Spin Hall Effects in Metallic Antiferromagnets

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We investigate four CuAu-I-type metallic antiferromagnets for their potential as spin current detectors using spin pumping and inverse spin Hall effect. Nontrivial spin Hall effects were observed for FeMn, PdMn, and IrMn while a much higher effect was obtained for PtMn. Using thickness-dependent measurements, we determined the spin diffusion lengths of these materials to be short, on the order of 1 nm. The estimated spin Hall angles of the four materials follow the relationship PtMn > IrMn > PdMn > FeMn, highlighting the correlation between the spin-orbit coupling of nonmagnetic species and the magnitude of the spin Hall effect in their antiferromagnetic alloys. These experiments are compared with first-principles calculations. Engineering the properties of the antiferromagnets as well as their interfaces can pave the way for manipulation of the spin dependent transport properties in antiferromagnet-based spintronics.

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The positive polarity of the ISHE voltage with respect to the AMR indicates a positive spin Hall angle for all four materials. Since both the ISHE and AMR components have the same power dependence, the resultant dc voltage is a sum of the two [25, 26]. We define a parameter $W_{\text{ISHE}}$, which represents the weight of the symmetric component (ISHE). $W_{\text{ISHE}}$ can be further expressed in the form of $W_{\text{ISHE}} = 1/(1 + V_{\text{AMR}}/V_{\text{ISHE}})$, and the ratio of the two components can be written as [27]

$$
\frac{V_{\text{ISHE}}}{V_{\text{AMR}}} = \frac{\gamma_{\text{SH}} E f g_{\text{mix}} t_{\text{AF}}}{R_{\text{CPW}} I_{\text{CPW}} \Delta R_{\text{FM}} / R_{\text{FM}} \Delta H R_{\text{FM}} \tan(\frac{t_{\text{AF}}}{2 \lambda_{\text{sf}}})},
$$

(1)

where $\gamma_{\text{SH}}$ is the spin Hall angle, $L$ is the device length, $E$ is the ellipticity correction, $R_{\text{CPW}}$ is the CPW resistance, $I_{\text{CPW}}$ is the microwave current passing along the CPW, $\Delta R_{\text{FM}} / R_{\text{FM}}$ is the FM anisotropic magnetoresistance, $h_{\text{rf}}$ is the Oersted field, $\Delta H$ is the linewidth, and $\rho_{\text{FM}}$ is the FM resistivity [27]. Notably, the AF thickness dependence is only contained in the distribution of the spin accumulation $\tan(\frac{t_{\text{AF}}}{2 \lambda_{\text{sf}}})$. Therefore, the spin diffusion length can be extracted from fitting the thickness-dependent ratio $W_{\text{ISHE}}$ (Fig. 3). This model was initially used for Py/NM bilayers, but it can be applied to the current scenario due to the facts that (1) Cu has a negligible spin Hall effect and long spin diffusion length (on the order of hundreds of nm), and (2) Cu shunts the voltages from both AMR and ISHE; therefore, the parameter $W_{\text{ISHE}}$ is weakly affected. Experimentally, we found that the absolute values of $W_{\text{ISHE}}$ for Py/AF are almost the same as with Py/Cu/AF but only slightly smaller, and the thickness dependence of $W_{\text{ISHE}}$ is almost identical for the two sets of samples. One possible explanation of the smaller $W_{\text{ISHE}}$ values for Py/AF could be the exchange spring effect [28, 29] causing an additional AMR contribution from the AF moments oscillating together with the Py moments [10, 13]. We obtain the spin diffusion lengths for the four AF materials (Fig. 3): $\lambda_{\text{sf}}(\text{FeMn}) = 1.8 \pm 0.5$ nm, $\lambda_{\text{sf}}(\text{PdMn}) = 1.3 \pm 0.1$ nm, $\lambda_{\text{sf}}(\text{IrMn}) = 0.7 \pm 0.2$ nm, and $\lambda_{\text{sf}}(\text{PtMn}) = 0.5 \pm 0.1$ nm. Such small spin diffusion lengths are comparable to the mean free paths estimated from their resistivity values; however, the diffusion model, which lays the foundation of the above analysis, is still valid in this limit [30].

In Eq. (1), only the spin Hall angle $\gamma_{\text{SH}}$, spin mixing conductance $g_{\text{mix}}$, and spin diffusion length $\lambda_{\text{sf}}$ are specific parameters related to the AF materials (spin current detector). Therefore, $W_{\text{ISHE}}$ can be rewritten as

$$
W_{\text{ISHE}} = \left(1 + \frac{1}{C_{\gamma_{\text{SH}} g_{\text{mix}} \lambda_{\text{sf}} \tan(\frac{t_{\text{AF}}}{2 \lambda_{\text{sf}}})}}\right)^{-1},
$$

(2)

where $C$ depends on $f$ and is otherwise only a function of the CPW and FM layer, and is thus independent of the AFs. We note that for very thin AF layers, it is likely that the Néel temperatures may be below RT due to the finite size effects [31, 32], which may affect the determination of $\lambda_{\text{sf}}$. Using a phenomenological model [31] and realistic parameters [33] we estimate the critical thicknesses for paramagnetic to antiferromagnetic transitions at RT to be $\sim 1$–2 nm for all the AFs studied here. However, at sufficiently high AF thickness, $t_{\text{AF}} > 3 \lambda_{\text{sf}}$, the AF layers are antiferromagnetically ordered and the $W_{\text{ISHE}}$ also saturates with the AF thickness $\tan(\frac{t_{\text{AF}}}{2 \lambda_{\text{sf}}}) \approx 1$, i.e., $W_{\text{ISHE}}^{\text{sat}} = \left[1 + (1 / C_{\gamma_{\text{SH}} g_{\text{mix}} \lambda_{\text{sf}}})\right]^{-1}$. We use the $W_{\text{ISHE}}^{\text{sat}}$ values obtained from the thicker samples ($t_{\text{AF}} \gtrsim 5$ nm) for the determination of $\gamma_{\text{SH}}$ of the antiferromagnetically ordered phases. According to Fig. 3, $W_{\text{ISHE}}^{\text{sat}}$ values of the four AFs at 9 GHz are $0.22 \pm 0.01$ (FeMn), $0.18 \pm 0.01$ (PdMn), $0.18 \pm 0.01$ (IrMn), and $0.31 \pm 0.01$ (PtMn). Using the $\lambda_{\text{sf}}$ values determined above and neglecting the variation of...
attributed the origin of the anomalous Hall effect of IrMn [16] to the transfer of spin-orbit coupling through their orbital hybridization [16,34]. This result is not unexpected, since recent theoretical work [19,36], For Py/Cu/AF structures, it has been found that the spin mixing and the spin-to-charge conversion are driven by the Cu/AF interface and the AF spin absorption, respectively [18,22,23]. For Py/AF structures, the spin mixing is affected by interface magnetic ordering and exchange coupling.

We present data for IrMn as an example. As illustrated in Fig. 4, the linewidth for Py/IrMn is higher than that for Py/Cu/IrMn, and both are greater than for pure Py. The damping enhancement can thus be extracted from a linear fit for the frequency dependence of the linewidth broadening ($\Delta H$) [25,27]. As shown in Fig. 4(a), the damping enhancement for Py/Cu/IrMn is around $\Delta \alpha = 1 \times 10^{-3}$. $g_{\text{mix}}$ is estimated around 12 nm$^{-2}$ via $g_{\text{mix}} = (4\pi M_r \mu_B / g \mu_B) \Delta \alpha$ [21,25,27], where $g$, $\mu_B$, and $M_r$ are the Landé $g$ factor, Bohr magneton, and saturation magnetization of Py, respectively. Measurements on FeMn, PdMn, and PtMn show less discernable damping enhancement. In order to compare our measurements to the theoretical calculations, we assume the same value of $g_{\text{mix}}$ for all measured AFs. Using the predetermined resistivity values of each AF, we estimated the spin Hall angle of the AFs to be 0.008 $\pm$ 0.002 for FeMn, 0.015 $\pm$ 0.005 for PdMn, 0.022 $\pm$ 0.005 for IrMn, and 0.060 $\pm$ 0.010 for PtMn, respectively. The corresponding spin Hall conductivities ($\sigma_{\text{exp}}$) are also calculated and shown in Table I. These estimated values are in agreement with the previous analysis using the ratio

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**FIG. 3** (color online). Thickness dependence of the weight $W_{\text{SHE}}$ of the symmetric component and estimation of the spin diffusion length for FeMn, PdMn, IrMn, and PtMn at RT.

$g_{\text{mix}}$ for now, we estimated the relative strength of the spin Hall angle (using FeMn as reference) of the four AF materials, FeMn:PdMn:IrMn:PtMn = 1:1:1:2.1:5.7. This result is not unexpected, since recent theoretical work [16] attributed the origin of the anomalous Hall effect of IrMn, not only to the triangular spin textures but also to the large spin orbit coupling of the heavy Ir atoms which is transferred to the magnetic Mn atoms by their hybridization. Further, another experiment showed that the spin-orbit strength of a nonmagnetic metal in a magnetic alloy directly dictates the magnitude of the anomalous Hall effect [34]. Therefore the 5$d$-metal-alloy AFs (PtMn and IrMn) are expected to exhibit stronger intrinsic SHE than the 4$d$-metal-alloy PdMn, which is further stronger than the 3$d$-metal-alloy FeMn. Recent work has demonstrated quantitative scaling of the spin Hall angle with the atomic number in a series of noble metals [35]. Our results further highlight the important role of the spin-orbit coupling of the heavy elements for the properties of their simple alloys which also acquire a sizable spin-orbit effect due to the transfer of spin-orbit coupling through their orbital hybridization [16,34].

In order to determine the spin Hall angle and spin Hall conductivity quantitatively from the experimental data, it is also necessary to determine the spin mixing conductance $g_{\text{mix}}$, which is usually extracted from the damping enhancement ($\Delta \alpha$) in the spin pumping experiment. For para-magnetic metals, the damping enhancement is mostly related to bulk spin absorption. However, additional contribution may arise due to magnetic ordering of interfaces [19,36]. For Py/Cu/AF structures, it has been found that the spin mixing and the spin-to-charge conversion are driven by the Cu/AF interface and the AF spin absorption, respectively [18,22,23]. For Py/AF structures, the spin mixing is affected by interface magnetic ordering and exchange coupling.

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**FIG. 4** (color online). (a) Frequency dependence of the linewidth broadening for the IrMn set of samples, and (b) the extraction of the damping enhancement with respect to pure Py at RT and 10 K.
between ISHE and AMR. Finally, we note that the spin Hall angle for PtMn is comparable to that of Pt obtained by the same experimental approach [24], which opens up the possibility of using PtMn as spin current detector in a variety of AF-spintronics applications.

The Py/AF bilayers showed additional damping enhancement, which is 3 or 4 times larger than in the Py/Cu/AF samples (Fig. 4). We attribute this additional damping enhancement to the FM/AF exchange coupling at the interface. Since AFs usually have relatively high anisotropy, the transfer of the spin angular momentum from precessing FM(Py) to the AF experiences additional dissipation induced by the rigid AF spin lattices under direct FM/AF exchange coupling. We also measured the frequency dependent linewidth at 10 K for one sample, 

\[
\sigma_{ij}^x = -\frac{2e\hbar}{N} \sum_k \sum_{c_{kn}} \sum_{Q_s} \frac{\langle k|n|Q_s|k|m\rangle\langle k|m|v_i|k\rangle}{(\epsilon_{kn} - \epsilon_{km})^2},
\]

(3)

where \(E_F\) is Fermi energy, \(N\) is the number of \(k\) points, \(\epsilon_{kn}\) is the band energy, \(v_i\) is the velocity component in direction \(j\), and \(Q_s = (\hbar/4V)\sigma_i v_i + \sigma_j\) is the spin current density operator for spin current flowing in the \(i\) direction with spin pointing in the \(s\) direction. Here, \(V\) is the volume of the unit cell and \(\sigma_s\) a Pauli matrix. We employed the Wannier interpolation technique [42] to make the evaluation of Eq. (3) computationally efficient. We constructed 18 maximally localized Wannier functions per atom describing the valence states [43,44]. A \(512 \times 512 \times 512\) Monkhorst-Pack \(k\) mesh [45] was employed to perform the Brillouin zone summation in Eq. (3). The resulting SHE conductivities are listed in Table I.

Assuming that the thin layers of Pt\(_{50}\)Mn\(_{50}\), Ir\(_{50}\)Mn\(_{50}\), Pd\(_{50}\)Mn\(_{50}\), and Fe\(_{50}\)Mn\(_{50}\) on Py are (111) textured and that the individual crystallites are rotated randomly around the (111) direction we obtain polycrystalline average SHE conductivities listed as \(\bar{\sigma}\) in Table I for fixed collinear staggered magnetization directions along the \(a\) and \(c\) directions within the crystallites. Assuming additionally that the direction of the staggered magnetization in the crystallites is random, we compute averaged SHE conductivities as

\[
\sigma_{av} = \frac{2\bar{\sigma}(a - \text{axis}) + \bar{\sigma}(c - \text{axis})}{3},
\]

(4)

which are also given in the Table I. It is noted that the values for PtMn, PdMn, and IrMn follow qualitatively the trend established by the experiments, which validates the averaging of magnetization directions for polycrystalline films. The larger discrepancy between experiment and theory

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<tr>
<th>TABLE I. Calculated SHE conductivities (\sigma_{ij}^x) for staggered magnetization along (a)-axis and (c)-axis directions and comparison with experimental values ((\sigma_{exp})) [units in ((\hbar/e^2))] for PtMn, IrMn, and PdMn exhibiting c4 symmetry around the (c)-axis, which results in (\sigma_{xc} = -\sigma_{cx}, \sigma_{xz} = -\sigma_{zx}), and (\sigma_{yx} = -\sigma_{xy}). Because of the spontaneous magnetic moments on Fe, this symmetry is absent in FeMn.</th>
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observed for FeMn (including a sign difference) may be due to its pronounced noncollinear magnetism.

In summary, we studied four metallic AF materials with the same chemical structure as possible spin current detectors using spin pumping and inverse spin Hall effect. By thickness-dependent measurements, we determined the spin diffusion length of these materials to be all rather short, on the order of 1 nm. We estimated the strength of the spin Hall angle of the four materials, in which the PtMn showed a large value, comparable to that of Pt. The estimated spin Hall angles of the four materials follow the relationship PtMn > IrMn > PdMn > FeMn, corroborating the important role of the spin-orbit coupling of the heavy metals for the properties of the Mn-based alloys through orbital hybridization. By comparing samples with and without a Cu spacer, we confirmed an additional damping enhancement due to the exchange coupling at the FM/AF interface, in addition to that induced by the transfer of spin angular momentum from spin pumping. We also performed first-principles calculations of ordered alloys and showed that the value of spin Hall conductivity can vary significantly with crystal orientation and staggered AF magnetization; calculations of averaged spin Hall conductivities for polycrystalline systems are in agreement with our experimental results for PdMn, IrMn, and PtMn. Future works on ordered epitaxial systems may allow tailoring the sign and magnitude of the spin Hall conductivities by manipulating the growth along different crystal orientations in these antiferromagnets.

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