Deposition monitoring of rare earths doped silicon rich oxide films by grazing incidence X-ray fluorescence and reflectivity analysis

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MOTIVATION

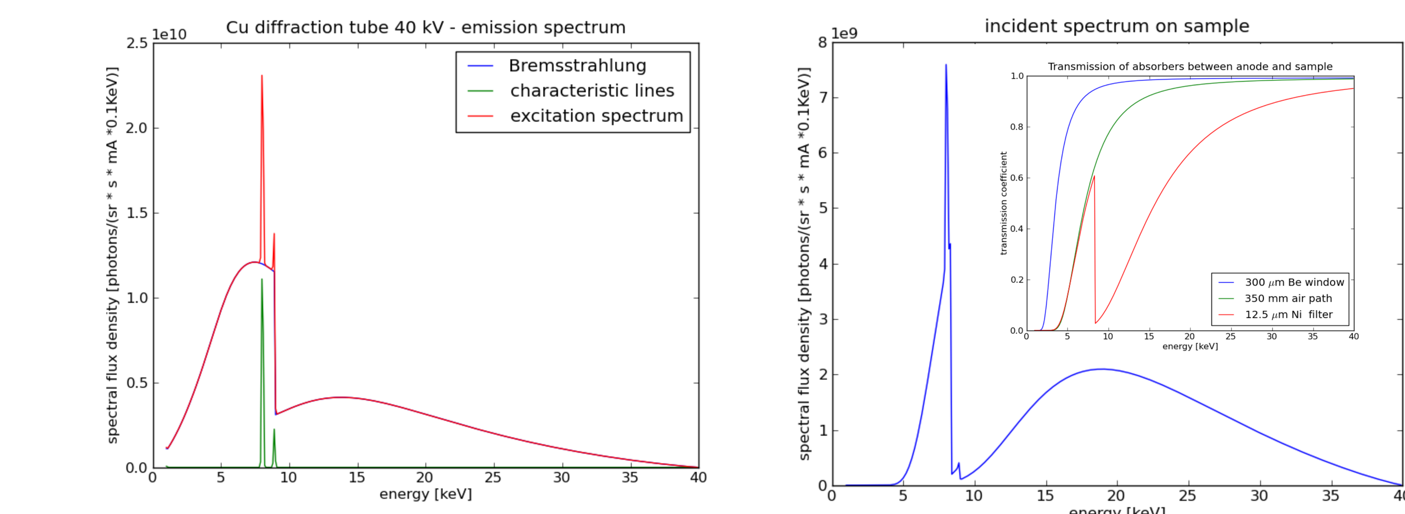
The efficiency of thin film solar cells can be improved with the addition of a photon down-conversion top layer. In this down converter layer, incident photons with energy larger than twice the band gap energy will give rise to two photons with lower energy, and then a solar cell efficiency of around 40% can be achieved.
In this work, Pr co-doped silicon rich oxide (SRO) thin films were investigated for their suitability as a down-converter layer.

GIXRF analysis

A KETEK Silicon Drift Detector (SDD) has been placed inside the cabin of a Panalytical X'Pert X-ray Diffractometer.
The detector was positioned to look in the middle of the sample aligned in the centre of rotation of the theta-theta goniometer at an angle chi=22° and an angle phi=0° fixed in the laboratory and sample frames. Acquisition of the data was performed on two different computers. The acquisition were manually started at the same time.
The acquisition was performed in continuous mode. Hence the angle of measurement for the two detectors was set as the mid motor position if the interval of acquisition and differed for the two techniques (XRR and XRF).

Excitation spectrum

The Panalytical X'Pert X-ray Diffractometer was equipped with a Cu tube operated at 40 krV and 40 mA. No monochromator in the primary beam side was present. The primary excitation spectrum was calculated according to:
Horst Ebel, X-ray Tube Spectra, X-Ray Spectrom. 28, 255-266 (1999)



Samples

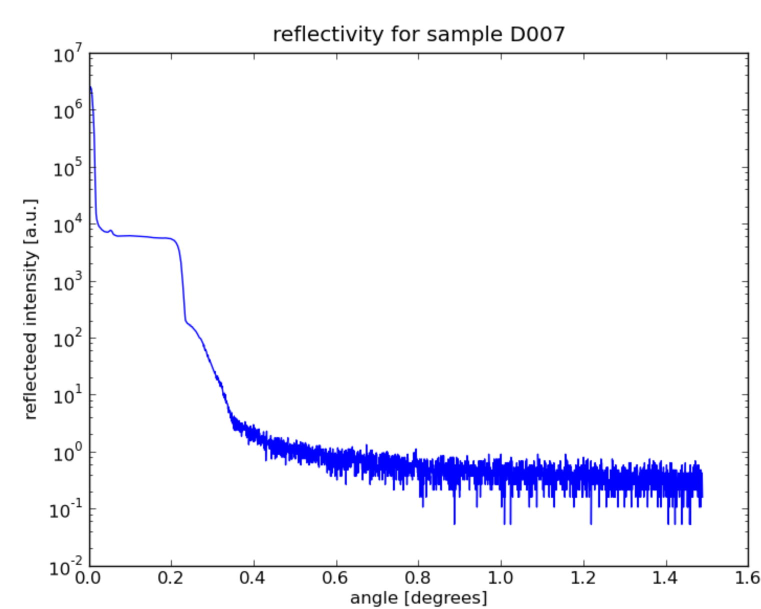
Sample were fabricated by reactive magnetron co-sputtering in two different chambers (C1 and C2) to check the deposition and check for possible contamination due do previous uses of the chambers. A pure silica target SiO_2 topped with Pr_6O_{11} pellets was sputtered.

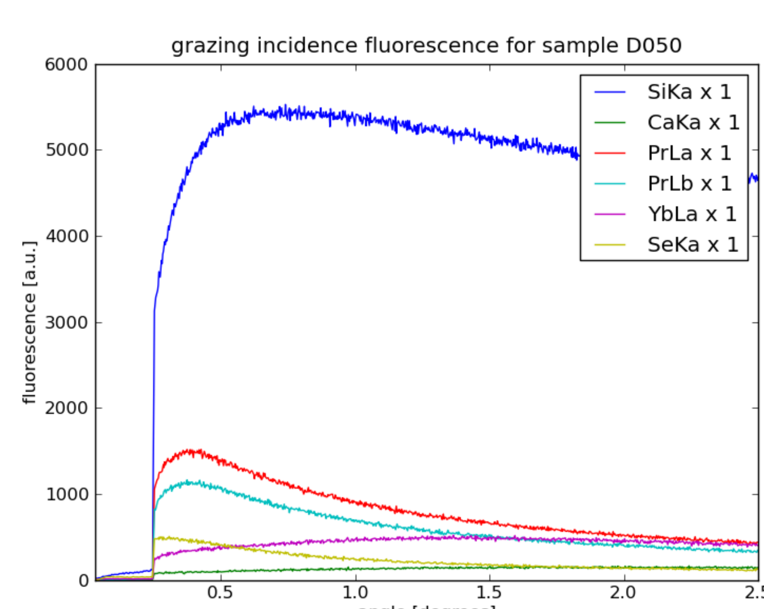
Hence some samples were annealed at different temperatures for 1 hour under nitrogen gas flow. Such treatment improves the phase separation between Si and SiO2 and allows recovering of the defects.

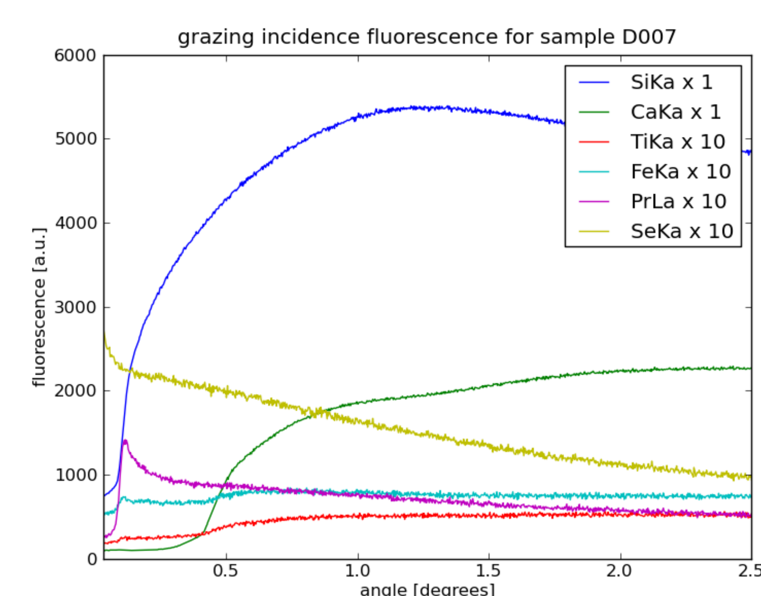
Experimental parameters are shown in the tables below.

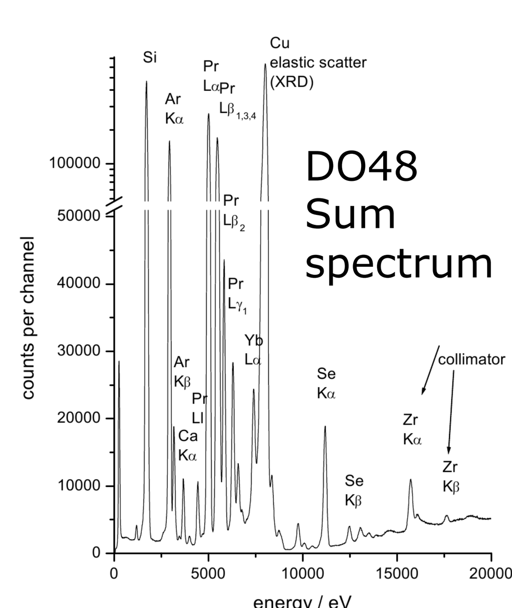
Sample name	Target type: SiO_2	Target type: Pr_6O_{11} chips	Target type: Yb_2O_3 chips	Substrate: Type	Substrate: Treatment	Deposition condition: Temp (°C)	Deposition condition: Ptot µbar	Deposition condition: rH_2 %	Deposition condition: Power (W)	Deposition condition: SRSO/SiO_2 layer
A1182	SiO_2	11	0	silicon	no	25	20	0	60	Mono SiO_2
A1183	SiO_2	11	0	Silicon	no	25	20	44%	60	Mono SRSO
A1195	SiO_2	11	0	silicon	no	400	20	44% (0% for SiO_2 layer)	60	Xc22 SRSO 2nm SiO_2 4nm
A1199	SiO_2	11	0	silicon	no	400	20	44% (0% for SiO_2 layer)	60	Xc25 SRSO 1nm SiO_2 4nm

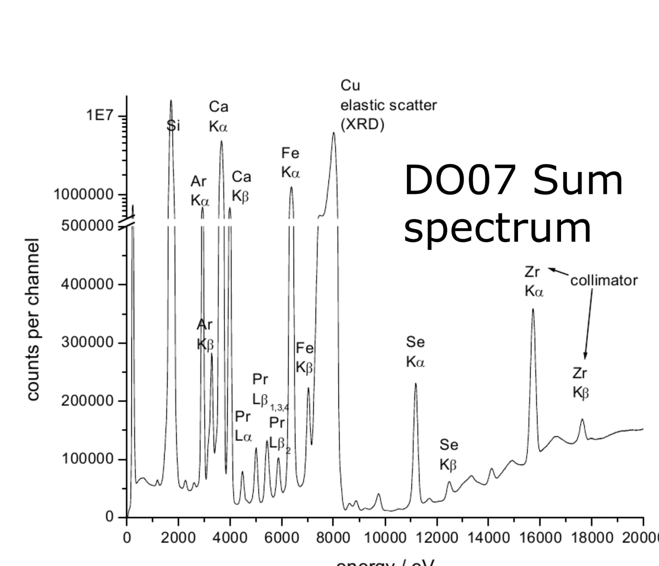
Sample name	Target type: SiO_2	Target type: Pr_6O_{11} chips	Target type: Yb_2O_3 chips	Substrate: Type	Substrate: Treatment	Deposition condition: Temp (°C)	Deposition condition: Ptot µbar	Deposition condition: rH_2 (sccm)	Deposition condition: Power (W)
D007	SiO_2	10	0	silicon	no	25	20	10	60
D022	SiO_2	17	0	silicon	no	25	20	50	100
D023	SiO_2	17	0	silicon	no	25	20	50	200
D048	SiO_2	0	0	silicon	no	25	20	10	200
D050	SiO_2	0	0	silicon	no	25	20	50	200
D054	SiO_2	0	0	silicon	no	25	20	0	200
D061	SiO_2	11	0	silicon	no	130	20	10	200
D071	SiO_2	11	0	silicon	no	25	20	10	180
D072	SiO_2	11	0	silicon	no	25	20	20	180
D080	SiO_2	0	0	silicon	no	25	20	50	60
D082	SiO_2	0	0	silicon	no	25	20	10	60

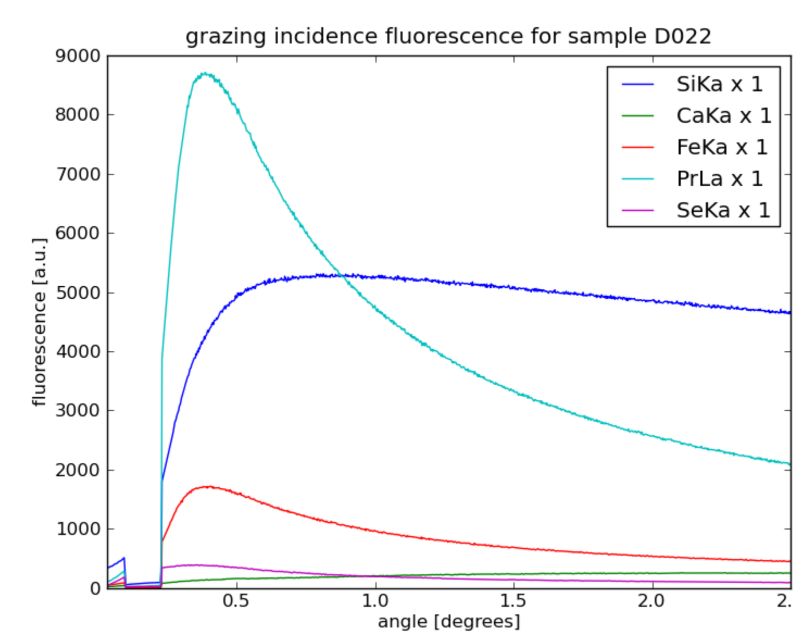


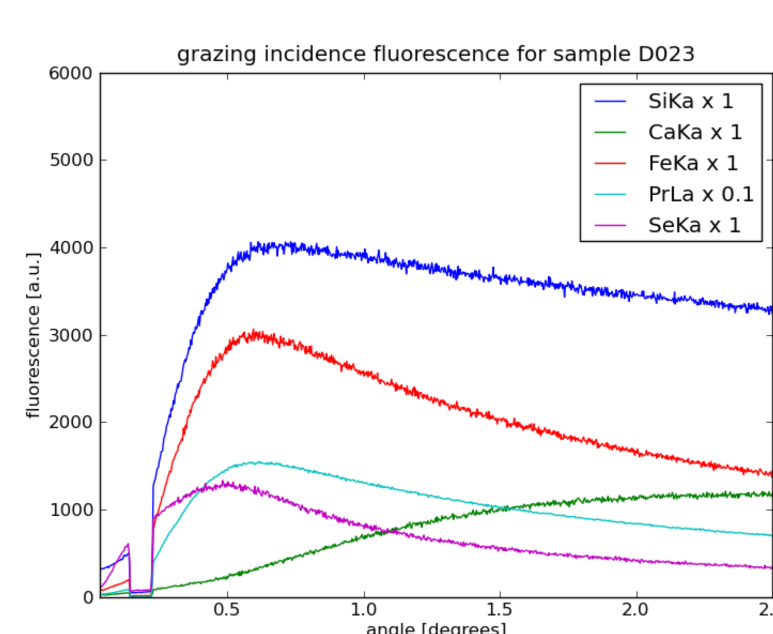


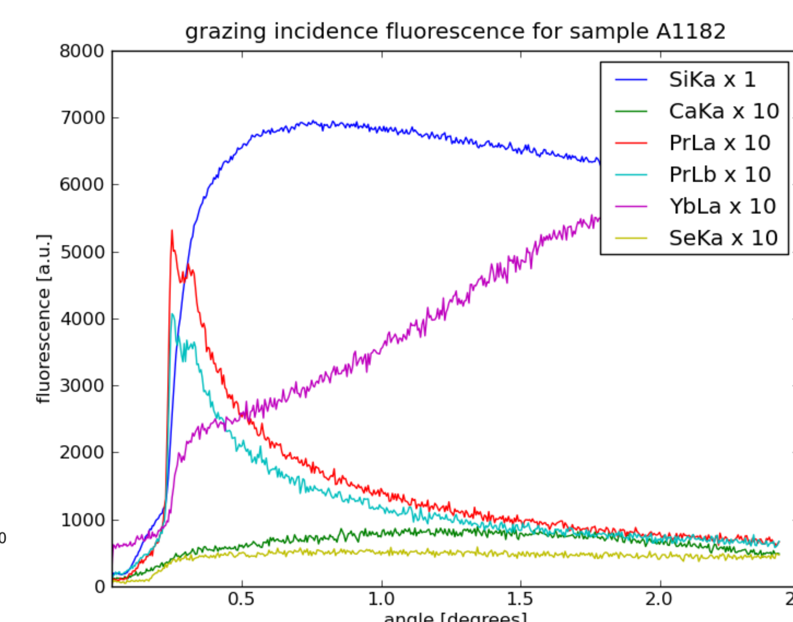


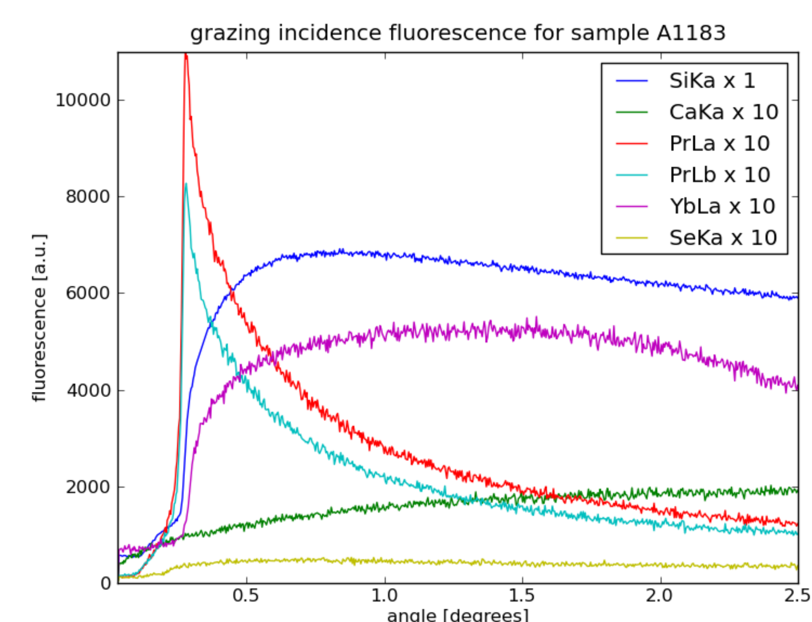












DISCUSSION and CONCLUSION

The reflectivity spectrum does not show any fringes. After long investigations this could be attributed to a misalignment of the instrument and a highbackground in the reflectivity. This strange behaviour of the reflectivity to to a primary beam not well collimated changed according to the height adjustment of the sample. The problem did not affect as much the gixrf spectra in the higher angle range.
Qualitatively the scan on D007 shows that the Pr is confined in the top layer whereas teh contaminations Ca, Ti, Fe give a signal at a higher angle indicating that they are accumulated ina deeper region, and were probably present on the substrate surface before deposition. The Selenium signal also shows a presence in the top layer but its shape is different from the Pr signal since the Pr L fluorescence is mainly excited by the Cu Kα line whereas the Se K fluorescence is excited by the high energy part of the Bremsstrahlung spectrum.

Analysis of samples D022 and D023 showed that the increase in the power caused a higher mass deposition of the Pr but also of the contamination.

Analysis of samples D048 and D050 where no Pr_6O_{11} chips where on teh target showed that Se contamination probably comes from the chamber from previous deposition. In fact also Pr and Yb could be clearly detected.

Analysis of Samples A1182 and A1183 showed no se contamination confirming that it was coming from previous work in chamber C1. A small As contamination could be seen. The distribution of Pr in these samples was much more peaked at low angles showing that the Pr accumulated in a region much closer to the surface then samples deposited in chamber C1.

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