High-Q Polymer Microcavities Integrated on a Multicore Fiber Facet for Vapor Sensing

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The further development of “lab on fiber” technology demands more powerful optical microsystems and optical fibers with special functions to create sensors with better performance. Whispering gallery microcavities (WGMs) made of different materials with high quality factors show very good sensitivity to gases, nanoparticles, and biocomponents. However, realizing the excitation and detection of the resonant modes of WGMs on the facets of optical fibers is still a great challenge. This work employs the advanced 3D manufacturing technology of two-photon lithography to fabricate an optical microsystem on the end facet of a multicore optical fiber, which enables excitation and detection of whispering gallery modes on the end facet of an optical fiber. Whispering gallery modes with high quality factors are observed, and the sensing characterization of this device for vapors of three types of volatile organic compounds is investigated. Such optical fiber vapor sensors can be applied in the fields of medical care, environmental monitoring and chemical manufacturing. Additionally, the out-of-plane light coupling strategy may be useful for the design of integrated optical circuits on chips.

Whispering gallery microcavities (WGMs) can be combined with optical fibers to create powerful tools that have the advantages of both, such as high sensitivity, flexibility, and remote sensing. The unique properties of WGMs, such as the high quality factors of their resonant modes,[1] the high response sensitivity of these modes to gases,[2–5] nanoparticles,[6–7] and biocomponents,[8] and various material compatibility,[9] make them one of the best candidates for sensing applications.[10] However, the complex excitation and detection setup for the resonant modes of WGMs lacks flexibility in practical applications. Therefore, a flexible, easy-to-use, and robust strategy to achieve excitation and detection of the resonant modes of WGMs is desirable. Integrating WGMs directly on or on optical fibers is an effective way to achieve this aim. For example, fabricating whispering gallery mode resonators directly in optical fibers has been demonstrated to be an effective strategy for combing WGMs and optical fibers.[11–18] In addition, placing a glass microsphere resonator near the end of a single mode optical fiber by putting it in a thin wall capillary fusion spliced to the optical fiber is another effective strategy. Based on this configuration, researchers demonstrated a chemical vapor sensor with high sensitivity.[14]

Additionally, in view of advanced nanofabrication technology, the end facets of optical fibers are intrinsically extensible platforms for various micro- and nano-optical structures.[15–19] The many works on this topic have made this research direction an important branch of optical fiber optics. The concept of “lab on fiber” defines this field well, which describes the vision that physical, chemical, or biological functions can be added to optical fibers by integrating different micro- and nanoscale optical structures composed of various materials into or onto these fibers. The way light is coupled out of and returned back into the optical fibers must be considered during the design of the structures. For single core optical fibers, only one channel for coupling light out of and into the fibers exists. Therefore, for “lab on fiber” devices based on single core optical fibers, only the structures that can return part of the light back into the core are suitable, which is a strong limitation on the realization of more powerful “lab on fiber” devices. Some important and classical optical configurations that need at least two or more ports for coupling light in or out are excluded from this concept if only single core optical fibers are employed. This limitation can be broken if...
multicore optical fibers are employed during the design of the devices. At the end facets of multicore optical fibers, each core can be treated as a port for coupling light into or out of the optical fibers. Optical interconnection between the cores can be realized on such end facets of multicore optical fibers, which enormously expands the functionalized optical configuration design space of the end facets of optical fibers. Furthermore, the advanced 3D manufacturing technology of two-photon lithography enables us to create tiny 3D “optical building blocks” at the microscale, such as microprisms, tapers, waveguides, and ring resonators, which are the basic optical elements used to manipulate light. Various 3D optical microsystems on the platforms of multicore optical fiber end facets can be realized by integrating optical microelements on them to establish connections between different cores. At the same time, the “lab on fiber” concept will be advanced from the stage of “lab on single core fiber” to the next stage of “lab on multicore fiber.”

In this work, we demonstrate an optical microsystem that consists of micropillars, prisms, tapers, a waveguide, and ring resonators on the end facet of a cleaved seven-core optical fiber, as shown in Figure 1A. Six of the seven cores are arranged in the form of hexagon, and the remaining core is located at the center of the optical fiber as well as at the center of the hexagon. Two cores with opposite positions in the hexagon are chosen as the ports for coupling light in or out. The micropillars located on these two cores play double roles, i.e., supporting the microprisms and tapers connected to them and guiding light out of the core and into the microprisms or vice versa. The two ends of the waveguide are welded to the tips of the tapers, forming a miniaturized “tapered fiber,” i.e., a typical optical element to couple light into WGMs. Two hollow circular truncated cones with different bottom and top radii are designed at both sides of the waveguide. Two rings are designed on top of the circular truncated cones to act as the WGMs. As shown in Figure 1A, assuming that the light is coupled out of the left core, it first passes through a microprism, then is reflected by a microprism and further squeezed into the “tapered fiber,” finally is coupled into the two ring WGMs to excite resonances, and arrives at the right core with the opposite component order. The entire system with the two micropillars with light coupled out-of-plane is reminiscent of the pins of the diode on a circuit board; therefore, we name this system WGMs with receptacle-type ports (WGMs_RTP).

For the design of the entire system, demands from both optics and mechanics must be comprehensively considered. To achieve the highest quality factor of the WGMs, the outer surface of the ring must be as smooth as possible, and the gap between the ring and the waveguide must be accurately controlled to ensure that the condition of the critical coupling points is satisfied. To ensure the stability of the system against disturbance from the environment, the waveguide must be fixed by another support. Therefore, at its middle position, the waveguide is embedded into one of the rings and supported by one of the hollow circular truncated cones while maintaining a gap with the other ring. The light in the waveguide can be coupled into the two ring WGMs at the same time; hence, in the transmission spectra, two types of resonances might occur. To make distinguishing these two types of resonances easier, the two rings are designed with different radii. Considering that the smaller the radius of the WGMs is, the lower the quality factor, the ring with a small radius is used to fix the waveguide, and the ring with a large radius is used to obtain whispering gallery resonant modes with high quality factors.

Based on the technology for 3D optical structure creation on the end facets of optical fibers we previously developed the aforementioned WGMs_RTP is realized on the end facet of a seven-core optical fiber (SM-7C1500(6.1/125), from Fibercore Ltd., UK) with cladding and core diameters of 6.1 and 125 μm, as shown in Figure 1B–E. The photoresist IP-L (from Nanoscribe GmbH, Germany) with a refractive index of 1.50 is used to fabricate the WGMs_RTP. Figure 1B shows a scanning electron microscopy (SEM) image of the top view of the WGMs_RTP on the end facet of the seven-core optical fiber. Figure 1C shows the details of the gap between the waveguide and the large ring with inner and outer radii of 13 and 15 μm, the weld connection between the waveguide and the small ring with inner and outer radii of 3 and 5 μm, and the tips of the
which is close to the highest value \([24,25]\) achieved thus far for polymer) with such a high quality factor is a good candidate to result in Fano resonance, as shown in Figure 2C.

Another six small dips are observed in the spectrum, which are 4 \(\mu\)m and 33.4°. Figure 1E shows the details of the side surface of the large ring: a round and very smooth surface can be observed, which is one of the necessary conditions to achieve high quality factors of the resonant modes. The ring protrudes out approximately 0.6, 0.8, and 1 \(\mu\)m from the hollow circular truncated cones, which makes the resonant light confined near the edge of the ring and not leak into the cones (see Figure 2E).

Figure 2 shows the typical transmission spectra of the WGMs_RTP and the electric field distribution of the resonant modes in the cross section of the large ring. Figure 2A shows three strong dips in the wavelength range from 1530 to 1570 nm. The two narrow dips with wavelengths of 1540.58 and 1558.69 are attributed to the resonant modes of the large ring, while the broad dip at the wavelength of 1551.35 nm originates from the resonant mode of the small ring. The quality factor of the resonant mode with a wavelength of 1540.58 nm is as high as 1.2 \(\times\) 10^5 (Figure 2B), which is close to the highest value \([24,25]\) achieved thus far for WGMs fabricated with the two-photon lithography method. Another six small dips are observed in the spectrum, which are higher order modes weakly coupled with the light in the waveguide. Fano resonance with a transform symmetric shape can be observed when overlap occurs between a narrow resonant mode and a broad mode due to the interference between them.\([26-28]\) In the WGMs_RTP, the whispering gallery resonant modes in the two rings and the resonant modes in the waveguide can be excited at the same time and interfere with each other to result in Fano resonance, as shown in Figure 2C.

The WGMs_RTP made of IP-L photoresist (a type of organic polymer) with such a high quality factor is a good candidate for organic vapor sensors. The narrowband whispering gallery modes will be shifted to longer wavelengths when the WGMs_RTP is placed in an environment with organic vapor due to the swelling effect\([29]\) of the photoresist in response to the organic vapor. To characterize the sensing performance of the WGMs_RTP for various organic vapors with different concentrations, three types of organic vapor environments with stable and quantitative concentrations are prepared. The volatile organic compounds (VOCs) propylene glycol monomethyl ether acetate (PGMEA), isopropanol (ISO), and alcohol (ALC) are mixed with pure water at various concentrations, and 100 mL of the mixed liquids is put into glass chambers with a volume of 125 mL. Then, the chambers are sealed with corks with pinholes through them. The pinholes are used to keep the pressures in the areas of the chambers above the mixed liquids the same as that of the outside atmosphere. The chambers are kept at a constant temperature of 22 °C, and the pressure of the air in the environment is the standard atmospheric pressure (101 325 Pa). As the concentration of the mixed liquid, the temperature and the pressure of the atmosphere are kept stable and fixed, the concentrations of the organic vapors in the areas of the chambers above the mixed liquids will also be stable and fixed.

Figure 3A shows the transmission spectra of the WGMs_RTP when it is put into the vapor of the PGMEA-water mixed liquid in chambers with mixed liquid volume ratio concentrations varying from 0% to 50% in steps of 5%. As shown, all the resonant modes in the spectra are redshifted with increasing concentration. The resonant modes shift faster in the lower concentration range (0–10%) than in the higher concentration range (10–50%). For example, the resonant mode at a wavelength of 1525.88 nm for the pure water environment redshifts to 1552.67 nm for the 5% concentration mixed liquid, with a wavelength shift of 26.79 nm, while this resonant mode shifts to 1558.69 nm for the 100% concentration mixed liquid.

For the PGMEA-water mixed liquid in the chambers, the narrowband modes will shift to longer wavelengths when the WGMs_RTP is placed in an environment with organic vapor due to the swelling effect of the photoresist in response to the organic vapor.
concentrations varying from 0% to 2% in steps of 0.2% are used to characterize the sensing performance of the WGMs in the RTP configuration. To more accurately characterize the sensing performance, the shapes of the resonant modes change from symmetric to antisymmetric, which is due to the interference between the whispering gallery mode of the WGMs and the waveguide resonant mode. To more accurately characterize the sensing performance of the WGMs_RTP for various concentrations of organic vapor, dilute mixed liquids with varying concentrations. Transmission spectra when the device is placed in the vapor environment produced by a PGMEA aqueous solution are shown in Figure 3A, where the solid circles, diamonds, and triangles correspond to the PGMEA, isopropanol and alcohol vapors, respectively. Such low LODs show the high sensitivity of the device to the PGMEA vapor compared to isopropanol and alcohol vapors. The device shows higher sensitivity to the PGMEA vapor than to the isopropanol and alcohol vapors, mainly because both the molecule weight and refractive index of PGMEA are larger than those of isopropanol and alcohol (Table 1). Therefore, the same amount of PGMEA molecules absorbed on the surface of the ring will cause larger expansion of the radius and increase of the refractive index of the large ring are proportional to the volume of adsorbed molecules, the wavelength shift can be expressed as \[ \Delta \lambda = \Delta \lambda_{\text{max}} \times \frac{c}{Kc} \] where \( \lambda \) is the wavelength of the resonant modes, \( c \) is the concentration of the saturated vapor, \( Kc \) is the Henry’s coefficient, and \( \Delta \lambda_{\text{max}} \) is the wavelength shift due to the swelling effect. Assuming that the changes in the radius and refractive index of the large ring are proportional to the volume of adsorbed molecules, the wavelength shift can be expressed as 

\[ \Delta \lambda = \frac{c}{Kc} \times \frac{\mu}{\rho} \times \Delta V \] 

where \( \mu \) is the molar volume of the solvent, \( \rho \) is the density of the solution, and \( \Delta V \) is the volume of adsorbed molecules.

The sensitivities at concentrations below 200 ppm for the PGMEA, isopropanol and alcohol vapors are estimated to be 21.700, 3.382, and 3.873 pm ppm\(^{-1}\), respectively. Such sensitivities are much higher than those previously reported, with the main reason being that the ring resonator in our configuration more easily expands after the absorption of molecules. The ring resonator is not adhered to the hard glass surface of the optical fiber; instead, it is supported by a soft and deformable hollow circular truncated cone with a height of 20 µm. Furthermore, the hollow circular truncated cone also expands due to the swelling effect, hence, the expansion of the ring resonator encounters almost no resistance. Based on such high sensitivities and the high wavelength sweeping accuracy of the measurement system, with a standard deviation \( \sigma \) less than 1 pm, the lower limits of detection (LODs), defined as 3σ/sensitivity, can be calculated as 0.138, 0.887, and 0.775 ppm for PGMEA, isopropanol and alcohol, respectively. Such low LODs show the advantage of the WGMs_RTP configuration.

Table 1. Physical properties and steady-state sensor response parameters for all VOC analytes.

<table>
<thead>
<tr>
<th>VOC</th>
<th>( H_p ) ( [\text{mol} (\text{m}^2 \text{pa})^{-1}] )</th>
<th>( P ) ( [\text{g cm}^{-3}] )</th>
<th>( M ) ( [\text{g mol}^{-1}] )</th>
<th>RI</th>
<th>Sensitivity ( [\text{pm ppm}^{-1}] )</th>
<th>LOD ( [\text{ppm}] )</th>
</tr>
</thead>
<tbody>
<tr>
<td>PGMEA</td>
<td>0.990</td>
<td>0.960</td>
<td>132.16</td>
<td>1.4020</td>
<td>21.700</td>
<td>0.138</td>
</tr>
<tr>
<td>Isopropanol</td>
<td>1.679</td>
<td>0.7855</td>
<td>60.1</td>
<td>1.3776</td>
<td>3.382</td>
<td>0.887</td>
</tr>
<tr>
<td>Alcohol</td>
<td>2.355</td>
<td>0.789</td>
<td>46.07</td>
<td>1.3169</td>
<td>3.873</td>
<td>0.775</td>
</tr>
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Figure 3. Sensing characterization of the WGMs_RTP for organic vapors with varying concentrations. Transmission spectra when the device is placed in the vapor environment produced by a PGMEA aqueous solution with concentration varying from A) 0% to 50% in steps of 5% and B) 0% to 2% in steps of 0.2%. C) Relationship between the wavelength shift (\( \Delta \lambda \)) of the resonant modes and the concentrations of the PGMEA, isopropanol and alcohol vapors.
A lineshape evolution of the resonant modes from symmetric to antisymmetric occurs, as shown in Figure 3A, which is a typical characteristic of Fano resonance. The Fano resonance originates from the superposition of two Lorentz resonances with different bandwidths. Indeed, in the WGMs RTP, the large ring supports narrowband whispering gallery resonant modes, while the small ring supports broadband modes, as illustrated in Figure 4. The two modes weakly couple with each other when the narrowband mode is located outside of the broadband mode. However, the two modes strongly couple when the narrowband mode is located inside the broadband mode. The change in the relative positions of the two modes occurs due to the different wavelength shifts with increasing PGMEA concentration. The smaller the radius of the ring is, the larger the resonant mode shifts for the same increase in PGMEA concentration.

The time response performances of the devices for the three types of vapors are also measured, as shown in Figure 5. The wavelength of the swept-frequency laser is set to periodically sweep from 1533.68 to 1534.00 nm within 0.04 s. Initially, the device is put into a chamber with 99.8 mL pure water, and then, 0.2 mL organic compounds are injected into the water with a syringe. The resonant modes immediately show a redshift, with wavelength shifts within 0.3 s of 35, 51, and 37 pm corresponding to PGMEA, isopropanol, and alcohol (Figure 5B). The results indicate that for a response of a 0.1 pm wavelength shift, the average times are 0.86, 5.9, and 0.81 ms for organic vapors with concentrations less than 150 ppm. Furthermore, considering that time is needed to reach the equilibrium state in the mixing processes of the organic compounds and water and of the vapors and air above the surface of the mixed liquids, the concentrations of the vapors during the measurements are much less than those in the stable cases (less than 150 ppm); therefore, the time response performance may be better than the values estimated above. Furthermore, the reversibility of the device is verified experimentally. The device is put into and taken out from the ISO vapor on top the chamber for four cycles, an ISO aqueous solution with concentration of 20% is used to produce the vapor. For each cycles, both the time for the device in the vapor and air is 100 s. Exponential increasing and deceasing behaviors of the Δλ are observed after the device being put into and taken out from the vapor, as shown in Figure 5C (where the solid circles are the experimental results, and the solid curves are exponentially fitting). For each cycles, the Δλ increases and reaches to the same value of 8 nm, and then decrease to 0 nm again, which demonstrate the good reversibility of the device. It is worth to note that it takes almost 90 s for the Δλ increasing to reach to the maximum value, while less than 50 s to the minimum value, which is determined by the absorption and evaporation process of the ISO molecules on the polymer of the photoresist.

In conclusion, we have demonstrated that the end facets of a multicore optical fiber are more extensible platforms for...
micro-optics. Each core can be treated as the input or output port of light, and optical microsystems can be built on the end facets of the multicore optical fiber by interconnecting different cores with various optical elements, such as prisms, waveguides, tapers, and ring resonators, thereby producing better optical fiber-based devices. As an example, using the 3D manufacturing technology of two-photon lithography, we build WGMs on the end facet of a seven-core optical fiber, and we design and equip a receptacle-type setup to guide and couple light into the microcavities. Whispering gallery resonant modes with high quality factors have been obtained on the end facet of a multicore optical fiber. The sensing performances of such devices for organic vapors are investigated, showing sensitivities of 21.700, 3.382, and 3.873 pm ppm⁻¹ for PGMEA, isopropanol, and alcohol vapors in the low concentration range and a fast time response of 0.3 s, with wavelength shifts of 35, 51, and 37 pm for organic vapors with concentrations less than 150 ppm. Furthermore, such a strategy of out-of-plane receptacle-type coupling to the microcavities may effectively increase the integration degree in the application of such microcavities in array form, which is useful for research on optical integrated circuits. In the future, if various sensitive structures or materials are added into or on the surface of the microcavities, then optical fiber sensors based on this design for biocomponents, magnetic fields, and pressure can also be realized.

Acknowledgements
S.Y.Z. and S.-J.T. contributed equally to this work. This research was supported by the National Natural Science Foundation of China (Nos. 61735002, 11474206, 11404224, 1174243, and 11774246); Youth Innovative Research Team of Capital Normal University (No. 008/19530050146); Beijing Nova Program (No. Z161100004916100); and Capacity Building for Sci-Tech Innovation–Fundamental Scientific Research Funds (Nos. 008/19530050170, 008/19530050180, 008/18530500186, and 025185305000/142). S.-J.T. and Y.-F.X. were supported by the National Key R&D Program of China (Grant No. 2016YFA0301302) and the NSFC (Grants Nos. 61435002, 11474206, 11404224, 1174243, and 11527901).

Conflict of Interest
The authors declare no conflict of interest.

Keywords
lab on fiber, two-photon lithography, vapor sensing, whispering gallery microcavities

Received: April 11, 2019
Revised: June 30, 2019
Published online: