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Flexible microresonators: lasing and sensing

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ABSTRACT

Microresonators have drawn a great deal of interest for their importance in both practical applications and fundamental physics in light-matter interaction. The optical confinement provided by a microresonator greatly enhances the interaction between optical spatial mode and the light emitting materials. Conventional fabrication of microresonators adopting semiconductor processing technology (no matter top-down or bottom-up approach) still faces some challenges. Here we report the feasibility of constructing solid state microresonators with various configurations including spheres, hemispheres and fibres from organic polymer in a flexible way. We realize optically pumped lasing from these structures after incorporating organic dye materials and/or colloidal quantum dots into the resonators. The lasing characteristics have been systematically examined in terms of size dependence and polarization. The longitudinal optical modes are well defined by whispering gallery modes. We are also able to tune the resonance modes by deforming the shape of micro-spheres, representing the facile manipulation of light-matter interaction. Finally, refractive index sensing with high sensitivity can be readily realized from these structures enabled by the existence of evanescent waves and improved by Vernier effect in coupled resonators.

Keywords: Microresonators, whispering gallery mode, lasers, droplets, hemispheres, microfibers, polymers, sensors, Vernier effect, coupled resonators,

1. INTRODUCTION

Microresonators or microcavities offer optical confinement that increases the light-matter interaction, and therefore, are key building blocks in the investigation of quantum electrodynamics, information transmission/processing and novel light sources [1]. Generally, microcavity fabrication relies on semiconductor processing with either top-down or bottom-up technique [2, 3]. For the top-down approach, cavities can be obtained by etching a planar cavity [2] while for the bottom-up approach, microstructures are self-assembly by crystallization [3]. These two methods, however, face some challenges that are usually required costly equipments and series of complicated fabrication steps. Recently, cavities formed by surface tension such as, microspheres, microtoroids have attracted great attention due to their simple fabrication [4, 5]. These resonators confine light by multiple total internal reflections at the cavity-its surrounding interface known as whispering gallery modes (WGM), which provide ultrahigh quality (Q) factor and small mode volume. They are beneficial for studying nonlinear optics, low threshold lasers, and highly sensitive label-free biosensors. However, the surface tension based technique has applied mostly to inorganic (silica, glass) materials.

Compared with traditional semiconductor and inorganic materials, organic polymers are better in mechanical flexibility and simpler process. Here, we demonstrate microresonators fabricated from surface tension induced polymer droplets, hemispheres and fibers [6-11]. By incorporating dye molecules into these structures, microlasers with excellent performances such as, narrow spectral linewidth, clear mode spacing and sensitive refractive index sensors have been successfully realized.

2. FABRICATION

2.1 Microspheres

Convenient structure for realization of whispering gallery modes is a dielectric sphere or droplet. High Q factor droplets based on liquid crystals [12] and organic solvents [13] have been reported. Herein, we introduce surface tension induced polymer droplets in a PDMS carrier [6]. The droplet solution was made by subsequently dissolving polystyrene (PS), epoxy resin (Araldite 506) [7] and Rhodamine B (RhB) in dichloromethane (DCM). Concentration of PS in DCM is

about 14 wt% and weight ratio between epoxy and PS is around 15:1. To create droplets, we used a metal tip to draw a line of the dye doped solution into PDMS as shown Figure 1(a). The solution has strong surface tension and is immiscible in PDMS. Thanks to those factors, it breaks into number of pieces and from that droplets are self-assembly created. Interestingly, the droplets tend to have decreasing sizes along the drawing direction as depicted in Figure 1(c). Droplets with diameters ranging from 100 to 10 μm can be readily obtained, convenient for investigation of size dependent characteristics. Figures (d)-(i) show a real self-assembled process from breaking pieces to spherical droplets. In addition, the droplets are well-aligned following the drawing line as shown in Figure 1(i).

Figure 1. (a)-(c) Fabrication technique and scheme of self-assembled polymer droplets [6]. (d)-(h) Top-down optical image of a self-assembled process that induces droplets. This process takes only about a minute. (i) Image of a well-aligned and uniform droplets.

2.2 Hemispheres

Hemispherical cavities on top of a distributed Bragg reflector (DBR) can be obtained from a fiber thanks to the flexibility of organic materials. The fiber is fabricated by directly drawn from a solution as shown in Figure 2(a) [10].

Figure 2. (a) Direct drawing technique for fiber fabrication. The hydrophobic layer reduces adhesion of the DBR surface. (b) Hemispherical cavities are self-assembly induced from a fiber by surface tension. This process takes about tens of minutes.

We dissolved poly(methyl methacrylate) (PMMA) in DCM and then added the epoxy and RhB. Weight ratio between PMMA and epoxy is about 6:1 and RhB concentration is around 0.2% [8]. The DBR surface is coated with a layer of 1H, 1H, 2H, 2H-perfluorooctyltriethoxysilane [14], the so called hydrophobic layer because it increases hydrophobic effect of the DBR surface. Similarly to the droplets, as shown in the Figure 2(b) the fiber that were placed on top the DBR after fabrication broke into many pieces and from that hemispheres were self-assembly formed. These cavities have a circular shape and smooth outer surface, which guarantee for high Q factor.

2.3 Microfibers

Interestingly, the above fiber will not breakdown but maintain its shape after drawing if we decrease the amount of the epoxy added. Generally, we use a solution in which weight ratio between PMMA and epoxy is around 1: (1.5–2) to make solid fibers [10]. After fabrication, we hang the fibers on a special design substrate that has a gap as depicted in Figure 3(a). Part of the fibers located above this gap can be considered as free-standing fibers with surrounding medium is the air, and therefore, convenient for study their optical properties. Highly uniform fibers with length up to 1 m can be readily fabricated using our approach. Fiber size can be tuned by drawing speed. Figures 3(b)–(d) shows top-down optical images of three fibers with decreasing diameters. The fibers can be employed for high Q factor microlasers and sensors.

Figure 3. (a) Polymer fibers are fabricated by directly drawing from a solution [10]. The gap offers free-standing fibers, convenient for optical characterization. (b)–(d) Top down optical images of various fibers with decreasing size. All fibers exhibit smooth outer surfaces that significant for achievement of high Q factor.

3. OPTICAL MEASUREMENT AND CHARACTERIZATIONS

3.1 Optical setup

To investigate optical properties of our cavities, we use a micro-photoluminescence (μ -PL) whose scheme shown in Figure 4. The excitation source is a common Q-switched Nd:YAG laser that provides green pulses at 532 nm. Linewidth of the pulse is 1 ns and repetition rate is 60 Hz. We construct optical alignment using number of lens and mirrors that enable an excitation spot in a range of 1.5 to 0.35 mm in diameter. The PL emission from the sample can be collected from the top or even from the edge by either 50 $\times$ or 10 $\times$ magnified objectives. The signal beam then splits into two directions, one is delivered to a silicon charge-coupled device (CCD) camera for PL image and the other comes to a

monochromator (spectral resolution about 0.05 nm) connected with a different CCD for spectra recoding. In addition, to prevent any possible damage to the CCD caused high intensity of the excitation laser, we usually use a long-pass filter to block its transmission. To identify polarization characteristic of light laser emission, we use a polarizer that can rotate with high accuracy angles. In this case, the emission is normally collected from the edge instead of from the top.

Figure 4. Micro-photoluminescence (μ -PL) setup for characterization of optical properties of the droplets, hemispheres, and fibers.

3.2 Low threshold microlasers

Microlasers are realized from droplets, hemispheres and fibers upon optical pumping. In this part, we demonstrate a typical case of hemisphere lasers. Figure 5(a) presents optical and PL image of a typical highly circular hemisphere. Under increasing pumping energy, the hemisphere exhibits evidently stronger brightness. Particularly, the intensity is highest at the edge that formed a bright ring-shape. This observation supports the WGM mechanism. Figure 5(b) plots a lasing spectrum from a 27.5 μ m diameter hemisphere. Polarization of the laser light was identified and assigned to

Figure 5. (a) Optical image and its PL image under pulse excitation. The bright ring predict the whispering gallery mode mechanism. (b) PL spectra from a 27.5 μ m-diameter hemisphere. The inset shows the spectrum below and just above lasing threshold.

transverse magnetic (TM) [8]. Using the asymptotic formulas [15] and consider the cavity size is $27.52\text{ }\mu\text{m}$, effective refractive index of the hemisphere and surrounding air are 1.46 and 1, respectively then lasing mode can be well fitted with the first order and mode number 203–205. The inset provides additional information about a revolution from spontaneous emission to lasing emission. The lasing threshold is relatively low that only around $0.37\text{ }\mu\text{J}$ per pulse. It is important to note that the lasing threshold is significantly reduced thanks to the presence of the DBR. Stop-band of the DBR is designed to well-match with the emission of RhB so it prevents optical loss through the substrate, leading to three-dimensional (3D) optical confinement [9]. Very recently, we able to realize quantum dots (QD) lasing by coating CdSe/CdS/ZnS core-multi-shells QD around a solid hemispherical cavity. It is expected that the hemispheres can be employed for realization of microlasers with different active materials such as, organic semiconductor, quantum rods...

3.3 Tunable microlasers via mechanical deformation

PMDS exists in form of semi-liquid so we can draw a line of another solution in it as shown in the Figure 1(a). However, the PDMS can be easily cured by adding a little amount of curing agent (usually ratio 10:1). We leave the sample in the ambient air at room temperature for 2 days and the PDMS becomes solid that can be taken out for its forme. We place and stick it on a movable substrate as shown in Figure 6(a). The mechanical flexibility of PDMS is well-known and thanks to this property, the shape of droplet cavities can be modified by stretching the PDMS film. The modification is shown figures 6(b) and (c) in which a spherical droplet changes to ellipsoid. As the lasing spectrum is related to the cavity size, it is tunable through the mechanical deformation. Suppose the original cavity diameter is D and after deformation is D_f , we can express the deformation ratio as $\Delta D\% = |D_f - D| \times 100\% / D$. Figure 6d plots lasing spectra of a $76\text{ }\mu\text{m}$ diameter droplet as function of $\Delta D\%$. It can be seen that the wavelength can be tune in a range of 0.5 nm for $\Delta D\%$ up to 5% . Even though the wavelength tunable range is limited, it opens a facile method for development of tunable laser.

Figure 6. (a) Scheme of tunable cavity size by mechanical deformation (stretch) [6]. (b) and (c) optical image of a typical droplet without and with deformation, respectively. Tunable optical modes of a $76\text{ }\mu\text{m}$ -diameter sphere versus deformation ratio $\Delta D\%$.

3.4 Coupled fiber lasers and refractive index sensing

Coupled cavity is fascinating structures that has rich novel physical phenomena. One interesting effect offered by a coupled resonator is the Vernier effect that increases the free spectral range (FSR) of adjacent modes, and has been

widely used for optical filters, optical wavelength division multiplexing [16]. Vernier effect enhances only optical modes that full-fill the resonance condition of two isolated cavities while depresses others. Its working principle is schematically plotted in Figure 7. Let's assume the FSR of the first and second cavity are FSR_1 and FSR_2 , matching condition for realization of Vernier effect is $N \times FSR_1 = FSR_2 \times M$, where N and M are co-prime integers. Obviously, FSR of the coupled cavity is determined as $FSR_{12} = N \times FSR_1 = FSR_2 \times M$ [16].

Figure 7. Principle of increase of free spectral range in a couple resonator via Vernier effect. (a)-(c) Optical modes of the first, second, and coupled cavity, respectively.

Characteristic of the Vernier effect is the increase of FSR, and therefore, FSR is a key parameter to verify its presence. For WGM cavities, FSR can be expressed as following:

$$FSR = \frac{\lambda^2}{\pi n D} \quad (1)$$

where, λ is resonant wavelength, n and D are refractive index and cavity diameter.

For big cavities, lasing mode can be extremely dense and FSR is small so it is not convenient to compare the FSR of coupled cavities with FSR of their isolated cavities. There is another way that we can directly confirm the Vernier effect by comparing experimental FSR of a coupled cavity with calculated value based on Vernier equation [17]:

$$FSR_{12} = \frac{\lambda^2}{\pi n (D_2 - D_1)} \quad (2)$$

We have successfully realized the Vernier effect in a coupled fiber lasers. Figure 8a plots a lasing spectrum from a fiber B ($D_B \approx 59 \mu\text{m}$). FSR_B is about 1.4 nm, which is well agreement with the equation (1), assuming $\lambda = 630 \text{ nm}$ and $n = 1.48$. Interestingly, when this fiber is coupled to another fiber (fiber A, $D_A \approx 39 \mu\text{m}$) their FSR (FSR_{AB}) largely increases to 4.4 nm (Figure 8b) and we claim this achievement to the Vernier effect. Indeed, calculated FSR_{AB} using the equation (2) is 4.3 nm that is very close to the experimental data. Furthermore, FSR_{AB} (4.4 nm) is almost triple the FSR_A (1.4 nm), which is well consistent with the Vernier theory.

Recently, application of WGM cavities as biosensors have been achieved intensive attentions due to their ultrasensitive even to single molecules [18]. These sensors are label-free and working based on interaction of sensing molecules with the cavity's surface that leading to spectral shift of resonant modes. For those sensors, both high Q factor and large FSR are highly demand but normally are conflicting in a single cavity. Large FSR is obtained from a small cavity while high

Q factor is possible in a big one. Fascinatingly, we found that this problem can be solved by using a coupled cavity where the Vernier effect enhanced the FRS without reducing cavity size. We have used the coupled fiber lasers for refractive index sensing of ethanol solution and a sensitivity of about 400 nm per refractive index unit (RIU) was achieved, compared to only 210 nm/RIU for a single one. This finding opens a promising direction for improving performance of biosensors.

Figure 8. Lasing spectrum from (a) a single fiber B and (b) a coupled fiber AB. The coupled structure exhibit large free spectral range and reduced lasing modes.

4. CONCLUSION

We have demonstrated feasible methods to fabricate high Q factor cavities including droplets, hemisphere, and fibers based on soft-matter polymers. All are surface tension induced so they have circular shapes and smooth outer surfaces. By doping organic dyes into these structures, optically pumped lasers with low lasing threshold and narrow spectral linewidth are obtained. Quantum dot lasing, which has many advantages such as, temperature stability and color tunable is possible using the hemisphere as a cavity temple. We also present the possibility to have tunable microlasers by using cavity deformation. More interestingly, we have explored the ability to integrate between fiber cavities for increasing FSR via Vernier effect, which enable to achieve single mode operation. The coupled structures offer another advantage that enhanced sensor's sensitivity compared with single one. Our finding is not only meaningful for the development of flexible microlasers and sensors but also important for investigation of light-matter interaction.

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