Ultrafast Electron Dynamics in Magnetic Thin Films

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Abstract: In past decades, ultrafast spin dynamics in magnetic systems have been associated with heat deposition from high energy laser pulses, limiting the selective access to spin order. Here, we use a long wavelength terahertz (THz) pump–optical probe setup to measure structural features in the ultrafast time scale. We find that complete demagnetization is possible with <6 THz pulses. This occurs concurrently with longitudinal acoustic phonons and an electronic response.

Keywords: terahertz spectroscopy; ultrafast demagnetization; charge-spin dynamics

1. Introduction

The study of ultrafast magnetism is crucial in advancing our understanding of magnetic systems, as well as developing ultrafast memory devices. The difficulty of this subject lies in spin dynamics, and its role as a part of a larger picture of the barely understood coupled structural dynamics.

As an example, when a ferromagnetic film is excited by a femtosecond (fs) laser pulse, partial demagnetization of the material occurs within ∼100 fs, followed by a remagnetisation to the original state on a longer time scale. This was first observed in 1996 for optical laser pulses [1], and later also for THz laser pulses [2–5]. While this phenomenon has led to intense investigations in the field by both experimentalists and theorists, it has now become evident that this process is mediated by a complex puzzle of strongly entangled pieces [6,7]. To elaborate on this, it is essential to understand the physical mechanisms that can influence the spin angular momenta of the electrons responsible for magnetisation.

In order to simplify the almost unfeasible task of tackling coupled structural dynamics, we examine the phenomena responsible and their respective timescales. This allows us to decouple mechanisms which have infinitesimal effects on each other.

As Born and Oppenheimer pointed out, a separation of atomic nucleus dynamics and the surrounding electrons can be made. This is because electrons will adiabatically follow any comparatively slow changes of the lattice. That is, typical dynamics of phonons are on the picosecond timescale, whereas electron dynamics in metals can occur in as short a time span as the attosecond timescale.

Similarly, a separation of electron charge and spin degrees of freedom is reasonable, due to the slower time scales that are observed for the dynamics of the spins, ∼100 fs, than that for the electron scattering, ∼10 fs.

The dynamics of magnetism in a stimulated material could be affected by any number of combinations of the mechanisms described above. In particular, there may be different channels of light–matter interactions; here, we elaborate on the two extremes: direct coupling of the laser pulses to electronic spins, and intermediate charge and/or phonon excitations which induce magnetisation dynamics [1,8–13].
In the direct scenario, the local electro-magnetic field of the stimulus directly couples with the electronic spin angular momenta [10,14], such that there is minimal heating of the electronic structure.

In the indirect channel, the laser photons do not primarily change the magnetisation. Instead, photon energy is transferred to the system in the form of an increased electronic and/or atomic temperature. This process can excite electron-hole pairs, lattice phonons and magnons, which will non-selectively spin-flip scatter with the electrons responsible for magnetisation, modifying their angular momenta [15–18].

Demagnetization effects in past reports with optical laser pulses are mediated through this indirect channel. This results in a non-equilibrium state, where the electronic temperature is raised above the phononic temperature, and an imbalance exists between chemical potentials of electrons with differing spins. This difference is the driving force for ultrafast demagnetization [19,20]. The system then evolves through the above discussed spin-flip scatterings, leading to changes in the orientations of atomic magnetic moments. As a result of this, a temporary demagnetization is observed, followed by remagnetisation to the original state through the balance of the chemical potentials, and the thermalisation of the electrons and phonons. Furthermore, in samples with metallic substrates, there is also the contribution of a superdiffusion process [21]. In this process, the excited mobile spin carriers are transferred onto a conducting substrate.

Lastly, the total effective magnetic field exerts a Zeeman torque onto the atomic magnetic moments. This minute contribution on the magnetisation $M(t)$ is a coherent oscillation in time, following the action of the electro-magnetic wave. However, excited electrons with the above discussed spin flip scattering alter $M(t)$ incoherently in time, leading to fluctuations of the phase; dephasing the system. Therefore, the precessional motion is not typically observable after optical laser pulse excitations. Although the effects of Zeeman torque have been disproven to influence electronic spins to the point of demagnetization in the Mott insulator NiO [22], this effect has not been shown in metals.

Optical pulse/thin film demagnetization has been extensively examined, and the majority of these studies report results on indirect and indiscriminate spin excitations [23–25]. Therefore, to increase the coherency and control of the interaction, experiments have been preformed with THz laser stimuli. The THz field cycle oscillates on a similar timescale as the natural speed of electronic spin motions, as opposed to optical pulse stimuli, which oscillate at a much faster timescale.

At low field amplitudes, THz lasers are therefore expected to directly and coherently couple with electronic spin dynamics. This interaction leads to a precessional motion of $M(t)$ due to the Zeeman torque, allowing for selective control of the magnetic phase. Moreover, THz photon energies are three orders of magnitude smaller than optical photon energies, inducing significantly less heating at low field amplitudes, and reducing the possibility for spin flip scattering between electrons and excited particles. This direct coupling of THz and demagnetization was reported for ferromagnetic cobalt films [26], where a coherent, phase-locked demagnetization response was observed under excitations from THz pulses.

With the recent advances of THz source technology, THz laser pulses with field amplitudes of several Teslas are now accessible. This opens new venues for exploring the complete precessional reversal of $M(t)$. From a technological perspective, obtaining such a reversal would be a milestone, as the time scales involved would be significantly shorter than the Larmor precession obtained in present-day technology. Clearing a potential pathway to increase processor speeds.

However, high field amplitudes come at the cost of a strong heating of the sample, and a loss of the coherent interaction between $M(t)$ and the THz laser field. In this regime, incoherent electronic excitations with subsequent spin-flip scatterings induce a dephasing of $M(t)$ without precessional motion. This is similar to the processes observed after optical pulses. Finally, an additional challenge at large field comes in the form of per-
manent modifications of film properties, and possible damage observable with scanning electron microscopy.

Here, we approach the ultrafast magnetisation dynamics from the photon energy perspective, where we compare excitation dynamics from high (optical) to low (THz) photon energies. We experimentally study the THz-induced ultrafast electronic and structural dynamics in nickel thin films.

In the THz frequency range, partial demagnetization in Ni has been previously reported, followed by permanent demagnetization [2]. Here, we carefully design the experiment to accurately map the THz-induced demagnetization. We report that sufficiently intense THz pulses lead to full demagnetization on the ultrafast time scale without sample damage. The process is combined with strong electronic excitation and the generation of acoustic phonons.

2. Materials and Methods

For our experiment, a 15 nm sputtered Ni thin film on Si substrate was chosen as the sample. This is due to the many extensive studies performed upon the ultrafast magnetism of the material, first reported by Beaurepaire et al. in 1996.

The setup is a tightly focused terahertz bullet scheme, where the THz pulse is focused at the diffraction limit to reach the maximum possible intensity. The THz pulse is generated by the optical rectification of near infrared pulses (1550 nm central frequency, 100 Hz, 50 fs, 3.5 mJ, light conversion OPA system) and an organic crystal DAST (Swiss Terahertz GmbH), 350 µm thick, and is modulated via a mechanical chopper at 37 Hz. The beam is expanded then focused on the sample (three off axis parabolic mirrors scheme). The probe beam is 800 nm centred, 70 fs, 100 Hz, and collinear with the THz pump. The sample was placed to allow for an incident angle of 45°. Both probe and pump were linearly polarized, and the full setup is illustrated in Figure 1a. The THz-induced magnetisation dynamics lead to birefringence measured with a collinear 800 nm probe (observed as an electrical signal on the balanced detector, which is linearly proportional to the amount of birefringence). The temporal and spectral contents of the THz pulse are shown in Figure 1b,c. All measurements of the ratio between the magnetisation and its saturation value $M(t)/M_s$, as shown in Figure 1, were performed by modulating the external magnetic field at 40 mT and 25 Hz. This frequency was used as a reference for our acquisition system to eliminate any non-magnetic contribution to the measured signal.

The magnetic field modulation is locked to the laser clock frequency, and its magnitude is larger than the saturation magnetic field of the sample. Therefore, the measured birefringence angle without the THz pump can be characterised as complete magnetisation of the sample. When the sample is pumped, the birefringence changes linearly with the sample magnetisation, which can then be extracted accordingly. This experimental setup was also characterised in-depth in a previous work [27].

The THz source contains significant spectral components up towards ~18 THz (as characterised through electro-optical sampling of a GaP crystal and ABCD detection technique); these components were eliminated through a set of low pass filters with cut-off frequencies at 3, 6 and 9 THz. This results in a set of pulses that are well defined in spectral content; we expect these pulses to contain several oscillation pulses in the time domain. To calculate the pulse fluence, we refer to a previous work on the same THz source: [28].

In the cited text, a one-to-one correspondence between the pulse fluence and the peak electric field strength was found through the Kerr effect in diamond, allowing us to infer the fluence of a filtered pulse by measuring the pulse duration and spot size (measured previously in [29]).
Figure 1. (a) The experimental setup for THz pump and optical MOKE probe. (b) The triggering unfiltered THz pulse. (c) Fourier transform of the unfiltered THz pulse (d) Terahertz-induced demagnetization taken with 3 THz low pass filter and THz excitation levels of 4.3, 7.7, 11.2 & 17.3 mJ/cm². (e) The demagnetization measurements taken with 6 THz low pass filter for 8, 16.7, 24.5, 29, 33.3, 39, 47.3 & 66.8 mJ/cm². (f) The fluence-dependent demagnetization taken with 6 THz low pass filter for delay of 0, 10 and 200 ps, with a straight line as a guide for the eye to showcase the near linear response.

In order to decouple the charge, spin and phonon dynamics without a detailed analysis of their specific relationships with one another, we exploit the timescale differences between them. This was achieved by modulating the THz pump excitation and repeating the experiment in Figure 1 with static magnetic fields of $B_+ = +40$ mT and $B_- = -40$ mT, corresponding to biases which are parallel and antiparallel to the THz pulse magnetic field, thus maintaining a constant magnetic bias during our experiments.

3. Results

Through the 3 THz filter, the maximum achievable THz fluence on the sample is 17.3 mJ/cm², leading to an instantaneous demagnetization ($dM/M_s$) of 60%, as shown in Figure 1d, where the time axis of the signals was shifted such that maximum demagnetization is observed at 0, and the timescale of remagnetisation can be compared for all pulse fluences. Extending the spectrum with the 6 THz filter, presented in Figure 1e, allows for much higher peak fluence up towards 66.8 mJ/cm² (equating to a peak incident magnetic field of 3.7 T in free space). This offers the possibility of complete demagnetization without sample damage at 47 mJ/cm², as alluded to in Figure 1f, where magnetisation is shown to recover. Measurements of the reflected pulse show that around 90% of the pulse fluence was reflected. This suggests that roughly 10% of the incident fluence is absorbed. Therefore, this corresponds to an absorbed fluence of $\sim$4.7 mJ/cm².

At low fluence, the extent of demagnetization increases with the excitation fluence almost linearly, suggesting a linear THz absorption mechanism in Figure 1f.
The dynamics under the different biases, characterised as the symmetric and antisymmetric components of the signal, are shown in Figure 2a for an excitation fluence of 190 mJ/cm². Individually, these dynamics contain all three degrees of freedom mentioned. If the signals under opposite magnetic biases are summed, we receive a bias-averaged total signal with features that do not depend on (the initial magnetisation state, and hence) the magnetic response of the sample. Consequently, the difference of opposite magnetic bias signals would give only the magnetic response of the material. Hence, the magnetic spin dynamics response (Figure 2b) could be separated from the charge and lattice degrees of freedom (shown in Figure 2c). The lattice and charge dynamics were further separated in Figure 2d by fitting Figure 2b to Figure 2c, with an overall amplitude variable, and calculating the difference.

![Figure 2](image)

**Figure 2.** (a) The THz-induced birefringence in Ni using an external mechanical chopper under and external magnetic bias B+ & B− with a fluence of 190 mJ/cm². (b) The extracted magnetic response (demagnetization) obtained from B+−B−. Blue, green, black, and magenta refer to the fluences of 47, 85, 123, & 190 mJ/cm². (c) the total electronic and structural response is obtained using (B+) + (B−) which contains both thermal excitation and acoustic phonons. The latter is independently shown in (d).

### 4. Discussion

We perform a rough calculation to compare the sample temperature at various fluences with the Curie temperature of Ni. The heat equation of the system is: \[ Q = m \times c \times dt, \] where \( m \) is the mass of the object, \( c \) is the heat capacity, and \( dt \) is the induced temperature change. Bulk nickel has a heat capacity of 0.44 J/gK and a density of 8.908 × 10⁶ g/m³. In our experiments, the sample thickness is 15 nm and we assume that about 10% of the incident THz pulse is absorbed by the sample. Using a low pass filter of 6 THz, we observe partial demagnetization at a fluence of 16.7 mJ/cm², corresponding to a temperature increase of
280 K. Complete demagnetization was observed at 47.3 mJ/cm\(^2\), corresponding to a 800 K temperature increase. Taking into account that the experiment was performed at room temperature, this gives us the temperature range of 500 K to 1070 K, which is comparable to the Curie temperature of Ni at 627 K.

This identifies that the demagnetization observed at very large fluence is due to the existence of hot electrons at the Fermi level (whereby electrons are excited by the intense sub ps heating out of the Fermi level. These electrons are prevented from thermalising with the Fermi sea due to the non equilibrium nature of the system, and therefore dominates the subsequent dynamics [30,31]). This is similar to what has been previously reported for optical laser sources, but has not been proven previously for THz sources.

Previous results on a similar sample [2] showed a maximum demagnetization of 58% at 89 mJ/cm\(^2\) before the sample becomes permanently damaged. This is in contradiction to the results we present here, where higher demagnetization was achieved at lower THz fluence without noticeable damage. We offer an explanation for this discrepancy: The MOKE probe in the previous report likely lacked the spatial resolution to pinpoint the demagnetised area exactly. Hence, both the areas of demagnetization and the areas that are unaffected contributed to the measurement, giving a lower demagnetization percentage. In this paper, we limit the spectral contents to <6 THz; this increases the diffraction-limited excitation spot size. This allows the area of demagnetization to be fully resolved, and we therefore achieved higher measurable demagnetization at lower fluence.

We use this quantitative result to address the main question of the role of photons in demagnetization; specifically, whether demagnetization was achieved through the direct or indirect channel discussed above. Conventionally, it was assumed that the direct mechanism was associated with excitations of low frequency electro-magnetic radiations, such as microwave radiation. In contrast, high frequency optical pulse excitations have been shown to demagnetise materials through the indirect mechanism dominantly. However, it is not impossible for both mechanisms to coexist.

It is generally considered that the higher the excitation pulse frequency, the higher the excitation field amplitude is needed in order to induce changes in the magnetisation. Therefore, to achieve magnetic switching in the THz regime, it would require practically unrealistic pulse intensities (>10 Tesla). In this limit, undesired structural changes would inevitably take place. Our experiments show that THz excitations are only capable of inducing small amplitude precessions, as is confirmed in Figure 1, where no precessions are observed in the measured signal. Therefore, our results point towards the indirect mechanism as the most likely hypothesis.

A lingering question remains: Do individual photon energies play a significant role towards the measured demagnetization effect? Indeed, this indirect mechanism critically depends on the presence of a high integrated photon energy to raise electron temperatures above phonon temperatures. Comparing the photon energies from the optical (eV energy scale) range to the THz (meV energy scale) range, we conclude that this effect is not directly possible without exponentially increasing the number of THz photons.

In contrast to this claim, the experimental results do not reveal a relationship between individual photon energy and demagnetization. First, there is no high fluence threshold for demagnetization; A nearly linear relationship between fluence and demagnetization is observed in our experiments (Figure 1f). Secondly, with comparisons between our results and a previous report on a similar sample under optical excitation [2], the absorbed fluence needed for demagnetization from optical and terahertz pulses are of the same order of magnitude. The differences in energy between individual optical and THz photons are three orders of magnitude apart, and thus confirms a negligible dependence of demagnetization on individual excitation photon energy, ruling out any significant direct interaction of the THz electric field frequency on demagnetization in our experiments, with evidence suggesting that only the net fluence absorbed influences the demagnetization in the sample.

In light of these results, we extended our search of the mechanisms behind demagnetization by including charge and lattice degrees of freedom. The ultrafast excitation of thin
films leads to rapid heating and excitation of electrons to higher non-equilibrium states. This absorbed energy is eventually transferred to the lattice through electron-phonon interactions, leading to spatio-temporal strain pulses and coherent acoustic phonons which reverberate inside the film. This model has been used to explain ultrafast optical generation of lattice strain waves of coherent acoustic phonons, which can manipulate and coherently control the magnetisation orientation in ferromagnetic films [8,11–13], thereby justifying our investigation into these additional dynamics.

The spin dynamics (as shown in Figure 2b) exhibit a rapid $\sim 1$ ps response, which decays over time, concurring with the results shown in Figure 1. Additionally, the charge-lattice-combined response (Figure 2c) is characterised by a sharp transient, followed by a slow relaxation with a fast damping oscillations. Here, the oscillations represent the coherent acoustic phonons and are independently extracted in Figure 2d, where $\sim$ ps to a few ps oscillations are observed.

To further justify the significance of these additional dynamics, the constant bias experiments was performed again at excitation fluence of 48 mJ/cm$^2$ and 85 mJ/cm$^2$ as shown in Figure 3a,c. Where the isolated spin and charge dynamics, corresponding to the magnetic and electronic responses are shown in Figure 3b,d. It is shown that the magnetic response lags behind the electronic response for all values of fluence. Furthermore, it is shown that a sub picosecond magnetic response exists from a THz pulse excitation.

![Figure 3](image)

**Figure 3.** (a) The measured Kerr signal ($B^+$ and $B^-$) under a 9 THz filtered fluence of 48 mJ/cm$^2$. (b) The extracted magnetic ($B^+ - B^-$) and electronic ($B^+ + B^-$) responses. (c,d) are the corresponding measurements to (a,b) under a fluence of 85 mJ/cm$^2$.

5. Conclusions

In the present paper, the physical mechanisms behind nickel thin film demagnetization due to femtosecond laser pulses are investigated. At low field amplitudes, we observe a linear response of demagnetization from the intensity of the pump electromagnetic field. The THz pump fluence needed to achieve full demagnetization is similar to the optical counterpart, and therefore suggesting that the frequency of the pump pulse does not play a major role in the demagnetization process. To support this argument, it was found that at
full demagnetization, the THz pulse induces heating within the sample that is comparable to the Curie temperature of nickel. Alluding to sample heating as the main cause of the demagnetization process.

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