Applied Surface Science xxx (2016) xxx–xxx

Contents lists available at ScienceDirect

Applied Surface Science

journal homepage: www.elsevier.com/locate/apsusc

Full length article

Experimental validation of the partial coherence model in spectroscopic ellipsometry and Mueller matrix polarimetry

M. Miranda-Medina[a,*], E. Garcia-Caurel[c], A. Peinado[c], M. Stchakovsky[d], K. Hingerl[b], R. Ossikovski[c]

[a] Austrian Center of Competence for Tribology – AC2T Research GmbH, Wiener Neustadt, Austria
[b] Center for Surface and Nanoanalytics, Johannes Kepler Universität Linz, 4040 Linz, Austria
[c] LPICM, CNRS, Ecole Polytechnique, Université Paris – Saclay, 91128 Palaiseau, France
[d] HORIBA Scientific, Avenue de la Vauve, 91120 Palaiseau, France

ARTICLE INFO

Article history:
Received 31 July 2016
Received in revised form
16 November 2016
Accepted 17 November 2016
Available online xxx

Keywords:
Mueller matrix polarimetry
Depolarization
Thick layers
Partial coherence

ABSTRACT

In this contribution, a recently advanced analytical approach for addressing partial coherence in spectroscopic polarimetric measurements is experimentally validated. The approach is based on the fundamental representation of the measurement process as the convolution of the polarimetric response of the sample and the instrumental function of the measurement system. Experimentally, the optical responses of two optically thick transparent layers were acquired by using spectroscopic Mueller matrix polarimetry at various angles of incidence over two spectral ranges (visible and infrared). The layers are considered isotropic and the loss of coherence is assumed to originate from the finite spectral resolution of the instrument. In parallel with the analytical approximation, the standard numerical approach implemented in commercial software was likewise used to reproduce the polarimetric responses. Excellent agreement between the analytical approximation, the commercial software one and the polarimetric measurements was found. The experimental validation of the analytical approximation represents a time-saving alternative to the numerical approaches used in commercial software and is of potential interest to real-time process monitoring by using spectroscopic ellipsometry or polarimetry.

1. Introduction

The interaction between electromagnetic radiation and matter is exploited by a variety of optical techniques to determine the physical properties of layered or bulk materials, interfaces, periodic structures or even more complex structures, such as biological specimens. More specifically, the polarization state of light is used as a probe in ellipsometry and polarimetry for the estimation of layer thicknesses and complex refractive indices. In the ideal case, ellipsometric and polarimetric systems operate under the assumption of totally polarized light; however, various sources of depolarization may arise during the experiment. The state of polarization of partially polarized and unpolarized light is described statistically and interpreted geometrically as the time average of points, representing totally polarized states, on the Poincaré sphere [1]. Features associated with the sample, such as thickness inhomogeneity, surface roughness or backside reflections of optically thick transparent layers can cause the depolarization of light. Thus, a thickness inhomogeneity of only ~2% in a 1-μm-thick film is sufficient to produce noticeable depolarization; the thickness nonuniformity in question is commonly modelled by implementing a thickness gradient [2]. Furthermore, finite spectral resolution, imperfectly collimated probing beam and spatially extended detector and/or light source are additional examples of depolarization sources attributable to the instrument as well as to the specific measurement configuration. The depolarization produced by the interaction of the instrument with the sample inherent to the measurement process is a consequence of the partial or total loss of coherence of the probing light traveling through the measured medium or system [3].

Depending on the amount of introduced depolarization, ellipsometry and polarimetry measurements can be seriously affected [4,5]. In transmission, birefringent slabs have been used to depolarize light as well as to illustrate the effect of the finite spectral resolution of the instrument on spectroscopic measurements [6]. In this case, the depolarization spectrum was modelled by superimposing incoherent multiple reflections. Similar models have been applied for describing depolarization in reflection configuration.

* Corresponding author.
E-mail address: maria.miranda@ac2t.at (M. Miranda-Medina).

http://dx.doi.org/10.1016/j.apsusc.2016.11.128
0169-4332/

Thin-film metrology of films deposited on transparent substrates requires the treatment of depolarization in order to obtain accurate results [7]. Additionally, the modeling of depolarization has been extended from isotropic [8] to anisotropic thick substrates by using the Mueller matrix formalism [9]. It was found that, despite the incoherent superposition of the reflected partial beams, the still present coherence between the *s*- and *p*- polarizations within a given beam in an anisotropic substrate can be used to determine the magnitude of anisotropy. For a generally anisotropic layer stack consisting of thin and thick layers, Mueller matrix spectra are modeled by a partially coherent summation of multiply reflected beams. A coherent summation of Jones vectors is used to model thin films, while thick films are characterized by Stokes vector formalism [10].

The common approach to modelling of spectra exhibiting depolarization typically combines stratified-medium electromagnetic modelling with numerical filtering to properly account for the observed partial coherence and depolarization. Recently, two of the authors advanced a novel, entirely analytical, general approach for addressing partial spectral coherence, based on the fundamental representation of the measurement process as the convolution (or the averaging) of the polarimetric response of the sample and the instrumental function of the measurement system [11]. The approach allows for handling the depolarization-inducing instrument parameters (finite spectral resolution, incidence angle spread, etc.) either separately or simultaneously.

The purpose of the present work is to demonstrate the validity of this methodology by applying it to experimental polarimetric data. Optically thick homogeneous transparent layers on 'semi-infinite' substrates are used as samples. A finite spectral resolution of the device is considered as the main source of depolarization. In addition to the comparison of the model to the experimental responses, the numerical solution of the fundamental convolution formulation of the polarimetric measurement is compared to the commonly used, numerical filtering based approach.

2. Experimental details

For the validation purposes, two structures consisting of optically thick (compared to the coherence length of the probing light) transparent layers on 'semi-infinite' substrates were used as samples. Sample 1 consisted of a silicon wafer coated with a thermally grown silicon dioxide layer of 1 μm thickness. This sample was measured in the visible range, from 1.2 to 2.8 eV, a spectral region within which the substrate is absorbing so that there was no undesired contribution from substrate backside reflections. The measurements were performed on a commercial spectroscopic Mueller matrix polarimeter *SmartSE* (HORIBA Scientific) operated in the visible to near-infrared spectral range (450–1000 nm with a nominal spectral step of 1.5 nm). The instrument uses ferroelectric liquid crystal retarders to generate and analyze the 16 independent polarization states needed to determine the full Mueller matrix of the sample. A more detailed description of the principle optical design of this polarimeter, including its operation and calibration methods, can be found in Ref. [12]. All measurements were done in reflection at the angle of incidence of 60°.

Sample 2 consisted of a boron-doped silicon wafer coated with a 35-μm-thick resin layer. This sample was measured in the infrared range, from 0.44 to 0.62 eV, which corresponds to a spectral window free of vibrational absorption bands of the resin. The backside of the substrate was roughened to ensure that backside reflection contribution could be effectively neglected. The infrared measurements were performed on a home-made spectroscopic Mueller matrix polarimeter installed on the infrared beamline (SMIS) at the French synchrotron facility SOLEIL. The infrared polarimeter features a full spectral range from 0.08 to 0.62 eV and employs achromatic optical components guaranteeing highly accurate measurements over a wide spectral range. All measurements were performed in reflection configuration at the angle of incidence of 69.1°. More information about the design, calibration and technical performances of the infrared Mueller ellipsometer can be found in Ref. [13].

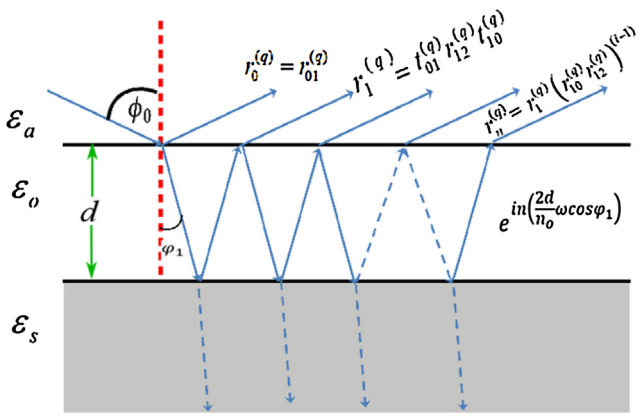

Fig. 1. Multiple-beam model of light reflection from the ambient (dielectric function ε_a) onto a thick layer (ε_o) lying upon a semi-infinite substrate (ε_s). The superscript (q) denotes either *p*- or *s*-polarization.

Experimental data were filtered out under Cloude decomposition method [14] in order to ensure the physical realizability of the Mueller matrices.

3. Description of the analytical approximation

Consider a sample with a structure as that shown in Fig. 1 where an optically thick transparent layer is lying on top of a semi-infinite substrate. If the sample consists of a thick substrate alone, then the latter is depicted by the layer whereas the substrate beneath becomes the ambient. Both the layer and the substrate are assumed isotropic. With all dielectric functions known, the Fresnel reflectances of the 0th (specularly reflected), 1st (bouncing once forth and back through the layer) 2nd, 3rd, and i^{th} (bouncing *i* times forth and back through the layer) partial beam are calculated as is shown in Fig. 1. From this picture, the Jones matrix elements of the optical structure, representing the overall reflectances for each combination of polarizations, can be readily obtained as sums of infinite geometric series with terms constituted by the reflectances of the partial beams.

The multiple passage of the probing light through the layer (i.e., the bouncing back and forth) retards the beam and lets the partial beams acquire a phase shift multiple of the factor

$$\alpha = \frac{2d\omega}{c}\sqrt{\varepsilon_o - \varepsilon_a \sin^2\phi_0} \tag{1}$$

Here, the symbol d corresponds to the thickness of the layer, ω is the cyclic frequency, c the velocity of light in vacuum, ϕ_0 is the incident angle, ε_o and ε_a are the refractive indices of the thick layer and the ambient, respectively. The retardation described by Eq. (1) is affected by the loss of coherence and leads to depolarization effects. If the finite spectral resolution of the measurement instrument is assumed as the main source of depolarization, the analytical approximation advanced in Ref. [11] shows that the average second-order moments of the Jones matrix elements describing the sample are given by

$$\langle J^{(i)} J^{(j)*} \rangle = \int J^{(i)}\left(\omega'\right) J^{(j)*}\left(\omega'\right) w\left(\omega - \omega'; \Delta\omega\right) d\omega' \tag{2}$$

Please cite this article in press as: M. Miranda-Medina, et al., Experimental validation of the partial coherence model in spectroscopic ellipsometry and Mueller matrix polarimetry, Appl. Surf. Sci. (2016), http://dx.doi.org/10.1016/j.apsusc.2016.11.128