Coherent description of the intrinsic and extrinsic anomalous Hall effect in disordered alloys on an \textit{ab initio} level

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A coherent description of the anomalous Hall effect (AHE) is presented that is applicable to pure as well as disordered alloy systems by treating all sources of the AHE on equal footing. This is achieved by an implementation of the Kubo-Středa equation using the fully relativistic Korringa-Kohn-Rostoker (KKR) Green’s function method in combination with the Coherent Potential Approximation (CPA) alloy theory. Applications to the pure elemental ferromagnets bcc-Fe and fcc-Ni led to results in full accordance with previous work. For the alloy systems fcc-Fe\textsubscript{x}Pd\textsubscript{1-x} and fcc-Ni\textsubscript{y}Pd\textsubscript{1-y} very satisfying agreement with experiment could be achieved for the anomalous Hall conductivity (AHC) over the whole range of concentration. To interpret these results an extension of the definition for the intrinsic AHC is suggested. Plotting the corresponding extrinsic AHC versus the longitudinal conductivity a linear relation is found in the dilute regimes, that allows a detailed discussion of the role of the skew and side-jump scattering processes.

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During the last years the anomalous Hall effect (AHE) has received great interest. This is partly caused by its close connection to the spin Hall effect (SHE), that 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 the ultimate origin for the AHE in ferromagnets is 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 \textit{ab initio} results could be obtained during the last years using an expression for the anomalous Hall conductivity (AHC) \(\sigma_{xy}\) 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 non-relativistic velocity operator connected to SOC. Interestingly, scaling laws connecting the AHC \(\sigma_{xy}\) and the longitudinal conductivity \(\sigma_{xx}\) (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 so 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 non-equilibrium Green’s function formalism. Using a suitable, but still tractable, model description for the electronic structure Onoda et al. [14, 15] could divide the range of \(\sigma_{xx}\) covered typically by real materials into three regimes with different scaling laws connecting \(\sigma_{xy}\) and \(\sigma_{xx}\).

In this communication results for the AHC obtained using the Kubo-Středa equation are presented. Using a fully relativistic Green’s 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 \(\sigma\) [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 \(\sigma_{xy}\) given by [18, 19]

\[
\sigma_{xy} = \frac{\hbar}{4\pi N\Omega} \text{Trace} \left\langle \hat{j}_x (G^+ - G^-) \hat{j}_y G^- \right\rangle \\
- \frac{e}{4\pi \Omega} \text{Trace} \left\langle \left( G^+ - G^- \right) (\hat{r}_x \hat{j}_y - \hat{r}_y \hat{j}_x) \right\rangle \\
+ \frac{e}{4\pi \Omega} \text{Trace} \left\langle \left( G^+ - G^- \right) (\hat{r}_x \hat{j}_y - \hat{r}_y \hat{j}_x) \right\rangle \\ (1)
\]
Here $\Omega$ is the volume of the unit cell, $N$ is the number of sites, while $\mathbf{r}$ and $\mathbf{j}$ 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 system considered here are metallic it has been omitted. The electronic structure of the system is represented in terms of the single-particle retarded $(G^+)$ and advanced $(G^-)$ Green’s 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 (KKR) formalism. The chemical disorder in the investigated random substitutional alloys has been accounted for by using the Coherent Potential Approximation (CPA). This alloy theory supplies a reliable framework to perform the configurational average indicated by the brackets $\langle...\rangle_c$ in Eq. (1). It includes, in particular, a clear definition for differences of configurational averages like $\langle j_x G^+ j_y G^- \rangle_c - \langle j_x G^+ j_y G^- \rangle_c$. These so-called vertex-corrected correspond to the scattering in terms within semi-classical Boltzmann transport theory.

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

$$\mathcal{H}_D = \alpha \cdot \mathbf{p} + \beta m c^2 + V + \beta \Sigma_z B .$$

Here $\mathbf{p} = -ih \nabla$ is the canonical momentum operator, $\alpha$ and $\beta$ are the standard Dirac matrices, 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 $\mathbf{j}$ is given by:

$$\mathbf{j} = -e |e| \alpha .$$

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

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

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

Recently, the intrinsic AHE of the pure ferromagnets Fe, Co and Ni as well as ordered FePt and FePd has been investigated as well as ordered FePt and FePd as $\alpha$ and $\beta$-functions in terms of the Berry curvature. Alternatively, the tensor element $\sigma_{xy}$ can be obtained directly from the expression given in Eq. (1) that is evaluated by Fourier transformation leading to a corresponding Brillouin zone integration. As the integrand shows a $\delta$-function like behavior for pure systems a small imaginary part $\epsilon$ has to be added to the Fermi energy $E_F$ and an extrapolation to zero has to be made for $\epsilon$. For the calculations of $\sigma_{xy}$ performed for bcc-Fe and fcc-Ni $\epsilon$ has been varied between $10^{-3}$ and $10^{-6}$ Ry. To ensure convergence of the Brillouin zone integration about $10^8$ 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.

| TABLE I: The intrinsic AHC of bcc-Fe and fcc-Ni from ab initio theoretical as well as experimental (Exp.) investigations. |
|------------|-------------|-------------|
| $\sigma_{xy}$ (mT cm) | bcc Fe | fcc Ni |
| present work | 0.638 | -1.635 |
| Yao et al. | 0.753 |
| Wang et al. | 0.751 | -2.203 |
| Yao | -2.073 |
| Exp. | 0.638 | -1.635 |

Exp. [4, 5] 1.032 -0.646
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-Streda equation in terms of Feynman diagrams. From this it can be seen that the skew and side-jump mechanisms are exclusively connected to diagrams involving the vertex corrections. The remaining diagrams are standing for products of the type $\langle j_x G^+(i) j_y G^-(i) \rangle$, 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 $\sigma_{xy}^{intr}$ to the case of diluted and concentrated alloys by combining all contributions not connected to the vertex corrections. This obviously allows to calculate the total AHC ($\sigma_{xy}$) and the intrinsic one ($\sigma_{xy}^{int}$) by evaluating the Kubo-Streda equation (Eq. (1)) with and without, resp., including the vertex corrections (VC); i.e. identifying $\sigma_{xy}$ $\equiv$ $\sigma_{xy}^{VC}$ and $\sigma_{xy}^{intr} = \sigma_{xy}^{intr no VC}$, respectively. As seen in Fig. 1 $\sigma_{xy}^{intr}$ gives a major contribution to the total AHC $\sigma_{xy}$ of fcc-Fe$_x$Pd$_{1-x}$ and shows in particular also a change in sign with varying concentration. For fcc-Ni$_x$Pd$_{1-x}$, on the other hand, $\sigma_{xy}^{intr}$ varies weakly with composition and extrapolates rather well to the intrinsic AHC of pure Ni (see Table I). For both alloy systems $\sigma_{xy}^{intr}$ $\approx$ 1 (mΩcm)$^{-1}$ 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 $\sigma_{xy}^{intr}$ to represent all contributions not connected to the vertex corrections.

The longitudinal conductivity $\sigma_{xx}$ of fcc-Fe$_x$Pd$_{1-x}$ and fcc-Ni$_x$Pd$_{1-x}$ lies nearly exclusively in the so-called super-clean regime with $\sigma_{xx} \gg (\mu\Omega cm)^{-1}$. For this regime the skew scattering mechanism should dominate $\sigma_{xy}$ obeying the relation $\sigma_{xy} = S \sigma_{xx}$, with $S$ being the so-called skewness factor. Accounting for all three mechanisms one is therefore led to the decomposition:

$$\sigma_{xy} = \sigma_{xy}^{intr} + S \sigma_{xx} + \sigma_{xy}^{sj} = \sigma_{xy}^{intr} + \sigma_{xy}^{sj} \cdot |\nabla|$$

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

In Fig. 2 the extrinsic AHC of fcc-Fe$_x$Pd$_{1-x}$ and fcc-Ni$_x$Pd$_{1-x}$ defined as $\sigma_{xy}^{extr} = \sigma_{xy} - \sigma_{xy}^{intr}$ is plotted versus the longitudinal conductivity $\sigma_{xx}$. 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$_x$Pd$_{1-x}$. Extrapolating for these regimes to $\sigma_{xx} = 0$ allows to deduce the corresponding skewness parameters and side-jump term $\sigma_{xy}^{sj} = (\text{Fe}_x \text{Pd}_{1-x})$ for $x_{Pd} \geq 0.9$: $S = -2.7 \cdot 10^{-3}$ and $\sigma_{xy}^{sj} \approx 0.1$ (mΩcm)$^{-1}$; Ni$_x$Pd$_{1-x}$ for $x_{Pd} \geq 0.9$: $S = 2.0 \cdot 10^{-3}$ and $\sigma_{xy}^{sj} \approx 0.4$ (mΩcm)$^{-1}$ and for $x_{Ni} \geq 0.9$: $S = -6.6 \cdot 10^{-3}$ and $\sigma_{xy}^{sj} \approx 4.8$ (mΩcm)$^{-1}$. These results show clearly that the skew scattering mechanisms by far dominates $\sigma_{xy}^{extr}$ 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 $\sigma_{xy}^{extr}$ according to their scaling behavior. An alternative way to define the side-jump term $\sigma_{xy}^{sj}$ is to make use of its connection with the anomalous velocity, that is a correction to the non-relativistic current density operator $J_{int} = -\frac{\sigma}{\mu} \cdot \nabla$. Within the relativistic approach used here, an estimate for $\sigma_{xy}^{sj}$ can be made using the alternative current density operator $J_B$ with the potential terms $V$ and $B$ suppressed (see Eq. 4). The corresponding ex-
trinsic AHE \( \sigma_{xy}^{\text{extr}} \mid \nabla \mid = \sigma_{xy} \mid \nabla \mid - \sigma_{xy}^{\text{intr}} \mid \nabla \mid \) allows to write \( \sigma_{xy}^{\text{s}\mid \nabla \mid} \approx \sigma_{xy}^{\text{s}\mid \nabla \mid} = \sigma_{xy}^{\text{extr}} \mid \nabla \mid - \sigma_{xy}^{\text{extr}} \mid \nabla \mid \). The results for \( \sigma_{xy}^{\text{s}\mid \nabla \mid} \) obtained this way for fcc-Fe\(_x\)Pd\(_1-x\) and fcc-Ni\(_x\)Pd\(_1-x\) 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 \( \sigma_{xy}^{\text{s}\mid \nabla \mid} \) 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 \textit{ab initio} level was presented based on a fully relativistic implementation of the Kubo-Středa equation using the multiple-scattering or KKR formalism in combination with the CPA alloy theory. The intrinsic AHC obtained this way for bcc-Fe and fcc-Ni was found in satisfying agreement with previous \textit{ab initio} work using an equivalent expression for \( \sigma_{xy} \) in terms of the Berry curvature. Corresponding calculations for the alloy systems fcc-Fe\(_x\)Pd\(_1-x\) and fcc-Ni\(_x\)Pd\(_1-x\) reproduced the available experimental data very well. Identifying the contributions to \( \sigma_{xy} \) that are not connected to the vertex corrections with the intrinsic AHE of an alloy allowed to decompose the remaining extrinsic AHE. Plotting \( \sigma_{xy} \) versus \( \sigma_{xx} \) 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 \( \sigma_{xy} \) due to the anomalous velocity.

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