Sub-pg mass sensing and measurement with an optomechanical oscillator

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Abstract: Mass sensing based on mechanical oscillation frequency shift in micro/nano scale mechanical oscillators is a well-known and widely used technique. Piezo-electric, electronic excitation/detection and free-space optical detection are the most common techniques used for monitoring the minute frequency shifts induced by added mass. The advent of optomechanical oscillator (OMO), enabled by strong interaction between circulating optical power and mechanical deformation in high quality factor optical microresonators, has created new possibilities for excitation and interrogation of micro/nanomechanical resonators. In particular, radiation pressure driven optomechanical oscillators (OMOs) are excellent candidates for mass detection/measurement due to their simplicity, sensitivity and all-optical operation. In an OMO, a high quality factor optical mode simultaneously serves as an efficient actuator and a sensitive probe for precise monitoring of the mechanical eigen-frequencies of the cavity structure. Here, we show the narrow linewidth of optomechanical oscillation combined with harmonic optical modulation generated by nonlinear optical transfer function, can result in sub-pg mass sensitivity in large silica microtoroid OMOs. Moreover by carefully studying the impact of mechanical mode selection, device dimensions, mass position and noise mechanisms we explore the performance limits of OMO both as a mass detector and a high resolution mass measurement system. Our analysis shows that femtogram level resolution is within reach even with relatively large OMOs.

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1. Introduction

During the past ten years various micro and nano-electromechanical (MEMS and NEMS) mass sensors have been proposed and demonstrated based on monitoring the shift of the mechanical resonant frequency (dynamic mode), or the bending induced by the added mass (static mode) [1–12]. Typically in the dynamic operational mode, the mechanical modes of the resonator/oscillator are excited thermally [5,10], piezoelectrically [11,12] or electrostatically [6–10]. The vibrational frequencies are then monitored using piezo-resistive [8,11], capacitive [6] or optical mechanisms [7,9,10].

Recently we have shown that optomechanical oscillator (OMO) can function as a compact, all-optical and low-power mass sensor [13]. Because of the high quality factor ($Q$-factor) of the circulating optical modes inside an OMO, a sub-mW optical beam can simultaneously function as an actuator and a sensitive probe. Beyond simplicity and low-power consumption the narrow linewidth (low-noise) of the OMO enables high-resolution frequency tracking and therefore high sensitivity mass measurement.

An optomechanical resonator is simply a high quality factor (high-$Q$) optical microcavity that also supports high-$Q$ mechanical modes that can efficiently modulate the optical path length [14]. In a high-$Q$ optomechanical resonator, the large circulating optical power results in a strong coupling between the optical and mechanical modes through radiation pressure. The interplay between resonant optical wavelength and the mechanical deformation may result in optomechanically induced gain or loss for the motion of the corresponding mechanical mode (depending on pump laser wavelength detuning) [14]. In the blue detuned regime ($\Delta \lambda = \lambda_{res} - \lambda_{laser} > 0$, $\lambda_{res}$: resonant wavelength of the optical mode, $\lambda_{laser}$: laser wavelength) above certain threshold power ($P_{th}$) the optomechanical gain cancels the mechanical loss and self-sustained optomechanical oscillation ensues [15–18]. Consequently the transmitted optical power (coupled out of microresonator) will be modulated at the mechanical oscillation frequency with an amplitude proportional to optical input power (pump power). So in an optomechanical oscillator (OMO) the optical power serves both as an efficient actuator and sensitive probe for monitoring the resonant frequencies of the oscillating mechanical modes. The optomechanical oscillation frequency ($f_{OMO}$) is almost equal to the frequency of the corresponding mechanical mode ($f_0$) and therefore depends on the size, geometry and material properties of the cavity structure [16]. The sensitivity slope for mass sensing can be defined as $\eta = |\Delta f_0/\Delta m|$, where $\Delta f_0$ is the frequency shift of the corresponding mechanical mode due to the added mass $\Delta m$. Accordingly, the sensing resolution is limited by $\eta$ and the resolution of frequency shift ($\Delta f_0 \sim \Delta f_{OMO}$) measurement. While $\eta$ is determined by the dimensions of the sensor [12], position of the mass and the selected mechanical mode, the minimum resolution for frequency measurement ($\Delta f_{OMO, min}$) is fundamentally limited by the linewidth of the mechanical oscillation ($\delta f_0$) and frequency jitter. The extremely narrow linewidth ($\delta f_0$) of OMO [16,19] makes it a high-resolution mass sensor [13].

It is worth pointing out that in the absence of the optomechanical interaction, high-$Q$ modes of optical microresonators have been extensively used for molecule detection and quantification based on effective refractive index change induced by molecular polarization [20]. Basically direct interaction between molecules and the evanescent field of the optical mode results in a detectable resonant optical wavelength shift proportional to the total number of interacting molecules. In contrast, in an optomechanical oscillator the particles/molecules interact with the mechanical mode that is activated and interrogated by a high-$Q$ optical mode. In an optomechanical mass sensor the evanescent optical field does not play any role and the optical mode is completely isolated from the particle/molecules (this is an advantage,
because direct optical interaction degrades the optical-$Q$ and therefore the performance of the device.

Here, we demonstrate sub-pg mass sensitivity of relatively large microtoroid OMOs enabled by frequency multiplication in optical domain. We also present a detailed analysis of optomechanical mass sensing mechanism and identify three possible operational modes. Our calculations show that when driven by a low noise and stable optical source, even large OMOs (diameter ~100 μm) can reach femtogram (fg) level mass sensitivity.

We have chosen the silica microtoroid OMO for our study because of its low phase noise, large capture area and well-studied optomechanical oscillatory behavior [15–18]. Recently monolithic micro/nano scale optomechanical oscillators have been proposed and demonstrated [21–25], however all of them have larger phase noise (due to the smaller effective mass) and relatively small capture areas. Also most nano-scale OMOs have very low-mechanical quality factor in air and can only oscillate in vacuum environment. The large optical-$Q$ of silica microtoroidal resonator not only reduces the optomechanical threshold power but also enables high sensitivity monitoring of the mechanical modes [26]. Even in atmospheric pressure and room temperature silica microtoroid OMO have relatively low threshold power and very narrow oscillation line-width (sub-Hz linewidth has been reported with only 300 μW of optical input power [16]). Compared to other OMO cavities, the silica microtoroid has a large sensitive area that is crucial for mass detection and measurement. Finally the fabrication process of silica microtoroids is relatively simple [27] compared to other OMOs.

2. Experimental results

Figure 1(a) shows the experimental arrangement used for mass measurement using microtoroid optomechanical oscillator. Optical input power is provided by a near-IR tunable laser (λ~1550 nm) that is coupled into and out of the microtoroid cavity using silica fiber-taper. When the laser wavelength is blue detuned (Δλ > 0) and the input power exceeds the optomechanical oscillation threshold power (i.e. $P_{in} > P_{th}$), the amplitude of the transmitted optical power will be modulated at $f_{OMO}$ with a modulation depth proportional to $P_{in}/P_{th}$ [17].

Upon detection in a photodetector the optical signal generates an electric signal that is measured and analyzed using an RF spectrum analyzer. Figure 1(b) shows the SEM image of one of the silica microtoroids used in this study. This microtoroid has a major diameter of $D = 131$ μm, minor diameter of $d = 7.4$ μm. The silica membrane is 2 μm thick and is attached on top of a silicon pillar with a diameter of $D_p = 5.2$ μm. The inset in Fig. 1(b) shows a polyethylene microbead landed on the toroidal section of the microresonator.

![Figure 1(a)](image)

**Fig. 1.** (a) Experimental arrangement used for mass measurement using microtoroid optomechanical oscillator. (b) SEM image of one of the silica microtoroids used in this study. $D = 131$ μm, $D_p = 5.2$ μm and $d = 7.8$ μm. Inset: a polyethylene microbead landed on the toroidal region of the resonator. (c) Schematic diagram showing the cross-section of a microtoroid, mechanical deformation of the silica membrane (associated with the 3rd mechanical mode), and the trajectory of the circulating WG optical mode (red line).

Figure 1(c) shows a schematic diagram of the microtoroid cross-section of the silica membrane deformation associated with 3rd mechanical mode, and the trajectory of
Whispering-Gallery (WG) optical mode (red line). Radiation pressure is applied to the structure in radial direction (blue arrows). To study the effect of the added mass on $f_{\text{OMO}}$, we have placed polyethylene microspheres (with red fluorescence) with diameters ranging from 0.5 µm to 2 µm and masses between 0.1 – 4 pg on the silica microtoroid OMOs. The microbeads are placed using a silica probe with spherical tip attached to a piezo micromanipulator. The silica microsphere was covered by micro-beads and then put in contact with the microresonator resulting in random distribution of micro-beads on the silica microdisk.

Fig. 2. (a) Measured RF spectrum (RBW = 30 Hz) of the OMO optical output power in the absence of the microbeads. For this OMO $D = 131$ µm, $D_p = 5.2$ µm and $d = 7.8$ µm and $f_{\text{OMO}} = 8.505$ MHz. Here: $Q_0 = 6.8 \times 10^7$, $Q_{\text{tot}} = 6.72 \times 10^6$, $Q_{\text{mech}} = 1300$, $P_{\text{in}} = 553.7$ µW, $P_{\text{th}} = 2.4P_{\text{in}}$ and $\Delta \lambda_{\text{m}} = 0.56$ ($\Delta \lambda_{\text{m}} = \Delta \lambda/\delta \lambda$, where $\delta \lambda = \lambda_{\text{res}}/Q_{\text{tot}}$). (b) Measured RF spectrum of the OMO in the vicinity of the fundamental frequency as microbeads are added sequentially (2 through 5). (c) Images of the distribution of microbeads on the microtoroid OMO. (d) SEM image of the microtoroid OMO and the microbeads distributed on it (the distribution corresponds to c-5). (e) FEM modeling of the mechanical deformation of the corresponding mechanical mode; the contours show the total displacements. (f) Measured RF spectrum (RBW = 30 Hz) of the OMO in the vicinity of the fifth harmonic frequency as microbeads are added sequentially (1 through 5). (g) Measured frequency shift of the fundamental oscillation $\Delta f_{\text{OMO}}$ and its 5th harmonic $\Delta (5f_{\text{OMO}})$ plotted against the loaded mass. The dashed red line depicts the calculated results using numerical modeling. The inset is a close-up view for $\Delta m< 5$ pg.
We used two distinct mechanical modes excited on two different microtoroids to evaluate the mass sensitivity of OMO and the enhancement obtained by monitoring optically generated harmonics. The wavelength detuning ($\Delta \lambda$) was kept constant during all measurements. Figure 2(a) shows the measured RF spectrum of the first OMO in the absence of external mass. The higher harmonics are generated by the nonlinear oscillatory optical transfer function of the optical microresonator as described in Ref. 28. Basically the oscillation of the Lorentzian transfer function at $f_{OMO}$ (stimulated by optomechanical interaction) modulates the transmitted optical power at $f_{OMO}$ and its harmonics ($n \times \Delta f_{OMO}$, $n = 1, 2, 3...$). Figure 2(b) shows the behavior of the RF spectrum of the detected optical power near the fundamental optomechanical oscillation frequency ($f_{OMO} = 8.505$ MHz) as micro-beads are loaded onto the OMO. For all spectral measurements the resolution bandwidth (RBW) of the RF spectrum analyzer is set to 30 Hz. Figure 2(c) shows the top-view micrograph of the microtoroid as the microbeads (red polyethylene microspheres) are randomly distributed on the microtoroid in four steps (1 is bare microtorid and 2-5 correspond to the measured spectrums and frequency shifts in Fig. 2(b), 2(f) and 2(g)). Figure 2(d) shows the corresponding Scanning Electron Microscope (SEM) images of the microtoroid and the added microbeads. The deformation of the corresponding mechanical mode (estimated by finite element modeling) is shown in Fig. 2(e). Figure 2(f) shows the measured RF spectrum of the OMO in the vicinity of the fifth harmonic frequency as microbeads are added sequentially (see Fig. 2(c)). A mass induced mechanical frequency shift of $\Delta f_{OMO}$ is translated to optical amplitude modulation frequency shift of $n \times \Delta f_{OMO}$ (where $n \geq 2$ is the harmonic component whose shift is measured). Since the linewidth of the oscillation is almost equal for all harmonics [28], the intrinsic optical harmonic generation effectively functions as an amplifying mechanism that enhances the frequency shift. Using larger optical input power and optimized detuning higher harmonics can be excited and monitored. Figure 2(g) shows the measured optomechanical oscillation frequency shift of the $f_{OMO}$ and its fifth harmonic plotted versus the added mass. The values of the added mass are estimated using the mass of the microbeads and their distribution obtained from the SEM and optical images after each step (as show in Fig. 2(d)). The mass of each microbead is calculated from the density of the corresponding material (polyethylene) and the volume of each microbead is estimated from the SEM image. The nonlinear behavior is due to the fact that mass is not added on a specific location of the disk (see section 3). The average sensitivity of the fundamental oscillation frequency to mass variation is about 266 Hz/pg and is in good agreement with the simulated results using finite element (FEM) modeling (red circles connected with a dashed line). As expected the sensitivity of the 5th harmonic is about five times larger (1380 Hz/pg).

Figure 3(a) shows the top-view photograph of another microtoroid as microbeads are deposited on its surface in 3 steps. This microtoroid has a major diameter of $D = 133$ µm, pillar diameter of $D_p = 11.25$ µm and minor diameter of $d = 7.24$ µm. On this microtoroid we have excited a mechanical mode with a fundamental optomechanical oscillation frequency of $f_{OMO} = 24.88$ MHz. Figure 3(b) shows the measured optomechanical oscillation frequency shift of the fundamental and its 5th harmonic plotted versus the added mass. The sensitivity of the fundamental oscillation frequency to mass variation is about 72 Hz/pg and is in good agreement with the simulated results using FEM simulation (red circles connected with a dashed line). The sensitivity of the 5th harmonic is about five times larger (~334 Hz/pg). The inset shows FEM modeling of the deformation of the corresponding mechanical mode. Figure 3(c) and 3(d) show the RF spectrum of the detected optical power near the fundamental optomechanical oscillation frequency (24.88 MHz) and its 5th fifth harmonic (~124.4 MHz) as microbeads are loaded onto the OMO.
Fig. 3. (a) Top view images of the distribution of microbeads on the second microtoroid OMO. For this OMO $D = 133 \ \mu m$, $D_p = 11.25 \ \mu m$, $d = 7.24 \ \mu m$ and $f_{OMO} = 24.88 \ \text{MHz}$, (b) Frequency shift of the fundamental oscillation $\Delta f_{OMO}$ and its 5th harmonic $\Delta(5f_{OMO})$ plotted against the loaded mass. The dashed line depicts the calculated results using numerical modeling. The inset shows FEM modeling of the mechanical deformation of the corresponding mechanical mode. (c) and (d) are the measured RF spectrum ((RBW = 30 Hz)) of the OMO in the vicinity of the fundamental frequency (c) and its fifth harmonic (d) as microbeads are added sequentially (1 through 4). Here: $Q_0 = 3.6 \times 10^6$, $Q_{tot} = 2.3 \times 10^6$ ($Q_{tot}$ is the loaded optical-$Q$), $Q_{mech} = 1466$, $\Delta \lambda_N = 0.58$ ($\Delta \lambda_N = \Delta \lambda / \delta \lambda$, where $\delta \lambda = \lambda_{res} / Q_{tot}$), $P_{in} = 212 \ \mu W$, and $P_{in} = 3.67$ $P_{th}$.

3. Analysis of mass sensing with optomechanical oscillation

The mass sensitivity slope of an OMO ($\eta = |\Delta f_{OMO}/\Delta m|$) is different for each mechanical mode and depends on the location of the added mass [29, 30]. The modal and position dependence of $\eta$ suggests that the optimal design for the OMO mass sensor varies for three different applications: 1) lumped mass measurement, 2) lumped mass detection and 3) distributed mass detection/measurement. For lumped mass measurement the OMO is used to measure the mass of a particle that can be placed at a desired position on the OMO. In this case for best resolution the OMO dimensions, excited mechanical mode (labeled by $i$) and the position of the particle should be selected such that $\eta_i(r, \theta)$ is maximized. For single particle detection the distribution of the particles on the sensor surface is unknown (because they randomly fall on different locations) and maximum probability of detection is desired. Therefore the OMO dimensions and the excited mechanical mode should be chosen to provide the largest area that translates the presence of the particles into a measurable frequency shift. Finally for detection and measurement of distributed mass, which is almost uniformly distributed over the OMO, the OMO dimensions and the selected mechanical mode should result in maximum oscillation frequency shift for a given surface density. For the theoretical study we focus on four mechanical eigen-modes of the silica microtoroid OMO with $D = 133 \ \mu m$, $D_p = 11.25 \ \mu m$, $d = 7.24 \ \mu m$ (similar to the one used in Fig. 3). Figure 4(a) shows the mechanical deformation of these modes (calculated using finite element modeling). These mechanical modes are chosen because they are the most commonly observed oscillating modes and have the lowest threshold powers (based on our experiment and previous studies [16,17]). Using first order perturbation theory and dynamic equations for an elastic medium, it can be shown that the frequency shift due to added mass with a magnitude $\Delta m$ at the position $r$ and $\theta$ on the microdisk can be written as:
\[ \frac{\Delta f_{\text{OMO}}}{f_{\text{OMO}}} = -f_{\text{OMO}} \frac{\Delta E_{\text{total}}}{2E_{\text{total}}} = -\pi^2 f_{\text{OMO}}^3 \frac{\Delta m(r,\theta)U^2(r,\theta)}{E_{\text{total}}} \]  

where \( E_{\text{total}} \) is the total energy stored in the OMO, \( \Delta E_{\text{total}} \) is the energy change due to external mass, \( \Delta m(r,\theta) \) is the external mass at position \((r,\theta)\), \( U(r,\theta) \) is the maximum total displacement (in three dimensions) at point \((r,\theta)\). Note that here we assume the external mass is positioned on the flat surface of the vibrating plate (so that \( z \) is constant). It is assumed that the added mass is so small that does not affect the mode shapes and its moment of inertia is negligible (i.e. nodal mass). Equation (1) implies that the modes with more localized displacement have better sensitivity. In other words for a given value of \( E_{\text{total}} \), \( U \) can be large only near certain position or very small all over the entire disk surface. We have calculated \( \eta \) using two methods. 1) **Energy Method** employing Eq. (1) where all the required parameters, that are independent of the added mass, of the passive mechanical resonator (i.e. \( f_{\text{OMO}}, U \) and \( E_{\text{total}} \)) are calculated with finite element modeling (FEM). 2) **Direct FEM** modeling of \( f_{\text{OMO}} \) before and after adding the mass and subsequent extraction of the frequency shift. The simulation results in Fig. 2(g) and 3(b) are calculated using the FEM method (considering the measured radial and azimuthal position of the all particles) verifying that outcomes of this method are in good agreement with experimental results. The FEM method takes into account possible perturbation of the mode shape due to added mass while the Energy method assumes \( U(r,\theta) \) is independent of added mass.

![Fig. 4. (a) FEM simulation of mechanical modes of a silica microtoroid. (a) Deformation of four eigen mechanical modes of a silica microtoroid with a pillar diameter \( D_p = 11.2 \) µm major diameter \( D = 133 \) µm, and minor diameter \( d = 7.4 \) µm. \( f_0 \) is the eigen frequency of the corresponding mode. (b) \( \eta \) plotted against radial position of mass \((0 = 0)\) for the four modes shown in part (a) using the FEM and Energy method. Compared to other modes, the \( \eta \) for the first mode is so small that appears as a straight line on the x-axis.](image)

Figure 4(b) shows the frequency shift for 1 pg mass (that is equal to sensitivity slope \( \eta \) with the unit of Hz/pg) plotted against radial position of mass (along \( 0 = 0 \) direction) for the modes shown Fig. 4(a) using both the **Energy** and **FEM** Methods. The sensitivity of the first mode is negligible compared to the other modes both because the displacement (mechanical
energy) is distributed all over the disk and the oscillation frequency is very small. As expected because 1 pg is small compared to the resonator mass \( m_r = 6 \times 10^4 \) pg, the outcomes of the two methods are very close. The agreement between the two methods justifies the use of Energy method that significantly simplifies the calculation processes for multiple masses at different positions. In addition, validity of the Energy method verifies the linearity of the response when \( \Delta m \ll m \), and the mass is added at a specific location (because based on Energy method \( \eta \) only depends on the location of the added mass and not its magnitude).

For mass measurement (where the particle under test can be placed at a desired position), the mode order \( i \) and optimal position \( (r_m, \theta_m) \) should be chosen such that the slope sensitivity is maximized (i.e. \( \eta_{\text{max}} = \eta_i(\pi, \theta_m) \)). According to Fig. 4(b) for the microtoroid in Fig. 2, the best choice is the 3rd mode and the best position is \( r = 17 \) \( \mu \)m (with an arbitrary value of \( \theta \) due to axial symmetry of this mode). Near this specific position \( \eta_3 \approx 500 \) Hz/pg for the fundamental oscillation and \( n \times 500 \) Hz/pg when the frequency of the \( n \)th harmonic is measured. Figure 5(a) shows \( \eta_{\text{max}} \) plotted against \( D/D_P \) for two different values of \( D \) and the three selected eigen modes \( (i = 1, 2, 3, 4) \). Generally \( \eta_{\text{max}} \) increases as \( D \) is decreased and for a given \( D \), shows an asymptotic behavior as a function \( D/D_P \) reaching a saturated value for \( D/D_P > 10 \). As \( D/D_P \) increases, a larger portion of energy is stored in the disk (\( U \) is larger) but the resonant frequency \( f_{\text{OMO}} = f_0 \) is reduced. According to Eq. (1), \( \Delta f_{\text{OMO}} \propto f_{\text{OMO}}^3 \times U^2 \) so these two effects balance each other for large values of \( D/D_P \).

Note that as expected this study suggests that the oscillation frequency of smaller OMOs and those with smaller effective mass, e.g. the zipper photonic crystal [23] and spoke-supported silica microtoroid or silicon nitride microrings [22] should be more sensitive to the added mass compared to the silica microtoroid. However, loading mass to the most sensitive location of these OMOs is a difficult task and the presence of the particle interferes with the resonant optical field degrading the optical-Q to a level that quenches the optomechanical oscillation. Moreover the smaller effective mass of these devices results in larger phase noise (larger oscillation linewidth) that limits the smallest measurable frequency shift.

For lumped mass detection, the goal is to detect the existence of added mass on the OMO. Therefore the capture probability should be maximized while maintaining the induced frequency shift above the minimum detectable frequency change (i.e. \( \Delta f_{\text{OMO}} \)). \( \Delta f_{\text{OMO}} \) is limited by different noise mechanisms and the resolution of RF spectrum analyzer (see Section 4). For a given mass change \( \Delta m \), we define the sensitive area \( (A_s) \) using the condition \( f_{\text{OMO}} > \Delta f_{\text{OMO}} \). Figure 5(b) shows \( A_s \) plotted against \( D \) for the three eigen-modes (2-4) when pillar diameter is \( D_P = 11.5 \) \( \mu \)m, particle mass is \( \Delta m = 1 \) pg and \( \Delta f_{\text{OMO}} = 40 \) Hz.
(chosen based on the limitation of our measurement system). For each eigen mode an optimized diameter ($D_{opt}$) can be identified that maximizes $A_s$. The inset shows $D_{opt}$ plotted versus mass value ($\Delta m$) suggesting for detection of heavier particles, larger microtoroids provide larger sensitive capture area.

Finally, the microtoroid OMO can be used for detection of uniformly distributed mass on its surface. This capability is effectively translated to measuring the surface density ($\rho_s$) of gas molecules that are adsorbed on the OMO surface. Since in thermodynamic equilibrium $\rho_s$ is proportional to the concentration of the molecules in the surrounding medium, one can extract the volume density from oscillation frequency shift. Note that in the case of biomolecules the adsorption probability can be significantly enhanced by functionalizing the surface (e.g. an organic monolayer deposited on top of the microtoroid). However at large densities, when the molecules are close enough to interact with each other, the intermolecular forces affect the resonant frequency through surface stress effects. As shown in the context of cantilever sensor, in this case both surface stress and mass effects should be taken into account [2]. For the sake of simplicity here we only consider a low-density but uniform distribution of non-interacting particles (ignoring surface stress effect). For a given microtoroid and mechanical mode Eq. (1) can be modified as:

$$\Delta f_\text{OMO} = -\pi^2 f_\text{OMO} \frac{\rho(r,\theta)\Delta a U^2(r,\theta)}{E_{\text{total}}}$$

where $\rho(r, \theta)$ is surface mass density at position $(r, \theta)$ on the microdisk and $\Delta a$ is the area covered by the external mass. To evaluate the contribution of the whole capture area, the figure of merit $\Lambda$ is defined as:

$$\Lambda = \frac{\pi^2 f_\text{OMO}^3}{E_{\text{total}}} \int_a \rho(r,\theta)U^2(r,\theta) da$$

$\Lambda$ has units of Hz and represents the total frequency shift due to the surface density $\rho_s$, $\Lambda$ also contains the effects of microtoroid dimensions (which directly defines the capture area), frequency of oscillation and mechanical deformation (for a specific mode) for a particular mass distribution.

$$\Delta \Lambda = -\pi^2 f_\text{OMO} \frac{\rho(r,\theta)\Delta a U^2(r,\theta)}{E_{\text{total}}}$$

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Figure 6 shows $\Lambda$ is plotted against $D/D_p$ for two different values of $D$ and the four eigen modes.

Figure 6 shows $\Lambda$ is plotted against $D/D_p$ for two different values of $D$ and the three selected eigen modes (2,3,4) as shown in Fig. 4(a), assuming a uniform mass distribution with a constant surface mass density of $\rho_s = 0.1$ ng/mm$^2$. As evident from the graph increasing $D/D_p$ beyond 10 does not affect $\Lambda$. 

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In conclusion depending on the desired mode of operation, the optimized dimensions of the microtoroid sensor correspond to maximum \( \eta_{\text{max}} / \Delta f_{\text{OMO}-\text{min}} \) (for lumped mass measurement), \( \Lambda / \Delta f_{\text{OMO}-\text{min}} \) (for detection/measurement of distributed mass) or \( \Lambda / \Delta f_{\text{OMO}-\text{min}} \) (for detection of lumped mass).

4. Ultimate performance of optomechanical mass sensor

The minimum mass resolution for an OMO sensor (for lumped mass measurement mode) is limited by the maximum mass sensitivity slope \( \eta_{\text{max}} \) and the minimum detectable frequency shift \( \Delta f_{\text{OMO}-\text{min}} \) through:

\[
\Delta m_{\text{min}} = \frac{\Delta f_{\text{OMO}-\text{min}}}{\eta_{\text{max}}} \tag{4}
\]

\( \Delta f_{\text{OMO}-\text{min}} \) is limited by OMO frequency noise and the resolution of frequency shift measurement instrument \( (\delta f_i) \). In classical regime, optomechanical oscillation frequency noise \( (\delta f_{\text{OMO}}) \) originates from fluctuations of the circulating optical power \( \delta (P_{\text{circ}}) \) and the thermo-mechanical (Brownian) noise. As shown before \([16,19]\) at room temperature Brownian noise \( (\propto k_B T) \) and mechanical quality factor of the corresponding mode \( (Q_m) \) set the lower limit for optomechanical oscillation linewidth \( (\delta f_i) \), while \( \delta (P_{\text{circ}}) \) generates oscillation frequency fluctuations through optical spring and opto-thermal effects \( (\Delta f_p = \Delta f_S + \Delta f_T) \) \([16]\). So the \( \Delta f_{\text{OMO}-\text{min}} \) can be expanded as:

\[
\Delta f_{\text{OMO}-\text{min}} = \delta f_i + \delta (\Delta f_p) + \delta f_i \tag{5}
\]

where \( \Delta f_p \) (\( = f_{\text{OMO}} - f_0 \)) is the shift due to the “optical spring effect” and “thermal effect” \([16]\). \( \Delta f_p \) translates the fluctuation of input parameters, \( \Delta f_N \) \( (\Delta f_N = \Delta f_S + \Delta f_T) \), where \( \delta f_S \) is the loaded linewidth of the corresponding optical mode, and \( P_{\text{in}} \), to fluctuations of \( f_{\text{OMO}} \) by affecting the circulating optical power inside the cavity. Although in our experiment \( \delta (Q_{\text{tot}} / Q_0) \) also contributes to \( \delta (\Delta f_p) \) (due to uncertainties in the coupling gap controlled by piezo stage), in new integrated OMOs \([21–25]\) this contribution is negligible. For given values of \( \delta (\Delta f_N) \) and \( \delta P_{\text{in}} \), \( \delta (\Delta f_p) \) depends on the microtoroid dimensions \([16]\). Note that the photodetection noise (i.e. shot noise, dark noise, and thermal noise) have negligible impact on the precision and resolution of the frequency monitoring.

The harmonic components \( (f_{\text{OMO}}^{(n)} = n \times f_{\text{OMO}}) \) are generated in the optical domain by the oscillatory transfer function of the cavity \([28]\), so their oscillation linewidth that is limited by the thermo-mechanical noise \([16,19]\) are almost equal to that of the fundamental oscillation \( (\delta f_L) \). Note that here we are ignoring the noise associated with the optical multiplication process because it is negligible compared to the thermo-mechanical noise. So When the \( n^{th} \) optically generated harmonic of the OMO is used for mass sensing, only \( \Delta f_p \) changes and Eq. \( (5) \) can be modified as:

\[
\Delta f_{\text{OMO}-\text{min}}^{(n)} = \delta f_L + n \times \delta (\Delta f_p) + \delta f_i \tag{6}
\]

The corresponding minimum detectable mass is:

\[
\Delta m_{\text{min}} = \frac{\delta f_L + \delta f_L + \delta f_p}{n \times \eta_{\text{max}}} \frac{1}{\eta_{\text{max}}} \tag{7}
\]

Using lasers with small relative intensity noise (RIN \(<100 \text{ dB/Hz}\)) and assuming \( \delta (Q_{\text{tot}} / Q_0) = 0 \), \( \delta (\Delta f_p) \) will be only limited by \( \delta (\Delta f_N) \). Modern frequency stabilization techniques that have enabled laser frequency fluctuation as small as 1 Hz \([31]\) combined with wavelength locking techniques, such as Hänsch–Couillaud \([32]\) or Pound–Drever–Hall \([33]\) methods, can easily keep \( \delta (\Delta f_N) \) below 500 Hz. Such level of stability results in \( (\Delta f_p) \sim 0.1 \text{ Hz} < \delta f_L \) enabling mass...
measurement/detection sub-fg resolutions (limited only by optomechanical oscillation linewidth).

Figure 7(a) shows $\Delta m_{\text{min}}$ plotted against $\Delta \nu_N$ for the microtoroid in Fig. 2(a) when $\delta(\Delta \nu_0) = 500 \text{ Hz}$. $\delta(Q_{\text{out}}/Q_0) = 0$ and $\delta f = 0$. Here the mass is located at the position of the maximum sensitivity ($\eta_{\text{max}} = \eta(t_m \theta_m)$) for each mode. By fixing the normalized detuning ($\Delta \nu_N$) at its optimal value (~0.28), in Fig. 7(b) we have plotted $\Delta m_{\text{min}}$ against the order of harmonic frequency used for the measurement ($n$) for two cases: $\delta f = 0$ and $\delta f = 30 \text{ Hz}$. As evident from this plot measuring the shift of the 7th harmonic frequency (instead of the fundamental frequency) can improve the sensitivity by almost one order of magnitude.

![Figure 7](image)

Fig. 7. (a) $\Delta m_{\text{min}}$ plotted against normalized detuning $\Delta \nu_N$ for the fifth harmonic ($n = 5$) of the mechanical modes of the microtoroid in Fig. 2 ($D = 131 \mu m$, $D_b = 5.2 \mu m$). The mass is located at the position of maximum sensitivity ($\eta_{\text{max}} = \eta(t_m \theta_m)$) for each mode. Here $Q_b = 6.8 \times 10^7$, $Q_{\text{max}} = 1300$, $Q_{\text{out}}/Q_b = 0.1$ and $P_b = 2 P_b$. We have assumed $\text{RIN} < 90 \text{dB}$, $\delta(\Delta \nu_0)$ is 500 Hz and ignored $\delta(Q_{\text{out}}/Q_0)$ and $\delta f$. (b) $\Delta m_{\text{min}}$ plotted against the order of harmonic frequency used for the measurement ($n$) for two cases: $\delta f = 0$ dashed lines and $\delta f = 30 \text{ Hz}$ (solid lines). The insets show the membrane deformation for the 2nd and 4th modes. The mass is located at the position of maximum sensitivity ($\eta_{\text{max}} = \eta(t_m \theta_m)$) for each mode shown as red zones in the insets. $\Delta \nu_N$ is fixed at 0.28 that is the optimal detuning according to part-a. Other parameters are the same as the ones in part-a. (c) $\Delta m_{\text{min}}$ plotted against the $D/D_b$ based on the fifth harmonic shift, for $D = 103 \mu m$ (dashed lines) and 133 $\mu m$ (solid lines). Here all parameters are chosen according to the actual experiment. $\Delta \nu_N = 0.56$, $Q_b = 6.8 \times 10^7$, $Q_{\text{max}} = 1400$, and $P_b=2 P_b$. $Q_{\text{out}}/Q_b = 0.1$. Uncertainty of $Q_{\text{out}}/Q_b$ and detuning $\delta(\Delta \lambda)$ are 1% and 400 KHz, respectively. $P_b = 2P_b$, $\delta f = 30 \text{ Hz}$.

Figure 7(c) shows $\Delta m_{\text{min}}$ plotted against the $D/D_b$ based on fifth harmonic shift, for $D = 103 \mu m$ (dashed lines) and 133 $\mu m$ (solid lines). Here all parameters are chosen according to the actual experiment. $\Delta \nu_N = 0.56$, $Q_b = 6.8 \times 10^7$, $Q_{\text{max}} = 1400$, and $P_b=2 P_b$. $Q_{\text{out}}/Q_b = 0.1$. Uncertainty of $Q_{\text{out}}/Q_b$ and detuning $\delta(\Delta \lambda)$ are 1% and 400 KHz, respectively. $P_b = 2P_b$, $\delta f = 30 \text{ Hz}$. In principle using our measurement system and the OMO shown in Fig. 2 a mass detection limit of 150 fg ($150 \times 10^{-15}$) is within reach.

Note that in electro-optomechanical oscillator driven by electro-mechanical force through external feedback [9], $\delta(\Delta f_0)$ may become negligible by reducing the optical power resulting to a fundamental sensitivity limited by thermal noise. However the impact of the feedback noise and frequency pulling mechanisms should be taken into account in these devices.

5. Conclusion

This work lays the foundation for a new class of mass sensors that can be used for characterization and detection of particles and molecules. Beyond all-optical operation the main advantage of OMO mass sensor is the simple and low-noise actuation and read-out mechanisms based on optical force and resonant optical path length modulation. Optical read-out based on high-$Q$ optical modes enables monitoring higher order mechanical modes with minimal out of plane displacement that are usually difficult to monitor using other techniques. The fact that due to reduced viscous damping high order mechanical modes have larger...
mechanical-$Q$s in air, enables narrow linewidth oscillation and therefore high sensitivity in atmospheric pressure. In principle all newly introduced integrated and nano-scale OMOs can be used as ultra-sensitive mass sensors, however the large capture area, narrow linewidth oscillation in air (due to larger effective mass) and simple fabrication method make the microtoroidal OMO a better candidate for practical applications. High-$Q$ optical modes can be also used to excite and interrogate resonant surface acoustic waves (as opposed to bulk mechanical modes) [34]. Although these modes may be used for mass sensing, their confinement near the small optical mode volume results in very small sensitive area.

We have shown the intrinsic harmonic optical modulation inside OMO can effectively serve as a low-noise amplification mechanism. Basically, independent of the specific mechanical mode used for sensing the induced shift for the corresponding $n$th harmonic frequency will be $n$ times larger than $\Delta f_0$ for a given mass and position. In principle this approach can be applied to all kinds of OMO based mass sensors to increase the slope response and signal-to-noise ratio (SNR) for frequency shift measurement and therefore high-resolution mass sensing. We have identified three distinct operational modes of the microtoroidal OMO sensor for lumped and distributed mass detection/quantification. Our analysis shows using low noise lasers and detectors combined with advanced laser locking mechanisms, microtoroidal OMO can serve as a mass sensor with fg level resolution for metrological applications.

Although the ultimate sensitivity of the mass sensor presented here is lower than the record sensitivities obtained for state-of-the-art nano-scale cantilever mass sensors, the microtoroid OMO’s simplicity, all-optical excitation/read-out and operation in atmospheric pressure make it a useful tool for applications where ultra-low sensitivity is not required. MEMS/NEMS mass sensors are fundamentally limited by the complexity and speed of the excitation mechanism (piezo, electro-static or thermal). The transducers of such sensors require relatively complex co-fabrication of the hybrid material systems integrated with a high-$Q$ mechanical resonator resulting in high cost [35]. Furthermore, piezoelectric and electrostatic sensors may have a limitation when working in the presence of strong electromagnetic fields. In contrast OMO based sensors have a relatively simple structure and material system and their all-optical operation make them immune to electromagnetic interference (EMI). An additional concern is that nano/microscale cantilever mass sensors commonly operate using out-of-plane vibrations that facilitate the read-out (specifically piezo-resistive techniques that rely on bending induced strain). These modes are affected by viscous damping when operating at atmospheric pressure and therefore have considerably lowered quality factors (approximately by factor of 10 or more; below 1000) [36–38]. However the displacements of microtoroid OMO based sensors are mostly in-plane and as such display higher mechanical quality factors, typically above 2000 [18], leading to a higher sensitivity at atmospheric pressure.

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