Stretchable organic printed microlaser

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Abstract. Miniaturized lasers with wavelength variability are of crucial importance for various ultracompact photonic devices. Here, we report fine tunable microlasers built by mechanically stretching the flexible microcavities and elastic substrate. The flexible microcavities fabricated by precisely printing dye-doped polymer solution on elastic substrate, enabled whispering-gallery-mode laser under optical pumping. Furthermore, on account of the outstanding structural flexibility of organic materials, reversible wavelength-tunable laser can be realized through adjusting the shape of microcavity \textit{via} mechanical stretching the elastic substrate. The results will provide guidance for the rational design of photonic devices with novel performances based on the characteristic of organic flexible materials.

1. Introduction

Miniaturized lasers attracting much interest for delivering intense coherent light signals at micro/nanoscale, have revolutionized many advanced applications, including optical communications, high-throughput sensing data storage, and laser display[1-3]. The ever-increasing demands for bandwidth and information density in the fields of communications and computing call for on-chip coherent optical sources capable of achieving wavelength variability[4-6]. In principle, the wavelength-tunable micro/nanolasers can be realized by tuning by the size, shape, temperature or electric field[7-10]. However, the poor responsive property of traditional materials leads to limited wavelength tuning range, slow response time and low sensitivity for their practical implementations.

Organic materials possess excellent responsiveness to external stimuli such as mechanical signals benefiting from various weak interactions between different molecules[11-13]. With outstanding processability and wide optical window, organic materials are ideal for the fabrication of microscale resonators by many methods, such as photolithography, electron beam lithography, laser direct writing and inkjet printing[14-16]. All these attractive characteristics make organic materials promising candidates to construct highly responsive microcavities, which would facilitate in-situ modulation of the resonant wavelengths through stimuli[17]. Therefore, developing a new method of fabricating organic flexible microlaser would promote the development of organic microlaser.

Herein, we demonstrate wavelength-tunable microlaser in organic spherical cap-shaped microcavities array by mechanical stretching. The spherical cap-shaped microstructures were fabricated by precisely depositing dye-doped polymer solution on elastic substrate \textit{via} an ultrasonic vibration-assisted inkjet printing. Each of these microspherical cap with smooth circular boundary structure can serve as an excellent high-quality whispering-gallery-mode (WGM) cavity to support laser oscillations under optical pumping. On the basis of the excellent flexibility of the organic
microstructures and substrate, we were able to reversibly tune the wavelength of the laser through the adjustment of microdisk cavity shape via mechanical stretching. The results afford an approach for simultaneous modulation of laser array output and enlighten the rational design of miniaturized photonic materials and devices with desired performances.

2. Materials and methods

2.1 Chemicals and reagents
Araldite 506 epoxy resin and polystyrene (PS), which were purchased from J&K Scientific Ltd. (Beijing, China) and Sigma-Aldrich (Shanghai, China) respectively, were selected as the matrix material to create the high-quality (Q) resonators due to their outstanding flexibility. Polydimethylsiloxane (PDMS, Sylgard 184 Silicon Elastomer) purchased from Dow Corning was selected as the elastic substrate because of its transparency, low refractive index, and good mechanical properties. Rhodamine B (RhB, 99%) purchased from TCI (Shanghai, China), which exhibits photoluminescence (PL) at red waveband, was selected as gain media for lasing. Dichloromethane was commercially available products. The materials were used as received without further treatment.

2.2 Instrument and characterization
Bright-field and fluorescence microscopy images were taken by inverted fluorescence microscope (Nikon, Ti-U) under the excitation of a mercury lamp (330–380 nm). The morphology of the as-prepared microspherical caps were examined by atomic force microscopy (AFM, NTEGRA Solaris, NT-MDT). The optically pumped lasing measurements for spherical caps were conducted on a custom micro-PL system. The excitation pulses (400 nm) were generated from an optical parametric amplifier (Light Conversion TOPAS) pumped by a regenerative amplifier (Spectra Physics, 800 nm, 150 fs, 1 kHz), which was in turn seeded by a mode-locked Ti: sapphire laser (Mai Tai, Spectra Physics, 800 nm, 150 fs, 80MHz). The excitation laser was filtered with a 450 nm short-pass filter and then focused down to a spot through an objective lens (Nikon CFLU Plan, ×20, N.A. = 0.5) as a nearly uniform pump source. The emissions from the spherical caps were collected by the same objective with a back-scattering configuration and analyzed by the spectrometer after removing the excitation beam with a 450-nm long-pass filter.

2.3 Fabrication of spherical cap shaped microlasers array
The PDMS was prepared by mixing a 10:1 mass ratio of liquid silicon base and a curing agent. Then the PDMS films were prepared by heating the PDMS solution. The spherical caps were prepared by depositing RhB-doped polymer solution on the PDMS substrates using a GIX™ Microplotter™ II from Sonoplot INC. The ink solution was prepared by dissolving 200 mg of PS in a mixed solution of 1 mL of dichloromethane and 10 mL of Araldite 506 epoxy resin. The printing process included imbibing ink solution with a glass needle via capillary action and spraying a drop assisted by ultrasonic vibration. Due to the hydrophobic effect between PDMS and polymer solution, the droplet self-assembled into a spherical cap-shaped structure. After drying in air for one day, solid-state spherical cap microstructures were obtained. The spherical cap laser arrays were fabricated by sequentially depositing luminescent inks at specific positions onto substrates according to predesigned digital patterns.

3. Results and discussion
Organic microlaser arrays were prepared by selectively printing RhB-doped polymer solution droplets at specific positions on substrates according to the predesigned digital patterns (Figure 1A). The fabrication process was spraying of a drop of organic ink solution assisted by ultrasonic vibration onto a PDMS substrate (Figure 1B). Then the hydrophobic effect between PDMS and polymer solution would drive each droplet to form the spherical cap morphology. As shown in Figure 1C, the ink droplet array dispersed on the PDMS substrate exhibit the geometry of spherical cap, and become solid after the evaporation of solvent. The atomic force microscopy (AFM) images presented in Figure 1D-F also
verify that the as-prepared microstructure has well-defined spherical cap morphology with smooth surfaces. This morphology can minimize the undesirable optical scattering and confine the guided emission efficiently in the microstructure, which is beneficial for achieving strong WGM microcavity effects[18]. Under UV light irradiation, the spherical caps emit bright yellow fluorescence (Figure 1G), confirming uniform doping of the dye in the polymer matrix. The size of spherical cap can be controlled by changing the amount of ink solution released.

Figure 1. (A) Schematic illustration of the fabrication of organic microlaser arrays by ultrasonic vibration-assisted inkjet printing. (B) Image of large-area ordered printed microstructures on PDMS substrate. The scale bar is 5 mm. (C) Microscopy image of the printed microstructure array showing uniform size and a well-defined pattern. The scale bars are 50 μm. (D-F) AFM images of a typical printed spherical cap. (D) 3D-AFM image. (E) 2D-AFM image, the scale bar is 5 μm. (F) Corresponding cross-sectional profile of the printed microstructure. (G) Fluorescence microscopy image of the printed microstructure array under UV light radiation (330–380 nm). The scale bar is 20 μm.
Figure 2. (A) Simulated electric field distribution of resonant cavity modes in a typical spherical cap shaped microstructure. (B) The microscopy image of the spherical caps under optical pumping. The scale bar is 5 μm. (C) PL spectra of an individual spherical cap under different pump pulse energies. (D) Plots of the photoluminescence peak intensity at 593 nm vs pump fluence, showing the lasing thresholds of 55.9 J/cm².

Figure 2A shows the electric field intensity distributions in the spherical cap obtained by using the finite-difference time-domain method. The energy of the photons is well-confined within the spherical cap, and the radial scattering into surrounding environment is quite limited. The optical mode profiles along the edge of the spherical cap clearly show efficient light guiding, indicating a typical WGM-type resonance. When the RhB-doped spherical cap was excited locally with a focused pulsed 400 nm laser beam in a home-built far-field micro-PL system, we could observe a brighter ring shape pattern located at the outer boundary of the spherical cap (Figure 2B). This result indicated total internal reflection of the emitted light along the edge of the spherical cap, which is a typical characteristic of WGM-type microresonator.

Figure 2C summarizes the PL spectra of an isolated spherical cap as a function of pump fluence. At low pump energy with fluence <54.8 J/cm², the PL spectra are dominated by broad spontaneous emissions. When the pump fluence exceeds a threshold, strong laser emission develops as a set of sharp peaks. The full width at half-maximum (FWHM) dramatically decreases down to ~0.4 nm above the threshold. The corresponding power dependence of the PL intensity (Figure 2D) showed a nonlinear behavior at the threshold of ~55.9 J/cm², indicating the transition from spontaneous emission via amplified spontaneous emission to full lasing oscillation.
Figure 3. (A) PL spectra of the spherical caps with different diameters. All scale bars are 20 μm. (B) Relationship between λ2/Δλ and the diameter of the spherical caps D. The red line is a fit to the function λ2/Δλ = nπD, which verifies the WGM cavity property of the printed spherical caps. (C) Plot of the experimental Q factor vs spherical cap diameter.

The lasing characteristics of these WGM microcavities, including the mode spacing and Q factor, can be well-tuned by altering the sizes of the spherical caps. Figure 3A presents PL images and lasing spectra of three distinct spherical caps with different sizes. Bright emission was observed along their ring-shaped boundaries, further indicating their typical WGM resonance. All spectra show a series of peaks with different values of mode spacing (Δλ). According to the WGM theory, the mode spacing Δλ and the diameter D should satisfy the equation λ2/Δλ = nπD, where λ is the wavelength of the guided light, and n is the group refractive index, respectively. On the basis of the experimentally measured relationship between λ2/Δλ and D, n = 1.57 was obtained. It is matching with the intrinsic refractive index of the PS polymer and Araldite 506 epoxy resin[19]. In addition, the lasing mode numbers decrease with reducing of spherical cap diameter, indicating that a single-mode laser can be obtainable by shortening of the cavity diameter. All these results indicate that the optical WGM modes are tightly confined in the spherical caps with negligible optical leakage to the substrate, which is beneficial for resonators to achieve high Q factors[20]. The measured Q factors of the cavities were on the order of 10^3, which is quite high for organic resonators. And the Q factor increased with the increasing of diameter.
Figure 4. (A) The design principle of tunable WGM lasing through mechanical stretching. (B) The top down optical images of a typical spherical cap before and after stretching, respectively. (C) Lasing spectra from a RhB-doped spherical cap with different stretching ratio ($\Delta D\%$). (D) Lasing wavelength shift of spherical cap as function of stretching ratio.

The spherical caps were printed on the PDMS substrate with a good elastic property. When the PDMS substrate was mechanically stretched from both sides (Figure 4A), the microstructures on PDMS would deform from sphere to ellipsoid (Figures 4B). As a result, the optical cavity of the spherical cap was changed under the deformation, which tuned the resonant modes. Figure 4C shows the lasing spectra of a typical spherical cap a function of stretching ratio ($\Delta D\%$). It can be seen that the lasing envelope exhibited a red-shift behavior with increasing of $\Delta D\%$, which is due to a slight increase of the optical microcavity. In addition, for the investigated range, the shift values of lasing wavelength showed a linear relationship with $\Delta D\%$ and depend on spherical cap’s size (Figure 4D).

4. Conclusion
In summary, we report that fine wavelength-tunable microlasers can be realized based on organic flexible microcavity and substrate. The flexible microcavities were prepared by printing ink solution on PDMS substrate, which can serve as a high-quality WGM cavity to support laser oscillations under optical pumping. With the outstanding mechanical flexibility of organic microstructure, the printed microstructures were proven to be an effective way to wavelength-tunable laser. These results provide a good inspiration for the rational design of high-performance, easy-to-fabricate, tunable laser source.

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