Supplemental Material: The chiral Hall effect in canted ferromagnets and antiferromagnets

J. Kipp,[1,2] K. Samanta,[1] F. R. Lux,[1,2] M. Merte,[1,2,3] D. Go,[3,1] J.-P. Hanke,[1]
M. Redies,[1,2] F. Freimuth,[1] S. Blügel,[1] M. Ležaić,[1] and Y. Mokrousov[1,3]

[1]*Peter Grünberg Institut and Institute for Advanced Simulation, Forschungszentrum Jülich and JARA, 52425 Jülich, Germany*
[2]*Department of Physics, RWTH Aachen University, 52056 Aachen, Germany*
[3]*Institute of Physics, Johannes Gutenberg University Mainz, 55099 Mainz, Germany*

Supplementary Note 1: Symmetry analysis of the crystal and chiral Hall effect for the hexagonal magnetic Rashba model

The Rashba model on the hexagonal lattice which is studied in the manuscript belongs to the crystallographic point group C_{6v} in the non-magnetic case. When the system is magnetic, it is characterized by the respective magnetization vectors $\mathbf{s}_i$ at each of the sites $i = A, B$. The direction cosine expansion in the manuscript is expressed in terms of $\mathbf{n}_\pm = \mathbf{s}_\mathrm{A} \pm \mathbf{s}_\mathrm{B}$, i.e,

$$\sigma_{xy}^{\mathrm{odd}} = \sum_{k,l=0}^{\infty} (c_{xy}^{\mathrm{odd}})^i : (\mathbf{n}_-^{\otimes 2k+1} \otimes \mathbf{n}_+^{\otimes 2l})_i \tag{11}$$

$$\sigma_{xy}^{\mathrm{even}} = \sum_{k,l=0}^{\infty} (c_{xy}^{\mathrm{even}})^i : (\mathbf{n}_-^{\otimes 2k} \otimes \mathbf{n}_+^{\otimes 2l+1})_i. \tag{12}$$

Accodringly, the lowest orders are thus given by the general expansions

$$\sigma_{xy}^{\mathrm{odd}} = (c_{xy}^{\mathrm{odd}})^i (n_-)_i + (c_{xy}^{\mathrm{odd}})^{ikl} (n_-)_i (n_+)_k (n_+)_l + \mathcal{O}(n^5) \tag{13}$$

$$\sigma_{xy}^{\mathrm{even}} = (c_{xy}^{\mathrm{even}})^i (n_+)_i + (c_{xy}^{\mathrm{even}})^{ikl} (n_+)_i (n_-)_k (n_-)_l + \mathcal{O}(n^5). \tag{14}$$

If a physical tensor T of rank $n = k + l$ connects k polar and l axial vectors, its transformation law under a symmetry transformation S is usually given by

$$T^{p_1 \cdots p_k a_1 \cdots a_l} \quad \to \quad T^{p_1 \cdots p_k a_1 \cdots a_l} |S|^l S_{p_1}^{\tilde{p}_1} \cdots S_{p_k}^{\tilde{p}_k} S_{a_1}^{\tilde{a}_1} \cdots S_{a_l}^{\tilde{a}_l}, \tag{15}$$

where $|S| = \det S$. Whether $\hat{\mathbf{n}}_\pm$ transforms as a polar or axial vector depends however on the position of the magnetic atoms and which symmetry transformations interchange the lattice sites. For example, while $P\hat{\mathbf{n}}_{A,B} = P\hat{\mathbf{n}}_{A,B}$, one has $P\hat{\mathbf{n}}_- = -\hat{\mathbf{n}}_-$ if P interchanges A and B, which would be the case for magnetic atoms on the hexagonal lattice. In other words, $\hat{\mathbf{n}}_-$ behaves as a polar vector under P while $\hat{\mathbf{n}}_+$ behaves as an axial vector. Taking the possible interchange of magnetic lattice sites into account for an arbitrary symmetry transformation S, the transformation law is generalized to

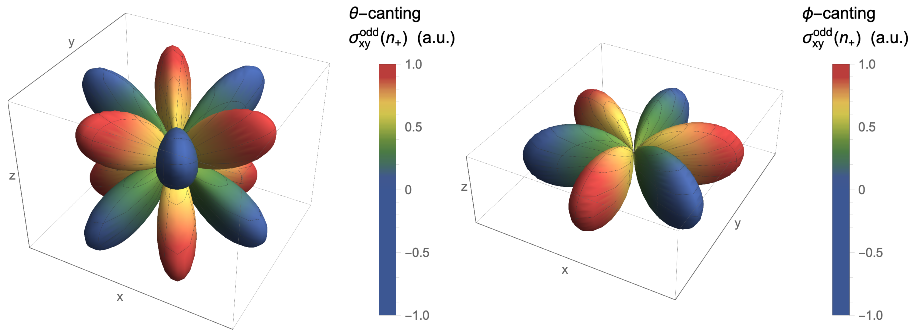
$$T^{i_1 \cdots i_n} \quad \to \quad T^{i_1 \cdots i_n} \mathrm{swap}(S)^\eta |S|^\epsilon S_{i_1}^{\tilde{i}_1} \cdots S_{i_n}^{\tilde{i}_k}, \tag{16}$$

where $\mathrm{swap}(S) = -1$, if S interchanges the lattice sites A and B and $\mathrm{swap}(S) = 1$ otherwise. As before, a tensor with $\epsilon = 0$ is called polar and with $\epsilon = 1$ it is axial. If $\eta = 1$ instead of $\eta = 0$, the tensor is called *staggered*, since its build to compensate the coupling to the staggered fields n_- and their unusual behavior under symmetry transformations. In this terminology, the coefficients of $\sigma_{xy}^{\mathrm{odd}}$ qualify as staggered axial tensors, while the coefficients of $\sigma_{xy}^{\mathrm{even}}$ belong to an ordinary axial tensor. This fixes their transformation behavior and one finds

$$\frac{\sigma_{xy}^{\mathrm{odd}} - \sigma_{yx}^{\mathrm{odd}}}{2} \in \mathrm{span}\{n_-^y n_+^y n_+^x + n_-^y n_+^x n_+^y + n_-^x n_+^y n_+^y - n_-^x n_+^x n_+^x\} \; + \mathcal{O}(n^5) \tag{17}$$

$$\frac{\sigma_{xy}^{\mathrm{even}} - \sigma_{yx}^{\mathrm{even}}}{2} = \in \mathrm{span}\{n_+^z, n_-^z n_-^x n_+^x + n_-^z n_-^y n_+^y,\; n_-^x n_-^z n_+^x + n_-^y n_-^z n_+^y,\; n_-^x n_-^x n_+^z + n_-^y n_-^y n_+^z\} \; + \mathcal{O}(n^5), \tag{18}$$

for the point group C_{6v} (the symmetry of the Rashba model on the hexagonal lattice as presented in the manuscript) and where the x-direction is defined as the axis which contains both atoms of the unit cell. Since $\mathbf{n}_+ \cdot \mathbf{n}_- = 0$, one has $\boldsymbol{\chi} \times \mathbf{n}_\pm = \mp \|\mathbf{n}_\pm\|^2 \mathbf{n}_\mp / 2$,



(a) Canting with respect to the θ-angle (b) Canting with respect to the ϕ-angle

Supplementary Fig. 1: Symmetry analysis of the chiral Hall effect in ferromagnets. The two figures show the parametric surface of the antisymmetrized contributions to $\sigma_{xy}^{\mathrm{odd}}$, parametrized by the direction of the ferromagnetic order parameter $\mathbf{n}_{+}$. The antiferromagnetic order parameter is induced by canting the ferromagnetic state either with respect to the polar angle θ, or the azimuthal angle ϕ. The x-axis is defined to contain the two atoms of the unit cell.

which can be used to introduce the chirality to these results. One finds

$$n_{-}^{y} n_{+}^{y} n_{+}^{x} + n_{-}^{y} n_{+}^{x} n_{+}^{y} + n_{-}^{x} n_{+}^{y} n_{+}^{y} - n_{-}^{x} n_{+}^{x} n_{+}^{x} = -\frac{2}{\|\mathbf{n}_{+}\|^{2}} \begin{pmatrix} -2 n_{+}^{x} n_{+}^{y} n_{+}^{z} \\ ((n_{+}^{y})^{2} - (n_{+}^{x})^{2}) n_{+}^{z} \\ 3(n_{+}^{x})^{2} n_{+}^{y} - (n_{+}^{y})^{3} \end{pmatrix} \cdot \boldsymbol{\chi}. \tag{19}$$

This result can be recast into the form

$$\frac{\sigma_{xy}^{\mathrm{odd}} - \sigma_{yx}^{\mathrm{odd}}}{2} \propto \mathbf{n}_{+}^{T} \Xi\, \boldsymbol{\chi} + \mathcal{O}(\chi^{3}), \tag{110}$$

with the angular-dependent coupling matrix

$$\Xi = -2 \begin{pmatrix} -2 \hat{n}_{+}^{y} \hat{n}_{+}^{z} & 0 & 0 \\ 0 & 0 & (3(\hat{n}_{+}^{x})^{2} - (\hat{n}_{+}^{y})^{2}) \\ 0 & ((\hat{n}_{+}^{y})^{2} - (\hat{n}_{+}^{x})^{2}) & 0 \end{pmatrix}, \tag{111}$$

and where $\hat{\mathbf{n}}_{+} = \mathbf{n}_{+}/\|\mathbf{n}_{+}\|$. In analogy to the topological Hall effect, one might expect the emergence of an isotropic coupling of the form $\mathbf{n}_{+} \cdot \boldsymbol{\chi}$, known as the *scalar spin chirality*. For the magnetic Rashba model however, this quantity is always zero, i.e., $\mathbf{n}_{+} \cdot \boldsymbol{\chi} = 0$. A finite effect can still arise through anisotropic deviations from this isotropic coupling which are ultimately induced by the spin-orbit interaction.

The symmetry of the odd tensor can be visualized by applying the θ-canting procedure as defined in Fig. 2a) of the manuscript. For $\mathbf{n}_{-} = \mathrm{const}$, Fig. 1a) explores the angular dependence of the tensor, by varying the spherical angles of $\mathbf{n}_{+}$ and plotting the magnitude of $(\sigma_{xy}^{\mathrm{odd}}(\theta) - \sigma_{xy}^{\mathrm{odd}}(-\theta))/2$ as a parametric surface. The result reveals that the effect vanishes when $\mathbf{n}_{+}$ is contained in the yz mirror plane, i.e., when $n_{+}^{x} = 0$. Likewise, the θ-canting is ineffective when $n_{+}^{z} = 0$. A dual situation occurs if the canting is not induced via the polar angle θ, but via the azimuthal angle ϕ. The parametric surface which corresponds to $(\sigma_{xy}^{\mathrm{odd}}(\phi) - \sigma_{xy}^{\mathrm{odd}}(-\phi))/2$ is shown in Fig. 1b). In this case, the effect is largest when $n_{+}^{z} = 0$ and it vanishes when $\mathbf{n}_{+}$ is contained in the xz mirror plane.

A: Influence of mirror planes

Since the mirror planes seem to define an important characteristic of the chiral Hall effect on the hexagonal lattice, it is worthwhile to study in detail how they enter the symmetry analysis. In a first step, one can define the auxiliary variables

$$m_{i} = \begin{cases} -1, & \text{if } i = y \\ 1, & \text{otherwise.} \end{cases}, \tag{112}$$

which are used to specify the xz mirror operator, given by $(M)^i_j = \delta^i_j m_i$. Therefore $|M| = -1$. The transformation law for the coefficients in Eq. (13) and Eq. (14) now reads

$$c_{xy}^{i_1\cdots i_n} \overset{!}{=} c_{xy}^{i_1\cdots i_n} \mathrm{swap}(M)^\eta |M|^\epsilon m_x m_y \prod_i m_i. \tag{113}$$

This means that $c_{xy}^{i_1\cdots i_n} = 0$, if

$$\mathrm{swap}(M)^\eta |M|^\epsilon m_x m_y \prod_i m_i = -1. \tag{114}$$

The two atoms are along the x-axis. M, therefore, does not swap the atoms. The criterion above then reduces to

$$\prod_i m_i = -1 \tag{115}$$

Couplings to an odd number of $n^y_\pm$ components are therefore forbidden by the mirror symmetry. Only an even number of $n^y_\pm$ factors is allowed. Considering instead the yz mirror plane amounts to the redefinition

$$m_i = \begin{cases} -1, & \text{if } i = x \\ 1, & \text{otherwise.} \end{cases} \tag{116}$$

In this case however, one finds $\mathrm{swap}(M) = -1$. And tensor elements for which

$$(-1)^\eta \prod_i m_i = -1, \tag{117}$$

are zero. If the tensor is staggered (odd in $\hat{\mathbf{n}}_-$) one has $\eta = 1$ and a coupling to an even number of $n^x_\pm$ factors is forbidden. On the other hand, if the tensor is not staggered (even in $\hat{\mathbf{n}}_-$), couplings to an odd number of $n^x_\pm$ is forbidden. Consider now the case where $n^x_+ = 0$. A finite staggered tensor can then only arise from an odd number of n^x_- couplings.

Supplementary Note 2: Linear perturbation of the Berry curvature

The goal of this section is to present a derivation of the first order derivative of the Berry curvature as presented in the manuscript in a manifestely gauge-invariant way. Consider a continuous partition of unity by a complete set of states:

$$\mathrm{id} = \sum_n |n(\boldsymbol{\lambda})\rangle \langle n(\boldsymbol{\lambda})| \,. \tag{21}$$

It is parametrized by a vector $\boldsymbol{\lambda}$ belonging to the phase space $\mathcal{M}$, representing for example the momentum in the first Brillouin zone and any additional adiabatic real-space coordinates. This parametrization is inherited from the Hamiltonian, which can be considered as a map from $\mathcal{M}$ to the space of bounded operators on the Hilbert space $\mathcal{H}$:

$$H\colon \mathcal{M} \to \mathcal{B}(\mathcal{H}) \tag{22}$$

$$\boldsymbol{\lambda} \mapsto H_{\boldsymbol{\lambda}}. \tag{23}$$

We consider the more general class of these maps which form the space $C^\infty(\mathcal{M};\mathcal{B}(\mathcal{H}))$ of smooth phase space operators. Given such an operator $\hat{o}$, we can differentiate it with respect to the phase space coordinates. By simultaneously expanding in terms of the instantaneous basis set, one obtains a notion of covariant differentiation:

$$\begin{aligned} \partial_i \hat{o}_{\boldsymbol{\lambda}} &= \partial_i \sum_{nm} \hat{o}^{nm}_{\boldsymbol{\lambda}} |n\rangle \langle m| \\ &= \sum_{nm} (\partial_i \hat{o}^{nm}_{\boldsymbol{\lambda}}) |n\rangle \langle m| + \hat{o}^{nm}_{\boldsymbol{\lambda}} \big(\partial_i |n\rangle\big) \langle m| + \hat{o}^{nm}_{\boldsymbol{\lambda}} |n\rangle \big(\partial_i \langle m|\big) \\ &\equiv \nabla_i \hat{o}_{\boldsymbol{\lambda}} - i[\mathcal{A}_i, \hat{o}_{\boldsymbol{\lambda}}]. \end{aligned} \tag{24}$$

Here, we have omitted the explicit notation for the parameter dependence of the states for simplicity. Further, we introduced the non-abelian Wilzcek-Zee connection

$$\mathcal{A}^{nm}_i \equiv i \langle n|\partial_i|m\rangle \,. \tag{25}$$

Using the adjoint representation, this result can be concisely expressed as

$$\partial_i = \nabla_i - i\mathfrak{A}_i, \tag{26}$$

where $\mathfrak{A}_i = \mathrm{ad}\,\mathcal{A}_i = [\mathcal{A}_i, \bullet]$. By construction, the associated curvature is flat. The reason is that the partial derivatives commute, i.e., one has $[\partial_i, \partial_j]\hat{o}_{\boldsymbol{\lambda}} = 0$. Nevertheless, we can construct it as

$$\begin{aligned}\mathfrak{F}_{ij} &= i[\partial_i, \partial_j] \\ &= i[\nabla_i - i\mathfrak{A}_i, \nabla_j - i\mathfrak{A}_j] \\ &= [\nabla_i, \mathfrak{A}_j] - [\nabla_j, \mathfrak{A}_i] - i[\mathfrak{A}_i, \mathfrak{A}_j] \\ &= \nabla_i\mathfrak{A}_j - \nabla_j\mathfrak{A}_i - i[\mathfrak{A}_i, \mathfrak{A}_j],\end{aligned} \tag{27}$$

which is the adjoint representation of

$$\mathcal{F}_{ij} = \nabla_i\mathcal{A}_j - \nabla_j\mathcal{A}_i - i[\mathcal{A}_i, \mathcal{A}_j] = \partial_i\mathcal{A}_j - \partial_j\mathcal{A}_i + i[\mathcal{A}_i, \mathcal{A}_j] = 0. \tag{28}$$

Since the right-hand side evaluates to zero for the flat connection, one has the two identities

$$\nabla_i\mathcal{A}_j - \nabla_j\mathcal{A}_i = +i[\mathcal{A}_i, \mathcal{A}_j] \tag{29}$$

$$\partial_i\mathcal{A}_j - \partial_j\mathcal{A}_i = -i[\mathcal{A}_i, \mathcal{A}_j]. \tag{210}$$

Let $|n(\boldsymbol{\lambda})\rangle$ now represent the parameter-dependent eigenstates of the Hamiltonian. The abelian Berry curvature tensor of the n-th band is then given by

$$\Omega^n_{\mu\nu}(\boldsymbol{\lambda}) = i\sum_{m\neq n}\frac{(\partial_\mu H)^{nm}(\partial_\nu H)^{mn}}{(\epsilon_n - \epsilon_m)^2} - (\mu \leftrightarrow \nu). \tag{211}$$

Since one has $(\partial_\mu H)^{nm} = i(\epsilon_n - \epsilon_m)\mathcal{A}^{nm}_\mu$ for $n \neq m$, this can also be written as

$$\Omega^n_{\mu\nu}(\boldsymbol{\lambda}) = -i[\mathcal{A}_\mu, \mathcal{A}_\nu]^{nn} = (\partial_\mu\mathcal{A}_\nu - \partial_\nu\mathcal{A}_\mu)^{nn} \equiv \Omega^{nn}_{\mu\nu}(\boldsymbol{\lambda}), \tag{212}$$

which is therefore in accordance with Eq. (210). We can take this as the definition of the abelian part of the more general, but flat curvature tensor:

$$\mathcal{F}_{\mu\nu} = \Omega_{\mu\nu} + i[\mathcal{A}_\mu, \mathcal{A}_\nu] = 0. \tag{213}$$

For the calculation of the anomalous Hall effect, one weights the Berry curvature of the respective bands with the electronic density matrix ρ, from which one obtains the total Berry curvature

$$\Omega^{\mathrm{tot}}_{\mu\nu}(\boldsymbol{\lambda}) = -i\ \mathrm{tr}\ \rho[\mathcal{A}_\mu, \mathcal{A}_\nu] = \Im\ \mathrm{tr}\ \rho[\mathcal{A}_\mu, \mathcal{A}_\nu], \tag{214}$$

and where the density matrix is given by

$$\rho(\boldsymbol{\lambda}) = \sum_n n_{\mathrm{F}}(\epsilon_n)\,|n\rangle\,\langle n|\,, \tag{215}$$

where n_{F} is the Fermi-Dirac distribution at the chemical potential μ. Having introduced the necessary terminology, the first order derivation of the Berry curvature tensor can now be constructed. Several terms can contribute:

$$\begin{aligned}\partial_\kappa\Omega^{\mathrm{tot}}_{\mu\nu} &= (\Im\ \mathrm{tr}\ \partial_\kappa\rho\mathcal{A}_\mu\mathcal{A}_\nu + \Im\ \mathrm{tr}\ \rho\partial_\kappa\mathcal{A}_\mu\mathcal{A}_\nu + \Im\ \mathrm{tr}\ \rho\mathcal{A}_\mu\partial_\kappa\mathcal{A}_\nu) - (\mu \leftrightarrow \nu) \\ &= \Im\ \mathrm{tr}\,(\nabla_\kappa\rho\mathcal{A}_\mu\mathcal{A}_\nu - i[\mathcal{A}_\kappa, \rho]\mathcal{A}_\mu\mathcal{A}_\nu + \rho[\partial_\kappa\mathcal{A}_\mu, \mathcal{A}_\nu]) - (\mu \leftrightarrow \nu).\end{aligned} \tag{216}$$

As $\Omega^{\mathrm{tot}}_{\mu\nu}$ is gauge-invariant quantity, its derivative is gauge-invariant as well, and so is the right-hand side of this equation. The first term is a Fermi surface contribution which we single out:

$$(\partial_\kappa\Omega^{\mathrm{tot}}_{\mu\nu})^{\mathrm{FS}} = \Im\ \mathrm{tr}\nabla_\kappa\rho[\mathcal{A}_\mu, \mathcal{A}_\nu]. \tag{217}$$

It vanishes for insulators in the zero temperature limit and we neglect it in the following. The remaining terms are

$$\begin{aligned}(\partial_\kappa \Omega^{\text{tot}}_{\mu\nu}) &= \Im\,\text{tr}\rho\left(-i[\mathcal{A}_\mu \mathcal{A}_\nu, \mathcal{A}_\kappa] + [\partial_\kappa \mathcal{A}_\mu, \mathcal{A}_\nu]\right) - (\mu \leftrightarrow \nu) \\ &= \frac{1}{2}\Im\,\text{tr}\rho\left([\Omega_{\mu\nu}, \mathcal{A}_\kappa] + [\mathcal{Q}_{\kappa\mu}, \mathcal{A}_\nu] + [\Omega_{\kappa\mu}, \mathcal{A}_\nu]\right) - (\mu \leftrightarrow \nu) \\ &= \frac{1}{2}\Im\,\text{tr}\rho\left([\Omega_{\mu\nu}, \mathcal{A}_\kappa] + [\mathcal{Q}_{\kappa\mu}, \mathcal{A}_\nu] + [\Omega_{\nu\kappa}, \mathcal{A}_\mu]\right) - (\mu \leftrightarrow \nu),\end{aligned} \tag{218}$$

where we have introduced

$$\mathcal{Q}_{\mu\nu} = \partial_\mu \mathcal{A}_\nu + \partial_\nu \mathcal{A}_\mu, \tag{219}$$

which is related to the quantum metric tensor.