Chapter 1

Theory and Instrumental Details

1.1 Principle of Ellipsometry

The measurements described in this thesis were made using spectrally resolved ellipsometry. While the basic experimental setup of an ellipsometer is relatively simple, the interpretation of the measured spectra is not trivial.

In ellipsometry, the change in polarization of a light beam upon reflection on a sample is measured. The basic setup of an ellipsometer is shown in figure 1.1. A light beam from a white light source or a monochromatic source such as a laser is polarized and incident on the sample under an angle ϕ_0 with the surface normal. The directly reflected light from the sample surface is analyzed using a second polarizer. The resulting intensity in dependence on the analyzer angle is then detected at the detector.

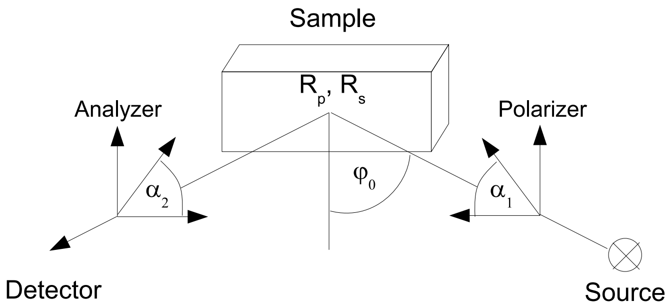

Figure 1.1: Schematic setup of an ellipsometer. R_p, R_s: complex reflection coefficients for parallel and perpendicular component, respectively; α_1, α_2: angle of the polarizer and analyzer; ϕ_0: angle of incidence.

The polarization change is quantified in terms of the ellipsometric parameters Ψ and Δ. These parameters are related to the sample properties by

$$\tan\Psi\, e^{i\Delta} = \frac{R_p}{R_s} \quad , \tag{1.1}$$

where R_p and R_s are the complex reflection coefficients for the parallel and perpendicular polarization component, respectively [6]. The parallel and perpendicular direction are defined with respect to the plane of incidence, which is the plane defined by the surface normal and the incident beam. The complex reflection coefficients describe how the linear polarization component in question changes in amplitude and phase factor due to the interaction with the sample. Therefore, Ψ basically describes the relative change in amplitude of the two components, while Δ describes the relative change in phase of the two linearly polarized components.

Ellipsometry is a technique highly sensitive to surface properties. The direct reflection configuration results in a technique that is not sensitive to misalignment of the sample. A slight deviation of the real angle of incidence from the expected angle defined by the position of the ellipsometer's goniometer arms is immediately recognized by a vanishing signal at the detector. The surface sensitivity is even more pronounced due to the fact that the method works at high angles of incidence close to the Brewster angle.

Since the complex reflection coefficients usually depend on the morphological properties of the sample such as layer thicknesses and interface roughnesses as well as on the optical parameters of the components making up the system, the interpretation of the ellipsometric spectra is complicated. In fact, only for the case of a homogeneous, isotropic, and within the penetration depth infinitely thick sample with a perfect surface can the ellipsometric parameters be converted analytically to the sample parameters. In this case the complex refractive index of the sample can be determined. In the case of more complex structured systems, the right hand side of equation 1.1 is a nonlinear, transcendent equation. It can therefore not be converted analytically to yield the sample parameters of interest. In such cases, a model of the system has to be developed and numerical optimization techniques have to be used to obtain the parameters of the sample from the ellipsometric measurements.

1.1.1 Maxwell Equations

The basic foundation for the interpretation of ellipsometric data are the Maxwell equations of electrodynamics [7, 8]. They are given by

$$
\begin{aligned}
\vec{\nabla} \times \vec{H} &= \frac{1}{c}\frac{\partial \vec{D}}{\partial t} + \frac{4\pi}{c}\vec{j} \\
\vec{\nabla} \times \vec{E} &= \frac{1}{c}\frac{\partial \vec{B}}{\partial t} \\
\vec{\nabla} \cdot \vec{D} &= 4\pi\rho \\
\vec{\nabla} \cdot \vec{B} &= 0 \quad ,
\end{aligned}
\tag{1.2}
$$

where $\vec{H}$, $\vec{D}$, $\vec{E}$ and $\vec{B}$ are the magnetic field strength, the electric displacement density, the electric field strength, and the magnetic flux density, respectively. The parameter $\vec{j}$ is the surface current and ρ is the charge density.

The associated material relations describe the influence of materials other than vacuum on the electromagnetic wave:

$$\begin{aligned} \vec{D} &= \epsilon \vec{E} \\ \vec{B} &= \mu \vec{H} \\ \vec{j} &= \sigma \vec{E} \quad . \end{aligned} \tag{1.3}$$

In the case of non magnetic materials the magnetic permeability $\mu = 1$. It is important to note that this theory describes only classical macroscopic parameters, e.g. dielectric function or complex refractive index. A detailed analysis of the scattering processes may require the use of quantum mechanics.

1.1.2 Mathematical Representation of Polarization

The term polarization refers to the time dependence of the electric field vector at a fixed point in space. The electric field vector is usually chosen to describe the electromagnetic wave, because the magnetic field vector is related to the electric field vector by the Maxwell equations and the interaction of electromagnetic waves with matter is usually largely determined by the electric interaction.

Assume that the electromagnetic wave in question is a monochromatic plane wave with a direction $\vec{k}$ parallel to the z-axis of the coordinate system. Using the fact that a monochromatic plane wave may be written as the superposition of two linear polarized and mutually orthogonal waves

$$\vec{E}(\vec{r},t) = \begin{bmatrix} E_x(\vec{r},t) \\ E_y(\vec{r},t) \end{bmatrix} = \begin{bmatrix} |E_x| e^{-i(\vec{k}\vec{r}-\omega t-\delta_x)} \\ |E_y| e^{-i(\vec{k}\vec{r}-\omega t-\delta_y)} \end{bmatrix} \quad , \tag{1.4}$$

it can be shown that in the most general case the endpoint of the electric field vector $\vec{E}$ traces the outline of an ellipse. The wave is said to be elliptically polarized, as opposed to e.g. linearly polarized, where the electric field vector stays in a plane [6].

An alternative way to represent the polarization of an electromagnetic wave is the Stokes vector [6, 7, 9]. The components of this four-dimensional vector $S = \{S_0, S_1, S_2, S_3\}$ are defined as

$$\begin{aligned} S_0 &= |E_x|^2 + |E_y|^2 \\ S_1 &= |E_x|^2 - |E_y|^2 \\ S_2 &= 2|E_x||E_y|\cos(\delta_y - \delta_x) = 2|E_x||E_y|\cos\Delta \\ S_3 &= 2|E_x||E_y|\sin(\delta_y - \delta_x) = 2|E_x||E_y|\sin\Delta \quad . \end{aligned} \tag{1.5}$$

The components of the Stokes vector all have the unit of intensity. It can be seen easily that only three of its components are independent

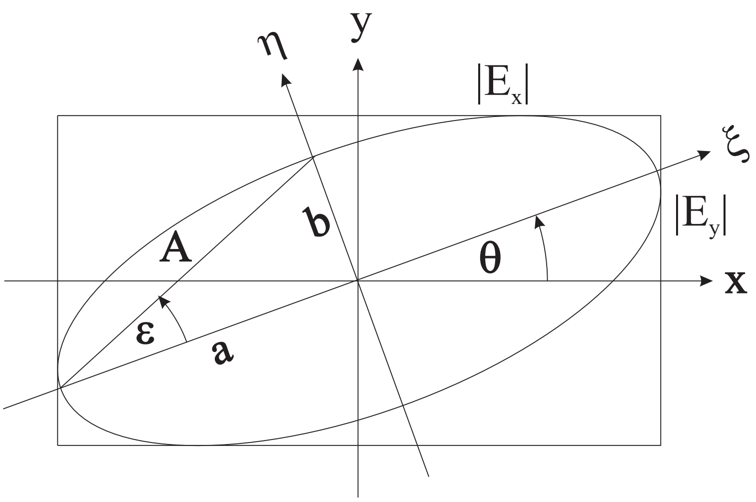

Figure 1.2: Parameters determining the Polarization Ellipse: Azimuth angle Θ, amplitude A and ellipticity $\tan\epsilon = b/a$. The time dependence of the electric field vector can be described by superposition of two linearly, along the coordinate axis polarized orthogonal waves $E_x(\vec{r}, t)$ and $E_y(\vec{r}, t)$.

$$s_0^2 = s_1^2 + s_2^2 + s_3^2 \quad . \tag{1.6}$$

The Stokes vector is important in photometric ellipsometry, because they are related to the intensity at the detector and the ellipsometric parameters Ψ and Δ, as explained in section 1.1.4.

1.1.3 Jones Vector and Jones Matrix Formalism

A representation of the polarization properties of an electromagnetic wave which is especially useful when discussing the propagation of polarized light through optical components is the Jones vector representation [6].

In the Jones vector formalism, the polarization of an electromagnetic wave is represented by the superposition of two different basis states. A typical choice of basis states are two orthogonal linear polarizations, but other choices such as two circular polarizations with different handedness are possible. The choice of basis states is usually guided by the system of optical elements under consideration, which may make one choice mathematically easier to handle. In the following, it is assumed that the basis states are two linear polarized waves oriented along the x and y axis of the coordinate system, respectively.

In ellipsometry only the polarization properties are of interest. Therefore, information about the absolute time dependence may be suppressed without loss of information. A complete representation of the polarization state is then given by

$$\vec{E} = \begin{bmatrix} E_x \\ E_y \end{bmatrix} \quad , \tag{1.7}$$

with

$$E_x = |E_x|e^{i\delta_x}, \quad E_y = |E_y|e^{i\delta_y} \quad . \tag{1.8}$$

The two components are complex numbers, also called phasors. They describe the amplitude of the two basis vectors and their phase difference. The superposition of these two components restores the polarization ellipse. In the Jones formalism, optical components are described by 2×2 matrices. For example, the effect of a linear polarizer with an azimuth angle α with respect to the xy-coordinate system on a linear polarized wave with Jones vector $\vec{E_i}$ is analogous to premultiplying this Jones vector by a matrix given by

$$\vec{E_o} = \begin{bmatrix} \cos\alpha & 0 \\ 0 & \sin\alpha \end{bmatrix} \vec{E_i} \quad . \tag{1.9}$$

Starting with an input Jones vector E_i, the resulting output Jones vector E_o after a sequence of optical elements can be obtained by subsequently premultiplying the input Jones vector E_i with the Jones matrices representing the different optical elements.

1.1.4 Photometric Ellipsometry

Several different ellipsometry setups exist, which differ in the way the ellipsometric parameters Ψ and Δ are derived, e.g., null ellipsometers and photometric ellipsometers. The ellipsometer used for the measurements in this thesis is of the latter type.

Given the simple ellipsometric setup of figure 1.1, the resulting ellipsometric parameters Ψ and Δ can be calculated as follows [9].

Using the Jones matrices of 1.1.3 for the polarizers and the sample, the intensity at the detector is equal to

$$I(\alpha_2) = \frac{1}{2}(s_0 + s_1\cos(2\alpha_2) + s_2\sin(2\alpha_2)) \quad , \tag{1.10}$$

where the coefficients s_x refer to the Stokes' vector components. The intensity is periodic with periodicity $2\alpha_2$. The Stokes vector components may, therefore, be calculated as the Fourier coefficients of the intensity by measuring several times at fixed polarizer angle and variable analyzer angle

$$\begin{aligned} s_1 &= 2\frac{\sum_i I(\alpha_{2i})\cos(2\alpha_{2i})}{\sum_i I(\alpha_{2i})} \\ s_2 &= 2\frac{\sum_i I(\alpha_{2i})\sin(2\alpha_{2i})}{\sum_i I(\alpha_{2i})} \end{aligned} \quad . \tag{1.11}$$

The ellipsometric parameters Ψ and Δ are given by

$$\cos 2\Psi = -\frac{s_1}{s_0}$$
$$\sin 2\Psi \cos \Delta = \frac{s_2}{s_0} \tag{1.12}$$

Since the ellipsometric parameters are included in expressions $\cos 2\Psi$ and $\sin 2\Psi \cos \Delta$, the parameter Δ can only be defined in the range $0° \leq \Delta \leq 180°$. By usage of an additional retarder with phase retardation δ, the expression $\sin 2\Psi \cos(\Delta + \delta)$ can be measured. In particular, for $\delta = 90°$, also the third Stokes vector component s_3 can be determined by

$$\sin 2\Psi \sin \Delta = \frac{s_3}{s_0} \quad , \tag{1.13}$$

so that Δ may be determined without ambiguity in the whole range $0° \leq \Delta \leq 360°$. Besides resolving the ambiguity of Δ, the usage of a retarder also solves a problem arising from very small values of Δ. In this case $|\cos \Delta| \approx 1$, and small errors in the measurement of $\cos \Delta$ lead to large errors in the determination of Δ. By transforming $\cos \Delta$ to $\sin \Delta$, the problem is shifted to a region where measurement errors have only a small impact and are not problematic in the numerical inversion.

1.2 Modelling Ellipsometric Spectra

As mentioned before, the interpretation of ellipsometric spectra usually requires the modelling of the morphological and optical properties of the sample. In the following, the basic ingredients for the modelling of layered samples, possibly with interface roughnesses, are described. Firstly, the reflection at a single interface is described and the inversion of the ellipsometric parameters to the complex refractive index is presented. In the following, mathematical tools for the modelling of layered systems with isotropic and homogeneous layers are shown. The effective medium approximation (EFM) for modelling systems consisting of several different constituents and interface roughnesses is introduced in section 1.2.3.

1.2.1 Reflection on a Bulk System - the Fresnel Coefficients

The equations most often used in the modelling of ellipsometric measurements are the Fresnel coefficients. The Fresnel coefficients describe the change in amplitude and phase of an electromagnetic wave upon reflection or transmission on a perfectly smooth interface between two homogeneous and isotropic media.

There are four Fresnel coefficients corresponding to reflection and transmission for electromagnetic waves polarized parallel and perpendicular to the plane of incidence. They can be found by stating the boundary conditions on the continuity of the field components normal and tangential to the surface:

$$\begin{aligned} \vec{n}_{12} \cdot \left(\vec{D}_2 - \vec{D}_1\right) &= 4\pi\vec{\rho} \\ \vec{n}_{12} \times \left(\vec{E}_2 - \vec{E}_1\right) &= 0 \\ \vec{n}_{12} \cdot \left(\vec{B}_2 - \vec{B}_1\right) &= 0 \\ \vec{n}_{12} \times \left(\vec{H}_2 - \vec{H}_1\right) &= \frac{4\pi}{c}\vec{j} \quad , \end{aligned} \tag{1.14}$$

where $\vec{n_{12}}$ is the unit vector normal to the surface and directed from the first into the second medium. In the case of vanishing surface charge and current $\vec{\rho}$ and $\vec{j}$, these equations reflect the continuity of the components of $\vec{D}$ and $\vec{B}$ perpendicular to the boundary, and the tangential components of $\vec{E}$ and $\vec{H}$. From these boundary conditions the so called Fresnel equations can be derived [7]:

$$\begin{aligned} r_p &= \frac{E_{rp}}{E_{ip}} = \frac{N_1 \cos\phi_0 - N_0 \cos\phi_1}{N_1 \cos\phi_0 + N_0 \cos\phi_1} \\ r_s &= \frac{E_{rs}}{E_{is}} = \frac{N_0 \cos\phi_0 - N_1 \cos\phi_1}{N_0 \cos\phi_0 + N_1 \cos\phi_1} \\ t_p &= \frac{E_{tp}}{E_{ip}} = \frac{2N_0 \cos\phi_0}{N_1 \cos\phi_0 + N_0 \cos\phi_1} \\ t_s &= \frac{E_{ts}}{E_{is}} = \frac{2N_0 \cos\phi_0}{N_0 \cos\phi_0 + N_s \cos\phi_1} \quad , \end{aligned} \tag{1.15}$$

with the complex refractive indices N_0 and N_1 of the first and the second medium, respectively. The subscripts r,t and i refer to reflected, transmitted and incident wave, while the subscripts p and s refer to parallel and perpendicular polarization with respect to the plane of incidence. The in general complex angle ϕ_1 can be derived as a function of ϕ_0, N_0 and N_1 by use of Snell's law [7]

$$N_0 \sin\phi_0 = N_1 \sin\phi_1 \quad . \tag{1.16}$$

In the case of a bulk sample with an infinite thickness when compared to the penetration depth, the complex reflection coefficients are equal to the Fresnel coefficients. In this case the ellipsometric equation can be written as [6]

$$\tan\Psi\, e^{i\Delta} = \frac{R_p}{R_s} = \frac{r_p}{r_s} \quad .$$

By substituting the Fresnel coefficients, the following explicit relation for the complex refractive index of the bulk medium N_1 in terms of the ellipsometric parameters Ψ and Δ and the angle of incidence ϕ_0 can be found [6]

$$N_1 = N_0 \tan\phi_0 \left[1 - \frac{4\rho}{(1+\rho)^2}\sin^2\phi_0\right]^{1/2} \quad , \tag{1.17}$$

where N_0 is the complex refractive index of the surrounding medium, which is usually air and can be set to 1, while ρ is related to the ellipsometric parameters Ψ and Δ by 1.1.

1.2.2 Layered systems

Ellipsometry is especially interesting when measuring layered systems and systems with interface roughnesses. The simultaneous measurement of interference phenomena in both the amplitude and phase information leads to a technique which can be used to characterize parameters such as layer thicknesses with high precision. In the following the standard ambient/film/substrate system is described as well as a general approach for modelling stratified planar isotropic systems. In addition, the effective medium approximation for modelling mixtures of optically different media and roughness effects is presented. Usually, the resulting formulae cannot be analytically inverted for the sample parameters, especially the layer thicknesses. Therefore numerical fit procedures have to be employed to invert them.

1.2.2.1 Ambient/Film/Substrate system

Figure 1.3 shows the paths taken by a light beam in the classic ambient/film/substrate model consisting of a homogeneous, isotropic film sandwiched between semi infinite homogeneous, isotropic ambient and substrate. The film layer leads to multiple internal reflections of the beam inside the film. These reflections lead to additional reflexes beside the main reflection from the film surface. The partial waves leaving the sample can interfere with each other, provided that the film's thickness and, therefore, the resulting lateral separation is not too large.

The resulting complex reflection coefficients are a function of the Fresnel coefficients of the different interfaces and the thickness of the film. They can be found by summing up the Fresnel coefficients of the paths taken by the different partial waves:

$$R_x = r_{01x} + t_{01x}t_{10x}r_{12x}e^{-i2\beta} + t_{01x}t_{10x}r_{10x}r_{12x}^2e^{-i4\beta} + \ldots \quad , \tag{1.18}$$

where the coefficients r_x and t_x are the Fresnel coefficients of equation 1.15 for reflection at and transmission through the interface, respectively. The subscript x stands for either p- or s-polarization with respect to the plane of incidence.

The parameter β is called the phase angle. It is the phase shift of the electromagnetic wave due to travelling a distance within the film and is therefore related to the thickness d of the film, the angle of incidence ϕ_0 and the complex refractive index of the film N_1.

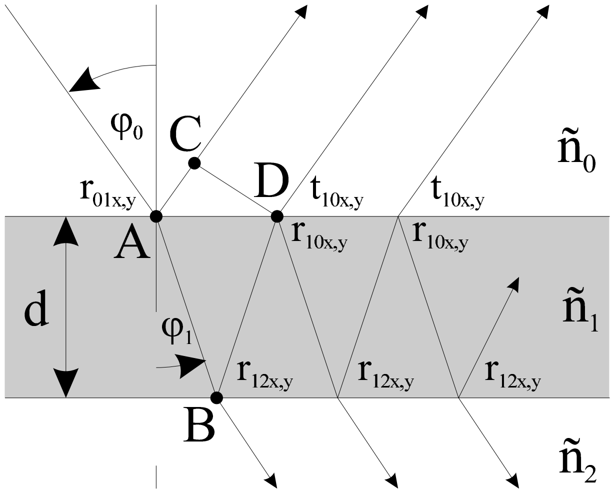

Figure 1.3: Interference in an ambient/film/substrate system

$$\beta = 2\pi\frac{d}{\lambda}(N_1^2 - N_0^2\sin^2\phi_0)^{1/2} \quad , \tag{1.19}$$

Equation 1.18 is an infinite geometric series and can be reduced to

$$R_x = \frac{r_{01x} + r_{12x}e^{-i2\beta}}{1 + r_{01x}r_{12x}e^{-i2\beta}} \quad . \tag{1.20}$$

These are the so-called Airy formulae [7]. From these equations expressions for the ellipsometric parameters according to eq. 1.1 can be derived:

$$\tan\Psi\, e^{i\Delta} = \frac{R_p}{R_s} = \frac{r_{01p} + r_{12p}e^{-i2\beta}}{1 + r_{01p}r_{12p}e^{-i2\beta}}\,\frac{1 + r_{01s}r_{12s}e^{-i2\beta}}{r_{01s} + r_{12s}e^{-i2\beta}} \quad . \tag{1.21}$$

1.2.2.2 Stratified planar isotropic systems

A uniform approach for the analysis of ellipsometric spectra from stratified planar isotropic systems is based on a 2×2-matrix formalism [6]. The influence of the sample on the polarization of the electromagnetic wave is described by a 2×2 scattering matrix S

$$S = \begin{bmatrix} S_{11} & S_{12} \\ S_{21} & S_{22} \end{bmatrix} \quad .$$

Let light be incident on a layered structure that is stratified along the z-axis. The scattering matrix relates the forward-travelling wave $E^{+}(z)$ and backward-travelling wave$E^{-}(z)$ at two planes z' and z''