Making soft solids flow: microscopic bursts and conga lines in jammed emulsions

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It is well known that jammed soft materials will flow if sheared above their yield stress – think mayonnaise spread on bread – but a complete microscopic description of this seemingly simple process has yet to emerge. What remains elusive is a microscopic framework that explains the macroscopic flow, derived from a 3-D spatially resolved analysis of the dynamics of the droplets or particles that compose the soft material. By combining confocal-rheology experiments on compressed emulsions and numerical simulations, we unravel that the primary microscopic mechanisms for flow are strongly influenced by the rate of the imposed deformation. When shearing fast, small coordinated clusters of droplets move collectively as in a conga line, while at low rates the flow emerges from bursts of droplet rearrangements, correlated over large domains. These regions exhibit complex spatio-temporal correlation patterns that reflect the long range elasticity embedded in the jammed material. These results identify the three-dimensional structure of microscopic rearrangements within sheared soft solids, revealing that the characteristic shape and dynamics of these structures are strongly determined by the rate of the external shear.

A hallmark of all soft disordered solids is their highly nonlinear response to the rate of ex-
ternally applied shear deformations. Experiments reveal that granular solids, colloidal glasses and foams exhibit shear localization that results in a spatially heterogeneous rheological response. The bulk flow behavior of soft solids is similar to various yield stress fluids, where the flow curve generically follows an empirical dependence of the shear stress $\sigma$ on the shear rate $\dot{\gamma}$, $\sigma = \sigma_Y + K \dot{\gamma}^n$, where $\sigma_Y$ is the yield stress and $K$ and $n$ are constants specific to the flowing material. For $n \sim 0.5$ the flow curve has a Herschel-Bulkley (HB) form which can be understood in terms of a non-local constitutive relationship. The underlying assumption is that, close to yielding, plastic and irreversible rearrangements are the local mechanism for shear stress dissipation. 2-D simulations and mean-field theories reveal that localised plastic rearrangements occurring within a nearly continuum elastic background may lead to avalanches. Experiments that focus on particle scale dynamics also point to the importance of quantifying local rearrangements and their correlations. Interestingly, these local rearrangements are expected to be rate dependent and could be used, in principle, to predict the form of the macroscopic flow. What remains elusive is a microscopic framework that explains the macroscale rheology derived from a three-dimensional spatially-resolved analysis of particle level dynamics.

One ideal model soft solid that exhibits universal rheological nonlinearities is an emulsion, a widely used system for the transport of highly value-added materials and for the efficient processing of immiscible fluids. By combining confocal-rheology experiments and molecular dynamics simulations, we quantitatively describe the droplet scale deformations in a sheared three-dimensional jammed emulsion. Our combined approach is an essential step, due in part to microscopic insights provided by numerical simulations that are often beyond the limits of experimental
resolution. In this work, we find that the elementary rearrangements reveal unexpected distributions of particle displacements and localised structures that are spatio-temporally correlated and highly dependent on the imposed shear rate. The physical picture is that the flow at high shear rates originates from the highly coordinated motion of small particle clusters that move collectively in same direction. Conversely, at low shear rates, the bulk flow emerges from elementary non-affine rearrangements which take place over much larger correlated domains that trigger events later in time, determined by the long-range elastic strain field of the initially solid amorphous material. The nature itself of the elementary and non-affine type of rearrangements, which are the primary microscale mechanism for flow, appears therefore to be dependent on the flow rate. The novel insight obtained here is a fundamental starting point for the development of new theories and constitutive models that are needed to design and control the flow of technologically relevant materials.

The experimental system is a direct emulsion of silicone oil droplets stabilized with the surfactant sodium dodecyl sulfate (SDS) in an index-matched continuous phase of water, glycerol, and fluorescein (see Methods). The droplets, with an average diameter of 6.0 µm and polydispersity of 0.15, were compressed above the jamming point with centrifugation. Measurements of the system under flow were performed with a custom stress-controlled rheometer integrated with a laser scanning confocal microscope. As shown in Fig 1, the rheometer’s gap is formed by a glass coverslip, which provides optical access from below for the microscope, and a parallel plate tool. Images were taken at a fixed relative position between the two instruments, where the local velocity, vorticity, and gradient axes will be referred to as x, y, and z, respectively.
We measure the influence of shear rate on the local droplet dynamics by acquiring and analysing time-resolved fluorescence confocal images while the rheometer simultaneously applies a continuous rotation at a fixed strain rate \( \dot{\gamma} \) (see Methods). For \( \dot{\gamma} \leq 10^{-2} \, s^{-1} \), time-resolved 3-D stacks are acquired, while at higher shear rates, we acquire 2-D images at \( z \)-positions that are equally spaced throughout the rheometer gap (Fig. 1b). In the case of 2-D imaging, where particle locating and tracking is not possible, time-resolved spatial cross-correlations are calculated between pairs of consecutive images at a given \( z \)-position to quantify the spatially-resolved particle-scale velocities \( v(x, y) | z \), which are compared against direct particle tracking. The averaged velocities are calculated as a function of \( z \) to determine the velocity profiles at each \( \dot{\gamma} \) and obtain the local shear rate \( \dot{\gamma}_l \); this analysis is performed for two different volume fractions and we present data for \( \phi = 0.70 \) (see Supplementary Information).

The flow curves for our emulsions exhibit HB rheology, with \( n \approx 0.51 \) for \( \phi = 0.70 \) and a corresponding dependence on \( \sigma_y \) that increases with \( \phi \) (see Fig. 2 inset). After removing the background flow in the confocal images, we obtain a vector displacement field, with components \( \Delta x \) and \( \Delta y \), of local fluctuations in the shear frame. In Fig. 2h, we plot the mean square displacement (MSD) \( \langle \Delta r^2 \rangle = \langle \Delta x^2 + \Delta y^2 \rangle \), as a function of the accumulated strain \( \Delta \gamma = \dot{\gamma} \Delta t \), over a wide range of applied shear rates. At the highest shear rates \( (\dot{\gamma} \geq 1 \, s^{-1}) \), the system exhibits super-diffusive behavior at small \( \Delta \gamma \), followed by a diffusive regime, whereas at lower strain rates the \( \langle \Delta r^2 \rangle \propto \Delta \gamma \) over the entire range. The distributions of droplet displacements for each \( \Delta \gamma \) are characterised by non-Gaussian statistics for all strains. The self-part of the van Hove correlation function computed for strain windows \( \Delta \gamma = 0.02 \) and \( \Delta \gamma = 0.10 \), shown in Figs. 2b,c indicates
that, in addition to the overall non-Gaussianity for all $\Delta \gamma$ and each $\dot{\gamma}$, another distinctive feature arises. At large $\Delta \gamma$, the distributions exhibit exponential tails for all $\dot{\gamma}$, whereas at small $\Delta \gamma$ we can clearly distinguish a power-law tail for the distributions measured at low $\dot{\gamma}$. These observations indicate that the underlying microscopic motion is far from the simple picture of shear induced diffusive motion. Moreover, the data also highlight distinctive features that depend strongly on the imposed shear rate. Interestingly, the exponential tails are reminiscent of correlated and heterogeneous dynamics in supercooled liquids\textsuperscript{25}. The power law tails, on the other hand, suggest a closer connection with plastic rearrangements taking place in an elastic background, and that the elasticity of the jammed solid plays a significant role in the limit of quasi-static shear\textsuperscript{21,26,27}.

One of the major challenges associated with experiments of jammed matter is that the characteristic length and time scales rapidly exceed the resolution of fast confocal microscopy methods. To overcome this, we employ complementary three-dimensional numerical simulations of polydisperse spheres that interact through a short ranged repulsion and are subjected to shear deformations. Our simulations are guided by the details of the experimental system where the emulsion droplets are generally stable to both ripening and coalescence and are nearly athermal in the range of $\phi$ explored. We utilise the WCA form of the truncated Lennard-Jones potential, defined as

$$U(r) = 4\epsilon \left[ \left( \frac{\sigma_{ij}}{r_{ij}} \right)^{12} - \left( \frac{\sigma_{ij}}{r_{ij}} \right)^6 \right] + \epsilon, \text{ for } r_{ij} \leq 2^{1/6} \sigma_{ij}, \text{ else } U(r_{ij}) = 0$$

defines the distance between the center of particles $i$ (with diameter $\sigma_i$) and $j$ (with diameter $\sigma_j$) at contact, and $\epsilon$ defines the energy unit, independent of particle diameter. We choose $\sigma_i$ from a Gaussian distribution with a variance of 10%. The numerical sample consist of 97556 particles at $\phi \sim 70\%$, which corresponds to a cubic simulation box dimension of $l = 42\sigma$. The finite shear rate
simulations were carried out solving, with the Lees-Edwards boundary conditions, the Langevin equation of motion

\[ m \frac{d^2 \vec{r}_i}{dt^2} = -\zeta \left( \frac{d\vec{r}_i}{dt} - \dot{\gamma} z_i \vec{e}_x \right) - \nabla \vec{r}_i U, \]  

(1)

where \( \dot{\gamma} \) is the applied shear rate, \( m \) is the particle mass, \( \vec{r}_i \) is the position vector of the particle \( i \), \( z_i \) is the \( z \)-coordinate of particle \( i \), \( \vec{e}_x \) is the unit vector along the \( x \)-axis, \( \zeta \) is the damping coefficient and \( m/\zeta = 2.0 \). \( \nabla \vec{r}_i U \) defines the force to interactions between the particle. \( \dot{\gamma} \) is expressed in units of \( \tau_0^{-1} \), where \( \tau_0 = \sigma \sqrt{m/\epsilon} \) is the time unit. All data presented are in the steady state flowing regime (see Methods).

The flow curve obtained from the simulations at \( \phi = 0.70 \) follows the HB form with a flow index \( n \approx 0.6 \) (Fig. 3a inset). We note that the shear stress at \( \dot{\gamma} = 10^{-4} \tau_0^{-1} \) does not vary much in comparison with the quasi-static simulations data (see Fig. 3a inset), which represents the zero shear rate limit.

To strengthen the connection between our experiments and simulations, we perform an identical analysis of simulated displacement statistics by computing the MSD and the self-part of the van Hove correlation functions (see Fig. 3). To calculate the non-affine displacements and correlations, we divide the simulated emulsion into slices of thickness \( 2\sigma \) along the \( z \)-direction and average the data over each slice. Each slice contains approximately 4000 particles (see Supplementary Information). Remarkably, for conditions equivalent to those shown in Fig. 2, the simulations capture all of the essential features found in the experimental results. Most notably, for small \( \Delta \gamma \) the MSD obtained in the simulations exhibits a crossover from ballistic to diffusive motion with
increasing $\dot{\gamma}$, while the van Hove functions exhibit the same change from exponential to power law behavior with decreasing $\dot{\gamma}$. The striking similarities in these essential microscopic metrics provide us enhanced confidence that other particle-scale features gleaned from our simulations will allow us to draw physically relevant conclusions about these materials.

The microscopic dynamics are explored in the numerical simulations through an analysis of the spatio-temporal correlations of the droplet displacements. We measure the displacement correlations $S_U$, and the correlations in the deviation from the average displacement $S_{\delta U}$, where $\vec{U} = U_x \hat{i} + U_y \hat{j}$ is the displacement vector of a droplet in the $x,y$-plane and $\delta U = |U - \langle U \rangle|$ is the average droplet mobility, (see Methods). Completely correlated (anti-correlated) displacements give $S_U = 1.0$ ($S_U = -1.0$). $S_{\delta U} = 1.0$ if the fluctuations in displacements are perfectly correlated and both functions go to zero for uncorrelated motion. $S_U$, computed at fixed $\Delta \gamma$ as a function of distance $r$ from a reference droplet, can be decomposed into longitudinal $S^L_U$ and transverse $S^T_U$ components (see Methods).

We compute $S_U(\Delta \gamma, \dot{\gamma})$ and $S_{\delta U}(\Delta \gamma, \dot{\gamma})$ at $\dot{\gamma} = 10^{-1}$ and $10^{-4}\tau_0^{-1}$ over a range of $\Delta \gamma$. In each case, we extract a correlation length $\xi$ by fitting the data with an exponential function and averaging the fit parameters over the $z$-direction. At high shear rates, and for all strain values, both correlations functions decay exponentially. For clarity, only $S_U$ is shown in Figs. 4a and b see Supplementary Information for $S_{\delta U}$. The corresponding correlation lengths are shown in Fig. 4c.

Interestingly $S^L_U$ is the dominant correlation component within the super-diffusive regime of the MSD indicating that the non-affine motion in this regime is strongly correlated in direction (see
Fig. 4d) The correlation length $\xi_{SU}$ grows with an increase of $\Delta \gamma$ from $2\sigma$ to $5\sigma$ in the same strain interval (Fig. 4c). Overall, these results suggest that at high shear rates and small strains, corresponding to the super-diffusive regime, the motion is localised to small groups of droplets that move in linear clusters whose size increases with $\Delta \gamma$; this behavior also carries over into the diffusive regime, where $S_U^T \simeq S_U^L$ (see Fig. 4d) and persists down to shear rates $\dot{\gamma} \to 10^{-2}\tau_0^{-1}$. Elementary flow events of this type are reminiscent of the string-like motion in the dynamical heterogeneities of supercooled liquids (see Supplementary Movies M3-M8).

When $\dot{\gamma} < 10^{-2}\tau_0^{-1}$ (Fig. 4b), there is a qualitative change in the nature of the elementary rearrangement events. At small $\Delta \gamma$, both $S_U$ and $S_{\delta U}$ show an exponential initial decorrelation that is followed by a power-law decay. The microscopic picture that emerges from these data is that at low $\dot{\gamma}$, the droplet rearrangements underlying the macroscopic flow may resemble plastic-like yielding regions embedded in an elastic medium. The picture is confirmed by the observed power law tails in the self part of the van Hove distributions (see Figs. 2 and 3). At these low deformation rates, $S_{\delta U}$ is significantly larger than $S_U$ at a fixed length scale and the related correlation length is significantly larger, $\xi \simeq 10\sigma$, roughly 25% of the total system size (Fig. 4c). These observations quantitatively indicate that the droplet rearrangements involve directionally cooperative mobile clusters that are embedded in an immobile elastic background. The origin of the large error bars in the correlation length at these low rates point to large fluctuations in the instantaneous velocity profiles, revealing a flow behavior that is both inhomogeneous and intermittent. At larger $\Delta \gamma$, a larger percentage of droplets become mobile and we measure a dramatic drop in the directional correlation length $\xi_{SU}$, whereas $S_{\delta U}$ continues to show long range correlations. These features clearly
indicate that droplet dynamics are highly dependent on shear rate and that local rearrangements have a fundamental different structure at low $\Delta \gamma$. The analysis of the spatio-temporal correlations indicate that their range and intensity change qualitatively with the rate. The two results together suggest that the change in the spatio-temporal correlations may be at the origin of (or must be coupled to) the differences in the elementary flow events when the material is sheared at different rates. Elaborating further, the differences and dependence of the microscopic non-affine dynamics with the imposed shear rate are an essential component of the non-linear dependence on the rate of the flow curve.

The non-affine displacement maps obtained at fixed shear rates qualitatively support our conclusions. Representative maps for experiments and simulations are shown in Fig. 5, where the colouring is based on the magnitude of non-affine displacements. At high $\dot{\gamma}$, we observe relatively small displacements that are rather spatially homogeneous (see Figs. 5a,c), while at lower values, the displacements become highly localised, bursty and heterogeneous (see Figs. 5b,d and movies M1 and M2 in the Supplementary Information).

Our results provide a first glimpse into the non-equilibrium flow properties of technologically significant materials by coupling the microscopic dynamics to the macroscopic flow behavior. We unravel particle scale rearrangements and spatio-temporal correlations that are directly connected to the imposed rate of shear deformation. The steady-state response of unjammed emulsions, defined by the bulk rheological flow curve of the material, is associated with changes in the nature of the elementary flow events that are governed by the onset of microscopic flow. When the
flow is parameterised by the local accumulated strain, two distinct and rate dependent microscopic mechanisms are observed as the initiators of stress relaxation and yielding. Correlated string-like motion, a signature of dynamical heterogeneities in liquids, is found in the flowing state well above the yield stress. In contrast, near the yield stress, the primary mechanism for stress relaxation is localised plastic bursts embedded within an elastic continuum. These bursts exhibit long range spatio-temporal correlations and are strongly reminiscent of the local elastically interacting rearrangements that underlie plasticity in amorphous solids, referred to as shear transformations. The distinct and rate dependent nature of these elementary excitations is a fundamental starting point for the development of microscopic theories and constitutive models. We provide a quantitative framework for the differences in the spatio-temporal patterns of the correlations in the microscopic dynamics, which have themselves a distinct and rate dependent structure. The new insight gained is a first 3-D microscopic picture of the spatial correlations in the flow and of the non-linear rheology of soft jammed materials.

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Methods

Sample preparation and data acquisition in experiments. A highly polydisperse emulsion at oil volume fraction $\phi = 0.7$ was initially formed by mixing silicone oil (50 cSt, from Clearco Products Co.) with a 10 mM solution of sodium dodecyl sulfate (SDS) in water/glycerol (9:1, by volume) with a laboratory disperser (IKA Ultra Turrax T25 basic). We fractionated the sample using an iterative creaming technique that takes advantage of the oil having a lower mass density than the continuous phase. Rather than relying on depletion from excess surfactant to flocculate droplets of a certain size as in the published technique, we found that the relatively large droplets of interest creamed in a reasonable time by themselves, so the process was carried out at 10 mM SDS. After 9 steps, the selected droplets had an average diameter of 6.0 $\mu$m with a polydispersity of 0.15, as determined by confocal imaging. For these experiments, large droplets were chosen so that small displacements (relative to the average droplet diameter) could be resolved. However, even larger droplets would require too large a gap (to avoid boundary effects) and would be more fragile (due to their low Laplace pressure).

In preparation for imaging, we index matched the emulsion with glycerol to obtain a transparent sample. The resulting continuous phase was dyed with fluorescein sodium salt at a 3 mM concentration. The SDS concentration was boosted to 50 mM, which we found increased the sta-
bility of the emulsion, particularly under shear with a high glycerol concentration. Even at the elevated surfactant levels, there was significant droplet coalescence for shear rates $\dot{\gamma} > 10 \, s^{-1}$. While 50 mM is well above the critical micelle concentration, we could not detect a dependence of the rheology on the SDS concentration, and thus suspect any depletion force had a minimal effect. The samples were compressed to the desired volume fraction $\phi$ with centrifugation, with the value of $\phi$ determined by boiling the continuous phase off.

The sample is placed on a glass coverslip at the bottom of a metal cup that is rigidly mounted to the rheometer. Optical access for the microscope is provided from below by a small hole on the underside of the cup. The flow experiments were performed using a parallel plate rheometer tool with a diameter $d = 25 \, \mu m$. The rheometer gap was set to a final value of $h = 100 \, \mu m$ by lowering the tool slowly over several minutes, while rotating at 0.1 rpm. This kept the normal force low, as monitored by the rheometer, and minimised damage to the sample. On the bottom plate coverslip, we lithographically defined a square grid of $10 \, \mu m$ wide, $5 \, \mu m$ high posts (spaced by $10 \, \mu m$) made from SU-8, a negative photoresist, to minimise surface slip. The posts were oriented in a consistent way with respect to the imaging axes. A similar slide was affixed to the rheometer tool for the same purpose. While some slip was present at high shear rates, determination of the local strain rate $\dot{\gamma}_l$, for example, came from direct optical measurements taken over the full gap, without regard for the nominal rheometer settings.

Images were acquired with a $63 \times$, 1.2 numerical aperture, water-immersion objective. Water was chosen over oil as the immersion fluid, as it produces smaller deflections of the coverslip with a better index-match to the sample. The objective was positioned at a radial distance of $d/3$ from
the central rheometer axis, a location where the local strain is equal to the plate-averaged strain.

When performing the flow measurements, the rheometer was set to a particular shear rate \( \dot{\gamma}_0 \), as referenced to the edge of the tool. In presenting the microscope data, we will refer to the shear rate at the radius where the images were acquired, \( \dot{\gamma} = 2\dot{\gamma}_0/3 \). After setting \( \dot{\gamma}_0 \), the imaging was not initiated until the shear stress stabilised. We also attempted to avoid imaging volumes where large coalesced drops were located. For \( \dot{\gamma} = 10^{-3} \text{s}^{-1} \) and below, it was possible to take 3-D stacks with the desired interval in time. At \( \dot{\gamma} = 10^{-2} \text{s}^{-1} \), it was only possible to acquire 3-D stacks that covered a fraction of the gap, so multiple stacks were acquired to cover the full sample. For higher rates, we acquired a rapid set of 2-D slices at fixed \( z \), incrementing the \( z \) position between sets. The time spacing between frames was set to \( 0.03/\dot{\gamma} \), which corresponds to a motion of 3 \( \mu \text{m} \) of the tool at the imaging location. The image size was 145 \( \mu \text{m} \times 145 \mu \text{m} \) with a resolution of 512 \( \times \) 512. The exception is for the data at \( \dot{\gamma} = 10 \text{s}^{-1} \), where, due to limitations of the microscope, the time spacing was set to \( 0.17/\dot{\gamma} \) with the resolution reduced to 512 \( \times \) 192. In all cases, we acquired at least 150 frames at each setting of \( z \).

Sample preparation, shearing protocol, steady state analysis in simulations. The initial sample on which rheological studies are carried out is prepared using the following protocol. A FCC crystal containing 97556 LJ particles at a desired volume fraction of 70\% (with a system dimension of \( l_x = l_y = l_z = 42.17\sigma \)) and a continuous polydispersity of 10\% is initially melted and equilibrated for 50000 molecular dynamics (MD) steps, with a timestep of \( \Delta t = 0.001 \), at a high temperature of \( T = 5.0\epsilon/k_B \). In the melt we do not find any signature of crystallinity from the FCC initialisation. For the purpose of measuring crystallinity, we measure the local orientational
ordering $q_{6}^{[35]}$. Note that the orientational ordering computation requires calculating the nearest neighbours for a reference particle. For this purpose we define all the particles within the first Voronoi shell as neighbours. The equilibrated melt is subjected to a temperature quench, wherein the system at $T = 5.0\epsilon/k_B$ is lowered to a temperature of $T = 0.001\epsilon/k_B$ at cooling rates of $5 \times 10^{-4}$. We then perform conjugate gradient minimisation to take the system to the zero temperature limit. In this configuration, we analyse the local orientational ordering and do not find signatures of crystallisation. The finite shear rate rheology studies were carried out by performing the shear deformation simulation using Lees-Edwards boundary conditions and solving the Langevin equation of motion (see Eqn. 1). The damping coefficient is chosen such that, after the elementary shear deformation, the system relaxes, within a accessible time to a potential energy (PE) which is comparable to the PE we would obtain if we performed a conjugate gradient minimisation.

We monitor the virial stress as well as the virial pressure evolution in the system with shear strain to make sure the system reaches a steady state. We find that even though the stress seems to have reached steady state at around 50% strain, the pressure is evolving. As the pressure saturates at roughly 300%, the system shows a homogeneous flow profile. We shear strain an additional 200% to obtain good statistics for the observables. Even though the velocity profile shows homogeneous flow, we find noticeable fluctuations in velocity profile, over small strain windows ($\sim 1\%$). To account for these fluctuations we compute the local shear rates $\dot{\gamma}_l = dV_z/dz$. The local rearrangement of particles is characterised by the non-affine mean square displacement (MSD). To compute non-affine displacement, we divide the system into slices of thickness $2\sigma$ along the $z$-direction.
Each slice contains approximately around 4000 particles. In each of these slice, we compute the shear frame displacements which are given by

\[
\Delta x_s = x(\gamma_2) - x(\gamma_1) - V_x(z) \Delta t
\]

\[
\Delta y_s = y(\gamma_2) - y(\gamma_1),
\]

where \(\Delta x_s\) and \(\Delta x_s\) are the displacements of the particles in shear frame measured as a function of local strain difference \(\Delta \gamma = \dot{\gamma} \Delta t\), \(\dot{\gamma}\) is the slab specific shear rate measured from the velocity profile, \(x(\gamma_\alpha)\) and \(y(\gamma_\alpha)\) are the coordinates of the particles at a particular accumulated shear strain of \(\gamma_\alpha\) and \(V_x(z) \Delta t\) measures the local accumulated affine displacement, where \(V_x(z)\) is the instantaneous velocity calculated over the time difference \(\Delta t\). The mean square displacement is computed as \(\langle \Delta r^2 \rangle = \langle \Delta x_s^2 + \Delta y_s^2 \rangle\). The MSD as a function of \(\Delta \gamma\) is averaged over different planes along the gradient direction. The MSD, for a wide range of strain, shows isotropic behaviour.

**Displacement correlation function** The displacement correlations associated with droplet displacements \(S_U\) and mobility (or deviation from average displacements) \(S_{\delta U}\) are defined as

\[
S_U(\Delta r, \Delta \gamma) = \frac{\langle \vec{U}_i \cdot \vec{U}_j \rangle}{\langle \vec{U}^2 \rangle}
\]

\[
S_{\delta U}(\Delta r, \Delta \gamma) = \frac{\langle \delta U_i \cdot \delta U_j \rangle}{\langle \delta U^2 \rangle}
\]

where \(\vec{U} = U_x \hat{i} + U_y \hat{j}\) is the displacement vector of a droplet in the shear frame, in the flow-vorticity plane and \(\delta U = |U - \langle U \rangle|\) is the fluctuation in the displacement of a droplet over the average displacement, which also known as mobility. The \(S_U\) is a vectorial correlation function which is indicative of directional correlation between the droplets. This function can be further
decomposed into longitudinal and transverse correlations given by

\[ S^L_U = \frac{\langle U^L_i U^L_j \rangle}{\langle U^2 \rangle} \]
\[ S^T_U = \frac{\langle \tilde{U}^T_i \cdot \tilde{U}^T_j \rangle}{\langle U^2 \rangle} \]

(4)

where \( U^L_{ij} = \tilde{U}_{ij} \cdot \hat{R}_{ij} \) is the displacement along the separation vector \( \hat{R}_{ij} \) and \( \tilde{U}^T_{ij} = \tilde{U}_{ij} - U^L_{ij} \hat{R}_{ij} \) is along perpendicular direction to the separation vector. The longitudinal and transverse correlation function are normalized such that \( S^L_U + S^T_U = S_U \).

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Supplementary Information is available in the online version of the paper.

Competing financial interests

The authors declare no competing financial interests.
**Figure 1**  Experimental system details. a, Cut-away diagram of the confocal rheometer system. Samples are loaded in the gap between the upper tool (yellow) and the lower glass plate (blue). b, Confocal micrograph of the $x, y$-plane showing a cross section of the emulsion at volume fraction $\phi = 0.70$, scale bar represents 20$\mu$m.

**Figure 2**  Flow curve and shear frame displacement from experiments. a, The shear-frame MSD measured in the $x, y$-plane for shear rates $10^{-3}$ s$^{-1}$, $10^{-2}$ s$^{-1}$, $10^{-1}$ s$^{-1}$, 1.0 s$^{-1}$ and 10.0 s$^{-1}$. The gray solid lines are indicative of diffusive behaviour. (inset) The shear stress $\sigma$ as a function of the shear rate $\dot{\gamma}$ measured at $\phi = 0.70$ and $\phi = 0.78$ and the solid lines represent Herschel-Bulkely fit, with flow indices of $n = 0.51$ and $n = 0.45$. The linear viscoelastic modulus of these emulsions is $G' = 180, 360$ Pa for $\phi = 0.70, 0.78$. The experimental van Hove correlation function $G_S(r, \Delta \gamma)$ is computed at $\phi = 0.70$ for two different strain windows b, $\Delta \gamma = 0.02$ and c, $\Delta \gamma = 0.1$ for two different shear rates 1.0 s$^{-1}$ and $10^{-3}$ s$^{-1}$. For the purpose of clarity we have shifted the abscissa of the distribution function by a constant factor of 0.2 for $10^{-3}$ s$^{-1}$. The dot-dashed line is the best fit with a Gaussian, the solid line represents the best fit with an exponential, whereas the dashed line is the best fit with a power law.

**Figure 3**  Flow curve and shear frame displacement from simulations. a, The non-affine MSD computed in the $x, y$-plane for shear rates $10^{-4}$ $\tau_0^{-1}$, $10^{-3}$ $\tau_0^{-1}$, $10^{-2}$ $\tau_0^{-1}$ and $10^{-1}$ $\tau_0^{-1}$ for simulations. (inset) Simulation flow curve. The shear stress $\sigma$ as a function of the
shear rate \( \dot{\gamma} \) measured at volume fraction \( \phi = 0.70 \) with solid lines showing a Herschel-Bulkely fit, with a flow index of 0.6. The solid square data obtained from the quasi-static simulations which corresponds to a zero shear rate condition. The \( G_s(r, \Delta \gamma) \) computed for \( b \), \( \Delta \gamma = 0.005 \) and \( c \), \( \Delta \gamma = 0.005 \) for shear rates \( \dot{\gamma} = 10^{-1} \tau_0^{-1} \), and \( \dot{\gamma} = 10^{-4} \tau_0^{-1} \). For the purpose of clarity we have shifted the abscissa of the distribution function by a constant factor of 0.2 for \( 10^{-4} \tau_0^{-1} \). The dot-dashed line is the best fit with a Gaussian, whereas the solid and dashed lines correspond respectively to the best fit with an exponential and power law.

**Figure 4** Displacement correlation function from simulations. a, At high shear rate \( \dot{\gamma} = 10^{-1} \tau_0^{-1} \) and b, low shear rate \( \dot{\gamma} = 10^{-4} \tau_0^{-1} \), the spatial correlation function \( S_U \) associated with droplet displacements is shown for shear strains \( \Delta \gamma = 0.005 \) (black), \( \Delta \gamma = 0.05 \) (red) and \( \Delta \gamma = 0.1 \) (green). c, the correlation length obtained from \( S_U \) as a function shear strain, for shear rates \( 10^{-4} \tau_0^{-1} \), \( 10^{-3} \tau_0^{-1} \), \( 10^{-2} \tau_0^{-1} \) and \( 10^{-1} \tau_0^{-1} \). d, the ratio of longitudinal to transverse displacement correlation function measured at a distance around \( 1\sigma \), for accumulated strains 0.005, 0.05, 0.1 and 0.2 as a function of shear rate.

**Figure 5** Displacement maps for experiments and simulations. Experimental trajectory snapshots for a, \( \dot{\gamma} = 3 \times 10^{-3} \text{ s}^{-1} \) and b, \( \dot{\gamma} = 1.0 \text{ s}^{-1} \). For the low shear rate, the snapshot is obtained at strain interval of \( \Delta \gamma \approx 0.02 \) (bottom plane), \( \Delta \gamma \approx 0.01 \) (middle plane) and \( \Delta \gamma \approx 0.001 \) (top plane). For the high shear rate, the accumulated strain intervals are \( \Delta \gamma \approx 0.025 \) (bottom plane), \( \Delta \gamma \approx 0.015 \) (middle plane) and \( \Delta \gamma \approx 0.02 \) (top plane). The
rendered particle positions are approximately taken from the middle third of the rheometer gap. Simulation trajectory snapshots for slow, $c \dot{\gamma} = 10^{-4} \tau_0^{-1}$, and fast $d \dot{\gamma} = 10^{-2} \tau_0^{-1}$, at $\Delta \gamma = 0.001$. Only three different layers in the 3D system are highlighted for the purpose of clarity. The colour scheme in all snapshots represent non-affine displacement magnitude. The arrows represent the non-affine displacement vector. To highlight the localisation of displacement vectors, the length of the vectors are amplified by 5 times in experimental snapshots and by 40 times in simulation snapshots. Renderings are made using freely available software.\textsuperscript{31}