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GLASSY DYNAMICS AND IDENTITY CRISES IN HARD-PARTICLE SYSTEMS

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EXECUTIVE SUMMARY

Glass formation is a well-known outstanding mystery in the physical sciences: It is a phenomenon with no canonical thermodynamic description, yet one with wide-ranging industrial applications, occurring in materials as disparate as superconductors and sand. We utilized our Blue Waters allocation to help shed light on this murky topic, by simulating and investigating the dynamics of several glass-forming soft matter systems on time scales spanning about six orders of magnitude. We used our open-source simulation toolkit HOOMD-blue [1], scalable software built to run in parallel using domain decomposition on both CPUs and GPUs. We found that all systems studied displayed dynamical signatures indicative of cooperative rearrangement and consequent dynamic heterogeneity, both hallmarks of glass-forming behavior [2,3]. We examined local structure in these systems on other machines, and found a link between glass formation and a structural identity crisis, or competition between various local structural motifs.

RESEARCH CHALLENGE

Soft matter systems, in which thermal fluctuations are strong enough to drive particle rearrangements, are capable of self-assembling into a staggering variety of simple and complex crystalline structures. Often, however, no such assembly occurs, and the system remains disordered, displaying instead dynamical signatures characteristic of glass-forming materials. To develop robust methods of self-assembly, then, an understanding of glass formation in soft matter systems is needed. Beyond the realm of soft matter, glasses have proven useful materials to humans for over four millennia, for applications ranging from dinnerware to phase-change memory devices [3].

Despite this ubiquity, a clear picture of the thermodynamics and structural change undergirding glass formation remains unknown. This is due in large part to the significant slowing down of any system as it approaches the glass transition [2,3], requiring investigations of glass formation to resolve, dauntingly, system dynamics on time scales that vary by orders of magnitude. We were able to tackle this problem using our highly parallel simulation package, HOOMD-blue, and by accessing both the Cray XK7 nodes hosted on Blue Waters and its very generous storage capabilities.

METHODS & CODES

To help shed light on the dynamics and structural underpinnings of glass formation in soft matter systems, we chose to simulate and analyze nonassembling monatomic systems of hard polyhedral particles, with no interactions aside from those of excluded volume. We chose particle shapes consisting of tetrahedra with varying degrees of edge and vertex truncation. These are simple systems mediated solely by entropy maximization. Entropy maximization is known to drive phase transitions in a variety of systems, from those composed of hard spheres [4] to hard rods [5] to colloids and polymer depletants [6]. Recently, it was shown that entropy maximization drives systems of hard polyhedral particles toward a vast array of ordered structures upon compression [7], and a mechanism involving emergent, directional entropic forces was proposed to explain this ordering [8]. In [7], however, a number of systems displayed no order at all, instead exhibiting the signature slow dynamics of glassiness. In these cases, directional entropic forces drove systems toward glassy disorder rather than crystallization. Our project further explores these entropic effects.

We used the hard particle Monte Carlo (HPMC) simulation method [9] included in HOOMD-blue to sample our systems in the isochoric ensemble in equilibrium. In brief, this classic simulation method proposes trial moves for particles, and accepts or rejects them based on an acceptance criterion that dictates rejection if there are any overlaps created in the system, and acceptance otherwise. We simulated systems of 4096 particles on a single GPU at a variety of densities, and subsequently measured structural and dynamical information. To gather trajectory information at a wide range of time scales for later analysis, we wrote our system trajectories to disk remarkably frequently—every 10 Monte Carlo (MC) sweeps—and collected data for the far longer period of 100 million MC sweeps. We produced trajectories as large as 3.25 terabyte (TB) MC sweeps per simulation, each containing about 10 million simulation frames.

RESULTS & IMPACT

For each trajectory, we measured several dynamical parameters known in the glass community to be indicators of glass-forming behavior, and confirmed that our hard particle systems are indeed glass-formers. These were the mean-squared displacement of all particles in the system, the van Hove autocorrelation function, the non-Gaussian parameter characterizing the particle displacement distribution, and the self-part of the intermediate scattering function [2,3]. The mean-squared displacement and self-intermediate scattering function for a sample system are shown in Fig. 1. Plateaus in both parameters at intermediate times and increasing densities indicate caging behavior typical of glass-formers; at these times, particles are trapped on average by a cage of their surrounding neighbors, and only escape due to cooperative structural rearrangements and consequent dynamic heterogeneity. The mean-squared displacement shows this caged behavior directly, while the plateau in the self-intermediate scattering function (a Fourier transform of the real-space van Hove autocorrelation function) shows a slowing-down of the relaxation of the system.

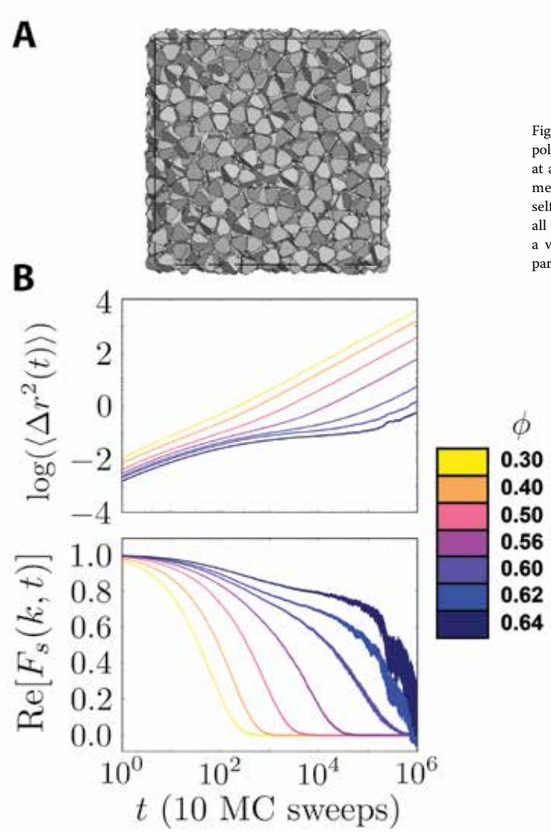

Figure 1: A glass-forming system of hard polyhedral particles. (A) A system snapshot at a volume density of 0.62. (B) Both the mean-squared displacement and the self-intermediate scattering function of all particles in the system over time, for a variety of densities. Plateaus in both parameters are indicative of caging.

We have determined via simulations on other platforms that observed glass-forming tendencies in these systems are related to competition between local structural motifs, mediated by entropic forces; future work will strive to make more concrete the connection between local structure and observed heterogeneous dynamics.

WHY BLUE WATERS

HOOMD-blue HPMC's parallel nature on both CPUs and GPUs allowed us to take unique advantage of the computational resources on Blue Waters, and, in particular, its GPU resources. We also produced tens of TB of raw data that we needed to sift through later, and we were able to accomplish this by storing it all on Blue Waters' generous Online file system, analyzing it, and storing resultant analysis results somewhere more permanently. Without the 500 TB quota afforded to us by Blue Waters, we would find it far more difficult to explore such fine-grained dynamical phenomena.

Erin Teich is a fifth-year Ph.D. student in applied physics at the University of Michigan. She is working under the direction of Sharon C. Glotzer, and expects to graduate in June 2018.