

Report STM: Chemical Detection with Liquid Microresonators

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1. Introduction

The demand for ultra-compact and inexpensive sensors for applications such as process monitoring, air quality monitoring and bio-medical analysis has steadily increased in the last decade. However, existing sensors often do not offer the needed sensitivity or do not have the required chemical specificity. In this project the application of liquid microresonators as chemical sensors has been studied, since they have the potential to overcome these drawbacks.

Optical cavities formed by silica microsphere resonators or silica toroids are known to be high finesse cavities (Q-factor $> 10^9$). The optical spectrum of microresonators is very sensitive to refractive index and light absorption by the surrounding environment and the resonator material itself. This makes them particularly interesting for chemical sensing applications of very small volumes. The problem with solid microresonators is that the main part of the light is contained inside the solid resonator and only the evanescent wave part interacts with the sample. We therefore decided to use a liquid droplet as resonator allowing us to have the analyte inside the resonator. In this way the main part of the mode interacts with the sample and it is expected that a very high sensitivity to optical absorption can be achieved and possibly even refractive index changes can be monitored.

2. Experiment and Theory

2.1 Theory

Whispering gallery modes can be thought as rays of light that are guided inside the circular, toroidal or spherical resonator along the circumference by total internal reflection. These modes can be observed either in the light scattered from the sphere, where the resonances appear as positive peaks in the spectrum, or in the transmitted light, where negative peaks indicate light missing due to coupling to the sphere. These modes are known to have low losses, e.g. the light circulates for many round-trips before it gets absorbed, scattered or transmitted. If an absorbing analyte is present, the losses increase resulting in a shorter photon lifetime.

In cavity ring-down spectroscopy this photon lifetime or ring-down time is measured by filling the optical cavity with light, switching off the laser and monitoring the intensity decay inside the cavity. The ring-down time of a microresonator with a circumference length L and round trip time t_{RT} is given by:

$$\tau = \frac{t_{RT}}{\text{losses}} = \frac{nL}{c(\text{losses}_{\text{scatter}} + \alpha_{\text{drop}}L + C\varepsilon L)} \quad (1)$$

The term $\text{losses}_{\text{scatter}}$ describes the losses per roundtrip due to scattering, α_{drop} is the absorption coefficient of the droplet material, C is the concentration of the sample, ε is

the molar extinction coefficient of the sample, c is the vacuum speed of light and n is the effective refractive index of the propagating mode.

Another way of measuring ring-down times is phase-shift CRD spectroscopy. There the intensity of the laser is modulated sinusoidally and the cavity emits light that is phase-shifted due to the lifetime of the photons in the cavity, i.e. the ring-down time. The ring-down time is related to the phase-shift $\Delta\phi$ between the light entering and exiting the cavity and modulation frequency ω by the following equation:

$$\tan(\Delta\phi) = -\omega\tau \quad (2)$$

This method has the advantage that short ring-down times, as usually observed for microcavities, can be compensated by increasing the modulation frequency ω .

2.2 Experiment

Paraffin oil was chosen as material for the droplet because it has a low vapour pressure and it has a low absorption at the laser wavelength of 1560 nm. The droplet has a diameter of about 1 mm and it is suspended at a tip of a silica fiber. The light is coupled into the microresonator by focusing a laser beam with an objective lens to a very small diameter so that the beam is touching the equator of the droplet. This allows some of the light to couple into the whispering gallery modes. Behind the droplet a second objective lens collects the transmitted light and a lens focuses it on the detector. A third objective lens collects the light scattered from the resonator and a lens focuses it on a second detector. A half-wave plate allows rotating the polarization to excite either TE or TM modes. The setup is depicted in Figure 1.

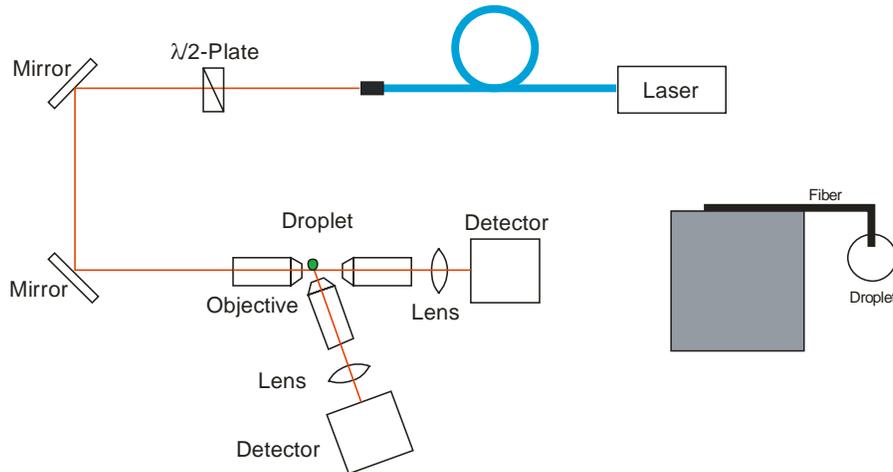


Figure 1: Setup of the experiment. The laser beam is tightly focused next to the droplet by an objective lens. Two mirrors allow aligning the beam and a $\lambda/2$ -plate gives control over the polarization. Objective lenses were used to collect the transmitted and the scattered light.

If the droplet was a perfect sphere, the modes would only appear at wavelengths for which the circumference of the droplet is an integer multiple of the wavelength. But due to gravity the droplet is slightly deformed which lifts the degeneracy and higher order modes appear at other wavelengths. The separation of formerly degenerate modes can then be used to determine the eccentricity of the droplet. Also note that TE and TM modes have different splitting (see Figure 2a).

To perform phase-shift cavity ring-down spectroscopy the laser needs to stay at the wavelength of the resonance. Therefore the laser wavelength was locked on resonance by using the Pound-Drever-Hall (PDH) technique. In the PDH method the deviation of the laser wavelength from centre of the resonance is measured and this information is fed back into the laser to correct the wavelength. This is done by varying the laser wavelength sinusoidally at radio frequencies. The light intensity will then also vary with an amplitude and phase depending on the wavelength deviation. The obtained signal has the shape of the derivative of the resonance (see Figure 2b). This signal can be used as an error signal for a servo loop, which then outputs a correction signal to suppress fluctuations of the laser wavelength relative to the center of the resonance.

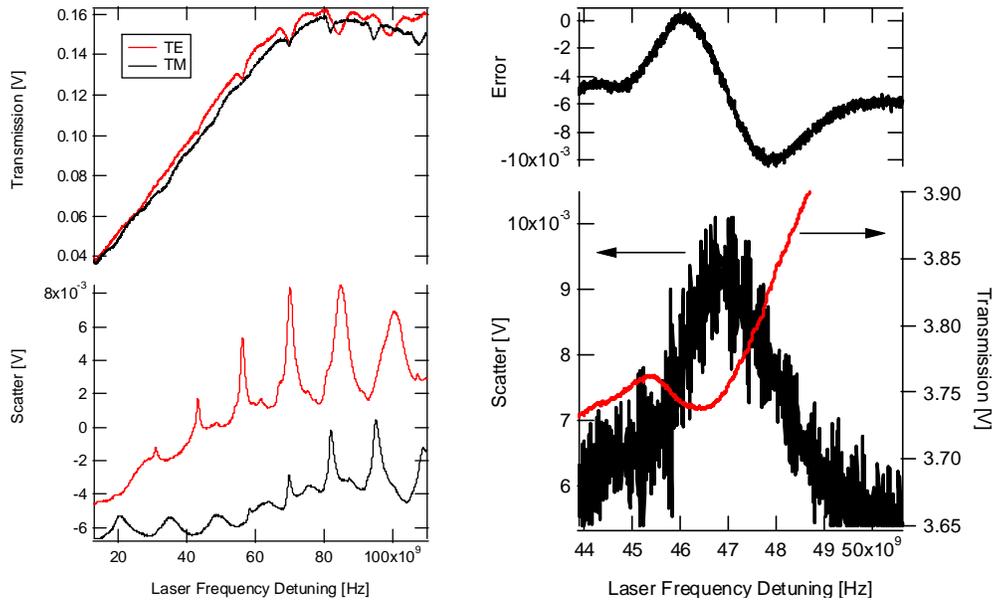


Figure 2: a) Transmission and scatter signal of the droplet. TE and TM modes have different splitting due to the eccentricity of the droplet deviating from a perfect sphere. b) Error signal generated by the scatter signal. This error signal allows the servo loop to give a feed-back to the laser current and therefore lock the laser to the center of the resonance.

To perform phase-shift cavity ring-down spectroscopy the laser intensity needs to be also intensity-modulated for measuring the phase-shift introduced by the resonator. In a first test the laser intensity was modulated by current modulation through the same bias-T as the modulation for the PDH-locking process, but the results were not satisfying. The phase-shift exhibited an offset and negative ring-down times were obtained for the scatter signal. A possible explanation is that an additional phase-shift was introduced by the current modulation, which results in wrong read-outs of the phase measurement. Therefore a fiber-coupled Mach-Zehnder modulator was implemented as external

modulator between the laser and the output collimator. No offset was observed anymore and a ring-down time of 0.87 ns was obtained by measuring the phase-shift of the scatter signal as a function of the intensity-modulation frequency (see Figure 3).

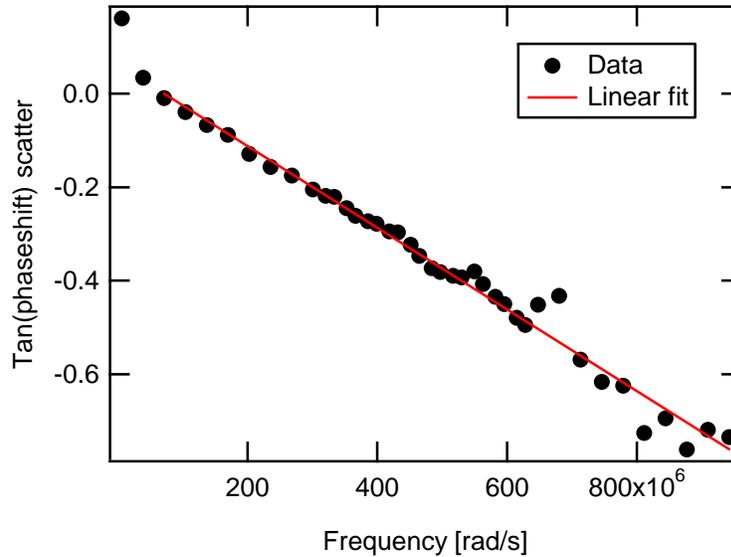


Figure 3: Tangent of the phase-shift versus modulation frequency. The ring-down time is 0.87 ns, which is in good agreement with the Q-factor obtained from the width of the resonance.

To perform chemical sensing with the microresonator, a sample needs to be introduced into the droplet. There are several ways of applying the analyte to the droplet, for example a small amount of it can be added to the pure droplet, or the pure droplet can be replaced by another droplet containing the analyte. The latter method has the advantage that the concentration in the droplet is known, whereas in the first case the concentration needs to be determined from the droplets volumes. One problem of both methods is that the size of the droplet changes from sample to sample, which results in a variation in ring-down time.

To examine the influence of the droplet size on the ring-down time, we measured the ring-down time of five droplets and, additionally, a small amount of paraffin oil was added to two droplets. The obtained ring-down times are all within a narrow range, the variations were smaller than 10% (see Figure 4). The larger droplets have a longer ringdown time because of the longer round-trip time. If an absorbing sample was introduced with a comparable high concentration, large changes in ring-down time could be expected, which should be clearly distinguishable from droplet to droplet variations.

Samples of cyclopentanamine in paraffin oil were prepared with a concentration of 10%. Unfortunately, the vapour pressure of cyclopentanamine is rather high and it evaporated quickly once a droplet was placed in the setup. It was possible to lock the laser to a resonance, but the wavelength position of the resonance shifted quickly due to the size change originating from the evaporation. The PDH-lock caused the laser current to follow the resonance decreasing laser current until it was zero. Therefore ring-down time measurements were not feasible with this substance.

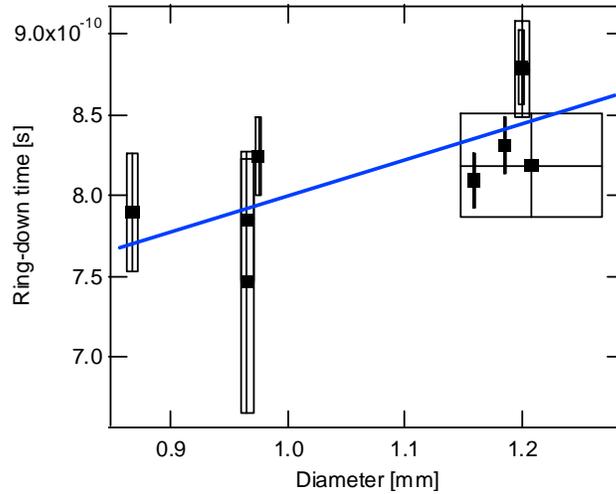


Figure 4: Ring-down times versus droplet diameter. As expected the ring-down time increases with increasing droplet size due to longer round-trip times.

3. Summary and Outlook

We demonstrated that it is possible to use a liquid droplet as a microresonator and to couple light into the whispering gallery modes by a free-space coupling scheme. We locked the laser wavelength to one of the resonances by using the Pound-Drever-Hall technique. With the laser locked on resonance, we performed phase-shift cavity ring-down spectroscopy by modulating the laser intensity with an external modulator. A ring-down time of 0.87 ns was obtained for a droplet of pure paraffin oil and the relationship between ring-down time and droplets size was studied. First attempts of concentration measurements were made, but the absorbing compound had a low vapor pressure and evaporates too fast for good measurements.

A next step will be to find compounds with a high vapor pressure and with a strong absorption feature at the required wavelength of 1560 nm. Also for the liquid droplet itself, more suitable materials can be found. A material that is a better solvent for a larger variety of compounds would be advantageous and an even lower absorption coefficient than that of paraffin oil would be desirable. The sample application method also could be improved.

Date

Signature
