Spin Hall angle versus spin diffusion length: Tailored by impurities

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We performed *ab initio* studies to search for materials where a large spin Hall effect caused by skew scattering is accompanied by a long spin diffusion length. Samples with such properties are promising candidates for all-metallic spintronics devices. Here we consider Cu, Au, and Pt hosts which are typical materials used in experiments. In particular, we identified light impurities such as C and N in Au and heavy impurities such as Bi in Cu to meet this criterion. They exhibit a large spin Hall angle ($\alpha > 0.06$) and an appropriate spin diffusion length of about 100 nm. In addition, a pronounced dependence of the spin diffusion length on the scattering properties of the impurity is found for Cu and Au hosts, in contrast to Pt where much smaller variations are obtained.

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I. INTRODUCTION

The spin Hall effect (SHE) is a key issue of spintronics since it allows for the creation of spin currents in nonmagnetic materials avoiding the problem of spin injection from a ferromagnet. Several groups[1–3] succeeded to measure the effect electronically in metallic devices. Particularly high values for the spin Hall angle α of about 0.1 were obtained for Au wires.[3] This angle is defined as the ratio of the spin Hall conductivity σ^s_{yx} to the longitudinal charge conductivity σ_{xx}. Two possible explanations are already proposed for this *gigantic* SHE in Au—a Kondo resonance at Fe impurities[4] and skew scattering at substitutional C impurities.[5] Both theoretical studies elucidate only one aspect of the measurement: is it possible to create a large spin current? However, for practical applications of the SHE the spin diffusion length is as well of crucial importance since it limits the size of a spin Hall device.

In this paper, we present a theoretical study of the extrinsic SHE and the spin diffusion length. Considering the influence of substitutional impurities in Cu, Au, and Pt hosts, we identify favorable systems for future spintronics applications.

II. METHOD

The Hall angle is calculated by means of an *ab initio* relativistic Korringa-Kohn-Rostoker method in combination with the solution of a linearized Boltzmann equation.[5–9] The method is valid in the dilute limit of the impurity concentration c, assuming well separated, noninteracting impurity atoms. In this limit the scattering term, included in the Boltzmann equation,[5] is proportional to the impurity concentration c. As a consequence, the conductivity tensor $\underline{\boldsymbol{\sigma}}$ is inversely proportional to c,

$$\underline{\boldsymbol{\sigma}} \sim \frac{1}{c}. \tag{1}$$

Thus, in the dilute limit the Hall angle is evidently independent on the impurity concentration.[5] If the scattering probabilities are known for a certain dilute alloy, the momentum relaxation τ, as well as, the spin-flip scattering time τ_{sf} can be derived.[9] Both scattering times scale inversely proportional to the impurity concentration and their ratio is independent of c. Using both times the spin diffusion length l_{sf} is obtained according to Valet and Fert.[10] In their theory this quantity

$$l_{sf} = \sqrt{\frac{\lambda \lambda_{sf}}{6}} \tag{2}$$

is determined by the (momentum) mean-free path $\lambda = \tau v_F$ and the spin-flip length $\lambda_{sf} = \tau_{sf} v_F$.[11] Here v_F is the Fermi velocity. In a free-electron model the expression above is equivalent to[12,13]

$$l_{sf} = \frac{\pi}{2k_F^2} \sqrt{\frac{3}{2}} \frac{h}{e^2} \sqrt{\frac{\tau_{sf}}{\tau}} \sigma_{xx}, \tag{3}$$

including the Fermi wave vector k_F. Although both formulas are based on a free-electron model they are nevertheless widely used in the literature. The advantage of our approach is that all parameters of Eq. (3) are calculated from first principles taking into account the anisotropy of the Fermi surface and the scattering. In other words, the used k_F, τ_{sf}, and τ are Fermi-surface averages. Moreover, σ_{xx} is obtained by a full solution of the Boltzmann equation including the scattering-in term.[5]

Our main objective is to identify systems of dilute alloys which exhibit a large spin Hall angle in combination with a long spin diffusion length. Such materials are highly desirable for potential applications of the SHE. In the first part of the next section we present results for the spin Hall angle, the longitudinal and the spin Hall conductivity for the considered hosts with different impurities. Then we discuss the spin diffusion length and estimate a reasonable impurity concentration for experimental samples.

III. RESULTS

A. Extrinsic spin Hall effect

In Table I the calculated values of the spin Hall angle are summarized for several dilute alloys. Since the conductivi-

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TABLE I. The spin Hall angle α as a function of the impurity atom for Cu, Au, and Pt hosts. In addition, the longitudinal charge conductivity σ_{xx} and the transversal spin Hall conductivity σ^s_{yx} are shown at an impurity concentration of 1 at. %.

	Cu			Au			Pt		
	α 10^{-3}	σ_{xx} $(\mu\Omega\ \mathrm{cm})^{-1}$	σ^s_{yx} $(10^3\ \mu\Omega\ \mathrm{cm})^{-1}$	α 10^{-3}	σ_{xx} $(\mu\Omega\ \mathrm{cm})^{-1}$	σ^s_{yx} $(10^3\ \mu\Omega\ \mathrm{cm})^{-1}$	α 10^{-3}	σ_{xx} $(\mu\Omega\ \mathrm{cm})^{-1}$	σ^s_{yx} $(10^3\ \mu\Omega\ \mathrm{cm})^{-1}$
Li	2.3	1.22	2.8	7.2	0.60	4.3	−2.3	0.29	−0.65
C	6.6	0.16	1.0	96.0	0.12	12.0	−2.8	0.26	−0.73
N	7.0	0.11	0.75	64.0	0.08	5.3	11.0	0.19	2.2
Mg	−1.5	1.57	−2.3	−8.2	0.67	−5.5	−3.8	0.29	−1.1
Cu				−0.44	2.96	−1.3	−5.2	0.42	−2.2
Ag	0.26	30.2	7.9	4.8	3.47	17.0	−2.7	0.48	−1.3
Pt	27.0	0.51	13.6	10.0	0.93	9.0			
Au	7.8	2.37	18.5				−1.1	0.83	−0.94
Bi	81.0	0.22	18.1	14.0	0.13	1.9	−1.2	0.25	−0.29

ties (longitudinal and transverse) are inversely proportional to the impurity concentration in the dilute limit, α is independent of the number of impurities. The conductivities are given for a fixed concentration of 1 at. %. As it was discussed already in Ref. 5, C and N impurities in a Au host provide a very high spin Hall angle. The values are comparable to α found experimentally.[3] Smaller values obtained by other experimental groups[14,15] can be explained by the presence of impurities such as Cu and Ag in Au samples. In addition, we find a very large spin Hall angle for Bi impurities in Cu.

The high α values in a Au host with light impurities are a quite counterintuitive result. Nevertheless, it can be explained by means of the scattering phase shifts at the impurity site. Such an approach is justified by the topology of the Fermi surface of Au and Cu,[8] which allows to use a spherical band approximation. To estimate the strength of the SHE, we can consider the differences of the scattering phase shifts $\delta_j(E_F)$ at the Fermi energy E_F for the total angular momenta $j=l\pm 1/2$ $(l>0)$ of an impurity atom and the host atoms. Moreover, for the considered systems it is sufficient to discuss just $l=1$ since scattering of p electrons dominates the effect. Thus, we can restrict our considerations to the p phase shift differences $\Delta\delta_{1/2}(E_F)=\delta^{imp}_{1/2}(E_F)-\delta^{host}_{1/2}(E_F)$ and $\Delta\delta_{3/2}(E_F)=\delta^{imp}_{3/2}(E_F)-\delta^{host}_{3/2}(E_F)$. They account for the scattering strength and for the spin-orbit interaction of the impurity, as well as, of the host atom. As it is well known, the splitting of the $p_{1/2}$ and $p_{3/2}$ levels is proportional to the spin-orbit interaction. In the same spirit the difference of $\Delta\delta_{3/2}(E_F)$ and $\Delta\delta_{1/2}(E_F)$ is a measure of the relevant spin-orbit splitting at the impurity site including both the spin-orbit interaction of the host and of the impurity atom. This difference of the p phase shifts is responsible for the left/right asymmetry of scattering for $s=1/2$ and $s=-1/2$ electrons.

Figures 1(a) and 1(b) show $\Delta\delta_{3/2}(E_F)$ and $\Delta\delta_{1/2}(E_F)$ for Cu and Au hosts, respectively. It turns out that the differences of the scattering phase shifts $\Delta\delta_{3/2}(E_F)-\Delta\delta_{1/2}(E_F)$ in Au are particularly large for light impurities. In combination with a strong scattering indicated by large absolute values of $\Delta\delta_{1/2}(E_F)$ and $\Delta\delta_{3/2}(E_F)$, a large spin Hall conductivity is expected. From this perspective, C and N in Au as well as Bi in Cu are the best candidates for a large extrinsic SHE. They are strong p scatterers with large differences of spin-orbit interaction between host and impurity atoms. The other considered impurities are mainly s or d (for example, Pt) scatterers which leads to small phase shifts in the $l=1$ channel. The picture is confirmed by C and N impurities in Cu. Al-

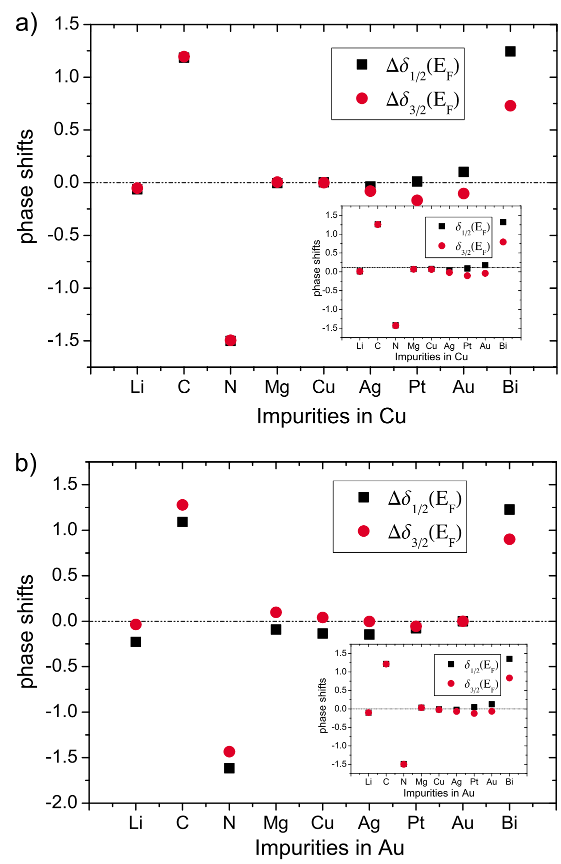

FIG. 1. (Color online) The differences between the scattering phase shifts of an impurity and the host atom for the levels $j=l\pm 1/2$ with $l=1$ (a) Cu host and (b) Au host. As insets the phase shifts of the impurity atoms only are shown.

though they are as well strong p scatterers, the SHE is comparable small since the spin-orbit interaction is negligible for these impurities and for the Cu host. Only heavy p scatterers such as Bi can produce a large α in Cu because of the strong spin-orbit interaction of Bi with respect to Cu. However, the Hall angle for Bi impurities in Au is smaller since the spin-orbit interaction of Bi and Au compensate each other partly. The difference of the phase shifts of Au and Bi atoms, the effective splitting $|\Delta\delta_{3/2}(E_F)-\Delta\delta_{1/2}(E_F)|$ is smaller in Au than in Cu [compare Figs. 1(a) and 1(b)].

To highlight the different nature of the large SHE in Cu with Bi impurities and in Au with C and N impurities, we present as insets of Fig. 1 the phase shifts $\delta_{1/2}^{imp}(E_F)$ and $\delta_{3/2}^{imp}(E_F)$ of the impurity atoms in the considered hosts. Here the phase shift differences between the $p_{1/2}$ and $p_{3/2}$ levels are a measure of the spin-orbit coupling strength of the impurity atoms, only. The light elements, such as C and N, show negligible spin-orbit coupling while for Pt, Au, and Bi the difference between the $p_{1/2}$ and $p_{3/2}$ level is clearly visible. With that knowledge the origin of the large SHE in the Cu(Bi) alloy can clearly be attributed to the spin-orbit coupling induced by Bi impurities. Whereas, for C and N in a Au host, the spin-orbit coupling at the Au atoms is responsible for the large SHE.

The strong variation in α by 2 orders of magnitude as a function of the impurity atom in Cu and Au is attributed to the relatively weak spin-orbit interaction of the hosts. In contrast, the dominant spin-orbit interaction of Pt suppresses the impurity-specific variations. It results in Hall angles which differ by 1 order of magnitude only. In the Pt-based alloys the effect is mainly provided by the host wave functions showing a much stronger spin-mixed character.[8] The spherical band approximation is not valid for Pt because of its complicated Fermi surface[8] with mainly d character of the wave functions at the Fermi level. Therefore, such a simple analysis as it was performed above for Au and Cu is impossible for Pt.

To summarize, C and N impurities in Au and Bi impurities in Cu are promising candidates for spintronics applications. However, the preparation of well-defined concentrations of C and N atoms in Au can be rather difficult. In this respect, the dilute Cu(Bi) alloy is preferable since it is known to exist. In addition, a Pt host with N impurities would be as well of interest since we predict a reasonable high spin Hall angle.

An important point to note is that the strength of the SHE in Cu- and Pt-based alloys is of comparable order. Of course, here we neglect the intrinsic SHE which should be much stronger for Pt than for Cu since for Pt several bands cross the Fermi level in contrast to Cu (and Au) with only one band.[8,16] Nevertheless, our calculated extrinsic spin Hall conductivities are of comparable magnitude to the intrinsic contribution.[16] Therefore, the latter one cannot drastically change α in Pt at low impurity concentrations. Thus, Cu based alloys are equally suited to measure the SHE as Pt based alloys.

B. Spin diffusion length

Now the question arises if it is possible to measure the obtained Hall angles in a real experiment where the system size is limited by the spin diffusion length. Equation (3) shows that for a given host material, with a fixed longitudinal conductivity, the ratio of the spin-flip scattering time and the momentum relaxation time determines l_{sf}. Figure 2 summarizes this ratio which is, in the used approximation of dilute alloys, independent on the impurity concentration. For Pt a very weak influence of the impurity character is found. It is different for Cu where the ratio changes over several orders of magnitude. For Au the character of the impurity atom influences the ratio by only 1 order of magnitude. This can be explained by the relatively weak spin mixing of the Au electron states in comparison to the strong mixing induced in Pt.[8] For a Cu host the spin-orbit coupling is weak and the main contribution arises from the spin-orbit coupling of the impurity atom. In contrast, the spin-orbit interaction in the Pt based alloys is essentially provided by the host.

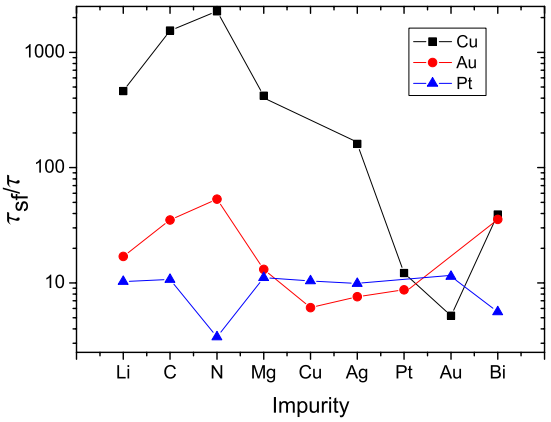

FIG. 2. (Color online) The ratio of the spin-flip scattering time τ_{sf} and the momentum relaxation time τ for different impurity atoms in Cu, Au, and Pt hosts.

C. SHE vs spin diffusion length

For the calculation of the spin diffusion length the longitudinal charge conductivity has to be considered as well. However, the conductivity is, in the dilute limit, inversely proportional to the impurity concentration which is not known in experiments. For this purpose the impurity concentration was deduced from experimental data for σ_{xx} published in relation to the SHE and the spin diffusion length.[14,17,18] We estimated the concentration c_{exp} of a specific impurity by assuming that only one type of impurity atom exists in the sample,

$$c_{exp} = \frac{\sigma_{xx}^{calc}}{\sigma_{xx}^{exp}} c_0. \tag{4}$$

Based on these concentrations, a spin diffusion length $l_{sf}(c_{exp})$ for experimental setups is obtained and summarized in Table II.

For all considered impurities in Pt the value of l_{sf} is nearly the same of about 7 nm, which is in reasonable agreement with experimentally found values in the range of 7–14 nm.[18,19] For a Au host moderate variations with respect to impurities occur between 42 and 120 nm, which is also in

TABLE II. The spin diffusion length l_{sf} calculated from Eq. (3) with a fixed σ_{xx} for different impurities in Cu, Au, and Pt hosts. The experimental concentration is estimated using Eq. (4). The used experimental values for the conductivity are $\sigma_{xx}(\mathrm{Cu})=0.7\ (\mu\Omega\ \mathrm{cm})^{-1}$, $\sigma_{xx}(\mathrm{Au})=0.48\ (\mu\Omega\ \mathrm{cm})^{-1}$, and $\sigma_{xx}(\mathrm{Pt})=0.08\ (\mu\Omega\ \mathrm{cm})^{-1}$ (Refs. 14, 17, and 18).

	Cu		Au		Pt	
	$l_{sf}(c_{exp})$ (nm)	c_{exp} at. %	$l_{sf}(c_{exp})$ (nm)	c_{exp} at. %	$l_{sf}(c_{exp})$ (nm)	c_{exp} at. %
Li	410	1.7	70	1.3	7.3	3.6
C	750	0.22	100	0.25	7.5	3.3
N	910	0.15	120	0.17	4.2	2.4
Mg	390	2.2	62	1.4	7.6	3.7
Cu			42	6.2	7.4	5.2
Ag	240	43.1	47	7.2	7.2	6.0
Pt	67	0.73	50	1.9		
Au	44	3.4			7.7	10.4
Bi	120	0.32	100	0.28	5.4	3.1

good agreement with experimentally found values of $l_{sf}=35-100$ nm.[3,20–24] The spin diffusion length in Cu varies between 44 nm for Au and 910 nm for N impurities. Experimentally reported values are in the range of 200–1000 nm.[2,17,21,23–25] Interestingly, the systems with the highest α in Au (C and N impurities) have also the longest l_{sf} which is provided by relatively small longitudinal conductivities. These impurities induce large potential changes which lead to a high resistivity already at low concentrations. The same holds for Bi impurities in Cu and Au lattices. The measured conductivity is reproduced with a small concentration of about 0.3 at. %. At such low concentrations the spin diffusion length is reasonably long ($l_{sf}\approx 100$ nm) for experimental requirements.

Combining the results for the spin Hall conductivity and the estimation of the spin diffusion length, the systems with N and C impurities in Au are good candidates for spintronics applications. In addition, Bi impurities in Cu provide both, a long spin diffusion length and a large spin Hall angle. On the other hand, there is no advantage to use Pt instead of Cu as a host material for spin Hall measurements. The spin diffusion length l_{sf} in Cu is more than one order of magnitude larger than in Pt which implies that Cu should be used instead of Pt.

IV. CONCLUSION

We have investigated the influence of several substitutional defects in Cu, Au, and Pt bulk crystals on the spin Hall angle and the spin diffusion length. It is shown that tailoring these quantities is possible for a light hosts such as Cu. Whereas, the role of impurities is minor for materials with strong intrinsic spin-orbit interaction like Pt. C and N impurities in a Au host and Bi impurities in a Cu host are identified as best candidates for all-metallic spin-current generation. At impurity concentrations feasible in experiment they show a spin Hall angle of 0.096, 0.064, and 0.081 combined with a spin diffusion length of 101 nm, 124 nm, and 120 nm, respectively. In addition, we find that Cu is a more attractive host material in comparison to Pt for the analysis of the skew scattering contribution to the SHE since Cu exhibits an extrinsic SHE comparable to Pt-based alloys and has at the same time a much longer spin diffusion length.

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