

Spatial refractive index sensor using whispering gallery modes in an optically trapped microsphere

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The authors propose the use of an optically trapped, dye doped polystyrene microsphere for spatial probing of the refractive index at any position in a fluid. Using the dye embedded in the microsphere as an internal broadband excitation source the authors eliminated the need for a tunable excitation source. They measured the full width at half maximum and frequency spacing of the transverse electric and transverse magnetic resonances as a function of the refractive index of the immersion fluid. The authors achieved a sensitivity of 5×10^{-4} in refractive index, even when the exact size of the microsphere was not known. © 2007 American Institute of Physics. [DOI: 10.1063/1.2722695]

Microspheres can act as high Q resonators in the optical regime^{1,2} and can store electromagnetic waves by repeated total internal reflection. Constructive interference results in an enhanced internal field which is called a whispering gallery mode (WGM). Because of the evanescent interaction between the WGM and the surrounding medium the WGM's resonance frequency is sensitive to changes in the refractive index of the sphere's surroundings. This sensitivity makes microspheres a useful tool in sensory applications. Recently Hanumegowda *et al.* demonstrated the use of microspheres in refractometric sensing.³ Moreover Vollmer *et al.*⁴ and White *et al.*⁵ demonstrated protein detection using microspheres. In all these studies³⁻⁵ narrowband light from a tunable diode laser was evanescently coupled to the microsphere via an optical fiber. Spatial mapping of the refractive index was not possible because the microsphere could not be freely moved through the sample.

In this letter we demonstrate the use of a dye doped polystyrene microsphere as a refractometric sensor. The polystyrene microsphere was trapped with optical tweezers, by which it could be positioned anywhere within the fluid. It can therefore be used to sense the refractive index in spatially inhomogeneous media, for example, near interfaces or in mixing flows. In this system we did not need a tunable laser to excite WGMs, instead we used the dye embedded in the microsphere itself as a broadband excitation source. The microspheres used in this study were synthesized in bulk and consequently showed a considerable size polydispersity. The absolute frequency of the WGM, which was used to probe the refractive index in previous studies,³⁻⁵ is very sensitive to the size of the sphere. Measurements of the absolute resonance frequency of a particle that has not been individually calibrated does not provide information about the refractive index of the medium. In contrast, we show that both the full width at half maximum (FWHM) of the emission peaks and the frequency spacing between adjacent transverse electric (TE) and transverse magnetic (TM) modes are sensitive and robust probes of the refractive index of the sphere's surroundings. We show that our method works over the whole

range of the microsphere size distribution without the need for individual calibration.

The FWHM of a WGM is determined by the loss upon total internal reflection at the curved surface of the microsphere. These losses depend on the mode number l and the refractive index contrast $m = m_0/m_l$, where m_0 and m_l are the refractive indices of the immersion medium and the microsphere, respectively. The FWHM of a WGM can be expressed as⁶

$$\frac{\Gamma}{2} = [Nx^2 n_l(x)^2]^{-1}, \quad (1)$$

with n_l the spherical Neumann function, $x = m_0ka$ the size parameter at which the WGM occurs, where k is the wave vector in vacuum and a the radius of the microsphere, and

$$N = \begin{cases} m^2 - 1 & \text{for TE modes} \\ (m^2 - 1)[\mu^2 + (\mu^2/m^2 - 1)] & \text{for TM modes,} \end{cases} \quad (2)$$

where $\mu = v/x$, with $v = l + 1/2$.

The frequency spacing between a TE and TM mode of the same mode number is due to a difference in phase shift of s and p polarized light upon reflection at the sphere surface. The absolute frequency at which a WGM occurs is defined as the real part of the pole of the scattering coefficient,⁷ and we calculated the frequency spacing by numerically calculating the poles for TE and TM modes.

In our experiments we used dye doped polystyrene microspheres, suspended in water (G1000, Duke Scientific Corporation). We measured the size distribution of the spheres by determining the absolute resonance frequencies of 24 microspheres in a reference fluid (water) and found a mean radius of $5.30 \mu\text{m}$ with a maximum deviation of only 0.5%. Taking into account a conservative error margin, we assumed that the size of any sphere we trap was $5.30 \pm 0.05 \mu\text{m}$. This size distribution is much narrower than the specifications of the manufacturer ($5.0 \pm 0.3 \mu\text{m}$). Although with our method it is not necessary to know the exact size of any individual sphere, the size distribution must be measured in advance. The microspheres were doped with 2% dye which emits in the green part of the spectrum ($\lambda = 480 - 540 \text{ nm}$). The sample holder was borosilicate glass capillary with inner dimensions $0.1 \times 2 \times 40 \text{ mm}^3$.

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With optical tweezers operating at 1064 nm we held a single microsphere far away from any surface. The lateral trap stiffness (measured using a quadrant photo diode⁸) was 190 ± 40 pN/ μm using 1 W of laser power on the sample. The excitation light source was a continuous wave argon laser (Spectra Physics Satellite 2016) emitting at 488 nm with a maximum output power of 30 mW, attenuated to 500 μW on the sample. We expect that similar results could be obtained with thermal illumination. The light of the excitation source was slightly defocused on the sphere, so that the whole sphere was illuminated. The emission of the dye was collected through a water immersed objective (numerical aperture of 1.2) and detected by an intensified charge-coupled device camera (Princeton Instruments ICCD 576), connected to a spectrograph (Oriel Instruments, MS257) which has a Gaussian response function with a width of 4.5 cm^{-1} .

Tuning of the refractive index of the immersion fluid m_0 was achieved by mixing water with ethylene glycol ($m_0=1.4317$ at 293 K and 589 nm) in various fractions. Ethylene glycol does not chemically affect polystyrene and is miscible with water in all volume fractions. Refractive indices of the mixtures were measured with an Abbe refractometer type 1 T which is temperature stabilized and has an accuracy of 2×10^{-4} in refractive index.

We compared our experimental emission spectra to an elaboration of Mie theory (EMT) developed by Chew^{9,10} describing the emission of dipoles inside a spherical cavity. The only parameters were the refractive index of the sphere m_1 , the refractive index of the immersion medium m_0 , and the sphere radius a . The refractive index of the sphere was measured¹¹ to be $m_1=1.586-1.5 \times 10^{-5}i$. The refractive index of the immersion fluid m_0 was measured with the refractometer. The only free parameter in our system was the radius of the sphere a . The theoretical emission spectra were corrected for the response function of the spectrograph.

In Fig. 1 we show the emission spectra obtained from a single dye doped polystyrene microsphere immersed in (a) water ($m_0=1.3325$) and (b) a mixture of water and ethylene glycol ($m_0=1.3978$). Sharp peaks were observed in the spectrum due to WGMs. The dashed lines show the radiation rates (normalized to the rate in bulk polystyrene) as predicted by EMT for (a) $a=5.33$ μm and (b) $a=5.319$ μm . Comparing the spectra in (a) and (b) we observe that the decrease in refractive index contrast induced an increase in FWHM of the WGMs and a decrease in the spacing between the adjacent TE and TM modes.

As we have shown in Fig. 1 it is possible to obtain m_0 and a by fitting the experimental spectra with a theoretical emission spectrum. However, the initial values for the parameters must be chosen rather carefully. The same parameters can be obtained in a more straightforward way by comparing only the FWHM and the TE_{96} - TM_{96} mode spacing to predictions from EMT. Both the FWHM and the mode spacing were determined by fitting a Voigt profile¹² to an individual WGM in the measured emission spectrum. The Voigt profile models the convolution of the Lorentzian width of the resonance and the Gaussian response function of the spectrograph. To accurately fit the resonance shape, we subtracted a quadratic fit of the background intensity.

In Fig. 2 we show the Lorentzian FWHM as a function of m_0 for the TE_{96} mode. The size of the error bars was taken from the error in the Lorentzian width of the fits. The large

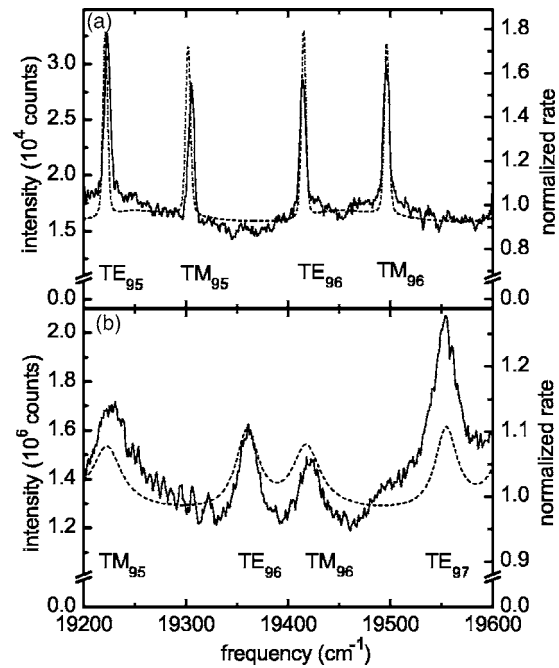


FIG. 1. Measured emission intensity (solid lines) of single dye doped polystyrene microspheres immersed in (a) water ($m_0=1.3325$) and (b) a mixture of water and ethylene glycol ($m_0=1.3978$). The dashed lines show the normalized radiation rates as predicted by EMT for (a) $a=5.33$ μm and (b) $a=5.319$ μm .

error at $m_0=1.41$ was due to a low signal to noise ratio resulting from a low refractive index contrast. The theoretical FWHMs (solid lines) were calculated for the biggest and smallest spheres in the measured size distribution and were calculated from Eq. (1). It can be seen that the FWHM is very insensitive to the sphere size as the calculated curves are almost identical.

In Fig. 3 we show the experimental TE_{96} - TM_{96} mode spacing, obtained from the central frequency of the fitted Voigt line shapes. The error bars are the uncertainty in the central frequency of the Lorentzian fits. The theoretical mode spacing is again calculated for the biggest and smallest spheres in the measured size distribution, and the result is shown as a gray area. It can be seen that the TE_{96} - TM_{96} mode spacing is slightly more sensitive to sphere size than the FWHM (Fig. 2).

Figures 2 and 3 show that our measurements correspond very well to the theoretical model, even though the size of the trapped microsphere varies between measurements due to

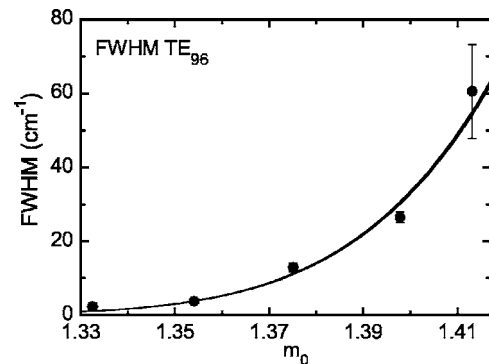


FIG. 2. Measured Lorentzian FWHM vs m_0 for the TE_{96} mode. The solid lines show the theoretical FWHM [Eq. (1)] for the biggest and the smallest spheres in the measured size distribution.

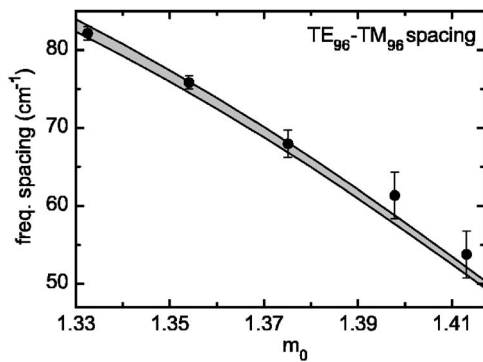


FIG. 3. Measured TE_{96} - TM_{96} mode spacing vs m_0 . The gray area shows the theoretical mode spacing calculated for the biggest and smallest spheres in the measured size distribution.

size polydispersity. The polydispersity in the sphere size introduced an inaccuracy in the theoretical values for the FWHM and TE-TM mode spacing of $<1\%$.

One can use either the FWHM or the mode spacing to measure the refractive index of the medium. However, the sensitivity and robustness of these methods is not the same. The TE_{96} - TM_{96} mode spacing is nearly linear in our refractive index range with a slope of 385 cm^{-1} per refractive index unit. An accuracy in width of 2 cm^{-1} was easily achieved for refractive indices below 1.39, resulting in a sensitivity of nearly 5×10^{-3} in refractive index. When measuring the FWHM of a WGM the sensitivity of the sensor is higher and varies throughout our refractive index domain. From the slope of the curve in Fig. 2 we obtained a sensitivity of 5×10^{-4} in refractive index around $m_0=1.41$.

The FWHM method leads to the most precise measurements in the ideal case. However, this method is very sensitive to line broadening due to contamination at the surface of the sphere. When contamination is suspected, both the TE-TM mode frequency spacing and the FWHM should be measured. If both methods lead to the same value of the refractive index, contamination can be excluded.

In conclusion we demonstrated a spatial probe for the refractive index comprising an optically trapped dye doped microsphere. The use of optical tweezers gives us the freedom to measure the refractive index anywhere in the fluid.

This freedom allows for spatial mapping of refractive index gradients or inhomogeneities caused by, for example, flow in microfluidic devices or chemical processes. The emission of an embedded dye acted as the internal excitation source. We measured the FWHM and the frequency spacing between TE and TM modes as a function of refractive index of the immersion fluid. The measured values showed excellent agreement with theoretical predictions, confirming the suitability of the system for spatial probing of the refractive index. Individual calibration of the sphere size was not necessary because the measured parameters are practically insensitive to sphere size within the size distribution found in commercially available microspheres. Very recently, Knöner *et al.*⁸ have demonstrated a method to determine the refractive index of a trapped microsphere by measuring the trapping force. Their method and ours are complimentary and could easily be combined, providing a versatile system for sensory applications.

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