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Survey of methods to characterize thin absorbing films with Spectroscopic Ellipsometry

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Abstract

Spectroscopic Ellipsometry (SE) is routinely used to measure thickness and optical constants of dielectric and semiconductor films. However, unique results for thin absorbing films, such as metals, are difficult to ensure. SE enables simultaneous determination of metal layer thickness and optical constants through several methods. These include multiple sample analysis, interference enhancement, optical constant parameterization, and simultaneous analysis of SE and intensity-based optical measurements. These methods are comparatively tested on a series of Chromium thin films prepared with multiple thicknesses and multiple substrate types. Thickness and optical constants for the thin Cr layers are determined, but sensitivity depends on the choice of method and its implementation.

Of the abovementioned methods, interference enhancement is demonstrated to increase sensitivity to Cr film properties significantly. This method requires a thick dielectric between the Cr film and Si substrate to enhance the information content of multiple angle measurements. The combination of SE and transmitted or reflected intensity is also shown to produce a unique result for the Cr films on fused silica substrates. Multi-sample analysis can increase the volume of measured information by providing multiple path lengths through the absorbing material. In addition, we demonstrate that multi-sample analysis is also effective with same Cr thickness under the condition that the overall light interaction is modified. Commonly, for multi-sample analysis, the optical constants of each film should be identical. This assumption is not necessarily valid for metal films, such as Cr, but the principle is demonstrated. Finally, optical parameterization is used to reduce the number of unknown model parameters. This reduces correlation while maintaining smooth, continuous, and often Kramers–Kronig consistent optical properties for the absorbing material. Optical parameterization is best utilized in combination with other methods to further improve model sensitivity. The advantages and limitations of each method are discussed.
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1. Introduction

Optical measurements, such as Spectroscopic Ellipsometry (SE) or Reflectance and Transmittance (*R*/*T*), are routinely used to measure thickness and optical constants of transparent and semi-transparent thin films. However, absorbing layers are often avoided due to two limiting factors: a) light does not penetrate absorbing films thicker than ~100 nm, and b) strong correlation between thickness and optical constants for absorbing layers may prevent a unique solution.

The first limitation is offset by the fact that many recent applications employ thinner absorbing layers. Such applications include graded metal alloys for advanced phase-shifting photomasks, thin silver layers in optical coating stacks for solar selectivity, and cermet films that mix metal and dielectric particles. The data storage industry uses thin absorbing films for

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applications ranging from magnetic DRAM to optical storage disks. TiN films are used as conductive diffusion barriers in copper-based semiconductor devices. Thin electrochromic and photochromic layers have an opaque and transparent state. This list of applications of sub-100 nm films continues to grow.

While optical measurements may not be ideally-suited for characterization of thin absorbing layers, they offer a few distinct advantages. Optical measurements are non-destructive, non-invasive, and can be performed in real-time. Thus, they are ideal for many in-line or in situ applications where film thickness and optical constants need to be determined quickly and precisely. This leads to the second limitation of optical methods — strong correlation between thickness and optical constants for very thin and absorbing films. The degree of correlation depends on characterization method and sample structure. Several methods can reduce correlation and ensure sensitive measurements for thin absorbing films. These include multiple sample analysis, interference enhancement, optical constant parameterization, and simultaneous analysis of SE and intensity-based optical measurements. Many of these methods were originally devised for single-wavelength characterization over three decades ago. Aspnes detailed many of these earlier references [1] and McGahan et al. provided a good overview of methods [2]. However, these techniques are still not commonly applied and deserve a fresh review.

The goal of this work is to demonstrate these methods and compare their advantages and limitations. The methods are applied to the characterization of a series of Cr thin films.

2. Spectroscopic Ellipsometry

Spectroscopic Ellipsometry (SE) measures the change of polarization in light that is reflected or transmitted from a sample. The polarization change is described by an amplitude ratio, tan(Ψ), and phase difference, Δ, between light oriented in the p- and s- directions relative to the sample surface:

$$\rho = \tan(\Psi)e^{i\Delta} = \frac{R_p}{R_s}, \tag{1}$$

where R_p and R_s represent the complex Fresnel coefficients for p- and s- polarizations, respectively. Ellipsometry measurements are used to determine optical constants and film thickness both for single- and multi-layer coatings. Many references discuss ellipsometry in further detail [3–6]. Spectrophotometric (R/T) techniques measure the change in light intensity reflected from or transmitted through a sample. While intensity measurements are more intuitive than polarization change, ellipsometry measurements offer a few distinct advantages: i) more information content — measurements capture both amplitude and phase, ii) greater sensitivity to ultrathin transparent layers less than 10 nm thick, and iii) more precise measurements. From a utilitarian point of view, SE and R/T measurements are complementary and when used together can often enhance the total information obtainable from a sample.

SE and R/T are not direct measurements of film thickness and optical constants. Instead, the sample structure must be inferred from polarization or intensity change versus wavelength and angle. To find the sample structure whose optical response best matches experimental measurements, regression analysis is used.

Optical measurements are often subject to the inverse problem, where the ‘result’ is measured but the ‘cause’ must be guessed. The ‘cause’ is the sample in question and it is depicted in a model which describes each material layer interacting with the measurement light. Optical responses (Ψ, Δ, R, T) from any model can be generated. Model-generated data can then be compared to experimental data while the sample properties (thickness, optical constants) are varied. Through regression analysis, unknown sample properties (“fit” parameters) whose response best matches the experimental data are found — this process is also referred to as “fitting” the experimental data.

The final model should be examined for accuracy, sensitivity and uniqueness. If too many “fit” parameters are varied, the solution may be under-determined, and the final model may not be unique. Choosing the correct method can make the difference between an under-determined solution and obtaining sufficient information to determine all “fit” parameters of an absorbing film.

2.1. Interpreting results

Several indicators are used to estimate sensitivity and uniqueness of the final model. These include the mean squared error, figure of merit (or 90% confidence limits), two-parameter correlation, and uniqueness testing. Jellison reviews these indicators in his chapter on data analysis [7]. Our implementation of regression analysis is detailed by Herzinger et al. [8], but reviewed briefly herein.

The first estimate of fit quality is a comparison between the model-generated curves and experimental data. The shape and structure of measured data curves indicate details of the materials and the film thickness. The final model accuracy depends first on whether it matches details of the experimental data curve. The mean squared error (MSE) quantifies the difference between model and experiment for all regressed quantities. The random measurement error for each data point is incorporated via the following biased MSE equation [8]:

$$\mathrm{MSE}^2 = \frac{1}{2N-M}\sum_{i=1}^{N}\left[\left(\frac{\psi_i^{\mathrm{mod}}-\psi_i^{\mathrm{exp}}}{\sigma_{\psi,i}^{\mathrm{exp}}}\right)^2+\left(\frac{\Delta_i^{\mathrm{mod}}-\Delta_i^{\mathrm{exp}}}{\sigma_{\Delta,i}^{\mathrm{exp}}}\right)^2\right] = \frac{1}{2N-M}\chi 2, \tag{2}$$

where $\sigma_{\Psi,\Delta}^{\mathrm{exp}}$ are the measurement error bars. The pre-factor normalizes the function so MSE is not directly affected by change in the number of wavelengths (N is the number of Ψ,Δ pairs) or number of “fit” parameters (M).

A good MSE is a necessary, but not sufficient judgment of the correct model. Correlation between fit parameters will often indicate that the model is not unique. The two-parameter correlation coefficients indicate the independence of any two fit parameters. If correlation does occur, the model can often be simplified by reducing the number of “fit” parameters. However, strong correlation between thickness and optical

constants (or variables that describe the optical functions) may indicate that a unique answer is not possible within the constraints of the current experiment.

Whenever correlation exists, the final result is called into question. To examine sensitivity and uniqueness of the final model, it is useful to adjust the parameter in question to a fixed value while refitting all other parameters. If similar "fit" quality is achieved, the fit is insensitive to this particular parameter, at least within the range it was adjusted.

A Uniqueness Test is a simulation of fit results for a model when one fit parameter is varied. The test parameter is fixed at a series of values while all remaining fit parameters are adjusted to find the best MSE. For example, a fit of optical constants and thickness may have a minimum MSE when the thickness is near 56.4 nm, as shown in Fig. 1. The Uniqueness Test would force the thickness to different values over a defined range. In this case, the thickness is fixed at different values between 55.5 nm and 57.5 nm. The optical constants are allowed to vary to find the best MSE value for each thickness that is tested. The MSE can then be graphed versus the tested fit parameter. In Fig. 1, the MSE has been normalized relative to the minimum MSE (from the best fit) for a convenient visual comparison. It is also convenient to record the parameter range that maintains MSE results within 10% (or any arbitrary percentage) of the minimum value. This parameter range will be referred to as the Uniqueness Range. As shown in Fig. 1, the Uniqueness Range is calculated as the difference between parameter values above and below the best fit result that provide MSE results 10% higher than the global MSE minimum.

Finally, all results (thickness and optical constants) must be consistent with other knowledge of the sample. A good test of final optical constants is to verify that they remain Kramers–Kronig (K–K) consistent [3]. This physical law connecting the real and imaginary dielectric functions provides a basis for "reasonable" shape that the final optical constants must adhere to. This test can become an inherent requirement if the model uses a K–K consistent dispersion equation (Section 3.3).

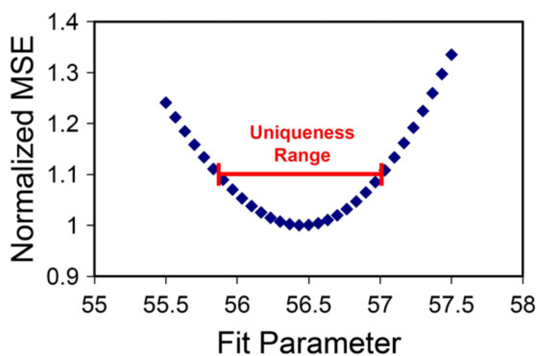


Fig. 1. Uniqueness Test of a fit parameter varies that parameter over a range of values, while remaining fit parameters are regressed to find the best match to experimental data. The MSE versus the varied fit parameter is graphed. Normalizing to the lowest MSE value allows convenient calculation of fit parameter range that provides values within 10% of the minimum MSE (referred to as the Uniqueness Range).

3. Characterization of absorbing films

Optical measurements require model-based analysis to determine the unknown sample properties. To find a unique solution, it is imperative that the measurement contains adequate information content to uniquely determine all unknown sample properties. For a single isotropic layer on known substrate, the three unknown parameters are film thickness (d) and complex dielectric constants (ε_1, ε_2). For a transparent film, only thickness and ε_1 need to be determined since ε_2 is zero. Even in this simple case, the two measured values (Ψ,Δ) from single-wavelength ellipsometry do not provide a unique solution. There are multiple thicknesses separated by a periodic interval that produce the same Ψ,Δ values [9]. To secure a unique solution, this simple case requires additional information which can often be found by varying wavelength or angle of incidence. Absorbing films complicate the situation as ε_2 is non-zero. A unique solution for thickness and dielectric constants requires either an increase of measurement information or reduction of unknown sample properties. This section details the various optical methods used for unique measurement of absorbing films.

The simplifying assumption of a single, homogenous layer with sharp interfaces will not be correct for many absorbing films. It is common for a film to have a thin surface oxide or contaminate layer just as it is possible to find microscopic roughness that will affect the measurement. Additionally, film dielectric constants may vary with depth. These features increase sample complexity to the extent that it may become impossible to arrive at a solution for the exact film microstructure with only optical measurements. However, combined with the methods described in this section, the optical measurement is often more sensitive to sample structure. Accurate characterization of material optical constants often requires a combination of optical methods and alternate metrology techniques that can provide a more complete understanding of the film.

3.1. Opaque film

A simple approach for absorbing films is to measure an opaque layer. Here the problem is reduced to two unknowns: $\varepsilon_1(\lambda)$ and $\varepsilon_2(\lambda)$. A direct inversion of $\Psi(\lambda)$ and $\Delta(\lambda)$ can provide the "pseudo" dielectric functions — so named because of the assumption that a single reflection comes from a sharp interface between material and ambient [10]:

$$\langle\varepsilon\rangle = \langle\varepsilon_1\rangle + i\langle\varepsilon_2\rangle = \sin(\phi)^2 \cdot \left[1 + \tan(\phi)^2 \cdot \left(\frac{1-\rho}{1+\rho}\right)^2\right] \quad (3)$$

For bulk materials with no surface layers, the "pseudo" dielectric functions are equivalent to the intrinsic dielectric functions. However, any roughness, oxidation or surface contamination will shift the "pseudo" dielectric functions away from the intrinsic values [1]. For many applications this approximation is adequate, as the "pseudo" dielectric functions represent the interaction of light with the complete sample.

However, these dielectric functions may not match the values for thinner layers.

This technique was recently applied to opaque films of iridium by Yan et al. [11]. To minimize surface effects, extremely smooth films were deposited. In addition, the remaining level of roughness was corrected in the optical model as quantified from atomic force microscopy measurements. This study was not extended to thinner layers and benefited from the absence of surface oxidation.

3.2. Transparent spectral region

When films are transparent over a portion of measured wavelengths, the problem is reduced to thickness and $\varepsilon_1(\lambda)$. Once thickness is determined, its value is fixed and both $\varepsilon_1(\lambda)$ and $\varepsilon_2(\lambda)$ can be determined at all measured wavelengths. This method is useful for dielectrics, organics, and semiconductors that are transparent at long wavelengths. Unfortunately, it does not apply to metal films, which absorb at all wavelengths.

Although this method is straightforward to implement, the result depends on how accurately the final model describes the true film structure (thickness, optical grading, roughness, etc.). Unlike modeling an opaque layer, where model inaccuracies are incorporated into the final optical constants, an incorrect model may not "fit" both transparent and absorbing regions. This helps identify an inaccurate model and allow further testing to find the correct model.

3.3. Optical constant parameterization

The total number of unknown properties can be reduced by describing the optical constants with a dispersion equation. For absorbing materials, the dispersion equation must describe both $\varepsilon_1(\lambda)$ and $\varepsilon_2(\lambda)$. These two values are not independent, as they are interrelated via the Kramers–Kronig (K–K) relations [3]. Many dispersion equations enforce K–K consistency to further reduce the number of parameters while maintaining a "physical" shape. Optical dispersion over a wide wavelength range often requires a summation of oscillator terms (Lorentz, Gaussian, Tauc–Lorentz, etc.). Although the dispersion equation reduces the set of possible $\varepsilon_1(\lambda)$ and $\varepsilon_2(\lambda)$ values, it still must describe the material accurately. Many references discuss further details of dispersion equations for absorbing materials [3,4,12].

3.4. Multiple angles

It is often assumed that measurement information will increase proportionately to the number of measured angles. However, the information content is not necessarily different. New information is only available if the change in angle significantly changes the light interaction with the sample.

The "pseudo" dielectric function offers a convenient test of whether multiple angles do provide new information. For a bulk-like material, data from multiple angles will map to the same "pseudo" dielectric functions. When the detected light comes from more than the first surface reflection, the "pseudo" dielectric function becomes simply a transformation of experimental data and no longer represents the bulk material dielectric function. If an angle change also significantly affects the measurement condition, the pseudo-optical constants will vary with angle. This confirms that new information about the sample structure is available by varying angle of incidence.

SE data measured at four angles on a thin TiN layer on Si substrate are shown in Fig. 2a. The data from all angles map to the same "pseudo" dielectric functions ($<\varepsilon_1,\varepsilon_2>$ from Eq. (3)) as

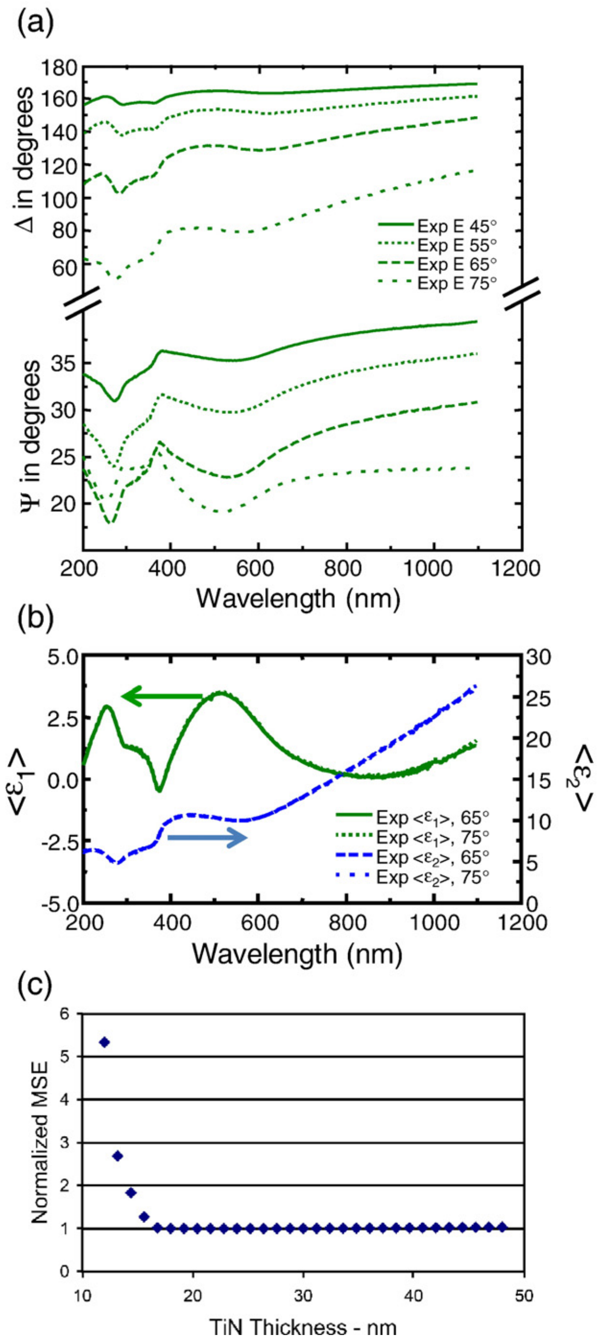


Fig. 2. (a) SE measurements at four angles for a TiN thin film on Si substrate. (b) The "pseudo" dielectric functions ($<\varepsilon_1, \varepsilon_2>$) for 65° and 75° are nearly identical, indicating that both angles provide essentially the same information. (c) Uniqueness Tests of SE data from four angles show that a similar low MSE value is achieved when fitting TiN optical constants with wide range of film thickness.

demonstrated in Fig. 2b for 65° and 75°. In this example similar information is obtained from different angles. The Uniqueness Test in Fig. 2c shows how the MSE varies when optical constants are “fit” to find best result for different TiN thickness values. The fit result is nearly equivalent for any thickness above 17 nm. In other words, a set of optical constants can be fit to match the data equally well for different film thicknesses. There is obviously strong correlation between thickness and optical constants and no unique result. Fig. 2c also makes it evident that no satisfactory optical constants could be fit to match the SE data with thickness below 17 nm. This is due to a restriction forcing ε_2 to positive values.

3.5. Interference enhancement

Adding a transparent film below the absorbing layer can provide new information by varying path length for different incident angles. The correlation between optical constants and thickness is reduced as long as multiple angles of incidence are measured and analyzed simultaneously. The thickness of the underlying film must be sufficient to provide interference in the absorbing film dispersion that is highly suspect. In fact, these features may not be K–K consistent. In other words, the presence of features in a section of absorbing film’s dielectric function where the dispersion should be smooth may be the result of incorrect model. This phenomenon is similar to the “Arwin–Aspnes” effect demonstrated for ultrathin transparent films on semiconductor substrates [13,14], where the spectral features correspond to critical points in the semiconductor dielectric functions. In the case of interference enhancement, however, the spectral features are created by the interference oscillations from the thick dielectric film below the absorbing layer. This effect is easier to identify if the dielectric layer is thicker than 500 nm because such thickness increases the number of interference oscillations. Also worth bearing in mind is the fact that the spectral position of these features will shift with change in angle of incidence.

Interference enhancement was demonstrated on thin films of a-C:H by McGahan et al. [2]. This method has found application in the data storage industry for thin magnetoresistive films [15].

3.6. Simultaneous SE and intensity

Another method to enhance the measured information is to combine ellipsometry with intensity-based optical measurements. Because the film is absorbing, Transmittance (T) measurements are often chosen to supplement the SE data. When SE and T are regressed simultaneously the correlation between thickness and optical constants is reduced and a unique solution is possible. The main limitation of this method is that it requires a transparent substrate and accurate Transmittance measurements. In addition, the substrate’s optical constants must be carefully characterized in advance, as any small absorption in the substrate (e.g. float glass) would be mixed into the film’s overall optical constants.

To help characterize many films including diamond-like carbon layers [16] and bimetal stacks of platinum and gold [17] ellipsometry has been combined with Transmission intensity. In this case, the combination of measurements is not limited to SE and Transmittance. Johs et al. have augmented reflected SE measurements with transmitted SE, reverse-side SE, and transmission intensity measurements for complex multi-layer structures on transparent substrates [18,19].

3.7. Multiple sample analysis

If thin film optical constants remain identical over a range of thickness, measurement of multiple samples can provide additional information. Traditionally, the multiple samples compare the same material at different film thicknesses. Simultaneous data regression, with one set of optical constants to describe all films, reduces the correlation. This method can be applied to films on any substrate — transparent or absorbing. Because this method depends on consistency of optical constants from multiple films, it is not typically applicable to films that are strongly dependent on process conditions or microstructure and may show grading versus film depth.

Multi-sample analysis has been applied to a variety of thin films including dielectrics, semiconductors, and metals. McGahan et al. applied this method to their study of a-C:H [2]. Characterization of semi-transparent films by multi-sample analysis has been performed by Jarrendahl and Arwin on Ta_2O_5, ScN, and CeO_2 [20]. It was used prominently by Herzinger et al. for compound semiconductors including InAs, AlAs, and AlSb layers [8,21]. As the semiconductor layers were ultrathin, in this study one of the layer thicknesses was often fixed at the nominal thickness. Thin metals have also been characterized with multi-sample analysis by Tompkins et al., with one of the samples represented by an opaque film [22].

3.8. In situ

SE measurements during film deposition can be considered an extension of multi-sample analysis. Each measurement time provides data for the same film with different thickness. Processing under vacuum helps avoid surface oxidation and contamination. Surface roughness needs to be considered, but can be dynamically monitored. In situ measurements have been combined with other previously listed methods. For example, Pribil et al. have applied in situ transmitted SE with in situ transmitted intensity measurements to characterize thin metal layers [23]. The additional transmission intensity data, collected via the same ellipsometer, helped to minimize correlation between thickness and optical constants. Another recent in situ application was demonstrated by Langereis et al. on ultrathin TiN films deposited by atomic layer deposition [24]. In situ ellipsometry allowed researchers to study the growth process and film resistivity variations.

3.9. Multiple ambient analysis

Another strategy to obtain additional information from a single sample is to change the ambient index of refraction. This change will modify the overall light interaction with the sample

which will test the model: the correct optical model must describe the film under multiple ambients. This approach often increases sensitivity to surface oxides or roughness. The main drawback of this method is the complexity of measuring within a liquid environment; this includes possible window birefringence and modeling the correct ambient index. In addition, the film must be optically stable within different ambient and some films are not. For example, porous films may change index as the ambient penetrates into the film. In fact, multiple ambient analysis is used to measure film porosity.

4. Experimental

4.1. Sample description

The samples in this experiment consisted of chromium layers with five thicknesses; these were deposited in one-inch squares on three different substrate-film combinations. The metal was deposited in an MRC 603 sputter system with argon pressure of 25 mTorr. The chromium thicknesses were nominally 10 nm, 15 nm, 20 nm, 25 nm, and 40 nm. The three substrate-underlayer film combinations were silicon wafer with 100 nm PECVD oxide, silicon wafer with 1000 nm PECVD oxide, and a fused silica wafer with 100 nm PECVD oxide. Thus, all fifteen Cr films were deposited on the same PECVD SiO_2 film. Using masks, each thickness was deposited simultaneously on each substrate-film underlayer until all five thicknesses were present.

4.2. Measurements

Measurements of films deposited on silicon were performed with an M-2000 spectroscopic ellipsometer [25]. This system incorporates a continuously rotating compensator before the sample and a CCD spectrometer for simultaneous spectral readout from 193 nm to 1700 nm.

Measurement of films on fused silica was performed with a V-VASE spectroscopic ellipsometer, which consists of a rotating analyzer with an adjustable Berek waveplate [26]. A dual-chamber monochromator selects measurement wavelengths between 190 nm and 2300 nm. This system captures both transmitted and reflected measurements over a wide range of incident angles.

Both instruments measure the percent depolarization. This is beneficial when working with samples which are non-uniform in thickness, since this causes the measured beam to become partially polarized. Partial polarization can also occur due to finite spectrometer bandwidth or incoherent backside substrate reflections. Depolarization and coherent/incoherent beam modeling are detailed elsewhere [19].

5. Results

Optical measurements of thin absorbing films can result in a high level of correlation between thickness and optical constants. Section 3 reviewed a number of methods used to reduce the correlation by increasing the amount of measured information or reducing the number of unknown sample properties. In the experiment, three series of Cr films are measured to test and compare these methods. For the purpose of demonstrating the capabilities of each method, the Cr optical constants are allowed to vary independently at all wavelengths for every study except the Optical Constant Parameterization. In other words, no dispersion relations are forced upon the Cr dielectric functions when interference enhancement, multi-sample analysis, and combined SE and Intensity methods are employed.

The goal of this work is not to characterize the Cr films with utmost accuracy. Rather, the goal is to demonstrate the analysis approaches reviewed in this paper. Thus, certain simplifying assumptions are made. Cr is known to oxidize readily to a stable thickness. However, for simplicity's sake, this work ignores all surface layers. This assumption should not affect the demonstration and comparison of various methods. Of course, a careful study of the Cr films themselves would benefit from additional metrology methods to measure surface roughness, oxidation, and possible optical variation with depth.

5.1. Reference samples

All three substrate-film underlayer reference samples were characterized with variable angle SE. Si substrate and thin interface (fixed at 1 nm) optical constants were fixed at values reported by Herzinger et al. [27]. The PECVD SiO_2 index was described using a Sellmeier-type dispersion equation. Oxide thickness and index were determined across the reference wafers and at corresponding locations outside the Cr-coated areas of each sample. Oxide thickness variation between Cr-coated and reference wafer was less than 0.6% and 0.3% for the 100 nm and 1000 nm SiO_2 samples, respectively.

Fused silica substrate optical constants were characterized with combined variable angle SE and transmitted intensity measurements. The SiO_2 film deposited on fused silica was characterized at a point outside the Cr-coated patterned regions. This result was fixed for all further modeling, although its effect on Cr measurements is minimal because of close index match between the PECVD SiO_2 film and the fused silica substrate.

5.2. Interference enhancement

While the addition of a thick dielectric layer between the absorbing film and the substrate raises sample complexity, it also increases available measurement information. Section 3.5 above, describes how 'interference enhancement' has been demonstrated to reduce correlation between thickness and optical constants for thin absorbing layers.

In our experiment, this method is demonstrated on two series of Cr films deposited on SiO_2 coated Si substrates. The interference enhancement provided by SiO_2 coating introduces two advantages. First, a change in path length through the layers generates "new" information about the absorbing film at different incident angles. Second, if the absorbing film "fit" is the correct thickness, the data structure from the underlying dielectric should not appear in the thin absorbing film optical constants. Both effects are discussed below.

The new information which is obtained as a result of variation in path length corresponding to angle variation can be shown by comparing "pseudo" dielectric functions. Data for Cr-coated glass at both 60° and 75° are only slightly separated (Fig. 3a). This is similar to the TiN layer on Si discussed in Section 3.4. However, the "pseudo" dielectric functions at 60° and 75° show significant separation for Cr on both 100 nm and 1000 nm underlying SiO_2 films on Si wafers (Fig. 3b and c, respectively).

When using interference enhancement, it is necessary to measure and fit data at more than one angle to directly determine optical constants and thickness for the thin Cr layer. This is illustrated in Fig. 4 using each SE data set shown in Fig. 3. Fits to single-angle data show strong correlation between thickness and optical constants, as demonstrated by the flat MSE in Uniqueness Tests for data taken only at 75°. Fig. 4 shows that the Cr on glass does not benefit from interference enhancement and that strong correlation exists even when one fits multiple angle data. However, there is a well-defined minimum in the MSE when fitting two angles for Cr films on SiO_2–Si. These films benefit from interference enhancement and exhibit a separation in "pseudo" optical constants at each angle.

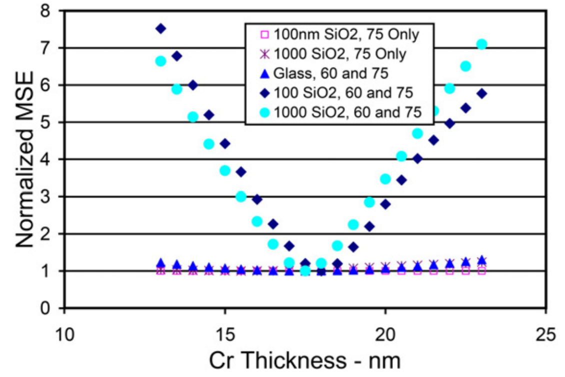


Fig. 4. Uniqueness tests are performed on the SE data shown in Fig. 3. Tests of single-angle 75° data are highly correlated, with no significant MSE minimum. Correlation also exists for multi-angle SE data with glass substrate. Tests of two angle data provide unique results with the addition of interference enhancement (films on SiO_2–Si).

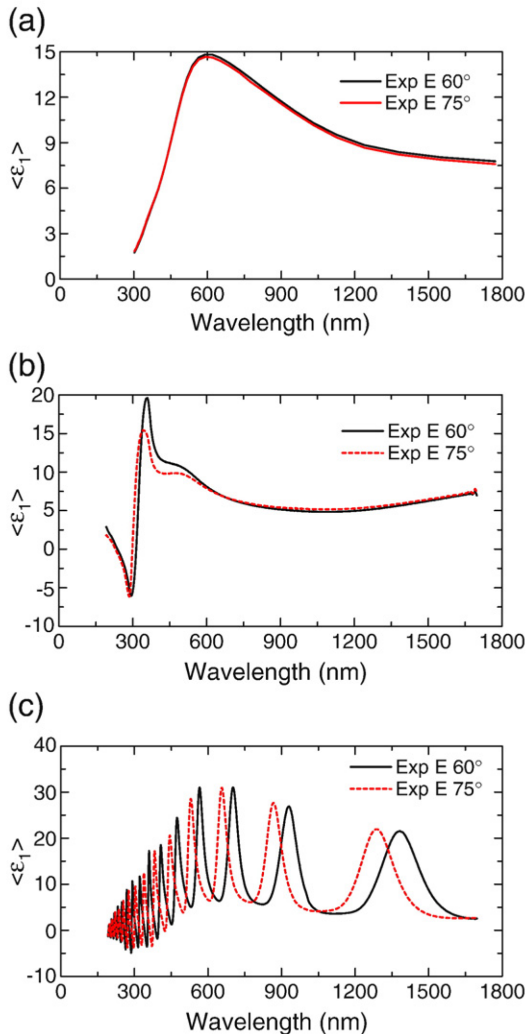


Fig. 3. "Pseudo" dielectric function for three variable angle SE measurements are compared. Samples include a thin Cr layer on (a) bare glass, (b) 100 nm SiO_2–Si, and (c) 1000 nm SiO_2–Si. Separation between "pseudo" dielectric function curves from different angles (in b and c) indicates that each angle produces new information.

It is worth mentioning that when interference enhancement is employed, the final choice of angles is not critical. It is only essential that data from more than one angle are analyzed simultaneously. In our experiment, similar results were obtained using two widely spread angles (45°, 75°) and two closely spaced angles (70°, 75°).

The second benefit provided by interference enhancement is similar to the "Arwin–Aspnes" effect from ultrathin dielectrics measured on semiconductor substrates [13]. Arwin and Aspnes point out that when the model thickness is incorrect, critical point structure from the semiconductor substrate will appear in the surface film optical constants. For Cr layers on 1000 nm SiO_2, it is easy to observe oscillatory features in the apparent Cr optical constants if the absorbing film thickness is not correct. These oscillations correspond to the interference pattern created by the thick SiO_2 layer. An errant model produces Cr optical constants with incorrect (unphysical) features, as shown in Fig. 5.

The effectiveness of interference enhancement depends on the thickness of both the absorbing layer and the underlying dielectric. Data from all ten Cr layers on SiO_2–Si were fit to

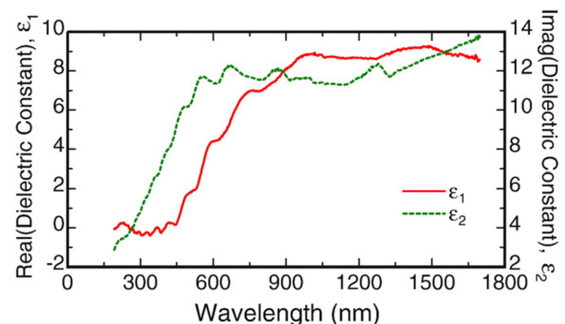


Fig. 5. Cr film dielectric functions from an errant model, where the thick SiO_2 interference features are prominent in the fit Cr optical constants.

determine their thickness and optical constants using interference enhancement. After each fit, a Uniqueness Test was performed on the Cr thickness. The Uniqueness Range (Cr thickness range where MSE remains within 10% of minimum) is compared for all ten samples in Fig. 6. The Uniqueness Range increases as Cr thickness increases. Results are similar for both 100 nm and 1000 nm underlying SiO_2 layers.

Obviously, sufficient interference enhancement depends on the choice of underlying dielectric's thickness. Its primary function is to significantly change the path length of light interacting with the sample at multiple angles to provide new information. For 100 nm SiO_2–Si, there is a single interference oscillation in the visible spectrum. The importance of this interference oscillation is demonstrated by comparing uniqueness tests over restricted spectral ranges. Fig. 7 shows that the fit sensitivity is reduced (Uniqueness Range increases) as analyzed wavelength range shifts to longer wavelengths. For analyzed data at wavelengths above 1000 nm, thickness is completely correlated with optical constants. From this test, one can estimate the minimum metal thickness to enable interference enhancement depending on measured spectral range. This type of study is straightforward and will not be discussed further in this paper.

As explained above, it is best to reduce the total number of unknown fit parameters to reduce correlation. If SiO_2 film thickness and index are well-known, these values should be fixed. Sensitivity to both SiO_2 thickness and index will be caused by the necessity to align interference features versus wavelength and angle of incidence. However, when underlying SiO_2 thickness and index are allowed to vary, as they were in our study, the results are offset compared to reference measurements. This offset is likely compensating for model imperfections related to the absorbing layer, such as ignoring surface oxidation or roughness.

The final Cr metal optical constants determined from individual samples on 100 nm SiO_2–Si utilizing interference enhancement and multi-angle SE data are shown in Fig. 8.

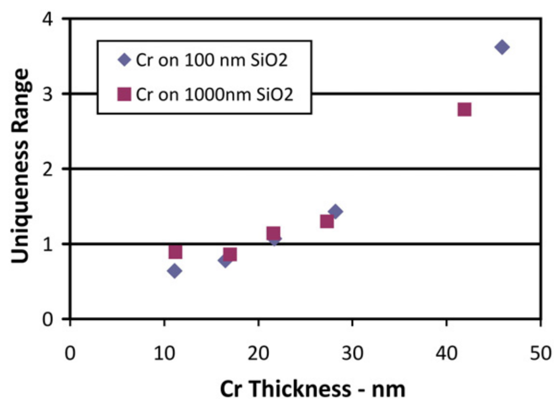


Fig. 6. Ten samples of Cr–SiO_2–Si were measured with variable angle SE. Interference enhancement allowed direct fit of Cr thickness and optical constants. The Cr thickness range that produces MSE values within 10% of the minimum (uniqueness range) is inversely related to thickness sensitivity. Results are similar for both 100 nm and 1000 nm SiO_2, but sensitivity decreases (range increases) for thicker Cr films.

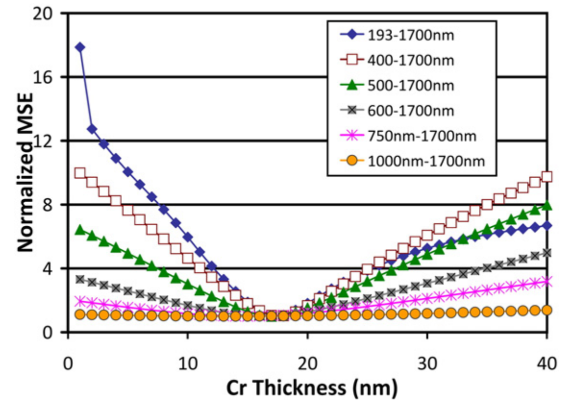


Fig. 7. Multi-angle SE data from a thin Cr film on 100 nm SiO_2–Si is analyzed. The resulting sensitivity decreases as wavelength range is confined to longer wavelengths. With data at wavelengths above 1000 nm, there is complete correlation between film thickness and optical constants.

Although some variation with Cr film thickness may be due to surface condition, the dielectric function variation with film thickness is expected to be realistic.

5.3. Combined ellipsometry and transmission

Correlation between thickness and optical constants for thin absorbing films on transparent substrates can be reduced by simultaneously analyzing SE and transmission intensity measurements. In our study, this method was applied to thin Cr samples on glass. To demonstrate correlation, the Cr optical constants were fit to match variable angle SE data with three different Cr thicknesses: 12 nm, 18 nm, and 24 nm. Each SE data fit, shown in Fig. 9a, is nearly perfect for each thickness choice. The dielectric functions determined from each thickness differ significantly, as shown in Fig. 9b. To determine which combination of thickness-dielectric functions is correct, each is used to generate a normal incidence transmission and a

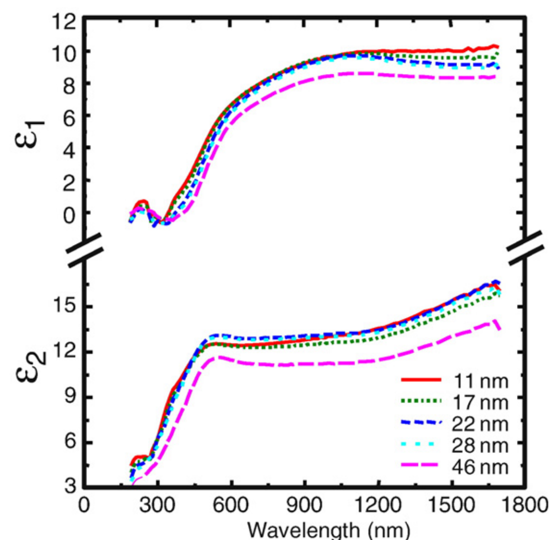
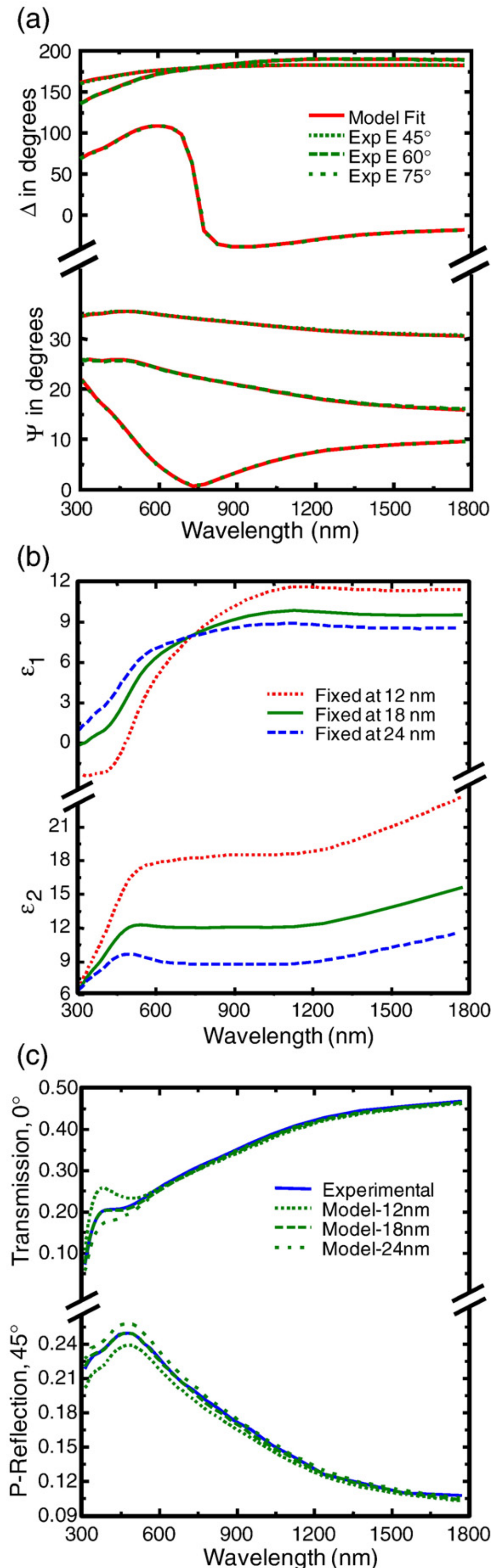


Fig. 8. Cr dielectric functions determined from direct fit of multi-angle SE data of thin Cr films on 100 nm SiO_2–Si.



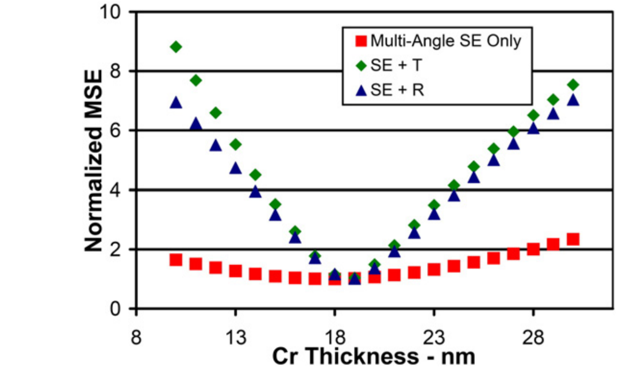


Fig. 10. Normalized MSE uniqueness test of multi-angle SE data from thin Cr film on glass shows a shallow minimum. Increased sensitivity results from simultaneous regression of SE and transmitted intensity or combined SE and reflected intensity data.

p-polarized 45°-angle reflection intensity curve. Comparison to experimental data in Fig. 9c shows that only the 18 nm fit result is in agreement with both *R* and *T*.

A slight separation in "pseudo" dielectric function is found for the multi-angle SE data on glass. In fact, multi-angle SE data yield a shallow MSE minimum versus Cr thickness, as illustrated in Fig. 10. This result will be readily affected by systematic errors. Simultaneous analysis of multi-angle SE and transmitted intensity data greatly improves sensitivity, as the well-defined minimum for the uniqueness test demonstrates in Fig. 10. A similar sensitivity improvement is found for simultaneous analysis of SE and reflected intensity data. Although each analysis provides similar thickness for the 18 nm Cr film, the resulting thickness varies for thicker Cr films along with increasing Uniqueness Range, as shown in Table 1.

5.4. Multi-sample analysis

Additional information can also be obtained by measuring multiple samples containing the same absorbing material. In simple terms, the measurement of three parameters (thickness, ε_1 and ε_2) requires more than a single Ψ,Δ mesurement. With additional data obtained from a second sample, four unknowns (2 thickness, ε_1 and ε_2) can be determined from four measurement values (Ψ_1, Δ_1, Ψ_2, Δ_2). In our study, multi-sample analysis is demonstrated using the fixed-angle 75° data from multiple Cr films on 100 nm SiO_2–Si. Each single-angle data set is correlated. This correlation is reduced by analyzing multiple samples with different thickness, as shown in Fig. 11 for two or more films. However, as thicker films are added to the multi-sample analysis, the MSE minimum increases. This indicates a poorer fit which is due to a variation in Cr optical constants as the film becomes thicker. When each sample has different optical constants, the multi-sample analysis returns an average set of optical constants to best match all data sets. This

Fig. 9. (a) SE data and model fit are nearly equivalent with Cr thickness fixed at 12 nm, 18 nm, or 24 nm. (b) The resulting fit's "thickness-dielectric functions" are used to generate normal incidence Transmission and 45° p-polarized Reflection curves. (c) These are compared to the corresponding experimental curves.

Table 1
Fit results and Uniqueness Range (Cr thickness range that provides fit MSE within 10% of the minimum MSE) for Cr films on glass

Location, nominal thickness (nm)	Data analysis results			
	SE only		SE and Transmission	
	Cr thickness (nm)	Uniqueness Range (nm)	Cr thickness (nm)	Uniqueness Range (nm)
A, 10	13.1	7.9	14.3	0.6
B, 15	17.7	5.6	18.6	1.1
C, 20	27.7	6.2	24.2	0.9
D, 25	23.2	9.3	28.9	2.2
E, 40	34.8	14.3	40.5	3.9

procedure combines information from multiple path lengths through similar Cr layers to determine thickness and optical constants uniquely.

Another multi-sample analysis approach combines samples with the same Cr layer thickness, but deposited on different substrates. The benefit of multiple path lengths in the Cr is lost, but the Cr optical constants may be more consistent since they are derived from the same thickness. For this multi-sample fit, the single-angle 75° SE data of the 18 nm Cr film on all three substrates were regressed simultaneously. Fig. 12 shows the Uniqueness Test for a multi-sample analysis of all three samples that contain nominally 18nm of Cr. In this test, the Cr thickness from 100 nm SiO_2–Si is systematically varied while allowing all additional parameters to fit, including the two remaining Cr film thicknesses. The MSE has a distinct minimum, which would not be present for any individual data set. The same result is achieved with any combination of two or more samples with the same Cr thickness on different underlying substrates.

5.5. Optical constant parameterization

When the Cr optical constants are parameterized, the set of possible curves that can be generated to match the data is reduced. The resulting set of optical functions may also maintain Kramers–Kronig consistency to further ensure the final results are physical. However, optical parameterization

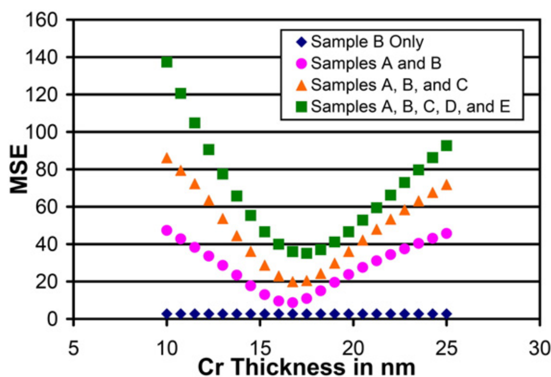


Fig. 11. MSE from uniqueness test comparing multi-sample results when restricting each data set to 75°. Two samples or more allow a unique result. However, the MSE increases significantly as combined fit quality decreases. This phenomenon is explained by the changing Cr optical constants with increasing thickness, which is not allowed in multi-sample analysis.

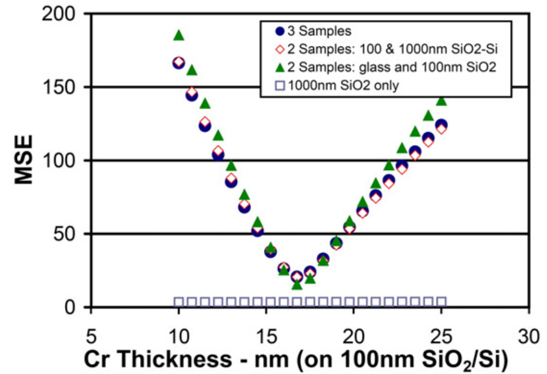


Fig. 12. MSE from multi-sample analysis of single-angle SE data from three samples, each with 18 nm Cr but different substrates: glass, 100 nm SiO_2–Si, and 1000 nm SiO_2–Si. Any combination of two or more samples reduces correlation between thickness and Cr optical constants.

requires careful placement of oscillators to match the optical structure of the absorbing film. For example, the final Cr optical constants may be generally described by the summation of three oscillator terms (Drude and two Lorentz). However, small features in the optical constants may require additional terms, increasing the number of free parameters. The total number of oscillator terms is not typically known until the final optical constants are established, and yet the terms play a role in the uniqueness of the optical model used to fit experimental data.

In Fig. 13, we show the Uniqueness Test for a single-angle of incidence. When the optical constants are allowed to vary independently at every wavelength (no parameterization), complete correlation occurs and any film thickness can provide the same fit quality. Fig. 13 compares the Uniqueness Test when the Cr optical properties are modeled as a combination of additional oscillators, starting with a Drude and Lorentz oscillator (two oscillators) and adding additional oscillator terms to better describe the dispersion of the Cr optical constants. As the number of oscillator terms increases, the MSE minimum reduces, showing improved match to the experimental data whenever Cr optical constants are properly described. However, the number of fit parameters and the set of possible optical functions also increase as the number of oscillator terms rises. This invariably causes the MSE minimum to widen, producing less sensitivity to a

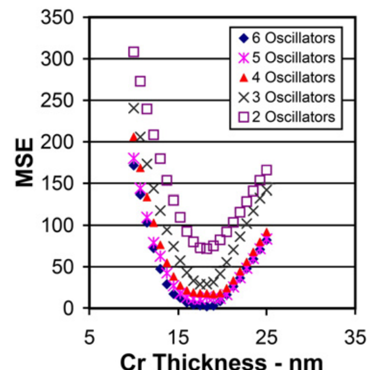


Fig. 13. Uniqueness test from optical parameterization of 75° data for 18 nm Cr on 100 nm SiO_2–Si.

final result. Much of the sensitivity shown in Fig. 13 is the result of interference enhancement. Although this analysis involves single-angle data, the underlying SiO_2 film interference combines with the restriction on dielectric function shape from the dispersion model to provide an MSE minimum versus Cr thickness. Performing the same test on Cr-coated Glass does not show significant sensitivity from optical constant parameterization alone.

For this reason, it is often best to combine optical parameterization with other methods such as interference enhancement, SE and intensity, or multi-sample analysis. Optical constant parameterization will help ensure that the results maintain a physical shape, and the combination of methods will improve sensitivity.

6. Conclusions

The correlation between film thickness and optical constants for thin Cr layers can be reduced by employing four different methods. Interference enhancement is demonstrated to increase measurement information at multiple angles of incidence. This method requires a thick dielectric between the Cr film and the Si substrate to modify the path length of light interacting with the Cr film. Combining SE and transmitted or reflected intensity is also shown to allow a unique result for the Cr films on fused silica substrates. This method also works by increasing the amount of information gained from measurement. However, transmission measurements require transparent substrates, and both transmission and reflection intensity values must be accurately measured. Multi-sample analysis augments the measured information by providing multiple path lengths through similar Cr films. However, this technique requires the optical constants of each deposited Cr film to be nearly identical. This assumption is not valid for the Cr films, but the principle is demonstrated both by combining data from multiple different thicknesses and combining data from the same thickness of Cr on three different underlying structures. Finally, optical constant parameterization can be used to reduce the number of unknown model parameters. This may reduce correlation with the added advantage of maintaining smooth, continuous, and often Kramers–Kronig consistent optical properties for the absorbing material. However, in our demonstration, the result was dependent on the correct optical function description and became less unique with increasing oscillator-model complexity. This method is best utilized when combined with the other methods to further improve model sensitivity. All methods allow the characterization of thin Cr films and, in principle, can be applied to any thin absorbing layer.

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