Linear response Kubo-Bastin formalism with application to the anomalous and spin Hall effects: A first-principles approach

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(Received 22 May 2015; revised manuscript received 19 August 2015; published 17 November 2015)

We present a general first-principles approach to treat various linear response phenomena relevant for spintronics. It is based on a Kubo-Bastin formalism and implemented within the multiple-scattering Korringa-Kohn-Rostoker (KKR) Green's function method with the underlying electronic structure determined by density functional theory. The symmetric (e.g., longitudinal electronic transport) as well as the antisymmetric (e.g., transverse transport) parts of the response tensor are determined, including both the so-called Fermi-sea and the Fermi-surface contributions. To describe spin-orbit-induced phenomena, such as the anomalous and spin Hall effects, a fully relativistic description is employed. Exploiting the adopted Green's function method substitutional disorder in the full concentration range of alloys is treated within the coherent potential approximation, taking full account of occurring vertex corrections in the averaging procedure for the linear response quantities. Extrinsic (scattering related, e.g., side-jump and skew scattering) and intrinsic (band structure–related) contributions to the transport tensors are treated on equal footing. Other phenomena, such as Gilbert damping and spin-orbit torques, are particular cases of the general framework and their determination is briefly addressed. The versatility of the method is demonstrated by presenting results for the anomalous and spin Hall conductivities for elemental transition metals and their alloys.

DOI: 10.1103/PhysRevB.92.184415 PACS number(s): 71.15.Rf, 72.10.Bg, 72.15.−v, 72.25.Ba

I. INTRODUCTION

There exist a number of transverse transport phenomena that have attracted a lot of attention in recent years due to their potential application in spintronics and their interesting underlying mechanisms. Among them are the anomalous (AHE) [1] and spin Hall (SHE) [2–4] effects and their spin-caloritronic counterparts [5], the anomalous and spin-Nernst effects [6–8], as well as the newly discovered spin-orbit torque in which a current exerts a torque on the magnetization in a ferromagnet [9–11]. Common to these effects is their relativistic origin, i.e., they are induced by spin-orbit coupling.

Quite generally, mechanisms giving rise to these effects are classified as band structure–related topological intrinsic or scattering-related extrinsic contributions (among the latter are skew and side-jump contributions). Many model calculations exist for these effects, each of which focuses on one or a few underlying mechanisms and typically rely on certain parameters [1,12–14]. Only recently have *ab initio* methods been developed that in most cases start from a density functional theory description of the electronic structure and that are able to provide a material-specific characterization of these phenomena. Several computer codes are now able to determine the intrinsic Berry-phase-associated contributions relying on the existence of well-defined energy bands in ordered systems [15–20]. Disorder in this particular approach can be introduced in a phenomenological way which allows one to include finite lifetime effects and can be used to describe systems with small content of impurities (dilute limit). On the other hand, the Boltzmann approach has been used to deal exclusively with extrinsic skew scattering contributions in the dilute limit. An approach that is capable of treating all the aforementioned linear response phenomena in a general way, i.e., treating intrinsic and extrinsic contributions on the same footing as well as being able to include disorder away from the dilute limit, is the Kubo linear response formalism in combination with a suitable alloy theory (see below).

The latter is our methodological starting ground in its Kubo-Greenwood (KG) formulation that is well established in describing longitudinal electronic transport, more precisely, giving the symmetric part of the transport tensor that connects a current with the electric field. Only states at the Fermi energy (Fermi surface) contribute to this part of the transport tensor. Many first-principles calculations have been performed employing the KG method implemented within the Korringa-Kohn-Rostoker (KKR) or the linear muffin-tin orbital (LMTO) electronic structure methods, demonstrating the viability to treat disordered systems and giving material-specific results [21–23]. Let us note in passing that already on the KG level the inclusion of vertex corrections (vc) becomes important and is readily incorporated in these approaches.

Going beyond the KG method and capturing the antisymmetric (transverse) parts of the transport tensors is methodologically and computationally much more demanding and only recently first-principles approaches have been devised that are based on the Kubo-Středa and Kubo-Bastin formalism [24–28]. There are several reasons for this: (i) As transverse transport phenomena like the AHE and SHE are manifestly spin-orbit induced, the effect of spin-orbit coupling has to be incorporated appropriately when calculating the electronic structure. (ii) One contribution to the tensor results exclusively from the states at the Fermi level and depends, in particular for pure systems, very sensitively on the topology of the Fermi surface. This implies the use of a huge number of k points needed for the Brillouin-zone integrations. Also, in the dilute limit of disordered alloys the vertex corrections have been shown to be of utter importance [25,29], again leading, together with the fine structure of the electronic states to be

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sampled, to great computational effort in the evaluation of k-space integrals. (iii) Finally, going beyond KG one either has to include a Fermi-sea term in the Kubo-Bastin formulation or to recast the transport equations into the Kubo-Středa equation. The latter then is often simplified by neglecting an orbital current term or relying on cancellation of terms in inversion symmetric systems, therefore restricting its range of application.

In this paper we present a Kubo-Bastin framework that in its formulation and implementation within relativistic multiple-scattering theory allows one to treat a variety of spin-orbit-induced linear response phenomena including the anomalous and spin Hall effect. It is applicable to pure systems as well as disordered alloys in the full concentration range and treats intrinsic (coherent) and extrinsic (incoherent) contributions within one and the same methodological approach. The application of the scheme to other phenomena (e.g., Gilbert damping, spin-orbit torques) is straightforward and is briefly discussed.

The paper is organized as follows: In Sec. II we formulate a generalized Kubo-Bastin theory within a fully relativistic framework. Based on the given expression we perform a symmetry analysis of the response tensor followed by a particular formulation for the anomalous and spin Hall effects. We then outline the linear response Kubo-Bastin approach within the relativistic KKR method, with more details given in the Appendix. In Sec. III we give technical details concerning the implementation. Finally, in Sec. IV we present results for the AHE and SHE in pure systems as well as disordered alloys. The paper is summarized in Sec. V.

II. THEORY

As we want to discuss, in particular, transverse spin-orbit-induced transport phenomena, we base our approach on the relativistic four-component Dirac formalism when dealing with the underlying electronic structure. This is motivated by the following reasons: (i) no approximation is involved when treating spin-orbit-induced properties, and (ii) it allows one to avoid problems to treat disorder [30] (vertex corrections) which would otherwise occur in a Pauli approach. The corresponding Dirac-Hamiltonian is given as

$$\hat{H}_{\mathrm{D}} = -i\hbar c\,\boldsymbol{\alpha}\cdot\boldsymbol{\nabla} + (\beta - \mathbb{I}_4)mc^2 + V_{\mathrm{KS}}(\mathbf{r})\,. \tag{1}$$

The single-particle potential V_{KS} appearing in Eq. (1) is determined in the framework of Kohn-Sham-Dirac (KSD) spin-density functional theory (KSD-SDFT) [31,32] and includes an exchange term $\beta\boldsymbol{\Sigma}\cdot\mathbf{B}_{\mathrm{xc}}$. The standard Dirac and spin matrices [31,33,34] α_μ, β, and Σ_μ are given as ($\mu \in \{\mathrm{x},\mathrm{y},\mathrm{z}\}$)

$$\alpha_\mu = \begin{pmatrix} 0_2 & \sigma_\mu \\ \sigma_\mu & 0_2 \end{pmatrix},\quad \beta = \begin{pmatrix} \mathbb{I}_2 & 0_2 \\ 0_2 & -\mathbb{I}_2 \end{pmatrix},\quad \Sigma_\mu = \begin{pmatrix} \sigma_\mu & 0_2 \\ 0_2 & \sigma_\mu \end{pmatrix}, \tag{2}$$

with the σ_μ being the Pauli matrices. The KSD Green's function (GF) is defined as the resolvent of the Dirac-Hamiltonian Eq. (1), $\hat{G}(z) = (z - \hat{H}_{\mathrm{D}})^{-1}$, with z being a complex energy variable.

A. Generalized Kubo-Bastin formalism

The starting point of our derivation is the Kubo-Bastin [35] like expression for the response tensor χ describing the reaction of the system in the observable represented by an operator $\hat{\mathbf{B}}$ due to the perturbation represented by the operator $\hat{\mathbf{A}}$:

$$\chi_{\mu\nu} = -\frac{\hbar}{2\pi\Omega}\int_{-\infty}^{\infty} f(E)\mathrm{Tr}\left\langle \hat{B}_\mu \frac{d\hat{G}^+}{dE}\hat{A}_\nu(\hat{G}^+ - \hat{G}^-) - \hat{B}_\mu(\hat{G}^+ - \hat{G}^-)\hat{A}_\nu\frac{d\hat{G}^-}{dE}\right\rangle dE\,. \tag{3}$$

Here $\mu,\nu \in \{x,y,z\}$ denote Cartesian coordinates, Ω is the volume of the system, $f(E) = [e^{(E-\mu)/k_{\mathrm{B}}T} + 1]^{-1}$ denotes the Fermi-Dirac distribution function with the chemical potential μ, the Fermi energy $E_{\mathrm{F}} = \mu(T = 0\,\mathrm{K})$, $\hat{G}^+$ and $\hat{G}^-$ are the retarded and advanced Green's function operators (for brevity their energy arguments will be suppressed), and $\langle\ldots\rangle$ denotes a configurational average. Following a procedure by Crépieux and Bruno [30], when deriving the Kubo-Středa equation we obtain (by keeping one half of the term and doing a partial integration on the second half) an expression that lends its hand to further insightful analysis as well as a first-principles implementation:

$$\chi_{\mu\nu} = \chi^{I}_{\mu\nu} + \chi^{II}_{\mu\nu} \tag{4}$$

$$\chi^{I}_{\mu\nu} = -\frac{\hbar}{4\pi\Omega}\int_{-\infty}^{\infty}\frac{df(E)}{dE}\mathrm{Tr}\langle \hat{B}_\mu(\hat{G}^+ - \hat{G}^-)\hat{A}_\nu\hat{G}^- - \hat{B}_\mu\hat{G}^+\hat{A}_\nu(\hat{G}^+ - \hat{G}^-)\rangle dE \tag{5}$$

$$\chi^{II}_{\mu\nu} = +\frac{\hbar}{4\pi\Omega}\int_{-\infty}^{\infty} f(E)\mathrm{Tr}\left\langle \hat{B}_\mu\hat{G}^+\hat{A}_\nu\frac{d\hat{G}^+}{dE} - \hat{B}_\mu\frac{d\hat{G}^+}{dE}\hat{A}_\nu\hat{G}^+ - \left(\hat{B}_\mu\hat{G}^-\hat{A}_\nu\frac{d\hat{G}^-}{dE} - \hat{B}_\mu\frac{d\hat{G}^-}{dE}\hat{A}_\nu\hat{G}^-\right)\right\rangle dE. \tag{6}$$

In the limit $T \to 0\,\mathrm{K}$, $f(E)$ becomes a step function and the first term Eq. (5) contributes to χ only in quantities to be evaluated at the Fermi energy E_{F}, whereas for the second term Eq. (6) the integration is over all occupied states. For this reason in what follows the term $\chi^{I}_{\mu\nu}$ Eq. (5) will be denoted as Fermi-surface and the term $\chi^{II}_{\mu\nu}$ Eq. (6) as the Fermi-sea term. Note that the last equation is a different but an equivalent form of the original equation by Bastin *et al.* [35].

B. Symmetry analysis

For the particular case of $\hat{\mathbf{A}} = \hat{\mathbf{B}} = \hat{\mathbf{O}}$ the Fermi-sea term is purely antisymmetric, $\chi^{II}_{\mu\nu} = -\chi^{II}_{\nu\mu}$. This can be seen by inspecting the first term in Eq. (6) containing only retarded ($\hat{G}^+$) as well as the second term in parenthesis containing exclusively advanced ($\hat{G}^-$) Green's functions. Both terms are antisymmetric, which can be shown by exploiting the property of the trace.

The analysis of the Fermi-surface term can be carried out by considering the symmetry-related subexpression of χ^{I}, i.e.,

$$C_{\mu\nu} = \mathrm{Tr}\langle \hat{B}_\mu(\hat{G}^+ - \hat{G}^-)\hat{A}_\nu\hat{G}^- - \hat{B}_\mu\hat{G}^+\hat{A}_\nu(\hat{G}^+ - \hat{G}^-)\rangle\,.$$

Extracting the symmetric part for $\hat{\mathbf{A}} = \hat{\mathbf{B}} = \hat{\mathbf{O}}$ leads to

$$\begin{aligned}\tfrac{1}{2}[C_{\mu\nu} + C_{\nu\mu}] &= -\mathrm{Tr}\langle \hat{O}_\mu(\hat{G}^+ - \hat{G}^-)\hat{O}_\nu(\hat{G}^+ - \hat{G}^-)\rangle \\ &= 4\mathrm{Tr}\langle \hat{O}_\mu \Im\hat{G}^+ \hat{O}_\nu \Im\hat{G}^+\rangle\,, \end{aligned} \tag{7}$$

a Kubo-Greenwood–like expression, where $\Im\hat{G}^+(E) = \frac{1}{2i}[\hat{G}^+(E) - \hat{G}^-(E)]$. This is frequently used in transport calculations, with $\hat{\mathbf{O}} = \hat{\mathbf{j}}$ being the charge current operator yielding the symmetric (and in particular, the longitudinal) contribution to the conductivity tensor $\sigma_{\mu\nu}$.

Extracting the antisymmetric part for $\hat{\mathbf{A}} = \hat{\mathbf{B}} = \hat{\mathbf{O}}$ gives

$$\begin{aligned}\tfrac{1}{2}[C_{\mu\nu} - C_{\nu\mu}] &= \tfrac{1}{2}\mathrm{Tr}\langle[\hat{O}_\mu(\hat{G}^+ - \hat{G}^-)\hat{O}_\nu \\ &\quad - \hat{O}_\nu(\hat{G}^+ - \hat{G}^-)\hat{O}_\mu](\hat{G}^+ + \hat{G}^-)\rangle \\ &= 2i\,\mathrm{Tr}\langle[\hat{O}_\mu \Im\hat{G}^+\hat{O}_\nu - \hat{O}_\nu\Im\hat{G}^+\hat{O}_\mu]\Re\hat{G}^+\rangle,\end{aligned}$$

where $\Re\hat{G}^+(E) = \frac{1}{2}[\hat{G}^+(E) + \hat{G}^-(E)]$. For the example of a charge-charge current response, this states that the (antisymmetric) anomalous Hall effect results from the Fermi sea as well as the antisymmetric surface contribution. It has been shown that in the latter case the Fermi-sea term can be transformed into a surface term [30,36] and the intrinsic AHE in a (pure) metallic ferromagnet is a topological Fermi-surface property [37].

To highlight the advantages of the presented scheme, let us note in passing that the case $\hat{\mathbf{A}} = \hat{\mathbf{B}} = \hat{\mathbf{T}}$, with $\hat{\mathbf{T}}$ being the magnetic torque operator, allows a formulation of the Gilbert damping [38–40]. Furthermore, the spin-orbit torque, i.e., the torque exerted on the magnetization in a ferromagnet resulting from a charge current [9,41], is obtained by using $\hat{\mathbf{B}} = \mathbf{T}$ and $\hat{\mathbf{A}} = \mathbf{j}$ [42,43].

Finally, we want to point out that further symmetry analysis of the response tensors on grounds of the (magnetic) space group of a bulk system can give additional relations, depending on the particular choice of operators $\hat{\mathbf{A}}$ and $\hat{\mathbf{B}}$ [44,45].

C. Conductivity within Kubo-Bastin linear response formalism

In the chosen relativistic formalism the electric current operator is given by $\hat{\mathbf{j}} = -|e|c\boldsymbol{\alpha}$, with $e > 0$ being the elementary charge. For describing the spin Hall effect, we here employ the relativistic spin (-polarization) current-density operator

$$\hat{\mathbf{J}}^\xi = \left(\beta\Sigma_\xi - \frac{\gamma_5\Pi_\xi}{mc}\right)|e|c\boldsymbol{\alpha}\,, \tag{8}$$

inspired by Bargmann and Wigner [46] and already used previously [25,47,48], with the kinetic momentum $\boldsymbol{\Pi} = (\hat{\mathbf{p}} + \frac{|e|}{c}\mathbf{A})\mathbb{1}_4$, the canonical momentum $\hat{\mathbf{p}}$, the vector potential $\mathbf{A}$, and [34]

$$\gamma_5 = \begin{pmatrix} 0_2 & -\mathbb{1}_2 \\ -\mathbb{1}_2 & 0_2 \end{pmatrix}. \tag{9}$$

For the remainder of the paper we consider the limit $T \to 0$ K of Eqs. (5) and (6) and two particular cases, both of which are characterized by choosing $\hat{\mathbf{A}} = \hat{\mathbf{j}}$ as charge current operator. The (longitudinal) charge and anomalous Hall conductivities are obtained by setting $\hat{\mathbf{B}} = \hat{\mathbf{j}}$. The spin Hall conductivity is obtained by setting $\hat{\mathbf{B}} = \hat{\mathbf{J}}^\xi$, where $\xi \in \{x,y,z\}$ characterizes the polarization direction of the spin current operator. With this Eqs. (5) and (6) read

$$\sigma^\xi_{\mu\nu} = \sigma^{\xi,I}_{\mu\nu} + \sigma^{\xi,II}_{\mu\nu}, \tag{10}$$

$$\sigma^{\xi,I}_{\mu\nu} = \frac{\hbar}{4\pi\Omega}\mathrm{Tr}\langle \hat{J}^\xi_\mu(\hat{G}^+ - \hat{G}^-)\hat{j}_\nu\hat{G}^- - \hat{J}^\xi_\mu\hat{G}^+\hat{j}_\nu(\hat{G}^+ - \hat{G}^-)\rangle, \tag{11}$$

$$\begin{aligned}\sigma^{\xi,II}_{\mu\nu} = \frac{\hbar}{4\pi\Omega}\int_{-\infty}^{E_\mathrm{F}} \mathrm{Tr}\Bigg\langle &\hat{J}^\xi_\mu\hat{G}^+\hat{j}_\nu\frac{d\hat{G}^+}{dE} - \hat{J}^\xi_\mu\frac{d\hat{G}^+}{dE}\hat{j}_\nu\hat{G}^+ \\ &-\left(\hat{J}^\xi_\mu\hat{G}^- j_\nu\frac{d\hat{G}^-}{dE} - \hat{J}^\xi_\mu\frac{d\hat{G}^-}{dE}\hat{j}_\nu\hat{G}^-\right)\Bigg\rangle dE\,,\end{aligned} \tag{12}$$

where in Eq. (11) the Green's functions are evaluated at the Fermi energy E_F and the energy arguments at the GFs have been omitted throughout. The conductivity tensor $\sigma_{\mu\nu}$ in terms of the charge-charge response is obtained by replacing $\hat{J}^\xi_\mu$ with $\hat{j}_\mu$ in the last expression. For the remainder of the paper we consider the special case $\xi = z$ and, if present, the following particular choice for the exchange field $\mathbf{B}_\mathrm{xc}(\mathbf{r}) = B(r)\hat{\mathbf{e}}_z$ in the Hamiltonian Eq. (1).

Note that in the discussion of longitudinal transport [49] and the AHE one can show that terms involving only retarded or advanced GF, i.e., terms of the type $\langle jG^+jG^+\rangle$ or $\langle jG^-jG^-\rangle$, can be neglected in the weak disorder limit [50], and this is indeed done in actual calculations [51]. In the present work all contributions are taken into account, in particular, because we discuss the full concentration range of alloys.

D. Kubo-Bastin linear response formalism within relativistic multiple-scattering KKR

The formalism presented here is inspired by previous implementations of the (relativistic) Kubo-Greenwood approach [52–56] which go back to a formulation by Butler [21]. These are restricted to the treatment of the symmetric part of the conductivity tensor evaluated at the Fermi energy. Here we report on a very general framework that (i) gives the symmetric as well as antisymmetric contributions by evaluating Fermi sea and surface contributions; (ii) is fully relativistic and therefore captures all important contributions to transverse transport (skew scattering, side jump); (iii) is easily extendable to any other operator pair for dealing with other phenomena like Gilbert damping [38,40,42] and spin-orbit torques; (iv) allows treatment of efficiently disordered systems, avoiding costly supercell approaches; and (v) lends its hand to straightforwardly include finite temperatures effects [57].

The evaluation and first-principles treatment of Eqs. (11) and (12) for solids requires a suitable representation of the GF, which in our chosen formalism will subsequently lead to a product expression containing matrix elements of the current operators with the basis functions and k-space integrals over scattering path operators. Disorder and ensuing vertex corrections in the averaging procedure will be treated by means of the coherent potential approximation (CPA) [21,58].

The real-space representation of the Green's function operator $\hat{G}(z)$ can be very efficiently obtained by using the spin-polarized relativistic version of multiple-scattering theory

[59–63]:

$$G(\mathbf{r},\mathbf{r}',z) = \sum_{\Lambda\Lambda'} Z^n_\Lambda(\mathbf{r},z)\,\tau^{nm}_{\Lambda\Lambda'}(z)\,Z^{m\times}_{\Lambda'}(\mathbf{r}',z) - \delta_{nm}\sum_\Lambda \big[Z^n_\Lambda(\mathbf{r},z)\,J^{n\times}_\Lambda(\mathbf{r}',z)\Theta(r'_n - r_n) + J^n_\Lambda(\mathbf{r},z)\,Z^{n\times}_\Lambda(\mathbf{r}',z)\Theta(r_n - r'_n)\big]. \quad (13)$$

Here $\mathbf{r},\mathbf{r}'$ refer to atomic sites at $\mathbf{R}_n$ and $\mathbf{R}_m$, respectively, where $Z^n_\Lambda(\mathbf{r},z) = Z_\Lambda(\mathbf{r}_n,z) = Z_\Lambda(\mathbf{r}-\mathbf{R}_n,z)$ as well as $J^n_\Lambda(\mathbf{r},z)$ are basis functions centered at positions $\mathbf{R}_n$. Note that here the basis functions are normalized according to the Oak Ridge–Bristol convention [64]. The four-component wave functions $Z^n_\Lambda(\mathbf{r},z)$ [$J^n_\Lambda(\mathbf{r},z)$] are regular (irregular) solutions to the single-site Dirac equation at complex energy z labeled by the combined quantum numbers Λ [$\Lambda = (\kappa,\mu)$], with κ and μ being the spin-orbit and magnetic quantum numbers [34]. The superscript $\times$ indicates the left-hand side solution of the Dirac equation [60]. The quantity $\tau^{nm}_{\Lambda\Lambda'}(z)$ is the scattering path operator that transfers an electronic wave coming in at site m into a wave going out from site n accounting for all possible intermediate scattering events. The retarded and advanced GF are obtained as the side limits $G^\pm(\mathbf{r},\mathbf{r}',E) = \lim_{\eta\to 0^+} G(\mathbf{r},\mathbf{r}',E \pm i\eta)$.

Inserting the real-space representation Eq. (13) into Eqs. (11) and (12) and cyclic permutation under the trace leads to sums of products of matrix elements evaluated on a given site and scattering path operators τ^{mn}. Pursuing the route of Butler [21] having a subsequent CPA averaging in mind, the conductivity tensor will partition into an on-site term σ^0 involving regular (Z_Λ) as well as irregular solutions (J_Λ) and an off-site term σ^1 containing only regular solutions (both for Fermi sea and surface terms):

$$\sigma^\xi_{\mu\nu} = \sigma^{\xi 0}_{\mu\nu} + \sigma^{\xi 1}_{\mu\nu} = \sigma^{\xi 0,I}_{\mu\nu} + \sigma^{\xi 1,I}_{\mu\nu} + \sigma^{\xi 0,II}_{\mu\nu} + \sigma^{\xi 1,II}_{\mu\nu}\,. \quad (14)$$

Working towards determining the energy derivative of the GF in terms of finite differences (see below) as well as representing the GF above and below the real axis leads to expressions of the form

$$\frac{1}{\Omega}\mathrm{Tr}\big\langle \hat{J}^\xi_\mu \hat{G}(z_a)\hat{J}_\nu \hat{G}(z_b)\big\rangle = \frac{1}{\Omega}\mathrm{Tr}\int_\Omega d^3r\,\hat{J}^\xi_\mu\,\Delta G_\nu(\mathbf{r},\mathbf{r},z_a,z_b) = \frac{1}{\Omega_n}\mathrm{Tr}\int_{\Omega_n} d^3r\,\hat{J}^\xi_\mu\,\Delta G^n_\nu(\mathbf{r},\mathbf{r},z_a,z_b)\,,$$

(Ω_n denotes the volume of the unit cell at site n), containing pairs of complex energies z_a and z_b and contributions to ΔG_ν with

$$\Delta G^n_\nu(\mathbf{r},\mathbf{r},z_a,z_b) = \sum_{\alpha\beta} x_\alpha x_\beta \Delta G^{1,\alpha\beta,n}_\nu(\mathbf{r},\mathbf{r},z_a,z_b) + \sum_{k=1}^{4} x_\alpha \Delta G^{0,\alpha,n}_{\nu k}(\mathbf{r},\mathbf{r},z_a,z_b)\,, \quad (15)$$

given in Appendix A. The Greek indices (α,β) denote alloy partners and x_α their concentrations. The terms in $\Delta G^{0,\alpha,n}_{\nu k}$ containing irregular solutions J_Λ, $k \in \{1,2,3,4\}$ are associated with the on-site contributions $\sigma^{\xi 0}$ only. The term $\Delta G^{1,\alpha\beta,n}_{\nu 1}$ containing exclusively regular solutions Z_Λ contributes to $\sigma^{\xi 1}$ requiring special treatment when performing the statistical average (done here within the CPA) in the case of an alloy. The appearing vertex corrections in this term are important and can, particularly in the dilute limit, give sizable contributions to the transverse conductivities (see Refs. [24,27] and below). As shown by Butler [21], they correspond to the scattering term in Boltzmann transport theory [25,65,66].

The evaluation of Eq. (15) leads to matrix elements of regular functions of the form $M^{ab\nu}_{\Lambda\Lambda'} = \langle Z^\times_\Lambda(z_a)|\hat{O}_\nu|Z_{\Lambda'}(z_b)\rangle_{\Omega_n}$ and matrix elements involving irregular functions whose evaluation is outlined in Appendix B. Let us note that the formalism is very general insofar as other linear response quantities (Gilbert damping, spin-orbit torques, etc.) are easily obtained by the appropriate choice of operators ($\hat{\mathbf{A}}$, $\hat{\mathbf{B}}$) and adaptation of their matrix elements to be inserted in the final multiple-scattering transport expressions.

The described formalism is applicable to pure systems as well as alloys in the full concentration range. For a pure system with a perfect band structure, the transverse (antisymmetric) component of the response is called intrinsic and is often associated with the existence of the Berry curvature coded in the band structure. An alloy, however, has no well-defined energy bands. Within the formalism presented here, one can separate the full response into coherent and incoherent contributions, with the latter exclusively caused by the vertex corrections. As a manner of speaking, the coherent contributions are here named intrinsic, the incoherent ones are called extrinsic, and the presented formalism captures both of them.

III. IMPLEMENTATION AND COMPUTATIONAL DETAILS

The expressions (11) and (12) as well as the following equations have been implemented into the MUNICH SPR-KKR package [63,67]. A fully relativistic Dirac four-component scheme for the basis functions Z_Λ and J_Λ has been used throughout with an angular momentum cutoff of $\ell_{\max} = 3$. The self-consistent field (SCF) potentials have been obtained within KSD-SDFT employing the Vosko-Wilk-Nussair (VWN) parametrization [68] for the exchange-correlation functional in the local density approximation (LDA). The involved energy integration has been performed on a semicircle in the complex plane using typically 50 energy points and 45^3 ($56^2 \times 30$) for cubic (hcp) system k-points in the BZ. As a shape approximation for the potential, the atomic sphere approximation (ASA) has been used. Experimental lattice constants have been used. Using these SCF potentials, subsequent Kubo-Bastin transport calculations have been performed. For the determination of the Fermi-surface term Eq. (11) in the concentrated regime of alloys approximately 10^5 k points in the BZ turned out to be sufficient due to smearing of the GF in k space for the disordered system. In the dilute limit with the concentration of an alloy partner becoming very small, around 10^6–10^7 k points had to be used to ensure convergence. In contrast to the disordered systems which for the calculations are carried out on the real-energy axis for pure elements, a small imaginary part has been added, $z = E_\mathrm{F} + i\eta$, and an extrapolation for $\eta \to 0^+$ has been carried out while ensuring

TABLE I. The AHC σ_{xy} in $(\Omega\,\mathrm{cm})^{-1}$ of the ferromagnetic transition metals bcc-Fe, hcp-Co, and fcc-Ni and the alloys $Fe_{50}Pd_{50}$ and $Ni_{50}Pd_{50}$ from *first-principles* theoretical (present work compared to other) as well as experimental (expt.) studies. The magnetization has been assumed to be oriented along the [001] direction.

	σ_{xy}^{0}	$\sigma_{xy}^{1,I}$	$\sigma_{xy}^{1,II}$	σ_{xy}	σ_{xy}^{theo}	σ_{xy}^{exp}
Fe	20	687	192	899	750,[d] 878,[e] 796[f]	1032[a]
Co	39	316	169	524	484,[g] 694,[e] 471[f]	813[h]
Ni	–84	–2654	57	–2681	–2500,[e] –2432[f]	–1100[b]
$Fe_{50}Pd_{50}$ (nvc)	–18	314	101	397		
$Fe_{50}Pd_{50}$ (vc)	–18	457	102	541		303[c]
$Ni_{50}Pd_{50}$ (nvc)	–113	–1830	130	–1813		
$Ni_{50}Pd_{50}$ (vc)	–113	–1417	130	–1400		–1293[c]

[a]Reference [69].
[b]Reference [70].
[c]Reference [71].
[d]Intrinsic, BCA, Ref. [16].
[e]Intrinsic, + scattering-independent side-jump, Ref. [72].
[f]Kubo-Bastin, TB-LMTO, Ref. [28].
[g]Intrinsic, BCA, Ref. [73].
[h]Estimated expt. value, Ref. [18].

for every value of η convergence with respect to the k mesh. Values of up to 10^9 k points have been used in this case.

For the treatment of the Fermi-sea contribution, Eq. (12) the energy path has been distorted to a semicircle in the upper (lower) half of the complex plane for the first (second) term containing the retarded (advanced) GF G^{+} (G^{-}) encompassing the valence states. The derivative of the GFs in the complex plane along a direction parallel to the real axis has been obtained by a two-point finite difference formula, $d\hat{G}^{\pm}(z)/dz \approx \frac{1}{h}[\hat{G}^{\pm}(z+h/2) - \hat{G}^{\pm}(z-h/2)]$, with $h \in \mathbb{R}$. A value of $h = 10^{-4}$ Ry turned out to be sufficient because of the smearing of the GF in the complex plane. The latter smoothing of the GF also leads to a fast k-mesh convergence, and it was sufficient to use around 10^3 k points at each energy point, except for the points near and next nearest to the real axis at E_F for which typically 10^6 k points have been used.

Here we restrict the spin current-density operator to z polarization, i.e., only $\hat{\mathbf{J}}^{z}$ is considered. Other polarization directions and the resulting tensor forms in a fully relativistic approach are discussed elsewhere [45]. Furthermore, as we here consider the SHE in paramagnetic systems without external fields, the vanishing vector potential in Eq. (8) results in a spin-polarization current-density operator with components

$$\hat{J}_{\mu}^{z} = \left(\beta\Sigma_{z} - \frac{\gamma_5 \hat{p}_z}{mc}\right)|e|c\alpha_{\mu}, \quad \mu \in \{x,y\}. \tag{16}$$

More details on the evaluation of matrix elements are given in Appendix B.

IV. RESULTS AND DISCUSSION

In Tables I and II we show the anomalous Hall conductivity (AHC) for various systems as calculated by the Kubo-Bastin approach [Eqs. (11) and (12)] for both pure systems as well as alloys.

TABLE II. The SHC σ_{xy}^{z} in $(\Omega\,\mathrm{cm})^{-1}$ of the nonmagnetic metals Cu, Pt, and Au and the alloys $Cu_{50}Au_{50}$ and $Au_{50}Pt_{50}$ from *first-principles* theoretical (present work compared to other) studies.

	σ_{xy}^{z0}	$\sigma_{xy}^{z1,I}$	$\sigma_{xy}^{z1,II}$	σ_{xy}^{z}	$\sigma_{xy}^{z,theo}$
Cu	–17	172	28	184	
Pt	98	4093	133	4324	4400[a]
Au	–16	743	90	817	700,[b] 800[c]
$Cu_{50}Au_{50}$ (nvc)	–20	605	71	656	
$Cu_{50}Au_{50}$ (vc)	–20	872	71	923	
$Au_{50}Pt_{50}$ (nvc)	34	2911	607	3553	
$Au_{50}Pt_{50}$ (vc)	34	2992	607	3634	

[a]Intrinsic, BCA, Ref. [17].
[b]Intrinsic, BCA, Ref. [76].
[c]Intrinsic, BCA, Ref. [77].

Let us first turn to the ferromagnetic systems and the determined values for the anomalous Hall conductivities. Table I shows the total conductivities σ_{xy} and the various contributions to it for the elemental ferromagnets Fe, Co, and Ni as well as for the two alloys $Fe_{50}Pd_{50}$ and $Ni_{50}Pd_{50}$. Discussing the overall numbers, one can state that for the systems considered the Fermi-surface contribution $\sigma_{xy}^{1,I}$ is the dominant one, but also the Fermi-sea term $\sigma_{xy}^{1,II}$ can give a significant contribution. This is seen, in particular, for the systems Fe, Co, and $Fe_{50}Pd_{50}$. Similar observations have been made before [28] in a tight-binding LMTO (TB-LMTO) framework (see the remarks below).

The site-diagonal term σ_{xy}^{0} is not significant, contributing only 2%–3% with a maximum of 10%. Note that we here show the sum of both Fermi sea and surface contributions to σ_{xy}^{0}. Both are numerically delicate, as they contain matrix elements involving the irregular solutions $J_{\Lambda}^{n}(\mathbf{r},z)$ and can become rather large. However, their sum σ_{xy}^{0} is small.

The AHC has been calculated recently within a Kubo-Bastin framework implemented in the TB-LMTO electronic structure method [28]. When comparing the results presented here to the latter ones, however, one has to be careful. First, in the TB-LMTO method the coherent potential functions and structure constants depend on the chosen representation. Even though the full conductivity is invariant with respect to the particular choice, some ambiguity in assigning terms contributing to the surface and sea terms arises, as only the sum of the antisymmetric part of the coherent surface term and sea term is invariant. Therefore only the numbers for the total conductivities should be compared. Second, in the LMTO transport approach there appear only intersite hoppings. A term equivalent to the site-diagonal contribution σ^{0} appearing in the present work does not exist. Third, the TB-LMTO method employs (configuration independent) effective velocity operators, i.e., the operator matrix elements are nonrandom while here the matrix elements as well as the scattering path operator are configuration dependent.

Turning now to the particular systems, for bcc Fe we find a total AHC that underestimates the experimental value by roughly 10%. On the other hand, this number is comparable to those obtained in calculations of the AHC employing the Berry curvature approach (BCA), including the intrinsic as well as a

scattering-independent side-jump term ($\sigma_{xy} = 878\,(\Omega\,\mathrm{cm})^{-1}$, Ref. [72]). Both of these contributions are included in the present formalism (coherent part). Note that calculations using the BCA leaving out the scattering-independent side-jump term give smaller values [15,16] [in the range of $\sigma_{xy} = 750\,(\Omega\,\mathrm{cm})^{-1}$]. For hcp Co the comparison to experiment as well as other theoretical results is less clear-cut, as there is a larger variation. Furthermore, the Fermi-sea term represents a significant contribution to the total AHC, as has been observed in another recent work [28]. For Ni notably all theoretical calculations employing the LDA grossly overestimate the experimental value. This has already been attributed to the deficiencies in properly describing the electronic structure, namely, the correlations are not fully captured by this approximation. Using the LDA + U or GGA + U approach, AHCs are obtained that are close to experimental values [20,57,72].

For alloys we show results for a particular concentration for $Fe_{50}Pd_{50}$ and $Ni_{50}Pd_{50}$ in Table I. Results for calculations including the vertex corrections (vc) as well as excluding them (nvc) are given. As can be seen the vc contribute substantially in the Fermi-surface term. On the other hand the Fermi-sea term does not contain any incoherent contribution, i.e. the vc do not occur in this case. This is in accord with the findings in Ref. [28], where it was analytically shown that for the AHE treated within the TB-LMTO CPA the vc are vanishing in the Fermi sea. Note, however, that this proof relied on a particular formulation of the CPA equations within the TB-LMTO formalism.

In Fig. 1 we show the AHC for Fe_xPd_{1-x} as a function of concentration. Overall the concentration dependence as well as the sign change is in good agreement to experiment. For all concentrations the dominant contribution to the AHC is given by the site-off diagonal Fermi-surface term ($\sigma_{xy}^{1,I}$). By analyzing the contribution dependence in more detail one observes

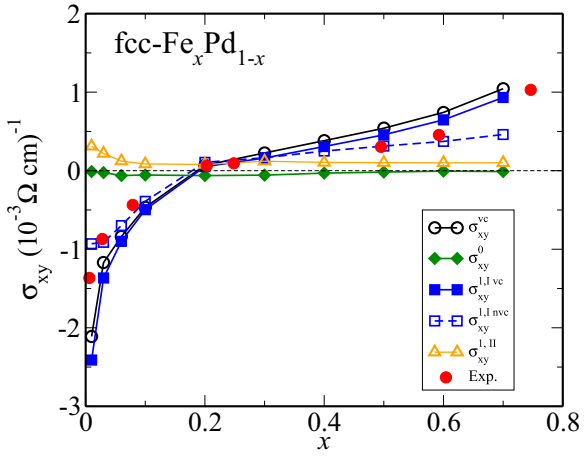

FIG. 1. (Color online) The AHC of fcc-Fe_xPd_{1-x} as a function of concentration x determined within the Kubo-Bastin formalism. The total AHC (σ_{xy}^{vc}) and different contributions to it are shown: the on-site term (σ_{xy}^{0}), the off-site Fermi-surface term, including vertex corrections ($\sigma_{xy}^{1,I,vc}$) and the off-site Ferm-sea contribution ($\sigma_{xy}^{1,II}$). Additionally, the off-site term omitting the vc ($\sigma_{xy}^{1,I,nvc}$) is shown for comparison. Experimental data [71] for σ_{xy} (full circles) determined at $T = 4.2$ K is also displayed.

that the incoherent contributions (vc) play a minor role in the middle of the concentration range but become very important at small concentrations. This dominance of extrinsic effects at small concentrations lends credibility to the Boltzmann formalism that is applicable to alloys in the dilute limit and captures the skew-scattering contribution [74]. Note that the formalism presented here gives all contributions to the AHC and allows one to extract intrinsic (coherent) as well as extrinsic (incoherent) contributions (e.g., skew scattering and side jump), as has been done before in the Kubo-Středa approach [24–26]. The site-diagonal term σ_{xy}^{0} gives only a minor contribution to the AHC over the whole concentration range and shows almost negligible variation. The Fermi-sea term $\sigma_{xy}^{1,II}$ follows the same trend, even though it is somewhat larger and shows stronger variation for vanishing concentration ($x \to 0$). One exception to the former statements is the range in which the total AHC changes sign ($x \approx 0.2$). There the site-diagonal as well as the Fermi-sea term gain larger relative weight that is, however, due to the fact that the Fermi-surface term approaches zero.

Let us turn now to the discussion of paramagnetic systems and the spin Hall conductivity (SHC). As both the AHE and SHE share the same relativistic origin and underlying mechanisms, observations made for the SHC can be discussed along the lines above for the AHC. In Table II we show the intrinsic SHC for Cu, Pt, and Au as well as the full SHC for the alloys $Cu_{50}Au_{50}$ and $Au_{50}Pt_{50}$. Overall, again the site-diagonal contribution σ_{xy}^{z0} is very small. The Fermi-sea contribution $\sigma_{xy}^{z1,II}$ is small but non-negligible, and for $Au_{50}Pt_{50}$ is largest and constitutes about 15% of the total SHC. For the pure systems Pt and Au, there is fair agreement to other theoretical BCA-based calculations. Let us note here that for Pt and Au the experimental spin Hall angle α_{sH}, i.e., the ratio between the SHC and the longitudinal charge conductivity for pure systems, is discussed rather controversially, with large scatter in the reported data (Pt: $\alpha_{sH} = 0.37 \ldots 12$, Au: $\alpha_{sH} = 0.8 \ldots 11.3$). Therefore we omitted a detailed list of experimental values in Table II and refer the interested reader to a recent compilation of experimental data [75]. Note further that in the case of the SHC for disordered systems the vertex corrections also vanish numerically in the Fermi-sea term, as has been observed for the AHC (see Table II). This can be seen as a result of the particular construction of the vc Eq. (A2), as these are only expressed in terms of scattering path operators τ and are independent of the chosen operators for the matrix elements.

In Fig. 2 we show the concentration-dependent SHC of the alloy Cu_xAu_{1-x}. For this system the total SHC is essentially given by the Fermi-surface term $\sigma_{xy}^{z1,I}$, with the site-diagonal and Fermi-sea term giving almost negligible contribution and having the largest relative contribution in the middle of the concentration range. The large diverging scattering contributions for $x \to 0$ and $x \to 1$ are of incoherent (extrinsic) origin, an observation already made for other dilute alloys [25]. A comment concerning both AHC and SHC in dilute alloys seems in due place here: together with the divergence of the SHC at the boundaries, also the longitudinal conductivities will diverge such that the ratio $\sigma_{xy}^{(z)}/\sigma_{xx}$, namely, the anomalous or spin Hall angle, that is usually determined in experiment, will have a finite value. Furthermore, as the intrinsic contribution

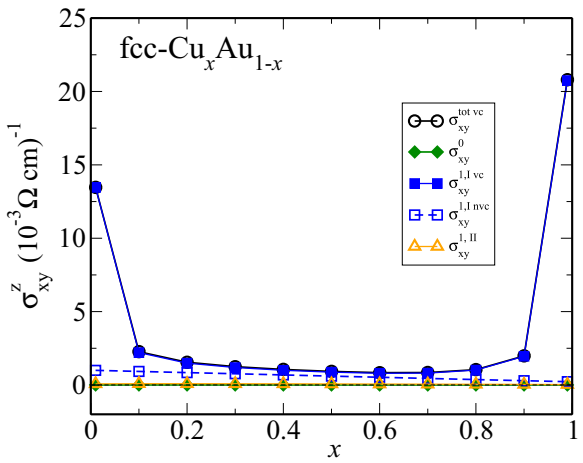

FIG. 2. (Color online) The SHC of Cu_xAu_{1-x} as a function of concentration x determined within the Kubo-Bastin formalism. The total SHC and different contributions (notation: see text and Fig. 1 caption) to it are displayed.

to the AHC/SHC for a perfect crystal (no disorder) is finite, its contribution to the Hall angle at $T = 0$ K will vanish. At finite temperature, however, induced scattering by lattice vibrations or impurities will lead to a finite σ_{xx} and indeed, experimental data for the AHE in metallic systems is often obtained by varying temperature or by doping.

V. SUMMARY

We presented a general linear response Kubo-Bastin approach and a subsequent implementation within a first-principles multiple-scattering Green's function method. The so-called Fermi-surface and Fermi-sea contributions are both treated on equal footing, employing a fully relativistic formulation spin-orbit-induced phenomena, particularly transverse transport quantities as the anomalous and spin Hall effect are properly described. The derived transport expression gives all elements of the (conductivity) tensor, namely, the symmetric and, in particular, antisymmetric components. Furthermore, the approach is not only able to deal with pure systems, but, using the CPA, substitutionally disordered alloys of any concentration can be treated, thereby avoiding inferior approximations as the virtual crystal approximation (VCA) and/or large supercells. The described method is able to capture both intrinsic as well as extrinsic (e.g., side-jump and skew-scattering) contributions to the transport tensors consistently within one and the same formulation. Vertex corrections (within the CPA) are fully taken into account. We presented applications for the AHE and SHE and discussed the various contributions to the (spin) transport tensors for pure systems as well as a number of transition-metal alloys.

As the derived expression within the KKR(-CPA) factorizes into matrix elements of the chosen operators and products of scattering path operators, the method can be straightforwardly adapted to deal with a number of linear response quantities by simply replacing the matrix elements. This concerns, e.g., the Gilbert damping [38] or spin-orbit torques. Spin-caloritronic quantities (e.g., spin and anomalous Nernst effects) will be accessible with minor effort within the presented Kubo-Bastin approach.

Finally, we want to point out that finite-temperature effects can be easily taken into account, as has already been done in Kubo-Greenwood–like formulations for longitudinal transport and Gilbert damping [38,78] using an alloy analogy model for lattice vibrations and spin fluctuations.

ACKNOWLEDGMENT

The authors would like to thank the Deutsche Forschungsgemeinschaft (DFG) for financial support within the SFB 689 and SPP 1538.

APPENDIX A: KKR-CPA TRANSPORT FORMALISM FOR THE KUBO-BASTIN FORMULATION

Starting from Eqs. (11) and (12) and due to the employed contour integration and the required energy derivative of the GF, matrix elements have to be calculated for pairs of complex energies. Note that in former approaches using the Kubo-Greenwood formulation for disordered alloys, calculations were performed on the real axis (symmetric surface term only). This very much simplified the expressions and implementation, as on the real axis the wave functions (Z_Λ and J_Λ) become real and one can neglect the second term in Eq. (13) containing the irregular solutions [see also Eq. (7)]. Furthermore, phase relations have been used to relate wave functions with energy $z = (\lim_{\eta\to 0^+} E_F + i\eta)$ to those with $z = \lim_{\eta\to 0^+}(E_F - i\eta)$, leading to transformation relations between matrix elements for the $\langle jG^+jG^+\rangle$, $\langle jG^+jG^-\rangle$, $\langle jG^-jG^+\rangle$, and $\langle jG^-jG^-\rangle$ terms in Eq. (11). Away from the real axis (i.e., when evaluating the Fermi-sea contribution and distorting the integration path for the energy into the complex plane) these are not applicable anymore for arbitrary operator pairs. For the τ matrix the following relation is, however, valid,

$$\tau^{nm}_{\Lambda\Lambda'}(z^*) = (-)^{l+l'}\left[\tau^{mn}_{\Lambda'\Lambda}(z)\right]^*, \tag{A1}$$

and can therefore be exploited. In what follows we work along the solution of the transport equation and notation introduced by Butler [21] and, however, extend it to the Kubo-Bastin formalism. For reasons of simplified notation, we here present only the case of having one atom per unit cell; the indices n and m therefore are numbering the unit cells in the crystal. With this the contributions to ΔG^n_ν Eq. (15) read as

$$\Delta G^{1,\alpha\beta,n}_\nu(\mathbf{r},\mathbf{r},z_a,z_b) = \sum_{\Lambda_1\Lambda_2\Lambda_3\Lambda_4} Z^{\alpha n}_{\Lambda_1}(\mathbf{r},z_a)\ j^{A\beta n}_{\nu\Lambda_2\Lambda_3}(r_{ws},z_a,z_b) Z^{\alpha n\times}_{\Lambda_4}(\mathbf{r},z_b)$$
$$\times \sum_{\Lambda_5\Lambda_6\Lambda_7\Lambda_8} \tilde{D}^{\alpha}_{\Lambda_8\Lambda_4}(z_b) D^{\alpha}_{\Lambda_1\Lambda_5}(z_a) \tilde{D}^{\beta}_{\Lambda_6\Lambda_2}(z_a) D^{\beta}_{\Lambda_3\Lambda_7}(z_b) \tilde{\chi}_{\Lambda_5\Lambda_6\Lambda_7\Lambda_8}(z_a,z_b),$$

$$\Delta G^{0,\alpha,n}_{\nu 1}(\mathbf{r},\mathbf{r},z_a,z_b) = \sum_{\Lambda_1\Lambda_2\Lambda_3\Lambda_4} Z^{\alpha n}_{\Lambda_1}(\mathbf{r},z_a)\; j^{A\beta n}_{\nu\Lambda_2\Lambda_3}(r_{ws},z_a,z_b) Z^{\alpha n\times}_{\Lambda_4}(\mathbf{r},z_b) \times \sum_{\Lambda_5\Lambda_6} D^{\alpha}_{\Lambda_1\Lambda_5}(z_a)\tilde{D}^{\alpha}_{\Lambda_6\Lambda_4}(z_a) \quad \bar{\tau}^{nn}_{\Lambda_5\Lambda_2}(z_a)\,\bar{\tau}^{nn}_{\Lambda_3\Lambda_6}(z_b),$$

$$\Delta G^{0,\alpha,n}_{\nu 2}(\mathbf{r},\mathbf{r},z_a,z_b) = -\sum_{\Lambda_1\Lambda_3\Lambda_4} \bar{\tau}^{nn\alpha}_{\Lambda_3\Lambda_4}(z_b)\left[J^{\alpha n}_{\Lambda_1}(\mathbf{r},z_a)\, Z^{\alpha n\times}_{\Lambda_4}(\mathbf{r},z_b)\, j^{A\alpha n}_{\nu\Lambda_1\Lambda_3}(r,z_a,z_b) + Z^{\alpha n}_{\Lambda_1}(\mathbf{r},z_a)\, Z^{\alpha n\times}_{\Lambda_4}(\mathbf{r},z_b)\, \bar{j}^{B\alpha n}_{\nu\Lambda_1\Lambda_3}(r,z_a,z_b)\right],$$

$$\Delta G^{0,\alpha,n}_{\nu 3}(\mathbf{r},\mathbf{r},z_a,z_b) = -\sum_{\Lambda_1\Lambda_2\Lambda_3} \bar{\tau}^{nn\alpha}_{\Lambda_1\Lambda_2}(z_a)\left[Z^{\alpha n}_{\Lambda_1}(\mathbf{r},z_a)\, J^{\alpha n\times}_{\Lambda_3}(\mathbf{r},z_b)\, j^{An}_{\nu\Lambda_2\Lambda_3}(r,z_a,z_b) + Z^{\alpha n}_{\Lambda_1}(\mathbf{r},z_a)\, Z^{\alpha n\times}_{\Lambda_3}(\mathbf{r},z_b)\, \bar{j}^{C\alpha n}_{\nu\Lambda_2\Lambda_3}(r,z_a,z_b)\right],$$

$$\Delta G^{0,\alpha,n}_{\nu 4}(\mathbf{r},\mathbf{r},z_a,z_b) = \sum_{\Lambda_1\Lambda_3}\left[J^{\alpha n}_{\Lambda_1}(\mathbf{r},z_a)\, J^{\alpha n\times}_{\Lambda_3}(\mathbf{r},z_b)\, j^{A\alpha n}_{\nu\Lambda_1\Lambda_3}(r,z_a,z_b) + Z^{\alpha n}_{\Lambda_1}(\mathbf{r},z_a)\, Z^{\alpha n\times}_{\Lambda_3}(\mathbf{r},z_b)\, \bar{j}^{D\alpha n}_{\nu\Lambda_1\Lambda_3}(r,z_a,z_b)\right],$$

with $\bar{\tau}$ denoting the CPA averaged τ matrix and the Greek indices α,β signify the atom type which occupies an atomic site n,m. In the last expression the auxiliary r-dependent quantities containing the charge current operator,

$$j^{A\alpha n}_{\nu\Lambda_1\Lambda_2}(r,z_a,z_b) = \int_0^r d^3r' Z^{\alpha n\times}_{\Lambda_1}(\mathbf{r}',z_a)\,\hat{j}_\nu Z^{\alpha n}_{\Lambda_2}(\mathbf{r}',z_b),$$

$$j^{B\alpha n}_{\nu\Lambda_1\Lambda_2}(r,z_a,z_b) = \int_0^r d^3r' J^{\alpha n\times}_{\Lambda_1}(\mathbf{r}',z_a)\,\hat{j}_\nu Z^{\alpha n}_{\Lambda_2}(\mathbf{r}',z_b),$$

$$j^{C\alpha n}_{\nu\Lambda_1\Lambda_2}(r,z_a,z_b) = \int_0^r d^3r' Z^{\alpha n\times}_{\Lambda_1}(\mathbf{r}',z_a)\,\hat{j}_\nu J^{\alpha n}_{\Lambda_2}(\mathbf{r}',z_b),$$

$$j^{D\alpha n}_{\nu\Lambda_1\Lambda_2}(r,z_a,z_b) = \int_0^r d^3r' J^{\alpha n\times}_{\Lambda_1}(\mathbf{r}',z_a)\,\hat{j}_\nu J^{\alpha n}_{\Lambda_2}(\mathbf{r}',z_b),$$

$$\bar{j}^{X\alpha n}_{\nu\Lambda_1\Lambda_2}(r,z_a,z_b) = j^{X\alpha n}_{\nu\Lambda_1\Lambda_2}(r_{ws},z_a,z_b) - j^{X\alpha n}_{\nu\Lambda_1\Lambda_2}(r,z_a,z_b), \quad \text{for} \quad X = A, B, C, D$$

have been used, where r_{ws} denotes the Wigner-Seitz (or ASA) radius. The following standard definitions [21,64] for the auxiliary matrices $D^\alpha,\tilde{D}^\alpha,x^\alpha,\Delta m^\alpha$, and $\tau^{nn\alpha}$ are employed:

$$D^\alpha = 1 + \bar{\tau}^{00}x^\alpha, \quad \tilde{D}^\alpha = 1 + x^\alpha\bar{\tau}^{00},$$
$$x^\alpha = [1 - \Delta m^\alpha\,\bar{\tau}^{00}]^{-1}\,\Delta m^\alpha,$$
$$\Delta m^\alpha = \bar{m} - m^\alpha, \quad \tau^{nn\alpha} = D^\alpha\bar{\tau}^{00} = \bar{\tau}^{00}\tilde{D}^\alpha,$$

where $\bar{m} = \bar{t}^{-1}$, with $\bar{t}$ being the CPA average of the single-site t matrices t^α and $m^\alpha = [t^\alpha]^{-1}$. In the solution of the transport equations the quantity

$$\tilde{\chi}_{\Lambda_1\Lambda_2\,\Lambda_3\Lambda_4} = \tilde{\chi}_{K_1K_2} = \{[1 - \chi\; w]^{-1}\chi\}_{K_1K_2}, \qquad \text{(A2)}$$

where the combined indices $K_1 = (\Lambda_1\Lambda_4), K_2 = (\Lambda_2\Lambda_3)$ play a crucial role as they contain the vertex corrections. The auxiliary quantity χ is given by

$$\chi_{K_1K_2} = \chi_{\Lambda_1\Lambda_2\,\Lambda_3\Lambda_4}(z_a,z_b) = \sum_{m,m\neq n} \bar{\tau}^{nm}_{\Lambda_1\Lambda_2}(z_a)\,\bar{\tau}^{mn}_{\Lambda_3\Lambda_4}(z_b) = \left[\frac{1}{\Omega_{\mathrm{BZ}}}\int_{\Omega_{\mathrm{BZ}}} \bar{\tau}_{\Lambda_1\Lambda_2}(\mathbf{k},z_a)\bar{\tau}_{\Lambda_3\Lambda_4}(\mathbf{k},z_b)\,d^3k\right] - \bar{\tau}^{nn}_{\Lambda_1\Lambda_2}(z_a)\,\bar{\tau}^{nn}_{\Lambda_3\Lambda_4}(z_b), \qquad \text{(A3)}$$

and is obtained via an integral over the BZ and results from the assumed periodicity of the CPA medium after a Fourier transformation. The determination of the four index quantity χ and the inversion Eq. (A2) are computationally very demanding, in particular, when many k points are needed and with growing system size (number of atoms per unit cell). A scheme to exploit symmetry when dealing with the BZ integral Eq. (A3) has been worked out previously [79]. This allows us to restrict $\chi_{K_1K_2}$ to its nonzero elements and to integrate only over the irreducible part of the BZ. The interaction term w is given as

$$w_{\Lambda_1\Lambda_2\,\Lambda_3\Lambda_4}(z_a,z_b) = w_{K_1K_2}(z_a,z_b) = \sum_\alpha c_\alpha x^\alpha_{\Lambda_1\Lambda_2}(z_a)x^\alpha_{\Lambda_3\Lambda_4}(z_b)\,,$$

where c_α in the last expression denotes the concentration of the alloy partner α (denoted as x_α in the main text).

Setting w to zero in Eq. (A2) amounts to neglecting the vertex corrections. Further note, however, that the formalism is equally well applicable to pure systems. In that case $\chi = \tilde{\chi}$ because $w = 0$.

APPENDIX B: MATRIX ELEMENTS

The regular and irregular solutions of Eq. (1) are expanded into four spinors of the form [80,81]

$$Z_\Lambda(\mathbf{r}) = \sum_{\Lambda'}\begin{pmatrix} g_{\Lambda'\Lambda}(r)\chi_{\Lambda'}(\hat{\mathbf{r}}) \\ if_{\Lambda'\Lambda}(r)\chi_{-\Lambda'}(\hat{\mathbf{r}})\end{pmatrix}, \qquad \text{(B1)}$$

where g and f are the radial functions of the large and small components, respectively, and χ_Λ are the usual spin-angular functions [34], being linear combinations of products of complex spherical harmonics Y_l^m and the spin functions $\chi_{m_s}, m_s \in \{-\frac{1}{2},+\frac{1}{2}\}$. The quantum number Λ in the latter expression is used to label the states which can have mixed spin-angular character. We use the notation $-\Lambda = (-\kappa,\mu)$.

Both, the AHE and SHE in the linear response framework originate from a perturbation given by the charge current. Within the relativistic framework used here the current operator is represented by $\hat{\mathbf{j}} = -|e|c\boldsymbol{\alpha}$. Therefore, matrix elements of the Dirac α_μ have to be evaluated. Note, when calculating these matrix elements significant errors can be introduced when using the shape approximation in the form of the ASA.

Here we use a scheme proposed by Shilkova and Shirokovskii [82] that has already been used before to correct for these errors. This can be checked by comparing to yet another form of the matrix elements for α that has been derived before [83] and relies on rewriting the matrix elements using the anticommutator $[\hat{H}_{\mathrm{D}},\boldsymbol{\alpha}]_+$ into an equivalent form containing the momentum operator $\hat{\mathbf{p}}$.

The calculations of matrix elements of the spin-polarization current-density operator Eq. (16) is naturally split into two components. The first component contains products of β,α_μ and Σ_z matrices which can be simplified using

$$\beta\Sigma_z\alpha_\mu = i\varepsilon_{z\mu\nu}\begin{pmatrix}0 & -\sigma_\nu\\ \sigma_\nu & 0\end{pmatrix},\quad \mu\in\{x,y\},\quad \nu\neq z,\mu, \tag{B2}$$

where in the last expression ε_{ijk} is the Levi-Civita symbol with the understanding of the mapping $x\to 1$, $y\to 2$, $z\to 3$ for the coordinate directions. By inspection of Eq. (2) it is seen that the matrix elements Eq. (B2) can be easily computed using the existing matrix elements of the current operator containing the α_μ matrices.

The second part involves matrix elements of the operator $\gamma_5\hat{p}_z\alpha_\mu$, i.e.,

$$\gamma_5\hat{p}_z\alpha_\mu = -\frac{\hbar}{i}\nabla_z\Sigma_\mu\,, \tag{B3}$$

which are evaluated using the gradient formula of Ref. [33], Eq. (2.57):

$$\begin{aligned}\nabla_M\,\phi_l(r)\,Y_l^m(\hat{\mathbf{r}}) = &\sqrt{\frac{l+1}{2l+3}}\,C(l\,1,l+1;m\,M)\\ &\times Y_{l+1}^{m+M}(\hat{r})\left[\frac{d\phi_l(r)}{dr}-\frac{l}{r}\phi_l(r)\right]\\ &-\sqrt{\frac{l}{2l-1}}\,C(l\,1,l-1;m\,M)\\ &\times Y_{l-1}^{m+M}(\hat{r})\left[\frac{d\phi_l(r)}{dr}+\frac{l+1}{r}\phi_l(r)\right],\end{aligned}$$

with $M\in\{-1,0,1\}$ denoting a spherical coordinate, ϕ_l a radial function, Y_l^m a complex spherical harmonic, and $C(j_1j_2j;m_1m_2)$ being a Clebsch-Gordan coefficient. (For phase conventions and definitions employed, see Ref. [33].) To use the latter formula the vector operator components Eq. (B3) have to be transformed from Cartesian coordinates {x,y,z} into spherical coordinates $\{-1,0,1\}$ using $A_{\pm1}=\mp\frac{1}{\sqrt{2}}(A_{\mathrm{x}}\pm iA_{\mathrm{y}}), A_0=A_{\mathrm{z}}$, both for the momentum operator and the relativistic Pauli-spin operator Σ_μ.

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