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Dense driven and active colloidal Systems

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We review recent theoretical and numerical developments in shear-driven colloidal suspensions and in dense active-particle systems, with a focus on the dynamics close to the glass transition. The review is guided by the idea that microscopic models of glassy sheared and active colloidal suspensions allow to understand the deep connection, but potentially also the differences between the two modes of driving a system out of equilibrium, in particular through the time-delayed memory effects in the many-particle dynamics.

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Introduction

Soft (colloidal) matter is easily brought out of equilibrium. The mesoscopic size of particles, R, and thermal energy, kT, conspire to cause Brownian motion, make mechanical moduli soft, $G \sim kT/R^3 = \mathcal{O}(Pa)$, corresponding forces weak, and time scales of particle motion large. External forces thus easily interfere with the relaxation of equilibrium fluctuations and cause nonlinear and non-equilibrium response phenomena. Colloidal systems are usually driven by externally imposed mechanical deformation [1] or by particle-intrinsic active forces [2].

Equilibrium is a powerful generic concept. A few well-understood state variables and transport coefficients encode system-specific microscopic details. They enter coarse-grained descriptions that take care of conservation laws and symmetries, and allow to predict large-scale emergent phenomena.

Non-equilibrium systems are less easy to categorize. In principle, each specific mode of driving a system out of equilibrium can break the fundamental symmetries of the system in its own way. This poses a quest: how to identify universalities in far-from-equilibrium systems, how to categorize nonequilibrium phenomena in coarse-grained models, and how to establish frameworks that allow to calculate suitable coarse-grained non-equilibrium transport coefficients from microscopic descriptions. Specifically, a comparison of driven (typically, sheared) and active colloidal systems suggests itself here.

A standard initial level of coarse graining for dense colloidal systems is Brownian dynamics (BD), where one abstracts from details of the driving mechanism and from the solvent. In the overdamped case, particles with positions $r_k (k = 1, \ldots, N)$ interact with potential-derived forces, $F_k = -\nabla_k U$, and are subject to non-equilibrium forces F_k^{ne},

$$dr_k = \mu F_k \, dt + \sqrt{2D} dW_k + \mu F_k^{\mathrm{ne}} \, dt. \qquad (1)$$

Any embedding solvent features only through random forces driving a Brownian motion dW_k with diffusivity D and mobility $\mu = D/kT$. This neglects hydrodynamic interactions (HI) that arise from mass and momentum conservation of the full system.

The form of the nonequilibrium force distinguishes driven systems from active ones. In bulk rheology, one considers a flow gradient $\kappa(r) = (\nabla v(r))^T$ in the suspending fluid, and writes $F_k^{\mathrm{ne}} = \kappa(r_k) \cdot r_k$. In microrheology, F_k^{ne} acts on only one particle. In active systems, on the other hand, $\mu F_k^{\mathrm{ne}} = v_0 n_k$ is typically a force proportional to the particle's self-propulsion velocity v_0 and coupled to some internal degree of freedom of each particle; for instance, its orientation n_k. Various models are available to describe the evolution of these internal degrees of freedom.

How to differentiate between an "external driving force" and an internal "active force" is quite subtle on the level of BD. Both show up as a drift term in Eq. (1), where not even velocity-driven and force-driven cases are easy to distinguish. On an operational level, part of the answer is contained in the flexibility of active forces: active particles can be "responsive" to their environment [3], and

they can be "programmed" to elicit desired single-particle and collective responses [4]. There is a veritable and growing zoo of increasingly sophisticated "microrobots" whose study marks the transition from the "mimetic" science of *observing* motion on the microscale to the "ludic"; one of *manipulating* it.[1]

Technically, global momentum conservation (including the fluid) dictates that active suspensions create quite different hydrodynamic flow patterns [5,6], because their self-generated forces create hydrodynamic force dipoles, and not monopoles. The neglect of HI is, however, common in models of dense suspensions, motivated by the observation that colloidal and molecular glasses share many aspects of their phenomenology. Also, the similarities observed between dense colloidal suspensions and driven systems without a viscous suspending medium, such as sheared granular materials and active microrobots, warrant an attempt at classification without recourse to HI.

The suggestion here shall be thus: a characterization of, and, if possible, a mapping between dense flow-driven and active suspensions should hinge on their collective many-body dynamics — in a microscopic description, on the *memory kernels* describing the non-equilibrium relaxation dynamics. The following review is written from this "glassy" and theoretical perspective, restricting to the simplest models of sheared and active BD systems, and restricting to the slow dynamics of dense suspensions.

The field of driven soft matter is vast and growing, and thus the following is necessarily a subjective perspective and incomplete. Recent reviews cover the physics of dense suspensions [7], and the rheology of aging soft glassy materials [8], of active systems [9], and of dense granular flows [10]. To call the field of work on active systems vast is an euphemism — a recent meta-review [11] alone lists over 1000 reviews and books on the topic. Both soft matter [12] and active matter [13] have been focal points of recent "roadmap" papers. Here we also focus on *microscopic* models; for a recent hydrodynamic approach to active matter, see, e.g., Ref. [14].

Theoretical and simulation techniques

The toolbox of non-equilibrium statistical physics still keeps evolving; this includes exciting discoveries that have not yet made their way into the "glassy" literature and are thus skipped over here. Let us just mention progress on path-integral formulations [15,16], or a recently discovered "hidden canonical gauge symmetry" under essentially arbitrary shifts of particle positions [*17] bearing a thought-provoking analogy of the deformation fields of disordered solids.

Projection operators and memory kernels

Projection-operator techniques play a prominent role in statistical physics — see Ref. [**18] for a thorough review. They place dynamical correlation functions of observables based on Eq. (1) at the center. A reduced description in terms of a few "relevant" dynamical variables is obtained, whose non-Markovian dynamics is captured in memory kernels of integro-differential equations; loosely speaking, they turn the Langevin equation Eq. (1) into a generalized Langevin equation (GLE). In the context of BD, let us denote by $\varrho^l_{t_0}(\vec{q},t)$ the result of propagating a density fluctuation $\varrho^l(\vec{q}) = \sum_{k=1}^{N} f_l(x_k)\exp[i\vec{q}\cdot\vec{r_k}]$ of the N-particle system (with particle positions $\vec{r_k}$ and some function f_l to capture the dependence on internal degrees of freedom x_k of a particle such as activity, orientation or other), from the starting time t_0 to t. One often deals with equations of the structure

$$\partial_t \varrho_{t_0}(t) + \omega_{t_0}(t)\varrho_{t_0}(t) + \int_{t_0}^{t} m(t,t'';t_0)\times \\ \times\Big\{(\partial_{t''} + \widetilde{\omega}_{t_0}(t''))\varrho_{t_0}(t'') - f^R_{t_0}(t'')\Big\}dt'' = f^R_{t_0}(t) \tag{2}$$

to be read as a matrix equation in the indices l. Here, f^R is a generalized force obtained from the projection-operator formalism; ω and $\widetilde{\omega}$ are matrix elements of the time-evolution operator; and m is the memory kernel. It is an intriguing and not fully solved problem how to determine the memory kernel given knowledge of the time evolution, e.g., from simulation — the inversion of Volterra integral equations is a hard inverse problem. Some progress could be made with the help of neural networks (NNs) [19].

Memory kernels in this framework become the key object through which to understand both the nature of collective effects, and the signatures of non-equilibrium driving forces. In experiments, specific features of the memory kernel allow to distinguish non-equilibrium from equilibrium GLE [*20]. From a theoretical point of view, different classes of memory kernels appear in different nonequilibrium situations: (i) driving by shear induces a time-dependence set by the external force; (ii) directional driving as in active microrheology causes a split into parallel and orthogonal contributions; (iii) internal degrees of freedom of active particles cause a delicate matrix structure of couplings between different l.

The integration through transients (ITT) approach connects memory kernels and correlation functions to non-equilibrium transport coefficients (much like Green-Kubo (GK) relations do in equilibrium) [21]. In essence, ITT describes the time-dependent non-equilibrium average of an observable f,

[1] I owe these terms to digital archeologist Bernard Frischer.

$$\langle f\rangle(t) = \langle f\rangle_{t_0} + \int_{-\infty}^{t} \langle f_{t''}(t) g^*_{t_0}(t'')\rangle_{t_0} \qquad (3)$$

where g^* is the conjugate variable to f (generalizing this notion from linear-response theory), and the integral is over a response function that is intimately connected to the memory kernels appearing in Eq. (2). One of the successes of ITT combined with the mode-coupling theory of the glass transition (MCT) is the derivation of constitutive equations for sheared colloids on the basis of Eq. (3). The same approach has been used in active systems, where ITT-MCT addresses non-trivial correlation functions [21] that are forbidden by symmetry in the passive case [**22,23].

The ITT approach to incorporate shear has recently been demonstrated also in direct comparison with granular rheology experiments [*24]. The quiescent agitated granular system is itself out of equilibrium, driven by forces that are modeled in the theory not much different than a thermostat. On this basis, MCT predicts flow curves that are governed by the same physical mechanisms as in colloidal suspensions; only at very high shear rates the difference is felt in the appearance of a "Bagnold" regime, $\sigma \sim \dot\gamma^2$, unique to granular materials. Agreement between experimental data and the MCT-like theory is very good (Figure 1), even though the experiment cannot guarantee the system to be in a fully homogeneously sheared state devoid of shear bands.

Glass-forming fluids and amorphous solids

For glass-forming fluids, MCT is still one of the most predictive, and thus also one of the most detectably falsified theories. It plays a specific role in combination with ITT, and hence for the dynamics of driven dense colloidal suspensions.

Hallmarks of the amorphous solid are peculiar low-frequency modes and elastic stress-correlations due to localized excitations, linked to Eshelby stress fields and a properly augmented elasticity theory [*25]. Recent advances to link the fluid-like and solid-like approaches to glassy phenomena came from revisiting old issues surrounding MCT and its approach to continuous solidity transitions [**26] (see also Ref. [27]). This work opens an exciting avenue into microscopic descriptions of glass and jamming transitions.

Numerical methods and machine learning

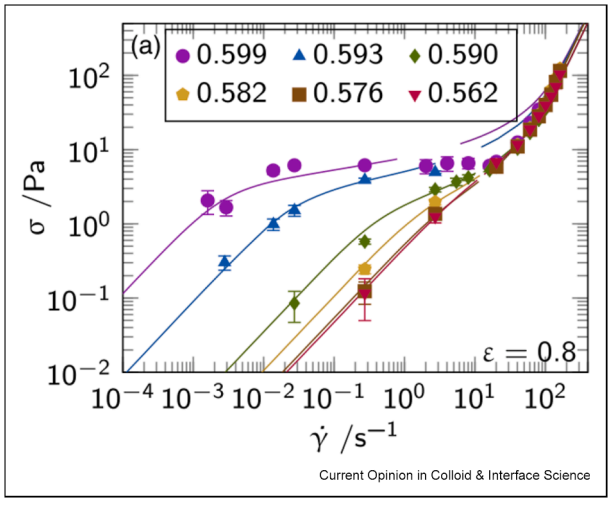
A major obstacle for molecular dynamics (MD) simulations of glass formers is the long equilibration time. The swap Monte Carlo (SWMC) method is now the de facto standard for the generation of equilibrated starting configurations, bringing MD simulations to previously inaccessible regions of the dynamics [28,29], and allowing to test theoretical predictions connected to replica theory and growing length scales deep in the glass [30].

Besides traditional simulations, machine learning (ML) tools play an increasing role in the study of the dynamics of structural glasses [*31]. Much work has gone into decoding specific particle rearrangements that are predictive of the slow relaxation dynamics [32,33]. A key quantity is the propensity of a particle to move: Given some static configuration, one constructs the "isokinetic ensemble"[2] by reassigning random velocities to the particles, and thus assigns to each particle in the configuration its mean displacement over some given time in this ensemble. Dynamic heterogeneity can be learned on this basis [34,35].

A next step along this route would be the application of machine-learning tools to classify different equilibrium systems. Note that NNs can help determine effective interaction [36] or even dynamical laws [37] in driven systems. Since non-equilibrium systems produce entropy, a non-trivial probability flux arises in the Fokker–Planck equation (FPE) associated to Eq. (1). Analyzing the FPE, a notoriously high-dimensional partial differential equation, is difficult; recent advances come from the application of generative modeling, allowing to learn the entropy production rate of active swimmers in NNs [*38].

The key point here is the *dynamics*, while many materials-physics applications of NNs focus on the statics problem of mapping interparticle interaction potentials.

Figure 1



Flow curve (stationary stress as function of shear rate), for a fluidized granular system: experimental values (symbols) at packing fractions as indicated, compared with predictions based on ITT and MCT (lines). From Ref. [*24] (figure published under CC BY 4.0).

[2] The author suspects that due to finite numerical precision and molecular chaos, this isokinetic ensemble is not in fact one where particle positions remain fixed; but for the purpose here this should not be relevant.

Machine-learned interaction potentials (MILPs) are now state of the art and enable fully microscopic studies of "real" molecular glass formers. An application to toluene indeed shows good agreement with experiment [*39]; however, close to the glass transition, large deviations appear in the relaxation times. This could point out deficiencies of the underlying density-functional theory (DFT) training the MILP. It could, however, also point to the high sensitivity of glassy dynamics to interaction details: even small changes in the static structure of the system can cause huge changes in the dynamics. While in principle detectable by an NN [40], a MILP might require additional training based on dynamical data in order to be accurate enough for that reason.

Sheared suspensions

Macroscopic flow

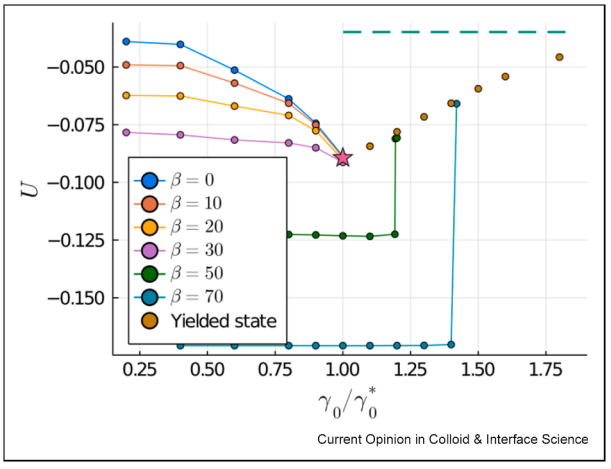
Shear-induced yielding of glasses is a process that is highly dependent on the preparation history of the glass, i.e., on the quenching and subsequent annealing. Simulations using SWMC have established the relation of preparation history to ductile and brittle yielding, and the nature of strain localization close to, and far below, the MCT crossover [*41] (see Figure 2).

We focus in the following on oscillatory shear, because of its surmised role in an analogy to active matter. Glasses under oscillatory shear can (effectively) remain solid, or yield, depending on the amplitude of the driving. This is an interesting non-equilibrium state transition that has received a lot of attention (reviewed in Ref. [42]), including recent simulations [43] and mean-field-like models [44,45] that look into the role of spatial heterogeneity and shear banding.

The preparation history plays a crucial role in yielding under oscillatory shear: different initial annealing brings glasses to different inherent-state energy levels. Oscillatory shear has two effects, depending on whether one exceeds a critical yielding amplitude or not: small-amplitude shear helps annealing of badly annealed glasses, while large shear pushes the system out of these energetic minima. Initially found in simulations, coarse-grained models describe this phenomenology (Figure 3) [46].

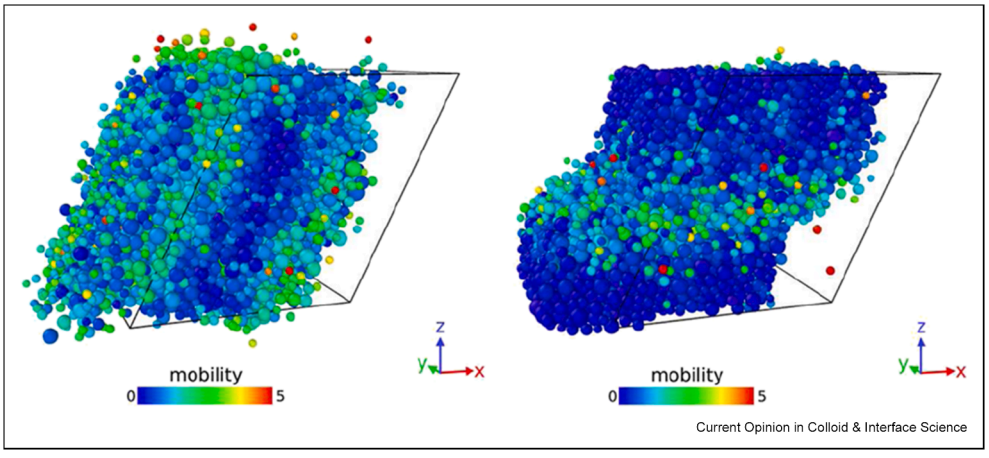
Large-amplitude oscillatory shear (LAOS) in the yielded state provides a wealth of detailed information

Figure 3



Energy U in oscillatory shear as a function of the strain amplitude γ_0, for initially poorly (small β) and well annealed (large β) glasses, according to a mean-field model. Mechanical strain helps annealing of poorly annealed glasses (U decreases initially with γ_0), but when yielding sets in U increases. For thermally well-annealed glasses, the yielding transition is discontinuous. Reprinted with permission from Ref. [46], Copyright (2022) by the American Physical Society.

Figure 2



Particle mobility (color-coded) during yielding of an amorphous solid just below the MCT transition T_c. Sample-dependent shear bands form that can be horizontal or vertical with respect to the deformation direction. Reprinted from Ref. [*41], with the permission of AIP Publishing.

about glass-forming suspensions, in particular if one is able to quantify the higher harmonics of the response. [47]. Connected to this, oscillatory-sheared glasses store information about reversible and irreversible parts of the deformation [48,49], and thus information on their deformation history [50]; with fine-tuned interparticle interactions, this leads the way towards trainable metamaterials [51].

Microrheology

For a more localized response, microrheology is the tool to go. It needs a detailed, microscopic understanding of the host fluid, however. MCT has been able to tackle the response in active microrheology, where the tracer particle is pulled by a constant external force. The elastic recoil turns out to reveal a lot about the fluid and glassy response [52] — this also connects to the mean back relaxation, a quality proposed to experimentally distinguish equilibrium from non-equilibrium memory effects [*20,53]. On a macroscopic level, it relates to the recoverable strain in LAOS [49].

Microrheology (passive and active[3]) is a convenient experimental tool, especially in biophysics. Thus, a theoretical understanding of how the non-equilibrium nature of the host system makes itself felt in the observables pertaining to the probe particle will provide further guidance regarding the link between shear-driven and active glassy suspensions.

Active glassy matter

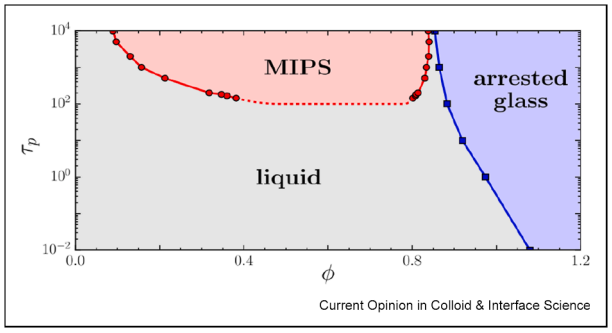
Glass transition and phase separation

Earlier work on collective dynamics in active suspensions has focused on motility-induced phase separation (MIPS): a phase-separation-like non-equilibrium clustering phenomenon driven by active forces (see, e.g., Ref. [54]). One relevant mechanism for MIPS is that the renormalized swim speed $v(\varrho)$ decreases below the bare self-propulsion velocity v_0 with increasing density ϱ. A microscopic description of this quantity hinges on the ITT-Eq. (3) [21].

A more recent shift is towards even denser states, the active glass. Simulation studies investigate diagrams in the density—activity plane where both the region of MIPS and the active glass can be identified [**22**,55] (see Figure 4).

Experimental methods of driving artificial active particles (through chemical gradients, electromagnetic or light fields, etc.) become difficult at high densities: external energy input to maintain the active motion by light assumes that each particle can be seen, so that "shadowing" by neighboring particles changes the activity depending on the density. Also, depletion of chemical fuel by one particle will cause a change in activity of the others. This cross-talk makes an experimental realization of the artificial active-particle glass significantly more complex than the usual theoretical models. Most experimental studies are also limited to quasi-two-dimensional settings. Few experimental realizations of synthetic three-dimensional active matter exist [56,57] (and unless in microgravity, the torque on Janus particles is not generally negligible, see Ref. [58] and SI therein). However, the discovery of glass-transition-like phenomena in tissues (speculated to be one of the driving forces behind wound healing or cancer metastasis; see e.g. Refs. [59,60]), bacterial colonies [23] or even biofilms, has spurred renewed interest in the active glass experimentally [61], but also in the development of new computational models like those based on the Voronoi fluid [62].

Figure 4



State diagram for dense active suspensions, including both motility-induced phase separation (MIPS) and a glassy state. Reprinted with permission from Ref. [**22], Copyright (2022) by the American Physical Society. See also Ref. [**55] for a similar figure.

Adding self-propulsion forces to the passive glass does not immediately cause fluidization, but a transition from "active glass" to a yielded state appears at finite activity. This motivates looking for an analogy to yielding under oscillatory shear. Note that just as with shear-driven systems, and despite colloquial imprecision,[4] one has to carefully distinguish glass- and jamming transitions [63], based on whether rigidity appears discontinuously or continuously. An interesting toy model in this context is also a glass out of active particles whose activity is infinitely persistent (termed "extreme" active matter) [64,65].

Beyond Brownian dynamics

An ongoing trend is to consider not just active colloidal particles, but larger "microrobots" [66]. Even some

[3] The term "active" here can be confusing: active microrheology refers to a passive system under a specific external driving; passive microrheology is a technique often used to understand active systems.

[4] The term "unjamming" is often used to describe the mobilization of cells in the fluidization of tissue, not specifying whether it might in fact be a "devitrification" transition.

simple toys behave dynamically like standard active-particle models, as an analysis of trajectories in terms of dynamical correlation functions reveals [67] (Also, the dynamics of transport by ants has been mapped onto a simple self-propelled particle model [68].)

But these systems are not overdamped like the standard colloidal suspensions, and thus they introduce new degrees of freedom that alter the dynamics. Theories thus need to be extended to include inertial active-matter models [69], orientational degrees of freedom, chirality [70,71], aligning and self-aligning [72], activity governed by multiple internal states [73], or non-reciprocal interactions [74].

This poses an interesting question in the context of glassy dynamics: in passive systems, the microscopic details of the dynamics (Brownian or Newtonian) do not matter for the glass transition. This is not a priori evident for the role of inertia or driving model — such as active Brownian particle (ABP), active Ornstein-Uhlenbeck process (AOUP), or run-and-tumble particle (RTP), to name a few — in the active glass. Such a dependence on individual-particle details would mark the active glass as truly different from the (possibly deformed) passive one. This extends from the glass transition to other dynamical transitions, such as jamming or localization in the Lorentz-gas model [75].

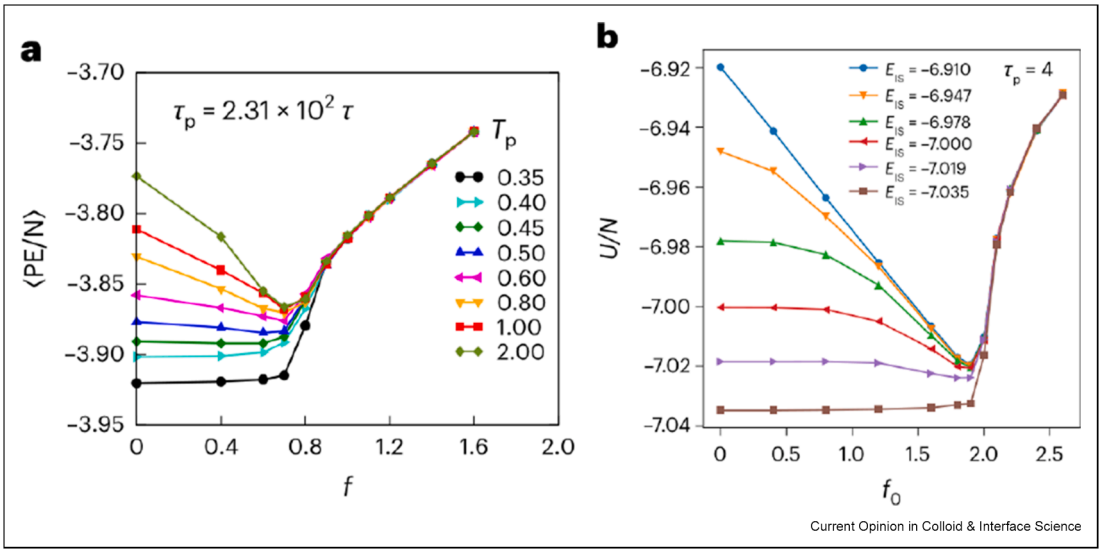
Sheared/active analogy

The power of physical descriptions is often to investigate seemingly different objects — such as sheared passive and active systems, and to discover the common mechanisms that underpin both (if any). Near the glass transition, a superficial analogy between active, granular, and colloidal systems is hard not to notice: The stationary dynamics bears the hallmarks of slow structural relaxation typical for the colloidal glass transition, and all these systems can yield in response to shear and active forces in similar ways.

Despite obvious similarities, differences also exist — most prominently, the continuously sheared glass is always fluidized, while the active glass remains a glass if the activity is not too strong, more akin to what is observed in oscillatory shear or active microrheology.

A straightforward idea starts by recognizing that most active models introduce *two* parameters associated with the driving force: a strength and a persistence time. Amplitude and frequency in oscillatory shear play a similar role [76]: Naïvely, observe the nonequilibrium driving term in Eq. (1), within the ABP model (2d, for simplicity), $\mu F_k^{\mathrm{ne}} = v_0 n_k = v_0(\cos\phi_k, \sin\phi_k)^T$, and in oscillatory shear, $\mu F_k^{\mathrm{ne}} = \gamma_0\omega\cos(\omega t)y e_x$; here ϕ_k is the active particle's orientation, while e_x is the direction of the shear gradient. Roughly speaking, the ABP loses sense of its direction after the correlation time of the random angle motion, set by its rotational diffusion coefficient, $1/D_r$; this suggests at least qualitatively the mapping $D_r \leftrightarrow \omega$. Similarly, the Péclet number relevant for oscillatory shear, $Pe_\omega = \gamma_0\omega\tau_0$, can be related to the free-active particle one, $Pe_0 = v_0^2/D_r D_0$, where $\tau_0 = \sigma^2/D_0$ (for a particle of size σ). This suggests to map $\gamma_0\omega \leftrightarrow v_0^2/D_r\sigma^2$. For dense systems, MCT instead predicts the dynamics to be governed by the Péclet number

Figure 5



Inherent state energies E_{IS} (or energy per particle U/N) as in Fig. 3, but for active systems from simulations of slightly different model systems (a: from Ref. [**78], published under CC BY 4.0; b: from Ref. [**79], reprinted with permission and published under CC BY-NC-ND 4.0), as a function of the activity parameter f entering the respective models.

$Pe_t = v_0\sigma/D_0$ that enters the memory kernel [21], so that a mapping $\gamma_0\omega \leftrightarrow v_0/\sigma$ appears equally plausible. Which is more relevant should be something to resolve with a fully developed microscopic theory.

This analogy is worth pursuing, because for oscillatory-shear-driven amorphous solids, it is known that memory of past deformations can be imprinted and read out, potentially turning the corresponding systems into functionalizable materials [77]. Recent simulation studies explore the analogy and parameter mapping in more detail [**78**,79]: the dependence on the initial quench and the annealing behavior of active glasses was found to be strikingly similar to the one observed in oscillatory shear — compare Figure 5 for active glasses with Figure 3 for oscillatory sheared glasses (Active forces even correspond more to stress-controlled shear than to strain-controlled shear [**79].) So far, these are numerical observations. They call for a theoretical exploration.

Outlook

Discovering universality across system classes that are, on some microscopic level, very different, is one of the most fruitful prospects in statistical physics. However, one should bear in mind that to date, the analogy between sheared colloidal, granular, and active glassy materials is still anecdotal. The microscopic theoretical framing of the observed analogies and differences should be a big step in understanding non-equilibrium dynamics of soft-matter systems in general. In particular, when it comes to collective, emergent behavior, one promising way to approach the topic appears to be a closer look at the features of the respective GLEs and their memory kernels.

Memory integrals, by virtue of ITT, lead to nonequilibrium transport coefficients. Thus, ideally, a theoretical description able to pinpoint the relevant characteristic features of memory kernels in shear-driven systems and those relevant for active systems will also be able to provide important guidance to coarse-grained models of nonequilibrium systems. At some point, this will surely also include living systems as the more complex cousins of active systems. For example, analogies like the ones shown in Figures 3 and 5 aside, field-theoretical approaches to oscillatory-sheared [46] and active [80] systems appear quite different. It is thus still an open exciting task to disentangle the phenomenology in driven colloidal systems and their microscopic origins, combined with continuum-model discovery guided by experimental data and helped with machine learning [81].

A topic that has been neglected so far in seeking this sheared—active analogy is the boundary conditions. Nonequilibrium systems can in principle strongly depend on the boundaries — a fact that is well known in particular for active systems. In the dense system, one hopes to be able to perform the microscopic coarse-graining step based on the notion of a homogeneous state, but the appearance of shear bands in driven colloids or MIPS in active colloids indicates that there might be a non-trivial coupling of macroscopic gradients back into the microscopic dynamics. Inhomogeneous extensions of microscopic theories can potentially address this. Conversely, confined systems might reveal more fundamental differences between active and sheared dense systems.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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