Direct verification of the fluctuation-response relation in dissipatively coupled oscillators

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The fluctuation-response relation, a central result in non-equilibrium statistical mechanics, relates equilibrium fluctuations in a system to its linear response to external forces. Here we provide a direct experimental verification of this relation for dissipatively coupled oscillators, as realized by a pair of optically trapped colloidal particles. A theoretical analysis in which fluid-mediated interactions are represented by non-local friction tensors matches experimental results and reveals a frequency maximum in the amplitude of the mutual response function for suitably chosen ratios of optical trap stiffnesses. This amplitude "resonance" is a sensitive function of the fluid viscosity and can be tuned by the trap stiffnesses suggesting the utility of the trap setup for accurate two-point microrheology.

The classical dynamics of a pair of coupled harmonic oscillators under a variety of boundary conditions and in different environments offers interesting study, and is often used to model systems encompassing diverse areas in physics. The environment dictates the dynamics, and thus, a careful analysis of the dynamics leads to a precise calibration of the environment. In this context, the coupled-oscillator problem in a fluid has interesting connotations, especially when the oscillators in question are of mesoscopic dimensions, since they would then be responsive to fluid properties at the microscopic level. Thus, while a preliminary perception would demarcate the problem to be a typical coupled oscillator that is in the over-damped regime - the fact that the particles are Brownian, and thus undergo position fluctuations influenced by stochastic forces - renders interesting possibilities. Moreover, the microscopic flow environment that the oscillators would sample could also lead to non-trivial effects, and even produce phenomenon that would otherwise be considered unlikely due to the boundary conditions governing the over-damped regime.

In this paper, we probe the coupled dynamics of a pair of colloidal particles trapped in close proximity in water using optical tweezers.The simple system of two colloidal spheres trapped in separate optical traps has earlier been used to study the effects on one of the spheres when the other is modulated sinusoidally \cite{pre, schmiddt}. The results seemed to match the theory well - however, there seemed to be limited interest in the consequences of the results and the possibilities offered by this system as a whole to study interesting aspects of basic physics. For example, a trapped particle under optical tweezers undergoes Brownian motion, which is a Markovian process and thus obeys detailed balance, so that the fluctuation dissipation relation is almost trivially satisfied in this case. However, the situation becomes more complicated when two particles are trapped in separate optical tweezers that are coupled by near-field hydrodynamic interactions with the entrained fluid. The particles undergo Brownian motion apparently independent of each other, and while both systems have linear responses individually - so that their relaxations are exponential, it would be interesting to probe whether fluctuation dissipation is stringently satisfied for the coupled system. This is what we achieve in this paper, where we first develop a theory to verify the fluctuation response relationship for two spheres trapped in optical tweezers and coupled by hydrodynamic interactions. Using appropriate fluid-mediated interactions represented by the mobility tensors, we check whether the response of one of the particles when the other is driven sinusoidally can indeed by equated with the cross-correlations of the Brownian fluctuations of the individual particles. We then verify this experimentally on two polystyrene microspheres trapped in very close proximity of each other (surface to surface separation lesser than the particle radius), and perform measurements to check whether the fluctuation-response theorem is indeed validated. In addition, we observe that when one of the microspheres is modulated sinusoidally, the response of the other is dependent on the driving frequency of the former, so that it displays an amplitude resonance. This is typically not feasible in a highly over-damped system such as optical tweezers where the liquid entirely damps out the inertial response of the trapped object. The resonance response is a critical function of the viscosity of the liquid and can, in principle, be used to perform two point micro-rheology measurements with high accuracy.

Experiment: The details of the experimental setup towards validation of Eq. 14 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

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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 imaging white light and detection beams also separated at the output by dichroic beam splitters. A very low volume fraction sample ($\phi \approx 0.01$) is prepared with 3 μm diameter polystyrene latex beads in 1 M NaCl-water solution for avoiding surface charges. A single droplet of about 20 μl volume of the sample is introduced on a standard 10 mm square cover slip so that the droplet radius is around 200 μm. We trap two spherical polystyrene beads (Sigma LB-30) of mean size 3 μm each, in two calibrated optical traps which are separated by a distance 4 ± 0.1 μm, so that the surface-surface distance of the trapped beads is 1 ± 0.2 μm, and the distance from the cover slip surface is 30 μm. From the literature \cite{**PRL 107, 248101 (2011)**}, this distance is still large enough to avoid optical cross talk and effects due to surface charges. In order to ensure that the trapping beams do not influence each other, we measure the Brownian motion of one when the other is switched on (in the absence of a particle), and check that there are no changes in the Brownian motion. One of the traps is sinusoidally modulated 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 using photodiode pairs PA1, PB1 and PA2, PB2, for the driving and driven particles, respectively. The voltage-amplitude calibration of our detection system reveals that we can resolve motion of around 12 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). The sampling frequency is 2 kHz, while we performed data blocking at the level of 100 points in order to ensure good Lorentzian fits [flyvbjerg] 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 modu-

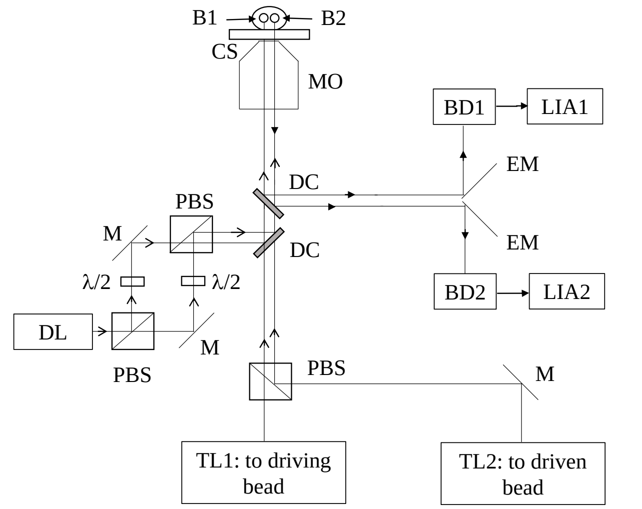

FIG. 1. A schematic of the setup is shown. TL1: Trapping laser for driving bead B1, TL2: trapping laser for driven bead B2, DL: detection laser, PBS: polarizing beam splitter cube, $\frac{\lambda}{2}$: half wave plate, DC: Dichroic mirror, MO: Microscope objective, CS: Cover slip, BD1 and BD2: balanced detection systems based on Thorlabs photodiodes PD-EC2, M: Mirror, EM: Edge mirror, LIA1 and LIA2: Lock-in amplifiers for B1 and B2, respectively.

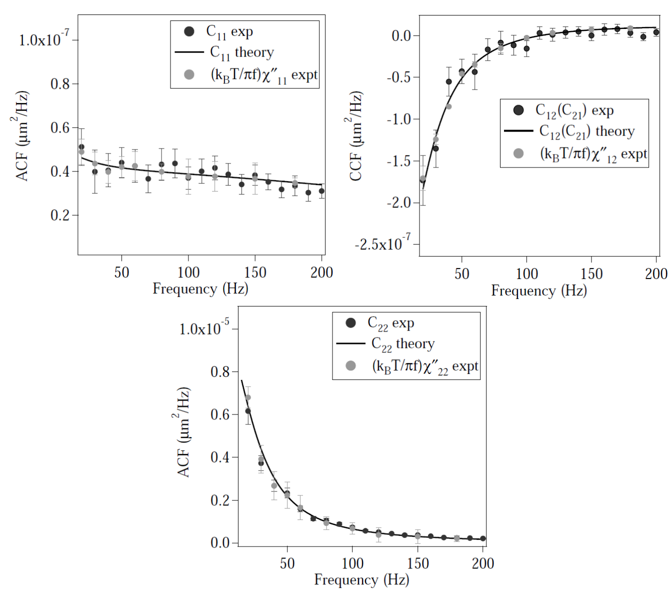

FIG. 2. Verfication of the Fluctuation-response relation for a pair of colloidal particles coupled hydrodynamically. We demonstrate this for individual matrix elements in the form of $\frac{k_B T}{\pi f}\chi"_{ij} = C_{ij}$, where (a) and (c) are for the diagonal matrix elements, and (b) is for the off-diagonal matrix element. We also fit the experimentally measured rhs, i.e. C_{ij} (the auto and cross-correlation functions), with the theoretically calculated values of the same. Note that while $C_{ij} = C_{ji}$ trivially, we are not able to measure the response $\chi"_{21}$, (response of B1 when B2 is driven), since the amplitude response of B1 (strongly trapped bead) when B2 is driven is lower than our detection sensitivity.

lated bead (B1) was 69.6 $\mu N/m$, while that of the driven is 4.8 $\mu N/m$. 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. 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 ($\chi"_{11}$, $\chi"_{22}$) 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 $\chi"_{12}$. This is obtained by measuring the amplitude and phase of B2 when B1 is driven. Note that we are not able to measure $\chi"_{21}$ 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. 2 in Supplementary Information) and obtain qualitatively similar data as reported in Ref. [Meiners and Quake].

Theory: It is naturally an interesting exercise to probe the underlying physics of the pair of trapped particles coupled by hydrodynamic interactions in a more quantitative manner. The force balance equation of an optically trapped bead is,

$$\begin{aligned} m\dot{\mathbf{v}}_i &= -\gamma_{ij}\mathbf{v}_j - \nabla_i U + \xi_i \\ \dot{\mathbf{R}}_i &= \mathbf{v}_i \end{aligned} \tag{1}$$

where, $\dot{\mathbf{R}}_i$ and $\dot{\mathbf{v}}_i$, respectively, are the position and velocity of the i-th particle of mass m, γ_{ij} is the many-body friction tensor, U is the potential, and ξ is the thermal stochastic noise. The optical potential is given by $U(t) = \frac{1}{2}\sum k_i(\mathbf{R}_i - \mathbf{R}_i^0)^2$ with $\mathbf{R}_i^0$ is the position of the potential minimum of the i-th optical trap and k_i is the corresponding stiffness. In the experimental set-up, we have two such optical traps with the minimum of one of them is being shifted with an external periodic signal, and the response of the two beads studied. Assuming momentum to be rapidly relaxing on the time scale of the trap motion, we neglect inertia and average over the noise to get,

$$-\gamma_{ij}\dot{\mathbf{R}}_j - \nabla_{\mathbf{i}}\mathbf{U} = 0 \tag{2}$$

Eq.2 can be inverted and presented in terms of mobility matrices as,

$$\begin{aligned} \dot{\mathbf{R}}_1 &= -\mu\delta k_1(\mathbf{R}_1 - \mathbf{R}_1^0) - \mu_{\mathbf{12}}k_2(\mathbf{R}_2 - \mathbf{R}_2^0) \\ \dot{\mathbf{R}}_2 &= -\mu_{\mathbf{21}}k_1(\mathbf{R}_1 - \mathbf{R}_1^0) - \mu\delta k_2(\mathbf{R}_2 - \mathbf{R}_2^0) \end{aligned} \tag{3}$$

We re-frame this equation using approximate mobility matrices with the separation vector to be the average distance between the two minima of the optical traps. Thus, one obtains,

$$\frac{d}{dt}\begin{bmatrix}\mathbf{R}_1\\ \mathbf{R}_2\end{bmatrix} = -\begin{bmatrix}\mu k_1\delta & \mu_{12}k_2\\ \mu_{21}k_1 & \mu k_2\delta\end{bmatrix}\begin{bmatrix}\mathbf{R}_1\\ \mathbf{R}_2\end{bmatrix} + \begin{bmatrix}\mu\delta & \mu_{12}\\ \mu_{21} & \mu\delta\end{bmatrix}\begin{bmatrix}\mathbf{F}_1^0\\ \mathbf{F}_2^0\end{bmatrix} \tag{4}$$

The steady state solution of the Eq.4 can easily be calculated by taking its Fourier transform. Assuming $\mathbf{A} = \begin{bmatrix}\mu k_1\delta & \mu_{12}k_2\\ \mu_{21}k_1 & \mu k_2\delta\end{bmatrix}$ and $\mathbf{M} = \begin{bmatrix}\mu\delta & \mu_{12}\\ \mu_{21} & \mu\delta\end{bmatrix}$, the solution in frequency domain is given by,

$$\mathbf{R}_i(\omega) = [-i\omega\delta + \mathbf{A}]_{ij}^{-1}\mathbf{M}\mathbf{F}_j^0(\omega) = \chi_{\mathbf{ij}}(\omega)\mathbf{F}_j^0(\omega) \tag{5}$$

which defines the response for χ. As the minimum of one of the optical traps is modulated by a sinusoidal signal of driving frequency Ω, $\mathbf{F}_j^0(\omega) = \frac{X_j}{2}(\delta(\omega-\Omega) + \delta(\omega+\Omega))$. Further, χ is a block-diagonal matrix in cartesian indices. Now, for a given experimental set-up, χ can be decomposed into $\chi_\parallel$ and $\chi_\perp$, which correspond to motion along the trap separation and that perpendicular to it, respectively. Inserting this form of the signal into Eq.5, and reverting to the time domain, we find

$$R_{\parallel i}(t) = \chi'_{\parallel ij}\cos(\Omega t)X_j + \chi"_{\parallel ij}\sin(\Omega t)X_j \tag{6}$$

The two time scales from the two traps can be calculated to be $\tau_i = \frac{1}{\mu k_i}$, so that,

$$\begin{aligned} \chi_\parallel(\Omega) &= \frac{1}{DetA_\parallel - \Omega^2 - i\Omega TrA_\parallel}\times \\ &\begin{bmatrix}(\mu k_1 - i\Omega) & \mu_{12}k_2\\ \mu_{21}k_1 & (\mu k_2 - i\Omega)\end{bmatrix}^{-1}\begin{bmatrix}\mu & \mu_{12}\\ \mu_{21} & \mu\end{bmatrix} \\ &= \frac{1}{DetA_\parallel - \Omega^2 - i\Omega TrA_\parallel}\times \\ &\begin{bmatrix}(\mu k_2 - i\Omega) & -\mu_{12}k_2\\ -\mu_{21}k_1 & (\mu k_1 - i\Omega)\end{bmatrix}\begin{bmatrix}\mu & \mu_{12}\\ \mu_{21} & \mu\end{bmatrix} \\ &= \frac{DetA_\parallel - \Omega^2 + i\Omega TrA_\parallel}{(DetA_\parallel - \Omega^2)^2 + \Omega^2(TrA_\parallel)^2}\times \\ &\begin{bmatrix}k_2 DetM_\parallel - i\mu\Omega & -i\Omega\mu_{12}\\ -i\Omega\mu_{21} & k_1 DetM_\parallel - i\mu\Omega\end{bmatrix} \end{aligned} \tag{7}$$

$$\begin{aligned} &= (DetA_\parallel - \Omega^2)^2 + \Omega^2(TrA_\parallel)^2\times \\ &\begin{bmatrix}\mu k_2^2 DetM_\parallel + \mu\Omega^2 & -(DetA_\parallel - \Omega^2)\mu_{12}\\ -(DetA_\parallel - \Omega^2)\mu_{21} & \mu k_1^2 DetM_\parallel + \mu\Omega^2\end{bmatrix} \end{aligned} \tag{8}$$

Note that we are interested in the resonance in amplitude of the second bead due to the forcing of the first bead that is given by maximizing the modulus of $\chi_{21\parallel}$ with respect to Ω.

$$
\begin{aligned}
\mid \chi_{21\parallel} \mid &= \mid \frac{-i\Omega\mu_{21}}{DetA_{\parallel} - \Omega^2 - i\Omega TrA_{\parallel}} \mid \\
&= \frac{\Omega\mu_{21}}{\sqrt{(DetA_{\parallel} - \Omega^2)^2 + \Omega^2(TrA_{\parallel})^2}}
\end{aligned} \tag{9}
$$

Clearly the resonance frequency in dimensionless unit is $\tau_1\Omega_{res} = \sqrt{\frac{DetA_{\parallel}}{\mu^2 k_1^2}} = \sqrt{\frac{k_2}{k_1}\left(1 - \frac{\mu^2_{\parallel 12}}{\mu^2}\right)}$ when $\mu_{21} \neq 0$.

Equation 5 is analogous to the expression of a forced harmonic oscillator where, resonance frequency $\omega_0^2 = DetA_{\parallel}$ and the damping term $\Gamma = TrA_{\parallel}$. The theoretical phase and amplitude response as a function of frequency are shown in Fig. 2(a) and (b). Interestingly, the expression of $\chi_{21\parallel}$ resembles that of the velocity of the forced damped driven oscillator in classical physics. This is due to the fact that the interactions in this case are mediated by the mobility of the fluid which essentially implies that amplitude (or displacement) in our system assumes the role of velocity in classical physics. Accordingly, the phase of the second bead lags behind the drive at 90 degrees at low frequencies, and leads it by the same amount at high frequencies, being in phase at resonance akin to the induced current against drive voltage in LCR circuits.

Let us now consider the situation where the external force is absent and the two trapped particles are executing Brownian motion in their respective traps. Then, from Eq.2 we obtain

$$
\begin{aligned}
\gamma_{ij}\dot{R}_j &= -k_{ij}R_j + \xi_i \\
\langle \xi_i \rangle &= 0 \\
\langle \xi_i \xi_j \rangle &= 2K_B T \gamma_{ij}
\end{aligned} \tag{10}
$$

Once again, the steady state solution in frequency space is derived easily by a Fourier transform, so that

$$
\mathbf{R}(\omega) = (-i\omega\delta + \mathbf{A})^{-1}\mathbf{M}\xi(\omega) \tag{11}
$$

Thus, the correlation matrix can be written as

$$
\langle \mathbf{R}(\omega)\mathbf{R}^{\dagger}(\omega)\rangle = (-i\omega\delta + \mathbf{A})^{-1}\mathbf{M}\langle \xi(\omega)\xi^{\dagger}(\omega)\rangle \mathbf{M}(i\omega\delta + \mathbf{A}^T)^{-1}
$$

$$
\frac{1}{2K_B T} C_{\Delta\Delta} = (-i\omega\delta + \mathbf{A})^{-1}\mathbf{M}(i\omega\delta + \mathbf{A}^T)^{-1} \tag{12}
$$

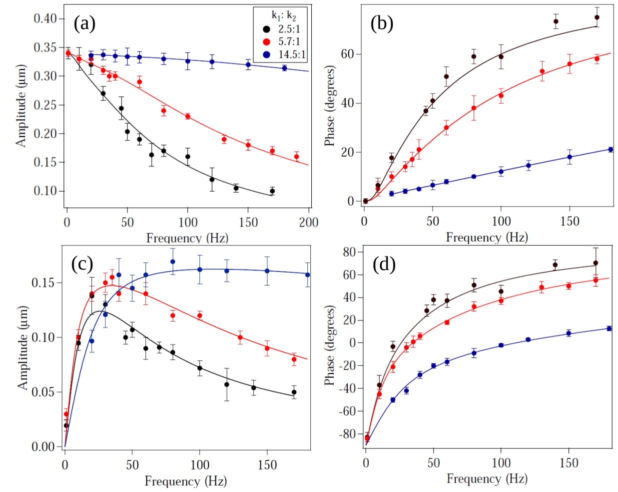

FIG. 3. Amplitude and phase response for driving (B1) and driven (B2) beads for different trap stiffness ratios. (a) and (b) demonstrate amplitude and phase responses (with respect to driving frequency) of B1, while (c) and (d) demonstrate that of B2 as functions of driving frequency, for trap stiffness ratios of 2.5:1 (black), 5.7:1 (red), and 14.5:1 (blue). 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.

$$
\begin{aligned}
&(-i\omega\delta + \mathbf{A})^{-1}_{\parallel}\mathbf{M}_{\parallel}(i\omega\delta + \mathbf{A}^T_{\parallel})^{-1} = \\
&\frac{1}{(DetA_{\parallel} - \omega^2)^2 + \omega^2(TrA_{\parallel})^2} \times \\
&\begin{bmatrix} \mu k_2 - i\omega & -\mu_{12}k_2 \\ -\mu_{21}k_1 & \mu k_1 - i\omega \end{bmatrix}
\begin{bmatrix} \mu & \mu_{12} \\ \mu_{21} & \mu \end{bmatrix}
\begin{bmatrix} \mu k_2 + i\omega & -\mu_{12}k_2 \\ -\mu_{21}k_1 & \mu k_1 + i\omega \end{bmatrix} \\
&= \frac{1}{(DetA_{\parallel} - \omega^2)^2 + \omega^2(TrA_{\parallel})^2} \times \\
&\begin{bmatrix} \mu k_2^2 DetM_{\parallel} + \mu\omega^2 & -(DetA_{\parallel} - \omega^2)\mu_{21} \\ -(DetA_{\parallel} - \omega^2)\mu_{12} & \mu k_1^2 DetM_{\parallel} + \mu\omega^2 \end{bmatrix}
\end{aligned} \tag{13}
$$

Finally, from Eqs. 7, 10, and 14, we see that the Fluctuation-response theorem holds. In matrix representation, it can be written as,

$$
2k_B T\chi^{"}(\omega) = \omega C_{xx}(\omega) \tag{14}
$$

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 for three different trap stiffness ratios of B1 and B2, the results of which are shown in Fig. 3(a) - (d). The amplitude and phase response of B1 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 changes 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). Thus, 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. In addition, the width of the resonance (Q factor) is also dependent on this ratio, and increases as the stiffness ratio is lowered. The phase response in 3(d) 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. As mentioned earlier, this is exactly similar to the relationship between velocity and driving force for a forced damped harmonic oscillator. Note that we fit each graph with the calculated values of the responses for the experimental parameters used, and obtain very good fits. An interesting point that needs to be mentioned here is that the resonance frequency is dependent on the viscosity of the fluid as shown in Eq.14, which implies that it would shift with changing viscosity. This property promises the measurement of this frequency shift as an accurate two-point micro-rheology probe of local viscosity of a fluid.

In conclusion, we study the coupled dynamics of two colloidal particles under near-field hydrodynamic interactions using optical tweezers. We verify the fluctuation-response directly in a model-free approach initially by measuring independently the auto and cross-correlations of the trapped particles, and comparing those with the measured amplitude and phase response when the particles are driven sinusoidally. The experiment is carried out on two polystyrene beads of diameter 3 um which are trapped in adjacent traps such that the surface-surface separation of the beads is 1 micron. The stiffness of the modulated bead is kept around 15 times higher than the driven bead to eliminate back-flows on the former and to ensure maximum amplitude response of the latter. To understand the dynamics of the problem more quantitatively, we develop a theoretical model where we solve the force balance equation invoking the mobility tensors for the system when both particles are at rest, as well as when is one of them is sinusoidally modulated. We generate amplitude and phase relationships between the response of the driving and driven particles with respect to the modulation frequency, and obtain the exact statement of the fluctuation-response theorem, which is rather non-trivial for a two particle system. In the process, we observe that the phase response of the driven bead resembles the current in an LCR circuit, and the amplitude actually undergoes a resonance at a particular driving frequency where the transfer of energy is maximum between drive and driven. We note that the resonance position and width are sensitive function of the ratio of the trap stiffnesses, so that a judicious choice of stiffness is required to observe effects with good SNR. Finally, the amplitude resonance frequency also depends on the fluid viscosity, which promises a direct application of this technique as a sensitive two-point microrheology probe of local viscosity.