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## Diffusive contact between randomly driven colloidal suspensions

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We study the relaxation process of two driven colloidal suspensions in diffusive contact to a steady state, similar to thermalization. We start by studying a single suspension, subjecting it to random driving forces via holographic optical tweezers, which agitate it to a higher effective temperature. Interestingly, the effective temperature of the suspension, defined by the Einstein relation, exhibits a non-monotonic dependence on the driving frequency. Next, we follow the flux of particles between two such suspensions in diffusive contact, starting from a uniform density and relaxing to a state with zero net particle flux. The density remains uniform for systems with different frequencies but equal effective temperatures. At high driving frequencies, we show that the density distribution at steady state is determined by equating the ratio of the chemical potential to the effective temperature in both systems, mirroring thermal equilibrium behavior.

A dilute suspension of spherical active or randomly driven colloidal particles provides a unique platform for investigating non-equilibrium statistical physics. Although clearly in a non-equilibrium state, this system often exhibits well-defined state functions. First, the osmotic pressure, denoted as II, coincides with the thermodynamic pressure when defined mechanically, under the condition that interactions between colloidal particles and between them and the system's boundaries are torque-free [1–4]. Second, an effective temperature, denoted as  $T_{\rm eff}$ , is well-defined through the generalized fluctuation-dissipation relation (GFDR) when active collisions occur at shorter timescales than particles' thermal relaxation to the potential landscape's minima. [5–8].

Notably, these suspensions adhere to an ideal-gas-like equation of state similar to the equation of state observed in dilute thermal colloidal suspensions  $\Pi = nk_BT$ ,[9–11], with  $T \to T_{\text{eff}}$ , where n is the particle number density and  $k_B$  the Boltzmann constant [1, 12, 13]. Experiments [13] and simulations [1, 14] have explored deviations from this equation of state as colloidal density increases. These timescales could arise from factors like the reorganization of the suspension or memory effects from complex collision dynamics [15].

The concept of effective temperature, a macroscopic variable governing heat flow independent of microscopic details, has proven valuable in non-equilibrium thermodynamics [16, 17]. Despite its widespread use, a crucial gap exists in the literature regarding the quantification of the effective temperature's sensitivity to microscopic details. This information is essential for defining its applicability and limitations. Furthermore, identifying state functions that predict equilibration upon contact between driven or active systems would offer significant insights into the underlying non-equilibrium thermodynamics governing these materials.

Here, we address both topics, creating systems of driven



FIG. 1. Illustration of the HOTs setup and the experiment protocol. Laser light is imprinted by a phase pattern by an SLM. The light is then relayed to the back aperture of an inverted microscope and focused on the sample plane, where it forms a random beam array. The phase mask is changed at a constant switching frequency, thus repeatedly creating new random beam arrays. The transparent beams in the figure represent one array, whereas the bolder beams represent the following beam array. The inset shows a top view of the suspension split into fictitious squares. A uniform beam distribution is achieved by assigning one beam per square.

colloidal suspensions, allowing us to change the microscopic dynamics of the particles in a controlled manner. To this aim, we apply random optical forces to the colloidal particles and measure the resulting suspension's effective temperature. We find that we can obtain the same effective temperature with two different driving protocols inducing different microscopic dynamics. When well defined, the effective temperature governs the thermodynamics of the driven suspensions independently of the underlying microscopic dynamics. Next, using the idealgas-like description of colloidal suspensions, we show that the equilibration between driven colloidal suspension in diffusive contact is reached when the ratio of the chemical potential and the effective temperature is equal on both sides, in accord with equilibrium thermodynamics.

We used holographic optical tweezers (HOTs) [18–20] to generate multiple randomly placed optical beams (Fig. 1) and switch their position at constant time intervals  $\tau_s = 1/f$ . Our HOTs setup, previously described in detail [21], is based on a Keopsys fiber laser (wavelength, 1083 nm). We control the position of the beams by imprinting a phase-only computer-generated hologram on the laser beam with a spatial light modulator (LCOS-SLM, X10468, Hamamatsu) capable of reliably switching its pattern with a frequency  $f \leq 20$  Hz. We couple the laser pattern on the SLM to the back aperture of a 100x oil immersion objective (NA=1.42) mounted on an Olympus IX71 inverted microscope. The light pattern is then formed in the sample plane.

Our samples consist of silica particles,  $1.50 \pm 0.08 \ \mu m$  in diameter (Polysciences,  $n_p \approx 1.42$ ), immersed in a 90% DMSO 10% water ( $n_m \approx 1.46$ ) mixture. The higher refractive index of the fluid ensures that particles are repelled from the laser beams. A quasi-2D suspension of colloidal particles is prepared, approximately 40  $\mu$ m high, by placing 8  $\mu$ L of the colloidal suspension between a slide and a cover-slip, both passivated with a 10 w/w% Bovine serum albumin (Sigma Aldrich) solution.

We drive the colloidal suspension, mimicking thermal forces in the following manner. An array of 36 randomly positioned beams is projected onto a region of interest (ROI) in the sample plane. To ensure homogeneous driving, we divide the ROI into a fictitious square array with a lattice parameter of  $5.0 \pm 0.1 \ \mu m$ . Each laser beam is assigned to one square in the lattice, i.e., each beam location is generated randomly within its respective square (Fig 1). The random distance between the laser beams and colloidal particles diffusing in the suspension ensures that particles are subjected to forces of varying strengths and directions. However, unlike thermal white noise, these tailored fluctuating forces change at constant time intervals.

The resulting motion of the colloidal particles is a type of Run-and-Tumble motion, where the direction of motion switches completely between runs with no memory or preference for directions. During the runs, the particle is pushed away from the laser beam towards the minimum of the potential landscape. The motion of a single particle of position x(t) under the influence of a laser beam located at y(t) can be described using a modified Langevin equation similar to that of a self-propelling particle [3],



FIG. 2. Extracting the effective temperature,  $T_D$ , from particle trajectories. a) Probability distribution of particle displacement, at f = 1 Hz, with lag time  $\tau = 1/30, 1/10, 1$  s (symbols) fitted to a Gaussian distribution (solid line). Inset: a typical trajectory of a particle at f = 1 Hz. b)  $\langle \Delta r^2 \rangle$  as a function of  $\tau$ , for experimental measurements of f = 0.1, 1, 10, 20 Hz. c) Experimental measurements (filled diamonds) and simulation results (empty squares) of the diffusion coefficient, friction coefficient, and effective temperature normalized by their value in the absence of driving as a function of switching frequency f.

$$\frac{d\vec{x}(t)}{dt} = \frac{F_{\text{opt}}(t)}{\gamma} + \sqrt{2D}\vec{\eta}(t),$$

$$\vec{F}_{\text{opt}} = \frac{A}{\sigma^2}(\vec{x} - \vec{y})e^{\frac{-(\vec{x} - \vec{y})^2}{2\sigma^2}},$$
(1)

where we have neglected inter-particle interactions. Here,  $\vec{F}_{\rm opt}$  is the stochastic Gaussian-shaped optical force driving the particle, with beam amplitude A and width  $\sigma$ . The beam's random position y changes at a frequency f, D is the diffusion constant, and  $\eta$  is a Gaussian white noise of unit variance.

To characterize the relation between the driving frequency and the resulting effective temperature of the colloidal suspension, we perform a set of experiments in which we change the driving frequency in the range of 0.1 Hz  $\leq f \leq 20$  Hz. We record 10 movies of 5.5 min at 30 fps using a CMOS camera (GS3-U3-2356M, FLIR) for each driving frequency from which we extract particle trajectories using standard particle tracking algorithms [22], see, for example, a typical trajectory at a driving frequency of f = 1 Hz (Fig. 2a, inset) [23]. The position probability distribution function (PDF) and mean square displacement  $\langle \Delta r^2 \rangle$  of the suspended particles are then calculated (Fig. 2a,b). Since the PDF is Gaussian at  $\tau > 1$  s and  $\langle \Delta r^2 \rangle$  increases linearly with lag time,  $\tau$ , we treat the long-time particle motion as normal diffusion [24]. The diffusion coefficient is then given by  $D = \langle \Delta r^2 \rangle / 4\tau$ .

In Fig. 2c (top pannel), we plot  $D/D_0$  as a function of the driving frequency, where  $D_0$  is the diffusion coefficient at room temperature with no driving. Interestingly, D is non-monotonic in f, reaching a maximum value at  $f \sim 2$  Hz. This behavior can be understood by looking at the two limiting cases. At the limit of very slow driving, the projected pattern barely changes while particles diffuse freely, avoiding the laser beams. Thus, we expect  $D/D_0 \simeq 1$ . In contrast, at very fast driving, the light pattern changes faster than the relaxation of the particles to the new potential landscape. Therefore, the particles experience a time-averaged uniform potential in which they diffuse freely, resulting in  $D/D_0 \simeq 1$ . We expect D to be maximal when the relaxation time of the particles to the new minima of the potential landscape,  $\tau_r$ , is equivalent to the switching time  $\tau_s$  of the pattern. In this regime, the particle's response to the optical forces results in maximal motion, leading to higher diffusivity levels and a higher effective temperature.

The friction coefficient of the suspension in the presence of driving is measured from a second set of experiments, in which we translate the suspension slowly through the light pattern and measure the average particle velocity [25]. The friction coefficient is given by  $\gamma = \gamma_0 v_0/v$ , where  $\gamma_0$  and  $v_0$  represent the friction coefficient and average velocity of the suspended particles without external driving. Figure 2c (middle pannel) shows the weak dependence of  $\gamma$  on the driving frequency.

Following previous studies [3, 5, 6, 12, 26], we use the Einstein relation to define the effective temperature of the suspension,  $k_B T_D = \gamma D$ . The effective temperature shown in Fig. 2c (bottom panel), exhibits a non-monotonic dependence on frequency. This behavior mirrors the trend observed in the diffusion coefficient, as the friction coefficient remains relatively constant across the measured frequency range.

We complement our experiments with Brownian dynamics simulation to extend the range of frequencies and driving strengths examined. In our simulations, particles are subjected to thermal and optical forces implemented according to Eq. 1. Particles interact via hard core repulsion implemented by the WCA potential [27]. In the first set of simulations, we use  $A = 35k_BT$ , T = 293 K,  $\gamma_0 = 6.178 \cdot 10^{-8}$  kg/s, and  $\sigma = 0.64 \ \mu m$  similar to the experimental conditions, and ran 10 simulations of 10 min duration to extract D and  $\gamma$  as a function of f (Fig. 2c). We observe only a qualitative agreement between simulations and experiments, mainly due to the lack of hydrodynamic interactions. However, simulations show a maximum effective temperature at a similar switching



FIG. 3. Driving characteristics extracted form simulated data with  $A = 35k_BT$ . A typical optical force acting on a single particle as a function of time for a) f = 0.1 Hz, and b) f = 50 Hz. c) The persistence particle motion time  $\tau_p$  as a function of the switching frequency. d) The average force acting on the particle as a function of the switching frequency.

frequency as experiments (Fig. 2c) [28].

We conducted further simulations in which we significantly enhanced the repulsion force of the laser beams (6 fold) and increased the driving frequency beyond experimentally attainable values (up to 200 Hz). This extended the range of examined conditions and allowed us to enhance the effect of optical forces in relation to thermal motion [7, 8].

From the simulated data, we can readily calculate the force acting on each colloidal particle as a function of time, as depicted for f = 0.1 Hz and f = 50 Hz in Fig. 3a,b, respectively. The initial amplitude of the force is determined by the random distance between the closest beam to each particle. The persistence time of particle motion is given by  $\tau_p = \min(\tau_s, \tau_r)$ . As the driving frequency increases,  $\tau_p$  transitions from  $\tau_r$ , 0.72 s in our simulations, to  $\tau_s$  (Fig. 3c). The average force amplitude acting on the particles increases with frequency and saturates towards a finite value (Fig. 3d [29]). For Runand-Tumble particles  $k_B(T_D - T) = v^2 \gamma / 2\alpha$  where v is the run speed and  $\alpha$  is the tumble rate for  $t \gg \alpha^{-1}$ [3]. By substituting  $v = F_{\rm av}\gamma$  and  $\alpha^{-1} = \tau_p$  we obtain  $k_B(T_D - T) \sim F_{av}^2 \tau_p / \gamma$ . The coupled effect of the non-linear change in mean force amplitude and persistent time further justifies the non-monotonic change in the effective temperature observed in our experiments and simulations.

In equilibrium, the transport of particles between two systems in diffusive contact is determined by the temperature and chemical potential [30]. The conventional assumption that thermalization is achieved by maximiz-



FIG. 4. Experimental measurement of particle flow between two driven systems in diffusive contact as illustrated on the right panel. The probability of finding a particle on the side of the higher effective temperature is plotted as a function of time for three typical cases: (1) same driving frequency and same effective temperature (orange), (2) different driving frequency but same effective temperature (purple), and (3) different driving frequency and different effective temperatures (green). Net particle flow is observed only when the effective temperature is different on the two sides. Here,  $f_1 = 1$  Hz,  $f_2 = 2$  Hz,  $f_3 = 0.1$  Hz resulting in  $T_1/T = T_2/T = 1.7 \pm 0.2$ and  $T_3/T = 1 \pm 0.2$ .

ing entropy with respect to energy and particle flow leads to two requirements,  $T_1 = T_2$  and  $\mu_1/T_1 = \mu_2/T_2$ , respectively. The chemical potential of dilute colloidal suspensions is given by [31],  $\mu = -k_BT \ln [(\rho_0/\rho)(T/T_0)^{3/2}]$ , where  $\rho$  is the number density of colloidal particles and  $\rho_0, T_0$  refer to some reference state. Following the generalization of pressure in terms of the effective temperature, we generalize the chemical potential of our randomly driven suspensions by substituting  $T \to T_D$ .

With this analogy in mind, our objective is to investigate the particle flow between two regions with different effective temperatures over time. To achieve this, we divide our ROI into two subsystems, each driven independently with a different switching frequency, as depicted in the right panel of Fig. 4. All experiments start with a uniform distribution of particles. We measure the probability of observing a particle in the system at the higher effective temperature  $P_{\rm hot}$  as a function of time. In our control experiment, we drive both sides of the system independently but with the same frequency  $f_1 = 1$  Hz, resulting in zero average current (orange line in Fig 4). Next, we chose a different frequency  $f_2 = 2$  Hz that drives the suspension to a similar effective temperature  $T_{1Hz}/T \sim T_{2Hz}/T = 1.7 \pm 0.2$ . We use  $f_1$  as the driving frequency on one side of the system and  $f_2$  on the other. While the effective temperature is similar on both sides, particle dynamics are different. Nonetheless, we observe no net particle flow between the two sides (purple line Fig 4). This result indicates that the effective temperature used here may be a state function of the system and is independent of microscopic dynamics. Finally, we drive the two sides of the system with different frequencies,  $f_1 = 1$  Hz and  $f_3 = 0.1$  Hz, resulting



FIG. 5. Comparison of the thermodynamic variables of two driven suspensions in diffusive contact (squares and circles) (a) The chemical potential as a function of the minimal switching frequency  $f_{\min}$ . (b) The effective temperature as a function of  $f_{\min}$ . (c)  $\mu/k_BT$  as a function of  $f_{\min}$ . Simulations were run with  $A = 200k_BT$ .

in different effective temperatures,  $T_1/T = 1.7 \pm 0.2$  and  $T_3/T = 1 \pm 0.2$ . Under these conditions particles flow towards the side with a lower effective temperature (green line Fig. 4 [32]).

Since the effective temperature is kept constant by our driving protocol, and particles can not carry with them effective heat, we expect that the two systems in contact will reach a steady state in which  $\mu/k_BT_D$  is equal on both sides. This condition is held trivially in our first two experiments since the effective temperature  $T_D$  and particle density  $\rho$  are equal from the start. However, for the third experiment, we have  $\mu_1/k_BT_3 \sim -14.7 \pm 0.8$  and  $\mu_3/k_BT_3 \sim -13.3 \pm 1.3$ . These values are equal within experimental measurement, suggesting that in dilute colloidal suspension, the effective temperature defined through the Einstein relation determines the thermodynamic behavior of the suspensions.

Next, we conduct a series of simulations to investigate the spectrum of driving frequencies wherein equilibrium conditions for reaching a steady state apply. Given that the GFDR holds for  $\tau_s < \tau_r$ , we examine our findings through a plot depicting the lower driving frequency,  $f_{\min}$ , of the two interacting systems. Specifically, we analyze both systems' effective chemical potential, temperature, and ratio (Fig. 5). We find that as  $f_{\min}$  increases, the values of  $\mu$  and  $T_D$  change less with the driving frequency. Moreover, while the values of  $\mu/T_D$  of both sides agree within measurement error at steady-state, the agreement is very good when  $f_{\min} > 1/\tau_r$ . Hence, we conclude that within our driving scheme, at sufficiently high frequencies, the driven colloidal suspensions behave as equilibrium colloidal suspensions at elevated temperatures.

Our experiment successfully establishes a system for controlling fluctuations in colloidal particles suspended in a fluid. These randomly driven particles exhibit enhanced normal diffusion at longer timescales. Following prior research on driven and active matter, we employed the Einstein relation to define an effective temperature. Specifically, when the switching time surpasses the average relaxation time of particles within the random driving potential, we ascertain the meaningful thermodynamic implications of this effective temperature. This threshold aligns with the condition previously identified for the temporal fluctuation-dissipation relation's validity in driven and active colloidal suspensions [7, 8], as well as in dry driven and active environments [33, 34].

Our study highlights two key findings in this regime. First, the specific mechanism driving the effective temperature does not influence the relaxation of connected systems toward a steady state. Second, the effective temperature serves a dual purpose: it defines a generalized chemical potential, and together, these two parameters govern the thermalization process in connected systems. In other words, the steady state of the connected systems is reached at maximum entropy, as in thermal equilibrium. Remarkably, this occurs notwithstanding the continuous investment of work and the concomitant entropy production required to sustain a driven, steady state.

In this study, our focus was on driven dilute colloidal suspensions. We anticipate that our effective chemical potential may prove inadequate in describing the equilibration of diffusive contacts in scenarios where the GFDR is violated. For instance, this could occur in denser driven/active colloidal suspensions [1, 13, 15], or in systems exhibiting long-wavelength density fluctuations [35, 36]. Conversely, a wide array of out-ofequilibrium systems, such as the fluctuation of red blood cells [37], driven granular gases [16, 33], collections of bristle robots [38], and active colloidal crystals [39], adhere to the GFDR. This observation suggests that, for these systems as well, the Einstein-based effective temperature may hold thermodynamics significance, for example, in contact where energy and particles are exchanged.

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### SUPPORTING MATERIALS FOR DIFFUSIVE CONTACT BETWEEN RANDOMLY DRIVEN COLLOIDAL SUSPENSIONS

### Typical trajectory shape

Figure 6 shows typical trajectories of particles at different driving frequencies. The particles are driven away from the optical beams toward a local minimum of the potential landscape, where they diffuse, unless a new random beam setting is projected driving them toward a new position. The typical trajectories, therefore, show either directed motion or diffusion. Consequently, at low switching frequencies, particles are confined to more compact areas on average.



FIG. 6. Typical trajectories of particles subjected to random repulsive forces switching pattern at a rate of 0.1 Hz, 0.5 Hz, 5 Hz, and 15 Hz. The particles are 1.5  $\mu$ m silica particles. They are suspended in a mixture of 90% DMSO 10% water ( $n_m \approx 1.46$ ). A HOTs apparatus creates and switches the laser beam pattern.

### The normal diffusion regime



FIG. 7. Characterization of the stochastic motion of the driven colloidal particles. a) Probability distribution of the particle's displacement after the shortest step measured ( $\tau = 1/30$  s) for different driving frequencies (various colors). The arrow indicates the trend with increasing driving frequency. b) The non-Gaussian parameter  $G(\tau)$  as a function of lag time for different driving frequencies (various colors). The arrow indicates the trend with increasing driving frequency. The inset shows the lag time at which  $G(\tau)$  reaches its maximal value,  $\tau_{\text{max}}$ , as a function of the switching frequency.

The diffusion coefficient of our driven particles is well-

defined on timescales much longer than the average duration of the particle's persistent motion. From the particle's displacement's probability distribution at the shortest lag time measured  $\tau = 1/30$  s, we observe a non-Gaussian propagator, especially at high driving frequencies (Fig. 7a). However, when plotting the non-Gaussian parameter  $G(\tau) = \frac{\overline{\langle \Delta x(\tau)^4 \rangle}}{3 \overline{\langle \Delta x(\tau)^2 \rangle}^2} - 1$  [1] we observe that the particle's motion transitions effectively to normal diffusion for lagtimes larger than  $\tau = 1$  s (Fig. 7b).

### Measuring the friction coefficient from simulation and experiments

Measurements of the effective friction coefficient were performed in the following manner. In experiments, the colloidal particle sample was translated at a constant velocity over the beam array while projected and switched (Fig. 8 top panel). Similarly, a particle was subjected to a drift force and random optical forces in simulation. The tracer particle's velocity was measured as a function of its position (Fig. 8lower panel).



FIG. 8. Illustration of the friction coefficient measurement scheme (top panel). A typical trace of tracer velocity as it is driven past the fluctuating beam pattern (lower panel).

# Effect of hydrodynamic interactions on the effective temperature

To examine the effect of hydrodynamic interactions (HI), we performed an additional set of Stocksian dynamic simulations [2, 3] in which the Rotne–Prager operator approximates HI and an unbounded 3D fluid suspension is assumed. We expect HI to enhance the agitation of the suspended colloidal particles since they transmit forces moving one particle to other particles. This is observed in Fig. 9; the diffusion coefficient of the simulated data with HI is higher than that of simulations without HI. Moreover, the experimental data are in between these limits since the actual HI in the experiment is partially suppressed due to the proximity of the particles to the bottom of the sample [4].



FIG. 9. Experimental diffusion coefficients (filled diamonds) are compared with simulation results. The simulations include (empty circles) and exclude (empty squares) hydrodynamic interactions. All data is normalized by the diffusion coefficient and effective temperature without driving and plotted as a function of the switching frequency.

# Relation between potential amplitude and average force



FIG. 10. Relation between the optical potential amplitude and the average force experienced by a particle in simulations;  $F_{\rm avg}$  as a function of switching frequency for different trap amplitudes. Error bars are omitted, since the error, defined as the standard deviation of  $F_{\rm avg}$  between the different particles, is much smaller than the marker size.

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