Influence of complex disorder on skew-scattering Hall effects in \(L_10\)-ordered FePt alloy

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We show by first-principles calculations that the skew-scattering anomalous Hall and spin Hall angles of \(L_10\)-ordered FePt drastically depend on different types of disorder. A different sign of the anomalous Hall angle is obtained when slightly deviating from the stoichiometric ratio towards the Fe-rich side as compared to the Pt-rich side. For stoichiometric samples, short-range ordering of defects has a profound effect on the Hall angles and can change them by a factor of 2 as compared to the case of uncorrelated disorder. This might explain the vast range of anomalous Hall angles measured in experiments, which undergo different preparation procedures and thus might differ in their crystallographic quality.

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Future information technology will heavily rely on spin-orbit effects, which enable the all-electric control of magnetization and spin degrees of freedom. Spin currents already play a vital role in state-of-the-art technology, for example, in spin-transfer torque magnetic access memories (STT-MRAM), and will become ever more important in emergent magnetic technologies. Bright prospects for relativistic spin currents are associated in particular with their key importance for the phenomena of spin-orbit torque [1], current-induced domain wall [2] and skyrmion motion [3], and ultrafast magnetic applications [4].

At the heart of spin-orbit transport effects lie the anomalous and spin Hall effects (AHE and SHE) [5], because they allow for an efficient conversion from a longitudinal charge current (that is, aligned parallel to an applied electric field) into a transverse charge and spin current, respectively. For these microscopically spin-orbit coupling (SOC) originated phenomena there is already a relatively established knowledge of their underlying mechanisms, which are partly rooted in topological properties, thus fundamentally relating the AHE and SHE to the physics of, e.g., skyrmions [6], orbital magnetism [7], and topological metals [8]. Conventionally, three relatively distinct contributions to the AHE and SHE are discussed: the so-called intrinsic Berry phase contribution stemming from the electronic structure of a pristine crystal, and two contributions which arise due to disorder, namely, the side jump and skew scattering [9]. Among the three, it is the skew scattering which dominates the Hall effects in the limit of small disorder. The reason is the linear scaling of the skew-scattering driven transverse conductivity \(\sigma_{xy}\) with the diagonal conductivity \(\sigma_{xx}\) for vanishing scattering. The corresponding scaling constants, the so-called anomalous or spin Hall angles (AHA or SHA), are respectively defined as \(\alpha^{AHE} = \sigma^c_{xy}/\sigma^c_{xx}\) and \(\alpha^{SHE} = \sigma^c_{xy}/\sigma^s_{xx}\) , where superscripts “c” and “s” refer to the charge- and spin-conductivity tensors, respectively.

From a materials perspective, while elemental ferromagnets Fe, Co, and Ni give rise to relatively large AHE, they have the disadvantage of weak SOC with corresponding small values of magnetic anisotropy energy [10,11]. Heavy transition metals with strong SOC can be successfully doped with magnetic impurities and give rise to large AHE, however, such systems suffer from low Curie temperatures [12]. The \(L_10\)-ordered FePt alloy is by now a classical example of a complex ferromagnet which combines strong SOC and large AHE with strong ferromagnetic ordering. Its crystal structure is depicted in Fig. 1(a). Remarkably, the strong SOC in combination with uniaxial symmetry of the tetragonal crystal structure leads to a gigantic out-of-plane magnetocrystalline anisotropy energy that is promising for perpendicular magnetic recording [13,14], strong anisotropy of the AHE, and large anisotropic magnetoresistance [15].

Much effort has been undertaken to analyze the AHE in this material from both theoretical and experimental sides [16–19]. Seemann et al. [20] deduced from a combined experimental and theoretical study that the intrinsic and side-jump contributions to the anomalous Hall conductivity (AHC) are dominant in their samples at elevated temperatures. By extrapolation to zero temperature they were also able to deduce a large magnitude of the skew-scattering Hall angle of 1.10%. However, experiments by He et al. [21] and Chen et al. [22,23] report an order of magnitude lower skew-scattering anomalous Hall angles of 0.05%. Recent ab initio calculations, which investigate the effect of long-range order by means of the coherent-potential approximation (CPA), find even smaller skew-scattering Hall angles of 0.02% [15]. In contrast, very large Hall angles of up to 1.5% have been reported in completely disordered FePt alloys [23]. This puzzling situation, as summarized in Table I, is the starting point of our investigation.

In this Rapid Communication, we show by density functional theory (DFT) calculations that the skew-scattering contribution to the AHE and SHE in \(L_10\) FePt drastically depends on the type of disorder present in real materials. As we show below, simple antisite defects of Fe and Pt lead to a different sign and magnitude of the AHA, comparable to the large values observed in experiment [20]. In contrast, our values for the AHA in stoichiometric samples with an uncorrelated distribution of defects are considerably lower in magnitude, in line with previous CPA results [15]. We additionally show that short-range ordering of defects (that is, a tendency to locate a Pt antisite defect next to an Fe antisite defect) has a profound effect on the AHE and SHE, and can change the corresponding Hall angles by a factor of 2 as compared to the case of uncorrelated disorder.

Our investigations are based on the local spin-density approximation (LSDA) to DFT employing the relativistic full-potential Korringa-Kohn-Rostoker Green’s function method
As described by FePt alloy. Experimental samples differ in their long-range order, 0.05% S

The green ellipse breaks the tetragonal symmetry of the lattice. In (b) an Fe atom by Pt or (c) vice versa. (d), (e) Swapping two nearest-ordered FePt. Large blue and small red spheres represent Pt and Fe.

\( \text{TABLE I. Summary of literature values for the absolute value of the skew-scattering anomalous Hall angle in the L}_{10} \text{-ordered FePt alloy. Experimental samples differ in their long-range order, as described by } S \text{, and film thickness } t \text{ (in nm).} \)

<table>
<thead>
<tr>
<th>( \alpha^{\text{AHE}} )</th>
<th>Sample</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.10%</td>
<td>( S \approx 0.8, \ t = 30 )</td>
<td>Seemann et al. [20]</td>
</tr>
<tr>
<td>0.05%</td>
<td>( S = 0.74, \ t = 10-20 )</td>
<td>Chen et al. [22]</td>
</tr>
<tr>
<td>0.05%</td>
<td>( S = 0.71, \ t = 20 )</td>
<td>He et al. [21]</td>
</tr>
<tr>
<td>0.8%–1.5%</td>
<td>( S = 0, \ t = 30–100 )</td>
<td>Chen et al. [23]</td>
</tr>
<tr>
<td>0.02%</td>
<td>TB-LMTO-CPA</td>
<td>Kudrnovsky et al. [15]</td>
</tr>
</tbody>
</table>

\( \alpha^{\text{AHE}} = \alpha^{\text{Fe}} + \alpha^{\text{Pt}} \).

\[ P_{kk}^{\text{avg}} = P_{kk}^{\text{Fe}} + P_{kk}^{\text{Pt}}. \]
where the concentrations for the two types of impurities are determined by the long-range order parameter $S$ according to $c_{Fe} = c_{Pt} = (1 - S)/2$. For this uncorrelated mixture of defects, the values of the diagonal conductivities are in between the ones of the previously discussed nonstoichiometric crystals, being much closer to the Pt-impurity case. Both the transverse charge and spin conductivities are significantly reduced as compared to the previous cases (see Table II). A heuristic argument for this reduction is a partial compensation of the opposite in sign skew scattering off Fe and Pt impurities. An anomalous Hall angle of approximately $-0.11\%$ deduced from very recent tight-binding linearized muffin-tin orbital method combined with the coherent-potential approximation (TB-LMTO-CPA) calculations \cite{33} is in very good agreement to our value of $-0.09\%$ (see Table II).

Let us now see whether we can arrive at this result by simpler means. First, it is seemingly plausible to think of electrons of opposite spin (up, $\uparrow$, and down, $\downarrow$) as distinct entities which do not interact with each other and which separately contribute to the charge and spin conductivity: $\sigma \equiv \sigma^\uparrow + \sigma^\downarrow$ and $\sigma^\downarrow = \sigma^\uparrow - \sigma^\downarrow$ (in fact, we used this picture above in our interpretation of diagonal conductivities in terms of the LDOS). The Matthiessen rule states that the resistivities can be simply added if the two scattering sources are independent of each other, i.e.,

$$\frac{1}{\sigma^\downarrow} = \frac{1}{\sigma^\uparrow} + \frac{1}{\sigma^\downarrow}\left[(\sigma^\uparrow)^{-1} + (\sigma^\downarrow)^{-1}\right], \quad (3)$$

and similarly for $\sigma^\downarrow$. This procedure yields values for the elements of the averaged charge-conductivity tensor that are about 20% too high in magnitude compared to the full calculation. In contrast, the transverse spin conductivity comes out by a factor of 8 too small. This discrepancy originates in the fact that, due to the strong spin-orbit coupling in FePt, the electronic wave functions are strongly spin mixed and the fact that, due to the strong spin-orbit coupling in FePt, the two-current ansatz evidently fails in this case.

An alternative approach would be to regard the charge and spin currents independently, and to perform the averaging in analogy to Eq. (3) directly on the level of the charge- and spin-conductivity tensors. This again gives a reasonable estimate for the elements of the charge-conductivity tensor, but as far as the transverse spin conductivity is concerned, not even its sign can be reproduced correctly ($\sigma^\downarrow = 400 \, S/cm$ as compared to $-930 \, S/cm$ for the full calculation). In conclusion, the Matthiessen rule approximations work quite well for charge transport, but greatly fail for spin-transport properties of the $L_1_0$ FePt alloy.

Equation (2) entails the approximations that (i) the wave-function phase is lost due to random positions of the impurities and (ii) the concentration is small enough that multiple-scattering effects between impurities can be neglected. However, in the case of correlated impurity positions these approximations are no longer valid. In order to estimate the impact of such effects on transport properties, we investigate the extremal case of two antisite defects being nearest neighbors, i.e., when nearest-neighbor Fe and Pt atoms swap their positions and form a dimer [Figs. 1(d) and 1(e)]. This class of defects simulates an ultimate case of short-range ordering (SRO) of defects. Generally, there are eight possible orientations of the dimer bond, and in a realistic situation they would appear, with equal probability, randomly distributed over the crystal. We emphasize that we perform a full calculation for each dimer orientation, i.e., we swap the two atoms in the impurity cluster, calculate the self-consistent impurity potentials, and finally obtain the

![FIG. 2. Spin-resolved local density of states of an Fe impurity and the Fe atom in a dimer as compared to the substituted host Pt atom (upper panel) and vice versa (lower panel). Arrows pointing downwards and upwards correspond to majority and minority spins, respectively \cite{29}.](image)
transition rates directly from Eq. (1). Next, we average over the dimer orientations on the level of the transition rates in analogy to Eq. (2), which neglects dimer-dimer interference effects. We choose the concentrations such that in total 1% of the crystal sites are defects.

Comparing first the local density of states (LDOS) of an Fe atom in the dimer to a simple antisite Fe impurity (see the upper panel of Fig. 2), we remark that the two LDOS are practically identical. The same is true for the Pt atom in the dimer compared to a Pt impurity (see lower panel), with minor modifications of the occupied states around 6 eV below the Fermi level. This similarity could suggest very similar transport properties between the uncorrelated mixture and the SRO case. Indeed, the full calculation reveals that SRO increases the diagonal conductivity by only 10% as compared to uncorrelated disorder (see Table II). This is qualitatively in line with Ref. [34], where a moderate decrease in the longitudinal resistivity upon inclusion of SRO in CuZn alloys was predicted from calculations based on the nonlocal coherent-potential approximation.

On the contrary, SRO has a profound impact on the transverse transport properties (see Table II). Interestingly, $\sigma_{xy}$ is increased by a factor of roughly 2, whereas $\sigma_{xy}^{c}$ is reduced by a factor of 1.6, with similar trends for the anomalous and spin Hall angles. Our results show that transverse transport properties depend on the fine details of scattering at the Fermi surface, and full ab initio calculations are required to describe complex disorder reliably.

To summarize, we have shown that the skew-scattering anomaly and spin Hall angles of $L1_0$-ordered FePt drastically depend on the disorder type. Remarkably, the sign of the AHE is changed when the composition of the alloy slightly deviates from the stoichiometric ratio towards the Fe-rich side as compared to the Pt-rich side. Short-range ordering of defects has a profound effect on the Hall angles and can change them by a factor of 2 as compared to the case of dilute uncorrelated disorder. This might explain the vast range of anomalous Hall angles measured in experiments on different samples of this alloy, which undergo different preparation procedures and differ in their crystallographic quality. The detailed microscopic understanding of skew scattering in such alloys paves the way towards an educated ability of engineering the desired Hall transport properties of transition metals.

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[29] Note that the sign of $\sigma_{xy}$ depends on the direction of the magnetization direction. Here, the magnetization direction (spin moment) is chosen along the $+z$ ($-z$) direction of the coordinate system.


