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Relation between spin Hall effect and anomalous Hall effect in 3d ferromagnetic metals

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We study the mechanisms of the spin Hall effect (SHE) and anomalous Hall effect (AHE) in 3d ferromagnetic metals (Fe, Co, permalloy (Ni81Fe19; Py), and Ni) by varying their resistivities and temperature. At low temperatures where the phonon scattering is negligible, the skew scattering coefficients of the SHE and AHE in Py are related to its spin polarization. However, this simple relation breaks down for Py at higher temperatures as well as for the other ferromagnetic metals at any temperature. We find that, in general, the relation between the SHE and AHE is more complex, with the temperature dependence of the SHE being much stronger than that of AHE.

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The spin Hall effect (SHE) and its inverse (ISHE) enable us to interconvert spin and charge currents in the transverse direction and are widely recognized as essential methods to generate and detect spin currents in spintronic devices. Since the original predictions of the SHE, it has been experimentally investigated in a variety of nonmagnetic materials with strong spin-orbit interactions such as III-V semiconductors,4–6 4d and 5d transition metals,7–16 alloys,17–20 oxides,21 and organic materials.22 The mechanism of the SHE can be extrinsic or intrinsic. The former depends on the combination of the host metal and impurities, while the latter depends on the detailed properties of the momentum-space Berry phase.23,24 These mechanisms are the same as for the anomalous Hall effect (AHE) in ferromagnetic metals (FMs), which has been intensively studied for many years.25 Thus, it has been commonly accepted that the SHE shares the same origin as the AHE26.

Recently, it was experimentally verified that the SHE also occurs in FMs with finite spin polarization.20–22 Intuitively, in FMs, both spin and charge accumulations can exist and are detected as the SHE and the AHE, respectively (see Fig. 1(a)). Thus, it was suggested that the SHE and AHE in FMs are related via the spin polarization.23 However, it has not been experimentally verified if this simple relation is general, and therefore valid for all the FMs and all the mechanisms. From a theoretical viewpoint, such a relation might hold in the limit of diffusive transport but is not expected to hold in general.

In this work, we present a detailed investigation of the relation between the SHE and the AHE in four different 3d FMs, i.e., Fe, Co, permalloy (Ni81Fe19; Py), and Ni. By changing the residual resistivity of the FM at low temperatures, the skew scattering contribution (one of the extrinsic mechanisms) can be separated from other contributions. It turns out that the aforementioned relation between the SHE and AHE holds for the skew scattering term in Py. However, this simple relation is not valid for the other mechanisms in Py and for the other FMs. The SHE in the 3d FMs has much stronger temperature dependence than the AHE. We discuss a possible scenario to explain the observed results.

To perform the SHE measurements in the 3d FMs, we adopted the spin absorption method in the lateral spin valve structure.13,15,16,18–21 This method enables us to estimate the spin diffusion length and the spin Hall angle (θSHE) on the same device. The SHE devices were fabricated on SiO2/Si substrates with multiple-step electron beam lithography followed by metal deposition and lift-off. We first patterned two 100-nm-wide wires and deposited Py by 30 nm in thickness by electron beam evaporation. The two Py wires are separated by a length (L) of 1 µm, as illustrated in Fig. 1(b). One of the Py wires is used as a spin current injector, while the other is used to estimate the spin diffusion length of our target wire, as detailed in Supplemental Material. In the second step, the target 3d FM wire (hereafter middle wire) with the width (wM) of 200 nm was placed just in the middle of the two Py wires and a 5- to 30-nm-thick 3d FM (Fe, Co, Py, or Ni) was deposited with electron beam evaporation. In the third step, a 100-nm-wide and 100-nm-thick Cu strip was bridged on top of the three wires with a Joule heating evaporator. Before the Cu evaporation, an Ar-ion milling treatment was performed to achieve transparent interfaces. For the AHE measurements, a 20-µm-long and 3-µm-wide Hall bar was patterned with...
electron beam lithography and the FM (5 to 30 nm in thickness) was deposited at the same time as the SHE devices were prepared. We then capped all the devices with Al₂O₃ using radio frequency magnetron sputtering to protect them from oxidization. All the electric transport measurements were performed in a ⁴He flow cryostat using the lock-in technique.

When an electric current $I_C$ is injected from Py to the left side of Cu as shown in Fig. 1(b), spin accumulation is created at the interface and diffuses in the Cu bridge. In this process, a pure spin current $(V_+ \leftrightarrow I_+, V_- \leftrightarrow I_-)$ on the same device and measuring the direct SHE, as detailed in Supplemental Material. It is well-known that the AHE occurs in FM as a result of the breaking of time reversal symmetry. However, the Onsager reciprocal relation holds for the SHE in FM because the total number of spin-up and spin-down electrons is always kept constant.

By using the spin transport model proposed by Takahashi and Maekawa, the spin Hall resistivity $-\rho_{xy}^{\text{SHE}}$ can be estimated as follows:

$$-\rho_{xy}^{\text{SHE}} = \rho_{yx}^{\text{SHE}} = \theta_{\text{SHE}}\rho_{xx} = \frac{\mu_{\text{FM}}}{x} (I_C/I_S) \Delta R_{\text{ISHE}}$$  \hspace{1cm} (1)$$

where $x$ is the shunting factor and $I_S$ is the effective spin current absorbed into the FM middle wire. When $I_S$ is converted into charge current in the middle wire, a part of the charge current is shunted by the Cu bridge on the middle FM wire. The shunting factor $x$ has been calculated with a finite elements method using SpinFlow 3D. $I_S$ can be determined from nonlocal spin valve measurements with and without the middle wire. $I_S$ is also related to the spin diffusion length of the middle FM wire.

We next measured the AHE with a Hall bar pattern, prepared at the same time as the SHE device. By applying an out-of-plane magnetic field and flowing $I_C$ in the longitudinal direction of the Hall bar, a transverse voltage drop $V_{\text{AHE}}(= V_+ - V_-)$ is detected as sketched in the inset of Fig. 1(d). Figure 1(d) shows a typical $R_{\text{AHE}} = V_{\text{AHE}}/I_C$ vs $H$ curve for Py at 10 K. Although there are two backgrounds, namely normal Hall resistance and planar Hall resistance in between $\pm 10$ kOe, a clear positive AHE signal $\Delta R_{\text{AHE}}$ can be extracted. From $\Delta R_{\text{AHE}}$, we obtain the anomalous Hall resistivity defined as,

$$-\rho_{xy}^{\text{AHE}} = \rho_{yx}^{\text{AHE}} = \theta_{\text{AHE}}\rho_{xx} = t\Delta R_{\text{AHE}},$$  \hspace{1cm} (2)$$

where $\theta_{\text{AHE}}$ is the anomalous Hall angle and $t$ is the thickness of the Hall bar.
By plotting $-\rho_{xy}^{\text{SHE}}$ and $-\rho_{xy}^{\text{AHE}}$, obtained with Eqs. (1) and (2), as a function of $\rho_{xx}$, the detailed mechanisms can be addressed as shown in previous works\cite{16,39,40}. For this purpose, the recent scaling equation proposed by Hou \emph{et al.}\cite{39} is useful:

$$-\rho_{xy}^{H} = \alpha_{ss}^{H}\rho_{xx,0} + \beta_{0}^{H}(\rho_{xx,0})^{2} + \gamma^{H}\rho_{xx,0}\rho_{xx,T} + \beta_{1}^{H}(\rho_{xx,T})^{2},$$

where $H$ refers to the SHE or AHE, $\rho_{xx,0}$ is the residual resistivity at low temperature (in the present case, at 10 K), $\rho_{xx,T}(=\rho_{xx} - \rho_{xx,0})$ is the resistivity induced by phonons and $\alpha_{ss}^{H}$ is the skew scattering angle due to impurities or grain boundaries. As detailed in Ref.\cite{39}, the side-jump terms due to static (impurities or grain boundaries) and dynamic (phonons) scattering sources as well as the intrinsic contribution originating from the band structure\cite{41,47} are entangled in $\beta_{0}^{H}$, $\gamma^{H}$, and $\beta_{1}^{H}$ in a complex manner. Nevertheless, as discussed in Ref.\cite{39}, the effect of the intrinsic Berry curvature is most strongly reflected in the $\beta_{1}^{H}$ term.

Firstly, to simplify Eq.\ (4), we focus on the low temperature part where the phonon contribution is negligible and consider the case of Py. By substituting $\rho_{xx,T} = 0$ in Eq.\ (3), a simplified equation can be obtained:

$$-\rho_{xy}^{H} = \alpha_{ss}^{H}\rho_{xx,0} + \beta_{0}^{H}(\rho_{xx,0})^{2}.$$  

In order to determine $\alpha_{ss}^{H}$ and $\beta_{0}^{H}$, the SHE and AHE of Py have to be measured in a wide $\rho_{xx,0}$ range. For this purpose, we changed the thickness of the Py wire (from 5 to 30 nm) and also the deposition rate (from 0.04 nm/s to 0.08 nm/s), as already demonstrated in our previous work for Py\cite{16}. Figures\ (a) and (b) show $-\rho_{xy}^{\text{SHE}}$ and $-\rho_{xy}^{\text{AHE}}$ of Py at 10 K as a function of $\rho_{xx,0}$, respectively. $-\rho_{xy}^{\text{SHE}}$ increases with increasing $\rho_{xx,0}$, while $-\rho_{xy}^{\text{AHE}}$ decreases with $\rho_{xx,0}$. By fitting $-\rho_{xy}^{\text{SHE}}$ and $-\rho_{xy}^{\text{AHE}}$ with Eq.\ (4), the skew scattering term $\alpha_{ss}^{H}$ and the combination of the side-jump and intrinsic contributions $\beta_{0}^{H}$ can be obtained as follows: $\alpha_{ss}^{\text{SHE}} = 1.0 \pm 0.4\%$, $\alpha_{ss}^{\text{AHE}} = 0.32 \pm 0.1\%$, $\beta_{0}^{\text{SHE}} = 131 \pm 60$ $\Omega^{-1}\text{-cm}^{-1}$ and $\beta_{0}^{\text{AHE}} = -76 \pm 20$ $\Omega^{-1}\text{-cm}^{-1}$. $\alpha_{ss}^{\text{AHE}}$ and $\beta_{0}^{\text{AHE}}$ are in good agreement with previous reports\cite{38,48}.

Interestingly, the ratio of the AHE and SHE in Py for the skew scattering contribution, $\alpha_{ss}^{\text{AHE}}/\alpha_{ss}^{\text{SHE}} = 0.32$, is a reasonable value for the spin polarization $(p = (3 - 2)/(3 + 2) = 0.2$ in the sketch of Fig.\ (a)).

In fact, this simple picture can be applied for diffusive scattering systems such as Py. As we detail in Ref.\cite{35}, Eq.\ (5) can be derived in Mott’s two current model\cite{50} under the specific assumption $\rho_{xy}^{\text{SHE}}/\rho_{xx}^{\text{SHE}} = -\rho_{xy}^{\text{AHE}}/\rho_{xx}^{\text{AHE}}$. Here $\rho_{ij}^{\uparrow}$ and $\rho_{ij}^{\downarrow}$ are the spin-up and spin-down resistivity tensor elements. Py is a random alloy composed of Ni and Fe. The anisotropy on the Fermi surface should be suppressed and lead to more isotropic scattering properties. Thus, the Hall angle is essentially a spin-independent property averaged over all the contributing states. This supports the finding that the simplified relation holds for the skew scattering in Py.

However, such a simple picture does not work for the other 3d FMs. The electronic states can be quite anisotropic because of the complicated band structure.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{Fig3}
\caption{(Color online) (a) Spin Hall angle $\theta_{\text{SHE}}$ and (b) anomalous Hall angle $\theta_{\text{AHE}}$ measured at 10 K as a function of the number of electrons in the outermost shell. The thickness of the 3d FMs is 20 nm. The dotted lines in (a) and (b) indicate $\theta_{H} = 0$.}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{Fig4}
\caption{(Color online) The temperature dependence of (a) spin Hall resistivity $-\rho_{xy}^{\text{SHE}}$ and (b) anomalous Hall resistivity $-\rho_{xy}^{\text{AHE}}$ in four FMs (Py, Fe, Co and Ni). The thickness of the four FMs is 20 nm both for the SHE and AHE measurements.}
\end{figure}
of 3d FMs. Those states should show distinct effective spin-orbit couplings for spin-up and spin-down electrons. Thus, the above specific assumption can break down. We show \( \theta_{\text{SHE}} \) and \( \theta_{\text{AHE}} \) at \( T = 10 \) K for the 3d FMs in Figs. 3(a) and 3(b), respectively. As in the case of the intrinsic SHEs in 4d and 5d transition metals, \( \theta_{\text{SHE}} \) is expected to change the sign from negative to positive with increasing the number of electrons in the outer shell. Such a tendency can be seen clearly in \( \theta_{\text{SHE}} \) of the 3d FMs in Fig. 3(a). However, the sign of \( \theta_{\text{SHE}} \) is opposite to that of \( \theta_{\text{AHE}} \) for Fe, Co, and Ni. Even in the case of Py, \( \theta_{\text{SHE}} \) is negative when \( \rho_{xx,0} \) is more than 40 \( \mu \Omega \cdot \text{cm} \), as shown in Fig. 2(b). This obviously shows that Eq. (5) is not general and the detailed band structure of the electron orbitals has to be taken into account, as mentioned above.

So far, we have focused on the low temperature parts of the SHE and AHE. To address the effect of dynamic disorders, we next discuss the temperature dependences of the SHE and AHE in Figs. 3(a) and 3(b), respectively. The temperature dependence of the SHE is much stronger than that of the AHE. For Fe, Py, and Ni, the sign of \( -\rho_{xy}^{\text{SHE}} \) is changed at 200-250 K, while such a sign change cannot be seen for \( -\rho_{xy}^{\text{AHE}} \). To specify the reason for such temperature dependences, we have fitted both \( -\rho_{xy}^{\text{SHE}} \) and \( -\rho_{xy}^{\text{AHE}} \) as a function of \( \rho_{xx,0} \) with Eq. (3) as shown in Fig. 3 and obtained \( \beta_1^{\text{SHE}} \) and \( \gamma^{\text{SHE}} \) as the quadratic and linear terms in Eq. (3), respectively (see Table I). For example, \( \beta_1^{\text{SHE}} \) of Py is more than two orders of magnitude larger than \( \beta_1^{\text{AHE}} \). Even for Py, the relations between the SHE and AHE for \( \beta_1^{\text{H}} \) and \( \gamma^{\text{H}} \) are not as simple as the skew scattering term.

A similar tendency can be seen for the other 3d FMs. For Fe, Co, and Ni, \( \beta_1^{\text{SHE}} \) is one or two orders of magnitude larger than \( \beta_1^{\text{AHE}} \), as shown in Table I. Note that \( \beta_1^{\text{SHE}} \) values in the present work are in good agreement with previous experiments (see Table I) and tight-binding calculations. On the other hand, \( \beta_1^{\text{H}} \) of the 3d FMs ranges between 4.9 and 17 \( \times 10^3 \) \( \Omega^{-1} \cdot \text{cm}^{-1} \), which is larger than that of a typical SHE material, Pt (1.6 \( \times 10^3 \) \( \Omega^{-1} \cdot \text{cm}^{-1} \)) and Ni (1.6 \( \times 10^3 \) \( \Omega^{-1} \cdot \text{cm}^{-1} \)). The relation between \( \gamma^{\text{SHE}} \) and \( \gamma^{\text{AHE}} \) strongly varies with the 3d FMs (see Table I).

Much larger \( \beta_1^{\text{SHE}} \) values than \( \beta_1^{\text{AHE}} \) ones would originate from the stronger temperature dependence of the SHE in 3d FMs. At the moment, we do not have a conclusive picture for the origin of this dependence. In general, the spin transport can be mediated not only by conductive electrons but also by magnons in FMs. One possible scenario is the contribution of electron-magnon interactions in 3d FMs. The electron-magnon interactions would induce additional spin-flip processes. We note that such spin-flip processes are equivalent in magnitude for up-to-down and down-to-up spin channels even in ferromagnetic systems. In such a situation, some asymmetric scatterings which are spin-dependent would contribute only to the SHE but not to the AHE, and thus would be associated with the fact that the strong temperature dependence is not present in the AHE of the 3d FMs or the SHE of nonmagnetic metals. Interestingly, a recent theoretical report claims that magnon spin current can be significant around room temperature in 3d FMs, which might be related to our case. However, there are some open questions: how large the asymmetric scatterings are quantitatively and whether any other mechanisms contribute to the observed spin Hall resistivity or not. These would be addressed in future.

In conclusion, we experimentally investigated the relation between the SHE and AHE in four 3d FMs (Fe, Co, Py (Ni81Fe19), and Ni). In a typical ferromagnetic alloy, Py, the skew scattering contribution of the AHE is related to that of the SHE via the spin polarization of Py, as can be understood intuitively. However, this relation does not hold for other mechanisms. This fact is highlighted by the temperature dependence of the SHE and AHE. For all the 3d FMs, one of the intrinsic mechanism terms \( \beta_1^{\text{SHE}} \) is much larger than \( \beta_1^{\text{AHE}} \). Asymmetric spin-dependent scatterings in the spin-flip processes induced by the electron-magnon interactions would be a possible explanation for the strong temperature dependence of the SHE in contrast to the AHE or even the SHE in nonmagnetic metals.

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FIG. 5: Spin Hall resistivity $-\rho_{xy}^{\text{SHE}}$ and anomalous Hall resistivity $-\rho_{xy}^{\text{AHE}}$ in four FMs (Py, Fe, Co and Ni) at finite temperatures. $-\rho_{xy}^{E_{K}}$ as a function of $x_{xx,T}$ in (a) Fe, (c) Co, (e) Py, and (g) Ni. Anomalous Hall resistivity $-\rho_{xy}^{\text{AHE}}$ as a function of $x_{xx,T}$ in (b) Fe, (d) Co, (f) Py, and (h) Ni. $x_{xx,T}$ varies by changing temperature from 10 K to 300 K. The solid lines are the best fits of the data to Eq. (3). The thickness of the four FMs is 20 nm both for SHE and AHE measurements except for the inset in (e). The inset in (e) shows $-\rho_{xy}^{E_{K}}$ of 5 nm thick Py wire. For the fitting with Eq. (3), the same parameter $\beta_{E_{K}}^{\text{SHE}} = -1.0(1 \times 10^{3} \text{ $\Omega^{-1}$ cm}^{-1})$ was used both for 20 nm and 5 nm thick Py wires.

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Within the semiclassical picture, the anomalous Hall conductivities arising from scatterings are given by \( \sigma_{xy} = \frac{1}{(2\pi)^2} \int_{FS} s_\alpha(k) \lambda_\alpha(k) \, dS \) and \( \sigma_{xy}^{SHE} = -\frac{2}{(2\pi)^2} \int_{FS} s_x(k) v_x(k) \lambda_y(k) \, dS \), respectively. Here \( v_x(k) \) are the Fermi velocities, \( \lambda_i(k) \) is the mean free path, \( s_x(k) \) is the spin polarization and we integrate over the Fermi surface (FS). In the limit of diffusive transport with an isotropic spin polarization \( s_x(k) = p \), we can take \( p \) in front of the integral and we find \( \rho_{xy}^{AHE} = \rho_{xy}^{SHE} \).


34 See Supplemental Material at https://... for details of the evaluation of the spin diffusion length of the middle wire and \( J_t \), the Onsager reciprocity in Py, and the relation between SHE and AHE using the simplified model.


48 Strictly speaking, \( \rho_{0}^{AHE} \) and \( \beta_{1}^{AHE} \) are different from \( b \) which is the coefficient of \( \rho_{xx}^{AHE} \) in Ref. 35. In general, \( b \) should be closer to \( \beta_{1}^{AHE} \), but in the case of Py, both \( \rho_{0}^{AHE} \) and \( \beta_{1}^{AHE} \) are comparable to \( b \).


54 The scattering probability related to the spin-flip process is defined as \( P_{kk'}^{\pm} \sim |\langle \Psi_{k'}^- | \hat{T} | \Psi_{k}^+ \rangle|^2 \) where \( \Psi_{k}^{\pm} \) is the wave function before (after) the scattering with the wave number \( k(k') \) and spin-up (spin-down −), and \( \hat{T} \) is a matrix in spin space including spin-orbit coupling and magnetism. By defining \( P_{kk'}^{\pm} \) in an equivalent way and integrating \( k \) and \( k' \), we find that on average it holds \( \tau_{\alpha}^{+} = \tau_{\alpha}^{-} \), with \( \tau_{\alpha}^{\pm} \equiv \int dk \int dk' P_{kk'}^{\pm} \), even in the magnetic case.