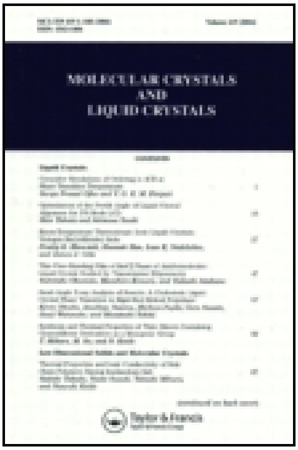

Molecular Crystals and Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl20

Wavelength-Variable Laser in a Hybrid Photonic Crystal Containing Ferroelectric Liquid Crystal

Yuko Matsuhisa [a], Akihiko Fujii [a], Masanori Ozaki [a], Wolfgang Haase [b] & Katsumi Yoshino [c d]

[a] Department of Electrical, Electronic and Information Engineering, Graduate School of Engineering, Osaka University, Suita, Osaka, Japan

[b] Institute of Physical Chemistry, Darmstadt University of Technology, Darmstadt, Germany

[c] Research Project Promotion Institute, Shimane University, Matsue, Shimane, Japan

[d] Collaborative Research Center for Advance Science and Technology, Osaka University, Yamada-oka, Suita, Osaka, Japan

Version of record first published: 22 Sep 2010

Mol. Cryst. Liq. Cryst., Vol. 477, pp. 245/[739]–254/[748], 2007

ISSN: 1542-1406 print/1563-5287 online
DOI: 10.1080/15421400701735616

Wavelength-Variable Laser in a Hybrid Photonic Crystal Containing Ferroelectric Liquid Crystal

Yuko Matsuhisa
Akihiko Fujii
Masanori Ozaki
Department of Electrical, Electronic and Information Engineering, Graduate School of Engineering, Osaka University, Suita, Osaka, Japan

Wolfgang Haase
Institute of Physical Chemistry, Darmstadt University of Technology, Darmstadt, Germany

Katsumi Yoshino
Research Project Promotion Institute, Shimane University, Matsue, Shimane, Japan; Collaborative Research Center for Advance Science and Technology, Osaka University, Yamada-oka, Suita, Osaka, Japan

We have investigated laser action in a hybrid photonic crystal (HPC) composed of ferroelectric liquid crystal (FLC) and dielectric multilayers. Single-mode laser action was observed in the HPC, which was based on a defect mode at the band edge of the FLC. The lasing threshold of the HPC became a thousandth lower than that of the simple FLC without the multilayers. The theoretical calculations of optical characteristics of the HPC and the simple FLC were performed, which was in good agreement with the experimental result. We have demonstrated tuning of the lasing wavelength by applying an electric field.

Keywords: ferroelectric liquid crystal; laser action; photonic crystal

This work is partially supported by a Grant-in-Aid for Scientific Research from the Ministry of Education, Culture, Sports, Science and Technology of Japan.
Address correspondence to Masanori Ozaki, Department of Electrical, Electronic and Information Engineering, Graduate School of Engineering, Osaka University, 2-1 Yamada-oka, Suita, Osaka 565-0871, Japan. E-mail: ozaki@opal.eei.eng.osaka-u.ac.jp

1. INTRODUCTION

There are various materials with riot of beautiful colors in nature. Some of them, such as opals, tail feathers of peacocks and so on, are attributed to the periodic structures with a periodicity in a range of optical wavelength. On the other hand, the advance of technology has recently enabled to artificially fabricate the periodic structures [1–3]. Such periodic structures have drawn much attention as photonic crystals (PCs) because of appearance of a photonic band gap (PBG). In the PBG, a certain energy range of photons is forbidden, which is attributable to Bragg reflection [4]. It is same as the forbidden electronic energy band for electrons in a crystal with the periodic potential.

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 PCs. These liquid crystals have chirality in their molecules, and spontaneously form helical periodic structures in an optical range. 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 a pseudo-PBG. Laser actions in such liquid crystals with a dye doped have been reported at the band edges, because the mediums work as distributed feedback cavities and photonic density of state (DOS) is enhanced [5–13]. Especially, FLCs form tilted chiral periodic structures, and are easily deformed upon applying an electric field with a fast response time [14]. Therefore, a fast modulation of the lasing wavelength can be expected in the FLCs [8–10]. However, the lasing threshold was high energy. Modifications of molecular structure of FLCs or resonator structures are needed for the low-threshold laser.

On the other hand, another interesting characteristic of PCs is a defect mode, which is caused in the PBG by the introduction of an imperfection into the PC [15]. At the defect mode, spontaneous emission is suppressed in the PBG and photons localize in the defect, which is due to amplification of the DOS. Hence, low-threshold lasers based on the defect mode are expected [16–18].

We have, so far, investigated a hybrid PC (HPC) which is composed of an inorganic PC and liquid crystal, and have proposed a tunable PC, in which a PBG and defect modes can be controlled [19–26]. In such tunable PCs, we have achieved the variable-wavelength laser actions. In this report, we have investigated laser action in a HPC containing FLC and dielectric multilayers fabricated with SiO_2 and TiO_2. The optical characteristics have been theoretically calculated to confirm the experimental result. We have also demonstrated tuning of the lasing wavelength by applying an electric field.

2. EXPERIMENTAL SETUP

Figure 1 shows a HPC containing FLC. A dielectric multilayer, consisting of five pairs of alternately stacked SiO_2 and TiO_2 layers, was deposited on a glass. 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 solid line in Figure 2 shows the transmission spectrum of the multilayer. A decreasing of the transmittance was observed in a spectral range from 550 nm to 700 nm, which coincides with the PBG. 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 the configuration, the electric field is applied perpendicular to the FLC helix 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. The extraordinary and ordinary refractive indices of the FLC are 1.71 and 1.53, respectively. The tilt angle and the helical pitch at a room temperature are 23.0° and 390 nm, respectively. The dashed line in Figure 2 shows the transmission spectrum of the FLC. A dip due to the PBG was observed at the wavelength of 594 nm, which was inside of the PBG of the multilayer. As a laser dye dopant, [2-[2-4(dimethylamino)pheny1]etheny1]-6-methy1-4H-pyran-4-ylidene propanedinitrile (Exciton, DCM) was doped in the FLC, whose concentration was 0.76 wt%. The FLC was

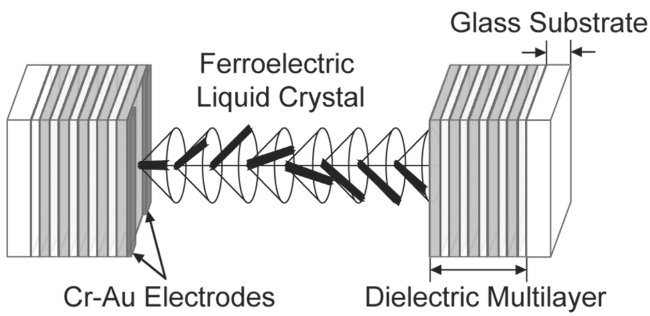

FIGURE 1 HPC containing FLC.

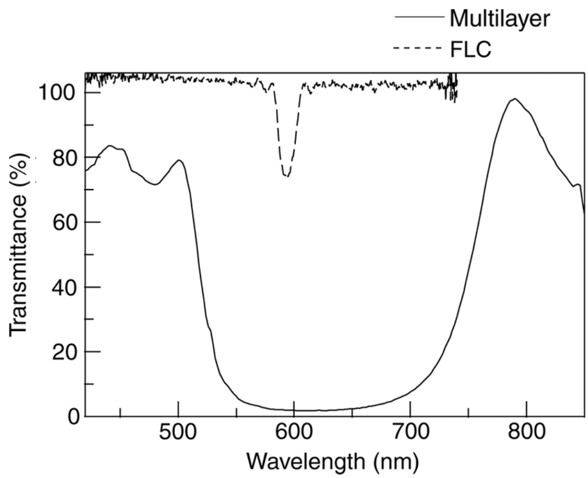

FIGURE 2 Transmission spectra of simple FLC and a dielectric multilayer.

injected between the multilayers using 18-μm spacer as the helical axis was perpendicular to the multilayers. In order to make comparison, emission characteristics of the simple FLC cell without the multilayers were also investigated.

The second harmonic light of a Q-switch Nd:YAG laser (Spectra Physics, Quanta-Ray INDI) was used for the excitation of the dye doped in the FLC, whose wavelength, pulse width and pulse repetition frequency were 532 nm, 8 ns and 10 Hz, respectively. The excitation laser beam irradiated the sample perpendicularly to the cell plate and was focused with a lens. The illumination area on the cell is about 0.2 mm^2. The emission spectra from the sample were measured from the backside of the sample using the CCD multichannel spectrometer having a spectral resolution of 2 nm (Hamamatsu Photonics, PMA-11) or that having a spectral resolution of 0.1 nm (Oriel, MS257).

3. EMISSION CHARACTERISTICS

First of all, we have investigated the emission characteristics of the FLC without the multilayers. Figure 3 shows the emission spectrum of the simple FLC at the excitation energy of 1.8 μJ/pulse. The emission spectrum was dominated by broad spontaneous emission. A decreasing in transmittance was observed at the wavelength of 600 nm, which coincides with the wavelength of the PBG of the FLC.

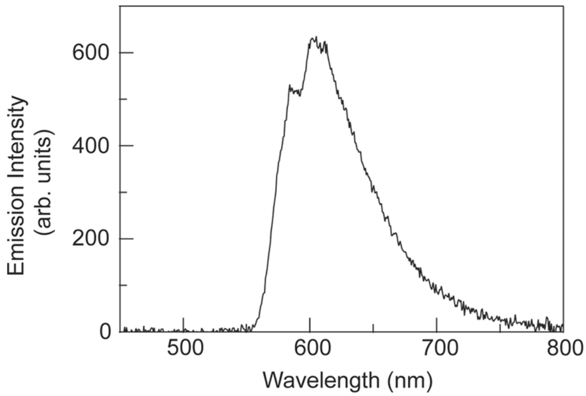

FIGURE 3 Emission spectrum of simple FLC.

The emission intensity increased as the excitation energy increased, however the laser action was not observed at any pumping energies up to the damage threshold.

Next, we have investigated the emission characteristics of a HPC containing FLC. Figure 4 shows the emission spectra of the HPC at the pumping energy of (a) 38 nJ/pulse and (b) 380 nJ/pulse. We observed several emission peaks at the low pumping energy, as shown in Figure 4(a). The emission peaks are related to defect modes originating from introduction of the FLC, as a kind of a defect, between the dielectric multilayers. This suggests that the emission peaks can be attributed to the spontaneously emitted light passing through narrow spectral windows owing to the defect modes. On the other hand, at the high pumping energy, one sharp emission line appeared at 605.8 nm, which coincides with one of the defect modes, as shown in Figure 4(b). Figure 5 shows the intensity and the width (the full width at half-maximum; FWHM) of the emission peak at 605.8 nm as a function of pumping energy. At the low pumping energy, the emission intensity increased as increasing the pumping energy. However, the emission intensity increased drastically above the threshold of 190 nJ/pulse. Simultaneously, the FWHM of the emission peak decreased from 0.4 nm to 0.1 nm which was the spectral limitation in this experimental setup. As a result, the sharp emission peak in Figure 4(b) was based on the laser action.

The dashed line in Figure 4(b) shows the transmission spectrum of the simple FLC without the multilayers. Note that the lasing wavelength of the HPC coincides with the particular defect mode at the band edge of the FLC, despite many defect modes existing. It indicated

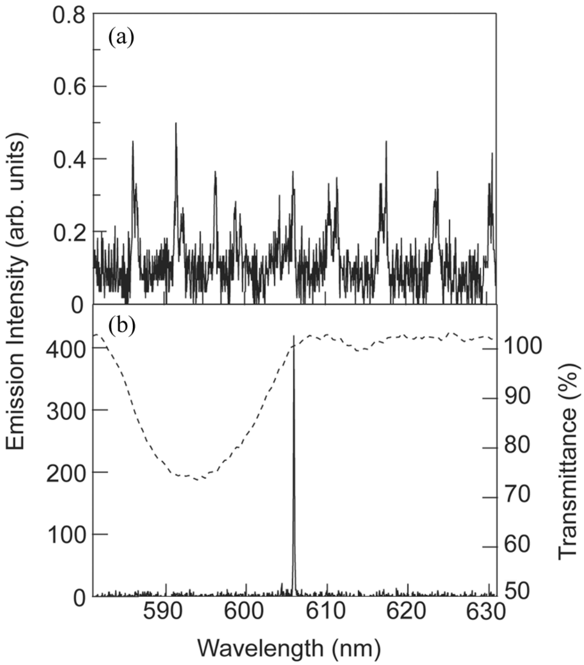

FIGURE 4 (a) Emission spectrum of HPC containing FLC at the pumping energy of 38 nJ/pulse. (b) Emission spectrum of HPC containing FLC at the pumping energy of 380 nJ/pulse and transmission spectrum of simple FLC.

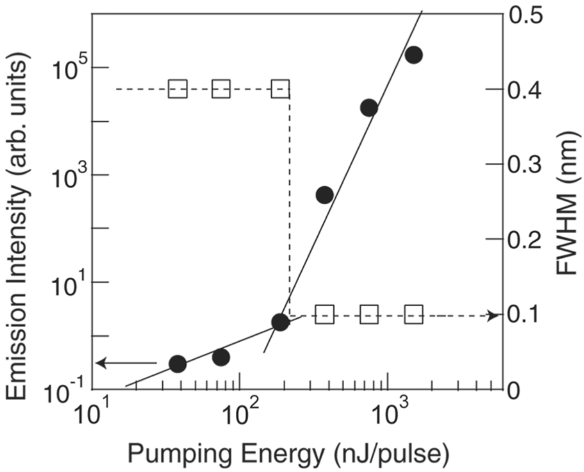

FIGURE 5 Emission intensity and FWHM of HPC as a function of pumping energy.

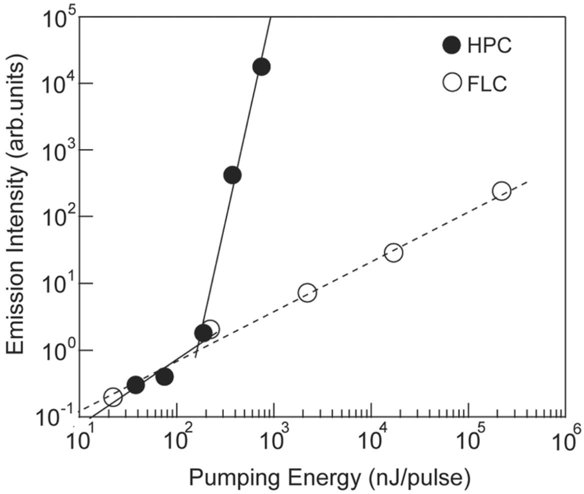

FIGURE 6 Emission intensity of simple FLC and HPC containing FLC as a function of pumping energy.

that this laser action originates from the double optical confinement of not only the band-edge effect of the FLC but also the light localization based on the defect mode of the dielectric multilayer PC.

We have compared the lasing threshold of the HPC containing the FLC with that of the simple FLC without the multilayers. Figure 6 shows the pumping energy dependence of the emission intensity in the HPC and the simple FLC. In the case of the simple FLC, the emission intensity increased as the excitation energy increased, however the laser action was not observed at any pumping energies up to the damage threshold of 1.0 mJ/pulse. This result indicates that the lasing threshold in the simple FLC is higher than the damage threshold in this system. On the other hand, in the HPC, the laser action was observed above the threshold of 190 nJ/pulse. Hence, the lasing threshold of the HPC became more than one thousand times lower than that of the simple FLC without the multilayers, which is attributed to the double optical confinement of both the FLC and the multilayers.

4. THEORETICAL CALCULATION

We have performed the theoretical calculation of optical characteristics of the simple FLC and the HPC containing the FLC by a method of 4×4 matrix [21]. Figures 7(a) and 7(b) show the calculated

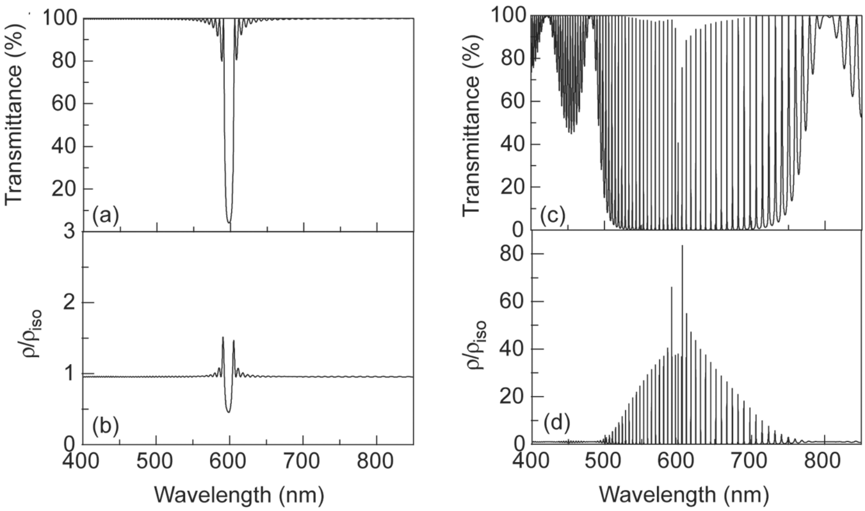

FIGURE 7 (a) Transmission spectrum and (b) DOS of simple FLC. (c) Transmission spectrum and (d) DOS of HPC containing FLC.

transmission spectrum and DOS of the simple FLC. The PBG was observed at the wavelength of 600 nm, and the DOS increased to 1.5 at the band edges of the PBG, 591.2 nm and 605.6 nm, which is attributed to the band-edge effect. On the other hand, the transmission spectrum and the DOS of the HPC are shown in Figures 7(c) and 7(d). In the transmission spectrum, many defect modes were observed in the PBG. The wavelengths of the defect modes corresponded to those of the emission peaks in Figure 4(a), which supports that the emission peaks in Figure 4(a) were caused by the defect modes. The DOS of the HPC increased at each defect mode. Especially, the DOS was markedly enhanced at the band edges of the FLC, which is much higher than the DOS of the simple FLC. Consequently, lowering the lasing threshold in the HPC can be explained by the high DOS at the band edges of the FLC.

5. LASING WAVELENGTH TUNING

Figure 8 shows the emission spectra at 2.3 μJ/pulse above the lasing threshold as a function of an applied electric field. The laser action was observed at the wavelength of 588 nm at 0 kV/cm. The lasing wavelength shifted toward the longer wavelength with increasing the applied electric field above the applied electric field of 0.5 kV/cm.

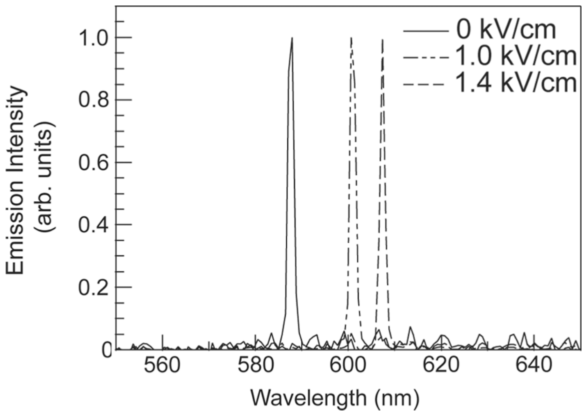

FIGURE 8 Emission spectra of HPC containing FLC as a function of applied electric field.

The FLC has a 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 PBG of the FLC shifts toward longer wavelength with increasing an applied electric field. The lasing wavelength of the HPC containing the FLC corresponded to the defect mode wavelength at the long-wavelength edge of the PBG of the FLC at any applied electric fields. Therefore, the red-shift of the lasing wavelength shown in Figure 8 originates from the shift of the PBG of the FLC caused by the field-induced elongation of the helix pitch. This result supports the single-mode laser action was contributed by the band-edge effect of the FLC.

6. CONCLUSION

We achieved single-mode laser action in the HPC containing the FLC at the band edge ofthe FLC. The lasing threshold in the HPC became a thousandth lower than that of the simple FLC because of the double optical confinement. By the theoretical calculation of optical characteristics of the HPC, the high DOS was observed at the band edge of the FLC, which was in good agreement with the experimental result. We also achieved tuning of the lasing wavelength by applying an electric field.

REFERENCES

[1] Yamamoto, N., Noda, S., & Sasaki, A. (1997). *Jpn. J. Appl. Phys.*, *36*, L1907.
[2] Assefa, S., Petrich, G. S., Kolodziejski, L. A., Mondol, M. K., & Smith, H. I. (2004). *J. Vac. Sci. Technol.*, B *22*, 3363.
[3] Fukuda, K., Sun, H., Matsuo, S., & Misawa, H. (1998). *Jpn. J. Appl. Phys.*, *37*, L508.
[4] John, S. (1987). *Phys. Rev. Lett.*, *58*, 2486.
[5] Il'chishin, I. P., Tikhonov, E. A., Tishchenko, V. G., Shpak, M. T., & John, (1980). *JETP. Lett.*, *32*, 24.
[6] Kopp, V. I., Fan, B., Vithana, H. K. M., & Genack, A. Z. (1998). *Opt. Lett.*, *23*, 1707.
[7] Funamoto, K., Ozaki, M., & Yoshino, K. (2003). *Jpn. J. Appl. Phys.*, *42*, 1523.
[8] Ozaki, M., Kasano, M., Ganzke, D., Haase, W., & Yoshino, K. (2002). *Adv. Mater.*, *14*, 306.
[9] Kasano, M., Ozaki, M., Yoshino, K., Ganzke, D., & Haase, W. (2003). *Appl. Phys. Lett.*, *82*, 4026.
[10] Ozaki, M., Kasano, M., Kitasho, T., Ganzke, D., Haase, W., & Yoshino, K. (2003). *Adv. Mater.*, *15*, 974.
[11] Cao, W., Munoz, A., Palffy-Muhoray, P., & Taheri, B. (2002). *Nat. Mat.*, *1*, 111.
[12] Schmidtke, J. & Stille, W. (2003). *Eur. Phys. J. B*, *31*, 179.
[13] Song, M. H., Ha, N. Y., Amemiya, K., Park, B., Takanishi, Y., Ishikawa, K., Wu, J. W., Nishimura, S., Toyooka, T., & Takezoe, H. (2006). *Adv. Mater.*, *18*, 193.
[14] Beresnev, L. A., Chigrinov, V. G., Dergachev, D. I., Poshidaev, E. I., Fünfschilling, J., & Schadt, M. (1989). *Liq. Cryst.*, *5*, 1171.
[15] Yablonovitch, E. (1987). *Phys. Rev. Lett.*, *58*, 2059.
[16] Noda, S., Chutinan, A., & Imada, M. (2000). *Nature*, *407*, 608.
[17] Ozaki, M., Ozaki, R., Matsui, T., & Yoshino, K. (2003). *Jpn. J. Appl. Phys.*, *42*, 472.
[18] Wu, X., Yamilov, A., Liu, X., Li, S., Dravid, V. P., Chang, R. P. H., & Cao, H. (2004). *Appl. Phys. Lett.*, *85*, 3657.
[19] Yoshino, K., Satoh, S., Shimoda, Y., kawagishi, Y., Nakayama, K., & Ozaki, M. (1999). *Jpn. J. Appl. Phys.*, *38*, 961.
[20] Yoshino, K., Shimoda, Y., Kawagishi, Y., Nakayama, K., & Ozaki, M. (1999). *Appl. Phys. Lett.*, *75*, 932.
[21] Shimoda, Y., Ozaki, M., & Yoshino, K. (2001). *Appl. Phys. Lett.*, *79*, 3627.
[22] Ozaki, R., Matsui, T., Ozaki, M., & Yoshino, K. (2002). *Jpn. J. Appl. Phys.*, *41*, L1482.
[23] Ozaki, R., Matsui, T., Ozaki, M., & Yoshino, K. (2003). *Appl. Phys. Lett.*, *82*, 3593.
[24] Ozaki, R., Ozaki, M., & Yoshino, K. (2003). *Jpn. J. Appl. Phys.*, *42*, L669.
[25] Matsuhisa, Y., Ozaki, R., Ozaki, M., & Yoshino, K. (2005). *Jpn. J. Appl. Phys.*, *44*, 629.
[26] Matsuhisa, Y., Ozaki, R., Yoshino, K., & Ozaki, M. *Thin solid film*s, *in press*.
[27] Berreman, D. W. (1973). *J. Opt. Soc. Am.*, *63*, 1374.