Supplementary material for "Orientation Dependence of the Intrinsic Anomalous Hall Effect in hcp Cobalt"

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In this supplementary material we first present the phenomenological expansion of the anomalous Hall conductivity $\boldsymbol{\sigma}^a$ of an hcp ferromagnet to third order in powers of the magnetization $\mathbf{M}_s$, and for fixed magnitude of $\mathbf{M}_s$ we re-express it in terms of $l=1$ and $l=3$ spherical harmonics. Secondly, the computational details of the first-principles calculations are given. Finally, the relation between the Hall resistivity and the Hall conductivity is derived and applied to hcp Co.

PHENOMENOLOGICAL EXPANSION OF THE HALL CONDUCTIVITY OF AN HCP CRYSTAL

The orientation dependence of $\boldsymbol{\sigma}^a(\mathbf{M}_s)$ can be described phenomenologically by expanding in powers of the direction cosines $\{\alpha_i\}$ of $\mathbf{M}_s$ [1, 2]:

$$\sigma^a_{ij}(\hat{\mathbf{m}}_s) = a_{ijp}\alpha_p + a_{ijpqr}\alpha_p\alpha_q\alpha_r + \ldots \qquad (1)$$

Here $\hat{\mathbf{m}}_s = \alpha_x\hat{\mathbf{x}} + \alpha_y\hat{\mathbf{y}} + \alpha_z\hat{\mathbf{z}}$ is the unit vector along the spin magnetization $\mathbf{M}_s$, and it is assumed that the magnitude of $\mathbf{M}_s$ is independent of orientation, as is the case for hcp Co to a very good approximation. Crystal symmetry and other considerations reduce the number of independent coefficients in Eq. (1). They are tabulated for the hcp structure to third order in Ref. [1]:

$$\begin{cases} \sigma^a_1 = a_{231}\alpha_1 + a_{23111}(\alpha_1^3 + \alpha_1\alpha_2^2) + a_{31233}\alpha_1\alpha_3^2 \\ \sigma^a_2 = a_{231}\alpha_2 + a_{23111}(\alpha_2^3 + \alpha_2\alpha_1^2) + a_{31233}\alpha_2\alpha_3^2 \\ \sigma^a_3 = a_{123}\alpha_3 + a_{12113}(\alpha_3\alpha_1^2 + \alpha_3\alpha_2^2) + a_{12333}\alpha_3^3 \end{cases} . \qquad (2)$$

The uniaxial nature of the hcp structure implies $a_{231} \neq a_{123}$, producing a misalignement between $\boldsymbol{\sigma}^a$ and $\mathbf{M}_s$ to first order in the expansion.

There are six coefficients in Eq. (2), the number needed to describe $\boldsymbol{\sigma}^a$ to third order as a function of the orientation *and* magnitude of $\mathbf{M}_s$. For fixed magnitude, the angular dependence requires only four independent parameters, which are conveniently chosen as the coefficients of an expansion in orthonormal real spherical harmonics $\overline{Y}_{lm}(\theta,\varphi)$. The projections $\int \sigma^a_i(\theta,\varphi)\overline{Y}_{lm}(\theta,\varphi)d\Omega$ have been calculated using a symbolic manipulation package, yielding

$$\begin{cases} \sigma^a_1 = c_{11}\overline{Y}_{11} + c_{31}\overline{Y}_{31} \\ \sigma^a_2 = c_{11}\overline{Y}_{1,-1} + c_{31}\overline{Y}_{3,-1} \\ \sigma^a_3 = c_{10}\overline{Y}_{10} + c_{30}\overline{Y}_{30} \end{cases} . \qquad (3)$$

Using the conventions of Ref. [3] to define the real spherical harmonics, the coefficients are given by

$$\begin{cases} c_{10} = \frac{2}{5}\sqrt{\frac{\pi}{3}}(5a_{123} + 2a_{12113} + 3a_{12333}) \\ c_{11} = -\frac{2}{5}\sqrt{\frac{\pi}{3}}(5a_{231} + a_{31233} + 4a_{23111}) \\ c_{30} = -\frac{4}{5}\sqrt{\frac{\pi}{7}}(a_{12113} - a_{12333}) \\ c_{31} = -\frac{4}{5}\sqrt{\frac{2\pi}{21}}(a_{31233} - a_{23111}) \end{cases} . \qquad (4)$$

Eq. (3) is the same as Eq. (4) in the main text, where the four coefficients c_{10}, c_{11}, c_{30}, and c_{31} were obtained from a least-squares fitting to the first-principles calculations at $T=0$. The rather good fit and, more generally, the smooth orientation dependence of the calculated intrinsic AHC seen in Fig. 1 of the main text should be contrasted with the oscillatory behavior found when scanning the chemical potential [4]. The comparatively less dramatic angular dependence can perhaps be rationalized by noting that the zero-spin-orbit limit is the same regardless of the magnetization direction (e.g., same crossings between Fermi surface sheets).

According to the spin-fluctuation model [5, 6], the thermal average $\langle\boldsymbol{\sigma}^a\rangle_T$ as a function of the polar and azimuthal angles θ and ϕ of $\langle\hat{\mathbf{m}}_s\rangle_T$ is given by Eq. (3), with the coefficients c_{lm} therein replaced by

$$c_{lm}(T) = c_{lm}(0)\left[\frac{M(T)}{M(0)}\right]^{l(l+1)/2} . \qquad (5)$$

This "$l(l+1)/2$ power law" model was used to generate the plot in the inset of Fig. 4 in the main text.

In the main text we evaluated at $T=0$ the orientational average $\sigma^a_{\rm av} = \langle\boldsymbol{\sigma}^a\cdot\hat{\mathbf{m}}_s\rangle$ of the anomalous Hall conductivity (AHC) from the data in Fig. 1, finding $\sigma^a_{\rm av} = 226$ S/cm. The value of $\sigma^a_{\rm av}$ can also be obtained from the fitted coefficients in Eq. (3). Expressing $\hat{\mathbf{m}}_s$ in terms of $l=1$ spherical harmonics and invoking the orthonormality condition, one finds

$$\sigma^a_{\rm av} = \frac{c_{10} - 2c_{11}}{2\sqrt{3\pi}} = 221\,\text{S/cm}, \qquad (6)$$

where on the right-hand side we have used the values of c_{10} and c_{11} from the main text. The good agreement between the two values confirms the validity of the phenomenological expansion. Eq. (6) is valid to all orders in the spherical-harmonic expansion. If the nonlinear terms in Eq. (2) are small, $\sigma^a_{\rm av}$ can be estimated from the single-crystal AHC evaluated at $\theta=0$ and $\theta=\pi/2$ only:

$$\sigma^a_{\rm av} \simeq \frac{1}{3}\sigma_m(0) + \frac{2}{3}\sigma_m(\pi/2) = 238\,\text{S/cm}, \qquad (7)$$

where $\sigma_m(\theta) = \boldsymbol{\sigma}^a\cdot\hat{\mathbf{m}}_s$.

COMPUTATIONAL DETAILS

We have carried out fully-relativistic band-strucutre calculations for hcp and fcc Co at the experimental lattice constants of 4.74 and 6.68 bohr respectively, using the `PWSCF` code [7]. The pseudopotential was generated using similar parameters as in Ref. [8]. The plane-wave basis cutoff for the expansion of the valence wave functions was set at 140 Ry, and the PBE generalized-gradient approximation[9] was used for the exchange-correlation functional. The self-consistent ground state was obtained using a $16\times16\times16$ Monkhorst-Pack mesh [10] of k points and a fictitious Fermi smearing [11] of 0.02 Ry for the Brillouin-zone integration. The calculation was initialized with the spin magnetization pointing along a specified direction; when self-consistency was achieved using a convergence threshold of 10^{-8} Ry for the total energy, the final cell-averaged magnetization [12] was found to be parallel to the initial magnetization. Hence it was not necessary to impose an energy-penalty constraint to fix $\mathbf{M}_s$ during the energy minimization.

For each spin magnetization direction we froze the self-consistent potential and performed a non-self-consistent calculation of the lowest 48 (28 for fcc Co) Bloch eigenstates and eigenvalues over a $10\times10\times10$ uniform k point mesh including the Γ point. From these, maximally-localized Wannier functions were then calculated using the method of Refs. [13, 14], as implemented in the `wannier90` code [15, 16]. For both fcc and hcp Co we chose 18 WFs per atom, covering the s, p, and d characters and both spins. During the disentanglement step used to select the Wannier subspace [14] the upper limit of the "outer energy window" was set at 41.4 eV above the Fermi level. All states up to 11.4 eV above the Fermi level were kept in the subspace by setting the "inner energy window" accordingly. The maximally-localized Wannier functions spanning the resulting subspace were then calculated by minimizing the spread functional [13]. The functional minimization procedures carried in both steps (subspace selection and localization) were initialized by projecting onto trial orbitals of the same type as used in Ref. [17] for bcc Fe: three t_{2g} d-like orbitals and six sp^3d^2 hybrids per spin channel and per atom.

The AHC was calculated using Eq. (1) of the main text. The k space integral of the Berry curvature was carried out using a Wannier-interpolation scheme [17] to sample efficiently the Brillouin zone over a $125\times125\times125$ uniform k-point mesh ($200\times200\times200$ for fcc Co), with a $5\times5\times5$ adaptively refined mesh around the points where the magnitude of the Berry curvature exceeded 10 Å^2. The magnetic circular dichroism (MCD) spectrum was computed in a similar manner [18] on the same interpolation mesh used for calculating the AHC.

INVERSION OF THE RESISTIVITY TENSOR

The comparison between the calculated Hall conductivities and the measured Hall resistivities requires inverting the resistivity tensor. For $\theta=0$ and $\theta=\pi/2$ the vectors $\boldsymbol{\sigma}^a$ and $\mathbf{M}_s$ are parallel. It then suffices to consider the components of ρ and σ in the orthogonal plane. The 2×2 in-plane resistivity matrix reads

$$\rho=\begin{pmatrix}\rho_{11} & \rho_{12}\\ -\rho_{12} & \rho_{22}\end{pmatrix}, \tag{8}$$

with inverse

$$\sigma=\rho^{-1}=\frac{1}{\rho_{11}\rho_{22}+\rho_{12}^2}\begin{pmatrix}\rho_{22} & -\rho_{12}\\ \rho_{12} & \rho_{11}\end{pmatrix}. \tag{9}$$

For $\theta=0$ this yields the familiar relation between the Hall conductivity $\sigma_z^a=\sigma_{xy}$, the Hall resistivity $\rho^a=\rho_{yx}$, and the longitudinal resistivity ρ_{xx}:

$$\sigma_z^a=-\frac{\rho_{xy}}{\rho_{xx}\rho_{yy}+\rho_{xy}^2}=\frac{\rho_{yx}}{\rho_{xx}^2+\rho_{xy}^2}\simeq\frac{\rho^a(\theta=0)}{\rho_{xx}^2}, \tag{10}$$

where we used $\rho_{xx}=\rho_{yy}$ and $\rho^a\ll\rho_{xx}$. For $\theta=\pi/2$ the in-plane resistivity is anisotropic ($\rho_{yy}\neq\rho_{zz}$) and thus

$$\sigma_x^a\simeq\frac{\rho^a(\theta=\pi/2)}{\rho_{yy}\rho_{zz}}=\frac{\rho^a(\theta=\pi/2)}{\rho_{xx}\rho_{zz}}. \tag{11}$$

Dividing Eq. (10) by Eq. (11) yields the relation

$$\frac{\sigma^a(\theta=0)}{\sigma^a(\theta=\pi/2)}\simeq(\rho_{zz}/\rho_{xx})\frac{\rho^a(\theta=0)}{\rho^a(\theta=\pi/2)} \tag{12}$$

used in the main text.

The anomalous Hall conductivities of single crystals magnetized along the c and a axes can now be calculated from the experimental resistivities. We take the room-temperature anomalous Hall resistivities from Fig. 2 of Ref. [19]: $\rho^a(\theta=0)=2.5\times10^{-8}\,\Omega$ cm and $\rho^a(\theta=\pi/2)=0.853\times10^{-8}\,\Omega$ cm. Since the corresponding longitudinal resistivities are not given in that work, we use the room-temperature values from Ref. [20], $\rho_{zz}=10.280\times10^{-6}\,\Omega$ cm and $\rho_{xx}=5.544\times10^{-6}\,\Omega$ cm. Plugging these numbers into the previous two equations we find the values given in Table I of the main text, $\sigma_z^a(\theta=0)\sim813$ S/cm and $\sigma_x^a(\theta=\pi/2)\sim150$ S/cm. Given the disparate experimental sources used to obtain them, these numbers should be taken as approximate.

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