Thin-film Characterization

5

The third intended application for the proposed dispersion-encoded low-coherence interferometry is the evaluation of thin-film characteristics on substrate materials. Due to the usage of thin-film technologies in high-volume production in e.g. the photovoltaics and semiconductor industry, process monitoring becomes relevant in order to ensure functional parameters such as solar cell efficiency, [289]. In this context, film thickness as well as film homogeneity over large areas are important criteria for quality assurance. This section describes the modifications and developments of the DE-LCI approach in order to measure film thickness of nm-sized films on comparatively thick substrates. Furthermore, results of different sample measurements are presented.

5.1 Setup Considerations

In order to achieve the goal of scan-free, spatially resolved film-thickness measurements of transmissive samples, a Mach-Zehnder interferometer was combined with an imaging spectrometer analogous to the approaches used in surface profilometry (see chapter 3) and polymer characterization (see chapter 4), Fig. 5.1. The collimated beam of a white-light source ($\Delta\lambda = (400–1000)\,\mathrm{nm}$) was divided by a broadband plate beamsplitter into sample and reference arm. The reference arm contained a dispersive element (N-BK7, $t_{DE} = 6.23\,\mathrm{mm}$) while the sample arm was equipped with the particular sample which was a thin-film on a transparent substrate. If the sample contained a film thickness gradient, it was mounted in such a way that the

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thickness gradient was aligned with the x-axis. The thickness and refractive index properties of the sample were dependent on the composition of the substrate material (t_s, n_s) and on the number of film layers (t_{fn}, n_{fn}) so that $t_{smp} = f(t_{sub}, t_{fn})$ and $n^{smp} = f(n_{sub}, n_{fn})$. The transmitted light was superimposed with light from the reference arm after passing the secondary beamsplitter BS2 and imaged onto the slit of an imaging spectrometer. The used imaging magnification was typically M = 0.6. The spectrally decomposed signal was detected with a two-dimensional CMOS-array of a camera.

The signal formation and analysis were largely based on the theory of dispersion-controlled low-coherence interferometry established for precision profilometry and cross-linking characterization in polymers, chapter 3. However, due to the composition of the sample as a multilayered system, a mathematically correct attribution has to be performed for all signal components to the sample composition. Typically, a transfer-matrix formalism is used to describe the transmission and reflection of light at every material interface, [290], Fig. 5.2. The basic idea of this formalism is that one singular matrix is used to describe the propagation of the electric field through a whole system of multiple layers. As of the design of the experiment within this work, only light with normal incidence regarding the front surface of the material system was taken into account. The formalism calculates the electric

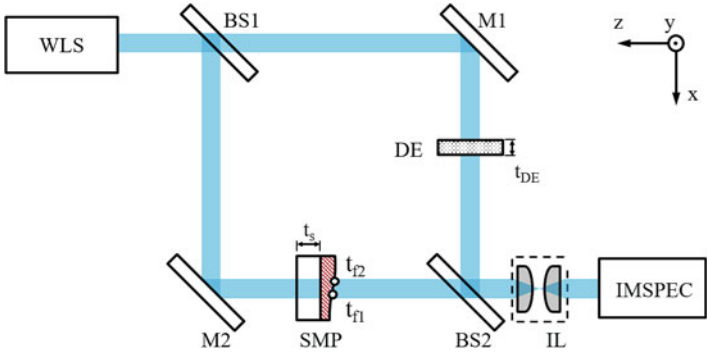

Figure 5.1 Experimental setup based on a Mach-Zehnder configuration with a WLS—white light source which is splitted 50:50 by BS1—first beamsplitter into a reference beam which is directed by M1—reference arm mirror and manipulated by a DE—dispersive element (N-BK7, t_{DE} = 6.23 mm) and the sample beam which is directed by M2—sample arm mirror through the SMP—sample with thin-film (thickness slope $t_f(x)$) on a substrate. Both are recombined by BS2—second beamsplitter and imaged using the IL—imaging lens on a IMSPEC—imaging spectrometer where the analysis is performed

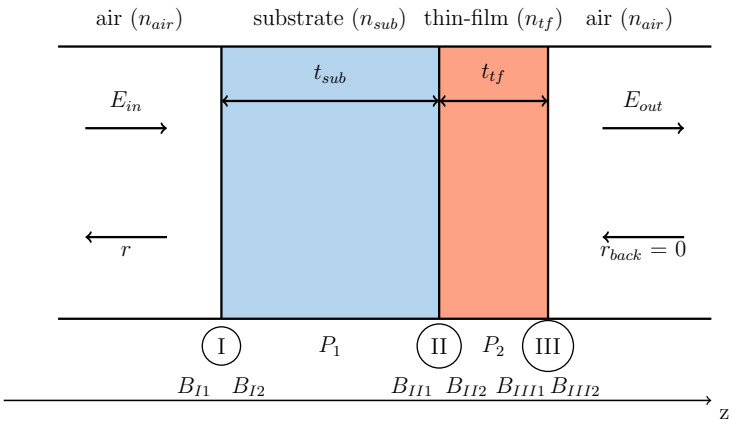

Figure 5.2 Simplified schema of the transfer-matrix formalism used to calculate the film thickness of a layer t_{tf} on a substrate using the information of the present boundaries B_{ij} and materials the electric field propagates through P_j, [290]

field components for a forward traveling wave (positive x-direction) E_{in} and for a backwards traveling wave E_{out} in order to account for transmission and reflection at every material interface due to the material properties by using the wavenumber k'_a and the propagation vector x

$$E_{in} = \Xi e^{-ikx} + \Pi e^{+ikx}, \qquad E_{out} = \Upsilon e^{-ikx} + \Omega e^{+ikx}, \tag{5.1}$$

where the propagation coefficients $\Xi, \Pi, \Upsilon, \Omega$ hold information on the materials and interfaces. Considering the fact that the tangential components of the electric field must be continuous at a material interface as a boundary condition and that the matrices of every single layer have to be multiplied, a notation of the electric field component of every material and subsequent material boundary can be found as a matrix describing the electric field when entering, M_i, and exiting the material system, M_o,

$$\underbrace{\begin{pmatrix} e^{-ikx} & e^{+ikx} \\ -ike^{-ikx} & +ike^{+ikx} \end{pmatrix}}_{M_i} \begin{pmatrix} \Xi \\ \Pi \end{pmatrix} = \underbrace{\begin{pmatrix} e^{-ikx} & e^{+ikx} \\ -ike^{-ikx} & +ike^{+ikx} \end{pmatrix}}_{M_o} \begin{pmatrix} \Upsilon \\ \Omega \end{pmatrix} \tag{5.2}$$

which leads to the final transfer matrix notation M_T,

$$M_T = M_{0i}^{-1} \cdot M_{0o} \cdot M_{0o}^{-1} \cdot M_{1i} \cdot ....M_{(j-1)o}^{-1} \cdot M_{ji}. \tag{5.3}$$

The description of the electric field in this notation represents the resulting wave in the sample arm. In combination with a notation for the reference arm, the complete propagation of light through the Mach-Zehnder interferometer can be described. In combination with the estimated equalization wavelength, measured data sets could be fitted according to the methods mentioned before, see section 3.3. A detailed derivation of a simple, one-layer sample material which was used for analysis of the majority of measurements presented in this chapter, can be found in the appendix in the Electronic Supplementary Material (ESM).

One important requirement in the practical implementation of this approach is the consideration of the substrate material as it shows incoherent behavior. Otherwise, the calculated signal inhibits higher order interferences which could reduce the signal-to-noise ratio significantly. Methods to minimize the influence of this problem would either be averaging the propagation in the substrate material by applying random phases or utilize a net-radiation method to get rid of the disturbing interference overlays, [291, 292].

5.2 Characterization of Thin-films on Bulk Substrates

In order to evaluate the characteristics of the setup regarding its capabilities in thin-film characterization, samples of single-layer ITO coatings on polished float glass substrates (CEC020S, PGO GmbH, Germany) were prepared. The samples were half-sided chemical etched in a ferric chloride bath at 230 K for 3 hrs.

The determination of height profiles on substrates with coated thin-films can be performed by classical methods like spectral photometry or tactile profilometry either in a point wise or scanning fashion, Fig. 5.3 a). The acquisition of surface profiles using a tactile profilometer over a range of 4 mm can be performed within about 30 s, depending on the desired lateral resolution. The results show that the thickness gradient was captured with a mean height of 65.9 ± 12.3 nm. Additionally, some significant noise with a value of ± 3.6 nm was observed in the data. In contrast to the scanning acquisition, the thickness gradient could be determined in a single acquisition along the lateral domain using the DE-LCI approach, Fig. 5.3 b). In direct comparison, the captured profile, representing the average of ten successive data acquisitions (10 x 30 ms), shows significantly lower noise of about 1.8 nm. Accordingly, the thickness gradient over 4 mm lateral measurement range was determined with (63.0 ± 1.6) nm. The lateral resolution was about 4.2 µm, determined by the designed magnification of M = 0.6. The significantly lower noise as

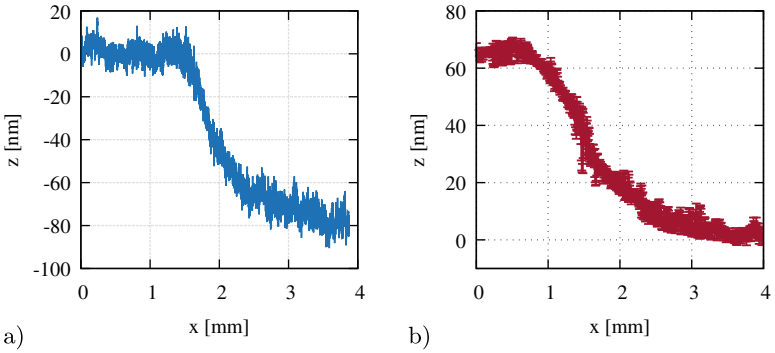

Figure 5.3 a) Plot of the thickness gradient, measured with a stylus profilometer as well as b) plot of the thickness gradient by DE-LCI

well as the higher resolution and measurement speeds are clear advantages of the DE-LCI approach in thin-film characterization.

5.3 Characterization of Flexible Substrate Materials

As described above, the transfer-matrix formalism is used as a mathematical basis for the fitting of experimentally acquired spectra. The influence of the substrate material parameters is usually low when analyzing thin-films on bulk substrates. Some applications of thin-films are fabricated on thin, flexible substrates such as polymer sheets, [289]. Thus, a more accurate knowledge of the material properties, especially of the thickness of these substrate materials, is necessary. In order to account for this, a dual-channel variation of the originally developed Mach-Zehnder interferometer was developed, Fig. 5.4.

In this setup, light is guided as before, see Fig. 5.1. Additionally, light from all interfaces but specifically that from the substrate was collected in a secondary detection path at the first beamsplitter. This path was equipped with a high-resolution grating spectrometer (σ_{spec} = 0.3 nm, Avaspec ULS3648 VB, Avantes BV, Apeldoorn, The Netherlands). In this context, the surfaces of the substrate material can be considered as a resonator where interference occurs dependent on the distance of the surfaces, hence the thickness. By utilizing the spectrometer to record the spectral modulations of this interference signal, the actual thickness was calculated according to the principles of Fourier-domain optical coherence tomography, [105].

Sheets of polyethylene terephthalate (PET) foil substrate (t_s = 135 µm) with a coated ITO layer (nominal thickness t_{fn}^{ITO} = 150 nm), typically applied in the photovoltaics industry, were used as samples. The ITO coating was partly removed from the sample by means of chemical etching to generate a film-thickness gradient for spatial investigations. From the measured back-reflected interference signal, a Fourier analysis with appropriate x-axis scaling could be performed, Fig. 5.5 a). The data shows a significant amount of DC-signal components and noise, but also very clear peaks. These peaks could be attributed to the first and second order reflections of the substrate surfaces. The peak information was extracted by applying a Blackman-Harris window function and fitted with a Gaussian function. From the fitted data, the optical path distances of the reflections could be estimated with (450.4 and 443.2) µm respectively. With the knowledge of the refractive index of the substrate material, the thickness was calculated for both reflections with (137.3 and 135.1) µm. The thickness of the substrate material could be confirmed by mea-

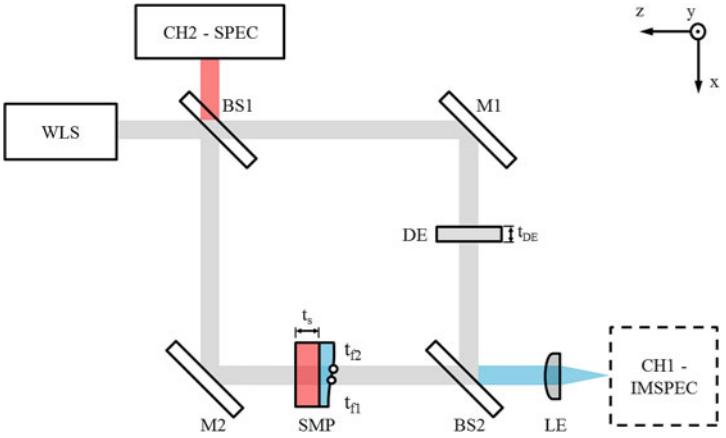

Figure 5.4 Dual-channel setup based on a Mach-Zehnder configuration with a WLS—white light source which is splitted 50:50 by BS1—first beamsplitter into a reference beam which is directed by M1—reference arm mirror and manipulated by a DE—dispersive element (t_{DE} = 6.23 mm) and the sample beam which is directed by M2—sample arm mirror through the SMP—sample with thin-film (thickness slope $t_f(x)$) on a substrate (t_s = 135 µm) and both are recombined by BS2—second beam splitter. The analysis is performed using a CH1—IMSPEC—primary, imaging spectrometer on which the recombined beam is imaged to with a LE—lens, here marked in blue as well as with the CH2—SPEC—secondary spectrometer which records the interference of the back-reflected signal from the substrate, here marked in red

surements on a tactile profilometer. With this result, an appropriate start value for the calculation of the film thickness with the imaging spectrometer is given. The method only allows the thickness calculation of the substrate material as the thin-film generates high-frequent interference which is not resolvable with a standard spectrometer.

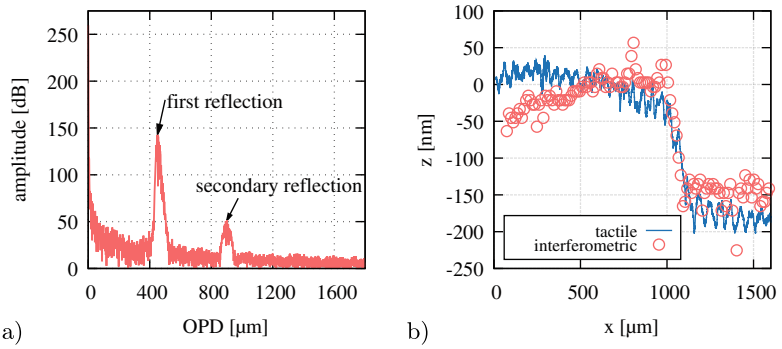

Figure 5.5 Results of film thickness measurements using a dual-channel interferometer with a) Fourier-analyzed data of the secondary channel with the x-axis scaled to the optical path difference (OPD) showing peaks from interference of two reflections which were used to calculate the substrate thickness as 137.3 and 135.1 µm respectively and b) slope of the ITO coating on this substrate measured in the primary channel having a ITO thickness of 151.6 nm in comparison to the slope measured on a tactile profilometer where the height was measured with 152.4 nm

By making use of the measured substrate thickness, data from the transmission measurement could be analyzed with the transfer-matrix formalism and appropriate fitting according to the model described with Eq. (3.4) and (3.6), Fig. 5.5 b). The results show that the slope of an ITO film on a PET substrate can be sampled with the appropriate resolution. From this slope the film thickness was measured with 151.6 nm. This corresponds well with a measurement of the sample with a tactile profilometer (Talysurf i-Series, Taylor Hobson Ltd, UK) showing a measured thickness of 152.4 nm. In the data, it is noticeable that the tactile profilometer shows significant, periodic noise which can be attributed to the surface roughness of the substrate material. In direct comparison the surface roughness is less pronounced in the interferometric data. Furthermore, the interferometric system was capable of resolving the slope as well as the film thickness with high precision. This simultaneous measurement helped to improve the accuracy of the fit model for the thin-film thickness determination without compromising the actual measurement.

While ellipsometric approaches can be error-prone on e.g. flexible substrates, the demonstrated approach is capable to measure film thickness in this setting with high accuracy. In comparison to spectral reflectometry, another state-of-the-art technology, the developed approach enables comparable resolutions on film thickness measurements while also maintaining a far larger measurement range of about 80 µm. This enables the simultaneous capture of the film thickness as well as the thickness of substrate materials. In summary, the DE-LCI approach shows significant advantages regarding accuracy, measurement time and the variety of possible samples, Tab. 5.1.

Table 5.1 Comparison of state-of-the-art technologies with the developed DE-LCI approach for thin-film characterization

approach	meas. time	resolution [nm]	spatial resolution	inline	flexible substrates	comments
reflectometry [179]	60 s	10 nm	no	yes	yes	single point measurements, large integration times necessary
ellipsometry [211]	8 s/λ	0.01 nm	yes	yes	no	gold standard, high accuracy
DE-LCI	50 ms	0.1 nm	yes	yes	yes	combines advantages