

Smooth interfaces of multilayer monochromators

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Abstract

Monochromators used in neutron or X-ray experiments sometimes have unintended higher order contributions. In most cases filters are used to remove them. We sputtered multilayer monochromators with smooth interfaces to reduce the higher order contributions removing the need for a filter. Ni and Ti are selected as sublayer materials due to their high scattering length density contrast. We have strongly reduced the higher order contributions by annealing. This reduces the roughness and off-specular scattering, however the second and third-order Bragg peak intensities increase unexpectedly in neutron reflectivity experiments.

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1. Introduction

Conventional multilayers used as monochromators have higher order contributions due to their sharp scattering length density profiles normal to the planes (in the following labelled $\overline{\rho(z)}$). Multilayers can be considered as one-dimensional crystals with Bragg peaks for the following condition: $\sin(\theta) = \lambda/2d$ with the period d .

The interfaces are characterized by two main properties. The first is the roughness of the interface which gives rise to off-specular scattering. The second is the smoothness of $\overline{\rho(z)}$, which is the laterally-averaged density profile of the multilayer perpendicular to its surface. It can be regarded as a step function convoluted with a gauss function. To prevent off-specular scattering and to reduce the higher order contributions, the approach of producing multilayers with blurred interfaces is followed.

The smoothness of the interface is dependent on the intermixing between neighbouring sublayers.

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A controlled interdiffusion can lead to an amorphisation of the interfaces and reduced roughness [1]. As we have shown in previous work [2–4], it is indeed possible to influence the smoothness. By modifying the interfaces in bilayer monochromators, a change in the Bragg peak intensities is observed. A sinusoidal $\overline{\rho(z)}$ should prevent all higher order contributions. This has to be paid for by increasing the number of periods by a factor of $\pi/2$ relatively to a multilayer with ideal sharp interfaces to have the same intensity in the first-order Bragg peak.

By annealing the multilayer, diffusion can lead to a blurring of the interfaces. We additionally introduced thin intermediate sublayers at the interface [5], which artificially increased the limited diffusion length. In the following, the results from specular neutron reflectivity experiments with respect to the annealing process of the multilayer monochromators with intermediate sublayers are presented.

2. Sample preparation

The multilayers consisting of Ni and Ti sublayers have been produced by magnetron sputtering on a Z600 Leybold at PSI, Switzerland. An additional inserted aperture limited the sputtering rate allowing for thinner layers. A silicon wafer with surface roughness of 4 Å was used as a substrate. The temperature within the sputtering chamber was up to 318 K. An overview over the investigated samples is given in Table 1. Intermediate sublayers of the same materials have been introduced, increasing the number of layers within a period from 2 to 22. The difference between the individual samples is the ratio of Ni to Ti thickness

in a period. The investigation of the temperature dependence upon annealing was the reason for the different ratios. The annealing has been performed in situ with a vacuum of 1.6×10^{-6} mbar.

3. The experiments

The neutron experiments have been performed on the time-of-flight reflectometer AMOR at SINQ in Switzerland. There a furnace was installed on the sample holder to anneal the sample in situ. The temperatures used are 323, 373, 423, 472 and 523 K. The annealing temperature was increased slowly to prevent a overshoot in the temperature. The illuminated sample size was 6 cm². The experimental range in intensity was over five orders of magnitude. Off-specular scattering measurements have been performed around Bragg peak positions, to separate the contributions of roughness and smoothness upon the damping of the intensity. X-ray diffraction has been performed with an incident energy of 8040 eV.

4. Results

As written above, $\overline{\rho(z)}$ can be regarded as a step function convoluted with a Gaussian from the roughness. The roughness is in our case 4 Å from the substrate. The diffusion length is depending on the annealing temperature. The temperature can not be set to a too high value because the sample can be destroyed (e.g. melted). Due to the numbers of the layers within a period and the diffusions length, the period thickness is limited. Increasing of the period thickness is possible, but the

Table 1
Layer sequences of sample 1 and 2. The thicknesses used in a simulation are shown

Layer structure																						
Material	Ti	Ni	Ti	Ni	Ti	Ni	Ti	Ni	Ti	Ni	Ti	Ni	Ti	Ni	Ti	Ni	Ti	Ni	Ti	Ni	Ti	Ni
thickness in Å	25	4	18	4	6	4	4	6	4	18	4	25	4	18	4	6	4	4	6	4	18	4

Fifteen periods of the shown sequence have been used in both samples. Sample 2 has an increased Ti thickness by 18% compared to sample 1.

sputtering time is increased drastically since the numbers of sublayers has to be increased accordingly. Above a critical thickness of the individual sublayers, $\overline{\rho(z)}$ will begin to deviate from a sinusoidal function. Because of this, here the period of the samples is chosen to be 194 Å. As simulation of the reflectivity is given in Fig. 1.

The time dependence for the annealing process is addressed for sample 1, presented in Fig. 2. First the sample was annealed for 5 min at 473 K then

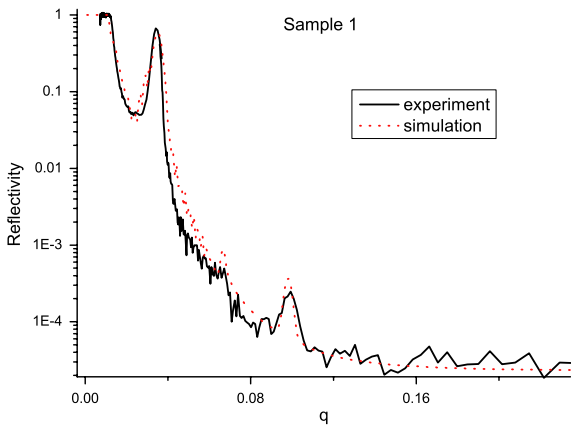


Fig. 1. Comparison of the simulation and the reflectivity of the not annealed sample 1.

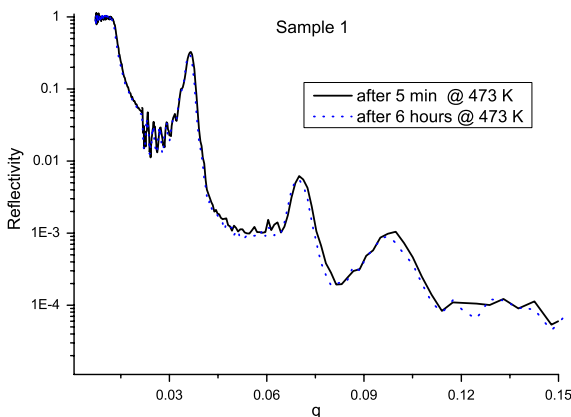


Fig. 2. Time dependence for sample 1 and one temperature. The dotted line is the annealed sample (6 h at 473 K). As is visible no time dependence is found. Several additional measurements have been performed, for clarity reasons only two are shown.

measured. The second measurement was performed after 6 h annealing at the same temperature. As shown the intensities for the Bragg peaks remained the same.

As shown for the same sample 1 in Fig. 3, the not annealed sample has a reduction in higher order contributions intensities by three orders of magnitude compared to a $\overline{\rho(z)}$ with sharp interfaces. The annealing process enhanced strongly the second-order Bragg peak and higher order contributions are also distinguishable. The intensity gain could originate from a change in the sublayer structure since it is possible that Ni_3Ti or NiTi_2 is formed leading to rather sharp interfaces again. Further experiments are needed to confirm or reject this theory.

If the thickness ratio of the sublayers is modified as in sample 2, the annealing is less enhancing the higher orders of the multilayer, see Fig. 4. Still all higher order contributions up to the fourth Bragg peak become distinguishable. Off-specular scattering does not show an increase or reduction in intensities after the annealing.

The X-ray diffraction of the Si wafer, the not annealed and the annealed sample 2 indicated, that the Ni and Ti formed small crystallites of Ni, Ti, NiTi and NiTi_2 during the annealing process.

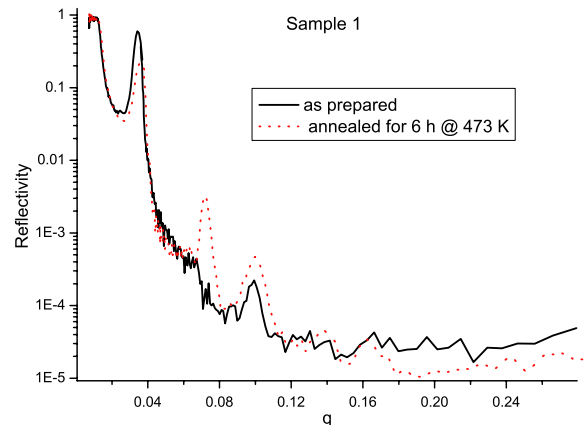


Fig. 3. Effect of annealing on sample 1, with almost no high order contributions before the annealing process. As one clearly can observe the annealing increased the higher orders intensities strongly. The temperature was 473 K during 6 h. As expected the first-order is reduced.

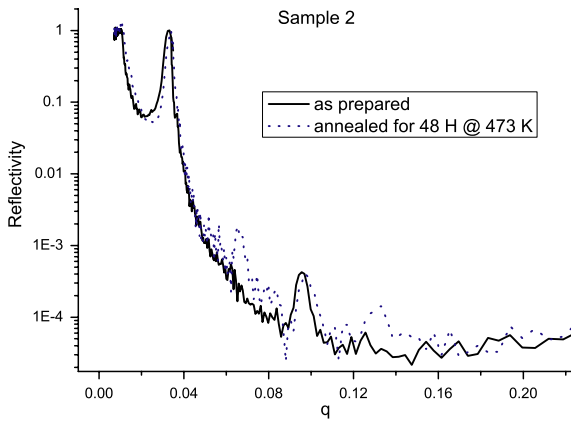


Fig. 4. Effect of annealing on sample 2, with small high-order contributions. Increase in the higher orders intensities is much smaller than in sample 1. Annealing temperature was 473 K and the time was 48 h.

5. Conclusion

Multilayers (Ni/Ti) with different scattering length density profiles $\overline{\rho(z)}$ have been sputtered. Neutron reflectivity measurements show less higher order contributions for multilayer monochromators with intermediate layers. This intensity reduction is caused by a blurring of the interfaces. The roughness at the interfaces is neither increased nor reduced and remained 4 Å after the annealing. The annealing process resulted in an unanticipated increase in the higher order intensities whereas the

first-order intensity is reduced, as expected. The reason for the enhancement of the higher order contributions is unclear. One reason for this behaviour could be the crystallization of the Ni/Ti sublayers to Ni₃Ti or NiTi₂ during the annealing at the interfaces. X-ray diffraction indicated that small crystallites are formed during the annealing of the multilayer.

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