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Bi₂Te₃/Fe₄GeTe₂ probed by terahertz spectroscopy

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Ultrafast electron transport in 2D van der Waals heterostructures $\text{Bi}_2\text{Te}_3/\text{Fe}_4\text{GeTe}_2$ probed by terahertz spectroscopy

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The research on two-dimensional (2D) magnetic materials and their heterostructures is crucial in fields like spintronics, materials science, and condensed matter physics. This study uses terahertz (THz) time-domain spectroscopy to investigate ultrafast electron transport properties in both van der Waals Fe_4GeTe_2 films and $\text{Bi}_2\text{Te}_3/\text{Fe}_4\text{GeTe}_2$ ferromagnetic/topological heterostructures. Our results show that these heterostructures exhibit effective THz electromagnetic shielding. The complex conductivity spectra of Fe_4GeTe_2 films and $\text{Bi}_2\text{Te}_3/\text{Fe}_4\text{GeTe}_2$ heterostructures with varying Fe_4GeTe_2 thicknesses are analyzed using the Drude–Smith model. We quantitatively examine how Fe_4GeTe_2 layer thickness affects the direct current conductivity, plasma frequency, carrier momentum scattering time, and back-scattering coefficient. As the number of Fe_4GeTe_2 layers increases, intra-layer back-scattering events for charge carriers become more frequent. This work provides THz frequency spectra for both Fe_4GeTe_2 and $\text{Bi}_2\text{Te}_3/\text{Fe}_4\text{GeTe}_2$, aiding in the design and optimization of THz modulators and detectors.

Keywords: terahertz time domain spectroscopy, $\text{Bi}_2\text{Te}_3/\text{Fe}_4\text{GeTe}_2$ heterostructures, terahertz conductivity spectrum, Drude–Smith model

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1. Introduction

Van der Waals (vdW) heterostructures, constructed by stacking different two-dimensional (2D) layered materials through vdW forces, not only overcome traditional lattice matching limitations but also exhibit unique physical properties via interfacial coupling.^[1–3] The vdW heterostructures show great potential in photodetectors and nonvolatile memories, making them a key research focus in condensed matter physics and materials science.^[4,5] In recent years, vdW heterostructures combining 2D magnetic materials and topological insulators have gained attention.^[6,7] The 2D magnetic vdW heterostructures have opened new avenues for the design of low-energy and high-efficiency spintronic devices.^[8–12]

Fe_4GeTe_2 , an emerging vdW ferromagnetic material with a higher Curie temperature and interlayer magnetic anisotropy, is considered a promising candidate for room-temperature spintronics devices.^[13] The magnetic properties of Fe_4GeTe_2 can be efficiently modulated by external electric fields or strain.^[14] Wang *et al.* demonstrated that the interfacial cou-

pling between Fe_4GeTe_2 and sapphire substrate can increase the Curie temperature of Fe_4GeTe_2 up to 530 K.^[13] Recently, Wang *et al.* constructed a $\text{Bi}_2\text{Te}_3/\text{Fe}_3\text{GeTe}_2$ vdW heterostructure by combining Fe_3GeTe_2 with the topological insulator Bi_2Te_3 .^[15] The heterostructures not only utilize the high mobility surface state of Bi_2Te_3 to enhance the charge transport efficiency, but also induce a topological magnetic structure through interfacial spin–orbit coupling and realize efficient spin-charge conversion.^[15,16] Through magnetic and electrical characterizations, it was demonstrated that the topological insulator Bi_2Te_3 could increase the Curie temperature of Fe_3GeTe_2 up to 400 K through the proximity effect. Thus, the Bi_2Te_3 is essential for achieving 2D room-temperature ferromagnetism^[15] and worm-like magnetic domain structure through heterostructure engineering.^[17]

In addition to the investigations of magnetic and electrical properties, the ultrafast dynamical properties of 2D magnetic materials and their heterostructures have attracted much attention.^[18] Liu *et al.* experimentally demonstrated that the magnetization strength and magnetic anisotropy of Fe_3GeTe_2

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can be modulated by femtosecond laser pulses.^[19] Li *et al.* found that, in the Fe/Bi₂Se₃ heterostructures the Bi₂Se₃ topological surface states not only accelerated the ultrafast demagnetization process, but also significantly enhanced the magnetic damping factor.^[20] Gong *et al.* showed that the amplitude, frequency and lifetime of spin waves in the vdW material Fe_{3.6}Co_{1.4}GeTe₂ can be effectively manipulated by modulating the polarization state of the laser pulse.^[21] Chen *et al.* investigated the generation and control of terahertz (THz) spin flow in the Fe₃GeTe₂/Bi₂Te₃ heterostructures, and the Fe₃GeTe₂ significantly enhances the THz pulse emission radiated from Bi₂Te₃.^[22] Recently, Tomarchio *et al.* found a correlation between the photo-induced THz electrodynamics of Fe₃GeTe₂ and Fe₅GeTe₂ films and their ultrafast demagnetization under femtosecond laser pulse irradiation.^[23] Based on current research, the Bi₂Te₃/Fe₄GeTe₂ heterostructures show promise for next-generation opto-electromagnetic information technology.

Optical properties in the THz range, such as complex permittivity and conductivity, help understand material electronic structures and carrier dynamics, and are crucial for developing ultrafast optoelectronic devices.^[24–29] THz time-domain spectroscopy (THz-TDS) uses coherent detection to measure the waveform of THz pulses, obtaining both amplitude and phase information.^[30] This technique features a wide bandwidth and high signal-to-noise ratio, enabling the acquisition of the THz conductivity of thin-film sample in a single measurement.^[31–37] In 2015, Jin *et al.* used THz-TDS to determine the fundamental parameters of the Mott conduction process, including the spin-resolved electron scattering time and electron density.^[31] In 2020, Jin *et al.* designed a silicon-based magnetic tunneling junction, which realizes the magnetic modulation of THz pulses by modulating the spin tunneling flow on the picosecond time scale.^[32] In addition, THz-TDS has been applied to study the ultrafast electron transport properties in antiferromagnetic Mn₂Au thin films,^[33] and ferromagnetic Fe thin films.^[34] The experimental results demonstrate that variations in the sample thickness affect the scattering and localization behavior of charge carriers.

Recently, Li *et al.* prepared Fe₃GeTe₂ single crystals with good electrical, magnetic, and electromagnetic wave absorption and THz shielding properties.^[38] Suo *et al.* investigated the photoinduced charge transport properties in vdW ferromagnetic semiconductors such as CrSiTe₃ and CrGeTe₃.^[39,40] THz emission spectroscopy and THz-TDS have been used to investigate the photoexcited currents in Sb₂Te₃-GeTe and photoconductivity in Sb₂Se₃/Si heterojunctions, respectively.^[41,42] However, there is insufficient exploration of ultrafast charge carrier transport in vdW ferromagnetic film and ferromagnetic/topological heterostructures. THz electrodynamic characterization is essential to reveal ul-

trafast optoelectronic properties and guide device design.

In this work, we employ the method of THz-TDS to investigate the THz conductivity spectra of both Fe₄GeTe₂ films and Bi₂Te₃/Fe₄GeTe₂ heterostructures. It is demonstrated that the Bi₂Te₃/Fe₄GeTe₂ heterostructures have good shielding efficiency against THz electromagnetic interference. The prepared Bi₂Te₃(8 nm)/Fe₄GeTe₂(12 nm) heterostructure achieves an average THz electromagnetic interference shielding value of 344.8 ± 27.8 dB/ μ m. The complex conductivities of both Fe₄GeTe₂ films and Bi₂Te₃/Fe₄GeTe₂ heterostructures were fitted by the Drude–Smith (DS) model. The extracted DC conductivity, plasma frequency, carrier momentum scattering time, and back-scattering factor were quantitatively analyzed as functions of the Fe₄GeTe₂ layer thickness. This study demonstrates the ultrafast charge carrier transport in both Fe₄GeTe₂ films and Bi₂Te₃/Fe₄GeTe₂ heterostructures within the THz frequency range, supporting the design of THz modulation and detector devices based on vdW ferromagnetic/topological heterostructures.

2. Sample preparation

High-quality 2D Fe₄GeTe₂ films and Bi₂Te₃/Fe₄GeTe₂ heterostructures were fabricated by molecular beam epitaxy (MBE) on the 500- μ m Al₂O₃ sapphire substrate. The background vacuum of the system was maintained at 10^{-10} Torr (1 Torr = 1.33322×10^2 Pa) during the experimental procedure. During the growth process, the substrate was first annealed at 600 °C for 1 h to remove the impurity layer, and then cooled down to 280 °C. For the Fe₄GeTe₂ films, elemental Fe, Ge, and Te were evaporated from standard Knudsen cells as the source materials. For the Bi₂Te₃/Fe₄GeTe₂, the high-purity Bi, Fe, Ge, and Te were then evaporated sequentially from a standard Knudsen evaporation source. The flux ratio of Bi and Te was precisely controlled to be 1:15 for the growth of the Bi₂Te₃ layer, while the flux ratio of Fe, Ge, and Te was 3:1:10 for the growth of the Fe₄GeTe₂ layer. After the growth of Bi₂Te₃/Fe₄GeTe₂, a 2-nm-thick Ge protective layer was deposited on the surface of the Fe₄GeTe₂ layer to prevent oxidation by ambient gases. The thicknesses of the Fe₄GeTe₂ films were 2 nm, 6 nm, 10 nm, and 12 nm. Within the Bi₂Te₃/Fe₄GeTe₂ heterostructures, the Bi₂Te₃ layer is 8-nm thick.

3. Experimental setup

The THz electrodynamic properties of both Fe₄GeTe₂ films and Bi₂Te₃/Fe₄GeTe₂ heterostructures were investigated by a transmitted THz-TDS, and the experimental setup is shown in Fig. 1. Laser pulses with a center wavelength of 800 nm, a repetition frequency of 1 kHz, and a pulse width of 120 fs were generated by titanium sapphire lasers (Mira 900

and Legend Elite Duo, Coherent Inc). The femtosecond laser pulses were split into two beams by a beam splitter: the high power beam was used to excite the ZnTe crystal to emit THz pulses radiation, and the low power beam was used for coherent detection of the THz pulses. The high power beam is frequency modulated by a chopper and then normal incident on a 1-mm thick $\langle 110 \rangle$ ZnTe crystal to generate THz radiation. The generated THz pulses are collected by off-axis parabolic mirrors and focused onto a 1-mm thick $\langle 110 \rangle$ ZnTe probe crystal. A low-power probe beam is time-delayed and incident on a ZnTe crystal, which propagates co-linearly with the THz pulse. The time-domain waveforms of the THz pulses were recorded using a free-space electro-optical sampling method. In the experiment, the sample was placed at the focal point of the THz beam. Weak signal acquisition was realized using a balanced photodetector and a lock-in amplifier. All THz spectral data were measured at room temperature and in a dry air environment (relative humidity below 10%).

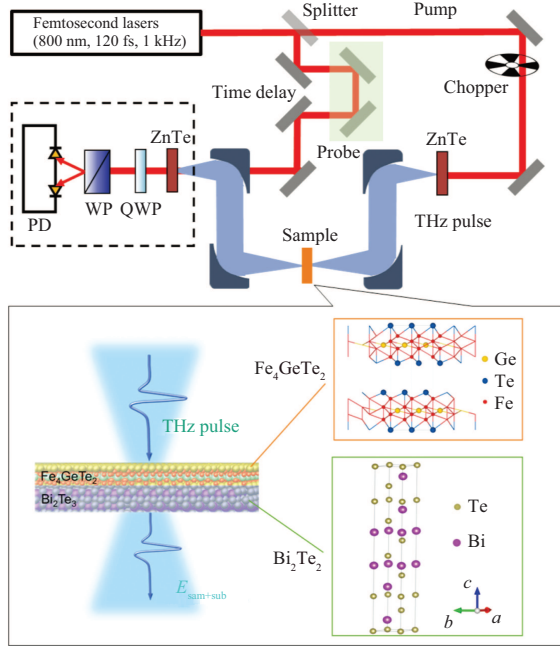


Fig. 1. The schematic diagram of the THz-TDS measurement for both Fe_4GeTe_2 films and $\text{Bi}_2\text{Te}_3/\text{Fe}_4\text{GeTe}_2$ heterostructures. WP: Wollaston prism, QWP: quarter wave plate, PD: photo-diode.

4. Experimental data analysis

Figure 2(a) shows the time-domain THz transmission signal $E_{\text{sam+sub}}(t)$ and $E_{\text{sub}}(t)$ through Fe_4GeTe_2 films on the sap-

phire substrate and the bare sapphire substrate, respectively. These signals are transformed into the frequency-domain using Fourier transform. The corresponding frequency-domain signals of Fe_4GeTe_2 films $\tilde{E}_{\text{sam+sub}}(\omega)$ and $\tilde{E}_{\text{sub}}(\omega)$ are shown in Fig. 2(b). Figure 2(c) shows $E_{\text{sam+sub}}(t)$ and $E_{\text{sub}}(t)$ for $\text{Bi}_2\text{Te}_3/\text{Fe}_4\text{GeTe}_2$ heterostructures with Fe_4GeTe_2 layer thicknesses of 2 nm, 6 nm, 10 nm, and 12 nm and the sapphire substrate. Figure 2(d) shows $\tilde{E}_{\text{sam+sub}}(\omega)$ for $\text{Bi}_2\text{Te}_3/\text{Fe}_4\text{GeTe}_2$ heterostructures and $\tilde{E}_{\text{sub}}(\omega)$ for the sapphire substrate. It can be seen that THz transmittances decrease gradually with increasing Fe_4GeTe_2 thickness for both Fe_4GeTe_2 films and $\text{Bi}_2\text{Te}_3/\text{Fe}_4\text{GeTe}_2$ heterostructures.

The THz electric field pulse induces a time-varying current in the sample based on its conductivity, causing attenuation and phase delay as it propagates. The THz complex conductivity spectra of both Fe_4GeTe_2 films and $\text{Bi}_2\text{Te}_3/\text{Fe}_4\text{GeTe}_2$ heterostructures can be obtained from $\tilde{E}_{\text{sub}}(\omega)$ and $\tilde{E}_{\text{sam+sub}}(\omega)$,^[43,44]

$$\frac{\tilde{E}_{\text{sam+sub}}(\omega)}{\tilde{E}_{\text{sub}}(\omega)} = \frac{1 + n_{\text{sub}}}{1 + n_{\text{sub}} + Z_0 \tilde{\sigma}(\omega) d_{\text{sam}}} \times \exp \left[\frac{i\omega}{c} (n_{\text{sub}} - 1) \Delta L \right], \quad (1)$$

where the free-space impedance $Z_0 = \sqrt{\mu_0/\epsilon_0} = 377 \, \Omega$, and $\tilde{\sigma}(\omega)$ is the complex conductivity. n_{sub} is the refractive index of the substrate within the THz frequency range. d_{sam} is the total thickness of the heterostructures. $\omega = 2\pi f$ and f is the THz frequency. ΔL is the difference in thickness between the substrate on which the samples were grown and the reference substrate. For Fe_4GeTe_2 (2, 6, 10, 12 nm), ΔL values are 1.2, 1.5, 1.4, and 2.6 μm , respectively. ΔL values for $\text{Bi}_2\text{Te}_3/\text{Fe}_4\text{GeTe}_2$ (2, 6, 10, 12 nm) are 2.6, 2.0, 4.0, and 3.3 μm . The ΔL values were experimentally measured using a high-precision digital micrometer. When applying Eq. (1) to calculate $\tilde{\sigma}(\omega)$, the conductivity error caused by the phase error $(\omega/c)(n_{\text{sub}} - 1)\Delta L$, which is induced by ΔL , has been compensated.^[45]

The refractive index of the Al_2O_3 substrate is calculated by the following equation based on the reference signal from air $E_{\text{air}}(t)$ and the substrate signal $E_{\text{sub}}(t)$, as shown in Fig. 2(e). We assume a plane wave impinging on the substrate at normal incidence. The transmittance is given by

$$T(\omega) = \frac{\tilde{E}_{\text{sub}}(\omega)}{\tilde{E}_{\text{air}}(\omega)} = A(\omega) \exp(-i\Delta\phi) = T_1(\omega) T_2(\omega) \cdot \exp \left(-i \frac{(\tilde{n}_{\text{sub}}(\omega) - 1) \omega d_{\text{sub}}}{c} \right), \quad (2)$$

where $\Delta\phi$ is the phase difference between $\tilde{E}_{\text{air}}(\omega)$ and $\tilde{E}_{\text{sub}}(\omega)$, $\tilde{n}_{\text{sub}}(\omega)$ is the complex refractive index of the substrate, c is the speed of light in vacuum, and d_{sub} is the thickness of Al_2O_3 substrate ($d_{\text{sub}} = 0.433 \, \text{mm}$). The Fresnel transmission coefficients for the air-sample and sample-air interfaces are $T_1(\omega) = 2/(1 + \tilde{n}_{\text{sub}}(\omega))$, $T_2(\omega) = 2\tilde{n}_{\text{sub}}(\omega)/(1 + \tilde{n}_{\text{sub}}(\omega))$, respectively. Here, $\tilde{n}_{\text{sub}}(\omega) = n_{\text{sub}}(\omega) - i\kappa_{\text{sub}}(\omega)$ is considered, thus

$$T(\omega) = \frac{4(n_{\text{sub}} - i\kappa)}{(1 + (n_{\text{sub}} - i\kappa))^2} \cdot \exp(-\alpha \cdot d_{\text{sub}}) \exp \left(-i \frac{\omega(n_{\text{sub}} - 1) \cdot d_{\text{sub}}}{c} \right), \quad (3)$$

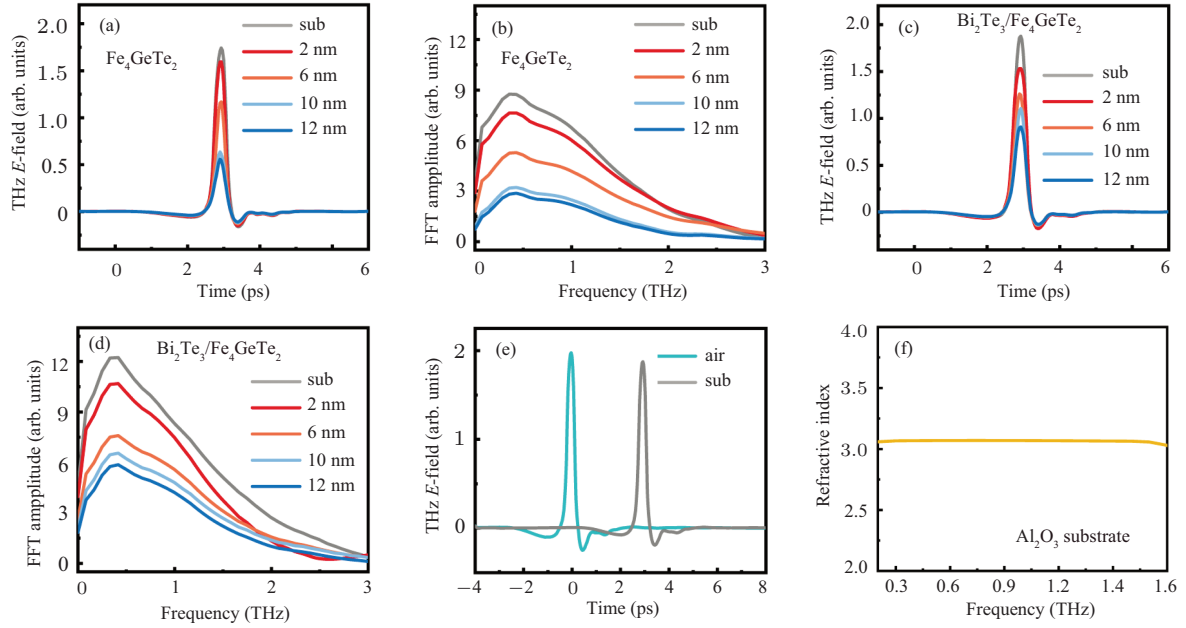


Fig. 2. (a) THz electric fields transmitted through Fe_4GeTe_2 films with varying thicknesses and Al_2O_3 substrate. (b) The transmitted THz signals in the frequency domain. (c) THz electric fields transmitted through $\text{Bi}_2\text{Te}_3/\text{Fe}_4\text{GeTe}_2$ heterostructures and Al_2O_3 substrate. (d) Transmitted THz signals in the frequency domain. (e) THz pulse transmitted through air as a reference and through the Al_2O_3 substrate. (f) Refractive index of the Al_2O_3 substrate at room temperature.

where $\alpha_{\text{sub}} = 2\omega\kappa_{\text{sub}}/c$ is the absorption coefficient. Owing to the small attenuation of THz propagation inside the Al_2O_3 substrate, and the condition $\kappa \ll n$ holding true, $n_{\text{sub}}(\omega)$ and $\alpha_{\text{sub}}(\omega)$ can be determined by^[46–48]

$$n_{\text{sub}}(\omega) = \frac{c\Delta\varphi}{\omega d_{\text{sub}}} + 1, \quad (4)$$

$$\alpha_{\text{sub}}(\omega) = \frac{2}{d_{\text{sub}}} \cdot \ln \left[\frac{4n_{\text{sub}}(\omega)}{A(\omega) \cdot ((n_{\text{sub}}(\omega) + 1))^2} \right]. \quad (5)$$

The average refractive index value of the Al_2O_3 substrate was calculated to be about 3.07 in a range of 0.2 THz–1.8 THz, as shown in Fig. 2(f).

Figures 3(a) and 3(b) show the THz complex conductivities of both Fe_4GeTe_2 films and $\text{Bi}_2\text{Te}_3/\text{Fe}_4\text{GeTe}_2$ heterostructures at room temperature, calculated by Eq. (1). The blue squares and the red circles represent the real part $\sigma_{\text{real}}(\omega)$ and the imaginary part $\sigma_{\text{imag}}(\omega)$ of the complex conductivity, respectively. As shown in Figs. 3(a) and 3(b), the $\sigma_{\text{real}}(\omega)$ are positive from 0.2 THz–1.8 THz. In addition, the $\sigma_{\text{real}}(\omega)$ increases with the thickness of the Fe_4GeTe_2 film. The $\sigma_{\text{real}}(\omega)$ of $\text{Bi}_2\text{Te}_3/\text{Fe}_4\text{GeTe}_2$ heterostructures are larger than that of pure Fe_4GeTe_2 films. While, $\sigma_{\text{imag}}(\omega)$ are near to zero. It is noteworthy that $\sigma_{\text{imag}}(\omega)$ becomes negative at high-frequencies for the thicker Fe_4GeTe_2 (12 nm) film and $\text{Bi}_2\text{Te}_3/\text{Fe}_4\text{GeTe}_2$ (10 nm and 12 nm) heterostructures.

The THz transmission spectrum allows calculating the THz electromagnetic interference shielding (EMI SE) for both Fe_4GeTe_2 films and $\text{Bi}_2\text{Te}_3/\text{Fe}_4\text{GeTe}_2$ heterostructures,

$$\text{EMI SE} = -20 \log \left(\frac{\tilde{E}_{\text{sam+sub}}(\omega)}{\tilde{E}_{\text{sub}}(\omega)} \right). \quad (6)$$

The THz EMI SE of Fe_4GeTe_2 films and $\text{Bi}_2\text{Te}_3/\text{Fe}_4\text{GeTe}_2$ heterostructures are plotted in Fig. 3(c). The THz EMI SE of the samples increases with the thickness of the Fe_4GeTe_2 layer. The average values of THz EMI SE coefficient for Fe_4GeTe_2 (12 nm) and $\text{Bi}_2\text{Te}_3/\text{Fe}_4\text{GeTe}_2$ (12 nm) are about 5.1 ± 0.3 dB and 6.9 ± 0.5 dB, respectively. In order to objectively evaluate the THz EMI shielding performance, the normalized thickness EMI SE value is utilized.^[49–55] The average normalized thickness EMI SE for the $\text{Bi}_2\text{Te}_3/\text{Fe}_4\text{GeTe}_2$ (12 nm) was 344.8 ± 27.8 dB/ μm within the frequency range of 0.2 THz–1.8 THz. This value surpasses that reported for Fe_3GeTe_2 single-crystal thin films^[38] and Mxene thin films,^[49] but is lower than the values observed in Mxene thin films with a pleated structure^[54] and PdTe_2 semimetallic films.^[55]

In order to deeply investigate the carrier transport properties in both Fe_4GeTe_2 films and $\text{Bi}_2\text{Te}_3/\text{Fe}_4\text{GeTe}_2$ heterostructures in the THz frequency range, we analyzed the experimentally measured $\tilde{\sigma}(\omega)$ with the DS model,^[56,57]

$$\tilde{\sigma}(\omega) = \frac{\epsilon_0 \omega_p^2 \tau}{1 - i\omega\tau} \left[1 + \frac{c}{1 - i\omega\tau} \right], \quad (7)$$

where ϵ_0 denotes the vacuum permittivity, ω_p is the plasma frequency, and τ is the momentum scattering time. The parameter c , ranging from -1 to 0 , is a phenomenological coefficient that quantifies the backscattering or restoring force, thereby characterizing the degree of carrier localization. As shown in Figs. 3(a) and 3(b), the blue solid lines are the fits of $\sigma_{\text{real}}(\omega)$, and the red solid lines are the fits of $\sigma_{\text{imag}}(\omega)$. The DS model shows good agreement with the experimental results. Through DS fitting, we extract the Fe_4GeTe_2 thickness-

dependent charge transport parameters, including ω_p , τ , c , and σ_{dc} , for Fe_4GeTe_2 films and $\text{Bi}_2\text{Te}_3/\text{Fe}_4\text{GeTe}_2$ heterostructures, which are summarized in Fig. 4.

The plasma frequency ω_p is a characteristic frequency. Below it, materials behave like metals; above it, they are transparent to light. As shown in Fig. 4(a), the ω_p increases as the thickness of the Fe_4GeTe_2 increases, indicating an increase in carrier concentration. As shown in Fig. 4(b), τ of Fe_4GeTe_2 and $\text{Bi}_2\text{Te}_3/\text{Fe}_4\text{GeTe}_2$ decrease from $\tau = 31.9 \pm 4.0$ fs to 7.1 ± 0.9 fs, and from $\tau = 49.7 \pm 3.9$ fs to 19.2 ± 1.3 fs, respectively, as the Fe_4GeTe_2 thickness decreases from 12 nm to 2 nm. The average momentum scattering times are about 16.8 fs and 33.8 fs for Fe_4GeTe_2 and $\text{Bi}_2\text{Te}_3/\text{Fe}_4\text{GeTe}_2$, respectively, which are nearly equivalent to that of a single-layer Fe film.^[58]

The DS model is commonly employed to describe semiconductor nanostructures or metal films, in which the charges experience backscattering or localization effects during transport. Backscattering refers to the phenomenon that carriers have a certain probability of reversing direction upon collision with other particles or grain boundaries. In this model, the parameter c is associated with the persistence of veloc-

ity effect and preserves the memory of the initial velocity. When $c = -1$, it indicates that the carriers are in a fully backscattered state; when $c = 0$, the DS model reduces to the Drude model, indicating that carriers are in a completely randomized state.^[59] It is worth noting that when applying the DS model to the conductivity spectroscopy of vdW ferromagnetic films and ferromagnetic/topological heterostructures, one must adequately account for not only the scattering of electrons within the inter-layer and at grain boundaries, but also the multiple back-scattering between different 2D layered Fe_4GeTe_2 . As shown in Fig. 4(c), the c -factor of Fe_4GeTe_2 and $\text{Bi}_2\text{Te}_3/\text{Fe}_4\text{GeTe}_2$ gradually decrease from -0.32 ± 0.06 to -0.67 ± 0.11 and from -0.14 ± 0.04 to -0.57 ± 0.16 , respectively, as the thickness of Fe_4GeTe_2 increases from 2 nm to 12 nm. Using the extracted ω_p , τ , and c factor of the samples, we can further determine the DC conductivity $\sigma_{dc} = \epsilon_0 \omega_p^2 \tau (1 + c)$.^[60,61] The σ_{dc} of Fe_4GeTe_2 and $\text{Bi}_2\text{Te}_3/\text{Fe}_4\text{GeTe}_2$ increase monotonically from 899 ± 124 S/cm to 2477 ± 319 S/cm and from 1139 ± 295 S/cm to 5142 ± 237 S/cm, respectively, as the thickness of Fe_4GeTe_2 is increased from 2 nm to 12 nm, as shown in Fig. 4(d).

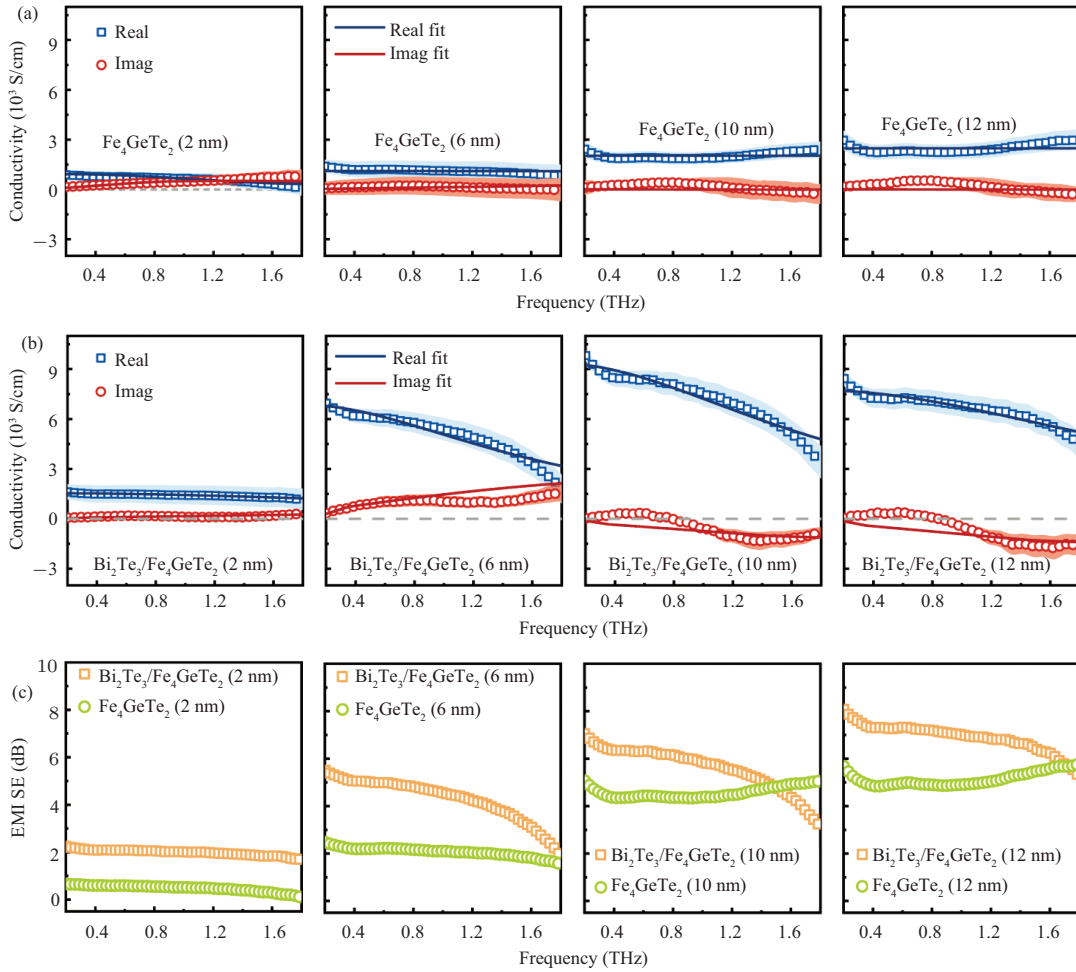


Fig. 3. The real (blue squares) and imaginary (red circles) components of the THz conductivity spectra for (a) Fe_4GeTe_2 films and (b) $\text{Bi}_2\text{Te}_3/\text{Fe}_4\text{GeTe}_2$ heterostructures with different thicknesses of Fe_4GeTe_2 layers at room temperature. The solid lines are the fitting results of the Drude-Smith model. (c) THz EMI SE of both Fe_4GeTe_2 films and $\text{Bi}_2\text{Te}_3/\text{Fe}_4\text{GeTe}_2$ heterostructures with varying Fe_4GeTe_2 thicknesses as a function of THz frequency.

As shown in Fig. 4(e), the thickness of an Fe_4GeTe_2 monolayer is $\sim 9.7 \text{ \AA}$ and adjacent Fe_4GeTe_2 monolayer are separated by a vdW gap. As the thickness of the Fe_4GeTe_2 layer increases, the number of included 2D ferromagnetic layers also increases, leading to a higher probability of intra-layer backscattering events for charge carriers. Consequently, the c factor gradually approaches -0.67 ± 0.11 and -0.57 ± 0.16 for Fe_4GeTe_2 and $\text{Bi}_2\text{Te}_3/\text{Fe}_4\text{GeTe}_2$, respectively. Our experimental results indicate that the topological insulator Bi_2Te_3 increases the conductivity and reduces the localization of carriers in the heterostructures, without changing the thickness de-

pendence of electron transports within vdW Fe_4GeTe_2 films. However, a mixing band structure of $\text{Bi}_2\text{Te}_3/\text{Fe}_4\text{GeTe}_2$ heterostructure is required to know whether the surfaces states of Bi_2Te_3 contribute to the conductivity spectra, which will be our future research focus. Furthermore, we plan to employ both optical pumping-THz probing techniques and THz emission spectroscopy^[62,63] to investigate the photoinduced transient conductivity and photocurrents in $\text{Bi}_2\text{Te}_3/\text{Fe}_4\text{GeTe}_2$ heterostructures, as well as its correlation with ultrafast spin dynamics.

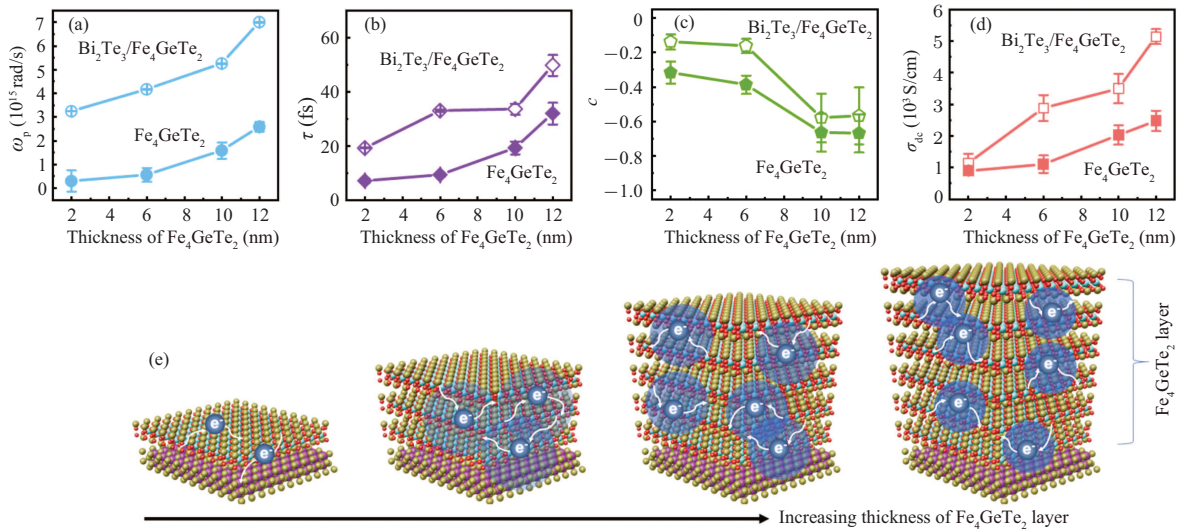


Fig. 4. The Drude-Smith fit parameters are plotted. (a) ω_p , (b) τ , (c) c , and (d) σ_{dc} , for both Fe_4GeTe_2 films and $\text{Bi}_2\text{Te}_3/\text{Fe}_4\text{GeTe}_2$ heterostructures as functions of Fe_4GeTe_2 layer thickness at room temperature. (e) Schematic diagram of carrier back-scattering in layered Fe_4GeTe_2 structures with different thicknesses.

5. Summary

To summarize, the THz transmission spectrum of both Fe_4GeTe_2 films and $\text{Bi}_2\text{Te}_3/\text{Fe}_4\text{GeTe}_2$ heterostructures was characterized by THz-TDS to extract the THz complex conductivity. The electromagnetic interference shielding properties of heterostructures with varying thicknesses in the THz frequency range was evaluated using time-resolved THz spectroscopy. Additionally, the THz conductivity spectra of both Fe_4GeTe_2 films and $\text{Bi}_2\text{Te}_3/\text{Fe}_4\text{GeTe}_2$ heterostructures with different Fe_4GeTe_2 layer thicknesses were fitted using the DS model to quantitatively determine the microscopic transport parameters of charges. It was observed that the DC conductivity increases with the thickness of the Fe_4GeTe_2 layer. As the number of 2D ferromagnetic layers increases, the c factor of Fe_4GeTe_2 and $\text{Bi}_2\text{Te}_3/\text{Fe}_4\text{GeTe}_2$ approaches -0.67 ± 0.11 and -0.57 ± 0.16 , respectively, indicating enhanced back-scattering processes for electrons. The spectral information and physical parameters elucidated in this work have implications for the potential application of the vdW $\text{Bi}_2\text{Te}_3/\text{Fe}_4\text{GeTe}_2$ heterostructures in fields such as electronics and optoelectronics.

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