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High spin polarization of the anomalous Hall current in Co-based Heusler compounds

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Abstract. Based on first principles density functional calculations of the intrinsic anomalous and spin Hall conductivities, we predict that the charge Hall current in Co-based full Heusler compounds Co\textsubscript{2}XZ (X = Cr and Mn; Z = Al, Si, Ga, Ge, In and Sn), except Co\textsubscript{2}CrGa, would be almost fully spin polarized, even though Co\textsubscript{2}MnAl, Co\textsubscript{2}MnGa, Co\textsubscript{2}MnIn and Co\textsubscript{2}MnSn do not have a half-metallic band structure. Furthermore, the ratio of the associated spin current to the charge Hall current is slightly larger than 1.0. This suggests that these Co-based Heusler compounds, especially Co\textsubscript{2}MnAl, Co\textsubscript{2}MnGa and Co\textsubscript{2}MnIn which are found to have large anomalous and spin Hall conductivities, might be called anomalous Hall half-metals and could have valuable applications in spintronics such as spin valves as well as magnetoresistive and spin-torque-driven nanodevices. These interesting findings are discussed in terms of the calculated electronic band structures, magnetic moments and also anomalous and spin Hall conductivities as a function of the Fermi level.

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1. Introduction

Spin electronics (or spintronics) [1] has recently become an emergent field because of the exciting promise of such spin-transport devices as magnetic field sensors for reading magnetically stored information based on giant magnetoresistance (GMR) [2, 3], spin-valves-based tunneling magnetoresistance (TMR) [4, 5] and spin-torque switching-based magnetic nanodevices such as magnetic random access memories [6, 7]. The materials, which can provide a highly spin-polarized current, are a key ingredient of spintronics. In this context, half-metallic ferromagnets [8], which are characterized by the coexistence of metallic behavior for one spin channel and insulating behavior for the other, are particularly attractive. Their electronic density of states at the Fermi level is completely spin polarized, and thus they could, in principle, offer a fully spin-polarized current. Therefore, since the first prediction of half-metallicity for the half Heusler compound NiMnSb in 1983 [8], intensive research on half-metallic materials has been carried out [9–15]. Indeed, a large number of materials have been predicted to be half-metallic and the half-metallicity of some of these such as CrO$_2$ with a Curie temperature $T_C = 392$ K [16] has also been verified experimentally.

Most Co-based full Heusler compounds in the cubic L2$_1$ structure are ferromagnetic with a high Curie temperature and a significant magnetic moment [10]. In particular, Co$_2$MnSi has a Curie temperature as high as 985 K and a large magnetic moment of 4.96 $\mu_B$. Furthermore, many of them were predicted to be half-metallic [11, 14, 15] and hence are of particular interest for spintronics. Therefore, the electronic band structure and magnetic properties of the full Heusler compounds have been intensively investigated both theoretically and experimentally in recent years [10, 11, 14, 15]. For example, the total magnetic moments of these materials were found to follow the Slater–Pauling-type behavior and the mechanism was explained in terms of the calculated electronic structures [11]. The Curie temperatures of Co-based Heusler compounds were also determined ab initio and the trends were related to the electronic structures [14].

In this paper, we study the anomalous Hall effect (AHE) and spin polarization of the Hall current in the Co-based full Heusler compounds Co$_2$XZ ($X = \text{Cr and Mn}; Z = \text{Al, Si, Ga, Ge, In and Sn}$) by ab initio calculations of intrinsic anomalous and spin Hall conductivities. The AHE, discovered in 1881 by Hall [17], is an archetypal spin-related transport phenomenon and hence has received renewed attention in recent years [18]. Indeed, many ab initio studies on the AHE in elemental ferromagnets [19–22] and intermetallic compounds [23, 24] have recently been reported. However, first principles investigations into the AHE in half-metallic
ferromagnets, which is interesting on its own account, have been very few [25]. Therefore, the principal purpose of this work is to understand the AHE in Co-based full Heusler compounds, especially those of half-metallic ones. The results may also help experimentally search for the Heusler compounds with a large AHE for applications, e.g., in magnetization sensors [26].

The intrinsic AHE is caused by the opposite anomalous velocities experienced by spin-up and spin-down electrons when they move through the relativistic energy bands in solids under the influence of the electric field [18]. In ferromagnets, where an unbalance of spin-up and spin-down electrons exists, these opposite transverse currents would give rise to the spin-polarized charge current (i.e. anomalous Hall current). In non-magnetic materials where the numbers of the spin-up and spin-down electrons are equal, the same process would result in a pure spin current, and this is known as the intrinsic spin Hall effect [27]. Interestingly, the pure spin current is dissipationless [27] and is thus important for the development of low-energy consumption nanoscale spintronic devices [28]. We note that high spin polarization ($P$) of the charge current ($I_C$) from the electrode is essential for large GMR and TMR. However, since the current-induced magnetization switching results from the transfer of spin angular momentum from the current carriers to the magnet [6], a large spin current ($I_S$) would be needed for the operation of the spin-torque switching-based nanodevices [6, 7], i.e. a large ratio of spin current to charge current [$\eta = |(2e/\hbar)I_S/I_C|$] would be crucial. For ordinary charge currents, this ratio $\eta$ varies from 0.0 (spin unpolarized current) to 1.0 (fully spin-polarized current). Interestingly, $\eta$ can be larger than 1.0 for the charge Hall currents and is $\infty$ for a pure spin current. Excitingly, spin-torque switching of ferromagnets driven by an intense pure spin current from the large spin Hall effect in tantalum has recently been reported [28]. Therefore, because of this and also the topological nature of the intrinsic AHE [18], it might be advantageous to use the Hall current from ferromagnets for magnetoelectronic devices, rather than the longitudinal current. Another purpose of this work is therefore to investigate the nature and spin polarization of the Hall current in Co-based Heusler compounds, knowledge of which is required for possible spintronic applications of the Hall current.

2. Theory and computational method

The intrinsic anomalous and spin Hall conductivities of a solid can be evaluated by using the Kubo formalism [19, 21, 22, 29]. The intrinsic Hall effect comes from the static limit ($\omega = 0$) of the off-diagonal elements of the optical conductivity [29]. Here we first calculate the imaginary part of the off-diagonal elements of the optical conductivity. Then we obtain the real part of the off-diagonal elements from the corresponding imaginary part by a Kramers–Kroning transformation. The intrinsic Hall conductivity $\sigma_{xy}^H$ is the static limit of the off-diagonal element of the optical conductivity $\sigma_{xy}^{\omega}(\omega = 0)$ (see [22] for more details). We note that the anomalous Hall conductivity (AHC) of bcc Fe [19] and the spin Hall conductivity (SHC) of fcc Pt [30] calculated in this way are in good agreement with that calculated directly by accounting for the Berry phase correction to the group velocity.

Since all the intrinsic Hall effects are caused by the spin–orbit coupling (SOC), first principles calculations must be based on a relativistic band theory. We calculate the relativistic band structure of the Co-based Heusler compounds ($Co_2XZ$) considered here using the highly accurate full-potential linearized augmented plane wave (FLAPW) method, as implemented in the WIEN2K code [31]. The self-consistent electronic structure calculations are based on the density functional theory (DFT) with the generalized gradient approximation (GGA) for
Table 1. Calculated total spin magnetic moment \( m_{\text{tot}}^s \) (\( \mu_B \) f.u.\(^{-1} \)), atomic spin magnetic moment \( m_s \) and orbital magnetic moment \( m_o \) (\( \mu_B \) atom\(^{-1} \)) as well as spin-decomposed density of states at the Fermi level \([N^\uparrow(E_F), N^\downarrow(E_F)]\) (states eV\(^{-1}\) f.u.\(^{-1}\)) of all the considered Heusler compounds Co\(_2\)XZ, together with the lattice constants \( a \) (Å) used and the number of the valence electrons \( n_v \) per formula unit (f.u.). For comparison, the available experimental magnetic moments are also listed. The orbital magnetic moment for the non-transition metal atoms \( m_o^Z \) is negligible, i.e. being less than 0.0001 \( \mu_B \) atom\(^{-1} \), and hence not listed here.

<table>
<thead>
<tr>
<th>Co(_2)XZ</th>
<th>( a )</th>
<th>( n_v )</th>
<th>( m_{\text{tot}}^s )</th>
<th>( m_s )</th>
<th>( m_s^X )</th>
<th>( m_o^s )</th>
<th>( m_o^X )</th>
<th>( N^\uparrow(E_F) )</th>
<th>( N^\downarrow(E_F) )</th>
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<td>Co(_2)CrAl</td>
<td>5.727(^{(a)})</td>
<td>27</td>
<td>2.998</td>
<td>0.804</td>
<td>1.533</td>
<td>−0.061</td>
<td>0.016</td>
<td>0.007</td>
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<td>5.630(^{(b)})</td>
<td>28</td>
<td>3.997</td>
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<td>−0.045</td>
<td>0.025</td>
<td>−0.008</td>
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<td>Co(_2)CrGe</td>
<td>5.805(^{(a)})</td>
<td>27</td>
<td>3.051</td>
<td>0.758</td>
<td>1.651</td>
<td>−0.053</td>
<td>0.016</td>
<td>0.006</td>
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<tr>
<td>Co(_2)CrGa</td>
<td>5.740(^{(b)})</td>
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<td>3.997</td>
<td>0.950</td>
<td>2.129</td>
<td>−0.033</td>
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<td>−0.013</td>
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<td>Co(_2)MnAl</td>
<td>5.755(^{(c)})</td>
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<td>0.760</td>
<td>2.730</td>
<td>−0.079</td>
<td>0.011</td>
<td>0.019</td>
<td>1.183</td>
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<td>5.654(^{(d)})</td>
<td>29</td>
<td>4.997</td>
<td>1.059</td>
<td>2.995</td>
<td>−0.052</td>
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<td>0.015</td>
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<tr>
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<td>0.754</td>
<td>2.794</td>
<td>−0.062</td>
<td>0.009</td>
<td>0.021</td>
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<tr>
<td>Co(_2)MnGe</td>
<td>5.743(^{(d)})</td>
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<td>4.999</td>
<td>0.999</td>
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<td>−0.041</td>
<td>0.030</td>
<td>0.019</td>
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<td>Co(_2)MnIn</td>
<td>5.990(^{(b)})</td>
<td>28</td>
<td>4.460</td>
<td>0.812</td>
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<td>0.017</td>
<td>0.023</td>
<td>2.321</td>
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<tr>
<td>Co(_2)MnSn</td>
<td>6.000(^{(d)})</td>
<td>29</td>
<td>5.033</td>
<td>0.974</td>
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<td>0.021</td>
<td>1.215</td>
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</tbody>
</table>

\(^{a}\) Experimental data [33].
\(^{b}\) GGA calculations (see footnote 4).
\(^{c}\) Experimental data [36].
\(^{d}\) Experimental data [10].

The lattice constants of Co\(_2\)CrSi, Co\(_2\)CrGe and Co\(_2\)MnIn were determined theoretically by the GGA [32] calculations by using the accurate projector augmented-wave method, as implemented in the Vienna Ab-initio Simulation Package [34, 35]. A large plane-wave cutoff energy of 350 eV and a \( 12 \times 12 \times 12 \) \( k \)-point mesh were used. 

The exchange correlation potential [32]. We consider only the fully ordered cubic Heusler compounds structure (L\(_2\)_1) and use the experimental lattice constants for all the considered Co\(_2\)XZ Heusler compounds except Co\(_2\)CrSi, Co\(_2\)CrGe and Co\(_2\)MnIn, as listed in table 1. Since the lattice constants of Co\(_2\)CrSi, Co\(_2\)CrGe and Co\(_2\)MnIn have not been reported, we have determined their lattice constants theoretically\(^4\). The SOC is included using the second variation technique [31] with the magnetization along the \( c \)-axis in all the present calculations. The wave function, charge density and potential were expanded in terms of the spherical harmonics inside the muffin-tin spheres and the cutoff angular moment \( L_{\text{max}} \) used is 10, 6 and 6, respectively. The wave function outside the muffin-tin spheres was expanded in terms of the augmented plane waves.
waves (APW) and a large number of augmented plane waves (about 70 APWs per atom, i.e. the maximum size of the crystal momentum \(K_{\text{max}} = 8/R_{\text{mt}}\)) were included in the present calculations. The improved tetrahedron method is used for the Brillouin-zone integration [37]. To obtain accurate ground-state charge density as well as spin and orbital magnetic moments, a fine \(36 \times 36 \times 36\) grid with 1240 \(k\)-points in the irreducible wedge in the first Brillouin zone was used.

### 3. Magnetic moments and half-metallicity

Let us first examine the calculated magnetic properties and band structures near the Fermi level of the considered Co-based Heusler compounds. Since the electronic structure and magnetism in the full Heusler compounds have been extensively studied (see, e.g., [11, 14, 15] and references therein), here we focus on only the salient features which may be related to the anomalous and spin Hall effects as well as spin polarization of the Hall current to be presented in the next two sections. The calculated total spin magnetic moment, local spin and orbital magnetic moments as well as spin-decomposed densities of states (DOSs) at the Fermi level (\(E_F\)) of all the considered Co-based Heusler compounds are listed in table 1, together with the available experimental total spin magnetic moments for comparison. The site-decomposed DOSs for the three representative Heusler compounds Co\(_2\)CrAl, Co\(_2\)MnAl and Co\(_2\)MnSi are displayed in figure 1.

Firstly, table 1 shows that Co\(_2\)CrZ (Z = Al, Si and Ge) and Co\(_2\)MnZ (Z = Si and Ge) are half-metallic, since the spin-down DOS at the Fermi level \([N^\downarrow(E_F)]\) for these compounds is zero. Therefore, their calculated total spin magnetic moments are almost an integer, as all the half-metallic ferromagnets should be. The tiny deviation of the total spin magnetic moment from an integer for these compounds is due to the inclusion of the SOC in the present calculations. Note that, in principle, including the SOC could induce a non-vanishing \(N^\downarrow(E_F)\) in the spin-down band gap of half-metals and hence could destroy the half-metallicity. Nevertheless, the \(N^\downarrow(E_F)\) was found to be very small in the Heusler compound NiMnSb although it could be large in heavy element compounds such as MnBi [38]. Indeed, the \(N^\downarrow(E_F)\) in the above-mentioned Co\(_2\)XZ compounds obtained with the SOC is negligible (see table 1). Co\(_2\)CrGa, Co\(_2\)MnAl, Co\(_2\)MnGa, Co\(_2\)MnSn and Co\(_2\)MnIn are not half-metallic (see table 1 and figure 1). Nevertheless, in Co\(_2\)CrGa, Co\(_2\)MnAl and Co\(_2\)MnSn, because the \(E_F\) is located only slightly below the top of the spin-down valence band (figure 1), the total spin magnetic moment deviates only slightly from an integer (see table 1 and figure 1).

Secondly, we find that the local spin magnetic moments on the 3d transition metal sites, namely Co and X (X = Cr and Mn), are large and coupled ferromagnetically (see table 1). The Co atoms have a spin magnetic moment ranging from 0.7 to 1.0 \(\mu_B\), and the spin magnetic moment of the Mn (Cr) atoms is around 3.0 (2.0) \(\mu_B\). In contrast, the local spin magnetic moments on the non-transition metal atoms Z (Z = Al, Si, Ga, Ge, In and Sn) are small and aligned antiparallel to that of the Co and X atoms. Not surprisingly, the orbital magnetic moments on the transition metal atoms (Co, Mn and Cr) are rather small, being about two orders of magnitude smaller than the spin magnetic moments, because of the weakness of the SOC in these 3d transition metal compounds. All the non-transition metal (Z) atoms have a practically zero orbital magnetic moment (i.e. \(\leq 0.0001 \mu_B\) atom\(^{-1}\)) and thus are not listed in table 1. The calculated total spin magnetic moments are in good agreement with previous theoretical calculations [11, 14] and also the experimental results [10].
Thirdly, we note that the calculated total spin magnetic moments for all the Heusler compounds more or less follow the so-called \( n_v - 24 \) rule (\( n_v \) is the number of valence electrons), as has already been reported in [11]. For 3d transition metals and their binary compounds, the total spin magnetic moment (\( m_{\text{tot}}^s \)) shows the well-known Slater–Pauling behavior [39]. This \( m_{\text{tot}}^s = n_v - 24 \) rule is essentially a generalized Slater–Pauling rule for the full-Heusler compounds. The reason is that in these compounds, the number of occupied spin-down states \( (n^\downarrow) \) is found to remain fixed at 12, at least when they are in the half-metallic state [11]. Therefore, \( m_{\text{tot}}^s = n_v - 2n^\downarrow = n_v - 24 \). For example, the \( n_v \) for Co\(_2\)CrAl and Co\(_2\)CrGa is 27, and the calculated \( m_{\text{tot}}^s \) are 3 \( \mu_B \). Similarly, the total spin magnetic moment is 4 \( \mu_B \) for Co\(_2\)MnAl, and 5 \( \mu_B \) for Co\(_2\)MnSi, Co\(_2\)MnGe, Co\(_2\)MnSn, also following this \( n_v - 24 \) rule. The obvious exceptions are Co\(_2\)MnGa and Co\(_2\)MnIn because they deviate strongly from the half-metallicity (table 1).

Displayed in figure 1 are the total and site decomposed DOSs of three selected Heusler compounds Co\(_2\)CrAl (a), Co\(_2\)MnAl (b) and Co\(_2\)MnSi (c). Figure 1 shows clearly that Co\(_2\)CrAl

![Figure 1. Total and site decomposed DOSs for (a) Co\(_2\)CrAl, (b) Co\(_2\)MnAl and (c) Co\(_2\)MnSi. The Fermi level is set to zero.](http://www.njp.org/)

and Co$_2$MnSi are half-metallic with the $E_F$ falling in the spin-down insulating gap, while Co$_2$MnAl is a normal ferromagnetic metal with the $E_F$ being located just below the top of the spin-down valence band. The DOS spectra for Co$_2$MnAl and Co$_2$MnSi are, in general, similar except for the location of the Fermi level. As one goes from Co$_2$MnSi to Co$_2$MnAl, the $n_v$ decreases by 1 and hence the $E_F$ is lowered from the middle of the spin-down insulating gap to that just below the top of the spin-down valence band. Similar situations occur for Co$_2$MnGa and Co$_2$MnGe, and thus their DOS spectra are not shown here. However, pronounced differences in the band structure between Co$_2$CrAl and Co$_2$MnAl exist. These differences are mainly caused by the different exchange splittings of the Cr and Mn 3d bands. The Mn atoms have a larger spin moment of $\sim 3.0 \mu_B$ and hence a larger 3d band exchange splitting, whilst the Cr atoms have a smaller spin moment of $\sim 2.0 \mu_B$ and hence a smaller exchange splitting (see table 1 and figure 1). Consequently, the Fermi level is located at the center of the spin-up 3d band in Co$_2$CrAl (figure 1(a)), while the spin-up 3d band in Co$_2$MnAl is mostly below the Fermi level (figure 1(b)). In short, the Co-based Heusler compounds considered here can be divided into two families, namely Co$_2$CrZ and Co$_2$MnZ. Within each family, the electronic structure and other physical properties for one member can be approximately obtained from another member by appropriately shifting the Fermi level. One exception is the pair of Co$_2$MnSn and Co$_2$MnIn.

4. Anomalous and spin Hall conductivities

A dense $k$ point mesh would be needed for obtaining accurate anomalous and spin Hall conductivities [19, 21, 29]. Therefore, we use several fine $k$-point meshes with the finest $k$-point mesh being $58 \times 58 \times 58$. To obtain the theoretical anomalous and spin Hall conductivities, we first calculate the AHC and SHC as a function of the number ($N_k$) of $k$-points in the first Brillouin zone. The calculated anomalous ($\sigma_{xy}^A$) and spin ($\sigma_{xy}^S$) Hall conductivities versus the inverse of the $N_k$ are then plotted and fitted to a first-order polynomial to obtain the converged theoretical $\sigma_{xy}^A$ and $\sigma_{xy}^S$ (i.e. the extrapolated value at $N_k = \infty$) (see [21, 22]). The theoretical $\sigma_{xy}^A$ and $\sigma_{xy}^S$ determined this way are listed in table 2.

Table 2 shows that the calculated $\sigma_{xy}^A$ and $\sigma_{xy}^S$ are large for Co$_2$MnAl, Co$_2$MnGa and Co$_2$MnIn, but are five to ten times smaller for the other compounds. This suggests that Co$_2$MnAl, Co$_2$MnGa and Co$_2$MnIn may find applications in, e.g., magnetic sensors [26]. Interestingly, the calculated $\sigma_{xy}^S$ seems to be about half of the $\sigma_{xy}^A$ for every compound considered here except Co$_2$CrGa. This may be attributed to the half-metallic behavior of these Heusler compounds. In the half-metallic materials, the charge current would flow only in one spin channel, and no charge current in the other spin channel, resulting in $\sigma_{xy}^S$ being twice as large as $\sigma_{xy}^A$. We will further discuss this point in the next section.

We note that the difference in the AHC between Co$_2$MnAl (Co$_2$MnGa) and Co$_2$MnSi (Co$_2$MnGe) is rather dramatic, while the number of valence electrons in Co$_2$MnAl (Co$_2$MnGa) differs from that in Co$_2$MnSi (Co$_2$MnGe) only by 1 (see table 1). In particular, the AHC in Co$_2$MnAl (Co$_2$MnGa) is about six times larger than that in Co$_2$MnSi (Co$_2$MnGe). This is somewhat surprising because one would expect a half-metallic metal with a larger magnetization to have a larger AHC than a normal ferromagnet with a smaller magnetization. To better understand this, we display the relativistic band structure as well as the AHC and $n_v$ as a function of the $E_F$ for three selected compounds Co$_2$CrAl, Co$_2$MnAl and Co$_2$MnSi in figure 2. Figures 2(d) and (g) suggest that the band structures of Co$_2$MnAl and Co$_2$MnSi are rather similar. The key difference is the location of the $E_F$ due to the difference in $n_v$ in these
Table 2. Calculated anomalous $[\sigma^A_{xy} \text{ (S cm}^{-1}\text{)}]$ and spin $[\sigma^S_{xy} \text{ (hS e}^{-1}\text{ cm}^{-1}\text{)}]$ Hall conductivities as well as spin-decomposed Hall conductivities ($\sigma^H_{xy}^{\uparrow}$, $\sigma^H_{xy}^{\downarrow}$) (S cm$^{-1}$), Hall current spin polarization $P^H$ (%), spin polarization of the electronic states at the Fermi level $P^D$ (%) and the ratio of spin current to charge current $\eta$.

<table>
<thead>
<tr>
<th>Co$_2$XZ</th>
<th>$\sigma^A_{xy}$</th>
<th>$\sigma^S_{xy}$</th>
<th>$\sigma^H_{xy}^{\uparrow}$</th>
<th>$\sigma^H_{xy}^{\downarrow}$</th>
<th>$P^H$</th>
<th>$P^D$</th>
<th>$\eta$</th>
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<td>Co$_2$CrAl</td>
<td>241 (438$^{(a)}$)</td>
<td>137</td>
<td>258</td>
<td>$-17.0$</td>
<td>114</td>
<td>100 (100$^{(a)}$)</td>
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<td>189</td>
<td>$-13.8$</td>
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<td>101</td>
<td>188</td>
<td>$-13.9$</td>
<td>116</td>
<td>75 (82$^{(a)}$)</td>
<td>1.16</td>
</tr>
</tbody>
</table>

$^a$ Theoretical results [25].
$^b$ Experimental data [40].
$^c$ Experimental data [26].

compounds. In other words, the band structure and other physical properties of Co$_2$MnSi may approximately be obtained from that of Co$_2$MnAl by raising the $E_F$ by about 0.5 eV due to one extra $p$ valence electron. Interestingly, there is a pronounced peak sitting at the $E_F$ in the $\sigma^A_{xy}$ spectrum of Co$_2$MnAl. This feature may be attributed to the prominent contributions to the AHC from the spin-down band pocket at the $\Gamma$ point and also the narrow spin-up bands along the Brillouin zone edges X–W and W–K (see figure 2, and also figure 5 in [25]). These band features also appear in the isoelectronic compound Co$_2$MnGa (not shown here), which has a large AHC too (table 2). However, when the Fermi level is raised to the highly dispersive Co and Mn d-orbital hybridized spin-up band region to accommodate one more valence electron as one goes from Co$_2$MnAl (Co$_2$MnGa) to Co$_2$MnSi (Co$_2$MnGe), these band features are now significantly below the $E_F$ (figure 2) with much diminished contributions to the AHC, thus resulting in the much reduced $\sigma^A_{xy}$ and $\sigma^S_{xy}$ in Co$_2$MnSi (Co$_2$MnGe) (table 2). Interestingly, the calculated $\sigma^A_{xy}$ is larger in Co$_2$MnGa than in Co$_2$MnAl. This difference in the $\sigma^A_{xy}$ could be attributed to the larger SOC in the Ga atoms than in the Al atoms. This finding prompted us to calculate the $\sigma^A_{xy}$ and $\sigma^S_{xy}$ for Co$_2$MnIn. Unfortunately, the calculated $\sigma^A_{xy}$ in Co$_2$MnIn is even smaller than that of Co$_2$MnAl.

However, as described in the preceding section, significant differences in the band structure between Co$_2$MnAl and Co$_2$CrAl exist and they are not simply due to the different locations of the $E_F$ in these compounds (figure 2). In particular, the top of the spin-down valence band which sticks out of the Fermi level in Co$_2$MnAl becomes completely submerged below the Fermi level in Co$_2$CrAl (figures 2(a) and (d)). These result in rather pronounced differences in the $\sigma^A_{xy}$ at the
**Figure 2.** Band structure (left panels), AHC ($\sigma^A_{ij}$) (middle panels) and the number of valence electrons per formula ($n_v$) (right panels) for Co$_2$CrAl (upper panels), Co$_2$MnAl (middle panels) and Co$_2$MnSi (bottom panels). The Fermi energy is shifted to zero. The red dashed ellipses in the left panels mark the predominantly spin-down energy bands.

$E_F$ and below. For example, the large broad peak in the $\sigma^A_{xy}$ spectrum between $-0.8$ and $0.0$ eV in Co$_2$MnAl (figure 2(e)) becomes a much reduced narrow peak in Co$_2$CrAl (figure 2(b)), and Co$_2$CrAl has a much smaller $\sigma^A_{xy}$ value of 241 S cm$^{-1}$ (table 2). Nevertheless, the calculated $\sigma^A_{xy}$ spectra above the $E_F$ in the two compounds are rather similar, both becoming negative at $\sim 1.0$ eV.

The AHC for Co$_2$CrAl was experimentally found to follow closely the magnetization with the ratio ($\sigma_{HI}$) of the two quantities being constant over a large temperature range [40], suggesting the intrinsic origin. Nonetheless, the measured AHC conductivity for Co$_2$CrAl is $\sim 125$ S cm$^{-1}$ [40], being only about half of our theoretical value (table 2). We note that the measured magnetization is only $1.65 \mu_B$ f.u.$^{-1}$ [40], being about two times smaller than our theoretical value of $3.0 \mu_B$ f.u.$^{-1}$ (table 1). Previous experimental studies and *ab initio* calculations showed that the intrinsic AHC is proportional to the magnetization [23, 40]. Therefore, the smaller experimental AHC may be attributed to the smaller observed magnetization in Co$_2$CrAl due to the presence of imperfections such as structural disorder [41]. If we use the measured $\sigma_{HI}$ and extrapolate the linear relation $\sigma^A_{xy} = \sigma_{HI} \times M$ to the theoretical...
magnetization (\(M\)) value, we obtain an ‘experimental’ \(\sigma_{xy}^A\) value of 227 S cm\(^{-1}\), being quite close to the theoretical \(\sigma_{xy}^A\) value of 241 S cm\(^{-1}\). The AHE in Co\(_2\)MnAl was recently measured by Vidal et al\cite{26}, and the Hall resistivity \(\rho_{xy}\) was found to be about 20 and 15 \(\mu\Omega\) cm in clean and dirty conditions, respectively. Using these experimental values together with the longitudinal resistivity of about 100 \(\mu\Omega\) cm, one can estimate the AHC to be in the range of 1500–2000 S cm\(^{-1}\). We note that our theoretical \(\sigma_{xy}^A\) of 1264 S cm\(^{-1}\) for Co\(_2\)MnAl is smaller than the estimated experimental values\cite{26} by about 15–40%. The discrepancy could be due to the fact that the Co\(_2\)MnAl samples used in the experiments contain mostly the B2 phase, instead of the L\(_2\)\(_1\) structure considered here. Note that the measured AHC values would contain both the intrinsic \(\sigma_{xy}^A\) and the contributions from extrinsic skew scattering and side jump mechanisms\cite{18}. Therefore, another source of the discrepancy could come from the extrinsic AHC which is not addressed here. In fact, recent \textit{ab initio} calculations based on the short-range disorder in the weak scattering limit\cite{42} indicated that the scattering-independent side jump contribution is of the order of \(\sim 100\) S cm\(^{-1}\) in 3d transition metal ferromagnets and L\(_1\)\(_2\) FePd and FePt. Similar values of the extrinsic AHC in L\(_1\)\(_2\) FePd\(_{1-x}\)Pt\(_x\) alloys were derived recently by comparing the measured AHC with the calculated intrinsic AHC\cite{24, 43}.

Our theoretical \(\sigma_{xy}^A\) value for Co\(_2\)MnSi agrees rather well with the theoretical value obtained recently by computing Berry curvatures\cite{25} (table 2). However, for Co\(_2\)CrAl, Co\(_2\)MnAl and Co\(_2\)MnSn, our theoretical \(\sigma_{xy}^A\) value differs significantly from that reported in\cite{25} by about 30–100\% (see table 2). We note that in\cite{25}, only 2000 \(k\)-points in the Brillouin zone were used and the convergence of the \(\sigma_{xy}^A\) was reported to be about 20\%. Also, here we used the accurate FLAPW method while, in contrast, the atomic spherical wave method with the atomic sphere approximation (ASA) was used in\cite{25}. Since the calculated AHC is sensitive to both the number of \(k\)-points in the Brillouin zone used and the details of the energy bands in the vicinity of the Fermi level\cite{19, 21, 22, 29}, the discrepancies in the calculated \(\sigma_{xy}^A\) between the two theoretical studies may perhaps be due to the fewer \(k\)-points in the Brillouin zone and the ASA used in\cite{25}.

### 5. Hall current spin polarization

The AHE has recently received intensive renewed interest mainly because of its close connection with spin-transport phenomena\cite{18}. Indeed, it could be advantageous to use the Hall current from a ferromagnet as a spin-polarized current source, instead of the longitudinal current, as mentioned before. Therefore, it would be interesting to know the spin polarization of the Hall current. The spin polarization \(P^H\) of the Hall current may be written as

\[
P^H = \frac{\sigma_{xy}^H\uparrow - \sigma_{xy}^H\downarrow}{\sigma_{xy}^H\uparrow + \sigma_{xy}^H\downarrow},
\]

where \(\sigma_{xy}^H\uparrow\) and \(\sigma_{xy}^H\downarrow\) are the spin-up and spin-down Hall conductivities, respectively. The \(\sigma_{xy}^H\uparrow\) and \(\sigma_{xy}^H\downarrow\) can be obtained from the calculated AHC and SHC via the relations

\[
\sigma_{xy}^A = \sigma_{xy}^H\uparrow + \sigma_{xy}^H\downarrow,
\]

\[
\frac{2e}{h^*}\sigma_{xy}^S = \sigma_{xy}^H\uparrow - \sigma_{xy}^H\downarrow.
\]

Note that the absolute value of \(P^H\) can be greater than 1.0 because the spin-decomposed Hall currents can go either right (positive) or left (negative). In the non-magnetic materials, the charge
Hall current is zero, and hence, \( \sigma_{xy}^{H} = -\sigma_{xy}^{H} \) results in \( P^H = \infty \). Also of interest is the ratio of the spin current to charge current \( \eta \) which can be written as \( \eta = |(2e/h)\sigma_{xy}^S/\sigma_{xy}^A| \). It should be pointed out that the above decomposition of the Hall conductivity into the simple spin-up and spin-down contributions (\( \sigma_{xy}^{H\uparrow} \) and \( \sigma_{xy}^{H\downarrow} \)) is valid only for metals containing light elements such as 3d transition metals. Because the SOC generally mixes spin-up and spin-down states, the Hall conductivity contains both the spin-conserving part and spin-non-conserving (spin-flipping) part [44], and the occurrence of the pronounced spin-non-conserving contribution would ruin the perfect two-current model (i.e. the above simple decomposition). The calculated spin-non-conserving contribution is indeed large in L1\(_0\) FePt but becomes small already in L1\(_0\) FePd [44].

The calculated \( \sigma_{xy}^{H\uparrow}, \sigma_{xy}^{H\downarrow}, P^H \) and \( \eta \) are listed in table 2. Interestingly, table 2 indicates that unlike the longitudinal charge current, the spin-up and spin-down Hall currents would flow in opposite directions in all the Co-based Heusler compounds considered except Co\(_2\)MnIn. Nonetheless, the spin-up Hall conductivity is more than ten times larger than the spin-down Hall conductivity in all these compounds except Co\(_2\)CrGa. In other words, the Hall current carriers are mostly of spin-up particles. Remarkably, this gives rise to the nearly 100% spin polarization of the Hall current in all the Heusler compounds except Co\(_2\)CrGa (table 2). Furthermore, the spin current to charge current ratio \( \eta \) in these compounds is also very high, being just over 1.0. Therefore, both the \( P^H \) and \( \eta \) of the Hall current in all the Co-based Heusler compounds except Co\(_2\)CrGa are close to or even better than the corresponding values of the longitudinal current from an ideal half-metallic ferromagnet, even though some of them are not half-metallic. This suggests that the charge Hall current from these compounds is promising for spintronic applications. Surprisingly, the spin-down Hall conductivity is non-negligible even in the half-metallic metals, corroborating that the occupied states well below the \( E_F \) would also contribute to the AHE [19].

The spin polarization of a magnetic material is usually described in terms of the spin-decomposed DOSs at the Fermi level as follows:

\[
P^D = \frac{N_\uparrow(E_F) - N_\downarrow(E_F)}{N_\uparrow(E_F) + N_\downarrow(E_F)},
\]

where \( N_\uparrow(E_F) \) and \( N_\downarrow(E_F) \) are the spin-up and spin-down DOSs at the \( E_F \), respectively. The spin polarization \( P^D \) would then vary from \(-1.0\) to \(1.0\) only. For the half-metallic materials, \( P^D \) equals either \(-1.0\) or \(1.0\). The calculated \( N_\uparrow(E_F) \) and \( N_\downarrow(E_F) \) for the Heusler compounds are listed in table 1, and the corresponding \( P^D \) are listed in table 2. In terms of \( P^D \), only Co\(_2\)CrAl, Co\(_2\)CrSi, Co\(_2\)CrGe, Co\(_2\)MnSi and Co\(_2\)MnGe are half-metallic. Interestingly, we note that in the non-half-metallic metals Co\(_2\)MnAl, Co\(_2\)MnGa, Co\(_2\)MnIn and Co\(_2\)MnSn, the \( P^D \) is significantly smaller than the \( P^H \), which is close to 100% (see table 2). Therefore, we may perhaps regard these compounds as anomalous Hall half-metals. As pointed out by researchers previously [45], the spin polarization \( P^D \) defined by equation (3) is not necessarily the spin polarization of the transport currents measured in the experiments. This can be clearly illustrated by magnetically anisotropic materials such as hcp Co. In hcp Co, the spin-decomposed DOSs at the \( E_F \) and hence \( P^D \) are independent of the magnetization direction, while, in contrast, the Hall conductivities and hence \( P^H \) change dramatically as the magnetization rotates [22]. From the viewpoint of spintronic applications, only the current spin polarizations such as \( P^H \), instead of the \( P^D \), count.
6. Conclusions

The anomalous and spin Hall conductivities as well as the electronic and magnetic properties of Co-based full Heusler compounds $\text{Co}_2\text{XZ}$ ($\text{X} = \text{Cr}$ and Mn; $\text{Z} = \text{Al}$, Si, Ga, Ge, In and Sn) have been calculated within the DFT with the GGA. The highly accurate FLAPW method is used. Interestingly, it is found that the spin-up and spin-down Hall currents would flow in opposite directions in all the Co-based Heusler compounds considered except $\text{Co}_2\text{MnIn}$, although the spin-up Hall conductivity is more than ten times larger than the spin-down Hall conductivity in these compounds except $\text{Co}_2\text{CrGa}$. As a result, the charge Hall current in all the Heusler compounds considered except $\text{Co}_2\text{CrGa}$ would be almost fully spin-polarized, even though $\text{Co}_2\text{MnAl}$, $\text{Co}_2\text{MnGa}$, $\text{Co}_2\text{MnIn}$ and $\text{Co}_2\text{MnSn}$ are not half-metallic from the viewpoint of the electronic structure. Moreover, the ratio of the accompanying spin current to the charge Hall current is slightly larger than 1.0. Based on these results, therefore, these Heusler compounds may be called anomalous Hall half-metals. These anomalous Hall half-metals, especially $\text{Co}_2\text{MnAl}$, $\text{Co}_2\text{MnGa}$ and $\text{Co}_2\text{MnIn}$ which have large anomalous and spin Hall conductivities, could find valuable applications in spintronics such as magnetoresistive and spin-torque switching nanodevices as well as spin valves. The calculated electronic band structures and magnetic moments as well as anomalous and spin Hall conductivities as a function of the Fermi level, are used to analyze these interesting findings. It is hoped that these interesting results would stimulate further experimental investigations into the AHE and also the characteristics of the Hall current in these Co-based Heusler compounds.

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