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Selective THz control of magnetic order: new opportunities from superradiant undulator sources

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Abstract

Recent advancements of accelerator technology enable the generation of carrier-envelope-phase stable THz pulses with high fields at adjustable high repetition rates. The appropriate choice of THz radiator allows generation of narrow-band, spectrally dense, multicycle THz transients of tunable THz frequency which are ideally suited to selectively excite low-energy excitations such as magnons or phonons. They also allow one to study the frequency dependence of nonresonant THz-field interactions with various order parameters with high dynamic range. In this paper, we discuss the future prospects of this new type of THz light source for studying the coherent control of magnetic order based on recent results.

Keywords: terahertz, THz control, magnetic order, multicycle pulses

(Some figures may appear in colour only in the online journal)

Introduction

The control of magnetic order by intense transient THz fields is an emerging area of ultra-fast science which has been enabled by the advancements of THz generation techniques that meanwhile routinely provide THz fields in the few-10 to 1000 kV cm⁻¹ regime. High-field THz sources fall into two main classes: table-top sources based on down-conversion of

strong near-infrared femtosecond lasers [1] and superradiant THz sources based on ultra-short, highly charged electron bunches at modern linear accelerators [2–6].

Various studies have investigated the interaction between THz fields and magnetic order on sub-THz-cycle timescales and have demonstrated different mechanisms for THz control. For instance, THz-driven demagnetization by nonresonant ponderomotive acceleration of free or weakly bound electrons

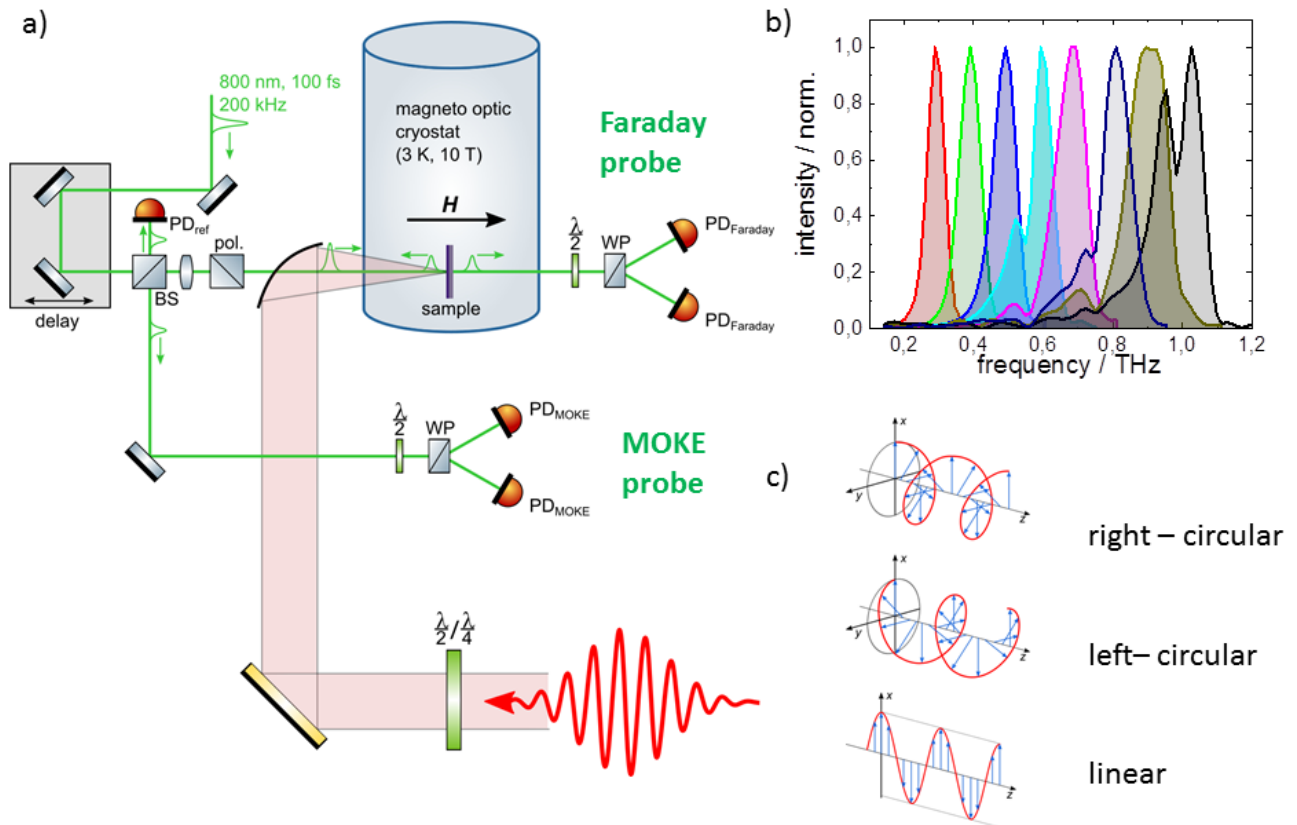


Figure 1. Currently available end station for THz control experiments of magnetic order at TELBE. (a) The measurements can be performed in a commercial 10 T split-coil-magnet system [20] providing for sample temperatures between 3 and 300 K and magnetic fields of up to 10 T. In the currently available end station, magnetization dynamics can routinely be probed on ultra-fast timescales with a synchronized femtosecond laser (green line) by transient MOKE and Faraday measurements with a temporal resolution in the few 10 fs regime [14]. Narrow-band, Fourier-limited carrier-envelope-phase (CEP) stable THz pulses are provided as pump pulses at a typical repetition rate of 100 kHz (red line), offering a spectral density that is (already during early stage operation) a factor of 30 higher than those available from laser-based, single-cycle THz pulses [6]. (b) THz spectra of different THz tunes of the TELBE undulator source. During early stage operation frequencies between 0.1–1.2 THz are available with pulse energies of up to 2 μJ . Once operational at its design parameters, THz fields in the MV cm^{-1} regime will become available. (c) Waveplates allow one to control the THz polarization state between circular and linear. This figure has been obtained by the author(s) from the Wikimedia website https://commons.wikimedia.org/wiki/File:Circular.Polarization.Circularly.Polarized.Light_Plane.wave_Left.Handed.svg, where it is stated to have been released into the public domain. It is included within this article on that basis.

via the electric-field component has been observed in conducting samples [7]. Selective and direct interaction of the spin system with the transient magnetic field component via the Zeeman torque has been demonstrated in antiferromagnetic films [6, 8, 9]. More recently, selective THz control of magnetic order via phonon pumping has been successfully exercised [10, 11] and the control of atomic-scale magnetic structure has been achieved via resonant excitation of a specific electro-magnon [12]. Most recently low-energy orbital transitions [13] have been utilized to drive large-angle excitations of magnetic lattices.

Many of these experiments benefit strongly from the uniquely high spectral densities of the narrow-band tunable THz pulses that have become available from superradiant undulator THz sources at modern linear accelerators. THz pulses with pulse energies up to the few-100 μJ regime and a relative spectral bandwidth between 10 and 20% can be generated yielding spectral densities that are orders of magnitude beyond that available from laser-based THz sources [5, 6]. In this article, we present selected results from the

first year of early-stage user operation of the TELBE THz facility [6] that demonstrate the capabilities of this new type of large-scale user facility for studying selective THz control phenomena.

Methods

TELBE currently produces multi-cycle THz pulses at a maximum repetition rate of 100 kHz, which are tunable between 0.1 and 1.2 THz with pulse energies of up to 2 μJ while operation at its design parameters will provide pulse energies of up to 100 μJ in a frequency range up to 3 THz [6] (see figure 1). The generated THz pulses consist of eight cycles and yield a spectral bandwidth of roughly 20%. The polarization is linear but can be controlled between circular, elliptical and linear by means of appropriate quarter- and half- waveplates. Transient Faraday and MOKE measurements have been established as standard ultra-fast probes for the THz-driven magnetization dynamics. A novel pulse-resolved data acquisition system enables us to achieve timing between the probe laser and THz

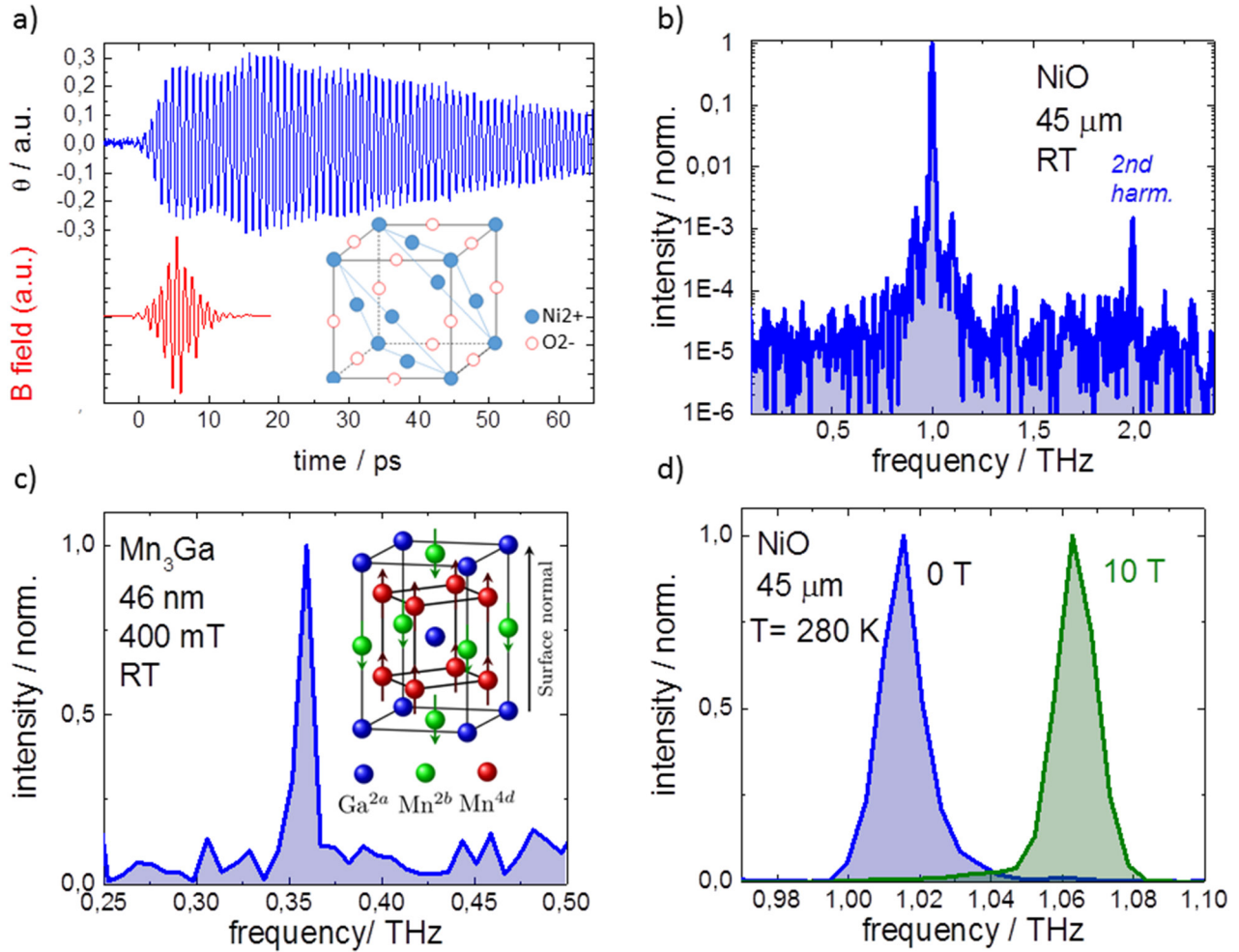


Figure 2. Probing resonant excitation of coherent spin waves by transient Faraday/MOKE spectroscopy (a) THz-driven spinwave experiment on a 45 μm NiO film, where the dynamics is probed by transient Faraday measurements with a 800 nm fs laser. The frequency of the pulsed THz magnetic field (red) can be tuned into resonance with the antiferromagnetic (AFM) mode of NiO at 1 THz yielding a spectral density that is by a factor of 30 higher than those available from laser-based single cycle THz pulses [6]. (b) Pulse-resolved detection in combination with the high spectral density and the high repetition rate of 100 kHz allows to achieve a dynamic range of better than 10^4 in routine measurements [14] which permits observation of the recently described emergence of higher-order features in the Faraday signal [9]. (c) The excellent dynamic range is important for the transfer of the concept of THz-driven spinwave excitations probed by time-resolved Faraday/MOKE measurements to thin films. As an example, the recent Faraday measurement of the coherently driven ferromagnetic (FM) resonance in a 46 nm thin Mn_3Ga film is shown. Reprinted from [18], with the permission of AIP Publishing. (d) The high dynamic range also aids experiments in extreme sample environments such as high magnetic fields and low temperatures. Shown here is an example of the effect of a 10 T magnetic field on the AFM resonance in NiO which is shifted to higher THz frequencies by ~ 0.06 THz [16].

pulses with a resolution of 12 fs (rms) and an exceptional dynamic range exceeding 10^6 in experiments that probe THz-driven dynamics in solids [14].

Results

Figure 2 shows experimental results of one typical class of early-stage experiments at TELBE. A multicycle THz pulse is tuned into resonance with specific magnetic modes and the coherent excitation is traced on sub-THz-cycle timescales by the transient Faraday rotation of a synchronized NIR fs laser. The first benchmark experiment, the results of which are shown in figures 2(a) and (b), has been performed on the prototypical antiferromagnet NiO [15].

The antiferromagnetic resonance in this material at a frequency of ~ 1 THz frequency has been utilized in the seminal

first demonstration experiments of THz coherent control of spinwaves using a laser-based THz radiation source [8]. Using the spectrally dense narrow-band TELBE THz pulses allows one to achieve a significantly larger spin deflection amplitude at similar pulse energies, since the energy is more efficiently directed selectively into the narrow resonance [6]. A dynamic range of better than 10^4 can be achieved easily on short measurement timescales. This permits the observation of only recently demonstrated higher-order effects such as the occurrence of a 2nd harmonic component of the 1 THz fundamental frequency in the Faraday response as demonstrated recently [9] with a superior signal-to-noise ratio. The increased sensitivity furthermore enables detection of such THz-driven spin excitations even in materials with small Verdet constants [17] or in ultra-thin films. An example is shown in figure 2(c), where the THz-driven ferromagnetic (FM) mode in an only 46 nm thick ferrimagnetic pseudo-Heusler-alloy Mn_3Ga film

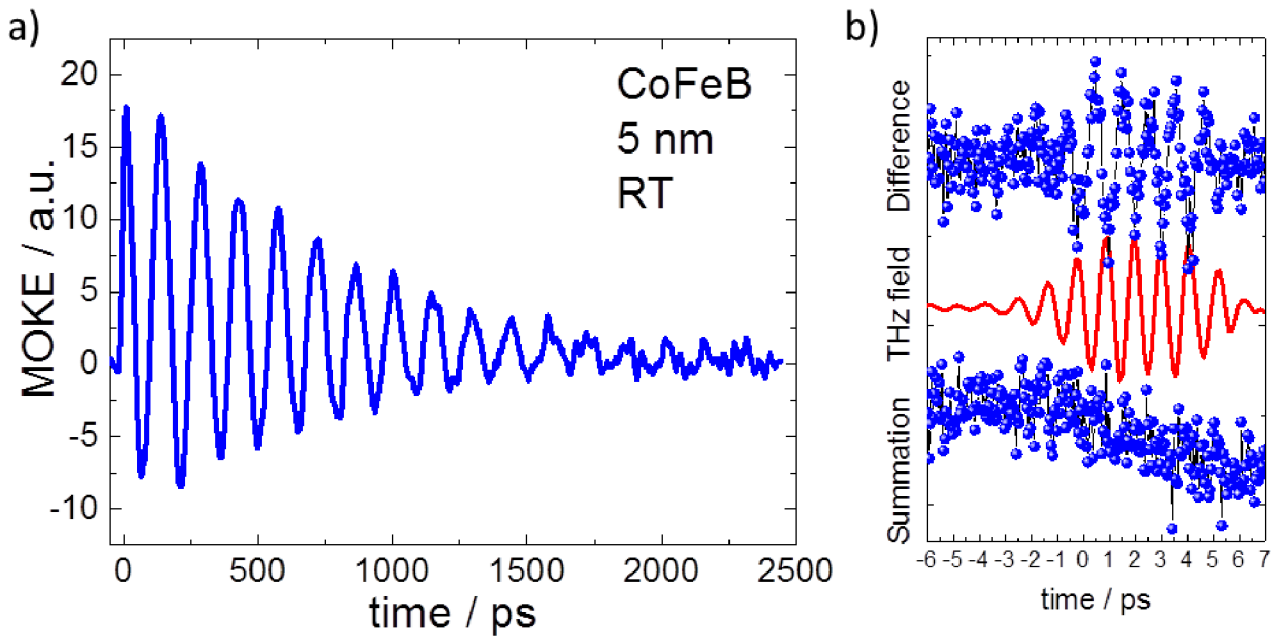


Figure 3. Probing non-resonant THz-driven demagnetization in ferromagnetic films by transient MOKE measurements. (a) Multicycle THz pulses drive the recently observed demagnetization in a 5 nm CoFeB film. The initiated demagnetization leads to the excitation of the ferromagnetic resonance on nanosecond timescales which can be followed at TELBE by shifting the electronic phase between the probe laser and the accelerator masterclock. (b) The onset of the demagnetization on ultra-fast timescales can be probed at few 10 fs timescales by measuring transient MOKE response at THz fields of opposite polarity (prepared by use of THz HWP's). Plotting the sum and difference of the derived response allows one to disentangle the fast coherent response following the driving THz field (Difference) and the picosecond-timescale noncoherent demagnetization response (Summation). The THz excitation pulse is also shown (red).

has been observed at a resonance frequency of 0.359 THz. In this experiment, another advantage of selective THz excitation becomes apparent: when comparing with concurrent laser-stimulated THz emission measurements, one finds that the observed resonance frequency is shifted noticeably by 0.09 THz to lower frequencies in comparison to the TELBE experiment [18]. Subsequent temperature dependence measurements of the THz emission process [19] show that this can be understood by an increase of the average sample temperature by 15 K due to the off-resonant laser excitation at 800 nm corresponding to 375 THz or 1.55 eV which is 3 orders of magnitude higher than the energy of the actual resonance. This type of residual excitation of the multiple energy degrees of freedom between the eV and meV range at once can be completely avoided when narrow-band THz pulses are used to exclusively address the FM resonance in Mn_3Ga at 0.36 THz (1.49 meV) as has been done in the TELBE experiment [18].

The end station at the TELBE facility has recently been equipped with a split-coil cryomagnet [20] that allows one to employ external magnetic fields of up to 10 T at temperatures between 3 and 300 K. Again utilizing NiO as a benchmark material, this enabled the observation of a clear shift of the AFM mode towards higher frequencies by 0.06 THz when applying a magnetic field of 10 T in the Faraday configuration (i.e. Hllk) [16]. To this end, the near-Gaussian-beam-like spatial properties of undulator radiation proves of high importance in keeping the inevitable transport losses of THz radiation into the 10 T split-coil magnet at minimum and by allowing achievement of close-to-optimal focal spot sizes on the sample. In combination with the high repetition rate, this

enables a dynamic range of better than 10^3 . The decrease in dynamic range compared to ambient conditions is because of the loss in fluency/THz field due to the larger focal length enforced by the ~ 200 mm distance between the sample and the optical viewports of the magnet.

Another class of experiments that benefits from the narrow bandwidth and tunability are those investigating the frequency dependence of the efficiency of THz-driven phenomena. An important consequence of the interaction of strong THz transients with matter is the acceleration of free or weakly bound electrons by the transient THz electric field component. Here, the interaction mechanism is based on the ponderomotive force exerted by the THz field and is thus scaling up with $1/\nu^2$, where ν is the radiation frequency. This means that processes based on this interaction, e.g. the THz-driven demagnetization of metallic ferromagnetic thin films [7], should become significantly more efficient at lower THz frequencies. TELBE has meanwhile successfully been utilized to drive demagnetization in CoFeB thin films at different THz frequencies [21]. An example for excitation with a 1 THz pulse is shown in figure 3. The induced ultrafast demagnetization also leads to the subsequent coherent excitation of the FM resonance at a few GHz frequency which can be followed over ns time scales by electronically shifting the phase between the probe laser and the electron accelerator (shown in figure 3(a)). On ultra-fast timescales, the THz polarization control by half-waveplates allows one to perform measurements of the transient MOKE signal at exactly antiparallel THz magnetic field directions. This enables the detection of the actual demagnetization (quadratic with respect to the THz field) from the resulting summation signal (see figure 3(b)) [21], and to disentangle it

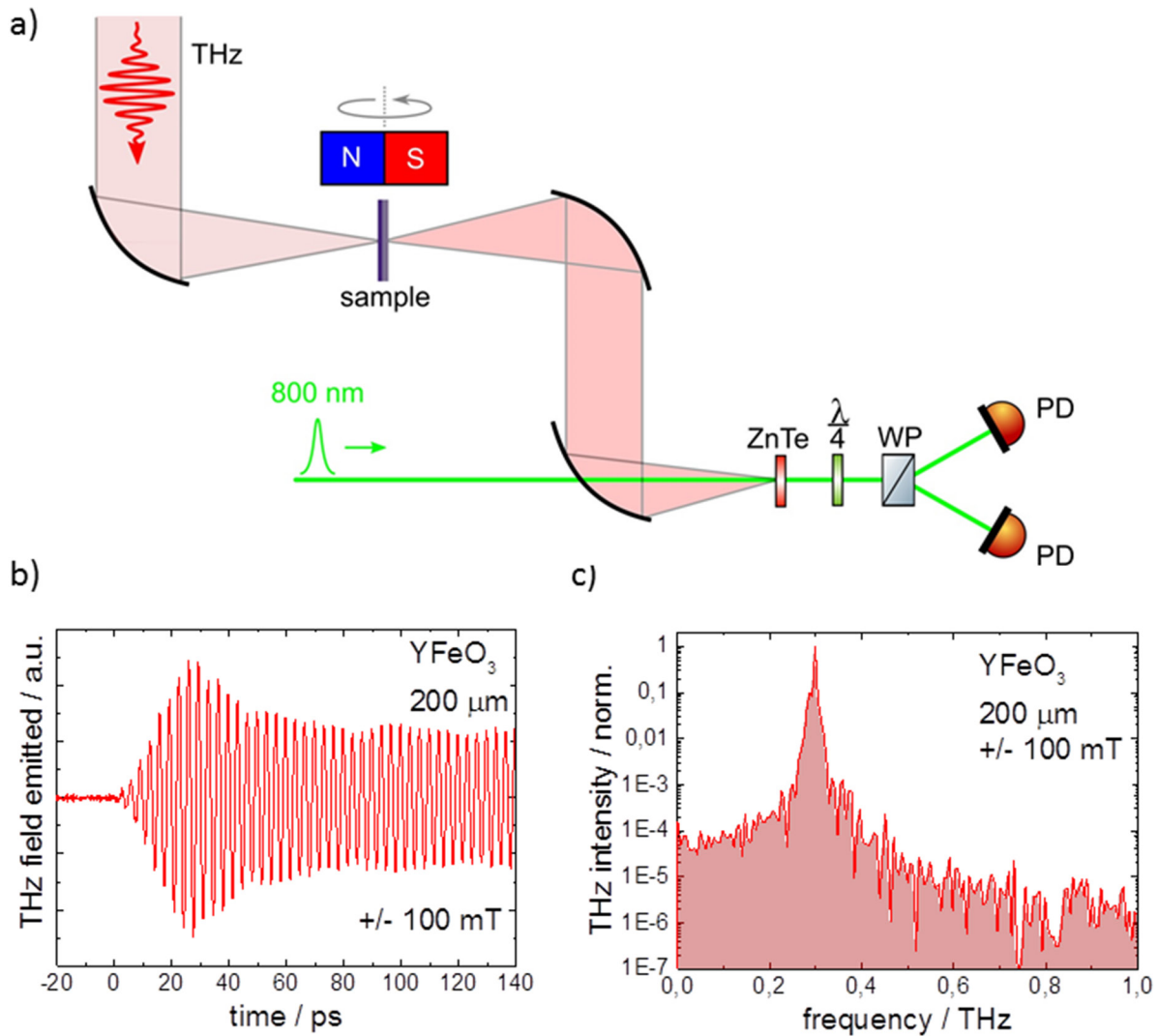


Figure 4. Selective driving and probing of a spinwave by THz emission spectroscopy: (a) experimental set-up for THz emission spectroscopy. The incident narrow-band spectrally dense THz pulses drive spin excitations coherently. An electromagnetic wave is re-emitted, with a field strength that is directly proportional to the 2nd derivative of the temporal magnetization change. The re-emitted field can be separated from the excitation pulse by subtraction of measurements for two opposite external magnetic fields. (b) Emitted THz transient of a resonantly driven FM spin excitation in YFeO₃ and (c) corresponding intensity spectrum. A dynamic range of better than 10⁶ has been achieved.

from the coherent precession (linear with the THz field) that can be isolated by taking the difference signal.

Discussion and conclusion

This paper demonstrates the potential of superradiant undulator facilities for research on magnetic materials that requires selective excitation with intense CEP stable, narrow-band THz light. Transient Faraday/MOKE spectroscopy has become the standard technique to probe selective THz control of magnetic order in the early-stage user experiments at TELBE. The reason for this is its comparatively straightforward implementation. The technique benefits from the high spectral density and the high repetition rate by achieving very high dynamic range. Thereby, THz-driven dynamics can be investigated in systems and in sample environments that are presently inaccessible by all-laser-based approaches. Another

easy-to-implement technique that benefits tremendously from the combination of high field and high repetition rate is THz emission spectroscopy (see figure 4).

Over the past 12 years several seminal experiments demonstrated that rapid changes of the magnetic state [22, 23], ultrafast spin currents [24] and, more recently, spinwave excitations [18, 25, 26] can lead to the emission of transient THz pulses. These pulses carry information on the spin dynamics involved (e.g. timescales or the nature of the magnetic resonances involved). Figures 4(b) and (c) shows a first example experiment of this type at TELBE where THz emission has been observed after selective excitation of the ferromagnetic mode at 0.3 THz in the model antiferromagnet YFeO₃ [25]. In this benchmark experiment, an excellent dynamic range of better than 10⁵ has been achieved. The ultimate advantage of this technique is that the only perturbation of the sample is the selective excitation of the specific low energy resonance of interest with the narrow-band THz excitation pulse. Parasitic

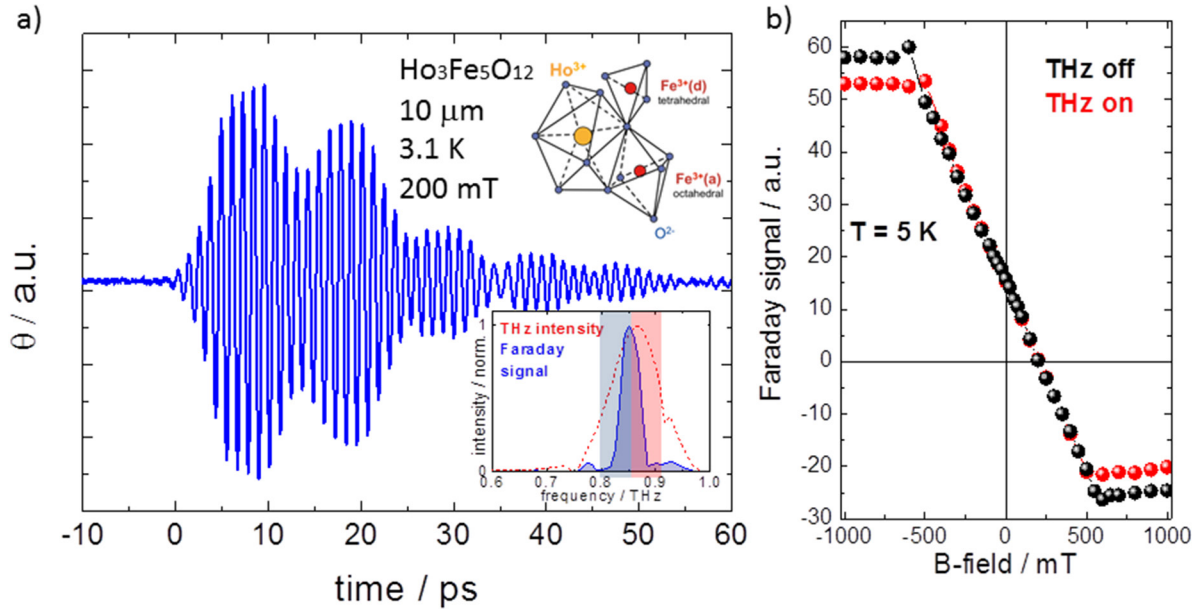


Figure 5. Effect of the average heating via direct THz lattice pumping in HoIG. (a) Transient Faraday signal after resonant THz excitation of 10 μm thick $\text{Ho}_3\text{Fe}_5\text{O}_{12}$ (HoIG) sample. (Inset) Spectrum of the THz excitation pulse (red) tuned in resonance with the optical magnon mode of HoIG (blue shaded area). The THz spectrum also overlaps with the Ho^{3+} crystal field resonance between 0.85 and 0.89 THz (red shaded). (b) A clearly observable temperature increase is quantified to be less than 15 K by measurement of the hysteresis with and without THz radiation impacting on the sample. The average THz power incident on the sample in this experiment was roughly 100 mW.

effects from additional probe light pulses are completely avoided. The dynamics (e.g. dephasing) of the coherently excited magnetic mode is followed by determination of the properties of the re-emitted THz wave.

Other versatile probe techniques for studying THz-driven changes of magnetic order that require more efforts in implementation are time-resolved scanning nearfield optical microscopy (TR—SNOM) [27, 28] and time, angle and spin-resolved photoelectron spectroscopy (TR—ARPES) [29]. These two techniques fully rely on the high repetition rate presently uniquely available from TELBE and shall enable probing of THz-driven dynamics on nanometer length scales and to follow the THz-field-induced changes directly in the electronic structure. End stations for both of these techniques are currently under development [30, 31].

Besides the advantages of the combination of high-field and high-repetition-rate THz sources one obvious obstacle arises when a significant part of the incident THz power leads to the subsequent heating of the sample. In the current early stage operation, TELBE provides average powers of 100 mW in the frequency range of 0.1–1.2 THz, but these powers will reach up to 10 W once the design parameters are achieved [6]. So far, samples that exhibited exclusively spin resonances in the addressed THz frequency range did not show a detectable increase of the average temperature, although the indication of magnon-lattice coupling was observed in the decay dynamics of THz-excited magnon in some experiments [26]. This has been different in cases where the spectral content of the THz excitation could directly couple to lattice degrees of freedom such as phonon modes [32]. In a recent study on THz-driven spin dynamics in holmium iron garnet (HoIG), the THz excitation pulse not only overlapped with a magnon

but also with a crystal-field resonance (see figure 5) [33]. In this case, a clear magnitude change of the hysteresis has been observed that could be associated with a temperature increase of ~ 15 K (see figure 5(b)), which appeared due to the low thermal conductivity of HoIG at the steady-state temperature of 5 K.

The motivation for the experiments shown in this paper stems mostly from an interest in: (i) background free spectroscopic measurements in the electronic ground state at high dynamic range (e.g. magnetic field and temperature dependence of resonances in NiO and YFeO_3 or the dependence of the FM resonance on the composition of Mn_{3-x}Ga nanofilms) or (ii) a better understanding of the underlying physical processes of recently discovered THz control phenomena like the demagnetization in CoFeB films [7] by determination of their frequency dependence. Furthermore, the dephasing of the coherent excitations can be followed and analyzed in the time-domain which allows to directly observe how and when the THz energy initially pumped selectively into a specific low energy mode such as an optical magnon is distributed to other degrees of freedom in the solid. Once TELBE can be operated with its design bunch charge of 1 nC, the pulse energies are expected to increase by 2 orders of magnitude (see figure 6) corresponding to field strengths $> 1 \text{ MV cm}^{-1}$ or few 100 mT. In this case, experiments will be able to explore selective THz control of magnetic degrees of freedom beyond the currently available perturbative regime and access intriguing phenomena such as THz-induced spin flip processes. Directing such exceptionally high fields selectively into narrow magnetic resonances will help to avoid recently discussed destructive effects of the THz electric fields [34] since off-resonant spectral components are kept at a minimum.

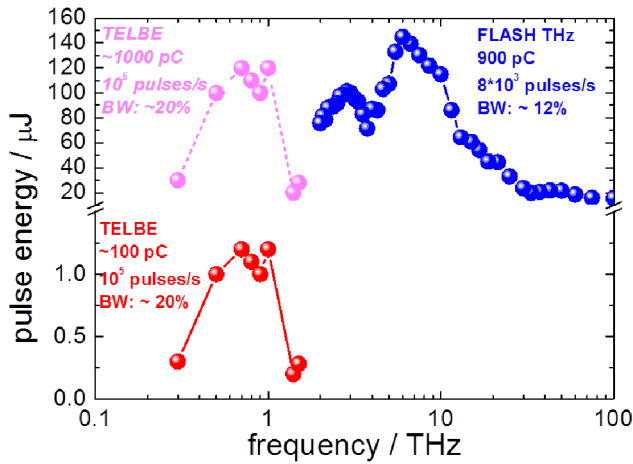


Figure 6. Pulse energies available between 0.1 and 100 THz from the two presently existing superradiant undulator based user facilities. The pulse energies scale quadratically with the bunch charge. Provided the electron bunch can be made short enough, this yields pulse energies in the 100 μJ regime for bunch charges in the 1 nC regime as already available with repetition rates up to 8000 pulses s^{-1} from the FLASH THz source [5] (blue). TELBE can currently be operated at a high repetition rate of 100 kHz but only with a moderate bunch charge of below 100 pC yielding pulse energies in the 1 μJ regime (red). After the planned upgrade, bunch charges of up to 1 nC should be available yielding pulse energies of up to 100 μJ in the frequency range between 0.2 and 1.2 THz. Reproduced from [6]. CC BY 4.0.

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