Trace Gas Detection Utilizing Optical Spectroscopy of Microresonant Cavities

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Trace gas detection is attempted by using spherical and disk-shaped microresonant optical cavities. Several different experimental configurations were examined: waveguide and disk, waveguide and sphere, and etched fiber and sphere. The etched fiber and sphere setup proved to be the simplest to align and couple significant amounts of input radiation into the resonant modes of the cavity. A 300 micron sphere coupled to a tapered bare optical fiber created a microresonant mode with a free-spectral range of 1.25nm, a bandwidth of 700 MHz, and an optical Q factor of 3x10⁴. Cavity ringdown measurements were also attempted near 1516.3nm.

1. INTRODUCTION

For ultra-sensitive detection of trace gas species, cavity ringdown spectroscopy (CRDS) has been previously shown to be a compact method for enhancing molecular absorption techniques. Through the use of a resonant cavity, absorption path lengths can be increased from the physical size of the resonator (usually the order of 1m for meso-scale CRDS) to several kilometers. Through the selection of appropriate absorption transitions, this method can allow sub-ppm detection of trace species with a wide dynamic range (~ 10⁶). Cavity ringdown spectroscopy was developed as an enhancement of cavity attenuated phase shift technique for the characterization of mirror reflectivities, using pulsed lasers to reduce the dependence on cavity mode matching [O’Keefe and Deacon, 1988].

CRDS offers great enhancement of the absorption optical path length with minimal coupling to the cavity mode resonances. However, due to the small fraction of energy storage in the cavity (and thus leaking out per cavity pass), high power lasers are required to detect the ring down signal. As long as the pulse duration of injected light is shorter than the round-trip time within the cavity, and the laser linewidth is smaller than the absorption features of interest, pulsed CRDS is a straightforward and very sensitive technique for obtaining the absorption spectrum of dilute or weakly absorbing gas-phase species [Zalicki et al., 1998], particulate matter [Sappey et al., 1998], and acetylene using femtosecond lasers [Gherman and Romanini, 1998]. Pulsed CRD offers high sensitivity over a potentially wide spectral range by use of a broadly tunable pulsed laser and a relatively simple setup [Scherer et al., 1997].

However, the use of pulsed lasers also imposes some limitations on the meso-scale CRDS technique. The intensity of the light coupled into and out of the cavity is small, as a consequence of reduced spectral overlap between cavity modes and laser linewidth as well as a lack of significant light buildup within the cavity. Moreover, for most practical pulsed laser systems, interference effects within the ring-down cavity preclude the use of simple models to describe the decay of light within the cavity as has been extensively demonstrated. In addition, the spectral resolution of pulsed CRDS is limited by the bandwidth of the pulsed laser (~typically 0.2 cm⁻¹), which is often too large for direct recording of high-resolution spectra [Nakagawa et al., 1994]. Finally, the relatively high cost of typical pulsed tunable laser systems in the infrared spectrum often prohibits the use of pulsed CRD for many industrial applications.

With the development of narrow bandwidth diode lasers in the near infrared (NIR) spectrum due to advancements in optical communication, trace gas species with infrared active transitions is now common using continuous wave (CW) laser sources [Romanini et al., 1997]. For typical CW cavity ringdown spectroscopy, two mirrors (usually R > 99.9%) are used to form a stable optical resonator (linear or ring resonators are common) with a discrete set of resonance frequencies (Figure 1). To couple a narrow linewidth laser effectively into the cavity, the laser wavelength must overlap with a cavity resonance

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The absorption coefficient can be calculated from the difference between the two measured ringdown times.

Typically, a threshold circuit coupled with the optical detector is used to detect sufficiently strong build-up events and trigger an acousto-optical modulator (AOM), which serves to deflect the laser beam and decouple it from the resonant cavity. After the beam is deflected, the light intensity in the cavity decays exponentially (known as the cavity ringdown) owing to fixed roundtrip losses due to mirror reflectivity, Rayleigh and Mie scattering in the measurement volume, and optical absorption from the test gas. The decay time can be expressed as:

\[
\frac{1}{\tau} = \frac{T + R + k_v(L)}{L} \tag{1}
\]

where \(\tau\) is the decay time constant, \(L\) is the cavity length, \(T\) is the propagation loss, \(R\) is the mirror reflection loss, \(k_v\) is the sample absorption coefficient, and \(c\) is the speed of light. The ringdown decay time without the presence of the absorbing sample is measured independently \((\tau_0)\) and thus the sample absorption coefficient can be calculated from difference between the two measured ringdown times.

\[
k_v = \frac{1}{c} \left( \frac{1}{\tau} - \frac{1}{\tau_0} \right) \tag{2}
\]

The measurement sensitivity depends on the rms shot-to-shot noise \((\sigma/\tau)\) of ringdown decay time constant \(\tau\) and on the equivalent path length \((c\tau_0)\) in the optical resonator.

Significant advances in chemical sensing using CRDS have been developed which build upon the CW excitation technique. Tosching et al., (2000) made sensitive measurements for gas phase solvant detection simultaneous with water concentration measurements. Hallock et al., (2003) proved the used of CW-CRDS for liquid samples using broadband diode lasers with the cavity selection the resonating mode and thus creating a narrowband laser filter, which is an ideal scheme for broadband absorbers. Paldus et al., (1998) employed a single-beam, dual-resonant approach to CRDS to continuously lock a narrow-band external-cavity diode laser to a high-finesse resonator with one polarization, while performing CRDS on the locked system using a second, orthogonally polarized arm. This enabled shot noise limited detection of the cavity ringdown time while the ring cavity geometry eliminates dangerous back reflection into the CW laser system which reduces laser intensity noise and mode hop issues. Berden et al., (1999) measured trace ammonia concentration in a low speed jet showing application in fluid flows. Engeln et al., (1998) used a swept laser technique with random cavity mode oscillations to record high-speed, narrowband absorption spectra within one cavity free spectral range. Finally, Kosterev et al., (2001) extended CW CRDS measurements into the mid-IR using a quantum cascade laser to measure nitric oxide concentration at the fundamental vibrational mode (5.2 \(\mu\)m).
WGM resonators utilize total internal reflection of evanescently coupled light to create extremely high Q cavities with reduced round trip losses. This resonator geometry and energy storage capability make WGMs the idea nanoscale building block for a novel cavity ringdown spectrometer with high sensitivity and ultra wide tunability (as total internal reflection is a broadband phenomenon which does not rely on a specialized reflective coating).

While advances in CRDS systems have increased rapidly, the systems are still bulky laboratory-scale devices which are neither portable nor field deployable, in general. Additionally, specialized high reflectivity mirrors are required for the resonant cavity, usually with limited spectral range (~50 nm). With the development of nanofabrication technologies and microscale optical system, entirely new areas of CRDS exploration and development are possible using microresonant cavities based on whispering galley mode (WGM) resonators. WGM resonators utilize total internal reflection of evanescently coupled light to create extremely high Q cavities with reduced round trip losses (Figure 2). This resonator geometry and energy storage capability make WGMs the idea nanoscale building block for a novel cavity ringdown spectrometer with high sensitivity and ultra wide tunability (as total internal reflection is a broadband phenomenon which does not rely on a specialized reflective coating).

This study looks to examine the feasibility of building and testing microresonant cavities for CRDS detection of trace gas species. For this type of investigation, the challenged lie mostly in the optical engineering and alignment issues associated with building an appropriate resonant cavity system. We explore many of these challenges while progressing towards producing resonant modes in a tapered fiber-microsphere geometry. This setup is then tested to see if CRDS is feasible in these microscale geometries.

II. EXPERIMENTAL SETUP

Many different configurations have been examined for proof-of-concept experiments for micro-scale cavity ringdown experiments. Optical whispering gallery mode resonances (WGM) have been explored extensively in the physics and fiber optic communities for the past 15 years. As a result, there have been many successful configurations, each with their own manufacture and alignment difficulties. Each of the setups examined externally used the setup shown in Figure 3. Though the elements within the shaded box (i.e. waveguides, tapered fibers, etc.) may
change, the external detectors, lasers, and optics were constant.

The first configuration attempted for this study was a combined waveguide-disk WGM resonator. This system had a 1 x 3 micron waveguide on a silicon substrate coupled to a 30 micron disk. The chip had 12 different resonator waveguide combinations with varying gap widths (from 100 nm to 1000 nm) to explore the effects to gap width on resonator performance. A high resolution SEM of this device is shown in Figure 4. While this setup was ideal in that the waveguide and resonator were already coupled together, this design actually introduced several very difficult optical engineering challenges in order to couple and de-couple light from the waveguide. Typically optical fibers are carefully aligned to the edges of waveguide to pitch the light from one to the other. However, with SM fiber utilizing at minimum a 6 micron core fiber, the geometrical mismatch coupled with the 3-axis nm precision required for alignment made coupling light into this waveguide an extreme challenge. Many methods were explored: using taper fibers, lensed fibers, index matching fluids, and physical contact. Unfortunately, these methods did not achieve significant amounts of light coupled into the waveguide. Much more sophisticated methods such as grating couplers, tapered waveguide elements, and micro-Bragg mirrors can be employed to improve this coupling; however, these solutions are all very costly from a fabrication standpoint and did not add substantively to the goals of this study.

The second attempted experimental setup involved a specially fabricated polymer waveguide with a 40 x 40 micron structure (shown schematically in Figure 5). This waveguide was then easier to couple light into as typical multimode fiber (50 micron diameter core size) could be used with minimal alignment difficulties. This setup achieved microwatt laser power levels from a 10 W, 1064 nm source. However, these waveguides did not have integral disk resonators placed on-chip, as a result of the lower resolution manufacturing process for polymer waveguides. Thus, a microsphere approach was employed as has been very common in the WGM literature. The microsphere is then suspended above the waveguide with a micron sized gap to induce the resonances in the meridional plane of the sphere. This configuration has been successful previously using SPARROW type waveguides coupling to microspheres. However, since the size of the polymer waveguide is rather large, it operates as a multi-mode waveguide. This multi-mode behavior, in turn, results in very little power coupled into the resonant modes of the sphere. Also, the sphere typical size is only 6-8 times that of the waveguide width, resulting in the excitation of still further polar modes within the sphere. Each of these excited modes has its own bandwidth, FSR, and Q values. With several modes competing for gain, detection of the resonant modes proved too challenging due to low signal levels.

The third experimental setup employed is a more simple tapered fiber, microsphere system (Figure 6). A single-mode fiber is stripped of most of its cladding and extruded to produce a taper down to ~ 1.5 microns. The microsphere is then brought close to the tapered fiber, maintaining a gap distance, to produce the resonant modes. This system is much simpler to control many of the important elements of the
alignment, since the remaining cladding on the fiber acts as an extra gap producing element allowing the sphere to be in directed contact with the fiber. This makes alignment on surface of the sphere much easier, and tends to produced higher Q results for the WGM modes. While the fiber taper/microsphere is not optimal for mass producing sensors based on this concept, it does allow for the basic physics of cavity ringdown in microspheres to be examined. Once a working optical setup has been achieved and light propagates both through the waveguide or tapered fiber and is coupled into the WGM resonator, a cavity ringdown type experiment can be attempted.

III. RESULTS AND DISCUSSION

With a working experimental setup as described above, a set of exploratory experiments was conducted to look at the optical capabilities of the microsphere-tapered fiber configuration for use in cavity ringdown experiments. With the end goal being CRDS experiments using WGM resonators, some basic scaling of the systems must be examined. Several WGM configurations are shown in Table 1 which led toward a WGM-CRDS design. For cavity ringdown experimentation to be effective, a long pathlength must be developed within the resonating element to increase the optical absorption of the target species. Clearly, the sphere resonator is the best geometry currently for the task as well created spheres can easily achieve Q factors in excess of $10^8$, ringdown times which are easily detectable in the micro-second range, and path lengths of several meters. Though a several meter path length device for macro-scale CRDS is not considered a significant enhancement over direct absorption methods, this micro-resonator CRDS system could be packaged in a much smaller device. It is worth noting that current research into nanofabrication techniques will continue to reduce the size of the minimum feature to be produced on the chip surface, enhancing the capability to make more accurate ring and disk formations. This higher accuracy fabrication will increase the best Q factors from these configurations and allow for sphere-type decay lengths to be achieved using all on-chip methods.

So beginning with the microsphere-tapered fiber system, the basic equations were examined to look the proper scaling of dimensions. The free spectral range (FSR) of a optical waveguide sphere is given by:

$$ FSR = \frac{c}{2\pi n R} \quad [1] $$

where $c$ is the speed of light, $n$ is the index of refraction in the media and $R$ is the radius of the sphere. This very simple solution is effective is mostly meridional modes are excited and only TE modes propagate within the sphere. If other polar modes are resonating, the FSR will include polar and azimuthal mode numbers associated with those propagating modes.

<table>
<thead>
<tr>
<th>Table 1 Example WGM configurations and their characteristic values at 1500 nm resonant wavelength and 300 micron sphere</th>
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<tbody>
<tr>
<td>Best rings, disks</td>
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<tr>
<td>Q factor</td>
</tr>
<tr>
<td>Resonant Linewidth</td>
</tr>
<tr>
<td>Decay Time</td>
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<td>Decay Length</td>
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Figure 7: Scattering spectra for WGM resonator sizes of D = 10, 12.5, and 15 microns, respectively [from Quan et al., 2004].

Figure 8: Gap effects on quality factor Q and FWHM [from Quan et al., 2005].

The cavity Q, or optical storage efficiency, is calculated primarily using experimental data. Its value is difficult to determine \textit{a priori} as it requires exact knowledge of the surface geometry of the sphere ($Q_{\text{surf}}$), internal inhomogeneities ($Q_{\text{rad}}$), adsorbed species layers ($Q_{\text{contam}}$), and material absorption ($Q_{\text{mat'}}$). These can be summed together as:

$$Q^{-1} = Q_{\text{rad}}^{-1} + Q_{\text{surf}}^{-1} + Q_{\text{contam}}^{-1} + Q_{\text{mat'}}^{-1}$$  \hspace{1cm} [2]

Most of these variables are very difficult to determine before the sphere is constructed and also very difficult to alter or improve once a particular sphere has been constructed. However, prior research has shown that using fused silica microspheres with near-infrared sources yields a maximum Q factor of $\sim 10^{11}$. Therefore the degradation of this peak cavity Q which occurs for real devices makes the creation of microspheres with cavity Q’s sufficient for CRDS a relatively straightforward task. Numerical simulations of WGM cavities have been performed previously to confirm these scaling laws [Quan et al., 2004 and Quan et al., 2005]. These show the FSR to be a repeatable and predictable value. In addition, it also showed the scaling of the cavity Q factor, WGM linewidth, and the evanescent gap (Figures 7 and 8). As shown in this study as well as others, the quality factor is strongly dependent upon the gap size, with larger gaps promoting larger Q values as well as smaller amounts of energy coupling (as energy transfer depends exponentially on the gap size). This means that there exists and optimum gap size for each type and diameter of WGM resonator/waveguide combination.

Experimental measurements of the spectral width of the resonance mode of the WGM cavity can also be used to determine the cavity Q (Equation 3).

$$Q = \frac{\omega_0}{\Delta \omega} = 2\pi \omega_0 \tau$$  \hspace{1cm} [3]

This formula can also be used to determine the effective ringdown time of the WGM cavity, which is the most important element of a CRDS measurement. Typically, the larger the non-perturbed ringdown time, the more sensitive the CRDS diagnostic. The Q factor can also be measured by the exponential ringdown time of the WGM resonator. However, this method is inferior to the spectral method as the wavelength stability and tuning capability of the external cavity diode lasers used as the excitation source is much better than detecting a weakly decaying signal on the pico-second timescale.

These scaling laws and information from previous studies led to the design of a tapered-fiber/microsphere arrangement that was thought to produce a large enough Q to attempt cavity ringdown measurements. Fused silica microspheres were created from bare single-mode fiber by melting the fiber with a lean propane torch system. As the fiber melts, the surface tension coalesces the melt into the highly accurate sphere shape. When the heat source is removed, the melt quickly solidifies into a sphere shape. Previous studies have shown that eliminating the carbon content in the melting flame (hydrogen or arc-heated sources) can reduce the internal inhomogeneities and increase the Q factor of the microsphere. This type of fabrication technique will be used in the future.

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For this study, several microspheres were constructed, ranging in size from 150 to 400 microns in diameter. These sphere sizes were chosen to enable simple and accurate alignment with the tapered fiber to maximize the energy flow to the WGM resonator as well as minimize the amount of energy coupled to non-meridional modes in the sphere. The sphere sizes in this size range also had FSR of the range of 1 nm which matched the mode-hop-free tuning capability of the diode laser used. Thus, the diode laser could be scanned through more than one resonant mode during a single laser scan.

For the tapered fiber, previous research has shown that the smallest fiber possible which still allows a significant energy flow through the tapered section is appropriate for coupling energy into the correct mode of the microsphere. This was achieved by stripping the cladding off of a single mode fiber, and then stretching that fiber in an in-house constructed stretching device driven with micro-stepping motor assemblies. The fiber was heated with the same propane torch during stretching to ensure an even expansion. This stretching process reduced the diameter of the fiber from 6 microns to approximately 1.5 microns. Finally, the sphere was placed in contact with the tapered fiber to ensure energy flow from the fiber to the sphere.

The first results of this experimental setup are shown in Figure 9. These traces represent signal voltage from the fiber optic detected using an IR photodiode versus time. The diode laser was scanned using a sawtooth driver current at 100 Hz over a spectral range of 1.23 nm near 1516.6 nm. With no WGM sphere in place, the signal voltage rises linearly with time. Therefore, each “dip” seen in the profile represents a WGM resonance mode in the sphere. As the laser temperature is varied, the dips appear to move within the time series data, as different spectral regions are accessed as the variation of the laser temperature also affects the output wavelength of the diode laser. Clearly, strong resonating modes are present with maximum energy couplings greater than 20%. While this result is encouraging, there are several significant very weak elements of the signal which make the systems use in CRDS challenging.

While the FSR calculation above describes that 1-2 resonant modes should be present when scanning the laser through this wavelength range, more than 10 modes are seen resonating along the profile. This is due to the excitation of multiple TE modes within the WGM sphere. This arises due to several issues with construction and alignment. When the

\[ T_{\text{laser}} = 18.72 \, ^\circ \text{C} \quad T_{\text{laser}} = 27.94 \, ^\circ \text{C} \quad T_{\text{laser}} = 37.12 \, ^\circ \text{C} \]

Figure 9  Multi-resonant modes excited within the microresonant sphere for different laser tuning ranges (for different laser temperatures). Resonant modes are represented by reduction in detected power and are seen on these traces a “dips” in the detector voltage. When no resonant modes are present, the laser power transmitted through the tapered fiber is linear in time.

Figure 10 Schematic of deformed WGM sphere  Figure 11 Couple of fiber taper to deformed WGM sphere causing other propagating modes.
The tapered fiber – sphere method proved to be much more successful. A bandwidth and Q factor were achieved which are below the best attainable values for the sphere, but are a strong stepping point towards completing CRDS experiments at the microscale. Future improvements to alignment techniques and sphere fabrication methods should lead to large improvements in the resonant quality factor. Once the method is

IV. CONCLUSIONS

The use of WGM microresonators for cavity ringdown spectroscopy is an interesting application of a macroscopic spectroscopic technique to the microscale. The simple scaling argument presented in this paper show that very long pathlengths and ringdown times are achievable through the use of microspheres coupled to tapered fibers. While this method is not readily scalable or able to be mass fabricated like other chip-based nanofabrication techniques, it does off the quickest pathway to utilizing microresonant structures in CRDS experiments. Several optical methods were tested in the study with limited success. Waveguide-based methods proved difficult due to light coupling issues at the entrance and exit of the waveguide. Additionally, coupling inefficiencies led to very limited signal levels.

Figure 12 Experimental output of the tapered fiber/sphere WGM resonator for computing the resonant width of the mode. The line segment a-b is placed at the FWHM of the resonant mode to determine $\Delta \omega_0$. When a liquid sphere is created, the flame melting is that intrinsic to the propane jet flame imposes a significant aerodynamic force on both the tip of fiber and the liquid sphere, which will make a bent fiber tip and an elliptically deformed sphere (Figure 10). This sphere deformation can alter the strong coupling to the meridional mode and allow for leakage to other polar and azimuthal modes. With the addition of the fiber taper alignment, multi-mode WGM resonance is very possible (Figure 11). The shape of the sphere as well as the precise alignment of the fiber taper is fundamental for eliminating these competing modes.

Realizing these elements degrade but do not eliminate the ability to make a Q measurement, the WGM sphere resonator was ultrasonically cleaned and had its alignment optimized with respect to the tapered fiber. Figure 12 shows the output of this effort, which the labeled line segment a-b drawn at the FWHM of the strongest resonating mode of the WGM. Converting from variation in time to frequency, this resonant mode has a bandwidth of 700 MHz, giving a Q factor of $2.8 \times 10^5$. This represents a ringdown time of 0.2 nsec using Equation 3. Clearly, this is two to three orders of magnitude below those values given for a typical sphere in this type of setup (Table 1), strongly suggesting that issues lie in the fabrication process of the sphere. With improved optical fabrication, elimination of the problems shown in Figure 11 is likely and the Q factor can be increased up to levels which are meaningful for CRDS experiments.

Due to the limitations of the Q factor in this system, rudimentary cavity ringdown measurements were attempted using a typical setup (Figure 3) with a fast modulator to act as a shutter for the beam. Unfortunately, due to the multimode behavior, multiple ringdown times and a very low signal to noise ratio made this measurement very difficult to make. With a much larger Q factor sphere and a reduction in the multimode behavior, this CRDS measurement would be much more straightforward. Future work will be aimed at improving the optical fabrication techniques to the point where useful CRDS signals can be detected.
confirmed in the tapered fiber – sphere geometry, the other waveguide-based geometries will be revisited to determine if on-chip based micro-CRDS detection is currently feasible.

V. REFERENCES


