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Hyperuniform dispersal of non-Brownian particles in viscous liquids
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The goal of this project is to establish methods for homogenizing suspensions of hard particles in extremely viscous liquids. Suspensions appear in a wide range of chemical, biological, and industrial settings, and dispersing particles throughout a fluid is an important challenge for a wide range of purposes and settings. For instance, concentrated suspensions exhibit dramatic mechanical responses including shear thickening, which can clog or damage equipment. Breaking up aggregates and re-suspending settled particles can ease processing demands. With the support of the American Chemical Society Petroleum Research Fund (PRF) over the last year (Sep. 1, 2017 to Aug. 31, 2018), we have made significant progress towards this goal. Below, I summarize key results during this funding period in three thrusts of the project. Finally, I summarize the impact of this funding on my students and my scientific career.

(1) *Self-organized hyperuniformity*. The aim of this thrust was to discover how shear protocols can be used to control the spatial distribution of non-Brownian particles that are settling under gravity in a viscous liquid. We used a simulation model, originally developed by Laurent Corté and coworkers, which has been found to reproduce the self-organization processes that occur in experiments. Through careful systematic study, we established a new dimensionless parameter that controls the vertical density gradients of the particles in the steady state, and we provided analytic arguments to rationalize the strength of these gradients. At low sedimentation speeds, the density gradients are smallest; to dig deeper we measured the density fluctuations across a wide spectrum of lengthscales using both real-space and Fourier-space methods. Figure 1 shows our main result: at sufficiently low sedimentation speed, density fluctuations vanish at long lengthscales, indicated by a vanishing structure factor S(k) as the wavevector k approaches 0. These findings have unveiled a new method for obtaining hyperuniformity that does not require fine tuning, so it can be applied in the same way regardless of the local volume fraction in a given application, and can be used in cases where the volume fraction cannot be measured easily.

Beyond its practical value, our work addresses fundamental questions in the field: our numerical simulations over a wide range of parameters show that the critical state is robust at producing hyperuniform distributions under another external drive (gravity). Furthermore, our quantitative theoretical arguments show how hyperuniformity breaks down in these systems. We established a phase diagram for homogenization in a suspension as a function of the lengthscale of interest. Surprisingly, we find that although the overall density of the re-suspended pack is governed by a single dimensionless number, there are two additional expressions that determine the vertical density gradients and the breakdown of hyperuniformity. These results were published in the current funding year in Nature Communications.

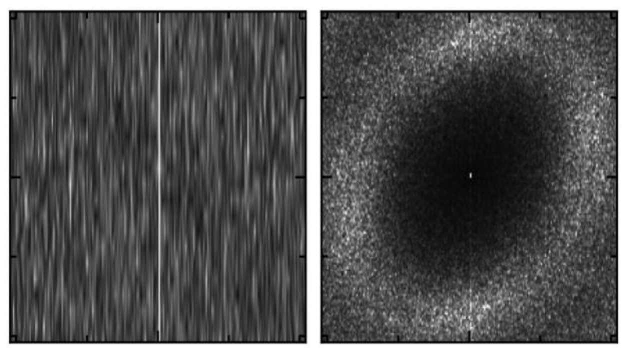

FIG 1. Structure factor, S(k), in simulations of a cyclically-sheared suspension of particles that are sedimenting under gravity. At high sedimentation speed (left image), nothing remarkable occurs. At low sedimentation speed (right image), density fluctuations vanish at long lengthscales.

(2) *History-dependence and memory formation in viscous suspensions.* Because they have by definition fallen out of thermal equilibrium, non-Brownian suspensions are acutely sensitive to shear protocols or other driving that is applied to them. Such suspensions are therefore constantly forming and erasing memories of their past. Understanding this memory formation on a fundamental level will enable rational design of methods for handling and homogenizing suspensions of hard particles in viscous liquids.

To build a deeper fundamental understanding of memory effects in such suspensions, we considered the effect of cyclic driving at two different strain amplitudes. We then abstracted the various states that may occur in terms of the memory content of the sample, as well as the transitions between them. We thus established a simple graph structure that underlies the evolution of suspensions. The advantage of this approach is that the graphs are sufficiently general that they may also be applied to related systems such as sheared amorphous solids, which exhibit different memory behaviors. By identifying defining features of these transition graphs, we provided simple diagnostic tools for distinguishing different classes of memory behaviors. To show the validity of this approach, we constructed minimal yet physically-motivated models that exhibit these various behaviors and possess the same distinctive transition graph structures. Notably, one of these models (the "park bench" model) solves a significant outstanding problem regarding the role of criticality, nonlinear diffusion, and disorder in a type of memory that occurs in cyclically sheared suspensions (i.e., multiple transient memories). This work led to the submission of a manuscript entitled "Minimal descriptions of cyclic memory behaviors", which was jointly authored with collaborator Nathan Keim.

This grant also supported my work on a review article on memory formation in matter, which I am co-authoring with several colleagues. This manuscript will be submitted in the near future.

(3) *Experiments.* As can be seen from the results above, we have been making considerable progress on the computational and theoretical side of this project in the current funding year, including the publication of a manuscript in Nature Communications, the submission of a manuscript on transition graphs for cyclic memory behaviors, and the co-authorship of a review article on memory formation in matter. One side-effect of this major activity is that the experimental component of the project is not as far along. These experiments will be a significant focus in the coming year, and an excellent senior graduate student will transition her effort to this project. We have already constructed an apparatus for testing cyclic shear protocol in experiments, and we built a custom shear cell and imaging system around a strain-controlled rheometer that we installed in the lab. We will apply these shear protocols to suspensions that we have made from 125-micron PMMA spheres in a ternary mixture of water, zinc chloride, and Triton X-100 to match the index of refraction of the particles.

Impact on my students and my career. This grant provided support for one graduate student, Jikai Wang. This is the second year Jikai has been funded on this project; he continued to manage the numerical simulations and analyze all the data. He presented his latest results at the 2018 APS March Meeting in Los Angeles. These funds also provided a portion of my summer salary. Finally, as this was my first research grant, it was instrumental in helping me establish my independent research program, leading to my first major publication where I am listed as the main PI.