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Laser Action in Dye-doped Photopolymerized Cholesteric Liquid Crystal

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Optically pumped mirrorless laser action has been observed in a dye-doped photopolymerized cholesteric liquid crystal (PCLC). At high excitation energy above the threshold, the laser action is observed at an edge of the one-dimensional photonic band of the PCLC helical structure. It is also manifested that the well-ordered alignment of liquid crystal molecules is essential for the realization of the laser action of PCLC.

KEYWORDS : cholesteric liquid crystal, photonic crystal, laser, stop band

色素をドープした光重合コレステリック液晶高分子薄膜におけるレーザー発振

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色素をドープした光重合コレステリック液晶高分子薄膜において、光励起によるレーザー発振の観測に成功した。閾値以上の励起光強度において、高分子コレステリック液晶の一次元螺旋周期構造により形成されるストップバンド端においてレーザー発振が観測された。レーザー発振には、液晶分子の良好な配向状態が必要であることも明らかにした。

1.introduction

Recently, a photonic crystal (PC) which is made from a periodic dielectric structure with a periodicity in a range of optical wavelength have attracted much attention from both fundamental and practical points of view[1, 2]. In the PC, the propagation of light is inhibited under the Bragg condition, which results in an appearance of an optical stop band or a photonic band gap (PBG). In the PBG, spontaneous emission is inhibited, so that a low-threshold laser action can be expected. The PC having a one-dimensional (1-D) periodic structure is called a 1-D PC. Although, in the 1-D PC, the complete localization of photon such as in the 3-D PC cannot be achieved, a laser action can be expected at the stop band edge, where the photon group velocity approaches zero[3].

Liquid crystals such as a cholesteric liquid crystal (CLC) and a chiral smectic liquid crystal, which have periodic helical structures with a periodicity of the optical wavelength, can be regarded as the 1-D PCs. In such liquid crystals with the helical structure, a circularly polarized light with the same handedness as the helix propagating along a helical axis is selectively reflected (selective reflection) and a stop band appears. Recently, the laser actions have been observed in both CLC[4,5] and chiral smectic liquid crystal[6]. In these compounds, 1-D periodic helical structures act as laser cavities and mirrorless distributed feedback (DFB) laser action was achieved at the photonic stop band edges.

In this letter, the laser emission of a photopolymerized CLC (PCLC) is presented. In such a flexible film, it can be investigated whether one-dimensional helical structure necessary for laser action is maintained even in deformed film. Moreover, the helical pitch of the photo-polymerized CLC polymers has no temperature dependence. This means that the operation wavelength of laser action is stable thermally. This is favorable from the view point of the laser device (Fig. 1).

2. Experiment

For the fablication of PCLC, two types of photo-polymerizable CLC mixtures (Merck KGaA) were used, which have right-handed helix and reflection bands around 779 nm and 440 nm. The helical pitch of the CLC sample, that is, the wavelength of the optical stop band can be adjusted by mixing these two CLC compounds at a proper ratio. As a laser dye dopant in the CLC, [2-[2-4-(Dimethylamino)phenyl]ethenyl]-6-methyl-4H-pyran-4-ylidene] propanedinitrile, DCM (Exciton) was used. The concentration of the dye was 0.4 wt.-%.

Two types of the PCLC cell were prepared as follows. One of them is consisted of two glass plates without any surface treatment for an alignment of liquid crystals. The monomer sample was inserted by capillary action into the cell. In order to obtain a homogeneously aligned cell, a mechanical shear was applied to the cell (type I). Another cell is made from two glass substrates which surfaces were coated with a polyimide (Japan Synthetic Rubber, AL1254) and rubbed (type II). The monomer sample was inserted by capillary action into the cell. A mechanical shear wasn't applied to the type II cell. The gap of these cells was 25 μm. The CLCs in these cells align these molecular long axes (director) parallel to the glass plates, that is, the helical axis is perpendicular to the glass substrate. Then UV light irradiation was performed using a Xe lamp to induce the photo-polymerization of the UV-curable CLC monomer. The transmission spectrum measurement was performed using a spectrometer (Shimadzu, UV-3150).

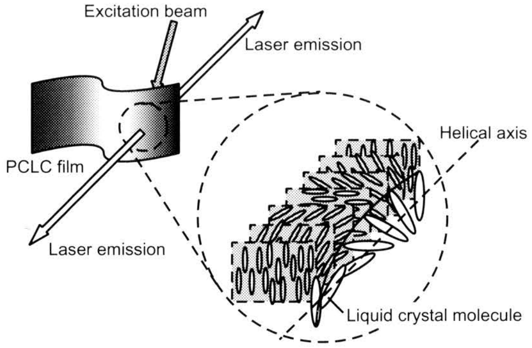

Fig.1. Schematic representation of the PCLC laser

A second harmonic light of Q-switched Nd:YAG laser (Spectra Physics, Quanta-Ray INDI) was used for an excitation, whose wavelength, pulse width and pulse repetition frequency were 532 nm, 8 ns and 10Hz, respectively. The illumination area on the sample was about 0.2 mm^2. The excitation laser beam irradiated the sample at an angle of about 45° with respect to the cell plate normal. The emission spectra from the dye-doped PCLC film were measured from an opposite side of the cell using a charge-coupled device (CCD) multichannel photodetector (Hamamatsu Photonics, PMA-11) having a spectral resolution of 3 nm. The collecting direction was perpendicular to the cell surface, which is along the helical axis.

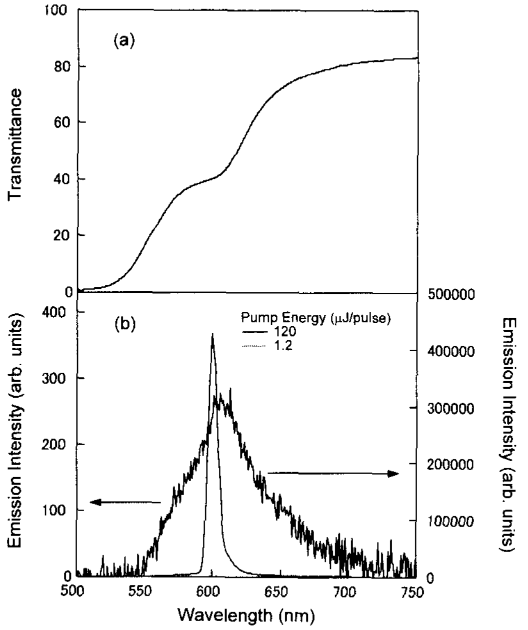

Fig.2. (a) Transmission spectrum of PCLC type I cell. (b) Emission spectra off a dye-doped PCLC type I cell at 3.0 μJ/pulse (dotted line) and 30 μJ/pulse (solid line) of the pump pulse energy.

3. Results and Discussion

Figure 2 (a) shows the transmission spectrum of the dye-doped PCLC type I cell. In Fig. 2 (a), the drop of the transmittance due to the selective reflection is observed around 600 nm which corresponds to longer wavelength stop band edge. Transmittance in shorter wavelength than about 570 nm is suppressed by the absorption of the doped laser dye.

Figure 2 (b) shows the emission spectra of the dye-doped PCLC film for various values of the excitation pulse energy. For low excitation energy (< 1.2 μJ/pulse), the spectrum is dominated by a broad spontaneous emission (SE) as shown in dotted line. The suppression of SE due to the stop band can be observed in shorter wavelength than 600 nm, which coincides with the result of the transmission spectra shown in Fig. 2 (a). At high excitation energy (30 μJ/pulse), a nallowing of linewidth of the emission spectrum is observed at the edge of the dip as shown in solid line. The full width at half maximum (FWHM) of the emission peak is about 8nm.

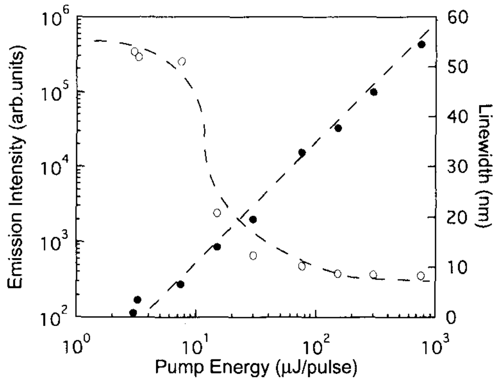

Fig.3. Pump energy dependence of the peak intensity and linewidth (FWHM) of the emission spectrum of the dye-doped PCLC type I cell.

The peak intensity and linewidth of the emission spectrum are shown in Fig. 3 as a function of the pump pulse energy. Although the linewidth of the emission spectrum drastically decreases above excitation energy of about 10 μJ/pulse, the emission intensity increases in proportion to the pump energy. This indicates that the nallowing of linewidth of the emission spectrum is not due to the lasing.

Figure 4 (a) shows the transmission spectrum of the dye-doped PCLC type II cell. In Fig. 4 (a), the drop of the transmittance due to the selective reflection is observed around 600 nm same as type I cell but sharper than that of type I.

Figure 4 (b) shows the emission spectra of the dye-doped PCLC film for various values of the excitation pulse energy. At high excitation energy (30 μJ/pulse), laser action is observed at the edge of the dip as shown in solid line. The full width at half maximum (FWHM) of the emission peak is about 2nm, which is limited by the spectral resolution of our experimental setup.

The peak intensity and linewidth of the emission spectrum are shown in Fig. 5 as a function of the pump pulse energy. This indicates the presence of a lasing threshold. At lower excitation energies, the emission intensity increases in proportion to the pump energy. Above the threshold at a pump pulse energy of

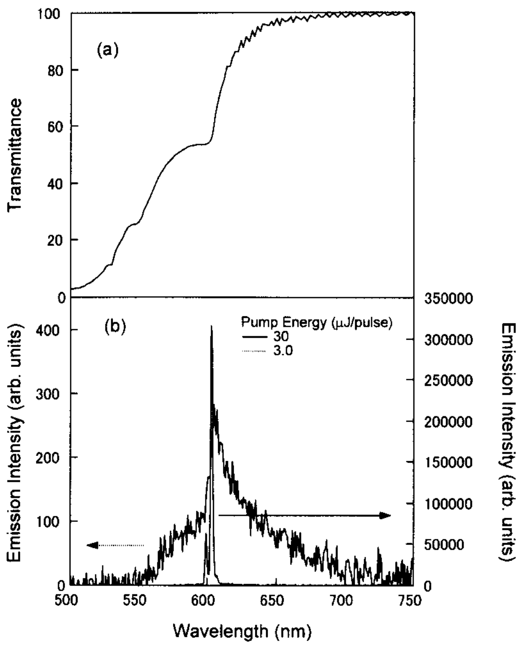

Fig.4. (a) Transmission spectrum of PCLC type II cell. (b) Emission spectra off a dye-doped PCLC type II cell at below (3.0 μJ/pulse, dotted line) and above (30 μJ/pulse, solid line) the threshold pump pulse energy. A sharp lasing peak appears at the edge of the stop band for high-energy excitation.

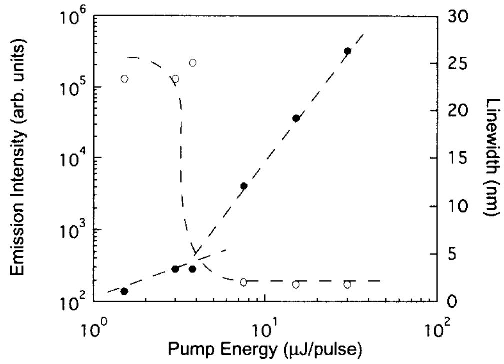

Fig.5. Pump energy dependence of the peak intensity and linewidth (FWHM) of the emission spectrum of the dye-doped PCLC type II cell.

about 3 μJ, the emission intensity increases nonlinearly. The linewidth of the emission spectrum also drastically decreases above the threshold. These results confirm that lasing occurs above the threshold of the pump energy at the edge of the photonic stop band in the spontaneous emission.

The reason why the laser action wasn't observed in type I cell can be attributed to the inhomogeneity of the alignment of liquid crystal molecules. From the comparison of transmittance of both cells (Figs. 2 (a) and 4 (a)), it can be considered that well-ordered alignment of liquid crystals is not obtained in type I cell. The well-ordered alignment of liquid crystal molecules is considered to be essential for the realization of the laser action of the PCLC.

4. Conclusions

In conclusion, optically pumped mirrorless laser action was observed at the edge of the stop band of the dye-doped PCLC. At high excitation energy above the threshold, the laser action is observed at an edge of the one-dimensional photonic band of the PCLC helical structure. It is also manifested that the well-ordered alignment of liquid crystal molecules is essential for the realization of the laser action of the PCLC.

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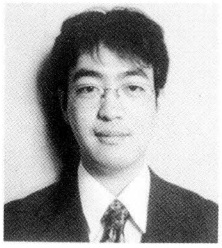

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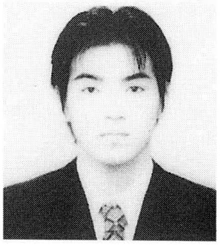

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