



Experiment title: Structural dynamics of photoexcited uranyl(VI) halide complexes

Experiment number:
CH-4567

Beamline:
ID09B

Date of experiment:
from: September 15, 2016 to: September 19, 2016

Date of report:
February 26,
2018

Shifts:
18

Local contact(s):
Dr. Denis Leshchev

Received at ESRF:

Names and affiliations of applicants (* indicates experimentalists):

S.Tsushima*, Institute of Resource Ecology, Helmholtz-Zentrum Dresden-Rossendorf, Dresden, Germany

D.Khakhulin*, European XFEL, Germany

K.Kvashnina, ESRF BM20

Report:

We performed pump-probe experiments to observe the photo-excited states of uranyl(VI). To be more specific, we studied the photoexcited states of uranyl(VI) in the presence and absence of a halide quencher by means of time-resolved wide angle X-ray scattering (TR-WAXS) in solution. Based on a series of density functional theory (DFT) calculations we predicted that the quenching in uranyl(VI) halide involve electron transfer from halide to uranium and concomitant structural rearrangement. The idea of the experiment was to confirm this hypothesis. Uranium concentration of the samples (50 mM) were adequately optimized so that at least 10–20% of the excited state population would be expected. We also made theoretical calculations of the WAXS signal and concluded that these experiments were feasible (see proposal).

Despite of the extensive efforts, no meaningful signal was detected (see Fig.1). The reason for this could not be narrowed down. It can be that the intensity of the laser was still not strong enough to get high amount of excited states although we basically worked with the maximum laser power. Another possibility is that the thickness of the glass capillaries. For safety reason, we were obliged (by the ESRF safety standard) to use double confinement, and the wall thickness of the glass capillaries were 0.01 mm and 0.2 mm. The latter could have been too thick.

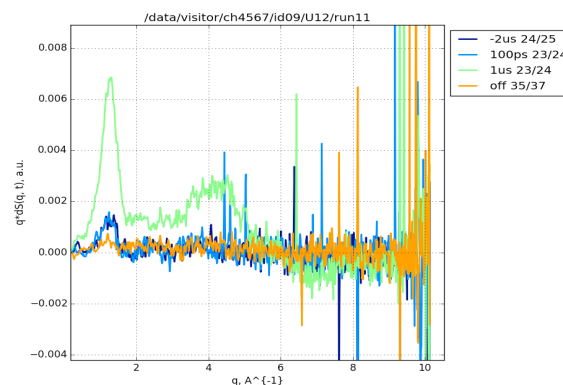


Figure 1 Measured WAXS signal for sample U12 (U50mM TBABr 250mM in CH₃CN)