

# Heavy water detection using ultra-high- $Q$ microcavities

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Ultra-high- $Q$  optical microcavities ( $Q > 10^7$ ) provide one method for distinguishing chemically similar species. Resonators immersed in  $\text{H}_2\text{O}$  have lower quality factors than those immersed in  $\text{D}_2\text{O}$  due to the difference in optical absorption. This difference can be used to create a  $\text{D}_2\text{O}$  detector. This effect is most noticeable at 1300 nm, where the  $Q(\text{H}_2\text{O})$  is  $10^6$  and the  $Q(\text{D}_2\text{O})$  is  $10^7$ . By monitoring  $Q$ , concentrations of 0.0001% [1 part in  $10^6$  per volume] of  $\text{D}_2\text{O}$  in  $\text{H}_2\text{O}$  have been detected. This sensitivity represents an order of magnitude improvement over previous techniques. Reversible detection was also demonstrated by cyclic introduction and flushing of  $\text{D}_2\text{O}$ . © 2006 Optical Society of America

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Detecting a single species in a mixture of chemically similar molecules can be difficult. Several different techniques have been applied to this problem, as it concerns  $\text{D}_2\text{O}$  in  $\text{H}_2\text{O}$  mixtures. While spectroscopic (emission and absorption) techniques might seem the most logical, since they give the chemical fingerprint of the individual molecules in a mixture, this technique has not achieved low detection sensitivities. Alternative methods, such as protonic conductors<sup>1</sup> and nuclear magnetic resonance,<sup>2</sup> have succeeded in detecting 5% and 10%  $\text{D}_2\text{O}$  in  $\text{H}_2\text{O}$ , respectively. To date, the most sensitive technique is optothermal detection. While this is similar to infrared detection, the signal can be further enhanced to give sensitivities of 30 parts per million per volume (ppmv) of  $\text{D}_2\text{O}$  in  $\text{H}_2\text{O}$ .<sup>3</sup>

Ultra-high- $Q$  microcavities can offer an enhancement of this type of measurement.<sup>4,5</sup> Changes in absorption modify the cavity's intrinsic quality factor, and under constant loading conditions (provided by a coupling waveguide) these modifications to the  $Q$  factor can be observed as changes in the on-resonant optical transmission through the coupling waveguide. In this work, we demonstrate this method for the first time, using the chemically similar species  $\text{D}_2\text{O}$  and  $\text{H}_2\text{O}$ .<sup>6</sup> In a single-pass measurement, which is typical of an optothermal detector or spectrophotometer, the light will pass through the sample, giving an absorption reading over a very small distance. However, the light orbiting inside an ultra-high- $Q$  microcavity ( $Q > 10 \times 10^6$ ) interacts with the sample over a large effective length, and, as shown here, can increase sensitivity to 1 part in  $10^6$  by volume (ppmv) or 0.0001% without increasing the testing footprint. To demonstrate this alternative to conventional chemical detection techniques, planar arrays of ultra-high- $Q$  microtoroids were fabricated as shown in Fig. 1 and described in Ref. 7. The testing proceeded as follows: (1) immerse the microtoroid in 100%  $\text{D}_2\text{O}$ , (2) gradually increase the concentration of  $\text{H}_2\text{O}$  in  $\text{D}_2\text{O}$  until 100%  $\text{H}_2\text{O}$  is reached, and (3) increase the concentration of  $\text{D}_2\text{O}$  to 100%. As shown previously, the difference between the quality factors

in  $\text{H}_2\text{O}$  and  $\text{D}_2\text{O}$  is liquid limited.<sup>8</sup> Therefore the quality factor can be described by  $Q_{\text{liq}} = 2\pi n / \lambda \alpha$ , where  $n$  is the effective refractive index,  $\lambda$  is the wavelength, and  $\alpha$  is the absorption rate introduced into the resonator whispering gallery owing to the presence of the liquid. The refractive indices of  $\text{H}_2\text{O}$  and  $\text{D}_2\text{O}$  are the same, and the resonant wavelength is constant. Since the absorption of  $\text{H}_2\text{O}$  is larger than that of  $\text{D}_2\text{O}$ , the quality factor of the resonator in  $\text{H}_2\text{O}$  is smaller than when the resonator is in  $\text{D}_2\text{O}$ , at 1300 nm.<sup>6</sup>

All detection events were performed by using a single-mode, tunable external cavity laser that was centered at 1320 nm. This laser was coupled to a single-mode optical fiber containing a short, tapered section that acts as a waveguide. The tapered section was used to couple power into and out of the whispering gallery modes of the ultra-high- $Q$  microtoroids.

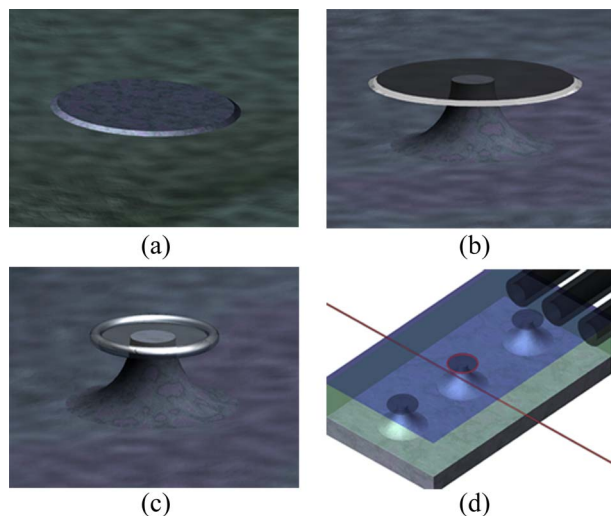


Fig. 1. (Color online) Fabrication outline consisting of (a) patterning the silica into an array of circular oxide pads, (b) etching the silicon wafer substrate with  $\text{XeF}_2$  to form the microdisk, and (c) reflowing the silica disk with a  $\text{CO}_2$  laser to form the toroidal resonator. (d) Using tapered optical fiber waveguides, the 1300 nm tunable laser is coupled into and out of the toroid.

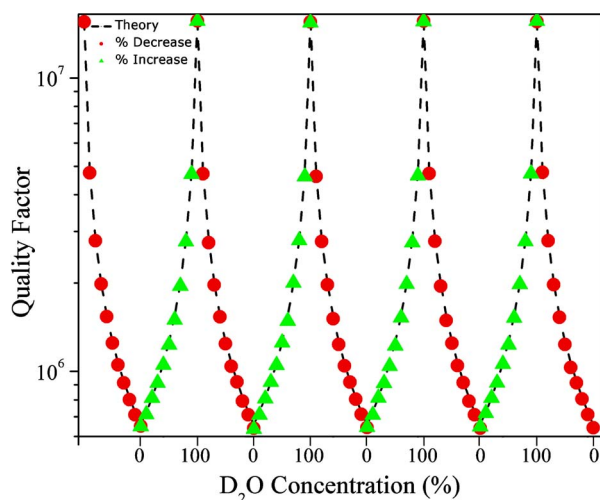


Fig. 2. (Color online) The microtoroid resonator is initially immersed in a solution of 100% D<sub>2</sub>O. The D<sub>2</sub>O concentration of solution is diluted with H<sub>2</sub>O in increments of 10%, until the toroid is immersed in 100% H<sub>2</sub>O. This process of controllably changing D<sub>2</sub>O and H<sub>2</sub>O is repeated five times.  $Q$  is systematically degraded (red circles) and recovered (green triangles) as the D<sub>2</sub>O and H<sub>2</sub>O are exchanged repeatedly.

These waveguides are high-efficiency probes of microcavities that function both in air and in solution and are fabricated by heating an optical fiber using an oxyhydric torch while stretching the fiber.<sup>9</sup> Tapered fibers for testing at 1300 nm were pulled from SMF-28 optical fiber to an average waist diameter of 1  $\mu$ m. During testing, the microtoroids were placed on a high-resolution translation stage and were monitored by two cameras (top and side view) simultaneously. With the taper waveguide in close proximity to the microtoroid, pure water was added and a coverslip was placed on top (Fig. 1), forming a water-filled aquarium.<sup>8</sup> After each solution was added, the resonance spectra were recorded. Then the solution around the toroid was removed and replaced with the next solution in the series.

Both the intrinsic  $Q$  and the resonance position were determined from the transmission spectra. The intrinsic  $Q$  factor was determined by scanning the single-mode laser and measuring both the transmission and the loaded linewidth (full width at half-maximum) in the undercoupled regime over a range of coupling conditions. The intrinsic modal linewidth (and hence intrinsic  $Q$ ) is then computed by using a simple coupling model. The laser scan frequency is optimized to ensure that neither scan direction (increasing frequency versus decreasing frequency) nor scan frequency has any observable impact on linewidth. The position of the resonant frequency was determined by scanning the laser over a 0.1 nm range and monitoring—recording the resonance position from an oscilloscope.

In a first series of measurements, the solutions were prepared in 10% increments (10% H<sub>2</sub>O in D<sub>2</sub>O, 20% H<sub>2</sub>O in D<sub>2</sub>O, etc). The toroid was initially in 100% D<sub>2</sub>O. After the quality factor was determined, all of the D<sub>2</sub>O was removed, until the toroid was in

air. The chamber was then flushed five times with the next-concentration solution (in this case, the 10% H<sub>2</sub>O in D<sub>2</sub>O solution), and the quality factor was again determined. This flushing process was followed for all solutions to remove trace amounts of higher- or lower-concentration solutions. Initially, with the toroid immersed in 100% D<sub>2</sub>O, the quality factor was  $1.55 \times 10^7$ . As can be seen in Fig. 2 and in Table 1, when the concentration of D<sub>2</sub>O was reduced, the quality factor of the toroidal resonator began to decrease. The  $Q$  of the toroid in 100% H<sub>2</sub>O was  $6.4 \times 10^5$ . The theoretical values for each concentration of D<sub>2</sub>O in H<sub>2</sub>O were calculated and are also shown. These theoretical and experimental values are also listed in Table 1 for direct comparison. This  $Q$  decrease was reversible, and by increasing the D<sub>2</sub>O concentration, the quality factor is recovered. This cyclical refreshing process was repeated several times, demonstrating the reproducibility of this detector.

To determine the lower bound on the detection capabilities, larger dilutions of D<sub>2</sub>O in H<sub>2</sub>O were prepared, ranging from 0.01% to  $1 \times 10^{-9}$ %. Starting at 100% H<sub>2</sub>O and slightly increasing the D<sub>2</sub>O concentration using the low-concentration solutions, it was possible to set a lower limit on the detection. As can be seen in Fig. 3 and in Table 1, there is a strong signal at 0.001% D<sub>2</sub>O in H<sub>2</sub>O. However, a small but detectable shift occurs with the 0.0001% D<sub>2</sub>O solution. These values are not believed to reflect a fundamental limit on the detection sensitivity of this device as no attempt to address operational sources of noise in the system has been attempted here.

**Table 1. Experimental and Theoretical Values of  $Q$  for High- and Low-Concentration Detection<sup>a</sup>**

%D <sub>2</sub> O in H <sub>2</sub> O	$Q$ ( $\times 10^6$ )	
	Experimental	Theoretical
100	15.574	15.9879
90	4.6936	4.8015
80	2.7718	2.82494
70	1.9750	2.00116
60	1.5150	1.54935
50	1.2387	1.26398
40	1.0466	1.06738
30	0.915177	0.923707
20	0.803907	0.814123
10	0.712847	0.727783
0.01	0.653315	0.664370274
$1 \times 10^{-3}$	0.647645	0.65863098
$1 \times 10^{-4}$	0.646268	0.65806309
$1 \times 10^{-5}$	0.645152	0.65800631
$1 \times 10^{-6}$	0.644906	0.65800063
$1 \times 10^{-7}$	0.644837	0.658000063
$1 \times 10^{-8}$	0.645483	0.658000006
$1 \times 10^{-9}$	0.644127	0.6580000063
0	0.643987	0.658000

<sup>a</sup>Theoretical values were calculated from  $Q_{\text{liq}}$ , where  $\alpha$  was taken from Ref. 6. All solutions are expressed as a percentage of D<sub>2</sub>O in H<sub>2</sub>O.

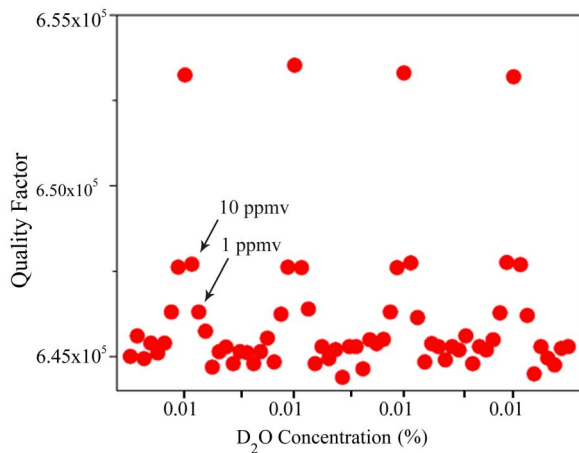


Fig. 3. (Color online) Starting with 100% H<sub>2</sub>O, the concentration of D<sub>2</sub>O was gradually increased by using low-concentration solutions ranging from  $1 \times 10^{-9}\%$  to 0.01%. A large change in the quality factor could be detected at 0.001% (10 ppmv). An additional change in  $Q$  could be detected at 0.0001% (1 ppmv).

The ultra-high- $Q$  microcavity has demonstrated the ability to detect the difference between two chemically similar species, H<sub>2</sub>O and D<sub>2</sub>O, at low concentrations. This detection is based on the subtle difference in optical absorptions between D<sub>2</sub>O and H<sub>2</sub>O, which is then magnified by the quality factor of the resonator. Previous technologies have measured 30 ppmv of D<sub>2</sub>O in H<sub>2</sub>O.<sup>3</sup> This is the equivalent of 0.003% D<sub>2</sub>O in H<sub>2</sub>O. Using resonant cavities, it was possible to improve upon this detection sensitivity by over an order of magnitude. The ability to actively

monitor the presence and simultaneously determine the quantity of D<sub>2</sub>O is very important, especially given its significance in current strategic locations.

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