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ULTRAFAST MAGNETISATION DYNAMICS AT THE LOW-FLUENCE LIMIT SUPPORTED BY EXTERNAL MAGNETIC FIELDS

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Abstract

We report on ultrafast magnetisation dynamics in ferromagnetic cobalt/platinum multilayers upon pumping by near and mid to far infrared radiation, utilizing sub-100 femtosecond free-electron laser pulses. The evolution of the excited magnetic state is studied on femtosecond timescales with nanometre spatial resolution and element selectivity, employing time-resolved magnetic small-angle X-ray scattering. The obtained results contribute to the ongoing discussion to what extent either coupling of the electromagnetic field or rather quasi-instantaneous heating of the electron-system is the driving force for phenomena like ultrafast demagnetization or all-optical helicity-dependent switching.

INTRODUCTION

Ultrafast demagnetisation on sub-100 femtosecond to picosecond time scales [1] and the related phenomenon of all-optical helicity-dependent switching (AO-HDS) of magnetisation [2] are fascinating effects in fundamental physics with potential for, e.g., energy-efficient future data-storage devices [3,4]. First discovered in amorphous ferrimagnets like GdFeCo [2,5], AO-HDS has also been found recently in various ferromagnets including Co/Pt multilayers [6]. In the latter case, the impact of the laser pulse is controversially debated. Reports on deterministic single-pulse switching in micron-sized magnetic domains [7] contrast with the stochastic nature of a multi-pulse switching mechanism found in different experiments, see, e.g., Ref. [8]. Modelling the deterministic process by means of the magnetic two-temperature model resulted in thresholds for the pump fluence and the duration of the inverse Faraday effect (IFE)-induced magnetic field, set by the length of the pump pulse [9,10]. In that interpretation, the impact of the laser pulse is assumed to be twofold. First, quasi-instantaneous heating of the electron-system above the Curie temperature mediates a transient paramagnetic state (demagnetisation). Second, optomagnetic coupling of the laser-induced effective magnetic field ($\mathbf{H}_{\mathrm{IFE}}$) to the spin system during subsequent cooling down to the paramagnetic phase can result in nucleation and consecutive growth of reversed magnetic domains, depending on the helicity of the laser light. As the spatial resolution in commonly used Kerr microscopy experiments is limited to the micron range, information on changes in nanoscopic domain systems during demagnetisation is scarce. In this respect, free-electron lasers (FELs), like FERMI and FLASH, providing sub-100 femtosecond extreme ultraviolet (XUV) pulses, allow to explore femtosecond dynamics also in nanometre-sized magnetic domains [11-15]. Tuning the photon energy to resonance with one of the dichroic transitions in the magnetic element, here the $M_{2,3}$-absorption edge of Co at 59.6 eV, adds element selectivity to studies of magnetisation dynamics [16].

This paper reports on a near-infrared (NIR)-pump–FEL-probe experiment (pump wavelength of 800 nm), in which the pump helicity-dependent response of a magnetic multi-domain state was studied in the presence of an external magnetic field, $\mathbf{H}_{\mathrm{ext}}$, following observations in Ref. [17]. The evolution of the domain system was monitored by time resolved resonant magnetic small-angle X-ray scattering (tr-mSAXS). Complementarily, we show the feasibility to study the laser beam's $\mathbf{H}$-field influence on demagnetisation by using polychromatic mid to far-IR pump radiation (wavelength of 30– 150 μm) for simplicity denoted in the following as THz-radiation (frequency of 2– 10 THz). Since the oscillation periods of the electromagnetic field are slower than for NIR radiation, a direct field-induced response of the magnetic system can be expected [18-20].

EXPERIMENTAL

Cobalt/platinum multilayers with perpendicular magnetic anisotropy were deposited by sputtering techniques on freestanding 50 nm-thin Si_3N_4 membranes [21]. The experiment using NIR-pump wavelength was conducted in tr-mSAXS geometry (Fig. 1) at the DiProI beamline of FERMI at ELETTRA [22]. A multilayer with 6 bilayer repetitions of Co/Pt was used, which is fully remanent with a coercive field of $\mu_0 H_{\mathrm{c}} \approx 15$ mT. The global energy minimum, obtained after out-of-plane demagnetization, is a multi-domain state with a characteristic periodicity of 460 nm. A perpendicular magnetic field increases the characteristic length scale as predominantly small domains, oriented oppositely to the field, are erased. The characteristic periodicity at a field of 15 mT is 532 nm.

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This rather strong susceptibility enhances the probability to induce features of AO-HDS which might result, e.g., in a second order scattering ring [23]. The *in situ* Helmholtz coil was operated in pulse mode, shaping the driving voltage such that the target field strength was reached well within 200 µs, kept constant for 250 µs, before it was switched off and decreased with a time constant of ≈ 1 ms. The field pulses were triggered with a time delay of −250 µs at the operation frequency of the FEL (50 Hz), ensuring a constant magnetic field at the time of the pump-probe events. A continuous pulse operation of the coils without detectable heating effects was feasible due to the low duty cycle of ≈ 3%. Circularly left (CL) or right (CR) polarized pump pulses with a fluence of $3.4\ \mathrm{mJ/cm^2}$ and a pulse duration of 3.5 ps were utilized, following recent predictions in Refs. [9, 10].

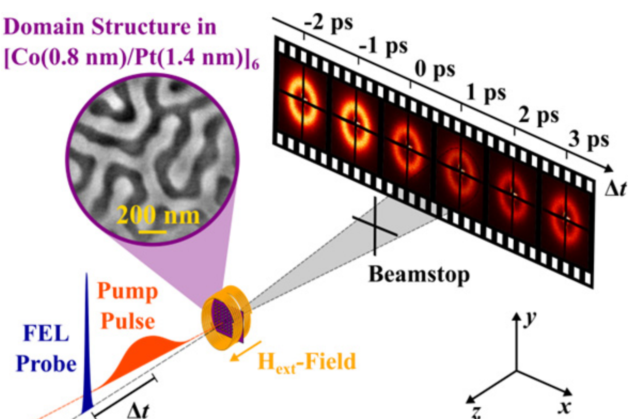

Figure 1: Schematics of the tr-mSAXS experiments. The magnetic multi-domain state was pumped by either circularly polarized NIR or linearly polarized THz pulses. The magnetic response was monitored by a CCD camera detecting scattering of delayed FEL-probe pulses resonantly tuned to the $M_{2,3}$ edge of Co. Magnetic fields were applied normal to the sample surface in the former case.

Complementarily, we used THz radiation to excite the magnetic multi-domain state. Our custom-made endstation allowing for THz-pump–FEL-probe experiments using the tr-mSAXS technique was installed at FLASH's BL3 beamline. A Co/Pt sample with 8 bilayer repetitions and a characteristic periodicity of 440 nm was pumped by polychromatic 10-cycle THz pulses containing wavelengths between 30 µm and 150 µm, produced by the additional electromagnetic undulator available at FLASH [24]. The pulses consisted of the fundamental frequency and, predominantly, the third and fifth harmonics. Higher frequency components of the spectrum (wavelengths shorter than 30 µm) were blocked using a low-pass filter. The actual THz trace as well as the weight of the frequency components could not be measured at the time of the experiment. The pump and probe pulses were geometrically separated and guided along two different beam paths to the endstation where they were focussed quasi-collinearly to the sample.

In both experiments the response of magnetisation was probed by sub-100 femtosecond FEL pulses resonantly tuned to the Co $M_{2,3}$-absorption edge (photon energy of 59.6 eV ≙ wavelength of 20.8 nm). The scattered FEL radiation was detected by a CCD camera with 2048 × 2048 pixels and a pixel size of 13.5 µm.

RESULTS AND DISCUSSION

Results from tr-mSAXS at FERMI's DiProI beamline are shown in Fig. 2. Scattering from a disordered magnetic maze-domain structure results in an isotropic scattering ring (Fig. 2a)) that is azimuthally averaged and fitted with a split Pearson type VII function (Fig. 2b)). Higher order scattering rings are not observed revealing that there is no significant disparity in up and down magnetised domains. The maximum of the scattered intensity $I(Q_\mathrm{c})$ is a measure for the out-of-plane component of the magnetisation $M_z \propto \sqrt{I(Q_\mathrm{c})}$ with Q_c representing the characteristic scattering length. The inset of Fig. 2c) shows the temporal evolution of magnetisation $M_z(\Delta t)$, normalized to the magnetisation $M_{z,0}$ in the unpumped case for CR and CL polarized pump-pulses and external fields of 0 mT and 15 mT. We observe a quenching of magnetisation to 55% during the pulse duration of 3.5 ps. Importantly, a dependence of the strength of demagnetisation on the external field or the pump pulse's helicity is not observed. This is understandable as the disparity in up and down magnetised domains is found to be negligibly small, so that differences in absorption for CR and CL polarized pump pulses average out. Note, that in a homogeneously magnetised Co/Pt sample a small polarization dependent difference in the strength of demagnetisation was assigned to the magnetic circular dichroism effect [17].

The time evolution of the shift in Q_c, $\Delta Q_\mathrm{c}(\Delta t)$, normalized to the unpumped $Q_{\mathrm{c},0}$ for both CR and CL polarized pump pulses and different external fields is shown in Fig. 2c) and d), respectively. In the absence of an external bias field $\Delta Q_\mathrm{c}(\Delta t)$ remains constant upon NIR-pumping. This behaviour is in line with the results of Ref. [12] where a fluence dependent onset of magneto-structural changes was found for Co/Pt multilayers. However, when applying external bias fields of 5 mT and 15 mT, ΔQ_c temporarily reduces within the pump-pulse duration of 3.5 ps and almost fully recovers within ≈ 100 ps, which is faster than the remagnetisation (inset in Fig. 2c)). For a bias field of $\mu_0 H_\mathrm{ext} = 15$ mT a maximum reduction of ΔQ_c by $(2 \pm 0.5)\%$ is found for CR polarization (Fig. 2c)), while for CL polarization the effect is about a factor of 2 stronger, $\Delta Q_\mathrm{c} = (5.5 \pm 0.5)\%$ (Fig. 2d)). As speckles are observed (Fig. 2a) even in images averaged over 100 FEL pulses, a significant rearrangement of magnetic domains can be excluded. The observed shift has rather to be interpreted in line with Ref. [12], i.e., as a consequence of superdiffusive spin currents.

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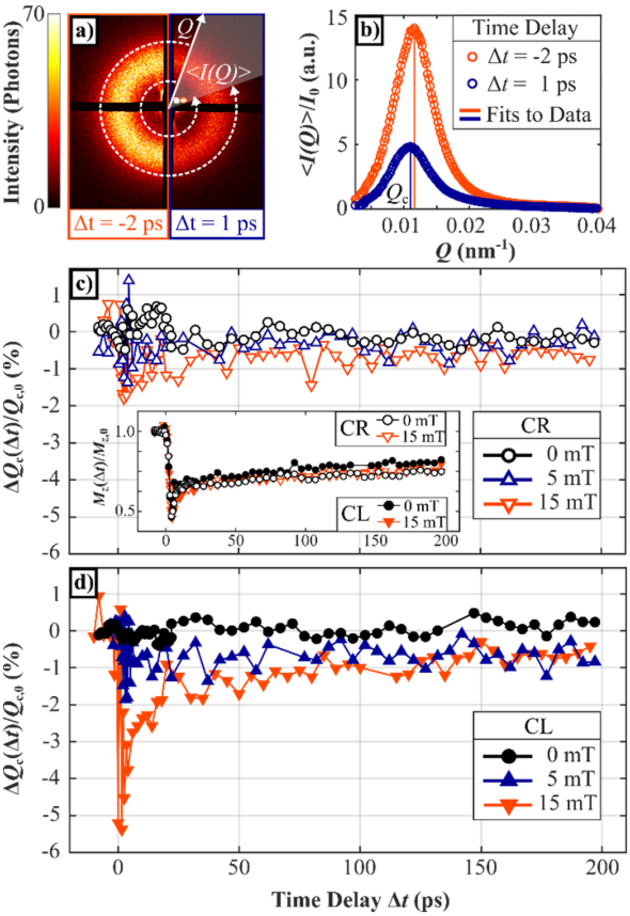

Figure 2: Ultrafast magnetization dynamics using NIR radiation and external magnetic fields. a) Examples of mSAXS images for the unpumped ($\Delta t = -2$ ps) and pumped ($\Delta t = 1$ ps) case (sum of 100 FEL pulses each). b) Respective azimuthally averaged intensities, normalized to the incoming FEL-intensity. The characteristic scattering vector Q_c and the maximum scattering intensity $I(Q_c)$ are obtained from fitting with a split Pearson type VII function. c),d) Time evolution of the relative change of the characteristic scattering length $\Delta Q_c(\Delta t) = Q_c(\Delta t) - Q_{c,0}$ for zero and external fields upon pumping by c) CR and d) CL polarized pump-pulses. The inset in c) shows the corresponding demagnetization for both polarizations and external fields of 0 mT and 15 mT. Lines are guides to the eye.

In that picture, the observed helicity dependence is interpreted to originate from the interplay of the polarization dependent induced magnetic field $\mathbf{H}_{\text{IFE}}$ with the external bias field $\mathbf{H}_{\text{ext}}$. The effect of superdiffusive spin transport on the domain-wall is either enhanced or suppressed. A spin-up-polarized current preferentially gets scattered in down-magnetised magnetic domains (and vice versa) in the vicinity of a domain wall, resulting in its transient broadening and the observed shift of Q_c. A quantification of the process using the results of Ref. [25] for the strength of IFE in ferromagnets and heavy paramagnets is planned.

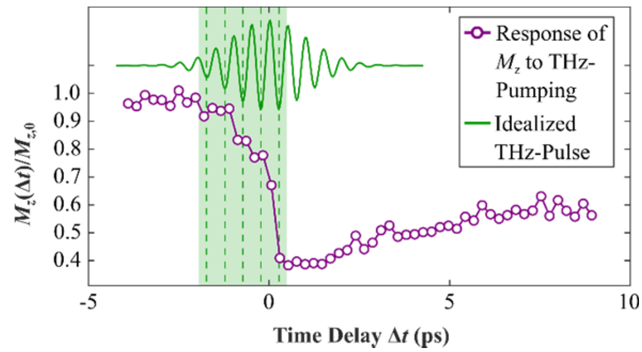

Figure 3: Ultrafast demagnetization using THz radiation (purple). The line is a guide to the eye. The unknown pulse shape is sketched as an ideal 10 cycle 2 THz pulse with Gaussian envelope (green).

Since for Co $\mathbf{H}_{\text{IFE}}$ points along the same direction for CR and CL polarized light, the optomagnetic behaviour of Pt might be the driving force as it shows a sign change of $\mathbf{H}_{\text{IFE}}$ for CR and CL polarized light [25].

For investigating the direct response of magnetisation to the pump pulse's **H**-field, we utilized polychromatic THz radiation, as described above, to pump the magnetic multi-domain state in Co/Pt. Prominently, we observe a strong quenching of the magnetisation to ≈ 40%, using a fluence of $\approx 10\ \text{mJ/cm}^2$ corresponding to a magnetic field component of ≈ 290 mT (Fig. 3). The falling edge of the demagnetisation curve is not smooth but rather shows a step-like behaviour. We interpret the overall decrease of magnetisation as a result of quasi-instantaneous Joule heating, in line with Ref. [18]. In contrast, the plateaus can be interpreted as the coherent, i.e., directly field-induced, part of the sample's response to the THz magnetic field. It is noted that the demagnetisation effect is large when compared to results in other studies [18-20]. We speculate that the spectral properties, especially the high-frequency part of the pump spectrum, are responsible for this difference.

In conclusion, we studied the influence of optomagnetic fields on demagnetisation dynamics. We found a reduction of the characteristic scattering length already for low-intensity NIR pump-pulses in presence of out-of-plane magnetic fields. The effect evolves on the timescale of the pulse duration and shows strong helicity dependence. A possible explanation lies in a helicity dependent strength of $\mathbf{H}_{\text{IFE}}$ in Co and/or sign of $\mathbf{H}_{\text{IFE}}$ in Pt. The magnetic response to pumping with THz radiation hints at coherent coupling of the THz magnetic field to the spin system. Recent and planned upgrades of the FLASH facility [26] and the THz beamline [24] will provide strongly improved conditions for further, more quantitative studies of THz-induced magnetisation dynamics.

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