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Ultrafast element-specific magnetization dynamics of complex magnetic materials on a table-top

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ABSTRACT

We review recent progress in femtosecond magnetization dynamics probed by extreme ultraviolet pulses from high-harmonic generation. In a transverse magneto-optical Kerr geometry, we established an ultrafast, element-specific experimental capability – on a table-top – for the measurement of magnetization dynamics in complex multi-sublattice magnets and multilayer magnetic structures. We show that this newly introduced technique is an artifact-free magnetic sensor, with only negligible non-magnetic (optical) contributions from the transient variation of the refractive index due to the presence of a non equilibrium hot-electron distribution. We then use these new experimental capabilities of ultrahigh time-resolution, combined with element-specific simultaneous probing, to disentangle important microscopic processes that drive magnetization dynamics on femtosecond timescales. We elucidate the role of exchange interaction on magnetization dynamics in strongly exchange-coupled alloys, and the role of photo-induced superdiffusive spin currents in magnetic multilayer stacks.

1. Introduction

Femtomagnetism, which is the manipulation of magnetic order on femtosecond timescales by ultrashort laser pulses, was first observed by Eric Beaurepaire et al. in 1996 [1]. Since then, femtomagnetism has become a challenging research topic of increasing interest because of its importance for uncovering fundamental new science and for technological applications. Typically, experiments that study femtomagnetism are carried out in a pump-probe geometry. An intense femtosecond laser pulse first excites a magnetic system, and the resulting ultrafast changes of the magnetization are then probed magneto-optically and by spin-resolved photoemission. The dynamical response of the magnetic material to the excitation by an ultrashort laser pulse is governed by non equilibrium interactions between photons, electrons, spins, and phonons (see Fig. 1). Despite nearly two decades of research, the fundamental microscopic processes involved in femtomagnetism are not well understood, and indeed are still a topic of intense debate [2–13]. One of the key challenges is to carefully disentangle the various dynamical processes shown in Fig. 1 to establish how they contribute to the behavior of a complex magnetic system far from equilibrium. This quest demands the development of new experimental capabilities. For example, the investigation of coherent magnetization dynamics [7,14] in the time domain requires extremely high time-resolution (<20 fs), while the influence of exchange-coupling on magnetization dynamics [15] requires element-specificity. Finally, to capture superdiffusive spin-transport in magnetic multilayer stacks [16], we require ultrafast element-specific, layer-selective probes of the magnetization state [16–18].

The ideal experimental technique would therefore combine sensitivity to the magnetization state with femtosecond-to-attosecond time-resolution, be able to distinguish the signal from different elements in an alloy or multilayer system, and be able to image with nanometer spatial resolution. This is a challenging task, but one that can be achieved by use of femtosecond slicing technique [17,18], the newly developed large-scale X-ray free-electron lasers (XFELs) [19,20], and table-top-scale high-harmonic generation (HHG) light sources [15,16,21–23]. Comparing magnetic dynamics probed by HHG and femtosecond slicing sources or XFELs, it is becoming clear that all of these techniques are complementary and have unique advantages. High harmonic probes at the M absorption edges have the advantages of probing the magnetic state of multiple

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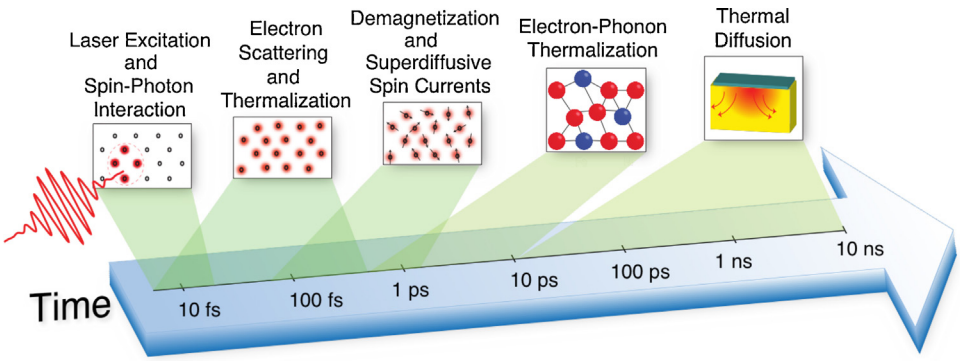

Fig. 1. Adapted from [15]. Schematic timeline of ultrafast photon–electron–spin–lattice interactions after an ultrafast laser excitation. During the ultrafast excitation of the electron system by a femtosecond laser pulse, ultrafast spin-photon interaction can be a source of magnetization dynamics [6,7,14,29]. On a longer femtosecond timescale, various scattering processes between electrons, phonons, and magnons, as well as superdiffusive spin-currents [8,16,30,31] determine the dynamic response of the material. The strongly excited electron system thermalizes by predominantly electron-electron scattering to a Fermi-Dirac distribution. Spin-flip electron–electron [5,11], electron–phonon [3,4,9–11,32], and electron–magnon [10,33] scattering processes, together with superdiffusive spin currents [8,16,30,31] mediate the magnetization dynamics. Electron-phonon scattering transfers the energy from the excited electron gas to the lattice, and thermal equilibrium is typically reached on picosecond timescales. Finally, on nanosecond timescales, the material cools by thermal diffusion. The different contributions of the above-mentioned processes to the ultrafast magnetic dynamics are widely debated.

elements simultaneously, allowing the fastest coupled dynamics to be uncovered with very high precision. This source is also compact and accessible. Synchrotrons and XFELs can probe dynamics at the higher-energy L-shell absorption edges, allowing spin and orbital contributions to be extracted, and enabling higher spatial resolution imaging.

In this paper, we review how HHG light sources can uncover the fastest dynamics in femtomagnetism [15,16,21,22]. HHG is an extremely nonlinear process that produces coherent short-wavelength beams with the shortest pulse durations in the few femtosecond to attosecond regime demonstrated to date for any light source [24–28]. Bright harmonic beams now span from 10 eV to greater than 2 keV [28] while retaining the polarization and coherence properties of the driving laser under phase-matched generation conditions. We show that table-top HHG sources are ideal probes of femtomagnetism because of their artifact-free sensitivity to the magnetization [22], femtosecond time resolution, and element-specificity at multiple sites simultaneously [21]. These unique new experimental capabilities make it possible to solve long-standing problems in femtomagnetism [16], and also enable more complex and technologically relevant magnetic materials to be studied [15]. HHG has already been used to capture the fastest magnetization dynamics in elemental materials, complex magnetic alloys, and multilayer systems, thereby elucidating the role of exchange interaction and superdiffusive spin currents in ultrafast magnetization dynamics.

2. The transverse magneto-optical Kerr effect in the XUV regime

Femtosecond extreme ultraviolet (XUV) pulses from HHG are produced by focusing 90% of an amplified, femtosecond laser pulse (780 nm wavelength, 2–3 kHz repetition rate, 1.5–2.5 mJ per pulse) into a capillary-waveguide filled with Ne (Fig. 2). For high-efficient phase-matched high-harmonic up-conversion, we optimize the gas pressure in the waveguide at values around 800 Torr [25,27]. A broad range of harmonics spanning photon energies from 35 eV to 72 eV are emitted simultaneously. The cutoff at 72 eV is due to the absorption edge of the Al filters that are used to block the laser light that co-propagates with the HHG beam. The duration of the high-harmonic XUV pulses is less than 10 fs [34].

We use a transversal magneto-optical Kerr (T-MOKE) geometry to probe the magnetic state of our samples (Fig. 2, inset), where, for a single magnetic layer, the polarization-dependent reflected intensity of the XUV beam for p-polarized incident light is [35,36]:

$$I_{\pm}^{\mathrm{P}} = I_0 \left| \frac{n\cos\theta_i - \cos\theta_t}{n\cos\theta_i + \cos\theta_t} \pm \frac{2\,\sin\theta_i \cos\theta_i}{n^2(n\cos\theta_i + \cos\theta_t)^2} \in_{xy} \right|^2 \tag{1}$$

and for s-polarized incident light is

$$I^{\mathrm{s}} = I_0 \left| \frac{\cos\theta_i - n\cos\theta_t}{\cos\theta_i + n\cos\theta_t} \right|^2 \tag{2}$$

where I_0 is the incident beam intensity, n the refractive index of the material, θ_i the angle of incidence, $\theta_t = \sin^{-1}(\sin\theta_i/n)$ the refractive angle, the plus-minus sign depends on the direction of the in-plane magnetization, and $\in_{xy}$ the magnetization-dependent off-diagonal element of the dielectric tensor. For p-polarized light, the first term inside the absolute square of Eq. (1) is the non-magnetic Fresnel coefficient for the sample reflectivity. The dependence of the reflectivity on the magnetic state is contained in the second term

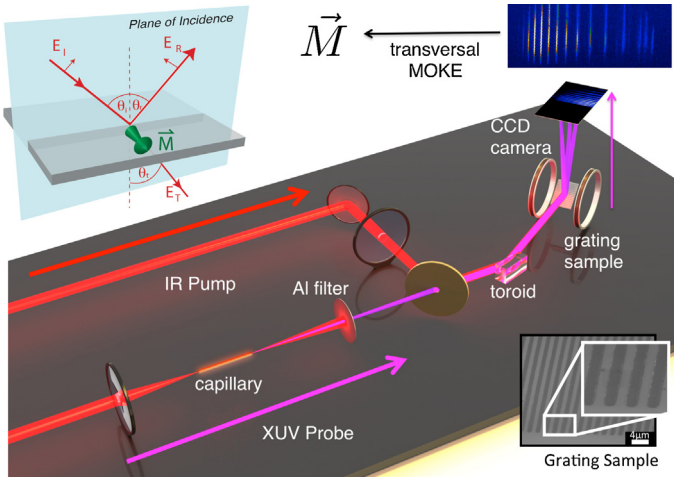

Fig. 2. Adapted from [15]. Schematic of the experiment. Ultrafast XUV pulses are reflected from a magnetic sample with a grating on top, which spatially separates the harmonics to form a spectrum on a CCD camera. The reflected HHG intensity at the shallow-core absorption edges depends on the magnetization transverse to the optical plane of incidence (T-MOKE, see inset) that is periodically reversed by transverse-mounted Helmholtz coils. Exciting the sample with an ultrashort laser pulse (red) initiates the magnetization dynamics. (For interpretation of the references to color in the artwork, the reader is referred to the web version of the article.)