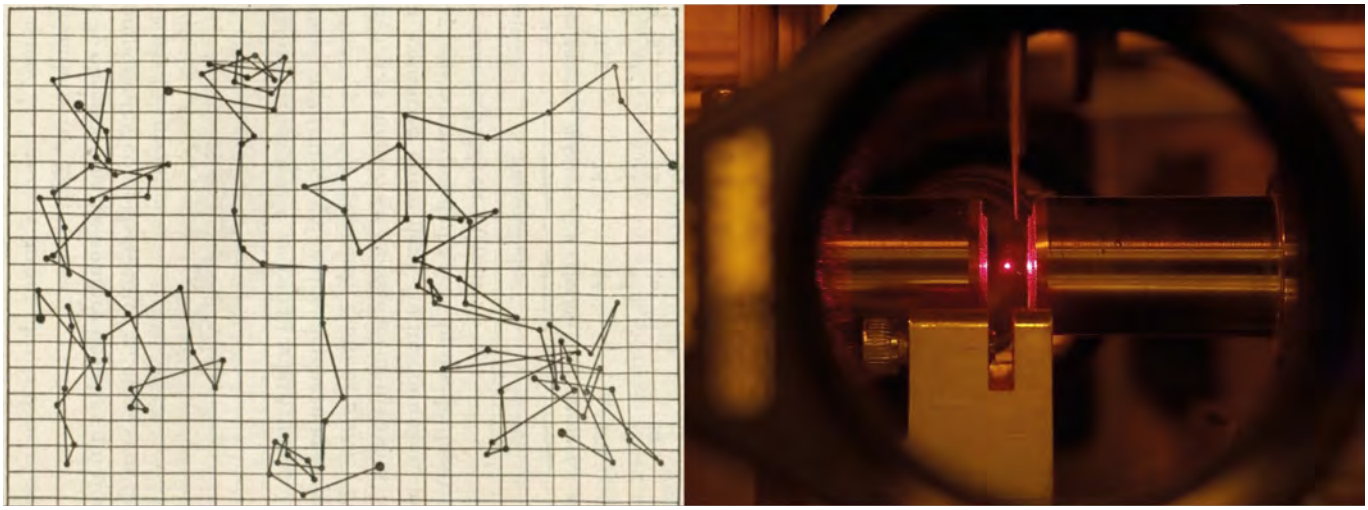

ZOOMING IN ON BROWNIAN MOTION WITH OPTICAL TRAPPING MICROSCOPY

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Optical trapping microscopy unravels open questions in statistical physics by making ultra-precise measurements of Brownian motion in diverse fluid environments. Moreover, the advent of pulsed lasers promises to push the boundaries even further, offering insights on how the compressibility of fluid flow affects Brownian particles at previously unexplored time scales.

Albert Einstein studied diffusion in his PhD thesis with a specific aim to determine then-unknown Avogadro's constant N_A. He derived a value for the translational diffusion coefficient D for a particle of radius a in a fluid of viscosity η

$$D = k_B T/6\pi\eta a, \quad (1)$$

where k_B is Boltzmann constant ($k_B = R/N_A$, where R is the gas constant) and T is the temperature. For his determination of N_A, he used known values of D and η for sugar dissolved in water. Shortly afterwards, with the same aim of determining N_A, he published a paper [1] where he proposed a formula for the mean square displacement $<(\Delta x)^2>$ of one-dimensional translational Brownian motion

$$<(\Delta x)^2> = 2Dt. \quad (2)$$

N_A could be estimated by observing micrometer-sized particles diffuse in water, over an observable distance within a minute. The root-mean-squared displacement of a Brownian particle grows with the square-root of time, a defining feature of diffusive motion. However, basic kinematics demands that at any instant t, the particle has a continuous — though fluctuating — instantaneous velocity $v(t)$ with which it moves from one point to the next.

Einstein realized this and published a paper [2] where he estimated a time θ below which Eq. (2) would fail:

$$\theta = m/(\log_{10} e\ 6\pi\eta a), \quad (3)$$

where m is a mass of the Brownian particle. He obtained Eq. (3) by considering how long it takes for a particle's velocity to decrease to one tenth of its initial value due to Stokes friction. In equilibrium, the root-mean-square velocity of a Brownian particle quantifies its typical velocity scale and is given by the variance of the Maxwell-Boltzmann distribution

$$\langle v^2 \rangle = k_B T/m. \quad (4)$$

Soon after Einstein's work, Jean Baptiste Perrin used Eq.(2) to determine the value of N_A using time-lapse microscopy of tree resin particles with varying sizes [3]. A reproduction of his hand-drawn Brownian motion trajectories are shown in the cover image of this article. Over half a century later, Arthur Ashkin invented optical trapping [4] and developed its application to colloidal particles [5]. In the decades since, accompanying detection techniques [6] evolved greater resolution [7] and enabled direct measurement of coloured thermal noise [8]. This form of optical trapping microscopy finally granted access to times below θ, allowing direct measurement of the Maxwell-Boltzmann distribution of Brownian velocities in gases [9] and in liquids [10], though Einstein famously expressed his view that such a precise measurement would be impossible [2].

A probe for non-equilibrium physics

It is clear from Eq. (3) and (4) that by measuring θ one can estimate particle mass or fluid viscosity while by measuring $\langle v^2 \rangle$ one can estimate particle mass or fluid temperature. Rapid measurement of these properties offers a new probe of non-equilibrium systems where thermodynamic quantities are well defined. To convey the concept, consider a micron-sized polystyrene sphere in air, where θ = 8 μs. Position tracking of the particle has to remain accurate at times shorter than θ in order to directly measure the Maxwell-Boltzmann distribution of velocities, see Figure 1 for a demonstration with experimental data. The duration of measurement (integration time) tmax is determined by the acceptable uncertainty in $\langle v^2 \rangle$. In the data shown in Figure 2, an integration time of 1 ms gives an uncertainty of 30%, *i.e.* two independent 1 ms long measurements of $\langle v^2 \rangle$ differ by 30%. A longer integration time of 10 ms reduces this uncertainty to 10%.

In water, θ is much smaller than in air due to the different viscosities of these fluids. Therefore, changes in a system's properties must be tracked more rapidly in liquids. For example, a micron-sized polystyrene sphere has θ = 0.14 μs in water. By measuring θ, the viscosity of various

> **The Maxwell-Boltzmann distribution is now routinely measured in the lab using optical trapping microscopy**

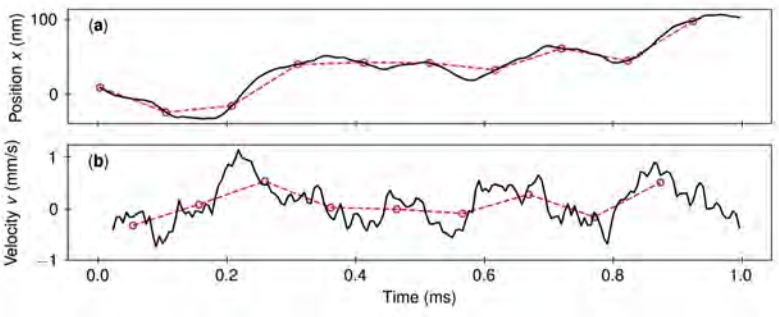
fluids have been measured using only 20 μs of high-resolution data [11]. However, liquids induce hydrodynamic memory effects, so the thermal force is no longer random white noise. It acquires a colour, *i.e.* it is correlated in time and the bare mass m appearing in Eq. (4) is replaced by the added mass $m^* > m$ that includes the inertia of the liquid dragged by the particle.

These time scales of optical trapping microscopy have to be contrasted with measurements in changes in diffusion coefficient D from Eq. (1), a quantity that is defined for times greater than θ for particles in air. Ability to measure θ instead of D thus introduces a finer time scale to explore a physical system of interest.

These preliminary studies might offer a probe for non-equilibrium systems, but their use in actual systems is at an early stage. The ability to weigh a changing mass is important for studies of nucleation. For example, we lack a complete understanding of how ice is formed in clouds. Controlling nucleation processes is crucial for optimized cloud seeding, a technique used to encourage precipitation.

Applications of optical trapping microscopy

High-precision optical trapping microscopy is primarily used to investigate equilibrium systems. For example, a particle in an incompressible bulk fluid or next to a wall with either stick or slip boundary condition was investigated: $\langle v^2 \rangle$ has been measured directly for particles in both gas and liquid, and it has been shown that the color of thermal noise is suppressed for a particle in liquid next to a wall [12]. Thus, this experimental technique can be used as a probe of *e.g.* temperature, viscosity or even surface wettability. In these applications, integration times can be much longer than in the case of non-equilibrium systems and accuracy can be significantly increased.



▼ **FIG. 1:** Time traces of (a) position and (b) velocity for a sphere with a diameter of 3 μm optically trapped in air and recorded at two different time resolutions, ~ θ/20 (black) and ~ θ theta (red). Velocity fluctuations can only be resolved for time resolutions below θ.

Future instrumentation with picosecond resolution is expected to tackle phenomena of compressibility in fluid flows.

In a recent example of such an application a microsphere was optically trapped in air and weighed using Eq. (4) [13]. Mass of 24 picograms was measured with uncertainty of 4%. This application is interesting in the context of ground-state cooling and sensitive force-transduction where accurate knowledge of the trapped particle's mass is required.

In addition to thermal fluctuations, a trapped particle may also be driven by the flow of surrounding fluid. Flow oscillations with up to a few hundred Hz have been measured with video microscopy. High-resolution optical trapping microscopy in air has recently extended this bandwidth to nearly 1 MHz, enabling measurement of flows associated with acoustic radiation [14]. This technique can be compared to other, more established acoustic sensors, such as a microphone.

Figure 3 compares a high-bandwidth microphone to an optically trapped particle when exposed to an impulsive sound generated by laser ablation of an aluminum target. Compared to the microphone, optical trapping microscopy offers a superior measurement bandwidth that is capable of resolving a steeper rising edge and a higher peak pressure. Interestingly, accurate calibration of optical trapping microscopy for high bandwidth acoustic sensing must account for hydrodynamic memory effects, even in air. Even though this example is limited to particles in air, it can easily be extended to liquid environments.

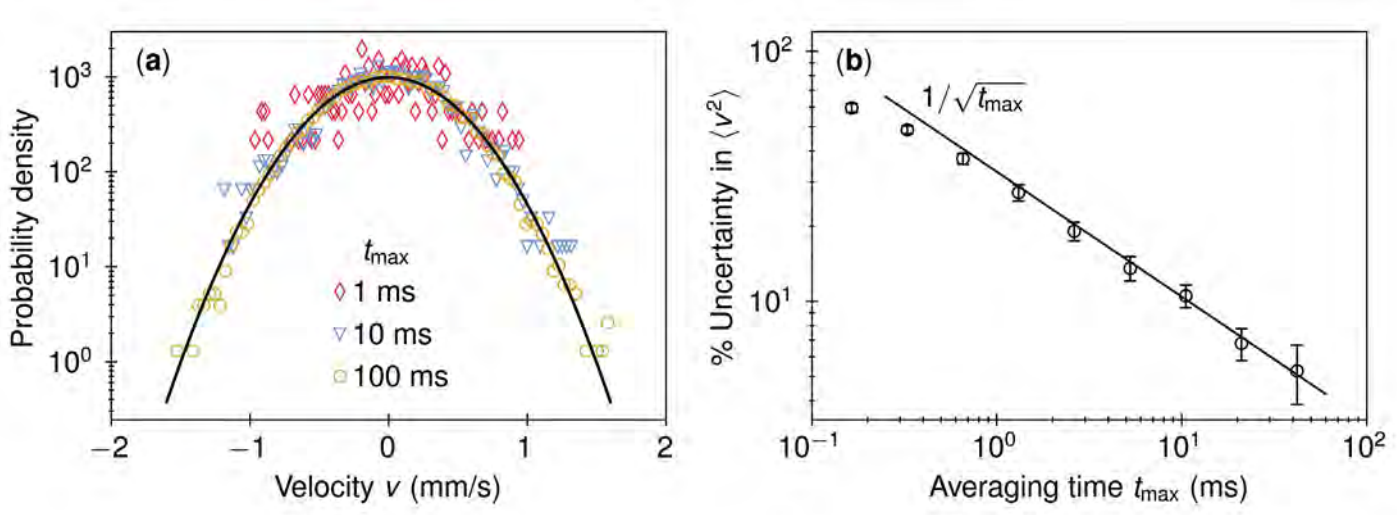
Future instrumentation and experiments

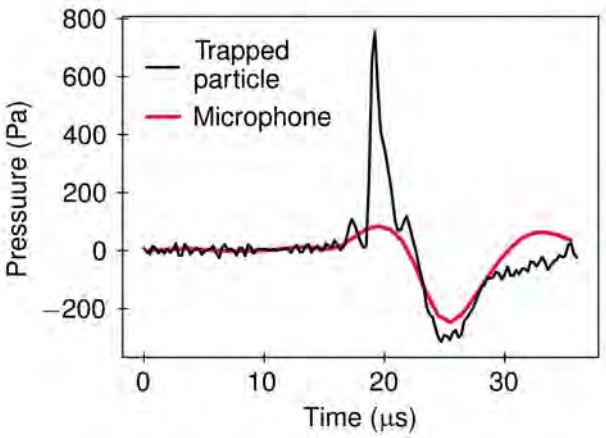
A next step in instrumentation development is the use of pulsed lasers for the position detection [12]. It is expected that the temporal resolution will reach the picosecond regime. Accessing these short time scales will, for the first time, reveal the effects of flow compressibility for a Brownian particle, *e.g.* the onset of viscosity and the bare mass of the Brownian particle in liquid.

Compressibility effects are expected to be especially pronounced in confined fluids, a regular situation in microfluidics. Numerical simulations [15] predict correlations between molecular collisions arising from density fluctuations in confined fluids, unlike those arising from hydrodynamic memory effects in bulk fluids, observed by optical trapping microscopy two decades ago [7]. A convincing experimental confirmation of these correlations in confined fluids is still lacking and there is a hope that optical trapping microscopy might provide it.

In conclusion, we turn our gaze towards the 19th century when the atomic hypothesis found utility in kinetic-molecular theory. Boltzmann proposed that collisions between these discrete microscopic entities are entirely random. This led Maxwell to propose the first distribution for a physical phenomenon. Einstein's and Perrin's work was a culmination of the centennial effort to determine Avogadro's constant, *i.e.* the physical size of these entities. The Maxwell-Boltzmann distribution is now routinely measured in the lab using optical trapping microscopy and it is authors' hope that the text here showed that the simple noise produced by atomic motions is far from being a conceptually closed subject. ■

▾ **FIG. 2: (a)** Maxwell-Boltzmann distribution of velocity traces recorded for different integration times tmax, give a different uncertainty in $\langle v^2 \rangle$ which is inversely proportional with root of tmax. The black line is theoretical expectation of the distribution. **(b)** The uncertainty in $\langle v^2 \rangle$ decreases as tmax-1/2.





▲ **FIG. 3:** Detecting the sound induced by laser ablation of an aluminum target. The trapped particle reacts quicker to an acoustic perturbation than a high-bandwidth microphone and better resolves the peak of a sound wave.

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