

# Anomalous Hall/Nernst Effects in Magnetic Semiconductors and Magnetic Insulators

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Collaborators:

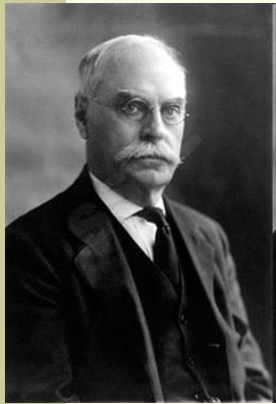
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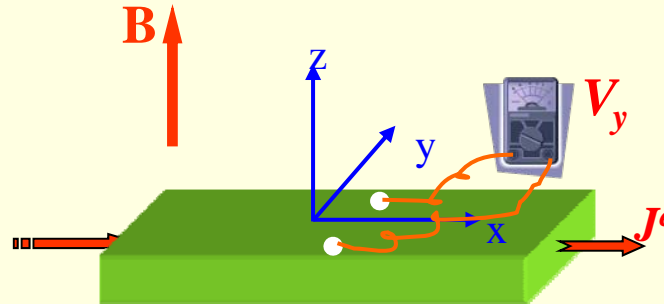
# Outline

- Introduction
  - Anomalous Hall effect (AHE)
  - Anomalous Nernst effect (ANE)
- GaMnAs: Dilute Magnetic Semiconductor (DMS)
  - AHE/ANE in absence of B-field
  - Validity of Mott relation with  $n=2$
  - Hole-mediated ferromagnetism, probably via impurity band
- $\text{Fe}_3\text{O}_4$ : Ferrimagnetic Insulator (FMI)
  - Robust AHE power-law scaling with  $n=0.3$
  - Preliminary ANE data: absence of ANE
- Summary

# Anomalous Hall Effect (AHE)



In ferromagnets,  $\rho_{xy}$  contains two parts:



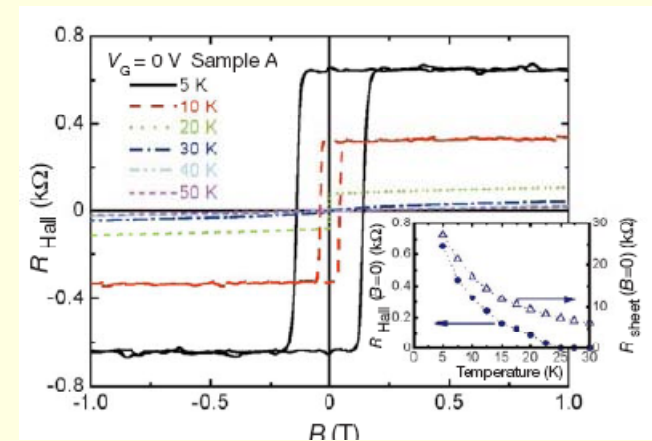
Normal or ordinary Hall effect (OHE)

$$\rho_{xy} = R_0 B + R_s M$$

$\rho_{AH}$ : anomalous or extraordinary Hall effect (AHE)

- $\rho_{xy}$  exists even if  $B=0$
- AHE is more than an order greater than OHE
- ➔  $\rho_{xy} \sim \rho^{AH}$

AHE is not caused by magnetic field, but by spin-orbit coupling (SOC)



AHE from 5 nm-thick (In, Mn)As layer

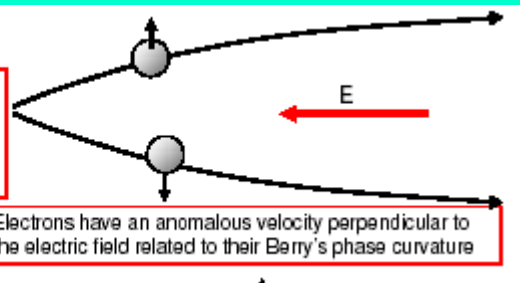
# Physical Origin of AHE

Spin-orbit effect: extrinsic (scattering) or intrinsic (band structure)

## ❖ Karplus & Luttinger (intrinsic: inter-band effect)

### Intrinsic deflection

Interband coherence induced by an external electric field gives rise to a velocity contribution perpendicular to the field direction. These currents do not sum to zero in ferromagnets.



$$\frac{d\langle \vec{r} \rangle}{dt} = \frac{\partial E}{\hbar \partial \vec{k}} + \frac{e}{\hbar} \vec{E} \times \vec{b}_v$$

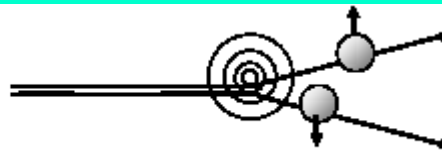
Electrons have an anomalous velocity perpendicular to the electric field related to their Berry's phase curvature

See excellent review articles: N.A. Sinitsyn, J. Phys.: Condens. Matter (2008); N. Nagaosa et al., Rev. Mod. Phys. (2010).

## ❖ Smit (extrinsic: skew scattering)

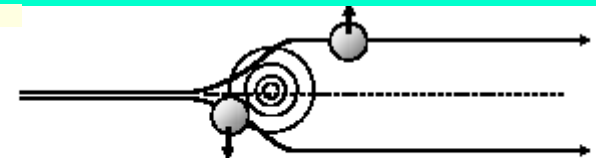
### Skew scattering

Asymmetric scattering due to the effective spin-orbit coupling of the electron or the impurity.



## ❖ Berger (extrinsic: side-jump)

### Side jump



The electron velocity is deflected in opposite directions by the opposite electric fields experienced upon approaching and leaving an impurity. The time-integrated velocity deflection is the side jump.

## ❖ Niu & MacDonald (intrinsic: Berry's phase)

# Power-Law

Power-law:

$$R_s = \lambda \rho_{xx}^n$$

❖ Exponent  $n=2$ :

$$\sigma_{xy} \sim \rho_{xy} / \rho_{xx}^2 \quad (\rho_{xx} \gg \rho_{xy})$$

→ independent of  $1/\tau$ !

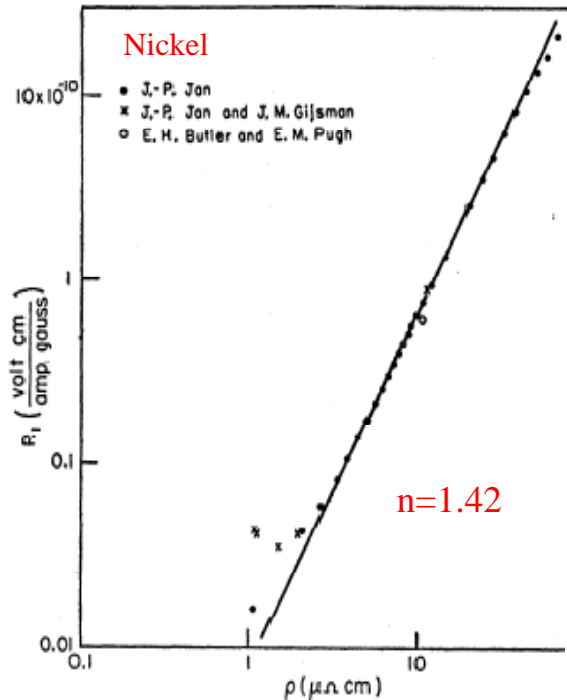
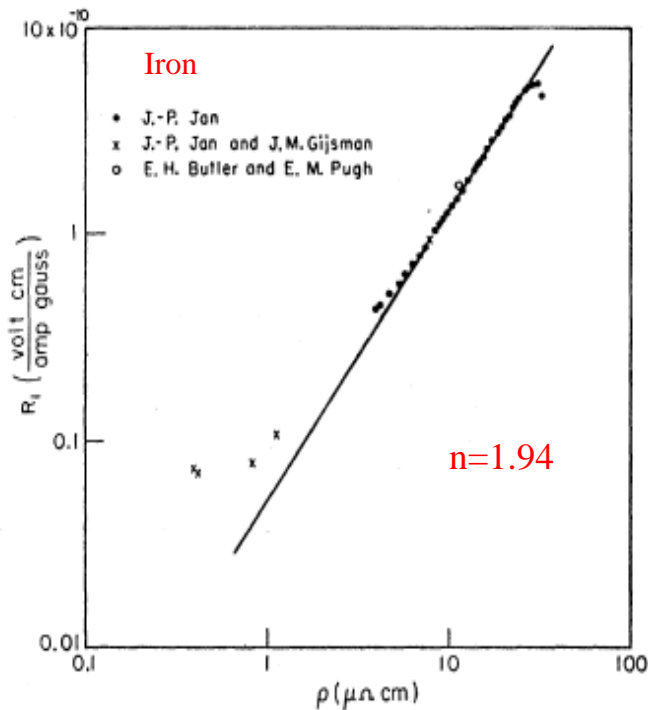
→ Special Hall current:  $J_H = \sigma_{xy} E_x$

❖ Exponent  $n=1$ :

$$\sigma_{xy} \sim \sigma_{xx}$$

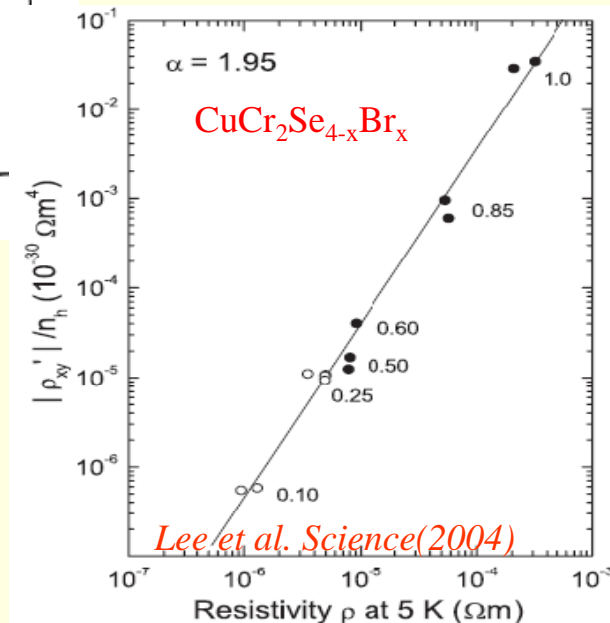
skew scattering (extrinsic)

# Experimental Data



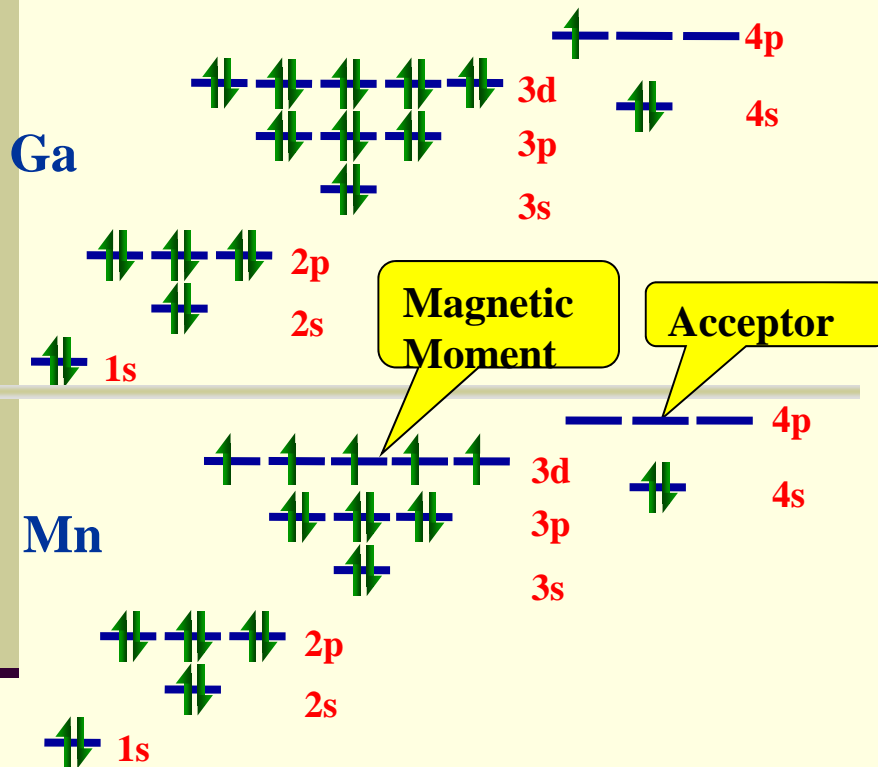
Pure metals are not ideal

- Scattering rate can be tuned by impurity, temperature or magnetic field
- Semiconductors or alloys are preferred



# Dilute Magnetic Semiconductors (DMS)

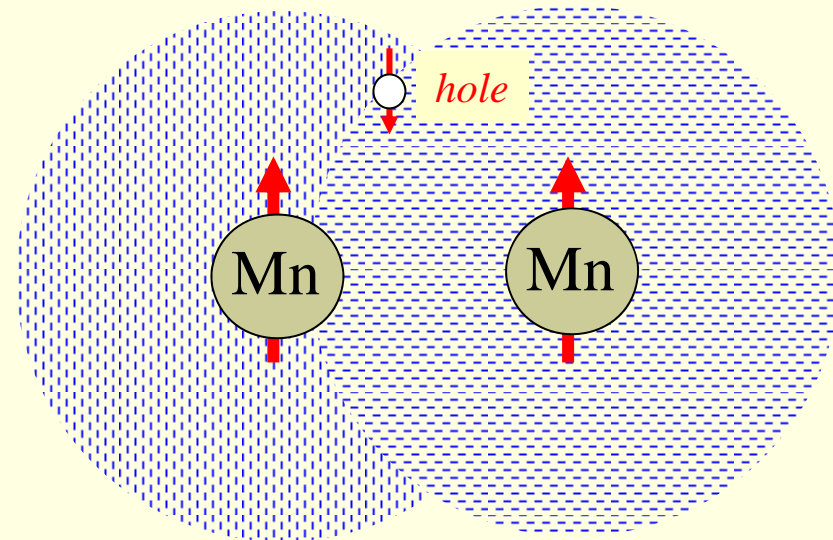
$\text{Ga}_{1-x}\text{Mn}_x\text{As}$ : most studied DMS



Mn substitutes Ga in GaAs: introducing spin and charge carriers!

## Carrier-mediated interaction

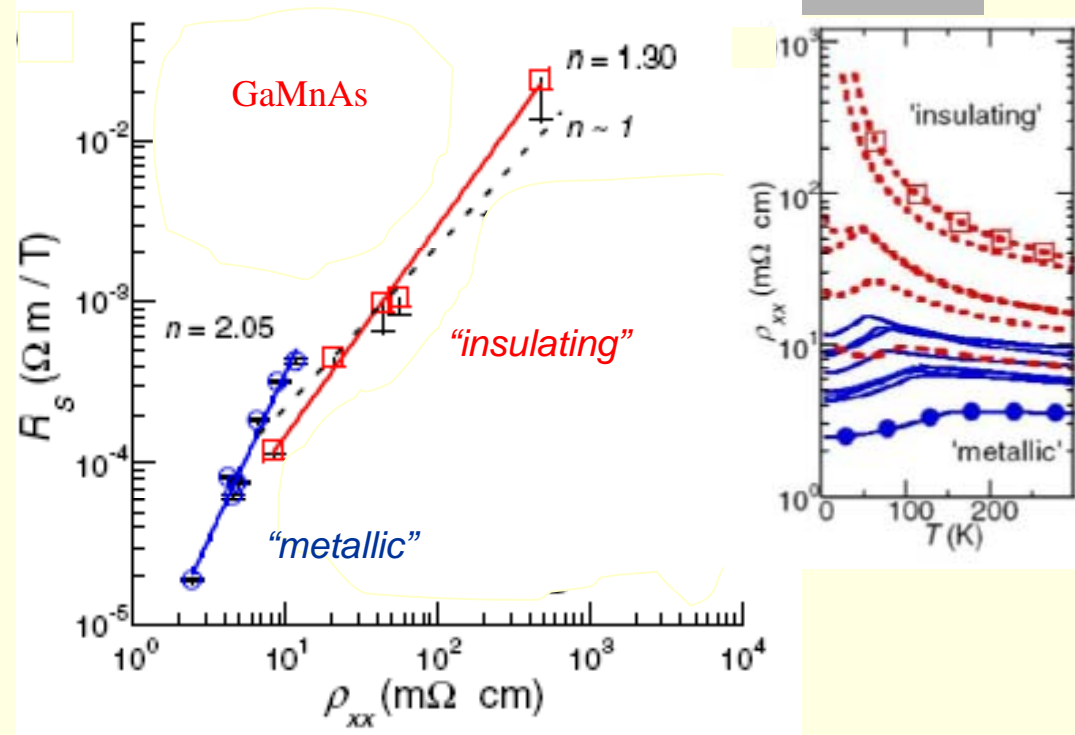
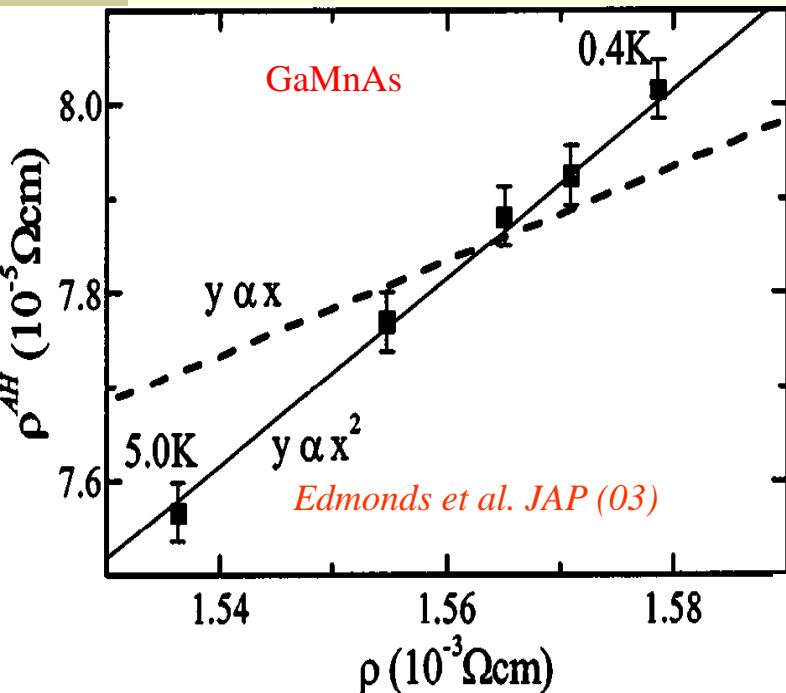
Zener model:  $T_c \sim x \cdot p^{1/3}$



Highest  $T_c \sim 150$  K!

- Strong SOC for holes in GaAs
- Strong impurity scattering

# AHE in DMS



Chun et al., PRL (07)

Results support intrinsic mechanism (but the resistivity range is too narrow)

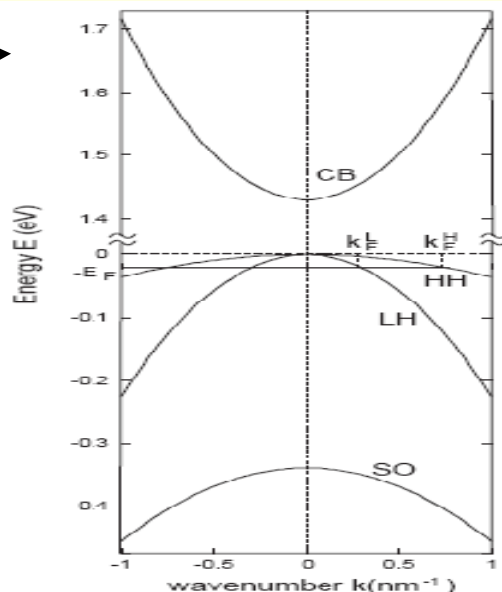
$n \sim 2$  and AHE's carrier density dependence in "metallic" regime  $\rightarrow$  intrinsic origin



# DMS: Intrinsic AHE

GaAs band structure →

- Spin-orbit coupling
- External electric field  $\vec{E}$



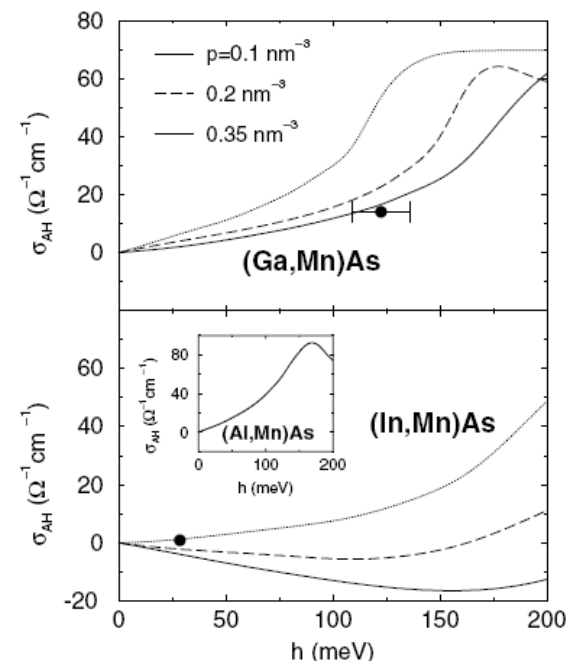
Electron wavepackets acquire additional velocity:

$$\dot{\vec{x}}_c = \frac{\partial \epsilon}{\hbar \partial \vec{k}} + \frac{(e/\hbar)\vec{E} \times \vec{\Omega}}{\omega}$$

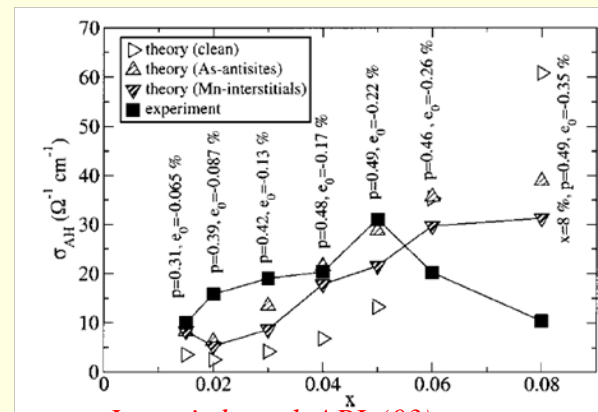
Anomalous velocity

Under broken time reversal symmetry, this Berry phase effect alone gives rise to AHE comparable with experimental values

→ Intrinsic origin of AHE in DMS

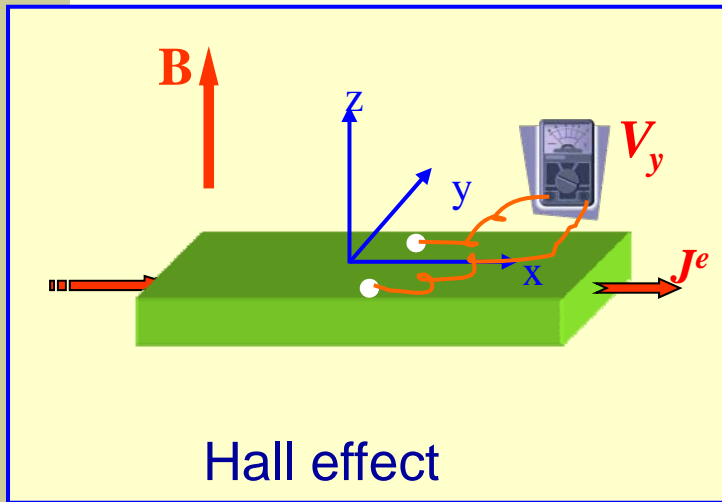


Jungwirth, Niu and McDonald, PRL (02)



Jungwirth et al. APL (03)

# Nernst Effect



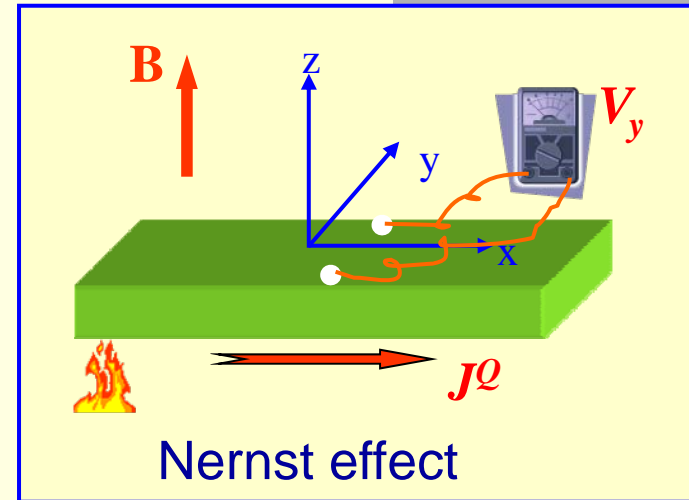
$$\sigma_H = \frac{E_y}{J_x}$$

- Normal Hall Effect

$$\sigma_H \propto B$$

- Anomalous Hall Effect (AHE)

$$\sigma_{AH} \propto M$$



$$S_N = \frac{E_y}{(\nabla T)_x} = - \frac{\Delta V_y}{\Delta T_x} \cdot \frac{L}{W}$$

- Normal Nernst Effect

$$S_N \propto B$$

- Anomalous Nernst Effect (ANE)

$$S_{AN} \propto M$$

# Other Transport Effects

**Stimulus:**  $\Delta V_x$  or  $\Delta T_x$

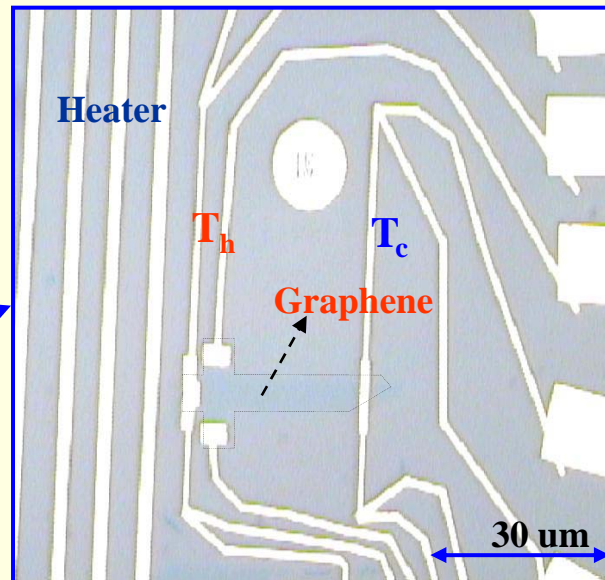
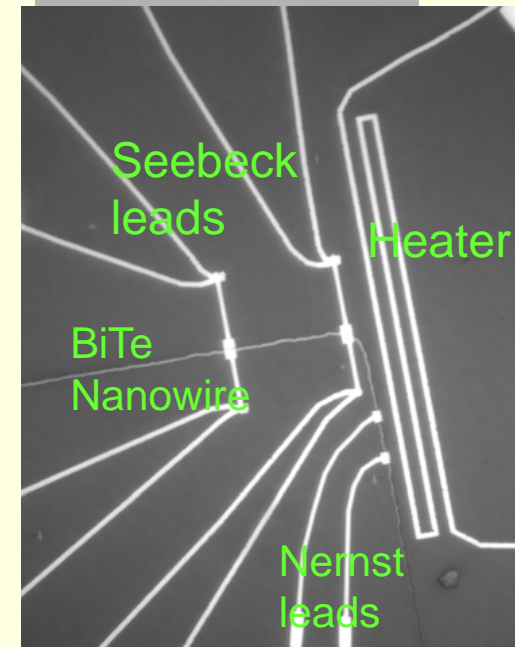
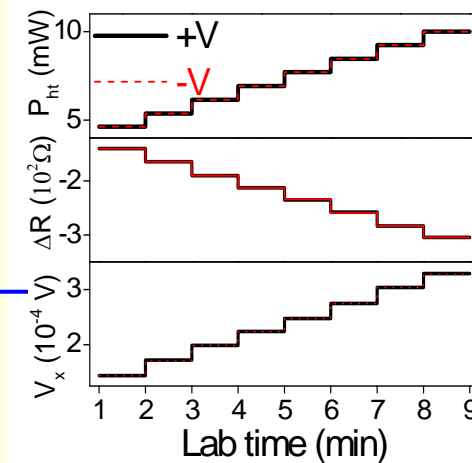
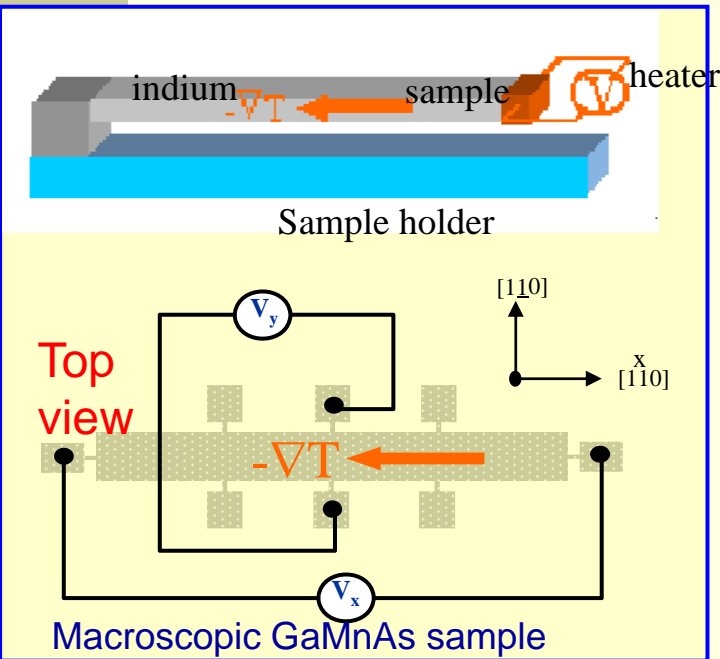
**Response:**  $\rightarrow I_x, I_x^Q$  and  $I_y, I_y^Q$  if there is B or M.

**Open-circuit condition:** measuring  $\Delta V$  instead of  $I$ ;  $\Delta T$  instead of  $I^Q$

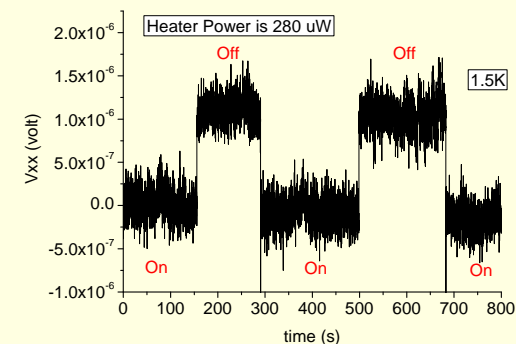
Response Stimulus	$I_x$	$I_y$	$I_x^Q$ or $\Delta T_x$	$I_y^Q$ or $\Delta T_y$
$\Delta V_x$	Conductivity	Hall effect	Peltier effect	Ettingshausen effect
$\Delta T_x$	Seebeck effect	Nernst effect	Thermal conductivity	Righi-Leduc effect

Various coefficients are connected by Onsager relations and other relations (e.g. Wiedemann-Franz law, Mott relation, etc.)

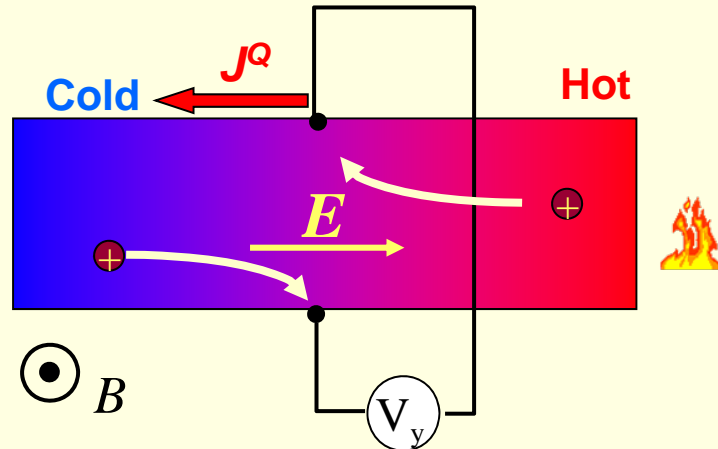
# Thermoelectric Measurements



Typical  $\Delta T \sim 50$  mK;  $\Delta T$  is measured by microfabricated thermometers.



# Nernst Effect



Open circuit voltage ( $J_x$  and  $J_y=0$ )

$$\vec{J} = \sigma \vec{E} + \alpha (-\nabla T)$$

$$S_{yx} = \frac{1}{\sigma_{xx}} (\alpha_{yx} - \sigma_{yx} S_{xx})$$

diffusion
drift

Nernst current
Hall current

- Nernst effect is a net effect resulting from both Hall current (drift) and Nernst current (diffusion). In metals, it is a very small effect.

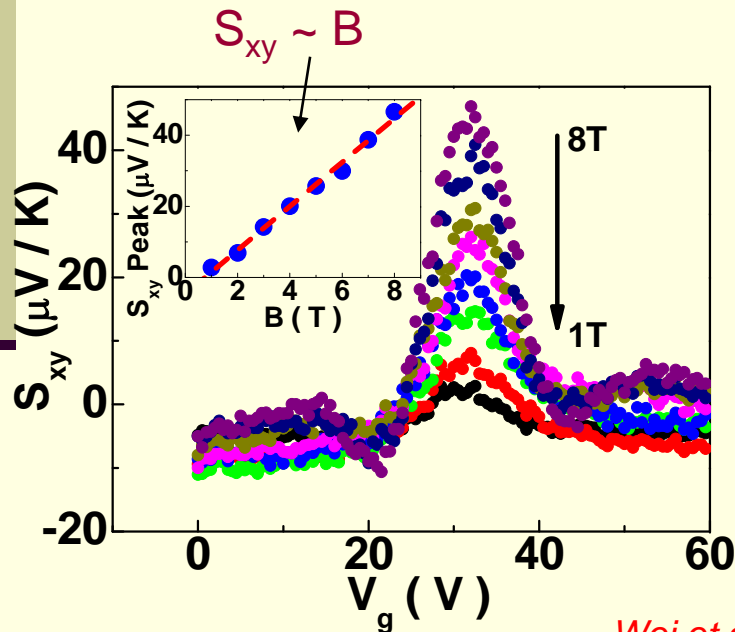
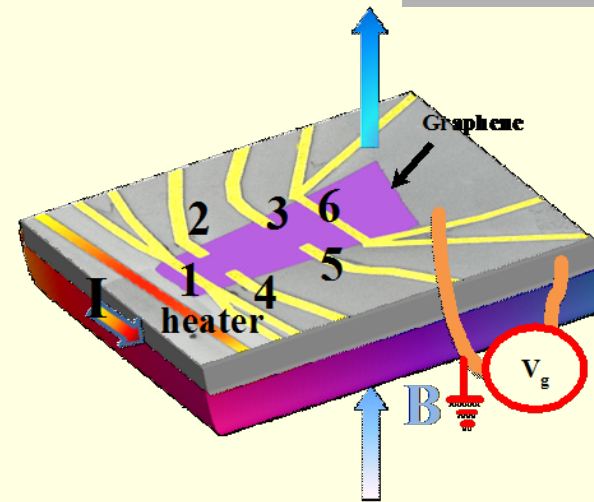
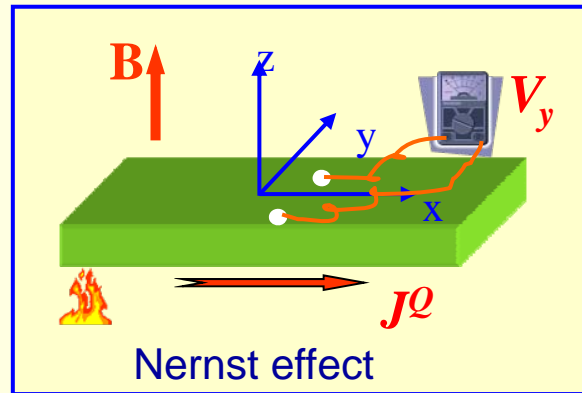
Mott relations

$$\alpha_{xx} = \frac{\pi^2 k_B^2 T}{3e} \left( \frac{\partial \sigma_{xx}}{\partial \varepsilon} \right)_{\varepsilon_F}$$

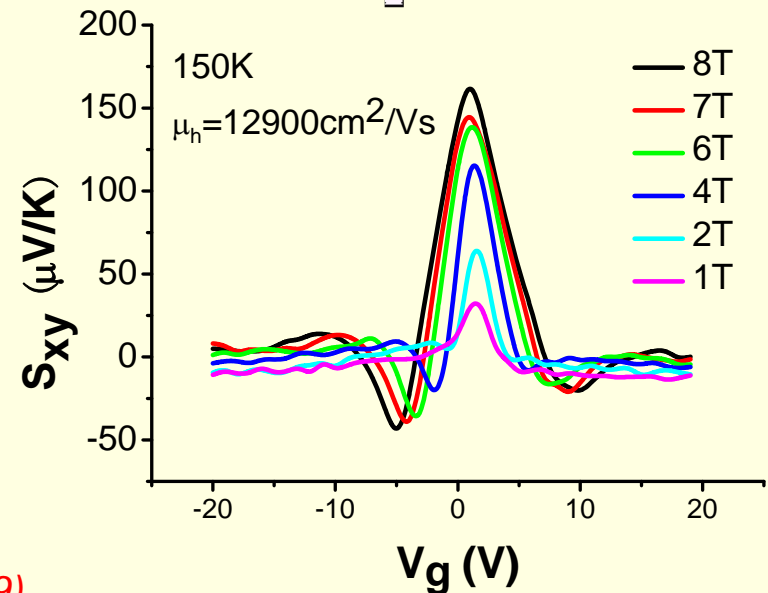
$$\alpha_{yx} = \frac{\pi^2 k_B^2 T}{3e} \left( \frac{\partial \sigma_{yx}}{\partial \varepsilon} \right)_{\varepsilon_F}$$

$$S_{yx} = \frac{\pi^2 k_B^2 T}{3e} \left[ \arctan \left( \frac{\sigma_{yx}}{\sigma_{xx}} \right) \right]' = \frac{\pi^2 k_B^2 T}{3e} \left( \frac{\partial \Theta_H}{\partial \varepsilon} \right)_{\varepsilon_F}$$

# Normal Nernst Effect in Graphene



Wei et al. PRL (09)



# Why Anomalous Nernst Effect?

- Does ANE exist if AHE is caused by intrinsic effect?
  - Only statistical force (no E-field)

$$\dot{\vec{x}}_c = \frac{\partial \epsilon}{\hbar \partial \vec{k}} + (e/\hbar) \vec{E} \times \vec{\Omega}.$$

- If  $I$  does not depend on Fermi energy, there will be no ANE

$$\boxed{\rho_{AH} = \lambda \rho_{xx}^2 M_z} \rightarrow \boxed{\sigma_{AH} = \lambda M_z} \quad \text{Finite } S_{AN} \rightarrow \lambda = \lambda(\epsilon_F)!$$

- How are AHE and ANE related?
  - Validity of Mott relation (D. Xiao PRL 07)
  - Does ANE help us understand physical origin of AHE
- What can we learn about ferromagnets?
  - Spin-orbit coupling, magnetic ordering

# Subtleties with DMS

- Films with in-plane anisotropy often require high magnetic fields to obtain finite AHE (i.e.  $M_z$ ). To completely saturate  $M_z$ , it requires magnetic fields in excess of 10 T!
- Normal Hall/Nernst signals become large at high fields.
- High magnetic fields cause significant magneto-resistance (i.e. change in  $\rho_{xx}$ ), even when AHE saturates.
- In DMS, all spins contribute to magnetization, but only those in hole-rich regions contribute to AHE; therefore, it is difficult to separate these two for  $\rho_{AH} = \lambda \rho_{xx}^2 M_z$ .

## Solutions:

- DMS films with perpendicular anisotropy (no need to have B-field)
- ANE and AHE measured simultaneously from the same area (no need to measure M)



# GaMnAs with Perpendicular Anisotropy

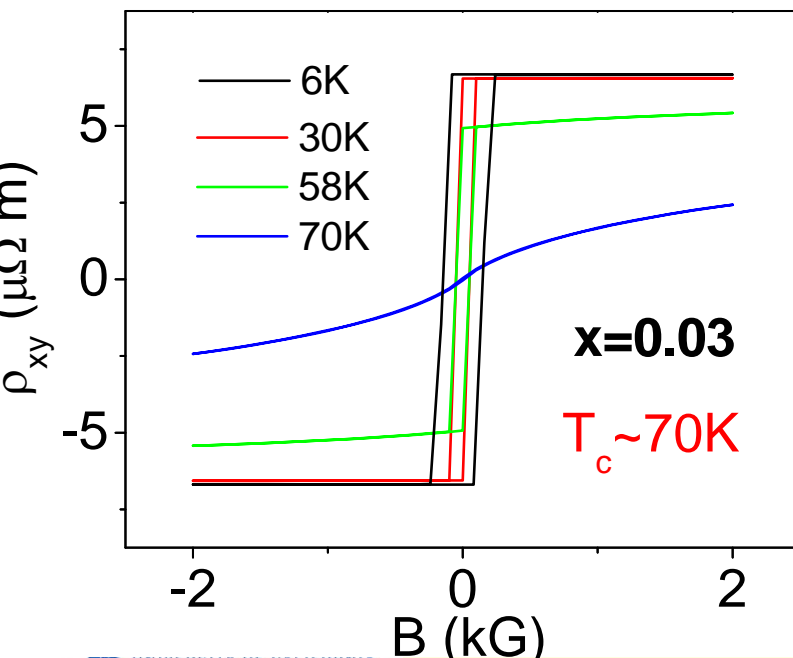
(Ga <sub>1-x</sub> Mn <sub>x</sub> )As 50 nm
(In <sub>y</sub> Ga <sub>1-y</sub> )As 500 nm
GaAs 50 nm
S. I. GaAs (001) sub.

InGaAs buffer layer → tensile strain  
→ perpendicular anisotropy

“B=0” is a special field

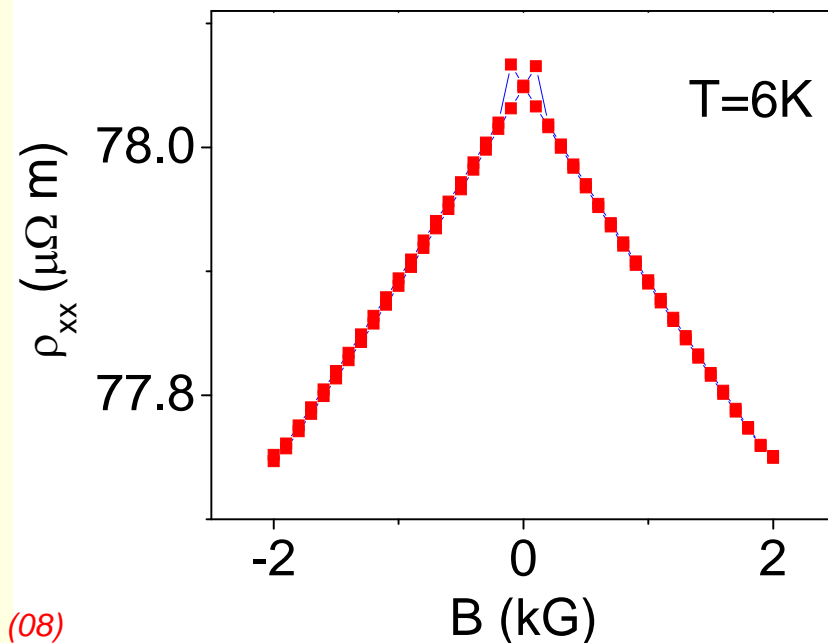
A set of films with different Mn, so different  $T_c$ 's.

*AHE (OHE is negligible)*



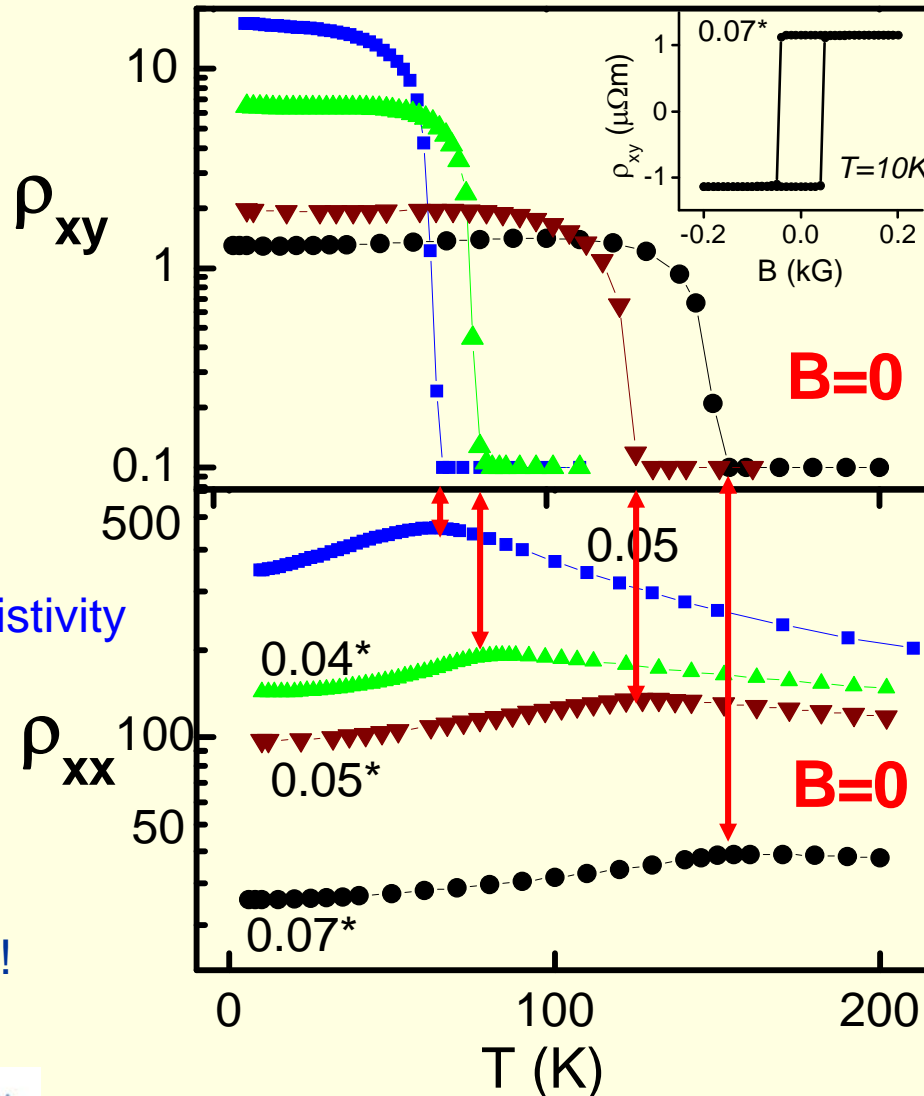
*Pu et al. PRL (08)*

*Longitudinal resistivity*



# AHE in GaMnAs

Hall resistivity



AHE remains positive below  $T_c$

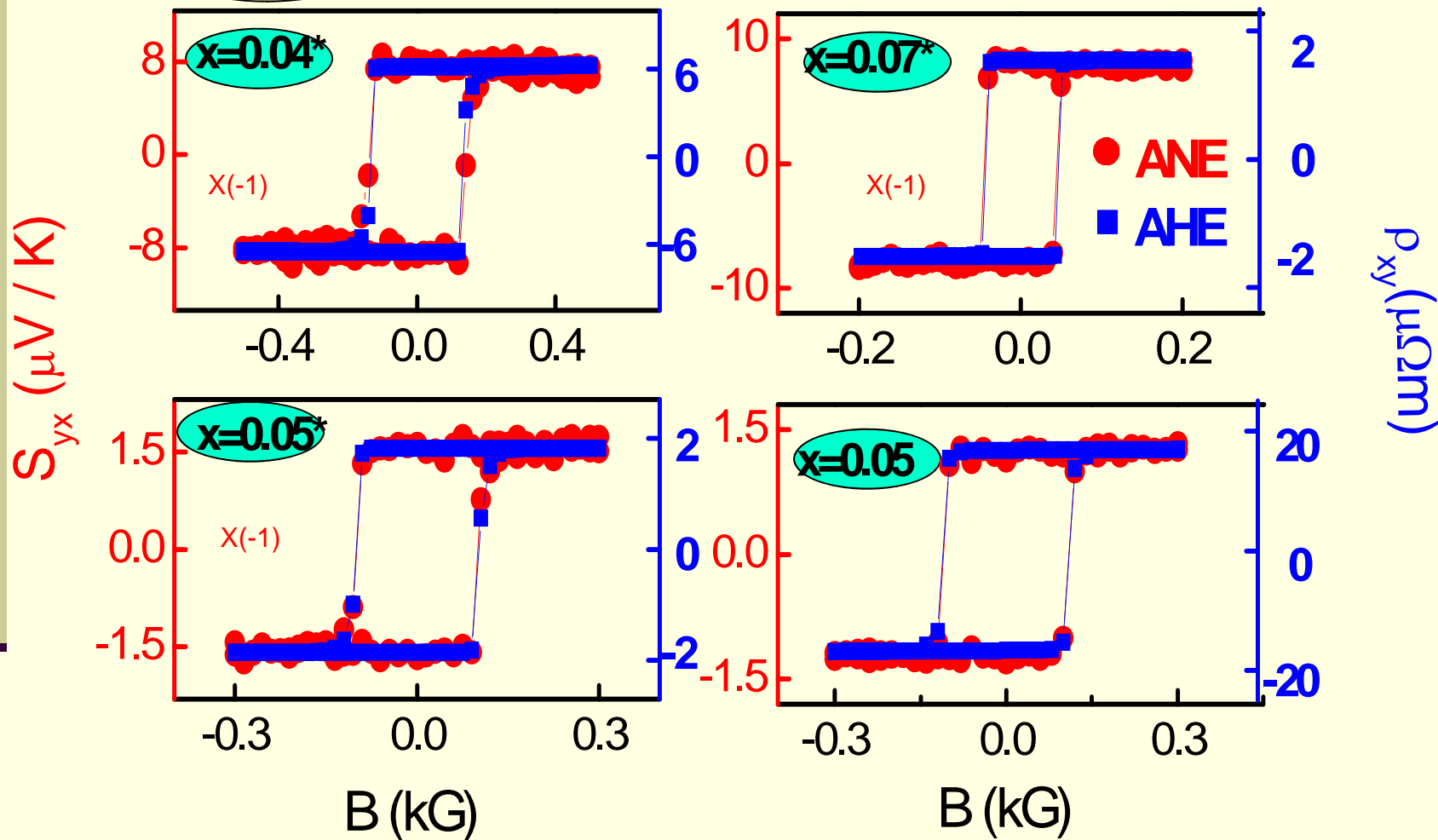
$x=0.04^*$ ,  
 $0.05^*$ , and  
 $0.07^*$  are  
annealed but  
 $x=0.05$  is not.

Usual power-law  
analysis is highly  
unreliable because  
of uncertainty in  
determining  $M$

All “metallic”!

# AHE & ANE in Different Samples

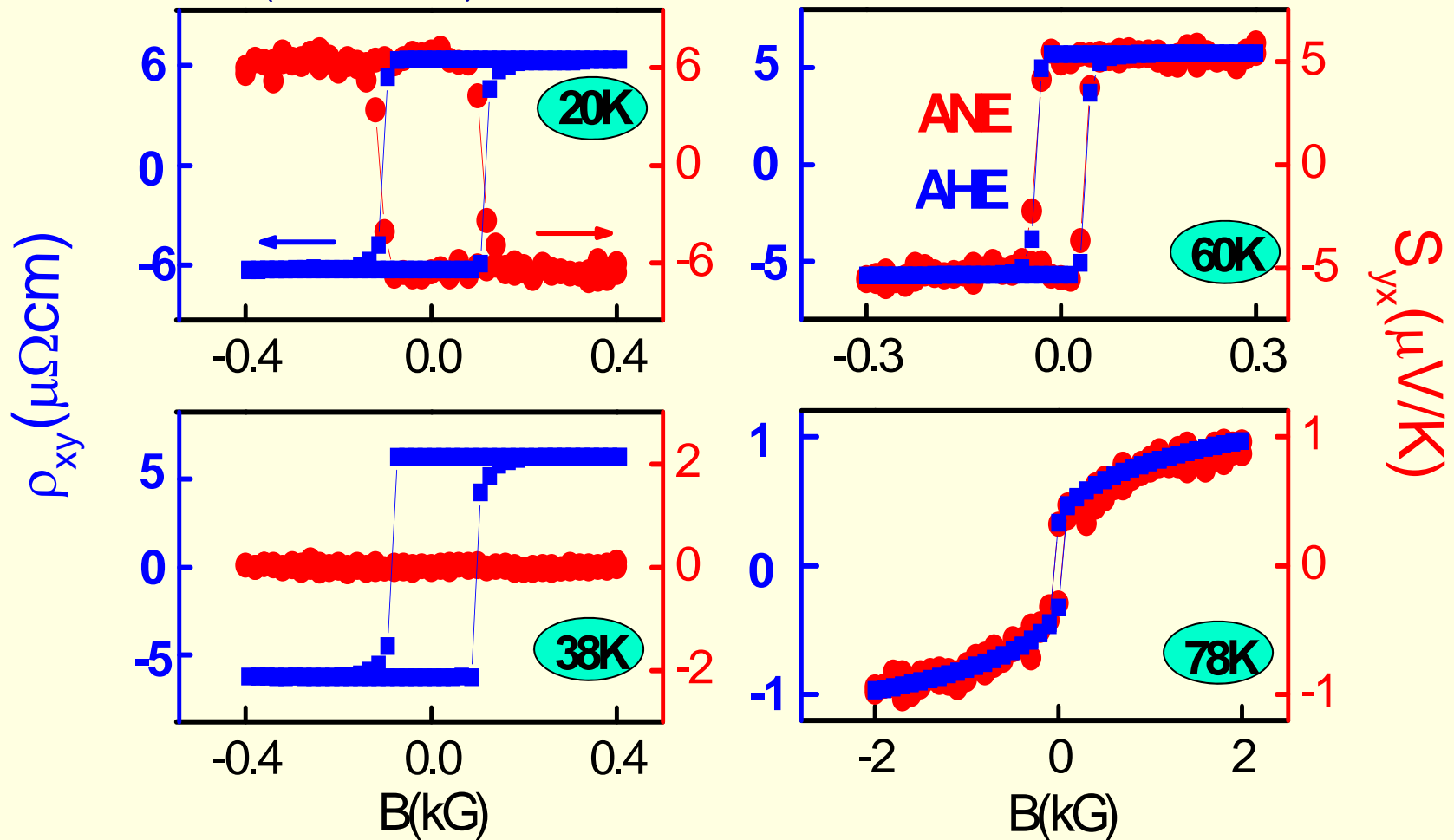
$T=10K$



No ordinary Nernst effect is visible;  $S_{yx}$  goes with  $\rho_{xy}$

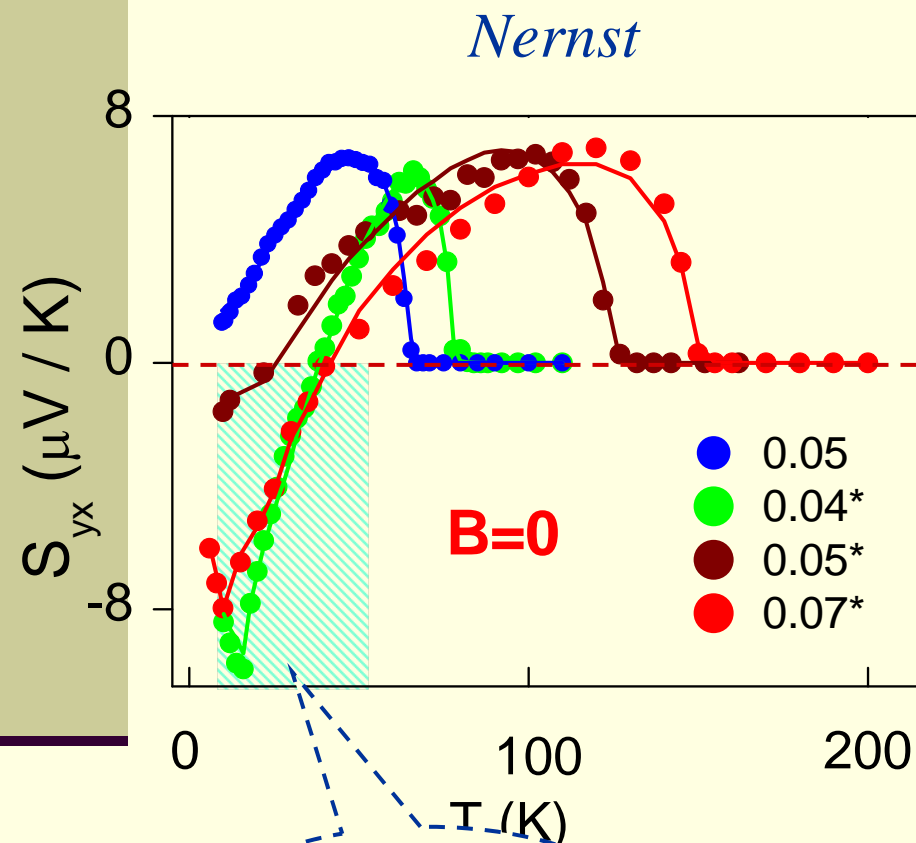
# AHE & ANE at Different Temperatures

■  $x=0.04^*$  (annealed)

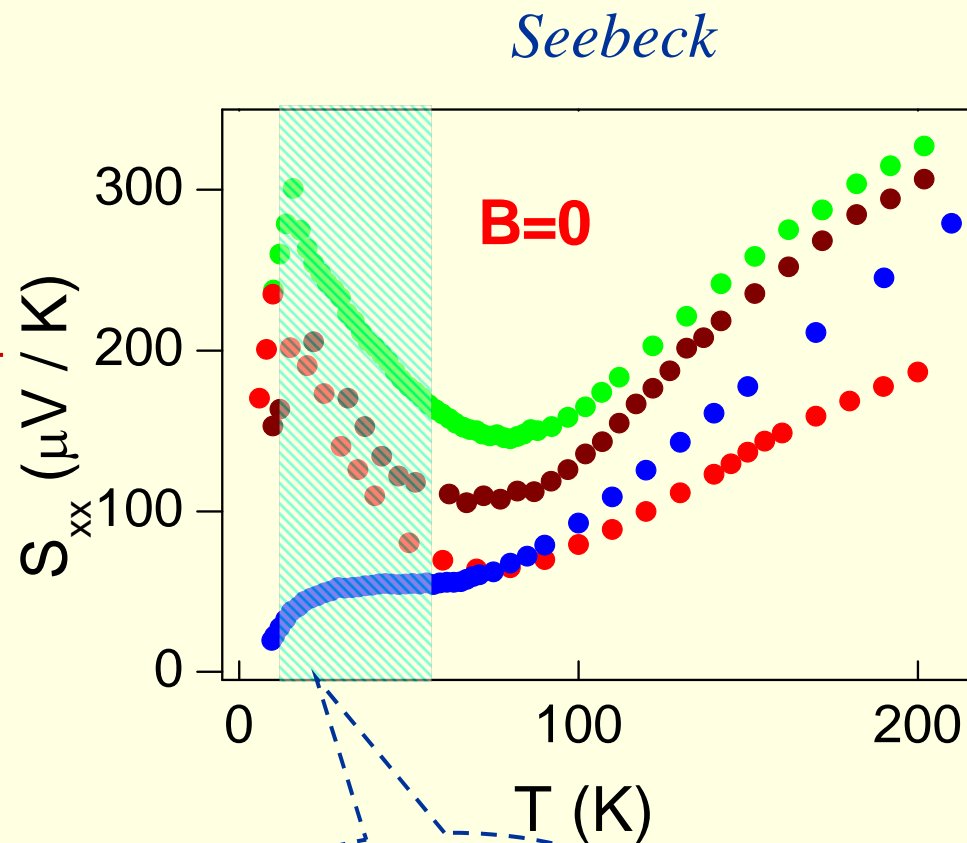


- Except for sign, AHE and ANE scale with each other  $\rightarrow$  share the same physical origin!

# Zero-Field $S_{xx}$ and $S_{yx}$



$S_{yx}$  changes sign!



$S_{xx}$  develops peak!

# Mott Relation

$$\rho_{xy} = \lambda M \rho_{xx}^n$$

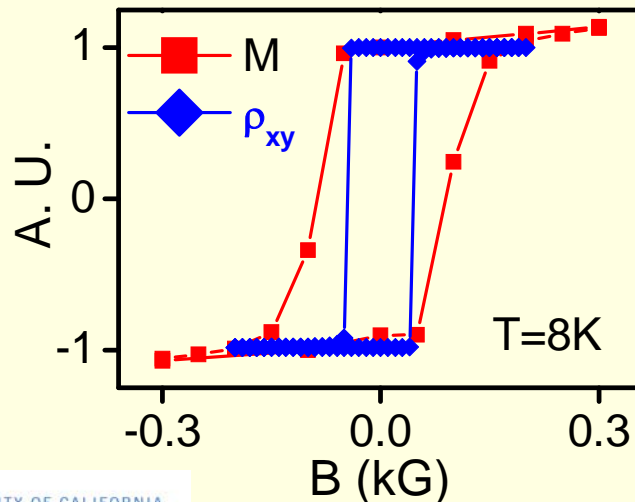
Mott relation  $\rightarrow$

$$S_{yx} \propto \left( \frac{\rho_{xy}}{\rho_{xx}} \right)' = \lambda M \rho_{xx}^{n-1} \left( T \frac{\pi^2 k_B^2}{3e} \frac{\lambda'}{\lambda} - (n-1) S_{xx} \right)$$

Replace M by  $\sigma_{xx}$  &  $\sigma_{xy}$  by reusing the power-law

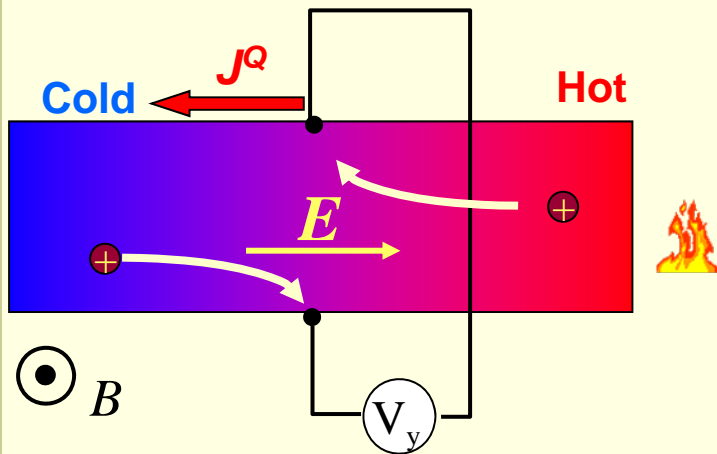
We have

$$S_{yx} = \frac{\rho_{xy}}{\rho_{xx}} \left( T \frac{\pi^2 k_B^2}{3e} \frac{\lambda'}{\lambda} - (n-1) S_{xx} \right)$$



- No longer need to measure M independently  $\rightarrow$  removing uncertainty in M measurements
- All transport coefficients are from exactly the same region (Hall bar cross)

# $S_{yx}$ Sign Change



Open circuit voltage ( $J_x$  and  $J_y=0$ )

Nernst current:

$$J_y = \alpha_{yx} (-\nabla T)_x$$

Nernst current Hall current

$$S_{yx} = \left( \frac{\sigma_{xy}}{\sigma_{xx}} \right)' = \frac{1}{\sigma_{xx}} (\alpha_{yx} - \sigma_{yx} S_{xx})$$

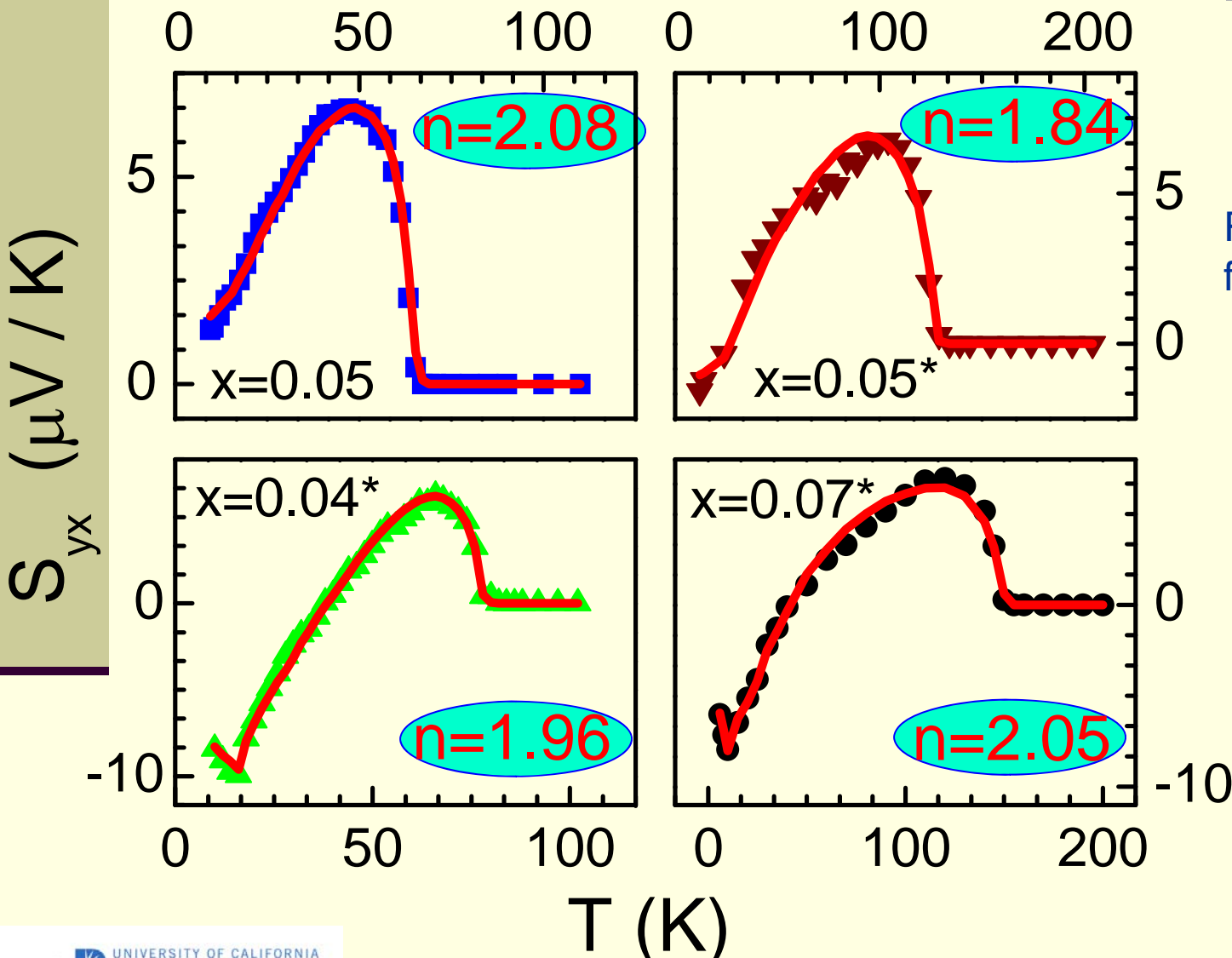
$$S_{yx} = \frac{\rho_{xy}}{\rho_{xx}} \left( T \frac{\pi^2 k_B^2}{3e} \frac{\lambda'}{\lambda} - (n-1) S_{xx} \right)$$

Nernst current Hall current

Sign change is only possible if  $n > 1$ ,  $\rightarrow$  NOT skew scattering!

At low-T where  $S_{xx}$  is large, Hall current exceeds Nernst current  $\rightarrow$  Sign Change!

# Exponent “n”



Red solid lines are fits to Mott relation

$$S_{yx} = \frac{\rho_{xy}}{\rho_{xx}} \left( T \frac{\pi^2 k_B^2}{3e} \frac{\lambda'}{\lambda} - (n-1) S_{xx} \right)$$

Mott relation works well for  $n=2$ !

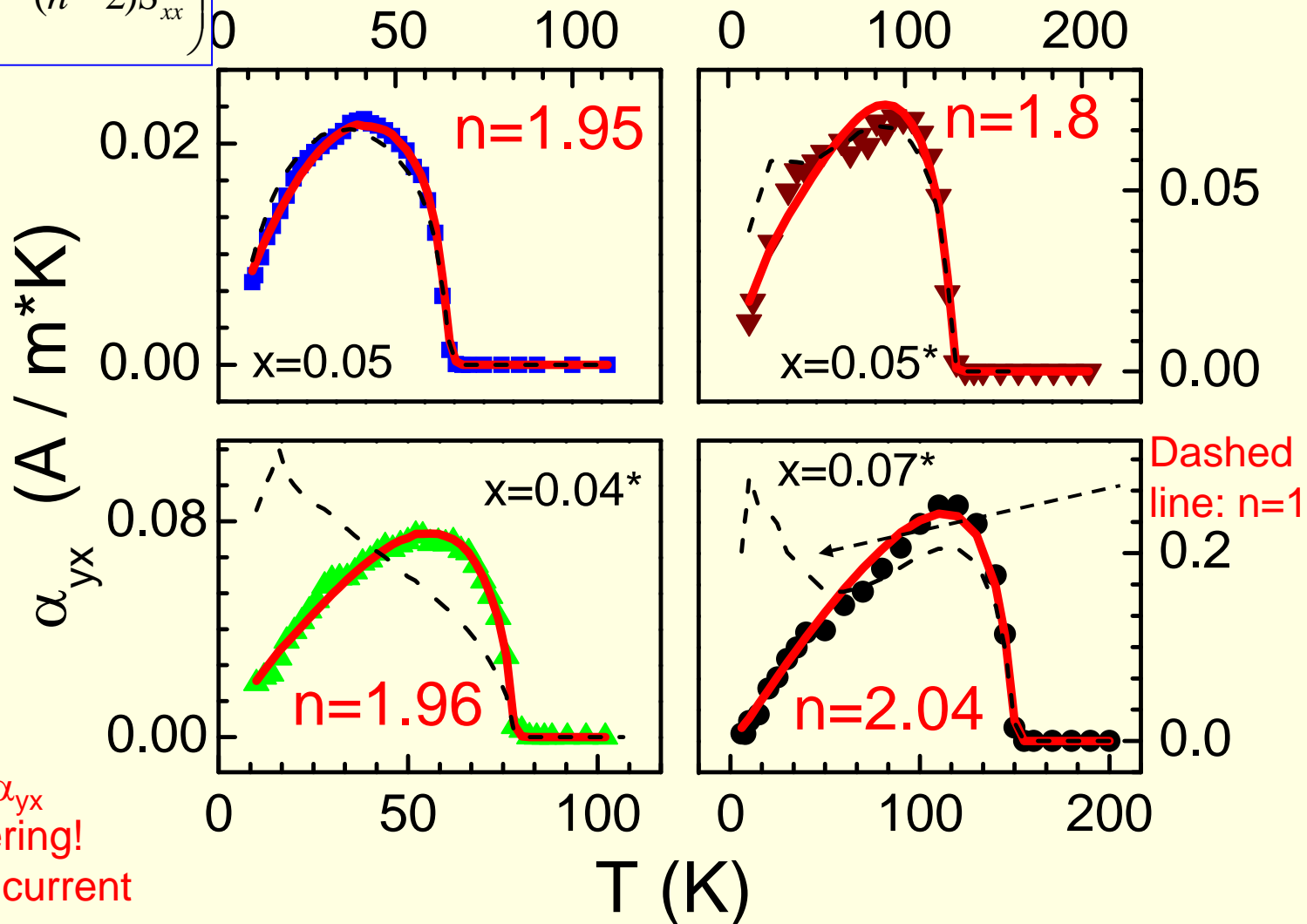


# Nernst Current: $J_N$

$$\alpha_{yx} = \frac{\rho_{xy}}{\rho_{xx}^2} \left( T \frac{\pi^2 k_B^2}{3e} \frac{\lambda'}{\lambda} - (n-2) S_{xx} \right)$$

Nernst current:

$$J_N = -\alpha_{yx} (\nabla T)_x$$



$n=2 \rightarrow$  nothing in  $\alpha_{yx}$  depends on scattering!  
 $\rightarrow$  intrinsic Nernst current

# Intrinsic vs. Side Jump

- $n=2 \rightarrow$  intrinsic or side jump (SJ)

In GaMnAs,

- ❖ Intrinsic mechanism can account for most of AHE magnitude
- ❖ SJ displacement  $\Delta y < 0.1$  nm, and MFP is about  $\sim 20$  nm.  
Hall angle  $\Theta_H < 0.05$  for SJ; observed Hall angle  $\Theta_H \sim 0.1$
- ❖ AHE/ANE is likely dominated by intrinsic mechanism

Mott relation holds for AHE/ANE with intrinsic (Berry's phase) mechanism.

# Meaning of $\lambda / \lambda'$

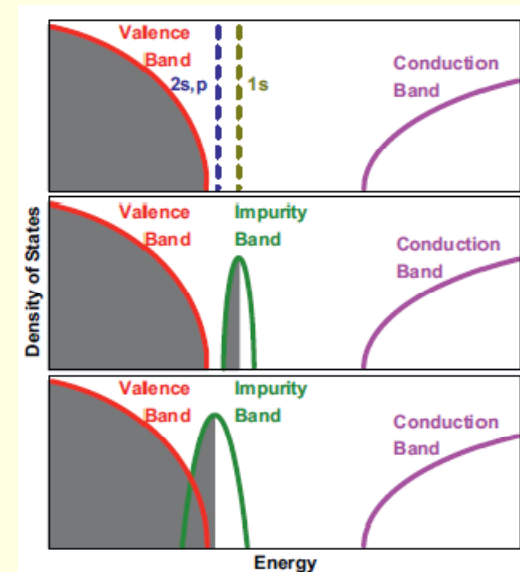
- $\lambda$  depends on Fermi energy
- We obtain the value of  $\lambda / \lambda'$

	#4*	#5	#5*	#7*
<b>n</b>	1.96	1.95	1.8	2.04
$\lambda / \lambda' (\text{eV})$	0.046	0.049	0.040	0.096

For small  $\varepsilon_F$ , we assume  $\lambda \propto \varepsilon_F^\alpha$

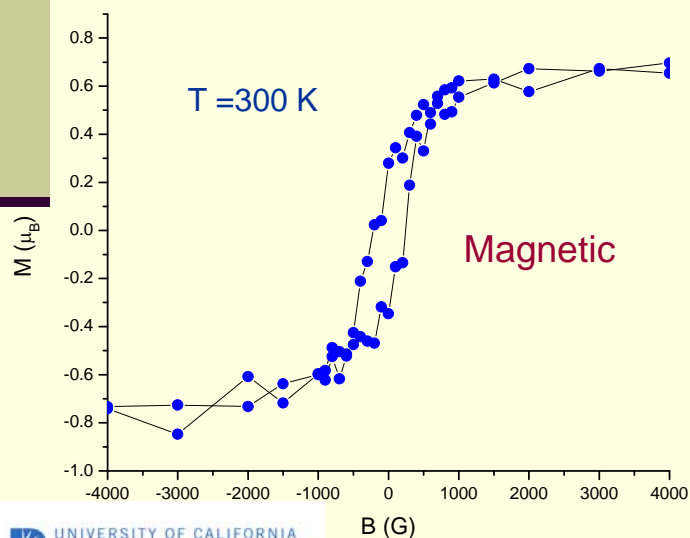
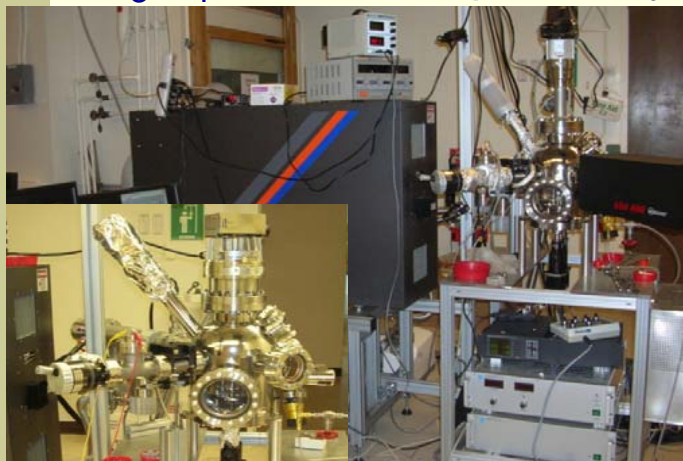
$$\frac{\lambda}{\lambda'} = \frac{\varepsilon_F}{\alpha} \approx 0.05 - 0.1 \text{ eV}$$

For  $p \sim 10^{19} \text{ cm}^{-3}$ , obtained  $\varepsilon_F$  is an order smaller than expected from valence band, suggesting the impurity band picture.

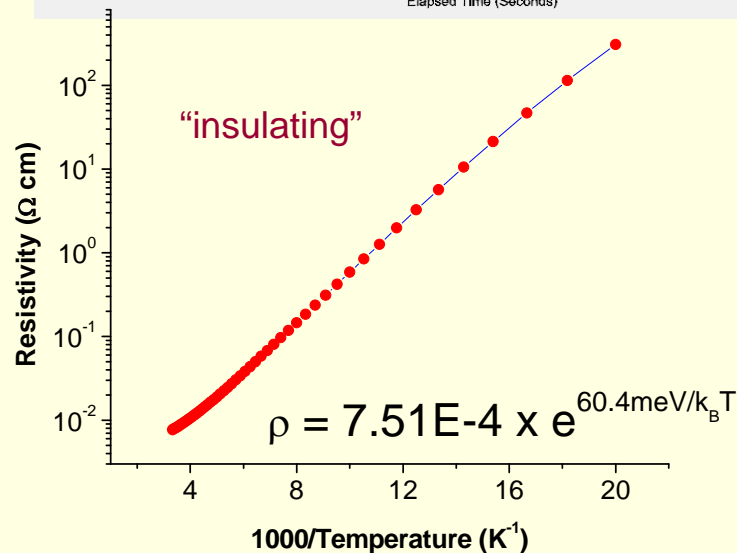
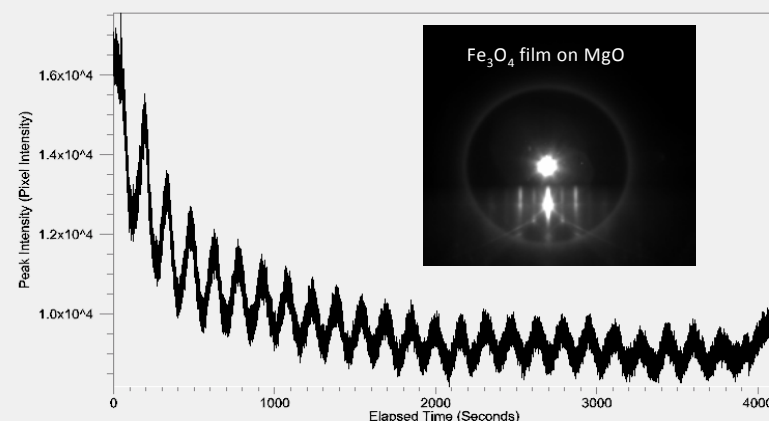


# Magnetic Insulator Films: $\text{Fe}_3\text{O}_4/\text{MgO}$ (001)

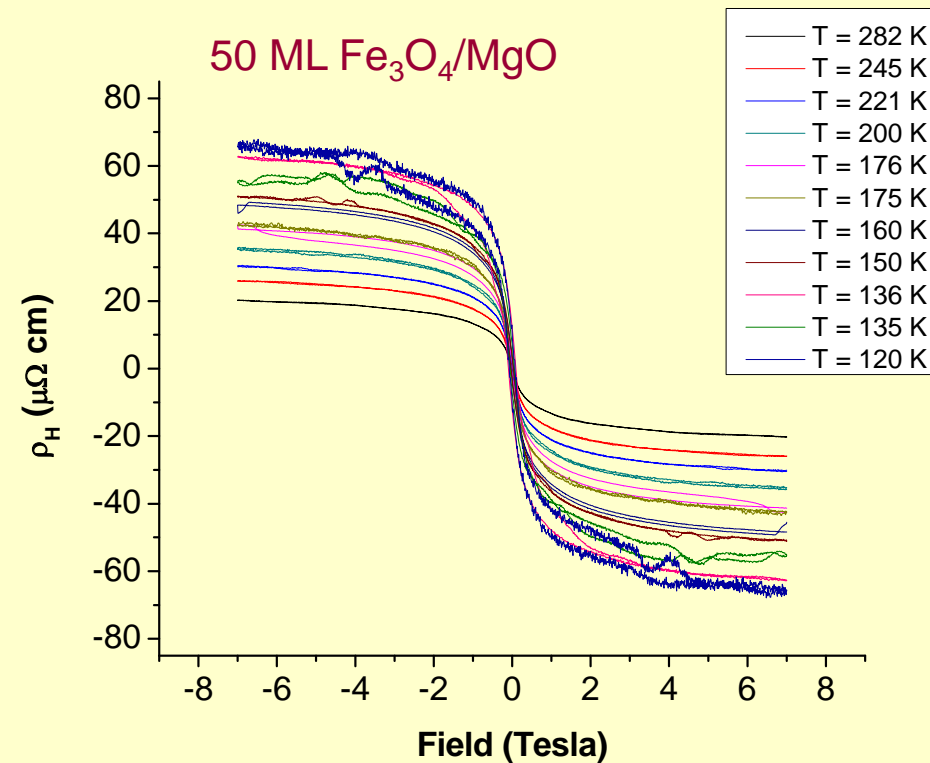
$\text{Fe}_3\text{O}_4$  films are epitaxially grown with laser MBE



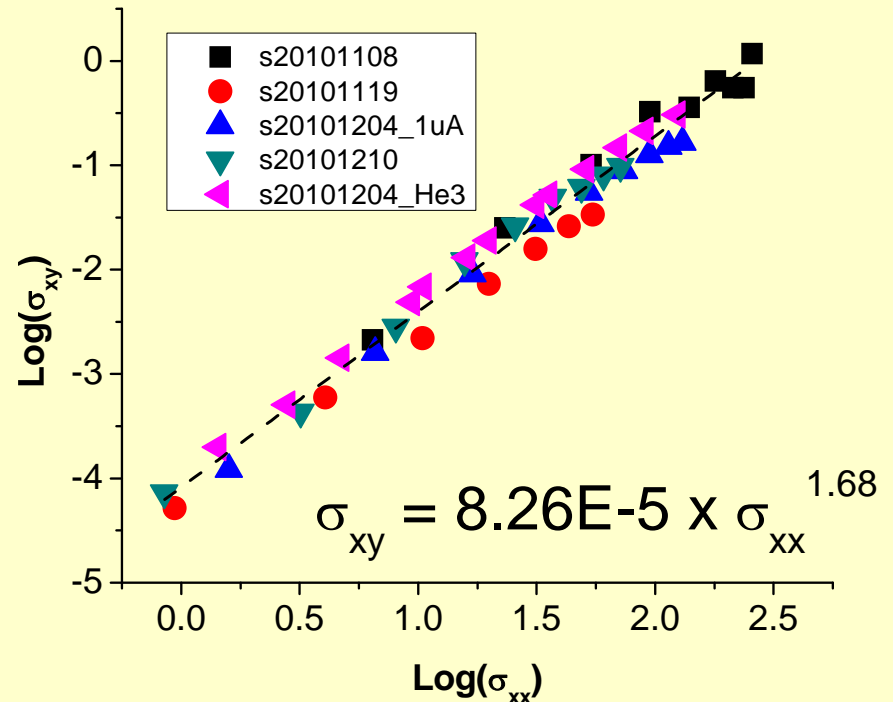
RHEED pattern and oscillations



# Power-Law Scaling

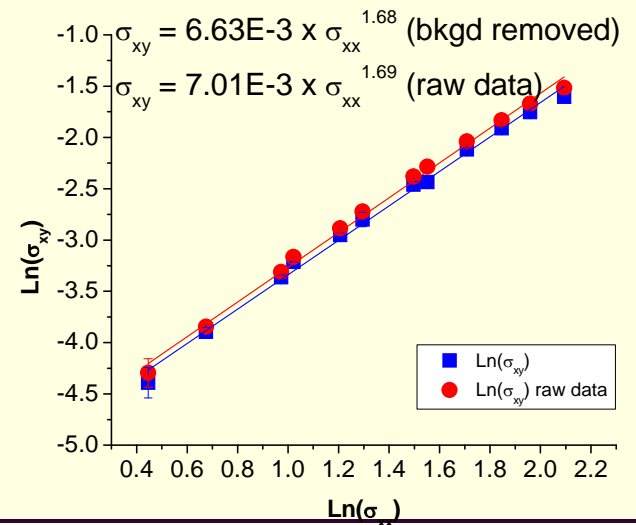
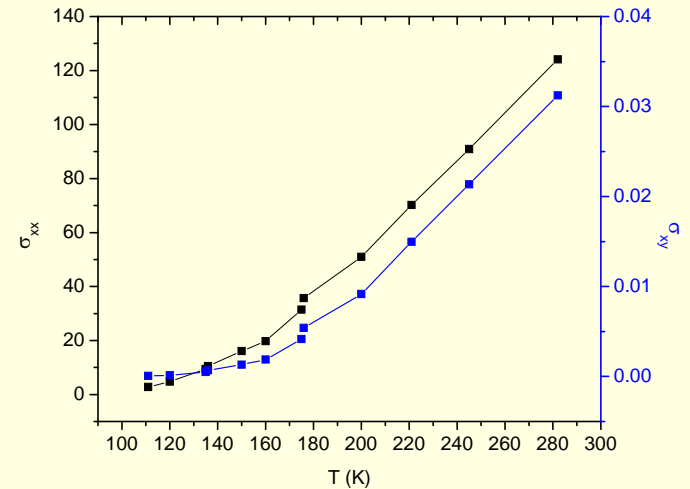
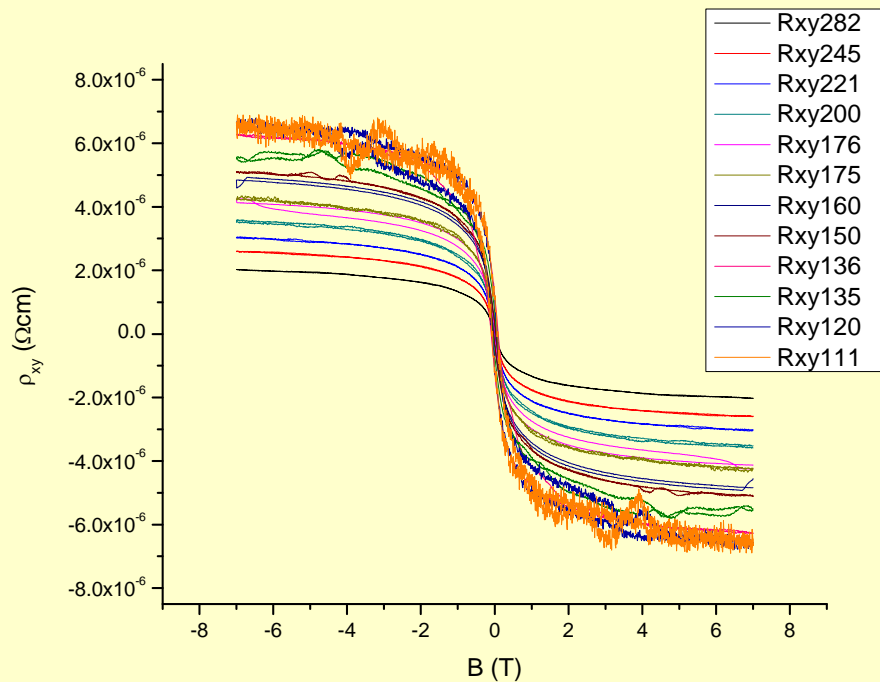


AHE signal above Verwey temperature



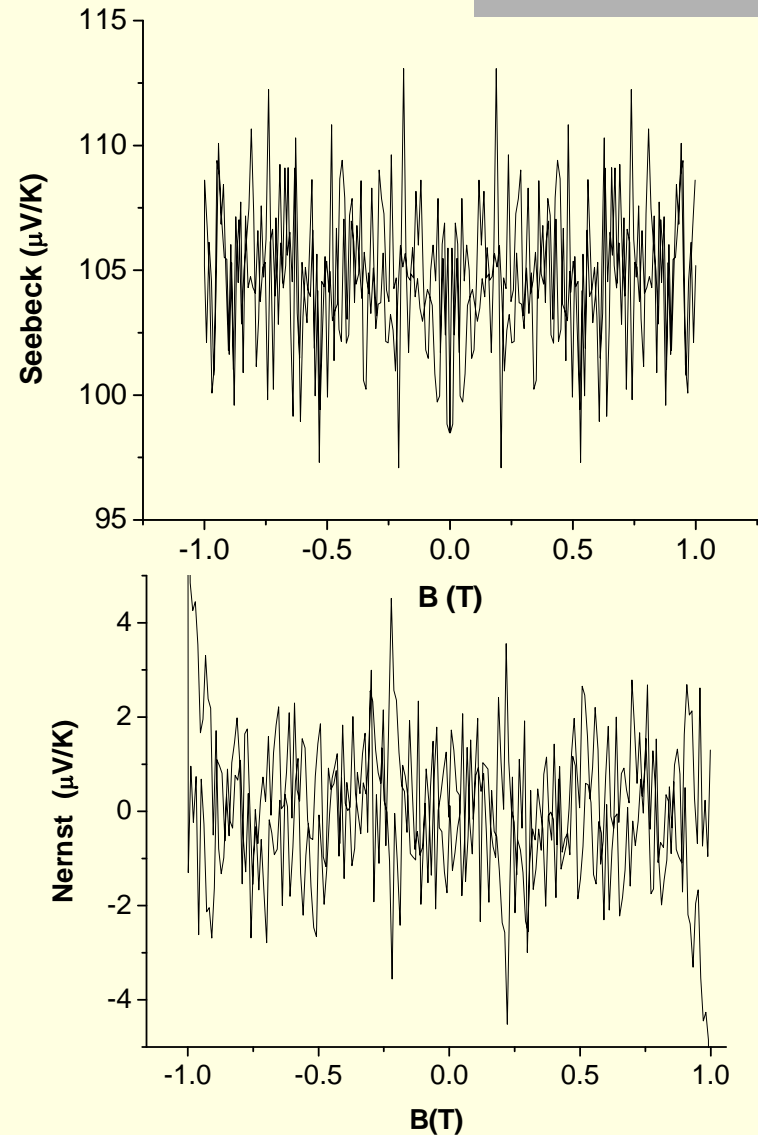
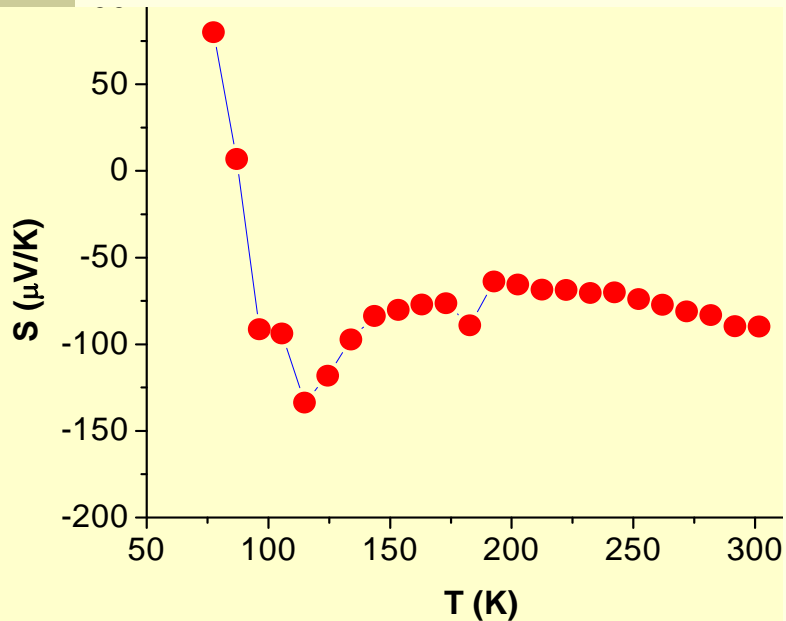
A much smaller  $n=0.32$ !  
Consistent with other reported value.

# AHE and Power-Law



# Thermoelectric Measurements

Zero-field Seebeck



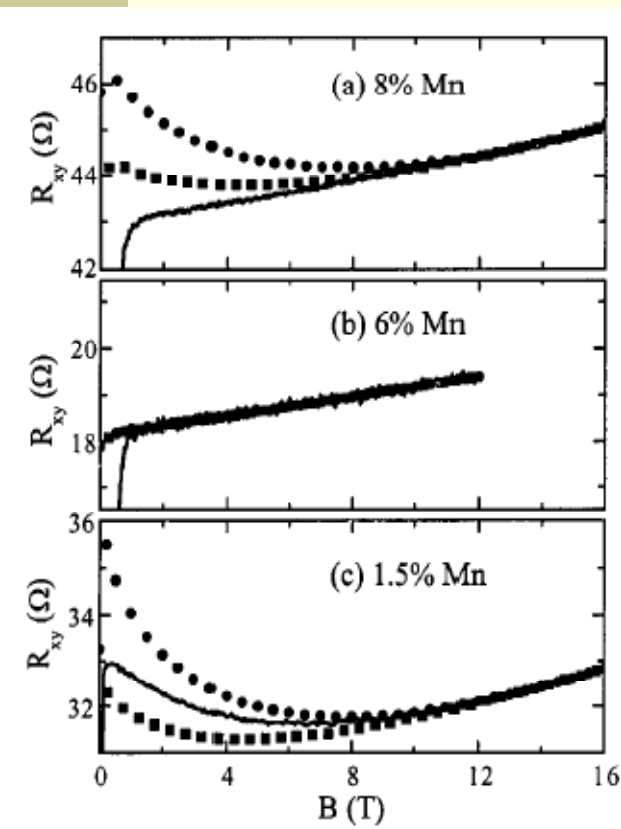
# Summary

- Large ANE is observed in GaMnAs [ $\lambda=\lambda(\varepsilon_F)$ ].
- AHE and ANE share the same physical origin.
- “n=2” is obtained from zero-field AHE and ANE without uncertainty in magnetization measurements.
- Our results suggest intrinsic Nernst current  $J_N$ .
- Mott relation is experimentally validated for scattering rate-independent anomalous transport.
- Small magnitude of  $\lambda/\lambda'$  suggests impurity band picture.
- “n=0.3” is found in epitaxial  $\text{Fe}_3\text{O}_4$  films, but ANE has not been observed.

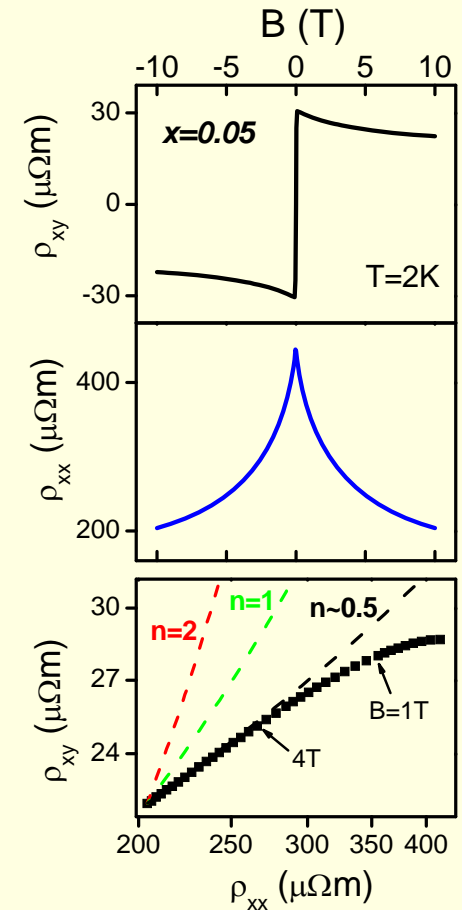
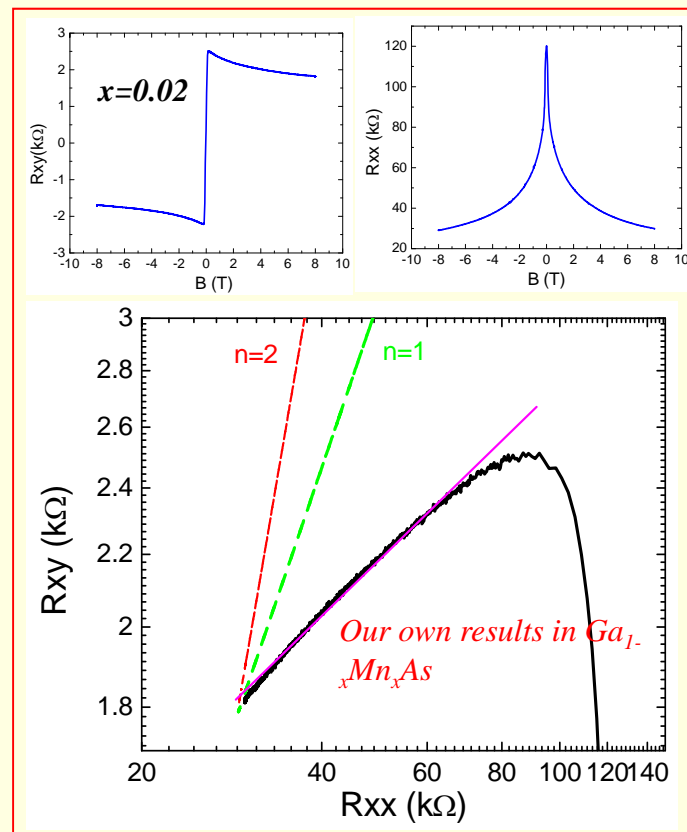


# Discussion: 1. Effect of High B-Field

In most experiments with in-plane anisotropy samples, high magnetic fields are used, but the effect of B-field on the power-law scaling is unclear. Which field is the proper one?

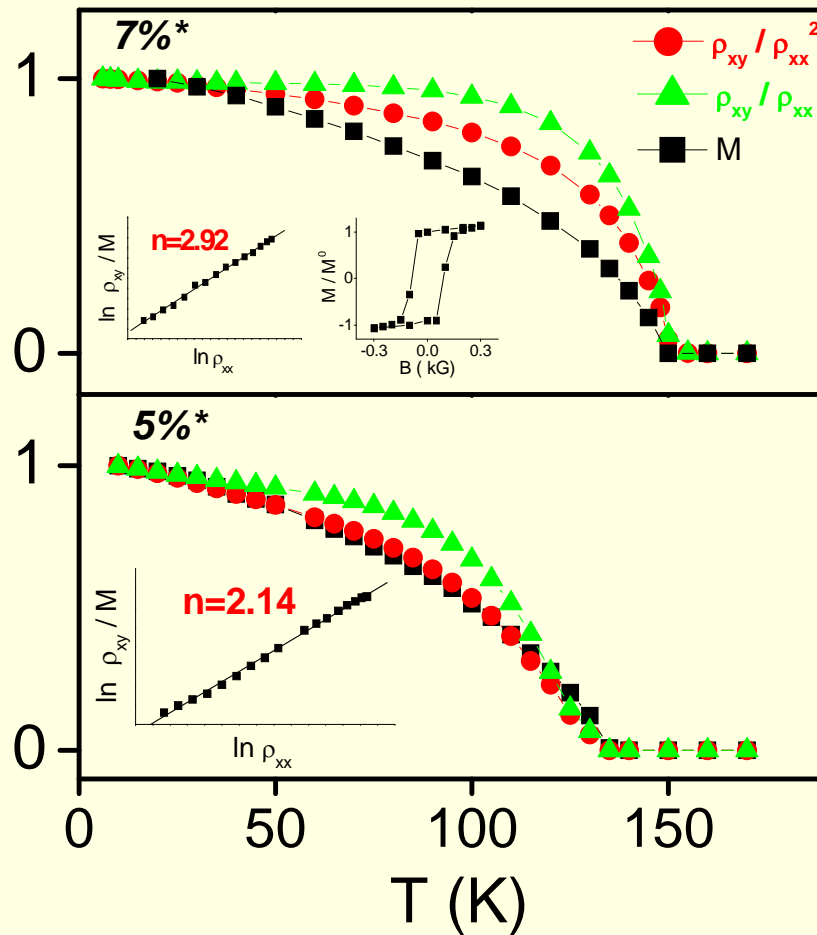


Edmonds et al. JAP (03)



# Find Exponent n

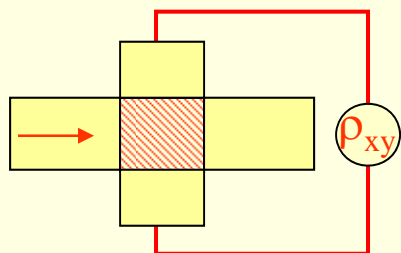
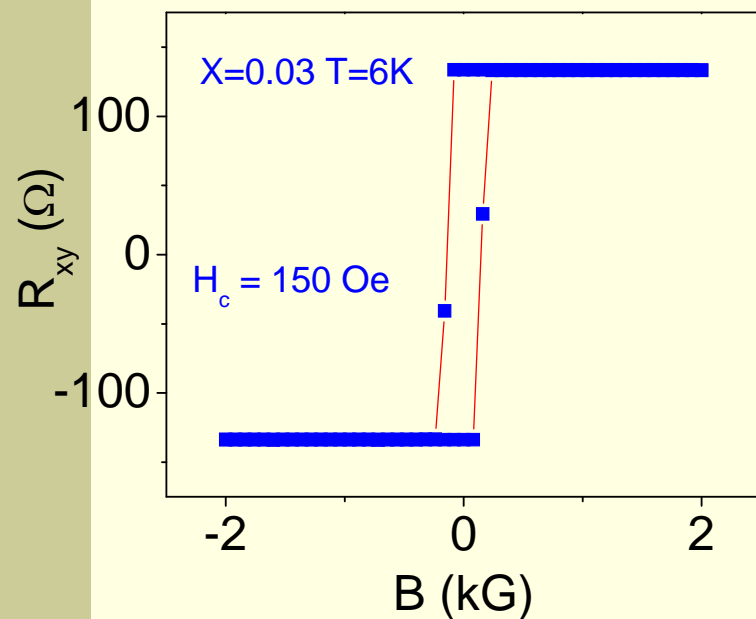
A.U.



- Exponent is not always one or two
- Separate magnetization measurement is needed
- Hall loops are different from MH loops

# $R_{xy}$ and M Measurements

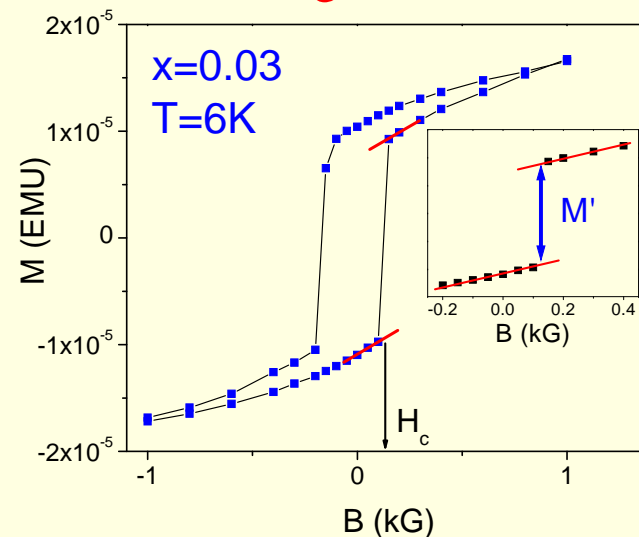
Transport



Probing area  $\sim (100 \mu\text{m})^2$

Magnetization

Good



Bad

