Ultrafast time and spin resolved photoemission experiment at FLASH

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1 Ultrafast demagnetization observed by spin resolved photoemission spectroscopy

Magnetism and its dynamical properties are at the basis of our information society. On long time scales and large length scales ferromagnetic cores of transformers prove essential for the electric power grid. On the nanosecond and nanometer scale magnetic domains are used to store information on hard disk drives. One of the forefront areas in modern magnetism is the use of ultrashort infrared or optical photon pulses to manipulate the magnetization. It has been observed that the magnetization of a ferromagnet seams to drop within less than a picosecond after excitation by a femtosecond laser pulse. This phenomenon is surprising as demagnetization requires the transfer of angular momentum from the spin system to the lattice of the solid.

The most direct way of measuring demagnetization processes is to detect the spin polarization of photoelectrons. All other optical methods like magneto-optical Kerr as well as circular dichroism measurements depend on the delicate band structure of the valence band, which is altered by the infrared pump beam. In contrast spin polarized photoelectron spectroscopy offers direct insight into the true magnetization of the ferromagnet. In order to obtain a reliable measure of the magnetization, which is a Brillouin zone average over all filled spin polarized band states, one needs to employ high-energy probe photons in the form of the soft x-rays available at a free electron laser. As spin is conserved in elastic photoemission with linearly polarized radiation, the spin polarization of the elastic valence electrons provide an accurate measure of the magnetization, or more precisely, a measurement of the spin part of the magnetization.

In our experiment a ferromagnetic sample is exposed to a 800 nm pump laser beam. The pump pulse of 100 fs duration excites a 10 nm Fe film serving as the magnetic sample. After a variable time delay the probe pulse from FLASH excites photoelectrons from the sample (see figure 1). As the FEL probe pulse has a photon energy of 180 eV it excites electrons from the valence band and core electrons. As the high energy electrons scatter with valence electrons on the way to the surface a cascade of lower energy electrons is created. This cascade carries an average spin polarization of the valence band. In order to measure the spin polarization of the photoelectrons a Mott spin polarimeter is used.

In 2011 we had two beam times at FLASH. During our first beamtime in March 2011 we successfully demonstrated the feasibility of time and spin resolved photoemission experiments using FEL radiation. This experiment had the goal of confirming the existence of ultrafast demagnetization processes in iron. The FEL induced photoelectrons were collected by an electrostatic lens and transferred into the Mott spin polarimeter. As this experiment did not contain an energy filter the detected electrons were mostly part of the cascade. Figure 2 shows the time dependence of the spin

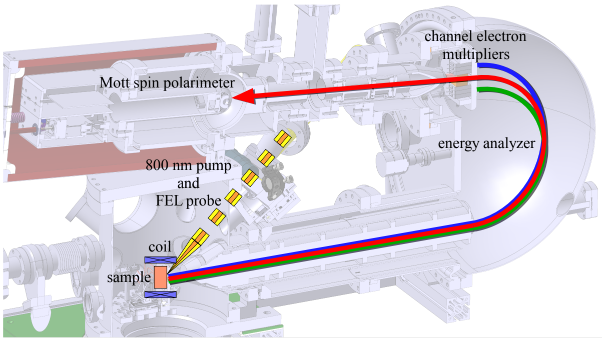

Figure 1: Principle of the experiment: The ferromagnetic sample is exposed to the laser pump beam followed by the FEL pulse. The pump- and probe beams are collinear. The FEL pulse photo-excites electrons which are energy analyzed and accelerated into the Mott spin polarimeter. The spin polarization is measured along the magnetization direction of the sample.

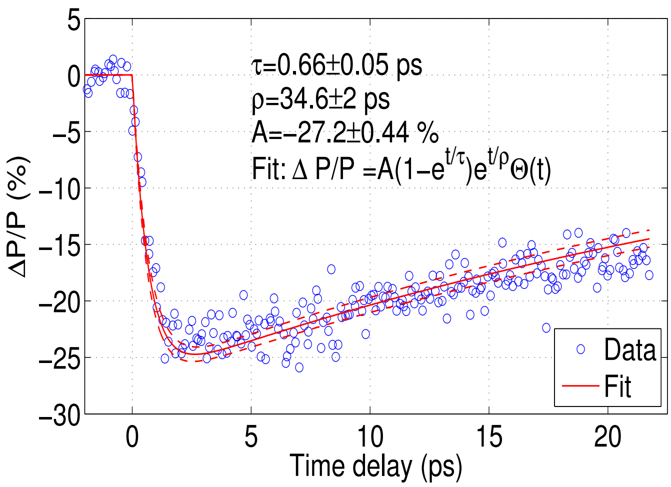

Figure 2: Results from our first beamtime at FLASH in March 2011. The spin polarization drops as a function of the pump probe delay time within less than a picosecond. As more data points were taken on the transition, the noise level is smaller around zero delay time than for longer delay times. The time resolution is limited by the timing jitter between the FEL and the 800 nm laser, which will be improved.

polarization after demagnetization by the 800 nm laser pulse. The spin polarization drops within less than a picosecond and slowly recovers on the 100ps time scale. This clearly demonstrates the feasibility of time and spin resolved photoemission experiments and shows that the magnetization indeed drops within less than a picosecond after excitation. Here we are not just looking at the spin polarization of electrons close to the Fermi energy, but at the cascade electrons which represent an average of the whole valence band.

For our second beam time in July 2011 we added a hemispherical electron energy analyzer to the experimental setup. This way we can select the energy of the photoelectrons before detection in the Mott spin polarimeter. For future experiments we intend to detect the demagnetization effect for different parts of the valence band. During the second beam time the timing diagnostic system at FLASH was improved so we achieved a higher temporal resolution than during the first beam time.