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Whispering-Gallery Mode lasers for biosensing: a rationale for reducing the lasing threshold

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ABSTRACT

Whispering-gallery modes have been studied extensively for biosensing applications. Whilst the vast majority of work undertaken has focused on high Q factor resonators, with the main improvement being a reduction of the resonator size to improve sensitivity, we have chosen a different pathway by starting with resonators that exhibit extremely high refractive index sensitivity but low Q factor. A way forward to overcome this limitation is to introduce a gain medium and operate the resonator above its lasing threshold. This has been shown to result on average in a 5 fold increase in the Q factor. With the lasing threshold itself being dependent on the Q factor, amongst other parameters, the Q factor enhancement can be exploited to either reduce the lasing threshold or alternatively enable smaller resonators to be operated above their lasing threshold. As a demonstration we present a 10 μm diameter polystyrene microsphere lasing in aqueous solution for refractive index sensing applications, which to the best of our knowledge is the smallest polystyrene microsphere laser ever demonstrated in these conditions.

Keywords: Whispering-gallery modes, microresonators, lasing.

1. INTRODUCTION

Since the discovery of Whispering-Gallery Modes (WGM) by Lord Rayleigh in the late 19th century with the observation of sound wave propagation inside the whispering-gallery of St Paul's Cathedral in London, a wealth of work has been pursued to unlock the potential of WGMs at optical frequencies for a large range of applications. Whispering-Gallery Modes occur when light is trapped inside a resonator by total internal reflection, circulating along the inner surface and returning in phase after single or multiple round trips to satisfy the resonance conditions [1]. WGMs are especially suited to refractive index sensing, since the spectral position of the resonances is dictated not only by the resonator geometry (e.g. diameter, sphericity) and optical properties, but also by the surrounding refractive index. Two distinct approaches have been developed to exploit WGMs, in a wide range of resonator geometries ranging from rings/toroids [2] and spheres [3] to cylinders and capillaries [4]. The first approach is based on using evanescent field coupling between a phase-matched tapered optical fiber (or prism) and the resonator, and scanning across a narrow wavelength range with a distributed feedback laser to identify the resonance positions and linewidths [5]. While this approach has allowed for unprecedented sensing performance and Q factors [5], it is limited in practice because any variation of the distance between the tapered fiber and resonator results not only in changes in the coupling efficiency but also in fluctuation of the resonance positions [6, 7]. Furthermore, using small optical resonators, which would enable improved performance, since the refractive index sensitivity is inversely proportional to the resonator diameter [8], is challenging from a practical point of view using this approach. The alternative approach involves using resonators that contain a gain medium. Upon illumination of these active resonators by a remote light source, light is emitted by the gain medium. At the resonance wavelengths of the microcavity, the Purcell effect increases the emission rate of the gain medium [9]. As a consequence, the resulting WGM spectrum observable remotely in the far field shows up as a fluorescent signal modulated by sharp peaks corresponding to the microcavity resonances [10]. While this approach offers advantages in terms of practicality such as robustness of the coupling scheme, easy excitation and collection of the WGM modulated fluorescence, the Q factor observed for the radiation modes are often 3 or 4 orders of magnitude lower than those observed using the evanescent field coupling approach, eventually limiting the use of such a WGM excitation strategy in terms of resolution for sensing applications [11]. A way forward to address this issue is to operate the microresonator beyond its fluorescence

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regime, inducing stimulated emission, resulting in a higher Q factor [10]. Examples of such lasing WGMs have been published by Kuwata, Gonokami and Takeda, who used dye doped polymer microspheres [12]. To the best of our knowledge, the smallest lasing polymer microspheres used in aqueous solution are 15 μm in diameter and no work has been reported on investigating the size limitations of polymer microspheres for inducing lasing in aqueous media where all refractive index sensing applications and especially biological sensing are performed.

Here, we present a strategy to produce WGM lasers consisting of 10 μm diameter polystyrene microspheres, which to the best of our knowledge are the smallest polystyrene microspheres ever reported to lase in water for refractive index sensing applications.

2. MATERIALS AND METHODS

Polystyrene microspheres with a nominal diameter (Ø) of 10.52 μm with a standard deviation (ΔØ) of 0.25 μm and a refractive index of 1.591 (Polysciences Inc., USA) were doped with a fluorescent laser dye (Nile Red, λ_{abs} ~ 532 nm, λ_{em} ~ 590 nm, Sigma Aldrich) using a liquid two phase system [10]. The fluorescent dye was first dissolved into xylene until the solubility limit was reached. The resulting solution was poured on top of an aqueous solution of diluted microspheres (5 mL H_2O + 100 μL microsphere solution 2.5% solid) and left on a magnetic stirrer plate until the xylene had completely evaporated. As xylene and water are immiscible and the fluorescent laser dye used hydrophobic, when the xylene evaporates, the fluorescent dye is transferred into the microspheres that come into contact with the dye solution. This method, compared with other approaches previously described consisting of coating the microresonator surface with either quantum dots or organic dye molecules [13, 14], enables a variation of the dye content to be loaded within the polymer sphere by simply changing the volume of the liquid phase containing the dye. After the doping procedure, the microsphere solution was heated at 95 °C for 1 hour to facilitate the removal of the solvent from the microspheres. The microspheres were then washed by centrifugation, the supernatant removed and the lost volume replaced by Millipore water. Several microsphere samples were prepared following the same procedure but with increasing volume of dye saturated xylene solution, increasing the amount of dye diffusing into the polystyrene microspheres.

A frequency doubled YAG laser (λ = 532 nm, ~ 800 ps pulse duration, 10 kHz repetition rate, Alphalas GmbH, Germany) was used for the excitation of the active microspheres. The beam emerging from the laser was first spatially filtered using a single mode fiber (SMF28, $Ø_{core}$ = 8 μm) before being coupled into the back port of an inverted microscope (IX 71, Olympus, Japan) equipped with a 532 nm dichroic filter, effectively using the microscope as a confocal setup. The WGM modulated fluorescence spectra from isolated microspheres deposited onto a microscope glass cover slip and observed through the microscope were spectrally resolved using a monochromator (iHR550, Horiba, Japan) equipped with three different gratings; 600, 1200 & 2400 mm^{-1} and a cooled CCD (Synapse 2048 pixels, Horiba, Japan).

3. RESULTS AND DISCUSSION

3.1 Theoretical modelling

The lasing threshold is a function of the gain dictated by the dye concentration, its quantum efficiency, and the Purcell enhancement effect that occurs at the resonance wavelengths, and also the optical losses of the microsphere, which are described by the Q factor. Only a limited number of publications have ever investigated the relationship between lasing threshold and the parameters listed above, and only Spillane et al. [15], provides an analytical expression for the lasing threshold as function of the Q factor, mode volume (V) and gain coefficient (specifically for a WGM Raman laser). One of the conclusions that can be drawn from that paper is that the lasing threshold should depend on V/Q^2. In earlier work by Sandoghdar et al. [16] the lasing threshold of neodymium doped silica microspheres was found to have a linear dependency of Q^{-1}. The Q factor can be directly inferred from spectral measurements of the WGM following the standard definition of $Q=\Delta\lambda/\lambda$. In order to assess the prospects of lasing in smaller microspheres in liquid, we calculated the variation of the mode volume of the WGMs for both a resonator in aqueous solution where sensing applications are performed, and when the resonator is in a dry environment using the standard definition [17].

$$V = \frac{\int \varepsilon(r)|\psi(r,\theta,\phi)|^2 r^2 \sin\theta dr d\theta d\phi}{max(\varepsilon(r)|\psi(r,\theta,\phi)|^2)} \qquad (1)$$

where $\varepsilon(r)$ is the radial dependence of the relative permittivity and $\psi(r, \theta, \phi)$ is the electric field of a given whispering-gallery mode in spherical coordinates. The mode fields were calculated using the tried and tested finite element method (FEM). From this calculation, it was found that the surrounding environment has only a limited impact on the mode volume which varies for the TE mode located around 590 nm from 10.1 μm^3 in air to 10.4 μm^3 in water. The proportion of energy outside the cavity increases from < ½ % to around 6%. The polar mode number l closest to 590 nm was chosen (l = 77, 78 for air and water clad spheres, respectively) for the simulations as determined by [18, 19]. Similarly, the TM mode within the same spectral region increases from 10.7 μm^3 in air to 11.9 μm^3 in water, and the increase in energy outside the cavity is again approximately tenfold. The relatively low dependency of mode volume on the specific surrounding index (provided the index contrast remains significant) can also be inferred from Figure 1, in which changing the surrounding medium from air to water only marginally lowers the peak intensity as the mode fields shift outwards slightly.

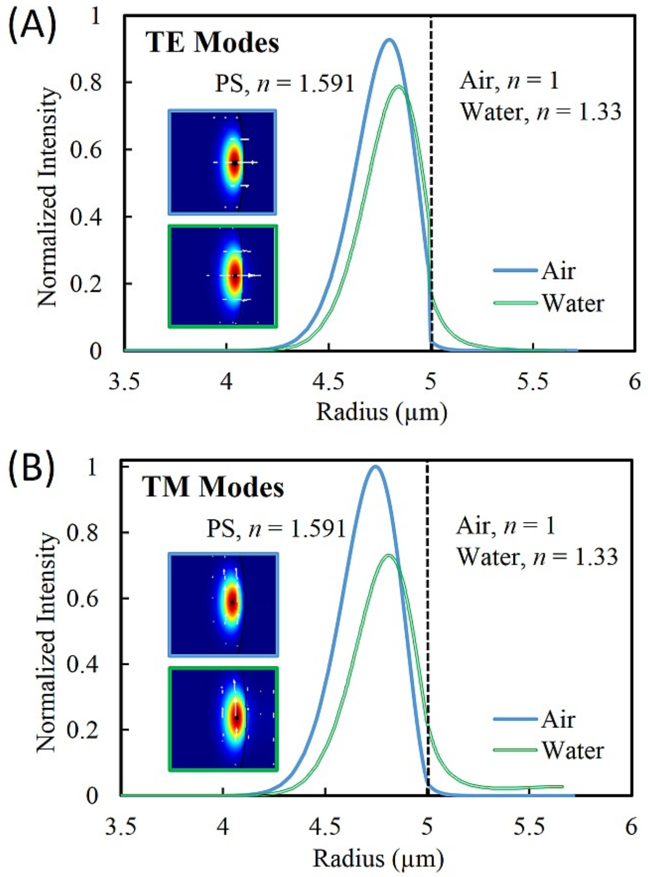

Figure 1. The radial dependence of the (A) TE and (B) TM mode intensities, of the 10 μm diameter polystyrene microsphere around 590 nm. Both air and water clad microspheres are simulated. Intensity distributions in the equatorial plane are shown in the insets with arrows showing the magnetic field directions.

Typical WGM spectra measured from a 10 μm diameter polystyrene microsphere in air and immersed in water are shown in Figure 2 (A) and (B). The immersion of the microsphere in water has a significant impact on its Q factor – it is reduced by a factor two from ~4×10^3 in air to 2×10^3 in water. This suggests that the lasing threshold of a 10 μm diameter polystyrene microsphere should only increase by a factor 10 once it is immersed in water assuming that the gain factor and especially the dye concentration and quantum efficiency remain the same, despite the modification of the local environment surrounding the sphere. These assumptions are justified by the fact that the dye is embedded into the polymer matrix constituting the microsphere and is therefore not affected by the change of solvent. Furthermore, as the organic dye used in this experiment is not soluble in water there is no reason for the concentration to change due to diffusion out of the polymer matrix.

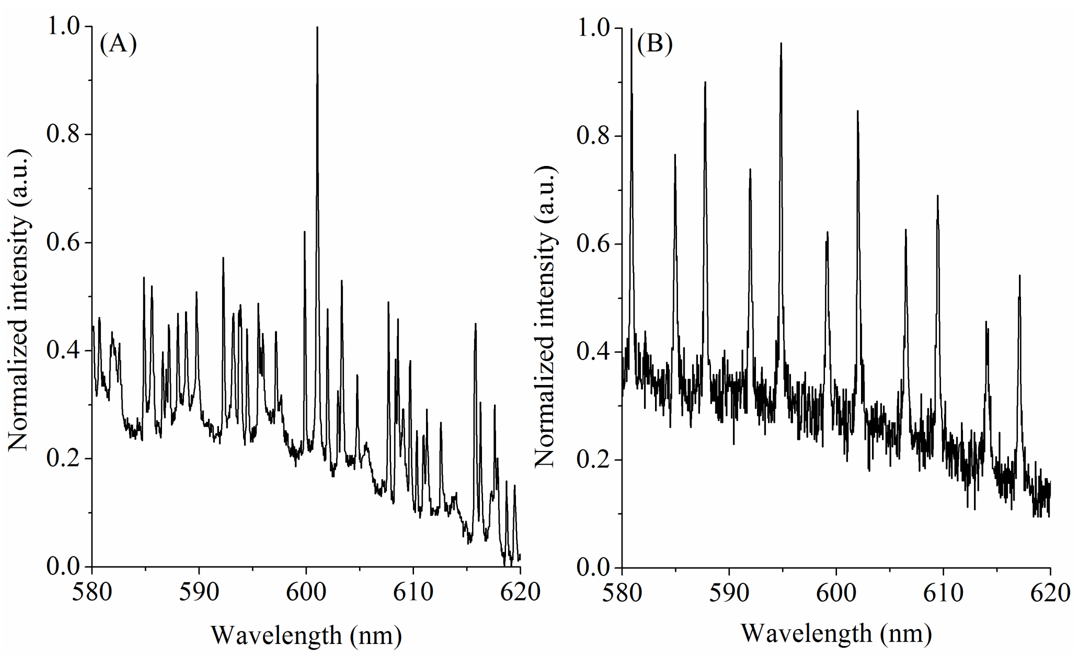

Figure 2. WGM spectra of a 10 μm diameter polystyrene microsphere (A) in air and (B) immersed in water.

3.2 Lasing Whispering-Gallery Mode microsphere in air

A typical series of WGM spectra measured from a 10 μm diameter polystyrene microsphere in air with increasing pump power are shown in Figure 3 (A).

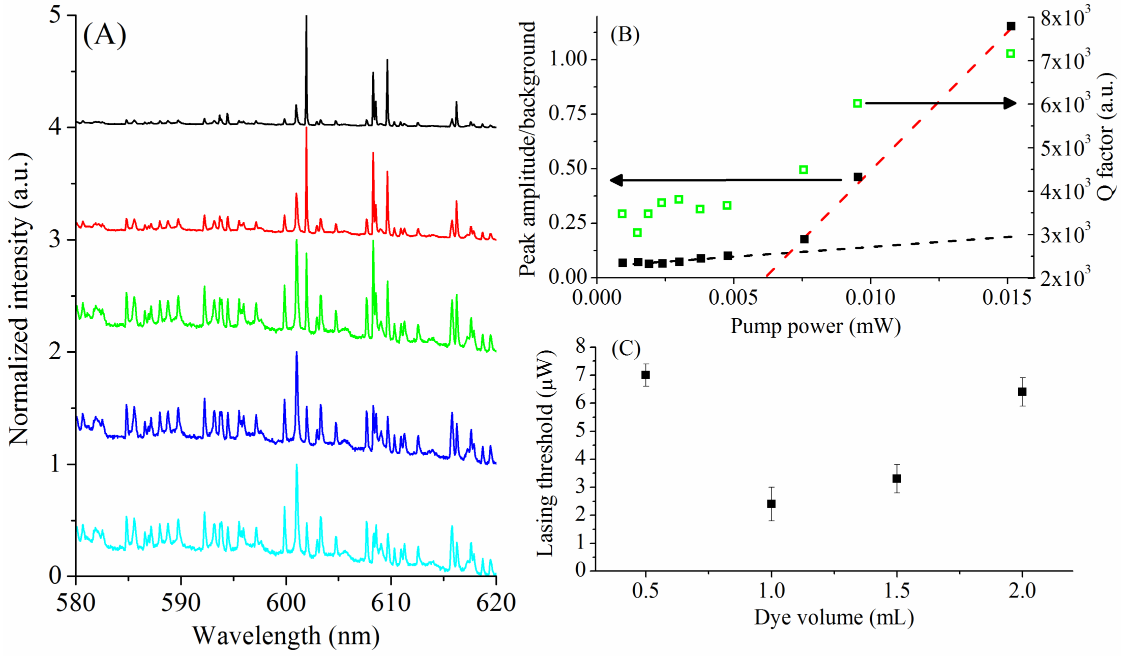

Figure 3. (A) Normalized WGM spectra of a 10 μm diameter dye doped polystyrene microsphere in air with increasing pump power (1 μW to 15 μW). (B) Ratio between the highest intensity WGM resonance and fluorescence background, and measured Q factor as function of the pump power for the same polystyrene microsphere in air. (C) Measured lasing threshold of the WGM in air as a function of the volume of saturated dye solution used in the doping process.

As the pump power increases, some modes become more intense, especially around the 600 nm region of the spectra. Figure 3 (B) shows the ratio between the most intense WGM resonance and the background level as a function of the pump power. The transition between the fluorescence and the stimulated emission regimes can be clearly seen around 7 μW. Similarly the measured Q factor, defined as λ/Δλ also increases beyond the lasing threshold, from 4×10^3 to almost 8×10^3. As the Q factor describes the stored energy in the resonator, a higher gain in the resonator, especially upon lasing will increase the stored energy and therefore the Q factor. This increase in Q factor is highly beneficial for sensing purposes as it increases the resolution of the sensor, enabling the detection of smaller changes in the resonance wavelength positions [11] and an improved detection limit [20]. Another parameter which could influence the lasing threshold is the gain medium concentration. To determine the optimum doping conditions, variations of the doping procedure described previously were followed which essentially increased the volume of dye solution. Similar measurements were performed for each microsphere sample, and the lasing threshold in air as a function of the volume of dye solution used was extracted, as is shown in Figure 3 (C). This figure illustrates clearly that upon reaching the optimum dye concentration within the microsphere, the lasing threshold can be significantly reduced, in this case down to 2.4 μW. However, once the optimum dye concentration is reached, any subsequent increase in the dye content within the microsphere results in self-quenching of the organic dye [21], and hence a higher lasing threshold.

3.3 Lasing Whispering-Gallery Mode microsphere in water

Similar measurements were performed with the same microsphere samples in aqueous solution, drastically decreasing the refractive index contrast between the microresonator and its surrounding environment, resulting in higher confinement losses of the propagating modes as the reflectivity at the interface is reduced and a higher portion of the evanescent field leaks out of the resonator. As a consequence, all the higher order modes are quenched and only the first order TE and TM modes can be seen in Figure 4 (A), which shows a typical WGM spectrum of a 10 μm dye doped polystyrene microsphere in water [22, 23].

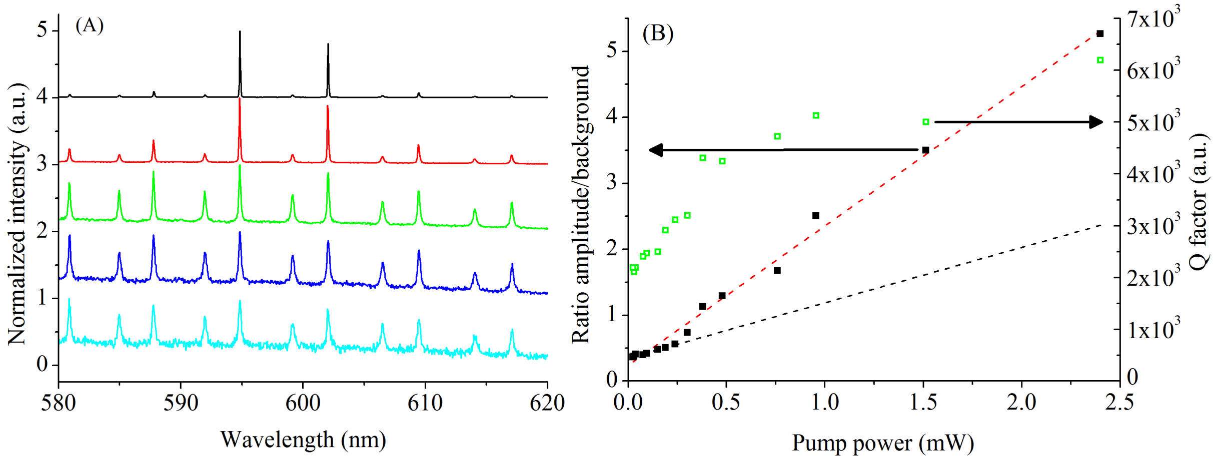

Figure 4. (A) Normalized WGM spectra of a 10 μm diameter dye doped polystyrene microsphere in *water* with increasing pump power (20 μW to 2.5 mW). (B) Ratio between the highest intensity WGM resonance and fluorescence background, and measured Q factor as function of the pump power for the polystyrene microspheres in water.

The Q factor of the first order mode is also strongly affected, dropping from 4×10^3 to 2×10^3. This decrease of Q factor, which defines the stored energy within the resonator, as function of the surrounding refractive index is consistent with the results reported for microspheres below 10 μm in diameter as function of surrounding refractive index [24]. For all the different batches of the microspheres prepared with different dye concentration, lasing was observed repeatedly in water for only one sample, shown in Figure 4 (A) and (B), which initially exhibited the lowest lasing threshold in air as shown in Figure 3 (C) for 1 mL of saturated dye solution. For all the other samples, lasing was not achievable before damaging the microsphere with the pump source. The lasing threshold of the 10 μm dye doped polystyrene microsphere measured in water (245 ± 2 μW) is shown in Figure 4 (B). For the entire population of 10 μm polystyrene microspheres measured, the lasing threshold in water is about 2 orders of magnitude higher than the lasing threshold observed for the same sample in air. However neither the V/Q^2 nor the Q^{-1} reported dependencies mentioned earlier for the lasing threshold explain why the lasing threshold is increased by two orders of magnitude when the measurements are performed in water. Our results therefore suggest that the Q factor has a much stronger influence on the lasing threshold than initially anticipated although further investigations could be pursued beyond the work reported here to understand fully the dependency of the lasing threshold on the Q factor. Nevertheless, as measurement of the Q factor has been used in previous work on WGMs to characterize changes of surrounding refractive index and biomolecular binding onto high Q factor resonator [25], one might appreciate that measuring the lasing threshold might be an interesting alternative to the standard resonance wavelength characterization, especially if the dependency of the lasing threshold varies as a power function of the Q factor.

CONCLUSION

We have shown that a 10 μm diameter dye doped polystyrene microsphere can support WGM lasing when operated in water. This is the smallest polystyrene microsphere ever demonstrated to lase in an aqueous environment. This was made possible by investigating the lasing threshold first in air as a function of the dye concentration within the microsphere and then selecting the optimum dye concentration resulting in the lowest possible lasing threshold. With regards to the application of refractive index sensing, a reported refractive index sensitivity of 50 nm/RIU [22] and a Q factor of 7×10^3 upon lasing (FWHM ~ 80 pm at 600 nm), means that the resolution is now only limited by the monochromator used for the characterisation of the WGM signal (resolution of 4 pm), which enables a detection limited 8×10^{-5} RIU. This experiment has also shed light on the dependence of the lasing threshold on Q factor, which is likely to have a far stronger influence than the quadratic dependency reported in the literature. While this would require further investigation, this paper has provided insight into the possibility of inducing lasing in even smaller microresonators. Since the lasing threshold reported here for 10 μm diameter polystyrene microspheres is already very close to the damage threshold at 532 nm, inducing lasing in even smaller polystyrene microspheres would require relying on alternative excitation schemes, using either microstructured optical fiber [10], plasmonic effects or a combination of both.

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