# Analysis of single nanoparticle detection by using 3-dimensionally confined optofluidic ring resonators

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Abstract: We theoretically analyze the ability of 3-dimensionally confined optofluidic ring resonators (OFRRs) for detection of a single nanoparticle in water and in air. The OFRR is based on a glass capillary, on which bottleshaped and bubble-shaped ring resonators can form. The spectral position of the whispering gallery mode in the OFRR shifts when a nanoparticle is attached to the OFRR inner surface. For both ring resonator structures, the electric field at the inner surface can be optimized by choosing the right wall thickness. Meanwhile, different electric field confinement along the capillary longitudinal axis can be achieved with different curvatures. Both effects significantly increase the sensitivity of the ring resonator for single nanoparticle detection. It is found that the sensitivity is enhanced about 10 times, as compared to that of a solid microsphere biosensor recently reported, and that the smallest detectable nanoparticle is estimated to be less than 20 nm in radius for a  $\Delta\lambda\lambda$  resolution of  $10^{-8}$ . The high sensitivity and the naturally integrated capillary based microfluidics make the OFRR a very promising sensing platform for detection of various nano-sized bio/chemical species in liquid as well as in air.

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OCIS codes: (230.5750) Resonators; (280.1415) Biological sensing and sensors.

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# 1. Introduction

Viral particles are responsible for the majority of human fatal diseases, including Ebola fever, influenza, HIV, SARS, dengue fever, and so on. Those small infectious agents with radius ranging from 1 nm to 800 nm spread and transmit extremely rapidly, and leave very limited time for treatment if humans are infected [1, 2]. The prevention and early diagnosis of those diseases require fast and trace amount detection of virus in liquid and in air. Among many approaches employed, the optical ring resonator based biosensor is one of the most sensitive devices, capable of detecting a single virion or nanoparticle in a real-time and label-free manner [3, 4]. In a ring resonator, light circulates and forms whispering-gallery modes (WGMs). When a virion or nanoparticle binds onto the resonator surface, its interaction with the WGM leads to a spectral shift or mode splitting [3, 4]. To date, by measuring the wavelength shift, a single influenza particle (50 nm in radius) in liquid has been detected experimentally with a solid microsphere [3]. Recently, by measuring the mode splitting, the detection and sizing of a single nanoparticle (30 nm in radius) in air have also been demonstrated with a microtoroid [4]. However, despite their excellent sensing performance, both structures lack of an efficient fluidic system to rapidly deliver samples to the sensing head (i.e., the ring resonator), which may significantly lengthen the detection time, in particular, when detecting a single nanoparticle.

In this work, we investigate the ability of the optofluidic ring resonator (OFRR) for single nanoparticle detection [5]. The OFRR is based on a glass capillary, whose cross section forms the ring resonator. In contrast to the solid microsphere and microtoroid where the outer surface is used for detection, the OFRR utilizes its interior surface to capture the analyte. Its naturally integrated capillary microfluidics enables efficient and rapid delivery of analytes in liquid and in air. Furthermore, by changing the wall thickness of the OFRR, the electric field of the WGM in the radial direction can be optimized for detection of different sizes of molecules near the OFRR inner surface [6]. In addition, bottle-shaped and bubble-shaped OFRRs can be created along the capillary, which strongly confine the WGM in the axial direction and significantly reduces the mode volume [7, 8]. Both radial and axial effects tremendously enhance the sensitivity of the OFRR in detecting a single nanoparticle. In this paper, we will theoretically analyze the sensing capability of the microbottle and microbubble ring resonators under different conditions (wall thickness, poloidal curvature, and nanoparticle size, etc.). It is shown that about 10-fold sensitivity enhancement over a microsphere biosensor could be achieved and that the smallest detectable nanoparticle is estimated to be less than 20 nm in radius. The high sensitivity in combination of the naturally integrated microfluidics makes the OFRR a very promising sensing platform for detection of various sizes of bio/chemical species in liquid and in air.

# 2. Model and theory

The geometries of the cylindrical OFRR, microbottle OFRR, microbubble OFRR, and solid microsphere, along with their respective parameters, are shown in Fig. 1. The cylindrical



Fig. 1. Schematic of different ring resonators under study. (a) Cylindrical OFRR in cylindrical coordinates (r,  $\theta$  and z with the origin at the arbitrary location on z axis). (b) Microbottle OFRR in cylindrical coordinates (r,  $\theta$  and z with the origin at the bottle center). (c) Microbubble OFRR in spherical coordinates (r,  $\theta$  and  $\varphi$  with the origin at the bubble center). (d) Solid microsphere in spherical coordinates (r,  $\theta$  and  $\varphi$  with the origin at the sphere center).

OFRR is essentially a 2-dimentional ring resonator without any confinement along the axial direction where the other three structures provide 3-dimensional confinement. In this study, we focus on the microbottle OFRR and microbubble OFRR, whose WGMs are analyzed separately.

For the microbottle OFRR, we use the adiabatic invariants method [9, 10]. As shown in Fig. 1(b), we assume that the profile of the microbottle radius along the z direction, R(z), can be described as:

$$R(z) = R_0 / \sqrt{1 + (\Delta k z)^2}, \qquad (1)$$

where  $\Delta k$  is a parameter to describe the curvature of the profile. When  $(\Delta kz)^2 < 0.05$  (or  $\Delta k < 0.22$  assuming the field extension in the *z* direction is 1  $\mu$ m) the adiabatic approximation is applicable and the field distribution  $E(r, \varphi, z)$  can be separated as  $E=E_r(r, z)E_{\varphi}(\varphi)E_z(z)$ .  $E_{\varphi}(\varphi)$  can be solved as  $E_{\varphi}(\varphi)=\exp(im\varphi)$  and  $E_r(r, z)$  can be written as [11, 12]:

$$E_{r} = \begin{cases} AJ_{m} \left( k_{\varphi}^{(m,l)} n_{1} r \right), & r \leq R_{1} \left( z \right) \\ BJ_{m} \left( k_{\varphi}^{(m,l)} n_{2} r \right) + CH_{m}^{(1)} \left( k_{\varphi}^{(m,l)} n_{2} r \right), & R_{1} \left( z \right) < r \leq R_{2} \left( z \right) \\ DH_{m}^{(1)} \left( k_{\varphi}^{(m,l)} n_{3} r \right), & r > R_{2} \left( z \right) \end{cases}$$
(2)

where  $J_m$  and  $H_m^{(1)}$  are the *m*th Bessel function and *m*th Hankel function of the first kind, respectively.  $k_{\varphi}^{(m,l)}$  is the amplitude of the resonant wave vector component in  $\varphi$  direction labeled by the azimuthal index *m* and the radial index *l*.

In the z direction, the equation for  $E_z$  is the same as the harmonic oscillator problem and the corresponding solution can be expressed as [10]:

$$E_{z} = E_{0}H_{q}\left(\sqrt{\frac{m\Delta k}{R_{0}}}z\right)\exp\left(-\frac{m\Delta k}{2R_{0}}z^{2}\right),$$
(3)

where  $H_q$  is the *q*th Hermite polynomial. The relevant resonant wave vector component in the *z* direction is  $k_z = [m^2/R_0^2 + (2q+1)m\Delta k/R_0]^{1/2}$  and the total wave vector is  $k = (k_{\varphi}^2 + k_z^2)^{1/2}$ . Note

that when  $\Delta k=0$ , the microbottle OFRR is the same as the cylindrical OFRR in Fig. 1(a), which can be described by Eq. (2).

The electric field distribution of the microbubble OFRR,  $E(r, \theta, \varphi)$ , can be separated into  $E_r(r)Y_l^m(\theta, \varphi)$ , where  $Y_l^m$  is the spherical harmonics of the *l*th degree and the *m*th order.  $E_r$  can be written as [11]:

$$E_{r} = \begin{cases} Aj_{m} \left( k^{(m,l)} n_{1} r \right), & r \leq R_{1} \\ Bj_{m} \left( k^{(m,l)} n_{2} r \right) + Ch_{m}^{(1)} \left( k_{\phi}^{(m,l)} n_{2} r \right), & R_{1} < r \leq R_{2} , \\ Dh_{m}^{(1)} \left( k^{(m,l)} n_{3} r \right), & r > R_{2} \end{cases}$$
(4)

where  $j_m$  and  $h_m$  are the *m*th order spherical Bessel function and the *m*th order spherical Hankel function of the first kind. Note that when  $n_1=n_2$  in Fig. 1(c), the microbubble OFRR becomes a solid microsphere.  $E_r$  can be simplified as:

$$E_{r} = \begin{cases} Aj_{m} \left( k^{(m,l)} n_{1} r \right), & r \leq R_{1} \\ Bh_{m}^{(1)} \left( k^{(m,l)} n_{2} r \right), & r > R_{1} \end{cases},$$
(5)

In order to compare the field distribution form different structures, we also introduce the normalization condition  $\int |E_r|^2 dr = 1$  and  $\int |E_z|^2 dz = 1$  for all modes, where the unit of r and z is chosen to be  $\mu$ m.

The resonant wave vectors for all three structures (microbottle, microbubble, and solid microsphere) are numerically solved by home-made programs based on the Mie scattering theory [11].

In the presence of a nanoparticle on the ring resonator surface, the resonant wavelength shifts, which can be numerically calculated by [13, 14]:

$$\frac{\Delta\lambda}{\lambda} = \frac{\int_{particle} \left(n_p^2 - n^2\right) |E|^2 dV}{2\int n^2 |E|^2 dV}$$
(6)

where  $n_p$  and n are the refractive indices of the nanoparticle and the medium and n is usually a function of space coordinates. The integration in numerator is taken within the nanoparticle and the integration in denominator is calculated in the whole space. For simplicity, in this work only the modes with the electric field parallel to the surface are studied. The other modes with the magnetic field parallel to the surface will generate slightly better improvement in the sensitivity (approximately 25% increase in  $\Delta\lambda\lambda$ ) due to the electric field discontinuity at the interfaces.

#### 3. Results and discussion

The relation between the radial distribution of the electric field of the 3rd order mode and the wall thickness is studied in Fig. 2 for the microbottle OFRR [Fig. 2(a), (b) and (c)] and microbubble OFRR [Fig. 2(d), (e) and (f)], respectively. The core of the OFRR is filled with water and the outside is air. For microbottle, the field is plotted at z=0. We choose the 3rd order mode because it can be experimentally realized based on our previous studies [6].

In Fig. 2(a) and (d), the wall thickness is 2.5  $\mu$ m and the most part of the electric field resides inside the wall. According to Eq. (6), the sensitivity for surface mass detection is proportional to the field strength near the surface. Therefore, when the field at the water-glass interface is only a tail of an exponential decay, the sensitivity is low. In Fig. 2(b) and (e), the wall thickness is reduced to 1.6  $\mu$ m and the first electric field peak emerges near the water-



Fig. 2. The electric field distribution of the 3rd order mode in the radial direction with different wall thicknesses for the microbottle (a-c) and the microbubble (d-f). In all case,  $R_2=36$  mm,  $n_1=1.33$ ,  $n_2=1.45$ ,  $n_3=1$ , m=288 for the microbottle, l=288 for the microbubble. (a)  $R_1=33.5 \,\mu$ m,  $\lambda=1022.543860$  nm. (b)  $R_1=34.4 \,\mu$ m,  $\lambda=979.4255256$  nm. (c)  $R_1=34.7 \,\mu$ m,  $\lambda=969.5206896$  nm. (d)  $R_1=33.5 \,\mu$ m,  $\lambda=1020.959929$  nm. (e)  $R_1=34.4 \,\mu$ m,  $\lambda=977.9460628$  nm. (f)  $R_1=34.7 \,\mu$ m,  $\lambda=967.9444502$  nm. (g) Field distribution of the 1st order mode of a microsphere in the radial direction.  $R_1=36 \,\mu$ m,  $n_1=1.45$ ,  $n_2=1.33$ , l=288,  $\lambda=1040.227437$  nm. (h)  $|E_{r0}|^2$  vs.  $R_1$ . Dashed line represents  $|E_{r0}|^2$  of the microsphere in (g).

glass interface, which increases the sensitivity for surface mass or surface adsorption detection. When the wall thickness further decreases to  $1.3 \,\mu\text{m}$ , as shown in Fig. 2(c) and (f), the first peak of the field is pushed into the core and the surface mass sensitivity deteriorates. For comparison, the electric field of a microsphere with a radius of 36  $\mu\text{m}$  surrounded with water is also drawn in Fig. 2(g). Due to the light confinement of the mode, the field peak never reaches the water-glass interface.

In Fig. 2(h),  $|E_{r0}|^2$  is plotted as a function of  $R_1$  for the microbottle and microbubble where  $E_{r0}$  is the electric field amplitude at the water-glass interface. Although the mathematical field expressions of the microbottle and microbubble are different, their  $|E_{r0}|^2$  curves are virtually the same. There is a maximum value for both curves, which represents the best wall thickness for surface mass detection. The curve near the maximum changes slowly, suggesting that the requirement for the optimal wall thickness is not quite critical. In comparison with the microsphere of the same size, the microbottle and microbubble have twice as large  $|E_{r0}|^2$  when the wall thickness is between 1.5 µm to 2 µm, showing the advantage of the OFRR based structures in surface mass detection. Practically, this wall thickness and the related tolerance are experimentally obtainable [6, 8, 15].

The electric field distribution along the capillary axis (*i.e.*, the *z* direction for the microbottle and the  $\theta$  direction for the microbubble and microsphere) are also investigated. In Fig. 3(a), the field distributions of the 0th order mode in Eq. (3) for the microbottle with different  $\Delta k$  is shown and compared with that of the microbubble. The inset shows the actual geometries for those structures. It is clear that the large curvature can efficiently decrease the field amplitude at *z*=0 (in the subsequent studies, we assume that the nanoparticle is always attached to the equator).  $|E_{z0}|^2$  as a function of  $\Delta k$  is plotted in Fig. 3(b) and is compared with that for the microbubble. Note that at the point that  $(R_0\Delta k)=1$ , the microbottle becomes the microbubble and has the same field intensity, suggesting that our original adiabatic approximation to describe the microbottle is always end that for spherical



Fig. 3. (a) The normalized 0th order mode field distribution of the microbottle and microsphere along the capillary with different  $\Delta k$ . For the microbottle, m=288,  $R_0=36$  µm. For the microsphere, l=288,  $R_1=36$  µm. Inset is their actual geometries. (b) The relation between  $|E_{z0}|^2$  and  $\Delta k$ . z=0.

structures. Therefore, in this region, the microbottle has the better sensing performance.

After the 3-dimensional electric field distribution is computed, the fractional resonant wavelength shift  $\Delta\lambda\lambda$  caused by a single nanoparticle in water is numerically calculated using Eq. (6) and the results are shown in Fig. 4. In order to obtain the maximum shift, the location of the nanoparticle is assumed to be at the equator and is at the inner surface of the OFRR based structures (or at the outer surface of the microsphere). Note that when the nanoparticle location is off the equator, the corresponding shift can be deduced by comparing the local field intensity to the peak value in Fig. 3(a).

In Fig. 4(a),  $\Delta\lambda/\lambda$  for different  $R_1$  is plotted while  $R_2$  and the nanoparticle radius,  $R_p$ , are fixed at 36 µm and 50 nm, respectively. Polystyrene beads ( $n_p$ =1.59) are chosen as a model system to simulate viral particles [3, 4, 18]. As expected, there is a maximum shift when the wall thickness is around 1.6 µm for both microbottle and microbubble. Under this wall thickness, the electric field peak is near the water-glass interface. Also we can see that the structure with a bigger  $\Delta k$  has a larger shift due to the light confinement in the z direction. In Fig. 4(b), the maximum shift vs. the nanoparticle radius is plotted for different structures. The change of  $\Delta\lambda/\lambda$  is basically proportional to the  $R_p^3$ , which is identical to the previously reported linear relationship between  $\Delta\lambda/\lambda$  and the nanoparticle volume [3, 14]. The smallest detectable nanoparticle radius is related to the smallest detectable  $\Delta\lambda/\lambda$ . Practically, by controlling the instability of the light source, mechanical vibration, thermo fluctuation, and noise from the photodetector,  $\Delta\lambda/\lambda$  of  $10^{-8}$  can be achieved as demonstrated in Ref [3, 6]. Under this detection limit, we arrive at the smallest detectable nanoparticle radius of approximately 17 nm for the curve of  $\Delta k$ =0.2.

The dependence of the sensitivity on  $\Delta k$  is studied by calculating the maximum shift caused by a single nanoparticle with 50 nm in radius for different  $\Delta k$ . As shown in Fig. 4(c), the wavelength  $(\Delta \lambda/\lambda)_{\text{max}}$ - $\Delta k$  curve exceeds the microsphere value when  $\Delta k$  is about 0.003 and intersects with the microbubble value at  $\Delta k$ =0.028 where the shape of the microbottle is equivalent to microbubble. Note that in the simulation, there is no boundary for the curvature of the microbottle profile, which can increase indefinitely by continuously increasing  $\Delta k$ . But considering the adaptive range of adiabatic invariant assumption and fabrication limitations, we predict approximately 10 times enhancement by comparing a large  $\Delta k$  microbottle ( $\Delta k$ =0.2) with a microsphere.

Figure 4(d) plots the linear relationship between the nanoparticle size and the best wall thickness, which is related to the maximum wavelength shift. The slope of this curve is virtually unity, which may result from the fact that the peak of the electric field needs to be at the center of the nanoparticle to obtain the maximum field overlap. Note that the needed change in the best wall thickness is almost negligible when the nanoparticle radius varies less than 100 nm. As a result, in actual applications, one thickness may be sufficient to detect



Fig. 4. Results of single nanoparticle detection in water. The modes with the 3rd order in the *r* direction and the 0th order in the *z* direction are used. For the microbottle and microbubble,  $R_2=36 \ \mu\text{m}$ , m=288, l=288,  $n_1=1.33$ ,  $n_2=1.45$ ,  $n_3=1$ . For the microsphere,  $R_1=36 \ \mu\text{m}$ , m=288, l=288,  $n_1=1.45$ ,  $n_2=1.45$ ,  $n_3=1$ . For the microsphere,  $R_1=36 \ \mu\text{m}$ , m=288, l=288,  $n_1=1.45$ ,  $n_2=1.33$ . The nanoparticle is located at the equator and at the inner surface of the OFRR based structures or at the outer surface of the microsphere.  $n_p=1.59$ . (a) Normalized wavelength shift caused by a single nanoparticle as a function of  $R_1$ . Wavelength shift from the microsphere is also shown for comparison. Nanoparticle radius is 50 nm. (b) The maximum wavelength shift vs. the nanoparticle radius for the microbottle with different  $\Delta k$ , microbubble, and microsphere. (c) The wavelength shift from the microbottle as a function of  $\Delta k$ . (d) The best wall thickness that gives the largest spectral shift as a function of the nanoparticle radius.

nanoparticles whose radii are in the range of 100 nm.

Finally, the detection of single nanoparticle in air is also studied. Here we choose the microbubble as a representative structure. The typical electric field distribution of the 3rd order mode in the r direction is shown in Fig. 5(a). Because the capillary is filled with air (n=1), the refractive indices inside and outside the capillary are the same. Consequently, the peak of the field intensity will always be inside the wall. The Q factor related to the radiation loss of the mode with the 3rd order in the r direction and the 0th order in the  $\theta$  direction is also calculated for different wall thicknesses and is shown in Fig. 5(b). Unlike the nanoparticle detection in water, in which the radiation loss is low and optical absorption of water limits the Q, the Q for the nanoparticle detection in air can be drastically degraded due to the radiation loss, which may reduce the wavelength spectral resolution and deteriorate the detection limit [13].

In Fig. 5(c),  $\Delta\lambda\lambda$  is plotted as a function of  $R_1$  and compared with that of the microsphere.  $\Delta\lambda\lambda$  increases when the fraction of light in the air core increases. Considering Fig. 5(b) and (c) in which the sensitivity is increased at the expense of the Q factor, a trade-off wall thickness is chosen to be around 1 µm where the Q factor is still about 10<sup>10</sup> and the shift is more than 10 times larger than that of the microsphere. In Fig. 5(d),  $\Delta\lambda\lambda$  for different nanoparticle radii is plotted for a microbubble with a 1-µm thick wall. The smallest detectable nanoparticle radius is also about 20 nm and the  $\Delta\lambda\lambda\lambda$  is 10 times larger than that of a microsphere across a wide range of nanoparticle radii.

## 4. Conclusion

In the summary, the single nanoparticle detection capability of 3-dimensional confined OFRRs is studied analytically and is compared with microsphere biosensors. In the radial direction, the electric field at the inner surface can be optimized by choosing the right wall



Fig. 5. Results of single nanoparticle detection in air.  $R_2=36 \ \mu m$ ,  $n_1=1$ ,  $n_2=1.45$ ,  $n_3=1$ , m=288, l=288. (a) Field distribution of the 3rd order mode of the microbubble.  $R_1=35 \ \mu m$ ,  $\lambda=784.0225833 \ m$ . (b) Q factor of the 3rd order mode of the microbubble as a function of the inner radius. (c) Fractional wavelength shift as a function of the inner radius. Shift from a solid microsphere is also plotted for comparison.  $R_p=50 \ m$ ,  $n_p=1.59$  and the single nanoparticle is located at the equator and at the inner surface of the microbubble (or at the outer surface of the microsphere). (d) Fractional wavelength shift for the microbubble and for the microsphere with different nanoparticle radii. The wall thickness of the microbubble is 1  $\mu m$ .

thickness. Along the long axis, the electric field can be efficiently confined by the curvature. Both effects significantly increase the sensitivity of the OFRR for single nanoparticle detection. It is found that the sensitivity is enhanced about 10 times, as compared to that of a solid microsphere biosensor in Ref [3], and that the smallest detectable nanoparticle is estimated to be less than 20 nm in radius (assuming  $\Delta\lambda\lambda$  of  $10^{-8}$  spectral resolution). The extension of the detection capability to smaller nanoparticle sizes enables the detection of more types of important and lethal viruses (such as SARS virus and dengue virus with radii below 50 nm) [16, 17]. Very recently,  $\Delta\lambda/\lambda$  of  $10^{-9}$  is achieved on a solid microsphere using the frequency-doubling technology via a PPLN [18]. The corresponding smallest detectable nanoparticle is reduced to 20 nm in radius [18]. Applying such a high spectral resolution technology to the OFRR should enable the detection of a nanoparticle of only approximately 7 nm in radius (see Fig. 4(b) and 5(d)). Similarly, detection of a single molecule (such as proteins whose size is around 5 nm in radius), which is the "holy grail" in label-free sensing, may even become possible. The high sensitivity for nanoparticle detection presented in this paper and for the detection of smaller molecules studied earlier [6], and the naturally integrated microfluidics make the OFRR a very promising sensing platform for detection of various sizes of bio/chemical species in liquid and in air.

#### Acknowledgments

This work is supported by the Wallace H. Coulter Foundation Early Career Award. H.L. is supported by China Scholarship Council (No. 2009610120) and by the University of Michigan.