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Infrared ellipsometry of nanometric anisotropic dielectric layers on absorbing materials

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HIGHLIGHTS

- The inversion problem of infrared ellipsometry is resolved on the basis of a fresh mathematical approach.
- The novel method possesses very high sensitivity because it is founded only on the phase conversion measurements.
- The method is successfully applicable for nanometric layers in the infrared spectral region.

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ABSTRACT

An inversion problem of infrared ellipsometry is resolved on the basis of a fresh mathematical approach, which does not use the traditional regression analysis for data handling and has no need of initial guesses for the desired parameters. It is shown that obtained simple analytical equations for ellipsometric quantities open up new possibilities for determining optical parameters of an anisotropic ultrathin layer. The novel method possesses very high sensitivity because it is based on the phase conversion measurements of polarized reflected light. The method is tested using a numerical simulation and the results demonstrate clearly that it is successfully applicable for nanometric layers in the infrared spectral region.

1. Introduction

An appropriate technique for determining the parameters of ultrathin films (thickness d much less than radiation wavelength λ) is, obviously, the ellipsometric method [1–7]. This is because ellipsometry is characterized by high sensitivity, is nondestructive, noninvasive, and can successfully be performed in real-time for investigating the dynamics of surfaces and nanostructures. The ellipsometric method is now well established in a fairly wide spectral region – from vacuum UV to far-IR. The bands that appear in an infrared spectrum contain the highly individual fingerprint of a material and, in consequence, infrared ellipsometry can also be employed to characterize surface layers in the low nanometer range, i.e. thinner than 1/1000th of the wavelength [8–16].

However, ellipsometry is an indirect method, i.e. the desired parameters must be inferred from measured ellipsometric characteristics by solution of an inversion problem, for which, as a rule, the widely accepted model-based regression analysis is used. Unfortunately, in the case of ultrathin films, the model-based regression analysis, where unknown parameters result from fitting the experimental data for ellipsometric quantities (prevalently frequency spectrum) to the computed values of these quantities on the basis of a presumed optical model of the structure under study, is characterized by a strong correlation between optical constants and film thickness, i.e. by this method it is not possible to determine the thickness and dielectric constants of an ultrathin film simultaneously [2].

Because of this, in the case of an ultrathin film it is appropriate to separate the contributions of an ultrathin film to the ellipsometric angles Δ and Ψ from the corresponding contributions Δ_0 and Ψ_0 of the bare substrate, i.e. to use the differential quantities $\delta\Delta = \Delta - \Delta_0$ and $\delta\Psi = \Psi - \Psi_0$ for determining the parameters of an ultrathin film. The quantities $\delta\Delta$ and $\delta\Psi$ are expressible analytically as a power series in the small parameter d/λ. Such an approach gives us relatively simple analytical equations which offer a clearer view of how the measured ellipsometric quantities depend on the incident angle and unknown film parameters, which in turn makes it possible to analyze the equations in the light of the resolution of the inverse problem by multi-angle measurements, which produce the effect of enhancing information content.

However, in doing so, it is very important to pay attention to the precision of ellipsometric measurements. If $\delta\Psi$ or $\delta\Delta$ becomes smaller than experimental error, then the use of this quantity in

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data processing fails. Generally, it has been known that ellipsometric angle $\delta\Psi$ is unsuitable for treatment of nanometric films in the infrared spectral range because nowadays a properly aligned ellipsometer with high-quality optics is capable of precision only of about several hundredths of degrees in Ψ and Δ. For clarity, in this paper the corresponding calculations are also presented for anisotropic films (Section 2) and it is shown that $\delta\Psi$ is that quantity which at first becomes smaller than experimental error if $d/\lambda < 10^{-3}$. Therefore, in this case, we can use only one measurable quantity, i.e. Δ. Naturally, this brings up the question: What parameters of ultrathin films can we determine solely on the basis of Δ? In general, one evident advantage of ellipsometry, by comparison with intensity-based measurements, definitely lines in the fact that it involves simultaneous measurement of two independent quantities that contain much more information about a sample than just the one measurable quantity in photometry. For example, in a recently published paper [17], it was shown that by applying a new mathematical approach for solving the inverse problem it is possible on the basis of two ellipsometric parameters ($\delta\Psi$ and $\delta\Delta$) to determine the thickness and all optical constants of a uniaxially anisotropic ultrathin layer on an absorbing isotropic substrate simultaneously, i.e. without the usual correlation between the thickness and optical constants. However, in light of the issues mentioned above, this method is no longer valid in the case of a nanometric film in the infrared spectral range, where $\delta\Psi$ is still not attainable by present-day ellipsometric techniques.

A purpose of this paper is to analyze the inverse issue for uniaxially anisotropic ultrathin films on isotropic absorbing substrates by a technique which is based only on the phase conversion measurements, i.e. by using nothing else but Δ.

The paper is organized as follows. In Section 2, the formulas for ellipsometric angles of uniaxially anisotropic transparent ultrathin films on absorbing isotropic substrates are derived. The third section is concerned with the solution of the ellipsometric inverse problem for such films.

2. Ellipsometric angles for ultrathin films

Let an ultrathin ($d << \lambda$) uniaxially anisotropic non-absorbing dielectric film be located upon a semi-infinite isotropic homogeneous and absorbing substrate with complex dielectric constant $\hat{\varepsilon}_s = \varepsilon_{sR} + i\varepsilon_{sI} \equiv \hat{n}_s^2 = (n_{sR} + in_{sI})^2$. The dielectric tensor for a uniaxially anisotropic transparent material in the $x\,y\,z$ coordinate system is given by

$$\begin{bmatrix} \varepsilon_{11} & \varepsilon_{12} & \varepsilon_{13} \\ \varepsilon_{21} & \varepsilon_{22} & \varepsilon_{23} \\ \varepsilon_{31} & \varepsilon_{32} & \varepsilon_{33} \end{bmatrix} = \mathbf{A} \begin{bmatrix} \varepsilon_o & 0 & 0 \\ 0 & \varepsilon_o & 0 \\ 0 & 0 & \varepsilon_e \end{bmatrix} \mathbf{A}^{-1}, \tag{1}$$

where $\mathbf{A}$ is the coordinate rotation matrix [18]. Therefore,

$$\varepsilon_{11} = \varepsilon_o \cos^2\varphi + (\varepsilon_o \cos^2\theta + \varepsilon_e \sin^2\theta)\sin^2\varphi, \tag{2}$$

$$\varepsilon_{12} = \varepsilon_{21} = (\varepsilon_o - \varepsilon_e)\sin^2\theta \sin\varphi\cos\phi, \tag{3}$$

$$\varepsilon_{22} = \varepsilon_o \sin^2\varphi + (\varepsilon_o \cos^2\theta + \varepsilon_e \sin^2\theta)\cos^2\varphi, \tag{4}$$

$$\varepsilon_{13} = \varepsilon_{31} = (\varepsilon_e - \varepsilon_o)\sin\theta\cos\theta\sin\varphi, \tag{5}$$

$$\varepsilon_{23} = \varepsilon_{32} = (\varepsilon_o - \varepsilon_e)\sin\theta\cos\theta\cos\varphi, \tag{6}$$

$$\varepsilon_{33} = \varepsilon_o \sin^2\theta + \varepsilon_e \cos^2\theta, \tag{7}$$

where $\varepsilon_o \equiv n_o^2$ and $\varepsilon_e \equiv n_e^2$ are the real principal dielectric-tensor components in the crystal-coordinate system, and θ and φ are the Euler angles with respect to a fixed $x\,y\,z$ coordinate system (the Cartesian laboratory coordinate system). The laboratory x, y, and z axes are defined as follows. The reflecting surface is the xy plane, and the plane of incidence is the zx plane, with the z axis normal to the surface of the layered medium and directed into it. The incident linearly polarized time-harmonic (the complex representation is taken in the form $\exp(-i\omega t)$, where $\omega = 2\pi c/\lambda$, and λ is a vacuum wavelength) electromagnetic plane wave in a transparent ambient medium with isotropic and homogeneous dielectric constant $\varepsilon_a \equiv n_a^2$ makes an angle ϕ_a with the z axis. Assume that all the media are nonmagnetic.

Since in anisotropic systems the Jones matrix contains off-diagonal terms, a so-called generalized ellipsometry [4,19] is designed for anisotropic systems. There are three normalized complex ratios, which may be chosen for the measurement in the reflection mode ellipsometry, for example,

$$r_{pp}/r_{ss} \equiv \tan\Psi_{pp}\exp(i\Delta_{pp}), \tag{8}$$

$$r_{ps}/r_{ss} \equiv \tan\Psi_{ps}\exp(i\Delta_{ps}), \tag{9}$$

$$r_{sp}/r_{ss} \equiv \tan\Psi_{sp}\exp(i\Delta_{sp}), \tag{10}$$

where the first subscript indicates the incident light.

In the following, we examine the major practical issue, which arises in connection with ellipsometric quantities $\delta\Delta$ and $\delta\Psi$. Namely, on the one hand, it is apparent that these quantities are rather small when $d/\lambda \ll 1$. On the other hand, ellipsometers determine ellipsometric angles always with a certain precision. This brings up the question: What ellipsometric angles are at all measurable for nanometric films?

Figs. 1–3 show the differential ellipsometric angles $\delta\Delta_{pp} = \Delta_{pp} - \Delta_0$, $\delta\Psi_{pp} = \Psi_{pp} - \Psi_0$, Δ_σ, and Ψ_σ (Δ_0 and Ψ_0 are the ellipsometric angles for the bare substrate; $\Psi_\sigma = \Delta_\sigma = 0$ if $d = 0$; $\sigma = ps$ or sp) as functions of d/λ for some different values of refractive indexes of anisotropic films and substrates. The ellipsometric angles $\delta\Delta_{pp}$, $\delta\Psi_{pp}$, Δ_σ, and Ψ_σ in Figs. 1–3 are computed by the rigorous electromagnetic theory for anisotropic layered systems on the basis of Eqs. (8)–(10) (the relevant computational technique is outlined, e.g., in [20]). Note that the ambient refractive index $n_a = 1$ in Figs. 1–9. The machine-performed computations demonstrate that for ultrathin films with $d/\lambda < 10^{-3}$ the measurable ellipsometric quantities are only $\delta\Delta_{pp}$ and Δ_σ, the others ($\delta\Psi_{pp}$ and Ψ_σ) are smaller than the measurement error (currently front-ranking ellipsometers are capable of precision of about several hundredths of degrees in Δ and Ψ). In other words, in this case, the ellipsometric angles which are bound up with the phase changes in the reflection process are chiefly applicable for ultrathin films.

Further, for the contribution of an anisotropic ultrathin layer to ellipsometric angle Δ_{pp}, i.e. for the quantity $\delta\Delta_{pp} = \Delta_{pp} - \Delta_0$ one can obtain the following approximate formula [20]:

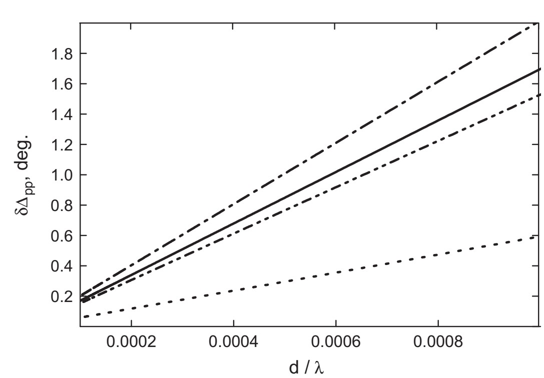

Fig. 1. Differential ellipsometric angle, $\delta\Delta_{pp} = \Delta_{pp} - \Delta_0$, as a function of d/λ for an anisotropic film with $n_o = 1.7$, $n_e = 1.8$, and $\theta = \varphi = 40°$ on an absorbing isotropic substrate with $\hat{n}_s = 1.5 + i0.6$ at $\phi_a = 50°$ (dotted curve) and for an anisotropic film with $n_o = 2.8$, $n_e = 2.6$, $\theta = 60°$, and $\varphi = 30°$ on different substrates with $\hat{n}_s = 3.9 + i0.5$ (solid curve), $\hat{n}_s = 2.5 + i0.1$ (dash-dotted curve), and $\hat{n}_s = 0.3 + i4$ (dash-dot-dotted curve) at $\phi_a = 70°$.