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Lowering lasing threshold in ferroelectric liquid crystal sandwiched between dielectric multilayers

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The authors have investigated laser action in ferroelectric liquid crystal (FLC) sandwiched between dielectric multilayers composed of SiO_2 and TiO_2. The single-mode laser action was observed at the band edge of FLC. The lasing threshold was much lower than that of simple FLC because of the double optical confinement caused by the sandwich structure. They have also demonstrated the tuning of lasing wavelength by applying an electric field. © *2006 American Institute of Physics*.
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Liquid crystal is a phase of matter with the property between that of liquids and solids. It has been actively studied as optical devices such as displays. Especially, liquid crystals including chiral molecules such as cholesteric liquid crystals, ferroelectric liquid crystals (FLCs), and cholesteric blue phases have recently attracted much attention as self-organized periodic structures. These liquid crystals have chirality in their molecular structure, and spontaneously form helical periodic structures with a periodicity of light wavelength. Light propagating in the helical periodic structure is selectively reflected depending on the polarization states if the light wavelength matches the optical pitch of the helical structure, which can be considered as pseudoband gap. At the edge of the stop band, a photon density of state and a photon dwell time in the helical periodic structure are enhanced, which are higher and longer than that in a layered periodic dielectric media without helix.[1] Therefore low-threshold laser is expected in such liquid crystals, and laser actions in the dye-doped liquid crystals with chirality have been recently reported.[2–8] Especially, FLCs form tilted chiral periodic structures with spontaneous polarizations, and easily deformed upon applying electric field with a fast response time.[9] Namely, a fast modulation of lasing wavelength can be expected in FLCs.[5–7] However, as the lasing thresholds of the FLCs so far reported were high, the considerations of molecular structures of the FLCs or resonator structures are needed for the low-threshold laser.

In this letter, we have investigated laser action in FLC sandwiched between dielectric multilayers and achieved to lower the lasing threshold compared with simple FLC without dielectric multilayers. We also demonstrate the tuning of lasing wavelength by applying an electric field.

Figure 1 shows the schematic structure of FLC sandwiched between dielectric multilayers. A dielectric multilayer, which was consisted of five pairs, alternately stacked the SiO_2 and TiO_2 layers, deposited on a glass substrate. The refractive indices of the SiO_2 and TiO_2 are 1.46 and 2.35, and their thicknesses are 103 and 64 nm, respectively. The center wavelength of the stop band was adjusted to be 600 nm, and an excitation wavelength for lasing (532 nm) was out of the stop band. In order to apply an in-plane electric field in the FLC layer, Cr–Au electrodes were evaporated with 2 mm separation on the multilayer. The top surface of the dielectric multilayer was coated with polyimide (JSR, JALS-2021-R2) in order to obtain a homeotropic alignment. In this configuration, an electric field is applied perpendicular to the FLC helical axis.

The FLC compound used in this study was a multicomponent mixture having the chiral smectic C (SmC^*) phase in a wide temperature range including a room temperature. As a laser dye dopant, [2-[2-4(dimethylamino)pheny1]etheny1]-6-methy1-4H-pyran-4-ylidene propanedinitrile (Exciton) was doped in the FLC, whose concentration was 0.76 wt %. The FLC was sandwiched between the multilayers using 16 μm spacers, as the helical axis was perpendicular to the multilayer surfaces. In order to make comparison, emission characteristics of simple FLC cell without the multilayer were also investigated.

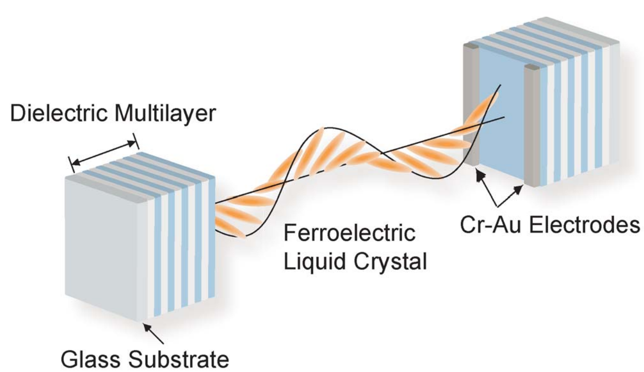

FIG. 1. (Color online) FLC sandwiched between dielectric multilayers. Electrodes on multilayer surface allow to apply the electric field perpendicular to the helical axis.

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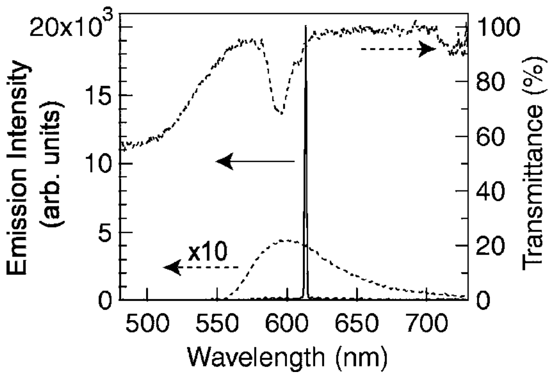

FIG. 2. Emission and transmission spectra of simple FLC (dashed line) and emission spectrum of FLC sandwiched between dielectric multilayers.

First of all, we have investigated transmission and emission spectra of simple FLC without dielectric multilayers, which are shown with the dashed lines in Fig. 2. The detailed experimental setup was described in Ref. 10. In the transmission spectrum, a drop of transmittance was observed at 595 nm, which was due to the stop band of the FLC. Decreasing of the transmittance at the shorter wavelength was attributed to absorption of the laser dye. The emission spectrum at the pump energy of 54 mJ/cm^2 pulse is shown with the dashed line, which was dominated by broad spontaneous emission of the laser dye. The emission intensity increased as the pump energy increased; however, laser action was not observed at any pump energies up to the damage threshold of 580 mJ/cm^2 pulse. This result indicates that the lasing threshold is higher than the damage threshold in this material.

We have investigated emission characteristics of FLC sandwiched between dielectric multilayers, as shown in Fig. 1. The solid line in Fig. 2 shows the emission spectrum of the FLC with the dielectric multilayers at the pump energy of 1.4 mJ/cm^2 pulse. Although many emission peaks appeared at low pump energy, above the threshold, a sharp emission line was observed, as shown in Fig. 2, at 614 nm which corresponds to the long wavelength band edge of the FLC. Figure 3 shows the peak intensity and the full width at half maximum (FWHM) of the emission spectrum as a function of pump energy. Above the pump energy of 600 $\mu J/cm^2$ pulse, the emission intensity increased drastically. Simultaneously, the FWHM of the emission peak decreased from 5 nm to less than the spectral resolution of 2 nm. This result indicates single-mode laser action occurred above the threshold of 600 $\mu J/cm^2$ pulse. Note that the lasing threshold is 10^3 times lower than that of the simple FLC without the multilayer, which must be attributed to the sandwich structure with the dielectric multilayers.

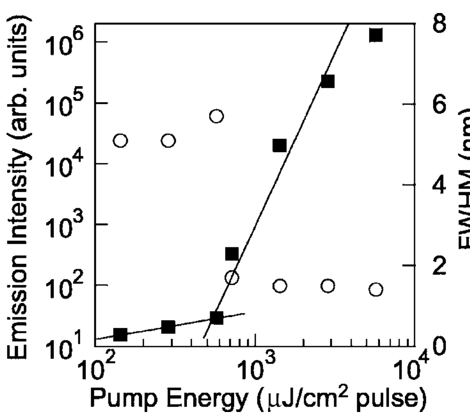

FIG. 3. Emission intensity and FWHM as a function of pump energy.

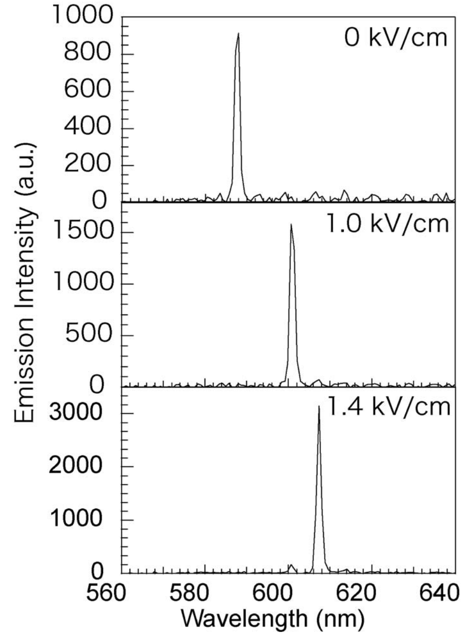

FIG. 4. Emission spectra as a function of applied voltage.

So far, we have investigated a one-dimensional photonic crystal containing cholesteric liquid crystal as a defect. In the photonic crystal, defect modes appeared due to introducing the defect into the photonic crystal. Among defect modes, we found a peculiar defect mode with high Q factor at the band edge of cholesteric liquid crystal. The Q factor of the peculiar defect mode is much higher than that of simple cholesteric liquid crystal because of double optical confinement of the band edge effect of the cholesteric liquid crystal and the light localization effect based on the defect mode.[10,11] We also investigated laser action in the photonic crystal and observed the single-mode laser action based on the peculiar defect mode with the high Q factor at the band edge of cholesteric liquid crystal. The lasing threshold was lower than that of the simple cholesteric liquid crystal because of the high Q factor due to the double optical confinement of not only the band edge effect but also the light localization effect.[12]

Same optical effect should work in the case of the FLC sandwiched between dielectric multilayers although the FLC layer was too thick to be considered as a defect. That is to say, this laser action was based on double optical confinement of not only the band edge effect of the FLC but also the light localization effect of the photonic crystal composed of the dielectric multilayers with the defect. Addition of the optical confinement by the dielectric multilayer to that by the FLC must lead to lower the lasing threshold of the FLC sandwiched between the dielectric multilayers compared with that of the simple FLC.

Figure 4 shows the emission spectra at pump energy of 7.5 mJ/cm^2 pulse, which is above the lasing threshold, as a function of an applied electric field. Under the applied electric field of 0.5 kV/cm, the laser action was observed at the wavelength of 588 nm. The difference of lasing wavelength at 0 V/cm between Figs. 2 and 4 was due to the difference of a measurement temperature. Above 0.5 kV/cm, the lasing wavelength shifted toward longer wavelength with increasing applied electric field, whose shift range was 19 nm. The emission peak intensity increased with redshift of the lasing wavelength. It might be related to spontaneous emission of the doped dye, which shows the peak of the emission efficiency at 610 nm.

FLC has spontaneous polarization which points normal to the molecules and parallel to the smectic layers. When an electric field is applied perpendicular to the helical axis, the FLC molecules intend to orient to the direction normal to the field because the polarization intends to point along the field direction. It causes the deformation of the helix and the elongation of the helical pitch. Therefore, the stop band shifts toward longer wavelength with increasing applied electric field. The shifted lasing wavelength upon the electric field (Fig. 4) corresponds to the long wavelength band edge of the FLC deformed by the electric field. This result supports that the low-threshold laser action in the FLC with the multilayers was attributed to not only the addition of the dielectric multilayers but also the band edge effect of the FLC.

In conclusion, we achieved single-mode laser action in FLC sandwiched between dielectric multilayers at the band edge of the FLC. The lasing threshold was thousand times lower than that of simple FLC because of the double optical confinement effect of the FLC and the dielectric multilayers. We also achieved the tuning of lasing wavelength by applying an electric field.

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