Coherent Description of the Intrinsic and Extrinsic Anomalous Hall Effect in Disordered Alloys on an *Ab Initio* Level

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A coherent description of the anomalous Hall effect is presented that is applicable to pure and disordered systems. This is achieved by an implementation of the Kubo-Středa equation using the fully relativistic Korringa-Kohn-Rostoker method in combination with the coherent potential approximation. Applications to the pure ferromagnets Fe and Ni led to results in full accordance with previous work. For the alloy systems Fe_xPd_{1-x} and Ni_xPd_{1-x} very satisfying agreement with experiment could be achieved for the anomalous Hall conductivity. To interpret these results a detailed discussion of the skew and side-jump scattering processes is given.

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During recent years the anomalous Hall effect (AHE) has received great interest. This is partly caused by its close connection to the spin Hall effect, which possesses a large potential for application in the rapidly growing field of spintronics [1]. On the other hand, many theoretical investigations are devoted to the development of a coherent description of these quite complex phenomena [2].

As was already pointed out by Karplus and Luttinger [3], the ultimate origin for the AHE in ferromagnets is the spin-orbit coupling (SOC) that—together with the spontaneous magnetization—leads to a symmetry breaking. As was demonstrated by experiment [4,5] and is obvious from the work of Karplus and Luttinger, the AHE is present even in pure systems. This so-called intrinsic AHE could later be connected to the Berry phase [6], and corresponding *ab initio* results could be obtained during recent years using an expression for the anomalous Hall conductivity (AHC) σ_{xy} in terms of the Berry curvature [7,8]. For diluted and concentrated alloys, on the other hand, the occurrence of the AHE was primarily ascribed to the spin-dependent skew or Mott [9,10] and the so-called side-jump [11] scattering mechanisms. The latter one is caused by the anomalous velocity, a first-order relativistic correction to the nonrelativistic velocity operator connected to SOC. Interestingly, scaling laws connecting the AHC σ_{xy} and the longitudinal conductivity σ_{xx} (see below) could be derived for these two extrinsic mechanisms [2]. Their treatment in connection with a description of electronic transport in terms of wave packet dynamics was discussed in detail recently by Sinitsyn [12]. When dealing with the extrinsic AHE in disordered systems, however, disorder was treated thus far only by model potentials [13] or by a damping parameter [14,15]. Crépieux and Bruno [16] performed qualitative investigations on the AHE on the basis of the Kubo-Středa equation. This equation is derived from Kubo's linear response formalism supplying a suitable basis for investigations based on a realistic description of the underlying electronic structure (see Ref. [17] and below). An alternative description of the AHE with a wider regime of applicability is achieved by using the nonequilibrium Green function formalism. Using a suitable, but still tractable, model description for the electronic structure, Onoda *et al.* [14,15] could divide the range of σ_{xx} covered typically by real materials into three regimes with different scaling laws connecting σ_{xy} and σ_{xx}.

In this Letter results for the AHC obtained using the Kubo-Středa equation are presented. Using a fully relativistic Green function formulation in combination with a reliable alloy theory, a coherent description for pure as well as diluted and concentrated alloys could be achieved that treats intrinsic and extrinsic sources of the AHE on equal footing.

The Kubo linear response formalism supplies an appropriate basis to deal with electronic transport in magnetic metallic systems. Making use of a single-particle description of the electronic structure and restricting to the case $T = 0$ K, one is led to the Kubo-Středa equation for the electrical conductivity tensor $\boldsymbol{\sigma}$ [18]. For cubic systems with the magnetization along the z direction, the AHE is described by the corresponding off-diagonal tensor element or anomalous Hall conductivity σ_{xy} given by [18,19]

$$\begin{aligned}\sigma_{xy} = & \frac{\hbar}{4\pi N\Omega}\mathrm{Tr}\langle \hat{j}_x(G^+ - G^-)\hat{j}_y G^- \\ & - \hat{j}_x G^+ \hat{j}_y (G^+ - G^-)\rangle_c \\ & + \frac{e}{4\pi i N\Omega}\mathrm{Tr}\langle (G^+ - G^-)(\hat{r}_x\hat{j}_y - \hat{r}_y\hat{j}_x)\rangle_c. \end{aligned} \quad (1)$$

Here Ω is the volume of the unit cell, N is the number of sites, while $\hat{\mathbf{r}}$ and $\hat{\mathbf{j}}$ are the position and current density operators, respectively. For the cubic systems considered here the last term is site diagonal for symmetry reasons. As furthermore all systems considered here are metallic, it has been omitted [17]. The electronic structure of the system is represented in terms of the single-particle retarded (G^+)

and advanced (G^-) Green functions at the Fermi energy E_F. Within the present work these functions have been evaluated by means of the multiple-scattering Korringa-Kohn-Rostoker formalism [20]. The chemical disorder in the investigated random substitutional alloys has been accounted for by using the coherent potential approximation [21]. This alloy theory supplies a reliable framework to perform the configurational average indicated by the brackets $\langle \cdots \rangle_c$ in Eq. (1). It includes, in particular, a clear definition for differences of configurational averages like $\langle \hat{j}_x G^+ \hat{j}_y G^- \rangle_c - \langle \hat{j}_x G^+ \rangle_c \langle \hat{j}_y G^- \rangle_c$. These so-called vertex corrections (VC) correspond to the scattering-in terms within semiclassical Boltzmann transport theory [22].

Dealing with the AHE requires us to account for the influence of spin-orbit coupling in an appropriate way. This is achieved by using the four-component Dirac formalism [23]. In combination with spin-density functional theory in its local approximation (LSDA) the corresponding Dirac Hamiltonian is given by [24]

$$\mathcal{H}_D = c\boldsymbol{\alpha} \cdot \hat{\mathbf{p}} + \beta mc^2 + V + \beta \Sigma_z B. \quad (2)$$

Here $\hat{\mathbf{p}} = -i\hbar\boldsymbol{\nabla}$ is the canonical momentum operator, $\boldsymbol{\alpha}$ and β are the standard Dirac matrices [23], while V and B represent the spin-independent and spin-dependent, respectively, effective LSDA potentials for the magnetization along z. Within the fully relativistic framework adopted here, the current density operator $\hat{\mathbf{j}}$ is given by [23]

$$\hat{\mathbf{j}} = -c|e|\boldsymbol{\alpha}. \quad (3)$$

To allow for a more detailed discussion on the origin of the AHE it is useful to introduce the alternative current density operator [25],

$$\hat{\mathbf{j}}_{\hat{\mathbf{p}}} = \frac{-|e|}{m + E/c^2}\left(\hat{\mathbf{p}} + \frac{V}{c}\boldsymbol{\alpha} + \frac{B}{c}\beta\Sigma_z(\alpha_x, \alpha_y, 0)^T\right), \quad (4)$$

that is equivalent to $\hat{\mathbf{j}}$ given by Eq. (3) and that can be derived from the anticommutator of $\hat{\mathbf{j}}$ and the Dirac Hamiltonian $\mathcal{H}_D$ given by Eq. (2).

Recently, the intrinsic AHE of the pure ferromagnets Fe, Co, and Ni [7,8,26] as well as ordered FePt and FePd [27] has been investigated theoretically on an *ab initio* level using the formulation for σ_{xy} in terms of the Berry curvature. Alternatively, the tensor element σ_{xy} can be obtained directly from the expression given in Eq. (1) that is evaluated by Fourier transformation leading to a corresponding Brillouin zone integration [22]. As the integrand shows a δ-function-like behavior for pure systems, a small imaginary part ϵ has to be added to the Fermi energy E_F and an extrapolation to zero has to be made for ϵ. For the calculations of σ_{xy} performed for bcc Fe and fcc Ni, ϵ has been varied between 10^{-3} and 10^{-6} Ry. To ensure convergence of the Brillouin zone integration, about 10^9 $\mathbf{k}$ points have been used. The resulting AHC of bcc Fe and fcc Ni is given in Table I together with experimental data as well as results of previous *ab initio* work [7,8,28]. Taking into account that SOC was treated by approximate schemes within the corresponding calculations, the agreement is quite satisfying.

TABLE I. The intrinsic AHC of bcc Fe and fcc Ni from *ab initio* theoretical as well as experimental (Exp.) investigations.

σ_{xy} $(\mathrm{m}\Omega\,\mathrm{cm})^{-1}$	bcc Fe	fcc Ni
This work	0.638	−1.635
Yao *et al.* [7]	0.753	
Wang *et al.* [8]	0.751	−2.203
Yao [28]		−2.073
Exp. [4,5]	1.032	−0.646

The expression for σ_{xy} in terms of the Berry curvature used within previous work is completely equivalent to the Kubo-Středa equation used here, as both approaches are based on Kubo's linear response formalism and adopt a single-particle description for the electronic structure for $T = 0$ K [17]. However, it should be stressed that the Berry curvature is usually formulated in terms of Bloch states implying translational symmetry this way. Calculations of σ_{xy} for disordered alloys with broken translational symmetry are therefore not possible on this basis while the Kubo-Středa equation supplies an adequate framework for such investigations. As the $\mathbf{k}$-dependent integrand connected with Eq. (1) gets smeared out in this case due to the chemical disorder, it loses its δ-function-like behavior. For that reason, broadening via a complex Fermi energy with $\epsilon > 0$ is not necessary for calculations on alloys.

Within the present work, corresponding calculations have been done for the alloy systems fcc Fe_xPd_{1-x} and fcc Ni_xPd_{1-x}. Both systems are formed by the nearly ferromagnetic transition metal Pd with an elemental $3d$ ferromagnet leading to a very low critical concentration x_{crit} for the onset of spontaneous ferromagnetic order ($x_{\mathrm{crit}}^{\mathrm{Fe}} \approx 0$ for Fe_xPd_{1-x} and $x_{\mathrm{crit}}^{\mathrm{Ni}} \approx 0.02$ for Ni_xPd_{1-x}). As can be seen in Fig. 1, calculation of the AHC via Eq. (1) leads to a very satisfying agreement with the experimental data [29], in particular, for Fe_xPd_{1-x}, that are available over a wide range of composition. In particular, the change in sign of σ_{xy} with composition observed for both alloy systems is well reproduced by the calculations. As one might speculate from the data for elemental bcc Fe and fcc Ni, in Table I the sign of σ_{xy} on the Pd-poor side of fcc Fe_xPd_{1-x} is indeed positive while it is negative for fcc Ni_xPd_{1-x}. For the Pd-rich side, the situation is reversed, clearly showing that the elemental ferromagnet is the primary source for the AHE in these two alloy systems (see below).

To get a more detailed insight into the mechanism responsible for the AHE in the investigated alloys, a decomposition of the AHC has been performed. A formal basis for this is provided by the representation of the Kubo-Středa equation in terms of Feynman diagrams [16]. From this it can be seen that the skew and side-jump mechanisms

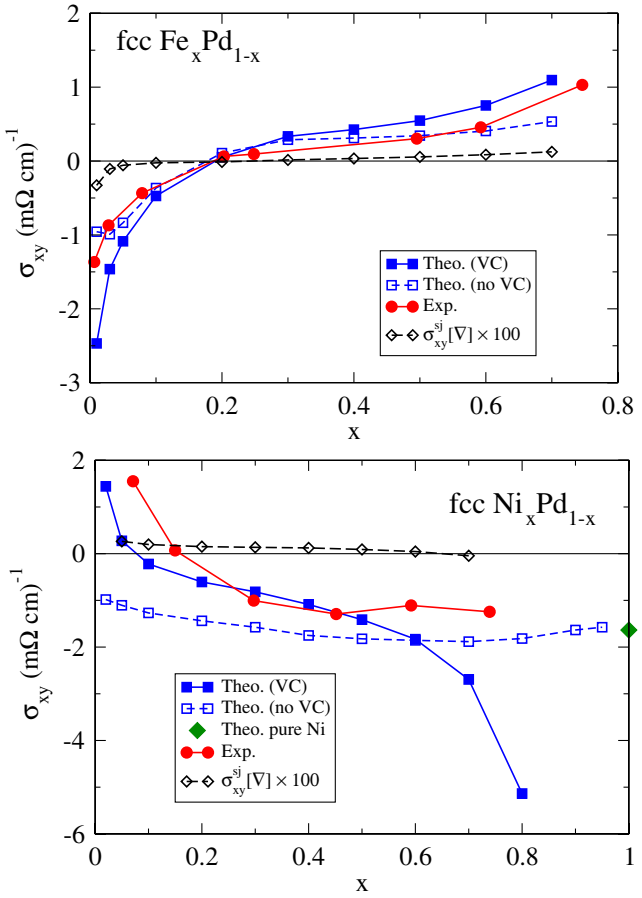

FIG. 1 (color online). The AHC of fcc Fe_xPd_{1-x} and fcc Ni_xPd_{1-x}. The total AHC σ_{xy} (full squares) has been calculated including the vertex corrections ($\sigma_{xy} \equiv \sigma_{xy}^{\mathrm{VC}}$), while the intrinsic AHC $\sigma_{xy}^{\mathrm{intr}}$ (open squares) has been obtained by omitting them ($\sigma_{xy}^{\mathrm{intr}} \equiv \sigma_{xy}^{\mathrm{no\ VC}}$). In addition, experimental data [29] for σ_{xy} (full circles) determined at $T = 4.2$ K are shown. Further, an estimation for the side-jump contribution $\sigma_{xy}^{\mathrm{sj}}$ to the extrinsic AHC of fcc Fe_xPd_{1-x} and fcc Ni_xPd_{1-x} calculated as the difference $\sigma_{xy}^{\mathrm{extr}} - \sigma_{xy}^{\mathrm{extr}}[\nabla]$ is shown (open diamonds; see text).

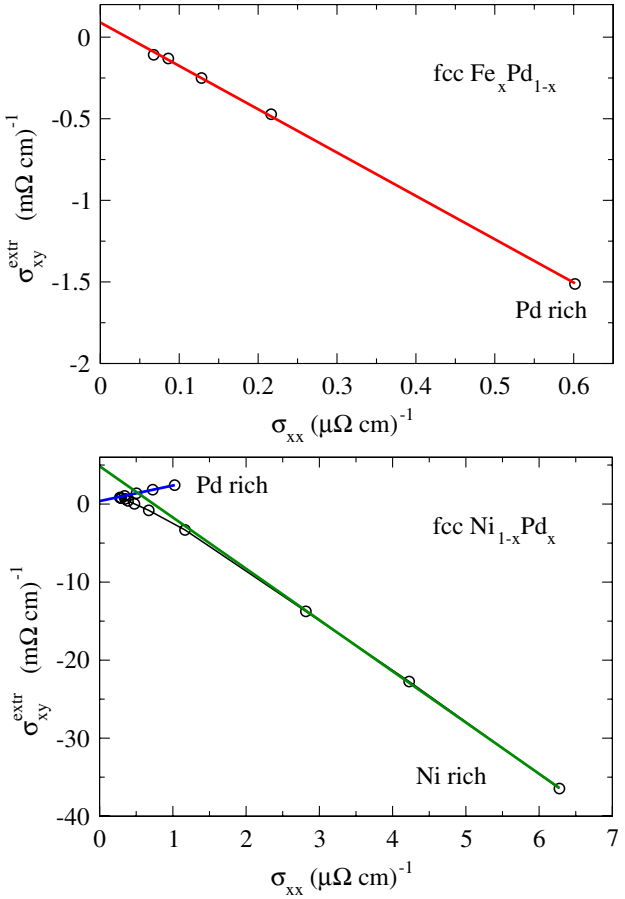

FIG. 2 (color online). The extrinsic AHC $\sigma_{xy}^{\mathrm{extr}}$ versus σ_{xx} for fcc Fe_xPd_{1-x} and fcc Ni_xPd_{1-x}. The straight lines represent extrapolations of the data for $x_{\mathrm{Pd}} \geq 0.9$ (Pd rich) and $x_{\mathrm{Ni}} \geq 0.9$ (Ni rich) to $\sigma_{xx} = 0$.

are exclusively connected to diagrams involving the vertex corrections. The remaining diagrams are standing for products of the type $\langle \hat{j}_x G^+ \rangle_c \langle \hat{j}_y G^- \rangle_c$ that correspond to the intrinsic AHE and correction terms due to chemical disorder. It seems therefore sensible to extend the definition of the intrinsic AHC $\sigma_{xy}^{\mathrm{intr}}$ to the case of diluted and concentrated alloys by combining all contributions not connected to the vertex corrections. This obviously allows us to calculate the total AHC (σ_{xy}) and the intrinsic one ($\sigma_{xy}^{\mathrm{intr}}$) by evaluating the Kubo-Středa equation [Eq. (1)] with and without, respectively, including the VC, i.e., identifying $\sigma_{xy} \equiv \sigma_{xy}^{\mathrm{VC}}$ and $\sigma_{xy}^{\mathrm{intr}} \equiv \sigma_{xy}^{\mathrm{no\ VC}}$, respectively. As seen in Fig. 1, $\sigma_{xy}^{\mathrm{intr}}$ gives a major contribution to the total AHC σ_{xy} of fcc Fe_xPd_{1-x} and shows, in particular, also a change in sign with varying concentration. For fcc Ni_xPd_{1-x}, on the other hand, $\sigma_{xy}^{\mathrm{intr}}$ varies weakly with composition and extrapolates rather well to the intrinsic AHC of pure Ni (see Table I). For both alloy systems $\sigma_{xy}^{\mathrm{intr}} \approx 1\ (\mathrm{m\Omega\,cm})^{-1}$ when x_{Pd} approaches 1, indicating that the intrinsic AHC is primarily determined by the properties of the Pd host in the dilute regime. These findings obviously justify the extension of the definition for $\sigma_{xy}^{\mathrm{intr}}$ to represent all contributions not connected to the vertex corrections.

The longitudinal conductivity σ_{xx} of fcc Fe_xPd_{1-x} and fcc Ni_xPd_{1-x} lies nearly exclusively in the so-called superclean regime with $\sigma_{xx} \gtrsim (\mu\Omega\,\mathrm{cm})^{-1}$ [14,15]. For this regime the skew-scattering mechanism should dominate σ_{xy} obeying the relation $\sigma_{xy} = S\sigma_{xx}$, with S being the so-called skewness factor [14,15]. Accounting for all three mechanisms one is therefore led to the decomposition [2],

$$\sigma_{xy} = \sigma_{xy}^{\mathrm{intr}} + S\sigma_{xx} + \sigma_{xy}^{\mathrm{sj}} = \sigma_{xy}^{\mathrm{intr}} + \sigma_{xy}^{\mathrm{extr}}, \qquad (5)$$

that may be seen as a definition for the side-jump contribution $\sigma_{xy}^{\mathrm{sj}}$ [2]. In fact, a plot of σ_{xy} versus σ_{xx} with the concentration as an implicit parameter was used in the past to decompose the experimental AHC of alloy systems accordingly [30–32].

In Fig. 2 the extrinsic AHC of fcc Fe_xPd_{1-x} and fcc Ni_xPd_{1-x} defined as $\sigma_{xy}^{\mathrm{extr}} = \sigma_{xy} - \sigma_{xy}^{\mathrm{intr}}$ is plotted versus the longitudinal conductivity σ_{xx}. Obviously, the relation

suggested by Eq. (5) is well fulfilled on the Pd-rich side of both systems as well as on the Ni-rich side of Ni_xPd_{1-x}. Extrapolating for these regimes to $\sigma_{xx} = 0$ allows us to deduce the corresponding skewness parameters and side-jump term σ_{xy}^{sj} [Fe_xPd_{1-x} for $x_{Pd} \geq 0.9$, $S = -2.7 \times 10^{-3}$ and $\sigma_{xy}^{sj} \approx 0.1\ (m\Omega\,cm)^{-1}$; Ni_xPd_{1-x} for $x_{Pd} \geq 0.9$, $S = 2.0 \times 10^{-3}$ and $\sigma_{xy}^{sj} \approx 0.4\ (m\Omega\,cm)^{-1}$; for $x_{Ni} \geq 0.9$, $S = -6.6 \times 10^{-3}$ and $\sigma_{xy}^{sj} \approx 4.8\ (m\Omega\,cm)^{-1}$]. These results show clearly that the skew-scattering mechanisms by far dominate σ_{xy}^{extr} in the dilute regimes. For the two alloy systems the corresponding skewness factor S is found comparable in magnitude but different in sign on the Pd-rich side (see above). This once more demonstrates that the skew-scattering mechanism has to be associated primarily with the solute component Fe or Ni, respectively.

As emphasized above, Eq. (5) can be seen as a definition for various extrinsic contributions to σ_{xy}^{extr} according to their scaling behavior. An alternative way to define the side-jump term σ_{xy}^{sj} is to make use of its connection with the anomalous velocity, that is a correction to the non-relativistic current density operator $\hat{\mathbf{j}}_{nr} = -\frac{|e|}{m}\frac{\hbar}{i}\boldsymbol{\nabla}$. Within the relativistic approach used here, an estimate for σ_{xy}^{sj} can be made using the alternative current density operator $\hat{\mathbf{j}}_{\hat{\mathbf{p}}}$ with the potential terms V and B suppressed [see Eq. (4)]. The corresponding extrinsic AHC $\sigma_{xy}^{extr}[\nabla] = \sigma_{xy}[\nabla] - \sigma_{xy}^{intr}[\nabla]$ allows us to write $\sigma_{xy}^{sj} \approx \sigma_{xy}^{sj}[\nabla] = \sigma_{xy}^{extr} - \sigma_{xy}^{extr}[\nabla]$. The results for σ_{xy}^{sj} obtained this way for fcc Fe_xPd_{1-x} and fcc Ni_xPd_{1-x} are also shown in Fig. 1. As one notes, there is obviously a non-negligible concentration dependency for both alloy systems, in particular, on the Pd-rich side. In both cases, however, the numerical results are much smaller than for the intrinsic as well as the skew-scattering contributions. While this once more supports the conclusion that the AHE of the investigated alloy systems is dominated by the latter mechanisms, it also shows that the quantitative results for the side-jump term σ_{xy}^{sj} may depend strongly on the definition used.

In summary, a coherent description of the AHE for pure metals and diluted as well as concentrated alloys on an *ab initio* level was presented based on a fully relativistic implementation of the Kubo-Středa equation using the multiple-scattering or Korringa-Kohn-Rostoker formalism in combination with the coherent potential approximation alloy theory. The intrinsic AHC obtained this way for bcc Fe and fcc Ni was found in satisfying agreement with previous *ab initio* work using an equivalent expression for σ_{xy} in terms of the Berry curvature. Corresponding calculations for the alloy systems fcc Fe_xPd_{1-x} and fcc Ni_xPd_{1-x} reproduced the available experimental data very well. Identifying the contributions to σ_{xy} that are not connected to the vertex corrections with the intrinsic AHE of an alloy allowed us to decompose the remaining extrinsic AHE. Plotting σ_{xy} versus σ_{xx} it was found that the skew-scattering term by far dominates the side-jump contribution in the dilute alloy regime. This conclusion could be supported by model calculations that supplied an estimate for the contribution to σ_{xy} due to the anomalous velocity.

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