Nonlocal anomalous Hall effect in ternary alloys based on noble metals

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We present a theoretical study of the nonlocal anomalous Hall effect induced by heavy-metal impurities in dilute magnetic alloys based on noble metals. The results of our first-principles calculations are shown in comparison to those obtained within a model consideration via Matthiessen's rule. Based on the transport properties of the constituent binary alloys, we reveal optimal host-impurity combinations to enhance the phenomenon. In particular, this allows us to explain experimental findings showing a strong effect in Cu-based alloys but a vanishing effect in the case of the Au host.

The spin Hall effect (SHE) [1–4] and the anomalous Hall effect (AHE) [5–7] are two related phenomena caused by spin-orbit coupling (SOC), which are very promising to be employed in novel spintronic devices. The efficiencies of these effects are described by the spin Hall angle (SHA) and the anomalous Hall angle (AHA),

$$\alpha_{\text{SHE}} = \frac{\sigma_{yx}^s}{\sigma_{xx}} = \frac{\rho_{xy}^s}{\rho_{xx}} \quad \text{and} \quad \alpha_{\text{AHE}} = \frac{\sigma_{yx}}{\sigma_{xx}} = \frac{\rho_{xy}}{\rho_{xx}}, \quad (1)$$

respectively. Here, σ_{xx} and ρ_{xx} are the conductivity and resistivity corresponding to the longitudinal charge transport. For the spin quantization axis pointing in the z direction, the spin Hall conductivity and the related resistivity are given by σ_{yx}^s and ρ_{xy}^s, respectively. Finally, σ_{yx} and ρ_{xy} describe the anomalous Hall conductivity and resistivity. In the case of dilute alloys, the skew-scattering mechanism [8,9] provides generally the dominant contribution to the SOC-driven transverse transport [10–14].

For practical applications, materials with large SHA and AHA are of interest. The authors of Ref. [15] proposed to enhance the AHE in noble-metal hosts with magnetic $3d$ impurities by the codoping of heavy-metal nonmagnetic impurities with strong SOC. As it was anticipated, this enhancement is due to the skew scattering of the spin-polarized current, induced by the magnetic impurities, from heavy-metal impurities [15]. Fert *et al.* [15] explained the idea within a simplified model approach and confirmed it experimentally on Cu-based alloys. However, for the case of Au as the host material, a contribution from the nonmagnetic impurities to the AHE was not detected. This puzzling point, combined with a general question on the optimal host-impurity combination for a strong AHE in ternary alloys, required further and deeper insight into the underlying physics.

Importantly, our study is related to the recent observations of the AHE in a nonmagnetic heavy-metal layer (Pt) contacted to a ferromagnetic insulator [16–18]. Since the magnetic proximity effect can be ruled out [17,19], Zhang and Vignale [20]

proposed an explanation [21] based on the combined action of spin-dependent scattering from the magnetic interface and the SHE in the bulk of the metal. This spatial separation of the SOC and the magnetization, both required for the AHE, has the same concept as the aforementioned mechanism proposed by Fert *et al.* [15] for ternary alloys. That is why we adopt the name *nonlocal anomalous Hall effect* [20] for the phenomenon considered in the present work.

Recently, we did provide an accurate theoretical analysis [22] serving as a detailed background for the idea of Fert *et al.* [15] to enhance the AHE in ternary alloys. The efficiency of our approach was demonstrated for Cu(Mn)-based ternary alloys by means of both first-principles computations and model calculations based on Matthiessen's rule. In this Rapid Communication we perform a further exhaustive investigation considering Cu, Ag, and Au hosts with various magnetic $3d$ impurities as well as heavy nonmagnetic impurities, which allows us to answer the open questions mentioned above. To this end, we use the approach of Refs. [22,23], which is shortly described in what follows.

Our model calculations are based on Matthiessen's rule [24] applied to an alloy of the form $H(A_{1-w}B_w)$. This corresponds to the host H containing impurities of two types, A and B, as described in Table I. In its conventional macroscopic sense, Matthiessen's rule states to add the resistivities of the two constituent binary alloys, in order to obtain the resistivity of the ternary alloy. Within the two-current model [25] this implies [23,26]

$$\hat{\rho}^\pm(A_{1-w}B_w) = (1-w)\hat{\rho}^\pm(A) + w\hat{\rho}^\pm(B), \quad (2)$$

where "+" and "−" relate to the "spin-up" and "spin-down" channel, respectively. Then, the AHA of the ternary alloy $H(A_{1-w}B_w)$ can be written as [23]

$$\alpha_{\text{AHE}}^{AB}(w) = -\frac{\frac{\rho_{xx}^{-AB}}{\rho_{xx}^{+AB}}\rho_{yx}^{+AB} + \frac{\rho_{xx}^{+AB}}{\rho_{xx}^{-AB}}\rho_{yx}^{-AB}}{\rho_{xx}^{+AB} + \rho_{xx}^{-AB}}, \quad (3)$$

where the involved quantities are the components of the resistivity tensor of Eq. (2) depending on the weighting factor w.

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TABLE I. Short description of the considered systems as ternary alloys of the form $H(A_{1-w}B_w)$ with the weighting factor $w \in [0,1]$ and the total impurity concentration 1 at. %.

Considered hosts	(H):	Cu, Ag, Au
Magnetic impurities	(A):	V, Cr, Mn, Fe, Co
Nonmagnetic impurities	(B):	Sb, Lu, Ta, Ir, Bi

Based on the assumption

$$\frac{(1-w)\rho_{yx}^+(A)}{w\rho_{yx}^+(B)} \approx \frac{(1-w)\rho_{yx}^-(A)}{w\rho_{yx}^-(B)} \approx 0, \quad (4)$$

one obtains [23]

$$\alpha_{\mathrm{AHE}}^{AB}(w) \approx -\frac{w\rho_{yx}^{+B}\left(\frac{\rho_{xx}^{-AB}}{\rho_{xx}^{+AB}} - \frac{\rho_{xx}^{+AB}}{\rho_{xx}^{-AB}}\right)}{\rho_{xx}^{+AB} + \rho_{xx}^{-AB}}. \quad (5)$$

Considering the extremum of the ratio $\alpha_{\mathrm{AHE}}^{AB}/\alpha_{\mathrm{SHE}}^B$ as a function of w leads to [23]

$$\left.\frac{\alpha_{\mathrm{AHE}}(A_{1-w}B_w)}{\alpha_{\mathrm{SHE}}(B)}\right|_{w_{\max}} \approx \frac{\sqrt{\rho_{xx}^-(A)} - \sqrt{\rho_{xx}^+(A)}}{\sqrt{\rho_{xx}^-(A)} + \sqrt{\rho_{xx}^+(A)}}, \quad (6)$$

where

$$w_{\max} \equiv w_{\max}(AB) \approx \frac{\sqrt{\rho_{xx}^+(A)\rho_{xx}^-(A)}}{\rho_{xx}^+(B) + \sqrt{\rho_{xx}^+(A)\rho_{xx}^-(A)}}. \quad (7)$$

The right hand side of Eq. (6) is independent of the type of nonmagnetic defects and is solely determined by the longitudinal resistivities of the binary system composed of host and magnetic impurities. The assumption of Eq. (4) implies negligible skew scattering at magnetic defects in comparison to nonmagnetic impurities, taking into account their relative concentrations. Consequently, for moderate values of the weighting factor ($w \sim 0.5$), the approximative description of the AHA in ternary alloys by Eq. (5) is valid, if the condition $|\rho_{yx}^\pm(B)| \gg |\rho_{yx}^\pm(A)|$ is fulfilled. For higher values of w, the approximation should work better, whereas for $w \to 0$ it fails unless $\rho_{yx}^+(A) \approx \rho_{yx}^-(A) \approx 0$.

The straightforward first-principles transport calculations performed for ternary alloys are based on the microscopic analog of Matthiessen's rule [26]

$$P_{kk'}(A_{1-w}B_w) = (1-w)P_{kk'}(A) + wP_{kk'}(B) \quad (8)$$

applied to the probability of the transition from an initial state k to a final state k' [22]. This means that the microscopic transition probabilities are added here, in contrast to the macroscopic resistivities summed up for the conventional Matthiessen's rule of Eq. (2). The resulting transition probability $P_{kk'}(A_{1-w}B_w)$

TABLE II. The anomalous Hall angle α_{AHE} and the spin Hall angle α_{SHE} (both in %) caused by Mn as well as Ir and Ta impurities, respectively, in the Cu, Ag, and Au hosts.

Host	Cu	Ag	Au
α_{AHE} [Mn]	0.05	0.19	1.13
α_{SHE} [Ir]	3.58	2.62	1.34
α_{SHE} [Ta]	1.51	1.15	0.66
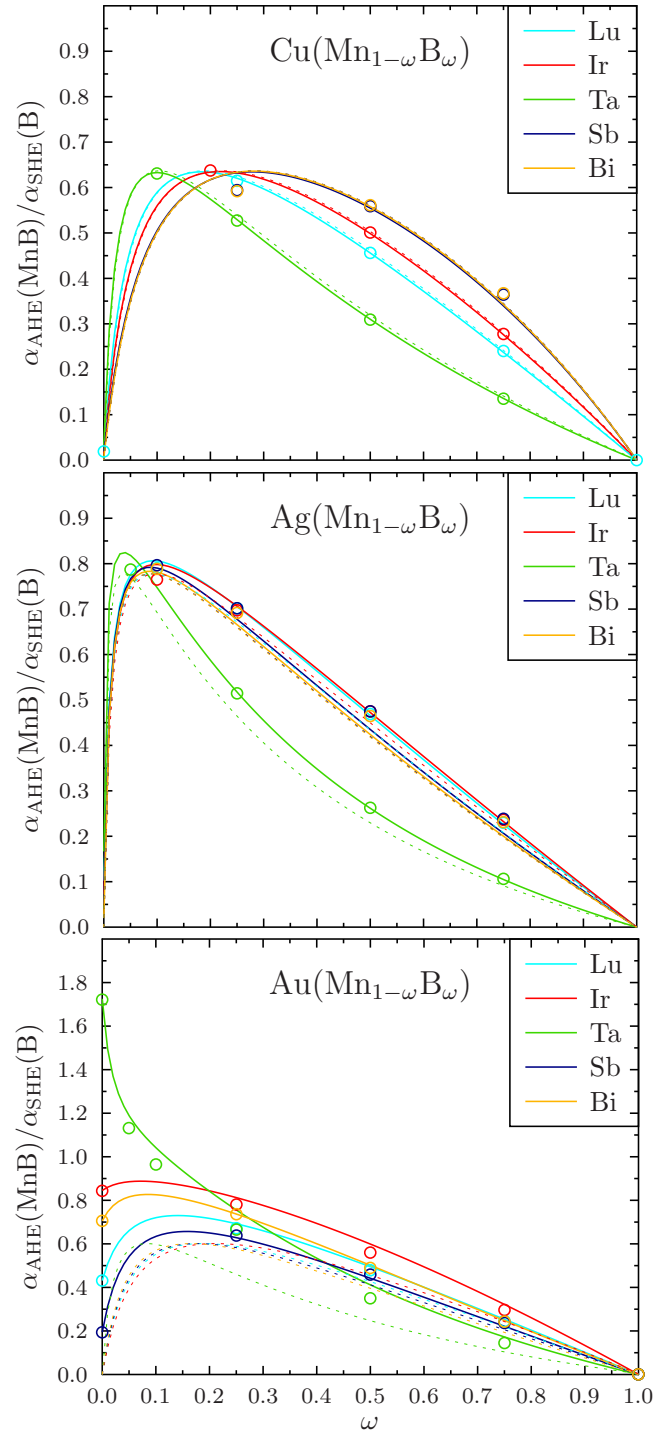
FIG. 1. The anomalous Hall angle of the $Cu(Mn_{1-w}B_w)$, $Ag(Mn_{1-w}B_w)$, and $Au(Mn_{1-w}B_w)$ alloys divided by the spin Hall angle of the related $Cu(B)$, $Ag(B)$, and $Au(B)$ alloys, where B denotes the nonmagnetic impurities of Table I. The solid lines provide the results obtained by Matthiessen's rule via Eq. (3) and the dashed lines with the approximation of Eq. (5). The dots are results of straightforward *ab initio* transport calculations. They are performed according to the approach of Ref. [23], where Fig. 2 relates to the upper graph of this figure.

TABLE III. The spin-resolved longitudinal charge resistivities (in $\mu\Omega\,\text{cm}$) and the anomalous Hall angle (in %) of the magnetic Cu-based binary alloys.

Alloy	Cu(Co)	Cu(Fe)	Cu(Mn)	Cu(Cr)	Cu(V)
ρ_{xx}^{+}	1.0	0.3	0.8	10.1	35.9
ρ_{xx}^{-}	22.0	34.0	15.8	11.4	23.0
α_{AHE}	0.179	0.004	0.050	-0.030	-0.095

is used to solve the Boltzmann equation [27]. Following the approach of Refs. [28,29], this provides the longitudinal charge conductivity as well as the skew-scattering contribution to both the spin and anomalous Hall conductivities. The same *ab initio* approach applied to the constituent binary alloys provides their transport properties required as the input parameters for our model calculations.

The theoretical formalisms sketched above are applied to 75 ternary alloys based on the Cu, Ag, and Au hosts with different magnetic and nonmagnetic impurities, as represented in Table I.

We start our discussion of the results by considering ternary alloys where the magnetism is induced by Mn atoms. The corresponding AHA normalized to the SHA of the constituent nonmagnetic binary alloys is shown in Fig. 1. For all systems, the *ab initio* results are well reproduced by Eq. (3) within the conventional Matthiessen's rule. In addition, for the Cu and Ag hosts, similar good agreement is reached via the approximative approach of Eq. (5), that is, without skew scattering at magnetic impurities. This is not the case for the Au(Mn)-based alloys, where generally the approximation of Eq. (5) differs significantly from the first-principles calculations. The latter show that Ir impurities in Au provide practically no enhancement of the AHE, whereas adding Ta impurities even leads to a reduction of the effect. This is in full agreement with the findings of Fert *et al.* [15], who observed that Ir and Ta impurities added to the Au(Mn) alloys do not enhance the effect. A simple qualitative explanation can be given with the help of Table II. The trends for the AHA caused by Mn impurities and the SHA provided by Ir and Ta impurities clearly indicate the increase of the AHE and the reduction of the SHE going from Cu over Ag to Au. As a result, one can see already for silver prominent deviations from the *ab initio* results if the approximation of Eq. (5) is exploited. The effect is much more drastic for the Au host, where the assumption of Eq. (4) fails significantly since the AHE induced by Mn impurities is not negligible. According to Table II, the related skew scattering becomes comparable to Ir impurities and much stronger than for Ta impurities. This explains why Fert *et al.* [15] could not enhance the AHE in the Au(Mn) alloys doped with Ir and Ta impurities.

Figure 2 shows Cu-based ternary alloys [30] with the same nonmagnetic impurities as in Fig. 1 but considering further $3d$ magnetic impurities. For the $\text{Cu}(\text{Co}_{1-w}\text{B}_w)$ and $\text{Cu}(\text{Fe}_{1-w}\text{B}_w)$ systems a similar behavior as for the Cu(Mn)-based alloys is observed. By contrast, for Cr and V as magnetic impurities the effects are strongly reduced. This can be understood by means of Eq. (6) stating that the extremum is determined by the difference of the spin-resolved longitudinal resistivities. Large
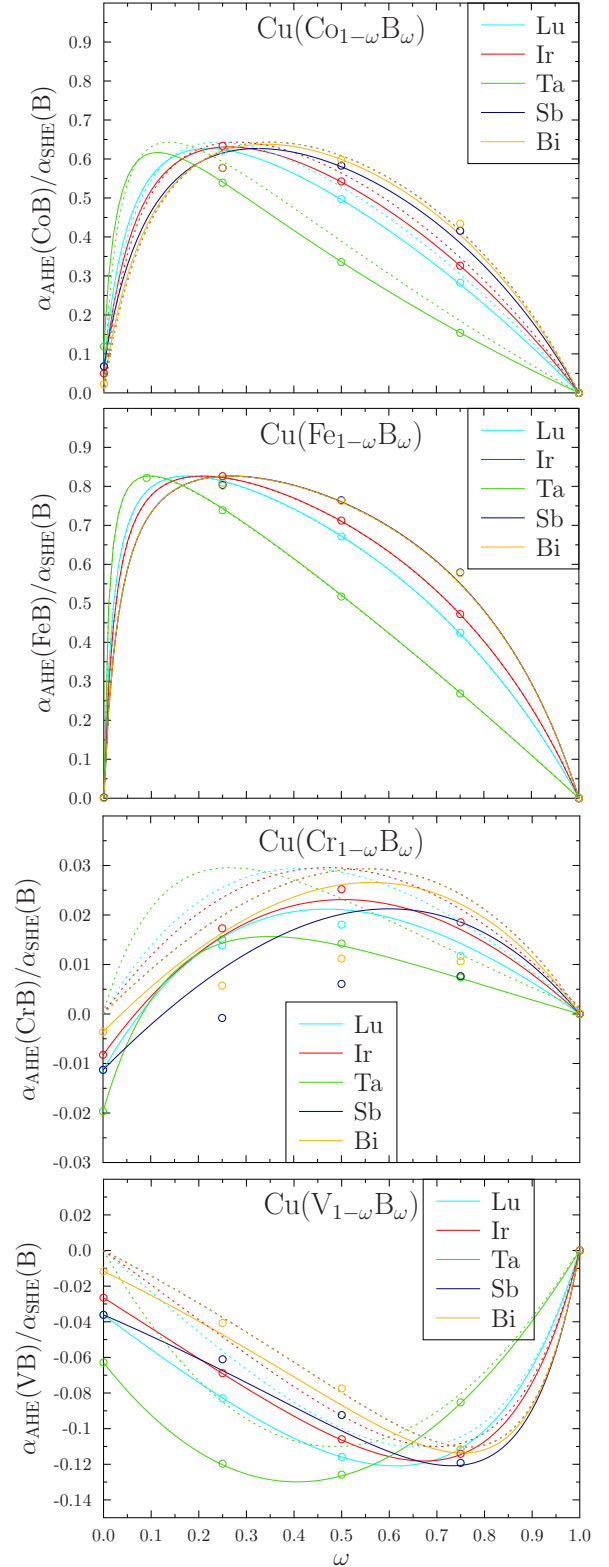

FIG. 2. The anomalous Hall angle of the $\text{Cu}(A_{1-w}B_w)$ alloys divided by the spin Hall angle of the related $\text{Cu}(B)$ alloys, where A and B denote the magnetic and nonmagnetic impurities of Table I, respectively. The solid lines provide the results obtained with Matthiessen's rule via Eq. (3) and the dashed lines with the approximation of Eq. (5). The dots are results of straightforward *ab initio* transport calculations according to the approach of Ref. [23].
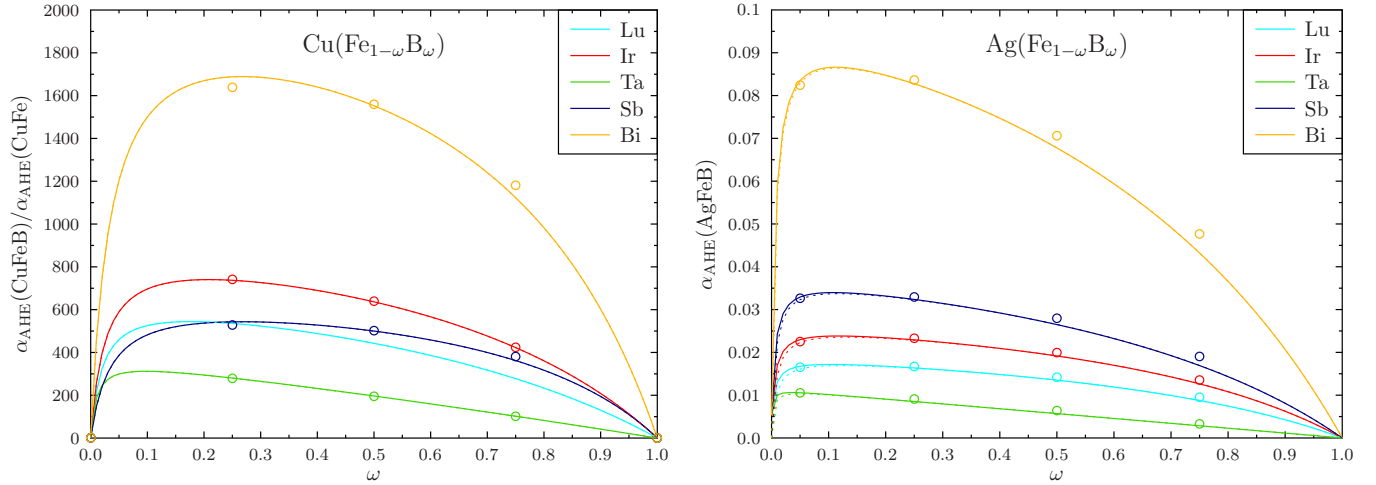
FIG. 3. The AHA of the $Cu(Fe_{1-w}B_w)$ alloys divided by the AHA of the $Cu(Fe)$ alloy (left) and the AHA of the $Ag(Fe_{1-w}B_w)$ alloys (right), where B denotes the nonmagnetic impurities of Table I. The dashed lines corresponding to the approximation of Eq. (5) practically coincide with the solid lines, providing the results obtained with Matthiessen's rule via Eq. (3). The dots are results of straightforward *ab initio* transport calculations performed according to the approach of Ref. [23].

differences between $\rho_{xx}^-(A)$ and $\rho_{xx}^+(A)$ give rise to highly spin-polarized currents and the strong AHE induced by their skew scattering on heavy-metal impurities. As shown in Table III and Fig. 2, this is found for the Cu-based alloys with Co and Fe impurities. In comparison to them, Cr impurities induce a much smaller difference, which suppresses the enhancement of the AHE via the SHE. For the case of V impurities, the magnitude of the AHA shown in Fig. 2 is slightly larger than for Cr impurities, but the opposite sign is present for its maximal value. This is caused by the fact that for the Cu(V) binary alloy we find $\rho_{xx}^-(A) < \rho_{xx}^+(A)$.

Furthermore, Table III provides the AHA for the considered magnetic impurities in copper. This explains the trend for the deviations between the results of the approximation of Eq. (5) and Matthiessen's rule via Eqs. (2) and (3). Indeed, the corresponding perfect agreement for the Cu(Fe)-based ternary alloys derives from the very small AHA in the Cu(Fe) binary alloy. For the Cu(Mn)-based systems, presented in Fig. 1, deviations are visible due to the enhanced α_{AHE} shown in Table III. For Co impurities, which provide an even larger AHA, the deviations are more remarkable for the ternary alloys. Although the strength of the skew scattering in the Cu(Cr) and Cu(V) binary alloys is comparable to that of the Cu(Mn) alloy, the largest deviations are obtained for Cr and V impurities. However, this is entirely due to the extremely small AHA of the related ternary alloys, which becomes comparable to the errors induced by the approximation of Eq. (4). The small magnitude of the AHA for ternary alloys with Cr and V impurities is also responsible for the remarkable deviations between the corresponding results of Matthiessen's rule according to Eq. (3) and the *ab initio* calculations. These deviations are most pronounced for Bi and Sb impurities, which are known to be strong *p* scatterers [31]. Therefore, they cause a strong anisotropy in the scattering over the Fermi surface of the host, whereas Matthiessen's rule holds exactly for an isotropic scattering [32].

For a better representation of the results, the AHAs of the ternary alloys shown by Figs. 1 and 2 were normalized to

the SHA of the related nonmagnetic binary alloys. However, in practice the aim is to increase the strength of the AHE itself. Therefore, it is important to consider the maximal enhancement of the AHE as well as the maximal absolute AHA attained for the considered systems. This is shown in Fig. 3, which identifies Fe combined with Bi as magnetic and nonmagnetic impurities, respectively, to create the best results. The maximal relative enhancement of the AHE, expressed as $\frac{\alpha_{AHE}(AB)}{\alpha_{AHE}(A)}$, is achieved for the $Cu(Fe_{0.75}Bi_{0.25})$ alloy and corresponds to the value of 1700. This is 17 times larger than what was obtained previously for the $Cu(Mn_{0.72}Bi_{0.28})$ alloy [23]. The much larger value can be explained by the very small AHA of the Cu(Fe) binary alloy. Searching for systems with large absolute AHAs, we have identified the $Ag(Fe_{0.9}Bi_{0.1})$ alloy providing the maximal AHA among the considered systems. As shown in Fig. 3, it corresponds to $\alpha_{AHE} = 8.6\%$, which is comparable to the SHA caused by Bi impurities in copper and silver obtained [33] as 8.1% and 9.5%, respectively.

In summary, we have investigated the nonlocal anomalous Hall effect caused by the skew scattering at heavy-metal nonmagnetic impurities of spin-polarized currents induced by $3d$ magnetic atoms in Cu, Ag, and Au. We reveal a crucial role of the type of magnetic impurities for the magnitude and the sign of the anomalous Hall angle. As optimal systems to enhance the effect, we identify Cu- and Ag-based alloys where the AHE induced by the $3d$ atoms is weak and the skew scattering caused by nonmagnetic impurities is strong. The situation with the Au host is shown to be opposite, which explains finally the experimental data where no enhancement of the effect was found in the case of gold. Our detailed analysis of the material-specific calculations provides general rules to handle and enhance the anomalous Hall effect in dilute ternary alloys. They imply the need for heavy nonmagnetic impurities in a relatively light host in combination with magnetic impurities providing no strong skew scattering but a large difference in the spin-resolved longitudinal resistivities. This knowledge is important to optimize

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[21] We should also mention an alternative explanation of the observed phenomenon proposed in Refs. [17,34], where it was assumed that both the SHE and the inverse SHE occur successively in the heavy metal. This was called the double-spin-Hall mechanism by the authors of Ref. [20], who criticized it for the short spin diffusion length required to fit the experimental data. However, a number of different experiments (see, for instance, Table I in Ref. [35]) confirm the spin diffusion length of the order of 1 nm assumed in Refs. [17,34]. Nevertheless, the double-spin-Hall mechanism is of second order in the SOC or SHA. By contrast, the theory of Zhang and Vignale [20] proposes a first-order mechanism to describe the puzzling experiments, which should dominate over the other one.

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