Abstract

Line- and hexagonal-like periodic textures were fabricated on aluminium zinc oxide (AZO) using direct laser interference patterning method. It was found that hexagonally patterned surfaces show a higher performance in both transparency and diffraction properties compared to line-like textured and non-patterned substrates. Furthermore, the electrical resistance of the processed AZO coated substrates remained below the tolerance values for transparent conducting electrodes.

Keywords: Light management; TCO; AZO; Direct laser interference patterning; Laser ablation; Organic Photovoltaics

1. Motivation / State of the Art

Thin film electrodes for organic photovoltaics and electronics are a major issue at present. For enhanced performance of organic solar cells and light emitting diodes, an optimized saturation of light is important. This so-called light management can be achieved by surface patterning of the front respectively back electrodes of the cell with a microstructure, which has the same diffractive effect on light like a Bragg grid [1-3].

The method of Direct Laser Interference Patterning (DLIP) enables a fast and simple large-scale fabrication of microscale patterns with spatial periods normally between 200 nm and 50 μm. Because high energetic pulsed laser systems are utilized, this method requires only one single processing step to generate the surface textures, avoiding the use of chemicals (for example for etching or development) or masks. Depending on the laser-type as well as materials engaged in the procedure, the processing speed varies from 10 to 100 cm²·s⁻¹. A broad variety of materials such as polymers, metals, semiconductors and transparent conducting oxides (TCOs) have been already patterned using DLIP. The basic pattern that can be created with DLIP using a two-beam set-

* Corresponding author. Tel.: +49 351 83391 3007
E-mail address: andres-fabian.lasagni@iws.fraunhofer.de
up (Fig. 1a) is a relief of parallel lines ("line-like pattern", see Fig. 1b). For grid-like patterns, the substrate has to be turned by an angle between 0°-90° and patterned a second time with the line-like texture. Using three (Fig. 1c) or more beams, more complex patterns can be produced [4-9]. The spatial period Λ of the pattern can be calculated using equation 1:

\[
\Lambda = \frac{\lambda}{2 \cdot \sin(\alpha)}
\]  

with \(\lambda\) being the laser wavelength, 2\(\alpha\) the angle between the two partial laser beams (Fig. 1a). In this work, aluminum zinc oxide (AZO) coated float glass substrates were patterned with line-like and hexagonal textures using DLIP. The intention was to improve the efficiency of organic solar cells and organic LEDs by enhanced light management.

Fig. 1. Scheme of (a) a two beam set-up for the Direct Laser Interference Patterning (DLIP) method and simulation of the intensity distribution for (b) a two beam and (c) three beam interference pattern.

2. Experimental

900 nm thin films of aluminum doped zinc oxide (AZO) were magnetron-sputtered on 3 mm thick float glass substrates. Subsequently, the substrates were cut into squared pieces with an edge length of 2.54 cm. After that, all samples were irradiated with laser pulses ranging from 275 to 300 mJ\(\text{cm}^{-2}\). The principle beam emitted from the laser source is split into two equal partial beams by 50-50 a beam splitter (Fig. 1a). An
arrangement of three highly reflective mirrors guides these two beams to the sample, where they congruently overlap. For the patterning process only single pulses were used (further information about the experimental setup can be found in [5, 6]).

The prepared samples were structured with line-like and hexagonal textures. In order to fabricate hexagonal patterns, the line-textured sample was turned by 60° and processed again in the same way. The hexagonal characteristic originates from the material ablation processes at the crossing sections between the lines. A pulsed Nd/YAG-laser with pulse duration of 10 ns, a repetition rate of 10 Hz and a wavelength of 355 nm were utilized for the experiments.

The DLIP-processed samples were imaged with a photospectrometer which is based upon an optical two path system to measure sample and reference simultaneously. A polarization filter was used to minimize measurement errors caused by internal polarization effects of the photospectrometer. SEM-scans were realized with a Philips XL-30 ESEM SEG microscope at a tilted object slide with an angle of 30°. Pattern depth and surface features were measured on squares of 10 x 10 μm by atomic force microscopy using a Jeol JSPM 5200 AFM-device.

3. Results and Discussion

The AZO coated glass substrates were irradiated varying laser fluence and pulse number. Using a laser fluence of 288 mJ.cm⁻², high qualitative surface patterns were produced (Fig. 2) [10].

![Fig. 2 SEM images of line-like and hexagonally patterned AZO thin film samples [10].](image-url)

Higher fluences destructed the pattern while lower energies led to imperfect textures. In all cases, the grained AZO surface was partially molten by the beam energy and formed into line-like (Fig. 1a), and pillar-like (Fig. 1b) shapes with comparably smooth superficies.

Optical spectrometric measurements of the treated surfaces showed an increase of the optical transmittance due to the partial laser ablation of the AZO thin film (Fig. 3a). For both line-like and pillar-like patterns, the diffuse transmission was significantly increased (Fig. 3b) and therefore presenting a higher haze value (quotient between diffuse and total transmission). Furthermore, the improvement for the pillar-like hexagonal texture is clearly higher than for the line-like structure [1,10].
The AZO sheet resistance was also measured using a four point method. This technique uses four terminals to sense the ohmic resistance without the influences by impedance of contact and wiring. The results showed that the electrical resistance smoothly increased with ascending laser fluences i.e. increasing pattern depth. This performance corresponds with the model of Fuchs and Sondheimer that predicts an increasing electrical resistance at a shrinking layer thickness [11, 12]. However, the measured electrical resistance for all pattern geometries and laser processing parameters are close to the initial Resistance ($2.31 \Omega_c$) of the AZO film (an increase of 4.3 and 11.3% are reported for the line- and hexagonal-like structure for a laser fluence of 288 mJ.cm$^{-2}$, respectively).

Although the results here presented show an efficient method to improve light management on AZO coatings, DLIP has been utilized in the past basically only at the laboratory scale. Generally, the laser interference experiments are carried out using high power pulsed Nd:YAG laser systems. Typical pulse durations range from 6 to 30 ns. In most of the cases, the infrared wavelength (1064 nm) of Nd:YAG lasers can not be absorbed properly by several materials, the wavelength is normally shortened using non-linear crystals. For instance, through frequency doubling the photons interacting with a nonlinear material are combined to form new photons with twice the energy, and therefore half the wavelength of the initial photons. Similarly,
Ultra violet (UV) wavelengths (e.g. 355 and 266 nm) can be generated by third (frequency tripling) or fourth (frequency quadrupling) harmonic generation.

Recently, compact interference patterning systems have been developed [13]. These systems offer the possibility not only to process planar surfaces but also complex three dimensional mechanical components including mechanical seals, piston rings, and bearings. Moreover, the system can be adapted in order to precisely fabricate micro or nanometer patterned arrays. The specification data of such a system is given in Table 1. Figure 4, illustrates a DLIP system to produce 1.5 μm line-like periodic arrays for the treatment of 300 x 300 mm² substrates. The DLIP optical head is located 50 cm over the working plane and has a size of 15 x 20 x 30 cm³.

Table 1: Specification Data of DLIP Systems at Fraunhofer IWS [13].

| Specification                      | Details                                      |
|-----------------------------------|----------------------------------------------|
| Working distance                  | 5 - 60 cm                                    |
| Working area                      | up to 300 x 300 mm² (standard dimension, can be also enlarged) |
| Laser power (wavelength dependent)| up to 25 W (Infra-red)                       |
| Pulse energy (wavelength dependent)| up to 2.5 J (Infra-red)                      |
| Laser wavelengths                 | 1064, 532, 355 and 266 nm                    |
| Repetition rate                   | 10 - 50 Hz                                   |
| Optical Head dimensions           | ~ 15 x 20 x 30 cm³                           |
| Structure period                  | from ~ 180 nm to 30 μm                       |
| Fabrication speed                 | 1 - 100 cm²/s (material dependent)           |

![Fig.4 Schematic of DLIP System (Fraunhofer IWS) [13].](image-url)
4. Conclusions

In this study, thin film layers of Aluminum doped Zinc oxide (AZO) were surface textured with line- and hexagonal-like patterns using two beam Direct interference laser patterning (DLIP). It was found that, especially for the hexagonal patterns, the treated substrates show a significantly higher haze profile as well as a slightly higher transmittance compared to the unpatterned reference samples. On the other hand, a minor increase of the AZO sheet resistance was observed, which can be attributed to the local reduction of the film thickness at the interference maxima positions, however the reported values are still acceptable for their use as functionalized substrates for organic electronics. Furthermore, it could be proven that DLIP provides a fast, cost-effective and efficient method for the production of transparent thin film AZO electrodes with enhanced light management properties.

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