Ultra-High Time Resolved XAS

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A pump-probe setup for transient x-ray absorption spectroscopy for the study of ultrafast phenomena like the transfer of angular momentum between spins and lattice in ferromagnetic and other materials has been built recently at BESSY. The setup is particularly devoted to X-ray magnetic circular dichroism (XMCD) measurements as the difference of X-ray absorption between left and right circularly polarized X-rays in ferromagnets on a ~100 fs timescale [1].

Short pulses are possible at BESSY II in a single-bunch mode (~ 50 ps) and in the “low-alpha” mode down to 1-3 ps. A temporal resolution of <120 fs was achieved at BESSY with the Femtoslicing Source [2,3]. Here the electron pulse is “sliced” with a femtosecond optical laser where the field of the laser pulse causes an energy modulation in the electron bunch. In the BESSY scheme the energy-modulated electrons are separated from the main pulse in a dipole magnet and subsequently generate fs pulses by passing through a helical undulator.

A major advantage of the Femtoslicing scheme is the perfect synchronization between fs laser and fs X-ray pulses. This is due to the fact that the fs optical pulses from the same laser source are used both for the slicing and the pump processes. The temporal phase between the laser pulse and the energy modulated electrons has to be preserved during propagation from the modulator to the sample. This translates into controlling the optical path length difference between optical and X-ray pulses. The femtoslicing radiator is optimized for polarization dependent spectroscopy in the soft X-ray range and yields 10^6 ph/s/0.1%BW. Behind the plane grating monochromator the fs X-ray flux for 0.1 % bandwidth is reduced to $<10^4$ photons per second which sets tight constraints for the feasibility of certain experiments. This leads to the necessity of highly efficient optics to transfer X-rays to a sample with minimum losses. The optics must perform a focusing of X-rays onto the sample, provide reasonable energy dispersion and has to have a highest possible x-ray transmission. The solution was found using only a single optical element – a reflection zone plate (RZP).

A setup for laser-pump x-ray probe absorption spectroscopy is shown in figure 1. X-ray absorption spectra are recorded by measuring the transmitted X-ray intensity by an avalanche photodiode [4].

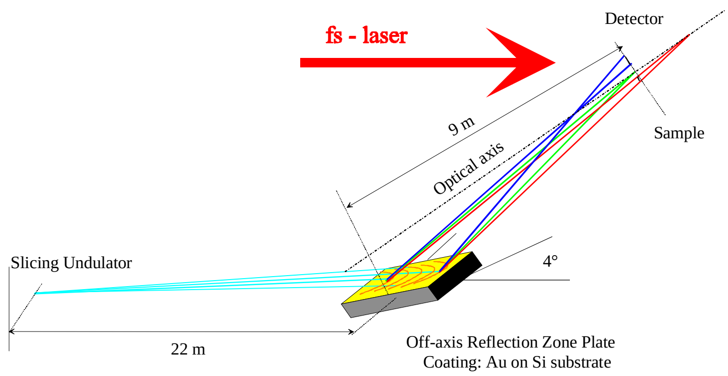


Fig. 1 Optical layout of the experiment.

The undulator source UE56 covers an energy range between 50 eV and 1400 eV and full polarization control. The polarization can be varied between linear (horizontal or vertical) and left and right handed elliptical. A RZP under total external reflection conditions features the important property of not changing the polarization of the beam.

The elliptical zone plates fabricated on a super-flat reflection mirror surface were effectively used for an x-ray monochromatization and beam focusing at photon energies below 1500 eV. This element can be applied in the beamlines with specific beam conditions such as very high heat load or very low flux. Since the whole setup involves only one reflection it meets the requirement of minimum losses. It is important, that the RZP must be off-axis, to provide the best energy and spatial resolution.

Another demand of slicing users is the preservation of the time resolution from the source. The residual temporal blurring of the short x-ray pulse is in our case a characteristic time of the order of 30 fs. The number of grooves in the diffraction structure along the beam direction should not exceed the value of 5200. This value defines a maximum aperture and maximum energy resolution of the optical element.

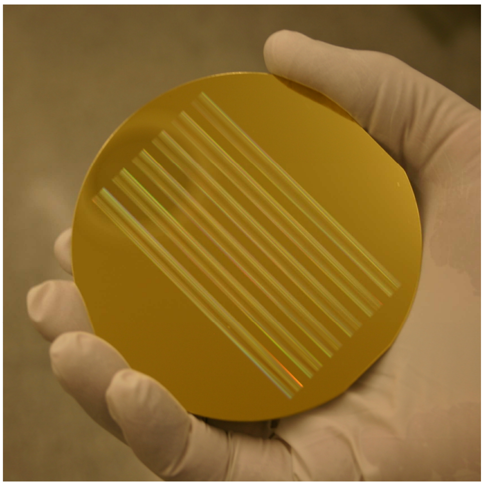
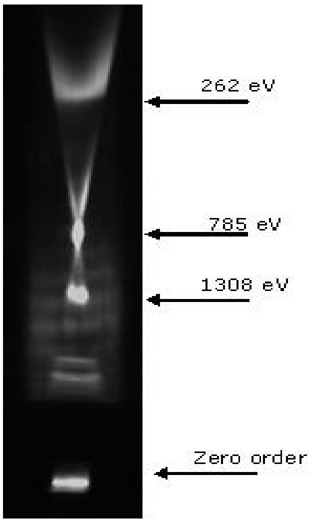


Fig. 2 Reflection zone plates on a Si substrate.

The reflection Fresnel zone plates were produced at the BESSY Centre for Microtechnology in cooperation with Microlithography and Consulting (ML&C) Company in Jena. Six Fresnel lenses designed for the edge energies between 500 eV and 1200 eV were fabricated on the same substrate (figure 2).

The monochromator was tested both with direct undulator beam and white light radiation operating at detuned gap settings. With direct undulator radiation a fluorescence screen was used to visualize an intensity distribution in the focal plane of the reflection zone plate. The result is shown in figure 2 which represents an image obtained on a fluorescence screen with light containing three undulator harmonics: the first at 262 eV; the third at 785 eV and the fifth at 1308 eV. The RZP is optimized for the energy of 765 eV and produces a focused spot at this energy. The other lenses produce a similar distribution in the focal plane but are optimized for five other photon energies. It must be mentioned that the beamline has only one optical element, therefore the total flux delivered by the beamline is of the order of 20 times higher than the photon flux from a traditional plane grating monochromator (PGM) beamline with a minimum of four reflecting surfaces. In figure 4 a comparision of absorption spectra from the L3 and L2 edges of Co is shown.



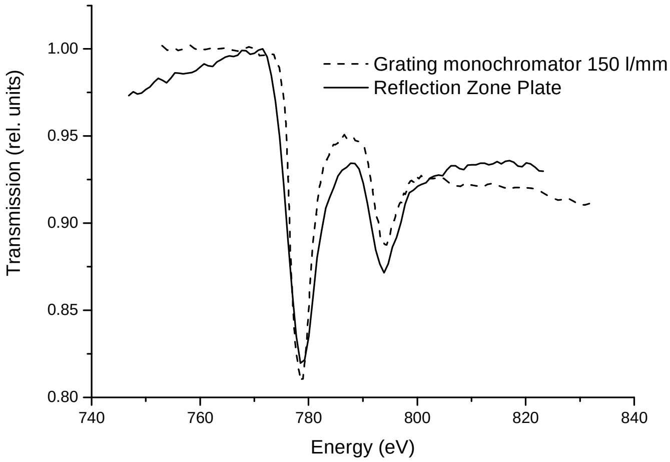


Fig. 3 Footprint of reflected light from the RZP in the focal plane on a P43 phosphor screen.

Fig. 4 L-Edge Absorption spectra of a 30 nm thin film of Co taken at different beamlines under identical conditions.

One can see the identical quality of the (single sweep) spectra regarding energy resolution. The difference is that the RZP monochromator the higher flux and easily allows for a parallel registration of the absorption spectra. Specially developed avalanche diodes arrays, consisting of 25 single diodes were tested to record simultaneous spectra from the full dispersion area. An optical fiber is used to improve the spatial resolution of the diode line array down to 50 µm, which corresponds to an energy resolution in experimental geometry of 1.2 eV. The gain in flux of more than order of magnitude has allowed to address a new class of ultrafast user experiments within the past year. Very recent examples will be presented.

References:

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