CHAPTER 6

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6 Summary

This thesis treats the characterization of multilayer mirror systems by means of the combination of several indirect methods based on reflection, fluorescence and scattering using EUV and X-ray radiation. In chapter 4, a methodology for a profound analysis on the validity and accuracy of the structural characterization obtained from extreme ultraviolet reflectivity, X-ray reflectivity, resonant extreme ultraviolet reflectivity and X-ray fluorescence experiments was presented and applied to different multilayer mirror samples. The uniqueness and accuracy of a solution to the inverse problem is an important aspect to justify the relation of the model parameters to actual sample properties. Thus, for the reconstruction of the layer systems structure, a particle swarm optimization was applied to fit the model parameters to the measured data and a Markov-chain Monte Carlo algorithm was employed to deduct the confidence intervals based on the measurement and model uncertainties addressing this problem. Depending on the sample properties, different methods have to be combined to improve the accuracy of the result and thereby provide the desired significance or to deliver a consistent solution in the first place. In particular, the different layer thicknesses of the two sample types under investigation in this thesis, the Mo/Si mirrors designed for 13.5 nm wavelength and the Cr/Sc mirrors for the water window, do have a significant influence on the applicability of the model and the analytic experiments required to deduct a consistent result.

Starting from the results for the structural properties of the layer systems, the characterization of the interface morphology of the samples was addressed in chapter 5. While delivering a result on the influence of both, the effect of roughness and intermixing at the interfaces in combination through the Névot-Croce parameter, none of the analytic experiments conducted for the structural reconstruction can distinguish those two effects. This issue was approached by establishing the analysis of EUV diffuse scattering with radiation impinging near-normal incidence as a suitable technique to deliver this distinction method. Sec. 5.1 introduces the method by analyzing a state-of-the-art $Mo/B_4C/Si/C$ mirror reaching (68.5 ± 0.7) % peak reflectance at its operation wavelengths of 13.5 nm. It was revealed that the high quality, and thus reflectivity, of the sample causes resonant enhancement of diffusely scattered radiation within the stack to significantly contribute to the diffuse scattering intensities. These dynamic effects require to be considered in the analysis by employing the theoretical framework of the distorted-wave Born approximation including multiple reflections at the interfaces of the multilayer. With this approach, the roughness properties for the samples could be extracted consistently.

In conclusion, comparing and combining the results of the structural characterization from chapter 4 and the roughness analysis in chapter 5 delivers a consistent characterization of the multilayer mirrors. The scope of this thesis is to yield an explanation for the lack of peak reflectivity compared to the theoretical threshold observed.

In Sec. 4.2 and Sec. 5.2, two sets of Mo/Si/C multilayer samples were investigated. In both sets, the thickness of the molybdenum layer was increased from sample to sample from nominally 1.7 nm to 3.05 nm crossing the threshold for crystallites forming in these layers. Furthermore, the second set was treated using an ion polishing technique during deposition with the goal to reduce roughness at the interfaces. Sec. 4.2 reveals that the combination of EUV reflectivity and X-ray reflectivity yields an unambiguous result for the molybdenum layer thickness following the nominal trend in both sets. The confidence intervals for the molybdenum thickness could be determined ranging from 0.43 nm down to 0.24 nm, depending on the sample. In comparison, the analysis of EUV reflectivity alone for the comparison sample Mo/B_4C/Si/C in Sec. 4.1 only yielded a confidence interval of approximately 1 nm, demonstrating the need for combining multiple datasets despite an excellent agreement of the calculated and measured curves as multiple solutions exist.

The sum of thicknesses of all layers in a period shows a distinct increase for both sets at a certain molybdenum layer thickness associated with a dip in peak reflectance with respect to the theoretical expectation as illustrated in Fig. 4.14. This effect, while observed in both sets, happens at significantly different molybdenum thicknesses comparing the unpolished with the polished samples. For those samples, the diffuse scattering intensity was analyzed in Sec. 5.1 to assess the interface morphology. The comparison with the structural analysis revealed an increase of roughness associated with the jump in the period thickness and the dip in peak reflectance, which is compensated again at even larger thicknesses. At this point, it may be concluded that these effects are caused by the crystallites forming in the molybdenum layer causing increased interface disturbances. By applying the ion polishing technique during the deposition of the sample, this threshold was moved towards lower molybdenum thicknesses. This is beneficial to the reflectance at the optimum molybdenum ratio with respect to the rest of the period, which in the polished set is now unaffected by roughening due to crystallization. However, comparing the roughness values found in the diffuse scattering analysis with the Névot-Croce factor from the optimized layer structure model, it becomes clear that while overall roughness was reduced significantly in the polished set, the Nevót-Croce parameter only reduced slightly indicating that intermixing is largely responsible for the remaining gap to the theoretically achievable reflectivity and thus requires to be addressed for even further improvement of the mirror.

In the case of the Cr/Sc multilayers for the water window spectral range, nominal layer thicknesses within a bilayer period are between 0.7 nm and 0.8 nm and thus noticeably thinner than for the Mo/Si systems. It was shown in Sec. 4.3.1, that an approach to the structural characterization based on a discrete layer model for the chromium and scandium layers does not yield a solution to both, the EUV reflectivity and XRR experiments, with the same set of parameters. Thus, a solution fitting the EUV reflectivity experiment fails to describe the XRR curve and vice verse. The discrete layer model is not suitable to describe the physical structure of the sample sufficiently and any solution found for either one of the experiments can not be related to the physical properties of the sample. Instead, a model describing a gradual interface profile and layers composed of a mixture of both materials was introduced in Sec. 4.3.2. Based on this gradual model, the intermixing and roughness were parametrized separately and asymmetric interface profiles could be described explicitly.

For this system the scope was to consistently determine all parameters of the improved model and thereby identify the cause for the low reflectivity. The question of uniqueness and accuracy of the solution was therefore addressed by performing EUV reflectivity, REUV, XRR and XRF experiments. Those were analyzed with respect to their utility to deliver solutions for that model in Sec. 4.3.4, by evaluating each dataset individually and by combining all in a single analysis. The solutions and confidence intervals determined prove, that only the combination of all datasets could yield a consistent result. It was found, that none of the regions within the Cr/Sc stack are pure chromium or scandium. Furthermore, the interface regions show a strong asymmetry, which could not be determined with the required significance by any of the analytic experiments alone. For the same reason as stated above, not even the combined analysis could distinguish roughness and intermixing. Those two parameters were shown to have a strong correlation requiring either one to exists in the sample. Thus identifying these as the explanation for the low reflectivity as compared to the theoretically possible values. To distinguish roughness and intermixing, the EUV diffuse scattering was measured and analyzed in Sec. 5.3. The result shows a roughness value of $\sigma_r = 0.17(-0.01/+0.02)$ nm. Consequently, the intermixing could be determined to be $47(-4/+3)$ %, leaving any of the nominal chromium or scandium layers of the stack to contain at least that amount of the other material on average. In conclusion, the roughness determined agrees with the roughness observed for the polished Mo/Si/C samples, leaving intermixing as the main cause for the small reflectivity of the Cr/Sc multilayer mirrors for the water window and thus the point were the improvement of the fabrication process should be made.

In summary, the work presented in this thesis highlights the importance of assessing the uniqueness and accuracy of indirect characterization methods to deduct a meaningful result. As shown on several occasions in the analysis of the multilayer mirrors, reconstructions in very good agreement with the data curves were proven to show ambiguities and inconsistencies, when adding complementary information from other experiments or even when analyzing the data of a single experiment using global optimization algorithms. With the approach of combining multiple analytic techniques and determining confidence intervals of the reconstructed parameters shown here, conclusions on the physical properties of the samples could be drawn reliably. This thesis thus augments the existing characterization methods for multilayer mirrors in that respect. Finally, with the inclusion of EUV diffuse scattering, a technique to assess the interface morphology was established suitable for at-wavelength characterization near-normal incidence offering an alternative to grazing-incidence methods such as grazing-incidence small-angle X-ray scattering (GISAXS).