Ab initio theory of galvanomagnetic phenomena in ferromagnetic metals and disordered alloys

I. Turek

Institute of Physics of Materials, Academy of Sciences of the Czech Republic, Žižkova 22, CZ-616 62 Brno, Czech Republic

J. Kudrnovský and V. Drchal

Institute of Physics, Academy of Sciences of the Czech Republic, Na Slovance 2, CZ-182 21 Praha 8, Czech Republic

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We present an ab initio theory of transport quantities of metallic ferromagnets developed in the framework of the fully relativistic tight-binding linear muffin-tin orbital method. The approach is based on the Kubo-Středa formula for the conductivity tensor, on the coherent potential approximation for random alloys, and on the concept of interatomic electron transport. The developed formalism is applied to pure 3d transition metals (Fe, Co, Ni) and to random Ni-based ferromagnetic alloys (Ni-Fe, Ni-Co, Ni-Mn). High values of the anisotropic magnetoresistance (AMR), found for Ni-rich alloys, are explained by a negligible disorder in the majority spin channel while a change of the sign of the anomalous Hall effect (AHE) on alloying is interpreted as a band-filling effect without a direct relation to the high AMR. The influence of disorder on the AHE in concentrated alloys is investigated as well.

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I. INTRODUCTION

Two galvanomagnetic phenomena discovered in the 19th century, namely, the anisotropic magnetoresistance (AMR) and the anomalous Hall effect (AHE) in ferromagnets, attract ongoing interest both in basic and applied physics. The AMR of bulk systems together with the giant magnetoresistance of magnetic multilayers plays an important role in magnetic data storage whereas the AHE is not fully understood at present despite the tremendous past and recent research activity. However, the basic origin of both phenomena is well known for a long time and it was identified with the simultaneous presence of spin polarization and spin-orbit (SO) interaction. Among open problems, one can list, e.g., correlations between the anomalous Hall conductivities and the longitudinal conductivities reported for a number of systems or the occurrence of large AMR values and the change of sign of the AHE at the same composition (around 15 at. % Fe) of random NiFe alloys. Let us mention that the AMR and AHE are related, respectively, to the symmetric and antisymmetric parts of the resistivity tensor of the solid. For this reason, an internally consistent theory of all transport phenomena (resistivity, AMR, AHE, etc.) represents an important task of condensed-matter physics.

Existing theoretical techniques for investigation of transport properties include the Boltzmann equation and the Kubo linear response theory which represent appropriate tools to describe electron scattering on impurities or phonons. The latter approach uses the Kubo-Greenwood formula for longitudinal resistivities and the AMR while the Kubo-Středa formula is a starting point for the AHE. However, the AHE contains not only an extrinsic contribution due to the electron scattering, but also an intrinsic one that is solely due to the band structure of an ideal crystal. The intrinsic AHE seems to dominate over the extrinsic part in a number of cases; its values are related to the Berry curvatures of the Bloch states of the crystal. These theoretical concepts and techniques have recently been used to address various problems on a model level, such as, e.g., to describe quantitatively the AHE and AMR in diluted magnetic semiconductors or to develop a unified scheme for the AHE of systems with high and low conductivities.

Materials-specific ab initio theory of galvanomagnetic phenomena is a straightforward extension of the model-level techniques only for the intrinsic AHE of pure magnetic crystals. Transport properties due to impurity scattering, in particular residual resistivities, of substitutionally disordered alloys without the SO interaction have been systematically studied by means of the Korringa-Kohn-Rostoker (KKR) method and the coherent potential approximation (CPA). An alternative first-principles technique has recently been developed within the tight-binding (TB) linear muffin-tin orbital (LMTO) method. Both approaches agree quantitatively for resistivities of metallic alloys and diluted magnetic semiconductors. The AMR of random cubic alloys – based on diagonal elements of the conductivity tensor, i.e., on the Kubo-Greenwood formula – have been studied in the fully relativistic KKR method. Similar results of the TB-LMTO method with the SO interaction included compare well to those of the KKR method again. These studies prove that the SO interaction can have a dramatic effect also on residual resistivities of ferromagnetic alloys of light (3d) elements.
An early attempt to calculate the AHE in random alloys from the Kubo-Greenwood formulaa used the KKR-CPA method was shown to be incorrect. A correct formulation based on the Kubo-Středa formula has appeared only very recently. The authors of Ref. 26 have suggested to interpret the coherent part of the anomalous Hall conductivity in a random alloy as the intrinsic contribution to the AHE while the incoherent part – the so-called vertex corrections – has been identified with the extrinsic contribution to the AHE. Moreover, they have studied the case of dilute alloys and have shown that the intrinsic contribution exhibits a well-defined limit for vanishing concentration of impurities in contrast to a divergence of the extrinsic contribution, in reasonable agreement with experimental data for FePd and NiPd random alloys.

The purpose of the present paper is to formulate the full conductivity tensor of ferromagnetic metals and substitutionally disordered alloys in the relativistic TB-LMTO-CPA method in the atomic sphere approximation (ASA) and to illustrate its applicability to the galvanomagnetic phenomena of systems containing 3$d$ transition-metal elements (Fe, Co, Ni, Mn). The paper is organized as follows. The developed method is presented in Section II. Section II A summarizes the most important relations of the fully relativistic TB-LMTO formalism relevant for the subsequent development of the transport theory, which is given in Section II B. Technical parts of the derivations are left to Appendices while details of the numerical procedures employed can be found in Section II C. The calculated results for selected systems and their discussion are contained in Section III. The AMR of Ni-based alloys is discussed in Section III A while Sections III B and III C are devoted to the AHE in pure metals and in random alloys, respectively. The main conclusions of the paper are summarized in Section IV.

II. METHOD

A. Relativistic TB-LMTO-ASA method

The Hamiltonian in the orthogonal LMTO representation for a ferromagnetic system treated in the fully relativistic LMTO-ASA method can be written as:

$$H = C + (\sqrt{\Delta})^+ S^0 (1 - \gamma S^0)^{-1} \sqrt{\Delta},$$

(1)

where the $C$, $\sqrt{\Delta}$ and $\gamma$ are site-diagonal matrices of potential parameters and the $S^0$ denotes the matrix of canonical structure constants. The form of the Hamiltonian $H$ is similar to the non-relativistic or non-magnetic cases, however, the structure of $H$ is more complicated in the spin-polarized relativistic case for two reasons. First, the matrices involved in Eq. (1) have different kinds of indices: the site index $R$ is combined either with the usual relativistic index $\Lambda = (\kappa\mu)$, or with a composed index $\hat{\Lambda} = (\ell\mu\lambda)$. Here the quantum number $\mu$ is related to $z$-component of the total angular momentum, the $\ell$ is related to the orbital angular momentum, the non-zero integer $\kappa$ is related in a well-known manner to the total angular momentum quantum number $j$ ($2j + 1 = 2|\kappa|$) and to the $\ell$ ($2\ell + 1 = 2\kappa + 1$), and the index $\lambda$ labels inequivalent regular solutions of the single-site problem in each $\ell\mu$-channel.

The Hamiltonian matrix has thus indices $H_{R\Lambda'} R\Lambda = H_{R\ell'\mu'\lambda'} R\ell\mu\lambda$ while the structure constant matrix has indices $S^0_{R\Lambda' R\Lambda} = S^0_{R\kappa'\mu' R\kappa\mu}$. Second, the site-diagonal matrices $C$, $\sqrt{\Delta}$ and $\gamma$ are not fully diagonal in the corresponding internal indices, $(\kappa\mu)$ or $(\ell\mu\lambda)$, see Refs. 27,29 for details. Moreover, the $\sqrt{\Delta}$ is defined as a matrix in the mixed indices $(\kappa'\mu', \ell\mu\lambda)$ which cannot be understood as a square root of any matrix $\Delta$. However, for the transport theory developed in Section II B the detailed structure of the matrices is less important and only their general properties are relevant, such as, e.g., the hermiticity of $C$, $\gamma$ and $S^0$.

The treatment of transport properties of disordered systems requires the Green’s functions (GF). The basic one – called the physical GF – is defined as the resolvent of the Hamiltonian $H$:

$$G(z) = (z - H)^{-1},$$

(2)

where $z$ denotes a complex energy variable. Other GF’s are introduced in the TB-LMTO formalism. The matrix of screened structure constants $S^\alpha$ in the TB-LMTO representation (superscript $\alpha$) is defined by:

$$S^\alpha = S^0 (1 - \alpha S^0)^{-1},$$

(3)

where $\alpha$ denotes a site-diagonal matrix of screening constants, and the site-diagonal matrix of screened potential functions $P^\alpha(z)$ is defined as:

$$P^\alpha(z) = \left[\sqrt{\Delta}(z - C)^{-1}(\sqrt{\Delta})^+ + \gamma - \alpha\right]^{-1}.$$

(4)
The auxiliary GF in the TB-LMTO representation is then defined as
\[ g^\alpha(z) = [P^\alpha(z) - S^\alpha]^{-1}, \]  
which represents a simpler quantity for theoretical and numerical treatments than the physical GF \( G(z) \), Eq. (2). Both GF’s are related to each other by linear rescaling
\[ G(z) = \lambda^\alpha(z) + \mu^\alpha(z)g^\alpha(z)\tilde{\mu}^\alpha(z), \]
where the quantities \( \lambda^\alpha(z), \mu^\alpha(z) \) and \( \tilde{\mu}^\alpha(z) \) denote site-diagonal matrices
\[ \lambda^\alpha(z) = \mu^\alpha(z)(\gamma - \alpha), \]
\[ \mu^\alpha(z) = (\sqrt{\Delta})^{-1}[1 + (\alpha - \gamma)P^\alpha(z)], \]
\[ \tilde{\mu}^\alpha(z) = [1 + P^\alpha(z)(\alpha - \gamma)]\left[(\sqrt{\Delta})^+\right]^{-1}. \]

The relation (6) is indispensable for an efficient treatment of bulk and layered systems, its proof is sketched in Appendix A. The substitutional disorder in a system on a fixed, non-random lattice can be best treated in the CPA applied to the auxiliary GF (5). The configurationally averaged \( g^\alpha(z) \) is then given by
\[ \langle g^\alpha(z) \rangle = \bar{g}^\alpha(z) = \left[P^\alpha(z) - S^\alpha \right]^{-1}, \]
where the \( P^\alpha(z) \) denotes the site-diagonal matrix of the coherent potential functions. Their determination from the CPA selfconsistency condition in the relativistic spin-polarized case as well as details of the LSDA selfconsistent procedure can be found elsewhere.28,29

### B. Full conductivity tensor

The conductivity tensor at zero temperature is given according to the Kubo-Streda formul\( \text{a}^29 \) as
\[ \sigma_{\mu\nu} = \sigma_0 \text{Tr} \{ V_\mu (G_+ - G_-) V_\nu G_- - V_\mu G_+ V_\nu (G_+ - G_-) + i (X_\mu V_\nu - X_\nu V_\mu) (G_+ - G_-) \}, \]
where the subscripts \( \mu \) and \( \nu \) denote indices of Cartesian coordinates \( (\mu, \nu \in \{x, y, z\}) \), the trace (Tr) is taken over all orbitals of the system, the energy argument of the GF’s is omitted since it equals the Fermi energy \( E_F \), and we abbreviated \( G_\pm = G(E_F \pm i\epsilon) \). The symbols \( X_\mu \) and \( V_\mu \) denote, respectively, the coordinate and velocity operators. The numerical prefactor \( \sigma_0 \) reflects the units employed and the size of the sample; with \( \hbar = 1 \) assumed here, it is given by \( \sigma_0 = e^2/(4\pi V_0 N) \), where \( V_0 \) is the volume of the primitive cell and \( N \) is the number of cells in a big finite crystal with periodic boundary conditions.

In analogy to the non-relativistic formulation of electron transport,20 the coordinate operator \( X_\mu \) is represented – in an orthonormal LMTO basis leading to the Hamiltonian (1) – by a matrix diagonal in the \((R\Lambda)\)-index as
\[ (X_\mu)_{R\Lambda,R\Lambda} = \delta_{R\Lambda} \delta_{\Lambda\Lambda} X^\mu_R, \]
where \( X^\mu_R \) is the \( \mu \)-th component of the position vector \( R \). The velocity operator \( V_\mu \) is then defined as a quantum-mechanical time derivative of \( X_\mu \) (\( \hbar = 1 \)):
\[ V_\mu = -i [X_\mu, H], \]
where \([A,B] = AB - BA\) denotes a commutator. The physical idea behind the simple rule (10) is an approximation of the true continuous coordinate by its step-like “integer” part that is constant inside each atomic sphere. This leads to a systematic neglect of any intraatomic currents so that the resulting conductivity \( \sigma_{\mu\nu} \) describes only the net electron motion between neighboring atomic sites.21 The final simple result for the \( X_\mu \) (10) is then obtained from basic properties of the phi orbitals \( |\phi_{R\Lambda}\rangle \) and phi-dot orbitals \( |\dot{\phi}_{R\Lambda}\rangle \) used in the definition of the orthonormal LMTO basis.20,31 In particular, one employs their orthonormality, \( \langle \phi_{R\Lambda} | \phi_{R\Lambda} \rangle = \delta_{R\Lambda} \delta_{\Lambda\Lambda} \) and \( \langle \phi_{R\Lambda} | \dot{\phi}_{R\Lambda} \rangle = 0 \), together with a neglect of small quantities \( \langle \dot{\phi}_{R\Lambda} | \phi_{R\Lambda} \rangle \) which is consistent with the second-order accuracy of the LMTO Hamiltonian (1).
For practical calculations, one can recast the original form of the conductivity tensor $\sigma_{\mu\nu}$ into a more suitable version in which the velocities $V_\mu$  are replaced by effective velocities $v_\mu^\alpha$ defined by
\begin{equation}
  v_\mu^\alpha = -i [X_\mu, S^\alpha],
\end{equation}
and in which the physical GF's $G_\pm$ are replaced by the auxiliary GF's $g_\pm^\alpha = g^\alpha (E_F \pm i0)$, see Eq. (5). This transformation rests on two relations. The first one connects both velocity operators by
\begin{equation}
  V_\mu = (\sqrt{\Delta})^+ (F^\alpha)^{-1} v_\mu^\alpha ((F^\alpha)^+)^{-1} \sqrt{\Delta},
\end{equation}
where
\begin{equation}
  F^\alpha = 1 + S^\alpha (\alpha - \gamma)
\end{equation}
and, consequently, $(F^\alpha)^+ = 1 + (\alpha - \gamma) S^\alpha$. The velocity relation (13) can easily be obtained from the explicit form of $H$ and from an identity $S^0 (1 - \gamma S^0)^{-1} = (F^\alpha)^{-1} S^\alpha (F^\alpha)^+^{-1}$, which is a direct consequence of the screening transformation (3). Note that the coordinate operator $X_\mu$ in the definition of the effective velocity $v_\mu^\alpha$ (12) has to be understood as a diagonal matrix in the $(\text{RA})$-index, i.e.,
\begin{equation}
  (X_\mu)_{\text{RA}R^\alpha A} = \delta_{\text{RA} R^\alpha} \delta_{A \Lambda} X^\mu_{\Lambda R},
\end{equation}
and that both operators $X_\mu$, Eqs. (11) and (15), commute with all other site-diagonal operators $(C, \sqrt{\Delta}, \gamma, \alpha)$. The second relation connects the physical and auxiliary GF's by
\begin{equation}
  G(z) = (\sqrt{\Delta})^{-1} (F^\alpha)^+ [((\alpha - \gamma) + g^\alpha (z) F^\alpha)] ((\sqrt{\Delta})^+)^{-1},
\end{equation}
which represents a complementary relation to Eq. (9) and which is proved in a similar way in Appendix A. The use of Eqs. (13) and (16) in the conductivity tensor (9) yields another form of the latter, namely,
\begin{equation}
  \sigma_{\mu\nu} = \sigma_0 \text{Tr} \{ v_\mu^\alpha (g^\alpha_{\mu\nu} - g^\nu_{\mu\nu}) v_\nu^\alpha - v_\mu^\alpha g^\nu_{\mu\nu} v_\nu^\alpha (g^\alpha_{\mu\nu} - g^\nu_{\mu\nu}) + i (X_\mu v_\nu^\alpha - X_\nu v_\mu^\alpha) (g^\alpha_{\mu\nu} - g^\nu_{\mu\nu}) \}.
\end{equation}
The proof of equivalence of both expressions for the $\sigma_{\mu\nu}$ is given in Appendix E.

The similarity of both conductivity formulas, Eqs. (9) and (17), is obvious. However, the latter one is better suited for configuration averaging since the effective velocities $v_\mu^\alpha$ (12) are non-random operators in contrast to the original velocities $V_\mu$ (11) and to the velocities in the KKR-CPA method. The CPA-average of the $\sigma_{\mu\nu}$ (17) can thus be reduced to averages of the auxiliary GF’s (8) and to the standard form of vertex corrections for the CPA-average of products $g^\alpha (z_1) \otimes g^\alpha (z_2)$. The same transformation was achieved in our previous study for longitudinal conductivities within the Kubo-Greenwood formula and the non-relativistic TB-LMTO method. The derived result, Eq. (17), represents an extension in two directions: to spin-polarized relativistic systems and to the full conductivity tensor, including the anomalous Hall conductivities.

C. Implementation and numerical details

The fully relativistic theory developed in Section II B has been applied to random binary alloys of 3d transition metals (Mn, Fe, Co, Ni), where the effects of the SO interaction are rather weak as compared to the alloy bandwidths and exchange splittings. For this reason, most of the results shown in the next section were calculated by means of a simplified version of the theory, in which the SO coupling was included as an on-site perturbation term of the $\xi \mathbf{L} \cdot \mathbf{S}$ form to the LMTO Hamiltonian in the scalar-relativistic approximation (SRA). This SRA+SO approach proved to yield results in surprisingly good quantitative agreement with results of fully relativistic LMTO and KKR techniques, both for magnetic moments and densities of states and for longitudinal resistivities of 3d transition-metal systems. In a few selected cases, results of the fully relativistic Dirac (FRD) theory will be shown for a direct comparison of both approaches.

The employed LMTO basis comprised $s$, $p$, and $d$-type orbitals, and the LSDA selfconsistency was achieved with the Vosko-Wilk-Nusair exchange-correlation potential and for magnetization pointing along $z$-axis of the alloy cubic structures. More details about the ground-state electronic structure calculations were published elsewhere. The numerical prefactor $\sigma_0$ used in the final conductivity formula, Eq. (17), is given by $\sigma_0 = e^2/(4\pi \hbar V_0 N)$, where $N$ is the number of $\mathbf{k}$ vectors sampling the whole Brillouin zone (BZ). The terms bilinear in the auxiliary GF’s $g_\pm$ in Eq. (17) were configurationally averaged in the CPA with the corresponding vertex corrections included, whereas
FIG. 1. (Color online) The anomalous Hall conductivity $\sigma_{xy}$ of fcc Ni versus the imaginary part of energy $\varepsilon$ for different numbers $N$ of $k$ vectors in the full BZ. The converged value obtained by methods based on the Berry curvature is $\sigma_{xy} = 22.0 \times 10^4$ S/m, see Ref. 17 and Table II below.

the term linear in the $g_{\pm}$ has been omitted here for symmetry reasons (it vanishes identically due to the inversion symmetry of the Bravais lattice). In a general case, the latter term in Eq. (17) is essentially equivalent to the so-called Fermi-sea contribution to the AHE, which can be neglected in metallic systems.

Particular attention was paid to convergence behavior of the conductivities with respect to the number $N$ of sampling $k$ points. We found that large $N$’s were necessary and that energy arguments of the GF’s had to contain a non-zero imaginary part, $\varepsilon = E_F \pm i\varepsilon$. The quantity $\varepsilon$ should be as small as possible for purely theoretical values of $\sigma_{\mu\nu}$; however, for a comparison with experiment, finite values of $\varepsilon$ can also be used in order to simulate additional scattering on phonons, magnons and defects in real samples.

A. AMR in random fcc Ni-based alloys

For cubic systems with magnetization along $z$-axis, the conductivity tensor has three independent non-zero matrix elements, namely, $\sigma_{xx} = \sigma_{yy}$, $\sigma_{zz}$, and $\sigma_{xy} = -\sigma_{yx}$; the same structure is found for the resistivity tensor $\rho_{\mu\nu}$. The AHE is obtained quantitatively from the non-diagonal elements, $\sigma_{xy}$ or $\rho_{xy}$, while the diagonal elements define the isotropic resistivity $\rho = (2\rho_{xx} + \rho_{zz})/3$ and the AMR ratio $\Delta\rho/\rho$, where $\Delta\rho = \rho_{zz} - \rho_{xx}$.

III. RESULTS AND DISCUSSION

A brief summary of the present TB-LMTO results and their comparison to available experimental data is shown in Table I for three Ni-based ferromagnetic alloys with the same average number of electrons per atom $\bar{Z} = 27.7$. One can see that the resistivities of the Ni-Co and Ni-Fe alloys are smaller by a factor of two to four as compared to experimental values. Similar underestimation of the resistivity values has been obtained by the relativistic KKR-CPA method while the scattering theory combined with a supercell TB-LMTO approach yields resistivities that are only $\sim 20\%$ smaller than the measured ones. The calculated AMR values of Ni-Co and Ni-Fe values exceed slightly the measured ones while the opposite discrepancy is found for the ferromagnetic Ni-Mn system: for a Ni$_{0.97}$Mn$_{0.03}$ alloy, the measured AMR amounts to 7.3%, whereas the present calculation yields the AMR of 3.3% and 2.4% in the FRD and the SRA+SO approach, respectively.

The first application of the developed scheme has been done for the AMR of fcc Ni-based alloys. A preliminary study of the Ni-Co and Ni-Cu alloy systems including a comparison with the results of the KKR method was published elsewhere here we show the AMR for the fcc Ni$_{1-x}$Fe$_x$ alloy calculated in the SRA+SO approach, see Fig. 2. One can see relatively high values of the AMR for the Ni-rich alloys with the maximum AMR ratio slightly exceeding 20%. Similar trends and values were obtained in the fully relativistic KKR method and in experiments.
TABLE I. Comparison of the calculated and measured transport quantities for three fcc Ni-based alloys with the average number of electrons per atom $\bar{Z} = 27.7$: the isotropic resistivity $\rho$ (in $\mu\Omega\text{cm}$), the AMR ratio and the anomalous Hall conductivity $\sigma_{xy}$ (in kS/m). The displayed calculated values correspond to the FRD approach; in parentheses, results of the SRA+SO approach are shown. The experimental values are taken from Ref. [42].

| alloy          | $\rho$         | AMR    | $\sigma_{xy}$   |
|---------------|----------------|--------|-----------------|
| Ni$_{0.7}$Co$_{0.3}$ calc. | 1.06(0.70)     | 0.47(0.42) | $-88(-74)$ |
| exp.          | 2.8            | 0.3    |                 |
| Ni$_{0.85}$Fe$_{0.15}$ calc. | 2.21(1.57)     | 0.29(0.24) | $-40(-29)$ |
| exp.          | 4.2            | 0.18   |                 |
| Ni$_{0.9}$Mn$_{0.1}$ calc. | 22.8(23.6)     | 0.009(0.004) | 65(54)  |

FIG. 2. (Color online) The calculated values of the anisotropic magnetoresistance of random fcc Ni$_{1-x}$Fe$_x$ alloy as a function of Fe concentration and their comparison to experimental values [42].

The high AMR values for the Ni-rich Ni-Fe alloys and even higher theoretical values as well as experimental values for the Ni-rich Ni-Co alloys deserve attention and call for explanation. Existing theoretical approaches to the AMR in metals and alloys are based on inclusion of the SO interaction in the lowest-order perturbation expansion [42,47,48]. However, the validity of such schemes for the Ni-rich alloys is limited due to the well-known fact that the SO interaction increases the resistivity by a factor of two or more as compared to the resistivity within the two current model [23,25]. In order to reveal the possible origin of the high AMR in these systems, we considered also the fcc Ni-Mn alloy with a ferromagnetic order in the whole concentration range studied (which represents the true ground state only for Mn concentrations below 15 at.% Mn [49,50]) and an artificial fcc Ni-Fe(*) alloy in which the majority spin (spin-up) potential of Fe atoms was replaced by that of Ni atoms leading thus to a system with no spin-up disorder. The real Ni-Co and Ni-Fe alloys are featured by a very weak disorder in the majority spin channel [23], the four ferromagnetic alloys considered in the present study form a set in which the spin-up disorder decreases along the sequence Ni-Mn, Ni-Fe, Ni-Co, and Ni-Fe(*).

The resulting AMR values of the four systems are plotted in Fig. 3 as functions of the change of the number of valence electrons $\Delta Z$ due to alloying. The latter quantity is defined as $\Delta Z(x) = x$ for the Ni$_{1-x}$Co$_x$ alloy, $\Delta Z(x) = 2x$ for the Ni$_{1-x}$Fe$_x$ alloy, and $\Delta Z(x) = 3x$ for the Ni$_{1-x}$Mn$_x$ alloy. The calculated AMR increases along the sequence Ni-Mn, Ni-Fe, Ni-Co, Ni-Fe(*), in qualitative agreement with experiment for the real alloys [42]. The obtained trend indicates that the AMR ratios are directly related to the disorder strength in the majority spin channel, with the highest AMR ratios obtained for alloys with negligible spin-up scattering. This conclusion does not depend on the particular approach (SRA+SO, FRD) employed in the calculation, as can be seen from a comparison of results for $\Delta Z = 0.3$, see Table [42].
FIG. 3. (Color online) The calculated values of the anisotropic magnetoresistance of random fcc Ni-based alloys as functions of the valence charge difference $\Delta Z$.

TABLE II. The calculated and experimental values of the anomalous Hall conductivity $\sigma_{xy}$ (in kS/m) for 3$d$ transition-metal ferromagnets. The displayed values obtained in this work correspond to the FRD approach; in parentheses, results of the SRA+SO approach are shown.

|                | bcc Fe  | fcc Co  | fcc Ni  |
|----------------|---------|---------|---------|
| this work      | $-65(-55)$ | $-36(-34)$ | $241(243)$ |
| Berry curvature | $-75^a$ | $-25^b$ | $220^c$ |
| KKR method     | $-64^d$ |         | $164^d$ |
| experiment     | $-103^f$ |         | $65^f$  |

$^a$ Reference 16.  $^b$ Reference 18.  $^c$ Reference 17.  $^d$ Reference 26.  $^e$ Reference 51.  $^f$ Reference 52.

B. AHE in elemental ferromagnets

The calculated anomalous Hall conductivities $\sigma_{xy}$ of three cubic $3d$ transition metals are listed in Table II together with other theoretical and experimental results. The Co was treated here in the fcc structure, which represents the natural cubic approximation to the ground-state hexagonal close-packed structure. It should be noted that our sign convention for the $\sigma_{xy}$ differs from that adopted in the previous studies; all signs in the table are thus taken consistently with our convention.

One can see that the results of the TB-LMTO method agree reasonably well with the data based on the Berry curvature and on the fully relativistic KKR Green’s-function technique. The biggest discrepancy in Table II, related to the measured and calculated AHE of fcc Ni, might reflect effects of strong correlations on the electronic structure.

C. AHE in random fcc Ni-based alloys

The effect of alloying on the AHE has been studied for the random fcc Ni-based alloys discussed in Section III A.

In order to assess the role of chemical disorder on the AHE, the Ni$_{1-x}$Co$_x$ alloy has also been treated in a simple virtual crystal approximation (VCA) in which the alloy constituents are replaced by an effective atom with the atomic number $Z(x) = 28 - x$; the resulting system is denoted as Ni-Co(*)).

The concentration trends of the $\sigma_{xy}$, calculated in the SRA+SO approach, are summarized in Fig. II. We note that the dilute limit of the alloys, where the $\sigma_{xy}$ diverges due to the diverging incoherent contribution, has not been studied here. All studied systems exhibit a change of the sign of the AHE due to alloying. This can be understood as
FIG. 4. (Color online) The calculated values of the anomalous Hall conductivity $\sigma_{xy}$ of random fcc Ni-based alloys as functions of the valence charge difference $\Delta Z$.

a consequence of band filling and of the opposite signs of the $\sigma_{xy}$ for pure fcc Co and Ni, see Table III. A similar sign change has been observed a long time ago for Ni-rich Ni-Fe and Ni-Co alloys. This feature was related to the occurrence of the high AMR values encountered in these systems with roughly the same composition. However, our calculated data for the ferromagnetic Ni-Mn alloy witness, that the sign change of the AHE can be obtained also for a system with much smaller AMR values.

Let us discuss finally the role of disorder on the AHE from a comparison of the calculated $\sigma_{xy}$ for the Ni-Fe and Ni-Fe(*) systems as well as for the Ni-Co and Ni-Co(*) alloys. As one can see from Fig. 4, the neglect of the disorder in the majority spin channel (Ni-Fe) or in both spin channels (Ni-Co) has a strong influence on the resulting $\sigma_{xy}$. This analysis indicates that results of simplified treatments of alloying, such as, e.g., in Ref. 56, should be taken with caution. Moreover, the $\sigma^\text{VCA}_{xy}$ for the Ni-Co(*) alloy differs significantly both from the total $\sigma_{xy}$ for the corresponding Ni-Co alloy in the CPA and from its coherent part, $\sigma^\text{coh}_{xy}$. For the equiconcentration Ni-Co alloy, we get: $\sigma^\text{VCA}_{xy} = -14$ kS/m, $\sigma_{xy} = -88$ kS/m, and $\sigma^\text{coh}_{xy} = -93$ kS/m. This example proves that the coherent part of the anomalous Hall conductivity of a real concentrated alloy is not related directly to that of the corresponding effective crystal, in contrast to the case of the dilute alloys.

IV. CONCLUSIONS

We have reformulated the Kubo-Stˇreda expression for the conductivity tensor in the fully relativistic TB-LMTO-CPA theory. This extends the applicability of our previous transport formalism to all elements of the conductivity tensor, indispensable for the anomalous Hall effect. The effect of the spin-orbit interaction has been included and implemented numerically in the Dirac four-component formalism as well as in a simple perturbative manner. The former approach is inevitable for systems with heavy elements while the latter scheme is advantageous for interpretation of the results in terms of the underlying electronic structure owing to the non-relativistic (or scalar-relativistic) basis set of orbitals. First applications to galvanomagnetic phenomena in pure metals (Fe, Co, Ni) and in ferromagnetic Ni-based alloys yield results in reasonable agreement with other techniques. The performed calculations for the real and artificial alloy systems clarified some aspects of the anisotropic magnetoresistance and the anomalous Hall effect and their possible interrelation. The developed technique has already been generalized and applied to alloy structures with several sublattices and different degree of atomic ordering. Further questions, such as, e.g., the influence of complex magnetic orders on the transport properties in systems with competing ferro- and antiferromagnetic interactions, remain a task for future.
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Appendix A: Relations between the Green’s functions

The proof of Eqs. (9) and (16) is based on a general identity valid for matrices \( \hat{x}, x, \hat{y}, y, \) and \( f, \) that are coupled by

\[
\hat{x} = x(1+f)x^{-1} = (1+xf)^{-1}x, \quad x = \hat{x}(1-\hat{x})^{-1} = (1-\hat{xf})^{-1}\hat{x},
\]

\[
\hat{y} = y(1+fy)^{-1}y, \quad y = \hat{y}(1-\hat{y}f)^{-1}\hat{y},
\]

if corresponding inverses exist. This yields relations

\[
\hat{x} - \hat{y} = (1+xf)(x-y)(1+fy)^{-1},
\]

\[
(\hat{x} - \hat{y})^{-1} = (1+fy)(x-y)^{-1}(1+xf),
\]

and the general identity

\[
(\hat{x} - \hat{y})^{-1} = -(1+fx)f + (1+fx)(x-y)^{-1}(1+xf).
\]

This identity can be used for \( \hat{x} = [(\sqrt{\Delta})^+ - (z-C)(\sqrt{\Delta})^{-1}, \hat{y} = S^0(1 - \gamma S^0)^{-1}, \) and \( f = \alpha - \gamma, \) so that \( x = P^\alpha(z) \) and \( y = S^\alpha. \) Substitution into Eq. (A3) and subsequent multiplication by \( (\sqrt{\Delta})^{-1} \) from the left and by \( [(\sqrt{\Delta})^+]^{-1} \) from the right leads to the relation (6) with the matrices \( \hat{x} = (\sqrt{\Delta})^+, \hat{y} = (\sqrt{\Delta})^{-1}, \) and \( f = \alpha - \gamma, \) so that \( y = P^\alpha(z) \) and \( x = S^\alpha. \) This yields the complementary relation between the Green’s functions, Eq. (16).

Appendix B: Equivalence of expressions for the conductivity tensor

For the proof of equivalence of Eqs. (9) and (17), we drop the superscript \( \alpha \) at \( v^\alpha_\mu, g^\alpha_\pm, S^\alpha \) and \( F^\alpha, \) and abbreviate \( a = \alpha - \gamma. \) The definition (14) is thus written as

\[
F = 1 + Sa,
\]

and the two relations (13) and (16) are now written as

\[
V_\mu = (\sqrt{\Delta})^+ F^{-1}v_\mu(F^+)^{-1}\sqrt{\Delta}, \quad G_\pm = (\sqrt{\Delta})^{-1}F^+(a + g_\pm F)[(\sqrt{\Delta})^+]^{-1}.
\]

Substitution of Eq. (B2) into the original Eq. (9) yields after trivial modifications

\[
\sigma_{\mu\nu} = \sigma_0 \text{Tr}\left\{ F^{-1}v_\mu(g_+ - g_-)v_\nu (a + g_- F) - v_\mu (a + g_+ F)F^{-1}v_\nu (g_+ - g_-) + i (X_\mu F^{-1}v_\nu - X_\nu F^{-1}v_\mu) (g_+ - g_-) F \right\} = \sigma^{(1)}_{\mu\nu} + \sigma^{(2)}_{\mu\nu},
\]

where the \( \sigma^{(1)}_{\mu\nu} \) and \( \sigma^{(2)}_{\mu\nu} \) comprise, respectively, all terms bilinear and linear in \( g_\pm. \) The first contribution is obvious,

\[
\sigma^{(1)}_{\mu\nu} = \sigma_0 \text{Tr}\left\{ v_\mu (g_+ - g_-)v_\nu g_- - v_\mu g_+ v_\nu (g_+ - g_-) \right\},
\]

while the second contribution is more complicated. It has a form

\[
\sigma^{(2)}_{\mu\nu} = \sigma_0 \text{Tr}\left\{ N_{\mu\nu} (g_+ - g_-) \right\},
\]

where the \( N_{\mu\nu} \) can be written in terms of the effective velocities \( v_\mu \) (12) as

\[
N_{\mu\nu} = v_\nu a F^{-1}v_\mu - v_\mu a F^{-1}v_\nu + i F (X_\mu F^{-1}v_\nu - X_\nu F^{-1}v_\mu)
\]

\[
= i ([X_\mu, S]a + FX_\mu) F^{-1}v_\nu - i ([X_\nu, S]a + F X_\nu) F^{-1}v_\mu.
\]

Here, the last two brackets can be modified using Eq. (B1) and \([X_\mu, a] = 0,\) so that \([X_\mu, F] = [X_\mu, S]a,\) hence \([X_\mu, S]a + F X_\mu = X_\mu F\) and Eq. (B6) reduces to

\[
N_{\mu\nu} = i (X_\mu v_\nu - X_\nu v_\mu).
\]
Consequently, the second contribution in Eq. (B3) simplifies to
\[ \sigma_{\mu\nu}^{(2)} = \sigma_0 \text{Tr} \{i (X_{\nu} v_{\nu} - X_{\nu} v_{\mu}) (g_+ - g_-)\}. \] (B8)

The sum of Eq. (B4) and Eq. (B8) is now identical to the transformed expression for the \( \sigma_{\mu\nu} \).
48 S. Kokado, M. Tsunoda, K. Harigaya, and A. Sakuma, J. Phys. Soc. Japan 81, 024705 (2012).
49 H. Akai and P. H. Dederichs, Phys. Rev. B 47, 8739 (1993).
50 J. Kudrnovský, V. Drchal, and P. Bruno, Phys. Rev. B 77, 224422 (2008).
51 P. N. Dheer, Phys. Rev. 156, 637 (1967).
52 J. M. Lavine, Phys. Rev. 123, 1273 (1961).
53 H. R. Fuh and G. Y. Guo, Phys. Rev. B 84, 144427 (2011).
54 L. Berger, Physica 30, 1141 (1964).
55 E. I. Kondorskii, A. V. Cheremushkina, and N. Kurbanyazov, Fiz. Tverd. Tela 6, 539 (1964).
56 H. Zhang, S. Blügel, and Y. Mokrousov, Phys. Rev. B 84, 024401 (2011).
57 J. Kudrnovský, V. Drchal, S. Khmelevskyi, and I. Turek, Phys. Rev. B 84, 214436 (2011).