

Chapter 1

Introduction

Abstract The topic of thin film optics/thin film optical spectroscopy can be tackled as a particular case of a broader class of phenomena, concerned with the interaction of electromagnetic irradiation with matter. A grammar description of this class of phenomena requires detailed analysis of the specific effects arising from material properties, as well as from the particular sample geometry. The analysis may be performed on both classical and quantum mechanical levels of description.

1.1 General Remarks

Whenever one is involved in spectroscopic experiments with electromagnetic waves, knowledge on the interaction of electromagnetic irradiation with matter is in the fundament of the theoretical understanding of the experimental results. This is true, for example, in molecular as well as in solid state optical spectroscopy. The light-with-matter interaction is the basis of numerous analytical measurement methods, which are applied in physics as well as in chemistry and biology. There is a tremendous amount of scientific publications and textbooks which deal with this subject. So what was the reason for writing this book?

The main reason was that in the present monograph the subject is described from the specific viewpoint of the *thin film spectroscopist*, and not from the viewpoint of general solid state or molecular optical spectroscopy. Caused by the specific geometry of a thin film sample, in thin film spectroscopy one needs a substantially modified mathematical description compared to the spectroscopy of other objects. The reason is that a thin film has a thickness that is usually in the nanometer- or micrometer region, while it may be considered to extend to infinity in the other two (lateral) dimensions. Of course, there also exist monographs on thin film optics (and particularly on optical coatings design). It is nevertheless the experience of the author that there appears to be a discrepancy between the typical reader's knowledge on the subject and the scientific level that is presumed to understand the highly specialized scientific literature. Moreover, the interaction of light with matter is

usually not taught as a separate university course. An interested student must therefore complete his knowledge referring to different courses or textbooks, such as those on general optics, classical continuum electrodynamics, quantum mechanics and solid state physics.

It is therefore the authors aim to provide the reader with a short and compact treatment of the interaction of light with matter (within an approach that is adapted to the specifics of thin solid films), and thus to bridge the gap between the readers basic knowledge on electrodynamics and quantum mechanics and the highly specialized literature on thin film optics and spectroscopy.

1.2 To the Content of the Book

In most practical cases, a thin film is built from a solid material. Therefore, the particular treatment in this book will mostly concern the specifics of the spectroscopy of solid matter. However, there appear situations where a general spectroscopic principle is easier to be explained referring to other states of matter. Inhomogeneous broadening of spectral lines is a typical example, as it is most easily explained in terms of the Doppler broadening as observed in gases. In such cases, we will happily leave the solid state specifics and turn to gases, in order to make the general principle more transparent.

Crystalline solids may be optically anisotropic. It is absolutely clear that a general and strong treatment of solid state spectroscopy must consider anisotropy. Nevertheless, in this book we will mostly restrict on optically isotropic materials. There are several reasons for this. First of all, many physical principles relevant in spectroscopy may be understood basing on the mathematically more simple treatment of isotropic materials. This is particularly true for many optical coatings, in fact, in optical coatings practice it is in most cases sufficient to work with isotropic layers models. There are exclusions from this rule, and in these situations anisotropy will be taken into account. This concerns, for example, the Giant Birefringent Optics (GBO) effects treated in connection with Fresnel's equations (Chap. 6). We will also refer to material anisotropy when discussing nonlinear optical effects at the end of this book (Chap. 13). By the way, the depolarization factors introduced in the first part of this book allow to a certain extent calculating the anisotropy in optical material constants as caused by the materials morphology (Chaps. 3 and 4). However, this book does definitely not deal with specifics of *wave propagation* in anisotropic materials.

Having clarified these general points, let us turn to the overall structure of this book. First of all it should be clear, that the reader is presumed to have certain knowledge on general optics, electrodynamics and quantum mechanics. It is not the purpose of this book to discuss the transversality of electromagnetic waves, nor to introduce the terms of linear or elliptical light polarization. The reader should be familiar with such kind of basic knowledge, as well as simple fundamentals of thermodynamics such as Boltzmann's and Maxwell's statistics.

Basing on this knowledge, the first part of the book (Chaps. 2–5) deals with the classical treatment of optical constants. In that classical treatment, both the electromagnetic field and the material systems will be described in terms of classical (non-quantum mechanical) models. Basing on Maxwell's equations, we will start with a rather formal introduction of optical constants and their frequency dependence (dispersion). We will have to introduce such important terms like the susceptibility, the polarizability, the dielectric function and the complex refractive index. We will then derive the main classical dispersion models (Debye-, Drude-, and the Lorentzian oscillator model). Starting from the Lorentz-Lorenz-formula, there will be a broad discussion of the optical properties of material mixtures. The first part of this book will be finished by the derivation of the Kramers-Kronig-relations for the dielectric function.

The second part (Chaps. 6–9) describes wave propagation in thin film systems. We start from Fresnel's equations for transmission and reflection at a single interface. This is an utmost important matter in thin film optics. For that reason, the discussion of these equations will fill up the full Chap. 6. In order to emphasize the physical value of these equations, we will derive a variety of optical and spectroscopic effects from them. Namely, this chapter will discuss Brewster's angle, total and attenuated total reflection of light, metallic reflection, propagating surface plasmon polaritons and the already mentioned GBO effects. In Chap. 7, the reader becomes familiar with the optical properties of thick slabs and single thin films. Chapter 8 deals with gradient index layers and film stacks; in particular, the matrix method for calculating transmittance and reflectance of an optical coating is introduced. In Chap. 9, some special cases are discussed, such as simple quarter-wave stacks, chirped mirrors, and the so-called grating waveguide structures.

The third part of the book (Chaps. 10–12) deals with the semiclassical treatment of optical constants. In this approach, the electromagnetic field is still described by Maxwell's equations, while the material system is described in terms of Schrödinger's equation. The goal is to obtain a semiclassical expression for the dielectric function, and consequently for the optical constants. Again, the reader is presumed to be familiar with basic knowledge on quantum mechanics and solid state physics, such as general properties of the wavefunction, simple models like the harmonic oscillator, perturbation theory, and Bloch waves. We start from the derivation of Einstein coefficients (Chap. 10). As a side effect of this derivation, we become familiar with quantum mechanical selection rules and Planck's formula for blackbody irradiation. By the way, we get the knowledge necessary to understand how a laser works. In Chap. 11, a density matrix approach will be presented to derive a general semiclassical expression for the dielectric polarizability of a quantum system with discrete energy levels. In Chap. 12, the derived apparatus will be generalized to the description of the optical constants of solids.

Finally, Chap. 13 (which forms the very short fourth part of the book) will deal with simple effects of nonlinear optics.

1.3 The General Problem

The basic problem we have to regard is the interaction of electromagnetic irradiation (light) with a specific kind of matter (a thin film system). In order to keep the treatment compact and “simple”, we will restrict our discussion to the electric dipole interaction. We will assume throughout this book, that among all terms in the multipole expansion of the electromagnetic field, the electric dipole contribution is the dominant one, and that other (higher order electric and all magnetic) terms may be neglected.

It is also worth emphasizing, that this book does definitely not deal with optical coatings design. It rather pursues the physical understanding of the information that may be drawn from a thin film spectrum as obtained from the experiment. We will therefore start from the experimental situation a thin film spectroscopist is confronted with.

In the frames of classical electrodynamics, any kind of light (which is used in optics) may be regarded as a superposition of electromagnetic waves. The idea of optical spectroscopy (or in more general optical characterization) is quite simple: If we have an object to be investigated (we will call it a *sample*), we have to bring it into interaction with electromagnetic waves (light). As the result of the interaction with the sample, certain properties of the light will be modified. The specific modification of the properties of electromagnetic waves resulting from the interaction with the sample shall give us information about the nature of the sample of interest.

For sufficiently low light intensities, the interaction process does not result in sample damage. Therefore, the majority of optical characterization techniques belong to the non-destructive analytical tools in materials science. This is one of the advantages of optical methods.

Although the main idea of optical characterization is quite simple, it may be an involved task to turn it into practice. In fact one has to solve two problems. The first one is of entirely experimental nature: The modifications in the light properties (which represent our *signal*) must be detected experimentally. For standard tasks, this part of the problem may be solved with the help of commercially available equipment. The second part is more closely related to modelling: From the signal (which may be simply a curve in a diagram) one has to conclude on concrete quantities characteristic for the sample. Despite of the researcher’s intuition and ability to identify or develop suitable models, this part may include severe computational efforts. Thus, the solution of the full problem requires the researcher to be skilled in experiment and theory (even mathematics) alike.

Let us now have a look at Fig. 1.1. Imagine the very simplest case—a monochromatic plane light wave impinging on a sample which is to be investigated. Due to the restriction on electric dipole interaction, we will only discuss the electric field of the light wave. In a complex notation, it may be written according to:

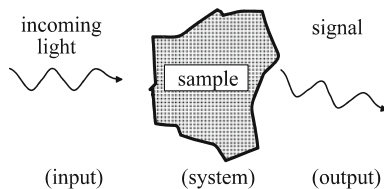


Fig. 1.1 Optical signal as the result of interaction of an electromagnetic wave with the sample

$$\mathbf{E} = \mathbf{E}(t, \mathbf{r}) = \mathbf{E}_0 e^{-i(\omega t - \mathbf{k} \cdot \mathbf{r})} \quad (1.1)$$

The parameters characterizing the incoming light (angular frequency ω , intensity (depends on the amplitude E_0), polarization of the light (direction of \mathbf{E}_0), propagation direction (direction of \mathbf{k})) are supposed to be known. Imagine further, that as the result of the interaction with the sample, we are able to detect an electromagnetic wave with modified properties. Which properties of the electromagnetic wave may have changed as the result of the interaction with the sample?

In principle, all of them may have changed. It is absolutely possible, that the interaction with the sample leads to changes in the frequency of the light. Typical examples are provided by Raman Scattering, or by several nonlinear optical processes. The polarization direction of the light may change as well. Ellipsometric techniques detect polarization changes and use them to judge the sample properties. Clearly, the light intensity may change (in most cases the light will be attenuated). This gives rise to numerous photometric methods analysing the sample properties basing on the measurement of intensity changes. And finally, anybody knows that the refraction of light may lead to changes in the propagation direction. Any refractometer makes use of this effect to determine the refractive index of a sample.

So we see, that the diversity of parameters characterizing electromagnetic radiation (in practice they are more than those mentioned here) may give rise to quite diverse optical characterization techniques.

We have now formulated our task: Starting from the analysis of certain parameters of the electromagnetic irradiation after having interacted with the sample, we want to obtain knowledge about the properties of the sample itself. Which kind of sample properties may be accessible to us?

Shortly spoken, the electromagnetic wave coming from the sample carries information about both the *sample material* and *sample geometry* (and the experimental geometry, but the latter is usually known to us). And if one is interested in the pure material properties, the geometrical influences on the signal have to be eliminated—experimentally or by calculations. In worse cases (and thin film spectra belong to these worse cases), geometrical and material informations are intermixed in the spectrum in a very complicated manner. In thin film systems, this is caused by the coherent superposition of electric fields arising from multiple internal reflections of light at the individual film interfaces. An experimental elimination of the geometrical sample contributions is then usually impossible, so that the

derivation of material properties often becomes interconnected with the instantaneous derivation of the geometrical properties by a corresponding mathematical treatment. As the result, we obtain information about both the sample material properties (for example the refractive index) and the geometry (for example the film thickness).

In order to make the theoretical treatment of thin film spectra more understandable, we will therefore develop the theory in two subsequent steps. The first step deals with the description of pure material parameters, such as the refractive index, the absorption coefficient, the static dielectric constant and so on. We will present several models that describe these parameters in different relevant physical systems.

The second step will be to solve Maxwell's equations in a system with given material parameters and a given geometry. In our particular case, we will do that for thin film systems. As the result, we obtain the electric field of the wave when it has left the system. Its properties will depend on the systems material *and* geometry. Having calculated the electric field, all the signal characteristics mentioned before may be theoretically derived. In the present book, the treatment will follow this philosophy.

In spectroscopy practice, one will proceed in a similar manner. The theoretical analysis of a measured spectrum starts from a hypothesis on the sample properties, including its material properties and geometry. Then, Maxwell's equations are solved, and the calculated characteristics are compared to the experimental values. From that, one may judge whether or not the assumptions previously made on the system were reasonable. If not, the assumed sample properties have to be altered, until a satisfying agreement between experiment and theory is achieved.

1.4 One Remark Concerning Conventions

Let us make an important remark concerning a convention implicitly made when writing down (1.1). Of course, the natural writing of the electric field in a monochromatic plane wave would operate with real functions and coefficients only. For such real fields, we could use a description of the type:

$$E_{real}(t, \mathbf{r}) = E_{0,real} \cos(\omega t - \mathbf{k}\mathbf{r} + \varphi) \quad (1.2)$$

However, the cosine function appears to be quite inconvenient with respect to our further mathematical treatment. On the other hand, it can be written as:

$$E_{real}(t, \mathbf{r}) = \frac{1}{2} \left[E_{0,real} e^{-i(\omega t - \mathbf{k}\mathbf{r})} e^{-i\varphi} + E_{0,real} e^{i(\omega t - \mathbf{k}\mathbf{r})} e^{i\varphi} \right] \equiv E_0 e^{-i(\omega t - \mathbf{k}\mathbf{r})} + c.c \quad (1.3)$$

Here “c.c.” denotes the conjugate complex to the preceding expression. It turns out, that the initially real electric field may be expressed as the sum of a complex field and its conjugate complex counterpart, while the latter does not contain any new physical information. Hereby, we have introduced the complex field amplitude E_0 as:

$$E_0 \equiv \frac{E_{0,real}e^{-i\varphi}}{2} \quad (1.4)$$

In practice, it appears much more convenient to build the further theory using complex electric fields $E(t, \mathbf{r})$ according to (1.3) and (1.4) instead of working with the real version (1.2). Therefore, in our treatment we make use of the complex field defined by (1.1), keeping in mind that the initially real field will be obtained when adding the complex conjugate to (1.1). Or, in other words:

$$E_{real}(t, \mathbf{r}) = 2\text{Re}E(t, \mathbf{r}) \quad (1.5)$$

where $E(t, \mathbf{r})$ is given by (1.1) and (1.4). But the choice of (1.1) for the complex writing of the electric field defines a particular convention, which is used throughout this book. When looking at (1.3), it becomes evident that we could have used the writing:

$$E_{real}(t, \mathbf{r}) = \frac{1}{2} \left[E_{0,real}e^{+i(\omega t - \mathbf{k}\mathbf{r})}e^{+i\varphi} + E_{0,real}e^{-i(\omega t - \mathbf{k}\mathbf{r})}e^{-i\varphi} \right] \equiv E_0e^{+i(\omega t - \mathbf{k}\mathbf{r})} + c.c.;$$

$$E_0 \equiv \frac{E_{0,real}e^{+i\varphi}}{2}$$

as well. It makes absolutely no physical difference whether, in (1.1), the plus or minus sign is chosen in the exponent. These are only two different conventions. But once we have decided on one of these conventions, we should strictly adhere to it in the following, in order to avoid convention confusion. In our particular treatment, we will use the minus-sign as fixed in (1.1). In other sources, the other convention may be used, which results in differences in the equations to be derived in the following.

Having clarified the general features of our approach, let us now turn to the introduction of the linear optical susceptibility.

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An Introduction

Stenzel, O.

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