Experimental report for beamtime CH-6200

Due to a non-functioning flood-gun we had to change sample, from non-conducting Al_2TiO_5 to a conducting CrFeNiTa alloy. HAXPES measurements of a non-conducting sample without charge neutralisation is not possible. We have very similar scientific questions for the CrFeNiTa alloy as for the planned Al_2TiO_5, so the change of sample did not affect the planned experiments. Overall this beamtime was very successful.

The studied samples were thin films of equimolar CrFeNiTa deposited on oxidized Si. The aim was to follow the amorphous to crystalline transformation of the CrFeNiTa alloy during annealing. XRD and HAXPES was measured simultaneously using a photon energy of 9072 eV. XRD was measured with 2theta angles ranging from 15 to 45 degrees. For HAXPES we measured Cr $2p_{3/2}$, Fe $2p_{3/2}$, Ni $2p_{3/2}$ and Ta 4d core level spectra as well as overview spectra. The valence band edge was visible in the overview spectra and will be used for binding energy calibration. The samples were studied in its pristine state as well as after annealing in several steps between 250°C to 750°C for 10 min each.

At all temperatures we observe a diffraction peak at a 2Theta angle of 35.5° that originates from Mo in the sample holder, see figure 1. After annealing to 350°C several sharp peaks appeared in the diffractograms, on the detector image these were visible as bright spots. Their origin is unclear, but as they were also visible outside of the sample area we believe they come from the sample holder or chamber. In figure 2 these sharp peaks have been removed. It is also unclear why they were not seen below 350°C. It could, however, be related to a re-alignment of the beam that was carried out at this point.

Figure 2 shows the results of XRD and HAXPES measured simultaneously after each annealing step. The broad diffraction peak at 37° shows that the CrFeNiTa alloy is amorphous at low temperatures. After annealing to 350°C the diffractograms change shape and the Si substrate becomes visible. At the same time a chemical shift is observed in the photoelectron spectra for Cr, Fe and Ni. The latter indicates that short-range changes are induced in the sample when annealing to 350°C. No clear additional chemical shifts are observed in the photoelectron spectra at higher temperatures for these core levels. The photoelectron spectra of Ta 4d show a more gradual shift during annealing, first to higher and then back to lower binding energies. The diffractograms clearly shows a complete crystallisation of the sample after annealing to 750°C. In addition, after longer annealing times at 750°C, we observe in the photoelectron spectra that Cr increased in intensity relative the other elements. We interpret this as elemental segregation with Cr diffusion towards the sample surface.

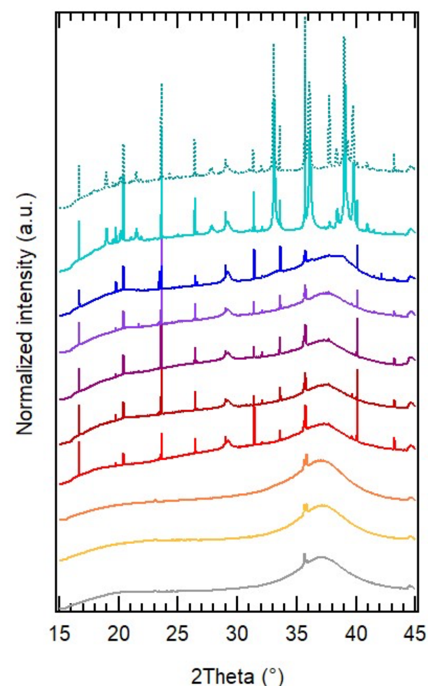

Figure 1. X-ray diffractograms as measured but shifted in y-direction for clarity.

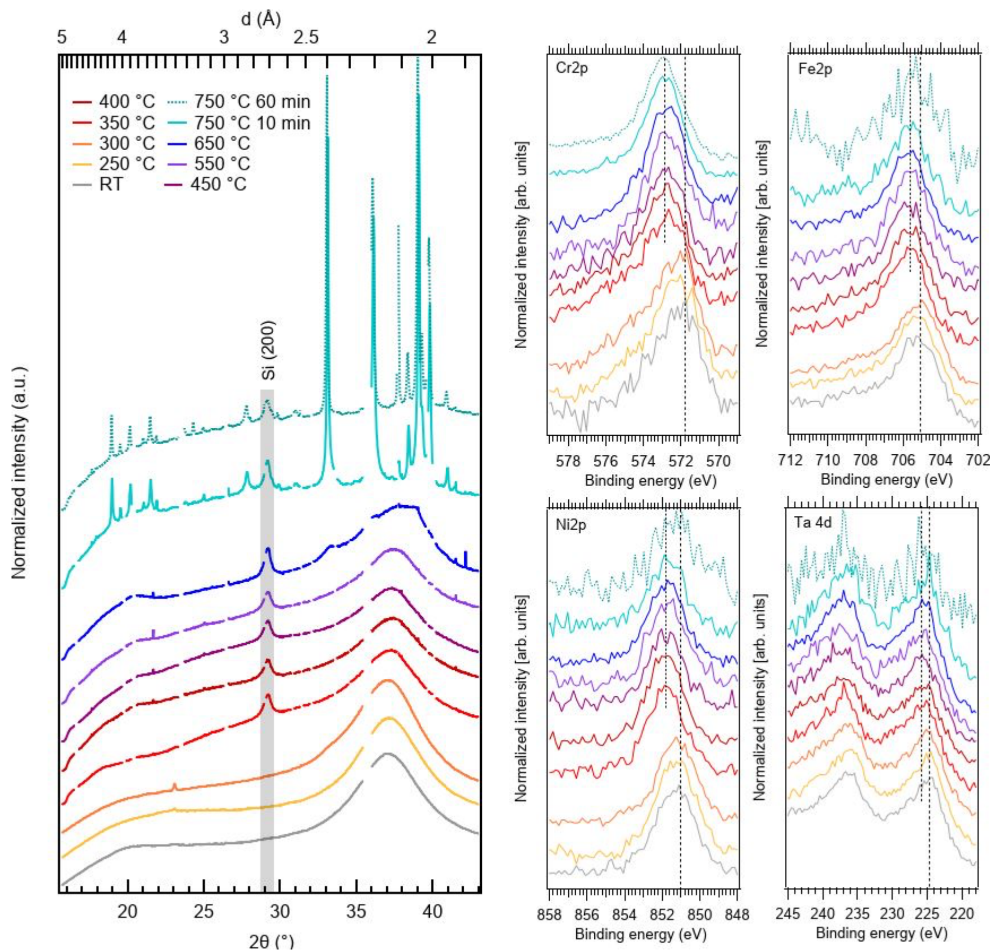

Figure 2. Left: X-ray diffractogram with peaks related to the sample holder removed. The observed Si peak originates from the sample substrate. Right: HAXPES measurements of the Cr $2p_{3/2}$, Fe $2p_{3/2}$, Ni $2p_{3/2}$ and Ta 4d core levels. Core level spectra are scaled in intensity to show the same height.