Spin Seebeck coefficient enhancement by using Ta$_{50}$W$_{50}$ alloy and YIG/Ru interface

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Abstract
This paper reports that Ta$_{50}$W$_{50}$ alloy has a higher spin Hall angle than that of $\beta$-W by measuring the spin Seebeck coefficients for Fe$_5$Y$_3$O$_{12}$ (YIG)/Ru/Ta$_{50}$W$_{50}$ and YIG/Ru/W systems. The spin Seebeck coefficient increased by inserting Ru, and the YIG/Ru/Ta$_{50}$W$_{50}$ system achieved 2.4 times the magnitude of spin Seebeck coefficient of the conventional YIG/Pt system although Ru and Ta$_{50}$W$_{50}$ have spin Hall angles with opposite signs. Ru seems to recover the spin mixing conductance that is decreased by Ta–W oxidation at its interface with YIG. To enhance the spin Seebeck coefficient, materials having a high spin Hall angle can be combined with materials having a high spin mixing conductance.

Keywords: spin Seebeck effect, spin Hall angle, spin mixing conductance

(Some figures may appear in colour only in the online journal)

1. Introduction

The spin Seebeck effect in a magnetic material is the spin accumulation caused by a temperature gradient. In a nonmagnetic metal adjacent to the magnetic material, the spin currents converge to the charge current through the inverse spin Hall effect [1]. These mechanisms have been studied extensively and the power generation possible with such devices has increased [2–8]. Since the voltage is perpendicular to the temperature gradient, power can be produced by using a uniform film, which could expand the range of applications for spin Seebeck devices. However, the generated power is too small a practical use. The spin Seebeck power is determined by the charge current generated in the nonmagnetic metal, which is calculated as below [9];

$$ j_c = -\theta_H \frac{2e}{h} j_s \times \sigma, $$

where $j_c$ is the charge current density, $(h/2e) \times j_s$ is the spin current density, $\sigma$ is the unit vector for spin polarisation and $\theta_H$ is the spin Hall angle of the nonmagnetic metal. In this paper, we focus on the spin Hall angle, which is an important parameter in determining the power generated by the spin Seebeck effect.

2. Approaches

The spin Hall angles $\theta_H$ of typical nonmagnetic metals such as Pt [10–20], $\beta$-Ta [13, 21] and $\beta$-W [22, 23] have been studied extensively. Although the magnitudes of the spin Hall angles of $\beta$-Ta and $\beta$-W are generally larger than that of Pt, the spin Seebeck voltages of $\beta$-Ta and $\beta$-W are smaller than that of Pt [24, 25]. To understand the disagreement between the spin Hall angle and the spin Seebeck voltage in a material, we attended to whether the magnetic material is a metal or an oxide. In this study, we took rough averages to understand the difference between the magnetic metals and oxide, and the reported spin Hall angle values were estimated from various sample structures and measurement systems. Figure 1 shows the average value of the spin Hall angle estimated simply from the values...
listed in table 1. For each structure of magnetic metal and nonmagnetic metal (blue data), $\beta$-Ta and $\beta$-W have a larger $\theta_H$ than that of Pt. In contrast, for each structure of magnetic oxide and nonmagnetic metal (red data), $\beta$-Ta has a smaller $\theta_H$ than that of Pt and the difference in $\theta_H$ between $\beta$-W and Pt is small. Even if we consider the effects of varying fabrication conditions and measurement methods on the $\theta_H$ distribution, it is clear that the spin Hall angle $\theta_H$ depends on the nonmagnetic material and on the pairing of magnetic and nonmagnetic materials.

According to the Ellingham diagrams, Ta and W are easily oxidised as compared to Fe, which is the main element of magnetic oxides, e.g. Fe$_5$Y$_3$O$_{12}$ (YIG) and Fe$_3$O$_4$, whereas Pt oxidises with more difficulty compared with Fe [30, 31]. Therefore, we hypothesise that oxygen moves slightly from YIG to Ta and W at the interface. In that case, the spin mixing conductance at the interface between a magnetic oxide and a nonmagnetic metal will be reduced. In this study, to obtain both a large spin Hall angle $\theta_H$ as that of $\beta$-W and high oxidation resistance as that of Pt, two approaches were utilised. First, we used a nonmagnetic metal Ta$_{50}$W$_{50}$ alloy, which has a higher oxidation resistance than that of Ta or W alone [32]. Second, we inserted a thin layer of Ru between the YIG and nonmagnetic materials because Ru has high oxidation resistance. Although the sign of the spin Hall angle $\theta_H$ of Ru is opposite to those of Ta and W, the magnitude of $\theta_H$ for Ru is very small [33]. The contribution of the Ru layer to the spin Hall angle can be restrained using a very thin sheet.

### 3. Experimental method

Figure 2 shows a schematic of the sample structure and the spin Seebeck measurement system. The sample structures prepared for the first test are YIG/Pt (5 nm), YIG/Ta (5 nm), YIG/W (5 nm) and YIG/Ta$_{50}$W$_{50}$ (5 nm) and those for the second test are YIG/Ru (0.5 nm)/Ta (4.5 nm), YIG/Ru (0.5 nm)/W (4.5 nm) and YIG/Ru (0.5 nm)/Ta$_{50}$W$_{50}$ (4.5 nm), where YIG is the 1 mm-thick sintered bulk. The details of sample preparation are reported in a previous paper [34]. The nonmagnetic layer shows resistivity of 77 $\mu\Omega$ cm, 1000 $\mu\Omega$ cm, 50 $\mu\Omega$ cm and 500 $\mu\Omega$ cm for Pt, Ta, W and Ta$_{50}$W$_{50}$, respectively. The resistivity of Ta, W and Ta$_{50}$W$_{50}$ are high enough to consider the materials as $\beta$-phase tetragonal crystalline structures.

A temperature gradient was applied with a pair of Peltier modules, and we monitored the temperature difference $\Delta T$ between the top and bottom of the sample. The temperature was monitored at the Peltier module surface within 1 mm from the sample surface; however, the monitored $\Delta T$ could be slightly larger than the actual temperature difference between the top and bottom of sample. Once $\Delta T$ was stabilised at 13 K or 18 K, a magnetic field was swept from $-3$ kOe to $+3$ kOe. The voltage generated was detected by 2 probes separated by 32 mm.

### 4. Experimental results and discussion

Before measuring the spin Seebeck voltage, we analysed the nonmagnetic metal Ta using x-ray photoelectron spectroscopy (XPS) to confirm oxidation at the interface. The sample structure for XPS was YIG (1 mm)/Ta (1 nm)/Al (2 nm). The thickness of the Ta layer was kept at 1 nm to emphasise the effect of the interface, and the top Al layer was sputtered to avoid atmospheric Ta oxidation. The XPS profiles for Al and Ta are shown in figures 3(a) and (b), respectively. The majority of the Al layer was oxidised but some metallic Al remained, which indicates that the Al layer successfully blocked the
atmospheric Ta oxidation. The Ta–O peaks in figure 3(b) indicate that Ta was oxidised at the interface with YIG.

Figure 4 shows the V–H curves for all samples when \( \Delta T \) was 15 K. The spin Seebeck voltage depends on the sign of the spin Hall angle. We found that the Ta 50W50 alloy has a spin Hall angle with the same sign as Ta and W independently. Since the Ru layer is sufficiently thin, samples with Ru show the same before measuring the spin Seebeck voltage as those without Ru. We estimated the spin Seebeck coefficient \( |S| \) using the expression \( \frac{t \Delta V}{l \Delta T} \), where \( l \) is the probe distance of 32 mm and \( t \) is the YIG thickness of 1 mm.

Figure 5 plots the absolute values of the spin \( |S| \) for all the studied samples. The YIG thickness that contributes to the spin Seebeck effect is about 100 nm. However, because our \( |S| \) was obtained by multiplying with the YIG total thickness of 1 mm, \( |S| \) is four orders of magnitude lower than its potential value. In figure 5, YIG/Ta has a smaller \( |S| \) than that of YIG/Pt, and YIG/W has mostly the same \( |S| \) as that of YIG/Pt. These results agree well with results reported in the literature [24, 25]. However, YIG/Ta50W50 did not exceed \( |S| \) as that of YIG/W. This indicates that the Ta50W50 alloy does not provide oxidation resistance at the YIG interface.

Ru is more resistive to oxidation than the Ta–W system, and its spin Hall angle is smaller than that of other noble metals such as Pt. As a result, Ru insertion improves the absolute spin Seebeck coefficient \( |S| \) for nonmagnetic metals of Ta, W and Ta50W50, despite the fact that Ru has an spin Hall angle with an opposite sign. YIG/Ru/Ta50W50, in particular, achieved a 2.4 times higher \( |S| \) than the standard YIG/Pt. When comparing \( |S| \) between YIG/Ru/W and YIG/Ru/Ta50W50, we can neglect the difference of spin mixing conductance because both samples have the same interface, YIG/Ru. In that case, the spin Seebeck coefficient \( |S| \) corresponds to the spin Hall angle. Therefore, the higher \( |S| \) of YIG/Ru/Ta50W50 relative to YIG/Ru/W indicates that Ta50W50 has a higher spin Hall angle than that of W. We found that Ta50W50 is the material with the highest spin Hall angle.

To estimate the effect of the spin Hall angles of Ru and Ta50W50 with opposite signs, we fabricated samples with YIG (1 mm)/Ru (\( t \) nm)/Ta50W50 (5 – \( t \) nm), where the Ru thickness \( t \) was varied from 0 to 5 nm and the Ta50W50 thickness was varied from 5 to 0 nm. The negative value of spin Seebeck coefficient \( -S \) in relation with the Ru thickness \( t \) is plotted in figure 6. \( -S \) was increased to a value more than its double by increasing the Ru thickness to 0.5 nm. However, once the Ru thickness is increased over 0.5 nm, \( -S \) starts decreasing and the sign changes at \( t = 4 \) nm, which indicates that the inverse spin Hall effect in Ru becomes dominant when the Ru layer is more than 4 nm thick. However, \( |S| \) is small when the Ru thickness is over 4 nm, the spin Seebeck voltage by the Ru layer is negligible compared to that by Ta50W50. In addition, there is possibility that the voltage is produced by the Nernst effect in the Ru layer. As \( |S| \) of the Ru 5 nm is sufficiently small, it was found that the Nernst effect in Ru makes a limited contribution.

Next, we estimated the ratio of improvement in \( |S| \) due to Ru insertion to gain insight into the influence of Ru insertion on nonmagnetic materials; \( \frac{|S|_{YIG/Ru/50W50}}{|S|_{YIG/Ta50W50}} = 2.2 \). We found that structures with a large amount of Ta are sensitive to the improvement due to Ru insertion. Since Ta has a lower Gibbs standard energy than that of W according to the Ellingham diagram [30, 31] 4, the recovery of spin mixing conductance is more pronounced in nonmagnetic metals with a large amount of Ta.

\[ \text{Figure 2. Schematic of the sample structure and measurement system.} \]

\[ \text{Figure 3. XPS profiles of Al (a) and Ta (b) for the YIG (1 mm)/Ta (1 nm)/Al (2 nm) structure.} \]

4 See footnote 3.
Figure 4. Generated voltage $V$ as a function of magnetic field $H$ in a temperature gradient $\Delta T = 15$ K for YIG 1 mm/Pt, Ta, W or Ta$_{50}$W$_{50}$ 5 nm (a) and YIG 1 mm/Ru 0.5 nm/Pt, Ta, W or Ta$_{50}$W$_{50}$ 4.5 nm (b), respectively.

Figure 5. Magnitude of spin Seebeck coefficient $|S|$ for YIG/nonmagnetic metals in 5 nm layers.

Figure 6. Negative value of spin Seebeck coefficient $-S$ for YIG (1 mm)/Ru ($t$ nm)/Ta$_{50}$W$_{50}$ ($5 - t$ nm).
As noted above, Ru insertion is intended to limit the oxidation of Ta, W and Ta50W50. The improvement of $|\Delta|$ caused by Ru insertion suggests that the Ru layer indeed limits the oxidation of Ta, W and Ta50W50, which improves the spin mixing conductance at the interface with YIG. In contrast, the spin Hall angle of W can be increased by intentional oxidation – [35]. We consider that optimum oxidation will improve the spin Hall angle of W but are easily oxidised. To design devices that require the interface with YIG will intercept the spin current injection and thereby decrease the spin mixing conductance.

5. Summary

We showed that YIG/Ru/Ta50W50 has a 2.4 times higher spin Seebeck coefficient than conventional YIG/Pt. By comparing the spin Seebeck coefficients for YIG/Ru/W and YIG/Ru/Ta50W50, we found that the spin Hall angle of Ta50W50 is larger than that of W. Since inserting Ru into the interface between YIG and nonmagnetic metal successfully improved the spin mixing conductance at the interface, Ru proves to be a suitable insertion material of improving the spin mixing conductance for YIG/Ta–W systems. These systems have a high spin Hall angle but are easily oxidised. To design devices that require specific spin Hall angle and spin mixing conductance, materials for the bulk and interface should be selected separately to improve the spin Seebeck coefficient.

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