Unidirectional Lasing From a Spiral-Shaped Microcavity of Dye-Doped Polymers

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***Abstract*— We experimentally demonstrate fabrication of a spiral-shaped polymer microdisk cavity by femtosecond laser direct writing via two-photon polymerization of dye-doped resins. This spiral microresonator supports highly directional emission from the notch with a high-Q factor exceeding 1.6 × 10^3 and a low lasing threshold at room temperature. Our numerical simulation results reveal that clockwise propagating high-Q whispering gallery-like modes may couple to the counterclockwise modes and give the directional output. Benefiting from the low cost and good chemical compatibility of polymer materials, the fabrication of such spiral-shaped polymer microlasers with high-Q and unidirectional emission holds great potentials for use as elements of integrated organic optoelectronic devices.**

***Index Terms*— Microfabrication, cavity resonators, optical polymers.**

I. Introduction

OPTICAL microcavities such as microdisks, microrings and microspheres are becoming the basic building blocks of integrated optoelectronic devices, and may find broad applications in optical communications, photonic circuits and optical sensors [1]–[4]. Light confinement in such microcavities with so-called whispering-gallery modes (WGMs) can be produced based on the total internal reflection of photons by the surface of rotational symmetrical cavities, leading to unique lasing features including high quality (Q) factor, low lasing threshold and small mode volume [3]–[6]. However, isotropic lasing emission of WGMs microresonators with rotational symmetry results in unavoidable low collection efficiency by lens coupling. One of the proposed solutions to overcome such limitation is to break the rotational symmetry by deforming the cavity shapes, e.g., the annular cavity, the spiral cavity, and the limaçon cavity, which can radiate light at certain directions without significantly degrading the high-Q factor [7]–[13].

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Because of the intrinsic three-dimensional (3D) prototyping capability with nano-scale spatial resolution, femtosecond laser direct writing (FsLDW) has shown its versatility in fabricating a variety of microsystems such as microoptical, micromechanic and microfluidic devices [14]. Recently, the fabrications of microcavities by FsLDW have also been demonstrated, e.g., in glass and polymer, which provides high-Q factors for both passive and active WGMX cavities [15], [16]. Owing to the low cost, easily engineering (such as doping with laser dyes), and good chemical/biological compatibility of polymer materials, FsLDW-fabricated polymer microcavities featuring unidirectional emission would be more desirable for organic optoelectronic integrated devices. Most recently, the fabrication of a passive asymmetric disk-shaped cavity was reported in zirconium/silicon hybrid sol-gel with highly directional coupling [17]. However, to our best knowledge, the demonstration of unidirectional lasing emissions of an active polymer microcavity fabricated directly by FsLDW has not been reported yet. Therefore, in this letter, we report the fabrication of a spiral-shaped polymer micro-disk cavity by FsLDW via two-photon polymerization of dye-doped resin, which produces low-threshold unidirectional laser emission.

II. Fabrication and Characterization

The boundary of the spiral-shaped microcavity was defined in polar coordinates (r, φ) as $r(\varphi) = r_0\{1 + \varepsilon\varphi/(2\pi)\}$ with ε being the deformation parameter and r_0 the radius at $\varphi = 0$. The radius $r(\varphi)$ jumps back to r_0 at $\varphi = 2\pi$, creating a notch. In our experiment, r_0 was set to $r_0 = 15\ \mu$m; ε was set to $\varepsilon = 0.067, 0.100$ and 0.133, which correspond to a notch size of 1.0, 1.5 and 2.0 μm, respectively. The spiral-shaped microcavities were fabricated by two-photon polymerization of dye-doped epoxy-based negative resins (SU-8, MicroChem Corp.) on a microscope cover slide substrate with a thickness of 200 μm. Laser dye of Rhodamine B was used as the gain medium with a concentration of 1.4 wt.%. The dye-doped resin sample was dip-coated on the substrate with a thickness of about 1 mm for subsequent fabrication by FsLDW with detailed procedures described elsewhere [18]. Briefly, femtosecond laser pulses (120 fs, 800nm, 76 MHz) from a Ti:Sapphire oscillator (Tsunami, Spectra-Physics) was focused into the sample by an oil-immersion objective lens (NA = 1.35, 100x) working under an oil condition. The waist of the laser beam filled the entire aperture of the lens, and the calculating spot

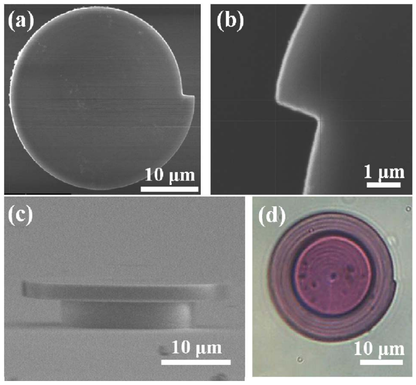

Fig. 1. (a) The top view SEM photograph of the microdisk; (b) the local and amplifying view SEM of the microdisk; (c) the side view SEM photograph of the microdisk; (d) the optical microscope image of the microdisk.

size was ∼361 nm. However, due to the two-photon polymerization in the laser direct writing technique, the fabrication resolution limit should be much smaller than the focused spot size. Based on the design, the laser focus in the sample was scanned by a 3D stage with a resolution of <1 nm. The laser power before the objective lens was fixed to be 7.0 mW, and the exposure time at each focusing spot was 1 ms. The patterned photoresists were post-baked at a temperature of 95 °C for 10 minutes, and then rinsed in acetone for 15 minutes to remove unsolidified resins.

The fabricated spiral-shaped microcavities were investigated by a scanning electron microscope (SEM) (JEOL JSM-7500F). As an example, Fig. 1(a) shows the SEM image of a typical sample with a diameter of ∼30.1 μm, and Fig. 1(b) is a magnified image which gives a notch size of 1.6 μm. Fig. 1(c) shows the side view of the microdisk SEM image, from which the thickness of the fabricated spiral-shaped microresonator is determined to be 2.1 μm. These measured values are in good agreements with the fabrication designs of 30, 1.5 and 2.0 μm respectively, indicating the fabrication precision of FsLDW on the deformed cavity even with sharp edges. Under optical microscope, the spiral microcavity (Fig. 1(d)) shows light pink color for the outer portion, while a relatively deeper pink color in the central portion, which reflects the structure of the disk with a supporting pillar beneath.

The absorption spectrum of Rhodamine B dye-doped resin SU-8 ranges from 450 to 600 nm [18], and thus we chose a frequency-doubled 532 nm Nd:YLF laser (Amberpico-Q-2000, Guoke) with a pulse width of 15 ps and a repetition rate of 50 KHz as the excitation source. An objective lens (NA = 0.25, 10x) was used to focus the pump laser beam onto a single sample at normal incidence. An electric shutter was used to confine each laser exposure time to 20 ms in order to reduce the bleaching effect of dye molecules in the cavity, which strongly depends on the pump laser power. Emission light in the microdisk plane was collected by a lens (convex lens f = 30mm), and the spectra were measured by a spectrometer (SR-303I-A, Andor) equipped with a CCD camera (DV420A-OE, Andor). The fabricated microcavity was mounted on a rotary stage, which was sitting on a 3D stage, so that both the orientation and the position of the microcavity can be controlled precisely.

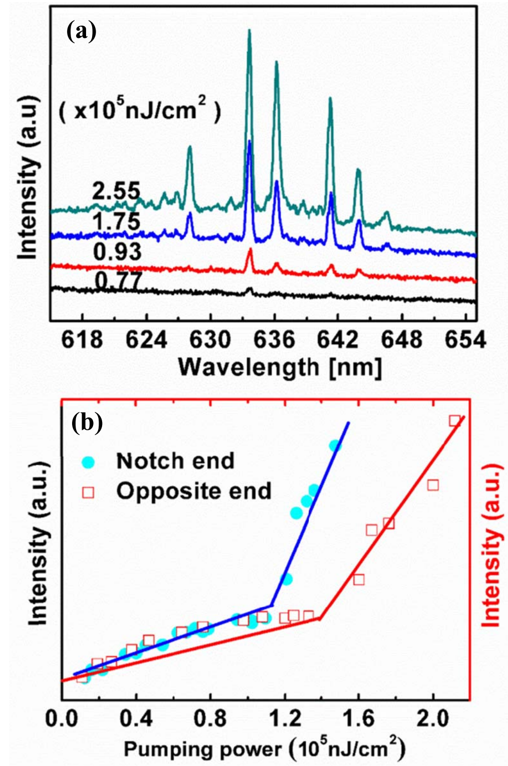

Fig. 2. (a) The measured spectra of the spiral-shaped microcavity with a notch size of 1 μm for five different pumping powers; (b) the measured signal intensities of the output from the notch and the opposite direction versus the power of the pump laser.

III. Results and Discussions

Figure 2(a) shows five emission spectra recorded at room temperature from the spiral-shaped microcavity with the notch size of 1.0 μm, with different pump energy densities of 0.77×10^5, 0.93×10^5, 1.75×10^5, 2.55×10^5 nJ/cm^2, respectively. In this case, a variable neutral filter was used to control the pump laser power. It can be clearly seen in Fig. 2(a) that multiple sharp peaks emerge on the broad emission spectrum of dye in the spectral range of 615-655 nm when the pump power reaches to certain values, which can be ascribed to lasing actions occurring in the microcavity resonator. In order to determine the lasing threshold, as shown in Fig. 2(b), we measured the light intensity of the output at 633.1 nm as a function of the pump laser power from the direction facing the notch section (blue circles), which gives a lasing threshold of $\sim 1.1 \times 10^5$ nJ/cm^2. In addition, based on the measured lasing spectra, the Q factor can be estimated according to the relation of $Q = \nu/\delta\nu$, where $\nu = c/\lambda$ is the frequency of the lasing emission and $\delta\nu$ is the full width at half maximum (FWHM) of the lasing line frequency, respectively. However, for an active microcavity the measured linewidth of the lasing emission may not be accurate for calculating the Q-factor since the emission linewidth can be substantially less than the cold cavity linewidth due to stimulated emission. Therefore, the lasing linewidths obtained with the pump powers at around the lasing threshold were used for estimating the Q-factor, in order to minimize the effect of stimulated emission on the measured line width. In this case,

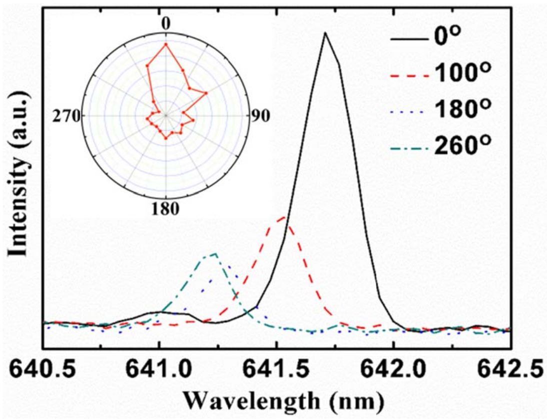

Fig. 3. The emission spectra of the spiral-shaped microdisk cavity laser at room temperature. Inset: the lasing intensity distribution measured with the signal emitted from different angles.

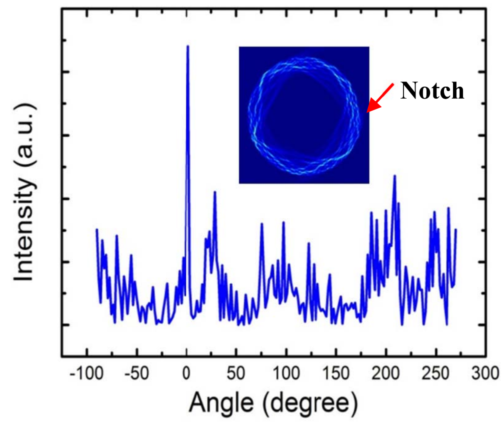

Fig. 4. FDTD simulation results of spectral mode structure. Inset: the intensity distribution of the mode at 631.65 nm.

the linewidth of the laser line at 633.6 nm was measured to be ~0.51 nm. Furthermore, the instrumental broadening was determined to be ~0.3 nm by measuring the spectral line of a single-mode HeNe laser at 632 nm, since the linewidth of the HeNe laser is typically 1GHz or less, which is much narrower than 0.3 nm and thus negligible. That is, the measured profile of the HeNe laser line reflects the instrumental broadening of the spectrometer. By deconvoluting the experimental curve with the instrumental broadening profile, the Q value is estimated to be $\sim 1.6 \times 10^3$. However, it should be pointed out that the Q-factors estimated based on near-threshold laser emission linewidths are an upper limit to the cold cavity Q values. For comparison, we also fabricated a circular WGM microdisk with the diameter of 30 μm. By measuring the lasing spectra of the circular microcavity, the Q-factor value was determined to be $\sim 3.8 \times 10^3$, which is larger than, but of the same order as, that of the spiral-shaped microdisk, indicating the notch effect on the Q factor.

To examine the directional emission property of the spiral-shaped microcavity, we measured the angular dependence of the emission spectrum with a fixed pump laser energy density of 1.3×10^5 nJ/cm^2. In Fig. 3, we show the emission spectra from four different collection angles of 0, 100, 180, 260 degrees, where 0 degree refers to the direction facing the notch section. It can be seen from the measured spectra that for several lasing modes, the lasing intensity from 0 degree is much stronger than those measured from other angles. We further plot the emission intensities in a polar coordinate in the inset of Fig. 3, from which it can be seen that the intensity distribution pattern of the spiral cavity exhibits a good unidirectional lasing emission property with a far field divergence of about 40°.

To get more detailed information about the properties of lasing modes, we carried out two-dimensional Finite-Difference Time-Domain (FDTD) numerical simulation. The spiral structure was set with the parameters of a notch size of 1 μm and a radius of $r_0 = 15$ μm, and the orientation of the structure was set as the same as that shown in Fig. 1(a). TE-polarization was calculated in our case. A broadband pulse was first used to excite all the optical modes in the cavity, and the residue energy spectrum in the cavity long after the pulse was calculated to get the resonant wavelengths of high-Q modes. However, the calculated spectrum is very sensitive to the exact shape of the spiral cavity due to the chaotic nature, and thus the simulation result may not represent the experimental one exactly because the numerical simulation is based on a cavity with an ideal spiral shape. Nonetheless, both of the simulation spectrum and the experimental spectrum show irregular spectral spacing between different peaks.

These calculated modes have typical Q values of about $10^4 \sim 10^3$. Since the lasing modes measured in the experiments are normally high-Q modes which are well overlapped with the gain spectrum, we focus on high-Q modes within certain spectral range. Individual mode was investigated at its particular wavelength, and far-field intensity distribution was calculated. Inset of Fig 4 shows the intensity distribution of a particular high-Q mode at 631.65 nm. As one can see, the mode exhibits a WGM-like distribution with energy mainly localized near the boundary of the cavity. Such mode distribution makes the light propagation inside the cavity satisfying total internal reflection condition when encounters the boundary other than the notch section. However unlike regular WGMs from circular cavities, the high-Q modes of the spiral cavity show irregular patterns that usually are different from each other, indicating the chaotic nature of spiral cavity. As a consequence, the far-field intensity distributions can be different. This is verified by our experimental observations. As shown in Fig. 3, the intensity variations at different angles behave differently for individual modes. Furthermore, all the high-Q modes we studied propagate in clockwise (cw) direction. This is understandable, since the counter-clockwise (ccw) propagating modes will likely hit the notch section and leak out easily, while the cw modes may avoid such leakage and maintaining high Q factors. However, some of these high-Q modes can be coupled to low-Q ccw modes via weak scattering at the notch area, due to the discontinuity of the boundary [10]. In this way, part of the energy will eventually escape from the cavity from the notch section. Meanwhile, part of the cw propagating evanescent wave can be reflected at the spiral notch [19], [20]. Thus high-Q modes usually exhibit relative strong far-field intensity distribution at 0 degree angle. Due to the chaotic nature of the spiral cavity, the resonant modes are very sensitive to the exact boundary shape. Though our numerical simulation results may not reproduce the exact lasing modes because of certain fabrication imperfection, the far-field intensity distribution shown in Fig. 4 for the calculated high-Q mode reflects the directional output observed in the experiments.

Furthermore, the existing of the circular pedestal under the spiral cavity will also impact the angular emission characteristics of the microlaser. Due to the discontinuity of refractive index, modes that extend greatly into the pedestal area will encounter strong scattering. As a result, these modes may not have a low threshold to achieve lasing. This effect makes the cavity behaviors similarly as selective area excitation, which will improve the directionality of laser output [21].

Other than angular dependence of the laser output intensity, one interesting phenomenon we have observed in our experiment is that the lasing threshold may be different when measuring from different angles. For the same lasing mode at 633.1 nm shown in Fig. 2, we measured the lasing threshold from the emission light collected at 180 degree, as shown in Fig. 2b (red rectangle), from which a lasing threshold of 1.5×10^5 nJ/cm^2 is obtained. This value is much larger than that obtained for the emission at 0 degree. Similar behavior of such position-dependent lasing thresholds has also been reported previously [22]. The underlying mechanism for this phenomenon is still not totally clear. However, our observation indicates that the far field measurement from certain angle may not reveal the exact information of a lasing mode in such cavity. As shown in Fig. 3, as we check the spectra more carefully, we find the peaks of a lasing mode shift with the measurement angles. This might be due to the chromatic dispersion when lights hit the boundary with different incident angle and escape from the cavity via tunneling, which induces the observed difference of lasing thresholds measured at different angles.

IV. Conclusions

In summary, we have designed and fabricated a spiral-shaped polymer microcavity by FsLDW via two-photon polymerization. The spiral microcavity shows a good capability of producing high-Q factor and low threshold lasing with highly unidirectional emission. FDTD numerical simulation results reveal that the lasing modes may correspond to cw propagating high-Q modes. Due to the scattering at the notch section, such high-Q modes exhibit strong far field intensity distribution at 0 degree. Because the precise 3D fabrication capability of FsLDW and the good compatibility of polymer to other materials, the realization of the creation of active unidirectional lasers in polymer in this letter provides an important step towards the functional integrated organic optoelectronic devices.

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