# Robust Zero-Field Skyrmion Formation in FeGe Epitaxial Thin Films

Fengyuan Yang

The Ohio State University

Acknowledgements:

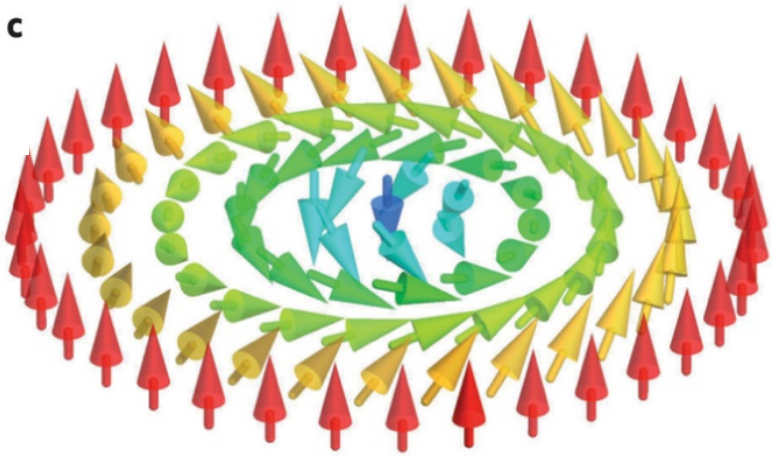
James Gallagher, Keng-Yuan Meng, Jack Brangham, Hailong Wang, Bryan Esser, Vidya Praveen Bhallamudi, David McComb, P. Chris Hammel

U.S. Department of Energy (DOE), Office of Science, Grant No. DE-SC0001304
Center for Emergent Materials, NSF-funded MRSEC, Grant No. DMR-1420451

# Outline

- Magnetic skyrmions and topological Hall effect
- Growth of FeGe thin films by UHV off-axis sputtering
- Structural properties of FeGe films
  - X-ray diffraction
  - Scanning transmission electron microscopy (STEM)
- Hall effect measurement of FeGe films
  - Extraction of topological Hall effect
- Robust skyrmion phase
  - Highest topological Hall resistivity observed to date
  - High remanent topological Hall resistivity at H = 0
- Summary

# Magnetic Skyrmions




3 – 230 nm

Pfleiderer, *Nat. Phys*. 7, 673 (2011)

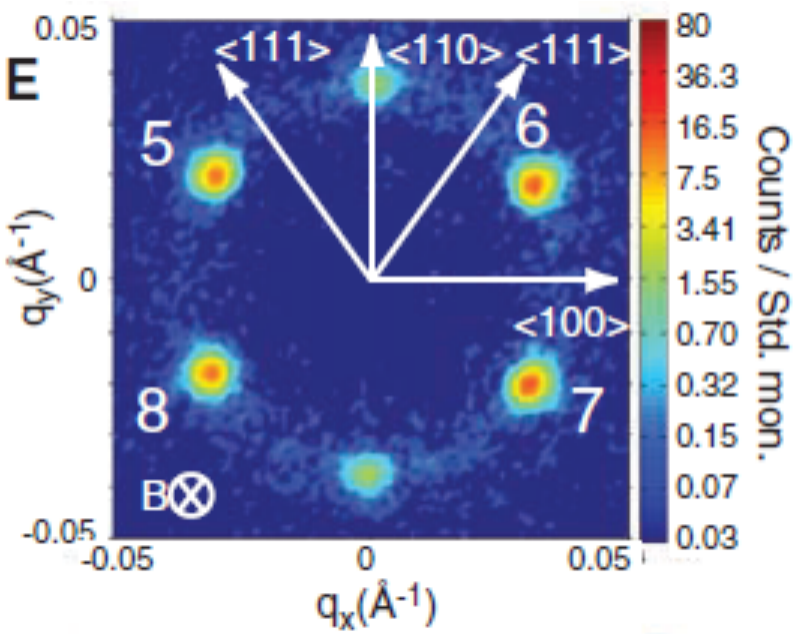
$$H = -J(\boldsymbol{S}_i \cdot \boldsymbol{S}_j) + \boldsymbol{D}_{ij} \cdot (\boldsymbol{S}_i \times \boldsymbol{S}_j)$$

↑↑ Heisenberg Exchange

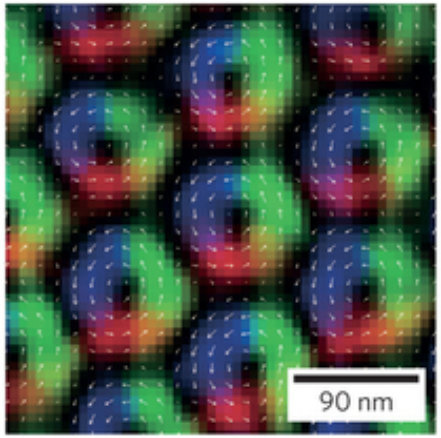
↑→ Dzyaloshinskii-Moriya Interaction (DMI)

# Magnetic Skyrmions

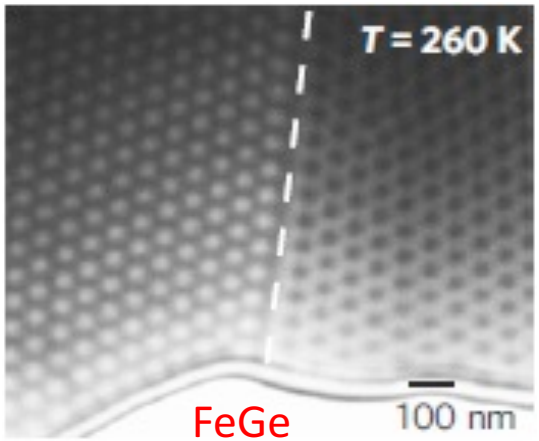
MnSi




Mühlbauer, et al. *Science* 323, 915 (2009)

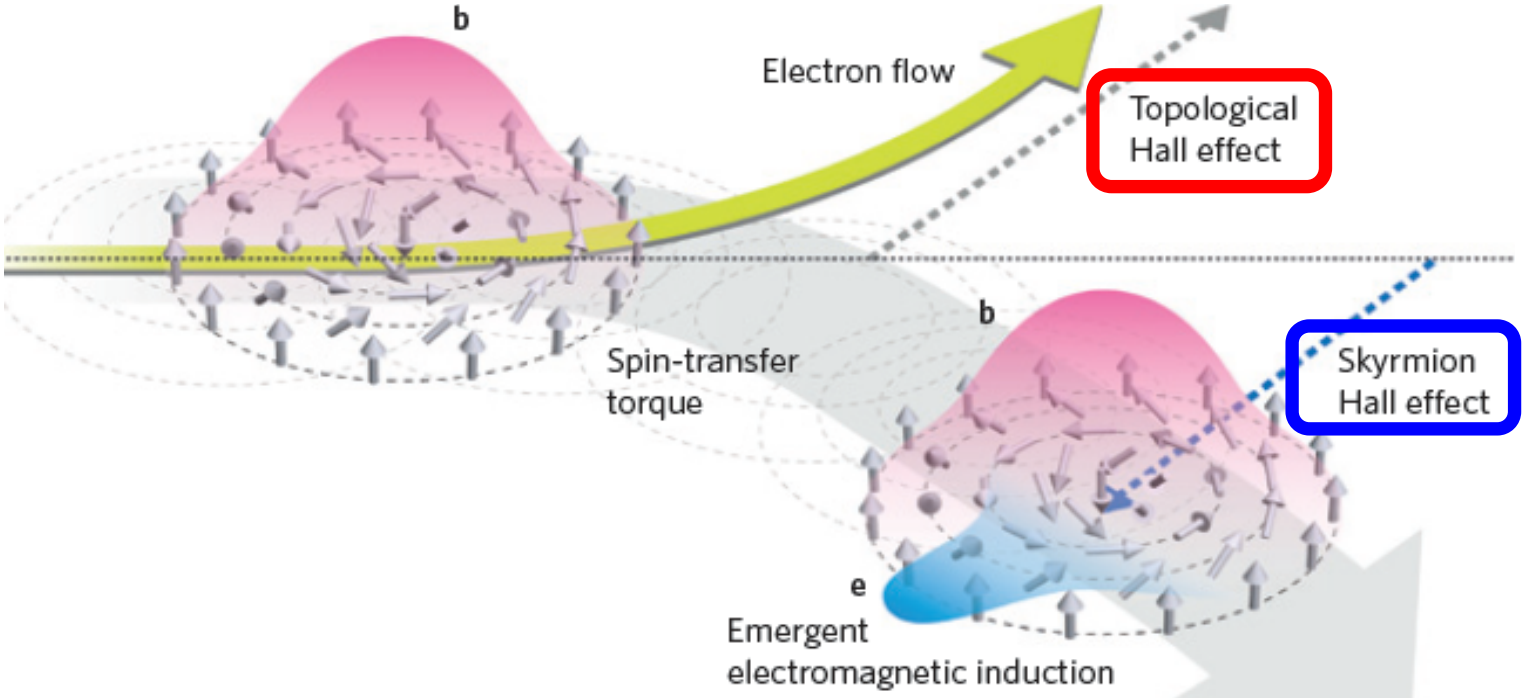



Fert, et al. *Nat. Nano*. 8, 152 (2013)




Yu, et al. *Nat. Mater.* 10, 106 (2011)

# Current-driven skyrmion motion




- Skyrmions can be manipulated by very low electric current
- Potential for low energy memory/logic application

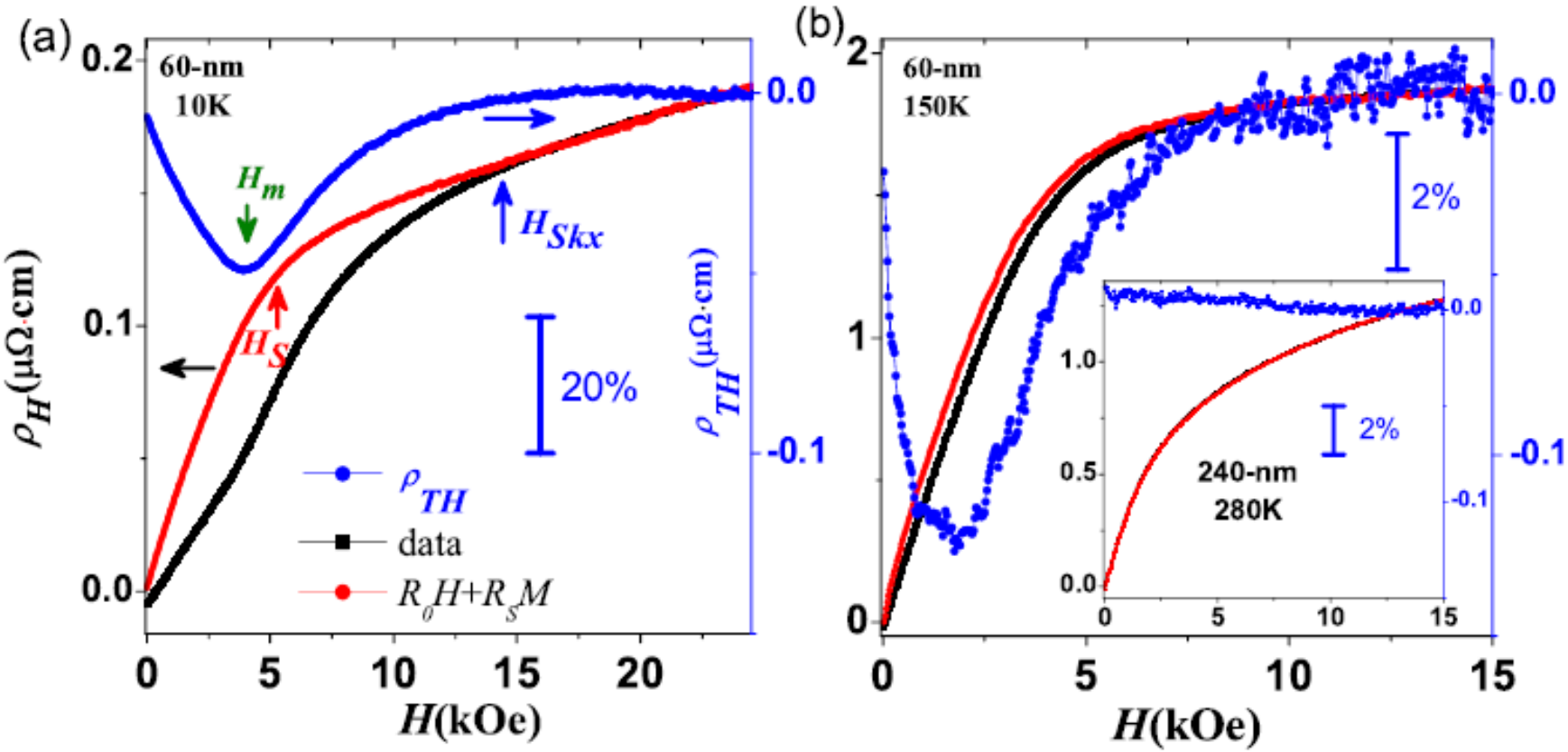
Nagaosa & Tokura, *Nat. Nano.* 8, 899 (2013)

# Topological Hall effect in skyrmion films

PRL 108, 267201 (2012) PHYSICAL REVIEW LETTERS week ending 29 JUNE 2012

## Extended Skyrmion Phase in Epitaxial FeGe(111) Thin Films

S. X. Huang* and C. L. Chien†




Huang & Chien, *Phys. Rev. Lett*. 108, 267201 (2012)

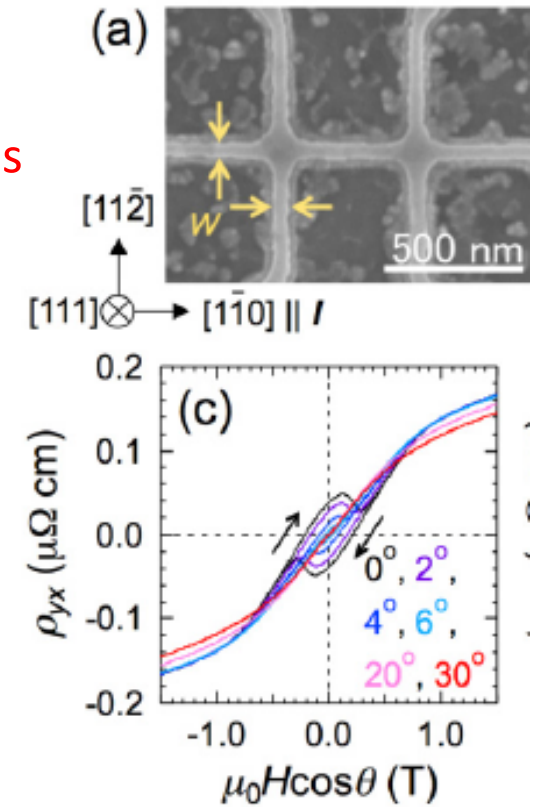
# Topological Hall effect in skyrmion films

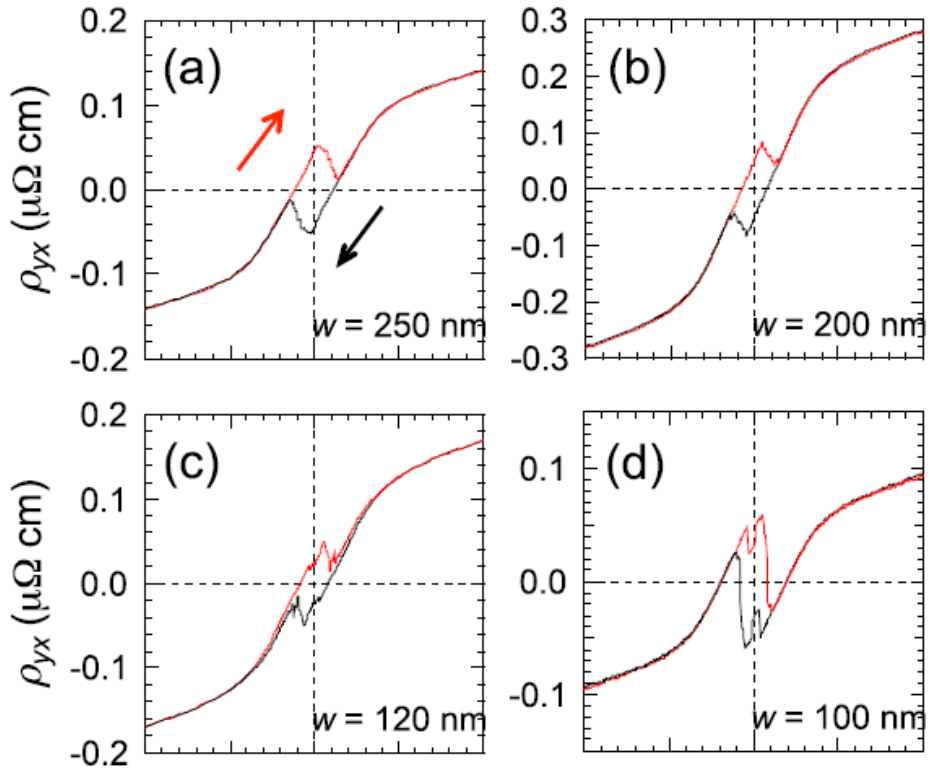
PHYSICAL REVIEW B **91**, 041122(R) (2015)

## Discretized topological Hall effect emerging from skyrmions in constricted geometry

N. Kanazawa,[1] M. Kubota,[2,3,*] A. Tsukazaki,[4] Y. Kozuka,[1] K. S. Takahashi,[2] M. Kawasaki,[1,2]
M. Ichikawa,[1] F. Kagawa,[2] and Y. Tokura[1,2]

FeGe
Epi-films



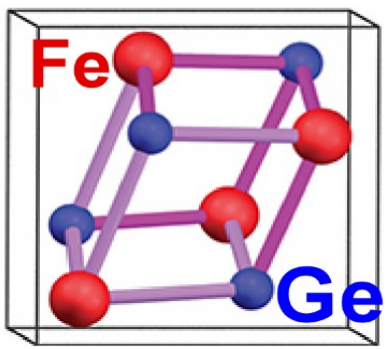




N. Kanazawa, et al. *Phys. Rev. B* 91, 041122(R) (2015)

# B20 skyrmion materials

Technologically important if B20-phase skyrmions exist

- At zero magnetic field
- At room temperature.




B20 cubic lattice
noncentrosymmetric

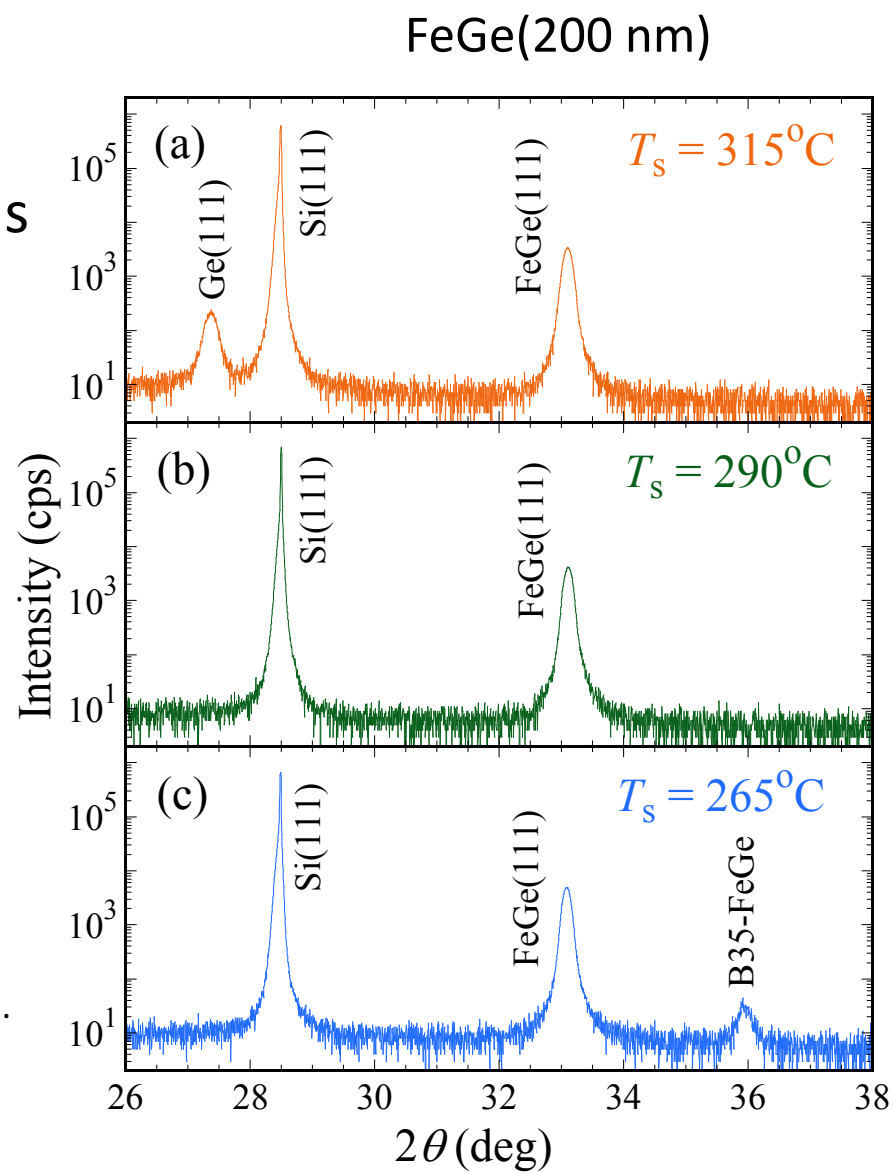
| Material | | $T_N$ (K) | λ (nm) |
|---|---|---|---|
| MnSi | Bulk | 30 | 18 |
| | Epitaxial thin film | 45 | 8.5 |
| $Mn_{1-x}Fe_xSi$ | $x = 0.06$ | 16.5 | 12.5 |
| | $x = 0.08$ | 10.6 | 11 |
| | $x = 0.10$ | 6.8 | 10 |
| $Fe_{1-x}Co_xSi$ | $x = 0.10$ | 11 | 43 |
| | $x = 0.5$ | 36 | 90 |
| | $x = 0.6$ | 24 | 174 |
| | $x = 0.7$ | 7 | 230 |
| MnGe | $T = 20$ K | 170 | 3 |
| | $T = 100$ K | - | 3.4 |
| | $T = 150$ K | - | 5.5 |
| $Mn_{1-x}Fe_xGe$ | $x = 0.35$ | 150 | 4.7 |
| | $x = 0.5$ | 185 | 14.5 |
| | $x = 0.7$ | 210 | 77 |
| | $x = 0.84$ | 220 | 220 |
| FeGe | Bulk | 278 | 70 |
| $Cu_2OSeO_3$ | Bulk | 59 | 62 |
| | Thinned plate | - | 50 |

Nagaosa & Tokura, *Nat. Nano.* 8, 899 (2013)

# X-Ray Diffraction of FeGe films

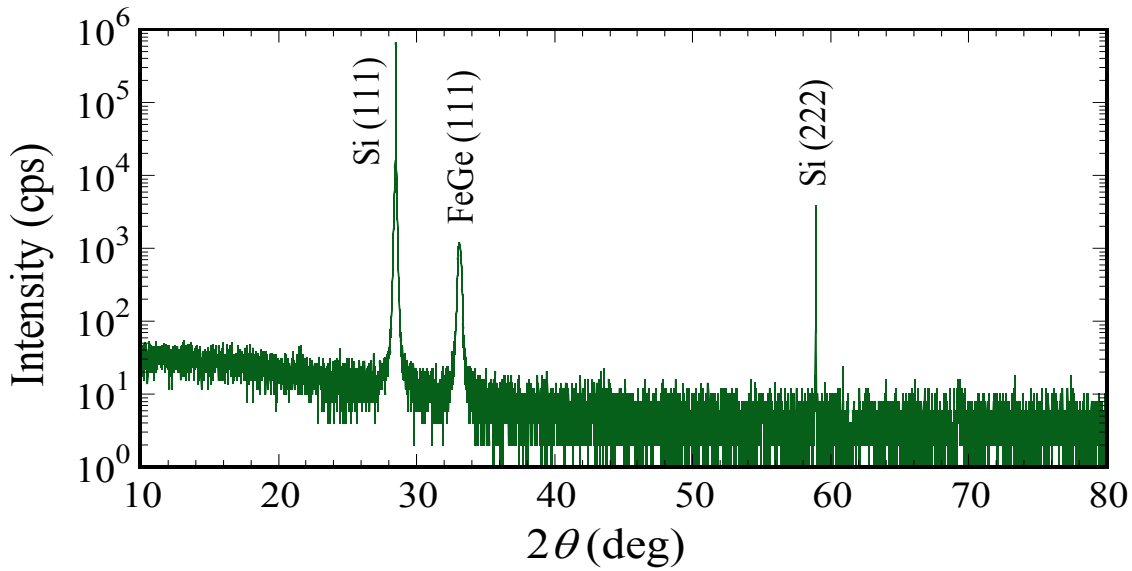
Deposition of FeGe epitaxial films

- UHV off-axis sputtering
- Base pressure: $10^{-11}$ Torr
- Gas: 5 mTorr Argon
- Growth rate: 0.67 nm/min
- FeGe film quality sensitive to substrate temperature

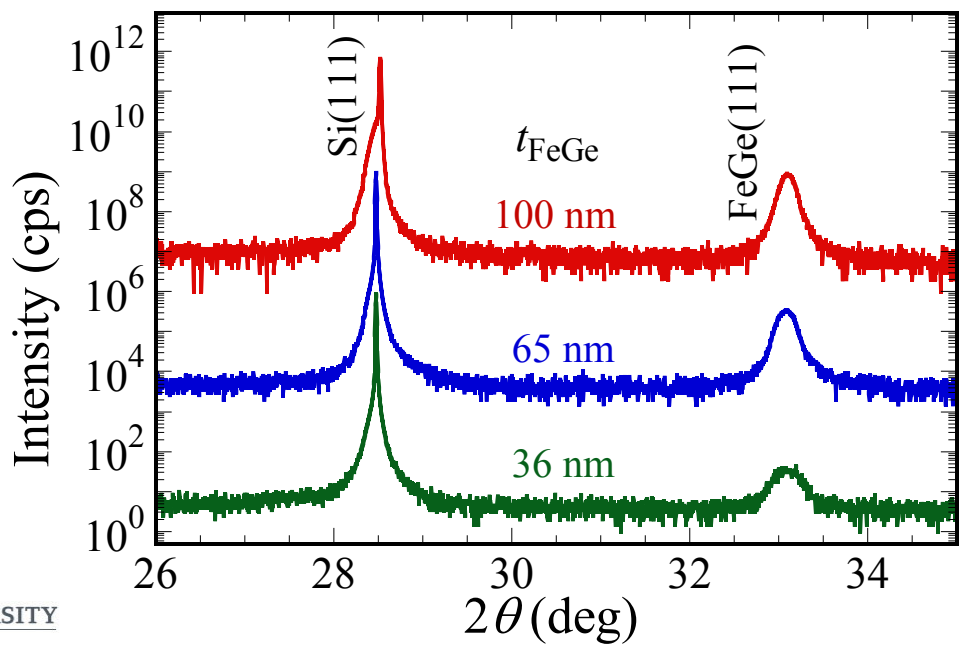



J. C. Gallagher, *et al.* Phys. Rev. Lett. 118, 027201 (2017).

# X-Ray Diffraction of FeGe films

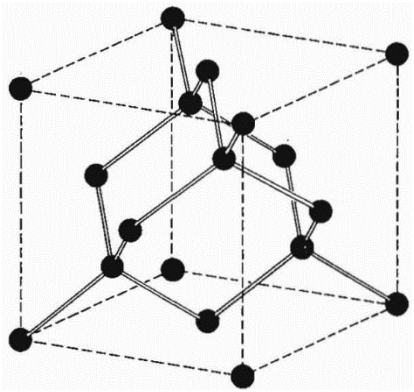


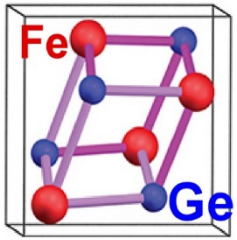

FeGe(200 nm)

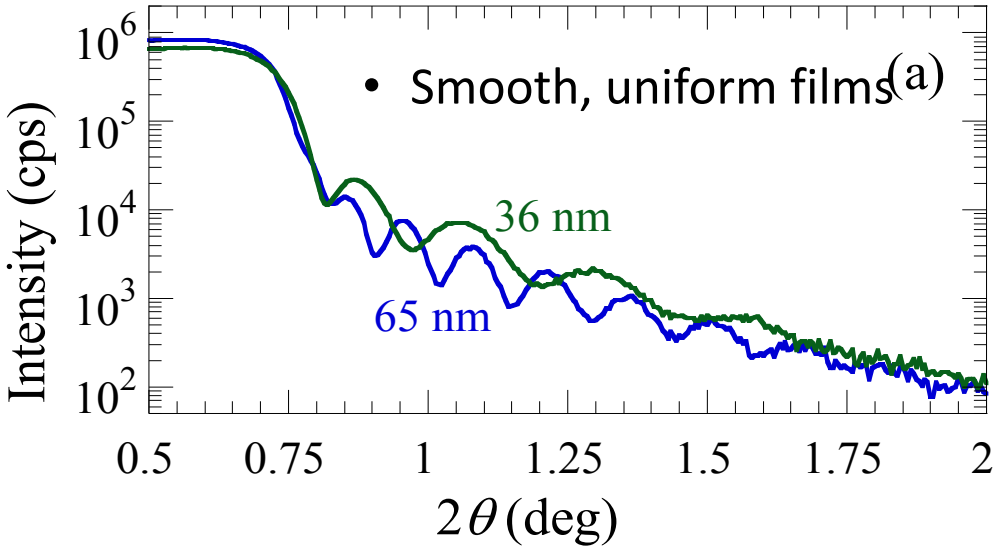


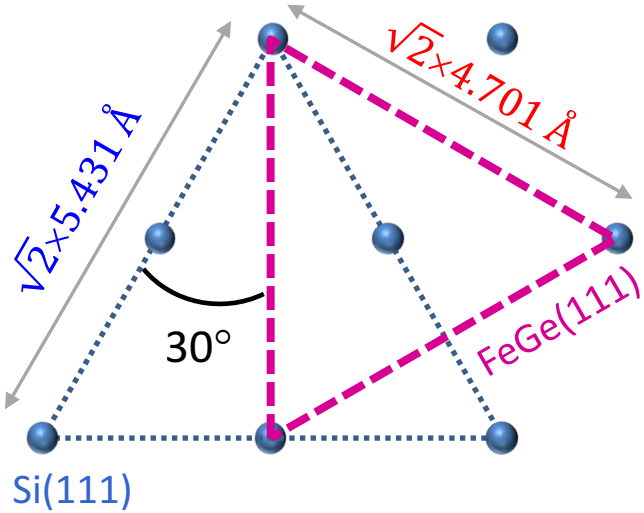

Pure B20 phase

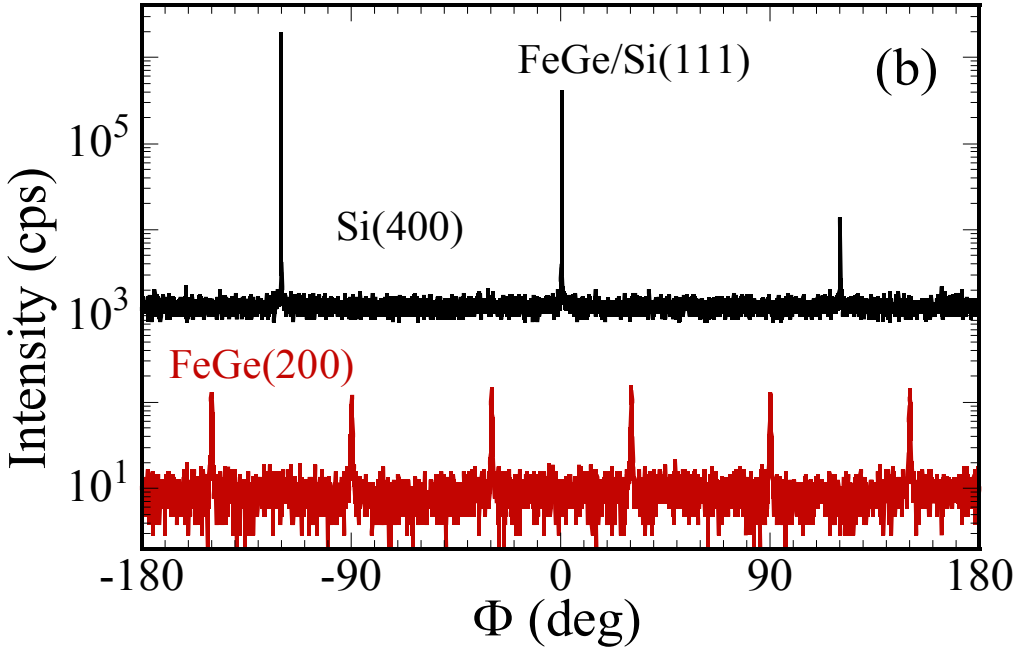
# X-Ray Diffraction of FeGe films





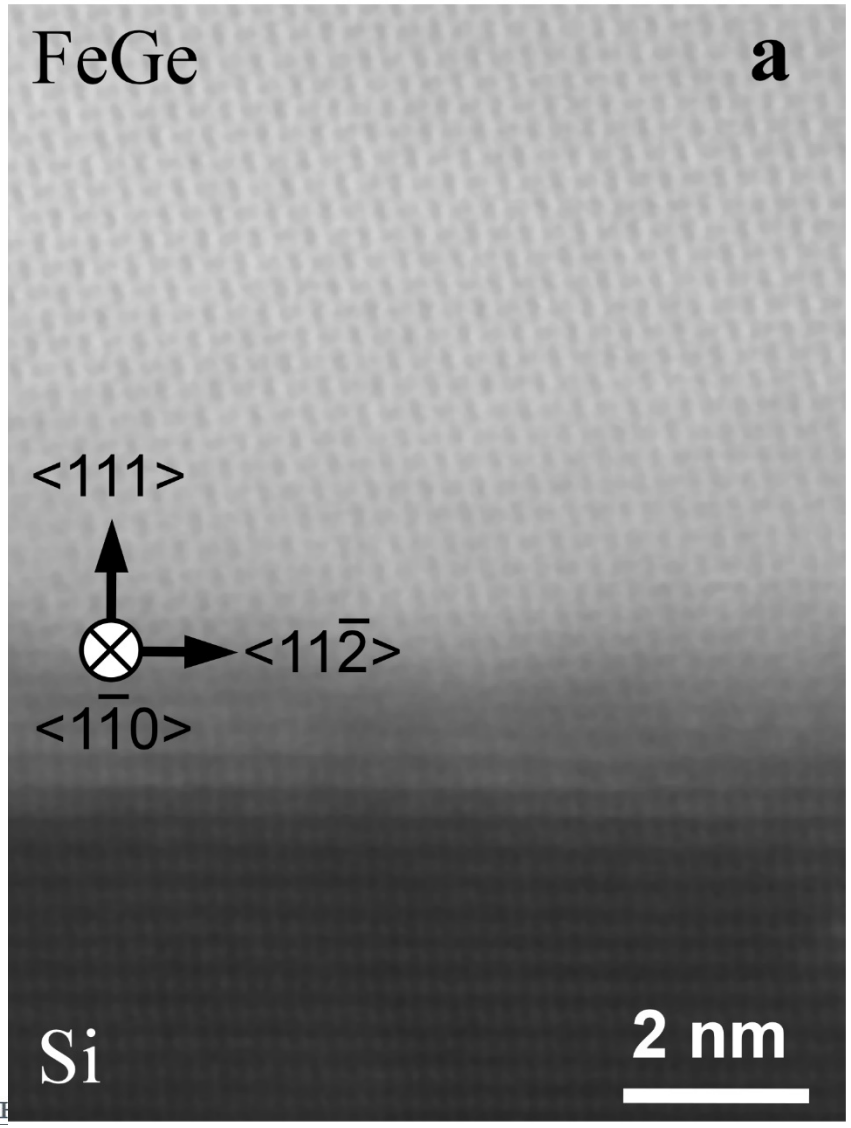



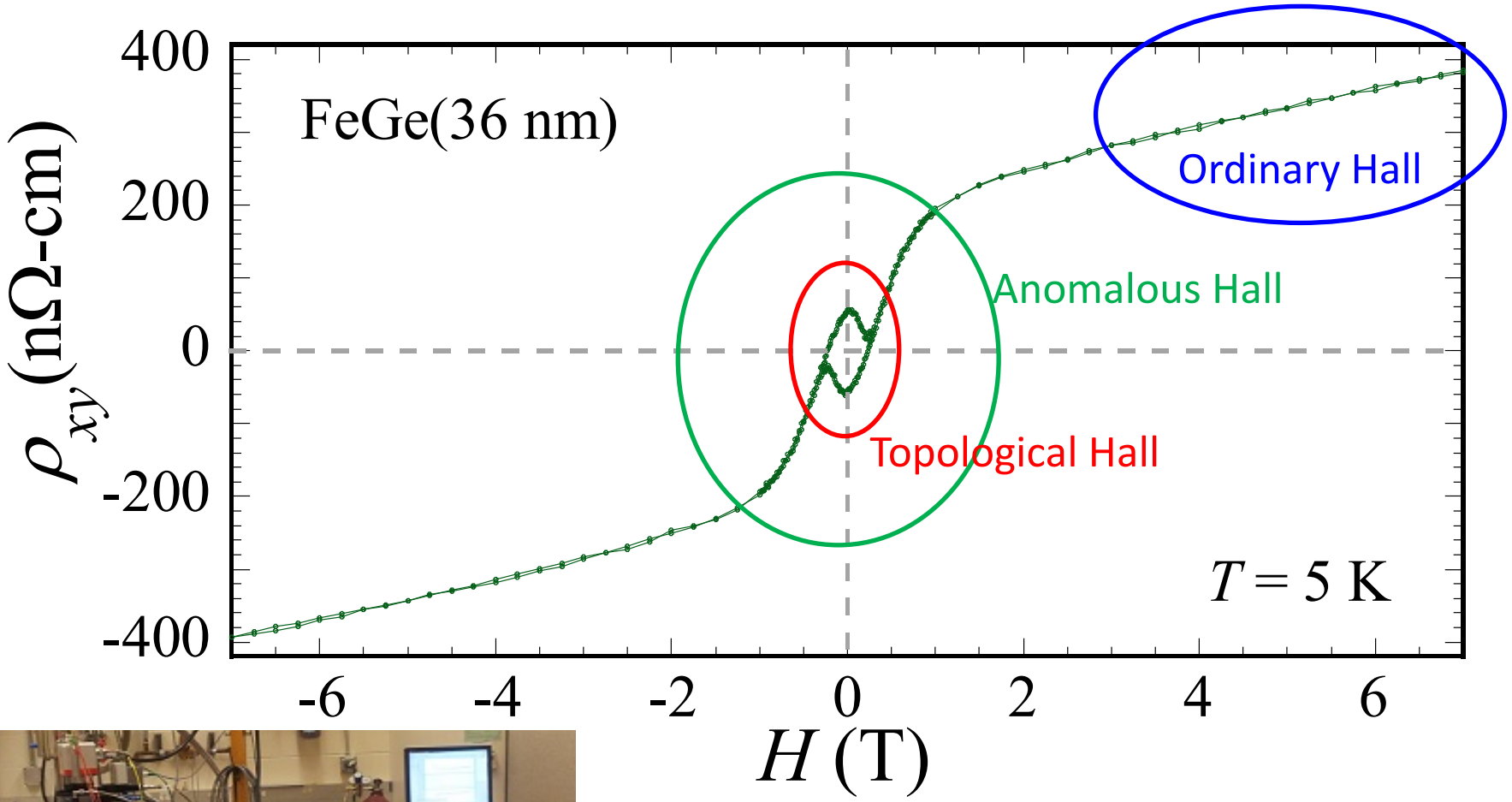




- Fully epitaxial
- 30° rotation

# STEM imaging of FeGe films

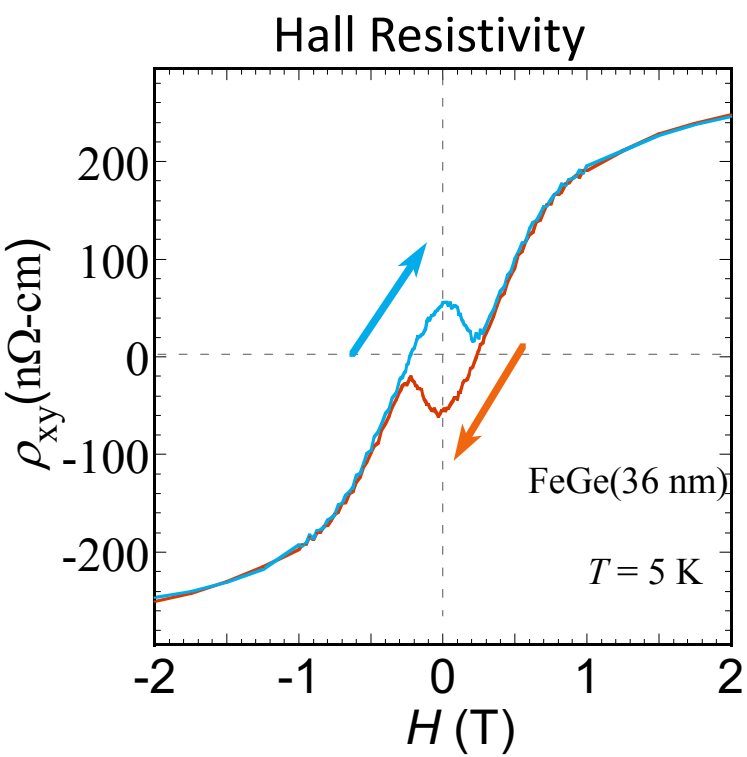


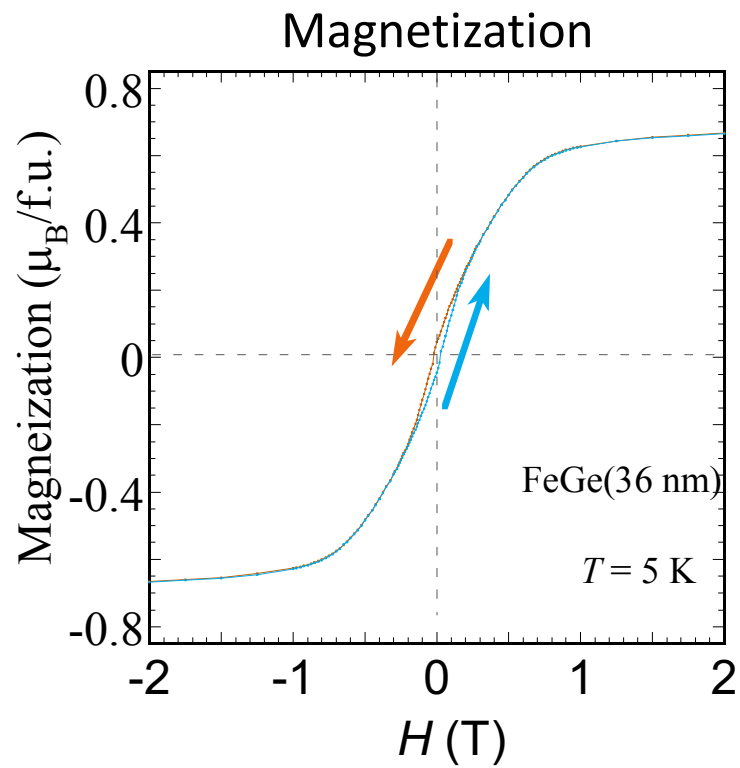
# Hall resistivity in FeGe films



$$\rho_{xy} = R_0 H + R_s M + \rho_{TH}$$

# Hall resistivity in FeGe films

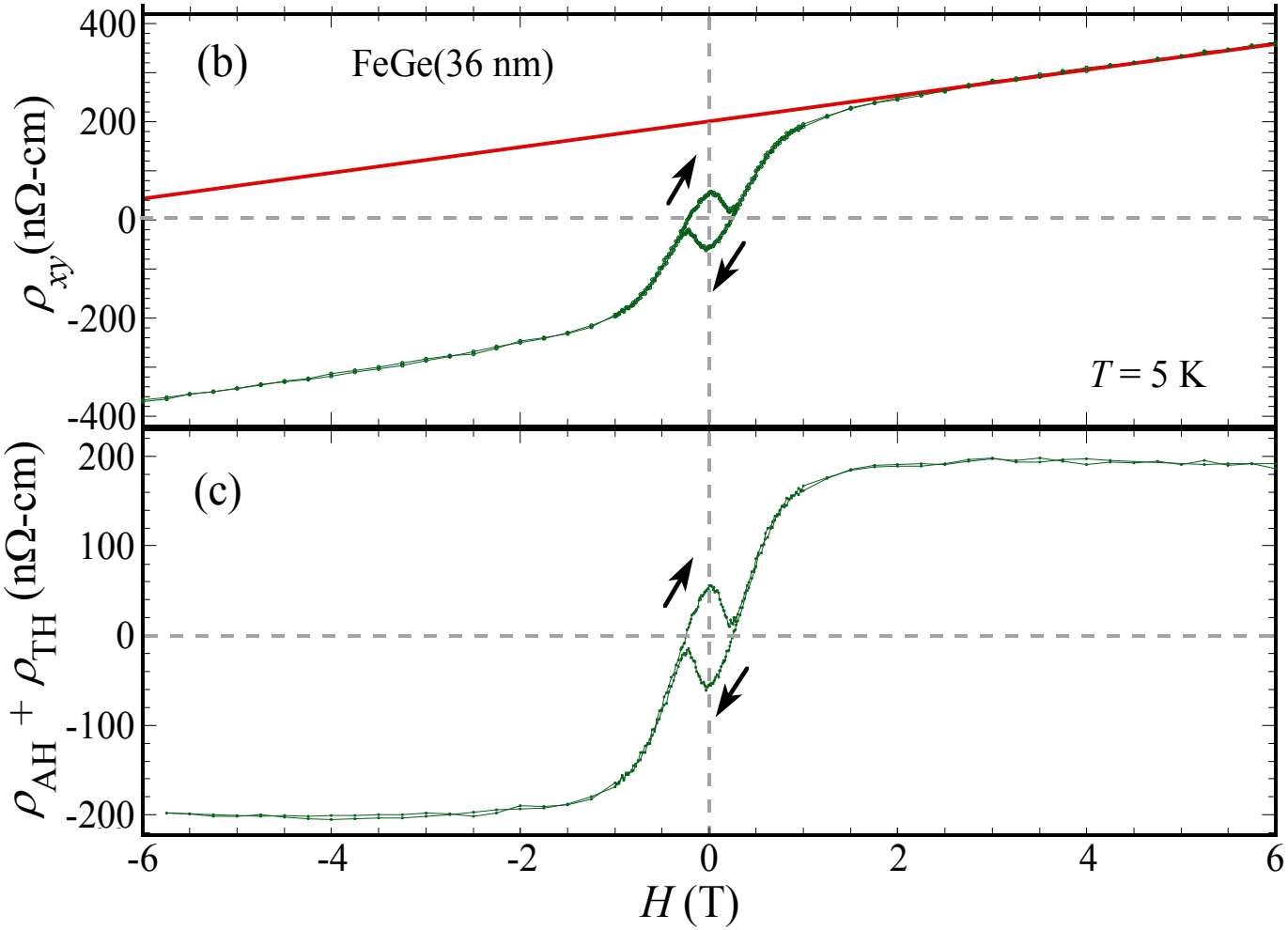



$$\rho_{xy} = R_0 H + R_s M + \rho_{TH}$$

# Extracting topological Hall resistivity

$$\rho_{xy} = R_0 H + R_s M + \rho_{TH}$$



# Extracting topological Hall resistivity

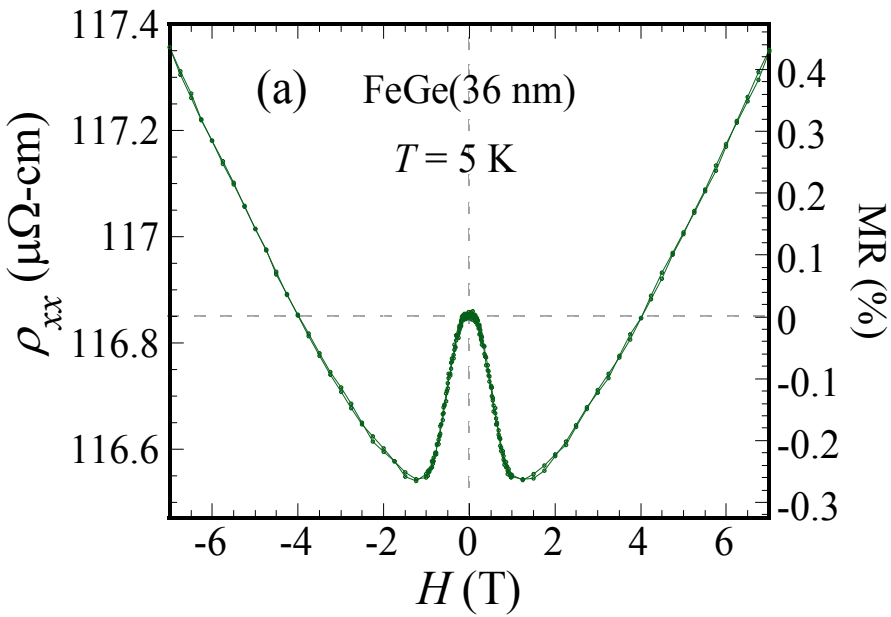
$$\rho_{xy} = R_0 H + R_s M + \rho_{TH}$$

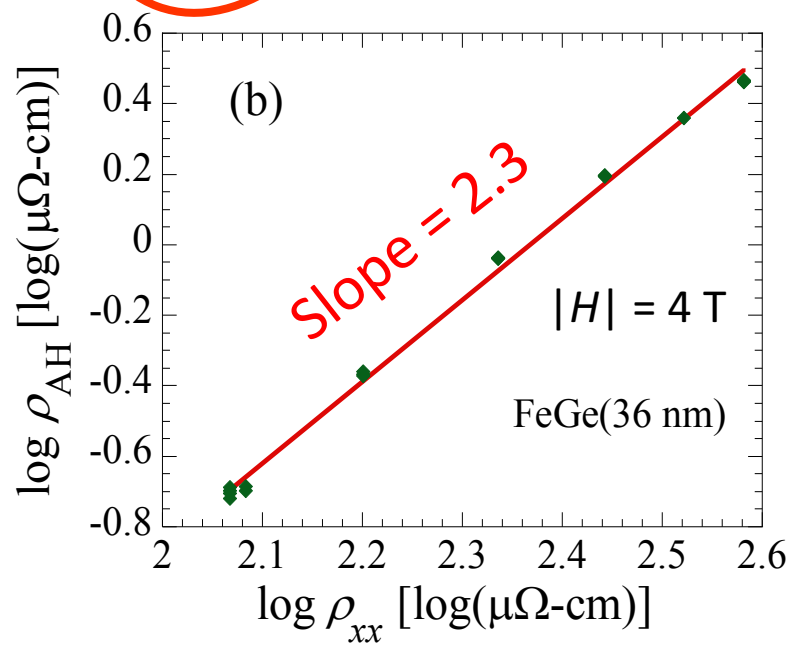
$$R_s M = (b\rho_{xx}^2 + c\rho_{xx})M$$

Scattering independent contribution

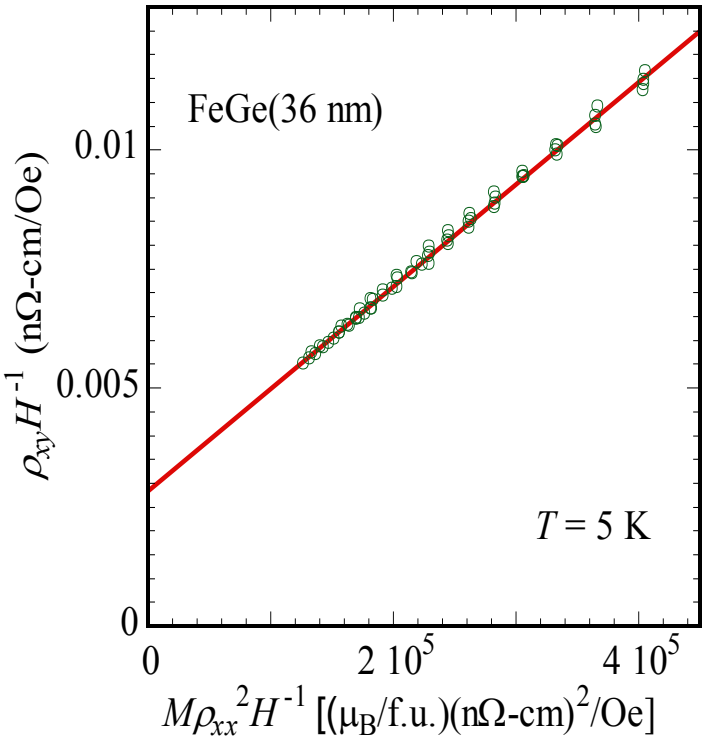
Skew scattering

Small MR

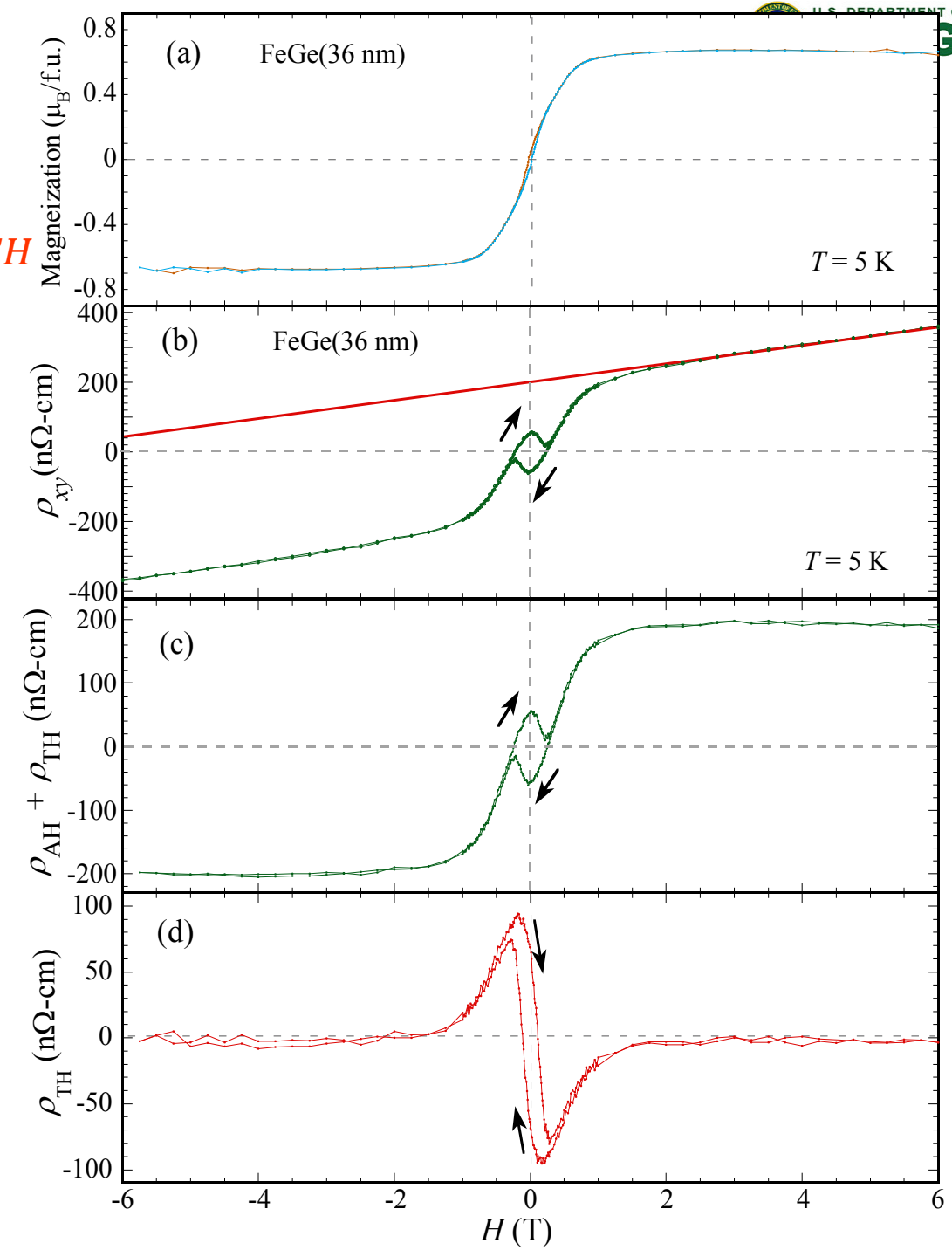


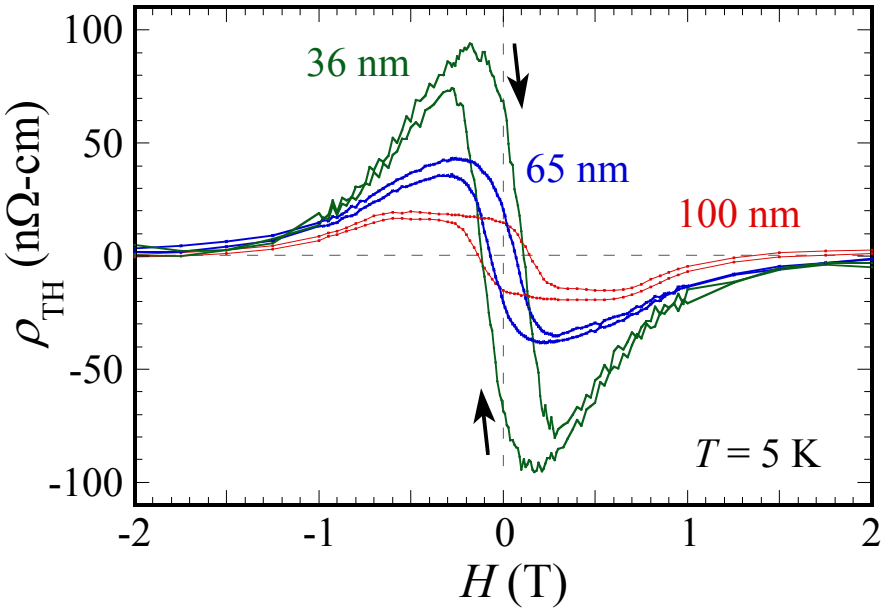
$$\rho_{xy} = R_0 H + b\rho_{xx}^2 M + \rho_{TH}$$

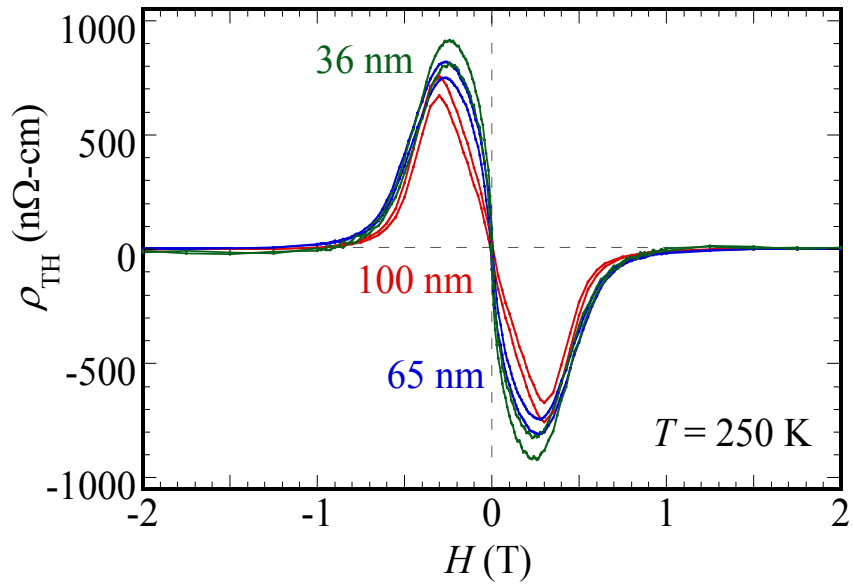



$$\frac{\rho_{xy}}{H} = R_o + b\frac{\rho_{xx}^2 M_s}{H}$$

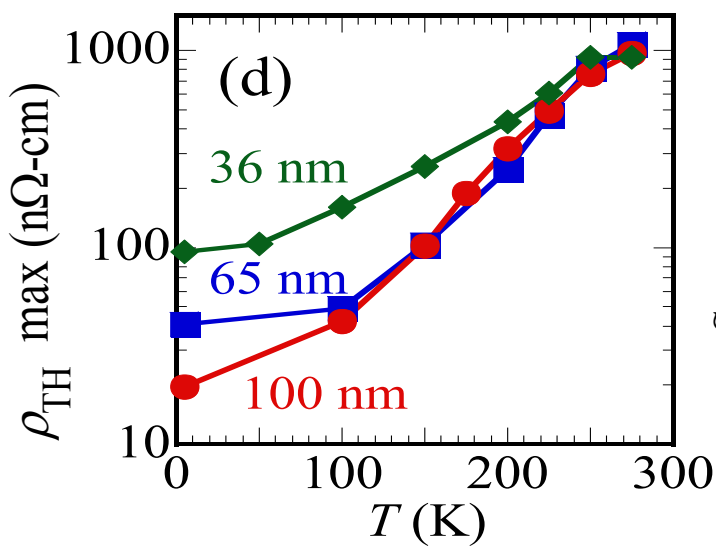


# Extracted Topological Hall Resistivity

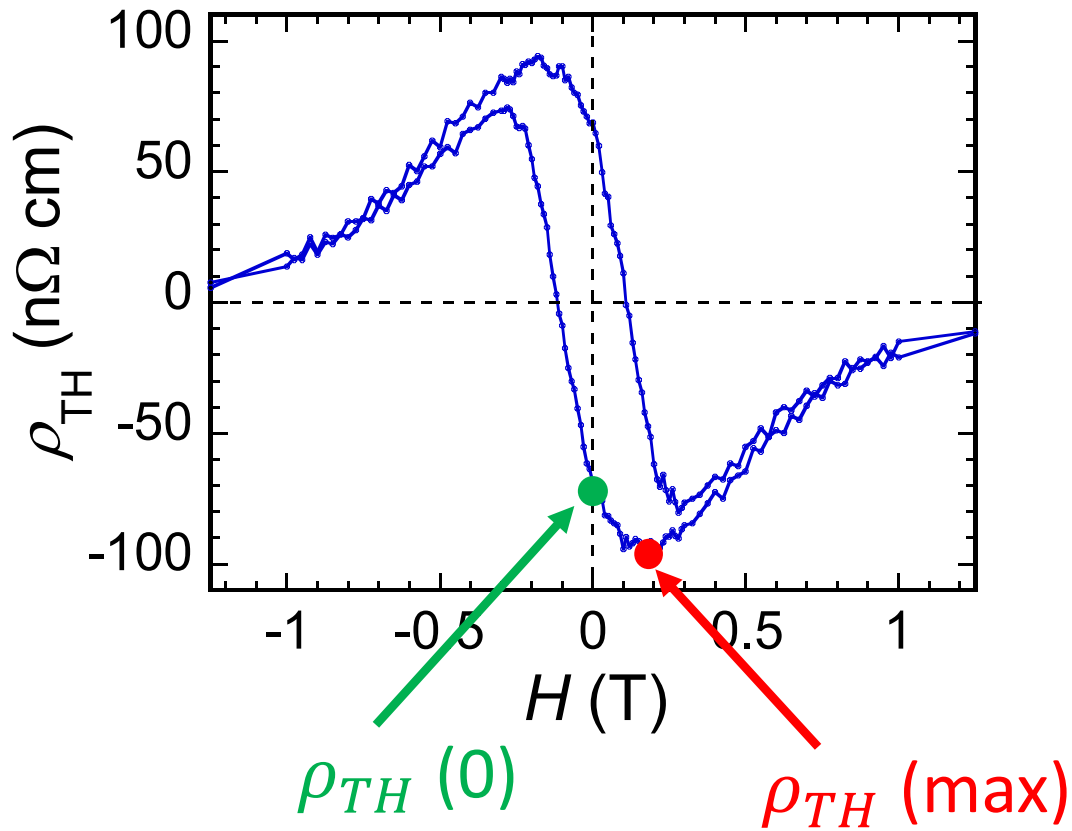


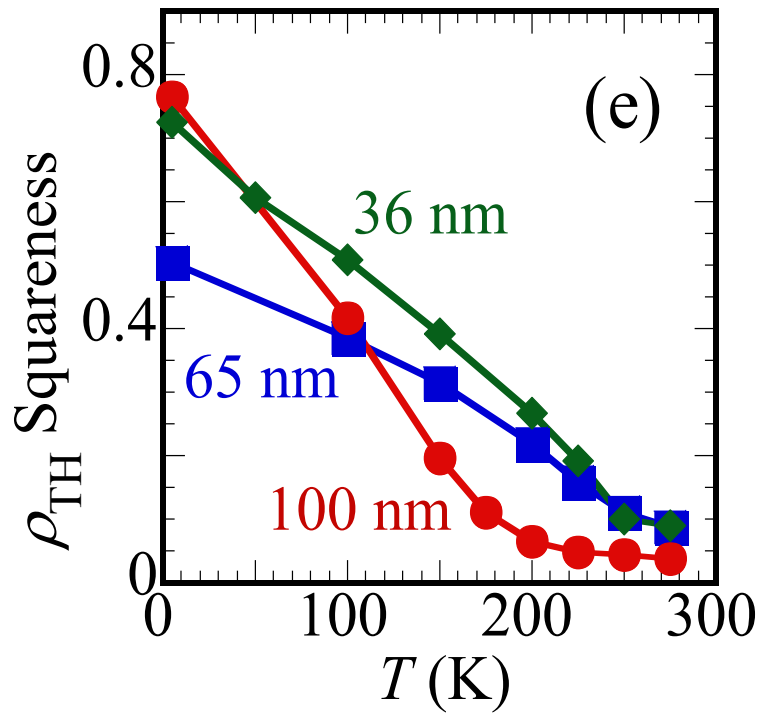

- Large topological Hall resistivity (up to 970 nΩ-cm)
- 6x larger than previous results



# Remanent Topological Hall Resistivity

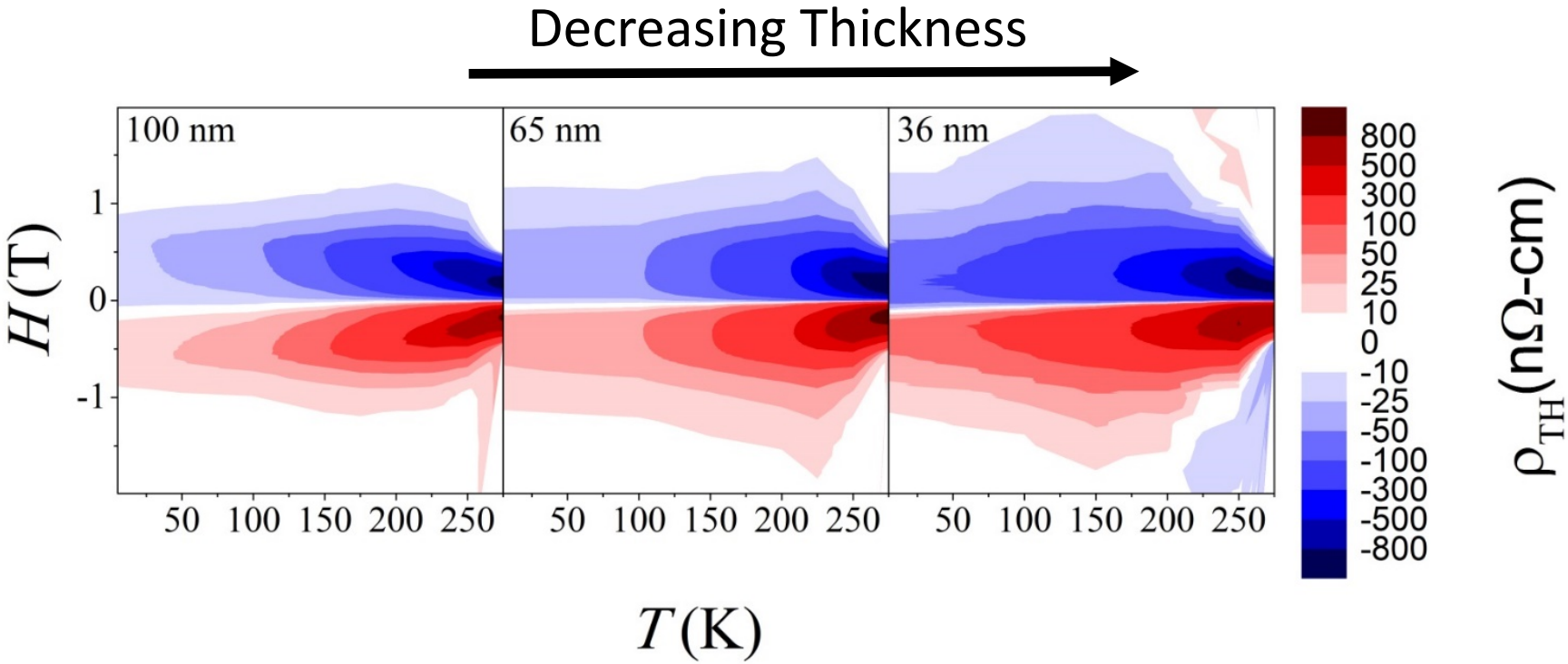



$$Squareness \equiv \rho_{TH}(H=0)/\rho_{TH}(\max)$$



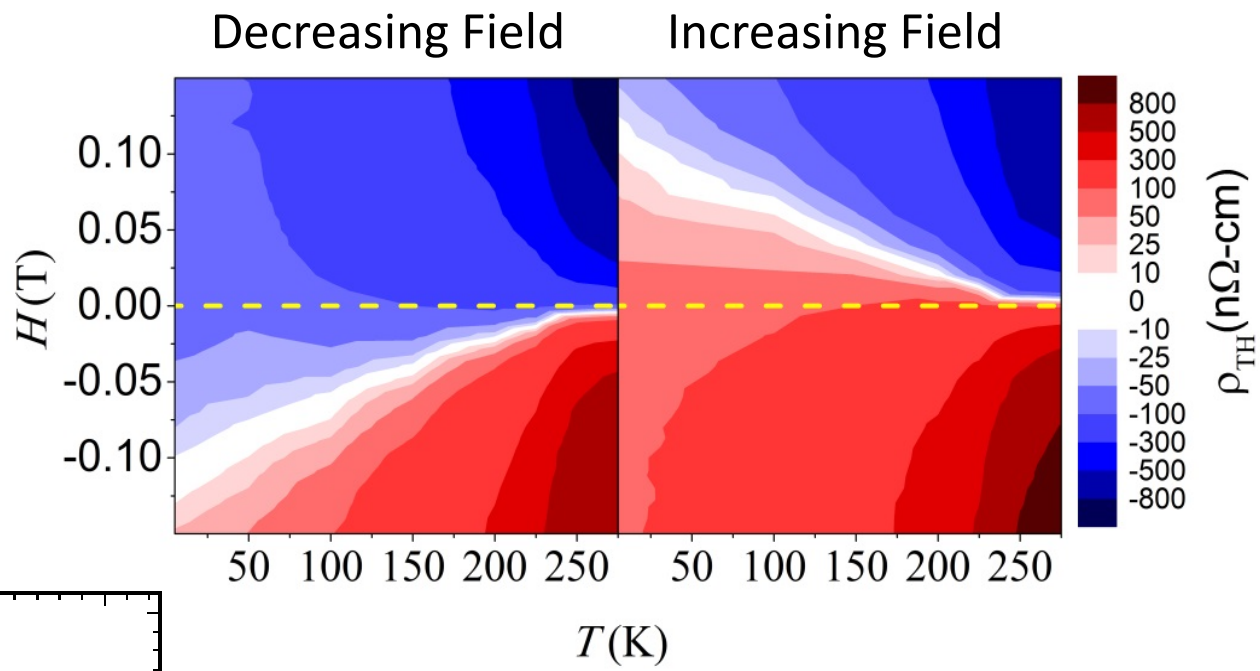

- Robust zero-field Skyrmion phase at all temperatures
- Squareness as high as 0.78

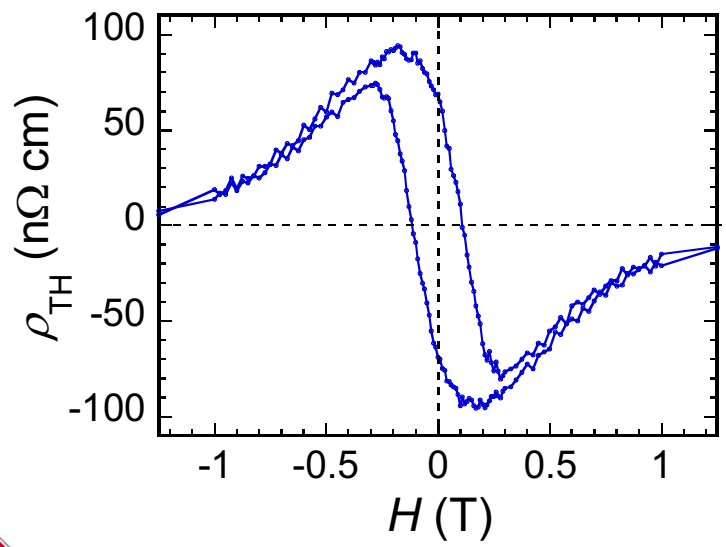
# Topological Hall Effect: variable Temperatures




- Topological Hall effect detected up to $H$ = 2 T
- Enhanced topological Hall effect at lower thicknesses
- Evidence for robust skyrmions at zero field

# Zero-Field Topological Hall Effect




- Sizable zero-field Skyrmion phase at all temperatures

J. C. Gallagher, *et al.* Phys. Rev. Lett. 118, 027201 (2017).

# Interface-driven skyrmions

ARTICLES
PUBLISHED ONLINE: 19 SEPTEMBER 2016 | DOI: 10.1038/NPHYS3883
nature physics

## Direct observation of the skyrmion Hall effect

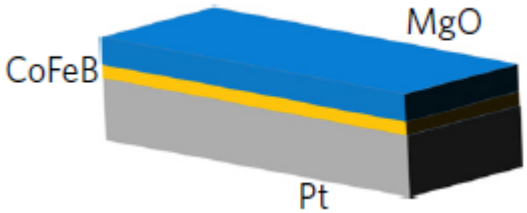
Wanjun Jiang[1,2,3★†], Xichao Zhang[4†], Guoqiang Yu[5], Wei Zhang[1,6], Xiao Wang[7], M. Benjamin Jungfleisch[1], John E. Pearson[1], Xuemei Cheng[7], Olle Heinonen[1,8], Kang L. Wang[5], Yan Zhou[4], Axel Hoffmann[1★] and Suzanne G. E. te Velthuis[1★]

Ta(5 nm)
CoFeB(1.1 nm)
$TaO_x$ (3 nm)

Jiang, *et al. Nature Phys.* **13**, 162-169 (2016).

ARTICLES
PUBLISHED ONLINE: 26 DECEMBER 2016 | DOI: 10.1038/NPHYS4000
nature physics

## Skyrmion Hall effect revealed by direct time-resolved X-ray microscopy




Kai Litzius[1,2,3], Ivan Lemesh[4], Benjamin Krüger[1], Pedram Bassirian[1], Lucas Caretta[4], Kornel Richter[1], Felix Büttner[4], Koji Sato[5], Oleg A. Tretiakov[6,7], Johannes Förster[3], Robert M. Reeve[1], Markus Weigand[3], Iuliia Bykova[3], Hermann Stoll[3], Gisela Schütz[3], Geoffrey S. D. Beach[4★] and Mathias Kläui[1,2★]

Litzius, *et al. Nature Phys.* **13**, 170-176 (2016).

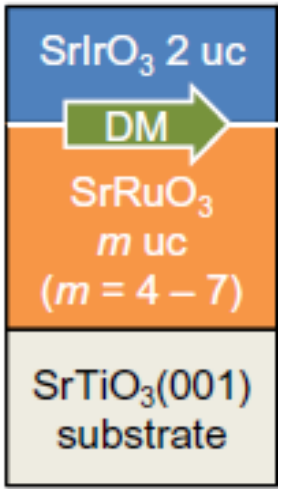
# Skyrmions in oxide bilayers

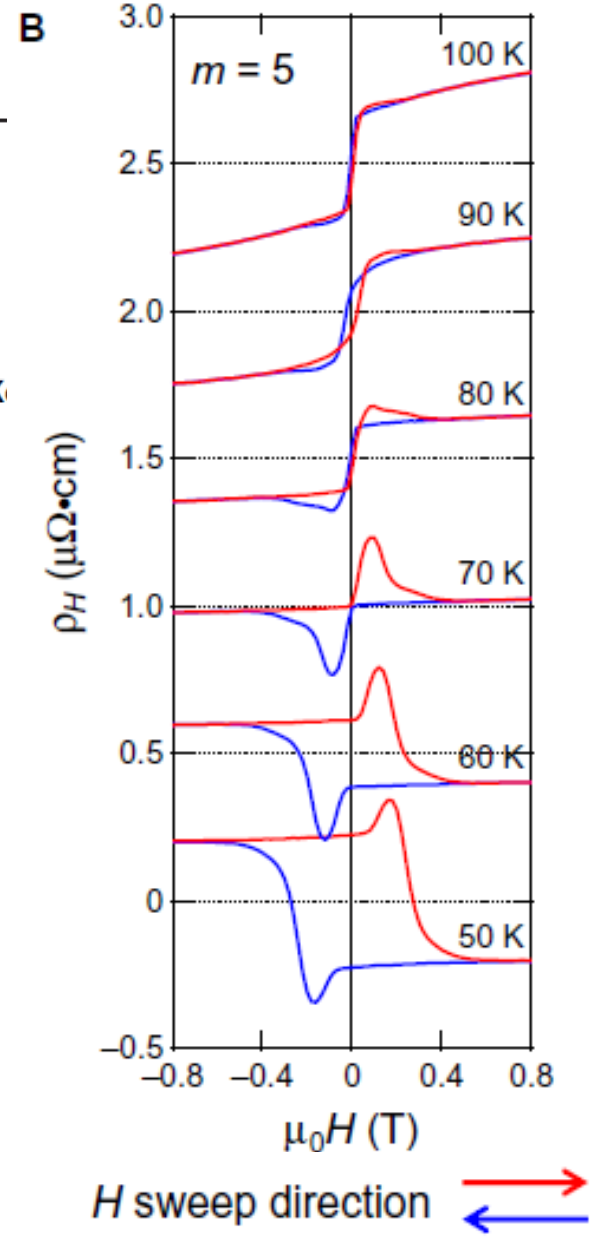
RESEARCH ARTICLE

ELECTROMAGNETISM

## Interface-driven topological Hall effect in $SrRuO_3$-$SrIrO_3$ bilayer

Jobu Matsuno,[1]* Naoki Ogawa,[1] Kenji Yasuda,[2] Fumitaka Kagawa,[1] Wataru K
Yoshinori Tokura,[1,2] Masashi Kawasaki[1,2]




Matruno et al., *Science Advances* 2 : e1600304 (2016)

# Skyrmions in oxide bilayers

RESEARCH ARTICLE

ELECTROMAGNETISM

## Interface-driven topological Hall effect in $SrRuO_3$-$SrIrO_3$ bilayer

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10.1126/sciadv.1600304

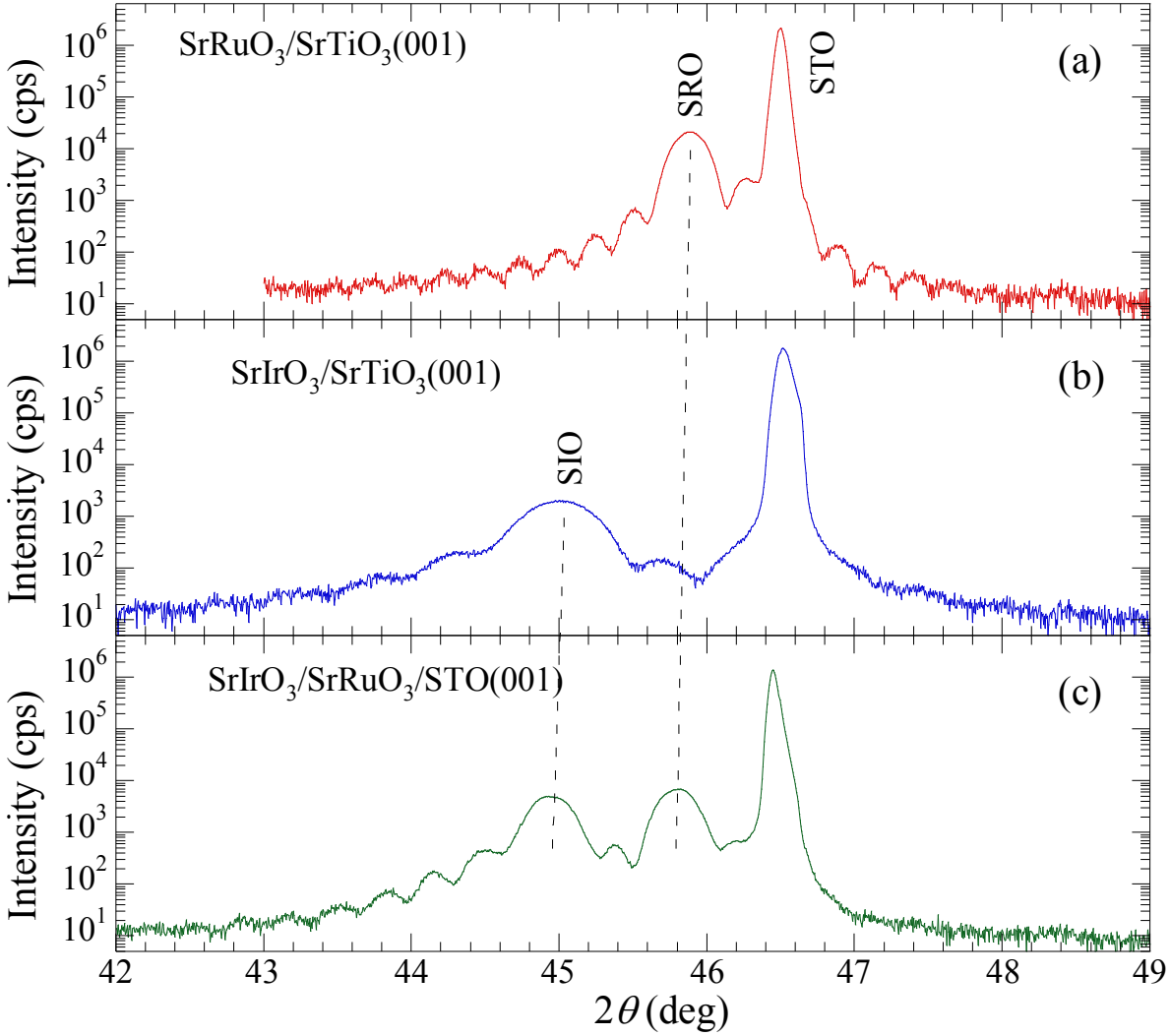
Jobu Matsuno,[1]* Naoki Ogawa,[1] Kenji Yasuda,[2] Fumitaka Kagawa,[1] Wataru Koshibae,[1] Naoto Nagaosa,[1,2] Yoshinori Tokura,[1,2] Masashi Kawasaki[1,2]

"Such interaction is expected to realize a 10-nm-sized magnetic skyrmion."

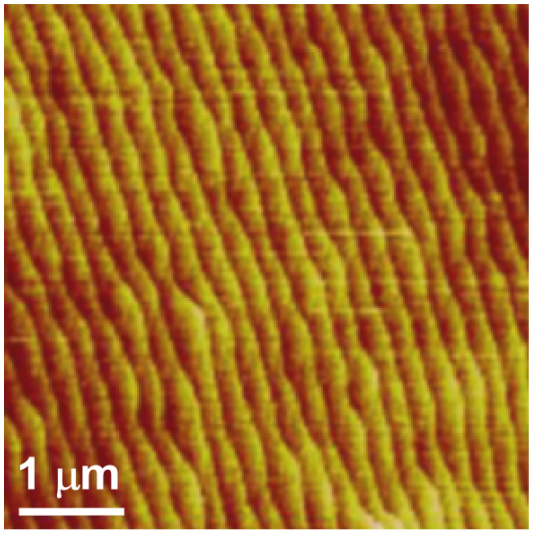
the $m \leq 6$ samples. The real $|D|$ value defined at the interface is thus deduced to be $m|D_{\mathrm{eff}}| = 14$ meV. This is much larger than those in the interface between metals: −2.2 meV for permalloy/Pt (*39*) and −1.05 meV for a Co/Ni multilayer on Pt(111) (*16*).

Matruno et al., *Science Advances* 2 : e1600304 (2016)

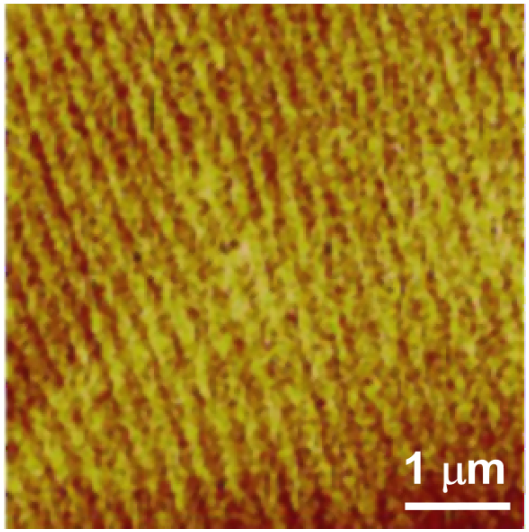
# $SrRuO_3/SrIrO_3$ bilayers



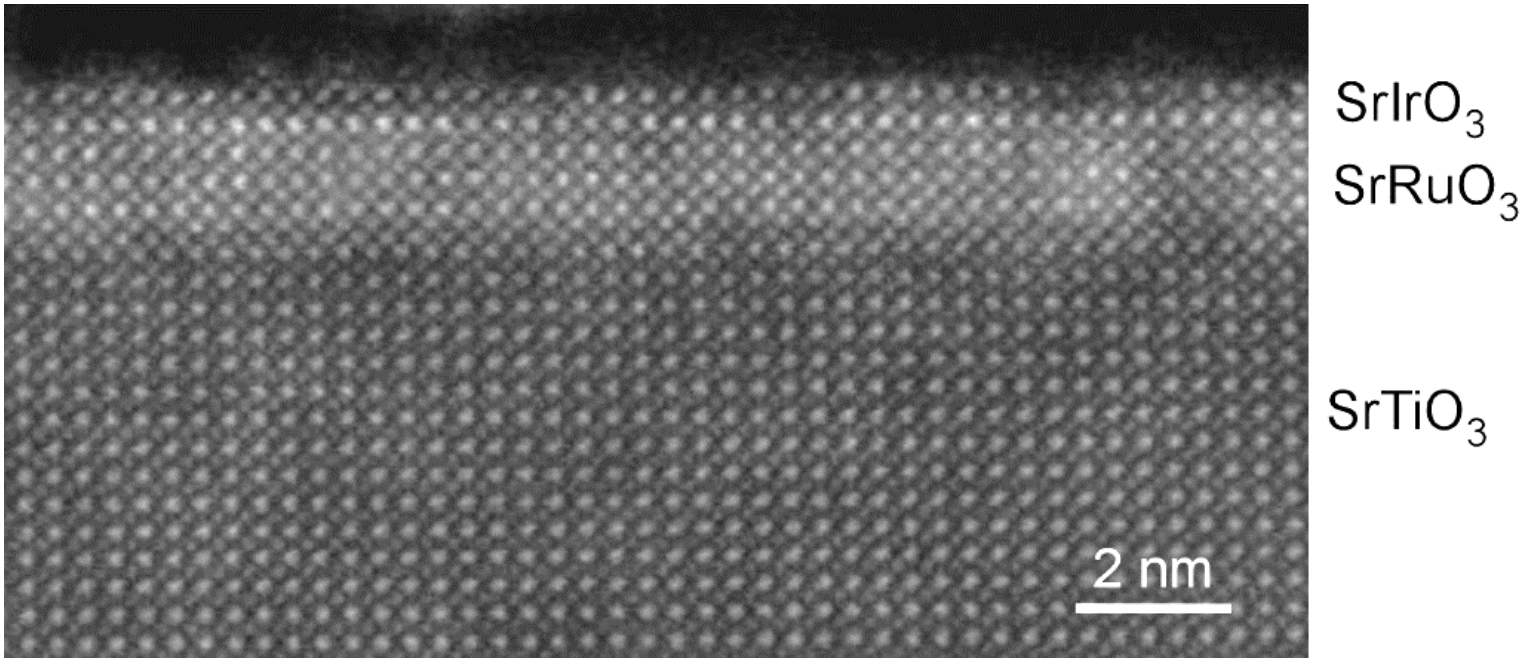

$SrTiO_3$ (001)




$SrRuO_3$(18 nm)/$SrTiO_3$ (001)



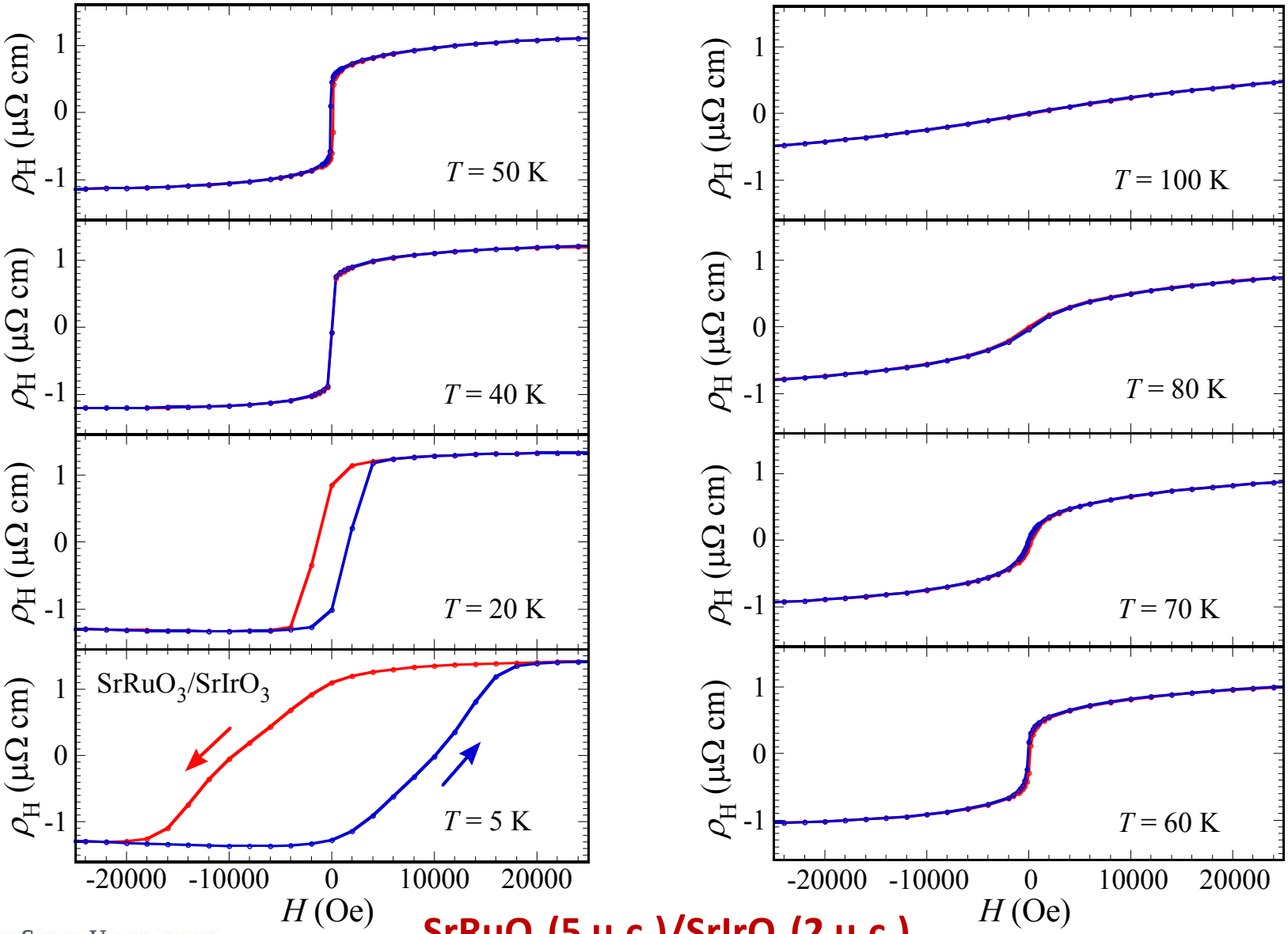
# $SrRuO_3/SrIrO_3$ bilayers




$SrRuO_3$: FM with $T_C$ ~ 160 K

Esser & McComb (OSU)

# Topological Hall Effect in $SrRuO_3/SrIrO_3$ bilayers




$SrRuO_3$(5 u.c.)/$SrIrO_3$(2 u.c.)

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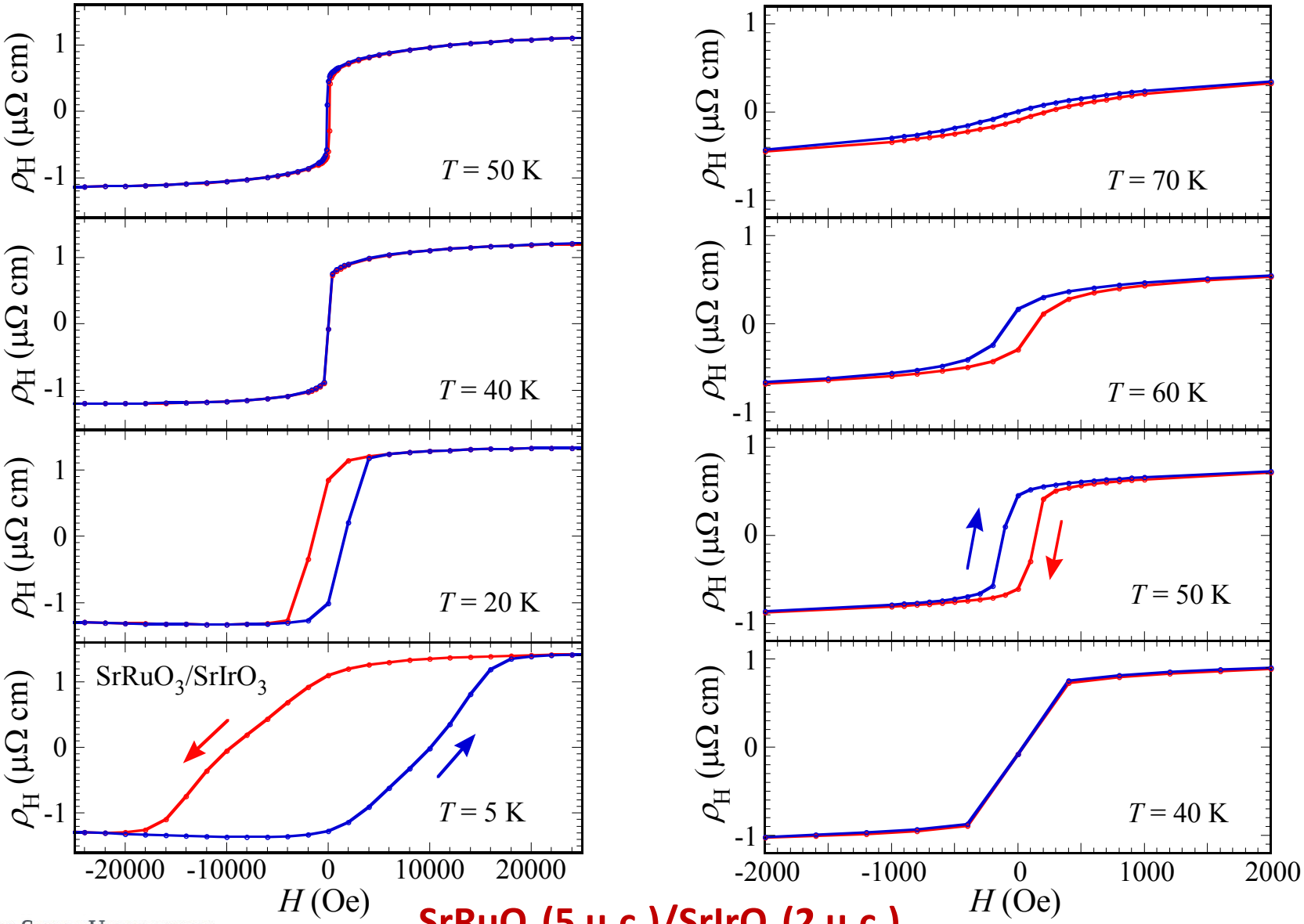


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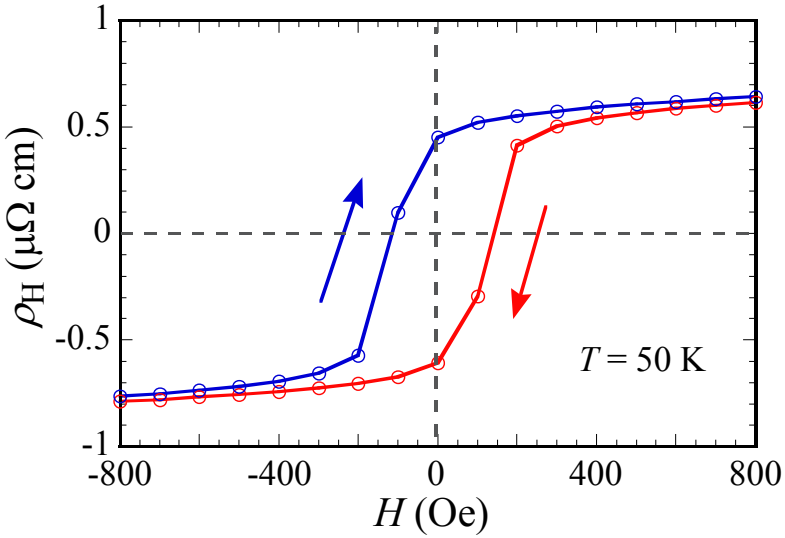
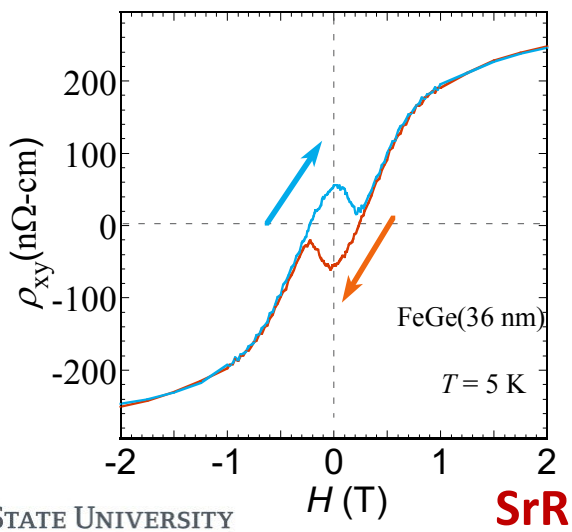
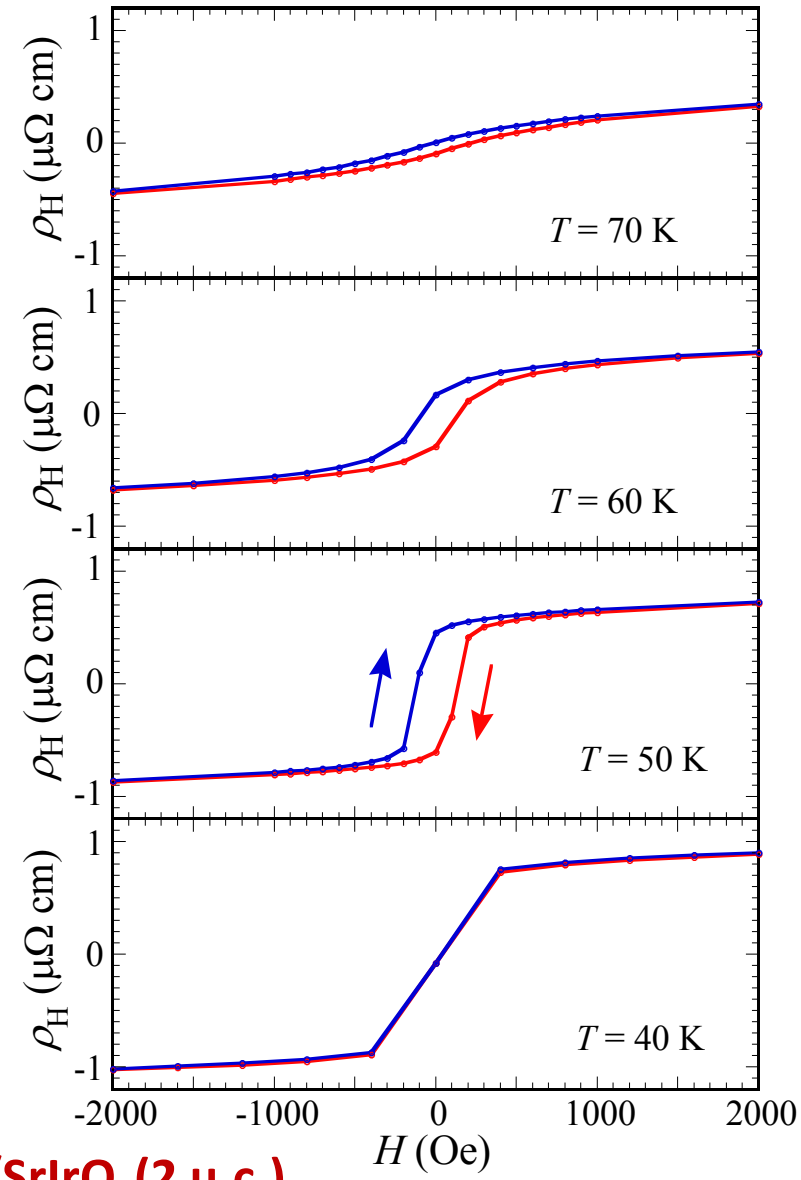
# Topological Hall Effect in $SrRuO_3/SrIrO_3$ bilayers

Raw Hall resistivity

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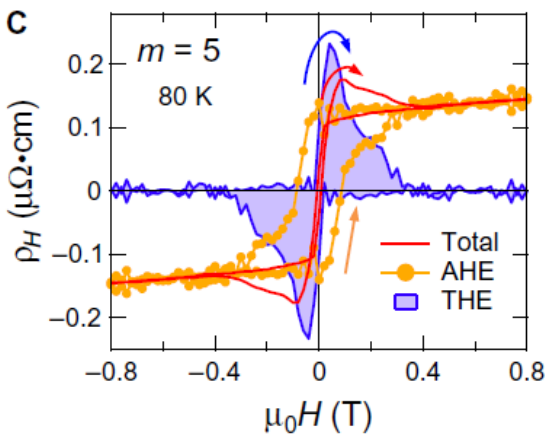
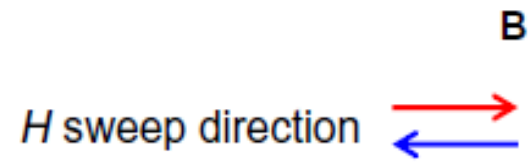
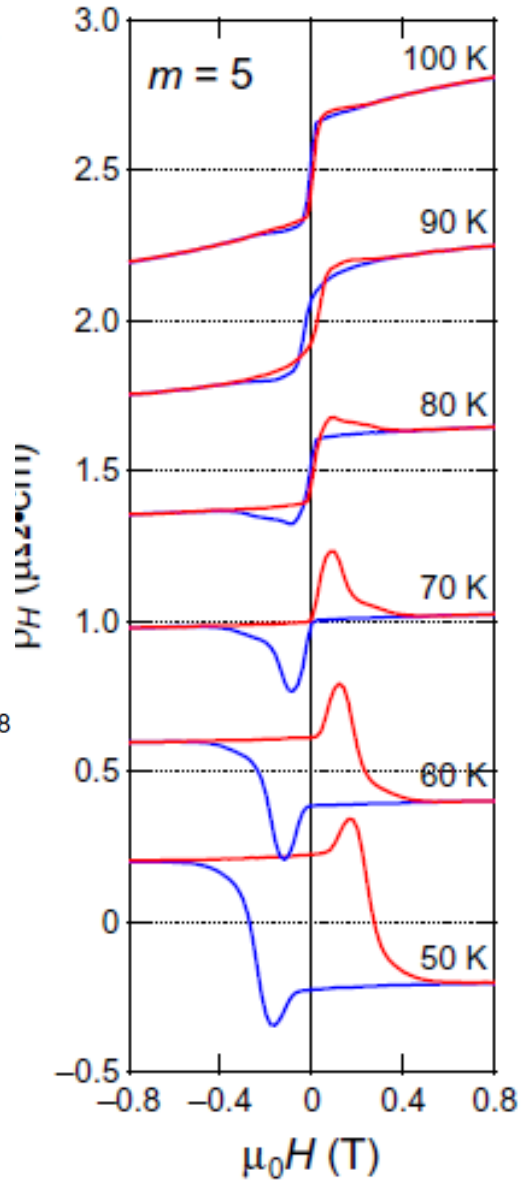
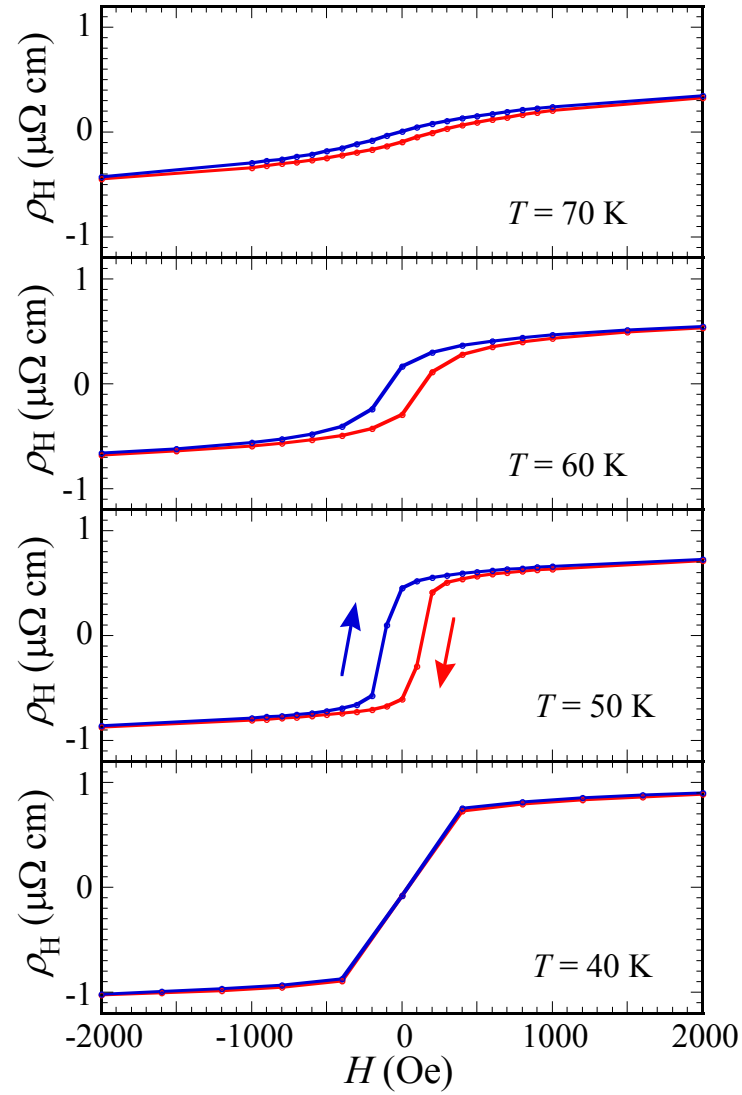


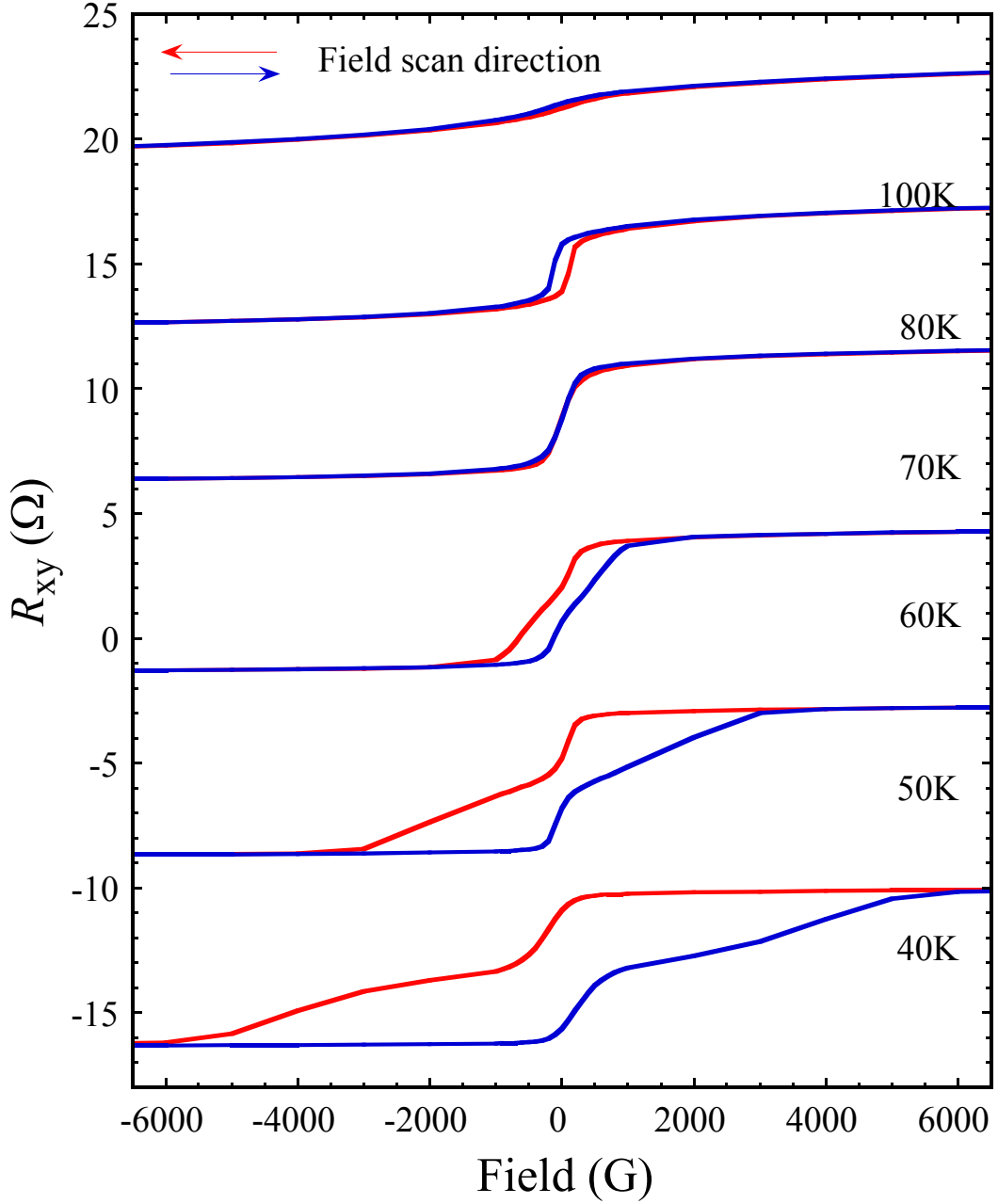




Matruno et al., *Science Advances* 2:e1600304 (2015)

$SrRuO_3$(5 u.c.)/$SrIrO_3$(2 u.c.)

## Single $SrRuO_3$ (6uc) layer on STO(001)



# Room-temperature oxide skyrmions

- High-$T_C$ FM oxides (LSMO, double perovskites)
- Strain-controlled anisotropy

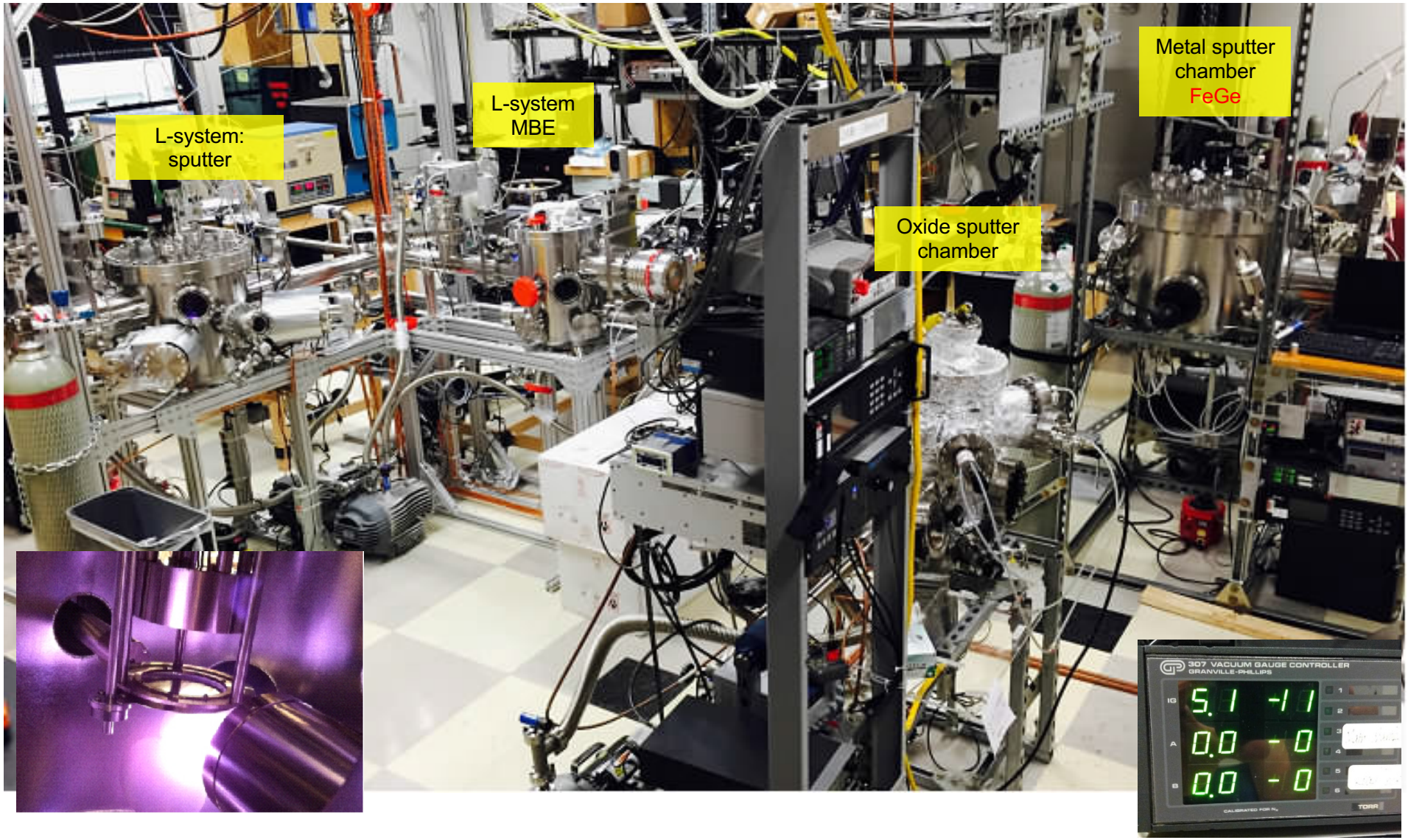
"Such interaction is expected to realize a 10-nm-sized magnetic skyrmion."

Matruno et al., *Science Advances* 2 : e1600304 (2016)

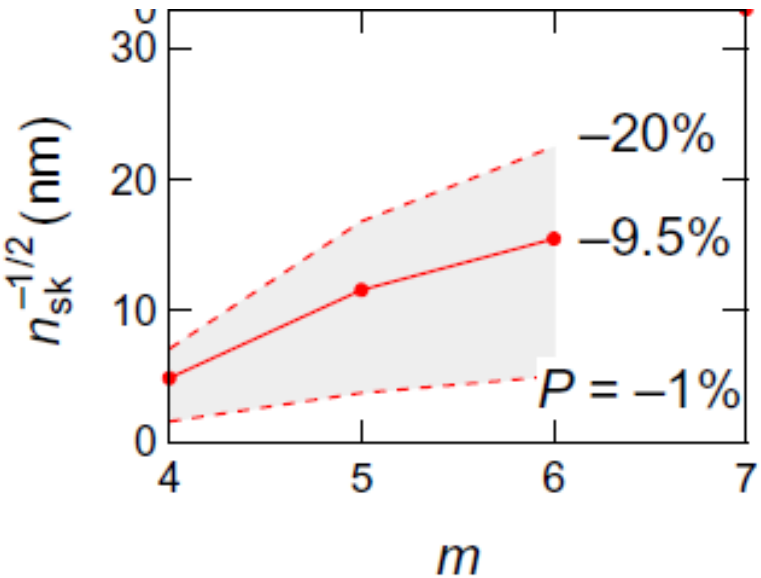
# Summary

- Growth of pure B20 phase, epitaxial FeGe films using UHV off-axis sputtering
- Confirmation of the pure B20 crystal structure using XRD and STEM
- Large topological Hall resistivity up to 970 nΩ-cm
- Robust Skyrmion formation at zero field
- Skyrmions in oxide FM/NM bilayers

# Sputter deposition of FeGe films



# Skyrmions in oxide bilayers




$$\rho_H^T = PR_0 b = PR_0 n_{sk} \phi_0$$

Matruno et al., *Science Advances* 2 : e1600304 (2016)

**The skyrmion number**

The swirling structure of a skyrmion is characterized by the topological skyrmion number defined by

$$N_{sk} = \frac{1}{4\pi}\iint d^2\mathbf{r}\,\mathbf{n}\cdot\left(\frac{\partial\mathbf{n}}{\partial x}\times\frac{\partial\mathbf{n}}{\partial y}\right) \quad \text{(B1)}$$

as the integral of the solid angle, and counts how many times $\mathbf{n}(\mathbf{r}) = \mathbf{n}(x,y)$ wraps the unit sphere[14,15]. Using the symmetry of the skyrmion, one can write

$$\mathbf{n}(\mathbf{r}) = (\cos\Phi(\varphi)\sin\Theta(r), \sin\Phi(\varphi)\sin\Theta(r), \cos\Theta(r)) \quad \text{(B2)}$$

where we introduce the polar coordinates $\mathbf{r} = (r\cos\varphi, r\sin\varphi)$. Putting this form into equation (B1), we obtain

$$N_{sk} = \frac{1}{4\pi}\int_0^\infty dr\int_0^{2\pi} d\varphi\,\frac{d\Theta(r)}{dr}\,\frac{d\Phi(\varphi)}{d\varphi}\sin\Theta(r) = [\cos\Theta(r)]_{r=0}^{r=\infty}[\Phi(\varphi)]_{\varphi=0}^{\varphi=2\pi}$$

Accordingly, one can classify the skyrmion structures as follows. Suppose the spins point up at $r \to \infty$ while they point down at $r = 0$. Then, $[\cos\Theta(r)]_{r=0}^{r=\infty} = 2$. Now, there are several possibilities for $\Phi(\varphi)$. The vorticity is defined by the integer $m = [\Phi(\varphi)]_{\varphi=0}^{\varphi=2\pi}/2\pi$. Therefore, the vorticity determines the skyrmion number as $N_{sk} = m$, once the boundary condition at $r \to \infty$ is fixed. We further define the helicity by the phase $\gamma$ appearing in

$$\Phi(\varphi) = m\varphi + \gamma \quad \text{(B3)}$$

We show in Fig. 1g various skyrmion structures corresponding to vorticity $m = \pm 1$, and $\gamma = 0, \pm\pi/2$ and $\pi$, Fig. 1a corresponds to $m = 1$ and $\gamma = \pi/2$.

From equations (B2) and (B3), the magnetic charge $\rho_{mag} = \nabla\cdot\mathbf{n}$ is calculated as

$$\rho_{mag} = \cos[(m-1)\varphi + \gamma]\left(\frac{d\Theta}{dr}\cos\Theta + \frac{m}{r}\sin\Theta\right)$$

On the other hand, the DM interaction is given by

$$H_{DM} = D\mathbf{n}\cdot(\nabla\times\mathbf{n}) = D\sin[(m-1)\varphi + \gamma]\left(\frac{d\Theta}{dr} + \frac{m}{2r}\sin 2\Theta\right)$$

or

$$H_{DM} = D\mathbf{n}\cdot(\mathbf{e}_z\times\nabla)\mathbf{n} = D\sin[(m-1)\varphi + \gamma]\left(\frac{d\Theta}{dr} + \frac{m}{2r}\sin 2\Theta\right)$$

Therefore, the state with vorticity $m = +1$ and $\gamma = \pm\pi/2$ has the lowest energy, where the sign of $\gamma$ is determined by the sign of $D$, which in turn is determined by the crystal structure. (When the DM vector is rotated by $\pm\pi/2$, $\gamma = 0$ or $\pi$ becomes the stable configuration[109].)

In the case of the four-spin interaction (case 3) or the frustrated exchange interaction (case 4), there is no distinction between the skyrmion and anti-skyrmion, that is, $m = \pm 1$ and $\gamma$ can take an arbitrary value.

**Emergent electromagnetic field**

One can describe the interaction between the spin textures and the conduction electrons in terms of the EEMF expressed by the spin directions. We consider the double-exchange model where the conduction electrons and spins are coupled ferromagnetically at each site by Hund's rule coupling. In the strong coupling limit, the spin wavefunction $|\chi(\mathbf{r})\rangle$ of the conduction electron at $\mathbf{r}$ corresponding to the localized spin $\mathbf{n}(\mathbf{r})$ in equation (B2) is given by

$$|\chi(\mathbf{r})\rangle = \left(\cos\frac{\Theta(\mathbf{r})}{2}, e^{i\Phi(r)}\sin\frac{\Theta(\mathbf{r})}{2}\right)^T$$

where $^T$ means the transpose. Therefore, when a conduction electron hops between two sites $\mathbf{r}$ and $\mathbf{r} + c\boldsymbol{\eta}_\alpha$ ($\boldsymbol{\eta}_\alpha$ is the unit vector along $\alpha(=x,y,z)$ direction, $\alpha$ is the space index and $c$ a lattice constant), the matrix element is given by

$$t_\alpha(\mathbf{r}) = t\langle\chi(\mathbf{r})|\chi(\mathbf{r} + c\boldsymbol{\eta}_\alpha)\rangle$$

where $t$ is the original transfer integral of the conduction electron. $t_\alpha(\mathbf{r})$ is in general a complex number, and can be written as $t_\alpha(\mathbf{r}) = |t_\alpha(\mathbf{r})|e^{ica_\alpha(r)}$. This phase factor $e^{ica_\alpha(r)}$ is analogous to the Peierls factor in the presence of the external magnetic field and hence we can regard $a_\alpha(\mathbf{r})$ as the vector potential of an effective electromagnetic field. Assuming the slowly varying spin configuration over the lattice constant $c$, we obtain $a_\alpha(\mathbf{r}) = -i\langle\chi(\mathbf{r})|\partial_\alpha\chi(\mathbf{r})\rangle = \frac{1}{2}\partial_\alpha\Phi(1 - \cos\Theta)$. From this expression, one can easily confirm that the emergent magnetic field $b_z$ is related to the solid angle as

$$b_z = \frac{\partial a_y}{\partial x} - \frac{\partial a_x}{\partial y} = \frac{1}{2}\mathbf{n}\cdot\left(\frac{\partial\mathbf{n}}{\partial x}\times\frac{\partial\mathbf{n}}{\partial y}\right)$$

Therefore, the total emergent magnetic flux associated with a skyrmion is $2\pi N_{sk}$. These considerations can be easily generalized to the three-dimensional case and also to the emergent electric field $e_\alpha$ as

$$b_\alpha = \tfrac{1}{2}\varepsilon^{\alpha\beta\gamma}\mathbf{n}\cdot(\partial_\beta\mathbf{n}\times\partial_\gamma\mathbf{n}) \quad \text{(B4a)}$$

$$e_\alpha = \mathbf{n}\cdot(\partial_\alpha\mathbf{n}\times\partial_t\mathbf{n}) \quad \text{(B4b)}$$

where $\partial_\mu = \partial/\partial x_\mu$, $\varepsilon^{\alpha\beta\gamma}$ is the totally antisymmetric tensor in three dimensions, and the coupling to the conduction electrons is described by the Lagrangian

$$L_{int} = j_\mu a_\mu \quad \text{(B5)}$$

with $j_\mu$ being the density and current density of conduction electrons as in the case of the Maxwell electromagnetic field $A_\mu$. Here $\mu$ is the space–time index.

We mention here that the well-known spin-transfer torque arises from equation (B5). Namely, the variation of $a_\mu$ with respect to $\mathbf{n}$ is given by

$$\delta a_\mu = -\tfrac{1}{2}\delta\mathbf{n}\cdot(\mathbf{n}\times\partial_\mu\mathbf{n}) \quad \text{(B6)}$$

Combined with the variation of the Berry phase term $\omega$ as $\delta\omega = -\frac{1}{2}\delta\mathbf{n}\cdot(\mathbf{n}\times\partial_t\mathbf{n})$, equation (B6) leads to the equation of motion as

$$[\partial_t + (\mathbf{j}\cdot\nabla)]\mathbf{n}(\mathbf{r},t) = 0$$

which describes the current-driven motion of the spin texture with spatially varying $\mathbf{n}$.

U.S. DEPARTMENT OF ENERGY

Skyrmions exhibit topological phenomena due to the emergent electromagnetic field (EEMF)

→ The topological Hall effect is induced by the emergent magnetic field of the skyrmions on conduction electrons.

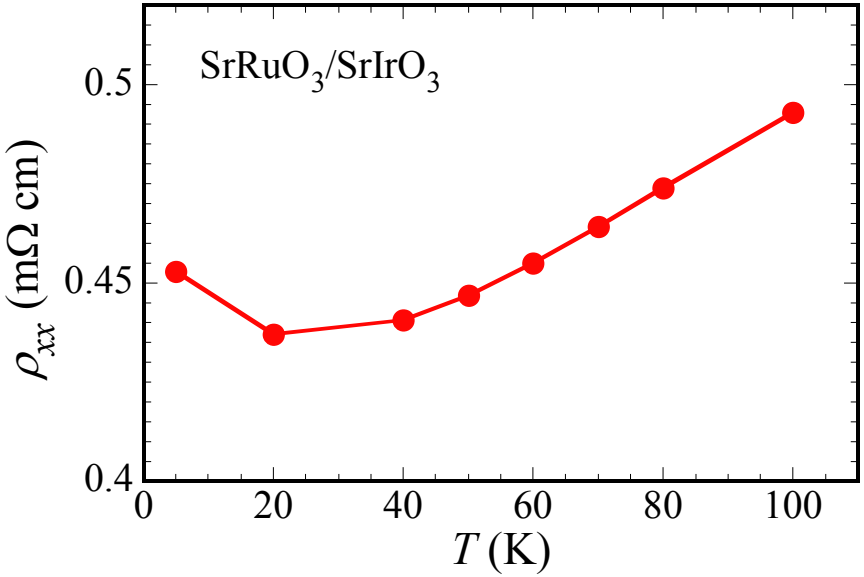
→ The motion of the skyrmions leads to the temporal change of the emergent magnetic field

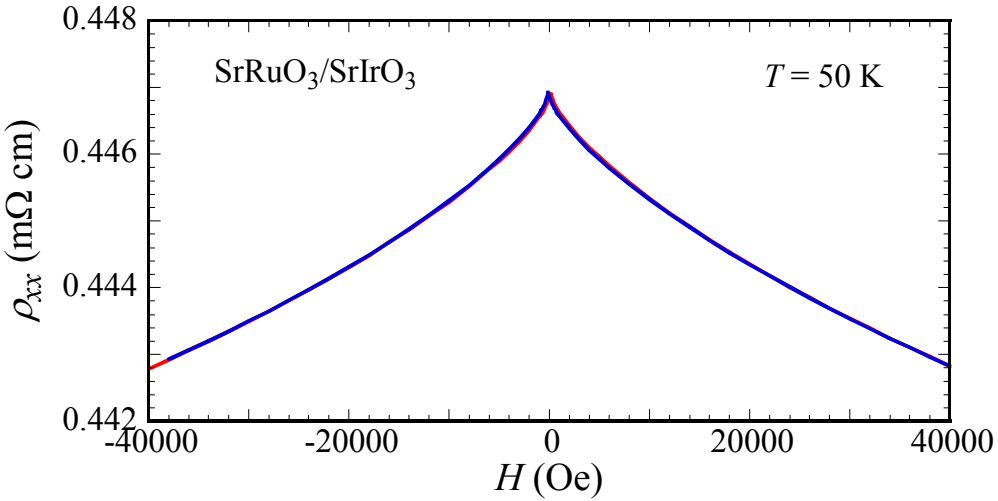
→ An electromagnetic induction.

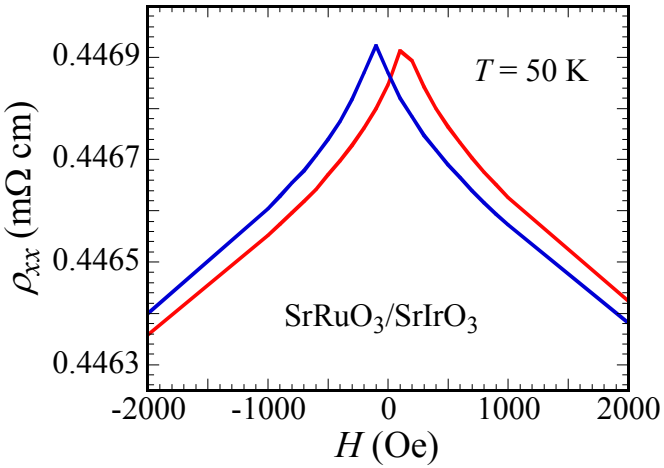
→ The induced emergent electric field contributes to the Hall effect when the skyrmions move

Nagaosa & Tokura, *Nature Nano.* 8, 899 (2013)

# Longitudinal resistivity of $SrRuO_3/SrIrO_3$ bilayers



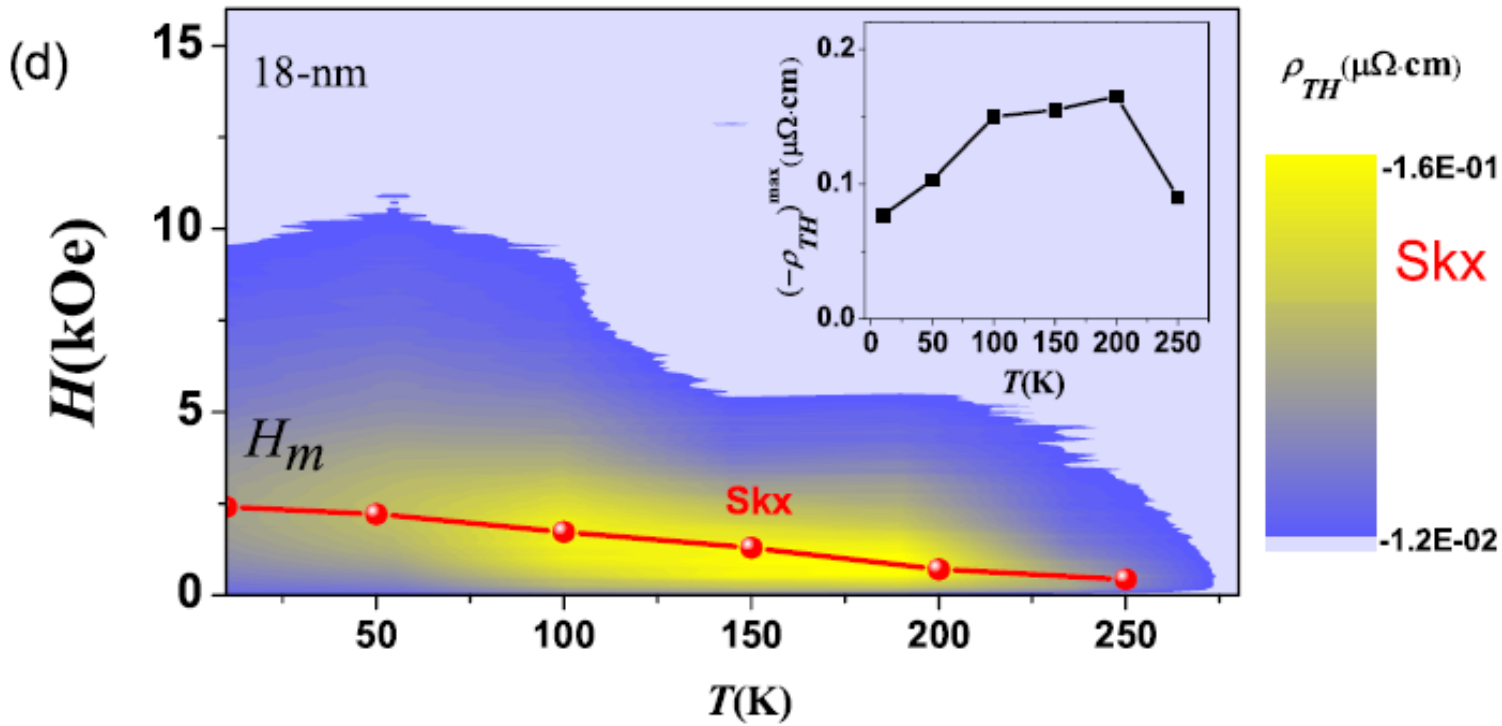



# Topological Hall effect in skyrmion films

PRL 108, 267201 (2012) PHYSICAL REVIEW LETTERS week ending 29 JUNE 2012

**Extended Skyrmion Phase in Epitaxial FeGe(111) Thin Films**

S. X. Huang* and C. L. Chien†



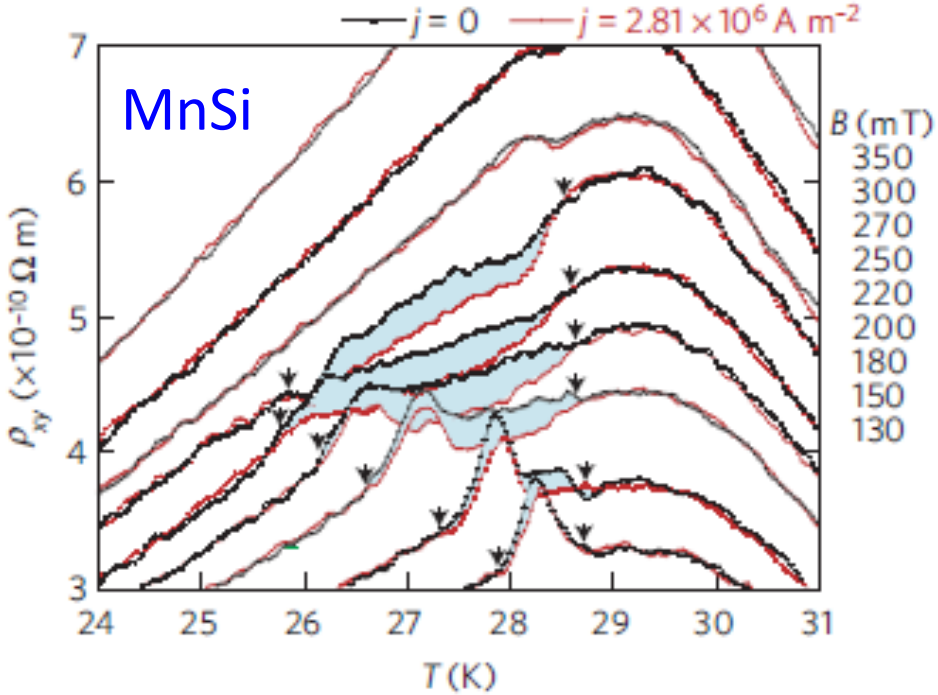

Huang & Chien. *Phys. Rev. Lett*. 108, 267201 (2012)

# Topological Hall effect in skyrmion films

nature physics LETTERS
PUBLISHED ONLINE: 19 FEBRUARY 2012 | DOI: 10.1038/NPHYS2231

## Emergent electrodynamics of skyrmions in a chiral magnet

T. Schulz[1], R. Ritz[1], A. Bauer[1], M. Halder[1], M. Wagner[1], C. Franz[1], C. Pfleiderer[1]*, K. Everschor[2], M. Garst[2] and A. Rosch[2]*




Schulz, et al. *Nat. Phys.* 8, 301 (2012)