

Monitoring laser-induced magnetization in FeRh by transient terahertz emission spectroscopy ^{EP}

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ABSTRACT

In this study, a conceptually different approach for investigating magnetic phase transitions in ultra-thin films is presented. THz emission from a laser-excited material is used to monitor the magnetization dynamics during the laser-driven antiferromagnetic to ferromagnetic transition in FeRh. The emitted THz signal is calibrated against static magnetometry data measurements, giving a direct measure of the absolute magnetic moment of the sample on the sub-picosecond timescale. The technique is, therefore, highly complementary to conventional time-resolved experiments such as time resolved magneto-optic Kerr effect (MOKE) or x-ray magnetic circular dichroism.

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The first observation of ultrafast demagnetization on the sub-picosecond timescale¹ led to intensive research in the field of ultrafast magnetization dynamics because of its prospective applications in spintronics. However, despite more than 20 years of effort, many mysteries remain in ultrafast magnetism, and so there is a clear need for alternative approaches. Here, we present a conceptually different technique to probe ultrafast magnetization dynamics to complement those in frequent use.

Pump-probe techniques are commonly employed to study ultrafast magnetization dynamics. The magneto-optical Faraday and Kerr effects are widely used and extremely powerful tools to study magnetization dynamics,^{2–4} but both techniques have limitations arising from the complex nature of the detected magneto-optic signals^{5,6} (e.g., coherent artifacts^{7–9} arising from the overlap of the pump and probe laser pulses). Another common approach is time-resolved x-ray

magnetic circular dichroism (XMCD). It is a local and element-specific probe, providing dynamical information at key absorption edges and has been used to disentangle, for example, the dynamics of spin and orbital angular momentum,¹⁰ but it needs large x-ray facilities.

The scheme presented in this Letter employs laboratory-based THz emission spectroscopy as a probe, which allows us to follow changes in the magnetic structure of thin magnetic films with sub-picosecond time resolution. We will show that THz emission spectroscopy is a powerful complementary technique to study magnetic phase transitions. There are several mechanisms in magnetic film samples that can contribute to the emission of a THz pulse.¹¹ In the pioneering work by Beaupaire *et al.*,¹² THz generation through the ultrafast demagnetization process of ferromagnetic Ni films was observed. This was confirmed by later work of Jian *et al.*,¹³ who demonstrated that THz emission from Fe–Ni alloys could be attributed to the

demagnetization process and that it carries information on the timescales of the energy transfer between the electron, spin, and phonon subsystems. Recently, it was shown that the THz emission can be enhanced through spin current diffusion in Fe and CoFeB hetero-structures.^{14,15} When materials possess magnetic resonances within the excitation bandwidth, terahertz emission spectroscopy can also be applied to observe the dynamics of those antiferromagnetic (AFM) and ferromagnetic (FM) modes.^{16,17} Here, we experimentally demonstrate that using a double pump technique, we can study the in-plane magnetization dynamics of ultrathin magnetic films without restriction on longer time scales that a single pump pulse entails. Furthermore, we have calibrated the THz emission against static magnetometry data (SQUID) from the same sample, allowing us to measure the absolute value of magnetization with sub-picosecond time resolution. As a proof of principle, we studied the laser-assisted magnetization dynamics of an iron-rhodium (FeRh) thin film.

Stoichiometric FeRh exhibits a first order phase transition from antiferromagnetic (AFM) to ferromagnetic (FM) spin order at approximately 370 K. The transition temperature depends on the exact composition and growth procedure of the samples.^{18,19} The phase transition can be driven by temperature or magnetic field and is associated with a lattice expansion, making it a favorable candidate for many spintronics applications.^{20–24} Laser induced, time-resolved MOKE experiments on FeRh suggested that the AFM to FM transition takes place on timescales of a few hundred femtoseconds followed by a phonon-driven lattice expansion over several picoseconds (ps).^{25,26} Bergman *et al.* have proposed a mechanism of the phase transition dynamics based on the nucleation and growth of microscopic magnetic domains.²⁷ The nucleation and growth of the microscopic magnetic domains and the coexistence of the AFM and FM phases have been imaged using photoemission electron microscopy (PEEM) imaging.²⁸ A time-resolved (tr) XMCD study by Radu *et al.*^{29,30} reported that the generation of ferromagnetic ordering for both Fe and Rh occurs on a timescale of several picoseconds. The discrepancy of the timescale observed in the AFM to FM phase transition between tr-MOKE and tr-XMCD measurements is still debated.

In this article, we present results on the laser-assisted AFM to FM phase transition as well as ultrafast demagnetization of the FM phase in FeRh thin films. Our results show that the induced magnetization takes place over several tens of picoseconds, which is in accordance with observations by time-resolved XMCD. A stoichiometric FeRh film of 35 nm was grown epitaxially onto (001) textured MgO on 20 nm-thick Si₃N₄ on a Si substrate. The details on the FeRh sample growth can be found in Ref. 31 and references therein. Samples were initially characterized by temperature-dependent magnetic small angle x-ray scattering, as well as static and tr-MOKE.

The AFM-FM phase transition and magnetization dynamics were studied using an optical pump–pump–probe technique. The experimental scheme is shown in Fig. 1(a). We used a laser system operating at a 1 kHz repetition rate with a 100 fs pulse duration at an 800 nm central wavelength. Beam splitters (BS1 and BS2) separate two pump beams, which were later recoupled using a polarizing beam splitter (PP). After the polarizing beam splitter, both pump beams have orthogonal polarization and passed through a 2 mm hole in a parabolic mirror to irradiate the sample at normal incidence. The first pump (reflected from BS1) is used to initiate the magnetization dynamics, while the second one is used to generate THz radiation from the sample. In order to avoid measurements of any THz

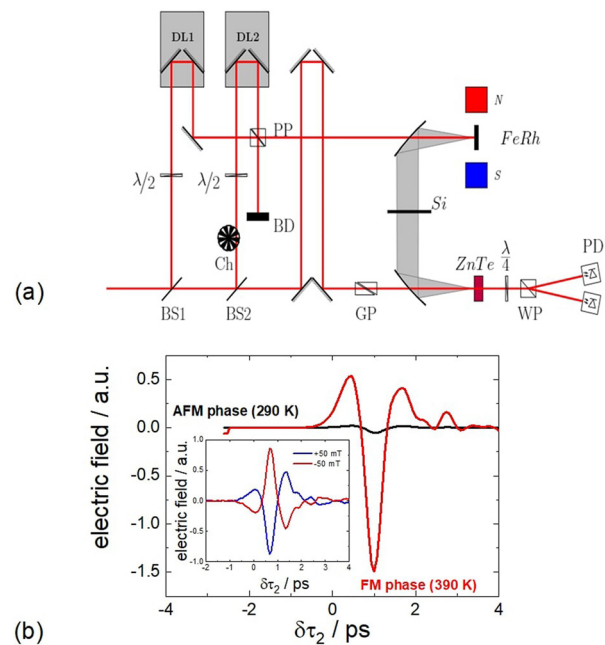


FIG. 1. (a) Experimental setup for THz emission spectroscopy. (b) THz pulses emitted after laser excitation at 290 K (AFM phase) and at 390 K (FM phase). The inset shows the polarity reversal of the THz electric field by the direction of the weak in-plane magnetic field.

radiation emitted because of the first pump pulse, we modulated the second pump with a mechanical chopper and employed heterodyne (lock-in) detection at the modulated frequency. THz radiation generated in the backward direction was refocused on a 2 mm thick ZnTe (110) crystal, and standard electro-optical (EO) sampling was performed. The thin film of FeRh was placed in a permanent magnetic field of 50 mT. The sample was mounted on a heater, which allows changing its temperature from 290 K to 400 K.

To calibrate our technique to the absolute magnetic moment of the sample, we performed temperature dependent measurements with THz emission and, separately, using a commercial magnetometer on the same sample. The first pump beam (pump-1) was blocked, while the fluence of the second pump beam (pump-2, termed as $\delta\tau_2$) is set to ~ 10 mJ/cm². As can be seen in Fig. 1(b), at room temperature (AFM phase), we observe comparably weak broadband THz emission from the sample, which is likely coming from an interfacial FM state.³² We then heated the sample to 390 K, which is above the AFM-FM phase transition temperature. At 390 K, we observed strong THz emission [Fig. 1(b)] roughly 50 times higher in field amplitude than the emission observed at 290 K. The emitted THz radiation has linear polarization, orthogonal to the applied in-plane magnetic field direction. The Fourier transform of the emitted THz radiation exhibits a nearly identical shape for both FM and AFM phases (not shown here), and the frequency spectrum is limited by the pulse duration of the probe pulses and the thickness of the EO detector. The polarity of the emitted THz radiation reverses by reversing the applied in-plane magnetic field, confirming the FM state of the FeRh [see the inset of Fig. 1(b)].

The magnetic moment of the studied FeRh thin film was measured using a commercial SQUID magnetometer as a function of

temperature. We observed the typical hysteretic behavior of the magnetization within the temperature sweep, as displayed in Fig. 2(a) as a black line, clearly indicating the AFM to FM phase transition. It can be seen that the film transforms to the FM phase completely at ~ 380 K. We also plot the peak of the emitted THz radiation from the same thin film at various temperatures upon heating and cooling from 300 K to 390 K with an excitation pump fluence of $40 \mu\text{J}/\text{cm}^2$. The data are shown as blue dots in Fig. 2(a). The normalization is done with respect to the maximum electric field of the emitted THz radiation in the full FM phase. We see qualitatively good agreement between static magnetometry and THz emission spectroscopy data as the latter shows the same shape and width as the static data with only

a slight shift on the temperature scale. This allows us to convert the THz amplitude to the magnetic moment of the sample under investigation. Small differences between the two datasets can be attributed to the heating of the sample, as discussed below.

It is critical to this approach that the THz generation pump pulse does not affect the magnetism of the sample. Stroboscopic pump-probe experiments require the sample to return to the same starting condition between cycles. This does not necessarily occur in systems that exhibit hysteresis, where several states can be stable at the same parameters. For this reason, for example, it is complicated to observe temperature hysteresis loops (two stable phases—AFM and FM are stable at the same temperatures inside the loop) using the pump-probe approach.³³ Here, we experimentally demonstrate that using excitation pulses with appropriate energies, we can map the dynamics of both stable phases and observe the hysteresis loop with a pump-probe approach. To find suitable pulse parameters, we recorded hysteresis loops for FeRh thin films using THz emission spectroscopy at various pump fluences, see Fig. 2(b). The fluence dependence of the hysteresis loop shows that for fluences between $0.04 \text{ mJ}/\text{cm}^2$ and $1.2 \text{ mJ}/\text{cm}^2$, the hysteresis width decreases, i.e., the apparent AFM-FM transition temperature becomes lower for higher fluences. At fluences of $6 \text{ mJ}/\text{cm}^2$ and above, there is a complete collapse of the loop along with an overall shift of the curve to apparently lower temperatures. These two effects on the hysteresis loops induced by the pump excitation can be explained as follows: we start with the sample in the AFM phase at room temperature and increase the equilibrium temperature to point (1) on the hysteresis curve in Fig. 2(c). A weak excitation pulse (shown as red circle) will heat the sample only slightly, but it will not visibly change the observed hysteresis, and the sample spot under irradiation will cool down before the next pulse arrives at the sample 1 ms later. This is symbolized by the red dashed loop. As we increase the pump fluence, the excitation pulse becomes strong enough to drive the sample into the FM phase, as shown by the black arrow emanating from point 1. The sample will then cool back to the equilibrium temperature of the sample holder; however, it will now remain in the stable FM phase (point 2). All subsequent excitation pulses will find the sample in the FM state, and this results in THz emission that is characteristic of the FM phase. With every excitation pulse, the irradiated spot will go through a heating-cooling-loop that is entirely located within the FM phase, as symbolized by the black dashed loop. Eventually, a reduction of the apparent equilibrium temperature of the AFM-FM transition is observed. Like Maat *et al.*,³⁴ we also observed irreversible remanence magnetization dynamics during the heating cycle, but to distinguish the laser-induced heat effects from the intrinsic nature of the irreversible AFM-FM transition process requires further investigations. As we increase the fluence of the excitation pulses further, this apparent temperature becomes lower and lower and the hysteresis width is further reduced until it completely collapses. At high enough fluences ($6 \text{ mJ}/\text{cm}^2$ and above), the average heating of the sample starts to induce a second effect: the collapsed hysteresis curve moves to a lower apparent temperature. In this case, the irradiated spot is at a higher actual temperature than the sample holder, which reduces the apparent FM-AFM transition temperature. Our fluence dependence experiments and the comparison with SQUID measurements clearly show that the hysteresis loop recorded at the fluence of $40 \mu\text{J}/\text{cm}^2$ indicates negligible average and dynamical heating due to excitation pump pulses and we use this fluence for the remaining studies.

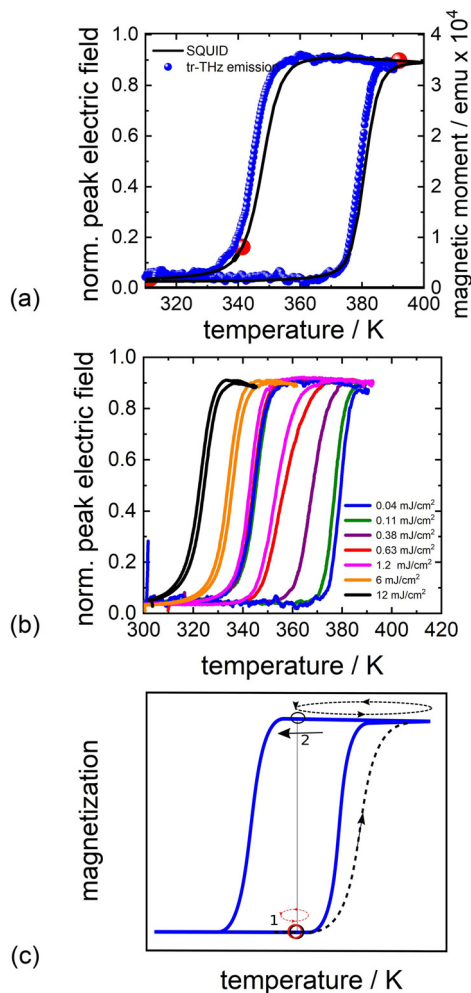


FIG. 2. (a) Temperature dependence of the peak field amplitude of the THz electric field [the maximum modulus of the field measurement displayed in Fig. 1(b)] compared to a conventional SQUID measurement. (b) Fluence dependence of thermal hysteresis loops recorded using THz emission spectroscopy. The temperature scale is for the sample holder, not the sample volume excited by laser, which can be higher. (c) Schematic of the effect of the excitation pump fluence on the hysteresis loop. Points 1 and 2 show the two stable states of the sample at a given temperature.

After calibrating the THz emission to the magnetic moment of the sample in non-perturbative pump energy, we now proceed to examine the ultrafast magnetization dynamics of the FeRh using the pump-pump-probe setup at room temperature. Irradiation of the FeRh thin film with pump-1 at a fluence of 10 mJ/cm^2 initiates the AFM-FM phase transition. It is probed by measuring the emitted radiation peak field initiated by pump-2, which was kept at the delay position where we observe a maximum amplitude of the emitted THz radiation, i.e., approximately 500 fs, with a fluence of $40 \mu\text{J/cm}^2$. Then, we vary the pump-1 delay with respect to pump-2, termed as $\delta\tau_1$ in Fig. 3. The lower fluence of pump-2 ensures that the dynamics of the thin film is defined by pump-1 solely. In Fig. 3(a), we show the magnetization dynamics induced in the FeRh thin film at opposite polarities of the applied in-plane magnetic field. The normalization is done with respect to the maximum electric field of the emitted THz radiation from the FM state of the thin film, which is 40% of the FM phase achieved by 10 mJ/cm^2 excitation pulses over the 300 ps timescale.

Next, we studied the fluence dependence of the laser induced AFM-FM phase transition in FeRh thin films at room temperature. The fluence of pump-1 was varied between 1 mJ/cm^2 and 10 mJ/cm^2 and used to drive the AFM-FM phase transition. This FM ordering is probed using THz emission spectroscopy by making use of pump-2 at a constant fluence. As can be seen in Fig. 3(b), we observe no THz emission from the FM phase at a pump fluence of 1 mJ/cm^2 . This fluence is not sufficient to induce any magnetization in the FeRh thin film at room temperature. At 5 mJ/cm^2 , we observe THz emission

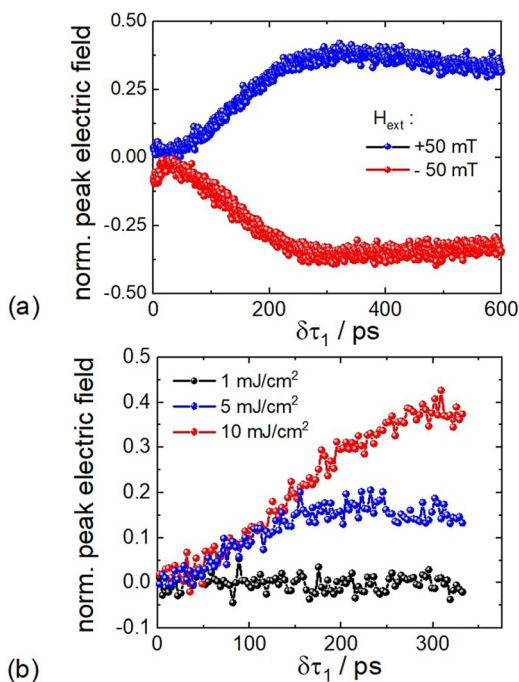


FIG. 3. (a) The peak field amplitude of the THz electric field generated by pump-2 pulse evolution as an indicator for the in-plane magnetization dynamics initiated by the first pump pulse. (b) Transient THz emission spectroscopy measurements of the laser-driven AFM to FM phase transition in FeRh at three different fluences of pump-1.

from the formation of magnetic order in the FM phase on the timescale of hundreds of picoseconds. The amount of magnetization (FM) reaches a maximum in the 200 ps timescale for 5 mJ/cm^2 , whereas at 10 mJ/cm^2 , the induced magnetization in the FM phase is still increasing on the scale of 300 ps. This shows that there is a threshold behavior for the induced magnetization with respect to the excitation pump fluence, consistent with the finding of Radu *et al.*²⁹ A fluence of 1 mJ/cm^2 (black) is below threshold, 5 mJ/cm^2 is near threshold (blue), and 10 mJ/cm^2 is above threshold (red).

The THz emission probe technique is not limited to the AFM-FM phase transition but applies equally to the more commonly studied (de)magnetization dynamics. In the following, we use it to study laser-driven demagnetization of the same FeRh sample from the FM phase. We performed a similar experiment to that shown in Fig. 3 but at various temperatures of the FeRh thin films. The first pump pulses have a fluence of 1 mJ/cm^2 . The THz emission pulse (pump-2) was kept at $40 \mu\text{J/cm}^2$. In Fig. 4, we plot the demagnetization dynamics of the FeRh thin film at three different temperatures, namely, 300 K, 340 K, and 390 K [which are shown in Fig. 2(a) as red dots]. At 300 K, the sample is in its AFM phase and 1 mJ/cm^2 optical excitation is not enough to excite magnetization within a timescale of a few tens of picoseconds. In the case of 340 K, the sample was at first heated up to 400 K and slowly cooled down to 340 K. The sample is, thus, partially magnetized (20% of saturation magnetization, being in the middle of FM to AFM transition). For this case, laser excitation induces ultrafast demagnetization, and the magnetization drops within 100 fs to 10% and then slowly recovers back to 20% of the original magnetization. For 390 K, the sample is in the FM phase and laser excitation induces 30% demagnetization. The observed timescale of the demagnetization process of 100 fs is limited by the pulse duration of the laser. These results are in agreement with XMCD measurements²⁹ and show that the induced magnetization in a laser-driven AFM-FM phase transition occurs on a timescale of hundreds of picosecond, while demagnetization can occur on sub-ps timescales.^{29,35}

In conclusion, we have shown that THz emission spectroscopy is a powerful and complementary tool to study magnetization dynamics at ultrafast timescales. The AFM-FM phase transition of an FeRh thin film was studied using THz emission spectroscopy as a function of

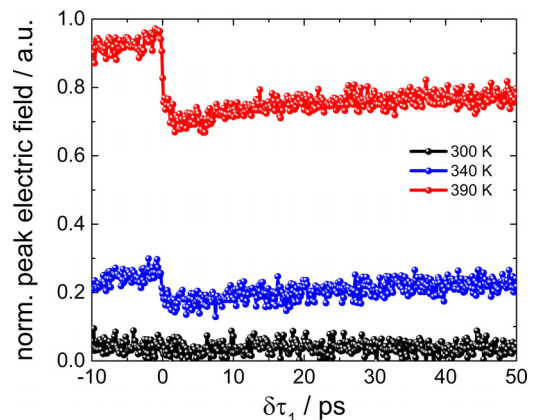


FIG. 4. Laser induced demagnetization at three different temperatures: 390 K (FM phase), 340 K (transition between FM and AFM), and 300 K (AFM phase).

temperature and calibrated against a commercial magnetometer, giving direct access to the sample's magnetic moment on ultrafast timescales. THz emission spectroscopy can measure magnetization dynamics averaged over the illuminated area and projected parallel to the external magnetic field.³⁶ Using 800 nm pump pulses, the illuminated area can be as small as 400 nm as defined by the diffraction limit. This limit can be surpassed by employing tip enhancement techniques, e.g., near field emission microscopy,³⁷ which should provide a spatial resolution of about 100 nm. The typical penetration depth of light into a metal is approximately a few 10 nm, which along with the lateral resolution limit defines a volume resolution for thick samples. We have applied the calibrated THz emission experiment to the laser-induced AFM-FM and demagnetization of the FM phase. This study confirms that the induced magnetization in the FM phase in FeRh takes place over hundreds of picoseconds and demagnetization in less than 100 fs. Our results agree well with tr-XMCD, but in contrast to tr-MOKE, we do not observe any changes associated with the AFM-FM phase on the timescales of tens of femtoseconds.

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DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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