Spectrochimica Acta Part B 64 (2009) 756–760

Contents lists available at ScienceDirect

Spectrochimica Acta Part B

journal homepage: www.elsevier.com/locate/sab

Development of soft X-ray multilayer laminar-type plane gratings and varied-line-spacing spherical grating for flat-field spectrograph in the 1–8 keV region ☆

Masato Koike [a,*], Masahiko Ishino [a], Takashi Imazono [a], Kazuo Sano [b], Hiroyuki Sasai [c], Masatoshi Hatayama [d], Hisataka Takenaka [d], Philip A. Heimann [e], Eric M. Gullikson [e]

[a] *Japan Atomic Energy Agency (JAEA), Japan*
[b] *Shimadzu Emit Co. Ltd, Japan*
[c] *Shimadzu Corp., Japan*
[d] *NTT-AT Nanofabrication Co., Japan*
[e] *Lawrence Berkeley National Laboratory, USA*

ARTICLE INFO

Article history:
Received 10 November 2007
Accepted 19 May 2009
Available online 28 May 2009

Keywords:
Diffraction grating
Holographic grating
Multilayer
Spectrometer
Soft X-ray

ABSTRACT

W/C and Co/SiO_2 multilayer laminar-type holographic plane gratings (groove density $1/\sigma = 1200$ lines/mm) in the 1–8 keV region are developed. For the Co/SiO_2 grating the diffraction efficiencies of 0.41 and 0.47 at 4 and 6 keV, respectively, and for the W/C grating 0.38 at 8 keV are observed. Taking advantage of the outstanding high diffraction efficiencies into practical soft X-ray spectrographs a Mo/SiO_2 multilayer varied-line-spacing (VLS) laminar-type spherical grating ($1/\sigma = 2400$ lines/mm) is also developed for use with a flat field spectrograph in the region of 1.7 keV. For the Mo/SiO_2 multilayer grating the diffraction efficiencies of 0.05–0.20 at 0.9–1.8 keV are observed. The FWHMs of the measured line profiles of Hf-Mα_1(1644.6 eV), Si-Kα_1(1740.0 eV), and W-Mα_1 (1775.4 eV) are 13.7 eV, 8.0 eV, and 8.7 eV, respectively.

1. Introduction

There has been a pressing need in the nanotechnology community for developing new functional materials for obtaining a density of states, DOS, in the valence band with nm-scale spatial resolution. The DOS can be observed by X-ray emission spectroscopy, XES, both excited by soft X-rays or electron beams. The advantages of this method are its applicability to gas, liquid, and solid samples as well as the excellent selectability of the elements and symmetries. However, the obtainable emission signal is usually very small even if the sample is excited by the use of the high brightness synchrotron radiation sources. Also it is essential to avoid stray light due to the excitation beam and obtain signal-to-noise ratio high enough to observe the weak emission spectra. To fulfill these requirements Terauchi et al. [1–3] proposed and constructed a flat-field grating spectrograph for a transmission electron microscope (TEM) using a commercially available mechanically-ruled varied-line-spacing (VLS) grating [4,5]. This system is attractive because it enables one to observe the DOS by X-ray emission spectroscopy as well as the conduction band by electron energy-loss spectroscopy(EELS) without scarifying the spatial resolution of the TEM.

The flat-field grating spectrograph has no moving mechanism and is a very stable system that is essential for characterization with nm-scale spatial resolution. Maintaining these advantages we have initiated a project to develop the flat-field grating spectrograph to cover an energy region of 1–2 keV. In the first stage of the project we have fabricated multilayer laminar-type holographic plane gratings and confirmed their high diffraction efficiencies. Based on the experimental results we designed a multilayer VLS laminar-type spherical grating for the flat-field grating spectrograph [6,7]. The purpose of this paper is to describe the performance of the multilayer laminar-type holographic gratings in terms of diffraction efficiency and spectral resolution.

2. W/C and Co/SiO_2 multilayer plane gratings

To assess the diffraction efficiency experimentally we designed and fabricated W/C and Co/SiO_2 multilayer plane gratings. The material pairs were chosen through a comparative study on the reflectivity of the various multilayer material pairs in the energy region of 1–8 keV using optical constants published by Henke et al [8]. The two multilayers were deposited on two laminar-type holographic plane gratings, respectively. The material of the grating blanks and grating

---

☆ "This paper was presented at the 19th "International Congress on X-ray Optics and Microanalysis" (ICXOM-19) held in Kyoto (Japan), 16–21 September 2007, and is published in the Special Issue of Spectrochimica Acta Part B, dedicated to that conference".

* Corresponding author.
E-mail address: koike.masato@jaea.go.jp (M. Koike).

0584-8547/$ – see front matter © 2009 Elsevier B.V. All rights reserved.
doi:10.1016/j.sab.2009.05.013

constant were synthetic quartz and 1/1200 mm respectively. For the W/C (or Co/SiO$_2$) multilayer grating the groove depth (h) was 3 nm (or 4 nm) with a land-to-period ratio (a/D) of 0.45 (or 0.50). The nominal multilayer period length was 6.64 nm and the ratio of (W or Co thickness)/(periodic length) was 0.4. The number of periods were 50 and 30 for the W/C and Co/SiO$_2$ multilayers, respectively. The resultant multilayer period lengths were found to be 6.66 nm and 6.62 nm for the W/C and Co/SiO$_2$ multilayers, respectively, as the result of an analysis of the Bragg peaks obtained by an X-ray diffractometer with a Cu-Kα (8.05 keV) source.

The diffraction efficiency measurements were performed at the BL5.3.1 (2.5–8.0 keV) and BL6.3.2 (0.6–1.2 keV) at the Advanced Light Source of Lawrence Berkeley National Lab., USA, and the evaluation beamline for soft X-ray optical elements BL-11 (0.6–1.5 keV), operated by Japan Atomic Energy Agency, at the SR Center of Ritsumeikan Univ., Shiga, Japan as well as at an X-ray diffractometer equipped with a Cu-Kα source at 8.05 keV.

Fig. 1 shows the measured diffraction efficiencies of the W/C and Co/SiO$_2$ multilayer gratings. For the Co/SiO$_2$ grating the 1st order's diffraction efficiencies of 0.41 and 0.47 at 4 and 6 keV, respectively, and for the W/C grating 0.38 at 8 keV were observed [9]. To assess the performance of the fabricated grating the theoretical diffraction efficiency was calculated by use of a simulation code [10] based on a differential method. The difference between the measured and theoretical efficiencies should be attributed to the surface roughness at the top and at the boundaries of the multilayer as well as exotic layers generated by inter-diffusion. To taking into account these effects we applied the Debye–Waller factor [11] to the theoretical diffraction efficiency. The comparison of the measured and theoretical efficiencies thus obtained suggests that both multilayer gratings have imperfections corresponding to an rms roughness of ~1 nm.

From these results our design and fabrication techniques to produce multilayer plane gratings were found to be ready for more advanced multilayer gratings.

3. Fabrication of Mo/SiO$_2$ multilayer VLS spherical grating

3.1. Flat field spectrograph and grating design

Schematic diagram of the soft X-ray flat-field spectrograph is shown in Fig. 2. In this figure a holographic recording system which is used to record the VLS grooves pattern on the spherical grating blank is also illustrated in gray lines.

A spherical holographic grating G accepts the light emerging through an entrance slit E. The light diffracted by G is focused on an image plane Σ that is parallel to x-axis and is located at a distance of L from the grating normal at the vertex O. The diffracted principal ray of wavelength λ in mth order intersects Σ at a point B_0. The distance OB$_0$ is denoted as r_0'.

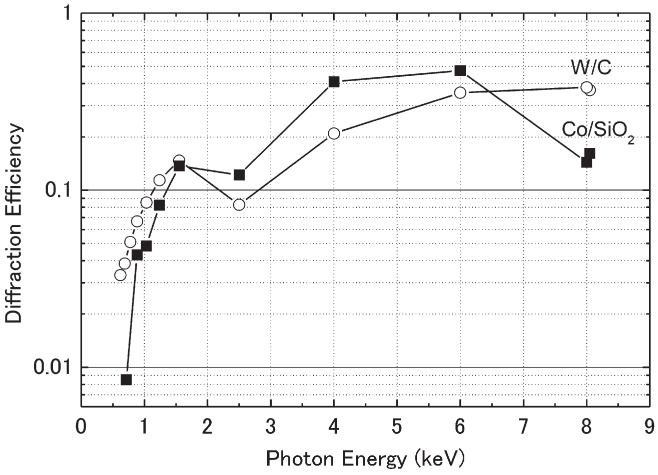

Fig. 1. Diffraction efficiencies of the W/C and Co/SiO$_2$ multilayer plane gratings.

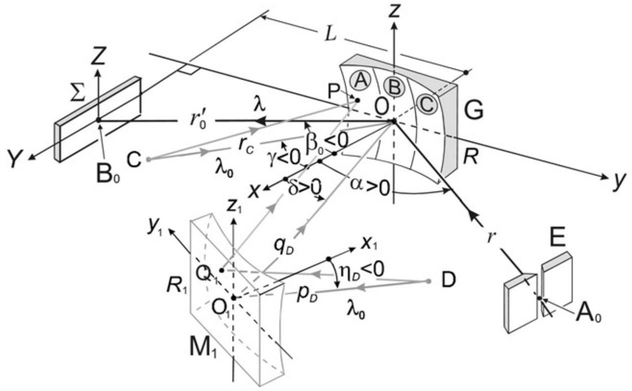

Fig. 2. Schematic diagram of the soft X-ray flat-field spectrograph and holographic recording system. For detail see text.

Fig. 2 also shows an aspheric wave-front recording optical system consisting of two coherent point sources, C and D, of wavelength λ_0, an auxiliary spherical mirror, M_1, and a spherical grating blank, G. A detailed description of the system and the definitions of quantities are given in Ref. [12]. The equation for the interference fringes formed on the surface of G, i.e., the equation for the grating grooves is expressed as

$$n\lambda_0 = [CP - (DQ_1 + Q_1P)] - [CO - (DO_1 + O_1O)]. \tag{1}$$

In Eq. (1), n is the groove number at the point $P(\xi,w,l)$ counted from the zeroth groove that passes through O. The sign of n is positive or negative according to whether the central point $(\xi,w,0)$ of the nth groove lies on the positive or negative y-axis.

The point Q_1 on M_1 is determined so that the ray $D-Q_1-P$ satisfies Fresnel law. The groove number n is expressed by a power series of the coordinates w and l of a point 1 P on the nth grooves as

$$n\sigma = w + \Gamma\Bigg[\frac{1}{2}\left(n_{20}w^2 + n_{02}l^2 + n_{30}w^3 + n_{12}wl^2\right) + \frac{1}{8}\left(n_{40}w^4 + 2n_{22}w^2l^2 + n_{04}l^4\right) + \cdots\Bigg], \tag{2}$$

where σ is the effective grating constant, $\Gamma = \sigma/\lambda_0$ for the holographic gratings and $\Gamma = 1$ for the ruled gratings. The explicit expressions of the groove parameters Γn_{ij} for the holographic grating recorded with aspheric wave-front recording optics and for the ruled grating are given in Refs. [12,13], respectively. The effective grating constant σ for holographic gratings is given by [14]

$$\sigma \equiv 1/[\partial n/\partial w]_{w=l=0} = \lambda_0/(\sin\delta - \sin\gamma). \tag{3}$$

The instrumental parameters of the standard soft X-ray flat-field spectrograph are as follows (see Fig. 2 [4–6]: r (distance A_0O) = 237.0 mm; D (distance between O and Σ) = 235 mm; W (effective width) = 46 mm; H (effective height) = 26 mm; m (spectral order) = +1; α (angle of incidence) = 87.0°. In designing the grating we assumed σ (effective grating constant) = 1/2400 mm, λ_0 (wavelength of the recording laser) = 441.6 nm, and λ_i (i = 1,2, and 3, design wavelengths) = 0.754 nm (1644.6 eV), 0.713 nm (1740.0 eV), and 0.698 nm (1775.4 eV). The wavelengths, λ_1, λ_2, and λ_3, are relevant to emission lines of hafnium Mα_1, silicon Kα_1, and tungsten Mα_1.

The following parameters were determined for the grating blank and recording system by means of the analytical design method (see Fig. 2) [13]: R (radius of curvature of G) = 4833.00 mm, r_C (distance CO) = 554.00 mm, R_1 (radius of curvature of M_1) = 400.00 mm, p_D (distance DO_1) = 1073.00 mm, q_D (distance O_1O) = 250.00 mm,