Parametric Resonance of Optically Trapped Aerosols

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The Brownian dynamics of an optically trapped water droplet are investigated across the transition from over to under-damped oscillations. The spectrum of position fluctuations evolves from a Lorentzian shape typical of over-damped systems (beads in liquid solvents), to a damped harmonic oscillator spectrum showing a resonance peak. In this later under-damped regime, we excite parametric resonance by periodically modulating the trapping power at twice the resonant frequency. The power spectra of position fluctuations are in excellent agreement with the obtained analytical solutions of a parametrically modulated Langevin equation.

Parametric resonance provides an efficient and straightforward way to pump energy into an under-damped harmonic oscillator\textsuperscript{[1]}. In general, if the resonance frequency of an oscillator is dependent upon a number of parameters modulating any of these at twice the natural oscillation frequency parametrically excites the resonance. Such behavior leads to surprising phenomena in the macroscopic world (pumping a swing, stability of vessels, surface waves in vibrated fluids)\textsuperscript{[2, 3]}. On the microscopic scale, where stochastic forces become important, one refers to Brownian parametric oscillators\textsuperscript{[4]}. As an example, the parametric driving of Brownian systems has been shown to be at the origin of some peculiar behaviors such as the squeezing of thermal noise in Paul traps\textsuperscript{[5]}. Parametrically excited torsional oscillations have also been reported in a single-crystal silicon micro electromechanical system\textsuperscript{[6]}. What makes parametric resonance useful is that in many cases it is easier to modulate a system parameter rather than applying an oscillating driving force. Moreover, for finite but low damping rates, we may never reach a stationary state with the damping forces dissipating all of the input power and consequently the amplitude of oscillations diverge.

Optically trapped microparticles constitute a beautiful example of Brownian damped harmonic oscillator (DHO)\textsuperscript{[12]} and they are becoming an increasingly common tool for the investigation of different fields of basic and applied science\textsuperscript{[7]}. The possibility of pumping mechanical energy into optically trapped particles could open the way to many applications. In optical tweezers, even though it is easy to periodically modulate the laser power, parametric excitation is usually ineffective because of the heavy damping action of the surrounding fluid.

Recently it has been reported that modulating the laser power at the parametric resonant frequency in an overdamped system increased the amplitude of mean squared fluctuations\textsuperscript{[8]}. However, these findings have been difficult to reproduce and are in strong contrast with the prediction of Langevin dynamics\textsuperscript{[9, 10, 11]}.

In this Letter we show how parametric resonance can be excited in optically trapped water droplets suspended in air, due to the reduced damping force. We measure power spectra of position fluctuations and find an excellent agreement with the theoretical expectations based on Langevin dynamics with a parametric forcing.

The dynamics of an optically trapped droplet is described by the Langevin equation\textsuperscript{[12]}:

\[
\ddot{x}(t) + \Omega_0^2 x(t) + \Gamma_0 \dot{x}(t) = \xi(t) \tag{1}
\]

where \( \Omega_0^2 = k/m \) is the natural angular frequency of the oscillator depending on trap stiffness \( k \) and particle mass \( m \). \( \Gamma_0 = 6\pi \eta a/C, m \) is the viscous damping due to the medium viscosity \( \eta \) and depending on particle radius \( a \) and mass \( m \). One must be careful to consider the non-continuum effects of Stokes’ Law in air due to the finite Knudsen number of the particles under study. To correct Stokes’ Law the empirical slip correction factor, \( C, \), is introduced, with a 5.5-1.6\% reduction in drag for 3-10\( \mu m \) diameter droplets respectively\textsuperscript{[13]}.

The stochastic force \( \xi \), due to thermal agitation of solvent molecules, is generally assumed to be uncorrelated on the time-scale of particle’s motion:

\[
\langle \xi(0)\xi(t) \rangle = 2\Gamma_0 K_B T/m \delta(t) \tag{2}
\]

The corresponding power spectrum \( S_\xi(\omega) \) of the stochastic variable \( \xi \) can be defined as:
obtain the power spectrum of position fluctuations as:

\[ \langle \dot{\xi}^*(\omega)\dot{\xi}(\omega') \rangle = S_\xi(\omega)\delta(\omega - \omega') \]  

(3)

where:

\[ \dot{\xi}(\omega) = 1/2\pi \int_{-\infty}^{\infty} \xi(t) \exp[-i\omega t] dt \]  

(4)

By putting (4) and (2) in (3) it is easily shown that \( \xi \) has a white noise spectrum

\[ S_\xi(\omega) = \frac{\Gamma_0 K_B T}{\pi m} \]  

(5)

By Fourier transforming (1) and using (5) we can easily obtain the power spectrum of position fluctuations as:

\[ S_x(\omega) = \frac{K_B T}{k} \frac{1}{\pi (\omega^2 - \Omega_0^2)^2 + \Gamma_0^2 \omega^2} \]  

(6)

When trapping objects in liquid solvents the ratio \( \Gamma_0/\Omega_0 \) is always larger than one, i.e. the system is overdamped. The ratio \( \Gamma_0/\Omega_0 \) depends only slightly on particle’s size and, in solvents with water-like viscosities, is always greater than 1 up to power levels of some tens of Ws. For typical trapping powers of order 10 mW in water \( \Gamma_0/\Omega_0 \) is typically > 10. As a result only those frequencies smaller than \( \Gamma_0 \), and hence much smaller than \( \Omega_0 \), have a significant amplitude in the power spectrum of \( x \). Under these conditions we can therefore neglect \( \omega^2 \) with respect to \( \Omega_0^2 \) in the first term of the denominator in (6) and obtain the usual Lorentzian power spectrum characterised by \( \omega^2 \) tails [14]. Such an overdamped condition precludes the possibility of exciting significant oscillations either directly or parametrically. To probe oscillations in the liquid damped regime we would need to be able to increase typical trap power by four orders of magnitude - introducing uncontrollable heating and damage of the trapped object. A more feasible route is to reduce viscosity by two orders of magnitude. This last condition can be readily obtained by trapping particles in air whose viscosity is approximately \( 1/55^{th} \) of water \( (\eta = 1.8 \times 10^{-5} \text{ Pa s}) \) [13].

For these experiments, our optical tweezers are based around an inverted microscope with a high numerical aperture oil immersion microscope objective (1.3NA, 100×). The continuous wave laser is a Nd:YAG, frequency doubled to give 0.2 W of 532 nm light. To couple the beam into the air medium, a single cover slip is rested over the objective on a thin oil layer. A water aerosol is produced using a nebulizer and usually a 3-10 \( \mu \text{m} \) diameter water droplet is trapped at the beam focus [12][10], see Fig. 1. One should note that whereas for particles trapped in fluid, a laser power of 10s of mW is typical, here, to maximise the stiffness of the trap, we use powers of order 1 W.

FIG. 1: The measured power spectra of trapped aerosol particle at two different powers. At lower power (black circles) it is overdamped and the mean squared amplitude of the high frequency motion decays as \( \omega^{-2} \). At higher powers (white circles), the aerosol is underdamped and the mean squared amplitude decays as \( \omega^{-4} \). The inset shows an optical image of a trapped aerosol particle.

A quadrant photo detector, placed in the back focal plane of the condenser lens, receives the light transmitted through the droplet. By measuring the imbalance of the light collected by the quadrants, the lateral displacement of the droplet is deduced with a bandwidth of several kHz and a precision of better than 5 nm [17].

The power spectra of the measured displacement, for two different trap powers, are shown in Fig. 1. It is clearly visible how the particle dynamics crosses over from an overdamped dynamics with a Lorentzian spectrum with a high frequency roll-off proportional to \( \omega^{-2} \) to an underdamped regime with a faster roll-off, \( \omega^{-4} \), and the appearance of a resonance peak at a frequency of about 1 kHz. The emergence of such a peak arises from the fact that the inertial terms in (1) are no longer negligible. As a consequence an average trajectory starting away from the equilibrium position crosses the equilibrium position with a finite velocity.

In this situation the parametric resonance is excited by modulating the strength of the trapping potential. Ideally the potential is made shallower when the particle traverses the equilibrium position and steeper again when the particle is far from the equilibrium position. This is maximally efficient when we modulate the potential at twice the natural oscillation frequency \( \Omega_0 \). To consider this model in quantitative terms we can rewrite (1) in the presence of a parametrically modulated external potential:
\[
\ddot{x}(t) + \Omega_0^2 [1 + g f(t)] x(t) + \Gamma_0 \dot{x}(t) = \xi(t)
\] (7)
\[
f(t + T) = f(t), \quad -1 < f(t) < 1
\] (8)

where \(0 < g < 1\) measures the strength of modulation. By Fourier transforming (8) we obtain:

\[
(-\omega^2 + \Omega_0^2 + i\omega \Gamma_0) \hat{x}(\omega) + \Omega_0^2 g \sum_{k=-\infty}^{\infty} a_k \hat{x}(\omega + k \Omega_1) = \hat{\xi}(\omega)
\] (9)

where \(a_k\) is the coefficient of the \(k2\pi/T = k\Omega_1\) frequency component of the Fourier series expansion of \(f(t)\). It is clear from equation (9) how parametric modulation introduces a coupling between all those frequencies differing by an integer number of \(\Omega_1\). We now introduce the vectors \(X_n(\omega) = \hat{x}(\omega + n \Omega_1)\) and \(R_n(\omega) = \hat{\xi}(\omega + n \Omega_1)\) and write the recursive relations:

\[
-(\omega^2 + \Omega_0^2 + i\omega(\omega + n \Omega_1)) X_n(\omega) + \Omega_0^2 g \sum_{k=-\infty}^{\infty} a_k X_{n+k}(\omega) = R_n.
\] (10)

To obtain the power spectrum \(S_x(\omega)\), for each frequency \(\omega\) we should compute \(X_0(\omega)\). This will be, in turn, coupled to all other components in the array \(X_n\). However the strength of the coupling will decay for large \(|n|\) so that we can limit ourselves to a finite number of components and write the matrix equation for the array \(\mathbf{X}(\omega) = [X_{-N}(\omega), ..., X_N(\omega)]\):

\[
\mathbf{G}^{-1}(\omega) \mathbf{X}(\omega) = \mathbf{R}(\omega)
\] (11)

with

\[
\mathbf{G}^{-1}_{nk}(\omega) = \frac{1}{-(\omega+n \Omega_1)^2 + \Omega_0^2 + i(\omega+n \Omega_1) \Gamma_0} \delta_{nk} + \Omega_0^2 g a_{k-n}
\] (12)

By matrix inversion we obtain the power spectrum as:

\[
\langle X_n^* (\omega) X_0(\omega') \rangle = \sum_{k,n=-N}^{N} G_{0k}(\omega) G_{0l}(\omega') \langle R_k^*(\omega) R_n(\omega') \rangle = \frac{\Gamma_0 K_B T}{\pi m} \sum_{n=-N}^{N} |G_{0n}(\omega)|^2 \delta(\omega - \omega')
\] (13)

and from the definition of power spectrum:

\[
S_x(\omega) = \frac{\Gamma_0 K_B T}{\pi m} \sum_{k=-N}^{N} |G_{0k}(\omega)|^2
\] (14)

If \(\Omega_0\) and \(\Gamma_0\) are known, we can use (14) to predict the power spectrum of a Brownian particle in a modulated trap. The white circles of Fig.2 show the measured power spectrum for a trapped water droplet when the trap has constant power. The presence of the peak suggests that we are in an underdamped regime. By fitting to equation (14) (solid line) we can directly extract the resonant frequency \(\Omega_0/2\pi = 2.0\) kHz and the damping term \(\Gamma_0 = 6.8\) kHz relevant to our experimental conditions. The fitted value of \(\Gamma_0\) corresponds to the Stokes drag on an aerosol droplet of radius 3.4\(\mu\)m.

We then apply the square-wave modulation of the trapping power, with the laser adjusted to give the same average power as before, \(\Omega_1 \approx 2\Omega_0\) and \(g = 0.4\). The black circles of Fig.2 show the power spectrum of the lateral motion for the trapped water droplet with modulated laser power. The excitation of a resonance appears as a higher and narrower peak in the power spectrum. This observed spectrum matches closely the expected behavior (solid line) obtained by applying the measured parameters \(\Omega_0, \Gamma_0, \Omega_1, g\) to equation (14), strongly supporting our interpretation of this peak being due to a parametric excitation of the resonance. Higher harmonics, characterizing the response of parametrically driven systems, are suppressed in our case, being only slightly underdamped.

Using these parameters with (14) we can make general predictions about the dynamics of a particle that can then be verified against experiments. One comparison to make is the predicted and observed form of the power spectra as a function of the modulation frequency, both above and below the parametric resonance condition. These results are shown in Fig.3, the white circles are the experimental points and the black lines are the predicted spectra. Again, there is an excellent agreement between the observed and predicted particle motion. In particular the parametric excitation of oscillations manifests as a narrowing of the peak (or a reduced apparent damping \(\Gamma\), defined as the full width half maximum), occurring when modulating at twice \(\Omega_0\). A shift in peak position \(\Omega_p\) is also apparent close to parametric resonance. Both of these signatures are compared with theoretical expectations in Fig.4 further supporting our interpretation of the system as being a Brownian parametric oscillator.

We recognise that the system we report here relates to study of only the lateral motion of the trapped droplets. In keeping with other work we note from examination of the video images that the axial movement of the trapped droplet has significant amplitude on much longer timescales, corresponding to frequencies in the region of 10-50Hz. This reflects the comparatively weak axial trapping, possibly arising from aberrations associated with non optimised objectives or the increased scattering force. It may be possible to use doughnut or Laguerre-Gaussian beams having zero on-axis intensity, and improved axial trapping.

We have reported the first observation of a parametrically excited resonance within a Brownian oscillator.
FIG. 2: The measure power spectrum of a trapped water droplet for no modulation of the laser power (white circles) and modulation at 3.9 kHz ($\Omega_1 \approx 2\Omega_0$) (black circles). The peak is higher and narrower on the resonant condition thus indicating parametric excitation. Solid line below black circles is the predicted spectrum from [14].

FIG. 3: Evolution of position power spectra on varying the modulation frequency $\Omega_1$. Parametric excitation of oscillations is evident at the parametric resonance condition $\Omega_1/\Omega_0 = 2$. The solid lines are the theoretical predictions from [14].

The demonstration of this effect within optical tweezers for this purpose was made possible by relying on the viscosity of air to lightly damp the motion of a trapped aerosol droplet. The detailed observed dynamics match closely the power spectra predicted from a parametrically modulated Langevin equation, where all parameters were fixed from examination of the non modulated system.

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