Nano- and Micro-Corruations in Photo-Aligned Polymer Thin-Films: Correlations Between Topologies and Directional Light Scattering

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Isotropic and anisotropic nano- and micro-corruations in polymer thin-films are shown to be independently tunable via photo-controlled diffusion of liquid crystalline pre-polymer molecules on substrate surfaces. Amplitude and shape of MC-topologies are quantitatively correlated with light scattering. Scattering from monomer corrugated (MC)-topologies is correlated with the averaged power spectral density (PSD) function of MC-height fluctuations and shown to be proportional to PSD. Paper-white MC-diffusers with optimal field of view, minimal glare and brightness enhancement exceeding 25 are demonstrated. Photo-patternning of MC-topologies is shown to render multidirectional diffusers and diffuse reflectors feasible. Generation of ultra thin MC-reflectors does not require micro tools and renders integration into flat panel displays feasible. [DOI: 10.1143/JJAP.42.6896]

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1. Introduction

Diffuser- and diffuse-reflector films with directional properties have become integral parts of reflective liquid crystal displays (LCDs). They redistribute incident light into a defined viewing cone such that uniform illumination, maximum brightness and minimal glare result. To meet the minimal power consumption requirements of portable displays and to make optimal use of environmental light — such as in mobile phones, or electronic books — efficient diffuse reflectors with directional reflectivity are key.

Conventional reflective LCDs use metallic-type reflectors that are brightest at the glare angle; i.e. at the angle of maximum reflection. However, due to specular reflection, legibility and contrast markedly degrade at this critical angle. The viewer is forced to tilt the display off-glare, which dims the image and de-saturates colors. To remedy this problem, holographic reflectors1—4) and reflectors with asymmetric, mechanically machined topologies5—7) have been developed. However, because of their limited scattering efficiency and coloration, present reflectors are not yet optimal. Their limitations are: peak brightness over a narrow angle of view, colored appearance, light-scattering that depends on the state of polarization of incident light and machining limitations. Moreover, most existing reflectors are too thick for integration into displays.

We have recently shown that high-resolution nano- and micro-topologies can be generated in sub-micron thin polymer films without use of nano-tools or high-resolution photo-masks. The new technology was designated monomer corrugation (MC)-technology.8,9) Isotropic as well as anisotropic MC-film topologies were shown when exposing a thin-film of MC-material to flux uv-light; where the MC-material in its simplest form consists of two types of functional monomers A and B. Component A is photosensitive and cross-linkable under uv-irradiation, whereas component B is optically inert. Light-induced differential surface diffusion of A and B and simultaneous cross-linking of the pre-polymer molecules A generate the corrugations.8,9) Moreover, we have shown that photo-aligned and photo-patterned substrate surfaces10,11) induce anisotropic surface diffusion in mesogenic pre-polymer MC-materials, enabling anisotropic MC-polymer films with liquid crystalline properties.

The MC-topologies range from nano- to micro-meters over arbitrarily large areas and substrate shapes.8) The MC-technology is applicable to virtually any planar or non-planar substrate. MC-films can be photo-patterned and integrated with our surface-aligned liquid crystalline polymer (LCP) functional thin-films.10—14) MC-applications are: ultra-thin, nano-grooved aligning layers for liquid crystal molecules in optically bi-stable LCDs, strongly birefringent optical retarders based on low birefringence materials, multidirectional diffusers and reflectors, wide-view films with corrugated topologies, optical security elements, corrugated reflective and transmissive polarizers or diffraction optical thin-films.8,9,15,16)

In this paper we show that amplitude- and shape distribution of MC-topologies can independently be tuned. Therefore and because amplitude and shape of the grooves of an optical grating individually contribute to light scattering,17,18) scattering synergies can be achieved. Diffuse MC-reflectors are shown to defy most of the deficiencies of single-frequency gratings, such as restricted field of view, coloration or color shift versus angle of view. Quantitative correlations between light scattering, MC-groove amplitude and groove periodicity are derived. The optical characteristics of MC-diffusers and reflectors are shown to correlate with the amplitude distribution, groove periodicity and groove shape of MC-corruations. Moreover, the power spectral density (PSD) of MC-corruations is shown to be proportional to MC-light scattering. This enables ultra-thin diffusers or directionally diffuse MC-reflectors for achromatic, reflective LCDs with large optical gain, high contrast and minimal glare. Photo-patterned MC-topologies are shown to further enhance directional reflectivity of MC-diffusers and reflectors.

2. Theory; Light Scattering by Thin Film Topologies

Figure 1(b) schematically depicts the optics of a single-frequency sinusoidal relief diffraction grating with period A and amplitude h. Light of intensity I0 and wavelength \( \lambda \) incident at the angle \( \theta_i \) is diffracted into a plane perpendicular-
Fig. 1. (a) Schematic scattering geometry: polar angle $\theta_s$; azimuthal angles $\phi_s$; incident light intensity $I_i$; scattered light intensity $I_s$. (b) Schematic of a single frequency sinusoidal grating illuminated perpendicular to the groove direction ($\phi=180^\circ$) with the diffracted orders $m=0, 1, 2$ following from grating eq. (1). Zero order ($m=0$) specular reflection $R_0$ in polar direction $\theta_0=\theta_i$; first order ($m=\pm 1$) diffraction $R_1$ along $\theta_1$; etc. $T_0$ and $T_1$ are the respective zero and first transmission orders.

ular to the groove direction. $\theta_{in}$ are the angles between the diffracted rays $R_m$ of order $m (m = 0, \pm 1, \pm 2, \text{etc.})$ and the normal to the grating surface. The diffracted rays obey the grating equation:\footnote{17,18}

$$m\lambda f = \sin \theta_i - \sin \theta_s,$$

(1)

where $f = 1/\Lambda$ is the groove frequency. For a particular wavelength $\lambda$, only diffraction orders $m$ which meet

$$|m\lambda/\Lambda| < 2$$

(2)

actually exist. In Fig. 1, specular reflection $R_0$ at the glare angle $\theta_0 = \theta_i$ corresponds to zero order diffraction ($m=0$), whereas the reflections $R_1$ and $R_2$ are the first and second orders respectively. The dielectric grating in Fig. 1 can be operated in reflection or in transmission. $T_0$ and $T_1$ are the zero and first order transmissions. According to coupled wave theory the diffraction efficiency of a grating for a given wavelength of incident light depends on period, amplitude and shape of its grooves.\footnote{17,18} The geometry of a single frequency grating can therefore be chosen such that, optimal diffraction into a specific angle of reflection occurs for this wavelength. Single frequency gratings exhibit narrow reflection/transmission cones around $\theta_{in}$. Therefore, diffusers and reflectors based on holographic or photolithographic gratings are usually coloured and exhibit narrow fields of view. Moreover, their scattering often depends on the state of polarization of incident light.

To remedy the deficiencies of single frequency gratings requires gratings with more complex topologies, such as multi-frequency groove patterns combined with amplitude-modulated groove topologies. Such gratings could meet the demanding scattering characteristics required for display applications, such as high gain, broader angle of view, white appearance and polarization-independent reflectance. For a particular set of grating parameters $- f = 1/\Lambda = \text{groove frequency}$, $\phi_s = \text{angle of light incidence}$ and $\phi_{in} = \text{diffraction angle}$ — grating eq. (1) holds for more than one wavelength. Therefore, several discrete wavelengths $\lambda_i$ exist which — when multiplied by successive integers $m$ — satisfy the condition for constructive interference of eq. (1). Analogously for a particular set of incident wavelengths $\lambda_i$ and angles ($\phi_i$, $\phi_{in}$), eq. (1) holds for more than one groove frequency; where successive diffraction orders may overlap within a given viewing cone. From the grating equation it follows for example that first order ($m=1$) reflected light of wavelength $\lambda$ will coincide with second order ($m=2$) diffracted light of wavelength $\lambda/2$, etc. If the groove pattern of a grating consists of different frequencies, the same holds for each groove frequency; i.e. incident light of wavelength $\lambda$ is diffracted by the groove frequency $2\phi_i$ into the first order angle $\theta_1$ ($m=1$) which coincides with the direction of the second order diffraction ($m=2$) of groove frequency $\phi_i$, etc. As a result the diffraction intensity, or brightness of the grating at $\theta_i$ increases. The field of view of multi-frequency gratings can be enlarged by diffracting the 1st order of wavelength $\lambda$ by the groove frequency $\phi_i$ into $\theta_i$, while diffracting the 1st order of $\phi_i$ into a different angle $\theta_i$.

In the following we show that surface scattering from MC-reflectors and diffusers can be described by their respective bi-directional reflectance or transmission distribution functions BRDF and BTDF.\footnote{19-26} The BRDF function relates the differential luminance $dL_i$ of incident light on a surface with the reflected and scattered differential luminance $dL_s$ and correlates the respective intensities with the angles of incidence and reflection and with polarization. The dependence between BRDF function and angular-resolved, differentially scattered light intensity ($dL_s/d\Omega$) follows from:\footnote{23}

$$BRDF_{\alpha,\beta}(\theta_i, \phi_i, \theta_s, \phi_s) = \frac{dL_s}{d\Omega_i} = \frac{1}{\cos \theta_i} \frac{1}{I_i} \left( \frac{dL}{d\Omega} \right)_{\alpha,\beta},$$

(3)

where the angles ($\theta_i$, $\phi_i$) and ($\theta_s$, $\phi_s$) define the respective directions of light incidence and scattering [Fig. 1(a)]. $\phi_i = 0$ is the azimuthal angle in the plane of the grating perpendicular to the groove direction; $I_i$ and $I_s$ are the respective incident and scattered light intensities in the far-field (power per steradian). The subscripts $\alpha$ and $\beta$ denote the state of linear polarization of incident and scattered light (note the right to left ordering). In those cases where the electric field vector is either perpendicular or parallel to the plane of light incidence, the respective polarization states are $\alpha, \beta = \perp, \parallel$.

Any perturbations $p(x, y)$ on an optically ideal surface — such as fluctuations of height $Z(x, y)$, or variations of
dielectric permittivity \( \varepsilon(x, y, \lambda) \) — disturb the interference conditions for rectilinear light propagation from the surface and cause light scattering.\(^{22,23}\) Modelling scattering requires the solution of an integral equation that relates incident and scattered fields. In general, experimentally determined BRD functions are difficult to correlate with the statistics of surface fluctuations. Therefore, correlating surface fluctuations with light scattering usually requires approximations. The most elementary scattering approximation is the first-order Born approximation; i.e. the first term in the iterative solution of the scattering equation \(^{[\text{c.f. ref. 23 and refs. therein}]}\). In the Born approximation the bi-directional scattering function BDSF — which is the scattering part of the BDRF — is essentially proportional to the power spectral density PSD\((f)\) of the perturbation \(p(x, y)\). The surface fluctuation of \(p(x, y)\) is described by the spatial frequency vector \(f\), yielding the power spectral density function \(S_p(f) = p(x, y)\). In first order scattering theory the frequency components of the surface fluctuations are related with the respective scattering angles by the conservation law of linear momentum; i.e. by the grating equations:\(^{33,34}\)

\[
f = \left( \begin{array}{c} f_x \\ f_y \\ \lambda \\ \sin \theta_x \\ \sin \theta_y \end{array} \right) = \frac{1}{\lambda} \left( \begin{array}{c} \sin \theta_x \cos \varphi_x - \sin \theta_y \\ \sin \theta_y \sin \varphi_x \end{array} \right).
\] (4)

Equations (4) are a generalisation of grating eq. (1) for the case of first-order diffraction from a grating \((m=1)\) with spatial wavelength \(\Lambda=1/f\). The frequencies \(f_x\) and \(f_y\) are the corrugation frequencies in \(x\) and \(y\) direction of the grating, with the \(x\)-axis perpendicular to the groove direction [fig. 1(b)].

For spatially isotropic surface corrugations, the power spectral density PSD is independent of the azimuthal direction \(\varphi_x\) of the grooves and depends only on the surface spatial frequency \(f\). In the case where surface corrugations contain periodic components — such as sinusoidal groove patterns — scattered light becomes angularly distributed and exhibits a series of discrete, sharp diffraction peaks. With decreasing groove length the diffraction peaks broaden in groove direction and light scattering in azimuthal direction increases. Moreover, and as depicted in fig. 1, for grooves perpendicular to the plane of light incidence \(\varphi_x = \pi n\) with \(n=1, 2, 3,\ldots\) diffraction occurs within this plane. For grooves aligned out of the plane of incidence \(\varphi_x \neq \pi n\), at least partial scattering out of the plane occurs. This qualitatively illustrates the dependence of polar and azimuthal scattering on groove geometry factors, such as groove alignment and groove lengths distribution. It also indicates that bright LCD reflectors with moderate directivity, which scatter major amounts of ambient light into a targeted polar viewing cone, should be feasible without unduly narrowing diffuse azimuthal scattering.

In the visible wavelength range light scattering is mainly caused by surface roughness; i.e. dielectric surface fluctuations \(\Delta \varepsilon(x, y, \lambda) = 0\) can be neglected. The cause for this topological scattering are phase fluctuations in the reflected wave-front due to amplitude fluctuations \(Z(x, y)\) of the surface topology. In the smooth-surface limit, where surface roughness is smaller than the wavelength of light, the bi-directional scattering distribution (BDS) function becomes:\(^{15-26}\)

\[
\text{BDSF}_1 = F_1(\theta_x, \theta_y, \varphi_x, \varphi_y, \lambda, \varepsilon) S_p(f).
\] (5)

The product \(F_S S_p\) in eq. (5) is the first term of the expansion of the Fourier integral, where \(S_p(f)\) is the two-dimensional power spectral density (PSD) of the surface roughness \(Z(x, y)\). The PSD is a pure surface quantity that depends only on topological parameters, such as surface periodicity and corrugation shape. In the case of rough surfaces, eq. (5) holds only if the higher terms of the Fourier expansion are negligible. For isotropic or grating-like topologies, the angular distribution of light scattering can be described by the one-dimensional form of the BDS function of eq. (5). In our geometry — where the MC-grooves are aligned perpendicular to the plane of light incidence \(\varphi_x = 180^\circ\) — scattering occurs in the same plane \(\varphi_y = 0\). In this case the one-dimensional power spectral density function of the height fluctuations in eq. (5) becomes:

\[
S_p(f) = \frac{1}{2L} \left( \int_{-L}^{L} e^{2\pi f x} Z(x) dx \right)^2.
\] (6)

We will show in §5, that eq. (6) contains all topological information on MC-surface roughness within the range of surface frequencies:

\[
\frac{1}{L} < f < \frac{N}{2L};
\] (7)

where \(L\) is the MC-profile length and \(N\) the number of data points along a given MC-profile. Equation (7) defines the angular limitations of light scattering of eq. (5). Equations (5) and (6) will be shown to quantitatively describe the light scattering characteristics of our MC-topologies.

Within the smooth surface limit of eq. (5), the form factor \(F_1\) in eq. (5) is independent of surface roughness. Furthermore, in the one-dimensional case, \(F_1\) takes the simple form:\(^{22,26}\)

\[
F_1 \approx 16 \frac{\pi^2}{\lambda^2} \cos \theta_x \cos \theta_y Q_{Lov};
\] (8)

where \(Q_{Lov}\) are the polarization factors which describe the dependence of light scattering on the polarisation of incident light and the polarizing properties of the scattering surface. For scattering within the plane of light incidence \((\varphi_y = 0)\), the cross-polarisation factors vanish \((Q_{Lov} = Q_{Lov} = 0)\) and the remaining terms are:\(^{25}\)

\[
Q_{Lov} = |\varepsilon - 1|^2 \left( \frac{1}{(\cos \theta_x + \sqrt{\varepsilon - \sin^2 \theta_x})(\cos \theta_y + \sqrt{\varepsilon - \sin^2 \theta_y})} \right)^2,
\]

\[
Q_{Lov} = |\varepsilon - 1|^2 \left( \frac{\sqrt{\varepsilon - \sin^2 \theta_x} \sqrt{\varepsilon - \sin^2 \theta_y} - \varepsilon \sin \theta_x \sin \theta_y}{(\varepsilon \cos \theta_x + \sqrt{\varepsilon - \sin^2 \theta_x})(\varepsilon \cos \theta_y + \sqrt{\varepsilon - \sin^2 \theta_y})} \right)^2.
\] (9)
where \( \varepsilon \) is the relative permittivity of the interface.

The wavelength dependence of the BDS function \( F_\lambda \) is governed by the form factor \( F_\gamma \) and by the PSD function of eqs. (6) and (8) respectively. From eq. (4) and \( \varphi = 0 \) it follows that the argument of PSD(\( \lambda \)) is proportional \( \lambda^{-1} \). Changing the wavelength of incident light therefore shifts the fraction of the PSD pertinent to light scattering. The one-dimensional PSD of the MC-surface roughness \( S_\lambda(f_\lambda) \) in eq. (6) is determined by the statistical amplitude fluctuations of the MC-topology. \( S_\lambda(f_\lambda) \) describes the angular distribution of scattered light for a given wavelength. Conversely, for a fixed set of scattering angles \( (\theta, \varphi) \) the PSD function may depend on the wavelength of incident light. Therefore and according to eq. (4), changing the wavelength shifts the frequency bandwidth fraction of the power spectrum that governs the distribution of scattered light. Without scattering contribution from the PSD function, the form factor \( F_\gamma \) in eq. (8) suggests that scattering decreases with wavelength as \( \lambda^{-3} \). However, and as we will show in Fig. 8, this does not quite apply to MC-topologies. Therefore, other scattering mechanisms apart from topological scattering must contribute to MC-scattering, or the assumption that the power density spectrum of MC-topologies is wavelength-independent does not hold.

Apart from surface roughness, density fluctuations in the dielectric \( \varepsilon(x, y, \lambda) \) of a surface — such as spatial refractive index fluctuations — may also cause light scattering. In the special case of perfectly smooth surfaces (\( Z = 0 \)), the influence of density fluctuations on surface scattering can be approximated with models. Such models correlate magnitude and distribution of material composition fluctuations with light scattering.\(^{22,23,26}\) Dielectric scattering was reported for surfaces consisting of randomly oriented anisotropic metallic grains, such as Beryllium.\(^{20}\) Non-aligned liquid crystal polymer (LCP) films with randomly oriented birefringent domains could also cause dielectric scattering. Analogous to the above topological scattering case described by eq. (5), the bi-directional dielectric scattering distribution function BDSF\( \gamma \) is in the smooth-fluctuation limit proportional to the power spectral density of the dielectric fluctuations \( S_\lambda(f_\lambda) \). However, unlike its topological counterpart \( F_\gamma \), the dependence of the one-dimensional dielectric form factor \( F_\gamma(\theta, \varphi, \varphi, \lambda, \varepsilon(\lambda)) \) on wavelength scales with \( 1/\lambda^2 \) and not with \( 1/\lambda^3 \);\(^{22,26}\) where \( F_\gamma(\theta, \varphi, \varphi, \lambda, \varepsilon(\lambda)) \) is assumed not to depend on dielectric fluctuations.

In general, bi-directional reflectance distribution functions of real surfaces comprise contributions from different scattering mechanisms. For statistically independent scattering mechanisms, or if different sources contribute to scattering, the respective BDSFs are additive. In §7 we will show that this applies to two-dimensional, photo-patterned MC-surface corrugation profiles, enabling multidirectional diffusers and reflectors with virtually any angular scattering profile. In the case of different, but related scattering mechanisms, interference effects can occur which lead to intermediate wavelength dependences.\(^{22,26}\)

3. Experimental; Preparation of Monomer Corrugated (MC)-Optical Thin-Films

MC-diffusers and reflectors with different optical properties were made using two different MC-solutions S1 and S2 which comprise the respective amounts of 22wt% and 15wt% of nematic MC-material. The nematic MC-material dissolved in butyl acetate/ethanol (19/1 parts) consists basically of the uv-curable pre-polymer LCP-mixture (CB483) and the non-cross-linkable ethane liquid crystal (LC)-component 1-ethoxy-4-[2-(4-pentylcyclohexy)ethyl]-benzene 5CAPO2\(^{27}\) — both from ROLIC — and 1% of photo-initiator Irgacure 184 from CIBA. The respective nematic phases of CB483 and 5CAPO2 are 18–55°C and 15–41°C. The pure MC-material is nematic at room temperature and designed to immediately copy alignment information from underlying films upon phase-transition from the isotropic into the nematic phase during solvent evaporation. The solutions S1 and S2 were homogenized in an ultrasonic bath for 5 minutes and filtered via 0.2 μm pore filters. The extraordinary and ordinary refractive indices of CB483 at 550 nm are \( n_{e,LCP} = 1.69 \) and \( n_{o,LCP} = 1.56 \).

For MC-diffuser \( D_0 \) with its isotropic topology (Fig. 2), MC-solution S1 was spin-coated at 2000 rpm onto a non-

![Fig. 2](image-url)
aligned glass substrate. After spin-coating, the sample $D_0$ was exposed to non-polarized uv-light of 14 mW/cm² during 60 s to cross-link the respective pre-polymer MC-fraction. Finally, the non-cross-linkable LC-component 5C0A52 was removed from the cross-linked MC-polymer film by rinsing the substrate with ethanol, followed by tempering at 150°C for 5 min.

Prerequisite for the formation of groove topologies in MC-diffusers and reflectors is uniaxial alignment of the substrates prior to spin-coating the MC-material. Ten glass substrates were photo-aligned for the diffusers $D_1$-$D_{10}$ by our linear photo-polymerization (LPP) technology, using a 2% cyclopentanone LPP-solution of JP265 from ROLIC. After tempering for 10 min at 180°C, the resulting 60 nm thin LPP-films were photo-aligned at vertical light incidence at room temperature with linearly polarized uv-light at $\lambda = 315$ nm, leading to 0° bias-tilt. MC-solution S2 was spin-coated at 2000 rpm on each substrate and cross-linked with non-polarized uv-light of 7 mW/cm² during different exposure times $t_i$. The different groove topologies of $D_1$-$D_{10}$ result from the different exposure times $t_i$:

- $t_1=30$ s, $t_2=60$ s, $t_3=90$ s, $t_4=120$ s, $t_5=150$ s, $t_6=180$ s, $t_7=240$ s, $t_8=300$ s, $t_9=360$ s and $t_{10}=600$ s.

The different topologies of the anisotropic MC-reflectors $R_1$-$R_4$ were achieved by varying the MC-film thickness (at constant cross-linking energy). MC-solution S2 was spin-coated on photo-aligned glass substrates using different spin-coating speeds. Subsequent vacuum deposition of a 100 nm silver layer on top of the MC-films converted the diffusers into reflectors. The rpm and the total film thickness for $R_1$-$R_4$ are:

- $3000$ rpm, $2000$ rpm, $1700$ rpm, $1200$ rpm and $242$ nm, $300$ nm, $320$ nm, $350$ nm, respectively.

Both, the cross-linking intensity 7 mW/cm² and the uv-exposure time of 60 s are constant for $R_1$-$R_4$. The resulting average groove periods and groove heights of $R_1$-$R_4$ are: $0.97$ μm, $1.53$ μm, $1.9$ μm, $2.32$ μm and $57$ nm, $104$ nm, $123$ nm, $147$ nm, respectively.

The topologies of the MC-corrugations were investigated by atomic force microscopy (AFM) in the contact-mode. AFM scans of 30x30 μm perpendicular to the grooves at two different pixel resolutions 256x256 or 512x512 were made. Their one-dimensional power spectral density (PSD) functions were directly derived from AFM surface-profile data. According to eq. (6), the one-dimensional average Fourier transformation of the AFM data were squared. Fast Fourier transformation (FFT) was used to calculate the average Fourier transformation. This was done by averaging the Fourier amplitudes of $N$ individual AFM profiles — with $N = 256$ or 512 AFM-scans — perpendicular to the groove direction (y-axis) using the scanning probe image processor software from Image Metrology. Since we are interested in the average light scattering characteristics of MC-topologies and because the height-fluctuations along MC-corrugation profiles are random (Fig. 3), we will show in §5 that the averaged one-dimensional Fourier transform approach is appropriate and yields smooth power density functions with noise immunity.

The absolute angular reflectance of MC-diffusers and reflectors was determined with a Perkin-Elmer Lambda 900 spectrometer equipped with an integrating sphere detector of large aperture. The incident monochromatic light of the instrument generates a relatively large (8x10 mm) spot that can be polarized. The two-dimensional spatial distribution of the scattered light was determined by conoscopic means with an EZ-Contrast 160R instrument from ELDM, using bright, collimated incident daylight D65. A BaSO4 white standard DIN 5033 from E. Merck served as a reference to determine gain and brightness of MC-reflectors. The certified diffuse reflectance of BaSO4 is $R_d = 0.975$ (400 nm), 0.983 (500 nm), 0.987 (600 nm) and 0.987 (700 nm).

4. Monomer Corrugation (MC)-Topologies and MC-Mechanisms

Monomer corrugated (MC)-materials consist of blends of liquid-crystal pre-polymers and photo-chemically inert monomer liquid crystals (LCs) which undergo optically induced phase-separation upon photo-cross-linking the liquid crystalline pre-polymer (LCP) components. Subsequent removal of the monomer LCs leaves a solid liquid crystal polymer film on the substrate with nano- or micro-pores in the MC-film. Shape, amplitude and periodicity of MC-topologies and their spatial distributions are controlled by MC-material parameters, MC-film thickness, LCP-content in the MC-mixture, substrate alignment, anchoring strength of the alignment, and by the direction and dose of cross-linking uv-light. The individual MC-components affect the final MC topology primarily via their miscibility and photo-reactivity. Because molecules tend to phase-separate prior to cross-linking, decreasing miscibility generally induces larger size topologies; whereas strong reactivity or fast speed of polymerization tends to decreases groove dimensions. Vice versa, the period of MC-topologies can be decreased by increasing LCP and/or photo-initiator content, or by increasing the dose of uv-cross-linking. Moreover, we found that the initial MC-film thickness prior to cross-linking strongly affects the MC-topology. In general, groove period and height increase with MC-film-thickness (c.f. MC-reflectors $R_1$-$R_4$ in §3).

The influence of substrate alignment and cross-linking energy on shape and amplitude of MC-corrugations are illustrated by the AFM images in Fig. 2 and Fig. 3. Figure 2(a) shows the AFM image of MC-diffuser $D_0$ made on a non-aligned glass substrate as described in the previous paragraph. Because of the random substrate surface of $D_0$, surface phase-separation is random too, which leads to the isotropic distribution of micro-corrugations depicted in Fig. 2(a). To quantitatively confirm the isotropic character of $D_0$, its two-dimensional fast Fourier transformed (2D-FFT) power spectrum was determined (c.f. §2) and depicted in the insert of Fig. 2(a). The perfectly circular 2D-FFT spectrum confirms the geometrical isotropic MC-topology of $D_0$ suggested by the AFM images and shows that MC-phase-separation indeed proceeds isotropically on non-aligned substrate surfaces.

2D-FFT power spectra enable determination of a characteristic length scale $\Lambda$, of the isotropic MC-topology of $D_0$. According to §2 the characteristic period of the morphology of $D_0$ is $\Lambda = 1/f = 2$ μm, where $f$ is the spatial frequency corresponding to the maximum of the averaged one-dimensional power spectrum of the MC-topology in Fig. 2(a). It is interesting to note the resemblance of the MC-morphology
in Fig. 2(a) with the morphologies of spinodally decomposed polymer dispersed liquid crystals (PDLCs) and ultra-thin LC-films.23-52 This similarity and the existence of a characteristic length in the MC-morphology of D0 indicates that spinodal decomposition may occur during MC-phase separation. Unlike in the case of ordinary diffusion, spinodal decomposition causes molecules to diffuse from depleted into dense regions.

Prerequisite for the generation of anisotropic MC-topologies on aligned substrates is that the degree of anisotropic interaction between surface alignment and long-range order of the MC-molecules is strong enough. Figure 3 shows the AFM images and the corresponding AFM profiles of the monomer-corrugated LCP-layers of the directional MC-diffusers D1, D5 and D10 with their different scattering profiles. As outlined in the previous paragraph, all MC-topologies were generated on uniaxially photo-aligned glass substrates. Analogous to the isotropic diffuser D0 [Fig. 2(a)], the anisotropic character and the degree of anisotropy of the MC-topologies of D1, D5 and D10 were quantitatively verified by 2D-FFT power spectroscopy [c.f. inserts in Fig. 3(a)]. Unlike the geometrically and optically isotropic diffuser D0, the power spectra of the diffusers D1, D5 and D10 in Fig. 3(a) exhibit two distinct elongated-ellipses perpendicular to the groove direction; i.e. perpendicular to the substrate aligning direction. This result confirms the pronounced AFM anisotropy of the topologies of D1, D5 and D10. The markedly different shapes of the ellipses in Fig. 3(a) due to the different processing steps described in §3 confirm that the degree of anisotropy of MC-topologies is indeed strongly affected by MC-processing.

The MC-process is basically governed by two mechanisms, namely by photoreaction and molecular-diffusion. In our present MC-materials, polymer network formation occurs via free radical polymerisation33) induced by photon absorption in the photo-initiator. Upon uv-exposure, the dispersed photo-initiator molecules form radicals, thereby causing polymerisation reactions at random positions in the MC-film. Because cross-linked MC-moieties do not blend with non-cross-linkable LC-components, local phase separation occurs. This causes diffusion of LC-molecules into film regions with lower polymer content.

The occurrence of anisotropic MC-phase-separation on uniaxially aligned substrates was further investigated by
means of contact-mode AFM-scans of the anisotropic diffuser topologies D₁–D₁₀ depicted in Fig. 3. As outlined in the previous paragraph, D₁–D₁₀ were cross-linked with successively increasing uv-doses. From Fig. 3 it follows that the MC-grooves are parallel to the photo-alignment direction. This indicates that phase separation proceeds preferentially perpendicular to the alignment direction. Qualitatively, this corresponds with the uniaxial nature of the anisotropic Van der Waals anchoring forces exerted by the alignment layer on the mesogenic MC-molecules and with the tendency of LC-molecules of the type SCAPO2 to diffuse via rotation around their long molecular axis; i.e. perpendicular to substrate alignment.

We found that at the very beginning of uv-exposure, large lateral LCP domains and correspondingly large groove structures occur in MC-films. At this early stage, small pores appear within large lateral LCP domains [c.f. D₁ in Fig. 3(a)]. With increasing uv-exposure, large LCP-domains split into smaller domains, the pores act as nucleation centres. As illustrated by D₂ in Fig. 3(a) this leads to a more uniform groove structure with larger aspect ratio. With further increasing uv-exposure the lateral LCP-domains continue to grow and simultaneously start to coalesce with neighbouring domains. This causes to increase LCP surface coverage further [c.f. the AFM image of D₁₀ in Fig. 3(a)].

The lateral LCP-domain growth process is qualitatively illustrated in Fig. 4. Figure 4 shows the increase of the surface coverage parameter ϕLCP of the LCP domains versus uv-exposure for D₁–D₁₀. ϕLCP was determined from the respective AFM scans (Fig. 3). From Fig. 4 it occurs that lateral LCP-domain growth is exponential. However, more detailed investigations of the cross-linking process are required to confirm the scaling factor.

Vertical MC-domain growth was found to be qualitatively governed by the following parameters: MC-film thickness d (from the alignment surface to the MC peak), average groove height ha and uv-exposure time t. Figure 5 depicts the relationships between these three parameters for diffusers D₁–D₁₀. From Fig. 5 it follows that the LCP-film thickness d increases exponentially with uv-exposure. This corresponds with the cross-linking kinetics of photo-resist films. Also from Fig. 5 follows that the average height of the LCP-grooves hₐ grows for exposure times up to 240 s with a similar exponential law as the growth rate of film thickness d. For exposure times up to ~120 s, the growth of hₐ parallels the growth of LCP-film thickness, whereas for longer exposure times MC-film thickness continues to grow and the average groove height starts to decrease (Fig. 5). The decrease of hₐ for extended uv-exposure is an averaging artefact. It is due to the strong irregular modulations in the LCP-groove profile which occur with increasing exposure [c.f. D₁₀ in Fig. 3(c) versus the AFM profiles of D₁ and D₂]. The pronounced modulation of groove amplitudes during vertical LCP-domain growth is probably due to the increased immiscibility of LC and LCP components with extended uv-exposure. To minimise interfacial energy, the LC-molecules diffuse towards the film-air interface. This corresponds with our finding that completed MC-films comprise virtually no residual LC-molecules but consist entirely of LCPs.

Apart from controlling MC-film thickness, average groove height and groove period, MC-processing also controls the size-distribution of MC-corrugations. The diffusers D₁ and D₅ in Figure 6 illustrate this. Figure 6 shows the different
distributions of MC-period versus uv-dosage. The distribution was determined from the respective averaged one-dimensional power spectral density (PSD)-functions of D1 and D2 using eq. (6). The PSD-function correlates distance and height of MC-corrugations. Figure 6 also shows that the frequency distribution of MC-diffuser D1 is broader than that of D2. We will show in the next paragraph that light scattering from MC-films is mainly governed by their topologies. Therefore, and as shown next, power spectral density functions are a powerful tool for correlating MC-topologies with light scattering from MC-films.

5. Correlations Between MC-Topologies and MC-Light Scattering

Due to the high degree of spatial order of MC-corrugations, MC-coatings act as optical gratings as well as diffuse scattering elements; i.e., they combine diffraction and scattering. In the following we quantitatively show that diffraction of light by anisotropic MC-topologies into targeted off-specular viewing cones is feasible. Figure 7(a) shows conoscopic plots of the spatial light distribution of MC-diffusers D0, D1 and D2 under normal illumination ($\theta_{i}=0^\circ$) for collimated, white D65 daylight. The diffraction pattern of the non-aligned MC-diffuser D0 in Fig. 7(a) with its isotropic topology is circular and agrees well with the 2D-FFT power spectrum of its AFM-image in the insert of Fig. 2(a). Unlike scattering from D0, the angular distribution of light scattered by the anisotropic MC-diffusers D1 and D2 is elliptical, with the long axes of the ellipses perpendicular to the aligning direction. Comparing the scattering patterns of D1 and D2 in Fig. 7(a) with the 2D-FFT power spectra of their respective topologies in the inserts of Fig. 3 shows good qualitative agreement. Figure 7(b) depicts the cross sectional scattering profile of D1 and D2 perpendicular to the groove direction ($\phi_{g}=0^\circ$) versus polar viewing angle $\theta_{v}$. The directly transmitted beam of zero diffraction order ($m=0$) is not shown in Fig. 7(b). The profiles in Fig. 7(b) exhibit two strongly diffuse scattering peaks which are symmetrical with respect to the directly transmitted beam at $\theta_{v}=0^\circ$. The two peaks correspond with the respective positive ($m=1$, right) and negative ($m=-1$, left) first diffraction orders of the MC-groove topologies.

As was shown in §3, the total amount of scattered light $T_{d}$ from a directional MC-diffuser, versus the fraction of non-diffracted light $T_{0}$ (i.e., zero order) is controlled by process parameters. This is illustrated by the transmission measurements of D1-D10 depicted in Fig. 8. Figure 8 shows that the initially dominant non-diffracted portion $T_{0}$ decreases with increasing uv-exposure during processing, whereas diffuse scattering strongly increases and becomes the dominant contribution for extended exposure.

In our attempt to correlate MC-film topology with light
scattering we found that a simple sinusoidal grating approach, such as the Raman–Nath theory, is not adequate. However, the bi-directional diffuse scattering (BDS) formalism outlined in §2 and the averaged one-dimensional power spectral density (PSD) functions of MC-topologies were found to properly model light scattering of MC-corrugations. Quantitatively this is shown in Figs. 9 and 10, where experimental results are compared with the theory of §2. Figure 9(a) shows the fit between the experimental light scattering profiles of MC-diffusers D₁ and D₅ of Fig. 7(b) for collimated, non-polarized incident white light and the respective one-dimensional power spectral density functions S₁(D₁) and S₅(D₅) following from AFM-scans and eq. (6). The averaged one-dimensional PSD functions used for the fit in Fig. 9(a) are those of Fig. 6. The good agreement between experiment and theory in Fig. 9(a) confirms that averaged 1D-PSD functions indeed properly describe light scattering of MC-topologies. The scaling factor which follows from the 1D-PSD functions and light scattering along θₛ=0 [Fig. 7(b)] is \( F = 0.43 \times 10^{-3} \) nm⁻³. This factor holds for MC-diffusers D₁–D₅. However, for D₆ and D₁₀ with their longer uv-cross-linking times, the factor increases to \( F = 0.5 \times 10^{-3}, 0.8 \times 10^{-3} \) and \( 1.0 \times 10^{-3} \) nm⁻³ respectively. This could be caused by an increase of the non-topological contribution to MC-scattering with increasing uv-exposure. Another cause could be an increase of the higher order terms which eq. (5) does not take into account. Also the occurrence of spikes, which we found to occur in MC-profiles with increasing uv-exposure, could detrimentally affect the PSD averaging process [c.f. the height-profile of D₁₀ in Fig. 3(c)].

To further compare experiment with theory, absolute measurements of the angular light scattering of our MC-diffusers at a defined wavelength and state of polarization of incident light were made with a Perkin Elmer Lambda900 photo-spectrometer. Figure 9(b) shows the angular dependence of measured and calculated forward scattering intensity of MC-diffuser D₁₂ for S and P polarized monochromatic input light of \( \lambda = 550 \text{ nm} \). The averaged one-dimensional PSD function from Fig. 6 was used for the fit. The good agreement between theory and experiment that follows from Fig. 9(b) for the angular-shape of the scattering by D₁₂ shows that averaged 1D-PSD functions not only hold for white light, but also for monochromatic light. The respective scaling factors between the 1D-PSD function determined from AFM and light scattering data are \( F_P = 0.85 \times 10^{-5} \) nm⁻³ for P-polarized incident light and \( F_S = 1.04 \times 10^{-5} \) nm⁻³ for S-polarized light. The difference between the factors is due to the respective different dielectric constants in \( \varepsilon_{Q_P} \) and \( \varepsilon_{Q_S} \) of eq. (9). Since the optical axis of the LCP molecules is parallel to the MC-groove direction for P-polarized light, \( \varepsilon_{Q_P} = (n_{11,\text{LCP}})^2 \) holds for \( Q_P \) and \( \varepsilon_{Q_S} = (n_{11,\text{LCP}})^2 \) for \( Q_S \) (S-polarized light). Considering the complexity of MC-topologies, the good
agreement between experiment and theory of the S and P polarized scattering profiles of $D_5$ shown in Fig. 9(b) is surprising and convincing.

From the grating eq. (4) it follows that it is primarily the argument of the PSD-function (6), i.e., the spatial frequency, that depends on the wavelength of incident light. In the case of topological scattering the PSD-function is a pure geometrical quantity and independent of incident radiation. However, if dielectric material fluctuations exist, the magnitude of the PSD-function can become wavelength dependent. According to eq. (4), the spatial frequency that corresponds with a given scattering direction is inversely proportional to the radiation wavelength. This geometrical dependence can be eliminated by plotting scattering data at different wavelengths not versus scattering angle but versus spatial frequency. To eliminate the geometrical scattering contribution of MC-topologies, the monochromatic scattering of diffuser $D_5$ was determined for $\phi_i=0$, using the respective incident wavelengths 400 nm, 550 nm and 700 nm. The data were recorded versus viewing angle $\theta_v$ and converted into the respective frequency dependencies by grating eq. (4). The results for $D_5$ are shown in Fig. 10(a). Similar results were obtained for $D_1$--$D_4$. Based on our geometrical model the scattering intensities versus frequency in Fig. 10(a) show good agreement between experiment and calculations. However, the frequency shift of the three peaks in Fig. 10(a) versus wavelength of incident light is proportional to $\lambda^2$, with $n=2.2$ and not $n=3$. As outlined in §2, $1/\lambda^3$ holds for the wavelength dependency of the geometrical scaling factor $F_2$ in the case of pure topological scattering; whereas $1/\lambda^2$ holds for $F_1$ and pure dielectric scattering. The intermediate exponent $n=2.2$ suggests a non-topological mechanism to contribute to MC-scattering. The cause could be order fluctuations of the optical anisotropy of the LCP molecules within the grooves, or nano-grooves superimposed onto the micro-topology of $D_5$. From the observed shift of the frequency distribution of $D_5$ in Fig. 6, nano-grooves seem to be the more likely cause for the discrepancy.

To further investigate the influence of PSD on light scattering from MC-topologies, experimental scattering data of MC-diffusers $D_1$ and $D_5$ were normalised with the proportionality factor $F(\lambda)=C/\lambda^n$, $C=$constant, and compared with the respective power spectral densities from Fig. 6. Figure 10(b) shows the results. From Fig. 10(b) it follows that the position and shape of the three scattering plots of MC-diffuser $D_1$ agree very well over the entire frequency range with the shape of the averaged 1D-PSD function in Fig. 6. This indicates that MC-diffuser $D_1$ essentially acts as a topological scattering device in the visible spectral range. This finding agrees with the calculated wavelength dependence $1/\lambda^{2.7}$ of $F$ for $D_1$ which approaches the topological exponent $n=3$. Except for higher frequencies where deviations occur at the wavelengths 550 nm and 700 nm, analogous results were obtained for MC-diffuser $D_5$. [Fig. 10(b)]. Below, the deviations are shown likely to be due to nano-grooves.

Changing the wavelength of incident light causes, according to eq. (4), the frequency bandwidth fraction for the power spectrum which governs the distribution of scattered light to shift. Indeed, only groove frequencies that

meet the condition $|\lambda \times f_s|<1$, i.e., $f_s<1.43 \mu m^{-1}$ at 700 nm and $f_s<2.5 \mu m^{-1}$ at 400 nm contribute to scattering at $\theta_v=0$, $\phi_i=0$ according to eq. (5). From Fig. 6 it follows that for MC-diffuser $D_1$ virtually all grooves contribute to scattering in the visible spectral range. However, for MC-diffuser $D_5$ this holds only at 400 nm where scattering fits the PSD function. At longer wavelengths, for instance at 700 nm, grooves with $f_s \geq 1.43 \mu m^{-1}$ no longer contribute to scattering and cause tailing of the 1D-PSD plot above $f_s=1.43 \mu m^{-1}$ for $D_5$ in Fig. 6. Long frequency grooves are more likely to cause dielectric scattering and to decrease the exponent $n$ in $1/\lambda^n$. Although our topological approach properly describes the proportionality of angular MC-scattering with PSD, the wavelength dependence of MC-scattering appears to be more complex. Indeed, within the above approximation of the $1/\lambda^n$ dependence in the visible spectral range, we found that $n$ decreases with increasing $uv$.

![Diagram](attachment:image.png)

**Fig. 10.** (a) Light scattering profiles of diffuser $D_5$ along $\phi_i=0$ for monochromatic incident light of the respective wavelengths 400, 550 and 700 nm, versus groove frequencies $f_s$. The profiles are measured versus viewing angle $\theta_v$ and converted into the grating frequency domain using eq. (4). Light incident normal to the substrate (i.e., $\theta=0$, $\phi_i=0$). (b) Normalised angular dependence of the scattering profiles of MC-diffusers $D_1$ and $D_5$ along $\phi_i=0, \theta(\lambda)/F(\lambda)$, illuminated with monochromatic light of 400 nm (squares), 550 nm (circles) and 700 nm (triangles). The scattering profiles compare with the corresponding 1D-PSD functions $S_i(\lambda) \times 10^{-6} m^{-2}$ and $S_i(D_5) \times 10^{-4} m^{-2}$ from Fig. 6 (solid lines).
exposure from \( n = 2.7 \) for D₁ to \( n = 1 \) for D₁₈, requiring further investigation.

For polar viewing angles between 0°–80°, the above analysis shows, that the shapes of the light scattering graphs of D₁ and D₃ in Figs. 9 and 10 are essentially governed by the spectral density functions of the respective MC-topologies. To illustrate MC-design flexibility, the topology of MC-diffuser D₁ was designed such that incident light is redirected into a 38° cone to the right, i.e. from \( \theta_{\text{min}} = 0^\circ \) to \( \theta_{\text{max}} = 38^\circ \) with maximum off-normal scattering at \( \theta = 10^\circ \).

The polar viewing cone of diffuser D₃ covers the range from \( \theta_{\text{min}} = 0^\circ \) to \( \theta_{\text{max}} = 80^\circ \) with maximum scattering at \( \theta = 15^\circ \) [c.f. Fig. 7(b)]. The respective angles of maximum scattering \( \theta₁ \) of both diffusers D₁ and D₃ agree well with their first order diffraction angles following from grating eq. (4) as well as with the respective average spatial groove frequencies \( f_s \) following from the averaged one-dimensional power spectral density functions in Fig. 6.

6. High-Gain, Paper-White Directional MC-Reflectors

Diffuse and directional MC-reflectors result when coating the anisotropic groove topologies of MC-diffusers with a thin layer of aluminum. This enables efficient diffuse MC-reflectors for displays as well as for optical security elements. MC-reflectors combine directional light scattering and diffraction without glare. By tailoring the geometry of the MC-topology, the amount of specular versus scattered light can be controlled and combined such that strong brightness enhancement results. Moreover, and as was shown in the previous paragraph, MC-topologies strongly reduce the wavelength dependence of reflectivity. To illustrate this effect, the MC-reflectors R₁–R₄ were used using different processing procedures that result in different size distributions of grooves (c.f. §3).

The scattering distribution of R₁–R₄ was determined for collimated daylight D65 at −30° illumination incidence. The reflected light consists of the specular peak at +30° and two strong, diffuse off-specular peaks that are symmetrical with respect to +30°. The averaged one-dimensional power spectral density functions of R₁–R₄ were determined from AFM scans of the reflector topologies. Their 1D-PSD functions are depicted in Fig. 11(a). Figure 11(b) shows the brightness enhancement of R₁–R₄ relative to a standard-white BaSO₄ diffuser perpendicular to the groove direction (\( \phi = 0^\circ \)). The topology of reflector R₁ was chosen such that incident light is directed into a cone of ±15° with maximum reflectance at \( \theta = 0^\circ \), i.e., normal to the reflector surface and 30° off specular reflection. From Fig. 11(b) follows for R₁ a brightness enhancement of a factor 2–4. To illustrate the design flexibility of MC-reflectors and to further increase gain, the topology of R₂ was chosen such that maximum reflectance occurs at \( \theta = 11^\circ \) off-axis for vertical light incidence. Fig. 11(b) shows that this causes the gain of R₂ to increases to 11. Increasing the respective angle of maximum reflectance \( \theta₁ \) further leads for R₃ and R₄ to a gain of 18 [Fig. 11(b)]. The bright MC-reflectors R₁–R₄, with \( \theta₁ = 18^\circ \), combines an exceptionally large gain of 25 with a reasonably broad viewing cone [Fig. 11(b)]. The angles of maximum reflectance \( \theta₁ \) in Fig. 11(b) correspond with the first order diffraction angles \( \theta₁ \) in grating eq. (4). \( \theta₁ \) was determined by inserting the average groove frequency \( f_s \) of the respective MC-reflectors into grating eq. (4); where the average groove frequencies \( f_s \) follow from the averaged one-dimensional PSD functions in Fig. 11(a).

Apart from the above correlations found between reflective MC-topologies and directional brightness enhancement, correlations exist between the viewing angle dependence of reflectivity and MC-topology. This is suggested from comparing the reflectivity peaks of the MC-reflectors R₁–R₄ in Fig. 11. The comparison of the reflectivity peaks of the graphs in Fig. 11(a) and Fig. 11(b) suggests a scaling factor \( F \) to exist between the ratios of corresponding peaks. Indeed, from Fig. 11(a) and Fig. 11(b) follows for R₁–R₄ and their respective peak ratios a scaling factor \( F \approx (1\text{-PSD}) \) with brightness \( F \approx 1.0 \times 10^{-6} \text{ nm}^{-3} \). Slightly decreases for R₄ to \( F \approx 0.8 \times 10^{-9} \text{ nm}^{-3} \). To quantitatively correlate diffuse light scattering of MC-reflectors with their PSD functions, absolute measurements at the wavelengths 400 nm, 550 nm and 700 nm were made. This was done by recording the scattered intensities of monochromatic light incident at \( \theta = 30^\circ \) versus viewing angle \( \phi \) along \( \phi = 0^\circ \) and convert-
The observed discrepancy between experiment and theory could also be the cause for the “wrong” exponent that follows from the wavelength shift of the reflectance peaks of $R_1$ and $R_4$ in Figs. 12(a) and 12(b). Using the same procedure as for the diffusers in the previous paragraph, we found for the wavelength dependency of the reflectance of $R_1$ and $R_4$ the respective exponents $n \approx 2.34$, $n \approx 1.66$, and $n \approx 0.72$, and $n \approx 0.15$, requiring further investigation.

As shown in §5 for MC-diffusers, the wavelength dependency of MC-topologies can virtually be eliminated by properly designed topologies; i.e. paper-white MC-topologies are feasible, provided small exponents $n$ can be realized. The small exponent $n \approx 0.15$ of the high gain MC-reflector $R_4$ ideally meets this requirement, rendering light scattering from $R_4$ virtually wavelength independent over the entire visible spectral range. The photograph in Fig. 13(a) depicts the paper-white appearance of the high gain MC-reflector $R_4$ whose reflectance is virtually polarization and wavelength independent.

7. Photo-Patterned MC-Topologies for Multi-Directional Light Scattering

To illustrate the effect of photo-patterned alignment of diffuse MC-reflectors, Fig. 13(b) shows a pixelated MC-reflector on a plastic substrate. The image of the reflector is a result of its binary pixel topology with two different pixel scattering directions. By synergistically combining the directionality of photo-patterned substrate alignment and MC-scattering, pixelated MC-reflectors are achieved whose pixels scatter light into different azimuthal directions. Alignment pixelation renders multidirectional MC-reflectors feasible redirecting light that is incident from different directions towards the viewer, thereby further increasing brightness and/or uniformity of MC-reflectors. This is illustrated in Fig. 14(a) that shows the optical microscope photograph of a pixelated MC-diffuser. The two different LPP-photo-alignment directions of adjacent areas and the two corresponding groove directions $a_1$ and $a_2$ differ by the azimuthal angle $90^\circ$ and shown in the AFM image of Fig. 14(b). Due to pixelation, the MC-reflector topology of Fig. 14(a) redirects incident light into two different orthogonal directions. This is confirmed by the conoscopic image of the pixelated MC-reflector in Fig. 14(c). In terms of BSDR functions, the optics of the MC-reflector in Fig. 14 can be described by two BSDR functions which are additive and which follow from the respective MC-pixel geometries. Therefore, the scattering intensities from the grooves of the pixels ($a_1$) in direction $\phi_1 = 45^\circ$ superimpose the intensities from the grooves of pixels ($a_2$) along $\phi_2 = -45^\circ$. This demonstrates that pixelation renders future MC-diffusers and reflectors with still further improved light scattering uniformity, directionality and gain possible.

8. Conclusions

We have shown that amplitude- and shape distribution of the nano and micro topologies of monomer corrugated (MC) polymer thin-films can independently be tuned by substrate alignment and uv-cross-linking. Light scattering synergizes result, enabling bright, directional MC-reflectors that defy most of the deficiencies of single-frequency gratings, such as narrow field of view, coloration, or color shift versus angle.
of view. Based on a bi-directional diffuse scattering (BDS) model and averaged one-dimensional power spectral density (PSD) functions, quantitative correlations between MC-groove amplitude, groove periodicity and diffuse light scattering are derived. Despite the complex MC-topology and its somewhat chaotic character, we found that the angular scattering intensity in the plane of light incidence is proportional to the averaged one-dimensional power spectral density of MC-height fluctuations determined by atomic force microscopy. The complex and surprisingly small wavelength dependence of MC-diffusers and reflectors was found to depend sample preparation and MC-material properties. It enables achromatic, sub-micron thin diffusers and directionally diffuse reflectors with large optical gain,
high display contrast and minimal glare. Moreover, photo-patterned MC-topologies on photo-aligned substrates are shown to further enhance directional reflectivity of MC-diffusers and reflectors, converting detrimental glare into desired brightness, etc.