Orientational arrest in dense suspensions of elliptical particles under oscillatory shear flows

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Abstract – We study the rheological response of dense suspensions of elliptical particles, with an aspect ratio equal to 3, under oscillatory shear flows and imposed pressure by numerical simulations. Like for the isotropic particles, we find that the oscillatory shear flows respect the Cox-Merz rule at large oscillatory strains but differ at low strains, with a lower viscosity than the steady shear and higher shear jamming packing fractions. However, unlike the isotropic cases (i.e., discs and spheres), frictionless ellipses get dynamically arrested in their initial orientational configuration at small oscillatory strains. We illustrate this by starting at two different configurations with different nematic order parameters and the average orientation of the particles. Surprisingly, the overall orientation in the frictionless case is uncoupled to the rheological response close to jamming, and the rheology is only controlled by the average number of contacts and the oscillatory strain. Having larger oscillatory strains or adding friction does, however, help the system escape these orientational arrested states, which are evolving to a disordered state independent of the initial configuration at low strains and ordered ones at large strains.

Introduction. – Mechanical properties of soft materials and complex fluids, such as suspensions in concentrated regimes, emulsions and granular materials, are challenging to describe due to their often complicated and dynamical many-body effects. Such systems have appeared as an important field of study not only from a pure physics perspective but also due to their practical applications in materials science and numerous other areas [1,2]. Oscillatory shear flows have been broadly used to investigate dense suspensions mechanical properties [3–5]. Previous studies have shown that the rheology of dense suspensions under oscillatory shear does not necessarily follow the Cox-Merz rule, which states that the oscillatory shear and steady shear viscosities should be equal to each other [6]. At constant shear rate \(\dot{\gamma}\) the viscosity \(\eta\) of dense non-Brownian suspensions consisting of rigid particles diverges as \(\eta/\eta_f \sim (\phi_c - \phi)^{-\alpha}\), where \(\eta_f\) is the viscosity of the background fluid, \(\phi\) is the particles packing fraction and \(\alpha\) is a positive exponent typically close to 2 [6,7]. The shear jamming packing fraction \(\phi_c\) is dependent on various parameters, including particles shape [8–15], friction [16–18] and interactions [19–22]. The rheology of dense suspensions can be quite complicated at unsteady shear conditions, showing non-trivial transient rheological behaviour [23–25]. It has been shown that oscillatory shear perpendicular [26–28] or parallel to the primary constant shear [29] can help flowability of dense suspensions by reducing the viscosity in a controlled way [28,29].

The viscosity reduction in suspensions is generally attributed to a restructuring of the microstructure [29] which, for instance, at orthogonal shear flows, happens through a tilting and ultimate breakage of the force chains [26] or more generally through random organization [28,30,31]. These studies were done on isotropic granules, although in reality, particles have some anisotropy.

The present study will explore the effect of shape anisotropy on the rheology of dense non-Brownian suspensions under oscillatory shear.

Simulation method. – Using a discrete element method we study two-dimensional suspensions consisting of \(\sim 1000\) ellipses with \(\pm 50\%\) poly-dispersity in major axis with a flat distribution and an aspect ratio of \(\alpha = a/b = 3\).
where \(a\) and \(b\) are the major and minor radius, respectively. Having poly-dispersity in particles’ sizes is to avoid (oscillatory shear-induced) crystallization. The eccentricity of an ellipse is given as \(e = \sqrt{1 - \alpha^2}\). We apply a constant external pressure \(P_{\text{ext}}\) on the two rough confining walls in their normal direction (now denoted \(y\)-direction). The two walls have a relative oscillatory velocity difference in transverse \(x\)-direction which leads to a macroscopic shear rate \(\dot{\gamma}(t) = \gamma_0 \cos(\omega t)\), where \(\gamma_0\) is the amplitude and \(\omega\) is the frequency of the oscillatory shear. Periodic boundary condition is considered in the \(x\)-direction (see fig. S2 in the Supplementary Material).

**Supplementary material (SM)**. Wall particles have the same properties as the flowing ellipses. The corresponding strain will be \(\gamma(t) = \gamma_0 \sin(\omega t)\), where \(\gamma_0 = \dot{\gamma}_0 / \omega\) is the amplitude of the oscillatory strain. Particles interact with each other via harmonic forces, \(f^{ij} = f^{i\text{ext}} + f^{i\text{visc}} = k_n \delta^{ij} \mathbf{u}^{i0} + k_t \delta^{ij} \mathbf{v}^{i0}\), where \(k_n\) and \(k_t = k_n / 2\) are the normal and the tangential spring constants, respectively, and where \(\delta^{ij}\) is the normal overlap and tangential displacement between the particles and \(i\) and \(j\) [13]. Coulomb friction \(|\mathbf{f}^{ij}_\text{friction}| \leq \mu_p |\mathbf{f}^{ij}_\text{ext}|\), is the constraint for the tangential force with \(\mu_p\) being the friction coefficient of the particles which can be either \(\mu_p = 0\) (frictionless) or \(\mu_p = 0.4\) (frictional) unless otherwise stated. Likewise, when particles \(i\) and \(j\) are in contact, they exert torques \(\tau^{ij}\) on each other. To have rigid particles, the ratio \(k_{nt}/P_{\text{ext}}\) between the normal spring constant and the external pressure is set to be \(3 \times 10^4\). The suspending fluid is Newtonian and treated as a continuum. We consider a viscous incompressible Stokes flow with the nonlinear inertia terms being negligible. Applying the superposition principle in the linear Stokes regime, \(f\), and torque, \(\tau\), imposed by the uniform background shear flow on each elliptical body \(i\) are described by \(f^{i\text{visc}} = 3 \pi \eta f c_f a (u^{i\text{y}}(y) - u^{i\text{y}})\) and \(f^{i\text{ext}} = f^{i\text{visc}} (1 + k_n / P_{\text{ext}})\), and \(\tau^{i\text{visc}} = 4 \pi \rho a b (c_{M\text{a}} e_i \dot{e}_i + (c_{M\text{b}} e_i \dot{e}_i)^2) \omega - c_{M\text{r}} \omega_i\) [32,33], where \(x\) and \(y\) are the unit vectors in \(x\) and \(y\) coordinates, respectively, \(u\), \(\omega\), and \(c\) are the translational and angular velocity of the particle \(i\), \(e_i = (e_i \mathbf{x}, e_i \mathbf{y})\) is its unit direction vector along the major axis and \(u_i = (u, e_i)\) and \(u_i = u - u_i\) are the particle’s velocity in major and minor axes directions, respectively. \(f^{i\text{y}}(y) = (\gamma y, 0)\) is the fluid velocity with \(y\) as the \(y\)-coordinate of the ellipse \(i\) and \(\omega = \dot{\gamma} / 2\) is the fluid angular velocity. Applying the boundary conditions such as the no-slip condition on the surface of the particles, coefficients are determined as (see ref. [32])

\[
c_{fa} = \frac{4}{3} e_3^2 [-(2e^2 + 1 + e^2) \log(\frac{1 + e}{2})], \quad c_{fb} = \frac{2}{3} e_3^2 [2e^2 + (3e^2 - 1) \log(\frac{1 + e}{2})], \quad c_{M\text{a}} = c_{fa}, \quad c_{M\text{b}} = (1 - e^2)^{-1} c_{fa}, \quad \text{and} \quad c_{M\text{r}} = 4 \pi \alpha [\frac{2}{3} (\frac{1 + e}{2})^2 [2e^2 + (3e^2 - 1) \log(\frac{1 + e}{2})]]^{-1}.
\]

The dynamics of the ellipses are overdamped, which leads to force and torque equations given as \(f^{i\text{int}} + f^{i\text{visc}} = -\sum_j f_{ij}\) and \(\tau^{i\text{int}} + \tau^{i\text{visc}} = -\sum_j \tau_{ij}\), where \(f^{i\text{int}}\) and \(\tau^{i\text{int}}\) are the external forces and torques, respectively. The characteristic time is given by \(\ell_0 = 3 \pi \eta f c_f a / (k_n \sqrt{\alpha})\). The equations of motion are integrated by using \(\delta t / \ell_0 = 0.1\) and the Heun method.

We select two preparation protocols for our suspensions, being either frictional or frictionless. The first one uses a pre-sheared protocol, where suspensions are first steadily sheared, and once they reached a steady state, we turn off the shear rate and let the suspension settle under the external pressure. This leads to a well-defined orientation of the ellipses. The second one corresponds to a non-directional/random protocol. We randomly placed and oriented ellipses in a dilute regime and then subjected them to compression by our external pressure. Once the two initial configurations are settled, the oscillatory shear is applied.

Average properties are measured after the suspensions come to a steady cyclic state; that is, the averages of each oscillation period only fluctuate around particular mean values. The lowest accumulative strain for which averaged data is collected is \(10\) \((\gamma_{\text{acc}} = \int |\gamma| dt \geq 10\) and for the largest \(\gamma_0\) a minimum of one full oscillation period is considered. Within each oscillation, 60 measurements are made (having \(2\pi / \omega > 60\)) \(\delta t\). We excluded the 5 closest layers to each wall in our measures to remove possible boundary effects. The stress tensor is calculated according to \(\sigma^{ijkl} = 1 / (2A) \sum_{i<j} f^{ij}_v r^{ij}_v\), where \(A\) is the area over which the stress is measured and \(r^{ij}_v\) is the two-particles center-to-center vector. The nematic order parameter \(S_2\) is taken as the largest eigenvalue of the director tensor \(Q^{ijkl} = 1 / N \sum_{i<j} (2e_i^2 e_j^2 - \delta_{ij})\), where \(N\) is the number of particles over which the measure is taken and \(\delta_{ij}\) the Kroenecker delta. \(\theta_{y\tilde{y}}\) is measured as the average particle angle with respect to the \(y\)-direction, normal to the lower surface (see fig. S2 in the SM). It is also possible to measure another direction angle, the particles’ angle with respect to the flow direction and parallel to the lower surface [34]. Here we only report the former angle.

**Suspensions oscillatory-shear rheology.** – Despite the steady shear rheology with solely viscous stresses and viscosities, at pure oscillatory shear flows, the stress and the viscosity will be a combination of both elastic and viscous responses. The complex stress in the linear response regime is given as [5,35] \(\sigma^{ijkl} = \eta'' \gamma_0 \sin(\omega t) + \eta' \gamma_0 \cos(\omega t)\), where \(\eta''\) and \(\eta'\) are the corresponding elastic (imaginary) and viscous (real) part of the complex viscosity \(\eta^* = \eta' - i\eta''\) and \(t\) the time. The magnitude of the complex viscosity is given by \(|\eta^*| = \sqrt{\eta''^2 + \eta'^2}\), with \(\eta'' = \int_{2\pi / \omega}^{t} \sigma(t) \cos(\omega t) dt\) and \(\eta' = \int_{2\pi / \omega}^{t} \sigma(t) \sin(\omega t) dt\) [29]. In a similar manner and analogous to the dimensionless viscous number \(J = \eta_\tau / \gamma P\) for suspensions in the steady viscous regime, we can define a shear-rate–weighted average viscous number \(J'\) as \(J' = \eta_\tau (\int_{2\pi / \omega}^{t} \gamma P(t) dt) / \int_{2\pi / \omega}^{t} |\gamma| dt\) [29]. \(J'\) is a measure of average shear rate (formally shear-rate–weighted) of the suspension and is directly related to how far away one is from a suspensions jamming point (i.e., \(\phi_\alpha - \phi\)). Following the same approach as in [29] we have generalized this shear-rate–weighted average definition for other operating parameters as well (see the SM for the
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Fig. 1: Evolution of (a), (b)) the direction angle $\theta_{e} \cdot \hat{y}$, (c), (d)) the nematic ordering $S_2$, (e), (f)) the packing fraction $\phi$ and (g), (h)) the number of contacts $Z$ for the frictional ($\mu_p = 0.4$) (left column) and the frictionless ($\mu_p = 0$) (right column) configurations at $\gamma_0 = 0.1$ and $J' \approx 0.1$. Empty and full symbols correspond to pre-sheared and non-directional preparations, respectively. Black lines are best fits of the relaxations. The blue dashed lines in (a), (b) indicate zero lines.

Results and discussion. – Figure 1 shows some typical numerical time evolutions of $\theta_{e} \cdot \hat{y}$, $S_2$, $\phi$ and $Z$ for oscillatory shear (OS) from the two preparation protocols, pre-sheared and random packings, at a low oscillatory strain ($\gamma_0 = 0.1$) for both frictional and frictionless particles. The packings are initially at rest. For the frictional case, the two starting configurations at the same $J'$ converge to the same packing fraction higher than the corresponding steady-shear value. They also converge to a directional disordered state with a low nematic ordering and average orientation fluctuating around zero. These orientational values are close, if not identical, to the initial random configuration, i.e., the pre-sheared samples relax to random configurations. One also notices that the packing fraction relaxes much faster than the two direction parameters. The number of contacts also relaxes quite rapidly and becomes identical for the two protocols. On the other hand, interestingly, this is not the case for the frictionless ellipses, where all quantities except the number of contacts of the two preparation protocols stay separated and without any detectable relaxation within our numerical time window.

To get further insight, we estimate strains over which the various quantities possibly relax. We assume exponential relaxations as $A_\infty \equiv A_\infty + (A_0 - A_\infty) \exp(-\kappa \gamma_{acc})$, where $A$ can be either $\theta_{e} \cdot \hat{y}$, $S_2$, $\phi$ or $Z$, and $\kappa^{-1}$ represents a relaxation strain. Figure 1 shows that the relaxation matches well an exponential description, shown by solid black lines, in the frictional case. Generally, the definition of the shear-rate–weighted averages of $\phi$, $Z$, $S_2$, and viscous/elastic stress ratio $\mu = \sigma/P$).

Fig. 2: Rescaled relaxation parameter of the nematic order $\kappa_{S_2}^{-1}$ for the pre-sheared configuration as a function of $\mu_p$ at $\gamma_0 = 0.1$ for various $J'$. Each $J'$-curve has been normalised by its corresponding value at $\mu_p \to \infty$. The dashed line indicates the best fit of the data with $\kappa_{S_2}^{-1} \sim a S_2 \mu_p^{-\beta} + c$. The relaxations of $S_2$ and $\theta_{e} \cdot \hat{y}$ follow each other (see fig. 1(a) and (c) and the SM), while $\phi$ and $Z$ relax an order of magnitude faster than the two other quantities (see fig. 1(e) and (g)). The relaxation strains depend on friction coefficient, oscillatory strain (see the SM), and $J'$. For purely frictionless particles and at a typical small $\gamma_0$ (e.g., 0.1), the packing fraction relaxes almost instantaneously, but to different values depending on if one starts from a random or pre-sheared configuration (see right panels in fig. 1). For the same system, the orientational relaxation from a
pre-sheared configuration does, however, show a seemingly infinite relaxation strain and at odds with its corresponding frictional case. To further investigate the role of frictional interaction, we simulate and measure $S_2$ relaxation strains, from pre-sheared configurations, for systems with other friction coefficients. Figure 2 shows the relaxation strain in $S_2$ as a function of the friction coefficient $\mu_p$. According to fig. 2 relaxation strains in $S_2$ possibly diverge as $\kappa_{S_2} \sim \mu_p^{-\beta}$ (with $\beta = 3.06 \pm 0.29$). Due to its sharp divergence, it is hard to tell if such divergence occurs at a finite critical friction coefficient $\mu_{p,c}$ (i.e., $\kappa_{S_2} \sim (\mu_p - \mu_{p,c})^{-\beta}$) rather than $\mu_{p,c} = 0$ (see other alternative fits in the SM). However, in the present study, $\mu_p = 0.1$ is the smallest friction coefficient corresponding to which we extracted the slowest detectable relaxation strains for the nematic ordering $S_2$. For $\mu_p < 0.1$, and the same as for the frictionless case, we could not detect any relaxations (beyond our noise level) at low $\gamma_0$. So, the friction coefficient threshold for the orientational arrest at small $\gamma_0$ lies between 0 and 0.1, and the exact value is left as an open question. Adding a finite amount of friction does, however, help the system to relax its orientation, a process similar to ratcheting [36]. Above $\mu_p = 1$, the relaxation strains saturate and only depend on $J'$ and $\gamma_0$. Smaller $J'$ values display smaller relaxations in terms of strains (see the inset in fig. 2), reflecting the higher packing fractions in those cases, with a larger number of contacts and number of collisions per strain.

Similarly we find that these infinite relaxation strains for the frictionless particles appear first when $\gamma_0 < 0.3$, with a power-law divergence $\kappa_{S_2} \sim (\gamma_0 - \gamma_{0,c})^{-\nu}$, with $\nu \sim 3.6$ and $\gamma_{0,c} \approx 0.1$ (see the SM).

Further evidence that at low oscillatory strains $\gamma_0 \lesssim 0.1$, frictional ellipses relax but frictionless ones do not can be found when studying the transient stress response. Figure 3 shows the stress response of pre-sheared samples after an initial relaxation in packing fraction. The frictional case shows a clear asymmetric response when sheared along the pre-sheared direction (gray zones in fig. 3(a)) compared to opposite to it (white zones in fig. 3(a)), while the frictionless case (fig. 3(b)) lacks this (within the noise, see the SM for more details). This asymmetry leads to an imbalance in dissipation and elastic storage in various directions that helps to evolve the system. While this asymmetric stress response is only transient, it persists up to 100 accumulated strains (and much larger than the typical turnover strain), corresponding to, e.g., 2700 oscillations at $\gamma_0 = 0.01$ at $J' \sim 0.1$.

Having determined that the frictionless particles do not fulfill one unique equation of state for low oscillatory strains, we investigate how the complex viscosity varies with packing fraction, and $\gamma_0$ for our two preparation protocols compared to frictional particles, see fig. 4(a), (b). For both (a) frictional and (b) frictionless ellipses, we find that the rheological response (i.e., $\eta' \mid \eta''$ vs. $\phi$) behaves as their corresponding steady shear (SS) cases for large $\gamma_0$'s. At lower $\gamma_0$'s, one finds a lower viscosity than SS at the same packing fraction, with an increased shear jamming packing fraction for the frictional particles, i.e., $\phi_{c,\text{OS}} < \phi_{\text{SS}}^{\max}$. These findings are in line with what has previously been reported for discs [29,37] and spheres [26,28]. Unlike for isotropic particles, frictionless ellipses show a preparation-dependent rheology (compare full and open symbols), with higher shear jamming packing fractions for pre-sheared preparations compared to non-directional/random ones. For the frictionless ellipses the non-directional preparation yields a shear-jamming below its corresponding point in SS (i.e., $\phi_{c,\text{OS}}^{\text{ran}} < \phi_{\text{SS}}^{\max}$). This shows that there exist at least two well-separated oscillatory shear-jamming points for frictionless elliptical particles. Nonetheless, once the complex viscosity $|\eta'|/\eta''$ is plotted against the number of contacts $Z$ in fig. 4(c) and (d), for frictionless particles, the two protocols with different $\phi$'s collapse on each other despite having both different packing fractions and orientational properties. Similar to the disc and sphere cases [27,29], the low oscillation strains follow the steady shear curve, but unlike

Fig. 3: Time series of the normalized shear stress for (a) frictional $\sigma'/\sigma_{\text{SS},I}$, and (b) frictionless $\sigma'/\sigma_{\text{SS},\text{nf}}$ pre-sheared configurations at $\gamma_0 = 0.01$ and $J' = 0.1$. $\sigma_{\max}$ and $\sigma'_{\max}$ are the stresses at the corresponding $J'$ in steady shear for frictional and frictionless ellipses, respectively. Gray and white zones represent forward and backward shearing, i.e., along with the average orientation of the particles and opposite to it, correspondingly. Panels (c) and (d) show the corresponding rescaled shear rates $\gamma'/\gamma_0$. 

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Fig. 4: Normalized complex viscosities $|\eta^*/\eta_f|$ vs. the packing fraction $\phi$ ((a) and (b)) and the number of contacts $Z_{c,||}$ ((c) and (d)) at various strain magnitudes $\gamma_0$. (a) and (c) belong to frictional particles while (b) and (d) express the frictionless ones. Empty and full symbols correspond to pre-sheared and non-directional preparations, respectively. The grey solid and dashed lines are the corresponding steady shear viscosity curves for frictional and frictionless particles, respectively. The brown dotted vertical lines in (a) and (b) show the steady shear jamming packing fractions for frictional $\phi_{c, SS}^{\gamma_0}$ and frictionless $\phi_{c, nf}^{\gamma_0}$ suspensions while in (c) and (d) they indicate the steady shear jamming number of contacts for frictional $Z_{c, SS}^{\gamma_0}$ and frictionless $Z_{c, nf}^{\gamma_0}$ configurations. The inset in panel (b) shows $\phi$ as a function of $J'$ for the frictionless ellipses, where the grey dashed line indicates the corresponding steady shear curve. Similarly, the inset in panel (d) illustrates $Z$ vs. $J'$ for the non-frictional particles, with the dashed line showing the respective steady shear case.

As for tapping [39], oscillations are thought to equilibrate these athermal systems, leading to a unique equation of state of a “thermalised” athermal system (i.e., the Edwards conjecture [40]). Hence, finding two packing fractions for frictionless particles at small oscillatory strains violates this conjecture, as ergodicity seems not to hold while fulfilled for the frictional cases. Instead, the frictionless cases end up in a dynamically arrested state, similar to a glassy state, keeping its original nematic ordering indefinitely. These findings shed light on the importance of preparation for granular systems composed of elongated particles, e.g., rice and cereals, on their mechanical properties and their reproducibility.

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Data availability statement: The data that support the findings of this study are available upon reasonable request from the authors.
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