From magneto-optics to ultrafast manipulation of magnetism

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The purpose of these lectures is to summarize the recent progress in laser-induced magnetization dynamics. Circularly polarized pulses are shown to act as strong transient magnetic field pulses originating from the non-absorptive inverse Faraday effect. An all-optical scheme of excitation and detection of different antiferromagnetic resonance modes with frequencies of several hundreds GHz is demonstrated, along with non-thermal excitation of phase transitions. On the other hand, in ferrimagnetic metals the magnetization could be reversed in a reproducible manner by single 40 fs circularly polarized laser pulses, without any applied magnetic field. The angular momentum of the magnetic system is shown to play a crucial role in this process. All these findings open new insights into the understanding of ultrafast magnetic excitation and, considering recent progress in the development of compact ultrafast lasers, may provide new prospects for applications of ultrafast opto-magnetic phenomena in magnetic storage and information processing technology.

Part 1. Magneto-optics and opto-magnetism.

The interaction of light with magnetized media is manifested in various magneto-optical phenomena. An example is the Faraday effect, observed as a rotation of the polarization plane of light transmitted through a magnetic medium:

$$\alpha_F = \frac{2\pi}{\lambda}\frac{\alpha M}{\varepsilon_0}, \qquad (1)$$

where α_F is the specific Faraday rotation, M is the magnetization, λ is the light wavelength, and α is the magneto-optical susceptibility, which is a scalar value in isotropic media. Various devices, such as magneto-optical isolators and modulators, make use of large values of Faraday rotation in transparent magnetic compounds.

Much less known is the inverse Faraday effect, where high intensity laser radiation induces a static magnetic field H(0):

$$\vec{H}(0) = \frac{\varepsilon_0}{\mu_0}\alpha\left[\vec{E}(\omega)\times\vec{E}^*(\omega)\right], \qquad (2)$$

where $E(\omega)$ and $E^*(\omega)$ are the electric field of the light wave and its complex conjugate, respectively [1-2]. It follows from equation (2) that circularly polarized light at frequency ω should induce a magnetization along the wave vector k. Note that symmetry considerations of equation (2) indicate equivalence between photoexcitation by circularly polarized light and action of an external magnetic field. Moreover, right- and left-handed circularly polarized waves should induce magnetizations of opposite sign.

Equations (1) and (2) show that both these phenomena are determined by the same magneto-optical susceptibility α [2]. Therefore, optical control of magnetization is expected to be most efficient in materials with high values of the Faraday rotation per unit magnetization. Another important property of the susceptibility α is that it has no symmetry restrictions and is thus allowed in all media, regardless of their crystallographic and magnetic structures. Moreover, the

inverse Faraday effect does not require absorption, and is based on a Raman-like coherent optical scattering process. This has the important consequence that the effect of light on the magnetization is non-thermal and practically instantaneous because it takes place on a femtosecond time scale.

The first demonstration of femtosecond opto-magnetic excitation was done in dysprosium orthoferrite $DyFeO_3$. Despite its small magnetization this material exhibits a giant Faraday rotation of about 3000^o cm^{-1} due to its strong spin-orbit interaction [3]. Thus non-thermal effects of light on the spontaneous magnetization are expected to be large in this material.

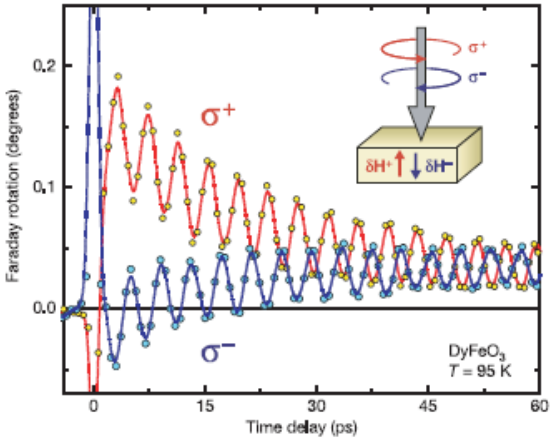

Figure 1 Magnetic excitations in $DyFeO_3$: the circularly polarized pumps of opposite helicities excite oscillations of opposite phase. Inset shows the geometry of the experiment. From reference [4]

Figure 1 shows the temporal evolution of the Faraday rotation for two circularly polarized pump pulses of opposite helicities [4]. At zero time delay instantaneous changes of the Faraday rotation are observed, that result from the excitation of virtual and real transitions in the Fe^{3+} ions from the high spin ground state S=5/2. The instantaneous changes of the Faraday rotation are followed by oscillations with a frequency of about 200 GHz, that can clearly be assigned to oscillations of the magnetization. It is seen from Fig. 1 that the helicity of the pump light controls the sign of the photo-induced magnetization. This observation unambiguously indicates that the coupling between spins and photons in $DyFeO_3$ is direct because the phase of the spin oscillations is given by the sign of angular momentum of the exciting photon.

The subsequent observation of the inverse Faraday effect also in a metallic magnet indicated that this mechanism does not rely on specific material properties but is a general phenomenon. Its strength, as well as the experimental possibility of observing it, depend on the material parameters. On the one hand, it is the value of the magneto-optical response that determines the non-thermal interaction. On the other hand, there are competing thermal effects that are usually dominating in metals. To make a classification, the role of the bandwidth may be invoked: narrowband materials, such as insulators, oxides, and to some extent also f metals can be excited much more selectively than the broadband transition metals. Similarly, the former exhibit more of a helicity dependence than the latter.

Part 2. The role of angular momentum in the ultrafast magnetization dynamics

Many peculiarities of the magnetization dynamics are related to the fact that a certain amount of angular momentum is associated with each magnetic moment. A typical way to reverse the

magnetization is to apply a magnetic field opposite to M. In such configuration the final state is well-defined, however the reversal only occurs in the presence of damping that transfers the corresponding angular momentum to the lattice.

Alternatively, the field can be applied in the orthogonal geometry thus creating a torque that changes the angular momentum directly. This accelerates the reversal though the latter may become non-deterministic, the problem getting worse at short times and strong fields [5].

Here I discuss our recent studies of the questions of angular momentum in ferrimagnetic rare-earth – transition metal alloys, e.g. GdFeCo, where both magnetization and angular momenta are temperature dependent. Depending on their composition, such ferrimagnets can exhibit a magnetization compensation temperature T_M where the magnetizations of the sublattices cancel each other and similarly, an angular momentum compensation temperature T_A where the net angular momentum vanishes. At the latter point, the frequency of the homogeneous spin precession diverges. As a consequence, ultrafast heating of a ferrimagnet across its compensation points results in a subpicosecond magnetization reversal [6].

Moreover, we have experimentally demonstrated that the magnetization can be manipulated and even reversed by a single 40 femtosecond circularly polarized laser pulse, without any applied magnetic field [7-8]. This optically induced ultrafast magnetization reversal is the combined result of laser heating of the magnetic system and circularly polarized light acting as a magnetic field with amplitudes of up to several Teslas. The direction of this opto-magnetic switching is determined only by the helicity, i.e. angular momentum, of light. This novel reversal pathway (see [5]) is shown to crucially depend on the net angular momentum reflecting the balance of the two opposite sublattices.

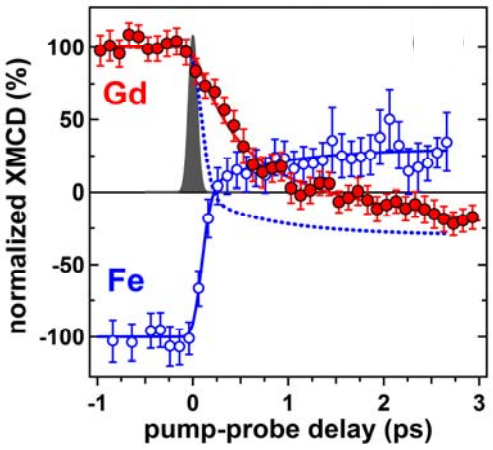

Figure 2 (adapted from [9]) Divergent dynamics of the Fe and Gd magnetic moments measured by time-resolved XMCD with fs time-resolution within the first 3 ps. Experimental time resolution of 100 fs is depicted by the solid Gaussian profile. The solid and dashed lines are guides to the eye. The dashed line depicts the magnetization of Fe-sublattice taken with the opposite sign.

In particular, optical excitation of ferrimagnetic GdFeCo on a time-scale pertinent to the characteristic time of exchange interaction between the rare earth (RE) and transition metal (TM) spins pushes the spin dynamics into a yet unexplored regime, where the two exchange coupled magnetic sublattices demonstrate substantially different dynamics, such as shown in Fig. 2. As a result, the reversal of spins appears to proceed via a novel transient state characterized by a

ferromagnetic alignment of the Gd and Fe magnetic moments, despite their ground-state antiferromagnetic coupling. The net magnetic moment of the RE(Gd) sublattice is found to reverse within 1.5 ps, which is substantially slower than the TM Fe reversal time of 300 fs. These surprising observations supported by atomistic simulations thus present a novel concept of manipulating magnetic order on a timescale of the exchange interaction.

To summarize, optical manipulation of magnetic order by femtosecond laser pulses has developed into an exciting and still expanding research field that keeps being fueled by a continuous stream of new and sometimes counterintuitive results. For a recent review the reader is referred to [10]. Considering the progress in the development of compact ultrafast lasers, optical control of magnetic order may also potentially revolutionize data storage and information processing technologies.

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