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Numerical ellipsometry: High accuracy modeling of thin absorbing films in the *n*–*k* plane

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ABSTRACT

A major challenge for those utilizing ellipsometry is numerical processing of the measured data. Our recent work shows how the transcendental, multivalued equations arising from the physics of reflection from layers can be solved in the *n*–*k* plane. This approach applies the mathematics of Complex Analysis to solve the equations numerically. The work presented extends the *n*–*k* method to obtain solutions within the accuracy limit of each measurement. The system treated here is that of a thin absorbing film (chromium) overlying a known substrate (silicon). Solutions for a three-layer model of the chromium film including film–substrate and film–air interfacial layers result in a mean square error (MSE) on the order of 0.01, a significant improvement over a single-layer model. Relaxing the constraint of vertical homogeneity provides a six-layer model with the same interfacial layers and four sublayers of chromium. The chromium layers have near-identical values of optical properties and an MSE of essentially zero (10^{-13}). It is anticipated that additional methods will be needed for other classes of problems.

1. Introduction

Analysis of ellipsometry data almost always requires computerized numerical methods because of the nature of the equations arising in the physics of light reflection from laminar surfaces [1]. Data acquisition can be very similar regardless of the configuration of the reflecting surface. On the other hand, data analysis may differ markedly depending upon the thickness, optical properties, and morphology of layers composing the reflecting surface. Consequently data analysis methods will be different for each class of problem.

The problem of solving the ellipsometry equations is difficult and can be improved. Prior to the early 1980s ellipsometry solutions were obtained using a book of tables, calculators, or a Fortran program [2–4]. These methods led to the use of least-squares, best-match numerical methods (Levenburg–Marquardt) implemented on digital personal computers [5]. Least squares methods have been useful for ellipsometry problems which are not adversely affected by correlation problems. On the other hand, errors introduced by the matching process can become very large as in the case of thin absorbing films.

The authors have been developing and publishing underlying mathematical and numerical methods for the modeling and solving processes. The requirement is that the number of independent equations equal or exceed the number of unknowns at each solution, commonly one per wavelength. The methods pose the mathematical problems to make it possible to solve the ellipsometry equations to the full limit of the accuracy of double precision numbers represented upon a digital computer, 10^{-13} typically. An important benefit of this is that the full extent of the instrument accuracy is made available for model improvement. How the overall solution accuracy is reduced by the effects of measurement error and model imperfection is more fully described in a prior publication [6].

Here we demonstrate a method which extends a single-layer model for chromium films on silicon to a much more accurate three-layer model with a mean square error (MSE) of 0.0105. The single-layer model has been examined earlier by the present authors using *n*–*k* plane methods [6–10] and by others using best-match [11]. The single-layer model used in both the earlier work of the present authors and those cited above were acknowledged as being a simplification of a real Cr film which was not atomically flat and which had been exposed to room air. Thus it is not surprising that the MSE could be improved by the addition of interface layers. A primary purpose of the work presented here is to describe a method for including additional layers to further develop the model. All MSE values were computed identically for constant thickness by making use of the standard deviations, σ, at each measurement to underweight those with wider variances as shown in the following, commonly-used equation.

$$\mathrm{MSE} = \sqrt{\frac{1}{2N-M}\sum_{i=1}^{N}\left[\left(\frac{\Psi_i^{\mathrm{mod}}-\Psi_i^{\mathrm{exp}}}{\sigma_{\Psi_i}^{\mathrm{exp}}}\right)^2+\left(\frac{\Delta_i^{\mathrm{mod}}-\Delta_i^{\mathrm{exp}}}{\sigma_{\Delta_i}^{\mathrm{exp}}}\right)^2\right]} \quad (1)$$

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where N is the number of measurement (Ψ, Δ) pairs, M is the number of variable parameters in the model, and σ is the variance at each measurement.

The number of degrees of freedom varied between the different models and methods. Many different methods were explored by Hilfiker et al. who did not specifically publish the degrees of freedom for each case but which, for the point-by-point solutions, clearly ranged from close to the number of measurements to perhaps half of the number of measurements depending upon the sample configuration and number of samples used for a given computation. On the other hand the number of degrees of freedom may have been as few as 10 for the work with oscillators. The work reported here had a similar number of degrees of freedom as the point-by-point work just mentioned, also depending on the specific computations. By any method, the single-layer model resulted in an inferior MSE compared to the three or more layer models because these more detailed and accurate multilayer models brought about a much closer match between measured and computed values of Ψ and Δ which in turn significantly brought down the size of the numerator terms in Eq. (1).

It must be emphasized that once solutions are found, solution testing may be readily carried out by any number of methods including any of the properly-working commercial software packages. The method described here does not require initial estimates. Problems with local minima do not occur and the numerical processes do not diverge. Note that while the solutions here are exclusively obtained using computerized numerical methods, they may be better understood when visualized graphically. These methods are now finding applications including the present paper on thin absorbing films, transparent conducting oxide films, and a basic physics study of optical absorption of lanthanum chromate films [12,13].

Hilfiker et al. have kindly shared their measured data to allow the authors to compare solution methods. The method presented here examines solutions at each individual wavelength within the set of measurements. At each wavelength there can be a very large, one or more orders of magnitude, variation in the effect of measurement accuracy upon the solution accuracy which depends on the underlying mathematics of the model. It makes little sense to solve, by any means, at wavelengths for which the underlying mathematics simply cannot provide accurate solutions and then to weight these solutions in the same way as those obtained at wavelengths for which the accuracy is better by an order of magnitude or more. In this type of approach, although only ellipsometric data at two wavelengths is required, spectroscopic measurements mainly are necessary to allow identification of the most appropriate wavelengths for analysis.

2. Theory

Spectroscopic ellipsometry and its mathematical formalism are well described elsewhere [1]. In general, light undergoes a change in polarization state upon reflection from a surface. The quantitative change in polarization state provides information pertaining to the reflecting surface. Computation of reflecting surface parameters presents a significant challenge normally requiring numerical methods to find solutions.

The surface configuration considered here is that of substrates with overlying films in which the media are uniform, homogeneous and isotropic with flat interfaces. The mathematics for multiple layers in the n–k plane analysis method is fully described in a previous publication [6]. Because the measurements were made on films of different thicknesses, the twisted curves in three-dimensional (n–k–d) space do not intersect; however, their projections onto the n–k plane do intersect if the optical properties n and k do not vary significantly with film thickness. These projections form a vertex at the film optical properties n and k [6]. The work here examines the vertices formed at each measured wavelength to take advantage of the fact that for ex-situ measurements the thickness of a film is a constant, a well-known solution constraint.

Descriptions of the way in which a number of theorems of Complex Analysis have been applied to the ellipsometry problem appear in previous publications, the details of which are not repeated here [6–10]. We shall identify individual solution curves by the planes upon which they pass through the point at infinity. All of the solution curves in the present work originated on the (0, −) log and root planes.

Solution curves may terminate only at singularities. The "S" type singularity is located at

$$\mathrm{S} = n_0 \sin(\alpha_0) \tag{2}$$

which corresponds to the critical value of film optical properties for total external reflection in which n_0 is the air optical index and α_0 is the angle of light incidence in the air ambient. Curves also may terminate at a "P" type singularity located at the "pseudo refractive index" of the reflecting surface

$$\mathrm{P} = n_0 \sin(\alpha_0)\sqrt{\tan^2(\alpha_0)\left(\frac{\rho-1}{\rho+1}\right)^2+1} \tag{3}$$

in which ρ is the measured complex parameter, $\rho = \tan(\Psi)e^{i\Delta}$. The physical interpretation of Eq. (2) is that it provides the optical properties of an infinite thickness (i.e. bulk) material which corresponds to the measured values of Ψ and Δ assuming homogeneous, isotropic bulk material with no surface roughness. Numerical solutions were calculated independently at each measured wavelength using a single incidence angle and thus problems with overdetermination of data were not encountered.

3. Modeling

All of the computer processes described were carried out using Matlab© GUI environments. Three measurements on three films of different thicknesses at a single wavelength define an "experiment." The fourth measurement was not used until later. The first step is to select the simplest model of the reflecting surface (single-layer) and to find the three intersections in the n–k plane for each experiment. At each intersection the associated values of n, k, and d (two values of the thickness, one for each of the intersecting solution curves) are recorded and plotted together with all of the error statistics for these values. The methods for computing error statistics are described fully in a prior publication so are not repeated here [6]. If the thicknesses at each solution are constant across wavelength then this simplest model is confirmed. For the data under examination here the simplest model was not confirmed and thus a more detailed model was required.

This more detailed, three-layer model includes both and an overlayer and an underlayer. In this case we choose to treat each as a thin effective medium approximation (EMA) layer composed of 50% of the material above it and 50% of the material below it. Clearly some material science expertise enters at this point and other choices may be examined. Solving the three-layer problem results in significantly improved agreement in the thickness as a function of wavelength. Optimal overlayer and underlayer thicknesses are obtained by computing the standard deviation of all the thicknesses for all experiments and summing them for a grid search of thicknesses. In principle, additional parameters could be varied. In the case presented in this work, both the underlayer and overlayer were significant in the solution and therefore were required. Intersections are found for each experiment considering the three-layer model just described including the error statistics.

Finally a solution for a six-layer model is presented. The model includes the same interfacial layers as the three-layer model but treats the chromium as four separate layers determined by each of the four measurements given the thicknesses determined for the three-layer model plus an estimated thickness for the fourth measurement using the optical properties of the three-layer model in a limited wavelength range described below.