Simultaneous electronic and the magnetic excitation of a ferromagnet by intense THz pulses

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Abstract

The speed of magnetization reversal is a key feature in magnetic data storage. Magnetic fields from intense THz pulses have been recently shown to induce small magnetization dynamics in a cobalt thin film on the sub-picosecond time scale. Here, we show that at higher field intensities, the THz electric field starts playing a role, strongly changing the dielectric properties of the cobalt thin film. Both the electronic and magnetic responses are found to occur simultaneously, with the electric field response persistent on a time scale orders of magnitude longer than the THz stimulus.

The speed of magnetization reversal is a key feature for ultrafast magnetic storage technology [1, 2]. In the state of the art data recording technology, the magnetization reversal, i.e., the complete inversion of the direction of the magnetization vector ($M$) occurs on a relatively long time scale on the order of the nanosecond. In the past, femtosecond optical pulses have been used to induce faster magnetic phase transition, but the impact of the optical pulse leads to thermal effects and therefore to a long recovery time (nanoseconds), which significantly limits the magnetic writing rate [3–6]. Using intense magnetic field pulses from a relativistic electron beam, magnetic switching on a picosecond timescale was observed post mortem [1, 2]. However, time-resolved exploration of the interplay of the electric and the magnetic field component with the ferromagnetic sample could not be performed at this large scale facility.

An alternative approach towards precessional magnetic switching on the picosecond timescale relies on the magnetic field component of a strongly asymmetric single cycle terahertz (THz) pulse [7–15] phase-locked to the spins. As the THz pulse carrier frequency matches well the natural timescale of the spin motions direct control of the spin dynamics with the THz magnetic field component becomes feasible by inducing a torque [16–18]. Recently, this concept has been proven experimentally by Vicario \textit{et al} where a small excursion of the spins was introduced non-resonantly by an intense 0.4 T THz field [17]. The observed coherent spin dynamics were more than an order of magnitude faster than in previous studies. Effects associated to the THz electric field were not observed. While this proof of principle experiment showed the great potential of intense THz radiation for precessional spin motion control, full magnetic switching requires much higher field intensities than what was available [19] in [17]. Here, we report on dynamics induced by an up-scaled THz pulse [20] intensity targeting large amplitude magnetization dynamics. We observed that at high field intensities, the THz electric field starts playing a significant role in changing the dielectric properties of the magnetic material.

The THz source used for the present studies provides intense THz bullets with maximum electric and magnetic field strengths of 6.7 MV cm$^{-1}$ and 2.23 T, respectively [20]. The corresponding peak intensity is 59 GW cm$^{-2}$. The THz was generated using optical rectification of a short wavelength infrared pulses ($\lambda = 1.5$ $\mu$m) in an organic crystal DSTMS. Figures 1(a) and (b) show the temporal field profile recorded by electro-optical sampling and the corresponding spectral amplitude. The THz spot in the focus, recorded using an uncooled micro-bolometer array (NEC Inc.) is shown in figure 1(c).

To measure the THz-induced nonlinear dynamics, we used a collinear THz-pump/optical Kerr effect (KE) probe scheme. The investigated sample is a 20 nm–thick Co film deposited on a Si substrate and capped with...
2 nm–thick layer of Pt. The latter is a protection layer which plays no role in the experiment presented here. We verified that by comparing the measurement against uncoated fresh sample. The beam configuration at the sample position is shown in figure 1(d): the THz pump and a collinear 800 nm centered probe beam impinge on the sample at an angle of 15° measured from the normal. The reflected probe beam was then collected and the THz-induced KE rotation was analyzed using a balanced detection scheme (a quarter wave plate followed by a Wollaston prism). An external magnetic field \( B \) bias (parallel to the sample plane and the plane of incidence) is used to prepare a well-defined in-plane magnetization state \( \mathbf{M} \) prior to pumping. During the measurements the THz polarization direction could be altered by rotating the THz generation crystal along with the near infrared pump beam polarization.

Nonthermal temporal magnetic evolution under the application of an external (THz) magnetic field is macroscopically governed by the Landau–Lifschitz–Gilbert (LLG) equation \([18]\). At relatively weak excitation, the amplitude of the induced ultrafast precessions is nearly a linear function of the THz magnetic field \( \mathbf{H}^{\text{THz}} \). The simplified LLG model does not take into account the effect of the THz electric field and thus may be inappropriate for the physics observed at high fields. High field intensities (on the order of several Tesla in the sub–THz range and much more as the excitation frequency increases \([18]\)) and proper pulse shaping \([21]\), however, are required for Terahertz-induced magnetization reversal. In this article we were aiming to increase

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**Figure 1.** (a) The temporal trace of the exciting THz pulse retrieved using electro–optical sampling technique (peak of 6.7 MV cm\(^{-1}\)) \([20]\). (b) The corresponding amplitude spectrum. (c) The THz spot size at the sample position measured using a micro–bolometer array–based camera. (d) Schematic diagram of the beams configurations on the sample.
the excitation intensity in order to study the role of the THz electric field component ($E_{\text{THz}}$) in the observed dynamics. Two major $E_{\text{THz}}$-induced effects can be considered. The first mechanism is the change in the dielectric tensor due to Kerr-like and thermal nonlinearities. The second effect is the thermal demagnetization of the sample induced by heating the electrons above the Curie temperature, similar to the majority of laser-induced magnetization dynamics in the optical regimes. The second mechanism depends on the excitation fluence. In our experimental setup, the sample is exposed to two external excitations, namely the THz (electric and magnetic) fields and the bias magnetic field. In phase-sensitive detection as we used here, one of the external excitations is modulated at a frequency which is used as a reference for our acquisition system. This allows us to distinguish between the magnetic and non-magnetic (electronic) dynamics. While modulating the THz pump reveals both dynamics, modulating the external bias shows only the magnetic ones.

The nonlinear dynamics induced by $E_{\text{THz}}$ and $H_{\text{THz}}$ are very different. For simplicity, we assume no coupling between $E_{\text{THz}}$ and $M$. This assumption is justified by the fact that, generally, temporal changes in $M$ require a nonzero torque $\mathbf{M} \times \mathbf{H}_{\text{eff}}$ (i.e. it vanishes for $M || |H_{\text{THz}}|$) and is a maximum for $M \perp |H_{\text{THz}}|$ [17, 18] and that the excitation of the electronic system does not depend on the direction of $M$. Second, while the $H_{\text{THz}}$-induced changes in the experimentally-dominant out-of-plane $M$ component are generally linear with the exciting $H_{\text{THz}}$ (at relatively low excitation as we show here), the $E_{\text{THz}}$-induced changes of the material refractive index are proportional to ($E_{\text{THz}}$)$^2$. Due to the limited signal to noise ratio in our 100 Hz laser system, we could not verify this scaling by scanning the fluence.

Figures 2(c) and (d) show the measured probe Kerr rotation under two conditions $\pm M \perp H_{\text{THz}}$ and $\pm M || |H_{\text{THz}}|$. A giant rotation is observed with an instantaneous rise time and duration over a long delay $> 20$ ps. A zoom-in on the initial dynamics is shown in figures 2(e) and (f). In the case of $\pm M \perp H_{\text{THz}}$, clear oscillations following the THz excitation field can be observed on the sub-ps scale superimposed on a large incoherent signal. The origin of these oscillations is the torque induced by the $M \perp H_{\text{THz}}$. Although the used 2 T magnetic field is strong, the instantaneous excitation frequency $f$ is high. In this regime, it was previously found that the $f = \gamma H_{\text{THz}}/2\pi$ with $\gamma$ being the gyromagnetic ratio. At $\sim 0.2$–0.3 THz, both theory [18] and experiment [1, 2]
showed a required switching field of $\sim 6$ T. If the depicted picture of precessional dynamics still holds, at our THz carrier frequency ($\sim 3$ THz), we need an order of magnitude higher magnetic field ($\sim 60$ T). To isolate the coherent precessions, we subtracted the trace pairs with $B^\pm$. The measured rotation has superimposed contributions from the excitation of both the electronic and magnetic systems. The sign of probe rotation does not depend on the sign of the THz electric field. On the contrary, the vectorial nature of the coherent precessional dynamics suggests that the application of two stimuli with opposite signs $H_{THz}^\pm$ leads to opposite torques $M_{H_{eff}}^\pm$ and thus perfectly reversed out-of-plane temporal magnetization dynamics. The probe polarization rotation direction (sign) follows the direction of this torque.

In order to distinguish between the two dynamics, we performed all the measurements in two distinct conditions with $+B$ and $-B$ (that is, $+M$ and $-M$). In this way, by subtracting the two traces the coherent precessions is obtained while the incoherent dynamics were extracted by adding the two measurements. The results are shown in figures 3(a) and (b). In the case of $M \perp H_{THz}$, (a) shows clear temporal oscillations representing magnetic precessions and corresponding to the THz temporal oscillations. (c) Reports the corresponding amplitude spectra to green curves in (a), (blue) and (b), (red).

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Finally, we would like to mention that the magnitude of the polarization rotation depends strongly on the probe initial polarization. We verified that by measuring the polarization rotation in the case of $M \parallel H_{THz}$ at different polarization of the probe (figure 4). Similar to [22], we obtained the maximum sensitivity for an input polarization of $\sim 45^\circ$. This effect may have magnetic or non-magnetic origin. In our case, it originates from the non-magnetic dielectric properties of the metal film as we discussed earlier. Nevertheless, the angle sensitivity depends on the dielectric properties of the film and the substrate.
In conclusion, we used ultra-intense THz bullets centered at 4 THz with an electric and magnetic fields of 6.7 MV cm$^{-1}$ and 2.3 T, respectively to trigger both the electronic and magnetic response in a thin Cobalt film. In addition to the previously observed THz magnetic field-induced spin precessions, we observed electric field-driven changes in the sample dielectric tensor and thus the optical properties. The two effects are found to occur simultaneously on the ultrafast time scale of the triggering THz pulse. While the magnetic response takes place only during the THz stimulus, the electric field-induced dynamics last up to a much longer time scale (>20 ps). We attribute that to THz-induced thermal dynamics in the material. We conclude that the role of the electric field cannot be excluded in the studies of THz-magnetism, particularly at higher field intensities. At even higher fluences, material demagnetization and damage may take place. Such effects generally scale quadratically with the electric field.

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Figure 4. Sensitivity of the probes rotation to the polarization. 0° corresponds to P-polarization and 90° corresponds to S-polarization.
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