nature photonics

LETTERS

PUBLISHED ONLINE: 11 AUGUST 2013 | DOI: 10.1038/NPHOTON.2013.209

Off-resonant magnetization dynamics phase-locked to an intense phase-stable terahertz transient

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Controlling magnetization dynamics with a femtosecond laser is attracting interest both in fundamental science and in industry because of the potential to achieve magnetic switching at ever faster speeds. Here, we report a coherent, phase-locked coupling between a high-field single-cycle terahertz transient and the magnetization of ferromagnetic cobalt films. The visualized magnetization dynamics follow the temporal terahertz field oscillation, are tightly locked to the terahertz phase and are induced in the absence of resonant excitations and energy deposition. The magnetic response occurs on the timescale of the stimulus and is thus two orders of magnitude faster than the Larmor precession response. The experimental results are excellently reproduced by the Landau–Lifshift–Gilbert semi-empirical model, indicating its applicability to ultrafast magnetization dynamics and also demonstrating the marginal effect of the co-propagating terahertz electric field. This novel phenomenon of phase-locked control of magnetization with a strong terahertz field suggests new opportunities for ultrafast data storage.

The coherent manipulation of magnetization at ever increasing speeds is of paramount importance in the development of future data storage and processing technologies if the growing demand for increased data rates and access speeds is to be met. Today's widely used mass storage devices, which are based on the giant magnetoresistance (GMR) effect[1,2], have access times limited to the scale of a nanosecond. The quest for faster manipulation of magnetization requires a stimulus that is capable of controlling the magnetization more rapidly. Following this approach, femtosecond near-infrared laser pulses have been used since 1996 to overcome GMR speed limits by inducing faster demagnetization in ferromagnets[3–5], ferrimagnets[6] and garnets[7]. In these experiments, optical pulses manipulate the magnetization by heating the spin system via ultrafast electronic excitation. The associated cooling dynamics, however, limit re-access times to nanoseconds. Moreover, the incoherent nature of heat precludes any possibility of imprinting the phase properties of the stimulus onto the magnetization dynamics. Coherent interaction of the laser with the magnetization has recently been shown to be favourable in regaining equilibrium on a faster timescale and counteracting heat-induced demagnetization[8–11]. However, to date, a magnetization response sensitive to the pulse phase has been observed only for resonant excitation of spin waves on a tens of picoseconds timescale. Here, we visualize, for the first time, off-resonant femtosecond magnetization dynamics following an ultrastrong, phase-stable laser terahertz field[12,13]. In this new interaction regime the laser's phase and field magnitude characteristics are directly imprinted onto the magnetization response, in the absence of any resonant mode. The off-resonant phase-locking mechanism presented here injects only minor entropy into the system, thus avoiding the speed limitation caused by the cooling process. Sub-terahertz pulses have been used before to off-resonantly excite magnetization dynamics[14]. However, the observed magnetic response returned to equilibrium over a timescale much longer than the driving picosecond stimulus, and the response seemed to be dominated by the intensity envelope rather than the **B**-field shape and phase. The much faster and phase-locked mechanism presented here is expected to affect next-generation storage technology, because the field-controlled mechanism enables ultrafast reversible interaction.

As a model system, a 10 nm thin cobalt film, which exhibits in-plane magnetization due to shape anisotropy, was used at room temperature. Before terahertz excitation, an external field $\mathbf{B}_{\mathrm{ext}}$, oriented 10° from the cobalt plane, was used to drive the macroscopic magnetic moment to saturation (Fig. 1). The linearly polarized terahertz pulse impinged onto the sample surface with an angle of incidence of 20° from the sample normal. This pulse propagated collinearly with a synchronous sub-50 fs probe pulse ($\lambda = 800$ nm), sampling the terahertz induced coherent magnetization dynamics via the magneto-optical Kerr effect (MOKE). In this geometry, MOKE primarily probes the out-of-plane magnetization component. The octave-spanning stimulus centred at 2.1 THz carries ~1.5 optical cycles with a field amplitude of ~0.4 T. Most important for the observation of subcycle magnetization dynamics is the fact that the terahertz magnetic field $\mathbf{B}(t)$ is phase-locked over consecutive shots to its intensity envelope $|\mathbf{B}^2(t)|$.

Figure 2a presents the observed time-resolved MOKE signal (red line), which shows the magnetization dynamics phase-locked with the terahertz magnetic field (blue dots). Apart from a quarter period phase shift, corresponding to a delay of ~50 fs, the MOKE signal exhibits a variation that is close to identical to the driving terahertz magnetic field. The MOKE frequency spectra depicted in Fig. 2b (red line) show the occurrence of terahertz constituents with the same frequencies as observed in the terahertz stimulus spectrum (blue dots). The magnetization dynamics do not rely on resonant excitation[9] nor Larmor precession. Contrary to previous results[14], the subpicosecond magnetic response presented here is fully dominated by the phase of the terahertz stimulus.

The observed dynamics can be understood within the empirical framework of the Landau–Lifschitz–Gilbert (LLG) formalism[15], in which magnetization evolution is given by $\mathrm{d}\mathbf{M}/\mathrm{d}t = -\gamma(\mathbf{M} \times \mathbf{B}) - \gamma\alpha(\mathbf{M} \times (\mathbf{M} \times \mathbf{B}))/|\mathbf{M}|$. Here, the first term on the right-hand side describes precession around an effective field **B** with Larmor

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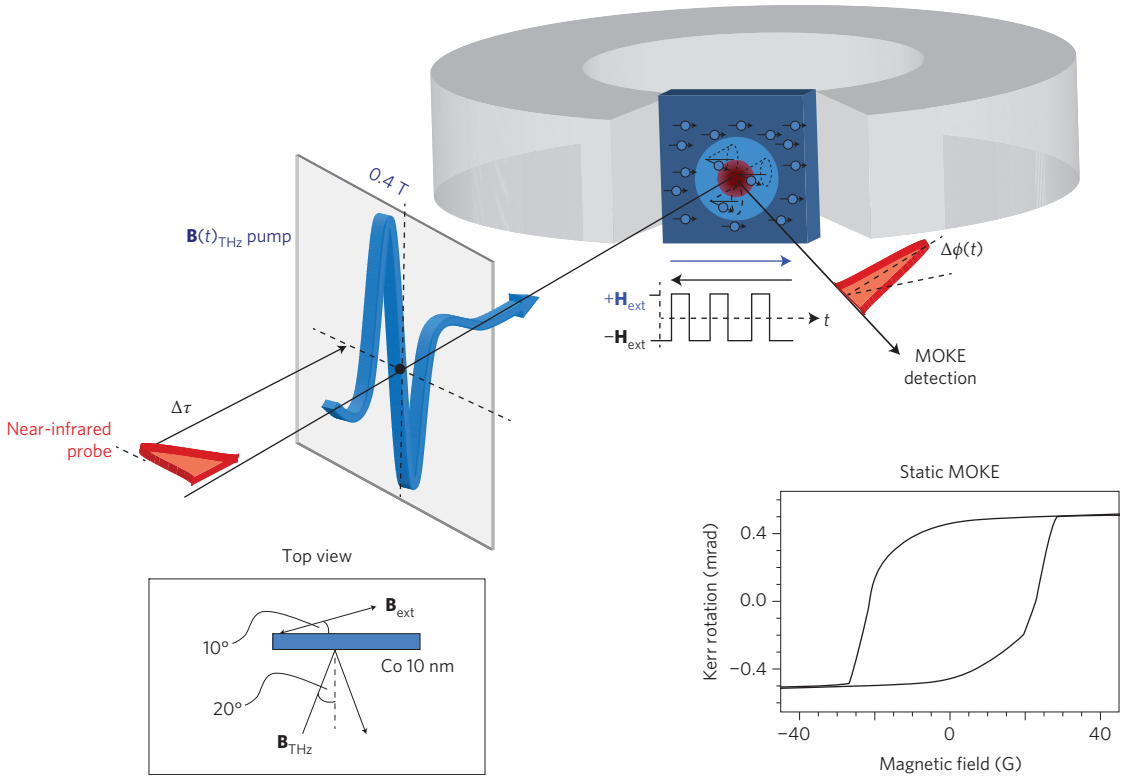

Figure 1 | Time-resolved magneto-optical Kerr effect (MOKE) set-up that allows for the measurement of ultrafast moment dynamics on a room-temperature cobalt sample surface. The strong 0.4 T single-cycle terahertz magnetic field (blue) is linearly polarized and carries an absolute phase, which remains constant for consecutive shots. The terahertz pump pulse hits the sample 20° off-normal incidence. The collinear 50 fs probe pulse originates from the same laser as used for terahertz generation and allows for jitter-free time-resolved measurement of magnetization dynamics via MOKE. The sample is placed at 10° in an external static magnetic field ($\mathbf{B}_{ext} = 0.01$ T), which can be inverted after a pump–probe cycle (by inverting the magnet current) in order to improve the signal-to-noise ratio of the Kerr rotation measurement. The static MOKE hysteresis curve as a function of $\mathbf{B}_{ext}$ is presented in the right inset. In our set-up, the maximum optical Kerr rotation is $\pm$0.5 mrad to fully reverse the cobalt magnetization.

frequency $\gamma|\mathbf{B}|$, and the second dissipative relaxation towards $\mathbf{B}$. In this equation, γ is the gyromagnetic constant and α is the empirical damping factor, which is taken to be 0.014 and embodies the diverse dissipative processes involving lattice and electronic degrees of freedom[16]. The $\mathbf{B}$ field has contributions arising from the terahertz pulse, the constant external magnetic field of 0.01 T, and the demagnetization field associated with the cobalt thin-film geometry. Using the LLG equation, a simulated MOKE response can be calculated via the micro-magnetics simulation package OOMMF[17]. This is plotted in Fig. 2 (black curves), and shows extremely good overall agreement with the experimental measurement. A more detailed account of the modelling method and the simulated three-dimensional magnetization dynamics is given in Supplementary section 'Micromagnetic simulations'. The LLG equation shows that the material's out-of-plane response, which is the component dominating the measured MOKE signal, is entirely determined by the LLG precessional term and the terahertz field contribution. Indeed, the damping effects and thin-film shape anisotropy can be ignored over the timescale of the magnetization dynamics (Supplementary Fig. S1). The out-of-plane magnetization dynamics is therefore considerably simplified and the LLG equation reduces to the form $\mathrm{d}\theta/\mathrm{d}t \cong -\gamma B_z^{\mathrm{THz}}$, with the solution $\theta(t) \cong \theta(t=0) + \gamma \int_0^t \mathrm{d}t B_z^{\mathrm{THz}}(t)$, where B_z^{THz} is the in-plane terahertz field perpendicular to the initial cobalt magnetic moments (Supplementary section 'Micromagnetic simulations'). Here, $\theta(t)$ is the angle between the magnetization and the thin-film plane. This integral dependence accounts for the observed phase shift between the stimulus and the magnetic response, which differs negligibly from the fully three-dimensional OOMMF LLG simulations shown in the figures. The quantitative agreement between simulated out-of-plane magnetization and the experiment demonstrates that both precession and damping effects are too slow to affect the out-of-plane response over the terahertz stimulus timescale. This good agreement also indicates that the co-propagating intense electric field has only a negligible influence on the magnetization dynamics at this timescale.

Such an inherently phase-locked interaction between stimulus and magnetization has never been reported previously and occurs on a femtosecond timescale, which is faster than expected[18]. The recorded ultrafast dynamics become observable due to the absence of hot electrons and ionizing effects. In fact, the terahertz photon energy is almost three orders of magnitude lower than that of a near-infrared photon, as generally used in ultrafast magnetization experiments (0.004 eV versus 1.6 eV). This avoids extensive heating and enables the discovery of unexpected phase- and field-sensitive coherent motions of the magnetization. The measured $\mathbf{M}(t)$ in Fig. 2 indicates that the phase properties of the magnetization are fully determined by the terahertz magnetic field, which we refer to as 'off-resonant coherent magnetization dynamics'. While conventional heating above the Curie temperature results in a decrease in the macroscopic magnetization magnitude $|\mathbf{M}|$, the coherent coupling demonstrated here alters exclusively the orientation of $\mathbf{M}$, and not its net magnitude.

Further insight into the magnetization dynamics may be gained by changing the vector direction of the terahertz field. In fact, it is experimentally seen that the magnetization response reduces as the terahertz magnetic field increasingly points towards the initial moment direction of the cobalt atoms (Fig. 3a). The LLG equation can also accurately describe this effect (Fig. 3b), indicating that the

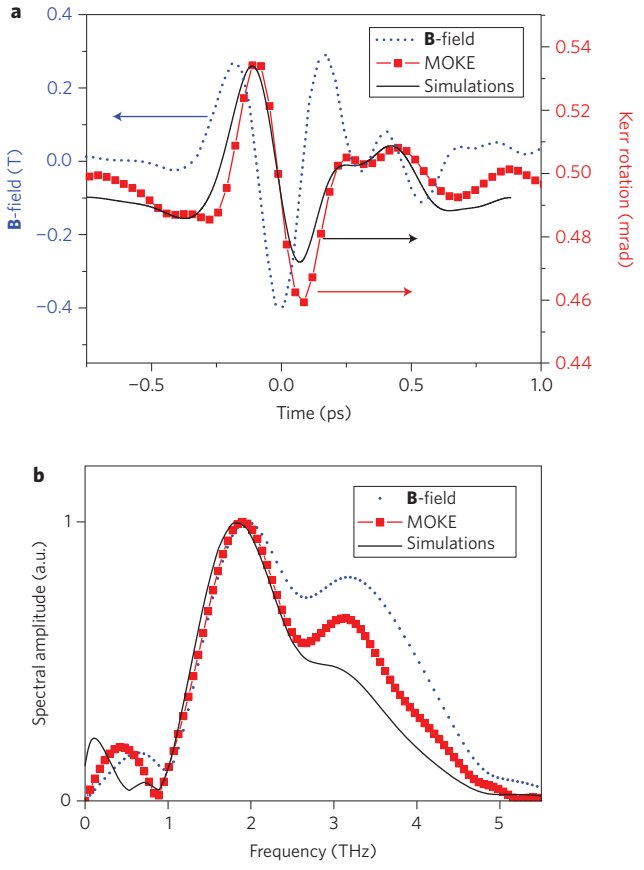

Figure 2 | Measurements of the phase-locked out-of-plane magnetization dynamics initiated by the strong terahertz transient. a, Femtosecond magnetization dynamics represented by the MOKE signal (red curve) and initiated by the phase-stable single-cycle terahertz magnetic field (blue dotted curve) follow the terahertz field oscillations. **b**, The corresponding spectral intensities of the terahertz pulse and MOKE response are almost identical, which illustrates that the coupling between the terahertz field and magnetization does not rely on any resonant excitation or spin waves. The magnetization vector evolution calculated using the OOMMF code[17] (black curve) as described by the LLG equation reflects the observed ultrafast magnetization dynamics well in both temporal and spectral domains.

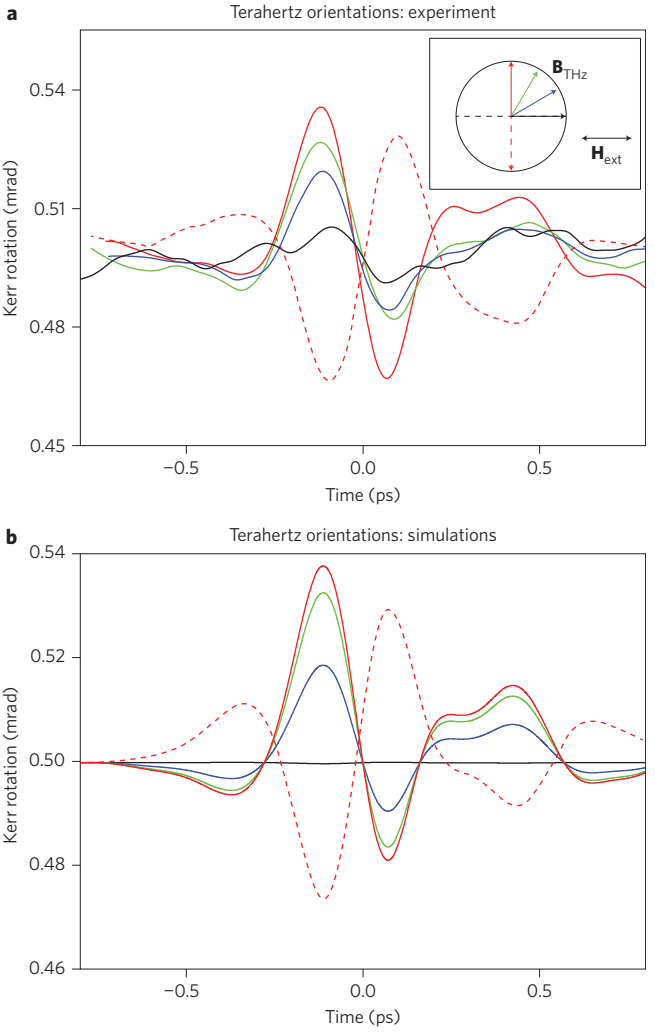

Figure 3 | Magnetization response dependence on the terahertz input polarization direction. a,**b**, Measured (**a**) and calculated (**b**) MOKE for different $\mathbf{B}_{THz}$ polarization angles oriented as shown in the inset. The maximum coupling occurs for the magnetic field orthogonal to the **M** vector (red curves) and decreases to the noise level for parallel alignment (black curve). For the blue and green curves, $\mathbf{B}_{THz}$ is oriented at 30° and 60° with respect to the **M** vector, respectively. **b**, OOMMF simulations[17] based on the LLG equation reproduce the experimental results with excellent agreement. When the $\mathbf{B}_{THz}$ field polarity is reversed, the magnetization response is equally reversed, demonstrating the absence of any incoherent heating stimulus and the phase-locked nature of the magnetization dynamics (solid and dashed red lines).

reduced angle between the terahertz field and the cobalt moments results in a smaller magnetic torque. The observed variation of the MOKE signal for parallel alignment of the terahertz **B**-field and sample magnetization (black curve in Fig. 3) reflects the noise level of our experiment.

The measurement and simulation performed with a terahertz stimulus with reversed magnetic field polarization result in a likewise inverted MOKE signal (solid and dashed red lines in Fig. 3a,b). The magnetic response for the two polarizations produces the same temporal shape and spectral components, providing evidence of the phase-locked coupling mechanism, which would not be expected for incoherent heating.

Over the investigated range, the out-of-plane magnetization is inherently linear with respect to the magnitude of the terahertz field (Fig. 4a). Here, three MOKE traces are recorded for different field strengths down to 40% of the maximum. The experimental and simulated curves demonstrate a proportional relation between the amplitude of the stimulus and the MOKE responses. As the terahertz magnetic field strength varies, the timescale of the magnetization response and its spectral components remain substantially unchanged. This confirms that the observed dynamics occur far from saturation and in the absence of dissipative processes.

In conclusion, we have shown that high-field terahertz pulses with a stable absolute phase offer an entirely new experimental avenue for the exploration and control of coherent subcycle femtosecond magnetization dynamics in ferromagnetic thin films. A strong phase-locking between the intense terahertz pulse and magnetization is established, with an absence of resonances (magnons, electromagnons and other modes) and heat injection. In this high-field regime, magnetization evolution is driven by the amplitude and phase of the terahertz laser. Previously inaccessible subcycle magnetization dynamics are therefore visualized. Remarkably, the magnetization response at this short timescale is very well understood by simple classical precession. This gives experimental insight into the bounds of applicability of the phenomenological LLG approach, and also shows that the terahertz electric field has little influence on the magnetization dynamics.

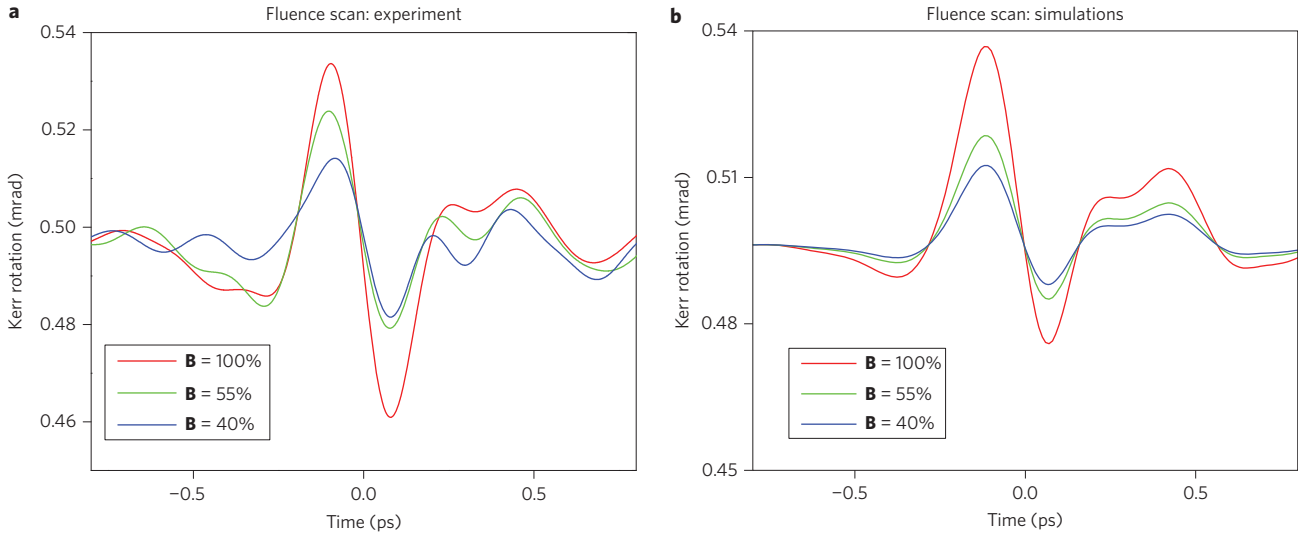

Figure 4 | Magnetic response as a function of terahertz field strength. a,**b**, Experimental (**a**) and simulated (**b**) MOKE response for different terahertz magnetic field strengths. For the measurement, the terahertz magnetic stimulus is oriented orthogonally with respect to the plane of incidence for maximum coupling with the magnetization vector. The curve indicates a linear dependence with respect to the amplitude of the magnetic terahertz field. The LLG equation (OOMMF) reproduces, with excellent agreement, the observed dependence (**b**).

The presented new concept of a phase-stable non-ionizing high-field stimulus opens the door to pushing light-induced phase-locked magnetization control towards the yet unidentified speed limit of magnetic switching.

Received 25 November 2012; accepted 2 July 2013; published online 11 August 2013

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Acknowledgements

This work was carried out at the Paul Scherrer Institute and was supported by the Swiss National Science Foundation (grant PP00P2_128493). The authors acknowledge A. Kleibert for helpful discussions and thank M. Paraliev for support. B.T. is supported by the ERASMUS Mundus program. J.L. acknowledges the DYNAVO project for financial support for upgrading the magnetron sputtering. C.P.H. acknowledges association with the National Center of Competence in Research on Molecular Ultrafast Science and Technology (NCCR-MUST).

Author contributions

C.P.H. and J.L. conceived the experiment. C.P.H. coordinated the project and wrote the manuscript, together with P.M.D. and C.V. The experiment was performed by C.V., C.R., J.L., F.A. and C.P.H., and B.T. fabricated and characterized the samples. P.M.D. performed numerical investigations of the magnetization dynamics. C.P.H., J.L., C.V., C.R. and P.M.D. contributed to data analysis.

Additional information

Supplementary information is available in the online version of the paper. Reprints and permissions information is available online at www.nature.com/reprints. Correspondence and requests for materials should be addressed to C.P.H.

Competing financial interests

The authors declare no competing financial interests.