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Thermo-optical resonance locking of an optically trapped salt-water microdroplet

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Abstract. We demonstrate that it is possible to lock the radius of an optically trapped salt-water microdroplet to the \(n = 1\) whispering gallery resonances (WGRs) at the trapping laser wavelength. The optical properties of the droplet are determined using stimulated Raman scattering. The droplet is in thermodynamic equilibrium with the surrounding vapour and the proposed locking mechanism consists of a balance between bulk heating and WGR heating. Raman measurements allow the size parameter \((nka)\) of the droplet to be determined with a precision of \(\sim 10^{-5}\) and the resonance linewidth to be estimated.

Contents

1. Introduction  2
2. Experimental setup  2
3. Observations and measurements  3
   3.1. Experimental observations  3
   3.2. Radius and refractive index determination  4
   3.3. Analysis of the Raman fingerprint  5
4. Modelling of the locked regime of the droplet  6
5. Conclusion  8
Acknowledgments  9
References  9

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1. Introduction

Spherical optical microresonators have been studied extensively and are characterized by high quality-factor resonances with very small mode volumes. Potential applications extend from their use as optical sensors [1], biosensors [2], Raman and dye-doped microlasers [3, 4] to metrology [5]. Fused-silica spheres offer the highest achieved quality factors in microspheres (∼8 × 10⁹) due to their low absorption in the telecoms spectral range [6]. An alternative approach uses liquid droplets, which can be rapidly created and adopt a spherical shape with a very high surface quality. Microspheres or microdroplets support high quality-factor whispering gallery resonances (WGRs) which can result in much lower excitation threshold intensities for nonlinear processes such as lasing, stimulated Raman scattering [7]–[9], and stimulated Brillouin scattering, than is commonly observed from bulk samples. In addition, liquid microlasers offer the possibility of tuneable frequency combs if the droplet diameter can be controlled precisely [5].

It has been observed [10] that the double resonance of excitation and fluorescence occurring in a microdroplet can induce thermally driven size-bistabilities. Arnold et al reported that the double resonance excitation of weakly absorbing aqueous droplets containing the dye crystal violet, ∼10⁻⁵ M, can exhibit a thermally driven size-bistability in cavity size (droplet radius) during wavelength tuning, dependent on the direction of wavelength scan. More recently, the control of droplet temperature through illumination by an infrared heating laser has provided an approach for regulating the WGR wavelengths observed in fluorescence from a dye-doped glycerol/water droplet on a super-hydrophobic surface [11].

In this paper, we demonstrate that the radius of a micron-size optically trapped salt-water droplet in a constant relative humidity (RH) atmosphere can be locked on the blue-shifted side of a WGR, providing fine control of its frequency-comb resonance spectrum. The locking mechanism involves laser-heating enhancement by TE- and TM-polarized WGRs at the trapping wavelength of 532 nm. In this experiment, the heating is due to the very weak absorption by water [12]. The size of a trapped salt-water droplet is a stationary state involving the surrounding RH, ambient temperature, droplet surface temperature and the droplet radius. Thus, by controlling the RH [13] or the optical trapping power (OTP) (which alters heating), the size of aqueous solution droplets can be regulated and the locked cavity size controlled.

2. Experimental setup

The experimental setup (figure 1) is based around a standard inverted microscope system (Nikon TE2000). The optical tweezers are created using a 532 nm Ventus laser (laser quantum) expanded to overfill the back aperture of a 1.25 NA oil-immersion microscope objective after reflection from a dichroic beam splitter. The back-scattered Raman light coming from the droplet is imaged, through an additional red-filter, onto the slit of a spectrograph, which is equipped with a 1200 g mm⁻¹ grating giving rise to a spectral dispersion of 0.082 nm per CCD pixel. The CCD camera (Andor, 1024 by 256 pixels) is cooled to −60 °C with a Peltier system. Aerosol droplets are generated by an ultrasonic nebulizer (Aerosonic) and flow into a custom-fabricated cell, mounted above the microscope objective, where they can be captured by the optical trap. As pure water droplets are stable only in oversaturated environments we make use of NaCl-doped droplets, which acts to decrease the vapour pressure of the droplets and enable
Figure 1. Schematic of the experimental setup. A 532 nm laser beam forms an optical tweezers system being focused through the microscope objective on a commercial Nikon TE2000 microscope. A droplet is trapped in a plastic trapping chamber, which replaces the conventional microscope slide sample in normal optical tweezers. An image of the droplet is formed on the slit of the spectrograph through a dichroic beam splitter, allowing Raman spectral information to be gathered from the trapped droplet.

stable formation in environments with RH lower than 100%. We use solutions with 50 g l\(^{-1}\) of NaCl.

The droplet’s radius and refractive index can be determined through examination of the wavelength-resolved resonant structure apparent in the OH-stretching Raman band of water \([14]\). Thus, the experimental frequency comb can be overlaid with a WGR fingerprint, which allows the measurement of the size and the refractive index of the droplet with nanometre and 10\(^{-4}\) accuracy, respectively.

3. Observations and measurements

3.1. Experimental observations

In figure 2, the OTP is alternatively increased and decreased inducing decrease and increase, respectively, of the measured whispering gallery optical path of a \(\sim 6 \mu m\) droplet at 650 nm. Although power increases induced a continuous blue drift of the Raman fingerprint with a characteristic response time of the order of 1 s, optical path increases were observed to be much quicker and ‘quantized’ to values commensurate with WGRs at the illuminating wavelength of 532 nm. The resonance stays stably locked over long periods of time (>10 min) at constant trapping power. Significant changes in RH would be required for the lock to become unstable. Figure 2 also indicates the calculated size parameters resonant with \(n = 1\) WGRs at the trapping wavelength, demonstrating an obvious correlation between the theoretically predicted resonant size and the experimentally determined ‘quantized’ sizes occurring during droplet growth, confirming a double resonance effect \([10]\).

The comparison between measured and calculated resonant sizes assumes that the droplet maintains a spherical shape allowing simple treatment by generalized Lorenz–Mie theory (GLMT) \([15, 16]\). Indeed, the average radiation pressure on the surface of the droplet is \(\delta p = I/c \sim 1\) Pa (on and off resonances), where \(I = P/(4\pi a^2)\) is the mean intensity on the surface of the droplet (\(P\) being the trapping power and \(a\) the droplet’s radius). This is much

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Figure 2. The variation in the WGR optical path converted into size parameter \( nka \) at 532 nm (bottom panel) taking dispersion in refractive index into account [19]. The measurements are presented as a function of time during which the OTP (measured at the back aperture of the microscope objective) is successively decreased and increased in the range between 50 and 20 mW (top panel). Spectra are taken every 10 s after changing the power. The optical path jumps observed during growth correspond to droplet sizes resonant with \( n = 1 \) TE (red solid lines) and TM (green dashed lines) WGR at 532 nm. The droplet size can remain resonant with TE resonances as robustly as TM resonances (although tends to favour TM resonances). The slow drift is due to non-constant RH in the trapping cell. The plateaus indicated relate to the plot shown in figure 3.

smaller than the capillarity-induced difference of pressure maintaining spherical shape, \( \Delta p = 2\gamma/a \sim 4 \times 10^4 \) Pa. The estimated eccentricity of the droplet [17] is then expected to be of the order of \( \delta a = 8a\delta p/\Delta p \sim 2 \) nm, possibly inducing a slight spectral shift of the resonance frequency comb but validating the use of GLMT for our experimental results [18]. Moreover, thermocapillary forces are expected to be of the same order of magnitude but to induce a deformation into an oblate ellipsoid.

3.2. Radius and refractive index determination

The refractive index was estimated from the Raman fingerprint around 650 nm. The \( 10^{-4} \) accuracy on this measurement relies on the strong dependence of the asymmetry between the
wavelengths of consecutive TE- and TM-polarized resonances with the refractive index \([19]\). The NaCl concentration was thus deduced with a 1% accuracy using an experimental fitting formula \([20]\). The refractive index at 532 nm can be numerically calculated with this same formula yielding the same \(10^{-4}\) accuracy. Moreover, a 1 in \(10^{-5}\) measurement of the product \(nka\) can be obtained by tracking Raman peaks \((n\) being the refractive index, \(k\) the wavenumber and \(a\) the radius of the droplet). An identical precision is obtained for the product \(nka\) at 532 nm as the 1% relative uncertainty on the salt concentration yields a \(2 \times 10^{-6}\) relative uncertainty on the product \(nka\) at 532 nm. This weak effect is due to the fact that the value of the dispersion coefficient is chiefly governed by water and only weakly depends on the salt concentration. Finally, in GLMT \([19]\), the product \(nka\) varies as \(\varepsilon/L\) where \(\varepsilon\) is the uncertainty on the refractive index and \(L\) the orbital angular momentum of the resonance. A \(10^{-4}\) accuracy on the refractive index is then sufficient to estimate the theoretical value of the resonant size parameters \((nka)_{\text{res}}\) with \(10^{-5}\) accuracy. Relevant and accurate comparison of experimentally determined size parameters with numerically calculated WGRs at 532 nm can then be performed.

The largest increase induced in the cavity size parameter, a change in mode number from 51 to 58 at the laser wavelength on reduction of trapping power, can be estimated to correspond to a decrease in the non-resonant heating by between 50 and 100 mK \([12]\). This decrease in non-resonant heating leads to condensational growth, which leads to the discrete steps in cavity size as the droplet sequentially tunes into resonance with modes of consecutive mode number. In addition, a slow drift in size is observed with time due to unsatisfactory long-term control of (and here increasing) RH in the trapping cell.

3.3. Analysis of the Raman fingerprint

The dominant mode polarization in the Raman signature is observed to match the polarization of the mode resonant with the illuminating wavelength as illustrated in figure 3. During the locked periods, we observed an increase of the resonant Raman peaks with decreasing OTP during a period at which the wavelengths are locked. As can be seen in figure 3, modes of polarization matching the resonance at 532 nm are observed to grow most significantly in stimulated Raman intensity. The increase factor in stimulated intensity was observed to be of the order of 4 while the total trapping power was diminished by a factor 1.5. This evolution was observed until the mode pattern hops to the next size parameter resonant with the 532 nm illumination. These observations suggest that the droplet continues to tune closer to the resonance condition as the trapping power is decreased.

A precise numerical analysis of the Raman resonance wavelengths allows the size parameter of the trapped droplet during the locked regimes to be determined with subpixel precision. In the cavity formed by the droplet, in the non-depleted pump approximation, the stimulated Raman scattering is expected to vary exponentially with the pumping intensity \([21]\). In figure 4, we plot the logarithm of the amplitude of a stimulated Raman peak as a function of the radius. Experimental data follow the Lorentzian profile of the WGR with the radius of the droplet. In these experiments, the stimulated Raman signals (SRSs) are due to both the contribution of resonating and non-resonating pumping intensities.

Comparison of the measured resonant radius with the theoretically determined resonant line centre suggests that the Raman size estimates are slightly smaller (by \(\sim 0.75\) nm). Although the mechanism described above leads to a locked size that remains to the ‘blue’ side of the resonance, this discrepancy is not consistent with calibration uncertainties (better than 0.25 nm...
Figure 3. Comparisons of Raman scattering amplitudes (RSA) recorded at the beginning and the ends of plateaus in figure 1 between times 320 and 400 s (top panel, radius locked on a TM54 resonance) and 590–680 s (bottom panel, radius locked on a TE55 resonance). Red dashed curves correspond to high tweezing powers for which the droplet’s radius is on the edge of a resonance. Blue solid curves correspond to low tweezing power for which the spontaneous scattering is lower and the droplet radius close to the top of the $n = 1$ WGR at 532 nm, giving efficient stimulated Raman Stokes-conversion. Raman maximum peaks are observed to match the resonating polarization at 532 nm.

on the radius of the droplet). However, while numerical simulations were performed for an assumed 532.000 nm tweezing laser, perfect superposition would be obtained for a value of 531.984 nm. We did not have any metrological way to measure the line of our laser with such a precision but this value appears to be consistent with the spectral width of Nd$^{3+}$ ions fluorescence.

4. Modelling of the locked regime of the droplet

We now consider the modelling of the locked regime of the droplets. The droplet being at all times in thermodynamic equilibrium with its vapour, its size is a stationary state and is the end-result of a balance between heating by the laser and cooling by thermal diffusion to the surrounding gas phase. In previous work, we have shown that a trapped droplet is a few tens of milli-Kelvins above the ambient temperature [12] and that when out of resonance, the absorption efficiency $Q_{\text{abs}}$ can be approximated by $2\alpha$ in good agreement with the experiment (where $\alpha$ is the absorption coefficient of water at 532 nm). On resonance, we now shall refer to the definition of the quality factor: $Q = \omega E / P_{\text{diss}} = \omega \tau$ where $E$ is the stored energy in the microcavity and $P_{\text{diss}}$ is the dissipated power. Rigorously, the quality factor is the harmonic mean of all losses: bend losses, absorption and surface scattering. At first approximation, we will assume the quality factor is dominated by intrinsic microcavity losses and that absorption and scattering are negligible in this regard. In this approximation, on resonance, the dissipated power is equal to the coupled impinging power in the resonating mode. Let $f$ be the fraction of...
Figure 4. A close inspection of the drifting of WGR allows a subpixel precision measurement of the droplet’s radius. In the label ‘a₀’ would correspond to the resonating radius for a 532.000 nm tweezing wavelength. The logarithm of the amplitude of the SRS of a TM40 peak (@650 nm) is shown (blue squares), which is proportional to the gain and thus follows the internal optical energy (on + off resonance). As the droplet gets closer to the peak of the resonance, the gain increases. The droplet is locked on a TM49 resonance at 532 nm but centred at a marginally smaller droplet size than the theoretical prediction (green curve). The Lorentzian fit of the experimental points (red line) gives a Q factor equal to 6.8 × 10³. The theoretical calculation gives 2.7 × 10⁴.

tweezing power coupled in the given resonating mode. The heating due to the stored energy can be approximated as αcE as high-Q mode volumes are almost completely contained inside the droplet. The contribution of the resonance to the absorption efficiency is then: \( fQ\alpha/k \), where \( k \) is the wavenumber at 532 nm in water. Thus, on resonance, the steady state elevation of the droplet above the room temperature can be estimated from the relation:

\[
\Delta T = \frac{PQ_{\text{abs}}}{4\pi K_a a} = \frac{\alpha P}{2\pi K_a a} \left( 1 + \frac{fQ}{2ka} \right),
\]

where \( P \) is the laser trapping power, \( a \) is the droplet radius and \( K_a \) is the thermal conductivity of the surrounding air (26.2 × 10⁻³ W m⁻¹ K⁻¹ at 300 K). The first term determines the non-resonant heating and the second term the resonant contribution.

During locked periods in figure 1, despite the significant decrease of trapping power, the droplet’s radius remains constant to within a fractional change \( \delta a/a \sim 10^{-4} \) resulting in an almost constant salt concentration. When the size is non-resonant, we can deduce that such a change in radius would correspond to a surface temperature change of the order of a few micro-Kelvins. Thus, from a thermodynamic point of view, the change in surface temperature of the droplet is
negligible compared with the decrease of trapping laser power and the locked regime is then characterized by marginally varying salt concentration, vapour pressure and temperature. The locked regime can be considered as an ‘isotherm’ curve, exhibiting the balance between bulk non-resonant heating—proportional to spontaneous Raman amplitude (figure 3)—and radius fine tuning of WGR resonant heating—varying linearly with the amplitude of the stimulated Raman peaks. While approaching resonance, the droplet is first off-resonance and finishes exactly on resonance, with a marginally varying temperature. The ratio of temperature between the beginning and the end of the locked regime is then equal to 1 during a plateau:

$$1 = \frac{P_{\text{locked}}}{P_{\text{unlocked}}} \left(1 + \frac{fQ}{2ka}\right).$$

For the locked regime of figure 4, the experimental ratio between optical powers at the beginning (off-resonance) and the end (on-resonance) of a locked period is equal to $\frac{P_{\text{unlocked}}}{P_{\text{locked}}} = 2.74$. This means that, assuming the tweezers geometry and the trapping height do not change significantly with laser power, resonant heating accounts for only 1.74 times non-resonant bulk heating.

In addition, in figure 4, we measured a quality factor equal to $6.8 \times 10^3$, four times as small as the theoretical value of $2.74 \times 10^4$. The difference cannot be due to an underestimate of the absorption as the imaginary part of the refractive index must reach $n'' = 4 \times 10^{-5}$ ($10^4$ as large as the theoretical value) to be consistent with such a marked reduction in $Q$. The full modelling of losses should take into account Rayleigh scattering by water and surface pollutants as well as the specified 30 GHz of spectral bandwidth of the Ventus laser we use, corresponding to a $1.9 \times 10^4$ $Q$-factor, unfortunately very close to the $Q$-factor of the droplets we study here.

The small effect of resonant, when compared with non-resonant, heating can be explained by the fact that the coupled-in power fraction $f$ is small even though the quality factor of the resonance is large (theoretically $2.74 \times 10^4$). From our experimental result, we can directly deduce the products $fQ = 144$. Our numerical simulations based on rigorous modelling of aberrated tightly focused beams [16] suggest $f$ is of the order of $3 \times 10^{-3}$ for the WGR with the $n = 49$ orbital momentum considered here. When combined with the value of the quality factor estimated from the Lorentzian fit in figure 3, $6.8 \times 10^3$, we expect the product $fQ$ to be smaller than 20. Thus, our numerical simulations underestimate the product $fQ$ by a factor 7, clearly not accounting for aberrations introduced by the optics which are expected to increase the coupled in power fraction $f$. Moreover, if the quality factor is altered by Rayleigh scattering, coupling between Mie modes should also be increased and the power coupled within the resonating mode is also expected to be larger.

5. Conclusion

We have reported observations of the locking of optically tweezed salt-water droplets to WGR resonant with the wavelength of the trapping laser. At constant RH, the increase of trapping power results in an increase in temperature. As a consequence, the droplet evaporates, increasing its salt concentration. When decreasing the trapping power, the droplet grows until it reaches a resonance onto which the loss of bulk heating is compensated by resonant heating. This stationary state can only be stable on the blue-detuned side of WGR (i.e. during the growth of the droplet). During a locked phase, the droplet grows marginally in size, slowly tuning into resonance with the incident field. In the fingerprint of stimulated Raman scattering, the locking of the droplet size and refractive index leads to prolonged stability of the observed WGR.
wavelengths. Some more precise experimental measurements might be of interest in the future for rigorous comparison with numerical simulations. In particular, the precise knowledge of the laser line, a narrower linewidth of the trapping laser and the measurement of the relative droplet height in the beam might be of interest to optimize the coupling with the high-$Q$ resonances of the droplet.

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References

[1] Le T, Savchenkov A, Yu N, Maleki L and Steier W H 2009 Optical resonant sensors: a method to reduce the effect of thermal drift Appl. Opt. 48 458–63
[2] Armani A M, Kulkarni R P, Fraser S E, Flagan R C and Vahala K J 2007 Label-free, single-molecule detection with optical microcavities Science 317 783–7
[3] Spillane S M, Kippenberg T J and Vahala K J 2002 Ultralow-threshold Raman laser using a spherical dielectric microcavity Nature 415 621–3
[4] Tzeng H M, Wall K F, Long M B and Chang R K 1984 Evaporation and condensation rates of liquid droplets deduced from structure resonances in the fluorescence-spectra Opt. Lett. 9 273–5
[5] Del’Haye P, Schliesser A, Arcizet O, Wilken T, Holzwarth R and Kippenberg T J 2007 Optical frequency comb generation from a monolithic microresonator Nature 450 1214–7
[6] Verwooy D W, Ilchenko V S, Mabuchi H, Streed E W and Kimble H J 1998 High-$Q$ measurements of fused-silica microspheres in the near infrared Opt. Lett. 23 247–9
[7] Chen G, Chang R K, Hill S C and Barber P W 1991 Frequency splitting of degenerate spherical cavity modes—stimulated Raman-scattering spectrum of deformed droplets Opt. Lett. 16 1269–71
[8] Lin H B, Huston A L, Eversole J D and Campillo A J 1990 Double-resonance stimulated Raman-scattering in micrometer-sized droplets J. Opt. Soc. Am. B-Opt. Phys. 7 2079–89
[9] Qian S X and Chang R K 1986 Multiorder stokes emission from micrometer-size droplets Phys. Rev. Lett. 56 926–9
[10] Arnold S, Leung K M and Pluchino A 1986 Optical bistability of an aerosol-particle Opt. Lett. 11 800–2
[11] Kiraz A, Karadag Y and Coskun A F 2008 Spectral tuning of liquid microdroplets standing on a superhydrophobic surface using electrowetting Appl. Phys. Lett. 92 191104
[12] Knox K J and Reid J P 2008 Ultrasensitive absorption spectroscopy of optically-trapped aerosol droplets J. Phys. Chem. A 112 10439–41
[13] Miles R E H, Guillon M, Mitchel L, McGlone D and Reid J P 2009 The influence of resonant absorption and heating on the equilibrium size of aqueous-solute aerosol droplets Phys. Chem. Chem. Phys. 11 7312–7
[14] Hopkins R J, Mitchel L, Ward A D and Reid J P 2004 Control and characterisation of a single aerosol droplet in a single-beam gradient-force optical trap Phys. Chem. Chem. Phys. 6 4924–7
[15] Lock J A and Gouesbet G 2009 Generalized Lorenz–Mie theory and applications J. Quantum Spectrosc. Radiat. Transfer 110 800–7
[16] Viana N B, Rocha M S, Mesquita O N, Mazolli A, Neto P A M and Nussenzveig H M 2007 Towards absolute calibration of optical tweezers Phys. Rev. E 75
[17] Lamb H 1945 Hydrodynamics (Dover, NY: Cambridge Mathematical Library)
[18] Lai H M, Leung P T, Young K, Barber P W and Hill S C 1990 Time-independent perturbation for leaking electromagnetic modes in open systems with application to resonances in microdroplets Phys. Rev. A 41 5187–98
[19] Lam C C, Young P T and Young K 1992 Explicit asymptotic formulas for the positions, widths, and strengths of resonances in Mie scattering J. Opt. Soc. Am. 9 1585–92
[20] Millard R C and Seaver G 1990 An index of refraction algorithm for seawater over temperature, pressure, salinity, density, and wavelength Deep-Sea Res. A 37 1909–26
[21] Campillo A J, Eversole J D and Lin H B 1996 Optical Processes in Microcavities (Singapore: World Scientific)