Nanoscale welding aerosol sensing based on whispering gallery modes in a cylindrical silica resonator

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Abstract: We report an experimental technique where one uses a standard silica fiber as a cylindrical whispering gallery mode (WGM) resonator to sense airborne nanoscale aerosols produced by electric arc welding. We find that the accumulation of aerosols on the resonator surface induces a measurable red-shift in resonance frequency, and establish an empirical relation that links the magnitude of resonance shift with the amount of aerosol deposition. The WGM quality factors, by contrast, do not decrease significantly, even for samples with a large percentage of surface area covered by aerosols. Our experimental results are discussed and compared with existing literature on WGM-based nanoparticle sensing.

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1. Introduction

Recently, research on silica-based optical resonators has attracted much attention. Due to the extremely low material absorption and smooth surface, such resonators can support high quality (Q) factor whispering gallery modes (WGMs) in various geometries including microspheres [1], microtoroids [2], and cylinders [3, 4]. Under appropriate coupling conditions, the existence of these WGMs can lead to very sharp transmission dips, with resonance linewidths inversely proportional to cavity Q factors. The center resonance positions and the Q factors of such transmission dips depend sensitively on the resonator environment. Taking advantage of this feature, various WGM resonators have been utilized for a wide range of sensing applications [4–8].

The main goal of this paper is to establish the feasibility of using a cylindrical resonator for airborne nanoscale welding aerosol sensing. Studies have shown that extended exposure to welding fumes can significantly increase the risk of bronchitis, lung cancer, and other diseases [9, 10]. In current practice, to monitor the presence of airborne aerosols, one typically must use a pump and a cascade impactor to collect aerosols in sufficiently large air volume, followed by measuring the weight of the collected aerosols [11, 12]. Since the resolution of weight measurement is typically limited to 0.1 μg or above, the sensitivity of this traditional approach is quite poor for airborne nanoparticle (NP) sensing, especially in comparison with the capability of WGM-based measurements. Yet, existing literature...
suggests that smaller aerosols, especially those with diameters less than 100 nm, are likely associated with the most significant health risks [13–16].

Currently, cylindrical WGM resonators are commonly used for detecting macromolecules and cells in liquids [4, 8]. To the best of our knowledge, cylindrical resonators have never been used for airborne aerosol sensing. For practical applications, cylindrical resonators have several obvious advantages. First of all, many cylindrical resonators require almost no fabrication. We can simply strip away the buffer coating of a standard communication fiber and use it as a WGM resonator with reasonably high-Q factors. Depending on specific geometries (e.g., hollow capillary tubes or solid cylinders) and cladding materials (e.g., air or liquid), silica-based cylindrical resonators with Q factors in the range of $10^4$ to $10^8$ have been reported [4, 17, 18]. Additionally, since the structural and material parameters of standard silica fibers are extremely well controlled, the properties of these WGMs remain highly consistent. This unique feature makes it possible to quantitatively investigate the relationship between WGM resonances and aerosols attached to the cylinder surface, which is very important for sensing applications.

In current literature, cylindrical resonators with a large number of randomly attached NPs have yet to be thoroughly investigated. Originally, we expect that due to surface scattering, the WGM Q factor of the cylindrical resonator should drop significantly, as suggested by our previous work on microsphere resonators covered with multiple gold NPs [19]. However, contrary to our original expectation, the main effect of aerosol accumulation is not a significant reduction in the WGM Q factor but rather a red-shift in its resonance wavelength. In fact, a key result of this paper is an empirical relation that links the amount of resonance shift with the quantity of aerosols accumulated on the silica resonator surface. At the same time, to our surprise, we find that even for a resonator with extremely rough surface (due to the adsorbed aerosols), the resonator can still support WGMs with moderately high Q factors in the range of $5 \times 10^4$ to $6 \times 10^5$. In comparison, the Q factors of a clean cylindrical resonator typically are in the range of $6 \times 10^4$ to $7 \times 10^5$. In section 5, we discuss several factors that may account for the resonance red-shift and the relatively high Q factors in the aerosol-covered resonators. Here we simply point out that due to the lack of axial confinement, the cylindrical resonator supports not just a discrete number of WGMs but a continuum of WGMs. This feature makes cylindrical resonators fundamentally different from other types of high-Q resonators (e.g., microsphere or microtoroids).

The experimental results reported here are practically relevant and theoretically interesting. The development of nanotechnology has made monitoring the presence of nanomaterials in environment an increasingly important task [16]. The WGM sensor presented here is low cost, requires no fabrication, is simply to align and to operate, and should be significantly more sensitive than the impactor-based existing technology [11, 12]. These features are very attractive for practical applications. From a theoretical point of view, it is possible to draw analogy between our results and existing literature on wave propagation in random media [20]. For example, the presence of aerosols on the resonator surface may produce significant multiple scattering that can effectively elongate the resonator optical path, which may explain the observed resonance red-shift. Interesting phenomena such as coherent back scattering [21] may also become important under certain circumstances. A complete understanding of our experimental results likely requires applying the theoretical framework of random media scattering [20], which, to the best of our knowledge, has yet to be systematically explored in the context of high-Q WGMs.
2. Experimental method

Fig. 1. Six major steps in WGM-based aerosol sensing: (a) clean fiber measurement, (b) welding aerosol deposition, (c) characterization of aerosol-covered sample, (d) SEM analysis, (e) aerosol removal, (f) control measurement of the re-cleaned fiber sample.

Our experimental procedure for aerosol sensing contains six major steps, which, as illustrated in Fig. 1, are: Fig. 1(a) clean sample characterization (step 1), Fig. 1(b) welding aerosol deposition (step 2), Fig. 1(c) measurement of the aerosol-covered sample (step 3), Fig. 1(d) scanning electron microscope (SEM) imaging of the aerosol-covered sample (step 4), Fig. 1(e) sample cleaning / aerosol removal (step 5), and Fig. 1(f) control measurement of the cleaned fiber sample (step 6).

Clean resonator characterization in Fig. 1(a) (step 1) is carried out by measuring the transmission spectrum through a thin fiber taper (~1 µm in diameter) that is placed in direct contact with the cylindrical resonator. The transmission spectrum of the coupled taper-resonator system depends on many parameters. To minimize experimental uncertainties, we require that: (1) all transmission measurements are carried out using the same fiber taper; (2) all taper-resonator coupling measurements are performed at the same taper location, with the taper in contact with the resonator surface; (3) all WGM parameters are extracted using the same WGM; (4) the WGM characteristics of the clean fiber resonator are obtained before aerosol deposition, which allows us to account for variations in fiber diameter.

Welding aerosol deposition (step 2) is carried out using the system in Fig. 2(a). The aerosols are generated by electric arc welding, where we utilize an electric current supply (AC-225 Arc Welder, Lincoln Electronic) to produce electric arc between an electrode (E6011 Fleetweld 180-3.2mm, Lincoln Electronic) and a carbon steel plate. The welding process takes place in a custom-built acrylic plate chamber with an outlet at the top. Some of the aerosols generated by welding drift upward towards the outlet at the top and are deposited onto the resonator surface. Unlike the traditional cascade-impactor-based approach, our approach does not require any “forced” air flow for aerosol collection.

Throughout the process of aerosol collection, the fiber resonator is placed in the V-groove of a fiber holder (FHS-025, Sumitomo Electric), which itself is fixed at the chamber outlet and faces downwards, as shown in Fig. 2(a). This configuration ensures that only the outer-half of the resonator surface is available for aerosol deposition. The remaining inner-half surface is covered by the V-groove and should remain aerosol-free. This expectation is clearly
confirmed by the SEM image in Fig. 2(b), where the side view image shows both the aerosol-covered outer surface and the clean inner surface. We control the amount of aerosols deposited onto the resonator surface by varying the amount of welding rods consumed in the process, while all other parameters being the same. After aerosol deposition, we fix the aerosol-covered sample onto a glass slide before we release the sample from the fiber holder. This allows us to distinguish the aerosol-covered outer surface from the clean inner surface, which is important for taper-resonator coupling.

![Diagram](image)

**Fig. 2.** (a) Illustration of the welding aerosol deposition system. (Insets: welding chamber and arc welder at the top-right and the bottom-right corner, respectively). The fiber resonator is placed in a V-groove such that only the outer half of the resonator surface is exposed to the flow of welding aerosols. The surface of the inner half remains clean and is used for fiber taper coupling. (b) A representative SEM image of a sample covered with aerosols. The side view image shows both the dirty outer half and the clean inner half surface.

In step 3, we utilize the same fiber taper to measure the transmission spectra of the aerosol-covered samples. We require that only the aerosol-free surface can possibly touch the taper during measurements. This requirement eliminates the possibility of inadvertently transferring aerosols from the resonators to the taper, and ensure that the same taper can be repeatedly used. After WGM characterization, we cut the fiber resonator in half, remove the section that has been used in WGM measurements, and use it for SEM imaging and analysis (step 4).

Finally, we perform a control experiment with the aim of excluding the possibility, however unlikely, that changes in WGM transmission spectra might be caused by physical or compositional damages to the silica resonator (due to exposure to welding aerosols, for example). In the control experiment, we first carry out step 5, where we remove aerosols from fiber surface by wiping the sample surface with alcohol, washing it in water for 30 seconds, sonicating for 10 minutes, again washing in water for 30 seconds, drying in air for 10 minutes, and finally wiping the sample surface with alcohol. Then, in step 6, we again measure taper transmission spectrum of the cleaned control sample. If the resonator remains intact during the process of aerosol collection / WGM characterization, then the transmission spectra of the clean and the control sample should be very similar.

Clean resonator calibration is a key step in our experimental procedure. Given its importance, studies of clean fiber resonator WGMs is separately addressed in the next section.
3. Calibration of WGMs in clean silica fiber resonators

For accurate sensing, all resonator parameters must be well controlled. Given this requirement, a natural choice for the cylindrical resonator is standard single-mode fiber (SMF, Corning SMF-28), with cladding diameter precisely set at 125±0.7 µm. Since the WGM is mainly confined near the cylindrical surface and does not “see” the fiber core, we can essentially treat the SMF as a uniform dielectric cylinder with refractive index \( n \cong 1.45 \) [17].

The taper used for WGM coupling is fabricated from the same SMF-28 fiber using a heat and pull method [22]. We monitor taper transmission throughout the pulling process to ensure that taper waist is approximately 1 µm in diameter. By placing the cylindrical resonator next to the thin taper, optical wave can be evanescently coupled into the resonator WGM. To reduce variability in evanescent coupling, we always place the cylindrical resonator in direct contact with the fiber taper. A polarization controller is used to adjust incident light polarization. A CCD camera captures the taper / resonator image, as shown in Fig. 3(b). An optical spectrum analyzer (OSA, si720, Micron Optics) is used to measure taper transmission in the range of 1520-1570 nm. The OSA is equipped with an internal laser source and a photodetector in one module. The spectral resolution of the OSA and the laser linewidth (~2.5-4 pm) is sufficient for identifying cylindrical resonator WGMs with \( \sim 10^5 \) Q factors. For confirmation, almost exactly the same transmission spectra are observed using a narrow-linewidth (< 300 kHz) tunable laser (TLB-6328, New Focus).

Besides taper-resonator gap distance (which is set at zero), transmission spectra may also depend on the fiber-resonator alignment angle \( \theta \), as defined in Fig. 3(b). In [23], it has been shown that a small amount of misalignment (\( 80°< \theta <100° \)) has no significant impact on the WGM characteristics of a cylindrical resonator. We have experimentally confirmed this observation. Figure 3(c) shows three transmission spectra measured at three different alignment angles \( \theta \).
but with $\theta = 90^\circ$, $85^\circ$, and $80^\circ$, respectively. (The $\theta$ values are estimated using CCD camera images.) For the data in Fig. 3(c), a WGM resonance dip can be clearly identified near 1522.7 nm and its position remains almost the same regardless of alignment angle. For quantitative comparison, we extract the WGM resonance frequency through Lorentzian fitting of the transmission resonance dip. Based on the results in Fig. 3(c), we find that for small misalignment (i.e., $80^\circ< \theta <100^\circ$), any resonance shift due to angle misalignment is less than 5 pm, which is close to the intrinsic limit of our measurement instruments.

![Diagram](image)

**Fig. 4.** (a) A schematic illustration of taper-resonator coupling is shown in the left panel. The right panel, which corresponds to the region in the dashed square in the left panel, illustrates five taper-resonator coupling locations considered in our experiment. (b) Transmission spectra at the five coupling locations in (a). Through Lorentzian fitting, we find that the center of the resonance dip for the desired WGM is located at 1522.7869, 1522.7817, and 1522.7826 nm for the 1st, 2nd, and 3rd location, respectively.

Next, we investigate the relationship between WGM transmission spectrum and the location for taper-resonator coupling. Altogether, we consider five coupling locations along the same taper, as indicated in Fig. 4(a). The 5th coupling location is closest to the taper waist, whereas the 1st one is farthest from the taper waist and the rest are in between. In Fig. 4(b), we show five distinct transmission spectra obtained at these five locations. At the 1st location, where taper-resonator coupling is perhaps the weakest, the WGM transmission dip near 1522.7 nm is closest to the ideal Lorentzian line-shape and perhaps most suitable for aerosol sensing. At the 2nd and the 3rd coupling location, the shape of the desired WGM resonance becomes clearly asymmetric and contains significant blue-shifted component. Similar phenomena are reported in [3, 24], where the blue-shifted component is attributed to the “spiral” modes that propagate along the axial direction of the dielectric cylinder. In contrast, at the 4th and 5th location, we can no longer clearly identify the WGM resonance near 1522.7 nm. Given the results in Fig. 4, the 1st coupling location is used for all subsequent measurements. To enforce this requirement, we simply mark the desired coupling location on the CCD camera image. As long as we do not move the microscope objective, the CCD camera, or the fiber taper, we only need to bring the cylindrical resonator to the marked position to ensure that the 1st location is used for taper-resonator coupling.

Since the diameter of the SMF-28 fiber slightly varies [25], we need to investigate its impact on WGM properties. For this study, we evenly divide a 2-meter-long fiber into 29 samples, with each sample is ~7 cm in length. For each section, we remove ~2 cm fiber coating and expose the silica surface. We then apply the aforementioned cleaning procedure (step 5) in Fig. 1(e) to prepare clean samples for transmission measurements. For proper statistical analysis, for every cylindrical resonator, we record transmission spectra at 20 random spots within an end-side 1 cm long segment marked in Fig. 5(a) and consider only the
WGM near ~1522.3 nm. In Fig. 5(b), we show four representative clean sample transmission spectra, obtained using samples at four different spool locations (approximately 21, 70, 126, and 147 cm away from the 1st sample, respectively). Compared with the WGM resonance in the 1st sample, the four WGM resonances in Fig. 5(b) are shifted by −5.5, −85.6, −16.7, and −81.3 pm, respectively. The overall spectral shapes, however, are almost exactly the same. For the 1st sample at 7 cm, we extract the resonance wavelengths and the Q factors for all 20 measurements obtained within the 1 cm long segment. The results are shown in Fig. 5(c). For each of the 29 samples, the average resonance wavelength and the Q factor of the WGM are shown in Figs. 5(d) and 5(e). Note that instead of absolute wavelength, Fig. 5(d) shows resonance shifts, where the average resonance wavelength of the 1st sample is subtracted from those of subsequent samples.

The results in Fig. 5(d) suggest that the WGM resonance shifts continuously along the fiber spool. Based on the data, we estimate that the rate of resonance shift varies in the range of 0.10 pm/cm to 4.79 pm/cm. This resonance shift roughly corresponds to fiber diameter change at the rate of 8 pm/cm to 390 pm/cm. Experimental results in [25] show similar changes in fiber diameter. Figure 5(e) suggests that the WGM Q factor varies randomly along the spool location with no noticeable trend.

4. WGMs in aerosol-covered resonators

After clean sample characterization, we deposit various amount of welding aerosols onto these resonators, measure their transmission spectra, and extract changes in WGM characteristics. Again, the same WGM, the same fiber taper, and the same taper coupling location are used for all experiments.
We use the characterization of a specific aerosol-covered sample #10 in Fig. 5(a) to highlight several important details in our experimental procedure. First of all, as shown in Fig. 1(a), we measure the resonance wavelength and the Q factor of the clean sample WGM at 20 randomly chosen locations within the 1 cm long segment. From the measured transmission spectra, we extract 20 clean sample WGM wavelengths ($\lambda_{\text{res}}^{\text{clean}}$) and Q factors ($Q^{\text{clean}}$) and their average values, $\bar{\lambda}_{\text{res}}^{\text{clean}}$ and $\bar{Q}^{\text{clean}}$. (Our convention is that individual measurement results and their averages are distinguished by the overhead bar.) Afterwards, we deposit welding aerosols onto the clean resonator. We then perform transmission measurements at 20 random locations of the same 1 cm segment to obtain the resonance wavelength ($\lambda_{\text{res}}^{\text{aerosol}}$) and Q factor ($Q^{\text{aerosol}}$) of the aerosol-covered sample. Afterwards, we remove the 1 cm segment for SEM imaging, followed by aerosol removal. Then, we measure the WGM resonance of the remaining 1 cm control sample at 20 random locations and determine its WGM wavelength ($\lambda_{\text{res}}^{\text{control}}$) and Q factor ($Q^{\text{control}}$). If the values of $\lambda_{\text{res}}^{\text{control}}$ and $Q^{\text{control}}$ are similar to $\bar{\lambda}_{\text{res}}^{\text{clean}}$ and $\bar{Q}^{\text{clean}}$, we can conclude that any changes in the WGM of the aerosol-covered samples are due to aerosol presence only. To eliminate effects due to fiber diameter changes, we mainly consider WGM resonance shifts (i.e., $\Delta\lambda_{\text{res}}^{\text{aerosol}} = \lambda_{\text{res}}^{\text{aerosol}} - \lambda_{\text{res}}^{\text{clean}}$, or $\Delta\lambda_{\text{res}}^{\text{control}} = \lambda_{\text{res}}^{\text{control}} - \lambda_{\text{res}}^{\text{clean}}$) in our analysis.

Figure 6(a) shows two representative transmission spectra of the clean (blue line) and the aerosol-covered (red line) sample #10. A magnified view of the same transmission spectra near the desired WGM resonance is given in Fig. 6(b), with the control measurement added as the green line. (The control data are slightly shifted vertically for easier visualization.) Obviously, the WGM in the aerosol-covered sample is red-shifted in comparison to the clean sample WGM. Additionally, after aerosol cleaning, the transmission spectrum of the control sample becomes almost indistinguishable from the clean sample transmission. These observations also hold statistically. In Fig. 6(c), we show individual WGM wavelengths and...
Q factors of the clean sample (blue circles), the aerosol-covered sample (red triangles), and the control sample (green squares). The average resonance wavelengths and the Q factors for the clean sample, the aerosol-covered sample, and the control sample are

\[ \Delta \lambda_{\text{clean}} = 0 \pm 2.5 \text{ pm}, \quad Q_{\text{clean}} = (6.8 \pm 1.5) \times 10^4, \quad \Delta \lambda_{\text{aerosol}} = 29.3 \pm 3.5 \text{ pm}, \]

\[ Q_{\text{aerosol}} = (6.2 \pm 0.5) \times 10^4, \quad \Delta \lambda_{\text{control}} = -2.8 \pm 2.0 \text{ pm}, \quad Q_{\text{control}} = (6.1 \pm 0.6) \times 10^4, \]

respectively.

Clearly, the WGM resonance shift in the aerosol-covered sample is much larger than any measurement uncertainties. In comparison, the 2.8 pm resonance shift between the control and the clean sample is almost within experimental errors, and can perhaps be partially attributed to the slight difference (~1 cm) in measurement locations. In contrast to the statistically significant red-shift in resonance wavelength, the presence of welding aerosols does not seem to induce significant changes in the WGM Q factors, as can be clearly seen from Fig. 6(c).

![Image](image.png)

Fig. 7. (a) A representative SEM image of sample #10. A magnified view of the yellow square in (a) is shown in (b). (c) is the image after applying the edge-detection algorithm. (d) shows the statistical distribution of aerosols with different sizes. Aerosols with effective diameter less than 50 nm are not considered, since it is difficult to reliably identify smaller aerosols. Results obtained using three different images (#5, #6, and #9) are individually shown. The average and the standard deviation obtained using all 10 SEM images are also given as the red bars.

The SEM images of the aerosol-covered resonator surface are taken in step 4. A representative SEM image and its magnified view are shown in Figs. 7(a) and 7(b), respectively. After applying the edge-detection function in MATLAB, we can readily identify aerosol boundary in Fig. 7(c) and use it to estimate the total area of any given aerosol. Then, under the constraint that total aerosol area being the same, we can estimate the effective aerosol diameter by assuming all aerosols to be perfect spheres. For each sample, we take SEM images at 10 random locations and apply this procedure to obtain the statistical distribution of aerosol diameters. Figure 7(d) shows three representative diameter distributions, as well as the mean distribution obtained by averaging over 10 SEM images. Clearly, most aerosols are less than 550 nm in diameter, with almost 50% less than 150 nm. Additionally, we note that the above procedure likely overestimates aerosol diameter rather than underestimates it, since it sometimes counts multiple aerosols in aggregation as a single...
larger aerosol. From the SEM images, we can also define aerosol density $D_{aerosol}$ as the percentage of resonator surface covered by aerosols. For each SEM image, we calculate its $D_{aerosol}$ by dividing the total surface area occupied by the aerosols by the SEM image area. Repeating this process for all 10 SEM images, the average $D_{aerosol}$ for the sample in Fig. 6 is found to be $4.7 \pm 1.0\%$.

For each resonator sample, the above procedure gives us important parameters such as the average resonance shift $\Delta \lambda_{res}$ and the average aerosol density $D_{aerosol}$. In Fig. 8(a), we plot $\Delta \lambda_{res}$ versus $D_{aerosol}$ for all 29 samples (red triangles). Empirically, we find that the experimentally measured resonance shift satisfies the following formula:

$$\Delta \lambda_{res} = \Delta \lambda_{res}^{max} \left[1 - \exp\left(-\frac{D_{aerosol}}{D_{thresh}}\right)\right]$$

where both $\Delta \lambda_{res}^{max}$ and $D_{thresh}$ are fitting parameters, with $\Delta \lambda_{res}^{max}$ representing maximum resonance shift and $D_{thresh}$ representing the “saturation” threshold for aerosol density. In Fig. 8(a), through least square fitting (red dashed line), we find $\Delta \lambda_{res}^{max} = 75.58 \text{ pm}$ and $D_{thresh} = 5.98 \%$. In Fig. 8(a), we also include the mean and the standard deviation for the resonance shift of the clean sample (blue circles) and the control sample (green squares). Based on the results in Fig. 8(a) and Eq. (1), we estimate that as long as $D_{aerosol}$ is greater than 0.95%, the presence of nanoscale welding aerosols should lead to a measurable shift in WGM resonance wavelength of the order of 10 pm. Additionally, the resonance shift starts to saturate once $D_{aerosol}$ exceeds 5.98%.
Fig. 9. The SEM image (a.1), the WGM transmission spectra (a.2), and the WGM resonance shift versus Q factor (a.3) for the sample with \( D_{\text{aerosol}} = 15.2 \% \). In (a.2), the transmission spectra of the clean, the aerosol-covered, and the control samples are represented as the blue, the red, and the green lines. The control sample result is slightly shifted vertically. (a.3) shows the individually measured resonance shifts and Q factors of the clean (blue circles), the aerosol-covered (red triangles), and the control (green squares) sample. Similar data sets for the sample with \( D_{\text{aerosol}} = 52.3 \% \) is shown in (b). (c) tabulates the WGM properties of the two sets of samples shown in (a) and (b).

In contrast to resonance shift, the presence of welding aerosols does not seem to induce significant changes in WGM Q factors. Figure 8(b) shows the average Q factors of the clean samples (\( Q_{\text{clean}} \), blue circles), the aerosol-covered samples (\( Q_{\text{aerosol}} \), red triangles), and the control samples (\( Q_{\text{control}} \), green squares) as a function of the average aerosol density \( D_{\text{aerosol}} \).

Our experimental results suggest weak relationship between aerosol density and WGM Q factor. For alternative visualization of the same data set, Fig. 8(c) directly plots the resonance shift of the aerosol-covered sample (\( \Delta \lambda_{\text{res}} \)) versus the Q factor of the same sample (\( Q_{\text{aerosol}} \)). The result suggests WGM Q factor slowly decreases as the aerosol-induced resonance shift increases. The dependence, however, is weak and only holds statistically.

Perhaps our most surprising result is that reasonably high-Q WGM can exist in a cylindrical resonator with extremely rough surface (due to aerosol coverage). To further confirm this observation, Fig. 9 presents additional data for two samples with relatively large aerosol accumulation: sample (a) with \( D_{\text{aerosol}} = 15.2 \% \), and sample (b) with \( D_{\text{aerosol}} = 52.3 \% \). Fig. 9(a.1) show a SEM image of the aerosol-covered sample. Figure 9(a.2) provides representative transmission spectra of the clean (blue line), the aerosol-covered (red...
line), and the control sample (green line), respectively. Figure 9(a.3) gives the individual resonance shift $\Delta \lambda_{\text{res}}$ versus Q factor of the clean (blue circles), the aerosol-covered (red triangles), and the control sample (green squares). Similar data sets for sample (b) are shown in Figs. 9(b.1)-9(b.3). In Fig. 9(c), we list the average values and the standard deviations for the resonance shifts and the Q factors of the clean, the aerosol-covered, and the control samples. All these results clearly confirm our previous conclusion, namely that: 1) the WGM largely retains its characteristic transmission dip and its Q factor remains relatively high, despite the extremely rough resonator surface; 2) the presence of welding aerosols induces significant red-shift in WGM resonance wavelength. Additionally, for large aerosol density, the WGM resonance shift clearly converges towards its “saturation” value, ~76 pm.

5. Discussion

To the best of our knowledge, currently there are no theoretical models for a cylindrical resonator attached with a large number of randomly distributed NPs. Most existing studies are limited to high-Q resonators (e.g., silica microspheres) that can only support a discrete number of WGMs [7, 19, 26]. For example, it has been shown that macromolecules adsorbed on the surface of a silica microsphere can induce a small but measurable red-shift of the WGM resonance [7, 26]. Such phenomenon has been validated using first order perturbation theory [7] and is sometimes referred to as the reactive sensing principle [26]. The reactive sensing principle is only valid for small perturbations, where the resonance shift is less than the WGM linewidth [26]. For larger perturbations, mode splitting has been observed in doubly degenerate WGM resonators [2, 27]. This mode splitting has also been found numerically in a cylindrical resonator embedded with one or more small dielectric cylinders as index perturbations [28, 29]. Additionally, for silica microspheres attached with multiple gold NPs, the resonator Q factor can be significantly reduced [19].

To qualitatively understand our experimental data, especially those in Fig. 8(a), we separately consider three distinct scenarios: 1) resonators attached with only a few NPs (reactive sensing regime); 2) resonators almost fully covered with NPs (saturation regime); and 3) those with intermediate NP coverages (intermediate regime). In the reactive sensing regime, our experimental data in Fig. 8(a) suggest that the WGM resonance undergoes red-shift, and the amount of resonance shift is directly proportional to the number of welding aerosols. These observations can be explained using the analyses in [7, 30]. Specifically, if we use $\tilde{E}_{\text{WGM}}(\tilde{r})$ to denote the WGM mode excited by the fiber taper, and consider $N$ welding aerosols located at $\tilde{r}_i$, $i = 1, \cdots, N$, the aerosol-induced frequency shift ($\delta \omega$) is [7]:

$$\delta \omega / \omega = - \sum_{i=1}^{N} \alpha_i |\tilde{E}_{\text{WGM}}(\tilde{r}_i)|^2 \left( \int 2 \int \int e_{\text{res}} |\tilde{E}_{\text{WGM}}(\tilde{r})|^2 dV \right)$$

(2)

where $\alpha_i$ denotes the excessive polarizability of the $i$-th welding aerosol, and $e_{\text{res}}$ represents the permittivity of the cylindrical resonator. For most materials, $\alpha_i$ is positive. Therefore, Eq. (2) easily explains the direction of resonance shift as well as the linear relationship between $\delta \omega$ and the amount of welding aerosols on the cylinder surface.

For the saturation regime, a resonator fully covered with NPs can be analyzed using an effective index approach. Here we can treat the composite air/NP layer as a homogenous cladding, where its effective index $n_{\text{eff}}$ is essentially determined by the NP filling fraction and should be greater than 1. Therefore, compared with a clean resonator’s air cladding, the WGMs in the aerosol-covered sample penetrates deeper into the composite air/NP cladding, which again leads to the red-shift observed in our experiments. The saturation of the resonance shift means that the penetration depth of the WGM mode in the composite cladding is limited, which suggests that the value of $n_{\text{eff}}$ is less than that of silica glass. In certain
aspects, these aerosol-covered resonators are similar to the random hole fiber in [31], where the fiber cladding is a silica matrix containing a large amount of random air bubbles.

Fig. 10. (a) The formation of a circulating WGM by placing a fiber taper in close proximity to the cylindrical resonator, as suggested in [32]. (b) Optical scattering by the adsorbed NPs can effectively increase the total cavity path of the cylindrical resonator, thus producing the red shift in WGM resonance.

For the intermediate regime, we believe it would be important to account for the effects of multiple scattering induced by the NPs. A rigorous analysis, which can be carried out using the framework in [32], is beyond the scope of this paper. Intuitively, however, we can understand the effect of multiple scattering as illustrated in Fig. 10. As shown in Fig. 10(a) and discussed in [32], the WGM resonance in a clean cylindrical cavity can be understood as the interplay of two processes: the evanescent coupling between the taper mode and the WGM, and the circulation / diffraction of WGM within the cylinder. If a sufficient amount of NPs are attached to the resonator surface, the multiple scattering of light may lead to a longer propagation path, as shown in Fig. 10(b). Naturally, a longer effective cavity path should produce the red-shift observed in our experiments. This intuitive picture may also explain why the resonator Q factor does not decrease significantly. Here it is important to realize a key difference between microspheres and cylinders. Since most WGMs in non-ideal microspheres are doubly degenerate, NP-induced optical scattering tends to remove energy from the WGM and reduces its Q factor [19]. However, cylindrical resonators support an infinite number of WGMs traveling at different angles, i.e., the “spiral” modes in [3, 24]. In this case, NP-induced optical scattering can simply redistribute energy within the continuum of WGMs. The scattering induced loss can effectively be much lower, which should lead to a higher Q factor.

Comparing existing literature with our experimental results, we observe that some of the simulation results in [29] are consistent with our experimental observations. For example, the existence of the saturation regime is supported by the study of WGMs in a cylinder resonator covered with a large amount of randomly distributed small cylindrical perturbations [29]. However, other predictions in [28, 29] (e.g., mode splitting) are not observed in our measurements. The discrepancy can perhaps be attributed to the fact that existing simulations [28, 29] assume both the resonator structures and index perturbations to be cylinders that are uniform along the longitudinal direction. As a result, the full 3D complexity of the problem, such as WGM diffraction, the existence of spiral modes, and the full effects of NP scattering, are excluded from their analyses. On the other hand, some predictions in [29] may prove to be relevant. For example, due to different behaviors in particle scattering, it is possible that the presence of micron-scale welding aerosols on the resonator surface may lead to different behaviors in WGM resonance shift and Q factors. Existing impactor technology, however, is sufficient for monitoring such micron-sized aerosols.
It is worth mentioning that our discussion is by no means complete. For example, the range of validity for the three regimes considered above is unknown. In fact, an in-depth understanding of our experimental results probably requires one to consider effects of wave propagation in random media [20], where many interesting phenomena such as coherent back scattering [21] may play important roles. Such analyses, however, are beyond the scope of this paper and will be considered in future publications.

Finally, we discuss two practical issues involved in cylindrical resonator based aerosol sensing. First, for applications that require real-time \textit{in situ} monitoring, we must make sure that the coupling components (e.g., the fiber tapers) remain free of aerosol attachment. In practice, it should be feasible to address this challenge through careful sensor packaging and design, where we ensure that coupling components are not exposed to aerosol flow. Second, the extremely high sensitivity of the WGM sensor can sometimes pose problems. For example, in our experiments, it takes approximately less than 1 minute (or less than 1 welding rod) to reach the saturation regime. (The distance between the welding arc and the resonator is approximately 50 cm.) For systems intended for long-term monitoring, we may need to significantly reduce the in-take of nanoscale aerosols to ensure proper function. Alternatively, we may use such sensors as an early warning system that can indicate, in real time, a dangerously high level of nanoscale aerosols. For such applications, the cylindrical resonator enjoys enormous advantages due to its low cost, ease of fabrication, small form factor, and wide commercial availability.

6. Conclusion

In this paper, we demonstrate the feasibility of using a standard silica fiber as a cylindrical WGM resonator to detect the presence of nanoscale aerosols generated by electric arc welding. We find that the accumulation of welding aerosols on the resonator produces a measurable red-shift in the WGM resonance wavelength. Furthermore, we establish an empirical relationship that quantitatively links the degree of resonance shift with the amount of aerosols deposition. Surprisingly, we find that the WGM Q factor does not decrease significantly, even for resonators with large aerosol accumulations and extremely rough surface profiles. We tentatively present three scenarios that may explain our experimental results under different regimes, and suggest that one may need to consider multiple scattering by randomly distributed aerosols in order to fully explain the experimental results.

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