Intracavity chemical absorption sensing using microresonator whispering-gallery modes

G. Farca, S. I. Shopova, A. T. Rosenberger Department of Physics, Oklahoma State University, Stillwater, OK, USA 74078-3072

ABSTRACT

Chemical absorption is investigated using the evanescent fraction of a whispering-gallery mode (WGM). An effective absorption path length of about 50 cm is obtained in methane sensing and about 100 cm in a liquid dye solution.

1. INTRODUCTION

The high-Q WGMs of dielectric microresonators are becoming increasingly useful in sensing applications. Spectroscopic techniques include evanescent-wave absorption sensing and WGM frequency-shift response.¹ We describe here a method for chemical detection by microcavity-enhanced evanescent-wave absorption sensing. Two different types of WGM resonators are used. For gas sensing the resonators used are microcylinders, 125 μ m in diameter, with the WGMs excited along the circumference. By stretching these microcylinders the WGMs can be tuned in resonance with the gas absorption resonances. Moreover, the WGMs can be locked to the laser frequency,² as it scans over the absorption lines of the gas, thus providing the opportunity to perform experiments analogous to wavelength-modulation spectroscopy (WMS). For the case of chemical sensing in liquids the whole system can be immersed in the liquid to be investigated. A microsphere is used in this case since no tunability of the WGMs is needed given the broad absorption feature of the chemicals involved.



Fig. 1. (Left) The tapered fiber excites WGMs in the cylindrical vertical cavity. The cavity can be tuned by applying a voltage to the PZT. (Right) For liquid sensing the WGM resonator is a microsphere obtained by melting the end of an optical fiber.

Light in a microresonator WGM is spatially confined near the dielectric boundary by total internal reflection and propagates around the cavity's perimeter. The mode has an evanescent fraction that extends out into the ambient and can thus be absorbed by an analyte therein. A WGM is excited via mode coupling from an adjacent tapered optical fiber (Fig. 1).

The Q of a WGM can be quite large because its losses, intrinsic (predominantly due to surface scattering) and coupling, can be very low. Because of the intrinsic loss in a microresonator, a WGM resonance appears as a Lorentzian

dip in the coupling-fiber throughput. The depth of this dip is sensitive to the change in WGM loss resulting from absorption in the surrounding analyte, and the primary sensitivity-determining factor is the intrinsic loss.

2. EXPERIMENTAL SETUP

Laser light is launched into a single-mode optical fiber. The fiber is adiabatically tapered in order to preserve single-mode propagation; it is brought into contact with the microresonator and its output is coupled into an optical detector and an oscilloscope. A microsphere is used for the detection of dyes in methanol whereas a vertical fused silica cylinder is used for gas sensing (Fig. 1). After immersing the microresonator in the solution the changes in dip depth of the WGMs overlapping with the broad absorption of the dye are monitored while increasing the concentration of dye. For the gas sensing the fused silica cylinder is attached to a mount that allows the tuning of the WGMs over the comparatively narrower absorption line of the gas. Once again the changes in dip depth are monitored as the WGM scans over the absorption line.

3. RESULTS

Figure 2 shows the variation of the dip depth as it scans over an absorption line of methane. The absorption line is obtained by using a beamsplitter to divert a fraction of the laser light through a 60-cm absorption cell in order to center the laser at that particular wavelength. Scanning the laser in frequency allows both the observation of the methane absorption line and the WGM spectrum. A voltage is then applied to the PZT in order to shift the desired WGM across the domain of interest. The effective absorption length in the case of gas sensing is determined to be approximately 50 cm.



Fig. 2. The position of the bottom of a WGM (square dots) as it scans over an absorption line (total width \sim 2 GHz) of methane at 1 Torr.

As a demonstration of absorbance sensing in liquid solvents, we previously used indocyanine green (ICG) in methanol and determined the effective absorption path length of an immersed 300-µm diameter microsphere. Very low concentrations (nM) of ICG were easily detected at 800 nm, and effective path lengths of about 1 m were measured. More recently, experiments have been done with a different dye (SDA 2072) in methanol at longer wavelengths (1510-1660 nm). Again, effective absorption lengths of about 1 m are found for nM concentrations, in spite of the very high absorption of methanol at these wavelengths (too high for reliable standard spectrophotometer measurements using a 1 cm cuvette). This holds promise for enabling detection of low chemical concentrations in highly absorbing solvents, and we are continuing to investigate this to clarify our understanding and develop a theoretical model that can reproduce the experimental results.

An exciting new development has recently been investigated. This technique allows us to make measurements of the evanescent fraction, the portion of a WGM that extends outside the microsphere. This fraction determines the sensitivity of chemical detection. We measure the resonance linewidth at critical coupling (the regime where the intrinsic loss of the microresonator equals the coupling loss), with and without analyte as depicted in Figure 3.



Fig. 3. WGM linewidth at critical coupling with (black trace) and without (grey trace) analyte (SDA 2072 dye). The change in linewidth for a known absorption coefficient of the analyte is directly related to the evanescent fraction of the WGM.

The linewidth measurements and known chemical absorption coefficient allow us to determine the evanescent fraction, and we compare the measured value of 2.5% to the calculated value. We find the measured value to be slightly larger, implying that the effective chemical concentration increases near the microsphere surface. This gives us a direct measurement of surface effects on sensitivity. This method also provides a means of detecting much larger chemical concentrations (μ M).

4. CONCLUSION AND FUTURE WORK

The viability of an inexpensive, accurate alternative to multi-pass absorption cells is demonstrated. This is so because using a microsphere for trace-gas sensing can improve the effective absorption path length by more than an order of magnitude. The sensitivity of our gas sensor can be further improved by at least an order of magnitude through the use of more advanced techniques such as WMS. Implementation of WMS is currently in progress in our lab; a WGM is locked to the laser so that the laser remains on resonance with the WGM as the wavelength modulation scans the laser over the molecular absorption resonance. Figure 4 shows our first results using this technique. Using a lock-in stabilizer and the PZT element the WGM is locked to the tunable laser as it scans over the absorption line of the gas (methane at 140 Torrs in this case). The lightest trace shows the evolution of the bottom of the WGM dip as it passes over the absorption line of methane. A small change in dip depth can be seen, as the WGM resonance becomes shallower when resonant with the absorption line(s) of methane depicted by the slightly darker trace. The top (black) trace shows the same signal after being noise-filtered and amplified by a lock-in amplifier; two orders of magnitude improvement in the sensing capability was thus realized. Moreover, by selecting the proper shape of the tapered fiber so that multimode propagation is achieved, the sensitivity can be increased by about two more orders of magnitude.



Fig. 4. The WGM is frequency-locked to the laser scan. The lightest curve shows the trace of the bottom of the WGM dip as it scans over the absorption line of the gas plotted as the slightly darker curve. The black curve shows again the WGM dip as it scans over the absorption line of the gas after being noise-filtered and amplified by a lock-in amplifier. The two sides of the graph represent the scan up and then down in frequency.

By coating the microresonators with Hg(Cd)Te nanoparticles we were able to build a microlaser³ that can be used as well for sensing experiments at wavelengths between 1200 and 1700 nm, the range over which lasing can be tuned by changing the size and composition of the nanoparticles. This would give us access to longer wavelengths where stronger gas absorption lines are present.

REFERENCES

- 1. I. Teraoka, S. Arnold and F. Vollmer, "Perturbation approach to resonance shifts of whispering-gallery modes in a dielectric microsphere as a probe of a surrounding medium," J. Opt. Soc. Am. B 20, 1937-1946, 2003.
- 2. J. P. Rezac and A. T. Rosenberger, "Locking a microsphere whispering-gallery mode to a laser," Opt. Express 8, 605-610, 2001.
- 3. S. I. Shopova, G. Farca, A. T. Rosenberger, W. M. S. Wickramanayake and N. A. Kotov, "Microsphere whisperinggallery-mode laser using HgTe quantum dots," Appl. Phys. Lett. 85, 6101-6103, 2004.