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Ultra-high-*Q* microcavity operation in H_2O and D_2O

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Optical microcavities provide a possible method for boosting the detection sensitivity of biomolecules. Silica-based microcavities are important because they are readily functionalized, which enables unlabeled detection. While silica resonators have been characterized in air, nearly all molecular detections are performed in solution. Therefore, it is important to determine their performance limits in an aqueous environment. In this letter, planar microtoroid resonators are used to measure the relationship between quality factor and toroid diameter at wavelengths ranging from visible to near-IR in both H_2O and D_2O, and results are then compared to predictions of a numerical model. Quality factors (Q) in excess of 10^8, a factor of 100 higher than previous measurements in an aqueous environment, are observed in both H_2O and D_2O. © *2005 American Institute of Physics*.
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High-Q and ultra-high-Q (UHQ) silica optical microcavities can perform as highly sensitive detectors;[1–3] they derive their excellent transduction abilities from long photon lifetimes within the whispering gallery of the microcavity. Unlike their optical waveguide counterparts, wherein the photon interacts with a functionalized surface only once,[4,5] recirculation within the microcavity allows photons to interact with the surface many times. Additionally, the surface of silica-based microcavities is easily sensitized using silanization agents,[6] amines, carbohydrates, the biotin-streptavidin system,[7] or antibodies.[8] For example, silica microsphere resonators, with a properly sensitized surface, were recently used to distinguish between two strands of DNA.[9] However, while detailed studies have been performed to determine the limits of a resonator's quality factor in air,[10–12] no comparable studies have been performed in water. Because most detection experiments are performed in a water-based solution, it is important to thoroughly understand the impact of this environment on the relationship between the diameter of the microtoroid resonator and the operational wavelengths of interest.

To this end, UHQ silica microtoroid resonators were fabricated over a wide range (50–250 μm) of major toroid diameters as shown in Fig. 1.[13] Experiments were performed in both water (H_2O) and deuterium oxide (D_2O). The D_2O was purchased from Aldrich. D_2O was chosen as the second liquid because it has the same refractive index and, in turn, the same radiation loss as H_2O. However, its absorption at all wavelengths tested is significantly less.[14] This allowed for selective probing of the absorption-loss mechanism and verification of the model developed to describe this system.

The model used finite element analysis to predict the Q-factor of microtoroid resonators immersed in water or D_2S and accounted for two loss mechanisms: radiation loss and absorption loss. The absorption loss was numerically calculated at (680, 1300, 1570) nm using published values of H_2O (0.0045, 1.12, 8.79) cm^{-1} and D_2O (2.46×10^{-4}, 0.105, 0.327) cm^{-1}.[14] The modeling of radiation loss uses a fully vectorial two-dimensional (2D) eigenmode/eigenvalue solver for the Helmholtz equation. The modeled structures consisted of a toroid with a circular cross section (n determined for silica through the Sellmeier equation) surrounded by a lower refractive index environment. As both D_2O and water have identified refractive indices, $n=1.33$, this aspect of the model was the same for both liquids. The minor diameter modeled was 6 μm. The mode solved for was the fundamental whispering gallery mode with, transverse magnetic polarization located nearest desired wavelength of interest. The accuracy of the code was verified for both the resonance wavelength and radiation Q by comparison with those of a microsphere (error $<10^{-4}$ for lambda and $<10\%$ for Q over range of data in the manuscript). The final $Q_{(\text{model})}$ value was arrived at using the expression: $Q^{-1}_{(\text{model})}=Q^{-1}_{(\text{radiation})}+Q^{-1}_{(\text{absorption})}$. The reduced refractive-index contrast for aqueous operation increases radiation loss at a fixed resonator radius, when compared to operation in air. While water or D_2O absorption within the environment is the central factor limiting Q at large radii. It is important to note that the refractive index of water and D_2O are identical, while the absorption of D_2O is significantly less than water.

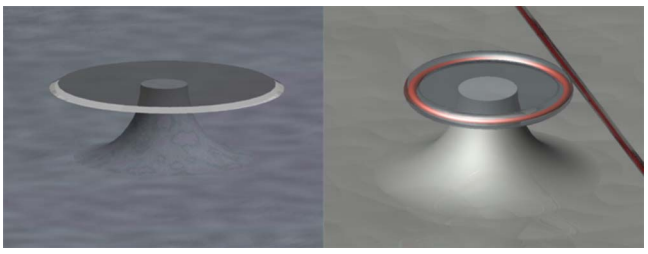

FIG. 1. Ultra-high-Q silica microtoroids are fabricated through a series of three steps. (a) Oxide is lithographically defined and etched using buffered oxide etchant, forming circular oxide pads of controllable diameter; the silicon is then isotropically etched using xenon difluoride gas, creating high-Q silica microdisks. (b) The oxide disks are reflowed using a CO_2 laser, forming the UHQ microtoroids. Power is coupled into and out of the microtoroid resonator using tapered optical fibers.

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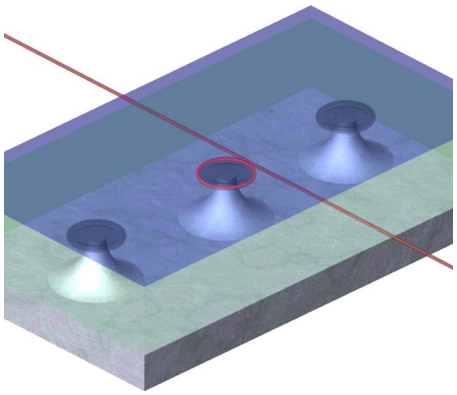

FIG. 2. The ultra-high-Q silica microtoroid is coupled to the fiber taper waveguide. After being immersed in either H_2O or D_2O, a coverslip is placed on top.

This results in identical radiation-loss quality factors for both liquids but significantly higher absorption-loss quality factors for D_2O.

Measurements of the resonator quality factor and analysis of the modal structure were performed at three wavelength bands (680, 1300, and 1550 nm). For testing, a single-mode, tunable external cavity laser was coupled to a single-mode optical fiber containing a short, tapered section. Tapered fiber waveguides are high-efficiency probes of microcavities.[15,16] The tapered-fiber waveguides are fabricated by heating an optical fiber using an oxyhydric torch, while stretching the fiber.[16] Tapered fibers for testing at 680 nm were pulled from F-SV fiber to an average waist diameter of 500 nm. Tapers for testing at 1330 and 1550 nm were pulled from SMF fiber to an average diameter of 1 μm. The tapered section was used to couple power into the whispering gallery modes of the UHQ microtoroids.

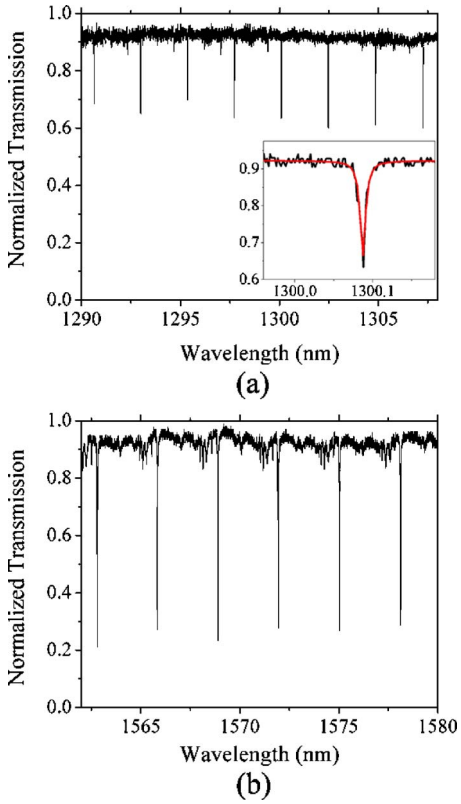

FIG. 3. Transmission spectra of (a) a highly undercoupled microtoroid resonator in H_2O at 1300 nm. The inset shows a single high-Q resonance (black line) with lorentzian fit (red line). (b) The transmission spectra of a microtoroid resonator in D_2O at 1550 nm bands. In this spectra, the resonator is also undercoupled but closer to being critically coupled.

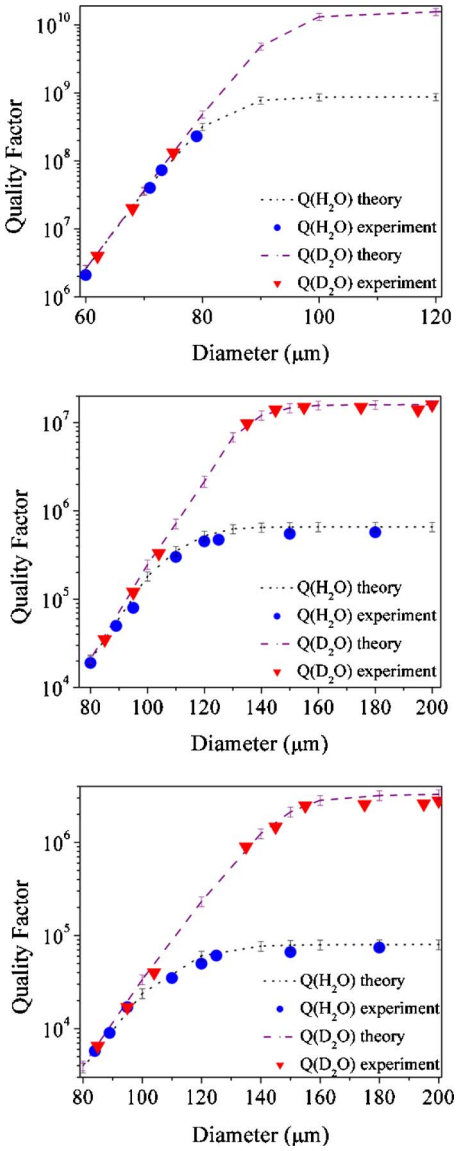

FIG. 4. (a) Quality factors measured and predicted in the 680 nm band plotted versus toroid major diameter. Q increases with major diameter over the range of diameters wherein radiation loss is the dominant loss mechanism. It then plateaus at values set by absorption of the aqueous environment. Above 5×10^8 data taking is unreliable due to laser-linewidth stability limitations. The maximum quality factor achieved in H_2O was 2.3×10^8 and in D_2O was 1.3×10^8. (b) Quality factors measured and predicted in the 1300 nm wavelength band. Both the radiation-loss-limited (small toroid diameter) and aqueous-absorption-loss limited regimes (Q plateau) are apparent. The measured absorptive-loss limits are 5×10^5 (in H_2O) and 1.6×10^7 (in D_2O). (c) Quality factors measured and predicted in the 1550 nm band. In H_2O, the maximum quality factor achieved is 7×10^4. By changing to D_2O, the maximum quality factors increased to 2.8×10^6.

During testing, the UHQ microtoroids were placed on a high-resolution translation stage (100 nm step resolution) and were monitored by two cameras (top and side views) simultaneously. The quality factor of the microtoroid resonator was first determined in air by calculating the resonance linewidth from an oscilloscope to ensure that it was above 10^8. With the taper waveguide in close proximity to the microtoroid, liquid was then added and a coverslip was placed on top (Fig. 2). A "liquid" gap between the toroid and the

taper was maintained when determining the quality factor in either H_2O or D_2O in order to maintain constant coupling between the microtoroid resonator and the taper waveguide.

Figure 3 shows typical transmission spectra in H_2O at 1300 nm and in D_2O at 1550 nm bands. The spectra are taken in the undercoupled regime.[16] The modal structure is dominated by principal transmission minima, confirmed subsequently, to be the fundamental transverse mode of the microtoroids. The intrinsic Q-factor (i.e., the Q-factor in the absence of waveguide loading) was determined by scanning the single-mode laser (short-term linewidth of 300 kHz) and measuring both the transmission and the loaded linewidth (full width at half-maximum) for several waveguide-resonator coupling conditions in the undercoupled regime. The intrinsic modal linewidth (and hence intrinsic Q) was then computed using a simple coupling model.[16] The laser scan frequency was optimized so as to ensure that neither scan direction (increasing frequency versus decreasing frequency) nor scan frequency had any observable impact on linewidth.

The intrinsic Q-factors measured in the 680 nm band plotted versus toroid major diameter are presented in Fig. 4(a). Q-factors trend to larger values with increasing toroid size. This behavior is in good agreement with predictions of the model [also shown in Fig. 4(a)] and results from radiation loss. The maximum quality factor achieved in H_2O was 2.3×10^8 and in D_2O was 1.3×10^8. These values are notable as they represent the highest Q-factors reported to date for operation in an aqueous environment. The highest Q previously reported in water was only 10^6 in a large diameter silica microsphere.[3] Accurate measurements of Q-factors beyond 5×10^8 were not possible in this experiment owing to laser linewidth stability. In principle, however, larger toroid diameters should exhibit quality factors as high as 1×10^9, in water, and 1×10^{10} in D_2O.

The measured intrinsic Q-factors for microtoroids in H_2O and D_2O at different toroid diameters and measured in the 1300 nm band are plotted in Fig. 4(b). Both the radiation-loss-limited regimes and the absorption-loss-limited regimes are clearly visible in these plots. Also plotted are predictions based on the model. Within this wavelength band, D_2O has a lower optical absorption and hence exhibits an absorption-limited Q plateau that is significantly higher than for H_2O (approximately 10^5 for H_2O versus above 10^7 for D_2O). The origin of this absorption limit is the vibration overtone of water. In D_2O this overtone is wavelength-shifted significantly, thereby increasing the observable Q plateau.

The measured intrinsic Q-factors versus toroid diameter in the 1550 nm band are shown in Fig. 4(c), along with the predictions of the model. Again, there is good agreement between measurement and the model, showing the transition between the radiation-loss-limited and absorptive-loss-limited regimes. The strong OH overtone absorption in H_2O lowers the Q plateau to 8×10^4, while for D_2O the value is higher, increasing to above 3×10^6.

In summary, we have determined the Q limits on an UHQ whispering gallery microcavity in a liquid bath at several wavelengths and over a range of sizes in both H_2O and D_2O. Both radiation-loss-limited operation and absorption-loss-limited operation were observed and agreed well with the predictions of a numerical model. Maximum observable Q-factors were greater than 10^8 and were obtained in the 680 nm wavelength band. These radiation-loss-limited values represent the highest Q-values ever reported for microresonator operation in an aqueous environment. Observation of much higher values in the absorptive-loss-limited regime is theoretically possible in this wavelength band. However, in the current experiment, laser-linewidth stability limited data taking to values below 5×10^8. The results presented are fundamental to all future research using resonators as a sensor and lay the groundwork for determining what diameter will maximize sensitivity. The very high values of Q observed here also bode well for development of this class of sensor technology.

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