Polarization-ratio reflectance measurements in the extreme ultraviolet

N. Brimhall, N. Heilmann, M. Ware, and J. Peatross*

Department of Physics and Astronomy, Brigham Young University, Provo, Utah 84602, USA
**Corresponding author: peat@byu.edu*

Received January 29, 2009; revised March 7, 2009; accepted March 25, 2009;
posted April 1, 2009 (Doc. ID 106958); published April 28, 2009

We demonstrate a technique for determining optical constants of materials in the extreme UV from the ratio of p-polarized to s-polarized reflectance. The measurements are based on laser-generated high-order harmonics, which have easily rotatable linear polarization but that are prone to brightness fluctuations and systematic drifts during measurement. Rather than measure the absolute reflectance, we extract the optical constants of a material from the ratio of p-polarized to s-polarized reflectance at multiple incident angles. This has the advantage of dividing out long-term fluctuations and possible systematic errors. We show that the reflectance ratio is as sensitive as the absolute reflectance to material optical properties. © 2009 Optical Society of America

OCIS codes: 340.7480, 120.4530.

The real and imaginary parts of the complex index of refraction ($N \equiv n + i\kappa$) describe the optical properties of a material as a function of wavelength. These constants need to be determined before materials can be meaningfully incorporated into the design process for optical coatings. In the extreme UV (EUV) wavelength range (10–100 nm), applications requiring reflective coatings include instrumentation for space-based astronomy, lithography, and microscopy. However, there are many potentially useful materials that have not yet been well characterized for EUV wavelengths [1–3].

Much of the available optical-constant data in the EUV has been obtained from absolute reflectance measurements, typically made at synchrotron facilities. For such measurements, the absolute reflectance (i.e., the ratio of the reflected intensity to the incident intensity) of a sample is measured at a range of angles. The reflectance measurements are then used to deduce optical constants by fitting to a theoretical model [4,5].

We recently constructed a polarimeter based on high-order harmonic generation [6] that can make similar absolute reflectance measurements in the EUV. This instrument uses 800 nm, 10 mJ, 35 fs laser pulses at 10 Hz to produce polarized and directional high-harmonic light in the wavelength range from about 10 to 60 nm. Compared to synchrotron-based systems, this polarimeter represents a compact and affordable "in-house" instrument helpful for developing EUV thin films. The instrument facilitates rapid turn-around time for film characterization and increases opportunities for *in situ* measurements during the thin-film deposition process, helping to circumvent challenges with sample contamination, oxidation, and deposition control.

Since the high-harmonic production depends nonlinearly on the generating laser, small drifts in the laser parameters over the course of a scan (~30 min) can lead to large drifts in the EUV intensity. These drifts can diminish the accuracy of absolute reflectance measurements and necessitate frequent measurements of the incident intensity, increasing the scan time and introducing possible errors in the spatial alignment.

In this Letter, we show that these challenges can be largely overcome by using the *ratio* of p-polarized to s-polarized reflectance measurements to determine optical constants (rather than the absolute reflectance measurement for a given polarization). This technique was first suggested for bulk samples by Avery [7] and for thin films by Miller and Taylor [8]. The ratio technique has been successfully employed to determine material optical constants in the visible and IR regions [9,10]. However, the technique has not previously been used in the EUV region, because of the lack of a source with easily rotatable polarization. Typical values for optical constants in the EUV ($n \approx 1$ and κ in the range 0.01–0.5) are quite different from visible and IR values. The efficacy of the ratio technique in this range of optical-constant values has not previously been explored.

The ratio technique greatly improves the performance of our instrument. We show here that the ratio curve is as sensitive to the material optical constants as the absolute reflectance curves in the range from a few degrees from grazing (where reflection is best) to near Brewster's angle. Thus, there is no disadvantage in extracting optical constants from ratio measurements, but there is a significant advantage to this technique when using an EUV source prone to fluctuations or if the detector is prone to misalignments (assuming that the EUV polarization can be easily controlled).

In our technique, the signal ratio is measured for multiple incidence angles, and then fit to a theoretical model to obtain optical constants. The essential requirement for implementing this technique is to have a linearly polarized EUV source that can be quickly rotated. Plasma sources are unpolarized, and while synchrotron light is naturally polarized, the polarization is not easily rotated. In the case of laser high harmonics, however, the polarization can be rotated with a half-wave plate in the generating laser beam. The harmonics preserve the same linear polarization as the laser [11], so the EUV polarization state can be changed in about 1 s. The measurement can be repeated many times for increased statistical accuracy against laser fluctuation. Since the ratio measurement does not rely on the value for the incident intensity, this technique eliminates systematic errors owing to drifts over time or alignment repeatability.

Another advantage of the ratio reflectance technique is that p- and s-polarized reflectances typically vary from each other by less than 1 order of magnitude over incident angles from 0° to 45° from grazing. This requires the detection system to have a linear response over only a modest range of intensities for an accurate measurement of the reflectance ratio. In contrast, the reflected EUV signal for typical materials is often 2–4 orders of magnitude smaller than the incident signal in this angle range, requiring wide dynamic range for absolute reflectance measurements.

In our ratio technique, a single layer of the material to be characterized is deposited on a known substrate. We determine the thickness of the layer using spectroscopic ellipsometry. Then a series of reflectance ratio measurements is obtained at a range of angles. Finally, the measured ratios are fit (using a least-squares fit) to the theoretical curve to find the optical constants of the layer.

In many circumstances it is necessary to consider more than one layer (see [12]), so we use a multilayer theory. The layers consist of vacuum (region 0), multiple thin films (regions 1 through k), and a substrate (region $k+1$). The thickness of the jth layer is specified by d_j, and the complex index of refraction in each region is $N_j=n_j+i\kappa_j$. Light with wavelength λ is incident from region 0 onto region 1 with incidence angle θ_0, measured from grazing, and propagates in region 1 with angle θ_1 and in region j with angle θ_j. The complex angles are determined in each region from Snell's law: $N_0 \cos\theta_0 = N_j \cos\theta_j$.

The reflectance from the multilayer stack is calculated using a standard matrix approach [13,14]. The total reflectance $R_{s,p}$ for either s- or p-polarized light is given by $R_{s,p}=|a_{21}^{s,p}/a_{11}^{s,p}|^2$. For s-polarized light, a_{21}^{s} and a_{11}^{s} are found from the matrix

$$\begin{bmatrix} a_{11}^{s} & a_{12}^{s} \\ a_{21}^{s} & a_{22}^{s} \end{bmatrix} = \frac{-1}{2N_0 \sin\theta_0} \begin{bmatrix} N_0\cos\theta_0 & -1 \\ N_0\cos\theta_0 & 1 \end{bmatrix}$$

$$\times \prod_{j=1}^{k} \begin{bmatrix} \sin\beta_j & i\cos\beta_j/(N_j\sin\theta_j) \\ iN_j \sin\theta_j \cos\beta_j & -\sin\theta_j \end{bmatrix}$$

$$\times \begin{bmatrix} -1 & 0 \\ N_{k+1}\sin\theta_{k+1} & 0 \end{bmatrix}. \tag{1}$$

For p-polarized light, a_{21}^{p} and a_{11}^{p} are found from

$$\begin{bmatrix} a_{11}^{p} & a_{12}^{p} \\ a_{21}^{p} & a_{22}^{p} \end{bmatrix} = \frac{1}{2N_0 \sin\theta_0} \begin{bmatrix} N_0 & -\sin\theta_0 \\ N_0 & \sin\theta_0 \end{bmatrix}$$

$$\times \prod_{j=1}^{k} \begin{bmatrix} \sin\beta_j & i\cos\beta_j \sin\theta_j/N_j \\ iN_j \cos\beta_j/\sin\theta_j & -\sin\theta_j \end{bmatrix}$$

$$\times \begin{bmatrix} \sin\theta_{k+1} & 0 \\ -N_{k+1} & 0 \end{bmatrix}, \tag{2}$$

where $\beta_j = \frac{2\pi}{\lambda} N_j d_j \sin\theta_j$.

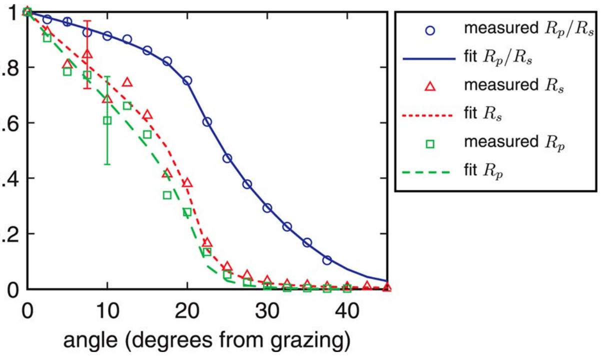

Fig. 1. (Color online) Measured and calculated s- and p-polarized absolute reflectances and the ratio of p-polarized to s-polarized reflectance from a silica sample at a wavelength of 29.6 nm. The error bars represent the average relative residual for each measured data set.

We tested the validity of the ratio measurement technique using our high-harmonics-based polarimeter. We used a naturally oxidized silicon sample with a SiO_2 layer of 1.0 ± 0.2 nm (determined by spectroscopic ellipsometry). The optical constants of SiO_2 and Si are well known [15], giving a standard by which to judge measurement errors. Figure 1 plots the measured absolute reflectance for s-polarized and p-polarized lights as well as the measured ratio reflectance from this sample at a wavelength of 29.6 nm. Each point represents an average of 400 shots at an effective frequency of about 3 Hz (because of laser-energy discrimination used in our instrument—see [6]). The lines show best-fit curves calculated from the previously described model with n and κ for the SiO_2 layer taken as free parameters in a least-squares fitting algorithm. Note that the scatter in measured values for the ratio reflectance is much lower than that of the two absolute reflectances. As discussed above, this is owing to the fact that the ratio measurement is insensitive to long-term drift in the EUV intensity. Since many of the systematic errors have been eliminated using this technique, the remaining error is mostly statistical. In principle, this type of error can be reduced by averaging more measurements.

Given that we can make a measurement of R_p/R_s with more accuracy than we can measure absolute reflectance of either polarization, an important question is whether we can derive optical constants from reflectance ratio measurements with the same efficiency as can be derived from ideal absolute reflectance measurements. To extract optical constants accurately, the theoretical curve for R_p/R_s must be sensitive to variations in the constants.

Figure 2 shows calculated plots of absolute reflectance and the ratio of p-polarized to s-polarized reflectance at a wavelength of 29.6 nm as n and κ are

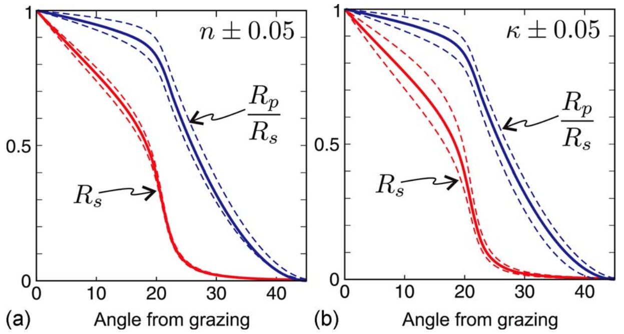

Fig. 2. (Color online) Calculated plots of s-polarized absolute reflectance and the ratio of p-polarized to s-polarized reflectance from a single 1-nm-thick layer of SiO_2 on Si. Solid curves show the accepted values of $n=0.911$ and $\kappa=0.085$. Dashed curves show the extent of the change when (a) n is varied by ±0.05 with κ held fixed, and (b) κ is varied by ±0.05 with n held fixed.

varied. The thickness of the film is chosen to be 1 nm. This relatively thin film allows variations in κ to result in noticeable variations in both the reflectance and the reflectance ratio. κ primarily influences absorption. To see the effects of absorption in a reflectance measurement, an appreciable fraction of light entering the film needs to be able to travel through the film, reflect from the substrate, and return and join the reflected signal. Once a film is thicker than a few absorption depths, essentially all of the light that enters the material gets absorbed.

The heavier lines in Fig. 2 show the reflectance and reflectance ratio for a film with n and κ chosen to be the constants of silicon dioxide at a wavelength of 29.6 nm ($n=0.911$, $\kappa=0.085$). The dashed curves show the variation in the curves as n and κ are each changed by ±0.05. We can see from these plots that the variation in the constants results in similar changes in the ratio reflectance and s-polarized absolute reflectance. (The R_p curve shows somewhat less variation than either of the ones plotted.) This indicates that we can deduce values for optical constants by fitting to the ratio data as well or better than to (noise-free) absolute reflectance data.

We used our measured data (shown in Fig. 1) to evaluate how well the optical constants for the SiO_2 film could be derived from the ratio data. We used a least-squares fitting algorithm to find the complex index of the SiO_2 film with the thickness fixed at 1 nm and the optical constants of the Si substrate fixed at their accepted values. We also included the effect of a thin contamination layer that formed on the sample (see [12]). For $\lambda=29.6$ nm, the algorithm returned values of $n=0.911\pm0.006$ and $\kappa=0.086\pm0.003$ for the SiO_2 film. These values are consistent with the known constants of SiO_2 at $\lambda=29.6$ nm ($n=0.911$ and $\kappa=0.085$). The measured data together with the best fit of the SiO_2 layer are shown in Fig. 1. At a second wavelength, $\lambda=34.8$ nm (not plotted), the fit of the ratio reflectance returned values of $n=0.898\pm0.002$ and $\kappa=0.120\pm0.003$ for the SiO_2 film (whereas the accepted values are $n=0.900$ and $\kappa=0.122$).

For comparison, we also determined the optical constants by fitting to the absolute reflectance measurements (also plotted in Fig. 1). As discussed above, these measurements were subject to systematic errors owing to possible detector alignment errors and long-term drifts in the high-harmonic EUV source. When these data points are used to determine the optical constants, they give less accurate results ($n=0.94$, $\kappa=0.043$), which vary somewhat from the known values for SiO_2.

We conclude that utilizing the ratio of p-polarized to s-polarized reflectance is a robust approach for extracting optical constants. This technique can significantly reduce the systematic errors to which our absolute measurements are sensitive. This approach makes for more viable polarimetry with laser-produced high harmonic, with the advantages of relatively low cost, small footprint, and opportunity for *in situ* measurements.

This work was supported by the National Science Foundation (NSF) under grant number PHY-0457316.

References and Note

1. B. Kjornrattanawanich, R. Soufli, S. Bajt, D. L. Windt, and J. F. Seely, Proc. SPIE **5538**, 17 (2004).
2. R. Soufli, A. L. Aquila, F. Salmassi, M. Fernández-Perea, and E. M. Gullikson, Appl. Opt. **47**, 4633 (2008).
3. D. Garoli, F. Frassetto, G. Monaco, P. Nicolosi, M. G. Pelizzo, F. Rigato, V. Rigato, A. Giglia, and S. Nannarone, Appl. Opt. **45**, 5642 (2006).
4. R. Soufli and E. Gullikson, Appl. Opt. **36**, 5499 (1997).
5. J. B. Kortright and D. L. Windt, Appl. Opt. **27**, 2841 (1988).
6. N. Brimhall, M. Turner, N. Herrick, D. D. Allred, R. S. Turley, M. Ware, and J. Peatross, Rev. Sci. Instrum. **79**, 103108 (2008).
7. D. G. Avery, Proc. Phys. Soc. London Sect. B **65**, 425 (1952).
8. R. F. Miller and A. J. Taylor, J. Phys. D **4**, 1419 (1971).
9. R. F. Miller, L. S. Julien, and A. J. Taylor, J. Phys. F **4**, 2338 (1974).
10. H. R. Philipp and E. A. Taft, Phys. Rev. **113**, 1002 (1959).
11. D. Schulze, M. Dorr, G. Sommerer, J. Ludwig, P. Nickles, T. Schlegel, W. Sandner, M. Drescher, U. Kleineberg, and U. Heinzmann, Phys. Rev. A **57**, 3003 (1998).
12. In this measurement, our samples typically become contaminated with a thin layer of hydrocarbons during transfer from the deposition chamber to the measurement chamber. Our algorithm also had to determine the constants for the contaminant layer (the thickness of the contaminant having been characterized by a separate ellipsometry measurement) to extract the constants for the SiO_2 layer. We used the optical constants of carbon for this first layer when determining the constants for SiO_2. Conversely, we obtained the optical constants of carbon when the constants for SiO_2 were used for the layer beneath the contaminant. In the future, we plan to do *in situ* deposition and measurement (an advantage for our instrument) for which sample contamination should be less of an issue.
13. E. Hecht, *Optics*, 4th ed. (Addison-Wesley, 2002).
14. A. Vasicek, *Optics of Thin Films* (North-Holland, 1960).
15. B. L. Henke, E. M. Gullikson, and J. C. Davis, At. Data Nucl. Data Tables **54**, 181 (1993).