Ultrafast and element-selective demagnetization dynamics

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We employ higher harmonics generated by a table-top femtosecond laser amplifier to detect element-selectively the ultrafast demagnetization in Permalloy at the M absorption edge of Fe and Ni in the soft X-ray regime. The magnetic signal at both edges is reduced simultaniously by about 60% within the first 300 fs after pump pulse excitation. This behaviour points towards a strong exchange coupling between Fe and Ni.

The study of magnetism, magnetic materials, and dynamics in magnetic systems is not only a topic of fundamental interest in our understanding of correlated systems, but also is it directly relevant to technology and information storage. In recent years, magnetism at the ultrafast timescale has been a topic of increasing interest. A thorough understanding of femtosecond magnetism will address the important question of how fast the magnetization can be modified in a material and what physical processes present fundamental limits to this speed. This consideration has motivated a variety of studies using magneto-optical effects with ultrafast light pulses.

Most magneto-optical studies of magnetization dynamics currently make use of either visible wavelength range from ultrafast lasers, or X-rays from large-scale synchrotron radiation facilities. Ultrafast lasers produce short pulses (around 50 fs), thereby attaining a time resolution in the femtosecond range [1, 2, 3], but with a spatial resolution that is generally limited by the wavelength of the probe light. X-rays, on the other hand, allow for a high spatial resolution, high contrast and element specificity by probing ferromagnetic materials at their absorption edges. However, the available time resolution to-date has been too slow (at the order of ps) to resolve the fastest domain reorientation dynamics. Because of this, significant efforts have been devoted to using laser pulses to select short bursts (less than 100 fs) of X-rays from a synchrotron light source (so-called femtosecond slicing) [4]. However, these experiments are time-consuming and challenging, due to the low flux of the sliced photons and the complexity of the technique.

In our work [5], we show for the first time that XUV light generated using high-harmonic generation [6, 7] can be used to observe magnetization dynamics of magnetic materials, exploiting the transverse magneto-optic Kerr effect. In this so-called T-MOKE geometry (Fig. 2b), the reflectivity of the material changes depending on the magnetization of the sample. In our experiment, we periodically reverse the magnetization direction of a grating structure made of Permalloy ($Ni_{80}Fe_{20}$), using an externally applied magnetic field from a Helmholtz coil (Fig. 2a). The strength of the magnetic signal is commonly quantified by the asymmetry, which is defined as $\frac{I_+ - I_-}{I_+ + I_-}$, where I_+ and I_- denote the reflected intensities recorded for two opposite magnetization directions. It can be shown that the asymmetry as a first approximation is linearly proportional to the magnitude of the magnetization [8].

A typical HHG spectrum is displayed in Fig. 1. It shows discrete harmonics diffracted off the Permalloy grating which are recorded by a X-ray CCD camera. The strong absorption at 72 eV is due to Al filters used to block the laser light co-propagating with the harmonics. We can map the magnetic asymmetry of the sample over the entire region around the Ni (67 eV) and Fe (54 eV) M absorption edge. We measured asymmetries of up to 4 % and 6 % at the Fe and Ni edge, respectively, depending on the angle of incidence on the sample. Due to the high photon flux and the high magnetic contrast in this spectral region, a good signal-to-noise ratio (more than 30) could be obtained in an acquisition time of only 100 seconds.

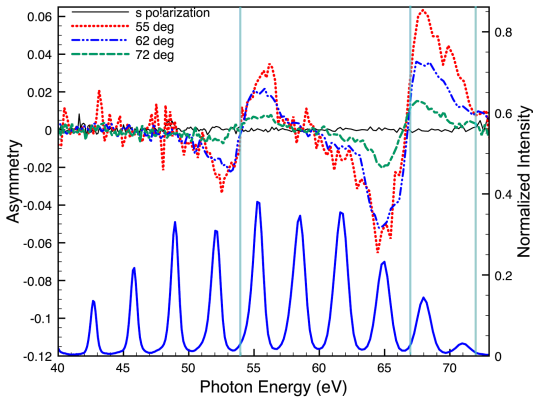

FIG. 1: Experimental data showing the HHG spectrum (bottom, right axis) and measured magnetic asymmetry (bottom, left axis). No asymmetry is measured with s-polarized light. The vertical lines denote Fe, Ni and Al M absorption edges.

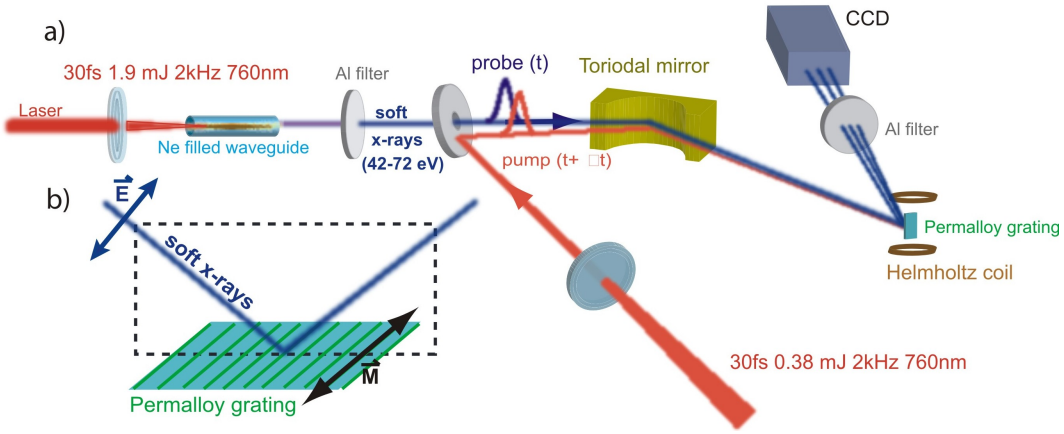

FIG. 2: a) Experimental setup: Intense light from an ultrafast laser amplifier is split by a beamsplitter (not shown). Most of the beam (about 80 %) is used to generate XUV higher harmonics, which are focused with a toriodal mirror onto a Permalloy grating. The diffracted light is recorded by a X-ray sensitive CCD. The rest of the beam is used as a pump to excite the sample. b) T-MOKE geometry: the large blue arrow depicts the polarization of the incident soft X-ray light (p-polarized light) whose plane of incidence is shown by the dashed black box. Black arrows indicate two directions of magnetization rotation, which are perpendicular to the plane of incidence of the probe light.

We demonstrate the use of a high harmonic beam from a table-top setup as an element-specific probe of ultrafast demagnetization processes after laser pulse excitation which demagnetizes the sample. Initially, the ultrafast intense pump pulse coherently interacts with the electron and spin system within the duration of the laser pulse [9]. Then, thermalization of the electron and spin system take place by means of incoherent scattering processes, resulting in a subsequent reduction/re-orientation of the magnetization vectors [1, 2, 3]. The data shown in Fig. 3 was obtained with a pump fluence of about $1\ \frac{mJ}{cm^2}$. We used harmonics around the Ni and Fe M absorption edge to measure the element-selective asymmetry as a function of the delay between pump and probe pulse. Fig. 3 shows clearly the simultanious reduction of the magnetic asymmetry for Ni and Fe within 300 fs after heating the sample by the pump beam. This behaviour points towards a tight exchange coupling of both elements and is in agreement with past measurements in the visible regime [3].

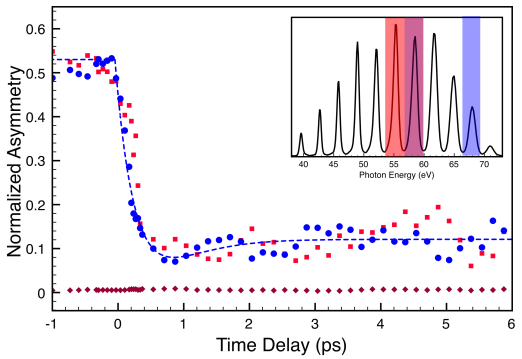

FIG. 3: Time-resolved measurement of the femtosecond laser-pulse induced demagnetization of the Permalloy film by high-harmonic photons around the Ni (blue circle), Fe (red square) M edge, and off-resonance (diamond). The dashed line corresponds to a fit of the expected response [3].

In summary, we exploit the T-MOKE to measure the magnetization dynamics of Permalloy at the M absorption edges of Fe and Ni using HHG light from a table-top laser. By using photon energies near absorption edges of the materials, element specific information about magnetic materials can be obtained with a high signal-to-noise ratio within short acquisition times. This experiment demonstrates the feasibility to investigate coherent magnetization dynamics [9] in the low femtosecond-to-attosecond regime using high-harmonic light with element specificity, and with the potential of a spatial resolution below 100 nm if coherent diffractive imaging techniques are employed [10].

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