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Determining the refractive index in the extreme ultraviolet using Kramers-Kronig on thin-film scandium oxide transmission data

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Whereas the real part of the refractive index is dependent on both transmittance and reflectance, the imaginary part can be determined from transmittance data alone. It is possible to use Kramers-Kronig analysis to calculate the real part if the imaginary part is known over a sufficiently broad range. We show that the delta calculated from reflection and transmission data without taking into account roughness may underestimate the real part of the refractive index of the scandium oxide samples we are studying by up to 40% near 270 eV.

I. INTRODUCTION

Although various methods for obtaining optical constants exist, accurate determination of these constants in the Extreme Ultraviolet (EUV) range is difficult[1]. One difficulty arises from the strong EUV absorption of carbon, nitrogen, and oxygen, which are common sample surface and bulk contaminants. Their effect on reflectance can be large. Changes in the apparent

refractive index calculate from reflectance measurements of an active metal surface are observed if its reaction with atmospheric gases are not blocked[1, 2]. The problem is not just the presence of the contaminant. It is also that the amount of the contaminant or oxidation may not be known, therefore the effect on the indices can not be properly taken into account. Blocking oxidation with a surface overcoat has been employed in some cases, but it has its own set of difficulties[2, 3]. The problem of an unknown composition is diminished if a thoroughly oxidized sample, such as scandium oxide, is the object of the study[4]. Though the material is absorbing because of the oxide, it is stable and known and very thin films can be sufficiently transparent for reliable measurements to be made.

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The index of refraction is normally defined as $n + i\kappa$. In the EUV and x-ray portions of the spectrum it is customary to define the index of refraction to be $\delta + i\beta$, where $\delta = 1 - n$. The real part, δ , is determined from the phase velocity and the imaginary part β is determined by the absorption. (In the visible wavelength range k is the symbol often used instead of β .) In 1926–1927 Kronig and Kramers showed that the real part of the index can be expressed as an integral of the imaginary part (and vice-versa)[5]. This optical dispersion relation, which exists in several forms, is the basis of an analytic method known as Kramers–Kronig (KK) analysis.

Although δ and β can both be calculated through measurements of absorption and phase shift of reflected light at lower photon energies, it is often difficult to accurately determine δ through such methods in the EUV and soft x-ray ranges. Surfaces are not ideal, absorption is high, and accessible instrumentation is not sufficiently advanced in this range[3–5]. However, it is usually possible to determine β through such methods, as it can be calculated from transmission data alone, which is not as dependent on surface conditions as reflectance is. The KK relations can be written in a form that involves what is known as the atomic scattering factors f_1 and f_2 . To use this form, it is necessary to note that δ and β are equal to C times f_1 and f_2 respectively, where $2\pi C = n_a r_c \lambda^2$. Here n_a is the number of atoms per unit volume in the optical medium and r_c is the classical electron radius[6].

The power of KK analysis is that it is only necessary to determine experimentally the imaginary part of the atomic scattering factor $f_2^0(\omega)$ to determine mathematically the real part, $f_1^0(\omega)$. KK of $f_2^0(\omega)$ for an element is the standard way of obtaining $\delta(\omega)$ of the material in the EUV[6, 7]. The relationship between $f_2^0(\omega)$ and $f_1^0(\omega)$ is

$$(1) \quad f_1^0(\omega) = Z - \frac{2}{\pi} P_C \int_0^\infty \frac{u f_2^0(u)}{u^2 - \omega^2} du$$

where the first-order term Z is the number of electrons per atom, u is energy, and the P_C indicates that only the non-divergent Cauchy principal part of the integral is to be considered[5, 6]. It is important to note that these equations require integration across the full spectrum of frequencies.

This paper describes the application of KK analysis to scandium oxide(Sc_2O_3) β data over a given range to produce the real part $\delta(\omega)$. The usual method for our group to obtain both $\beta(\omega)$ and $\delta(\omega)$ is directly from measurements of transmission and reflection at the Advance Light Sources (RT δ)[4]. We also show that, as expected, the KK method yields unphysical results when applied to experimental data of a finite range, but is reasonable when augmented for the range above and below our experimental data with approximate values of β for scandium oxide. The augmenting data we used were calculated via the isolated atom model using a program at the CXRO[8]. We believe that the $\delta(\omega)$ resulting from KK analysis may be more reliable over higher energy ranges, than the method of calculating $\delta(\omega)$ directly from reflection and transmission (RT method). The RT method may underestimate $\delta(\omega)$ by up to 40% at short wavelengths. This may be due to roughness scattering from the samples which will be more pronounced at higher frequencies. It is also possible that the KK analysis may be in error due to uncertainties in the density; and, to a lesser extent, the thickness of the film used to obtain $\beta(\omega)$.

II. COMPUTATIONAL METHOD

We report on the KK analysis on two sets of beta data. The first are synthetic beta values. The second are values of beta obtained from ref 4. The Kramers-Kronig relations seen in Eq.1 are meant to be used on equations, rather than sets of numerical data. The equations for δ and β still apply, but to use the KK relations on our data, we needed to change the equations from integrals to summations. Eq. 1 becomes:

$$(2) \quad f_1^0(\omega) = Z - \frac{2}{\pi} \sum \frac{u f_2^0(u)}{u^2 - \omega^2} \Delta u.$$

The arguments of these summations can be evaluated on a point-by-point basis and summed to parallel the integrals in the original equations. To avoid a singularity, we skip over the points for which $u^2 - \omega^2 \approx 0$. This summation is equivalent to the trapezoid rule of integration. More precise ways of integrating could have been used. But these were judged to be unnecessary since the $\beta(\omega)$ values were close together and slowly varying.

We tested KK first on synthetic values of β . These were calculated using the indices program at the CXRO website[8]. We wanted to see how well our method works for data similar to the experimental data we would like to analyze.

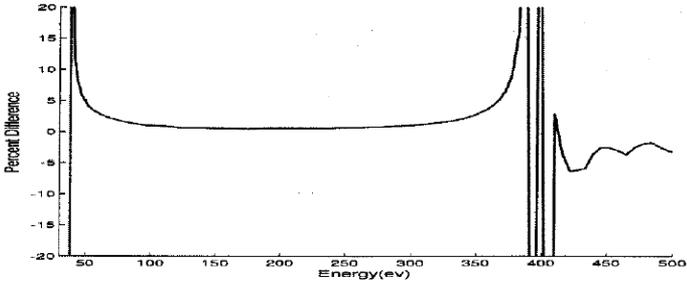


Figure 1: Kramers-Kronig analysis on CXRO $\beta(E)$ data to get δ – Plot of the percent difference between CXRO δ values and the KK δ (calculated using CXRO β data from 30 eV to 30 keV). They are plotted as $\beta(E)$, where $E = \hbar\omega$. We will use ω and E interchangeably in the text.

Values provided by CXRO included both the real and imaginary parts of the index of refraction for a specified material over a finite range. Each set of values corresponds to a specific density of the given material. The values of these data were calculated through KK analysis. Therefore, by performing KK analysis on the imaginary part of the values, we should be able to replicate successfully the real part. The difference between the CXRO values of δ and those from KK analysis for scandium oxide (for a chosen density of 3.4 g/cm^3) can be seen in Fig. 1. They are plotted as percent difference versus energy. We use ω and E interchangeably in the text. A feature present in δ around 400 eV causes high amplitude oscillations in the percent difference between the KK δ and the δ provided by CXRO. This is the L edge of scandium. We also noted in our calculations that the percent error is significantly smaller at high energy (not shown), where there are no absorption features[9].

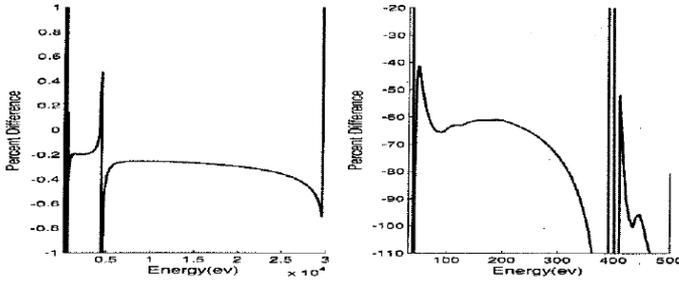


Figure 2: Truncated data sets—Percent difference between the KK calculation and the value provided by CXRO are plotted. The truncation is at 30 keV for the figure on the left and only 500 eV for that on the right. The percent difference increases dramatically when the range of data being analyzed in the KK sums is shortened. This is particularly true near the ends of the ranges, as expected.

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We chose to investigate the effect of not having access to the full energy range. This is because the values of β derived from measured data only covers the energy range of about 40 eV to 270 eV causing our summations to only be approximate. Therefore, a test on truncated sets of CXRO β data reveals that KK analysis is most accurate on sets of data that cover the broadest range of energy. Aside from the obvious trend for KK analysis to lose accuracy as the range of data being used in the sum is shortened, it is also important to note that, as expected, the ends of each set of data have the highest error. We are applying a finite energy range to a summation that is meant to be over an infinite range of energy. However, we see that when using the full range of available CXRO values (even though these do cover the entire EUV and x-ray range), we are able to calculate δ to within 1% of the CXRO values (see Fig. 2). The reason that it does so well is that function in the integrand in Equation 1 becomes very small for values of $u \gg \omega$. The CXRO numbers do go to 30 keV.

The range for which we measure experimental data is even shorter than the smaller range shown in Fig. 2, which had an error of over 60%. We, therefore, understand that small ranges of experimental data must be augmented with synthetic values for energies beyond those measured to achieve realistic results from KK. The approach is standard in KK[2].

III. APPLICATION OF KRAMERS-KRONIG TO EXPERIMENTAL β DATA TO OBTAIN $\delta(\omega)$ OF SC_2O_3

We finally turn to the KK analysis of the $\beta(\omega)$ obtained from RT measurements of an scandium oxide film[4]. The measured sample was 30 nm thick and was deposited on a PIN detector from International Radiation Devices. An initial run of the analytic program reveals that experimental RT data must be smoothed for KK analysis to yield a smooth δ . Our calculations show that in areas where the data points don't form a smooth line, KK is unable to produce a smooth δ . Instead, the resulting δ is jagged, frequently switching between positive and negative slopes [9]. After we smooth the RT data, KK analysis yields a much cleaner, smoother line. For obvious reasons, the summation used in KK analysis works better on smooth data than on rough data.

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When we apply KK analysis, truncating the sums to include only our experimental data, the resulting difference between our KK calculation and our RT calculation is over 100%. As observed with the synthetic data in the aforementioned numerical experiment, the KK δ did not match the CXRO values any closer than this. As concluded above, it is necessary to extend the range of the data being analyzed by attaching synthetic values at both ends of our experimental data, more nearly completing the summation.

However, we need to know what density to use for scandium oxide in our calculations. Each set of synthetic values corresponds to a single density. If we use CXRO values based on the wrong density, our KK analysis will not work as well as it should. Unfortunately, the exact density of thin films is rarely known. However, testing the effects of using different densities provides some guidance.

Both the KK δ and the CXRO δ scale with the density chosen in the generation of the synthetic data, but the experimental RT δ calculated by Acosta[4] does not depend on presumed density. Matching RT δ and KK δ can suggest what is the proper density. We believe that it is most important to match δ at low frequencies (energy). At high energies wavelengths are shorter, causing light to be more easily scattered by the roughness of the surface, causing less of the reflected light to be collected by the detector. Due to the increase in scattering at these higher energies, we expect our calculation to be greater than those from reflection and transmission measurements. Therefore, the RT calculation would have a lower δ than that calculated by KK analysis.

A δ calculated from the RT data could underestimate the KK δ for rough samples over spectral ranges where scattering occurs. On the other hand, the KK calculation of δ is based on β , which is not strongly affected by roughness scattering. Therefore, we might expect our KK calculation for δ to be greater than those from reflection and transmission calculations. We also expect our calculation to match these values more closely at lower energies, where scattering due to roughness is less of a factor, and to match synthetic data more closely than they match RT values for higher energies. With decreasing energy, scattering becomes less of a problem, and we expect our calculation to converge to the values calculated from reflection and transmission data.

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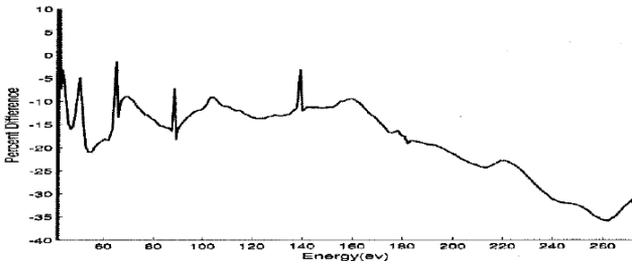


Figure 3: Percent difference in $\delta(\hbar\omega)$ for scandium oxide measured at the ALS, $(\delta_{RT} - \delta_{KK}) / \delta_{KK}$. As energy increases the difference between the two deltas increases. The negative sign on the percent difference shows that the KK δ is greater than the RT δ .

We ran the KK program to calculate δ using several different densities, including 3.86 and 3.2 g/cm³. The only density that gives us a δ that is greater than the RT data at low energies and closely matches CXRO values at higher energies was 3.86 g/cm³, which corresponds to 100% dense scandium oxide. The δ calculated using KK analysis for this density is higher than Acosta's RT calculation from reflection and transmission data at lower energies and follows CXRO values more closely than the values calculated from reflection and transmission at the higher energies, just as we expect. These results lead us to believe that our scandium oxide thin film is close to 100% dense, without large voids in the layer. For further discussion on density see [4]. The results of KK analysis on our RT β data using 100% density (3.86 g/cm³) differ from the δ calculated from reflection and transmission data by less than 50% across the entire range of measured data. The percent difference

between the KK δ values and RT values seen in Fig. 3 is negative because the KK values are greater than those calculated from reflection and transmission data. At the higher energies, the magnitude of the percent difference increases significantly. This is the area in which we expect our calculation to follow the CXRO values more closely than the δ calculated from reflection and transmission data due to roughness scattering effects.

IV. CONCLUSIONS

We have determined the δ of scandium oxide from 40 to 270 eV via KK analysis. This δ matches CXRO values quite closely except in regions with high absorption where it is expected to differ because of chemical shifts. The results of our computational analysis show that the δ calculated from reflection and transmission data for these samples without taking into account roughness may underestimate δ by up to 40% near 270 eV. It is noteworthy that these KK calculations more closely match the synthetic values provided by CXRO in the areas where they least match the RT calculations, as expected[5]. For this method of calculating $\delta(\omega)$ to be accurate, it is necessary to smooth the experimental data and determine the density of the thin film being measured.

V. ACKNOWLEDGMENTS

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