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#### *MONOX* : a characterization tool for the X-UV range

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#### Abstract

A new laboratory apparatus devoted to the characterization of various devices for the X-UV range (100-5000 eV), such as mirrors, diffraction gratings, spectrometers or detectors is described. The apparatus includes open x-ray tubes as x-ray sources, a two-crystal monochromator for wavelength selection and a goniometer. Various examples of its use are presented : dispersive mode where the radiation coming from the x-ray tube is dispersed by the two-crystal monochromator, spectrometric mode where the goniometer is used as a plane x-ray spectrometer and reflectometric mode where a selected wavelength is used to perform absolute reflectivity measurements.

Keywords : Instrumentation for x-rays, 07.85.Fv X-ray reflectometry, 61.10.Kw X-ray optics, 41.50.+h Optical properties of multilayers, 78.67.Pt

Short title : MONOX : a characterization tool for the X-UV range

#### Introduction

The development of new sources in the X-UV range (100 - 5000 eV) as well as the optical components and the associated detectors requires the characterization of their performances in this spectral range [1]. In this context, we have developed a spectrogoniometer called MONOX, comprising a two-crystal monochromator and a  $\theta$ -2 $\theta$  goniometer. At the present time, the apparatus is operated with x-ray tubes as x-ray sources. However, it is also designed to work in a synchrotron radiation facility.

We present various examples of the ways to use the MONOX apparatus :

- 1) *Spectrometric mode* : Dispersion by the crystal or the multilayer in the goniometer of the polychromatic radiation emitted by a target located inside the x-ray tube, to deduce the physico-chemical state of this target from the analysis of its emission spectrum. In this case, the two-crystal monochromator is not used. This mode is analogous to the Wavelength Dispersive Spectrometry (WDS).
- 2) *Dispersive mode* : Dispersion by the two-crystal monochromator of the polychromatic radiation emitted by a target located inside the x-ray tube. In this case, MONOX is used as a tunable x-ray source in the soft x-ray and X-UV ranges.
- 3) « *Reflectometric* » mode : A wavelength selected by the two-crystal monochromator is used to perform absolute measurements of reflectivity with the help of the goniometer.

#### **Description of the apparatus**

The MONOX apparatus includes three different parts enclosed within their own vacuum vessel. A schematic presentation is shown in the Figure 1. The first part is the x-ray source, the second the monochromator and the third the goniometer. The whole apparatus works under a vacuum of the order of  $5.10^{-7}$  Torr ( $7.10^{-5}$  Pa) produced by turbomolecular pumps.

The x-ray sources

We use two open (windowless) Coolidge tubes equipped with exchangeable anodes. The radiation used for the measurements is either the Bremsstrahlung coming from a target of high atomic number (tungsten, gold, ...) or a characteristic emission line : K line of light elements (Be to Si), L $\alpha$  line of transition metals (Cr to Pd) and the elements up to Sb, and M $\zeta$  lines of the elements whose atomic number is between 37 and 42. Thus, these lines cover the spectral range between 12.9 nm (Rb M $\zeta$ ) and 0.34 nm (Sb L $\alpha$ ), that is to say from 96 eV to 3600 eV [2].

The maximal excitation conditions are 10 kV, 100 mA. The high voltage is delivered by a stabilized high-voltage supply. The current is regulated within 0.1 mA. The anode is water-cooled. In spectrometric mode, the sample to be analysed is placed on the anode.

The upper x-ray tube (Figure 1) is used for the two-crystal monochromator working in the dispersive and reflectometric modes. Otherwise, in spectrometric mode, the lower x-ray tube is used.

#### The monochromator

This device is used when working in the reflectometric mode to select a given photon energy, generally corresponding to a characteristic line in the spectrum of the source, or when working in the dispersive mode to perform the dispersion of the radiation emitted from the x-ray tube.

It is a two-crystal monochromator with separated dispersive elements and a fixed exit. The system that we have developed is different from the "classical" two-crystal systems [3]. The dispersive elements (mirror 1 and mirror 2 in Figure 1) are plane crystals or plane multilayer interferential mirrors (MIM) working according to the Bragg diffraction. The two elements are translated in parallel and oriented independently.

The mechanical scheme is given in the Figure 2. Two parallel translation units (T1 and T2) are put horizontally. Each unit can be rotated by a stepping motor (R1 and R2) whose angular resolution is equal to one millidegree. The horizontal distance X between the R1 and R2 units is given by  $X = h / tg(2\theta)$  where  $\theta$  is the Bragg angle and h the vertical distance between the T1 and T2 units. The adopted geometry enables a maximal distance X equal to 320 mm. Given the value of h, the Bragg angle can vary between 11° and 72°.

A mirror-holder containing the dispersive element, is fixed on each unit R1 and R2. In the spectrometric mode, a mechanism enables the retraction under vacuum of the dispersive element placed on R2, thus allowing the direct polychromatic beam to pass through the monochromator.

The dispersive elements generally used are MIM : W/C of small period (d = 2,54 nm) for the L emissions of the transition metals, Ni/C (d = 4,92 nm) for the K emission of carbon and La/B<sub>4</sub>C (d = 4,80 nm) [4] for the boron K emission. Artificial crystals (lead stearate, TlAP, KAP, ...) are also used for the K emissions of oxygen and nitrogen and eventually for boron. It should be noted that the bandwidth of the source available at the exit of the monochromator is generally determined by the natural width of the emission line (or emission band) when a multilayer mirror is used as a dispersive element.

An important point is the polarization of the radiation whose value is indispensable for the comparison with the theory in case of a measurement of the reflected power. The polarization rate depends on the Bragg angle and can be calculated assuming the radiation from the x-ray tube to be unpolarized. Near the Brewster angle (Bragg angle near  $45^{\circ}$ ) the quasi-monochromatic radiation is almost transverse electric.

#### The goniometer

This part consists of an adjustable entrance slit, a  $\theta$ -2 $\theta$ -type goniometer and a detection system. The goniometer comprises classically two coaxial rotation units. The first one carries the sample-holder containing the optical device (mirror, grating, ...) to be tested in reflectometric mode or the plane analysing element (crystal, multilayer, ...) in spectrometric mode. The second one moves the detection system. The two movements of rotation  $\theta$  and  $2\theta$  are independent to allow off specular diffusion studies. The angular resolution of the rotation movements is equal to one millidegree.

The sample-holder can move in three directions, two in the sample plane, the other perpendicular to it. The mechanical resolution of these movements is better than  $6.10^{-9}$  m. The distance between the sample-holder and the detector is adjustable to allow in particular the study of focussing optical devices (for example Bragg-Fresnel lenses). The detector can be moved perpendicularly to the specular direction for diffusion measurements.

The x-ray detection is performed either by a detector with a gaseous flux (argonmethane) working in the Geiger mode, by an X-UV diode (UVX 100 UDT) preceded by a screen opaque to the visible light, or by a channeltron system (Amptektron MD-501). In the present paper, the results are obtained with the Geiger counter.

#### **Examples of performances**

#### Spectrometric mode

As mentioned in Introduction, this mode can be viewed as WDS with a plane dispersive device (crystal, multilayer mirror or grating). Indeed this mode is an "opportunity" offered by MONOX but because no focusing dispersive device is implemented, the achieved detection sensitivity remains largely poorer than the one achieved with WDS commercially available system using a bent dispersive device. Nevertheless the use of a multilayer grating offers attractive possibilities illustrated by the following examples. We give two examples of spectrometric analysis obtained with a combination of MIM and a "high-resolution" multilayer mirror. The latter is a multilayer grating (MG) consisting of a lamellar grating etched to a multilayer structure. The principle as well as the recent developments of these MG can be found in references [6-8].

The first example presents the analysis of an iron target. Figure 3 shows the spectrum obtained with a MIM made of 150 Mo/B<sub>4</sub>C bilayers whose period is 6 nm, along with the spectrum obtained with the MG fabricated from this MIM. The peak at 8.61° corresponds to the Fe L $\alpha\beta$  doublet (705 eV), that at 8.89° to tungsten M emissions at the third diffraction order and the structure at 9.87° to the Fe Ll $\eta$  doublet (615 eV). The use of the MG allows the resolution of structures that are impossible to resolve with the MIM. For example, when the MIM is used, the W M emission is blended with the Fe L $\alpha\beta$  emission in a wide single peak around 8.9°. This does not permit to obtain a correct measurement of the iron intensity for quantitative analysis. It is observed that the same emission does not appear at the same angular position when analysed with the MIM or the MG, while both have the same period. This is due to the etching of the MIM, which lightened the structure and produced a MG having optical constants slightly different than those of the MIM [8].

The second example given in Figure 4 deals with a target made of a boron carbide film deposited onto a silicon substrate. With the MIM (the same as for the previous analysis) three lines are observed, whereas with the MG six structures are resolved and identified in Figure 6. The resolution of the C K emission in the first diffraction order from the O K emission in the second diffraction order is a real success for the etched multilayer gratings used as high-resolution dispersive elements in monochromators. It should be noted that the background is lowered when a MG is used (see also Figure 3). This improves the detection limit and allows detecting impurity elements such as silicon in the present example, which was omitted in the course of the MIM analysis.

We plan to produce in the near future cylindrically bent MG. Such devices would improve the luminosity of the spectrometer and after a modification of the mechanism of the goniometer, we hope to get a rather bright Rowland X-UV spectrometer for the detection of light chemical elements (boron, carbon, nitrogen, oxygen).

#### Dispersive mode

Figures 5 and 6 present the spectral distributions of two sources, obtained with the two-crystal monochromator equipped with W/C MIM. Figure 5 corresponds to the spectrum obtained with a copper target excited at 5 kV with a tungsten oxide deposit (coming from the sublimation of the thermoemissive filament). It displays the Cu L range (750-1100 eV). The Cu L $\alpha\beta$  (930 eV), Cu Ll $\eta$  (810 eV) et Cu L $\beta_{3,4}$  (1020 eV) emissions are observed, as well as a structure located at 910 eV corresponding the W M $\alpha$  emission in the second diffraction order.

Figure 6 is obtained with a tungsten target excited at 4 kV and displays the 500-600 eV range. The O K $\alpha$  emission band is observed at 525 eV. The oxygen comes from the tungsten oxide. We note that the full width at half maximum of the O K $\alpha$  emission is 12 eV, whereas it is determined to be 3 eV with a high-resolution bent crystal spectrometer [5].

Using the dispersive mode, it is possible to perform calibration of various x-ray devices such as detectors, filters, spectrometers and also the chemical characterizations of solid materials. Among the various kinds of solid surface characterization, MONOX provides an efficient tool for soft-x-ray reflection spectroscopy. The investigation method consists in recording the spectra of radiation reflected in the conditions of quasi-total reflection by solid surfaces around the absorption threshold of an element composing the medium. The evolution of the fine structures observed in the spectra, the so-called Refl XANES (reflection X-ray Absorption Near Edge Structure) versus the glancing angle provides information on the physico-chemical properties of the surfaces. We give an example of such a characterization in Figure 7. This figure shows the evolution of the reflection spectra in the vicinity of Si-K edge of a quartz crystal polished along the  $(11\overline{2}0)$  planes. The double-crystal monochromator is equipped with a pair of PET dispersive crystals, of chemical formula C(CH<sub>2</sub>OH), giving a resolving power about 6000 [9]. The spectra can be compared with the results reported in [10] obtained with a first generation synchrotron facility. The evolution of the spectra with the glancing angle corresponds to the phenomenon of anomalous reflection, which occurs in the domain of anomalous electronic scattering associated with an absorption edge. The details of this physics is beyond the scope of the paper but can be found in specialized papers [10,11,12]. Our results likely indicate that the surface of our sample is partly amorphous because the well-marked features observed in Ref. 10 are less pronounced in the present case. This structural state would be the result of the polishing process which tends to destroy the crystalline character of the surface [13].

The implementation of this mode for spectrometric analysis (especially for trace detection) is restricted by the low brightness of the double-crystal system due to the generally moderate reflectivity of the crystals and MIM in the X-UV domain. Indeed a double-crystal monochromator in the (+,-) configuration, like in our apparatus, has theoretically the same resolution than a single crystal monochromator, but a smaller output in terms of brightness because of the double reflection instead of a single one [14]. Moreover the attractive perspective of the association of a MIM with a MG is limited by the difficulty in matching the MIM with the MG. Indeed the effective period of a MIM is different from the effective period of the corresponding MG (i.e. the MG etched in the MIM) because of refractive effects [8]. This is the phenomenon observed previously in the Fe L $\alpha\beta$  spectrum. Since this mismatch depends on the photon energy [8], achieving the combination of a MIM with a MG working in a wide spectral domain is not a simple task.

#### Reflectometric mode

In this mode, a wavelength is selected by the double-crystal monochromator and is used to perform absolute reflectivity measurements with the help of the goniometer. Generally, the chosen wavelength corresponds to a characteristic emission line indicated in the "The x-ray sources" sub-section. As an example, we present the reflectivity measurements obtained using a Mo/Si MIM at the wavelength of the Cu L $\alpha\beta$  doublet (1.33 nm, 930 eV). A pair of W/C mirrors placed inside the two-crystal monochromator selects the line. The Cu L $\alpha$  and L $\beta$  lines are not resolved by the monochromator.

The Mo/Si mirror has a nominal period equal to 6.96 nm (2.92 nm Mo and 4.04 nm Si). Its reflectivity curve is presented in Figure 8. At each interface of the mirror, a transition layer is formed, whose nature (composition and thickness) has been determined by electron-

induced x-ray emission spectroscopy [15]. This spectroscopic analysis has shown that the transition layer is made essentially of  $MoSi_2$  having a mean thickness of 0.9 nm. Since the density of the various layers is lower than that of the bulk materials and the interfacial roughness is 0.75 nm, a theoretical model [16] can reproduce the experimental reflectivity curve (Figure 8).

#### **Conclusion and perspectives**

We have built a novel multi-purpose apparatus for the soft x-ray and X-UV range. At present, it works with classical laboratory sources but we plan to use it at synchrotron facility such as the future SOLEIL synchrotron. Besides the possibility of performing WDS-like measurements, this apparatus in the dispersive mode enables to study the spectra from a selected anode on a large spectral domain and also provides a tunable continuous source from the Bremsstrahlung of heavy materials. The latter possibility can be turned to account for the calibration of various devices and also for physico-chemical characterization as illustrated in this paper. But the low intensity of the Bremsstrahlung produced by the x-ray tubes in the energy range below 700 eV makes it difficult to use the dispersive mode with the present monochromator. Nevertheless the needs in this spectral range are numerous, in particular around the oxygen K edge to individually characterize oxides such as HfO<sub>2</sub>, ZrO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, ..., interesting the microelectronics industry [17]. In this context, to increase the signal, we plan to improve of the monochromator by using an optical system with a curved mirror to focus the divergent radiation coming from the x-ray tube and a plane grating to disperse this radiation under grazing incidence. The already existing mechanical components of the present two-crystal monochromator will be used for this improvement.

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#### **Figure captions**

Figure 1: Scheme of the MONOX apparatus. The lines represent the x-ray path. In dispersive or reflectometric mode, the upper x-ray tube is used. In spectrometric mode, the lower x-ray tube is used and the lower mirror (mirror 2) is retracted.

*Figure 2: Scheme of the two-crystal monochromator. Left : front view; right : side view.* 

*Figure 3: Spectrum of an iron target obtained with a*  $Mo/B_4C$  *multilayer mirror (dashed line) and with a "high resolution" multilayer grating (solid line).* 

Figure 4. Spectrum of a boron carbide film deposited onto a silicon substrate obtained with a  $Mo/B_4C$  MIM (dashed line) and with a "high resolution" MG (solid line).

*Figure 5 : Spectral distribution of the radiation emitted par a copper target and analysed by the double-crystal monochromator.* 

Figure 6: Spectral distribution of the radiation emitted by a tungsten target and analysed by the double-crystal monochromator in the range of the  $O K\alpha$  emission.

Figure 7: Reflection spectra of a (1120) quartz blade in the vicinity of the Si-K edge, for various orientation of the sample between 0.6 and 1.0°.

Figure 8. Experimental (open dots) and calculated (lines) reflectivity curves of a Mo/Si MIM. Solid line : perfect Mo/Si MIM; long dashed line : Mo/Si MIM with 0.9 nm thick  $MoSi_2$  interphases; dashed-dotted line : idem and densities equal to 90% of the densities of the bulk materials; dotted line : idem and 0.75 nm interfacial roughness.



Figure 1 : Scheme of the MONOX apparatus. The lines represent the x-ray path. In dispersive or reflectometric mode, the upper x-ray tube is used. In spectrometric mode, the lower x-ray tube is used and the lower mirror (mirror 2) is retracted.



*Figure 2 : Scheme of the two-crystal monochromator. Left : front view; right : side view.* 



Figure 3 : Spectrum of an iron target obtained with a  $Mo/B_4C$  multilayer mirror (dashed line) and with a "high resolution" multilayer grating (solid line).



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Figure 6 : Spectral distribution of the radiation emitted by a tungsten target and analysed by the double-crystal monochromator in the range of the O K $\alpha$  emission.



Figure 7 : Reflection spectra of a  $(11\overline{2}0)$  quartz blade in the vicinity of the Si-K edge, for various glancing angles between 0.6 and 1.0°.



Sample angle (°)

Figure 8 : Experimental (open dots) and calculated (lines) reflectivity curves of a Mo/Si MIM. Solid line : perfect Mo/Si MIM; long dashed line : Mo/Si MIM with 0.9 nm thick  $MoSi_2$  interphases; dashed-dotted line : idem and densities equal to 90% of the densities of the bulk materials; dotted line : idem and 0.75 nm interfacial roughness.