Ultra-high-Q microcavity operation in H₂O and D₂O

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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 H₂O and D₂O, and results are then compared to predictions of a numerical model. Quality factors (Q) in excess of 10⁸, a factor of 100 higher than previous measurements in an aqueous environment, are observed in both H₂O and D₂O. © 2005 American Institute of Physics. [DOI: 10.1063/1.2099529]

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 lifetime within the whispering gallery of the microcavity.

To this end, UHQ silica microtoroid resonators were 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 CO₂ laser, forming the UHQ microtoroids. Power is coupled into and out of the microtoroid resonator using tapered optical fibers.
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This results in identical radiation-loss quality factors for both liquids but significantly higher absorption-loss quality factors for D$_2$O.

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.\textsuperscript{15,16} The tapered-fiber waveguides are fabricated by heating an optical fiber using an oxyhydric torch, while stretching the fiber.\textsuperscript{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 $\mu$m.

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 surface was introduced. The 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 $5 \times 10^5$ data taking is unreliable due to laser-line width stability limitations. The maximum quality factor achieved in H$_2$O was $2.3 \times 10^8$ and in D$_2$O was $1.3 \times 10^8$. (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 $5 \times 10^5$ (in H$_2$O) and $1.6 \times 10^7$ (in D$_2$O). (c) Quality factors measured and predicted in the 1550 nm band. In H$_2$O, the maximum quality factor achieved is $7 \times 10^4$. By changing to D$_2$O, the maximum quality factors increased to $2.8 \times 10^6$.\textsuperscript{15,16}
taper was maintained when determining the quality factor in either H$_2$O or D$_2$O in order to maintain constant coupling between the microtoroid resonator and the taper waveguide.

Figure 3 shows typical transmission spectra in H$_2$O at 1300 nm and in D$_2$O at 1550 nm bands. The spectra are taken in the undercoupled regime.$^{10}$ 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.$^{10}$ 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$_2$O was $2.3 \times 10^8$ and in D$_2$O was $1.3 \times 10^8$. 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 $5 \times 10^8$ were not possible in this experiment owing to laser linewidth stability. In principle, however, larger toroid diameters should exhibit quality factors as high as $1 \times 10^9$, in water, and $1 \times 10^{10}$ in D$_2$O.

The measured intrinsic $Q$-factors for microtoroids in H$_2$O and D$_2$O 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$_2$O has a lower optical absorption and hence exhibits an absorption-limited $Q$ plateau that is significantly higher than for H$_2$O (approximately $10^5$ for H$_2$O versus above $10^7$ for D$_2$O). The origin of this absorption limit is the vibration overtone of water. In D$_2$O 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$_2$O lowers the $Q$ plateau to $8 \times 10^4$, while for D$_2$O the value is higher, increasing to above $3 \times 10^9$.

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$_2$O and D$_2$O. 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 $5 \times 10^8$. 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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