



Tauber, K., Hoenemann, A., Fedorov, D. V., Gradhand, M., & Mertig, I. (2015). Enhancement of the anomalous Hall effect in ternary alloys. *Physical Review B: Condensed Matter and Materials Physics*, 91(22), Article 220404. <https://doi.org/10.1103/PhysRevB.91.220404>

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Enhancement of the anomalous Hall effect in ternary alloys

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(Received 2 April 2015; published 8 June 2015)

We consider ternary alloys of the composition $\text{Cu}(\text{Mn}_{1-w}T_w)$, where T corresponds to different nonmagnetic impurities. As was discovered by Fert *et al.* [*J. Magn. Magn. Mater.* **24**, 231 (1981)], the anomalous Hall effect (AHE) in the binary $\text{Cu}(\text{Mn})$ alloy can be significantly enhanced by means of codoping using $5d$ impurities. Moreover, they attempted to quantify the spin Hall effect (SHE) in $\text{Cu}(T)$ binary alloys via the AHE measured in the related ternary alloys. Here, we present a theoretical study serving as a detailed background of the experimental findings by clarifying the conditions required for a maximal enhancement of the AHE as well as the relations between both Hall effects. Based on the proposed approach, we perform first-principles calculations for several $\text{Cu}(\text{Mn}_{1-w}T_w)$ [$T = \text{Au}, \text{Bi}, \text{Ir}, \text{Lu}, \text{Sb}, \text{or Ta}$] alloys, which are underpinned by theoretical investigations via Matthiessen's rule.

DOI: 10.1103/PhysRevB.91.220404

PACS number(s): 71.15.Rf, 72.25.Ba, 75.76.+j, 85.75.-d

In 1879, Edwin Hall discovered his famous effect, where moving electrons are deflected transversal to an applied magnetic field due to the Lorentz force [1]. Two years later, he reported [2] that in ferromagnetic materials this effect can be much larger than in nonmagnetic conductors. There, the Hall resistivity [3]

$$\rho_{yx} = R_H H_z = R_0 H_z + \underbrace{R_s M_z(H_z)}_{\rho_{yx}^E} \quad (1)$$

is not only directly proportional to the applied perpendicular field H_z , but includes also the so-called extraordinary or anomalous Hall resistivity ρ_{yx}^E , which is proportional to the magnetization M_z . Consequently, the Hall coefficient R_H contains both the ordinary contribution R_0 and the anomalous contribution $R_s \chi$, where $\chi = M_z(H_z)/H_z$ is the magnetic susceptibility. The second term on the right-hand side (r.h.s.) of Eq. (1) describes the phenomenon known as extraordinary or anomalous Hall effect (AHE). The AHE can be caused by an intrinsic mechanism related to the Berry curvature [4,5] or by two extrinsic mechanisms [3]. They are known as skew scattering [6,7] and side jump [8] and refer to spin-dependent scattering at impurities due to spin-orbit coupling (SOC). The same mechanisms also drive the spin Hall effect (SHE) [9–11], which describes the conversion of a charge current into a transverse spin current. The beauty of this phenomenon is that it also provides a useful tool for the detection of spin currents. Namely, via the inverse spin Hall effect (ISHE), a spin current causes a transverse charge current, which can be detected as Hall voltage [12].

Ferromagnetic materials have a spin imbalance in the number of electrons, whereby any longitudinal charge transfer is accompanied by a longitudinal spin current. Consequently, the SHE and ISHE coexist leading to a spin accumulation and transverse voltage, respectively. The created voltage corresponds to the AHE. In the dilute alloys considered in this Rapid Communication the skew scattering dominates the effects [13–16].

The aim of the presented work is to elucidate the conditions required for the enhancement of the AHE in ternary alloys by means of codoping. In contrast to our recent study [17], where the efficiency of the SHE could not be enhanced in a ternary alloy with respect to the constituent binary alloys, here it will be demonstrated that it is possible for the AHE. We consider ternary alloys of the composition $\text{Cu}(\text{Mn}_{1-w}T_w)$, which means a Cu host with magnetic Mn impurities and nonmagnetic impurities of $T = \text{Au}, \text{Bi}, \text{Ir}, \text{Lu}, \text{Sb}, \text{or Ta}$. The quantity $w \in [0, 1]$ describes the weighting factor between the impurities Mn and T [17], while the total impurity concentration is fixed to 1 at.%. Alloys of that composition were investigated experimentally by Fert *et al.* [18]. Particularly, they used measurements of the AHE in the ternary alloys to determine the strength of the SHE in the corresponding binary alloy $\text{Cu}(T)$. Their study showed at first that the skew scattering in $\text{Cu}(\text{Mn})$ alloys is negligible and therefore there is no significant anomalous contribution to the Hall effect. Nevertheless, the Mn impurities break the time-reversal symmetry, which leads to a longitudinal spin-polarized current as shown in Fig. 1(a). Then, the authors of Ref. [18] found that the skew scattering appears if nonmagnetic impurities with strong SOC are added to the $\text{Cu}(\text{Mn})$ alloy. These impurities alone would create a pure SHE in copper as visualized in Fig. 1(b). Combining

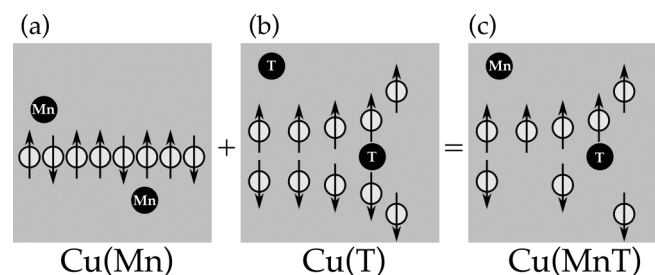


FIG. 1. (a) Longitudinal spin-polarized current due to magnetic Mn impurities. (b) Spin Hall effect due to skew scattering caused by nonmagnetic impurities with strong spin-orbit interaction. (c) Skew scattering for a spin-polarized current leading to the anomalous Hall effect.

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both, the resulting AHE in the considered ternary alloys can be understood as the skew scattering acting on a spin-polarized current. This is illustrated by Fig. 1(c). The investigation of the SHE via the AHE measurements performed in Ref. [18] was possible, since the skew scattering is assumed to be provided by the nonmagnetic impurities only. In the following, a comprehensive formalism is derived, which verifies the proportionality of the efficiencies between the SHE and AHE under this assumption.

Within the two-current model, the charge and spin current densities are given by

$$\begin{aligned} \mathbf{j} &= \mathbf{j}^+ + \mathbf{j}^- = (\hat{\sigma}^+ + \hat{\sigma}^-)\mathbf{E} = \hat{\sigma}\mathbf{E} = (\hat{\rho})^{-1}\mathbf{E}, \\ \mathbf{j}^s &= \mathbf{j}^+ - \mathbf{j}^- = (\hat{\sigma}^+ - \hat{\sigma}^-)\mathbf{E} = \hat{\sigma}^s\mathbf{E} = (\hat{\rho}^s)^{-1}\mathbf{E}, \end{aligned} \quad (2)$$

where \mathbf{j}^\pm are the spin-resolved current densities. Here, $\hat{\sigma}$ and $\hat{\sigma}^s$ are the charge and spin conductivity, respectively, while $\hat{\rho}$ and $\hat{\rho}^s$ are the corresponding resistivities. Under the assumption of negligible skew scattering at Mn impurities in copper one can use $\rho_{yx}^+ = -\rho_{yx}^-$, since it originates from nonmagnetic impurities only. Accordingly, the anomalous contribution to the Hall resistivity can be obtained as [18]

$$\rho_{yx}^E = \frac{j_x^s}{j_x} \rho_{yx}^+ = \frac{j_x^+ - j_x^-}{j_x^+ + j_x^-} \rho_{yx}^+, \quad (3)$$

using Eq. (2) up to first order in ρ_{yx}/ρ_{xx} [19]. Equation (3) implies that the anomalous Hall resistivity results from the product of the polarization of the spin-dependent currents caused by Mn impurities and the Hall resistivity ρ_{yx}^+ , which describes the skew scattering at the nonmagnetic impurities. This leads to a strong correlation between the SHE and AHE in the Cu(*T*) and Cu(Mn *T*) alloys, respectively. However, all quantities involved in the equation above depend on the weighting factor, if the total impurity concentration is fixed. This complicates a quantitative comparison of the two phenomena needed to attain optimal conditions delivered by the codoping.

In what follows, we solve this problem by deriving an expression for the maximal enhancement of the AHE in ternary alloys as a function of the weighting factor *w*. For this aim, let us compare the efficiencies of both effects. They are given by the anomalous Hall angle (AHA) and spin Hall angle (SHA):

$$\alpha_{\text{AHE}} = \frac{\sigma_{yx}}{\sigma_{xx}} \quad \text{and} \quad \alpha_{\text{SHE}} = \frac{\sigma_{yx}^s}{\sigma_{xx}}, \quad (4)$$

$$\alpha_{\text{AHE}}^{\text{Mn}T}(w) = \frac{\sigma_{yx}^{\text{Mn}T}}{\sigma_{xx}^{\text{Mn}T}} = - \frac{[(1-w)\rho_{yx}^{+\text{Mn}} + w\rho_{yx}^{+T}] \frac{\rho_{xx}^{-\text{Mn}T}}{\rho_{xx}^{+\text{Mn}T}} + [(1-w)\rho_{yx}^{-\text{Mn}} + w\rho_{yx}^{-T}] \frac{\rho_{xx}^{+\text{Mn}T}}{\rho_{xx}^{-\text{Mn}T}}}{\rho_{xx}^{+\text{Mn}T} + \rho_{xx}^{-\text{Mn}T}}. \quad (8)$$

Since *T* are nonmagnetic impurities, the identities $\rho_{xx}^{+T} = \rho_{xx}^{-T}$ and $\rho_{yx}^{+T} = -\rho_{yx}^{-T}$ are valid. For further simplification of Eq. (8), we set $\rho_{yx}^{+\text{Mn}} = \rho_{yx}^{-\text{Mn}} = 0$, according to the assumption of negligible skew scattering at Mn impurities, and obtain

$$\alpha_{\text{AHE}}^{\text{Mn}T} = - \frac{w\rho_{yx}^{+T} \left(\frac{\rho_{xx}^{-\text{Mn}T}}{\rho_{xx}^{+\text{Mn}T}} - \frac{\rho_{xx}^{+\text{Mn}T}}{\rho_{xx}^{-\text{Mn}T}} \right)}{\rho_{xx}^{+\text{Mn}T} + \rho_{xx}^{-\text{Mn}T}}. \quad (9)$$

The used assumption is supported by the spin-dependent resistivities of the binary alloys shown in Table I, where ρ_{yx}^\pm of the Cu(Mn) alloy is much smaller than for the other binary alloys. This has an exception for the Cu(Au) alloy showing comparable results, which should be kept in mind. It is also clear that this assumption leads to incorrect results near $w = 0$, where the efficiency according to Eq. (9) goes to zero instead of $\alpha_{\text{AHE}}^{\text{Mn}} = 0.0005$ obtained from first principles.

respectively. A comparison of these quantities is not trivial, since the efficiency for the ternary alloy Cu(Mn_{1-*w*}*T*_{*w*}) changes with the weighting factor. Therefore α_{AHE} has to be calculated for the whole range of $w \in (0, 1)$. In that respect, it is convenient to apply Matthiessen's rule. Here, the scattering at both types of impurities is considered independently and only the transport properties of the constituent binary alloys are needed for the description of the ternary alloy [20]. For a cubic host system with *z* as the spin quantization axis, the spin-dependent conductivity has the following tensor structure:

$$\hat{\sigma}^\pm = \begin{pmatrix} \sigma_{xx}^\pm & -\sigma_{yx}^\pm & 0 \\ \sigma_{yx}^\pm & \sigma_{xx}^\pm & 0 \\ 0 & 0 & \sigma_{zz}^\pm \end{pmatrix} = (\hat{\rho}^\pm)^{-1}. \quad (5)$$

Within our work, these quantities are obtained via the solution of the linearized Boltzmann equation [21,22]. This is based on the electronic structure of the host and impurity system calculated using the relativistic Korringa-Kohn-Rostoker method [23].

The off-diagonal components of the conductivity or resistivity tensor are normally much smaller than the diagonal ones. Therefore one can use the following approximations:

$$\begin{aligned} \sigma_{xx}^\pm &= \frac{\rho_{xx}^\pm}{(\rho_{xx}^\pm)^2 + (\rho_{yx}^\pm)^2} \approx \frac{1}{\rho_{xx}^\pm}, \\ \sigma_{yx}^\pm &= -\frac{\rho_{yx}^\pm}{(\rho_{xx}^\pm)^2 + (\rho_{yx}^\pm)^2} \approx -\frac{\rho_{yx}^\pm}{(\rho_{xx}^\pm)^2}. \end{aligned} \quad (6)$$

Using Matthiessen's rule as a further approximation for the ternary alloys, the resistivity tensor is built by the linear combination of the resistivities related to the two constituent binary alloys [20]:

$$\hat{\rho}^{\pm\text{Mn}T} = (1-w)\hat{\rho}^{\pm\text{Mn}} + w\hat{\rho}^{\pm T}. \quad (7)$$

The corresponding spin-dependent conductivities of the ternary alloy are obtained from Eq. (6). Finally, the total charge conductivity can be calculated via the two-current model of Eq. (2) and one obtains the anomalous Hall angle according to Eq. (4) as

TABLE I. Columns 1–4: Spin-resolved longitudinal and transversal resistivities (in $\mu\Omega\text{cm}$) of Cu(Mn) and Cu(T) with $T = \text{Au, Bi, Ir, Lu, Sb, or Ta}$. Column 5: Spin Hall angle (in 10^{-2}) for Cu(T) alloys. Column 6: Experimental results for the spin Hall angle by Fert *et al.* [18]. Column 7: Anomalous Hall angle (in 10^{-2}) in Cu(Mn T) at the extremum position. Column 8: Spin Hall angle (in 10^{-2}) for Cu(T) alloys calculated via Eq. (13).

Col.	1	2	3	4	5	6	7	8
Imp.	ρ_{xx}^+	ρ_{xx}^-	ρ_{yx}^+	ρ_{yx}^-	α_{SHE}^T	$\alpha_{\text{SHE}}^T(\text{exp.})$	$\alpha_{\text{AHE}}^{\text{Mn}T}(w_{\text{max}})$	α_{SHE}^T via w_{max}
Mn	0.78	15.83	-0.0004	0.0155	–	–	–	–
Au	0.85	0.85	-0.0067	0.0067	0.79	2.7	0.51	0.80
Bi	8.90	8.90	-0.7276	0.7276	8.17	–	4.92	7.73
Ir	12.36	12.36	-0.4413	0.4413	3.57	5.2	2.28	3.58
Lu	15.16	15.16	-0.3978	0.3978	2.62	-2.4	1.64	2.59
Sb	8.86	8.86	-0.2323	0.2323	2.62	2.3	1.58	2.48
Ta	30.29	30.29	-0.4532	0.4532	1.50	1.4	0.94	1.48

It is useful to build the ratio of the AHA in Eq. (9) and the SHA for the Cu(T) alloy, $\alpha_{\text{SHE}}^T = -\rho_{yx}^{+T}/\rho_{xx}^{+T}$, as

$$\frac{\alpha_{\text{AHE}}^{\text{Mn}T}}{\alpha_{\text{SHE}}^T} = \frac{w\rho_{xx}^{+T} \left[\frac{(1-w)\rho_{xx}^{-\text{Mn}} + w\rho_{xx}^{+T}}{(1-w)\rho_{xx}^{+\text{Mn}} + w\rho_{xx}^{+T}} - \frac{(1-w)\rho_{xx}^{+\text{Mn}} + w\rho_{xx}^{+T}}{(1-w)\rho_{xx}^{-\text{Mn}} + w\rho_{xx}^{+T}} \right]}{(1-w)(\rho_{xx}^{+\text{Mn}} + \rho_{xx}^{-\text{Mn}}) + 2w\rho_{xx}^{+T}}, \quad (10)$$

since this quantity depends only on the longitudinal transport properties of the Cu(Mn) and Cu(T) alloys. According to the aim of our work, the extremum of this ratio is of great interest. Its position in terms of the weighting factor can be obtained as

$$w_{\text{max}} = \frac{\sqrt{\rho_{xx}^{+\text{Mn}} \rho_{xx}^{-\text{Mn}}}}{\rho_{xx}^{+T} + \sqrt{\rho_{xx}^{+\text{Mn}} \rho_{xx}^{-\text{Mn}}}}. \quad (11)$$

This implies with $\rho_{xx}^{+T} = \sqrt{\rho_{xx}^{+\text{Mn}} \rho_{xx}^{-\text{Mn}}}$ the extremum occurs for a ternary alloy with equal weighting, $w = 0.5$, for both impurities. For $\rho_{xx}^{+T} > \sqrt{\rho_{xx}^{+\text{Mn}} \rho_{xx}^{-\text{Mn}}}$, the extremum shifts to the left ($w_{\text{max}} < 0.5$) and otherwise to the right ($w_{\text{max}} > 0.5$). Remarkably, the extremal value of Eq. (10), given by

$$\frac{\alpha_{\text{AHE}}^{\text{Mn}T}(w_{\text{max}})}{\alpha_{\text{SHE}}^T} = \frac{\sqrt{\rho_{xx}^{-\text{Mn}}} - \sqrt{\rho_{xx}^{+\text{Mn}}}}{\sqrt{\rho_{xx}^{-\text{Mn}}} + \sqrt{\rho_{xx}^{+\text{Mn}}}} \equiv C^{\text{Mn}}, \quad (12)$$

depends on the transport properties of the Cu(Mn) alloy only. In other words, at the extremum the AHA of the Cu(Mn T) alloy is directly related to the SHA of the Cu(T) alloy

$$\alpha_{\text{AHE}}^{\text{Mn}T}(w_{\text{max}}) = C^{\text{Mn}} \alpha_{\text{SHE}}^T \quad (13)$$

via the proportionality constant C^{Mn} defined by Eq. (12). It is obtained as 0.637 for a Cu host with Mn impurities using our first-principles calculations.

Finally, we confirm the accuracy of the presented theoretical model using a comparison to the results of full *ab initio* transport calculations, which are shown in Fig. 2 and Table I. First of all, the simplification from Eq. (8) to Eq. (9) is verified. For this purpose, the ratio $\alpha_{\text{AHE}}^{\text{Mn}T}/\alpha_{\text{SHE}}^T$ is shown in Fig. 2, where the quantity $\alpha_{\text{AHE}}^{\text{Mn}T}$ is calculated via Eqs. (8) and (9) for the solid and dashed lines, respectively. Both curves provide a good agreement except the region close to $w = 0$, where the solid lines converge to the AHA of the Cu(Mn) alloy while the dashed lines go to zero. The deviations are most pronounced for the Cu(MnAu) alloys, since the strength of the skew scattering is comparable for Au and Mn impurities

in copper, as was mentioned above. Figure 2 includes the results of full *ab initio* calculations for ternary alloys. They are obtained using the approach of Ref. [17]. It is based on the incoherent superposition of the microscopic transition probabilities for each scattering process. Evidently, the corresponding values marked by dots agree very well with the model predictions.

Furthermore, we compare our calculated SHAs in column 5 of Table I with the experimental results of Fert *et al.* [18] in column 6. Although for the Cu(Lu) alloy the opposite sign is obtained, the absolute value fits nicely for Lu, Sb, and Ta impurities. For the Cu(Ir) alloy the deviation is much larger, but still satisfactory. Only for the Au impurities a strong disagreement is observed. The deviations are caused by different approaches used to obtain the SHA via the AHE. In Ref. [18], the Hall coefficient $R_H = R_0 + R_s \chi$ given by Eq. (1) was first approximated by the function $R_H = R_0 + A/T$, assuming for the magnetic susceptibility the behavior $\chi \propto 1/T$ in the low field limit. The coefficient A was obtained via a linear fit of the observed R_H as a function of the inverse temperature. It was performed for a few Cu(Mn $_x$ T $_y$) alloys with different fractions x and y . Within a simplified resonant scattering model used by Fert *et al.* [18], A/x can be expressed as a product of α_{SHE}^T and a function of y/x . This allowed the authors of Ref. [18] to derive the spin Hall angle by a second fit of the experimental data. In addition, a few other parameters entered their analysis, which can also influence the final values of α_{SHE}^T derived in Ref. [18] for low temperature and shown in column 6 of Table I. Therefore the accuracy of this procedure may vary significantly for different systems. By contrast, our technique for determination of the SHA via the AHA is based on the knowledge of the spin-resolved resistivities for the binary alloy with magnetic impurities only. Its high reliability is well demonstrated by Table I, comparing columns 5 and 8, where the latter provides the SHAs obtained with the presented technique based on Eq. (13).

To estimate the enhancement of the AHE for the considered ternary alloys, *ab initio* transport calculations were performed at the predicted extremum positions given in Fig 2. The corresponding AHAs were calculated and shown in column 7. Dividing these quantities by the SHA of the related binary alloys Cu(T), presented in column 5, values between 0.6 and 0.65 are obtained. This agrees well with the constant

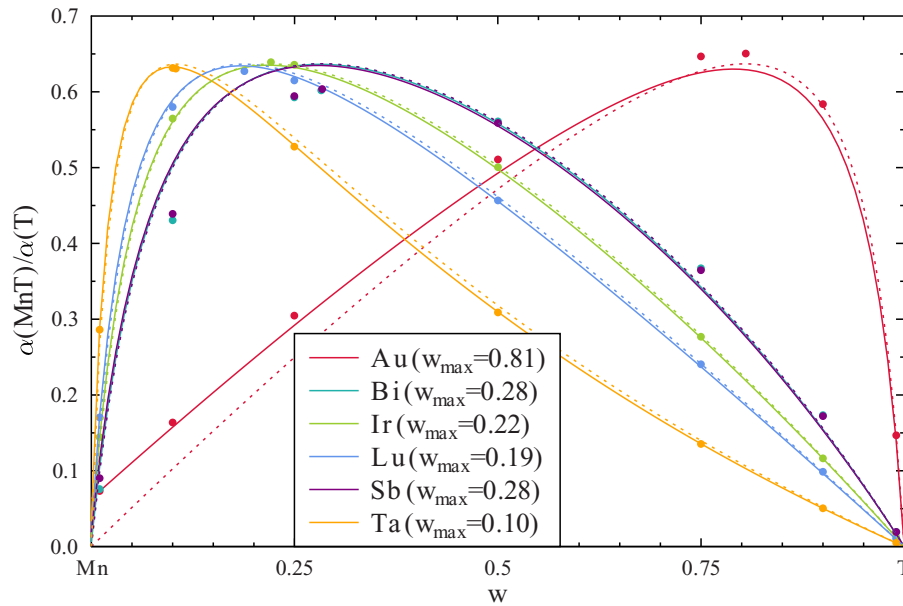


FIG. 2. (Color online) The anomalous Hall angle of the ternary alloy $\text{Cu}(\text{Mn}_{1-w}\text{T}_w)$ divided by the spin Hall angle of the corresponding $\text{Cu}(\text{T})$ alloy, where the nonmagnetic impurity is either Au, Bi, Ir, Lu, Sb, or Ta. The solid lines provide the results obtained with Matthiessen's rule via Eq. (8) and the dashed lines with the approximation of Eq. (9). The dots are results of full *ab initio* transport calculations performed according to the approach of Ref. [17].

$C^{\text{Mn}} = 0.637$ of Eq. (13) and is present in Fig. 2 as almost constant height of the maxima for the different nonmagnetic impurities. We obtain the enhancement of the AHE in the $\text{Cu}(\text{Mn})$ alloy due to codoping of $5d$ impurities, comparing the AHA for the ternary alloy in column 7 with the AHA of $\text{Cu}(\text{Mn})$, $\alpha_{\text{AHE}}^{\text{Mn}} = 0.05 \times 10^{-2}$. The initial efficiency of the AHE for Mn impurities in copper can be increased by almost a factor of 100, which is present for the $\text{Cu}(\text{Mn}_{0.72}\text{Bi}_{0.28})$ alloy. This shows that the AHE can be enhanced significantly, if magnetic impurities are accompanied by nonmagnetic ones providing a strong SHE.

Thus the determination of the SHA via the AHE measurements proposed by Fert *et al.* [18] is justified. However, we show that it can be done in a more reliable way using the following procedure. According to Eq. (13), the weighting w_{max} maximizing the AHA has to be identified. This is expressed entirely via the spin-resolved longitudinal conductivities of the two binary alloys according to Eq. (11). For the $\text{Cu}(\text{Mn})$ binary alloy they can be obtained from first-principles calculations or derived from measurements of the total charge resistivities combined with the observation of the negative magnetoresistance [18]. Alternatively, the required spin-resolved conductivities can be attained from measurements comparing the temperature dependence in the magnetic binary and ternary alloys [24–26]. Ultimately, based on the proportionality stated in Eq. (12), the SHA of the nonmagnetic binary alloy can be derived from the measurements

of the AHA in the ternary alloy. Such measurements have to be performed at the extremal weighting w_{max} according to Eq. (11).

In summary, a detailed theoretical study of the anomalous Hall effect in dilute $\text{Cu}(\text{Mn})$ alloys codoped with different $5d$ as well as Bi impurities is presented. We show that under the condition of constant total impurity concentration the efficiency of the AHE, given by the anomalous Hall angle, can be significantly enhanced due to strong skew scattering caused by the heavy nonmagnetic impurities. The optimal composition providing the maximum of this efficiency substantially depends on the type of these additional impurities. However, the maximal enhancement relative to the SHA of the nonmagnetic binary alloy can be expressed in terms of the transport properties of the magnetic $\text{Cu}(\text{Mn})$ alloy only. This shows the direct connection between the AHA and SHA of the ternary and nonmagnetic binary alloy, respectively. Thus our work supports the idea of Ref. [18] to investigate the SHE via the AHE measurements and significantly improves the reliability of its quantitative predictions.

We acknowledge helpful discussions with A. Fert. This work was partially supported by the Deutsche Forschungsgemeinschaft (DFG) via SFB 762. In addition, M.G. acknowledges financial support from the DFG via a research fellowship (GR3838/1-1) and from the Leverhulme Trust via an Early Career Research Fellowship (ECF-2013-538).

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