Controlling spins with light

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The interaction of sub-picosecond laser pulses with magnetically ordered materials has developed into an extremely exciting research topic in modern magnetism. From the discovery of sub-picosecond demagnetization over a decade ago to the recent demonstration of magnetization reversal by a single 40fs laser pulse, the manipulation of spins by ultrashort laser pulses has become a fundamentally challenging topic with a potentially high impact for future spintronics, data storage and manipulation, and quantum computation. We have recently demonstrated that one can generate ultrashort and very strong (teslas) magnetic field pulses via the so-called inverse Faraday effect. Such optically induced magnetic field pulses provide unprecedented means for the generation, manipulation and coherent control of spins on very short time scales. The basic ideas behind these so-called opto-magnetic effects will be discussed and illustrated with recent results, demonstrating the various possibilities of this new field of femto-magnetism.

Keywords: spintronics; magneto-optics; spin dynamics

1. Introduction

The demands for ever increasing speed of storage of information in magnetic media and the intrinsic limitations connected with the generation of magnetic field pulses by current have triggered intense research to find ways to control spins and magnetization by means other than magnetic fields [1]. Since the demonstration of sub-picosecond demagnetization by a 60fs laser pulse by Beaurepaire et al. [2], manipulating and controlling magnetization with ultrashort laser pulses has become a challenge. This ultrafast demagnetization process was subsequently confirmed by others and explained in terms of a two- or three-temperature model [3–6]. In addition to laser-induced demagnetization, the optical excitation of spin waves has been intensively studied in a large variety of materials [7–12]. However, in all of these laser-induced dynamic effects, the observed magnetic excitation was the result of optical absorption followed by a rapid increase in the electron temperature. This then leads to subsequent demagnetization or to changes in the magnetic anisotropy, resulting in a sudden change in the effective magnetic field that subsequently triggers the magnetization to precess. It would be far more exciting to be able to control spins by light in a non-thermal way. Femtosecond laser pulses offer the intriguing possibility of probing a magnetic system on a time...
scale that is equivalent to the exchange interaction, responsible for the existence of magnetic order, while being much faster than the time scale of spin–orbit interaction (1–10 ps) or magnetic precession (100–1000 ps). Because the latter is considered to set the limiting time scale for magnetization reversal, the option of femtosecond optical excitation immediately leads to the question of whether it would be possible to reverse magnetization faster than half a precessional period. Recently, we have demonstrated that, using circularly polarized 40 fs laser pulses, the magnetization of a 20 nm GdFeCo film could indeed be reversed in a controlled way [13]. The explanation for this surprising result was given in terms of an optically induced effective magnetic field, the responsible mechanism being the inverse Faraday effect [14]. In this paper, we will show how these optically induced fields can indeed be used to control spins in magnetically ordered media and how this can lead to new and unconventional ways to control spins at unprecedented time scales.

2. Interaction between photons and spins

The interaction between photons and spins can be derived using energy considerations [15]. The thermodynamic potential $\Phi$ of an isotropic, non-absorbing, magnetically ordered medium with static magnetization $M(0)$ in a monochromatic light field $\mathbf{E}(\omega)$ includes the term

$$F = \alpha_{ijk} E_i(\omega) E_j(\omega)^* M_k(0), \quad (2.1)$$

where $\alpha_{ijk}$ is the magneto-optic susceptibility [14,16–19]. In the electric dipole approximation, the linear optical response of a medium to a field $\mathbf{E}(\omega)$ is defined by the optical polarization $\mathbf{P}(\omega) = \partial \Phi / \partial \mathbf{E}(\omega)^*$. From equation (2.1), one can easily see that the optical polarization $\mathbf{P}(\omega)$ should have a contribution $\mathbf{P}^{(m)}$ proportional to the magnetization $\mathbf{M}$:

$$P_i^{(m)}(\omega) = \alpha_{ijk} E_j(\omega) M_k(0). \quad (2.2)$$

From this equation, one can find that, when linearly polarized light is transmitted through a magnetized medium, the polarization plane of the light gradually rotates over an angle $\theta_F$ given by

$$\theta_F = \frac{\alpha_{ijk} M_k(0) \cdot \omega L}{cn}, \quad (2.3)$$

where $c$ is the speed of light in vacuum, $n$ the refraction coefficient of the medium and $L$ the propagation distance of the light in the medium [20]. Equation (2.3) describes the so-called magneto-optical Faraday effect discovered by Faraday in 1846 [21].

From equation (2.1), one can find that an electric field of light at frequency $\omega$ will act on the magnetization as an effective magnetic field $\mathbf{H}^{\text{eff}}$ directed along the wavevector of the light $\mathbf{k}$:

$$H_k = -\frac{\partial F}{\partial M_k} = \alpha_{ijk} E_i(\omega) E_j(\omega)^*. \quad (2.4)$$
In isotropic media, $\alpha_{ijk}$ is a fully antisymmetric tensor with a single independent element $\alpha$. Therefore, equation (2.4) can be rewritten as

$$H = \alpha[\mathbf{E}(\omega) \times \mathbf{E}(\omega)^*].$$  \hspace{1cm} (2.5)

From this, it becomes obvious that right- and left-handed circularly polarized waves should act as magnetic fields of opposite sign $[14,16–18]$. Therefore, in addition to the well-known magneto-optical Faraday effect, where the polarization of light is affected by the magnetization $\mathbf{M}$, the same susceptibility $\alpha$ also determines the inverse opto-magnetic phenomenon: circularly polarized light affects the magnetization via the inverse Faraday effect. Note that, in contrast to conventional magnetic resonance, where microwave photons couple to the magnetic moments through the magnetic component of the electromagnetic field, here it is the electric component of the photon field that is responsible for the inverse Faraday effect.

The first demonstration of femtosecond opto-magnetic excitation of spin dynamics was the optical excitation of the antiferromagnetic resonance modes in DyFeO$_3$, in the hundreds of gigahertz frequency range $[22]$. Details of the structure of this antiferromagnet can be found in Wijn $[23]$. Owing to the Dzyaloshinskii–Moriya interaction, the antiferromagnetically coupled Fe spins are slightly canted, giving rise to a spontaneous magnetization $M_S \sim 8$ G. Despite the small magnetization, this material exhibits a giant Faraday rotation of about $3000^\circ$ cm$^{-1}$ owing to its strong spin–orbit interaction $[20]$. Such weak ferromagnets represent an intriguing combination of static properties of a ferromagnet with the dynamics of an antiferromagnet.

To detect the optically induced magnetization, we used the direct magneto-optical Faraday effect, which was possible due to the presence of a weak ferromagnetic moment. Figure 1 shows the temporal evolution of the Faraday rotation in a $z$-cut DyFeO$_3$ sample for two circularly polarized pump pulses of opposite helicities. On the scale of 60 ps, one can clearly distinguish two different processes that start after excitation with a pump pulse. At zero time delay, instantaneous changes of the Faraday rotation are observed, which 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 figure 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 the helicity of the exciting photon.

The amplitude of the oscillations corresponds to a photo-induced change in the magnetization $\Delta M \sim 0.06M_S$, where $M_S$ is the saturation magnetization.

From figure 1, one can distinguish not only oscillations, but also an exponential decay of the equilibrium level on a time scale of about 100 ps. This can be explained by a photo-induced change in the equilibrium orientation of the magnetization and a subsequent decay of this equilibrium orientation to the initial state. For the laser fluence used ($10$ mJ cm$^{-2}$), the increase in sample temperature was less than $10$ K, as follows from the observed temperature dependence of the measured frequencies.

Although, in principle, the effect of optically induced magnetization does not require the absorption of photons, the laser control of the spontaneous magnetization and the excitation of coherent spin oscillations are equivalent to
Figure 1. Magnetic excitations in DyFeO$_3$ probed by the magneto-optical Faraday effect at $T = 95\,\text{K}$. Two processes can be distinguished: (i) instantaneous changes of the Faraday effect due to the photoexcitation of the Fe ions and relaxation back to the high-spin ground state $S = 5/2$; and (ii) oscillations of the Fe spins around their equilibrium direction with an approximately 5 ps period. The circularly polarized pump pulses of opposite helicities excite oscillations of opposite phase. Inset shows the geometry of the experiment. Vectors $\delta H^+$ and $\delta H^-$ represent the effective magnetic fields induced by the right-handed $\sigma^+$ and left-handed $\sigma^-$ circularly polarized pump pulses, respectively [22]. (Online version in colour.)

Figure 2. The amplitude of the spin oscillations as a function of pump fluence for the same sample and conditions as in figure 1 [22].

photoexcitation of magnons and thus requires some energy. Figure 2 shows the amplitude of the photoexcited spin oscillations as a function of the pump intensity. The linearity of this dependence indicates that the photoexcitation of magnons is a two-photon process, which is in perfect agreement with the expression for the inverse Faraday effect (equation (2.5)). Note that extrapolation of the intensity dependence shows that the photo-induced effect on the magnetization would reach
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Figure 3. Coherent control of spins in DyFeO$_3$ with two circularly polarized laser pulses at $T = 95$ K. (a) Precession triggered by the first laser pulse; (b) amplification of spin precession by the second laser pulse that comes after an even number of full periods; and (c) stopping of the spin oscillations by the second pump that comes after an odd number of half periods. (Online version in colour.)

the saturation value of $M_s$ at a pump fluence of about 500 mJ cm$^{-2}$. The effect of such 100 fs laser pulses on the magnetic system would be equivalent to the application of a magnetic field pulse of about 5 T. According to our measurements, the absorption in DyFeO$_3$ in the near-infrared spectral range is of the order of 100–200 cm$^{-1}$. Given this low value of the absorption, a photoexcitation of 500 mJ cm$^{-2}$ is still below the damage threshold of DyFeO$_3$ and thus quite feasible, provided the sample is of high optical quality.

3. Double-pump coherent control of spin precession

The non-thermal effects of light on the magnetization allow for ultrafast coherent control of spin precession. Such control can be achieved by using multiple laser pulses in rapid succession.

In figure 3, the results of such coherent control experiments are shown for DyFeO$_3$. A pump pulse of helicity $\sigma^+$ arriving at $t = 0$ that acts as a strong pulse of magnetic field triggers precession of antiferromagnetic spins like in figure 1. A second pump pulse of helicity $\sigma^-$ arriving after an odd number of half precessional periods rotates the magnetization further away from $H_{\text{eff}}$, causing the subsequent precession to have almost twice the amplitude (figure 3b). If, however, this second pump pulse arrives after an integer number of full periods, the magnetization is rotated back to its original equilibrium orientation and no further precession takes place (figure 3c).
These experiments clearly demonstrate that femtosecond optical pulses can be used to directly and coherently control spin motions. Depending on the phase of the precession when the second pulse arrives, energy is either transferred from the laser pulse to the magnetic system (amplification of the precession) or from the magnetic excitation to the optical pulse (stopping of the precession). In view of the low intrinsic damping in the orthoferrites, and therefore the long lifetime of their magnetic excitations, it is remarkable how ultrashort laser pulses can completely and instantaneously stop the long-period coherent precession of spins. This process of transferring energy back to the optical pulse can also be viewed as coherent laser cooling of magnons.

The complex spin oscillations in orthoferrites triggered by a train of laser pulses have recently been studied theoretically using nonlinear Landau–Lifshitz–Gilbert (LLG) equations. It was demonstrated that such a periodic excitation of spins results in various patterns of spin oscillations, which depend on intensity and periodicity of the laser pulses [24].

4. Inertia-driven switching in antiferromagnets

The dynamics of spins in a ferromagnet is described by the LLG equation that does not contain inertial terms such as acceleration. Therefore, a switching between two minima requires that the system is dragged over the potential barrier. However, if the inertial motion of the magnetic moments, similar to that of a finite-mass object, was possible, one could ‘kick’ the system so that the following motion would take the system over the barrier and into the new equilibrium (figure 4). Such a mechanism would allow us to use pulses much shorter than the precessional periods.

The inertial motion can actually be realized in antiferromagnets such as the one discussed in the previous section [25]. The dynamics of an antiferromagnet with two sublattices $M_1$ and $M_2$ is described in terms of the motion of the antiferromagnetic unit vector $\mathbf{l} = (M_1 - M_2)/|M_1 - M_2|$. In angular variables, the equation of motion can be written as [25]

$$\frac{d^2 \varphi}{dt^2} + 2I \frac{d\varphi}{dt} + \omega_0^2 \frac{dw(\varphi)}{d\varphi} - \frac{\gamma^2 H_D}{\sin \theta_0} H(t) \cos \varphi = 0. \quad (4.1)$$

Here the presence of the acceleration $d^2\varphi/dt^2$ describes the appearance of inertia, similar to that in Newton’s equation of motion for unit mass; the second term presents a viscous force with damping coefficient $I$; terms with $d(w(\varphi))/d\varphi$ and $H_D$ represent the restoring and driving force, respectively; $w(\varphi)$ is a dimensionless function proportional to the magnetic anisotropy energy; and $\omega_0$ is the frequency of the lower antiferromagnetic mode.

Holmium orthoferrite (HoFeO$_3$) appeared to be an excellent candidate for observing such inertial reorientation of spins: between $T_1 = 38$ K and $T_2 = 58$ K, the magnetic system of HoFeO$_3$ is characterized by two thermodynamic potential minima [23]. One persists up to $T_2$ and corresponds to the $\Gamma_{12}$ magnetic symmetry, with the magnetic moment $\mathbf{M}$ along the $x$ crystallographic axis and the antiferromagnetic vector $\mathbf{l}$ in the $zy$-plane (figure 4). The second potential minimum is present above $T_1$ and corresponds to the $\Gamma_{24}$ phase having both $\mathbf{M}$ and $\mathbf{l}$ in the $xz$-plane. The transition between these two minima is a first-order phase

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Figure 4. Spin switching in HoFeO₃. \( \Gamma_{12} \) and \( \Gamma_{24} \) are the two metastable phases present between 38 and 52 K. \( \mathbf{H} \) represents an effective magnetic field pulse that initiates an inertial motion of spins from the \( \Gamma_{12} \) to the \( \Gamma_{24} \) magnetic phase. In the \( \Gamma_{12} \) phase, the antiferromagnetic vector \( \mathbf{l} \) is in the \( xy \)-plane and the \( z \)-component of magnetization \( M \) is zero. A transition towards the \( \Gamma_{24} \) phase occurs through a rotation of the \( \mathbf{l} \)-vector over an angle \( \delta \phi \) towards the new equilibrium in the \( zx \)-plane with a non-zero \( M_z \) component [25]. (Online version in colour.)

transition at \( T_t = 52 \) K, where the magnetization direction changes by about 15–20°, which can be achieved either by heating or by the application of a static magnetic field.

A short magnetic field pulse should trigger an inertial spin reorientation between the magnetic phases \( \Gamma_{12} \) and \( \Gamma_{24} \). To observe this, however, the pulse must be shorter than the characteristic time of the magnetic eigenmodes, a few picoseconds in the case of an antiferromagnet. Such short pulses can only be provided by the inverse Faraday effect.

Results of calculations based on equation (4.1) are shown in figure 5a. It is seen that the excitation of the spins by a 100 fs magnetic field pulse with an amplitude \( H_p \) triggers damped spin oscillations and may change the equilibrium value of the angle \( \phi \). The new equilibrium orientation depends on the amplitude of the optically induced field pulse: if the pulse amplitude \( H_p < H_c \), the system relaxes back to the initial state \( \phi = 0 \); for \( H_p > H_c \), the resulting state is the \( \Gamma_{24} \) phase with \( \phi = \pi/2 \). The value of the critical field for HoFeO₃ is of the order of 0.5 T. Figure 5 shows that the coordinate \( \phi \) of the spin hardly changes during the 100 fs action of the magnetic field. Instead, the spins acquire sufficient momentum to overcome the potential barrier afterwards and motion with practically the same value \( d\phi/dt \) persists long after the pulse is gone.

To observe such inertial motion of spins in HoFeO₃, pump–probe experiments were performed with 100 fs circularly polarized laser pulses. The pump-induced spin dynamics was monitored by detecting the \( M_z \) component of the magnetization via the magneto-optical Faraday effect in the probe pulse, measured as a function of the delay between the pump and probe pulses. The results of such time-resolved measurements are shown in figure 5b for the two opposite pump helicities. The experimental observations are in excellent agreement with the theoretical predictions.
Figure 5. (a) Numerical simulations and (b) experimental observation at $T = 50.5\,\text{K}$ of inertia-driven spin switching in HoFeO$_3$. In the calculations the reorientation is triggered by a 100fs Gaussian magnetic field pulse directed along the $z$-axis. $H_c$ is the critical field required for the transition $\Gamma_{12} \rightarrow \Gamma_{24}$. The simulations show that at $T = 50.5\,\text{K}$ the critical field is equal to 0.6T. In the experiment the effective magnetic fields along the $z$-axis were generated by right-handed $\sigma^+$ and left-handed $\sigma^-$ circularly polarized laser pulses via the inverse Faraday effect [25]. (Online version in colour.)

agreement with the results of the simulations and thus demonstrate the inertial motion of antiferromagnetically coupled spins. Outside the range between $T_1 = 38\,\text{K}$ and $T_2 = 52\,\text{K}$, such pulses cause only small-amplitude damped oscillations towards the initial state, similar to that discussed in §2 for DyFeO$_3$ above.

5. All-optical magnetization reversal

As is obvious from the previous sections, circularly polarized femtosecond laser pulses act as equally short magnetic field pulses via the inverse Faraday effect. This naturally leads to the following question. Can such optically induced field pulses be used to completely reverse the magnetization of a magnetic domain?

The experiments were performed by placing a sample of the GdFeCo magnetic alloy under a polarizing microscope, where domains with magnetization ‘up’ and ‘down’ could be observed as white and black regions, respectively. To excite the material, amplified pulses from a Ti:sapphire laser were used at a wavelength of $\lambda = 800\,\text{nm}$, at a repetition rate of 1kHz and a pulse width of 40fs. The laser pulses were incident normal to the sample surface, so that an effective optically generated magnetic field would be directed along the magnetization, similar to a conventional recording scheme.

In order to determine unambiguously whether excitation by a single 40fs laser pulse is sufficient to reverse the magnetization, the laser beam was swept at high speed across the sample, so that each pulse landed at a different spot.
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Figure 6. The effect of single 40 fs circularly polarized laser pulses on the magnetic domains in Gd\textsubscript{2}Fe\textsubscript{14.6}Co\textsubscript{3.4}. The domain pattern was obtained by sweeping at high speed (approx. 50 mm s\textsuperscript{-1}) circularly polarized beams across the surface so that every single laser pulse landed at a different spot. The laser fluence was about 2.9 mJ cm\textsuperscript{-2}, and the sample was at 300 K. The small size variation of the written domains is caused by the pulse-to-pulse fluctuation of the laser intensity [13].

(figure 6). One can see that each of the $\sigma^+$ pulses reverses the magnetization in the black domain, but does not affect the magnetization of the white domain. The opposite situation is observed when the sample is exposed to $\sigma^-$ pulses. Thus, during the presence of a single 40 fs laser pulse, information about the angular momentum of the photons is transferred to the magnetic medium, and subsequently recording occurs. These experiments unambiguously demonstrate that all-optical magnetization reversal can be achieved by single 40 fs circularly polarized laser pulses without the aid of an external magnetic field.

These surprising results immediately trigger the following question. What are the relevant time scales and mechanism of such an optically induced magnetization reversal? A precessional switching within 40 fs would require enormous effective magnetic fields above $10^2$ T and unrealistically strong damping. Besides, such strong and short magnetic field pulses are not expected to lead to a deterministic switching of magnetic domains [26]. To address these questions, femtosecond, single-shot, time-resolved optical imaging of magnetic structures was developed and used together with multi-scale modelling beyond the macro-spin approximation [27].

GdFeCo samples were excited by a single circularly polarized laser pulse. A single linearly polarized probe pulse delayed with respect to the pump was used for ultrafast imaging of the magnetic domain structure via the magneto-optical Faraday effect. Magnetic domains with magnetization parallel (‘up’) or antiparallel (‘down’) to the sample normal are seen as white or black regions, respectively, in an image on a charge-coupled device camera. After each write–read event, the initial magnetic state was restored by applying a magnetic field pulse. Taking images of the magnetic structure for different delays between the pump and probe pulses, we were able to visualize the ultrafast dynamics of the laser-induced magnetic changes in the material.

Figure 7a shows images of magnetic domains at different delays after excitation by right- ($\sigma^+$) or left-handed ($\sigma^-$) circularly polarized pulses. In the first few hundreds of femtoseconds, pump pulses of both helicities bring the originally magnetized medium into a strongly non-equilibrium state with no measurable net magnetization, seen as a grey area in the second column of figure 7. The size
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Figure 7. The magnetization evolution in Gd$_{24}$Fe$_{66.5}$Co$_{9.5}$ after the excitation with right- ($\sigma^+$) and left-handed ($\sigma^-$) circularly polarized pulses at room temperature. The domain is initially magnetized ‘up’ (white domain) and ‘down’ (black domain). The last column shows the final state of the domains after a few seconds. The circles show areas actually affected by the pump pulses [27]. (Online version in colour.)

of this demagnetized area is given by the laser beam diameter and the intensity profile. In the following few tens of picoseconds, either the medium relaxes back to the initial state or a small (approx. 5 $\mu$m) domain with a reversed magnetization is formed. It is thus obvious that: (i) the switching proceeds via a strongly non-equilibrium demagnetized state, clearly not following the conventional route of precessional motion; (ii) the final state is defined by the helicity of the 100 fs pump pulse (last column of figure 7); and (iii) the actual switching occurs long after the optical pulse is gone.

The time of the switching is in fact surprising because, in contrast to heat-assisted magnetic recording [28], the reversal time is much longer than that of the effective light-induced magnetic field pulse $H_{\text{eff}}$. The duration of the latter, $\Delta_{\text{eff}}$, is still an open question but it can be different from the full width at half-maximum (FWHM) of the optical pulse. In principle, $\Delta_{\text{eff}}$ can be estimated from the spectrum of the terahertz radiation generated by an Fe film excited by a sub-picosecond visible laser pulse. Based on a half-period oscillation with the lowest frequency in the spectrum reported in [29], the maximum $\Delta_{\text{eff}}$ is about 3 ps. The pulse amplitude $H_{\text{eff}}$, for a typical pump fluence of 2.5 J m$^{-2}$ and the magneto-optical constant of GdFeCo (approx. $3 \times 10^5$ $\circ$ cm$^{-1}$), reaches 20 T.

To understand this new reversal process, the Landau–Lifshitz–Bloch (LLB) equation was solved. This macrospin approach encapsulates very well the response of a set of coupled atomic spins subjected to rapidly varying temperature changes, including the reduction of the magnitude of $M$. The temperature-dependent parameters for the LLB equation, i.e. the longitudinal and transverse susceptibilities and the temperature variation of the magnetization, are calculated atomistically using Langevin dynamics combined with an LLG equation for each spin [30]. The laser-induced increase in the kinetic energy (temperature) of the electrons is simulated using a two-temperature model [31], the parameters for which were taken to be typical for a metal [32] (electron heat capacity $C_e = 1.8 \times 10^6$ J m$^{-3}$ K at room temperature and electron–phonon coupling $G_{\text{el–ph}} = 1.7 \times 10^{18}$ J K$^{-1}$ s). The simulations show that, in the first 100 fs, the electron temperature $T_{\text{el}}$ increases from 300 K up to $T^*_{\text{el}}$ and relaxes with a time constant
of 0.5 ps down to the vicinity of $T_C$. Simultaneously, the spins experience a short pulse of effective magnetic field with amplitude $H_{\text{eff}} = 20\,\text{T}$ and duration $\Delta t_{\text{eff}}$. The possibility of magnetization reversal under these circumstances has been analysed numerically for a volume of 30 nm$^3$, as a function of $T^*_e$ given by the two-temperature model, $\Delta t_{\text{eff}}$ as an unknown parameter and an $H_{\text{eff}}$ of 20 T.

The results of the simulations are plotted in figure 8a as a phase diagram, defining the combinations of $T^*_e$ and $\Delta t_{\text{eff}}$ for which switching occurs for the given $H_{\text{eff}}$. As can be seen from the diagram, a field duration as short as $\Delta t_{\text{eff}} = 250\,\text{fs}$ is needed to reverse the magnetization. For better insights into this reversal process, we simulated the latter for $\Delta t_{\text{eff}} = 250\,\text{fs}$ and $T^*_e = 1130\,\text{K}$. The result is plotted in figure 8b, showing that, already after 250 fs, the effective fields of the two different polarities bring the medium into two different states, while the magnetization is nearly quenched within less than 0.5 ps. This is followed by relaxation either to the initial state or to the state with reversed magnetization, achieved already within 10 ps. The considered pulse duration $\Delta t_{\text{eff}}$ of 250 fs is only 2.5 times larger than the FWHM of the optical pulse in the experiments and well within the estimated lifetime of a medium excitation responsible for $H_{\text{eff}}$. Importantly, in simulations, $\Delta t_{\text{eff}}$ was found to be sensitive to the parameters of the two-temperature model. In particular, an increase in $G_{\text{el--ph}}$ leads to a reduction of the minimum field pulse duration. This shows that the suggested mechanism may, in principle, explain the experimentally observed laser-induced magnetization reversal. This magnetization reversal does not involve precession;
instead, it occurs via a linear reversal mechanism, where the magnetization first vanishes and then reappears in the direction of $H_{\text{eff}}$, avoiding any transverse magnetization components, just as seen in figure 7. Exactly as in the experiments, the initial 250 fs effective magnetic field pulse drives the reversal process, which takes one to two orders of magnitude longer.

Such a theoretically predicted reversal window of electron temperature can be easily verified in the experiment when one changes the intensity of the laser pulse. Figure 8c shows the switchability, i.e. the difference between the final states of magnetization achieved in the experiment with $\sigma^+$- and $\sigma^-$-polarized pulses, as a function of $T_{\text{el}}^*$, calculated from the laser pulse intensity. It is seen that, indeed, switching occurs within a fairly narrow laser intensity range. For intensities below this window, no laser-induced magnetization reversal occurs, whereas if the intensity exceeds a certain level, both helicities result in magnetization reversal, since the laser pulse destroys the magnetic order completely, which is then reconstructed by stray fields [33]. Despite this qualitative agreement between simulations and experiments, the experimentally observed reversal time is several times greater than the calculated 10 ps. The latter, however, is calculated for a 30 nm domain, whereas in our experiments the magnetization in a 5 $\mu$m spot is manipulated.

6. Conclusions and outlook

Optical manipulation of spins by femtosecond laser pulses has developed into an exciting and still expanding research field. The discovery of ultrafast demagnetization by a 60 fs laser pulse, that is, demagnetization on a time scale much faster than the spin–lattice relaxation time, has triggered a wealth of experimental and theoretical research focused on the questions: How fast and by what means can magnetic order be changed and manipulated?

We have demonstrated that, with circularly polarized laser pulses, ultrashort magnetic field pulses can be generated via the so-called inverse Faraday effect, with a strength up to 20 T. With such an optically induced magnetic field, one may selectively excite different modes of magnetic resonance, realize quantum control of spin oscillations and trigger phase transitions non-thermally on a sub-picosecond time scale. Such uniquely short and strong magnetic field pulses were even used to demonstrate an inertia-driven phase transition in an antiferromagnetically ordered material. In addition, it was shown that a single 40 fs circularly polarized laser pulse can controllably reverse magnetization, where the direction of this switching was solely determined by the helicity of the light pulse.

All of these results demonstrated that femtosecond laser pulses are indeed very novel and effective stimuli for the manipulation of magnetic order, which may even lead to all-optical magnetic bits (figure 9). Given the development of compact femtosecond laser sources [34], such an application may become feasible within the next decade.

Further progress in this field of ‘femto-magnetism’ will strongly rely on further developments in theory, combining ab initio theory, atomistic and micromagnetic simulations. The same can be said for the experimental approaches: for a further and detailed understanding of ‘femto-magnetism’, a combination of

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Figure 9. Demonstration of compact all-optical recording of magnetic bits. This was achieved by scanning a circularly polarized laser beam across the sample and simultaneously modulating the polarization of the beam between left and right circularly polarized [13]. (Online version in colour.)

experimental approaches such as combining optical excitation with probing via X-ray magnetic circular dichroism will allow us to get an atomic-scale picture of what actually happens during and after the excitation of a magnetic medium by a femtosecond laser pulse. The development of novel femtosecond slicing approaches at synchrotron sources is therefore very promising.

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