Title

The fabrication and characterization of Spintronic Terahertz Emitter using magnetic/non-magnetic bilayer with nanometer thickness

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Abstract

Recent innovations in spintronics have introduced a new approach to THz-wave generation by exploiting ultrafast spin dynamics within magnetic/nonmagnetic heterostructures. This approach is promising as a compact and efficient terahertz wave source based on rapid advances in magnetic material fabrication and ultrafast laser technology. In this paper, nano-order controlled magnetic (Fe)/non-magnetic (Pt) bilayers are fabricated on glass substrates using magnetron sputtering, a cost-effective method for large area deposition. The thin films are characterized and their thickness dependence on spintronic terahertz emission performance is investigated. The magnetic films obtained in this study are characterized by XPS and identified as Fe3O4 rather than Fe metal; the Fe3O4 films function as the magnetic layers in STE devices and show good and stable performance over a wide range of film thickness (5-10 nm) compared to that of Fe metal films (1.5-2.5 nm). To analyze the performance, a one-dimensional model is constructed, and the analytical results are discussed.

Introduction

Research in terahertz (THz) technology has been marked by significant advances due to its potential in a wide range of applications in fields such as communications, security, and biomedical sciences [1,2]. Despite the technological progress, the generation and manipulation of THz waves continue to pose significant challenges, primarily due to the limitations of available sources that are compact, efficient, and capable of covering the broad THz spectrum.

Recent innovations in spintronics have introduced a novel approach to THz wave generation by exploiting ultrafast spin dynamics within magnetic heterostructures [3,4]. This new class of THz emitters, known as Spintronic THz emitters (STEs), utilizes the inverse spin Hall effect (ISHE) to convert spin currents generated by femtosecond pulse laser excitation of ferromagnetic layers into THz radiation. This method not only promises a compact and efficient source of THz waves, but also takes advantage of the rapid developments in magnetic materials and ultrafast laser technology. STEs are characterized by their ability to operate over a wide bandwidth, from 100 GHz to 30 THz, positioning them between conventional microwave and far-infrared technologies. They operate by using ultrafast laser pulses to excite electrons in a ferromagnetic layer, which then diffuse as a spin-polarized current into an adjacent non-magnetic layer with strong spin-orbit coupling. The ISHE in the non-magnetic layer converts this spin current into a charge current, thereby generating THz radiation [5].

In this paper, nano-order controlled magnetic/non-magnetic bilayers are fabricated on glass substrates using magnetron sputtering method, which enables large area size, and the respective thin film characterizations and respective thickness dependence of STE performance are shown. For STE performance, a one-dimensional computational model is constructed, and the analytical results are discussed.

Principle and One-Dimensional Model of Spintronic Terahertz Emitter

The principle of terahertz wave emission by irradiation of a femtosecond pulse laser onto a bilayer composed of a magnetic and a non-magnetic material with nanometer thickness is explained by Figure 1. The bilayer is subjected to a magnetic field of about 20 mT. Figure 1(b) is an enlarged view of the circled area in Figure 1 (a), and notations ①, ②, ③ correspond to steps 1), 2), 3) and equations (1), (2), and (3), respectively, as mentioned the following.

1) When a femtosecond pulse laser irradiates the bilayer of magnetic and non-magnetic material, a spin current is generated in the magnetic material. Kampfrath et al. [3] explained that the majority of the light-excited spins in the magnetic material consist of s or p electrons, and the minority of spins consist of d electrons, resulting in a difference in migration speed between them, generating a transient spin current. This explanation is based on previous studies [3] in which spin pulses were generated by optical excitation with femtosecond pulses.

2) When the dynamic spin current generated in step 1) super-diffuses through the magnetic material and reaches the interface with the non-magnetic layer, it is converted into an in-plane charge current via the inverse spin Hall effect [6], diffusing into the non-magnetic layer.

3) The transient charge current generated in step 2 corresponds to the femtosecond pulse and propagates as a terahertz wave whose frequency is the inverse of the femtosecond pulse duration.

According to the basic principle of STE mentioned above and the derivation of the simple model shown in Reference 7, we have constructed a simple one-dimensional model to treat these phenomena quantitatively, assuming that steps 1) to 3) occur in sequence and almost simultaneously, using the parameters such as the power of the laser, the absorption coefficient of the laser light (800 nm wavelength), the diffusion length of the spin current, and the absorption coefficient of terahertz light in each of the NM/FM layers, respectively, which do not appear overtly in Reference 7. In Fig. 1 (b), coordinate X is defined the coordinate in the film thickness direction of the FM layer with the interface between the NM and FM layers as the origin, and coordinate Y is defined the coordinate in the film thickness direction of the NM layer with the interface between the NM and FM layers as the origin. Both X and Y are positive values.

Consider the generation and diffusion of spins at position x in the magnetic layer (with thickness d_{FM}) and the spin current reaching the non-magnetic layer as TSC(d_{FM}):

$$TSC(d_{FM}) = \int SPG(x) \cdot DF(x)\, dx \quad (1)$$

The integration is performed within the thickness d_{FM} of the magnetic layer, where SPG(x) is the spin current generated at position x by femtosecond pulse laser irradiation and DF(x) is the fraction of the spin current at position x that can diffuse to the interface with the non-magnetic layer. Furthermore, if the spin current TSC(d_{FM}) that reaches the interface diffuses into the non-magnetic layer (with a thickness d_{NM}) and is converted into a charge current JC(y) at each position y via the inverse spin Hall effect, then

$$JC(y) = \theta_{SH} \cdot TSC(d_{FM}) \cdot DN(y) \quad (2)$$

where θ_{SH} is the conversion coefficient for converting the spin current into a charge current, corresponding to the spin Hall angle in the inverse spin Hall effect. Finally, if the charge current generated in step 3 corresponds to the femtosecond pulse laser and its conversion coefficient is η_{THz}, the intensity of the generated terahertz wave TH(d_{FM}, d_{NM}) is

$$TH(d_{FM}, d_{NM}) = \eta_{THz} \cdot \int JC(y) \cdot ABSNM(y)\, dy \quad (3)$$

Here, $ABSN(y)$ represents the fraction of the terahertz wave emitted from the outside at position y in the non-magnetic layer. The integration is performed within the thickness d_{NM} of the non-magnetic layer.

Experimental

The magnetic and non-magnetic thin films on the glass substrate (D263, borosilicate glass) used in this study were prepared by magnetron sputtering. The magnetic thin film was prepared using a 2-inch iron target (99.99% purity), and the non-magnetic thin film was prepared using a 2-inch platinum target (99.9% purity). The substrate temperature was room temperature. The background pressure before deposition was less than $6x10^{-5}$ Pa, and it was controlled to 1 Pa during deposition. The deposition rates were 0.05 nm/sec for the non-magnetic platinum layer and 0.15 nm/sec for the magnetic iron layer, with RF power of 30 W and 60 W, respectively. The substrate size was up to 4 inches in diameter and the in-plane uniformity was within ±5%. All fabricated thin films were evaluated for sheet resistance using the four-point probe method and light transmittance at a wavelength of 800 nm and were characterized by XPS measurements. As for STE devices, nanometer-thick bilayers of glass/non-magnetic/magnetic structures and glass/magnetic/non-magnetic structures were prepared. Some samples were observed by cross-sectional TEM. The STE performances were measured using terahertz time domain spectroscopy (THz-TDS) set up in our laboratory.

Experimental results, analysis and discussion

Figure 2 (a) shows the cross-sectional TEM observation of the fabricated glass/Pt/Fe structure STE device and the measured thicknesses of the Pt and Fe films. In Figure 2 (b), the sheet resistance versus the calculated –ln (T/(1-R)); Absorbance from the transmittance T and reflectance R of 800 nm wavelength light measured for the simultaneously fabricated single films. The red plots in (b) show the results of a single layer thin film sample that was simultaneously deposited by magnetron sputtering with the sample shown in Fig.2 (a). In Figure 2(b), the horizontal axis corresponds to the product of the optical absorption coefficient and the film thickness, and the known film thickness obtained from cross-sectional TEM measurements can be used to convert the horizontal axis to the film thickness and calculate the absorption coefficient of the film. As a result, the optical absorption coefficients of Pt and Fe films at 800nm wavelength light were found to be $2.40x10^6$ cm^{-1} and $2.98x10^5$ cm^{-1}, respectively. According to reference URL [14], the absorption coefficients of Pt and Fe bulk for 800 nm light are $1.27x10^6$ cm^{-1} and $1.63x10^6$ cm^{-1}, respectively. The absorption coefficient of Pt is overestimated relative to the bulk, while that of Fe is underestimated relative to the bulk. The former may be due to the renormalization of multiple reflections, while the latter may be due to oxidation of the Fe film (to be discussed later).

We observed the cross-sectional TEM on the glass/Pt/Fe structure and the glass/Fe/Pt structure, together with the THz TDS measurement results of the terahertz waves emitted when irradiated with a femtosecond pulse laser of 800 nm wavelength from the glass substrate side. The glass/Pt/Fe structure showed relatively strong THz emission, while no THz emission was observed for the glass/Fe/Pt structure. In the Fe layer of the glass/Fe/Pt, a transition layer-like region was observed near the interface between the glass and the Fe layer, which was not observed in the Fe layer of the

glass/Pt/Fe structure. This result led us to focus the study on the glass/Pt/Fe structure.

Figure 3(a) shows the Fe2p spectrum of the XPS measurements of the bilayers fabricated in this study. The Fe 2p electron spectra were almost identical for all samples except the thinnest Fe layer, and by comparison with the standard data of Fe metal, FeO, Fe_3O_4, and Fe_2O_3, the Fe film fabricated in this study was identified as Fe_3O_4. Magnetite (Fe_3O_4) is known to function as a ferri-magnetic material that can serve as a spin source to generate spin currents [9] and is half-metallic (only polarized spin electrons exist in the conduction band), and a large spin current can be expected from spin pumping [10]. The FM(Fe) layer in our devices study is identified as Fe_3O_4 and is thought to play a role in FM layer. Figure 3(b) shows the magnetization curve of a typical thin film prepared in this study and identified as Fe_3O_4, measured using the magneto-optic Kerr effect. As can be seen from the figure, the magnetization changes slowly after an abrupt change and saturates at about 10 mT. This is the reason why a magnetic field of 20 mT is applied perpendicular to the paper surface in Figure 1(a).

Furthermore, experiments were conducted to understand the dependence of STE on the thickness of Pt and Fe_3O_4 films in this study. Based on preliminary experiments, the dependence of the STE on the thickness of the Pt film was investigated with the Fe_3O_4 film thickness controlled to 6 nm (5-7 nm). The dependence of the STE on the thickness of the Fe_3O_4 film was investigated with the Pt film thickness controlled to 3 nm (2.5-3.5 nm). The measurement of STE in this study was carried out by TDS system [12].

In this case, equation (1) is expressed as

$$SP(x)=(LP)\cdot\exp(-\alpha_{Pt}\cdot d_{Pt})\cdot\exp(-\alpha_{Fe3O4}\cdot x)\cdot\eta_{sp} \qquad (4)$$

where LP is the power of the femtosecond laser, α_{Pt} and α_{Fe3O4} are the absorption coefficients of the laser light in Pt and Fe_3O_4, respectively, and η_{sp} is the conversion coefficient from laser light absorption to spin current. In eq. (1), $D(x) = \exp(-x/\lambda_{Fe3O4})$ represents the fraction of the spin current that can diffuse in Fe_3O_4, where λ_{Fe3O4} is the diffusion length of the spin current in Fe_3O_4.

Equation (2) can be expressed using the result of (1) and $D(y)=\exp(-y/\lambda_{Pt})$, where λ_{Pt} is the spin current diffusion length in Pt. As for the spin-interface reflection, a large spin-mixing conductance of the interface suppresses the reflection; it has been reported that the spin-mixing conductance is large at the Pt/Fe and Pt/Fe_3O_4 interfaces [11]. So, we ignore the interfacial reflection here and perform the rest of the analysis

Equation (3) can be expressed using the result of (2) and $ABSN(y) = \exp(-\alpha^{THz}{}_{Pt}\cdot y)\cdot\exp(-\alpha^{THz}{}_{Fe3O4}\cdot d_{Fe3O4})$, where $\alpha^{THz}{}_{Pt}$ and $\alpha^{THz}{}_{Fe3O4}$ are the absorption coefficients of terahertz light in Pt and Fe_3O_4, respectively. Therefore, the intensity of terahertz radiation emitted by the samples used in this study can be calculated using the above equations and is expressed by rewriting $T(d_{FM},d_{NM})$ in Eq.(3) as $TH(d_{Fe3O4}, d_{Pt})$.

The intensity of terahertz radiation was measured using a TDS system in our laboratory [12]. The vertical axes in (a) and (b) of Figure 4 show the intensity of terahertz radiation, but not the absolute

intensity, which is a comparative value based on the signal obtained with a 1 cm thick ZnTe (110) crystal that we own. Figures 4 (a) and (b) show the experimentally obtained thickness dependence of STE on Pt and Fe_3O_4 films, respectively, where the plotted points represent the experimental data and the solid lines represent the fitting results using the above calculations. The thickness dependence of the Pt film in Figure 4 (a) agrees well with the results presented by Torosyan et al. [8] for the Pt film in STE.

Similarly, Figure 4(b) shows the experimental results of Torosyan et al. [8] when the magnetic layer is Fe, with the fitting results of the calculations using our one-dimensional model shown as solid lines. The results show that when Fe is used as the magnetic layer, the optimal thickness for STE is around 2 nm, and the performance decreases rapidly on both sides of this thickness. In contrast, when Fe_3O_4 is used as the magnetic layer, the optimal thickness is around 6-7 nm, with the performance gradually decreasing on the thicker side. From the results of the obtained parameters used for the fitting calculations, the femtosecond laser light (800 nm wavelength) absorption coefficient of the Fe film ($1x10^6$ cm^{-1}) is larger than that of Fe_3O_4 ($3x10^5$ cm^{-1}) [13], so the probability of spin current generation per unit film thickness is large, while the spin diffusion length is 0.5 nm for the Fe film and 2 nm for Fe_3O_4. That is the reason that the performance of both STEs is considered to be at the same level.

Conclusions

Good STE with NM/FM bilayers are uniformly deposited by magnetron sputtering using platinum and iron targets on a 100 mmΦ glass substrate. The FM film obtained in this study is characterized by XPS and identified as Fe_3O_4. However, experimental results show that the Fe_3O_4 film plays the role of an FM layer in STE devices. We report for the first time that the performance of STE devices fabricated with Pt/Fe3O4 bilayers may be equal to or superior to that with Fe metal thin films. In addition, compared to Fe metal thin film, high emission intensity can be stably obtained over a wide film thickness range (5-10nm) as shown in Fig.4 (b), and the material and structure are considered suitable for the development of 2-dimensional devices that require in-plane uniformity of properties.

Using our 1D model in this study, to analyze the dependence of NM(Pt) and FM (Fe, Fe_3O_4) as a function of film thickness, we determine each parameter that characterizes the performance of STE devices. Although the electrical conductivity of the Fe_3O_4 film obtained in this study is one to two orders of magnitude smaller than the Fe film, it is comparable in STE performance due to its longer spin diffusion length and lower terahertz absorption.

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Fig. 1 Diagram showing the femtosecond pulse laser and terahertz wave radiation used in the STE measurements in this study: (a), and schematic diagram of the process from femtosecond pulse laser irradiation to the generation of terahertz wave radiation: (b)

Fig. 2 (a) is a cross-sectional TEM photograph of the glass/Pt/Fe structure, from which the thicknesses of the Pt and Fe thin films were measured. (b) is a plot of the measured absorbance vs. sheet resistance of the Pt and Fe films on the fabricated glass substrate, respectively. The red plot points in the figure are the points of the measured results of the Pt and Fe films on the glass substrates that were fabricated at the same time as the sample fabrication in (a), respectively. The horizontal axis of those points indicates the film thickness of 2.7 nm and 5.0 nm, respectively.

Fig. 3 (a) XPS measurement results of the bilayer on the glass substrate fabricated in this study. The spectra show various thickness of Fe layer of the narrow scan measurement results for Fe 2p electrons. The Fe films fabricated in this study were identified as Fe_3O_4.

(b) The magnetization curve of a typical Fe thin film prepared in this study (identified as Fe_3O_4 by XPS) is shown using the magneto-optic Kerr effect.

Fig. 4 (a) and (b) show the experimentally obtained thickness dependence of STE on Pt and Fe_3O_4 films as well as Fe films [8], respectively, where the plotted points represent the experimental data and the solid lines represent the fitting results using our 1D model .

-Authors Contributions

All authors contributed to the study. Material preparation, data collection and analysis were performed by Mikihiko Nishitani, Kohei Ejiri, Shinya Isosaki, Rouchen Dai, Shojiro Nishitani, Makoto Nakajima.
The first draft of the manuscript was written by Mikihiko Nishitani and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

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-Competing Interests

The authors declare no competing interests.

-Data availability

The datasets generated during and/or analyzed during the current study are available from the corresponding author on reasonable request.