Terahertz Generation via Picosecond Spin-To-Charge Conversion in IrMn₃/NiFe Heterojunction

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Experimental investigations of ultrafast electro-optical properties in magnetic materials manifest their great potential for emerging spintronic optoelectronic devices. Here, using time-resolved terahertz emission spectroscopy, we construct a spintronic terahertz emitter consisting of an IrMn₃/NiFe heterojunction. A femtosecond spin current pulse is generated in the thin film of the NiFe layer when it absorbs a femtosecond laser pulse, and then the spin current is converted into a transient charge current by the metallic IrMn₃ layer on picosecond timescales. We timely record the terahertz emission associated with this ultrafast conversion process by means of electro-optic sampling. Besides, the spin-to-charge conversion efficiency of the IrMn₃/NiFe heterojunction is determined via quantitative analysis of the spin torque ferromagnetic resonance results. We have both optically verified and electrically studied the spin-to-charge conversion of the IrMn₃/NiFe heterojunction. Our results enlarge the material choice range of spintronic terahertz emitters, which may promote further investigations of ultrafast spin-to-charge conversion in different heterojunction materials.

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I. INTRODUCTION

The central principle of spintronics is the manipulation of spin currents and spin orders in various categories of materials [1,2]. One significant and key step is efficient spin current generation and measurement, over a very long period of time. The platforms to realize spin-electricity conversion are limited to nonmagnetic materials or interfaces, including heavy metal [3,4], topological insulator [5–7], Rashba interface [8,9], and two-dimensional electron gas [10–12]. Recently, some groundbreaking works have demonstrated that antiferromagnetic materials can also act as efficient generators, detectors, and transmitters of spin current [13–17], which makes these antiferromagnetic materials good candidates for spin current generators and detectors in spintronic applications. However, the timescales in these pioneering studies are limited to

nanoseconds, which cannot support applications in the terahertz field. To tackle this issue, the approach of applying a femtosecond laser pulse in a specifically designed magnetic heterostructure [18] is proposed, which reduces the timescale of spin-electricity conversion to the subpicosecond range, enabling the engineering of terahertz spintronic devices. Therefore, this holds out the promise of spintronic terahertz emitters. In this respect, emerging terahertz devices comprised of a variety of nonmagnetic materials or interfaces, such as heavy metal [19–24], topological insulator [25–28], and Ag/Bi Rashba interface [29,30], have been validated. Apart from that, by using the aforementioned spin-to-charge conversion of antiferromagnetic materials, Chen *et al.* proposed a compensated magnetic heterostructure consisting of ferrimagnet and antiferromagnet to operate as a terahertz emitter [31]. Meanwhile, Zhou *et al.* observed terahertz emission in a non-collinear antiferromagnetic Mn₃Sn heterostructure [32], which extends the material selection for spintronic terahertz emitters. Nevertheless, neither detailed experimental analysis nor quantitative calculation is explicitly presented

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in those studies, let alone a detailed investigation of terahertz properties. Therefore, a combination of optical verification and electrical calculation of a spintronic terahertz emitter comprised of an antiferromagnet/ferromagnet heterojunction may provide a more complete understanding. The terahertz measurements on an ultrafast timescale and the calculation of quasistatic regime enable an in-depth insight to clarify the inside properties.

II. EXPERIMENTAL DETAILS

A. Device structure

In this work, we construct a spintronic terahertz emitter based on picosecond spin-to-charge conversion in an IrMn₃/NiFe heterojunction. The detailed device configuration of our scheme is depicted in Fig. 1(a), the complete material stack of which is Ta(1)/MgO(2)/NiFe(5)/IrMn₃(6)/Ta(4), with the values in parentheses being nominal thickness in nanometers. The MgO and top Ta serve as the capping layer to prevent oxidation, and the bottom Ta is the adhesion layer,

FIG. 1. (a) Schematic illustration of ultrafast spin injection and conversion in IrMn₃/NiFe heterojunction. θ is the polarization angle of the external magnetic field with respect to the y axis. (b) The magnetization moment M of IrMn₃/NiFe heterojunction as a function of magnetic field H_x , where θ is set as 0° .

which can improve greatly the growth quality of materials. The IrMn₃/NiFe heterojunction for terahertz emission and spin-torque ferromagnetic resonance (STFMR) measurements is grown using a multisource high-vacuum magnetron sputtering system with a base pressure of 1×10^{-6} Pa. Therefore, the lattice structure of the heterojunction is polycrystalline. The substrates are selected to be glass. For comparison, materials of pure NiFe on a glass substrate with the same capping layer are also prepared. During the electrical measurement of device fabrication, photolithography and Ar-ion milling are employed to pattern the multilayers into micrometer-scale Hall bars with a size of $65 \times 20 \mu\text{m}^2$. Next, Ti(20)/Au(100) electrical contact pads are fabricated by photolithography, electron beam evaporation, and a lift-off process. Figure 1(b) shows the MH loops of the IrMn₃/NiFe sample measured by a superconducting quantum interference device magnetometer. The direction of the magnetic field is set as the long axis of the material stack, which shows that the saturated magnetic field of NiFe is about 150 Oe. In order to fully saturate the magnetization of the ferromagnetic layer, we apply a bias magnetic field with constant magnitude (1000 Oe) in the sample plane (xy direction). Here, θ represents the azimuthal angle of the bias magnetic field with respect to the y axis, which enables us to manipulate the spin polarization of injected electrons.

B. Terahertz measurement setup

For terahertz emission measurements, the detailed experimental setup is shown in Fig. 2. The incident femtosecond laser pulse is derived from a 1-kHz Ti:sapphire regenerative amplifier, where the central wavelength of the laser beam is 800 nm, the pulse width of the femtosecond

FIG. 2. Sketch of terahertz emission experimental measurement setup.

laser is 150 fs, and the pump power is 0–150 mW. The laser beam is split into two for the pump and probe, the pump laser beam being used to excite the sample for terahertz generation. The generated terahertz radiation is collected by a parabolic mirror and then focused onto a 1-mm-thick ZnTe crystal. The Pockel effect induces a birefringence generation when the terahertz radiation is incident on the crystal. Therefore, a polarization rotation of the subsequent probe laser beam can be analyzed by a balanced photodetector system. The diameter of the incident laser beam is about 4.5 mm. In order to minimize the terahertz wave absorption by water molecules in the atmosphere, all terahertz generation and detection measurements are performed in a dry environment with a humidity level of 1.5%.

III. RESULTS AND DISCUSSION

When electrons absorb energy of the femtosecond laser pulse, they are excited from the bands below the Fermi energy to the bands above, resulting in a nonequilibrium electron distribution and diffusion. Therefore, injection

of spin-polarized current into the IrMn₃ layer (in the z direction) occurs. Then, the spin current is converted into a charge current in the IrMn₃ layer. According to previous experimental results, the spin diffusion length of IrMn is usually smaller than 1 nm [14], so the injected spin current will not diffuse into the bottom Ta layer and there is no subsequent spin-to-charge conversion. The generated charge current in the IrMn₃ layer can be described by the following equation:

$$\vec{J}_C = \theta_{SH}(2e/\hbar)\vec{J}_S \times \vec{\sigma}, \quad (1)$$

where $\vec{J}_C$ and $\vec{J}_S$ are the charge current and spin current, respectively, θ_{SH} represents the spin-to-charge conversion efficiency of the IrMn₃ layer, e and $\hbar$ are the charge of electron and reduced Planck constant, respectively, and $\vec{\sigma}$ is the spin polarization. Terahertz electromagnetic radiation is observed in the transient process of spin current conversion. However, it should be clearly noted that the antiferromagnetic IrMn₃ layer appears to be polycrystalline, so the spin-to-charge conversion in IrMn₃ cannot

FIG. 3. Ultrafast spin-current-induced terahertz emission from IrMn₃/NiFe heterojunction. (a) Typical terahertz emission in the time domain emitted from IrMn₃/NiFe heterojunction and pure NiFe film. The azimuthal angle of the external magnetic field is 0° and the pump power of the incident laser is 100 mW. Inset: the corresponding Fourier transformation spectra. (b) The corresponding Fourier transforms for the terahertz emission in the time domain. (c) The terahertz emission in the time domain with various pump powers. (d) The peak-to-peak magnitude of terahertz radiation with respect to the pump power. The solid line is a fit to the data.

be linked to the antiferromagnetic order directly, which may also account for the disordered Ir in IrMn_3 .

A. Terahertz emission in heterojunction

The experimental results of terahertz emission induced by ultrafast spin current injection and conversion in the $\text{IrMn}_3/\text{NiFe}$ heterojunction are shown in Fig. 3. Here, the azimuthal angle of the external magnetic field and the pump power of the incident laser are set as 0° and 100 mW, respectively, for all measurements unless otherwise specified. Previous investigations show that the ultrafast demagnetization of NiFe and IrMn_3 also causes terahertz emission [33]. Therefore, in order to differentiate the underlying mechanism of terahertz emission in our sample, a material-dependence study is performed on $\text{IrMn}_3/\text{NiFe}$ heterojunction and pure NiFe. As illustrated in Fig. 3(a), the $\text{IrMn}_3/\text{NiFe}$ heterojunction exhibits a considerable terahertz emission on a picosecond timescale,

while no terahertz emission is observed from the pure NiFe layer. It can be seen that the femtosecond laser-induced demagnetization of pure NiFe contributes negligibly to the terahertz emission, while the heterojunction and inside spin-to-charge conversion is the prerequisite for effective terahertz generation. Figure 3(b) presents the corresponding Fourier transformation spectra of the $\text{IrMn}_3/\text{NiFe}$ heterojunction, in which the bandwidth of the measured signal is 3 THz, which is consistent with the previous investigation of a heavy metal/ferromagnet heterojunction. However, the measured bandwidth of emission is restricted to the thickness of the ZnTe crystal; a much wider bandwidth fully covering the 1–30-THz range without a gap can be achieved in the spintronic terahertz emitter [23]. More importantly, the magnitude of the terahertz signal is directly related to the intensity of conversion of electron charge, so more powerful femtosecond lasers will result in a stronger terahertz emission. The pump power dependence of the emitted terahertz emission at an azimuthal

FIG. 4. (a) Terahertz emission in the time domain from $\text{IrMn}_3/\text{NiFe}$ heterojunction for two typical azimuthal angles of external magnetic field θ . (b) The corresponding Fourier transforms for the terahertz emission in the time domain. (c) The dependence of peak-to-peak magnitude of terahertz radiation on the azimuthal angle of external magnetic field θ from 0° to 360° . The solid curve is a fit to the data of the complete period. (d) The peak-to-peak magnitude of terahertz radiation measured in $\text{IrMn}_3/\text{NiFe}$ heterojunction as a function of azimuthal angle of external magnetic field θ and time delay.

angle of 0° is shown in Figs. 3(c) and 3(d), where the intensity of emission of terahertz pulses increasing with the incident laser power is clearly observed. Figure 3(d) shows a good linear relationship between the pump power and the peak-to-peak amplitude, which reveals that a more powerful incident laser would result in a larger spin current injection. Therefore, we can effectively set the specific intensity of the terahertz signal by adjusting the power of the incident laser.

Next, in order to confirm the central role of magnetization in terahertz emission, we reorient the magnetization of the NiFe layer using an applied in-plane magnetic field. The magnetization variation allows for the tuning of the spin polarization of injected electrons. As shown in Fig. 4(a), the polarity of the emitted terahertz pulses is reversed when the direction of the magnetic field is flipped, which indicates that the terahertz emissions are directly related to the magnetization of the ferromagnet film, while the corresponding Fourier transformation spectra are almost identical. Figure 4(c) plots the azimuthal-angle-dependent peak-to-peak amplitude of the generated terahertz electric field, which can be fitted well by a sine function. This phenomenon shows good consistency with the relationship between the spin polarization of the spin

current and the magnetization of the ferromagnet layer. Moreover, the saturation magnetization of the heterojunction induces the largest terahertz emission; the terahertz emission is minimized when the magnetization of the ferromagnet layer parallel to the y axis. The modulation depth can reach as high as 100% when the angles of applied magnetic field are 0° and 90° . Figure 4(d) shows a detailed terahertz emission phase diagram for the azimuthal angle and time delay, which shows a typically periodic evolution trend. Therefore, the application of external magnetic field may enable us to manipulate the terahertz emission effectively, which shows promise for terahertz modulation.

B. The quantitative determination of charge-spin conversion efficiency

The ultrafast spin current injection and conversion in the IrMn₃/NiFe heterojunction are the primary mechanisms of terahertz emission. The spin Hall angle is the key parameter for the efficiency evaluation of charge-spin conversion. Therefore, to measure the current-induced spin orbit torque and quantitatively determine the spin Hall angle of the IrMn₃ layer, we employ the widely used STFMR technique [34]. The detailed measurement setup is shown in

FIG. 5. STFMR measurements. (a) A typical STFMR signal (symbols) of an IrMn₃/NiFe device at 6 GHz with fits (lines), where the violet and red lines represent the symmetric and antisymmetric Lorentzian components, respectively. (b) The STFMR signal of the IrMn₃/NiFe device at 6 GHz with different input powers. (c) Resonance frequency f as a function of the resonant field H_0 . The solid curve represents a fit to the Kittel formula. (d) Resonance linewidth ΔH as a function of the resonant frequency f .

the Supplemental Material [35], in which the multilayer is patterned into a microstrip with length of 65 μm and width of 20 μm . In the STFMR experiments, the rectified dc voltage V_{dc} shown in Fig. 5(a) can be decomposed as follows: $V_{\text{dc}} = V_S F_S + V_A F_A$, where F_S and F_A represent the symmetric and antisymmetric Lorentzian functions, respectively, and V_S and V_A are the amplitudes of symmetric and antisymmetric components. The external magnetic field H_{ext} is applied in the film plane. In order to maximize the rectified voltage, the angle θ is set as 45° , and the input power of microwave current is fixed at 15 dBm. By employing the established analysis method, the efficiency of spin current generation can be mathematically expressed as follows:

$$\theta_{\text{SH}} = \frac{V_S e \mu_0 M_S t d}{V_A \hbar} [1 + (4\pi M_{\text{eff}}/H_0)]^{1/2}. \quad (2)$$

Here, e and μ_0 are the electron charge and vacuum permeability, respectively, M_S and $\hbar$ are the saturated magnetization and reduced Planck constant, respectively, and t and d are the thickness of the ferromagnetic layer and IrMn₃ layer, respectively. By fitting the typical STFMR signal, the symmetric and antisymmetric component amplitudes can be quantitatively determined. Thus, the spin Hall angle is extracted to be $\theta_{\text{SH}} = 0.024$ for the IrMn₃/NiFe heterojunction, which is consistent with previous results [14,16]. In Fig. 5(b), we compare the FMR signals under different microwave powers, which reveals that rectified voltage signal increases with microwave power, and the maximum rectified dc voltage corresponds to the maximum microwave power of 15 dBm. Figure 5(c) shows the resonant frequency as a function of resonant field, which is well fitted by the Kittel formula: $f = (\gamma/2\pi) [H_0 (H_0 + 4\pi M_{\text{eff}})]^{1/2}$. This enables the extraction of the saturation magnetization ($M_S = 7.27 \times 10^5$ A/m) in the microstrip. Figure 5(d) summarizes the FMR linewidth ΔH as a function of resonant frequency, which can be fitted by the equation of linewidth: $\Delta H = \Delta H_0 + \alpha\omega/\gamma$, where the damping coefficient α is calculated to be 0.016. The detailed experimental measurements are provided in the Supplemental Material [35].

IV. CONCLUSION

In conclusion, we construct a spintronic terahertz emitter based on ultrafast spin-to-charge conversion in an IrMn₃/NiFe heterojunction. The ultrafast spin current injection and spin-to-charge conversion are comprehensively investigated using terahertz emission spectroscopy. Particularly, the charge-to-spin conversion efficiency of the IrMn₃/NiFe heterojunction is quantitatively determined using the STFMR technique. By rotating the applied magnetic field, the modulation depth of terahertz emission can reach as high as 100%, which points to excellent potential for constructing terahertz sources. The findings

reported here not only lay the groundwork for further research into ultrafast spin Hall effect of IrMn₃ material, but also pave a promising pathway for the design and construction of optospintronic devices and antiferromagnet devices. However, due to the fact that the antiferromagnetic IrMn₃ layer is polycrystalline, the spin-to-charge conversion in IrMn₃ cannot be linked to the antiferromagnetic order directly. Our observation of terahertz emission may originate from disordered Ir in the polycrystalline IrMn₃ layer. Further exploration of epitaxial samples is needed to thoroughly understand the mechanism of the conversion process, which will benefit the development of spintronic optoelectronic devices.

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