Ballistic and diffusive dynamics in a two-dimensional ideal gas of macroscopic chaotic Faraday waves

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We have constructed a macroscopic driven system of chaotic Faraday waves whose statistical mechanics, we find, are surprisingly simple, mimicking those of a thermal gas. We use real-time tracking of a single floating probe, energy equipartition, and the Stokes-Einstein relation to define and measure a pseudotemperature and diffusion constant and then self-consistently determine a coefficient of viscous friction for a test particle in this pseudothermal gas. Because of its simplicity, this system can serve as a model for direct experimental investigation of nonequilibrium statistical mechanics, much as the ideal gas epitomizes equilibrium statistical mechanics.

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Classical kinetic theory requires molecular chaos and homogeneity [1,2]. Atomic-scale collisions provide the source of homogeneous random motion in thermal systems and form the foundation of classical thermodynamics. In equilibrium thermal systems the crossover from ballistic motion to diffusive motion is a fundamental link between microscopic statistical mechanics and macroscopic thermodynamics [3]. In Einstein's classic thought experiment of a pollen grain in water a thorough study of the grain's ballistic motion would require simultaneous temporal resolution of $\simeq 10 \ \mu s$ and spatial resolution of $\simeq 1 \ nm$ [4]. Therefore, this crossover from ballistic to diffusive has only recently been experimentally demonstrated in the equilibrium thermal systems of rarefied gases [5,6] and liquids [7,8].

By contrast, macroscopic systems allow studies of their constituent dynamics that are impossible in the thermal world. Macroscopic systems dilate the characteristic length and time scales but lose the stochastic excitations of thermal systems. This means that any of the random motion necessary for mimicking equilibrium thermal behavior macroscopically must be produced by some stochastic energy input. Because chaotic Faraday waves are very well understood and characterized they present an ideal source of randomness. The Faraday instability is excited on the surface of a fluid subject to vertical oscillations beyond some critical amplitude [9–11]. Above a second, higher, critical amplitude the surface waves transition from stable ordered waves to spatiotemporal chaos [12]. Surface flows have been measured using fluorescent dyes [13,14] and tracer particles considerably smaller than the characteristic wave size [13,15–17]. A ballistic to diffusive crossover has been observed in chaotic Faraday waves using virtual tracer data drawn from particle image velocimetry measurements [18]. However, for real tracer measurements to date, diffusive motion and fractional Brownian motion have been observed at long and at relatively short time scales, respectively [17,19]; the ballistic regime for real particles has not been demonstrated.

It remains unproven, therefore, whether a driven (and therefore nonequilibrium) athermal system such as chaotic Faraday waves still exhibits all characteristics of equilibrium statistical mechanics, such as a ballistic-diffusive crossover and a well-defined temperature derived from atomistic chaos, as it is in classical kinetic theory. A variety of attempts to define

pseudotemperatures have been proposed for nonequilibrium systems [20]. These have had limited success, describing aspects of the systems' dynamics only over narrow parameter ranges and for few measured properties. A typical approach is to use the Stokes-Einstein relation [21,22] or fluctuationdissipation theorem [22] to define an effective temperature. While successful in producing a well-defined temperature, these studies do not comment on whether or not their internally determined quantities exhibit behavior consistent with classical kinetic theory. In the present study we achieve random excitation and homogeneity by floating a particle large relative to the characteristic length of the Faraday waves on a chaotic fluid surface (Fig. 1). Despite the decidedly nonequilibrium nature of chaotic Faraday waves, we show that they drive a buoyant tracer to undergo fully ballistic (short times) and diffusive (long times) Brownian motion, a hallmark of isotropic equilibrium statistical mechanics. This system admits of a well-defined temperature, diffusion constant, and drag coefficient consistent with the system being a nearly ideal gas of excitations. We have created a macroscopic pseudothermal system (which, if treated like a conventional thermal system, would be 10^8 times hotter than the center of the sun).

We generate the Faraday waves in a circular aluminum dish brimful with water (Fig. 1). We use a circular dish to discourage ordering of the waves [23]. The inner portion, where the water is held, has a radius of 9 cm and a depth of 1.27 cm. A gutter to collect spillover is carved around this inner region. The dish is vertically agitated by a shaker (Vibration Test Systems VTS-100) whose frequency f and peak-peak amplitude Awe control independently using a digital function generator (Stanford Research Systems Model DS335) and a voltage amplifier (Behringer Europower EP4000). Mounted on the bottom of the dish is an accelerometer (CTC AC244-2D/010) used to measure stroke amplitude and frequency. The dish is filled to brimful conditions [11] against a knife edge to avoid pinning of the contact angle, which would create a "cold" zone near the edges. However, the fluid position remains pinned to the knife edge, leading to some damping of the waves near the edges. Our floating test particle is a three-dimensional printed section of cone (selectively laser sintered nylon, coated in black spray paint, with a density of approximately 0.7 g/cm^3) with a top radius of 0.75 cm, height 0.25 cm, side angle

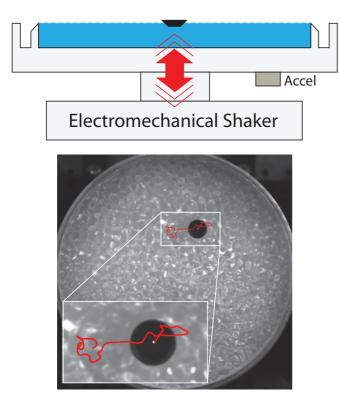


FIG. 1. (Color online) Shown on the left is a cross-sectional diagram (cartoon) of the shaker assembly. The inner portion of the dish is filled with water to brimful conditions. The electromechanical shaker vertically agitates the dish, exciting the Faraday instability on the surface of the water. A buoyant particle is buffeted by the surface waves. An accelerometer (Accel) is attached to the bottom of the dish to directly measure the peak-peak amplitude *A*. Shown on the right is a photo of the experimental apparatus from above. A floating particle is shown, along with a typical position trace for 7.5 s of data collection. The surface of the water is illuminated from an angle to show the chaotic Faraday waves.

45°, and mass 216 mg. The size of the particle is chosen such that it is always larger than the characteristic size of the Faraday waves, which are on the order of millimeters. The particle is colored black and the dish is spray painted white for maximum contrast. The system is imaged from \simeq 1.3 m above the surface of the water with a digital camera (Pointgrey Flea3 FL3-U3-32S2M-CS with a Pentax C32500 KP lens) with a 2080 × 1552 CMOS sensor.

We know *a priori* that certain regions of our parameter space are inaccessible (Fig. 2): Below a critical amplitude line chaotic Faraday waves do not exist; above a sufficiently high amplitude line the surface undergoes a topological transition and begins to splash, which typically sinks the particle. The shape of these critical lines is nontrivially dependent on the properties of the fluid (i.e., viscosity, capillary length, and skin depth), the frequency and amplitude of oscillation, and the container aspect ratio [24]. The details of the transition from ordered to chaotic Faraday waves have been the subject of a great deal of scientific interest [25,26]. However, for this work we are only concerning ourselves with systems in which spatiotemporal chaos is fully developed but the vertical oscillation is not so great that the fluid splashes.

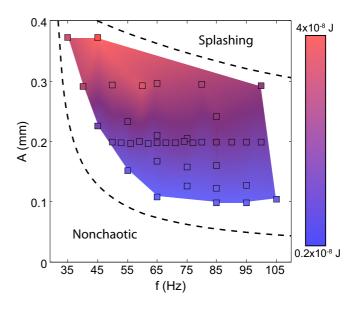


FIG. 2. (Color online) Map of the parameter space. The parameter space is formed by the two externally controlled parameters: shaker frequency f and peak-peak amplitude A. This map shows approximate loci between the nonchaotic, accessible, and splashing regimes. Plotted within the accessible region are points in parameter space where measurements were taken, with color indicating pseudotemperature τ . The color gradient goes from blue (dark gray) for cold to red (light gray) for hot. Plotted behind these points is an interpolated contour map of τ .

The floating test particle is set into the dish near the center and tracking is initiated. We track the particle with subpixel accuracy using a radial symmetry algorithm [27]. By restricting our region of interest of the current frame to the area near where the particle was found in the previous frame, we are able to track in real time at 60 frames/s with a signal to noise ratio $\gtrsim 200:1$ and a subpixel real-space resolution of $<10 \ \mu$ m. The particle is tracked for 7.5 s, chosen to allow for maximum data collection while reducing the likelihood that the particle will reach the edge of the dish, where the physics is fundamentally different. The process is repeated for N = 10 or 100 trials.

We measure the mean square displacement $\langle \Delta r^2 \rangle$ and use it to characterize the particle's motion. If motion is ballistic then

$$\langle \Delta r^2 \rangle = \langle v^2 \rangle \Delta t^2, \tag{1}$$

where $\langle v^2 \rangle$ is the mean square ballistic velocity and Δt is the lag time. If motion is diffusive, then

$$\langle \Delta r^2 \rangle = 4D\Delta t,\tag{2}$$

where *D* is the diffusion constant. Figure 3 shows a representative mean square displacement demonstrating ballistic motion $(\langle \Delta r^2 \rangle \sim \Delta t^2)$ for short times and diffusive $(\langle \Delta r^2 \rangle \sim \Delta t)$ for long times. We measure the coefficients $\langle v^2 \rangle$ and *D* by fitting Eqs. (1) and (2) to the data in the ballistic and diffusive regimes, respectively.

The insets of Fig. 3(a) show the scaled distributions of displacement magnitude for various lag times Δt . For short

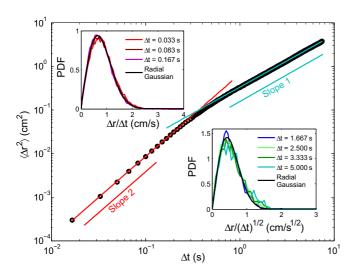


FIG. 3. (Color online) Representative mean square displacement $\langle \Delta r^2 \rangle$ (f = 85 Hz and A = 0.159 mm). Here $\langle \Delta r^2 \rangle$ is plotted against lag time Δt . At short lag times the mean square displacement shows a power law with an exponent of 2, indicating ballistic motion. For longer lag times the curve turns over to a power law with an exponent of 1, indicating diffusive motion. The upper left inset is the distribution of displacement Δr divided by a factor of lag time Δt for lag times in the ballistic regime. The lower right inset is the distribution of Δr divided by $\Delta t^{1/2}$ for lag times in the diffusive regime. Both distributions are shown to collapse onto a 2D Gaussian distribution integrated over all directions (radial Gaussian distribution).

times in the ballistic regime the distributions collapse when scaled by a factor of Δt and for long times in the diffusive regime the distributions collapse when scaled by a factor of $\Delta t^{1/2}$. In both regimes, the distributions are well described by a two-dimensional (2D) Gaussian distribution integrated over all directions uniformly. This random walk behavior is an emergent result of allowing a particle to interact with a chaotic surface.

In thermal systems the quantities $\langle v^2 \rangle$ and *D* are related to temperature *T* and the coefficient of viscous friction ζ . In contrast to studies such as [21,22], we use the average kinetic energy of the tracer as it undergoes ballistic motion to define an effective temperature. By equipartition the temperature is proportional to the average kinetic energy per particle as

$$\frac{1}{2}m\langle v^2\rangle = k_B T \equiv \tau, \qquad (3)$$

where *m* is the mass of a particle (m = 216 mg) and k_B the Boltzmann coefficient. The thermal energy scale k_BT , *D*, and ζ are related by the Stokes-Einstein relation as

$$D\zeta = k_B T \equiv \tau, \tag{4}$$

where *D* and ζ both depend on τ . For our system we regard τ , the energy scale, as a pseudotemperature; τ may be treated with the same physics as a conventional temperature, as opposed to an effective temperature such as those used to describe granular and other athermal systems. The friction term ζ is primarily associated with the viscosity of the chaotic Faraday waves as a 2D medium through which the floating particle is moving.

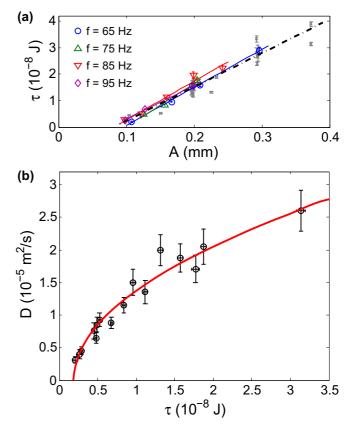


FIG. 4. (Color online) Gas behavior. (a) Pseudotemperature τ plotted against shaker peak-peak amplitude A for various shaker frequencies f, with linear fits shown for frequencies plotted in large color symbols (circles, upward triangles, downward triangles, and diamonds). Gray crosses are the remaining frequencies shown in Fig. 2, ranging from 35 to 105 Hz, and the dashed line is a linear fit to all data points shown. (b) Diffusion constant D plotted against pseudotemperature τ . Data points are taken for data sets where N = 100 trials. Shown in red is a fit to the data of Eq. (7). Error bars shown in (a) and (b) are 1σ confidence intervals resulting from fits used to determine D and τ .

Thus, by measuring $\langle v^2 \rangle$ and D we can determine the intrinsic properties τ and ζ of a 2D pseudothermal system.

Analysis of the data reveals the dependence of $\langle v^2 \rangle$ and D, and therefore τ and ζ , on our externally controlled parameters A and f. Figure 4(a) demonstrates that pseudotemperature τ is proportional to shaker amplitude A, independent of frequency. This trend is also apparent in the pseudotemperature contours shown in Fig. 2. Thus, driving amplitude is the external control for pseudotemperature. Since the typical Faraday wavelength is primarily determined by the driving frequency and τ is independent of frequency, we may also deduce that τ is independent of the ratio of particle size to wavelength over the range of parameters for which we have conducted our study. We find a calibration curve between driving amplitude and the pseudotemperature of our bath:

$$\tau \approx (1.3 \times 10^{-7} \text{ J/mm})A - 1.0 \times 10^{-8} \text{ J.}$$
 (5)

The linearity of this relationship is expected because the height of the chaotic Faraday waves should be proportional to the driving amplitude and the particle moves in response to the height difference between the two largest waves in contact with it. However, there is a curious feature to this fit in that τ crosses zero at finite A. This is due to the fact that below a particular driving amplitude the pseudotemperature is ill defined because the Faraday waves are not chaotic. Thus, A must reach a critical value before τ can exist and begin to increase.

While the existence of ballistic and diffusive motion is strongly suggestive that the system of driven Faraday waves can be considered as a gas at some pseudotemperature τ , we can more rigorously examine whether other general properties of thermal gasses, such as the temperature dependence of their viscosity, manifest themselves in this driven system. Recalling now the Stokes-Einstein relation (4), we examine the behavior of the viscous drag coefficient ζ . Stokes' equation for incompressible fluids states that $\zeta \propto \eta$ where η is the viscosity. Sutherland's formula for the viscosity of a nearly ideal 2D gas (in which particles have finite collision radii and soft long-range interaction potentials) [28] further relates η to τ as

$$\eta = \lambda' \frac{\tau}{\sqrt{\tau + C'}},\tag{6}$$

where λ' and C' are constants of the gas. We can now solve for *D* as a function of τ for an ideal gas to find

$$D = a\sqrt{\tau + b},\tag{7}$$

where all of our constants have been absorbed into *a* and *b*. Figure 4(b) shows our data on τ and *D* to be well fit by this simple ideal gas model with constants $a = 0.16 \pm 0.01 \text{ m/kg}^{1/2}$ and $b = (-1.87 \pm 0.65) \times 10^{-9}$ J. The parameter *a* relates diffusion to energy and the parameter *b* contains information on the interactions within the gas.

As *a* relates diffusion to energy it has units of length/mass^{1/2}. Plugging in the length and mass scale of the particle we would predict from simple dimensional analysis that *a* should be on the order of 0.75 cm/(0.2 g)^{1/2} $\simeq 0.5$ m/kg^{1/2}, which is comparable to the fitted value of *a*.

Since *b* is negative there is a positive value of τ at which the diffusion constant should go to zero and thus the viscosity to infinity. This is a singular point that does not exist for real ideal gases but is difficult to probe directly because the time scales for diffusion diverge as $\tau \rightarrow -b$ and, more practically, it is near the transition from chaos to order in our system.

Above $\tau = -b$ the viscosity and diffusion constant maintain functional behavior consistent with that of a nearly ideal gas, suggesting that the chaotic Faraday waves should be considered a nearly ideal gas of random surface excitations. The sign of the constant *b* tells us the sign of the interaction between the elements of the gas: positive for attractive and negative for repulsive. One would expect that the interactions in our system are repulsive, as gravity will cause waves to repel one another over short distances.

We have demonstrated that chaotic Faraday waves can be described as a pseudothermal, nearly ideal gas of random surface excitations for which a thermal energy scale, diffusion constant, and viscosity are well defined. We directly observe the particle to receive normally distributed isotropic kicks from the chaotic waves. The particle's displacement scales with lag time exactly as would be expected for a Brownian particle and exhibits a clear transition from ballistic to diffusive behavior, showing that we have produced a laboratory-scale Brownian system by Aristotelian means. This pseudothermal gas takes difficult-to-access time and length scales and dilates them to easily studied levels. In so doing we have defined a macroscopic effective temperature in an out-of-equilibrium system functionally identical to, but completely divorced in origin from, a *microscopic* thermal temperature. The present single-particle study outlines the method by which one would define a temperature for a system of chaotic Faraday waves and future studies should examine the dynamics of the multiparticle case.

This experimentally tractable system will be useful for studying the statistical mechanics of actively driven nonequilibrium systems in general, especially interparticle interactions and driven assembly on 2D substrates, which is a subject of active scientific interest [29]. An outstanding theoretical question for 2D systems is whether it is possible to create long range order from short-range interactions [30–32]. Our system offers a fully 2D environment in which these questions can be studied.

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