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Abstract: Dye-doped polystyrene (DDPS) encapsulated in a silica-glass capillary with a diameter of 300 μm was fabricated through radical polymerization of styrene within the capillary. The coherent random lasing (RL) with full width at half maximum (FWHM) of 0.36 nm and a quality factor of 1608 was produced in the DDPS with the capillary when pumping at 532 nm. However, the incoherent RL with FWHM of 6.62 nm and a quality factor of 92 was produced in the DDPS without the capillary. A detailed investigation on this phenomenon by changing the diameter of the capillary and core refractive index (RI) reveals that there exists a strong whispering gallery mode (WGM) resonance in the capillary, which helps generate the coherent RL. The findings may open up a new approach for the fabrication of highly efficient photonic devices.

Keywords: whispering gallery modes; random lasers; coherent random lasing (RL)

1. Introduction

Whispering gallery mode (WGM) is optical resonance arising from light being trapped due to total internal reflection (TIR) at the boundary. The light propagating along the inner surface of the resonator gives rise to constructive interference when returning in phase after each round trip. This creates resonance features, with spectral positions and linewidths that depend on the geometry of the resonator, as well as the surrounding environment [1]. Recently, WGM resonance has been widely used to enhance the sensitivity of gas sensors, such as for detecting carbon dioxide and water [2–9]. WGM enhanced lasing has been observed in some materials and circular structures [10–18]. It has been reported that random lasings (RLs) are generated by WGM resonance in disordered systems. Chen et al. [19] modified SiO_2 nanospheres on the surface of ZnO nanorods, pumping out random laser light with the aid of WGM resonance effect of spherical particles. As for a glass capillary, WGM resonance easily occurs in the wall of the capillary if the refractive index (RI) of the filling medium in the capillary is greater than the RI of the glass capillary.

Many studies have been carried out pumping RLs by filling a capillary with medium [20–23]. In these works, the capillary was pumped laterally, so the emitted light was likely to generate WGM resonance. However, the effect of the WGM resonance of the capillary on the random lasing has not been reported yet.

It is well known that gain medium and scatterers are two necessary elements for RLs. For example, longitudinal plasmon resonance of nanorod [24] can increase fluorescence intensity, which may pump random lasing. In the fields of solar cells, a flexible polypyrrole-coated fabric counter electrode [25] was used, because of low production costs and conversion efficiencies. Removal of pollutants from wastewater also used organic dyes [26]. Here, N,N′-di-[3-(isobutyl polyhedral oligomeric silsesquioxanes) propyl] perylene diimide (DPP) [22,23], which is composed of perylene diimide (PDI) as the gain group and polyhedral oligomeric silsesquioxanes (POSS) with a diameter of 0.54 nm as the scattering group was selected for the RL medium. π–π stacking of the chromophores and other physical interactions further makes these materials suffer from the well-known aggregation caused quenching (ACQ) effect, so it is important to reduce these influences [27]. Such a molecule not only acts as a gain medium but also as a scatterer medium in the RL system. A dye-doped polystyrene (DDPS) solid sample encapsulated in the capillary was obtained by bulk polymerization of refined styrene with DPP. RL phenomenon was observed in the random lasing system. By comparing the random lasing behaviors of the polymer solid sample with and without capillary, the influence of WGM resonance on the random lasing was verified. In addition, the impacts of WGM of different sizes of capillary diameters and WGM under various RI environments on RLs were investigated.

2. Materials and Methods

2.1. DPP Synthesis and Random Lasing Measurements

DPP was prepared according to the previously reported procedures [28]. A Q-switched neodymium-doped yttrium aluminum garnet (Nd: YAG) laser (pulse duration 10 ns, repetition rate 10 Hz) with an output of 532 nm was used to pump the sample with a focus lens of f = 10 cm. Pump pulse energy was controlled by a Glan prism group. The pumping spot size was 100 nm. Emitted light was collected by using a fiber spectrometer (Ocean Optics, QE65 Pro, resolution 0.4 nm, integration time 100 ms).

2.2. The Preparation of Dye-Doped Polystyrene (DDPS)

5 mL of 10^{-3} M DPP in CH_2Cl_2 solution was prepared via volumetric flasks. The solution was transferred to a 25 mL glass test tube at room temperature. After the solvents were volatilized completely, the purified styrene solution containing 1 wt% refined 2,2′-Azobis (2-methylpropionitrile) (AIBN) was added and treated by ultrasonic for 10 min and cooled at room temperature. Five kinds of capillaries with inner diameters of 100 µm, 200 µm, 300 µm, 500 µm, and 700 µm were added into the glass test tube and sealed with a stopper. The glass test tube was placed in an oven at 60 °C for 3 h and then taken out and polymerized into a solid at room temperature for about 2 months to avoid bubbles in the polymer. The capillary was lightly shredded from the glass tube. The outer surface of the capillary was carefully wiped with CH_2Cl_2. The capillary encapsulated DDPS samples were then obtained.

2.3. The Preparation of Dye-Doped Mixed Solution (DDMS)

In order to reduce the additional influence of solvent polarity, the solvent was a mixture of $CHCl_3$ and CS_2. The volume ratios of $CHCl_3$ and CS_2 in the five samples were 10/90, 20/80, 30/70, 40/60, and 50/50, respectively. Every five capillaries, with inner diameters of 300 µm and outer diameters of 500 µm, were inserted into the five solutions so that a liquid column of about 10 cm was formed by the capillary action. The two ends of the capillary were then sealed by optical adhesive, and a capillary encapsulated dye-doped mixed solution (DDMS) sample was obtained.

3. Results

DDPS solid sample encapsulated in the capillary was obtained by bulk polymerization of refined styrene with DPP. The DDPS solid sample was obtained after peeling off the capillary. Under pumping of 532 nm light with energy of 50 μJ and 70 μJ, the emission spectra of DDPS with and without the capillary (inner diameter: 300 μm; outer diameter: 500 μm) are shown in Figure 1a, respectively. There were several sharp laser-like emission peaks emerging in the emission spectrum of DDPS with capillary. The position and intensity of the sharp peaks randomly changed at different moments, which is the inherent nature of random lasing behavior. Interestingly, after peeling off the capillary, sharp emission peaks disappeared and only a single narrowed peak at 580 nm with a full width at half maximum (FWHM) of about 6.62 nm ($\Delta\lambda_1$) was observed. Based on the experimental observation, it is easily deduced that the laser-like emission was possibly caused by the WGM of the capillary.

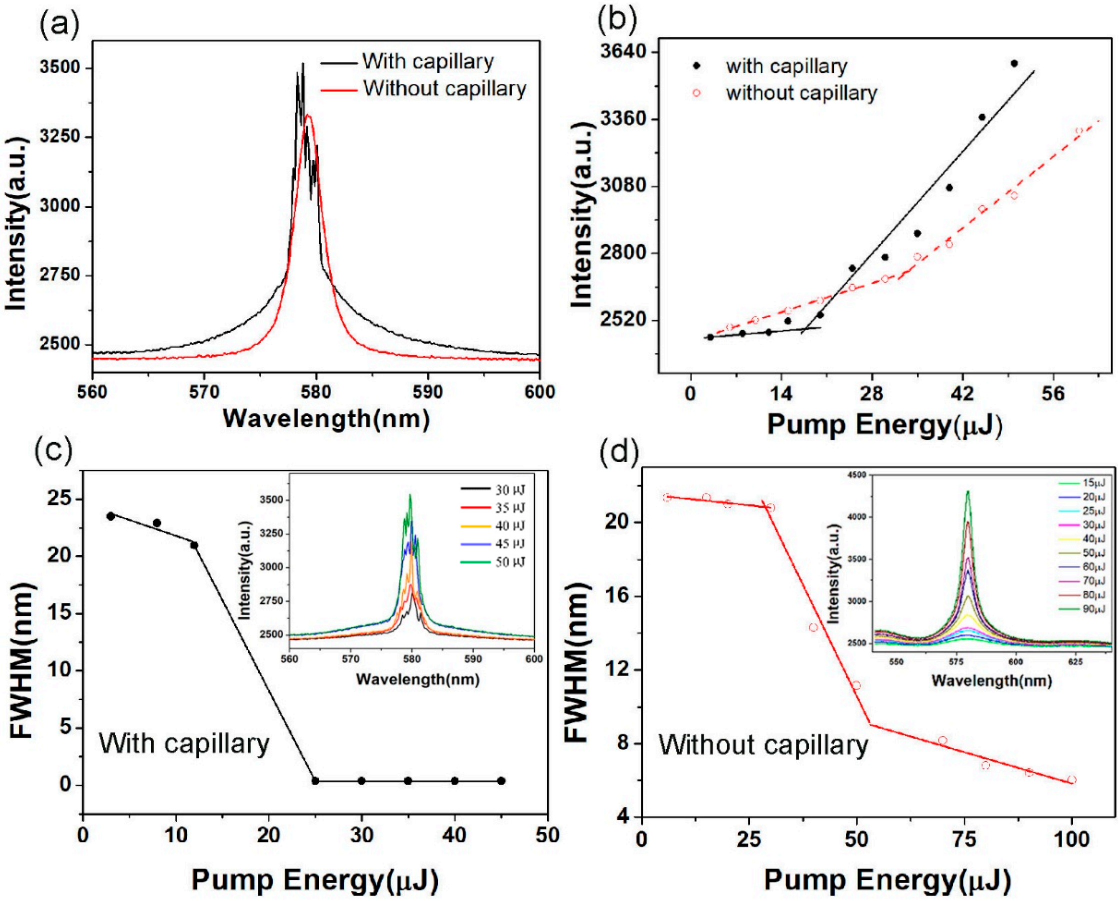

Figure 1. Emission spectra of 10^{-3} M N,N′-di-[3-(isobutyl polyhedral oligomeric silsesquioxanes) propyl] perylene diimide (DPP)-doped dye-doped polystyrene (DDPS) with and without a capillary (inner diameter 300 μm) under a pump of 532 nm light with energy of 50 and 70 μJ, respectively (**a**). The dependence of the emission intensity on pumping energy for emission from 10^{-3} M DPP-doped DDPS with and without a capillary (inner diameter 300 μm) (**b**). The dependence of the full width at half maximum (FWHM) on pumping energy with a capillary (**c**) and without a capillary (**d**) in the same condition.

It is worth pointing out that the single narrowed peak in Figure 1a is over 3 times narrower than that of the normal spontaneous emission spectrum of DPP in previous work [22]. It is well known that RL is divided into two classes based on the optical feedback: the incoherent [29–33] and coherent RLs [34–38]. For incoherent optical feedback, firstly suggested by Letokhov [39], light amplification occurs via a diffusion process, which results in spectral narrowing, typical of amplified spontaneous emission (ASE). On the contrary, coherent RLs exhibit a sequence of narrow lines [40] with FWHM < 1 nm, and the feedback mechanism is the closed loop scattering in the disorder medium. Figure 1a clearly shows that the stimulated emission with a capillary belongs to coherent lasing and the stimulated emission without a capillary belongs to incoherent lasing.

To further investigate the laser action, the dependence of the emission intensity on pumping energy was determined. As shown in Figure 1b, there are two abrupt changes of the slope at about 17.7 μJ in the black line and 33.4 μJ in the red dash, respectively. Both of two changes provided a signature for the appearance of the stimulated emission. Moreover, abrupt narrowing of FWHM observed in the dependence of FWHM on pumping energy in Figure 1c,d also verified the stimulated emission. Apart from these, the position and intensity of the separated sharp peaks from the sample of DDPS with capillary were found to randomly change at different moments, which is the inherent nature of coherent random lasing behavior. Therefore, coherent RL takes place with the sample of DDPS in the capillary and incoherent RL with DDPS alone. Corresponding thresholds were about 17.7 μJ (with a capillary) and 33.4 μJ (without a capillary), respectively, as shown in Figure 1b. This result implies that it is easier to generate RL with the assistance of the capillary. For DDPS with a capillary, the FWHM of each peak was about 0.36 nm ($\Delta\lambda_1$) and the quality factor (QF) was estimated to be around 1608 by the definition QF = $\lambda/\Delta\lambda$, where λ is the peak wavelength and $\Delta\lambda$ is the line width of the peak. The QF value of DDPS without a capillary was approximately 92, which is much smaller than 1608.

The inner diameter of a glass capillary is usually several hundred microns. After a beam of light is incident from the side, WGM resonance easily occurs in the capillary regardless of whether the refractive index of the filling medium in the capillary is greater than or less than the refractive index of the glass [28,41]. For example, when the refractive index of the capillary tube is less than that of the medium filled inside it and the angle of light incident from the side is appropriate, total reflection occurred at the interface between the capillary tube and the medium. On the other hand, the emitted light may have also been totally reflected at the interface between capillary and air, thus making the light continuously propagate forward along the annular tube wall. When the light propagation path formed a loop, interference was generated to form WGM resonance at both interfaces.

In light of the facts underlined above, the hidden mechanism for the different experiment phenomena above is depicted in Figure 2a. When there was a capillary around DDPS, there were exactly two interfaces: (1) an interface between the inner DDPS (RI: 1.59) and the inner face of capillary (RI: 1.5059); (2) an interface between the outer face of the capillary and air (RI: 1) around the capillary, which were recorded as the DDPS-Capillary interface and the Capillary-Air interface, respectively. When a 532 nm pulsed laser perpendicularly irradiated DDPS from the side, pump light entered DDPS and pumped DPP aggregates to produce fluorescence. Due to the scattering of DPP, many loops can be formed, as the yellow arrows show in Figure 2b, and each loop corresponds to a RL peak. When the fluorescence reached the boundary between DDPS and the capillary, due to the larger RI of DDPS than that of capillary glass, with the appropriate angle of incident light, most of the light was totally reflected in the DDPS-Capillary interface to continue propagation, which can form the first WGM, as the yellow arrows show in Figure 2b. The generation of the first WGM can be verified by Figure 6a, showing the RL spot located in inner wall of the capillary. At the same time, there was still a little part of the light propagating along the annular outer wall of the capillary, as the red dash arrows show in Figure 2b, which can generate a second WGM in the Capillary-Air interface. It is well known that if b/a (as shown in Figure 2b) is close to 1, high-mode-order WGMs can penetrate into the DDPS-Capillary interface so that its distribution extends into the gain medium [28,41]. This implies that the second WGM also makes a contribution to the gain. Next, we should discuss the WGM's impacts on RLs.

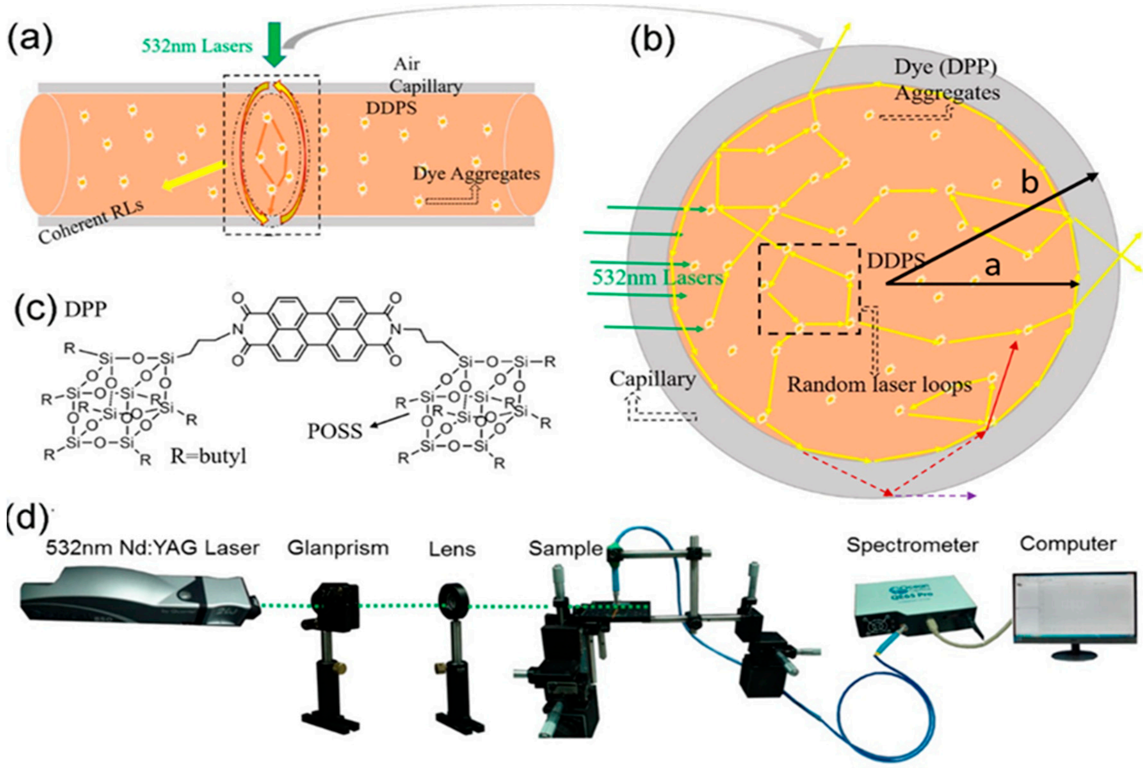

Figure 2. Schematic illustration of the mechanism responsible for the coherent random lasing (RL) from DDPS in the capillary (**a**,**b**). DPP molecular structure diagram (**c**). Random laser device diagram (**d**).

It was assumed that a beam of resonance light was generated from a small loop formed by the scatterers first and then was enhanced by WGM resonance at the boundary, and a laser was formed. Under such an assumption, the spectrum of RLs should be relatively regular, but the RLs shown in Figure 1a were not very regular. Therefore, it is very likely that there are three possibilities. One possibility is mentioned above, and the second possibility is that the light localized in WGM was scattered in the gain medium and was selected by the loop formed by scattered particles and then passed through the glass tube wall. Another possibility is that the closed loop paths dynamically changed owing to the vibration of the DPP aggregates, which makes the laser action more random.

Multiple scattering within the DDPS still exists after the capillary is removed, in which the cylindrical DDPS is exposed to air. Although the photon may generate WGM resonance at this interface, there was only one WGM, while for DDPS with capillary there were two WGMs, which greatly increased the stay time of photons in the gain medium and gave more chances for more photons getting into a loop (similar to resonant cavity) to obtain coherent RL. In contrast, for DDPS without a capillary, the residence time of the photon in the gain medium was significantly reduced. Therefore, it was less likely to produce coherent RLs but more likely to produce incoherent RLs.

As mentioned above, WGM is sensitive to optical cavity size, and we desire to know whether the WGM of various diameter capillaries (or various optical cavity sizes) can always boost RLs. Figure 3 shows the emission spectra of DDPS (10^{-3} M DPP) in capillaries with various inner diameters. Interestingly, coherent RLs can be pumped out for the sample with the small inner diameter (100–500 μm) of the capillary. When the inner diameter raised to 700 μm, the emission peak turned into a broader fluorescence peak, suggesting that the effect of WGM was so weak that it led to no RLs. For 100, 200, 300, and 500 μm diameter samples, the WGM was strong enough to make the gain larger than loss and boost RLs. Moreover, we also found that the thresholds of 100 μm, 200 μm, 300 μm, and 500 μm were around 30 μJ, 46 μJ, 90 μJ, and 27 μJ, respectively, and the QFs of them were about 1938, 1549, 1157, and 2690, respectively, which are shown in Figure 4. The high QF value indicates the existence of

WGM. The above results indicate that 500 μm DDPS with a capillary was the most ideal RL material in this work.

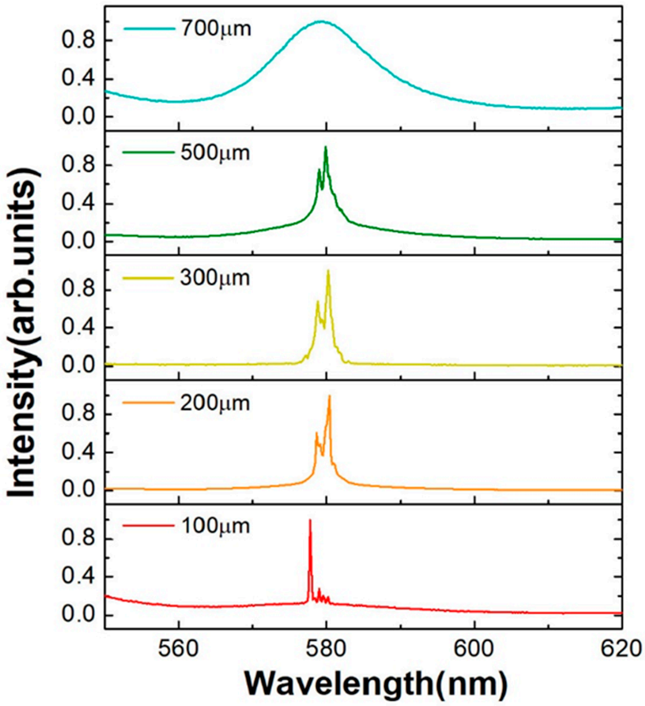

Figure 3. Emission spectra of confirmed concentration of DPP (10^{-3}M) in DDPS with a series of inner diameter capillaries from 100 to 700 μm.

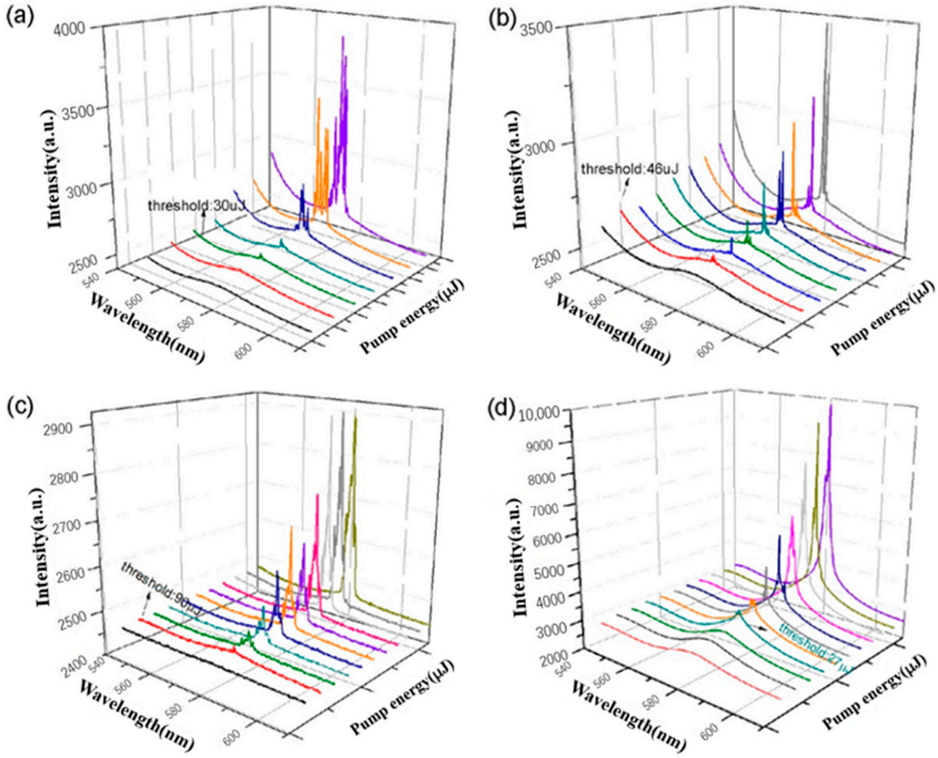

Figure 4. Evolution of emission spectra of DDPS in capillaries with different diameters under various pump energies, 100 μm (**a**), 200 μm (**b**), 300 μm (**c**), and 500 μm (**d**).

To explore the effect of WGM resonance with different refractive indexes (RIs) on RLs, the different refractive index samples were prepared by changing the proportion of mixed solution (or dye-doped mixed solution (DDMS)) in the capillary, which is not only simple, but also can reduce the influence of polarity. Five samples, shown in Figures 5 and 6, were named as S1 (10/90), S2 (20/80), S3 (30/70), S4 (40/60), and S5 (50/50), respectively; their RIs (n_1) were 1.6031, 1.5778, 1.5585, 1.5401, and 1.5201, respectively, and the RI of the fused-silica capillary (n_2) was 1.5059, so the refractive index contrasts $\Delta n = n_1 - n_2$ between them were 0.0972, 0.0719, 0.0526, 0.0342, and 0.0142, respectively. In all cases, the inner liquid had a higher refractive index than the surrounding capillary. As shown in Figure 5a, the emission peaks of S1, S2, and S3 were sharp, while the emission peaks of S4 and S5 were broad fluorescence peaks, indicating that WGM of the capillary does not always enhance RLs and only DDMS with high RI contrast can obtain coherent RLs with WGM. It has been reported that intrinsic leakage loss of given WGM increases with the decrease of the refractive index [42]. This means that the effect of WGM is inversely proportional to the environmental RI. The results above show that only strong WGM can boost RLs, which is in line with the results of the effect of inner diameters of capillaries on the RLs. As shown in Figure 5b, the thresholds were ordered by 0.66 μJ (S1) > 0.79 μJ (S2) > 1.06 μJ (S3). This result indicates that the threshold decreased with an increasing in the RI of the inner liquid, which illustrates that the stronger the WGM resonance, the more favorable it was to obtain RLs. This conclusion is further confirmed by the observation in Figure 6. Figure 6a,c,e shows the lasing microscope images from S1, S2, and S3 under dark environment. Figure 6b,d,f shows S1, S2, and S3 under white light irradiation. We detected the random laser from S1, S2, and S3. The bright spot in the picture was the brightest, which belonged to RLs. It is clear that the distance we marked in Figure 6a was 300 μm, which was, rightly, the inner diameter of the capillary. Therefore, the bright emission of RLs came from the capillary inner wall, where the WGM existed, which confirms that WGM boosted RLs. Additionally, the emission intensities of the RLs were ordered by S1 > S2 > S3 under the same pumping energy, which confirms that the bigger Δn brought stronger WGM and further led to stronger coherent RLs. Figure 6 also shows that, although the closed-loop was formed in the disorder medium, a coherent random laser was not produced from DDPS in the intracavity of the capillary owing to the fact that the pumping energy was lower than the pumping threshold.

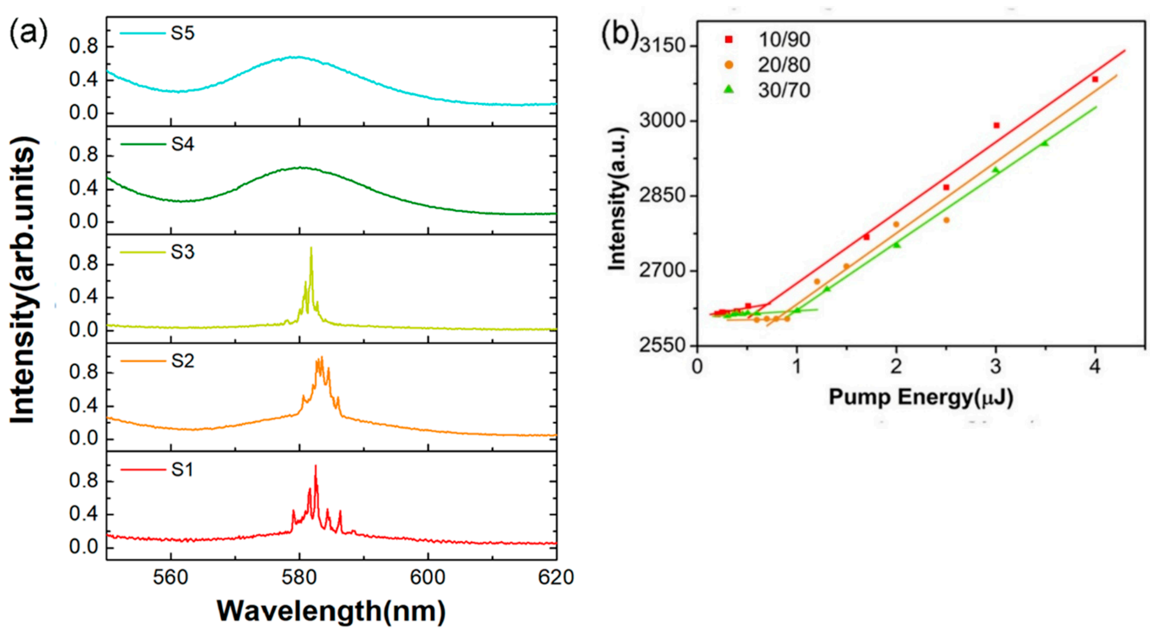

Figure 5. (**a**) Emission spectra of the confirmed concentration of DPP (10^{-3}M) in DDMS with a series of RIs. (**b**) The dependence of emission intensity on pump energy.

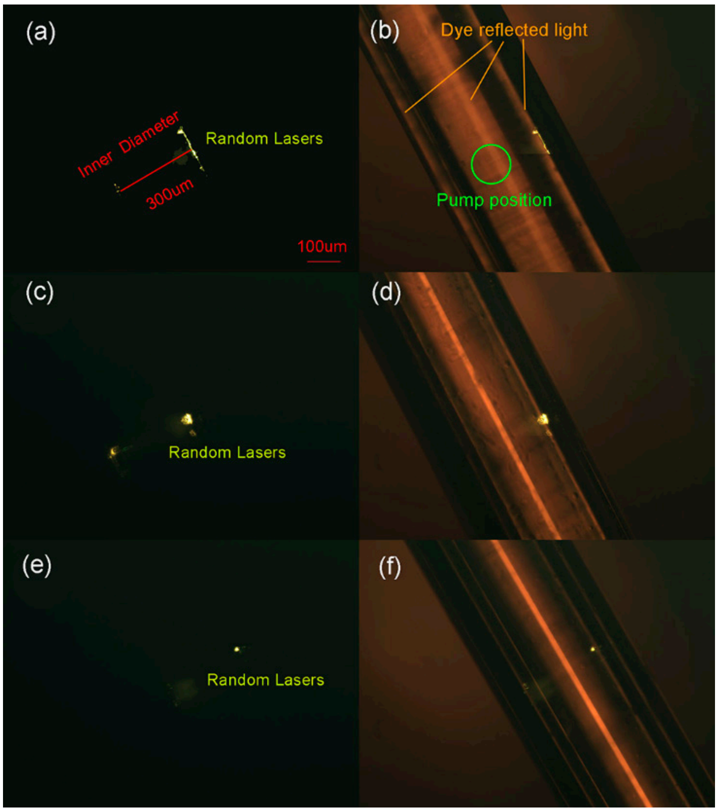

Figure 6. (**a**,**c**,**e**) are the lasing microscope images from S1, S2, and S3 under dark environment. (**b**,**d**,**f**) are pictures from S1, S2, and S3 under white light irradiation. The pump position is marked by green circle marks.

4. Conclusions

In conclusion, dye-doped polystyrene was made by slow polymerization in fused-silica capillaries with various diameters. The coherent RLs with FWHM of 0.36 nm and QF of 1608 were obtained in the DDPS with the capillary, which was attributed to the WGM waveguide of the capillary. In contrast, the incoherent RLs with FWHM of 6.62 nm and a QF of 92 were observed in the DDPS without the capillary. The experimental phenomena was explored under various conditions and explained in detail. The results demonstrate that WGM resonance excites coherent RLs through the small diameter of the capillary (100–500 µm) and there is a high RI contrast between DDMS and glass ($\Delta n \geq 0.03$). The high optical gain and low lasing threshold were attained in the WGM waveguide of the capillary, which makes it a promising candidate for efficient photonic devices.

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Conflicts of Interest: The authors declare no conflict of interest.

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