Electrically-Tunable Chiral Laser based on Ferroelectric Liquid Crystals

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1. INTRODUCTION

Photonic crystals (PCs) having a three-dimensional (3-D) ordered structure with a periodicity of optical wavelength have attracted considerable attention from both fundamental and practical points of view, because in such materials 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.   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. In order to realize the photonic crystal, a large number of intensive studies on a micro-fabrication based on a semiconductor processing technology and a self assembly construction of nano-scale spheres have been carried out.

Liquid crystals (LCs) including chiral molecule have a self-organized helical structure which can be regarded as a 1-D periodic structure.   In such systems, there is a so-called stop band in which the light can not propagate, which is considered as a 1-D pseudo-bandgap.   Lasing at the band edge has been reported in the cholesteric liquid crystal (CLC), chiral smectic liquid crystal[3] and polymerized cholesteric liquid crystal.[4]   These laser action in the 1-D helical structure of the chiral liquid crystals are interpreted to be based on the band edge of the 1-D photonic band gap in which the photon group velocity is suppressed.

Chiral smectic liquid crystals with a tilted structure show a ferroelectricity, which are called ferroelectric liquid crystal (FLC), and have an expected potential for the electrooptic applications because of a fast response to the electric field. 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 CLC.   The helix of FLC can be easily deformed upon applying electric field and its response is fast because of the strong interaction between the spontaneous polarization and electric field. Therefore, a fast modulation of the lasing wavelength upon applying electric field can be expected in a dye-doped FLC.

In this study, we report tunable lasing in dye-doped LCs with 1-D periodic structure. We successfully demonstrate a lasing wavelength tuning in a wide range upon applying electric field for the first time.

2. EXPERIMENTAL

For the host compound of FLC laser, a multi-component mixture having the chiral smectic C (SmC*) phase in a wide temperature range including a room temperature was used. As a laser dye doped in the FLC, a Coumarin 500 (Exciton) was used. The concentration of the dye is 0.2 wt%. The sample was filled into sandwich cells of two types of cell configurations; one is a homeotropically aligned cell and the other is a planar cell configuration for waveguide laser. In

the homeotropically aligned cell, aluminum foils were used as both spacers determining cell thickness and electrodes, which allows us to apply the electric field parallel to the glass substrates. The cell gap was 50μm. The electrodes distance is 2mm.　In order to obtain a homeotropically aligned cell in which the smectic layers are parallel to the substrates, surfaces were coated with a polyimide (Japan Synthetic Rubber, JALS-2021-R2). In the homeotropically aligned cell, the helicoidal axis is perpendicular to the glass substrates. For a planar cell in which the helical axis is parallel to the glass substrates, surfaces of glass substrates were coated with polyimide (JSR, AL1254) and were rubbed unidirectionally. In order to apply an electric field normal to the helical axis, glass plates coated with an In-Sn oxide (ITO) were used as substrates. Thin glass plates were used as spacers, whose thickness was about 150 μm.

As an excitation source for FLC laser, a 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 were 150fs, 400nm and 1 kHz, respectively. For the homeotropically aligned cell geometry, the excitation laser beam irradiated the sample at an angle of 45° with respect to the cell plate normal.　The illumination area on the sample was about 0.2 mm². The emission spectra from the dye-doped FLC were measured from the opposite side of the cell. The collecting direction was perpendicular to the cell surface, which is normal to the smectic layers and along the helical axis. On the other hand, for the waveguide laser, the excitation laser beam irradiated the sample perpendicularly to the cell plate and was rectangularly focused using two lenses, concave lens and cylindrical lens, to area of 0.3 mm². The long axis of the irradiated area on the sample is parallel to the helical axis of the FLC. The emission along the helical axis passed through the spacer glass film and went outside the cell.

3. RESULTS AND DISCUSSION

3.1　Electrical tunability of lasing wavelength in dye-doped FLC

The lasing characteristics of FLC in the absence of the electric field have been investigated. Figure 1 shows emission spectra of dye-doped FLC as a function of pump energy. For low pump energy (1.76μJ/pulse), the spectrum is dominated by a broad spontaneous emission and the dip is observed in the broad spectrum. The dip originates from the selective reflection band due to the helix of FLC. As the excitation energy increases, the emission intensity is enhanced.　At high excitation energy (10.4 μJ/pulse), lasing appears as a sharp peak at the lower energy edge of the dip. The full width at half maximum (FWHM) of the emission peak is less than 0.5nm.

Figure 1: Emission spectra of a dye-doped FLC as a function of pump pulse energy.

The laser light emitted from the FLC is circularly polarized and the sense of the polarization is right-hand, which coincides with the helical sense of the FLC used here. In the stop band due to the helical structure, only the circularly polarized light with the same sense as that of the helix selectively interacts with the periodic helical structure and is reflected, which is known as selective reflection. Therefore, the result of the polarization dependence measurement strongly supports that the laser action in the dye-doped FLC is based on the band effect in a periodic structure of the FLC helix.

FLC has a spontaneous polarization P_s which points normal to the molecules and parallel to the smectic layers. When the electric field is applied in the layer, for lower field P_s intends to point along the field direction and FLC molecules starts to reorient toward the direction normal to the, resulting in the deformation of the helix. In the equilibrium state, the deformation of the helix might cause the elongation of the periodicity of the helix. Above the threshold field, all FLC molecules orient to the same direction and the helix is unwound.
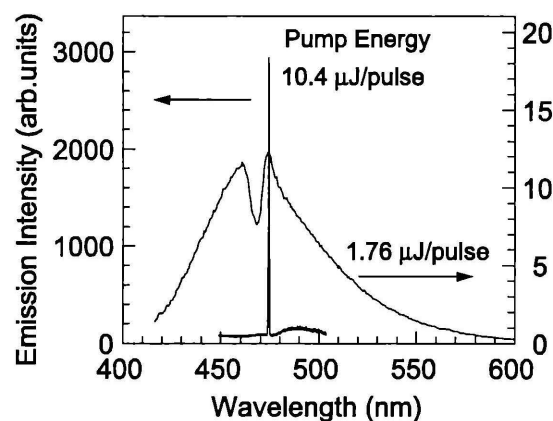
The fact that the periodicity of the helical structure of the dye-doped FLC can be controlled upon the field application allows us to expect the electric field tuning of the laser emission wavelength. Figure 2 shows the lasing spectra of the dye-doped FLC at high excitation energy (24μJ/pulse) as a function of the applied electric field. It should be noted that lasing wavelength largely shifts toward longer wavelength with increasing the field, which corresponds to the shift of the selective reflection band. In spite of a low field (3.5kV/cm), a wide tuning of the lasing wavelength was achieved.[5]
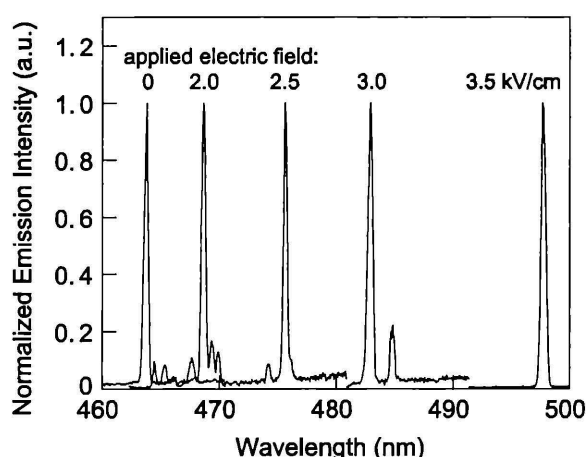

Figure 2: Normalized emission spectra at high excitation energy in a dye-doped FLC as a function of applied electric field.
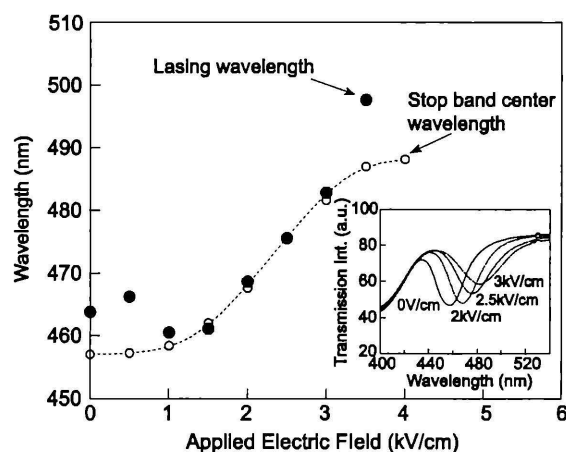
Figure 3: Applied electric field dependences of lasing wavelength and stop band center wavelength of a dye-doped FLC.

Figure 3 shows electric field dependence of the lasing wavelength. The selective reflection center wavelength determined from the transmission spectra (the inset) is also shown as a function of the field. It can be confirmed that the shift of the lasing wavelength is based on the band shift caused by field-induced change in the helix periodicity. It should be noted that for the medium field range the lasing is observed at the center of the stop band.

The electrooptical effects originating from the field-induced deformation of the helix in the FLC have been proposed. Especially, the response of the electrooptical switching based on a small deformation of the helix of a short pitch FLC is as fast as several μs order and the application to an optical communication as well as to a display device has been proposed. The relaxation times of the helix deformation of FLC is represented by the following equation,

$$\tau \propto \frac{p^2 \gamma}{K},$$

where p is the helix pitch, γ is the rotational viscosity and K is the elastic constant.

According to this relation, the response time is inversely proportional to p^2, and high frequency modulation of the periodicity of the helix can be expected in a short pitch FLC. In deed, the electrooptic modulation device using a similar compound to the FLC material used in this study has of the order of several μs response. Consequently, the fast modulation of the lasing is possible in the dye-doped FLC with a short pitch.

3.2　Waveguide laser of FLC

Lasing in the FLC mentioned above has been performed in the cell configuration in which a helical axis is perpendicular to the substrates and laser light is emitted out perpendicularly to the cell surface, as shown in Fig. 4(a). In this configuration, it is not easy to achieve alignment of high quality in a thick cell, so that it is difficult to extend an

active region for the lasing. Moreover, a pump beam is absorbed in the vicinity of the interface between liquid crystal and substrate, and doped dye in a bulk is not effectively excited. We design a planar cell configuration of dye-doped FLC for lasing as shown in Fig.4(b) and demonstrate optically pumped lasing in a waveguide.[6] In this waveguide liquid crystal laser, emission wavelength can be widely controlled by applying electric field.

As the same manner as the case of homeotropically aligned cell configuration, there exists a lasing threshold in the pump energy dependence of the peak intensity and linewidth of the emission spectrum in the waveguide configuration. At lower excitation energy, the emission intensity increases in proportion to the pump energy. Above the threshold at a pump pulse energy of about 10 μJ/pulse, the emission intensity nonlinearly 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 lasing wavelength shifts toward longer wavelength with increasing voltage. In spite of a low field (2.0 kV/cm), a wide tuning of the lasing wavelength was achieved.
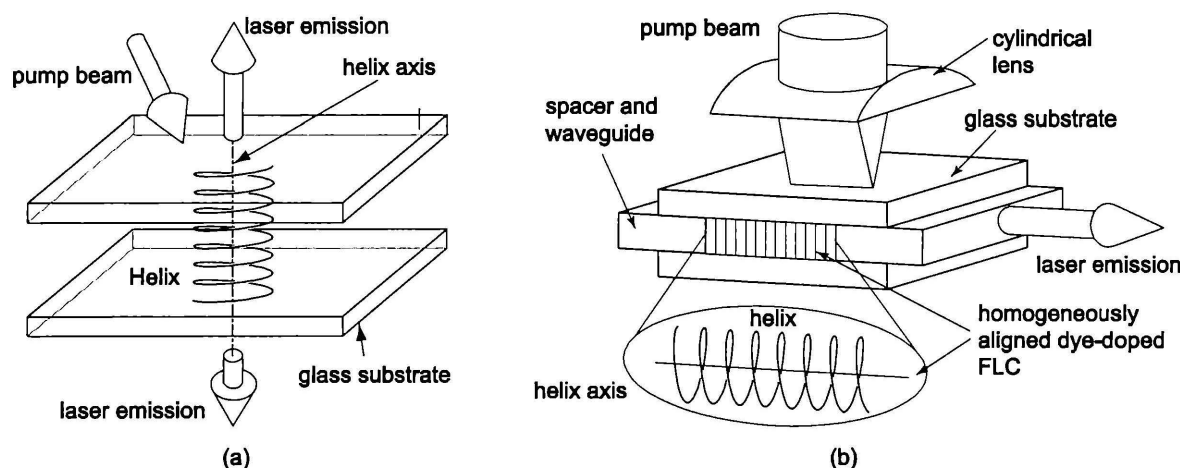

Figure 4: Cell configurations for the laser action; (a) homeotropically aligned cell, (b) planarly aligned cell for waveguide lasing.

SUMMARY

Electrically-tunable lasing in dye-doped chiral liquid crystal with one dimensional periodic structure was reported. We also designed a planar cell configuration of dye-doped FLC for lasing and demonstrated optically pumped lasing in a waveguide. In this waveguide liquid crystal laser, emission wavelength could be widely controlled by applying electric field.

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