Persistent and extremely persistent dense active fluids

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L. Berthier, E. Flenner & G. Szamel, *How active forces influence nonequilibrium glass transitions*, New J. Phys. **19**, 125006 (2017)

L. Berthier, E. Flenner & G. Szamel, *Perspective: Glassy dynamics in dense systems of active particles*, J. Chem. Phys. **150**, 200901 (2019)

G. Szamel & E. Flenner, *Perspective: Long-ranged velocity correlations in dense systems of self-propelled particles*, EPL **133**, 60002 (2021)

G. Szamel & E. Flenner, *Extremely Persistent Dense Active Fluids*, Soft Matter, **20**, 5237 (2024)

Thanks to: NSF

Outline

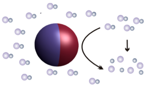
1. Introduction
2. Glassy dynamics (moderately persistent active fluids)
3. Extremely persistent active fluids
4. Velocity correlations in systems of persistent active particles

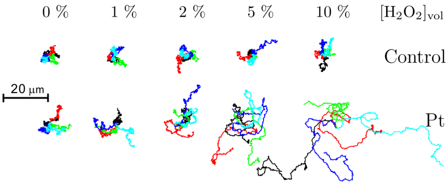
Active Matter

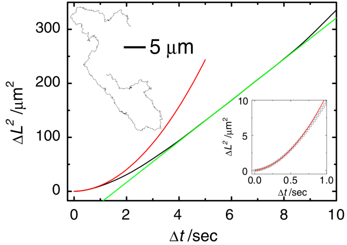
Active matter: constituents "consume" energy and convert it into (systematic) motion

- Important feature: driving/breaking of detailed balance/breaking of time reversal symmetry at the level of individual particles.
- Result: single-particle and collective behavior very different from that exhibited by non-active (*i.e.* passive) systems:
  - Monothermal cyclic engine.
  - Motility-induced phase separation (MIPS)
    - liquid-gas-like phase separation in systems with purely repulsive interactions.
  - Non-trivial dynamics in extremely persistent active fluids. $\Leftarrow$
  - Non-trivial equal-time correlations between velocities of different active articles. $\Leftarrow$

Example: active colloidal particles



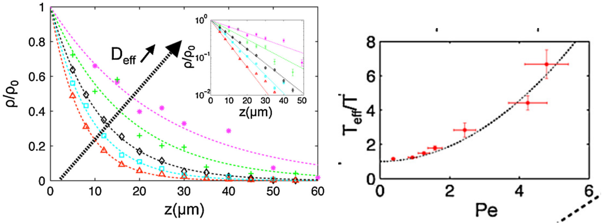




Janus colloids

Howse, ..., Golestanian, PRL 2007

- One side of a spherical colloid covered by a catalyst; chemical reaction $\rightarrow$ self-propulsion.
- Short times: ballistic motion.
- Long times: diffusive motion.



- Dilute active colloids in gravity $\rightarrow$ barometric distribution with effective temp. Palacci et al. PRL 2010.

Active Brownian Particles

ten Hagen *et al.* JCPM 2011

- Overdamped dynamics, no hydrodynamic interactions (dry active matter).
- Each particle is endowed with self-propulsion velocity of magnitude v_0.
- The direction of the self-propulsion velocity diffuses freely;
  τ: persistence time of the direction.
- Equations of motion in two spatial dimensions:

$$\gamma_t \dot{\mathbf{r}}_i = -\sum_j \nabla_i V(r_{ij}) + \gamma_t v_0 \mathbf{n}_i + \boldsymbol{\zeta}_i \quad \left\langle \boldsymbol{\zeta}_i(t)\boldsymbol{\zeta}_j(t')\right\rangle = 2\boldsymbol{I}\gamma_t T \delta_{ij}\delta(t-t'),$$

$$\mathbf{n}_i = (\cos(\varphi_i), \sin(\varphi_i))$$

$$\gamma_r \dot{\varphi}_i = \eta_i, \qquad \left\langle \eta_i(t)\eta_j(t')\right\rangle = 2\gamma_r T \delta_{ij}\delta(t-t')$$

  γ_t & γ_r - friction coefficients; rotational diff. coeff. $D_r = T/\gamma_r$; persistence time $\tau = D_r^{-1}$

- If thermal noise in the equation of motion for the position is neglected $\rightarrow$ athermal ABP model; active temperature $T_a = \gamma v_0^2 \tau/2 = \gamma v_0^2/(2D_r)$.

Active Ornstein-Uhlenbeck particles

GS PRE 2014, Martin *et al.* PRE 2021

- Overdamped dynamics; self-propulsion force evolves according to the Ornstein-Uhlenbeck process.

$$\gamma \dot{\mathbf{r}}_i = -\sum_j \nabla_i V(r_{ij}) + \mathbf{f}_i + \boldsymbol{\zeta}_i \qquad \left\langle \boldsymbol{\zeta}_i(t)\boldsymbol{\zeta}_j(t')\right\rangle = 2\gamma T \delta_{ij} \boldsymbol{I} \delta(t-t'),$$

$$\tau_p \dot{\mathbf{f}}_i = -\mathbf{f}_i + \boldsymbol{\eta}_i \qquad \left\langle \boldsymbol{\eta}_i(t)\boldsymbol{\eta}_j(t')\right\rangle = 2\gamma T_a \delta_{ij} \mathbf{I} \delta(t-t')$$

γ - friction coefficient; $\mathbf{f}_n$ - self-propulsion force; τ_p - persistence time;
$\boldsymbol{\eta}_i$ - noise of the reservoir coupled to the self-propulsion; T_a - noise strength

- In contrast to an ABP, for an AOUP both the direction and the magnitude of the self-propulsion evolve stochastically.
- Analogue of v_0 - root-mean-squared self-propulsion $f = \sqrt{3T_a/\tau_p}$.
- Advantage: for a single particle many properties can be evaluated analytically.
- If the noise in the equation of motion for the position is neglected $\rightarrow$ athermal AOUP model.

Short persistence time limit

- In the short persistence time limit, $\tau_p \to 0$, and at constant active temperature T_a, an active system becomes equivalent to a passive system at a higher temperature.

- For example, an athermal system of AOUPs in the $\tau_p \to 0$ limit becomes equivalent to a thermal Brownian system at temperature $T = T_a$.
- A thermal system of AOUPs at temperature T in the $\tau_p \to 0$ limit becomes equivalent to a thermal Brownian system at temperature $T + T_a = T_{\text{eff}}$.

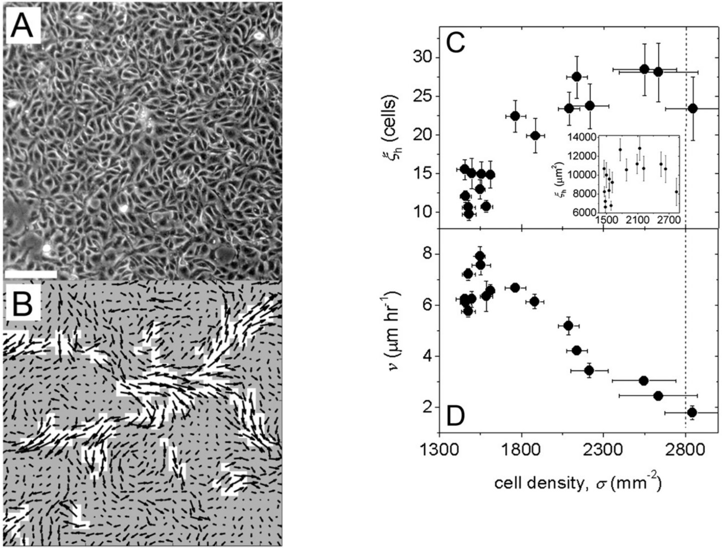
Active systems: very large parameter space compared to passive systems

- Passive system: temperature T and number density n or volume fraction ϕ.
- Active system: two more parameters that characterize the strength of the self propulsion and the persistence of the self-propulsion.
  - ABP: T, ϕ, self-propulsion velocity v_0 and persistence time τ.
  - athermal ABP: ϕ, self-propulsion velocity v_0 and persistence time τ.
  - athermal AOUP: ϕ, active temperature T_a and persistence time τ_p.
- Very different behavior can be observed while changing one parameter, depending on the path in the parameter space.
  - Increasing persistence time at constant T_a.
  - Increasing persistence time at constant root-mean-squared self-propulsion $f \propto \sqrt{T_a/\tau_p}$.

Outline

Glassy dynamics of cell motion

Angelini *et al.* PNAS 2011



- Confluent cell layer

Glassy dynamics:

- Dynamic heterogeneity
- Slowing down

AOUPs with purely repulsive interactions

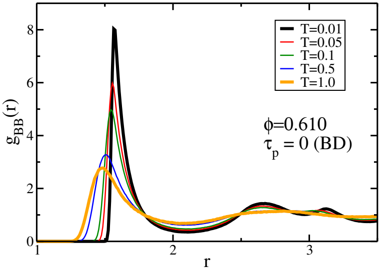
Berthier, Flenner & GS, NJP **19** 125006

- 50:50 mixture of athermal active Ornstein-Uhlenbeck particles
- interactions: WCA truncation of the LJ potential

$$V_{\alpha\beta}(r) = \begin{cases} 4\epsilon\left(\left(\frac{\sigma_{\alpha\beta}}{r}\right)^{12} - \left(\frac{\sigma_{\alpha\beta}}{r}\right)^{6}\right) & r \leq 2^{1/6}\sigma_{\alpha\beta} \\ 0 & r > 2^{1/6}\sigma_{\alpha\beta} \end{cases}$$

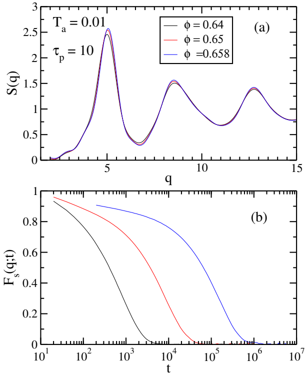
$$\sigma_{AA} = 1.0; \quad \sigma_{AB} = 1.2; \quad \sigma_{BB} = 1.4$$

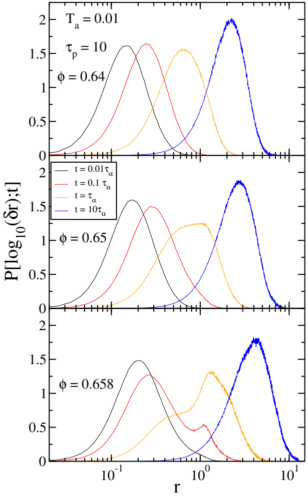
- In the $\tau_p \to 0$ limit this system is equivalent to a thermal system $T = T_a$; for this thermal system in the $T \to 0$ limit we get a binary hard sphere mixture:



Glassy dynamics: dependence on ϕ at $T_a = 0.01$

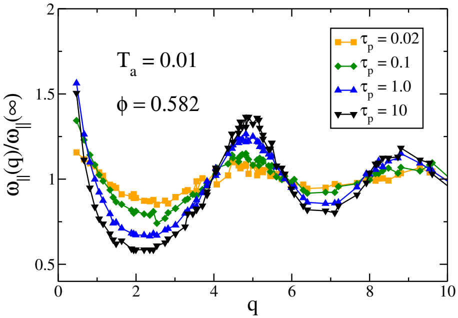
$$S(q) = \frac{1}{N}\left\langle \sum_{i,j} e^{i\mathbf{k}\cdot(\mathbf{r}_i - \mathbf{r}_j)}\right\rangle \qquad F_s(q;t) = \frac{1}{N}\left\langle \sum_i e^{i\mathbf{q}\cdot(\mathbf{r}_i(t) - \mathbf{r}_i(0))}\right\rangle$$





- Small changes of $\phi \rightarrow$ small changes in the structure factor and big changes in the dynamics.
- Dynamic heterogeneity

Active glassy dynamics: non-trivial *equal time* velocity correlations

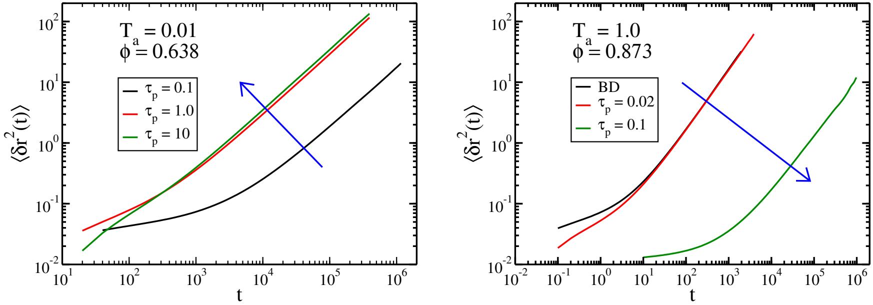


- Velocity of particle i: $\frac{1}{\gamma}(\mathbf{f}_i + \mathbf{F}_i) \equiv \frac{1}{\gamma}(\mathbf{f}_i + \sum_j \mathbf{F}_{ij})$
- Longitudinal velocity correlations:

$$\omega_{\|}(q) = \frac{1}{N\gamma^2} \left\langle \hat{\mathbf{q}} \cdot \sum_i (\mathbf{f}_i + \mathbf{F}_i) e^{-i\mathbf{q}\cdot\mathbf{r}_i} \; \hat{\mathbf{q}} \cdot \sum_l (\mathbf{f}_l + \mathbf{F}_l) e^{i\mathbf{q}\cdot\mathbf{r}_l} \right\rangle$$

- For a thermal Brownian system (in the limit $\tau_p \to 0$, $T_a =$const.),velocities of different particles are uncorrelated & $\omega_{\|}(q)\tau_p = T$.

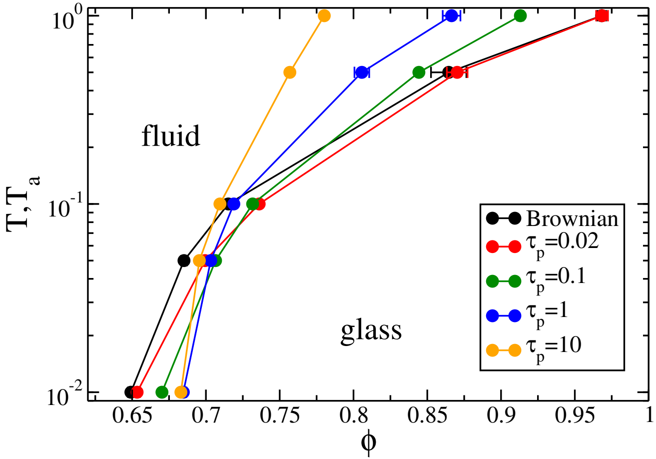
Dependence of the dynamics on τ_p at const. T_a



Arrows indicate increasing persistence time.

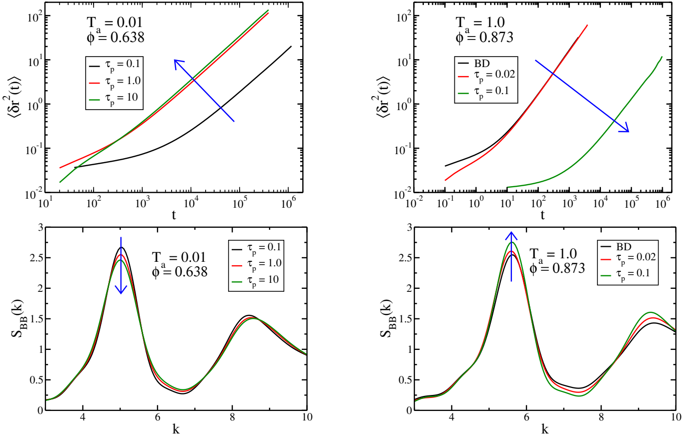
- Short-time dynamics slows down with increasing τ_p.
- Long-time dynamics speeds up at $T_a = 0.01$ and slows down at $T_a = 1.0$.

Dependence of the apparent glass transition line on τ_p



Increasing persistence time at constant active temperature T_a fluidizes or glassifies the active system, depending on T_a.

Long-time dynamics correlates with steady state structure factor



Arrows indicate increasing persistence time.

Newer, alternative interpretation: dynamics is the fastest when cage size coincides with the persistence length Debets *et al.*, PRL **127**, 278002 (2021).

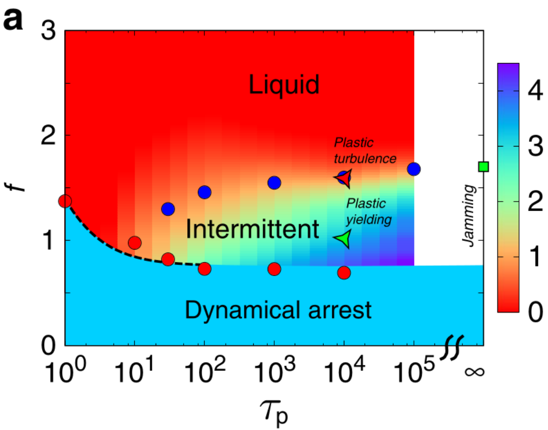
Summary

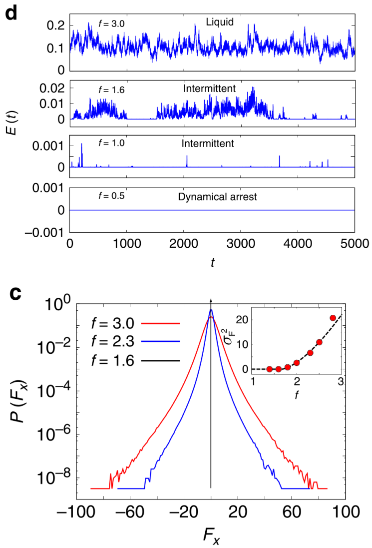
- At low active temperatures, increasing persistence time fluidizes the active system.
- At higher active temperatures and higher volume fractions, increasing persistence time makes the active system more glassy.
- Changes in the dynamics correlate with the changes in the peak value of the steady state structure factor.
- Note: an equilibrium system with the same pair correlations would be completely arrested.

Outline

Extreme active matter at high densities

Mandal *et al.* Nat. Comm. 2020

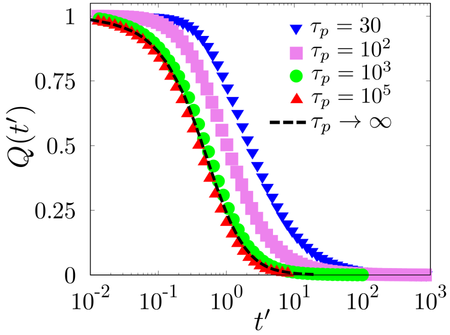


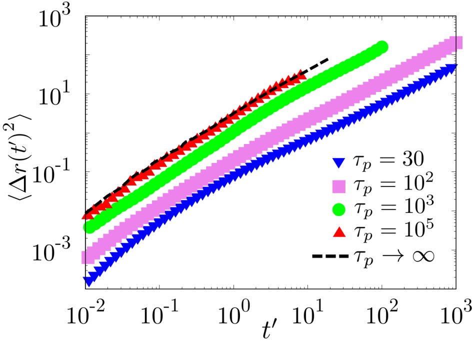


Intermittent dynamics at very large persistence times and intermediate active forces;
exponential tails of “total force” distribution.

Motion on the time scale of the persistence time

Mandal & Sollich, JPCM **33**, 184001 (2021)





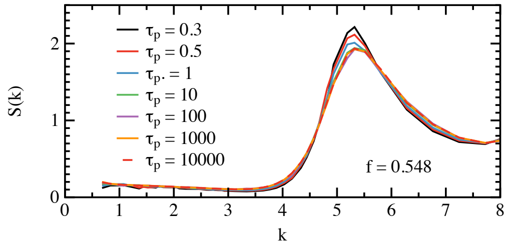
In the $\tau_p \to \infty$ limit the overlap function and the mean-squared displacement evolve on the time scale of the persistence time, $t' = t/\tau_p$.

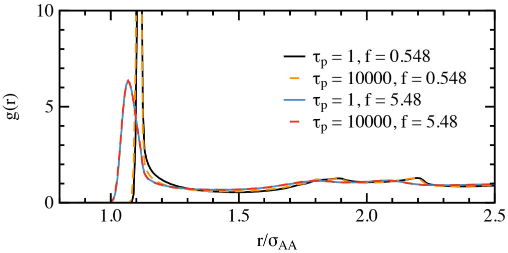
Extremely persistent active fluids

GS & Flenner SM **20**, 5237 (2024)

- Model system: the same as in the glassy dynamics study: athermal active Ornstein-Uhlenbeck particles with purely repulsive interactions.
- Goal: dynamics of active fluids in the $\tau_p \to \infty$ limit.
- To mimic Mandal *et al.* we keep the strength of active force $f = \sqrt{3T_a/\tau_p}$ constant and increase τ_p, *i.e.* we also increase $T_a \propto \tau_p$!

For a given f, structure depends very weakly on τ_p

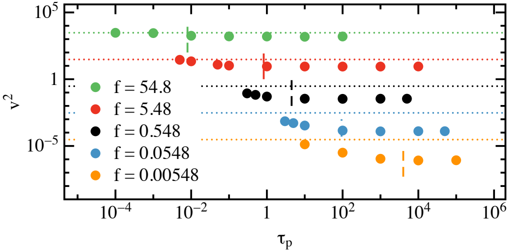




- For a fixed value of self-propulsion force f, the structure of our active system depends only weakly on persistence time τ_p.
- Structure factor saturates at large persistence times.
- At all values of self-propulsion force that we studied, there is pronounced short-range stucture.

- The dynamics is ballistic up to persistence time; the large τ_p characteristics of the long-time dynamics depend as power laws on self-propulsion force.

Single-particle motion: mean squared velocity GS & Flenner SM **20**, 5237 (2024)

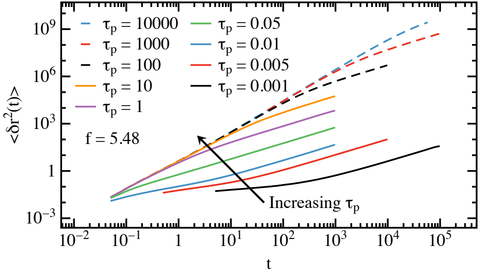


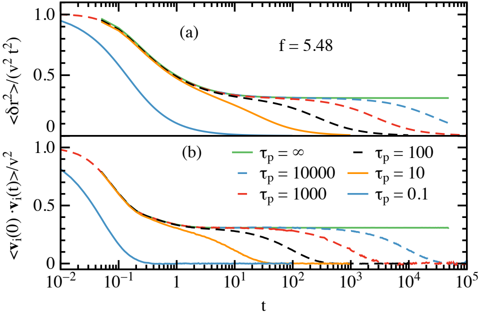
$$v^2 = \frac{1}{N}\left\langle \sum_n \dot{\mathbf{r}}_n^2 \right\rangle \equiv \frac{1}{N\gamma^2}\left\langle \sum_n \left(\mathbf{f}_n + \sum_m \mathbf{F}_{nm} \right)^2 \right\rangle$$

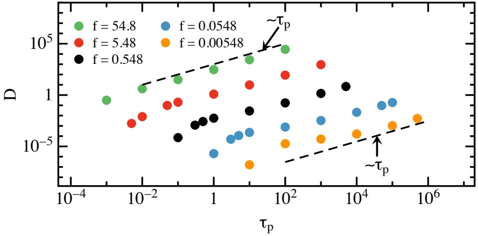
- v^2 quantifies how well the self-propulsion force is balanced by the interparticle interactions.
- Short persistence times: $v^2 \approx f^2/\gamma^2$.
- Long persistence times: v^2 saturates at a smaller but finite value.

Single-particle motion: MSD & D

GS & Flenner SM **20**, 5237 (2024)

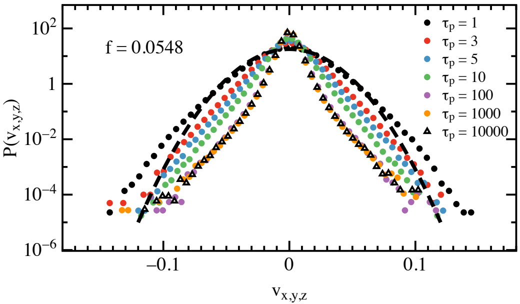




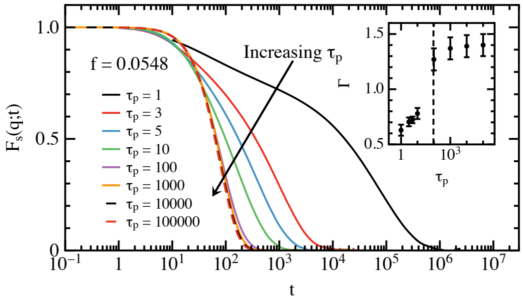


- MSD is ballistic at short times (expected) and at intermediate times (surprising).
- Time-dependent velocity correlation function develops a plateau that reflects the second ballistic regime.
- At large τ_p, $D \propto \tau_p$.

Single-particle motion: velocity distrib. & $F_s = \frac{1}{N}\left\langle \sum_n e^{i\mathbf{q}\cdot(\mathbf{r}_n(t)-\mathbf{r}_n(0))}\right\rangle$



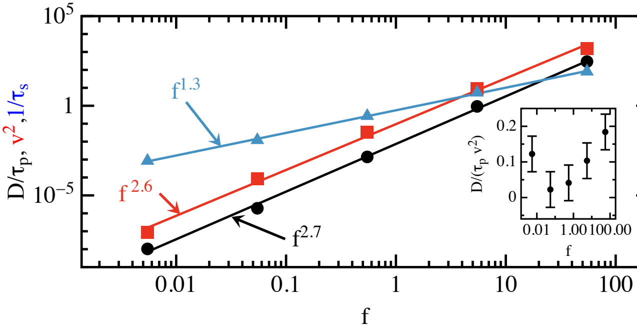
Broad, exponential tails!



Compressed exponential:

$$F_s(q;t) \propto e^{-(t/\tau_s)^{\Gamma}},\ \Gamma > 1.$$

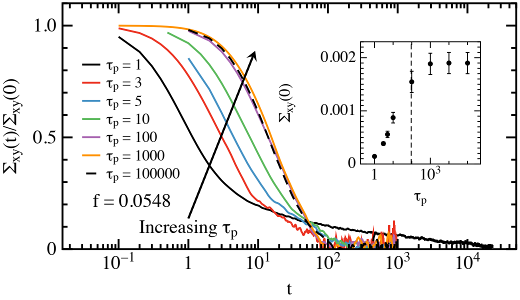
Single-particle motion: scaling laws for $\tau_p \to \infty$ quantities



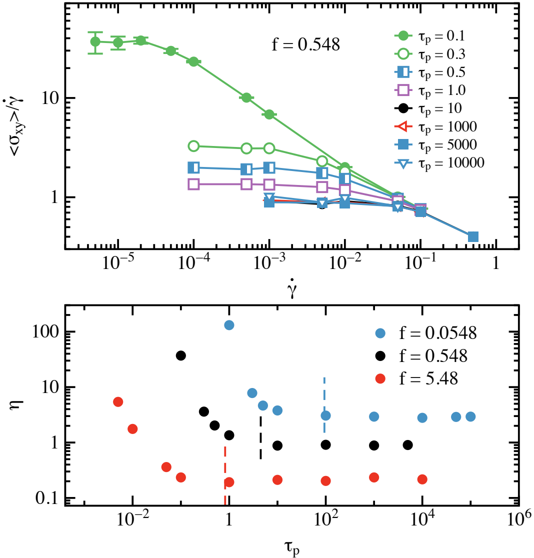
Large τ_p limits of the mean-squared velocity, D/τ_p and the F_s relaxation time depend on the strength of active forces as power laws.

Non-interacting particles: $v^2 \propto D/\tau_p \propto f^2$ & $1/\tau_s \propto f$.

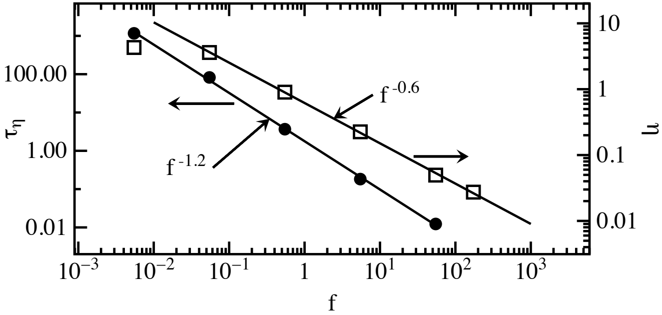
Shear stress correlations and shear viscosity



Exponential time dependence in the $\tau_p \to \infty$ limit.

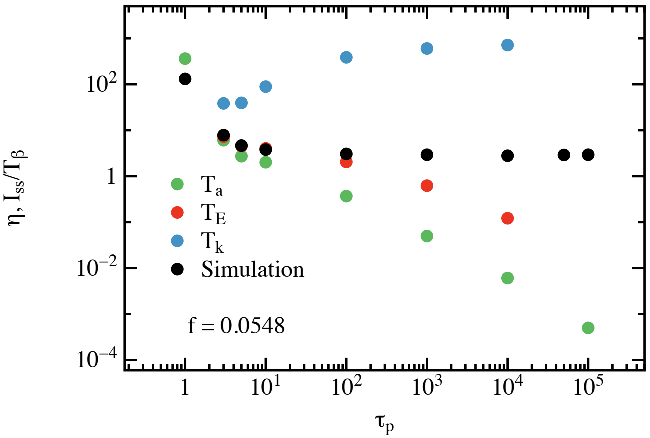


Rheology: scaling laws for $\tau_p \to \infty$ quantities



Large τ_p limits of the shear-stress correlation time and the viscosity depend on the strength of active forces as power laws.

Equilibrium FDT implies $\eta = T^{-1}\int_0^\infty \Sigma_{xy}(t)dt$



- black circles: η from direct simulations
- green: $T_a^{-1}\int_0^\infty \Sigma_{xy}(t)dt$

  T_a - active temperature
- red: $T_E^{-1}\int_0^\infty \Sigma_{xy}(t)dt$

  $T_E = \frac{\text{diffusion}}{\text{mobility}}$

  T_E - Einstein effective temperature
- blue: $T_K^{-1}\int_0^\infty \Sigma_{xy}(t)dt$

  $T_K = v^2$ - kinetic temperature

Summary

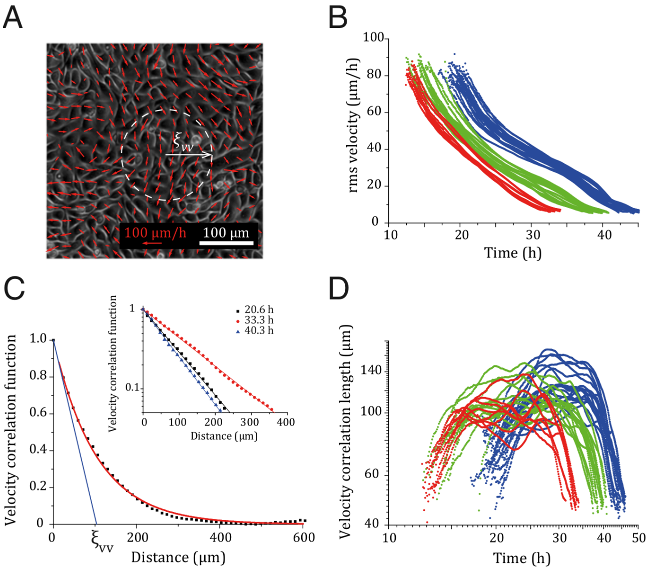
- Extremely persistent active fluids.
- Ballistic but non-trivial single-particle dynamics.
- Power-law dependence of dynamic quantities on the strength of active forces.

- Complete phase diagram?
- Theory for extremely persistent fluids?

Outline

Collective motion cells in a monolayer

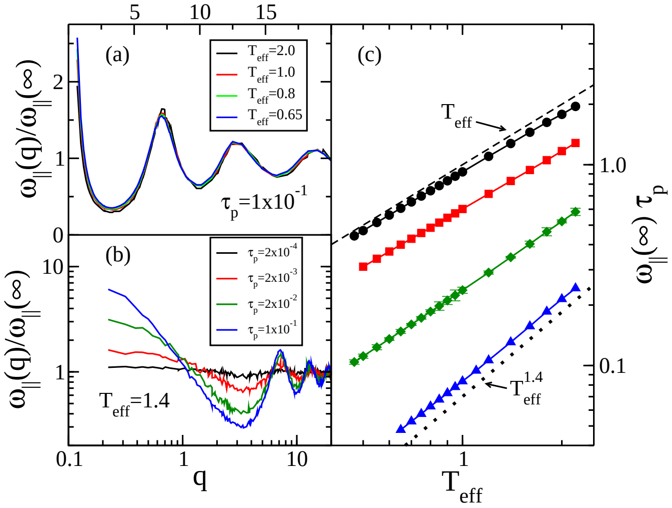
Garcia *et al.* PNAS 2015



Active slow cellular motion:

- Non-trivial equal time velocity correlations

Velocity correlation functions appear naturally in theories for active dynamics

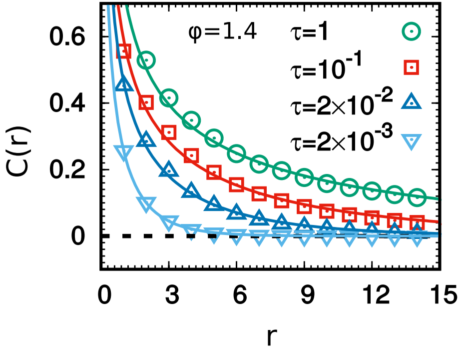


Flenner, GS & Berthier, Soft Matter 2016

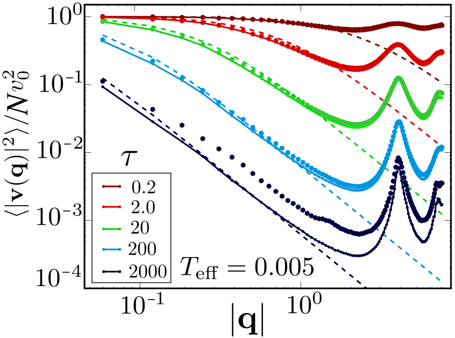
- Panels (a) and (b): non-trivial *equal-time* velocity correlations
- Panel (b): the range of velocity correlations increases with increasing persistence time (which quantifies departure from equilibrium).

T_{eff} - effective/active temperature (one of possible quantitative measures of the strength of the activity)

Velocity correlations were found in active ordered and amorphous solids



Caprini, Marconi & Puglisi PRR 2020



Henkes *et al.* Nature Comm 2020

- Velocity correlations in active ordered solids (left figure) or in active amorphous solids (right figure) increase with increasing persistence time.

Our focus: velocity correlations in active fluids

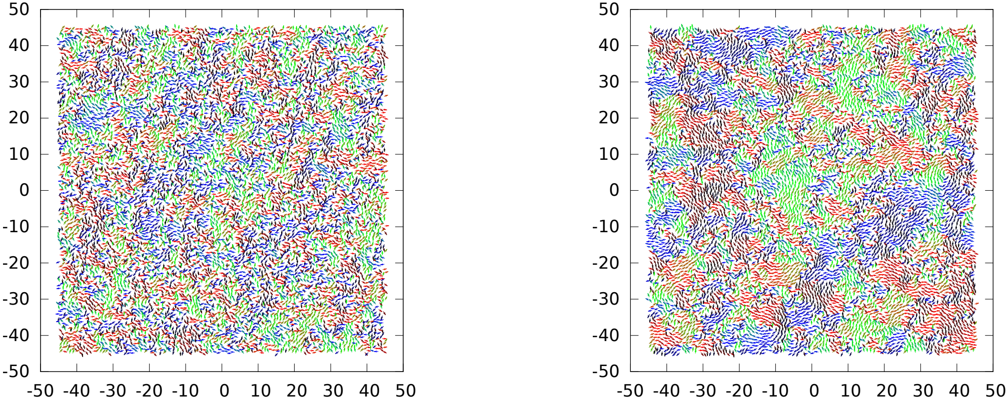
GS & Flenner EPL 2021

Goals:

- Characterize the long-range character of equal-time velocity correlations in active fluids.
- Explain theoretically the long-range velocity correlations.
- Understand their importance for the structure and the dynamics of active matter systems.

- Model: a system of athermal active Brownian particles, with purely repulsive interactions;
  polydisperse, with non-additive cross-diameters to avoid MISP and crystallization.

Qualitative picture → snapshots of configurations



$\tau = 0.14$ $\qquad$ $\tau = 10.0$

Arrows show orientations of the velocities; specific velocity directions are also color-coded.

Quantitative analysis: velocity correlation functions

- Fourier transform of the velocity field:
  $\mathbf{v}(\mathbf{q}) = \sum_j \dot{\mathbf{r}}_j e^{-i\mathbf{q}\cdot\mathbf{r}_j} \equiv \sum_j \left(\gamma^{-1} \sum_l \mathbf{F}_{jl} + v_0 \mathbf{n}_j\right) e^{-i\mathbf{q}\cdot\mathbf{r}_j}$.
- Longitudinal velocity correlation function

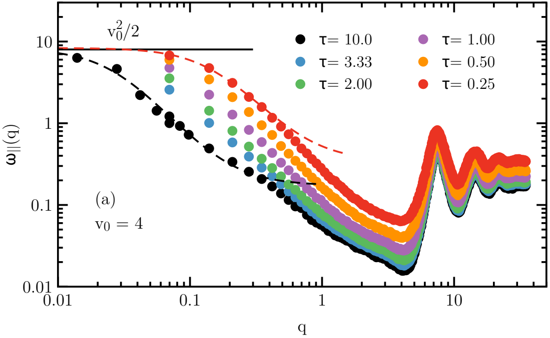
$$\omega_{\parallel}(q) = \frac{1}{N} \left\langle \left|\hat{\mathbf{q}} \cdot \mathbf{v}(\mathbf{q})\right|^2 \right\rangle$$

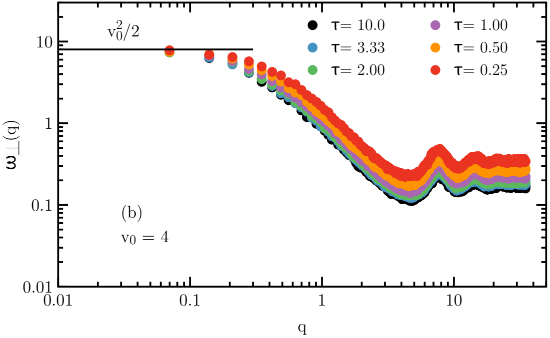
  appears in the theoretical analysis of the dynamics $\rightarrow$ GS, Flenner & Berthier PRE 2015.
- The complementary, transverse velocity correlation function

$$\omega_{\perp}(q) = \frac{1}{N} \left\langle \left|\mathbf{v}(\mathbf{q}) - \hat{\mathbf{q}}(\hat{\mathbf{q}} \cdot \mathbf{v}(\mathbf{q}))\right|^2 \right\rangle$$

  .
- Recall: in “passive” fluids equal-time velocity correlations are trivial but longitudinal and transverse velocity correlations exhibit different time-dependence.

Persistence time dependence of velocity correlations

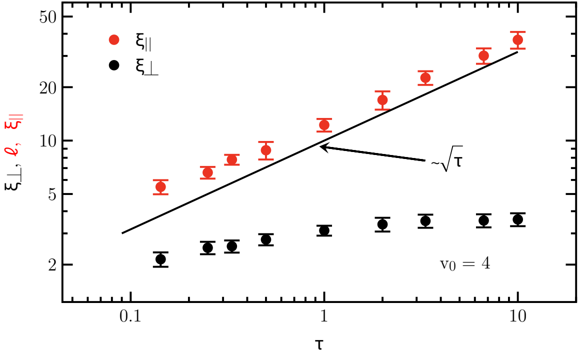




dashed lines: Ornstein-Zernicke-like fits

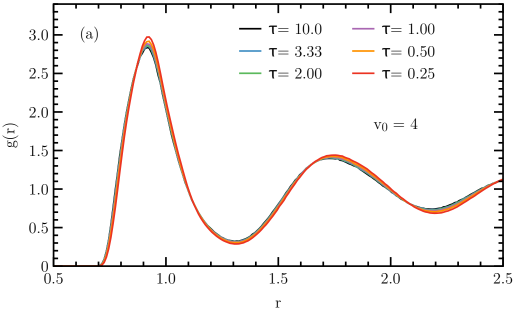
At constant v_0, the range of the longitudinal velocity correlation function increases with increasing persistence time τ, whereas the range of the transverse correlation function changes little.

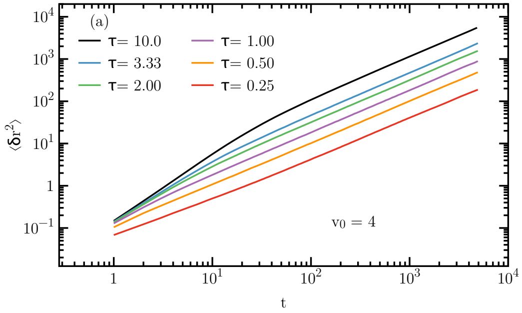
Velocity correlation length



At constant v_0, the longitudinal velocity correlation length increases approximately as $\sqrt{\tau}$, whereas the transverse velocity correlation length grows little and then saturates.

Long-range velocity correlations in active fluids!





Fluid-like local structure ($g(r)$, left figure) and dynamics (mean-squared displacement, right figure) for all persistence times investigated.

Approximate theory

- We re-write the first term in the Fourier transform of the velocity field,

  $\gamma \mathbf{v}(\mathbf{q};t) = \sum_j \sum_{k\neq j} \mathbf{F}_{jk} e^{-i\mathbf{q}\cdot\mathbf{r}_j} + \gamma v_0 \sum_j \mathbf{n}_j e^{-i\mathbf{q}\cdot\mathbf{r}_j(t)}$

  in terms of the interaction part of the pressure tensor,

  $\sum_j \sum_{k\neq j} \mathbf{F}_{jk} e^{-i\mathbf{q}\cdot\mathbf{r}_j} = i\mathbf{q}\cdot \sum_j \sum_{k\neq j} \mathbf{r}_{jk} \frac{\mathbf{r}_{jk}}{2r_{jk}} V'(r_{jk}) \left[\frac{e^{i\mathbf{q}\cdot\mathbf{r}_{jk}}-1}{i\mathbf{q}\cdot\mathbf{r}_{jk}}\right] e^{-i\mathbf{q}\cdot\mathbf{r}_j} \equiv -i\mathbf{q}\cdot\mathbf{\Pi}_v(\mathbf{q};t)$

  where $\mathbf{r}_{jk} = \mathbf{r}_j - \mathbf{r}_k$ and $\mathbf{\Pi}_v$ is the interaction (virial) part of the pressure tensor.

- Approximation:

  $$\mathbf{\Pi}_v(\mathbf{r};t) \approx P_v + \mathbf{I}\left(\partial_\rho P_v\right)\left(\rho(\mathbf{r};t) - \rho\right) \quad \text{where} \quad P_v = \langle \mathbf{\Pi}_v(\mathbf{r};t)\rangle$$

  and $\rho(\mathbf{r};t) = \sum_i \delta(\mathbf{r} - \mathbf{r}_i(t))$ is the microscopic (instantaneous) density.

- Time derivative of $\mathbf{v}(\mathbf{q};t)$ can now be expressed in terms of $\mathbf{v}(\mathbf{q};t)$ itself:

  $$-\gamma i\omega \mathbf{v}(\mathbf{q};\omega) = -\gamma v_o i\omega \mathbf{n}(\mathbf{q};\omega) - \mathbf{q}\left(\partial_\rho P_v\right)\mathbf{q}\cdot\mathbf{v}(\mathbf{q};\omega)$$

Approximate theory: final formulae

- After some manipulations we get an approximate small-wavevector result:

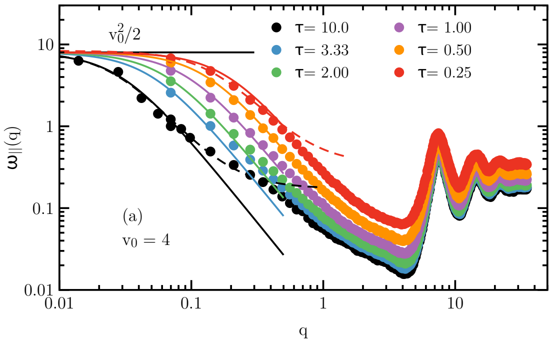
$$\left\langle |\hat{\mathbf{q}} \cdot \mathbf{v}(\mathbf{q})|^2 \right\rangle = \frac{N v_0^2}{2} \frac{1}{1 + q^2 \tau B_v / (\gamma \rho)}.$$

  where $B_v = \rho \partial_\rho P_v$ is the virial bulk modulus of the active fluid.

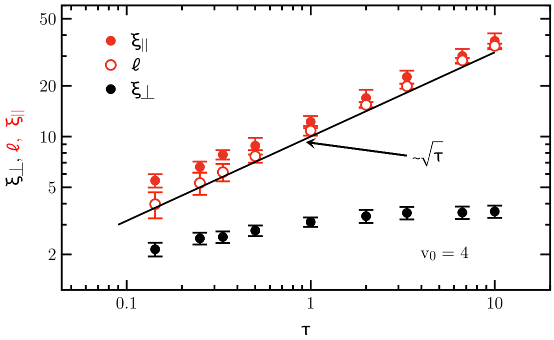
- Longitudinal velocity correlation length:

$$\xi_{||} = \sqrt{\tau B_v / (\gamma \rho)}.$$

Approximate theory: comparison with simulations

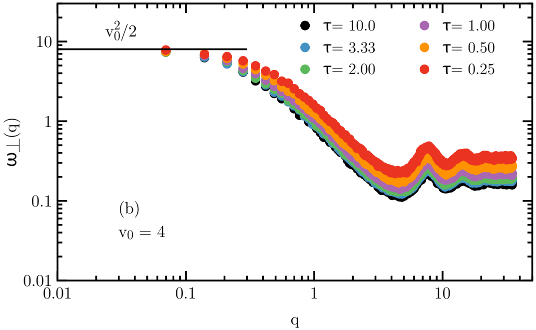


solid lines: approximate theory

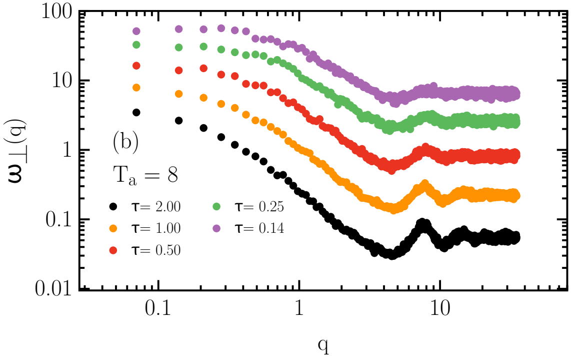


open symbols: approximate theory

Missing piece: transverse correlations

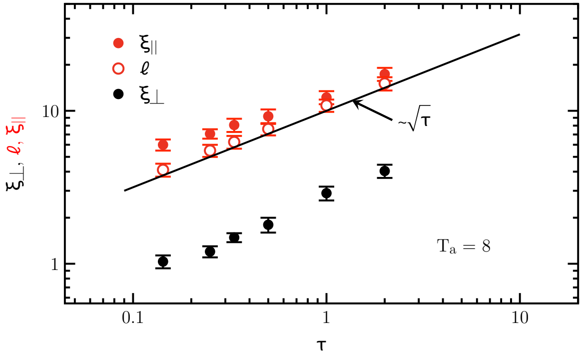


constant self-propulsion velocity



constant active temperature $T_a = \gamma v_0^2/(2D_r)$

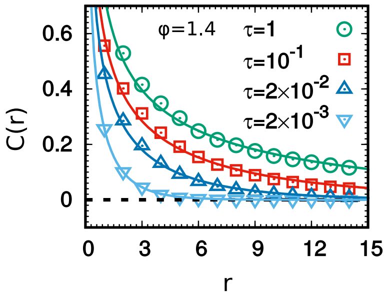
Transverse velocity correlation length increases when monitored at constant T_a



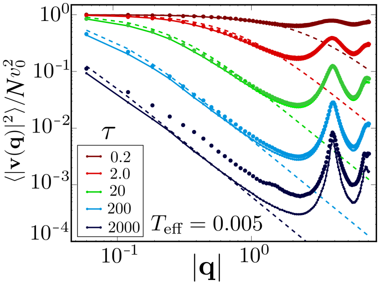
solid symbols: simulations; open symbols: approximate theory

- Approximate theory for velocity correlations in active fluids is missing the transverse velocity correlations.

Earlier theoretical approaches



Caprini, Marconi & Puglisi PRR 2020



Henkes *et al.* Nature Comm 2020

Assumptions:

- $2d$ hexagonally ordered crystal
- small displacements from lattice sites

Assumptions:

- 2d elastic amorphous solid
- velocity correlations in terms of elastic moduli

Summary

- Equal-time velocity correlations are ubiquitous in solid-like and fluid-like active matter systems.
- The velocity correlations increase with increasing persistence of active motion.
- Correlations in solid-like systems can be explained by the combined effect of rigidity and persistent motion.
- Longitudinal correlations in fluid-like systems can be explained by the combined effect of compressibility (bulk modulus) and persistent motion.

- Are long-range equal-time velocity correlations a side effect of the activity or do they modify the structure and/or the dynamics of active systems?