

Loss of interface spin transparency from conductivity mismatch in topological insulator based spintronic terahertz emitters

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Terahertz emissions from magnetic heterostructures comprising Fermi-level engineered 3D topological insulators, Bi_2Se_3 , are compared to assess how tuning the chemical potential absent any compositional change affects the resultant emission properties. Despite an enhancement in spin-orbit torque efficiency exhibited by bulk-insulating Bi_2Se_3 heterostructures, an ~ 5 -fold reduction in the terahertz emission amplitude is observed as compared to its bulk-conducting counterpart. Such discrepancy between reciprocal measurements of spin-charge conversion is understood to result from a loss of interface transparency to spin currents due to the low carrier density and high sheet resistance of bulk-insulating Bi_2Se_3 , resulting in a pronounced conductivity mismatch between the metallic ferromagnet and topological insulating thin films that constitute our spintronic terahertz emitters.

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I. INTRODUCTION

Information storage and processing has become one of the largest consumers of energy worldwide [1] necessitating the need for novel, ultralow-power electronics to meet ever-growing demands. Replacing CMOS-based SRAM with nonvolatile memory is a promising avenue for reducing energy consumption and allowing for more efficient computing architectures. Of particular interest are MRAM devices that operate on the principle of field-free magnetic switching through spin-orbit torques (SOT) [2–4] in which a charge-to-spin conversion takes place through the spin Hall [5] or Rashba-Edelstein [6] effects and can be experimentally quantitated by measuring the harmonic Hall response [7]. However, lithographic fabrication of Hall bar devices within these experiments is not always conducive to the high-throughput characterization needed for SOT material selection, opening the door for an alternative, contact-free means of assessing SOT performance, having the added benefit of providing useful dynamical insights [8]. Here, terahertz (THz) emission spectroscopy has emerged as such an alternative, where relative conversion efficiencies in SOT devices, known as spintronic THz emitters, can be determined through measuring an inverse, spin-to-charge conversion

process, which is fundamentally related to the forward process through Onsager reciprocal relations [9,10].

THz emission from spintronic magnetic heterostructures [11–15] follows from the introduction of an ultrafast spin current into a nonmagnetic layer brought on by photoinduced demagnetization within a neighboring metallic ferromagnet by a femtosecond light pulse. This spin current is converted to a subpicosecond charge current through an inverse spin Hall [16] or Rashba-Edelstein [17,18] effect, ultimately resulting in the emission of THz radiation. In this way, temporal dynamics intrinsic to the spin-to-charge conversion process in such heterostructures can be understood on the ultrashort timescales that underlie current generation and decay. To date, the majority of spintronic THz emitters are based on nonmagnetic, heavy metals, where spin-to-charge conversion is rooted in an inverse spin Hall effect and traditionally understood to arise from extrinsic spin-dependent Mott scattering [19–21]. Here, both high-field >300 kV/cm (cf. Ref. [22]) and broadband >20 THz (cf. Ref. [11]) emission have been observed, reflecting both the efficiency and ultrafast timescales inherent to this conversion process in heavy metals. Improving on such conversion efficiency entails strengthening the correlation between spin- and charge degrees of freedom, as exemplified by the spin-momentum locking found in three-dimensional (3D) topological insulators.

Three-dimensional topological insulators (TIs) can exhibit highly efficient spin-to-charge conversion due to strong spin-orbit coupling and an inherent spin-momentum locking present in helical Dirac surface states [23], enabling a high degree of spin polarization to accumulate at the interface between the TI and a magnetic layer of a spin torque device [24–26]. Despite these favorable qualities, THz emission efficiencies from TI-based spintronic emitters underperform with respect to more conventional, heavy-metal emitters

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comprising elemental W or Pt. Here, interfacial and bulk defects in the TI film are expected to play a contributing role by shifting the chemical potential away from the topological surface states (TSS) that span the bulk band gap, placing it within the bulk conduction or valence bands [27,28]. Together with surface charge transfer [29] and band bending [30] that often accompanies heterostructuring, these changes to the chemical potential lead to an interfacial electronic structure that is largely dominated by bulk carriers. This has sparked efforts to improve charge-to-spin conversion efficiencies in TI-based SOT devices through Fermi level (E_F) engineering by alloying films comprising *n*-type Bi_2Te_3 and *p*-type Sb_2Te_3 to ensure predominant TSS transport [31–33].

Our recent synthesis of “bulk-insulating” TI films possessing an E_F within the bulk band gap through growth on a $(\text{Bi}_{0.5}\text{In}_{0.5})_2\text{Se}_3/\text{In}_2\text{Se}_3$ buffer layer allowed us to compare the SOT efficiency to that of conventional “bulk-conducting” Bi_2Se_3 films used for switching in an adjacent $\text{Ni}_{80}\text{Fe}_{20}$ layer grown *in situ* [34]. This approach circumvents the reduction in band gap and mobility that accompanies alloying and compensatory doping, and ensures an identical interfacial structure, allowing for direct comparison where only the chemical potential differs. There, we found a significant ($4\times$) reduction in the critical current density needed for switching, along with a 5–10-fold improvement in SOT efficiency within bulk-insulating Bi_2Se_3 , begging the question as to whether a similar improvement in THz emission efficiency could be likewise observed.

In this article, THz emission from the same $\text{Ni}_{80}\text{Fe}_{20}/\text{Bi}_2\text{Se}_3$ magnetic heterostructures comprising E_F -engineered, bulk-insulating (BI) and bulk-conducting (BC) 3D TI (Bi_2Se_3) thin films as used for SOT measurements in Ref. [34] are compared in order to assess how tuning the chemical potential affects the resultant emission properties. Despite the significant enhancement in SOT efficiency exhibited by $\text{Ni}_{80}\text{Fe}_{20}/\text{BI-Bi}_2\text{Se}_3$ as compared to $\text{Ni}_{80}\text{Fe}_{20}/\text{BC-Bi}_2\text{Se}_3$, an $\sim 5\times$ reduction in the THz emission amplitude is observed. Such a discrepancy between reciprocal measurements of spin-charge conversion is understood to result from a conductivity mismatch across the metallic ferromagnet/TI interface, becoming more pronounced in $\text{BI-Bi}_2\text{Se}_3$ due to a lower carrier density and higher sheet resistance. This reduced interface transparency leads to weaker spin current injection into the TI layer following ultrafast demagnetization. Our results underscore the importance of conductivity mismatch when assessing high-frequency spin-to-charge conversion efficiencies in magnetic heterostructures consisting of quantum material systems.

II. EXPERIMENT

Bulk-insulating and bulk-conducting Bi_2Se_3 thin films [<8 Se-Bi-Se-Bi-Se quintuple layers (QL) ~ 8 nm] were grown in an ultrahigh-vacuum environment [$P \sim 2 \times 10^{-10}$ Torr (base)] by molecular-beam epitaxy and characterized by magnetotransport and angle-resolved photoemission spectroscopy (ARPES) prior to carrying out THz emission measurements [34]. While $\text{BC-Bi}_2\text{Se}_3$ films were deposited directly on sapphire (c- Al_2O_3 ; $t \sim 0.3$ mm), $\text{BI-Bi}_2\text{Se}_3$ samples

were deposited on a $(\text{Bi}_{0.5}\text{In}_{0.5})_2\text{Se}_3$ (20 QL)/ In_2Se_3 (20 QL) buffer layer, which serves as an electrically insulating, virtual substrate that is both isostructural and better lattice matched to Bi_2Se_3 (1.6%/3.3%, respectively), as compared to Al_2O_3 (14%) [35]. This produces higher quality Bi_2Se_3 films having fewer interfacial and bulk defects [35], as evidenced by the higher carrier mobility ($\mu = 1608 \text{ cm}^2/\text{Vs}$ at 300 K) and lower carrier density ($n = 1.87 \times 10^{12} \text{ cm}^{-2}$ at 300 K) of $\text{BI-Bi}_2\text{Se}_3$ as compared to $\text{BC-Bi}_2\text{Se}_3$ ($\mu = 282 \text{ cm}^2/\text{Vs}$; $n = 3.72 \times 10^{12} \text{ cm}^{-2}$ at 300 K). This reduced carrier density is reflected in the ARPES spectra, where the E_F of $\text{BI-Bi}_2\text{Se}_3$ lies within the bulk band gap, while that of $\text{BC-Bi}_2\text{Se}_3$ crosses above the conduction band, revealing a dominant contribution of topological surface states in the surface electronic structure of $\text{BI-Bi}_2\text{Se}_3$ [34].

Magnetic heterostructures of $\text{Ni}_{80}\text{Fe}_{20}/\text{BI-Bi}_2\text{Se}_3$ and $\text{Ni}_{80}\text{Fe}_{20}/\text{BC-Bi}_2\text{Se}_3$ were fabricated by *in situ* deposition of Permalloy ($\text{Py} = \text{Ni}_{80}\text{Fe}_{20}$ - 8 nm) on Bi_2Se_3 thin films using an interconnected sputter system. In this way, clean, well-controlled, and reproducible interfaces between Bi_2Se_3 and $\text{Ni}_{80}\text{Fe}_{20}$ thin films can be created, which can facilitate a strong hybridization between the topological insulator and ferromagnetic metal, as well as the formation of descendant states whose spin-momentum locking underlies the efficient spin-to-charge conversion processes responsible for THz emission [34,36].

THz emission experiments were performed in a transmission geometry using a Yb-fiber oscillator, outputting 140-fs optical pulses with a central wavelength of 1040 nm (1.19 eV) at an 80-MHz repetition rate. Spintronic THz emitters were excited film side first at normal incidence using loosely focused (full width at half maximum = 195 μm) optical pulses at a fluence, $F < 250 \text{ mJ}/\text{cm}^2$. The emitted THz was collected as a function of excitation polarization (θ), sample azimuth (α), and magnetic-field (0.2 T) orientation (ϕ) using free space electro-optic sampling in a 1-mm-thick $\langle 110 \rangle$ GaP crystal. The driving optical beam underwent a 2-kHz modulation via mechanical chopping, while the in-plane $[1, -1, 0]$ axis of GaP was preferentially oriented to detect vertically polarized THz pulses. All experiments were performed at room temperature (295 K) in a dry air atmosphere ($\text{RH} < 20\%$) to reduce the effect of water vapor absorption on the emitted THz signal.

III. RESULTS AND DISCUSSION

THz emission spectra from bare $\text{Ni}_{80}\text{Fe}_{20}$ and $\text{BI-Bi}_2\text{Se}_3$ thin films are shown in Fig. 1. A 10-nm Se cap was deposited on Bi_2Se_3 to protect the surface from atmospheric oxidation. Here, we exploit the sensitivity of THz emission to magnetic point-group and dipole symmetries to characterize the emission dependence of these constituent thin films prior to heterostructuring into spintronic emitters.

Figure 1(a) schematically illustrates the THz emission geometry for $\text{Ni}_{80}\text{Fe}_{20}$, where an added in-plane, external magnetic (H -) field is applied to align the magnetic domains. Given the low in-plane coercive field for $\text{Ni}_{80}\text{Fe}_{20}$ ($H_c < 6 \times 10^{-4}$ T), our 0.2-T H field ensures the magnetization to fully saturate along the applied field direction. The emitted THz trace, shown in Fig. 1(b), is polarized mutually perpendicular to the propagation and magnetization directions ($\mathbf{k} \times \mathbf{M}$), and

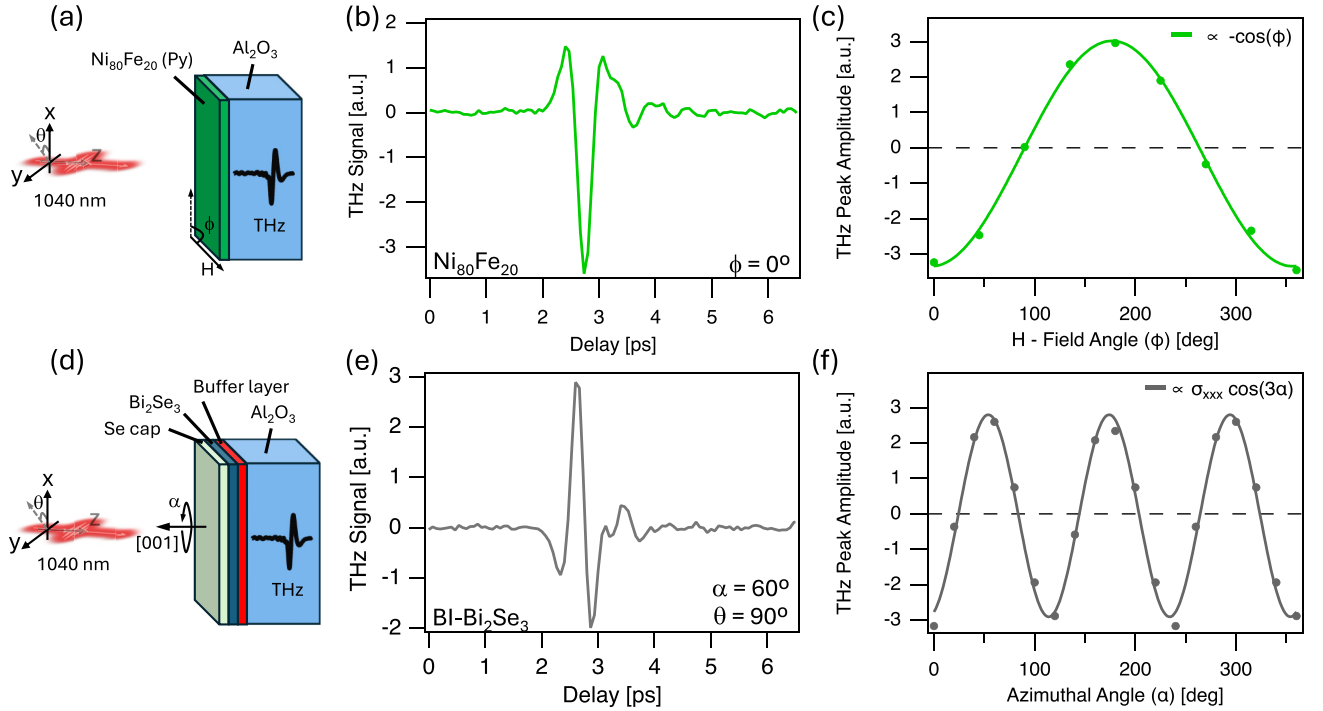


FIG. 1. THz emission from $\text{Ni}_{80}\text{Fe}_{20}$ and Bi_2Se_3 thin films. Schematic illustrating THz emission from (a) Permalloy, $\text{Ni}_{80}\text{Fe}_{20}$ (5.0 nm) and (d) $\text{BI-Bi}_2\text{Se}_3$ [8 QL on a $(\text{Bi}_{0.5}\text{In}_{0.5})_2\text{Se}_3$ (20 QL)/ In_2Se_3 (20 QL) buffer layer], thin films deposited on sapphire (c- Al_2O_3 ; $r0.3$ mm) substrates. Representative THz traces from (b) $\text{Ni}_{80}\text{Fe}_{20}$ and (e) $\text{BI-Bi}_2\text{Se}_3$ driven by an ultrashort, horizontally polarized, near-infrared light pulse [$\Delta t < 140$ fs; $\lambda = 1040$ nm ($h\nu = 1.19$ eV)] normally incident on the topmost surface of the thin film. (c) THz peak amplitude (green points) from $\text{Ni}_{80}\text{Fe}_{20}$ as a function of an applied, in-plane magnetic field ($H < 0.2$ T) angle (ϕ) and fit to the $\cos(\phi)$ dependence for ultrafast demagnetization (solid green). (f) THz peak amplitude (gray points) from $\text{BI-Bi}_2\text{Se}_3$ as a function of azimuthal sample rotation (α) about a [001] normal (symmetrized with respect to 180°) and fit to the $\cos(3\alpha)$ dependence of the shift current (solid gray).

undergoes a polarity reversal with field ($\phi = 0^\circ$ vs $\phi = 180^\circ$) as shown in Fig. 1(c), with ϕ denoting the direction of the H field relative to the y axis. Both the sinusoidal H -field dependence in Fig. 1(c) as well as the weak ($<17\%$) dependence of THz emission on pump polarization suggest the underlying mechanism to be rooted in ultrafast demagnetization, whereby a photoinduced quench of the magnetic moment introduces a time variation to the magnetic dipole, yielding $E_{\text{THz}} \propto \partial^2 M / \partial t^2$ [37].

Broadband THz emission from $\text{BI-Bi}_2\text{Se}_3$ is shown in Fig. 1(e), and is similar in amplitude to the emission spectrum of $\text{Ni}_{80}\text{Fe}_{20}$ [Fig. 1(b)]. Despite possessing a centrosymmetric, trigonal space group ($R\bar{3}m$; No. 166), a nonlinear optical response in Bi_2Se_3 thin films (<8 QL) can originate from contributions arising from the surface/interface, where inversion symmetry is naturally lost. As a prototypical 3D topological insulator, Bi_2Se_3 has been extensively studied using THz techniques [38–41] where helicity-dependent injection [42] and photon drag effects [43] have been observed alongside helicity-independent shift photocurrents [44]. This latter mechanism dominates following normal-incident excitation by linearly polarized light [Fig. 1(d)] and can be attributed to a shift in electron charge density along the Bi–Se bond following photoexcitation.

Optical excitation along the [001] surface normal using linearly polarized light yields a $\sigma_{xxx} \cos(3\alpha)$ dependence to the THz amplitude [Fig. 1(f)], which is fully consistent with shift current excitation within the $3m$ point-group symmetry

of a Bi_2Se_3 surface (Appendixes 1 and 2). Here, α denotes the azimuthal rotation angle, while σ_{xxx} defines the sole nonlinear tensor element of a (001)-oriented Bi_2Se_3 surface under normal incident excitation [45]. This mechanism for photocurrent generation is further supported by a pump-polarization dependence, which results in a $\sigma_{xxx} \cos(2\theta)$ dependence on the THz amplitude as a function of polarization from a normally incident optical driving pulse relative to the vertical, x axis (θ). Such behavior (see Fig. 4 in Appendix 1) is similarly consistent with shift current generation, and when taken together with the azimuthal dependence in Fig. 1(f) suggests this to be the dominant mechanism underlying THz emission from our bare $\text{BI-Bi}_2\text{Se}_3$ thin film.

Despite a larger (~ 26 nm [46]) optical penetration depth in Bi_2Se_3 at $\lambda = 1040$ nm (1.19 eV) as compared to our 8-QL film thickness, THz emission from $\text{BI-Bi}_2\text{Se}_3$ is spectroscopically indistinguishable from the emission spectrum of $\text{BC-Bi}_2\text{Se}_3$ from literature [43,44], where no buffer layer is present, as both materials host the same $3m$ point-group symmetry. This is due to the fact that shift current generation follows from an optical excitation that predominately involves transitions between topologically trivial states within the full 1.19-eV window of the optical driving pulse [47]. In order to exploit the improved film quality brought on by better lattice matching through inclusion of this buffer layer, our THz emission experiments must necessarily become sensitive to the behavior of carriers in the vicinity of E_F , where the effects of topological surface states can dominate the electronic

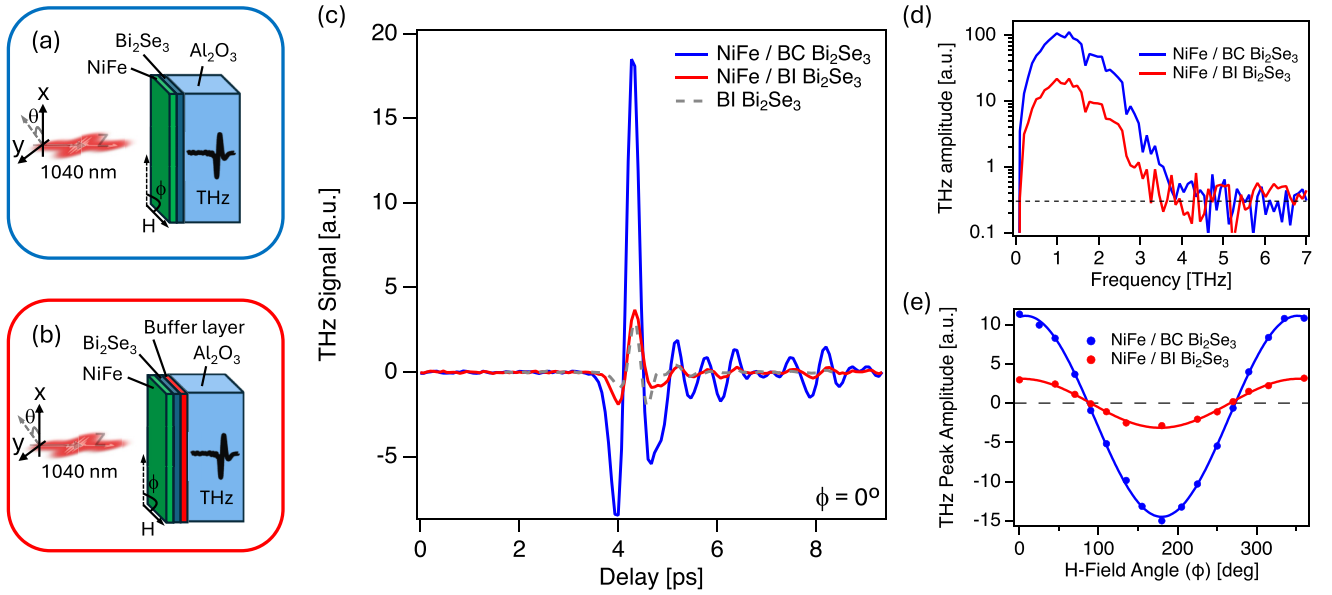


FIG. 2. THz emission from $\text{Ni}_{80}\text{Fe}_{20}/\text{Bi}_2\text{Se}_3$ heterostructures. Sketch of the spintronic THz emitters (a) $\text{Ni}_{80}\text{Fe}_{20}$ (8.0 nm)/BC- Bi_2Se_3 (8 QL) and (b) $\text{Ni}_{80}\text{Fe}_{20}$ (8.0 nm)/BI- Bi_2Se_3 (8 QL). (c) THz waveforms generated from $\text{Ni}_{80}\text{Fe}_{20}/\text{BC-Bi}_2\text{Se}_3$ (blue trace), $\text{Ni}_{80}\text{Fe}_{20}/\text{BI-Bi}_2\text{Se}_3$ (red trace), and bare BI- Bi_2Se_3 (gray trace) under a saturating magnetic field (0.2 T) applied perpendicular to THz polarization and propagation directions ($\phi = 0^\circ$). (d) The corresponding power spectra of the time-domain traces in (c), where noise floor is denoted by the dotted gray line. (e) THz peak amplitude from $\text{Ni}_{80}\text{Fe}_{20}/\text{BC-Bi}_2\text{Se}_3$ (blue circles) and $\text{Ni}_{80}\text{Fe}_{20}/\text{BI-Bi}_2\text{Se}_3$ heterostructures (red circles) as a function of an applied, in-plane magnetic-field angle (ϕ). Solid lines show a $\sin(\phi)$ dependence on the experimental data.

transport [48]. This can be accomplished by injecting spin carriers directly into Bi_2Se_3 thin films following photoexcitation within a magnetic heterostructure, where spin-momentum locking of 3D TIs leads to an efficient spin-to-charge conversion, resulting in the emission of THz radiation from transiently excited photocurrents in direct proximity to E_F [49–51].

THz emission spectra from the magnetic heterostructures, $\text{Ni}_{80}\text{Fe}_{20}/\text{BC-Bi}_2\text{Se}_3$ and $\text{Ni}_{80}\text{Fe}_{20}/\text{BI-Bi}_2\text{Se}_3$, are shown in Fig. 2. Here, a normally incident, ultrashort light pulse transiently quenches the magnetic moment of the metallic ferromagnetic alloy, $\text{Ni}_{80}\text{Fe}_{20}$, leading to an excess of spin angular momentum that is partially carried away by photoexcited hot electrons [52] with the remainder being lost to the lattice [53]. In the conventional framework of the Stoner model, the resulting spin current brought on by this demagnetization process is driven across the interface by a generalized spin voltage [54], where it can subsequently undergo a spin-to-charge conversion at the surface of Bi_2Se_3 by way of an inverse Rashba-Edelstein effect (IREE) [50].

THz emission traces shown in Fig. 2(c) reveal a weak ($1.15\times$) enhancement of the emitted THz amplitude in $\text{Ni}_{80}\text{Fe}_{20}/\text{BI-Bi}_2\text{Se}_3$ as compared to bare BI- Bi_2Se_3 . In contrast, $\text{Ni}_{80}\text{Fe}_{20}/\text{BC-Bi}_2\text{Se}_3$ shows a more significant enhancement ($4.8\times$) as compared to $\text{Ni}_{80}\text{Fe}_{20}/\text{BI-Bi}_2\text{Se}_3$, with both spectra exhibiting broadband emission [Fig. 2(d)] that is limited by the time resolution of our 140-fs driving pulse as well as the detection bandwidth of the 1-mm $\langle 110 \rangle$ GaP crystal used for electro-optic sampling. Similar to the emission dependence of $\text{Ni}_{80}\text{Fe}_{20}$ in Figs. 1(a)–1(c), the emitted THz traces in Fig. 2(c) are polarized mutually perpendicular to the propagation and magnetization directions

($\mathbf{k} \times \mathbf{M}$), with both traces undergoing a polarity reversal with an externally applied H field ($\phi = 0^\circ$ vs $\phi = 180^\circ$) as shown in Fig. 2(e). Here, a higher emission amplitude for $\text{Ni}_{80}\text{Fe}_{20}/\text{BC-Bi}_2\text{Se}_3$ is observed over the entire sinusoidal cycle, where the THz field dependence of both TI spintronic emitters exhibits a 180° phase shift with respect to $\text{Ni}_{80}\text{Fe}_{20}$ [Fig. 1(c)]. Hence, despite a modest enhancement of THz amplitude in magnetic heterostructures, both the change in polarity with respect to $\text{Ni}_{80}\text{Fe}_{20}$ as well as a lack of pump-polarization dependence in the emitted THz trace of $\text{Ni}_{80}\text{Fe}_{20}/\text{BC-Bi}_2\text{Se}_3$ and $\text{Ni}_{80}\text{Fe}_{20}/\text{BI-Bi}_2\text{Se}_3$ (see Fig. 5, Appendix 3) highlight the distinct microscopic mechanism underlying spin-to-charge conversion in spintronic emitters as compared to the respective magnetic dipole radiation and shift current generation exhibited by their constituent $\text{Ni}_{80}\text{Fe}_{20}$ and Bi_2Se_3 elements. In other words, THz spin transport is still strongly linked to demagnetization, where charge- and spin pumping have been proposed to directly follow from this process [55], but the resultant emission pattern, polarity, and bandwidth (see Fig. 6, Appendix 4) contrast with that arising from either dipole radiation or photocurrent excitation.

In comparison to Fig. 2, angle-dependent second-harmonic Hall measurements reveal markedly different behavior for the SOT efficiencies of $\text{Ni}_{80}\text{Fe}_{20}/\text{BI-Bi}_2\text{Se}_3$ and $\text{Ni}_{80}\text{Fe}_{20}/\text{BC-Bi}_2\text{Se}_3$ [34]. Namely, $\text{Ni}_{80}\text{Fe}_{20}/\text{BI-Bi}_2\text{Se}_3$ showed an ~ 4 -fold reduction in the critical current density needed to switch an adjacent $\text{Ni}_{80}\text{Fe}_{20}$ layer, and a 5–10-fold enhancement in SOT efficiency as compared to its $\text{Ni}_{80}\text{Fe}_{20}/\text{BC-Bi}_2\text{Se}_3$ counterpart. Based on Onsager reciprocal relations, the corresponding charge-to-spin conversion process underlying SOT likewise gives rise to spin-to-charge conversion as captured by THz emission. However, the charge current ratio,

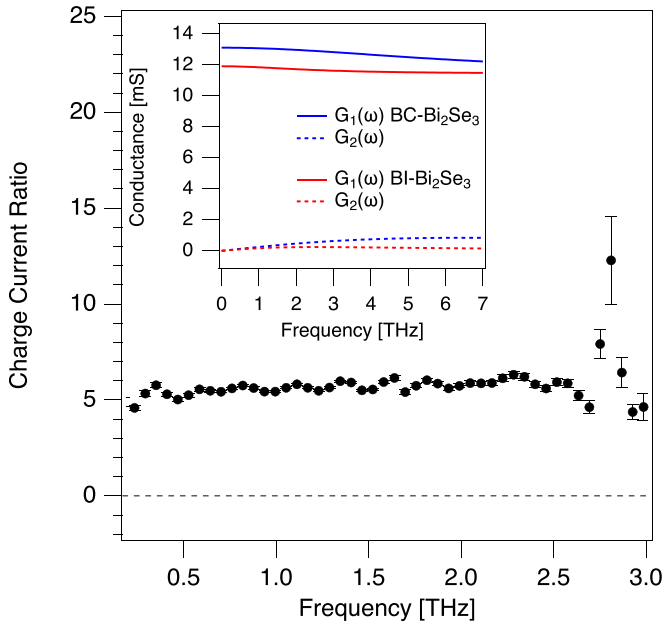


FIG. 3. THz charge current ratio. THz frequency-dependent charge current ratio between $\text{Ni}_{80}\text{Fe}_{20}/\text{BC-Bi}_2\text{Se}_3$ and $\text{Ni}_{80}\text{Fe}_{20}/\text{BI-Bi}_2\text{Se}_3$. Error bars are computed by considering the standard error ($\sigma/\sqrt{N}$; $\sigma = \pm 15\%$ at 1 THz) following computation from the frequency-dependent noise distribution of the THz power spectrum for $n = 380$ separate THz traces collected on each sample. The inset depicts the complex, frequency-dependent conductance [$G_{1,2}(\omega)$] of BC - Bi_2Se_3 (blue) and BI - Bi_2Se_3 (red), where solid (dashed) lines denote the real (imaginary) components.

$J_{\text{BC}}(\omega)/J_{\text{BI}}(\omega) = G_{\text{BC}}(\omega)E_{\text{BC}}(\omega)/G_{\text{BI}}(\omega)E_{\text{BI}}(\omega)$, in Fig. 3 reveals an $\sim 5\times$ weaker response across the emission spectrum in $\text{Ni}_{80}\text{Fe}_{20}/\text{BI-Bi}_2\text{Se}_3$ as compared to $\text{Ni}_{80}\text{Fe}_{20}/\text{BC-Bi}_2\text{Se}_3$, where the frequency-dependent conductance, $G(\omega)$, between these two compounds differs by $<10\%$ (Appendix 5), suggesting BI- Bi_2Se_3 to either possess a smaller IREE length compared to its BC- Bi_2Se_3 counterpart, or that interface transparency between $\text{Ni}_{80}\text{Fe}_{20}$ and Bi_2Se_3 plays a significant role in determining how much spin current is injected into the TI film following photoexcitation.

While the former scenario leads to an inconsistency with second-harmonic Hall measurements when assuming that an identical spin current deriving from ultrafast demagnetization in $\text{Ni}_{80}\text{Fe}_{20}$ is injected into BC/BI- Bi_2Se_3 , the latter can arise from an interfacial resistance attributed to either roughness or a conductivity mismatch between hot electron spin carriers experiencing distinct electronic environments across the interface. Here, interfacial roughness and chemical disorder are known to erode the transparency of spin currents, resulting in less efficient spin injection, and therefore weaker, polarization-independent THz emission [56]. However, a unique advantage of using the buffer layer method to tune E_F is that nearly identical interfaces can be maintained between $\text{Ni}_{80}\text{Fe}_{20}$ and BI-/BC- Bi_2Se_3 as no compositional changes to the TI are required to shift the position of E_F relative to the bulk bands (Appendix 6). This contrasts with the more conventional approach of tuning E_F through Sb doping in $(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_3$, where interface transparency in

$(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_3/\text{Fe}$ magnetic heterostructures is known to strongly affect THz emission efficiency [57]. In our study, we were able to maintain the same chemical and compositional makeup across the interface, enabling us to rule out these differences between BI- Bi_2Se_3 and BC- Bi_2Se_3 interfaces, and allowing us to focus on how electronic properties alone influence the THz emission spectrum.

Namely, the lower carrier density in BI- Bi_2Se_3 results in a sheet resistance that is $\sim 4\times$ larger than that of BC- Bi_2Se_3 , leading to a substantial ($>100\times$) conductivity mismatch between the metallic, ferromagnet, $\text{Ni}_{80}\text{Fe}_{20}$, and E_F -engineered TIs. Within a superdiffusive model of THz spin transport [58] the spin accumulation across the interface depends on its electrical conductivity, as spin carriers are unable to inject from a highly conductive ferromagnet into a highly resistive semiconductor [59]. In this scenario, the spin-to-charge conversion efficiency is roughly unchanged between BI- Bi_2Se_3 and BC- Bi_2Se_3 , but the amount of spin current crossing the interface and resulting in an ultrafast charge current will be markedly different.

Absent resistance, the spin polarization (SP) at an interface between two semi-infinite ferromagnetic (F) and nonmagnetic (N) films is given by $\text{SP} = \frac{\beta}{1+r_N/r_F}$, where $r_N = \rho_N^* l_{sf}^N$ and $r_F = \rho_F^* l_{sf}^F$ are the products of the resistivity and spin-diffusion length for the nonmagnetic and ferromagnetic materials, respectively, while β denotes the intrinsic spin polarization of the ferromagnet [59]. Here, r_N for both BI- Bi_2Se_3 and BC- Bi_2Se_3 is computed from measured carrier densities/mobilities by applying a dc Drude model ($\rho^* = 1/\sigma = 1/n e \mu$) along with a spin-diffusion length of 1.6 nm [60]–6.2 nm [61] for Bi_2Se_3 , yielding respective values of $r_N = 1.0 \times 10^{-13}$ – $2.7 \times 10^{-14} \Omega \text{ m}^2$ and 2.9×10^{-14} – $7.6 \times 10^{-15} \Omega \text{ m}^2$ for BI- Bi_2Se_3 and BC- Bi_2Se_3 . Similarly, an $r_F = 2.0 \times 10^{-15} \text{ W m}^2$ was found for $\text{Ni}_{80}\text{Fe}_{20}$, where an experimentally measured resistivity of $2.9 \times 10^{-7} \Omega \text{ m}$ ($T = 300 \text{ K}$), and spin-diffusion length of 7 nm [62] was used to compute this quantity. Based on these values, interfacial spin polarizations of $\sim \beta/14$ – $\beta/51$ and $\sim \beta/4.8$ – $\beta/15.5$ are estimated for $\text{Ni}_{80}\text{Fe}_{20}/\text{BI-Bi}_2\text{Se}_3$ and $\text{Ni}_{80}\text{Fe}_{20}/\text{BC-Bi}_2\text{Se}_3$, respectively, meaning an ~ 2.9 – 3.3 -fold reduction to the spin polarization occurs at the BI- Bi_2Se_3 interface as compared to its BC- Bi_2Se_3 counterpart. Such a reduction in interfacial spin polarization roughly agrees with the charge current ratio presented in Fig. 3, supporting a loss of interface transparency from conductivity mismatch in the $\text{Ni}_{80}\text{Fe}_{20}/\text{BI-Bi}_2\text{Se}_3$ spintronic THz emitter.

On the increasingly short timescale of the pulse duration, alternate spin transport mechanisms that fail to rely upon such conductivity mismatch can likewise exist. This includes optical intersite spin transfer (OIST), in which a spin-selective charge redistribution occurs between electronic subsystems across the interface [63,64]. As a coherent process governed solely by optical excitation, spin transport by OIST occurs over much faster timescales as compared to superdiffusion, but similarly exhibits a short, subunit-cell relaxation length, making it especially fragile to decoherence. Given the spin-relaxation length of Bi_2Se_3 to span multiple QLs, ranging from 1.6 nm [60] to 6.2 nm [61], we can exclude OIST as an alternate spin transport mechanism. Furthermore, a notable absence of linewidth broadening was

observed in the frequency-dependent ferromagnetic resonance of both $\text{Ni}_{80}\text{Fe}_{20}/\text{BC-Bi}_2\text{Se}_3$ and $\text{Ni}_{80}\text{Fe}_{20}/\text{BI-Bi}_2\text{Se}_3$ as compared to bare $\text{Ni}_{80}\text{Fe}_{20}$ (Appendix 7). Such a null finding is indicative of there being few spins that cross the interface, consistent with a conductivity mismatch between hot electron spin carriers being the most likely explanation for the difference in THz emission strength between $\text{Ni}_{80}\text{Fe}_{20}/\text{BI-Bi}_2\text{Se}_3$ and $\text{Ni}_{80}\text{Fe}_{20}/\text{BC-Bi}_2\text{Se}_3$, as observed in our experiments.

Several strategies to overcome the limitations imposed by conductivity mismatch on THz spin transport in TI-based magnetic heterostructures are known to exist. These include replacing metallic $\text{Ni}_{80}\text{Fe}_{20}$ with a magnetic insulator, where magnon mediated spin transport can proceed via a spin Seebeck effect [65]. Alternatively, interfacing Bi_2Se_3 with a heavy metal, like Bi, has been shown to enhance THz emission, which is argued to result from the additive contribution of topological surface states and the Rashba split bands of a metallic, 2D electron gas [57,66,67]. However, enhancing the emitted THz response while retaining the same constituent elements as employed in our current study necessitates that a higher degree of spin-polarized carriers must pass through the $\text{Ni}_{80}\text{Fe}_{20}/\text{Bi}_2\text{Se}_3$ interface. This can be accomplished by incorporating a tunnel junction between the $\text{Ni}_{80}\text{Fe}_{20}$ and Bi_2Se_3 layers, to provide the interface resistance needed to regain full advantage of the enhanced spin-to-charge conversion efficiency of TIs [68]. Finally, parallel/antiparallel spin-momentum locking in the Kramers-Weyl semimetals provides another avenue to reduce the interface resistance between metallic ferromagnets such as $\text{Ni}_{80}\text{Fe}_{20}$, and spin-momentum locked surface states [69]. However, this comes with the added technical challenge of coupling the emitted THz arising from longitudinal charge currents into free space [70].

In closing, terahertz emission from magnetic heterostructures comprising E_F -engineered Bi_3Se_3 , are compared to assess how the position of the chemical potential affects the resultant emission properties. The emission spectra of bare TI films reflect the $3m$ point-group symmetry of Bi_2Se_3 , while that from magnetic heterostructures follows from the magnetic dipole symmetry of ultrafast demagnetization. Bulk-insulating Bi_2Se_3 heterostructures show an ~ 5 -fold reduction in the terahertz emission amplitude as compared to their bulk-conducting counterparts. This is attributed to a conductivity mismatch across the $\text{Ni}_{80}\text{Fe}_{20}/\text{Bi}_2\text{Se}_3$ interface brought on by the lower carrier density and higher sheet resistance of the bulk-insulating film. Tunnel junctions and Kramers-Weyl semimetals are proposed as an effective strategy to enhance the degree of spin polarization injected across the interface, providing a promising future direction for designing spintronic THz emitters using advanced, functional materials beyond elemental heavy metals.

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J.K.G. and N.S.S. contributed equally to this work.

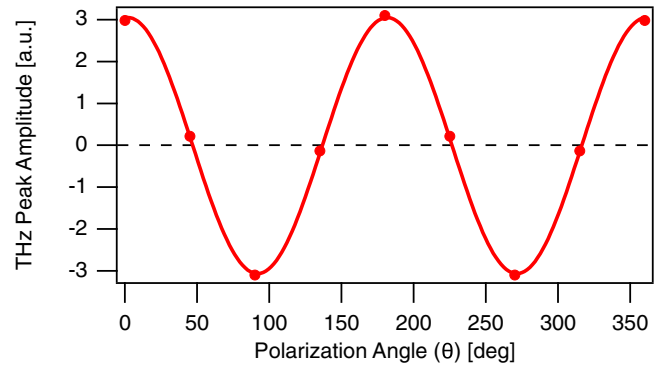


FIG. 4. THz peak amplitude (red points) from (001)-oriented BI- Bi_2Se_3 as a function of polarization angle (θ) and fit to the $\cos(2\theta)$ dependence of the shift current (solid red trace). Here, data were symmetrized about $\theta = 180^\circ$ for clarity.

DATA AVAILABILITY

The data that support the findings of this article are not publicly available because of legal restrictions preventing unrestricted public distribution. The data are available from the authors upon reasonable request.

APPENDIX

1. Bi_2Se_3 polarization dependence

Photocurrent generation arises from a second-order process in which a dc current, J_i^{dc} , is created in response to the frequency-dependent electric $[E(\omega)]$ field of a driving light pulse:

$$J_i^{\text{dc}} = \sigma_{ijk} E_j(\omega) E_k^*(\omega).$$

Here, $E_k^* = E_k(-\omega)$, while σ_{ijk} denotes the photocurrent response tensor. Given this photocurrent to be real, it must remain unchanged under complex conjugation, making the real part of σ_{ijk} symmetric following coordinate exchange. This yields a response tensor for the linear photogalvanic effect that results from a transient shift current following above-gap excitation by linearly polarized light [71]. Based on the $3m$ point-group symmetry of Bi_2Se_3 , there are five independent tensor elements of σ_{ijk} [45]: $\sigma_{xzx} = \sigma_{yzy}$, $\sigma_{xxz} = \sigma_{yyz}$, $\sigma_{zxx} = \sigma_{zyy}$, σ_{zzz} , and $\sigma_{xxx} = -\sigma_{xyy} = -\sigma_{yyx} = -\sigma_{yxy}$, where we have adopted the laboratory coordinate system defined in Fig. 1(d). At normal incidence, $E_z = 0$, the polarization-dependent photocurrent response of (001) oriented Bi_2Se_3 is given by

$$\begin{aligned} J_x &= \sigma_{xzx} E_z E_x^* + \sigma_{xxz} E_x E_z^* + \sigma_{xxx} E_x E_x^* + \sigma_{xyy} E_y E_y^* \\ &= \sigma_{xxx} (E_x E_x^* - E_y E_y^*) = \sigma_{xxx} |E|^2 (\cos^2 \theta - \sin^2 \theta) \\ &= \sigma_{xxx} |E|^2 \cos(2\theta), \end{aligned}$$

where θ denotes the projection of the polarization relative to the vertical, x axis as defined by a $\theta/2$ half-waveplate angle. Such polarization dependence is illustrated in Fig. 4, where a 2θ dependence in the THz peak amplitude is clearly evident.

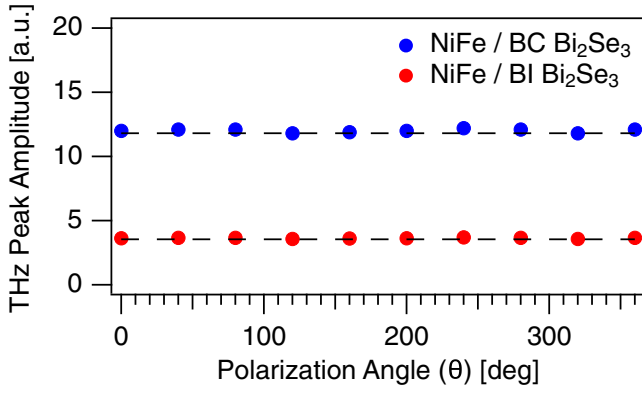


FIG. 5. THz peak amplitude from $\text{Ni}_{80}\text{Fe}_{20}/\text{BC-Bi}_2\text{Se}_3$ (blue points) and $\text{Ni}_{80}\text{Fe}_{20}/\text{BI-Bi}_2\text{Se}_3$ (red points) as a function of pump polarization (θ). Dashed lines denote the constant, average.

2. Bi_2Se_3 azimuthal dependence

Under azimuthal rotation (α) about the $[001]$ normal, the photocurrent response tensor will transform according to

$$\sigma'_{i'j'k'} = R_{i'i} R_{j'j} R_{k'k} \sigma_{ijk},$$

where $R_{i'i}$ denotes the i' th element of the z -axis rotation matrix: $R_{x'x} = R_{y'y} = \cos(\alpha)$, $R_{x'y} = -R_{y'x} = \sin(\alpha)$, $R_{zz} = 1$. Following normal incident excitation, the sole nonlinear tensor element, σ_{xxx} , will then transform as

$$\begin{aligned} \sigma_{x'x'x'} &= R_{x'x} R_{x'x} R_{x'x} \sigma_{xxx} + R_{x'x} R_{x'y} R_{x'y} \sigma_{xyy} \\ &+ R_{x'y} R_{x'y} R_{x'x} \sigma_{yyx} + R_{x'y} R_{x'x} R_{x'y} \sigma_{yxy}. \end{aligned}$$

Making use of the fact that $\sigma_{xxx} = -\sigma_{xyy} = -\sigma_{yyx} = -\sigma_{yxy}$, the above expression simplifies to

$$\sigma_{x'x'x'} = \sigma_{xxx} (\cos^3 \alpha - 3 \cos \alpha \sin^2 \alpha) = \sigma_{xxx} \cos(3\alpha),$$

revealing the 3α dependence expected for shift current generation as shown in Fig. 1(f).

3. Polarization dependence of spintronic THz emitters

In contrast to bare Bi_2Se_3 thin films, magnetic heterostructures of $\text{Ni}_{80}\text{Fe}_{20}/\text{BI-Bi}_2\text{Se}_3$ and $\text{Ni}_{80}\text{Fe}_{20}/\text{BC-Bi}_2\text{Se}_3$ lose the polarization dependence of the $3m$ space group, as illustrated in Fig. 5. Here, THz peak amplitudes are constant with respect to pump polarization (θ), akin to that observed from the emission spectrum of $\text{Ni}_{80}\text{Fe}_{20}$ following ultrafast demagnetization. Thus, despite sharing similar amplitudes, THz emission from $\text{Ni}_{80}\text{Fe}_{20}/\text{BI-Bi}_2\text{Se}_3$ exhibits no trace of shift current excitation from bare BI- Bi_2Se_3 , supporting the assignment of a spin-to-charge conversion via an inverse Rashba-Edelstein effect in the magnetic heterostructure.

4. Comparison of emission bandwidth

The normalized emission bandwidth from bare $\text{Ni}_{80}\text{Fe}_{20}$ and $\text{Ni}_{80}\text{Fe}_{20}/\text{BI-Bi}_2\text{Se}_3$ magnetic heterostructures in Fig. 6 are nearly identical, reflecting the fact that THz spin transport is strongly linked to ultrafast demagnetization. However, the normalized emission bandwidth of bare BI- Bi_2Se_3 is distinct, reflective of the microscopic origin for THz generation resulting from photocurrent excitation. In all cases our THz

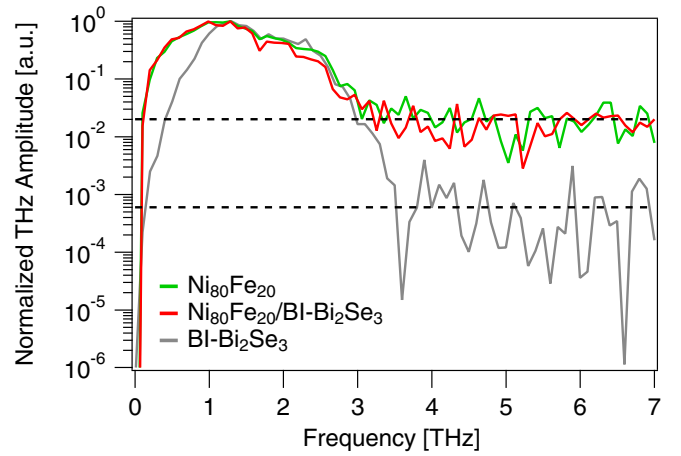


FIG. 6. Normalized emission bandwidth resulting from bare $\text{Ni}_{80}\text{Fe}_{20}$ and $\text{Ni}_{80}\text{Fe}_{20}/\text{BI-Bi}_2\text{Se}_3$ magnetic heterostructures, and bare BI- Bi_2Se_3 .

detection bandwidth is limited to <3.5 THz as determined by the time resolution of our 140-fs driving pulse, as well as the detection bandwidth of the 1 mm (110) GaP crystal used for electro-optic sampling.

5. Charge current ratio

Determining the charge current ratio from the emitted THz response begins with the consideration of Ohm's law in frequency space $\langle \mathbf{J}_C(\omega) \rangle = G(\omega) \mathbf{E}(\omega)$. $\langle \mathbf{J}_C(\omega) \rangle$ is the thickness-averaged charge current density, $G(\omega)$ is the thickness-averaged THz conductivity (conductance), and $\mathbf{E}(\omega)$ is the emitted THz electric field. For a thin-film heterostructure between two half-spaces (air and substrate), the conductance is given by $G(\omega) = \frac{n_{\text{air}} + n_{\text{sub}}}{Z_0} + \int_0^d dz \sigma(z, \omega)$ [12]. Here, $n_{\text{air}} = 1$ is the refractive index of air, $n_{\text{sub}} = 3.31$ is the refractive index of the sapphire substrate, $Z_0 = 377 \Omega$ is the impedance of free space, and $\sigma(z, \omega)$ is the frequency-dependent conductivity, which we integrate over film thickness, d . To evaluate this integral, we estimate $\sigma(z, \omega)$ to be roughly constant over d and apply a Drude model $\sigma(\omega) = \frac{ne\mu}{1 - i\omega\tau}$. For both BC- Bi_2Se_3 and BI- Bi_2Se_3 , $\sigma(\omega)$ is computed from the measured carrier densities/mobilities, yielding the complex frequency-dependent conductance curves as shown in the inset of Fig. 3. The small ($<10\%$) difference in the bulk electronic properties between these two films is taken into account by multiplying the complex conductance of BC- Bi_2Se_3 and BI- Bi_2Se_3 with their respective THz emission spectra and taking the ratio, $J_{\text{BC}}(\omega)/J_{\text{BI}}(\omega)$, to isolate the behavior of the charge, and hence injected spin, current response.

6. Surface morphology of Bi_2Se_3 and $\text{Ni}_{80}\text{Fe}_{20}/\text{Bi}_2\text{Se}_3$ heterostructures

Figure 7(a) presents a representative atomic force microscopy (AFM) image ($2 \mu\text{m} \times 2 \mu\text{m}$) of an as-grown BI- Bi_2Se_3 surface, consisting of triangular QL height islands. The $\text{Ni}_{80}\text{Fe}_{20}$ films and MgO capping layers were deposited *in situ* at room temperature, without exposure to ambient

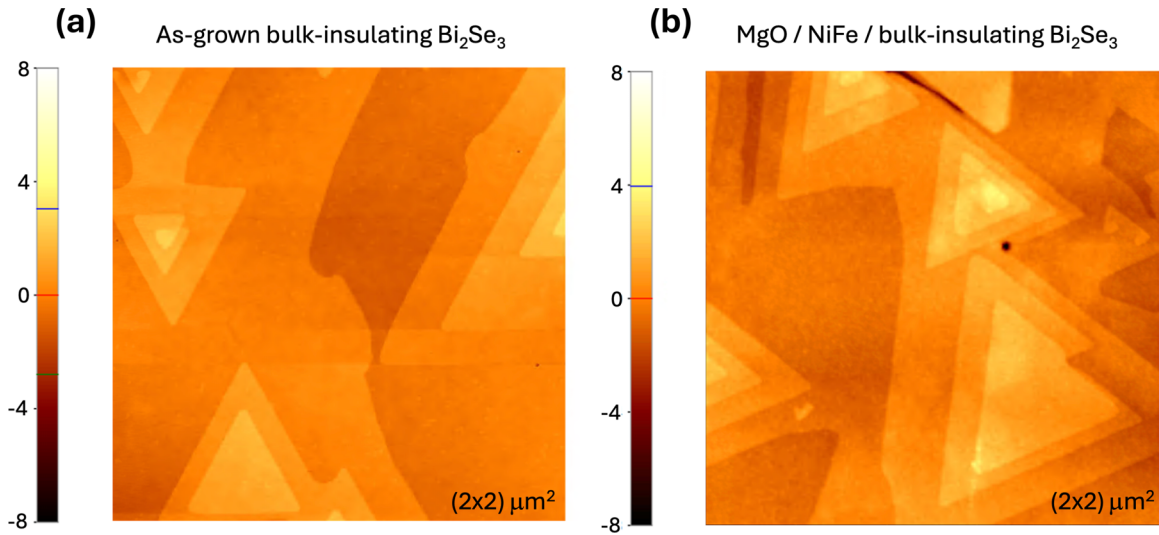


FIG. 7. Representative atomic force microscopy (AFM) images of (a) an as-grown BI-Bi₂Se₃ thin film and (b) a MgO-capped Ni₈₀Fe₂₀/BI-Bi₂Se₃ heterostructure.

atmosphere. AFM images of the magnetic heterostructure [Fig. 7(b)] clearly shows a similar surface morphology, indicating that the Ni₈₀Fe₂₀ is conformal to the Bi₂Se₃ underlayer.

7. Ferromagnetic resonance

Frequency-dependent linewidth broadening of the ferromagnetic resonance (FMR) allows for an estimate of the spin mixing conductance, G_{eff} , to be made [72]. Here, the effect of spin pumping across the magnetic/nonmagnetic interface leads to increased spin dampening in the ferromagnet, resulting in a characteristically broadened linewidth as illustrated for yttrium-iron-garnet (YIG)/Bi₂Se₃ in Fig. 8. In contrast, frequency-dependent FMR linewidths for both Ni₈₀Fe₂₀/BC-Bi₂Se₃ and Ni₈₀Fe₂₀/BI-Bi₂Se₃ exhibit no change in comparison to Ni₈₀Fe₂₀ alone. Such null findings reflect the inherent challenge of performing these experiments on Ni₈₀Fe₂₀/Bi₂Se₃. This includes the fact that spin injection efficiency is significantly hindered when the conductivity of the ferromagnetic metal is much higher than that of the nonmagnetic medium (e.g., Bi₂Se₃) [59], meaning few spins are transferred/injected across the interface into the Bi₂Se₃ layer. Additionally, the larger Gilbert damping in Ni₈₀Fe₂₀ as compared to YIG (Fig. 8) makes it more challenging to detect the signature changes in linewidth broadening that result from spin pumping, particularly under such small spin transfers. Hence, despite our null result, these findings are indicative of

there being few spins that cross the interface, consistent with a conductivity mismatch as proposed on the basis of our THz emission experiments.

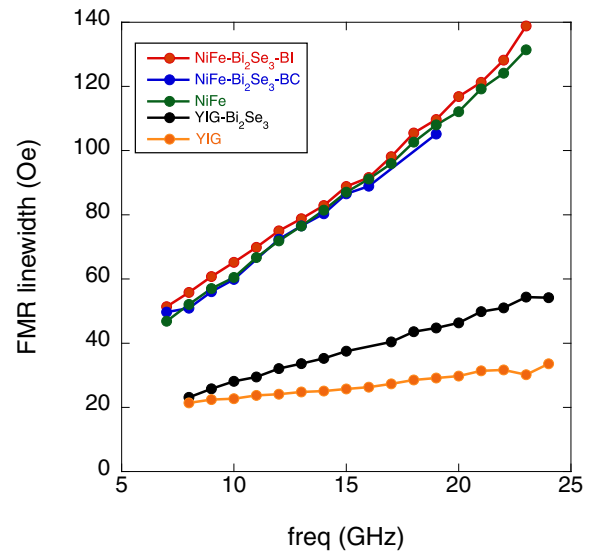


FIG. 8. Frequency-dependent FMR linewidths of YIG (orange), YIG/Bi₂Se₃ (black), Ni₈₀Fe₂₀ (green), Ni₈₀Fe₂₀/BC-Bi₂Se₃ (blue), and Ni₈₀Fe₂₀/BI-Bi₂Se₃ (red).

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