VUV and XUV reflectance of optically coated mirrors for selection of high harmonics

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Abstract: We report the reflectance, ∼1.15° from normal incidence, of six different mirrors as a function of photon energy, using monochromatic vacuum ultraviolet (VUV) and extreme ultraviolet (XUV) radiation with energies between 7.5 eV and 24.5 eV. The mirrors examined included both single and multilayer optical coatings, as well as an uncoated substrate. We discuss the performance of each mirror, paying particular attention to the potential application of suppression and selection of high-order harmonics of a Ti:sapphire laser.

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1. Introduction

Recent developments in multilayer interference coating capabilities have enabled efficient reflection of vacuum ultraviolet (VUV) and extreme ultraviolet (XUV) photons [1-5], despite the multitude of atomic and molecular electronic resonances that strongly absorb radiation at these wavelengths. These effects are achieved by alternating thin layers of two materials with different indices of refraction, which can satisfy a particular Bragg reflection condition for incident radiation [6]. Since the result is constructive interference across a range of wavelengths, certain mirrors can be fabricated that are optimized for reflecting particular photon energies.

Despite the advances in optical coating technologies, characterization of the VUV and XUV reflectance of these coatings relies almost exclusively on theoretical models [7] that are not well tested for VUV light. The atomic and material properties of the elements are tabulated by the Center for X-ray Optics (CXRO) database for photon energies from 30 eV - 30,000 eV, which represents an energy regime where atomic scattering factors can be used reliably for calculating the optical properties of condensed matter. This is because across this energy range,
modeling the atoms within a material as independent scatterers of radiation is a good approximation [7]. Below 30 eV, photon energies approach the valence ionization thresholds for all materials and atomic scattering factors become less dependable for calculations, since the coating material can no longer be treated as a collection of non-interacting atoms. For VUV photon energies, experimental data becomes important to accurately model the optical properties of condensed matter, but is largely unavailable. Synchrotrons, free-electron lasers, excimer lasers, and nonlinear optical processes, such as strong-field driven high harmonic generation (HHG) [8,9], provide access to photons in the VUV and XUV range, and are widely used by scientists and engineers working in an array of fields, making mirror reflectance measurements beneficial to a broad community. Reflectance measurements additionally provide the theoretical optics community with benchmarks for calculations.

Of the light sources mentioned above, HHG uniquely enables the table-top production of attosecond and femtosecond, coherent pulses of VUV and XUV radiation and has become a valuable tool of the ultrafast science community in the years since its discovery. The pulse durations and energies of these harmonics make them suitable for studying photon-driven physical and chemical processes in the time domain. For example, an electronic transition in an atomic or molecular system could be accessible using a relatively narrow band of the HHG spectrum, such as one odd harmonic of the fundamental. However, HHG techniques typically produce many orders of odd harmonics that are of comparable intensity, which can lead to unnecessary complexity in the measurements of interest. This has triggered research interest within the atomic and molecular physics, photochemistry, materials, and photonics communities to select and isolate narrow bands of harmonics. Since HHG produces a broad spectrum of discrete harmonics, and the ability to control this process to produce a relatively narrow spectrum, such as a single harmonic, is very limited in its current state, other methods for beam filtration have been explored. While techniques exist to disperse harmonics in time [10] and space [11], they generally result in significant losses and are limited in application to specific ranges of energies. An alternative approach is to use specialized optically coated mirrors that reflect the desired photons, while suppressing the undesired harmonics [12-15].

Since VUV and XUV radiation is absorbed significantly during reflection by most material interfaces, these photons can be difficult to steer and focus without substantial losses. This makes characterizing the reflectivity of various materials in the VUV and XUV range of the electromagnetic spectrum useful for developing a wide range of experiments and technologies, such as current- and next-generation photolithographic techniques [3,16], attosecond pulse compression methods [17,18], and table-top coherent diffraction imaging [19]. Here we report measurements of the reflectance, $\sim 1.15^\circ$ from normal incidence, of five optically coated mirrors and one polished substrate for photon energies between 7.5 eV and 24.5 eV (51 - 165 nm).

2. Experiment

Measurements were taken at the Advanced Light Source (ALS) using the synchrotron radiation produced at Beamline 9.0.2. The energy of the photons was variable through the adjustment of the beamline’s undulator gap, and finely tunable through the use of the beamline’s three meter off-plane Eagle monochromator. The higher harmonics typically present from the undulator were suppressed through the use of a differentially pumped He or Ar gas filter, with a path length of 115 mm, and operating at 20 and 30 Torr, respectively. In addition to the gas filter, a removable MgF$_2$ window could be placed in the beam path upstream from the monochromator to further suppress the undulator harmonics with energies above 11 eV. The typical photon energy bandwidth downstream from the monochromator was approximately 0.1% (10 - 50 meV resolution). The spot size of the collimated beam at the endstation was approximately $500 \times 350 \mu m$, with a photon flux of $\sim 10^{14}$ photons/second.
Fig. 1. Detector position 1, measuring full photon flux at normal incidence, and position 2, measuring reflected photon flux $\sim 1.15^\circ$ from normal incidence. The angle of reflection from the mirror is exaggerated for illustrative purposes. Not to scale.

To measure the reflectance of the different materials across a desired range, the interchangeable mirror was situated inside of a six-way cross high vacuum chamber. The mirror was placed in the beam path, $\sim 1.15^\circ$ from normal incidence, which reflected incident photons to a detector. The detector, consisting of an VUV/XUV photodiode (Opto Diode Corp, AXUV100G), was mounted to a rotatable, linear manipulator on top of the chamber, which permitted the measurement of both the incident ($I_0$) and reflected ($I_R$) photon flux with the same detector, without changing the angle or position of the mirror. The detector and mirror were separated by about 100 mm horizontally inside the chamber itself, hence the reflected beam propagated approximately 4 mm vertically before reaching the detector, corresponding to an angle of incidence of $\sim 1.15^\circ$. The beam was well collimated, resulting in no significant difference in the beam profile at either detector position for flat mirrors, while the curved mirrors generated a loose focus that occurred near the detector. It was verified that the response of this detector was far from saturation and earlier tests of the same detector with a 158 nm wavelength beam demonstrated that the variation of detection efficiency with spot position on its surface was below 1%. The photocurrent from the photodiode was measured after the dark current, which was $<10^{-4}$ of the measured photocurrent, was subtracted. The resulting analog signal was digitized by a 486 Keithley picoammeter and an ADC. This digital signal was then acquired and stored on a PC using an in-house data acquisition program written in LabVIEW.

The detector was located and oriented in either of two positions (Fig. 1): one for measuring the full photon flux, position 1, and another for measuring the reflected photon flux, position 2. To record the full signal, the photodiode was lowered into the beam path at normal incidence
to position 1. To record the reflected signal, the photodiode was rotated by 180° and retracted to position 2. In this configuration, the beam freely passed under the detector, and propagated to the mirror, \( \sim 1.15° \) from normal incidence, upon which some portion of the photons were reflected to the detector.

First, the gas filter was filled with \( \sim 30 \) Torr of Ar. The photodiode was then lowered to position 1 and the photon flux at normal incidence was recorded across the range of 7.5 - 15.25 eV. Following this measurement, the photodiode was rotated and retracted to position 2, where the collimated radiation then propagated to the mirror. The reflected photon flux was then recorded by the rotated and retracted photodiode across the same energy range of 7.5 - 15.25 eV. Next, the gas filter was purged and filled with \( \sim 20 \) Torr of He gas, and the MgF\(_2\) window moved into the beam path. The same series of steps for measuring incident and reflected beam were performed across the range of 7.5 - 11.0 eV. Last, the MgF\(_2\) window was moved out of the beam path, and the same measurement procedure was performed across the range of 12.5 - 24.5 eV.

This simple protocol was repeated for each of the six different mirrors. The mirrors tested include a precision polished flat Si substrate (Gooch & Housego), a protected Al for deep UV coating (Lattice Electro Optics (LEO), PALEUV-B-1025), a B\(_4\)C multilayer coating (CXRO), a dielectric optical coating fabricated for 270 nm (LEO, RX-270-0-B-1025), a dielectric optical coating fabricated for 160 nm (Layertec, 103091), and a multilayer optical coating fabricated for 52 nm (CXRO). With the exception of the Si mirror, each of the mirrors tested were spherical with a 20 cm radius of curvature.

The CXRO fabricated B\(_4\)C mirror consisted of a single B\(_4\)C/Cr stack, which was a 30 nm coating of B\(_4\)C deposited on top of a 5 nm coating of Cr. The CXRO fabricated multilayer mirror for 52 nm consisted of four periods of SiC/Mg stacks, with a bilayer thickness of 35 nm for each stack. The Layertec mirror for 160 nm was a dielectric coated substrate, designed to provide \( > 92 \% \) reflectance at the intended wavelength, while reflecting \( < 10 \% \) of the 800 nm light and \( < 10 \% \) of the light at 266 nm. The LEO mirror designed for 270 nm was a dielectric coated substrate, exhibiting high reflectance at the intended wavelength. The LEO deep UV protected Al mirror was a MgF\(_2\) protected Al metal coated substrate, while the Gooch and Housego Si mirror was an uncoated precision polished plano Si substrate.

### 3. Results

The reflectance, as a function of photon energy, is given by

\[
R(E) = \frac{I_R(E)}{I_0(E)}
\]

where E was measured in incremental steps of 0.25 eV. Three scans were taken per mirror, each with a different photon beam filtration scheme and across a different region of the energy domain. In the regions of the scans that overlapped in energy, the arithmetic mean of the reflectance values was taken to admit a single value at each energy. The resulting data set was then used to generated curves by cubic spline interpolation. The uncertainties were dominated by the pressure stability of the Ar/He gas filter, which was static to within 1\% for a given scan and 2\% between scans. We estimate one standard deviation of the overall uncertainty in our measurements to be \( \Delta R(E) < 0.03 \).

Figures 2 and 3 show the reflectance of the various materials as a function of energy. As a guide to the reader interested in reflectance at the odd harmonics of a Ti:sapphire laser, we indicate the location of various harmonic orders of an 800 nm fundamental wavelength. We have also included a table below showing the energies and wavelengths of various harmonics for different Ti:sapphire driving wavelengths. Below, we discuss which mirrors are best suited for reflecting and selecting various 800 nm harmonics.
Table 1. Wavelength and energy values for odd harmonic orders 5 - 15 with driving wavelengths 780 nm, 800 nm, and 810 nm.

| Harmonic Order | 780 nm | 800 nm | 810 nm |
|---------------|--------|--------|--------|
|               | nm     | eV     | nm     | eV     | nm     | eV     |
| 5             | 156    | 7.95   | 160    | 7.75   | 162    | 7.65   |
| 7             | 111    | 11.13  | 114    | 10.85  | 116    | 10.72  |
| 9             | 86.7   | 14.31  | 88.9   | 13.95  | 90.0   | 13.78  |
| 11            | 70.9   | 17.49  | 72.7   | 17.05  | 73.6   | 16.84  |
| 13            | 60.0   | 20.66  | 61.5   | 20.15  | 62.3   | 19.90  |
| 15            | 52.0   | 23.84  | 53.3   | 23.25  | 54.0   | 22.96  |

At low energies, the 160 nm mirror performs the best, with reflectance above 0.90 at 160 nm (7.75 eV). At 114 nm (10.88 eV), the deep UV protected Al mirror demonstrates the highest relative reflectance, with a value above 0.25. Radiation at 89 nm (13.93 eV) is best reflected by the 160 nm mirror, with a value just over 0.20. Between 54 - 83 nm (14.0 - 23.0 eV), the B₄C coating is the most reflective, maintaining a relatively constant value between 0.11 - 0.18, thus the B₄C coating has the highest relative reflectance at 73 nm (16.98 eV) and 62 nm (20.00 eV). At wavelengths below 54 nm (energies above 23.0 eV), the 52 nm mirror exhibits the highest reflectance.

Fig. 2. Reflectance of the Si substrate, the deep UV protected Al mirror, and the 270 nm and 160 nm optical coatings from 7.5 - 24.5 eV (51 - 165 nm). Measured data are indicated by colored shapes. The curves connecting points were generated using cubic spline interpolation. Labeled vertical lines correspond to odd harmonic orders 5 - 15 of an 800 nm driving field.
relative reflectance, with a value above 0.25 at 53 nm (23.39 eV).

The 160 nm optical coating performs well at its designed wavelength, and it additionally reflects at values near 0.2 at 114 nm (10.88 eV) and 89 nm (13.93 eV). This makes the coating suitable for reflecting the 5th, 7th, and 9th harmonics of 800 nm while suppressing higher orders. The Si substrate has reflectance greater than 0.35 at 160 nm, while exhibiting a steep cutoff with increasing energy, which is favorable for isolation of the 5th harmonic of 800 nm. The deep UV protected Al coating has a reflectance value just under 0.50 at 160 nm, and more than 0.25 at 114 nm, making it an eligible candidate for suppressing harmonics above the 7th. For the 270 nm optical coating, reflectance above 0.20 is exhibited at 160 nm, hence this coating is suitable for suppressing harmonics above the 5th. In the case of the 52 nm optical coating, greater than 0.20 reflectance is demonstrated across the range of 103 - 160 nm (7.75 - 12.0 eV), hence a portion of the 5th and 7th harmonics of an 800 nm fundamental beam are reflected, in addition to the 15th.

Since photons are absorbed at particular energies during transmission through a medium, certain materials transmit photons across particular energy bands, while absorbing across others. By employing transmissive beam filters, such as gas, glass, and metal filters, further suppression of the undesired harmonics and selection of appropriate harmonic may be realized [10,20], in addition to blocking out the co-propagating driving field used to generate the harmonics [21]. Hence through the use of multilayer coated mirrors in parallel with other beam filtration methods, such as transmissive solid and gas filters, significant harmonic isolation can be achieved.

It should be noted that the present B4C and MgF2 protected Al reflectance measurements agree only in overall shape with previous measurements taken [22-25], while the reflectance is overall about a factor of 2 less than for a new B4C coating before exposure to atmosphere. The experiment presented in this paper was conducted in 2015, while the B4C mirror used in this experiment was fabricated by CXRO in 2009 and the protected Al mirror was purchased.
six months before these measurements were taken. It is possible that since the mirrors had not been recently fabricated, the coatings could have degraded in quality over time, in addition to potentially suffering some effects of localized surface oxidation. As pointed out in [22], degradation in the reflectance of the B$_4$C coating is commonly observed depending on the history of the mirror, even when the mirror is stored in a relatively well pressure-, humidity-, and temperature-controlled environment, such as a dry N$_2$ storage box. This is the same case as in MgF$_2$ protected Al mirrors, as demonstrated in [25]. This reflectance degradation is not well understood and its characterization is beyond the scope of this study. Nevertheless, in the case of B$_4$C, since the reflectance has been observed to decrease by as much as 0.10 - 0.20 within six months of storage in a desiccator, a similar degradation provides a possible explanation accounting for the discrepancy in reported measurements, as all mirrors used in this experiment were stored in an N$_2$ desiccator for extended periods of time. This may also serve to explain any disparities in reported MgF$_2$ protected Al mirror reflectance measurements, since the reflectivity of thin MgF$_2$ films have been found to degrade by up to 10% within 15 minutes at atmospheric pressure [25].

4. Conclusion

The reflectance of six mirrors, $\sim$1.15$^\circ$ from normal incidence, was measured across a portion of the VUV and XUV spectrum using tunable, monochromatic photons at the Advanced Light Source. The energy range examined, 7.5 - 24.5 eV (51 - 165 nm), contained photon energies corresponding to the 5$^{th}$ through the 15$^{th}$ harmonic of an 800 nm fundamental wavelength laser. These measurements should be useful for the communities employing VUV and XUV photons and may enable laboratories utilizing broadband VUV and XUV light sources, such as HHG laboratories, to carry out new and more precise measurements and experiments through improved filtration of a relatively narrow band from a typical HHG spectrum. Five out of the six materials tested exhibit modest reflectance of two or more harmonics, making many of them suitable for low or high pass optical filters. By combining multilayer mirror coatings with other simple beam filtration protocols, such as the employment of gas, glass and metal transmissive filters, harmonic selectivity and isolation in HHG beamlines can be achieved.

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