Microring resonators with flow-through nanopores for nanoparticle counting and sizing

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Abstract: This paper proposes a high precision method for nanoparticle counting and sizing using a microring resonator-waveguide system that contains a flow-through nanopore. Theoretical analysis is carried out based on the coupled-mode theory, showing that when the nanoparticle passes the nanopore a temporal pulse signal can be detected and that the peak amplitude depends linearly on the nanoparticle volume. It is estimated that a nanoparticle of sub-10 nm in size may be detectable.

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References and links

1. Introduction

Nanoparticle counting and sizing are essential for a broad range of applications such as nanotechnology, virology, disease diagnosis, and biomedical research [1, 2]. Electron microscopes such as SEM and TEM have long been used for particle sizing, but they are very expensive and bulky. Dynamic laser scattering provides a much simpler way to measure the particle size down to the order of 10 nm, but requires relatively large sample concentration. Nanopore technology based the Coulter principle has also been used for nanoparticle counting and sizing. Recently, scanning ion occlusion spectroscopy was developed based on size-tunable micro/nanopores fabricated on a polymer membrane and was able to measure the particle size down to 50 nm [1, 2].

The optical microring resonator is an emerging sensing technology that has been used for highly sensitive biomolecular detection in the past decade. Recently, the ring resonator was also employed for nanoparticle detection, counting, and sizing [3–14]. For example, using the whispering gallery mode (WGM) frequency shift method, Arnold et al. detected single Influenza A viral particles (~100 nm in diameter) attached to the ring resonator surface [3]. Furthermore, by using the plasmonic enhancement mechanism, his group was able to detect and size single MS2 viral particles (~25 nm in diameter) with the ring resonator [14]. Using the self-referencing mode splitting method, Yang et al. demonstrated detection and sizing of single nanoparticles on the order of 30 nm in diameter [9, 12, 13]. Koch et al. and Yi et al. employed backscattering caused by the nanoparticles attached to the ring resonator surface to detect nanoparticles [5–8]. Despite unprecedented achievement, the aforementioned approaches may suffer from the following limitations. First, they all rely on the direct attachment of the nanoparticles to the ring resonator. Accumulation of nanoparticles on the ring makes it difficult to continuously monitor and count nanoparticles, as the signal generated by later nanoparticles may significantly be affected by the presence of nanoparticles deposited earlier. Second, they rely primarily on diffusion for nanoparticles to reach the ring resonator surface, which process is slow, less controllable, and does not generate accurate information about the nanoparticle concentration. Third, the attachment position of the nanoparticle on the ring resonator surface is random. Consequently, the interaction between the nanoparticle and the WGM varies, which is likely to produce erroneous size information, in particular, when the frequency shift method [3, 10, 11, 14] or the backscattering method [5–8] is used.

In this paper, we propose and analyze a microring resonator with flow-through nanopores, which overcomes the aforementioned problems. The proposed device is illustrated in Fig. 1. A dielectric microring resonator and an adjacent waveguide bus are embedded in a low-index medium. A flow-through nanopore (or nanopores) can be created in the coupling region between the ring and the waveguide. When a nanoparticle of interest is present in the nanopore, the coupling between the ring resonator and the waveguide changes. In addition, the scattering arising from the nanoparticle in the nanopore causes additional loss in the WGM. Both effects may result in a change in transmitted light intensity at the detector located at the distal end of the waveguide.

The device has a few distinct advantages over the current microring resonator designs. First, the flow-through structure allows nanoparticles to be transported directly to the detection zone by convection, which process is rapid and highly controllable. Second, the detection zone is well defined, which renders much better detection consistency and hence

accuracy in size measurement. Third, the device enables continuous particle counting and sizing, thus enabling measurement of the particle concentration. Fourth, it is compatible with the conductance-based nanopore technologies. Thus, hybrid dual mode (optical and electrical) detection becomes possible.

In this paper, we present detailed theoretical analysis of the coupling effect and the scattering effect due to the presence of a nanoparticle inside the nanopore. It is shown that the coupling effect is dominant in determining the sensing signal, and that detection and sizing of nanoparticles below 10 nm is possible.

2. Theory

Referring to Fig. 2 and assuming that the laser is on resonance with the WGM of the ring resonator we can write the transmitted light intensity $T$, as:

$$T = |b_1|^2 \left( \frac{1 - \chi}{1 + \chi} \right)^2,$$

where $t$ is the transmission coefficient and is related to the total coupling coefficient $\kappa$ by $t^2 + \kappa^2 = 1$, $\chi = |a_2/b_2|$, which is related to the fractional round-trip intensity loss $L$, by $\chi = (1 - L)^{1/2}$. When $\kappa << 1$ and $L << 1$, Eq. (1) can be approximated as:

$$T = \left( \frac{\kappa^2 - L}{\kappa^2 + L} \right)^2,$$

In the presence of a nanoparticle in the nanopore, the coupling between the ring and the waveguide as well as the loss will be modified, which will affect the transmitted light intensity, \textit{i.e.}:
\[ \Delta T = (\frac{\partial T}{\partial \kappa^2}) \cdot \Delta (\kappa^2) + (\frac{\partial T}{\partial L}) \cdot \Delta L, \]  

(3)

where \( \frac{\partial T}{\partial \kappa^2} = S_c \) and \( \frac{\partial T}{\partial L} = S_L \) is the device sensitivity with respect to the coupling change and the loss change. \( \Delta (\kappa^2) \) and \( \Delta L \) have different dependence on the nanoparticle size and refractive index, as discussed later.

### 2.1 Contribution from the coupling change

Let us consider \( S_c \) first. Figure 3(a) plots the transmitted light intensity as a function of \( \kappa^2 \) for different intrinsic ring resonator losses, \( L \). In contrast to the featureless transmission seen in a typical waveguide-waveguide system, which is linearly proportional to the total coupling \( \kappa^2 \), the existence of the ring resonator completely changes the characteristics of the transmission. The transmission becomes much more sensitive when the ring resonator-waveguide system is operated in the under-coupling regime where \( \kappa^2 < L \). The lower the loss (or the larger \( Q_0 \)) is, the steeper the transmission curve is in the under-coupling regime. When \( \kappa^2 = L \), the critical coupling at which \( T = 0 \) is achieved. As discussed later, we will employ these phenomena for enhanced nanoparticle detection. Figure 3(b) provides the more quantitative details about \( S_c \) as a function of \( \kappa^2 \) for various ring resonator losses. For example, according Fig. 3(b), for \( L = 0.2, 0.1 \) and 0.01, \( S_c \) can be as large as \(-13.5, -27, \) and \(-270, \) respectively for \( \kappa^2/L = 0.1 \). When \( \kappa^2 = L \) or when \( \kappa^2 \gg L \), \( S_c \) approaches zero.

The total coupling coefficient, \( \kappa^2 \), between the ring resonator and the waveguide bus in the absence of a nanoparticle can be calculated based on the coupled-mode theory [15, 16]:

\[
\kappa = \int_{-\infty}^{\infty} C(z)dz = \sqrt{\frac{2\pi R}{\gamma}} \cdot \exp\left( -\frac{\Delta \beta^2 R}{2\lambda_0} \right) \cdot C(0),
\]

(4)

where \( R \) is the ring resonator radius. \( \gamma = (2\pi / \lambda_0) \sqrt{n_{rr}^2 - n_M^2} \) is the decay constant of the WGM evanescent field outside the ring resonator. \( n_{rr} \) and \( n_M \) are the refractive index of the ring resonator and the surrounding medium, respectively. \( \lambda_0 \) is the wavelength in vacuum. \( \Delta \beta \) is the difference propagation constant in the ring resonator and in the waveguide. In the reminder of our calculation, we assume that \( \Delta \beta = 0 \) without losing generality. \( C(0) \) is the local coupling coefficient at \( z = 0 \) and is given by the following overlap integration [17]:

![Fig. 3. (A) Normalized power transmission as a function of coupling coefficient \( \kappa^2 \) based on Eq. (1). The critical coupling (zero transmission) occurs when \( \kappa^2 = L \). The under-coupling regime refers to \( \kappa^2 < L \). (B) \( S_c \) as a function of coupling coefficient \( \kappa^2 \) for various ring resonator losses.](image-url)
where $H_i^x$ is the magnetic field component along the x-direction. Based on $H_i^x$, we can obtain the E-field along the y-direction $E_i^y$ for the ring resonator and $E_{W}^y$ for the waveguide bus:

$$E_i^y = \frac{-\mu_0 \omega_0 H_i^x - \frac{1}{\epsilon_0 n_i^2 \beta_i} \frac{\partial^2 H_i^x}{\partial y^2}}{\beta_i} = -\frac{\mu_0}{\beta_i} H_i^x, \quad i = RR, W$$

(6)

where $\epsilon_0$, $\mu_0$, and $\omega$ are permittivity, permeability, and the angular frequency, respectively. $\beta$ is the propagation constant.

In the presence of a nanoparticle (assumed to be a cube with a lateral size of $d$) as illustrated in Fig. 4, the additional local coupling coefficient and total coupling coefficient can be expressed as:

$$\Delta C_{NP}(0) = \frac{\omega_0 \epsilon_0 (n_{NP}^2 - n_{water}^2) \int_0^{d/2} \int_0^{d/2-d/2} (E_{Lab}^y)^* E_{W}^y dxdy}{2\int_0^{-\infty} \int_{-\infty}^{-\infty} (E_{Lab}^y)^* H_{Lab}^x dxdy}$$

(7)

and

$$\Delta \kappa = \int_0^{d/2} \Delta C_{NP}(0) dz = d \cdot \Delta C_{NP}(0),$$

(8)

where $n_{NP}$ and $n_{water}$ are the refractive index of the nanoparticle and water that fills the nanopore, respectively. Note that the overlap integration in Eq. (7) takes place only in the region where the nanoparticle is present. In Eqs. (7) and (8), the nanoparticle is located at $(x, y, z) = (0, 0, 0)$, i.e., the center of the nanopore. Detailed calculation shows that $\Delta C_{NP}(0)$ and hence $\Delta \kappa$ remain nearly unchanged when the nanoparticle moves within the x-z plane inside the nanopore. This can be explained by the fact that any decrease of the electric field of one mode (e.g., $E_{Lab}$) when the nanoparticle moves off the center in the x-z plane is compensated for by the increase of the electric field of another mode (e.g., $E_{W}$). This phenomenon is important for detection consistency, as the detection signal will remain the same regardless the nanoparticle’s position in the x-z plane. In contrast, when the nanoparticle moves along the y-direction, the largest $\Delta C_{NP}(0)$ and hence $\Delta \kappa$ are obtained when the nanoparticle is located at $y = 0$ (the middle of the nanopore along the y-direction). Therefore, when a nanoparticle flows through the nanopore, a temporal peak in the coupling will emerge, which can be used in particle counting. For simplicity, in the reminder of the paper, all the calculations are carried out for the nanoparticle located at the origin. Furthermore, we assume that the WGM of the ring resonator has the same mode field distribution as a straight waveguide of the same dimension. Based on these assumptions, we have

$$\Delta \kappa = \frac{\omega_0 \epsilon_0 (n_{NP}^2 - n_{water}^2) E_{Lab}^y(r_0) E_{W}^y(r_0) d^3}{2\int_0^{-\infty} \int_{-\infty}^{-\infty} (E_{Lab}^y)^* H_{Lab}^x dxdy} = \frac{k_0^2 (n_{NP}^2 - n_{water}^2) E^2(r_0) d^3}{2\beta \int_0^{-\infty} \int_{-\infty}^{-\infty} |E_{Lab}^y|^2 dxdy},$$

(9)

where $E(r_0) = E_{W}^y(r_0) = E_{Lab}^y(r_0)$ is the electric field at the location of the nanoparticle (i.e., at the origin). It is important to note that $\Delta \kappa$ is proportional to $d^3$, which is the volume of the nanoparticle. For a spherical nanoparticle with a diameter of $d$, $d^3$ in Eq. (9) should be replaced by $\pi d^6/6$, i.e., the sphere volume.

Table 1 lists $\kappa$ and $\Delta \kappa$ for different gaps between the ring resonator and the waveguide.
Fig. 4. E-field distribution of the mode \((E_y)_{11}\) inside the gap between the ring resonator and the waveguide. Here we assume that the WGM of the ring resonator has the same mode field distribution as a straight waveguide of the same dimension. The squares show the cube-shaped nanoparticle at different locations within the gap.

bus based on Eqs. (4) and (9). In the calculation, we use the following parameters: ring resonator radius \(R = 15 \mu m\); width \(w = 0.45 \mu m\) and height \(h = 0.1 \mu m\) for both the ring and the waveguide; \(n_{RR} = n_W = 1.7\), \(n_M = 1.45\), \(n_{water} = 1.33\), and \(n_{NP} = 1.55\); \(\lambda_0 = 1.55 \mu m\); particle size \(d = 100 \text{ nm}\). From Table 1, it is important to observe that for a given ring resonator-waveguide system, \(\Delta \kappa / \kappa\) remains nearly the same regardless of the size of the gap. This is understandable by comparing Eqs. (5) and (7), as both \(C(0)\) and \(\Delta C_{NP}(0)\) (hence \(\kappa\) and \(\Delta \kappa\)) have the same dependence on the gap.

### Table 1. \(\kappa\) and \(\Delta \kappa\) for a 100 nm nanoparticle with different gaps

<table>
<thead>
<tr>
<th>Gap (nm)</th>
<th>(\kappa)</th>
<th>(\Delta \kappa)</th>
<th>(\Delta \kappa / \kappa)</th>
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<tbody>
<tr>
<td>370</td>
<td>0.31834</td>
<td>5.7193e-4</td>
<td>1.7966e-3</td>
</tr>
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<td>450</td>
<td>0.2520</td>
<td>4.5281e-4</td>
<td>1.7969e-3</td>
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<td>2.9217e-4</td>
<td>1.7969e-3</td>
</tr>
<tr>
<td>1000</td>
<td>0.0506</td>
<td>9.0958e-5</td>
<td>1.7976e-3</td>
</tr>
</tbody>
</table>

2.2 Contribution from the loss change

The Rayleigh scattering induced loss arising from the presence of the nanoparticle, \(L_s\), can be written as:

\[
L_s = \frac{n_{water}^2}{2} E^2(r_0) \cdot \sigma,
\]

where

\[
\sigma = \frac{2\pi^2 d^6}{3} \left( \frac{n_{NP}^2 - n_{water}^2}{\lambda_0^2 n_{NP}^2 + 2n_{water}^2} \right)^2
\]

is the Rayleigh scattering cross section. Therefore,

\[
\Delta L = \frac{n_{water}^2 E^2(r_0) \sigma}{n_{RR}^2 \int \int |E_{RR}|^2 \ dx \ dy},
\]

We now compare the relative contribution of the coupling and scattering to the sensing signal, \(\Delta T\). According to Eq. (3), we have

\[
\Delta T = S_c \cdot \Delta (\kappa^2) + S_s \cdot \Delta L,
\]
Comparison $\Delta(\kappa^2)$ and $\Delta L$ using Eqs. (9) and (12) shows that

$$\frac{\Delta(\kappa^2)}{\Delta L} = \frac{3\kappa}{\pi^2} \left(\frac{n_{NP}^2 + 2n_{water}^2}{n_{NP}^2 - n_{water}^2}\right) \cdot \frac{n_{RR}^2}{n_{water}^2} \cdot (\lambda_{0}\kappa)^3. \quad (14)$$

Using $n_{NP} = 1.55$, $n_{water} = 1.33$, $n_{RR} = 1.7$, we have,

$$\frac{\Delta(\kappa^2)}{\Delta L} = 1.65\kappa(\lambda_{0}\kappa)^3, \quad (15)$$

At $\lambda_0 = 1550$ nm, $\kappa$ is usually much larger than 0.01 (see, for example, Table 1). Therefore, we have $\Delta(\kappa^2)/\Delta L$ exceeding $10^2$ and $10^3$ for $d = 100$ nm and 10 nm, respectively. In addition, according to Fig. 3(b) and Fig. 5, when the ring resonator is operated in the under-coupling regime or near the critical point, $|S_2|$ is larger than or close to $|S_1|$. Therefore, the contribution of the scattering loss to the transmitted light intensity change, $\Delta T$, can be ignored, especially when we deal with nanoparticles whose size is well below 100 nm.

Our analysis performed so far reveals that the coupling between the ring resonator and waveguide plays a dominant role in determining the nanoparticle sensing signal, which concurs with recent experimental results that suggest that the change in the coupling between the ring resonators induced by protein molecules may significantly modulate the characteristics of the ring resonator system [18].

### 3. Detection principle and sensor design

In actual measurement, we use the fractional change of the transmitted light intensity, $\Delta T/T$, as the sensing signal. Based on the discussion in Section 2.2 and Eq. (2), we have

$$\frac{\Delta T}{T} = \frac{\Delta \ln(T)}{\Delta(\kappa^2)} \cdot \Delta(\kappa^2) = \left(\frac{1}{\kappa^2 - L} - \frac{1}{\kappa^2 + L}\right) \cdot 4\kappa^2 \cdot \frac{\Delta \kappa}{\kappa}, \quad (16)$$

Considering a situation in which $\kappa^2$ is near $L$, Eq. (16) can be simplified as:

$$\frac{\Delta T}{T} = \frac{4f}{1-f} \cdot \frac{\Delta \kappa}{\kappa}, \quad (17)$$

where $f = \kappa^2/L$. Figure 5 plots $4f/(1-f)$. When $\kappa^2$ approaches the critical coupling point ($\kappa^2 \rightarrow L$), $1/(\kappa^2 - L)$ approaches infinity.
To highlight the advantage of using a ring resonator for nanoparticle detection, we consider a situation in which the ring resonator is simply replaced by a curved waveguide that has the same $\kappa^2$. In this case, the fractional change in the transmitted light ($\Delta T/T$) induced by the nanoparticle would be the same as the coupling induced loss (i.e., $\Delta(\kappa^2)$). Therefore, with the ring resonator, the sensing signal (i.e., $\Delta T/T$) is enhanced by a factor of approximately $2/[L(1-f)]$. Apparently, the lower $L$ is, the higher enhancement is for given $f$.

Here we present a quantitative example of detecting and sizing nanoparticles with the proposed ring resonator-waveguide system. For the parameters used in Table 1, we choose a gap of 370 nm so that $\kappa = 0.31834$ and $\kappa^2 = 0.1$. To optimize the sensitivity, we place the ring resonator system close to the critical coupling point (but not exactly at the critical coupling point, as there would be no light transmitted). In the actual experiment, this can be done through appropriate designs by adjusting the gap between the ring resonator and the waveguide to find the optimal coupling (i.e., $\kappa^2$). Another approach is to tune $L$ to match $\kappa^2$, which can be accomplished by first fabricating a ring resonator with $L$ being slightly lower than needed for the critical coupling and then introducing a small additional loss (for example, using an AFM tip [5]) to increase $L$. For the purpose of discussion, we can set $L = 0.09$, which corresponds to $Q_0 = 6.9E3$ and should be quite easy to achieve with the state-of-the-art lithographic method. Based on Fig. 6 ($f = 1.1$) and Table 1, we have

$$\frac{\Delta T}{T} = 44 \cdot \frac{\Delta \kappa}{\kappa} = 7.9\%,$$

for a 100 nm nanoparticle, which can easily be detected. Using a normalized standard deviation in light intensity of 8E-6 that has been demonstrated earlier [19], we estimate that a nanoparticle of 5 nm in size is detectable. Certainly a larger signal ($\Delta T/T$) (and hence smaller detectable nanoparticles) can be achieved, if $\kappa^2$ and $L$ are brought even closer to each other or a ring resonator with a higher Q-factor is used. In actual experiments, once $\Delta T/T$ is measured, the nanoparticle volume (and its cube- or sphere-equivalent diameter) can be deduced by $\Delta \kappa/\kappa$ through Eq. (9) and Table 1.

5. Conclusion

We have performed detailed analysis of using the ring resonator-waveguide system for possible nanoparticle detection. It is found that the coupling between the ring resonator and the waveguide bus can be significantly modulated by the presence of a nanoparticle in the gap, thus generating a sensing signal that depends linearly on the nanoparticle volume. Our
work presents a new method for rapid and accurate counting and sizing of nanoparticles ranging from a few hundred nanometers to sub-10 nm.

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