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Ultrafast and terahertz spintronics: Guest editorial

Special Collection: Ultrafast and Terahertz Spintronics

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ABSTRACT

Spin-based electronics (spintronics) aims at extending electronic functionalities, which rely on the electron charge as information carrier, by the spin of the electron. To make spintronics competitive and compatible with other information carriers like photons and electrons, their speed needs to be pushed to femtosecond time scales and, thus, terahertz frequencies. In ultrafast and terahertz spintronics, femtosecond optical and terahertz electromagnetic pulses are used to induce spin torque and spin transport and to monitor the subsequent time evolution. The two approaches, sometimes referred to as femto-magnetism and terahertz magnetism, have provided new, surprising, and relevant insight as well as applications for spintronics. Examples include the ultrafast optical switching of magnetic order and the generation of broadband terahertz electromagnetic fields. This APL Special Topic Collection is dedicated to provide a platform for the newest developments and future trends in the very active, dynamic, and exciting research field of ultrafast and terahertz spintronics.

I. INTRODUCTION

A. Spintronics

Spin-based electronics (spintronics) is a highly active subfield of magnetism and, more generally, solid-state research. It aims at extending electronic functionalities, which rely on the electron charge as information carrier, by the spin of the electron.[1]

Spintronics has large application potential in information storage and processing. Elementary spintronic operations (Fig. 1) include (a) control over magnetic order and, thus, over magnetically stored information by spin torque (Zeeman torque, spin-transfer torque, and spin-orbit torque) [Fig. 1(a)], (b) the transport of spin angular momentum in the form of spin-polarized currents [Fig. 1(b)] or spin waves (magnons), and (c) the detection of spin dynamics [Fig. 1(c)].[2,3] These operations have already been implemented in a commercial device, a magnetic random-access memory, with writing rates potentially reaching 1 GHz.[4]

B. Why ultrafast spintronics?

To make spintronic operations competitive and compatible with other information carriers like photons[5] and electrons,[6] their speed needs to be pushed to femtosecond time scales and, thus, terahertz frequencies. Working toward this goal comes with a number of exciting benefits.

First, one obtains a better understanding of fundamental magnetic and spintronic effects. Examples include spin-electron and spin-lattice coupling, magnon generation and relaxation, and the initial stage of transport phenomena, such as giant magnetoresistance (GMR),[7] tunneling magnetoresistance (TMR),[8] anisotropic magnetoresistance (AMR),[9] the spin Seebeck effect (SSE),[10,11] and spin pumping.[12,13] Second, the occurrence of new physical effects can be expected because the terahertz range overlaps with important excitations of magnetic solids.[14] Examples are the frequencies of phonons and magnons and the relaxation rates of electronic intraband transport. Third and finally, applications beyond spintronics, in fields such as ultrafast and terahertz photonics,[15] arise.

C. Implementation

To implement ultrafast spintronics, one needs to realize established spintronic operations, in particular those of Fig. 1, at femtosecond time scales. This goal requires the development of suitable

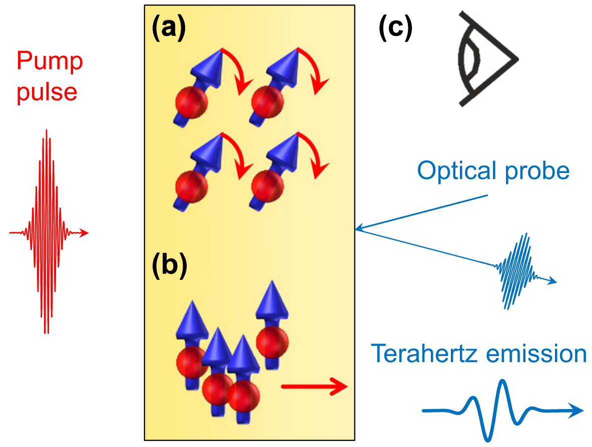

FIG. 1. Elementary spintronic operations. (a) Changing the orientation of spins requires torque, and (b) transport of spin angular momentum implies spin currents. (c) These processes need to be monitored. In ultrafast and terahertz spintronics, the operations (a)–(c) are implemented using femtosecond optical pulses (visible to infrared) and terahertz electromagnetic pulses (1–30 THz).

experimental stimuli and probes of ultrafast spin dynamics and of theoretical methods to model them.

Experimentally, femtosecond optical laser pulses and, more recently, terahertz electromagnetic pulses have been shown to be powerful tools to induce spin torque[16–21] [Fig. 1(a)] and spin transport[22–32] [Fig. 1(b)] and to monitor the subsequent time evolution [Fig. 1(c)]. The two approaches, sometimes referred to as femto-magnetism and terahertz magnetism, have provided relevant fundamental insight into spintronics and new surprising applications. Examples include the ultrafast optical switching of magnetic order[33,34] and the generation of broadband terahertz electromagnetic fields.[15,35–42]

Theory work is confronted with severe challenges because spontaneous magnetic ordering is a genuine many-electron phenomenon. Its description is largely based on model approaches, such as the Stoner and Heisenberg model.[43] In combination with ultrafast and highly nonequilibrium dynamics, the situation becomes even more complex. As a consequence, dedicated tools have been developed to address various phenomena like ultrafast demagnetization[33] and spin transport.[22]

D. This Special Topic

Ultrafast and terahertz spintronics is a highly active research field, which not only becomes clear from the numerous research works being published but also by concerted efforts like the Collaborative Research Center SFB-TRR 227 “Ultrafast Spin Dynamics” of the German Research Foundation, and the Actions “Ultrafast opto-magneto-electronics for non-dissipative information technology (MAGNETOFON)” and “Novel Spin-Based Building Blocks for Advanced Terahertz Applications (s-Nebula)” of the European Union.

As detailed below, the articles of this Special Topic address latest trends in ultrafast spintronics with regard to (1) understanding of fundamental effects of ultrafast magnetic-order quenching and (2) spin transport, and with regard to (3) applications in magnetic switching and generation of terahertz electromagnetic pulses.

II. FUNDAMENTAL EFFECTS: ULTRAFAST DEMAGNETIZATION

A. Relevance and current status

Ultrafast demagnetization, i.e., quenching of ferromagnetic order following excitation by an ultrashort laser pulse[44–50] [Fig. 2(a)], is a key phenomenon of ultrafast and terahertz spintronics. For example, it reveals the time scale of the equilibration of the ordered electron spins with electron-orbital and crystal-lattice degrees of freedom. Furthermore, ultrafast demagnetization is a central component in all-optical ultrafast magnetization switching[34] (Sec. IV).

From a phenomenological viewpoint, excitation by the pump pulse induces (i) a spin accumulation, also known as spin voltage, i.e., a pump-induced excess of magnetization, and (ii) a pump-induced temperature difference between spin-up and spin-down electrons.[51–56] Both (i) and (ii) act as driving forces of the temporal rate of change of the magnetization.

One can refine this model by splitting the electronic system in subsystems, e.g., localized d-or f-type electrons, which carry the ferromagnetic order and magnons, and sp-type conduction electrons,

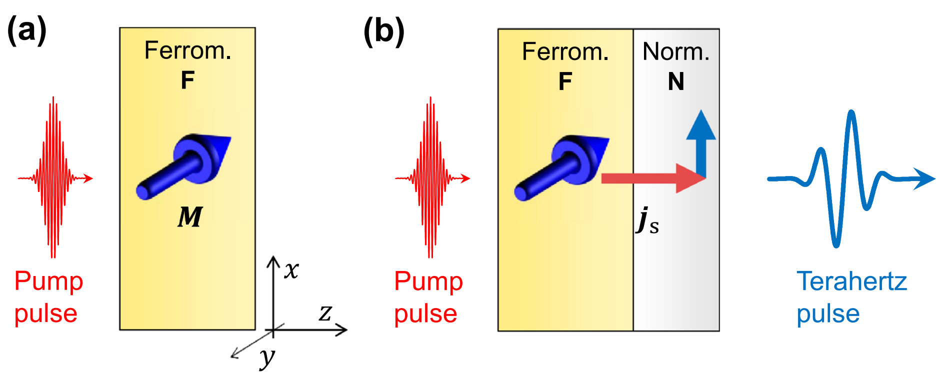

FIG. 2. Ultrafast demagnetization and ultrafast spin transport. (a) Excitation of a ferromagnetic thin film F with a femtosecond laser pulse (pump) leads to a transient quenching of the magnetization M. (b) Femtosecond laser-pulse excitation of a F|N stack consisting of a ferromagnetic layer F and normal-metal layer N triggers a transient ultrafast spin current with density j_s from F to N. Spin-to-charge-current conversion converts the longitudinal spin current into a transient charge current that acts as a source of a terahertz electromagnetic pulse. This figure was adapted from Ref. 55.

which are highly mobile and may strongly interact with the crystal lattice.[57–60]

Microscopically, elementary processes such as electronic spin-flip scattering or electron-magnon and phonon-magnon scattering[61–64] need to be considered. They relate to distinct models, for instance, Stoner-[62] or sd-type[57,59] approaches.

B. Challenges and this Special Topic

Even though ultrafast demagnetization is a relatively mature research subject, there are still numerous open questions, the number of which naturally increases with the complexity of the material under study. Already, in simple 3d-type model ferromagnets, such as Ni, the role of the shape of the pump-induced nonequilibrium electron distribution[49,50] of spin excitations[65–67] and the static crystalline and spin order[68] on the magnetization dynamics has been addressed only recently.

In rare-earth metals, such as Gd and Tb, the magnetization is dominated by highly localized 4f electrons at several electronvolts below the Fermi energy, resulting in potentially more complex ultrafast spin dynamics. Accordingly, Decker *et al.*[69] address ultrafast energy equilibration of Gd 4f and 5d electrons with the crystal lattice. On the other hand, Andres *et al.*[70] report on a one order of magnitude faster characteristic time of ultrafast demagnetization in Tb than in Gd, which is in part ascribed to the very different coupling of 4f spins and crystal lattice in the two materials.

In alloys and multilayers, (anti)ferromagnetically coupled spin sublattices enable additional interactions, particularly in energy and spin transfer between the sublattices. Here, element- and/or state-selective probing is highly desirable and facilitated by extreme ultraviolet and soft x-ray pulses. Remarkably, time-delayed dynamics on the 10–50 fs scale between spin subsystems are observed, for instance between Ni and Fe in the $Fe_{50}Ni_{50}$ alloy,[71] between Pt and Co in $[Co|Pt]_n$ multilayers,[72] and between electrons above and below the Fermi energy in $[Co|Pd]_n$ multilayers.[73] A delayed response of a laser-excited ferrimagnetic iron garnet is also reported when weak vs strong external magnetic fields are applied.[74] Theoretical models allow one to elucidate the role of spin transfer by exchange scattering.[75]

Understanding of the probing mechanism is of central importance in ultrafast spintronics. Time-dependent density-functional theory (TD-DFT) is a powerful approach here. For example, it allows one to study to which extent the sum rules of x-ray magnetic circular dichroism (XMCD), which are used to determine instantaneous magnetic moments, are still valid during and following ultrafast laser excitation.[76] To probe electron-magnon coupling in ferromagnetic Fe, the renormalization of electronic surface states shows larger benchmark values.[77]

C. Outlook

The microscopic understanding of excitations and couplings in solids demands very specific, high-resolution, and spin-sensitive probes. There is an ongoing development of free-electron lasers and high-repetition-rate high-harmonic-generation light sources in combination with dedicated multichannel spectrometers. These efforts will push experimental techniques, such as depth-sensitive resonant magnetic scattering, resonant inelastic x-ray scattering and spin-resolved photoemission to a higher level of performance and, thus, foster new microscopic insight into ultrafast spin dynamics. For example, first experiments[78] have combined terahertz pump pulses with photoemission probes and delivered unprecedented views on electron dynamics, which may open the field of light wave-controlled spintronics. Future work is expected to further proceed from elemental materials[50,68] to complex compounds or multilayers,[72,73] including recent material developments, such as magnetic Weyl semimetals.[79]

III. FUNDAMENTAL EFFECTS: ULTRAFAST SPIN TRANSPORT

A. Relevance and current status

Studying the generation and manipulation of ultrafast charge and spin currents using femtosecond laser pulses is of prime importance. Fundamental questions revolve around the interaction of light with spintronic heterostructures, the mechanisms leading to ultrafast spin transport,[31,53–56,60,80–82] and its relaxation,[55] the dominant carriers[83] (conduction electrons or magnons), and the subsequent transport dynamics (ballistic, superdiffusive, and diffusive).[80] From an applied perspective, ultrafast spin currents are highly relevant for applying spin torque, the acceleration or slowing down of ultrafast demagnetization (Sec. II) and magnetization switching (Sec. IV). Another interesting application is the generation of terahertz electromagnetic pulses (Sec. V).

It is well established that optical excitation of two-layer stacks F|N, where F is a ferro- or ferrimagnetically ordered material and N is a normal metal, leads to the generation of ultrafast spin currents from F to N[22] [Fig. 2(b)]. Following the optical pump pulse, ultrafast spin transport is typically probed by a time-delayed optical probe pulse taking advantage of magneto-optic effects[22] [Fig. 1(b)] or the probe-emitted photoelectrons.[32] Alternatively, one can exploit spin-to-charge-current conversion, resulting in an in-plane charge current that emits a measurable terahertz electromagnetic pulse[29,79,84,85] [Fig. 2(b)].

Phenomenologically, and similar to ultrafast demagnetization (Sec. II), two possible driving forces are discussed: a pump-induced difference of electron temperature (spin Seebeck effect[10,11]) and/or spin voltage[31,52–56] between F and N. Both differences also apply to nonthermal electron distributions in certain models.[10,55] The resulting spin-polarized electron current injected into N starts out ballistically and, once scattering sets in, becomes superdiffusive[80] and eventually diffusive.

Remarkably, in stacks F|N made of rather simple metals, such as $Ni_{80}F_{20}$ for F and Pt for N, the ultrafast spin current from F to N has identical time evolution as the rate of change of the magnetization in ultrafast demagnetization of a single F layer.[55] This behavior indicates that ultrafast spin transport in F|N and ultrafast demagnetization in F are predominantly driven by the same force: the transient spin voltage of F.[31,53–55]

In addition to such band-like transport, a shift-type mechanism,[86] optical intersite spin transfer (OISTR), was suggested recently[87] and studied experimentally.[88–91] In OISTR, optical excitation transfers electronic occupation into states with a shifted center of mass of spin density, resulting in spin transfer from F to N.

A further spin-current mechanism can arise when circularly polarized pump pulses are applied.[92] Microscopically, the pump-helicity-dependent spin transport was explained by inverse spin-orbit torque.

Theoretical approaches to ultrafast spin transport are based on the Boltzmann transport equation[93] or time-dependent density-functional theory.[87,94,95]

B. Challenges and this Special Topic

A central goal of ultrafast spintronics is to maximize the spin current amplitude for a given incident laser fluence, i.e., pulse energy per area [Fig. 2(b)]. To obtain maximum driving spin voltage, materials such as magnetic Weyl semimetals,[79] half-metals,[96] and Co_xFe_{1-x} alloys $(0 < x < 1)$[84] are studied. Maximum interface spin transmission is another crucial parameter and addressed by interface engineering[96] and introducing intermediate layers between F and N.[85] The spatiotemporal structure of the spin current can be controlled by spin-sink layers[96] and depends on the time scales of electron-phonon coupling[97] and the transition from ballistic to diffusive transport.[98] Finally, new spin-current sources need to be explored, for example, chiral antiferromagnets.[99]

Future modeling needs to take all interactions between electron spins, electron orbital motion and phonons, as well as spin, charge, and heat transport on multiple time and length scales into account.

C. Outlook

Even though central aspects of ultrafast spin transport are not yet well understood, particularly, those related to interfaces, promising applications have emerged and will motivate further studies. While previous works have so far mostly considered ultrafast spin transport by conduction electrons, ultrafast magnon transport[100] is expected to yield new and exciting insight and applications such as frequency-comb generation.[101]

IV. APPLICATIONS: MAGNETIC-ORDER SWITCHING

A. Relevance and current status

Ultrafast all-optical switching of magnetic order can be of crucial importance for the development of future generations information-technology elements with low power consumption and fast operation. Ultimately, we aim for operation at terahertz rates. To reach this goal, a solid material base is required, that is, magnetic materials whose properties permit operation at such frequencies. Consequently, studies of magnetic materials in the terahertz range have been expanding strongly in the last decade. In particular, the goal is set to achieve full switching of magnetic order at the time scale of a few picoseconds, or even below that.

Several mechanisms of all-optical switching of magnetic order have been discovered by now. Among them, the most investigated is the ultrafast heating-only-induced switching of the magnetization of ferrimagnetic alloys, witnessed by several contributions in this issue.[102–106] The switching occurs at the sub-picosecond time scale and is toggle by nature. The mechanism has been known for about 15 years and is relatively well understood by now.[102–105] Therefore, ferrimagnetic alloys are considered to be the most reliable possible material system for applications, for example, as recording medium in an ultrafast concept of opto-magnetic memory.

There are several compositions of the ferrimagnet alloys that exhibit single-shot toggle switching. The important ingredient is the presence of two coupled spin sublattices with different intra-sublattice exchange interaction. This feature could easily be realized not only in rare-earth/transition-metal alloys and multilayers but also in rare-earth-free ferrimagnets such as Mn-based Heusler alloys.

B. Challenges and this Special Topic

The most important question related to application is how reliably the magnetization switching can be repeated. During the switching process, the magnetization first crosses zero on a sub-picosecond scale, provided the pump pulse was short enough. However, the subsequent complete recovery of the magnetization along its new (reversed) direction is slower and ultimately limits the repetition rate of the possible re-recording process. Various schemes can be applied to improve this situation.[102] Materials with strong magnetic anisotropy can both accelerate the recovery and assure the stability of small bits. This idea motivated a search among alloys containing various rare-earth materials.[103]

Another important aspect is the energy required for the magnetization reversal process, which is an issue dominating the data-storage industry as a whole. One could reduce this energy considerably by applying smart multilayer schemes.[105] Last but not the least, the theoretical description of the switching process is not yet fully complete,[104] and novel schemes can be proposed for fast and less energy-hungry switching.[106]

C. Outlook

In view of energy consumption as well as residual heating effects, nonthermal mechanisms of ultrafast magnetization reversal attract more and more attention. To avoid heating, magnetic insulators will become materials of choice. The enabling mechanisms will of course be completely different from those in metals. The most interesting possibility may be to use the crystal lattice as the driving parameter. Thus, one can envisage magnetic switching by lattice strain on the femtosecond scale. Alternatively, an optically driven electron charge-transfer transition similarly affects the crystal fields and should, therefore, be able to induce a sufficiently strong transient magnetic anisotropy to achieve the switching.

V. APPLICATIONS: SPINTRONIC SOURCES OF TERAHERTZ ELECTROMAGNETIC PULSES

A. Relevance and current status

Upon excitation by a femtosecond laser pulse, a spintronic terahertz emitter (STE) emits an ultrashort terahertz electromagnetic pulse [Fig. 2(b)]. Such pulses can be used as sensitive and ultrafast probes of all infrared-active resonances of all phases of matter, for example, molecular rotations and vibrations in gases and liquids as well as phonons, intraband electron transport, Cooper pairs, excitons, and magnons in solids. Due to this specificity, terahertz radiation has also found application in imaging and quality control.

As reviewed in detail in an Editorial of this Special Topic,[15] a typical STE consists of a ferromagnetic metal layer F, which acts as a spin-current source, in contact with one or two metal layers N for spin-to-charge-current conversion [Fig. 2(b)]. In the current understanding of STEs, the incident femtosecond laser pulse deposits energy in F and, thus, induces a spin voltage that drives a spin current into the adjacent N (Sec. IV). Due to spin-to-charge-current conversion, the out-of-plane spin current is converted to an ultrafast in-plane charge current that acts as a source of a terahertz electromagnetic pulse [Fig. 2(b)].

Among other attractive features,[15] STEs are broadband without gaps in the emission spectrum at 0.3–30 THz,[35] more efficient than state-of-the-art emitters, such as ZnTe and GaP,[35] independent of the pump wavelength from the infrared[107] to the visible[108] to the extreme ultraviolet,[109] scalable,[110,111] and their linear polarization can be set by an external magnetic field in a contactless manner.[112]

B. Open questions and this Special Topic

An important goal regarding STEs is to extend the amplitude and bandwidth of the emitted terahertz pulses. It can be achieved by larger and faster spin currents (Sec. IV) and more efficient spin-to-charge-current conversion through material optimization and photonic measures.[15] For example, antiferromagnets are found to be interesting candidates for spin-to-charge-current conversion.[85] At the same time, the peculiar features of STEs need to be studied in detail to develop and demonstrate new functionalities.

This Special Topic also reports on exciting functionalities and applications. Modulation of the terahertz polarization direction by an electric voltage rather than an external magnetic field is demonstrated.[113] The voltage is applied to the piezo-electric substrate on which the STE is grown, and the resulting strain-induced magnetic anisotropy rotates the STE magnetization.

The STE area can be upscaled straightforwardly. By simultaneously increasing the driving laser power, peak terahertz fields can be generated that are sufficiently strong (here, 60 kV/cm) to modulate a second, air-plasma-based terahertz source.[114] By further upscaling, terahertz peak fields exceeding 1.5 MV/cm and fluences of about 1 mJ/cm^2 are obtained,[111] which are highly interesting for nonlinear terahertz spectroscopy in the elusive 1–10 THz window.

Finally, the lateral extent of the terahertz near-field directly behind the STE layer equals that of the intensity of the driving femtosecond laser beam. By placing the sample in the terahertz near-field, imaging beyond the diffraction limit can be realized.[115] At 1 THz, a spatial resolution that is a factor of 60 smaller than the wavelength of 300 μm is achieved. A similar approach is used to gain spatially resolved insight into a laterally nanostructured STE.[116]

C. Outlook

To further improve STEs, future research is expected to include novel materials as ultrafast spin-current sources and spin-to-charge-current converters and to explore the so far under-researched role of the F/N interface.

VI. CONCLUSION

Ultrafast and terahertz spintronics have significantly extended the scope and application range of spintronics and magnetism research. In latest applications, ultrafast magnetization switching was achieved all-optically with the assistance of ultrafast spin currents[117] or by the SOT resulting from picosecond charge-current pulses.[118] Optimization and scaling of STEs[15] enabled the spintronic generation of ultrabroadband terahertz pulses with peak fields >1.5 MV/cm,[111] which can be used for nonlinear terahertz spectroscopy.[14,119,120] For the faithful detection of such strong and broadband electromagnetic fields, Zeeman torque is a promising approach.[121] Future directions may take advantage of coherent spin dynamics[122] for neuromorphic computing and other such applications.[123]

Fundamentally, there are strong efforts to push genuine spintronic effects to terahertz frequencies. Examples include magnetoresistive effects such as the anomalous Hall effect,[124–129] anisotropic magnetoresistance,[9,125,130,131] giant magnetoresistance,[7] and tunnel magnetoresistance[8] as well as torques such as the magnetic-field-induced Zeeman torque,[132] the electric-field-driven SOT[17] in magnetic multilayers, and the Néel SOTs[118,133] in novel antiferromagnets for antiferromagnetic spintronics.

Finally and remarkably, angular momenta other than electronic spin are currently under consideration as information carriers: the orbital angular momentum of electrons[134,135] and the angular momentum of the crystal lattice and its vibrations (phonons).[136] Early works have shown that the transfer of electron orbital angular momentum[137,138] and lattice angular momentum,[139] the latter making use of chiral materials,[140] can be pushed to ultrafast time scales. State-of-the-art theory, in particular, the calculation of response functions[141,142] and time-dependent density-functional theory,[94,95] will crucially support and possibly even guide these developments.

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