Complex Hydrodynamic Interactions and Stokes-Einstein Relation in Quasi-Two-Dimensional Colloidal Fluids

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Abstract

Long-ranged hydrodynamic interactions are intrinsic to diverse physical systems [1–7] and, in fact, are the most prominent ones for low Reynolds number systems, such as flows involving micro-organisms and particle-laden suspensions [6–9], and protein diffusion across membranes [10, 11]. While hydrodynamics is well-understood in the far-field (asymptotic limit) [5, 7, 12], it has been less explored in the near-field [13]. We experimentally elucidate the hydrodynamic correlations in quasi-two-dimensional colloidal fluids in the near-field, uncovering a new filling-the-void mode formed by the combination of well-known drag and anti-drag hydrodynamic dipolar modes. These modes superpose, creating a rugged potential landscape for the diffusing colloids that leads to the violation of the Stokes-Einstein relation (SER), whose validity is believed to be a hallmark feature of liquids [14]. We find pair-wise colloid interactions unravel complex emergent dynamics due to hydrodynamics, which in contrast to dynamics emanating from single particles are observed to be anisotropic. Remarkably, insights gleaned enabled us to pick out
the directions unique to each pair, in which hydrodynamic correlations are the weakest, thereby recovering the validity of SER along these. Our experiments revealing the effects of distinct modes of hydrodynamic interactions on SER will have significant ramifications for fluids’ underlying dynamic structure and transport properties in two dimensions.

The microscopic underpinnings of hydrodynamic interactions between suspended particles at low Reynold numbers continue to be an area of intense research in condensed matter physics \[4–19\]. As the effects of inertia can be neglected, the suspended particles interact over lengthscales much longer than their sizes, affecting their transport \[5, 7, 12\]. In both the near and far-field, the particle interaction in pairs become strongly dependent on the spatial confinement of the fluid and boundary conditions \[20, 21\]. For example, in two-dimensions (2D), both the analytical/numerical predictions of the asymptotic far-field solutions and experimental findings highlight a dipolar flow profile with longitudinal drag and transverse anti-drag coupling between suspended particles \[5, 7\]. This contrasts the monopole-like interactions with drag component along both the directions in three-dimensions (3D) \[19, 20\]. Moreover, with the emergence of structural features in the near field as particulate density increases, the longitudinal drag correlations show modulations in phase with the structural signatures \[22\]. However, the nature of transverse anti-drag coupling in the near-field, for example, its possible phase difference with the longitudinal correlations, let alone influence of hydrodynamics on the well-known transport quantifier relations like the Stokes-Einstein relation, is yet to be elucidated. Note that the presence or absence of a phase difference between different (inherent/external) control parameters leads to rich and complex emergent dynamics in various physical systems \[23–26\].
In addition to numerous applications, colloids are valuable model systems to peek into diverse physical phenomena at single-particle resolution [27–31]. Here, we employed optical video microscopy to probe the hydrodynamic interactions, focusing on near-field correlations, in colloidal fluids in quasi-two-dimensional (q2D) geometric confinement (Methods). Experiments were performed with polystyrene latex beads with diameter, $\sigma = 1.04 \, \mu m$, at seven different particle packing area fractions, $\phi$, in the range $0.15 \leq \phi \leq 0.61$; well below the hexatic phase transition for 2D packings [32]. The displacement distributions of particles, irrespective of $\phi$, are observed to be spatially symmetric about their mean position (Fig. 1a) and are devoid of any information about the hydrodynamics. The nature of hydrodynamic interaction (HIs) is revealed only when the conditional probability distribution of displacements for the particles in pairs is measured in the body frame of reference of the respective pairs. In the body frame, longitudinal ($L$) and transverse ($T$) axes for particles in a pair separated by distance, $r$, are defined along and perpendicular to the line joining the particles in the pair at the initial time, $t_0$, respectively. Our data clearly show well-known hydrodynamic dipolar modes [5, 7]; co-diffusion (drag) along the longitudinal (SI Fig. S1 & SI Video S1) and anti-symmetric (anti-drag) correlations along the transverse direction (SI Fig. S1 & SI Video S2). Moreover, for $r \sim 1.1 \sigma$, we also observe the linear combination of drag and anti-drag modes and identify it as filling-the-void mode (Fig. 1b). Here, one particle of the pair moves circumferentially around the other, eventually filling the void created by the motion of the other particle (see SI Video S3). To the best of our knowledge, this mode has not been observed in experiments before. These modes can also be seen in the ensemble-averaged hydrodynamic flow profile of particle pairs separated by vector $\mathbf{r}(r, \theta)$ at $\phi = 0.15$ (Methods, Fig. 1c).
The longitudinal hydrodynamic correlation, $H_L$, for particles in the pairs located at $\{r', r + r'\}$ and separated by distance, $r$, can also be quantified as $H_L(r,t) = \langle \Delta r_L(r',t) \Delta r_L(r' + r, t) \rangle_{r',t_0}/D^*$ [5, 7]. Here, $\Delta r_L$ is the displacement of particles in a pair along $L$ over the lag time, $t$, the averaging, $\langle \rangle$, is performed over $t_0$ and $r'$, and $D^*$ is the monomer self-diffusivity. A similar definition is used for the transverse mode, $H_T$. Figure 1 d shows $H_L$ and $H_T$ versus $r$ for two different $\phi$. At low $\phi$, $\phi = 0.15$, $H_L$ and $H_T$ mimic dipolar decay profile in the far-field, i.e., decay as $1/r^2$. The positive correlations in $H_L$ and negative in $H_T$, albeit with different magnitudes, is a defining signature of q2D rigid confinement (SI Fig. S2). At higher density, $\phi = 0.61$, concomitant with the emergence of local structural features in the near-field (SI Fig. S3), non-monotonic spatial modulations appear in $H_L$ and $H_T$ and they begin to deviate from the dipolar form. Nevertheless, the far-field, $r > 8\sigma$, decay profiles continue to be dipolar (Figure 1 d). As drag is sensitive to underlying structure of the fluid, modulations in $H_L$ are in phase with structural features (SI Fig. S3) [5, 22]. Surprisingly, however, anti-drag, $H_T$, is found to have a spatial phase difference of around $0.25\sigma$ with $H_L$ (inset to Fig. 1 d), which is also revealed in $Z_{rel}(r,t) \equiv \langle r(t + t_0) \rangle_{r',t_0} - 1$, defined for each pair. Since anti-drag leads to pair rotation and eventually their separation, it is succinctly captured by $Z_{rel}$. At the highest $\phi$ studied, $\phi = 0.61$, $Z_{rel}$ shows in phase oscillatory decaying modulations with $H_T$ in the near-field (Fig. 1 e), and has a spatial phase difference of around $0.25\sigma$ with $H_L$. Note that the positive coupling in $H_T$ for $r < 2\sigma$ at $\phi = 0.61$ is due to multi-body effects and can be reconciled by measuring only the pairwise transverse correlations for certain configurations that lowers multi-body hydrodynamic effects (SI Fig. S4).

Taken together, the insights offered by $H_L$ and $H_T$ (and $Z_{rel}$) allow us to construct the spatiotemporal evolution of a pair with $r(t)$ (Fig. 1 f). The
particles in a pair separated by $r \sim 1.0\sigma$ diffuses to $r \sim 1.25\sigma$ due to the paucity of accessible configurations (because of hard-sphere interactions). Around $r \sim 1.25\sigma$, pairs are the most stable as indicated by the minima in $Z_{rel}$ (Fig. 1 e) and drag is dominant. Once the separation between particles of the pairs is $r \sim 1.5\sigma$, rotation of the particles around the transverse direction leads to their radial separation, which becomes maximum around $r \sim 1.75\sigma$, and is the most unstable configuration for a given pair. With time as $r$ increases, anti-drag weakens and drag kicks in and it once again becomes prominent at $r \sim 2.0\sigma$, and the cycle repeats (Fig. 1 f).

The emergent spatiotemporal mobility landscape discovered above for spatially confined colloidal suspensions will eventually lead to fluctuations in the local viscosity (or diffusivity of colloid-pairs). As a case study, here, we probe the Stokes-Einstein relation (SER) for q2D colloidal fluids, which has come into the limelight in recent years due to discordant observations. Per SER, 

$$D = \frac{k_B T}{6\pi \eta (\sigma/2)},$$

where $k_B T$ is the thermal energy and $\eta$ is the viscosity of suspending fluid. Considering structural relaxation time, $\tau_\alpha$, as a proxy for $\eta$ [33, 34], $D \propto \tau_\alpha^{-\xi}$, with SER exponent, $\xi = 1$, a hallmark feature of liquids [14]. Surprisingly, recent computer simulations and experiments have observed $\xi > 1$ for 2D fluids [35–37]. The unusual behaviour of $\xi$ has alluded to the presence of long-wavelength Mermin-Wagner fluctuation in 2D liquids [38]. In fact, taking cues from solids [39, 40], these correlations have been removed by considering the relative motion of particles with respect to their cages, which recover $\xi \sim 1$ [38]. This approach to filter out correlated motions naturally assume that $D$ and $\tau_\alpha$ are isotropic (no $\theta$-dependence of dynamics) in both near- and far-field.

Therefore, to check the possible influence of spatial phase difference between longitudinal and transverse modes of HIs on the dynamics of q2D
colloidal fluids, we measure $D(r, \theta)$ and $\tau_\alpha(r, \theta)$ (Methods and SI Fig S5). At low $\phi$, $\phi = 0.15$, while diffusion of pairs are isotropic and homogeneous for all $r$ (Fig. 2a & c), pairs relaxation for $r < 2.5\sigma$ are found to be anisotropic (Fig. 2b & d). One the one hand, since drag leads to co-diffusion of particles in pairs, $r$ at which $H_L$ is most-prominent ($r \sim 1.0\sigma$), pairs will take longer to relax along the longitudinal direction than along the transverse one, i.e., $\tau^L_\alpha(r \sim 1.0\sigma) > \tau^T_\alpha(r \sim 1.0\sigma)$ (Fig. 2b & d). On the other hand, anti-drag helps in pair rotation and separation, and hence, $r$ corresponding to points when $H_T$ (or $Z_{rel}$) is comparatively stronger ($r \sim 1.75\sigma$), an opposite trend is expected and observed (Fig. 2b & d). With the increase in particle packing area fractions, concomitant with the appearance of structural features in the near-field, diffusion of pairs become spatially inhomogeneous and are in phase with $H_L$ (Fig. 2e & g). However, pairs’ relaxation are found to be not only spatially inhomogeneous, but anisotropic also (Fig. 2f & h), with spatial phase lag of $\sim 0.25\sigma$ being reflected between $\tau^L_\alpha$ and $\tau^T_\alpha$ also (Fig. 2d & h).

Motivated by these observations, we next measured $D(r, \theta)$ and $\tau_\alpha(r, \theta)$ for all $\phi$ and focussed along longitudinal ($\theta = 0^\circ$) and transverse ($\theta = 90^\circ$) directions at different $r$ for testing the validity of SER. For each $r$, we used $D^{L,T}$ and $\tau^{L,T}_\alpha$ from all $\phi$ to extract $\xi^{L,T}$. Obviously, the variation of $D$ and $\tau_\alpha$ with $r$ and anisotropy in $\tau_\alpha$ along $L$ and $T$ are reflected in SER exponent, $\xi^L$ and $\xi^T$, respectively (Fig. 3). Strikingly, the spatial anisotropy and phase lag between $\xi^L(r)$ and $\xi^T(r)$ with $r$ disappear and become equal along different randomly chosen directions (SI Fig. S6), suggesting the unusual trend in $\xi(r)$ is due to the distinct modes of hydrodynamic interactions in 2D spatial confinement. The self-diffusivity of particles, $D^s$ and their relaxation, $\tau^s_\alpha$, that has been traditionally utilized hitherto, yields $\xi^s = 1.14 \pm 0.01$ (inset to Fig. 3); the spatial modulations in $\xi$ decay with $r$ and converge to $\xi^s$ in the far-field,
$r > 8\sigma$. In total, these observations indicate that competing modes of HIs are a fundamental feature of rigid spatial confinement that precludes any global correction to remove the correlated dynamics.

Finally, in the quest to reconcile the unusual behaviour of $\xi$ for q2D colloidal fluids and recover $\xi \sim 1$, we propose a simple approach based on pairwise interactions and their correlated displacements. SER’s validity ($\xi = 1$) is expected only for a purely random process (fluids) and not for (complex) correlated dynamics, as seen here. Nevertheless, the motion of the particles of pairs in the direction perpendicular to their centre-of-mass displacements is predominantly due to thermal fluctuations (Fig. 4 a). Hence, extracting $D^{CM\perp}$ and $\tau^{CM\perp}_\alpha$ along this direction should yield $\xi^{CM\perp} \sim 1$. Figure 4 c corroborates our arguments. In the far-field, $r > 8\sigma$, as spatial inhomogeneities in $H_L$ and $H_T$ disappear (Fig. 1 e), $\xi^{CM\perp}$ decays and saturates to $1.01 \pm 0.01$ (Fig. 4 c). In the near field, $r < 8\sigma$, $\xi^{CM\perp}$ oscillates around 1. Along with the phase difference between $H_L$ and $H_T$, huge spatial inhomogeneities with $r$ in the near-field imply that hydrodynamic correlations cannot be completely filtered out (Fig. 1 e). Interestingly, in this regime, too, $\xi^{CM\perp}$ at specific $r$, where hydrodynamic correlations are the weakest along the orthogonal direction to the centre-of-mass displacements of pairs, which corresponds to extrema of $Z_{rel}$ or $\frac{d(H_L+H_T)}{dr}$, $\xi^{CM\perp} \rightarrow 1$ (Fig. 4 b and SI Fig. S7). In the inset to Figure 4 b, $\xi^{CM\perp}$ at extrema of $Z_{rel}$ are compared to the corresponding value of $\xi^L$. Remarkably, the change in $\xi$ is enormous; for instance, $\xi^L(r = 1.25) = 1.33 \pm 0.02$ changes to $\xi^{CM\perp} = 0.99 \pm 0.02$, by just estimating $\xi$ in the directions in which the motions are uncorrelated and are predominantly due to thermal fluctuations, a prerequisite for ascertaining the validity of SER.

Our experiments highlight the importance of phase, in this case, presence, between the two hydrodynamic dipolar modes with contrasting magnitudes,
which are inherent to 2D spatially confined systems, on the structure and dynamics of the colloidal fluids. Thermal fluctuations randomize the hydrodynamic dipoles in passive fluids studied here, unlike in nematics [41], driven suspensions [7], and polar flocks [16]. Nonetheless, two-body interactions can still account for the complex spatially inhomogeneous and anisotropic dynamics of the fluids. It is intriguing to note that even \( D \) at low \( \phi \), \( \phi \leq 0.35 \), are anisotropic when extracted from short time duration, \( t < 20 \) s, timescales over which hydrodynamic interactions are significant (SI Fig. S8). At higher \( \phi \), the dynamics become mildly sub-diffusive over short timescales, precluding extraction of \( D(r, \theta) \), and hence, we cannot comment on whether \( D(r) \) continues to be anisotropic at these densities. Our studies demonstrate that the unusual magnitude of the Stokes-Einstein exponent \( \xi > 1 \) observed for 2D colloidal fluids is due to complex hydrodynamics in confined geometry; however, the origin of \( \xi > 1 \) in 2D simulations wherein hydrodynamics is absent is still not clear and could be due to long-wavelength Mermin-Wagner fluctuations [38]. These microscopic fundamental insights of hydrodynamics will profoundly impact numerous phenomena like cluster aggregation, translocation of proteins, nucleation and growth kinetics of crystal, active systems, and clogging and jamming of channels, especially in spatially constrained systems, and are worth re-examination.

Methods

**Experimental details.** We have used polystyrene microspheres of diameter \( 2\sigma = 1.04 \) µm with polydispersity of < 5% suspended in water. The particles were loaded into a wedge-shaped cell and allowed to sediment under gravity into the thin quasi-two-dimensional (q2D) region of the cells. Once a desired packing area fraction, \( \phi \), was achieved, the cell was allowed to equilibrate for
at least six hours before video microscopy. It was ensured that data for all \( \phi \)s were taken from the same region of the cell. The images, at each \( \phi \), were captured at 10 frames per second (fps) for 20 minutes. The trajectories of the particles were obtained using standard tracking algorithms \([42]\). The dynamic spatial resolution was found to be 20 nm. All the subsequent analysis were performed using in-house developed codes.

**Dynamics measurement.** Particles’ self-diffusivity, \( D^s \), and structural relaxation time, \( \tau^s_\alpha \), in the lab frame were measured from the mean squared displacements, \( \langle \Delta r(t)^2 \rangle \), and self-intermediate scattering functions, \( F_s(q,t) \) \([35]\) (SI Fig. S5). \( \langle \Delta r(t)^2 \rangle = \langle \frac{1}{N} \sum_{k=1}^{N} (r_k(t + t_0) - r_k(t_0))^2 \rangle_{t_0} \) and \( F_s(q,t) = \langle \frac{1}{N} \sum_{k=1}^{N} e^{i q \cdot (r_k(t + t_0) - r_k(t_0))} \rangle_{t_0} \). Here, \( N \) is the total number of particles, \( (r_k(t + t_0) - r_k(t_0)) \) is the displacement of \( k \)th particle over the lag time, \( t \), and the averaging, \( \langle \rangle \), were performed over \( t_0 \). For all the analyses presented in this study, the magnitude of probe wave-vector, \( q = 2\pi/a \), where \( a \) is the position of the first peak in the pair correlation function, \( g(r) \), at \( \phi = 0.61 \). The direction of \( q \) is chosen to be along \( x- \)axis. For the pair-dynamics measurement in the body frame, \( D(r, \theta) \) and \( \tau_\alpha(r, \theta) \) of particle-pairs were measured using similar definitions used for lab frame analysis. However, \( r \) is the pair-separation, and \( \theta \) is the angle between the chosen direction and the line joining the particles at \( t_0 \). \( q = \frac{2\pi}{a} \hat{\theta} \). While dynamics at low \( \phi \) is diffusive over the entire experimental duration for each \( r \) and \( \theta \), at higher \( \phi \) (\( \phi \geq 0.58 \)), dynamics are found to be diffusive only for \( t > 20 \) s. Therefore, for consistency, \( D \) at all \( \phi \) and each \( r \) and \( \theta \) studied here have been obtained for \( 20 \) s \( \leq t \leq 50 \) s. The decay of \( F_s(q,t) \) to \( 1/e \) is read-off as structural relaxation time, \( \tau_\alpha \), i.e., \( F_s(q,t = \tau_\alpha) = 1/e \).

**Hydrodynamic flow profile.** For the flow field, we first computed the displacements of particles \( \Delta r_k(t = 0.5s) \), and then plotted by rotating all the
displacements, \( \langle \Delta r_j(t = 0.5s) \rangle_{r,t_0,k \neq j} \), on the same plane such that \( \Delta r_k(t) \) is along the \( x \)-axis [12].

**Supplementary information.** This article contains supplementary files/videos.

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**Declarations**

- Authors declare no competing financial interests.
- Correspondence and requests of materials should be addressed to C.K.M.
Fig. 1 | Hydrodynamic modes and spatiotemporal evolution of pairs. **a**, Colormap of $P(\Delta r(t))$ for particles in pairs separated by an arbitrary $r$. **b**, Conditional $P(\Delta r(t))$ measured for the particle on the left of the pair when the particle on the right moves by $\geq 1.0\,\sigma$ over $t$ for $r = 1.1\,\sigma$ along $T$ depicted by yellow arrow. Solid red and white circles represent the mean positions of the particles at $t_0$ and $(t_0 + t)$, respectively. The displacement color map for **a** & **b** are normalized by the maximum displacements in each case. **c**, Polar colormap, $r(r,\theta)$, of hydrodynamic flow profile when the colloid at origin (solid red circle) moves towards the right (open red circle) over $t$. The dashed radial circles represent $r = \{2, 3, 4\}\,\sigma$. The direction of the field line, $\psi$, with respect to moving colloid at origin is also shown in the background. Representative red arrows, with their head and length represent the direction and strength, respectively, of the field. The measurements for **a**–**c** were performed at $\phi = 0.15$ and $t = 0.5\,\text{s}$. **d**, $H_L$ (solid symbols) and $H_T$ (open symbols) versus $r$ for $\phi = 0.15$ (red circles) and $\phi = 0.61$ (green squares). The black solid and dashed lines show $\pm A/r^2$ dependencies, respectively, where $A$ is a constant. The inset shows $H_T$ at $\phi = 0.61$. **e**, $Z_{rel}$ versus $r$ at $\phi = 0.61$ for $t = 0.5\,\text{s}$ (black squares), $t = 1.5\,\text{s}$ (red circles) and $t = 3.0\,\text{s}$ (blue triangles). The inset shows typical schematics depicting configurations corresponding to the peak position in $Z_{rel}$. **f**, Schematic construction of spatiotemporal evolution of a pair of particles due to hydrodynamics.
Fig. 2 | Elucidating the influence of hydrodynamics on transport quantifiers. Polar colormaps $D(r,\theta)$ at a, $\phi = 0.15$ and e, $\phi = 0.61$, and $\tau_\alpha (r,\theta)$ at b, $\phi = 0.15$ and f, $\phi = 0.61$ versus $r$ with colormaps shown on their left. The dashed radial circles are at $r = \{2, 3, 4, \ldots\} \sigma$. $D(r)$ at c, $\phi = 0.15$ and g, $\phi = 0.61$, and $\tau_\alpha (r)$ at d, $\phi = 0.15$ and h, $\phi = 0.61$ along $L$ and $T$ directions corresponding to $\theta = 0^\circ$ and $\theta = 90^\circ$, respectively. The errors bars in $D$ are from fittings.
Fig. 3 | Influence of hydrodynamics on Stokes-Einstein relation in near-field. SE exponents $\xi^L$ and $\xi^T$ versus $r$. The inset shows self-diffusivity of particles, $D_s$, versus their relaxation, $\tau_s^\alpha$; the solid line shows $D_s \propto \tau_s^{1.14\pm0.01}$ dependency. Black dashed and dotted lines at $\xi = -1.00$ and $\xi = -1.18$ depict the ideally expected and asymptotic obtained values of $\xi$, respectively. Top and bottom panels shows representative $D^L$ and $D^T$ versus $\tau^L_\alpha$ and $\tau^T_\alpha$, respectively, for different $r$ as shown in the figures. The solid lines depict linear fits to determine $\xi$. The errors bars are from fittings.
Fig. 4 | Reconciling Stokes-Einstein relation. a, Schematic to visualize the direction in which the displacements of particles in the pair are least correlated. A particle pair (red spheres) at $t_0$ with the centre-of-mass, $r_{CM}(r', r + r', t_0)$ (solid black circle), moves to a new position (green spheres), $r_{CM}(r', r + r', t + t_0)$, over $t$ (open black circle). The displacements of the particles in the pair located at $r'$ and $(r + r')$, i.e., $\Delta r(r', t)$ and $\Delta r(r + r', t)$, respectively, in the direction perpendicular to the centre-of-mass displacement direction are least influenced by HIs. b, Schematic showing representative $r$ at which influence of $(H_L + H_T)$ is weakest in the near-field. c, $\xi_{CM\perp}$ versus $r$. $\xi_{CM\perp} \rightarrow -1.01 \pm 0.01$ for $r > 8\sigma$; ideal expected value of $\xi$ is shown by black dashed line. The inset shows comparison of $\xi_{CM\perp}$ at $r$ corresponding to b (red spheres) with $\xi_{H_L}$ (solid black circles). $\xi_{CM\perp} = -1.00 \pm 0.01$ and $\xi_{H_L} = -1.19 \pm 0.02$ corresponding to points shown in the insets are shown as dashed lines. The errors bars are from fittings.
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