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Electrically tunable lasing based on defect mode in one-dimensional photonic crystal with conducting polymer and liquid crystal defect layer

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Electrical tuning of the wavelength of the 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. © *2004 American Institute of Physics.* [DOI: 10.1063/1.1686891]

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,[1,2] and various applications of PCs have been proposed. 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 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, a tunable PC has been proposed in opal or inverse opal infiltrated with LC.[4–8] Although opal and inverse opal are simple and inexpensive approaches to realize a 3D PC using self-organization of colloidal particles,[9,10] the introduction of a defect into the 3D periodic structure is a problem that must be resolved.

Not only 3D PCs but also one-dimensional (1D) PCs are an attractive subject. Recently, we have introduced a nematic LC (NLC) layer in a dielectric multilayer structure as a defect in a 1D PC, in which the wavelength of defect modes was 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] 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.[12] In this 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,[13–15] 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.

The schematic structure of the 1D PC with the defect is shown in Fig. 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 and 69 nm, respectively. The number of SiO_2–TiO_2 pairs on the substrate is ten. The 1D PC fabricated on the above-mentioned conditions has a stop band in a transmission spectrum between 550 and 770 nm.

A poly (2-methoxy-5-dodecyl-oxy-1, 4-phenylene-vinylene) (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 in-

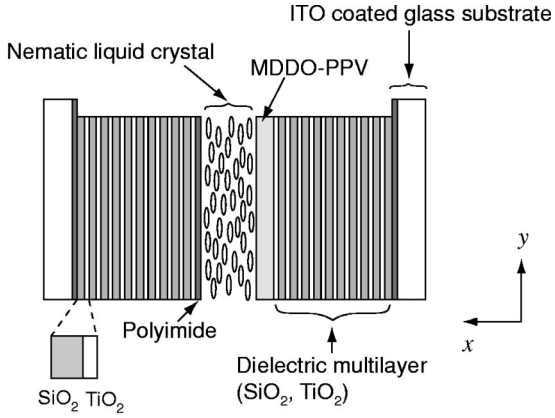

FIG. 1. Schematic view of a one-dimensional (1D) photonic crystal (PC) with a poly (2-methoxy-5-dodecyl-oxy-1, 4-phenylenevinylene) (MDDO-PPV) and nematic liquid crystal defect layer.

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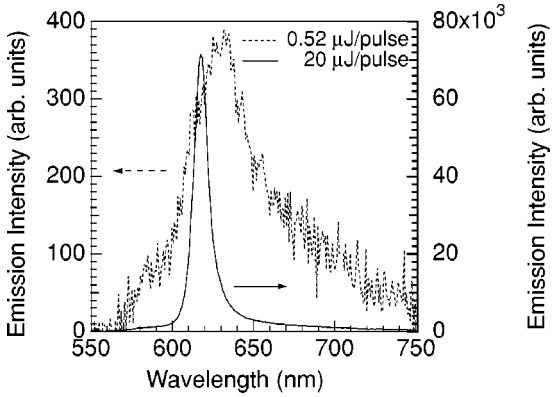

FIG. 2. Photoluminescence spectrum of the MDDO-PPV as a function of the excitation energy.

dex 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 the dielectric multilayers, which is uncoated with MDDO-PPV, is coated with a polyimide (Japan Synthetic Rubber, AL1254) and unidirectionally rubbed along the *y* axis shown in Fig. 1. In the absence of an electric field, the long molecular axis of the NLC aligns parallel to the substrate (*y* 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. A halogen lamp was used as a light source. For the measurement of lasing emission spectra with a higher resolution, a spectrograph (Oriel, MS257) with a CCD detector having spectral resolution of 0.5 nm was used.

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.

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 changed upon applying a rectangular wave voltage of 1 kHz.

Figure 2 shows emission spectra of MDDO-PPV film spin-coated on a glass substrate as a function of the pump energy. The photoluminescence spectrum of the MDDO-PPV film appears in the wavelength range between 550 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 (20 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 and 10 nm, respectively. This spectral narrowing upon high energy excitation is common in π-conjugated polymers[16–19] and is attributed to an amplified spontaneous emission (ASE).

Figure 3(a) shows the transmission spectrum of the 1D PC with the PPV-LC defect. PBG is observed in the spectral range between 550 and 770 nm. The suppression of the transmittance at shorter wavelengths ($<$550 nm) is attributed

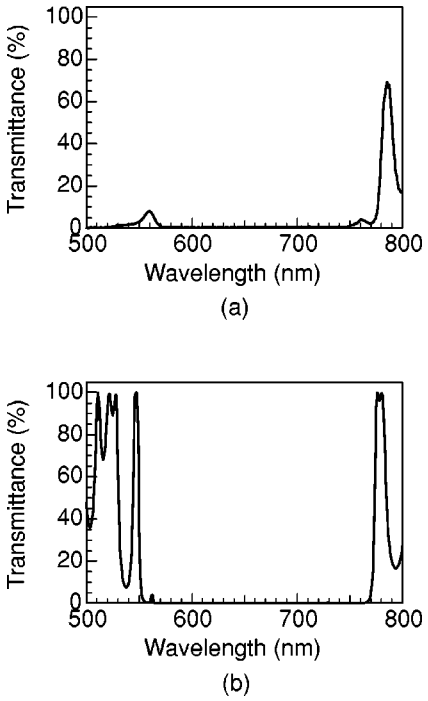

FIG. 3. (a) Transmission spectrum of the 1D PC with a PPV-LC defect. (b) Theoretical transmission spectrum of the 1D PC with a PPV-LC defect calculated by a 4×4 matrix method.

to the absorption of the MDDO-PPV. Regardless of the introduction of the PPV-LC defect layer in the 1D PC, defect mode peaks were not observed in PBG. For a theoretical verification of the above-noted result, we have performed a theoretical calculation of the light propagation in the 1D PC with the PPV-LC defect using characteristic matrices.[20] In the calculation, the absorption of PPV was not taken into account. Figure 3(b) shows the calculated transmission spectrum of the 1D PC with the PPV-LC defect. The theoretical calculation indicates the appearance of the PBG in the transmission spectrum, which agrees with the experimental result shown in Fig. 3(a). However, 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 recognized in the transmission spectrum.

We have also analyzed the distribution of the electric field in the 1D PC with the PPV-LC defect using a finite difference time domain calculation. This calculation is an analysis of the Maxwell equations by according to the Yee algorithm in discrete time and lattices.[21] Figures 4(a) and 4(b) show electric field distribution in the 1D PC with the defect for a defect mode and a PBG wavelength, respectively. Although, in both cases, the incident light hardly transmitted through the 1D PC with PPV-LC defect, at the defect mode wavelength, a standing wave appears in the defect layer as shown in Fig. 4(a). On the contrary, at the PBG wavelength, the defect layer does not act effectively for the light propagation and the incident light simply decreases. Namely, the characteristics of the light propagation in the 1D PC with PPV-LC defect are different between the defect mode and the PBG wavelengths.

Figure 5 shows the emission spectra of the 1D PC with

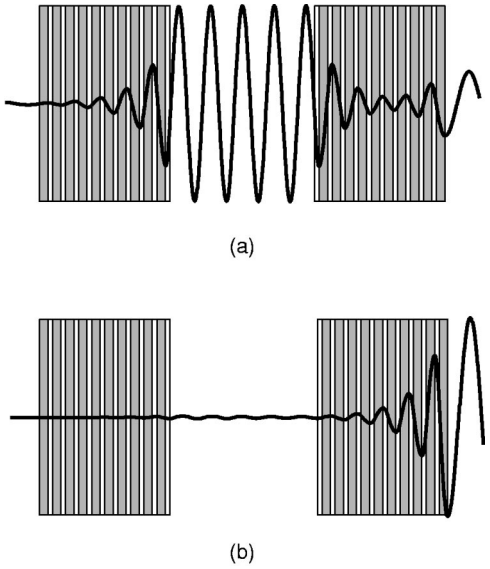

FIG. 4. A distribution of the electric field calculated by a finite difference time domain (FDTD) method (a) at a defect mode wavelength and (b) at a photonic bad gap wavelength.

the PPV-LC defect as a function of the applied voltage. The excitation energy for the emission is 130 nJ/pulse. A sharp emission peak appears above the threshold excitation energy (26 nJ/pulse) as shown in Fig. 5. The FWHM of the emission peaks is about 0.5 nm, which is limited by a spectral resolution of our experimental setup. These narrow peak widths are much narrower than that of ASE emission spectrum of the MDDO-PPV without 1D PC (10 nm) at a high excitation energy (20 μJ/pulse) shown in Fig. 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 15 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 shows the pump energy dependence of the peak intensity at 5 V. Although, at lower pump energy, emission spectra cannot be observed due to the strong confinement of the photons at the defect, there is a clear threshold ($\sim$26 nJ/pulse) for the intense emission. This result also supports that the sharp emission spectrum under applying voltage originates from the laser action.

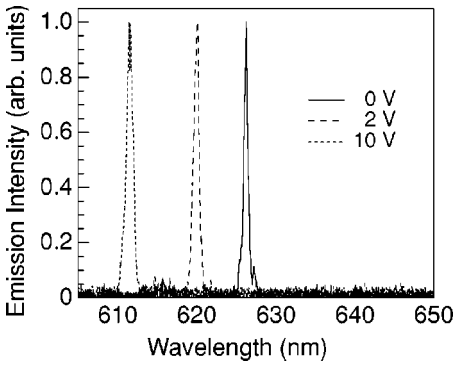

FIG. 5. Emission spectra of the 1D PC with PPV-LC defect as a function of applied voltage.

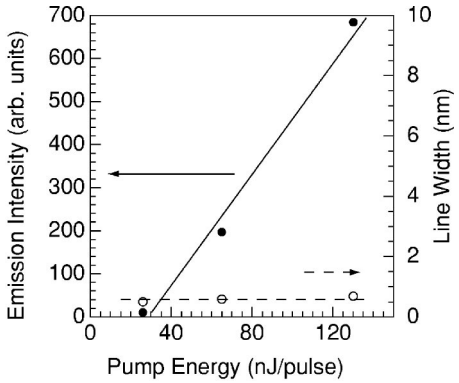

FIG. 6. Pump energy dependence of the peak intensity and the linewidth.

In conclusion, 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. The calculated transmission spectrum agreed well with the experimental one. At the defect mode wavelength, peak intensity was very weak. However, a standing wave in the defect layer was confirmed by calculating the distribution of the electric field. By pumping MDDO-PPV defect layer in the 1D PC, the sharp lasing spectra were observed and lasing wavelength was widely tuned upon a low voltage.

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