Laser Action at the Edge of Stop Band in Dye-doped Ferroelectric Liquid Crystal

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Optically pumped laser emission has been observed at the edge of one-dimensional photonic band of dye-doped chiral smectic liquid crystal with a periodic helical structure which is a so-called ferroelectric liquid crystal. Lasing wavelength was tuned with temperature. A large wavelength shift to higher wavelength was observed with increasing temperature, which corresponds to the increase in the helical pitch for the ferroelectric liquid crystal.

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

色素をドープした強誘電性液晶のストップバンド端におけるレーザー発振

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色素をドープしたカイラルスメクチック液晶（強誘電性液晶）を光励起したところ、螺旋周期構造に起因するストップバンドの端からレーザー発振を観測した。カイラルスメクチック液晶におけるレーザー発振の観測ははじめてである。温度の変化に伴って、螺旋ピッチの変化すなわちストップバンドのシフトに対応して、レーザー発振波長がシフトすることが確認された。

1.Introduction

Photonic crystals having an ordered structure with a periodicity of optical wavelength have attracted considerable attention from both fundamental and practical points of view, because novel physical concepts such as photonic band gap have been theoretically predicted and various applications of photonic crystals have been proposed.[1-2]
Especially, the study of stimulated emission in photonic band gap is one of the most attractive subjects, since, in the band gap, a spontaneous emission is inhibited and low-threshold lasers based on photonic crystals are expected.[2-7]So far intensive studies on one- and two- dimensional band gap materials have been performed.In a one-dimensional periodic structure, the laser action has been expected at the photonic band edge where the photon group velocity approaches zero.[8]

Liquid crystals including chiral molecule have a self-organized helical structure which is a one-dimensional periodic structure and shows characteristic optical properties.[9] In cholesteric liquid crystals with helical structure, light propagating along the helical axis is selectively reflected depending on the polarization states if the wavelength of the light matches to the optical pitch of the helical structure, which is a so-called selective reflection.The wavelength region in which the light can not propagate is the stop band, which is considered as a one-dimensional pseudo-bandgap. Lasing at the band edge has been reported in the cholesteric liquid crystal.[10-12]

Chiral smectic liquid crystals with tilted structure show a ferroelectricity, which is called ferroelectric liquid crystal (FLC), and have an expected potential for the electrooptic applications because of the fast response to the electric field.[13]The FLC also has a helical structure and shows the selective reflection due to the one-dimensional periodic structure as the almost same manner as the cholesteric liquid crystal.[14]However , although studies on the enhancement of a nonlinear effect using the helical structure of FLC have been performed,[15]the control of the stimulated emission and laser action in the helical structure of FLC have not been demonstrated yet.

In this study, we prepare FLC mixtures having a well-controlled short pitch of the helical structure whose periodicity is equivalent to the visible optical wavelength, and successfully demonstrate a laser action at the edge of the photonic stop band of the dye-doped FLC for the first time.

2. Experiment

The FLC compound used in this study is a multi-component mixture having the chiral smectic C (SmC*) phase in a wide temperature range including a room temperature (~0 oC to68oC).A helical pitch of the FLC changes from less than300nm to500nm with temperature.As a laser dye doped in the FLC, a Coumarin500(Exciton) was used.The concentration of the dye is0.2wt%.The sample was filled into a sandwich cell which consists of two glass plates.The cell gap was50mm.In order to obtain a homeotropically aligned cell, surfaces were coated with a polyimide (JALS-2021-R２, Japan Synthetic Rubber). In the homeotropically aligned cell, the helicoidal axis is perpendicular to the glass substrates.

For excitation source of emission measurement, second harmonic light of a regenerative amplifier system based on a Ti:sapphire laser (Spectra Physics) was used. The pulse width, wavelength and pulse repetition frequency of the output laser beam were150fs, 400nm and 1 kHz, respectively.The excitation energy can be varied within the range from0.01to28mJ/pulse.The illumination area on the sample was about0.2mm2. The excitation laser beam irradiated the sample at an angle of45o with respect to the cell plate normal.The emission spectra from the dye-doped FLC were measured from the opposite side of the cell using a CCD multichannel photodetector (Hamamatsu Photonics, PMA-11) having spectral resolution of3nm. For a high resolution measurement, a spectrograph (Oriel, MS257) with a CCD detector was used, whose spectral resolution is0.15nm.The

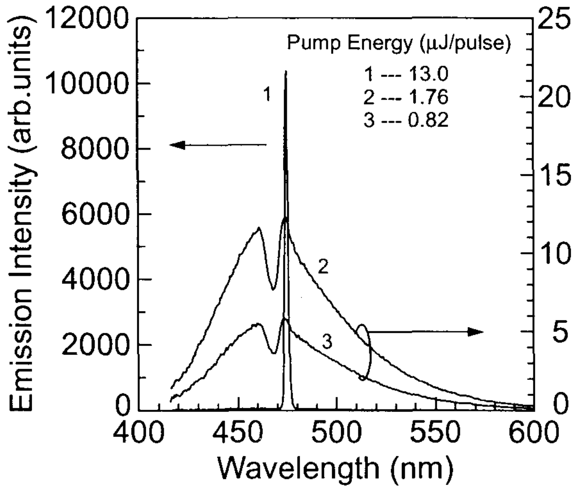

Fig.1: Emission spectra of a dye-doped FLC as a function of pump pulse energy.A sharp lasing spectrum appears at the edge of the stop band by the high-energy excitation (13.0mJ/pulse).

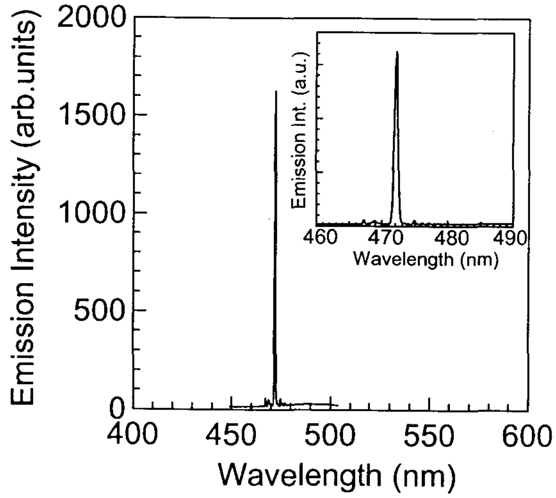

Fig.2: High resolution emission spectra of a dye-doped FLC at high-energy excitation.

collecting direction was perpendicular to the cell surface, which is normal to the smectic layers and along the helical axis.

3.Results and Discussion

Figure 1 shows the emission spectra of the dye-doped FLC as a function of the excitation pulse energy at20oC.For a low excitation energy (<1.76mJ/pulse), the spectrum is dominated by a broad spontaneous emission and the dips are observed in the broad spectra.The wavelength of the dip coincides with that of the stop band for the half pitch.With increasing the pump energy, the emission intensity is enhanced.At high excitation energy (13.0mJ/pulse), lasing appears as a sharp peak at the edge of the dip.Figure 2 shows a high resolution emission spectrum of the dye-doped FLC at high energy excitation (17.7mJ/pulse).The full width at half maximum (FWHM) of the emission peak is about0.6nm , which is limited by the spectral resolution of our experimental setup.

The peak intensity and linewidth of the emission spectrum are shown in Fig.3as a function of the pump pulse energy.This clearly indicates the presence of a lasing threshold.Above the threshold at a pump pulse energy of about3mJ , the emission intensity non-linearly increases.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 threshold pump energy for lasing would be lowered by optimizing a doped laser dye, a concentration of the dye, a molecular tilt angle of FLC with respect to the smectic layer and cell geometries such as the thickness and smectic layer arrangement.The design of molecular structure of the host FLC for a high birefringence might be one of the most important factors for the low threshold lasing.

Figure 4 shows the emission spectra below and above the lasing threshold at various temperatures.The

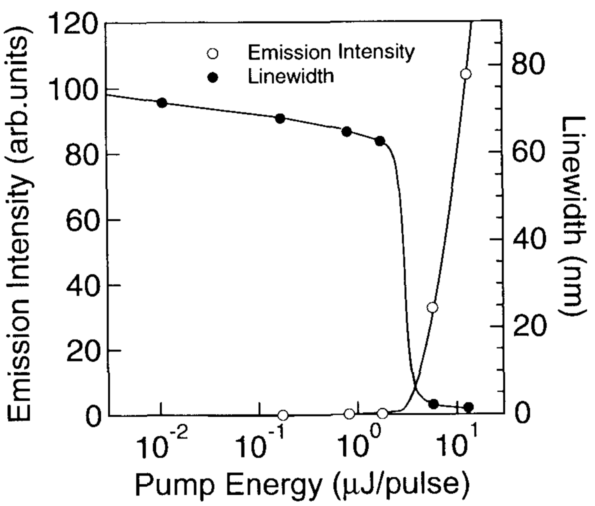

Fig.3: Pump energy dependences of peak intensity and linewidth (FWHM) of the emission spectrum of dye-doped FLC.

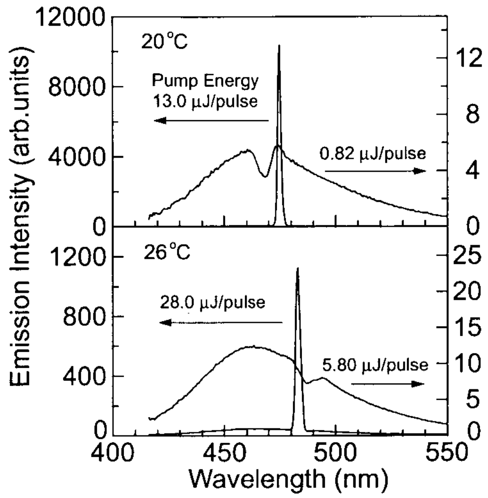

Fig.4: Temperature dependence of the emission spectra of dye-doped FLC at low and high energy excitations.

dip in the spontaneous emission spectrum at low pump energy shifts with the temperature.This stop band shift corresponds to the change in the helical pitch of the host FLC.The helical pitch increases with increasing temperature. Therefore, the dip shifts toward longer wavelength as the temperature increases, as shown in Fig.4.As is evident from Fig.4, the lasing wavelength also shifts with the temperature, which follows the dip shift in the spontaneous emission spectra.It should be noted that, at both temperatures, lasing occurs at the edge of the stop band dips.This indicates that lasing is strongly associated with the presence of the stop band.At higher temperature (29oC), lasing is not observed at any pump energy up to the damage threshold. This might be because the stop band wavelength deviates far from the spontaneous emission peak, which also supports the fact that the photonic stop band is essential to the laser action.

As shown in Fig.4, the lasing wavelength can be easily tuned in a wide spectral range by changing the temperature.In addition, the helical pitch of the FLC can be controlled also by the electric field, and FLC shows fast response of the molecular reorientation to the field because of the strong interaction between the field and a spontaneous polarization.Therefore, a high-speed modulation of the laser action is expected using the dye-doped FLC proposed in this study.

4.Conclusions

In conclusions, optically pumped laser emission was observed at the edge of one-dimensional photonic band of dye-doped chiral smectic liquid crystal with a periodic spiral structure.Lasing wavelength was tuned by adjusting the period of the helical structure with temperature.

Acknowledgment

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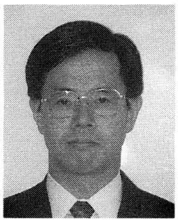

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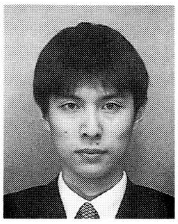

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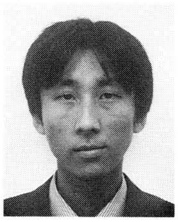

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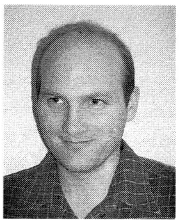

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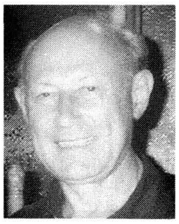

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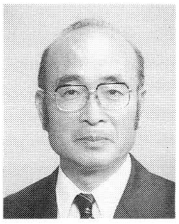

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