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ABSTRACT

Spintronic terahertz emitters are broadband and efficient sources of terahertz radiation, which emerged at the intersection of ultrafast spintronics and terahertz photonics. They are based on efficient spin-current generation, spin-to-charge-current conversion, and current-to-field conversion at terahertz rates. In this Editorial, we review the recent developments and applications, the current understanding of the physical processes, and the future challenges and perspectives of broadband spintronic terahertz emitters.

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I. INTRODUCTION

A. Terahertz time-domain spectroscopy

Terahertz (THz) radiation covers the range from about 0.1 to 30 THz. It holds great promise for basic research and future applications,^{1,2} because the THz frequency range coincides with many low-energy modes in all phases of matter, i.e., plasmas, gases, liquids, and solids.³ For example, THz radiation can resonantly couple to conduction-electron transport, plasmons, excitons, Cooper pairs, phonons, or magnons.⁴ Thus, THz spectroscopy is a powerful tool to study fundamental processes in a wide range of materials.

THz radiation not only serves as a probe: The development of high-amplitude THz sources enables the control of collective excitations of matter^{5–7} such as magnons in magnets^{8–11} or driving of phonons.^{12–16} Currently, THz electric fields reach peak strengths of the order of 1 MV/cm in table-top systems, and they exceed ~ 10 MV/cm in large-scale user facilities such as free-electron lasers.¹⁷ Upon excitation with intense THz pulses, ultrafast switching between different phases of matter (e.g., topological, magnetic, and structural) was observed recently.^{8,18–25} THz excitation can also be combined with other well-established experimental probes such as angle-resolved photoemission spectroscopy,²⁶ scanning tunneling microscopy,^{27–29} or x-ray diffraction.^{30,31} Merging THz spectroscopy with such powerful

probing techniques can yield unique multidimensional insight into fundamental processes on ultrafast time scales.

In terms of applications, THz imaging and sensing have recently gained considerable interest.³² For instance, THz imaging holds great promise for biomedical and security applications since THz waves can be transmitted through living biological systems without harm, in contrast to wavelengths in the ultraviolet to x-ray range. THz imaging can be used for quality control and inspection such as in pharmaceutical,³³ manufacturing,^{34,35} and security applications.³⁶ Meanwhile, a THz radar is also under intense development because of its potential for autonomous driving and military applications.^{37,38}

The recent development of sub-wavelength THz microscopy, for instance, by using sub-wavelength apertures,^{32,39} tip-like probes, or the near-field of the THz source,^{32,40} led to spatial resolutions deep into the micrometer range or even down to tens of nanometers, i.e., far below the THz wavelengths of about $10\ \mu\text{m}$ to 3 mm. With the combined advantages of high spatial resolution and low-frequency ultrafast optical response, THz near-field techniques are foreseen to become versatile tools to reveal the THz response of materials at the nanoscale.

Based on these promising prospects for THz radiation, the exploration and development of more capable THz sources with high efficiency, broad frequency bandwidth, and easy access have become one

of the most important goals in the THz-photonics community. Thus, we will focus on table-top laser-driven approaches for the generation of THz radiation in the following.

B. Generation of THz radiation

The generation of electromagnetic radiation, including THz radiation, requires a time-dependent charge-current density \mathbf{J} . In the frequency domain, the electric field \mathbf{E} of the resulting electromagnetic wave is determined by the wave equation⁴¹

$$-\nabla \times (\nabla \times \mathbf{E}) + \frac{n^2 \omega^2}{c^2} \mathbf{E} = -\frac{iZ_0 \omega}{c} \mathbf{J}. \quad (1)$$

Here, ∇ is the spatial derivative (Nabla operator), $\omega/2\pi$ is the frequency, n is the refractive index, c is the vacuum speed of light, and $Z_0 \approx 377 \Omega$ is the free-space impedance. In general, \mathbf{E} , \mathbf{J} , and n are complex-valued and depend on the spatial position and frequency.

According to Eq. (1), two conditions must be fulfilled to obtain a broadband and high-amplitude THz emitter: (a) maximum amplitude and bandwidth of \mathbf{J} and (b) constructive superposition, i.e., phase matching, of all THz waves generated throughout the volume of the emitter.⁴² Condition (b) critically depends on the spatial distribution of the material-specific parameter n .

1. Ultrafast photocurrents

When the current \mathbf{J} is dominated by the electrons of the emitter material, it has contributions from the orbital motion of the electrons and from the spinning current of their spins.⁴³ Accordingly, the total source current density can be written as a sum

$$\mathbf{J} = \nabla \times \mathbf{M} + \mathbf{J}_{\text{orb}} \quad (2)$$

of terms due to (1) the electron spin density (magnetization) \mathbf{M} and (2) the orbital electron currents \mathbf{J}_{orb} .⁴¹ Both contributions can be driven resonantly and off-resonantly when a material is illuminated with light, as discussed in more detail in the following.

- (1) A time-dependent magnetization $\mathbf{M}(t)$ gives rise to the emission of magnetic-dipole radiation. An example is the THz emission that arises from the ultrafast magnetization quenching in a ferromagnet by resonant excitation of conduction electrons with a femtosecond laser pulse.^{44–46} A nonresonantly driven term (1) is, for instance, possible by the inverse Faraday effect in an optically transparent material, in which torque is exerted on \mathbf{M} , that is, magnon modes are excited by the driving laser pulse.^{47–49} While mechanism (1) is highly interesting for the detection of magnetization dynamics, it typically lacks efficiency compared to other THz-generation mechanisms described by term (2), because magnetic dipoles radiate less efficiently than electric dipoles.
- (2) In crystalline solids and for resonant optical excitation of electrons, \mathbf{J}_{orb} can be further divided into shift and injection currents.^{50–53} In a simplified picture, a shift current is a displacement of charges within the solid's unit cell. The current density typically has bipolar dynamics owing to the forward and subsequent backward flow of the electronic charge density, convoluted with the pump-pulse intensity envelope. A typical example of a resonant shift current is the above-bandgap

excitation of bulk GaAs⁵⁴ or the excitation of the surface-near regions of the topological insulator Bi₂Se₃.⁵³

In contrast, injection currents arise from optically induced changes in the group velocity of electrons, and the current amplitude is proportional to a typically unipolar response function describing electronic relaxation, again convoluted with the pump-pulse intensity envelope. Injection currents can be spin-polarized and are maximized for circularly polarized driving light.⁵² They are by far less popular for THz-pulse generation and are found by, for instance, resonant excitation of electrons with circularly polarized light in wurtzite semiconductors⁵² or in topological insulators.⁵⁵

For below-bandgap ultrafast optical excitation of transparent nonlinear optical crystals such as ZnTe, GaP, and GaSe,^{1,2} the photocurrent generation mechanism is referred to as optical rectification. This off-resonant process can be understood as a shift current with vanishing relaxation time.

2. Emission of the THz pulse

The THz-generation efficiency of nonlinear optical crystals typically increases with thickness, because a larger volume emits THz waves. However, an increased thickness often lowers the generated THz bandwidth due to a phase-mismatch of the pump field and the emitted THz field and because of THz-radiation absorption by infrared-active phonons, thus violating condition (b). For instance, the reststrahlen band in polar semiconducting crystals typically strongly attenuates emission of THz radiation in the range between about 5 and 10 THz. Thus, a broad bandwidth and high efficiency are usually difficult to achieve simultaneously.

In general, the emitted THz-field amplitude is only sizable if the ultrafast photocurrent exhibits a nonvanishing transient electric dipole moment and, thus, broken inversion symmetry. While in materials such as GaAs, ZnTe, or GaP, inversion asymmetry is intrinsically provided by the crystal structure, it may also be induced by external means. An example is a photoconductive antenna, in which inversion symmetry is broken by applying a static electric field, which accelerates the photo-generated charge carriers. The resulting transient charge current emits a THz pulse.^{1,2} Photoconductive antennas are currently among the most widely used THz emitters due to their high efficiency and excellent usability. Typically, their bandwidth is limited to 0.1–5 THz [detrimental to condition (a)] by the relatively long-lived current flow in the used semiconductor materials (e.g., LT-GaAs).^{56–59}

A notable exception is Ge, which is a nonpolar semiconductor and, therefore, lacks one-phonon absorption. Thus, Ge photoconductive THz emitters allowed one to extend the THz bandwidth $\Delta\omega_{\text{THz}}^{10\%}/2\pi$ up to about 10 THz, yet with reduced THz-generation efficiency.⁶⁰ Note that $\Delta\omega_{\text{THz}}^{10\%}$ is defined as the full width at 10% of the maximal THz electric field amplitude in the spectral domain at the detector position.

Another approach to generate THz radiation relies on laser-induced plasmas, in which the driving laser fields break the inversion symmetry.⁶¹ Such THz plasma sources⁶² can reach 0.1 mJ pulse energy at a large bandwidth of up to about 100 THz or up to 50 mJ pulse energy at 1 THz bandwidth. However, their energy fluctuation is typically larger than that of other commonly used THz sources due to the high degree of nonlinearity of the generation process. The large pump-pulse energies require costly amplified laser systems.

Therefore, new THz-emitter concepts need to be explored. They should simultaneously satisfy the requirements of high THz-generation efficiency, broad bandwidth, stability, and easy access.

C. Spintronic THz emitters

The recent development of ultrafast spintronics and femtomagnetism paved the way toward a promising THz-emitter concept: a spintronic thin-film stack FM|NM consisting of layers of a ferromagnetic metal (FM) and a normal metal (NM) (see Fig. 1).^{63,64} After optical excitation of such a FM|NM heterostructure, an ultrafast spin current flows across the interface from the in-plane magnetized FM into an adjacent NM layer. The ultrafast out-of-plane spin current is converted into an in-plane charge current through a spin-to-charge-current conversion (S2C) process. If the NM consists of a heavy metal like Pt, S2C predominantly happens in the NM layer by the inverse spin Hall effect (ISHE).^{63–66} The resulting sub-picosecond transient charge current emits electromagnetic waves with THz frequencies.

Such spintronic THz emitters (STEs) have interesting properties:

1. They were shown to emit radiation with frequencies between 0.1 and 30 THz without any spectral gaps, thus outperforming photoconductive switches or nonlinear optical crystals (e.g., ZnTe and GaP) in terms of bandwidth.⁶⁴
2. STEs can be driven by virtually any pump wavelength,^{67,68} in stark contrast to THz sources based on semiconductors.
3. The efficiency and long-term stability of STEs are comparable with or even better than commonly used THz sources (see Fig. 7).
4. The fabrication procedure is based on well-established thin-film growth processes, which enable high-quality emitters with an outstanding homogeneity.^{64,69}
5. The emitted THz field of the STE scales linearly with the excitation fluence up to about 0.1 mJ/cm². Only at about 5 mJ/cm², permanent STE damage is induced.⁷⁰
6. One can conveniently grow large-area STEs to enable THz-high-field applications. In fact, by expanding the pump-laser beam to avoid optical damage of the STE, high pump powers of

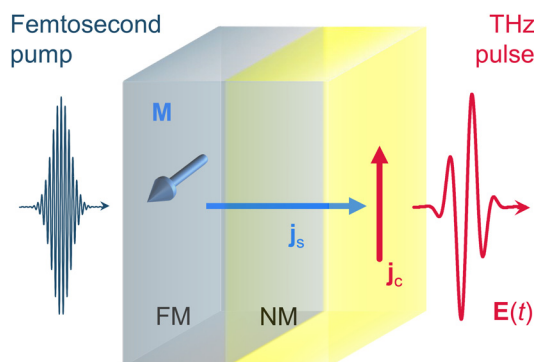


FIG. 1. Electromagnetic-wave generation process in a spintronic THz emitter. An ultrashort laser pulse triggers a spin-current injection from the in-plane magnetized ferromagnetic metal (FM) into the normal metal (NM). In the next step, the spin-orbit interaction in the NM converts the spin current j_s into an in-plane charge current j_c , which finally emits a THz electromagnetic pulse. Directly behind the sample, the electric field of the linearly polarized THz pulse is perpendicular to the sample magnetization M .

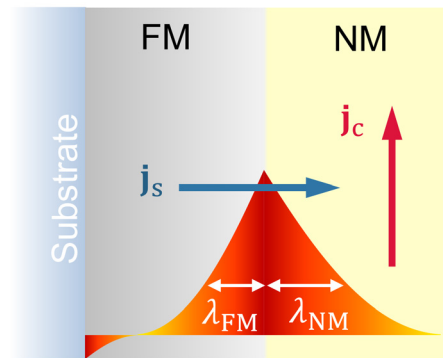


FIG. 2. Ultrashort spin current following optical excitation in FM|NM stacks. The spin current decays over a length λ_{NM} inside the NM layer, which depends on the material and on frequency. In the FM layer, an analogous relaxation length λ_{FM} quantifies the mean FM layer depth from which spin-polarized FM electrons still reach the FM|NM interface. Reproduced with permission from Gueckstock *et al.*, *Adv. Mater.* **33**, e2006281 (2021). Copyright 2021 Authors, licensed under a Creative Commons Attribution (CC BY) license.

7. As a STE is made of thin metallic films, it can be easily micro-structured, allowing for photonic nano-engineering to tailor the STE performance.^{65,71–73}
8. The emitted THz pulse is linearly polarized, and the THz electric field is to a very good approximation perpendicular to the FM magnetization, which can be conveniently controlled by an external magnetic field,⁷⁴ thereby enabling modulation of the THz emission at kHz rates.⁷⁵
9. Employing more complex magnetization patterns inside the STE allows one to create exotic THz-beam polarization states.⁷⁶
10. As the STE is only a few nanometers thick, no phase-matching conditions need to be fulfilled, and the substrate can act as an efficient heat sink. Moreover, the pump beam profile is preserved during the THz generation mechanism, making tight focusing of the generated THz beam possible.
11. The STE enables near-field imaging approaches with deep-subwavelength resolution.⁷⁷

Thus, the STE combines ultrabroad bandwidth, high efficiency, ease of use, and flexibility in terms of geometry and design.^{64,66} These benefits demonstrate the potential of STEs in terms of THz applications and as a THz source even beyond the traditional THz community, which might be further fostered by the recent commercial availability of the STE (TeraSpinTec GmbH).^{69,78,79}

Recently, several reviews were published that highlight specific aspects of spintronic THz emission and optically generated ultrafast currents, which we would like to recommend to the reader.^{80–86} In this Editorial, we review the fundamentals of STEs, various THz generation mechanisms, their optimization, applications, challenges, and future perspectives. Special attention lies on the microscopic mechanisms and the resulting potential in terms of optimizing spintronic THz sources.

II. KEY EMPIRICAL RESULTS OBTAINED FROM STEs

STEs take advantage of ultrafast spin dynamics in FM|NM heterostructures to reach high THz-generation efficiency and broad bandwidth. They complement the capabilities of traditional THz emitters with their spintronic principle of operation. In this section, the key empirical results for different STE realizations are presented.

A. Model of the STE operation

A typical STE is a heterostructure containing FM and NM layers with thicknesses of several nanometers. When excited with a femtosecond laser pulse, the structure emits THz radiation. We model this phenomenon as a result of four elementary processes (see Fig. 1):

- (1) absorption of the optical pump pulse,
- (2) generation of a spin current flowing from the FM into the NM layer,
- (3) conversion of the spin current into a transverse charge current, which
- (4) acts as a source of a THz electromagnetic pulse.

In the following, we address processes (1)–(4) from a phenomenological viewpoint, whereas the microscopic mechanisms of processes (1)–(3) will be discussed in more detail in Sec. III.

- (1) The absorbed pump-pulse energy is initially deposited primarily in the electronic system of the FM|NM stack. The resulting transient electron distributions are expected to be significantly different for the FM and NM layers.
- (2) Macroscopically, a net spin transport from the FM to the NM layer is allowed, because the FM|NM stack exhibits a broken inversion symmetry. Microscopically, the spin current can be carried by spin-polarized conduction electrons and/or magnons. Phenomenologically, spin currents can be driven by gradients of electrostatic potential, electron temperature, and the spin accumulation, which is also called spin voltage.⁸⁷ If the FM is insulating and not excited by the pump pulse, the transient temperature gradient between the cold FM and the hot NM layer is the dominant driver of a magnon-type spin current through the interfacial spin Seebeck effect (SSE).⁸⁸

In contrast, a metallic FM layer, such as Fe, Co, or Ni, exhibits a higher electronic temperature following the pump absorption and, thus, aims at reducing its magnetization. The spin angular momentum can be released to the FM crystal lattice or by transport to the NM layer. In the framework of the Stoner model of ferromagnetism, one can show that the electron spin current arises from a gradient of the electronic temperature and the spin voltage between FM and NM layers.⁸⁹ The spin voltage μ_s can be understood as an excess of spin density (magnetization) in the FM layer that needs to be released to attain equilibrium. For a more detailed discussion, please see Sec. III A.

The resulting spin current with density $\mathbf{j}_s(z, t)$ flows parallel to the normal of the FM|NM stack (Fig. 2). Upon crossing the FM/NM interface, the spin current undergoes spin loss and reflection, and only a fraction of the spins is injected into the NM layer.^{64,90,91} Inside the NM layer, the spin-current density $\mathbf{j}_s(z, t)$ decays with increasing depth $z > 0$ due to various electron scattering processes.⁶³ As shown in Fig. 2, the spatial decay is characterized by a relaxation length λ_{NM} , which depends on the material and on frequency.⁹² In the FM layer, an analogous

relaxation length λ_{FM} exists. It quantifies the mean propagation length of spin-polarized FM electrons that still reach the FM|NM interface.

- (3) While flowing, the spin current $\mathbf{j}_s = j_s \mathbf{u}_z$ is partially converted into a transient charge current $\mathbf{j}_c = j_c \mathbf{u}_z \times \mathbf{M}/|\mathbf{M}|$ by S2C. The direction of \mathbf{j}_c is perpendicular to the sample normal \mathbf{u}_z and the FM magnetization. Phenomenologically, S2C can in the frequency domain be described by

$$\mathbf{j}_c(z, \omega) = \gamma(z, \omega) \mathbf{j}_s(z, \omega), \quad (3)$$

where the unitless parameter γ quantifies the S2C strength. Both j_s and j_c have the unit $\text{s}^{-1} \text{m}^{-2}$.

S2C arises from spin-orbit coupling, and two major mechanisms are considered: the ISHE and the inverse Rashba-Edelstein effect (IREE). For the ISHE, γ is accordingly called the spin Hall angle. While the ISHE is symmetry-allowed in the bulk of any material, the IREE requires a locally broken inversion symmetry. Therefore, the IREE contributes to $\gamma(z, \omega)$ only at positions z close to interfaces such as of the FM and NM layers or in materials with inversion asymmetry in the bulk. Furthermore, the ISHE can often be considered instantaneous over the relevant frequency range 0–40 THz,⁷⁹ implying a negligible frequency dependence of γ . In contrast, the IREE scales with the accumulated spin density and may, therefore, exhibit memory effects, which imply a frequency dependence of γ .

- (4) In the final step, the ultrafast in-plane \mathbf{j}_c emits THz radiation according to Eqs. (1) and (2). For FM|NM-stack thicknesses much smaller than the THz wavelength and the THz attenuation length inside the FM and NM layers, the THz electric field is in the frequency domain approximately equal to⁹³

$$\mathbf{E}(\omega) = -eZ(\omega) \int dz \mathbf{j}_c(z, \omega). \quad (4)$$

Here, $-e$ is the electron elementary charge, and Z is the frequency-dependent impedance of the STE, which quantifies the current-to-electric-field conversion. It can be calculated according to

$$Z(\omega) = \frac{Z_0}{n_1(\omega) + n_2(\omega) + Z_0 G(\omega)}, \quad (5)$$

with the free-space impedance $Z_0 \approx 377 \Omega$ and the refractive indices $n_1(\omega) \approx 1$ and $n_2(\omega)$ of air and the substrate, respectively. The THz sheet conductance $G(\omega)$ of the metal stack is $\int_0^{d_{\text{NM}}+d_{\text{FM}}} dz \sigma(z, \omega)$, where σ is the local THz conductivity of the metal layers.

B. Experimental implementation

1. STE fabrication

Because typical STEs consist of metallic films that are only a few nanometers thick, a variety of well-established thin-film deposition techniques can be used to grow STEs. The most prominent one is sputter deposition, which typically results in polycrystalline metallic films.^{64–66} STEs made of single-crystalline FM and NM layers can be obtained via post-growth thermal annealing⁹⁴ or growth by molecular beam epitaxy.^{64,95}

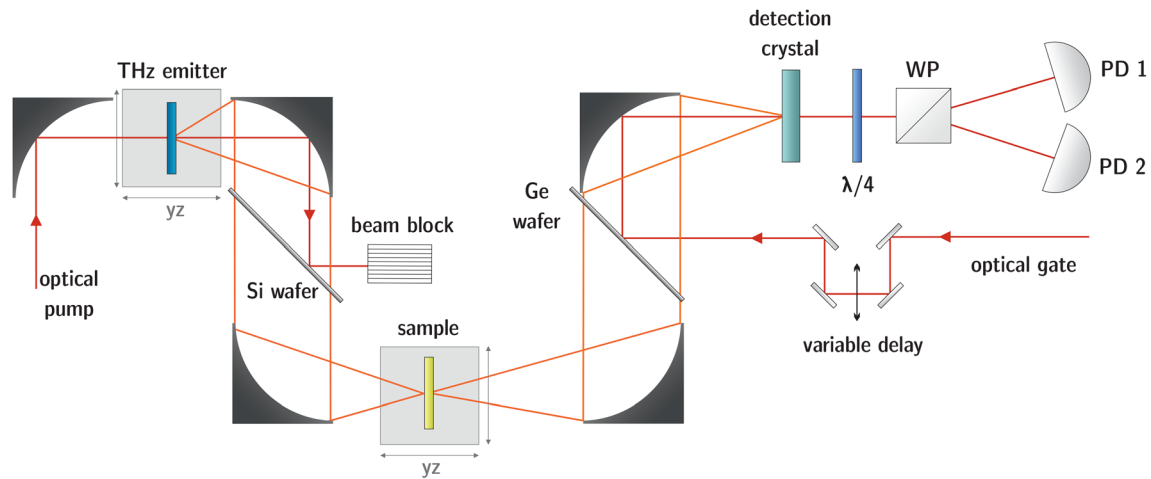


FIG. 3. A typical THz time-domain spectrometer. An optical pump pulse is focused onto a THz emitter. The emitted THz wave is collimated by an off-axis parabolic mirror and subsequently focused onto the sample. A Si wafer is used to block the residual pump pulse. After recollimation, the optical-gate beam is combined with the THz beam by using a Ge wafer. THz and gate beams are focused into the electro-optic detection crystal. By delaying the two pulses with respect to one another, the THz-field-induced birefringence, a measure of the THz-electric-field strength in the detection crystal, is mapped out using a balanced detection scheme consisting of a quarter-wave plate ($\lambda/4$), a Wollaston prism (WP), and two photodiodes (PD1 and PD2).

2. Optical/THz setup

The STE is usually driven by a pulsed femtosecond laser and used in combination with THz time-domain spectroscopy (THz-TDS). This spectroscopic mode allows one to retrieve not only the amplitude but also the phase information of the THz pulse directly in the time domain.⁹⁶ To this end, the THz pulse is measured in a phase-resolved manner by electro-optic (EO) detection in a nonlinear optical crystal such as GaP or ZnTe. In these crystals, the THz electric field induces a transient birefringence via the linear EO effect. The refractive index of the EO crystal changes in proportion to the instantaneous THz electric field, which is sampled by a time-delayed optical probing pulse. By measuring the acquired probe-pulse ellipticity vs the delay between the THz and the probe pulse, the entire THz waveform is mapped out. A typical THz-TDS setup including the STE is schematically shown in Fig. 3.

Note that EO sampling (EOS) does not record the THz electric field directly. Instead, the measured THz signal $S(t)$ is determined by the convolution of the THz electric field $E_{\text{Det}}(t)$ right in front of the detector with the response function $H_{\text{EOS}}(t)$ of the THz detection process. In the frequency domain, $S(\omega)$ and $E_{\text{Det}}(\omega)$ are connected by a multiplication⁶⁴

$$S(\omega) = H_{\text{EOS}}(\omega)E_{\text{Det}}(\omega). \quad (6)$$

Figure 4 shows the spectra $E_{\text{Det}}(\omega)$ and $S(\omega)$ of a typical THz pulse emitted from a STE upon pumping with ~ 10 fs optical laser pulses and detection using a 50- μm -thick GaP EO detection crystal. The minimum of $|S(\omega)|$ at ~ 8 THz is due to $H_{\text{EOS}}(\omega)$ [Fig. 4(b)] and results from the compensation of purely electronic and Raman-type contributions involving the crystal lattice to the EO effect in GaP.⁹⁷ As with the THz-generation process discussed above, by increasing the EO crystal thickness, the detected signal $S(t)$ increases in amplitude, but the bandwidth of $H_{\text{EOS}}(\omega)$ is reduced simultaneously because of the increased THz-field attenuation and phase mismatch between the THz and the probe pulse. Therefore, in the study of the STE, thin GaP

is a suitable choice as the EO detector due to its relatively large bandwidth [see Fig. 4(b)] and sufficient signal strength.

C. Typical THz-emission signals from STEs and their optimization

Based on our model [Eqs. (3)–(5)], we now turn to implementations of STEs based on various material systems and mechanisms, which can strongly influence their performance and properties. We present the different optimization steps that, in hindsight, provided valuable insight into the underlying STE physics.

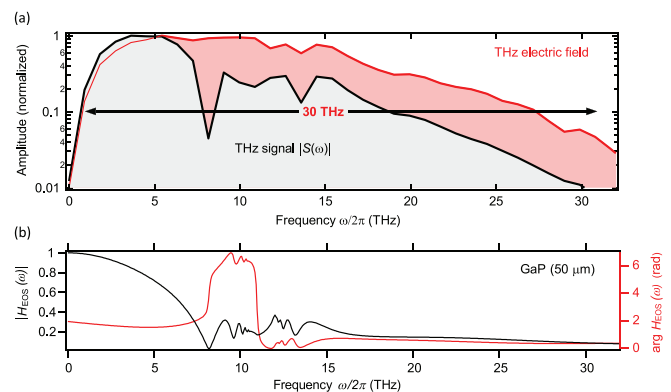


FIG. 4. Typical data obtained from a spintronic THz emitter with a THz time-domain spectrometer. (a) Amplitude spectrum $|S(\omega)|$ (black curve) of the electro-optic signal of a THz pulse from a FM|NM stack and the corresponding extracted THz electric field spectrum $|E_{\text{Det}}(\omega)|$ (red curve) at the position of the 50- μm -thick GaP detector. (b) Complex-valued response function $|H_{\text{EOS}}(\omega)|$ of the 50- μm -thick GaP electro-optic crystal for the case of a 10 fs, 800 nm gate pulse used for THz detection.⁶⁴ Reproduced with permission from Seifert *et al.*, Nat. Photonics **10**, 483 (2016).⁶⁴ Copyright 2016 Macmillan Publishers Limited.

1. Typical THz-emission signals from STEs

In 2013, THz emission from metallic spintronic FM|NM heterostructures was reported.⁶³ The THz waveforms emitted from the studied Fe|Au and Fe|Ru thin films differed in polarity and dynamics, yet both THz signals reversed entirely upon reversing the in-plane sample magnetization [Fig. 5(a)]. The corresponding THz emission spectrum of Fe|Au peaked at 4 THz and had a bandwidth of about 10 THz [Fig. 5(b)], which was much broader than the spectrum obtained from Fe|Ru. This finding indicated different dynamics of the ultrafast currents in Au and Ru. *Ab initio* calculations provided an explanation of the different dynamics within the superdiffusive spin-transport model (Sec. III A 1).⁹⁸ This work identified a prototype FM|NM STE based on the ISHE and showed the potential of THz-emission spectroscopy for the study of ultrafast spin-related transport dynamics.

Subsequent work⁶⁴ put Eq. (3) to test by various experimental checks. First, the THz signal was found to reverse upon reversing the sample magnetization [Fig. 6(a)].^{63,64} Second, by growing the FM|NM bilayer in the reversed order, i.e., NM|FM, the polarity of the detected THz signal reversed because j_s flowed from FM to NM and, thus, changed its sign [Fig. 6(b)]. Third, a sinusoidal dependence of the THz amplitude on the direction of the external magnetic field was found when a THz polarizer was inserted into the optical path, which demonstrated that the emitted THz pulse was linearly polarized with the THz electric field perpendicular to M [Fig. 6(c)]. Fourth, the ISHE was confirmed as the dominant S2C mechanism by comparing the sign and the amplitude of the THz emission for different NM layers⁶⁴ (see Sec. II C 3).

Following these checks, an in-depth optimization of the STE's geometry and materials was conducted, as detailed in Secs. I and II. The resulting optimized STE trilayer exhibits a remarkable performance as a THz source,⁶⁴ as compiled in Sec. I C. In particular, the electric field [Fig. 7(a)] and bandwidth [Figs. 4(a) and 7(b)] of the

emitted THz pulse from the STE right in front of the EO detector were compared to other commonly used pulsed THz sources. The results highlight the high THz-emission efficiency from the STE, as well as its ultrabroad continuous spectrum. The extracted electric-field bandwidth $\Delta\omega_{\text{THz}}^{10\%}/2\pi$ at the detector position reaches about ~ 30 THz when using 10 fs pump pulses centered at a wavelength of 800 nm (Fig. 7). Therefore, spintronic multilayers are a promising ultrabroadband THz source with excellent performance and offer interesting perspectives for implementing future spintronic developments aiming at, for instance, even higher S2C performances.⁹⁹

2. Optimization of the STE THz-emission amplitude

The STE has the advantages of simple device structure, easy fabrication, adjustable polarization, and ultrabroad bandwidth, making it a promising THz source. However, to widen its applications, its THz-generation efficiency needs to be optimized. In recent years, several studies aimed at optimizing the STE in terms of THz-emission efficiency,^{64–67,69,71,73,94,95,100–106} the ease-of-use and compatibility^{68,107–109} as well as tunability of the THz-emission properties such as the polarization.^{72,74,110,111} From such optimization studies, one might not only expect an enhanced STE performance but also a more in-depth understanding of the physical phenomena governing the STE operation.

To analyze the STE performance, we start with Eqs. (3)–(5) and make the following assumptions: The spatial profile of the spin current j_s is calculated from spin-diffusion equations,¹¹² the spin-current amplitude is assumed to scale linearly with the energy density deposited by the pump pulse, and S2C is dominated by the ISHE in the NM layer. Thus, for metal films that are thin compared to the THz wavelength and the THz as well as the optical penetration depth. In other words, for film thicknesses well below 20 nm, the emitted THz electric field can be described in the plane-wave approximation by^{64,105}

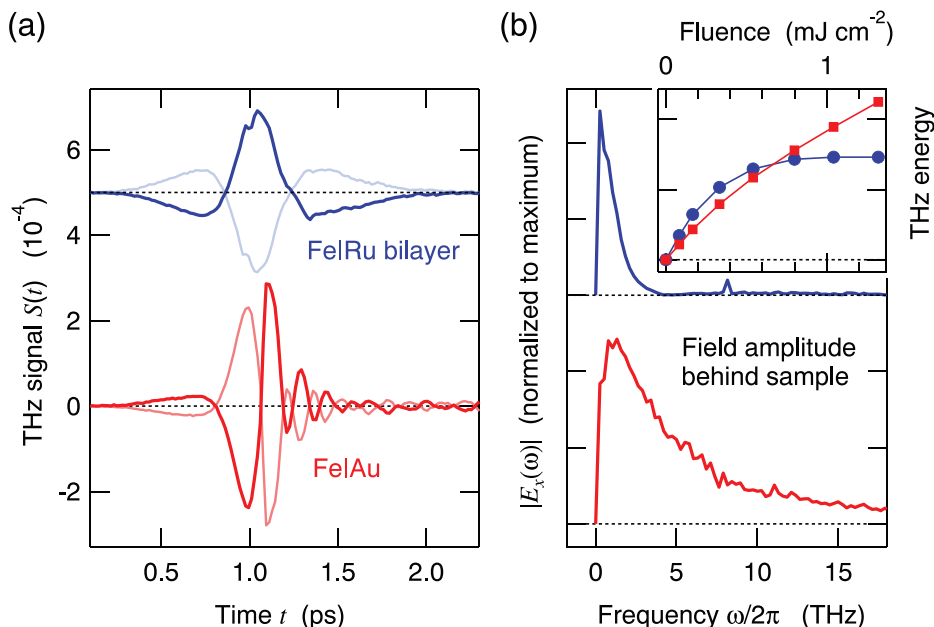


FIG. 5. Experimental demonstration of THz emission in spintronic FM|NM stacks. (a) Emitted THz waveforms from Fe|Ru and Fe|Au thin-film heterostructures for opposite sample magnetizations. (b) The Fourier transform of (a), the inset is the pump-fluence-dependence of the THz energy. Reproduced with permission from Kampfrath *et al.*, Nat. Nanotechnol. **8**, 256–260 (2013).⁶³ Copyright 2013 Macmillan Publishers Limited.

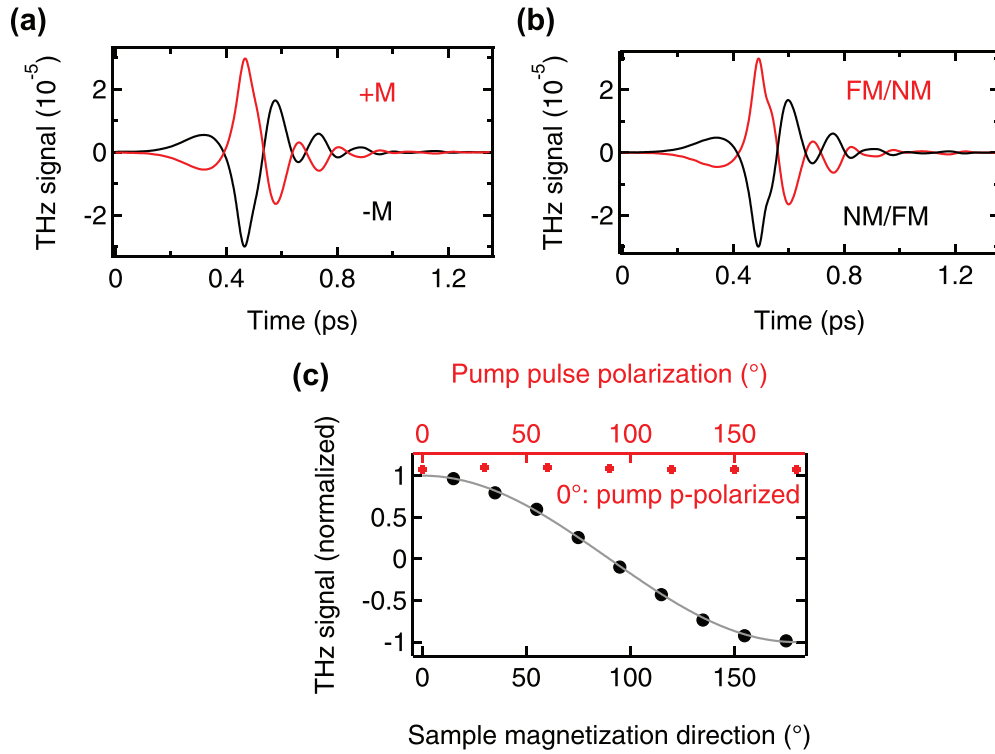


FIG. 6. THz-signal symmetries from STEs consisting of FM|NM thin-film stacks with large spin-orbit coupling. (a) Polarity dependence of the detected THz signal in the direction of the sample magnetization M . (b) Detected THz-emission signals from FM|NM and NM|FM bilayers, i.e., with reversed growth order. (c) Dependence of the THz emission signal on the angle of the external magnetic field in the sample plane with a polarization-sensitive THz detection (black) and in the polarization direction of the linearly polarized pump pulse (red). Reproduced with permission from Seifert *et al.*, Nat. Photonics **10**, 483 (2016).⁶⁴ Copyright 2016 Macmillan Publishers Limited.

$$E_{\text{THz}} = \frac{AF_{\text{inc}}}{d_{\text{NM}} + d_{\text{FM}}} \cdot \underset{(1)}{j_s^0} \underset{(2)}{t_{\text{FM/NM}}} \underset{(3)}{\lambda_{\text{NM}}} \underset{(4)}{\tanh \frac{d_{\text{NM}}}{2\lambda_{\text{NM}}}} \cdot \gamma \cdot \frac{eZ_0}{n_1 + n_2 + Z_0G}. \quad (7)$$

Here, the terms (1)–(4) correspond to the different model steps introduced in Sec. II A: (1) pump-pulse absorption, (2) spin-current generation, (3) spin-to-charge current conversion, and (4) charge-current-to-electric-field conversion. In Eq. (7), the quantity A is the absorbed fraction of the incident pump-pulse fluence F_{inc} , j_s^0 is the generated spin-current density per pump-pulse excitation density, $t_{\text{FM/NM}}$ is the interfacial spin-current transmission amplitude between the FM and NM layers, λ_{NM} is the spin-current relaxation length in the NM layer, and γ is the NM spin Hall angle. The quantities n_1 , n_2 , e , and G were already introduced following Eqs. (4) and (5). All parameters except d_{NM} , e , Z_0 and those in term (1) depend, in principle, on the THz frequency. Table I summarizes typical values of the parameters that enter Eq. (7) found in optimized STEs (see Sec. II C 2).

A recently introduced model¹¹³ that involves spin voltages (see Sec. III A 3) allows one to derive a relation between material-specific parameters and j_s , which reads as

$$j_s(t) \propto \int d\tau I(\tau) H_{j_s, I}(t - \tau).$$

with the impulse-response function

$$H_{j_s, I}(t) = \Theta(t) \left[A_{\text{es}} e^{-\Gamma_{\text{es}} t} - A_{\text{ep}} e^{-\Gamma_{\text{ep}} t} \right]. \quad (8)$$

Here, $I(t)$ is the pump-pulse intensity envelope, Θ is the Heaviside step function, Γ_{es}^{-1} and Γ_{ep}^{-1} are the time constants of electron-spin and electron-phonon equilibration, respectively, $A_{\text{es}} = (\Gamma_{\text{es}} - R\Gamma_{\text{ep}})/(\Gamma_{\text{es}} - \Gamma_{\text{ep}})$ and $A_{\text{ep}} = (1 - R)\Gamma_{\text{ep}}/(\Gamma_{\text{es}} - \Gamma_{\text{ep}})$, and R is the ratio of the electronic and total heat capacity of the sample. In essence, the spin-current dynamics inside the STE¹¹⁴ are determined by a rise time given by the pump-pulse duration and a decay that has contributions of Γ_{es}^{-1} (typically 100 fs) and Γ_{ep}^{-1} (typically several 100 fs).¹¹³ This result implies that the bandwidth of the STE is ultimately only limited by the pump-pulse duration and by the electron-spin relaxation time.

In summary, the THz-emission efficiency depends strongly on the material properties of the FM and NM layers, as well as the photonic and geometric design of the STE. The impact of all these parameters on the STE performance will be discussed in the following.

3. Material choice

In view of Eq. (7), intrinsic sample parameters that impact the THz generation efficiency are γ , λ_{rel} , $t_{\text{FM/NM}}$, j_s^0 , A , and $Z = Z_0/(n_1 + n_2 + Z_0G)$.

a. NM variation. Seifert *et al.*⁶⁴ performed systematic studies of the STE's NMs including Cr, Pd, Ta, W, Ir, Pt₃₈Mn₆₂, and Pt. It was found that the emitted THz amplitude was highly dependent on the

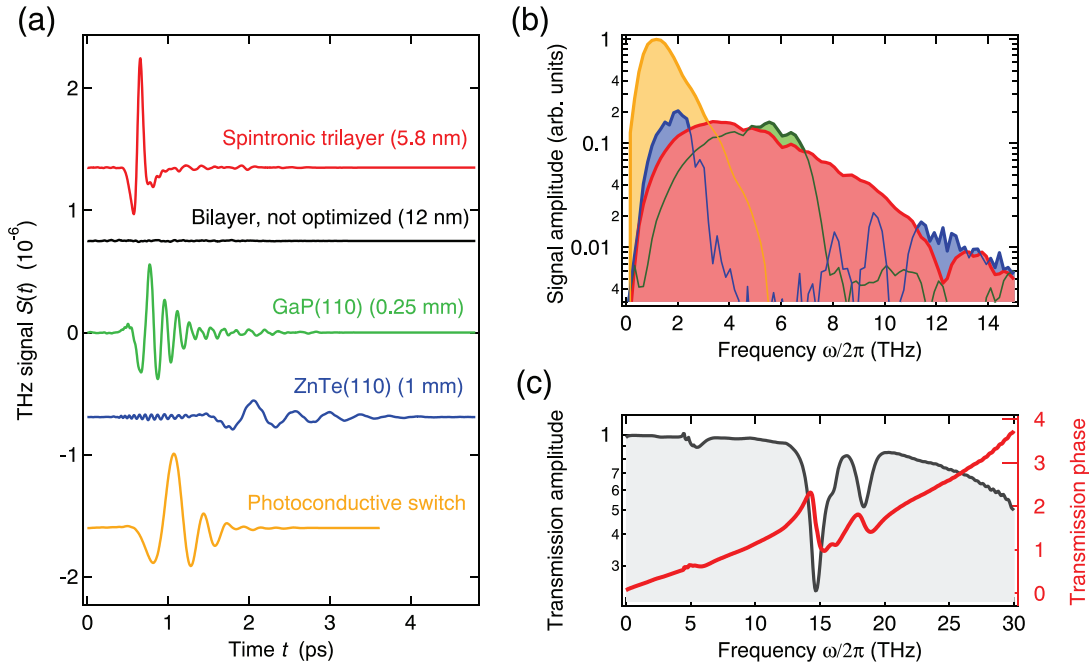


FIG. 7. Comparison of the emitted THz waveforms from different THz emitters under identical conditions in (a) the time domain and (b) the frequency domain employing a poled-polymer electro-optic detector. (c) Spectral amplitude and phase of the THz transmission of a 7.5- μm -thick polytetrafluoroethylene (PTFE) thread-seal tape measured with a spintronic emitter using a 10- μm -thick ZnTe electro-optic sensor. Reproduced with permission from Seifert *et al.*, Nat. Photonics 10, 483 (2016).⁶⁴ Copyright 2016 Macmillan Publishers Limited.

NM material (Fig. 8). Compared to FM|Pt, the THz-emission signals with opposite sign from FM|Ta and FM|W originated from their negative spin Hall angle γ as confirmed by *ab initio* calculations. Related works¹¹⁸ could confirm these results⁶⁴ and extended the NMs to Au,¹¹⁹ Ru,^{63,102} Al,¹²⁰ IrMn₃,¹²¹ and Mn₂Au.¹²²

An interesting approach to enhance the S2C amplitude is by alloying, which might enhance the skew-scattering contribution to the S2C significantly. Accordingly, Gueckstock and co-workers performed a systematic study that showed the profound impact that alloying layers at the FM/NM interface can have on the STE performance.¹¹⁵ To date, however, Pt-based STEs that rely on the ISHE yield the highest THz-emission amplitudes.

For the IREE-based STE, the range of the studied material systems is rather limited. To date, only Ag/Bi interfaces,^{117,123} monolayer MoS₂,¹²⁴ and the surface of Bi₂Se₃¹²⁵ were investigated. Fe|Ag|Bi and Co|Bi₂Se₃ stacks showed a THz-emission efficiency of about 1/5 of ZnTe.^{117,123,125} Previous DC-transport studies indicated a large potential in terms of S2C for IREE systems.¹²⁶ Exploiting this promising feature for STEs implies maximizing the Rashba S2C parameter λ_{IREE}

(see Sec. III B 2), which has the unit of length and is equivalent to the term $\lambda_{\text{NM}}\gamma\tanh(d_{\text{NM}}/2\lambda_{\text{NM}})$ for the ISHE-based STE [see Eq. (7)]. However, experiments could not yet demonstrate the expected large THz-emission efficiencies for IREE-based STEs, which might indicate limitations in terms of $t_{\text{FM}/\text{NM}}$. In another approach, THz emission from ISHE and IREE could be implemented in the same device. Adding the two respective THz signals could boost the efficiency to a much higher level in future STE designs.¹¹⁷

b. Metallic FM variation. The choice of the FM material for the STE was also investigated,⁶⁴ and the results indicated that Co, Fe, or the binary alloys containing Ni, Fe, or Co as the FM layer have much higher THz-emission efficiency than pure Ni [Fig. 9(a)]. The lower THz-emission efficiency of Ni-based STEs might be related to its lower Curie temperature or a lower value of $t_{\text{FM}/\text{NM}}$.⁹⁰ Related works studied the detailed composition of Co and Fe in (Co_xFe_{1-x})₈₀B₂₀|Pt structures and found that the THz-emission efficiency is maximized at $x = 0.1$ – 0.3 ^{66,104} or in the range of $x = 0.5$ – 0.8 for Co_xFe_{1-x}|Pt.¹²⁷ A temperature-dependent study found a minor impact on the

TABLE I. Typical frequency-averaged parameter values of optimized STEs [see Eq. (7)].^{64,115–117} To calculate the value of Z , typical values of $n_1 = 1$, $n_2 = 2.5$, and $G = 4$ S were assumed.

Parameter	A	F_{inc}	d_{NM}	d_{FM}	$t_{\text{FM}/\text{NM}}^0$	λ_{NM}	γ	Z
Typical value	0.5	$0.1 \frac{\text{mJ}}{\text{cm}^2}$	3 nm	3 nm	$10^{32} \frac{1}{\text{s m}^2}$	2 nm	0.1	50 Ω

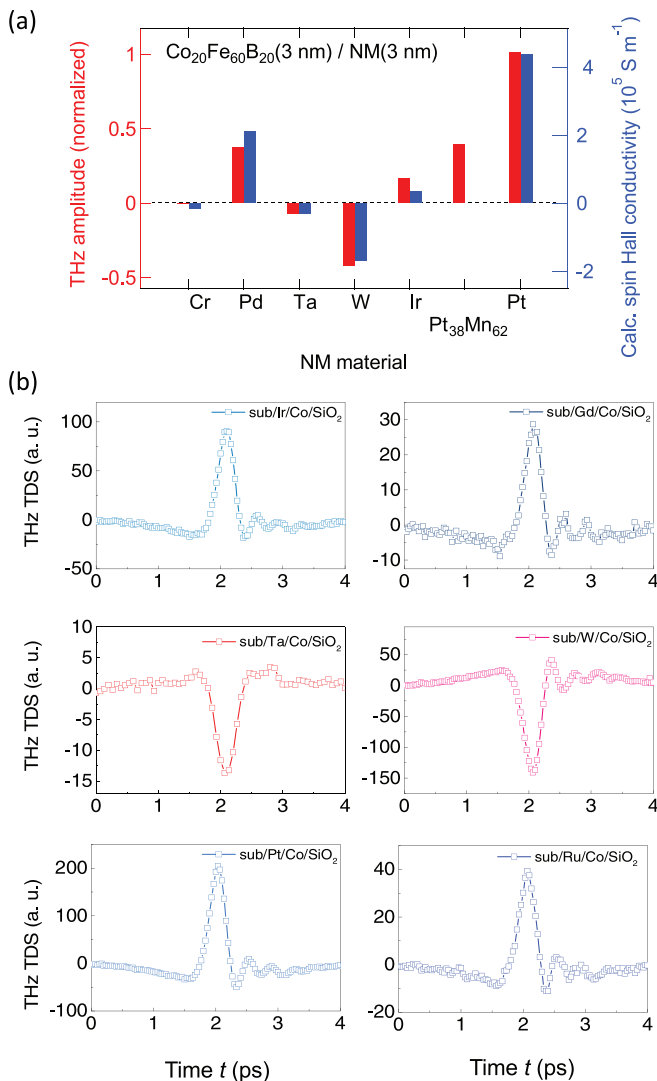


FIG. 8. Comparison of THz-emission amplitudes from FM|NM heterostructures based on the inverse spin Hall effect for different NMs. (a) THz emission from CoFeB|NM with different NMs (red, normalized to CoFeB|Pt) along with *ab initio* calculations of the spin Hall conductivity (blue). Reproduced with permission from Seifert *et al.*, Nat. Photonics **10**, 483 (2016).⁵⁴ Copyright 2016 Macmillan Publishers Limited. (b) THz emission from Co|NM with different NMs. The figures are adapted from Refs. 64 and 66. Reproduced with permission from Wu *et al.*, Adv. Mater. **29**, 1603031 (2017).⁶⁶ Copyright 2017 Wiley-VCH.

THz-emission amplitude in Co|Pt structures between 10 and 300 K,¹²⁸ consistent with a related study in Co|Mn₂Au.¹²² Among the studied samples, sputter-deposited Co₄₀Fe₄₀B₂₀ is the most efficient FM for the STE so far.

In the future, half-metallic FM systems such as Heusler alloys,^{129–132} which promise large spin polarizations, might help increase the STE performance. However, it should be noted that it is not only the value of the spin polarization alone that determines the STE performance but that also the change in magnetization per electronic-temperature increase has a major impact.¹¹⁵

c. Beyond ferromagnets. As an interesting alternative to FMs, ferrimagnets (FIMs),^{73,133–138} antiferromagnets,^{73,133,139–141} and magnetic insulators (MIs)^{88,142} were studied as the spin-current-generating magnetic layer in the STE. Seifert *et al.* compared the THz-emission signals from CoFeB|Pt and M|Pt samples, where M = DyCo₅, Gd₂₄Fe₇, Fe₃O₄, and FeRh [Figs. 9(b) and 9(c) and Ref. 133]. A parameter, defined as the figure of merit $FOM_M = \|t_{FM/NM}^0\|_M / \|t_{FM/NM}^0\|_{ref}$, was introduced to compare the spin current generated in a M|Pt bilayer with that of a CoFeB|Pt reference sample, for which $FOM_{CoFeB} = 1$ by definition. For the metallic FIM DyCo₅ and Gd₂₄Fe₇, the corresponding FOMs were extracted to be 0.85 and 0.79, respectively. These observations were ascribed to the reduced saturation magnetization due to the ferrimagnetic order. The spin polarization of DyCo₅, Gd₂₄Fe₇, and CoFeB is 40%, 36%, and 65%, respectively. Evidently, the FM order plays an important role in the performance of the STE.

Moreover, the experimental data of Fe₃O₄|Pt gave $FOM = 0.09$, which were attributed to a low value of $t_{FM/NM}^0$. These results suggest that low-conductivity magnetic materials are not suitable for efficient STEs, and it is essential for the spin-current-generating layer to have a metal-like conductivity. Furthermore, the THz emission from FeRh|Pt was found to be strongly temperature-dependent, which can be attributed to the antiferromagnet-to-FM phase transition of FeRh around room temperature.^{133,143} Although these alternative structures have not yet outperformed the FM-based STE, interesting functionalities might arise. For instance, it was shown that the spin current is governed by only one of the magnetic sublattices in an alloyed ferrimagnet such as GdFeCo.¹³⁴

d. Magnetic-insulator|NM heterostructures. The spin-current generation and injection in MI|NM bilayers are based on a different mechanism compared to the fully metallic STE. A typical example is a ferrimagnetic-insulator|NM (FII|NM) stack. Because of the insulating nature of the magnetic layer, no conduction electrons are available to carry a spin current from the FII to the NM. Instead, the spin transport is mediated by magnons inside the FII.

In the common DC version of the so-called spin Seebeck effect (SSE), a temperature gradient throughout the FII|NM sample leads to a magnon accumulation at the FII/NM interface. In contrast, the ultrafast SSE relies on the selective heating of the NM layer by the femto-second laser pulse creating a step-like temperature profile across the FII/NM interface. The ultrafast increase in the electronic temperature in the NM layer leads to an enhanced scattering of NM electrons off the interface. During these scattering events, the NM electrons couple to the magnetic order in the FII through the interfacial exchange interaction. Through a second-order effect, the pump-induced enhanced exchange-scattering noise inside the NM gets rectified, giving rise to a net \mathbf{j}_s . The spin current is polarized along the FII magnetization and flows perpendicular to the interface.

The ultrafast SSE was experimentally observed in ferrimagnetic YIG|Pt bilayers by means of optical pump-probe spectroscopy with ~ 1 ps time resolution¹⁴⁴ and THz emission spectroscopy with 27 fs resolution,⁸⁸ confirmed for different YIG thicknesses¹¹⁴ and extended to low temperatures.¹⁴⁵ The observed THz emission signal was of magnetic origin and related to a spin current inside the NM followed by the ISHE [see Fig. 10 and Eq. (3)]. Notably, the ultrafast SSE can coexist with the spin-voltage-driven spin transport in magnetic materials such as the half-metallic ferrimagnet Fe₃O₄ (magnetite).¹¹⁴ In the

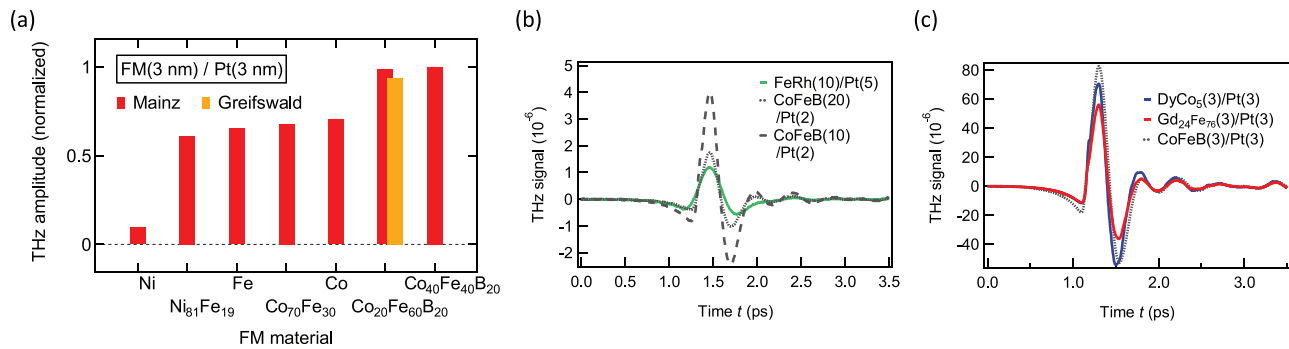


FIG. 9. Comparison of THz-emission amplitudes from FM|NM heterostructures based on the inverse spin Hall effect for different FMs. (a) THz emission from FM|Pt with different FM materials grown in different labs. Reproduced with permission from Seifert *et al.*, Nat. Photonics **10**, 483 (2016).⁶⁴ Copyright 2016 Macmillan Publishers Limited. (b) and (c) THz emission from heterostructures containing complex magnets such as DyCo₅, Gd₂₄Fe₇₆, or FeRh and Pt as the ISHE material. The figures are adapted from Refs. 64 and 133. Reproduced with permission from Seifert *et al.*, Spin **7**, 1740010 (2017).¹³³ Copyright 2017 Authors, licensed under a Creative Commons Attribution (CC BY) license.

future, the ultrafast SSE may provide an all-optical access to study magnons in FIIs and enable the generation of ultrafast spin currents using MIs.¹⁴⁶ In terms of THz-emission strength, the SSE scenario in FII|NM samples is about 2 to 3 orders of magnitude less efficient than the spin-voltage mechanism in all-metallic structures.

Another progress in this field is the THz generation in antiferromagnet insulator|NM (AFMI|NM) stacks such as NiO|Pt bilayers.¹³⁹ In the case of NiO, the spin-current generation was ascribed to the

inverse Faraday effect.⁴⁸ Although its efficiency is much lower than for the fully metallic STEs, this approach demonstrates the potential of AFMIs and AFMs, in general, in THz spintronics.^{147,148}

4. Thickness optimization

The thickness of each layer in the STE's FM|NM heterostructure can strongly affect the THz-emission efficiency. According to Eq. (7),

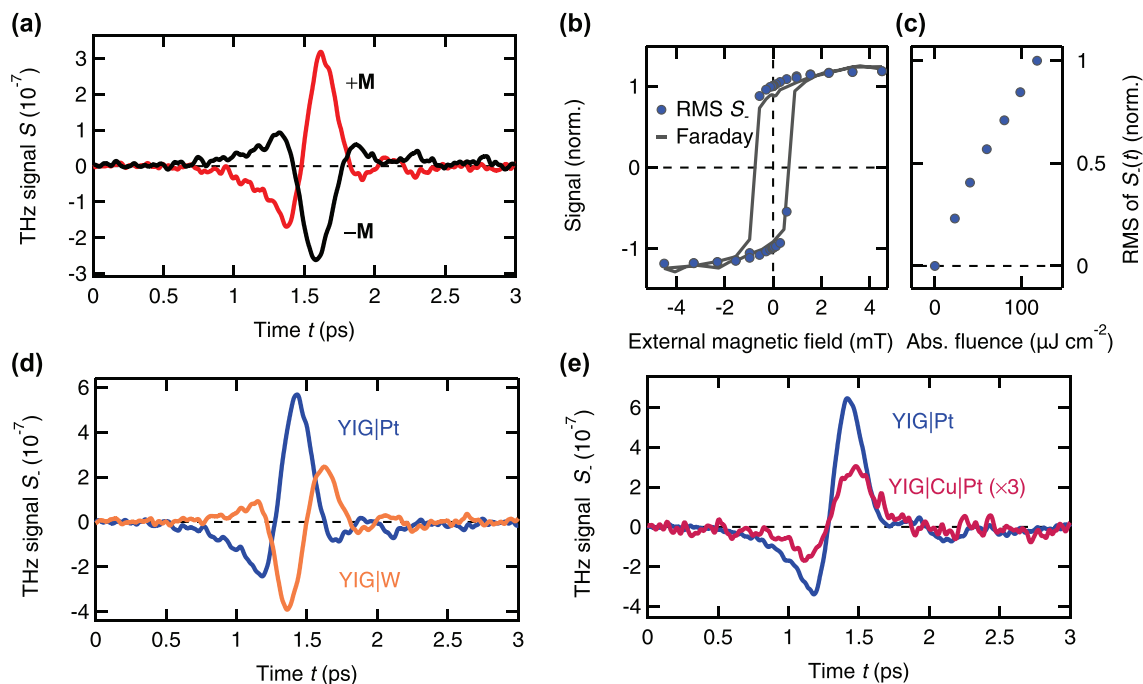


FIG. 10. THz emission of photoexcited YIG|Pt heterostructures. (a) THz emission signals $S(t, \pm M)$ from a YIG|Pt sample in opposite directions of the in-plane YIG magnetization M as a function of time t . (b) Amplitude of the THz signal $S_{-}(t) = S(t, M) - S(t, -M)$ and the Faraday rotation of a continuous-wave laser beam (wavelength 532 nm) as a function of the external magnetic field. (c) Fluence dependence of the THz-emission amplitude. (d) THz-emission signal from YIG samples capped with Pt and W. (e) THz emission signal from a YIG film capped with Pt or Cu|Pt. This figure is adapted from Ref. 88. Reproduced with permission from Seifert *et al.*, Nat. Commun. **9**, 2899 (2018). Copyright 2018 Authors, licensed under a Creative Commons Attribution (CC BY) license.

d_{NM} and d_{FM} should be optimized with respect to the factor A , the total metal-film conductance $G = \int_0^{d_{\text{NM}}+d_{\text{FM}}} dz \sigma(z)$, and the spin-current-propagation factor $\lambda_{\text{NM}} \tanh(d_{\text{NM}}/2\lambda_{\text{NM}})$.

Notably, the factor A was found to be approximately independent of d_{NM} and d_{FM} for total thicknesses in the range from 3 to 10 nm.⁶⁴ For larger thicknesses, however, A follows an exponential decay plus an offset.^{64,105}

In terms of the spin-current propagation, the factor $\tanh(d_{\text{NM}}/2\lambda_{\text{NM}})$ in Eq. (7) can minimize the THz-emission amplitude significantly if $d_{\text{NM}} < \lambda_{\text{NM}}$, because multiple spin-current reflections superimpose destructively.¹⁰⁵ If, on the other hand, $d_{\text{NM}} \gg \lambda_{\text{NM}}$, the spin current has decayed before reaching the NM/substrate or NM/air boundary. Thus, an optimal total thickness for the STE exists, which maximizes the THz-emission amplitude. One can estimate that this optimum occurs at $d_{\text{NM}} \sim 2\lambda_{\text{NM}}$, which corresponds to the peak in Fig. 11(a). Therefore, the optimized thickness of the NM layer should be about 2 nm for Pt ($\lambda_{\text{NM}} = 1$ nm) and in a similar range for other NMs with large S2C efficiency.^{64,105,106}

Importantly, for DC spin currents, λ_{NM} in Eq. (7) equals the spin-diffusion length but is expected to be shortened and approach the electron mean free-path length λ_{mfp} for THz spin transport (TST).⁹² This prediction was confirmed recently by measuring spin-current propagation lengths at GHz and THz rates, revealing a fourfold reduction in the case of THz spin currents.¹⁴⁹ Accordingly, the measured spin-diffusion length of Pt is of the order of 10 nm,¹⁵⁰ whereas, for THz spin currents, a λ_{NM} of approximately 1 nm was found for thin Pt films. Similar values were reported for other ISHE materials.^{91,105} This observation allows one to draw the following important conclusion: For the typical metallic thin films used in STEs, the THz conductivity is well described by the Drude model¹⁵¹ and one often finds to a good approximation $\sigma_{\text{NM}}(\omega) \approx \sigma_{\text{NM}}(\omega = 0)$ due to the small Drude scattering time τ in the range of a few femtoseconds.^{79,105} This fact directly allows us to conclude that $\sigma_{\text{NM}} \propto \tau \propto \lambda_{\text{mfp}} = \lambda_{\text{NM}}$.

We can go one step further and address the impact of the above argumentation on the emitted THz electric field E . For typical NM film thicknesses of a few nanometers, one typically has $\tanh(d_{\text{NM}}/2\lambda_{\text{NM}}) \approx 1$ and $n_1 + n_2 + Z_0 G \approx n_1 + n_2$. Equation (7), thus, implies that

$E \propto \sigma_{\text{NM}} \gamma$. In other words, E is rather proportional to the spin Hall conductivity $\sigma_{\text{NM}}^{\text{SH}} = \sigma_{\text{NM}} \gamma$ and not to the spin Hall angle γ .^{64,91} Note that this simple relationship only holds in cubic/isotropic materials such as the typical solids used for STEs. It might be more complex otherwise.¹⁵²

Similar to d_{NM} , the thickness of the FM layer has a profound impact on the THz-emission amplitude, too [Fig. 11(b)].¹¹⁷ Here, a critical minimal thickness d_0 of typically about 0.5 nm exists, below which the magnetic order of the FM layer is not well established,¹⁵³ as also confirmed by recent magneto-optical Kerr effect (MOKE) studies.⁶⁵ The impact of this magnetically dead layer can be described by a correction factor $\tanh[(d_{\text{NM}} - d_0)/\lambda_{\text{FM}}]$ to Eq. (7),⁹⁵ where λ_{FM} accounts for the finite spin propagation length inside the FM layer, i.e., the region of the FM that still contributes to the spin current into the NM as shown in Fig. 2.⁹⁵ Based on recent studies,^{64–66,106,154} the optimized thickness of the FM layer (FM = Ni, Co, Fe, and CoFeB) is 1–4 nm.

To summarize, thickness-dependent studies of the FM and the NM layers show that the THz spin and charge currents are localized within a few nanometers at the FM/NM interface and can, thus, be considered interfacial probes of ultrafast charge and spin dynamics.

5. Crystal structure, degree of disorder, and alloying

Controlling the atomic arrangement and degree of disorder in the bulk and at the interfaces of the STE's metallic layers might optimize the generation (j_s^0), the injection efficiency ($t_{\text{FM}/\text{NM}}$), and propagation (λ_{NM}) of the spin current as well as the two parameters A and Z .

Li and co-workers systematically studied the influence of interfacial roughness, crystal structure, and interface intermixing on the THz emission from polycrystalline Co/Pt heterostructures.¹⁰⁰ As shown in Fig. 12(a), the THz emission based on the spin voltage (see Secs. II A and III A 3) strongly depended on the interfacial roughness and could be largely improved by increasing the smoothness of the samples. A possible reason is that the spin-flip probability is strongly correlated with the roughness and/or other types of disorder at the Co/Pt

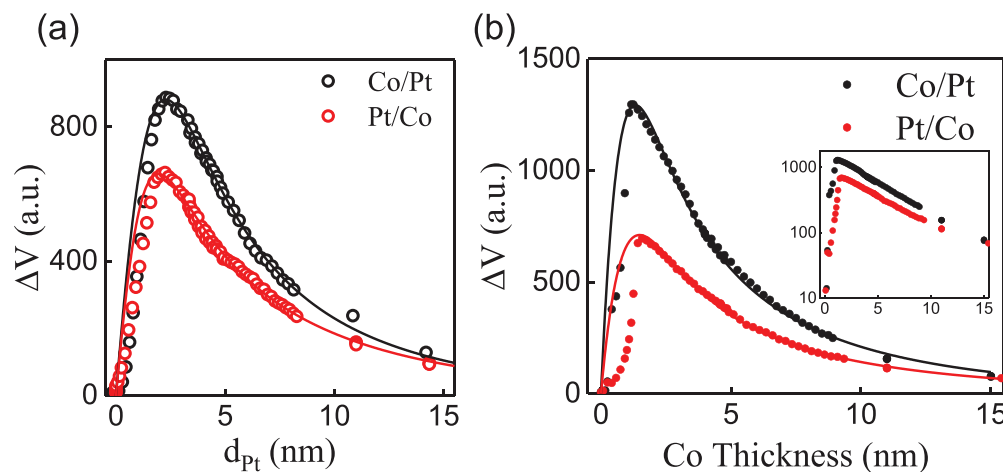


FIG. 11. Dependence of the THz-emission amplitude on the thickness of (a) the NM and (b) the FM layers under identical experimental conditions. Reproduced with permission from Zhou *et al.*, Phys. Rev. Lett. 121, 086801 (2018).¹¹⁷ Copyright 2018 American Physical Society.

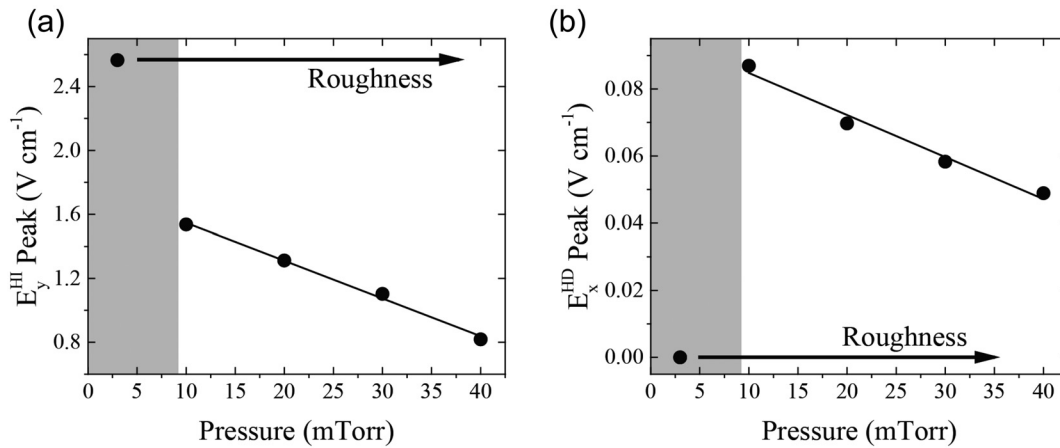


FIG. 12. Influence of the interfacial roughness on (a) the spin-voltage-based and (b) the THz-emission amplitude related to the inverse spin-orbit torque from Co|Pt. Reproduced with permission from Li *et al.*, Phys. Rev. Mater. **3**, 084415 (2019).¹⁰⁰ Copyright 2019 American Physical Society.

interface. It is argued that more disorder decreases the spin-current injection across the Co/Pt interface, i.e., the factor $t_{\text{FM}/\text{NM}}$ in Eq. (7).

However, the same study reported that the THz-emission based on the inverse spin-orbit torque [ISOT, Fig. 12(b), see Sec. II C 10] increases first within the same sample for small degrees of disorder before also decreasing for larger disorder. This surprising behavior might be attributed to the interfacial origin of the ISOT mechanism. In contrast, an intermixing layer at the interface, i.e., a $\text{Co}_x\text{Pt}_{1-x}$ spacer layer, resulted in an increased spin-voltage-driven THz-emission amplitude. At least two explanations are possible: (i) The $\text{Co}_x\text{Pt}_{1-x}$ alloy layer increases t , or (ii) an additional IREE arises from the $\text{Co}_x\text{Pt}_{1-x}$ alloy layer. Clearly, these results indicate that a suitable interlayer can control the THz-emission efficiency. Similar conclusions were reached in a related work¹⁰³ that reported a strong impact of electron-defect scattering on the STE performance by comparing Fe|Pt bilayers with different degrees of crystallinity. Along these lines, epitaxial Fe films were shown to exhibit an anisotropic THz emission performance, which was attributed to strain-induced effects.¹⁵⁵

Another work revealed the strong influence that interfacial intermixing can have on the STE performance.¹¹⁵ In this study, Gueckstock *et al.* showed that the order, in which the layers are deposited during the sputtering growth process, determines the sign and magnitude of the S2C in the interfacial alloy layer (see Sec. III B).

The crystal structure of FM|NM layers can be further optimized by post-annealing of sputter-grown STEs.^{94,104} As seen in Fig. 13, in CoFeB|Ta heterostructures,⁹⁴ the annealing process had different effects: (a) B atoms diffused into the Ta layer, which enlarged the S2C efficiency and decreased λ_{NM} . (b) An enhanced crystallization of the CoFeB layer can have a profound impact on the electronic transport properties. (c) The saturation magnetization decreased because of the atomic mixing between CoFeB and Ta, which diminished j_s^0 and likely also impacted $t_{\text{FM}/\text{NM}}$. The best THz-emission performance for CoFeB|Ta bilayers was found for post-growth annealing at 300 °C for 1 h. On the other hand, THz emission from $\text{Co}_x\text{Fe}_{1-x}\text{B}_{20}$ |Ta bilayers¹⁰⁴ was reported to peak for $x = 0.2$ and an annealing temperature of ~ 400 °C, where the largest saturation magnetization of CoFeB was obtained.

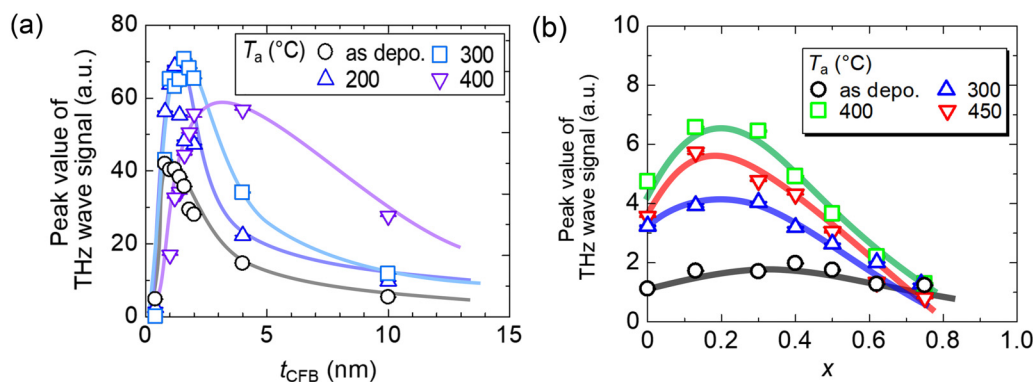


FIG. 13. Effect of post-growth annealing on the THz emission from spintronic THz emitters based on (a) CoFeB|Ta for different CoFeB thicknesses t_{CFB} and (b) $(\text{Co}_x\text{Fe}_{1-x})_{80}\text{B}_{20}$ |Ta for varying Co-content x . Reproduced with permission from Sasaki *et al.*, Phys. Rev. B **100**, 140406(R) (2019).¹⁰⁴ Copyright 2019 American Physical Society. Reproduced with permission from Appl. Phys. Lett. **111**, 102401 (2017).⁹⁴ Copyright 2017 AIP Publishing.

6. Impact of the substrate

According to Eq. (7), the substrate, on which the STE is grown, impacts the THz emission directly through the impedance Z . Thus, spectral features in the THz emission can arise, which are footprints of the substrate phonon modes.⁷⁵ Moreover, the substrate material influences the heat transport away from the excited STE area, which becomes an important factor for STE excitation conditions with large pump powers.^{69,70,156}

7. Pump-pulse characteristics

Specific pump-pulse parameters, such as the wavelength, the duration, and the temporal structure, play a critical role for the STE performance. A major experimental challenge for wavelength-dependent studies is to maintain a constant pump-pulse duration for different center wavelengths. Following this approach, for 1550, 800, and 400 nm center wavelengths^{68,157} and for the range 900–1500 nm,⁶⁷ it was found that the structure of the emitted THz pulse is independent of the pump wavelength. The majority of the recent studies^{67,68} also showed that the THz-emission efficiency is largely independent of the pump wavelength. One exception from this behavior¹⁰⁹ was ascribed to a wavelength-dependent absorptance of the STE.

From a fundamental-science viewpoint, the pump-wavelength independence of STEs is fully consistent with the spin-transport model of Eq. (8). From an applied viewpoint, it is distinctly different from conventional THz sources, e.g., nonlinear optical crystals and semiconductor-based THz emitters. It enables the integration of the STE into systems driven by low-cost and compact femtosecond fiber lasers without loss of efficiency.

Regarding the pump-pulse duration $\Delta t_{\text{pump}}^{50\%}$ (defined as the full-width at 50% maximum of the intensity envelope), experimental results^{64,69,158} suggest that the product of THz bandwidth $\Delta\omega_{\text{THz}}^{10\%}/2\pi$ of the focused THz beam at the detector position (see Figs. 3 and 4) and $\Delta t_{\text{pump}}^{50\%}$ is a constant that is given by

$$\frac{\Delta t_{\text{pump}}^{50\%} \Delta\omega_{\text{THz}}^{10\%}}{2\pi} \approx 0.5. \quad (9)$$

Here, $\Delta\omega_{\text{THz}}^{10\%}$ is defined as the full width at 10% of the maximal THz electric field amplitude in the spectral domain. The temporal structure of the pump pulse is a further degree of freedom that allows one to generate double or multiple THz pulses by exciting the STE with multiple time-delayed pump pulses.^{111,159}

8. Impact of nano- and microstructuring

Aside from FM|NM bilayer structures, more complex STE designs were proposed to increase the THz-emission efficiency. The basic idea is to gain more THz electric field per pump-pulse energy by (a) increasing j_s and the S2C strength and by (b) better photonic management, which includes an optimized pump-pulse absorption (absorptance A) and a maximized current-to-electric-field conversion efficiency (THz impedance Z).

Regarding (a), a promising approach utilizes trilayer STEs with the structure NM₁|FM|NM₂ [Fig. 14(a)].^{64,69,101} This design takes advantage of the spin currents in the forward as well as the backward propagation direction. Accordingly, the spin currents injected into NM₁ (j_s^1) and

NM₂ (j_s^2) have opposite directions. To avoid cancelation of the net charge current (the sum of j_c^1 and j_c^2 inside the two NM layers), the spin Hall angles of NM₁ and NM₂ should have opposite signs [see Eq. (3)]. Indeed, measurements of the THz-emission amplitude from W|Co₄₀Fe₄₀B₂₀|Pt trilayers revealed a doubling as compared to the Co₄₀Fe₄₀B₂₀|Pt bilayer counterpart with identical total thickness.⁶⁴

Regarding (b), to increase A at the expense of Z , a multilayer structure [Pt|Fe|MgO]_{*n*} was fabricated [Fig. 14(b)],⁶⁵ where n is the index number of the repeating periods. A 2-nm-thick MgO layer is used to isolate the spin current from the neighboring Fe layers to avoid spin-current leakage into the adjacent bilayer unit. The maximum THz-emission amplitude was found for $n = 3$ with a ~70% increase in the THz-emission amplitude in comparison to $n = 1$ for the bilayer STE. For $n > 3$, the lowered excitation density and the increased STE impedance can decrease the THz-emission efficiency.¹⁶⁰

The two approaches of STE stacking and trilayer design were combined for trilayer STEs based on W|Fe|Pt films. In Ref. 101, the THz emission from (W/Fe/Pt/SiO₂)_{*n*} periodic structures for $n = 1, 2, 3$ with different SiO₂ thicknesses (d_{SiO_2}) was measured. Controlling d_{SiO_2} allows for the increase in the incident light absorption via minimizing the reflection at interfaces/surfaces and, hence, boosts the THz-emission efficiency. An optimized structure with $n = 2$ and $d = 100$ nm could be identified with a corresponding efficiency that is ~1.7 times larger than that for $n = 1$.

Note, however, that the best stacking index n depends on the individual NM and FM layer properties and, thus, not for each trilayer STE, $n > 1$ results in an increased THz-emission amplitude (unpublished results by some of the authors).

The study of Ref. 111 arranged multiple STEs in one setup and superimposed the two emitted THz pulses. By controlling the external magnetic fields for the two STEs separately, the polarization of the combined THz pulse could be tuned. Clearly, the two THz emitters placed in a cascaded geometry can optimize the THz-generation efficiency by recycling the otherwise wasted reflected or transmitted fraction of the pump pulse. Similarly, future approaches might aim at combining the THz pulses already from a single STE that are propagating in forward and backward directions with respect to the pump pulse propagation direction, which would increase the THz-emission amplitude by a factor of 2.

Spin-valve or synthetic antiferromagnet designs, that is, samples that contain multiple, possibly magnetically coupled FM layers with different magnetic properties, such as coercive field strength, offer another promising approach to enrich the STE functionality.^{161–163} Alternatively, magnetic tunnel junctions can also be used to tune the spintronic THz emission.¹⁶⁴

Additionally, some works reported THz emission from microstructured FM|NM samples (also see Sec. V C), which allow one to control the emitted THz pulse shape and polarization to some extent.^{65,71,72,165–168}

9. Upscaling of the STE

A major advantage of STEs over conventional THz sources is their straightforward scalability. Accordingly, an optimized large-area trilayer STE with a diameter of 7.5 cm was able to generate strong THz pulses, the peak electric field of which reached ~300 kV cm⁻¹ at a bandwidth of ~15 THz upon pumping with 60 fs pulses with an

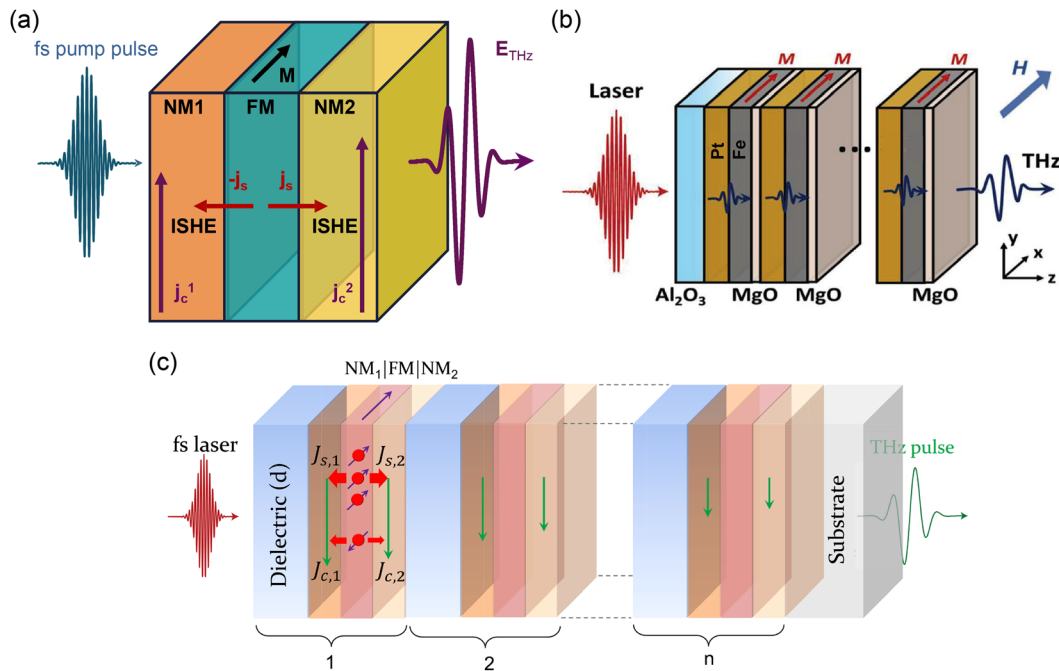


FIG. 14. Structure design concepts for the spintronic THz emitter. (a) $NM_1|FM|NM_2$ structure, where NM_1 and NM_2 have spin Hall angles of opposite signs. Reproduced with permission from Seifert *et al.*, *Appl. Phys. Lett.* **110**, 252402 (2017).⁶⁹ Copyright 2017 AIP Publishing. (b) $[Pt|Fe|MgO]_n$ periodic structure. Reproduced with permission from Yang *et al.*, *Adv. Opt. Mater.* **4**, 1944–1949 (2016).⁶⁵ Copyright 2016 Wiley-VCH. (c) $[W|Fe|Pt|SiO_2]_n$ periodic structure. Reproduced with permission from Feng *et al.*, *Adv. Opt. Mater.* **6**, 1800965 (2018).¹⁰¹ Copyright 2018 Wiley-VCH.

energy of 5 mJ) and a center wavelength of 800 nm.⁶⁹ This THz electric field strength allowed one to measure the THz Kerr effect in diamond, which scales quadratically with the THz electric field. Future improvements in the STE THz-emission efficiency are, therefore, foreseen to enable studying THz strong-field phenomena over a wide THz frequency range.¹⁶⁹

10. THz emission induced by other spintronic phenomena

a. Spin-to-charge current conversion inside the magnetic material. Aside from the ISHE and the IREE inside the NM, S2C can also occur inside the magnetic material such as in single-layer FM samples^{170–172} but also in the FM|NM heterostructure. In single-layer FM samples, the out-of-plane spin current may arise from pump-induced gradients or structural gradients inside the sample. Accordingly, the measured THz-emission signals from MgO|FM|quartz [FM = $(Fe_{0.8}Mn_{0.2})_{0.67}Pt_{0.33}$, $Fe_{0.8}Mn_{0.2}$, $Co_{0.2}Fe_{0.6}B_{0.2}$, and $Ni_{0.8}Fe_{0.2}$] were shown to follow the same symmetries as the ISHE-based STE [Eq. (3)]. However, the THz generation efficiencies for STEs based on S2C inside the FM are far below state-of-the-art STEs based on the ISHE inside NMs such as Pt.

b. Magnetic dipole radiation. According to Eq. (2), a time-varying magnetization can emit a THz pulse.⁴⁴ Recent studies found that the corresponding magnetic-dipole radiation from metallic FM layers of a few nanometers thickness is on the order of 1% of the electric dipole

radiation obtained from optimized STEs under identical excitation conditions.¹¹³ However, care needs to be taken when assigning electric- or magnetic-dipole characteristics to the observed THz-emission signal.⁴⁴ It requires a thorough symmetry analysis of the THz signal as shown recently.^{46,113}

c. Inverse spin-orbit torque. Apart from spin voltage, THz emission triggered by the inverse spin-orbit torque (ISOT) was reported in Co|Pt¹⁷³ and CoFeB|Ag|Bi¹²³ thin films under excitation with circularly polarized femtosecond laser pulses as well as for NiO|Pt¹³⁹ thin films with linearly polarized pump pulses. It was suggested that the optical spin transfer torque, which is also known as the inverse Faraday effect, leads to a tilting of the in-plane magnetic order in the sample plane.^{48,173}

For Co|Pt¹⁷³ and CoFeB|Ag|Bi,¹²³ the subsequent magnetization dynamics injects a spin current polarized perpendicular to \mathbf{M} into the adjacent NM, e.g., NM = Pt. The spin current is converted via the ISHE inside the NM into a transverse charge current. This ISOT-based THz generation is phenomenologically described by¹⁷³

$$\mathbf{E}_{THz} \propto \mathbf{j}_c \propto \mathbf{n} \times [\mathbf{M} \times \boldsymbol{\sigma}]I, \quad (10)$$

where \mathbf{n} is the unit vector normal to the thin-film stack, $\boldsymbol{\sigma}$ is the axial unit vector pointing parallel or antiparallel to the propagation of the circularly polarized light and I is the pump-pulse intensity envelope. Importantly, the polarization of \mathbf{E}_{THz} for the ISOT mechanism is oriented along \mathbf{M} in the studied FMs, whereas it is perpendicular to \mathbf{M} for the spin-voltage mechanism [see Eq. (3)].

Thus, upon illuminating a Co|Pt heterostructure with circularly polarized femtosecond laser pulses, two THz emission signals can be detected with different polarizations, perpendicular and parallel to \mathbf{M} [Fig. 15(a)]. The ISOT-component follows Eq. (10), where the sign change of \mathbf{M} and σ leads to the reversal of the emitted THz waveform [Fig. 15(b)]. In CoFeB|Ag|Bi, a similar dependence of the emitted THz signal was observed.¹²³ However, the measured THz-signal amplitudes from the ISOT effect strongly depend on the sample preparation processes, and the ISOT-based THz emission tends to decrease with lower FM/NM interface quality; the ISOT-based structures were found to

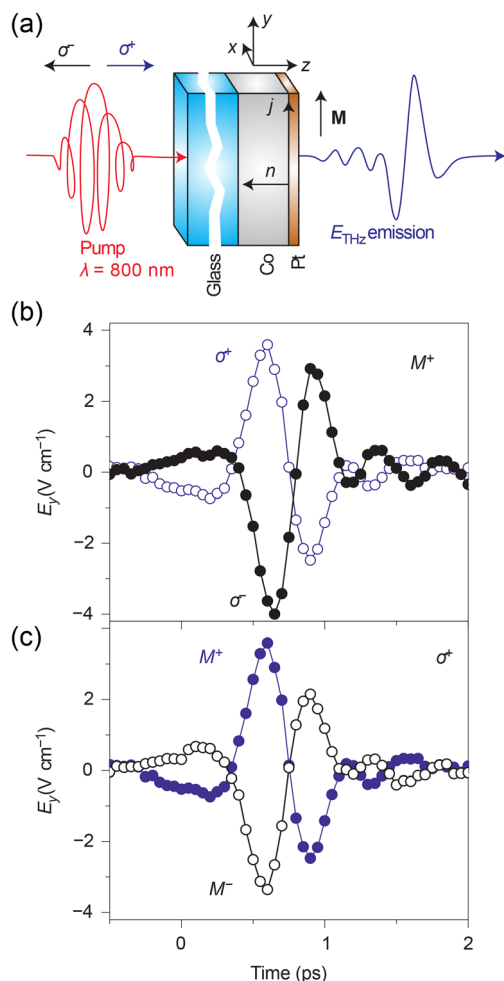


FIG. 15. THz emission from Co|Pt based on the inverse spin-orbit torque. (a) Upon exciting a FM|NM thin film heterostructure with circularly polarized optical femtosecond laser pulses, the optical spin transfer torque tilts the in-plane magnetization \mathbf{M} of the FM layer. Subsequently, the magnetization relaxes back to its equilibrium orientation thereby emitting a spin current into the adjacent Pt layer. The inverse spin Hall effect converts the spin current into an in-plane charge current that radiates at THz frequencies. The polarization of the THz pulse is along \mathbf{M} . (b) THz emission signals for the two pump helicities ($\sigma^{+,-}$) at constant \mathbf{M} . (c) THz emission signal for opposite \mathbf{M} at constant pump helicity. The figures are adapted from Ref. 173. Reproduced with permission from Huisman *et al.*, Nat. Nanotechnol. 11, 455–458 (2016). Copyright 2016 Macmillan Publishers Limited.

deliver THz amplitudes that were significantly smaller than from spin-voltage driven STEs.^{123,173}

As of now, none of the above alternative spintronic phenomena are competitive with respect to STEs based on the ISHE S2C and the transient spin voltage.

In conclusion, all these efforts demonstrate that different designs of the STE can greatly enhance its efficiency as well as functionality and promote this type of the THz emitter toward real-world applications.

III. CURRENT UNDERSTANDING OF ELEMENTARY PROCESSES IN THE STE

The physical phenomena in STEs are manifold, including ultrafast spin dynamics, transport, and S2C mechanisms. To improve the STE performance, the understanding of the microscopic mechanisms behind these different steps is mandatory, including a tailoring of the STE to specific applications. We focus on STEs that can be described according to the four processes introduced in Secs. II A and II C 2. In the following, we address processes (1)–(3) in more detail.

A. Processes (1) and (2): Laser-driven spin-current generation

At frequencies much smaller than ~ 1 THz, it is known that spin currents can be triggered by gradients of the electrostatic potential (leading to longitudinal spin-polarized electron currents in metallic ferromagnets or transverse spin currents by the ISHE in metals), by temperature gradients (e.g., leading to longitudinal spin-polarized electron currents in metals due to the spin-dependent Seebeck effect or magnon currents due to the spin Seebeck effect), and by gradients of the spin voltage.⁸⁷ The latter is also known as spin accumulation and quantifies the excess (or lack) of the local spin density, i.e., how far the local spin density of a solid is away from its equilibrium value. Transferring these concepts to THz frequencies and nonthermal states is a matter of current research.^{88,113,174}

The spin-current generation and injection triggered by femtosecond laser pulses in FM|NM heterostructures are strongly related to the ultrafast spin and carrier dynamics in the FM layer. Usually, in a NM (e.g., Au or Ag), the ultrafast relaxation of photoexcited electrons happens on a sub-picosecond timescale, and the associated dynamics of energy transfer from the orbital degrees of freedom to the phonons can be described by the two-temperature model (2TM).^{175,176} In the limit of weak excitation, the 2TM can be extended to nonthermal electron distributions.^{113,174} In a FM, however, ultrafast optical excitation, additionally, causes a magnetization quenching on a sub-picosecond timescale, as first observed by Beaurepaire *et al.*¹⁷⁷

In thin single FM films on insulating substrates, such ultrafast demagnetization (UDM) is governed by transfer of spin angular momentum from the electrons to the crystal lattice.^{44,45,177–198} In FM films thicker than the penetration depth of the pump or in stacks such as FM|NM, an additional channel for the transfer of spin out of the excited FM regions becomes possible: ultrafast or THz spin transport as visualized in Fig. 16.^{98,199–208}

1. Superdiffusive spin transport

To simulate the ultrafast electron spin current following ultrafast laser excitation, Battiato *et al.* developed a semiclassical model of spin transport.^{98,199} Their theory captures both ballistic transport (without

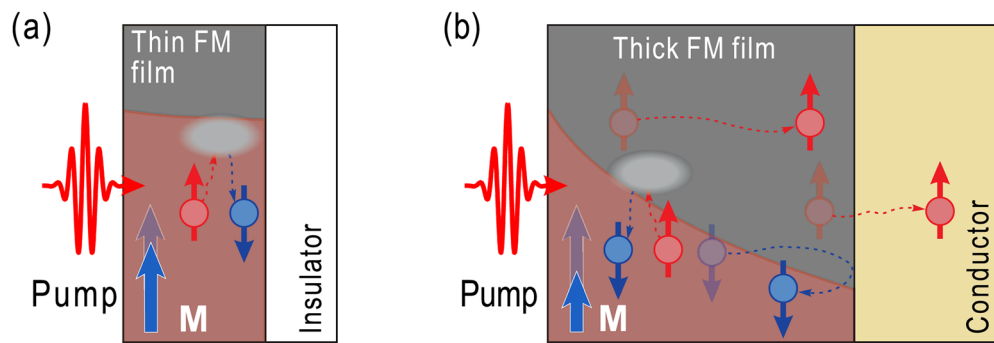


FIG. 16. Schematics of femtosecond laser excitation of (a) a thin FM on an insulating substrate, allowing for local relaxation processes only, and (b) a thick FM on a conducting substrate, where both local and nonlocal relaxation processes are present. The red-shaded areas represent the in-depth profiles of the laser excitation. Reproduced with permission from Rzdolski *et al.*, *J. Phys.* **29**, 174002 (2017).²⁰⁰ Copyright 2017 IOP Publishing Ltd.

electron scattering) and diffusive transport (dominated by electron scattering), which are summarized as superdiffusive spin transport (SDST).^{201,208} The simulations based on the SDST model are supported by different ultrafast optical studies,^{202–207} although some experimental results indicated that other mechanisms might significantly contribute, too.^{209–213}

In the SDST model, rate equations describe the spin-dependent carrier population, together with the *ab initio* input of the carrier velocities and lifetimes. Several events are taken into consideration, including multiple spin-conserving electron collisions and inelastic electronic scattering cascades.⁹⁸ In 3D transition FMs, the photoexcited sp electrons generally have higher band velocities and longer lifetimes than the excited minority spins. According to the SDST model, this effect leads to spin transport out of the FM layer upon optical excitation.

SDST simulations could reproduce the qualitative shape of the spin-current dynamics and its order of magnitude in Fe|Au and Fe|Ru bilayers.⁶³ However, the connection of the SDST model to spintronic concepts, such as gradients of spin voltage, temperature, or electrostatic potential, is not obvious and was addressed only recently.¹¹³

In addition to the superdiffusion model, a particle-in-cell simulation based on the Boltzmann equation was developed recently.²¹⁴ It can reproduce the superdiffusive transport behavior and also captures the main physical process of the laser-induced demagnetization.

2. Optically induced intersite spin transfer

The term optically induced intersite spin transfer (OISTR) refers to a coherent, ultrafast spin transfer between two subsystems that occurs while the pump laser pulse overlaps with the sample.²¹⁵ It was predicted by *ab initio* calculations and experimentally observed in Ni|Pt bilayers.²¹⁶ As typical spin-current dynamics in STEs were found to strongly exceed the pump pulse duration,^{113,114,116} OISTR is considered to make a minor contribution in the THz-emission process. However, the experimental observation of the OISTR effect using THz spectroscopy poses a highly interesting challenge for future studies.

3. Spin voltage as a major spin-current driving force

In general, three different forces can drive a spin current:^{87,217} gradients of the electrostatic potential, the temperature, or the spin voltage. The latter is also known as the spin accumulation and

quantifies how much the local spin density deviates from its instantaneous equilibrium value. A transient spin voltage was experimentally observed in laser-excited single Fe films.²¹⁸ Moreover, theoretical works suggest that the spin voltage has a major impact on driving ultrafast demagnetization in FMs.^{89,219,220}

Recently, Rouzegar *et al.*¹¹³ made an experimental and theoretical effort to connect ultrafast spin transport and ultrafast demagnetization to differences or gradients of macroscopic parameters such as temperature and spin voltage. To this end, they used Boltzmann-type rate equations and the Stoner model, which describes the electronic structure of a typical 3d FM qualitatively well. The magnetization M shifts the energy of spin-up and spin-down electronic Bloch states by an energy proportional to $|M|$ with respect to each other (see Fig. 17).

When such a FM is excited by an ultrafast optical pulse, the chemical potential of each spin channel changes differently. As a result, a splitting of the spin chemical potential, an ultrafast spin voltage, arises.^{89,217,219} The relaxation of this spin voltage, which causes a magnetization quenching, can occur via local spin-flip scattering events inside the FM or via spin transport into an adjacent layer (Ref. 113 and Fig. 17), which is also consistent with observations in other THz emission studies.²²¹ It is noteworthy that the concepts of temperature, chemical potential, and, thus, spin voltage can be extended to nonthermal electron distributions that prevail directly after optical excitation.^{113,222}

Experimental evidence that spin currents and demagnetization evolve with the same ultrafast dynamics in FM|NM heterostructures was found directly in THz emission studies comparing FM and FM|NM samples¹¹⁵ or by probing the magnetization dynamics via the magneto-optical Kerr effect (MOKE) in more complex magnetic multilayers.^{223,224} An important insight from the results in Ref. 113 is that the ultrafast spin current in the STE is only determined by the amount of excess energy that is deposited in the electronic system. In other words, only ultrafast heating of the FM is required, regardless of the precise shape of the resulting nonequilibrium electron distribution and, thus, the pump photon energy. This notion is in line with previous experimental findings for infrared and visible^{67,68,157} down to THz pump photons.¹⁷⁴

In terms of the STE performance, the connection between ultrafast demagnetization and spin transport can be of great value. It means that the vast knowledge from the ultrafast-magnetism community⁸⁴ can be transferred and exploited to enhance the THz-emission

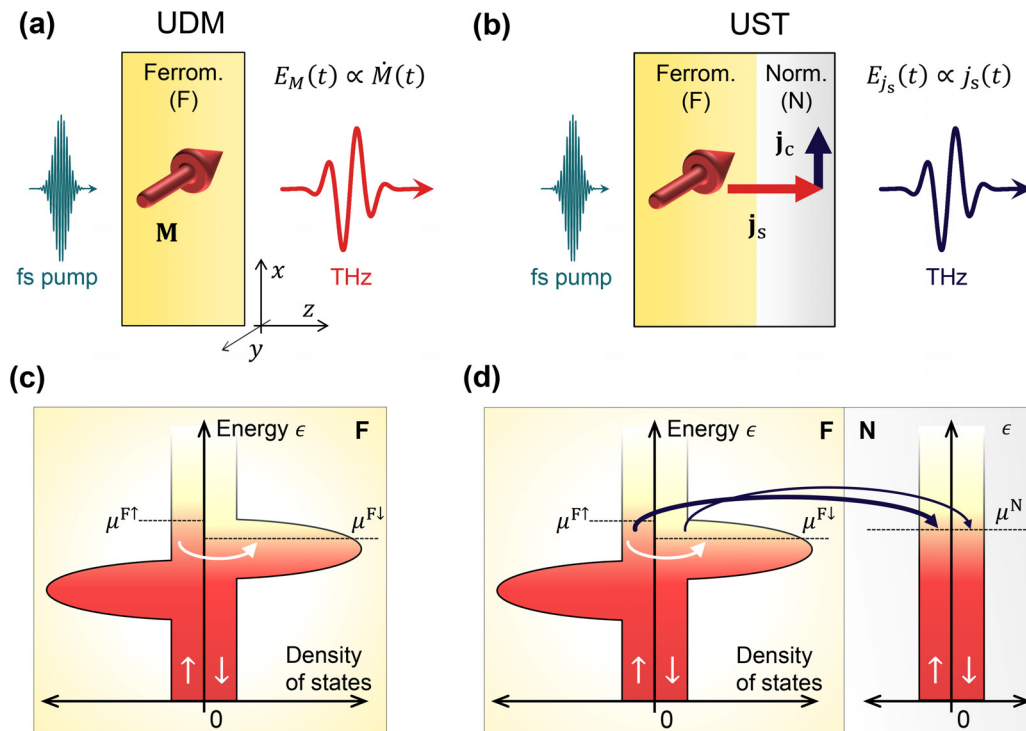


FIG. 17. Comparison of (a) optically driven ultrafast demagnetization (UDM) and (b) THz spin transport (TST). While the former causes magnetic dipole radiation proportional to the magnetization dynamics $\dot{\mathbf{M}}(t)$, the latter drives electric dipole radiation that is usually found in the STE. (c, d) Excitation by a femtosecond optical laser pulse triggers the splitting of the spin chemical potentials $\mu^{F\uparrow}$ and $\mu^{F\downarrow}$ inside the ferromagnet. This ultrafast spin voltage can relax via (c) local spin-flip events or via (d) spin transport into an adjacent nonmagnetic material. The figure is adapted from Ref. 113. Reproduced with permission from Rouzegar *et al.*, arXiv:2103.11710 (2021). Copyright 2021 Attribution-NonCommercial-NoDerivatives 4.0 International (CC BY-NC-ND 4.0).

strength of the STE. A possible route to an enhanced STE performance would be the suppression of ultrafast local spin relaxation inside the FM and the corresponding enhancement of the spin transport.

4. Spin injection across the FM/NM interface

As shown in Fig. 2, the spin current inside the STE is strongly localized at the FM/NM interface,¹¹⁵ which is why, in addition to the bulk, the interfaces have a strong impact on the spatial distribution of \mathbf{j}_s .¹⁷¹ For simplicity, theoretical calculations often assume a transparent FM/NM interface for electron or spin transport, i.e., all electrons can pass through the interface without any loss. However, in reality, electron reflection and spin loss at the interface have to be considered and can be seen as an interfacial spin resistance.

Recently, a theoretical study calculated the reflection of a superdiffusive spin current inside a FM|NM heterostructure (see Fig. 18) and revealed a possible impact of the interfacial spin resistance on ultrafast spin currents.⁹⁰ The calculated results show that the spin-up electrons (majority) have a lower reflectivity, that is, a higher interface transmittance than the spin-down electrons (minority) at an Fe/NM interface. Accordingly, the authors of Ref. 90 argued that the spin current would have a higher spin polarization after crossing the interface. Such a “positive” spin filter effect might enhance the demagnetization in the FM layer and the THz-emission efficiency. For the Ni/NM interface,

the spin filter effect is of opposite (“negative”) character and would lead to a decrease in the demagnetization and THz-emission amplitude.⁹⁰

A well-established method for DC characterization of spintronic properties of FM|NM heterostructures is spin pumping.²²⁵ However, the quantity that determines the spin injection into the NM layer across the interface in spin-pumping experiments is the effective spin mixing conductance $g_{\text{eff}}^{\uparrow\downarrow}$, which is different from the diagonal

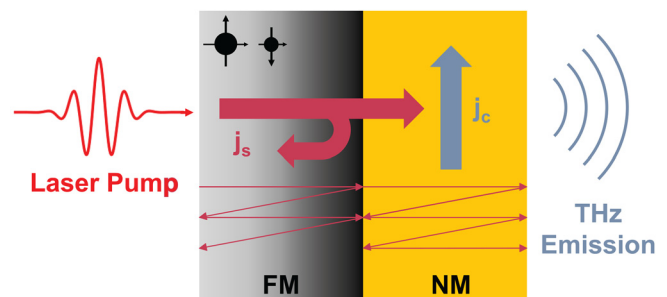


FIG. 18. Ultrafast spin transport and THz emission in the FM|NM heterostructure including the interface reflection. Reproduced with permission from Lu *et al.*, Phys. Rev. B **101**, 014435 (2020). Copyright 2020 Authors, licensed under a Creative Commons Attribution (CC BY) license.

counterparts $t_{\text{FM}/\text{NM}}$. Accordingly, a recent study, which directly measured $g_{\text{eff}}^{\uparrow\downarrow}$ and $t_{\text{FM}/\text{NM}}$ in the same samples by spin pumping and THz-emission spectroscopy,²²⁶ respectively, found that these two quantities are different but can be related to one another if the spin loss by interfacial spin-orbit coupling, that is, a spin memory loss during the THz emission process,²²⁷ is included. The spin memory loss, however, needs to be characterized by independent experiments such as tr-MOKE measurements.²⁰⁵ The experimental data suggested that, in Ni|Au heterostructures, $\sim 50\%$ of the spin current is lost in the spin transfer process at the interface due to spin-flip scattering processes.

Indeed, a recent work¹⁰⁰ (see Sec. II D 5) further confirmed that the quality of the interface plays an important role in the spin injection process, and the interfacial disorder can result in a reduction of the spin current from FM to NM. Future interface-modification studies might aim at disentangling the role played by changes in $t_{\text{FM}/\text{NM}}$, S2C by interfacial alloys,¹¹⁵ or a change in the interfacial magnetic properties such as reduced Curie temperatures^{228,229} that would impact the spin voltage¹¹³ close to the interface.

B. Process (3): Spin-to-charge current conversion

S2C plays an important role in the THz emitter made of FM|NM heterostructures. S2C phenomena, such as ISHE and IREE, are a consequence of spin-orbit coupling (SOC) mainly in the NM. Therefore, materials such as heavy metals, topological insulators, two-dimensional transition metal dichalcogenides (2D-TMDCs), and Weyl semimetals can all be candidates for the NM layer. This flexibility, thus, enables to probe spin-related phenomena in a wide range of materials beyond purely metallic FM|NM heterostructures using THz-emission spectroscopy.

1. Spin Hall effect and inverse spin Hall effect

The SHE is a transport phenomenon that generates spin currents from charge currents via SOC.^{230–233} The inverse process, the ISHE, which converts a spin current into an electric current, is driven by the same mechanism: SOC-induced spin-dependent scattering (Fig. 19). Different from the extrinsic SHE that originates from impurity scattering, the intrinsic SHE is directly related to the band structure of the material and has a sizable S2C efficiency in heavy metals (e.g., Pt, W, and Ta) and topologically nontrivial materials (e.g., topological insulators and Weyl semimetals).

The relation between charge- and spin-current density j_c and j_s is given in Eq. (3), which is repeated here for convenience²³¹

$$j_s = \gamma j_c. \quad (11)$$

The parameter γ is the spin Hall angle that describes the S2C efficiency [see Eq. (3)]. In some heavy metals, such as Pt and Ta, γ reaches values between 10^{-2} and 10^{-1} ,^{234–239} which is much larger than for materials with minor SOC (Cu, Au, or some semiconductors).^{240,241} To date, there is a huge number of DC spintronic studies of the SHE/ISHE in various sample systems that are indispensable for realizing efficient STEs.

Aside from γ , another critical parameter in the SHE/ISHE is the relaxation length λ_{NM} , which describes how far the spin current can flow in a material. At THz frequencies, it equals the electronic mean free path rather than the spin diffusion length, as discussed in Sec. II C 4.

2. Rashba-Edelstein effect and inverse Rashba-Edelstein effect

Different from the SHE and ISHE, which are allowed in regions with inversion symmetry, additional S2C mechanisms can occur in regions with broken inversion symmetry such as interfaces or surfaces. Prominent examples are the Rashba-Edelstein effect (REE) and its inverse, the IREE (Fig. 20). In a simple microscopic model of the REE, an applied electric field drives a charge current and, thus, shifts the Fermi surface, that is, it induces an asymmetry in the electron distribution in wavevector space [Fig. 20(b)]. Due to spin-momentum locking, such a nonequilibrium electron distribution can lead to an asymmetric spin distribution, and, thus, form a net spin polarization in regions with broken inversion symmetry. The spin polarization can diffuse as a spin current or exert spin torque to an adjacent material.²⁴²

The REE and IREE were observed experimentally in many material systems, such as at Ag/Bi interfaces,^{117,123,126,243–246} in topological insulators,^{125,247–252} two-dimensional electron gases (2DEGs),²⁵³ 2D-TMDCs,^{254–256} Heusler compounds,²⁵⁷ and at metal/oxide interfaces.²⁵⁸

The REE and the IREE can be characterized by the following S2C coefficients:

$$q_{\text{REE}} = \frac{j_s}{J_c}, \quad (12)$$

$$\lambda_{\text{IREE}} = \frac{J_c}{j_s}, \quad (13)$$

where j_s is a 3D spin current with dimension $\text{s}^{-1} \text{m}^{-2}$ and J_c is a 2D charge current with dimension $\text{s}^{-1} \text{m}^{-1}$. Therefore, the conversion coefficients q_{REE} and λ_{IREE} have dimensions m^{-1} and m , respectively.

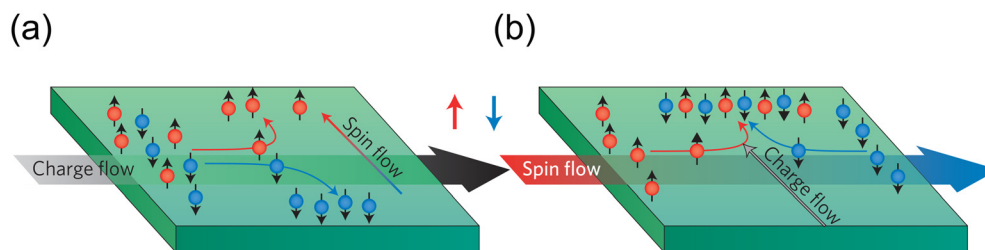


FIG. 19. Schematic of (a) spin Hall effect and (b) inverse spin Hall effect. Reproduced with permission from K. W. Kim and H. W. Lee, Nat. Phys. **10**, 549–550 (2014).²³³ Copyright 2014 Macmillan Publishers Limited.

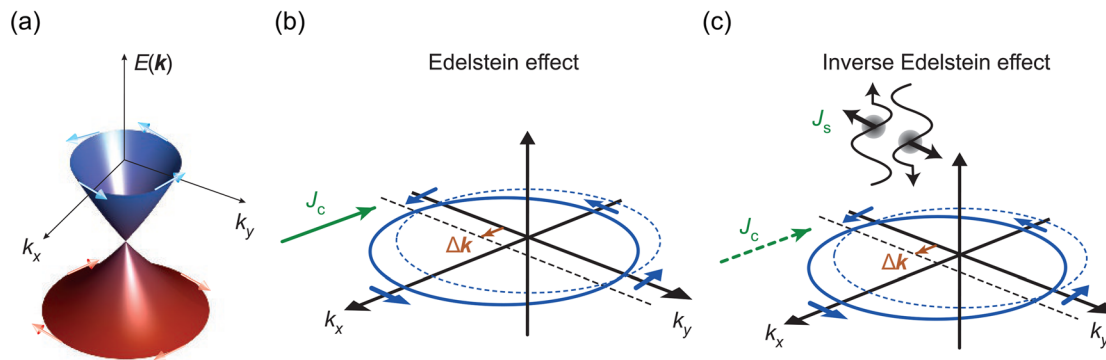


FIG. 20. (a) Schematic of the band structure of the surface states of a topological insulator. Schematic of the (b) Rashba–Edelstein effect and (c) inverse Rashba–Edelstein effect. Reproduced with permission from Soumyanarayanan *et al.*, *Nature* **539**, 509 (2016). Copyright 2016 Macmillan Publishers Limited.

In addition, q_{REE} and λ_{IREE} are constants that typically depend on the properties of the interface/surface.^{126,248,251,259}

To compare the REE/IREE with the SHE/ISHE, a parameter called the effective spin Hall angle or spin-torque ratio, γ^{eff} , is frequently used. This parameter can be defined as $\gamma^{\text{eff}} = \lambda_{\text{IREE}} t$ for the IREE and $\gamma^{\text{eff}} = q_{\text{REE}}/t$ for the REE. Here, t is the thickness of the interface or surface. Although the thickness of a 2D interface/surface is, in principle, hard to define, t is often approximated as the thickness of the first few atomic layers, in which the S2C happens. For example, in the spin-pumping experiments of four-layer MoS_2 ,²⁵⁵ the IREE coefficient $\lambda_{\text{IREE}} = 0.4$ nm and $\gamma^{\text{eff}} = 0.96$ were found by taking the thickness of the interface equal to the sample thickness, that is, $t = 2.4$ nm. To date, the materials that show a sizable REE/IREE usually have extremely high S2C efficiencies, which raises questions about the exact meaning of λ_{IREE} . For instance, the value of γ^{eff} is between 0.14 and 0.96 for MoS_2 ,^{254,255} between 2.0 and 3.5 for Bi_2Se_3 ,^{234,247,249} and ~ 1.5 for the Ag/Bi interface.¹²⁶

In principle, distinguishing the ISHE from the IREE can be possible by the different thickness dependences of the induced charge currents that are given by¹²⁶

$$J_c = e\gamma\lambda_{\text{NM}}\tanh\frac{d_{\text{NM}}}{2\lambda_{\text{NM}}}j_s \quad (14)$$

in the ISHE case [see also Eq. (7)] and by

$$J_c = e\lambda_{\text{IREE}}j_s \quad (15)$$

in the IREE case. However, an experimental separation of the ISHE from the IREE remains challenging in transport^{260,261} and especially in THz-emission experiments. The latter might be eased in the future by the observation that in contrast to the ISHE, the IREE scales with the accumulated spin density and should, therefore, exhibit memory effects, which imply a frequency dependence of λ_{IREE} and q_{REE} .

Experimentally, strong evidence for the IREE mechanism was provided by the opposite polarities of the emitted THz pulses depending on the stacking order, i.e., by comparing Ag/Bi to Bi/Ag interfaces (Fig. 21) although certain ambiguities remain.²⁶² The bandwidth of the detected THz pulses was about 3 THz and limited by the relatively narrow bandwidth of the detection crystal.^{117,123} It appears reasonable to the authors that the IREE-based STEs can reach the record bandwidths of their ISHE-based counterparts given the observation that

many other spintronic phenomena also remain operative at THz rates.^{64,78,142,263}

Another example of IREE-based THz emission appears in Co| MoS_2 heterostructures.¹²⁴ Previous spin-torque-ferromagnetic-resonance and spin-pumping measurements revealed a sizable S2C via the IREE in the atomically thin 2D system MoS_2 .^{254–256} Note that MoS_2 is a semiconductor with a large bandgap of ~ 1.9 eV, which is quite different from the conducting NM layers discussed before. Such material difference may lead to a much smaller spin-current-injection efficiency²⁶⁴ but could also enable functionalities, as will be discussed further below.²⁶⁵

Remarkably, the THz-emission amplitude from IREE-based STEs can reach up to 20% of a 1-mm-thick ZnTe crystal under the same experimental conditions^{117,125} and, thus, also about 20% of the best ISHE-based STEs.

The high S2C efficiency of the IREE-based STE can potentially generate intense THz radiation and provide a promising approach for broadband THz emission.

3. Other S2C mechanisms

Apart from the ISHE and the IREE, there are other S2C mechanisms such as the valley Hall effect (VHE)²⁶⁶ and valley Edelstein effect (VEE).²⁶⁷ Both mechanisms are associated with the valley degree of freedom that is coupled with the spin degree of freedom, for instance, in 2D-TMDCs.²⁶⁸

For example, a monolayer of MoS_2 has two nonequivalent valleys in its band structure that are located in the vicinity of the K and K' point. Due to spin-valley coupling, a spin flip is always accompanied by a change of the electronic valley and vice versa.²⁶⁸ Therefore, the VHE, which separates charges transversely to an applied electric field, can be accompanied by the formation of a transverse spin accumulation. In contrast, for the VEE, the electric-field-induced spin polarization is predicted to be parallel to the applied electric field. However, the reciprocal effects of VHE and VEE, which would be useful for the STE design, have not yet been observed. Its observation would require the injection of a spin current into the VHE/VEE material and the separation from other S2C mechanisms such as the ISHE or IREE. Another challenge in terms of strong THz emission might arise from the difficulty in efficiently injecting spin currents from a magnetic metal into the typically semiconducting VHE/VEE material.²⁶⁴

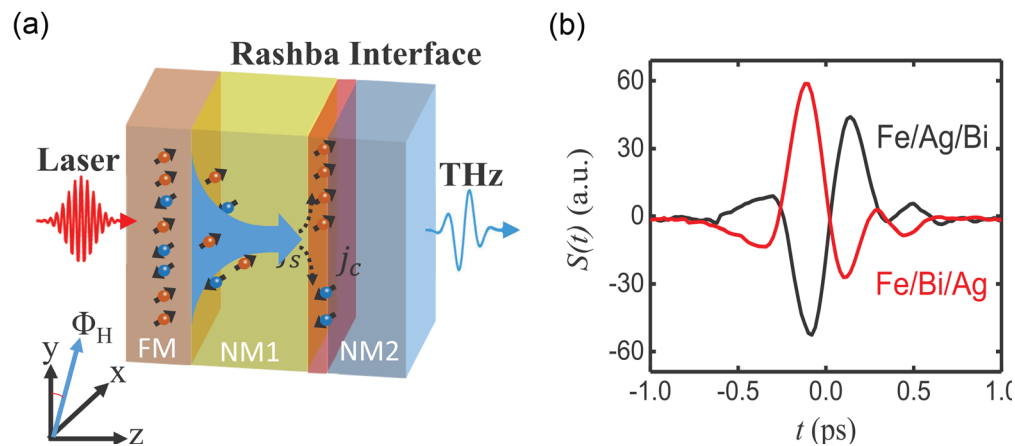


FIG. 21. THz emission from Ag/Bi Rashba interfaces under femtosecond-laser excitation. (a) Excitation of a thin-film heterostructure with a femtosecond laser pulse triggers the injection of a spin current into the NMs (NM1 = Ag, NM2 = Bi). The spin current is transformed into an in-plane charge current by the Rashba spin-orbit field at the Ag/Bi interface. (b) The polarity of the emitted THz pulse depends on the orientation of the Rashba interface with respect to the spin current source (FM layer). Reproduced with permission from Zhou *et al.*, *Phys. Rev. Lett.* **121**, 086801 (2018).¹¹⁷ Copyright 2018 American Physical Society.

In summary, S2C is a critical ingredient for spintronic THz emission and has a strong impact on the THz-emission efficiency. Conversely, THz-emission spectroscopy is an excellent tool itself to unravel the properties of S2C in various material systems.⁸² It is, therefore, a promising approach to find efficient spintronic materials.

IV. APPLICATIONS

The unique advantages of the STE over other THz emitters enable a manifold of applications. For each of them, different aspects of the STE are exploited and, thus, require a tailored optimization that may be guided by our introduced STE model [Eqs. (3)–(5), (7), and (8)]. In the following, we will highlight certain emerging STE applications that may have a profound impact beyond the field of THz photonics.

A. Broadband linear THz spectroscopy

In several recent studies, STEs were used to perform broadband THz spectroscopy. In an early work, Seifert *et al.* demonstrated the spectroscopic characterization of a thin Teflon tape from 1 to 30 THz, which could reveal the characteristic THz phonon absorptions of this material.⁶⁴ Subsequent studies used STEs and linear THz spectroscopy to measure the frequency dependence of central spintronic phenomena such as the anomalous Hall effect or the anisotropic magnetoresistance.^{79,263}

Davies *et al.* used a STE to measure the transient photoconductivity over an extended THz spectral range in perovskites²⁶⁹ and to determine the temperature-dependent THz refractive index of quartz.²⁷⁰ In a related study, the equilibrium and the transient THz conductivity of a perovskite sample as a function of the doping concentration were measured with a STE.²⁷¹

In a near-field-type approach, Balos and co-workers measured the THz complex-valued refractive index of water that was in direct physical contact to the STE's metal surface from 0.3 to 15 THz.²⁷² This technique can be applied to any liquid and is particularly useful for THz-opaque liquids.

B. Spatiotemporal THz beam modulation

One major advantage of the STE over other THz sources is its tunability by external magnetic fields, which allows for the global variation or even spatial patterning of the linear THz polarization.

Hibberd and co-workers⁷⁶ demonstrated reversible magnetic patterning of the FM layer inside the STE using inhomogeneous external magnetic fields as shown in Fig. 22(a). This approach permits the creation of different THz polarization states, including doughnut-like modes or radially polarized THz electric fields.^{76,273} A related study generated THz pulses from a STE that had two perpendicularly magnetized regions close to each other.¹¹⁰ This approach allowed for the creation of elliptically polarized THz electric fields. Due to the fact that it is based on double pulses, it is applicable only to a relatively narrow frequency range.

Shaping the THz polarization from linear to circular behind the STE was demonstrated with the help of a large-birefringence liquid crystal, onto which the STE was deposited.¹¹⁰ Another approach exploits different THz-emission mechanisms in STEs including topological materials. It superimposes THz electric fields with perpendicular linear polarization, yet with different phases to obtain elliptically polarized THz pulses over a relatively narrow THz frequency interval.²⁷⁴ Future STE designs might additionally incorporate multilayer structures with a pinned FM layer to allow for a magnetic-field-free operation¹⁰⁷ as required in many application environments.

Recently, Gueckstock *et al.* succeeded in modulating the THz pulses from a STE with a bandwidth of 30 THz in the polarity and polarization direction at rates of up to 10 kHz and with a contrast exceeding 99%. This achievement relies on the rapid variation of the STE magnetization as shown in Fig. 22(b).^{75,275} It enables low-noise broadband THz spectroscopy or rapid ellipsometric sample characterization without the need for mechanical choppers or other modulators of the optical pump beam.

Finally, shaping of THz beam properties, such as the divergence, can be achieved by depositing the STE on flexible substrates as shown

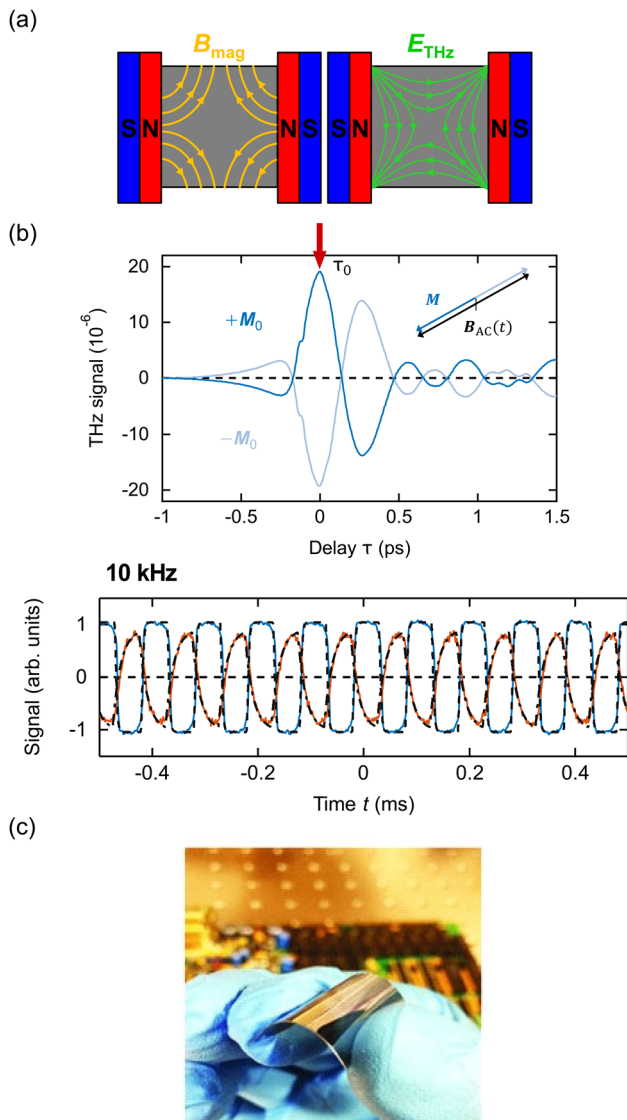


FIG. 22. Spatiotemporal THz beam shaping using the STE. (a) Spatially structured magnetic fields can generate doughnut-like THz-electric-field distributions from the STE. Reproduced with permission from Hibberd *et al.*, *Appl. Phys. Lett.* **114**, 031101 (2019).⁷⁶ Copyright 2019 AIP Publishing. (b) Modulation of the STE magnetization at kHz rates enables rapid THz-polarity modulation. Top panel: THz-emission signals for opposite magnetizations of the STE. Bottom panel: Time evolution of the peak THz signal (red arrow in left panel) upon modulation of the STE magnetization at 10 kHz (blue line) and magneto-optic signal tracking the STE magnetization (orange line). Reproduced with permission from Gueckstock *et al.*, *Optica* **8**, 1013 (2021).⁷⁵ Copyright 2021 Authors, licensed under a Creative Commons Attribution (CC BY) license. (c) Flexible substrates or deposition on curved surfaces of STE stacks broaden their applicability. Reproduced with permission from Wu *et al.*, *Adv. Mater.* **29**, 1603031 (2017).⁶⁶ Copyright 2017 Wiley-VCH.

in Fig. 22(c).⁶⁶ A straightforward extension of this approach involves growing the STE on top of curved surfaces. It allows for a built-in THz focusing functionality by using, for instance, parabolic mirrors or THz/optical lenses as the STE substrate.

C. THz imaging

The STE design provides a unique platform for near-field imaging applications, because the THz emitter is just a few nanometers thick. Therefore, probing THz near-fields with lateral features above the STE surface becomes possible whose sizes are given by the lateral structure of the optical pump beam that is incident onto the STE. In principle, the lateral THz field can have feature sizes comparable to the wavelength of the optical pump, which often lies in the submicrometer range.²⁷⁶

Recently, such THz near-field imaging with a STE was first demonstrated,⁷⁷ which allowed the authors to resolve subwavelength structures with a resolution of $6.5 \mu\text{m}$ by placing the object in the near-field region of the STE. In their experiment, the STE was illuminated with a spatially structured pump beam, and the emitted THz signal was recorded using EO detection. After illumination with a specific set of spatially structured pump pulses, an inversion algorithm was used to extract the image of the object. Polarization-induced imaging artifacts could be reduced by switching the THz polarization with an external magnetic field. Depth information of the object could be obtained by exploiting the different arrival times of the THz pulses at the detector.

A similar approach aimed at combining the STE near-field imaging concept with resonant microstructures, i.e., split ring resonators, to enhance certain THz frequencies locally. In this way, Bai and co-workers could detect a change in the emitted THz pulse upon covering their STE device with cancer cells, which might be useful for future biological sensing applications.²⁷⁷ More recent studies reported on the implementation of a trilayer STE into a THz-emission microscope²⁷⁸ and on achieving a spatial resolution down to $5 \mu\text{m}$ using THz near-field imaging with a STE.²⁷⁹

Because of its compactness, a STE could be combined with an electro-optic detection unit to yield a magnetic field sensor with a relatively small footprint.²⁸⁰ Following this approach, Bulgarevich *et al.* demonstrated a sensitivity in the millitesla range with submillimeter spatial resolution.

D. Nonlinear THz spectroscopy

Strong THz pulses can trigger a nonlinear response of the studied system.⁴² As illustrated in Figs. 23(a) and 23(b), a large-area STE was used to generate THz electric fields with a peak strength of 300 kV/cm^{-1} (Ref. 69) in the diffraction-limited focus of a Gaussian beam. The pulses drove a third-order nonlinear effect, i.e., the THz Kerr effect, inside a diamond sample.

A completely different nonlinear application of the STE was demonstrated by Muller *et al.*²⁸¹ They coupled THz pulses with a bandwidth exceeding 20 THz generated from a STE by using 8 fs pump pulses into the junction of a scanning tunneling microscope (STM) as shown in Figs. 23(c) and 23(d). Because of the inherent nonlinear response of the tunnel current to the incident THz electric field, the THz pulses were rectified. The resulting DC current was detected by the low-bandwidth STM electronics. The broad THz spectrum delivered by the STE allowed the authors to extract the THz response function of the tunnel junction in the range between 1 and 15 THz, which revealed a pronounced low-pass behavior.

E. THz-emission spectroscopy of spintronic transport

Note that the spintronic-THz-emission process itself is nonlinear in the pumping field and, thus, a nonlinear spectroscopic approach. It

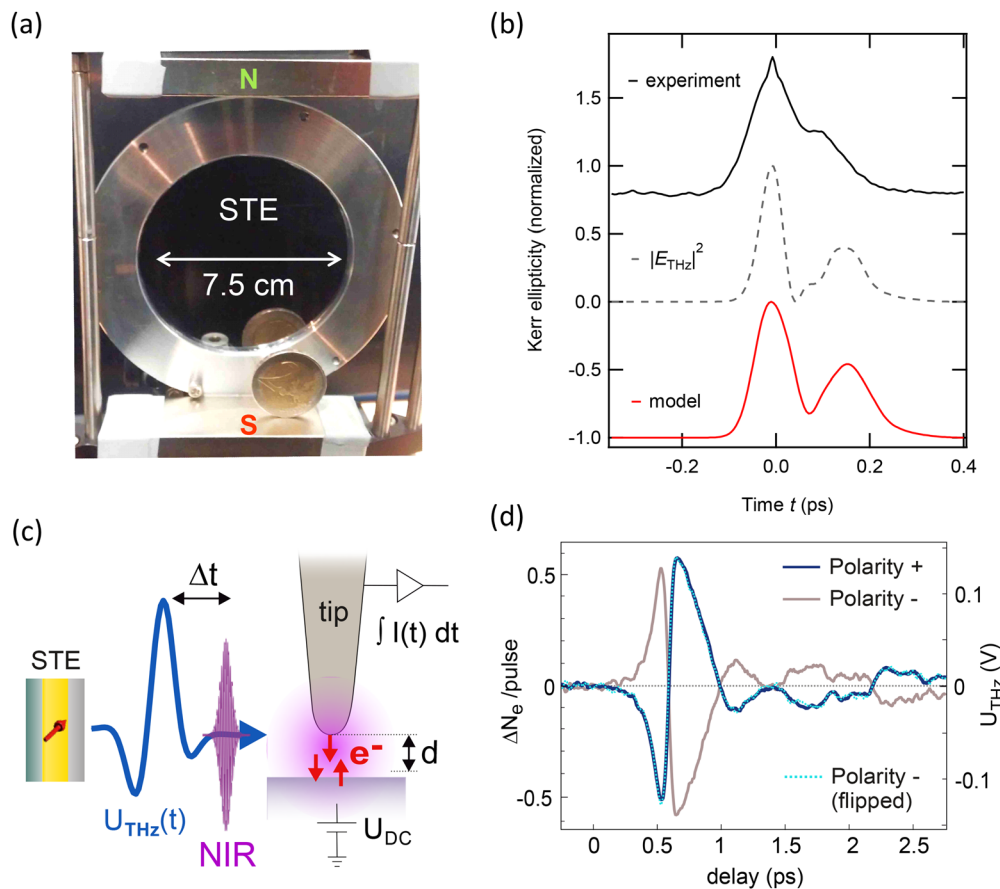


FIG. 23. Nonlinear-optics applications of STEs. (a) Large-area STE emitter that can generate THz electric field strengths of 300 kV cm^{-1} when excited by femtosecond laser pulses at 800 nm central wavelength and 5 mJ pulse energy. (b) Third-order nonlinear effect (Kerr effect) in diamond driven by the strong THz electric field generated with the STE shown in (a). Reproduced with permission from Seifert *et al.*, *Appl. Phys. Lett.* **110**, 252402 (2017).⁶⁹ Copyright 2017 AIP Publishing. (c) THz-STM setup, in which the STE generates THz-electric-field pulses that serve to apply an ultrafast voltage across the STM junction. The resulting THz voltage U_{THz} drives photoelectrons that are generated by a preceding near-infrared (NIR) pulse. (d) Rectified tunneling electrons as a function of the delay time between the THz and the NIR pulse. The resulting waveform is odd in STE magnetization and low-pass filtered by the response function of the STM junction. Reproduced with permission from Muller *et al.*, *ACS Photonics* **7**, 2046 (2020).²⁸¹ Copyright 2020 Authors, licensed under a Creative Commons Attribution (CC BY) license.

was used to characterize spintronic transport in FM|NM STEs as a function of the material composition. Such THz emission studies were shown to be fully consistent with low-frequency electronic characterization approaches in terms of the extracted S2C efficiencies.^{78,142}

Importantly, however, the all-optical approach does not require any sample microstructuring and, thus, allows one to extract spintronically relevant parameters such as the S2C efficiency and the spin-current propagation length with large sample throughput.^{73,88,105,117,124,125,133–136,140,142,159,173,282}

A further interesting approach along these lines is to study the propagation of spin currents through interlayers^{66,133,283} that might even be magnetically ordered,^{284,285} or of the FM magnetization dynamics^{159,286} that might even involve its laser-induced reversal.²⁸⁷

Recent THz-emission studies provided insight into the magnetic structure of the ferrimagnetic half-metal Fe_3O_4 ¹¹⁴ and revealed the initial formation dynamics of the ultrafast spin Seebeck effect (Ref. 88 and Sec. II C 3).

V. FUTURE PERSPECTIVES

A. Semiconductors and 2D materials

In contrast to metal-based STEs, ultrafast spin injection (and THz emission) from a FM into a semiconductor (SC) can be influenced by an interfacial Schottky barrier that may reduce the spin injection efficiency, but simultaneously increase the spin polarization of the injected carriers (Fig. 24 and Refs. 264, 265, 288, and 289). Moreover, the propagation length of spin currents inside SCs can be significantly larger than in metals,²⁹⁰ which could boost the STE THz-emission performance. Consequently, FM|SC bilayers are a promising platform for efficient STEs.

2D materials have promising properties, such as spin-valley locking or large S2C efficiencies,^{124,291–295} for their use in STEs. Recently, a giant ultrafast spin injection into a SC was experimentally confirmed in FM|MoS₂ heterostructures,^{124,254} where MoS₂ is an atomically thin SC with a bandgap of around 1.9 eV and strong SOC.²⁹⁶ The latter

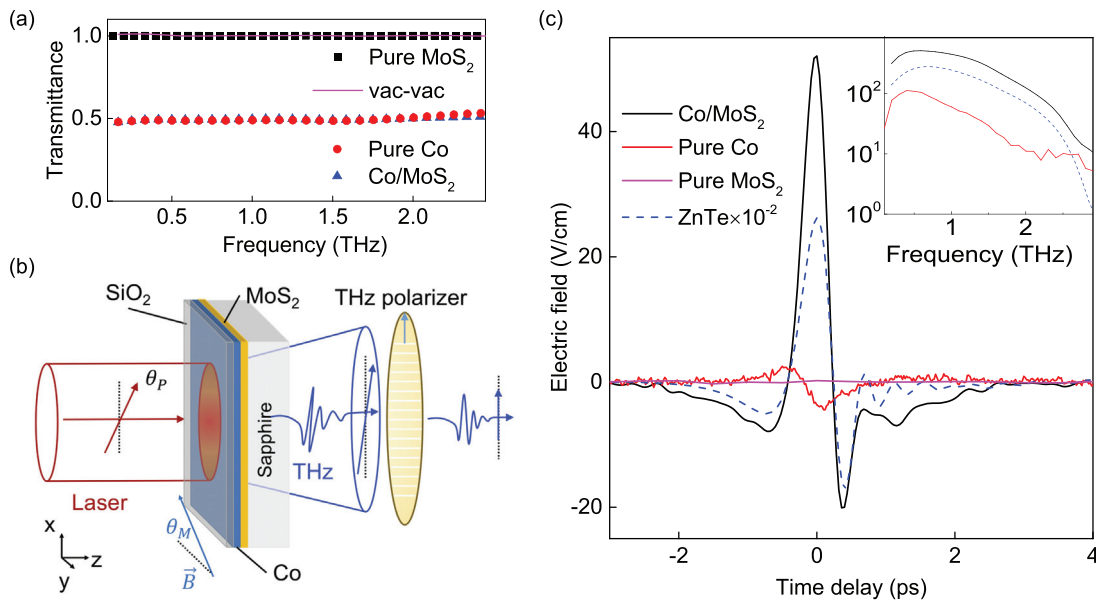


FIG. 24. Measured spin injection from the FM Co into the semiconductor MoS₂. (a) THz transmittance through vacuum, Co, MoS₂, and Co|MoS₂ showing the transparency of MoS₂ to THz radiation. (b) Experimental geometry for measuring the THz emission upon optical pumping. (c) Comparison of emitted THz electric fields showing the enhanced emission from Co|MoS₂ bilayers due to spin injection into the semiconductor. Reproduced with permission from Cheng *et al.*, Nat. Phys. **15**, 347–351 (2019).¹²⁴ Copyright 2019 Macmillan Publishers Limited.

property is essential for a large S2C efficiency that leads to the emission of THz radiation. The optically injected spin-current density was estimated to be $4 \times (10^6\text{--}10^8) \text{ A cm}^{-2}$,¹²⁴ which is several orders of magnitude larger than the values obtained by other methods, such as spin pumping ($10^2\text{--}10^3 \text{ A cm}^{-2}$)²⁹⁷ and spin-transfer torque in magnetic tunnel junctions ($10^{-1}\text{--}10^0 \text{ A cm}^{-2}$).^{298,299}

Future engineering of the Schottky barrier and the SC bandgap could optimize optically injected spin currents. Along these lines, Vetter *et al.*³⁰⁰ investigated the spintronic THz emission from NiFe|n-GaN heterostructures. They found a significant change in the emitted THz amplitude as a function of the GaN doping level. The more metallic the SC was, the smaller the emitted THz pulse became. However, disentangling the spin currents injected into the SC layer from other effects, such as a decrease in the sample impedance with increased doping level [last factor in Eq. (7)] or a S2C already inside the metallic FM layer,¹⁷¹ remains a challenge.

B. Topological materials

Topological materials bear a large potential for an efficient S2C due to the inherent spin-momentum locking realized, for instance, in the surface states of topological insulators. The impact of topological surface states on the THz S2C was investigated in Co|Bi₂Se₃ heterostructures.¹²⁵ As shown in Fig. 25, the THz-emission signal is strongly depended on the Bi₂Se₃ thickness: The signal increased significantly if the Bi₂Se₃ layer was thicker than five quintuple-layers. This thickness is known to be the lower limit for the formation of the topological surface states.³⁰¹ However, a thorough quantification of the impact of the topological surface state on the THz emission process poses an interesting question for future studies.

Interestingly, the THz-emission amplitude from Co|Bi₂Se₃ was found to be constant for a sample temperature between 10 and 300 K, which may make this type of emitter design suitable for applications in environments with large temperature variations such as space missions [Fig. 25(b)]. Implementing emerging material classes such as magnetic or nonmagnetic Weyl semimetals³⁰² as S2C converters or spin-current-generating layers might in the future further enrich the STE by topological functionalities such as quantized photocurrents.³⁰³ Accordingly, a recent study highlighted the efficient spin-current generation in Weyl semimetals that might help one enrich the STE by topological functionalities.³⁰⁴ For instance, the combination of the ISHE-based THz-emission mechanism with the circular photogalvanic effect, that is, the injection current, in topological materials was shown to allow for elliptical THz polarization states.³⁰⁵

C. Photonic optimization

Optimizing the STE from a photonic perspective mainly follows two goals: To increase (i) the pump-pulse absorption and (ii) the THz-outcoupling efficiency into free space.

Anti-reflective coatings were shown to be a promising approach to reach (i). Herapath *et al.*⁶⁷ could enhance the pump absorption from about 50% to 100%, thereby increasing the emitted THz pulse amplitude by a factor of 2 for certain pump wavelengths.

To reach goal (ii), THz antenna structures were deposited onto the STE by using patterned gold overlayers. As a result, the THz electric field from the STE was enhanced by a factor of 2 compared to an identical bare STE,¹⁰⁸ as confirmed by a more recent study.³⁰⁶ In another work, the STE performance was significantly improved in a narrow THz frequency range by adding a horn antenna to the STE.³⁰⁷

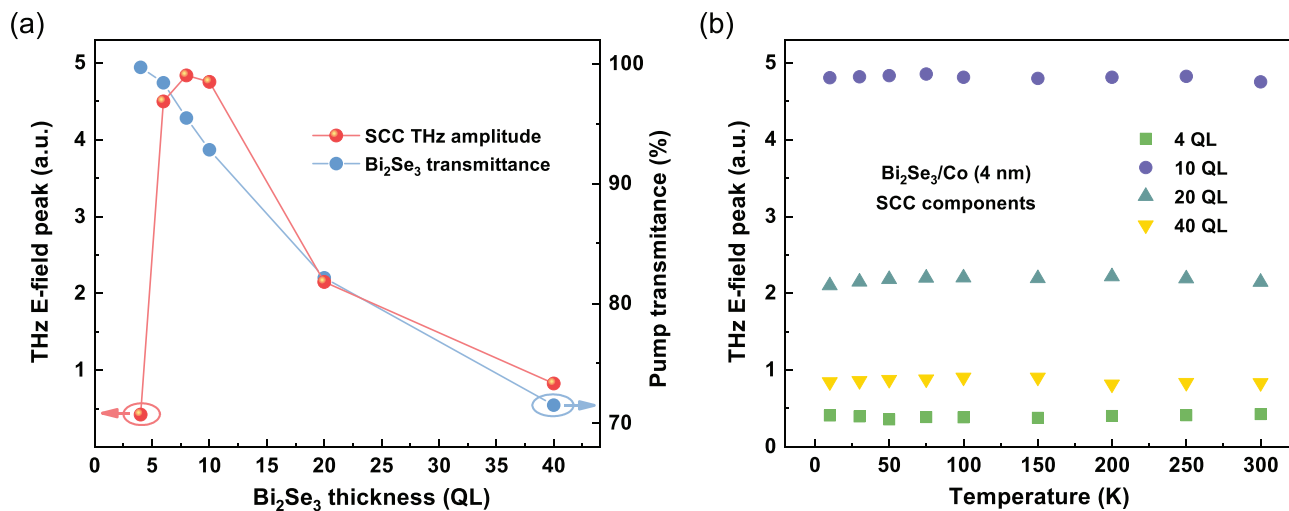


FIG. 25. THz emission from thin film heterostructures containing Co and the topological insulator Bi₂Se₃. (a) THz-signal amplitude and THz transmittance as a function of the thickness of Bi₂Se₃. (b) THz-signal amplitude related to S2C as a function of the temperature for different thicknesses, i.e., the number of quintuple layers (QLs). Reproduced with permission from Wang *et al.*, *Adv. Mater.* **30**, e1802356 (2018).¹²⁵ Copyright 2018 Wiley-VCH.

It should, however, be noted that antenna designs and antireflection coatings typically only perform well in a certain interval of THz and pump frequencies, respectively. Thus, finding broadband solutions remains a future challenge, and there is much optimization potential for THz-antenna designs. An interesting strategy might be to guide the pump field to spatial regions, such as the FM layer, that enable larger THz-emission amplitudes per pump energy, for instance, by plasmonic enhancement as used in designs of photo-conductive switches.^{57,308}

D. Integration, combination with existing technologies and readiness for real-world applications

Hybrid emitter concepts hold great promise for combining the benefits from different THz-emitter designs. Chen and co-workers³⁰⁹ combined the ISHE-based STE with a photoconductive switch and demonstrated a modulation of the THz amplitude and the center frequency by the bias current with a significant emission enhancement at low THz frequencies.

In terms of operational temperature T_0 , the choice should aim at a region in the $M(T)$ curve of the magnetic layer that maximizes $\Delta M/\Delta T$.¹¹³ In other words, the pump-induced transient change in the electronic temperature $\Delta T = T_{e,\text{peak}} - T_0$, with the electronic peak pump-induced temperature $T_{e,\text{peak}}$, should lead to a maximized change in magnetization ΔM . Accordingly, cooling down of typical STEs did not change their performance drastically^{122,128} since typical Curie temperatures, that is, regions where $\Delta M/\Delta T$ is large, are found at around 1000 K for typical bulk 3d ferromagnets. Note, however, that these critical temperatures might be reduced in thin films²²⁸ and close to interfaces.²²⁹

Future goals include the on-chip integration of a STE,^{310,311} which might boost the bandwidth of on-chip Auston switches that were recently exploited to trigger ultrafast switching of magnetic nanostructures,³¹² and pave the way toward THz photonic integrated circuits.

E. The ideal STE: Estimating future improvements

Finally, we would like to present an estimate of what improvements might be achievable in terms of the performance of the STE. Starting from the trilayer STEs with optimized thickness and materials (TeraSpinTec GmbH), future developments might ideally reach the following goals: (i) increase in the S2C from currently about 10% to 100%, i.e., enhance the efficiency of the ISHE or IREE S2C to fully use the initial spin current,⁹⁹ (ii) utilization of 100% of the pump beam energy instead of currently about 50%, (iii) combination of forward and backward propagating THz pulses, which would yield a factor of 2 in the THz-electric-field amplitude, (iv) optimized outcoupling efficiency of the THz electric field from the metal stacks into free space from currently about $Z = Z_0/(n_1 + n_2 + Z_0G) \approx Z_0/5$ to an ideal value of $Z_0/2$ ($n_1 = n_2 = 1, G = 0$), implying an increase by a factor of 2.5 that might be feasible by proper photonic design,¹⁰⁸ and (v) maximizing the injected spin current into the NM, i.e., the factor $t_{\text{FM}/\text{NM}}^0$, which can be estimated from a combined spin pumping/THz emission study but has so far only been characterized for a very limited set of FM|NM combinations.²²⁶ However, the value of $t_{\text{FM}/\text{NM}}$ has an upper limit that can be calculated from the Sharvin conductances of the materials that form the interface for spin current transmission.^{90,313} In terms of damage threshold that is currently already at 5 mJ/cm², an advanced heat-sinking strategy might help one push this limit even further.⁷⁰

In total, the above points promise an ideal increase in the amplitude of the THz-electric field from the STE at a given pump-pulse energy by a factor of 100 without even considering possible improvements in the factor $t_{\text{FM}/\text{NM}}^0$. This consideration implies an increase in the THz pulse energy at constant pump pulse energy by 4 orders of magnitude, which demonstrates the enormous potential of future STE optimizations.

VI. CONCLUSIONS AND OUTLOOK

The field of spintronic THz emitters continues to evolve rapidly with developments in terms of materials and of the understanding of the fundamental physics behind the STE. We see great potential

to increase the emitted THz field strength by exploiting photonic designs, emerging material classes, such as topological materials and ferromagnet/semiconductor interfaces, or emerging spintronic phenomena such as the orbital Hall effect.^{314,315}

The manifold of existing STE utilizations, for instance, spintronic characterization, THz near-field imaging, or THz-driven scanning tunneling microscopy, clearly demonstrates the STE's potential for both, basic research and future applications. We hope that this reviewing Editorial will be a useful introduction for the readers to the basic concepts and ideas of STEs, thereby allowing them to push the research field of spintronic THz emitters forward in the near future. Finally, the broadband and efficient detection of THz pulses using spintronic principles remains a major open research field that might significantly benefit from the knowledge acquired from studying STEs.

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AUTHOR DECLARATIONS

Conflict of Interest

T.S.S. and T.K. are shareholders of TeraSpinTec GmbH, and T.S.S. is an employee of TeraSpinTec GmbH.

Author Contributions

T.S.S. and L.C. contributed equally to this work.

DATA AVAILABILITY

The data that support the findings of this study are available within the article.

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