Magnetic properties of Ferromagnetic Semiconductor (Ga,Mn)As

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In collaboration with:

- T. Dietl, et al., Warsaw
- B. Gallagher, et al., Nottingham
- L.W. Molenkamp, et al., Wuerzburg
- H. Ohno, et al., Sendai

Support by: Japanese **ERATO,** EU **FENIKS**, Polish **MNiI**

Outlook

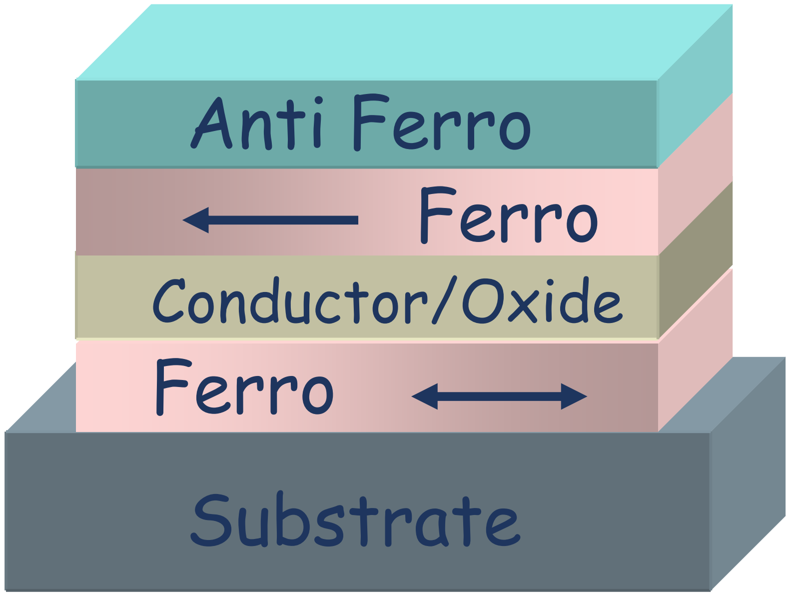
- Introduction
  - motivation/history
- T_C and M_S
- Uniaxial magnetic anisotropy due to confinement and/or biaxial (epitaxial) strain
  - reorientation transition
- Biaxial (cubic, 4-fold) in-plane anisotropy
- Uniaxial in-plane anisotropy
  - reorientation transition
  - single domain behaviour

Hole driven ferro-DMS, mostly (Ga,Mn)As

Making spins to:

- store and reveal information in a faster way
- transmit information **(supplementing charge and light)**
- process information **(supplementing charge)**

Spin valve (or MTJ)



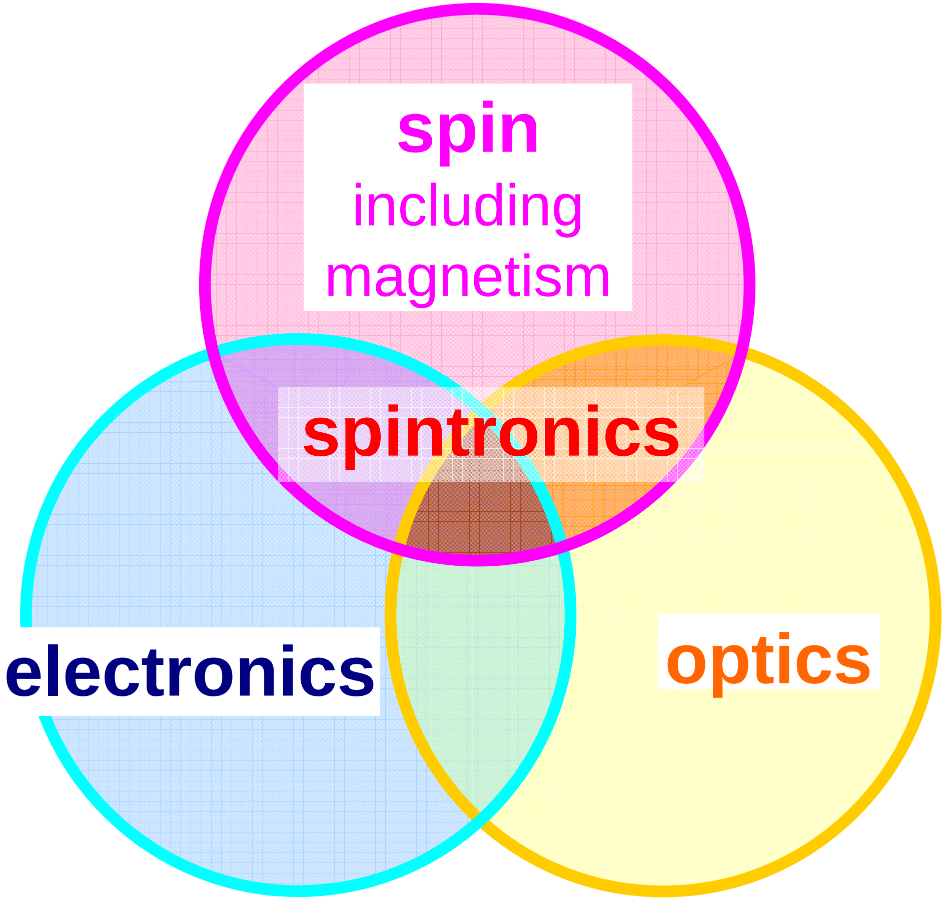
Main applications:

- magnetic field sensors
- read heads
- galvanic isolators
- Magnetoresistive RAMs

Why semiconductor spintronics?

Semiconductor Spin-electronics (Spintronics)

Spin-related phenomena in semiconductors →
an additional degree of freedom (spin + charge → **spintronics**)



Ferromagnetic semiconductors

May offer a possibility to replace of

'All metal' Spin-Based Electronic Devices

- they posses both spins and mechanism that effectively couples spins with carriers.
- technological compliance with semiconductor industry.

Towards ferromagnetic semiconductors

- **magnetic semiconductors**

  magnetic semiconductors and insulators: short-range antiferromagnetic superexchange

  EuTe, ..., NiO, ...

  short-range ferromagnetic super- or double exchange

  EuS, $ZnCr_2Se_4$, $La_{1-x}Sr_xMnO_3$, ...

  EuS/KCl,...

- **diluted magnetic semiconductors**

  Standard semiconductor + magnetic ion

  II-VI: $Cd_{1-x}Mn_xTe$, ..., $Hg_{1-x}Mn_xSe$, ...

  IV-VI: $Sn_{1-x}Mn_xTe$, ..., $Pb_{1-x}Eu_xS$

  III-V: $In_{1-x}Mn_xSb$, ..., $Ga_{1-x}Er_xN$

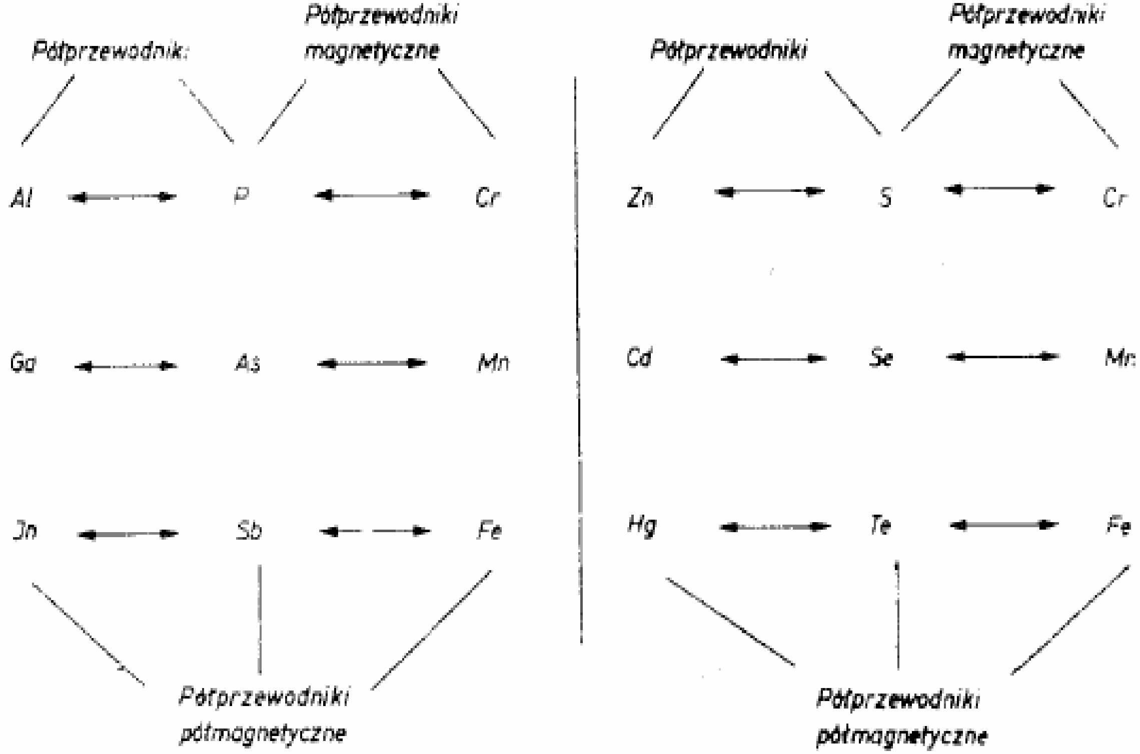
  IV: $Ge_{1-x}Mn_x$, ..., $Si_{1-x}Ce_x$

History of DMS

POSTĘPY FIZYKI — TOM 28 — ZESZYT 5 — 1977

Robert R. Gałązka

Instytut Fizyki PAN
Warszawa

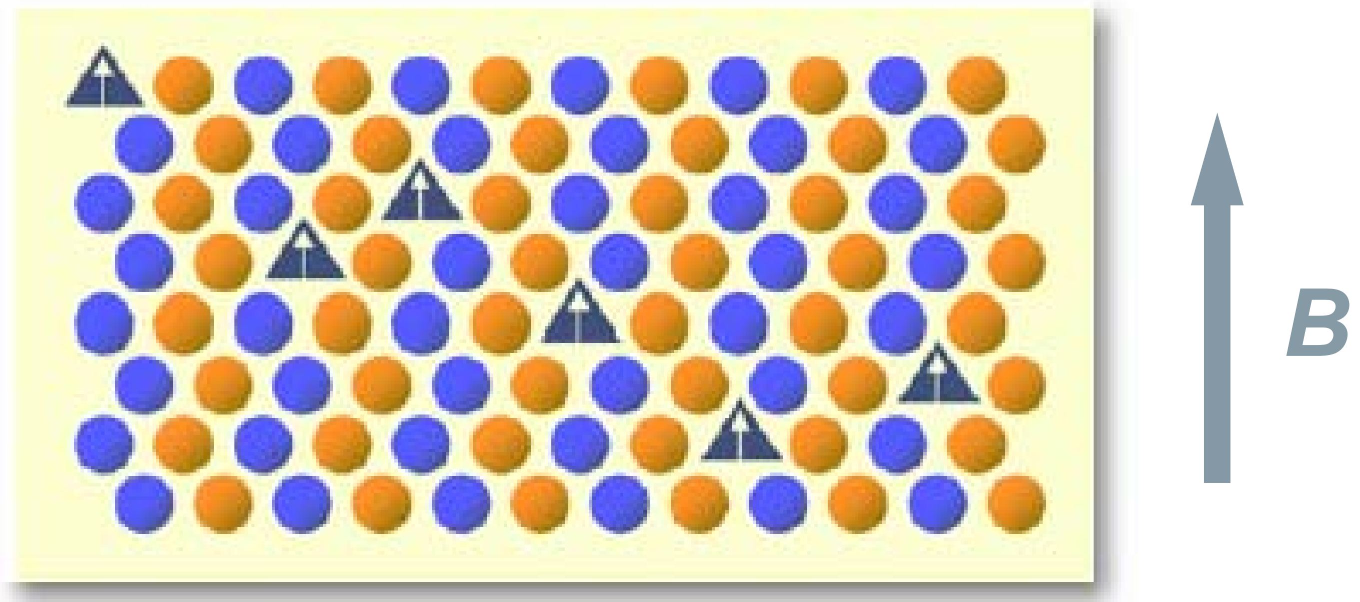


Rys. 2. Przykład jak można tworzyć półprzewodniki półmagnetyczne. Oczywiście można również tworzyć skośne połączenia np GaMnSb, ZnFeSe...

Abstract: The paper considers a new group of solid states — alloys between semiconducting and magnetic compounds. The materials conserve main properties characteristic for semiconductors (doping in wide range of concentration on n and p type, well defined band structure $E(k)$) but contain strong localized spins introduced by transition elements. New physical phenomena are observed mainly at low temperatures and in the presence of magnetic field. Experimental results are presented for HgMnTe and CdMnTe type of mixed crystals.

Most of DMS: random antiferromagnet

short range antiferromagnetic superexchange



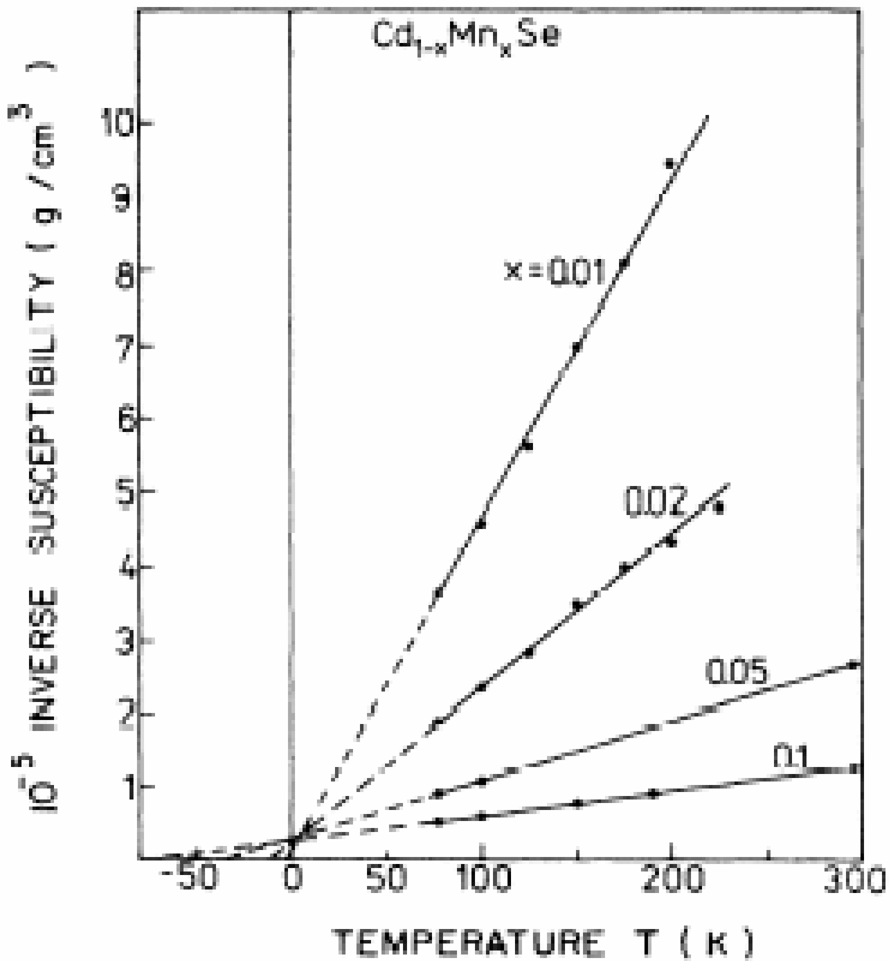
Evidences for antiferromagnetic interactions: magnetic susceptibility

Curie-Weiss law

$$\chi = C/(T - \Theta)$$

$$C = g\mu_B S(S+1)xN_o/3k_B$$

$\Theta < 0$ antiferro



A. Lewicki et al.

Magnetisation of localized spins

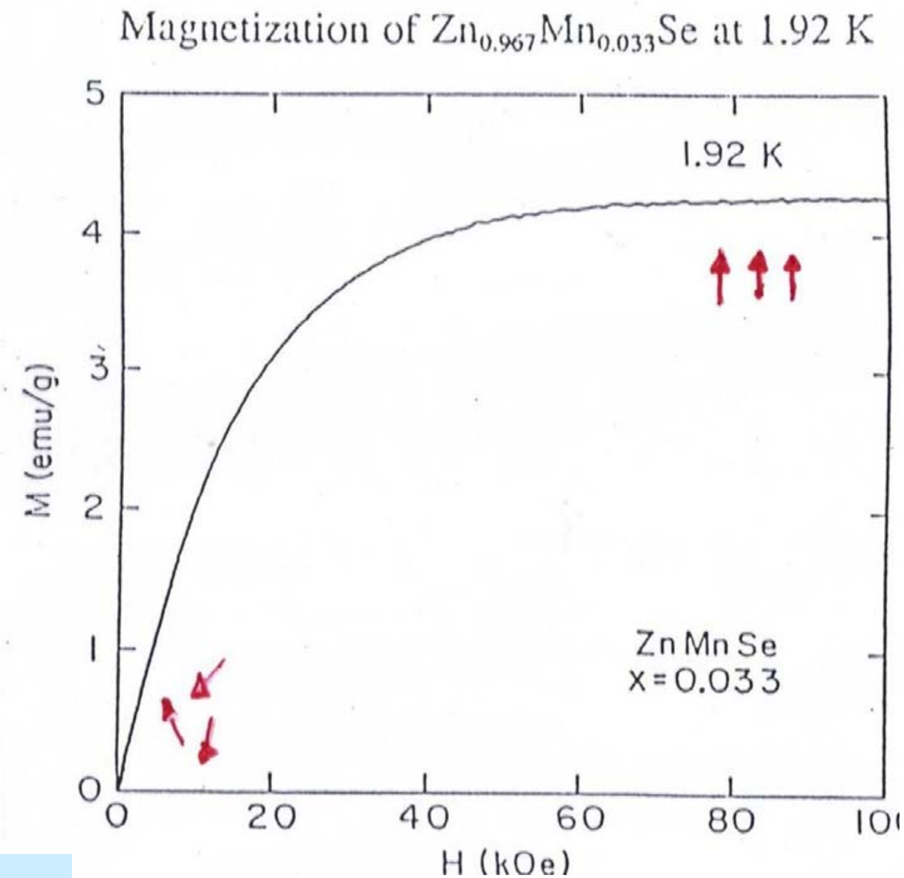
$M(T,H) = g\mu_B S x_{eff} N_o B_S[g\mu_B H/k_B(T + T_{AF})]$

antiferromagnetic interactions

$x_{eff} < x$

$T_{AF} > 0$

Modified Brillouin function

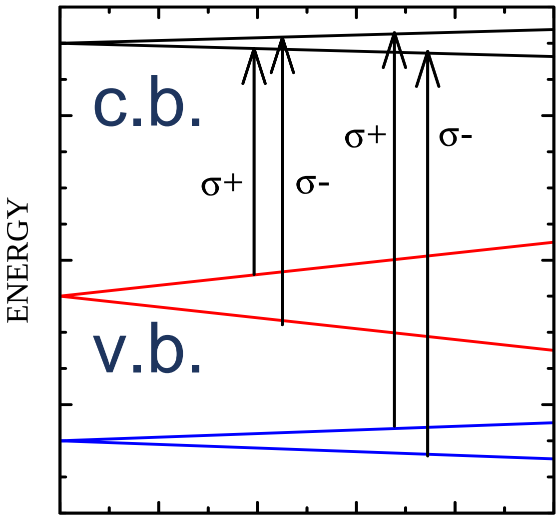


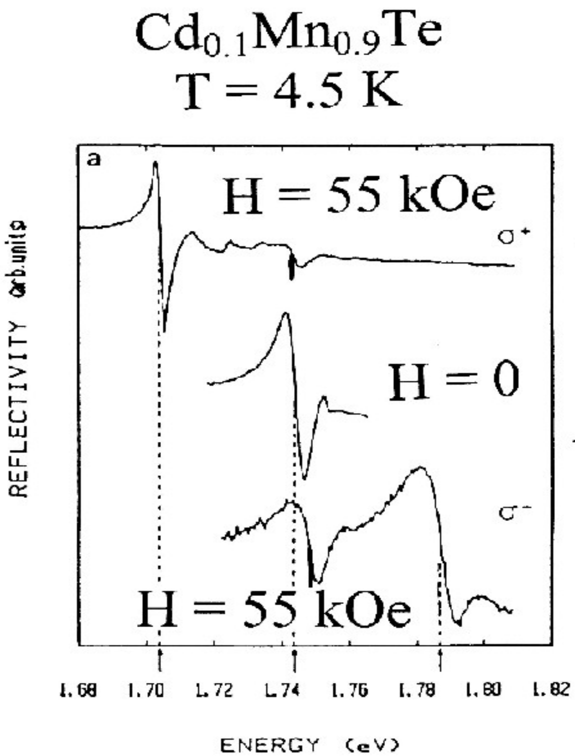
no spontaneous magnetisation …

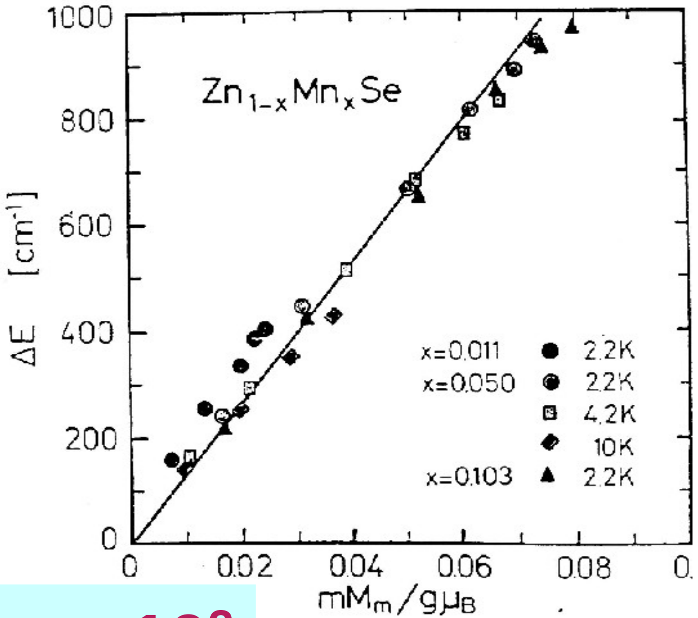
Y. Shapira et al.

Determination of sp-d exchange integrals: - giant splitting of exciton states

$\Delta E \sim M \sim B_S(H)$







$g_{eff} > 10^2$

J. Gaj et al., R. Planel,..
A. Twardowski et al.
G. Bastard, …

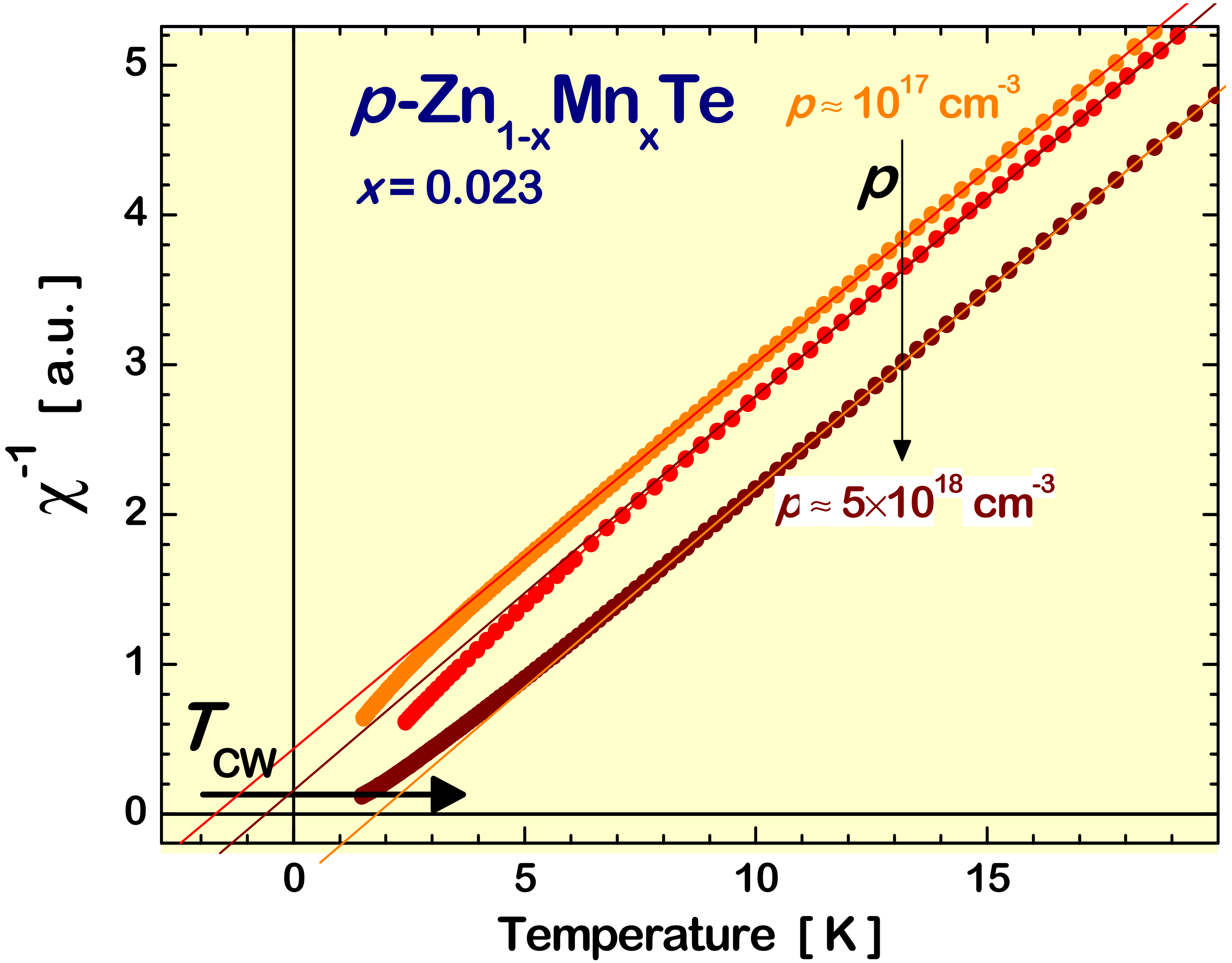
-- s-d: $I_{sd} \equiv \alpha N_o \approx 0.2$ eV

no s-d hybridization => potential s-d exchange

-- p-d: $I_{pd} \equiv \beta N_o \approx -1.0$ eV

large *p-d* hybridization and large intra-site Hubbard *U* => kinetic p-d exchange

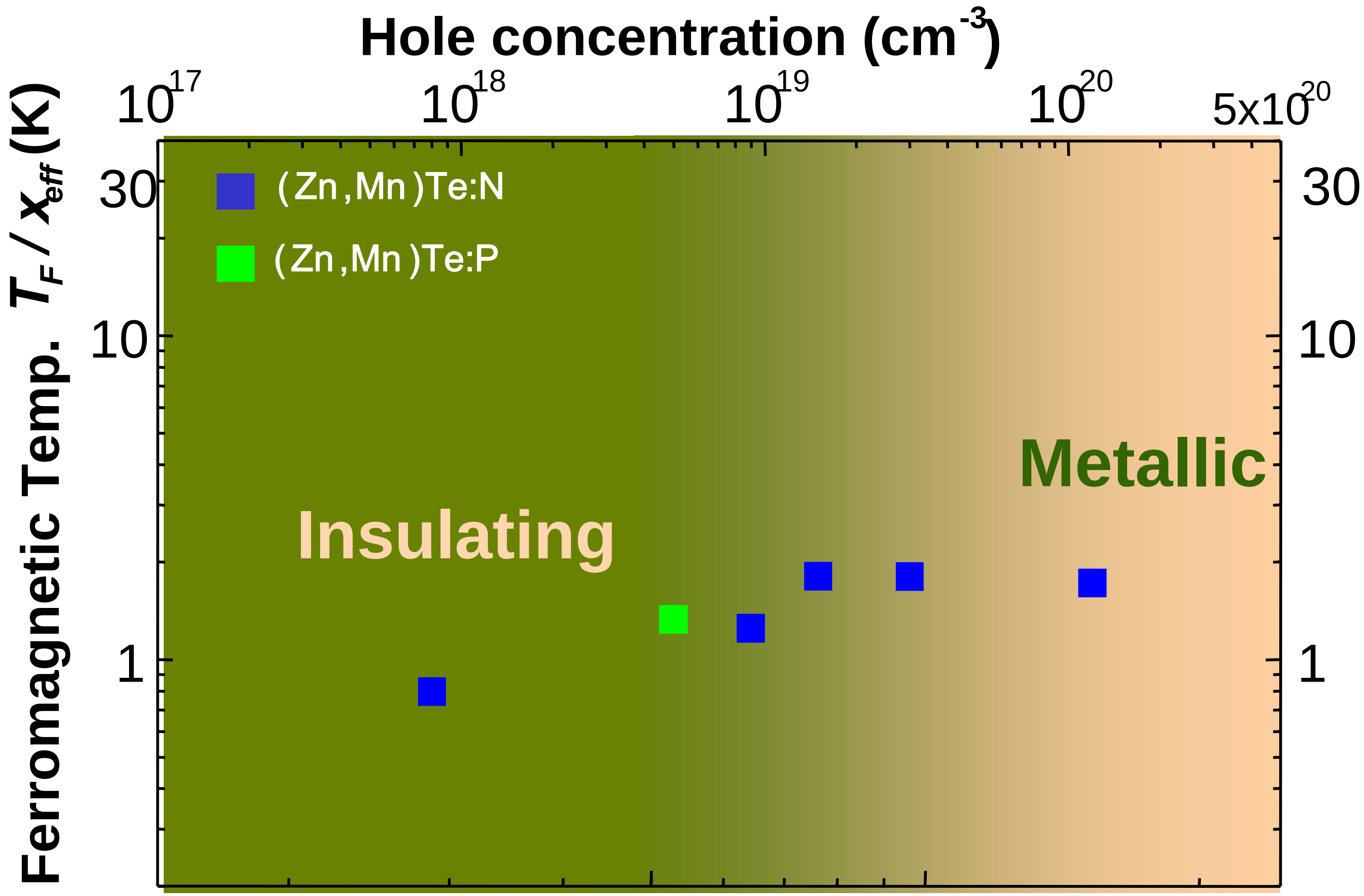
Effect of acceptor doping on magnetic susceptibility in $Zn_{1-x}Mn_xTe$:P



χ^{-1} vs. T

Sawicki et al. (Warsaw) pss'02
Kępa et al. (Warsaw, Oregon) PRL'03

Ferromagnetic temperature in p-(Zn,Mn)Te



- ferromagnetism disappears in the absence of holes
- ferromagnetism on both sides of metal-insulator transition

Ferrand et al. (Grenoble, Warsaw) PRB'01
Sawicki et al. (Warsaw) pss'02

Ferromagnetism in DMS – the origin

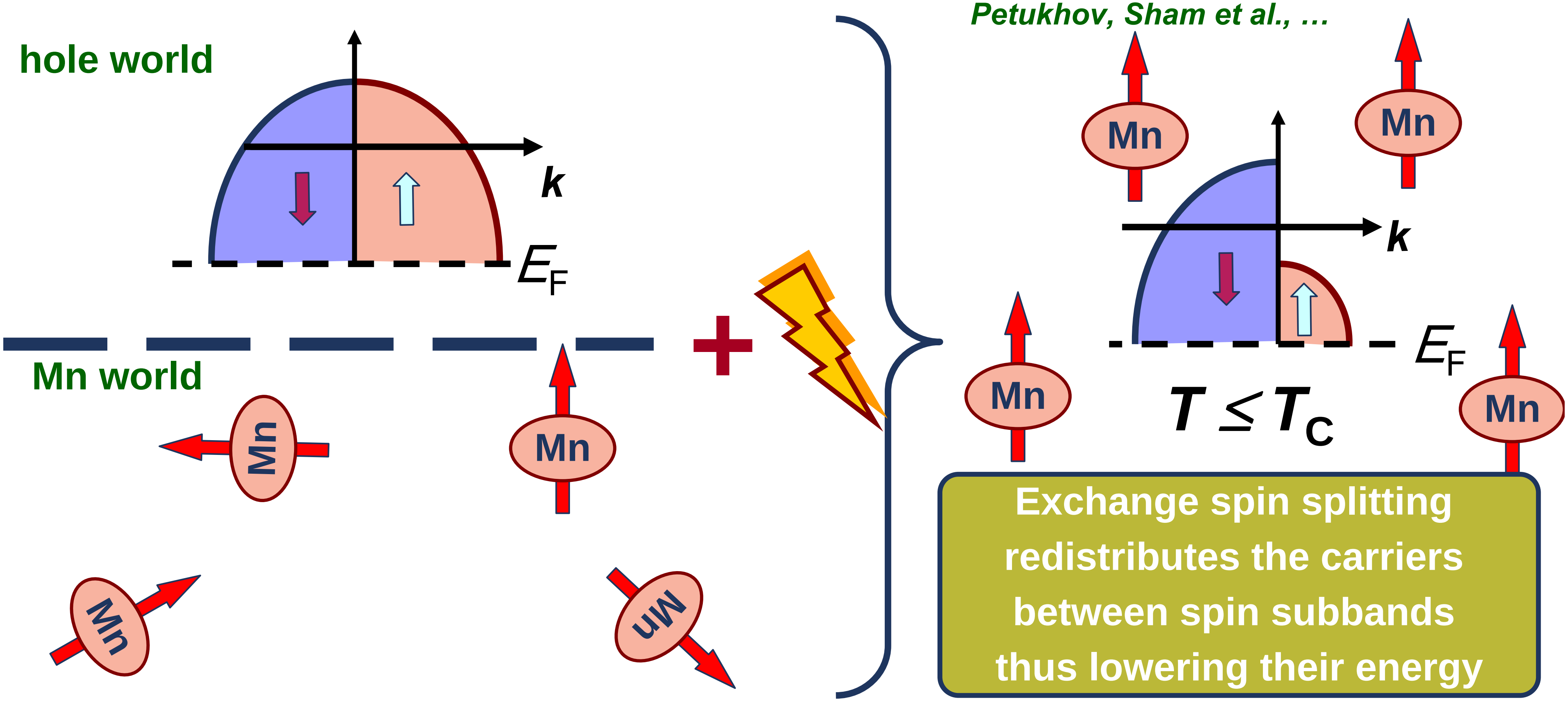
-- carriers localized by impurities (BMP): inoperative

Bhatt et al., Dugaev et al., Inoue et al., Das Sarma et al., Dagotto et al.,

...

-- delocalized carriers (Zener/RKKY model)

Ryabchenko, et al., Dietl et al., MacDonald et al., Boselli et al., Petukhov, Sham et al., …



Exchange spin splitting redistributes the carriers between spin subbands thus lowering their energy

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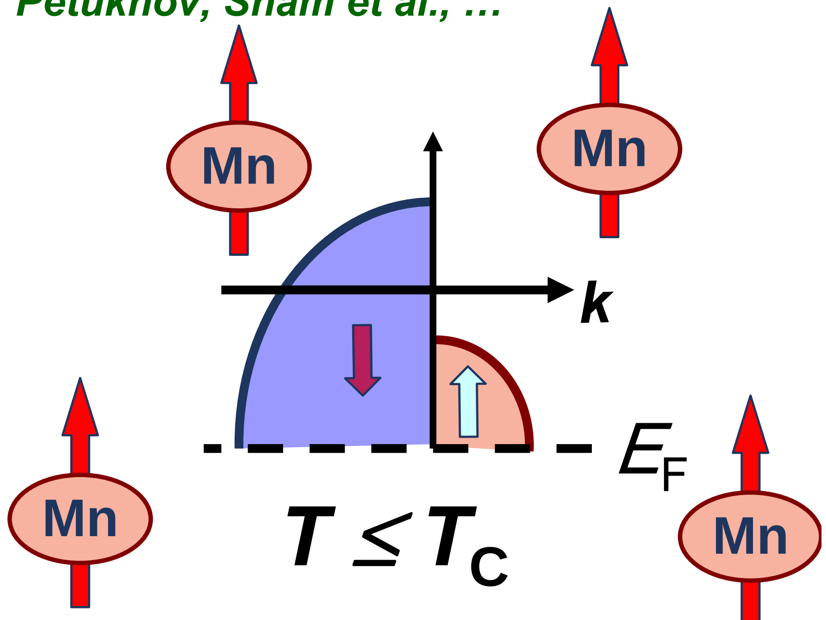
$$T_C = x_{eff} N_0 S(S+1) J^2 A_F \rho(\varepsilon_F)/12k_F$$

holes!!! = valence band

-- s-d: $I_{sd} \equiv \alpha N_o \approx 0.2$ eV
no s-d hybridization

-- p-d: $I_{pd} \equiv \beta N_o \approx -1.0$ eV
large *p-d* hybridization

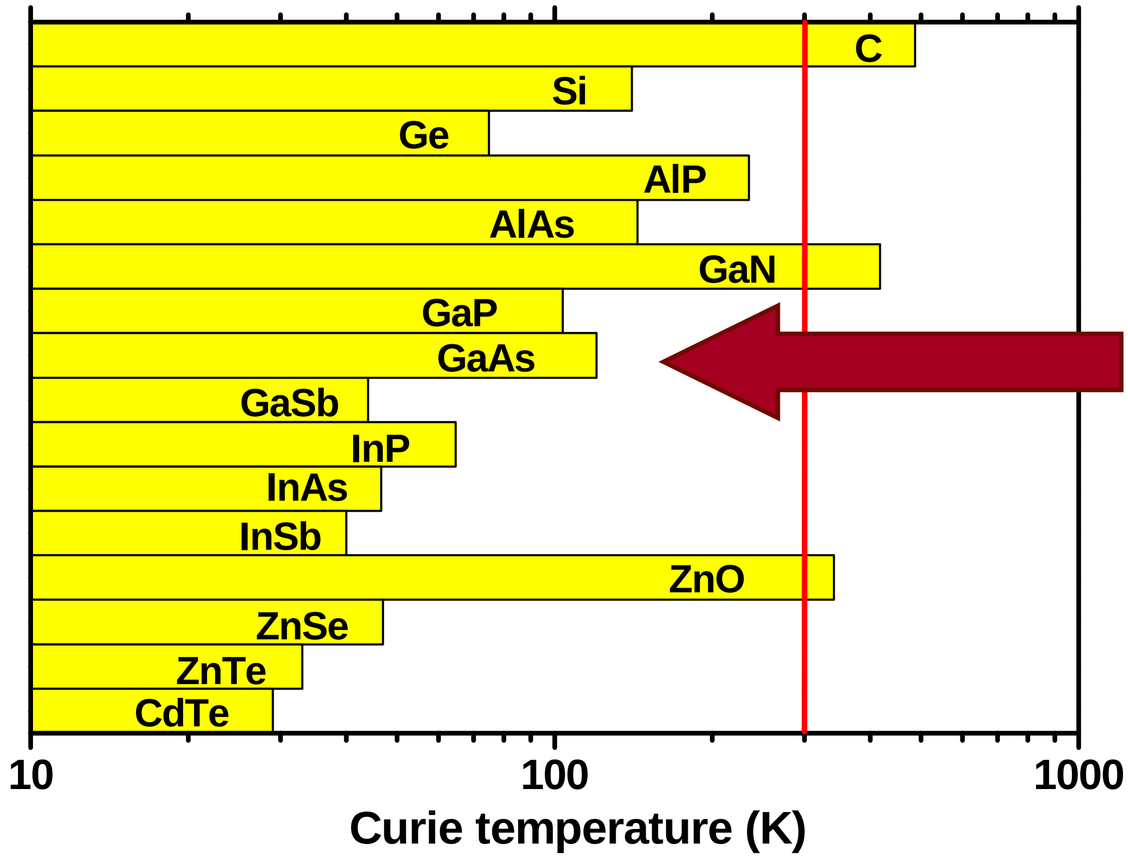


Exchange spin splitting redistributes the carriers between spin subbands thus lowering their energy

Why DMS, why (Ga,Mn)As?

Carrier mediated ferromagnetism in semiconductors:

$x = 0.05$, $p = 3.5\times10^{20}$ cm^{-3}



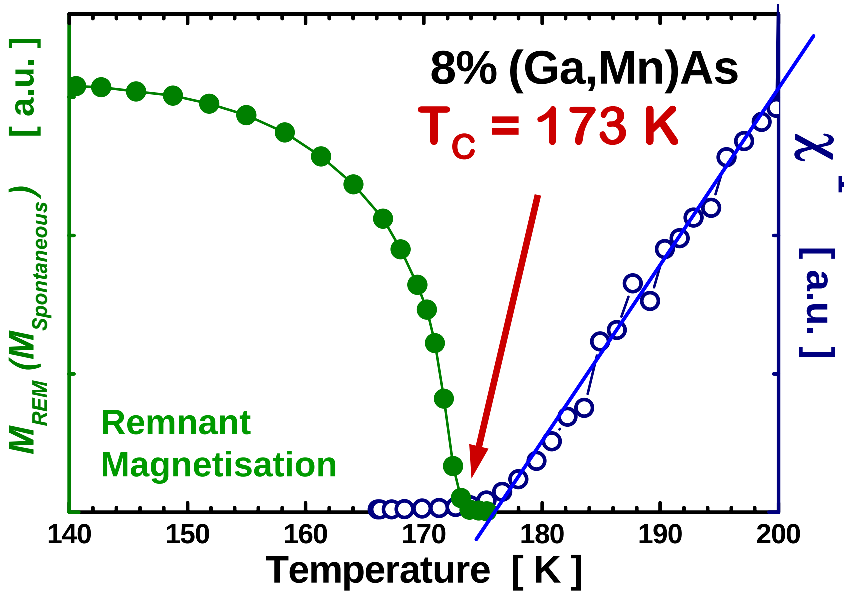
T. Dietl, et al., Science 2000

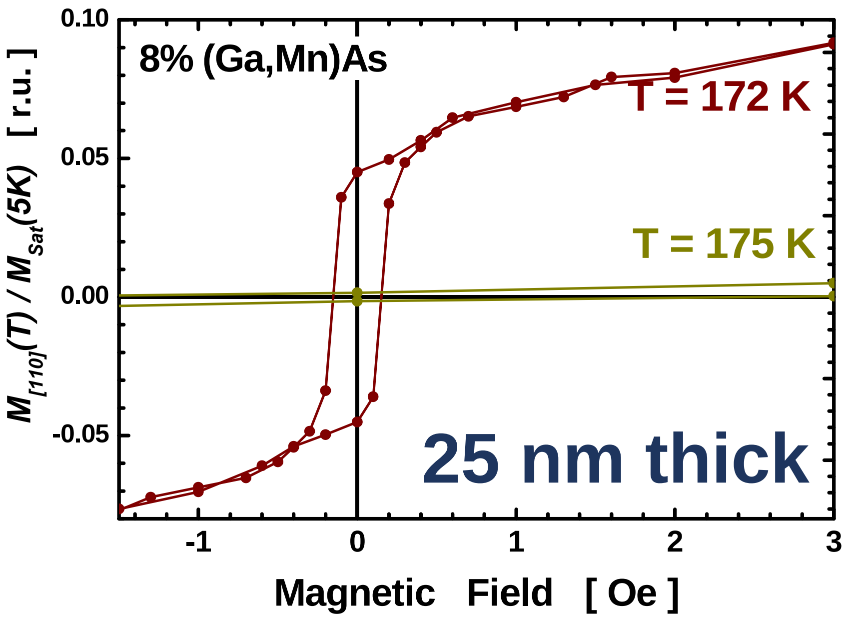
More than 20 compounds showed ferro- coupling so far

Operational criteria:

- Scaling of T_C and M with x and p
- Interplay between semiconducting and ferromagnetic properties

(Ga,Mn)As: single phase ferro-DMS

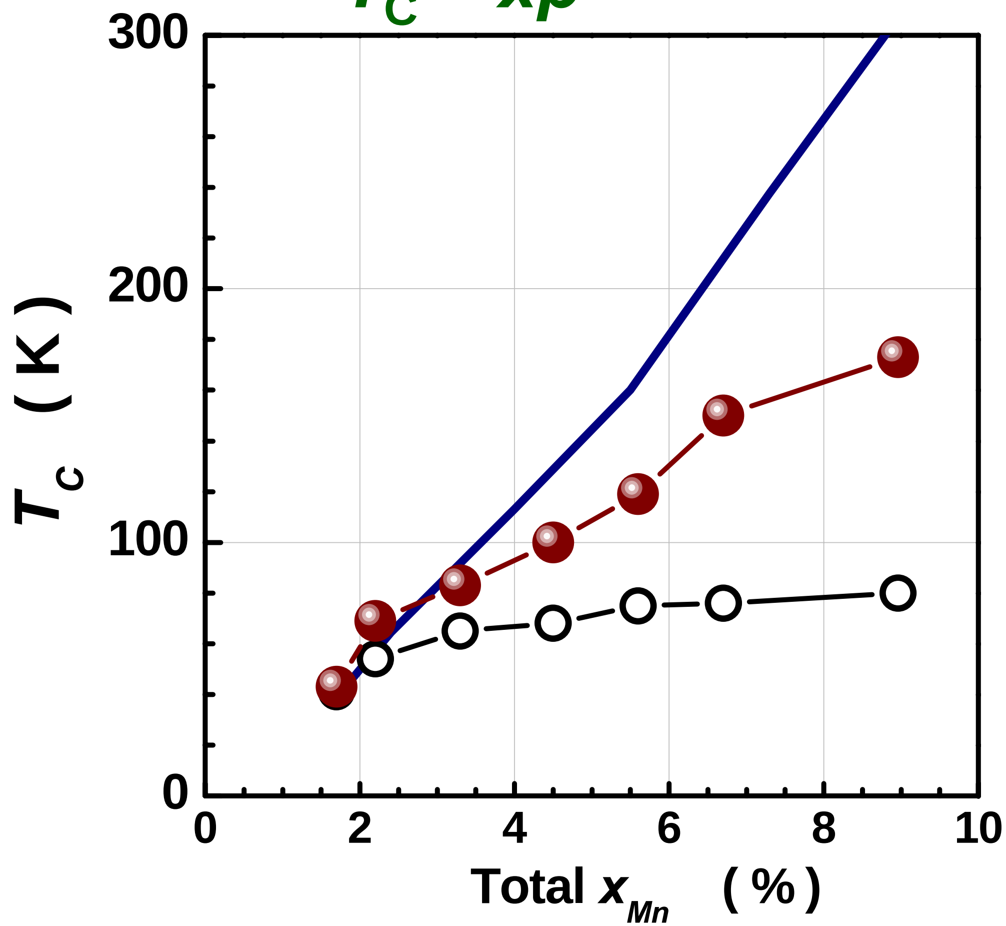




K-Y. Wang, et al., JAP '04 & ICPS'27

$T_F = x_{eff} N_0 S(S+1) J^2 A_F \rho(\varepsilon_F)/12k_F$

$T_C \sim xp^{1/3}$

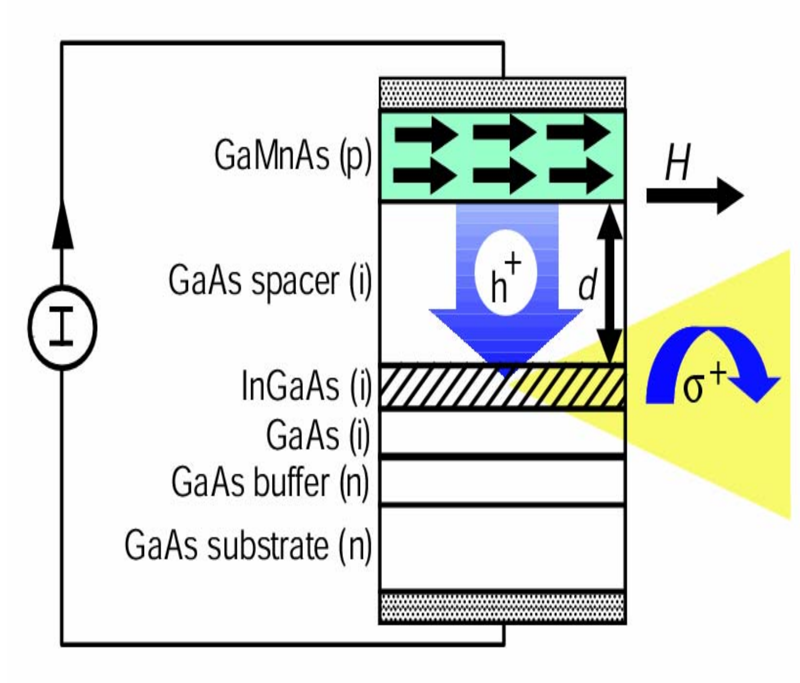


T. Dietl, H. Ohno, F. Matsukura, PRB '01

T_C = 173 K = -100° C

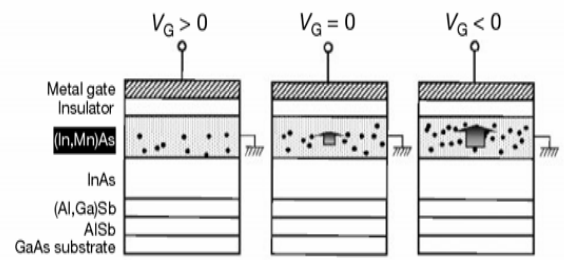
Operational criteria for carrier-controlled ferromagnetic semiconductors

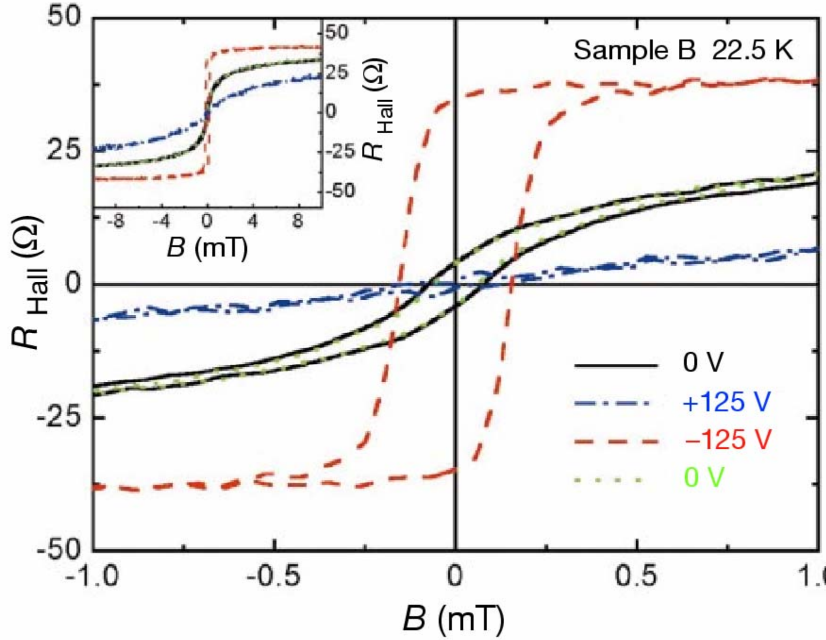
Spin-LED



Y. Ohno et al., Nature'99

Ferro-FET





H. Ohno et al., Nature'00

Also:

- Current induced domain wall switching **J_C~10^5 A/cm^2**

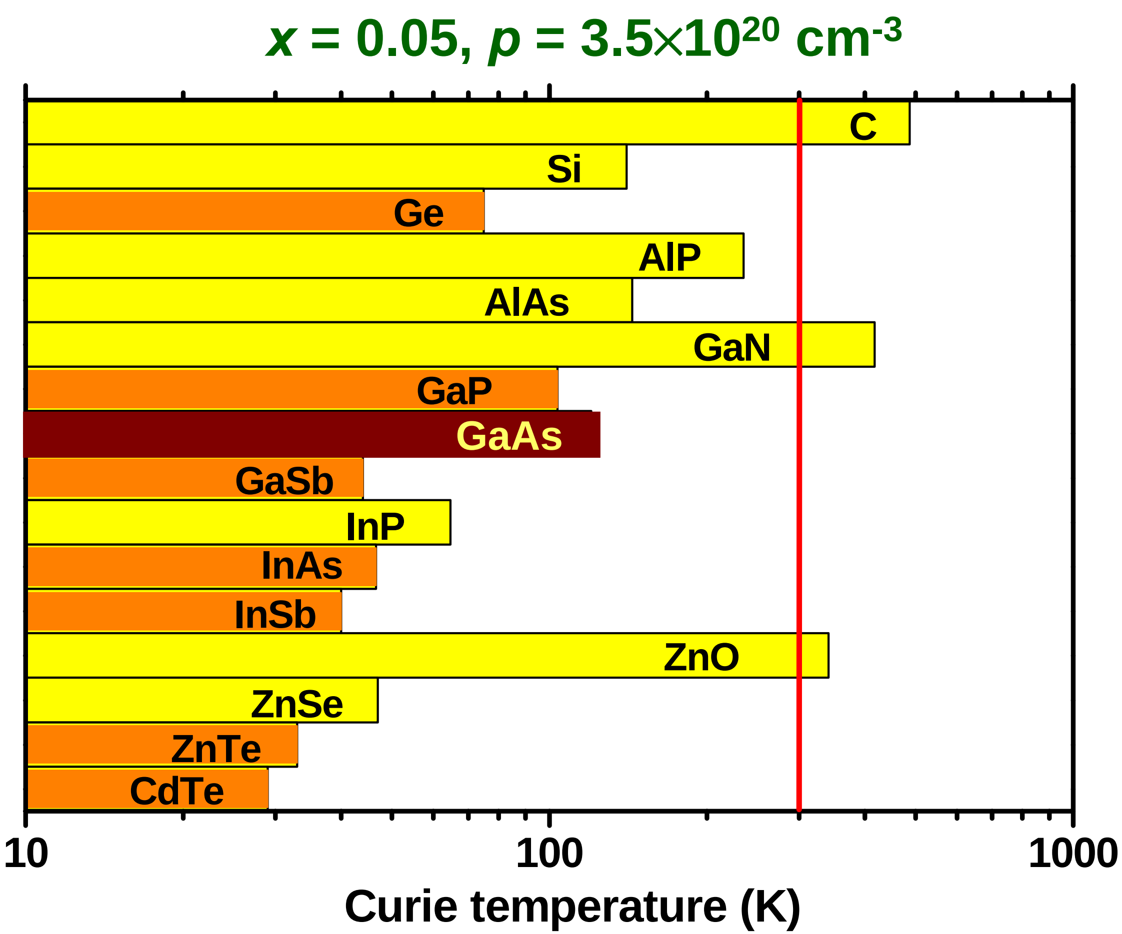
  M. Yamanouchi, et al., Nature'04

- Electrically assisted magnetisation reversal

  D. Chiba, et al., Science'03

Why DMS, why (Ga,Mn)As?

Carrier mediated ferromagnetism in semiconductors:



T. Dietl, et al., Science 2000

More than 20 compounds showed ferro- coupling so far

Operational criteria:

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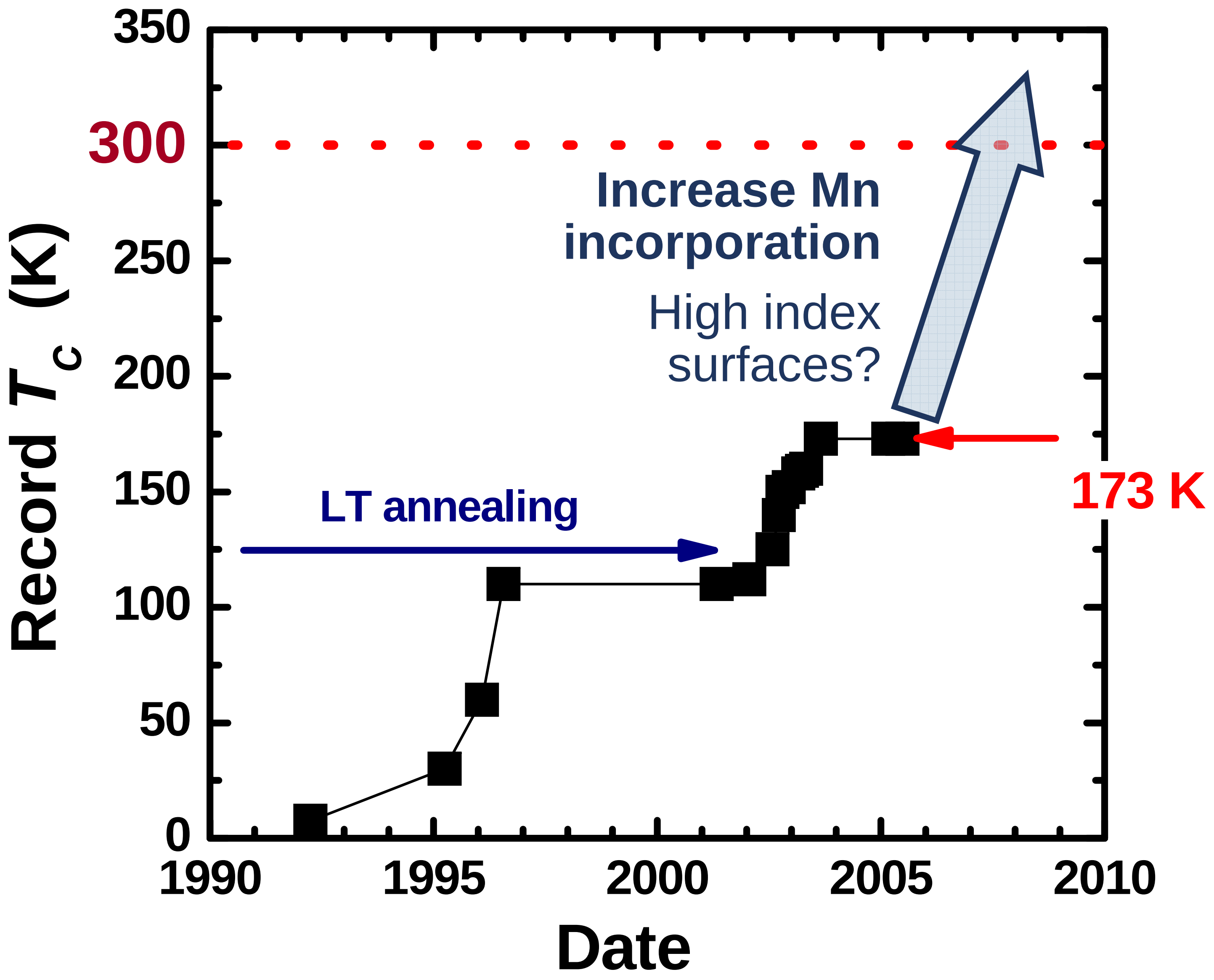
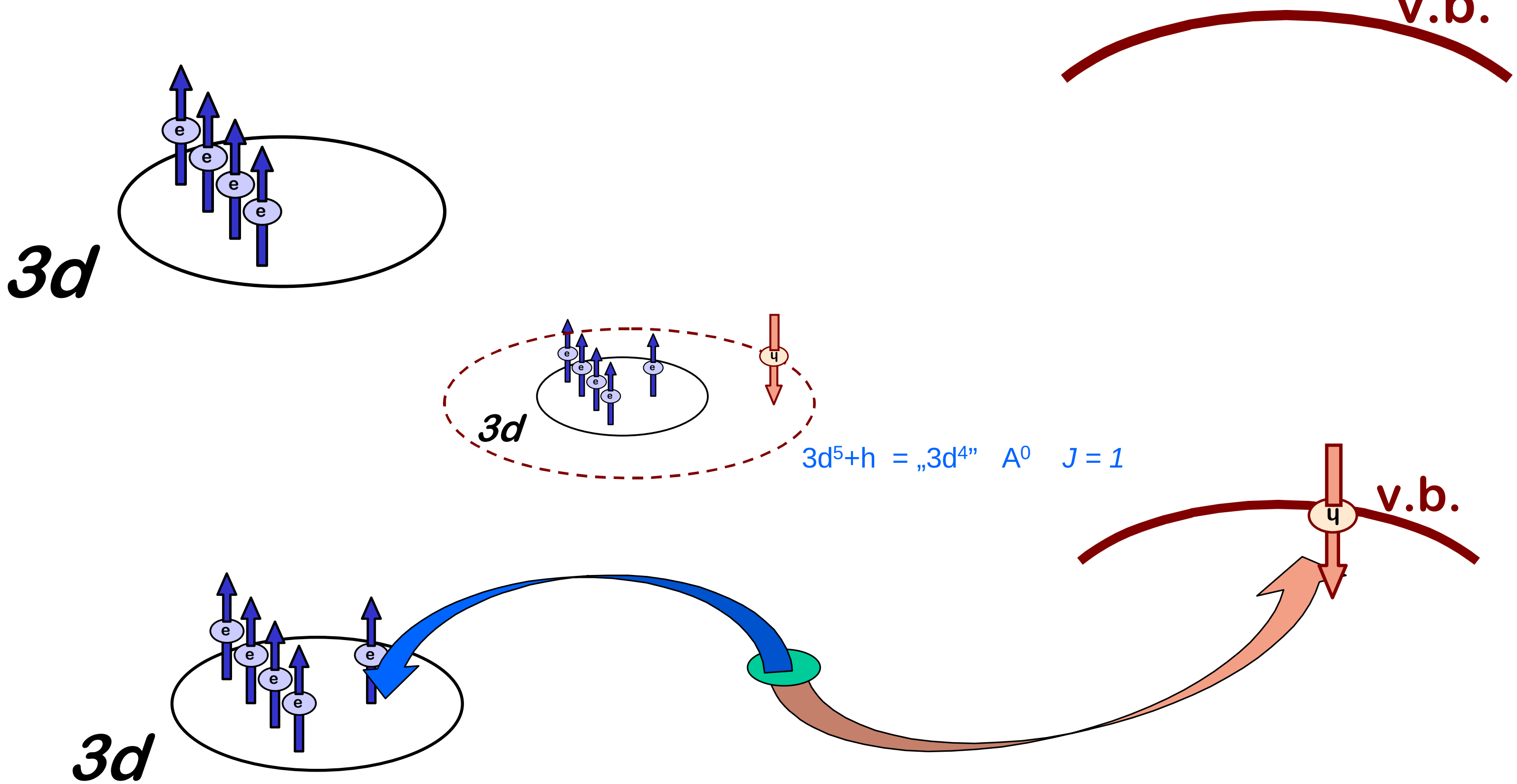
- Interplay between semiconducting and ferromagnetic properties

T_C in (Ga,Mn)As: prospects



Mn in GaAs

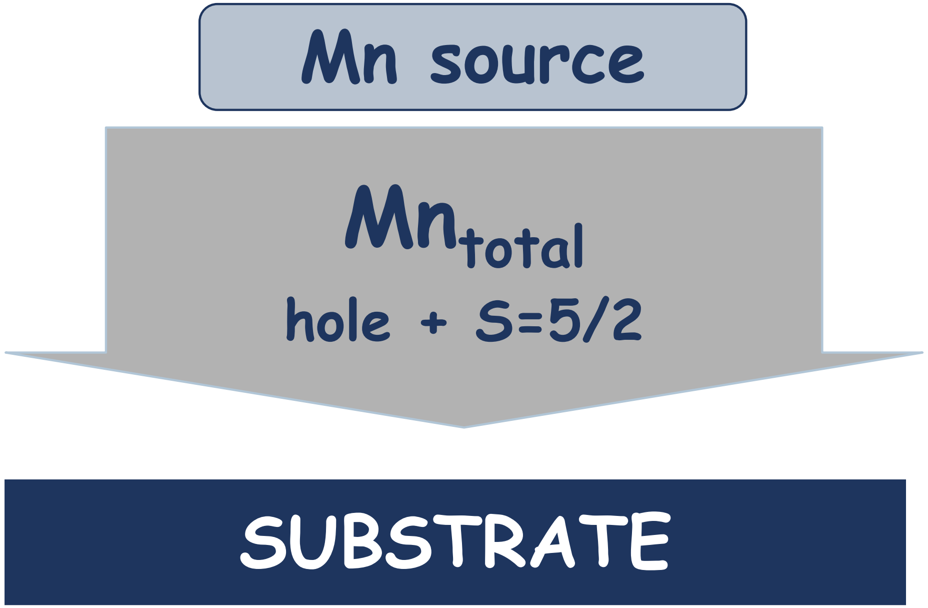
Mn(...$3d^5 4s^2$) + GaAs = $3d^4$ (A^0) + e+e+e

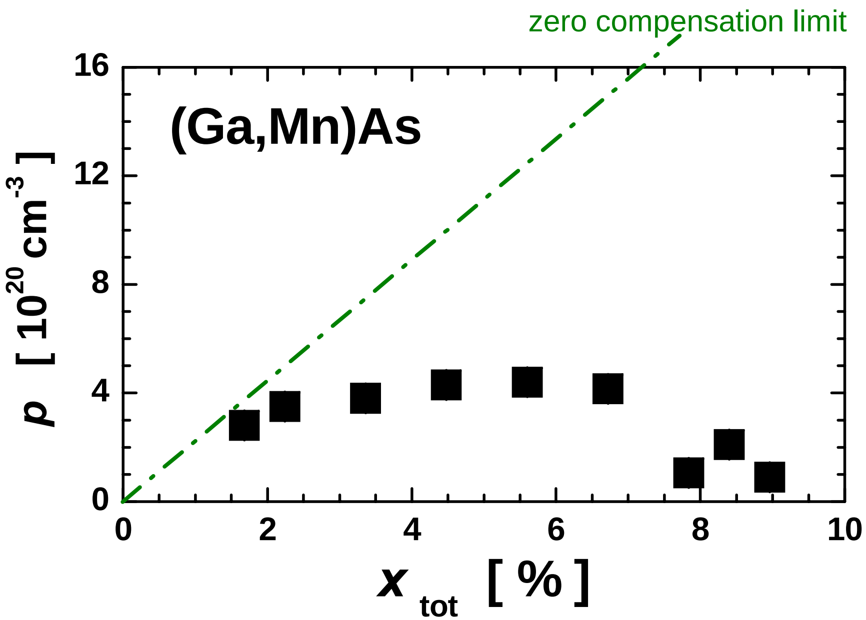


$3d^{4+1}$ = „$3d^5$" A^- $S = 5/2$, $L = 0$

Mn = spin 5/2 + hole

Growth of (Ga,Mn)As





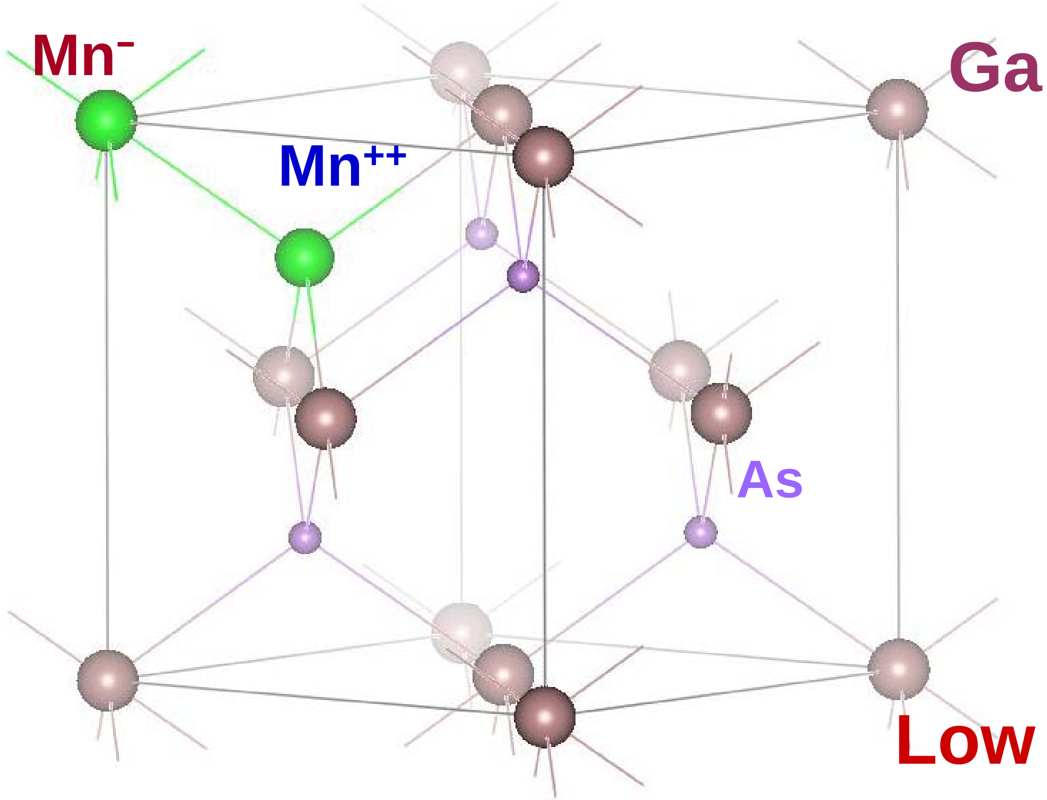
Something went wrong!

K. Yu, et al.

Mn interstitials

c-RBS and **c-PIXE** reveal:
in low-temperature MBE grown ferromagnetic (Ga,Mn)As
Mn atoms occupy three distinct positions in the lattice

substitutional Mn_{Ga}, interstitial Mn_{I}, and random (MnAs)
in proportions depending on annealing.



K. Yu, et al., PRB'02

interstitial Mn_{I}:

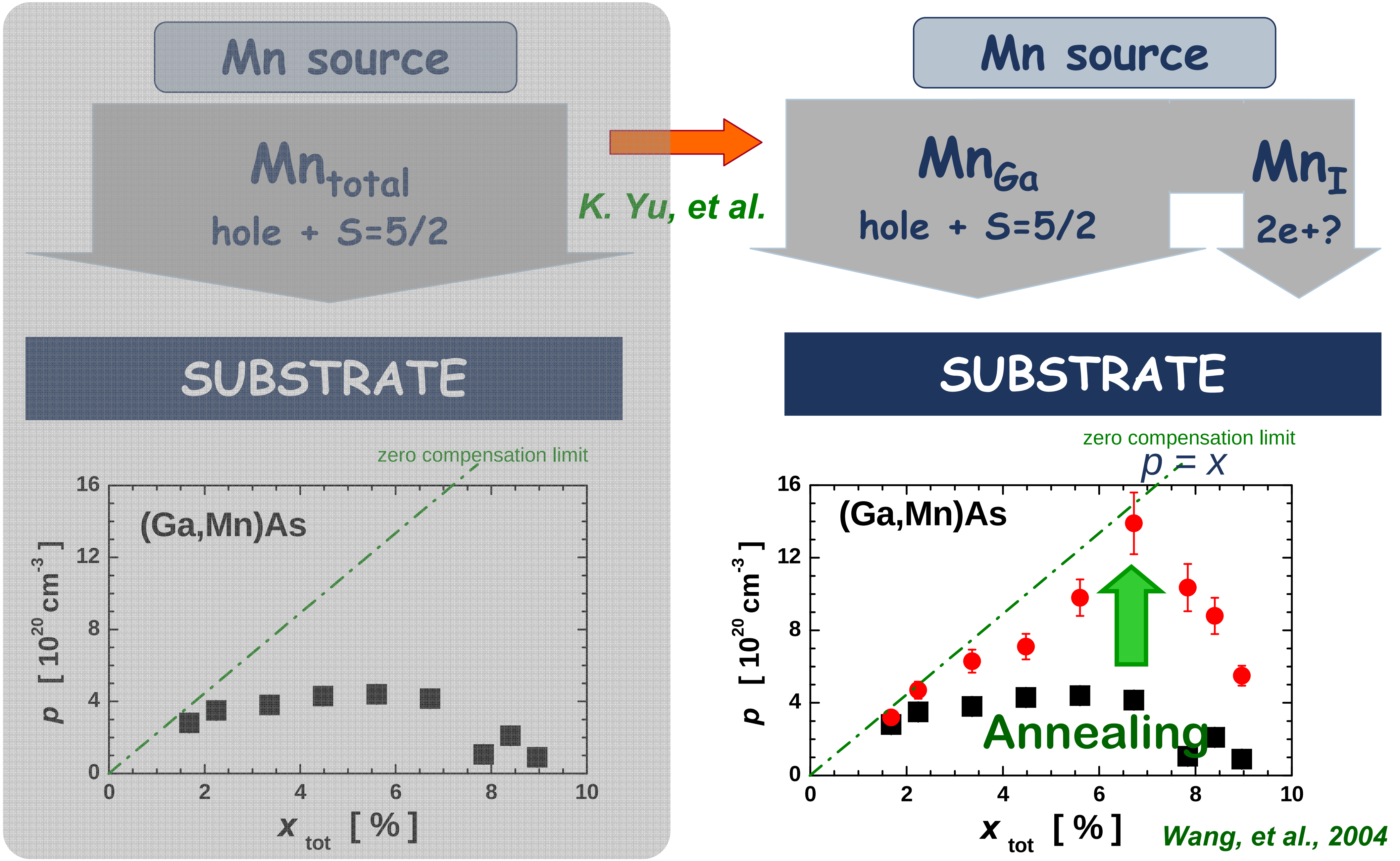
- ☹ **Double donor**
- ☹ **Does not play ferro**
- ☹ **AF bonds to Mn_{Ga}**

Blinowski, Kacman, PRB'03

Low temperature annealing!!

Potashnik et al.,'02

Growth of (Ga,Mn)As



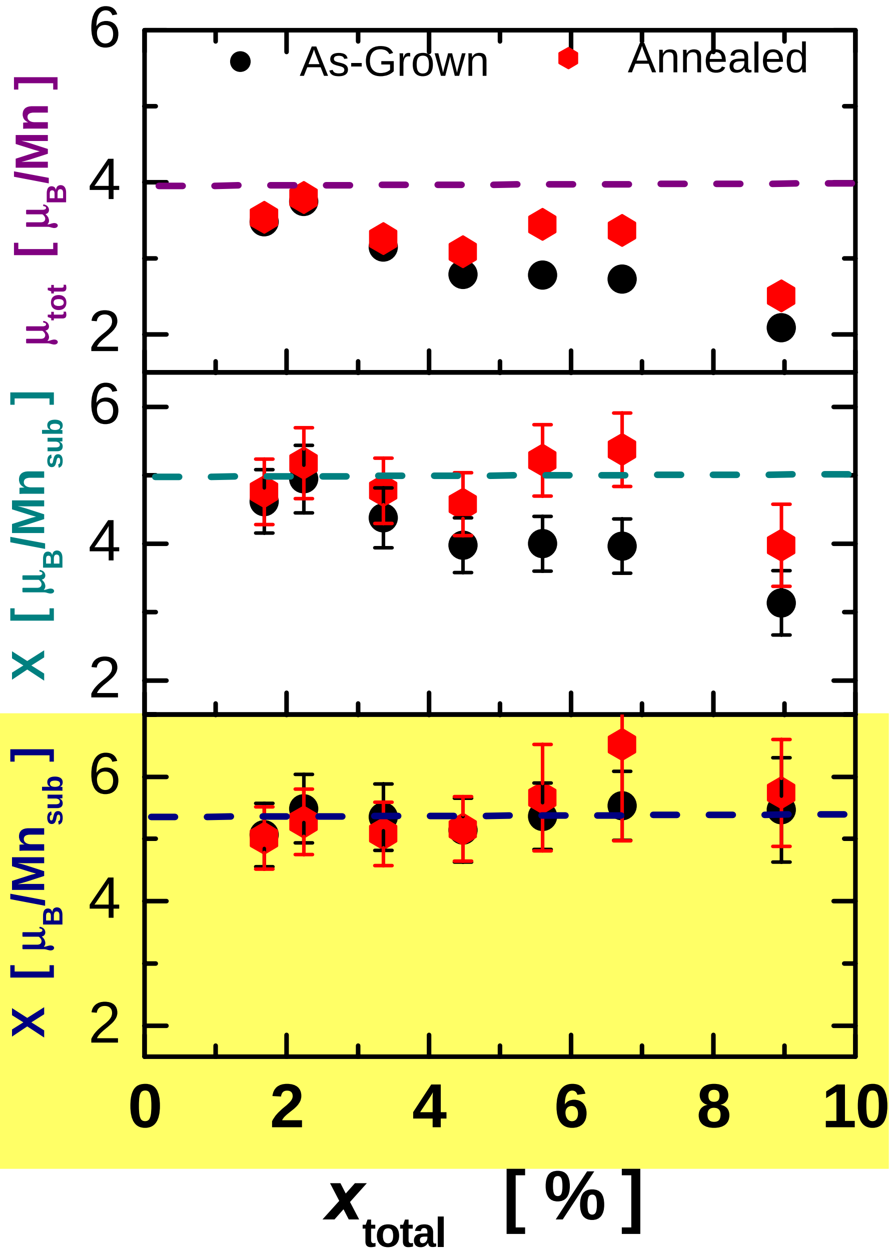
(apparent) 'Magnetisation deficit'

$\mu_{tot} = M_S / x_{tot}$

$M_S = N_0\, x_{eff} \cdot X + p \cdot (-1)$

Mn$_I$ paramagnetic: $x_{eff} = x_{Sub}$

Mn$_I$ AF to Mn$_{Ga}$: $x_{eff} = x_{Sub} - x_I$



... summary

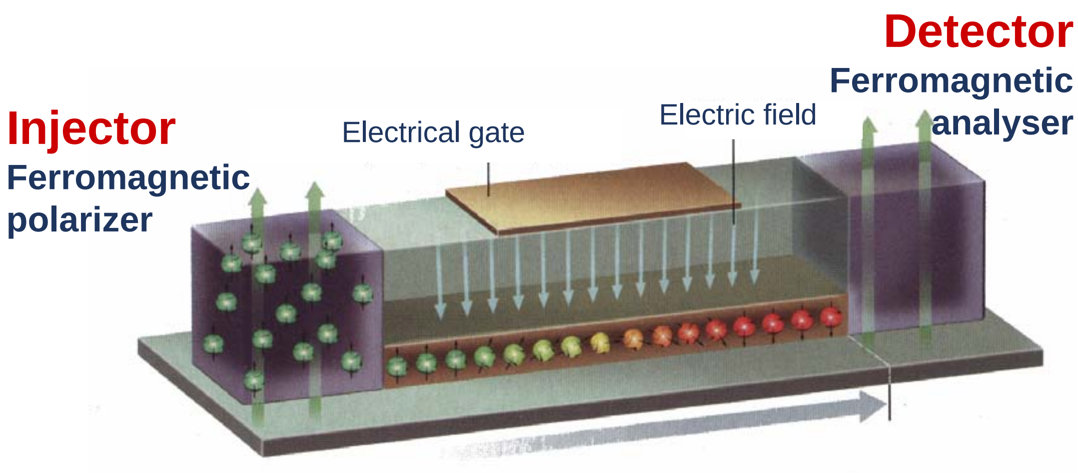
- (Ga,Mn)As emerges as the best understood model ferromagnet with a number of attractive functionalities

- Control of magnetism and magnetization direction is possible by external means

- Beginning of the road for high temperature ferromagnetic semiconducting system

The magnetic anisotropy

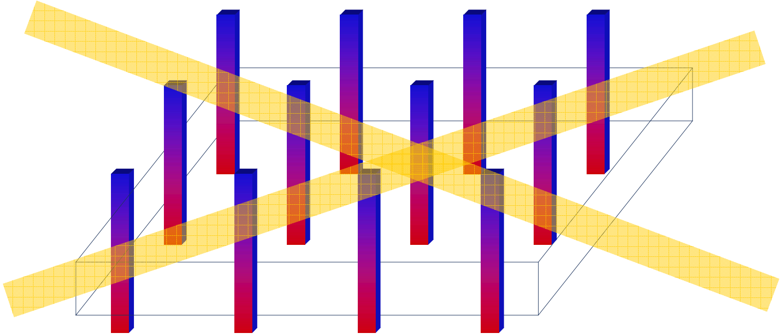
- **Testing/verification for models**
- **Device engineering**
  - magnetoresistive AMR ~ $\cos^2(\measuredangle\, \vec{j}, \vec{M})$
  - spin injection/detection

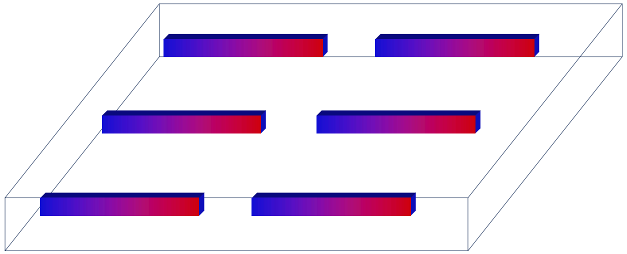


Datta & Das (1990)

- **utilisation of the magnetic anisotropy**

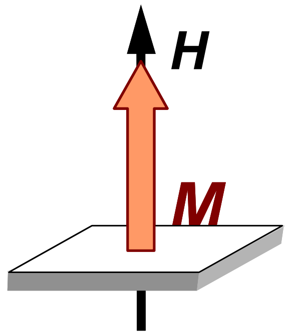
Magnetocrystalline vs. shape anisotropy





Despite the expected for the layered material in-plane arrangement of ***M*** ($H_A = M_S$), relatively strong **perpendicular (uniaxial) magnetic anisotropy** has been observed since the very beginning of the studies:

- (In,Mn)As/GaAs – Munekata '93
- some (Ga,Mn)As/InGaAs – Ohno, Shono '96-'00
- QW (Cd,Mn)Te – Haury '97
- (Ga,Al,Mn)As/GaAs – Takamura '02
- (Ga,Mn)As/GaAs – Sawicki '02



$H_A >> M_S$ ⇒ magnetocrystalline anisotropy dominates over the shape effects

M_S in 5% (Ga,Mn)As ≅ 600 Oe

22000 Oe for Fe

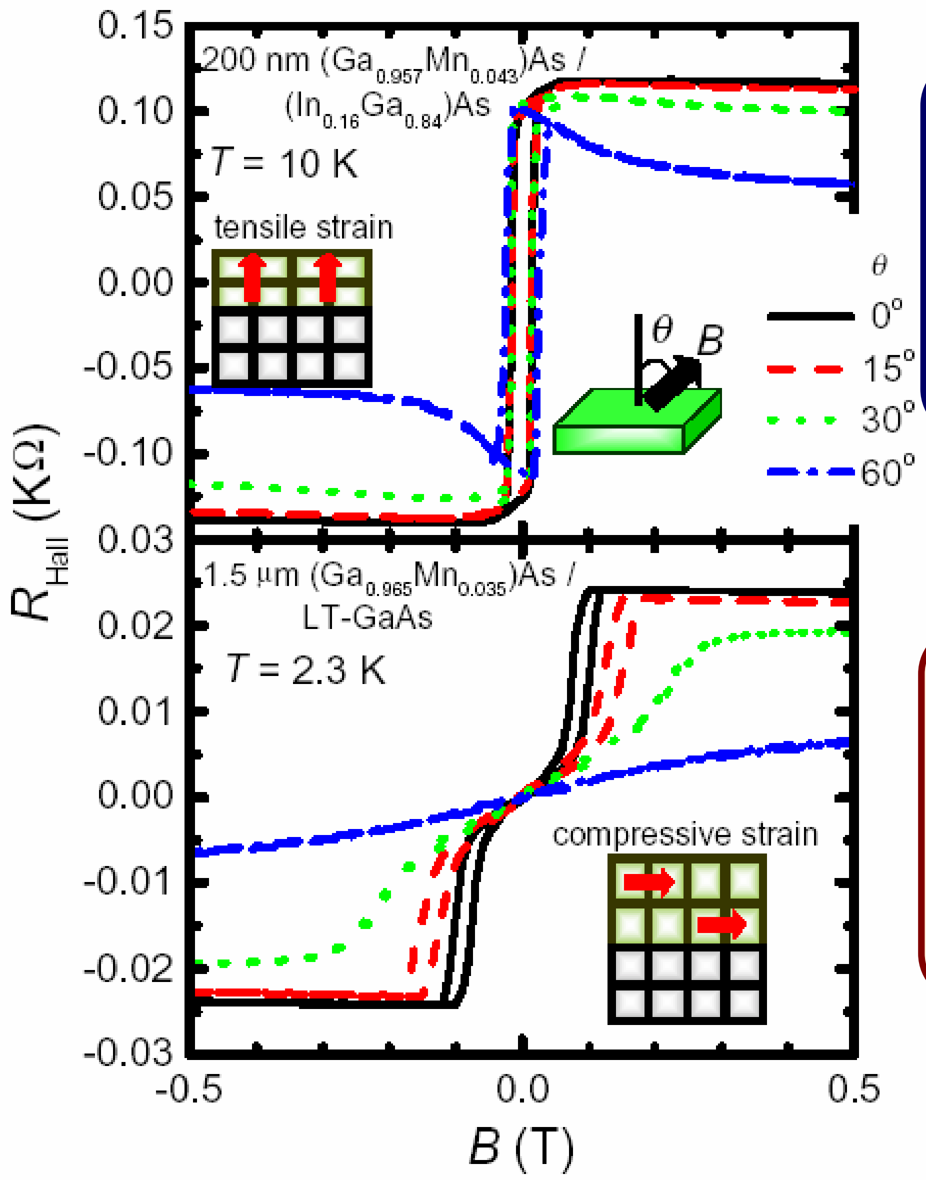
Magnetic anisotropy in cubic materials

T_d symmetry of the host lattice

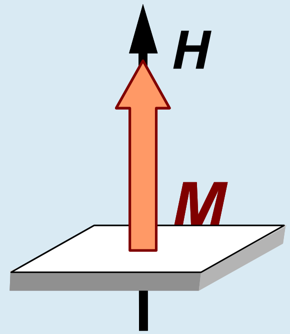
⇓

magnetic anisotropy is expected on **<100>** and **<111>** directions

MA of *p*-DMS: *the epitaxial origin*



Shen et al. 1997 (Sendai)



Tensile strain

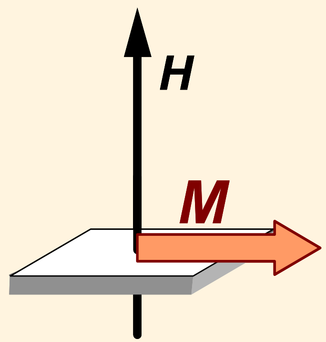
⇓

Perpendicular

Magnetic Anisotropy

Marginal role of the shape anisotropy!!

$K_s = \boldsymbol{M}^2(D_a - D_c)/2$



Compressive strain

⇓

In plane

Magnetic Anisotropy

+ lots of confusing information

? [100], [110] ?

about in plain easy axis

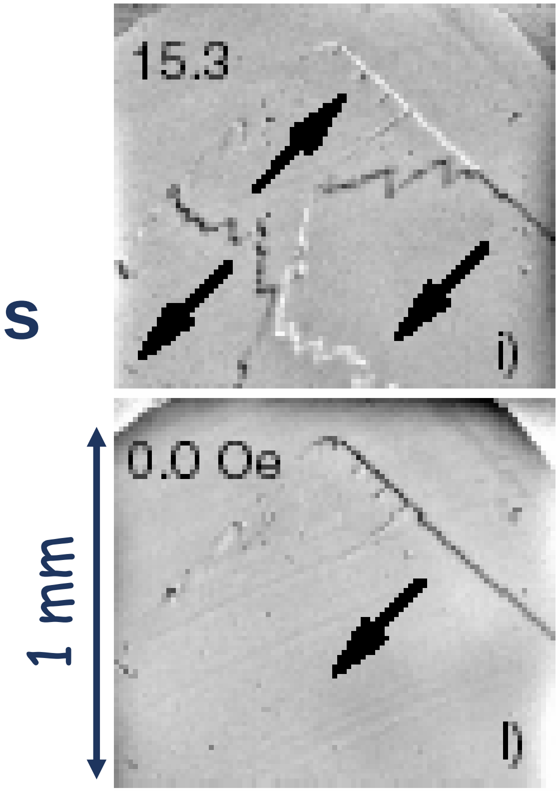
Excellent micromagnetic properties

- **Large values of K_a (= $M_S H_a / 2$) and A hinder domain formation**

Domain wall energy $E = (K_a A)^{1/2}$

- **Dilute systems: low M_S**

(Ga,Mn)As



Welp et al., PRL'03

Magnetic anisotropy – the origin

•EPR studies shows that Mn single ion anisotropy is negligible *Fedorych et al., 2002*

• p-d Zener Model - Mn - Mn interaction is mediated by holes, characterised by a non-zero orbital momentum

Dietl, Ohno, Matsukura, PRB 2001
(cf. Abolfath, Jungwirth, Brum, MacDonald, PRB 2001)

↓

It is the anisotropy of the carrier-mediated exchange interaction stemming from spin-orbit coupling of hole gas.

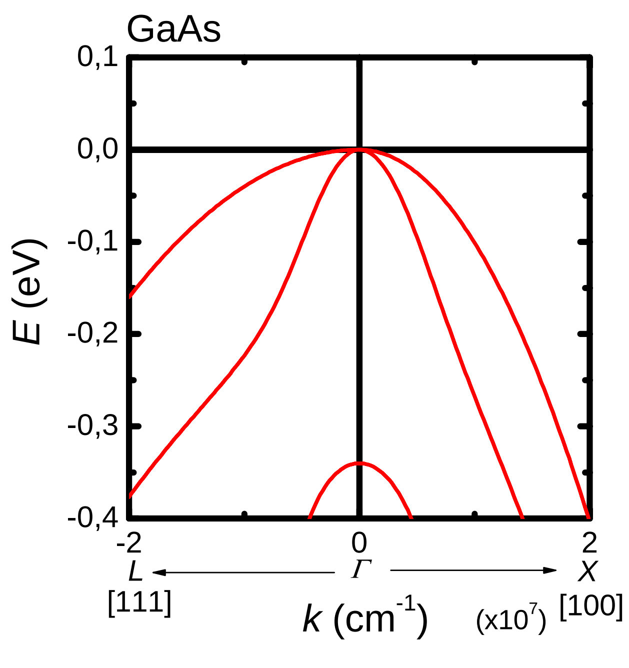
Valence band structure (Zinc-blende Γ_7 and Γ_8)

Schrödinger equation: $(H_{kp} + H_{pd} + H_{bs})\Psi = E\Psi$

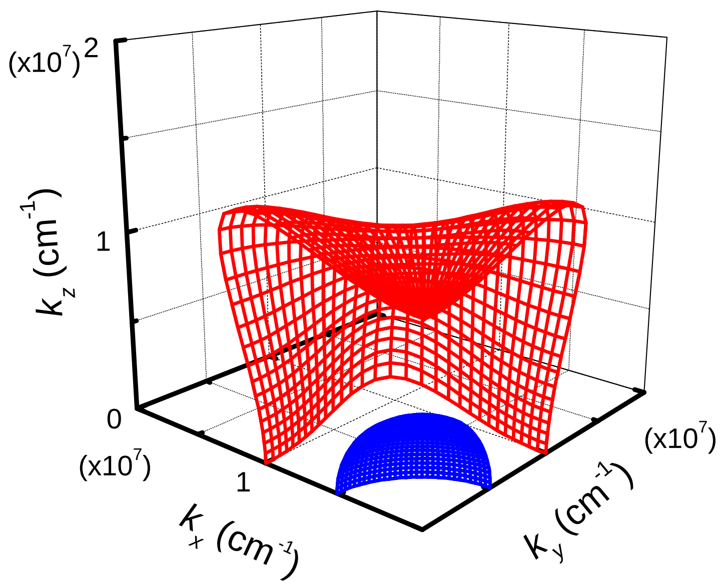
basis function:

$$u_1 = \frac{1}{\sqrt{2}}(X + iY)\uparrow, \quad u_2 = i\frac{1}{\sqrt{6}}[(X + iY)\downarrow - 2Z\uparrow], \quad u_3 = \frac{1}{\sqrt{6}}[(X - iY)\uparrow + 2Z\downarrow],$$

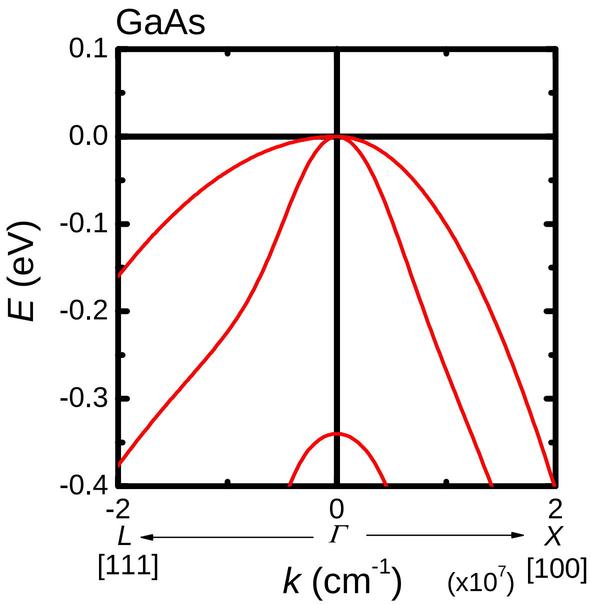
$$u_4 = i\frac{1}{\sqrt{2}}(X - iY)\downarrow, \quad u_5 = \frac{1}{\sqrt{3}}[(X + iY)\downarrow + Z\uparrow], \quad u_6 = i\frac{1}{\sqrt{3}}[-(X - iY)\uparrow + Z\downarrow].$$

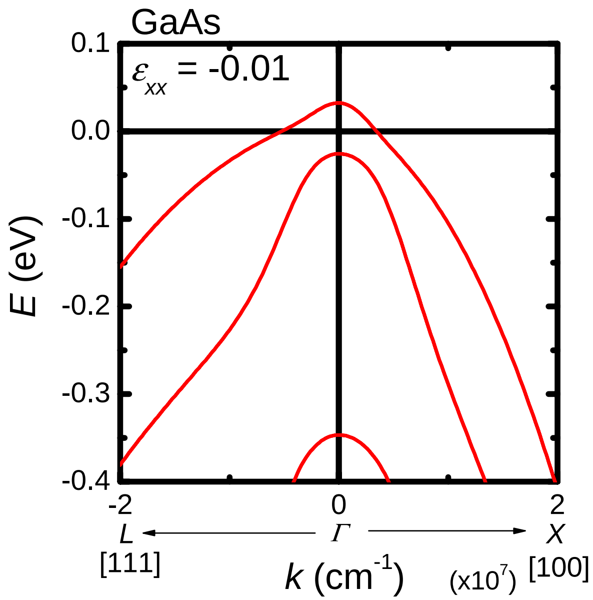


Fermi Surface at E_F = 100 meV



Dispersion of strained (Ga,Mn)As





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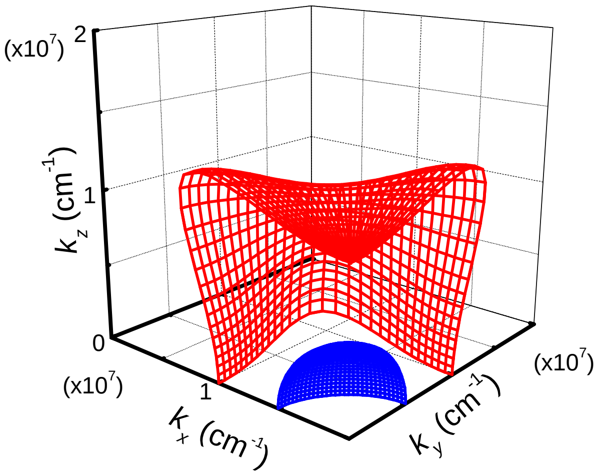
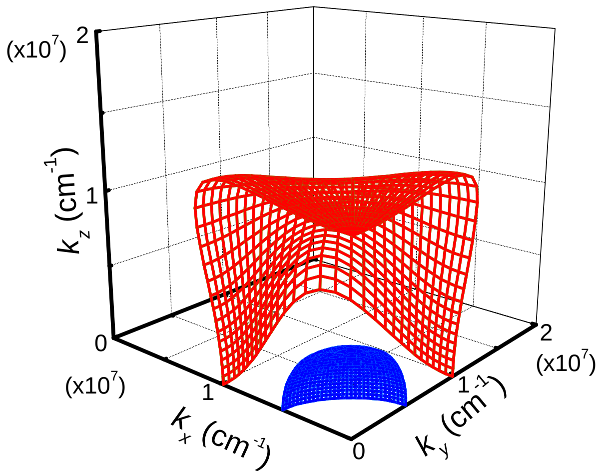
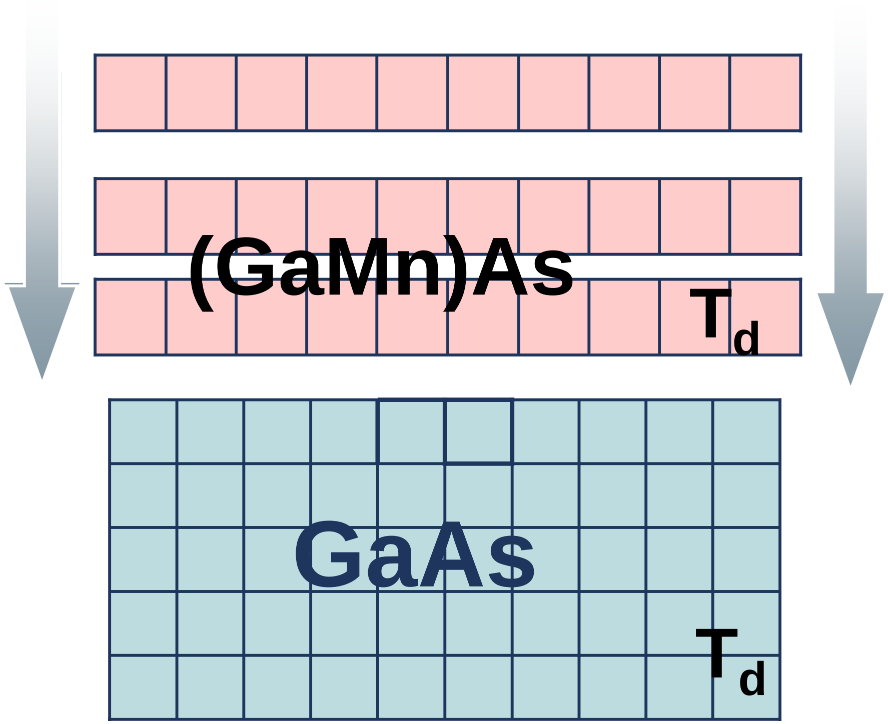
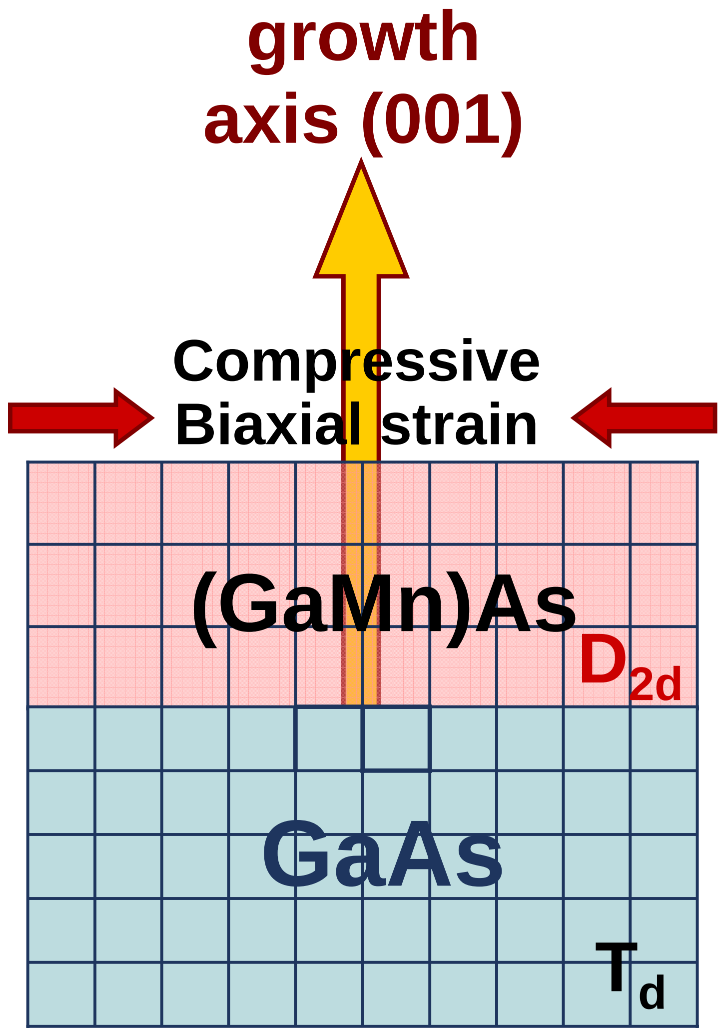




Uniaxial MA – epitaxial origin

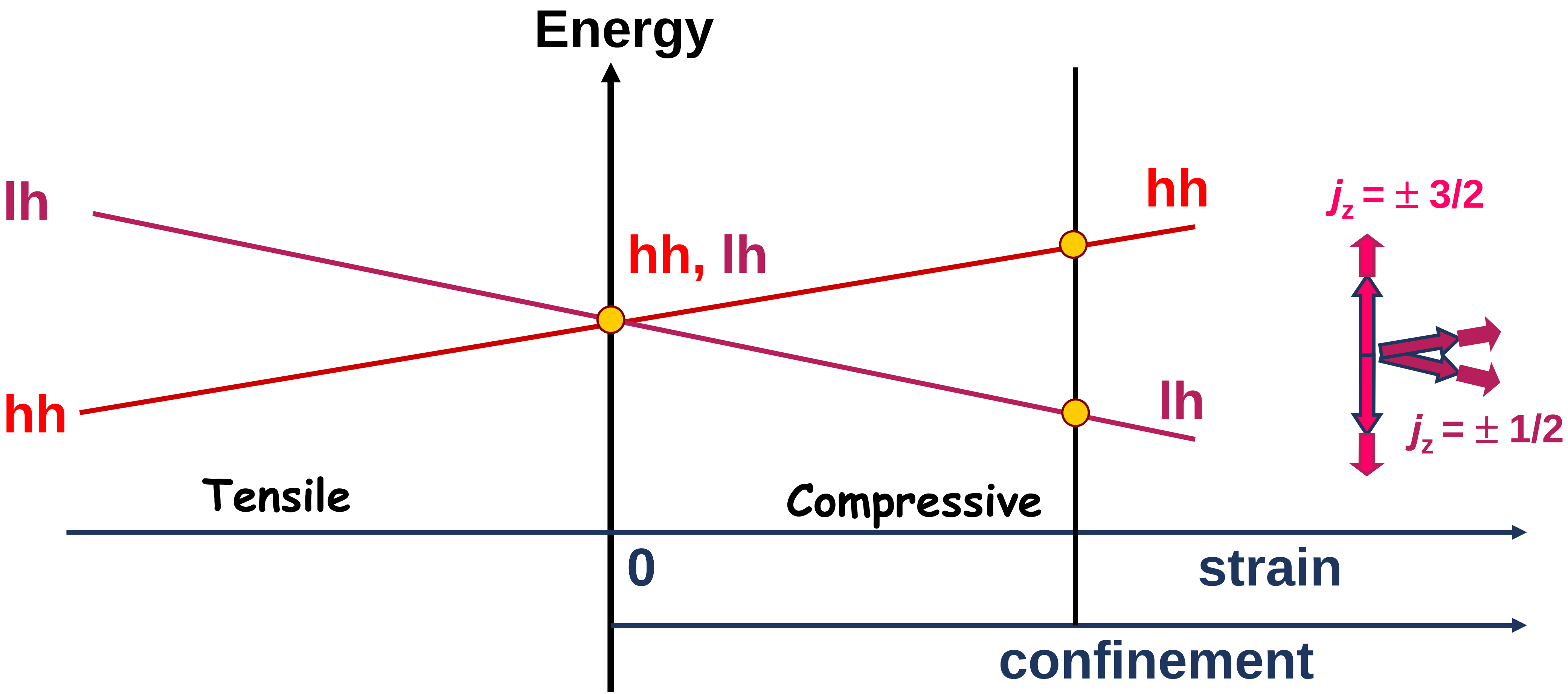
Pseudomorphic low temperature MBE growth:





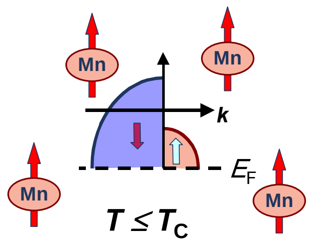
Uniaxial MA – epitaxial origin

1. strain, confinement or both split the hh from lh

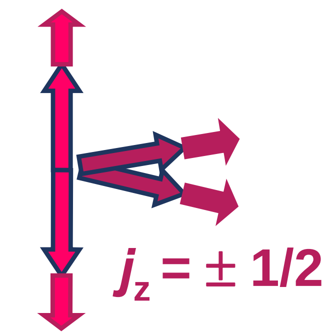


Uniaxial MA – epitaxial origin

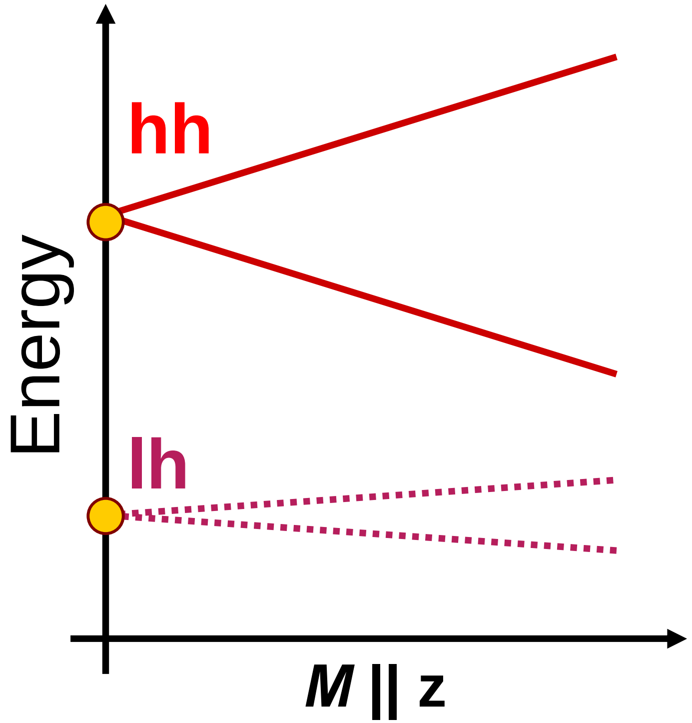
1. **strain, confinement or both split the hh from lh**
2. if $M \neq 0$ the lower energy state: • for **hh** (l=±1) when $k \perp M$

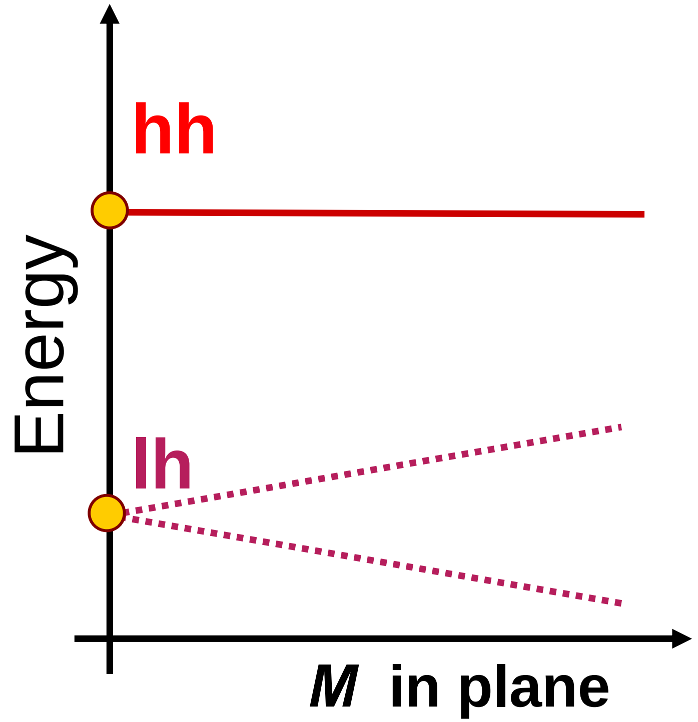


Compressive case, low hole density



$\sim J\, s \cdot S$

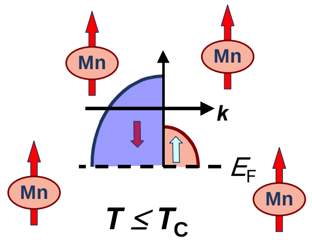




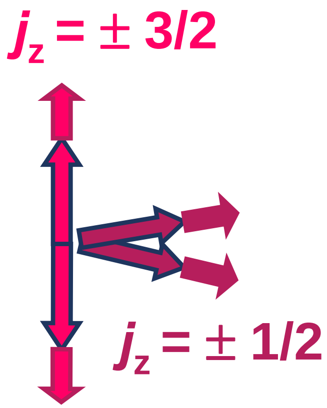
• **hh. subband occupied ➔ easy [001] ($K_U > 0$; strong)**

Uniaxial MA – epitaxial origin

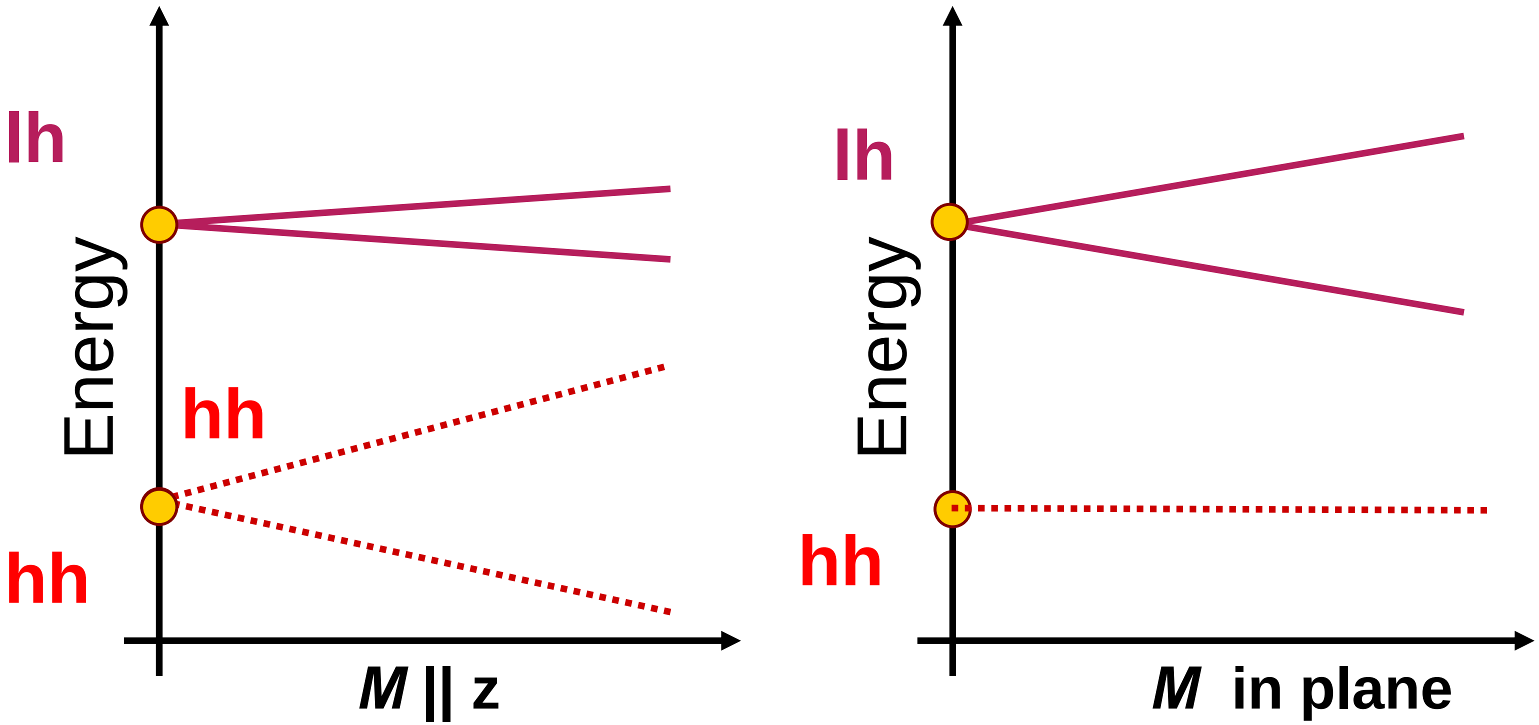
1. **strain, confinement or both split the hh from lh**
2. if $\boldsymbol{M} \neq \boldsymbol{0}$ the lower energy state: • for **lh** (l=0) when $\boldsymbol{k} \parallel \boldsymbol{M}$



Tensile case, low hole density



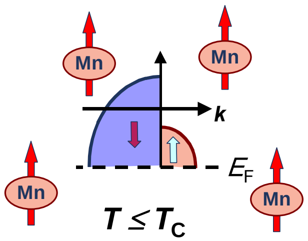
$\sim J\, s \cdot S$

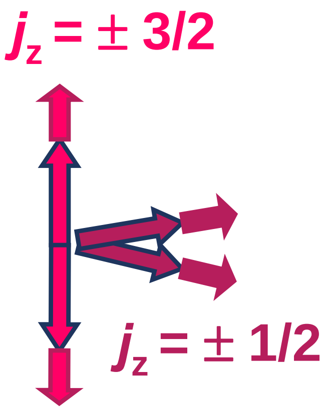


• **hh. subband occupied ➔ easy (001) ($K_U < 0$; weak)**

Magnetic anisotropy – epitaxial origin

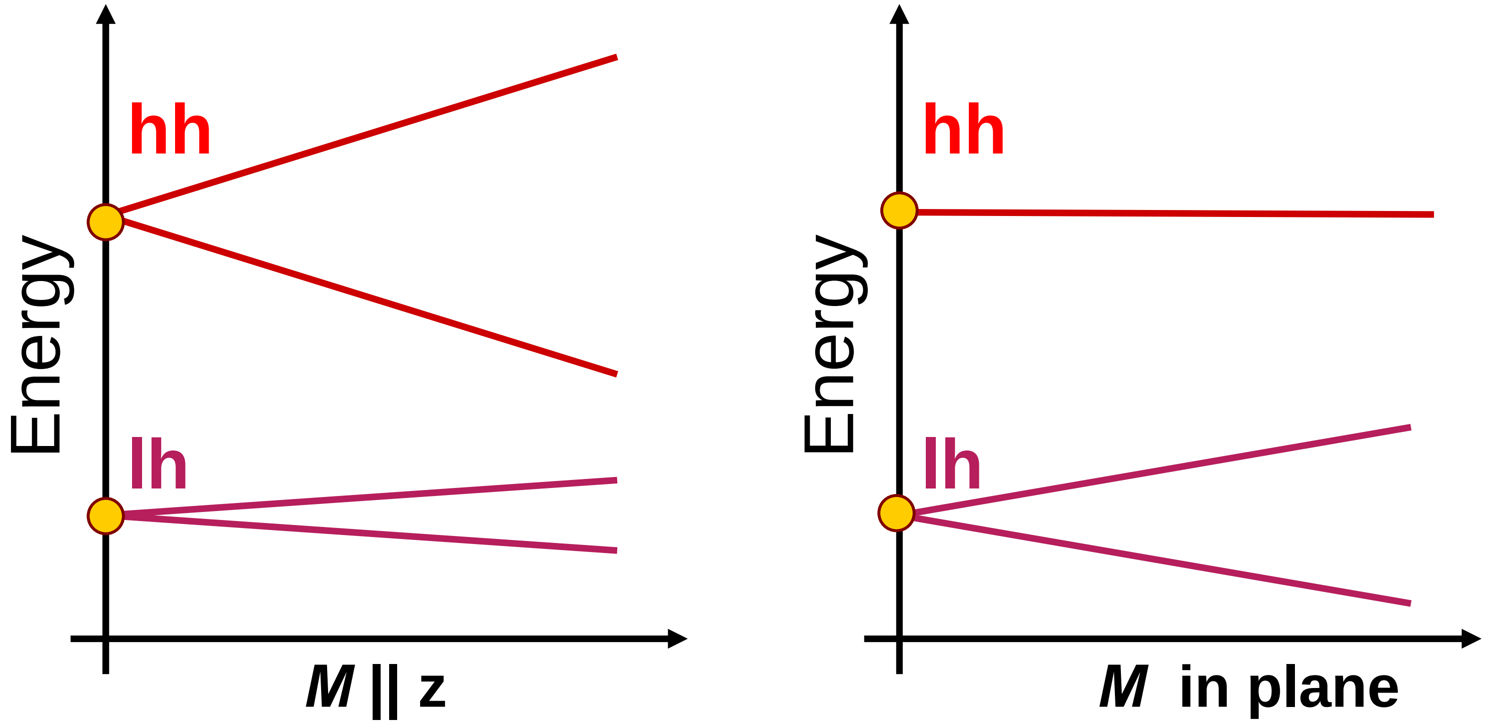
Epitaxial (biaxial) strain ⇒ Splitting of the hole states





$\sim J\, s \cdot S$

Compressive case:

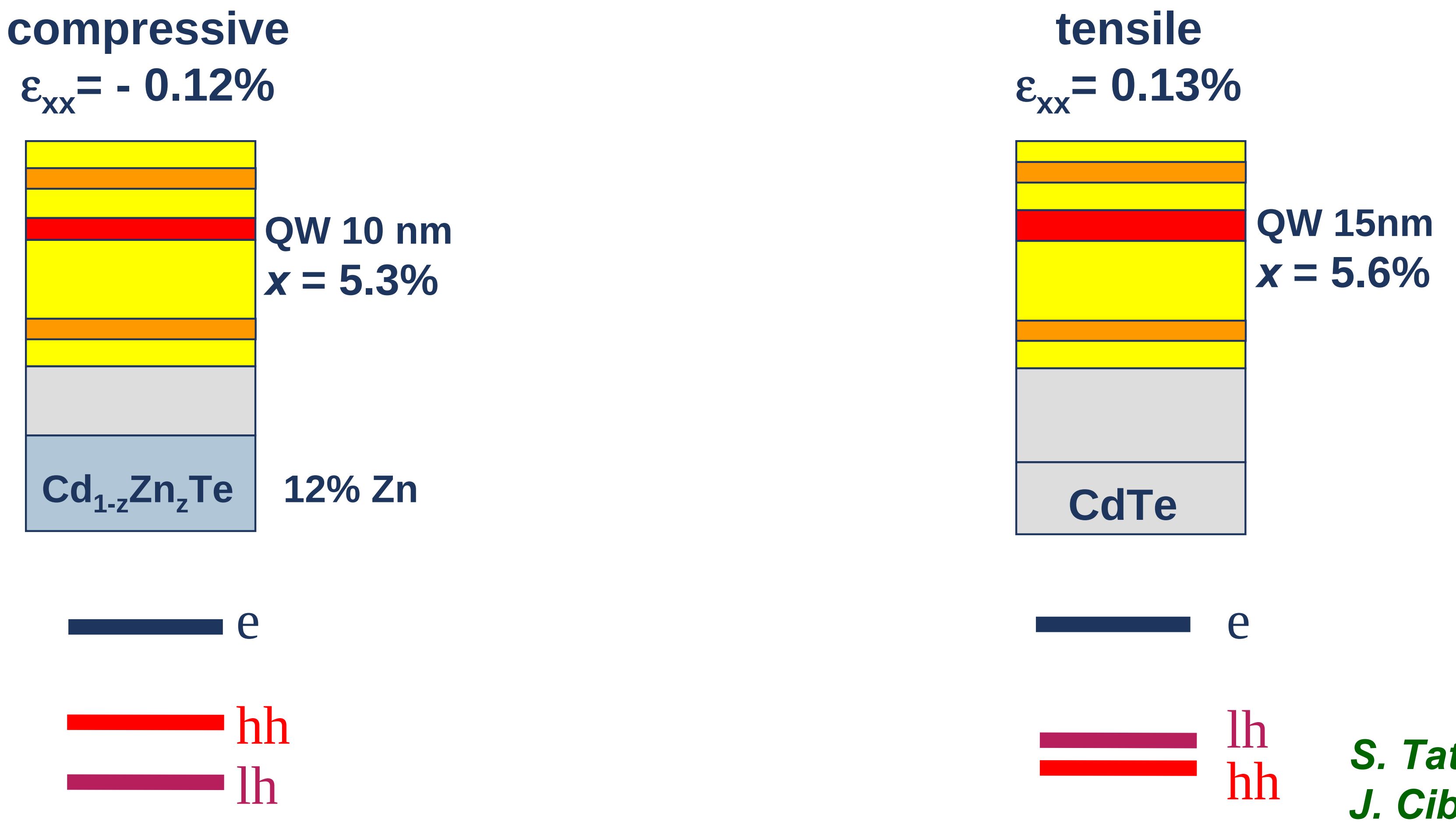


⇒ uniaxial anisotropy

- hh. subband occupied ➔ perpendicular anisotropy (strong)
- lh. subband occupied ➔ in-plane anisotropy (weak)

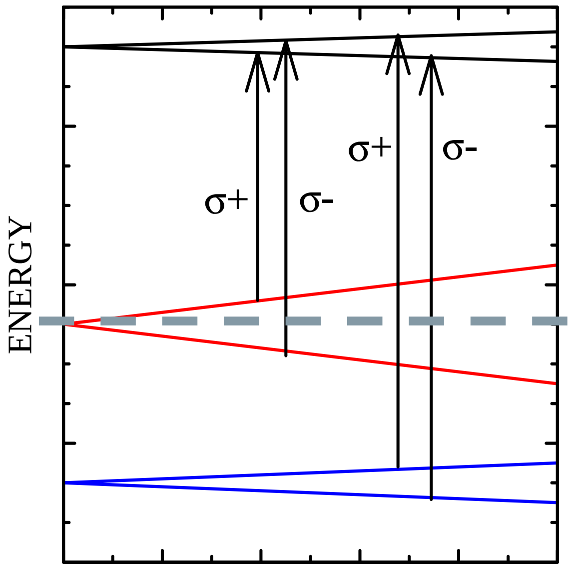
Valence band engineering – (Cd,Mn)Te QW

Compensation of confinement induced hh/lh splitting by epitaxial tensile strain

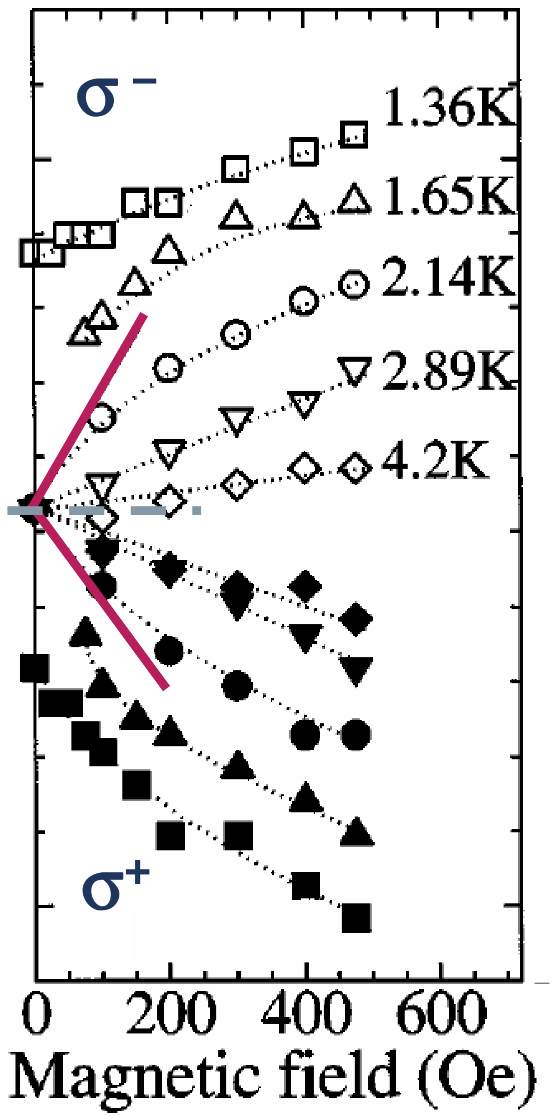


S. Tatarenko
J. Cibert
(Grenoble)

Faraday configuration



ΔE ~ M



$\chi =$
$\delta(E^{-} - E^{+})/\delta H$
at $H \rightarrow 0$

(Warsaw, Grenoble)

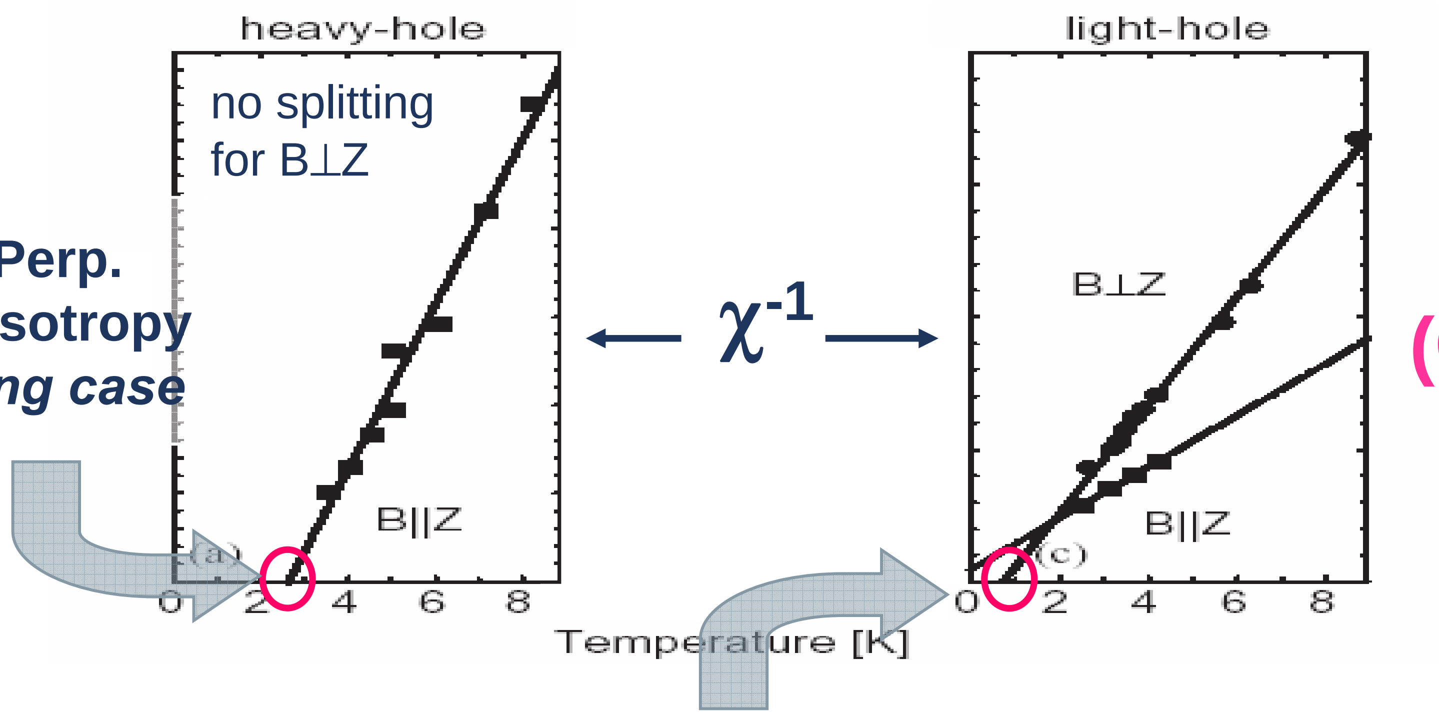
Tailoring the magnetic anisotropy

P. Kossacki et al., Physica E'04

By strain

(Cd,Mn)Te QW

Perp. anisotropy ***Ising case***



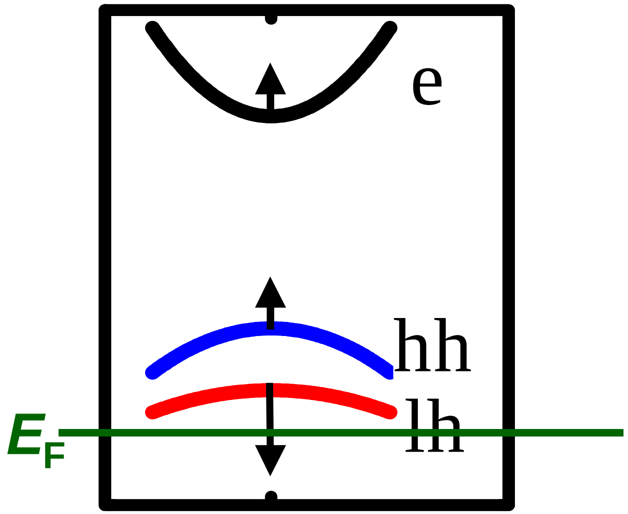
In-plane like anisotropy

hh/lh influence on uniaxial anisotropy

$T_{2d} \Rightarrow D_{2d}$ symmetry lowering, growth direction is the quantisation axis, hh/lh population plays decisive role

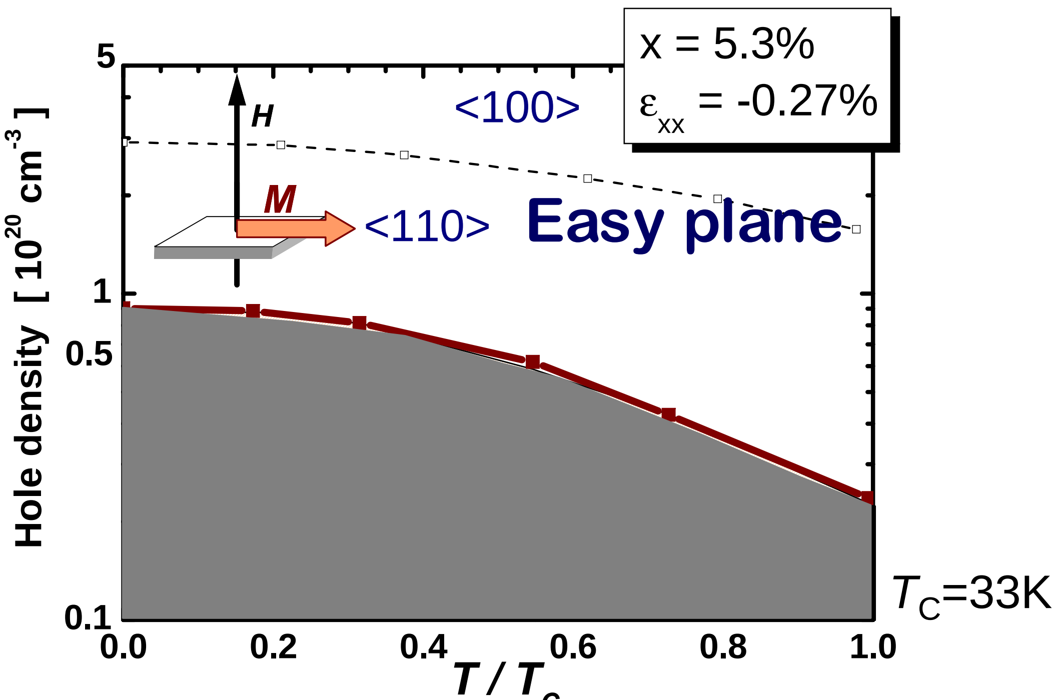
$$K_u = f\,(\,k{\cdot}p\ 6{\times}6\ H + \ H_{p\text{-}d},\ H_{Strain}\,)$$

For biaxial **compression**



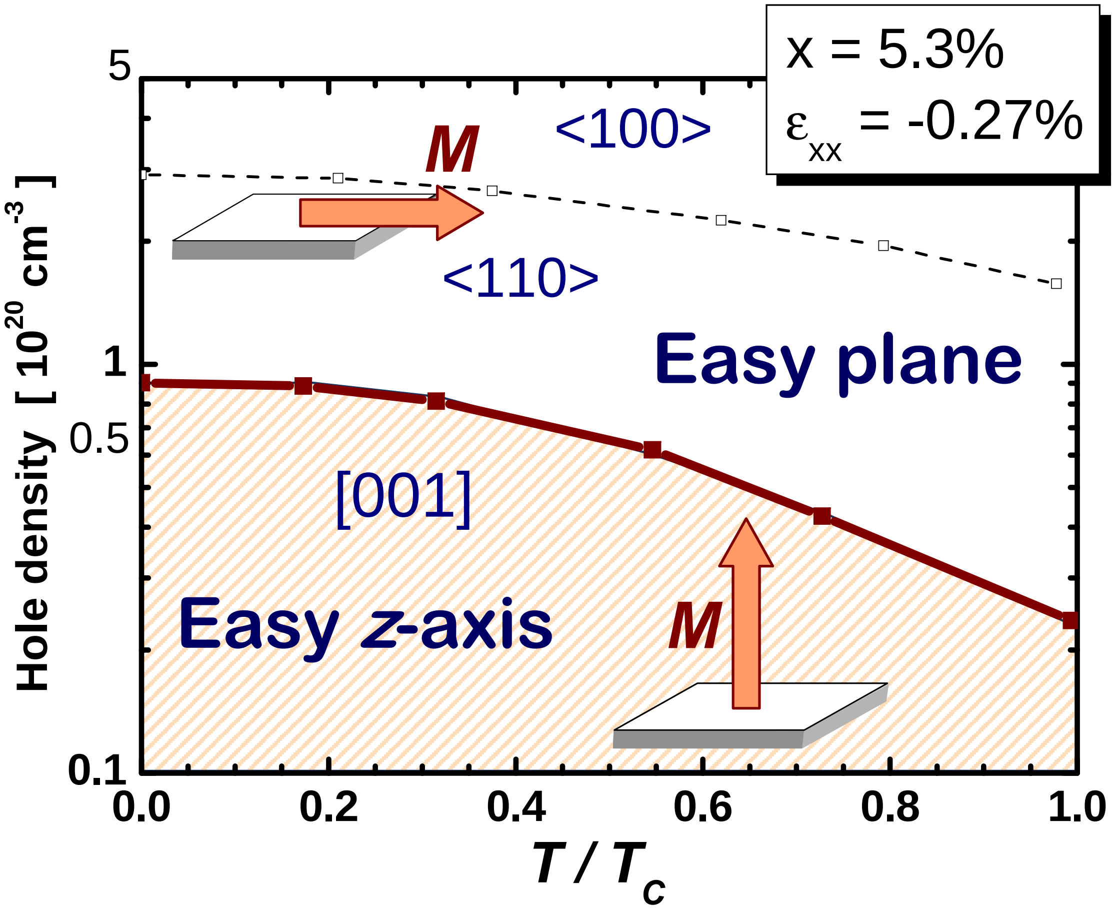
Typically,

easy axis in plane



Calculations: Dietl, Ohno, Matsukura, PRB 2001

hh/lh influence on anisotropy

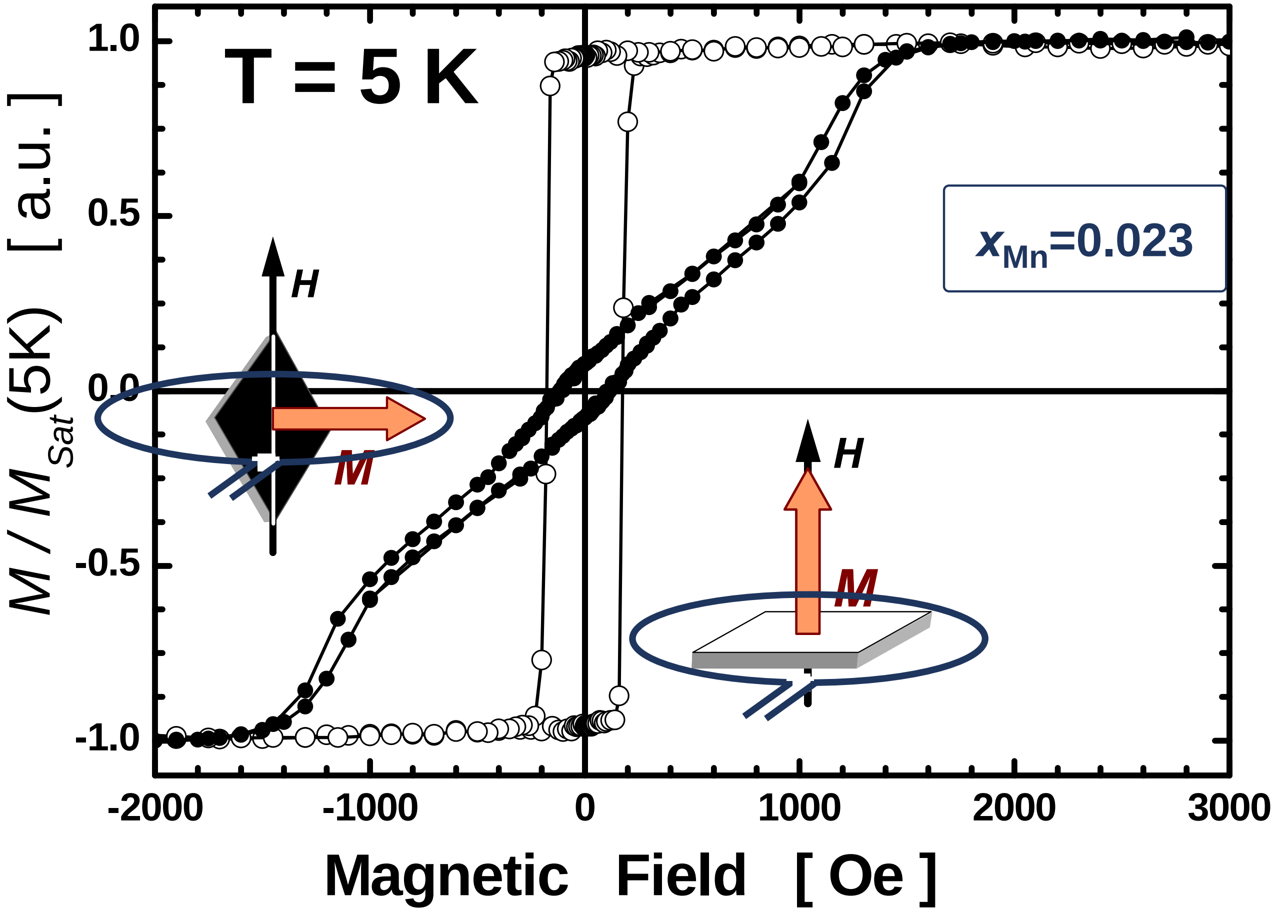


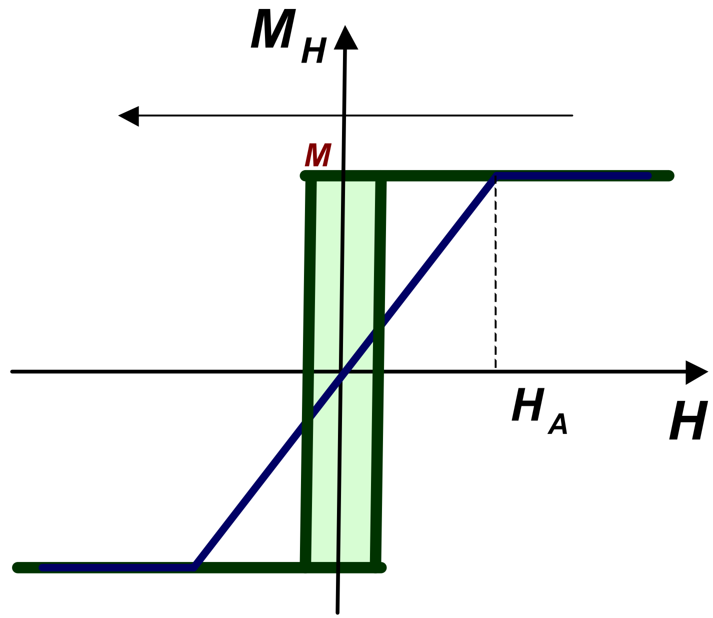
Two important features emerge:

1) **Both types of anisotropy possible**
2) **2nd order phase transition in-between**

Perpendicular magnetic anisotropy

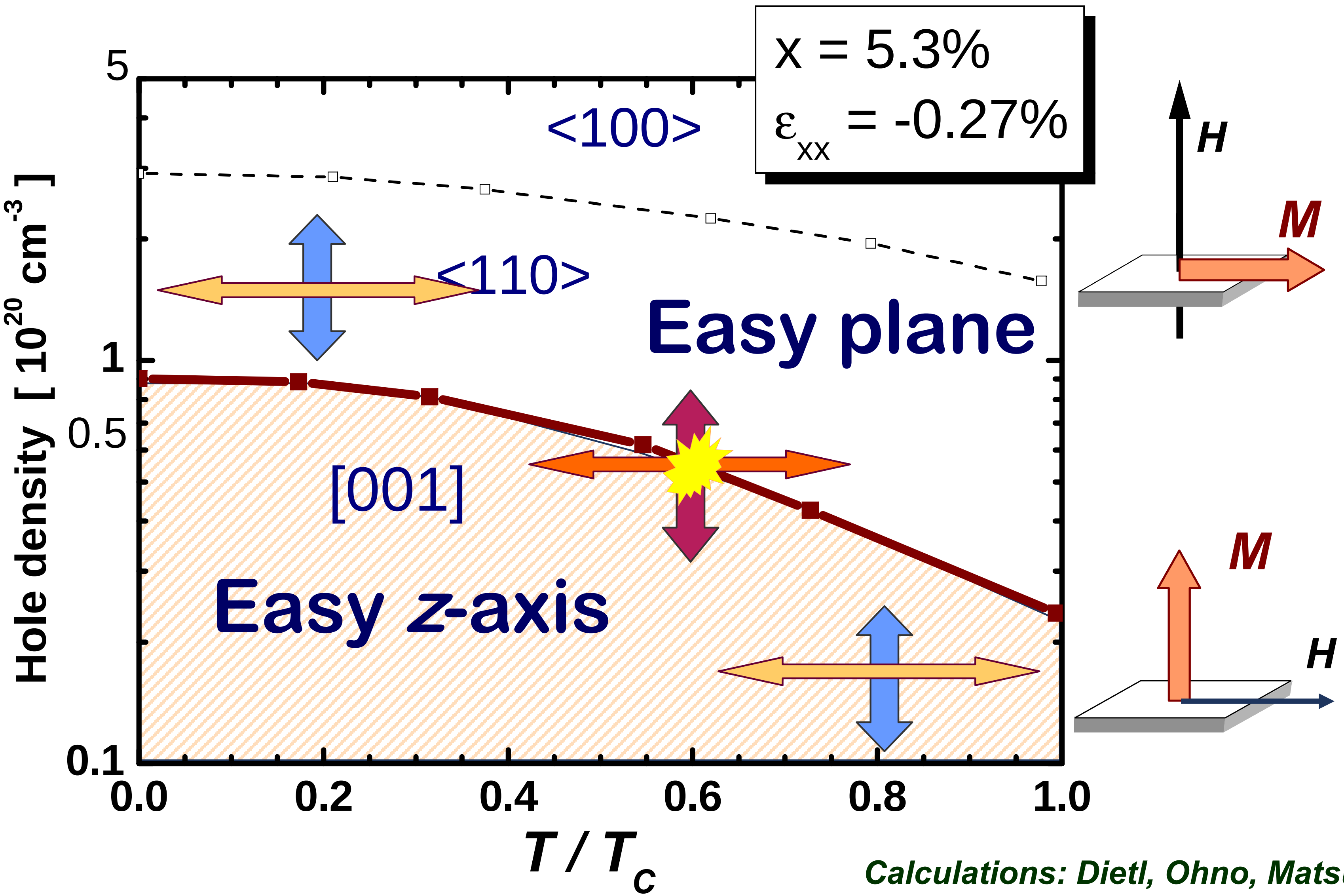
1) For low enough *p* perpendicular magnetic anisotropy in compressively strained (Ga,Mn)As/GaAs is observed (in-plane for tensile case)





hh/lh influence on anisotropy

2) The reorientation: easy axis ⇔ easy plane

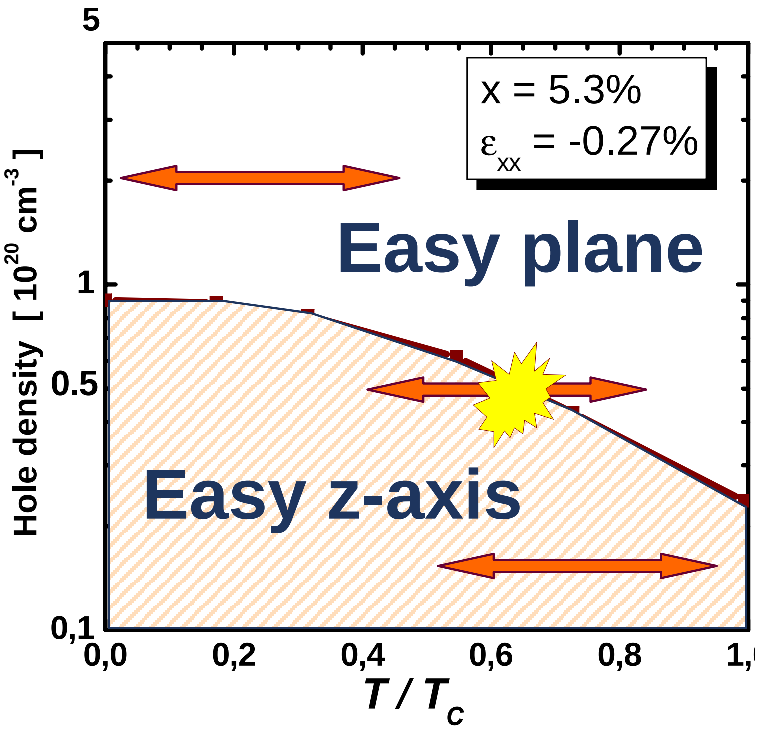


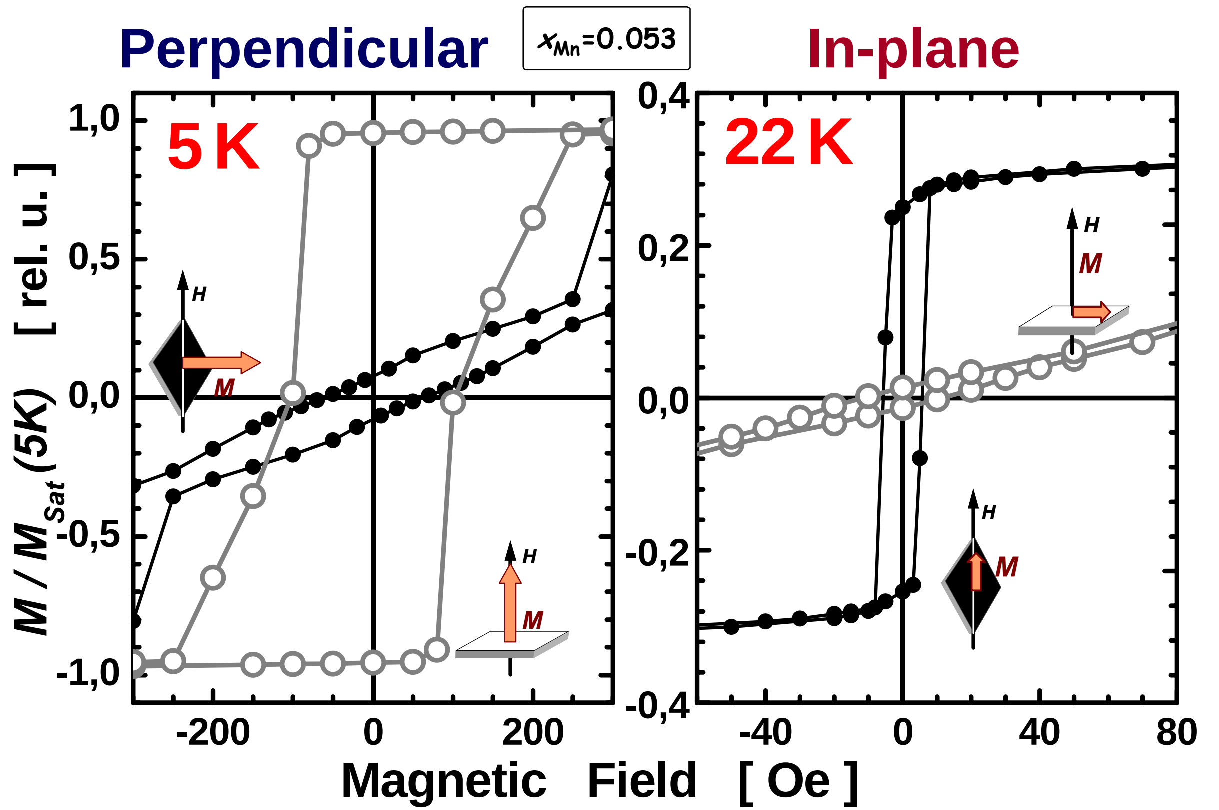
Calculations: Dietl, Ohno, Matsukura, PRB 2001

The reorientation transition: temperature

Temperature influence on hh/lh population ratio:

$$h\omega_s \sim M = f(T)$$





M. Sawicki, et al., PRB '04

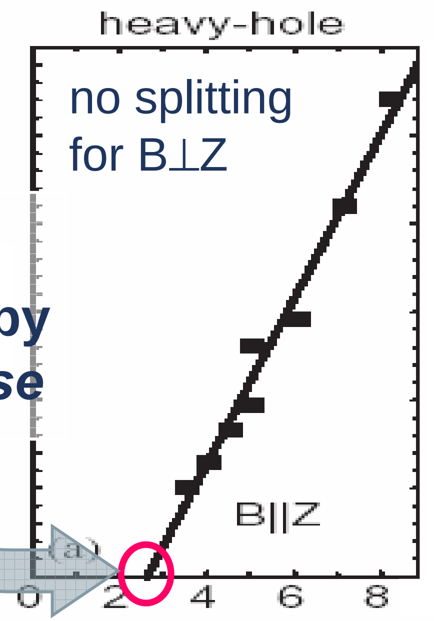
Tailoring the magnetic anisotropy

P. Kossacki et al., Physica E'04

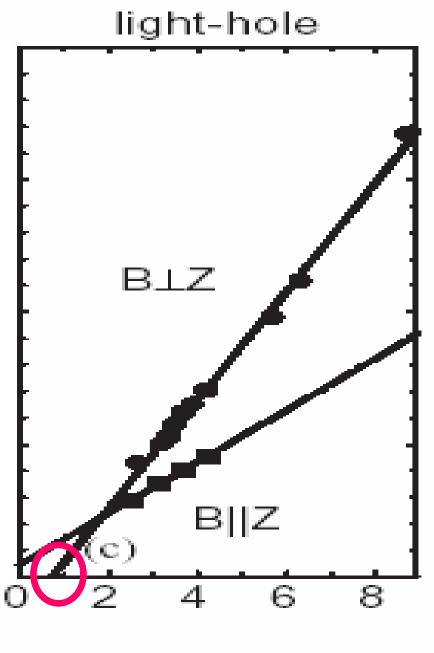
By strain

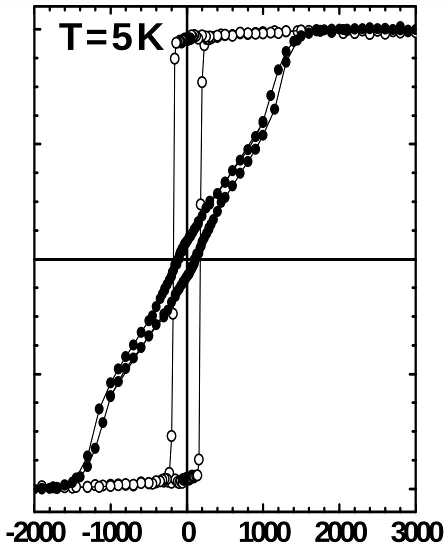
(Cd,Mn)Te QW

Perp. anisotropy *Ising case*

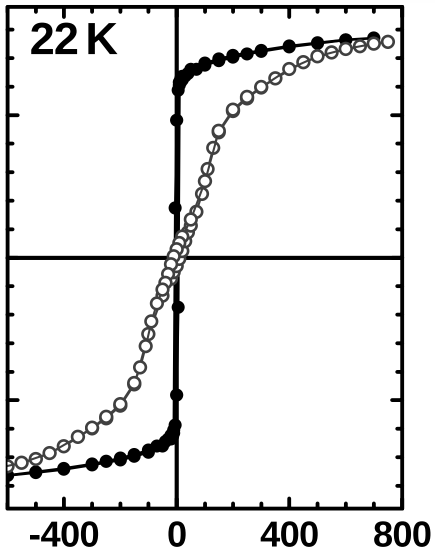


χ^{-1}





In-plane like anisotropy



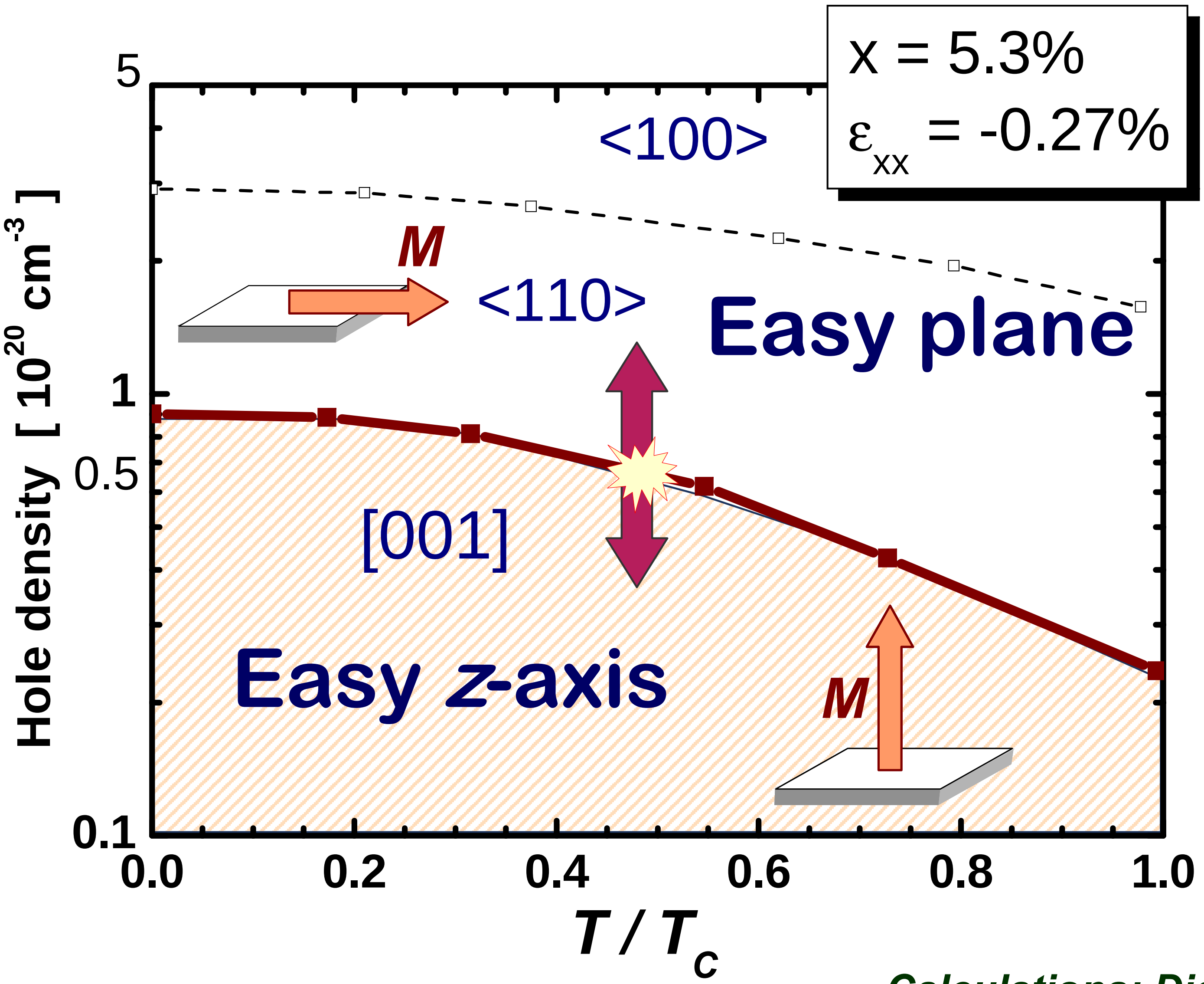
Magnetic Field [Oe]

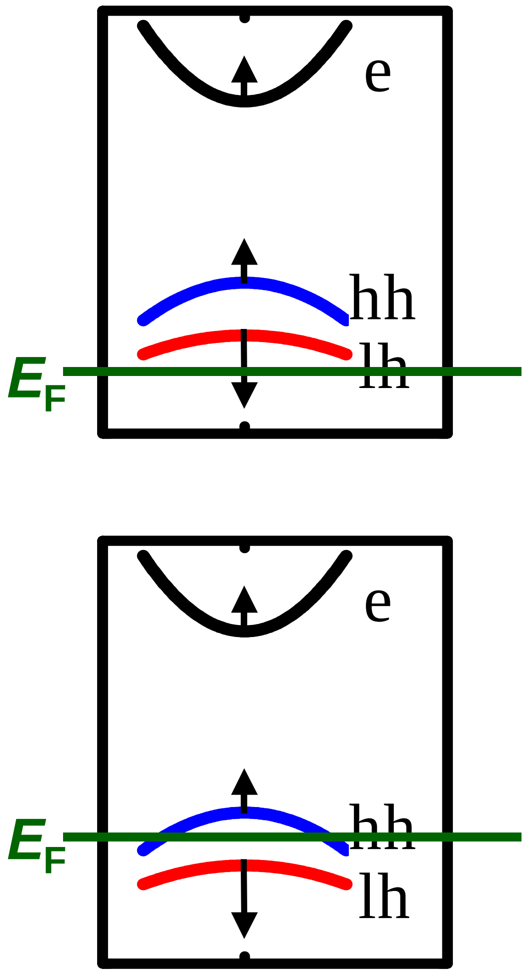
Compressed (Ga,Mn)As

By temperature

(and hole density)

The reorientation transition: hole density

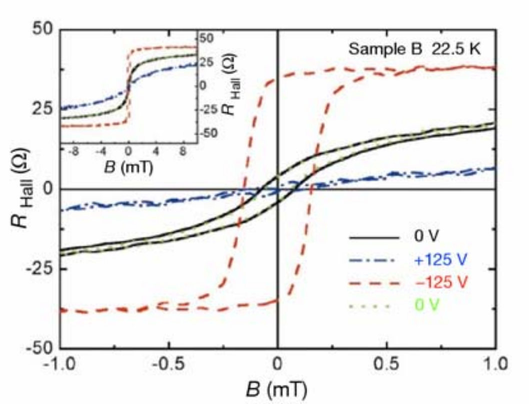




Calculations: Dietl, Ohno, Matsukura, PRB 2001

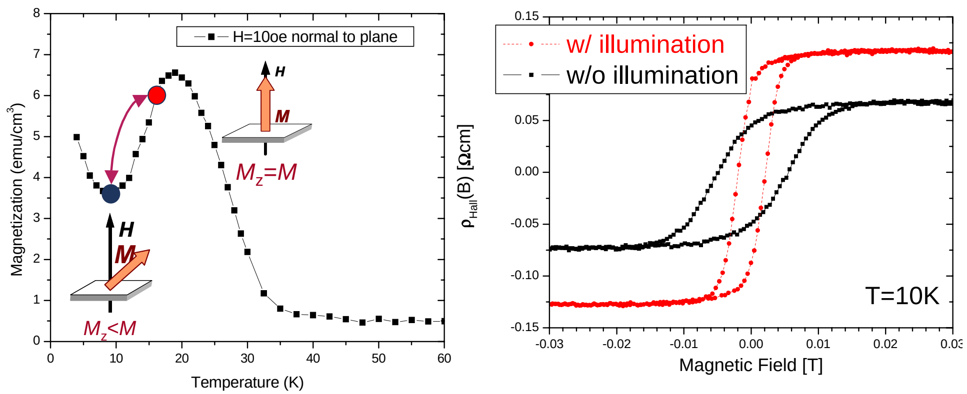
The reorientation transition: hole density

- **Gate**



H. Ohno et al., Nature'00

- **Light** InMnAs/GaSb heterojunction



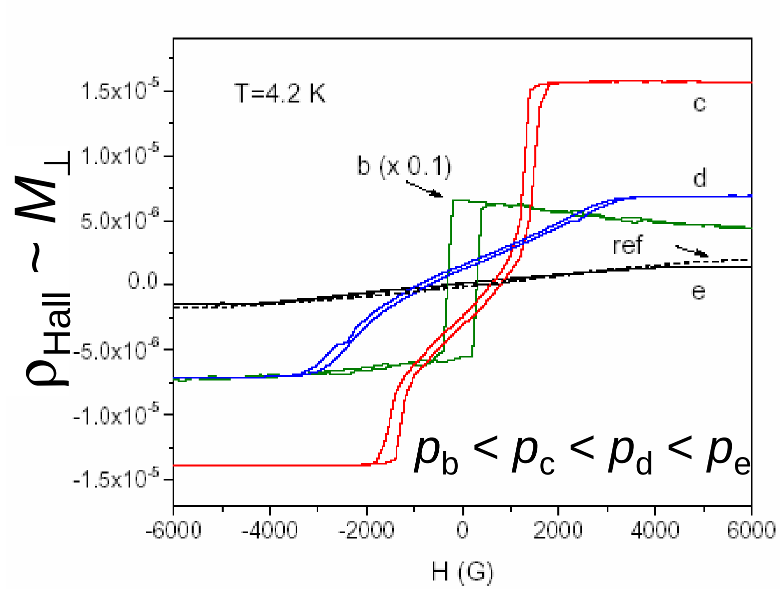
Koshihara et al.,'97; X. Liu et al., '04

- **Compensation**
  - Hydrogenation

    Lemaitre et al., 27 ICPS '04

    Brandt et al., '04

  - **LT annealing**



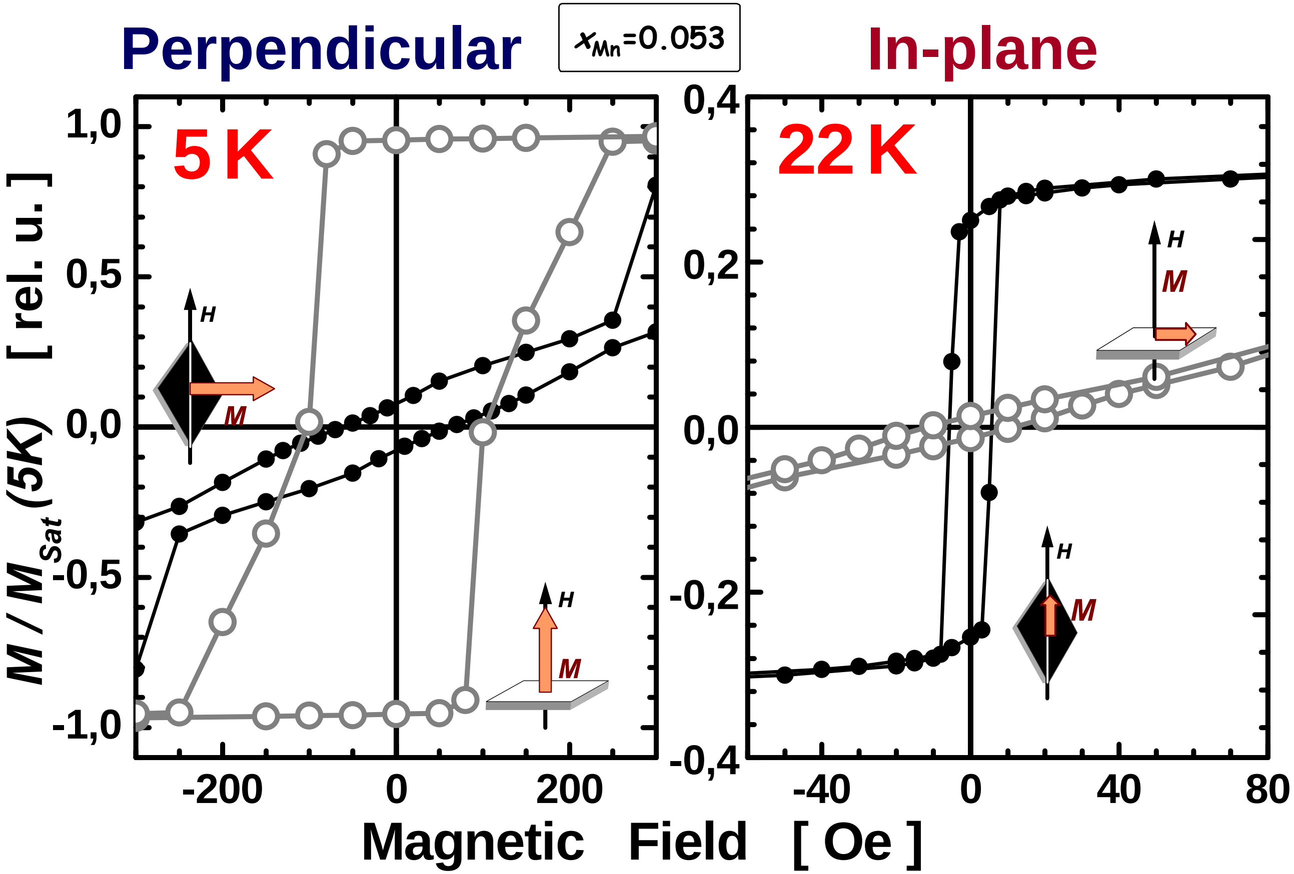
Penn State '02, Nottingham '03, & everywhere else

Thevenard, et al., '05

Hole density change: LT annealing

Post growth LT annealing increases hole density

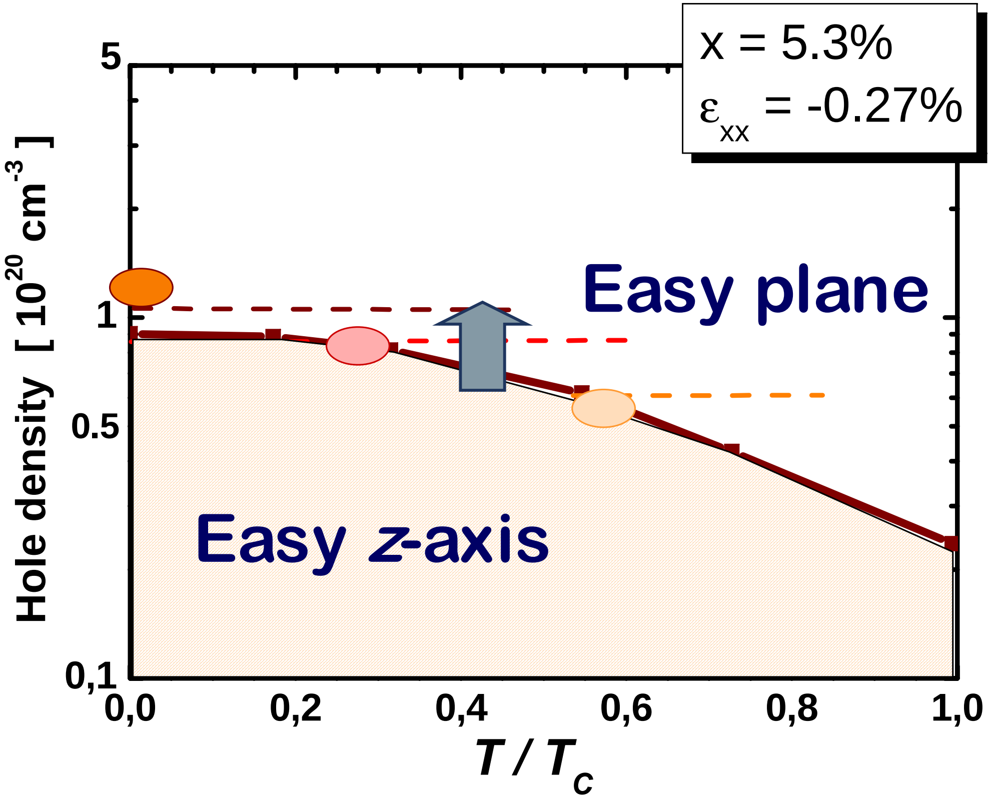
Annealing influence on magnetic anisotropy/ reorientation transition } **REM(T) needed**

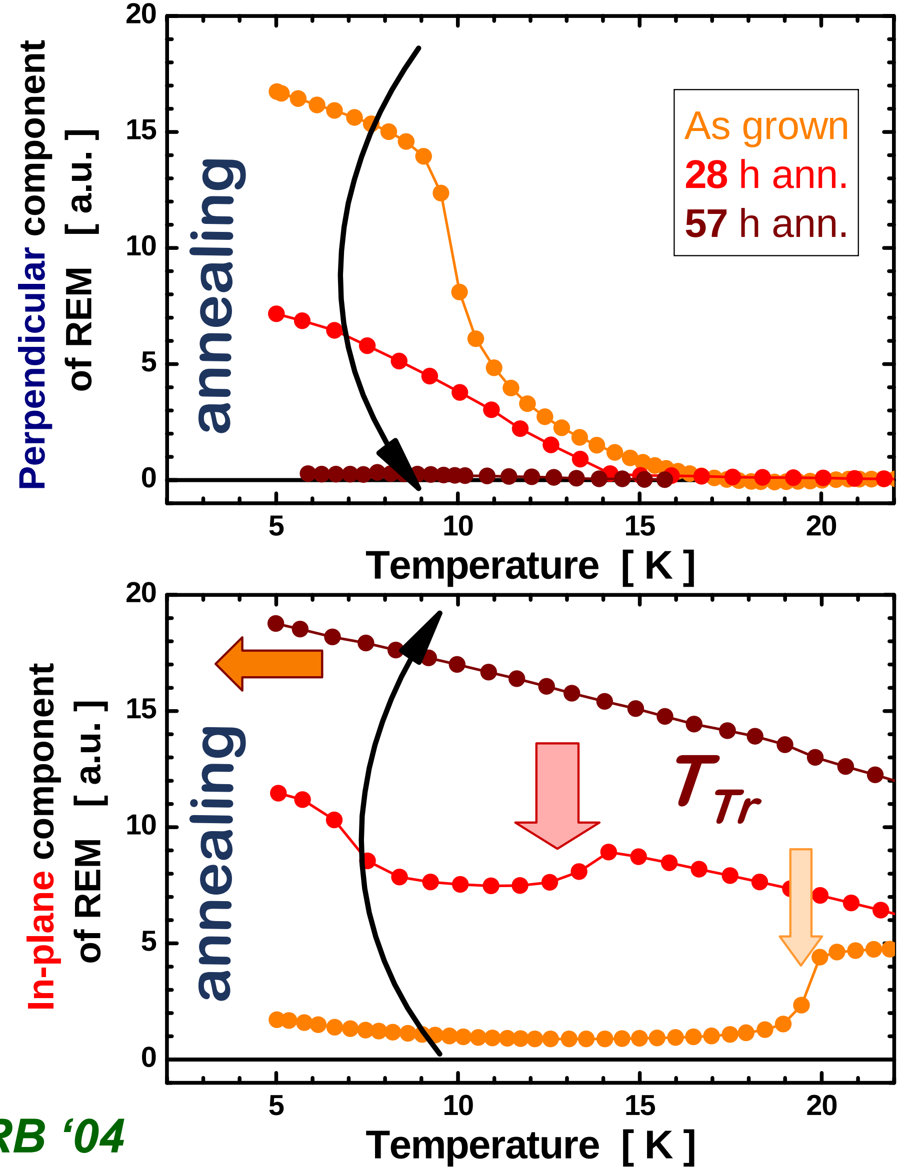


Hole density change: LT annealing

Post growth LT annealing increases hole density

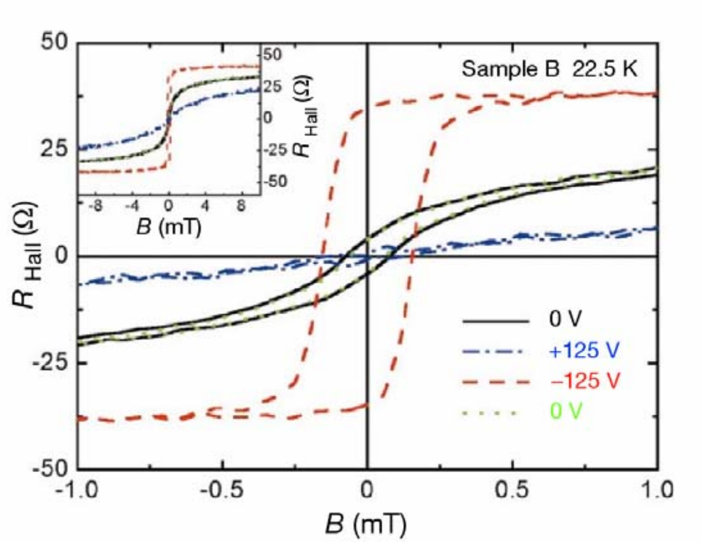
Annealing influence on magnetic anisotropy/ reorientation transition



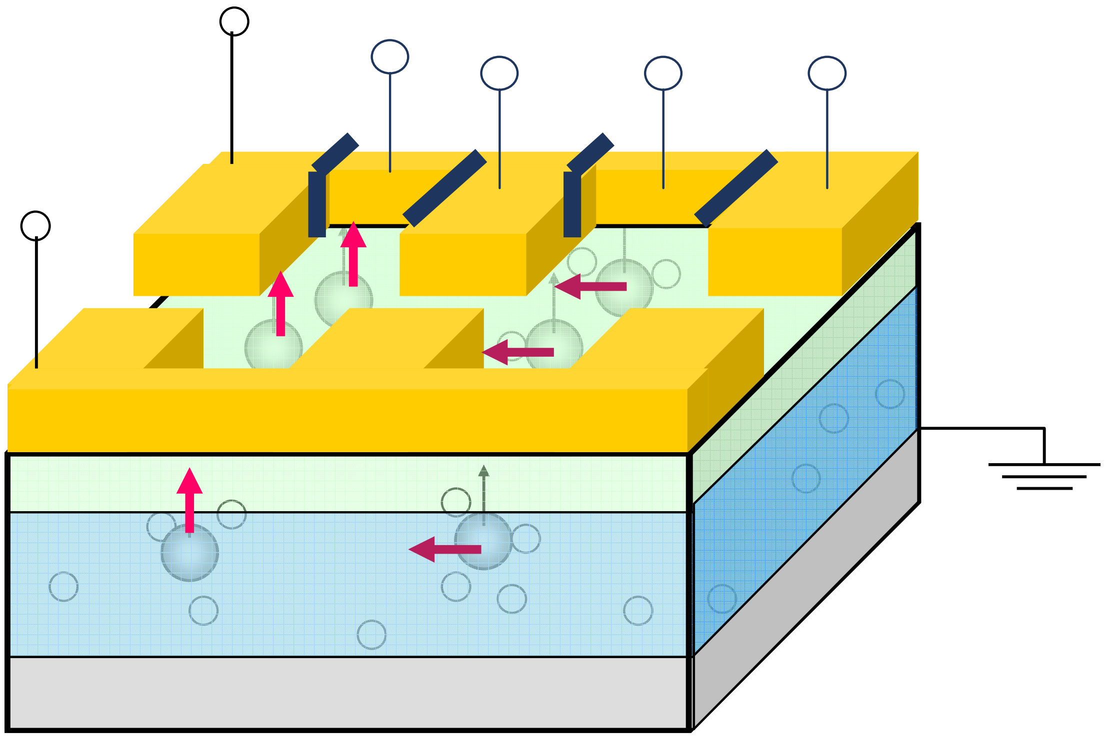


M. Sawicki, et al., PRB '04

Control of the magnetism in nano scale

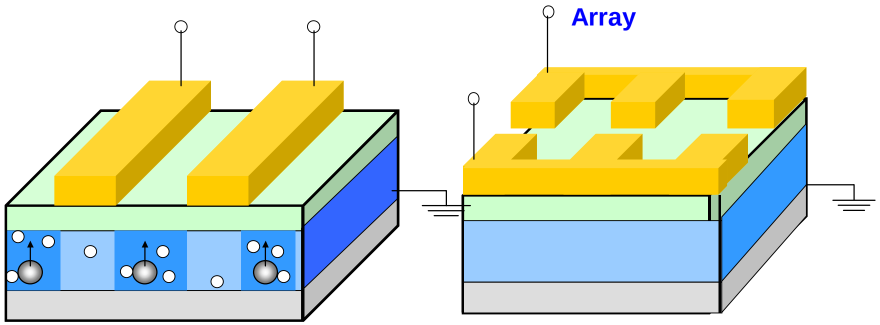


Controlling quantum magnetic dots



Patterning magnetic nanostructures

Ferromagnetic Quantum Wires **Ferromagnetic Quantum Dot Array**



....summary

- Confinement and Strain induced magnetocrystalline anisotropy observed.

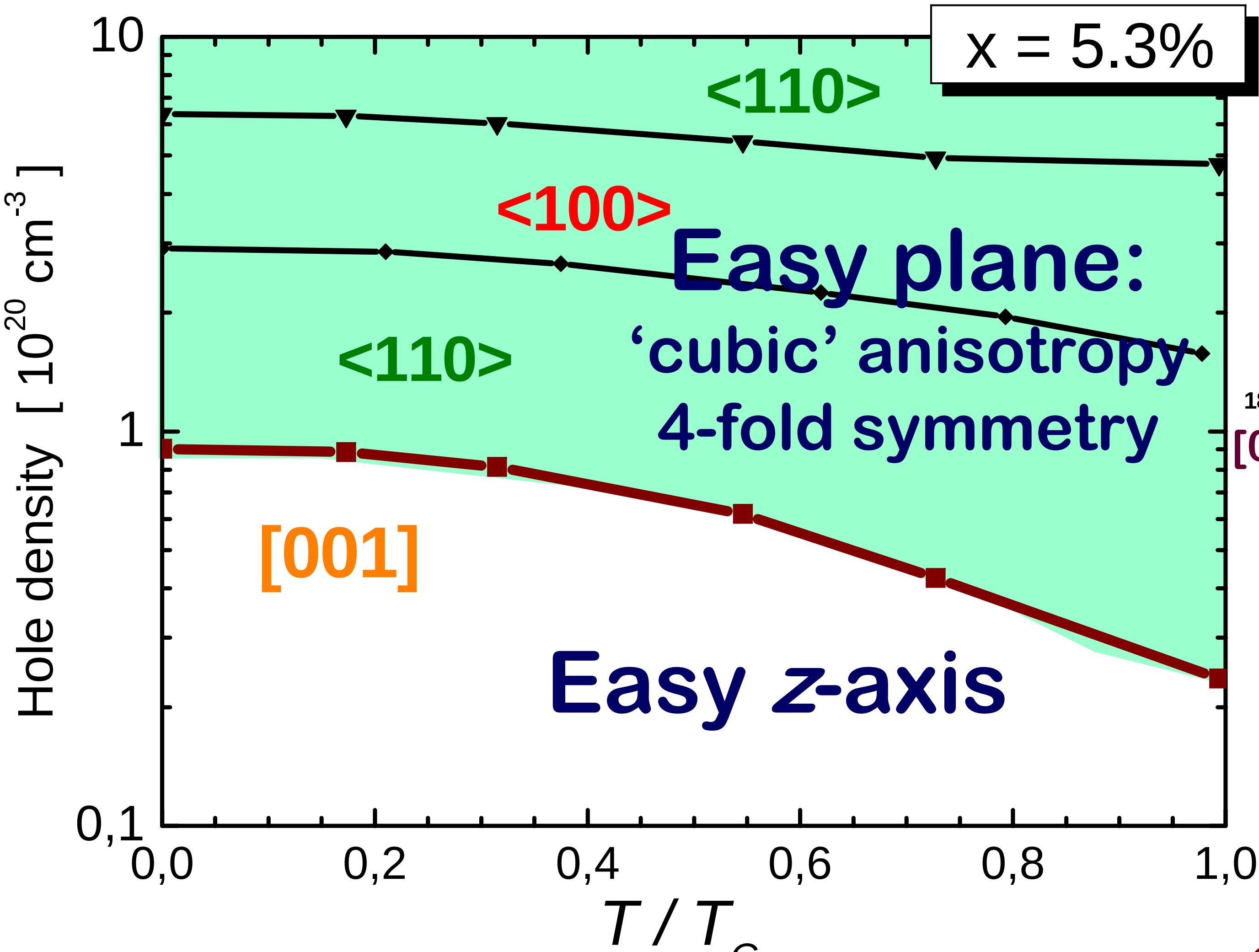
  - character
  - magnitude
  - reorientation transition

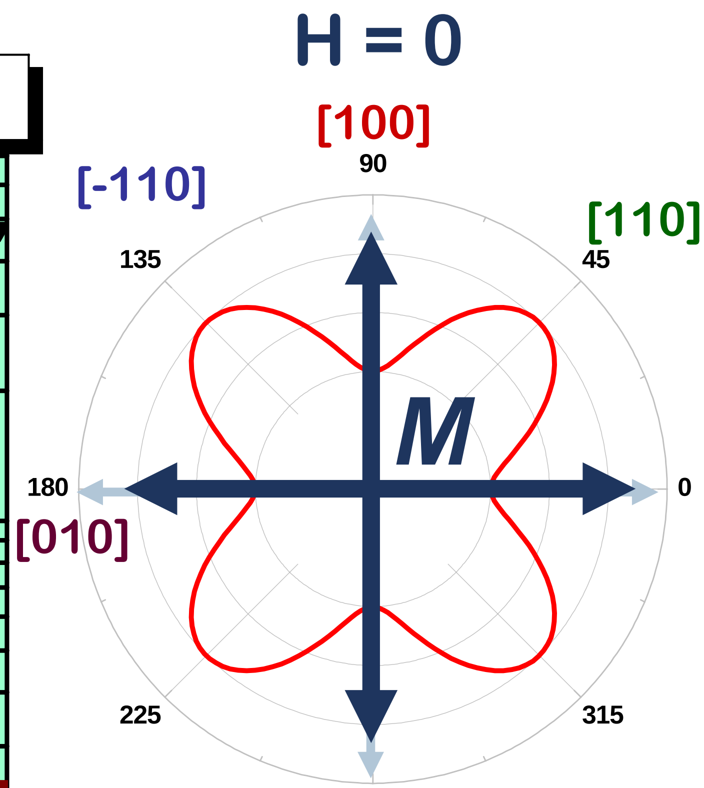
- consistent with p-d Zener model

The *in-plane* magnetic anisotropy

The epitaxially induced D_{2d} symmetry suggests 4-fold (biaxial) magnetic in-plane anisotropy

4-fold in-plane magnetic anisotropy

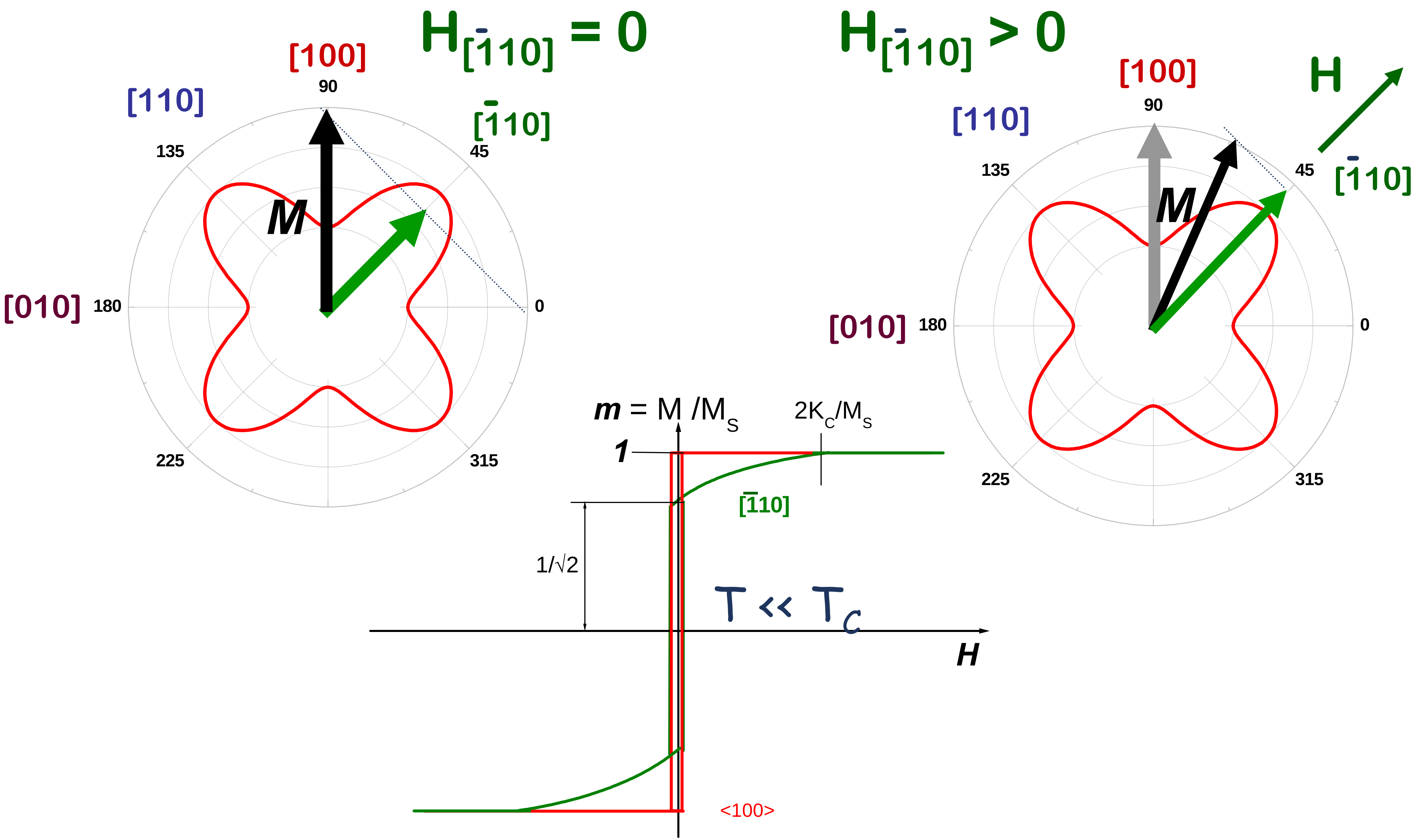




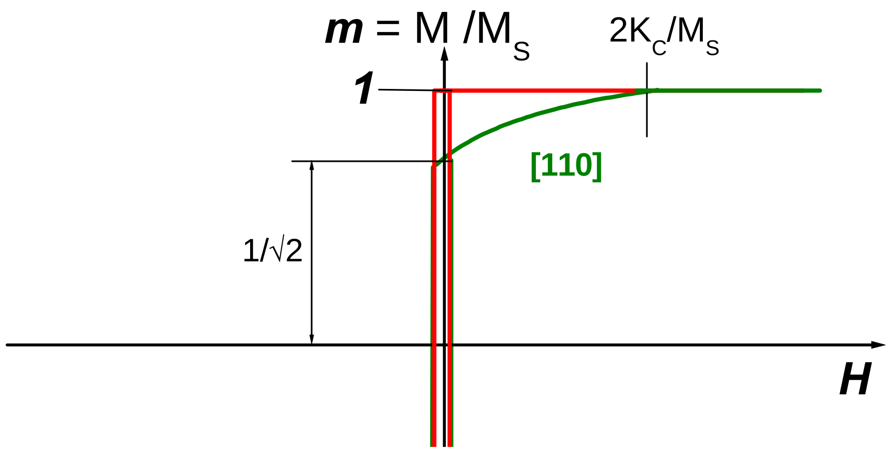
Can we observe this?

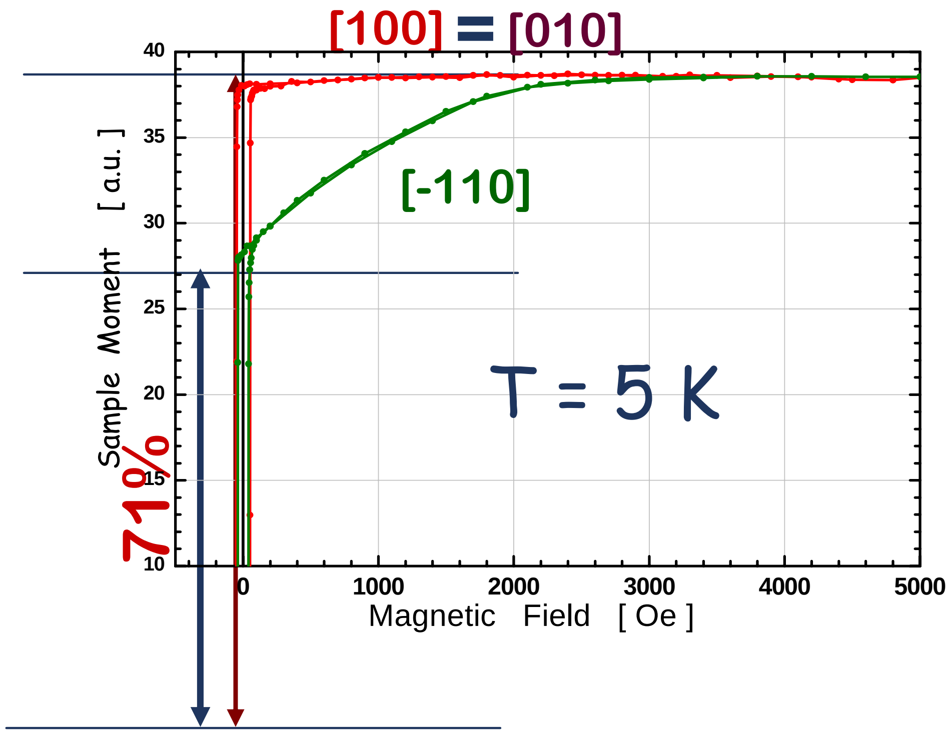
Calculations: Dietl, Ohno, Matsukura, PRB 2001

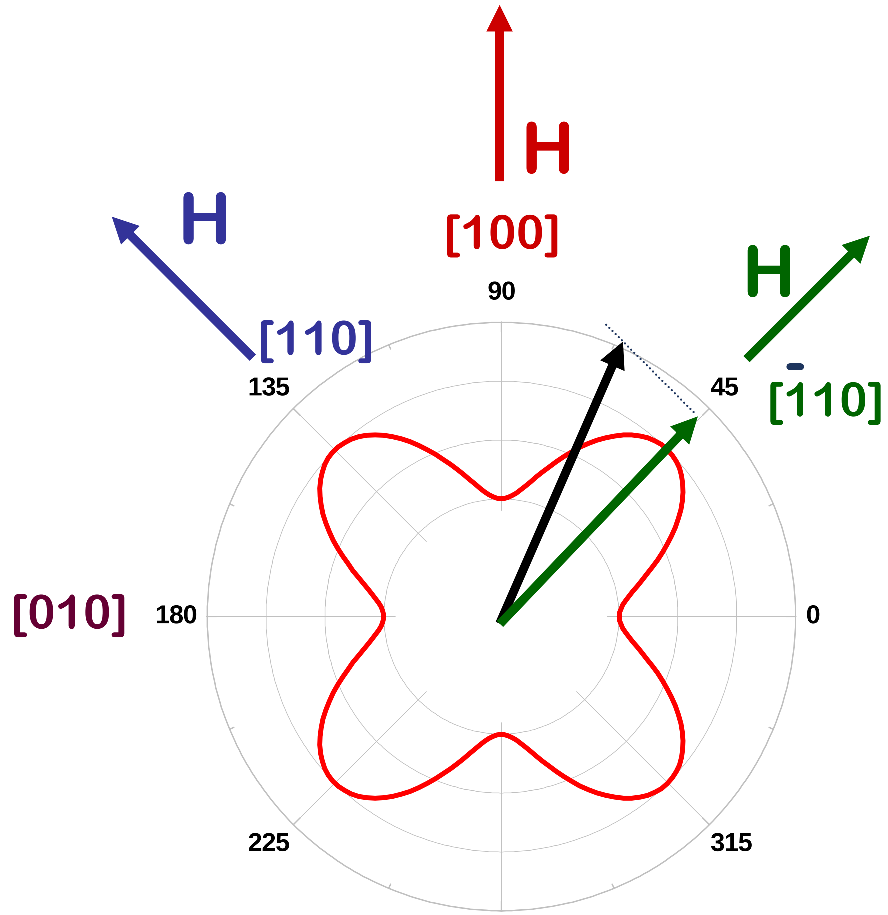
Field induced coherent rotation



Field induced coherent rotation: low T



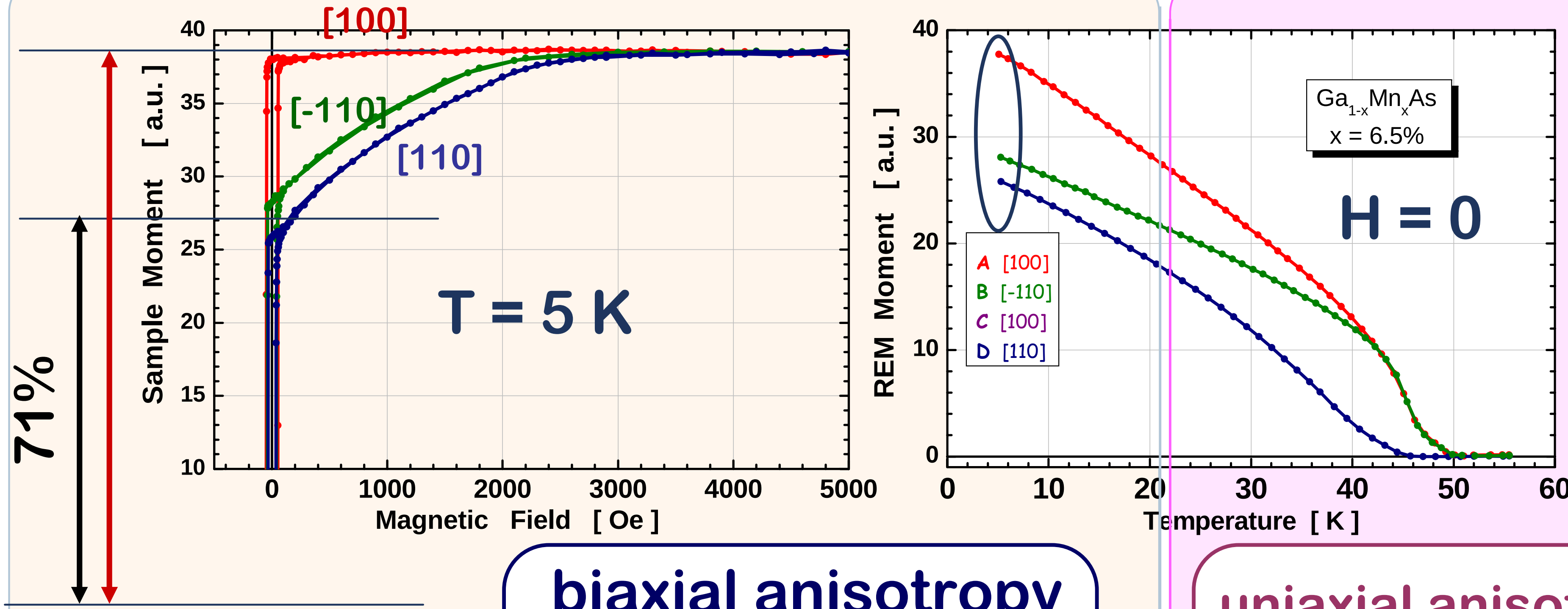




A proof of:

- Formation of macroscopically large domains
- 4-fold magnetic symmetry

Temperature dependence of in-plane magnetic anisotropy



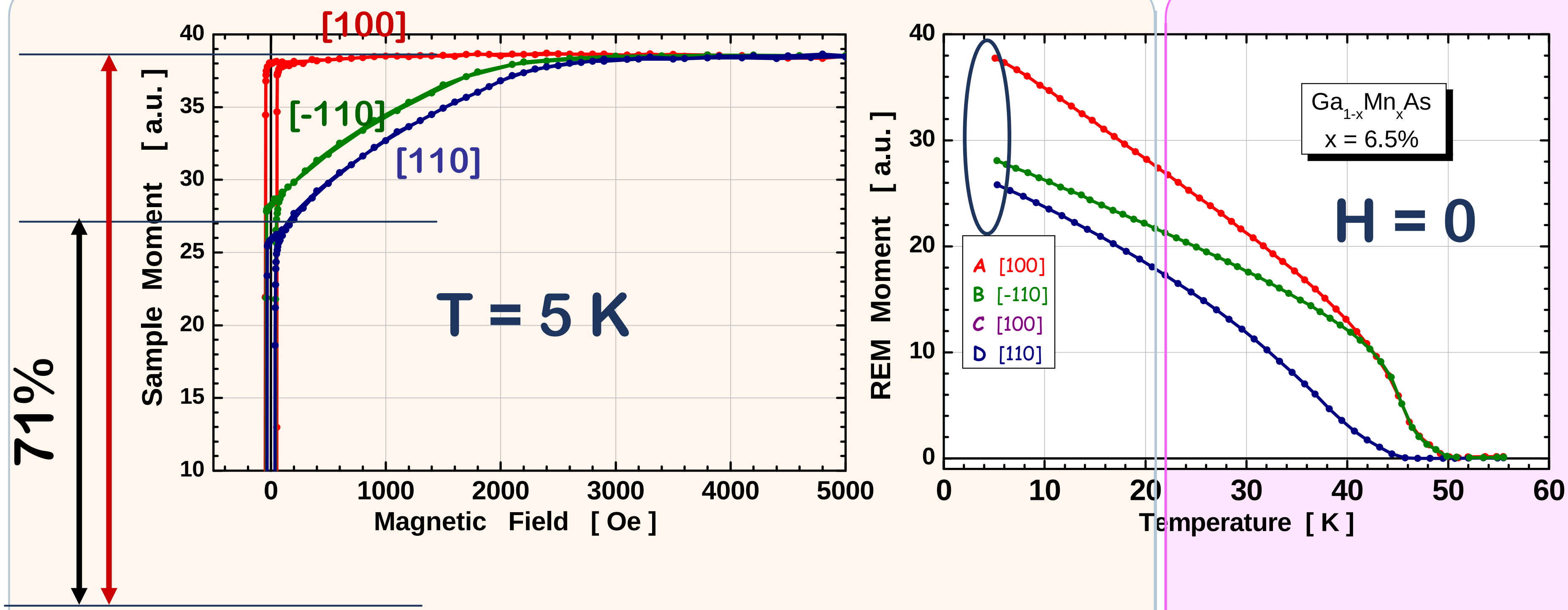
biaxial anisotropy
4-fold symmetry
winning at low *T*

uniaxial anisotropy
2-fold symmetry
winning at high *T*

cf. Katsumoto et al., Hrabovsky et al., Tang et al., Welp et al., Ferre et al., Liu et al.,...,
& EVERYONE ELSE.

The new reorientation transition when system crosses from biaxial to uniaxial anisotropy dominating temperature range

Temperature dependence of in-plane magnetic anisotropy

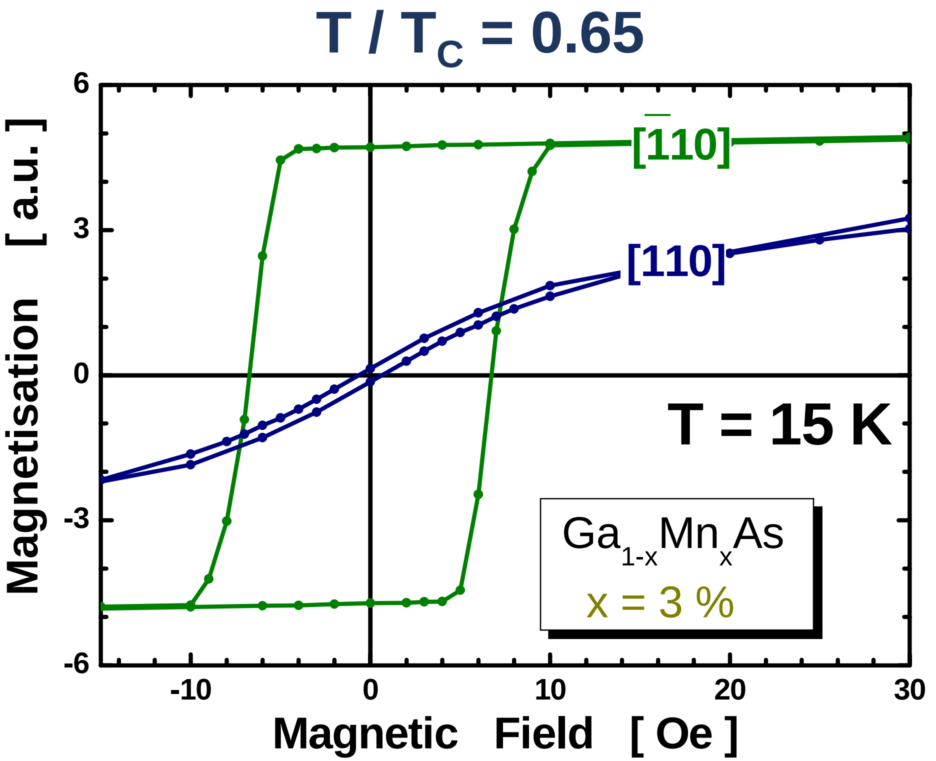


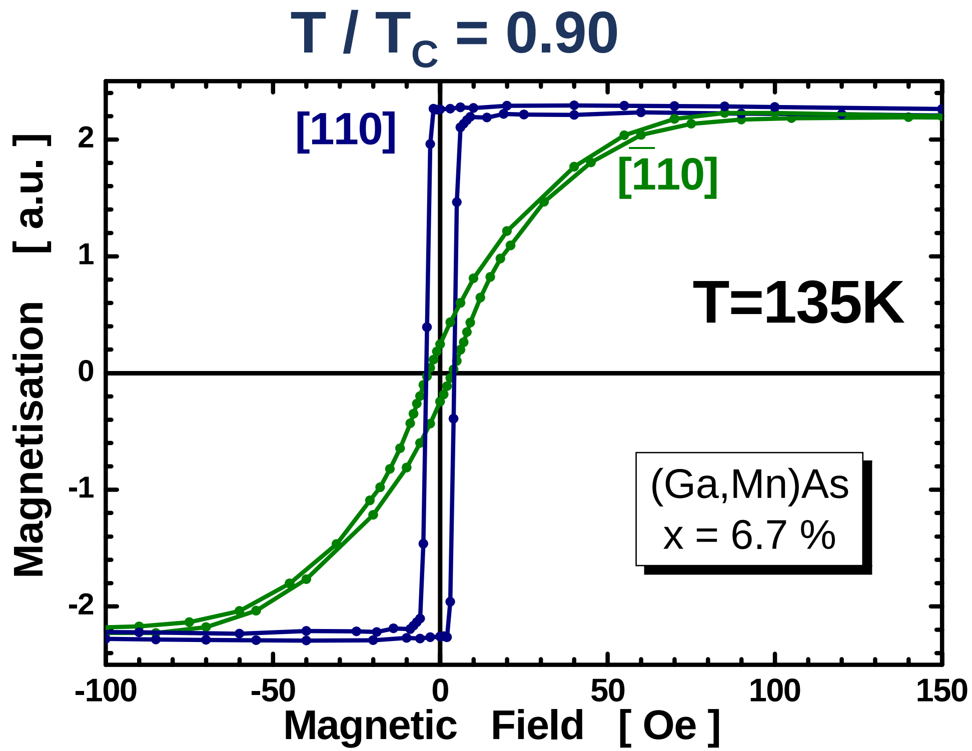
As grown samples: **Uni_easy [-110]**

Cubic_easy <100> **Uni_hard [110]**

In-plane uniaxial magnetic anisotropy

Strong uniaxial behaviour with either [-110] or [110] the easy axis, seen on all studied samples, *usually* dominating close to T_C





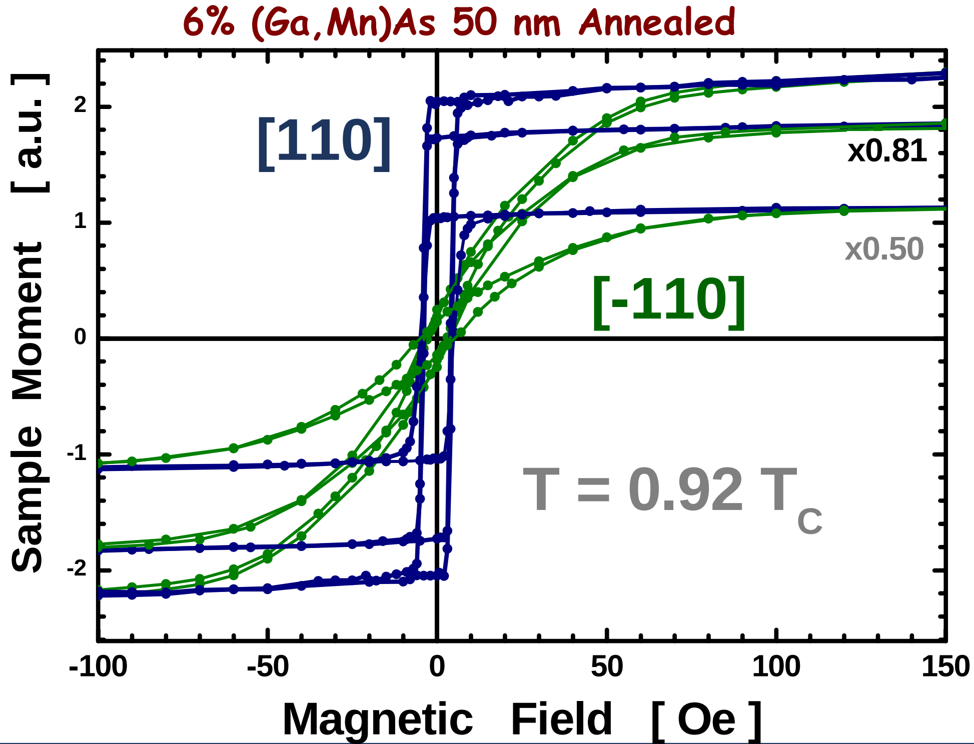
(near perfect single domain behaviour!!)

M. Sawicki, et al.,, PRB '05

In-plane uniaxial magnetic anisotropy

Precluded by symmetry considerations. **Not expected in D_{2d}.**

- Thickness independent: seen from 7 μm down to 5 nm
  Welp et al., '04 *Nottingham, '04*
- Not sensitive to etching



Before: **50nm**
2x etch
4x etch: **25nm**

Not sensitive to the state of the surface; surface/interface anisotropy not important

In-plane uniaxial magnetic anisotropy

$D_{2d} \rightarrow C_{2v}$ **symmetry lowering:**

(In C_{2v} [110] and [-110] are not equivalent)

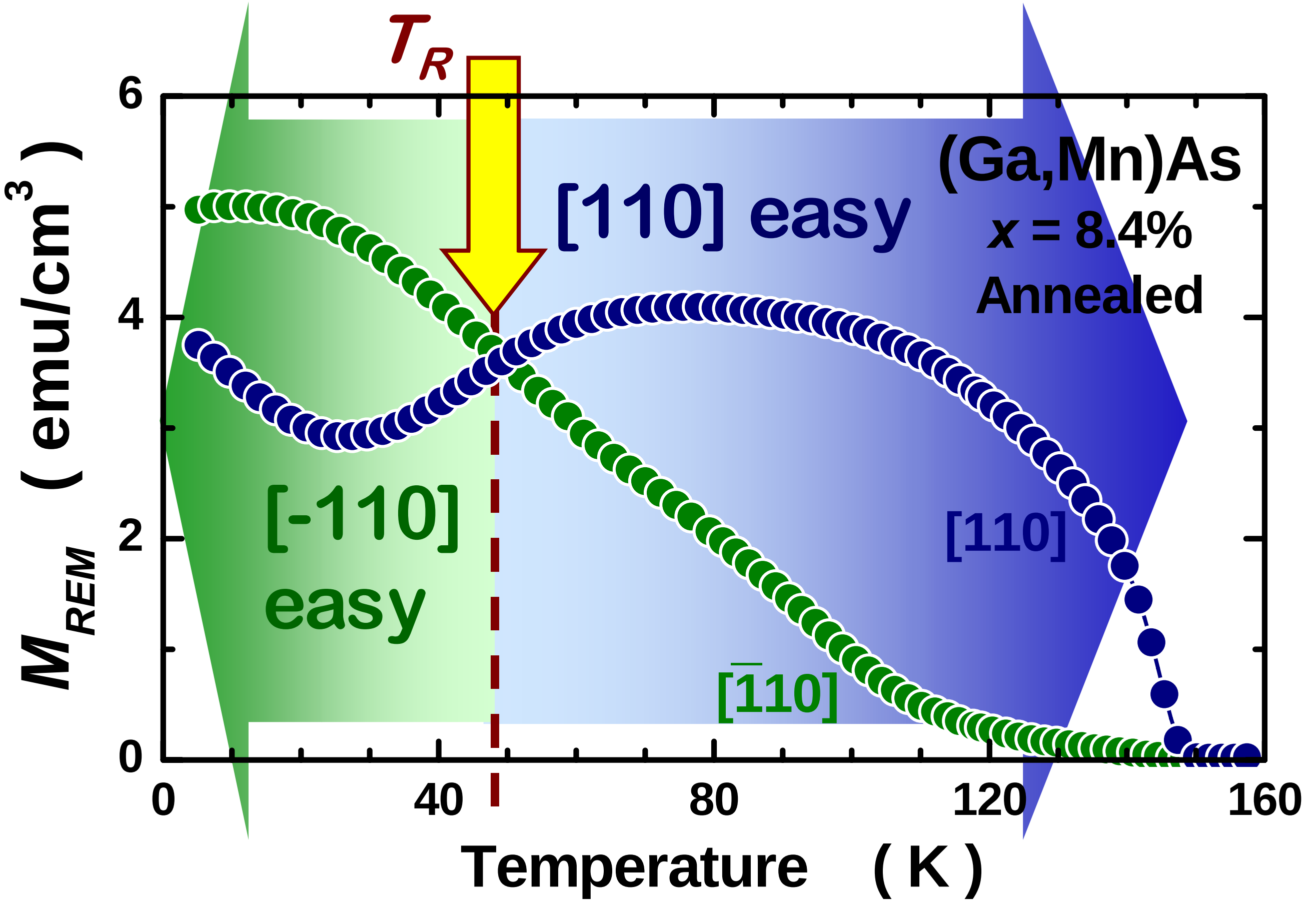
- Mn concentration gradient along growth axis *Sadowski et al., 2004*

- preferential incorporation of Mn during growth *Welp et al., 2004*

More information required.....

In-plane uniaxial magnetic anisotropy

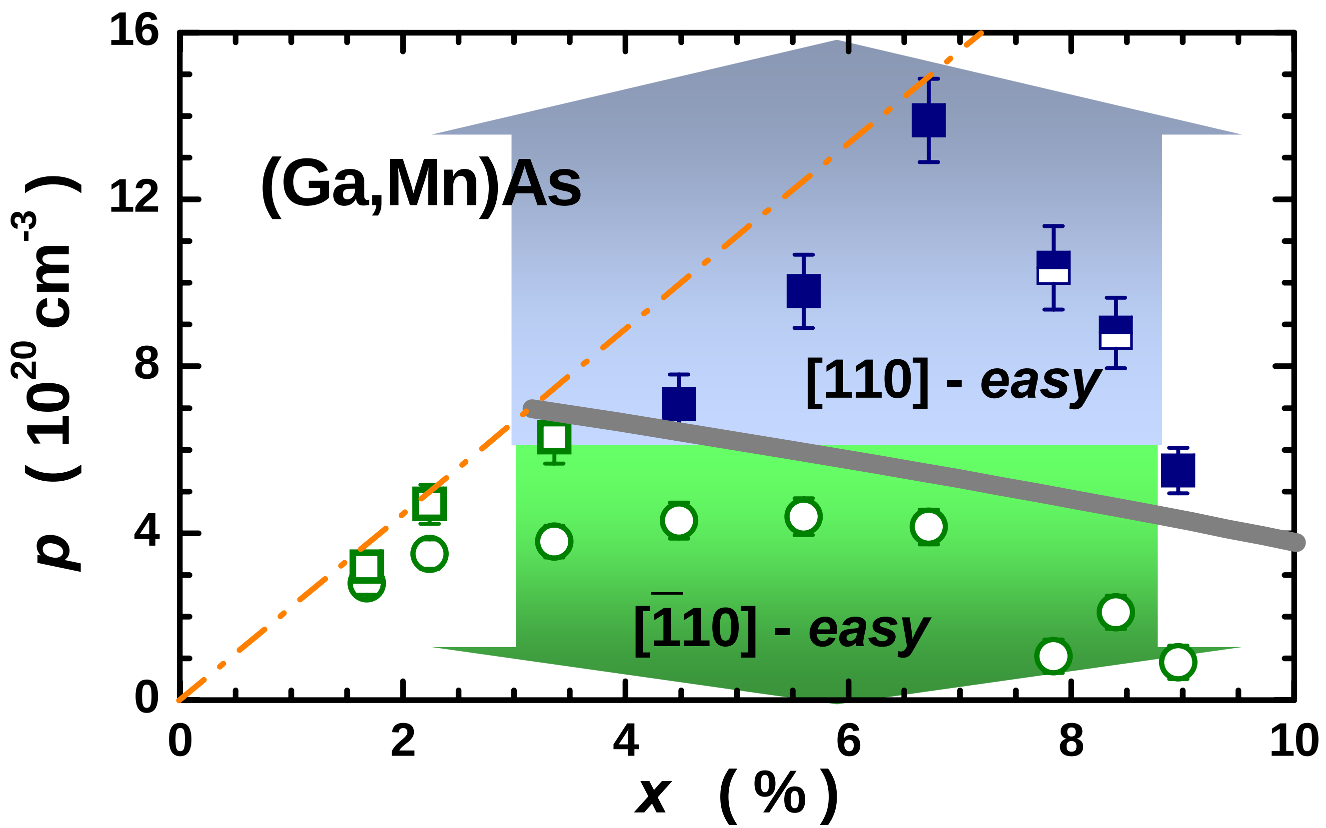
- There are samples with the easy axis switching from [-110] to [110] on increasing T



M. Sawicki, et al., PRB'05

In-plane uniaxial magnetic anisotropy

- There are samples with the uniaxial easy axis switching from [-110] to [110] on increasing T;
- It switches also upon annealing if $p \gtrsim 6\times10^{20}$ cm^{-3}

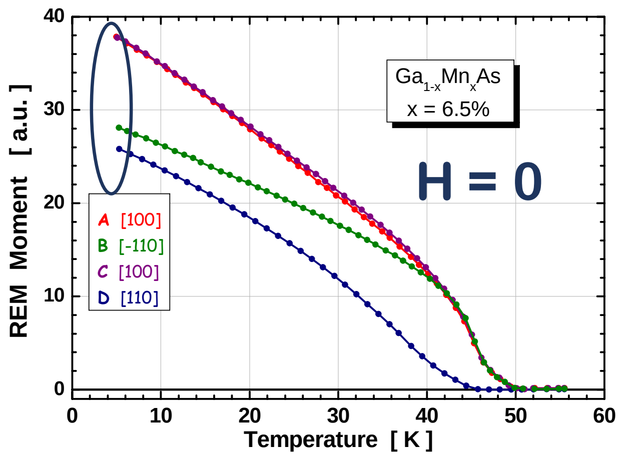


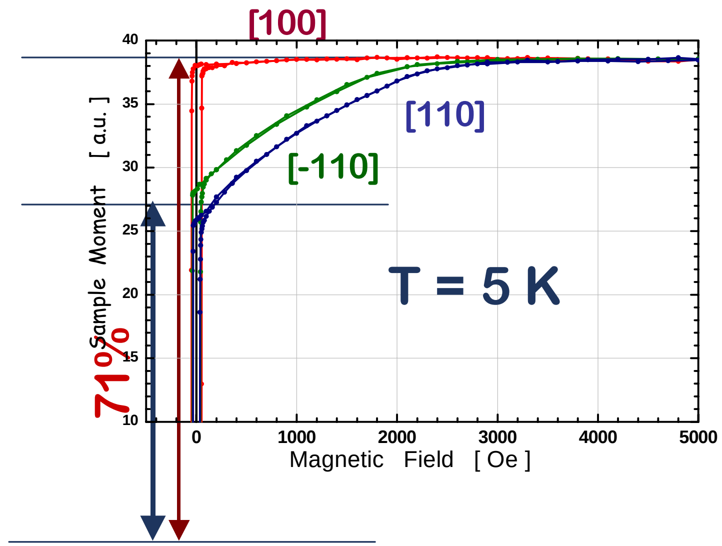
M. Sawicki, et al., PRB'05

$M(T)$ in presence of two competing in-plane anisotropies: single domain case

$$E_m = -K_C/4\, \sin^4(2\theta) \;+\; K_U \sin^2\theta - MH\cos(\varphi - \theta)$$

$K_C \sim M_S^4 \Leftarrow$ *expected* $\Rightarrow K_U \sim M_S^2$





Two 'competing' terms

⇓

Magnetic easy axis reorientation transition

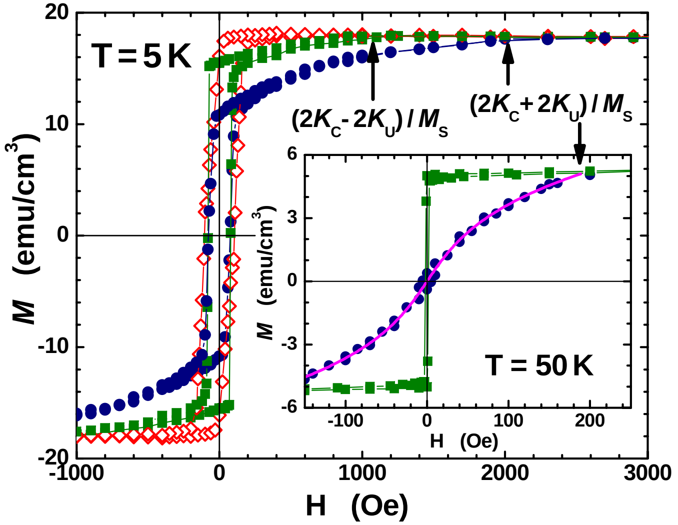
when $K_C = K_U$

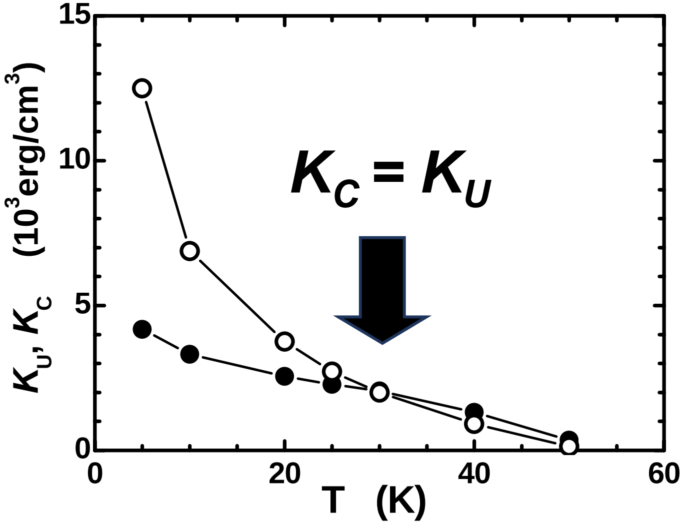
K. Wang, et al., cond-mat '05

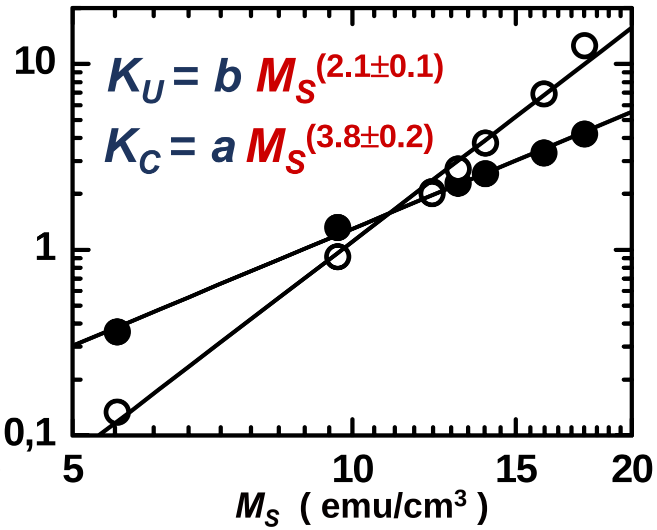
Phenomenological description of magnetic anisotropy in single domain (Ga,Mn)As

$$E_m = -K_C/4\,\sin^4(2\theta) + K_U \sin^2\theta - MH\cos(\varphi - \theta)$$

$K_C \sim M_S^4 \Leftarrow$ *expected* $\Rightarrow K_U \sim M_S^2$ $\quad M_{[-110]}^2 + M_{[110]}^2 = M_S^2$







K. Wang, et al., cond-mat '05

Conclusions

Magnetic anisotropy in hole-controlled ferro-DMS:

- ***magnetic anisotropy – effect of s-o interaction in the valence band***
- ***z-axis (perpendicular)/in plane anisotropies controlled by confinement and epitaxial strain***
- ***in-plane anisotropy: competition of biaxial(cubic) and uniaxial anisotropy – origin not yet understood***
- ***three Spin Reorientation Transitions observed:***
  - ***perpendicular ⇔ in plane***
  - ***<100> ⇔ [-110]***
  - ***[-110] ⇔ [110]***

  possibility of easier magnetisation manipulation
- ***phenomenological self-consistent description possible in single domain model***