

P8 – Quantitative Analysis of Fluctuations and Irreversibility of Optically Trapped Microspheres

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We present an experimental verification of several universal theorems of stochastic thermodynamics by means of controllable optical traps, so-called “optical tweezers”. The theoretical and experimental investigation of this novel branch of modern thermodynamics promises in various ways to be of great benefit for the quantitative understanding and future application of processes at the micro- and nanometre scale in the fields of physics, biology, chemistry and applied technology. Utilizing the formal framework established by *U. Seifert* and others around 2005 and following the experimental approaches of *G. M. Wang et. al* first published in 2002, we experimentally demonstrate the validity of several pertinent fluctuation theorems for special non-equilibrium states of optically trapped colloids. Our results in this context exceed the scope of *Wang et al.* 2002-2005 by a considerable margin. Fluctuation theorems which came into the focus of theoretical research about 15 years ago describe the emergence and quantitative evolution of macroscopic irreversibility from the microscopic point of view of stochastic thermodynamics. Moreover, we present a new method for the experimental determination of the radius and the temperature of a single optically trapped colloid by directly analyzing the thermal equilibriums fluctuations of the bead. This technique may be utilized for the optimisation of several other optical tweezers experiments which show a high requirement for quantitative precision.



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Fluctuations and irreversibility

Stochastic thermodynamics verified by optically trapped colloids

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Abstract

We experimentally verify several so-called *fluctuation theorems* related to the novel and promising field of *stochastic thermodynamics* by means of optical tweezers. Generally speaking, those fluctuation theorems describe the emergence and quantitative evolution of macroscopic irreversibility in thermodynamics from a microscopic point of view. In this way, the corresponding, usually distinct levels of microscopic and macroscopic description of irreversible processes are linked and merged. As a result, the macroscopic state function entropy proves to be the ensemble average of a fluctuating microscopic quantity which can be measured on a single colloid level. Surprisingly, the virtually impossible macroscopic case that the ice cubes in a warm drink cool down further to heat up the surrounding liquid, turns out to be probable in the microscopic world. Utilizing the formal framework established by U. Seifert [2, 3] and others and following the experimental approaches of G. M. Wang et al. [4-7], our results exceed the pertinent scope of Wang et al. 2002-2005 by a considerable margin.



Several dissipation theorems

Using the definition of $Q(t)$ from eq. (1), several so-called fluctuation theorems [1-10] can be deduced for this quantity.

Generally, the universal integral fluctuation theorem (UIFT) holds regardless of the particular initial and final system states:

$$\langle e^{-Q(t)} \rangle = 1 \quad \forall t \geq 0. \quad (3)$$

Applying the Jensen inequality, the UIFT implies

$$\langle Q(t) \rangle \geq 0$$

which represents a generalization of the macroscopic second law of thermodynamics, i.e. $\Delta S_{tot} = \langle \Delta S_{tot} \rangle \geq 0$.

Moreover, for an equilibrium or non-equilibrium steady state initial system state, one can show that the detailed transient fluctuation theorem (TFT)

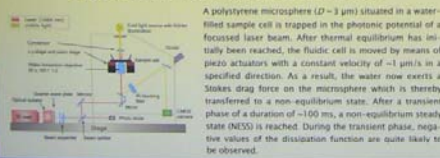
$$\frac{\Pr(Q = -A)}{\Pr(Q = +A)} = e^{-A} \quad (4)$$

holds. Its integral version, the integrated transient fluctuation theorem (ITFT)

$$\frac{\Pr(Q(t) < 0)}{\Pr(Q(t) \geq 0)} = \langle e^{-Q(t)} \rangle_{Q(t) \geq 0} \quad (5)$$

hold:

Experimental setup and non-equilibrium system state



A polystyrene microsphere ($D = 3 \mu\text{m}$) situated in a water-filled sample cell is trapped in the photonic potential of a focussed laser beam. After thermal equilibrium has initially been reached, the fluidic cell is moved by means of piezo-actuators with a constant velocity of $-1 \mu\text{m/s}$ in a specified direction. As a result, the water now exerts a Stokes drag force on the microsphere which is thereby transferred to a non-equilibrium state. After a transient phase of a duration of ~ 100 ms, a non-equilibrium steady state (NESS) is reached. During the transient phase, negative values of the dissipation function are quite likely to be observed.

Measuring broken detailed balance and irreversibility

We observe one time-dependent coordinate $x(t)$ of a microsphere which changes due to thermal noise (i.e. Brownian motion) and an external driving protocol. The argument Q of all fluctuation theorems considered on this poster is a logarithmic measure of broken detailed balance under non-equilibrium conditions. This quantity, in literature usually referred to as *dissipation function*, is reasonably defined as

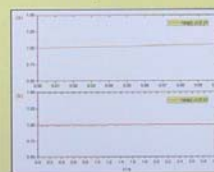
$$Q(t) = \ln \frac{p_1(x(t))Pr(x(t) | x(0))}{p_2(x(0))Pr(x(t) | x(0))} \quad (1)$$

where $Pr(x(t) | x(0))$ denotes the probability of the forward path whereas $Pr(x(t) | x(0))$ denotes the corresponding probability of the backward path. The probability distributions of the completely arbitrary initial and final states are given by $p_1(x(0))$ and $p_2(x(0)) = p_1(x(t))$. Thus, $Q(t)$ quantifies the degree of irreversibility that is associated with the regarded non-equilibrium trajectory.

In order to experimentally measure the dissipation function, it has to be expressed in terms of directly accessible quantities. For instance, one can show that the dissipation function in a certain frame of reference is equal to the total work done on the particle over Boltzmann constant times temperature, i.e.

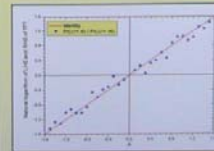
$$Q^{tot}(t) = \frac{1}{k_B T} W^{tot}(t) = \frac{1}{k_B T} \int_0^t \dot{x}(t') F^{tot}(t') dt' \quad (2)$$

Experimental confirmation



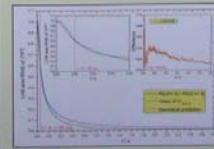
$$\langle e^{-Q(t)} \rangle = 1 \quad \forall t \geq 0$$

Fig. 1: The quantity $\langle e^{-Q(t)} \rangle$ shown in appropriate time domains for two dissipation functions deduced for deterministic (top) and stochastic (bottom) dynamics. For larger time scales, rare events play an increasingly dominant role.



$$\frac{\Pr(Q = -A)}{\Pr(Q = +A)} = e^{-A}$$

Fig. 2: Comparison of the left-hand side (LHS, blue spheres) and the right-hand side (RHS, red line) of the TFT for 6000 single trajectories 100 ms after the initiation of the perturbation.



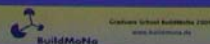
$$\frac{\Pr(Q(t) < 0)}{\Pr(Q(t) \geq 0)} = \langle e^{-Q(t)} \rangle_{Q(t) \geq 0}$$

Fig. 3: Comparison of the LHS and RHS of the ITFT (blue and green lines). Additionally, the theoretical prediction by means of stochastic dynamics is shown (red line). As expected, these three graphs almost collapse. Insets: Magnified detail view of the initial region and LHS-RHS of ITFT.

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