Large area PLD of nanometer-multilayers

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Abstract

The deposition of nanometer-multilayers on technical relevant substrates, used as X-ray optics, makes extreme demands on the deposition process concerning precision, reproducibility and long-term stability. Across a stack of more than 150 layers with single layer thicknesses in the range between 1 and 10 nm, a variation of single layer thickness considerably lower than \( \sigma_D = 0.1 \) nm and an interface roughness below \( \sigma_R = 0.25 \) nm have to be realized. Thickness homogeneity \( \Delta d/d < 1\% \) and lateral thickness gradients \( \Delta d/\Delta x \approx 10^{-3} \) have to be guaranteed across macroscopic substrate dimensions.

Magnetron sputtering and e-beam evaporation are well-established deposition techniques to fabricate X-ray optical multilayers. For particular material combinations and for tailored thickness profiles, PLD has become an interesting alternative to these predominant technologies.

Within the last years the established 4 in. large area PLD technology of X-ray optical metal/carbon and carbon/carbon multilayers has been up-scaled to the deposition of substrates up to 6 in. diameter.

An improvement of long-term stability, which is necessary due to the increased substrate dimensions, is achieved by the modified target geometry and an increase in the accuracy of target and substrate positions in the range below 0.1 mm.

Metal/carbon, metal/B₄C, carbon/carbon X-ray optics both with homogeneous thickness distribution and with tailored thickness gradients can be deposited on 6 in. substrates by means of a completely automated deposition process. A period thickness homogeneity better than 1% across 6 in. substrate length, a layer by layer reproducibility of \( \sigma_D \leq 0.01 \) nm and a run-to-run stability \( \Delta d < 0.05 \) nm are achieved. As a result of the realized precision, Ni/C gradient multilayers, used as Göbel-Mirrors in X-ray analysis, can be deposited both on flat and on pre-curved substrates.

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1. Introduction

In contrast to the visible light X-rays cannot be deflected effectively by means of ordinary lenses and mirrors. Sufficiently reflected intensities can be achieved only on the condition of total reflection or at Bragg reflection on periodic structures.

For Bragg reflection either natural crystals with lattice constant \( d_{hkl} \) or—if for particular wavelengths or geometries no crystals are available—particularly fabricated nanometer-multilayers with the period thickness (one double layer) \( d \) are used.

Reflectivities of more than 80% can be achieved with such multilayers, consisting of more than 150 nm thick single layers. Besides the period number \( N \), the X-ray optical performance of real X-ray optical multilayers is determined by the period thickness \( d \), the interface roughness \( \sigma_R \), the variation of period thickness across
the total layer stack $\sigma_D$ and the X-ray optical constants of the alternating deposited spacer and absorber material, in principal. Typical values of these characteristic parameters of a nanometer-multilayer for X-ray optical applications are shown in Fig. 1. Extremely high qualified deposition techniques are demanded to meet these requirements across macroscopic substrate dimensions. Corresponding to the planned application multilayers with homogeneous thickness distribution and with tailored lateral or depth thickness gradients have to be deposited as well. A special challenge represents the synthesis of gradient multilayers on pre-curved substrates. Here the mean period thickness has to be fitted to the radius of curvature across the total substrate length. A mismatch between actual and set value in the range of $\Delta d \approx (\pm 0.05)$ nm can cause a perceptible decrease of reflectivity at this point.

Up to now magnetron sputtering and e-beam evaporation are well-established technologies to deposit X-ray optical multilayers. For selected material combinations as well as for the deposition of multilayers with tailored layer thickness profiles, PLD represents an interesting alternative to these predominant technologies [1]. In the following, selected results of large area PLD are demonstrated, achieved at metal/carbon X-ray optics deposited on flat and pre-curved substrates.

2. Large area PLD

A particular configuration of target and substrate handling was developed to solve the problem of the deposition of layers with large non-uniformity in thickness, initiated by the $\cos^n$-distribution ($n > 1$) of the particles in the strongly directed plasma plume. Because the plasma plume direction is close to normal direction of the target, the plume can be elongated by means of scanning the surface of a cylindrical target across the fixed position of a focused laser beam. Special thickness profiles are realized by applying a tailored relative motion of plasma plume and substrate. The principal arrangement of this dual-beam Fig. 1. Typical parameters of nanometer-multilayers for X-ray optical applications.

Fig. 2. Principle of dual-beam PLD source, designed for large area PLD of nanometer-multilayers.
PLD source is demonstrated in Fig. 2 and has already been published [2].

Keeping up the successful tested principle of plasma plume and substrate relative motion by means of ablation at the curved surface of a cylindrical target the previous 4 in. technology was up-scaled to the deposition of substrates up to 6 in. diameter.

For the 6 in. technology, the target dimensions were extended to a length of maximum 165 and 20 mm in diameter. A new substrate handling system was designed to carry substrates of 6 in. diameter and 2 in. height. The motions of all four targets and of the substrate can be automatically controlled and synchronized to the laser operation by a main computer. The target–substrate distance can be tuned in the range between 150 and 250 mm. A reproducibility of target and substrate positions in the range lower than 0.1 mm is realized under UHV (base pressure \( p < 2 \times 10^{-8} \text{ mbar} \)).

To find the best deposition conditions, the laser parameters are optimized for each material in the multilayer system. The optimization is necessary in order to get smooth interfaces and high contrast in the material density between spacer and absorber layers. Subsequently the morphology of multilayers is adjusted by varying the high reproducible geometry parameters like target and substrate motion. To guarantee long-term stability, target surfaces are pre-conditioned by laser ablation before deposition.

The complete system for the automated deposition of substrates up to 6 in. diameter under UHV conditions was realized as a cluster-tool in modular design. It consists of a PLD and a magnetron sputtering module. Both modules are connected by a completely automated substrate handling system with load lock and substrate magazine. The cluster-tool system with PLD module (left) and sputter module (right) is shown in Fig. 3. The installation of these two deposition

Fig. 3. Combination of large area PLD (left) and magnetron sputter deposition (right) in a UHV cluster-tool system for substrates up to 6 in. diameter.
methods represents an useful combination to produce a wide variety of X-ray optical multilayers.

3. Results

Previous results have shown, that by means of $\lambda = 1064$ nm moderate pulse energies are required to achieve high-performance Ni/C X-ray optics [2]. Using similar laser parameters for the 6 in. large area PLD, comparable results are observed in Ni/C, Ni/B$_4$C and Mo/B$_4$C multilayers. As demonstrated in Fig. 4, layer stacks show a regular morphology and smooth interfaces also at small period thicknesses of $d = 3.15$ nm. Comparable results are achieved for Mo/C and W/C multilayers with $\lambda = 355$ nm.

In continuous operation mode a high long-term stability is required for precise reproducibility and adjustment of layer parameters (thickness, material density) both within one multilayer deposition cycle and from run-to-run.

Within one multilayer cycle, which can take several hours, a variation of period thickness across layer stack lower than $\sigma_d = 0.01$ nm and a shift of $\Delta d < 0.001$ nm/period is measured in a 75-period Ni/C multilayer deposited on a 6 in. substrate (Fig. 5).

Cu K$\alpha$ X-ray reflectometry results show a peak width (FWHM) of the 1st order BRAGG peak of $\Delta(2\Theta) = 0.068^\circ$, which is close to the value of $\Delta(2\Theta) = 0.058^\circ$ achieved with an ideal layer stack. Reflectivities $R > 80\%$ for Cu K$\alpha$ radiation of the 1st order BRAGG peak indicate high material density contrast and smooth interfaces in the range of $\sigma_R \approx 0.25$ nm too.

Across 6 in. substrate length homogeneities of mean period thickness better than $\Delta d/d = 1\%$ are achieved in the $z$-direction using the substrate motion (Fig. 6). These multilayers are applied as rectangular monochromators for synchrotron radiation. Therefore homogeneity in $x$-direction (plume motion) better than $\Delta d/d = 1\%$ is limited to a range of $\Delta x = (\pm 25)$ mm.

Extreme process stability is required for synthesis of gradient multilayers on pre-curved substrates. Mean period thickness varies in sub-nanometer-scale across macroscopic substrate length because the set value of mean period thickness is defined by the incident angle at each point corresponding to the Bragg equation [3]. For applications as Göbel-Mirrors in the hard X-ray

Fig. 4. Layer stack morphology of metal/carbon and metal/B$_4$C multilayers, deposited at $\lambda = 1064$ nm at optimized laser parameters.
Fig. 5. X-ray reflectometry results (Cu Kα radiation) of a 75-period Ni/C multilayer (PL 106): mean period thickness $d = 4.4$ nm; results of simulation: variation of mean period thickness $\sigma_d = 0.01$ nm, period thickness drift $\Delta d \leq 0.001$ nm/period.

Fig. 6. Homogeneity of mean period thickness $d$ across 6 in. substrate length: $z$-direction (substrate motion): $d = (4.02 \pm 0.01)$ nm, $\Delta d/d < 0.3\%$ across 6 in.; $x$-direction (plume motion): $\Delta d/d < 1\%$ across 2 in.
regime, the maximum deviation from set value of mean period thickness has to be lower than 
$$\Delta d_{\text{rel}} = (\pm 0.9 – 1.3)\%$$ (depending on focal distance) at each substrate point to provide sufficient X-ray optical performance. Across a 60 mm length pre-curved substrate, a 75-period Ni/C gradient multilayer was deposited. The measured deviation between set and actual value of mean period thickness across 55 mm was in the permitted thickness range (Fig. 7). Only in the low period thickness range (small focal length) at the beginning of the mirror, there is a deviation of more than 0.9% and the limit is exceeded by +0.016 nm.

4. Conclusions

The established dual-beam source for 4 in. large area PLD was successfully up-scaled to the 6 in. geometry.

Deposition of metal/carbon and metal/B_{4}C multilayers can be realized on substrates up to 6 in. in diameter because of the improved long-term stability of the deposition process. In particular, the deposition of pre-curved substrates was successfully realized, which is necessary for the fabrication of high-performance X-ray optics, used as Göbel-Mirrors in X-ray analysis.

In combination with magnetron sputter deposition, a wide variety of material combinations for X-ray optics can be deposited under optimized technological conditions. Further work will be focused on the improvement of long-term stability and thickness homogeneity across the total 6 in. substrates.

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References


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Fig. 7. Mean period thickness difference of set and actual value of a 75-period Ni/C gradient multilayer, deposited across a pre-curved substrate (Göbel-Mirror).