Polarization holographic gratings in PAZO azopolymer recorded with different recording-beams polarizations

G Mateev1,2, L Nedelchev1,2, D Ivanov1, R Tomova1, P Petrova1, V Strijkova, N Berberova1 and D Nazarova1

1Acad. J. Malinowski Institute of Optical Materials and Technologies, Bulgarian Academy of Sciences, Acad. G. Bonchev Str., Bl. 109, 1113 Sofia, Bulgaria
2University of Telecommunications and Post, 1 Acad. St. Mladenov Str., 1700 Sofia, Bulgaria

E-mail: g_mateev@abv.bg

Abstract. Polarization holographic gratings (PHG) were recorded using a laser emitting a wavelength of 491 nm in thin films of the (poly[1-[4-(3-carboxy-4-hydroxyphenylazo)benzenesulfonamido]-1,2-ethanediyl, sodium salt]) azopolymer, shortly denoted as PAZO. Thin azopolymer films with various thicknesses were spin-coated on plastic and glass substrates. Four different polarization states of the recording beams were used, and the results compared: a) two vertical linear polarizations, b) horizontal and vertical linear polarizations, c) linear polarizations at +45° and −45° relative to the recording plane, and d) two orthogonal circular polarizations – left- and right-handed (LCP and RCP). The diffraction efficiency in the +1 diffraction order was monitored in real time by a probing laser beam at the wavelength of 635 nm. The results indicate that the highest diffraction efficiency is achieved when recording with orthogonal polarizations – linear at ±45° or left and right circular. This is explained by the ability of the azopolymer material to record the variations in the polarization state of light better than the variations in its intensity. The holographic gratings obtained can be used to enhance the light-extraction efficiency of an OLED device.

1. Introduction

Azo-containing polymers are widely investigated materials because of their photo-anisotropic properties [1-3]. The photoinduced phenomena, such as linear and circular photoinduced birefringence, are due to the trans-cis-trans isomerization in azomaterials when illuminated by polarized light.

As measured earlier by our group, the maximal photoinduced birefringence of the azopolymer PAZO is \( \Delta n \approx 0.07 \) [4]. This high photoinduced birefringence is of essential importance for achieving high efficiency of polarization diffraction gratings. These gratings are interesting because of their achromaticity and the theoretical possibility to achieve 100% diffraction efficiency (DE), even in thin films [2].

There are different ways to inscribe polarization gratings. In this study, we compared the efficiency of gratings recorded with four different polarization states of the recording beams: a) two vertical linear polarizations (denoted shortly as VV), b) horizontal and vertical linear polarizations (HV), c) linear polarizations at +45° and −45° relative to the recording plane (PM), and d) two orthogonal circular polarizations – left- and right-handed (LCP and RCP). The diffraction efficiency in the +1 diffraction order was monitored in real time by a probing laser beam at the wavelength of 635 nm. The results indicate that the highest diffraction efficiency is achieved when recording with orthogonal polarizations – linear at ±45° or left and right circular. This is explained by the ability of the azopolymer material to record the variations in the polarization state of light better than the variations in its intensity. The holographic gratings obtained can be used to enhance the light-extraction efficiency of an OLED device.

3 To whom any correspondence should be addressed.
circular polarizations – left- and right-handed (LR). The use of different polarizations results in different polarization patterns of the interference field on the samples surface and in different properties of the gratings and values of their DE.

2. Theory

The properties of the polarization gratings were investigated theoretically in [2]. When one uses two pumping beams with defined polarization, one obtains a specific interference field on the sample. The cases with VV, HV, PM and LR polarizations of the pumping beams are shown in figure 1.

![Figure 1](image)

**Figure 1.** Interference field patterns for different polarizations of the two pumping beams: (a) VV, (b) HV, (c) PM, and (d) LR. \(2\delta\) is the phase difference between the pump beams.

The measured diffraction efficiency strongly depends on the polarization of the reading beam. Hence, for each of these cases, one uses a reading beam with polarization coinciding with the polarization of one of the recording beams. Thus, for the HV, VV and PM geometries, one obtains diffracted beams at \(+1\) orders, while for the LR geometry one should expect to get diffraction only in the \(+1\) order.

In the case when the photoinduced changes are relatively small in materials with linear and circular photoanisotropy, and one irradiates the medium with two beams with vertical and horizontal polarizations, the intensities of the \(+1\) diffracted orders are:

\[
I_{+1} = \frac{1}{4} \left[ (\Delta \phi^L)^2 + (\Delta \phi^C)^2 \pm 2 \Delta \phi^L \Delta \phi^C \sin(2\alpha) \right],
\]

where \(\Delta n^L\) and \(\Delta n^C\) are the amplitudes of modulation of linear and circular birefringence and \(\alpha\) is the azimuth of the linearly polarized reading laser beam [2]. If the azimuth \(\alpha\) is \(45^\circ\), one obtains the maximal value for the intensity of the \(+1\) diffracted order and the minimal value for the intensity of the \(-1\) diffracted order. Using these two values, one can calculate the linear and circular birefringence. In the case when the two values are equal, the circular birefringence is 0:

\[
\Delta \phi^C = I_{+1, max}^{1/2} + I_{-1, min}^{1/2}, \Delta \phi^C = I_{+1, max}^{1/2} - I_{-1, min}^{1/2}.
\]

The calculations are made under the assumption that the recording beams have equal intensities.

3. Experiments

3.1. Samples preparation and thickness measurement

We prepared samples for the experiments by dissolving two different concentrations of PAZO azopolymer in water, namely 100 mg/ml and 125 mg/ml, and then spin-coating the solutions on glass substrates at 1500 rpm for 30 s. As a result, thin homogeneous films were formed for optical testing.
Their thickness was determined by a Filmetrics F20 device using the non-destructive method of thin-film interference. The thicknesses measured were 470 nm for the samples produced from the solution with concentration 100 mg/ml, and 790 nm for the others.

3.2. Recording of the PHG

The experimental setup for recording the PHG is presented in figure 2. For recording, a 491-nm diode laser was used, while the reading diode laser was operating at 635 nm. The recording laser radiation was split into two beams by a polarizing beam-splitter (PBS). Half- and quarter-wave plates were used to adjust the polarization state of the pumping and reading beams according to the four different geometries. The recording beams had diameters of 1 cm, and power of 10 mW. The recording angle was $\theta = 30^\circ$.

![Figure 2. Experimental setup for the PHG recording.](image)

The experimental procedure was as follows: First we measured the background with the reading laser for one minute. Then we started the holographic recording with the pumping laser irradiating the sample for ten minutes. After ten minutes, we turned the pumping laser off and measured the relaxation. The power of the transmitted first-order diffracted beam was measured continuously by a power meter and the data stored on a computer. After five minutes of relaxation, we measured the power of the other diffracted orders (if any). Using the results of these measurements, we calculated the temporal dependence of the diffraction efficiency (DE).

3.3. Results

We recorded PHG in thin azopolymer films with two thicknesses and four different cases of the recording beams’ polarization, denoted as HV, VV, PM and LR. In each case, the polarization state of the reading laser coincided with the state of one of the recording beams. Figure 3a shows the results for the DE as a function of the time for the films with 470-nm thickness, figure 3b shows the same for the films with 790-nm thickness.

For the 470-nm, one can see that the LR case leads to a much higher DE than the other cases. This result is repeated with the thicker layers (790 nm). Also, for the thicker layers one can see clear differences between all cases. Thus, the highest DE is obtained with the LR geometry, followed by the PM and VV geometries, with the lowest DE obtained with the HV geometry. The above is explained by the fact that in the LR case the resultant optical field has linear polarization only with a varying azimuth.
Figure 3. Diffraction efficiency (DE) of the gratings recorded at different polarization configurations: (a) samples with a thickness of 470 nm, and (b) samples with a thickness of 790 nm.

In all other cases, the field has elliptical polarization. On the other hand, the PAZO azopolymer exhibits a significant photoinduced linear, but no circular, birefringence. Thus, the LR geometry offers a better opportunity to utilize the PAZO properties. Also, in the LR geometry there exists one diffracted beam only, while for all other cases there are two approximately equal diffracted beams in ±1 order, in full accordance with the theoretical expectations. This fact doubles up the DE in LR geometry.

Figure 4. (a) Comparison of the DE for LR polarizations and different thickness of the samples – 470 nm and 790 nm. (b) Diffraction efficiency of the 790-nm thick sample for LR polarizations and recording time 1.5 hours.

For the geometry with the highest DE (LR), we compared the performances of the PHGs formed in the films of different thickness. As expected, the thicker sample yielded a higher (by a factor of 3.5) DE. Further, we observed that 10 minutes of recording were sufficient to reach saturation for the thin film, but clearly not enough for the thick one. We, therefore, recorded a PHG with longer exposure in the sample with 790-nm thickness, as shown in figure 4b, and obtained about four times as high DE.

4. Conclusions
We prepared samples of PAZO azopolymer with two different thicknesses and recorded PHGs in four cases (VV, HV, PM and LR recording geometries) for the two thicknesses. The results showed that the highest DE is achieved using an LR geometry, because in this geometry the resulting optical field is
linearly polarized and there is only one diffracted beam. Furthermore, the thicker sample yields a higher DE but requires much longer time to reach saturation.

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**References**
[1] Todorov T, Nikolova L and Tomova N 1984 Polarization holography. 1: A new high-efficiency organic material with reversible photoinduced birefringence *Appl. Opt.* **23** 4309
[2] Nikolova L and Ramanujam P S 2009 *Polarization Holography* (Cambridge University Press, Cambridge)
[3] Wang X 2017 *Azo Polymers: Synthesis, Functions and Applications* (Springer-Verlag, Berlin)
[4] Nedelchev L, Nazarova D, Mateev G and Berberova N 2015 Birefringence induced in azopolymer (PAZO) films with different thickness *Proc. SPIE* **9447** 94471I-1