Attosecond broadband multilayer mirrors for the water window spectral range

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ABSTRACT

Recent advances in the development of attosecond soft X-ray sources ranging into the 'water window' spectral range, between the carbon 1s and oxygen 1s states (284 eV - 543 eV), are also driving the development of suited broadband multilayer optics for attosecond beam steering and dispersion management. The relatively low intensity of current High Harmonic Generation (HHG) soft X-ray sources calls for an efficient use of photons, thus the development of low-loss multilayer optics is of uttermost importance. Here, we report about the realization of atomically smooth interfaces in broadband CrSc multilayer mirrors by an optimized ion beam deposition and assisted interface polishing process.

Keywords: Multilayers, Polishing, Roughness, Mirrors, Attosecond Pulses, High Harmonics, Ion Beam Deposition

1. INTRODUCTION

Highly reflective multilayer mirrors for the water window spectral range, the spectral range defined by the K-edges of carbon and oxygen (284 eV and 543 eV, respectively), have become of great interest over the last decade [1-3]. Many different applications such as high-resolution microscopy [4], time-resolved attosecond (soft) X-ray spectroscopy [5] or X-ray astronomy [6] demand multilayer mirrors for beam steering, beam focusing or spectral shaping. Here we focus on the extension of mirror technology for single attosecond (1 as=10^{-18} s) pulses from currently sub-200 eV [7] to the water window spectral range. This will not only provide new insight into the dynamics of ultrafast core-shell electron wave packets, measured by time-resolved spectroscopy, it will also push forward the investigation of biomolecules and cells in attosecond time-resolved soft X-ray microscopy, for which the water window is an ideal testing environment. There is a high demand for highly reflective multilayer mirrors in the water window wavelength range due to the lack of sources with sufficient photon flux. The dominating generation process for attosecond pulses is High Harmonic Generation (HHG), which can be generated to the keV region [8]. However, the available photon numbers are, especially for low cross section experiments, not sufficiently high. The most suitable multilayer material combination, covering the lower spectral half of the water window up to about 400 eV, is chromium (Cr) and scandium (Sc) [9], with the highest achievable reflectivity in the vicinity of the Sc L_3-absorption edge (≈ 398 eV). Multilayer mirrors allow for spectral shaping of attosecond pulses based on the pulse-bandwidth requisite. To achieve sufficient reflectivity, it requires interface-optimized multilayer mirrors with almost perfect interfaces due to the huge loss in reflectivity, which originates from interface imperfections.

Here, we report about our achievements in minimizing the interface roughness of ion beam deposited Cr/Sc multilayer mirrors by optimizing the kinetic energy of krypton ions in the deposition but also by assisted ion-beam interface polishing process. We show experimental results on our ion-beam polished (sub-) nanolayers from measurements using X-ray reflectometry, spectral ellipsometry as well as transmission electron microscopy (TEM) cross section analysis.

2. METHODOLOGY AND MIRROR PARAMETERS

2.1 Ion beam deposition and ion beam polishing

All mirrors were produced using a dual ion-beam load-locked deposition machine at a constant background pressure of 10^{-9} mbar. A detailed explanation of the setup can be found in Guggenmos et al. [10]. Inside the two radio frequency inductively-coupled ion beam sources, krypton (Kr) gas is ionized and the ions are accelerated by means of the applied sputtering grid voltage, either towards the selected target material (deposition plasma source) or directly towards the substrate (assist plasma source). The krypton ions are neutralized directly after leaving the sources to prevent the targets and substrates from charging effects which might result in fluctuating deposition rates. Layer thicknesses are controlled over sputter times calculated from material-dependent bulk sputtering rates. The model takes into account diffusion, height factors and additional process parameters, such as shutter response time. Typical deposition rates for chromium and scandium are in the range of 0.5 Å/s. For the purpose of the carried out examinations, depositions were performed on crystalline Si (100) wafers with a native SiO_2 layer of approximately 20 angstroms.

We have composed three different sets of deposition parameters, referred to as *default*, *optimized* and *optimized+assist*. The *default* parameter set includes deposition parameters which are previously established for material systems suitable for attosecond multilayer mirrors beyond 200 eV, like silicon (Si), molybdenum (Mo), lanthanum (La) or boron carbide (B_4C) [7,11]. For the optimized sets we refined the deposition of chromium and scandium in terms of kinetic energy. Refinement of deposition parameters includes Monte-Carlo simulations (Figure 1) using the software SRIM [12] in order to find the best trade-off between the optimum kinetic energy of the sputtered atoms for layer-by-layer growth [13] and a low penetration depth into the subjacent layer, thus preventing layer intermixing [14].

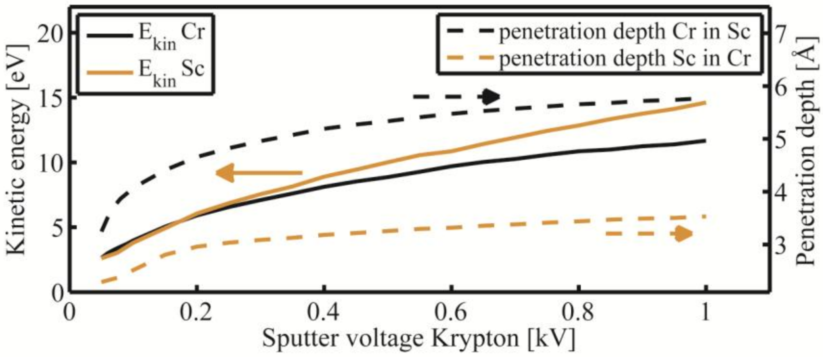

Figure 1. Monte Carlo simulation: Kinetic energy (solid lines) and penetration depth (dashed lines) at the interface for chromium and scandium atoms dependent on the sputter voltage being used for the krypton ions.

The last parameter set *optimized+assist* is identical to the parameter set *optimized*, with one important difference: the assist plasma source was used for performing ion polishing on every 10^{th} Cr layer, thus every 10 periods. Previous experiments showed that no loss in height is caused by the polishing process with the chosen parameters and only surface smoothening occurs. The polishing step for 60 seconds, after every 10^{th} period, has been chosen as a trade-off between the overall deposition time and the smoothening effect.

2.2 Mirror parameters

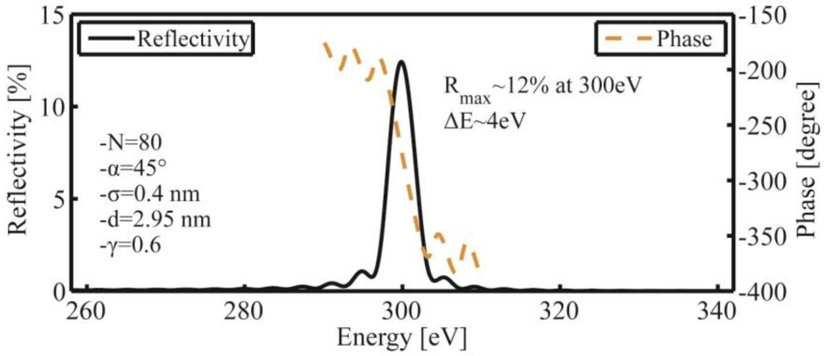

Figure 2. XUV simulation of the reflectivity (solid) and phase (dashed) with the chosen mirror parameters.

With these three sets of parameters, Cr/Sc multilayer mirrors with a period number of N=80, a period thickness of d=2.95 nm and a gamma factor of γ=0.6 (d_{Sc} / (d_{Sc}+d_{Cr})) were realized, resulting in individual layer thicknesses for Cr and Sc of 1.18 nm and 1.77 nm, respectively. The mirrors were designed for a central energy of 300 eV and a FWHM bandwidth of 4 eV at an angle of incidence of 45 degree (Figure 2). The mirrors were optimized for the reflection of sub-500 as pulses. Simulations reveal a peak reflectivity of ~12% when assuming an interface roughness of σ=0.4 nm, which originates from XUV measurements, on mirrors with comparable designs, which are not shown here. The theoretical total stack height of 237.1 nm (including the top chromium oxide layer) was experimentally proven by surface profilometry with values of (237.4 ± 0.9) nm.

3. ANALYSIS METHODS AND RESULTS

3.1 Characterization by hard X-ray reflectometry

Hard X-ray reflectometry (XRR), using a molybdenum K_α source with a wavelength of $\lambda\approx$0.71 Å, was performed for a first hint on the impact of the different kinetic energies and of the polishing effect on the interface roughness σ. A comparison of the measured XRR data of the three mirrors is shown in Figure 3.

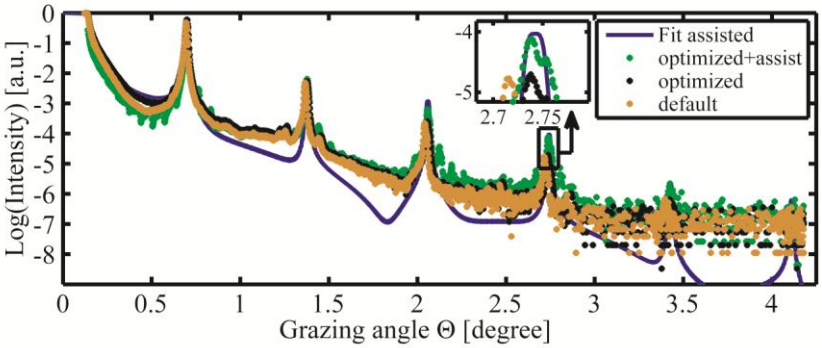

Figure 3. Hard XRR data for the three different mirrors. The lowest interface roughness, thus the highest peak intensities, shows the *optimized+assist* mirror. Only the fit of the interface polished mirror is shown.

When comparing the intensities of the higher order Bragg peaks, where the impact of interface roughness is high, one can clearly see the improvement caused by the tailoring of the deposition parameters. Out of the fitting procedure for each measurement, we obtained a mean interface roughness value of σ=0.26 nm for the *default*, σ=0.24 nm for the *optimized* and σ=0.21 nm for the *optimized+assist* parameter set.

3.2 Characterization by transmission electron microscopy

We have seen that a higher reflectivity could be gained by a lower interface roughness. To confirm these findings TEM measurements were carried out on Si (100) witness samples produced under the three different parameter sets (Figure 4). Due to their period number of N=80 and their period thickness of d=2.95 nm, these coatings were perfectly suited for TEM analysis, which is limited to a stack height of approximately 250 nm. The TEM images of the coatings are shown in Figure 4 with the upper row displaying images of the top six periods of each coating (topmost layer Cr_2O_3 [10]) and the lower row showing images from the bottom seven start periods of each stack with the bottom-most layer Sc. In the TEM images chromium layers show up darker than scandium layers due to their higher atomic number.

For the *default* parameter set (Figure 4, left column), layers are clearly distinguishable at the bottom of the stack (Figure 4d) whereas they show a high degree of intermixing at the top (Figure 4a). This is confirmed by the diminishing image contrast. With the *optimized* parameter set the layers at the bottom and the top (Figure 4, center column) are comparable with regard to image contrast, while for the additionally polished multilayer the layers at the top (Figure 4c) seem to show even a decreased intermixing and increased layer-to-layer interface abruptness. In summary, the best layer characteristics and stack evolution so far has been achieved by optimized kinetic deposition energy and chromium layer ion polishing of every tenth period resulting in clearly distinguishable layers, both at the bottom and at the top of the stack and significantly improved layer-to-layer interfaces without roughness accumulation over an 80 period stack.

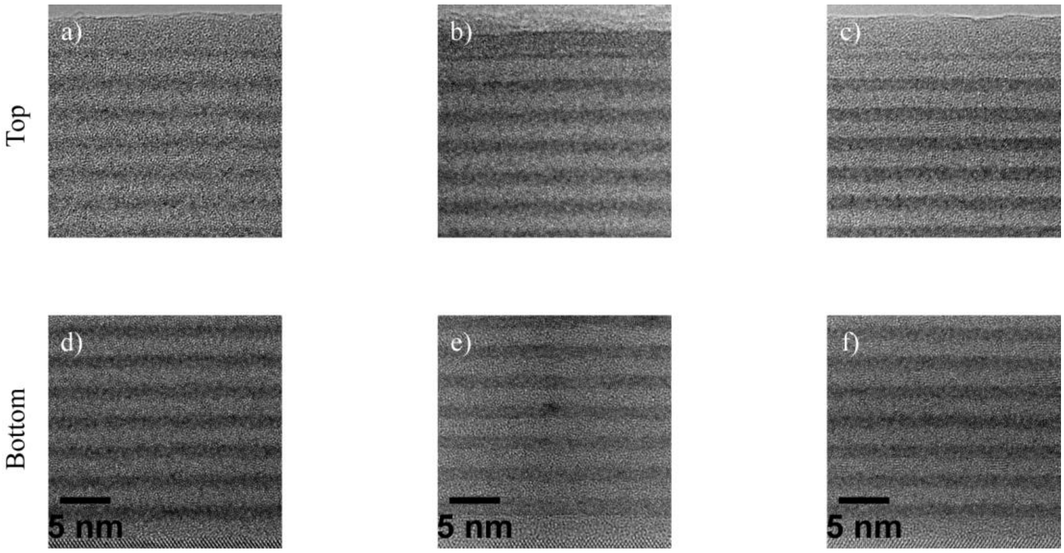

Figure 4. TEM cross section images of the multilayer evolution from bottom (lower panel) to top (upper panel) with the *default* [a) + d)], optimized [b) + e)] and optimized parameters including the assisted interface polishing [c) + f)].

A complete TEM image of the polished multilayer stack is shown in Figure 5. It demonstrates the homogeneity of the multilayer stack over the whole film thickness.

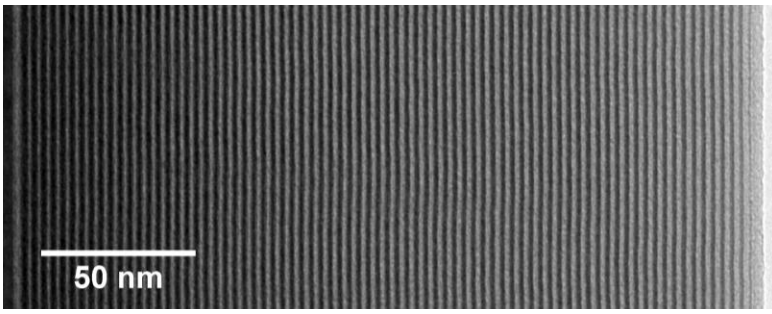

Figure 5. TEM cross section image of the whole multilayer stack for the additional polished multilayer.

3.3 Ellipsometry

Our ion-beam deposition machine is equipped with an in-situ spectral ellipsometer from Woollam (EC-400 / M-2000VI, λ=370-1695 nm) allowing for ellipsometric measurements under vacuum conditions. We have analyzed the produced mirrors with this tool during deposition and directly afterwards. The effect of interface polishing is even clearer visible for samples with a higher number of periods, as roughness evolves through the stack, thus we have increased the period number to N=500 and thus 999 interfaces. Since metals get 'optically thick' in the visible/infrared spectral region at a thickness of approximately 80 nm, only the last ~30 periods contribute to the ellipsometric analysis. Roughness has an impact on the measurement, thus it is resolvable by spectral ellipsometry [15]. We have investigated the influence of the assisted polishing effect on the multilayer stack evolution by measuring the ellipsometry parameters Ψ and Δ and calculating the stack's mean dispersion and absorption $\langle n \rangle$ and $\langle k \rangle$. We compare two multilayer stacks, where both are identical in terms of mirror parameters N, d, and gamma, but only one got polished with the assist source after every 10^{th} period. Since the total stack height, proven by surface profilometry, is identical for both multilayer mirrors, the ellipsometrically determined overall absorption is also expected to be the same (material amount is equal). The only measurable effect remaining is decreased dispersion due to decreased interface roughness and thus reduced scattering. These effects, nearly constant absorption and decreasing dispersion, are visible in the comparison of our polished and non-polished multilayer mirrors, measured via spectral ellipsometry, and are shown in Figure 6.

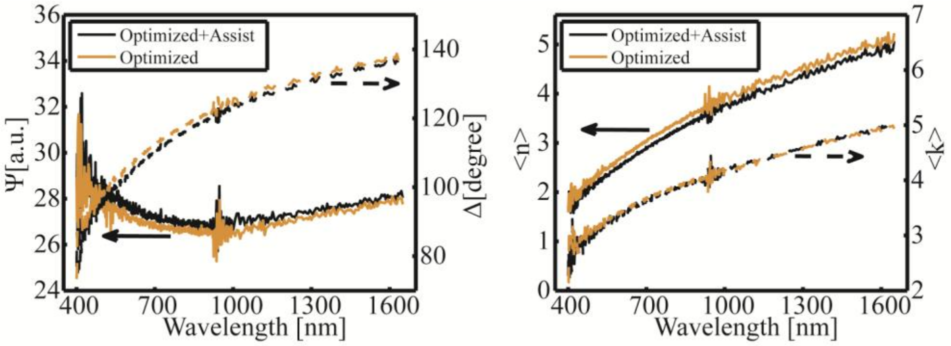

Figure 6. Measured ellipsometry parameters Ψ and Δ for the assisted polished and non-polished multilayer mirrors (left) and the calculated mean dispersion $\langle n \rangle$ and absorption $\langle k \rangle$ for both systems (right). The absorption stays nearly constant while the dispersion could be decreased by polishing, thus reduced scattering due to lower interface roughness.

4. CONCLUSIONS

We have proven the reduction of interface roughness of water window CrSc multilayer mirrors. Broadband attosecond mirrors for the reflection of sub-500 as pulses have been fabricated. The ion beam deposited nanolayer growth was optimized by tweaking the deposition parameter of CrSc multilayer mirrors, mainly by tailoring the kinetic energy of the target atoms, where the optimization was theoretically accompanied by Monte-Carlo simulations. Besides, we have realized an important step towards 'perfect' layer growth by additionally assisted ion beam interface polishing. This technique even enabled a decreasing interface roughness during the layer growth from the substrate to the top layer. Using a well-suited kinetic energy for the target materials (chromium and scandium) and additional interface polishing for tailored attosecond multilayer mirrors will increase the reflectivity dramatically. Such low-loss mirrors will thus enable for the first time attosecond-resolved experiments on biological samples in the water window spectral range or pave the way towards shorter pulse durations [16,17], by filtering a multi-octave cut-off spectrum from future HHG soft X-ray sources.

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REFERENCES

[1] Eriksson, F., Johansson, G. A., Hertz, H. M., Gullikson, E. M., Kreissig, U. and Birch, J., "14.5% near-normal incidence reflectance of Cr Sc x-ray multilayer mirrors for the water window," Optics Letters 28(24), 2494-2496 (2003).

[2] Kuhlmann, T., Yulin, S., Feigl, T., Kaiser, N., Gorelik, T., Kaiser, U. and Richter, W., "Chromium-scandium multilayer mirrors for the nitrogen K_α line in the water window region," Applied Optics 41(10), 2048-2052 (2002).

[3] Eriksson, F., Ghafoor, N., Hultman, L. and Birch, J., "Reflectivity and structural evolution of Cr/Sc and nitrogen containing Cr/Sc multilayers during thermal annealing," Journal of Applied Physics 104, 063516 (2008).

[4] Gorniak, T., Heine, R., Mancuso, A. P., Staier, F., Christophis, C., Pettitt, M. E., Sakdinawat, A., Treusch, R., Guerassimova, N., Feldhaus, J., Gutt, C., Grübel, G., Eisebitt, S., Beyer, A., Gölzhäuser, A., Weckert, E., Grunze, M., Vartanyants, I. A. and Rosenhahn, A., "X-ray holographic microscopy with zone plates applied to biological samples in the water window using 3rd harmonic radiation from the free-electron laser FLASH," Optics Express 19(12), 11059-11070 (2011).

[5] Schultze, M., Fieß, M., Karpowicz, N., Gagnon, J., Korbman, M., Hofstetter, M., Neppl, S., Cavalieri, A. L., Komninos, Y., Mercouris, Th., Nicolaides, C. A., Pazourek, R., Nagele, S., Feist, J., Burgdörfer, J., Azzeer, A. M., Ernstorfer, R., Kienberger, R., Kleineberg, U., Goulielmakis, E., Krausz, F. and Yakovlev, V. S., "Delay in Photoemission," Science 328(5986), 1658-1662 (2010).

[6] Santos-Lleo, M., Schartel, N., Tananbaum, H., Tucker, W. and Weisskopf, M. C., "The first decade of science with Chandra and XMM-Newton," Nature 462, 997-1004 (2009).

[7] Hofstetter, M., Aquila, A., Schultze, M., Guggenmos, A., Yang, S., Gullikson, E., Huth, M., Nickel, B., Gagnon, J., Yakovlev, V. S., Goulielmakis, E., Krausz, F. and Kleineberg, U., "Lanthanum-molybdenum multilayer mirrors for attosecond pulses between 80 and 130 eV," New Journal of Physics 13(6), 063038 (2011).

[8] Popmintchev, T., Chen, M.-C., Popmintchev, D., Arpin, P., Brown, S., Ališauskas, S., Andriukaitis, G., Balciunas, T., Mücke, O. D., Pugzlys, A., Baltuška, A., Shim, B., Schrauth, S. E., Gaeta, A., Hernández-García, C., Plaja, L., Becker, A., Jaron-Becker, A., Murnane, M. M. and Kapteyn, H. C., "Bright Coherent Ultrahigh Harmonics in the keV X-ray Regime from Mid-Infrared Femtosecond Lasers," Science 336(6086), 1287-1291 (2012).

[9] Schäfers, F., Mertins, H.-C., Schmolla, F., Packe, I., Salashchenko, N. N. and Shamov, E. A., "Cr/Sc multilayers for the soft-x-ray range," Applied Optics 37(4), 719-728 (1998).

[10] Guggenmos, A., Rauhut, R., Hofstetter, M., Hertrich, S., Nickel, B., Schmidt, J., Gullikson, E. M., Seibald, M., Schnick, W. and Kleineberg, U., "Aperiodic CrSc multilayer mirrors for attosecond water window pulses," Optics Express 21(19), 21728-21740 (2013).

[11] Hofstetter, M., Schultze, M., Fieß, M., Dennhardt, B., Guggenmos, A., Gagnon, J., Yakovlev, V., Goulielmakis, E., Kienberger, R., Gullikson, E. M., Krausz, F. and Kleineberg, U., "Attosecond dispersion control by extreme ultraviolet multilayer mirrors," Optics Express 19(3), 1767-1776 (2011).

[12] Ziegler, J. F., Ziegler, M. D. and Biersack, J. P., "SRIM - The stopping and range of ions in matter (2010)," Nuclear Instruments and Methods in Physics Research B 268, 1818-1823 (2010).

[13] Hubler, G. K. and Sprague, J. A., "Energetic particles in PVD technology: particle-surface interaction processes and energy-particle relationships in thin film deposition," Surface and Coatings Technology 81(1), 29-35 (1996).

[14] Gilmore, C. M. and Sprague, J. A., "Interface mixing of energetic metals deposited onto metals," Surface and Coatings Technology 83(1), 146-150 (1996).

[15] Nagib, N. N., Mahmoud, N. A., Ismail, L. Z., Amer, M. A. and Abd-El-Sabour, Kh., "Effect of surface roughness on the optical constants of bulk polycrystalline gold samples," Optik 125, 1085-1087 (2014).

[16] Kleineberg, U., "New routes towards even shorter attosecond soft X-ray pulses," Annalen der Physik 525(12), A188-A190 (2013).

[17] Goulielmakis, E., Schultze, M., Hofstetter, M., Yakovlev, V. S., Gagnon, J., Uiberacker, M., Aquila, A. L., Gullikson, E. M., Attwood, D. T., Kienberger, R., Krausz, F. and Kleineberg, U., "Single-cycle nonlinear optics," Science 320, 1614 (2008).