Compact soft x-ray reflectometer based on a line-emitting laser-plasma source

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We describe a compact soft x-ray reflectometer for in-house characterization of water-window multilayer optics. The instrument is based on a line-emitting, liquid-jet, laser-plasma source in combination with angular scanning of the studied multilayer optics. With a proper choice of target liquid and thin-film filters, one or a few lines of well-defined wavelength dominate the spectrum and multilayer periods are measured with an accuracy of 0.003 nm using a multi-line calibration procedure. Absolute reflectivity may also be estimated with the instrument. The typical measurement time is currently 10 min. Although the principles of the reflectometer may be used in the entire soft x-ray and extreme ultraviolet range, the current instrument is primarily directed towards normal-incidence multilayer optics for water-window x-ray microscopy, and is thus demonstrated on W/B_4C multilayers for this wavelength range. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1327307]

I. INTRODUCTION

Multilayer optics are becoming increasingly important for the soft x-ray and extreme ultraviolet (EUV) wavelength region ($\lambda=1$–20 nm). To date, such optics are primarily characterized by synchrotron-based reflectometers. Unfortunately, the accessibility to such instruments is limited, making rapid feedback to the manufacturing process difficult. Furthermore, accurate measurements of multilayer period require a nontrivial calibration procedure. In the present article we describe a table-top reflectometer based on a line-emitting laser-plasma source and demonstrate its applicability for accurate multilayer period measurements and reflectivity measurements on water-window ($\lambda=2.4$–4.4 nm) normal-incidence multilayer optics.

The motivation and purpose of the compact line-source reflectometer described in this article is to characterize normal-incidence water-window multilayer optics[1] to be used as condensers in the Stockholm compact soft x-ray microscope.[2,3] This microscope is based on a 4π steradian, line-emitting, liquid-jet-target laser-plasma source. In order to reduce exposure times, the condenser mirror reflectivity must be high, which requires not only high-quality multilayers but also very accurate control of the multilayer period in order to match the mirror peak reflectivity at normal incidence to the fixed-wavelength line emission of the source. The water-window reflectometer has therefore been designed to provide fast and accurate measurements of especially multilayer period but also reflectivity.

Currently, most of the measurements on soft x-ray multilayer optics are performed with synchrotron radiation in combination with gratings for wavelength selectivity.[4,5] These reflectometers have the advantage of a continuously tunable monochromatic radiation source with high average power making direct near-normal-incidence measurements possible. Unfortunately, there are few such facilities worldwide and even fewer close to multilayer fabrication laboratories. Thus, the turnaround time for measurements are long, making optimization of multilayer structures time consuming. In order to solve this problem, a few compact reflectometers have been developed for in-house mirror characterization.[6–9] These laser-plasma based reflectometers are, compared with synchrotrons, compact and cost effective in development and maintenance. The laser-plasma sources utilized in these instruments are all based on a solid target with a relatively high Z value, producing broadband soft x-ray emission. Wavelength selectivity is provided by a grazing-incidence monochromator and a grazing-incidence spherical mirror is then used to focus the radiation onto the sample. Although these instruments do not provide the same intensity as synchrotron-based reflectometers, they can still be very useful in the development of multilayer optics.

In the present article we describe a compact angle-scanning soft x-ray reflectometer based on a high-brightness water-window line-emitting liquid-jet laser-plasma source.[10–12] This arrangement has several advantages compared to previous table-top instruments. The use of line emission with well-known wavelengths eliminates the need for a complicated grazing-incidence grating monochromator and makes accurate measurements of multilayer period possible without the uncertainty in the calibration of the monochromator. By use of suitable target liquids and free-standing

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metal foils for spectral filtering, the emission-line wavelengths of the reflectometer may be chosen to suit the particular investigation to be performed. Furthermore, the laser-plasma source has a substantially higher brightness than electron-exited line-emission sources[13] and, thus, provides faster measurements. In addition, the source is practically debris free, making damages to x-ray optics, windows, and samples negligible, and it may be operated unattended and without interruptions for a very long time. This article describes the principles, design, and operation of the reflectometer system and initial characterization of W/B_4C mirrors for water-window radiation.

II. PRINCIPLES OF OPERATION

The reflectometer experimental arrangement consists of a line-emitting laser-plasma soft x-ray source and an angular scanning device for measuring the angular distribution of the reflected intensity from the multilayer sample. The two important parameters of multilayer mirrors measured with the reflectometer are multilayer period and reflectivity. In this section we outline the principles for measuring these parameters.

The angular dependence of multilayer reflectivity is given by the Bragg equation compensated for refraction[14]

$$m\lambda = 2\Lambda \sin\theta \sqrt{1 - \frac{2\bar{\delta}(\lambda)}{\sin^2\theta}}, \tag{1}$$

where m is the order of reflection, λ the wavelength, θ the angle of incidence measured from the surface of the mirror, Λ the multilayer period, and $\bar{\delta}$ is the weighted average refractive index of the two materials in the bilayers originally retrieved from the complex refractive index $(n = 1 - \delta + i\beta)$. Equation (1) is usually rewritten as

$$\sin^2\theta = \frac{1}{4\Lambda^2}(m\lambda)^2 + 2\bar{\delta}(\lambda). \tag{2}$$

In conventional single-line Cu $K\alpha$ hard x-ray reflectometry several reflection orders are recorded at grazing incidence and the multilayer period can be determined from the slope $(1/4\Lambda^2)$ of Eq. (2). In our soft x-ray measurements we increase the accuracy of the multilayer period determinations by simultaneously using several first and second order reflection peaks from the well-defined laser-plasma emission lines in one angular scan. This means, however, that $\bar{\delta}(\lambda)$ can no longer be considered as a constant, since the refractive index for, e.g., W and B_4C will vary significantly within the water window. The wavelength dependence of the refractive index has therefore been included in the analyzing procedure which is thoroughly discussed in Sec. IV.

The reflectivity of a multilayer mirror is dependent on the wavelength, angle of incidence, and parameters characterizing the multilayer structure such as multilayer period, relative layer thickness, layer roughness, and interdiffusion. When measuring the absolute reflectivity of the multilayer mirrors, the simplest approach is to use monochromatic radiation. The reflectivity is then directly given as the ratio between the incident intensity and the reflected intensity, both measured simultaneously by two detectors in the reflectometer. If the source is not monochromatic, as in the general case of the line-emitting laser-plasma source, the reference detector will detect all emission lines simultaneously while the other detector will only detect the one line that is reflected from the multilayer mirror. This will result in underestimation of the multilayer reflectivity.

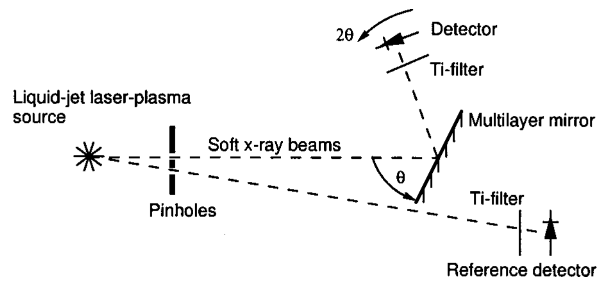

FIG. 1. Experimental arrangement for the compact reflectometer.

In our experimental arrangement the monochromatic radiation is accomplished by using a suitable combination of line-emitting target material in the laser-plasma source and free-standing metal foils for spectral filtering so that only one emission line is transmitted onto the multilayer sample. If no such target/filter combination is available to achieve monochromatic radiation, calibration of the absolute reflectivity measurements may still be performed for a specific emission line using a multilayer mirror of known reflectivity.

To measure the reflectivity at a specific angle θ, e.g., normal incidence, is in the general case not possible since the choice of wavelength is not completely free with a line-emitting laser-plasma source. Furthermore, true normal-incidence measurements of the reflectivity are hard to perform with a plane mirror sample since the detector would block the incident x-ray beam. Our reflectometer can at present perform near-normal-incidence measurements up to a grazing angle of 85°. It is often possible to overcome these limitations by choosing an emission line close to the desired wavelength which gives a reflection peak just below 85°. Although this will not give the reflectivity at the correct angle and wavelength, it is possible to combine the measured data with theoretical models to estimate the reflectivity at the desired angle and wavelength.

III. EXPERIMENTAL ARRANGEMENT

The compact soft x-ray reflectometer is depicted in Fig. 1. The experimental arrangement consists of a laser-plasma source generating soft x rays, a mechanical angular scanning device, a soft x-ray detector system, and a computer for controlling and collecting the data.

The principles of liquid-jet laser-plasma source are described in detail in Ref. 10. In the present experiment laser plasmas are generated when 50–80 mJ, $\lambda = 532$ nm, 3 ns pulses from a 100 Hz Nd: YAG laser are focused onto a target consisting of a liquid jet with a diameter of about 10 μm. Ethanol[11] and ammonium hydroxide[12] were chosen as target materials to generate line emission at suitable wavelengths in the water window. The emitted spectrum from the laser-plasma source using ethanol target is shown in Fig. 2. In this experiment the CVI and CV lines at $\lambda = 2.847$, 3.374,

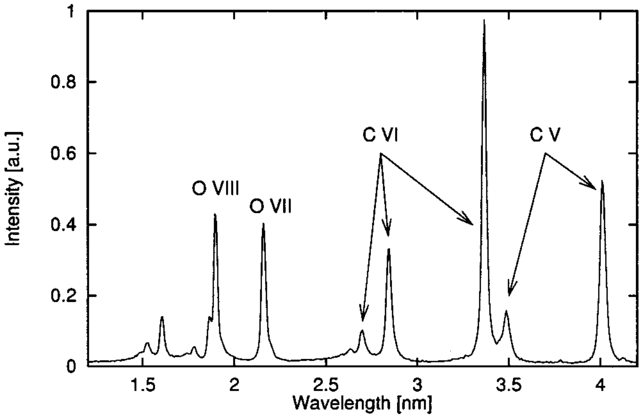

FIG. 2. Emission spectrum from the laser-plasma source utilizing an ethanol liquid-jet target.

and 4.027 nm were utilized for the measurement of multilayer period. The ammonium hydroxide spectrum is shown in Fig. 3, which also includes the transmission curve for the, in total, 1 μm titanium filters. The titanium filter will strongly suppress the nitrogen and oxygen emission lines below $\lambda=2.7$ nm, providing single-line radiation from the $\lambda=2.879$ nm NVI line. The source diameter and flux depends on target material and laser parameters. In the present experiment these are estimated to be 20–30 μm and $5\times10^{10}-5\times10^{11}$ ph/(sr×pulse×line), respectively.

The main difference from earlier experiments with this type of source is that the source is contained in a separate small vacuum chamber of 40 mm in diameter. The source chamber is then connected to the main vacuum chamber (450 mm diameter), where the reflectometer and multilayer mirror sample are located. A vacuum valve in combination with two pinholes separates the two chambers. With this arrangement it is possible to evacuate the vacuum chambers separately and keep a pressure of $\sim10^{-6}$ mbar in the main chamber, while the vacuum in the source chamber is $\sim10^{-3}$ mbar. It also results in a fast sample exchange time (5 min.) since it is possible to open up and change the sample in the main vacuum chamber without venting the source chamber.

The two 500-μm-diam pinholes 70 mm from the source define two collimated soft x-ray beams, each with an angular divergence of 0.4° and an angular separation of 8.1°, see Fig. 1. One of the soft x-ray beams from the source chamber is incident on the multilayer mirror sample under test and it has a spot size of 1 mm diameter on the surface of the mirror. The sample can be rotated by a stepper motor in the range of zero grazing angle θ to normal incidence. Synchronized with this movement is a detector which is rotating with twice the angle of the multilayer mirror in order to detect the reflected x rays from the mirror. The sample can be stepped with a resolution of 0.009° and the zero position ($\theta=90°$) can be reached with an error of less than one step.

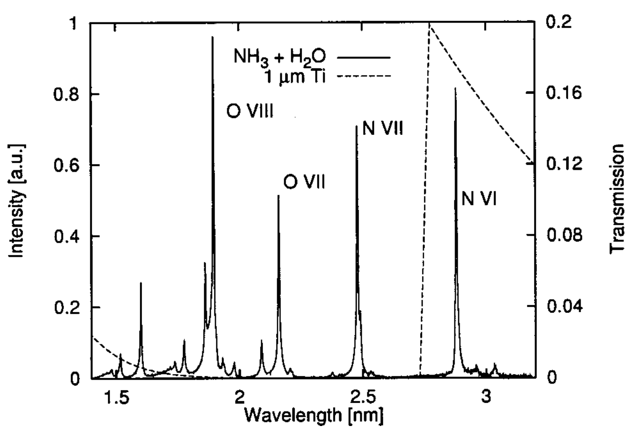

FIG. 3. Emission spectrum (left) from the laser-plasma source utilizing a liquid-jet target of concentrated ammonium hydroxide and the transmission (right) of 1 μm free-standing titanium foil.

The detector system consists of two identical assemblies of a titanium-covered soft x-ray photodiode (AXUV-5, IRD Inc.) and a charge sensitive amplifier (A250, Amptek Inc.). One assembly is mounted on a rotating arm on the reflectometer to measure the reflected intensity from the multilayer mirror. The other, which is placed in a fixed position at the second beam from the x-ray source, is used as reference to monitor the x-ray source intensity. By dividing the first signal by the second it is possible to compensate for long term (10^0-10^3 s) fluctuations in source intensity. The soft x-ray sensitive diode has a quantum efficiency for water-window wavelengths of 80–130 e^-/photon and the charge sensitive amplifier has a sensitivity of 0.12 μV/e^-. The output signal from the amplifier is measured by a digital oscilloscope and range from a few mV (detector) to 3 V (reference detector).

A computer system equipped with an input–output card, stepper motor driver card, and a general purpose interface bus (GPIB) card is used to control the reflectometer and collect the data. A LABVIEW program ties everything together and it features a fully automated scanning and measuring procedure. Once the start, stop, and stepping angles have been given, the program performs the scan and simultaneously collects the measured data from the oscilloscope through the GPIB interface.

IV. EXPERIMENTS AND DISCUSSION

The performance of the reflectometer system was demonstrated on W/B_4C multilayer mirror samples. The multilayer structures were manufactured by a magnetron sputtering process on 20 mm×40 mm silicon substrates with a nominal $\Gamma=0.2$ (ratio between W layer thickness and multilayer period) and were optimized for normal-incidence second-order reflection at $\lambda=3.374$ nm.

A typical angular scan performed on one of the samples is shown in Fig. 4. The scan consists of more than 300 data points, each point an average of 20 laser pulses. The typical data recording time for the measurements presented in this article is 10 min, which is mainly due to the slow data transfer rate from the oscilloscope and may be significantly reduced by using a more rapid data acquisition device. Furthermore, by use of variable step length along the angular scan, the time may be reduced an order of magnitude. The target material in the laser-plasma source was ethanol and the titanium filters were 1 μm thick in total. The three peaks at 25.84°, 31.01°, and 37.03° are the first-order reflections of the $\lambda=2.847$, 3.374, and 4.027 nm lines, respectively, and the last peak at 59.02° represents the second-order reflection

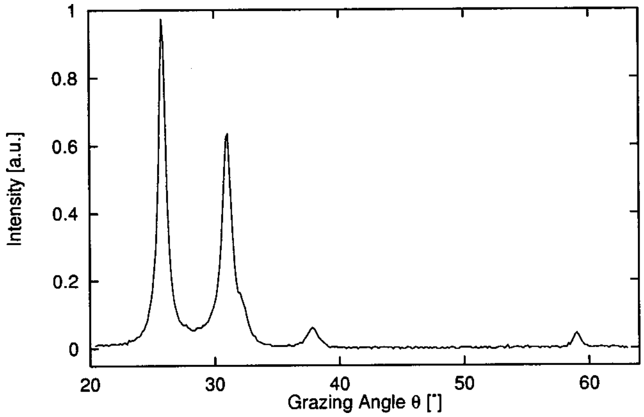

FIG. 4. Reflected intensity from a multilayer sample measured with a laser-plasma source utilizing ethanol target. The peaks at 25.84°, 31.01°, and 37.03° are first-order reflections of the $\lambda=2.847$, 3.374, and 4.027 nm lines and the last peak at 59.02° represent, the second-order reflection at $\lambda=2.847$ nm.

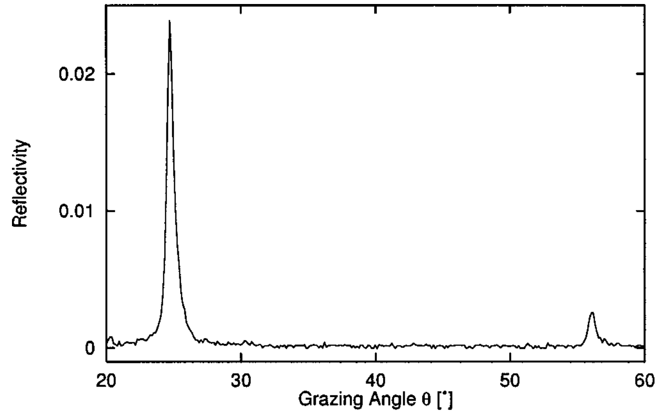

FIG. 5. Reflected intensity from a multilayer sample measured with a laser-plasma source utilizing ammonium hydroxide target. Peaks at 24.99° and 56.32° are first- and second-order reflection of the 2.879 nm NVI line, respectively.

of the $\lambda=2.847$ nm line. From this data the multilayer period was calculated to $\Lambda=3.340$ nm using the procedure described below.

To achieve a high accuracy in the multilayer period measurements, we use as many reflection peak positions as possible when calculating the multilayer period. The first step is to reduce the noise in the recorded data and this is performed by averaging over 20 pulses from the laser-plasma source for each data point, which reduces the noise to typically 3%. When an angular spectrum has been recorded the positions of all reflection peaks are identified by fitting a Gaussian shaped curve to each peak using at least ten data points distributed over the peak. From this the angular peak positions are determined with an accuracy of 0.01°. Note that angular step size normally used is in the range from 0.09° to 0.135° in order to reduce the recording time. Depending on the number of available reflection peaks, different analyzing methods to calculate the multilayer period are used. All methods, however, originate from Eq. (1) or Eq. (2).

If at least two reflection peaks are available, e.g., $\lambda=2.847$ and 3.374 nm, Eq. (1) is used. Here the angular measurements are first compensated for any offset error, due to, e.g., misalignment, by assuming that both measurements should result in the same multilayer period. After the compensation, the final error in the multilayer period value is less than 0.01 nm, provided the peaks are located within grazing angles of 20°–90°. Note that this method requires prior knowledge of the refractive index of the multilayer materials and also the Γ value, since the $\overline{\delta}(\lambda)$ has to be calculated. If the two peaks are first- and second-order reflection from the same emission line, Eq. (2) can be used with $\overline{\delta}(\lambda)$ as a constant.

If four or more peaks are available, as shown in Fig. 4, another method can be used to increase the accuracy even further. This is based on Eq. (2) which is developed into a model equation

$$\sin^2(\theta+\Delta\theta)=c_1(m\lambda)^2+c_2\lambda+c_3, \tag{3}$$

where $\Delta\theta$ is the unknown angular offset error, and $\overline{\delta}(\lambda)$ in Eq. (2) has been replaced by a linear function of λ which is a valid estimate for W/B_4C multilayers in the range of $\lambda=2.847-4.027$ nm. The constant c_1, and thus the multilayer period, is found with high accuracy using a least-square fit. The accuracy of the multilayer period value using this method has been estimated to 0.003 nm. The error analysis has been done by applying random errors on each peak position and a random angular offset error with known standard deviations and studying how they influence the final result.

The requirements on the accuracy of the multilayer period measurements performed with the soft x-ray reflectometer are guided by the demands on the microscope condenser performance. Assuming a bandwidth of $\lambda/\Delta\lambda=80$ for a multilayer mirror optimized for the $\lambda=3.374$ nm line, an error of 0.003 nm in the multilayer period would reduce the reflectivity of a second-order mirror to 98% of the peak reflectivity. For a first-order mirror the same error would reduce the reflectivity to 94% of the peak reflectivity.

Measurements of absolute reflectivity, without the calibration using a multilayer mirror with known reflectivity, require monochromatic radiation, as stated in Sec. II. To obtain such monochromatic x-ray radiation the laser-plasma source was operated with ammonium hydroxide as target and a 1-μm-thick titanium filter to remove all lines except the $\lambda=2.879$ nm line from nitrogen ion emission. Figure 5 shows a reflectivity measurement of a W/B_4C multilayer sample. The multilayer period for this sample is 3.48 nm and the first and second order Bragg reflections appear at 24.99° and 56.32° grazing angle, respectively. The first-order reflectivity is 2.4% and the second-order reflectivity is 0.3%.

The accuracy of the reflectivity measurements depends on the ability of the titanium filter to suppress emission outside the $\lambda=2.879$ nm line, cf. Fig. 3. If the spectral filtering is not complete, the reference detector will record a small flux from, e.g., the short-wavelength nitrogen and oxygen lines, resulting in an underestimation of the reflectivity. Measurements performed in Refs. 12 and 15 indicate that the radiation is sufficiently monochromatic for this type of measurement, although we do not yet have a quantitative error analysis and the accuracy will be improved by calibration with multilayer mirror of known reflectivity. Finally, from Fig. 5 it can be seen that a reflectivity down to 0.1% may be measured due to the high signal-to-noise ratio.

The reflectometer described in this article is now involved in the development of multilayer condenser optics for water-window soft x-ray microscopy. Although the primary purpose of the reflectometer is directed towards this application, it is a general tool that may be used over a wide wavelength range. By combining suitable target liquids and free-standing metal foils for spectral filtering, spectrally tailored line emission for significant parts of the soft x-ray and EUV range may be produced.

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