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# Molecular Crystals and Liquid Crystals

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### Design and Fabrication of High-Performance Liquid Crystal Gratings

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## Design and Fabrication of High-Performance Liquid Crystal Gratings

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We present basic principles of designing and fabricating high-performance liquid crystal (LC) gratings with binary alignment structures which produce a variety of the diffraction properties. The periodic binary structure in the LC layer was fabricated through a selective irradiation of the UV light using the single-masking process. An alternating homeotropic and hybrid alignment configuration produces the input polarization-dependent diffraction suitable for the polarization-separating devices while an oppositely twisted binary alignment configuration gives the input polarization-insensitive diffraction. It is found that a dye-doped bidirectional alignment configuration can be used for obtaining optically controllable polarization properties.

Keywords: binary configuration; LC grating; photopolymer; polarization insensitivity

#### INTRODUCTION

Binary optical elements (BOEs) play an important role in many optical technologies [1–7] because of their compactness as well as their high potential for various modifications of light phase and/or polarization.

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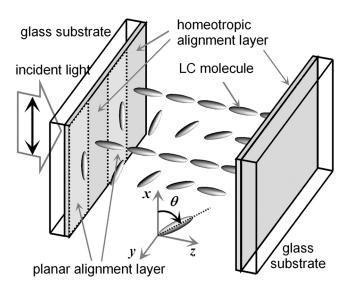
As the simplest form of the BOE, several binary gratings with onedimensional periodicity have been theoretically studied [8], and fabricated to achieve high diffraction efficiency [8,9]. In general, the BOEs were fabricated by utilizing complicated micro-lithographic techniques [10,11] or multi-layered dielectric coatings [12] in recent years.

The LCs are very useful for constructing a variety of gratings since the LC director modulation can be electrically and geometrically controlled in a simple way. For producing such director modulation through the LC layer, three types of binary structures are typically used; polymer structures with the space filled with the LC [13], dual alignment structures [14,15], and multi-electrode structures [16]. Recently, a rather simple and powerful method of constructing LC binary structures has been developed using the selective exposure of a linearly polarized ultraviolet (LPUV) light onto photosensitive polymers [7,17–19].

We report on several high performance LC binary gratings fabricated using a single-masking process of photoaligning the LCs, and present the design rules and basic principles for practical applications. The binary alignment structures fall into three different categories: i) an alternating homeotropic and hybrid alignment structure applicable for polarization-separating devices, ii) an oppositely twisted alignment structure possessing the polarization-invariance, and iii) a bidirectionally aligned structure having the polarization-insensitivity. The surface-controlled LC alignment technique was employed to construct the polarization-separating phase grating and the oppositely twisted polarization grating. In the bidirectional LC polarization grating, the polymer networks formed in a dye-doped LC medium play an essential role in achieving the optic axis modulation in the homogeneously aligned LC layer. In the following sections, the device configurations together with basic concepts of the three LC binary gratings will be described. Some concluding remarks will be made in the remaining section.

#### POLARIZATION-SEPARATING LC GRATING

We first describe the LC binary phase grating, having alternating homogeneous and hybrid domains, which gives the polarization-separating diffraction property. The basic structure of the LC polarization-separating grating is illustrated in Figure 1 where the LC molecules are aligned periodically in an alternating homeotropic and hybrid geometry. In the alternating hybrid aligned LC geometry as shown in Figure 1, the polarization-dependent intensity of the diffracted beam becomes well separated for various input wavelengths

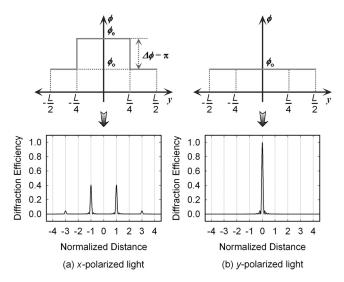


**FIGURE 1** The LC binary grating structure in an alternating homeotropic and hybrid geometry.

in the presence of an applied voltage. Considering that a grating vector lies along the *y*-direction, the input beam polarized along the grating vector (*y*-polarized light) will not experience any relative phase difference on passing through two adjacent domains. However, the input beam polarized along the *x*-direction has a modulated phase depending on the molecular tilt angle in the hybrid domain that is associated with

$$n_{\rm eff} = \frac{1}{d} \int_0^d \frac{n_o n_e}{\sqrt{n_e^2 \sin^2 \theta(z) + n_o^2 \cos^2 \theta(z)}} dz, \tag{1}$$

where  $n_e$  and  $n_o$  are the extraordinary and ordinary refractive indices of LC, respectively. Here,  $\theta(z)$  is the molecular tilt angle in the x-y plane and d is the cell thickness. Since the relative phase difference experienced by the polarized input beam is governed by the amount of the effective refractive index  $(n_{\rm eff}-n_o)$  through the molecular tilt  $\theta(z)$ , the magnitude of an applied voltage can control precisely the polarization-separating diffraction properties of the LC cell. In the ideal binary configuration, at the relative phase retardation  $\Delta \phi = (2m+1)\pi$  (m is an integer), there exist only odd orders of the diffraction that have the maximum intensities as shown in Figure 2.



**FIGURE 2** The phase profiles and the corresponding diffraction patterns of an ideal binary grating for (a) *x*-polarized light and (b) *y*-polarized light at the phase retardation of  $\Delta \phi = \pi$ .

For the ideal binary grating, the diffraction efficiency of the k-th order is given by

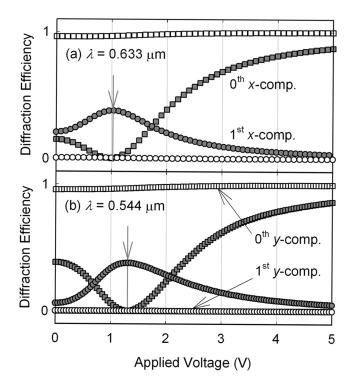
$$\eta_k = \begin{cases} \cos^2\left(\frac{\Delta\phi}{2}\right) & k = 0\\ \left(\frac{2}{\pi k}\right)^2 \sin^2\left(\frac{\pi k}{2}\right) \sin^2\left(\frac{\Delta\phi}{2}\right) & k \neq 0 \end{cases}, \tag{2}$$

where  $\Delta\phi$  is the relative phase difference between two adjacent domains in the binary grating system. When the off-diagonal elements in the Jones matrix are non-zero, any linearly polarized light can be decomposed into two linearly polarized states that are orthogonal to each other [20]. In our case, the diffracted intensity of a linearly polarized light can be decomposed into the x- and y-components. As shown in Figure 1, only the input beam polarized along the x-component experiences the phase difference of  $\Delta\phi=2\pi(n_{\rm eff}-n_o)d/\lambda$  where  $\lambda$  is the wavelength of the input beam. For the x-component of the incident light, there exist only odd orders  $(k=\pm 1,\,\pm 3,\,\ldots)$  of the diffraction that have the maximum intensities at  $\Delta\phi=(2m+1)\pi$  (m is an integer). Note that the zeroth order or non-diffracted beam has the maximum intensity at  $\Delta\phi=2m\pi$  and the minimum (zero) intensity at  $\Delta\phi=(2m+1)\pi$ . Consequently, when an applied voltage

is adjusted to satisfy that for given wavelength of the input beam,  $\Delta \phi = (2m+1)\pi$ , all the diffracted beams of  $k=\pm 1, \pm 3, \ldots$  are polarized along the y-direction and non-diffracted beam of k=0 is polarized along the x-direction. Therefore, such polarization-separating features of our LC binary phase grating can be used as a polarizing beamsplitter (PBS) with the wavelength tuning capability under an applied voltage.

In the light of the above idea, we fabricated the LC binary phase gratings with alternating hybrid alignment structures using a single step process of photoalignment. The periodicity of the grating was chosen as 20 μm [17,21]. The photopolymer material, aligning the LC molecules homogeneously by the polarized ultraviolet (UV) exposure and homeotropically by the non-treatment with the UV light, was LGC-M1 of LG Cable, Ltd [17]. An array of one-dimensional binary phase gratings was then produced on the substrate by only one step exposure of the LPUV beam through an amplitude photomask at 2.0 mW for 2 minutes. The thickness of the LC cell was maintained using glass spacers of 8.8 µm thick. A commercial nematic LC, MLC-6012 of E. Merck, was filled into the cell at 95°C in the isotropic state. The ordinary and extraordinary refractive indices of MLC-6012 are  $n_o = 1.4620 + 5682/\lambda^2$  and  $n_e = 1.5525 + 9523/\lambda^2$ , respectively, where  $\lambda$  is the wavelength of light in nm. The dielectric anisotropy and the elastic constants are  $\Delta \varepsilon = 8.2$ ,  $K_1 = 11.6 \times 10^{-12}$  N,  $K_1 = 5.5 \times 10^{-12}$  N,  $K_1 = 16.1 \times 10^{-12} \text{ N, respectively } [22].$ 

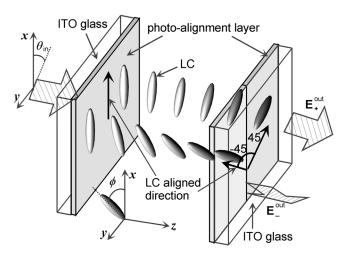
Figure 3 shows the x- and y-components of the zeroth- and firstorder diffractions as a function of the applied voltage for wavelengths of  $\lambda = 0.633 \,\mu m$  and  $\lambda = 0.544 \,\mu m$ . As shown in Figure 3, the y-components are quite insensitive to the applied voltage  $V_{
m app}$  while the x-components depend on  $V_{app}$ . In other words, the only x-component of the incident light experiences the phase modulation controlled by the applied voltage. The y-polarized light is just passing through the LC grating with no experience of the phase modulation. Note that the y-components for all higher order diffractions and the x-component for the zeroth order diffraction are essentially zero at a certain voltage called a tuning voltage. For example, the y-component for the first order diffraction and the *x*-component for the zeroth order diffraction vanish at  $V_{\rm app} \approx 1.0 \, \rm V$ , which corresponds exactly to the case of  $\Delta \phi = 2\pi (n_{\rm eff} - n_o) d/\lambda = \pi$  at the wavelength of  $\lambda = 0.633 \, \mu {\rm m}$ . The tuning voltages for the wavelength of  $\lambda = 0.544 \,\mu m$  was achieved to be about 1.2 V. This means that at such voltage, the LC cell behaves as an ideal PBS which separates an input beam into a pair of the diffracted beam polarized in the y-direction and that polarized in the x-direction by combining only lower order diffractions.



**FIGURE 3** The measured *x*- and *y*-components of the zeroth- and first-order diffractions of the input beams with (a)  $\lambda=0.633\,\mu\mathrm{m}$  and (b)  $\lambda=0.544\,\mu\mathrm{m}$  passing through the alternating hybrid LC binary grating with the cell thickness  $d=8.8\,\mu\mathrm{m}$  as a function of the applied voltage, respectively. Gray solid arrows indicate the tuning voltages that correspond exactly to the case of  $\Delta\phi=\pi$ .

#### OPPOSITELY TWISTED LC GRATING

We now present the LC polarization-invariant grating in an oppositely twisted configuration fabricated by a single-masking process. The basic structure of the LC polarization grating is described in Figure 4 where the LC molecules are twisted periodically in an alternating oppositely twisted geometry. In the optically adiabatic limit [23], the polarization of incident light becomes rotated an angle of either  $+45^{\circ}$  or  $-45^{\circ}$  with respect to the *x*-axis on passing the other surface with two oppositely twisted domains. The alternating rotation of the polarization vector naturally gives the polarization-invariant optical modulation. Using the Jones matrix formalism [23], the outcoming optical fields,  $\mathbf{E}_{1}^{\text{out}}$  and  $\mathbf{E}\mathbf{1}_{2}^{\text{out}}$ , in the positively and negatively twisted domains are given by [24]



**FIGURE 4** The LC polarization grating structure with two oppositely twisted domains. At one surface, the LC director is uniform along the *x*-axis and at the other, two directors in the two domains make angles of  $\pm 45^{\circ}$  with respect to the *x*-axis.

$$\begin{split} \mathbf{E}_{+}^{\text{out}} &= \frac{1}{\sqrt{2}} \begin{pmatrix} \exp(-i\varphi)\cos\theta_{\text{in}} + \sin\theta_{\text{in}} \\ -\exp(-i\varphi)\cos\theta_{\text{in}} + \sin\theta_{\text{in}} \end{pmatrix}, \\ \mathbf{E}_{-}^{\text{out}} &= \frac{1}{\sqrt{2}} \begin{pmatrix} \exp(-i\varphi)\cos\theta_{\text{in}} - \sin\theta_{\text{in}} \\ \exp(-i\varphi)\cos\theta_{\text{in}} + \sin\theta_{\text{in}} \end{pmatrix}, \end{split} \tag{3}$$

where  $\theta_{\rm in}$  represents the polarization angle of incident light with respect to the *x*-axis. The phase retardation of the LC layer is given by  $\varphi = 2\pi\Delta nd/\lambda$  where  $\Delta n$  and d are the birefringence of the LC and the cell thickness, respectively.

Let us describe the diffraction properties of our LC polarization grating in an oppositely twisted binary configuration. The Fraunhofer diffraction patterns of a periodic grating device are calculated by the Fourier transform [25]. In the case of the binary grating with identical periodicities of two different domains as shown in Figure 4, the optical fields of the zeroth and the first order diffraction beams,  $\mathbf{E}_0^{\text{diff}}$  and  $\mathbf{E}_1^{\text{diff}}$  are [26]

$$\begin{split} \mathbf{E}_{0}^{\mathrm{diff}} &= \frac{1}{2} (\mathbf{E}_{+}^{\mathrm{out}} + \mathbf{E}_{-}^{\mathrm{out}}) = \frac{1}{\sqrt{2}} \begin{pmatrix} \exp(-i\varphi)\cos\theta_{\mathrm{in}} \\ \sin\theta_{\mathrm{in}} \end{pmatrix}, \\ \mathbf{E}_{1}^{\mathrm{diff}} &= \frac{i}{\pi} (\mathbf{E}_{+}^{\mathrm{out}} - \mathbf{E}_{-}^{\mathrm{out}}) = \frac{i\sqrt{2}}{\pi} \begin{pmatrix} -\sin\theta_{\mathrm{in}} \\ \exp(-i\varphi)\cos\theta_{\mathrm{in}} \end{pmatrix}. \end{split} \tag{4}$$

Note that the optical fields of the two diffraction beams are orthogonal to each other irrespective of the polarization state,  $\theta_{\rm in}$ , of the incident beam and the phase retardation,  $\varphi$ , of the LC layer since  $\mathbf{E}_0^{\rm diff} \cdot \mathbf{E}_1^{\rm diff} = 0$ . As a consequence, the zeroth and first order diffraction efficiencies,  $\eta_0$  and  $\eta_1$  are

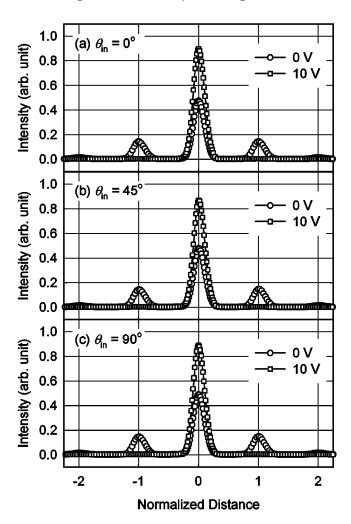
$$\begin{split} &\eta_0 = E_0^{\rm diff} E_0^{\rm diff^+} = \frac{1}{2} \left(\cos^2 \theta_{\rm in} + \sin^2 \theta_{\rm in}\right) = \frac{1}{2}, \\ &\eta_1 = E_1^{\rm diff} E_1^{\rm diff^+} = \frac{2}{\pi^2} \left(\sin^2 \theta_{\rm in} + \cos^2 \theta_{\rm in}\right) = \frac{2}{\pi^2}. \end{split} \tag{5}$$

It should be noted that both diffraction efficiencies are expressed as certain constants independent of the polarization of incident light and the phase retardation through the LC cell.

In the presence of an applied voltage, the LC molecules with positive dielectric anisotropy are reoriented out of the substrates in the both domains, positive and negative regions. In the vertically deformed state, the polarization modulation disappears and the diffracted beams disappear. Such switchable features of our LC polarization grating are suitable for a switchable 2-dimensional/3-dimensional (2D/3D) display system where in the 3D mode, the switchable polarization grating divides the 3D images into the left- and right-images in a stereoscopic display [6]. On the other hand, in the 2D mode, the switchable grating passes the 2D images with no modulation.

The LC polarization grating with oppositely twisted domain structures was fabricated using glass substrates coated with the photopolymer of LGC-M2 (LG Cable Ltd., Korea) [27]. The photopolymer can align the LC molecules homogeneously under the illumination of a LPUV light and alter repeatedly the direction of the LC alignment depending on the polarization of the LPUV light. The substrate having binary LC domain structures was prepared by a single-masking process of two step-LPUV illumination [27]. After the LPUV exposure onto the whole region, the LPUV light through an amplitude photomask, whose direction of the modulation made an angle of  $45^{\circ}$  with respect to the initial LC director, was illuminated for aligning the LC molecules perpendicular to the initial director. The grating period was  $100\,\mu\text{m}$ . The cell thickness was maintained using glass spacers of  $9.48\,\mu\text{m}$ . The LC material used in this study was MLC-6012 of E. Merck.

Figure 5 shows the diffraction intensity profiles of our LC polarization grating for different polarization states of an incident light. Here, the normalized distance represents the distance in unit of the order of diffraction along the *y*-axis. Open circles and open rectangles denote the diffraction intensities at the applied voltages of 0 V and 10 V,



**FIGURE 5** Diffracted intensity profiles through the oppositely twisted LC polarization grating with the cell thickness  $d=9.48\,\mu\mathrm{m}$  ( $\Phi\approx3\pi$ ) as a function of the normalized distance at different applied voltages for three polarization states (a)  $0^{\circ}$ , (b)  $45^{\circ}$ , and (c)  $90^{\circ}$  with respect to the *x*-axis.

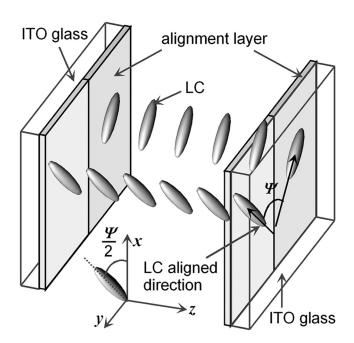
respectively. As shown in Figures 5(a)–(c), in the absence of an applied voltage, the incident light always experiences the polarization modulation and the diffraction appears on passing through our LC grating. In the presence of an applied voltage, however, the LC molecules are reoriented perpendicular to the substrates in the two twisted regions and thus no polarization modulation, i.e., no diffraction appears. It

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should be noted that the diffraction intensities for all different polarization states of the incident light are exactly same. As expected from Eq. (5), any polarization state of an incident light produces the identical diffraction property in our oppositely twisted LC grating where the twist angles,  $\pm 45^{\circ}$ , in two alternating domains are symmetric with respect to the *x*-axis. In a special case that  $\varphi = m\pi$  (*m* is an integer) in Eq. (4), the zeroth and the first order diffracted beams are linearly polarized and the resulting polarization states are perpendicular to each other irrespective of the polarization state of the incident light [27].

#### **BIDIRECTIONALLY ALIGNED LC GRATING**

We now present a bidrectionally aligned LC grating, having the polarization-insensitivity, based on a dye-doped LC system with polymer networks. The polarization-insensitive diffraction was achieved by periodically modulating the optic axis of a homogeneously aligned nematic LC layer as shown in Figure 6. Periodic domains were produced by a single UV exposure onto a dye-doped LC system where



**FIGURE 6** The bidirectional LC polarization grating structure with two periodically modulated optic axes in the homogeneous LC layer.

the polymer networks were periodically formed by the UV irradiation through a patterned photomask [28].

We describe the diffraction properties of the LC binary grating with the optic axis modulation and the phase retardation. Within the Fraunhofer diffraction formalism for a binary anisotropic grating [26], the optical field of the first-order diffraction is given by [28]

$$\mathbf{E}_{+1}^{\text{diff}} = -\frac{2}{\pi} \sin \frac{\Gamma}{2} \sin \Psi \begin{pmatrix} A_x \\ A_y \end{pmatrix}, \tag{6}$$

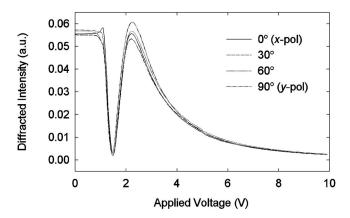
where  $A_x$  and  $A_y$  represent the x- and y-components of an incident light in a coordinate bisecting two easy axes,  $\Gamma$  is the phase retardation, and  $\Psi$  is the angle between the two easy axes. It should be noted that the first order diffraction efficiency is expressed as a function of the phase retardation and the optic axis modulation that are independent of the polarization of the incident light.

The bidrectionally aligned LC grating based on the dye-doped LC with polymer networks was fabricated using two steps of a patterned UV light exposure and a uniform illumination. The cell was first irradiated with the UV light from a Xe-Hg lamp at  $10\,\mathrm{mW/cm^2}$  for 50 minutes through an amplitude photomask to form polymer networks selectively in the bulk. The irradiated region has the higher polymer concentration than the not-irradiated region. In the polymer-stabilized state with a low concentration (2 wt.%) of the polymer, the uniform alignment of the LC along the rubbing direction was produced. After the patterned UV exposure, a uniform illumination was carried out to generate the optical reorientation of the LC molecules in the periodically modulated polymer networks as shown in Figure 6. As a pump beam source, an Ar-ion laser with wavelength of 488 nm and the power of 100 mW/cm<sup>2</sup> was used. For the linearly polarized pump beam, the LC molecules in the UV-illuminated region with high density of polymer networks are reoriented perpendicular to the polarization of the pump beam since the polymer networks formed in the LC medium provide the internal surface for the dye adsorption [28]. On the other hand, the LC molecules in the not-illuminated region with low density of polymer networks are essentially maintained in the initial state along the rubbing direction in spite of the irradiation of the pump beam. The single pump beam used was linearly polarized at an angle of 45° with respect to the rubbing direction  $(\Psi=45^{\circ})$  so that the maximum optical torque was produced. In such configuration, the diffraction efficiency of our LC grating can be electrically controllable and insensitive to the polarization of the input 374/[776] C.-J. Yu et al.

beam in the entire voltage range of operation since the periodically modulated optic axis is preserved under an applied voltage.

The materials used in our study were a nematic liquid crystal of ZLI-2293 (E. Merck Industries), azo-dye of Methyl Red (MR, Sigma-Aldrich), and an UV-curable photopolymer of NOA65 (Norland Products Inc.). The dye-doped LC/polymer composite system was prepared from a homogeneous mixture of 97 wt.% of ZLI-2993, 1 wt.% of MR, and 2 wt.% of NOA65. The mixture was injected into the sample cell at temperature above the clearing point of the LC. The inner surfaces of the substrates were coated with polyvinyl alchol (PVA) layers and rubbed unidirectionally so as to promote the initial homogeneous alignment of the LC. The cell thickness was maintained using glass spacers of  $6\,\mu m$  thick.

Figure 7 shows the first-order diffraction of our bidrectionally aligned LC grating as a function of the applied voltage for various states of the input polarization. The valleys and peaks observed in the diffraction intensities correspond to the phase retardation of  $2\pi$  and  $\pi$ , respectively. At the applied voltage of  $10\,\mathrm{V}$ , the phase retardation becomes nearly zero and the diffraction disappears. This tells us that the first-order diffraction is polarization-insensitive in the entire voltage range we studied. In our dye-doped LC binary grating, the diffraction efficiency can be also controlled by the polarization state of the pump beam with respect to the initial rubbing direction.



**FIGURE 7** The diffracted intensities through the bidirectionally aligned LC polarization grating as a function of the applied voltage for various states of the input polarization.

#### CONCLUSION

We presented several high performance LC gratings fabricated by a single-masking process of photoalignment together with the design rules and basic principles. The LC polarization-separating grating would be useful for constructing a polarizing beamsplitter with the wavelength tuning capability. Both the oppositely twisted LC grating and the dye-doped bidirectionally aligned LC grating posses the polarization-insensitivity in the absence of an applied voltage. Moreover, the bidirectionally aligned LC grating was polarization-insensitive in the entire voltage range we studied. Along with analytic calculations for an ideal binary grating, the diffraction properties considering the distortions of the LC at domain boundaries were discussed in the linearly graded phase approximation [21].

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