Escape and Synchronization of a Brownian Particle

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The escape and synchronization of a 1-μm Brownian particle from an optical trap is presented. The probability distribution of residence times within a given well is exponential with a cutoff at short time. Temporal modulation of the well leads to synchronization.

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Brownian movement is a classic of statistical physics. In his seminal work, Kramers developed a theory on the thermally activated escape from a potential well [1]. In this Letter we present a visualization of the escape of a macroscopic particle and show that the theory is quantitatively verifiable. Rather than make analogy to a particle in a well [2], we consider a fluctuating Brownian particle. Using the technique of optical traps [3,4], we localize the particle in an optical potential well and study its escape. We then enrich the dynamics by temporal modulation of the well and observe the proposed stochastic synchronization [5].

This is a model system for a wide range of phenomena in nature, from neuronal action potentials to muscular motility, and even climate change. The concept invoked is always synchronization of a stochastic escape within a double potential well. As far as we know, this is the first experiment directly analyzing the process for a particle in a double well, subjected to intrinsic thermal activation.

Optical trapping is conceptually simple. A dielectric object in an electric field is polarized. In the presence of an electric field gradient, the polarized particle moves towards the region of highest field. The transverse Gaussian intensity profile across the width of the beam of standard lasers pulls the object towards the beam axis [3]. To counter the destabilizing radiation pressure, one sharply focuses the laser, imposing an electric field gradient along the beam direction [4].

By constructing an optical double potential well using two beams for a Brownian particle, direct observation of the thermally driven escape is possible. The process is visualized for a 1-μm glass sphere in water at room temperature using video microscopy. The probability distribution of residence times, i.e., of the intervals of time between escape events from well to well, is exponential with a cutoff at short times. By evaluating the mean of these distributions ($\sim$2 sec) and extracting an upper limit to the cutoff (≤ 0.1 sec), we quantitatively verify Kramers model and measure interwell energy barriers around $(2-3)kT$.

Synchronization of a stochastic process by periodic external forcing was proposed by Benzi *et al.* [5]. To observe this phenomenon, the double well was built so that one could modulate the depths of the wells. We have observed and measured a synchronization of the escape process by small amplitude modulation.

Let us first describe the experimental aspects. The experiment is built around and through an inverted microscope (Fig. 1). The 488-nm TEM_{00} output of an argon ion laser, expanded by lenses $L1$ ($f=38.2$ mm) and $L2$ ($f=175.0$ mm), enters into the microscope and is strongly focused into the sample by a 100× objective, the diverging part of the beam acting as the optical trap. The power needed is about 10 mW per trap. The sample sits on a translation stage. The charge-coupling-device camera signal is recorded on an S-VHS video recorder. Images were analyzed directly off the monitor.

The optical setup divided and recombined the original beam with two beam splitters ($BS1$ and $BS2$) which allowed independent positional control via the two mirrors ($M2$ and $M3$). We positioned the two laser beams at a distance d from each other and the beam waist at a height s above the bottom coverslip of the experimental cell. When the two beams are brought close together, the transverse potential along the line connecting their centers is reduced, allowing the particle to escape from one trap to the other, defining the one-dimensional axis of the experiment.

To have a barrier height Q sufficiently small, the interwell spacing d must be about 1 μm. Changes in d by as little as 100 nm dramatically changed Q. We chose to add neutral density filter wheels ($FW1$ and $FW2$) to each beam, providing independent control of the intensity.

The various control parameters of the experiment include the geometry of the double well, i.e., the particle radius a, interwell spacing d, and beam waist position s. The laser intensity I sets the barrier height Q and the overall depth of the double well. The particle moves in water with viscosity η, at the ambient temperature T, stable to less than 1 °C.

Samples are prepared by diluting a stock of 1-μm silica spheres down to a number concentration of roughly one sphere per microliter. Cells are built out of a pair of No. 1 coverslips separated by strips of "parafilm" and sealed with epoxy and wax. We used bright-field microscopy with an overall magnification on the TV monitor of 12500.

The video image of intrawell fluctuations and interwell escape events were recorded. Analysis of these escape "events" was done on a Macintosh computer, the video tape being played at reduced speed. The probability distribution $p(\tau)$ of residence times is calculated, normal-

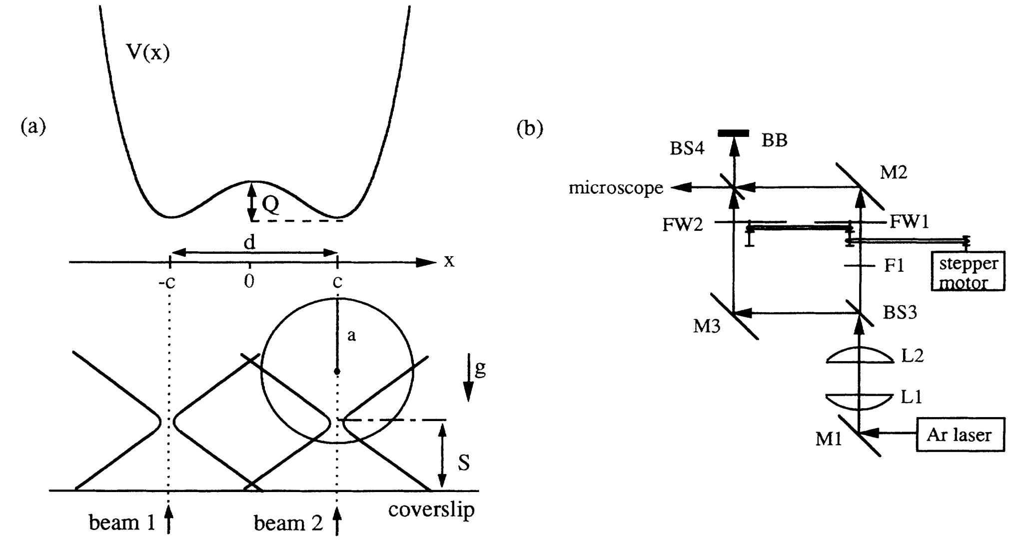

FIG. 1. Experimental setup. (a) Geometry and schematic of the double well. (b) The optical arrangement. Lenses $L1$ and $L2$ expand the beam; beam splitters $BS3$ and $BS4$ define the two beams. BB is an absorber. Neutral density filter wheels $FW1$ and $FW2$ are driven by the stepper motor.

ized by the total number of events N.

We conservatively estimate an experimental temporal resolution of $\Delta t = 120$ msec. To determine the functional form of the probability distribution we used $\frac{1}{2}$-sec bins. The interwell crossing time, below the experimental uncertainty, was ignored (usually two video frames or 67 msec). The first three moments of the distribution are used—the mean value, the standard deviation, and the skewness.

We now turn to the residence time probability distributions. Within his paper, Kramers derived the rate of escape for a Brownian particle in the large viscosity limit [1]. One finds that the mean residence or Kramers time is

$$\bar{\tau}_K \cong (6\pi\eta a/m\omega^2)\exp[Q/kT] = \tau_R \exp[Q/kT] , \quad (1)$$

where a and m are the particle radius and mass, ω the associated quadratic frequency of the potential $U \propto \frac{1}{2} m\omega^2 x^2$, assumed to be the same both at the base of the well and at the top of the barrier, and barrier height Q. The prefactor τ_R is the relaxation time within the well. Since we are observing a homogeneous Poisson process, each escape event is random and of small probability. It is expected then that the residence time distribution should have an exponential density of the form $p(\tau) = \lambda \exp[-\lambda\tau]$.

Figure 2 presents a typical measured probability distribution. This distribution contains $N = 219$ escape events sampled over a 15-min period. The first moment of the distribution defines a mean Kramers time of $\bar{\tau}_K = 1.3$ sec, the standard deviation $\sigma = 1.3$ sec, and the skewness $s = 2.0$. An exponential distribution $p(\tau) = \lambda\exp(-\lambda\tau)$ has a mean equal to the standard deviation $\lambda^{-1} = \bar{\tau} = \sigma$ and a skewness of 2. Thus, the statistical description of Fig. 2 fits the description of an exponential distribution. We then fitted the values of each $\frac{1}{2}$-sec bin (inset of Fig. 2) with the associated statistical uncertainties, using a weighted exponential of the form $p(\tau) = A\exp(-\lambda\tau)$, yielding $\lambda = 0.76 \pm 0.05$; thus $\bar{\tau} = 1.3 \pm 0.1$ sec, consistent with the data itself.

Figure 2 also shows a cutoff in the distribution at short times, reflecting the relaxation time τ_R of the particle within the well. From this cutoff, one can estimate the relaxation time $\tau_R \approx 0.1 \pm 0.1$ sec, a large uncertainty. From the estimated τ_R and the measured mean $\bar{\tau}_K$ one

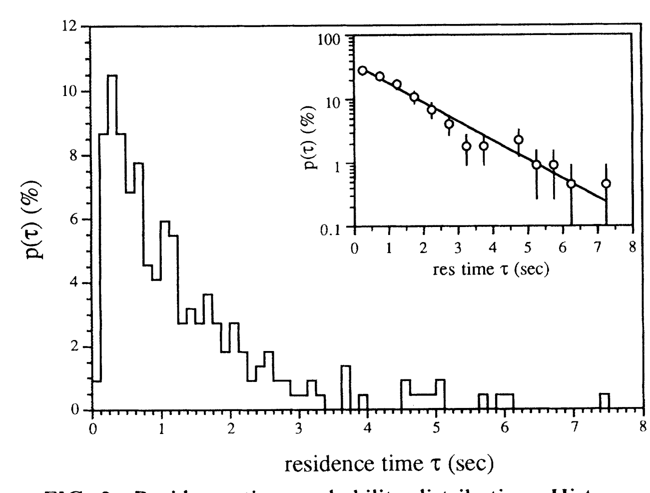

FIG. 2. Residence time probability distribution. Histogram of the distribution showing the exponential decay and the cutoff at short time. $N = 219$ events, $\bar{\tau} = 1.3$ sec, $\sigma = 1.3$ sec, skew $= 2.0$. Bin width $\frac{1}{8}$ sec. Inset: Logarithmic plot showing weighted exponential fit to the data. $\lambda_{\rm fit} = 0.76 \pm 0.05$. Bin width $= \frac{1}{2}$ sec.

deduces a barrier height according to Eq. (1), $Q/kT = 2.6 \pm 1.0$. The uncertainty comes mainly from the estimate of the relaxation time.

As a check, one can estimate an upper bound of the barrier height by extending the parabolic shape to the crossover point between the two wells using the harmonic oscillator energy. For separation $d = 1.0$ μm and particle radius $a = 1.0$ μm, one finds $Q/kT = 5.8 \pm 6.1$ consistent with the previous values.

Further, one can independently estimate the relaxation time from different measurements of the well depth [6] using the harmonic oscillator energy and the definition of the relaxation time. This leads to $\tau_R = 90$ msec, quite close to the cutoff estimate from the distribution.

Let us now discuss synchronization, an idea first proposed by Benzi *et al.* [5]. A double well potential within the Langevin formalism is used. The model, in the overdamped limit where the inertial term has been dropped, is

$$\dot{x} = ax - bx^3 + A\cos(\Omega t + \theta) + \xi(t)\,, \tag{2}$$

where a and b are quartic potential parameters, A is the modulation amplitude, Ω is the modulation frequency, θ is the phase, and $\xi(t)$ represents the fluctuations. The only analytic treatment is in the adiabatic limit where the period of modulation is long relative to the mean escape time [7,8]. Starting from Benzi *et al.*'s early publications, a large set of theoretical work [9–12] some with numerical simulations [13,14] or analog simulations [15–17] as well as experiments [18,19] describe various developments of the so-called "stochastic resonance" idea.

In the experiment, we have observed two characteristic times—the mean Kramers time $\bar{\tau}_K$ and a cutoff or relaxation time τ_R. To synchronize this escape process, we add a third time, a modulation period τ (in effect $\tau/2$ is the relevant time). There are then three regimes depending on whether the dimensionless time $\tilde{\tau} = \tau/\bar{\tau}_K$ is greater than, equal to, or less than 1. Experimentally, to periodically modulate the depths of the potential wells one varies the intensity of each laser beam, since well depth is proportional to intensity. By oscillating the neutral density filter wheels ($FW1$ and $FW2$ in Fig. 1) over a small angle $\Delta\theta$, one varies the intensity of each beam in phase opposition. Modulation amplitude A and period τ are set by two inputs to the stepper motor controller.

The so-called adiabatic limit corresponds to a long modulation period relative to the mean Kramers time $\bar{\tau}_K \leq \tau$. We observe in this regime synchronization about the modulation period τ, and some of the "natural" distribution at short times, as shown in Fig. 3(a). This is physically understandable since the wells are modulated slowly enough for the particle to have a reasonable probability to escape at short time.

In contrast, when the modulation period is such that $\tau_R \leq \tau \leq \bar{\tau}_K$, one observes a structure of odd harmonics in the probability distribution and the disappearance of the natural distribution at very short time [Fig. 3(b)]. The physics of the harmonic structure is fairly evident. As the modulation is too short, the particle might not escape every τ and must wait extra periods 2τ, 4τ, or $2n\tau$, with a weaker probability to be in the initial well each time.

When the modulation period is equal to the mean Kramers time $\bar{\tau}_K = \tau$, a maximal synchronization occurs, meaning no observable harmonic structure and distribution at short time (Fig. 4). It is important to mention here that there is no evidence for a resonance phenomenon. The line shape is independent of modulation period, changing only with amplitude. There is merely a particular condition, $\tilde{\tau} = 1$, at which both the harmonic structure and the natural distribution at short time are strongly reduced. For $\tilde{\tau} \gg 1$, the short time distribution reappears

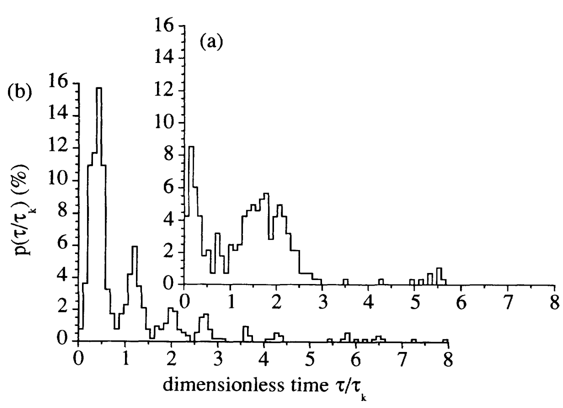

FIG. 3. Probability distributions with a modulation period different from the mean Kramers time. Histogram of residence times for (a) $\tilde{\tau} = \tau/\bar{\tau}_K = 1.54$ showing synchronization and a distribution at short times ($N = 282$ events) and (b) $\tilde{\tau} = 0.38$ showing a structure of odd harmonics ($N = 224$ events). Bin width $= \frac{1}{8}$ sec. $A = \pm 7\%$.

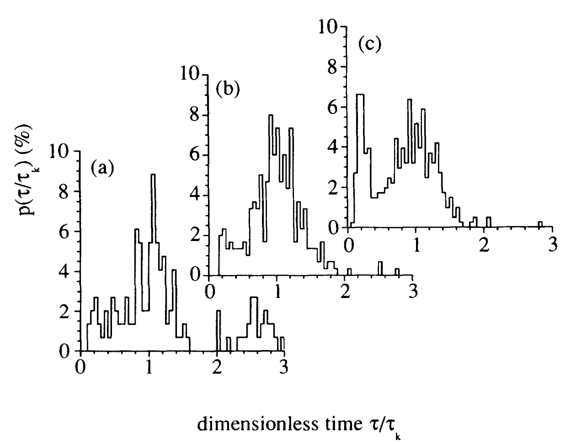

FIG. 4. Probability distributions with a modulation period near the mean Kramers time. (a) $\tilde{\tau} = 0.89$ ($N = 147$ events), (b) $\tilde{\tau} = 1.00$ ($N = 300$ events), and (c) $\tilde{\tau} = 1.11$ ($N = 408$ events). Bin width $= \frac{1}{8}$ sec. $A = \pm 7\%$.

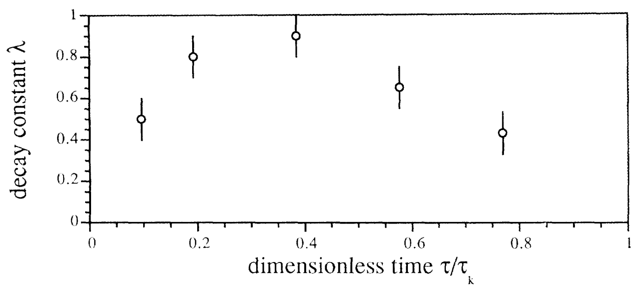

FIG. 5. Exponential decay constant of odd harmonic peaks vs dimensionless modulation time. $A = \pm 7\%$.

and for $\bar{\tau} \ll 1$ one samples it.

An odd harmonic structure was first seen in analog simulations [15]. A theory to describe the exponential decay of the harmonic peaks was worked out in the limit where the amplitude A goes to zero while the amplitude to noise intensity ratio A/D goes to infinity [17]. As our potential is of the order of $3kT$, the results of this theory do not apply. We nonetheless observe an exponential decay. Figure 5 shows the behavior of the decay constant λ.

Let us note a surprising result of the experiment. Beyond the exponential decay of the harmonics, one sees a resurgence of them. For example, in Fig. 3(a), the eleventh and twelfth harmonics can clearly be seen, which seems to appear when $m\bar{\tau}_K = n\tau$.

In conclusion, we have observed the Brownian escape of a 1-μm particle in water at room temperature from an optical potential well and its synchronization. Further, the present systems will allow one to study the interplay between noise and temperature, an area not well understood in most dynamical systems experiments.

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