

A W:B₄C multilayer phase retarder for broadband polarization analysis of soft x-ray radiation

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A W:B₄C multilayer phase retarder has been designed and characterized which shows a nearly constant phase retardance between 640 and 850 eV photon energies when operated near the Bragg condition. This freestanding transmission multilayer was used successfully to determine, for the first time, the full polarization vector at soft x-ray energies above 600 eV, which was not possible before due to the lack of suitable optical elements. Thus, quantitative polarimetry is now possible at the 2*p* edges of the magnetic substances Fe, Co, and Ni for the benefit of magnetic circular dichroism spectroscopy employing circularly polarized synchrotron radiation. © 2008 American Institute of Physics. [DOI: 10.1063/1.2841803]

INTRODUCTION

Soft x-ray synchrotron radiation with variable polarization is a sophisticated probe of the physical properties of matter. Many of the most advanced experiments take advantage of the inherently high degree of linear and/or circular polarization of such a source which is, in general, an elliptical undulator. However, the experimental determination of the polarization by, e.g., measuring the Stokes parameters of the monochromatic radiation becomes a necessity, because the beamline and monochromator may seriously influence and modify the polarization of the light source.¹

Polarimeters designed to deliver the four Stokes parameters of a source rely on a phase retarder and analyzer in combination.^{2–5} In the VUV range, the preferred retarders and analyzers are reflection optics,^{2,6–11} while in the soft x-ray region, they are transmission and reflection multilayers.^{3,12–17} Because of the enhancement of the multilayer performance near absorption edges, most multilayers have been designed to be used near the 2*p* absorption edges of the constituting materials (Cr:C, Cr:Sc, Mo:Si, Ni:Ti, and Ni:V). At best, they can operate at two distinct energies [e.g., Sc at 397 eV and Cr at 550 eV (Refs. 16 and 18)]. In the hard x-ray region, various standard monochromator crystals in Laue or Bragg geometry have been used.¹⁹ However, in the intermediate energy range between 600 eV and 4 keV, a complete polarization analysis has so far not been possible due to a lack of suitable phase retarding multilayers. This arises from the small multilayer period required (<1 nm) at these energies being of the same order as the interface roughness. Similarly, the unavailability of crystals which can be thinned down to less than 1 μm has prevented the adoption of the hard x-ray, single crystal approach to polarizing optics in this region.

The use of a nonresonant transmission multilayer as a phase retarder is reported in this paper. It describes new optical elements capable of a complete polarization analysis of

a synchrotron radiation beamline over an extended and continuous range of energies. This method of polarimetry determines the optical properties of the optical components and of the source in a single measurement—self-calibrating (primary) optical metrology.^{2–4}

This paper presents results from a W:B₄C phase retarder that gains its contrast from the broad W–N shell absorption (4*s*, 4*p*, 4*d*, 4*f*) between ~200 and ~500 eV. Since there are no strong resonances in the atomic scattering factors between ~500 and ~1800 eV in any of the constituents,²⁰ it acts as a nonresonant phase retarder. We present measurements of the transmission multilayer showing nearly constant phase retardance at energies between 640 and 850 eV.

EXPERIMENTAL

The measurements were carried out using the BESSY 6 axes polarimeter³ on the helical undulator beamline UE56/2 PGM1 at BESSY.^{21,22} The polarimeter houses two optical elements on two rotating stages, α (for the transmission multilayer polarizer) and β (for the reflecting multilayer analyzer). The angle of incidence θ_p and θ_a of these elements can be independently set to match the Bragg angle of the structure in question. There is also a two-theta arm on the reflection stage to hold and position the GaAsP-photodiode detector in two dimensions.

Two different reflection multilayers were used. The first one was manufactured by the X-ray Company, Russia and is a W:B₄C ($d=1.38$ nm, $\Gamma=0.44$, $N=350$) deposited on a solid silicon wafer substrate. It consists of 350 bilayers of W and B₄C with the bilayer spacing of 1.38 nm and a ratio of W thickness to total thickness of 0.44 (reported by the manufacturers). We shall use this shorthand notation to describe multilayers throughout the rest of this paper. The limit of 350 bilayers was chosen so that the transmission through the

multilayer would not be too small ($>0.2\%$) and that the rocking curve of either multilayer not be too small [$>0.2^\circ$ full width at half maximum (FWHM)].

The second, reflection W:B₄C multilayer ($d=1.23$ nm, $\Gamma=0.5$, $N=300$), was deposited onto a silicon wafer. With this period, the multilayer satisfies the Bragg condition at an angle of incidence of 45° near the Fe-2*p* absorption edge at 708 eV. This reflection multilayer when operated at the Brewster angle is essentially perfectly polarizing.²³ As the angle of incidence for the Bragg condition moves away from the Brewster angle, when changing the photon energy, the analyzer multilayer becomes less polarizing.

The transmission multilayer, W:B₄C [$d=1.38$ nm, $\Gamma=0.44$, $N=350$ (nominal)], was codeposited on to a silicon wafer with the first reflection multilayer described above. It is an unsupported multilayer on a $12 \times 12 \times 0.5$ mm³ Si frame with a central, circular 8 mm diameter unsupported region. The back etching process used to remove the silicon may have removed also some of the deposited layers—thus, the number of layers in the design is a maximum and may not represent the true structure of the manufactured optical element.

The atomic scattering factors for tungsten, boron, and carbon show no strong resonances between 500 and 1800 eV, thus, there is no resonant enhancement near absorption edges and the multilayer phase retarder is “non resonant.” In other words, provided that the angle of incidence of the multilayer is close to the Bragg condition, the device acts as a phase retarder independent of photon energy over a wide range of photon energies.

We report on three types of measurements.

The first was to measure the reflectivity of the reflection multilayers for linearly polarized light. The *s* and *p* geometries were selected by rotating the multilayers about the optical axis (the α and β axes, respectively).

The second was a transmission measurement of the free-standing multilayer as a function of angle of incidence θ_p for several photon energies in the range of 470–780 eV, again, for *s*- and *p*-linearly polarized light.

The third type of measurement was a full polarization measurement using both optical elements. Measurements were performed at four different photon energies between 640 and 850 eV. Measuring the reflected intensity as a function of α and β allows us to determine simultaneously the Stokes parameters of the source and the optical properties of the two multilayers.²³ At 639 eV, the W:B₄C (1.38 nm, $\Gamma=0.44$, $N=350$) reflection multilayer was used, with the Bragg angle near 45° close to the Brewster angle, thus, giving optimum polarizance. At the other energies, the W:B₄C (1.23 nm, $\Gamma=0.5$, $N=300$) multilayer was used. As mentioned above, this gives optimum polarizance at 708 eV.

RESULTS

The peak reflectivity of the analyzer (reflection) multilayer codeposited with the polarizer was found to be 4.6% for *s*-polarized light at 640 eV. The FWHM of the Bragg peak was found to be 0.26° . This is satisfactorily modeled using an interface roughness of 0.275 nm as the error

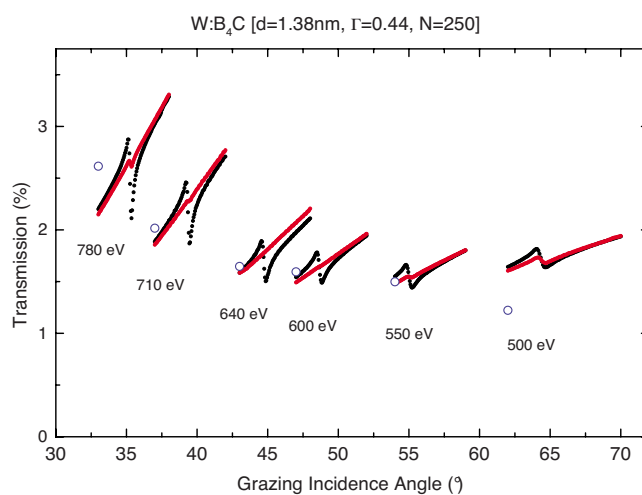


FIG. 1. (Color online) The measured, absolute transmission of the W:B₄C unsupported multilayer at various photon energies vs grazing angle. The small solid circles indicate transmission for *p*-polarized light (red) and *s*-polarized light (black). The large open circles indicate a best fit to the measured off-resonance transmission data using a simple model that ignores any interference effects due to the multilayer structure of the sample.

function using the program IMD.²⁴ Reflection data on the other multilayer have been previously reported.²³

The transmission of the polarizer (transmission) multilayer as function of its incidence angle at various photon energies between 500 and 780 eV is shown in Fig. 1 for both *s*- and *p*-linearly polarized light. Around the Bragg angle, the transmission is modulated differently for *s* and *p* polarizations.

To determine the bilayer spacing of the transmission multilayer, it is necessary to determine the Bragg angle for the multilayer. The wavelength calibration of the beamline and monochromator is assumed to be accurate, while there may be an error (offset) in the absolute value of the tilt angle θ_p . Thus, the offset in the measured θ_p and the multilayer spacing were fitted to the Bragg equation over the range of wavelengths and angles measured. The bilayer spacing was found to be 1.380 nm—in agreement with the manufacturer, who measured the Bragg angle with a Fe *L* $\alpha_{1,2}$ source. All data have been corrected for the angular offset.

The modeled transmission data shown in Fig. 1, were calculated from the reported W and B₄C absorption coefficients²⁵ and the amount of material in the light path as derived from the number of bilayers, the layer spacing, and the grazing incidence angle, and does not include interference effects. The reflection of the multilayer away from resonance is implicitly assumed to be negligible. The best fit to the data was achieved using 250 bilayers. Thus, it appears as if the back etching of the substrate has removed some of the deposited layers.

The ratio of *p* to *s* transmission of the multilayer T_p/T_s (polarizance) is another indicator of the quality of the multilayer and thus of its ability to act as a phase retarder. This ratio is presented in Fig. 2 (data points) along with data modeled using IMD (lines). The previously determined roughness (0.275 nm) and number of bilayers (250) are used in this calculation which agrees well with the on-resonance

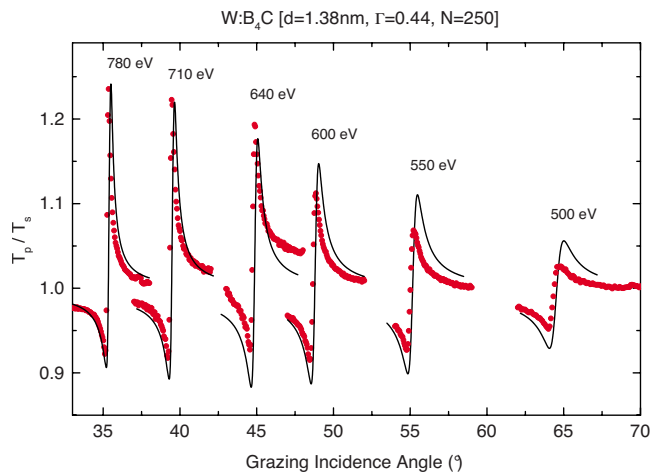


FIG. 2. (Color online) Ratio $T_p:T_s$ measured with linearly polarized light from the UE56/2 PGM1 undulator (points). Also shown are modeled results (lines).

transmission without further adjustments. We thus have a model describing the multilayer. The phase shift predicted by this model will be used later in the paper to extract the circular polarization from the data.

The polarization of the incident radiation was measured at 639, 708, 780, and 850 eV. These data sets consist of α and β scans at several closely spaced θ_p around the relevant Bragg angle.

Figure 3 shows a typical measurement and its fit. In the fitting, it is implicitly assumed that the angles of α and β are correctly aligned to each other as was regularly checked by visual inspection within $\pm 0.5^\circ$. It was further known that the background (dark) level of the detectors was negligible

The fit results in Fig. 4 show an almost constant maximum phase retardance, Δ of approximately -8° . The data for each photon energy are fit simultaneously, keeping the

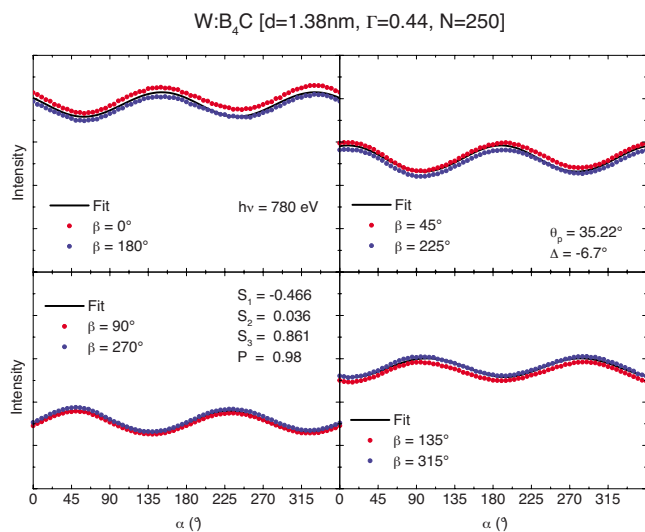


FIG. 3. (Color online) Measured polarimetry data and fit. The vertical scale is the same for all panels. The data are normalized to the ring current. By symmetry, the experimental curves should be identical. The deviation from the ideal case is due to mechanical misalignment and systematic errors (e.g., nonlinear brilliance changes of the light beam with beam current). The data were fit as presented without further correction.

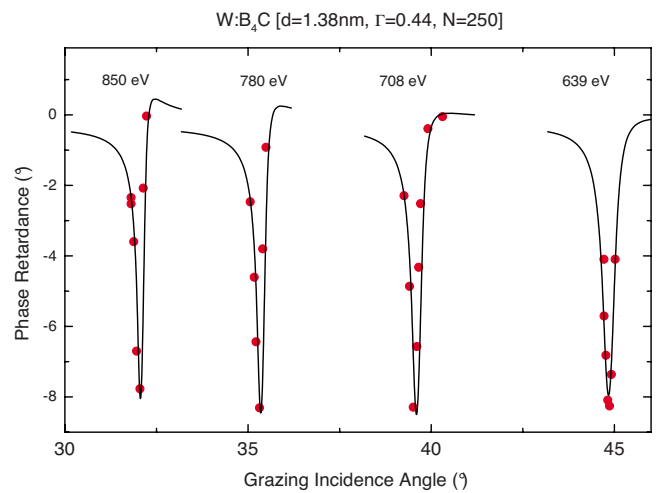


FIG. 4. (Color online) The phase retardance of the W:B₄C multilayers at four different photon energies. The points represent the measured phase retardance and the lines are modeled results using a roughness of 0.275 nm and 250 bilayers.

Stokes parameters and the analyzer parameters the same for all θ_p but allowing the phase retarder parameters to vary. The data were fit without any weighting.

The determination of Δ comes from two types of terms in the fitting equation.²⁻⁴ The first type is dependent upon the linear polarization, (S_1 and S_2) appears as $\cos(\Delta)$ and shows a modulation which scales approximately as $[1 \pm \cos(\Delta)]$. The second type is dependent upon the circular polarization (S_3) and shows a modulation which scales as $\sin(\Delta)$. For small Δ , $\cos(\Delta) \sim 1$ and $\sin(\Delta) \sim \Delta$. Thus, for small Δ , there exists a range of Δ , where it is not possible to uniquely determine Δ , from the fit alone, but where the product $S_3 \cdot \Delta$ can be determined.

We note that knowing the degree of polarization $P = \sqrt{(S_1^2 + S_2^2 + S_3^2)}$ is equivalent to knowing S_3 since S_1 and S_2 are obtained from the fit with a small uncertainty of ± 0.002 . The remaining intensity is shared between S_3 and the unpolarized fraction. Several fits were made each with an additional constraint in that the total polarization P was fixed ($P=1.0, 0.99, 0.98, \dots$). The value of P was selected that best reproduced the phase retardance, Δ , obtained from the multilayer model above. Care was also taken to ensure that there was one point measured at the inflexion point of the T_p/T_s versus θ_p curve which corresponds to maximum phase shift. The range in P over which Δ agrees with the model is indicative of the uncertainty of P . Thus, S_3 is determined.

WAVE (Ref. 26) was used to calculate the undulator output between 638 and 850 eV where the total polarization P was found to be very close to 1.0

A summary of the Stokes parameters measured on beamline UE56/2 PGM1 at BESSY is given in Table I. The results are compared to modeled data and include a data point measured with a Cr:Sc multilayer¹⁸ which has a much larger phase retardance (-27°).

IV. SUMMARY AND OUTLOOK

We have designed and characterized a nonresonant multilayer phase retarder on the basis of a freestanding

TABLE I. The Stokes parameters of beamline UE56/2 PGM1 at BESSY and the polarizing power of the analyzers used.

Energy (eV)	S_1	S_2	S_3	P	Polarizer material	R_p/R_s	W:B ₄ C analyzer (nm)	Undulator model (P)
571	-0.409	-0.008	0.890	0.98 ± 0.02	Cr:Sc	0.013	1.38	1.00
639	-0.482	-0.008	0.865	0.99 ± 0.02	W:B ₄ C	0.000	1.38	1.00
708	-0.456	0.046	0.866	0.98 ± 0.02	W:B ₄ C	0.000	1.224	1.00
780	-0.466	0.036	0.861	0.98 ± 0.02	W:B ₄ C	0.041	1.224	1.00
850	-0.465	0.040	0.816	0.94 ± 0.03	W:B ₄ C	0.102	1.224	1.00

W:B₄C multilayer used in transmission. This multilayer was used successfully to determine, for the first time, the full polarization vector at soft x-ray energies above 600 eV, which was not possible before due to the lack of optical elements. The data show a nearly constant (maximum) phase retardance of -8° between 640 and 850 eV in the vicinity of the respective Bragg angle. This is far away from ideal quarterwave behavior (90°), nevertheless, it is sufficient to fully and unequivocally characterize a beam of unknown polarization with an uncertainty of 3% relative error by taking highly redundant data.

Extending the working range of nonresonant multilayer phase retarders toward higher photon energies beyond the 850 eV achieved here faces different technical problems.

Absolute transmission will increase as long as the absorption decreases with photon energy. However, at the same time, the phase retarding capability decreases as the index of refraction approaches 1. Increasing the number of layers increases the phase retardance, but at the expense of a simultaneous increase in the demand for precision in the alignment and flatness of the transmission multilayer. This will place further constraints on the tolerances in the mechanical precision and stability of the ultrahigh vacuum rotation stages. For the measurements presented here, our apparatus worked near its limits of angular resolution.

The (maximum) phase retardance of the transmission multilayer as modeled at approximately 1200 eV and 22° grazing angle is -5° . W:B₄C (1.38 nm, $\Gamma=0.44$, $N=250$) with a roughness of 0.275 nm. This is near the limit of usefulness of this multilayer, both in terms of phase retardance and grazing angle. However, it remains to be proven that the phase shifting properties remain as observed at lower photon energies.

The nonresonant nature of the phase retardance means that data can be taken at any energy that is required, which was not previously possible due to the resonant behavior of other multilayer phase retarders used at lower energies. Hence, quantitative polarimetry is possible now at the $2p$ edges of the magnetic substances Fe, Co, and Ni, for the benefit of the magnetic circular dichroism-spectroscopy work done there.

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