LETTERS

Ultrafast non-thermal control of magnetization by instantaneous photomagnetic pulses

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The demand for ever-increasing density of information storage and speed of manipulation has triggered an intense search for ways to control the magnetization of a medium by means other than magnetic fields[1–5]. Recent experiments on laser-induced demagnetization[6–8] and spin reorientation[9] use ultrafast lasers as a means to manipulate magnetization, accessing timescales of a picosecond or less. However, in all these cases the observed magnetic excitation is the result of optical absorption followed by a rapid temperature increase. This thermal origin of spin excitation considerably limits potential applications because the repetition frequency is limited by the cooling time[10]. Here we demonstrate that circularly polarized femtosecond laser pulses can be used to non-thermally excite and coherently control the spin dynamics in magnets by way of the inverse Faraday effect. Such a photomagnetic interaction is instantaneous and is limited in time by the pulse width (~200 fs in our experiment). Our finding thus reveals an alternative mechanism of ultrafast coherent spin control, and offers prospects for applications of ultrafast lasers in magnetic devices.

The interaction of light with magnetized media is manifested in various magneto-optical phenomena. A good example is the Faraday effect, observed as a rotation of the polarization plane of light transmitted through a magnetic medium[11]:

$$\alpha_F = \frac{\chi}{n}\mathbf{M}\cdot\mathbf{k} \qquad (1)$$

where α_F is the specific Faraday rotation, **M** is the magnetization, n is the refractive index, **k** is the wave vector of light, and χ is the magneto-optical susceptibility, which is a scalar value in isotropic media[12,13]. 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 magnetization $\mathbf{M}(0)$:

$$\mathbf{M}(0) = \frac{\chi}{16\pi}[\mathbf{E}(\omega)\times\mathbf{E}^*(\omega)] \qquad (2)$$

where $\mathbf{E}(\omega)$ and $\mathbf{E}^*(\omega)$ are the electric field of the light wave and its complex conjugate, respectively[13–16]. 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 χ (refs 14, 15). In particular, in the case of the inverse Faraday effect, χ is the ratio between the induced magnetization and the laser intensity. 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 χ 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 can be considered as instantaneous because it takes place on a femtosecond timescale. Indeed, if one stimulates an optical transition into a virtual state with a strong spin–orbit interaction, the following relaxation into the ground state may be accompanied by spin switching and re-emission of a photon with a fixed phase shift and lower energy with respect to that of the incident photon. In magnetically ordered materials, this process is known as excitation of magnons by light[17]. Recent theoretical work has indicated the possibility of laser-induced spin reversal on a femtosecond timescale[18]. However, the experimental demonstration of such non-thermal ultrafast optical control of magnetization has remained an intriguing challenge until now.

The material of choice for our study was dysprosium orthoferrite

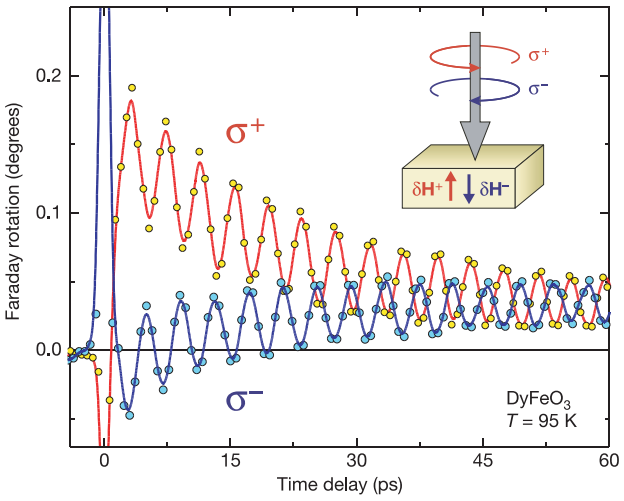

Figure 1 | Magnetic excitations in $DyFeO_3$ probed by the magneto-optical Faraday effect. Two processes can be distinguished: (1) instantaneous changes of the Faraday effect due to the photoexcitation of Fe ions and relaxation back to the high spin ground state $S = 5/2$; (2) oscillations of the Fe spins around their equilibrium direction with an approximately 5 ps period. The circularly polarized pumps of opposite helicities excite oscillations of opposite phase. Inset shows the geometry of the experiment. Vectors $\delta\mathbf{H}^+$ and $\delta\mathbf{H}^-$ represent the effective magnetic fields induced by right-handed σ^+ and left-handed σ^- circularly polarized pumps, respectively.

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$DyFeO_3$, which belongs to the group of rare-earth orthoferrites[19]. This material crystallizes in an orthorhombic perovskite-type structure with four molecular units per unit cell, with a space-group symmetry D_{2h}^{16} (*Pbnm*). The spins of the dysprosium ions are not ordered above 4 K, being in a paramagnetic state. The spins of the Fe^{3+} ions ($3d^5$, ground state $^6A_{1g}$, $S = 5/2$) are coupled antiferromagnetically by isotropic exchange. The Dzyaloshinskii–Moriya interaction[20,21] leads to a slight canting of opposite spins with an angle of about 0.5°, giving rise to a spontaneous magnetization $M_s = 8$ G. Despite the small magnetization, this material exhibits a giant Faraday rotation of about 3,000° cm^{-1} owing to its strong spin–orbit interaction[22]. Thus non-thermal effects of light on the spontaneous magnetization are expected to be large in this material.

The studied $DyFeO_3$ samples were prepared from X-ray oriented boules grown by a floating zone method. They were cut perpendicular to the *z* and *x* crystal axes and were 60 μm thick. For the detection of the optically induced magnetization, we used the direct magneto-optical Faraday effect. Figure 1 shows the temporal evolution of the Faraday rotation in a *z*-cut 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 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, which 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 the angular momentum of the exciting photon.

Figure 2 shows the difference between the Faraday rotations induced by right- and left-handed circularly polarized pump light in the *z*-cut sample for the temperature range between 20 K and 175 K. It is seen that an increase of the temperature results in an increase of the frequency of the oscillations up to 450 GHz at 175 K, while the amplitude of the oscillation decreases. This behaviour is in excellent agreement with previous Raman experiments in $DyFeO_3$ (refs 23–25). The damping of the oscillations in the range of 200 ps is due to magnon scattering on phonons and spins of dysprosium ions. The highest value of the amplitude of the photo-induced oscillations is observed between 20 K and 50 K. The amplitude of the oscillations corresponds to a photo-induced magnetization $M = M_S/16$, where M_S is the saturation magnetization. This ratio is obtained from hysteresis measurements in a static magnetic field that show that the saturated Faraday rotation in a single domain *z*-cut sample is equal to 1°.

From Figs 1 and 2 one can distinguish not only oscillations but also an exponential decay of the equilibrium point on a timescale of about 100 ps. This can be explained by the photo-induced change of the equilibrium orientation of the magnetization and subsequent decay of the equilibrium orientation to the initial state.

Although in principle the effect of optically induced magnetization does not require the absorption of photons, laser control of the spontaneous magnetization and the excitation of coherent spin oscillations is equivalent to photoexcitation of magnons and thus requires some energy. Such photoexcitation of magnons can occur via a process similar to Raman scattering. The inset in Fig. 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 one-photon process. Note that

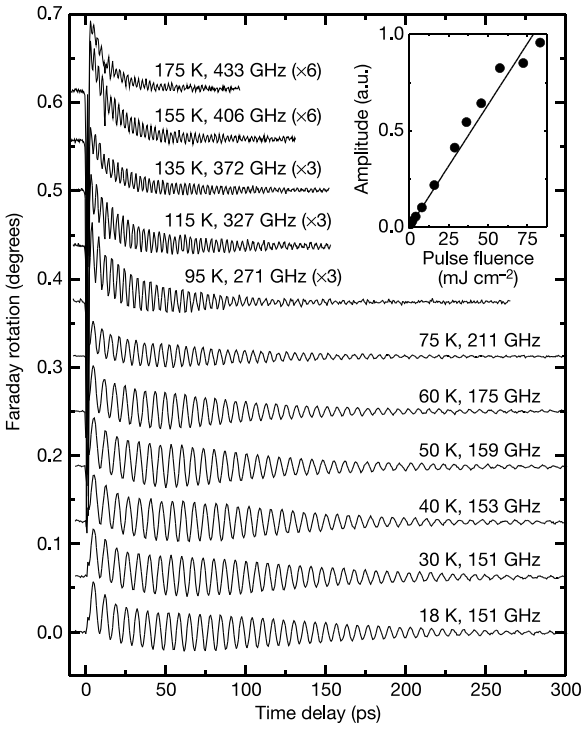

Figure 2 | Excitation of the spin oscillations in $DyFeO_3$ measured at different temperatures in the range between 20 K and 170 K. In order to exclude effects not relevant to magnetic excitations, the difference between the signals for right- and left-handed circularly polarized pump pulses is plotted. Every new curve is shifted from the previous one along the vertical axis over 0.06°. Inset shows the amplitude of the spin oscillations as a function of pump fluence.

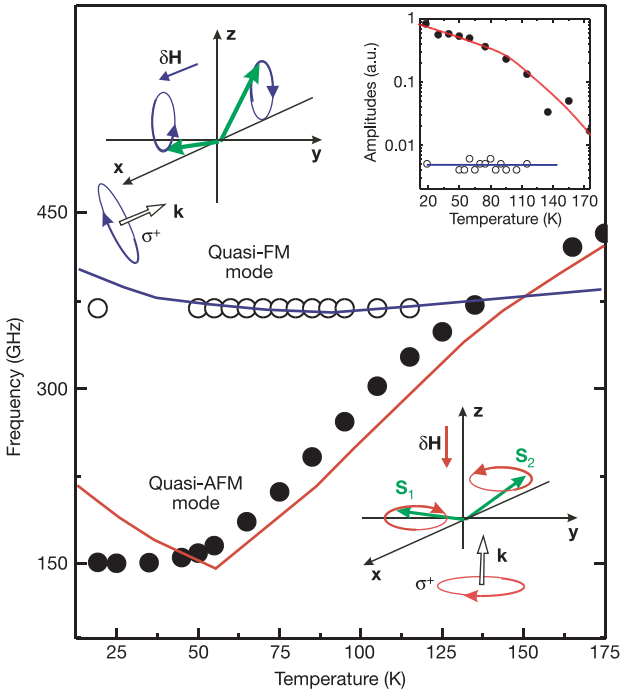

Figure 3 | Temperature dependence of the frequencies of the observed spin oscillations. Filled and open circles show the frequencies of the excited oscillations for laser pulses propagating along the *z* axis and *x* axis, respectively. Red and blue lines show the frequency of the quasi-antiferromagnetic (quasi-AFM) and the quasi-ferromagnetic (quasi-FM) resonance modes from refs 23–25. Top right inset shows the temperature dependence of the oscillation amplitudes. Top left and bottom right insets are respectively schematic representations of the quasi-FM and quasi-AFM modes of the spin resonance. Vectors δH show the directions of the instantaneous magnetic field that is equivalent to the photoexcitation.

extrapolation of the intensity dependence shows that the photo-induced effect on the magnetization would reach the saturation value of M_S at a pump fluence of about 500 mJ cm^{-2}. The effect of such a 200 fs laser pulse on the magnetic system is 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 is thus quite feasible, given a sample of high optical quality.

Owing to the strong anisotropy of the magnetic susceptibility in $DyFeO_3$, magnetic fields in different directions should trigger different types of spin oscillations (Fig. 3). A magnetic field pulse directed along the z axis excites oscillations that correspond to the quasi-antiferromagnetic resonance mode, whereas a field pulse along the x axis will excite the quasi-ferromagnetic resonance mode[23]. These predictions are in excellent agreement with the experimentally observed temperature dependence of the frequency of the oscillations for z-cut and x-cut samples, which closely resemble the temperature dependence for the quasi-antiferromagnetic and the upper quasi-ferromagnetic resonance mode in $DyFeO_3$, respectively (see Fig. 3). All these observations unambiguously show that an ultrashort laser pulse acts on the ensemble of strongly correlated spins as a magnetic field pulse directed along the wave vector of the photons. Simple estimates show that such optical pulses are equivalent to magnetic field pulses with an amplitude of 0.3 T and a full-width at half-maximum of about 200 fs.

Note that the application of a static external magnetic field up to 0.05 T in a direction parallel to the wave vector of light only resulted in a slight change of the frequency (about 1%), again confirming that the effective photo-induced field is dominating the dynamics.

We have demonstrated that with circularly polarized femtosecond laser pulses one can purely optically and thus non-thermally excite and coherently control spin oscillations in the weak ferromagnet $DyFeO_3$. Such optical pulses are shown to be equivalent to 200 fs magnetic field pulses up to 5 T. In view of the great variety of magnetic materials, the direct effect of light on spontaneous magnetization in other materials and at higher temperatures is foreseen. Our findings open new insights into the understanding of ultrafast magnetic excitation and, regarding recent progress in the development of compact ultrafast lasers[26], may provide new prospects for applications of ultrafast photomagnetic phenomena.

METHODS

The measurements were performed in a pump and probe configuration at a photon energy of 1.55 eV using amplified 200 fs pulses from a Ti:sapphire laser at a repetition rate of 1 kHz. The pump beam was circularly polarized, while the probe beam had linear polarization. The intensity ratio between the pump and probe pulses was about 100. Both beams were focused on the sample to a spot diameter of about 200 μm for the pump and somewhat smaller for the probe beam. The pump fluence on the sample was around 30 mJ cm^{-2}. The measurements were done in a cold finger cryostat where the temperature could be stabilized in the range 15–300 K with a precision better than 0.5 K.

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