A Practical Guide to Optical Metrology for Thin Films

Michael Quinten

This book presents a comprehensive overview of optical metrology for thin film thickness determination by optical means, from electrodynamic basics, and hardware components, to methods of measurement and evaluation. The author, an expert in the field with both academic and industrial experience, concentrates on the spectral reflectance measurement with miniaturized spectrometers, one of the most flexible techniques for inline and offline thickness determination.

From the contents:

- Propagation of Light and other Electromagnetic Waves
- Spectral Reflectance and Transmittance of a Layer Stack
- The Optical Measurement
- Thin Film Thickness Determination
- The Color of Thin Films
- Applications

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A Practical Guide to Optical Metrology for Thin Films
An artificially oxidized bismuth crystal superimposed on a background of medieval glass. After a long period in soil alteration formed a thin nanometric film of layer silicates with locally different thickness on the glass. The thickness of the nanometric oxide film on the crystal is locally different too. (Photograph of crystal by U. Quinten; photograph of glass by G. Müller, www.mueller-mineralien.de).
To my family
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Preface

The optical response of a thin film is determined by several parameters: its thickness, its optical properties, and the surrounding (other layers, substrates). Among them, its thickness $d$ is the most important. Compared to the vacuum wavelength of light $\lambda$, it must have a certain value to establish characteristic features in reflectance, transmittance, or ellipsometric parameters by interference. The size can be reduced by a factor $n$, with $n$ being the refractive index of the film. The reason is that the optical thickness $t = n \cdot d$ is the intrinsic parameter that must be compared with $\lambda$.

The measurement of reflectance, transmittance, or ellipsometric parameters has become a major tool for in-line inspection, process control, and quality control of thin films since it is fast, contactless, nondestructive, and even cheap compared to other methods.

During my successful stint with industry from 2001 till date, I have become acquainted with several aspects of optical thin-film metrology. It is a very fascinating subject since it connects electrodynamics with solid-state physics. The input parameters of any evaluation algorithm are never constant but may vary from one measurement task to the next because the optical material functions strongly depend on film manufacturing, composition, and stochiometry. Film thickness determination then becomes also a question of refractive index determination.

The purpose of this book is to introduce in optical metrology for thin film thickness determination. It provides information on the electrodynamic basics and methods of measurement and evaluation. Hence, it is directed at all people who are involved in measuring film thickness by optical means, whether as manufacturer, in process and quality control, or in research and development. Hopefully, university lecturers and students of natural sciences and engineering will also find this book beneficial.

To write this book required reading and evaluating many monographs and a still larger number of publications on this subject. To my surprise, a lot of work has been done in ellipsometry, but spectral reflectance measurement for film thickness determination is sparsely described in literature although it is a well-established method. The total amount of published work is, however, too immense to consider them all in such a book. Therefore, I hope to have included the most relevant up to date, and apologize for all the contributions not considered here.
Last but not least, I want to greatly acknowledge all the people who supported me with data material, helpful information, and measurements, namely, Thomas Fries and Juergen Koglin from FRT GmbH, Anke Orth, Faiza Houta, and Bjoern Lewald from FRT GmbH, Alexei Maznev from the Massachusetts Institute of Technology, and Leif J. Hoglund from Semilab AMS. Many thanks to Gerhard Mueller for the picture of the glass on the cover, to my wife Ulrike for the picture of the oxidized Bi crystal on the cover, and finally to my family for their support and patience with me during writing this book.

Aldenhoven,  
February 19, 2012

Michael Quinten
1 Introduction

Thin films of transparent or semitransparent materials play an important role in our life. A variety of colors in nature are caused by the interference of light reflected at thin transparent layers. Examples are the iridescent colors of a peacock feather, the impressive colors of lustrous butterfly wings, or simply the play of colors of thin oil films on water.

Much more demonstrative is, however, the use of thin films in technical applications. Films with maximum thickness of a few hundred nanometers are used as protective layers, hard coatings, antireflection coatings, adhesion and antiadhesion coatings, decorative coatings, transparent conductive layers, absorbing layers, in biosensors, and for tinted and annealed architectural glass. The combination of many thin films in multilayer stacks even lead to optical filters with sharp edges in reflection and transmission and almost 100% reflectivity in certain desired spectral ranges. The highest commercial impact these films have in microelectronics. Most microelectronic parts (processors, RAMs, flat screens, CDs/DVDs, hard disks, and some more) are manufactured with the help of thin-film technology. Thicker films of mainly transparent plastics are almost everywhere present as food packaging, wrapping, foils, membranes, lamination, and in display technology and solar cells, to give some examples.

Hence, it is our attempt to get as much information as possible on the properties and composition of surfaces and surface coatings. The two main classes of thin-film measurements are optical and stylus-based techniques. When measuring with a (mechanical) stylus, the thickness and roughness are obtained by monitoring the deflections of the fine-tipped stylus as it is dragged along the surface of the film. Stylus instruments, however, require a step in the film to measure thickness, even when using comparable optical sensors such as chromatic white light sensors. They are often the preferred method when measuring opaque films, such as metals.

Optical techniques determine the thin-film properties by measuring how the films interact with light. They can measure the thickness, roughness, and optical constants of a film. Optical techniques are usually the preferred method for measuring thin films because they are accurate, nondestructive, and require little or no sample preparation. The two most common optical measurement types are the spectral reflectance measurement and the ellipsometry. They form the main subject of this book. Besides, there exist other nondestructive methods for film thickness determination.
with more or lower capabilities. Among them we find magnetoinductive and capacitive methods and the eddy current method, as well as the indirect measurement by a vibrating quartz or the measurement with ultrasound. Optical methods comprise light section, X-ray total reflection, photothermal deflection, and confocal chromatic measurement.

Spectral reflectance measurement or reflectometry uses the intensity of the light and measures the amount of light reflected from a thin film or a multilayer stack over a range of wavelengths, with the incident light normal (perpendicular) to the sample surface. Spectral reflectance can also measure the thickness, roughness, and optical constants of a broad range of thin films. However, if the film is very thin so that there is less than one reflectance oscillation, there is insufficient information available to determine the film parameters. Therefore, the number of film properties that may be determined decreases for very thin films. If on the other hand one attempts to solve for too many parameters, a unique solution cannot be found, but more than one possible combination of parameter values may result in a calculated reflectance that matches the measured reflectance. Depending upon the film material and the wavelength range of the measurement, the minimum single-film thickness that can be measured using spectral reflectance is in the 20–100 nm range. Additional determination of optical constants increases this minimum thickness. Nevertheless, as spectral reflectance is much simpler and less expensive than the second most common optical measurement—the ellipsometry—it is often used for quick and easy offline and in-line thickness determination in laboratories, production, and process control. To our knowledge, no comprehensive book on reflectometry as it is being practiced exists except for the one by Tompkins and McGahan [1], published in 1999. Therefore, one intention of this book is to bring the reflectometry closer to the practitioner.

In the late 1800s, Paul Drude [2] used the phase shift induced between the perpendicular components of polarized light to measure film thickness down to a few nanometers. This was the first study on film thickness measurement with a method that was later called ellipsometry. When the perpendicular components of polarized light are out of phase, the light is said to be elliptically polarized, for which this technique came to be called ellipsometry. Ellipsometry measures reflectance at nonnormal incidence (typically around 75° from normal) and is rather sensitive to very thin layers. The two different polarization measurements provide twice as much information for analysis. Variable-angle ellipsometry can be used to take reflectance measurements at many different incidence angles, thereby increasing the amount of information available for analysis. In 1977, Azzam and Bashara [3] authored the book Ellipsometry and Polarized Light, which has been the key source to be cited in most technical writing on the subject. Later on, several handbooks were published [4–6] that cover the theory of ellipsometry, instrumentation, applications, and emerging areas, in which experts in the field contributed to various aspects of ellipsometry. Fundamental principles and applications of spectroscopic ellipsometry are to be found in the recently published work of Fujiwara [7].

This book starts with Chapter 2 with an introduction to the basics of the propagation of light and other electromagnetic radiation in space and matter. Beyond the general properties of electromagnetic waves, we consider mainly the deviations
from the straightforward propagation by reflection, refraction, and diffraction since they are important for understanding the optical layer thickness determination and the functioning of the optical measuring devices. Interference of electromagnetic waves is a key effect not only for the diffraction of light but also for the optical layer thickness determination as it causes characteristic deviations in the reflectance spectrum of a thin film. From this characteristic interference pattern, all the film parameters are finally deduced.

Optical thickness determination is not only a question of electrodynamics but also a question of solid-state physics. The reason is that propagation in matter also means interaction of the electromagnetic wave with the matter. This interaction can be described with the complex dielectric function, while when discussing wave propagation in and through media the complex refractive index is appropriate. Both are connected via Maxwell’s relation. In Chapter 2, we discuss physical models for the dielectric function and present empiric formulas for the refractive index.

The main topics of this book, the determination of the thickness of a layer in a layer stack from measurement of the spectral reflectance or transmittance, is treated in Chapters 3–5. The first step is taken in Chapter 3 with the modeling of the spectral reflectance \( R \) and transmittance \( T \) of a layer stack. Giving the thicknesses and complex refractive indices of all layers and substrates of the layer stack as input parameters, two common models – the propagating wave model and the \( r-t-\phi \) model – can be used to calculate \( R \) and \( T \) of the stack (see Figure 1.1). The models are introduced in

![Figure 1.1](image-url)  
**Figure 1.1** Modeling the reflectance \( R \) and transmittance \( T \) or ellipsometric data of a layer stack.
Chapter 3 and extensions on surface roughness and incoherent substrates are discussed. Absorption of light in the layer restricts the measurability of the thickness to a material-dependent maximum thickness.

In Chapter 4, we introduce the reflectometric and ellipsometric measurement and further optical methods, and discuss the optical components needed for the measurements. In all setups for optical thickness determination, the sample gets illuminated. Hence, light sources and their spectral distribution play a key role in the layer thickness determination, as well as the second key component, spectrometers. With the spectrometer, the reflected light modulated by the thickness interference gets spectrally resolved and analyzed.

Reflectometric and ellipsometric measurements do not measure the physical properties themselves but the optical response of the system caused by the physical properties. Hence, one needs to solve an inverse problem in order to find the value of actual physical properties of interest, such as thicknesses of the layers and optical properties of the materials. This inverse problem is solved numerically by finding the best fit between measured and calculated data, and physical properties are inferred from the model that gives the best fit (see Figure 1.2). To get reliable results, it is important to check the validity of the used model and to understand the sensitivity of the measured data to parameters of interest. In Chapter 5, we present and discuss numerical methods for determination of layer thickness and determination of optical constants of the layer material.

Chapter 6 is devoted to the apparent color of thin films. As the photographs on the cover of this book demonstrate, the interference in thin films leads to various colors depending on the thickness and refractive index of the film. However, not all colors

![Figure 1.2](image_url)

Figure 1.2 Fit procedure when analyzing measured $R$, $T$, or ellipsometric data for film thickness.
are available from one single layer. Instead, multilayer systems are needed to cover a certain color gamut.

Finally, in Chapter 7 we present several technical applications where film thickness measurement is important. They are accompanied by corresponding measuring results. The applications can be classified into the following:

- Applications with a single unsupported layer, for example, glass, sapphire, or semiconductor wafers, and transparent polymer films.
- Applications with one layer on a substrate, for example, protective layers (hard coats), broadband antireflection coatings, photoresists, and transparent conductive layers (TCF and TCO).
- Applications with two layers on a substrate. Examples of two layers on a substrate are photoresists on silica on a wafer, bonded wafers, and SOI wafers (SOI, silicon on insulator).
- Multilayer applications, for example, high reflective (HR) and antireflective (AR) coatings, beam splitter coatings, dielectric mirrors, optical filters, thin-film solar cells, and OLEDs (organic light emitting diodes).

We want to point out that all calculations of reflectance and transmittance spectra, the evaluation of thickness parameters and color, and the determination of optical constants were carried out with self-made software packages, MQLayer, MQNandK, and MQColor [8].
2
Propagation of Light and Other Electromagnetic Waves

This chapter introduces the basics of the propagation of light and other electromagnetic radiation in space and matter. Beyond the general properties of electromagnetic waves, we consider mainly the deviations from the straightforward propagation by reflection, refraction, and diffraction since they are important for understanding the optical layer thickness determination and the functioning of the optical measuring devices. Last but not least, propagation in matter also means interaction of the electromagnetic wave with the matter for what we also discuss the dielectric function and the refractive index in this chapter.

2.1
Properties of Electromagnetic Waves

When discussing the properties of electromagnetic waves, it seems appropriate to give first a definition of a wave. A wave generally is a process that is periodic in time and space. That means there exists a periodicity \( T \) in time after that the wave looks the same as at a certain time point \( t \), and a periodicity \( R \) in space where the wave looks the same as at a certain point \( r \):

\[
A(r + R, t + T) = A(r, t).
\]

Mathematically, \( A(r,t) \) fulfills the wave equation (in Cartesian coordinates):

\[
\left( \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right) A(r, t) = 0,
\]

with \( c \) being the propagation velocity. That means, in general, we search for a vector with its second derivative in time being proportional to its second derivative in space. The actual solution, however, is additionally determined by the boundary conditions of this differential equation.
When talking about electromagnetic waves, we often find, though it is not mandatory, that solutions of the wave equation are harmonic functions in time and space like

\[ A(\mathbf{r}, t) = A_0 \cdot \exp(i(\mathbf{k} \cdot \mathbf{r} - \omega t + \phi)), \]

with \( k = |\mathbf{k}| = 2\pi/|\mathbf{R}| \), \( \omega = 2\pi/T \), and \( \phi \) an arbitrary constant. The length \( |\mathbf{R}| \) is called \textit{wavelength} \( \lambda \) and describes the distance between two successive identical phases of the wave in space, for example, the distance between two maxima or two minima. The propagation velocity corresponds to the vacuum velocity of light \( c = 299 792 458 \text{ m/s} \).

\( k \) and \( \omega \) fulfill the \textit{dispersion relation}

\[ k^2 = \frac{\omega^2}{c^2}. \]

Note that in (2.3) we used the notation with complex numbers, with \( i = \sqrt{-1} \) being the complex unit. This notation will be used throughout the book. For an introduction to the numerics with complex numbers, we refer to Appendix A.

For electromagnetic waves, we have to consider an electric field \( \mathbf{E}(\mathbf{r}, t) \) and a magnetic field \( \mathbf{H}(\mathbf{r}, t) \) that must fulfill on the one hand the above conditions for \( A(\mathbf{r}, t) \) and on the other hand Maxwell’s equations:

\[ \text{div}(\mathbf{D}) = \rho, \]

\[ \text{div}(\mathbf{B}) = 0, \]

\[ \text{curl} \ \mathbf{E} = \frac{\partial \mathbf{B}}{\partial t}, \]

\[ \text{curl} \ \mathbf{H} = \mathbf{J} + \frac{\partial \mathbf{D}}{\partial t}. \]

These equations, however, contain three more vectors, the current density \( \mathbf{J} \), the electrical flux density or displacement \( \mathbf{D} \), and the magnetic induction \( \mathbf{B} \). To resolve Maxwell’s equations for \( \mathbf{E} \) and \( \mathbf{H} \), it is therefore necessary to supplement them by relations that connect \( \mathbf{J} \), \( \mathbf{D} \), and \( \mathbf{B} \) with \( \mathbf{E} \) and \( \mathbf{H} \).

When applying an electric field \( \mathbf{E} \) on any material, the electric field forces unbound charge carriers to move, resulting in a current with density \( \mathbf{J} \). The current is proportional to the applied field, with the \textit{conductivity} \( \sigma \) being the proportionality constant:

\[ \mathbf{J} = \sigma \mathbf{E}. \]

For bound charge carriers, the situation is different. They cannot move but are displaced. Inside the body of condensed matter the electric field usually displaces only the electrons while the ions are too inert as to follow the electric field. Thereby, each atom becomes an electric dipole with dipole moment \( \mathbf{p} \). These dipole moments add up to a macroscopic net polarization \( \mathbf{P} \) of the material that is related to the electric
field \( \mathbf{E} \) by the general equation

\[
P = \varepsilon_0 \chi \mathbf{E}.
\] (2.10)

The factor \( \chi \) is the macroscopic susceptibility of the matter. The polarization \( P \) contributes to the electrical flux density or displacement \( \mathbf{D} \):

\[
\mathbf{D} = \varepsilon_0 \mathbf{E} + \mathbf{P} = \varepsilon_0 (1 + \chi) \mathbf{E}
\] (2.11)

defining the dielectric constant or permittivity \( \varepsilon \) as

\[
\varepsilon = 1 + \chi.
\] (2.12)

Susceptibility \( \chi \) and dielectric constant \( \varepsilon \) are the optical material functions. In the framework of Maxwell’s theory, they enter the field relations as constants that are valid for the bulk material under consideration.

Applying a time harmonic electric field, the dipoles oscillate with the same frequency as the applied field. That means the center of gravity of the displaced electrons changes from one side to the other side, resulting in a displacement current \( \partial \mathbf{D} / \partial t \).

Similar to the electric field \( \mathbf{E} \), the magnetic field \( \mathbf{H} \) also causes two reactions of the material on the applied field, a magnetization \( \mathbf{M} \), and a current displacement \( \partial \mathbf{D} / \partial t \). The latter is the result of the Lorentz force on bound and free electrons. The magnetization \( \mathbf{M} \) contributes to the magnetic induction \( \mathbf{B} \) in the material

\[
\mathbf{B} = \mu_0 \mathbf{H} + \mu_0 \mathbf{M} = \mu \mu_0 \mathbf{H}
\] (2.13)

due to reorientation of permanent magnetic dipoles in the applied field. Looking at frequencies of electromagnetic waves, permanent magnetic dipoles are too inert as to follow a rapidly oscillating magnetic field. This holds true for frequencies ranging from the far infrared to infinity. Therefore, the relative permeability \( \mu \) can be assumed to be 1 throughout the above frequency range, even for magnetic materials.

Finally, we point to the fact that when dealing with electromagnetic waves, static charges are absent, that is, \( \varrho = 0 \). If we further restrict only to time harmonic fields for the sake of simplification, the time dependence of the fields can be separated with the ansatz

\[
\mathbf{E} = \mathbf{E}(r) \cdot \exp(-i \omega t) \quad \text{and} \quad \mathbf{H} = \mathbf{H}(r) \cdot \exp(-i \omega t)
\] (2.14)

and the corresponding Maxwell equations for the unknown parts \( \mathbf{E}(r) \) and \( \mathbf{H}(r) \) now read

\[
\text{div}(\mathbf{E}) = 0,
\] (2.15)

\[
\text{div}(\mathbf{H}) = 0,
\] (2.16)

\[
\text{curl}(\mathbf{E}) = i \omega \mu_0 \mathbf{H},
\] (2.17)

\[
\text{curl}(\mathbf{H}) = (-i \varepsilon \varepsilon_0 \omega + \sigma) \mathbf{E}.
\] (2.18)