G Model
APSUSC-27577; No. of Pages 7
ARTICLE IN PRESS
Applied Surface Science xxx (2014) xxx–xxx
Contents lists available at ScienceDirect
Applied Surface Science
journal homepage: www.elsevier.com/locate/apsusc

Optical and structural characterization of the $Co/Mo_2C/Y$ system

Yanyan Yuan[a,b,*], Karine Le Guen[a,b], Jean-Michel André[a,b], Zhanshan Wang[c], Jingtao Zhu[c], Christian Mény[d], Philippe Jonnard[a,b]

[a] *Sorbonne Universités, UPMC Univ Paris 06, Laboratoire de Chimie Physique-Matière et Rayonnement, 11 rue Pierre et Marie Curie, F-75231 Paris cedex 05, France*
[b] *CNRS UMR 7614, Laboratoire de Chimie Physique-Matière et Rayonnement, 11 rue Pierre et Marie Curie, F-75231 Paris cedex 05, France*
[c] *Institute of Precision Optical Engineering, Department of Physics, Tongji University, Shanghai 200092, China*
[d] *Institut de Physique et Chimie des Matériaux de Strasbourg, UMR 7504 CNRS-Université De Strasbourg, 23 rue du Loess, 67034 Strasbourg, France*

ARTICLE INFO

Article history:
Received 19 December 2013
Received in revised form 28 February 2014
Accepted 25 March 2014
Available online xxx

Keywords:
$Co/Mo_2C/Y$ multilayer
Interface analysis
Thermal behaviour
Optical properties
Nuclear magnetic resonance spectroscopy
X-ray reflectivity
X-ray diffraction

ABSTRACT

We study the thermal behaviour of a tri-layer multilayer, designed by inserting a third material, yttrium, into the previously studied Co/Mo_2C system. The system is designed to work at near-normal incidence at the wavelength of 14.1 nm. The theoretical reflectivity of Co-based multilayer ($Co/Mo_2C/Y$ system) is improved up to 54% after the addition of yttrium. Two types of multilayers with different orders of yttrium layer are deposited: $Co/Mo_2C/Y$ and $Co/Y/Mo_2C$. The samples are annealed up to 600 °C. The multilayers were characterized using hard x-ray and extreme ultraviolet reflectivity, nuclear magnetic resonance (NMR) spectroscopy and x-ray diffraction (XRD). The results show that the reflectivity of the $Co/Mo_2C/Y$ multilayer is 27.5% at near normal incidence around 14.6 nm for as-deposited sample, and then it decreases gradually after annealing up to 600 °C. A significant period compression is observed from 300 °C annealing and above. The $Co/Y/Mo_2C$ multilayer shows low reflectivity, less than 2.5%. NMR spectra reveal that the pure Co layers are completely mixed with other elements since there is no signal from ferromagnetic Co in the annealing samples of the $Co/Mo_2C/Y$ multilayer and all $Co/Y/Mo_2C$ samples. Based on the NMR and XRD results, we fit the EUV data for both multilayers with two different models in one period taking into account the formation of the interfacial compounds.

Introduction

Multilayer mirrors for the soft x-ray and extreme ultraviolet ranges consisting of two materials with high and low optical constants are studied since a long time. The potential applications include telescope [1], projection lithography [2,3], synchrotron beamline [4] and x-ray microprobe [5]. Bi-layer multilayers have been studied for many years and some of them have achieved high optical performance and good thermal stability. Tri-layer multilayers, resulting from the addition of a third layer into bi-layer [6–9], are proposed with the aim of improving the optical performance and thermal stability.

The optical performance of Co/Mo_2C multilayer has been recently studied in our group [10] (experimental reflectivity is 25% @ 11° and 1.59 nm). The optical performance is related to the chemical and physical environments at the interfaces. In the bi-layer system, it is not possible to know clearly which interface, Co-on-Mo_2C or Mo_2C-on-Co, has more influence on the optical performance. Thus in order to distinguish these two interfaces contributions and improve the optical performance of multilayer, we inserted a third layer, yttrium, into Co/Mo_2C multilayer stack. Two types of multilayers are formed: $Co/Mo_2C/Y$ with interfaces Mo_2C-on-Co, Y-on-Mo_2C and Co-on-Y; $Co/Y/Mo_2C$ with interfaces Co-on-Mo_2C, Y-on-Co and Mo_2C-on-Y.

Yttrium is a promising material for application in the multilayer and can provide good thermal stability. Indeed, Bosgra et al. have reported that 0.2 nm thick Y layer significantly reduced the silicon diffusion towards Mo in the $B_4C/Mo/Y/Si$ system [11]. A theoretical study about the Y-based multilayers, for example, Pd/Y, Ag/Y, Mo/Y, Nb/Y, has shown that the combination of yttrium and these materials could give normal incidence peak reflectivity ranging from 50% to 65% in the range of 8-13 nm [12]. The study of Mo/Y multilayer demonstrated that this stack is stable up to 400 °C [13].

In the present work we study the optical performance of the $Co/Mo_2C/Y$ tri-layer multilayer with different interface orders. The order of the Y layer in the system cannot only significantly affect

* Corresponding author at: Sorbonne Universités, UPMC Univ Paris 06, Laboratoire de Chimie Physique-Matière et Rayonnement, 11 rue Pierre et Marie Curie, F-75231 Paris cedex 05, France Tel.: +33 0 1 44 27 62 22; fax: +33 0 1 44 27 62 26.
E-mail addresses: yanyan.yuan@upmc.fr, yyyuan106@gmail.com (Y. Yuan).

http://dx.doi.org/10.1016/j.apsusc.2014.03.176
0169-4332/© 2014 Elsevier B.V. All rights reserved.

Please cite this article in press as: Y. Yuan, et al., Optical and structural characterization of the $Co/Mo_2C/Y$ system, Appl. Surf. Sci. (2014), http://dx.doi.org/10.1016/j.apsusc.2014.03.176

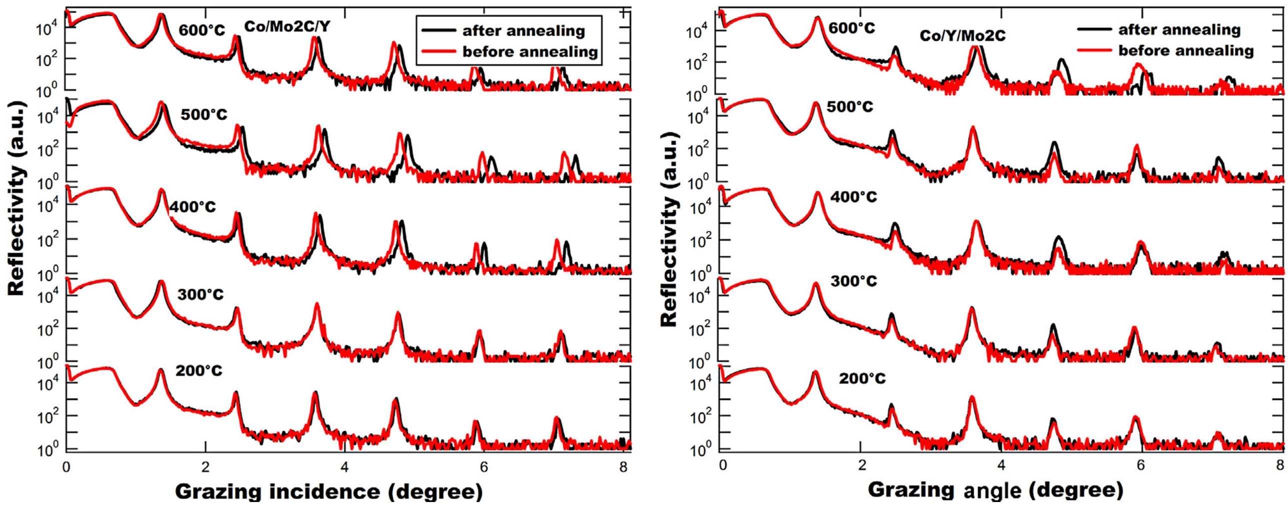

Fig. 1. Reflectivity curves at 0.154 nm of the $Co/Mo_2C/Y$ and $Co/Y/Mo_2C$ systems before and after annealing as a function of the grazing angle. For the sake of clarity, spectra of the sample annealed at 200, 300, 400, 500 and 600 °C as well as corresponding as-deposited samples are shifted vertically.

the reflectivity but also the interfacial diffusion, as reported in the Co/Mg system with an insertion of Zr layer [9]. We observed that the addition of the third layer of yttrium improves the optical performance of the Co/Mo_2C multilayer (45% theoretical reflectivity). The two designed systems $Co/Mo_2C/Y$ and $Co/Y/Mo_2C$ with different interface orders present the theoretical reflectivity of 54% and 11% at near normal incidence, respectively. The reason for the significant difference of theoretical reflectivity of these two systems is the difference of the optical path of the incident beam into the multilayer stacks. In order to assess how the two sets of interfaces affect the optical performance and the interfacial diffusion, we fit the reflectivity curves performed in the EUV range to provide the structural parameters and the interface width of the multilayer. Results are complemented by nuclear magnetic resonance (NMR) spectra and x-ray diffraction (XRD) experiments.

Experimental

Sample fabrication

The $Co/Mo_2C/Y$ and $Co/Y/Mo_2C$ systems were deposited by direct current magnetron sputtering. The substrates were sliced and polished Si (1 0 0) wafers, with surface roughness of 0.4 nm (rms) determined by atomic force microscopy in the range of 10^{-2}–10 μm^{-1} spatial frequencies. The designed period is 7.71 nm and the thickness of Co, Mo_2C and Y layer was 1.00 nm, 2.67 nm and 4.04 nm, respectively. The base pressure was 10^{-5} Pa before deposition. The working gas was argon (99.999% purity) at a constant working pressure 0.1 Pa. The number of periods is 30. There were six 10 × 10 mm^2 substrate sections cleaved from a single wafer for each system. Then six multilayers were deposited separately in the same conditions. The first layer on the substrate was the Co one. With our notation, the layers are indicated from the substrate toward the surface. A 3.5 nm thick B_4C capping layer was deposited to prevent oxidation. Five as-deposited samples of each system were annealed at 200, 300, 400, 500 and 600 °C during 1 h under high vacuum to study their thermal behaviour.

Sample characterization

The quality of the multilayer was checked through grazing incidence x-ray reflectometry (GIXR) using Cu Kα wavelength (0.154 nm) in the θ–2θ mode. The angular resolution is 5/1000°. The fit of GIXR data allowed the determination of the thickness and roughness of the different layers in each structure and also the estimation of the density of the Co, Mo_2C and Y layers.

In the extreme ultraviolet range, the reflectivity measurements were performed with *s*-polarized radiation at the application wavelength of 14.6 nm on the BEAR beamline in the Elettra synchrotron radiation facility [14]. The photon energy is calibrated by the Si $2p_{3/2}$ binding energy at 99.2 eV. The reflectivity curves were obtained at the angle of 4° off-normal incidence. The fits of reflectivity data were performed with IMD software [15] and provided information about interdiffusion.

In order to probe the chemical state of the Co atoms within the multilayer, we analyzed the samples by using zero field NMR spectroscopy. To enhance the sensitivity, the testing temperature was set at 4.2 K. The NMR spectra represent the distribution of the Co atoms as a function of their resonance frequency, that is to say the hyperfine field experienced by the Co nuclei. The NMR resonance frequency is sensitive to the local environment of the probed atoms: the number, nature and symmetry of atoms in its neighbourhood [16,17].

X-ray diffraction is a well-established technique for determining the crystal structure of thin films. To identify the phase present in our systems as a function of the annealing temperatures, we performed x-ray diffraction of the $Co/Mo_2C/Y$ (as-deposited, 300 and 600 °C) and $Co/Y/Mo_2C$ (as-deposited) samples with Cu Kα radiation (0.154 nm).

Results and discussion

Grazing incidence x-ray reflectometry at 0.154 nm

We show in Fig. 1 the GIXR curves of the $Co/Mo_2C/Y$ and $Co/Y/Mo_2C$ systems prior and following annealing. Six well-defined peaks are observed for both systems, indicating that these multilayers possess a well-defined periodic structure. After 400 °C annealing the peak positions of the $Co/Mo_2C/Y$ multilayers shift toward high angle, which implies a period decrease according to the Bragg law. Concerning the $Co/Y/Mo_2C$ systems, it is observed that the Bragg peaks position of the 600 °C annealed sample generate a significant shift toward high angle. This is due to the variation of period upon annealing.

We performed the fit of the GIXR data with Bede Refs software (genetic algorithm) [18] to estimate the structural parameters (thickness, roughness and density of each layer) using a tri-layer model (no interlayer is taken into account). A good agreement

Please cite this article in press as: Y. Yuan, et al., Optical and structural characterization of the $Co/Mo_2C/Y$ system, Appl. Surf. Sci. (2014), http://dx.doi.org/10.1016/j.apsusc.2014.03.176