Spintronic terahertz emission from Ni/Pt bilayer grown on MgO

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Abstract. Spintronic THz emission from Ni/Pt bilayer grown on MgO is reported based on the novel THz emitter using metallic structures. The Ni metal was deposited first on a MgO substrate and capped with a thin Pt metal via electron beam deposition. The THz emission data was obtained using a standard terahertz time-domain spectroscopy setup using a Ti: sapphire laser excitation source. Initial measurements were done using 800nm excitation with 7 mW and 185 mW pump powers under upward and downward magnetic field orientations. Polarity reversal of the terahertz signal was observed upon changing the orientation of the magnetic field. Maximum amplitude was found at 0.5 THz with bandwidth up to ~6 THz. A saturation fluence of 85.04 mJ/cm² was calculated from the pump fluence-dependence plot of the THz peak-to-peak signal. The results are consistent with the spintronic THz emission due to the inverse spin-Hall effect and provide insights for future development and optimizations.

1. Introduction

Terahertz (THz) radiation, or electromagnetic radiation in general, has been known to be generated when charge carriers accelerate [1]. This has been utilized in THz emission of materials, especially in semiconductors which main mechanisms are by drift-related current and/or diffusion-related current [2,3]. To optimize these mechanisms, different methods have been employed, such as varying dopant concentrations [2], low-temperature growths [4], epitaxial layer designs [5], and quantum structures [6]. Fabrication techniques have also been implemented like the photoconductive antenna (PCA) designs which accelerate excited electrons from one electrode to another in the presence of an electrical bias [7].

Recently, a different THz mechanism was reported by Kampfrath et al., which involves the spin property of the electrons [9]. This opens up spintronics, or spin electronics, in the THz research or possibly vice-versa. The designed emitter source consists of a ferromagnetic, FM, and nonmagnetic, NM, (FM/NM) metal thin film heterostructure. This emitter utilizes the inverse spin-Hall effect (ISHE), a phenomenon that converts the spin current (coming from the FM material) into a transient transverse
charge current (into the NM metal) [8-10]. An FM/NM sample is magnetized along a direction parallel to the plane of the metal layer/substrate. When pumped with an ultrafast femtosecond laser, non-equilibrium spin-polarized electrons will be generated in the FM layer. These electrons will diffuse to the NM layer through a superdiffusive process. Due to spin-orbit coupling, spin-up and spin-down electrons will be deflected at opposite directions with a mean angle (spin Hall angle), $\gamma$, which will then manifest into a transient charge current [9-11]. This becomes the source of THz radiation [8].

Spintronics, on its own, is popular research due to its potential for future efficient devices (less energy loss) [12]. Aside from this, the interest in spintronic THz emitters comes from their advantages in terms of contactless structures (no need of fabrication process), durability, and ease of operation (no to minimal sample alignment) [8-11]. With this, different material compositions and strategies for FM/NM systems have been developed, including defect engineering [13], NM metal investigations [14], and FM/NM heterostructure [15]. One of the common designs is the FM/Pt bilayer structure. Pt as the NM metal delivered higher THz amplitude due to its larger $\gamma$ compared to other metals [9,14]. Intensive research has already been done for Co/Pt, and Fe/Pt bilayer structures and only few can be found for Ni/Pt structures [10,11,15,16]. In this work, spintronic THz emission from Ni/Pt as the FM/NM structure is reported as a proof-of-concept demonstration. The Ni/Pt bilayer was deposited on MgO using the electron beam deposition facility of the Condensed Matter Physics Laboratory (CMPL) of the University of the Philippines – Diliman. This work is in collaboration with the Research Center for Development of Far-Infrared Region in the University of Fukui.

2. Experimental details
A 0.5mm thick MgO substrate was prepared and cleaned successively in trichloroethylene, acetone, and methanol using an ultrasonicator. The sample was then rinsed with deionized water. Using an electron beam deposition facility, nickel was deposited on the MgO substrate at a deposition rate of 0.5 Å/s. It was then capped with platinum at a deposition rate of 0.3 Å/s. The pellets used have a purity of 4N and was obtained from Kurt J. Lesker. Pre-calibrations were done using a quartz crystal. The estimated thickness of nickel and platinum are 15 nm and 6 nm, respectively.

A standard THz-TDS setup in transmission geometry was employed in obtaining the THz emission of the Ni/Pt on the MgO sample. The setup is equipped with a mode-locked Ti: sapphire femtosecond (fs) laser source, emitting 100 fs pulses at a central wavelength of 800nm and repetition rate of ~80 MHz. The laser beam is split into a pump and probe beam by a polarizing beam splitter. The pump beam (s-polarized) is mechanically chopped (set to 2 kHz) and sharply focused on the sample with an effective spot diameter of 1.53µm. The sample (mounted perpendicular to the pump beam) is oriented such that it is pumped on the metal side rather than the MgO substrate. This is to prevent any reflections coming from the MgO-air interface, where the metal layers also serve as an anti-reflection for THz [10]. A magnetic field, B, of 20 mT, is also applied perpendicular to the pump beam. The flat side of the hyper-hemispherical Si lens was attached directly at the back of the sample (MgO side) to collimate the THz emission. This emission was detected by an LT-GaAs dipole PCA with a 3.4µm gap, gated by the probe beam. Initial runs were done using 7 mW and 185 mW average power for different magnetic field directions (up and down). Excitation power-dependent THz-TDS measurements was then performed by varying pump power at constant magnetic field direction.

3. Results and discussion
Figure 1a and 1b shows the resulting THz-TDS spectra of the Ni/Pt sample on MgO taken at 7 mW and 185 mW pump power excitations, respectively. It can be seen from both graphs that the THz signal waveform inverted as the magnetic field is reversed. This is consistent with the THz emission process relating to spin current due to the spin-orbit interaction which is responsible for ISHE [8,17].

To discuss the phenomenon thoroughly, introducing an ultrafast laser in a magnetized FM/NM layer will cause a strongly spin-polarized current. The net current from the FM layer to the NM layer (axis is perpendicular to the plane of the film) is caused by the difference in the transport properties of the two layers [9]. Additionally, since there is an abundance of the spin-up electrons (majority carriers) as well as higher band velocity and lifetime as compared to spin-down electrons (minority carriers) in the FM layer, the resulting net current is a strongly spin-polarized current [10,18]. As the electrons diffuse in the NM layer through a superdiffusive process [18], spin-up and spin-down electrons are deflected at opposite directions due to spin-orbit coupling [17]. This will convert the spin current, $\vec{J}_s$, into a perpendicular charge current, $\vec{J}_c$, according to ISHE,

$$\vec{J}_c = \gamma \vec{J}_s \times \vec{\sigma}$$

(1)

where $\gamma$ is the spin Hall angle and $\vec{\sigma}$ is the spin-polarization vector parallel to the direction of the sample magnetization [8,17]. Thus, the direction of the transverse charge current is dependent on the direction of the applied magnetic field. Changing the direction of the magnetic field to the opposite direction will change the direction of the transverse current to the opposite side. This results in the reversal of the polarity of the THz signal.
The corresponding fast Fourier transform (FFT) power spectra of the power dependence plot is shown in Figure 2a and 2b. Changing the direction of the magnetic field does not affect the characteristics of the Fourier transform. This shows that the magnetic field only affects the polarity of the THz signal. Signal-to-noise ratio (SNR) can be visually observed to improve for 185 mW which is measured to be around 34 dB. The Ni/Pt on MgO was found to have bandwidth up to ~6 THz. The wide bandwidth is due to the fast dynamics and relaxation time of electrons in the metal layer having also been observed in early studies [8-10]. The peak power of the THz signal was found at 0.5 THz. An observable decrease in the signal starting at 1 THz is attributed to the frequency response of the dipole PCA [19]. Additionally, there is also the THz absorption of MgO above 3 THz [10].

An increase in the THz signal amplitude (Figure 1) and an improvement of the SNR (Figure 2) are observed for 185 mW laser pump power as compared to 7 mW laser pump power. This shows a relationship between the THz emission of the Ni/Pt sample and the laser pump power. The THz signal was obtained for different pump laser fluence (pump power of the laser over its diffraction-limited beam spot) at a constant magnetic field (B up). Figure 3 shows the pump fluence dependence of the THz peak-to-peak amplitude. The pump fluence is observed to be proportional to the resulting THz signal. Increasing the pump fluence allows more spin-polarized electrons generated in the Ni layer, which diffuses into the Pt metal. This increases the converted transient charge current, which in return also increases the THz emission.
A nonlinear relationship of the THz signal to the fluence can also be observed, which indicates saturation. This occurs due to the limited spin accumulation in the NM material at high fluence [8,15]. There is also possible heating of the sample due to the laser, which can cause changes in conductivity and a decrease in magnetization [10, 16]. The saturation fluence, F_{sat}, can be obtained from fitting in a saturation equation,

\[ E_{THz}(F) = \frac{AF}{F + F_{sat}} \]

where E is the THz field, A is a radiated field constant, and F is the pump fluence [20]. The calculated saturation fluence is 85.04 mJ/cm^2.

4. Conclusion
Terahertz emission from Ni/Pt bilayer grown via an electron beam deposition had been shown. Polarity reversal was observed from reversing the direction of the magnetic field, which is consistent with ISHE. The bandwidth of the THz signal was observed up to ~6THz, which is suitable for data transmission applications. The wide bandwidth corresponds to the fast relaxation time of the electrons. Optimization of the thickness of the metal layers should be implemented first to increase the output signal. The peak amplitude of the signal was found at 0.5 THz. Saturation fluence was calculated, obtaining a value of 85.04 mJ/cm^2. The saturation is due to the limited spin accumulation in the Pt metal and possible heating of the sample induced by the laser. As a proof-of-concept demonstration, the results serve as a promising start for future research regarding the spintronic-THz mechanism. This may also lead to potential studies about spintronics in general.

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References

[1] Griffiths J 2008 Introduction to Electrodynamics (Pearson Benjamin Cummings)
[2] Zhang X and Auston D 1992 Journal of Applied Physics 71 326
[3] Dekorsy T, Auer H, Bakker H, Roskos H and Kurz H 1996 Phys. Rev. B 53 4005-4014
[4] Prieto E A, Viscaara S A, Somintac A, Salvador A, Estacio E, Que C, Yamamoto K and Tani M 2004 J. Opt. Soc. Am. B 31 291-295
[5] Takeuchi H, Yanagisawa J, Hasegawa T and Nakayama M 2008 Applied Physics Letters 93 081916
[6] Roskos H, Nuss M, Shah J, Leo K, Miller D, Fox A, Schmitt-Rink S and Köhler K 1992 Physical Review Letters 68 2216-2219
[7] Tani M, Herrman M and Sakai K 2002 Meas. Sci. Technol. 13 1739
[8] Kampfrath T, Battiato M, Maldonado P, Eilers G, Nötzold J, Mährlein S, Zbarsky V, Freimuth F, Mokrousov Y, Blügel S, Wolf M, Radu I, Oppeneer P and Münzenberg M 2013 Nature Nanotech. 8 256–260
[9] Seifert T, Jaiswal S, Martens U, Hannegan J, Braun L, Maldonado P, Freimuth F, Kronenberg A, Henrizi J, Radu I, Beaurepaire E, Mokrousov Y, Oppeneer P M, Jourdan M, Jakob G, Turchinovich D, Hayden L M, Wolf M, Münzenberg M, Kläui M and Kampfrath T 2016 Nature Photon 10 483–488
[10] Torosyan G, Keller S, Scheuer L, Beigang R and Papaioannou E Th 2018 Sci Rep 8, 1311
[11] Papaioannou E Th, Torosyan G, Keller S, Scheuer L, Battiato M, Mag-usara V K, L’huillier J, Tani M and Beigang R 2018 IEEE Transactions on Magnetics 54 9100205
[12] Walowski J and Münzenberg M 2016 J. Appl. Phys. 120 180901
[13] Nenno D, Scheuer L, Sokoluk D, Keller S, Torosyan G, Brodianski A, Lösch J, Battiato M, Rahmi M, Binder R, Schneider H, Beigang R and Papaioannou E Th 2019 Sci Rep 9 13348
[14] Seifert T, Tran N, Gueckstock O, Rouzegar S, Nadvornik L, Jaiswal S, Jakob G, Temnov V, Münzenberg M, Wold M, Kläui M and Kampfrath T 2018 J. Phys. D: Appl Phys. 51 364003
[15] Yang D, Liang J, Zhou C, Sun L, Zheng R, Lou S, Wu Y and Qi J 2016 Adv. Optical Mater. 4 1944-1949
[16] Huismann T, Mikhailovsky R, Costa J, Freimuth F, Paz E, Ventura J, Freitas P, Blügel S, Mokrousov Y, Rising Th, Kimel A 2016 Nature Nanotechnology 11 455-458
[17] Saitoh E, Ueda M, Miyajama H and Tatara G 2006 Applied Physics Letters 88 182509
[18] Battiato M, Carva K and Oppeneer P M 2010 Physical. Review Letters 105 027203
[19] Jepsen P, Jacobsen R and Keiding S 1996 J. Opt. Soc. Am. B 13 2424-2436
[20] Sakai K and Tani M 2005 Terahertz optoelectronics (Springer-Verlag Berlin Heidelberg)