

# Magnetyzm w półprzewodnikach

**Tomasz DIETL**

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Laboratory for Cryogenic and Spintronic Research  
Institute of Theoretical Physics, Warsaw University*

**współpracownicy:**

*M. Sawicki et al. – Warszawa*

*J. Cibert et al. – Grenoble*

*H. Ohno et al. – Sendai*

*S. Kuroda et al. – Tsukuba*

*A. Bonanni et al. – Linz*

*B. Gallagher et al. – Nottingham*

*L. Molenkamp et al. – Wuerzburg*

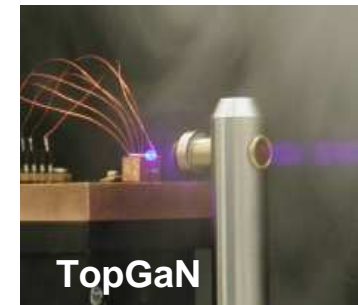
**finansowanie:** *ERATO – JST; NANOSPIN -- EC project;*

*SPINTRA – ESF; Humboldt Foundation, AdG ERC „FunDMS”*

*artykuły przeglądowe, patrz arXiv; : J. Phys. C'07; JAP'08, JPSJ'08*

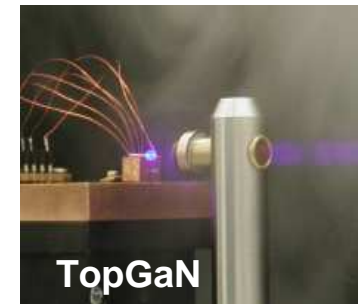
# Spintronics -- materials aspect

*Why to do not combine complementary resources of ferromagnets and semiconductors?*



# Spintronics -- materials aspect

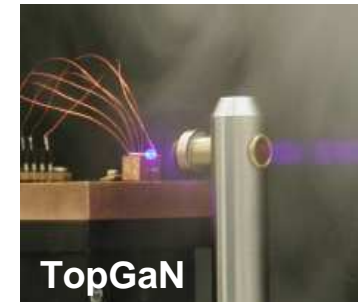
*Why to do not combine complementary resources of ferromagnets and semiconductors?*



- **hybrid ferromagnetic-metal/semiconductor structures**
  - **spin injection** *A. Hanbicki et al. (NRL) APL'03, Nature Phys. '07*
  - **optical isolators** *H. Shimizu et al. (Tokyo) JJAP'04*
  - **Stern-Gerlach apparatus** *J. Wróbel et al. (Warsaw) PRL'04*
  - .....

# Spintronics -- materials aspect

*Why to do not combine complementary resources of ferromagnets and semiconductors?*



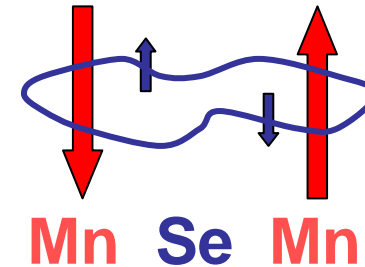
- **ferromagnetic semiconductors – multifunctional materials**

# Making *semiconductors* ferromagnetic

- ***Antiferromagnetic* superexchange dominates**  
in magnetic insulators and semiconductors

→ **no spontaneous magnetization**

NiO, MnSe, EuTe, ...

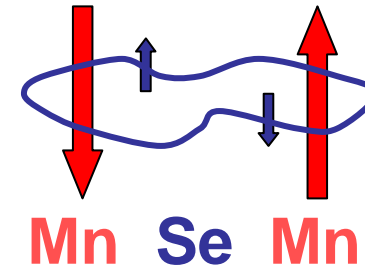


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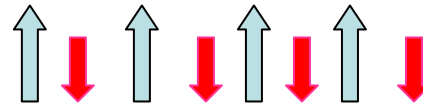
NiO, MnSe, EuTe, ...



- **Exceptions**

-- **ferrimagnets (two ions or two spin states co-exist)**

NiO(Fe<sub>2</sub>O<sub>3</sub>), Mn<sub>4</sub>N, ...

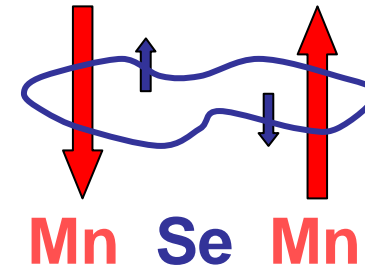


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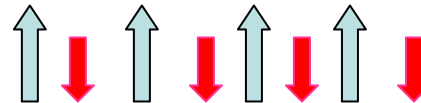
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- **Exceptions**

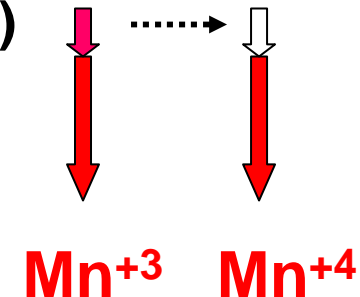
- **ferrimagnets (two ions or two spin states co-exist)**

NiO(Fe<sub>2</sub>O<sub>3</sub>), Mn<sub>4</sub>N, ...



- **double exchange (two charge states co-exist)**

LaMnO<sub>3</sub> → La<sub>1-x</sub>Sr<sub>x</sub>MnO<sub>3</sub> (holes in d band)

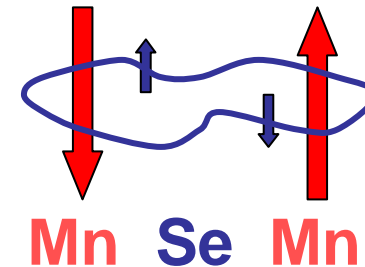


# Search for ferromagnetic semiconductors

- **Antiferromagnetic superexchange dominates**  
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→ **no spontaneous magnetization**

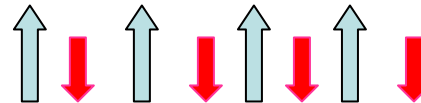
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- **Exceptions**

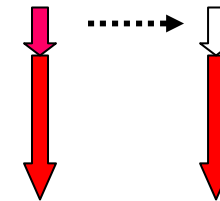
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**Mn<sup>+3</sup> Mn<sup>+4</sup>**

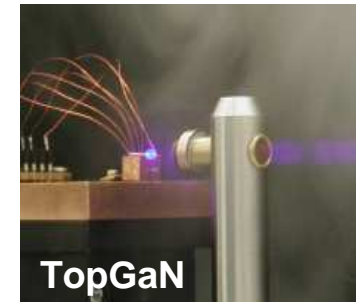
- **ferromagnetic superexchange dominates**

EuO, ZnCr<sub>2</sub>Se<sub>4</sub>, ...  $T_C \approx 100$  K *IBM, MIT, Tohoku, ... '60-'70*



# Spintronics -- materials aspect

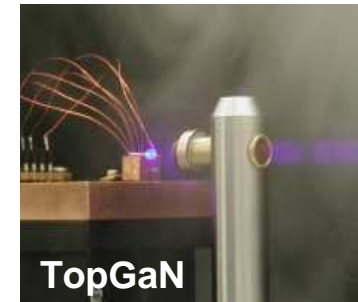
*Why to do not combine complementary resources of ferromagnets and semiconductors?*



- **ferromagnetic semiconductors – multifunctional materials**
  - **making good semiconductors of magnetic oxides**  
*cf. J. Fontcuberta, R. Gross, N. Keller, ....*

# Spintronics -- materials aspect

*Why to do not combine complementary resources of ferromagnets and semiconductors?*

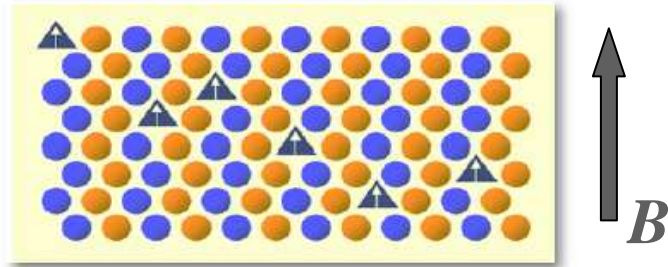


- **ferromagnetic semiconductors – multifunctional materials**
  - **making good semiconductors of magnetic oxides**
  - **making good semiconductors magnetic**

*R.R. Gałazka et al. (Warsaw)'77- ; H. Ohno et al. (IBM, Tohoku) '89 – see, Spin Physics, Nature Milestones (2008)*

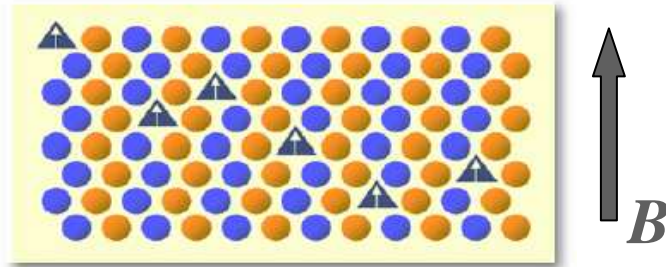
# Making DMS ferromagnetic

- Intrinsic DMS – random *antiferromagnets*

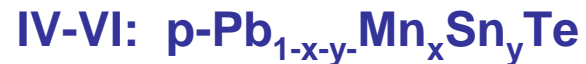


# Making DMS ferromagnetic

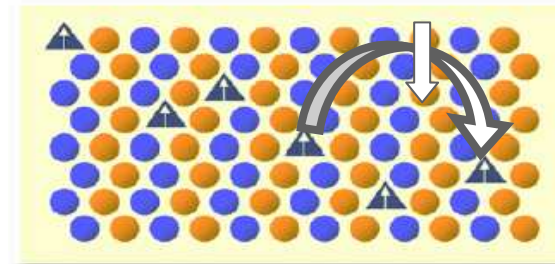
- Intrinsic DMS – random *antiferromagnets*



- p<sup>+</sup>-type DMS – Zener/RKKY *ferromagnets*



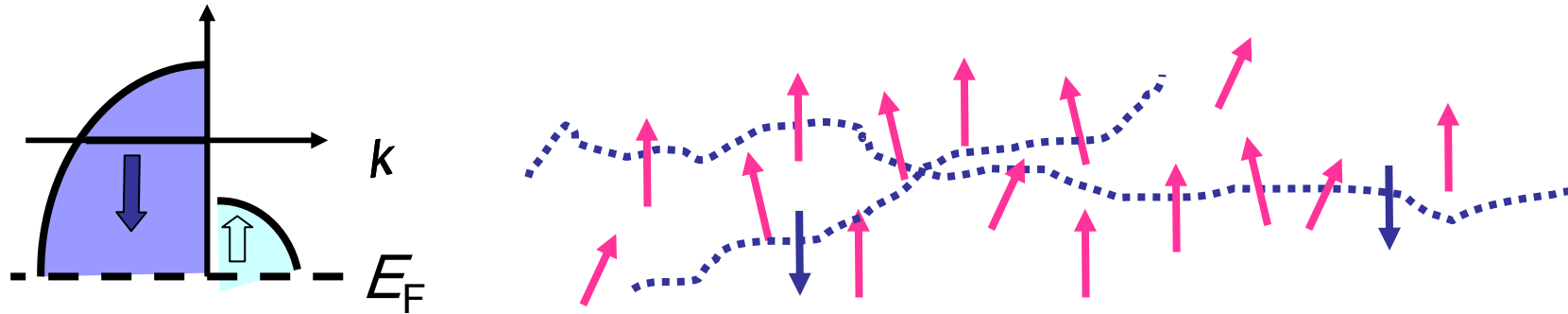
*Story et al. (Warsaw, MIT) PRL '86*



$T_C \approx 100 \text{ K for } x = 0.05$



# p-d Zener/RKKY model of hole-controlled ferromagnetism in DMS

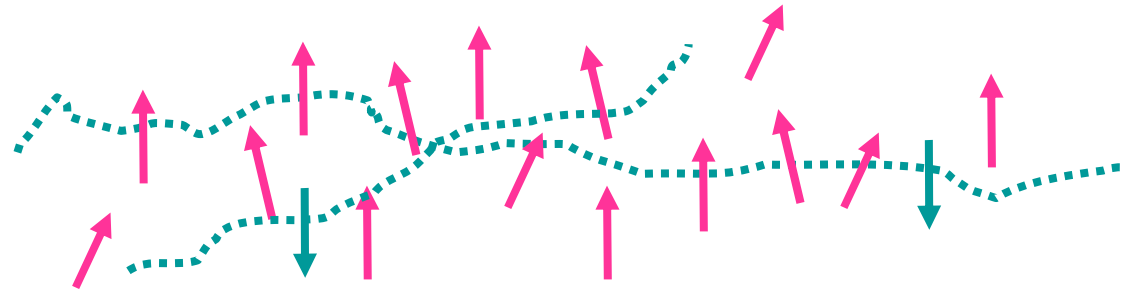
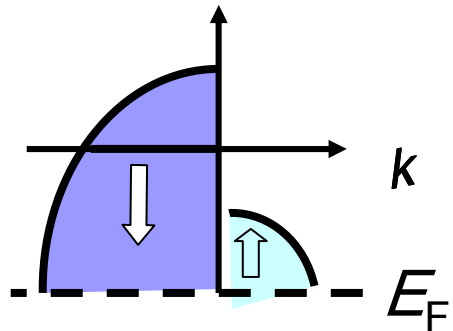


*Driving force:*

lowering of the hole energy due to redistribution between hole spin subbands split by p-d exchange interaction

*T.D. , Y. Merle d'Aubigné PRB'97-  
T. D, H. Ohno – Science'00 -  
Jungwirth et al. (Austin/Prague) '99-*

# p-d Zener/RKKY model of hole-controlled ferromagnetism in DMS

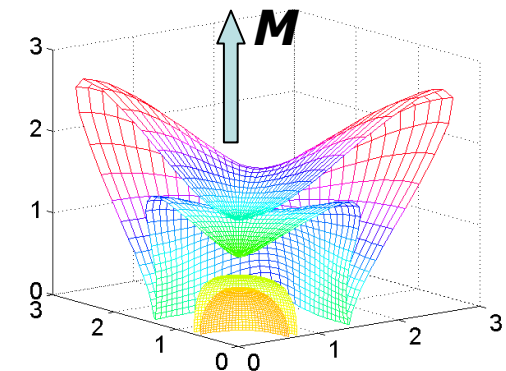


*Driving force:*

lowering of the hole energy due to redistribution between hole spin subbands split by *p-d* exchange interaction,  $\Delta \sim \beta M$

*Essential ingredient:*

Complexity of the valence band structure has to be taken into account



No adjustable parameters

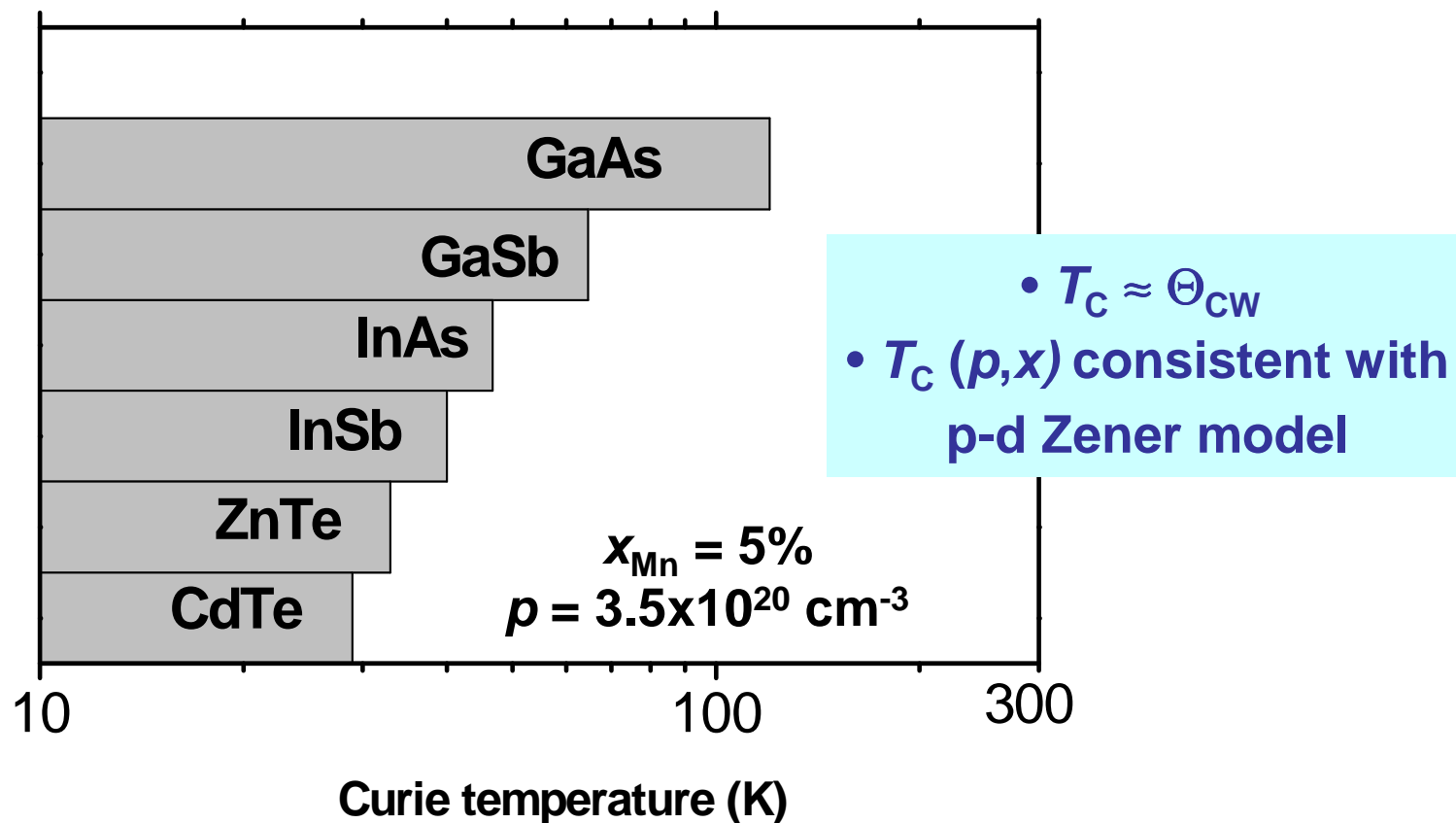
$$T_C \sim x\beta^2\rho^{(s)}_{\text{DOS}}$$

T.D. , Y. Merle d'Aubigne PRB'97-

T. D, H. Ohno – Science'00 -

T. Jungwirth et al. (Austin/Prague) '99-

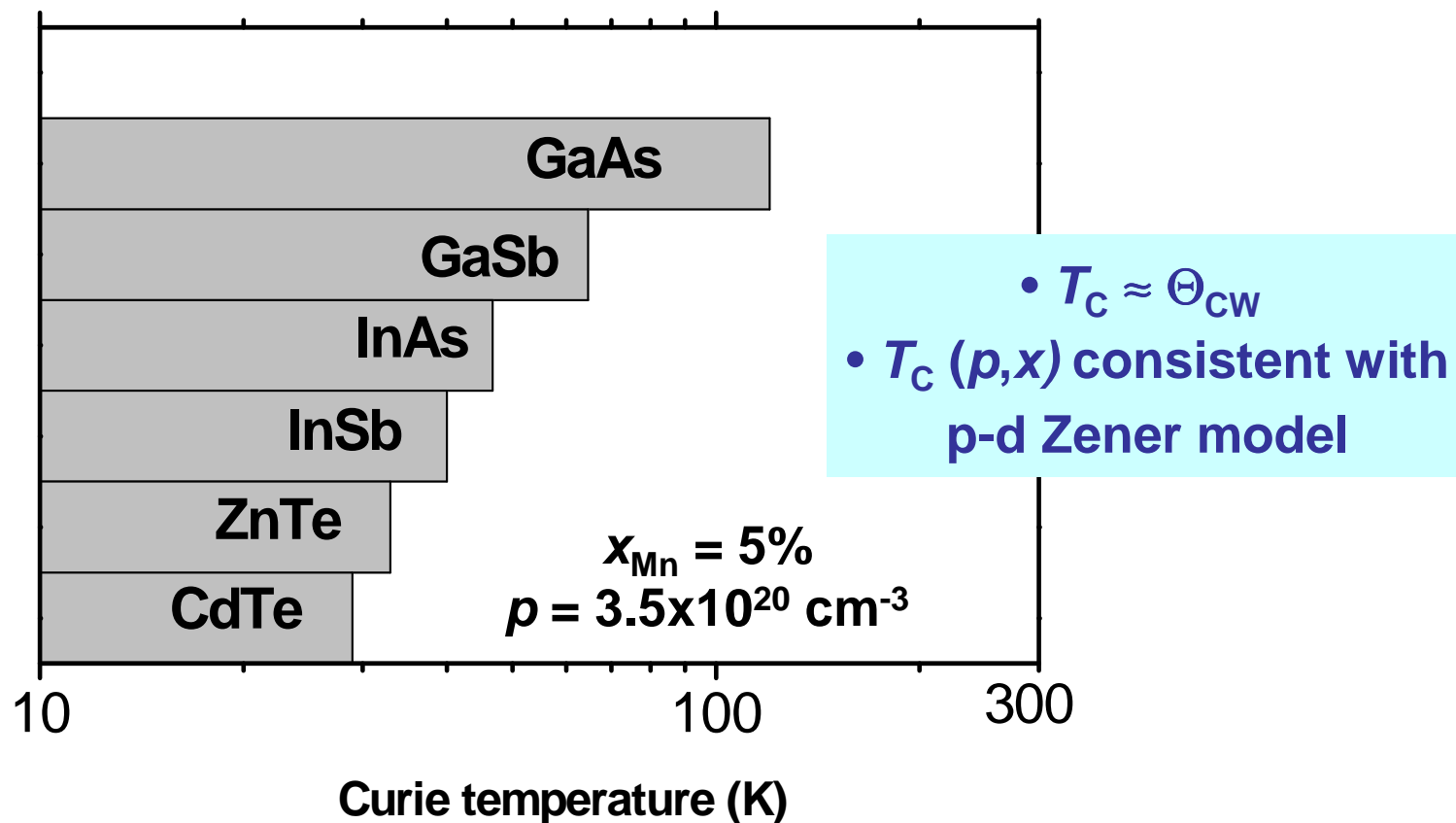
# Mn-based p-type DMS with valence-band hole mediated ferromagnetism



Expl.:

*Tohoku, Tokyo, Grenoble, Wuerzburg, PSU, Notre Dame, Lund, UCSB, Nottingham, Prague...*

# Mn-based p-type DMS with valence-band hole mediated ferromagnetism



Expl.:

*Tohoku, Tokyo, Grenoble, Wuerzburg, PSU, Notre Dame, Lund, UCSB, Nottingham, Prague...*

- not double exchange
- not impurity band models

*see, T. Jungwirth, ... T.D., PRB'07*



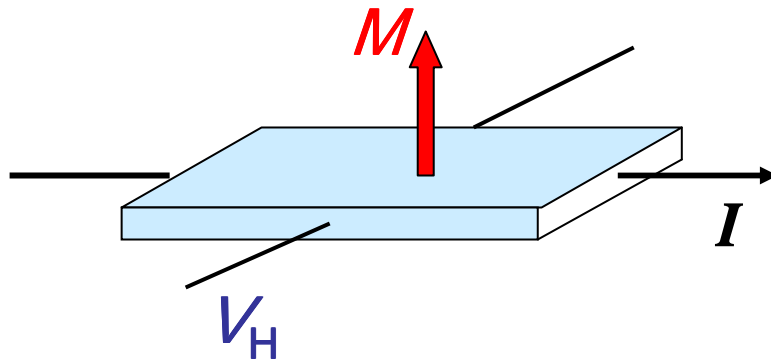
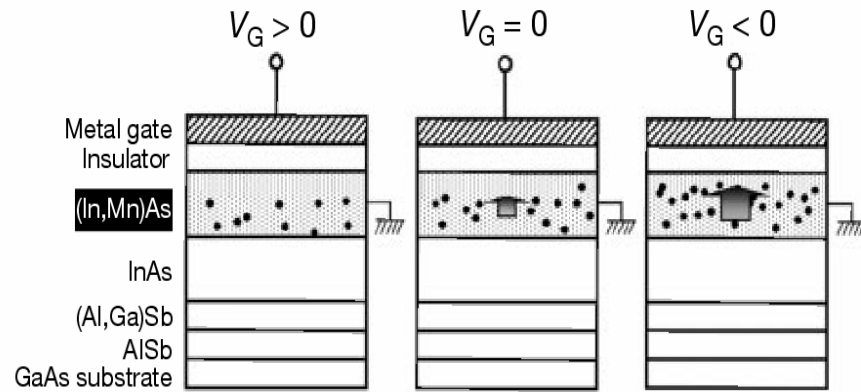
# Spintronic functionalities of ferro DMS

- **spin injection** (*Tohoku/St. Barbara Nature'99, JJAP'01, PRB'02  
APL'03,'06; IMEC/Warsaw APL'04, PRB'05*)
- **GMR, TMR, TAMR** (*Tohoku Physica'00,'04; Thales PRL'03,  
Wuerzburg PRL'04,'05, Nottingham PRL'05*)
- **RTD** (*Tohoku APL'98, Tokyo APL'06; Thales PRL'07*)
- **''''''''**

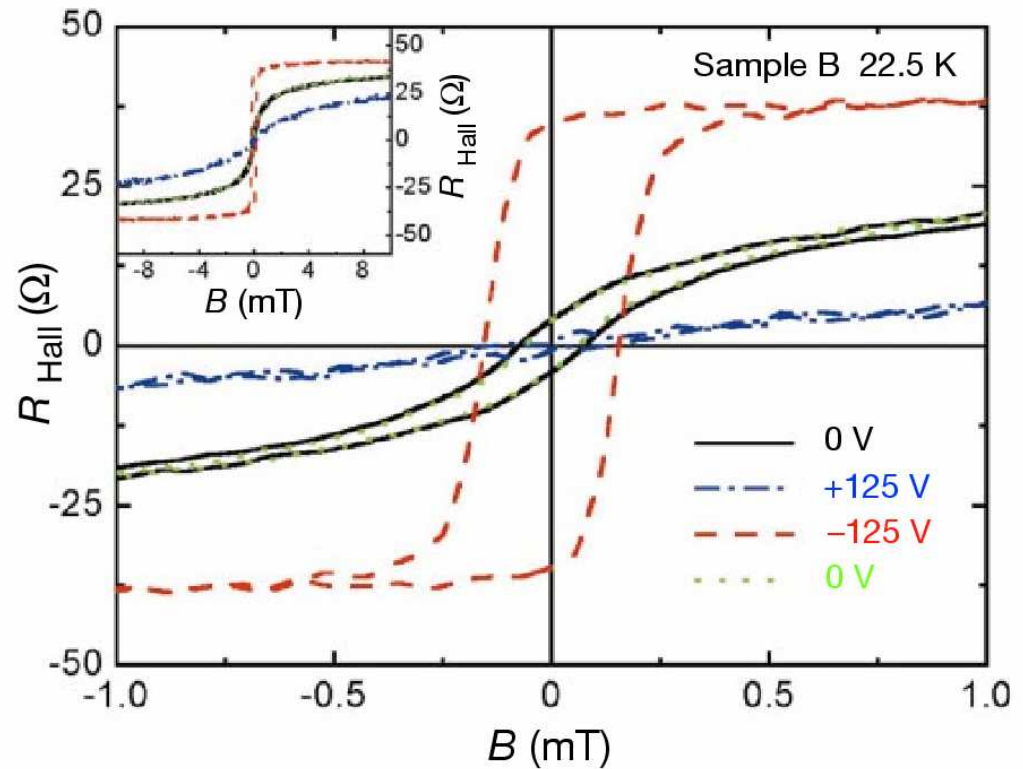
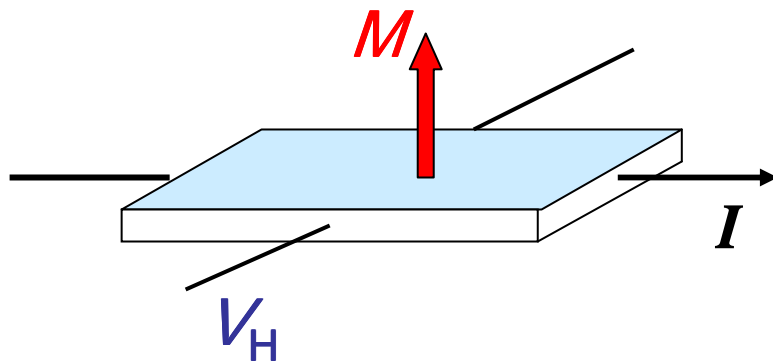
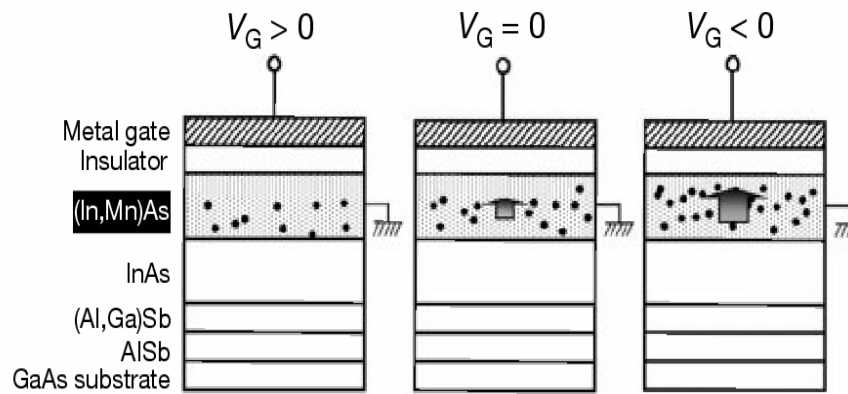
## Magnetisation manipulation by:

- **light** (*Tokyo PRL'97, Grenoble/Warsaw PRL'97,'02*)
- **electric field** (*Tohoku/Warsaw Nature'00,'08 Grenoble/Warsaw PRL'02*)
- **electric current in trilayer structures**  
(*Tohoku PRL'04, Orsay PRB'06*)
- **domain-wall displacement induced by electric current** (*Tohoku, Nature '04, Science'07, Tohoku/Warsaw PRL'06*)

# Tuning of magnetic ordering by electric field (ferro-FET) (In,Mn)As



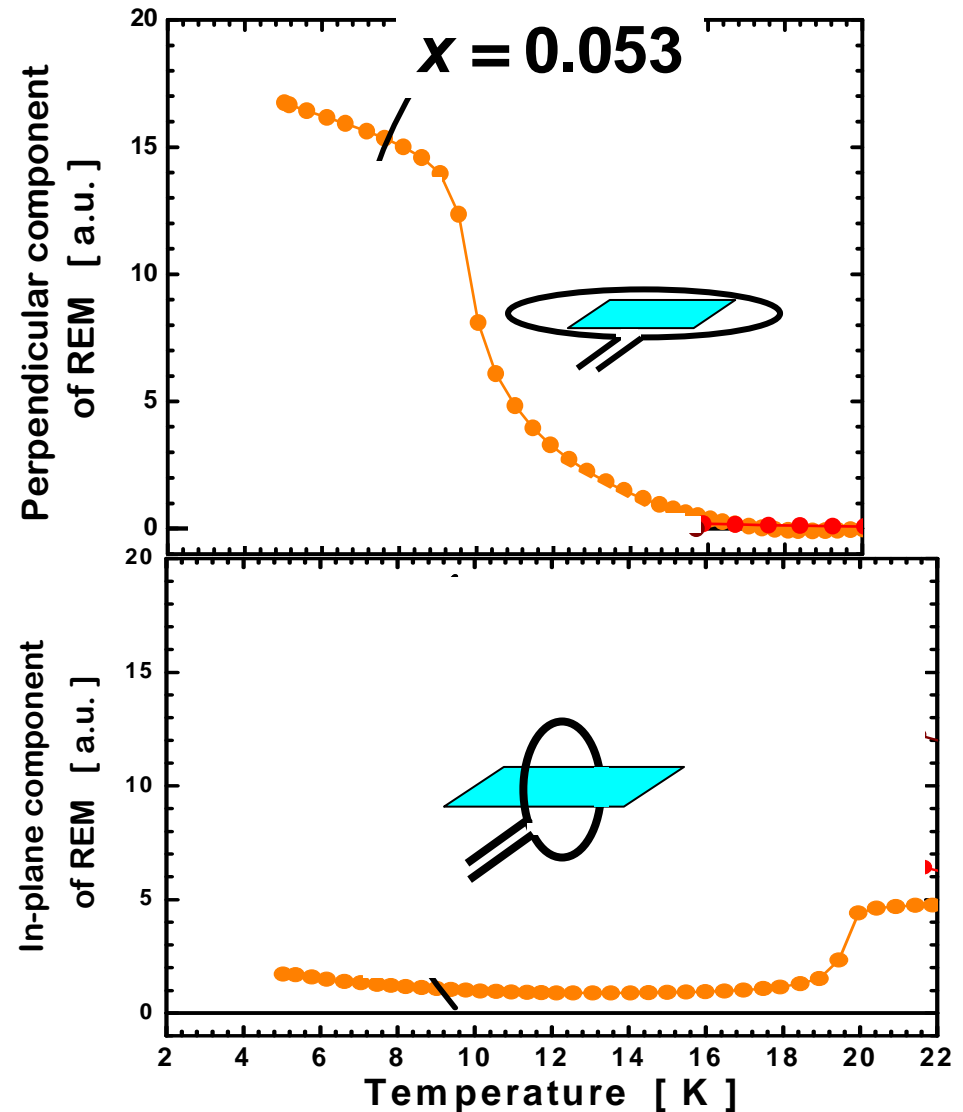
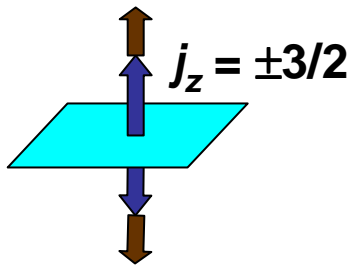
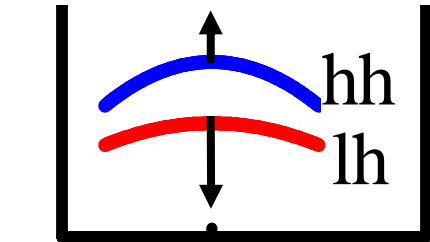
# Tuning magnetic ordering by electric field (ferro-FET) (In,Mn)As



*H. Ohno ... TD., Nature 2000*

# Epitaxial-strain-induced magnetic anisotropy

$\text{Ga}_{1-x}\text{Mn}_x\text{As}/\text{GaAs}$   
 → compressive strain

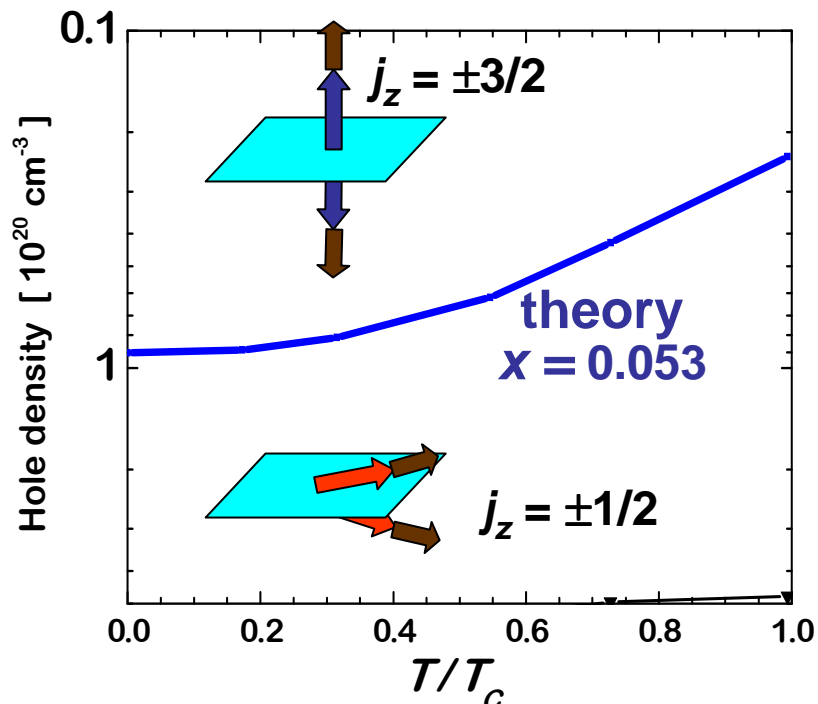
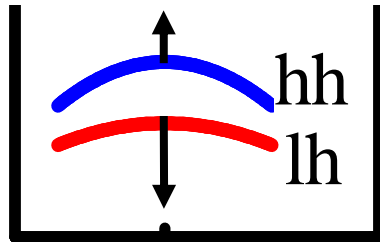


after T.D. et al. (Warsaw, Tohoku) PRB '01

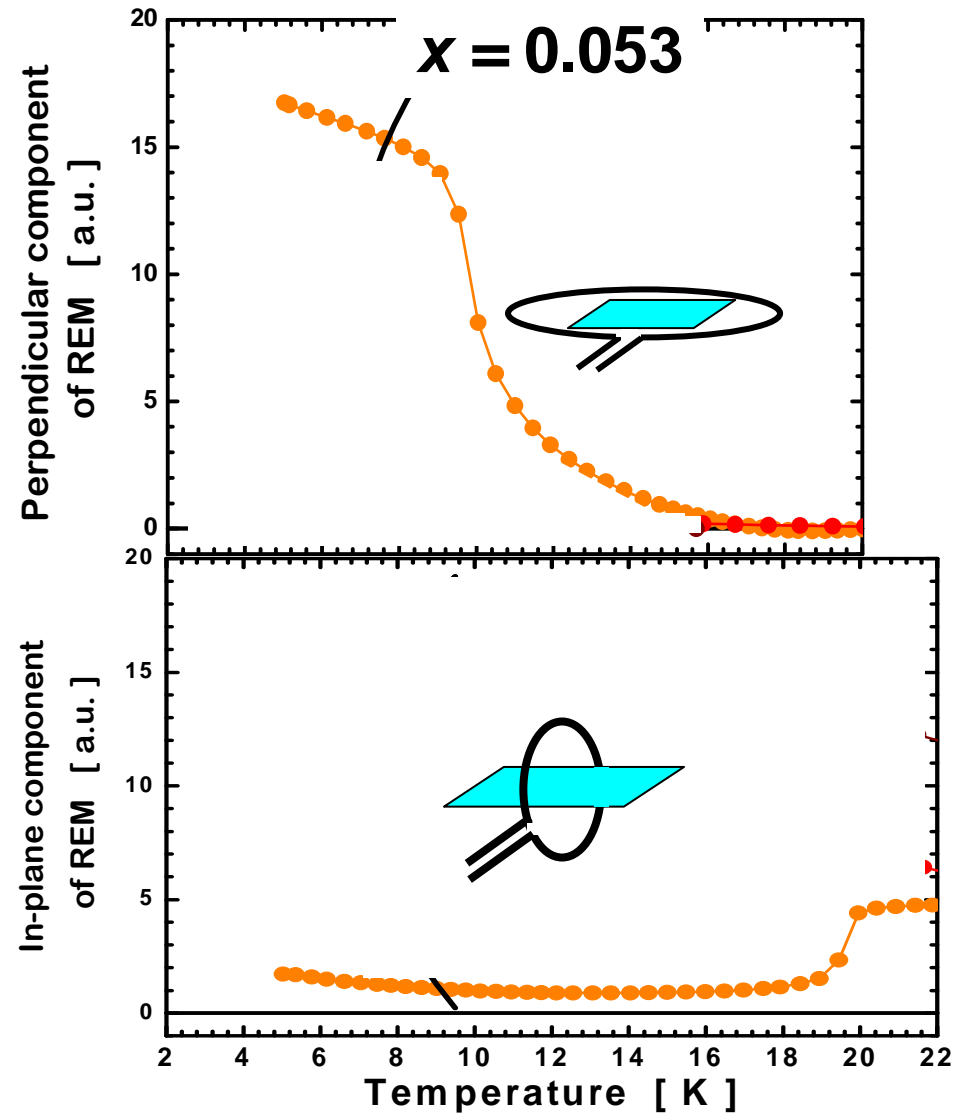
Sawicki et al. (Warsaw, Wuerzburg) PRB'04

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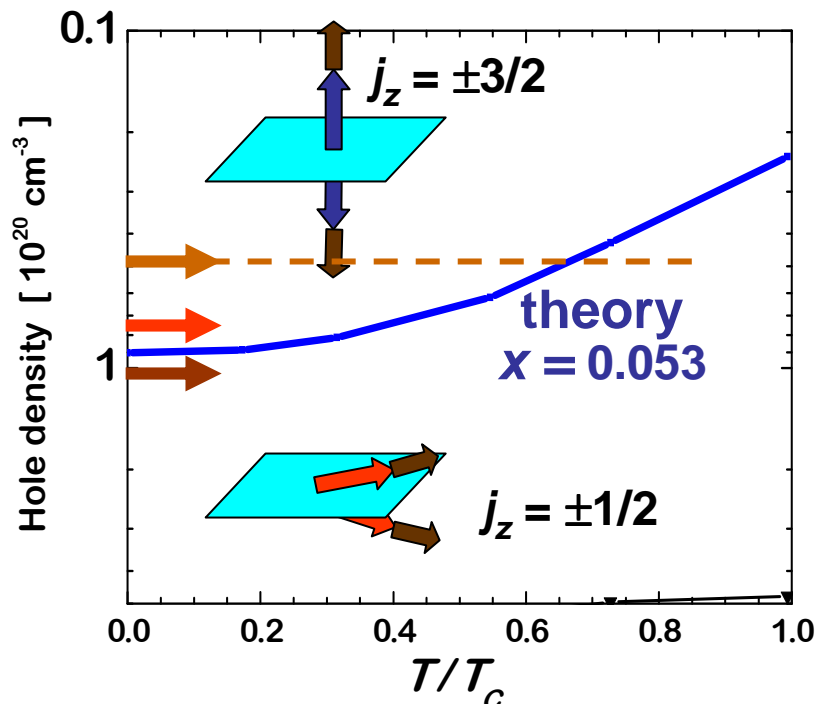
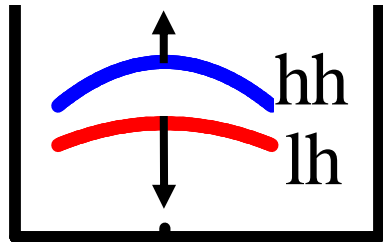
after T.D. et al. (Warsaw, Tohoku) PRB '01



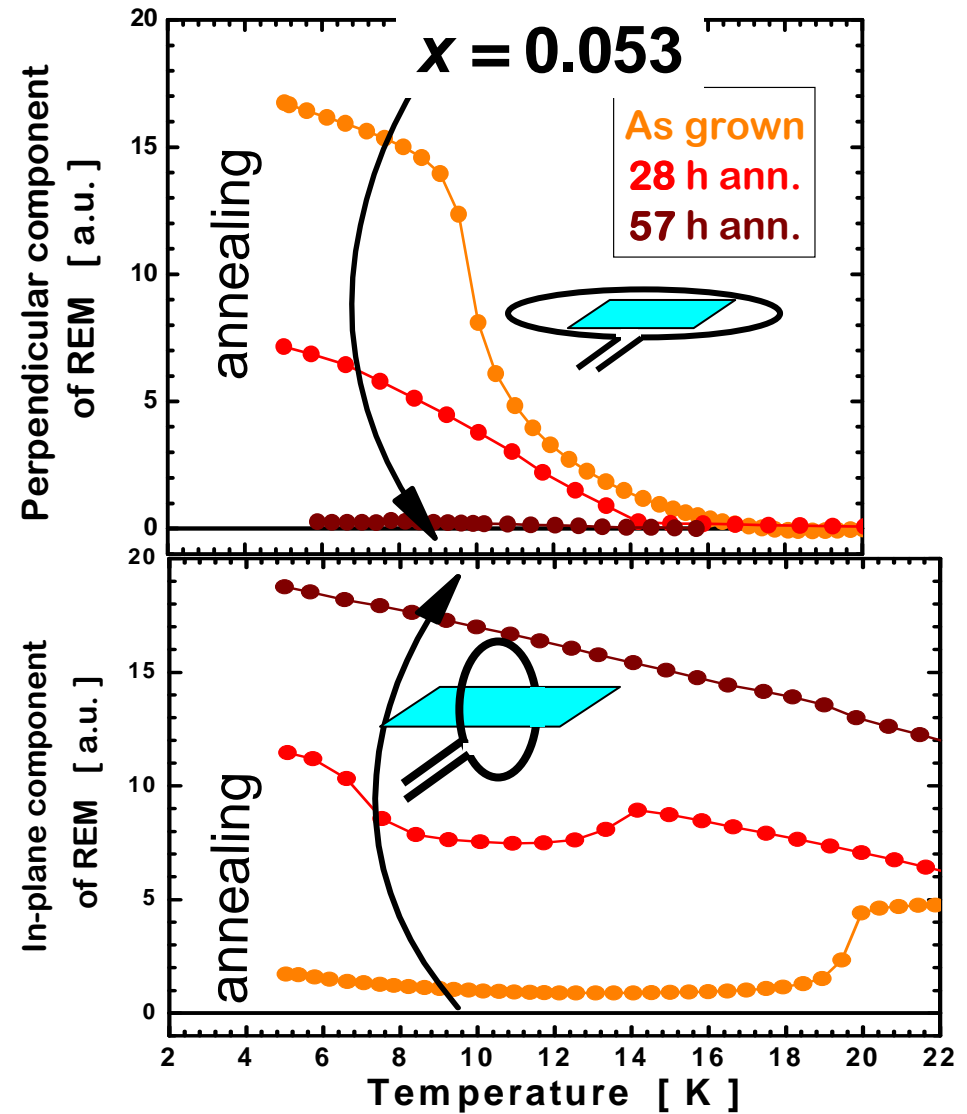
Sawicki et al. (Warsaw, Wuerzburg) PRB'04

# Reorientation transition –theory and expt.

$\text{Ga}_{1-x}\text{Mn}_x\text{As}/\text{GaAs}$   
 → compressive strain



after T.D. et al. (Warsaw, Tohoku) PRB '01



Sawicki et al. (Warsaw, Wuerzburg) PRB'04  
 (Warsaw, Nottingham) RB'05, PRL'05

# Combining FET and anisotropy

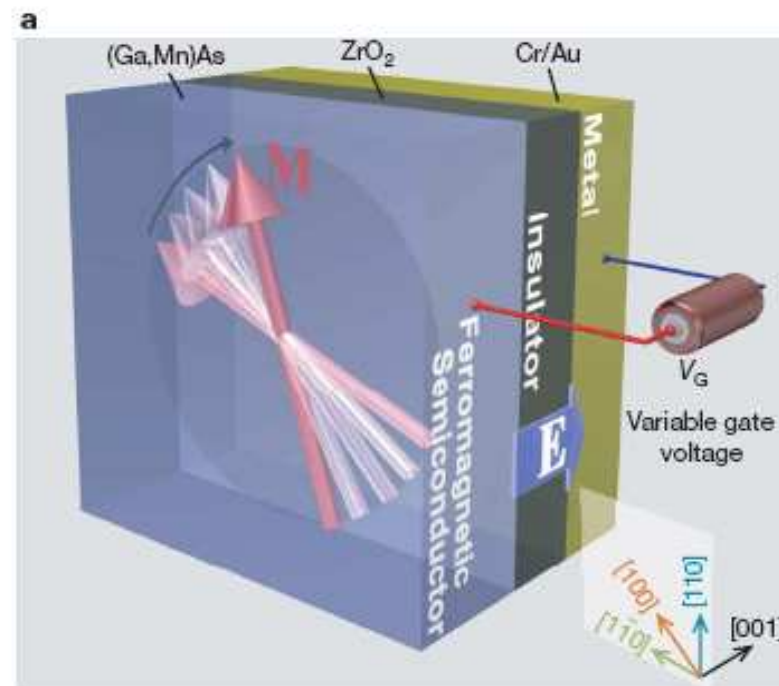
Vol 455|25 September 2008|doi:10.1038/nature07318

nature

## Magnetization vector manipulation by electric fields

D. Chiba<sup>1,2</sup>, M. Sawicki<sup>2,3</sup>, Y. Nishitani<sup>2</sup>, Y. Nakatani<sup>4</sup>, F. Matsukura<sup>2,1</sup> & H. Ohno<sup>2,1</sup>

<sup>1</sup>Semiconductor Spintronics Project, Exploratory Research for Advanced Technology, Japan Science and Technology Agency, Sanban-cho 5, Chiyoda-ku, Tokyo 102-0075, Japan. <sup>2</sup>Laboratory for Nanoelectronics and Spintronics, Research Institute of Electrical Communication, Tohoku University, Katahira 2-1-1, Aoba-ku, Sendai 980-8577, Japan. <sup>3</sup>Institute of Physics, Polish Academy of Sciences, Al. Lotników 32/46, PL-02668, Warszawa, Poland. <sup>4</sup>University of Electro-communications, Chofugaoka 1-5-1, Chofu, Tokyo 182-8585, Japan.



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# Strategies for high $T_C$ in hole-controlled DMS

- **Strategies:**
  - increasing  $p$  and/or  $x$  in existing ferromagnetic DMS (Ga,Mn)As

*T.D. et al. (Warsaw, Tohoku, Grenoble) Science'00*



# Strategies for high $T_C$ in hole-controlled DMS

- **Strategies:**
  - increasing  $p$  and/or  $x$  in existing ferromagnetic DMS (Ga,Mn)As
  - searching for DMS with greater coupling constant  $x\beta^2\rho(E_F)$   
nitrides, oxides

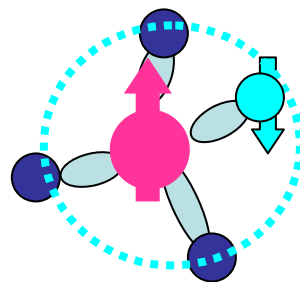
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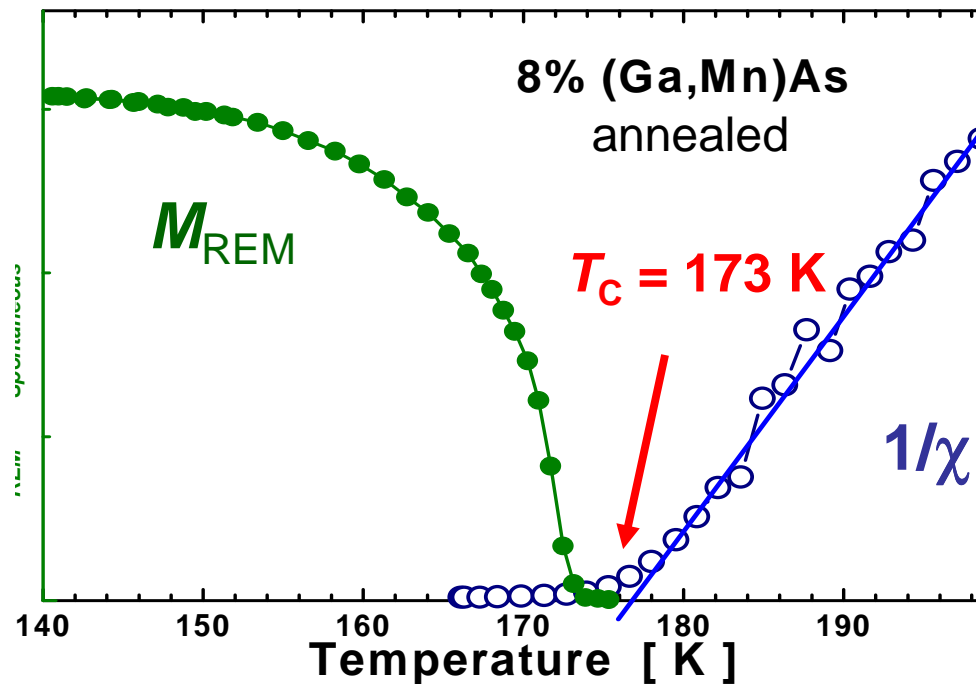
- **Obstacles:**

- self-compensation
- tight binding of holes by TM ions  
(Zhang-Rice singlet)
- solubility limits

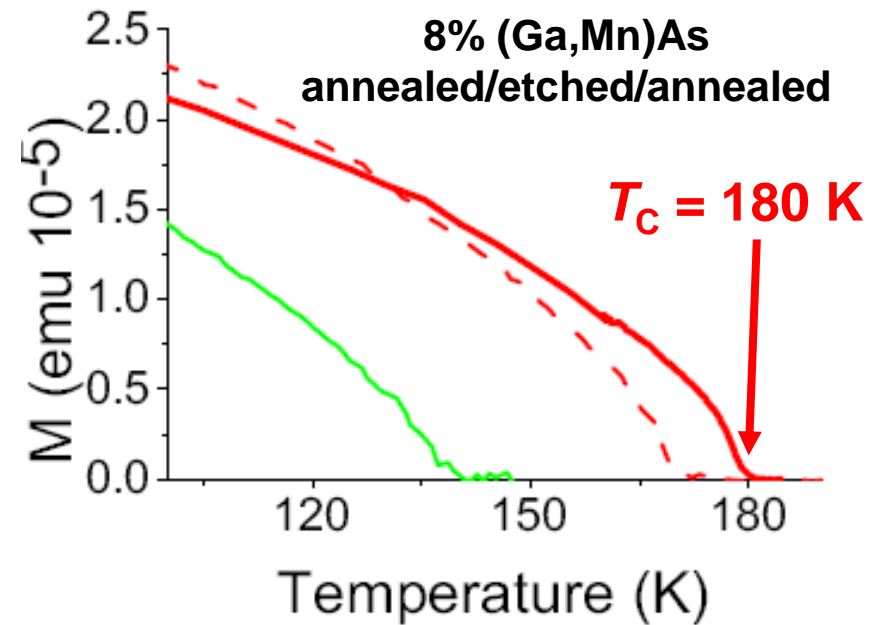


# Where are we? (Ga,Mn)As

remanent magnetization and  $1/\chi$  vs.  $T$



remanent magnetization



*Olejnik et al. (Prague) cond-mat/08*

*Wang/ Sawicki (Nottingham, Warsaw) ICPS'04*

*Sendai, Tokyo, Notre Dame, PSU, Lund, Wuerzburg, IMEC, UCSB,...*

# Self-compensation

# $T_C$ in (Ga,Mn)As

The progress due to increase of  $p$  by low temperature annealing →

**out diffusion of  $Mn_I$ :**

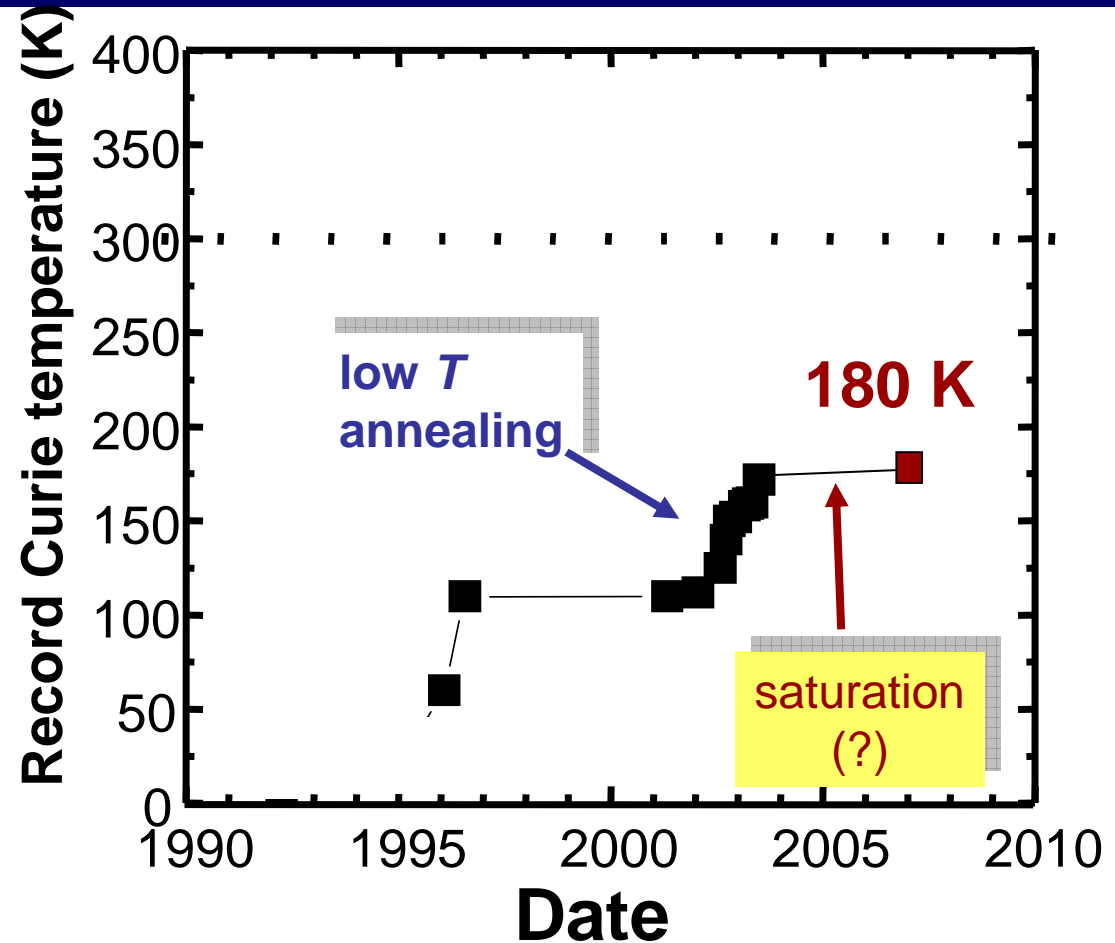
*Máca and Mašek (Prague)*

*PRB'02, Yu et al. (Berkeley,*

*Notre Dame) PRB'02; APL'04*

*Edmonds et al. (Nottingham,*

*Warsaw) PRL'04*



*Nottingham, Sendai, Tokyo, Notre Dame, PSU, Lund, Wuerzburg, IMEC, St. Barbara, Prague...*

**Beyond (Ga,Mn)As**  
**Strong coupling**

# Chemical trends in exchange energy $\beta N_0$

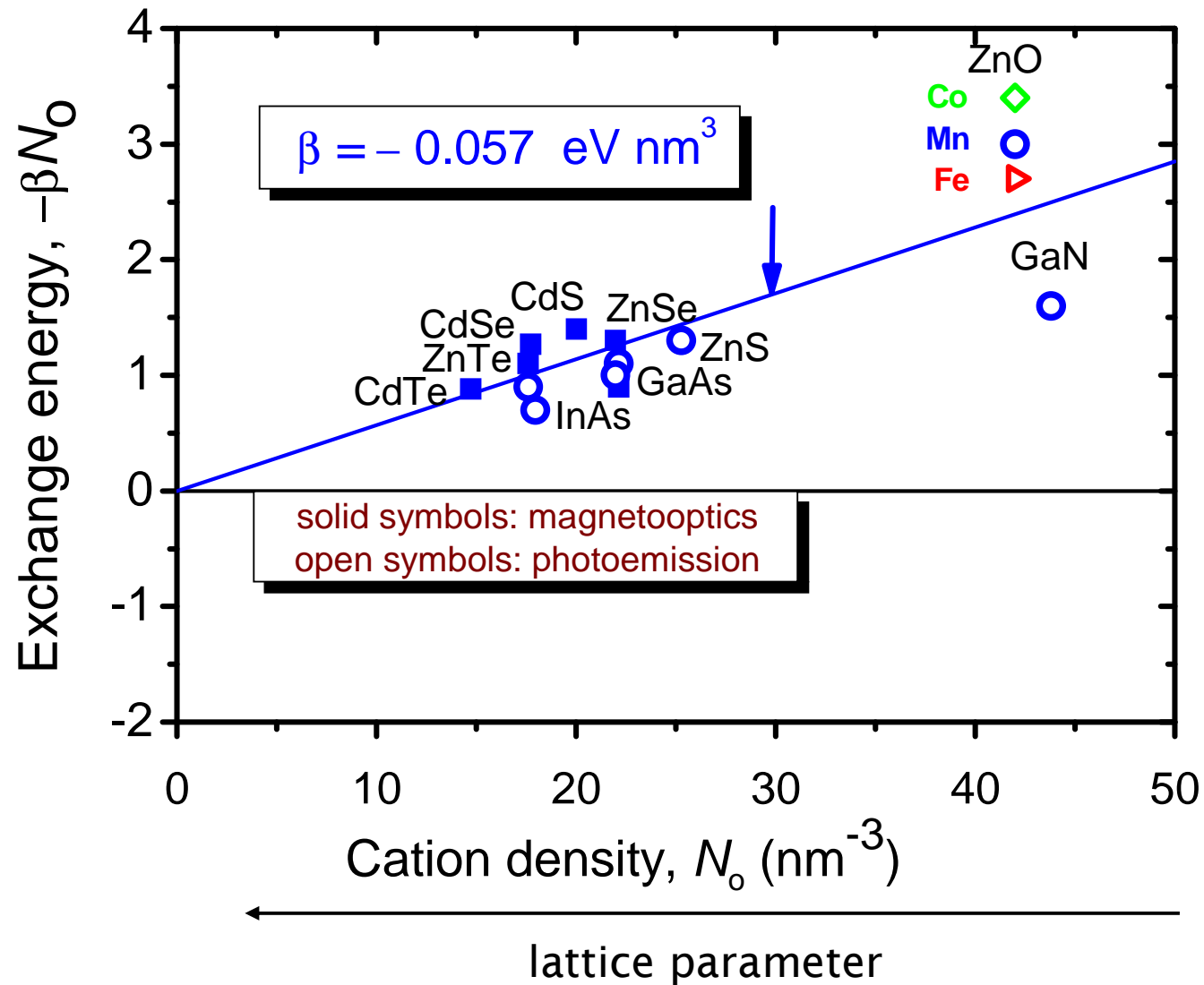


photo-emission  
XAS  
Fujimori et al.  
(Tokyo)

# Exchange energy $\beta N_0$

## Photoemission vs. magneto-optics

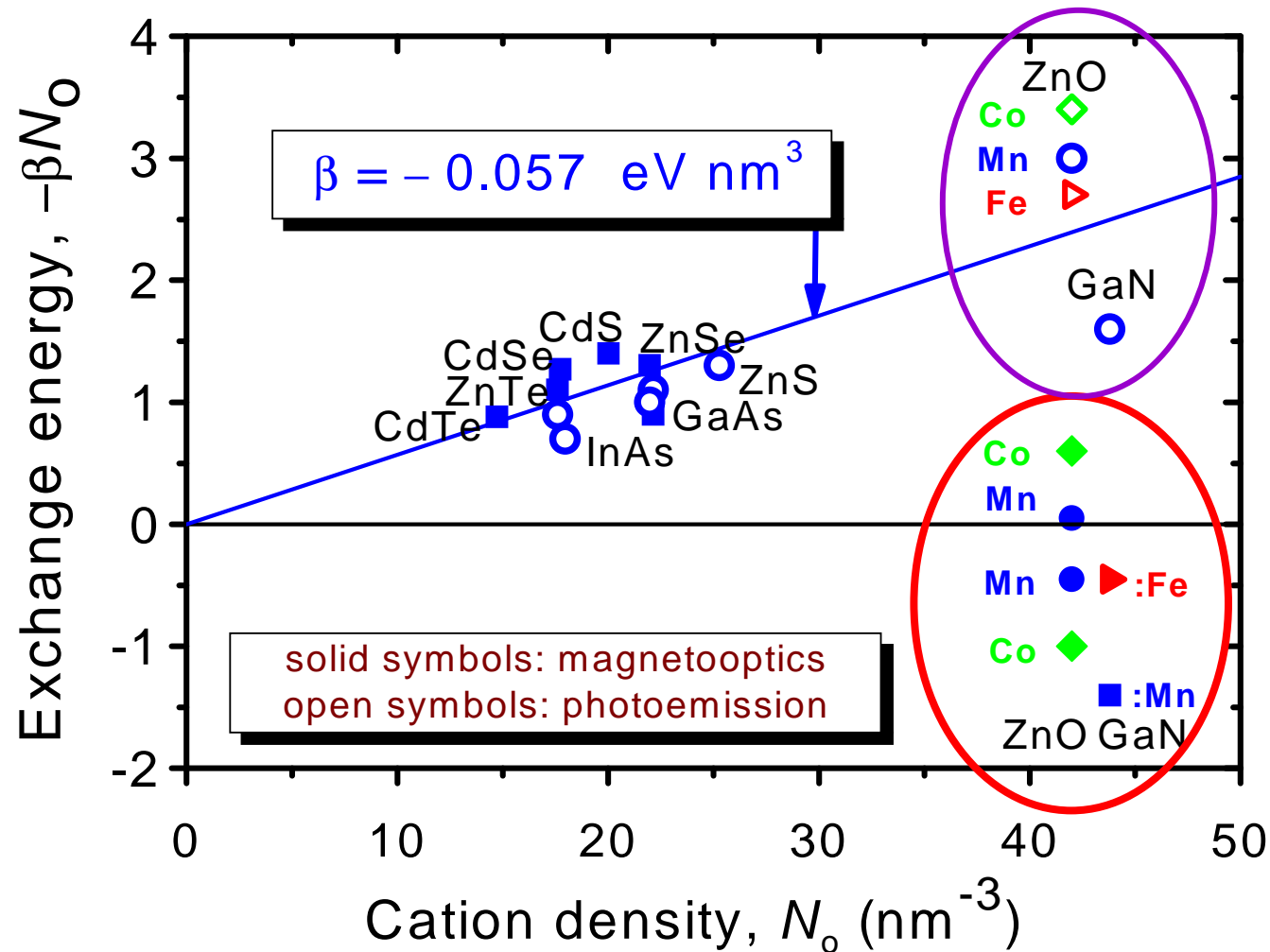


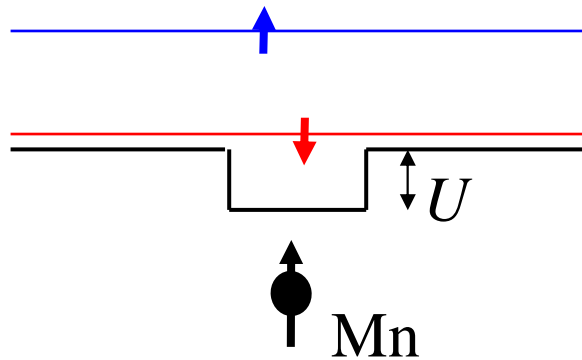
photo-emission  
XAS  
*Fujimori et al.*  
(Tokyo)

magneto-optics  
*Pacuski et al.*  
(Warsaw/Grenoble/Linz)  
PRB'06'07, PRL'08



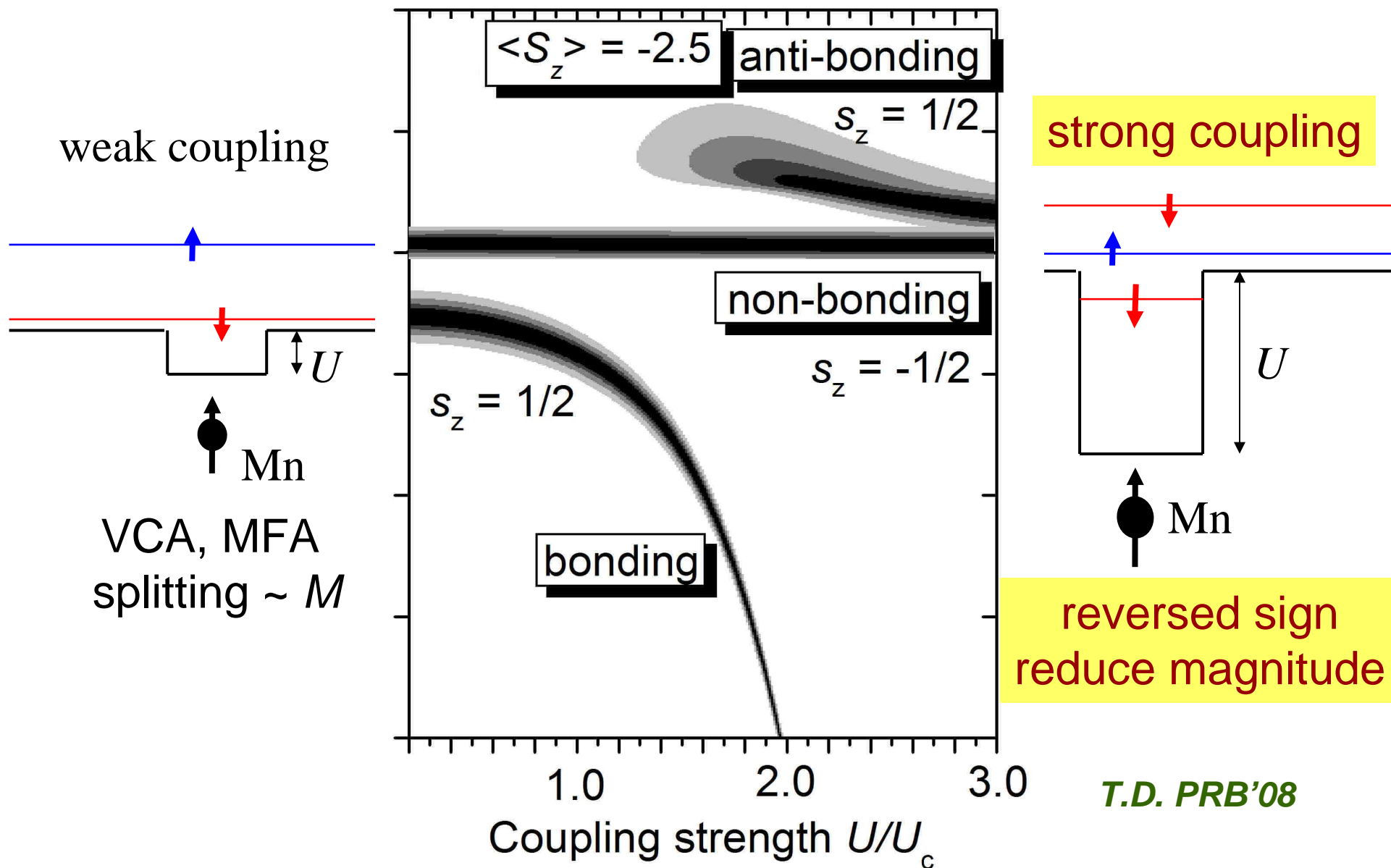
# Spin splitting

weak coupling



VCA, MFA  
splitting  $\sim M$

# Spin splitting



# Effect of strong coupling on ferromagnetism

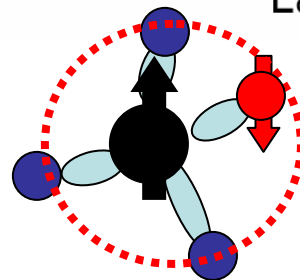
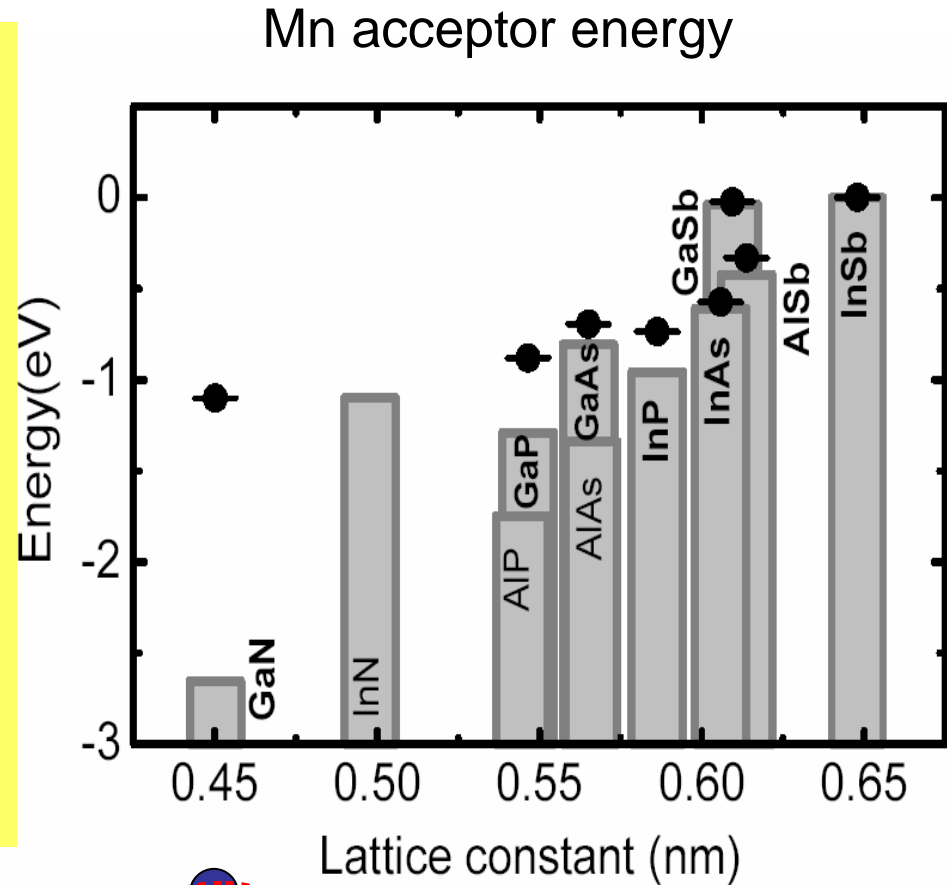
deep hole trap formed



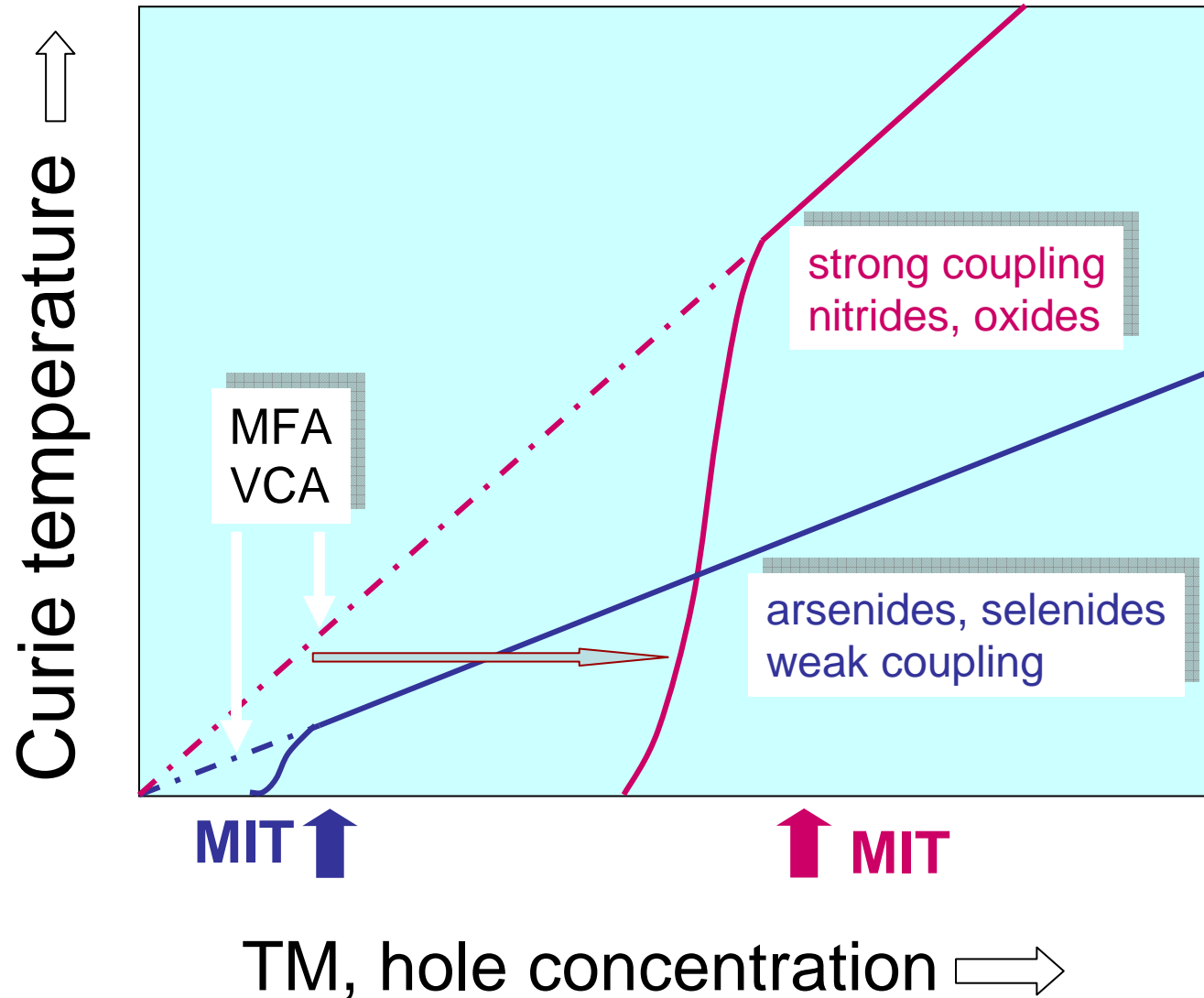
no holes



no efficient ferromagnetism according to *p-d* Zener model

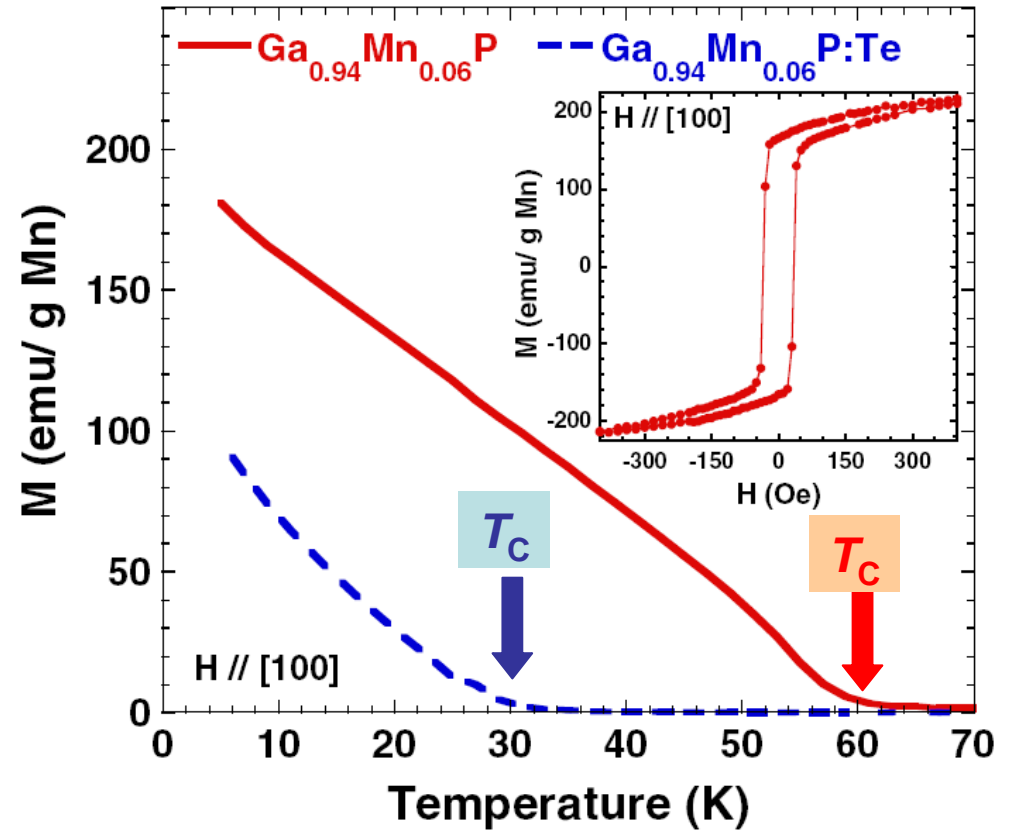
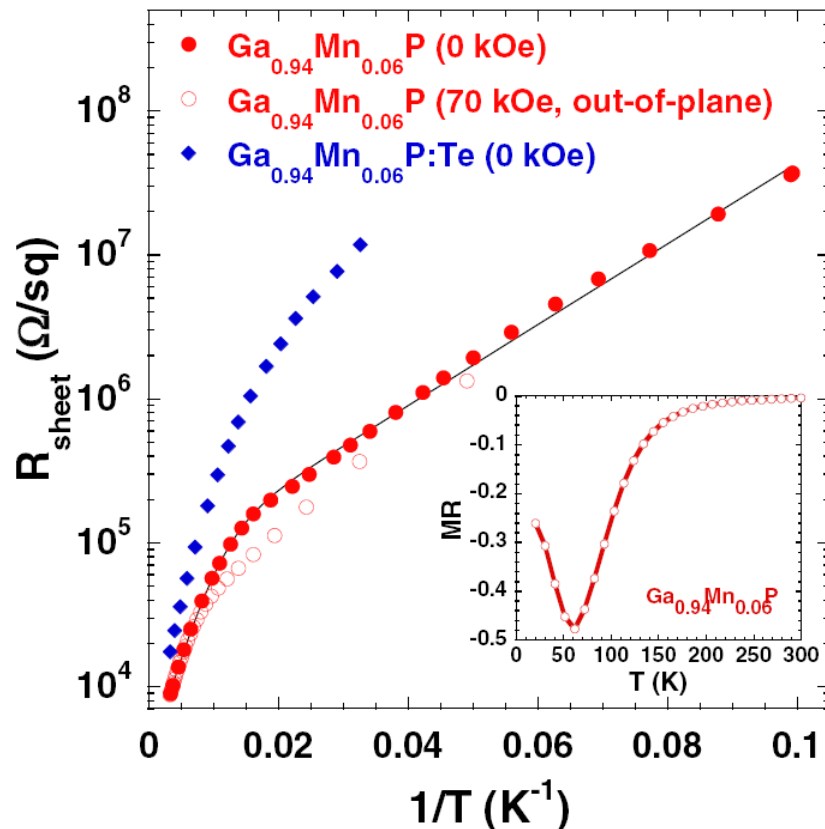


# Effect of strong $p$ - $d$ coupling on $T_C$ in p-type DMS



# Properties of implanted $\text{Ga}_{1-x}\text{Mn}_x\text{P}$

$x = 6\%$ ,  $T_C \cong 55 \text{ K}$

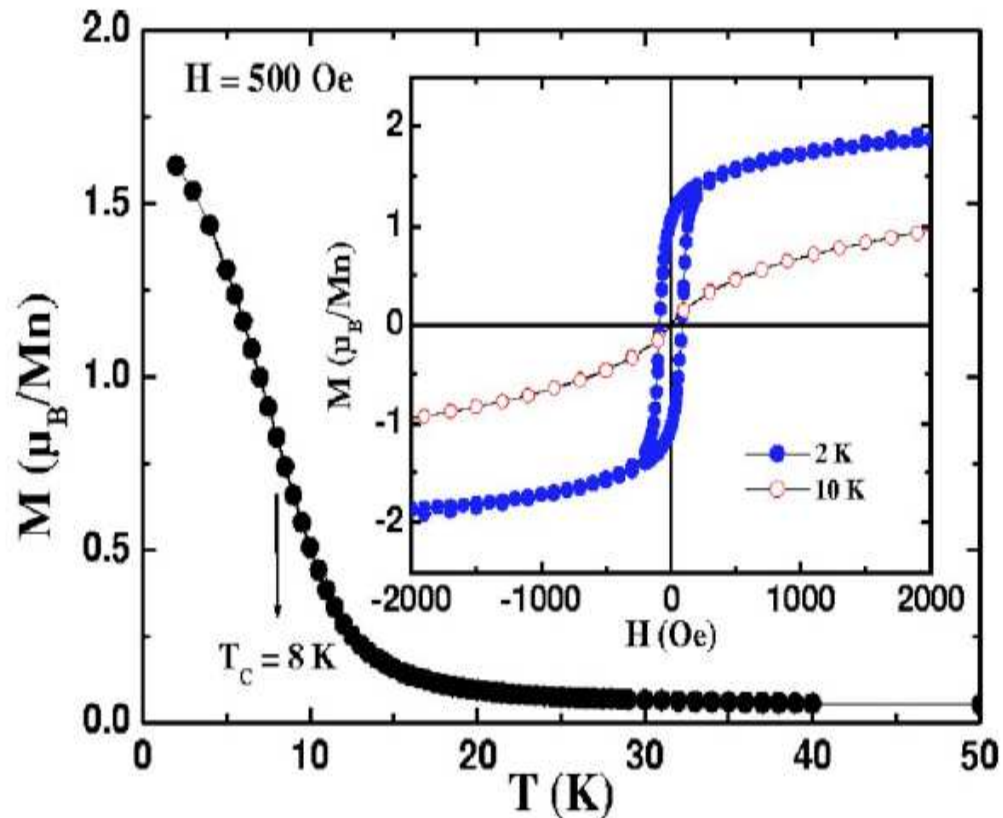


localisation

Scarpulla et al. (Berkeley) PRL '05

# Magnetic properties of uniform $\text{Ga}_{1-x}\text{Mn}_x\text{N}$

$x = 6\%$ ,  $T_C \cong 8 \text{ K}$



*Sarigiannidou et al. (Grenoble) PRB'06*

weak FM

# DMS, DMO, and *non*-magnetic materials showing spontaneous magnetization at 300 K

wz-c-(Ga,Mn)N, (Ga,Fe)N, (In,Mn)N, (Al,Mn)N, (Ga,Cr)N, (Al,Cr)N

(Zn,Mn)O, (Zn,Ni)O, (Zn,Co)O, (Zn,V)O, (Zn,Fe,Cu)O, (Zn,Cu)O

(Zn,Cr)Te

(Ti,Co)O<sub>2</sub>, (Ti,V)O<sub>2</sub>, (Ti,Cr)O<sub>2</sub>, (Sn,Co)O<sub>2</sub>, (Sn,Fe)O<sub>2</sub>, (Hf,Co)O<sub>2</sub>

(Ga,Mn)As, (In,Mn)As, (Ga,Mn)Sb, (Ga,Mn)P:C

(Cd,Ge,Mn)P<sub>2</sub>, (Zn,Ge,Mn)P<sub>2</sub>, (Cd,Ge,Mn)As<sub>2</sub>, (Zn,Sn,Mn)As<sub>2</sub>

(Ge,Mn), (Ge,Cr), (Ge,Mn,Fe), (Si,Fe), (Si,Mn), (SiC,Fe)

no valence band holes

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(Ge,Mn), (Ge,Cr), (Ge,Mn,Fe), (Si,Fe), (Si,Mn), (SiC,Fe)

(La,Ca)B<sub>6</sub>, CaB<sub>2</sub>C<sub>2</sub>, C, C<sub>60</sub>, HfO<sub>2</sub>, ZnO, (Ga,Gd)N, (Ga,Eu)N ...

no valence band holes, no TM ions



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no valence band holes, no TM ions  
often supported by DFT computations  
... no devices ...

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origin of UFO ?

# SQUID studies of DMS and DMO in Warsaw

*M. Sawicki et al. (Warsaw):*

wz-c-(Ga,Mn)N, (Ga,Fe)N

(Ga,Mn)As

(Zn,Mn)Te:N, P

(Cd,Mn)Te, (Cd,Mn)Se

(Cd,Cr)Te, (Zn,Cr)Se

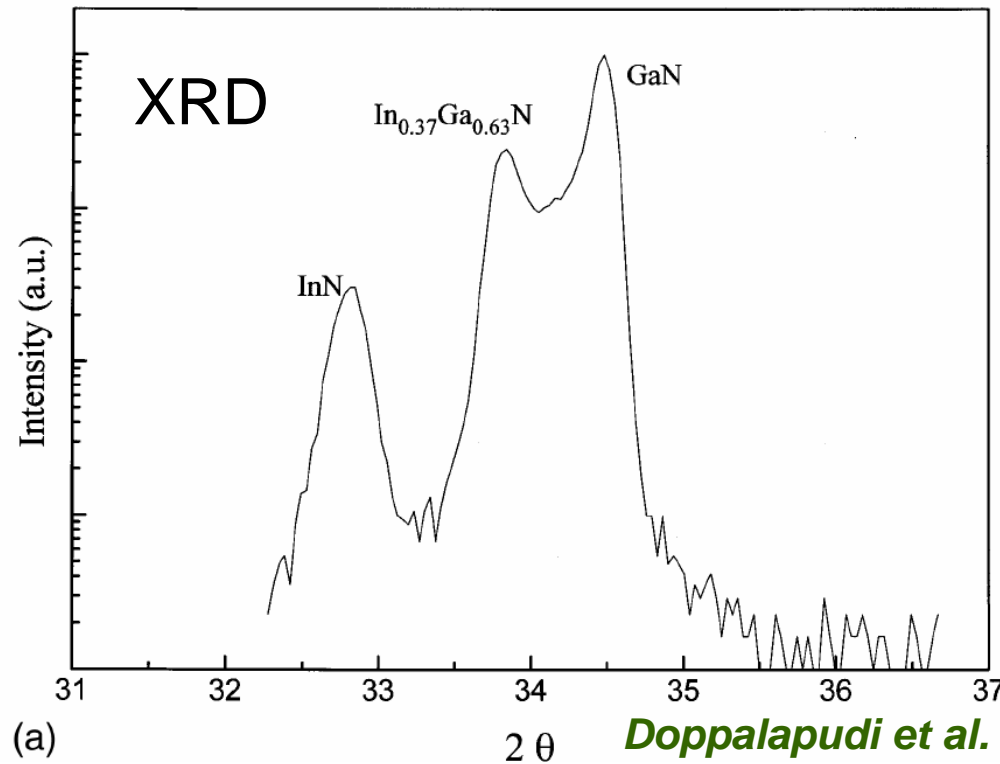
(Zn,Mn)O, (Zn,Co)O, (Zn,Cr)O

# Experimental challenges

- precipitates of magnetic metals (Co, Fe)
- contamination by magnetic nanoparticles (growth, processing)
- sample but also substrate, interface, holder,...
- SQUID: residual fields, software, ....

**Beyond solubility limit**

# Spinodal decomposition in MBE-grown (Ga,In)N



*Doppalapudi et al. (Boston) JAP'98*  
*Liliental-Weber et al. (Berkeley), JEM'05*

**controlled by:**

- solubility limit?
- surface phase separation and kinetic barrier?
- ??

# Model for high $T_C$ DMS

## *Model for DMS and DMO beyond solubility limit*

- phase separation or spinodal decomposition into regions with low and high concentration of magnetic constituents
- FM or AFM regions with high concentration determine magnetic properties (through high blocking temperature)

*T. D. ICPS'04, MRS'04, Nature Mat. Sept.'06 -  
K. Sato, H. Katayama-Yoshida, P. Dederichs, JAAP'05-  
S. Kuroda et al. (Tsukuba/Warsaw) Nature Mat.'07*



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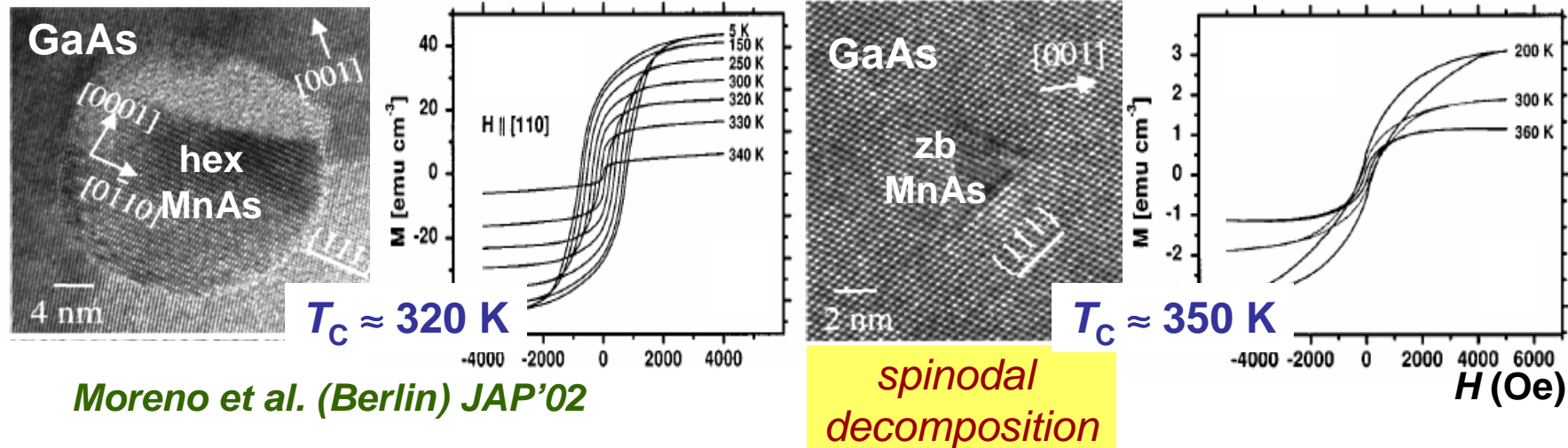
*T. D. ICPS'04, MRS'04, Nature Mat. Sept.'06 -  
K. Sato, H. Katayama-Yoshida, P. Dederichs, JAAP'05-  
S. Kuroda et al. (Tsukuba/Warsaw) Nature Mat.'07*

### **spinodal decomposition** hard to detect:

- crystallographic structure remains uniform
- magnetic regions may gather at surface or interface
- element specific 3D nanoanalyzer required

# Phase separations in (Ga,Mn)As

- depending on growth conditions precipitates or spinodal decomposition



*Moreno et al. (Berlin) JAP'02*

*spinodal decomposition*

*Mn-rich regions:*

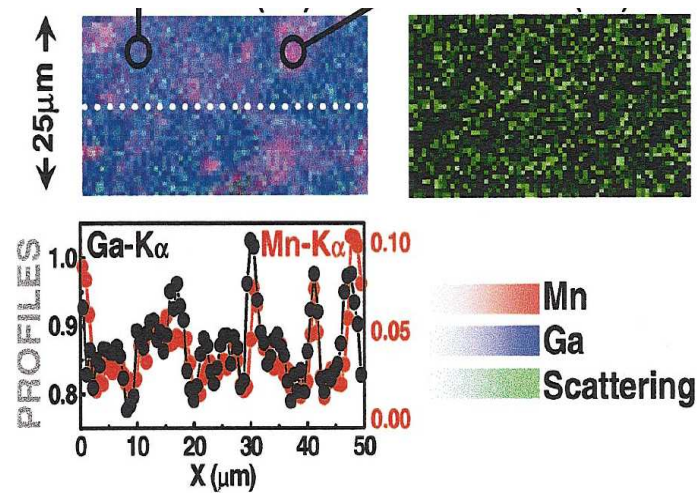
- control magnetic properties *De Boeck et al. (IMEC) APL'96*
- enhance magneto-optical effects (MCD) *Akinaga et al. (Tsukuba) APL'00;*  
*Shimizu et al. (Tokyo) APL'01; Yokoyama et al. (Tokyo) JAP'05*
- affect conductance and Hall effect *Heimbrodt et al. (Marburg) PRB'04*

DMS with spinodal decomposition  
→ functional high  $T_C$  composite systems

# Spinodal decomposition in DMS from TEM + EDS

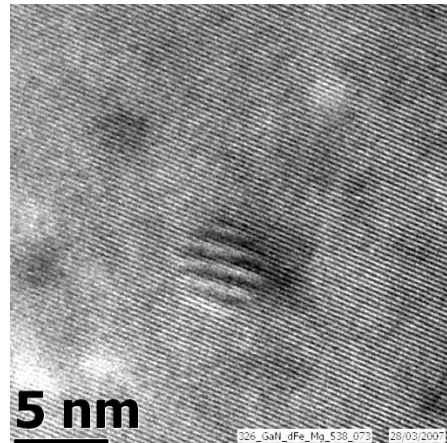
## Dots

(Ga,Mn)N



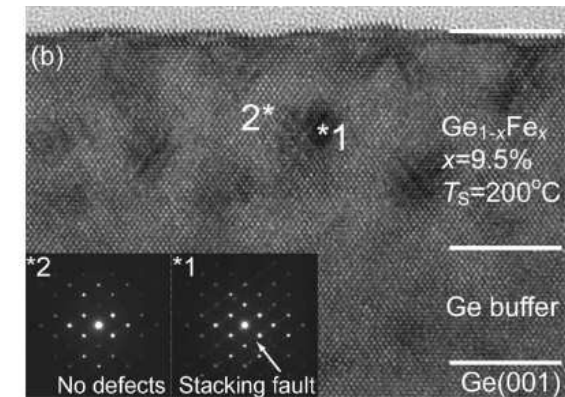
*Martínez-Criado, et al.*  
(Grenoble) APL'05

(Ga,Fe)N



*Bonanni (Linz/Warsaw)*  
PRB'07

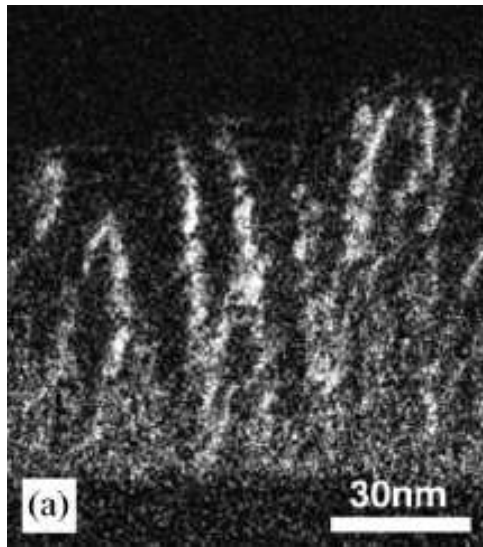
(Ge,Fe)



*Shuto et al. (Tokyo) APL'07*

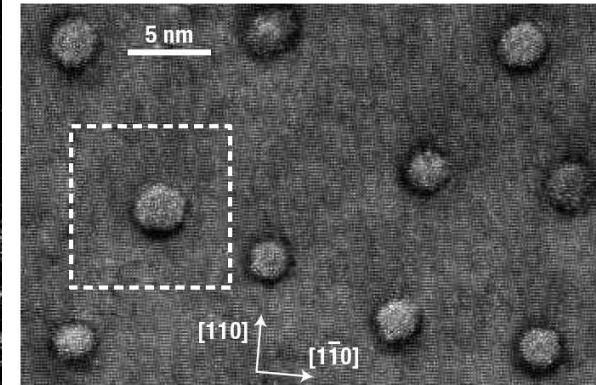
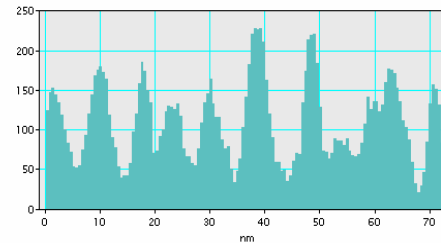
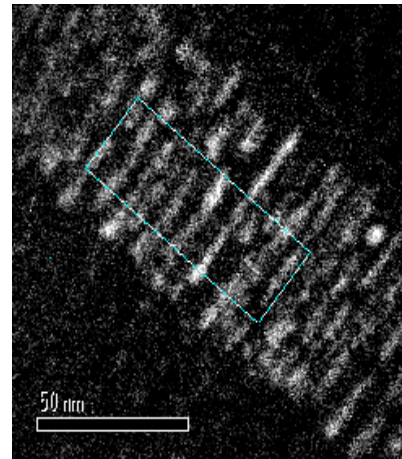
# Nanocolumns

(Al,Cr)N



*Guo et al. (Arizona)  
JMMM'06*

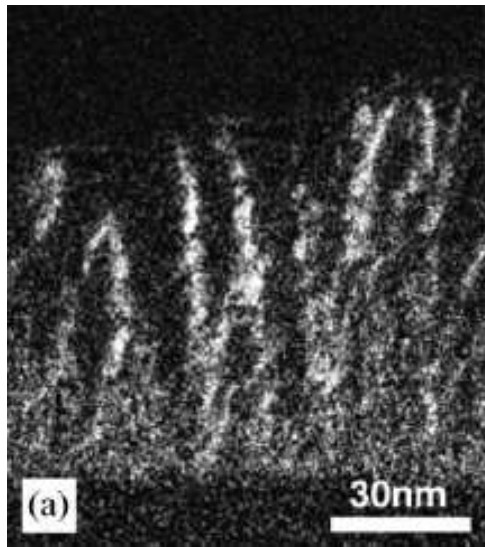
(Ge,Mn)



*Jamet et al. (Grenoble)  
Nature Mat. '06*

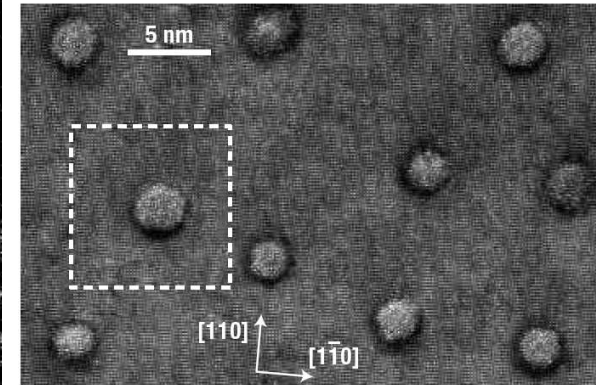
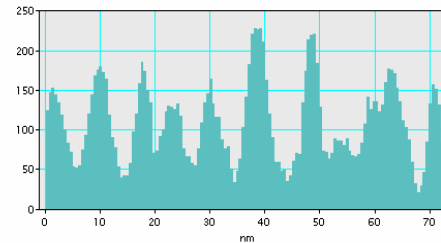
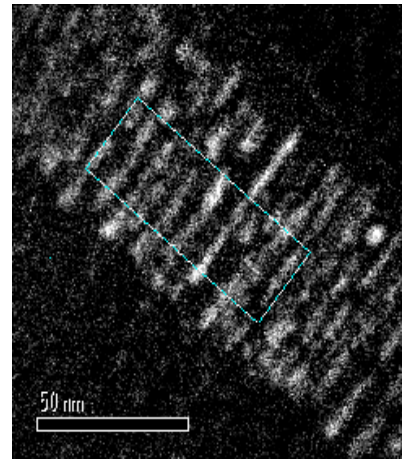
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*Guo et al. (Arizona)  
JMMM'06*

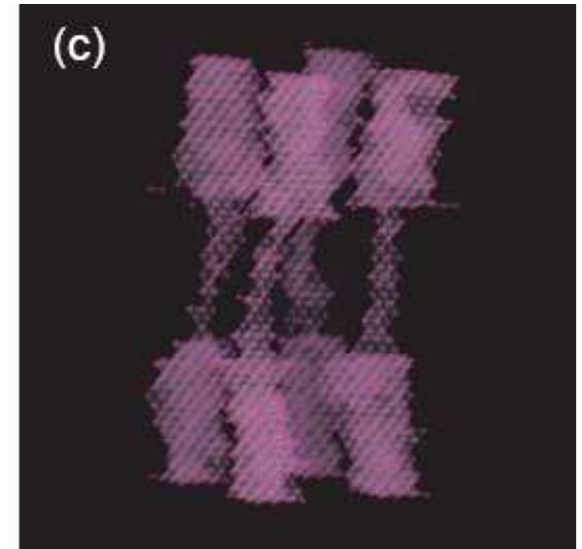
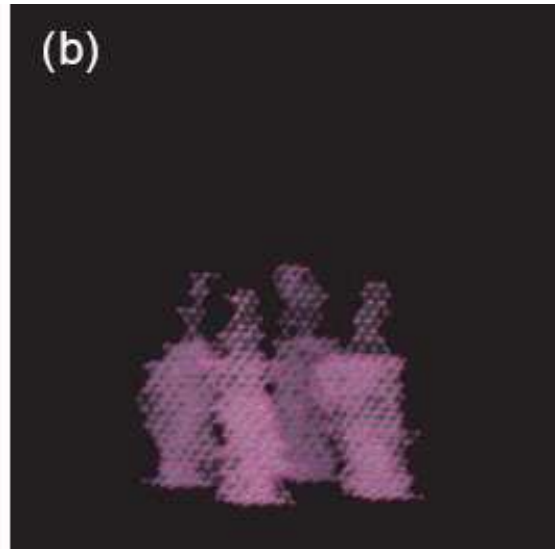
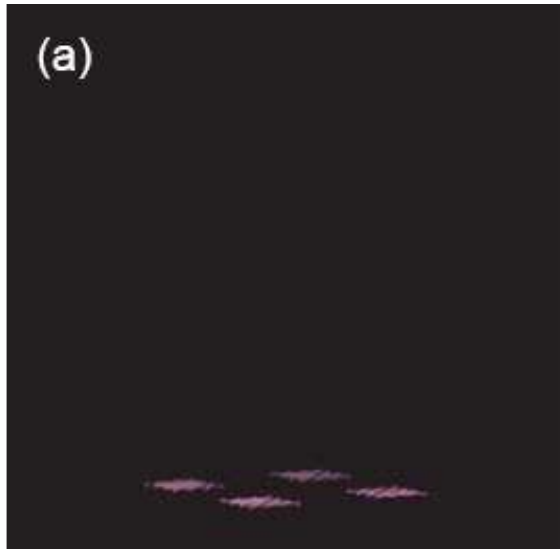
(Ge,Mn)



*Jamet et al. (Grenoble)  
Nature Mat. '06*

**Nanomagnets in form  
of dots or nanocolumns**

# Formation of tunnel/Coulomb blockade junctions

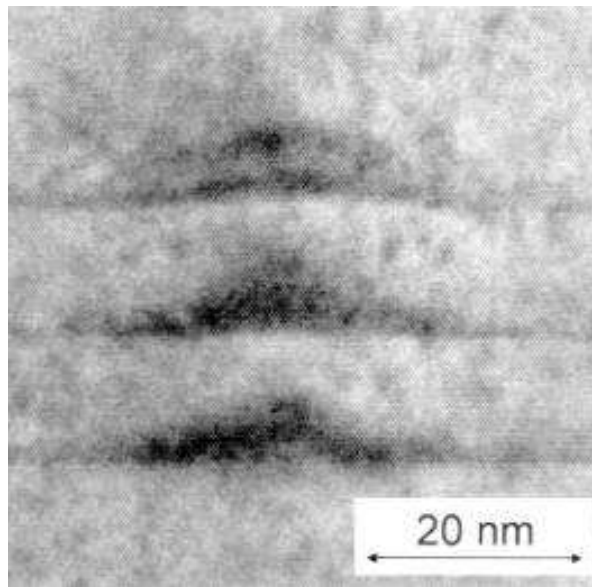
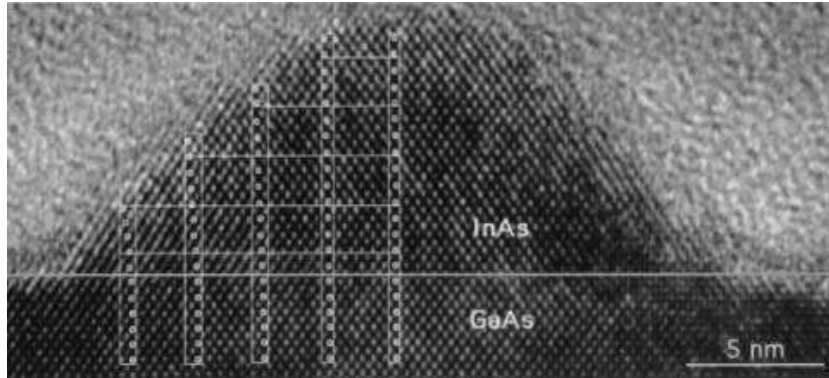


*Fukushima et al. (Osaka) phys.stat.sol. (c)'06*

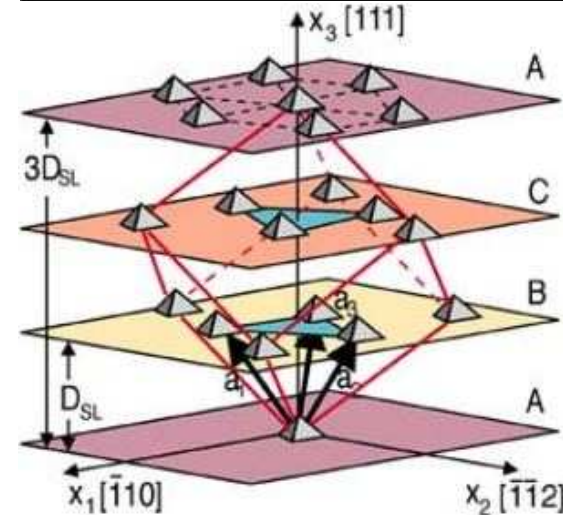
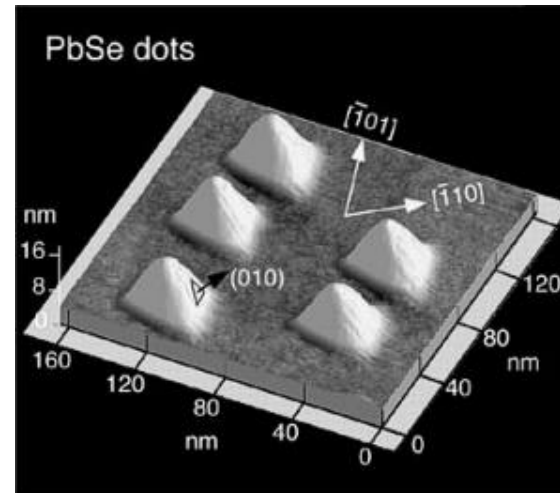
How to control spinodal decomposition?

# Self-organized quantum dots in semiconductors

InAs in GaAs



PbSe in (Pb,Eu)Te



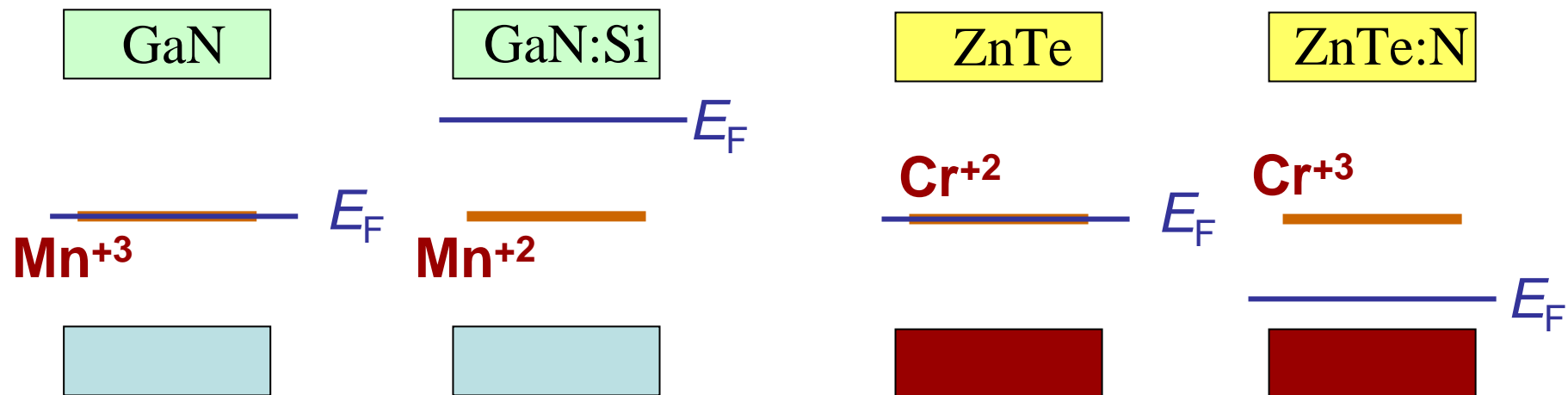
*Springholz et al.  
(Linz) Science'98*

**spinodal decomposition controlled by epitaxial strain**



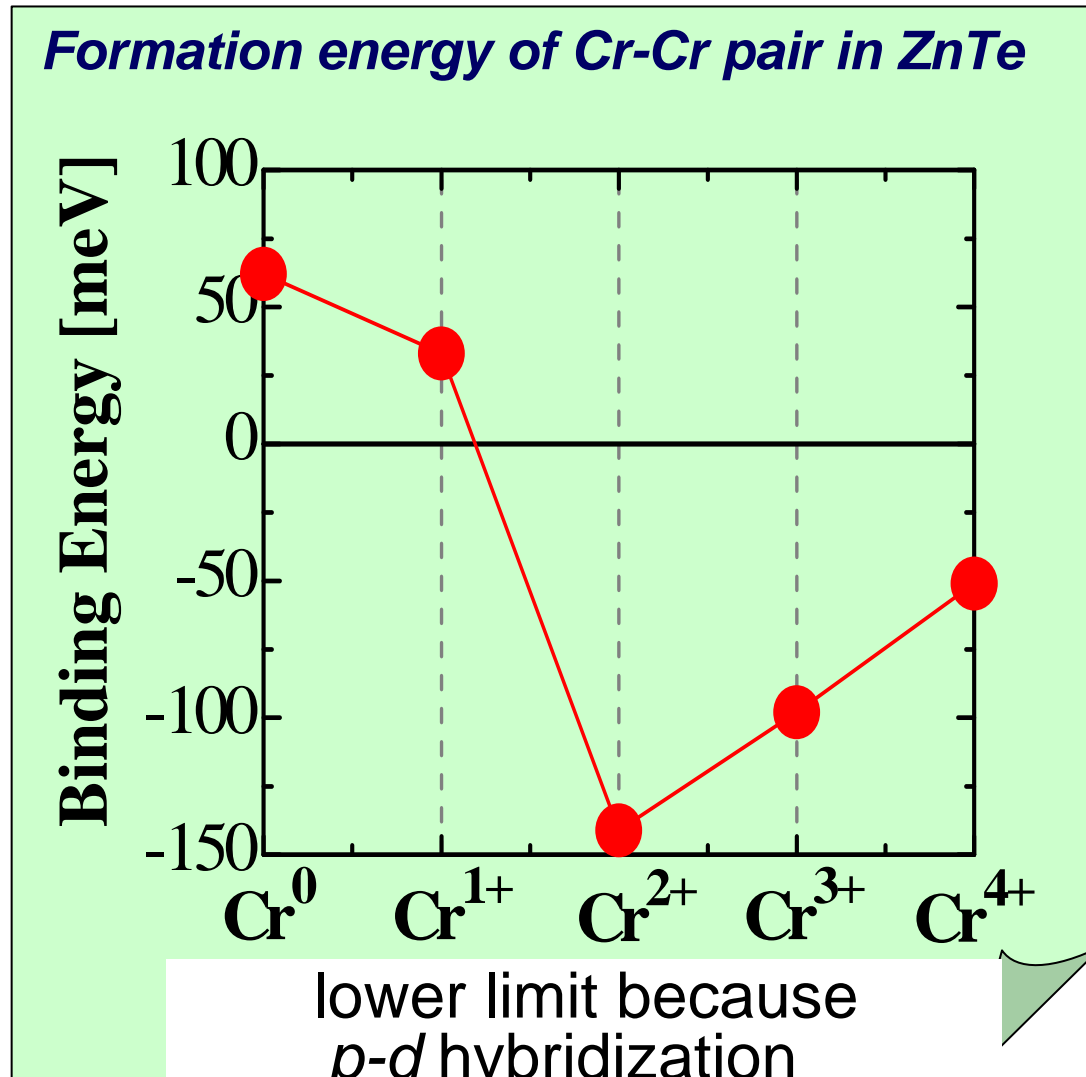
# Effect of doping on spinodal decomposition

TM charge state is controlled by co-doping with shallow impurities. Because of Coulomb repulsion **spinodal decomposition is blocked if TM is charged**



*T.D., Nature Mat.'06  
L.-H. Ye et al. (NWU) PRB'06*

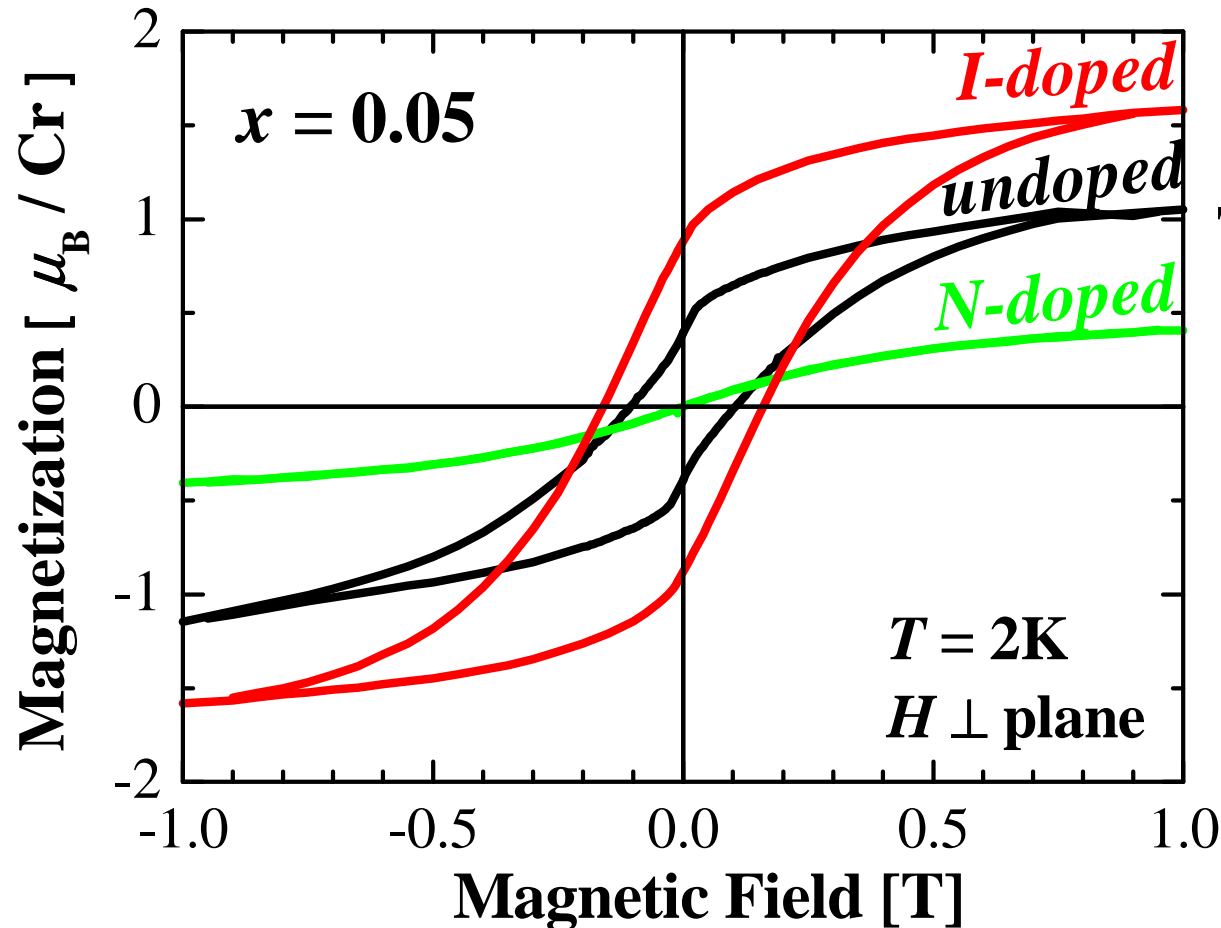
# Effect of charge state on binding energy of Cr pair



lower limit because  
*p-d* hybridization  
overestimated by LSDA

*S. Kuroda ... T.D.  
(Tsukuba, Warsaw)  
Nature Mat., '07*

# Effect of co-doping on magnetism



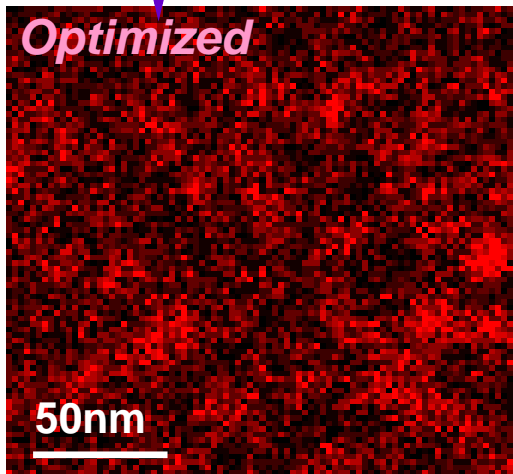
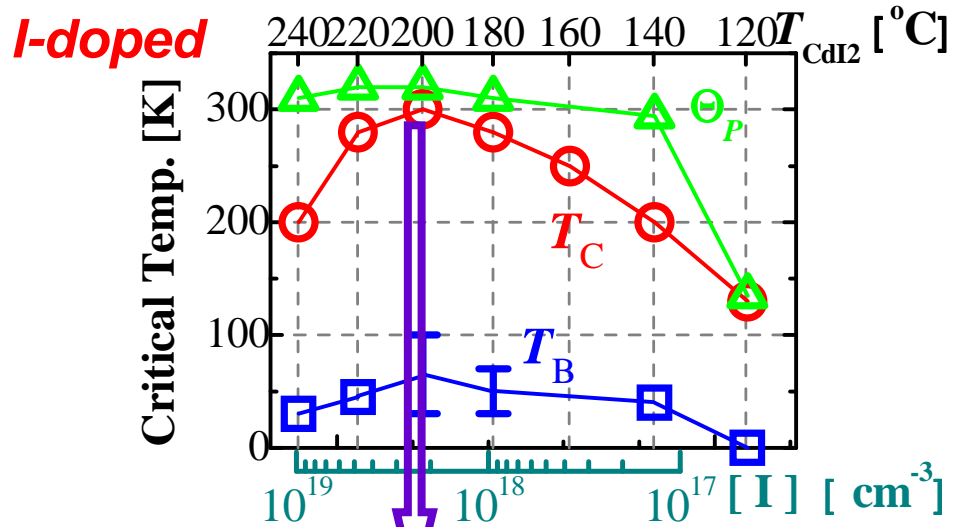
[I]  $\sim 2 \times 10^{18} \text{ cm}^{-3} \text{ Cr}^{2+}$

Te-rich growth  $\text{Cr}^{2+}/\text{Cr}^{3+}$

[N]  $\sim 3 \times 10^{20} \text{ cm}^{-3} \text{ Cr}^{3+}$

S. Kuroda ... T.D. (Tsukuba, Warsaw) Nature Mat., '07

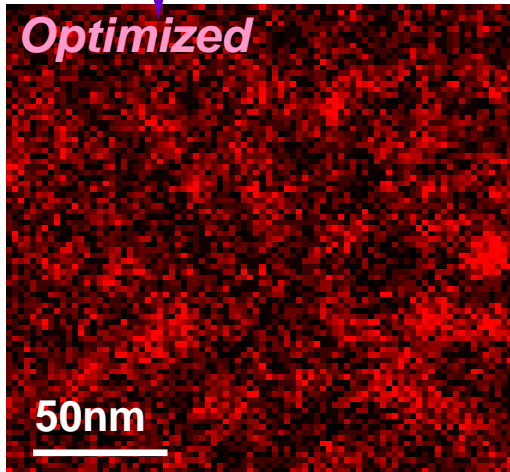
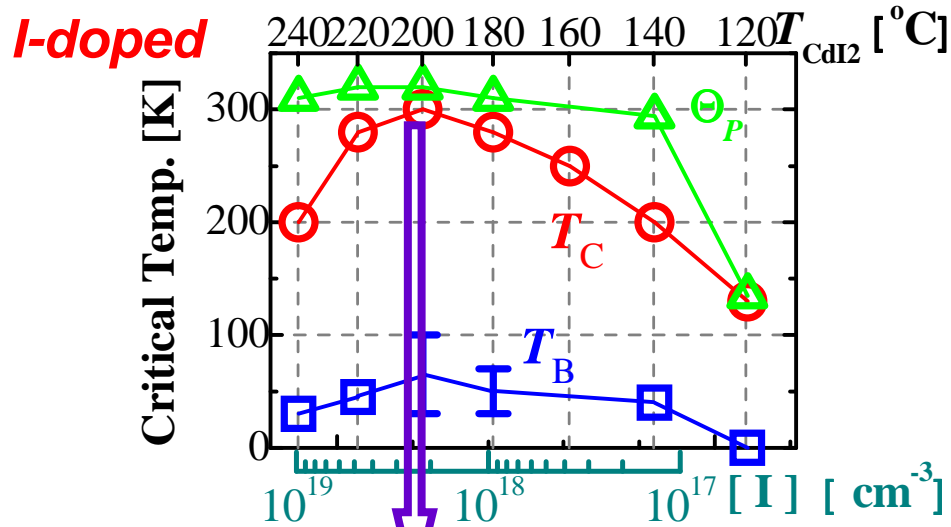
# Effect of co-doping on Cr distribution



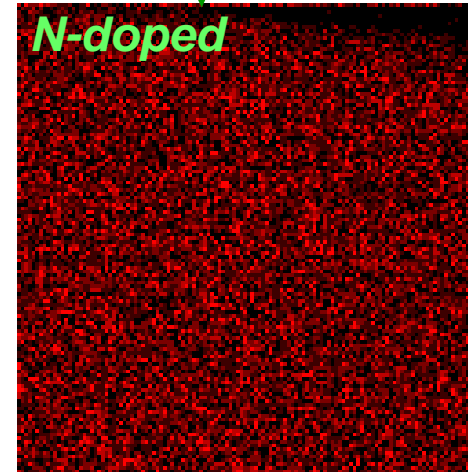
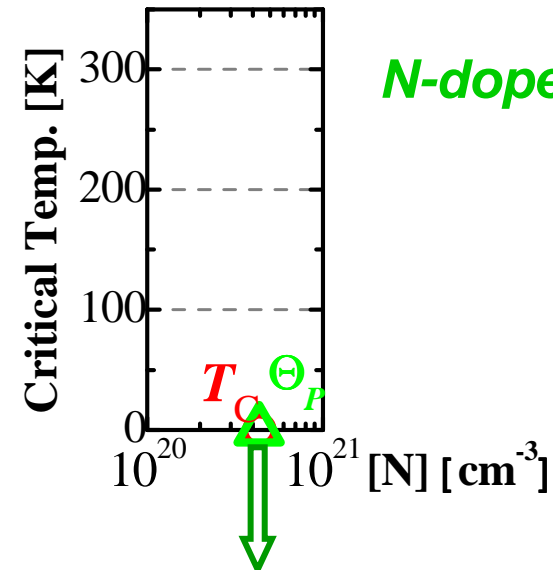
*Inhomogeneous*

S. Kuroda ... T.D. (Tsukuba, Warsaw) Nature Mat., '07

# Effect of co-doping on Cr distribution



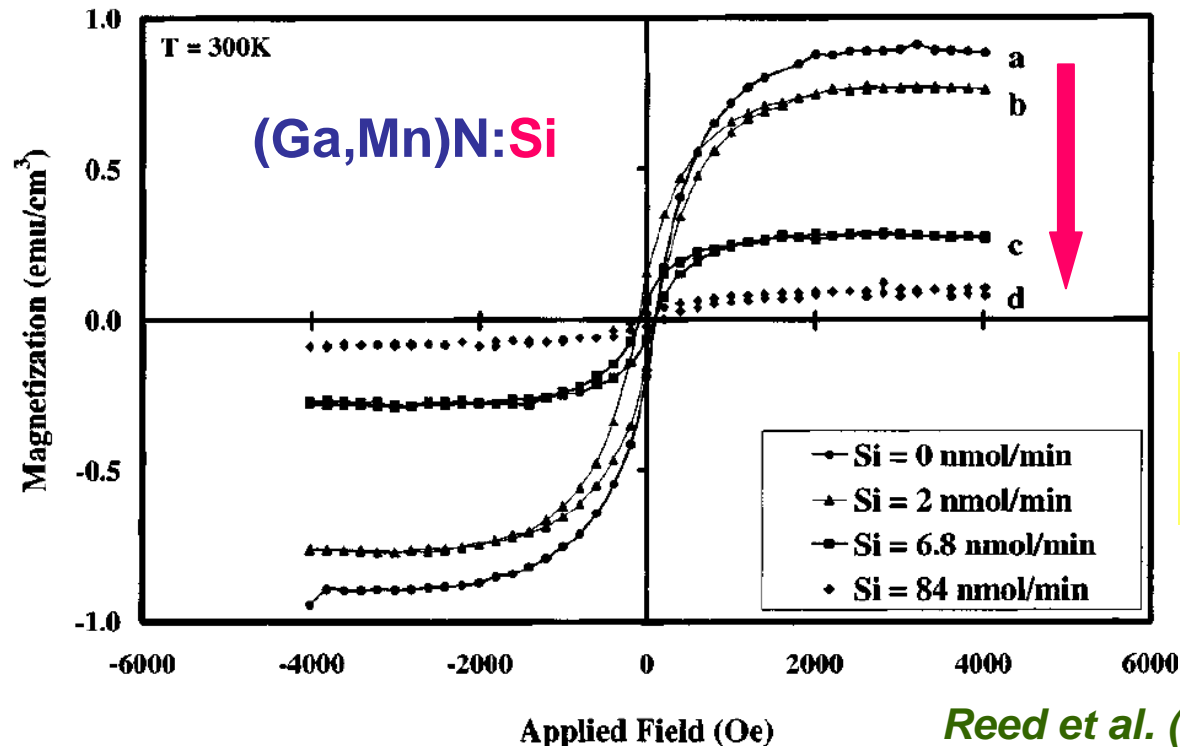
**Inhomogeneous**



**Homogeneous**

S. Kuroda ... T.D. (Tsukuba, Warsaw) Nature Mat., '07

# Ferromagnetism of (Ga,Mn)N – effect of co-doping



**(Ga,Mn)N**  
 **$x = 0.2\%$**   
 **$T_C \gg 300\text{K}$**

**(Ga,Mn)N,  $x = 0.2\%$**   
 **$T_C \rightarrow 0$  for Si doping**

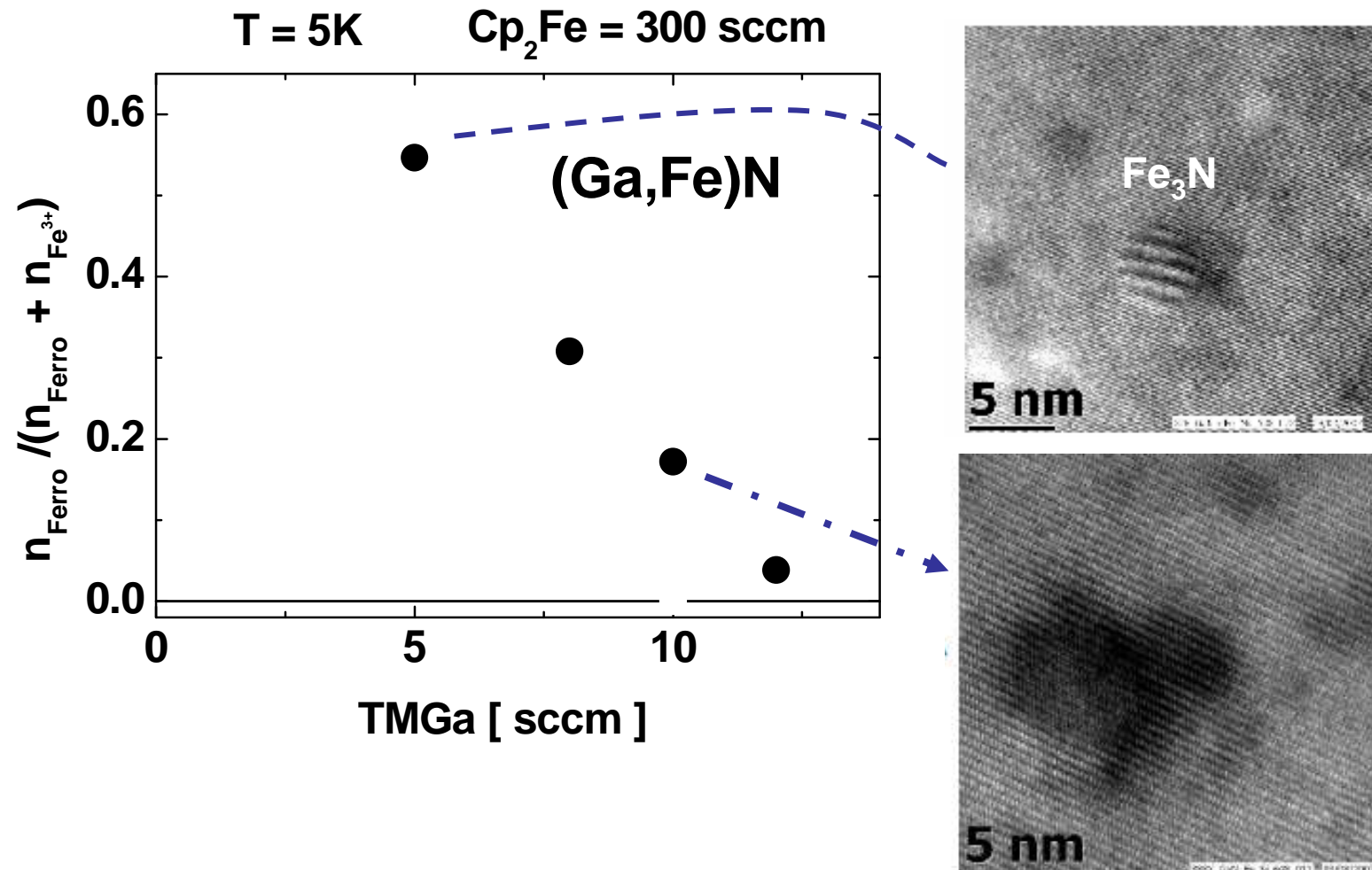
*Reed et al. (NCSU) APL '05*

*cf. Kane et al. (Georgia) JCG '06*

Effect of co-doping and stoichiometry on  $T_C$  also in (Zn,Mn)O and (Zn,Co)O

*cf. Kittilstved et al. (Washington) PRL '05*

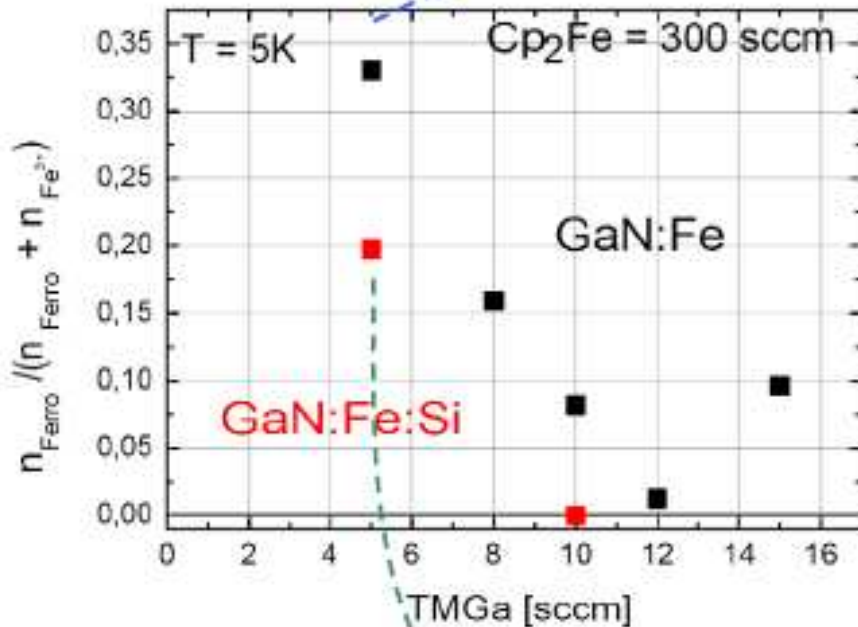
# (Ga,Fe)N -- ferromagnetic response vs. growth rate



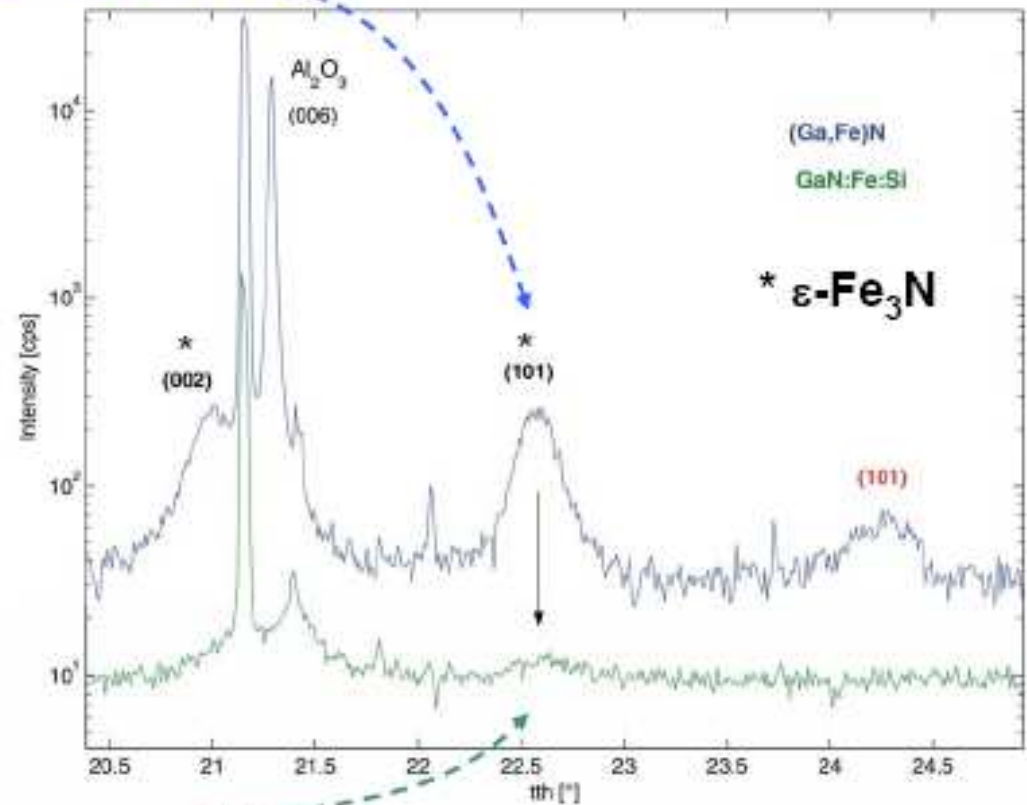
*A. Bonanni .... T.D. (Linz/Warsaw/Grenoble/Prague) PRL '08*

# Ferromagnetism of (Ga,Fe)N – effect of co-doping

ferromagnetic response  
vs. growth rate



synchrotron XDR

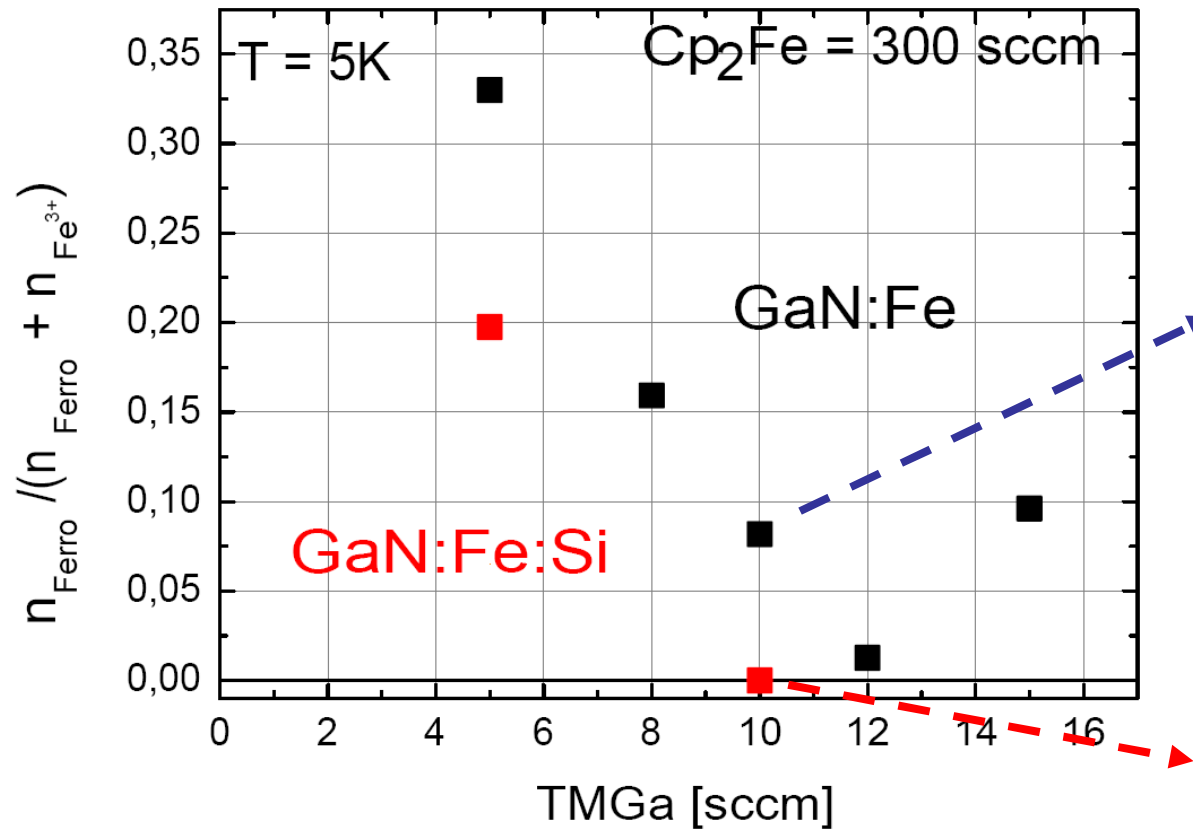


Reduction/dissolution of second phases

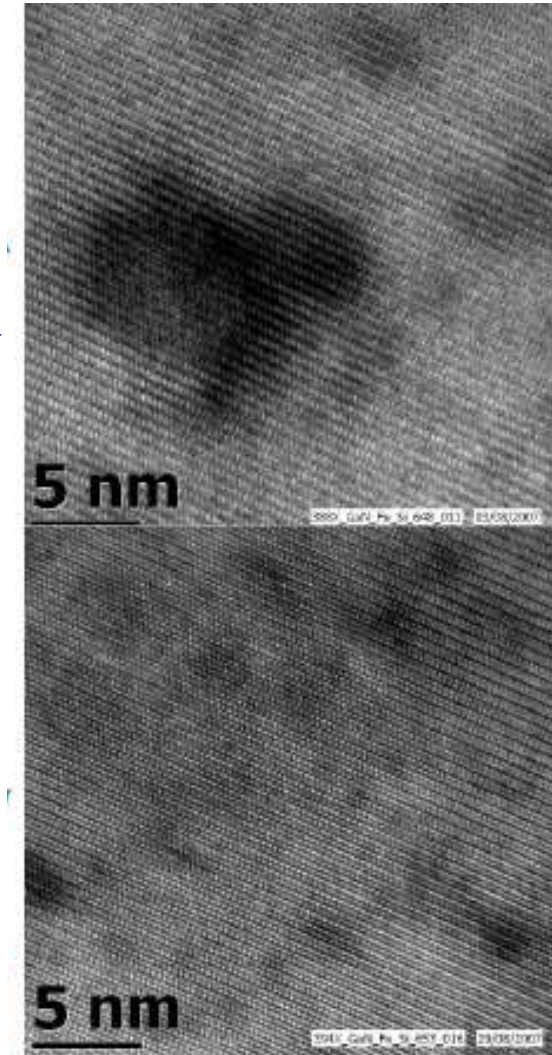
A. Bonanni .... T.D. (Linz/Warsaw/Grenoble/Prague) PRL'08



# Ferromagnetism of (Ga,Fe)N – effect of co-doping



disolution of spinodal decomposition

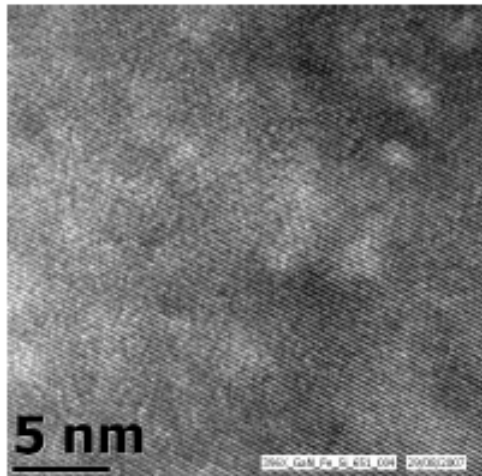


A. Bonanni .... T.D. (Linz/Warsaw/Grenoble/Prague) PRL'08

# Summary: distribution of TM in DMS

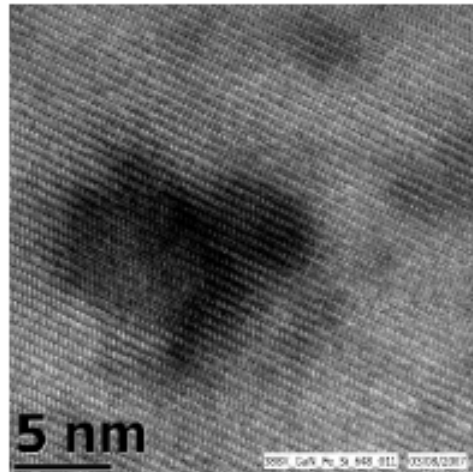
## diluted material

paramagnetic behavior  
of  $\text{Fe}^{3+}$  ions



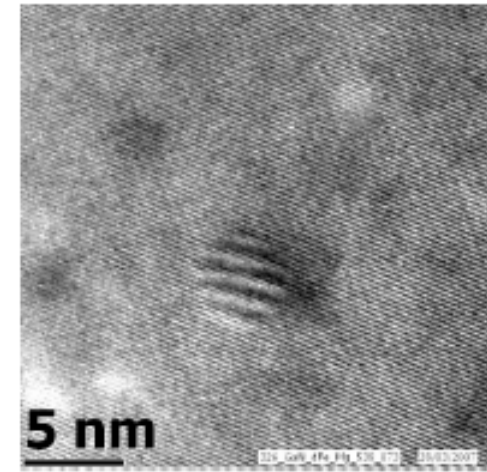
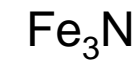
## spinodal decomposition

nano-scale chemical  
phase separation  
/ regions with high  
magnetic ions  
concentration  
/ novel magnetic  
phases stabilized



## precipitation

of known ferromagnetic,  
ferrimagnetic,  
antiferromagnetic  
compounds in  
semiconductor matrix



*A. Bonanni .... T.D. (Linz/Warsaw/Grenoble/Prague) PRL '08*

# SUMMARY cont.

1. New coherent magnetic compounds formed
2. High  $T_C$ ,  $T_N$ ; high blocking temperature
  - spontaneous magnetization at high temperatures
3. Nanoassembling (shape, size) can be controlled *in situ* by growth conditions/co-doping

# SUMMARY cont.

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4th generation epitaxy

# SUMMARY cont.

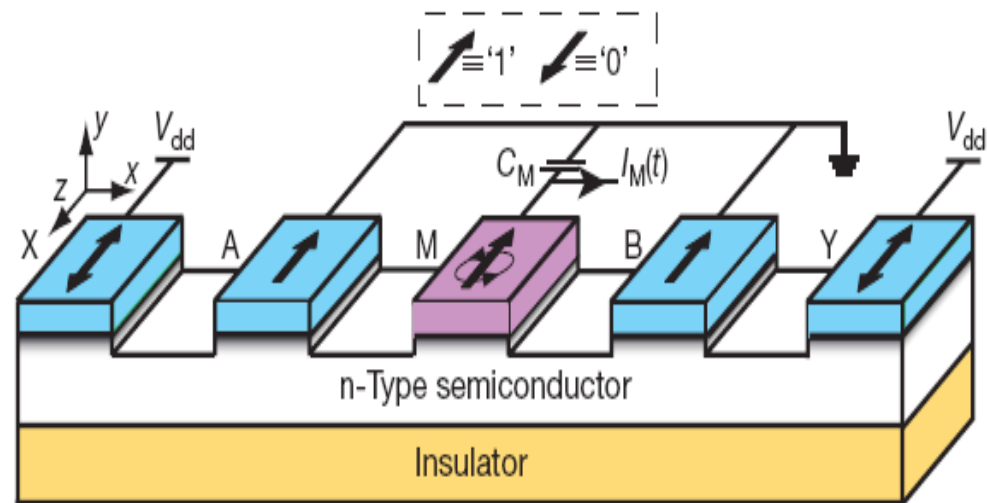
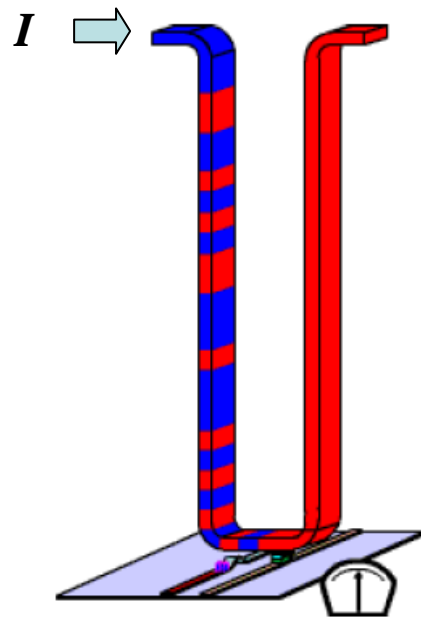
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2. High  $T_C$ ,  $T_N$ ; high blocking temperature
  - spontaneous magnetization at high temperatures
3. Nanoassembling (shape, size) can be controlled *in situ* by growth conditions/co-doping
4. Functionalities:
  - nanometalization → nanoelectronics, optoelectronics, plasmonics
  - large magnetotransport effects → field sensors
  - large magneto-optical effects → optical isolators, tunable photonic crystals
  - spintronic structure → high density MRAMs/race track memories/logic
  - large spin entropy → thermoelectricity
  - ....

# OUTLOOK

*Self-organised nanomagnets in semiconductors -- media for:*

race track memory

logic gates



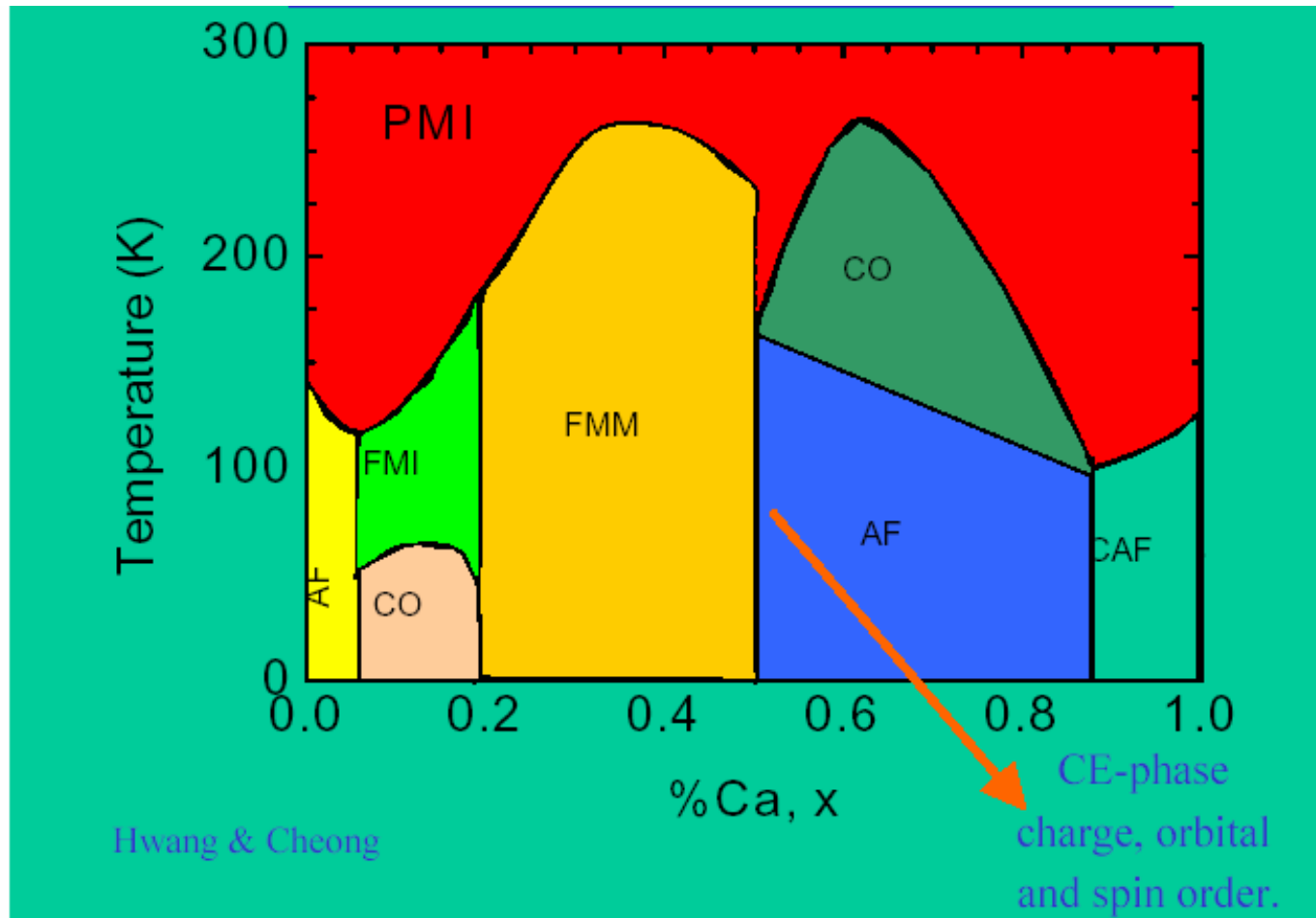
*Dery et al. (UCSD) Nature '07*

*Parkin et al. (IBM) SPINTECH '05, Science '06*

magneto-optical devices

metallization

# Phase diagram of $\text{La}_{1-x}\text{Ca}_x\text{MnO}_3$



**DMS and DMO: interactions determine spatial distribution of both carriers *and* localised spins**

