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Wavelength-tunable organic semiconductor lasers based on elastic distributed feedback gratings

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Abstract: Wavelength-tunable organic semiconductor lasers based on mechanically stretchable polydimethylsiloxane (PDMS) gratings were developed. The intrinsic stretchability of PDMS was explored to modulate the period of the distributed feedback gratings for fine tuning the lasing wavelength. Notably, elastic lasers based on three typical light-emitting molecules show comparable lasing threshold values analogous to rigid devices and a continuous wavelength tunability of about 10 nm by mechanical stretching. In addition, the stretchability provides a simple solution for dynamically tuning the lasing wavelength in a spectral range that is challenging to achieve for inorganic counterparts. Our work has provided a simple and efficient method of fabricating tunable organic lasers that depend on stretchable distributed feedback gratings, demonstrating a significant step in the advancement of flexible organic optoelectronic devices.

Key words: stretchable electronics; organic semiconductor lasers; elastic lasers; distributed feedback (DFB) gratings; wavelength tunability

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1. Introduction

With good capability of producing intense coherent light at (sub)wavelength scale, organic lasers can serve as important tools in various fields including chemical sensing and optical communication[1–5]. The growing demands on optoelectronic devices with high level of integration call for organic lasers with wavelength switching[6–8]. Efforts have been concentrated on the design and development of various tunable organic lasers because many photonic applications can be enabled or enhanced by wavelength tunability[9–13]. Current methods of modulating the wavelength of organic lasers mainly involve the reconfigurable Förster resonance energy transfer (FRET) process[14] and the photoisomerization-activated intramolecular charge-transfer (ICT) process[15]. On one hand, the output wavelength of FRET lasing can be tailored by adjusting the FRET efficiency, which is associated with the stoichiometric ratio of donor and acceptor. On the other hand, typical photoisomerizable molecules can be employed to achieve a wide gain region tuning without compromising the lasing characteristics via using its own photoisomerization to modulate the ICT state level. In addition, photoisomerization will induce a reversible refractive index change of the optical cavity where the photoisomerizable dyes locate and in turn adjust the resonant modes continuously[16].

During the past few decades, great progress has been achieved in developing optically pumped organic semiconductor lasers in both solutions and solid states for different molecules, and in resonators from simple waveguides to complex geometries including microrings, microcavities and DFB structures[17–22]. Among these, DFB structures are efficient to achieve organic lasing owing to their long gain path, great optical mode confinement and high reflectivity[23]. Generally speaking, the device consists of a thin film on top of a corrugated fused silica substrate. DFB lasers can demonstrate low thresholds, particularly for first-order feedback where output coupling losses are low. In this case, the Bragg condition is demonstrated as Eq. (1)[1]:

$$m\lambda_{\mathrm{Bragg}} = 2n_{\mathrm{eff}}\Lambda. \qquad (1)$$

Here, m is the diffraction order, λ is the vacuum wavelength, n_{eff} is the effective index of the waveguide and Λ is the periodicity of the corrugation. Consequently, the laser wavelength λ strongly relies on the effective index or the grating period of the DFB structure. Limited alteration of the effective index can be obtained by adjusting the thickness of gain media. The wide wavelength modulation of organic lasers can be achieved by adjusting the structural parameters of DFB resonators. However, the kinds of grating that were previously reported are limited. In addition, most gratings are made from rigid patterns, which makes the structural parameters hard to modulate. The characteristics can suppress their ability of light manipulation and hinders the utilizations in various fields. In addition, common grating fabrication approaches such as electron-beam[24] and nanoimprint lithography[25], are complex and expensive. Consequently, it is very promising that elastic DFB lasers can be optically tuned on the surface just by mechanical stretching[26–33]. Under this circumstance,

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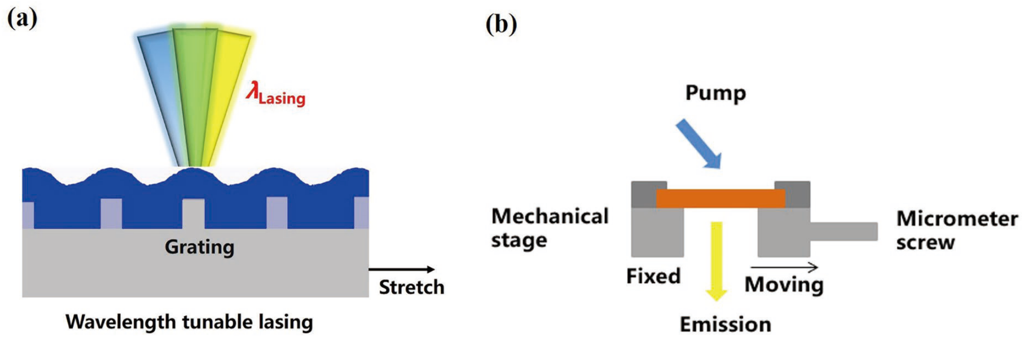

Fig. 1. (Color online) (a) Device structure of wavelength-tunable organic semiconductor lasers. (b) Graphical illustration of the stretching setup.

the wavelength tuning was achieved by means of modulating the periodicity with mechanical strain[27, 28]. For instance, Görrn[31] developed a novel method of fabricating DFB gratings on the surface of elastomeric PDMS by self-organization. The emission wavelength can be continuously modulated from 633.6 to 638.3 nm (4.7 nm) by uniaxially stretching DFB lasers. However, the tuning range was small and the process involved many complicated steps.

In this contribution, efficient and stable lasing wavelength tuning was achieved in a low-cost and elastic DFB laser. The intrinsic stretchability of PDMS was explored to change the period of the DFB gratings to adjust the lasing wavelength. The individual elastic laser was continuously tuned about 10 nm with a stretch of 3%, which was higher than 4.7 nm achieved by Görrn[31] and 4 nm obtained by Klinkhammer[34] through employing a switchable liquid crystal cladding for organic lasers. More importantly, the threshold for each device was just increased by 5 times with a stretch range from 0% to 3%. According to our results, these devices show great advantages of low cost, simple preparation processes and excellent compatibility. Thus, a simple and straightforward method has been developed to modulate the lasing wavelength.

2. Experiment

T-m and SPL(2)-1 were synthesized by our group. F8BT was bought from Xi'an Polymer Light Technology Corporation. Elastic gratings were fabricated by replicating a silicon master grating fabricated by electron beam lithography. These rectangular gratings possessed three periods between 280 and 360 nm and a depth of 60 nm. PDMS is selected because of its superior optical transparency and elasticity. The elastic gratings were made by casting and curing PDMS on the top of a silicon master. The feedback structure was determined by electron beam lithography master with different periods. After coating the surface with PDMS, the samples were heated at 50 °C for 2 h for curing, and the grating was subsequently separated from the substrate by peeling off carefully. Finally, a self-standing elastomeric grating was obtained. Three typical light-emitting materials including T-m, SpL(2)-1 and F8BT were dissolved in toluene with the concentrations of 20 mg/mL. The solutions were stirred and heated at 50 °C for 2 h. The film thicknesses were modulated by changing spin-coating speeds. Three different laser gain materials were spin-coated on the fabricated gratings to form lasers, respectively, as shown in Fig. 1(a). The organic lasers were optically pumped with an optical parametric oscillator (OPO), which was excited by pulsed Nd^{3+}:YAG nanosecond laser (Continuum Ltd, Surelite, frequency 10 Hz, pulse width 5 ns) at room temperature. The output beam from OPO was shaped by an adjustable circular slot and then focused with a cylindrical focal lens (focus length, 10 cm). The lasing output intensity from organic lasers detected by using a spectrometer upon various pump energies was recorded to determine their lasing thresholds. Consequently, the thresholds of organic lasers were calculated by the change in the slope of the output intensity with the increment of the excitation fluence.

As shown in Fig. 1(b), the elastic lasers were put in the stretching setup. The laser was uniformly stretched by carefully rotating the micrometer. The stretching setup consisted of two three-way translation stages and the moving directions were adjusted by rotating the vernier adjustment knob. The largest stretch distance can approach 40 nm with the smallest stretch step length of 0.01 mm.

3. Results and discussion

To fabricate elastic gratings, PDMS and the curing agent were mixed at 15 : 1 volume ratio and spin-coated on top of the silicon master grating. Subsequently, the mixture was subjected to thermal treatments to solidify, and peeled off carefully, as depicted in Fig. 2(a). During the solidification of PDMS, the parts in contact with the formwork organized themselves into a highly ordered grating, and its depth and period were determined by the silicon master grating. According to atomic force microscopy (AFM) images in Fig. 2(b), DFB structures of three PDMS gratings possessed a period of about 280, 320, and 360 nm, respectively, with the modulation depth of approximately 60 nm. Based on the results, such a grating made from PDMS exhibited higher precision and fewer structural defects compared to those previously reported.

The gain materials used in this experiment are three typical light-emitting molecules: T-m, SPL(2)-1 and F8BT. The chemical structures are shown in Figs. 3(a)–3(c). In this system, organic media showed two distinguished merits: the first is the mechanical flexibility that can maintain the lasing properties in the range of a certain stretch; the second is the wide emission spectra that can guarantee the spectral tuning in a large scope. Fig. 3(d) demonstrates the absorption, photoluminescence (PL), and amplified spontaneous emission (ASE) spectra of three molecules in the film states. The ASE spectra of films shown as the filled curves were recorded when exciting the planar waveguide by using Nd^{3+}:YAG laser as the pumping source with incident energy well above the ASE threshold. The ASE peaks are located at 426, 483 and 569 nm,

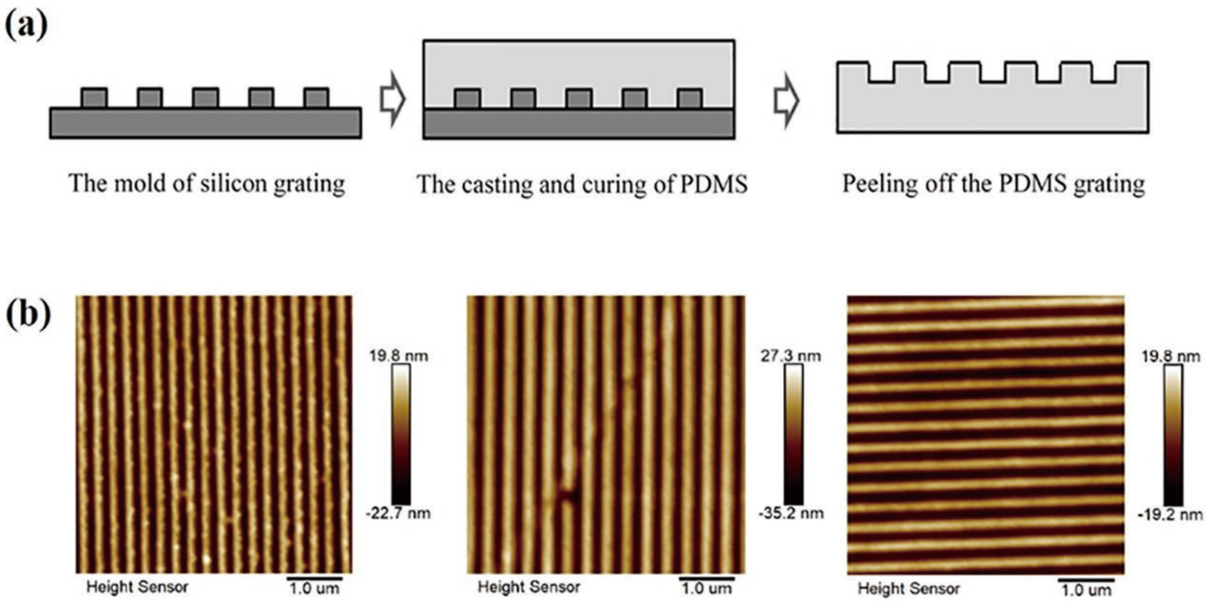

Fig. 2. (Color online) (a) Fabrication process. (b) AFM images of the PDMS gratings. The period is 280, 320, and 360 nm from left-hand to right-hand, respectively, and the groove depth are both ~60 nm.

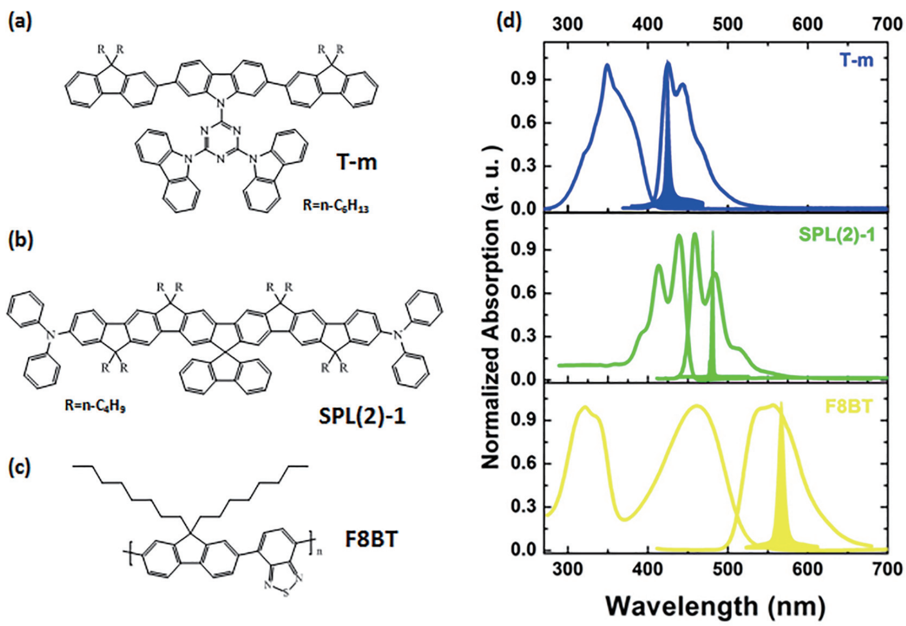

Fig. 3. (Color online) Chemical structures of (a) T-m, (b) SPL(2)-1, (c) F8BT from top to bottom. (d) Absorption, PL and ASE spectra for a thin film of T-m (blue), SPL(2)-1 (green), F8BT (yellow).

respectively, which corresponded to the blue, green and yellow wavelengths.

The organic laser was prepared by depositing organic semiconductors on top of DFB gratings. These gain medium supported lasing between blue and yellow. A 120-nm-thick layer was spin-coated from a toluene solution on the PDMS gratings. As shown in Fig. 4(a), the lasing characteristics of the devices were similar to rigid DFB lasers based on quartz structured substrates. When T-m was optically pumped at a wavelength of 355 nm, the lasing wavelength was observed at 426 nm with a grating period of 280 nm. Upon optical pumping at the wavelength of 420 nm, SpL(2)-1 exhibited the lasing wavelength of 476 nm with a grating period of 320 nm. As for F8BT optically pumped at the wavelength of 450 nm, the lasing wavelength was recorded at 574 nm with a grating period of 360 nm. The chromaticities of laser outputs were calculated from the three molecules of different gratings, which were marked on the CIE1931 chromaticity diagram (Fig. 4(b)). Obviously, the calculated colors covered the ultra-wide range from blue to yellow. Experimentally, a minimal lasing threshold of 150, 69 and 123 nJ were recorded for T-m, SpL(2)-1 and F8BT films with a grating period of 280, 320 and 360 nm, respectively, which indicated comparatively low lasing threshold in the range of several nJ orders. As for organic lasers, laser power can be interpreted as the output intensity, which can be observed from Figs. 5(a)–5(c). The laser power of devices based on T-m, SpL(2)-1 and F8BT films corresponding to its minimal lasing threshold were 120, 95 and 83 a.u., respectively. The lasing output intensity was increased with the increment of the pump intensity, resulting in a narrowing of the emission spectra. During the process, a change in the slope of the output intensity was recorded for

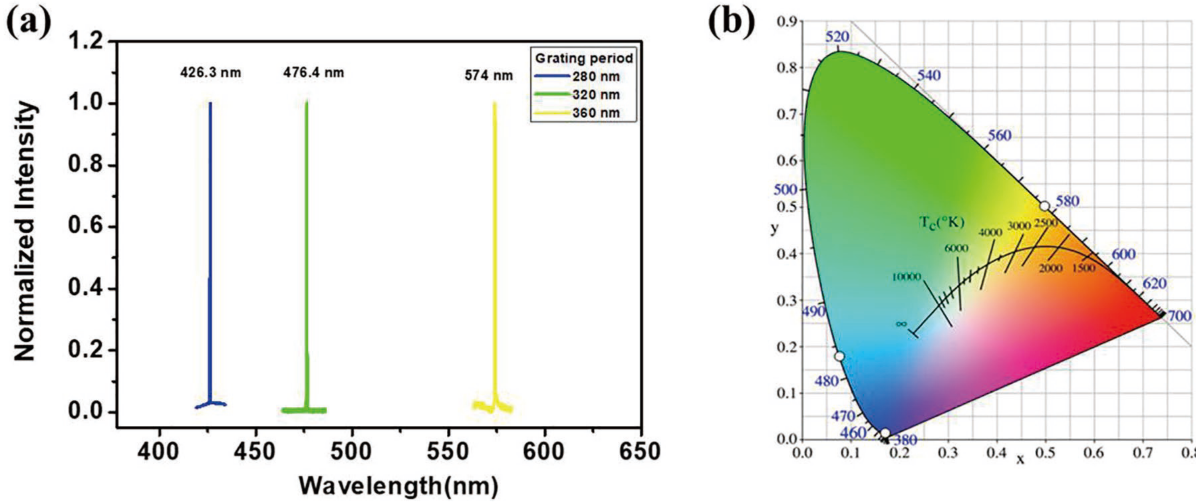

Fig. 4. (Color online) Lasing characteristics of elastic organic DFB lasers. (a) Spectra of elastic DFB lasers based on active films of T-m, SPL(2)-1 and F8BT, respectively. (b) Chromaticity coordinates of the elastic lasers in (a) on the CIE 1931 chromaticity diagram.

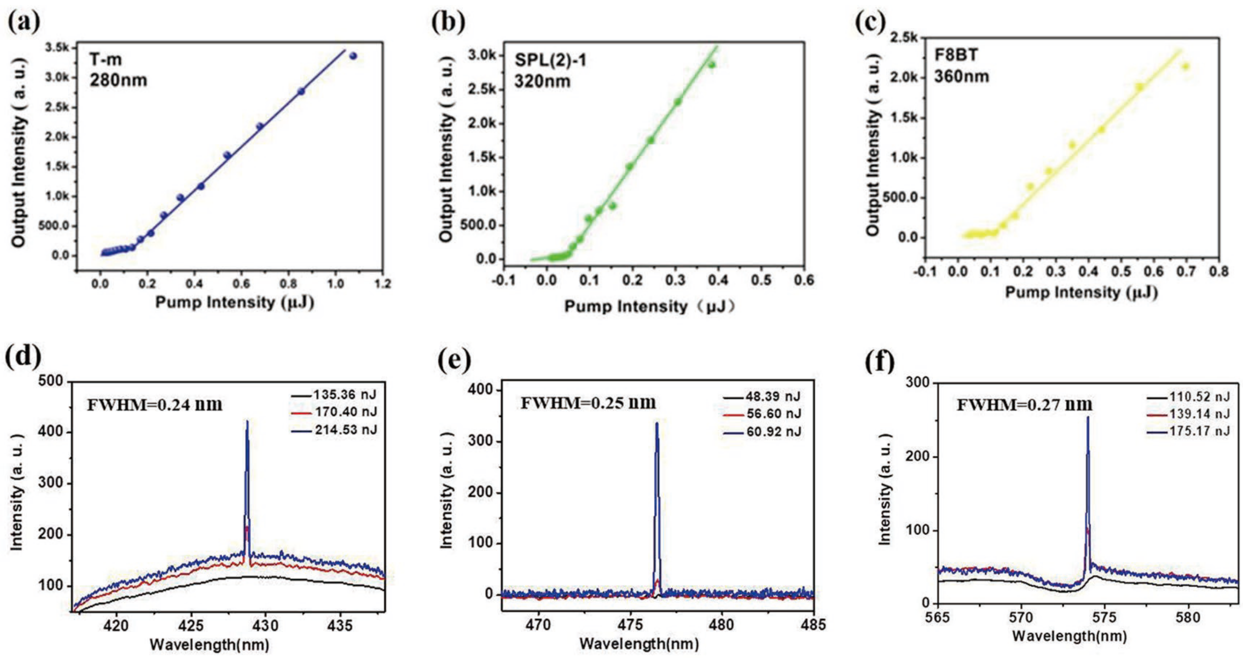

Fig. 5. (Color online) (a–c) Output intensity as a function of the pump fluence and (d–f) emission spectra with pump pulse energy slightly above the lasing threshold for the three devices.

each device, which denotes the thresholds of the DFB lasers. The slope of this curve is often called the laser efficiency. In terms of organic lasers, the laser threshold is usually considered as an important parameter to compare the performance of organic lasers. Our elastic lasers based on F8BT demonstrated the lasing threshold of 123 nJ, which was comparable with that of previously reported devices (63 nJ)[35]. Although the threshold was relatively higher, our lasers had attractive elastic characteristics and this result was acceptable to organic lasers with both good elastic and optical properties. Meanwhile, emission spectra for the corresponding three devices were obtained with the pump pulse energy slightly above the lasing threshold (Figs. 5(d)–5(f)). According to these results, PDMS gratings in our work possess good feedback effects although the transfer method is facile. In other words, the lasing performance of devices based on our method has been excellent compared with traditional approaches such as e-beam lithography on oxidized silica wafers or the direct fabrication of gratings in a photo-resist by laser interference lithography.

A significant property of elastic lasers is the possibility to modulate the λ_{DFB}. Owing to the elastic characteristics of lasers, the grating periods can be increased with an increment of the stretch when the stress was applied to laser devices. Elastic lasers were mounted in an apparatus by uniaxial stretching. Device operation was conducted by focusing the pump beam on a certain section of the laser.

An elastic laser based on active films of uniform thickness (~150 nm) and variable periods of different gratings (Λ = 280, 320, 360 nm) provided a wide tunability range of ~160 nm (emission: 426–586 nm). Each laser was mechanically stretched by up to 3%. Figs. 6(a)–6(c) demonstrate the influence of the strain on lasing wavelength. In this case, the lasing spectra were observed from 426 to 436 nm for T-m with a

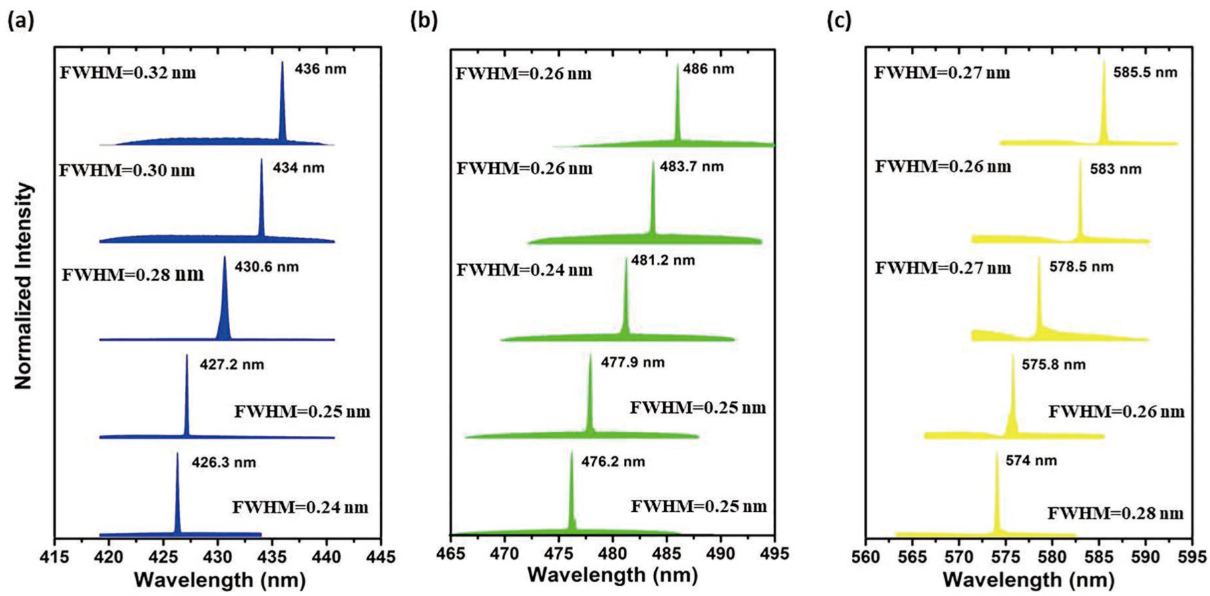

Fig. 6. (Color online) Lasing spectra measured by straining the elastic DFB lasers based on (a) T-m, (b) SPL(2)-1, and (c) F8BT.

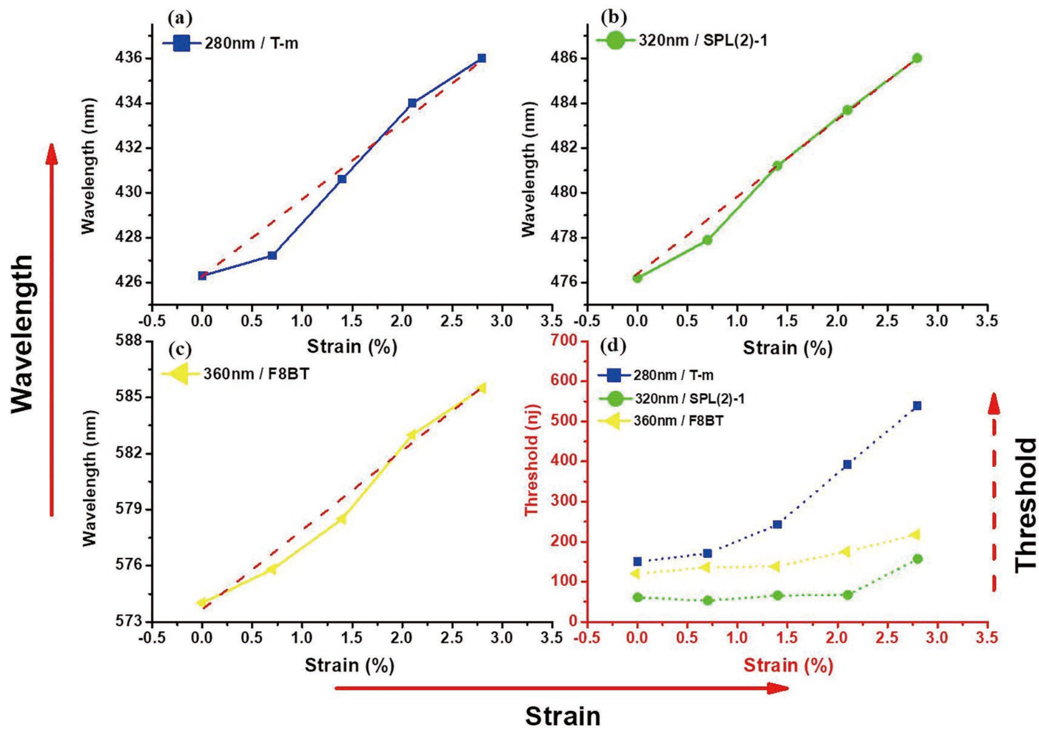

Fig. 7. (Color online) The dependence of lasing wavelength of the elastic DFB lasers based on (a) T-m, (b) SPL(2)-1, and (c) F8BT. (d) The dependence of threshold variations of these elastic DFB lasers on the applied strain.

grating period of 280 nm. Accordingly, the lasing spectra of SPL(2)-1 were recorded from 476 to 486 nm with a grating period of 320 nm. F8BT possessed the lasing spectra from 574 to 585 nm with a grating period of 360 nm. The individual elastic laser was continuously tuned about 10 nm with the stretch of 3%. The corresponding full-width at half-maximum (FWHM) values were provided, which were not greatly increased during the stretch. More importantly, the threshold was just increased 5 times if the laser was stretched from 0% to 3%. The device performance will be degraded once above a certain range of the stretch. For the purpose of providing a direct insight into the wavelength tunability of the elastic lasers, the dependence of the wavelength and threshold on the applied stretch was demonstrated in Fig. 7. According to the plot, the emission of organic lasers was gradually red-shifted with the increase of the strain. Meanwhile, the lasing threshold was slightly increased with the increment of the strain, which may result from the large optical loss caused by the deteriorated gain media. Furthermore, the CIE coordinates were marked in Fig. 8, demonstrating the change of the lasing color.

The mechanical stability of the device is closely associated with not only the elastic PDMS grating but also the organic gain media. Since T-m and SpL(2)-1 are synthesized by our group and F8BT is a commercial material, we choose F8BT to investigate the mechanical stability in details. According to

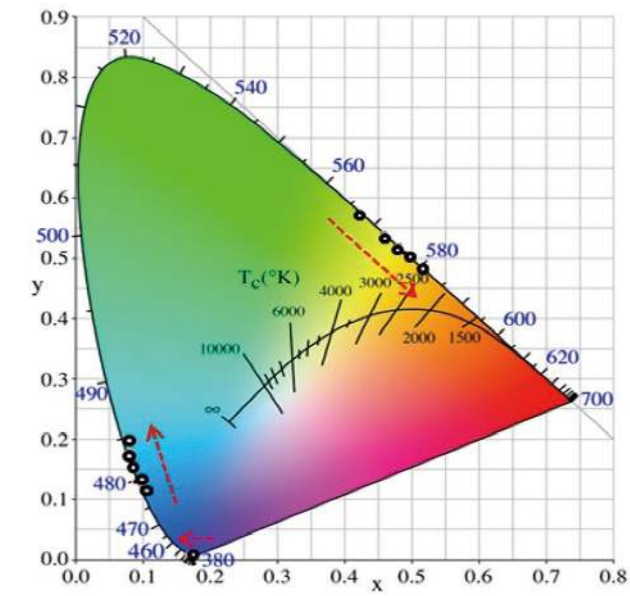

Fig. 8. (Color online) Chromaticity coordinates of the varied lasing wavelength for T-m, SPL(2)-1, and F8BT on the CIE 1931 chromaticity diagram (the red arrows indicate the direction of spectral shift).

Fig. 9, the device's performance can be largely maintained after three stretches. After the first stretch, the organic laser based on F8BT retained its original state and possessed the threshold of 114 nJ with the lasing wavelength at 574.82 nm (Figs. 9(a) and 9(b)). After the second stretch, the device based on F8BT showed the threshold of 135 nJ with the lasing wavelength at 575.02 nm, whose threshold was a little increased compared with that after the first stretch (Figs. 9(c) and 9(d)). In the case of the third stretch, the threshold of 136 nJ was observed and the lasing wavelength was recorded at 574.98 nm (Figs. 9(e) and 9(f)). Meanwhile, the corresponding full-width at half-maximum (FWHM) values were not greatly altered after three stretches. According to these results, three stretches did not cause a great deterioration of the performance of the device, such as the lasing threshold and the lasing wavelength. Although the lasing threshold was relatively increased, it can be acceptable under this circumstance.

Tunable lasers are of paramount significance in modern telecom systems because they can be employed to populate various channel slots. As indicated, the wavelength-tunable characteristics have enabled organic lasers with great importance and wide applications. For instance, by using a tunable laser it is possible to generate 3D topographical images with chemical specificity, which can be utilized in medical diagnostics such as breadth analysis and non-invasive glucose monitoring. They can also be employed in environmental sensing, pollution monitoring and industrial process monitoring which requires a specific wavelength. Meanwhile, other tunable lasers with integrated properties have also been fabricated, including electro-absorption modulators, semiconductor optical amplifiers and electro-absorption modulator integrated lasers.

In addition to the elastic properties, the PDMS resonator in our DFB lasers has unique characteristics, which can be employed repeatedly. Under normal conditions, the PDMS resonator can only be used once since the gain medium is deposited onto it and it is difficult to be separated from the PDMS resonator. In our work, we have optimized the parameters to fabricate the PDMS resonator via a transfer method. It is worth mentioning that the gain medium can be easily peeled from the PDMS resonator. Consequently, the PDMS resonator can be used repeatedly and the performance of the corresponding lasers is still comparable to that based on the fresh PDMS resonator first used. Nowadays, inspired by the needs of people, it is desirable to develop organic lasers that are compatible with mechanically-flexible platforms. In our work, soft DFB lasers can be optically modulated on the surface just by the mechanical stretch. This feature would launch a wide range of novel applications. Besides applications in biology, pharmaceuticals, explosive detection and chemistry, other utilizations involving conformable sensitive optical skins for monitoring the structural health of civil infrastructure are expected[36–38]. In brief, our study has contributed to the development of flexible organic lasers.

4. Conclusion

In summary, organic lasers with tunable wavelength based on stretchable DFB gratings were manufactured. To prepare the gratings, liquid PDMS was solidified by thermal annealing and then peeled off from the master grating. The deposition of organic semiconductors from solution exemplified a laser structure of facile manufacturing. In the case of mechanical stretching, a tuning interval of around 10 nm was achieved from three different devices from blue to green and yellow dyes, respectively. It is worth mentioning that the individual elastic laser shows a comparable lasing threshold analogous to rigid devices. In brief, our work has developed a practical transfer printing method for achieving wavelength tunable organic laser devices with outstanding level of precision and few structural defects. High flexibility of PDMS gratings and the low cost of fabrication have provided a prominent method of developing superior lasers that take full advantage of the characteristics of organic semiconductors.

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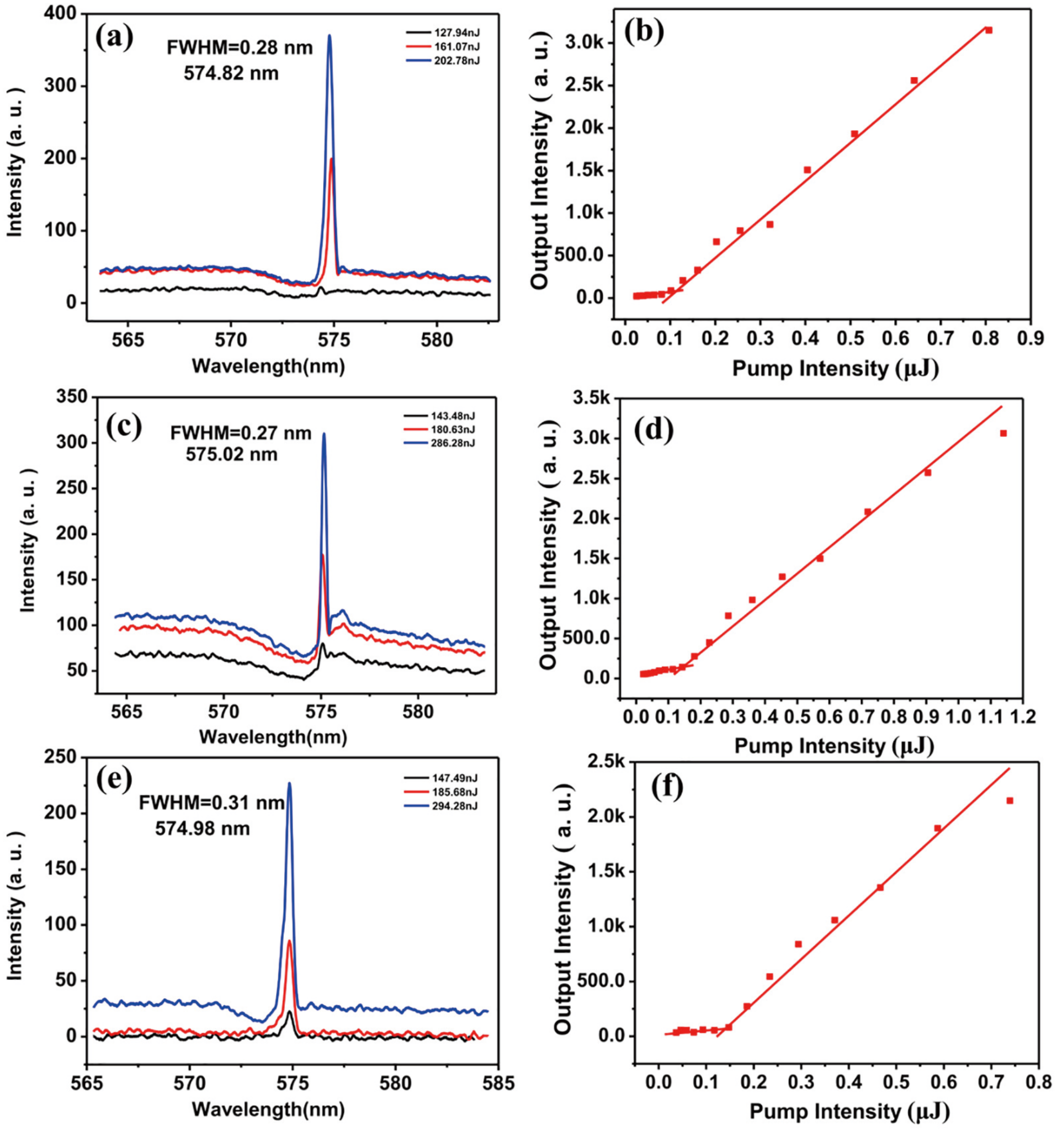

Fig. 9. (Color online) Emission spectra with pump pulse energy slightly above the lasing threshold for the organic laser based on F8BT, and output intensity as a function of the pump fluence (a, b) after the first stretch, (c, d) after the second stretch and (e, f) after the third stretch, respectively.

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