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Precise optical constant determination in the soft X-ray, EUV, and VUV spectral range

Najmeh Abbasirad^a, Qais Saadeh^a, Richard Ciesielski^a, Alexander Gottwald^a, Vicky Philippsen^b, Igor Makhotkin^c, Andrey Sokolov^d, Michael Kolbe^a, Frank Scholze^a, and Victor Soltwisch^a

^aPhysikalisch-Technische Bundesanstalt (PTB), Abbestr. 2-12, 10587 Berlin, Germany

^bIMEC, Kapeldreef 75, B-3001 Leuven, Belgium

^cMESA, Institute for Nanotechnology, University of Twente, Netherlands

^dHelmholtz-Zentrum Berlin für Materialien und Energie, Hahn-Meitner Platz 1, 14109 Berlin, Germany

ABSTRACT

Optical constants of materials are essential for predicting and interpreting optical responses, which is crucial when designing new optical components. Although accurate databases of optical constants are available for some regions of the electromagnetic spectrum, for the vacuum ultraviolet (VUV), the extreme ultraviolet (EUV), and soft X-ray spectral ranges, the available optical data suffer inconsistencies, and their determination is particularly challenging. Here, we present a selected example of ruthenium (Ru) for the determination of optical constants from the VUV to the soft X-ray spectral range using reflectivity measurements performed with synchrotron radiation. The subtleties of reflectivity measurements are discussed for a large wavelength range, from 0.7 to 200 nanometers.

Keywords: optical constant, X-ray reflectometry, soft X-ray, vacuum ultraviolet, extreme ultraviolet, synchrotron radiation

1. INTRODUCTION

The design of state-of-art EUV lithography optics is highly dependent on the optical constants (n & k) of different materials within the extreme ultraviolet (EUV) to the vacuum ultraviolet (VUV) spectrum. The accuracy of the optical constants is extremely important for the design and optimization of optical systems, as imprecise optical constants can lead to deviations in the desired performance. The optical constants of many materials for the spectral range of EUV to VUV either have not been reported yet or show noticeable discrepancies. Moreover, when it comes to the optical response of thin films, it varies from its bulk counterpart. For the thin film, its density, contamination, oxidation, and interdiffusion layers affect the optical response of the system leading to more complexity in determining the optical constants. For this reason, the Physikalisch-Technische Bundesanstalt (PTB) has established a new database to fill the gap of optical constants for the EUV to VUV spectral ranges. The Optical Constants Database (OCDB) of PTB is online and can be accessed through the web address (<https://OCDB.ptb.de/download>).

Another common technique to determine optical constants is photo-absorption measurements, which require transmissive, free-standing films.¹ For the soft x-ray to VUV spectral ranges, this approach is challenged by strong absorption. Hence, We use angle-resolved reflectometry measurements with synchrotron radiation to determine optical constants by implementing advanced optimization methods. We demonstrate the results for a broad spectral range from 0.7 nm - 200 nm using the measured reflectance data of ruthenium and discuss the challenges in data evaluation and optical constant determination.

Corresponding author: najmeh.abbasirad@ptb.de

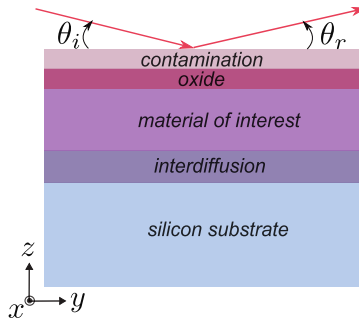


Figure 1. Schematic of the experimental configuration inside the soft x-ray and VUV reflectometers. The s-polarized beam is scanned with the angle θ_i from grazing angle to normal incidence. The reflected light is measured by the detector at the angle θ_r . The thin film usually includes contamination, oxide, and interdiffusion layers.

2. VUV TO SOFT X-RAY REFLECTOMETRY

The Angle-resolved reflectivity measurements at VUV spectral range (photon energy 3 eV- 36 eV) were carried out in the new VUV beamline at the Metrology Light Source (MLS) electron storage ring at the PTB laboratory.² The reflectivity measurements for the soft x-ray (photon energy range from 75 eV -1300 eV) were conducted at the electron storage ring BESSY II at Helmholtz institute Berlin.^{3,4}

Figure 1 illustrates the experimental geometry inside the soft x-ray and VUV reflectometers. A monochromatic s-polarized beam with the angle of θ_i illuminates the sample, and a photodiode detects the reflected light at the exit angle of θ_r where $\theta_i = \theta_r$. For each photon energy, θ_i is scanned from the grazing angle to an almost normal angle of incidence. The position of the detector is changed according to the incident angle. The measured ruthenium (Ru) thin film sample was produced on a silicon wafer substrate using DC magnetron sputtering under a base pressure of about 10^{-7} mbar. Sputtering was performed using argon as process gas at working pressures between 10^{-4} mbar and 5×10^{-3} mbar. The thin film sample has a nominal thickness of 30 nm; however, in several studies of thin films in the EUV spectral range, the reflectance profile represented a multilayered system, including contamination, oxide, and interdiffusion layers as shown in Fig. 1.^{5,6} Particularly, surface sensitivity leads to strong absorption and scattering because of interfacial imperfections that should be addressed when solving the optimization problem.

3. RESULTS AND DISCUSSION

The optical properties of a multilayered system are characterized by material properties, layers' thickness, and optical constants at the desired wavelength λ and the angle of incidence. The optical constant in EUV and the soft x-ray is expressed as a complex refractive index, $n(\lambda) = 1 - \delta - i\beta$ or where δ and β are attributed to the phase velocity and the absorption of electromagnetic radiation in the material, respectively. However, for the VUV, the optical constant is represented by $n - ik$ where n is the refractive index, and k is the extinction coefficient. Hence for the sake of consistency, we stick to $n - ik$ representation to demonstrate the optical constants for EUV to VUV spectral range

Figures 2(a) and 2(b) illustrate the reflectance maps measured at the soft X-ray beamline (75eV- 1300eV) and at the VUV beamline (3eV - 36eV). The reflectance map in Fig. 2(a) depicts the reflectance profiles for the very grazing angles $< 20^\circ$. In the soft x-ray spectral range (550 eV - 1300 eV), it is observed that by increasing the grazing angle, more photon energies are absorbed, resulting in almost zero reflectance from the Ru thin film. The observed high absorption in the energy range of 250 eV- 550 eV in the vicinity of the carbon and oxygen absorption edges is expected due to the carbon contamination of the optics in the beamline path. The reflectance measurements for EUV spectral range from 75 eV to 250 eV were carried out from the grazing angle to a very close angle to the normal incidence, although here we only show the reflectance profiles for the angles $< 20^\circ$. The reflectance profiles at this energy range present the Kiessig fringes indicating the properties of a multilayered system which in turn provides us with more information to precisely determine the different thicknesses of the multilayered structure and its optical constants.

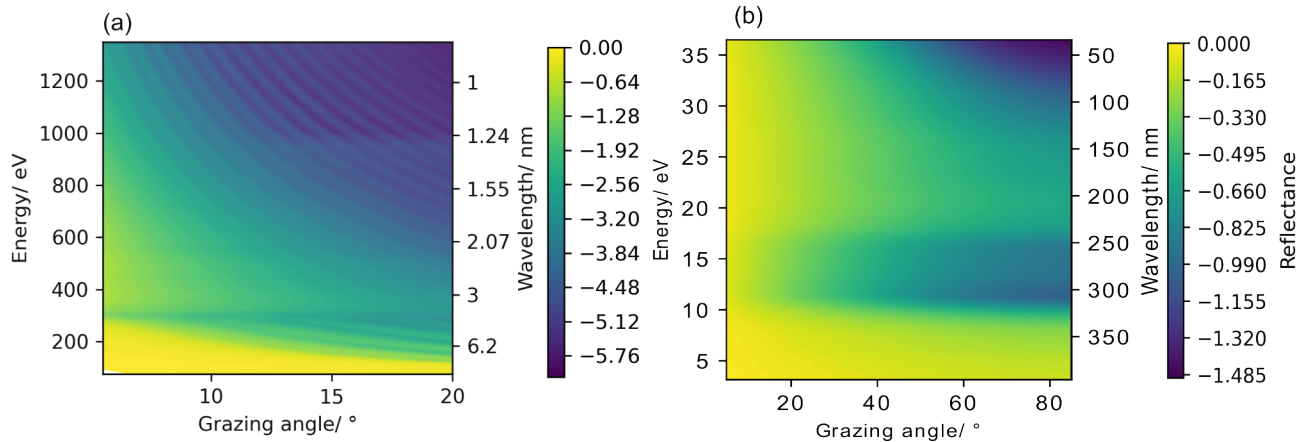


Figure 2. (a) Measured reflectance map of the thin film of ruthenium on the silicon substrate for the photon energy between 75 eV- 1300 eV (b) Measured reflectance map of the same thin film of ruthenium for the photon energy between 3 eV- 36 eV obtained from another beamline.

Figure 2(b) demonstrates the measured reflectance map from energy 3 eV - 36 eV (36 nm - 400 nm) enabled by a new VUV beamline at MLS. The reflectometer of this beamline allows the incident beam from the grazing angle to the almost normal angle of incidence for all the available photon energies. These measurements were successfully carried out after the commissioning time of the beamline for the entire available spectral range of the beamline. The uncertainty budget of this beamline is well established, allowing the more precise determination of the optical constant with the known uncertainties, which is lacking in most of the reported optical constant literature data. The broad spectral range measured in this beamline requires different optics for supporting measurement at different wavelength ranges. Thus, the precise alignment of the optics is essential before conducting the measurements at desired wavelength range. Since the wavelength of the incident beam is larger than the thickness of the Ru thin film, no Kiessig fringes are observed on the reflectance map, which also leads to the more demanding computation time and slow convergence of the optimization problem to retrieve the optical constants of the Ru for VUV spectral range.

Figure 3(a-d) shows the calculated n and k for the photon energy from 3 eV-35 eV and 75 eV - 200 eV, which are the most interesting energy ranges for lithography purposes. In the optimization problem, first, the reflectance from the multilayered systems is computed using the transfer matrix method.⁶ Then a differential evolution algorithm⁷ optimizes the optical constants, layers' thickness, and roughnesses of the measured thin film. The observed Kiessig fringes in the reflectance profile of Ru in the EUV range provide insight into the presence of the contamination, oxide, and interdiffusion layers.⁵ For energy range from 75 eV - 200 eV, the calculated n and k in Figs. 3(a-b) overlapped with the reported optical constants of the Ru in OCDB and CXRO database.^{1,5} The determination of the optical constant in the VUV range is more challenging due to the very long convergence time of the optimization problem. To verify the still partly unknown experimental uncertainties in the VUV spectral range, no simple meta-heuristic optimizer like differential evolution can be used in the optimization. Monte Carlo simulations (Markov Chain Monte Carlo) are necessary to calculate the probability distributions of the n & k values. This increases the numerical effort considerably. Figures 3(c-d) show preliminary obtained n and k for the energy range 3 eV - 36 eV. We compared the simulated optical constant of the Ru with the reported values in literature.⁸ The discrepancy is observed between the simulated values for n for the calculated refractive index of the Ru compared to the reported values in the literature. Nevertheless, the thickness of the Ru 28.3 ± 0.3 nm was attained, solving the optimization problem in both EUV and VUV spectral ranges.

4. SUMMARY AND OUTLOOK

The measured reflectance profiles of the thin film of Ru from the soft x-ray to the VUV spectral range were demonstrated. Using advanced optimization techniques, the optical constants of the Ru for the EUV and VUV range were reported. The obtained thickness for the ruthenium was consistent for both EUV and VUV data

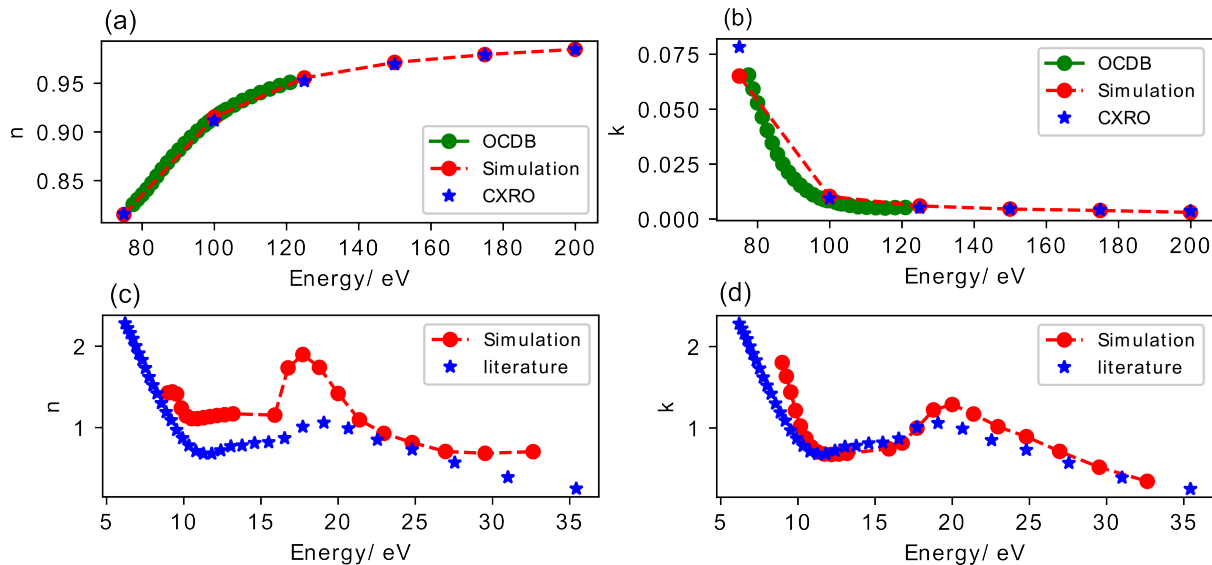


Figure 3. (a) and (b) simulated n and k of the Ru for the EUV photon energies 75 eV - 200 eV compared with the OCDB and CXRO optical constant database. (c) and (d) simulated n and k of the Ru for the VUV photon energies 3 eV-35 eV compared to the reported values in the reference⁸

evaluation. It is apparent that the determination of the optical constants for the VUV spectral range is more challenging and time-consuming. The optical constants of ruthenium and more measured materials for the entire range of X-ray to VUV are a work in progress and will soon be available on the OCDB database.

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