Enhancement of thermal spin pumping by orbital angular momentum of rare earth iron garnet

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\textbf{ABSTRACT}

In a bilayer of ferromagnetic and non-magnetic metal, spin pumping can be generated by a thermal gradient. The spin current generation depends on the spin mixing conductance of the interface and the magnetic properties of the ferromagnetic layer. Due to its low intrinsic damping, rare earth iron garnet is often used for the ferromagnetic layer in the spin Seebeck experiment. However, it is actually a ferrimagnetic with antiferromagnetically coupled magnetic lattices and the contribution of rare earth magnetic lattice of rare earth iron garnet on thermal spin pumping is not well understood. Here we focus on the effect of magnetic properties of lanthanide and show that the orbital angular momentum of rare earth iron garnet enhances thermal spin current generation of lanthanide substituted yttrium iron garnet.

1. Introduction

The increased Ohmic losses that are associated with decreasing size of integrated circuits is expected to the breakdown of Moore’s law [1]. By employing spin degree of freedom to existing technologies, such as thermoelectric device, the manipulation of spin current alongside electric current can lead to a better efficiency and delay the breakdown\cite{2]. The generation of a pure spin current with no charge current also widely research because it is expected to have less Ohmic loss \cite{3}. At a magnetic interface, a spin current can be generated by electric field \cite{4, 5}, ferromagnetic resonance \cite{6, 7} or thermal gradient \cite{8, 9}.

One of the mechanism of the spin current generation at an interface of ferromagnet and non-magnetic metal is spin Seebeck effect \cite{10}. The magnitude of the spin current\cite{11}

\begin{equation}
J_s = \frac{\hbar g^{11}}{2\pi M_s V_c} \Delta T
\end{equation}

is proportional to temperature different $\Delta T$ across the interface, spin mixing conductance $g^{11}$ of the interface \cite{12}, gyromagnetic ratio $\gamma$ and the inverse of the saturated magnetization $M_s$ of the ferromagnetic layer. $V_c$ is the magnetic coherence volume of the magnetic layer \cite{13}. At the ferromagnet side, the spin angular momentum loss can be detected as enhancement in the magnetic damping. At the non-magnetic metal side, the spin current can be detected as an electric voltage by means of the inverse spin Hall effect \cite{5}. The direction of the detected electric current is perpendicular to both spin current direction and magnetization of the ferromagnet.

Thermal spin pumping can also occurs when the magnetic layer is antiferromagnetic, such as MnFe\textsubscript{2} \cite{14, 15} or a ferrimagnetic insulator such as Y\textsubscript{3}Fe\textsubscript{5}O\textsubscript{12} (YIG) \cite{16, 17}. Furthermore, transition near the magnetization compensation point of Gd\textsubscript{3}Fe\textsubscript{5}O\textsubscript{12} is observed as the switch of the polarization of the generated spin current \cite{18, 19}. Recently, it has been observed that spin Seebeck effect can be enhanced by substituting yttrium in ferrimagnetic YIG by rare earths \cite{20, 21}. While machine-learning has been employed for optimizing the spin Seebeck effect \cite{21}, a deeper physical understanding on the role of lanthanide on thermal spin pumping is required.

In this article, we investigate the role of lanthanide magnetic moment in the thermal spin pumping of lanthanide substituted YIG, Y\textsubscript{3}Fe\textsubscript{5}O\textsubscript{12}. Sec. 2 describes thermal spin pumping from ferrimagnet with two magnetic sublattices. Sec. 3 discuss the coupling of Fe and Lanthanide (R) magnetic lattices and its effect on $V_c$, $M_s$ and $\gamma$. While $M_s$ and $\gamma$ of rare earth iron garnet is widely studied, their combined relation to thermal spin pumping are not so well described. We analyze the effect of R angular momentum on $g^{11}$ and the resultant spin current in Sec. 4 and summarize the thermal spin pumping of Y\textsubscript{3}Fe\textsubscript{5}O\textsubscript{12}|Pt in Sec. 5.

2. Thermal spin pumping from ferrimagnet

Ref. \cite{11} derived Eq. 1 for describing spin Seebeck effect from ferrimagnet to non-magnetic metal. Thermally generated spin current arises from spin pumping from ferromagnetic layer to non-magnetic layer \cite{25}

\begin{equation}
J_{sp} = g^{11}\mathbf{m} \times \mathbf{h},
\end{equation}

and backflow spin current due to Johnson Nyquist thermal noise.

\begin{equation}
J_{bn} = M_s V_c\mathbf{m} \times \mathbf{h}.
\end{equation}

Here $\mathbf{m}$ is magnetization direction, $\mathbf{h}$ is zero-averaged magnetic field noise \cite{26}

\begin{equation}
\langle h(t)h(t') \rangle = \frac{2k_B T \alpha}{\gamma M_s V_c} \delta(t-t'),
\end{equation}

$\alpha$ is damping constant. At the magnetic interface $\alpha$ dominantly arise from the spin mixing at the interface and the averaged spin current is proportional to the temperature difference across the interface\cite{11}

\begin{equation}
J_s = \langle g^{11}\mathbf{m} \times \mathbf{m} + M_s V_c \mathbf{m} \times \mathbf{h} \rangle = \frac{\hbar g^{11}}{2\pi M_s V_c} \Delta T.
\end{equation}

While the spin pumping of Y\textsubscript{3}Fe\textsubscript{5}O\textsubscript{12} can be described by the dynamic of Fe magnetic moment alone \cite{27}, the total spin current generation of Y\textsubscript{3}Fe\textsubscript{5}O\textsubscript{12} require two magnetic sub-lattices model. Since the spin mixing conductance is shown to arise from the exchange interaction between the localized spin of ferromagnet and conduction spin of the non-magnetic metal \cite{27, 28}

\begin{equation}
g^{11} \propto J_{ex}^2 \left( S_{Fe} - |g_J - 1| J_g \right)^2,
\end{equation}

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For $x$ between spins. Itums of Fe and R at tice effect of the exchange coupling of magnetic lattices. The exchange affect spin current generations, in this article we will focus on the using two sub lattice model of Fe and R [31].

The magnetization of rare earth garnet arises from the magnetic moment of trivalent iron and lanthanide (R). As seen in Fig. 1, Fe are antiferromagnetically coupled to non-magnetic metal, which mainly depend on the non-magnetic metal when the ferromagnet is an insulator, such as rare earth iron garnets [30].

3. Coupled dynamics of rare earth iron garnet

The magnetization of rare earth garnets arises from the magnetic moments of trivalent ions of iron and lanthanide (R). As seen in Fig. 1, Fe occupies tetrahedral (d) and octahedral (a) site, while R occupy dodecahedral (c) site [24]. Since a and c sites are antiferromagnetically coupled to d-site, the total magnetization of RY$_2$Fe$_5$O$_{12}$ is

$$M_s = M_{Fe} - M_R.$$  

(7)

For $x = 1$, the magnetization of RY$_2$Fe$_5$O$_{12}$ can be determined using two sub lattice model of Fe and R [31].

While demagnetizing fields and crystallographic anisotropy may affect spin current generations, in this article we will focus on the effect of the exchange coupling of magnetic lattices. The exchange coupling can be written using interaction Hamiltonian consists of ferromagnetic coupling between Fe spins of neighboring lattice and antiferromagnetic coupling between R and Fe spins at the same lattice

$$H_{M} = \lambda \sum_\alpha [g_\alpha - 1|J_{R}^\alpha|S_{Fe}^\alpha - J_{Fe}^\alpha + S_{Fe}^{\alpha+1}].$$  

(8)

Here $S_{Fe}^\alpha = M_{Fe}/(2\gamma_0)$ and $J_{Fe}^{\alpha} = M_R/(g_\alpha g_\alpha T)$ are angular momentums of Fe and R at $n$-th site, respectively. $g_\alpha$ is Lande g-factor. $I$ and $\lambda$ are the exchange constants that depends on the distances between spins.

The Landau–Lifshitz equations of the spins under magnetic field $H$ are

$$\frac{dS_{Fe}^\alpha}{dt} = S_{Fe}^\alpha \times \left(2\gamma_0 H + I \sum_{m=\alpha+1}^{\alpha} S_{Fe}^m - |g_\alpha - 1|J_{Fe}^\alpha \right).$$  

(9)

where $\gamma_0$ is the classical gyromagnetic ratio. The coupled equations can be linearized by setting $F_\pm = F_{Fe}^\pm + iF_{R}^\pm = F_0 e^{i(\omega_0 t)}$,

$$\frac{d}{dt}\begin{bmatrix} S_{Fe}^\alpha \\ J_{Fe}^\alpha \end{bmatrix} = \mathbf{W} \begin{bmatrix} S_{Fe}^\alpha \\ J_{Fe}^\alpha \end{bmatrix},$$  

(10)

$$\mathbf{W} = \begin{bmatrix} 2\gamma_0 H - 4I S_{Fe} \sin^2 \frac{ka}{2} + |g_\alpha - 1|\lambda J_{Fe} & |g_\alpha - 1|\lambda S_{Fe} \\ \frac{|g_\alpha - 1|\lambda J_{Fe}}{g_\alpha g_\alpha T} H & |g_\alpha - 1|\lambda S_{Fe} \end{bmatrix}.$$  

The eigenvalues of the matrix is

$$\omega_{1,2} = \frac{4I S_{Fe} \cos \frac{ka}{2} + |g_\alpha - 1|\lambda J_{Fe}}{2} \pm \sqrt{1 + 4\left(\frac{S_{Fe} g_\alpha J_{Fe}}{g_\alpha g_\alpha T} H\right)(4I S_{Fe} \cos \frac{ka}{2} + |g_\alpha - 1|\lambda J_{Fe} + 4I S_{Fe} J_{Fe})}.$$  

(11)

where $\lambda' = |g_\alpha - 1|\lambda$. Here $a$ and $k$ are lattice constant and wave vector. Two limits, $k = 0 (H \neq 0)$ and $H = 0 (k \neq 0)$ are useful for analyzing the effect of rare earth magnetic sub-lattice on $V_\gamma$, $M_s$ and $\gamma$.

3.1 Magnetic moment of lanthanide sublattice

The spin wave modes in Fig. 2 can be found from eigenfrequencies of matrix in Eq. 11 for $H = 0$,

$$\omega_1(H = 0) = 4I S_{Fe} \cos \frac{ka}{2} + |g_\alpha - 1|\lambda J_{Fe}$$  

(12)

$$\omega_2(H = 0) = -|g_\alpha - 1|\lambda S_{Fe}$$  

(13)

for $IS_{Fe} \gg \lambda(S_{Fe} - J_{Fe})$. Eq. 12 indicates that the changes of the spin wave stiffness

$$D = \lim_{k \to 0} \frac{\partial^2 \omega_1}{2 \partial k^2} \approx IS_{Fe}a^2$$  

(14)

due to rare earth sublattice only depends on the change of lattice constant. The effects of spin wave on the spin current generation is widely researched [32, 33, 34] and the effect of the spin wave on thermal spin pumping is captured on magnetic coherence volume $V_\gamma$, which is directly related to the spin wave stiffness [13]

$$V_\gamma \propto D^{1/2} \propto a^3.$$  

(15)

While lattice constants of R$_2$Fe$_5$O$_{12}$ is well studied [35], in this article we will assume that the changes of $a$ due to small R substitution RY$_2$Fe$_5$O$_{12}$, which is less than 1% [36], is negligible.

Eq. 13 indicates that rare earth magnetic sub-lattice of rare earth iron garnet gives an optical branch on its spin wave dispersion [31]. The optical spin wave mode $\omega_2$ can be associated with the paramagnetic response of R to Fe molecular field. The temperature dependence of $M_R = g_\alpha J_{Fe} N$ with quantum number of total angular momentum $J$ can be determined from the statical averaged of $J_R$

$$J_{R}(T) = \left(\frac{\sum_{J_\alpha} J_\alpha^2 e^{-\omega_0(J_\alpha^2)/k_BT}}{\sum_{J_\alpha} e^{-\omega_0(J_\alpha^2)/k_BT}}\right) = \frac{J \beta J}{k_BT}.$$  

(16)

$\beta J$ is the Brillouin function. Since $\lim_{x \to 0} B_J(x) = (J + 1)x/3$, $M_R$ at a high temperature can be approximated using Curie law

$$J_{R}(T) = \frac{|g_\alpha - 1|J(J + 1)\lambda S_{Fe}}{3k_BT}.$$  

(17)
\[ \omega_1 = \omega_1(\lambda = 0) = 4S_{Fe} \sin^2(ka/2) \]

\[ \omega_2 = -\lambda S_{Fe} \]

**Figure 2**: Spin-wave spectra of two-sublattice models of \( \text{RY}_2\text{Fe}_5\text{O}_{12} \) (red lines). The spectrum of ferromagnetic spin wave \( \omega_1 \) of iron chain with spin \( S_{Fe} \) is approximately the same as the unperturbed one (black dashed line). The antiferromagnetic coupling between Fe and R generate an optical branch \( \omega_2 = -\lambda S_{Fe}, \lambda \) is exchange constant of Fe and R.

The sign switching of \( M_s = 2S_{Fe} - xg_J J_R \) happen at compensation point [24, 37]

\[ T_{comp} = \frac{x\lambda g_J S_{Fe} - 1}{6k_B} J(J + 1) \] (18)

The sign switching of \( M \) near compensation point is observed as a switching of the spin polarization of the spin current [18, 19]. Fig. 3 illustrate the relation in Eq. 18 for

\[ \lambda = \begin{cases} 
\lambda_1 = 2.78 \times 10^{-2} k_B T_C, \\
\lambda_2 = (8.2 + 0.7g_J(g_J - 1)J(J + 1)) \times 10^{-2} k_B T_C, 
\end{cases} \] (19)

obtained from linear and parabolic fitting of \( T_{comp} \) to experimental data from Ref. [24].

3.2. Gyromagnetic ratio

Gyromagnetic ratio \( \gamma \) of \( \text{RY}_2\text{Fe}_5\text{O}_{12} \) can be determined by considering \( k = 0 \) with \( H \neq 0 \) limit of Eq. 11

\[ \begin{align}
\omega_1(k = 0) &= |g_J - 1| \lambda (J_R - S_{Fe}) + \mathcal{O}(H), \\
\omega_2(k = 0) &= \gamma_0 H \frac{2S_{Fe} - g_J J_R}{S_{Fe} - J_R} + \mathcal{O}(H^2).
\end{align} \] (20, 21)

Eq. 20 is related to Kaplan–Kittel frequency of two-sublattice system [35, 38] in THz regime [39]. The resultant gyromagnetic ratio can be obtained from the lower eigenvalue (Eq. 21)

\[ \gamma = \left. \frac{\partial \omega_1(k = 0)}{\partial H} \right|_{H=0} = \gamma_0 \frac{2S_{Fe} - g_J J_R}{S_{Fe} - J_R}. \] (22)

Since orbital angular momentum is \( L = (2 - g_J)J_s \), one can see orbital angular momentum increase \( \gamma \) of the rare earth element

\[ \lim_{\lambda g_J S_{Fe}} \gamma \approx 2\gamma_0 \left( 1 + \frac{2 - g_J}{2} \lambda \frac{J_R}{S_{Fe}} \right) = 2\gamma_0 \left( 1 + \frac{L_R}{S_{Fe}} \right). \] (23)

**Figure 3**: Compensation temperature \( T_{comp} \) of \( \text{R}_2\text{Fe}_5\text{O}_{12} \) is proportional to \( \lambda g_J - 1 |J(J + 1) \). The red line corresponds to \( \lambda_1 = 2.78 \times 10^{-2} k_B T_C \), obtained from linear fitting to experiment data from Ref. [24]. The blue lines corresponds to \( \lambda_2 = (8.2 + 0.7g_J(g_J - 1)J(J + 1)) \times 10^{-2} k_B T_C \) obtained from parabolic fitting for better agreement. \( T_C \) is the Curie temperature. At the compensation temperature total magnetization \( M_s = M_{Fe} - M_R \) changes sign.

4. Enhanced spin current generation

By combining the influence of lanthanide on \( V_c, M_s, \gamma \) and \( g^{11} \), we can arrive at the following proportionality

\[ J_s \propto \frac{(S_{Fe} - g_J - 1)J_R}{S_{Fe} - J_R} \] (24)

For heavier lanthanide \( g_J > 1 \), one can see that the spin current is enhanced by orbital angular momentum

\[ \lim_{\lambda g_J S_{Fe}} J_s \propto \left( 1 + \frac{3 - 2g_J J_R}{S_{Fe}} \right) \left( 1 + \frac{L_R - S_R}{S_{Fe}} \right). \] (25)

Here, \( L_R \) and \( S_R \) are coupled to \( J_R \), which expectation value is described in Eq. 17. Fig. 4 illustrates the values of thermal spin pumping of \( \text{RY}_2\text{Fe}_5\text{O}_{12} \)/Pt compared to those of \( \text{Y}_2\text{Fe}_5\text{O}_{12} \)/Pt in room temperature \( T \sim 0.5T_C \) and shows the agreement to experiment data by Ref. [21].

The larger value of \( \text{LaY}_2\text{Fe}_5\text{O}_{12} \) compared to \( \text{LaY}_2\text{Fe}_5\text{O}_{12} \) may come from the decrease of rare earth garnet lattice constant. The decreasing value may also rise from the low heat conductance of YIG, which reduces temperature different at the interface [12]. By adding a corrected exchange constant \( \lambda \) obtained from parabolic fitting in Fig. 3, the resultant thermal spin pumping agrees with the values for heavy end of lanthanide series.
5. Conclusion

To summarize, we consider two magnetic sub-lattice model for describing the thermal spin pumping of rare earth iron garnet. The effect of lanthanide magnetic moment on the thermal spin pumping of rare earth iron garnet arise from its contribution on $M_s$, $\gamma$ and $g^{11}$. While the angular momentum of R does not affect $V_c$ and reduces $g^{11}$, it enhances the thermal spin pumping by reducing $M_s$ and increasing $\gamma$.

Magnetic moment of lanthanide is paramagnetically responsive to the molecular field of Fe. Generally, the exchange coupling constant between Fe and R can be assumed constant ($\lambda = \lambda_1$) for all lanthanide elements. A correction factor may be added from parabolic fitting in Fig. 3. The resultant thermal spin pumping agrees with the values for heavy end of lanthanide series. The maximum value of thermal spin pumping is achieved when the difference between orbital and spin angular momentum of the rare earth is maximum.

The deviation between theoretical and experimental values may arise from the dependency of crystalline and interface parameters to the lanthanide substitution, such as lattice parameter, g-factor and crystalline anisotropy. The present modeling is also applicable for other ferrimagnetic materials.

Table 1

| R    | GGG  | SGGG  | weighted averaged |
|------|------|-------|-------------------|
| Y    | 2.46±0.09 | 1.32±0.24 | 2.14±0.58        |
| La   | 2.23±0.59 | 2.36±0.10 | 2.34±0.31        |
| Ce   | 3.09±0.36 | 3.17±0.19 | 3.14±0.21        |
| Pr   | 2.89±0.18 | 3.51±0.08 | 3.32±0.32        |
| Nd   | 2.09±0.29 | 2.96±0.12 | 2.69±0.46        |
| Sm   | 3.23±0.18 | 2.71±0.10 | 2.90±0.28        |
| Eu   | 1.72±0.14 | 0.65±0.13 | 1.18±0.55        |
| Gd   | 2.70±0.15 | 0.72±0.09 | 1.43±0.99        |
| Tb   | 2.74±0.10 | 1.04±0.34 | 2.36±0.86        |
| Dy   | 3.87±0.31 | 3.17±0.19 | 3.44±0.39        |
| Ho   | 3.92±0.09 | 3.31±0.09 | 3.62±0.31        |
| Er   | 4.77±0.20 | 4.48±0.33 | 4.66±0.24        |
| Tm   | 4.54±0.12 | 3.81±0.24 | 4.31±0.39        |
| Yb   | 5.35±0.30 | 4.37±0.16 | 4.71±0.52        |
| Lu   | 3.91±0.05 | 2.27±0.13 | 3.45±0.82        |

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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