state. After relaxation, the overlaps change to new values, \( x_{ij}^{\prime} \neq x_{ij}^{\text{affine}} \), due to non-affine displacements of the particles, where we observe four kinds of changes (from \( x_{ij} \) to \( x_{ij}^{\prime} \) as shown in Figs. (c) and (d): \( x_{12} > 0 \) and \( x_{13} < 0 \) change to 12 > 0 and \( x_{13}^{\prime} < 0 \), respectively, where they do not change their signs and thus contacts are neither generated nor broken. We name these changes “contact-to-contact (CC)” and “virtual-to-virtual (VV)”, respectively. On the other hand, \( x_{14} < 0 \) and \( x_{15} > 0 \) change to \( x_{14}^{\prime} > 0 \) and \( x_{15}^{\prime} < 0 \), respectively, where a new contact is generated and an existing contact is broken, respectively. We call these changes “virtual-to-contact (VC)” and “contact-to-virtual (CV)”, respectively.

The restructing of the force-chains, attributed to the changes, (CC), (VV), (VC), and (CV), is well captured by the PDFs of the generalized overlaps. Figure (b) displays the PDFs of the overlaps scaled by the averaged overlap before compression, \( \bar{x} = \bar{x}(\phi) \), \( x_{ij}^{\text{affine}} = x_{ij}^{\text{affine}}/\bar{x}(\phi) \), and \( \bar{x}^{\prime} = x_{ij}^{\prime}/\bar{x}(\phi) \), where we omit the subscript \( i \) from the scaled overlaps. As can be seen, the difference between affine and non-affine deformations is clear: The affine deformation just shifts the PDF before compression to the positive direction, while non-affine deformations broaden the PDF in positive overlaps and reconstruct the discontinuous “jump” around zero. Note that, however, the new PDF in negative overlaps is comparable with that after affine deformation (see the inset in Fig. (b)).

![Fig. 1](Color online) (a) Sketch of the generalized force-chain network with contacts (red lines) and virtual contacts (blue lines), where overlaps are defined as positive and negative, respectively. The widths of red lines are proportional to the strength of forces. (b) The PDFs of scaled overlaps, \( P_{\phi}(\xi) \) (squares), \( P_{\phi + \delta \phi}(\xi^{\text{affine}}) \) (triangles), and \( P_{\phi + \delta \phi}(\xi^{\prime}) \) (circles), for \( \phi - \phi_J = 1.2 \times 10^{-3} \) and \( \delta \phi = 1.2 \times 10^{-3} \). The inset is the zoom-in to the PDFs of virtual contacts. (c) and (d): Sketches of the DT around a single particle (c) before compression and (d) after relaxation, where red solid and blue dashed lines represent contacts and virtual contacts, respectively. The circles are particles with centers placed on the Delaunay vertices.

To describe such non-affine evolution of the PDFs, we introduce the Chapman-Kolmogorov equation \( 24 \),

\[
P_{\phi + \delta \phi}(\xi^{\prime}) = \int_{-\infty}^{\infty} W(\xi^{\prime} | \xi) P_{\phi}(\xi) d\xi ,
\]

(2)
where \( W(\xi' | \xi) \) is a conditional probability distribution (CPD) satisfying the normalization condition, \( \int_{-\infty}^{\infty} W(\xi' | \xi)d\xi' = 1 \).

The CPD is the probability of overlaps becoming \( \xi' \) which were \( \xi \) before compression (i.e. a distribution of \( \xi' \) around a mean value which depends on \( \xi \)). For example, the CPD for affine deformation is a delta function, \( W_{\text{affine}}(\xi' | \xi) = \delta(\xi' - f_\alpha(\xi)), \) where the mean value is given by a linear function of \( \xi \), \( f_\alpha(\xi) = \xi + B_\alpha \gamma \) with a coefficient, \( B_\alpha = D_i/(2A\phi) \), which just shifts the PDF by \( B_\alpha \gamma \); i.e. \( P_\phi(\xi - B_\alpha \gamma) \), as shown in Fig. 1(b)[2].

On the other hand, the CPDs for non-affine deformations can be measured through scatter plots of the scaled overlaps, see Figures 2(a) and (b), where the four kinds of changes are mapped onto four regions: (CC) \( \xi, \xi' > 0 \), (VV) \( \xi, \xi' < 0 \), (VC) \( \xi < 0, \xi' > 0 \), and (CV) \( \xi > 0, \xi' < 0 \), respectively. In (CC) and (VV), the scaled overlaps after compression distribute around mean values which we describe by linear fitting functions for \( \xi' \),

\[
f_n(\xi) = (a_n + 1)\xi + b_n, \tag{3}
\]

where the subscripts, \( n = c \) and \( v \), represent the mean values in (CC) and (VV), respectively. If we introduce standard deviations of \( \xi' \) from \( f_\alpha(\xi) \) as \( v_n \), which are almost independent of \( \xi \), the systematic deviation from affine deformations can be quantified by the coefficients, \( a_n, b_n, \) and \( v_n \), as summarized in Fig. 2(c). Note that the differences are always present, but if not visible if the applied strain is small or the system is far from jamming, i.e. if \( \gamma \ll 1 \) (Fig. 2(a)), while \( \xi' \) deviates more from \( f_\alpha(\xi) \) and data points are more dispersed if we increase \( \gamma \) (Fig. 2(b)). For example, Fig. 2(d) shows a double logarithmic plot of \( a_\gamma \) against \( \gamma \), where all data collapse onto a linear scaling, \( a_\gamma \simeq A_\gamma \gamma \) with \( A_\gamma = 0.76 \pm 0.002 \). We also find other scaling relations, \( a_v \simeq 0 \), \( b_v \simeq B_v \gamma \), \( b_\gamma \simeq B_v \gamma \), \( v_v \simeq V_v \gamma \), and \( v_\gamma \simeq V_\gamma \gamma \) with \( B_v = 0.24 \pm 0.002 \), \( B_\gamma = 1.80 \pm 0.001 \), \( V_v = 0.32 \pm 0.01 \), and \( V_\gamma = 4.41 \pm 0.06 \), respectively, for \( \gamma < 1 \) (see the ESI[1]), so that all parameters characterizing the mean values and fluctuations are linearly scaled by \( \gamma \). Because \( a_v \simeq 0 \) and \( b_v \simeq B_v \gamma \), we have, for their large fluctuations (\( V_v \gg V_\gamma \)). In contrast to (CC) and (VV), the data of \( \xi' \) in (VC) and (CV) are concentrated in narrow regions (between the axes and the dashed lines in Fig. 2(c)), whereas \( f_\alpha(\xi) \) linearly increases with \( \xi \) in (VC) and there is no data of \( f_\alpha(\xi) \) in (CV), i.e. the affine deformation gives closing contacts only.

We then determine the CPDs for non-affine deformations as the distributions of scaled overlaps, \( \xi' \), around their mean values, \( f_\alpha(\xi) \). Figure 3(a) shows the CPDs in (CC), where all results with a wide range of \( \gamma \) are symmetric around \( f_\alpha(\xi) \) and collapse if we multiply \( W_{\text{CC}}(\xi' | \xi) \) and \( \xi' - f_\alpha(\xi) \) by \( \gamma \) and \( 1/\gamma \), respectively. The solid line is a Gaussian distribution function,

\[
\gamma W_{\text{CC}}(\xi' | \xi) = \frac{1}{\sqrt{2\pi V_\gamma^2}} e^{-\gamma^2/2V_\gamma^2}, \tag{4}
\]

with \( \Theta \equiv \left[ \xi' - f_\alpha(\xi) \right]/\gamma \). Figure 3(b) displays the CPDs in (VV), where all results are also symmetric around \( f_\alpha(\xi) \) and collapse as well, after the same scaling as for (CC). The solid line here is a stable distribution function

\[
\gamma W_{\text{VV}}(\xi' | \xi) = \frac{1}{2\kappa} \int_{-\infty}^{\infty} e^{-\left(\kappa|z|e^{\lambda z} + \mu z\right)/\kappa} dz, \tag{5}
\]

with \( \Omega \equiv \left[ \xi' - f_\alpha(\xi) \right]/\gamma \), where \( z \) is a dimensionless wave number, and the fitting parameters are given by \( \lambda = 1.65 \) and \( \kappa = 0.62 \), respectively, i.e. the CPD in (VV) is nearly a Holtsmark distribution (\( \lambda = 3/2 \) and \( \kappa > 0 \)). Figures 3(c) and (d) show the CPDs in (CV) and (VC) approximated by exponential distributions,

\[
\gamma W_{\text{CV}}(\xi' | \xi) = \left\{ 1 - L_{\text{CC}}(\xi) \right\} e^{\lambda_\nu q_\nu z}, \tag{6}
\]

\[
\gamma W_{\text{VC}}(\xi' | \xi) = \left\{ 1 - L_{\text{VV}}(\xi) \right\} e^{-\lambda_\nu q_\nu z}, \tag{7}
\]

\textit{**\gamma can be large, whereas \( \delta \gamma \) is always small.**}
respectively, where \( \Lambda \equiv \xi / \gamma \) and the dimensionless lengths are given by \( q_i = 6.10 \) and \( q_e = 0.65 \) (\( q_e \gg q_i \)), respectively. In curly brackets on the right hand sides, \( I_{CC}(\xi) \equiv \frac{1}{2} \text{erfc} \left( -\frac{\lambda}{\sqrt{2} \xi} \right) \) and \( I_{VV}(\xi) \equiv \int_{-\infty}^{0} W_{VV}(\xi'|\xi) d\xi' \) are the cumulative distribution functions of the CPDs in (CC) and (VV), respectively, which are required to satisfy the normalization conditions \( \int_{-\infty}^{\infty} I_{CC}(\xi) d\xi = 1 \) and well describe the dependence of the CPDs on \( \xi \) (see the ESI). In addition, if \( \gamma = 0 \), \( W_{CC} = W_{VV} = \delta(\xi - \xi^0) \) and \( W_{CV} = W_{VC} = 0 \) so that the Chapman-Kolmogorov equation \( \partial_t P(\xi; t) = T(\xi|\xi^0) P(\xi^0) \) does not change the PDF without deformations.

Now, we restrict \( \delta \phi \) to quite small values compared to \( \phi - \phi_j \) and define an infinitesimal scaled strain step as \( \delta \gamma \equiv \delta \phi / (\phi - \phi_j) \ll 1 \). Introducing a transition rate as \( T(\xi|\xi^0) = \lim_{\delta \gamma \to 0} W(\xi'|\xi) / \delta \gamma \), we rewrite the Chapman-Kolmogorov equation \( \partial_t P(\xi; t) = T(\xi|\xi^0) P(\xi^0) \) as a Master equation \( \partial_\gamma P_\phi(\xi) = \int_{-\infty}^{\infty} \left[ T(\xi'|\xi) - T(\xi'|\xi^0) P_\phi(\xi^0) \right] d\xi' \), where we use the CPDs, Eqs. (4)-(7), for the transition rates. Figures 3a) and (b) display the numerical solutions of the Master equation under incremental compression steps, where the increment of area fraction is fixed to \( \delta \gamma = 10^{-3} \) so that \( \delta \gamma \lesssim 2.5 \times 10^{-3} \) throughout the numerical integrations. Here, the initial condition is given by the PDF obtained through MD simulations with the distance from jamming, \( \phi_0 - \phi_j = 4 \times 10^{-3} \). The overlaps are scaled by the averaged overlap at the initial state, \( \bar{\xi}(\phi_0) \). Good agreements between the solutions (red solid lines) and MD simulations (open symbols) are established for small \( \delta \gamma \) even in the tails of the PDFs (the inset in Fig. 3b). In addition, the Master equation reproduces discontinuous jumps of the PDFs around zero-overlap as observed in Fig. 1b). We also confirmed that numerical solutions starting from different initial conditions, e.g. a step function and a Gaussian distribution (not consistent with mechanical stability), converge to a unique solution with discontinuous jumps around zero (see the ESI). In addition, MD simulations of decompression tests with the increment of area fraction, \( \delta \phi < 0 \), we find that the mean values and CPDs are given by just replacing the scaling parameter, \( \gamma \) with \( -\gamma \) in Eqs. (3)-(7), which does not change the form of the Master equation (8). Therefore, the linear scalings of the coefficients for non-affine deformations, \( a_n, b_n \), and \( v_n \), are maintained under decompression, and the functional forms of the CPDs are the same for both compression and decompression (see the ESI). However, note that the scattered data under compression and decompression are not symmetric with respect to the diagonal line, \( \xi' = \xi \). Thus, the transition rates for decompressions are \( T_{\delta \gamma;0}(\xi'|\xi) \neq T_{\delta \gamma;0}(\xi|\xi') \), which leads to irreversible responses of soft particle packings under quasi-static cyclic (de)compressions.

In summary, we provide, for the first time, a Master equation for the PDFs of forces in soft particle packings under quasi-static (de)compressions, where not only the changes of contacts and virtual contacts, but also their mutual exchange, i.e. opening and closing contacts, are included in the transition rates for the Master equation. The transition rates (or the CPDs of the generalized overlaps) are symmetric around mean values with finite widths, where both the mean and fluctuations are well characterized by a single scaling parameter, \( \gamma = \delta \phi / (\phi - \phi_j) \), quantifying the degree of non-affine deformations. We confirm that shapes of the CPDs and linear scalings for the mean and fluctuations are the same for compression and decompression. The Master equation can predict the incremental evolution of the PDFs, including discontinuous jumps around zero, that is, the multi-particle system is reduced to a single-contact picture, i.e. a mean-field like description.

The CPDs show by themselves important properties: Contacts respond in a non-affine way, especially near jamming, as quantified by the scaling, e.g. \( a_n \sim \gamma = \delta \phi / (\phi - \phi_j) \). Astonishingly, their fluctuations obey Gaussian statistics, indicating uncorrelated stochastic evolution of forces. In contrast, the nearly Holtsmark distributions feature much broader
The solutions develop in the directions indicated by the arrows. The open squares, circles, and triangles are the PDFs obtained from MD simulations with $\phi - \phi_J = 4 \times 10^{-3}$, $1.2 \times 10^{-2}$, and $4 \times 10^{-2}$, respectively. The insets show the semi-logarithmic plots. Overlaps are scaled by the averaged overlap at $\phi_0 - \phi_J = 4 \times 10^{-3}$.

tails for virtual contacts that deform affinely in average. Indicating much larger changes of interparticle gaps, this implies a strongly correlated stochastic evolution over a wide range of length-scales. The probabilities for opening and closing contacts are exponentially decaying with distance from zero (i.e. $e^{-|\Delta|/q_v}$ and $e^{-|\Delta|/q_c}$ in Eqs. 3 and 4, respectively), and cause the discontinuous jumps in the PDFs, since opening contacts are free to open widely whereas closing contacts are affected by repulsion, i.e. $q_v \gg q_c$. Because both the Gaussian and Holtsmark distributions are members of the stable distribution family, fluctuations of contacts and virtual contacts in soft particles should obey the generalized central limit theorem, which has consequences for the statistical description of disordered systems in general. The strong deviation from an affine approximation for contacts and the enormous fluctuations of overlaps for virtual contacts, as well as the probabilities for opening and closing of contacts, are all proportional to the scaled strain step, $\gamma$.

Clearly, there is the need of further studies on the physical origin of the statistics of overlaps described above. The functional forms of the CPDs can give very interesting insights into the micro-mechanics of soft particles, e.g. stochastic processes of overlaps in force-chain networks. Now, analytic solutions or asymptotic solutions of the Master equation are important next steps towards the understanding of the functional forms of the PDFs. The Master equation also poses a new challenge; it requires the increment $\delta\phi$ to be much smaller than $\phi - \phi_J$, i.e. $\gamma \ll 1$. Thus, strictly speaking, it can never reach $\phi_J$, and the result cannot be the PDF at $\phi_J$, albeit asymptotically. This means that the jamming transition is a singular limit of the Master equation.

Finally, our analysis can be easily extended to three dimensions and be examined and validated by experiments, e.g. by photoelastic tests or oedometer test of sands. The extension to other cases is also straightforward, e.g. the solutions under shear can be obtained if we apply our results for (de)compressions to each principal direction (in preparation).

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