

Full length article

Programmed Achromatic and Wide Field-of-View Retarder Using Form Birefringence of Template-Grown ZnO nanostructures

Manh-Thang Tran^a, Nguyen-Hung Tran^{a,*}, Ji-Hoon Lee^b

^a Division of Electronics Engineering, Jeonbuk National University, 567 Baekje-daero, Deokjin-gu, Jeonju-si, 54896, Jeollabuk-do, Republic of Korea

^b Future Semiconductor Convergence Technology Research Center, Division of Electronics Engineering, Jeonbuk National University, 567 Baekje-daero, Deokjin-gu, Jeonju-si, 54896, Jeollabuk-do, Republic of Korea

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ABSTRACT

Achromatic retarders with a wide field of view (FOV) can maintain a consistent performance across a broad range of wavelengths and wide viewing angles. Despite the widespread use of retarders, achieving a true achromatic retarder with a wide FOV remains challenging. Here, we present an achromatic quarter-wave plate (QWP) based on form birefringence (FB) of nanostructures operating in visible wavelengths from 450 to 650 nm. Our 1.8-μm-thick QWP was fabricated using a templated growth method that combines laser interference lithography and hydrothermal growth methods. A flat reflectance spectrum with an average reflectance of 1.37% was measured from the antireflection of a circular polarizer with the FB retarder, confirming the wideband efficiency of the QWP. The achromaticity of retardation was unchanged at greater incident angles.

1. Introduction

An achromatic retarder with a wide field of view (FOV) can be a useful component of optical systems due to its ability to convert the polarization of light independent of the wavelength and incident angle. Since the physical concept of an achromatic retarder was proposed by Pancharatnam [1], various approaches to this task have been attempted [1–33]. Two main approaches have been developed to fabricate achromatic retarders. First, a stack-type retarder combines multiple waveplates to obtain achromaticity [2–11]. The chromatic retardation of light after passing through the first retarder is compensated for by subsequent retarders, whose optical axes are obliquely oriented. Designed using optimization of in-plane retardation, the retarders inevitably suffer from when the effective optic axis orientation varies depending on the wavelength of light [5]. The second approach is based on anisotropic dispersion of the principal refractive indices using a single retarder layer. The chemical structures of the molecules are designed to possess different dispersions along different molecular axes for this approach [12–23].

The achromaticity of a retarder can be evaluated using the wavelength dependence of phase retardation Γ [5]:

$$\Gamma(\lambda) = \frac{2\pi d}{\lambda} \Delta n(\lambda), \quad (1)$$

where, d is the thickness of the medium, λ is the wavelength of light, and Δn is the material birefringence. $\Delta n(\lambda)$ must increase at a

longer λ , i.e., decrease with a high frequency of light, for $\frac{d\Gamma}{d\lambda} = 0$. In that case, the retardation $Re = d\Delta n(\lambda)$ is said to have a negative dispersion (ND) [20,21]. While a positive dispersion (PD) of retardation is observed in most natural materials, a ND is primarily induced in artificial materials [12–33]. Despite extensive efforts in recent decades, limitations in performance and challenges in fabrication of ND retarders persist. Dispersion of organic material-based retarders tends to be weaker at longer wavelengths [12–23]. ND retarders made from inorganic material have been rarely reported, and they produced low birefringence and required a sophisticated fabrication process [32]. In addition, the FOV of the conventional retarders is not wide, i.e., the retardation and the achromaticity of the retardation change significantly as the incident angle increases. Achromatic retarders and their characteristics are summarized in Table 1.

Another approach, form birefringence (FB) [24–33], is based on a completely different optical principle. While intrinsically anisotropic materials are used in previous approaches, optically isotropic materials can be used for FB. The theory of FB was suggested in the 1950s [34, 35]. It has been experimentally verified by artificial gratings [24–33] and observed in natural structures, such as the R8 cell in the eyes of mantis shrimp [36–38]. A subwavelength grating (SWG), the simplest structure showing FB, is composed of two periodically alternating isotropic materials, as shown in Fig. 1(a). Within a grating period of

* Corresponding authors.

E-mail addresses: trannguyenhung.work@gmail.com (N.-H. Tran), jihoonlee@jbnu.ac.kr (J.-H. Lee).

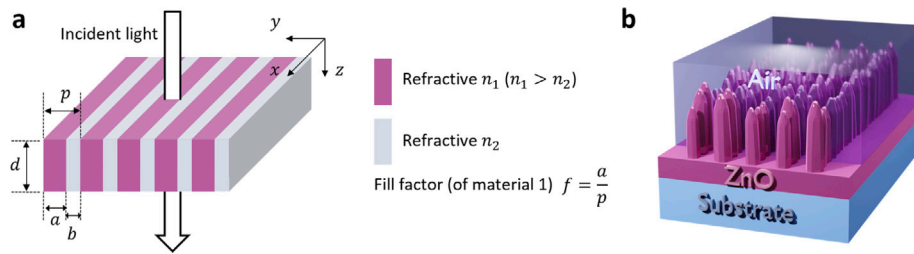


Fig. 1. (a) Subwavelength grating structure and (b) modeled ZnO NR-based structure.

Table 1
Classification of achromatic retarders.

Classification	Fabrication method	Limitations	References
Stack-type	Stack of the multi-retarders with oblique orientation of the optic axes	Complicated fabrication process and high cost; change of the effective optic axis orientation at different wavelengths of light	[2–11]
Reactive mesogen	Coating of the reactive mesogen molecules and UV exposure	Weak negative dispersion at long wavelength region; difficult to implement biaxiality	[12–23]

p , the denser material with a higher refractive index, n_1 , has a width of a , while the less dense material with a lower refractive index, n_2 , has a width of b . An ND of retardation can be achieved by optimizing n_1 , n_2 , p , and a fill factor of the denser medium $f = a/p$ [26,28,31,33].

Several top-down micro- and nanofabrication approaches have been used to fabricate ND FB retarders. A mid-infrared (3.5–5 μm) achromatic quarter-wave plate (QWP) was demonstrated by an SWG fabricated on silicon substrates with conventional photolithography and reactive ion etching (RIE) [27]. Imprint lithography is commonly used in SWG fabrication, although reports on ND waveplates are limited [29, 31]. Using electron beam (E-beam) lithography and RIE, a broadband retarder in the full visible region was reported, but the magnitude of phase retardation was limited to $34 \pm 7^\circ$ [28]. The phase retardation of the retarder must be greater than 90° for practical applications such as QWPs, but difficulties fabricating high-aspect-ratio gratings have limited our ability to achieve this threshold. Furthermore, achromatic SWG-based QWPs rarely operate in the visible range [31].

In this report, we present a programmed achromatic QWP operating in the visible wavelength range using FB of template-grown nanostructures (TGN) as illustrated in Fig. 1(b). The TGN were fabricated by combining laser interference lithography (LIL) and hydrothermal growth of ZnO nanorods (NRs), which offer structural flexibility and offer the potential to design different types of retarders. Our achromatic QWP achieved a phase retardation of 91° , with a large birefringence of $\Delta n(550 \text{ nm}) = 0.08$. Additionally, the achromaticity of retardation was unchanged as the incident angle of the light increased, implying a wide FOV. We also evaluated the antireflection performance of the QWP, showing its potential for practical applications.

2. Method

The fabrication process of our ZnO TGN is illustrated in Fig. 2. Initially, a ZnO seed layer was deposited on a glass substrate using RF sputtering. A one-dimensional photoresist (PR) grating was patterned on the ZnO-deposited substrate using LIL. Finally, the hydrothermal method was used to grow ZnO NRs within the open areas of the PR pattern over the ZnO seed layer. Finally, periodic ZnO columns formed on the substrate after stripping the PR grating.

2.1. ZnO seed layer deposition

A 1 inch $\times$ 1 inch glass slide (CITOGlas), used as a substrate, was sequentially cleaned with acetone, isopropanol, and deionized water (DI water) and then dried in an oven at 90°C for 2 h. A ZnO layer was deposited on the substrate using an RF sputtering system (DAEKI HI-TECH) at 150 W, an argon flow of 72 sccm, and a working pressure of 15 mTorr (abbreviated as ZnO/glass), as depicted in Fig. 2(a). The thickness of the ZnO layer (50 nm to 1 μm) was controlled by sputtering duration (5 to 90 min).

2.2. Template PR grating fabrication by LIL

Fig. 2(b) depicts the optical setup of LIL using a Lloyd's mirror configuration. A laser beam generated by a commercial 405 nm laser (TopMode 405, TOPTICA, coherent length $> 20 \text{ m}$) was continuously guided by mirrors 1–3 and entered a spatial filter, which consisted of a 10x objective lens (LMPlan 10x/0.3) and an 8- μm pinhole (#38538, Thorlabs). The laser beam was collimated by a plano-convex lens (LA1725-A, Thorlabs) and projected to a substrate by mirror 4. The exposed surface substrate and mirror were perpendicular to one another on a rotation stage. The laser beam center was aligned at the intersection of the substrate and the mirror. The incident angle θ_i shown in Fig. 2(c), can be adjusted by rotating the stage. To fabricate the template PR grating, a positive PR (AZ GXR-601 (14cp), AZ Electronic Materials) diluted with propylene glycol methyl ether acetate (PGMEA, $\geq 99.5\%$, Kanto Chemical) at a volume ratio of 2:3 was spin-coated at 7000 rpm for 60 s to achieve a thickness of 250 nm on the ZnO-deposited substrate. The PR-coated substrate was exposed at θ_i values of $25.5\text{--}36.6^\circ$ with a dose of $15\text{--}21 \text{ mJ}/\text{cm}^2$, followed by post-exposure baking at 110°C for 2 min. Development was achieved by immersing it in AZ 300 MIF Developer solution (AZ Electronic Materials) for 13 s then rinsing it in DI water. The PR-patterned ZnO/glass was dried with compressed air and heated at 110°C for 5 min to evaporate the residual solvent and water. The periodicity of the PR columns can be controllable by adjusting the θ_i , while the width of the columns was determined by controlling the exposure dose. A greater exposure dose produced a narrower width and a wider separation of the PR columns, as can be seen in Fig. 2(d).

2.3. ZnO NR growth

ZnO NRs were grown on PR-patterned ZnO/glass using a hydrothermal method [Fig. 2(a)]. A solution containing 40 mL of 25 mM hexamethylenetetramine (HMTA, $\geq 99.5\%$ pure, Aldrich) and 0–3 mL of ammonium hydroxide solution (28%–30%, Sigma-Aldrich) was dropped slowly into 40 mL of 16 mM zinc acetate dihydrate ($\geq 98\%$, Sigma-Aldrich) and stirred for 10 min before being transferred to a Teflon liner. The PR-patterned ZnO/glass was immersed vertically in the solution mixture. The temperature was kept at 90°C , and growth time was varied from 1 to 4 h to control ZnO rod height. The sample was extracted from the Teflon liner, rinsed immediately in DI water, dried by compressed air, and heated to 90°C in an oven for 2 h to remove residual water. Finally, the PR mask was stripped from the sample by

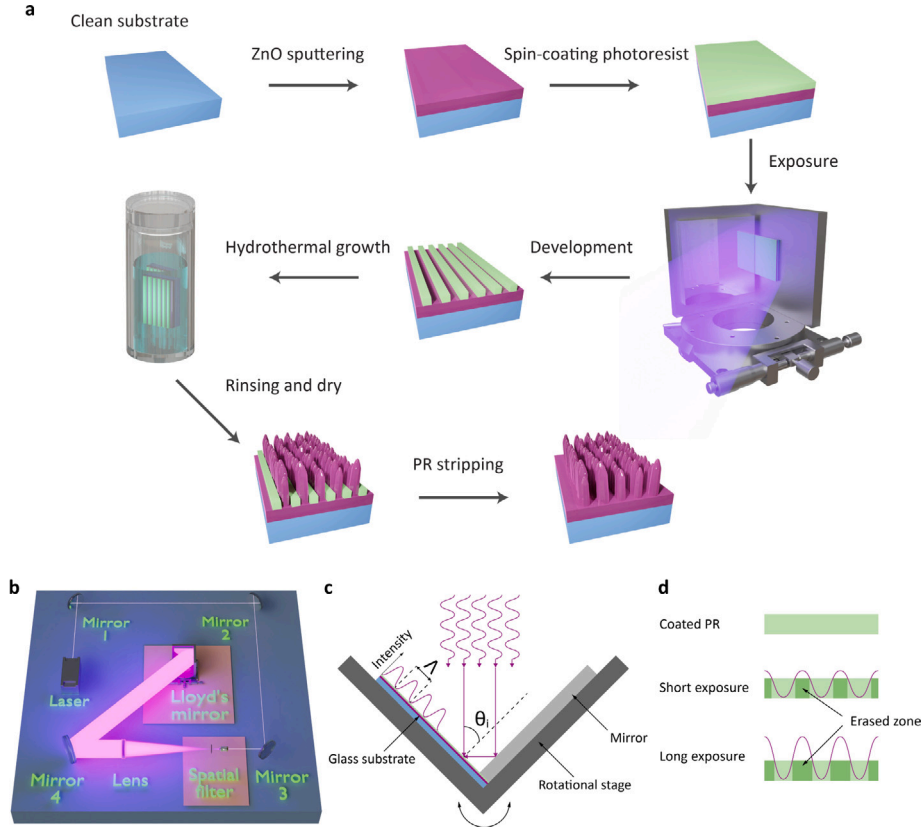


Fig. 2. (a) Schematic of ZnO TGN fabrication. (b) Optical setup of the laser interference system. (c) Lloyd's mirror interferometer structure. (d) A PR grating formation under short and long exposure.

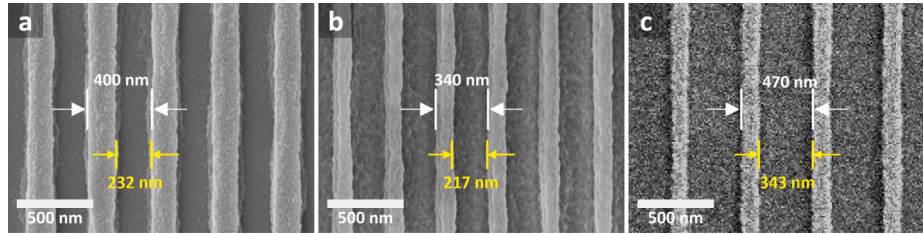


Fig. 3. Template PR gratings with varying periods and aperture ratios. The aperture ratio was defined as the ratio of PR separation (yellow) with respect to the period (white).

immersion in a PS-400, PRIEM stripper solution for 2 min, followed by rinsing with DI water and drying in an oven at 90 °C for 2 h.

Structural analysis of the samples was carried out using field emission scanning electron microscopy (SUPRA40VP, Carl Zeiss). The retardation properties (in-plane and out-of-plane) were measured according to the Stokes polarimetry method [39]. Transmittance, reflectance, and CIElab color were analyzed using an ultraviolet–visible light spectrometer (UV-2600, SHIMADZU) equipped with an integrated sphere (ISR-2600). Theoretical retardation of TGN was computed using Bloch wave analysis theory, and the details are described in the literature [33, 40].

3. Results and discussion

3.1. ZnO TGN fabrication

3.1.1. Template photoresist grating fabrication

Fig. 3 are scanning electron microscopy (SEM) images of the template PR gratings on glass substrates. The grating period p was controlled from 340 to 470 nm by varying the incident angle θ_i of LIL from

25.5 to 36.6 °, according to the following equation [41]:

$$p = \frac{\lambda}{2n \sin \theta_i}, \quad (2)$$

where $\lambda = 405$ nm is the laser wavelength and $n = 1$ is the refractive index of the air (see the Methods section). The width of the PR columns was determined by the exposure dose as shown in Fig. 2(d). A greater exposure dose produced a narrower width and a wider separation of the PR columns. The aperture ratio was defined as the spatial area ratio of the PR columns separation divided by the period. The aperture ratios of Figs. 3(a), 3(b), and 3(c) were 0.58, 0.64, and 0.73, respectively. The ZnO was grown from the opened area between PRs, and the period and aperture ratios of the PR columns are critical parameters that determine the geometrical parameters of the ZnO columns and the resulting retardation dispersion [25,33].

3.1.2. ZnO growth optimization

To form a ZnO grating structure, ZnO columns composed of NRs were grown through the open area of the PR grating patterns. For practical use as optical retarders, rods need to be uniform, vertical, dense, and transparent. Initially, the ZnO rods were synthesized on

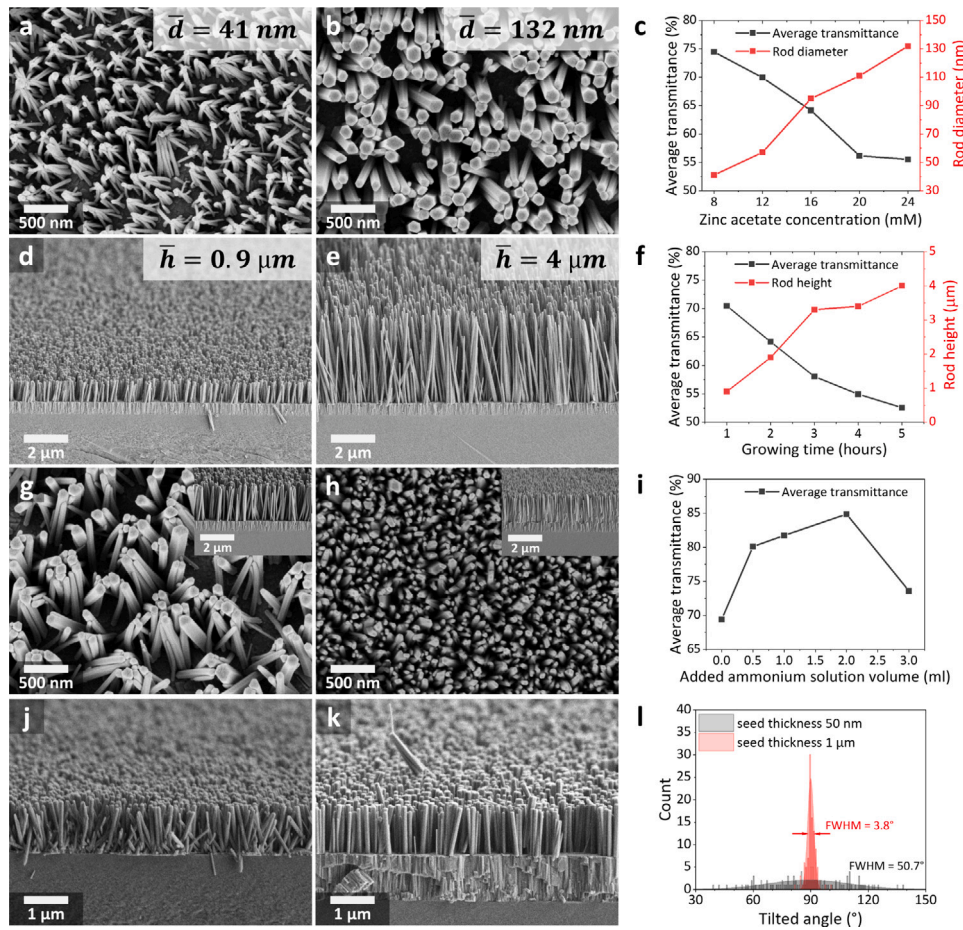


Fig. 4. ZnO morphology and transmittance under different hydrothermal growth conditions. (a, b) SEM images of ZnO NRs synthesized with a.Zn²⁺ concentrations of 8 mM (a) and 24 mM (b). In this experiment, $T_{\text{seed}} \approx 570$ nm, $C_{\text{HMTA}} = 25$ mM, $V_{\text{NH}_4\text{OH}} = 0$ mL, and $t = 2$ h. (c) Dependence of average transmittance and diameter of ZnO NRs on a.Zn²⁺ concentration. (d, e) SEM images of ZnO NRs grown over durations of 2 h (d) and 5 h (e). In this experiment, $T_{\text{seed}} \approx 570$ nm, $C_{\text{a.Zn}^{2+}} = 16$ mM, $C_{\text{HMTA}} = 25$ mM, and $V_{\text{NH}_4\text{OH}} = 0$ mL. (f) Dependence of average transmittance and height of ZnO NRs on growing time. (g, h) SEM images of ZnO NRs when 0 mL (g) and 3 mL (h) NH₄OH solution was added to the precursor mixture. In this experiment, $T_{\text{seed}} \approx 570$ nm, $C_{\text{a.Zn}^{2+}} = 16$ mM, $C_{\text{HMTA}} = 25$ mM, and $t = 2$ h. The insets are cross-sectional SEM images of ZnO NRs. (i) Average transmittance of ZnO NRs by volume of added NH₄OH solution. (j, k) SEM images of ZnO NRs synthesized on 50-nm- and 1-μm-thick seed layers (counted from 100 NRs).

a non-patterned seed layer, and the morphology and transparency of the sample were evaluated under a variety of hydrothermal growth conditions. The precursor concentration (C), volume of added ammonia solution ($V_{\text{NH}_4\text{OH}}$), reaction duration (t), and ZnO-sputtered seed thickness (T_{seed}) were the growth parameters.

Fig. 4(a–f) illustrates the morphology and transmittance of ZnO grown under various conditions. Figs. 4(a) and 4(b) show a clear increase in ZnO rod diameter from 41 to 132 nm when the zinc acetate dihydrate (a.Zn²⁺) concentration was increased from 8 to 24 mM. Figs. 4(d) and 4(e) are cross-sectional SEM images of samples fabricated with different growth times. The height of the ZnO NRs increased from 0.9 to 4 μm when the growth time was varied from 1 to 4 h. Both increases in rod diameter and rod height led to a degradation in average transmittance. Figs. 4(c) and 4(f) depict larger rod dimensions, resulting in more severe scattering [42,43]. These findings suggest that the precursor concentration and growth time are key experimental parameters for controlling the morphology and transmittance of ZnO rods.

Figs. 4(g) and 4(h) are SEM images of ZnO morphology at varying $V_{\text{NH}_4\text{OH}}$. The density of NRs increased when ammonium was added to the solution. The surface was covered by closely packed ZnO rods. Meanwhile, the growth rate was low when $V_{\text{NH}_4\text{OH}}$ was less than 1 mL, and the rods grew rapidly when $V_{\text{NH}_4\text{OH}}$ was greater than 2 mL (Supplementary Material, Fig. S3). As the volume of ammonium increased, ZnO nucleation was promoted, increasing number of hydroxyl molecules in

the precursor solution [44,45]. A high density of ZnO NRs is crucial for achieving the desired structure of ZnO grating (Fig. 1). Adding ammonium was an essential process for ZnO growth. As shown in Fig. 4(i), greater transmittance was observed in the high-density samples except $V_{\text{NH}_4\text{OH}} = 3$ mL, consistent with findings in the literature [42].

Figs. 4(j) and 4(k) are SEM images of ZnO NRs grown on seed layers with different thicknesses. On a 50-nm-thick layer, ZnO NRs grew in various directions, as reflected in the tilted-angle distribution, with a large full width at half maximum (FWHM) of 50.7°, as depicted in Fig. 4(l). The ZnO rod verticality was significantly improved when the seed layer was thickened to 1 μm, resulting in a reduced FWHM of 3.8°. Previous reports showed that an increase in thickness improved seed crystallinity, which facilitated vertical NR growth [46,47]. Straight ZnO NRs are essential to achieve the intended design structure of the TGN. After examining the morphology and optical properties of ZnO under various growth conditions, the optimal synthesis parameters for ZnO NRs were a precursor mixture consisting of 40 mL of 16 mM a.Zn²⁺, 40 mL of 25 mM HMTA, $V_{\text{NH}_4\text{OH}} = 3$ mL, $t = 2$ h, and a seed thickness of approximately 1 μm.

3.1.3. ZnO grating fabrication

Fig. 5 comprises SEM images of ZnO gratings grown on seed layers through the PR template. Uniform, periodic, and vertical ZnO columns were observed. Each ZnO column consisted of vertically grown ZnO NRs with a height of 1.76 ± 0.05 μm. The spatial density of the ZnO

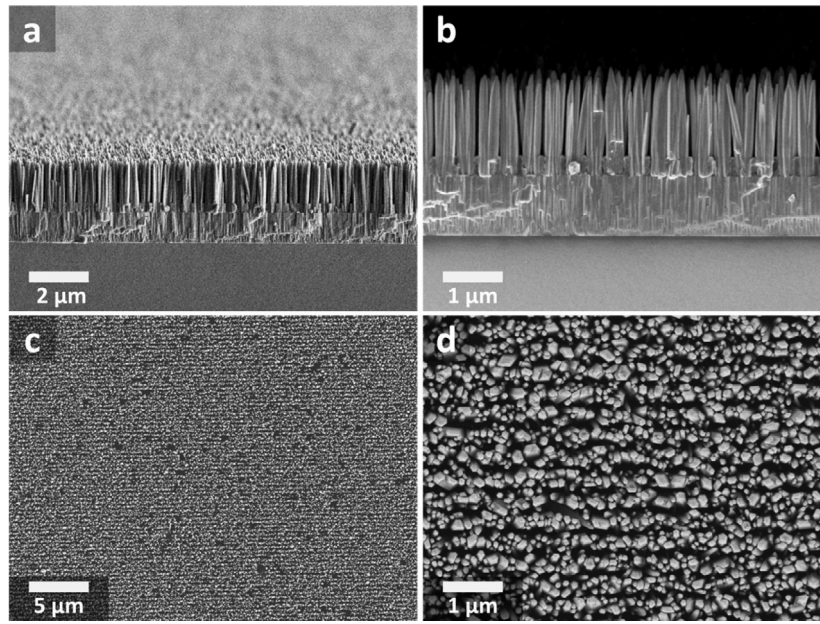


Fig. 5. SEM images of the fabricated ZnO grating viewed at oblique (a), side (b), and top directions (c,d).

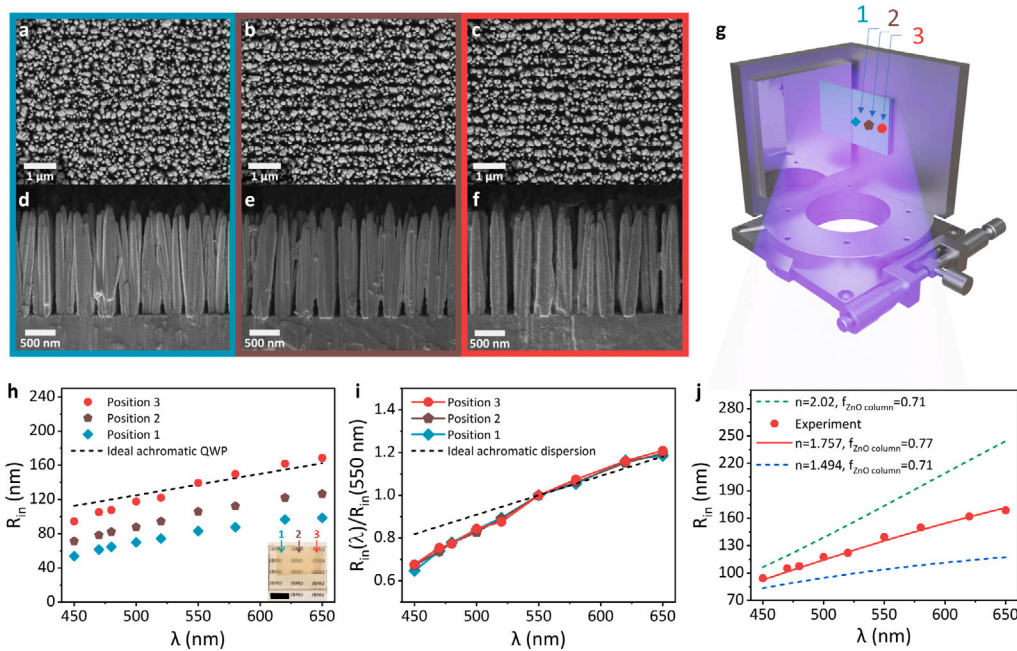


Fig. 6. (a–f) Top and side views of ZnO columns at different positions on the sample. The aperture ratios of the PR patterns at position 1 (a, d), position 2 (b, e), and position 3 (c, f) are 0.83, 0.77 and 0.71, respectively. (g) Measured positions on the sample. (h) In-plane retardation by wavelength at 3 different positions. The inset shows the measured positions. Scale bar: 5 mm. (i) Corresponding relative retardation dispersions. (j) Calculated and measured in-plane sample retardation at position 3.

column was the spatial ratio of ZnO NRs in each ZnO column, which was obtained by top-view SEM imagery (Supplementary Material, Fig. S4). The spatial density must be distinguished from the fill factor f of the ZnO columns, which is defined as the spatial ratio of the ZnO column width divided by the period of the ZnO column. The aspect ratio of each ZnO NR was 6.5 greater than the values found in the literature [27,28].

Fig. 6(a–f) shows the variation in the separation between ZnO columns. SEM images captured at three different positions on the samples revealed slight variance in the periodicity and spatial density of the ZnO NRs, depending on the distance from the Lloyd mirror, as depicted in Fig. 6(g). The spatial densities of the ZnO NRs were 0.595, 0.618, and 0.575 at positions 1, 2, and 3, respectively (Supplementary

Material, Fig. S4). As discussed in the PR grating fabrication section, the aperture ratio of the PR pattern is controlled by the exposure dose. Because the laser light source has a Gaussian intensity profile, the light intensity varies from the center to the edge of the beam, affecting the structure of the PR templates and the subsequently grown ZnO columns. This issue could potentially be mitigated using a beam shaper to flatten the Gaussian profile.

3.2. Optical property characterization

In-plane retardation is defined as $R_{in} = d\Delta n = d(n_x - n_y)$, where n_x and n_y are the in-plane refractive indices and d is thickness of

the retarder. Fig. 6(h) illustrates R_{in} retardation dispersions at three different positions on the sample. An ND of retardation was observed at all positions. At position 3 (red dots), the dispersion of retardation was close to the ideal achromaticity (black dashed line). Retardation was 139 nm at $\lambda = 550$ nm, corresponding to a phase retardation of 91° . $R_{in}(450 \text{ nm}) = 94.4 \text{ nm}$ deviated below the ideal curve by 18.1 nm, while $R_{in}(650 \text{ nm}) = 168.6 \text{ nm}$ was slightly higher than the ideal value of 162.5 nm. The relative dispersion, which was normalized by $R_{in}(550 \text{ nm})$, behaved in a similar way, as shown in Fig. 6(i). The variance of the birefringence at 3 positions was non-negligible, while the relative dispersion trends were nearly consistent (Supplementary Material, section S VI). As $d = 1.76 \mu\text{m}$, a birefringence of $\Delta n(550 \text{ nm}) = 0.08$ was achieved. Generally, the Δn of commercial ND retarders is smaller than that of the PD retarders [13,15–17,20,23]. The Δn of the ND stretched copolymer retarder was approximately 0.001, and that of the ND reactive mesogen film was approximately 0.05. The measured $\Delta n = 0.08$ was greater than that of the previous organic materials. This enabled fabrication of a thinner retarder films.

Fig. 6(j) shows the experimental in-plane retardation at position 3 and the theoretical calculation results. The refractive index of bulk ZnO without voids is 2.02 [48]. The upper limit of the theoretical in-plane retardation can be estimated by assuming that the fill factor of the ZnO columns is the same as the aperture ratio of the PR pattern (i.e., 0.71). This is reasonable because most of the seed layer surface is covered with the ZnO NRs. The calculated upper limit (dashed green line) was greater than the experimental value throughout visible wavelengths. To estimate the lower limit, the average spatial density of each ZnO column at position 3 was analyzed. Next, the refractive index of the ZnO column was calculated using the Lorentz–Lorenz equation by assuming each column was a composite of ZnO and air [49,50]. The spatial density of the ZnO column was 0.575 ± 0.02 (Supplementary Material, Fig. S4), and the refractive index was 1.494 ± 0.029 (Supplementary Material, Table S1). The calculated lower boundary of the in-plane retardation (dashed blue line) was less than the experimental value.

Because the densities of the ZnO NRs decreased moving away from the seed layer, the experimental in-plane retardation data were fitted by assuming that the refractive index of the ZnO column was the average of 2.02 (seed layer surface) and 1.494 (top of NRs), or 1.757. The experimental value was fitted while varying the fill factor of the ZnO columns. The fitted curve (red line) was closely approximated when the fill factor of ZnO columns was 0.77, with a coefficient of determination of $R^2 = 0.99$, as shown in Fig. 6(j). The slightly greater fill factor compared with the aperture ratio of the PR pattern (0.71) can be explained as the effective area of the ZnO columns increased due to the tilted growth of the ZnO NRs. The measurement results at positions 1 and 2 were also closely approximated, with $R^2 > 0.92$ (Supplementary Material, Fig. S5). The well-approximated theoretical calculation results mean that the experimentally measured FB-induced retardation can be explained by Gu and Yeh's model [25].

The Cauchy equation $\Delta n = A + \frac{B}{\lambda^2} + \frac{C}{\lambda^4}$ is generally used to explain the dispersion of the refractive index and birefringence of the medium. Assuming the coefficient C of the quartic term is negligible, B determines the type of dispersion. When the sign of B is positive and negative, Δn represents PD and ND, respectively [33]. The value of B, which was obtained by approximation of position 3 in Fig. 6 was $-0.0285 \pm 0.008 \times 10^{-2}$. On the other hand, that of a commercial ND retarder of RM material is -0.0078 ± 0.002 [16]. This indicates the FB approach has greater potential as a substance for the ND retarders.

The retardation at an oblique angle is given by:

$$Re(\theta, \phi) = \left[(n_x - n_y) + (n_z^2 - n_x n_y) \left(\frac{\cos^2 \phi}{n_x} - \frac{\sin^2 \phi}{n_y} \right) \frac{\sin^2 \theta}{2n_z^2} \right] d, \quad (3)$$

where ϕ and θ are the incident azimuthal angle and polar angle, respectively, n_x and n_y are the in-plane refractive indices, and n_z is the

refractive index along the thickness direction [5]. Given $\phi = 90^\circ$, the retardation is simplified as Eq. (4):

$$Re(\theta) = \left[(n_x - n_y) + \left(\frac{n_x n_y - n_z^2}{n_y} \right) \frac{\sin^2 \theta}{2n_z^2} \right] d. \quad (4)$$

The angular dependence of the retardation is eliminated in Eq. (4), i.e., a wide FOV is achieved, provided that the out-of-plane retardation R_{th} as defined by Eq. (5) converges to zero.

$$R_{th} = \left[\sqrt{n_x n_y} - n_z \right] d \approx \left(\frac{n_x + n_y}{2} - n_z \right) d. \quad (5)$$

The NZ coefficient, used for retarder classification, is given by:

$$NZ = \frac{R_{th}(\lambda)}{R_{in}(\lambda)} + 0.5 = \frac{n_x - n_z}{n_x - n_y}. \quad (6)$$

The condition of $R_{th} = 0$ in Eq. (5) is equivalent to $NZ = 0.5$ in Eq. (6), corresponding to the biaxial z-plate [Fig. 7(e)]. Biaxial retarders are extremely difficult to fabricate by the conventional methods using stretched-films or reactive mesogens. These approaches require control of the molecular orientation or chemical structure along the out-of-plane direction (z-axis) as well as in-plane direction (xy-plane) [53–56]. In contrast, the FB approach offers a relatively straightforward method for controlling the refractive index along the z-axis. This control can be achieved by adjusting the growth time of the ZnO nanorods, a potential strategy for realizing biaxial retarders.

Out-of-plane retardation characteristics of the retarder are presented in Fig. 7. Retardation values $R_e(\theta)$ at various incident light angles were measured at different wavelengths, as shown in Fig. 7(a). The sample was rotated with respect to the fast (y) axis, in Fig. 1(a). Fig. 7(b) displays retardation dispersion at position 3 under incident angles of 0° , 10° , 20° , and 30° . As the incident angle increases, the retardation increases slightly, reaching approximately 13% greater at 30° compared with normal incidence. Because the sample was rotated around the fast axis, the increase in retardation indicates its positive biaxial nature (+b plate). The achromaticity of retardation remained constant across incident angles, as in Fig. 7(c).

Experimentally, the retardation $R_e(\theta)$ can be expressed by [39]:

$$Re(\theta) = \frac{d}{\cos \theta'} \left[\frac{n'_x n'_z}{\sqrt{n_x'^2 \sin^2(\theta' - \theta_0) + n_z'^2 \cos^2(\theta' - \theta_0)}} - n'_y \right]. \quad (7)$$

Here, n'_x , n'_y , and n'_z are the projected refractive indices obtained by fitting $Re(\theta)$ data to Eq. (7). θ' is the transformed incident angle, and θ_0 is the tilt angle of the ZnO rods. n_x , n_y , and n_z are obtained by substituting the respective n'_x , n'_y , and n'_z into the Eqs. (8), (9), and (10).

$$n_x = \frac{n'_x n'_z}{\sqrt{n_x'^2 \sin^2(-\theta_0) + n_z'^2 \cos^2(-\theta_0)}} \quad (8)$$

$$n_y = n'_y \quad (9)$$

$$n_z = \frac{n'_x n'_z}{\sqrt{n_x'^2 \cos^2(-\theta_0) + n_z'^2 \sin^2(-\theta_0)}} \quad (10)$$

The $R_{in}(\lambda)$, $R_{th}(\lambda)$, and NZ are summarized in Fig. 7(d). The sign of R_{th} is negative, and its absolute values are significantly greater than R_{in} , which means n_z is greater than the average of n_x and n_y . NZ coefficients of the samples within a range of $(-2, 0)$ were obtained. Referring to Fig. 7(e), the grating-structured ZnO corresponds to the +b plate. To the best of our knowledge, the biaxiality of the ND retarders has been rarely reported. The RM-based ND retarders are generally negative biaxial due to the planar orientation of the molecular plane on the surface [51]. The polymer-based ND retarders are also negative biaxial and have NZ coefficients greater than 1 [12]. This intrinsic nature of the organic material-based retarders limits the potential improvement of the FOV property, which is difficult to overcome.

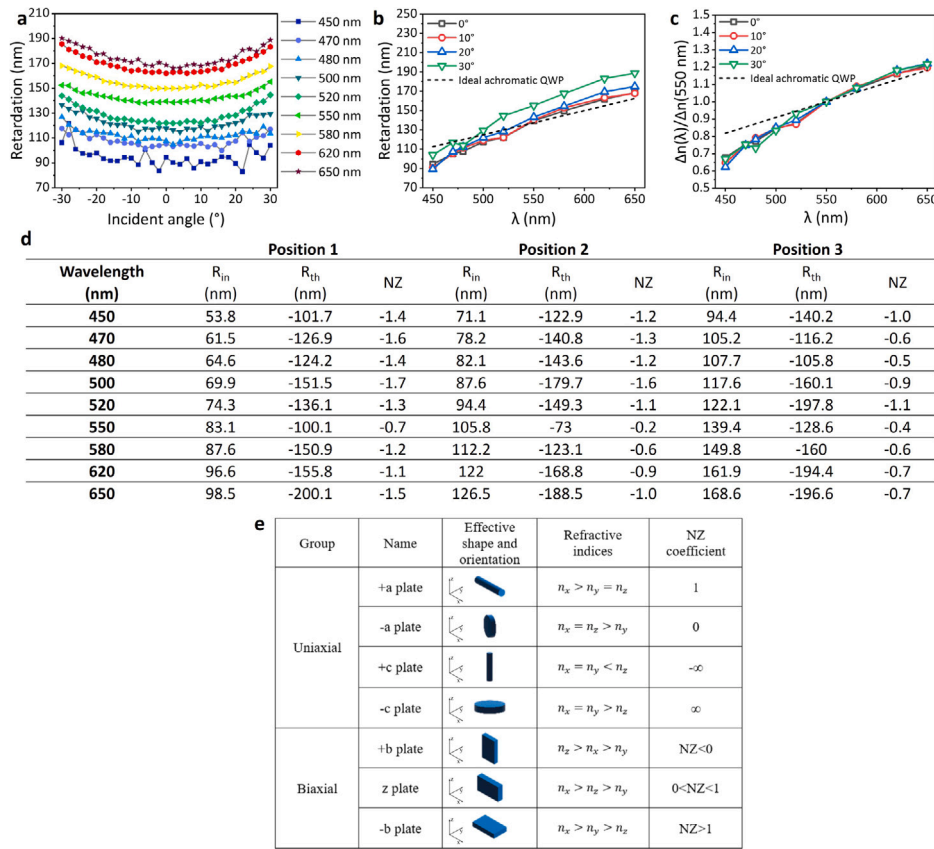


Fig. 7. (a–c) Retardation by incident angle (a), retardation dispersion with incident angles of 0°, 10°, 20°, 30° (b), and relative birefringence dispersion measured on position 1 (c). (d) Specifications of the FB retarder at 3 positions. (e) Classification of retarders [51,52].

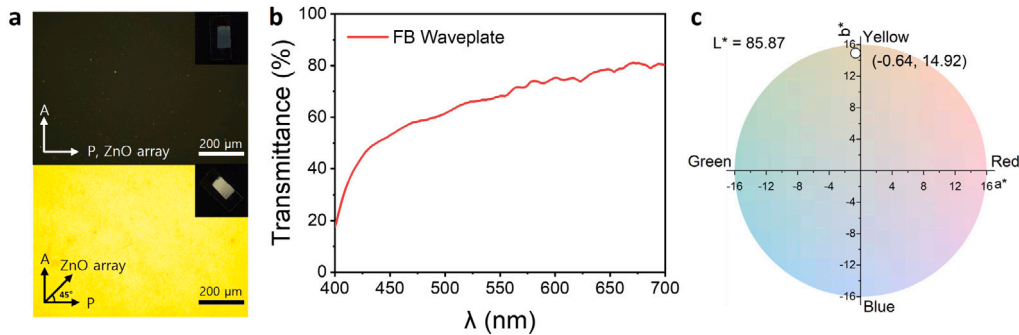


Fig. 8. (a) POM images with ZnO column direction at 0° and 45°, the insets are sample photos of areas between two crossed polarizers. (b) Transmittance spectrum of the FB retarder. (c) CIElab color coordinate of the FB retarder.

However, the +b plate and z-plate, which have been rarely reported, can be regarded as advanced optical components with wide FOVs [55–57]. The +b behavior of our FB retarder can be explained by its ZnO grating structure. Considering the effective shape and orientation, each ZnO NR can be regarded as the +c plate ($NZ \rightarrow -\infty$) [58], while the periodic ZnO column structure corresponds to the z plate ($0 < NZ < 1$). Therefore, ZnO columns consisting of ZnO NRs can show an intermediate property of +c and z plates, corresponding to the +b plate ($NZ < 0$). Based on the flexibility of our fabrication method, different types of achromatic retarders, including uniaxial and biaxial retarders, can be realized.

Fig. 8(a) shows polarized optical microscopy images of the FB retarder. A clear contrast in brightness between 0° and 45° orientation indicates a uniform alignment of the ZnO rods on the substrate. Despite the optimization of ZnO morphology, scattering was not completely eliminated. Fig. 8(b) shows the diffuse transmittance spectrum of the

FB retarder. Transmittance at a wavelength of 550 nm was 68%, and the average transmittance from 400 to 700 nm was 65%. Transmittance dropped significantly from 80% at 700 nm to 20% at 400 nm. Light with a shorter wavelength experiences greater scatter, resulting in lower transmittance, while light with a longer wavelength exhibits higher transmittance. Additionally, the absorption of ZnO in the near-UV region contributes to reduced transmittance at shorter wavelengths. For further transparency improvement, MOCVD or vapor–liquid–solid method are potential approaches for reducing the diameter and increasing the density of ZnO nanorods [59–63]. The coordinates of the transmitted color in the CIELAB color space in Fig. 8(c) indicate a yellowish tint, consistent with the obtained transmittance spectrum. For applications in displays, it is crucial to neutralize color scatter to prevent impact on the color production capability. Reducing the diameter of ZnO NRs can enhance transmission at shorter wavelengths, achieving a more neutral color [42].

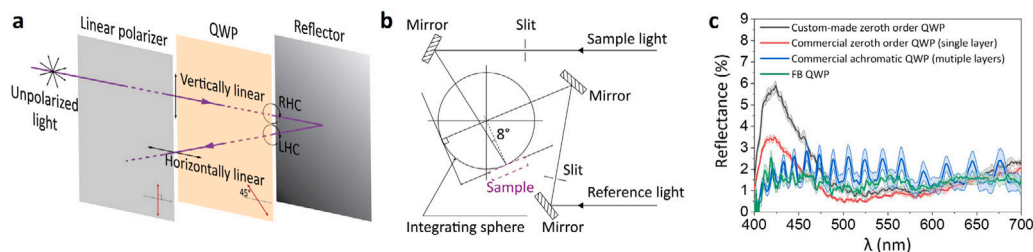


Fig. 9. (a) Antireflection structure using a linear polarizer and a QWP. Red arrows indicate the optical axis of components. (b) Optical setup of reflectance measurement (redrawing of a published structure from Shimadzu [64]). (c) Measured reflectance of four types of QWP. Upper and lower limits represent the measurement uncertainty.

3.3. Antireflection demonstration

To evaluate the performance of the proposed retarder, the antireflection (AR) efficiency was measured using a quasi-circular polarizer composed of a linear polarizer and various QWPs. The slow axis of the QWP was set at 45° to the transmission axis of the polarizer. Initially, incident unpolarized light was converted to vertically linearly polarized light upon passing through the linear polarizer. This light then became right-handed circularly polarized (RHC) after traversing the QWP. Upon reflection from a reflector, the RHC light was transformed into left-handed circularly polarized (LHC) light. As the LHC light passed back through the QWP, it was converted into horizontally linear-polarized light, which was blocked by the polarizer, preventing reflection. We measured the reflectance spectrum from 400 to 700 nm to evaluate the AR performance. The reflectance measurement was carried out using a UV–visible light spectrometer equipped with an integrating sphere (ISR 2600), as depicted in Fig. 9(a) and Fig. 9(b). Measurement uncertainty is analyzed in the Supplementary Material. Four types of retarders were tested: a custom-made zero-order QWP (fabricated by LC242 following the process in a previous publication [52]), a commercial zero-order QWP (#49219, Edmund), a commercial achromatic QWP (AQWP10M-580, Thorlabs), and our FB QWP. The AR performance is presented in Fig. 9(c). The commercial and custom-made zero-order QWPs, which are designed for a wavelength of 532 nm, showed similar reflection spectra, with reflectance less than 1% throughout the 470–600 nm range. The reflectance rose rapidly beyond this range. Below 470 nm, the average reflectance values of the commercial and custom-made zero-order QWPs were 2.44% and 4.14%, respectively. Despite the flat fluctuating reflection spectrum provided by a commercial achromatic QWP, its average reflectance through the measured range was 1.67%. Due to achromatic retardation and lower angular dependence (Supplementary Material, Fig. S6), the FB QWP had a flat reflectance with an average value of 1.37%. Overall, the FB QWP showed advanced properties when used in antireflection components, implying a potential use in organic light-emitting diode (OLED) panels.

4. Conclusion

We successfully demonstrated an FB-based achromatic QWP operating in visible light using a templated-grown nanostructure. Our FB retarder exhibited ND retardation with a phase retardation of 91° at 550 nm. To achieve the desired structural ZnO grating with vertical, dense, and transparent ZnO NRs, various growth parameters, including precursor concentration, growth duration, addition of ammonium, and seed layer thickness, were optimized. In addition, the achromaticity of the FB retarder did not change with an increase in the incident angle, retaining a wide FOV. An antireflection layer for OLEDs using our QWP performed competitively compared with other QWPs. Due to the flexibility in structure configuration, our proposed method of combining LIL and hydrothermal growth can enable a selective fabrication of uniaxial and biaxial retarders (i.e., programmable achromatic retarders with wide FOVs).

CRediT authorship contribution statement

Manh-Thang Tran: Writing – review & editing, Writing – original draft, Visualization, Methodology, Formal analysis, Data curation. **Nguyen-Hung Tran:** Writing – review & editing, Visualization, Supervision, Methodology, Formal analysis, Data curation. **Ji-Hoon Lee:** Writing – review & editing, Writing – original draft, Supervision, Resources, Project administration, Methodology, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Ji-Hoon Lee reports financial support was provided by National Research Foundation of Korea. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary material related to this article can be found online at <https://doi.org/10.1016/j.optlastec.2025.113011>.

Data availability

Data will be made available on request.

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