Flexible whispering gallery mode optical microcavities for lasers and sensors

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**FLEXIBLE WHISPERING GALLERY MODE OPTICAL
MICROCAVITIES FOR LASERS AND SENSORS**

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FLEXIBLE WHISPERING GALLERY MODE OPTICAL MICROCAVITIES FOR LASERS AND SENSORS

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To my wife, Trang

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Abstract

Optical microresonators or microcavities confine light to tiny volumes known as optical modes where the light-matter interactions are strongly enhanced. They are basic building blocks for the investigation of cavity quantum electrodynamics, nonlinear optical effects, and employed as optical switches, optical filters, lasers and sensors. Among different geometries, whispering gallery mode (WGM) cavities, working based on total internal reflection, are alternative structures for realization of low threshold lasers and sensitive sensors thanks to their intrinsically small mode volume and high quality (Q) factor. Up to date, most high Q factor WGM resonators rely on semiconductor techniques requiring costly apparatuses and a number of complex processes. This conventional approach has some limitations: high cost, difficulty for doping gain medium into the cavities and mechanical inflexibility. Therefore, exploiting soft matter that suppresses the above limitations of semiconductor technology is a significant task. In this thesis, we demonstrate our successful exploration of novel soft matter compositions for surface tension-induced high Q factor WGM cavities namely polymer droplets, hemispheres and microfibers. Because the structures are self-assembled, the fabrication is relatively simple. By incorporating organic dye molecules into these structures, lasing with excellent performances such as narrow spectral linewidth, well-defined polarization, clear mode spacing, and strong photostability have been observed under optical excitation. Single mode operation, being important for on-chip applications and optical integrated circuits, is achievable from the three configurations. Especially interesting, the hemisphere and fiber lasers can be used for refractive index sensing for gases and liquids. Complex structures like coupled fiber lasers are also studied for robust single mode operation and improving the sensor's sensitivity. This achievement indicates a full possibility to

integrate between fiber cavities or fiber cavity with fiber waveguide, enabling potential applications of polymer fibers for flexible optical integrated devices. Compared with the traditional semiconductor or inorganic resonators, our cavities have many advantages including low production cost, mechanical flexibility and being straightforward for doping functional materials such as organic molecules or semiconductor nanomaterials. Furthermore, we successfully employ the hemispherical resonators to realize colloidal quantum dot lasing. This new kind of laser is considered as future coherent light source due to their photo/temperature stability and color tunability. The result opens a prospect of using the hemispheres as template cavities for realization of various microlasers based on different gain mediums such as nanocrystal nano/microwires and organic semiconductors. In conclusion, we have contributed promising material compositions and straightforward techniques for creating flexible high Q factor cavities with promising applications as bendable/stretchable microlasers and sensors. Our finding should be also useful for investigation of novel optical nonlinear effects and active/passive components in optical plastic devices/circuits.

List of abbreviations

ASE	Amplified spontaneous emission
C540A	Coumarin 540A
CCD	Charged coupled device
CM	Confocal microscopy
CQD	Colloidal quantum dot
CW	Continuous wave
DBR	Distributed Bragg reflector
DCM	Dichloromethane
DFB	Distributed feedback
FEM	Finite element method
FSR	Free spectral range
LED	Light emitting diode
PDMS	Polydimethylsiloxane
PMMA	Poly(methylmethacrylate)
PPE	Pump pulse energy
PS	Polystyrene
Q	Quality
QW	Quantum well
R19P	Rhodamine 19 Perchlorate
R6G	Rhodamine 6G
RhB	Rhodamine B
RIU	Refractive index unit

SEM	Scanning electron microscopy
TE	Transverse electric
TEM	Transmission electron microscopy
THF	Tetrahydrofuran
TIR	Total internal reflection
TM	Transverse magnetic
TMM	Transfer matrix method
VCSEL	Vertical cavity surface emitting laser
WGM	Whispering gallery mode
μ-PL	Micro-photoluminescence

Chapter 1

Introduction

1.1 Motivation and contribution

The optical and computing technologies, together, have opened up the Information Age with the internet and telecommunication, which has offered unimaginable benefits to our society and significantly improved our daily life quality. Over decades, development of personal and super computer has achieved enormous progresses but today, the computing industry is starting to face two challenging issues: the bandwidth limitations and the heat generation [1]. Could optical technology offer a solution to those problems? This enquiry is fully addressed by a recently published paper on *Nature Photonics* [1], and the answer is probably yes.

Figure 1.1 Optical technology promotes the development of computers to a new level that is with high energy-efficiency and large bandwidth.

The challenges of electronic computers or devices result from using electrons for data transmission/processing. This mechanism is extremely reliable but probably not optimized because of the following reasons (i) it is required to apply electricity that costs energy; (ii) it generates heat because of the electron motion; (iii) it has a bandwidth limitation. In contrast, optical computers or systems use light for information transmission/processing. Light travels by itself without need for any voltage or heat generation. Recent achievements of ultrahigh bandwidth and energy efficiency silicon photonic integrated circuits attest to the key role of optical technology for next generation computers as depicted in Figure 1.1 [1-4].

Electronic computers are based on silicon transistors and metallic tracks. Equivalently, all-optical computers build on optical switches, filters and dielectric waveguides (Table 1.1) [1]. On other hand, optical microcavities are fundamental elements of optical switches/filters [2, 5], and therefore, they play important roles in modern photonic technologies. Furthermore, optical resonators enhance light-matter interaction so they are also significant for studying nonlinear optics such as bistability effect, optical chaos, cavity solitons, strong coupling cavity quantum electrodynamics [6-9] and widely used for lasers and biosensors [10].

	Electronic computers	All-optical computers
Basic elements	Transistors (electronic switches) Metallic tracks	Optical switches, filters Dielectric waveguides

Table 1.1 Comparison of basic elements between electronic and all-optical computers.

Optical cavities exhibit in many configurations including Fabry-Perot [11], photonic crystal [12, 13] and whispering gallery mode (WGM) cavities (Figure 1.2). Among them, WGM resonators are interesting cases because they provide extremely high quality (Q) factor which is important for low threshold microlasers, nonlinear optical effects, and ultrasensitive sensors [14-19].

Figure 1.2 Typical configurations of optical microcavities and their estimated Q factor based on Ref. [5].

Up to date, most of microcavities rely on traditional semiconductor materials with two main fabrication approaches namely top-down and bottom-up [20]. For the top-down technique, microcavities can be achieved by directly etching a planar cavity which is usually epitaxially grown [11]. This technique has some disadvantages including sophisticated epitaxial grown, limitation of material choices due to lattice matching condition, high surface roughness caused by the etching process. For the bottom-up approach, microstructures are normally obtained via self-assembled crystallization of nanostructures [21]. For this method,

surface roughness of the obtained cavities is better compared with the top-down approach but the bottom-up technique also has some drawbacks: (i) it is required to well and precisely control the growing conditions and (ii) the position and the size of the cavities are nearly uncontrollable. Generally, both fabrication methods rely on costly apparatuses and complicated nano/microfabrication.

Compared with semiconductor and inorganic materials, organic polymers are much easier to process and better in mechanical flexibility. They are considered as materials for future photonic devices [22]. Recently, soft matter structures such as organic and polymer cavities, polymer light emitting diodes, self-assembled droplets have been successfully demonstrated as advanced flexible optoelectronic structures which are ultrathin, bendable, and stretchable [23-30]. The development of polymer optoelectronics will lead to numerous amazing devices such as plastic light sources and flexible displays (Figure 1.3).

Figure 1.3 Scheme of a flexible display

Evidently, exploring novel photonic structures, especially optical microcavities, based on soft mater or organic material is significant and interesting work. Particularly, it has been

shown that WGM cavities are very attractive due to their potential applications in both fundamental studies and optical technologies.

Ideally, WGM cavities should have a circular cross-section, and therefore, spheres and cylinders are two best candidates (Figure 1.2). Another important requirement of WGM cavities is smooth outer surface, which prevents radiative loss via elastic scattering. The two factors are fulfilled in surface tension induced cavities (self-assembly formed by surface tension) which automatically form circular cross-section and the surface roughness is, in principle, close to atomic level. Recently, surface tension induced WGM cavities have been fascinated intensive research attentions [16, 17].

For surface tension induced microspheres, liquid droplets are common because they are easy to create. Up to date, water or organic solvents makes most of the droplet cavities [31]. These resonators have high Q factor, and are suitable for laser application. However, their instability and short lifetime due to evaporation hinder the liquid droplets from potential applications. To overcome the above drawbacks is an important task, and promising solutions will open opportunities to employ droplet cavities in optoelectronic devices.

For surface tension induced microcylinders, directly drawing or electrospinning polymer fibers are good examples due to their effective throughput and simple fabrication. However, lasers from these fiber cavities are mainly limited to waveguide effect and F-P cavity [27]. On these works, pieces of fibers are optically pumped and emission light propagated along the fiber axes and two endfaces served as mirrors for optical feedback. Typically, this mechanism suffers from high optical loss because of low reflectivity of the endface reflectors. Therefore, the fiber cavities have low Q factor which is undesired in many applications. WGMs in polymer fibers, which are significant for low threshold and narrow spectral line microlasers, have not been explored. Successful realization of WGMs in

polymer fibers will open opportunities to employ the fibers as highly sensitive optical sensors and alternative components in optical circuits.

This thesis dedicates to develop new type of flexible WGM cavities based on soft matter polymer materials and to investigate their applications as microlasers and sensors. In particular, we concentrate on solving the challenging issues of droplet lasing, which are instability and short evaporative lifetime, and exploring WGM polymer fiber lasers. Following this line, we successfully fabricate three novel flexible WGM microresonators namely polymer droplets, hemispheres, and fibers. These structures are self-assembled by surface tension, thus have almost atomic smooth outer surface and circular cross-section, enabling for achieving high quality factor resonator. By incorporating laser dyes into these cavities, multi and even single longitudinal lasing modes with excellent properties such as narrow spectral linewidth, clear mode spacing and low threshold are observed under optical pumping at room temperature. We have also successfully employed the hemispheres and microfibers to sense refractive index for gases and liquids. Furthermore, a coupling structure consists of two asymmetric fibers has been investigated to obtain single mode emission and to improve sensor's sensitivity. This achievement indicates a full possibility to employ the fibers in optical integrated circuits. Very recently, the solid hemispherical resonators are successfully used for realization of colloidal quantum dot lasing. This new kind of laser is considered as future coherent light sources due to their photo/temperature stability and color tunability. This achievement shows the promise of the hemispheres as template cavities for realization of various microlasers based on different gain media such as semiconductor nano/microwires and organic semiconductor, which is highly important for development of novel light emitting devices.

In summary, our work contributes promising material compositions, straightforward fabrication techniques and unique structures for the development of flexible optical cavities with potential applications as excellent bendable/stretchable microlasers and sensors. Due to high Q factor, our cavities would be used to explore novel optical nonlinear effects and passive/active components in optical plastic devices/circuits.

1.2 Thesis outline

This thesis consists of nine chapters. The first chapter briefly presents the motivation and author's contributions. The next two chapters give basic knowledge of dye lasers and the optical physics of whispering gallery mode cavities. Chapters 4-8 show author's main achievements and the final chapter gives summary and future work.

Chapter 2 introduces a basic principle of a laser device and working principles of its main components, followed by dynamics of dye lasers that includes energy diagram and extension of rate equations.

Chapter 3 describes optical modes of a dielectric microsphere/cylinder and reviews related works on whispering gallery mode microlasers. Firstly, we present characteristics of the optical modes including intensity field distribution, optical loss mechanism, quality factor, mode volume. We also show our own simulated profiles of field intensity of spheres/cylinders using finite element method supported by COMSOL multiphysics. Secondly, we discuss the development of droplet lasing and their current challenging issues. Finally, we briefly describe our optical measurement setup.

Chapter 4 demonstrates a simple but very effective method for fabrication of droplets. The technique allows us to create hundreds of droplets with different sizes via one-step fabrication. By doping laser dyes into these droplets, whispering gallery mode lasing is

observed upon optical excitation. Single longitudinal mode lasing, important for on-chip application, is obtained by reducing droplet size to around 14 μm . Furthermore, the microlasers wavelength can be tuned via mechanical cavity deformation. Our finding provides a versatile approach for stretchable lasers, which is promising for flexible optoelectronic devices. This work has appeared in *Scientific Reports* [32].

Chapter 5 describes an elegant technique for self-assembling hemispherical resonators (from droplets) on a planar dielectric mirror via hydrophobic effect. We show our development of hemisphere fabrication from manual to automatical and precise control (machine operation). By embedding dye molecules into these configurations, for the first time, lasing from hemispherical cavities has been achieved. Compared with droplet lasing shown in the chapter 4, the hemisphere laser herein has higher Q factor due to better light confinement. More interestingly, as the hemispheres are located on a planar surface and surrounding medium is the air, a waveguide can be approached closely to them for effective optical excitation and output laser collection. Due to high stability, long lifetime and easy for output coupling, the hemisphere lasers have overcome all the limitations of conventional liquid droplet lasers. Contents in this chapter have been published in *Advanced Materials* [33] and *Scientific Reports* [34]. Part of this chapter is from manuscript in preparation [35].

Chapter 6 shows a novel method for fabrication of one-dimensional arrays of hemispherical cavities. By doping another polymer (PMMA) into the previous hemisphere's solution, the hemispheres show better lasing performances including higher Q factor, lower lasing threshold and enhanced photostability. This improvement is due to the presence of PMMA which increases effective refractive index of the cavity and, at the same time, protects the dye from oxidation. Owing to the above advantages, the lasers have been successfully employed as refractive index vapor sensors with sensitivity about 130 nm per

refractive index unit. This achievement demonstrates the potential applications of the hemisphere lasers as sensitive chemical and biological sensors. Contents presented in this chapter have appeared in *Applied Physics Letters* [36].

Chapter 7 describes polymer microfibers fabricated via directly drawing from a solution embedding with dye molecules. Under optical pumping, whispering gallery mode lasing with narrow spectral linewidth (0.08 nm) and good photostability is achieved. Particularly, the single mode emission, which is important for integrated photonics and on-chip application, is possible in ~ 10 μm -diameter fibers. The fiber lasers can be used for refractive index sensing and sensitivity up to 300 nm per refractive index unit is obtained. Content in this chapter has been published in *Laser & Photonics Reviews* [37] and was selected for a Frontispiece image.

Chapter 8 investigates coupled fiber lasers. By coupling two asymmetric laser cavities, single mode emission is obtained via the Vernier effect. Compared with an isolated cavity, the coupled structure has two main advantages: single mode emission with higher cavity Q factor and higher sensor's sensitivity. The coupled cavity offers a solution to achieve two desired factors, large free spectral range or mode spacing and high Q factor, which are not possible in a single whispering gallery mode cavity. Furthermore, compared with other coupled cavities such as microdisks, microrings, our coupling fibers have several advantages in term of easy fabrication and alignment. This work has been appeared on *Advanced Optical Materials* [38] and selected for inside cover of the March 2014 issue [39].

Chapter 9 summarizes author's main contributions and presents perspective investigations. In the future works, firstly, we show our preliminary results of quantum dot lasers using the hemispheres as template resonators. Compared with dye laser, quantum dot laser is better in color tunability, temperature and photoluminescence stability. Secondly, we

suggest an indirect electrical pumping approach for achieving hemisphere lasers. Finally, we propose some complex integrated structures based on our cavities to realize their potential applications in optical and photonic devices.

List of publications shows the author's scientific contributions including journal publications, a patent, a book chapter, and conference proceedings/presentations.

Chapter 2

Laser principle and dynamics of dye lasers

2.1 Basic principle of lasers

2.1.1 Introduction

Laser is acronym for “light amplification by stimulated emission of radiation”. Laser principle was developed about six decades ago by American scientist C. H. Townes and Soviet physicists N.G. Basov and A.M. Prokhorov, and they were awarded Nobel Prize in physics (1964) for their breakthrough. However, T.H. Maiman was the first person who successfully fabricated a real laser. He described his invention in *Nature* journal with the title “*Stimulated Optical Radiation in Ruby*” [40] in 1960. Since then, enormous progress has been achieved in laser technology. Up to date, hundreds kinds of lasers have been invented such as solid-state (Neodymium, Yb: YAG, Titanium sapphire), semiconductor, gas (Helium-Neon, CO₂, CO), liquid (organic dye) lasers [41]. Today, lasers are well-recognized as one of the most essential devices in modern life due to their unique coherence (both spatial and temporal) properties, high directionality, strong brightness. Lasers have wide range applications in optical communications, information processing, common electronics (printers, barcode scanners), medical surgery, industry (welding and cutting materials) as well as scientific research.

2.1.2 Spontaneous and stimulated emission

The exceptional properties of laser light originally come from a process known as stimulated emission that was proposed, for the first time, by Einstein in 1917. For simplicity, let consider an atom with only two energy levels, level 1 and level 2 that stands for a ground

state and an excited state, respectively. Their energies are E_1 and E_2 . Generally, the atom is located at the lowest energy level, the ground state, but under excitation, it will absorb the provided energy and jump to the excited state. This transition is known as *absorption* (Figure 2.1a). Now, the atom stays at the excited state, which is not stable, so it quickly decays to the ground state. The decay can be either nonradiative (no photon is emitted, and the energy is transferred to, for example, heat) or radiative (known as *spontaneous emission*), producing a photon (Figure 2.1b) with frequency ν :

$$\nu = \frac{E_2 - E_1}{h} \quad (2.1)$$

where h is Planck's constant. This process is characterized by Einstein's coefficient A_{21} . The photon produced via the spontaneous emission can be in all directions and with random phases, and therefore, spontaneous emission is a loss process and not useful in a laser device. The situation will be more interesting if there is incident photon. This photon will force the atom undergo to the ground state via the radiative decay, known as *stimulated emission*, characterized by Einstein's coefficient B_{21} . Unlike the spontaneous emission, this process produces two photons (one is the incident photon) and they have the same phase and

Figure 2.1 (a)-(c) Schematic diagrams of absorption, spontaneous and stimulated emission, respectively.

direction (Figure 2.2c). The stimulated emission is necessary but not enough for laser generation. To achieve lasing requires a cavity/resonator which provides optical feedback to amplify the stimulated emission. Laser starts to appear when the number of produced photons equal to total number of loss photons within the same unit time and this state is referred to lasing threshold. The role of a cavity is very important because it provides the incident photons mentioned above and amplifies them via stimulated emission. Comprehensive principle of laser devices are available in many books, for instance, Refs [41, 42].

2.1.3 Main components of a laser device

Typically, a laser has three main components (Figure 2.2): a pumping source (electric or optical pump), a gain medium/material, and a cavity. The pumping element supplies energy to the laser system by excited atoms/molecules of the gain medium to excited states. Then, these atoms/molecules decay to ground states and produce light. In other words, pumping energy is converted to light via the gain. The optical cavity or resonator then provides optical feedback to amplify the emitting photons. A common resonator structure is the Fabry-Perot cavity that consists of two mirrors (Figure 2.2). One mirror has very high reflectivity while the other is partially transparent for laser light output, and therefore, usually called output coupler.

Figure 2.2 Scheme of a laser device

2.1.4 Four-level laser diagram

Laser emission is only possible when population inversion occurs. That means that the number of carriers in the upper state is larger than that of the lower state. This phenomenon, however, is not achievable for two-level (Figure 2.1) but three/four-level. Dynamics of dye lasers is generally described as the four-level system [43, 44] (Figure 2.3), which is characterized by two fast transition processes, $3 \rightarrow 2$ and $1 \rightarrow 0$. As a result, we can assume population of dye molecules at level 3 (N_3) and level 1 (N_1) close to zero [41], and therefore, the inversion population ($N_2 > N_1$) can easily be obtained by small number population N_2 . To describe the laser dynamics, a simple way is to use rate equations combined with the laser diagram. In the next sections, we show the energy scheme of organic dyes and rate equations of dye lasers.

Figure 2.3 Schematic diagram of four-level laser.

2.2 Dynamics of dye lasers

Comprehensive dynamics and characteristics of dye lasers can be found in several review papers [44-48] and their wide range applications are available in number of books [49, 50].

Below, we briefly introduce two important characteristic of dye lasers: their energy levels and extension of rate equations.

2.2.1 Energy level of organic dyes

Compared with solid state and gas laser, the dye lasers have broader emission so they are frequently used as tunable wavelength lasers. This characteristic is due to complex band structures of organic dyes (Figure 2.4) [44]. It consists of singlet and triplet states and in each state, there are many vibrational and rotational levels. The ground state is the lowest single state. At thermal equilibrium, all dye molecules are in the ground state, denoted as level 0 of the four-level laser diagram. Under excitation, these molecules absorb energy and jump to higher level of the excited singlet states (level 3), then it quickly relaxes to lower level of the excited single states (level 2). At this state, molecules can (i) go to higher level S_2 (known as *excited state absorption*); (ii) decay to low level of the triplet states (T_1) via *intersystem crossing*, characterized by rate constant K_{ST} ; (iii) decay to high level of the first single states (level 1) by either spontaneous emission (known as *fluorescence*) characterized by fluorescence lifetime τ or stimulated emission. Molecules at level 1 can quickly go to the bottom of ground state through nonradiative processes. For molecules at T_1 , there are two possibilities: (a) it can either jump to excited triplet states by absorbing excitation energy or laser emission itself. This process is referred to *triplet state absorption* [50]; (b) it decays to the ground state via either radiative (known as *phosphorescence*) or nonradiative transition, characterized by lifetime τ_T . It is important to note that $\tau \ll \tau_T$ because transitions within singlet state are spin-allow while the transitions from single to triplet are spin-forbidden [44].

Figure 2.4 Schematic diagram of energy levels of a typical organic dye molecule based on Ref. [44]. The intense and light horizontal lines indicate vibrational and rotational states, respectively.

2.2.2 Extension of rate equations

In a real case, the *excited state absorption* and *triplet state absorption* can be significantly averted by pumping process. Therefore, for simplicity, we ignore it in the rate equations. And notice that population at levels 3 (N_3) and 1 (N_1) are considered being zero ($N_3 \approx N_1 \approx 0$) so rate equations are required only for population of the upper laser state (N_2), ground state (N_0) and first triplet state T_1 (N_T) as following expressions:

$$\frac{dN_2}{dt} = B_{03}\rho_{pump}N_0 - \frac{N_2}{\tau} - N_2k_{ST} - B_{21}N_2\rho_{laser} \quad (2.2)$$

$$\frac{dN_T}{dt} = k_{ST}N_2 - \frac{N_T}{\tau_T} \quad (2.3)$$

$$\frac{dN_0}{dt} = -B_{03}N_0\rho_{pump} + \frac{N_T}{\tau_T} + \frac{N_2}{\tau} + B_{21}N_2\rho_{laser} \quad (2.4)$$

where, ρ_{pump} is pumping rate. ρ_{laser} is referred to laser intensity related to the stimulated emission. B_{ij} are Einstein stimulated emission or stimulated absorption coefficients. Note that the total population of dye molecules is a constant. Now, considering τ_p is the photon lifetime in the cavity laser then we have:

$$\frac{d\rho_{laser}}{dt} = B_{21}N_2\rho_{laser} - \frac{\rho_{laser}}{\tau_p} \quad (2.5)$$

In the equation (2.5), the first term of the right part correspond to photon production via stimulated emission while the second term is referred to optical loss of the cavity.

2.3 Summary

In conclusion, we have presented the basic knowledge of a laser device by introducing their major optical processes including spontaneous emission, stimulated emission, and population inversion. Laser components are briefly discussed and followed by dynamics of dye lasers. The knowledge of this chapter is meaningful for later investigation of dye lasers in our microcavities.

Chapter 3

Whispering gallery mode cavities and lasers

3.1 Introduction

In chapter 1, we have pointed out that WGM cavities serve as alternative structures for low threshold microlasers and ultrasensitive biosensors. Here, we discuss in depth the fundamental physics of WGMs and their important characteristics.

The expression “*whispering gallery*” was originally given by Lord Rayleigh in 1912 [51]. He used the above phrase to explain a fascinating phenomenon that he observed himself in the Saint Paul’s cathedral located in London where a person who whispered along the wall would hear his/her own voice after some time [52]. This effect was ascribed to the propagating of acoustical waves around of the dome of the cathedral. Later, it was found that electromagnetic waves in a dielectric sphere exhibited similar effect. Indeed, light can propagate along a circular cross-section of a sphere by multi total internal reflections (TIR) (Figure 3.1a). Furthermore, supposing there is a light source on the sphere that continuously emits light then this wave can constructive interfere with itself if it has the same phase after

Figure 3.1 (a) Light is trapped within a dielectric sphere/cylinder by multiple total internal reflections and then is amplified to form whispering gallery modes (b).

travelling one round. When it happens, light is amplified by resonant recirculation and the resonance is known as whispering gallery modes (WGMs), depicted in Figure 3.1b.

Understanding the WGMs, which is the aim of this chapter, is necessary to explore it for applications. We start from the Helmholtz equation that can provide description of optical

transition in a dielectric medium:
$$\nabla^2 E + k^2 E = 0 \quad (3.1)$$

where, ∇ is the Laplacian operator, E is electric field, k is wave number which has a relationship with refractive index (n), speed of light in vacuum (c), and angular frequency (ω)

and as follow:
$$k = n \frac{\omega}{c} \quad (3.2)$$

To get characteristic equations of optical modes in a dielectric sphere or cylinder, we need to solve the Helmholtz equation in the spherical or cylindrical coordinates. In the following sections, characteristics of WGMs including field intensity distribution, resonant positions, optical loss mechanism and effective mode volume are discussed.

3.2 Optical modes of a dielectric microsphere

3.2.1 Characteristic equations

Optical modes of a dielectric sphere have been studied for decades by several authors [52-55]. A comprehensive knowledge can be also found in recent review papers [56-58]. Here, we briefly present the description based on Ref. [59]. The Laplacian operation in spherical coordinates (Figure 3.2) is given:

$$\nabla = \frac{1}{r^2} \frac{\partial^2}{\partial r^2} \left(r^2 \frac{\partial}{\partial r} \right) + \frac{1}{r^2 \sin^2 \phi} \frac{\partial^2}{\partial \theta^2} + \frac{1}{r^2 \sin \phi} \frac{\partial}{\partial \theta} \left(\sin \phi \frac{\partial}{\partial \theta} \right) \quad (3.3)$$

where, r is the radius of the sphere, θ and ϕ are the polar and azimuthal angles. The amplitudes of either electric (E) or magnetic (B) fields inside the sphere are given in terms of the Bessel function of the first kind $j_l(x)$ and the generalized Legendre polynomial $P_l^m(x)$:

$$\begin{pmatrix} E_r^{in} \\ E_\theta^{in} \\ E_\phi^{in} \end{pmatrix} = \begin{pmatrix} a_1 \\ a_2 \\ a_3 \end{pmatrix} j_l(k_{in}r) P_l^m(\cos\theta) e^{im\phi} \quad (3.4)$$

where, E_r, E_θ, E_ϕ are the components of electric field, l and m are the angular and azimuthal mode numbers, a_1, a_2, a_3 are coefficients. Components of magnetic field (B_r, B_θ, B_ϕ) can be similarly expressed as the equation (3.4) but with different coefficients.

Figure 3.2 Path of light in a dielectric sphere with spherical coordinates based on Ref. [59]

Outside the sphere, the fields are evanescent waves and described by the Hankel function of the first kind $h_l(x)$:

$$\begin{pmatrix} E_r^{out} \\ E_\theta^{out} \\ E_\phi^{out} \end{pmatrix} = \begin{pmatrix} b_1 \\ b_2 \\ b_3 \end{pmatrix} h_l(k_{out}r) P_l^m(\cos\theta) e^{im\phi} \quad (3.5)$$

Considering the Maxwell boundary conditions

$$\begin{cases} E(B)_{\theta}^{in} = E(B)_{\theta}^{out} \\ E(B)_{\phi}^{in} = E(B)_{\phi}^{out} \end{cases} \quad (3.6)$$

and notice the definition of transverse electric (TE) mode $E_r^{in} = F_r^{out} = 0$ and transverse magnetic (TM) mode $B_r^{in} = B_r^{out} = 0$, the solution for TE polarization is given:

$$H_l'(k_{out}R)J_l(k_{in}R) = nJ_l'(k_{in}R)H_l(k_{out}R) \quad (3.7)$$

$$\text{and for the TM polarization: } nH_l'(k_{out}R)J_l(k_{in}R) = J_l'(k_{in}R)H_l(k_{out}R) \quad (3.8)$$

where $J_l(x) = xj_l(x)$ and $H_l(x) = xh_l(x)$ with ' is refer to derivative, R is radius the sphere.

Equations (3.7) and (3.8) are characteristic formulas for TE and TM optical modes of a dielectric microsphere.

3.2.2 Intensity field distribution of whispering gallery modes

Typically, the characteristic equations for WGMs of a microsphere can be obtained by matching the Maxwell boundary conditions. From these equations, the intensity distribution of WGMs can be derived by either analytical approach [60] or numerical calculation [61-63]. It depends on the polarization (TE or TM modes) and three quantum integer numbers: q , m , and l . q is number of peaks in the radial direction, m is number of field maximum inside and along the circular boundary between the sphere and the outside medium; $l-m+1$ is number of the peaks in the azimuthal direction [64].

Following the method that was carefully described in Refs. [63, 65], we perform the field distribution of WGMs in a cavity using finite element method (FEM) which is available in a commercial COMSOL Multiphysics. Figure 3.3 show magnetic intensity profiles of

WGMs in a 10 μm -radius dielectric sphere as function of three quantum numbers q , l and m . Generally, the first and second order fundamental modes ($l = m$, $q = 1$ or 2) are the most observation in experiments, and therefore, we only focus on these modes. Figure 3.4 plots the intensity profiles of first order WGM versus increasing radius of the sphere. The mode

Figure 3.3 Magnetic intensity profiles of whispering gallery modes of a 10 μm -radius sphere with TE polarization. (a) and (b) first and second radial mode order. (c) and (d) first radial order but different polar mode number.

Figure 3.4 Magnetic intensity profiles for sphere cavities with increasing radius. The numerical simulation for TE polarization and wavelength near 590 nm.

profile expands in the polar direction following the increase of the curvature. Nevertheless, ratio between cavity size and the mode area significantly increases, which denotes large mode number m in a big cavity.

To get a better view of field distribution outside the sphere (evanescence wave), we show in Figure 3.5 the intensity in the radial direction for the two fundamental order WGMs. There is a tiny fraction ($\sim 1\%$) of total energy is located outside the cavity indicated good optical confinement.

Figure 3.5 Intensity distributions in the radial direction of a $5\text{ }\mu\text{m}$ - radius microsphere for the first and second radial mode order. Refractive index of the sphere and outside medium is 1.5 and 1, respectively.

3.3 Optical modes of a dielectric microcylinder

Similarly, for a dielectric cylinder, description of whispering gallery modes can be derived by solving the Helmholtz equation in the cylindrical coordinate. Particularly, can see Ref. [66] for more information. Here, we briefly show the approach and solution based on Ref. [59].

The Laplacian operation in spherical coordinates is given:

$$\nabla = \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial}{\partial r} \right) + \frac{1}{r^2} \frac{\partial^2}{\partial \theta^2} + \frac{\partial^2}{\partial z^2} \quad (3.9)$$

Where r, θ, z are cylindrical coordinates (Figure 3.6)

Figure 3.6 Cylindrical coordinates.

Consider an infinite cylinder with radius R and refractive index n and assuming that electric or magnetic field F_z is supposed independent on θ , which can be expressed as

$$F_z(r) = J_0(\gamma r) \text{ for } r \leq R; \quad F_z(r) = AK_0(\beta r) \text{ for } r \geq R \quad (3.10)$$

where, J_0 and K_0 are the Bessel and modified Bessel function, respectively. A is a constant.

$$\gamma = \left(\frac{\omega^2 n^2}{c^2} - k_z^2 \right)^{1/2}, \quad \beta = \left(k_z^2 - \frac{\omega^2}{c^2} \right)^{1/2}$$

where, k_z is wavevector of light along the axis (z) of the cylinder.

The solution, for example, for TM polarization is given

$$-\frac{n^2 J_1(\gamma R)}{\gamma J_0(\gamma R)} = \frac{K_1(\beta R)}{\beta K_0(\beta R)} \quad (3.11)$$

Field intensity distribution in a dielectric microcylinder can also found by using FEM technique as demonstrated in Figure 3.7.

Figure 3.7 Magnetic intensity profiles of whispering gallery modes of a 10 μm -radius microfiber with TM polarization.

3.4 Asymptotic formulas

To find the resonant wavelengths from a spherical cavity, we can use the asymptotic formulas derived by Lam et al [67]. The author found that the resonance positions can be characterized as a series in $v^{-1/3} = (m + 1/2)^{-1/3}$, where m is the mode number. Let us consider the resonance modes in wavelength, the expansions are given [64]:

$$(\lambda_m^q)^{-1} = \frac{1}{\pi D n_1} \left[v + 2^{-1/3} A_q v^{1/3} - \frac{P}{\sqrt{n_r^2 - 1}} + \frac{2^{-2/3} A_q^2 v^{-1/3}}{10/3} - \frac{P \left(n_r^2 - \frac{2}{3} P^2 \right) A_q}{2^{1/3} (n_r^2 - 1)^{3/2} v^{2/3}} \right] \quad (3.12)$$

In this equation $n_r = n_1/n_2$, n_1 and n_2 are refractive index of the sphere and the outside medium, respectively. A_q is the roots of the Airy function. $A_q = 2.338, 4.088, 5.521$ for $q = 1, 2, 3$, respectively. P is a coefficient related to the polarization characteristic and given by:

$$P = n_r^{-1} = n_2/n_1 \text{ for TM modes, } P = n_r = n_1/n_2 \text{ for TE modes} \quad (3.13)$$

The equation (3.12) is applicable for either a sphere or a cylinder because they share the same two-dimensional (2D) circular cross-section.

The free spectral range (FSR) or mode spacing of the cavity, is defined as the distance between successive mode number m , can be derived from the asymptotic formulas.

$$FSR^q = \lambda_m^q - \lambda_{m+1}^q \quad (3.14)$$

For the first order ($q = 1$), the FSR can be approximately estimated as follow:

$$FSR \approx \frac{\lambda^2}{\pi n D} \quad (3.15)$$

It can be seen from Figure 3.8, the FSR is proportional to inverse of cavity diameter.

Figure 3.8 FSR as a function of cavity size

3.5 Quality factor and mode volume

Among many characteristic parameters of a microcavity, quality (Q) factor and mode volume (V) are basic and the most important ones.

3.5.1 Quality factor

Q factor characterizes the life time (τ) of light energy in the cavity mode [56]. In general form, Q factor is given

$$Q = \omega\tau \quad (3.16)$$

Q factor of optical modes in a WGC is a function of several loss factors and can be determined as below [68]:

$$Q^{-1} = Q_{cont}^{-1} + Q_{scatt}^{-1} + Q_{mat}^{-1} + Q_{rad}^{-1} \quad (3.17)$$

where, Q_{cont} refers to losses due to surface contaminants; Q_{scatt} denotes scattering losses inside the material and inhomogeneous of the cavity's surface; Q_{mat} is intrinsic material loss; Q_{rad} is radiative losses.

The losses introduced by surface contaminants, such as the presence of water molecules (diffusion from atmosphere) in cavity surface has strong effect on the upper limit for the intrinsic Q factor [58].

The scattering loss is mainly due to Rayleigh scattering and can be estimated based on the two parameters of surface inhomogeneities: the correlation length B and root-mean-square (rms) size σ [68].

$$Q_{scatt} = \frac{\lambda^2 D}{2\pi^2 \sigma^2 B} \quad (3.18)$$

It is clearly that Q_{scatt} linearly increases with cavity size. Let us consider a sphere with fixed diameter $D = 100 \mu\text{m}$, and $B = 5 \text{ nm}$, we have a relationship between Q_{scatt} and σ as plotted in the Figure 3.9. The increase of surface roughness of the cavity strongly affects the Q_{scatt} factor. For instance, with $\sigma = 0.5$ and 2 nm , Q_{scatt} are about 4.6×10^9 and 2.9×10^8 (one order magnitude smaller).

Figure 3.9 Q_{scatt} versus the rms size of surface inhomogeneities

Q_{mat} is limit due to absorption losses of the material and also the Rayleigh scattering.

It can be estimated as following expression:

$$Q_{mat} = \frac{2\pi n}{\alpha \lambda} \quad (3.19)$$

For poly(methylmethacrylate) or PMMA, the material used in our work, the optical attenuation at 600 nm can be down to 0.12 dB/m, refractive index $n = 1.49$, one can be expected $Q_{mat} \approx 10^8$.

Q_{rad} denotes intrinsic radiative decay due to curvature of the cavity. Whispering gallery modes are result of TIR on a curved surface so it has some optical loss.

Q_{rad} is defined as a ratio between real and imaginary part of the wave number [52]:

$$Q_{rad} = \frac{\text{Re}(k_{q,l})}{\text{Im}(k_{q,l})} \quad (3.20)$$

For $l = m$, Q_{mat} can be expressed as follows [62]:

$$Q_{rad} = \frac{1}{2} \left(m + \frac{1}{2} \right) n^{-(1-2p)} (n^2 - 1)^{\frac{1}{2}} e^{2T_{q,m}} \quad (3.21)$$

$$\text{where,} \quad T_{q,m} = \left(m + \frac{1}{2} \right) (\beta_{q,m} - \tanh \beta_{q,m}) \quad (3.22)$$

$$\beta_{q,m} = \arccos h \left\{ n \left[1 - \frac{1}{m+1/2} \left(A_q \left(\frac{2m+1}{4} \right)^{1/3} + \frac{n^{1-2p}}{\sqrt{n^2-1}} \right) \right]^{-1} \right\} \quad (3.23)$$

$p = 1$ and 0 for TM and TE polarization, respectively.

Q_{rad} depends on q and m . High order mode has lower Q_{rad} and a larger cavity has higher Q_{rad} . For $m = 140$, estimation of Q_{rad} based on the equation (3.21) is about 10^{35} .

3.5.2 Mode volume

Mode volume gives information about field localization. However, it is more convenient to use effective mode volume (V_{eff}) for characteristic of a mode [52]:

$$V_{eff} = \frac{\int E_{\kappa\eta}^2(r) dr}{E_{\kappa\eta}^2(r_{max})} \quad (3.24)$$

where κ is a function of r , θ , φ that denotes the field projection; η is associated with quantum

numbers q , l , m . Approximately, replacing the Bessel function by the Airy function, a simple form of V_{eff} can be obtained for electric modes as following [52]:

$$V_{eff} = \frac{2.639}{m^{1.142}} R^3 \quad (3.25)$$

For WGCs, the V_{eff} is small, which is significant for studying light matter interaction.

3.6 Whispering gallery mode microlasers

Stimulated emission was observed from a sphere for many decades ago [69]. However, dye laser from droplets was not achieved until 1980s [70-72]. Later, droplet cavities were further investigated for nonlinear optics such as stimulated Raman and Brillouin scattering [73, 74]. Since then, droplets have been attracted increasing research attentions due to their extremely high Q factor and simple creation [31]. To create droplet cavities, several techniques have been adopted. At initial stage, droplets were generated from a piezoelectric droplet generation, and they were free-falling (acceleration $g = 9.81 \text{ m/s}^2$). For this method, droplets move downward with high speed so it is inconvenient for optical characterizations. To avoid this problem and make the system more stable for measurement, several techniques have been developed, for instance, creating levitated droplets (free-standing) by trapping it in the air via a complex optical tweezers [75] or generating droplets on a plane surface using a hydrophobic substrate [76-78]. Despite those improvements, a droplet cavity generally has a

short evaporative lifetime. This drawback limits droplet cavities as a laboratory tool. In order to employ droplet cavities for real applications, the lifetime issue needs to be addressed.

Recently, droplet lifetime has been significantly improved thanks to microfluidic technology. In this idea the surrounding medium (air) is replaced by another liquid. That means liquid droplets are integrated in a microfluidic channel which carries another immiscible liquid [64, 79]. Based on this technique, droplet cavities have successfully made a transformation from a laboratory tool to real applications, for example, droplet lasers have been demonstrated as optical switches [80, 81] and bio/chemical sensors [82]. Nevertheless, there is room for improvements, to make them more suitable for specific purposes. Below are some suggestions. (i) Droplets fabricated by a microchannel typical have large size (tens of microns) that is not possible for single mode emission. (ii) Single microfluidic channel can create only one droplet's size so it is not convenient for investigation of size dependent lasing characteristics. (iii) Mobility of droplets (due to fluidic flow) leading to poor handling feasibility, and therefore, difficult for constructing optical devices. (iv) The instability of droplets causes difficulty in coupling out the emission from droplets via a waveguide. In this thesis, the above issues are solved one by one.

Besides the self-assembled droplets, there are other interesting flexible self-assembled structures: polymer fibers. However, up to date, most of nano/microfibers are mainly applied as waveguides while WGM lasing based on the circular cross-section of the fibers has not been explored. We have devoted our efforts for filling this gap and successfully realized, for the first time, high quality WGM lasing from directly drawing polymer fibers. Applications of fiber lasers for refractive index sensing and achievement of single mode emission via the Vernier effect are also demonstrated.

3.7 Optical measurements

We used a micro-photoluminescence (μ -PL) system whose images are shown in Figure 3.10 and schematical diagram is presented in Figure 3.11 to study optical properties of our structures. The pumping source was a Nd:YAG laser that generates green pulse at 532 nm, pulse duration of 1 ns and repetition rate of 60 Hz. The pump laser has linear polarization, which is parallel to the ground. We guided the excitation laser to the sample using series of mirrors and lens with incident angle of $\sim 45^\circ$ to the surface of the sample. The excitation spot has an ellipse shape whose size can be 0.3×0.4 to 1×1.2 mm that depends on optical alignment. Emission from the sample was collected on the top using an objective (50 $\times$, NA = 0.42) and the signal was further delivered to a monochromator linked to a charged coupled device (CCD) for spectrum recoding. Spectral resolution is about 0.05 nm and noise level of the CCD is around 6 e-rms at 100 kHz readout. To prevent damage of CCD caused by high intensity of the pumping laser, a long-pass filter was frequently used to prevent transmission of the green wavelength during the measurement.

Figure 3.10 Micro-photoluminescence systems. (a) Microscopy and (b) Monochromator integrated with fiber coupler and CCD.

Figure 3.11 Scheme of the micro-photoluminescence systems for optical characterization of droplet/hemisphere/fiber lasing. The XYZ movable state enables the sample to freely-move in three-dimensional space.

3.8 Summary

We have presented the characteristic equations of WGMs in a dielectric microsphere/cylinder. Intensity field distribution and position of optical modes are discussed. Using FEM supported by COMSOL multiphysics, we demonstrate the profiles of field intensity in a WGM cavity, which provides information of two most important parameters of WGM cavities, quality

factor and mode volume. Subsequently, we reviews related works on whispering gallery mode microlasers including their development and current challenging problems. In the last section, we briefly demonstrate our optical measurement setup for optical characterizations of all our structures.

Chapter 4

Tunable lasers based on self-assembled polymer droplets

4.1 Introduction¹

In the Chapter 3, we have shown that even though droplets in liquid microfluidic channel have many advantages over droplets in the air, they have their own drawbacks such as the mobility of droplets leading to vast difficulties in constructing and operating optical systems. To solve this issue, the idea of replacing liquid by elastic polymer was suggested [83, 84]. Furthermore, the authors also applied inject print technique to create small droplets with diameter down to about 10 μm . However, on these works, the cavity material was still liquid (organic solvent). The solvent was found to be slowly leaked into the outside medium, thus dis enabling for long time operation. To avoid this issue, we do not use liquid solvent but organic polymer to create droplet cavities. In this chapter, we demonstrate polymer droplets with varied sizes in a polymer elastomer. Our fabrication technique is simple which is based on a single process but very effective [32]. The diameter (D) of droplet cavities can be easily tuned from 5 to 200 μm , thus enables for size dependent characteristics and single mode operation. In addition, owing to the original elastic properties of both the cavity and carrier, cavity size hence lasing wavelengths can be modified under mechanical deformation. This finding is significant for the development of tunable lasers. The change of lasing modes with the cavity shape would be also useful for strain or force sensing.

¹ Content in this chapter has appeared in "Tuning Whispering Gallery Mode Lasing from Self-Assembled Polymer Droplets," [Sci. Rep.](#) **3**, 1362 (2013).

4.2 Self-assembled polymer microdroplets

The host material of droplets is a mixture of polystyrene (PS) and epoxy. We made this solution by subsequently mixing 14 wt% PS (molecular weight $m_w \sim 2000$) in 99.76% purity dichloromethane (DCM) with epoxy resin (Araldite[®] 506). The epoxy was dominated in the solution with weight ratio over PS is about (12 - 15): 1. The epoxy is needed for the self-assembled process due to its large surface tension. PS has a higher refractive index up to 1.59 at 630 nm [85] is believed to increase cavity index, and therefore, improve the Q factor of droplets.

For lasing generation, the lasing dye Rhodamine B (RhB), chemical formula $C_{28}H_{31}ClN_2O_3$ (Figure 4.1a), was doped in the polymer solution with concentration about 2 wt%. RhB has a wide wavelength emission from about 540 to 650 nm (Figure 4.1b).

Figure 4.1 (a) RhB molecular structure based on product information of Sigma-Aldrich and (b) photoluminescence emission of RhB (in DCM).

Figure 4.2 plots the schematic diagram of a direct drawing technique and self-assembled droplets in polydimethylsiloxane (PDMS) (fulfilled in a glass holder). We started to coat a little amount of solution outside a sharp tip of a metal rod (a needle). This step can be done by simply dipping the tip into and then retracted from the dye-doped solution (Figure 4.2a). After that, we vertically dipped the tip in the PDMS and subsequently moved across a

line (Figure 4.2b). The movement is done rapidly. The tip leaves its coating solution into the PDMS, creating a fiber-shape with gradual decrease of radius from the starting (S) position to the ending (E) position. After a minute, this structure breaks down to form numerous droplets spontaneously because (i) the surface tension of solution tends to minimize its energy by minimizing surface area and (ii) a sphere has the smallest surface area compared with other configuration with the same volume. Top-down

Figure 4.2 Self-assembled polymer droplets in a polymer carrier. (a) - (d) A direct drawing technique and illustration of the self-assembled process. (S) and (E) are starting and ending drawing points. (e) - (j) Typical self-assembled process with time. Droplet size gradually decreases from S to E. Droplets with diameters around 10 μm are obtained near the ending region (k). All optical images were captured using a 10× objective under halogen illumination.

view of a typical self-assembled process is presented in Figures 4.2e-j where the transformation from the line-shape to droplets is clearly observed. As expected, sizes of droplets normally decrease from S to E (Figures 4.2c-d) so series of droplets with small diameters about 10 μm can be found near the E region (Figure 4.2k). For these droplets, lasing mode spacing is large, which is suitable for single mode emission. It is noted that the droplets are fully covered by the PDMS. Furthermore, the PDMS is easily cured, fixing the position and size of the droplet embedded, which leads to stable and robust droplet lasers, thus enabling steady operation and applications. These are advantageous of our approach compared to the state-of-the-art water or organic liquid cavities which are unstable and have short evaporative lifetime.

4.3 Optical characterizations of droplet lasers

4.3.1 Whispering gallery mode lasers

Under laser pump, droplet emits light that can be captured by the CCD camera. From the PL images as shown in Figures 4.3a and 4.3b, there are two bright rims opposite to the droplet center while the intensity at other part of the droplet is relatively weak. This observation gives a clue that the optical loop may be located in vertical plane as depicted in Figure 4.3c instead of horizontal plane. Otherwise, brightness should come from the circumference of this droplet similar to hemispherical microlaser [33].

Figure 4.4 plots emission from a 24 μm RhB-doped droplet under pulse energy of 1.35 μJ . Two separated lasing envelopes were observed which are theoretically ascribed to TE and TM modes. Using the asymptotic solutions from Chapter 3 and assume (i) diameter of droplet is 24.22 μm , (ii) refractive index of droplet and PDMS are 1.53 and 1.41 [86],

respectively the lasing modes can be well-fitted with the first order $q = 1$ and mode numbers $m = 174$ -178 as shown below the spectrum. Therefore, we conclude that lasing mechanism is attributed to WGMs. Owing to smooth outer surface, the Q factor of droplet lasing is about 5×10^3 and spectral linewidth of 0.13 nm (the inset of Figure 4.4).

Figure 4.3 Cavity loop in a typical droplet. (a) and (b) Images of a droplet upon pulse excitation. The white cross indicates the pumping position. (c) Scheme of WGMs (emitting patterns) inside a droplet under green excitation beam with vertical electric field (depicted by the arrows).

Lasing threshold is, however, not the same for TE and TM polarization. Integrated PL intensities of TE and TM lasing modes versus excitation energy are different. The nonlinear increase of PL intensity with pumping energy indicates lasing action and the threshold for TE mode is 125 nJ, which is lower than that of 195 nJ for TM polarization (Figure 4.5a). This difference might due to less optical loss of TE emission compared with TM light, which has been reported in liquid droplet lasing [72]. From this observation, it is expected that if the pulse energy is higher than threshold for TE mode but lower than threshold for TM mode,

there should be only TE emission. We observed this effect as shown in Figure 4.5b, only TE modes, mode numbers $m = 175$ -178, were generated at pulse energy of 170 nJ.

Figure 4.4 Emission and polarization characteristics of a droplet laser. (a) Lasing spectrum from a 24 μ m-diameter droplet. The inset is close-up of lasing peaks, indicated spectrum linewidth of about 0.13 nm. Lasing wavelengths for TE and TM polarization are calculated and drawn below the spectrum, shows an agreement between experimental observation and theory.

Figure 4.5 (a) Lasing threshold characteristics for TE and TM polarization. (b) TE modes develop before appearance of TM modes.

4.3.2 Size dependent lasing characteristics

One advantage of our technique is the possibility to create droplets with a wide-range of sizes, beneficial for investigation of size dependent lasing characteristics. Figure 4.6 plots spectra of four droplets with decreasing diameters from about 31 to 16 μm . As the cavity size decreases, we observed three phenomena that are similar with previously reported [64] (i) a blue-shift behaviour of lasing envelop from 650 nm to 600 nm; (ii) number of lasing modes decreases and (iii) free spectral range (FSR) increases.

The first phenomenon was observed and well explained in Ref [64], which ascribed to the lasing power (P) coupled out the droplet.

$$P \propto \frac{V(D, \lambda)}{\lambda^3} g(\lambda) \beta(D, \lambda)$$

where, V is mode volume, g is the gain spectrum, D is the diameter, β is the out-coupling efficiency. $\beta = Q_{rad}^{-1} / Q_{total}^{-1} = Q_{rad}^{-1} / (Q_{rad}^{-1} + Q_{abs}^{-1})$

It is note that a blue-shift effect has been observed previously due to variation of dye concentration [87] but in our work, the dye concentration is unchanged so we exclude this reason.

The second effect is due to less overlap between cavity mode and gain profile. Finally, the increasing of FSR is well-explained with notice that $\text{FSR} = \lambda^2 / (\pi D n_e)$ where λ and n_e are resonant wavelength and effective refractive index of droplet. According to this equation, FSR increases with decreasing D . Furthermore, FSR should follow an α/D function (α is a constant) because the value $\lambda^2 / (\pi n_e)$ is almost unchanged. We experimentally measured FSR of series of droplets based on their spectra (Figure 4.6) and plot FSR as function of their diameters (Figure 4.7). The FSR curve is well fitted with the α/D function, verifies an agreement between experiment and theory.

Figure 4.6 Size dependent characteristics of droplet lasing.

Figure 4.7 Free spectral range (FSR) as function of droplet size.

Narrow linewidth and single mode operation are demanded for integrated photonic circuits, and therefore, are desired parameters of microlasers. It can be achieved based on various configurations including distributed feedback (DFB) [86], or Fabry-Perot (F-P) resonators [88]. In a DFB structure, the grating is typically designed for single wavelength reflection and only this wavelength is full filled the resonance condition so single mode lasing emission is normally generated. In contrast, for F-P cavity, a wide range of wavelengths can be fulfilled the resonance condition, leading to multiple lasing modes. To obtain single mode emission, FSR needs to be sufficiently large which is comparable with a lasing region [75]. It means that the cavity length is required to be short enough. Similar to F-P resonators, in WGC cavities, numerous of resonance modes can be generated and single mode lasing can be only obtained when the cavity diameter is in range of 10 μm . The Figure 4.6 shows number of lasing modes decrease with reducing cavity size because FSR increases.

Figure 4.8 Single mode lasing operation is observed from a 13 μm droplet (PL image is shown in the up-right inset). The up-left inset indicates close-up of lasing mode with Lorentz fitting.

When the diameter down to about 13 μm , we observed single mode emission with spectral linewidth of 0.15 nm (Figure 4.8). Compared with DFB and F-P resonators [86, 88], our structure is much easier in fabrication but the lasing linewidth is still in the same order of magnitude.

4.4 Tunable lasing emission

4.4.1 Wavelength tunability via gain doping

Doping flexibility is one main advantages of soft matter cavity compared **to** traditional semiconductor resonators. Here, we demonstrate that the lasing wavelengths can be easily tuned by changing the doping lasing dyes to Rhodamine 6G (R6G), and Rhodamine 19 Perchlorate (R19P).

Figure 4.9. Size dependent characteristics of droplet lasing for different gain doping (a) R19P and (b) R6G.

Figure 4.9 shows series lasing spectra from droplets ($D = 13\text{-}30\text{ }\mu\text{m}$) doped R19P and R6G. Lasing wavelengths are in a range of 555 nm to 600 nm and 557 to 617 nm for R19P and R6G gain, respectively. Single mode lasing emission was also achieved from droplets with diameter around 13 μm .

4.4.2 Wavelength tunability via mechanical cavity deformation

Tunable lasing wavelength without changing gain material is more interesting. It can be realized by changing the shape or refractive index of a cavity [24, 78, 83, 89]. In our work, both cavity and carrier have mechanical flexibility, offering us a possibility to manipulate lasing mode by mechanical deformation. The carrier PDMS can be cured by adding curing agent with ratio over the base material is 1:10. The sample was dried under ambient conditions and at room temperature for about two days. The PDMS became solid and it could be taken out from the glass holder and fixed in a state that is on two movable substrates (Figure 4.10a). By tuning this substrate, The PDMS film can be stretched thus altering the shape of the droplets embedded from sphere to ellipsoid (Figures 4.10b-c). As a result, the cavity length is modified which leads to variation of lasing modes.

Figure 4.11a plots spectra of a 90 μm droplet under increasing deformation ratio from 0 to about 6%. Lasing mode exhibits a red-shift behaviour, which is ascribed to a light increase in droplet cavity [83]. Moreover, the shifting value ($\Delta\lambda$) has a liner relationship with the deformation and is higher for small droplets as shown in Figure 4.11b. Under the same deformation, about 3%, the shifting is around 0.5 and 0.15 nm for droplets with diameters of 64 and 90 μm , respectively. It is expected that the change of lasing wavelength with cavity deformation can be employed for force sensor.

Figure 4.10 Droplet cavity changes under mechanical deformation. (a) Schematic diagram for stretching droplet. (b) and (c) Optical image of a droplet without and with deformation, respectively.

Figure 4.11 Tunable lasing mode via cavity deformation. (a) Lasing mode is red-shift with increasing deformation ratio ($\Delta D\%$). (b) Mode shifting ($\Delta\lambda$) with different deformation ratios and cavity sizes.

4.5 Summary

In this chapter, we contribute a facile and novel method to fabricate droplet cavities with series of size. The droplet material is a polymer composition that demonstrates better stability compared with water or organic liquids (Figure 4.12). The droplets are self-assembled so they have smooth outer surface, favourable for high cavity Q factor. Lasing emission with (i) low threshold (about 200 nJ/pulse) and (ii) clear separation mode and well-defined polarization have been achieved. Furthermore, we demonstrate the lasing wavelength tunability via gain doping and mechanical cavity deformation, which is significant for the development of tunable and flexible lasers.

Figure 4.12 Development diagram of droplet lasers.

Owing to simple fabrication, high Q factor and mechanical flexibility, the droplets may be used for flexible photonic devices. Interestingly, the cavity shape can be modified by controlling the ratio of epoxy and PS so it is expected that different configurations such as cylindrical cavity, ellipsoid and spindle-cavity can be fabricated. This achievement might contribute to study optical chaos and to develop directional emission microlasers. However, even though the polymer droplets exhibit many above advantages, there are also some drawbacks. For example, because the droplets are fully covered by the PDMS, there is no

free-space for approaching a waveguide to the droplets for efficient excitation and emission collection. To over these issues, we have successfully developed a new hybrid structure (combination of organic and inorganic materials), the so-called hemispherical microcavities which will be introduced and discussed in the following chapter.

Chapter 5

Lasing from hybrid flexible hemispherical microcavities

5.1 Introduction²

In the previous chapter, we have introduced the soft matter droplet cavities inside PDMS matrix. This structure has several advantages such as simple fabrication, excellent mechanical and doping flexibility. However, there are still some limitations: (i) Contrast refractive index between cavity ($n_1 = 1.53$) and surrounding environment (PDMS, $n_2 = 1.41$) is not high ($n_1 - n_2 = 0.12$) which reduces the Q factor of droplets; (ii) Due to the elastomer, coupling out the lasing emission by integrating or manipulating a waveguide closed to droplet surface is nearly impossible. We solve these drawbacks in this chapter.

Herein, we combine droplets with a planar distributed Bragg reflector (DBR) to make hybrid hemispherical cavities [90]. These structures take both the stability of the dielectric planar construction and the easy of soft materials. We propose a novel technique to create hemispherical cavities on a DBR substrate with precisely control the size and position. By doping laser dyes into these hemispheres, for the first time, optically pumped lasing has been observed. We address lasing mechanism, size dependent characteristics, and optical confinement effect of the hemispheres.

²The work in this chapter has appeared in "Self-Assembled Flexible Microlasers," *Adv. Mater.* **24**, OP60-OP64 (2012) ([Highlighted by Materialviews](#)) and "Single Mode Lasing from Hybrid Hemispherical Microresonators," *Sci. Rep.* **2**, 244 (2012).

5.2 Fabrication processes

5.2.1 Distributed Bragg Reflector

Distributed Bragg reflector (DBR) is a high reflectivity dielectric mirror, which is very important component in vertical cavity surface emitting lasers (VCSEL) [91-93]. It consists of numbers of repeat pairs of two dielectric quarter-wavelength layers. The thickness of the two layers is very critical so phase difference for an electromagnetic wave reflected at the layer's interface is multiple of 2π . Reflectivity of a DBR depends on number of the pair layers and their refractive indices, and can be obtained based on transfer matrix method (TMM) [59] and simulated by a commercial software like Matlab [94] or Mathcad [95]. A stop-band where the reflectivity is the highest and typically flat versus wavelengths is an important factor of any DBRs.

Figure 5.1 DBR structure fabricated by electron beam evaporation.

In our work, the DBR is a commercial structure that consists of 27 pairs of SiO₂ and TiO₂ with central wavelength at 630 nm, and fabricated by electron beam evaporation (Figure 5.1). Measured reflectivity of the DBR is plotted in Figure 5.2. The reflectivity is maximum ($> 98\%$) at a central flat band covers from 600 to 740 nm and drops off in either sides.

Figure 5.2 Measured reflectivity of the DBR structure.

5.2.2 Hemispherical microcavities fabricated manually

The solution used to form the hemispheres was made by dissolving the commercial available Araldite 506 epoxy resin (so-called epoxy for short) into chloroform and then adding Rhodamine 6G (R6G) dye. The process is shown in Figure 5.3. Volume ratio between chloroform and the epoxy is about 1:8. The function of chloroform is for well dispersion of the R6G. The solution still works without chloroform. However, it takes more time to get a uniform mixture. If the chloroform is too much, it can destroy the original viscosity of the epoxy, thus significantly dropping solution's surface tension and seriously affects the hemispheres' quality.

Figure 5.3 Fabrication recipes of R6G doped polymer solution.

Figure 5.4 RhB molecular structure based on product information of Sigma-Aldrich.

R6G is a common and high quantum yield lasing dye material [50] whose molecular structure is shown in Figure 5.4. This dye is well dissolve in water and most common organic solvents like chloroform, DCM, acetone and so on. R6G has a broad emission spectrum covers from about 520 up to near 720 nm that matches well with the flat stop-band of our DBR.

Figures 5.5a-e show a manual method to form the hemispheres. Firstly, a hydrophobic film was deposited on top surface of the DBR by spin coating or by spreading a droplet of a solution that contains one volume of 1H,1H,2H,2H-perfluorooctyl-Triethoxy-silane [90] and four volume of chloroform. This special layer enhances the hydrophobic effect of the DBR surface which plays important role for obtain perfect hemispheres. The same hydrophobic

layer was coated outside a microrod (can be common optical fibers, human hairs, metallic needles). Secondly, the dye solution was spread along the rod to create a thin layer (Figure 5.5c). This layer gives the birth of droplets, which are formed spontaneously by surface tension (Figure 5.5d). These droplets hang on the rod and they can be easily transferred to the DBR surface by a soft contact. Interestingly, not all but about half of droplets left the rod creating the hemispheres. Therefore, after the first deposition, droplets (smaller size) are still hanging on the rod and they can be reused to make smaller hemispheres (Figure 5.5e). By doing so, hemispheres with reduced sizes can be achieved. Figures 5.5f-j present typical hemispheres with diameters from about 100 to 20 μm . The hemispheres show smooth surface because they were self-assembly induced by surface tension. We can see nearly perfect circular ring shapes from edge of the hemispheres, thus enabling to achieve WGMs. High quality hemispheres with different sizes can be fabricated by the above approach. Size and position of a hemisphere can be controlled but the accuracy is limited. To improve the controllability, we use a microplotter system to make hemispheres that will be introduced in the next section.

Figure 5.5 (a)-(e) Hemispherical microcavities fabrication based on droplets. (f)-(j) Top-down optical images of various hemispheres with decreasing sizes

5.2.3 Hemispherical microcavities fabricated by a microplotter

Figure 5.6a shows the image of GIXTM MicroplotterTM II from Sonoplot, INC. This system is widely used for polymer electronics, microarrays. One important component in this apparatus is a dispenser connected with a glass tip (Figure 5.6c). Thanks to this small tip, the microplotter can deposit or dispense extremely small liquid volumes (picoliters) onto many kinds of planar surface. The tip can be moved freely in 3D space and its position is controlled by the control panel (Figure 5.6b) with help from a small available CCD camera. The microplotter can create many configurations such as hemisphere arrays, polymer line/arc waveguide as small as 5 μm . Here, we limit the application of the microplotter on creation of hemispheres only.

Figure 5.6 (a) Microplotter system and its magnitude of (b) control panel and (c) hollow glass needle.

To form a hemisphere, the solution first needs to be loaded into the hollow glass needle via its head. The fluid is dispensed by the ultrasonic pumping (automatically or manually activated during the fabrication process). Then, the needle is approached close to contact with the surface as shown in Figure 5.7. A droplet will be deposited on the surface and the ultrasonic is deactivated. After that, the tips is retracted from the surface and move to another position and the deposition process is repeated.

Figure 5.7 Fluid deposition on a planar substrate via hollow glass needle and ultrasonic pumping.

Figures 5.8a-e show step-by-step how hemisphere arrays can be fabricated by the microplotter. By using the glass tip with different sizes, corresponding hemispheres with a particular diameter can be obtained. Figures 5.8f-g present hemisphere arrays formed by glass needle with diameter of needle's head 30 and about 90 μm , respectively. For the 30 μm needle, diameter of created hemispheres is about 35 μm while for 90 μm needle, it is about 85 μm . From the images, hemispheres are almost the same or the uniformity is excellent. To examine further, we measured diameters of 173 hemispheres created by the 30 μm and the

size distribution is plotted in Figure 5.9. About 42% of the hemispheres have diameters either 35 or 36 μm and more than 95% of the hemispheres have diameters $35 \pm 3 \mu\text{m}$. The results indicate that the glass needle can precisely control cavity size.

Figure 5.8 (a)-(e) Array of hemispherical microcavities fabricated by microplotter. (f) and (g) Hemisphere arrays using a 30 and 90 μm glass tip, respectively.

Figure 5.9 Size distributions of hemispheres formed by the 30 μm glass needle.

Figure 5.10 NTU pattern based hemispheres using the 30 μm glass tip.

The glass needle is free to move in 3D space with movement distance as small as 1 μm . Therefore, it is expected that position of created feature can be controlled with accuracy in range of 2-3 μm . An example of the position controllability is shown in Figure 5.10 where a “NTU” pattern is formed by line of hemispheres. The ability to manage the hemisphere’s position with high precision is very important for fabrication of coupling cavities.

5.3 Optical characteristics of hemispherical microcavities

5.3.1 Spontaneous emission under continuous wave laser excitation

Individual R6G doped hemispherical structures were investigated by means of the $\mu\text{-PL}$ system (shown in the Chapter 3) under continuous wave (CW) and pulsed laser excitation. Figure 5.11a plots PL emission from a hemisphere with diameter (D) of 100 μm (whose image is shown in the inset of Figure 5.12c) under Cd-He CW laser line of 325 nm. It is clear that the PL intensity increase with rising in the power of the CW laser. The spectrum has broad linewidth which is therefore ascribed to spontaneous emission. The PL intensity (I_{PL}) should be expressed as a function of pumping power (I_{EX}), $I_{\text{PL}} = \eta(I_{\text{EX}})^\gamma$, where η is the

emission efficiency and γ denotes the radiative recombination mechanism [96]. To gain the above dependence, the integrated PL intensity versus CW excitation power is plotted in Figure 5.11b. The results indicate a linear relationship with exponent factor $\gamma = 1.1$, verifying the observation of the spontaneous emission from the R6G doped hemisphere.

Figure 5.11 (a) PL intensity from a 100 μm hemisphere and (b) the integrated PL versus increasing CW pump power.

5.3.2 WGM lasing emission under pulse laser pump

Lasing emission was observed from hemispheres under pulsed laser excitation from the YAG pulse laser. Figure 5.12a shows spectra from a 80 μm -diameter hemisphere (optical and PL image are present in the inset of Figure 5.12b) as function of pump pulse energy (PPE). At low PPE ($< 2.8 \mu\text{J}$), there is only spontaneous emission. However, at PPE = $2.8 \mu\text{J}$, sharp

Figure 5.12 (a) Emission spectra from individual hemisphere versus excitation energy. The inset presents the PL emission just above lasing emission. (b) The integrated PL intensity versus PPE. (c) Lasing mode calculation for (d) corresponding lasing spectrum under PPE = 3 μJ .

peaks appear. Close-up spectrum is plotted in the inset. The peak intensity sharply increases with increasing PPE. The integrated PL versus PPE (Figure 5.12b) displays a nonlinear dependence that is the indicator of lasing action. The matching point gives information of lasing threshold, which is determined to be about 2.5 μJ . The lasing modes can be well matched using simple equation based WGM theory: $\lambda_m = \pi n_e D / m$, where m is mode numbers and n_e is effective refractive index [90]. Considering $D = 80.14 \mu\text{m}$ and $n_e = 1.41$, the calculated m is found to be 623-625 as shown in Figure 5.12c. In addition, under increasing PPE the lasing modes exhibit blue-shift (Figure 5.12d) which probably caused by thermal effect [97].

5.3.3 Cavity size dependent properties and single mode operation

Lasing characteristics from hemispheres with different sizes were carried out. Figure 5.13 plots spectra emission from there hemispheres with increasing size. We observed similar effects compared to our previous droplet lasing. The FSR decreases with cavity size. For WGCs, FSR can be estimated by $\text{FSR} = \lambda^2 / (\pi D n_e)$, where the resonant wavelength λ can be almost considered as a constant. As a result, FSR is a linear dependence of $1/D$. To verify this hypothesis, we measured FSR of a number of hemispheres based on their spectra and plot the data as a function of $1/D$ (Figure 5.14a). The experimental data is well fitted with a linear function that supports the WGM lasing mechanism. In addition, the lasing Q factor, determined as $Q = \lambda / \delta\lambda$, where $\delta\lambda$ denotes the spectral linewidth of the lasing mode, increases with D (Figure 5.14b). The Q factor are about 6×10^3 to 8×10^3 for $D = 30$ and $100 \mu\text{m}$, respectively. This value is higher than those of droplet lasers [64] and semiconductor micropillar lasers [11].

Figure 5.13 Lasing spectra from hemispheres with different diameters. The inset shows optical and PL images of corresponding hemispheres.

Figure 5.14 (a) FSR as function of $1/D$ and (b) Q factor versus D .

In order to better confirm the lasing mechanism, we have fitted the lasing spectra for several hemispheres based on WGMs. Figure 5.15 plots energy of lasing mode and calculation as function of mode number m . The data displays the consistence between theory (small back symbols) and experiments (big symbols) for cavity sizes ranging from around 30 to 100 μm . We, therefore, conclude that the lasing action in the hemispheres is ascribed to WGMs.

Figure 5.15 Fitting lasing modes for different hemispheres. The big symbols refer to experimental mode energy and the small dots symbols are calculated energy based WGM theory.

In the previous chapter, the single mode emission, from droplets with diameter close to 13-14 μm , was obtained. Here, the single mode, which is highly demanded for information processing and on-chip application, can be also observed from a hemisphere with diameter

around 15 μm as shown in Figure 5.16. The single mode operation is possible because the FSR is comparable with the lasing region as depicted in the inset of Figure 5.16.

Figure 5.16 Single mode emission from a 15 μm hemisphere. The inset depicts the lasing region.

5.4 Three dimensional confinement of hybrid hemispherical resonators

The DBR substrate herein is not only for cavity stabilization but also, more importantly, for reducing lasing threshold. With the underneath DBR, light from hemisphere is confined in 3D [90]. In horizontal plane, light is trapped by total internal refraction. In vertical plane, light is also trapped inside the hemisphere because it cannot leak through the DBR substrate. This idea was confirmed by replacing the DBR with a normal glass substrate and an aluminium foil. Under the same condition, lasing was not observed from the hemisphere on the glass substrate (Figure 5.17a). From the PL image (Figure 5.17c), the optical loss was too high to generate the cavity loop. However, lasing emission was achieved from the hemisphere on the

aluminium foil (Figure 5.17d). The PL image exhibits the bright circular loop that ascribed to WGMs in the edge of the hemisphere (Figure 5.17f).

Figure 5.17 (a) and (d) PL from a hemisphere that deposited on top of a glass substrate and an Al foil, respectively. (b) and (c) Optical and PL images of the hemisphere on glass. (e) and (f) Optical and PL image of the hemisphere on Al foil.

5.5 Numerical simulation of electromagnetic field of a hemisphere

Cavity loop is formed inside and near the edge of a hemisphere and denoted as a white ring in Figure 5.18a. Electromagnetic field in the horizontal plane and radial direction was simulated by FEM method supported by COMSOL multiphysics. The simulated results are shown in Figures 5.18b-c. The light is confined to the hemisphere by multiple total internal reflections at the hemisphere-air interface. Figure 5.18c presents mode profile in the vertical cross-section following method in Ref. [63].

Figure 5.18 (a) Light is in 3D (xyz) confined inside a hemisphere to form WGMs that is denoted as the white ring. (b) Field distribution in a hemisphere with $D \sim 7 \mu\text{m}$ in (b) horizontal and (c) vertical plane.

5.6 Multicolor lasing from a single hemisphere

Recently, by embedding green and red emission lasing dye (Coumarin 540A and Rhodamine B) into a single hemisphere, dual color lasing was achieved under pulsed laser excitation (wavelength 420 nm, pulse duration 5 ns, repetition rate 20 Hz). Figure 5.19 plots spectrum from a hemisphere with $D \sim 200 \mu\text{m}$. Obviously, there are two lasing envelopes, one is green wavelengths and the other is yellow-red wavelengths. This achievement opens a possibility to generate white light lasing, which can be found applications for display technology [98] and solid-state lighting [99].

Figure 5.19 Lasing spectrum from a C540 and RhB doped hemisphere with green and yellow-red emission.

5.7 Summary

We have demonstrated versatile technique for fabricating high Q factor microcavities. This approach has advantages of both the flexibility of soft matter and the stability of traditional dielectric material. Furthermore, this combination leads to the 3D optical confinement cavities which have been demonstrated as outstanding candidates for lasing applications. By embedding lasing dye into the hemispheres, lasing with excellent characteristics such as low threshold, clear mode spacing and narrow spectral linewidth have been observed. Interestingly, different gain materials can be easily to incorporate into the structures for color tunability. Further development on coupled or arrays of cavities may open potential application for optical filters and bistability. Our structure also provides a good foundation for studying nonlinear optical effects.

Chapter 6

Self-assembled hemispherical microlasers and their sensing application

6.1 Introduction³

In the previous chapter, flexible hemispherical microlasers with excellent properties such as low threshold, clear mode spacing and high Q factor have been demonstrated. In this Chapter, we present hemispheres with better lasing performances including lower lasing threshold, higher photostability and potential applications as bio/chemical **sensors**. Compared with earlier structures, the dye-epoxy solution was modified by adding poly(methacrylicacidmethylester)/PMMA. It is well-known that PMMA, liner formula $[\text{CH}_2\text{C}(\text{CH}_3)(\text{CO}_2\text{CH}_3)]_n$, is a chemically stable material with high refractive index ($n_{\text{PMMA}} = 1.49$) and transparency at visible wavelengths. Figure 6.1 shows chemical structure of a single PMMA chain. The polymer chain number (n) affects viscosity of PMMA solution. In our work, molecular weight (m_w) of PMMA is about 350,000 corresponding to around 3,500 chains.

Owing to high refractive index, the presence of PMMA in the solution significantly increases index of hemispheres. Thus, light is better confined and cavity Q factor is improved. The introduction of PMMA in the new recipe is shown in Figure 6.2. Firstly, 10 wt% PMMA is dissolved in DCM and following epoxy is added. Weight ratio between epoxy and PMMA

³ Data in this chapter has published in "Application of self-assembled hemispherical microlasers as gas sensors," [Appl. Phys. Lett.](#) **102**, 031107-031104 (2013).

(WR_{ep}) is in a range of (4-8): 1. Finally, 0.2-0.25 wt% RhB is incorporating into the solvated PMMA-epoxy solution.

Figure 6.1 Monomer structure of PMMA. n is repeated number of polymer chain.

Figure 6.2 Fabrication recipes of dye doped polymer solution

6.2 Self-assembled hemispherical resonators

PMMA has been widely used for forming nano/microfibers waveguide [100]. The fibers can be realized by directly drawing from a solution [101]. Generally, polymer fiber can process a uniform cylindrical shape. Taking this advantage, we propose a new idea based on the polymer fiber for creating hemispheres with high uniformity in size. It is possible to transform a fiber to well-aligned hemispheres because the main component of the solution is still the epoxy resin (Chapter 5) which has large surface tension. Figure 6.3 shows schematic diagram of fiber fabrication. Firstly, solution is injected to deposit a droplet on top of a glass substrate (Figure 6.3a). Then, a metal needle is approached down to immerse into the droplet. Secondly, the needle is vertically retracted and a fiber is formed (Figure 6.3b). Finally,

fabricated fiber is carefully placed on a DBR substrate coated the thin hydrophobic layer (Figure 6.3c). It is note that the fiber size is controllable by drawing speed.

Figure 6.3 Schematic diagram of microfibers fabricated via directly drawing from solution.

Figure 6.4 Self-assembled process from a polymer fiber to hemispherical resonators as a function of time.

Figure 6.4 shows top-down optical images of the self-assembled process from a fiber ($WR_{ep} = 6:1$) to hemispheres on top of the DBR. Thanks to the hydrophobic layer and large surface tension of the solution, the fiber is shrunk (Figure 6.4a) and then broken down into many pieces (Figure 6.4b), which later self-assembly formed hemispheres (Figures 6.4c,d). The total time (T_{total}), counted when fiber was placed on the DBR until hemisphere formation, takes about 20 min. This time depends on WR_{ep} and size of fibers that is summarized in Table 6.1.

For $WR_{ep} < 4:1$, fiber can fabricate but the self-assembled process cannot be obtained because the fiber is strongly stuck on the DBR's surface. In contrast, when $WR_{ep} > 8:1$, concentration of PMMA is low, fiber cannot be formed. The self-assembled process can complete only for $4:1 < WR_{ep} < 8:1$. When WR_{ep} increases, T_{total} decreases. For instance, fiber with diameter about 20 μm , T_{total} is approximately 20 min for $WR_{ep} = 6:1$ and less than 5 min for $WR_{ep} = 8:1$. On other hand, the time for hemisphere formation is longer for a bigger fiber. For example, with fixed $WR_{ep} = 8:1$, T_{total} are around 5, 10, 20 min for fibers with diameters about 20, 60, 100 μm , respectively.

WR_{ep}	Total time of the self-assembled process		
	Fiber diameter 15-25 μm	Fiber diameter 50-70 μm	Fiber diameter 90-110 μm
4:1		~10 h	
6:1	10-20 min	50-70 min	100-115 min
8:1	<5 min	10-15 min	~20 min

Table 6.1 Total time of the self-assembled process as function of WR_{ep} and fiber size.

The hemisphere diameter is controllable by fiber size. Hemispheres with diameters from about 5-150 μm can be easily formed. Figure 6.5 typically shows hemispheres with diameters about 10, 50, and 100 μm .

Figure 6.5 Microresonators with various sizes from 100 to 10 μm can be easily obtained.

6.3 Lasing characteristics of hemispherical lasers

To prepare hemispheres for optical characterizations, we baked them at 60 $^{\circ}\text{C}$ for 90 min and additional dried at room temperature for 2-3 days. These processes help to solidify and stabilize the hemispheres. After that, the hemispheres are ready for optical excitation. Figures 6.6b-d show top-down PL images of a hemisphere (optical image, Figure 6.6a) upon increasing pumping energy. Under low excitation density, the hemisphere exhibits weak PL emission (Figure 6.6b) whose intensity largely increases upon higher excitation energy

(Figure 6.6c-d). When the pumping is above lasing threshold, emission intensity at the edge of the hemisphere is much higher than that of middle part of the hemisphere, which leads to a very bright ring shape. This observation predicts the origin of WGMs.

Figure 6.6 Images of a hemisphere (a) under halogen illumination and (b)-(d) under laser excitation with increasing pumping energy.

Polarization is an important feature of laser light. It can be transverse magnetic (TM) or transverse electric (TE) which identifies by orientation of electric field. Figure 6.7 indicates schematic view of TM (TE) modes when electric field is oriented in radial/horizontal (vertical) directions [24]. Based on this definition, the polarization can be identified by examining emission intensity after a polarizer. If at the initial position, polarizer's axis is in horizontal direction then the intensity should be maximum for TM modes and minimum for

TE modes. The result is reversed when the polarizer is rotated 90 degree (Figure 6.7). Following this idea, we constructed an optical setup depicted in Figure 6.8 to indentify polarization of laser light from a hemisphere. The emission is collected from the edge of the hemisphere. Polarizer's axis is oriented as in Figure 6.7.

Figure 6.7 schematic views of WGMs (yellow-red ellipsoids) with transverse magnetic (TM) or transverse electric (TE) polarization (indentified by electric field orientation-white arrows). TM and TE modes can be identified by monitoring light intensity after a polarizer's under certain rotation angles.

Figure 6.8 Emission is collected from the edge of hemisphere. Polarization of the laser light is studied using a polarizer.

Figure 6.9 plots spectra emission of the hemisphere versus rotation angle of the polarizer. The intensity of lasing modes reaches maximum at 0 and 180 degree, and decreases to the minimum at 90 degree. Integrated of PL intensity versus the rotation angle is shown in the inset, indicating a good agreement between experimental data and Malus' law. We conclude that the laser light is ascribed to only TM polarization. However, even though TM modes are dominated, TE modes can be also detected (much weaker intensity) under extremely high pumping density. Low Q factor for TE modes might due to diffraction scattering [102] and it is may the reason for their absence in our structure.

Figure 6.9 Lasing intensity with different rotation angles of the polarizer. The inset show integrated lasing mode versus the rotation angle. The fitting curve is based on Malus' law.

Low threshold lasing is a significant factor of any microlasers [103]. Previously, lasing threshold of R6G doped hemisphere was about 2 $\mu\text{J}/\text{pulse}$ corresponding to around 200 kW cm^{-2} , which can be easily achieved using common diode lasers. However, in order to be able to pump by a bright light emitting diode (LED) with pulse current, the lasing threshold need to be much lower. In this chapter, the lasing threshold can reach down to about 45 kW cm^{-2} , which is one order better than our previous work. This value can be improved by increase cavity's refractive index and doping suitable dye concentration. It is expected that the hemisphere lasers can be realized under LED pumping which minimizes the system and opens potential applications.

Figure 6.10a shows PL spectra of a hemisphere upon increasing pump pulse energy. The evolution from spontaneous emission with broad linewidth and weak intensity under lowest pumping density (216 nJ) to stimulated emission with sharp lasing peaks when pumping energy ≥ 396 nJ can be clearly seen. Furthermore, as shown in the inset, position of lasing modes remains the same at increasing pumping excitations. There is no evidence of the blue-shift that observed formerly (Chapter 5). This is a significant improvement in lasing photostability, thanks to the presence of the PMMA. In fact, PMMA helps to cure the hemispheres leading to solid structures and thus prevents the diffusion of oxygen from the air to the hemispheres, and therefore, avoiding oxidation of dye molecules.

Figure 6.10b plots the threshold behaviour of the hemisphere, which is about 380 nJ, equivalent to a fluence per pulse of 34 $\mu\text{J cm}^{-2}$. We have investigated numerous hemispheres and lasing thresholds are in a range of 30 to 45 $\mu\text{J cm}^{-2}$ corresponding to 30-45 kW cm^{-2} . These values are seven orders smaller than previous reports on droplet lasing [76, 77].

Figure 6.10 (a) Emission from a hemisphere with diameter about $104\ \mu\text{m}$ versus pump pulse energy. The inset shows a close-up image of lasing modes. (b) Lasing threshold behaviour and (c) Close up lasing mode at pulse energy 540 nJ with calculation mode position for first order with TM polarization.

Lasing wavelengths can be well fitted using WGM theory for the first order $q = 1$ and TM polarization (Figure 6.10c). Given diameter of the hemisphere $D = 104.01 \mu\text{m}$, refractive index of the hemisphere and the outside medium (the air) are 1.46 and 1, respectively, resonant modes are well matched with mode numbers $m = 759\text{-}762$. As a result, we conclude again that lasing mechanism is ascribed to WGMs.

Figure 6.11 Lasing characteristics versus cavity's size. Single mode emission is achieved when diameter of hemisphere closed to $10 \mu\text{m}$.

For WGCs, the FSR can be estimated as $\text{FSR} = \lambda^2/(\pi n D)$. That means when D decreases, FSR should increase. This effect is clearly observed as shown in Figure 6.11. The expanding of FSR also cuts off certain lasing modes due to less overlap of resonant modes with gain region. It is found that when D closed to $10 \mu\text{m}$, FSR is comparable to the lasing region and single mode emission is achieved [75].

6.4 Vapor refractive index sensor

The idea of using WGCs for sensing species bonded on the cavity's surface has been proposed in middle of 1990s [104] and well developed in earlier 2000s [105, 106]. Up to date, the WGM technology is considered as one of the most powerful technique for ultrasensitive biological and chemical sensing. This method is label-free and sensitivity can down to level of single particle and molecule [18, 107-110]. Mechanism of this method is based on the modification of cavity mode due to interaction between sensing molecules or particles with the cavity's surface. This reaction leads to shifting of resonance mode and is an indicator of a sensor. Following this approach, we employ our microlasers for refractive index vapor sensing.

Figure 6.12 shows schematic setup for sensing application of the hemisphere lasers. We placed and then fixed the DBR with a hemisphere $D \approx 53 \mu\text{m}$ on the bottom of a glass beaker (Figure 6.12a). Then, a sheet of parafilm was used to seal the top open part of the beaker (Figure 6.12b). To allow sensing vapor loading, excitation and signal collection, a small circular window was opened just above the hemisphere.

We excited the hemisphere before the vapor was loaded and recorded the signal. It is noted that in this experiment, the excitation source was the same but with a lower repetition rate, 20 Hz, for better emission stability. Following, the acetone vapor was pumped through the window for about 15 s. Subsequently, pumping laser was opened to excite the hemisphere in 4 s. The emission was continuously recorded with integrated time of 100 ms. The results show a red-shift of lasing mode after vapor gas is loaded (Figure 6.12d). Furthermore, after the vapor fully diffuses from the beaker to the air through the hole, lasing mode back to its original position. This observation verifies the red-shift is caused by the existence of acetone vapor.

Figure 6.12 (a)-(c) Schematic setup for investigation of vapor refractive index sensing. (d) Shifting of lasing mode is an indication of refractive index variation. Before, after and final mean before the acetone vapor is loaded, after the vapor is loaded and final of vapor diffusion. The final stage refers to the original condition (before vapor is pumped).

The sensing mechanism of our sensor is based on two factors: (i) The presence of acetone vapor leads to a thin layer of acetone molecules, which covers the hemisphere, and causes the wavelength to shift. This description will be discussed in detail in Chapter 8. In addition, the vapor may also cause the polymer to expand which leads to wavelength shift.

Figure 6.13a plots lasing spectra of the hemisphere with diffusion time of acetone vapor (T_d). During the 4 s excitation, acetone vapor continuously diffused from the beaker to the air. As a result, the concentration of acetone vapor inside the beaker decreased as function of time, thus reducing refractive index of the medium. This is the reason why lasing mode exhibits a blue-shift with increasing T_d . The shift of lasing mode is better recognized in Figure 6.13b where close up image of a lasing mode near 608 nm is shown.

Figure 6.13 (a) Lasing mode shifts with decreasing of acetone vapor density, proportional to the time for acetone diffuse from the beaker to the air (T_d). (b) Close-up image of lasing mode near 608 nm.

Figure 6.14 Shifting of lasing mode decreases with T_d

Shifted value ($\Delta\lambda$) of lasing mode compared with its original position (before acetone vapor is added) as function of T_d is plotted in Figure 6.14. The shift significantly drops in the first 600 ms, following with a gradual decrease. The maximum shift is about 0.11 nm

corresponding to refractive index change (Δn) of around 8×10^{-4} refractive index unit (RIU). Therefore, the achieved sensitivity is $S = \Delta\lambda/\Delta n \approx 135$ nm/RIU, which is one order better than earlier work based on a ring cavity [108].

6.5 Summary

In this Chapter, we demonstrate a new approach for fabrication of hemispherical microresonators with tunable size from about 5 to 150 μm . By incorporating lasing dye RhB into these structures, lasing modes with very narrow spectral linewidth (0.06 nm) are achieved. Laser Q factor is up to 10^4 and lasing threshold is about 30-45 kW cm^{-2} . Compared with our earlier work on hemispherical lasers, the lasing performances are significantly improved such as better photostability, higher Q factor and lower threshold. Furthermore, we have shown that this kind of lasers can be employed as refractive index vapor sensors with sensitivity about 135 nm/RIU. Our findings demonstrate the potential of the hemispherical cavities as sensitive chemical/biological sensors and active components for phonic devices.

Chapter 7

Microfiber lasers and its application as liquid refractive index sensors

7.1 Introduction⁴

Recently, one-dimensional (1D) semiconductor structures have attracted intensive research attentions due to their small size and many applications such as laser light sources, waveguides, photo detectors and LED [111-113]. They are considered as building blocks for on-chip technologies. However, typically fabrication of these structures is very challenging because they rely on high cost technologies combined with well and precise control nano/micro fabrication equipments.

Compare with traditional semiconductor 1D structures, polymer nano/microfibers are novel, cost-effective mass production, better mechanical flexibility, and ease assembling to ordered structures or arrays that is highly potential for functional nano/microscale optical devices [114]. Polymer fibers can be easily to fabricate by electrospinning deposition [114-119], vapor deposition [120], or directly drawing from liquid phase [101, 121-125]. The recent breakthrough of creating logic elements and circuits from polymer fibers arrays predicts their bright future in photonic technologies [126].

Up to date, applications of polymer fibers are mainly limited as waveguides. Nano/microlasers, which are significant importance for information processing in photonic

⁴Contents in this chapter has appeared in "Whispering gallery mode microlasers and refractive index sensing based on single polymer fiber," *Laser Photon. Rev.* **7**, 133-139 (2013). ([Frontispiece Image](#)).

devices and systems [127], however, are rarely investigated and most of them are based on waveguide effect [114, 119, 128]. The principle of these lasers is schematically depicted in Figure 7.1 where the two endfaces served as two mirrors to form a common F-P resonator. Generally, this approach suffers from low cavity Q factor due to natural poor reflectivity of the fiber endface reflectors. To overcome this drawback, other approaches need to study.

Figure 7.1 Nano/microfiber lasers based on F-P cavity.

Figure 7.2 (a) Nano/microfiber lasers based on whispering gallery cavities in which light is trapped inside the fiber by total internal reflections and (b) subsequently enhanced by resonances known as WGMs.

Generally, polymer fibers can possess cylindrical shapes that offer circular cross-sections (Figure 7.2a), and therefore, has a full possibility to generate WGMs (Figure 7.2b). The WGMs are formed by total internal reflections at the cavity's interface and surrounding medium, offers negligible optical loss. WGM cavities provide very high cavity Q factor. WGM lasing has realized in cylindrical capillary tubes [97], microrings [129, 130], microdisks [21], microbubbles [131] but it has not been realized in polymer fibers embedding a gain material.

In this chapter, we show, for the first time, high Q factor WGM lasing from directly drawing polymer fibers embedding dye molecules. Lasing mechanism is systematically investigated by analyzing and fitting lasing modes of a single fiber, supported by size dependent characteristics of series of fibers and numerical simulation. We also show that these fiber lasers can be used for sensitive refractive index sensing.

7.2 Direct drawing polymer fibers

The solution used for the fibers is similar with that in the previous chapter but in different component's ratio. It is a mixture of about 11 wt% PMMA in DCM with epoxy resin and lasing dye R6G. Weight ratio between PMMA and the epoxy is ~1: 2 and R6G concentration is nearly 0.06 wt%. To form the fibers, a little solution is injected and deposited (in form of a droplet) on top edge of a special design glass substrate (Figure 7.3a). Then, a needle, whose optical image shown in Figure 7.4a, is approached down to immerse into the droplet. Subsequently, the tip is vertically retracted from the droplet and fiber is formed (Figure 7.3b). Finally, fabricated fibers are carefully placed on a special design substrate with a gap (Figure 7.3c). The gap helps to obtain free-standing fibers, convenient for optical characterizations. Fibers with various sizes are fabricated by different drawing speeds. Typically, the fiber's

diameter can be tuned from 5 to 100 μm . Figures 7.3e-k show achieved fibers with well uniform diameters from 10 to 100 μm .

Figure 7.3 (a)-(c) Microfibers are fabricated by directly drawing from polymer solution. (e)-(k) Top-down optical images of typical fabricated polymer fibers with decreasing diameters from about 100 to 10 μm .

Figure 7.4 (a) Optical image of the metal tip used to draw the fibers. (b) Optical image of a fiber and (c) its circular cross-section examined by a confocal microscopy. (d)-(e) Scanning electron microscope (SEM) images of a fiber indicates a smooth outer surface, beneficial for high cavity Q factor.

The fiber's cross-section and surface roughness are studied by a confocal microscopy (CM) and a scanning electron microscopy (SEM), respectively. The CM image, captured by scanning a horizontal fiber from highest to lowest point, exhibits a nearly circular cross-section (Figure 7.4c). On other hand, the SEM images (Figures 7.4d,e) demonstrate smoothness of the outer surface of fibers. These parameters are significant to realize high Q WGM lasing from the fiber cavity.

7.3 Whispering gallery mode lasers

Figure 7.5a plots PL intensity from a fiber with $D \approx 32 \mu\text{m}$ versus increasing pump pulse energy (PPE). Under PPE = 300 nJ, there is only weak broad spontaneous emission but sharp lasing peaks start to appear upon PPE = 500 nJ. Intensity of the lasing peaks gradually increases with rising of PPE. Figure 7.5b shows integrated PL intensity of the emission as a function of PPE. The nonlinear dependent curve is indicator of lasing with threshold of about 400 nJ.

Figure 7.5 (a) PL intensity of a fiber (image shown in the inset) as function of pump pulse energy. (b) Integrated PL versus the pump pulse energy indicated the lasing threshold.

One of the most important characteristics of WGM lasers is their polarization. WGMs are formed by total internal reflection and this phenomenon dependent on the orientation of electric field. The boundary condition for TE wave is different for TM wave. In a cylindrical structure like our fibers, the light is TE (TM) wave if its electric field is oriented in radial direction (parallel to fiber's axis) as depicted in Figures 7.6a-b.

Figure 7.6 Schematic views of WGMs (red ellipsoids) with (a) transverse electric (TE) or (b) transverse magnetic (TM) polarization (characterized by electric field orientation-white arrows). These two kinds of polarizations can be identified by monitoring light intensity after a polarizer (depicted in the figure). (c) Integrated PL intensity of a fiber laser after the polarizer versus different rotation angles.

To examine what kind of polarization occurs in inside the fiber, we used a polarizer depicted in Figures 7.6a and b. At initial stage (rotation angle is referred to 0 degree), the polarizer's axis is set to parallel to the fiber. Figure 7.6c plots the integrated PL intensity of a typical fiber versus rotation angle of the polarizer. The results show that the intensity is

maximum at 0 and 180 degree and minimum at 90 degree. The data is well fitted by the Malus' law. Similar results were obtained for other fibers. That means the identical polarization of laser light is TM. It is noted that the fiber and PPE were fixed during the measurement.

The position of lasing modes can be well explained (Figure 7.7) using the equation (3.12) for TM polarization. Given that diameter of the fiber $D = 32.36 \mu\text{m}$, refractive index of the fiber $n = 1.46$, the highest intensity peaks (referred to the first order because they have highest Q factor) are well matched with mode numbers $m = 231$ -239. Similarly, the second order peaks with lower intensity are well fitted with $m = 225$ -228. As a result, the lasing mechanism is indeed ascribed to WGMs.

Figure 7.7 Analysis of lasing modes based WGM theory. The arrows refer to higher order ($p > 2$) modes.

7.4 Size-dependent lasing characteristics

Our approach offers polymer fibers with various sizes so we are able to investigate size-dependent lasing characteristics. Figure 7.8a plots lasing spectra from three fibers with

Figure 7.8 Size-dependent lasing characteristics. (a) Spectra of three fibers with diameters about 22, 16 and 10 μm. (b) FSR and Q factor versus fiber diameters. (c) Fitting lasing modes for different fibers whose diameters shown beside each curve. The rectangular symbols refer to experimental mode energy while the small dot symbols are calculated energy based WGM theory.

decreasing diameters from 22 to 10 μm . Similar to our previous works, as the cavity size decreases, the FSR increases, leading to less number of lasing modes and single mode is obtained when fiber diameter down to about 10 μm .

Figure 7.8b shows FSR and Q factor as function of fiber sizes. The FSR curve is fitted well with a α/D function (α is a constant), supporting the WGM mechanism. Q factor is high, about 7×10^3 for $D = 20 \mu\text{m}$, increases to 8×10^3 for $D = 70 \mu\text{m}$. Even the single mode has $Q \approx 6.7 \times 10^3$, corresponding to spectral linewidth $\delta\lambda \approx 0.085 \text{ nm}$, which is much better than that of the VCSEL polymer lasers ($\delta\lambda = 1.8 \text{ nm}$) [132] or F-P nanofiber lasers ($\delta\lambda = 0.3 \text{ nm}$) [114]. This result indicates our advantages compared with most recent achievements on nano/microfiber lasers. Figure 7.8c plots the experimental and calculated photon energy of lasing modes of different fibers as function of mode numbers m . The data illustrates an agreement between calculation and observation for series of fiber sizes, once again, supported the WGM mechanism. In addition, for the fiber whose has the sing mode emission m is found to be 73 with considering $D = 9.87 \mu\text{m}$. The field distribution of this 9.87 μm fiber will be simulated later and the mode number m , equal to number of field maximum, is expected to be 73, the same as analytical calculation.

Lasing stability under continuously excitation for three fibers with different sizes is plotted in Figure 7.9. The lasing intensity of the fibers decreased with the pumping time due to the thermal effect or/and fluorescence quenching [133, 134]. The thermal effect or heat is generated by the pulse laser. This effect decreases the absorption efficiency of the dye molecules, therefore, resulting in the decreasing of emission. Unlikely, the fluorescence quenching is referred to the diffusion of oxygen through the fiber surface. In particular, the lasing lifetime of a fiber with $D \approx 30 \mu\text{m}$ is around 20 minutes or 7.2×10^4 excitation pulses. This value is comparable with the emitting polymer in Ref. [134] (7.6-20 minutes or 4×10^4 -

12×10^4 excitation pulses) and the organic-based laser devices in Ref. [135] (approximately 0.2-2.5 minutes or 1.2×10^4 - 15×10^4 excitation pulses). The good lasing stability of our fibers is due to excellent properties of the mixture polymer, which well protects dye molecules from oxidation.

Figure 7.9 Normalized integrated lasing intensity as function of excitation time for different fibers. The inset shows a close view for the fiber with $D = 12 \mu\text{m}$.

7.5 Numerical calculation of field intensity inside a fiber

Electromagnetic field distribution inside the fiber whose emitted single mode emission ($D = 9.87 \mu\text{m}$) was simulated using FEM supported from COMSOL multiphysics and the results are shown in Figure 7.10. Obviously, TM waves are well confined in a circular cross-section

for both first and second orders ($q = 1, 2$). The mode number m is, indeed, equal to 73 as expected for $q = 1$. The results confirms the WGM mechanism is therefore gives a real picture of how the TM wave is trapped and selectively enhanced by total internal reflections. To gain better clarity, intensity in the radial direction for both cases are plotted in Figure 7.10c and d, respectively. Most of the energy ($\sim 99\%$) is located in the fiber boundary except a faction of evanescent wave existing outside the interface. This is the reason for low optical loss or high lasing Q factor observed in our work.

Figure 7.10 (a), (b) Numerical simulation of magnetic field inside a fiber's cross-section ($n = 1.46$, $D = 9.87 \mu\text{m}$) for the first and second order and (c), (d) their field intensity in the radial direction, respectively. The dash line illustrates the edge of the fiber.

7.6 Refractive index liquid sensor

In the previous chapter, we have demonstrated the application of hemispherical lasers as sensitive vapor refractive index sensors. Here, using the same idea, we employ the fiber lasers for liquid refractive index sensing by using a setup shown in Figure 7.11a. A fiber with $D \approx 36 \mu\text{m}$ was fixed in a glass holder that is later filled with with 23 mL water.

Figure 7.11 (a) Scheme for investigation of refractive index sensing. (b) Red-shift of lasing mode is indication of increasing in index of the medium. (c) Lasing spectra of a fiber versus increasing THF concentration. (d) The red-shift value ($\Delta\lambda$) as function of refractive index variation (Δn).

The fiber was then pumped with fixed PPE and emission was recorded. After that 0.2 mL tetrahydrofuran (THF) was added and the process was repeated. To make sure that the solution is uniform, the fiber was only excited after 45 seconds counted when THF was added.

The presence of THF leads to a thin layer formation (THF molecules) covered the fiber surface that expands the original cavity length, leading to red-shift of lasing mode as depicted in Figure 7.11b [18]. By monitoring the shift value ($\Delta\lambda$) with the change of refractive index (Δn), sensor's validity should be verified. Typically, relationship between Δn and $\Delta\lambda$ is linear dependence [136].

Figure 7.11c plots lasing spectra from the fiber with increasing THF concentration. The lasing envelope exhibits red-shifting. We measured the shifting value ($\Delta\lambda$) and calculated the variation of refractive index (Δn) following Ref. [137] and their relationship are shown in Figure 7.11d. The linear dependence of $\Delta\lambda$ on Δn validates the fiber sensor. The sensitivity is $S = \Delta\lambda/\Delta n$, which can be extracted from slope value of the fitting line, is found to be about 300 nm/RIU. This sensitivity is one order better than previous report based on microsphere resonators [136].

7.7 Summary

In conclusion, we have demonstrated WGM lasing from directly drawing polymer fibers with excellent lasing characteristics such as low threshold (400 nJ/pulse) and narrow spectral linewidth (0.08 nm). Single mode operation, which is important for information processing, is obtained from the fiber with diameter about 10 μm . Owing to high cavity Q factor, the fiber laser has been successfully employed for liquid refractive index sensing with sensitivity

up to around 300 nm/RIU. Due to the good lasing performances, low cost mass production, we believe our polymer fibers are potential for future applications in photonic devices. Further possible developments such as coupled fibers, array or ordered fiber structures should be interesting for not only improving lasing performances but also interesting for optical integrated circuit and nonlinear optical effects.

Chapter 8

Vernier effect and enhanced sensitive sensor of coupled polymer fiber lasers

8.1 Introduction⁵

In the previous chapter, WGM polymer microfiber lasers have been investigated with excellent lasing performances including, low threshold, high Q factor and long lifetime. Application of fiber lasers for refractive index liquid also demonstrated. In this chapter, we will show a significant development of fiber lasers to a new higher level: coupled fiber lasers. Here, we have realized single mode lasing operation from coupled fiber lasers via the Vernier effect. The coupled structure is also more favourable compared with a single cavity for biological sensing application because it has large mode spacing and high cavity Q factor, which are not possible to achieve in a single WGM cavity.

Single mode emission and high Q factor or narrow spectral linewidth are two most desired parameters of microlasers. Single mode operation is important for on-chip and optical integrated systems [88]. Generally, it can be easily realized by distributed feedback (DFB) cavities due to their strict resonant conditions [138-140]. However, it is not the case for WGCs, in which resonant modes are very dense, single mode can be obtained only when cavity size is very small, close to 10 μm [32, 36, 75]. On other hand, we know that cavity Q

⁵Contents in this chapter has appeared in "Coupled polymer microfiber lasers for single mode operation and enhanced refractive index sensing," *Adv. Optical. Mater.* **2**, 220-225, (2014). ([Inside cover image](#)).

factor of WGCs linearly decreases with their size. As a result, single WGM lasing typically has a low Q factor.

The conflict problem between high Q factor and single mode emission can be solved in a coupled cavity, in which the single mode is generated via the Vernier effect without deteriorating laser Q factor [141-147]. In addition, the possibility to couple with other component is not only significant for improving performances of real photonic devices [148-150] but also for fundamental studies with many novel phenomena such as slow light, induced transparency and absorption [151, 152]. Vernier effect has been realized in coupled semiconductor, optofluidic, silica/glass fiber resonators [143-147]. However, this effect has never been reported in coupled WGM polymer fiber lasers.

8.2 Vernier effect

The Vernier effect has been widely used in photonic technology especially for optical filters and wavelength division multiplexing devices [141, 142]. It is generally observed in a coupled cavity. The working principle of Vernier effect is depicted in Figure 8.1. The coupled cavity is supposed to consist of two asymmetric cavities with FSRs of individual cavities are FSR_1 and FSR_2 , respectively (Figure 8.1a and b). The effect happens when resonant modes in individual cavities interact with each other and the resonant modes that matching with the resonance condition of both resonators are enhanced while others are depressed. As a result, the FSR of the coupled structure has significantly increases to $\text{FSR}_{12} = M \times \text{FSR}_1 = N \times \text{FSR}_2$, where M and N are co-prime integer numbers (Figure 8.1c). The expansion of FSR is a key factor to achieve single mode lasing emission [75].

Figure 8.2 shows numerical calculation of field distribution of transverse magnetic (TM) modes in a coupled cavity using FEM supported by COMSOL Multiphysics. The

coupled structure consists of two cylindrical cavities with radii 1.5 and 2.237 μm , respectively. Refractive index of the two cavities is 1.48. The two structures are not contacted but separated by a small gap, about 0.05λ [153]. This small distance allows strong interaction between WGMs in each cavity.

Figure 8.1 (a)-(c) Resonant modes characteristic of the first, the second, and the coupled cavities. FSR of the coupled cavity largely increases due to the Vernier effect.

Herein, λ is resonance wavelength, calculated by using WGM theory and characterized by q - mode order and m - mode number. Figure 8.2a shows field distribution at $\lambda = 630$ nm, which is the resonant wavelength for both cavities. It is corresponding to $q = 1$, $m = 28$ for the big cavity and $q = 1$, $m = 18$ for the small one. In this case, the coupled cavity is considered to be on resonance supported by clear resonance patterns (referred to number of peaks in intensity, which is equal to mode number m , along the circumference of the fiber)

with high intensity. However, the situation is different in Figure 8.2b where only the big cavity is on resonance at $\lambda = 651.3$ nm ($q = 1$, $m = 27$), the field intensity is weak with unclear resonance patterns. This observation indicates the mode suppression. The simulation validate the Vernier effect in coupled WGCs, opens a possibility to realize single mode emission via Vernier effect.

Figure 8.2 Normalized magnetic field distribution in a coupled cavity for a wavelength that fulfilled resonance condition of (a) both cavities and (b) only the big cavity.

8.3 Vernier effect in coupled fiber lasers

Figure 8.3 show a scheme about how separated and coupled fibers are studied. The two single fibers with suitable sizes are placed on the top surface of a DBR then immersed into water and investigated subsequently (Figure 8.3a). After that, they are directly forced to

contact to form a coupled fiber and then it is fixed on the DBR using a PMMA glue (Figure 8.3b). The function of the DBR is to prevent optical loss leaked through the substrate, therefore, significant for achieving low lasing threshold [34].

Figure 8.3 (a) Single separated fibers and the coupled fiber are subsequently investigated under optical pulse pumping.

As discussed above, FSR is a convenient parameter to verify the existence of the Vernier effect. For WGMs generated inside cylindrical fiber 1 and fiber 2, as shown in Figure 8.3, their FSRs can be roughly estimated:

$$\text{FSR}_{1(2)} = \frac{\lambda^2}{\pi n_e D_{1(2)}} \quad (8.1)$$

where D_1 and D_2 are diameters of fiber 1 and 2, respectively; n_e is effective refractive index of the fibers. When these fibers are coupled and the Vernier effect takes place, the FSR largely increase and can be expressed as below [143]:

$$\text{FSR}_{12} = \frac{\lambda^2}{\pi n_e (D_2 - D_1)} \quad (8.2)$$

Figure 8.4 plots emission spectra of the fiber A under excitation with pump fluence of 1.06 and 1.70 $\mu\text{J mm}^{-2}$. At low energy, there is only broaden weak spontaneous but stimulated emission with sharp lasing modes appeared under the higher excitation energy.

Figure 8.4 Spectra from fiber A indicates the evolution from spontaneous to stimulated lasing emission.

Figure 8.5 shows single mode emission obtained via the Vernier effect from the so-called coupled fiber AB that consists of two asymmetric fibers, the fiber A and fiber B. Figures 8.5a and b plots emission from the fiber A and B under the same pumping energy of 2.12 $\mu\text{J mm}^{-2}$. The FSR_A and FSR_B are determined to be 1.4 and 2.2 nm and it is found that $3 \times \text{FSR}_A \approx 2 \times \text{FSR}_B$, so the Vernier's resonance condition is fulfilled. Furthermore, we have a quick check on the $\text{FSR}_{A(B)}$ to make sure it is product of WGMs. Using the equation (8.1)

with considering that the resonant wavelengths from fiber A and fiber B are the same $\lambda_A = \lambda_B = 608$ nm and $n_{\text{eff}} = 1.48$ then $\text{FSR}_{A(B)}$ can be estimated. It is found that $\text{FSR}_A = 1.46$ nm for $D_A = 54.5$ μm and $\text{FSR}_B = 2.24$ nm for $D_B = 35.5$ μm . $\text{FSR}_{A(B)}$ calculated by the equation (8.1) is consistent with observations that confirms the WGM mechanism is origin of lasing action in our fibers. It is noted that diameters of the fiber A, fiber B are estimated from their optical images.

Figure 8.5 (a), (b) Lasing spectrum of fiber A, B under upon pump fluence of $2.12 \mu\text{J mm}^{-2}$, respectively. (c) Single mode emission is generated in the coupled fiber AB versus increasing pumping energy. (d) Lasing threshold behavior of the fiber A, the fiber B and the coupled fiber AB. The inset shows Vernier mode spacing of the coupled fiber AB at pump fluence of $3.61 \mu\text{J mm}^{-2}$.

Figure 8.5c plots the emission from the coupled fiber AB versus pump fluence in which the single mode is successfully generated. The single mode lasing appears under pump fluence of $2.55 \mu\text{J mm}^{-2}$ and remains stable under higher pump fluence up to $3.40 \mu\text{J mm}^{-2}$. We claim this mode is Vernier mode based on two factors. Firstly, the single mode appears nearly wavelength of 612 nm that is close to resonant wavelengths of both fiber A and fiber B. Secondly, under pump fluence of $3.61 \mu\text{J mm}^{-2}$, second Vernier mode is developed and FSR_{AB} is determined to be 4.2 nm. On other hand, using the equation (8.2) prediction of FRS_{AB} is 4.18 nm that is very close to the observation (4.2 nm). We, therefore, conclude that the single mode operation in the coupled fiber AB is a production of the Vernier effect. Figure 8.5d plots integrated PL intensity of emission from the fiber A, fiber B and the coupled fiber AB. The lasing threshold of the fiber A(B) are around $1.6 \mu\text{J mm}^{-2}$ lower than that of the coupled fiber AB ($2.7 \mu\text{J mm}^{-2}$).

The Vernier effect in coupled fiber lasers is highly repeatable. Figure 8.6a plots lasing emission from a single fiber namely fiber D with $\text{FSR}_{\text{D}} = 1.5 \text{ nm}$. When this fiber is coupled with another fiber (fiber C) to form the so-called coupled fiber CD, FSR_{CD} largely increases to 4.4 nm (Figure 8.6b). This value is around triple FSR_{D} . Using the equation (8.2) with considering that $\lambda_{\text{C}} = \lambda_{\text{D}} = 640 \text{ nm}$, $D_{\text{C}} = 36 \mu\text{m}$, $D_{\text{D}} = 55 \mu\text{m}$, the calculated FSR_{CD} is 4.6 nm, highly consistent with the experiment.

Table 8.1 indicates FSR of coupled fibers versus diameters of individual diameters (ΔD) for experiment and calculation. The consistent between observation and prediction verifies the existence of the Vernier effect and its contribution for the expansion of FSR and the single mode operation.

Figure 8.6 (a) Multi lasing mode from single fiber D and (b) lasing mode with large FSR from coupled fiber CD. FSR of coupled fibers as function of $\Delta D = D_2 - D_1$, a comparison between experimental observation and calculation using the equation (8.2).

Diameter of fibers (μm)		FSR of coupled fibers (nm)	
D_1	D_2	Calculation	Experimental
35.5	54.5	4.2	4.2
36.0	55.0	4.6	4.4
24.1	38.1	5.7	5.5

Table 8.1 Experimental and calculation (based on the equation (8.2)) of FSR as versus diameters of isolated fibers in coupled structures [38].

8.4 Refractive index sensing and enhanced sensitivity mechanism

Similar to microlasers, large FSR and high Q factor are two desired parameters for any biosensors based WGM technology [18, 107, 154]. Great FSR is convenient for distinguish the shifting of cavity mode while high Q factor offers higher sensitivity. As a result, a coupled cavity is favourable tool for sensing application.

Here, we employ the coupled fiber CD for liquid refractive index sensing based on a setup depicted in Figure 8.7a. Ethanol is used to modify the water medium. The two channel “in” and “out” help to keep the volume solution inside the holder nearly unchanged. It is important to maintain the position of excitation spot.

Figure 8.7 (a) Schematic setup for investigation of refractive index sensing. Thin layer of sensing material on the fiber surface (b) is a main reason for red-shift of lasing mode (c).

The existence of ethanol leads to a thin layer covered surface of the coupled fiber CD as shown in Figure 8.7b. This layer affects the WGM cavity and leads to resonant shifting (Figure 8.7c). The sensor's sensitivity (S) is ratio between the shifting value ($\Delta\lambda$) and deviation of refractive index (Δn): $S = \Delta\lambda/\Delta n$.

Figure 8.8 depicts the origin of the red-shift due to the thin layer (thickness ΔR) and how to estimate the shifting value [18]. After formation of the layer, the WGM is expanded. Considering in a wavelength range as shown in Figure 8.8b, the shifting value $\Delta\lambda$ can be estimated using simple mathematic equation for two triangles AOB and A'OB', where $AB = \lambda$, $A'B' = \lambda + \Delta\lambda$, and $AO = BO = R$. We have,

$$\frac{\Delta\lambda}{\lambda} = \frac{\Delta R}{R} \quad (8.3)$$

Figure 8.8 (a) Depict of WGM in bare sphere cavity and (b) in cavity with addition thin layer based on Ref. [18].

Figure 8.9 (a) Spectra of the coupled fiber CD versus increasing ethanol concentration and (b) Lasing spectra of a single fiber namely fiber E under the same sensing condition, posses with increasing refractive index. (c), (d) Magnitude spectra around 641 nm in (a) and 645 nm in (b), respectively. To gain clarity, all curves in (a)-(b) are shifted vertically. (e) The shifting value $\Delta\lambda$ as function of Δn for the coupled fiber CD and fiber E.

As discussed earlier, using the coupled cavity for biosensing is favourable because of their larger FSR and higher sensitivity compared with single cavity. This advantage has been observed using the setup in Figure 8.7a. By continuous adding a little ethanol (0.1 mL) into water (fixed volume 20 mL), the spirit concentration (C_E) increases from 0 to 1.18 wt% corresponding to Δn from 0 to 7.1×10^{-4} RIU. Lasing emission is recorded with increase ethanol fraction and plotted in Figures 8.9a and b for the coupled fiber CD and the single fiber E, respectively. It is clear that lasing mode red-shifts versus increase ethanol concentration or refractive index, verifying the sensing mechanism. For gaining better view, spectra near main lasing modes are magnified (Figures 8.c, d). The coupled fiber CD obviously demonstrates a larger red-shift in comparison with that from the fiber E. For example, for the highest ethanol concentration, the shift value for the coupled fiber CD is 0.28 nm but only 0.15 nm for the single fiber E. When investigating relationship between $\Delta\lambda$ and Δn , we found a linear dependence as shown in Figure 8.9e. This result confirms the validity of our sensors. The calculated sensitivity for the coupled fiber CD is 398 nm/RIU and for the fiber E is 210 nm/RIU. The improvement factor in sensitivity is, therefore, $398/210 = 1.9$ or nearly double.

Using the thin layer theory and considering that both single cavities in the coupled structure contributed their own sensitivity to final sensitivity of the coupled one, we are able to explain the experimental observation. Given resonance shift of the coupled fiber CD ($\Delta\lambda_{CD}$) and fiber E ($\Delta\lambda_E$) are function of the thin layer ΔR then they can be estimated as: $\Delta\lambda_{CD}/\lambda = \Delta R/R_C + \Delta R/R_D$ and $\Delta\lambda_E/\lambda = \Delta R/R_E$ where R_C , R_D , R_E are radius of the fiber C, the fiber D, the fiber E, respectively. The calculated improvement factor is $\Delta\lambda_{CD}/\Delta\lambda_E = (R_C + R_D)R_E/R_C R_D = 2.1$ that is highly consistent with the experiment (1.9).

8.5 Summary

In conclusion, we demonstrate the full possibility of integrating between polymer fiber cavities. It is an important for the development of all polymer fiber optical switches/filters [155], beneficial for optical integrated circuits. Particularly, we have realized the Vernier effect in coupled fiber lasers with achievement of narrow spectral linewidth (0.09 nm) single mode lasing operation. Interestingly, our coupling structure is not only favourable for laser sources but also biosensors due to their large FSR and high Q factor. The results show the advantages of using coupled for liquid refractive index sensing such as easies to distinguish the resonant shift and better sensitivity compared with the single fiber sensor. Owing to low cost and excellent lasing properties, our findings demonstrate that polymer fibers lasers are very promising candidates for future flexible photonic devices.

Chapter 9

Summary and future directions

9.1 Summary

Investigation of organic materials for flexible photonic devices and technologies is a significant work that will open up new kind of thin and flexible optoelectronics devices. Particularly, optical WGM microcavities are interesting structures because they are the core of optical switches, filters and lasers, which have potential applications in optical integrated systems.

In first part of this thesis, we have presented the basic knowledge of a laser device and optical microcavities. Particularly, we focus on WGM cavities and their important factors such as characteristic equations, position of optical modes, quality factor and mode volume. We demonstrate that intensity field distribution of a microsphere/cylinder can be obtained using finite element method supported by COMSOL multiphysics. Following, whispering gallery mode microlasers including their development and current challenges are briefly discussed.

Next, we present our successfully fabrications and optical characterizations of three novel WGM microresonators including polymer droplets, hemispheres, and fibers. These structures are surface tension induced so they have almost atomic smooth outer surface and circular cross-section, thus enabling for high quality resonators. By doping laser dyes into these cavities, optically pumped multi and even single longitudinal mode lasers with narrow spectral linewidth (~ 0.08 nm), clear mode spacing and low lasing threshold (~ 400 nJ/pulse) are achieved. Owing to high Q factor and strong photostability, the hemisphere and microfiber laser have exhibits their potential applications as sensitive vapor and liquid

refractive index sensors. Especially interesting, we demonstrate the possibility to integrate the fiber-fiber cavities by realization of the coupling Vernier effect from coupled fiber lasers. This success allows us to get single mode operation (without reducing cavity size) and enhanced the sensor's sensitivity. In conclusion, the main advantages of our cavities are the straightforward fabrication, ease to incorporate functional active materials, mechanical flexibility (bendable and stretchable) and high Q factor. We believe our finding will promote the development of flexible optical devices.

Even though we have demonstrated some impressive performances of flexible microlasers and sensors, several works need to be continuously investigated to employ the structures as compact optical devices. Below, we show several promising directions.

9.2 Future directions

9.2.1 Quantum dot lasing based on hemispherical cavities

Quantum dot (QD) is a semiconductor nanostructure that consists of only hundreds to thousands of atoms [156]. Due to ultrasmall size, QDs are a bridge between solid state and single atoms, and have mix properties of them [157]. QDs can be fabricated by several techniques but here we focus on QDs that synthesized by colloidal chemistry, which generally called colloidal QDs (CQDs) [157]. QDs are novel and very promising gain material for lasing applications. Compared with traditional 2D confinement quantum wells (QWs) laser, QD lasing is better in temperature stability and narrower emission line [157]. However, realization of lasing action based QDs was challenging issue due to ultrafast nonradioactive Auger recombination.

In 2000, optical gain and stimulated emission of CQDs was reported by V. I. Klimov, et al, [158]. Since then, loads of effort have been spent to obtain CQD lasing by numbers of configurations such as DFB [138, 159, 160], random cavities [161], and especially WGCs [75, 162-166]. Very recently, multicolor (red, green, blue) vertical cavity surface emitting lasers (VCSEL) based on CQDs have been successfully fabricated [167]. Further development on CQD lasing is significant for the development of novel photonic devices.

Here, we demonstrate that our hemispheres can be served as high Q resonators for realization of CQD lasing. The CQDs herein are CdSe/CdS/ZnS core-multi-shells, fabricated by one-pot chemical method [168, 169]. Figure 9.1 shows low and high magnitude transmission electron microscopy (TEM) images of CQDs, indicating uniform dots.

Figure 9.1 (a) and (b) Low and high magnitude TEM images of CdSe/CdS/ZnS core-multishells colloidal QDs.

To make a solution for solid hemispherical cavities, we make a solution that consist of epoxy resin and hardner (ratio 10:1). By loading this composition into the hollow glass needle of the microplotter, hemisphere arrays can be deposited on top of DBR surface coated a hydrophobic layer. After formation, hemispheres will become solid by drying under ambient

conditions and at room temperature for 1-2 days. Later, we carefully immersed the whole structure in ethanol to remove the hydrophobic layer. To coat QDs on the hemisphere surface, QD solution was directly deposited on top the hemispheres the so-call drop casting technique.

To excite the QD coated hemisphere, we use the green pulse laser as previously. We achieved amplified spontaneous emission (ASE) at pump fluence of 0.95 mJ cm^{-2} and lasing emission at 1.01 mJ cm^{-2} . Spectral linewidth of lasing mode was about 0.12 nm.

Compared with traditional dye molecules, CQDs has many advantages for applications as gain material such as color tunability and photostability. Experimental has shown that QD has long PL lifetime compared with dye molecules [122]. Therefore, it is also expected that QDs lasing should have much longer lifetime in comparison with dye laser. To verify this hypothesis, we studied the degradation of lasing intensity under continuously excitation and found that the stability of QD lasing is at least one order magnitude better than dye lasing.

In conclusion, we have preliminarily employed the hemispheres as template resonators for realization of CQD lasing. Further development such as blue, green lasing emission is appreciated the potential application of CQDs for display and solid lighting. Furthermore, finding a suitable method to incorporating CQDs in a polymer matrix without reducing their photoluminescence quantum yield is needed to investigate. The polymer would protect the CQDs from oxidation and does not affect their optical properties. It is also interesting to replace the CQDs by other kind of gain materials such as semiconductor quantum nanorods and organic light emitting polymers to obtain different kind of lasers.

9.2.2 Electrically pumped hemisphere lasers

Even though the hemispheres microlasers have number of excellent lasing properties, they require a powerful pumping laser source, which is expensive and inconvenient. Recent development in flexible lasers has successfully realized LED pumped organic lasers [28, 170]. This achievement significantly reduces size of whole laser system and much convenient for applications. Following this idea, we propose to realize LED pumped hemisphere lasers. To achieve this target, we need to design an appropriate structure that combines a LED and a DBR. The DBR should have maximum transmission for the LED light but high reflection for the emission of the hemispheres.

Realization of LED pumped hemisphere laser is a challenging issue. Currently, the lowest achievable threshold is about $3 \times 10^4 \text{ W cm}^{-2}$ which is two order magnitude higher than the LED pumped organic laser [170]. However, we expect that the lasing threshold can be further reduced by optimization of the whole system. The hemisphere needs to have perfect circular shape with smooth outer surface to reach the highest possible Q factor. Optimization of dye concentration is needed to investigate because it will strongly reduce the lasing threshold.

9.2.3 Coupling between hemisphere and polymer waveguide

Coupling the hemisphere with a waveguide is a significant task. It allows to effectively excite the hemisphere via the waveguide. In comparison with the free space excitation, it is more convenient and much higher efficiency. The emission from the hemisphere can be also extracted out through the waveguide. Successful investigation of the coupling system will bring hemisphere laser in real optical devices such as all polymer optical filters, optical switches, and especially components in photonic integrated circuits.

Figure 9.2 shows a coupling configuration between a hemisphere and a polymer waveguide. Half of the fiber is an input port while the rest is an output port. The excitation can be guided into the input port. This light is coupled to and excites the hemisphere. Then, emission of the hemisphere is coupled to the fiber and is guided out via the output port for specific purpose. More complex configuration can be established as shown in Figure 9.3 where two hemispheres are integrated with a single fiber. In this system, one hemisphere can act as a filter for optical modes generated in the other one, the Vernier effect can be possibly realized. This system opens one more channel for controlling of the output signal.

Figure 9.2 Schematic of a polymer waveguide coupled hemisphere cavity.

Figure 9.3 Schematic of a polymer waveguide coupled two hemisphere cavities.

9.2.4 Coupling between fiber waveguide and fiber cavity

Owing to cost effective mass production and mechanical flexibility, polymer fibers are promising candidates for future flexible/plastic optical devices. Our fabrication approach (Chapter 7) offers fiber with wide-range tunability in size (5 to 150 μm in diameter). Small fibers can be served as low optical loss waveguide while big fibers can be used as whispering gallery cavities. Therefore, it is interesting and convenient to realize a polymer fiber waveguide coupled polymer fiber cavity as shown in Figure 9.4. Successful investigation is significant to achieve all-polymer-fibers optical components such as optical switches, optical filters.

Figure 9.4 Schematic of a polymer fiber waveguide coupled polymer fiber cavity.

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List of publications

A. Journal articles

1. **V. D. Ta**, R. Chen, and H. D. Sun, "Wide-range coupling between surface plasmon polariton and cylindrical dielectric waveguide mode," *Opt. Express* **19**, 13598-13603 (2011)
2. **V. D. Ta**, R. Chen, and H. D. Sun, "Self-Assembled Flexible Microlasers," *Adv. Mater.* **24**, OP60-OP64 (2012). ([Highlighted by Materialviews](#))
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4. **V. D. Ta**, R. Chen, L. Ma, Y. J. Ying, and H. D. Sun, "Whispering gallery mode microlasers and refractive index sensing based on single polymer fiber," *Laser Photon. Rev.* **7**, 133-139 (2013). ([Frontispiece image of January issue, 2013](#))
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11. Y. Wang, **V. D. Ta**, Y. Gao, T. C. He, R. Chen, E. Mutlugun, H. V. Demir, and H. D. Sun, "Stimulated emission and lasing from CdSe/CdS/ZnS core-multi-shell quantum dots by simultaneous three-photon absorption," *Adv. Mater.* **26**, 2954-2961 (2014).
12. Y. Wang, K. S. Leck, **V. D. Ta**, R. Chen, V. Nalla, Y. Gao, T. C. He, N. Zheludev, H. V. Demir, H. D. Sun, "Blue Liquid Lasers from Solution of CdZnS/ZnS Ternary Alloy Quantum Dots with Quasi-Continuous Pumping," ([Submitted](#)).

B. Patents

1. R. Chen, H. D. Sun, **V. D. Ta**, "Hemispherical microstructure arrays for GaN-based Light Emitting Diodes and Method for Forming the Arrays," [WO2013158038 A1](#) (2013).

C. Book chapter

1. R. Chen, **V. D. Ta**, and H. D. Sun, "Whispering Gallery Mode in ZnO with Hexagonal Cross Section," in *Handbook of Optical Microcavities*, A. H. W. Choi, ed. (Pan Stanford Publishing, 2014).

D. Conference proceeding and presentations

1. **V. D. Ta**, R. Chen, H. D. Sun, "Flexible Microresonators: Lasing and Sensing," SPIE Photonics West, San Francisco, California, United States (2014).
2. **V. D. Ta**, R. Chen, D.M. Nguyen, H.D. Sun, "Flexible Optical Microcavities and Their Sensing Application," European Conference on Lasers and Electro-Optics (CLEO Europe), Munich, Germany (2013).
3. Y. Wang, Y. Gao, **V. D. Ta**, H. V. Demir, H.D. Sun, "Coherent Random lasing from CdSe/CdS/ZnS quantum dots," The Japan Society of Applied Physics-Optical Society of America (JSAP-OSA) Joint Symposia, Kyoto, Japan (2013).

4. **V. D. Ta**, R. Chen, D.M. Nguyen, H.D. Sun, "Microlasers and Sensors from Self-Assembled Hemispherical Cavities," Asia Student Photonics Conference (ASPC), Osaka, Japan (2013).
5. **V. D. Ta**, R. Chen, H.D. Sun "Lasing from Flexible Microcavities and Their Applications," International Photonics and OptoElectronics Meetings (POEM), Wuhan, China (2013). (Invited talk)
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11. **V. D. Ta**, R. Chen, H.D. Sun, "Microlaser from Self-Assembled Hemispherical Resonator," Conference on Lasers and Electro-Optics (CLEO), San Jose, California, United States (2012).