ESRF	Experiment title: Implementing a laser pump x-ray probe technique on a sub-nanosecond time-scale	Experiment number: CH 587
Beamline: ID 09	Date of experiment: from: 10.07.1999 to: 16.07.1999	Date of report: 28.02.2001
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Report:

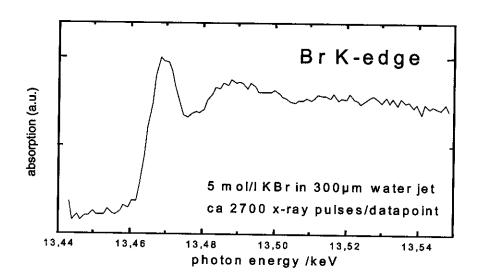
This project aims at probing real-time photoinduced structural modifications in a large class of media from simple systems as KBr/H₂O to highly complex biological systems as Hb-H₂S in its physiological medium. The techniques of choice are time-resolved x-ray absorption spectroscopies (Extended X-ray Absorption Fine Structure-EXAFS or X-ray Absorption Near Edge Structure-XANES) in the picosecond time domain.

The principle of the experiment is based on the pump-probe scheme, where an ultrafast laser pulse induces a structural modification in a chemical, physical or biological system. The picosecond hard x-ray pulse from a synchrotron source probes the evolution of the structure in real-time by means of an adjustable time delay between the pump and the probe pulse.

Our model system for the reported beamtime is 5mol/l aqueous solution of KBr in a liquide jet of 0.3mm thickness. An ultrafast laser pulse (400nm, 1.8 x 10¹⁴ photons/pulse (=90µJ), 100fs) generates Br radicals via 2-photon photoionisation. The time-delayed x-ray pulse (13.2-13.5keV, 2 x 10⁵ photons/pulse, ca.100 ps pulsewidth) probes the modified absorption features around the bromine K-edge. XANES is expected to show a photoinduced

pre-edge resonance (1s \rightarrow 4p) and EXAFS will exhibit a modified fine structure due to photoinduced changes of the solvent cage around the nascent bromine radical.

The following spectrum of laser pumped KBr/ H_2O was obtained by averaging ca. 2700 x-ray pulses (corresponding to 3 sec accumulation time) per datapoint, using a slow ion-chamber as I_0 and a gated avanlanche photo diode (Hamamatsu S2383) as I_1 , which records one synchronized x-ray pulse at 896 Hz.



Our calculation (based on the scheme presented in [1]) of the expected pump-probe signal for the $1s \rightarrow 4p$ white line predicts a signal-to-noise ratio of 9.8 after 2700 pump shots, i.e. an observable signal in principal. However, the recorded noise on the spectrum was 300 times larger than the shot-noise limit due to unknown additional electronic noise sources. Thus, our experimental sensitivity was too low for the detection of a photoinduced effect on the absorption spectrum. We have used the same electronic setup with the same instrumentation at other beamlines before, and we obtained spectra with signal-to-noise ratios of only 2-3 above the shot-noise limit. Therefore we conclude that the noise source is inherent at ID09 and not due to the chosen detection scheme.

^[1] C. Bressler, M. Saes, M. Chergui, R. Abela, P. Pattison, submitted to: Nucl. Instr. Methods A (2000)

^[2] M. Chergui et al., ESRF annual report 1998:"Optical X-ray pump-probe study of the vibrational and electronical relaxation of I₂ in liquid solvent"