Spin Hall Effect Induced by Resonant Scattering on Impurities in Metals

Albert Fert

Unité Mixte de Physique CNRS/Thales, 91767, Palaiseau, France and Université Paris-Sud, 91405, Orsay, France

Peter M. Levy

Department of Physics, New York University, 4 Washington Place, New York, New York 10003, USA

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The spin Hall effect is a promising way for transforming charge currents into spin currents in spintronic devices. Large values of the spin Hall angle, the characteristic parameter of the yield of this transformation, have been recently found in noble metals doped with nonmagnetic impurities. We show that this can be explained by resonant scattering off impurity states split by the spin-orbit interaction. By using as an example copper doped with 5*d* impurities we describe the general conditions and provide a guide for experimentalists for obtaining the largest effects.

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The spin hall effect (SHE), first described by Dyakonov and Perel in 1971 [1], is a subject of intense research as it allows for the generation of spin currents in nonmagnetic conductors and the developments of spintronic devices built without ferromagnetic materials. While much has been said about the SHE in semiconductors and two dimensional electron gases, in this Letter we focus on metals and present a calculation and discussion of the SHE induced by resonant scattering from impurity levels on nonmagnetic impurities in Cu. Whereas recent *ab initio* calculations of the SHE induced by nonmagnetic impurities in metals [2] considered only the skew scattering contribution (except for a recent Letter that appeared after submission of this Letter [3]), we calculate both the skew scattering and side-jump terms in an analytical model which aims at a general description of the main features of the impurity-induced SHE and at a prediction of the best conditions for large effects. By comparing the spin Hall angles due to skew scattering and to side-jump, we can predict the threshold concentration at which the side-jump contribution becomes predominant and can generate very large effects.

The SHE is due to spin-orbit (*S*-*O*) interactions and is associated with off-diagonal terms of the resistivity tensor having opposite signs for spin up and spin down electrons, respectively ρ_{xy} and -ρ_{xy} for $s_z = \pm 1/2$. It can include an intrinsic contribution due to the effect of *S*-*O* interactions on the wave functions of the pure material [4,5] and an extrinsic one resulting from spin-orbit interactions on impurity or defect sites [6,7]. Two mechanisms can contribute to the extrinsic SHE, the skew scattering [6,8] and the scattering with side-jump [7,8].

When the SHE is used to produce a transverse spin current, the maximum yield of the transformation of a longitudinal charge current into a transverse spin current is related to the Spin Hall Angle (SHA), defined as $\Phi_H = \rho_{xy}/\rho_{xx}$ where ρ_{xx} is the diagonal term of the resistivity tensor, i.e., the conventional resistivity for spin $\sigma =\uparrow\downarrow (\pm)$ channels. Consequently Φ_H is the important parameter for practical applications in spintronics. Until 2007, the largest values of Φ_H obtained for pure materials, metals, or semiconductors, had been obtained for Pt ($\Phi_H \approx 0.5\%$) [9,10]. The much larger value of 5% found in 2008 for Au [11] was surprising and has been ascribed to skew scattering by Fe or Pt impurities [12]. An even larger SHE ($\approx 15\%$) was recently obtained by doping Au with Pt impurities [13]. Actually, this brings to mind the large values of Φ_H of a few percent found 30 years ago [14,15] for the SHE induced by nonmagnetic 5*d* impurities in Cu, e.g., $\Phi_H = 2.6\%$, for Cu doped with Ir. This large SHE, with a typical change of sign between the beginning (Lu) and the end (Ir) of the 5*d* series, was ascribed to resonant scattering on the impurity 5*d* states split by the *S*-*O* interaction [14]. Recently measurements by Niimi *et al.* [16] on Cu doped with Ir have confirmed the large value ($\Phi_H \approx 2.1\%$) of the SHE induced by Ir in Cu and confirmed its skew scattering mechanism. Thus, impurity scattering appears as a promising way to obtain the most efficient transformation of charge currents into spin currents by SHE.

Our calculation done for $T = 0$ K is based on a partial wave analysis of the resonant scattering of free electrons from the $j = 5/2$ and $j = 3/2$ states of 5*d* impurities in a metal like Cu, as illustrated in the inset of Fig. 1. From the splitting between the 5/2 and 3/2 levels, $E_{5/2} - E_{3/2} = 5\lambda_d/2$, where λ_d is the impurity 5*d* *S*-*O* constant and by using the classical expression of the phase shift at the Fermi energy ε_F as a function of the resonant level energy E_j, $\cot(\eta_j) = (E_j - \varepsilon_F)/\Delta$ where Δ is the resonance width, we find to first order in λ_d/Δ, $\Delta\eta = \eta_{3/2} - \eta_{5/2} = 5/2 \frac{\lambda_d}{\Delta} \sin^2\eta_2$, where η_2 is the mean phase shift at the Fermi level expressed as a function of the number Z_d of 5*d* electrons on the impurity by Friedel's sum rule, $\eta_2 = (2\eta_{3/2} + 3\eta_{5/2})/5 = \pi Z_d/10$ [17]. By using the canonical expression for the scattering *T* matrix at the Fermi level as a function of the phase shifts [17], and after expanding the states $|j, m_j\rangle$ in terms of $|m, \sigma\rangle$ states and keeping only terms that will contribute to ρ_{xx} and ρ_{xy}, we find the following expression (to first order in λ_d/Δ only non-spin-flip terms contribute),

$$T_{\mathbf{k}'\sigma,\mathbf{k}\sigma} = \frac{2}{n(\varepsilon_F)}\Big[\sigma\frac{\lambda_d}{\Delta}e^{i2\eta_2}\sin^2\eta_2\sum_m mY_2^{m*}(\hat{\mathbf{k}})Y_2^m(\hat{\mathbf{k}}') - 2\sum_{lm}e^{i\eta_l}\sin\eta_l Y_l^{m*}(\hat{\mathbf{k}})Y_l^m(\hat{\mathbf{k}}')\Big], \tag{1}$$

where $\sigma = \pm 1$ and $n(\varepsilon_F)$ is the DOS for one direction of the spin. Note that interchanging $\hat{\mathbf{k}}$ and $\hat{\mathbf{k}}'$ in the first term in the bracket changes its sign, because the factor m in the sum over m becomes $-m$; this is the signature of the antisymmetric scattering. The second term is the usual symmetric term associated with charge scattering, and these phase shifts do not depend on m.

From the antisymmetric part of the scattering probability $W_{\text{antisym}}(k\sigma \to k'\sigma)$, associated with cross terms between the antisymmetric and symmetric parts of the T matrix, we find a term which produces transverse scattering at the Fermi level,

$$\sum_{k'} W_{\text{antisym}}(\mathbf{k}\sigma \to \mathbf{k}'\sigma)g(\mathbf{k}', \sigma) \equiv e\omega_{\text{skew}}(\sigma)\hat{\mathbf{e}}\cdot\hat{\mathbf{k}}\times\hat{\mathbf{z}}, \tag{2}$$

where $\hat{\mathbf{e}}$ is a unit vector along the electric field $\mathbf{E}$, $\hat{\mathbf{z}}$ the spin quantization axis, and we have used the normal out-of-equilibrium distribution function $g(\mathbf{k}', \sigma) \equiv -e\tau_0 v_F E\hat{\mathbf{e}}\cdot\hat{\mathbf{k}}'$

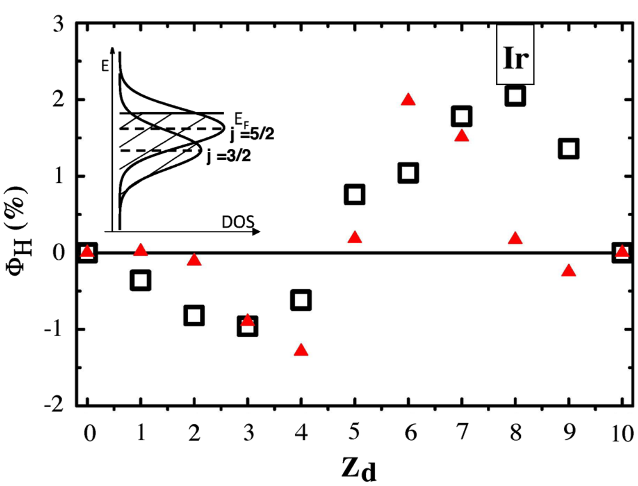

FIG. 1 (color online). Skew scattering (squares) and side-jump (triangles) contributions to the spin Hall angle calculated from Eqs. (11) and (13) as a function of the number of d electrons, Z_d, for $5d$ impurities in Cu. Z_d increases from about 1 for Lu to about 9 for Pt (the calculations in the text for Ir have been done with $Z_d = 8$, see text and Ref. [23]). All other parameters are indicated in the text. The side-jump contribution is calculated for an impurity concentration of 2%. Inset: Density of states (DOS) of a $5d$ virtual bound state with S-O splitting between $j = 3/2$ and $j = 5/2$ states.

where τ_0 is the isotropic relaxation time to arrive at this result. Only the cross terms in the T matrix between the $l = 2$ and $l \pm 1$ terms contribute to Eq. (2). As our aim is a general rather than numerical description we consider only $l = 2$ and $l - 1 = 1$, i.e., we neglect $l + 1 = 3$, and find

$$\omega_{\text{skew}}(\sigma) = -\sigma\frac{6N_i\tau_0 v_F}{\pi\hbar n(\varepsilon_F)}\frac{\lambda_d}{\Delta}\sin(2\eta_2 - \eta_1)\sin^2\eta_2\sin\eta_1, \tag{3}$$

where N_i is the number of impurities.

The contribution to the Hall effect from the side-jump mechanism, i.e., the anomalous velocity, arises from current driven electric dipoles transverse to the flow [18,19]. The expression for the anomalous velocity $\mathbf{w}_a(\mathbf{k}, \sigma)$, Eq. (2.13) in Ref. [19] is,

$$\mathbf{w}_a(\mathbf{k}, \sigma) = \frac{2N_i}{\hbar}\Big[\text{Re}\nabla_{\mathbf{k}}T_{\mathbf{k}\sigma,\mathbf{k}\sigma} + \sum_{\mathbf{k}'\sigma'}\text{P}\frac{1}{(\varepsilon_{k\sigma} - \varepsilon_{k'\sigma'})}\times\text{Re}T^\dagger_{\mathbf{k}\sigma,\mathbf{k}'\sigma'}\nabla_{\mathbf{k}'}T_{\mathbf{k}'\sigma',\mathbf{k}\sigma} - \pi\sum_{\mathbf{k}'\sigma'}\delta(\varepsilon_{k\sigma} - \varepsilon_{k'\sigma'})\text{Im}T^\dagger_{\mathbf{k}\sigma,\mathbf{k}'\sigma'}\nabla_{\mathbf{k}'}T_{\mathbf{k}'\sigma',\mathbf{k}\sigma}\Big]. \tag{4}$$

Only the last term contributes to the Hall effect [19] and by using Eq. (1), we find at $T = 0$ K,

$$\mathbf{w}_a(\mathbf{k}, \sigma)|_{T=0} = \omega_a(\sigma)\hat{\mathbf{k}}\times\hat{\mathbf{z}}, \tag{5}$$

where,

$$\omega_a(\sigma) = \sigma\frac{12N_i}{\pi n(\varepsilon_F)\hbar k_F}\frac{\lambda_d}{\Delta}\frac{E_F}{\Delta}\cos(3\eta_2 - \eta_1)\sin^3\eta_2\sin\eta_1. \tag{6}$$

The *transverse* Hall current comes from terms proportional to $\hat{\mathbf{z}}\times\hat{\mathbf{e}}$; see Ref. [19] for the details. For the anomalous velocity contribution the electric field enters when we use the out-of-equilibrium distribution function $g(\mathbf{k}, \sigma) \equiv -e\tau_0 v_F E\hat{\mathbf{e}}\cdot\hat{\mathbf{k}}$ in the expression for the current $\mathbf{J} = -e\sum_{\mathbf{k},\sigma}[\mathbf{v}_\mathbf{k} + \mathbf{w}_a(\mathbf{k}, \sigma)]f(\mathbf{k}, \sigma)$; see Ref. [19]. When we consider spherical conduction bands and average over Ω_k we find there are two transverse components in $\sigma_H \equiv \sigma_{yx}$. The skew scattering one at $T = 0$ K is,

$$\sigma_{\text{skew}}(\sigma = \uparrow\downarrow) = -\tfrac{1}{3}e^2 n(\varepsilon_F)v_F\tau_0\omega_{\mathbf{skew}}(\sigma), \tag{7}$$

and the anomalous velocity or side-jump contribution is

$$\sigma_{\text{anom}}(\sigma = \uparrow\downarrow) = -\tfrac{2}{3}e^2 n(\varepsilon_F)v_F\tau_0\omega_{\mathbf{a}}(\sigma). \tag{8}$$

The normal conductivity at $T = 0$ K *for each spin channel* is

$$\sigma_N^{-1} = \frac{20\pi\hbar N_i}{n_\sigma e^2 k_F}\sin^2\eta_2, \tag{9}$$

where k_F is the momentum at the Fermi level, and $n_\sigma = \frac{1}{2}n_{\text{total}}$.

By placing the expression for $\omega_{\rm skew}(\sigma)$, see Eq. (3), in the expression for $\sigma_{\rm skew}$, Eq. (7), and dividing by σ_N^2 we find to first order in σ_H/σ_N the skew scattering contribution to the Hall effect at $T = 0$ K is,

$$\rho_{xy}^{\rm skew}(\sigma = \uparrow\downarrow \rightarrow \pm) = \pm \frac{12\pi N_i \hbar}{n_\sigma e^2 k_F} \frac{\lambda_d}{\Delta} \sin(2\eta_2 - \eta_1)\sin^2\eta_2 \sin\eta_1. \quad (10)$$

The Hall angle from the skew scattering is

$$\Phi_H^{\rm skew} = \pm 3/5 \frac{\lambda_d}{\Delta} \sin(2\eta_2 - \eta_1)\sin\eta_1. \quad (11)$$

By placing $\omega_a(\sigma)$, see Eq. (6), in Eq. (8), and dividing by σ_N^2 we find the anomalous velocity contribution to the Hall effect at $T = 0$ K is

$$\rho_{xy}^{\rm anom}(\sigma = \uparrow\downarrow \rightarrow \pm) = \mp \frac{320 N_i \hbar}{n_\sigma e^2 k_F} \frac{c}{z} \frac{\lambda_d}{\Delta} \frac{E_F}{\Delta} \cos(3\eta_2 - \eta_1)\sin^5\eta_2 \sin\eta_1, \quad (12)$$

where c is the impurity concentration, and $z \equiv \frac{n_{\rm total}}{N_s} = \frac{2n_\sigma}{N_s}$, i.e., the number of conduction electrons per lattice site. Finally, the Hall angle from the side jump is

$$\Phi_H^{\rm anom} = \mp 16/\pi \frac{c}{z} \frac{\lambda_d}{\Delta} \frac{E_F}{\Delta} \cos(3\eta_2 - \eta_1)\sin^3\eta_2 \sin\eta_1. \quad (13)$$

Similar calculations can be performed in the presence of crystal field. With completely crystal field split t_{2g} and e_g states, for example, the prefactors of Eqs. (11) and (13) for the t_{2g} states are multiplied by $\frac{1}{3}$ and $\frac{1}{5}$ respectively, and η_2 is replaced by $\eta_{t_{2g}} = \frac{\pi}{6} Z_{t_{2g}}$, where $Z_{t_{2g}}$ is the number of electrons in the t_{2g} states.

We begin the discussion of our results by a glance at the expressions of the spin Hall angle (SHA), Eqs. (11) and (13), for the case without crystal field splittings. $\Phi_H^{\rm skew}$ is proportional to $\frac{\lambda_d}{\Delta}$, and $\Phi_H^{\rm anom} \sim \frac{\lambda_d E_F}{\Delta^2}$. Large effects are thus expected for narrow resonances when the *S-O* splitting induces significant differences in the scattering on $5/2$ and $3/2$ states. In the corresponding expressions for the intrinsic contribution to the SHA [4,5] the denominator Δ is replaced by an energy of the order of the bandwidth, therefore extrinsic effects due to resonant scattering should be generally larger in the usual case where the width of the resonance is smaller than the bandwidth.

The second important feature in the expressions for the Hall angle, arising from the symmetry rules for the SHE, is the interplay between the asymmetric scattering amplitude in the channel l and the symmetric amplitudes in the channels $l \pm 1$. It follows that the spin Hall angle, Eqs. (11) and (13), depends not only on the phase shift η_2 in the resonant channel ($l = 2$) but also on the phase shifts in the nonresonant channels $l \pm 1$. As the scattering in a nonresonant channel is generally weaker than in a resonant one, this selection of cross terms (rarely described in theoretical papers) contributes to the general smallness of the SHA. Here we will take into account η_1 and neglect η_3.

We now focus on the skew scattering. If one supposes that the main contribution to the scattering by $5d$ impurities in noble metals comes from the resonance on their $5d$ states [17], η_2 is much larger then η_1 and, in first approximation, Φ_H is proportional to $\sin 2\eta_2$; see Eq. (11). As $\eta_2 = \frac{\pi Z_d}{10}$, $\sin 2\eta_2$ changes sign from positive to negative between the beginning and end of the $5d$ series as shown in Fig. 1. This change arises from the difference in sign of the asymmetric resonant scattering on $5/2$ and $3/2$ states. This agrees with the observed change of sign for the skew scattering SHE induced by $5d$ impurities in Cu [14,15], $\Phi_H = -1.2\%$ for Lu impurities ($Z_d[\mathrm{Lu}] = 1$) and $\Phi_H = 2.6\%$ for Ir impurities ($Z_d[\mathrm{Ir}] = 8$); the positive SHE for Ir in Cu has been confirmed by recent experiments $\Phi_H = 2.1\%$ [16]. A similar change of sign is also observed for the intrinsic SHE of pure $5d$ metals, which suggests a similar explanation.

Now, we proceed to a quantitative discussion of the Hall angle for Ir-doped Cu for which different type experiments have consistently shown a predominant contribution from skew scattering with reasonably consistent values of Φ_H, $\Phi_H = 2.6\%$ in Ref. [14] and $\Phi_H = 2.1\%$ in Ref. [16]. Typical values of Δ for $5d$ impurities in noble metals are close to 0.5 eV from both experiments [20] and *ab initio* calculations [12,21]. With $\Delta = 0.5$ eV, $\lambda_d \approx 0.25$ eV [22], the more recent experimental value of the SHA for CuIr, $\Phi_H = 2.1\%$, is obtained by introducing $\eta_1 = -4.3°$ in Eq. (11) if we suppose $Z_d = 8$ for Ir impurities in Cu [23]. The decomposition of Eq. (11) into two factors, $\frac{3\lambda_d \sin(2\eta_2 - \eta_1)}{5\Delta} = -0.277$ and $\sin\eta_1 = -0.075$, shows that the interference between the resonant and nonresonant channels induces a significant reduction. The calculation for Ir in Cu can be extended to other $5d$ impurities. For an insight of the variation of Φ_H with Z_d we took the values of Δ and η_1 used for Ir, the *S-O* constants λ_d for the $5d$ series [22] and $\eta_2 = \frac{\pi Z_d}{10}$. As shown on Fig. 1, one obtains a wavy variation reflecting mainly the variation of $\sin(2\eta_2 - \eta_1) \approx \sin 2\eta_2$ modulated by the smooth increase of λ_d.

In contrast to the skew scattering contribution to the SHA, the side-jump one is proportional to the impurity concentration c. The side-jump SHA for $c = 2\%$, calculated with $E_F = 7$ eV for Cu and the values of the parameters λ_d, Δ, η_1 already used for skew scattering, is compared in Fig. 1 with the skew scattering one. For impurities at the beginning and the end of the $5d$ series (Lu, Hf, Ir, Pt) the side-jump contribution at $c = 2\%$ is much smaller than the skew scattering one. This is in agreement with the result that there is only skew scattering for concentrations of a few percent of Ir in Cu [16]. For impurities in the middle of the series, like W, Ta, or Os, concentrations as small as 2% yield side-jump and skew scattering contributions of the same order of magnitude and very large contributions from the side-jump ($\Phi_H^{\rm anom} \gtrsim 10\%$) are expected for concentrations of the order of 10%. This can be compared to the situation of the Anomalous Hall Effect of Gd doped with Lu impurities [24] in which the side-jump contribution exceeds the skew scattering for concentrations above about 6% of Lu. Finally, we point out that in the limit $\eta_1 \ll \eta_2$ the ratio between $\Phi_H^{\rm anom}$ and $\Phi_H^{\rm skew}$ does not depend on the free parameter η_1; this reinforces our conclusion as to the relative sizes of the two contributions.

The above discussion is altered when a crystal field splits the t_{2g} and e_g states. According to the results summarized after Eq. (13), this introduces a change of sign at midway through the filling of the t_{2g} states at $Z_{t_{2g}} = 3$, and a reduction by 3 (skew scattering) and 5 (side-jump) in the amplitudes of the Hall angles. This can change the variation through the 5d series but not really the order of magnitude of the SHA's. For quantitative predictions only an *ab initio* calculation of the scattering phase shifts can lead to realistic results. Our analytical calculation rather aims to predict the main features expected from 5d resonances and to identify the important parameters.

Summarizing, large SHE effects induced by the resonant scattering from impurity states, here d levels, are expected from the combination of: (i) a large S-0 coupling of the impurity states and a narrow resonance, which is the condition to obtain a large asymmetric scattering amplitude in the resonance channel l, and (ii) a large symmetric scattering in the channels ($l \pm 1$). The second condition is not really fulfilled for 5d impurities in Cu so that skew scattering SHA's of only a few percent are expected in agreement with the existing experimental results [14,16]. However, at least for some impurities (W, Ta, Os), large side-jump effects can be expected with Φ_H exceeding 10% for $c \gtrsim 10\%$. Spin Hall angles above 10% would be extremely interesting candidates for generating spin currents without magnetic materials in spintronic devices.

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