

Direct verification of the fluctuation-dissipation relation in viscously coupled oscillators

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(Dated: July 1, 2017)

The fluctuation-dissipation relation, a central result in non-equilibrium statistical physics, relates equilibrium fluctuations in a system to its linear response to external forces. Here we provide a direct experimental verification of this relation for viscously coupled oscillators, as realized by a pair of optically trapped colloidal particles. A theoretical analysis, in which interactions mediated by slow viscous flow are represented by non-local friction tensors, matches experimental results and reveals a frequency maximum in the amplitude of the mutual response which is a sensitive function of the trap stiffnesses and the friction tensors. This allows for its location and width to be tuned and suggests the utility of the trap setup for accurate two-point microrheology.

The relation between the generalized susceptibility and equilibrium fluctuations of the generalized forces, first obtained for a linear resistive circuit by Nyquist [1] and then proved for any general linear dissipative system by Callen and Welton [2], is a central result in non-equilibrium statistical physics. The relation can be used to infer the intrinsic fluctuations of a system from measurements of its response to external perturbations or, perhaps more startlingly, to predict its response to external perturbations from the character of its intrinsic fluctuations [3]. The fluctuation-dissipation relation is the point of departure for several areas of current research including fluctuation relations [4], relaxation in glasses [5], and response and correlations in active [6] and driven systems [7, 8].

The first experimental verification of the relation between fluctuation and dissipation was due to Johnson [9], whose investigation of the “thermal agitation of electricity in conductors” provided the motivation for Nyquist’s theoretical work [1]. Though the relation has been verified since in systems with conservative couplings, a direct verification in a system where the coupling is entirely dissipative is, to the best of our knowledge, not available. Colloidal particles in a viscous fluid interact through velocity-dependent many-body hydrodynamic forces whose strength, away from boundaries, is inversely proportional to the distance between the particles. The range of these dissipative forces can be made much greater than that of conservative forces such as the DLVO interaction [10, 11]. Therefore, it is possible to engineer a situation where the dominant coupling between colloidal particles is the viscous hydrodynamic force and all other interactions are negligibly small. Such systems, then, are ideal for testing the fluctuation-dissipation relation when couplings are purely dissipative.

In this Letter, we present a direct verification of the fluctuation-dissipation relation for a pair of optically trapped colloidal particles in water. We measure the equilibrium fluctuations of the distance between the par-

ticles and the response of one particle to the sinusoidal motion of another particle. Transforming both correlations and responses to the frequency domain, we verify the fluctuation-dissipation relation over a range of frequencies spanning two orders of magnitude. Remarkably, the response function has a peak in frequency, reminiscent of a resonance, though the system of oscillators is entirely overdamped. A theoretical analysis, assuming slow viscous flow of the ambient water, is in excellent agreement with the experiments. The analysis reveals that the location and width of the resonant peak can be tuned by altering the viscosity, the separation between the particles, the trap stiffnesses, and the colloidal diameters. It provides the inverse relations necessary for using the trap setup for accurate two-point microrheology. We now present details of our experiment and its analysis.

Experiment: The details of the experimental setup towards validation of the fluctuation response theorem are provided in Supplementary Information - here we provide a brief description. Thus, we set up a dual-beam optical tweezers (Fig. 1) by focusing two orthogonally polarized beams of wavelength $\lambda = 1064$ nm generated independently from two diode lasers using a high NA immersion-oil microscope objective (Zeiss PlanApo, 100×1.4). One of the lasers is modulated using an AOM located conjugate to the back-focal plane of the microscope objective, and a long optical path after the AOM ensures that a minimal beam deflection is enough to modulate one of the trapped beams, so that the intensity in the first order remains constant to around 2%. The modulated and unmodulated beams are independently coupled into the trapping microscope using mirrors and a polarizing beam splitter, while detection is performed using a separate laser at 671 nm generating two detection beams also orthogonally polarized and superposed on the respective trapping beams using dichroic beam splitters. The two trapped beads are imaged and their displacements measured by back-focal-plane-interferometry, with the imag-

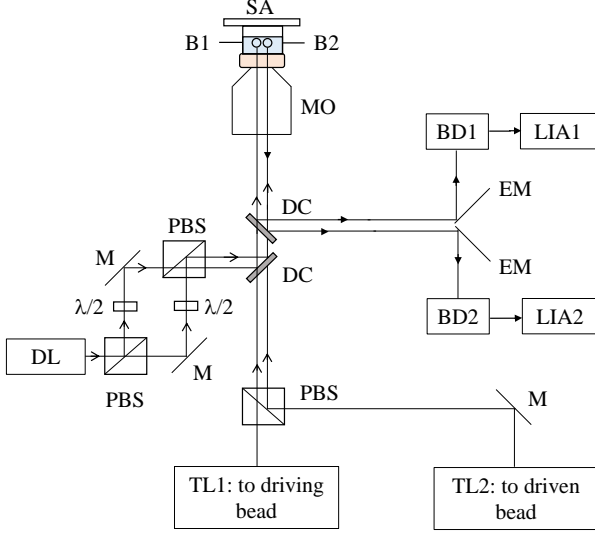


Figure 1. A schematic diagram of the experimental setup. TL1: trapping laser for driving particle B1, TL2: trapping laser for driven particle B2, DL: detection laser, PBS: polarizing beam splitter cube, $\frac{\lambda}{2}$: half wave plate, DC: dichroic mirror, MO: microscope objective, CS: cover slip, BD1 and BD2: balanced detection systems based on Thorlabs photodiodes PD-EC2, M: mirror, EM: edge mirror, LIA1 and LIA2: lock-in amplifiers for B1 and B2, respectively.

ing white light and detection beams also separated at the output by dichroic beam splitters, which along with the orthogonal polarization scheme ensures that cross-talk in the detection beams is absent. A very low volume fraction sample ($\phi \approx 0.01$) is prepared with $3 \mu\text{m}$ diameter polystyrene latex beads in 1 M NaCl-water solution for avoiding surface charges. We trap two spherical polystyrene beads (Sigma LB-30) of mean size $3 \mu\text{m}$ each, in two calibrated optical traps which are separated by a distance $4 \pm 0.1 \mu\text{m}$, so that the surface-surface distance of the trapped beads is $1 \pm 0.2 \mu\text{m}$ ($0.67a$, a being the particle radius) and the distance from the cover slip surface is $30 \mu\text{m}$ ($20a$, so as to overrule wall effects). From the literature [12], the particle separation is still large enough to avoid effects due to optical binding and surface charges. In order to ensure that the trapping and detection beams are not influencing each other, we measure the Brownian motion of a trapped particle when the trapping and detection beams for the other trap is switched on (in the absence of a particle), and check that there are no changes in the measured trap stiffness. One of the traps is sinusoidally modulated (amplitude around $0.2a$) and the phase and amplitude response of both the driving and driven particles with reference to the sinusoidal drive are measured by lock-in detection (Stanford Research, SR830). To get large signal to noise, we use balanced detection systems BD1 and BD2, for the driving and driven particles, respectively. The voltage-amplitude calibration of our detection system reveals that we can

resolve motion of around 5 nm with an SNR of 2.

Each of the optical traps are calibrated using equipartition and power spectrum methods considering the the particle temperature to be same as the room temperature. We verify that each of the potentials is harmonic in nature from the histogram of the Brownian motion which is satisfactorily Gaussian (Fig. 1 in Supplementary Information), even when both trapping beams are on. The sampling frequency is 2 kHz , while we performed data blocking at the level of 100 points in order to ensure good Lorentzian fits [13] for trap calibration. We maintain a considerably higher stiffness for the particle in the modulated trap so that it is not affected by the back-flow due to the driven particle. The low stiffness of the driven trap ensures that it has a maximal response to the drive. Thus, for validation of the fluctuation response theorem, the stiffness of the modulated bead (B1) was $69.6 \mu\text{N/m}$, while that of the driven is $4.8 \mu\text{N/m}$. Note that, to observe a clear amplitude resonance, a lower ratio of trap stiffness is required, as we demonstrate later. The verification of the fluctuation-response theorem is shown in Figs. 2. It is understandable that while the fluctuation-response theorem is in the form a simple equation for a single particle, for two particles the equations would be represented in the form of a matrix, which we discuss in more detail later. This is what we demonstrate in Fig. 2(a), (b), and (c), where the auto and cross-correlations for both particles are matched with the corresponding response functions. The auto-correlation function of B1 is shown in Fig. 2(a), while that of B2 is shown in Fig. 2(b). The corresponding response functions (χ_{11}^{\parallel} , χ_{22}^{\parallel}) are obtained by measuring the amplitude and phase of the individual particles when they are themselves driven. Fig. 2(c) shows the cross-correlation function which is again compared with the corresponding response function χ_{12}^{\parallel} . This is obtained by measuring the amplitude and phase of B2 when B1 is driven. Note that we are not able to measure χ_{21}^{\parallel} which is the response of B1 when B2 is driven since the much larger stiffness of B1 renders the amplitude of the response extremely small so that it is beyond our detection sensitivity. For the response measurements, each data point is the average of ten separate measurements at each frequency. It is clear from the figures that we obtain a good match between fluctuation and response - which essentially validates the fluctuation-response relations for a pair of colloidal particles coupled by hydrodynamic interactions. Note that for consistency check, we also plot the cross-correlation function in the time domain (Fig. 4 in Supplementary Information) and obtain qualitatively similar data as reported in Ref. [14].

Theory: The Langevin equations describing the stochastic trajectories of the colloids are [15]

$$m_i \ddot{\mathbf{v}}_i + \gamma_{ij} \cdot \mathbf{v}_j + \nabla_i U = \boldsymbol{\xi}_i \quad (1)$$

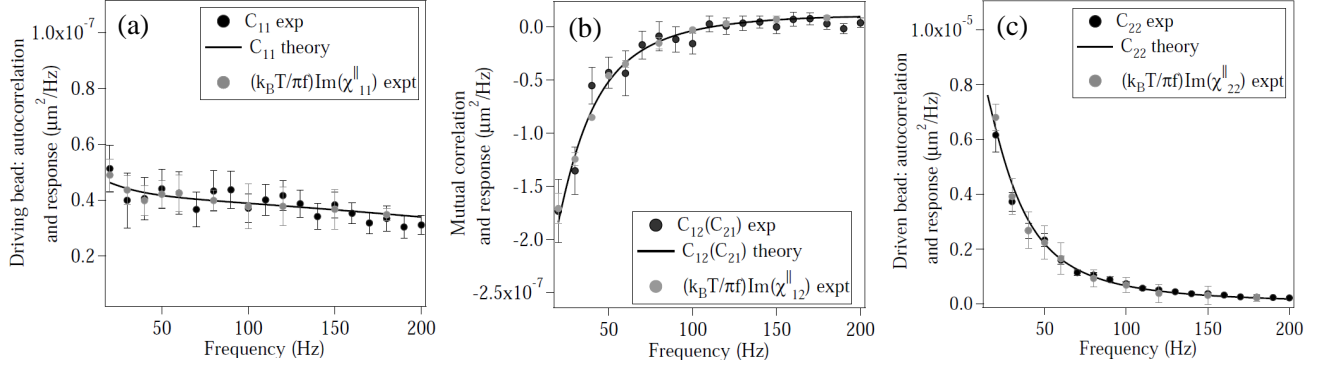


Figure 2. Verification of the fluctuation-response relation $C_{ij} = (k_B T / \pi f) \chi''_{ij}$ for a pair of viscously coupled colloidal particles in optical traps. The first and third panels compare the self-response and the position auto-correlation of, respectively, the driving and driven colloid, while the second panel compares their mutual-response and cross-correlation. Theoretically computed correlation functions, assuming over-damped motion of the colloids and slow viscous flow in the fluid, are shown as solid lines.

where $i, j = 1, 2$ refer to the driving and driven colloid, m_i are their masses, \mathbf{v}_i are their velocities, γ_{ij} are the second-rank friction tensors encoding the velocity-dependent dissipative forces mediated by the fluid, $U = U_1 + U_2$ is the total potential of the conservative forces, and ξ_i , the Langevin noises, are zero-mean Gaussian random variables whose variance is provided by the fluctuation-dissipation relation $\langle \xi_i(t) \xi_j(t') \rangle = 2k_B T \gamma_{ij} \delta(t - t')$. The bold-face notation, with Cartesian indices suppressed, is used for both vectors and tensors.

In the limit of slow viscous flow in the fluid, the friction tensors can be calculated from the Stokes equation using a variety of methods [16–20]. To leading order the result is

$$\gamma_{ij} = \delta_{ij} \mathbf{I} \gamma_i - (1 - \delta_{ij}) \gamma_i \gamma_j \mathcal{F}_i \mathcal{F}_j \mathbf{G}(\mathbf{r}_i, \mathbf{r}_j) \quad (2)$$

where $\gamma_i = 6\pi\eta a_i$ are the self-frictions, \mathbf{G} is a Green’s function of the Stokes equation [21], \mathbf{r}_i are the centers of the colloids and $\mathcal{F}_i = 1 + \frac{a_i^2}{6} \nabla_i^2$ are the Faxén corrections that account for the finite radius, a_i , of the colloids. We emphasize that this expression is not limited to the translationally invariant Green’s function of unbounded flow, $8\pi\eta \mathbf{G}(\mathbf{r}) = (\nabla^2 \mathbf{I} - \nabla \nabla) \mathbf{r}$, but holds generally for any Green’s function and is both symmetric and positive-definite [19, 20]. The mutual friction tensors decay inversely with distance in an unbounded fluid and more rapidly in the proximity of boundaries. The assumption of slow viscous flow is valid at frequencies $\omega \tau_\nu \ll 1$ where $\tau_\nu = \rho L^2 / \eta$ is the vorticity diffusion time scale [22].

The harmonic optical potentials are given by $U_i(t) = \frac{1}{2} k_i |\mathbf{r}_i - \mathbf{r}_i^0|^2$ where \mathbf{r}_i^0 are the centers and k_i are the stiffnesses of the optical traps. Note the absence of conservative mutual couplings. The system remains in equilibrium when the trap centers are stationary but is driven into non-equilibrium when they are modulated in time as $\mathbf{r}_i^0(t)$. For small modulations the response is linear.

For modulation frequencies $\omega \ll \gamma_i / m_i$ the veloci-

ties can be adiabatically eliminated from the inertial Langevin equations to yield inertialess Langevin equations for the positions [23]. The multiplicative noises in the resulting equations have clear interpretations within the adiabatic elimination procedure; there is no Itô-Stratonovich dilemma [15, 24–27]. Both correlation and response functions can be calculated in this limit. Linearizing about the mean separation between the trap centers and decomposing the motion into components parallel and perpendicular to the separation vector, the result for the parallel response function is

$$\text{Im} [\chi_{ij}^{\parallel}(\omega)] = \frac{\omega M_{ij}}{(\det A - \omega^2)^2 + (\omega \text{tr} A)^2} \quad (3)$$

where $A_{ij} = \mu_{ij}^{\parallel} k_j$ is a “response” matrix, the mobility matrix μ_{ij}^{\parallel} is the inverse of the friction matrix and

$$M_{ij} = \begin{pmatrix} \frac{k_2}{k_1} \mu_{22}^{\parallel} \det A + \mu_{11}^{\parallel} \omega^2 & -\mu_{12}^{\parallel} (\det A - \omega^2) \\ -\mu_{21}^{\parallel} (\det A - \omega^2) & \frac{k_1}{k_2} \mu_{11}^{\parallel} \det A + \mu_{22}^{\parallel} \omega^2 \end{pmatrix}.$$

The magnitude of the response of the driven bead to the driving bead is maximum at the “resonance” frequency

$$\omega_{res} = \sqrt{\det A} = \sqrt{\mu_{11}^{\parallel} \mu_{22}^{\parallel} k_1 k_2 \left(1 - \frac{\mu_{12}^{\parallel} \mu_{21}^{\parallel}}{\mu_{11}^{\parallel} \mu_{22}^{\parallel}} \right)}. \quad (4)$$

A simple analysis of the system with the two particles executing Brownian motion in the absence of the external drive leads us to write down the auto and cross-correlation functions (C_{ij}), so that by comparing with the response functions χ''_{ij} , we have $C_{ij} = (k_B T / \pi f) \chi''_{ij}$

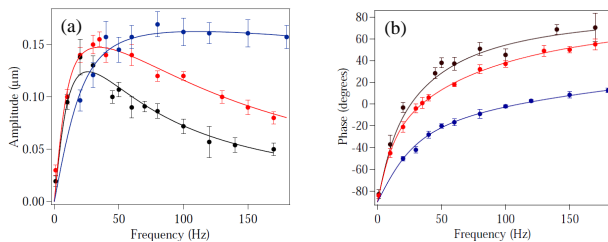


Figure 3. Amplitude and phase response for driven (B2) bead for different trap stiffness ratios with the inter-particle separation $0.67a$. (a) and (b) demonstrate amplitude and phase responses (with respect to driving frequency) of B2, for trap stiffness ratios of 2.5:1 (black), 5.7:1 (red), and 14.5:1 (blue) for (c) and (e). The resonance frequency in (c) is 20 Hz (black), 35 Hz (red), and 111 Hz (blue). The solid spheres denote experimental data points while the solid lines are corresponding theoretical fits.

which is the well known fluctuation-response relationship [28].

This is indeed what we validate in Fig.3(a)-(c). We now focus on a particularly interesting facet of our problem, namely the amplitude and phase response of B2 under the influence of the driven particle B1. We study this experimentally for three different trap stiffness ratios of B1 and B2, the results of which are shown in Fig. 3(a) and (b). Note that we fit each graph with the calculated values of the responses for the experimental parameters used, and obtain very good fits. The amplitude and phase response of B1 (Supplementary information) to the drive frequency is expected, with the amplitude decaying with increasing frequency, and the phase being in sync with the drive at low frequencies and gradually lagging behind as the frequency is increased. However, the amplitude response of B2 is rather interesting, and shows a clear resonance response at a certain frequency, the value of which increases as the stiffness ratio of the traps is increased. Thus, we have a resonance frequency of around 111 Hz (blue solid spheres in Fig.3(c)) with $k_1 : k_2 = 14.5$, a frequency of around 33 Hz with a ratio of 5.7:1 (red solid spheres), and a frequency of around 20 Hz with a ratio of 2.5:1 (black solid spheres). In fact, it is as if the entrained fluid has minimum impedance around this frequency, so that there is maximum energy transfer between the driving and the driven beads. The amplitude of the resonance is also linearly dependent on the particle separation, and falls off as the latter is increased, so that with our current detection sensitivity, we do not observe the resonance effects beyond a surface-surface separation greater than $3a$ - this increased distance also being smaller than that used in earlier experiments, which possibly explains the fact that this phenomenon has not been reported earlier. The width of the resonance (Q factor) is also dependent on the stiffness ratio, and increases as the latter is reduced. For a given medium, the

resonance can thus be tuned by changing the stiffness ratios (as well as the inter-particle separation and particle diameters). Interestingly, it is obvious that the value of the Q -factor as well as the resonance frequency also depends on the damping, and can be modified by changing the viscosity of the solution. This property promises the measurement of this frequency shift as an accurate two-point micro-rheology probe of local viscosity of a fluid. Finally, the phase response in 3(e) is easily explained: B2 lags 90 degrees in phase with respect to the drive at very low frequencies with the lag reducing until the drive and driven are in phase at resonance, after which the driven bead leads in phase, and asymptotically approaches 90 degrees at high frequencies. The rate of approach is also determined by the stiffness ratio, and is rather slow at large stiffness ratios. Indeed, this is exactly similar to the relationship between velocity and driving force for a forced damped harmonic oscillator, and arises due to the fact that the oscillators are dissipatively coupled.

In conclusion, we perform a direct experimental verification of the fluctuation-dissipation relation in a system consisting of two colloidal particles confined in a viscous medium (water) in very close proximity (surface-surface separation less than the particle radius) using separate optical tweezers. Our results provide a confirmation of the validity of the fluctuation-dissipation relation in the presence of long-ranged dissipative forces that are the only source of coupling of, otherwise, independent degrees of freedom. Surprisingly, we identify a resonance in the response in a system which is overdamped and suggest its use in accurate two-point microrheology. The present experiment can be extended in several directions: measurements at higher frequencies can uncover the effects of retarded hydrodynamic interactions and the role of particle inertia while holographic traps can be used to test the fluctuation-dissipation relation in the presence of many-body hydrodynamic interactions. Some of these will be presented in forthcoming work.

This work was supported by the Indian Institute of Science Education and Research, Kolkata, an autonomous research and teaching institute funded by the Ministry of Human Resource Development, Govt. of India. We acknowledge computing resources on the Annapurna cluster provided by The Institute of Mathematical Sciences.

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