Continuous spatial tuning of laser emissions with tuning resolution less than 1 nm in a wedge cell of dye-doped cholesteric liquid crystals

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Abstract: We report the design and fabrication of a wedge structured CLC film incorporating a spatial gradient of a chiral dopant concentration. A continuous spatial laser tuning in the broad visible spectral range with tuning resolution less than 1 nm is demonstrated, which renders a CLC-based micron-sized laser an important continuously tunable laser device.

OCIS codes: (140.3600) Lasers, tunable; (300.6360) Spectroscopy, laser.

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1. Introduction

The chiral photonic bandgap properties of a cholesteric liquid crystal (CLC) originate from the self-organized helical arrangement of birefringent nematic mesogenes, which permits a formation of a polarization standing wave inside the CLC [1–3]. In addition, a sharply enhanced density of mode at the band-edge of the selective reflection band enables a low-threshold lasing operation near the band-edge when a stimulated emission process in present [1–4]. When compared with a distributed feedback thin-film laser, which is in a quarter-stack multilayer structure supporting an intensity-modulated standing wave, the absence of an intensity modulation within the CLC helical structure results in a significant narrowing of the linewidth of the longitudinal mode closest to the band-edge [1–4]. Consequently, the band-edge lasing spectrum usually exhibits a single-mode structure under a moderate optical pumping, which is one major advantageous characteristics of the CLC over a linear cavity laser [2]. In the application of a single-mode CLC laser to examples such as absorption spectroscopy, fluorescent excitation, and telecommunications, it is necessary to have spectral tunability. Since the spectral position of the CLC laser is determined by the magnitude of the helical pitch, the external stimuli, including electric field and mechanical strain, are employed to control the helical pitch, with the spatial position of the pumping beam fixed [5–7]. Also there is the possibility of tuning the laser emission wavelength through the influence of dye dopant concentration on the lasing wavelength [8–9]. Another important scheme to attain spectral tunability has been to employ spatial tuning by scanning a tightly focused pumping beam across the CLC cell, possessing a built-in spatial gradient of a helical pitch. A spatial pitch gradient is built-in by various means, such as a frozen thermal gradient in a fixed chiral dopant concentration, a position controlled UV curing, and diffusion of two chiral dopant concentrations [10–15].

In all the above examples, however, the single-mode laser line takes on discrete spectral values, separated by the free spectral range $(\Delta\lambda)_{fsr} \approx \lambda^2/(2\cdot n_f \cdot d)$ for a thin film thickness d of several to $\approx 20\mu m$ of the CLC cell composed of two parallel alignment layers. In an attempt to achieve a spectral tuning covering the full visible spectral range, a $16\mu m$ thick cell was fabricated, obtaining a pitch difference $\sim 8nm$ [16–17], and resulting in a tuning resolution of $\Delta\lambda \approx 8nm$.

In our previous work [18], it has been shown that a continuous, non discrete, spatial tuning of laser emissions in the spectral range of $\approx 9nm$ can be attained in a wedge CLC cell prepared with a fixed concentration of the chiral dopant. As the focused pumping beam scans across the wedge cell, the cell thickness goes through a continuous change. Boundary conditions such as surface tension and surface anchoring of nematics on two alignment layers possessing a spatial gradient of geometric spacing incurs a gradual change in the optical helical pitch near $\approx 9nm$. Hence, the discrete lines of the laser spectrum disappear, permitting a continuous spatial tuning of the single-mode CLC laser emission. However, the spectral range of laser wavelength tuning is limited only to $\approx 9nm$, which is simply from the free spectral range allowed in the polarization standing wave of the wedge CLC cell.

The increase of the laser wavelength tuning range with a continuous spatial tuning could be achieved by implementing a gradual change of the optical helical pitch in the wedge cell structure. In other words, we combine a continuous concentration gradient of chiral dopants and the wedge CLC cell, which will facilitate a continuous spatial tuning in the broad visible spectral range with a laser spectral tuning resolution of $\Delta\lambda \leq 1nm$. By this combination, we demonstrate a continuous spatial tuning of laser emissions with tuning resolution less than 1 nm in the broad visible spectral range. Details of cell fabrication and lasing measurements are

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presented in Sec. 2. Experimental results on the characteristics of laser emission are presented in Sec. 3 with discussion. Conclusion is presented in Sec. 4.

2. CLC cell fabrication and lasing measurements

The wedge cells were fabricated by employing two different sizes of spacers (thin, 8.25 μm; thick, 12.0 μm), and a spatial gradient with a thickness change from 1.5 to 2.0 μm over a lateral distance of $1.0\,\mathrm{cm}$ was achieved. See Fig. 1. The concentration gradient was established by half-filling with a high (low) chiral dopant concentration CLC doped with two or three laser dyes at the thin (thick) spacer position, respectively. The CLC wedge cells (W-cell) were then kept at room temperature for one to two weeks to develop a pitch gradient through diffusion of the helical rotatory power. As a control, a parallel CLC cell (P-cell) with a uniform thickness, prepared with a $12.0\mu m$ spacer, was filled in the same manner as the CLC wedge cell. As an alignment layer, SE-5291 polyimide (pretilt angle of $6^o \sim 7^o$, Nissan Chemical Korea Co. Ltd., korea) was employed to fabricate the CLC cells.

In order to cover the full visible spectral range, five different laser dyes were employed. According to dye combinations, 3 kinds of CLC wedge cells, namely, WL-cell, WM-cell, and WS-cell, were fabricated. WL-cell (DCM (Aldrich, USA) and LDS698 (Exciton, USA), in $\approx 1\,wt\%$); WM-cell (Coumarin500 (Exciton), Coumarin 540A (Aldrich), and Rhodamine 590 (Exciton), in $\approx 1\,\mathrm{wt\%}$); WS-cell (Coumarin 500 (Exciton), in $\approx 1\,\mathrm{wt\%}$ and Coumarin 540A (Aldrich), in $\approx 0.6\,\mathrm{wt\%}$). For the CLC materials, CLC-x1 ($\lambda_B = 450nm$) and CLC-x2 ($\lambda_B = 670nm$) (both from Merck, Germany) were employed.

As an optical pumping source, third harmonic generation 355 nm light from a Q-switched Nd:YAG laser (pulse width of 7 ns and repetition rate of 10 Hz) was employed. The pumping laser beam with ~2mm beam diameter was focused by a lens with a focal length of 20 cm and the beam waist (w) at the focal point is calculated with the formula $w = \lambda / \sin\theta = 71\,\mu m$ (where, λ is the wavelength of the pump beam and $\sin\theta = 1/200$). In order to find out an angle where anomalously strong absorption happens [8], the focused beam was incident obliquely on the sample with incidence angle, $20^\circ \sim 45^\circ$. So, the beam size on the sample could be increased up to ~1.5 times. It is expected that a smaller pump spot size will provide a narrower linewidth with a higher spectral resolution, while a change in the wedge angle does not affect the characteristics of spatial tuning.The generated laser emission along the normal of the CLC cell was collected by a spectrometer with a resolution of 0.36 nm (HR 2000+ , Ocean Optics, USA).

Figure 1 shows schematic diagram of the CLC pitch gradient developed in a wedge cell. The cholesteric helical pitches were quantized with the number of half- turns [19] by the boundary condition. Along the positive x-direction, a linear increase of the helical pitch continuously took place. However, if the CLC pitch gradient was developed in a parallel cell along the dashed guide-line, 1 in Fig. 1, only two, s1 and L1, discontinuous pitches (gray color) were allowed by the boundary condition. The pitches between s1 and L1 were elongated to fit their length to the long L1 pitch. The energy was minimized when the pitch was elongated between s1 and L1. Similarly, if the pitch gradient was developed with a negative slope along the solid line 2 by half-filling with a high (low) chiral dopant concentration CLC at the thick (thin) spacer position, discontinuous pitches (s2 and L2, black color) were allowed by the boundary condition (Fig. 3(g)). The pitches between the s2 and L2 were subsequently elongated to satisfy the geometrical thickness of the wedge cell thickness. There was a discontinuous pitch jumping between s2 and j2 that lead to the behavior of laser tuning (Fig. 3(g)).

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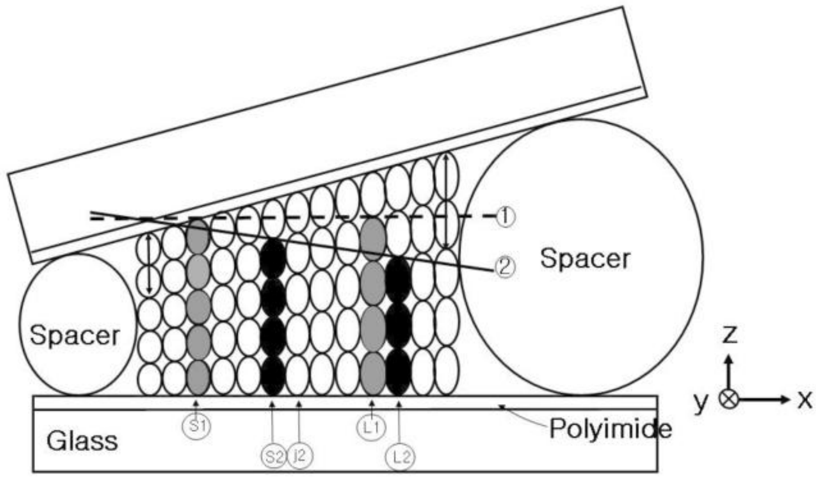

Fig. 1. Schematic diagram of the CLC pitch gradient developed in a wedge cell.

3. Experimental results and discussion

Figures 2 and 3 show the experimental measurements carried out on parallel cell (P-cell) and wedge cells (WL-, WM-, WS-cell), respectively. In the photographic images of Fig. 2(a) and 3(a, c, e), the P-cell and W-cell appear to be similar, with exception to the WM-cell, which shows a dye aggregation, Fig. 3(c). However, when examined with a polarization microscope, the area where the characteristics of the lasing and photonic band gaps (PBG) were measured revealed a difference between the parallel and wedge cells. In Fig. 2(b), the P-cell shows a discontinuous color change when crossing the Cano lines in each piece of the polarized microscope images. Nonetheless, in Fig. 3(b, d, f), the W-cells show a continuous color change stemming from a continuous pitch change, though dislocation lines can be observed at the sample edges where concentration mixing was not completed. When the generated laser spectra are compared in Fig. 2(c) and 3(h-1,-2,-3, and –4), there is a drastic difference in the tuning characteristics between the P-cell and WL-cell. As the tightly focused pumping laser (beam size $\approx 71\mu m$) scans across the P-cell, eight discrete laser spectral lines were observed in the P-cell, which were separated by a laser tuning difference of $\Delta\lambda \approx 10nm$ in a $12\mu m$ thick cell and could be estimated from the distortedly developed pitches of the parallel cell (gray colored 2 pitches in Fig. 1).

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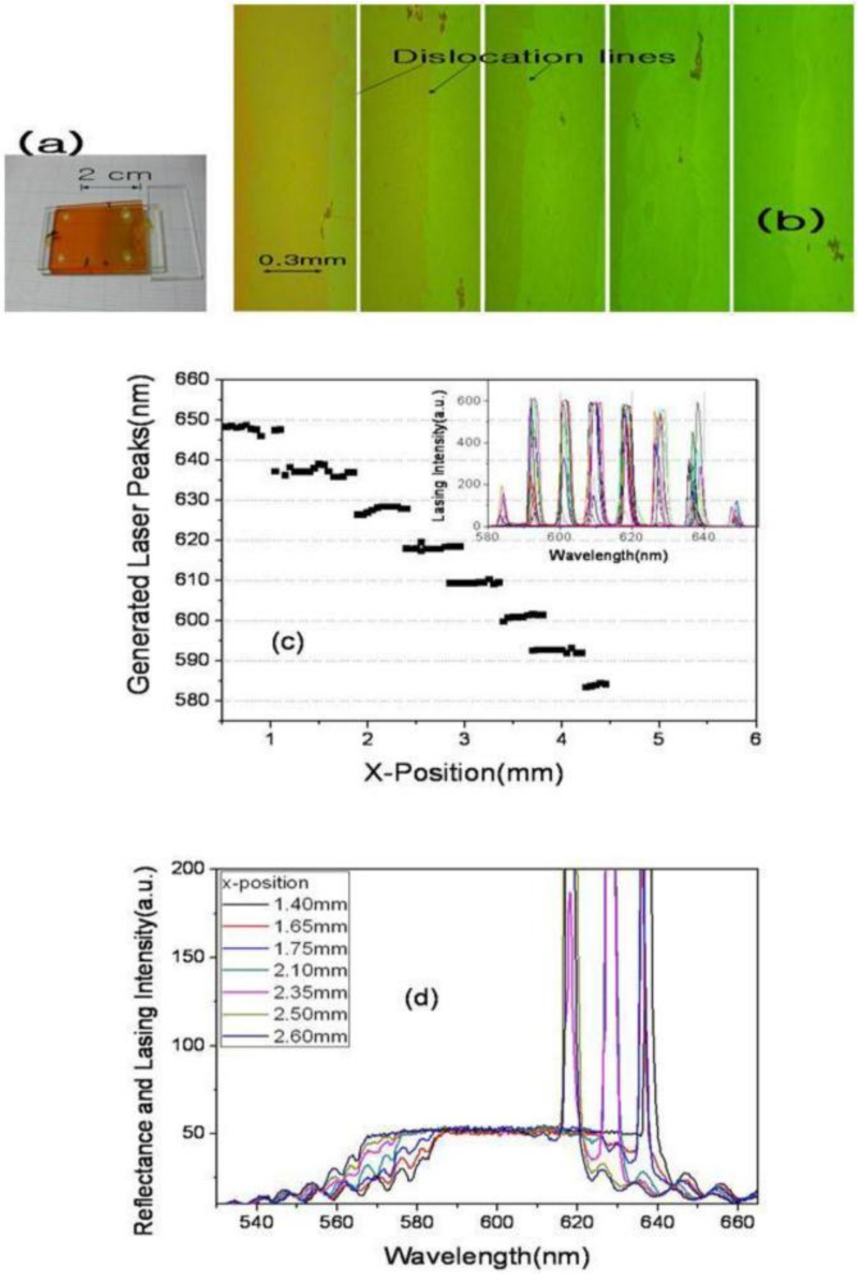

Fig. 2. (a) Photographic image of the P-cell; (b) juxtaposition of five pieces of polarized microscope images at different spatial positions; (c) laser line as a function of spatial position with the inset of the laser line spectrum; (d) stopband reflection and lasing spectrum for the P-cell.

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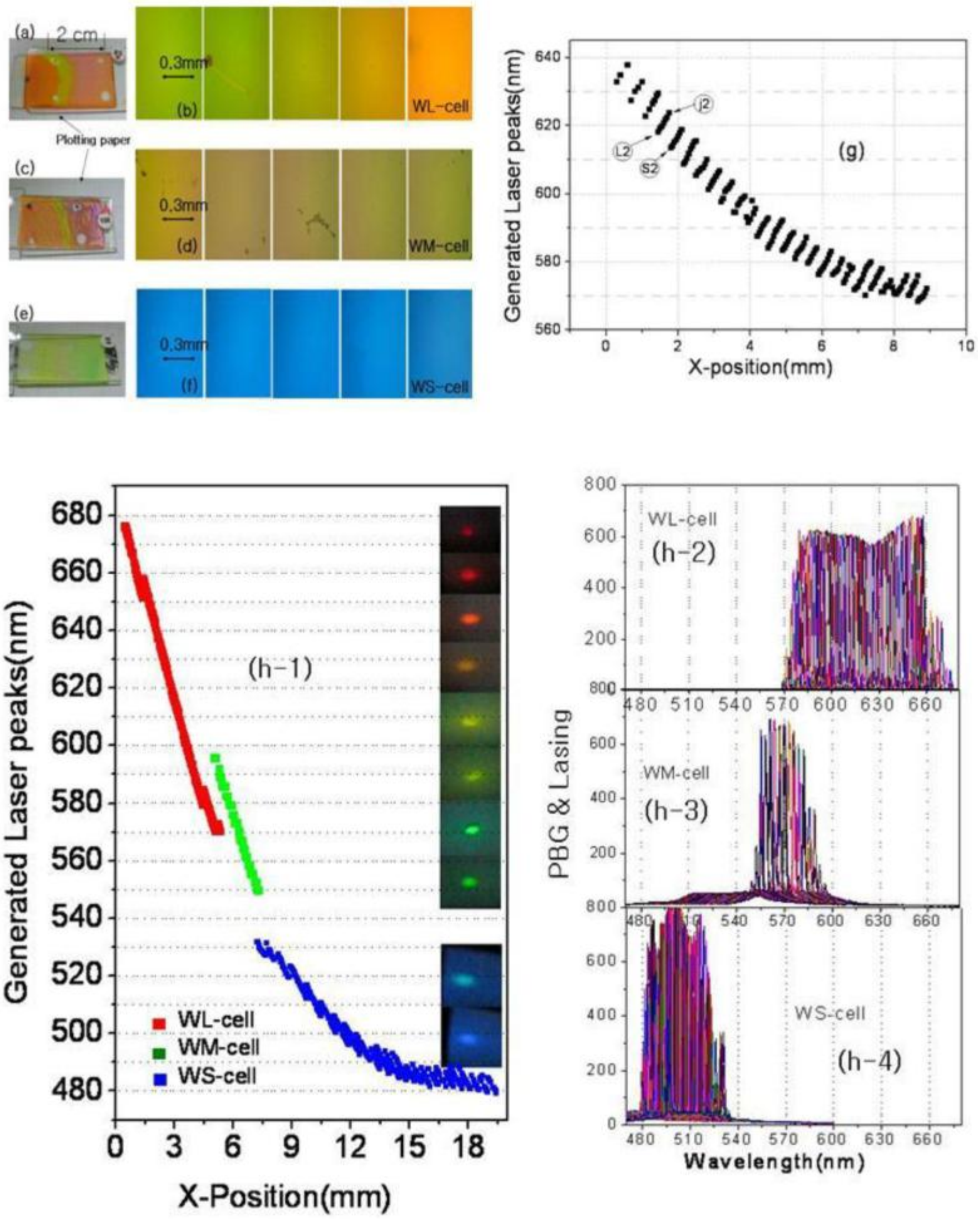

Fig. 3. Photographic images and juxtapositions of five pieces of polarized microscope images at different spatial positions of the WL-cell (a and b); the WM-cell (c and d); WS-cell (e and f); laser lines as a function of spatial position with the inset of the lasing photographs (h-1) and laser line spectra for each WL-, WM-, and WS-cell, respectively (h-2, –3, –4). Differently from the WL-cell, when the concentration gradient was developed with a negative slope (Fig. 1, solid line), the laser tuned like a toothed wheel due to boundary conditions (g). (Media 1)

However, in the WL-cell, a continuous tuning of the laser wavelength was achieved over a range of 107 nm, from 570.07 to 676.12 nm, with an accuracy of $\Delta\lambda \approx 1nm$, by a $50\mu m$ spatial movement of the cell in the x-direction (Fig. 4(a)). Moreover, the tuning resolution could be increased with an accuracy of $\Delta\lambda < 0.3nm$ by increasing the spatial

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resolution by a $10\mu m$ (resolution limit of the authors' system) movement in the x-direction, as shown in Fig. 4(b).

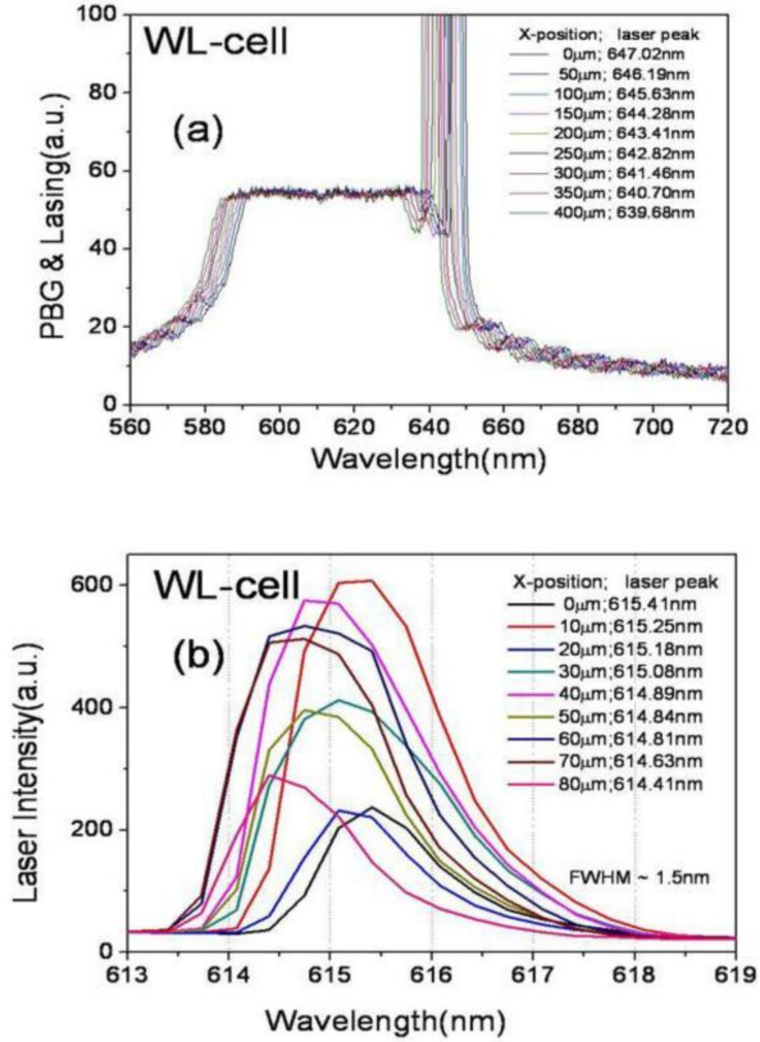

Fig. 4. (a) Stopband reflection and lasing spectrum change by a $50\mu m$ x-position movement; (b) lasing spectrum change by a $10\mu m$ x-position movement for the WL-cell.

Figures 2(d) and 4(a) show in detail the stop band reflection spectrum shape and laser line spectral position at different pumping positions. In the P-cell, Fig. 2(d), even in the presence of a continuous concentration gradient of the chiral dopant, discrete laser lines appear at the band gap edge when the pitch p takes on a value satisfying the polarization standing wave condition. Between the two laser lines, both the long and short edges of the stopband reflection spectrum suffer strong structural deformations since the pitch of the concentration gradient does not match with the helical pitch determined by the geometrical cell thickness. However, in the wedge cell (WL-cell, Fig. 4(a)), where thickness slope matches the pitch gradient of the CLC, the laser wavelength is tuned continuously without any structural deformation of band edges of the reflection spectrum.

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The wide continuous tunability was achieved through combination of a chiral dopant concentration gradient and wedge cell structure. In order to study the spectral position relation of stopband and lasing, the reflection and laser spectra are monitored simultaneously.

In order to extend the tuning spectral range to short wavelengths, another 2 kinds of wedge cells are introduced, namely, WS-cell and WM-cell. In the case of the WS-cell (Coumarin 500 and Coumarin 540A dyes; 8.25 and 15 μm spacers), a continuous tuning of the laser wavelength was achieved with the spectral width exceeding 53.18 nm, from 479.36 to 531.54 nm. The generated laser wavelength can be spatially tuned with the resolution of $\Delta\lambda \approx 1nm$, by a 50 μm movement in the x-direction. In this lasing action, the Forster energy transfer [20] from Coumarine 500 (sensitizer) to Coumarine 540A (emitter) is involved. Although Coumarin 540A exhibits a broad fluorescence emission spectra in the range of 480 ~600 nm in solvent, in the WS-cell case herein, the lasing range was not broad. In order to further extend the lasing range, the WM-cell (Coumarin 500, Coumarin 540A, Rhodamine 590 dyes were added) is introduced. With a fresh sample, a continuous tuning of laser wavelength was achieved in the range of 480 ~530 nm and 550 ~570 nm. However, after several days elapsed, the continuous lasing range was shrunk to 490 ~520 nm due to the fluorescence quenching through dye aggregation. When a thick cell was fabricated (32 and $35\mu m$ spaces used), the tuning range was 46.7 nm, from 549.8 ~595.5 nm, and the lasing intensity was high and stable (Fig. 3(g), WM-cell). In this case, the generated laser wavelength was spatially tuned with steps of $\Delta\lambda \approx 2.6\,nm$, by a $50\mu m$ movement in the x-direction. Due to a little lack of combination of a chiral dopant concentration gradient and wedge cell structure, the tuning step was larger than 1 nm. Furthermore, the lasing could be also generated in a long wavelength (more than 570 nm range) by a 532 nm laser pumping, with nearly the same results measured in other cells.

4. Conclusion

A continuous spatial tuning was achieved in the broad visible spectral range with a wavelength tuning resolution of $\Delta\lambda \le 1\,nm$ by developing a continuous helical pitch gradient of the CLC in a wedge cell. The authors found that in order to satisfy the boundary conditions within the CLC cell, the length of the pitch length required elongation up to 9 nm. In order to realize the CLC photonic structure as a continuous tunable laser device, a long-term temporal and thermal stability of the helical pitch gradient was an indispensable condition. Thus, by also introducing a thin polymerizable CLC wedge cell structure with a pitch gradient, this lab could present a facile route to fabricate continuous tuning laser devices. Currently, the authors are investigating fabrication of one single wedge cell device with a polymerizable CLC for continuous laser tuning in the full visible range. This wedge cell structure method could be widened to cover the optical spectrum to include UV, VIS, and the IR spectra. When this new type of CLC cell is employed in the optically pumped laser operation, the continuous spatial tuning range of the laser wavelength is only limited by the availability of appropriate laser dyes with the spectral linewidth and the CLC material. The scheme of spatial laser tuning in a wedge cell incorporated with a chiral dopant concentration opens a way to designing small-sized optical devices that allow for a wide tunability of single-mode laser emissions, which could potentially be an alternative for the nonlinear-optics based optical parametric oscillator normally adopted for wide spectral range tuning.

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