Design, simulation, and fabrication of longitudinal electro-optically tuned transparent-comb-electrode photonic crystals

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We designed advanced transparent-comb-electrode photonic crystals (TCE PCs) to use the longitudinal electro-optic (LEO) effect, so they are composed of Al-doped ZnO (AZO) layers, which serve as internal/external electrodes, and centered (Pb,La)(Zr,Ti)O3 (PLZT) layers, which serve as EO material. The simulated transmittance spectra of a multilayer interference (MLI) model revealed the conditions in the AZO and PLZT film thicknesses and the AZO/PLZT period number for a narrow photonic band gap (PBG). The TCE PCs consisted of four periods of AZO/PLZT, and it was fabricated by RF magnetron sputtering through metal mask patterning. Finally, a refractive index change induced by the LEO effect of the PLZT layers was accomplished by applying the voltage dependence of transmittance spectra that were well fitted with a revised (MLI) model considering light absorption and scattering. The PCs exhibit large refractive index changes of $1.3 \times 10^{-2}$ and $2.9 \times 10^{-2}$ at DC voltages of 4 and 8 V, respectively. The corresponding PBG shifts were 3.9 and 8.5 nm, implying high a PBG tunability of $\sim 1$ nm/V.

Key-words : Photonic crystals, Longitudinal electro-optic effect, Photonic band gap, Tunability, Multilayer model

1. Introduction

At the present day, the home network (i.e., the network between home and the outer field) is one of the bare essentials of daily life, and is growing at a rapid pace. To run a high-capacity home network with a lot of signals smoothly, more ultra-compact optical switches that enable high integration, simple fabrication, and low power consumption (i.e., low driving voltage) are strongly demanded. However, the conventional LiNbO$_3$ and (Pb,La)(Zr,Ti)O$_3$ optical switches, which use the electro-optic (EO) effect, are not reached until the end of the line, because their driving principle is a phase delay difference. Therefore, these optical switches are governed by the optical path length and refractive index change of EO materials, indicating a trade-off between size and driving voltage. This trade-off prevents to commercialize the low-driving-voltage ultra-compact optical switch. Moreover, researchers have reached the limit to the number of solutions for improving the EO properties of the material. Therefore, new optical modulation is needed to escape the trade-off.

In response to this demand, the researchers are presently interested in periodic nano-structured photonic crystals (PCs) as promising candidates for optical devices due to their integrable ultra-small size and special ability to manipulate photons. Among the many possible applications that PCs offer, the tunable photonic band gap (PBG) is especially desirable, because it allows optical modulation. The PBGs are fundamentally dependent on the refractive indices and thicknesses of the constituent materials. One effective way of achieving PBG tunability is to use ferroelectrics with an EO effect, because these ferroelectrics enable developers to vary refractive indices by simply applying voltages. In recent years, researchers have made great efforts to develop tunable PCs based on ferroelectrics. Indeed, many studies have reported the successful development of PCs with various geometries, but the PCs' two or three-dimensional structures are still limited by low PBG tunability or complicated processes.

On the other hand, one-dimensional (1D) PCs offer many advantages, such as simple fabrication, processability, and large areas. K. L. Jim et al. reported the 1D tunable PCs based on the (Ba,Sr)TiO$_3$ (BST)/MgO multilayer, but these ultra-compact optical devices are still not feasible due to their high driving voltage that results from their low PBG tunability. The EO type used in the PCs is based on a multilayer and has a transverse EO (TEO) effect, that is, the direction of light propagation and applied voltage is perpendicular. In order to apply voltage to the ferroelectric layers in this TEO type, two coplanar metal electrodes with a wide gap (20–60 $\mu$m) are generally used. Thus, very high voltage is required to obtain a large refractive index change in this type. In addition, the electric field is not uniform within each ferroelectric layer. Therefore, the development of an alternative is urgently required for achieving high PBG tunability.

In this report, we propose the advanced transparent-comb-electrode PCs (TCE PCs) based on the multilayer structure. We developed them through the viewpoint of device structure engineering, and then simulated the TCE PCs to find the suitable thickness and period number conditions for the constitution layers of a narrow PBG. After fabricating prototype TCE PCs and observing their microstructure, we evaluated their PBG tunability, which is induced by the EO effect of ferroelectric layers.

2. Device design

As shown in Fig. 1, the TCE PCs were designed as two comb-like sets of transparent conductive oxide (TCO) layers (A layers). They acted as an internal/external electrode and were connected to an anode/cathode. The centered layers were transparent ferroelectric oxide (TFO) layers (B layers). According to our understanding of the characteristics of the longitudinal EO (LEO) effect, we determined that the direction of light propagation and
applied voltage are parallel,\(^1\) which served as an insight into the design of the structure. This LEO type has high potential for obtaining a high refractive index change at a low voltage owing to this type’s short EO interaction distance (<1 µm), which is the same length as the EO film’s thickness. Moreover, its applied electric field is uniform across the TFO layers. Furthermore, the TCO films can be used both as electrodes and as one of the constituent materials in the TCE PCs, suggesting that the individual metal electrodes are no longer necessary. Results from our previous study also confirmed the high LEO effect and very high variations in the refractive indices of 100 preferentially oriented (Pb,La)(Zr,Ti)O\(_3\) (PLZT) films grown on Al-doped ZnO (AZO) films that were coated onto SrTiO\(_3\) (ST) substrates.\(^{14,15}\) Therefore, we selected TCO and TFO layers and substrates, AZO and PLZT films, and ST substrates for obtaining a large refractive index change at a low voltage to achieve high PBG tunability.

3. Simulations with the multilayer interference model

We simulated the designed TCE PCs based on the AZO/PLZT multilayer by using the multilayer interference (MLI) model, as shown in Fig. 2(a). P. Rouard introduced this method by which the derivation of the Fresnel coefficients for a single layer can be extended to any number of layers.\(^{16}\) Starting with the substrate and film 1 with Fresnel coefficients \(r_0, r_1, t_0, t_1\), we first computed the amplitude \(R_0\) and \(T_0\) of the light reflected and transmitted by the substrate as follows:

\[
N_i = n_i - ik_i
\]

\[
R_0 = \frac{r_1 + r_0 \exp\left(-\frac{4\pi N_1 d_0}{\lambda}\right)}{1 + r_1 r_0 \exp\left(-\frac{4\pi N_1 d_0}{\lambda}\right)}
\]

\[
t_1 + t_0 \exp\left(-\frac{2i\pi N_1 d_0}{\lambda}\right)
\]

\[
T_0 = \frac{1 + r_1 r_0 \exp\left(-\frac{4\pi N_1 d_0}{\lambda}\right)}{1 + r_1 r_0 \exp\left(-\frac{4\pi N_1 d_0}{\lambda}\right)}
\]

Here, \(N_i\) is the complex refractive index, \(n_i\) and \(k_i\) are respectively the refractive index and the extinction coefficient, \(d\) is the thickness of each layer, and \(\lambda\) is the wavelength. This model operates under the assumption that the films have no light scattering effect (i.e., have perfectly smooth interfaces) and no absorption [extinction coefficient \((k) = 0\). Thus, the total transmitted amplitude \(T_j\) and energy \(E_j\) can be calculated as follows:

\[
T_j = \frac{t_4 + T_{j-1} \exp\left(-\frac{2\pi N_j d_1}{\lambda}\right)}{1 + R_{j-1} t_4 \exp\left(-\frac{2\pi N_j d_1}{\lambda}\right)}
\]

\[
E_j = n_1 T_j T_j^* \quad (4)
\]

where \(n_1\) is the refractive index of the substrate, and \(T_j^*\) is the complex conjugate of \(T_j\). Films with perfectly smooth interfaces and no absorption do not exist outside textbooks, as shown in Fig. 2(b). The light scattered from the rough interfaces on a multilayer reduces the transmitted amplitude.\(^{17,18}\) A scalar scattering theory based on a randomly rough surface can be used to predict the effect of the roughness on the transmittance in terms of the root mean square (RMS) roughness at interfaces. The theory can be briefly expressed with the following equation:\(^{19}\)

\[
T_{\text{scatter}} = T_j - T_j \exp\left(-\frac{(4\pi \sigma \cos(\theta)/\lambda)^2}{2}\right) \quad (6)
\]

Here, \(T_{\text{scatter}}\) is the intensity of the scattered transmittance, \(\sigma\) is the total RMS roughness, and \(\theta\) is the incidence angle. By using a revised MLI model that considers the light scattering as calculated from RMS roughness as well as light absorption, we fitted the measured transmittance spectra of the TCE PCs based on the AZO/PLZT multilayer to our analysis of the ordinary complex refractive indices of AZO and PLZT films to obtain the applied voltage dependence of the refractive index variations induced by the LEO effect of PLZT films.

4. Experimental procedures

The AZO and PLZT films were fabricated using a conventional RF magnetron sputtering system (Seed Kab, Co) to optimize deposition conditions and thus achieve advanced preparation.\(^{14}\) Sputtered AZO (ZnO:Al\(_2\)O\(_3\) = 98:2) and PLZT composite targets\(^{20}\) were used. For the latter, a PbO pellet was placed at the center of the Pb\(_{0.95}\)La\(_{0.05}\)Zr\(_{0.65}\)Ti\(_{0.35}\)O\(_3\) [PLZT (9/65/35)] target with 10 mol% of excess PbO to supply a suitable amount of Pb to the PLZT films. ST single crystals (10 x 10 x 0.5 mm\(^2\), Shinkosha Ltd.) with both sides polished were also used as transparent substrates. The deposition chamber was evacuated to 6 x 10\(^{-6}\) Torr using a diffusion pumping system before deposition. The deposition of all films was carried out at 2 x 10\(^{-2}\) Torr using Ar as a sputtering gas. To achieve optimized deposition conditions, we heated the ST substrate at 600°C using a
kanthal A wire, and applied 150 W of RF power. The substrate-target distance was 3 cm. Then, based on the AZO/PLZT multilayer, the prototype TCE PCs was fabricated on a ST substrate through a metal shadow mask with square openings (6 × 8 mm$^2$; Fig. 3). First, the AZO layer was deposited on the ST substrate (AZO 1). After rotating the mask 90 degrees and moving it in a parallel direction, we deposited the PLZT layer (PLZT 2). Third, the mask was rotated another 90 degrees in the same direction and shifted along the opposite parallel direction to fabricate the AZO layer (AZO 3). The fourth step was solely a repeat of the second step for preparing the PLZT films (PLZT 4). The same four-step process was repeated to prepare four more periods of AZO/PLZT (AZO 5, PLZT 6, AZO 7 and PLZT 8).

After fabricating the TCE PCs, the cross-sectional image and surface morphology were observed using a field-emission scanning electron microscope (FE-SEM; JEOL JSM-6301F). The thicknesses and 1D RMS roughness values of the AZO and PLZT films were measured using the surface profilometer (Veeco Dektak150). Then, to confirm the ordinary refractive indices of AZO, PLZT films, and refractive index changes of PLZT films at applied voltage by using the MLI model, the transmittance spectra of the TCE PCs based on the AZO/PLZT multilayer were measured with a spectrophotometer (JASCO V-570).

5. Results and discussion

5.1 Transmittance spectrum simulations of TCE PCs with the AZO/PLZT multilayer

First, we simulated the transmittance spectra of the PCs based on the AZO/PLZT multilayer with 6 periods as a function of film thickness $d$ in order to find suitable conditions in AZO and PLZT film thickness and to find which AZO/PLZT period number achieves a narrow PBG with high switching sensitivity.\(^{21}\) Film 1 is AZO and film 2 is PLZT. The reference refractive indices that we used are $n_1 = 2.0^{22}$ and $n_2 = 2.4^{23}$ considering the ideal multilayer with no light absorption ($k_1 = k_2 = 0$) and no light scattering from interface roughness. While accounting for Snell’s law, we designated the thicknesses $d$ of the AZO and PLZT films as $m\lambda_s/4n$, where $m$ is a positive number at which the constructive interference occurs, and $\lambda_s$ is the central wavelength in PBG. The set value of the wavelength in this work was 1560 nm, which is widely used near the infrared range to achieve low optical loss in optical communication.\(^{24}\) When $m$ is changed from 1 to 5 (i.e., from 1 to 3 and then from 3 to 5), the thicknesses of AZO and PLZT films are 210 and 150 nm for $m = 1$, 630 and 450 nm for $m = 3$, and 1050 and 750 nm for $m = 5$, respectively. We selected the conditions with $m = 1$ and 3 for simulation, since the thickness summation of one period of AZO/PLZT is about 2 $\mu$m at $m = 5$, and we expected a long fabrication time for several periods of the AZO/PLZT multilayer. The results of the simulated transmittance spectra as functions of AZO and PLZT film thicknesses ($d_1$ and $d_2$) at $m = 1$ and 3 revealed that all central wavelengths of PBGs were confirmed at a wavelength of around 1560 nm, as planned [Fig. 4(a)]. From the results, we chose the narrower second PBG at $d_1 = 630$ nm and $d_2 = 450$ nm ($m = 3$). Then, while keeping the selected AZO and PLZT thickness conditions, we simulated the AZO/PLZT period number dependence of the transmittance spectra, as shown in Fig. 4(b). As the number of periods increased, the dip of the PBG became deeper, implying that PCs with a larger number of periods will exhibit better filtering properties. Moreover, we confirmed that PCs with over four periods and exhibiting narrow PBGs are enough to switch owing to an extinction ratio of over ~3 dB, which is below a transmittance of 50%. Given the above simulation results, we decided to fabricate TCE PCs consisting of four periods of an AZO/PLZT with 630-nm-thick AZO films and 450-nm-thick PLZT films.

5.2 Cross-sectional and surface microstructures of TCE PCs with the AZO/PLZT multilayer

Using the above simulation results and deposition conditions for AZO and PLZT films, we prepared a prototype TCE PCs consisting of four period numbers (8 layers) of AZO/PLZT through metal mask patterning. Then, a cross-sectional microstructure of the TCE PCs was observed with a FE-SEM, and the SEM image is shown in Fig. 5. The image confirms that dense PLZT layers with thicknesses ranging 440–450 nm were deposited between internal AZO layers with thicknesses ranging from 620 to 630 nm, as expected. However, as we increased the layer number, as shown in Figs. 6(a)–6(c), the surface morphology became rougher; the 1D RMS roughness also increased, as shown in Figs. 6(d)–6(f). We assumed that the multilayer coatings were roughened at the interfaces because of the high RF power in our fabrication process. To obtain a high deposition rate of AZO and PLZT films to fabricate a multilayer, we used relatively high RF power. Hayashi et al. also reported a high growth rate and rough surface morphologies of sputtered films at high RF power.\(^{25}\) This problem mainly results from the re-sputtering by energetic negative oxygen ions. In the case of the growth of oxide thin films...
by sputtering, it has been reported that the bombardment of energetic oxygen atoms and/or ions results in re-sputtering, and deteriorates the quality of growing films.\(^{26-28}\) Moreover, it was deduced that these rough interfaces created by the fabrication process will produce transmitted light scattering. Therefore, we attempted to analyze the measured transmittance of our TCE PCs based on the AZO/PLZT multilayer.

5.3 Transmittance spectrum analysis of TCE PCs with the AZO/PLZT multilayer by a revised MLI model

Figure 7 shows the transmittance spectra of the TCE PCs with a four-period AZO/PLZT multilayer during the experiment, simulation, and fitting curve using a revised MLI model that can consider light absorption and scattering effects. Our comparison of the experimental and simulated transmittance spectra revealed that both results exhibit almost the same PBG range with wave-lengths of 1470–1650 nm, but some of the transmittance values and central wavelengths in the PBG differed, which suggests that some of the refractive indices of deposited AZO and PLZT films differed from the reference values used in the simulation. We also assumed that the scattered and absorbed transmittance intensity differed. To confirm this assumption, we fitted the experimental transmittance by using a revised MLI model that considers light absorption and scattering by changing the extinction coefficients and introducing the values of the above-measured RMS roughness. As a result, the fitting curves are in good agreement with the experimental point data. However, the fitted refractive index \((n_1 \sim 2.105)\) of the AZO films was slightly higher than that of previously reported AZO films \((1.9 \sim 2.1)\)\(^{28}\) even though the fitted thicknesses of the AZO and PLZT films was almost the same as planned. This discrepancy might have been largely due to low carrier concentration.\(^{29}\) The resistivity \((1.2 \times 10^{-2} \Omega \text{cm})\) of the AZO films in our work\(^{14}\) was relatively high, implying low carrier concentration. In contrast, the refractive index \((n_2 \sim 2.334)\) of the PLZT films was in a low range compared to the previously reported measurements of PLZT films \((2.3 \sim 2.5)\).\(^{30,31}\) Moreover, the fitted extinction coefficients \((k_1,2 < 0.0024, \sim 0.0028)\) of the AZO and PLZT films were relatively high in comparison to previously reported values \((0.001 \sim 0.003)\).\(^{32,33}\) From the above results, we judged that these differences in the refractive indices of PLZT films and extinction coefficients in all films arose from the effect of decreased film density and some defects (porosity and voids) in the interfaces caused by roughness. Thus, to control tightly PBG position and transmittance values, it is important to improve the interface roughness of multilayers.

5.4 Photonic band gap tunability of TCE PCs with the AZO/PLZT multilayer

To evaluate the PBG tunability of the longitudinal electro-optically tunable TCE PCs based on a four-period AZO/PLZT multilayer, the applied voltage dependence of transmittance spectra were measured. Figure 8 shows the observed transmittance spectra at the DC voltages of 0, 4, and 8 V as well as the fitting curves as a function of wavelength. As the applied voltage increased, the observed PBG shifted to a longer wavelength, indicating changes in the refractive index due to the LEO effect of the PLZT films. To obtain the refractive index variations, the applied voltage dependence of transmittance spectra were also fitted using the revised MLI model by changing the refractive index of the PLZT layers. The refractive index variations were \(1.3 \times 10^{-2}\) and \(2.9 \times 10^{-2}\) at the DC voltages of 4 and 8 V, respectively.
and 8.5 nm, indicating a high PBG tunability of 4 and 8 V, respectively. The corresponding PBG shifts were 3.9 and 2.9 nm, respectively. To determine the PBG shift, we calculated the changed central wavelength \(\Delta_{\lambda}\) in PBG from the refractive index changes induced by the LEO effect of the PLZT multilayers by using the TEO effect.\(^{7,35}\) The PBG shifts were 3.9 and 8.5 nm at DC voltages 4 and 8 V, respectively, indicating an average PBG tunability of \(\sim 1\) nm/V. In comparison with previously reported PBG tunabilities,\(^{7,27-37}\) this value is very high. Specifically, the PBG tunability of this work is over 100-fold higher than that (0.008 nm/V) of reported tunable PCs based on a BST/MgO multilayer using the TEO effect.\(^{10}\)

Consequently, we simulated and fabricated the designed tunable TCE PCs using the LEO effect based on the AZO/PLZT multilayer, and we confirmed a very high PBG tunability of \(\sim 1\) nm/V. Our creation of a simple and highly tunable TCE PCs should enhance the development of ultra-compact optical devices, because it can serve as an alternative to using a switch that requires low driving voltage.

6. Conclusions

An advanced longitudinal electro-optically tunable TCE PCs was designed, simulated, and fabricated. This TCE PCs was composed of two sets of TCO layers to create an internal cathode electrode and to centered TFO layers as EO material. The results of the simulated transmission spectra by the MLI model indicate that the narrower PBG has AZO/PLZT films of 630 and 450-nm thickness. Four periods of AZO/PLZT films were then selected. After preparing our prototype TCE PCs consisting of the four selected periods of AZO/PLZT multilayers by using the RF magnetron sputtering method through metal mask patterning, the dense cross-sectional microstructure and rough surface morphologies were observed. Then, the transmission spectra as a function of wavelength of the prototype TCE PCs were measured, and we observed that the PBG falls within the near-infrared wavelengths from 1470 to 1650 nm. Moreover, the complex refractive indices of the AZO and PLZT layers were confirmed with the revised MLI model with light absorption and scattering, which showed well-fitted transmission spectra. The applied voltage dependence of the transmission spectra was also fitted, and the refractive index changes induced by the LEO effect of the PLZT layers were \(3.1 \times 10^{-2}\) and \(2.9 \times 10^{-2}\) at the DC voltages of 4 and 8 V, respectively. The corresponding PBG shifts were 3.9 and 8.5 nm, indicating a high PBG tunability of \(\sim 1\) nm/V.

![Graph](image)

**Fig. 8.** Observed and fitted transmittance spectra versus wavelengths at applied DC voltages of 0, 4, and 8 V. The points and solid curves are the experimental and fitting data. \(\Delta n\) and \(\Delta_{\lambda}\) indicate variations in the refractive indices of the PLZT films and central wavelength due to the various applied voltages.

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