Dynamic responses of a system describe how disturbances in any thermodynamic quantity relax with time via motion of the constituent particles [1,2]. This forms the microscopic basis of transport processes [1–4], like diffusion in liquids. In general particle dynamics depends on the structure of the system. Systems driven away from equilibrium often show the emergence of structural heterogeneity in steady states, known as pattern formation [5]. This is relevant in areas ranging from material science to biological systems [3,4]. The particle dynamics in non-equilibrium systems is nontrivial due to drive [6–9]. Despite large number of studies [3–9], the connections between structure and dynamics, and consequently, the transport processes, are not well understood in non-equilibrium situations. This motivates us to explore dynamic responses and relate them to the underlying structural morphology in non-equilibrium steady states.

Colloids are model systems to realise material properties both in and out of equilibrium [3,4,6,7]. Variety of structures can be induced in colloids by external perturbations [3,4,6–9]. Moreover, the particles are large and move relatively slowly so that their motions can be followed by optical imaging [1–4,6–9]. Several experimental and theoretical works show that an external uniform electric field of large strength drives a binary mixture of oppositely charged colloidal particles to form steady-state heterogeneous structures with lanes of dynamically locked-in like-charged particles [3,4,6–19]. The mixture is homogeneous at low field.

We consider the steady states of a binary charged colloidal system in 3-dimensions in the presence of an electric field. We study how a perturbation in local density relaxes via microscopic motions in steady states. The density relaxation, also known as the van Hove function (vHf) [20], is measurable from scattering experiments and routinely used to probe dynamic responses [1,2]. The vHf consists of two parts: 1) The probability distribution of displacements of individual particles in a given time interval (self-vHf), characterising particle motions. 2) The probability distribution of separations between pair of particles (distinct-vHf), describing relaxation of structural changes, over a time interval [20]. The steady-state structural correlations are given by pair correlation functions (PCF) [1] which are probability distributions of inter-particle separation at a given time. The PCF is the distinct vHf where the time interval is zero. The self-vHf for a normal liquid is Gaussian whose width increases linearly with time. The slope defines the self-diffusion coefficient, \( D \) [1,20]. The distinct-vHf in such cases decays exponentially in time with a rate \( Dq^2 \) where \( 2\pi/q \) is the characteristic length of density relaxation [1].
We generate particle configurations following the Brownian Dynamics simulations [21]. The PCFs show that the structural correlations grow with increasing electric field as the system undergoes transition from homogeneous to lane state. The system has normal liquid-like dynamic responses both in homogeneous and lane state. At intermediate field, the self-vHf develops a slow exponentially falling tail. The distinct vHf shows a stretched exponential structural relaxation. Both behaviors are quite unlike normal liquids. The single particle motions show that the anomaly is due to heterogeneity in self-diffusion of particles. The heterogeneity has a non-monotonic dependence on structural correlations. We come up with a model to explain this non-monotonic behavior.

Our simulated system consists of a binary mixture of equal number of positively ($N_+$) and negatively ($N_-$) charged colloidal particles of diameter $\sigma (= 1 \mu m)$ ($N_+ = N_- = 1000$) in a solvent of viscosity $\eta (= 1 \text{cP})$ in a cubic box of length ($L = 21.599\sigma$) at temperature ($T = 298K$) with the periodic boundary conditions.

The pair interaction between particles at positions $\vec{R}_i$ and $\vec{R}_j$ and separation $r_{ij} = |\vec{R}_i - \vec{R}_j|$ is given by [14] $V(r_{ij}) = V_{SC}(r_{ij}) + V_{Repulsion}(r_{ij})$, where $V_{SC}(r_{ij}) = \frac{V_0(q_i q_j/(1 + \frac{|\vec{R}_i - \vec{R}_j|}{\kappa\sigma}))^2}{|\vec{R}_i - \vec{R}_j|}$ and $V_{Repulsion}(r_{ij}) = \varepsilon (\sigma/r_{ij})^{12} - (\sigma/r_{ij})^6 + \frac{1}{4} \frac{\varepsilon}{\kappa\sigma}$ for $r_{ij} < 2^{1/6}\sigma$ and zero, elsewhere. Here $q_i (= q$ for all $i$) is the charge of the $i$-th particle, $\kappa$ the inverse screening length, $V_0$ the interaction strength parameter and $\varepsilon = 4[|V_0(1 + \kappa\sigma/2)]^2$ [14]. We fix $\kappa\sigma (= 5.0)$ and $V_0^2 = |q|^2 V_0/k_B T (= 50.0)$ as in ref. [14]. The BD simulations are carried out using discretized form of the Langevin’s equation [21] with viscous damping $\Gamma (= 3\pi\eta\sigma)$ and electric field $f_0$ in the $z$-direction. The fluctuating force $\vec{F}$ has mean zero and variance $(\langle F^{(\perp)}(t) F^{(\perp)}(t') \rangle) = 2D_\sigma^2\delta(\delta(t - t'))$. Here $\alpha$ and $\beta$ denote the Cartesian components and $D_0 = k_B T/3\pi\sigma$ ($k_B$ the Boltzmann constant) the Einstein-Stokes Diffusion coefficient. We take $\tau_\sigma(= \sigma^2/D_0)$ as time unit, $\sigma$ the length unit and $k_B T$ the energy unit. The integration time step $\Delta t = 0.00005$.

The analysis is carried out in steady state for a non-zero $f(= |q| f_0 \sigma/k_B T)$, applied after the system is equilibrated without electric field. We calculate the lane order parameter, $\Phi$ defined in ref. [14]. We compute single particle densities [1] for both species in $XY$ and $XZ$ planes denoted by $\rho^{(+)}(X, Y)$ and $\rho^{(\pm)}(X, Z)$, respectively. We calculate the self-vHf for ions $G^{(+)}(\Delta r, t)(= (1/N_+) \sum_{i=1}^{N_+} \delta(\Delta r + \vec{R}_i(t) - \vec{R}_i(0)))$ displaced by a distance $\Delta r(= \Delta z, \Delta r_{\perp})$ in time interval $t$ [1]. Here $\Delta z$ is the displacement parallel and $\Delta r_{\perp}(= \Delta x, \Delta y)$ that in plane transverse to the field. We define the individual-vHf for a pair of $+ve$ ions separated at a distance $\vec{r}(= z, r_{\perp})$ in time interval $t$, given by $G_0^{(+)}(\vec{r}, t)(= (2/N_+ (N_+ - 1)) \sum_{i,j=1}^{N_+} \delta(\vec{r} + \vec{R}_i(t) - \vec{R}_j(0)))$. Similarly, we calculate distinct vHfs between a pair of $+ve$ and $-ve$ charges, $G_{D}^{(+)}(\vec{r}, t)(= (1/N_+ N_-) \sum_{i=1}^{N_+} \sum_{j=1}^{N_-} \delta(\vec{r} + \vec{R}_i(t) - \vec{R}_j(0)))$. The distinct vHf for the $-ve$ particles are defined likewise. All the averages are over initial conditions and 20 different Brownian trajectories each starting from a different equilibrium configuration. The PCFs for the $+ve$ ions $g^{(+)}(r_{\perp}, z)$, the $-ve$ ions $g^{(-)}(r_{\perp}, z)$ and between $+ve$ and $-ve$ ions $g^{(+,-)}(r_{\perp}, z)$, are given by the $t = 0$ value of the corresponding distinct-vHFs. To capture the microscopic motions, we compute the probability distributions $P^{(+)}(\Delta r_{\perp}, t)$ of particle displacements in transverse plane in time $t$ of 40 randomly tagged particles over 30 Brownian trajectories.

The steady state is characterized by steady values of $\Phi$. The equilibrated system ($f = 0$) has $\Phi(0) \approx 0.55$. After turning on the field ($f \neq 0$), we observe that $\Phi(f)$ grows with time and finally reaches a steady value with small fluctuations. The mean value of the order parameter $\Delta \Phi(= \Phi(f) - \Phi(0))$ shows continuous rise with $f$ as observed earlier [14].

Both species behave similarly, and we focus on the $+ve$ species. We show the density profiles $\rho^{(+)}(X, Y)$ in the $XY$ plane and $\rho^{(+)}(X, Z)$ in the $XZ$-plane in fig. 1 for different $f$. For small $f(= 50)$, a nearly homogeneous mixed phase is obtained, where small domains of like-charged particles aligned parallel to the field is observed (figs. 1(a) and 1(b)). There is an intermediate pre-lane state having much bigger domains with increasing $f(= 150)$ (figs. 1(c) and 1(d)). Finally the lane state takes place, as in earlier observations [16], for sufficiently large $f (= 300)$. Here networks of large domains in the $XY$-plane along with vertical lanes in the $XZ$-plane are seen (figs. 1(e) and 1(f)). The structural morphologies in the $XY$-plane are very similar to those in pattern forming liquids [5].
functions of and panel), the correlations in $g_f^{(\perp)}(r_{\perp}, \pm z)$ (inset) in fig. 2(c) as functions of $z$ at $r_{\perp} \approx 1$ for different values of $f$. For $f = 0$, $g_f^{(\pm)}(r_{\perp}, z) \approx g_f^{(\pm)}(r_{\perp}, -z)$ (data not shown). For $f \neq 0$, the symmetry in $g_f^{(\pm)}(r_{\perp}, z)$ is lost, although $g_f^{(\pm)}(r_{\perp}, z)$ remains symmetric. For $f = 50$, there are unequal peaks in $g_f^{(\pm)}(r_{\perp}, z)$ at $z \approx \pm 1$. This peak gets broadened at $f = 150$. For $f = 300$, $g_f^{(\pm)}(r_{\perp}, z)$ decays as $(-z)^{\gamma}$ with $\gamma \approx 2.09$. This algebraic decay is consistent to the observations reported for two-dimensional systems [18].

The correlation energy [1] $E^{C+}(f)(= \int V(\pm\pm)(r) \times g_f^{(\pm\pm)}(r_{\perp}, z) d^2 r_{\perp} dz + \int V(\pm-)(r) g_f^{(\pm-)}(r_{\perp}, z) d^2 r_{\perp} dz)$ is the cost of energy for replacing a negatively charged particle by a positively charged particle in a domain of positively charged particles. The inset of fig. 2(b) shows the correlation energy $\Delta E^{C+} = E^{C+}(f) - E^{C+}(0)$ and the energy due to the external electric field, $E^{P+}(f) = 2 f q_0 L^{2/3} |\rho(z)| dz$ as functions of $f$. The energy cost of bringing like charges in a domain is compensated by the external electrostatic energy above $f_C = 200$.

**Dynamic responses.**– Now we focus on the self-vHfs. We find that in the presence of non-zero $f$, $G_S^{(\pm)}(\Delta z, t) \neq G_S^{(\pm)}(-\Delta z, t) \neq G_S^{(\pm)}(\Delta r_{\perp}, t)$, where $\Delta r_{\perp} = |\Delta r_{\perp}|$. The peaks in $G_S^{(\pm)}(\pm\Delta z, t)$ shift linearly in $t$ with slope $v_d \approx q f_\perp / \Gamma$. We account for drift velocity, $v_d$ by transforming to $\Delta z' = \Delta z - v_d t$. In fig. 3, we show the dependence of $G_S^{(\pm)}(\Delta z', t)$ on $\pm\Delta z'$ for different $f$. For all $f$, $G_S^{(\pm)}(-\Delta z', t)$ is Gaussian. In contrast, $G_S^{(\pm)}(\Delta z', t)$ behaves differently: $G_S^{(\pm)}(\Delta z', t)$ is Gaussian for $f = 50$ (fig. 3(a)), while it has an exponential tail for $f = 150$ (fig. 3(b)) close to $f_c$. Increasing $f = 300$ further, $G_S^{(\pm)}(\Delta z', t)$ takes the form of a double Gaussian (fig. 3(c)).

In the transverse plane structural morphologies show distinct changes as $f$ is varied. $G_S^{(\pm)}(\Delta r_{\perp}, t)$ behaves similarly as $G_S^{(\pm)}(\Delta z', t)$. For small $f(= 50)$, $G_S^{(\pm)}(\Delta r_{\perp}, t)$ is Gaussian as shown in fig. 4(a). We find spatially exponential decay tail in $G_S^{(\pm)}(\Delta r_{\perp}, t) as in $G_S^{(\pm)}(\Delta z', t)$ for $f = 150$ (fig. 4(b)). The amplitude of the Gaussian part relative to that of the exponential tail for large $t$ approaches the ratio $\Phi(f)/(1 - \Phi(f))$. This implies that the exponential tail develops due to movement of $+$ve particles in the neighborhood of $-$ve particles. Anomalies in self-vHfs have been reported earlier in systems with competing time scales [22]. Heterogeneous density relaxation in our system is not ruled out due to formation of domains where the particles near the domain boundary may behave differently from those inside the domain. The self-vHfs have Gaussian form with double peaks for $f(= 300 > f_C)$ in the fully developed lane phase (fig. 4(c)).
We further quantify the changes in self-vHF in the transverse \[\text{plane}.\] The changes in the self-vHF take place at critical values, \(\Delta r^c\). We fit the data to a form, \(\exp(-\Delta r^c)^2/\sigma_\perp^2(t))\) for \(\Delta r^c < r_c\) and \(\exp(-\Delta r^c/\lambda_\perp(t))\) for \(\Delta r^c > r_c\) for \(f = 150\) (see footnote 1). The data fitting for \(f = 300\) has been done by double Gaussians with width parameters \(\sigma_\perp^2(t)\) for \(\Delta r^c < r_c\) and \(\sigma_\perp^2(t)\) for \(\Delta r^c > r_c\). The fitted curves are shown for representative cases in insets of fig. 4. We find that \(r_c\) decreases with \(t\), but saturates to a finite value for at least two decades (data not shown), implying that the deviations of dynamical behaviors from normal liquid persist for very long time. The inset of fig. 4(a) shows that \(\sigma_\perp^2(t)\) depends linearly on \(t\) as in normal liquids for \(f = 50\). The inset of fig. 4(b) shows \(\lambda_\perp(t) \sim t^{0.5}\). On the other hand, \(\sigma_\perp^2(t) \sim t\) (data not shown) for \(f = 150\). This is characteristic of non-Fickian diffusion [22]. The slopes of \(\sigma_\perp^2(t)\) and \(\sigma_\perp^2(t)\) for \(f = 300\) show linear dependences on \(t\) (fig. 4(c), inset). Our data indicate the presence of two diffusion coefficients for \(f = 300\).

Next, we address the relaxations of structures given by \(G^{(\pm)}_D(r_c, t)\). We show the dependences of \(G^{(\pm)}_D(r_c, t)\)

\[\text{1We restrict our data up to time window } t \approx 15\tau_3 \text{ so that the noisy part of the very long time interval is avoided in the fitting. We fit } G^{(\pm)}_S(\Delta_{r^c}, t) = A \exp(-\Delta_{r^c})^2/\sigma^2(t) \text{ for } \Delta_{r^c} < r_c(t) \text{ and } G^{(\pm)}_S(\Delta_{r^c}, t) = B \exp(-\Delta_{r^c}/\lambda(t)) \text{ for } \Delta_{r^c} > r_c(t). \text{ We minimize } \chi^2 \text{ with respect to } r_c \text{ and the fitting parameters. Similar fitting procedures are repeated for } \Delta z \text{ dependences. The fitting for other values of } f \text{ has also been done similarly.}\]

Fig. 3: ln\(G^{(\pm)}_S(\Delta z', t)\) vs. \(\Delta z'\) for (a) \(f = 50\), (b) \(f = 150\) and (c) \(f = 300\) for \(t = 0.1\tau_3\) (filled triangles), \(1.0\tau_3\) (filled circles), \(10.0\tau_3\) (open circles). Exponential (dashed) and Gaussian (dot-dashed) lines are the fitted curves.

Fig. 4: Transverse plane self-vHFs: ln\(G^{(\pm)}_S(\Delta r^c, t)\) vs. \(\Delta r^c\) plots for \(t = 0.1\tau_3\) (filled triangles), \(1.0\tau_3\) (filled circles), \(10.0\tau_3\) (open circles): (a) \(f = 50\); dashed line: Gaussian fit. Inset: \(\sigma_\perp^2(t)\) as a function of \(t\) (b) \(f = 150\); dashed line: Gaussian and dot-dashed line: exponential tail; inset: \(\lambda(t)\) as a function of \(t\) (triangles), dotted line shows \(\lambda(t) \sim (t/\tau_3)^{0.5}\). (c) \(f = 300\); dashed line and dot-dashed line: double Gaussian fits. Inset: dependences of \(\sigma_\perp^2(t)\) (filled symbols) and \(\sigma_\perp^2(t)\) (open symbols) on \(t\). Lines show the best fitted linear curves.

and \(G^{(\pm)}_D(r_c, t)\) on \(r_c\) in fig. 5(a) for \(f = 150\) for two different \(t\). At \(t = 0\), there is a peak in \(G^{(\pm)}_D(r_c, t)\) at \(r_c = 0\) which decreases with \(t\). \(G^{(\pm)}_D(r_c, t)\) shows a strong peak at \(r_c \approx 1\) that flattens with \(t\). The wave vector \(q_c\)-dependent distinct-vHFs, \(G^{(\pm)}_D(q_c, t)\) corresponding to \(G^{(\pm)}_D(r_c, t)\) show a maximum at \(q_c = q_0\). On the other hand, \(G^{(\pm)}_D(q_c, t)\) corresponding to \(G^{(\pm)}_D(r_c, t)\) show a minimum around the same wave vector (fig. 5(a), inset). Both the maximum and minimum values decrease with \(t\).

The structural relaxations are given by the decays in the maximum or minimum values with \(t\). We quantify this by \(C^{(\pm)}_0(t) = G^{(\pm)}_D(q_0, t)\) and \(C^{(\pm)}_0(t) = 1/G^{(\pm)}_D(q_0, t)\). We show these quantities in fig. 5(b) in semi-logarithmic plots. The plots show that the relaxation of +ve particles in the neighbourhood of other –ve particles (denoted by \(C^{(\pm)}_0(t)\)) is slower than that in the vicinity of other +ve particles (denoted by \(C^{(\pm)}_0(t)\)). We observe that the decay is exponential in \(t\) in general indicating diffusive relaxation [1]. However, \(-\ln C^{(\pm)}_0(t) \sim t^{0.75}\) for \(f = 150\), implying a slow stretched exponential relaxation.

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The slow relaxation at $f = 150$ can be understood within the Vineyard approximation [1] where the distinct vHf is a convolution of self-vHf and PCF. Within this approximation, the time dependence of the distinct-vHf is governed by that of self-vHf. Our simulations indicate that $G_S^{(+)\perp}(q_\perp,t) = G_S^{(N)}(q_\perp,t) + \frac{\Phi}{\tau} G_S^{(T)}(q_\perp,t)$ for a given wave vector $q_\perp$, where $G_S^{(N)}(q_\perp,t)$ is the Fourier transform of the Gaussian component and $G_S^{(T)}(q_\perp,t)$ that of the exponential tail part. Let $\ln C_0(t) = \ln G_S^{(N)}(q_\perp,t) - C_r(t)$, where $C_r(t)$ is correction due to the tail. Assuming the correction to be small, one gets from the Vineyard approximation, to the first order in $\Phi$, $-C_r(t) \sim \frac{\Phi}{\tau} G_S^{(T)}(q_\perp,t)$. For the transverse plane, (in 2 dimensions), $G_S^{(T)}(q_\perp,t) \sim \int_{\mathbb{R}^2} e^{-r/\lambda}J_\lambda(\sqrt{q^2\cdot r})dr$. Substituting $\zeta = r/\lambda$ and using $J_0(\zeta) \sim 1/\sqrt{\pi\zeta}$, one gets, $G_S^{(T)}(q_\perp,t) \sim \lambda^{3/2}(t) \int \zeta^{-1/2}e^{-\zeta^2}d\zeta$. Using $\lambda(t) \sim t^{0.5}$ as observed in our simulations, we get $C_r(t) \sim t^{0.75}$. Thus, $-\ln C_0(t)$ has a slower decay with dependence $\sim t^{0.75}$. The stretched exponential decay in $-\ln C_0^{(++)}(t)$ indicates that this slowing-down is due to $+ve$ particles in the vicinity of $t/\tau = 300$ (triangles). Lines show the fitted curves.

Fig. 5: (a) $G_D^{(++)}(r_\perp,t)$ (lower curves) and $G_D^{(++)}(r_\perp,t)$ (upper curves, with vertical offset 1.0) as functions of $r_\perp$ for $t = 0$ (solid line) and $t = 10\tau_3$ (dashed line) for $f = 150$; inset: dependences of $G_D^{(++)}(q_\perp,t)$ (upper curves, with vertical offset 0.1) and $G_D^{(++)}(q_\perp,t)$ (lower curves) on $q_\perp$ for $f = 150$ for $t = 0$ (solid line) and $t = 10\tau_3$ (dashed line). (b) Dependence of $-\ln C_0^{(++)}(t)$ (open symbols) and $-\ln C_0^{(++)}(t)$ (filled symbols) on $t/\tau_3$ for $f = 50$ (squares), $f = 150$ (circles) and $f = 300$ (triangles). Lines show the fitted curves.

Fig. 6: (a) $\sigma_{\perp i}^2(t)$ vs. $t$ plots for two randomly tagged particles for $f = 150$ (open symbols) and $f = 300$ (filled symbols). (b) $P(D_r/D_B)$ vs. $D_r/D_B$ plots for $f = 50$ (dotted line with open circles), 150 (solid line with filled squares) and 300 (dotted line with filled circles); inset: $\Omega$ as a function of $f$. The dotted line is guide to the eyes.

**Particle resolved picture.** – In order to gain microscopic understanding of the dynamic behavior we measure the probability distribution of displacements $P^{(++)}(\Delta r_\perp,t)$ of tagged $+ve$ particles. We tag random particles from different structural regions. The second moments, $\sigma_{\perp i}^2(t) = \int \Delta r_\perp^2 P^{(++)}(\Delta r_\perp,t)d^2(\Delta r_\perp)$, are shown in fig. 6(a) for $f = 150$ and 300, respectively. We find different slopes for different particles in the given time window. The slopes give the self-diffusion coefficients, $D_r$ of the tagged particles. Figure 6(b) shows the distribution $P(D_r/D_B)$ where $D_B$ is the bulk diffusion coefficient. We observe a sharp peak in $P(D_r/D_B)$ for $f = 50$. The distribution is much broader for $f = 150$ but gets sharp again at $f = 300$. We take the width of $P(D_r/D_B)$ around the peak, $\Omega$, as a measure of heterogeneity in diffusion. $\Omega$ has a maximum around $f = 150$ (fig. 6(b), inset).

The exponential tail in self-vHf and stretched exponential structural relaxation in distinct vHf are seen when $P(D_r/D_B)$ is broad. This is in agreement to the phenomenological proposition that the exponential tail originates due to heterogeneity in diffusion [22]. Note that dynamic slowing-down is observed in super-cooled systems as well [23,24]. However, the slow dynamics in such systems is due to caging of the particles by their neighbours [25]. In contrast, the individual particle
motions here are always diffusive. However, the diffusion has heterogeneity.

This heterogeneous diffusion implies the heterogeneity in transport processes. The mean diffusion, \( \langle D \rangle = \int D(P) dD \approx 1.3D_B \) for \( f = 50 \). However, \( \langle D \rangle \) is ill-defined for \( f = 150 \). \( \langle D \rangle \approx 0.7D_B \) corresponding to the peak for \( f = 300 \) is lower than that for \( f = 50 \). However, the tail in \( P(D) \) for \( f = 300 \) corresponds to \( \langle D \rangle \approx 2D_B \) which is higher than that for \( f = 50 \). The presence of two different diffusivities indicates dynamic locked-in situation, qualitatively similar to the observations reported in ref. [16].

### Generalization

The width of distribution of diffusivities depends non-monotonically on the structural correlation length scale which grows with \( f \). This non-monotonic dependence can be understood in a general context. Let us consider two domains, consisting of particles denoted as A and B. \( V^{AA}(r) \) is the mutual interaction of the particles in a domain A and \( V^{BB}(r) \) that in a domain B. \( V^{AB}(r) \) is the interaction between particles of different domains. Let there be a relative drift, \( v_{rel} \) in the z-direction between the domains. Let us focus on one of the domains, say domain A. The local density in the domain changes due to single particle motion. The single particle motions can be of two different types: 1) diffusive and drive currents of particles A; 2) movement of particles A to domain B. The second type of movement across different domains costs interaction energy due to changes in the neighboring particles at mean density \( \rho_0 \), \( \delta V \approx [V^{AA}(r) = \rho_0^{-1/3} - V^{AB}(r) = \rho_0^{-1/3}] \rho_0 \) where \( \rho_0^{-1/3} \) is the mean separation between the particles. The probability, \( p \) of motion across the domain wall is \( \sim \exp(-\delta V/k_BT) \). These particles will experience a force of magnitude, \( f_{rel} = (\Gamma_{rel} \cdot r) \) corresponding to the drive with energy cost \( \delta V_{ext} \sim p f_{rel} \rho^{-1/3} \). The corresponding current is \( -[\rho_{rel} \exp(-\delta V_{ext}/k_BT)] \delta\rho^{\prime}(\vec{r}',z,t) \). Here \( \delta\rho^{\prime}(\vec{r}',z,t) = \rho^{\prime}(\vec{r}',z,t) \) where \( \rho^{\prime}(\vec{r}',z,t) \) is local density. The negative sign accounts for the loss of particles from domain A.

The linearized equation of continuity for density is

\[
- \frac{\partial}{\partial t} + D \nabla^2 + \lambda \partial_z \delta \rho^{\prime}(\vec{r}',z,t) = N(\vec{r}',z,t),
\]

where \( N(\vec{r}',z,t) \) is the noise with Gaussian statistics. Here \( \lambda = v_{rel}(1-\exp(-\delta V_{ext}/k_BT)) \). This term represents competition between drive and particle interaction. The diffusive current in \( z \) has been ignored in comparison to the drive current. The steady state implies \( \delta \rho^{\prime} = 0 \), and hence, eq. (1) yields

\[
D \nabla^2 + \lambda \partial_z \delta \rho^{\prime}(\vec{r}',z,t) = N(\vec{r}',z,t).
\]

Using \( \rho^{\prime}(\vec{r}',z) = \int e^{i\vec{k}' \cdot \vec{r}} e^{i\vec{k} \cdot \vec{r}} \rho_{\vec{k}'}(\vec{k}) d\vec{k}' \) and \( \rho_{\vec{k}'}(\vec{k}) = \int e^{i\vec{k} \cdot \vec{r}} e^{i\vec{k} \cdot \vec{r}} N(\vec{k},\vec{k}) d\vec{k} \) in eq. (2), one obtains \( \langle \rho(\vec{k},\vec{k}) \rangle = \frac{\langle N(\vec{k},\vec{k}) \rangle}{D^2(\vec{k}^2 + \lambda^2)} \).

In real space \( \langle \delta \rho^{\prime}(\vec{r}',z) \delta \rho^{\prime}(\vec{r},0) \rangle = \int e^{-i\vec{k} \cdot \vec{r}} e^{i\vec{k} \cdot \vec{r}} \delta \rho^{\prime}(\vec{r}',z) d\vec{k} \) for white noise, where \( \langle N(\vec{k},\vec{k}) \rangle = C \) (constant). First, we perform the \( k_z \) integral. The denominator of the integrand has the poles at \( k_z = 0 \) (second order) and \( k_z = \pm i(2k_2^2 + \lambda^2)^{1/2} \) for high \( \lambda \). We consider a semicircle with pole at \( k_z = -i(2k_2^2 + \lambda^2)^{1/2} \).

Using Cauchy theorem: \( \langle \delta \rho^{\prime}(\vec{r}',z) \delta \rho^{\prime}(\vec{r},0) \rangle \sim \int k_z dk_z e^{-ik_z \cdot r} e^{-i(2k_2^2 + \lambda^2)^{1/2} k_z} \). Substituting \( \zeta^2 = (2k_2^2 + \lambda^2)^{1/2} \), we get \( \langle \delta \rho^{\prime}(\vec{r}',z) \delta \rho^{\prime}(\vec{r},0) \rangle \sim e^{-z^2/\zeta^2} \int d\zeta \), so that the decay of correlation is \( e^{-z^2/\zeta^2} \) in \( r_z \).

The above scenario assumes the movement of a particle with diffusion \( D \). Let us now consider a pair of particles where the second particle has slightly different diffusion coefficient \( D + \delta D, \delta D \) is taken to be a random variable as suggested by our simulations. The relative diffusion of the particles \( \tilde{D} = 2D + \delta D \). If the particles are structurally uncorrelated, the separation between a pair of particles fluctuates with time with probability \( \sim \exp(-r_z^2/\tilde{D}t) \). Due to structural correlation, the probability is given by \( \sim \exp(-x_rz/\tilde{D}) \exp(-r_z^2/\tilde{D}t) \). Integration over \( r_z \) and \( t \) results in moments of probability distribution of \( \tilde{D} \). Let us consider the integral, \( \tilde{I} = \int \frac{e^{-z^2/\zeta^2}}{\sqrt{\zeta^2}} e^{-z^2/\tilde{D}t} d\zeta d\tilde{D} \). Substituting \( \zeta = \frac{z}{\sqrt{\tilde{D}}} \) in \( I \), we can rewrite the integral as \( I = \int e^{-2z^2/\tilde{D}t} d\tilde{D} \int \frac{d\zeta}{\sqrt{\tilde{D}}} e^{-\zeta} \). Further, substitution yields \( I = \frac{\sqrt{\pi} \varphi}{\tilde{D}z} \). yields \( \tilde{I} \sim \frac{1}{\tilde{D}^2} \) which can be interpreted as the moment of marginal distribution, in \( P(\tilde{D}) \). Since \( D \) is a constant one can as well take \( D = 0 \) without any loss of generality, so that one gets the moments of distribution of the random variable \( \delta D \).

Our simulated system, A and B stands for positively and negatively charged particles, respectively. Using \( V^{AA} = V^{(+)} \approx \frac{\nu_0}{(1+z^2)^{1/2}} \exp(-(z+1)/\sigma) \), \( V^{AB} = V^{(-)} \approx \frac{\nu_0}{(1+z^2)^{1/2}} \exp(-(z-1)/\sigma) \) and \( v_{rel} = \partial_r = f/\Gamma \), we obtain the maximum in \( \omega \) at \( f \approx 150 \) with the parameters taken from the simulation model. This is in agreement to our simulation results where the maximum in \( \Omega \) is at \( f \approx 150 \). Thus the model can qualitatively capture the anomalies observed in our BD simulations.

Our model is directly applicable to phase-separating liquids in the presence of drive both in two dimensions [12] and three dimensions [14]. There exist complex systems, where anomalies in dynamical responses have been ascribed phenomenologically to heterogeneity in diffusion [22,23,26-28]. The generality of our analysis points to exploring the possibility of relating such heterogeneity to non-equilibrium effects where the particle interactions compete with local drive [29-31]. The local drives need not be necessarily external, but may arise out of unbalanced thermodynamic forces due to non-uniform
distribution of pressure, temperature and chemical potential in the system.

Our theoretical analysis shows that the anomalies in dynamics of driven binary colloid are due to competition between particle interaction and drive in the system. These results can be verified by experiments. The heterogeneity in diffusion reflects heterogeneity in transport processes which is tunable by external drive. This may be harnessed in technological applications [30,31].

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