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Abstract

© 2020 American Physical Society. Magnetic tunnel junctions (MTJs) are a key technology in modern spintronics because they are the basis of read-heads of modern hard disk drives, nonvolatile magnetic random access memories, and sensor applications. In this paper, we demonstrate that tunneling magnetoresistance can influence terahertz (THz) wave propagation through a MTJ. In particular, various magnetic configurations between parallel state and antiparallel state of the magnetizations of the ferromagnetic layers in the MTJ have the effect of changing the conductivity, making a functional modulation of the propagating THz electromagnetic fields. Operating in the THz frequency range, a maximal modulation depth of 60% is reached for the parallel state of the MTJ with a thickness of 77.45 nm, using a magnetic field as low as 30 mT. The THz conductivity spectrum of the MTJ is governed by spin-dependent electron tunneling. It is anticipated that the MTJ device and its tunability scheme will have many potential applications in THz magnetic modulators, filtering, and sensing.

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Magnetic Modulation of Terahertz Waves via Spin-Polarized Electron Tunneling Based on Magnetic Tunnel Junctions

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Magnetic tunnel junctions (MTJs) are a key technology in modern spintronics because they are the basis of read-heads of modern hard disk drives, nonvolatile magnetic random access memories, and sensor applications. In this paper, we demonstrate that tunneling magnetoresistance can influence terahertz (THz) wave propagation through a MTJ. In particular, various magnetic configurations between parallel state and antiparallel state of the magnetizations of the ferromagnetic layers in the MTJ have the effect of changing the conductivity, making a functional modulation of the propagating THz electromagnetic fields. Operating in the THz frequency range, a maximal modulation depth of 60% is reached for the parallel state of the MTJ with a thickness of 77.45 nm, using a magnetic field as low as 30 mT. The THz conductivity spectrum of the MTJ is governed by spin-dependent electron tunneling. It is anticipated that the MTJ device and its tunability scheme will have many potential applications in THz magnetic modulators, filtering, and sensing.

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I. INTRODUCTION

Terahertz (THz) radiation lies in the frequency range from 0.1 to 10 THz, between the microwave and infrared regions [1,2]. Recent technological innovation in photonics and nanotechnology is enabling THz technology to be applied in information and communications [3,4], imaging and spectroscopy [5,6], biology and medicine [7,8], nondestructive evaluation and security [9]. Among the potential THz applications, imaging and communications

are two important research subjects that may bring great breakthroughs in modern optoelectronic information technology. Therefore, suitable functional devices and materials are in high demand for modulation or control of THz radiation [10,11]. THz modulation has been demonstrated by optical, electronic, thermal, and mechanical methods [12–16]. Usually, optical methods require an intense ultra-short femtosecond laser beam, while thermal methods are limited to the scale of tens of milliseconds. In addition, these traditional modulation methods are not entirely suitable for the THz wave bandwidth.

It is noteworthy that THz transients have become increasingly important for investigation of the light-matter interaction in spintronic materials, since many dynamic processes such as spin currents [17–19] and magnetic spin waves [20,21] oscillate with THz frequencies. There

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have been numerous THz devices controlled by external magnetic fields demonstrated for amplitude modulation [22–24], phase retardation [25], and polarization control [26,27]. Recently, liquid-suspended magnetic ferrofluid Fe_3O_4 nanoparticles and ferrofluid-filled photonic crystals have been widely used to realize THz magnetic modulators, based on magnetic induced birefringence and dichroism [22–24]. However, the THz modulation depths of 66% at 35 mT, 40% at 150 mT, and 34% at 194 mT require significantly long propagation lengths of 10 mm [22], 120 μm [23], and 1 mm [24], respectively. Therefore, the development of solid-state devices is still an important topic in terms of practical THz magnetic modulators.

In this respect, magnetoresistance is an important spintronic effect for changing the conductivity response and influencing THz electromagnetic wave propagation. Chau *et al.* demonstrated that anisotropic magnetoresistance (AMR) can be used to modulate the transmission of THz radiation in both ensemble subwavelength Co microparticles [28] and dense micrometer-sized Co particles coated with Au microparticles [29]. Recently, we have shown that the application of a magnetic field up to 100 mT to a giant magnetoresistive (GMR) structure induces a striking reduction of the transmission of the THz field by about 20%. The difference in conduction by electrons with opposite spins in ferromagnetic (FM) metals was accessed precisely in a GMR stack [30]. Although the tunneling magnetoresistance (TMR) effect (TMR ratio up to around 200%) has been probed by dc measurement statically [31] and studied within the infrared regime (15–150 THz) using contactless reflection spectroscopy [32], a quantitative investigation of the spin-polarized electron-tunneling-induced conductivity change in a TMR structure at THz frequencies has not yet been reported. In the present paper, we use single-cycle THz pulses to drive the spin-polarized charge across a FM/MgO/FM interface over picosecond time scales. We demonstrate that an external magnetic field (approximately 30 mT) tends to align the magnetization of the free FM layer with the pinned FM layer, leading to a strong attenuation of the propagating THz wave. We experimentally measure the increasing change of the sheet conductivity spectrum, as the magnetizations of FM layers are aligned antiparallel at zero magnetic field to co-alignment under the applied magnetic field. A modulation depth as high as 60% is reached for the parallel configuration of a magnetic tunnel junction (MTJ) with a thickness of 77.45 nm, using a magnetic field as low as 30 mT. In this work, we explore the THz TMR effect to offer a solution towards spintronic THz amplitude modulators.

II. EXPERIMENTAL METHODS

Figure 1(a) shows our MTJ multistack grown on a Si substrate. The MTJ multistack is Ta(5)/Ru(10)/ $\text{Ir}_{22}\text{Mn}_{78}$ (10)/ $\text{Co}_{75}\text{Fe}_{25}$ (2.5)/Ru(0.85)/ $\text{Co}_{40}\text{Fe}_{40}\text{B}_{20}$ (3)/MgO(1.9)/

free layer/Ta(5)/Ru(6). The numbers in parentheses indicate the thickness of each layer in nanometers. By sputter-depositing a ferromagnetic $\text{Co}_{75}\text{Fe}_{25}$ film on top of an antiferromagnetic layer of $\text{Ir}_{22}\text{Mn}_{78}$, the spin orientation of the FM thin films can be “pinned” by the exchange bias coupling between the net interfacial moment of antiferromagnetic layer and the FM layer, with a large coercive force to keep the direction of the magnetic moment in the x direction, as shown in Fig. 1(a). The magnetization of the top magnet can be switched by an external magnetic field, which is called a free layer. In this work, the free layer is chosen as $\text{Co}_{40}\text{Fe}_{40}\text{B}_{20}$ (3)/Ru(0.2)/ $\text{Co}_{70.5}\text{Fe}_{4.5}\text{Si}_{15}\text{B}_{10}$ (30). The soft magnetic material $\text{Co}_{70.5}\text{Fe}_{4.5}\text{Si}_{15}\text{B}_{10}$ layer has higher crystalline temperature than $\text{Co}_{40}\text{Fe}_{40}\text{B}_{20}$, which means that MTJs including this layer have both high TMR ratio and low coercivity of the free layer after 300–400 °C annealing. This technique has been demonstrated to provide high sensitivity and low noise for MTJs [33]. Within the free FM layer, the $\text{Co}_{40}\text{Fe}_{40}\text{B}_{20}$ (3) and $\text{Co}_{70.5}\text{Fe}_{4.5}\text{Si}_{15}\text{B}_{10}$ (30) are ferromagnetically coupled via 0.2-nm Ru. While within the pinned FM layer, $\text{Co}_{75}\text{Fe}_{25}$ (2.5) and $\text{Co}_{40}\text{Fe}_{40}\text{B}_{20}$ (3) are antiferromagnetically coupled via 0.85-nm Ru, which is typical for obtaining strong artificial antiferromagnetic coupling. Figure 1(b) shows the magnetic hysteresis loop of our MTJ multistack, using a vibrating sample magnetometer (see Fig. S1 of Ref. [34] for details). TMR is a consequence of spin-dependent tunneling, which originates from an imbalance of the charge current carried by majority (large hollow arrows) and minority spin electrons tunneling from a ferromagnet through an insulating barrier, as shown in the inset of Fig. 1(a). For the nanometer thin FM layers with the parallel configuration of magnetizations (black arrows), the electrons with the same spin orientation with respect to the magnetization can tunnel through the MgO layer easily. As the magnetizations of the two FM layers are opposite, the possibility of electron tunneling between two FM layers becomes smaller, resulting in a smaller tunneling charge current compared to the case for the same directions of the magnetizations. Using the dc four-probe method in a three-dimensional Helmholtz coil system, Fig. 1(c) shows the relative resistance change of our MTJ multistack: $\text{TMR} = (R_{\text{AP}} - R_{\text{P}})/R_{\text{P}} = 210\%$ at $\mathbf{H} = 10$ mT at room temperature, where R_{P} and R_{AP} are the tunneling resistance measured when the magnetizations are aligned in parallel and antiparallel configurations, respectively.

Our THz time-domain spectroscopy (THz-TDS) experiments are performed in a standard linear transmission configuration with the external magnetic field $\mathbf{H}$ applied in the plane of the sample. The single-cycle THz pulses are generated using a low-temperature (LT) GaAs photoconductive antenna with 800-nm, 100-fs Ti:sapphire laser pulses and repetition rate of 80 MHz (MaiTai HP, Spectra-Physics). The generated THz pulses are collimated into

FIG. 1. (a) Schematic illustration of the THz tunneling magnetoresistance effect and the MTJ sample structure used in this work. The application of an external magnetic field, $\mathbf{H}$, leading to parallel configuration creates a large flow of electrons as compared with antiparallel configuration ($\mathbf{H} = 0$ mT). The statically measured (b) in-plane magnetization curve and (c) TMR ratio of the MTJ stack sample.

parallel beams by a micro silicon lens. The sample is positioned at the beam waist of the THz beam. The THz pulses are detected by photoconductive switching, which consists of a metallic dipole antenna on LT-grown GaAs. The photoinduced carriers of LT GaAs are driven by the THz electromagnetic field, which produces a detected current pulse. By a variable optical delay, the THz pulses generated at the emitter can be continuously delayed with respect to the gated detector, which allows us to temporally scan the electric field. The THz pulse spectrum covers the range 0.5–1.5 THz. The electric field of the incident THz pulse is linearly polarized in a horizontal plane (x axis). The weak single-cycle, sub-picosecond THz transients are directed normally at the sample (z axis). As shown in Fig. 1(a), the magnetic field $\mathbf{H}$ is applied in the plane of the sample (x axis), perpendicular to the propagation of the THz beam and parallel to the electric field polarization of the THz pulse. The THz electric field induces a time-dependent current in the sample according to its conductivity, leading to the attenuation and phase delay of the THz pulse as it propagated through the sample. We note that the THz pulse used in the present experiment is too small to influence the sample magnetization by $\mathbf{B}_{\text{THz}}$, the magnetic field components of the THz pulse. The THz beam path in the spectrometer is purged with dry nitrogen in order to minimize absorption of THz radiation by

atmospheric water vapor. All of the THz transmissions are averaged over five measurements at room temperature in an ambient atmosphere.

III. EXPERIMENTAL RESULTS AND DISCUSSION

The blue trace in Fig. 2(a) represents the typical time-domain THz transmitted signal $E(t)$ through the MTJ multistack with applied magnetic field $\mathbf{H} = 30$ mT (along the x axis), in comparison with that measured at $\mathbf{H} = 0$ mT (red circles). We note a significant attenuation of THz peak-to-peak values of the parallel orientation E_P , which is compared with E_{AP} at the antiparallel configuration, $(E_{\text{AP}} - E_P)/E_{\text{AP}} \approx 36\%$. Figure 2(b) shows the instantaneous intensities of the THz waveforms, which are calculated from the square of the measured time-resolved THz electric fields in Fig. 2(a). It can also be found that these two THz waveforms are not accompanied by any observable temporal delays. The THz optical properties are essentially determined by the low-energy carrier dynamics and are especially sensitive to the electrical conductivity. This observation strongly indicates that the resistivity of the MTJ multistack depends on the relative orientation of electron spins in the pinned and free FM layers of the structure. In the absence of an applied magnetic field, the spins are aligned in an antiparallel fashion leading to a higher

FIG. 2. (a) Time-domain THz waveforms $E(t)$ through the MTJ multistack for magnetizations aligned in parallel (blue line) and antiparallel (red circle) states. Inset: the orientation of the magnetic field is along the x axis, which rotates $\mathbf{M}_{\text{free-layer}}$ (green arrow) along the x axis. In the case of $\theta = 90^\circ$ and $\varphi = 0^\circ$, $\mathbf{M}_{\text{free-layer}}$ is aligned parallel to $\mathbf{M}_{\text{pinned-layer}}$. The zero time delay corresponds to an arbitrary starting position. (b) Instantaneous THz intensity calculated from the square of the measured electric field.

resistivity of the structure. As the spins in FM layers are co-aligned in the applied magnetic field, the sample becomes more conductive, resulting in more THz absorption.

In our experiments, the applied magnetic field (x axis) orients $\mathbf{M}_{\text{free-layer}}$ along the x axis for all cases. While it is not strong enough to align $\mathbf{M}_{\text{pinned-layer}}$ along the x axis, the orientation of $\mathbf{M}_{\text{pinned-layer}}$ can be rotated by rotating the sample. We define the azimuthal angle θ as the angle between the magnetization of the pinned layer $\mathbf{M}_{\text{pinned-layer}}$ and the y axis. In addition, the relative angle φ is defined by the angle between $\mathbf{M}_{\text{free-layer}}$ and $\mathbf{M}_{\text{pinned-layer}}$, as shown in the inset of Fig. 2(b). By rotating the azimuthal angles $\theta = 180^\circ$, 270° , and 360° (0°), we obtain the THz waveforms $E(t)$ with different relative angles $\varphi = 90^\circ$, 180° , and 270° as the blue curves shown in Figs. 3(a)–3(c), respectively. Firstly, they are compared to $E(t)$ transmitted through the MTJ multistack measured at $\mathbf{H} = 0$ mT (red circles). The relative directions of $\mathbf{M}_{\text{free-layer}}$ and $\mathbf{M}_{\text{pinned-layer}}$ are shown in the insets of Fig. 3. As shown

in Fig. 3(b), for $\theta = 270^\circ$ and $\varphi = 180^\circ$, the THz waveform is almost the same as that measured at zero magnetic field antiparallel orientation, owing to the magnetic field not changing the orientation of $\mathbf{M}_{\text{free-layer}}$. In the cases of $\theta = 180^\circ$, $\varphi = 90^\circ$ [Fig. 3(a)] and $\theta = 0^\circ$, $\varphi = 270^\circ$ [Fig. 3(c)], $\mathbf{M}_{\text{free-layer}}$ is oriented perpendicular to $\mathbf{M}_{\text{pinned-layer}}$, the transmitted THz amplitudes (blue curves) are smaller than that measured at zero magnetic field antiparallel orientation (red circles), while they are larger than that measured at $\varphi = 0^\circ$ [blue curve in Fig. 2(a)]. From our experimental results, we can see that the change of the magnetic configurations between $\mathbf{M}_{\text{free-layer}}$ and $\mathbf{M}_{\text{pinned-layer}}$ within the MTJ has a modulating effect on the propagating THz electromagnetic pulses.

As the red circles show in Figs. 3(a)–3(c), the incident THz wave with the electric field polarized parallel to $\mathbf{M}_{\text{pinned-layer}}$ (i.e., $\theta = 90^\circ$, 270°) undergoes an increased absorption. While the THz electric field polarized orthogonal to the $\mathbf{M}_{\text{pinned-layer}}$ direction (i.e., $\theta = 180^\circ$, 360°) undergoes a reduced attenuation. $I = \int E^2(t)dt$ is used to characterize the intensity of THz waveform transmitted through the sample with ultrafast time resolution. The data quantified in Fig. 3(d) are well fitted with a $\cos^2\theta$ function by the solid line, which can be attributed to AMR. AMR originates from the scattering anisotropy due to the spin-orbit interaction. The THz pulse transmission through the MTJ sample is influenced by magnetically varying the resistivity. We find that the modulation of the THz intensity by AMR is around 16%. Note that for the sufficiently thin nanometer layers used in our MTJ, we do not observe any reshaping and delay of the THz waveforms, which are observed in subwavelength-size Co particles with sample length ranging from 2 to 9 nm [28] and Co/Au bimetallic particles with sample length of 3 nm [29].

The further implications between the magnetization configuration of the MTJ sample and THz transmitted intensity are emphasized in Fig. 3(e), which are strongly dependent on the rotation angle φ . It should be noted that the dc resistance of the MTJ multistack can be expressed as $R(\varphi) = (1/2)(R_{\text{AP}} + R_{\text{P}}) - (1/2)(R_{\text{AP}} - R_{\text{P}})\cos\varphi$. Since the THz transmission is correlated to the resistance, it is reasonable to expect that the intensity of THz transmission and the dc resistance share the same $\cos\varphi$ dependence. Thus

$$I(\varphi) = \frac{1}{2}(I_{\text{AP}} + I_{\text{P}}) - \frac{1}{2}(I_{\text{AP}} - I_{\text{P}})\cos\varphi, \quad (1)$$

where I_{AP} and I_{P} are the intensity of THz waveforms for parallel and antiparallel states, respectively. Obviously, Fig. 3(e) shows that $I(\varphi)$ follows mainly the $\cos\varphi$ dependence, which again provides strong and direct evidence of TMR. Actually, we can calculate $I(\varphi)$ at different (φ) using the measured values of I_{AP} and I_{P} . Besides TMR, the experimental measured THz transmission can also be determined by several other contributions: (1) intrinsic

FIG. 3. Time-domain THz waveforms $E(t)$ through the MTJ multistack measured at $H=0$ mT (red circles) and at $H=30$ mT applied along the x axis (blue traces) for selected azimuthal angles of (a) $\theta = 180^\circ$, (b) $\theta = 270^\circ$, and (c) $\theta = 360^\circ$. (d) THz intensities $I = \int E^2(t)dt$ are calculated from the THz waveforms transmitted through the MTJ versus θ in the configuration of antiparallel state. The solid line shows a $\cos^2 \theta$ fit. (e) As $\mathbf{M}_{\text{free-layer}}$ is fixed along the x axis, φ is dependent on I . The full fitting (blue line) includes both the AMR (green dotted line) and TMR (red dashed line) contributions.

resistance of the Si substrate, (2) nonmagnetic interface resistance between the metallic layers, and (3) AMR. Indeed, the first two terms do not depend on the rotation angle φ . Therefore, we simultaneously consider both AMR and TMR contributions to the entirety of measured data. The dotted line and the dashed line show independently AMR with $\cos^2 \theta$ dependence and TMR with $\cos \varphi$ dependence, respectively. We can see that TMR plays a significant role in the observed effect under an applied magnetic field.

One could argue that the magnitudes of magnetizations of free layer and pinned layer are not exactly the same, which builds up a net magnetization of the MTJ multistack. Therefore, in such a ferromagnetic anisotropic material, the role of magnetization-induced birefringence and dichroism in the propagating THz wave is required to be examined [22]. It should be mentioned that, as the magnetization is parallel (orthogonal) to the polarization plane of a propagating THz wave, linear (circular) birefringence can be induced, corresponding to the Voigt (Faraday) configuration in the THz regime. There have been a series of demonstrations of the Faraday rotation in two-dimensional electron gases, HgTe thin films, topological insulators, and high- T_c superconductors [35–38]. Because the THz polarization can be modulated by the Faraday effect, when either spatial inversion or time reversal symmetry in the materials is breaking. We test the polarization of transmitted THz waveforms using time-domain THz polarimetry [39,40] (see Fig. S2 of Ref. [34] for details). Figure S2 shows the three-dimensional trajectory for the transmitted THz

electric fields through the MTJ multistack with and without a magnetic field. We find that the magnetic field modulates the THz transmittance, while the polarization state of the THz pulse after it passes through the MTJ sample does not show any changes, relative to the incident one. These experimental results confirm again that due to the TMR effect, the magnetically varying conductivity causes a strong spin-dependent THz transmission modulation. This means that the THz propagation properties in our metallic sample mainly depend on the electronic transports on the Fermi level.

For a quantitative analysis, we Fourier-transform the transmitted THz signals. Figures 4(a)–4(c) display the frequency-dependent THz spectra $E_\varphi(\omega/2\pi)$ transmitted through the ensemble TMR multistack on the Si substrate for the antiparallel state ($\varphi = 180^\circ$), perpendicular state ($\varphi = 90^\circ$), and parallel state ($\varphi = 0^\circ$). We evaluate the efficiency of the modulation process using the energy spectral density modulation depth [22]:

$$M_\varphi(\omega) = \frac{|E_{\text{AP}}(\omega)|^2 - |E_\varphi(\omega)|^2}{|E_{\text{AP}}(\omega)|^2}. \quad (2)$$

Figure 4(d) shows the frequency-resolved modulation depth for the MTJ sample at three given states: $\varphi = 0^\circ$, 90° , 180° . In the case of the parallel state ($\varphi = 0^\circ$), $M_{\varphi=0^\circ}(\omega/2\pi)$ is found to be around 60% over the frequency range 0.6–1.2 THz. $M_\varphi(\omega/2\pi)$ shows a very weak THz frequency dependence in all three cases. In Fig. 4(e), the symbols show the dependency of the measured THz

FIG. 4. Example amplitude spectra of the measured THz time traces under different magnetization configurations. (a) Antiparallel state ($\theta = 270^\circ$, $\varphi = 180^\circ$), (b) perpendicular state ($\theta = 0^\circ$, $\varphi = 90^\circ$), and (c) parallel state ($\theta = 90^\circ$, $\varphi = 0^\circ$). For the Fourier transformation, the THz signal in the time domain $-5 \text{ ps} < t < 10 \text{ ps}$ is used, which limits the frequency resolution to be 0.26 meV . (d) The frequency-dependent modulation depth of the THz waveforms $M_\varphi(\omega/2\pi)$ calculated using Eq. (2) for $\varphi = 0^\circ$, 90° , and 180° . (e) The angle-dependent modulation depth $M_{\omega/2\pi}(\varphi)$ for $\omega/2\pi = 0.6 \text{ THz}$ and 1.0 THz . (f) The calculated $M(\varphi, \omega/2\pi)$ as functions of frequencies and angles, using the experimental $E_P(\omega/2\pi)$ and $E_{AP}(\omega/2\pi)$ as input.

modulation depth on the relative angle φ , at two selected frequencies of $\omega/2\pi = 0.6$ and 1.0 THz . Using Eqs. (1) and (2), we calculate the φ dependence of THz waveform modulation depth $M_{\omega/2\pi}(\varphi)$, as shown by the solid lines in Fig. 4(e). Excellent agreement between the measured data and the model is observed. Therefore, the modulation of THz signals through the MTJ can be readily calculated only using the experimentally determined $E_P(\omega/2\pi)$ and $E_{AP}(\omega/2\pi)$, as illustrated in the mapping of $M(\varphi, \omega/2\pi)$ in Fig. 4(f). In this regard, our results indicate the potential for heat- and contact-free TMR readout using THz transients. The maximum modulation depth obtained in our MTJ sample (approximately 60%) is comparable to that of the THz magnetic modulator based on Fe_3O_4 nanoparticles (66%) [22]. It should be noted that the thickness of our MTJ sample is 4–5 orders of magnitude less than that of the magnetically clustered particles [22,28,29].

Analogous to electrically driven current in the TMR system, the THz pulse has been demonstrated to drive the spin-polarized current optically due to the different conductivities of majority and minority spin electrons inside the FM layer [30]. We consider that the electron wave function is delocalized across multiple layers of the MTJ structure and assume a Fermi velocity of $v_F \sim 10^6 \text{ m/s}$ and

momentum scattering time range of 20–50 fs. The average mean free path of the electrons is estimated to be $\lambda_e = v_F \times \tau \sim 20\text{--}50 \text{ nm}$, of the order of our whole MTJ structure of 77.45 nm . Chau *et al.* also mentioned that in the THz regime, the skin depth of the current is around 100 nm within bimetallic microparticles [29]. Actually, due to the Heisenberg uncertainty principle, the spatial localization of an electron within one thin metallic layer can result in a significant component of its momentum in the direction perpendicular to the plane of the layer. Therefore, the effective medium approach can be applied to describe the electron conduction in TMR multilayers. It should be also noted that our weak THz transients are not sufficient to induce any electronic or magnetic non-linearity in the sample, providing for a linear conductivity measurement.

Now we present differential measurements to yield the spin-polarized electron-tunneling-induced complex sheet conductivity $\Delta\sigma(\omega)$ using the thin-film equation $E_\varphi(\omega)/E_{AP}(\omega) = (n+1)/[n+1+Z_0\Delta\sigma(\omega)]$ [5,6], where the measured $E_{AP}(\omega)$ (highest resistive case) is taken as a reference and $E_\varphi(\omega)$ is taken as a sample signal. $n = 3.2$ is the measured effective real THz refractive index of the whole sample structure [41,42] (see Fig. S3

FIG. 5. The magnetic-field-induced change in sheet conductivity $\Delta\sigma$ of the MTJ multistack for three representative magnetization configurations, calculated from the Fourier transform of the time-domain electric field E_φ through the sample and E_{AP} for the reference. Real and imaginary parts of $\Delta\sigma$ for (a) antiparallel state ($\theta = 270^\circ$, $\varphi = 180^\circ$), (b) perpendicular state ($\theta = 0^\circ$, $\varphi = 90^\circ$), and (c) parallel state ($\theta = 90^\circ$, $\varphi = 0^\circ$).

of Ref. [34] for details) and $Z_0 = 377 \Omega$ is the impedance of free space. $\Delta\sigma(\omega) = \sigma_\varphi(\omega) - \sigma_{AP}(\omega)$ is the modulation of the frequency-sensitive sheet conductivity owing to the change of electron tunneling in the MTJ multistack.

Figure 5 shows the measured *real* component $\Delta\sigma_1(\omega/2\pi)$ (square symbols) and *imaginary* component $\Delta\sigma_2(\omega/2\pi)$ (circle symbols) within the THz spectral range of 0.5–1.5 THz. It is important to see that $\Delta\sigma_1(\omega/2\pi)$ is markedly increased, as φ changes from 180° (antiparallel state), to 90° (perpendicular state), and finally to 0° (parallel state), indicating that the electron tunneling is increased. We note that the conductivity of the MTJ multistack depends on the angle between the respective magnetization of two magnetic layers [43]:

$$\sigma_{dc} = \sigma_0(1 + P \times P^* \cos \varphi), \quad (3)$$

where σ_0 is the mean conductance and P and P^* are the effective spin polarization coefficients of the ferromagnetic-barrier coupling, including the simple spin polarization of the free and the pinned FM layers, as well as the contribution from the interface [43]. The angular dependence as shown by Eq. (3) is verified by dc measurements. However, it is not demonstrated in the THz frequency range. According to Eq. (3), $\Delta\sigma_{\varphi=90^\circ}(\omega/2\pi) = \sigma_{\varphi=90^\circ}(\omega/2\pi) - \sigma_{AP}(\omega/2\pi) = \sigma_0(\omega/2\pi)P \times P^*$, while $\Delta\sigma_{\varphi=0^\circ}(\omega/2\pi) = \sigma_{\varphi=0^\circ}(\omega/2\pi) - \sigma_{AP}(\omega/2\pi) = 2\sigma_0(\omega/2\pi)P \times P^*$. It can be noted that $2 \times \Delta\sigma_{\varphi=90^\circ}(\omega/2\pi) = \Delta\sigma_{\varphi=0^\circ}(\omega/2\pi)$. The orange line in Fig. 5(c) is the calculated result of the measured $\Delta\sigma_{\varphi=90^\circ}(\omega/2\pi)$ after multiplication by 2, which is consistent with the measured $\Delta\sigma_{\varphi=0^\circ}(\omega/2\pi)$. Our experimental findings demonstrate a good agreement with the TMR theoretical prediction over the wide THz bandwidth.

Finally, we note here that for a prospective THz modulation device [10,11], on the one hand, a fast modulation speed is required. In principle, the magnetic fields of THz pulses radiating from high-dielectric-constant lithium niobite, large-size ZnTe, and two-color laser filaments are enough to act as ultrafast magnetic pulses to switch $\mathbf{M}_{\text{free-layer}}$ within picosecond time scales [44]. If the magnetic field $\mathbf{B}_{\text{THz}}$ of THz pulse results in a reorientation of $\mathbf{M}_{\text{free-layer}}$ through the Landau-Lifschitz-Gilbert formalism, the resistance of the MTJ changes through the TMR effect during the pulse duration. We will leave the investigation of the THz transmission depending on the strength of THz input pulse to future work.

On the other hand, we believe that the MTJ-based THz modulation demonstrated here can be integrated with the concept of spintronic THz emitters (such as W/FM/Pt [45] or FM/Ag/Bi [46,47] heterostructures). The noncollinear configuration of magnetizations in the synthetic spintronic structures will be useful for further designs of THz emitters with tunable amplitudes and polarizations [48,49].

IV. CONCLUSION

In summary, we demonstrate THz magnetic modulation using the THz TMR effect, which is governed by spin-dependent electron tunneling on an ultrafast (sub-) picosecond time scale. The MTJ-based THz modulation not only combines a high modulation depth and low magnetic field requirements, but also shows flexibility given by the tunability of THz conductivity in response to the relative magnetization orientations. We believe that the modulation amplitude can be further improved by optimizing structures and parameters of the FM material and insulating barrier. As the magnetization can be switched (or quenched) by ultrashort THz or laser pulses, it should be possible to achieve a rather high-speed modulation device with operating frequency up to THz. Moreover, the MTJ can be further integrated with other metamaterials and spintronic THz emitters, which will be useful for further designs of unique THz functional devices.

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