# Do we understand (Ga,Mn)As?: prospects for high temperature ferromagnetism in (Ga,Mn)As semiconductors

Jairo Sinova
Texas A &M University

**References: Jungwirth et al Phys. Rev. B 72, 165204 (2005) and Jungwirth et al, *Theory of ferromagnetic (III,Mn)V semiconductors,* to appear in Rev. of Mod. Phys. (2006).**

KITP, May 25th 2005

Support:

# THE TEAM

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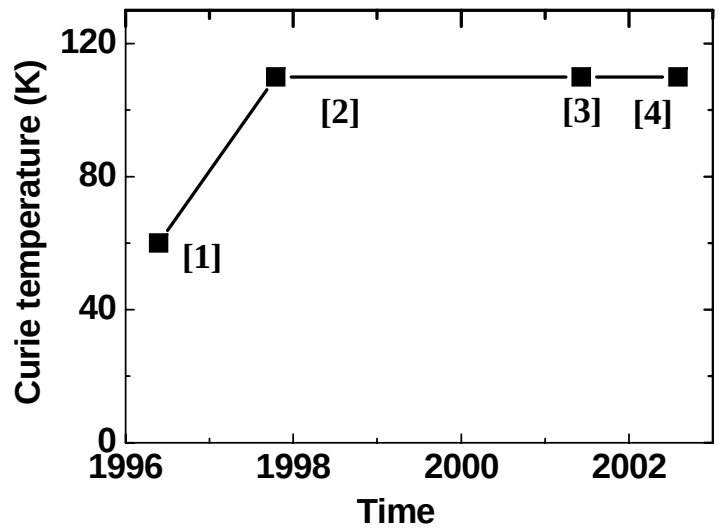
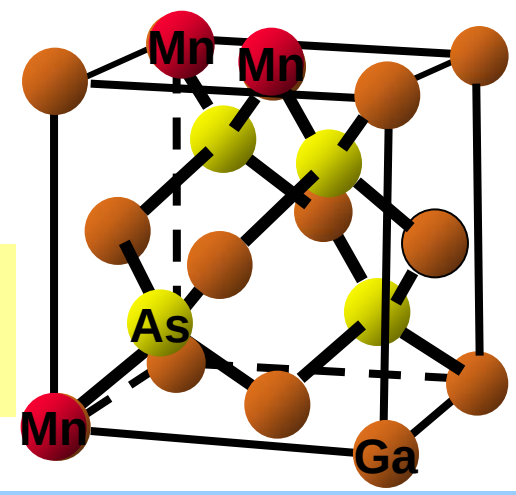
# OUTLINE

- DMS: intro to the phenomenology
  - Possible stumbling blocks to high Tc
  - Theoretical approaches to DMSs
- What is theory telling us about Tc trends
  - Is there an intrinsic limitation
  - Extrinsic limitations
- What is the data telling us: thumbs up or down?
- Other successful descriptions of system properties
  - Magnetic anisotropy
  - Temperature dependence of transport in metallic samples
  - Magnetization dynamics
  - Domain wall dynamics and resistances
  - Anisotropic magnetoresistance
  - TAMR
  - Anomalous Hall effect
- Remaining challenges:
  - Red shift in IR absorption peak
  - Seemingly large effective masses

# Problems for GaMnAs (late 2002)

- Curie temperature limited to ~110K.
- Only metallic for ~3% to 6% Mn
- High degree of compensation
- Unusual magnetization (temperature dep.)
- Significant magnetization deficit



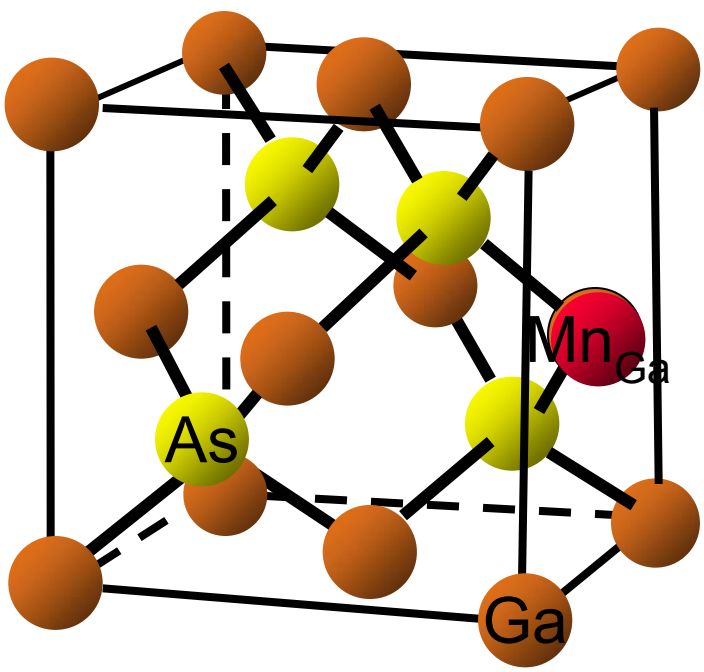




**"110K could be a *fundamental limit* on $T_C$"**

**But are these intrinsic properties of GaMnAs ??**

# (Ga,Mn)As diluted magnetic semiconductor

**Low-T MBE - random but nearly uniform Mn distribution up to ~ 10% doping**




GALLIUM 69.72

5.91 Ga 31

[Ar] $3d^{10}4s^2 3p^1$

4.51 ORC 1.695 1.001

303 240

MANGANESE 54.938

7.43 Mn 25

[Ar] $3d^5 4s^2$

8.89 CUB

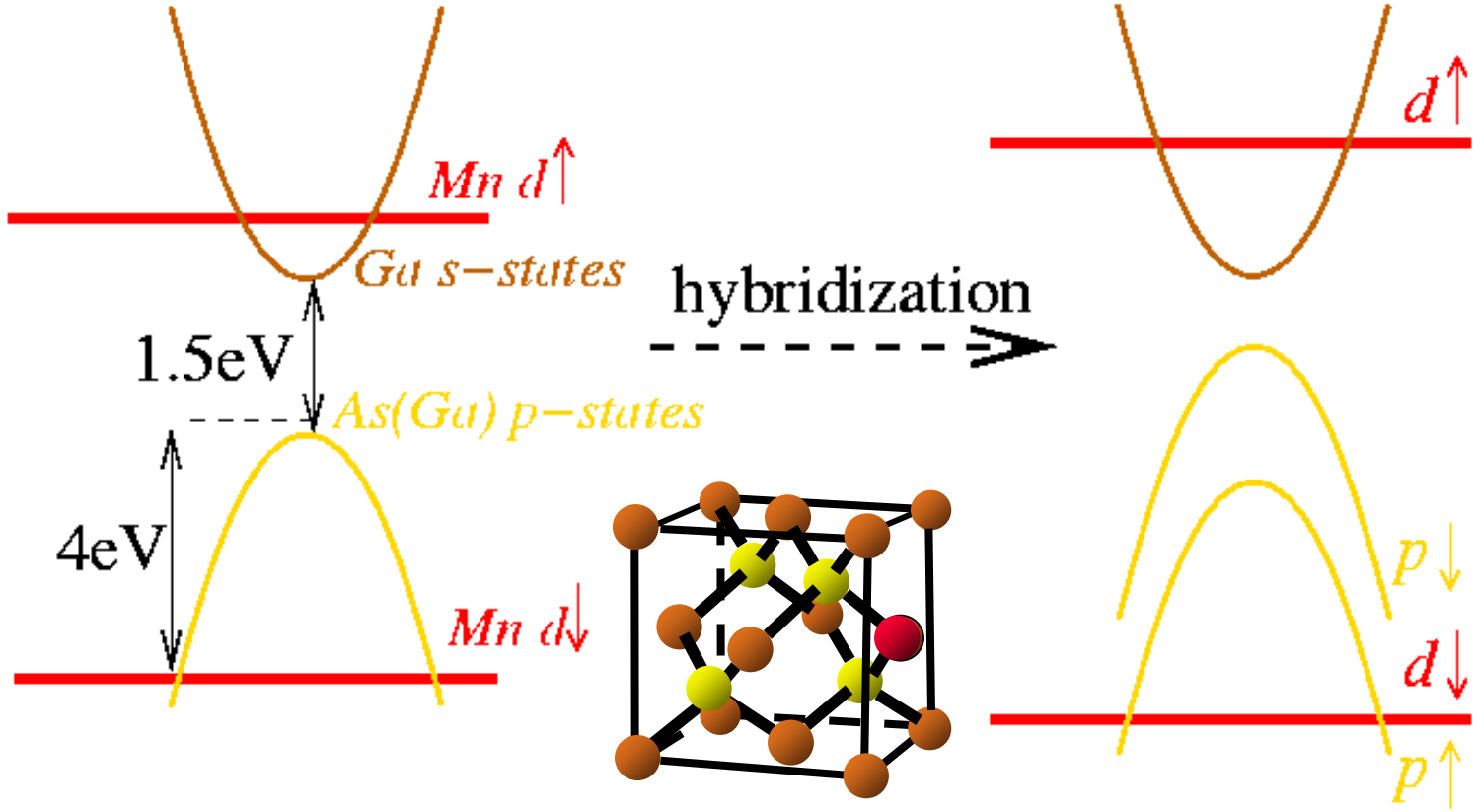
1518 400

***5 d*-electrons with L=0, S=5/2**

**moderately shallow acceptor (110 meV)**

# Theoretical descriptions

**Microscopic: atomic orbitals & Coulomb correlation of *d*-electrons & hopping**




**Effective magnetic coupling:** $J_{pd}\, S_{Mn} \cdot s_{hole}$

**Coulomb correlation of *d*-electrons & hopping →AF kinetic-exchange coupling**

# Which theory is right? High noon at KITP:

**Impurity bandit vs Valence Joe**

**KP Eastwood**

**Fast principles Jack**

# Theoretical Approaches to DMSs

- First Principles LSDA

PROS: No initial assumptions, effective Heisenberg model can be extracted, good for determining chemical trends

CONS: Size limitation, difficulty dealing with long range interactions, lack of quantitative predictability, neglects SO coupling (usually)

- Microscopic TB models

PROS: "Unbiased" microscopic approach, correct capture of band structure and hybridization, treats disorder microscopically (combined with CPA), very good agreement with LDA+U calculations

CONS: neglects coulomb interaction effects, difficult to capture non-tabulated chemical trends, hard to reach large system sizes

- k.p $\oplus$ Local Moment

PROS: simplicity of description, lots of computational ability, SO coupling can be incorporated,
CONS: applicable only for metallic weakly hybridized systems (e.g. optimally doped GaMnAs), over simplicity (e.g. constant Jpd), no good for deep impurity levels (e.g. GaMnN)

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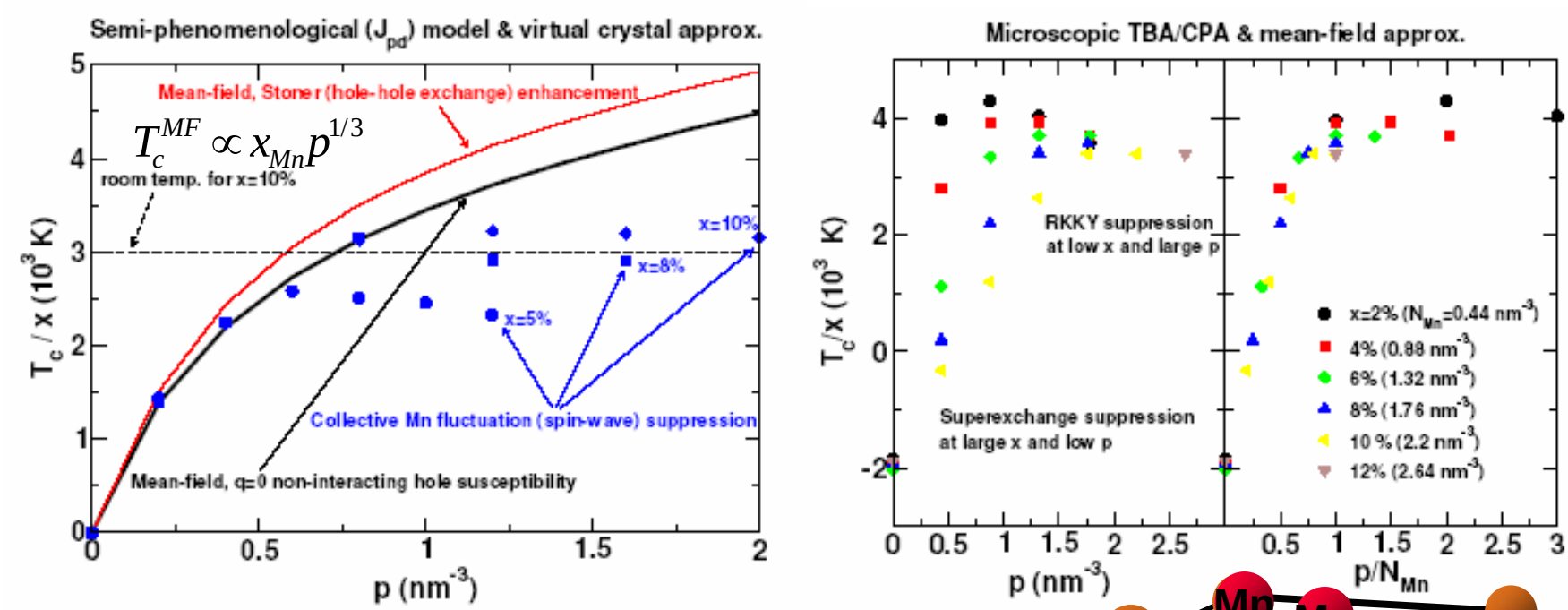
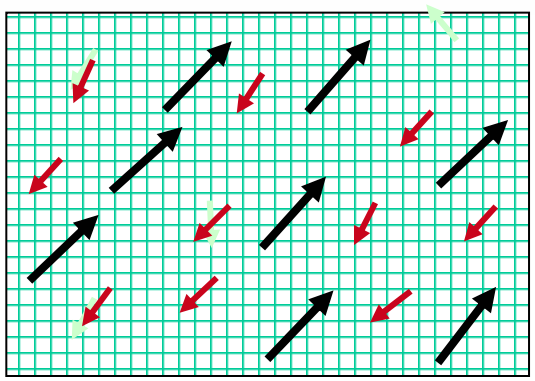
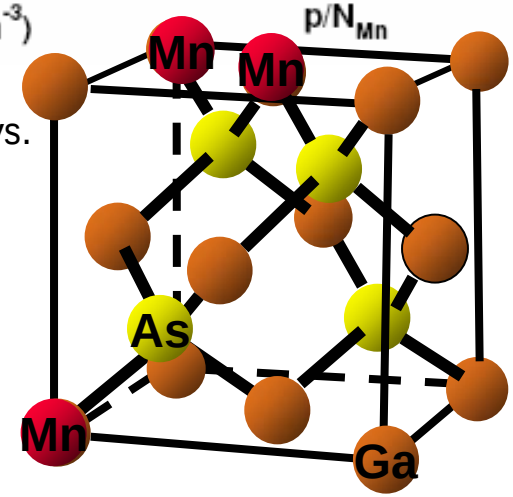


Jungwirth, Wang, et al. Phys. Rev. B 72, 165204 (2005)



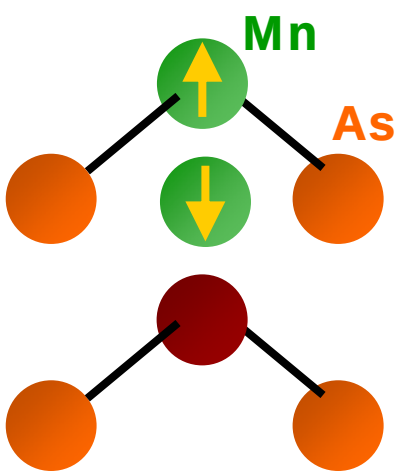

**Intrinsic properties of (Ga,Mn)As**: **$T_c$ linear in $Mn_{Ga}$ local moment concentration; falls rapidly with decreasing hole density in more than 50% compensated samples; nearly independent of hole density for compensation < 50%.**

# Extrinsic effects: Interstitial Mn - a magnetism killer

## Interstitial Mn is detrimental to magnetic order:

- **compensating double-donor – reduces carrier density**
- **couples antiferromagnetically to substitutional Mn even in low compensation samples**

***Blinowski PRB '03, Mašek, Máca PRB '03***

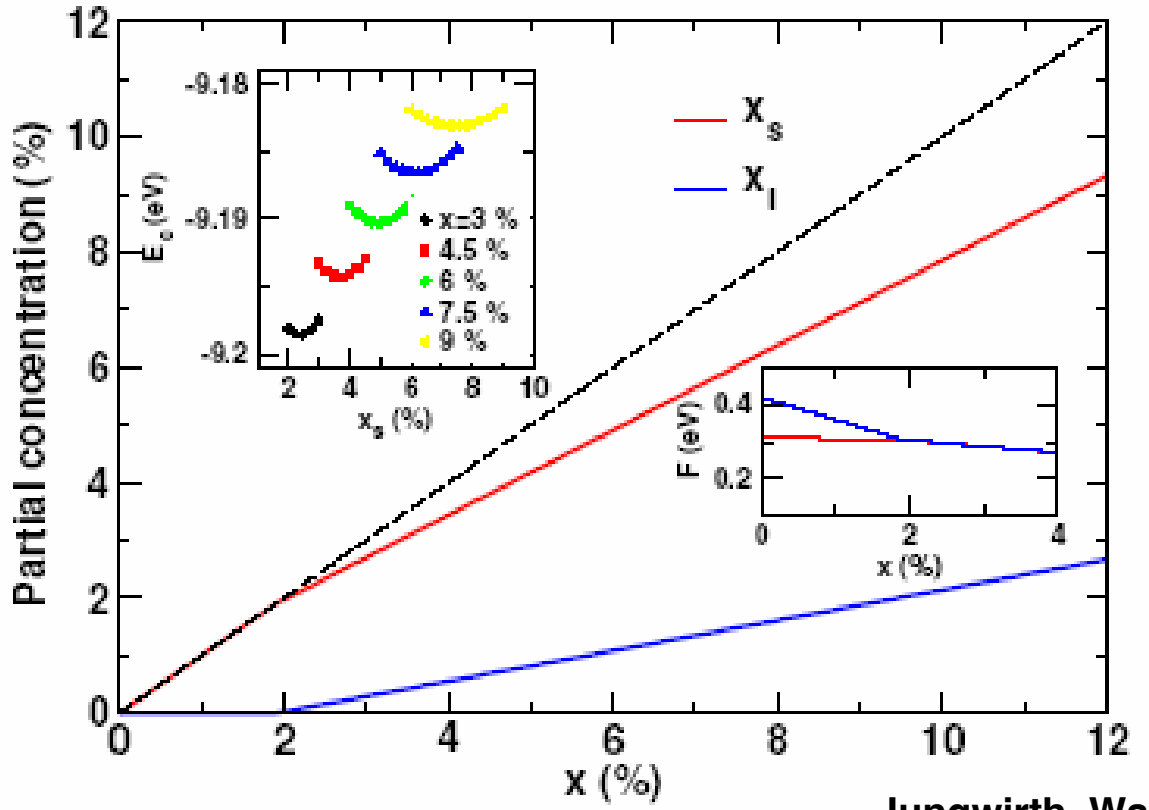



*Yu et al., PRB '02*:

**~10-20% of total Mn concentration is incorporated as interstitials**

**Increased $T_C$ on annealing corresponds to removal of these defects.**

# $Mn_{Ga}$ and $Mn_I$ partial concentrations




As grown Materials calculation

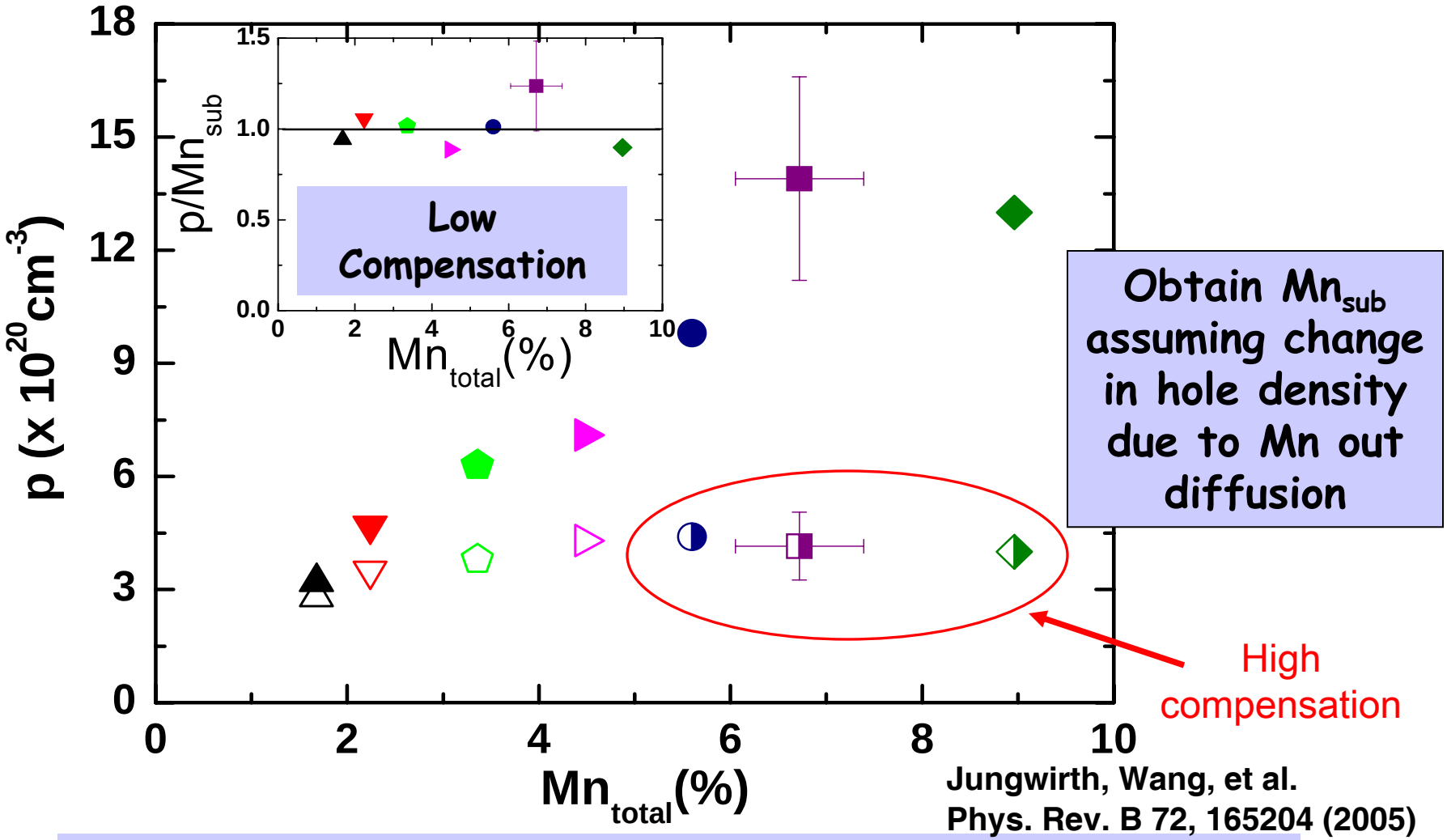
**Jungwirth, Wang, et al.**
**Phys. Rev. B 72, 165204 (2005)**

## Microscopic defect formation energy calculations:

**No signs of saturation in the dependence of $Mn_{Ga}$ concentration on total Mn doping**

## Experimental hole densities: measured by ordinary Hall effect

**Open symbols & half closed as grown. Closed symbols annealed**



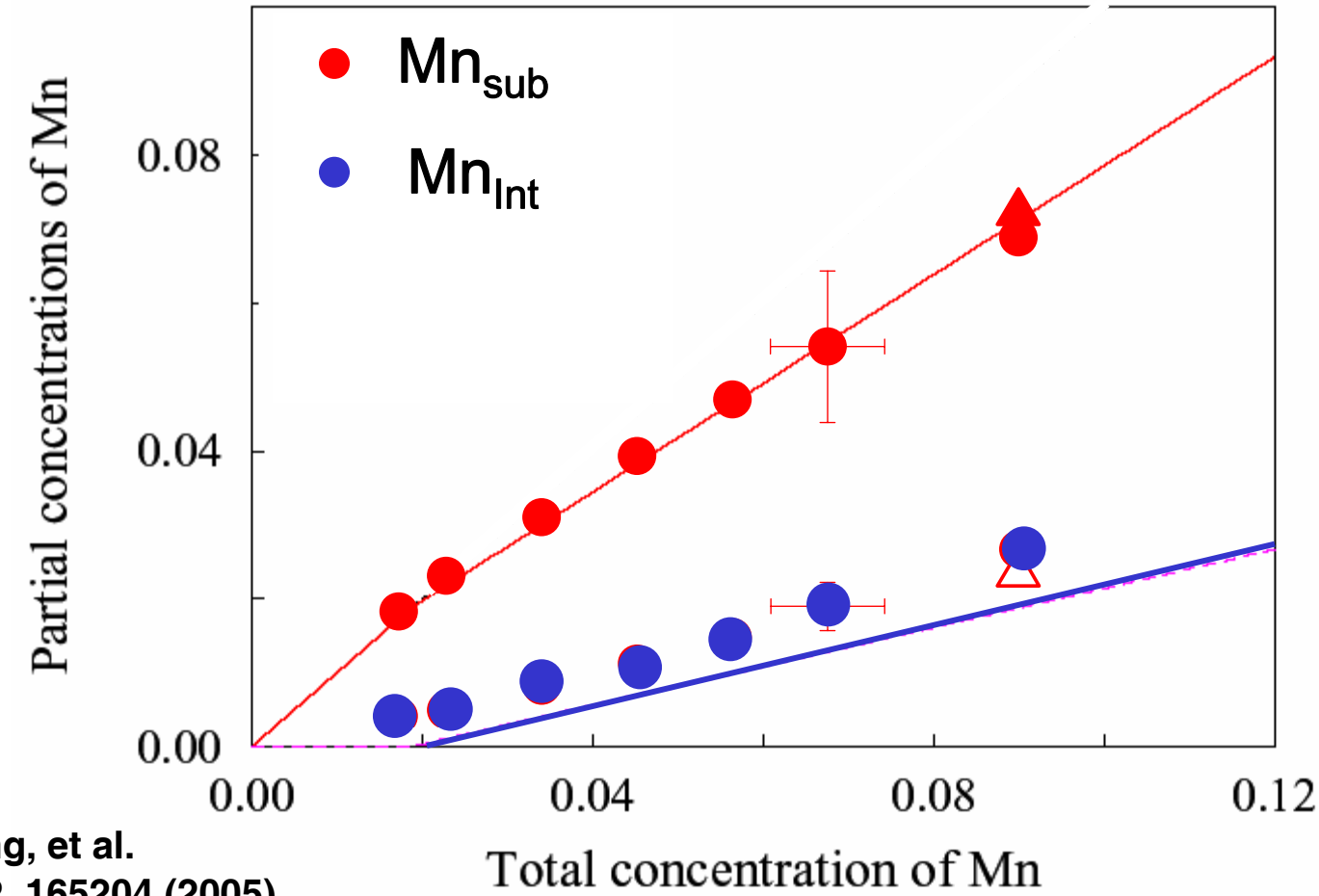

Jungwirth, Wang, et al.
Phys. Rev. B 72, 165204 (2005)

Annealing can very significantly increases hole densities.

## Experimental partial concentrations of MnGa and MnI in as grown samples

**Theoretical linear dependence of $Mn_{sub}$ on total Mn confirmed experimentally**

Obtain $Mn_{sub}$ & $Mn_{Int}$ assuming change in hole density due to Mn out diffusion




Jungwirth, Wang, et al.
Phys. Rev. B 72, 165204 (2005)

SIMS: measures total Mn concentration.
Interstitials only compensation assumed

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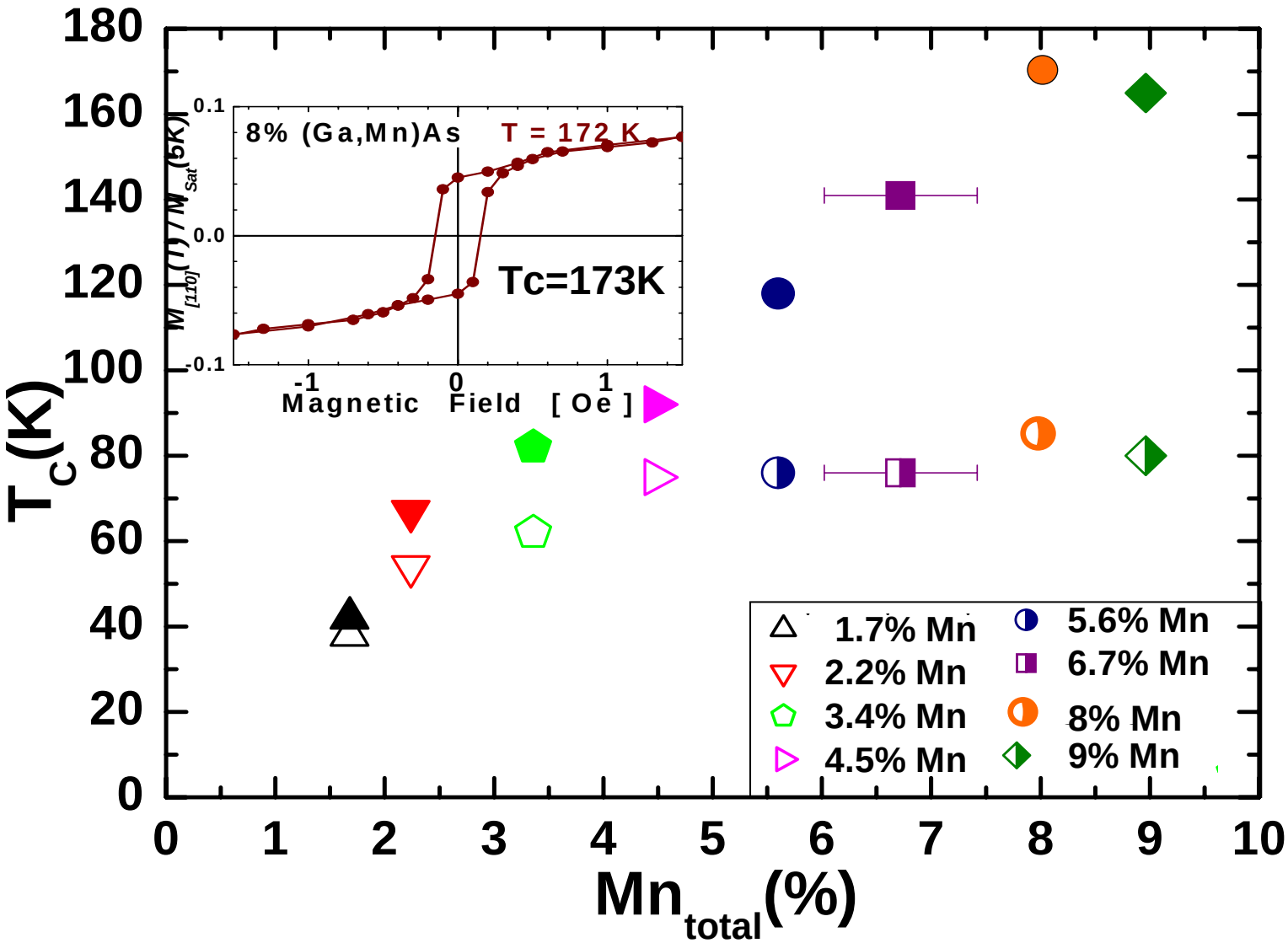
# Tc as grown and annealed samples

**Open symbols as grown. Closed symbols annealed**



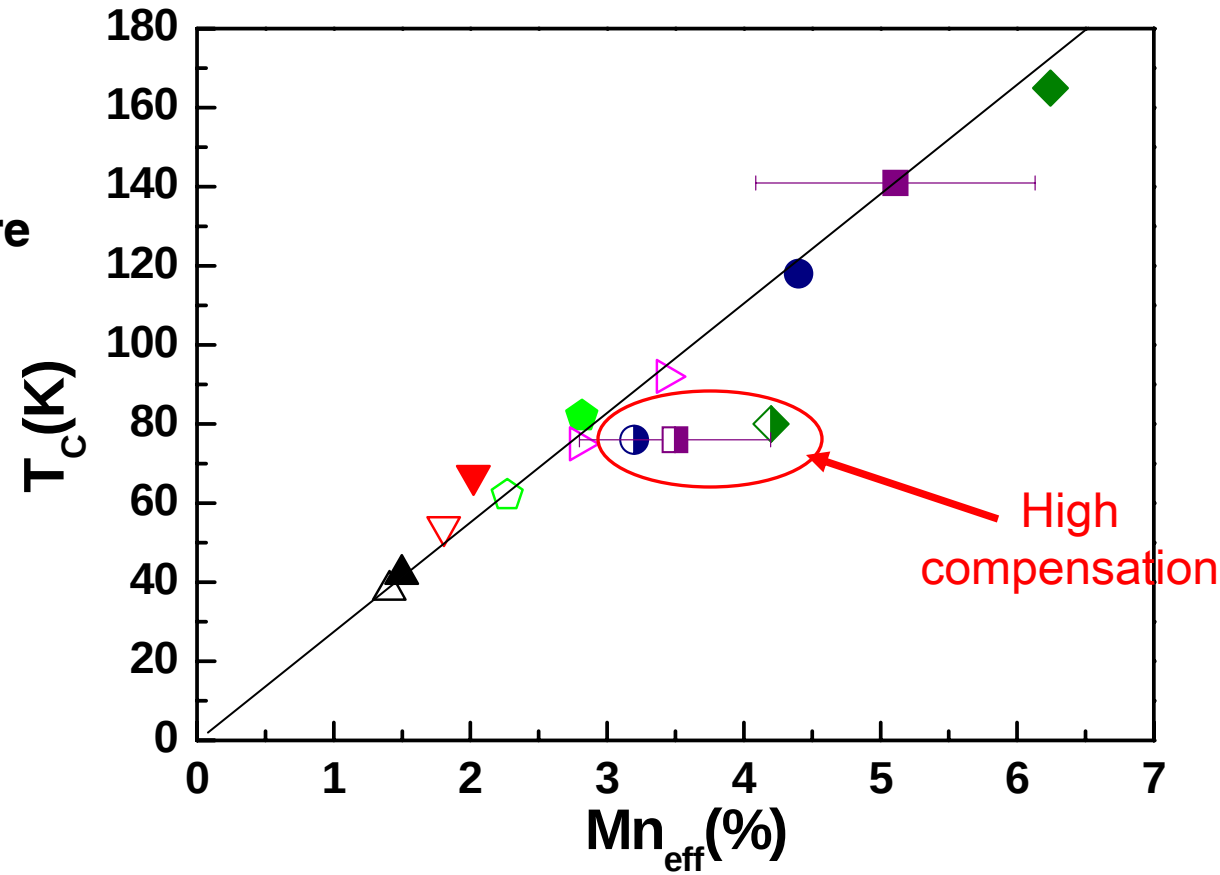
## Linear increase of Tc with effective Mn

**Effective Moment density, $Mn_{eff} = Mn_{sub}$-$Mn_{Int}$ due to AF $Mn_{sub}$-$Mn_{Int}$ pairs.**

**Tc increases with $Mn_{eff}$ when compensation is less than ~40%.**

**No saturation of Tc at high Mn concentrations**

**Closed symbols are annealed samples**



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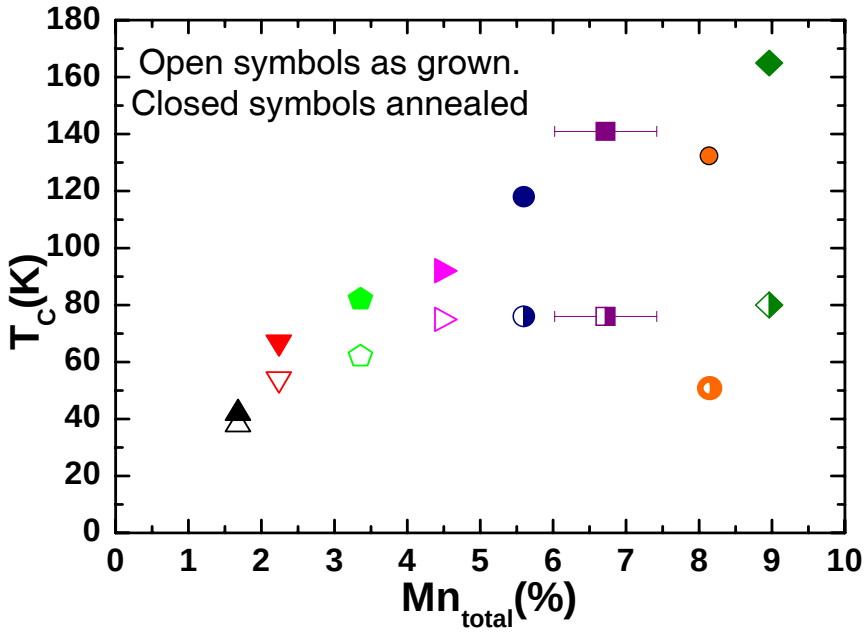
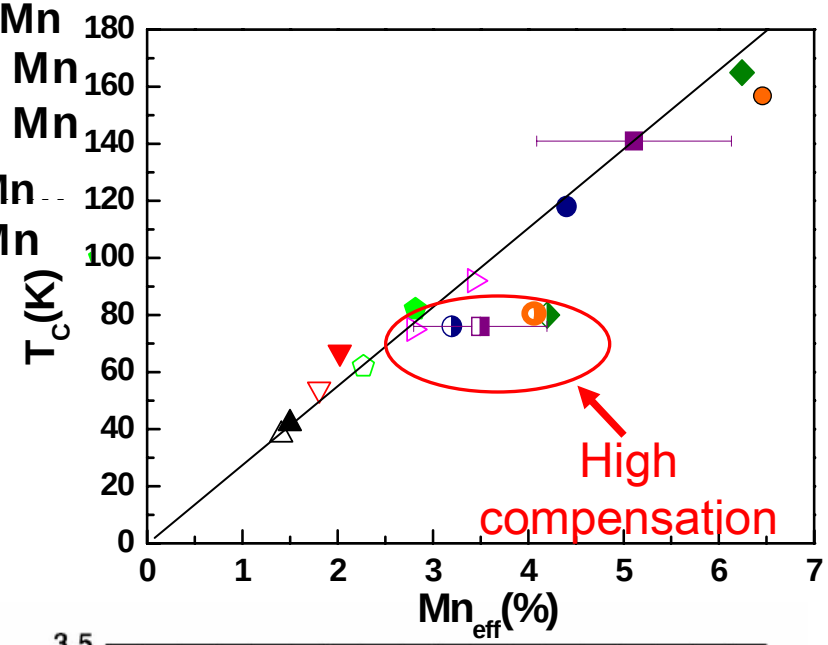
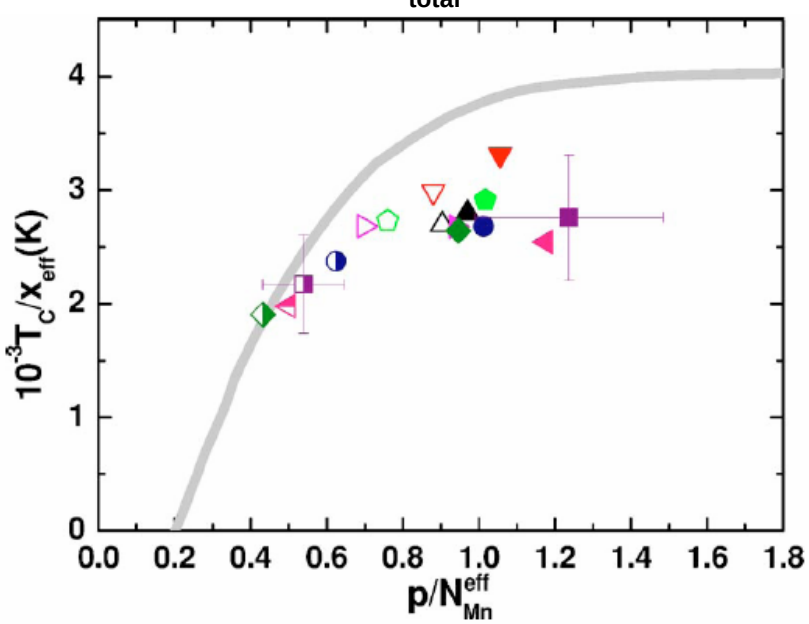
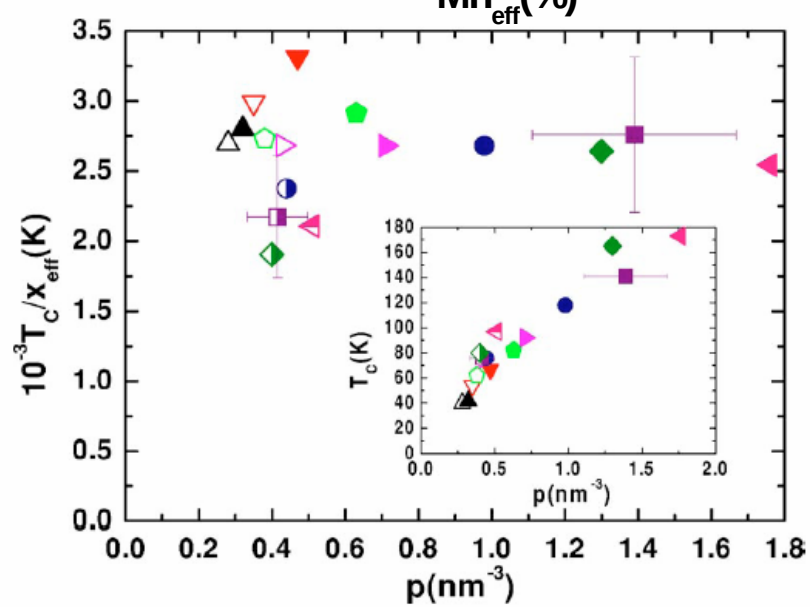



- 1.7% Mn
- 2.2% Mn
- 3.4% Mn
- 4.5% Mn
- 5.6% Mn
- 6.7% Mn
- 8% Mn
- 9% Mn

## Linear increase of Tc with $Mn_{eff} = Mn_{sub} - Mn_{Int}$

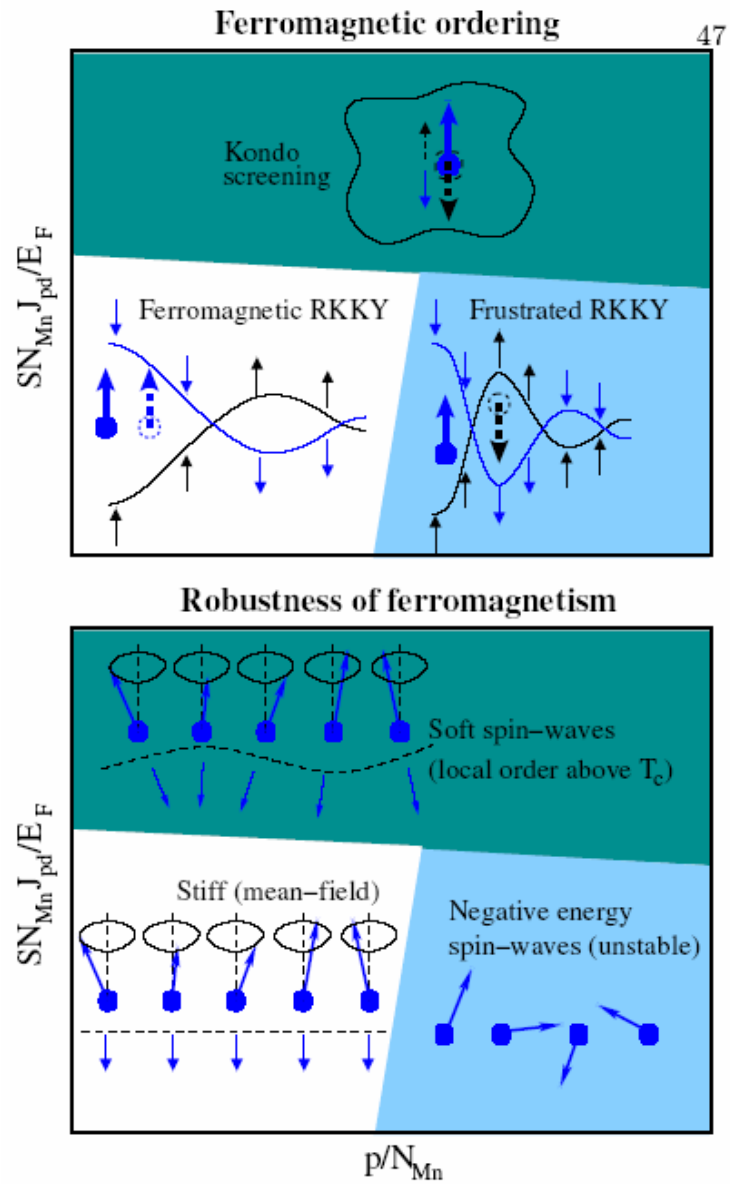


# Prospects of high Tc in DMSs

- Concentration of uncompensated $Mn_{Ga}$ moments has to reach ~10%. Only 6.2% in the current record Tc=173K sample
- Charge compensation not so important unless > 40%
- No indication from theory or experiment that the problem is other than technological - better control of growth-T, stoichiometry
- New growth or chemical composition strategies to incorporate more MnGa local moments or enhance p-d coupling
- Window in this difficult phase space is narrow and obtaining the optimal strength of the coupling and technical difficulties for GaMnAs may make it impossible to reach room Tc
- May want to look into materials close to this material but higher coupling strength to find the optimal system



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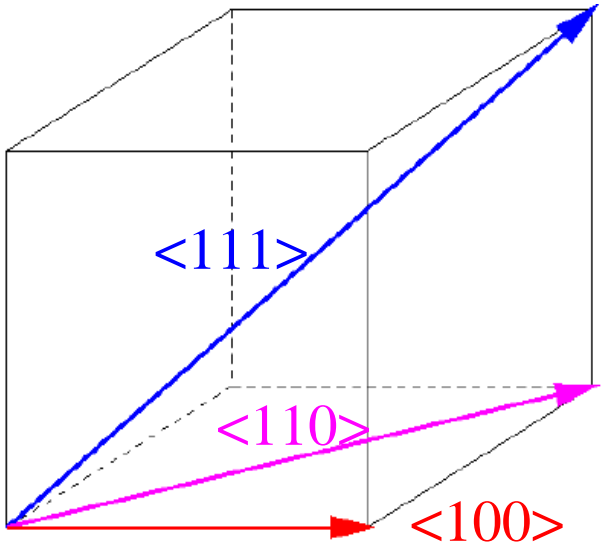
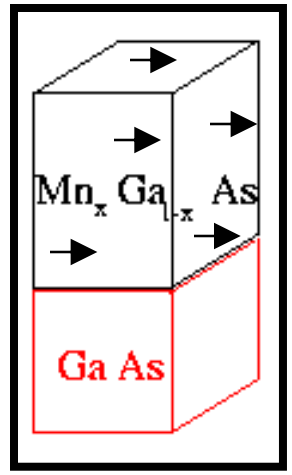
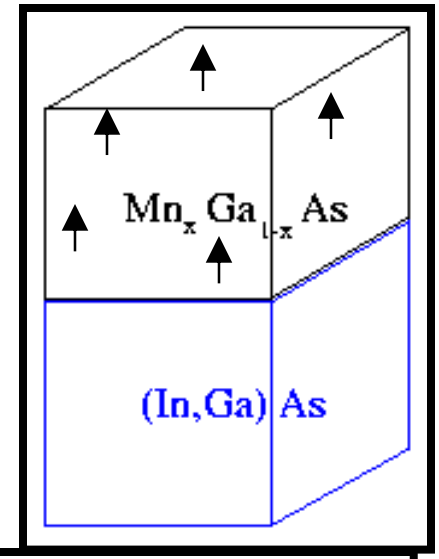
# MAGNETIC ANISOTROPY



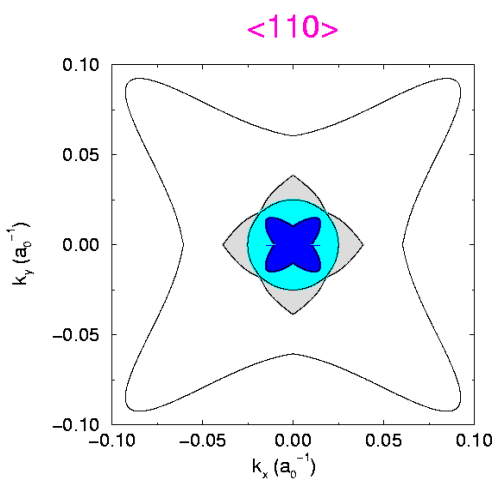

experiment:

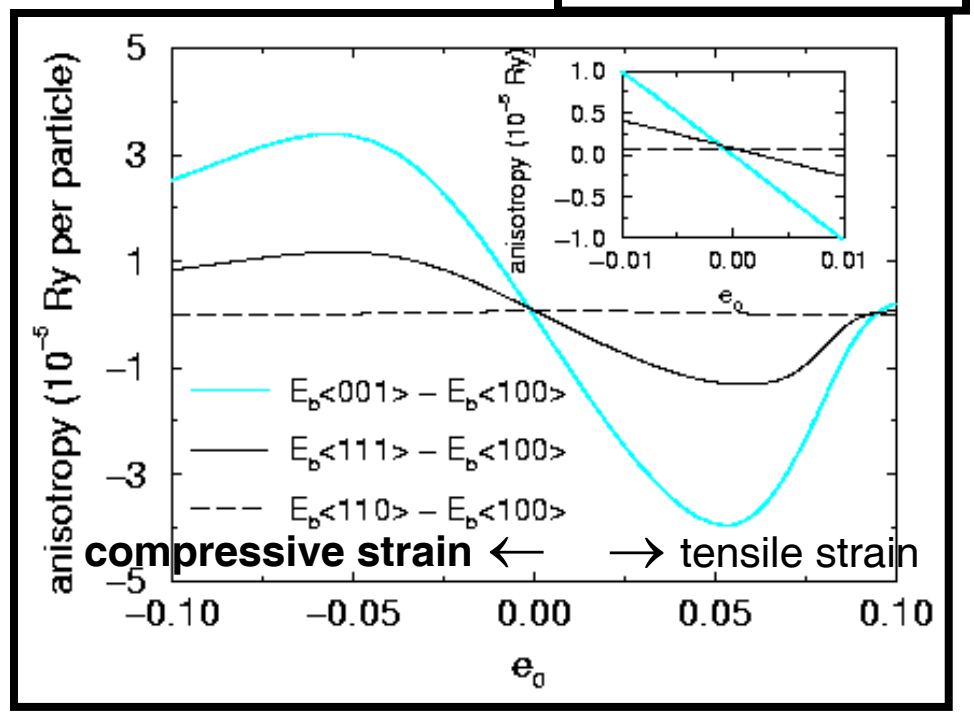


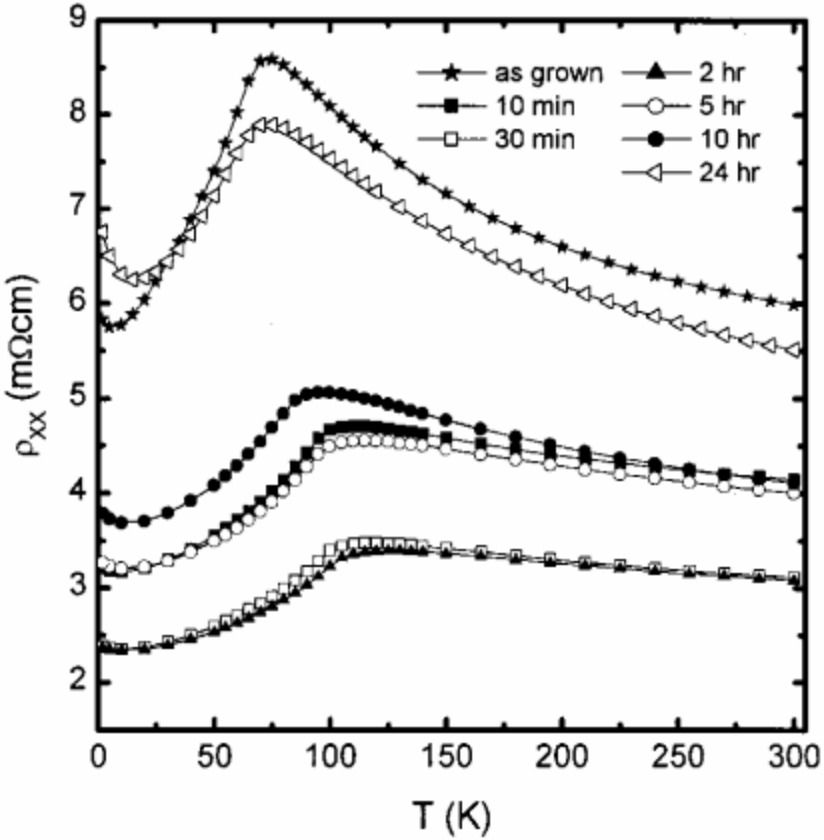

## Condensation energy depends on magnetization orientation

**M. Abolfath, T. Jungwirth, J. Brum, A.H. MacDonald, Phys. Rev. B 63, 035305 (2001)**

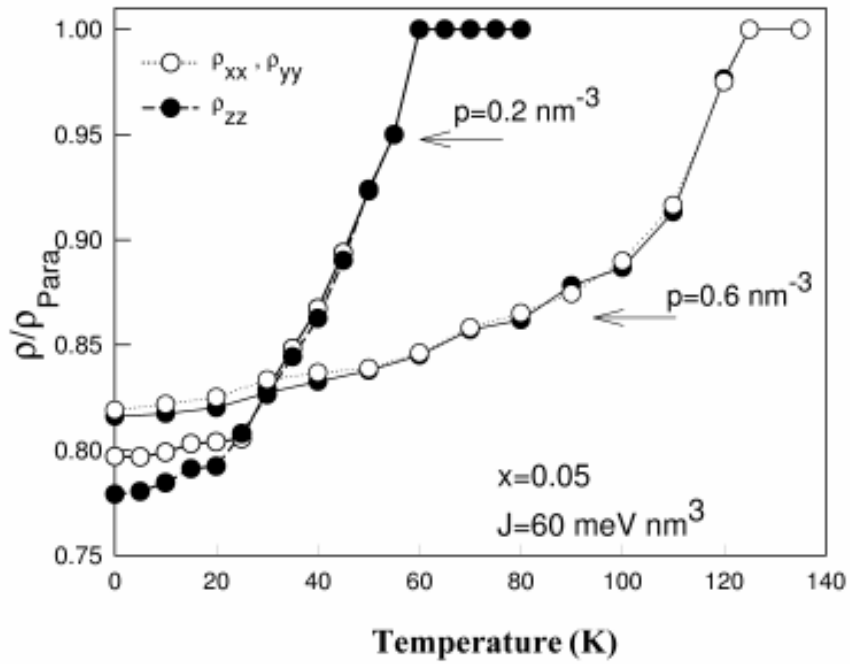


# Resistivity temperature dependence of metallic GaMnAs




Potashnik et al 2001

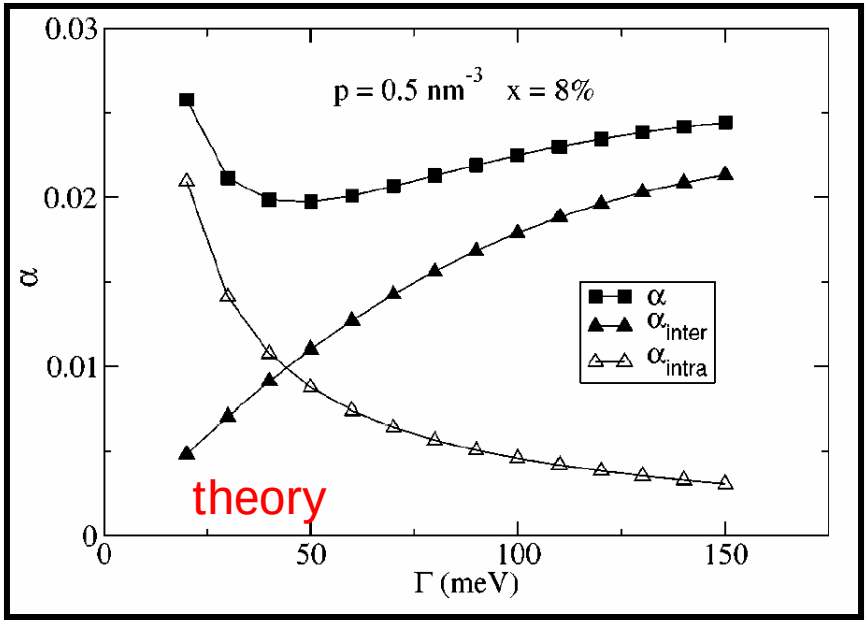


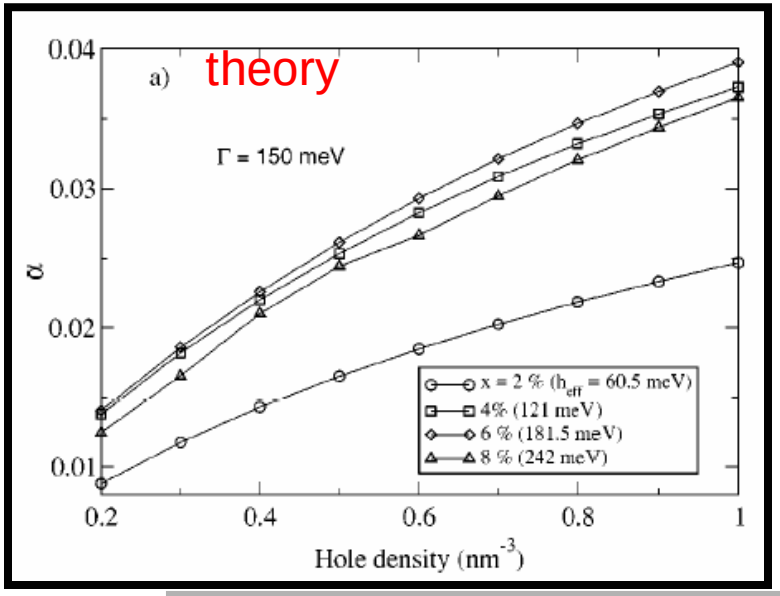

Lopez-Sanchez and Bery 2003
Hwang and Das Sarma 2005

# Ferromagnetic resonance: Gilbert damping

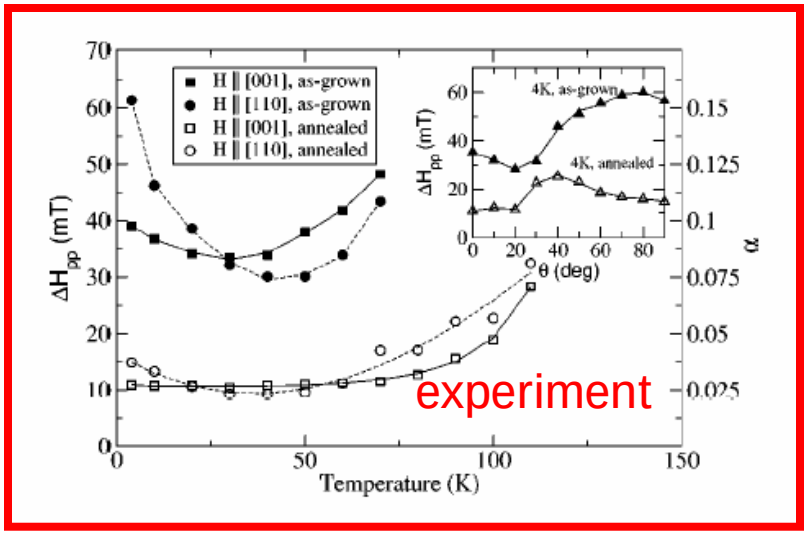
$$\alpha = \frac{J_{pd} h_{eff}}{4\hbar} \int \frac{d^3k}{(2\pi)^3} \sum_{a,b} |\langle \phi_a(\mathbf{k}) | s^+ | \phi_b(\mathbf{k}) \rangle|^2 \times A_{a,\mathbf{k}}(\epsilon_F) A_{b,\mathbf{k}}(\epsilon_F).$$

$$A_{a,\mathbf{k}}(\omega) = \Gamma / [(\epsilon - \epsilon_{a,\mathbf{k}})^2 + \Gamma^2/4].$$

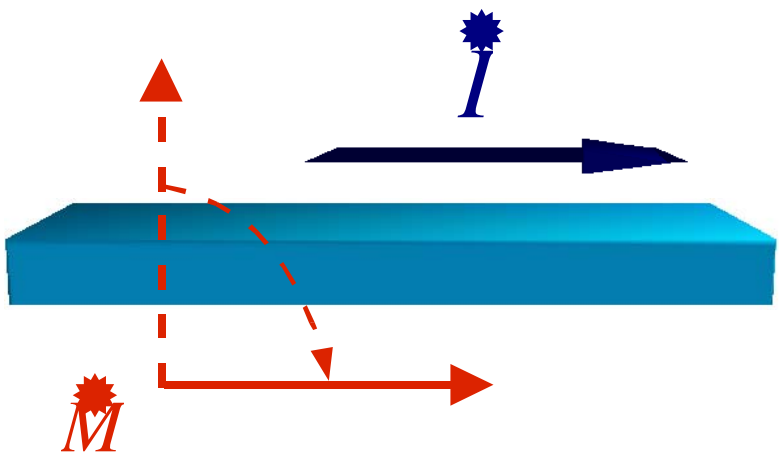


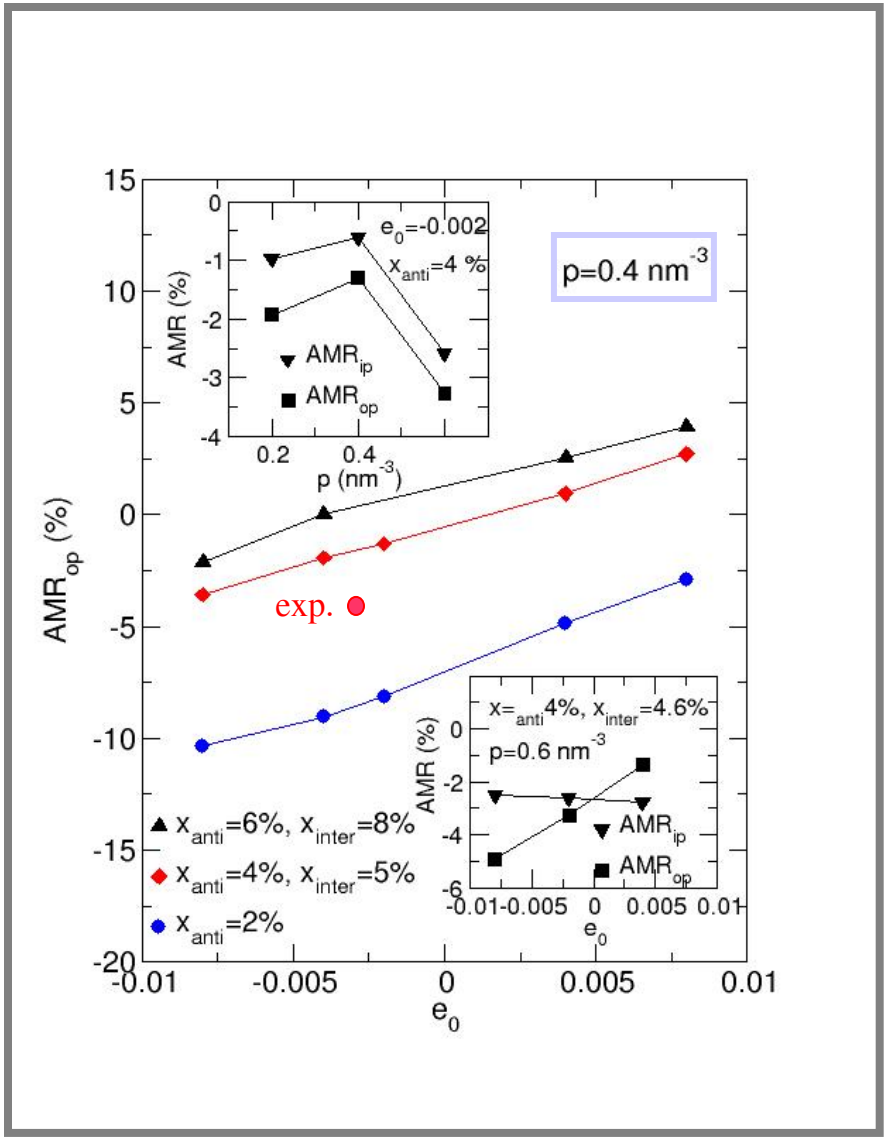




$$\Delta H_{pp}(\omega) = \Delta H_{pp}(0) + \frac{2}{\sqrt{3}} \frac{\omega}{g\mu_B} \alpha.$$



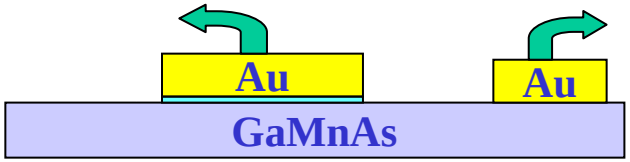
# Anisotropic Magnetoresistance



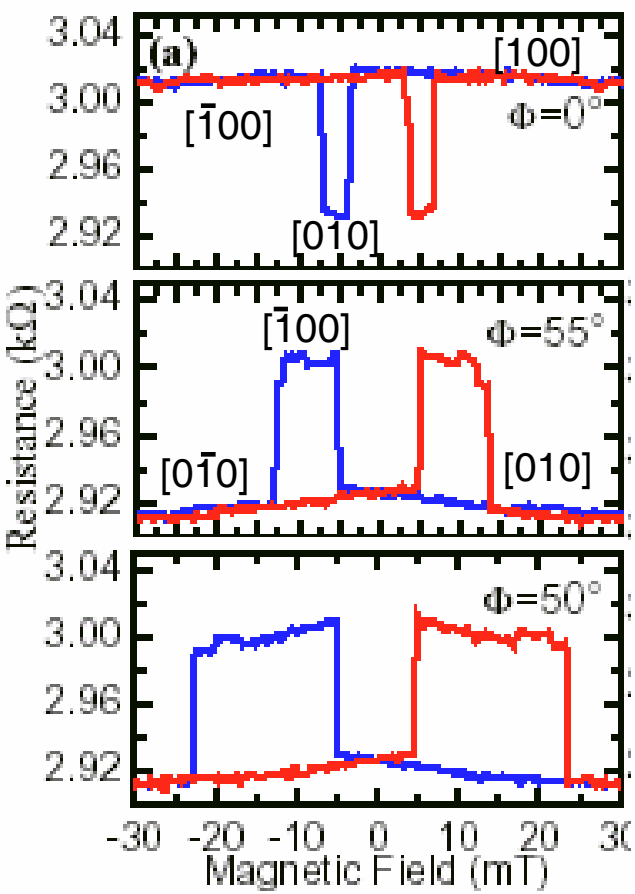



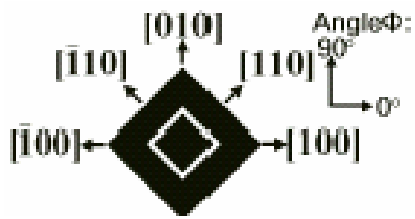

**T. Jungwirth, M. Abolfath, J. Sinova, J. Kucera, A.H. MacDonald, Appl. Phys. Lett. 2002**

# Tunneling anisotropic magnetoresistance (TAMR)



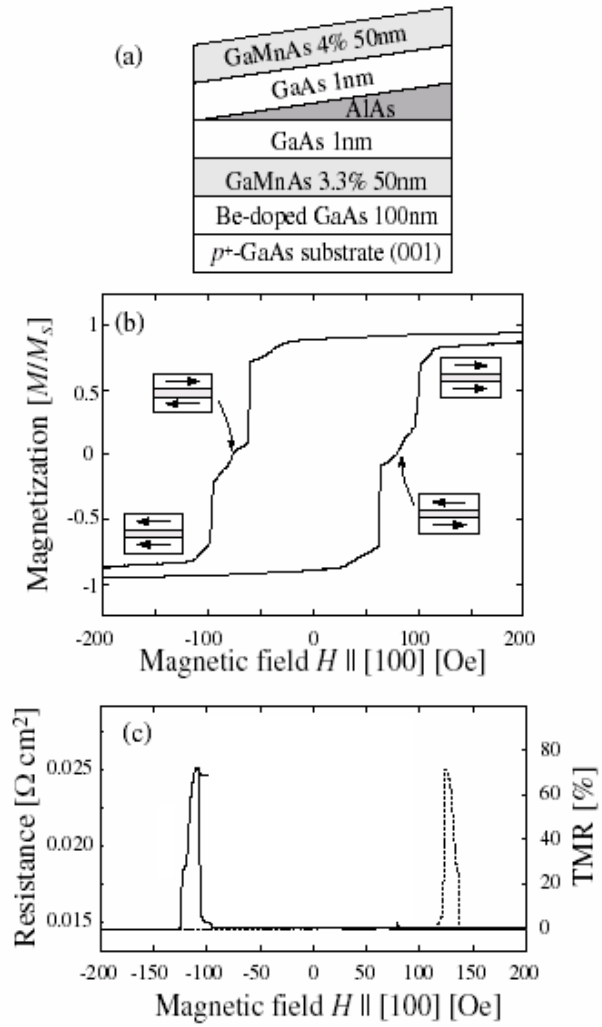

*Gould, Ruster, Jungwirth, et al., PRL '04*



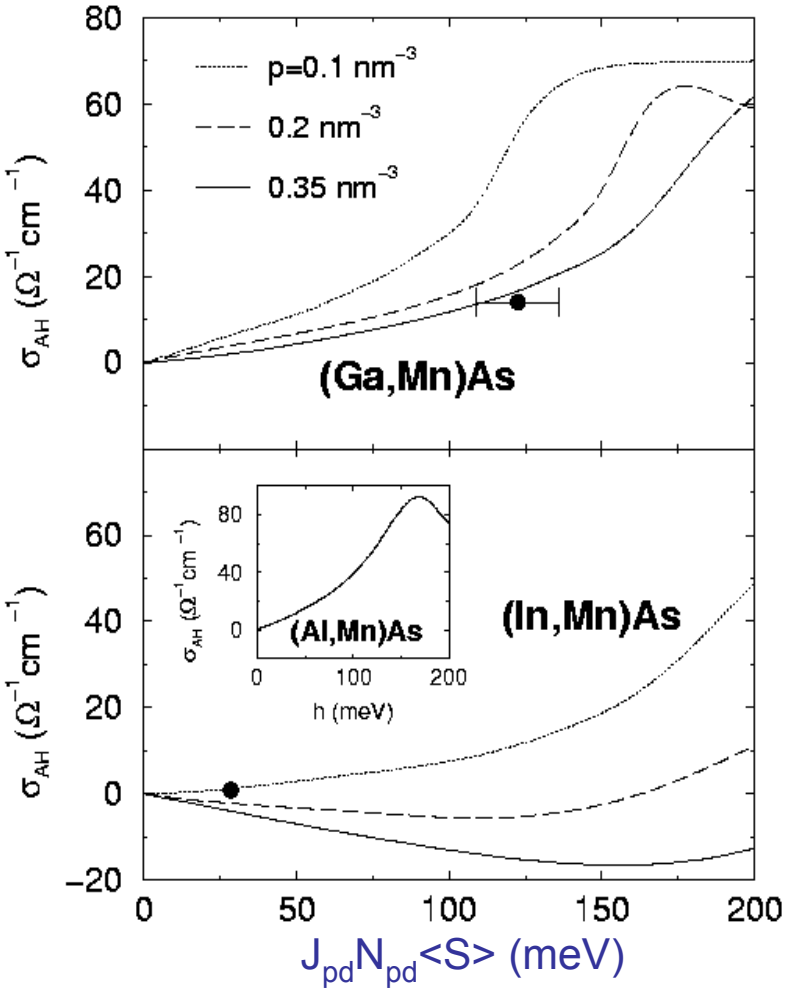

**Bistable memory device with a single magnetic layer only**

## Giant magneto-resistance



# ANOMALOUS HALL EFFECT

AHE without disorder




**T. Jungwirth, Q. Niu, A.H. MacDonald, Phys. Rev. Lett. 88, 207208 (2002)**

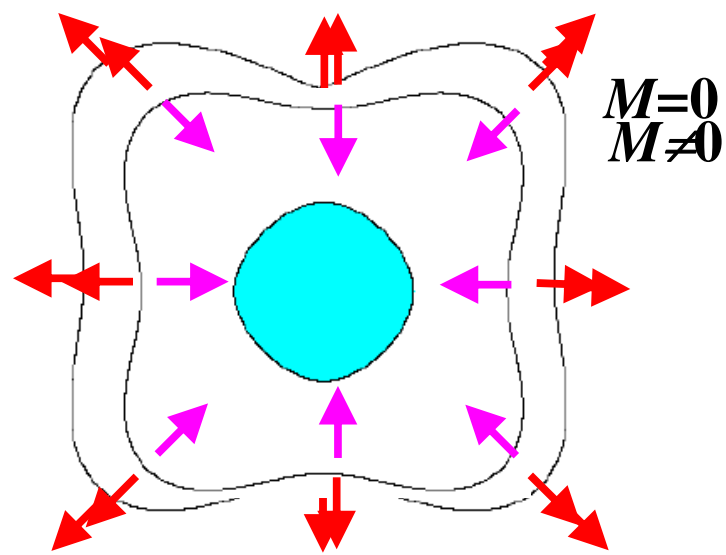
**anomalous velocity:**

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

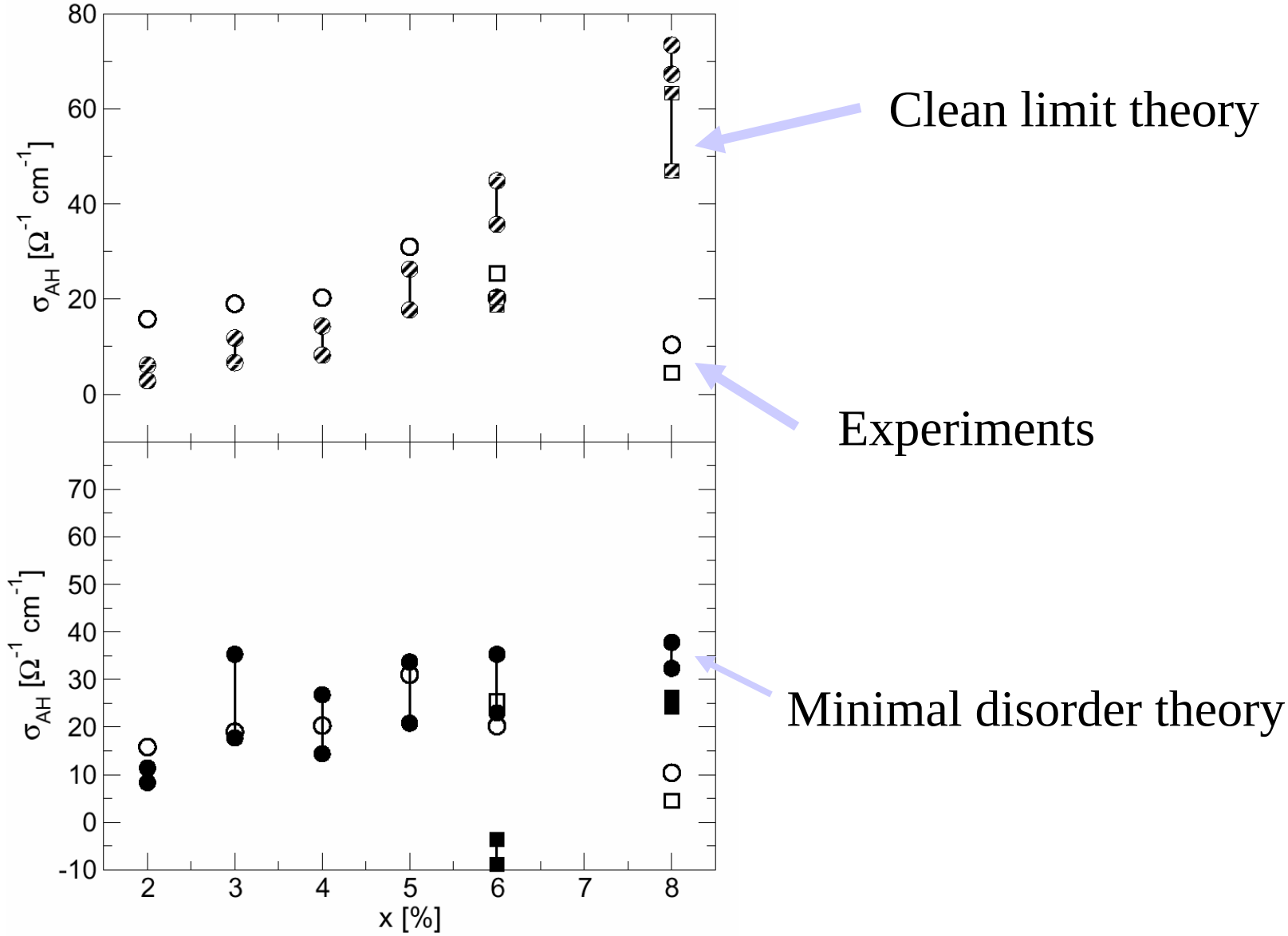
**Berry curvature:**

$$\Omega_z = 2\mathrm{Im}[\langle \frac{\partial u}{\partial k_y} | \frac{\partial u}{\partial k_x} \rangle].$$

$$\sigma_{AH} = -\frac{e^2}{\hbar} \sum_n \int \frac{d\vec{k}}{(2\pi)^3} f_{n,\vec{k}} \Omega_z(n, \vec{k}) \ ,$$



# ANOMALOUS HALL EFFECT IN GaMnAs



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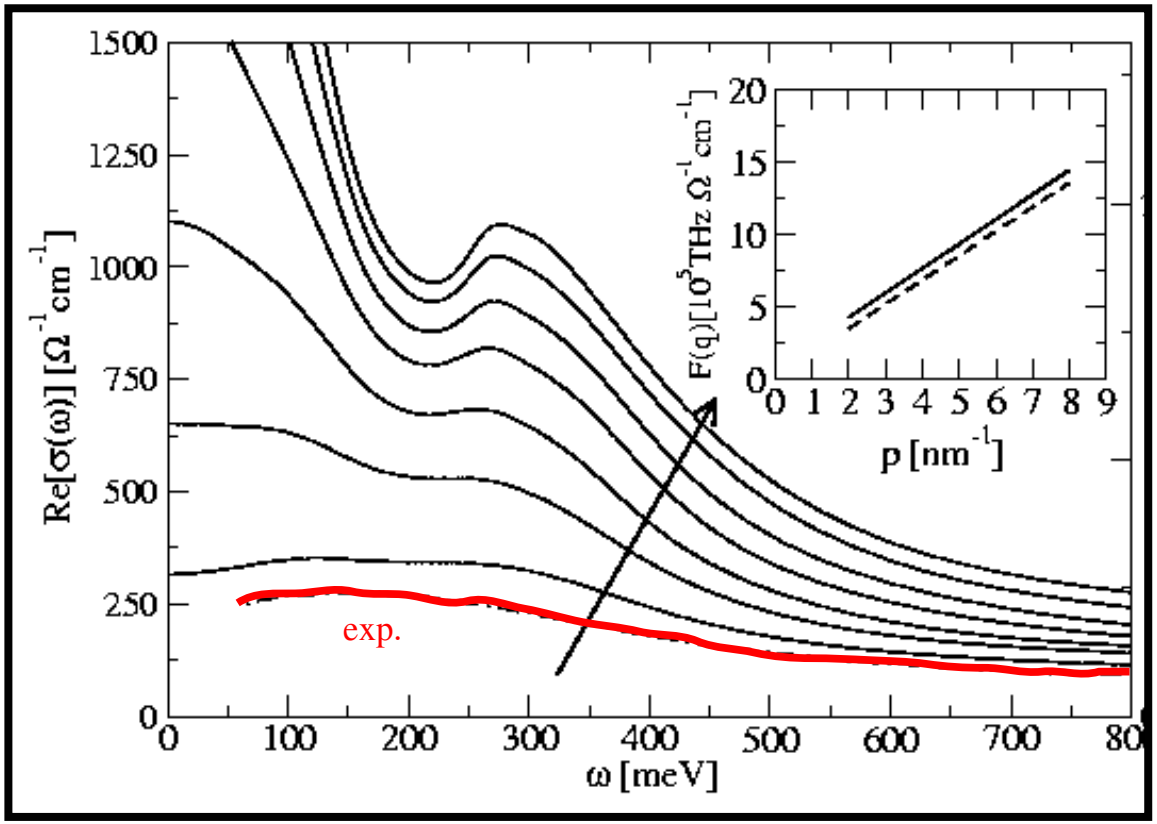
# The valence band picture of IR absorption

$$F = \int d\omega \mathrm{Re}[\sigma(\omega)] = \pi e^2 p / 2m_{opt}$$

hole density: $p$=0.2, 0.3, ....., 0.8 nm$^{-3}$

x=5%




$m_{opt}$ independent of (within 10%):

- density
- disorder
- magnetic state

***GaAs***

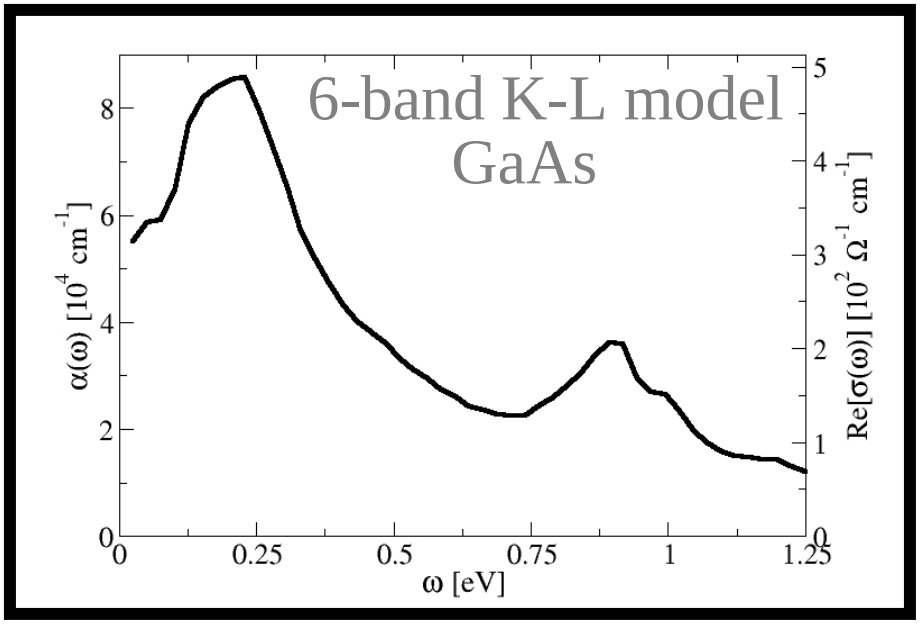
$$m_{op} \approx 0.24\, m_e$$

**J. Sinova, et al. Phys. Rev. B 66, 041202 (2002).**

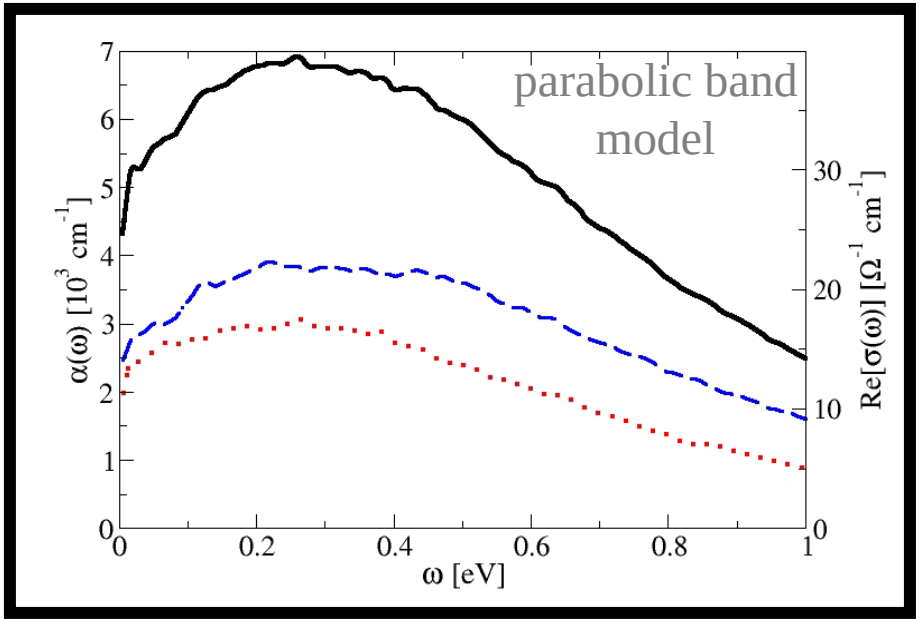
**Exps: Singley et al Phys. Rev. Lett. 89, 097203 (2002)**
**Hirakawa, et al Phys. Rev. B 65, 193312 (2002)**

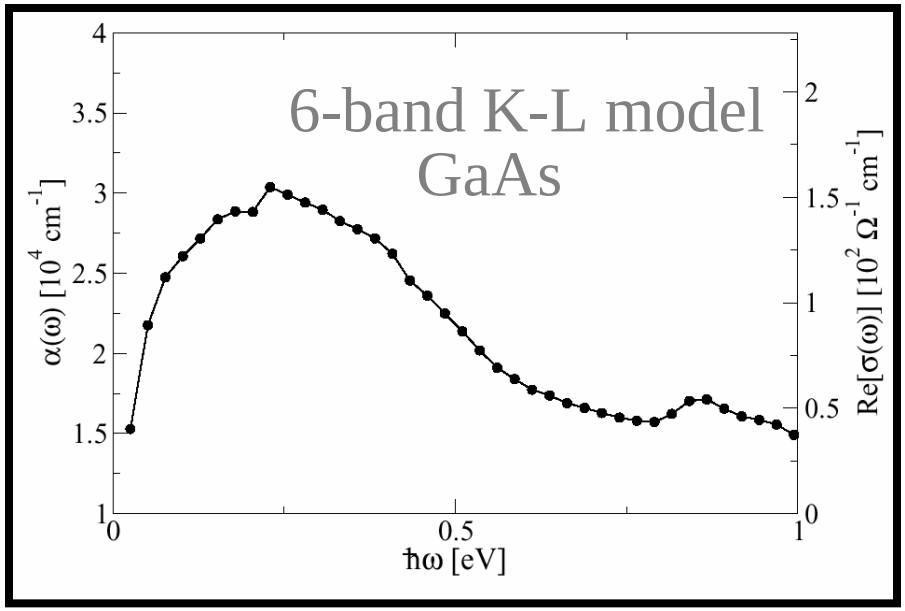
**infrared absorption →accurate density measurement**

# FINITE SIZE EXACT DIAGONALIZATION STUDIES




p=0.33 nm$^{-3}$, x=4.5%,
compensation from anti-sites




p=0.2 nm$^{-3}$, x=4.0%,
compensation from anti-sites

f-sum rule accurate within 10 %

**S.-R. E. Yang, J. Sinova, T. Jungwirth, Y.P. Shim, and A.H. MacDonald, PRB 67, 045205 (03)**

## Possible issues regarding IR absorption

- Energy dependence of Jpd
- Localization effects
- Contributions due to impurity states: Flatte's approach of starting from isolated impurities
- Systematic p and $x_{eff}$ study (need more than 2 $m_{eff}$ data points)

# Keeping Score

The effective Hamiltonian (MF) and weak scattering theory (no free parameters) describe (III,Mn)V shallow acceptor metallic DMSs very well in the regime that is valid:

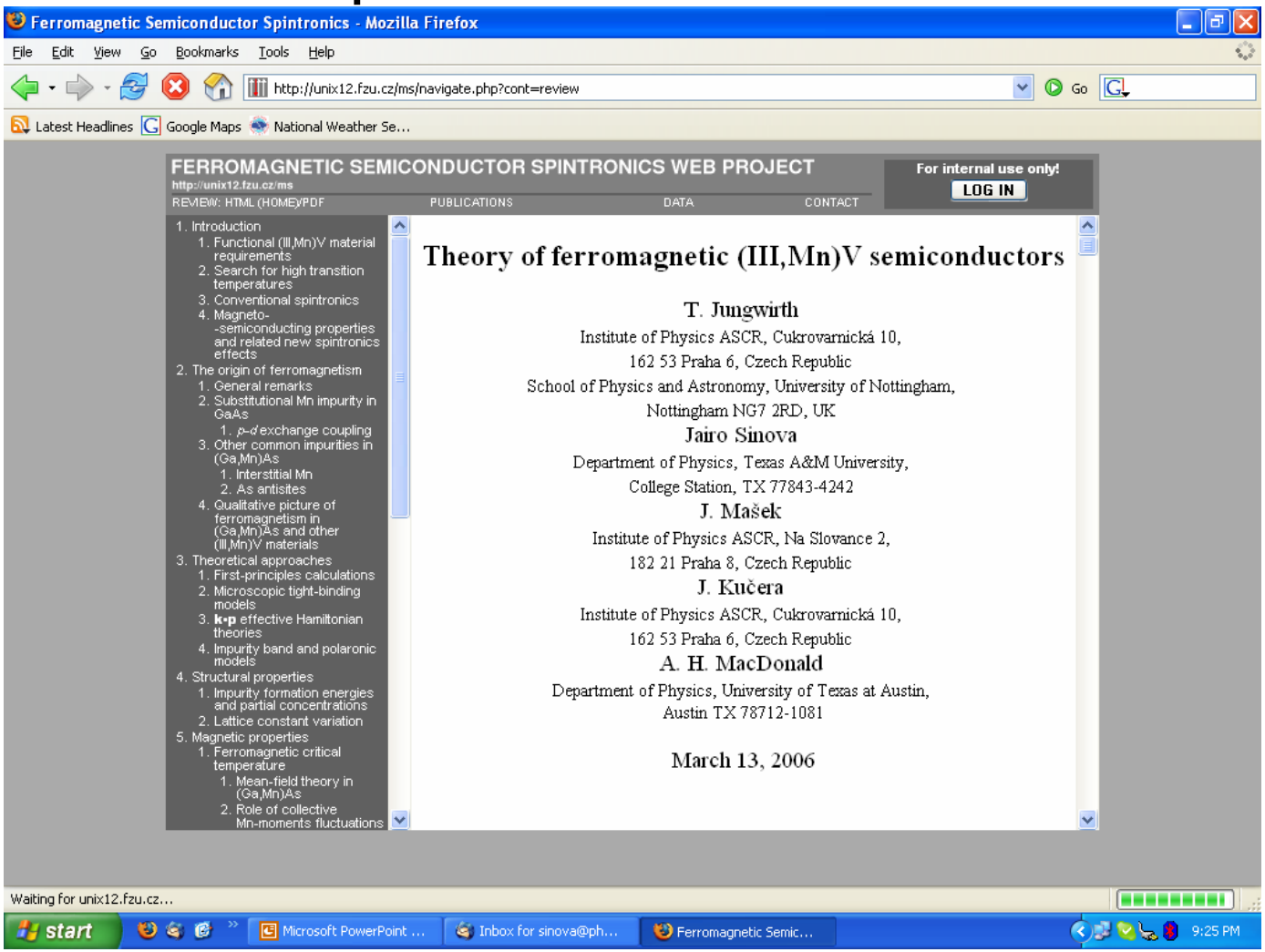
- Ferromagnetic transition temperatures √
- Magneto-crystalline anisotropy and coercively √
- Domain structure √
- Anisotropic magneto-resistance √
- Anomalous Hall effect √
- MO in the visible range √
- Non-Drude peak in longitudinal ac-conductivity √
- Ferromagnetic resonance √
- Domain wall resistance √
- TAMR √

**BUT it is only a peace of the theoretical mosaic with many remaining challenges!!**

**TB+CPA and LDA+U/SIC-LSDA calculations describe well chemical trends, impurity formation energies, lattice constant variations upon doping**

***Theory of ferromagnetic (III,Mn)V semiconductors***,
Jungwirth, Sinova, Masek, Kucera, and MacDonald,
**to appear in Rev. of Mod. Phys., in cond-mat/0603380**

# http://unix12.fzu.cz/ms



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# Publications:

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**Public. type**

- Review
- Other

**Experiment/Theory**

- Experiment

**Theory**

- First-principles calculations
- Microscopic tight-binding approximation
- **k·p** effective Hamiltonian theories
- Impurity band and polaronic models

**Material**

**Mn-doped III-V's**

| | |
|---|---|
| AlAs | AlSb |
| AlP | AlN |
| GaAs | GaSb |
| GaP | GaN |
| InAs | InSb |
| InP | InN |

**Related magnetic materials**

- Other transition metal-doped III-V's
- II-VI diluted magnetic semiconductors
- Group IV diluted magnetic

**Topic**

**Origins of ferromagnetism**

- Role of substitutional Mn
- Other impurities
- Pictures of ferromagnetism

**Bulk properties**

Structural properties

- Formation and binding energies
- Lattice constant

Done

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