

Terahertz time-domain signatures of the inverse Edelstein effect in topological-insulator/ferromagnet heterostructures

G. Bierhance^{1,2}, C. In¹, E. Rongione^{3,4}, R. Rouzegar¹, O. Gueckstock¹, E. Longo^{5,6},
L. Baringthon³, N. Reyren³, R. Lebrun³, J.-M. George³, P. Tsipas⁷, M. Wolf²,
T.S. Seifert¹, R. Mantovan⁵, H. Jaffrès³, A. Dimoulas⁷ and T. Kampfrath^{1,2,*}

¹Department of Physics, *Freie Universität Berlin*, 14195 Berlin, Germany

²Department of Physical Chemistry, *Fritz Haber Institute of the Max Planck Society*, 14195 Berlin, Germany

³Laboratoire Albert Fert, CNRS, Thales, *Université Paris-Saclay*, 91767 Palaiseau, France

⁴Catalan Institute of Nanoscience and Nanotechnology (ICN2), CSIC and BIST, Campus UAB, Bellaterra, 08193 Barcelona, Spain

⁵CNR-IMM Unit of Agrate Brianza, Via C. Olivetti 2, 20864 Agrate Brianza, Italy

⁶Institut de Ciència de Materials de Barcelona (ICMAB-CSIC), Campus de la UAB, 08193 Bellaterra, Spain

⁷Institute of Nanoscience and Nanotechnology, *National Center for Scientific Research "Demokritos"*, 15310 Athens, Greece



(Received 1 August 2024; revised 17 October 2025; accepted 2 December 2025; published 18 February 2026)

Three-dimensional topological insulators possess topologically protected surface states with spin-momentum locking, which enable spin-charge-current interconversion (SCI) by the inverse Edelstein effect (IEE). However, it remains experimentally challenging to separate the surface-related IEE from the bulk-type inverse spin Hall effect (ISHE). Here we search for distinct time-domain signatures of the two SCI phenomena in an $\mathcal{F}$ /TI model stack of a ferromagnetic-metal layer $\mathcal{F}$ (Co and Fe) and a topological-insulator (TI) layer (Bi_2Te_3 , SnBi_2Te_4 , and $\text{Bi}_{1-x}\text{Sb}_x$ with $x = 0.15$ and $x = 0.3$), where the focus is on Bi_2Te_3 . A femtosecond laser pulse serves to induce a transient spin voltage $\mu_s^{\mathcal{F}}$ in $\mathcal{F}$ and, thus, drive an ultrafast spin current out of $\mathcal{F}$. SCI results in a transverse charge current with a sheet density I_c that is detected by sampling of the emitted terahertz electric field. Analysis of the dynamics of $I_c(t)$ vs time t relative to $\mu_s^{\mathcal{F}}(t)$ reveals two components with distinct timescales: (1) a quasi-instantaneous response and (2) a longer-lived response with a relaxation time of 270 fs, which is independent of the $\mathcal{F}$ material chosen. Component (1) is consistently ascribed to the ISHE. In contrast, we interpret component (2) as a signature of interfacial spin accumulation and the IEE at the $\mathcal{F}/\text{Bi}_2\text{Te}_3$ interface, with a fraction of less than 10^{-2} of the incident spins participating. This assignment is fully consistent with respect to its dynamics and magnitude. We rule out other possible signal contributions, such as spin trapping in intermediate states, as less likely. Our results show that the femtosecond dynamics of photocurrents allow us to differentiate the ISHE and the IEE and, more generally, provide important insights into the mechanisms of spin transport and SCI in $\mathcal{F}$ /TI stacks.

DOI: [10.1103/n1nw-2wyg](https://doi.org/10.1103/n1nw-2wyg)

I. INTRODUCTION

Spin transport and spin-charge-current interconversion (SCI) are important functionalities in future spintronic devices. To keep pace with the speed of other information carriers, such as photons and electrons [1], one needs to

push the speed of these functionalities to terahertz frequencies. An important spintronic model system is an $\mathcal{F}/\mathcal{N}$ stack consisting of a ferromagnetic metal layer $\mathcal{F}$ (e.g., Fe, Co, Ni) and a nonferromagnetic layer $\mathcal{N}$ (e.g., Pt) [2,3].

Two major SCI mechanisms in $\mathcal{F}/\mathcal{N}$ stacks are the inverse spin Hall effect (ISHE) and the inverse Edelstein effect (IEE). The ISHE occurs in the $\mathcal{N}$ and/or $\mathcal{F}$ bulk regions [Fig. 1(a)] and converts a longitudinal spin current of density j_s into a transverse charge current of density j_c proportional to j_s , i.e., $j_c \propto j_s$. For the IEE, in contrast, a charge current arises from a transient excess magnetization or spin accumulation μ_s^{IF} at an interface, e.g., between $\mathcal{F}$ and $\mathcal{N}$ [Fig. 1(b)], where the local inversion symmetry is

*Contact author: tobias.kampfrath@fu-berlin.de

Published by the American Physical Society under the terms of the [Creative Commons Attribution 4.0 International](https://creativecommons.org/licenses/by/4.0/) license. Further distribution of this work must maintain attribution to the author(s) and the published article's title, journal citation, and DOI.

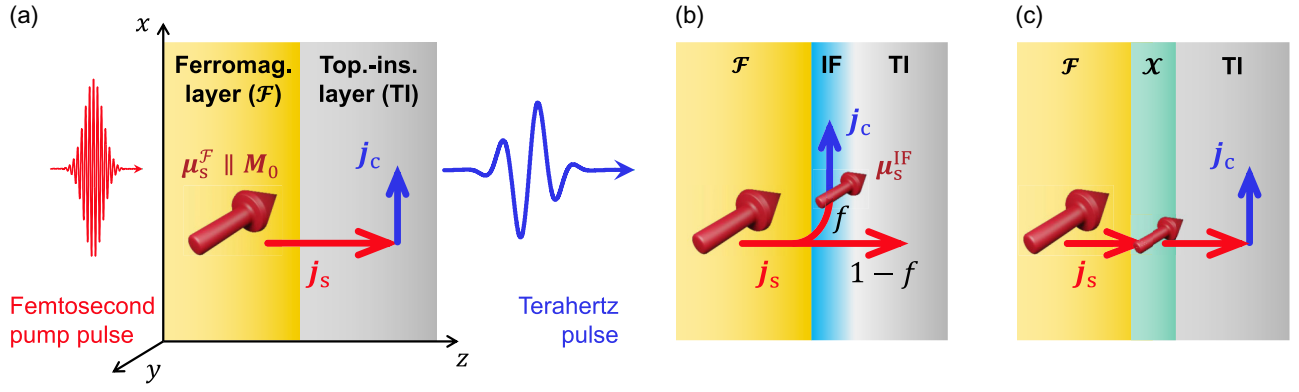


FIG. 1. Mechanisms of spin transport and spin-charge-current interconversion in $\mathcal{F}$ /TI stacks. (a) A spin current with density j_s flows from the ferromagnetic-metal layer $\mathcal{F}$ with magnetization $\mathbf{M}_0$ into a topological-insulator layer TI. j_s is converted into a transverse charge current with density j_c by the ISHE in the TI bulk, and (b) the IEE at the $\mathcal{F}$ /TI interface (IF), which hosts a transient spin accumulation μ_s^{IF} . Here f is the fraction of the total spin current j_s that is injected into the interfacial states and, thus, induces μ_s^{IF} . (c) Before the fraction $1 - f$ of j_s reaches the TI bulk, some of these spins may be trapped in an interlayer $\mathcal{X}$. As indicated in (a), the spin current is, in our experiment, driven by a femtosecond laser pulse through, e.g., the spin voltage $\mu_s^{\mathcal{F}}$ that arises from the optical heating of electrons in $\mathcal{F}$. The z -integrated charge current j_c is detected by measuring the emitted terahertz electromagnetic pulse.

broken. The IEE charge current is proportional to μ_s^{IF} , i.e., $j_c \propto \mu_s^{\text{IF}}$.

In three-dimensional topological insulators (TIs), such as Bi_2Te_3 [4], noteworthy contributions to SCI are expected from both the ISHE [5] and the IEE [6,7]. In particular, topologically protected surface states (TSS) and spin-momentum locking at the $\mathcal{F}$ /TI interface may lead to an enhanced IEE and, thus, a potential increase of SCI efficiencies [8,9]. At the same time, calculations predict that the contact between a ferromagnet $\mathcal{F}$, such as Co, and a TI, such as Bi_2Se_3 , shifts the TSS below the Fermi level, thereby potentially distorting the helical spin texture [10–12]. Note that Rashba-type states of two-dimensional electron gases can also contribute to SCI at interfaces. In the following, we use the term “IEE” for SCI by both TSS and topologically trivial Rashba states.

Importantly, as the $\mathcal{F}$ /TI interface is buried, its electronic structure around the Fermi energy and, thus, possible IEE features remain hidden to electron probes such as photoelectron emission spectroscopy [13]. Therefore, it has been a long-standing challenge to probe these states directly and unambiguously. Likewise, as the ISHE and IEE have identical macroscopic symmetry properties, they are hard to distinguish in transport experiments without measures such as stacking modification [14].

However, the ISHE and IEE may exhibit different temporal dynamics, which would, in principle, allow one to separate the two phenomena. For example, in heavy metals, the ISHE is expected to be instantaneous on a timescale of 50 fs and below because of the ultrashort relaxation time of electronic spin currents [15] and charge currents [16]. In contrast, the IEE has potentially slower dynamics because it requires an accumulation μ_s^{IF} of electron spin at the $\mathcal{F}$ / $\mathcal{N}$ interface [Fig. 1(b)] and, thus, a temporal

integration of the feeding spin current j_s . Consequently, we need to study SCI on the femtosecond timescale to look for possible temporal signatures of the IEE. Ultrafast studies on $\mathcal{F}$ /TI stacks based on terahertz emission spectroscopy have already been conducted [6,7,17,18], but they focused on signal amplitudes rather than dynamics.

In this work, we use femtosecond laser pulses to drive spin transport in $\mathcal{F}$ /TI stacks and $\mathcal{F}$ /Pt reference stacks, where we choose Co and Fe for $\mathcal{F}$. The resulting photocurrent j_c is measured by detecting the concomitantly emitted terahertz electromagnetic pulse [Fig. 1(a)]. We find that the photocurrent dynamics in $\mathcal{F}$ /TI contain an additional slower component relative to $\mathcal{F}$ /Pt. By assuming that j_c is predominantly caused by the optically induced transient spin voltage of $\mathcal{F}$, we infer a response function that summarizes the temporal response of spin transport and SCI following a fictitious $\delta(t)$ -like spin voltage. A noninstantaneous exponentially decaying response component with a time constant of 270 fs is found, which is independent of the choice (Co or Fe) of the $\mathcal{F}$ material. We consistently ascribe this contribution to noninstantaneous SCI by the IEE. The fraction of the spin current that is injected into the TSS is determined to be smaller than 10^{-2} . This small coupling is essential to achieve the relatively long relaxation time of 270 fs.

II. EXPERIMENTAL SETUP

To study spin transport and SCI on ultrafast timescales, we use broadband terahertz emission spectroscopy, as schematically shown in Fig. 1(a). In brief, a femtosecond laser pulse excites the sample, thereby driving an ultrafast longitudinal spin and, thus, transverse charge current that

is probed by measuring the terahertz electromagnetic pulse it emits.

A. Samples

Our samples, all grown on Si substrates, are $\mathcal{F}/\mathcal{N}$ or $\mathcal{N}/\mathcal{F}$ stacks with $\mathcal{F}$ being Co or Fe. For $\mathcal{N}$, we choose the TIs Bi_2Te_3 , SnBi_2Te_4 , and $\text{Bi}_{1-x}\text{Sb}_x$ ($x=0.15$ or $x=0.3$), and, as reference SCI material, Pt. Our main emphasis lies in the investigation of Bi_2Te_3 . Note that the structures containing Bi_2Te_3 or SnBi_2Te_4 are grown on top of an InAs seed layer (thickness 30 nm) on the Si substrate (see Supplemental Sec. 1 [19]). Samples are characterized by dc conductivity measurements with the use of a four-point probe system. More in-depth characterization of the same samples, e.g., by X-ray diffraction, angle-resolved photoelectron emission spectroscopy (ARPES), and scanning transmission electron microscopy, can be found in Refs. [17,18].

B. Terahertz emission spectroscopy

In the terahertz emission experiment [Fig. 1(a)], an external magnetic field of strength greater than 30 mT sets the direction of the static in-plane magnetization $\mathbf{M}_0$ of $\mathcal{F}$. An optical pump pulse (nominal duration 10 fs, center wavelength 800 nm, incident fluence up to 0.3 mJ/cm^2 , repetition rate 80 MHz) from a Ti:sapphire laser oscillator transiently heats the metallic heterostructure and induces a transient excess of spin angular momentum in $\mathcal{F}$ [15]. This so-called spin voltage $\mu_s^{\mathcal{F}}$ and, potentially, other forces such as temperature gradients [15] drive an ultrafast spin current with density j_s from $\mathcal{F}$ to $\mathcal{N}$. SCI due to, e.g., the ISHE and the IEE results in a transverse charge current j_c , which acts as a source of an electromagnetic pulse with frequencies extending into the terahertz range [20].

We measure the emitted terahertz electric field by electro-optic sampling with a linearly polarized gate pulse from the same laser in a GaP(110) crystal (thickness 250 μm ; see Supplemental Sec. 2 [19]) [21,22]. The resulting electro-optic signal $S(t, \mathbf{M}_0)$ is the terahertz-field-induced gate-pulse ellipticity as a function of the time delay t between the terahertz pulse and the gate pulse. We acquire terahertz signals for opposite $\mathcal{F}$ magnetizations $\pm \mathbf{M}_0$. Because we are interested in effects linear in the spin voltage $\mu_s^{\mathcal{F}}$, we focus on the signal component

$$S(t) = \frac{S(t, +\mathbf{M}_0) - S(t, -\mathbf{M}_0)}{2} \quad (1)$$

odd in $\mathbf{M}_0$. Signals even in $\mathbf{M}_0$ are typically 1 order of magnitude smaller than the odd-in- $\mathbf{M}_0$ signals for $\mathcal{F}/\mathcal{N}$ or $\mathcal{N}/\mathcal{F}$ stacks (see Supplemental Fig. S1 [19]). We observe no odd-in- $\mathbf{M}_0$ signal for a sample with $\mathcal{N} = \text{Bi}_2\text{Te}_3$ but without $\mathcal{F}$ (see Supplemental Fig. S2 [19]). Likewise, we confirm that the presence of an InAs seed layer between the substrate and

Fe has no impact on the signal dynamics obtained from the Fe/Pt reference sample (see Supplemental Fig. S2 [19]).

C. Terahertz-photocurrent extraction

The signal $S(t)$ contains electric-dipole and magnetic-dipole contributions [15]. The electric-dipole component arises from the charge current with sheet density

$$I_c(t) = \int dz j_c(z, t), \quad (2)$$

i.e., the charge-current density integrated along the stack normal [15,23]. A nonzero I_c requires broken inversion symmetry and includes the charge current due to SCI in the TI layer [Fig. 1(a)]. The magnetic-dipole contribution is induced predominantly by the optically induced magnetization dynamics in the ferromagnetic layer $\mathcal{F}$ [15,24–26]. It is ignored here because it is typically 1 order of magnitude smaller than the signals measured.

With this assumption, the terahertz signal in the frequency domain is given by the product [15]

$$\tilde{S}(\omega) = \tilde{H}_{SE}(\omega) \tilde{Z}(\omega) \tilde{I}_c(\omega), \quad (3)$$

where the tilde denotes Fourier transformation of the relevant quantity, and $\omega/2\pi$ is the frequency. The sample's sheet impedance $\tilde{Z}$ quantifies the conversion of $\tilde{I}_c$ into the terahertz electric field $\tilde{E} = \tilde{Z}\tilde{I}_c$ directly behind the sample. Finally, $\tilde{H}_{SE}$ captures the propagation of $\tilde{E}$ from the sample to the detector and the electro-optic detection process that result in the measured signal $\tilde{S}$.

By using Eq. (3), we can retrieve $\tilde{I}_c(\omega)$ and, thus, $I_c(t)$ [Eq. (2)] from the measured terahertz signal $\tilde{S}(\omega)$ [15,23,27]. To determine $\tilde{H}_{SE}(\omega)$, we choose the spintronic stack [28,29] W(2 nm)/CoFeB(1.8 nm)/Pt(2 nm) as a reference emitter, whose current evolution is known [15,30]. From terahertz time-domain reflection spectroscopy, we conclude that the sheet impedance $\tilde{Z}(\omega)$ has a negligible frequency dependence (see Supplemental Fig. S3 [19]). Therefore, we approximately determine $\tilde{Z}(\omega)$ from the dc sheet conductance (see Supplemental Table S1 [19]).

III. RESULTS AND DISCUSSION

A. Terahertz raw signals

Figure 2(a) shows normalized terahertz signals $S(t)$ odd in $\mathbf{M}_0$ from $\mathcal{F}(1.5 \text{ nm})/\text{Bi}_2\text{Te}_3(5 \text{ nm})$ and the $\mathcal{F}(2 \text{ nm})/\text{Pt}(4 \text{ nm})$ reference stack, where $\mathcal{F}$ is Co or Fe. For Co(1.5 nm)/ $\text{Bi}_2\text{Te}_3(5 \text{ nm})$, we confirm explicitly that the terahertz signal depends linearly on the pump power (see Supplemental Fig. S4 [19]). Note that we multiply the signals from the TI samples by -1 to account for the reverse growth order of $\mathcal{F}/\text{Pt}$ and $\text{Bi}_2\text{Te}_3/\mathcal{F}$.

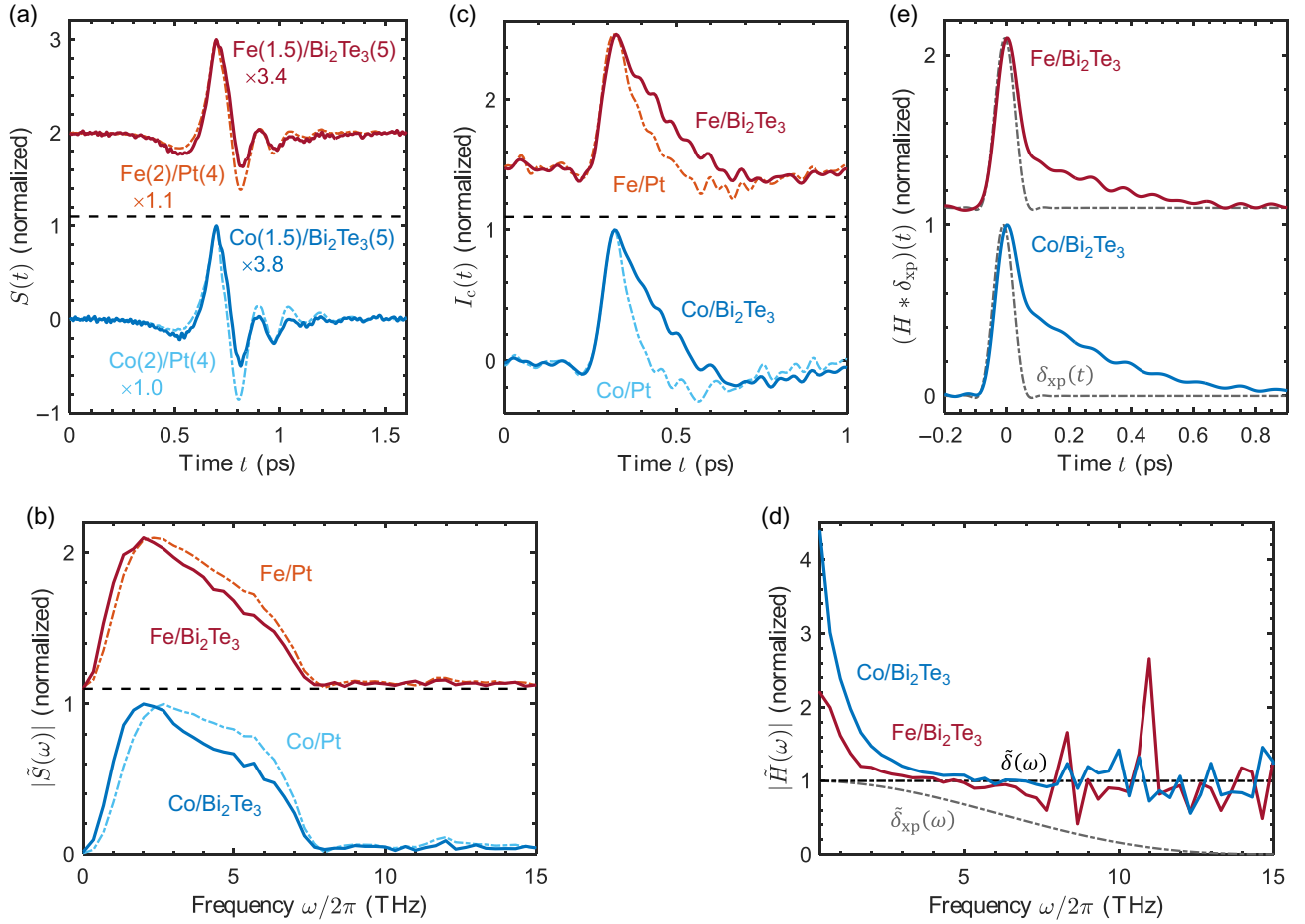


FIG. 2. From terahertz signals S to currents I_c to response functions H of $\mathcal{F}/\text{Bi}_2\text{Te}_3$ stacks. (a) Normalized electro-optic signals $S(t)$ of terahertz pulses emitted from the photoexcited samples. Solid lines show signals from $\mathcal{F}/\text{Bi}_2\text{Te}_3$, while dash-dotted lines indicate signals from the $\mathcal{F}/\text{Pt}$ reference, where $\mathcal{F}$ is Co or Fe. Layer thicknesses in nanometers are given as numbers in parentheses. All data are normalized to data for Co/Pt and scaled by the indicated factors. (b) Normalized magnitude of the Fourier amplitude $\tilde{S}(\omega)$ vs frequency $\omega/2\pi$ for the signals shown in (a). (c) Normalized charge-current sheet density $I_c(t)$ as extracted from the signals in (a) through Eq. (3). $I_c(t)$ of $\mathcal{F}/\text{Pt}$ has the same temporal dynamics as the transient spin voltage $\mu_s^{\mathcal{F}}(t)$ of $\mathcal{F} = \text{Fe}$ and $\mathcal{F} = \text{Co}$. (d) Fourier magnitude $|\tilde{H}(\omega)|$ of the $\mu_s^{\mathcal{F}}$ -to- I_c response function H [see Eqs. (5) and (6)], normalized to an average of 1 over the frequency interval 9–15 THz. The dash-dotted black line is the Fourier amplitude $\tilde{\delta}(\omega) = 1$ of the Dirac δ -function, whereas $\tilde{\delta}_{\text{xp}}(\omega)$ (dash-dotted gray line) accounts for the finite bandwidth of our setup. (e) Normalized $\mu_s^{\mathcal{F}}$ -to- I_c response function $(H * \delta_{\text{xp}})(t)$ in the time domain. It summarizes the spin-transport and SCI dynamics following an impulsive spin voltage $\mu_s^{\mathcal{F}}(t) \propto \delta_{\text{xp}}(t)$ (dash-dotted gray line), which is the inverse Fourier transformation of $\tilde{\delta}_{\text{xp}}(\omega)$ in (d). In some panels, waveforms are vertically offset for clarity and horizontally shifted such that signal maxima coincide.

While all waveforms in Fig. 2(a) have a somewhat similar shape, there are still notable qualitative differences. For example, the Co/Pt signal has sharper features than all other signals. This perception is reinforced by the magnitude of the Fourier-transformed signal $\tilde{S}(\omega)$ [Fig. 2(b)], which is enhanced at higher frequencies $\omega/2\pi$ within the accessible bandwidth compared with other signals. The spectral dip at $\omega/2\pi = 8$ THz is a characteristic feature of the electro-optic sampling in GaP [21].

B. Terahertz charge currents

To enable a more direct comparison of the four terahertz signals in Fig. 2(a), we extract the charge-current evolution $I_c(t)$ from $S(t)$ (Sec. II C) [15,23]. As seen in Fig. 2(c), $I_c(t)$ rises instantaneously within our time resolution for all samples, but decays fastest in Co/Pt, followed by Fe/Pt, Co/Bi₂Te₃, and Fe/Bi₂Te₃.

As shown previously [15], the photocurrent $I_c^{\text{ref}}(t)$ in the Fe/Pt and Co/Pt reference samples monitors the dynamics

of the spin voltage $\mu_s^{\mathcal{F}}(t)$ of $\mathcal{F}$, i.e.,

$$I_c^{\text{ref}}(t) \propto \mu_s^{\mathcal{F}}(t). \quad (4)$$

Figure 2(c) reveals that the spin voltage rises steplike for both $\mathcal{F} = \text{Co}$ and $\mathcal{F} = \text{Fe}$, but decays more rapidly in Co than in Fe. Assuming a biexponential-decay model [15], the dominant time constant, i.e., the electron-spin equilibration time, is 80 and 126 fs, respectively [31–34] (see Supplemental Sec. 3 [19]). This observation implies that spin relaxation in Co is substantially faster than in Fe [15].

C. Charge-current origin

Importantly, Fig. 2(c) indicates that the presence of $\mathcal{N} = \text{Bi}_2\text{Te}_3$ slows down the dynamics of the photocurrent relative to the presence of $\mathcal{N} = \text{Pt}$ for both choices of $\mathcal{F}$, i.e., Fe and Co. To understand this slowing down, we recall that for $\mathcal{N} = \text{Pt}$, the terahertz emission signal was previously shown to arise from a spin current j_s and its conversion into a charge current j_c , analogous to what is seen in Fig. 1(a) [15]. As we observe terahertz emission for both Co/Bi₂Te₃ and Fe/Bi₂Te₃, we assume an analogous two-step process for the Bi₂Te₃ samples and briefly review SCI and spin transport in the following sections.

1. SCI

The charge-current density j_c due to SCI generally scales linearly with the spin-current density $j_s(z, t)$ [Fig. 1(a)]. For metallic $\mathcal{N}$, SCI by the ISHE is quasi-instantaneous [16], resulting in $j_c(z, t) = \theta_{\text{SH}}(z)j_s(z, t)$, where the spin Hall angle θ_{SH} quantifies the efficiency of this kind of SCI.

In contrast, for the IEE, SCI is localized at the $\mathcal{F}/\mathcal{N}$ interface. The resulting interfacial charge-current sheet density I_c [Eq. (2)] is proportional to the instantaneous interfacial spin accumulation μ_s^{IF} through $I_c(t) \propto \mu_s^{\text{IF}}(t)$. Because a nonzero μ_s^{IF} requires an accumulation of the incident spin, we expect a time-integrating behavior of the feeding spin-current density j_s . In other words, $I_c(t)$ is expected to decay more slowly than $j_s(z, t)$, as also found in our measurements [Fig. 2(c)].

2. Spin transport in $\mathcal{F}/\text{TI}$

The spin current j_s (Fig. 1) can originate from a number of processes. First, a direct photoinduced shift-type spin transfer at the $\mathcal{F}/\mathcal{N}$ interface [35,36] would yield a very short derivativelike feature similar to $\partial\delta(t)/\partial t$ around the photocurrent onset [15], which is, however, not observed here [Fig. 2(c)]. We conclude that j_s arises from bandlike electron transport, which can, in general, be induced by gradients of electronic temperature (Seebeck-type contribution) or of spin voltage (pyrospintronic contribution) [15].

The Seebeck-type contribution is proportional to the difference between the electron temperature in $\mathcal{N}$ and the

magnon or electron temperature in $\mathcal{F}$ [15,23,27,37]. One can estimate the impact of the Seebeck-type contribution on j_s by increasing the pump fluence. As the electron heat capacity of $\mathcal{N} = \text{Bi}_2\text{Te}_3$ at 400 K is 30–40 times smaller than that of $\mathcal{F} = \text{Fe}$ and $\mathcal{F} = \text{Co}$ [15,32,38] (see Supplemental Table S2 [19]), the electronic temperature in Bi₂Te₃ directly after sample excitation has a much more sub-linear dependence on the pump fluence than in $\mathcal{F}$ (see Supplemental Fig. S5 and Supplemental Sec. 4 [19]). Consequently, the resulting Seebeck-type signal should depend very nonlinearly on the pump fluence for the fluences available in our experiment [39]. More precisely, the terahertz signal amplitude vs pump fluence would deviate from the initially linear trend by more than 70% already for an incident fluence of 0.1 mJ/cm². In contrast, we observe a highly linear dependence of the terahertz signal over the full range of pump fluences (see Supplemental Fig. S4 [19]). Therefore, a temperature difference between $\mathcal{F}$ and $\mathcal{N}$ contributes negligibly to j_s .

Thus, by exclusion, we deduce that the spin current in $\mathcal{F}/\text{Bi}_2\text{Te}_3$ with $\mathcal{F} = \text{Co}$ or $\mathcal{F} = \text{Fe}$ is solely driven by the pump-induced spin voltage in $\mathcal{F}$. Further, because the measured terahertz signals scale linearly with the pump fluence (see Supplemental Fig. S4 [19]), the spin-current density j_s , all spin accumulations, and the resulting charge-current density j_c are connected to the spin voltage $\mu_s^{\mathcal{F}}$ by a linear relationship. In particular, the charge-current sheet density I_c [Eq. (2)] is given by a linear response to the spin voltage $\mu_s^{\mathcal{F}}$ and, thus, is formally described by the convolution

$$I_c(t) = (H * \mu_s^{\mathcal{F}})(t) = \int dt' H(t - t') \mu_s^{\mathcal{F}}(t'). \quad (5)$$

3. Spin-transport and SCI response function

In Eq. (5), $H(t)$ can be understood as the charge current $I_c(t)$ that is induced by an impulsive spin voltage, i.e., $\mu_s^{\mathcal{F}}(t') = \delta(t')$, which has a much simpler temporal structure than the actual spin voltage [Fig. 2(c)] [30]. The impulse response function H summarizes all information on the subsequent spin-transport and SCI dynamics. We, thus, term it the “spin-transport and SCI response function” or “ $\mu_s^{\mathcal{F}}$ -to- I_c response function” of the given sample.

Importantly, any deviation of $H(t)$ from a $\delta(t)$ -like shape is a signature of noninstantaneous dynamics of spin transport and/or SCI [see Fig. 2(c)]. An example of a quasi-instantaneous process is spin transport into Pt, where the lifetime of j_s and any accumulation is less than 15 fs [40]. A noninstantaneous spin-current response is the trapping of spin-polarized electrons in a layer such as a MgO layer between $\mathcal{F}$ and $\mathcal{N}$ [30]. Because of its much simpler interpretation, we focus on the dynamics of H rather than of signals S [Fig. 2(a)] or currents I_c [Fig. 2(c)] in the following.

D. Analysis of response function H

1. Frequency domain

To extract the $\mu_s^{\mathcal{F}}$ -to- I_c response function H of an $\mathcal{F}/\text{Bi}_2\text{Te}_3$ stack, we Fourier-transform Eq. (5) into the frequency domain and obtain the simple product $\tilde{I}_c(\omega) = \tilde{H}(\omega)\tilde{\mu}_s^{\mathcal{F}}(\omega)$. For the $\mathcal{F}/\text{Pt}$ reference sample, one has $I_c^{\text{ref}}(t) \propto \mu_s^{\mathcal{F}}(t)$ [15,30], and, consequently, $\tilde{H}^{\text{ref}}(\omega)$ is constant over the whole bandwidth (0.3–15 THz) of our experiment. By applying Eq. (3) to both the sample signal $\tilde{S}(\omega)$ ($\mathcal{F}/\text{TI}$) and the reference signal $\tilde{S}^{\text{ref}}(\omega)$ ($\mathcal{F}/\text{Pt}$), we find

$$\tilde{H}(\omega) \propto \frac{\tilde{I}_c(\omega)}{\tilde{\mu}_s^{\mathcal{F}}(\omega)} \propto \frac{\tilde{S}(\omega)/\tilde{Z}(\omega)}{\tilde{S}^{\text{ref}}(\omega)/\tilde{Z}^{\text{ref}}(\omega)}, \quad (6)$$

where $\tilde{Z}^{\text{ref}}(\omega)$ is the terahertz sheet impedance of the $\mathcal{F}/\text{Pt}$ reference sample. Importantly, the instrument response function $\tilde{H}_{SE}(\omega)$ [Eq. (3)] cancels in Eq. (6). Further, $\tilde{Z}^{\text{ref}}(\omega)$ and $\tilde{Z}(\omega)$ are independent of frequency to a very good approximation (see Supplemental Fig. S3 [19]). Therefore, $\tilde{H}(\omega)$ is, apart from a global scaling factor, obtained by straightforward division of terahertz emission signals from the $\mathcal{F}/\text{Bi}_2\text{Te}_3$ and $\mathcal{F}/\text{Pt}$ samples in the frequency domain [Eq. (6)], without the need to measure any instrument response functions.

Note that $\tilde{S}(\omega)$ and $\tilde{S}^{\text{ref}}(\omega)$ exhibit a dip at 8 THz [Fig. 2(b)], which arises from a zero of the electro-optic response of our GaP detector [21]. To avoid division by the very small and, thus, relatively uncertain $\tilde{S}^{\text{ref}}(\omega)$ in Eq. (6), we determine $\tilde{H}(\omega)$ at 8 THz by interpolation of $\tilde{H}(\omega)$ at the adjacent frequencies.

The modulus of the extracted $\mu_s^{\mathcal{F}}$ -to- I_c response function $\tilde{H}(\omega)$ of $\text{Co}/\text{Bi}_2\text{Te}_3$ and $\text{Fe}/\text{Bi}_2\text{Te}_3$ is displayed in Fig. 2(d). The magnitude of $\tilde{H}(\omega)$ decreases from 0.3 to 5 THz and subsequently remains constant up to the maximum frequency covered of 15 THz. Therefore, the range 5–15 THz appears to be dominated by an instantaneous, scaled $\delta(t)$ -like response with constant Fourier transformation $\tilde{\delta}(\omega) = 1$ [Fig. 2(d)]. In contrast, in the interval 0.3–5 THz, $\tilde{H}(\omega)$ has significant variation, which suggests noninstantaneous contributions to the dynamics of spin current and/or SCI.

2. Time domain

It is highly instructive to consider the $\mu_s^{\mathcal{F}}$ -to- I_c response functions of $\text{Co}/\text{Bi}_2\text{Te}_3$ and $\text{Fe}/\text{Bi}_2\text{Te}_3$ in the time domain. Before the inverse Fourier transformation, we multiply $\tilde{H}(\omega)$ by a filter function $\tilde{\delta}_{xp}(\omega)$ to account for the finite usable bandwidth of our data [Fig. 2(d)]. We choose $\tilde{\delta}_{xp}(\omega)$ to be a Norton-Beer window function with a cutoff at $\omega/2\pi = \pm 15$ THz [41,42] because its time-domain counterpart $\delta_{xp}(t)$ is almost unipolar with a full width at half maximum of 74 fs [Fig. 2(e)]. By inverse Fourier transformation of the filtered $\tilde{H}(\omega)\tilde{\delta}_{xp}(\omega)$, we obtain $(H * \delta_{xp})(t)$

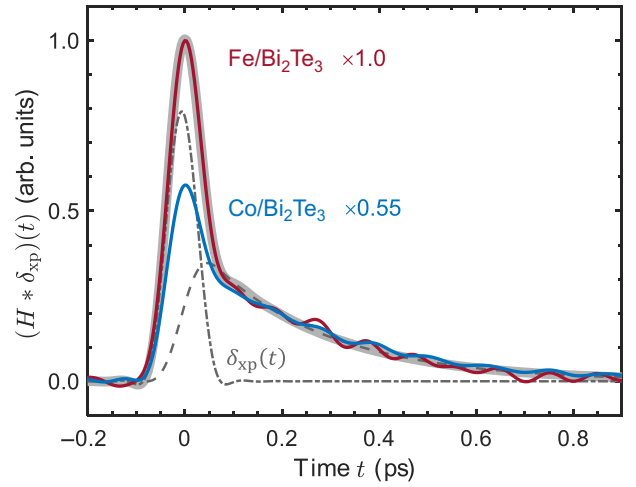


FIG. 3. Fit to the $\mu_s^{\mathcal{F}}$ -to- I_c response functions $(H * \delta_{xp})(t)$ of $\mathcal{F}/\text{Bi}_2\text{Te}_3$ with $\mathcal{F} = \text{Fe}$ and $\mathcal{F} = \text{Co}$ [see Fig. 2(e)]. The modeled H [Eq. (7)] for $\text{Fe}/\text{Bi}_2\text{Te}_3$ (thick gray line) contains the sum of a scaled instantaneous response $a\delta(t)$ (dash-dotted gray line) and a scaled single-sided exponential decay $bD_\tau(t)$ (dashed gray line). Both components include a convolution with the experimental δ -function $\delta_{xp}(t)$ [see Fig. 2(e)], which accounts for the finite bandwidth of our terahertz emission setup. The measured response functions are horizontally shifted such that signal maxima coincide. Note the scaling factors.

in the time domain. The convolution of the response function $H(t)$ with the unipolar peak $\delta_{xp}(t)$ possibly smoothens features of H that are finer than the width of δ_{xp} , but otherwise leaves the shape of H unchanged.

The extracted $(H * \delta_{xp})(t)$ of both $\text{Co}/\text{Bi}_2\text{Te}_3$ and $\text{Fe}/\text{Bi}_2\text{Te}_3$ is zero for $t < 0$ ps [Fig. 2(e)], as expected for a causal response. At time $t \approx 0$ ps, it turns into a peak with a shape similar to $\delta_{xp}(t)$, which characterizes the time resolution with which H is determined. Importantly, the scaled relaxation tail of $H * \delta_{xp}$ is identical within the accuracy of our experiment for both $\text{Co}/\text{Bi}_2\text{Te}_3$ and $\text{Fe}/\text{Bi}_2\text{Te}_3$ (Fig. 3).

Consequently, we tentatively parameterize the measured response function by

$$H(t) = a\delta(t) + bD_\tau(t) \text{ with } D_\tau(t) = \frac{1}{\tau}\Theta(t)e^{-t/\tau}. \quad (7)$$

It is the sum of a δ -function and a single-sided exponential decay $D_\tau(t)$ with $\int dt D_\tau(t) = 1$, where $\Theta(t)$ denotes the Heaviside step function. The weight coefficients a and b and the time constant τ are free parameters. Figure 3 shows that a fit using $H * \delta_{xp}$ with H from Eq. (7) excellently reproduces our experimental results for $\tau \approx 270 \pm 20$ fs for the $\mathcal{F}/\text{TI}$ stacks with the TI Bi_2Te_3 .

We summarize this significant result. Within our experimental accuracy, the charge-current response $H(t)$ induced by an impulsive spin voltage $\mu_s^{\mathcal{F}}(t) = \delta(t)$ in $\mathcal{F}$ is the sum of an instantaneous response $[a\delta(t)]$ and an exponential decay $[bD_\tau(t)]$. Remarkably, when the spin-current

source $\mathcal{F} = \text{Co}$ is replaced by Fe, only the amplitudes a and b change, whereas the changes in the relaxation time τ are minor. This finding indicates that the photocurrent $I_c(t)$ arises from a superposition of two processes of spin transport and/or SCI in Bi_2Te_3 rather than in $\mathcal{F}$. The first process $[a\delta(t)]$ is instantaneous within our time resolution, whereas the second one $[bD_\tau(t)]$ has a relaxation time τ of approximately 270 fs.

Supplemental Figure S6 [19] shows the corresponding response function $(H * \delta_{\text{xp}})(t)$ for $\text{Co}/\text{SnBi}_2\text{Te}_4$ and $\text{Co}/\text{Bi}_{1-x}\text{Sb}_x$ samples with differing thickness and composition ($x = 0.15$ and $x = 0.3$). SnBi_2Te_4 displays very similar dynamics in comparison with Bi_2Te_3 , in line with the model of Eq. (7). In contrast, $\text{Bi}_{1-x}\text{Sb}_x$ (5 nm) shows a faster and more complex response (see Supplemental Sec. 5 [19]).

E. Interpretation: ISHE and IEE

On the basis of the previous analysis and discussion, we suggest the following microscopic scenario to explain our observations. First, if we assume a δ -like spin voltage, it drives a δ -like spin current across the $\mathcal{F}/\text{TI}$ interface [Fig. 1(b)]. A fraction f of this current ($0 \leq f \leq 1$) is coupled into the interfacial TSS and/or Rashba-type states, whereas the remaining part $1 - f$ flows into the TI bulk [Fig. 1(b)].

We tentatively assign $H_{\text{ISHE}} := a\delta$ in Eq. (7) to the ISHE in the bulk of $\mathcal{F}$ and Bi_2Te_3 , whereas $H_{\text{IEE}} := bD_\tau$ is ascribed to the IEE at the $\mathcal{F}/\text{Bi}_2\text{Te}_3$ interface, which is slower because of the required spin accumulation μ_s^{IF} [Fig. 1(b)]. The coefficients a and b are proportional to $1 - f$ and f , respectively. We, thus, have

$$H(t) = H_{\text{ISHE}}(t) + H_{\text{IEE}}(t). \quad (8)$$

Importantly, this interpretation of our measured $\mu_s^{\mathcal{F}}$ -to- I_c response function H is consistent with respect to the sign, magnitude, and temporal shape of the ISHE and IEE, as discussed in the following sections.

1. ISHE

The δ -like spin voltage in $\mathcal{F}$ injects spin-polarized electrons into the TI bulk, where they propagate with the Fermi velocity v_F^{B} into the depth of the film. This ballistic current relaxes on the timescale of the bulk momentum-relaxation time τ^{B} and is, simultaneously, converted into an in-plane charge current by the ISHE. Therefore, H_{ISHE} is proportional to the product $(1 - f)v_F^{\text{B}}\tau^{\text{B}}\theta_{\text{SH}}$, where θ_{SH} is the spin Hall angle of the TI and relaxes according to $\Theta(t)e^{-t/\tau^{\text{B}}} \approx \tau^{\text{B}}\delta(t)$. The latter approximation is justified because the bulk Drude scattering time τ^{B} [16] is significantly shorter than our time resolution of 74 fs. Examples include ferromagnets such as Fe ($\tau^{\text{B}} = 30$ fs) [43,44] and typical TIs

such as Bi_2Te_3 ($\tau^{\text{B}} < 40$ fs) [45]. We, therefore, obtain

$$H_{\text{ISHE}}(t) \propto (1 - f)v_F^{\text{B}}\tau^{\text{B}}\theta_{\text{SH}}\delta(t) \quad (9)$$

in the ballistic limit, which is relevant on ultrafast timescales [46]. We note that Eq. (9) is fully consistent with the $a\delta$ term in Eq. (7). Further, the sign of a and, therefore, θ_{SH} is positive, analogous to that for Pt [Fig. 2(c)]. This finding agrees with the sign of the spin Hall angle from previous studies on Bi_2Te_3 grown by molecular-beam epitaxy [47]. We note that a possible ISHE in Fe and Co may contribute to a , but the ISHE in TIs dominates because the spin Hall angle of Fe and Co is negative relative to that of Pt [48].

2. IEE

Here the δ -like spin voltage in $\mathcal{F}$ injects spin angular momentum into TSS or Rashba-type states of the $\mathcal{F}/\text{TI}$ interface [Fig. 1(b)]. Because of spin-momentum locking of these states, each excess spin parallel to $\mathbf{M}_0$ is accompanied by one electron moving with the interface Fermi velocity v_F^{IF} in the interface plane but perpendicular to $\mathbf{M}_0$ [10]. Therefore, the IEE-related charge current H_{IEE} of TSS scales with $f v_F^{\text{IF}}$.

To model the dynamics of H_{IEE} , we note that the total electron spin in the $\mathcal{F}/\text{TI}$ interface is proportional to the interface spin accumulation μ_s^{IF} . The latter decays with a rate $\partial\mu_s^{\text{IF}}/\partial t = -\mu_s^{\text{IF}}/\tau^{\text{IF}}$ due, e.g., to spin-conserving transfer (Tr) into $\mathcal{F}$ or TI bulk states or, possibly, spin-flip (SF) scattering. We obtain $\mu_s^{\text{IF}}(t) \propto \Theta(t)e^{-t/\tau^{\text{IF}}}$ with a total inverse time constant $(\tau^{\text{IF}})^{-1} = \tau_{\text{Tr}}^{-1} + \tau_{\text{SF}}^{-1}$. Therefore, we have

$$H_{\text{IEE}}(t) \propto f v_F^{\text{IF}} \tau^{\text{IF}} D_\tau(t). \quad (10)$$

Equation (10) is in line with the bD_τ term in Eq. (7). Moreover, the sign of b and, thus, the assumed IEE is positive, analogous to the sign of the ISHE of Pt. This finding may suggest that bD_τ in Eq. (7) arises from the IEE of the TSS rather than trivial Rashba states [6]. The reason is that these two kinds of states usually contribute with opposite sign to SCI, as inferred from ARPES measurements on similar TIs such as Bi_2Se_3 and from spin-transfer ferromagnetic-resonance measurements on $(\text{Bi}_{0.4}\text{Sb}_{0.6})_2\text{Te}_3$ [49–51]. However, exceptions to this notion have already been observed [52].

Remarkably, the characteristic time τ^{IF} of 270 fs found in our experiment (Fig. 3) is compatible with interface spin relaxation times measured by time-resolved ARPES in Sb_2Te_3 samples under similar excitation conditions [53,54]. Further, terahertz-transmission and terahertz-reflection spectroscopy studies of Bi_2Te_3 suggest momentum scattering times of around 200 fs for the surface states at room temperature [45,55]. We emphasize, however,

that these time constants were determined for TI samples without any adjacent layer $\mathcal{F}$.

3. Estimation of f

It is instructive to estimate the fraction f of the spin current from $\mathcal{F}$ to TI that is coupled into the TSS [Fig. 1(b)]. To this end, we note that in the measured $H * \delta_{xp}$, the area under the peak ($a\delta$) and the area under the relaxation tail (bD_τ) are roughly equal (Fig. 3). This observation along with our interpretation implies $\int dt H_{\text{ISHE}}(t) \sim \int dt H_{\text{IEE}}(t)$. By using Eqs. (9) and (10), we conclude that

$$\frac{f}{1-f} \sim \frac{v_F^B \tau^B \theta_{\text{SH}}}{v_F^{\text{IF}} \tau^{\text{IF}}}. \quad (11)$$

To estimate f , we take the reported bulk Drude scattering time $\tau^B < 40$ fs of Bi_2Te_3 [45], while we use $\tau^{\text{IF}} = 270$ fs as found in our experiment (Fig. 3). The Fermi velocity v_F^{IF} associated with the TSS is 0.4 nm/fs [56,57], and for the bulk Fermi velocity, we make the reasonable assumption $v_F^B < v_F^{\text{IF}}$. Together with the calculated spin Hall angle $\theta_{\text{SH}} = 0.06$ of Bi_2Te_3 [58], Eq. (11) yields $f < 10^{-2}$.

Interestingly, the inferred $f \ll 1$ implies a relatively small probability that an electron spin traversing the interface from $\mathcal{F}$ to TI is transferred into the TSS. It, thus, suggests a relatively weak coupling of $\mathcal{F}$ and the TSS of the adjacent TI. This conclusion is consistent with the long lifetime $\tau^{\text{IF}} = 270$ fs, which is comparable to the lifetime for Bi_2Te_3 samples without any layer on top [45,55].

4. Static case

As a final check, we extend our considerations to the quasistatic regime and calculate the SCI efficiency for the IEE as $\lambda_{\text{IEE}} = \int dt I_c / \int dt j_s = \int dt H_{\text{IEE}}(t)$. We find $\lambda_{\text{IEE}} = f v_F^{\text{IF}} \tau^{\text{IF}}$, in agreement with the result $\lambda_{\text{IEE}} = v_F^{\text{IF}} \tau'$ for static currents [59], if we identify $\tau' = f \tau^{\text{IF}}$. Note, however, that the physically meaningful time constant is the spin relaxation time τ^{IF} of the TSS, and not the much smaller τ' , which was determined in previous work [60,61].

For $v_F^{\text{IF}} = 0.4$ nm/fs [56,57], $f < 10^{-2}$, and $\tau^{\text{IF}} = 270$ fs as inferred above, we obtain $\lambda_{\text{IEE}} < 1$ nm. This value is consistent with $\lambda_{\text{IEE}} \sim 0.1$ nm [60–62] as obtained in experiments with spin pumping of j_s and contact-based detection of the resulting static current I_c for similar TIs [8,9,63].

In summary, our assignment of $a\delta$ in Eq. (7) to the ISHE in the bulk of $\mathcal{F}$ and Bi_2Te_3 [Fig. 1(a)] and of bD_τ to the IEE at the $\mathcal{F}/\text{Bi}_2\text{Te}_3$ interface [Fig. 1(b)] is highly consistent with respect to the amplitude and the shape of the dynamics of the measured response function $H * \delta_{xp}$ (Fig. 3).

F. Alternative contributions and mechanisms

Even though our interpretation of our experimental results is consistent, other mechanisms may also explain the observed current dynamics in Fig. 2(c). According to Eq. (5) and the remarks preceding it, a noninstantaneous modification $D_\tau(t)$ [Eq. (7)] of the photocurrent relative to the reference $\mathcal{N} = \text{Pt}$ may arise not only from (1) changes in the SCI as discussed in Sec. III E but also from (2) changes in the spin-voltage dynamics and (3) the spin-current propagation.

Regarding scenario (2), the spin-voltage dynamics $\mu_s^{\mathcal{F}}(t)$ in $\mathcal{F}$ may change when $\mathcal{N} = \text{Pt}$ is replaced by Bi_2Te_3 due to a modified spin transfer from $\mathcal{F}$ to $\mathcal{N}$, thereby invalidating Eq. (6). This scenario is unlikely because spin transport into $\mathcal{N}$ was shown to only negligibly change the spin-voltage dynamics of $\mathcal{F}$ [15]. Alternatively, $\mu_s^{\mathcal{F}}(t)$ can be modified by a change in the electron-temperature dynamics in $\mathcal{F}$ due to heat flow from $\mathcal{N}$ to $\mathcal{F}$. In this case, the dynamics would sensitively depend on the difference of electronic temperature between $\mathcal{F}$ and $\mathcal{N} = \text{Bi}_2\text{Te}_3$. As discussed in Sec. III C, a highly non-linear fluence dependence of the terahertz signal would result from this hypothesis, which is not observed. A major reason why the heat flow from $\mathcal{N} = \text{Bi}_2\text{Te}_3$ to $\mathcal{F}$ is not relevant for the temperature dynamics in $\mathcal{F}$ is that the electronic heat capacity and electron-phonon coupling of Bi_2Te_3 are significantly smaller than those of $\mathcal{F} = \text{Co}$ and $\mathcal{F} = \text{Fe}$ (see Supplemental Sec. 4 [19]).

Concerning scenario (3), the noninstantaneous contribution D_τ may arise from a longer-lived ballistic spin-current propagation in $\mathcal{N} = \text{Bi}_2\text{Te}_3$. This assumption is, however, inconsistent with previous work [5] that found that $j_s(z, t)$ in Bi_2Te_3 relaxes over as little as 2 nm. Moreover, the characteristic bulk Drude scattering time of Bi_2Te_3 is less than 40 fs and, thus, below $\tau = 270$ fs [45]. Alternatively, D_τ may result from the buildup of a longer-lived spin accumulation in the bulk of $\mathcal{N} = \text{Bi}_2\text{Te}_3$. In this scenario, a non-negative δ -like spin voltage in $\mathcal{F}$ would, by transport, lead to a non-negative spin voltage in $\mathcal{N}$. The latter, in turn, would drive spin back to $\mathcal{F}$ and imply a longer-lived spin-current component, but with opposite sign compared with $a\delta$, which is not observed.

A final option for scenario (3) would rely on structural imperfections of the $\mathcal{F}/\text{Bi}_2\text{Te}_3$ sample, i.e., a third layer $\mathcal{X}$ between $\mathcal{F} = \text{Co}$ or $\mathcal{F} = \text{Fe}$ and $\mathcal{N} = \text{Bi}_2\text{Te}_3$ [Fig. 1(c)]. The emerging localized states of $\mathcal{X}$ could partially trap spin-polarized electrons from $\mathcal{F}$ and later release them into $\mathcal{N}$. Indeed, such relaxation behavior on timescales of 100 fs was recently observed for MgO tunnel barriers between CoFeB and Pt thin films [30]. In our $\text{Co}/\text{Bi}_2\text{Te}_3$ sample, the existence of a third intermixing layer $\mathcal{X}$ is not implausible: Previous studies observed the formation of a magnetically dead layer and interdiffusion between ferromagnets such as Fe or Co and TIs such as Bi_2Se_3 [64,65] or Sb_2Te_3 [66,67]. Likewise, for $\text{Co}/\text{Bi}_2\text{Te}_3$, the formation

of interfacial CoTe₂ was proposed [68]. Similarly, at interfaces between ferromagnetic-metal and organic-molecule layers, spins can be trapped for up to 1 ps in hybrid interface states [69]. Possibly, such trapping could even occur in $\mathcal{F}$ /TI interface states with negligible SCI. In any case, one would expect that the trap layer $\mathcal{X}$ and, thus, the trapping time depend sensitively on the adjacent layer $\mathcal{F}$. However, as an almost identical time constant τ of approximately 270 ± 20 fs of the delayed feature D_τ is observed for both $\mathcal{F} = \text{Co}$ and $\mathcal{F} = \text{Fe}$ (Fig. 3), we consider scenario (3) as rather unlikely.

A short discussion of the $\mu_s^{\mathcal{F}}$ -to- I_c response functions of Co/SnBi₂Te₄ and Co/Bi_{1-x}Sb_x can be found in Supplemental Sec. 5 [19].

IV. CONCLUSIONS

In conclusion, we use terahertz emission spectroscopy to investigate ultrafast spin-transport and SCI dynamics in $\mathcal{F}$ /Bi₂Te₃ stacks. Extraction of a suitable response function permits a detailed quantitative analysis of these processes on the basis of their temporal signatures. It reveals a non-instantaneous relaxation component with a relaxation time of 270 fs that is independent of the $\mathcal{F}$ material chosen, Fe or Co. It can consistently be ascribed to spin accumulation and SCI by the IEE at the $\mathcal{F}$ /TI interface [Fig. 1(b)].

In general, on the one hand, our study may serve as a blueprint to disentangle the complex spin transport and SCI responses in spintronic multilayers and, more generally, other conversion processes, such as trap and release [Fig. 1(c)]. On the other hand, this work reveals an approach toward emitters and detectors of broadband terahertz electromagnetic radiation [70,71] whose response can be tailored by interface tuning.

ACKNOWLEDGMENTS

We thank Sergio Valenzuela for fruitful discussions. We acknowledge funding by the European Union and the European Research Council (ERC) through the Future and Emerging Technologies project SKYTOP (Grant No. 824123), the ERC 2023 Proof-of-Concept Grant T-SPINDEX (Grant No. 101123255), the ERC 2023 Advanced Grant ORBITERA (Grant No. 101142285), and the German Research Foundation through the Collaborative Research Center SFB TRR 227 “Ultrafast spin dynamics” (Project No. 328545488, projects A05 and B02) and the Cluster of Excellence EXC 3112 “Center for Chiral Electronics” (EXC 3112/1, Project No. 533767171).

DATA AVAILABILITY

The data that support the findings of this article are openly available [72].

- [1] E. Y. Vedmedenko, R. K. Kawakami, D. D. Sheka, P. Gambardella, A. Kirilyuk, A. Hirohata, C. Binek, O. Chubykalo-Fesenko, S. Sanvito, B. J. Kirby, *et al.*, The 2020 magnetism roadmap, *J. Phys. D: Appl. Phys.* **53**, 453001 (2020).
- [2] R. Schneider, M. Fix, R. Heming, S. Michaelis de Vasconcellos, M. Albrecht, and R. Bratschkitsch, Magnetic-field-dependent THz emission of spintronic TbFe/Pt layers, *ACS Photonics* **5**, 3936 (2018).
- [3] Y. Sasaki, Y. Kota, S. Iihama, K. Z. Suzuki, A. Sakuma, and S. Mizukami, Effect of Co and Fe stoichiometry on terahertz emission from Ta/(Co_xFe_{1-x})₈₀B₂₀/MgO thin films, *Phys. Rev. B* **100**, 140406 (2019).
- [4] E. Longo, L. Locatelli, P. Tsipas, A. Lintzeris, A. Dimoulas, M. Fanciulli, M. Longo, and R. Mantovan, Exploiting the close-to-Dirac point shift of the Fermi level in the Sb₂Te₃/Bi₂Te₃ topological insulator heterostructure for spin-charge conversion, *ACS Appl. Mater. Interfaces* **15**, 50237 (2023).
- [5] V. Sharma, R. Nepal, W. Wu, E. A. Pogue, R. Kumar, R. Kolagani, L. Gundlach, M. B. Jungfleisch, and R. C. Budhani, Comparing spin injection in Fe₇₅Co₂₅/Bi₂Te₃ at GHz and optical excitations, *Appl. Phys. Lett.* **122**, 072403 (2023).
- [6] X. Chen, H. Wang, H. Liu, C. Wang, G. Wei, C. Fang, H. Wang, C. Geng, S. Liu, P. Li, *et al.*, Generation and control of terahertz spin currents in topology-induced 2D ferromagnetic Fe₃GeTe₂/Bi₂Te₃ heterostructures, *Adv. Mater.* **34**, 2106172 (2022).
- [7] X. Wang, L. Cheng, D. Zhu, Y. Wu, M. Chen, Y. Wang, D. Zhao, C. B. Boothroyd, Y. M. Lam, J.-X. Zhu, *et al.*, Ultrafast spin-to-charge conversion at the surface of topological insulator thin films, *Adv. Mater.* **30**, 1802356 (2018).
- [8] E. Longo, L. Locatelli, M. Belli, M. Alia, A. Kumar, M. Longo, M. Fanciulli, and R. Mantovan, Spin-Charge Conversion in Fe/Au/Sb₂Te₃ Heterostructures as probed by spin pumping ferromagnetic resonance, *Adv. Mater. Interfaces* **8**, 2101244 (2021).
- [9] E. Longo, M. Belli, M. Alia, M. Rimoldi, R. Cecchini, M. Longo, C. Wiemer, L. Locatelli, P. Tsipas, A. Dimoulas, *et al.*, Large spin-to-charge conversion at room temperature in extended epitaxial Sb₂Te₃ Topological insulator chemically grown on silicon, *Adv. Funct. Mater.* **32**, 2109361 (2022).
- [10] A. Manchon, J. Železný, I. M. Miron, T. Jungwirth, J. Sinova, A. Thiaville, K. Garello, and P. Gambardella, Current-induced spin-orbit torques in ferromagnetic and antiferromagnetic systems, *Rev. Mod. Phys.* **91**, 035004 (2019).
- [11] J. M. Marmolejo-Tejada, K. Dolui, P. Lazić, P.-H. Chang, S. Smidstrup, D. Stradi, K. Stokbro, and B. K. Nikolić, Proximity band structure and spin textures on both sides of topological-insulator/ferromagnetic-metal interface and their charge transport probes, *Nano Lett.* **17**, 5626 (2017).
- [12] J. Zhang, J. P. Velez, X. Dang, and E. Y. Tsybmal, Band structure and spin texture of Bi₂Se₃ 3d ferromagnetic metal interface, *Phys. Rev. B* **94**, 014435 (2016).
- [13] A. Dimoulas, Perspectives for the growth of epitaxial 2D van der Waals layers with an emphasis on ferromagnetic metals for spintronics, *Adv. Mater. Interfaces* **9**, 2201469 (2022).

- [14] J. Shen, Z. Feng, P. Xu, D. Hou, Y. Gao, and X. Jin, Spin-to-charge conversion in Ag/Bi bilayer revisited, *Phys. Rev. Lett.* **126**, 197201 (2021).
- [15] R. Rouzegar, L. Brandt, L. Nádvorník, D. A. Reiss, A. L. Chekhov, O. Gueckstock, C. In, M. Wolf, T. S. Seifert, P. W. Brouwer, *et al.*, Laser-induced terahertz spin transport in magnetic nanostructures arises from the same force as ultrafast demagnetization, *Phys. Rev. B* **106**, 144427 (2022).
- [16] T. S. Seifert, U. Martens, F. Radu, M. Ribow, M. Berritta, L. Nádvorník, R. Starke, T. Jungwirth, M. Wolf, I. Radu, *et al.*, Frequency-independent terahertz anomalous Hall effect in DyCo₅, Co₃₂Fe₆₈, and Gd₂₇Fe₇₃ thin films from DC to 40 THz, *Adv. Mater.* **33**, 2007398 (2021).
- [17] E. Rongione, S. Fragkos, L. Baringthon, J. Hawecker, E. Xenogiannopoulou, P. Tsipas, C. Song, M. Mićica, J. Mangeney, J. Tignon, *et al.*, Ultrafast spin-charge conversion at SnBi₂Te₄/Co topological insulator interfaces probed by terahertz emission spectroscopy, *Adv. Opt. Mater.* **10**, 2102061 (2022).
- [18] E. Rongione, L. Baringthon, D. She, G. Patriarche, R. Lebrun, A. Lemaître, M. Morassi, N. Reyren, M. Mićica, J. Mangeney, *et al.*, Spin-momentum locking and ultrafast spin-charge conversion in ultrathin epitaxial Bi_{1-x}Sb_x topological insulator, *Adv. Sci.* **10**, 2301124 (2023).
- [19] See the Supplemental Material at <http://link.aps.org/supplemental/10.1103/nlnw-2wyg>, which includes Refs. [73–83], for details of sample preparation and characterization; terahertz emission spectroscopy, terahertz emission signals, and signal pump fluence dependence; terahertz reflectivity signals of Bi₂Te₃/Co, dc sheet conductance and terahertz impedance; current-relaxation dynamics of reference samples, electron-phonon coupling parameters, and electron and lattice heat capacities; electron-temperature dynamics; and response function of Co/SnBi₂Te₄ and Co/Bi_{1-x}Sb_x and interpretation.
- [20] T. Kampfrath, M. Battiato, P. Maldonado, G. Eilers, J. Nötzold, S. Mährlein, V. Zbarsky, F. Freimuth, Y. Mokrousov, S. Blügel, *et al.*, Terahertz spin current pulses controlled by magnetic heterostructures, *Nat. Nanotechnol.* **8**, 256 (2013).
- [21] A. Leitenstorfer, S. Hunsche, J. Shah, M. C. Nuss, and W. H. Knox, Detectors and sources for ultrabroadband electro-optic sampling: Experiment and theory, *Appl. Phys. Lett.* **74**, 1516 (1999).
- [22] T. Kampfrath, J. Nötzold, and M. Wolf, Sampling of broadband terahertz pulses with thick electro-optic crystals, *Appl. Phys. Lett.* **90**, 231113 (2007).
- [23] T. S. Seifert, S. Jaiswal, J. Barker, S. T. Weber, I. Razdolski, J. Cramer, O. Gueckstock, S. F. Mährlein, L. Nádvorník, S. Watanabe, *et al.*, Femtosecond formation dynamics of the spin Seebeck effect revealed by terahertz spectroscopy, *Nat. Commun.* **9**, 2899 (2018).
- [24] E. Beaupaire, G. M. Turner, S. M. Harrel, M. C. Beard, J.-Y. Bigot, and C. A. Schmuttenmaer, Coherent terahertz emission from ferromagnetic films excited by femtosecond laser pulses, *Appl. Phys. Lett.* **84**, 3465 (2004).
- [25] W. Zhang, P. Maldonado, Z. Jin, T. S. Seifert, J. Arabski, G. Schmerber, E. Beaupaire, M. Bonn, T. Kampfrath, P. M. Oppeneer, *et al.*, Ultrafast terahertz magnetometry, *Nat. Commun.* **11**, 4247 (2020).
- [26] T. J. Huisman, R. V. Mikhaylovskiy, A. Tsukamoto, T. Rasing, and A. V. Kimel, Simultaneous measurements of terahertz emission and magneto-optical Kerr effect for resolving ultrafast laser-induced demagnetization dynamics, *Phys. Rev. B* **92**, 104419 (2015).
- [27] P. Jiménez-Cavero, O. Gueckstock, L. Nádvorník, I. Lucas, T. S. Seifert, M. Wolf, R. Rouzegar, P. W. Brouwer, S. Becker, G. Jakob, *et al.*, Transition of laser-induced terahertz spin currents from torque- to conduction-electron-mediated transport, *Phys. Rev. B* **105**, 184408 (2022).
- [28] T. Seifert, S. Jaiswal, U. Martens, J. Hannegan, L. Braun, P. Maldonado, F. Freimuth, A. Kronenberg, J. Henrizi, I. Radu, *et al.*, Efficient metallic spintronic emitters of ultrabroadband terahertz radiation, *Nat. Photonics* **10**, 483 (2016).
- [29] R. Rouzegar, A. L. Chekhov, Y. Behovits, B. R. Serrano, M. A. Syskaki, C. H. Lambert, D. Engel, U. Martens, M. Münzenberg, M. Wolf, *et al.*, Broadband spintronic terahertz source with peak electric fields exceeding 1.5 MV/cm, *Phys. Rev. Appl.* **19**, 034018 (2023).
- [30] R. Rouzegar, M. A. Wahada, A. L. Chekhov, W. Hoppe, G. Bierhance, J. Jechumtál, L. Nádvorník, M. Wolf, T. S. Seifert, S. S. P. Parkin, *et al.*, Terahertz spin-conductance spectroscopy: probing coherent and incoherent ultrafast spin tunneling, *Nano Lett.* **24**, 7852 (2024).
- [31] D. Zahn, F. Jakobs, H. Seiler, T. A. Butcher, D. Engel, J. Vorberger, U. Atxitia, Y. W. Windsor, and R. Ernstorfer, Intrinsic energy flow in laser-excited 3d ferromagnets, *Phys. Rev. Res.* **4**, 013104 (2022).
- [32] G. R. Stewart, Measurement of low-temperature specific heat, *Rev. Sci. Instrum.* **54**, 1 (1983).
- [33] D. Zahn, H. Seiler, Y. W. Windsor, and R. Ernstorfer, Ultrafast lattice dynamics and electron-phonon coupling in platinum extracted with a global fitting approach for time-resolved polycrystalline diffraction data, *Struct. Dyn.* **8**, 064301 (2021).
- [34] H. Jang, L. Marnitz, T. Huebner, J. Kimling, T. Kuschel, and D. G. Cahill, Thermal conductivity of oxide tunnel barriers in magnetic tunnel junctions measured by ultrafast thermoreflectance and magneto-optic Kerr effect thermometry, *Phys. Rev. Appl.* **13**, 024007 (2020).
- [35] J. K. Dewhurst, P. Elliott, S. Shallcross, E. K. U. Gross, and S. Sharma, Laser-induced intersite spin transfer, *Nano Lett.* **18**, 1842 (2018).
- [36] F. Willems, C. von Korff Schmising, C. Strüber, D. Schick, D. W. Engel, J. K. Dewhurst, P. Elliott, S. Sharma, and S. Eisebitt, Optical inter-site spin transfer probed by energy and spin-resolved transient absorption spectroscopy, *Nat. Commun.* **11**, 871 (2020).
- [37] F. N. Kholid, D. Hamara, M. Terschanski, F. Mertens, D. Bossini, M. Cinchetti, L. McKenzie-Sell, J. Patchett, D. Petit, R. Cowburn, *et al.*, Temperature dependence of the picosecond spin Seebeck effect, *Appl. Phys. Lett.* **119**, 032401 (2021).
- [38] M. Hajlaoui, E. Papalazarou, J. Mauchain, G. Lantz, N. Moisan, D. Boschetto, Z. Jiang, I. Miotkowski, Y. P. Chen, A. Taleb-Ibrahimi, *et al.*, Ultrafast surface carrier dynamics in the topological insulator Bi₂Te₃, *Nano Lett.* **12**, 3532 (2012).
- [39] W. Lu, Z. Fan, Y. Yang, J. Ma, J. Lai, X. Song, X. Zhuo, Z. Xu, J. Liu, X. Hu, *et al.*, Ultrafast photothermoelectric

- effect in Dirac semimetallic Cd_3As_2 revealed by terahertz emission, *Nat. Commun.* **13**, 1623 (2022).
- [40] R. Freeman, A. Zholud, Z. Dun, H. Zhou, and S. Urazhdin, Evidence for Dyakonov-Perel-like spin relaxation in Pt, *Phys. Rev. Lett.* **120**, 067204 (2018).
- [41] D. A. Naylor and M. K. Tahic, Apodizing functions for Fourier transform spectroscopy, *J. Opt. Soc. Am. A* **24**, 3644 (2007).
- [42] A. L. Chekhov, Y. Behovits, J. J. F. Heitz, C. Denker, D. A. Reiss, M. Wolf, M. Weinelt, P. W. Brouwer, M. Münzenberg, and T. Kampfrath, Ultrafast demagnetization of iron induced by optical versus terahertz pulses, *Phys. Rev. X* **11**, 041055 (2021).
- [43] S. Bonetti, M. C. Hoffmann, M.-J. Sher, Z. Chen, S.-H. Yang, M. G. Samant, S. S. P. Parkin, and H. A. Dürr, THz-driven ultrafast spin-lattice scattering in amorphous metallic ferromagnets, *Phys. Rev. Lett.* **117**, 087205 (2016).
- [44] K. L. Krewer, W. Zhang, J. Arabski, G. Schmerber, E. Beaupre, M. Bonn, and D. Turchinovich, Thickness-dependent electron momentum relaxation times in iron films, *Appl. Phys. Lett.* **116**, 102406 (2020).
- [45] B. Lee, Y. Ishida, J. Kim, J. Kim, N. H. Jo, S. Y. Kim, I. Kwak, M.-C. Lee, K. Kim, J. H. Kim, *et al.*, Ultrafast probes of photovoltage and two-dimensional electron gas in the topological insulator Bi_2Te_3 by angle-resolved photoemission and terahertz spectroscopy, *Phys. Rev. B* **106**, 195430 (2022).
- [46] O. Gueckstock, R. L. Seeger, T. S. Seifert, S. Auffret, S. Gambarelli, J. N. Kirchhof, K. I. Bolotin, V. Baltz, T. Kampfrath, and L. Nádorvnik, Impact of gigahertz and terahertz transport regimes on spin propagation and conversion in the antiferromagnet IrMn , *Appl. Phys. Lett.* **120**, 062408 (2022).
- [47] H. Wu, P. Zhang, P. Deng, Q. Lan, Q. Pan, S. A. Razavi, X. Che, L. Huang, B. Dai, K. Wong, *et al.*, Room-temperature spin-orbit torque from topological surface states, *Phys. Rev. Lett.* **123**, 207205 (2019).
- [48] H. Wang, C. Du, P. Chris Hammel, and F. Yang, Spin current and inverse spin Hall effect in ferromagnetic metals probed by $\text{Y}_3\text{Fe}_5\text{O}_{12}$ -based spin pumping, *Appl. Phys. Lett.* **104**, 202405 (2014).
- [49] F. Bonell, M. Goto, G. Sauthier, J. F. Sierra, A. I. Figueroa, M. V. Costache, S. Miwa, Y. Suzuki, and S. O. Valenzuela, Control of spin-orbit torques by interface engineering in topological insulator heterostructures, *Nano Lett.* **20**, 5893 (2020).
- [50] P. D. C. King, R. C. Hatch, M. Bianchi, R. Ovsyannikov, C. Lupulescu, G. Landolt, B. Slomski, J. H. Dil, D. Guan, J. L. Mi, *et al.*, Large Rashba spin splitting of a two-dimensional electron gas in Bi_2Se_3 , *Phys. Rev. Lett.* **107**, 096802 (2011).
- [51] M. S. Bahramy, P. D. C. King, A. de la Torre, J. Chang, M. Shi, L. Patthey, G. Balakrishnan, P. Hofmann, R. Arita, N. Nagaosa, *et al.*, Emergent quantum confinement at topological insulator surfaces, *Nat. Commun.* **3**, 1159 (2012).
- [52] M. Tong, Y. Hu, Z. Wang, T. Zhou, X. Xie, X. A. Cheng, and T. Jiang, Enhanced terahertz radiation by efficient spin-to-charge conversion in Rashba-mediated Dirac surface states, *Nano Lett.* **21**, 60 (2021).
- [53] J. Sánchez-Barriga, E. Golias, A. Varykhalov, J. Braun, L. V. Yashina, R. Schumann, J. Minár, H. Ebert, O. Kornilov, and O. Rader, Ultrafast spin-polarization control of Dirac fermions in topological insulators, *Phys. Rev. B* **93**, 155426 (2016).
- [54] K. Kuroda, J. Reimann, K. A. Kokh, O. E. Tereshchenko, A. Kimura, J. Gütde, and U. Höfer, Ultrafast energy- and momentum-resolved surface Dirac photocurrents in the topological insulator Sb_2Te_3 , *Phys. Rev. B* **95**, 081103 (2017).
- [55] L. Ding, M. Wu, S. Zhou, L. Zhu, H. Wen, X. Cheng, and W. Xu, Study of the surface and bulk states of Bi_2Te_3 topological insulators using terahertz time-domain spectroscopy, *Phys. Status Solidi RRL* **17**, 2300008 (2023).
- [56] Y. L. Chen, J. G. Analytis, J.-H. Chu, Z. K. Liu, S.-K. Mo, X. L. Qi, H. J. Zhang, D. H. Lu, X. Dai, Z. Fang, *et al.*, Experimental realization of a three-dimensional topological insulator, Bi_2Te_3 , *Science* **325**, 178 (2009).
- [57] D.-X. Qu, Y. S. Hor, J. Xiong, R. J. Cava, and N. P. Ong, Quantum oscillations and Hall anomaly of surface states in the topological insulator Bi_2Te_3 , *Science* **329**, 821 (2010).
- [58] S. M. Farzaneh and S. Rakheja, Intrinsic spin Hall effect in topological insulators: A first-principles study, *Phys. Rev. Mater.* **4**, 114202 (2020).
- [59] J. C. Rojas-Sánchez, S. Oyarzún, Y. Fu, A. Marty, C. Vergnaud, S. Gambarelli, L. Vila, M. Jamet, Y. Ohtsubo, A. Taleb-Ibrahimi, *et al.*, Spin to charge conversion at room temperature by spin pumping into a new type of topological insulator: α -Sn film, *Phys. Rev. Lett.* **116**, 096602 (2016).
- [60] B. B. Singh, S. K. Jena, M. Samanta, K. Biswas, and S. Bedanta, High spin to charge conversion efficiency in electron beam-evaporated topological insulator Bi_2Se_3 , *ACS Appl. Mater. Interfaces* **12**, 53409 (2020).
- [61] M. DC, J.-Y. Chen, T. Peterson, P. Sahu, B. Ma, N. Mousavi, R. Harjani, and J.-P. Wang, Observation of high spin-to-charge conversion by sputtered bismuth selenide thin films at room temperature, *Nano Lett.* **19**, 4836 (2019).
- [62] J. B. S. Mendes, O. Alves Santos, J. Holanda, R. P. Loreto, C. I. L. de Araujo, C.-Z. Chang, J. S. Moodera, A. Azevedo, and S. M. Rezende, Dirac-surface-state-dominated spin to charge current conversion in the topological insulator $(\text{Bi}_{0.22}\text{Sb}_{0.78})_2\text{Te}_3$ films at room temperature, *Phys. Rev. B* **96**, 180415 (2017).
- [63] C. N. Wu, Y. H. Lin, Y. T. Fanchiang, H. Y. Hung, H. Y. Lin, P. H. Lin, J. G. Lin, S. F. Lee, M. Hong, and J. Kwo, Strongly enhanced spin current in topological insulator/ferromagnetic metal heterostructures by spin pumping, *J. Appl. Phys.* **117**, 17D148 (2015).
- [64] J. Li, Z. Y. Wang, A. Tan, P. A. Glans, E. Arenholz, C. Hwang, J. Shi, and Z. Q. Qiu, Magnetic dead layer at the interface between a Co film and the topological insulator Bi_2Se_3 , *Phys. Rev. B* **86**, 054430 (2012).
- [65] L. A. Walsh, C. M. Smyth, A. T. Barton, Q. Wang, Z. Che, R. Yue, J. Kim, M. J. Kim, R. M. Wallace, and C. L. Hinkle, Interface chemistry of contact metals and ferromagnets on the topological insulator Bi_2Se_3 , *J. Phys. Chem. C* **121**, 23551 (2017).
- [66] E. Longo, C. Wiemer, R. Cecchini, M. Longo, A. Lamperti, A. Khanas, A. Zenkevich, M. Fanciulli, and R. Manton, Chemical, structural and magnetic properties of the

- Fe/Sb₂Te₃ interface, *J. Magn. Magn. Mater.* **474**, 632 (2019).
- [67] E. Longo, C. Wiemer, R. Cecchini, M. Longo, A. Lamperti, A. Khanas, A. Zenkevich, M. Cantoni, C. Rinaldi, M. Fanciulli, *et al.*, Fe/Sb₂Te₃ Interface reconstruction through mild thermal annealing, *Adv. Mater. Interfaces* **7**, 2000905 (2020).
- [68] R. P. Gupta, O. D. Iyore, K. Xiong, J. B. White, K. Cho, H. N. Alshareef, and B. E. Gnade, Interface characterization of cobalt contacts on bismuth selenium telluride for thermoelectric devices, *Electrochem. Solid-State Lett.* **12**, H395 (2009).
- [69] S. Steil, N. Großmann, M. Laux, A. Ruffing, D. Steil, M. Wiesenmayer, S. Mathias, O. L. A. Monti, M. Cinchetti, and M. Aeschlimann, Spin-dependent trapping of electrons at spininterfaces, *Nat. Phys.* **9**, 242 (2013).
- [70] T. S. Seifert, L. Cheng, Z. Wei, T. Kampfrath, and J. Qi, Spintronic sources of ultrashort terahertz electromagnetic pulses, *Appl. Phys. Lett.* **120**, 180401 (2022).
- [71] A. L. Chekhov, Y. Behovits, U. Martens, B. R. Serrano, M. Wolf, T. S. Seifert, M. Münzenberg, and T. Kampfrath, Broadband spintronic detection of the absolute field strength of terahertz electromagnetic pulses, *Phys. Rev. Appl.* **20**, 034037 (2023).
- [72] G. Bierhance, C. In, E. Rongione, R. Rouzegar, O. Gueckstock, E. Longo, L. Baringthon, N. Reyren, R. Lebrun, J.-M. George, *et al.*, *Terahertz time-domain signatures of the inverse Edelstein effect in topological-insulator/ferromagnet heterostructures, dataset (version v1)*, <https://doi.org/10.5281/zenodo.17872415> (Zenodo, 2025).
- [73] S. Fragkos, L. Baringthon, P. Tsipas, E. Xenogiannopoulou, P. Le Fèvre, P. Kumar, H. Okuno, N. Reyren, A. Lemaitre, G. Patriarche, *et al.*, Topological surface states in epitaxial (SnBi₂Te₄)_n(Bi₂Te₃)_m natural van der Waals superlattices, *Phys. Rev. Mater.* **5**, 014203 (2021).
- [74] L. Baringthon, T. H. Dang, H. Jaffrès, N. Reyren, J.-M. George, M. Morassi, G. Patriarche, A. Lemaitre, F. Bertran, and P. Le Fèvre, Topological surface states in ultrathin Bi_{1-x}Sb_x layers, *Phys. Rev. Mater.* **6**, 074204 (2022).
- [75] V. Sharma, W. Wu, P. Bajracharya, D. Q. To, A. Johnson, A. Janotti, G. W. Bryant, L. Gundlach, M. B. Jungfleisch, and R. C. Budhani, Light and microwave driven spin pumping across FeGaB–BiSb interface, *Phys. Rev. Mater.* **5**, 124410 (2021).
- [76] J. Han and L. Liu, Topological insulators for efficient spin–orbit torques, *APL Mater.* **9**, 060901 (2021).
- [77] T. S. Seifert, N. M. Tran, O. Gueckstock, S. M. Rouzegar, L. Nadvornik, S. Jaiswal, G. Jakob, V. V. Temnov, M. Münzenberg, M. Wolf, *et al.*, Terahertz spectroscopy for all-optical spintronic characterization of the spin-Hall-effect metals Pt, W and Cu₈₀Ir₂₀, *J. Phys. D: Appl. Phys.* **51**, 364003 (2018).
- [78] H. Park, S. Rho, J. Kim, H. Kim, D. Kim, C. Kang, and M.-H. Cho, Topological surface-dominated spintronic THz emission in topologically nontrivial Bi_{1-x}Sb_x films, *Adv. Sci.* **9**, 2200948 (2022).
- [79] H. Wang, J. Kally, J. S. Lee, T. Liu, H. Chang, D. R. Hickey, K. A. Mkhoyan, M. Wu, A. Richardella, and N. Samarth, Surface-state-dominated spin-charge current conversion in topological-insulator–ferromagnetic-insulator heterostructures, *Phys. Rev. Lett.* **117**, 076601 (2016).
- [80] Y. Zhang, K. He, C.-Z. Chang, C.-L. Song, L.-L. Wang, X. Chen, J.-F. Jia, Z. Fang, X. Dai, W.-Y. Shan, *et al.*, Crossover of the three-dimensional topological insulator Bi₂Se₃ to the two-dimensional limit, *Nat. Phys.* **6**, 584 (2010).
- [81] L. Braun, G. Mussler, A. Hruban, M. Konczykowski, T. Schumann, M. Wolf, M. Münzenberg, L. Perfetti, and T. Kampfrath, Ultrafast photocurrents at the surface of the three-dimensional topological insulator Bi₂Se₃, *Nat. Commun.* **7**, 13259 (2016).
- [82] M. Nagai, S. Watanabe, R. Imamura, M. Ashida, K. Shimoyama, H. Li, A. N. Hattori, and H. Tanaka, Characterization of ultrathin conductive films using a simplified approach for terahertz time-domain spectroscopic ellipsometry, *J. Infrared Millim. Terahertz Waves* **45**, 949 (2024).
- [83] Y. Li and T. F. Heinz, Two-dimensional models for the optical response of thin films, *2D Mater.* **5**, 025021 (2018).