Anharmonic aerosol particle dynamics at Mie resonances in modulated counter-propagating optical tweezers

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ABSTRACT

We analyze the trapping potential of a setup of counter propagating optical tweezers and test the validity of the harmonic approximation in the dimension parallel to the trapping beam axis. While the dynamics of spherical droplets is very well described by a harmonic oscillator, we find that the trapping potential changes drastically for particles at resonance with the trapping laser, at so called Mie resonances. Our findings are discussed with regard to possible origins of this phenomenon.

Keywords: optical trapping, harmonic oscillator, mass, aerosols, single particles

1. INTRODUCTION

Optical trapping is a potent tool to characterize various aerosol species, as it allows to confine and isolate single aerosol particles over extended periods of time, enabling the measurement of single particle properties in a controlled environment1-3. The literature on measurements performed on single optically trapped particles encompasses the determination of size, refractive index, chemical composition, shape and, for liquid droplets, viscosity and surface tension4-8. In addition, recent studies have demonstrated novel techniques to measure the mass of optically trapped particles in the submicrometer size range and above9-14, a property previously inaccessible to optical methods.

Most optical mass measurement techniques presented in the literature use the harmonic oscillator model to describe the particle in the trapping potential and deduce the particle’s mass from measurements of the particle dynamics in response to a given excitation force9, 11, 14. The validity and limitations of this model for the description of real trapping potentials are key factors determining the accuracy of a given mass measurement technique. Most studies, however, do not report tests of the validity of the underlying model, instead only quoting an overall uncertainty of the mass measurement, without disentangling model limitations from contributions of other experimental parameters. Investigating the applicability of the harmonic potential to improve modelling of particle dynamics in specific optical trapping setups will therefore help increase the accuracy of single particle mass measurements.

In the following we present a setup of counter-propagating optical tweezers (CPT) for the mass measurement of submicrometer to micrometer sized aerosol particles with accuracies of approximately 100fg (see also Reich et al.9). We focus on tests of the validity of the harmonic approximation to the trapping potential and report on a particular exception that occurs when the trapped particle is close to a Mie resonance, that is, if it has a size such that the monochromatic trapping laser resonates inside the particle. While this phenomenon is still awaiting a full theoretical explanation, we discuss possible origins.

2. EXPERIMENTAL SETUP

The CPT setup used for trapping single particles in nitrogen gas is shown in Fig. 1 (see also Reich et al.9). Light from a continuous wave laser (Laser Quantum, wavelength 532nm, typical power 1.5W) passes through an optical isolator (Thorlabs IO-5-532-HP) to avoid optical feedback into the laser system, before being expanded by a factor of 4 and split into two beams at the polarizing beam splitter (PBS) cube. The two counter-propagating beams are aligned on a single axis and focused into the trapping cell to create the trapping potential for the aerosol particles between the two foci. The power
ratio of the two beams depends on the polarization state before the PBS, which can be adjusted with the electro-optic modulator (EOM, ConOptics 350-50-01-RP). By adjusting the voltage applied to the EOM, the power ratio of the two beams and thereby the trapping position of the particle along the beam axis can be modulated on a timescale up to MHz. The polarization directions of the two beams after the PBS are orthogonal to each other. Half wave plates in each of them allow us to investigate the dependence of the trapping potential on the polarization of the two beams by adjusting their respective polarization axes.

Figure 1 Sketch of the counter-propagating optical tweezers. The laser beam is directed through the optical isolator (OI), a half-wave plate \(\lambda/2\) and the electro-optic modulator (EOM) and expanded by a factor of 4 using two aspherical lenses (AL) before it is split into two beams at the polarizing beam splitter (PBS) cube. The polarization direction of each beam is adjusted with a half-wave plate before the beams are focused into the trapping cell to form the electrodynamic trapping potential. (see also Reich et al.²)

During the modulation of the trapping beams, the particle position is recorded using a position sensitive photodiode (PSP Thorlabs PDP90A) as shown in Fig. 2. The elastically scattered light is collected under a central angle of 90° and focused into a single spot on the PSP to track the particle movement. The analog signal from the PSP is recorded using a lock-in amplifier (medium frequency lock-in, MFLI, Zurich Instruments). The MFLI signal output is used to drive the sinusoidal modulation of the voltage applied to the EOM. The position signal can be recorded either in real time or as a demodulated phase and amplitude using the signal output voltage as reference.

Figure 2 Sketch of the particle tracking setup. The elastically scattered light is collected under a mean angle of 90° before it is focused onto the position sensitive photodiode (PSP). The position signal of the PSP is recorded by the lock-in amplifier (MFLI) in real time or as demodulated amplitude and phase using the modulation signal of the electro-optic modulator (EOM signal) as reference, which is also generated by the MFLI. (see also Reich et al.²)
To control the relative humidity (RH) inside the cell during measurements, a mixture of wet and dry nitrogen flows with
individually adjustable flow rates passes the lower part of the cell as shown in Fig. 3. The flow is sufficiently far away
from the particle trapping position not to interfere with the measurements. The particles in this study are aqueous NaCl
droplets and aqueous glycine droplets, which are generated from solution using an atomizer (TSI 3076) with pressurized
humidified nitrogen and delivered to the trapping region via the inlet at the top of the cell. The use of humidified nitrogen
ensures that the particles remain in a liquid state until they reach the trapping cell. The water content of the liquid droplets
adjusts to the RH of the surrounding, which allows us to tune the size of the droplet by adjusting the RH in the cell. A
sensor (Sensirion SHT31) placed a few millimeters from the particle is used to measure temperature and RH during the
measurements.

The size of the trapped droplet is measured using broad-band light scattering (BLS)4. As shown in Fig. 3, light from a Xe
source (Energetiq LDLS EQ-99XFC, spectral range 190-2100nm) is focused into the cell through the bottom window and
the elastically scattered light is collected under approximately 37° through the top window. The spectrum of the scattered
light is recorded in the wavelength range 330-490nm using a spectrometer (Andor Kymera 328i) and analyzed in terms of
Mie theory to determine the radius and the wavelength dependent refractive index of the droplet.

![Figure 3 Sketch of the trapping cell.](image)

3. MODEL OF THE DYNAMICS IN THE OPTICAL TRAP

The dynamics of a particle trapped in the modulated CPT is calculated by Newton’s second law9. The force acting on the
trapped particle is given by the optical force exerted by the laser and by the damping due to the surrounding medium
(nitrogen):

$$ m \ddot{z} = F_{\text{opt}} + F_{\text{damp}} $$

where $m$ is the particle mass. We denote the direction along the trapping beams with $z$ and write the optical force along $z$
as

$$ F_{\text{opt}} = -k (z - z_0) $$

where $z_0$ is the position of the harmonic potential minimum and $k$ is the optical stiffness of the trap. In our CPT the typical
optical force on the particle perpendicular to $z$ is much stronger than parallel to $z$ so that the particle is assumed to be
confined to the $z$ axis at all times and the particle dynamics perpendicular to $z$ can be neglected. The damping force of the particle due to the surrounding nitrogen is proportional to the particle’s speed, therefore we write

$$F_{\text{damp}} = -\gamma \dot{z}$$

where $\gamma$ is the linear drag coefficient. Eq. (3) is applicable for low Reynolds numbers of the particle, which is fulfilled for the observed range of particle speeds in our CPT.

For a sinusoidal modulation of the EOM, $z_0$ can be written as a function of time as follows:

$$z_0(t) = Z_0 \sin(2\pi f t)$$

where $f$ and $Z_0$ are the modulation frequency and amplitude, respectively. Finally, combining Eq. (1) - (4), we find the following expression describing the particle dynamics in the CPT:

$$\ddot{z} + \gamma \frac{k}{m} \dot{z} + \frac{k}{m} z = Z_0 \frac{k}{m} \sin(2\pi f t)$$

Eq. (5) is solved by

$$z(t) = A(f) \sin(2\pi ft - \phi(f))$$

with the oscillation amplitude $A$ and phase $\phi$.

$$A(f) = Z_0 \frac{\Omega_0^2}{\sqrt{(\Omega_0^2 - f^2)^2 + (\Gamma_0 f)^2}}$$

$$\phi(f) = \tan^{-1}\left(\frac{\Gamma_0 f}{\Omega_0^2 - f^2}\right)$$

where we have introduced the eigenfrequency of the system $\Omega_0 = \frac{1}{2\pi} \sqrt{\frac{k}{m}}$ and the damping rate $\Gamma_0 = \frac{1}{2\pi} \frac{\gamma}{m}$. The factor of $1/2\pi$ converts the units from angular frequency to frequency.

4. DATA ANALYSIS

The following explains the data processing to determine the phase of the oscillation $\phi$ and the radius of the droplet $R$ using the raw data from the PSP and the BLS spectrum, respectively.

4.1 Position sensitive photodiode data

Two channels of the PSP are used for the particle tracking, SUM and Xdiff. The SUM signal is proportional to the intensity of the particle image on the PSP and the Xdiff signal is proportional to both the intensity and the position of the particle image on the PSP. Thus the position signal corresponds to $X_{\text{diff}}$, which is obtained using a homemade voltage divider. The impedance of the PSP and its read-out electronics artificially increases the measured phase compared with the true phase of the particle oscillation. To determine this increase, the PSP is placed in one of the trapping beams during modulation and the phase of the power modulation is measured for different modulation frequencies from 1-5000Hz. Since the EOM and the light propagation operate on much faster timescales, the observed phase is entirely due to the impedance of the PSP and its read-out electronics. This phase is subtracted from every phase measurement to obtain the true phase of the particle oscillation at a given frequency.

4.2 Broad-band light scattering spectra

The raw BLS spectra are first background subtracted and normalized by the spectrum of the broad-band source. The resulting spectra are then analyzed by fitting simulations of the light scattering cross-section in the given wavelength range of 330-490nm using Mie theory. The free fit parameters are the radius of the particle and the first two Cauchy expansion coefficients of the wavelength dependent refractive index, $n(\lambda) = n_0 + n_1 \left(\frac{475 \text{nm}}{\lambda}\right)^2 - 1$. The reference wavelength 475nm is largely arbitrary, here it corresponds to the wavelength where the raw spectrum typically has the largest signal. The
mean scattering angle is 37° and the half angle of the collected scattering cone is 13°. Experimental spectra and simulations are compared by eye and the typical accuracy for the fitted radius is 1%.

5. RESULTS AND DISCUSSION

Oscillation amplitude and phase of a liquid NaCl droplet in the modulated CPT are shown in Fig. 4. The modulation frequency is swept from 20 to 2000Hz and the measurement is repeated for four different modulation amplitudes. The four modulation amplitudes are indicated by the amplitude of the voltage applied to the EOM and correspond to approximate $Z_0$ values of 10, 20, 30, 40μm (1mV of EOM voltage ~ 1μm of particle displacement). As can be seen from Fig. 4a, the measured phase curves are virtually identical, which implies, that $\phi$ does not depend on $Z_0$ in agreement with Eq. (7). Since this is a feature specific to the harmonic potential, we conclude that the harmonic potential is a very good approximation for this particle. Fig. 4b confirms that $A$ scales linearly with $Z_0$ in agreement with Eq. (7).

Figure 4 Phase $\phi$ and amplitude $A$ as a function of modulation frequency $f$ in the frequency range 20-2000Hz. a Phase measurements for four different modulation amplitudes, 10, 20, 30, 40mV, and best fit using Eq. (7). The phase measurements coincide with each other in agreement with Eq. (7). The best fit parameters are $\Omega_0 = 1122Hz$ and $\Gamma_0 = 4762Hz$. b Amplitude measurements and best fits for the same modulation amplitudes and frequencies. The best fits are obtained using Eq. (7) by setting $\Omega_0 = 1122Hz$, $\Gamma_0 = 4762Hz$ according to the phase fit and optimizing $Z_0$. As seen by the y-intercept of the best fit curves, the $Z_0$ values are proportional to the modulation amplitude in agreement with Eq. (7). The data of Fig. 4a show the same trend as those in Fig. 2 of Reich et al.9, but compared with the latter they were recorded at higher laser power.

This first example represents a particle that is not at Mie resonance, and the harmonic oscillator model is able to describe the particle dynamics very well. This contrasts with the behavior of an aqueous glycine droplet in the trap at an RH adjusted so that the particle size closely matches the Mie resonance condition. Fig. 5 shows the position signal in the modulated CPT over a few oscillation periods for different orientations of the polarization directions of the trapping beams. Harmonic oscillation is again observed for cross-polarized beams (Fig. 5a and b), as evident from the sine shaped waveforms. For parallel-polarized beams (Fig. 5c and d), however, the particle movement is no longer sinusoidal at all. Instead the particle is occasionally locked at certain positions for a fraction of the period. In Fig. 5c at approximately 1.3s, the particle even stays locked for a whole period, seemingly insensitive to the change of the trapping potential over that period. The behavior in Fig. 5 is typical for particles at Mie resonance and is equally observed for NaCl droplets9, both at low frequencies and at frequencies in the order of kHz. This suggests that the effective trapping potential changes drastically when the particle size approaches a value matching the resonance condition for parallel-polarized trapping beams. Notably, this phenomenon is not observed when the beams are cross-polarized.
Figure 5 Position signal versus time of a particle at Mie resonance for different trapping beam polarizations. a and b correspond to cross-polarized trapping beams and the waveforms are approximately sinusoidal indicating harmonic oscillation. c and d correspond to parallel-polarized beams. Deviations from the harmonic oscillation are seen at the points of largest displacement of the particle, where the waves form plateaus and in c at approximately 1.3s. The modulation frequency for all measurements is 2Hz.

In the absence of a full theoretical explanation of the step-wise oscillatory motion observed for parallel-polarized trapping beams, we limit ourselves to a qualitative discussion of possible origins of the observed phenomenon. In CPT, anharmonic contributions to the trapping potential can in principle arise from interference between the two sufficiently coherent trapping beams. For example, simultaneous trapping of particles at multiple positions using CPT has been demonstrated in the literature\textsuperscript{15, 16} for setups where the path difference between the two trapping beams is shorter than the coherence length of the laser. In our setup, however, we chose a path difference between the two trapping beams of more than 50cm on purpose, far exceeding the coherence length of the laser of approximately 7mm. Therefore, we expect the two trapping beams to be completely incoherent and exclude coherence phenomena as an explanation of the pronounced non-harmonic oscillation observed.

For incoherent beams the impact of the two beams on the particle can be treated separately. If the strong trapping forces perpendicular to the direction of propagation confine the particle on the axis of propagation (the z axis) at all times, as argued above, and if the particles are spherical, as we assumed, the optical force exerted by either beam should not depend on the direction of the polarization. This is clearly not fulfilled close to Mie resonances, as the results in Fig. 5 demonstrate, hinting at a breaking of the (cylindrical) symmetry at the origin of the observed phenomenon. One source of symmetry
breaking could be a misalignment of the two trapping beams. Given however, that we observed the phenomenon consistently even after repeated attempts at realigning the trap, accidental trap misalignments would appear to be a rather unlikely explanation of the observed particle dynamics. If not the trap alignment, the particle itself must be responsible for the symmetry breaking.

We propose that the observed phenomenon arises from deformations of the particle at Mie resonances, temporarily breaking its spherical symmetry. Deformations of the spherical shape due to the optical pressure exerted by the laser beams are typically very small for micrometer sized aqueous droplets\textsuperscript{17}. For a particle at Mie resonance, however, the optical pressure is amplified considerably. The deformation of the particle is correspondingly larger and might explain why the phenomenon is only observed for particle sizes closely matching the resonance condition. Furthermore, the relative orientation of the polarization of the trapping beams also becomes relevant in this scenario because the effects from the individual beams add up for parallel polarization, resulting in a larger deformation of the droplet than for cross-polarized beams. While this hypothesis is consistent with the observed phenomena, a quantitative investigation of the effect of optical pressure on the droplet dynamics is called for. This will require precise simulations of all the optical forces in the modulated CPT together with the ensuing droplet deformation.

6. CONCLUSION

We have investigated the trapping potential of a CPT along the trapping beam axis and tested the validity of the harmonic approximation. While in general the harmonic oscillator model is able to reproduce the observed particle dynamics very well, a notable exception occurs for particles sizes matching closely the Mie resonance condition for the wavelength of the trapping laser. We propose that this phenomenon is a consequence of the droplet deformation due to the optical stress exerted on the particle by the trapping laser. Further quantitative investigations are needed to fully explain the observed phenomenon.

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