Secondary Spin Current Driven Efficient THz Spintronic Emitters

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Femtosecond laser-induced photoexcitation of ferromagnet (FM)/heavy metal (HM) heterostructures has attracted attention by emitting broadband terahertz frequencies. The phenomenon relies on the formation of an ultrafast spin current, which is primarily attributed to the direct photoexcitation of the FM layer. However, during the process, the FM layer also experiences a secondary excitation led by the hot electrons from the HM layer that travel across the FM/HM interface and transfer additional energy in the FM. Thus, the generated secondary spins enhance the total spin current formation and lead to amplified spintronic terahertz emission. These results emphasize the significance of the secondary spin current, which even exceeds the primary spin currents when FM/HM heterostructures with thicker HM are used. An analytical model is developed to provide deeper insights into the microscopic processes within the individual layers, underlining the generalized ultrafast superdiffusive spin-transport mechanism.

1. Introduction

Out-of-equilibrium excitation of spintronic materials using femtosecond laser pulses[1] has drawn immense interest in condensed matter physics by exploiting asymmetry in the lifetime and velocity of excited majority and minority spins.[2,3] Upon laser absorption, the ferromagnet (FM)/heavy metal (HM) heterostructure undergoes spin excitation, leading to spin flipping and scattering events in femtosecond timescale.[4–7] At the same time, the spins superdiffuse semi-ballistically, forming direct channels of spin transport between the FM and HM layers.[8–11] The phenomena have led to the development of a range of applications such as broadband terahertz sources,[12–16] ultrafast spin-transfer–torque driven logic devices,[17] terahertz (THz) magnetometry,[18–20] spin-resolved electron spectroscopy,[21] ballistic electron emission microscopy,[22] next-generation data-processors,[23–27] and ultrafast spin injection in semiconductors and topological insulators.[28–33] Theoretical calculations attributed to the superdiffusion predicted the scattering processes within the FM and HM to cause the flow of hot electrons both from the FM to HM and HM to FM,[8,34] leading to enhanced ultrafast demagnetization of FM.[34,35] However, the spintronic terahertz emission has so far been discussed only through the primary photoexcitation of the FM layer, assuming unidirectional carrier flow from FM to HM.[36–39] Instead, our results, in agreement with the theoretical predictions,[8,34] distinguish effects of simultaneous photoexcitation of the HM layer for inducing additional carrier flow from HM to FM, playing a crucial role in the generation of enhanced terahertz radiation.

The photoexcited carriers in the HM layer are spin-unpolarized and do not possess energy-dependent spin asymmetry. Yet, the excited hot electrons from HM experience a spin-differentiated diffusion as they travel across the FM/HM interface and deposit additional energy in the FM layer, leading to the secondary excitation of the FM.[8,9,35,40] Hence, the FM generates an additional spin-polarized current and constructively enhances the total spin current.[8,9,35,40] In this work, we quantify the effect of secondary FM excitation by investigating the terahertz radiation from the FM/HM heterostructure with varying thicknesses of the HM layer from 0 to 10 nm. By increasing the HM thickness, three concurrent events occur i) an increase in absorption of terahertz radiation within the HM layer, ii) an
Figure 1. a) Illustration of three major ultrafast demagnetization channels upon femtosecond laser pulse excitation. The green and red regions of the DOS distribution indicate the spin-up and spin-down populations, respectively. Process 1 and 2: spin-flip mechanism and FM $\rightarrow$ HM spin-transport mechanism, respectively, led by the primary photoexcitation of FM. Solid blue arrow indicate spin current formed by primary photoexcitation of FM. Process 3: (This work) spin-transport mechanism highlights both primary and secondary excitation of FM. Solid red arrow indicate spin current formed by secondary excitation. Solid green (solid purple) arrow represent transport of spin-up (spin-down) electrons from HM $\rightarrow$ FM. The solid brown arrow in the periphery indicates the coexistence of all three processes 1, 2, and 3. b,c) photoexcitation of the two heterostructures depicting the formation of THz wave with charge current vector $j_c$ shown by the vertical green arrow. White dashed curve indicates the path followed by spins while undergoing spin-to-charge conversion in the heterostructure; b) Schematic of Quartz/NiFe heterostructure and c) Schematic of Quartz/Pt/NiFe heterostructure. d,e) Measured terahertz electric field emitted from d) photoexcited Quartz/NiFe and e) photoexcited Quartz/Pt/NiFe.

increase in spin-to-charge conversion, and iii) an increase in the secondary spin current. We decouple the three events to estimate the enhancement in carrier generation in the FM layer. The theoretical model agrees well with the experimental results and unveils the microscopic processes contributing to superdiffusion. Moreover, the secondary spin currents even surpass the primary spin currents when heterostructures with higher HM thickness are used. The results thus emphasize the significance of primary and secondary excitation for ultrafast spin transport processes and establish a generalized spin transport mechanism, crucial for developing spintronics-based THz functional devices.

2. Results and Discussions

Figure 1a illustrates the spin-flip scattering and spin-transport processes\textsuperscript{[41]} for constituting the ultrafast demagnetization in the spintronic heterostructure. So far, the spin-flip scattering (process 1) and the spin transport FM $\rightarrow$ HM (process 2) have shown only the primary photoexcitation of the FM layer. However, as highlighted in process 3, the HM layer also experiences photoexcitation, due to which HM is able to excite the FM layer once again, leading to an enhanced superdiffusive spin current, $j_s$. In principle, when the spins from FM superdiffuse into the HM layer, they undergo an inverse spin Hall effect (ISHE) and generate a transient ultrafast charge current, $j_c$, where $j_c = \gamma j_s \times M/[M]$,\textsuperscript{[14]} $\gamma$ is the spin Hall angle of the HM layer, and $M$ is the magnetization vector of the FM layer. As a result, the heterostructure emits terahertz electric field according to $E(t) = \frac{\partial j_c(t)}{\partial t}$\textsuperscript{[12-14,42]} and terahertz pulse amplitude scales with the ultrafast charge current amplitude, given by, $E_{\text{peak}} \propto j_{\text{peak}} \propto j_{\text{peak}}^s$.\textsuperscript{[18,43]} Besides, within the heterostructure, the terahertz field is also produced from minor processes without involving superdiffusion of spin current across the FM/HM interface. The mechanism includes i) ultrafast change in magnetic dipoles,\textsuperscript{[20]} ii) ultrafast change in electric dipoles,\textsuperscript{[20]} and iii) back reflection of spins in FM that experience spin-to-charge conversion due to anomalous Hall effect.\textsuperscript{[44-47]} In Figure 1b,c, a comparison of the terahertz generation is performed between two spintronic heterostructures, Pt(0 nm)/Ni$_{80}$Fe$_{20}$(3 nm) [PT-0] and Pt(2 nm)/Ni$_{80}$Fe$_{20}$(3 nm) [PT-2]. From this point, Platinum is written as Pt, and Permalloy (Ni$_{80}$Fe$_{20}$) is written as NiFe. Refer to Sections S1, S4, S5, and S6 in the Supporting Information, for setup and experimental methods. The sample without heavy metal, [PT-0], is chosen to inhibit the superdiffusive spin-transport and represents the THz generation arising only due to the minor processes. On the other hand, [PT-2] exhibits superdiffusive spin transport along with minor processes. The resulting terahertz electric field emission is shown in Figure 1d,e, where the terahertz pulse amplitude is observed to be dominated by superdiffusion (Figure 1e) and an order of magnitude higher than the counterpart in Figure 1d. As such, the terahertz pulse amplitude can be essentially considered to provide a measure of superdiffusive spin current having a much lower contribution from the minor processes. The contributions from the minor processes are separately accounted for in the theoretical analysis.
Figure 2. a) Illustration of the superdiffusion process due to the photoexcitation of both FM and HM layers. The spatial profile indicates the decay of spin polarization in the HM layer. $z_{1/e}$ marks the depth in the HM layer where the spin polarization decreases to $1/e$ of its strength at $z = 0$. b) Experimental observation of terahertz transmission decay as observed in [PT-d] where $d = 0, 1, 2, 4, 6, 8, 10$ nm. $\tau_{trans} = 0.268$ nm$^{-1}$. c) The peak-to-peak amplitude of terahertz emission from all the samples is shown by solid green triangles. The dashed line divides the region, indicating the exponential decrease in terahertz transmission amplitude for $d > 4$ nm. A solid red line indicates an exponential fit for emission in [PT-d] when $d > 4$ nm; with $\tau_{emit} = 0.136$ nm$^{-1}$. d) The construction of spatial profile describes the decay of polarized spins in the HM layer through multiple spin reflections at the surface with the nth spin decay term given by $S_n(z)$. e) The formalism suggests that $z_{1/e}$ saturates at different depths of the HM layer ($\lambda_{sd}$) with varying $\lambda_{sd}$. In our case of Pt, $\lambda_{sd} = 1.3$ nm yields $\lambda_{sd} = 4$ nm.

We consider the net spin current generated in heterostructure to consist of two components represented by $j_1$ and $j_2$. Here, the primary spin current, $j_1$, arises from the direct photoexcitation of the FM layer and secondary spin current, $j_2$, arise due to HM-led photoexcitation of the FM layer (Figure 2a). To decouple the two components experimentally, we used a set of samples, given as Quartz(1 mm)/Pt($d$ nm)/NiFe(3 nm) with $d$ varying from 0 to 10 nm, and is referred to as [PT-d]. With the increase in HM layer thicknesses, three processes occur: i) increase in spin-to-charge conversion, ii) increase in spin diffusion length, and iii) enhancement of $j_2$. The transmissions from all the samples are recorded to estimate the attenuation of terahertz wave due to absorption in the HM layer. A separate ZnFe-based time-domain terahertz spectrometer was used to perform the transmission measurement, with spintronic emitters at the terahertz focus point (refer to Section S1, in the Supporting Information, for experimental details). In Figure 2b, we observe an exponential decrease in terahertz transmission amplitude for increasing HM layer thicknesses with terahertz transmission decay rate, $\tau_{trans} = 0.268$ nm$^{-1}$. As a control experiment, the transmission was also recorded in the photoexcited state of the heterostructure, where an additional laser beam ($\lambda = 800$ nm, 35 fs, 1 kHz repetition rate) was used to photo-illuminate the sample at 1.5 mJ cm$^{-2}$ (see inset of Figure 2b), far below the optical damage threshold. The terahertz transmission in the photoexcited state of the sample was found to be slightly higher than the non-excited scenario, where the transmission decay rate with increasing HM thickness is almost identical in both cases. Further, to estimate the increase in spin-to-charge conversion and $j_2$, the terahertz emission from all the samples were recorded as shown in Figure 2c. The emission initially increases up to samples [PT-d], with $d \geq 2$ nm in the left region but exhibits an exponential decrease beyond samples [PT-d], with $d \geq 4$ nm where the emission decay rate $\tau_{emit} = 0.136$ nm$^{-1}$. The region in the right (where $d \geq 4$ nm) is of particular interest where the HM thickness is more than the spin diffusion length of Pt, $\lambda_{sd} = 1.3$ nm.$^{[50,51]}$ Pt was chosen in particular for its consistent spin diffusion length and spin Hall angle below 10 nm.$^{[52,53]}$

A monotonic exponential decrease in samples with Pt thickness beyond 4 nm suggests a saturated spin-to-charge conversion; however, in contrast, one would expect the exponential decay to appear beyond $d \geq 2$ nm (since $\lambda_{sd} = 1.3$ nm). To understand the behavior, we develop a formalism and model the spin-to-charge conversion by estimating the spatial decay of polarized spins in the HM layer (Figure 2d). Within such nanometer-thin films of HM, the spin reflection at the surfaces becomes significant, forming the left- and right-propagation.$^{[54]}$ Thus, by applying the continuity boundary conditions of the fields at the interfaces, the propagation of the spins in HM is described using a generalized transfer matrix method (TMM). Moreover, to reproduce the emission process, we applied a modified TMM$^{[55]}$ treating HM as a THz source layer. The modified TMM yields a volume charge current, $j_z$, with a temporal and spatial dependence parallel to the sample surface. The temporal profile of $j_z$.

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is used to describe the transient THz radiation field, calculated in accordance with the experimental shape of the THz emission profile. The spatial profile thus accounts for the total $j_s \rightarrow j_c$ along the HM layer, as shown in Figure 2d. Here, the nth spin decay term $S_n(z)$ exhibits exponential decay as given by the equations below, but the overall spin dynamics profile becomes more complicated than a simple exponential decay due to both left and right direction propagation.

\[
\begin{align*}
S_1(z) &= e^{-\frac{z}{\lambda_{id}}} ; \quad S_2(z) &= \chi e^{-\frac{z}{\lambda_{sd}}} e^{-\frac{z}{\lambda_{et}}} \\
S_3(z) &= e^{\eta e^{-\frac{z}{\lambda_{sd}}}} e^{-\frac{z}{\lambda_{et}}} ; \quad S_4(z) &= \chi^2 e^{-\frac{z}{\lambda_{sd}}} e^{-\frac{z}{\lambda_{et}}} 
\end{align*}
\]

(1)

(2)

Here, $z$ is the axis along the thickness of the HM layer. $\chi$ is the spin reflection at the Pt/Quartz interface, $\eta$ is the spin reflection at NiFe/Pt interface, $\lambda_{id}$ is the spin diffusion length, and $d$ is the thickness of the HM layer. The spin losses due to reflections at the interface are almost negligible, and we assume $\chi = \eta = 1$, which is in agreement with the previous studies.\[54\] The direction of the arrows depict the spin decay path in $S(z)$. As given in Figure 2d, the overall spatial profile $S(z)_{overall}$ can therefore be constructed as

\[
S(z)_{overall} = (S_1 + S_2 + S_3 + \cdots) - (S_2 + S_4 + S_6 + \cdots) 
\]

(3)

\[
S(z)_{overall} = e^{\frac{z}{\lambda_{id}}} - e^{\frac{z}{\lambda_{sd}}} e^{\frac{z}{\lambda_{et}}} 
\]

(4)

We use the formalism to estimate the HM thickness required for a spin-to-charge conversion. The maximum amplitude of the spins near the FM/HM interface is calculated using Equation (4).

For example, at $z = 0$, $S(z) = S(z_0) = \frac{1-e^{-\frac{z_0}{\lambda_{id}}}}{1-e^{-\frac{z_0}{\lambda_{sd}}}}$ and $S(z)$ reaches $S(z_0)/e$ in the depth of the HM layer, given by $z = z_{1/e}$. Using Equation (4), $z_{1/e}$ can thus be calculated as

\[
S_{1/e} = \lambda_{id} \log \left( \frac{1}{2} e^{-\frac{z_{1/e}}{\lambda_{id}}} \left( -1 + e^{-\frac{z_{1/e}}{\lambda_{sd}}} \right) \\
+ e^{-\frac{z_{1/e}}{\lambda_{et}}} \sqrt{e^{-\frac{z_{1/e}}{\lambda_{sd}}} \left( 1 + 4 e^{-\frac{z_{1/e}}{\lambda_{sd}}} + e^{-\frac{z_{1/e}}{\lambda_{et}} - 2 e^{-\frac{z_{1/e}}{\lambda_{et}}}} \right)} \right) 
\]

(5)

Here, at the known spin diffusion length for Pt, $\lambda_{id} = 1.3$ nm,\[50,51\] $z_{1/e}$ is observed to saturate at Pt thickness of $\approx 4$ nm (Figure 2e). In other words, $\lambda_{sd}^{eff} \approx 4$ nm can be interpreted as the Pt thickness required for saturating spin-to-charge conversion with spin diffusion length $\lambda_{sd} = 1.3$ nm. Hence, no additional spin-to-charge conversion occurs in samples with Pt thickness beyond 4 nm. As a result, a monotonic exponential decay is observed in samples with Pt thickness beyond 4 nm, as shown in Figure 2c. Our methods are in line with recent works by Jon Gorchon et al.,\[52\] where the authors have likewise considered multiple reflections of spins and the THz field within the spintronic heterostructure to estimate the spin diffusion lengths.

Further, we simplify the visualization of THz emission as illustrated in Figure 3a. Since we account only for the ultrafast spin excitations (spin-flip and spin scattering processes) occurring in sub-picosecond timescales, the terahertz emission arising due to the $j_s \rightarrow j_c$ in the Pt layer can be assumed to interfere constructively. Here, the terahertz emission from HM/FM sample was detected from the THz side, as shown in Figure 3a. Such a scenario consists of two main phenomena i) the superdiffusive spin current experiencing ISHE in the HM layer generates the terahertz radiation in both parallel and anti-parallel directions of laser propagation (indicated as paths A and B), and ii) the emission in path B experiences a back-reflection which constructively interfere with the emission from path A. Notice, for samples [PT-d], with $d \geq 4$ nm, the relative terahertz radiation is observed to saturate at Pt thickness of $\approx 4$ nm (Figure 2e).

In other words, $\lambda_{et}$ is used to describe the transient THz radiation field, calculated in accordance with the experimental shape of the THz emission profile. The spatial profile thus accounts for the total $j_s \rightarrow j_c$ along the HM layer, as shown in Figure 2d. Here, the nth spin decay term $S_n(z)$ exhibits exponential decay as given by the equations below, but the overall spin dynamics profile becomes more complicated than a simple exponential decay due to both left and right direction propagation.
\[ E_{\text{THz}} (d) = \alpha E_{\text{excit}}^{Pt} [\lambda_{\text{sat}}] A_{\text{NiFe}}^{\text{NiFe}} [d] A_{\text{excit}}^{\text{NiFe}} [d] \]

with all possible characteristic parameters, still fails to explain the HM thickness-dependent terahertz emission profile (refer to the solid red line in Figure 3d). Therefore, a modified model, including the HM-led secondary excitation of FM, is used, as given in Equation (7).

In Equation (7), the third correction term indicates the aforementioned \( j_{s} \), with \( \beta \) as the scaling factor. The term, \( A_{\text{excit}}^{\text{NiFe}} [d] \) denotes the absorption of the excitation laser in Pt, and \( \int_{0}^{d} e^{-z} \, dz \) denotes the total amount of energy Pt diffuses into the NiFe.
represents the energy diffusion length which provides the depth inside NiFe, where the transported hot electrons from the Pt decay to $1/e$ of its energy. Using Equation (7), we achieve an excellent correspondence between the theoretical THz emission profile (solid black line in Figure 3d) and the experimental THz emission profile (black spheres in Figure 3d), where we find the best fit at $\lambda_d = 1.3 \, \text{nm}$ with $\lambda_s = 0.76 \, \text{nm}$, $\alpha = 0.65$, $\beta = 0.40$, and $\xi = 1.68$ (Figure 3c–e). In addition, when we calculate back the THz emission with only the first two terms (considering $j_s' = 0$; which is $\beta = 0$) keeping $\lambda_d = 1.3 \, \text{nm}$, the overall decay rate of the emission (dashed green line in Figure 3c) exhibits an identical decay as that of transmission (red spheres in Figure 3c). The observation is in good agreement with the relative decay behavior of the terahertz transmission and terahertz emission as a function of Pt thickness, shown in Figure 3b. Further, by decoupling the contribution of the primary spin current, $j_s$, and the secondary spin current, $j_s'$, a significant role of HM layer driven $j_s'$ is seen in Figure 3e. As such, the ratio of THz contribution from $j_s, j_s'$, which is 0.19 at Pt thickness of 1 nm, is found to increase to 1 at Pt thickness of 4 nm and further scales to 2.78 at the Pt thickness of 10 nm.

3. Conclusions

In summary, we highlight that the HM layer generates unpolarized spins upon femtosecond laser excitation and forms spin-differentiated diffusion of the hot carriers across the FM/HM interface, depositing additional energy into the FM layer. As a result, the FM undergoes a secondary spin excitation apart from the primary photoexcitation. The experimental and theoretical results indicate a prominent contribution of the secondary spin current generation in comparison to the primary spin current and provide a measure of energy transfer from HM to FM at an ultrafast time scale. As such, our study uncovers the contribution of primary and secondary spin current toward total spin current driven superdiffusion in FM/HM heterostructure, essential for developing efficient terahertz sources and ultrafast spintronics for future technologies.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Author Contributions

P.A., R.M., and R.S. conceived the project and designed the experiments. P.A. performed all the measurements and experimental analysis. Y.Y. and M.B. provided the theoretical model. H.A. and Y.F. fabricated the spintronic emitter. All the authors analyzed and discussed the results. P.A., Y.Y., R.M., and R.S. wrote the manuscript with input from all the authors. R.S. led the overall project.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

secondary excitation, secondary spin currents, spin-diffusion length, spintronic terahertz emission, superdiffusion

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