Signature of spin-dependent Seebeck effect in dynamical spin injection of metallic bilayer structures

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Abstract

The dynamical spin injection in a ferromagnetic/paramagnetic bilayer with various paramagnetic layers has been examined by using the inverse spin Hall effect. We adapt a CoFeB film as a ferromagnetic layer, which has a large spin dependent Seebeck coefficient. The contribution of the spin pumping was evaluated from the line-width change of the ferromagnetic resonant spectra while that of the thermal spin injection was evaluated from the heat conductivity for the paramagnetic layer. We find that the spin Hall voltage does not show the systematic change with respect to the line-width change. However, the normalized spin Hall voltage is found to increase with the heat conductivity for the paramagnetic layer. These results suggest that the thermal spin injection is a major contribution for the dynamical spin injection in CoFeB/paramagnetic bilayer systems.

Spin transports in solid states are of great interests owing to the intriguing quantum phenomena with their high potential application [1, 2]. In these phenomena, a spin current, a flow of spin angular momentum, plays various important roles such as manipulation and detection of the magnetization [3, 4]. Therefore, the efficient manipulation of the spin current is a key ingredient for deepening the understanding of the related physics as well as for developing spintronic applications. In most of cases, the spin current is generated electrically in a ferromagnetic metal (FM) and is injected into a nonmagnetic metal (NM) by applying an electric field across the FM/NM interface [5–9]. The generation of the spin current is proportional to the electrical spin polarization, which is defined as \( \sigma_{\uparrow} - \sigma_{\downarrow} \) / \( \sigma_{\uparrow} + \sigma_{\downarrow} \), where \( \sigma_{\uparrow} \) and \( \sigma_{\downarrow} \) are the electrical conductivity for the up-spin electron and that for the down-spin electron, respectively. This is known as the electrical spin injection, which has high controllability and flexibility.

Instead of the electrical means, recent development of understanding spintronics enable us to use the heat for manipulation of the spin current [10, 11]. In this case, the generation efficiency of the spin current is proportional to the difference between the Seebeck coefficient for up-spin electron and that for down-spin electron [12, 13]. Owing to the spin dependence of the Seebeck coefficient, the temperature gradient across the FM/NM interface produces the injection of the spin current from the FM into the NM, namely thermal spin injection. This is a unique and fantastic technique for controlling spintronic devices. In general, the efficiency of the thermal spin injection is much smaller than the electrical one. However, it often shows significant enhancement in several FMs depending on its band structure and can exceed the efficiency of the electrical spin injection. We showed that CoFe-based alloys have large spin dependent Seebeck coefficients because of their favorable band structures, which leads to excellent thermal spin injection properties [14–16].
additional damping effect due to the paramagnetic material in contact with the ferromagnet [2, 17, 23]. However, our recent study pointed out that the thermal spin injection could be the driving force of the dynamical spin injection in a CoFeB/Pt bilayer system [24, 25]. Since the dissipation due to the magnetization damping during the ferromagnetic resonance raises the temperature of the CoFeB, the temperature gradient is produced at the CoFeB/Pt interface. This results in the efficient thermal spin injection because of the relatively large spin-dependent Seebeck coefficient for CoFeB [14–16]. The several important signatures of the thermal spin injection have been observed in the power and structure dependences in the inverse spin Hall signals. However, these characteristics were only examined in the CoFeB/Pt bilayer system [25]. The systematic study with changing the paramagnetic materials may provide useful information for seeking the major contribution on the dynamical spin injection. For this purpose, in the present study, we investigate the dynamical spin injection in the bilayer systems consisting of the CoFeB and various paramagnetic metals (PM).

We experimentally investigate the dynamical spin injection property in CoFeB/PM bilayer systems. Figure 1 shows the schematic illustration of the spin Hall device used for the present study. Here, we adapt Pt, Ta and W as the PM. The CoFeB/PM bilayer films have been prepared on a FZ-Si substrate by using an ultra-high-vacuum magnetron sputtering system with the base pressure less than $1 \times 10^{-6}$ Pa. The lateral dimension of the bilayer film patterned by the stencil mask is 200 $\mu$m in width and 600 $\mu$m in length. The PMs on the FZ-Si substrate have 10 nm thickness. The CoFeB on PM is also 10 nm thickness, where the influence of the perpendicular anisotropy is negligible. After preparing the bilayer, the Cu strip line, which is a microwave generator for the FMR, is prepared on the CoFeB/PM bilayer film. Here, the strip line and the bilayer are electrically separated by inserting a 100 nm thick SiO$_2$ film, which is formed by a magnetron sputtering. The Cu strip line, 200 $\mu$m in width, 600 $\mu$m in length and 100 nm in thickness, is deposited by thermal evaporation. By exciting the FMR of the CoFeB film, both the spin pumping and the FMR heating will occur and the spin current is injected into the PM. The injected spin current is converted to the detectable electrical voltage along the PM wire because of the inverse spin Hall effect (SHE) in the PM.

First we evaluate the magnitude of the spin pumping effect for the bilayer films from the FMR spectra based on the reflection measurement using a vector-network analyzer. Figure 2(a) shows the representative $S_{11}$ spectra for various bilayer films under the input micro-wave power of 32 $\mu$W with the magnetic field of 60 mT along the strip line. As a reference, we measured the $S_{11}$ spectra for a monolayer CoFeB film prepared by the same deposition condition. In all bilayer systems, the resonant frequency $f_R$ shows good agreements with Kittel’s equation

$$f_R = \frac{n}{2\pi} \sqrt{\left(\mu_0 H + (1 - N_e) M_s\right)\left(\mu_0 H + N_e M_s\right)}.$$  \hspace{1cm} (1)

Here, $\gamma$ is the gyromagnetic ratio given by $1.76 \times 10^{11}$ rad s$^{-1}$ T$^{-1}$, $M_s$ is the saturation magnetization with $1.5 \pm 0.05$ T and $N_e$ is the demagnetizing factor with 0.98. However, the line width of the FMR spectra for all bilayer films becomes wider than that of the monolayer CoFeB film, implying the existence of the spin pumping [2]. Since the line width at a specific magnetic field involves not only the spin pumping effect but also other extrinsic effects such as inhomogeneous broadening and eddy current, it is difficult to discuss about the magnitude of the spin pumping only from these results. Therefore, we evaluated the frequency dependence of the line width of the FMR spectra from the direct measurement of the derivative FMR curve using the microwave irradiation with AC modulation coil. Here, we use two dimensional films which simultaneously prepared by the above device. Figure 2(b) shows a representative spectra for the derivative FMR where the peak-to-peak line width is clearly identified. We performed similar experiments with changing the microwave frequency and then plot the line width $\Delta H$ as a function of the microwave frequency $f_{RF}$ as shown in figure 2(c). All systems show linear relationship between $\Delta H$ and $f_{RF}$, supporting the following equation [26].
From the slope of each plot, we can estimate the damping constant $\alpha$ precisely. The CoFeB/Pt system shows the large increase of the damping constant while the CoFeB/W sample shows the smallest increase. These indicate that the spin pumping effect is maximum in the CoFeB/Pt system and is the minimum in the CoFeB/W system.

We then perform the inverse spin Hall measurements for the three bilayer systems in order to evaluate the dynamical spin injection property. Figures 3(a)–(c) show the spectra for the CoFeB/Pt, the CoFeB/Ta and the CoFeB/W bilayer sample, respectively. Here, the static electrical voltage along the PM wire under 6 GHz microwave irradiation is measured with sweeping the magnetic field. To perform the proper comparison for samples, we adapt the normalized voltage which is defined by the detected voltage divided by the electrical resistance for the area of the bilayer under the strip line. Despite of the largest spin pumping effect in the Pt, the normalized ISHE voltage does not show the maximum change. Moreover, we have observed the largest voltage in the CoFeB/W samples, which shows the smallest spin pumping effect. These imply that the spin pumping is not major contribution for the dynamical spin injection.

It should be noted that the inverse spin Hall voltage is proportional to the conversion efficiency from the spin to the charge currents, namely spin Hall angle $\theta_{\text{SHE}}$. Therefore, for the proper evaluation of the dynamical spin injection efficiency, the output voltage for each system should be divided by the spin Hall angle for the PM. About Pt, there is a systematic experimental study of the spin Hall angle with respect to the electrical resistivity [27]. By comparing with our experimental results, our Pt with the electrical resistivity is in the intrinsic dirty regime and the spin Hall angle can be assumed to be 0.1. We also emphasize that this value is approximately the same as the averaged valued for the sputtered Pt in the literatures [19, 28]. For Ta and W, the spin Hall angle is known to depend on the crystalline structure [29, 31, 33]. From the fabrication procedure and the electrical resistivity, which has been evaluated from the monolayer film, our Ta and W seem to have the alpha-phase structure. Therefore, as the value of $\theta_{\text{SHE}}$ for our PMs, we adapt the values $-0.07$ and $-0.14$ for Ta [19, 29, 32] and W [19, 31, 33], respectively. Then, we calculate the value $\Delta V/(R\theta_{\text{SHE}})$ for the proper evaluation for the
material dependence of the dynamical spin injection. We find that the CoFeB/W system shows the largest value of the dynamical spin injection, although this bilayer showed the smallest spin pumping effect. These facts suggest that the spin pumping is not the major contribution for the dynamical spin injection.

To understand these discrepancies, we consider the contribution from the thermal spin injection. Since the RF magnetic field is constant for all measurements, we assume that the temperature change of the CoFeB due to the FMR is same for all samples. The generated heat in the CoFeB distributes into the PM layer and the strip line through the SiO$_2$ insulating layer. The driving force of the thermal spin injection is the temperature gradient in the FM in the vicinity of the interface. The injection efficiency should increase with the heat conductivity $\kappa$ for the PM under the situation that the Si substrate does not change the temperature. Although it is difficult to experimentally evaluate the heat conductivity for thin films, we believe that the comparison of the bulk heat conductivity for each PM provide the similar tendency for the thin films. In table 1, we show the heat conductivity for each PM at the bulk structure and find that W has the largest heat conductivity, supporting the large thermal spin injection efficiency for the CoFeB/W system.

We also emphasize that the generated spin current $I_{S0}$ is not equal to the spin current injected into the PM $I_{S1}$ because of the back flow of the spin current $I_{S2}$ into the spin injector, by using the frame work of the spin resistance circuit similarly to the electrical spin injection, the injection efficiency of the spin current $\eta = I_{S1}/I_{S0}$ can be calculated by the following equation $R_{SP}/(R_{SP} + R_{SG})$ with the spin resistance for the FM $R_{SP}$ and that for the PM $R_{SG}$. Here, the spin resistance is defined by $2\rho\lambda/(S(1 - P^2))$, where $\rho$ is the electrical resistivity, $\lambda$ is the spin diffusion length, $S$ is the effective cross section for the spin current and $P$ is the spin polarization. Thus, the material dependence of the thermal spin injection efficiency should reflect $\kappa\eta$. As can be seen in table 1, $\eta$ for the CoFeB/Pt and the CoFeB/W is much larger than that for the CoFeB/Ta because of the small $R_{SP}$ and owing to the large $\kappa$ of W, the thermal spin injection efficiency becomes a maximum in the CoFeB/W system. Figure 4 shows material dependence of the thermal spin injection efficiency $\kappa\eta$ together with the experimentally obtained output signal. It was clearly confirmed that the thermal spin injection model provides the consistent description.

Table 1. Characteristic values of magnetic, electric and thermal properties for CoFeB, Pt, Ta and W.

| Material | Damping $\alpha$ | $|\Delta V|/R$ ($\mu$V/Ω) | $|\Delta V|/(R\theta_{SHE})$ ($\mu$V/Ω) | $\rho$ ($\mu$$\Omega$cm) | $\lambda$ (nm) | $R_{SP}$ (Ω/nm$^2$) | $\eta$ | $\kappa$ (W mK$^{-1}$) | $\kappa\eta$ |
|----------|------------------|-----------------------------|---------------------------------|-----------------|----------------|----------------|--------|------------------|
| CoFeB    | 0.0049           | —                           | —                               | 122             | 2              | 2440           | 80     | —                |
| Pt       | 0.0086           | 0.44                        | 0.100                           | 4.43            | 63             | 1260           | 0.66   | 70               | 46.2   |
| Ta       | 0.0066           | 0.19                        | -0.140                          | 1.36            | 140            | 2400           | 0.50   | 50               | 25.2   |
| W        | 0.0065           | 0.76                        | -0.070                          | 10.9            | 88             | 1760           | 0.48   | 180              | 104.6  |

Figure 4. Material dependence of thermal spin injection efficiency ($\kappa\eta$) together with the experimentally obtained normalized inverse spin Hall voltage $|\Delta V|/(R\theta_{SHE})$. The inset shows the schematic illustration of the back flow of the spin current under the thermal spin injection.
for the paramagnetic material dependence of the normalized inverse spin Hall signal $|\Delta V|/(R\theta_{\text{SHE}})$ under the dynamical spin injection.

For another signature of the thermal spin injection, we have measured the input RF power dependences of the inverse spin Hall signal. Figure 5 shows the output voltage change $\Delta V$ as a function of the input power for all bilayer samples. $\Delta V$ increases linearly below the input power of 100 mW, but it starts to deviate above the input power of 100 mW. This kind of saturation tendency is the signature of the thermal spin injection because the spin-injection efficiency is proportional to the longitudinal component of the magnetization, since the spin pumping is proportional to the transverse component of the magnetization, the power dependence should show the opposite tendency. Thus, the power dependence of the output voltage also supports the thermal spin injection mechanism for the dynamical spin injection.

In summary, the mechanism of the dynamical spin injection in the ferromagnetic/paramagnetic bilayer systems has been examined experimentally with changing the paramagnetic materials. Although the enhancement of the line width due to the spin pumping have been observed in all bilayer systems, it was difficult to confirm the correlation between the spin pumping and inverse spin Hall signal. On the other hand, by taking into account the thermal spin injection, we were able to obtain the consistent description about the material dependence of the inverse spin Hall signal. The obtained results strongly suggest that the thermal spin injection provides a significant contribution in the dynamical spin injection.

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