Fraunhofer Institut Werkstoff- und Strahltechnik

Preprint

High-Precision nm-Coatings for EUV and X-Ray Optical Applications

Proceedings of NanoFair (2002)

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November 2002

St. Braun, Th. Böttger, R. Dietsch, Th. Foltyn, P. Gawlitza, Th. Holz, H. Mai, M. Menzel, M. Moss, J. Schmidt, L. van Loyen, D. Weißbach, and A. Leson: *High-Precision nm-Coatings for EUV and X-Ray Optical Applications*, Proc. NanoFair (2002)

High-Precision nm-Coatings for EUV and X-Ray Optical Applications

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Abstract

In this paper we will give an overview of the preparation, characterization and typical results of nm-multilayer coatings prepared by **p**ulsed **l**aser **d**eposition (PLD) and **m**agnetron **s**putter **d**eposition (MSD). We have achieved particularly outstanding results for the Ni/C and Mo/Si material systems, e.g. **e**xtreme **u**ltra**v**iolet (EUV) reflectances of >71% with Mo/Si. The thickness deviations of the multilayer period on flat and curved substrates with macroscopic dimensions (150mm diameter) are reduced down to <0.05%. The standard deviation of the thicknesses from run-to-run is <0.2%.

1 Introduction

Multilayers with up to 1000 periods of layers with nm-thicknesses are used as reflectors for various EUV and X-ray optical applications. Besides the well-known laboratory applications like diffractometry, reflectometry and fluorescence analysis, the extreme ultraviolet lithography (EUVL), as the technology for the production of next-generation integrated circuits with pattern sizes <50nm, is one of the main driving forces for the development of nm-multilayers. However, independent of the application in mind, the multilayers have to meet stringent requirements concerning reflectivity, reproducibility, uniformity, resolution and stability.

In order to fulfill the crucial specifications needed, we use two ultra-high vacuum coating techniques: pulsed laser deposition and magnetron sputter deposition, each with special physical properties. Depending on the material system of the multilayer stack we can choose the most suitable deposition method. Moreover, we can combine both techniques – PLD and MSD - because they are available in one and the same machine.

2 Preparation of nm-Coatings

The deposition methods used for the preparation of multilayer coatings with single layer thicknesses in the nm-range and below have to meet stringent requirements concerning the quality of the layers. The following points are the most important ones:

- high reproducibility of the layer thicknesses throughout the whole stack,
- smooth layers and smooth interfaces between adjacent layers,
- sharp interfaces, i.e. steep concentration gradients at the transition from one layer material to the next,
- deposition of nm-layers having optical parameters close to the corresponding solid state values.

Four deposition techniques fit these requirements particularly well and are widely used: magnetron sputter deposition, ion beam sputter deposition, pulsed laser deposition and electron beam evaporation. Each of these techniques has its specific advantages and drawbacks. In this work we will focus on the MSD and PLD, which are both available in our laboratories.

2.1 Magnetron Sputter Deposition

The most decisive parameter which influences the quality of the coatings is the energy distribution of the particles coming from the target and being deposited on the substrate. Energies which are too low will result in rough interfaces because of the lack of surface mobility of the condensing particles, whereas energies which are too high will lead to intermixing on the interfaces connected with a decrease of the optical contrast between the layers within the stack. Typical energies of particles generated by MSD are in the range of 1-10eV. According to structure zone models, the deposited layers are in a dense or dense polycrystalline state [1]. The typical columnar layer growth can be reduced by the proper choice of deposition parameters (e.g. low sputter gas pressures) and geometries (e.g. optimization of target-substrate distance).

The set-up of our MSD system is similar to that used by other groups [2,3]. Inside an UHV sputter chamber with a square footprint, four magnetron sources are mounted along the diagonals (fig. 1). Typical operation parameters are: base pressure: $<2\cdot10^{-8}$mbar; sputter gas: argon with working pressures of $0.8\text{-}1.5\cdot10^{-3}$mbar; target size: $304.8\text{x}88.9\text{x}6.35\text{mm}^3$;

substrate size: 150mm diameter; target-substrate distance: 50-100mm; magnetron source power: 50-300W. The thicknesses of the individual layers are controlled by altering the angular frequencies ω_R depending on the angle ϕ relative to the starting position. This enables the accurate deposition of multilayers with uniform and axially symmetric thickness profiles on flat or curved substrates. The deposition of freely defined 2-dimensional thickness profiles requires the application of special masks.

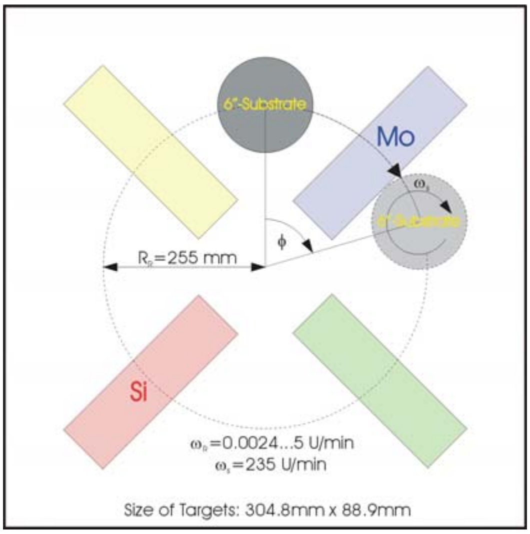


Fig. 1: Scheme of the target-substrate arrangement inside the sputtering chamber. In this example, the equipment for the deposition of Mo/Si multilayers is shown. The two positions between Mo and Si are occupied by barrier, buffer or capping layer materials.

2.2 Pulsed Laser Deposition (PLD)

The interaction of a focused laser beam and the solid state surface of the target leads to the emission of a plasma consisting of ions, atoms, clusters, electrons and photons of the ablated material. These particles with energies of up to several hundred eV condense on the substrate surface and form a thin film. Due to the high energy of the particles, the surface mobility on the substrate is also high. Consequently, the growing nm-layers are extremely smooth. According to the structure zone model, PLD prepared layers are in a vitreous amorphous state far from the thermodynamical equilibrium.

In order to increase the uniformity of the layers, a special target-substrate arrangement has been developed (fig. 2, [4]). Recently, this technology has been scaled up to substrate diameters of 150mm. Typical parameters of the PLD process are: base pressure: $<10^{-8}$mbar; working pressure: $<3\cdot10^{-8}$mbar; laser wavelength: 1064nm, 532nm, 355nm; laser pulse duration: 4-10ns, laser frequency: 1-10Hz; laser power density on the target surface: 10^8-10^9 W/cm^2; target length: up to 165mm; target diameter: 20mm.

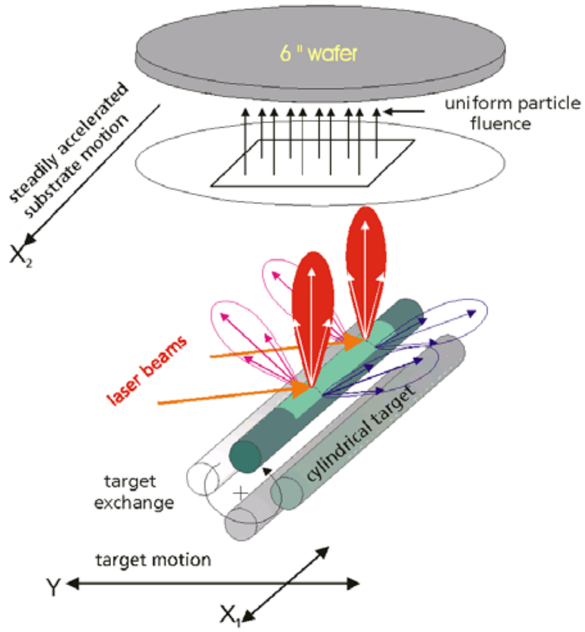
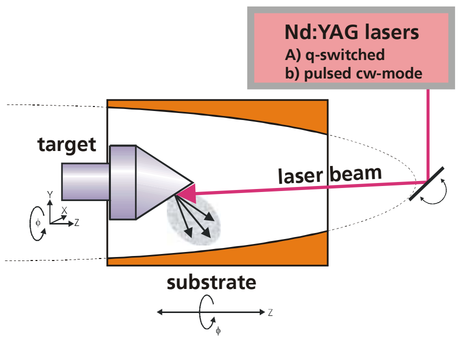
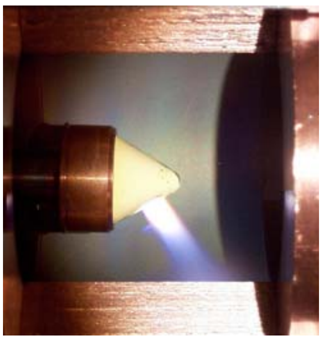


Fig. 2: Principle of thin film deposition by Pulsed Laser Deposition: Targets of cylindrical shapes are moved in two directions X_1 and Y. As a result of the Y motion, the plasma plume pivots and forms a uniform layer in the Y direction. The substrate movement parallel to X_2 is superposed and results – depending on the motion profile - in an uniform or a gradient layer thickness.

2.3 Internal Coating by PLD

The application of classical deposition methods available for internal coatings is often limited to substrates with inner diameters larger than a critical size or to coatings with no necessity of uniform thickness distributions. With the PLD, a uniquely small source is available which can succesfully be used to coat inner walls of substrates having diameters of down to 5mm with uniform layers. A schematic view is shown in fig. 3: in contrast to the standard PLD technique as described in section 2.2, the laser beam is directed to a conic target, which moves inside the substrate [5]. Hence, the inner diameter of the substrate is mainly limited by the cone size and the laser beam dimensions. The relative movement of target and substrate enables the deposition of uniform layers even on non-cylindrical substrates with rotational symmetry.





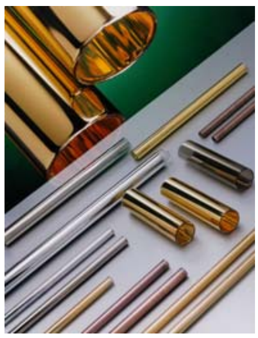


Fig. 3: Left: Schematic view of the inner wall deposition by PLD. Middle: Photograph of the target and the plasma plume inside a model segment during the deposition. Right: Examples for internal coated tubes: substrate: glass, coatings: Ni, Au, Pd, DLC, …

Interesting applications are e.g. total reflection X-ray and EUV optics, where the requirements concerning the surface quality are much more relaxed compared to the situation for multilayer mirrors. Since e.g. EUV radiation can be total reflected at grazing angles of up to 20°, ellipsoidal or parboloidal substrate surfaces coated with molybdenum single layers can be used to collect divergent radiation from EUV point sources. Depending on the application, surface roughnesses of up to σ_{rms}=10nm are tolerable. Therefore, availability and costs of such substrates are not restraints for the practical use of this kind of optics.

3 Metrology on nm-Coatings

The preparation of high-quality multi and single layer coatings is only possible if accurate metrology tools are routinely available, which enable the immediate inspection of the layer properties. Therefore the continued improvement of these measurements is necessary. At present, we are using Cu-, Mo-, Cr-Kα reflectometry to perform these inspections. Additionally, we are developing an EUV reflectometer for the wavelength range λ=10-16nm, which permits the metrology of Mo/Si multilayer coatings for EUV applications at the working wavelength.

3.1 Cu-Kα Reflectometry

Cu-Kα reflectometry is the standard method to characterize nm-multi and -single layers. From the measurement of the specular reflection depending on the grazing angle, information about reflectivity, stack regularity, multilayer period, layer thickness ratio and interface quality can be deduced. In order to decrease measurement time and to increase measurement accuracy, a new concept of the X-ray optical path has been developed [6]. With the so called twin-Goebel mirror (TGM) arrangement two parabolic shaped X-ray mirrors coated with Ni/C gradient multilayers are combined. The divergent beam of a commercial X-ray source is collimated by the primary mirror, the resulting parallel beam is then used to scan the sample and the reflected parallel beam is focused at the detector entrance slit (Fig. 4).

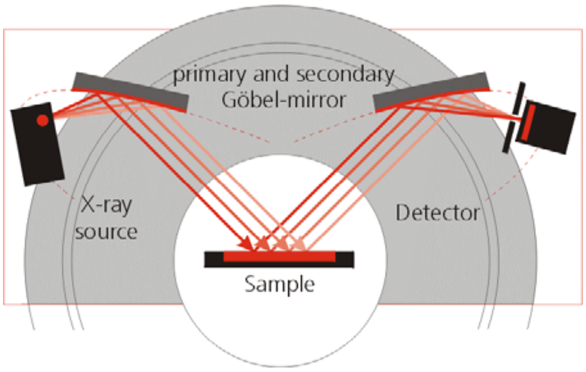


Fig. 4: Twin Goebel mirror arrangement: two parabolic X-ray mirrors coated with Ni/C multilayers are combined.

Using the TGM arrangement, the following parameters are typical values:

- Detection of relative thickness deviations <0.02% (1-σ)
- Primary beam intensity >$1.0 \cdot 10^{+9}$cps (Cu-Kα_1 and –Kα_2)
- Suppression of Cu-Kβ radiation at a ratio of 1000000:1
- Beam divergence <0.02°

3.2 EUV Reflectrometry

The characterization of multilayers for the reflection of EUV radiation also requires the metrology in this wavelength range. From Cu-Kα reflectometry only geometrical parameters and qualitative information about the multilayers can be derived. The exact reflectance of EUV mirrors with the accuracy needed can only be determined by at-wavelength reflectometry. Another problem is that concave curved substrates can not be measured under grazing angles. Also for this purpose, EUV metrology is mandatory. However, some additional difficulties arise with EUV reflectometry:

- Due to the high absorption of EUV radiation in air, the arrangement of source, monochromator, goniometer and detector has to be under vacuum.
- EUV sources with the necessary stability and power are not commercially available and require custom-made solutions.

Presently, measurements of the EUV reflectance are predominantly carried out at synchrotron beamlines, where a well-defined source with high power is available. The continued improvement of the measurement quality has resulted in an outstanding accuracy of these measurements. A total relative uncertainity of 0.25% for the peak reflectance and 0.025% for the peak wavelength is routinely achieved at PTB/BESSY2 [7,8].

However, for the production process of EUV optics for the lithography, the immediate access to metrology tools is necessary and the availability of stand alone devices is mandatory. Within the last years a stand alone EUV laboratory reflectometer for large samples has been developed, which meets the following specifications:

- Au-LPP-source ($3 \cdot 10^6$ shots/target),
- wavelength: 10 to 16 nm, wavelength uncertainty: 0.003 nm,
- angle of incidence: 3 to 60°
- measurement area: ∅ 500 mm, sample thickness: 200 mm, sample weight: 30 kg,
- measurement spot size: < 3 mm,
- uncertainty of reflectance: < 1%.

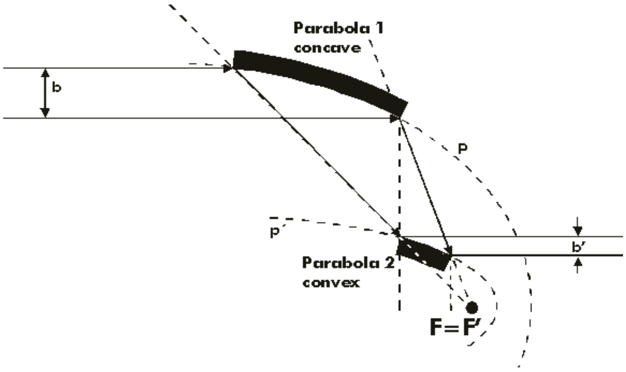
4 Experimental Results and Applications

4.1 Ni/C Multilayers

Besides other applications, Ni/C multilayers are preferably used as reflectors for Cu-Kα radiation. At this wavelength this material combination gives the highest reflectances. Depending on the multilayer period, these values can reach up to 90%. Another large advantage of Ni/C multilayers is the fact that, because of the Ni absorption edge between the Cu-Kα and –Kβ line, the undesired Kβ radiation is reduced by the multilayer itself and does not have to be suppressed by additional filters which would reduce the intensity.

The main applications of Ni/C multilayers are the so called Goebel mirrors, where a parabolic shaped surface is coated with a gradient multilayer [9]. With such Goebel mirrors, divergent radiation of commercial X-ray sources can be collected and collimated. A typical application in X-ray reflectometers and diffractometers has already been shown in section 3.1 (fig. 4). The main prerequisites for high intensities of Goebel mirrors are high reflectances of the Ni/C multilayers and precise gradients and surfaces according to the mathematical demands. High reflectances have been achieved by the optimization of the PLD process which is – due to the high particle energies – well-suited for the preparation of Ni/C multilayers with extremely smooth interfaces even at low multilayer periods. The gradient needed to fulfill the Bragg condition on every surface point of the mirror can be realized by the proper choice of the substrate motion in the X_2 direction (fig. 2 in section 2.2). Typical maximum deviations of the measured from the aim thickness are in the range of $\pm$0.03nm.

However, in addition to the rather simple Goebel mirrors, complex combinations of different gradient multilayers on curved substrates (parabolic, elliptic, concave, convex) have been investigated and tested [10]. Depending on the needs of our customers with respect to intensity, resolution, spot size and source geometry, we are able to design the optimum X-ray optical system. One example is given in fig. 5, where a so called beam collimator is shown. In this case, the aim was to get a small spot of X-rays to improve X-ray reflectometry and diffractometry on strongly curved substrates or on small samples. The final spot size reached is 0.3x0.6mm^2 with an intensity of >10^{5}cps within this area.



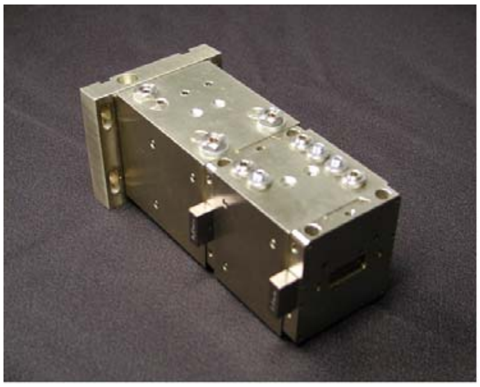


Fig. 5: Left: Scheme of the X-ray path for the beam compressor: the parallel part of the X-ray beam coming from the source is compressed from a size b to b'. Right: Photograph of the beam compressor.

4.2 Mo/Si Multilayers

For a long time, Mo/Si multilayers as near normal incidence mirrors for the EUV radiation have been of great interest and were first reported by Barbee [2]. During the last couple of years, the investigations of these multilayers have been increased due to their use in the EUV lithography, the next generation process for the semiconductor industry. Main features of this technology are the illumination of the mask and the demagnification of the mask structure onto the silicon wafer by the reflection of EUV radiation with a wavelength of λ=13.4nm on optics consisting of ultra-smooth substrates and high-reflection Mo/Si multilayers. In order to achieve the desired image quality, at least six EUV mirrors are necessary. Therefore a reflectance increase of every mirror of only 1% leads to a relative improvement of the overall throughput of nearly 10%. This shows that the optimization of the Mo/Si multilayer reflectance is one of the most urgent tasks. However, other conditions like uniformity, reproducibility and stability have to be fulfilled at the same time with high reflectance.

Multilayers prepared by MSD and consisting of two layers per period – Mo and Si – show a maximum reflectance of typically 69% [11,12]. The gap between this value and the theoretical limit (R=75.5%) is caused by the non-ideal structure of the multilayers. One of the main reasons for this discrepancy is the formation of intermixing zones on the interfaces between adjacent layers. We have found transition layer thicknesses of 1.2nm and 0.6nm on the Mo-on-Si and Si-on-Mo interfaces, respectively. Therefore we have carried out extensive investigations to reduce this intermixing by introducing thin barrier layers (C, B_4C) [12,13]. As a consequence, we succeeded to increase the reflectance to outstanding values of R_{EUV}=70.1% (λ=13.3nm, α=1.5°, fig. 6) and R_{EUV}=71.4% (λ=12.5nm, α=22.5°).

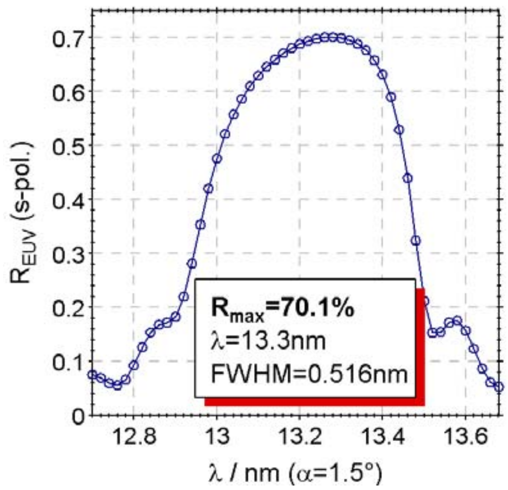


Fig. 6: EUV reflectance of interface-optimized Mo/Si multilayers at a photon wavelength of λ=13.3nm and at near normal incidence with α=1.5°. The mirror parameters are: Si-substrate/ 59(Mo/Si/C)/Mo/Si with a multilayer period of d=6.79nm. (Measurement was made at PTB/BESSY2)

Furthermore, the thermal stability has also been improved by the introduction of barrier layers. A multilayer with R_{EUV}=69.5% in the as-deposited state shows reflectances of 69.7% and 69.2% after annealing for 20 minutes at T=150°C and T=200°C, respectively. Typical uniformities of our multilayers are characterized by standard deviations of the multilayer period of 0.05% (flat), 0.06% (spherical concave) and 0.07% (spherical convex) across the substrate surface with diameters of 150mm. The run-to-run reproducibility of the deposition process results in uncertainties of the multilayer period of <0.1% (pure Mo/Si) and <0.2% (Mo/Si with barrier layers).

5 Summary

Multilayers with nm periods are deposited by different methods: MSD and PLD. Particularly the Mo/Si and Ni/C multilayer systems have been extensively investigated. Outstanding results have been obtained for both combinations (e.g. R_{EUV}>71% for optimized Mo/Si multilayers). The precision needed for the use of such multilayers as EUV and X-ray reflectors has been achieved on flat and curved substrates. The standard deviations of the multilayer period are <0.05% (flat), <0.06% (concave) and <0.07% (convex). The run-to-run reproducibility of the multilayer prepared by MSD is <0.2%.

Simultaneous to the improvements of the deposition technique, the metrology tools have also advanced. Several X-ray optical systems have been developed for Cu- and Mo-radiation, e.g. in order to improve the lateral resolution of X-ray reflectometry, which enables the accurate characterization of multilayers on curved substrates. Additionally, a stand-alone reflectometer for the EUV range has been developed, which permits the characterization of EUV optics at the working wavelength directly in the coating laboratories.

Acknowledgments

The authors would like to acknowledge the help of numerous people who have contributed to the success of recent research and development projects: R. Scholz (MPI Halle), F. Scholze, J. Tümmler (PTB Berlin), C. Rempel (Bestec Berlin), H. Stiel (MBI Berlin), F. Macco (Carl Zeiss Oberkochen), D. C. Meyer, P. Paufler (TU Dresden), Ch. Baerlocher (ETH Zürich), T. Sebald, S. Beyer (Astrium München).

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