Hydrodynamic Coupling Melts Acoustically Levitated Crystalline Rafts

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The acoustic levitation of small particles provides a versatile platform to investigate the collective dynamical properties of self-assembled many-body systems in the presence of hydrodynamic coupling. However, acoustic scattering forces can only generate attractive interactions at close range in the levitation plane, limiting self-assembly to rafts where particles come into direct, dissipative contact. Here, we overcome this limitation by using particles small enough that the viscosity of air establishes a repulsive streaming flow at close range. By tuning the size of particles relative to the characteristic length scale of the viscous flow, we control the interplay between attractive and repulsive forces. In this novel granular raft, particles form an open lattice with tunable spacing. Hydrodynamic coupling between particles gives rise to spontaneous excitations in the lattice, in turn driving intermittent particle rearrangements. Under the action of these fluctuations, the raft transitions from a predominantly quiescent, crystalline structure, to a two-dimensional liquid-like state. We show that this transition is characterized by dynamic heterogeneity and intermittency, as well as cooperative particle movements, that produce an effectively ‘cageless’ crystal. These findings shed light on fluid-coupling driven excitations that are difficult to isolate and control in many other hydrodynamic systems.

Acoustic levitation has been widely used as a contact-free technique to manipulate particles and direct particle self-assembly [10,11]. It has also been used to study the collective properties of many body systems [11,13]. Within the nodal plane of an acoustic cavity, scattered sound waves establish tunable attractions between particles [14], aggregating them together into a close-packed monolayer raft. Sub-millimeter granular particles are too large for thermal noise to drive their dynamics. However, other athermal fluctuations can be introduced by slightly detuning the cavity excitation away from its resonant frequency, which results in vertical oscillations [10,15,16] or spontaneous rotation of the entire raft [11].

Such fluctuations are a manifestation of hydrodynamic instabilities (HIs), where the configuration of particles moving relative to a surrounding fluid evolves unpredictably due to hydrodynamic coupling. HI-induced excitations in many-body systems can exhibit complex correlations that control collective properties, in contrast to uncorrelated thermal fluctuations. In the above-mentioned prior work, the fluctuations emerged from feedback between a moving object and the acoustic cavity mode, an instability found already for a single levitated particle [15,16]. A more subtle instability can arise from the hydrodynamic coupling among multiple particles. For example, such HIs can occur among neighboring particles settling in a fluid [17,18], and it has been associated with the chaotic trajectories of small numbers of weakly interacting steel spheres in an oscillating glycerol bath [19]. Acoustic levitation has proven to be a powerful technique to observe multiple forms of HIs in systems of hundreds of particles evolving over long time scales. However, strongly dissipative frictional particle contacts in prior studies [10,11] masked the presence of HIs arising from the relative motion of particles.

Here, we overcome this limitation by levitating particles small enough that a viscosity-driven repulsive streaming flow can have an appreciable effect on the structure of the particle assemblies. When the levitated particles are comparable in size to the extent of the streaming flows, the repulsive hydrodynamic forces can counteract attractive scattering forces at close range. By tuning the particle size, and by extension, the relative strengths of attraction and repulsion, we are able to vary the stable distance between pairs of levitated particles from contact to a particle diameter apart (Fig 1b-f). Underdamped and contactless, the particles can be now be driven by excitations arising from their hydrodynamic coupling.

We find that the sound energy density of the cavity determines the magnitude of HI-driven excitations, controlling the transition from a quiescent crystal-like state to a highly diffusive liquid-like state. As a result of fluid-induced spatio-temporal correlations, this transition is mediated by intermittent, avalanche-like dynamics where a single displacement event can trigger system wide rearrangements, a clear distinction from the spatially homogeneous thermal melting transition [20,21]. Fundamentally, the avalanche-like excitation arises as a consequence of the correlated motion between neighboring particles starting from the ballistic time scale, which produces an effectively ‘cageless’ crystal over long time scales. In contrast to similar phenomenon in kinetically trapped systems [22], avalanche-like events in levitated rafts do not relax by the descent of a rough configurational landscape and can continue indefinitely.

Our experimental setup is illustrated in Fig 1a. A resonating acoustic cavity is driven on one side by an ultrasonic transducer, which is bolted to an aluminum horn to maximize power output. The resonant frequency...
FIG. 1. Acoustically levitated particles with tunable interactions. a, Schematic of the experimental setup. The transducer consists of an aluminum resonating horn (bottom) and a piezoelectric driving element (top). Ultrasonic standing waves (of wavelength $\lambda$) form between the transducer and reflector. Particles are levitated at the pressure nodal plane. A high-speed video camera is used to obtain the bottom view. b-g, Images of levitated particle clusters (top) and zoomed-in (bottom) views of interparticle spacing. Average particle diameter and frequency are $D = 180\mu m$, $f = 34.7$ kHz ($\lambda = 9.55$ mm), $D = 40\mu m$, $f = 64.3$ kHz ($\lambda = 5.23$ mm), and $D = 40\mu m$, $f = 34.7$ kHz ($\lambda = 9.55$ mm). Scale bars correspond to 100 $\mu m$.

$f_0$ of the coupled transducer and horn is 34,870Hz, with an associated free space wavelength $\lambda = c/f_0 \simeq 9.8 mm$, where $c$ is the speed of sound in air (Supplementary Section 1). Here, we use polyethylene particles of diameter $D = 30 - 60\mu m$ (Supplementary Section 2 and 3), such that the setup operates deep in the Rayleigh scattering regime ($\lambda > D$). Unlike larger levitated objects [10, 15, 16], these particles compose a negligible volume fraction of the resonating cavity. As a result, changing the driving frequency or amplitude adjusts the energy density of the acoustic cavity without triggering spontaneous vertical oscillations of individual particles. Confined by single-scattering forces (i.e. primary forces) [23, 24], particles levitate at the pressure nodal plane, and are imaged from below using a high-speed camera (Fig 1b-f).

Tunable attractive and repulsive interactions. At low energy density in the cavity, particles levitate in stationary configurations and do not fluctuate. The equilibrium distances between particles are determined by the competition of attractive and repulsive interparticle forces. Attractive forces arise from the scattering of sound between particles. These long-range secondary radiation forces are anisotropic, with attraction for particle configurations in the nodal plane and repulsion out of the plane $\parallel$ (Fig. 2a). The magnitude of in-plane attraction as a function of distance scales as

$$F_{\text{rad}}(r) \propto -\frac{E_0 D^6}{r^4 \lambda^3},$$  \hspace{1cm} (1)$$

where $E_0 \equiv \rho_0 V_0^2/2$ is the energy density of the acoustic field in air with maximum velocity $V_0$, density $\rho_0$, wavelength $\lambda$, and $r$ is the center-to-center distance between particles of diameter $D$ [14].

Interparticle repulsion arises from a viscosity ($\nu$) effect, and is not captured by the above scattering analysis. Near particle surfaces a viscous boundary layer of characteristic thickness $\delta_v = \sqrt{\nu/\pi f}$ forms, over which the velocity of air relative to the particle surface decays to zero. Within the boundary layer, acoustic energy is coupled to steady, short-ranged flows. For particle sizes much larger than $\delta_v$, the influence of this boundary layer is negligible; however, for $D \approx \delta_v$, the time-averaged, non-linear microstreaming flows that develop within the viscous boundary layer surrounding the particle become significant. These flows form two toroidal vortices around the sphere, aligned with the axis of the exciting wave $\parallel$. Flow in the levitation plane near the particle surface is outward, opposing attraction from the radiation force (Fig. 2b). The magnitude of this microstreaming-induced repulsion depends primarily on $\delta_v$, which is similar for particles of all sizes at the same driving frequency $f$. Therefore, as particle size decreases, the relative magnitude of the viscous correction to the short-ranged interaction grows. As viscous forces are nearly constant, while $F_{\text{rad}}$ scales with $D^6$ (Eq. 1), interparticle separation increases with smaller particle size.

To analyze this quantitatively, we calculate interparticle forces using the Lattice Boltzmann method (LBM), a discretized direct fluid simulation technique (see Meth-
are opposite to that in a. The direction of flow-induced forces in the levitation plane represents the particle, while repulsive in the polar direction. The white circle represents the particle in the x-z plane. The interaction is attractive in the equatorial direction.

The competition between scattering and viscous forces produces stable in-plane equilibrium separations which are controlled by the Stokes number \( \Omega = 2\pi f R^2 / \nu \). Figure 2 compares the numerical results for the equilibrium, in-plane center-to-center particle distance \( r_{eq} \) from Fabre et al. [27] with our LBM simulations and experimental observations, showing close agreement. Importantly, we find no significant difference in \( r_{eq} \) when comparing isolated particle pairs with pairs of neighboring particles inside large rafts (Supplementary Section 4). These results are consistent with prior investigations of centimeter-scale spherical particles in a confined, oscillating viscous liquid [19, 28–30].

**Order-to-disorder transition.** In the following we focus on levitated rafts with \( \Omega \approx 6 \), and induce stochastic particle dynamics by increasing the applied oscillation amplitude, parameterized here by the acoustic pressure in the cavity, \( P_{acous} \). The intensity of acoustic driving controls the transition from a quiescent crystal-like state (Fig. 3a, i) with well-defined equilibrium interparticle spacings, to a liquid-like state (Fig. 3a, ii) with rapidly fluctuating particle positions and associated kinetic energy (Fig. 3b and Supplementary Video 1). We note that all motion in this liquid state occurs within the levitation plane and does not involve excursions into the vertical direction (Supplementary Video 2). The particles’ velocity distribution is isotropic and well-described as a traditional granular gas (Supplementary Section 5). The interaction of the raft with the acoustic field in the cavity produces a reduced local energy density at the edge of the raft, thus producing distinctive rings of particles that rarely leave their respective annuli (Supplementary Section 6).

To quantify how disorder emerges in the crystal-like state as \( P_{acous} \) is increased, we calculate the spatially and long-time averaged bond order parameter \( \Phi_b = \langle \sum_{n_{nn}} \sin(\theta_i) \rangle / N_{nn} \), where \( \theta_i \) is the angle between the bond and an arbitrary axis, \( n_{nn} \) denotes nearest neighbor, \( N_{nn} \) is the number of neighbor particles, and the average is over all particles \( N \). We find that \( \Phi_b \) monotonically decreases with increasing \( P_{acous} \) (Fig. 3c). At first glance this is surprising, given that the strength of interparticle, in-plane attractive forces increases with energy density in the cavity and thus \( P_{acous} \) (see Eq. 1 and Fig. 2c). Stronger interparticle binding promotes crystalline order; however, we find that the acoustic energy density increases the strength of HI-driven perturbations more rapidly, allowing us to observe an order-to-disorder transition.

FIG. 2. Short-ranged repulsive interactions from viscous effects. a. The acoustic scattering potential, \( U_{ac} \), in the x-z plane. The interaction is attractive in the equatorial while repulsive in the polar direction. The white circle represents the particle. b. Time-averaged microstreaming flow of magnitude \( v_{ms} \) around a sphere, as obtained by Lane [25]. The direction of flow-induced forces in the levitation plane are opposite to that in a. c. The magnitude of force in the equatorial direction \( F_r \) as a function of radial distance \( r \) for various particle diameter, as calculated with a lattice fluid simulation. The black curve represents the force from the scattering expansion following [14]. d. The magnitude of the microstreaming repulsive interaction, approximated by subtracting the scattering expansion from the lattice fluid force traces in c. The purple dashed line represents an exponential fit, \( a \cdot \exp(-(r - D)/\kappa) \); \( \kappa \approx 14 \mu m \). e. The equilibrium distance \( r_{eq}/D \) between a particle pair as a function of Stokes number \( \Omega \). Blue error bars represent the uncertainties for selected data points (Supplementary Section 2).
We observe that this order-to-disorder transition is characterized by sudden, avalanche-like rearrangements (Fig. 3a). To quantify these excursions, we calculate a spatially averaged, local order correlation function, \( h_6(t, \Delta t) = \langle \Phi_6(t - \Delta t) \cdot \Phi_6(t) \rangle_N \), which measures how much local 6-fold orientational order \( \Phi_6 \) changed over a short time interval \( \Delta t \) before time \( t \), averaged over \( N \) particles. \( h_6 = 1 \) indicates no change in ordering, while \( h_6 \) close to zero corresponds to the decorrelation of local bond order. A characteristic decorrelation time \( \Delta t = 10 \text{ms} \) was chosen for the liquid state, but the behavior of \( h_6 \) shown here is not sensitive to choices of \( \Delta t \) from 5 to 100ms.

Figure 3b-d shows the evolution of \( h_6 \) for three representative cases. Rafts exhibit intermittent dynamics (orange), where \( h_6(t) \) switches between low and high values at random intervals, indicating that the raft behaves as a crystal-like state with stochastic bursts of liquid-like rearrangements. The quiescent, crystal-like state has a wide range of lifetime, from fractions of a second to a few seconds, but will eventually be interrupted by a liquid-like excitation. In the crystal-like state the particle displacements are smaller than a lattice spacing, and account for all decorrelations \( h_6 > 0.9 \) (red shaded band). For example, the dip in \( h_6 \) for the red trace around 0.3s is due to phonon motion in an otherwise stationary crystal. Finally, at large \( P_{\text{acous}} \), the raft enters a liquid-like state, with \( h_6 < 0.5 \). Defining the probability \( P_{\text{cryg}} \) of finding the raft in the crystal-like state as the fraction of time that \( h_6 > 0.9 \), we observe a sharp drop around \( P_{\text{acous}} \approx 860 \text{Pa} = 1.1 P_{\text{acous}}^{\text{min}} \) (purple dashed line in Fig. 3b-d,e). \( P_{\text{acous}}^{\text{min}} = 780 \text{Pa} \) is the minimum pressure at which rafts can be stably levitated, signaling the transition from the stochastically interrupted crystal-like state, to a consistently liquid-like state.

**Intermittent dynamics from cooperative motion.** We now look in more detail at the structural rearrangements that are produced by HIs inside a raft. Intermittent excursions into the liquid-like state can be identified by pulses in the kinetic energy per particle (Fig. 3a inset), where each pulse is an intense, avalanche-like excitation. Between such events, the system settles into an idle, highly ordered crystal state (Supplementary Video 3). These excited and quiescent states exist for similar amounts of time and typically each last for a fraction of a second. At the onset of an event (Fig. 3a inset), the kinetic energy increases smoothly at about the same rate as it decays by viscous damping of individual particle motions in air. This similarity in the rates of energy increase and decrease implies that the energy is injected locally via viscous streaming flows, on a per particle basis, rather than through a sudden external, boundary-coupled perturbation (e.g., via an air current).

To better understand these intermittent excursions, we next examine the per-particle dynamics of a representative event. We define a reference time \( t_0 \), immediately before the onset of the event for comparison with later times, \( t \). In Fig. 3b, arrows indicate particle displacements from \( t_0 \) to \( t \). Particles positions at time \( t \) are shown and colored according to their kinetic energy. An event typically starts with chain-like rearrangements at the raft edge, the locations of which are correlated with defects that delineate the interface between the raft’s core and its outer layers, as shown in Fig. 3a, i–ii (Supplementary section 7) [31]. Particle motion creates more defects which propagate through the entire extent of the raft (Supplementary section 8), inducing an avalanche-like burst of collective rearrangement that persists until the kinetic energy is dissipated and the system attains some stable configuration. We note that there is significant spatial heterogeneity in the particle rearrangements during an event, with the most mobile particles tending to collectively move in a linear path through the raft, similar to what has been observed in glassy supercooled liquids [32, 34].

A crucial distinction between our levitated rafts and...
supercooled liquids is in the pre- and post-event configurations. Glassy systems drive avalanche events by descending a rough free-energy landscape, a process that starts with large displacements localized near ‘hot spots’, but eventually arrests as the ground state is approached. In contrast, levitated rafts undergo intermittent avalanche-like events indefinitely, and crystal-like configurations before and after events are not distinctly different (Fig. 4a, i–ii and Supplementary section 9). Furthermore, avalanche-like events in levitated rafts are not localized and eventually involve the entire system.

In order to illuminate the nonequilibrium nature of these hydrodynamically driven raft rearrangements, we compare our experimental results with predictions based on a thermal ensemble of particles. To this end, we plot the mean square displacement (MSD) for both the experiment and a molecular dynamics simulation (Supplementary section 10) with pair-wise interactions (Fig. 4c). To properly compare the data, we non-dimensionalize distances by the lattice spacing $L$, and times by the time taken for a particle to ballistically travel a distance of one lattice spacing $t^* = L/\langle v \rangle$. For particles in a thermal bath and with $k_B T/V_B = 0.04$ (the case most analogous to our experiments, see Supplementary section 10...
for details), where $V_B$ is the binding energy, the MSD traces exhibit ballistic behavior for $t << t^*$, caging for $t \approx t^*$, and diffusive motion for $t >> t^*$ [35]. In contrast, the experimental MSD does not exhibit any caging behavior. Instead, it transitions directly from ballistic to diffusive behavior around $t \approx 0.1t^*$. This behavior is reminiscent to that of a liquid or gas, without lattice ordering to induce intermediate-time caging. Notably, although the experiment has an effective temperature $\langle KE_{x,y} \rangle / V_B = 8 \times 10^{-4}$, nearly 50 times lower than the simulation counterpart, the experimental particles are more mobile due to the presence of intermittent avalanches.

To further examine this exotic behavior, we define the velocity-direction correlation parameter

$$\gamma(t) = \sum_{ij} (\hat{U}_i(t) \cdot \hat{U}_j(t)) w_{ij}(t),$$

(2)

where $\hat{U}_i$ and $\hat{U}_j$ are the unit vectors of displacement for neighboring particles $i$ and $j$, and $\hat{r}_{ij}$ is the displacement vector between particles. In addition, we define a weight density $w_{ij}(t) = (\hat{U}_i(t) \cdot \hat{r}_{ij}(t))^2$, indicated by the shading in the schematic of Fig. 4d, such that directions parallel to the velocity of the particle are weighted more than those perpendicular. Our results are plotted in Fig. 4d, and show that the levitated particles display highly correlated motions even at short times. In contrast, the motions of particles in the simulation are uncorrelated, and only begin to correlate after the ballistic regime $t > 0.1t^*$. Fundamentally, this contrast highlights the difference between excitation induced by HIs and by thermal fluctuation. Excitations from HIs display a high degree of spatio-temporal correlation and depend on the configuration of nearby particles.

We show that this spatio-temporal correlation results in the lack of particle caging by measuring the number of particles that escape their caging neighborhood over time. Here, particles are considered to have escaped their cage if they traversed at least one lattice spacing relative to $t_0$. Our results are plotted in Fig. 4e, and show that the levitated particles begin to uncage at $t \approx t^*$, which is the time particles will take to cross a distance $L$ if their motion is ballistic. In contrast, the simulated system only starts to uncage for $t \approx 10t^*$, which is when the particle motion becomes diffusive. This distinction shows how the correlated motions make the levitated crystal susceptible to perturbations, as a single uncaging event is amplified into a system-spanning event by the tendency of interparticle HIs to correlate particle motions.

Taken together, our results establish acoustically-levitated rafts as a potent platform for investigating how hydrodynamic coupling can spontaneously excite a many-body system. By tuning the relative strength of attractive scattering interactions and repulsive microstreaming flows, particle configurations exhibit hydrodynamic instabilities that scale in magnitude with applied acoustic pressure more quickly than any stabilizing acoustic binding. As a consequence, increasing acoustic pressure drives rafts through an order-to-disorder transition. Unlike the melting of a two-dimensional crystal in thermal equilibrium, this order-to-disorder transition occurs through intermittent periods of liquid-like behavior that eventually merge into a two-dimensional fluid-like steady state. These intermittent, spatially correlated events reveal that hydrodynamic-instability-driven excitations are not equivalent to their thermal counterparts, in line with recent results for active systems that demonstrate the central role of statistical decorrelation between driven and fluctuating degrees of freedom in order to recover thermal-like behavior [36]. Levitated rafts may also serve as a highly suitable system to explore experimentally the link between spatially-varying dissipation and dynamic transitions, as seen in simulations of active matter [37, 38]. Finally, our results provide valuable insight into the limits of acoustic many-body control, elucidating a key instability mechanism that affects the precision of acoustic manipulation at small scales.
EXPERIMENTAL SETUP AND METHOD

Experiment

Our acoustic cavity is driven on one side by a commercial ultrasonic transducer. An aluminum horn is bolted onto the transducer to amplify the acoustic power generation. The bottom of the metallic horn has a concave radius of curvature \( R = 50 \text{mm} \) and was painted black to minimize light reflection. The concave geometry weakly confines the particles at the center of the trap. A commercial heat tape, regulated with a PID temperature controller, is wrapped around the horn and maintains the transducer at \( 35 \pm 0.5^\circ \text{C} \) for experimental repeatability. The transducer is driven by applying a sinusoidal wave of peak-to-peak voltage \( V_{pp} (60-200 \text{V}) \) and frequency close to the resonance frequency of the horn, \( f_0 = 34.85 \text{kHz} \); this signal is generated from a function generator and amplified by a high-voltage amplifier (A-301 HV amplifier, AA Lab Systems). The reflector consists of an acrylic plate (bottom) and an ITO-coated glass slide (top). To reduce the effect of tribocharging, both the horn and reflector were grounded. The distance between the edge of the horn and the reflector can be adjusted with a translation stage to \( \lambda/2 = 4.9 \text{mm} \). In order to minimize the effect of air currents, the entire setup was enclosed within an acrylic box with dimensions much larger than the levitation region \((l \times w \times h = 61 \times 30 \times 46 \times \text{cm}^3)\).

We used polyethylene spherical particles (Cospheric, material density \( \rho = 1000 \text{kgm}^{-3} \), diameter \( d=30-60 \mu\text{m} \), see Supplementary Section 2 and 3 for particle size distribution). The particles are stored, and all experiments are performed within a humidity and temperature-controlled laboratory environment (45-50 % relative humidity, 22-24°C).

Before and after each experiment, the setup was cleaned with de-ionized water and dried with compressed air. For each experiment, the particles were added to the cavity with a tweezer. Videos are recorded with a high-speed camera (Vision Research Phantom T1340) at 3000 frames per second. The acoustic pressure generated is measured with Xarion optical microphone (Eta100 Ultra).

Lattice Boltzmann Simulation

The Lattice Boltzmann method (LBM) was used to perform simulations of particles interacting through acoustically-mediated forces. This method captures the full extent of the fluid-structure interactions, naturally including the effects of viscous dissipation and momentum transfer due to multiple scattering events. LBM simulations of the acoustic cavity were performed within the waLBerla framework [40]. Particles were simulated in periodic domains to avoid spurious boundary reflections. A spatially-homogeneous background force field was used to oscillate the background fluid. The PE functionality of the waLBerla framework was used to simulate the interaction of particles with the acoustic field [41]. Hydrodynamic forces between the particles and fluid were handled with the partially-saturated cells method [42]. A local cell size of \( D/15 \), where \( D \) is the particle diameter, was sufficiently high lattice resolution for accurate force calculations.
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Author contributions
B.W., B.V.S., and H.M.J. designed the study. M.X.L. designed the original experimental apparatus. B.W. modified the apparatus and performed the experiments. B.W. performed molecular dynamics simulations and analyzed the data. B.V.S. performed the lattice Boltzmann simulations and analyzed the data. All authors interpreted the results and contributed to writing the manuscript.

Competing interests
The authors declare no competing interests.