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Laser Action Based on Electrically Controllable Defect Mode in One-Dimensional Photonic Crystal Containing Conducting Polymer and Liquid Crystal Defect Layers

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Electrical tuning of the wavelength of a defect mode lasing in a one-dimensional photonic crystal has been demonstrated using a conducting polymer as an active emission layer and a nematic liquid crystal as an electrically tunable defect layer in the periodic structure. Lasing wavelength is widely tuned upon applying the electric field, which follows a defect mode shift due to the refractive index change in the nematic liquid crystal defect layer caused by field-induced realignment of the liquid crystal molecules.

Keywords: conducting polymer; defect mode; lasing; photonic crystal

INTRODUCTION

Photonic crystal (PC) having a three-dimensional (3D) ordered structure with a periodicity of optical wavelength has attracted considerable attention from both fundamental and practical points of view, because in such materials, a photonic band gap (PBG) exists in which the existence of a certain energy range of photon is forbidden, and various applications of PCs have been proposed [1,2]. In particular, the study of a defect mode in PBG is one of the most attractive subjects, since photons are localized and a low-threshold laser based on

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the defect mode of the PCs should be expected. However, complete 3D PCs with a periodicity equivalent to visible wavelength remain a technical challenge [3].

On the other hand, liquid crystals (LCs) have a large optical anisotropy and are sensitive to an external stress such as an electric field. Based on such optical anisotropy and field sensitivity, tunable PCs have been proposed in opal or inverse opal infiltrated with LC [4–8]. Although opal and inverse opal are simple and inexpensive approaches to realize 3D PC using self-organization of colloidal particles [9,10], the introduction of a defect into the 3D periodic structure is one of big problems to be resolved.

Not only 3D PC but also one-dimensional (1D) PC is an attractive subject. Recently, we have introduced a nematic LC (NLC) layer in a dielectric multilayer structure as a defect in the 1D PC, in which the wavelength of defect modes were controlled upon applying electric field based on the change in optical length of the defect layer caused by the field-induced molecular reorientation of the NLC [11]. We have proposed a novel technique of high speed electrooptic switching based on the tunable defect mode in the 1D PC with NLC defect [12]. Furthermore, the modulation of defect mode lasing upon applying a low voltage has been demonstrated in the 1D PC with a dye-doped NLC defect [13]. In this laser system, the solubility of the dye to LC was very important. In order to resolve the solubility problem of the dye, the emission layer should be separated from the LC defect as modulation layer.

In this study, a conducting polymer, which has highly extended π-conjugated systems in main chains and is expected as a possible high gain medium for laser application [14–18], is used as the emission layer, and a wavelength tunable laser based on an electrically controllable defect mode is demonstrated in the 1D PC including the conducting polymer and NLC defect layers. The lasing wavelength can be tuned over a wide range upon applying an electric field.

EXPERIMENTAL

A schematic view of a 1D PC with a defect is shown in Figure 1. A dielectric multilayer consisting of an alternating stack of SiO_2 and TiO_2 layers deposited on an In-Sn oxide (ITO)-coated glass substrate was used as the 1D PC. The refractive indices of SiO_2 and TiO_2 are 1.46 and 2.35, respectively. In order to adjust the center wavelength of the stop band to 650 nm, the optical thickness of both SiO_2 and TiO_2 should be one-quarter of 650 nm, and the thickness of SiO_2 and TiO_2 layers are determined to be 111 nm and 69 nm, respectively.

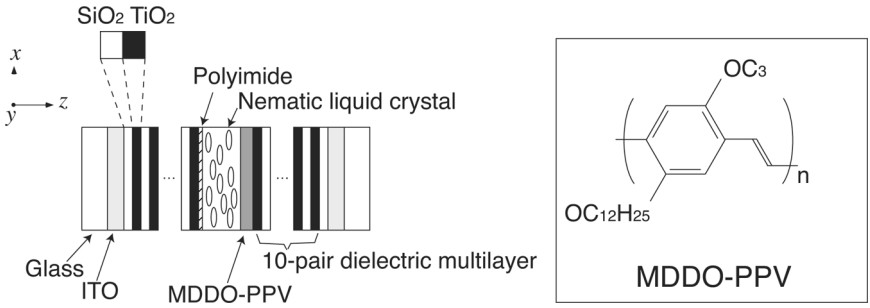

FIGURE 1 Schematic view of a one-dimensional (1-D) photonic crystal (PC) with a poly (2-methoxy-5-dodecyl-oxy-1, 4-phenylenevinylene) (MDDO-PPV) and nematic liquid crystal (NLC) defect layer.

The number of SiO_2-TiO_2 pairs on the substrate is ten. The 1D PC fabricated on above conditions has a stop band in a transmission spectrum between 570 nm and 750 nm.

A poly (2-methoxy-5-dodecyl-oxy-1, 4-phenylenevinylene) (MDDO-PPV) is used as an active material for the emission. A MDDO-PPV layer was coated on the top surface of the 1D PC by spin-coating from a chloroform solution of the polymer. The thickness of the MDDO-PPV layer was approximately 200 nm.

For the introduction of the NLC defect layer, an NLC (Merck, E47) was sandwiched between two substrates with dielectric multilayers using 2-μm spacers. The refractive index anisotropy Δn of E47 is 0.209 at room temperature and the dielectric anisotropy $\Delta\varepsilon$ is positive. In order to align the NLC molecules, one of dielectric multilayers, which is uncoated with MDDO-PPV, is coated with a polyimide (Japan Synthetic Rubber, AL1254) and unidirectionally rubbed along the x-axis shown in Figure 1. In the absence of an electric field, the long molecular axis of the NLC aligns parallel to the substrate (x-axis).

The transmission spectra were measured by a charge-coupled device (CCD) multichannel spectrometer (Hamamatsu Photonics, PMA-11) placed on the opposite side of the cell. Resolution of the CCD multichannel spectrometer is 3.0 nm. A halogen lamp was used as a light source. A second-harmonic light of a Q-switched Nd:YAG laser (Spectra Physics, Quanta-Ray INDI) is used for excitation, whose wavelength, pulse width and pulse repetition frequency are 532 nm, 8 ns and 10 Hz, respectively. Since the Nd:YAG laser beam of 532 nm as excitation light can transmit through the cell without the confinement of PBG appeared from 570 nm to 750 nm, the MDDO-PPV is excited efficiently. Nd:YAG laser beam for excitation irradiated the

sample perpendicularly to the cell plate and the illumination area on the sample is about 0.2 mm^2. In order to control the emission wavelength, the orientation of the LC molecules in the defect layer was gradually changed upon applying a rectangular wave voltage of 1 kHz.

RESULTS AND DISCUSSIONS

Figure 2 shows emission spectra of MDDO-PPV film spin-coated on a glass substrate without dielectric multilayer as a function of the excitation energy. The photoluminescence spectrum of the MDDO-PPV film appears in the wavelength range between 550 nm and 750 nm. Therefore, the emission light from the MDDO-PPV should be confined in the defect of the 1D PC. For a low excitation energy (0.52 μJ/pulse), the spectrum denoted by the dot line is dominated by a broad spontaneous emission. At a high excitation energy (26 μJ/pulse), a narrow emission spectrum denoted by the solid line appears. The full width at half maximum (FWHM) of emission peaks at low and high excitation energies are about 50 nm and 10 nm, respectively. This spectral narrowing upon high energy excitation is common in π-conjugated polymers and is attributed to an amplified spontaneous emission (ASE).

Figure 3 shows the transmission spectrum of the 1D PC with the PPV-LC defect. PBG is observed in the spectral range between

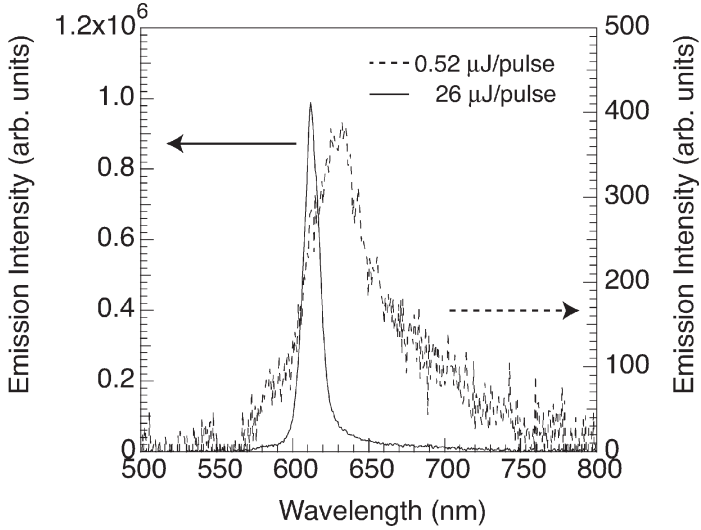

FIGURE 2 Photoluminescence spectrum of the MDDO-PPV as a function of the excitation energy.

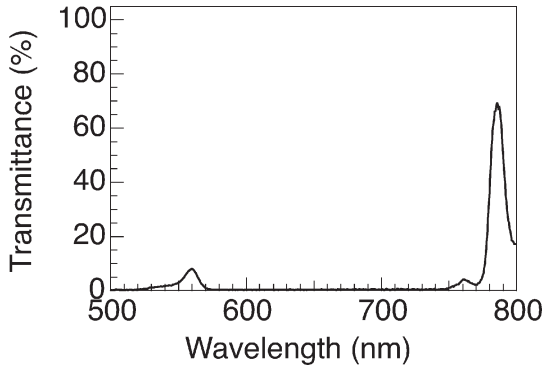

FIGURE 3 Transmission spectrum of the 1D PC with PPV-LC defect.

570 nm and 750 nm. The suppression of the transmittance at shorter wavelengths (< 570 nm) is attributed to the absorption of the MDDO-PPV. The transmittances of defect mode peaks in the PBG were less than 1%. This result indicates that photons of the defect mode wavelengths were strongly confined in the defect layer. Consequently, defect modes originating from the PPV-LC defect were hardly observed in the transmission spectrum.

Figure 4 shows the emission spectra of the 1D PC with the PPV-LC defect at low excitation energy. The excitation energy for the emission is 325 nJ/pulse. As is evident from Figure 4, mainly four defect mode peaks (P_1, P_2, P_3, P_4) appear in the PBG of the 1D PC with the PPV-LC defect. It should be noted these peaks are classified into two groups. P_2 and P_4 shift as the voltage increases, while the positions of P_1 and P_3 are independent of the voltage. The light field of the defect mode is localized in the defect layer and the field profile is sensitive to the optical length of the defect layer. Therefore, the spectral position of the defect mode should strongly depend on the refractive index of the defect layer. The lights propagating through the LC defect layer feel extraordinary refractive index n_e and ordinary refractive index n_o of the LC as an eigen mode. P_2 and P_4 correspond to the light propagating feeling n_e of the LC in the initial state (0 V), because the LC molecules align parallel to the x-axis shown in Figure 1. However, due to a positive dielectric anisotropy $\Delta\varepsilon$ of the LC used in this study, under the field above Fredericks threshold, the direction of LC molecular alignment changes and the refractive index decrease with increasing voltage. Finally, at 10 V the light feels nearly n_o of the LC which is smaller by 0.209 compared with n_e. As a result, the wavelengths of P_2 and P_4 shift upon applying voltage. On the other hand, peaks P_1 and P_3

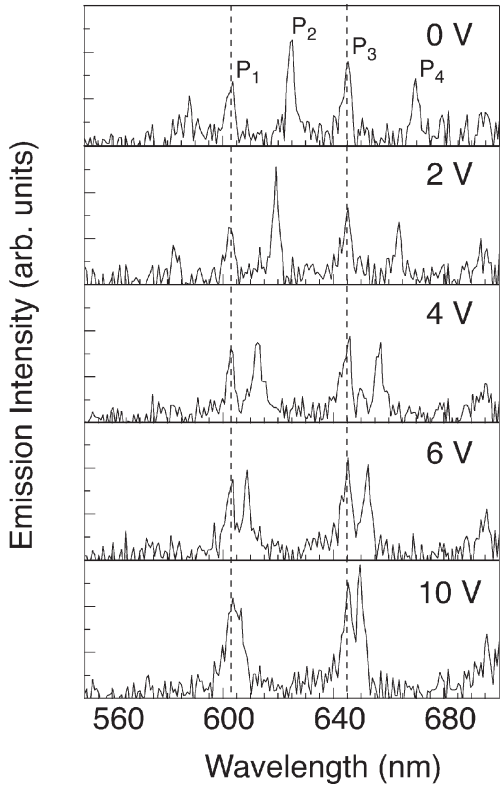

FIGURE 4 Emission spectra of the 1D PC with PPV-LC defect at a low excitation energy (325 nJ/pulse).

marked dotted lines in Figure 4 does not shift. This indicates that the lights propagating through the defect layer which correspond to P_1 and P_3 always feel n_o of the LC. Therefore, the optical length of the defect layer is independent of the molecular orientation of the LC and the profile of the spectrum does not change upon applying the electric field.

Figure 5 shows the emission spectra of the 1D PC with the PPV-LC defect as a function of the applied voltage. The excitation energy for the emission is 1.3 μJ/pulse. In order to eliminate light polarizing along the *y*-axis, polarizer is set in front of the detector of CCD multichannel spectrometer. A sharp emission peak appears above the threshold excitation energy (650 nJ/pulse) as shown in Figure 5. The FWHM of the emission peaks is about 2.0 nm, which is limited by a spectral resolution of our experimental setup. These peak widths are much narrower than that of ASE emission spectrum of the

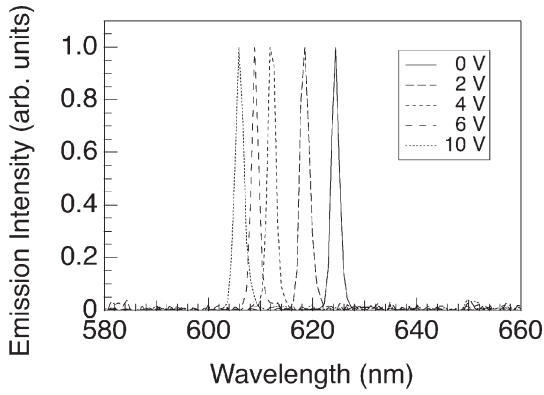

FIGURE 5 Emission spectra of the 1D PC with PPV-LC defect at a high excitation energy (1.3 μJ/pulse) as a function of applied voltage.

MDDO-PPV without 1D PC (10 nm) at a high excitation energy (26 μJ/pulse) shown in Figure 2. This indicates that these sharp emissions are due to a laser action. It should be noted that the lasing peak shifts toward shorter wavelengths with increasing voltage. The wavelength shift of the lasing peak is about 20 nm, even upon applying low voltage. This shift follows the defect mode shift, which originates from the decrease in the optical length of the NLC defect layer caused by the field-induced reorientation of the NLC molecules.

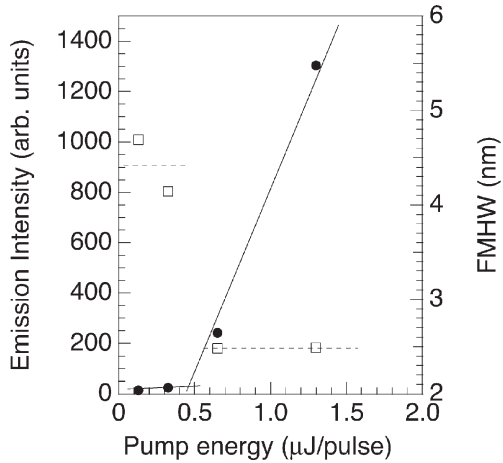

FIGURE 6 Excitation energy dependence of the peak intensity and the line width.

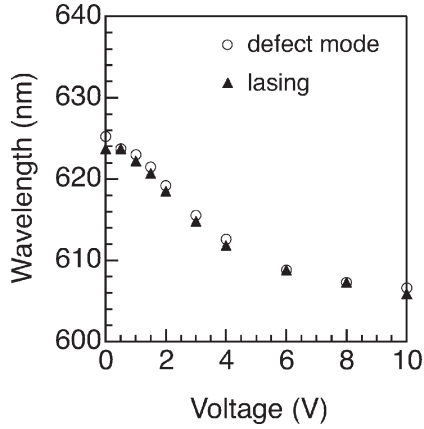

FIGURE 7 Voltage dependence of the defect mode and the lasing wavelength.

Figure 6 shows the excitation energy dependence of the peak intensity and FWHM at 10 V. Above the threshold at a excitation pulse energy of about 650 nJ/pulse, the emission intensity drastically increases and FWHM decreases. The FWHM of the emission spectrum is about 2 nm, which is limited by a spectral resolution of the CCD spectrometer used in this experiment. The FWHM below the threshold might be suppressed by the spectral width of the defect mode which is shown in Figure 4. These results indicate the sharp emission spectrum under applying voltage originates from the laser action.

Figure 7 shows voltage dependence of the lasing peak wavelength. As is evident from Figure 7, a threshold voltage exists for the peak shift and the lasing wavelength shifts toward shorter wavelengths above the threshold of 1.0 V. This is associated with Fredericks transition of the LC in the defect layer. It should be noted that the voltage dependence of the lasing peak position agrees well with that of the defect mode peak. This confirms that the wavelength tuning upon applying voltage is based on the defect mode shift caused by the molecular reorientation in the defect layer.

CONCLUSIONS

We demonstrated the electrical tuning of the defect mode lasing in a 1D PC PBG by using a conducting polymer and a nematic liquid crystal defect layers. Laser emission was observed upon the irradiation of excitation laser beam above the threshold energy. Lasing wavelength was widely tuned with a low voltage.

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