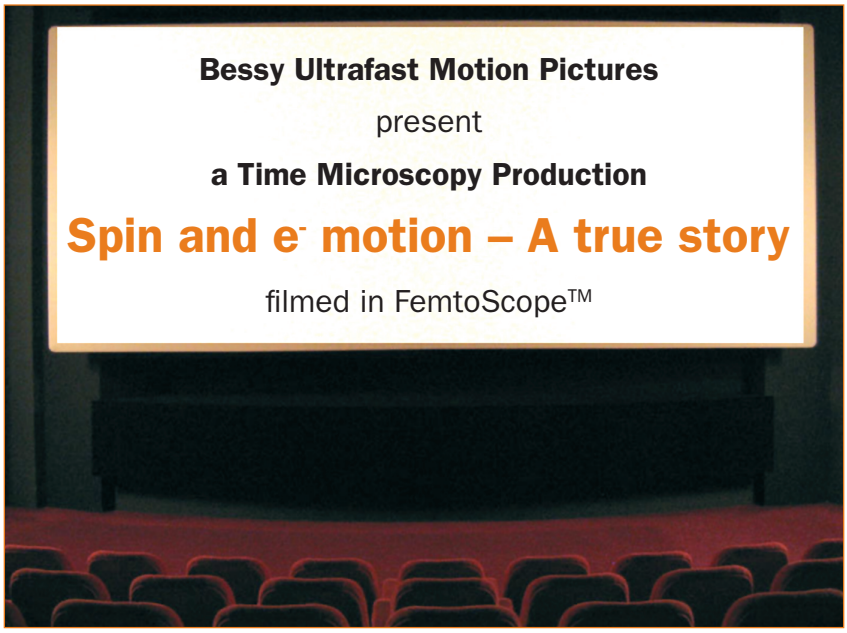
Chirping violins suggest the end of a movie in the seventh heaven. The couple got together eventually. How? You do not know since you had fallen asleep after the first sequence when he started to write an email … Well, never mind: You have something in common with physicists and chemists! They often know from where a process like a chemical reaction started and where it ends, but they neither know the storyboard. You would just have to stay awake but for them it is more complicated. Scientists need very short and very bright X-ray pulses to watch their movies.

On a timescale of 20-100 femtosceonds (fs) the atomic nuclei 'stand still' while the much lighter electrons are still moving. Spectroscopy on the fs time scale is, therefore, able to detect the breaking and formation of electronic bonds as the nuclei move during a chemical reaction. In magnetic solids the electronic orbital motion is intimately linked to spin motion. Such spin-orbit forces ultimately determine the direction of spontaneous spin order as well as energy barriers for magnetization reversal especially of nanometer scale devices. Spectroscopy of spin and orbital motion on the fs timescale, therefore, opens up radically new ways for determining the ultimate speed of magnetization reversal of bits in magnetic data storage devices. On a microscopic level it is of interest to observe the transfer of energy and angular momentum between electronic, spin, and lattice degrees of freedom in atoms, molecules and solids. The use of optical fs laser excitation leads to the fascinating prospect of observing these processes in real time by stroboscopically 'imaging' the electron and spin motion, thus, separating the influence of lattice vibrations.

X-rays as a probe are especially suited for this purpose due to the inherent element specificity by involving electronic core levels. Resonant core-valence excitation also enables an orbitally resolved probe of the electronic structure. X-ray polarization control finally is the key ingredient in obtaining magnetic sensitivity which is nowadays routinely employed at 3rd generation synchtrotrons for separating spin and orbital components of the magnetic moments. However, researchers were limited in time resolution due to the pulse length available at synchrotron radiation sources. 'To watch' the ultrafast changes in magnetization or chemical reactions shorter X-ray pulses are required.

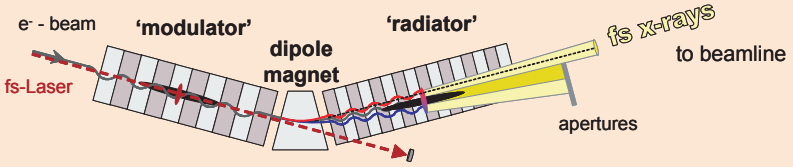


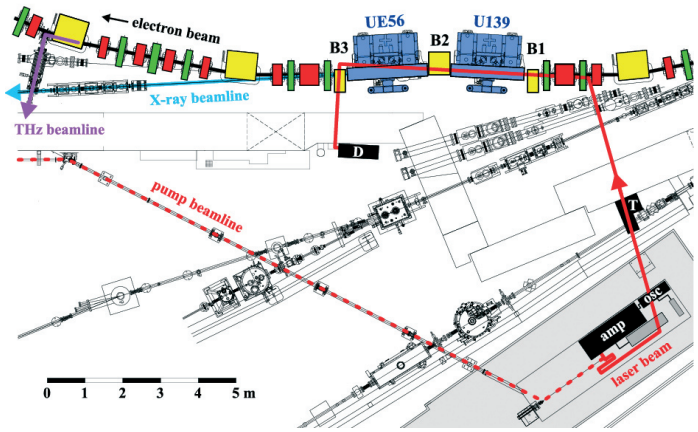
Synchrotron radiation meets fs laser

Synchrotron sources do not radiate their X-rays continuously but rather produce series of short pulses. In time-resolved experiments the shortest observation time is given by the duration of the X-ray pulses. At present 3rd generation synchrotron sources, X-ray pulses are typically 50 picoseconds (ps) long. Shorter pulses down to just a few ps are available at BESSY during special operation conditions, the so-called 'low-α' mode. The ultimate temporal resolution of <120 fs is reached with the Femtoslicing Source.

Femtoslicing scheme

Femtosecond pulses from a Ti:sapphire laser system (wavelength λ = 780 nm, pulse duration τ = 50 fs FWHM, pulse energy E = 2 mJ, repetition rate = 1 kHz) co-propagate with electron bunches in a planar U139 undulator (the 'modulator'). The electric field of a 50 fs laser pulse induces an energy modulation of up to ± 1% to the 1.7 GeV electrons within an ultrashort 'slice' of the 50-70 ps wide bunch. Within our scheme the energy modulated electrons are separated from the major (99.9%) non-sliced part of the bunch by a dipole bending magnet. It serves for angular separation within the helical UE56 soft X-ray source (the 'radiator'). Picosecond radiation from the main bunch is blocked by front end apertures.





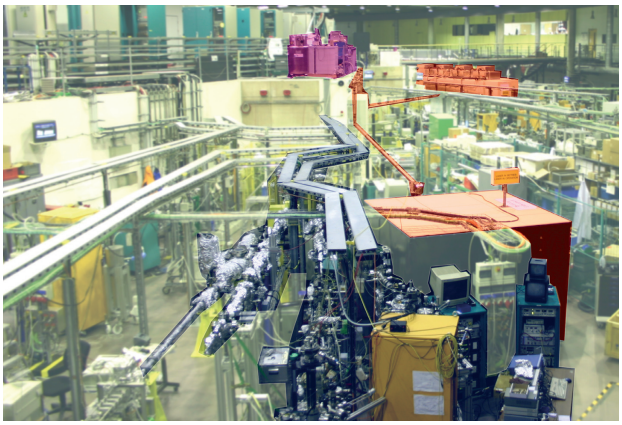


Fig. 1:
Left: Floorplan of the Femtoslicing source. The fs laser beam (red) is guided into the tunnel for 'slicing' the electron hybrid bunch within the U139 modulator. The 'pump'-part of the laser beam is transferred to the experiment (not shown).

Right: Photo of the installation with the experimental chamber in front, the laser hutch in the back (orange) and the THz setup (violet).

Here, the electron pulse is 'sliced' with an optical femtosecond laser where the field of the laser pulse causes an energy modulation in the electron bunch [1]. 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 (see box: Femtoslicing scheme). The price to be paid for the now improved time resolution (at storage rings) is a dramatic reduction in X-ray intensity since only 10^{-3} of the electrons of the initial bunch are involved in the fs-pulse generation.

A major advantage of the Femtoslicing scheme is the inherent rigid temporal correlation between fs laser and fs X-ray pulses. This is due to the fact that the fs laser pulses used for the slicing and the pump processes originate from the same laser pulse (see box: experimental setup). 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 by a distance better than 30 µm (corresponding to 100 fs) over a distance of typically 50 m (Fig. 1).

The implementation of the Femtoslicing Source started in 2004 with the insertion of the two undulator pieces (U139 and UE56) and the set-up of the high power laser [2]. Beyond these challenges, the key ingredient of getting the facility up-and-running was to achieve the overlap of the electron bunch and the laser pulse in the modulator. It turned out that our experiences with coherent synchrotron radiation and THz radiation played a major role in opening the door to the fs-pulses.

THz-Flashes

When the (optical) laser pulse hits the electrons, the path length differences of energy-modulated electrons also create a sub-ps density modulation in the longitudinal bunch profile, which gives rise to intense coherent THz pulses [3].

Using a dedicated THz beamline at a bending magnet 11 m downstream of the modulator (Fig. 1), strong pulses in the range up to 3 THz appear with each laser shot. The detection of the THz signal is a prerequisite for the generation of fs X-ray pulses. Featuring a high dynamics, these ultrashort THz flashes indicate a successful resonant interaction in the modulator, where the two beams (laser and electron beam, both having a diameter comparable to a human hair) have to co-propagate over a few meters with a temporal overlap of only some picoseconds.

We found at BESSY, that a THz flash returns after subsequent turns of the bunch along the storage ring lattice (240 m). The ratio of the THz intensity from the first and second turn sensitively depends on the initial energy modulation $\Delta E/E$ of the electrons, a phenomenon which can be explained by momentum compaction and a resulting longitudinal expansion of the initial density modulation on the bunch. This 'afterglow' of the THz flashes is routinely used to maximize the energy modulation process prior to fs X-ray experiments on a shot-to-shot basis. The spectral shape of the THz flashes as measured by advanced rapid-scan Fourier Transform Spectroscopy techniques is employed to pre-tune the ~100 fs length of the slice and hence, the resulting length and intensity of the fs X-ray pulses (see box: THz pre-tuning).

During routine operation of the fs X-ray facility, the laser-electron interaction is now stabilized over many days correcting for long term transverse drifts between laser- and electron beam and synchronous phase shifts

of the order of 20 ps of the decaying bunch. This is performed by feedback loops to keep the THz signal maximized by controlling two laser mirrors and the laser-electron timing.

Properties of fs X-rays

The UE56 femtoslicing radiator is optimized for polarization dependent spectroscopy in the soft X-ray range. The fs X-ray flux for 0.1% bandwidth is of the order of 10^4 photons per second. Our fs undulator source allows for wavelength tuneability between 300 and 1,200 eV photon energy and for full polarization control. The polarization can be varied between linear (horizontal or vertical) and left and right handed elliptical.

The horizontal angular separation has been shown a successful concept of suppressing non-sliced undulator radiation from the desired fs soft X-rays. By choosing appropriate settings of the beam bend in the dipole magnet and the front end beamline apertures, we have succeeded to obtain very good fs-signal/ps-background ratios (up to 100:1) over the full photon energy range for either linear or elliptical polarization.

The UE56 X-ray source can still act as a **normal BESSY undulator** when the laser is switched off and using 0° beam bend allowing for pump-probe experiments with 70 ps temporal resolution and significantly higher photon flux, which is also an important feature for setting up a new fs experiment.

Fs electronic motion in metals

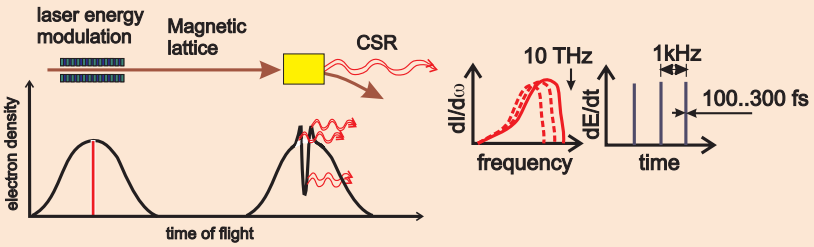
As a first proof-of-principle experiment utilizing full polarization control of fs X-rays at the Femtoslicing Source we studied the ultrafast transfer of angular momentum between spins and lattice in ferromagnetic materials.

When energy is pumped into the electronic system of a metal by absorption of a fs laser pulse, electrons are excited into unoccupied states above the Fermi level. These electron-hole excitations will ultimately decay into phonons on a characteristic timescale given by electron phonon coupling (~1 ps). On intermediate times of several 100 fs an increase of the electronic temperature of a metal is observed while the lattice remains cold. This effect can be observed in fs X-ray absorption spectroscopy (Fig. 2). The X-ray absorption especially at the threshold energy changes during laser heating. The observed rise time is given mainly by the X-ray pulse duration since the response of the electrons to laser excitations is essentially instantaneous. Increasing the X-ray pulse length from the Femtoslicing (<120 fs) to low-α mode (~10 ps) and standard operation (~50 ps) is clearly seen in the decreasing slope of the so-called pump-probe cross-correlation scans (Fig. 3).

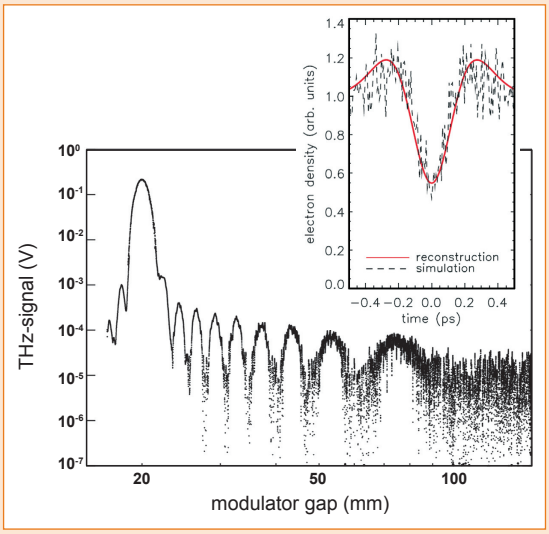
On ~1 ps timescale the energy deposited in the electronic system is dispersed to the lattice, this relaxation process can be seen in Fig. 3 at corresponding times. On closer inspection the fs changes of X-ray absorption spectra cannot be explained by a heated electronic system alone. An increased electronic temperature would result in a broadening of the electron distribution at the Fermi level. This was clearly observed in time-resolved photoelectron spectroscopy with fs laser radiation [4]. Naively we would

THz pre-tuning

A coherent THz pulse is emitted on a bend magnet (THz-radiator, yellow) by a 'dip' in the electron bunch, left behind by the sliced electrons after propagating the magnetic lattice. Since other electrons emit only weak incoherent radiation in that spectral range, the coherent flash exceeds the radiation from all other bunches by many orders of magnitude. The spectral shape of the THz flashes is essentially the Fourier Transform of the 'dip'. FTIR spectroscopy is employed to pre-tune the **~100 fs** length of the slice and the initial energy modulation



Total THz pulse energy (emitted by a 'dip', i.e. electron-density modulation measured in the inset) while the resonance at 19.85 mm gap of the U139 has been slowly detuned to a maximum gap of 150 mm. The periodicity of the side bands corresponds to λ/N, where N=10 is the number of periods and λ the fundamental wavelength. The measurements demonstrate that the resonance can be alternatively described as a convolution of the spontaneous undulator field and the laser field. Even if the undulator field is many orders of magnitude smaller, an interaction can still be detected. This high sensitivity of the THz signal is a key to establish and maintain the overlap between laser and electron beam necessary for fs X-ray experiments.



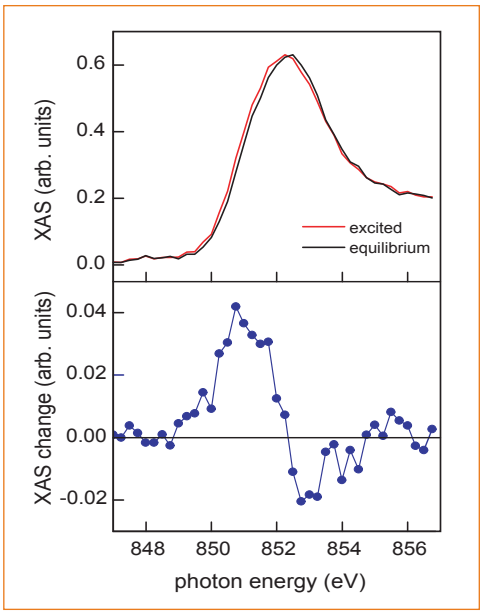


Fig. 2:
Top panel: Ni L_3 absoprtion edge studied with 120 fs soft X-ray pulses. Red: IR laser pumped Ni film, black: laser off.

Bottom panel: The difference between the laser pumped and not-pumped absorption spectrum reflects the observed peak shift of the Ni L_3 edge.

expect to observe a similar broadening of the leading edge in X-ray absorption spectroscopy of Ni metal. This is not observed as can be seen in the time-resolved X-ray absorption spectra in Fig. 2. Instead the fs laser excitation leads mainly to a rigid shift of the absorption threshold towards lower X-ray energy. This is typical for a core level shift as it is observed for differing chemical environments. These data are, therefore, the first indication of a transient core level shift due to fs laser heating in metallic Ni. Presently we can only speculate about the microscopic mechanism for such an effect. It seems, however, reasonable to assume that the scattering of laser excited electrons could lead to an increased localization of itinerant Ni valence electrons.

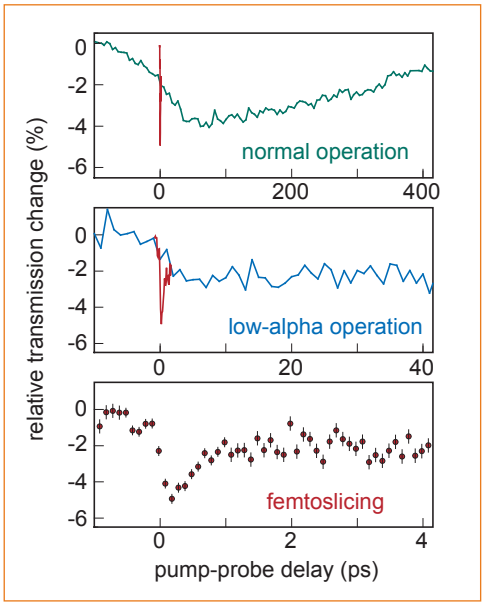


Fig. 3:
Transient shift of the Ni L_3 egde measured at the maximum of the laser induced difference curve (Fig. 2, bottom panel) vs delay time between laser pump and X-ray probe pulse. The bunch lengths are: 50 ps (top), 10 ps (middle) and 120 fs (bottom), with the latter superimposed on the two upper graphs (red line).

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Ultrafast heating of a ferromagnet

Optical transitions are usually spin conserving, even in a ferromagnetic material. In this respect the first observation of fs laser induced demagnetization of ferromagnetic Ni [5] has attracted enormous interest. Applications such as magneto-optical recording could become feasible on sub-ps timescales. These results raise the fundamental question about angular momentum conservation on ultrafast timescales. Since the electron spin represents an angular momentum quantum, any quenching of spin angular momentum by fs laser excitation must lead to an increase of angular momentum in another particle reservoir. It has been argued that spin angular momentum can be transferred to the lattice only on significantly longer timescales of ~100 ps [6]. On the fs timescale quenching of spin angular momentum should then be accompanied by an increase of orbital angular momentum. In the ground state the orbital motion of electrons contributes by only ~20% to the total magnetic moment in 3d transition metal ferromagnets. It could, therefore, act as a sink for angular momentum even on the fs timescale.

X-ray magnetic circular dichroism (XMCD), i.e. the difference of X-ray absorption between left and right circularly polarized X-rays in ferromagnets, is the ideal tool to observe such effects. For instance, at the Ni L_3 absorption edge sum rules relate the XMCD intensity to linear combination of spin, S, and orbital, L, angular moment as 2S+3L. The fs temporal evolution of the quantity, 2S+3L, following the absorption of a fs optical laser pulse. It represents the first direct demonstration that S is transferred to the lattice and not to L on a ~100 fs timescale (Fig. 4). This can be visualized for the following scenario, considering that in thermal equilibrium (at negative time delays in Fig. 4) L is typically only about 20% of S. The ~80% decrease of 2S+3L during the first ps is then mainly due to the reduction of S. If this change in S would be completely compensated by L, the quantity 2S+3L would actually increase by about 20% in contradiction to the measurements. The important conclusion drawn from the data in Fig. 4 is the existence of an ultrafast channel for transfer of spin angular momentum to the lattice. In analogy to the well known Einstein-de Haas-effect, a small suspended ferromagnet would start to rotate upon laser induced demagnetization (schematically shown at the top of Fig. 4).

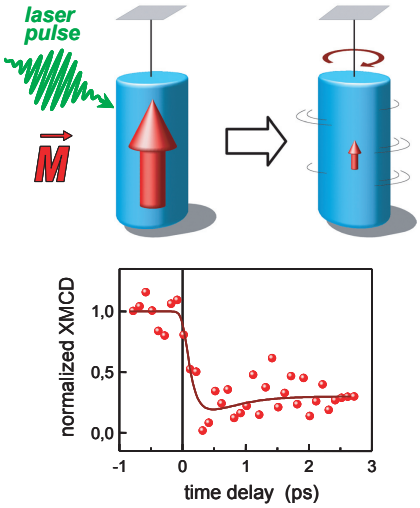


Fig. 4:
Transient normalized XMCD of the Ni L_3 edge vs delay time between laser pump and X-ray probe pulse. The curve is taken with elliptically polarized fs soft X-rays. The upper part illustrates the concept of angular momentum conservation upon laser induced demagnetization (Einstein-de Haas-Effect).

Where we are and where to go ...

Employing X-ray pulses of fs duration from the BESSY Femtoslicing Source and the laser for well correlated pump-probe experiments, we were able to demonstrate an ultrafast transfer of angular momentum between spins and lattice in ferromagnetic materials. This allows a glimpse at the future use of intense fs X-ray pulses from the planned BESSY soft X-ray Free Electron Laser source.

The results also show that the required level of stability of the laser-synchrotron setup can indeed be achieved. The Femtoslicing Source can be considered as a new user facility allowing for pump-probe experiments with X-rays (between 300 and 1,200 eV photon energy) and THz radiation on a sub-ps timescale

For more sophisticated experiments involving more intense pulses, however, it will be of paramount importance to avoid the intensity losses due to slicing. This can be achieved using linear accelerator based X-ray sources since the electron bunches are used only once for X-ray generation and can, therefore, be compressed down to sub-ps duration. In free electron laser (FEL) sources, the bunches contain enough electrons so that the resulting ultrashort X-ray pulses become coherent like a conventional laser.

In the proposed BESSY soft X-ray FEL, the lasing process is induced by seeding the electron bunches by external optical fs laser pulses. This new FEL principle utilizes the fact that the time structure of an optical fs laser pulse can be 'imprinted' upon an electron bunch in a way very similar to the Femtoslicing principle. In a sense the BESSY Femtoslicing Source represents the first step towards a seeded soft X-ray FEL including the required development of suitable electron bunch diagnostics capable of measuring a fs charge modulation of electron bunches.

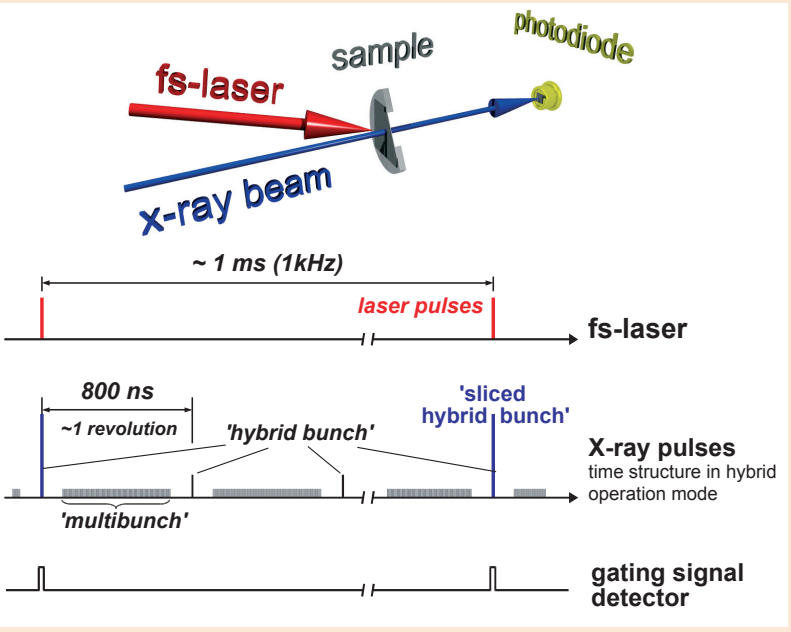
So, while we are waiting to be able to watch the whole movie using Free Electron Lasers, with Femtoslicing we can now get the first glance of the storyboard ... and this is already very exciting.

Experimental setup

A scheme of the experimental XAS setup is shown in the upper part of the figure. A 30 nm Ni-film deposited on a 500 nm thick aluminum membrane is carried by an aluminum frame. X-ray absorption spectra of the Ni-film are recorded by measuring the transmitted X-ray intensity by an avalanche photodiode. For XMCD measurements, an external magnetic field can be applied to the sample by an electromagnet.

Laser-induced ultrafast excitation and subsequent relaxation processes are studied employing the pump-probe principle. A well defined time prior to the arrival of the X-ray probe pulse a laser pulse focused on the probed area excites the sample. Recording X-ray absorption at a particular X-ray photon energy for a sequence of different time delays between pump and probe pulse images the progression of laser induced dynamics. The relative time delay is controlled by changing the geometric path length of the laser pulse (1 µm ≙ 3.3 fs).

The effective repetition rate of the experiment is determined by that of the laser. The laser is electronically synchronized to the synchrotron within less than the hybrid electron bunch width. Intrinsic synchronization of the fs laser pump and the fs X-ray probe pulses is obtained through the slicing process. An efficient suppression of remaining X-ray intensity from non-sliced electron bunches requires time gating of the photodiode (lower part of the figure).



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The significant changes within the design and modifications of machine, undulator, beamline and detector have been facilitated by a joint effort of many more BESSY staff members.

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