Raman lasing in a hollow, bottle-like microresonator

Yuta Ooka1,2, Yong Yang1, Jonathan Ward1, and Síle Nic Chormaic1

1Light-Matter Interactions Unit, Okinawa Institute of Science and Technology Graduate University, Onna, Okinawa 904-0495, Japan
2Department of Electronics and Electrical Engineering, Faculty of Science and Technology, Keio University, Yokohama 223-8522, Japan

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We report on the fabrication of an ultrahigh-quality-factor, bottle-like microresonator from a microcapillary and the realization of Raman lasing therein at pump wavelengths of 1.55 µm and 780 nm. The dependence of the Raman laser threshold on the mode volume is investigated. The mode volume of the fundamental bottle mode is calculated and compared with that of a microsphere. Third-order cascaded Raman lasing was observed under pumping at 780 nm. In principle, Raman lasing in a hollow bottle-like microresonator can be used in sensing applications. As an example, we briefly discuss the possibility of a high-dynamic-range, high-resolution aerostatic pressure sensor.

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Whispering gallery mode (WGM) microresonators (WGRs) have high quality (Q) factors and relatively small mode volumes. Because of these properties, they can be used to demonstrate and implement many nonlinear effects, such as stimulated Brillouin scattering,1) four-wave mixing (FWM),2) soliton generation,3) and Kerr optical switching.4,5) The Raman lasing observed in WGRs is based on the resonator material’s phonon band. Thus, in comparison with Brillouin scattering or FWM, phase matching is automatically satisfied; this makes Raman lasing in WGRs relatively easy to implement. The ultrahigh optical Q in WGRs guarantees a very low Raman lasing threshold, and even cascaded processes can be achieved, as has been demonstrated in silica microspheres,6,7) microtoroids,8,9) chalcogenide microspheres,10,11) and poly(dimethylsiloxane) (PDMS polymer) WGRs.12) Raman lasing in WGRs can be very useful for sensing applications, as (i) it does not require a dopant in the resonator’s material and (ii) it decreases the effective linewidth of the WGM through the Raman gain. Single nanoparticle detection using Raman lasing has been achieved13–15) thereby illustrating the high detection resolution attainable.

Conventional WGRs, such as microspheres, microdisks, and microtoroids, confine the optical mode to narrow rings along the equator. Even though the mode volume is small, the tuning range is limited by the design. Bottle-like microresonators (BLMRs) were developed16) because they have good strain and stress tunability. The good tunability makes them attractive devices for sensing, and a number of applications have recently been developed.17,18) As an alternative to heating a tapered optical fiber to achieve a solid bottle shape,18,19) a silica (or other material) microcapillary is used to create a hollow bottle. This creates a highly prolate shape with an empty channel inside. The bottle mode is sensitive to the material contained within the microcapillary, so this device is a promising candidate for optofluidic sensing; to date, refractive index sensing has been demonstrated.18,20) Here, we report on another nonlinear effect in hollow BLMRs, namely, Raman lasing. We demonstrate low-threshold Raman lasing in a BLMR with a Q factor as high as 108. The threshold for Raman lasing is higher than that for a microsphere of similar diameter owing to the larger mode volume of the BLMR. As with other WGRs, we observe a cascaded Raman process when the pump power is high enough.

A hollow BLMR [Fig. 1(a)] was fabricated using the setup illustrated in Fig. 1(b). A silica capillary with an outer diameter of 350 µm and an inner diameter of 100 µm was clamped onto two translation stages. Counterpropagating CO2 beams were focused on each side of the capillary, and a CCD camera was used to monitor the fabrication process from the top. A weak CO2 power was initially used to remove the polymer coating from the capillary. The power was then gently increased to soften the capillary while the two stages were pulled from both ends. Hence, a tapered capillary was fabricated with a decreasing diameter along its length. Next, the tapered capillary was connected to a nitrogen gas source, and the inner pressure of the capillary was kept at around 3 bar. Finally, the CO2 laser beams were focused back on the tapered zone. The molten wall of the tapered capillary swelled gradually until the desired outer diameter was achieved. After fabrication, the capillary with the hollow BLMR was removed from the stages and glued to a glass holder. The microscope image of the device in Fig. 2(a) shows that the BLMR appears as a small bump along...
The microcapillary. The outer diameter of the bottle was 119 µm, and the diameter of the tapered capillary was 103 µm. The thickness of the BLMR wall was estimated to be 25 µm.

The BLMR was placed on a three-dimensional nanopositioner. A tapered fiber was used to evanescently couple light from a tunable diode laser (Newfocus Velocity 6728) into the resonator. In the experiment, the coupling gap was adjusted to minimize the Raman lasing threshold. To maintain the coupling condition, the entire system was isolated from the environment using an enclosure. First, the pump laser was scanned over a range spanning 17.5 GHz at a speed from the blue side of the optical mode, and the transmission spectrum was recorded. To excite different WGMs, the BLMR was moved using the nanopositioner so that the tapered fiber traversed the equator from one side to the other. The position was changed until a ringing effect23 was observed. The ringing indicates an ultrahigh Q mode, and it occurs as the light stored in the microresonator beats with the light from the scanned laser in the taper before the cavity mode decays by intrinsic loss. Figure 2(b) shows the mode used for observing Raman lasing. The measured loaded Q of this mode is $2.3 \times 10^8$.

Light from the taper output was split; one portion was sent to a photodiode, and the other was sent to an optical spectrum analyzer (Anritsu MS9740A). The resolution of the spectrum analyzer was set to 0.07 nm with a wavelength range from 1540 to 1660 nm. The laser frequency was tuned into resonance with the cavity mode. Because of the thermal and Kerr effects, when the laser is tuned from the blue side of the WGM, the WGM shifts in the same direction as the laser tuning, so the pump and the WGM become thermally locked.

The input power was increased while the system was in this self-stabilizing state, and stimulated Raman scattering (SRS), i.e., Raman lasing, was observed on the spectrum analyzer. Figure 3 shows an example of this effect. The strong line to the left is the pump laser with a wavelength of 1545.1 nm. The peak at 1654.5 nm is the Raman lasing, which appears when the pump laser power is high enough; this is the SRS of the BLMR. The interval between the pump and Raman peaks is about 110 nm, which corresponds to the 13.5 THz peak on the Raman gain spectrum of silica. The Raman threshold was measured by varying the input pump power and is shown in the inset of Fig. 3. Note that the total transmission efficiency of the taper used for coupling was 87%, and the final measured threshold was about 1.6 mW.

The power threshold of SRS is given by20 $P_{th} \propto V/Q^2$, where $Q$ represents the optical $Q$ factor of the pump mode, and $V$ is the mode volume, which is defined as

$$V = \iiint e^{-[E(r, \varphi, z)]^2/[E_{\text{max}}(r, \varphi, z)]^2} r \, dr \, d\varphi \, dz.$$  \hspace{1cm} (1)$$

Here, $\varepsilon$ is the permittivity of the medium, $E$ is the electromagnetic field amplitude of the BLMR mode, and $E_{\text{max}}$ is the maximum of $E$.

The WGMs in a BLMR differ from those in a microsphere, as the BLMR mode propagation constant has a component along the $z$-axis [Fig. 1(a)].24,25 Let us assume that the profile of the BLMR is parabolic with $R(z) = R_0 [1 - 1/2(\zeta z)^2]$, where $R_0$ is the maximum radius, and $\zeta^2$ is the curvature of the profile. The first-order radial mode can be explicitly solved in cylindrical coordinates,20,24,25 yielding the wave number of the mode as

$$k_{lp} = \sqrt{\frac{l^2}{R_0^2} + \left(p + \frac{1}{2}\right) \Gamma_z}.$$  \hspace{1cm} (2)$$

The mode is numbered using two indices, $l$ for the azimuthal and $p$ for the axial mode numbers. Further, $p$ represents the number of maxima along the $z$-direction, and $\Gamma_z = 2\zeta^2 R_0$. Suppose the mode spirals back and forth along the $z$-axis and is confined within two caustics located at $\pm z_c$.  

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Fig. 2. (a) Microscope image of the hollow resonator. (b) Transmittance spectrum of a mode (red line) that was used for generating stimulated Raman scattering. The center wavelength of this mode is about 1545.09 nm. The transmission is normalized at the level of the laser background. $Q$ is found to be $2.3 \times 10^8$ by Lorentzian fitting (black line).

Fig. 3. Raman spectrum when the pump power is above the threshold. The wavelength of the pump laser and Raman scattering are 1545.1 and 1654.5 nm, respectively. Inset: relationship between the power of the pump and Raman scattering. The black line is a linear fit to the data.
Fig. 4. Electromagnetic field distribution of the \((l, p) = (160, 1)\) BLMR mode in cylindrical coordinates. \(z_c\) is the caustic point.

(see Fig. 4); then \(l \approx 2\pi R(z_c)/\lambda\), where \(\lambda\) is the wavelength of interest. \(E(r, \varphi, z)\) can be separated as \(E(r, \varphi, z) = \Psi_{lp}(r, z)Z_{lp}(z)\exp(il\varphi)\), where \(\Psi_{lp}(r, z)\) is \(24,25\)

\[
\Psi_{lp}(r, z) = \begin{cases} 
A_lJ_l(k_l r) & r < R(z) \\
H_l^{(2)}\left(\frac{k_l r}{n}\right) + B_lH_l^{(1)}\left(\frac{k_l r}{n}\right) & r > R(z)
\end{cases}
\]

and \(Z_{lp}(z)\) is

\[
Z_{lp}(z) = C_{lp}H_p\left(\sqrt{\frac{\Gamma_1}{2}} r\right) \exp\left(-\frac{\Gamma_1}{4} z^2\right).
\]

In Eq. (3), \(n\) is the refractive index of the BLMR material, and \(k_l\) is defined as the azimuthal component of the propagation constant \(k\), such that \(k_l = kR(z_c)/R(z)\). For a specific mode, \(k\) is represented by \(k_{lp}\). \(A_l\) and \(H_l^{(2)}\) are Bessel and Hankel functions, respectively. \(A_l\) and \(B_l\) are coefficients that can be derived by considering the boundary condition at \(r = R(z)\). In Eq. (4), \(H_p\) is a Hermite function, and \(C_{lp} = [\Gamma_1/(2^{2p+1}(p!)^2)]^{1/4}\) is a coefficient.

The mode volume of the fundamental BLMR mode, \(p = 1\), is the smallest. In our case, \(R_0 = 59.5\mu m, \zeta \approx 0.0052\mu m^{-1}\), and we choose \((l, p) = (160, 1)\) and \(\pm z_c = 10\mu m\). By substituting the above values into Eqs. (2)–(4), the mode volume, \(V_{160,1}\), is calculated to be about 15,000 \(\mu m^3\) from Eq. (1). The mode distribution is plotted in Fig. 4. For comparison, the volume of the fundamental mode of a silica microsphere can be deduced in a similar way.\(^{26}\) For a microsphere with a radius of 60\(\mu m\), the mode volume is about 3500 \(\mu m^3\). The larger mode volume in the BLMR corresponds to a Raman threshold that is more than twice that in a microsphere as obtained in earlier works.\(^{2,6}\)

Finally, we demonstrate cascaded Raman lasing using the same hollow microresonator. The pump light source was changed to one with a wavelength of 771.6 nm (NewFocus Velocity 6712). A BLMR mode was found with a \(Q\) factor of more than \(2 \times 10^7\). At a pump power greater than 1.5 mW, the first-order Raman laser peaking at 802.0 nm appears. When the pump power is increased to more than 2 mW, the first-order Raman laser peak becomes strong enough to excite the second-order Raman peak at 831.2 nm. By such a cascaded process, a third-order Raman laser at 861.3 nm was excited when the pump laser power exceeded 3.8 mW. A spectrum containing three orders of cascaded Raman lasing is shown in Fig. 5. In principle, such a process can continue with further increases to the input power. However, as shown by Min et al.,\(^7\) the cascaded Raman threshold increases at a cubic rate that is proportional to the Raman order number. Hence, in our case, the fourth-order threshold is beyond the maximum output of the laser source.

In conclusion, we have realized SRS in BLMRs. Even though the bottle-like WGR has a larger mode volume than other WGRs, a Raman threshold of 1.6 mW is still achievable. Cascaded Raman lasing can also be obtained. In the future, Raman lasing in the hollow BLMR could be used for high-sensitivity sensing applications, such as aerostatic pressure sensing. Aerostatic pressure tuning of WGRs was previously demonstrated in solid microspheres,\(^{27}\) but the sensitivity was low. In hollow structures, such as microbubbles, high sensitivity and resolution can be set during fabrication, as the sensitivity improves with reduced wall thickness.\(^{28,29}\) However, very thin walls cannot withstand high pressure; therefore, there is a trade off between sensitivity and dynamic range. The Raman laser linewidth is much narrower than the natural linewidth of the passive cavity, so, even with a thick wall, it is still possible to resolve the very small changes at high pressure. The BLMR in this work has an estimated\(^{28}\) sensitivity of 1.14 kHz/Pa, whereas the Raman laser linewidth is in the kHz range, so such a device is feasible for implementing an aerostatic pressure sensor with good resolution working in a high pressure range. Aside from the potential application to high-pressure sensing, laser sensing in WGRs\(^{13,14}\) may also be realized in this structure.

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