Anisotropic High-Order Anomalous Hall Effect in Epitaxial Nickel Thin Films

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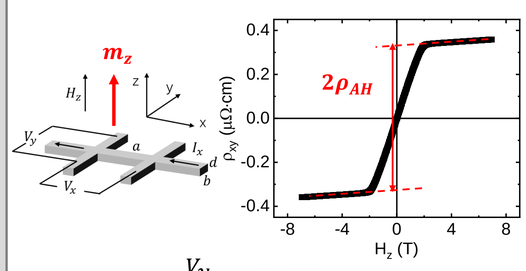
Introduction

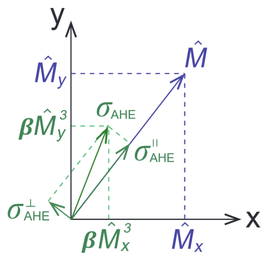
➢ **Anomalous Hall effect (AHE)**



$$\rho_{xy} = \frac{V_y}{I_x} = R_H H_z + \rho_{AH} m_z$$

Ordinary Hall Anomalous Hall

➢ **AHE always proportional to Mz?**



W. Peng *et al.*, arXiv:2402.15741 (2024)

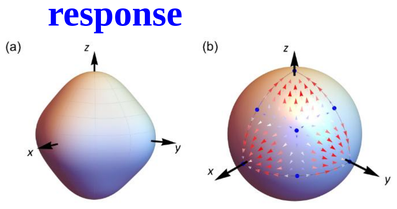
Conventional AHE

$\rho_{xy}^{AHE} \propto m_z$

Beyond Mz-proportional AHE?

$\rho_{xy}^{AHE} = \rho_{xy}^{AHE}(\theta_m, \phi_m)$

➢ **High-order AHE from multipolar response**



$$\sigma_i^H = \alpha \hat{M}_i + \beta \hat{M}_i^3$$

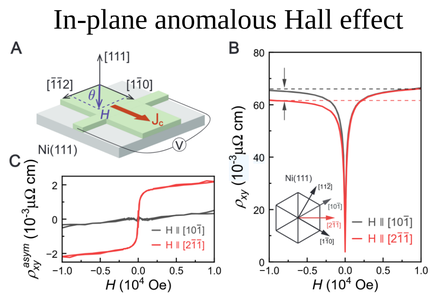
Liu et al., Phys. Rev. X 15, 031006 (2025).

High order Spin-orbit coupling

High order Spin-dependent transport

➢ **In-plane AHE: evidence beyond Mz**

In-plane anomalous Hall effect

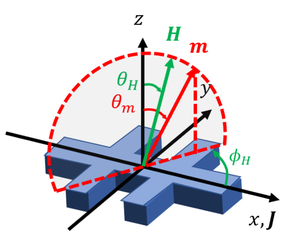


W. Peng *et al.*, arXiv:2402.15741 (2024)

Can high-order AHE harmonics carry in-plane crystalline anisotropy?

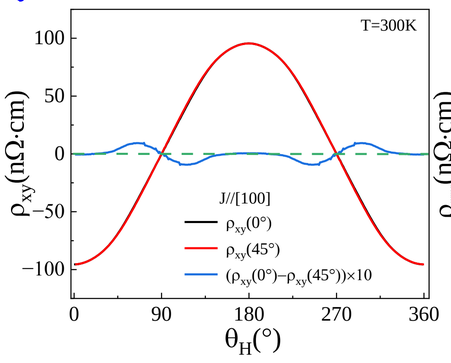
Observation of anisotropic angular-dependent AHE

Measurement geometry:



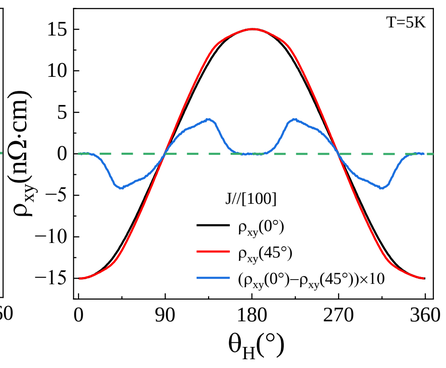
Ni(20 nm)/LAO(001)

T=300 K



$\rho_{xy}^{AHE}(\theta_m, 0°) \neq \rho_{xy}^{AHE}(\theta_m, 45°)$

T=5 K

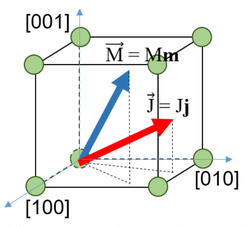


Anisotropy enhanced in 5K

AHE contains an in-plane-sensitive component

Temperature evolution of anisotropic high-order AHE

➢ **Phenomenological model of AHE based on crystal symmetry**



From Ohm's law $E_i = \rho_{ij} J_j$, expansion of ρ_{ij} on **m**:

$$\rho_{ij}(\mathbf{m}) = a_{ij} + a_{kij} m_k + a_{klmij} m_k m_l m_m + \cdots$$

Onsager relation: $\rho_{ij}(\boldsymbol{m}) = \rho_{ji}(-\boldsymbol{m})$

Cubic crystalline symmetry

W. Döring, Ann. Phys. 424, 259 (1938)

$$\rho_{ij}(\theta_m, \phi_m, \phi_j) = \sum_k \rho_{2k+1}^{(0)} \cos\big((2k+1)\theta_m\big) + \sum_k \rho_{2k+1}^{(4)} \cos\big((2k+1)\theta_m\big) \cos\big(4(\phi_m + \phi_j)\big)$$

➢ **High-order expansion of AHE**

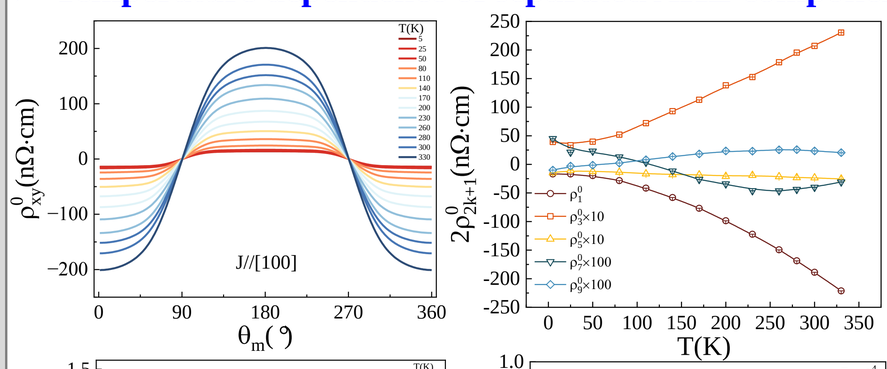
$$\rho_{xy}^{AHE}(\theta_m, \phi_m, \phi_j) = \rho_{xy}^{0}(\theta_m) + \rho_{xy}^{4\phi}(\theta_m, \phi_m, \phi_j)$$

$$\rho_{xy}^{0}(\theta_m) = \sum_k \rho_{2k+1}^{(0)} \cos\big((2k+1)\theta_m\big) \quad \rho_{xy}^{4\phi}(\theta_m, \phi_m, \phi_j) = \sum_k \rho_{2k+1}^{(4)} \cos\big((2k+1)\theta_m\big) \cos\big(4(\phi_m + \phi_j)\big)$$

$$\Delta\rho_{xy}^{0}(\theta_m) = \rho_{xy}^{AHE}(\theta_m, \phi_m = 0°, \phi_j = 0°) + \rho_{xy}^{AHE}(\theta_m, \phi_m = 45°, \phi_j = 0°) = 2\rho_{xy}^{0}(\theta_m)$$

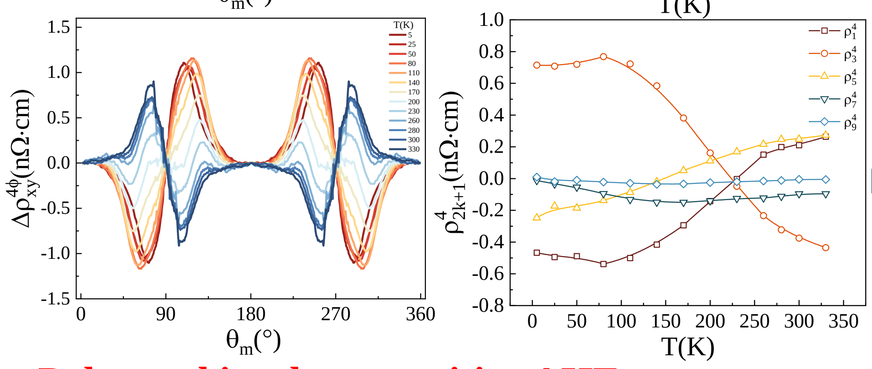
$$\Delta\rho_{xy}^{4\phi}(\theta_m) = \rho_{xy}^{AHE}(\theta_m, \phi_m = 0°, \phi_j = 0°) - \rho_{xy}^{AHE}(\theta_m, \phi_m = 45°, \phi_j = 0°) = 2\rho_{xy}^{4\phi}(\theta_m)$$

➢ **Temperature dependence of separated AHE components**



ρ_{xy}^0: polar component

regular temperature evolution dominated by the first−order term.



$\rho_{xy}^{4\phi}$: Fourfold anisotropic component

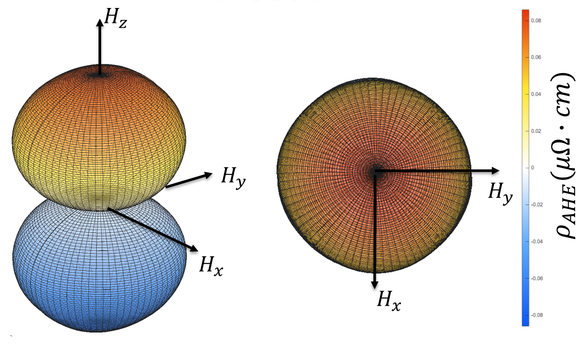
a sign reversal around ~200 K

Polar and in-plane-sensitive AHE components exhibit distinct temperature evolution.

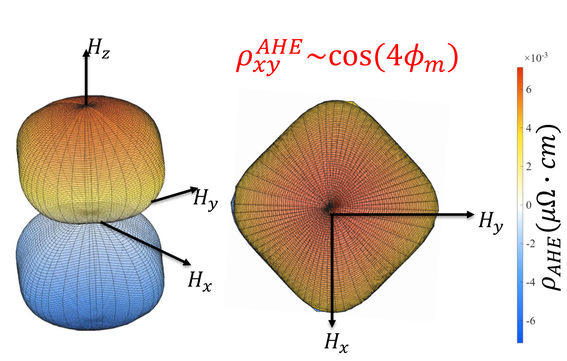
3D angular mapping of anisotropic AHE

➢ **3D full mapping of Ni(001) AHE**

T=300 K



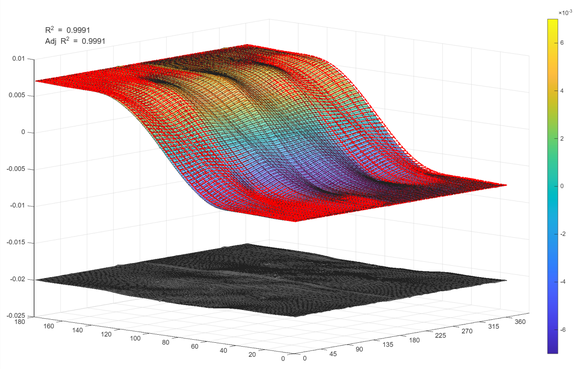
T=5 K



300K:Weak anisotropic

5K:Fourfold in-plane Symmetry

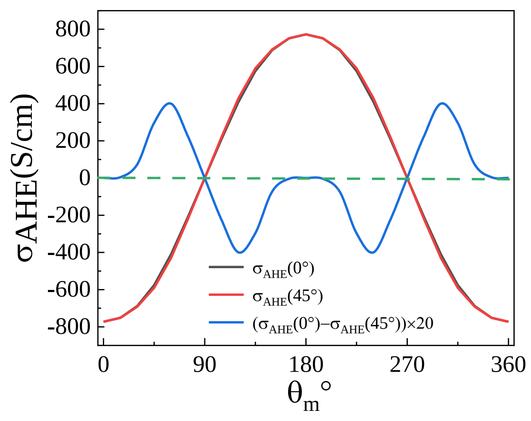
➢ **Global fitting by symmetry model**



Symmetry-model fitting formula

$$\rho_{xy}^{AHE} = \rho_{xy}^{0}(\theta_m) + \rho_{xy}^{4\phi}(\theta_m, \phi_m, \phi_j)$$

➢ **ϕ_m -dependent AHE confirmed by DFT**

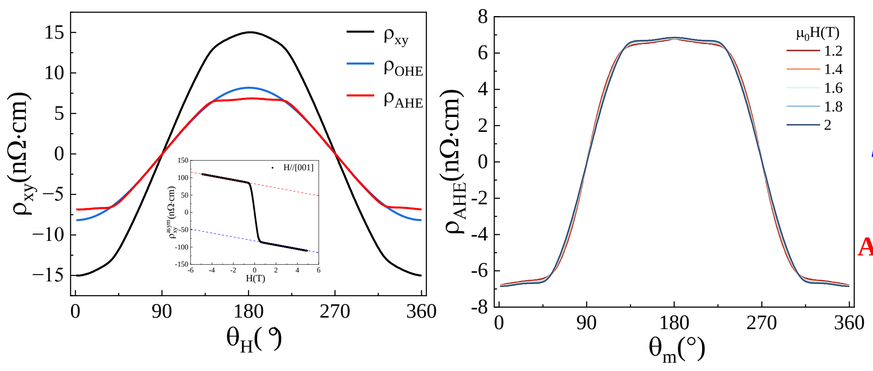


$$\sigma_{xy}^{AHE}(0°) \neq \sigma_{xy}^{AHE}(45°)$$

Full 3D angular mapping visualizes fourfold crystalline anisotropy in high-order AHE.

Extraction of intrinsic AHE and high-order harmonics

➢ **Ordinary Hall Subtraction**



$$\rho_{xy}^{raw} = R_0 B_z + \rho_{xy}^{AHE}$$

$$\rho_{xy}^{AHE} = \rho_{xy}^{raw} - R_0 B \cos(\theta_H)$$

Muti field verification

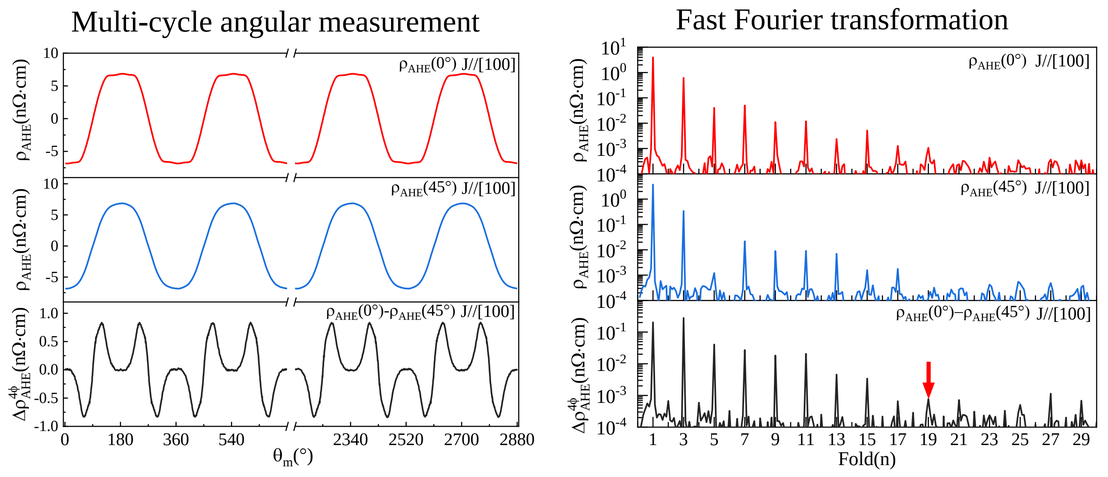
AHE curves nearly collapse

➢ **Fourier verification of high-order AHE**

Up to 19-fold AHE

Multi-cycle angular measurement

Fast Fourier transformation



Both polar and anisotropic AHE host high-order harmonics.

Beyond the conventional Mz picture.

OHE-subtracted AHE reveals robust odd-order harmonics up to 19 fold beyond the conventional Mz picture.

Summary

- **High-order AHE harmonics up to 19th order in Ni(001)**
- **In-plane-sensitive AHE beyond the conventional Mz picture**
- **Fourfold anisotropy visualized by full 3D angular mapping**
- **Distinct temperature evolution of two AHE components**