Insight into the skew-scattering mechanism of the spin Hall effect: potential scattering versus spin-orbit scattering

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We present a detailed analysis of the skew-scattering contribution to the spin Hall conductivity using an extended version of the resonant scattering model of Fert and Levy [Phys. Rev. Lett. 106, 157208 (2011)]. For 5d impurities in a Cu host, the proposed phase shift model reproduces the corresponding first-principles calculations. Crucial for that agreement is the consideration of two scattering channels related to $p$ and $d$ impurity states, since the discussed mechanism is governed by a subtle interplay between the spin-orbit and potential scattering in both angular-momentum channels. It is shown that the potential scattering strength plays a decisive role for the magnitude of the spin Hall conductivity.

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An intriguing direction in the development of spintronic devices is based on the spin Hall effect (SHE). This phenomenon, caused by spin-orbit coupling (SOC), provides an opportunity for spin current generation in nonmagnetic materials without injection from ferromagnets. Materials with a large spin Hall angle (SHA), described recently, offer an attractive route to obtain a giant SHE.

For the understanding of the essential conditions of such a strong SHE, a microscopic analysis of this mechanism is desired. In particular, models with input parameters provided by first-principles calculations are helpful for an intuitive picture and a detailed analysis of the material specific SHE. Recently, Fert and Levy have proposed a resonant scattering model and applied it to a Cu host with 5d impurities. The well-known behavior of the residual resistivity of noble metals with transition metal impurities is qualitatively well described by Friedel’s $d$ resonance model. Therefore, it was assumed that the spin-orbit driven transverse transport in these systems is mainly caused by impurity $d$ states. In this paper we show that despite such a seemingly sensible assumption, the contribution related to the spin-orbit scattering in the $p$ channel is comparably large for the considered systems. Consequently, the total spin Hall conductivity (SHE) is predominantly caused by two scattering channels related to both $p$ and $d$ states. This somewhat surprising result is caused by the vertex corrections, which play an ultimate role for the skew-scattering mechanism. They enter the semiclassical approach via the so-called scattering-in term of the Boltzmann equation. Furthermore, we show that for the considered 5d series of impurity atoms in copper the magnitude of the SHE is mainly determined by the potential scattering strength reflected in the momentum relaxation time.

The paper is organized in three parts. First, we provide a derivation of the proposed phase shift model as an extended version of the resonant scattering model considered in Ref. for a description of the SHE. Then, we present the results for 5d impurities in a Cu host obtained within this model in comparison to 	extit{ab initio} results, to confirm its validity. Finally, we perform a detailed analysis identifying the significant contributions to the SHE to elucidate our findings outlined above.

Within the semiclassical approach the conductivity tensor can be written, using the spherical band approximation, as

\[
\hat{\sigma} = \frac{e^2}{V} \sum_k \delta(\varepsilon_k - \varepsilon_F) \mathbf{v}_k \otimes \Lambda_k = \frac{e^2 \hbar^2}{k_v} \sum_i \int d\Omega_k \mathbf{v}_k \otimes \Lambda_k ,
\]

where the mean free path is given by the Boltzmann equation

\[
\Lambda_k = \Lambda_k^{\text{out}} + \Lambda_k^{\text{in}} = \tau_k (\mathbf{v}_k + \sum_{k'} P_{k'\rightarrow k'} \Lambda_{k'})
\]

with the momentum relaxation time

\[
\frac{1}{\tau_k} = \sum_{k'} \frac{P_{k'\rightarrow k}}{k_v'} = \frac{2\pi c_i N}{k_v} \sum_{k'} |T_{k'\rightarrow k}|^2 \delta(\varepsilon_k - \varepsilon_{k'})
\]

and the group velocity $\mathbf{v}_k = \hbar \mathbf{k}/m_e$. Here, the microscopic transition probability $P_{k'\rightarrow k}$ is the scattering rate...
from an initial state $k$ into a final state $k'$. This quantity is defined by the corresponding transition matrix $T_{k'\rightarrow k}$ and scales with the impurity concentration $c_i = N_i/N$, as valid for noninteracting impurities.\textsuperscript{16}

Of crucial importance for the further discussion of the considered phenomenon is the second term on the r.h.s. of Eq. (2). This scattering-in term, which corresponds to the vertex corrections of the Kubo theory in the dilute limit of impurity concentrations,\textsuperscript{16,17} is known to be responsible for the skew-scattering mechanism.\textsuperscript{16} Moreover, only the antisymmetric part of $P_{k\rightarrow k'}$ involved in Eq. (2) contributes to the effect.\textsuperscript{15}

Following Refs. \textsuperscript{8} and \textsuperscript{13} the transition matrix for the “spin-up” (\textsuperscript{“+”}) states can be written, to first order in $\lambda_1/\Delta l$, as

$$T_{k'\rightarrow k}^{++} = \frac{4\pi^2\hbar^2}{m_k}\sum_{l,m}\frac{\hbar^2}{2m_k}\sin^2\eta_l - 2e^{i\eta_l}\sin\eta_l \times \left(Y^{m}(\hat{k})\right)^* Y^{m}(\hat{k}').$$\hspace{1cm} (4)

Here, $\Delta l$ and $\lambda_1$ are the resonance width and the SOC constant for the corresponding $l > 0$ impurity level, respectively. This expression can be derived starting from the conventional nonrelativistic transition matrix and assuming the phase shifts to become $l$-dependent as $\eta_l \rightarrow \eta_l^m \approx \eta_l - \frac{\hbar^2}{2m_k}\sin^2\eta_l$ for a perturbative treatment of the SOC.\textsuperscript{18,19} Thus, in Eq. (4) the spin-orbit scattering strength is measured by $(\lambda_1/\Delta l)\sin^2\eta_l$, while $\sin\eta_l$ in the second term relates to the strength of the potential scattering.\textsuperscript{20}

As mentioned above, the skew-scattering mechanism is solely determined by the antisymmetric part of the microscopic transition probability. Thus, one needs to calculate $|T_{k'\rightarrow k}^{++}|^2 = \left(|T_{k'\rightarrow k}^{++}|^2 - |T_{k\rightarrow k'}^{++}|^2\right)/2$. Since the first term on the r.h.s. of Eq. (4) is antisymmetric while the second one is symmetric with respect to exchange of $k$ and $k'$ we obtain

$$|T_{k'\rightarrow k}^{++}|^2_{\text{antisym}} = \frac{64\pi^2\hbar^2}{m_k}\sum_{l,m}\sum_{l',m'}\frac{\hbar^2}{2m_k}\sin(2\eta_l - \eta_{l'}) \times \sin^2\eta_l \sin\eta_{l'} Y^{m}(\hat{k}) Y^{m}(\hat{k}')^* \left(Y^{m}(\hat{k})\right)^* Y^{m}(\hat{k}')^*.$$\hspace{1cm} (5)

Within the spherical band approximation, the skew-scattering contribution to the spin Hall conductivity, $\sigma_{yx}^\perp = 2\sigma_{yx}^{\perp}$, is given by\textsuperscript{15}

$$\sigma_{yx}^+ = \frac{e^2}{h} \int d\Omega_k \int d\Omega_{k'} \frac{\hbar k_y k'_y |T_{k'\rightarrow k}^{++}|^2_{\text{antisym}}}{(2\pi)^2}.$$\hspace{1cm} (6)

This is obtained assuming $\Delta k' \approx \Delta_k^\text{out}$, for the scattering-in term of Eq. (2) as well as an isotropic momentum relaxation time $\tau_0 \approx \tau_0$. The involved components of the crystal momentum can be expressed in terms of spherical harmonics as\textsuperscript{21}

$$k_x = \frac{k_y\sqrt{2\pi}|Y_0^1(k) - Y_1^1(k)|}{\sqrt{3}} , \hspace{1cm} k_y = \frac{ik_y\sqrt{2\pi}|Y_0^1(k) + Y_1^1(k)|}{\sqrt{3}}.$$\hspace{1cm} (7)

Taking into account Eq. (5) and the integrals over three spherical harmonics\textsuperscript{22}, which are related to the Clebsch-Gordan coefficients\textsuperscript{21}, we obtain\textsuperscript{23}

$$\sigma_{yx}^+ = \frac{e^2}{h} \left(\frac{2\pi^2\hbar^2 c_0}{\pi m_k^2 V_0}\right) \tau_0^2 \times \left\{ \frac{1}{3} \lambda_1 \sin^2\eta_l \sin(2\eta_l - \eta_0) \sin\eta_0 - \sin(2\eta_l - \eta_2) \sin\eta_2 \right.$$\hspace{1cm} + $\frac{21\lambda_1}{\Delta l} \sin^2\eta_l \sin(2\eta_l - \eta_2) \sin(2\eta_l - \eta_2) \right\},$$\hspace{1cm} (8)

where $V_0$ is the unit cell volume. Here, all scattering contributions involving $s$, $p$, $d$, and $f$ states are considered, neglecting terms with $l > 3$. A more general expression for $\sigma_{yx}^+$ is provided in the Supplemental Material.\textsuperscript{23} In addition, for the isotropic momentum relaxation time involved in Eq. (8) we neglect the influence of the SOC and apply the expression,\textsuperscript{12}

$$\frac{1}{\tau_0} = \frac{4\pi^2\hbar^2}{m_k}\sum_l (2l + 1) \sin^2\eta_l,$$\hspace{1cm} (9)

which is similar to the approach used in Ref. \textsuperscript{8}.

For practical applications of Eq. (8) one needs to know the SOC constant for an impurity atom in the considered host. Often this is approximated by the corresponding atomic SOC constant.\textsuperscript{23,24} However, $\lambda_1$ for an impurity embedded in a crystal can differ significantly from the value of an isolated atom.\textsuperscript{25,26} Therefore, it is preferable to calculate $\lambda_1$ via the self-consistent electronic structure of a real impurity system.\textsuperscript{26} The resonance width required additionally can be obtained from the partial local density of states at the impurity site. Of course, the assumption of a simple Lorentz shape for all impurity states with $l > 0$, as used in Eq. (4), is commonly difficult.\textsuperscript{15} However, we can generalize Eq. (8) using the following relations,\textsuperscript{27}

$$\frac{\lambda_1 \sin^2\eta_l}{\Delta l} = \frac{2(\delta_{l-1/2} - \delta_{l+1/2})}{2l+1}, \hspace{1cm} \eta_l = i\delta_{l-1/2} + (1 + i)\delta_{l+1/2},$$\hspace{1cm} (10)

based on the phase shifts related to the relativistic quantum number $j = l \pm 1/2$. This makes it possible to apply Eq. (8) for any impurity system, since no special assumptions for the impurity states are made. Nevertheless, the virtual bound states are treated by this generalized approach as well.

For our study, the corresponding relativistic phase shifts $\delta_{l \pm 1/2}$ are obtained by first-principles calculations performed with a relativistic Korringa-Kohn-Rostoker Green’s function method.\textsuperscript{28} Their values for the considered 5d impurities in a Cu host are presented in the Supplemental Material.\textsuperscript{23}

Figure 1 shows results for the SHC obtained using Eqs. (8)-(10) in comparison to direct first-principles calculations performed applying the Boltzmann equation\textsuperscript{29} and the Kubo-Středa formula.\textsuperscript{15} Also included are the corresponding results of the relativistic phase shift model, which was presented in Ref. \textsuperscript{12}. Throughout the paper, all values of the SHC are shown for the impurity concen-
and labeled with 2 shows the two discussed points to the crucial role of the potential scattering.

Surprisingly, the \( p \rightarrow d \) contribution, related to the spin-orbit scattering in the \( p \) channel, is of the same order as the \( d \rightarrow p \) contribution. Moreover, for Pt and Au impurities the \( p \rightarrow d \) contribution becomes even significantly larger than the \( d \rightarrow p \) one.

To get a clear insight into the origin of this unexpected result, it is worth to use another helpful property of Eq. \( \text{[8]} \). The Hall conductivity given by this expression is a result of the interplay between the spin-orbit and potential scattering, which were entering additively into the transition matrix only. Nevertheless, the specific structure of Eq. \( \text{[8]} \) still allows one to separate out the spin-orbit scattering contribution from the rest of the skew-scattering contribution to the SHE. In other words, the terms \( C_{p \rightarrow d} = -\frac{1}{2} C_{12} \) and \( C_{d \rightarrow p} = C_{21} \) providing the dominant contributions to Eq. \( \text{[8]} \) can be divided into two factors. The first one, \( (\lambda_l / \Delta_l) \sin^2 \eta_l \), is the spin-orbit scattering strength for the corresponding \( l \) channel, while the rest is described merely in terms of the nonrelativistic phase shifts. Figure \( \text{[3]} \) shows the two discussed parts separately. In contrast to the SOC part related to \( l = 1 \), which has just a small enhancement going from Lu to Au, the corresponding part for \( l = 2 \) shows the resonance behavior \( \text{[9,10]} \) caused by the 5d shell filling (see Table S2 in the Supplemental Material \( \text{[23]} \)). This part is enhanced around W and reduces towards both sides. As a result, in the case of Au impurities the spin-orbit scattering strength in the \( p \) channel is four times larger than for \( d \) states. However, from Hf to Ir both contributions are of comparable magnitude and how they add up is determined by the second part of the corresponding \( C_{ll'} \) terms, which is given by

\[
\sin (2\eta_l - \eta_{l'}) \sin \eta_l = 2 \sin \eta_l \sin \eta_{l'} \cos (\eta_l - \eta_{l'}) - \sin^2 \eta_{l'} . \tag{11}
\]

Here, the used identity helps us to highlight a further important point, since the first term on the r.h.s of Eq. \( \text{[12]} \) is the same for \( ll' \) and \( ll' \) contributions. The difference is caused solely by the second term, which depends on \( \eta_2 \) and \( \eta_1 \) for the \( p \rightarrow d \) and \( d \rightarrow p \) contributions, respectively. Thus, the nature of the scattering-in term, which is responsible for the structure of Eq. \( \text{[6]} \), leads to the coupling of the spin-orbit scattering in the \( p \) channel with the potential scattering in the \( d \) channel. For the considered systems, the latter one is enhanced by the resonance properties of the 5d impurities in copper. In contrast to the results of Ref. \( \text{[8]} \), the subtle combination of the two scattering mechanisms causes a positive SHE, and consequently a positive SHA, for all the considered systems.

Moreover, the main trend of the total SHE with a significant enhancement for Pt and Au, as seen in Figs. \( \text{[1]} \) and \( \text{[2]} \) is entirely provided by the behavior of \( \eta_0 \). This is illustrated by Fig. 4 where we show the momentum relaxation time in addition to the two dominant contributions in the braces of Eq. \( \text{[8]} \). With this expression, the strong correlation between \( \tau_0 \) and the total SHE of Figs. \( \text{[1]} \) and \( \text{[2]} \) points to the crucial role of the potential scattering strength by itself for the magnitude of the SHE.

In summary, we developed a phase shift model for the skew-scattering mechanism of the SHE as an extension of
the resonant scattering model of Ref. [8]. Exploiting this approach we show that the spin Hall conductivity results from a subtle interplay between the spin-orbit and potential scattering in different $l$ channels, caused by the structure of the scattering-in term (vertex corrections). For 5$d$ impurities in copper this leads to a crucial importance of the contribution related to the spin-orbit coupling of $p$ states, which explains the unanticipated behavior of the spin Hall conductivity. The proposed phase shift model provides good agreement with results of first-principles approaches and highlights the complexity of the specific problem. In combination with parameters derived from $ab \ initio$ calculations this model can be used for a simplified description of the SHE in dilute alloys based on noble metals. However, we emphasize the utmost importance to carefully consider all leading terms.

Fig. 3: (Color online) The two parts of the contributions $C_{p-d}$ and $C_{d-p}$ are shown separately.

Fig. 4: (Color online) The contributions $C_{p-d}$ and $C_{d-p}$ are shown versus $\tau_0^2$ (in arbitrary units).

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