

Spintronics and diluted magnetic semiconductors

DETI.2 workshop 2012

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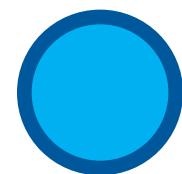
Outline

1. Principles of spintronics
2. Paramagnetic DMS, giant Zeeman splitting and bound magnetic polarons
3. Magnetic coupling
4. Half metallic materials
5. Magnetism and Transport in GaMnAs
6. Oxide based DMS by ion implantation

Origin of magnetism



Electron charge + movement
→ Magnetic moment



Nucleus

Orbital moment

$$\hat{L}^2 Y_l^{m_l} = \hbar^2 l(l+1) Y_l^{m_l}$$

$$\hat{L}_z Y_l^{m_l} = \hbar m_l Y_l^{m_l}$$

Dirac equation (Spin)

$$(c\vec{\gamma} \cdot \hat{p} + mc^2)\psi = i\hbar\gamma^0 \frac{\partial}{\partial t}\psi$$

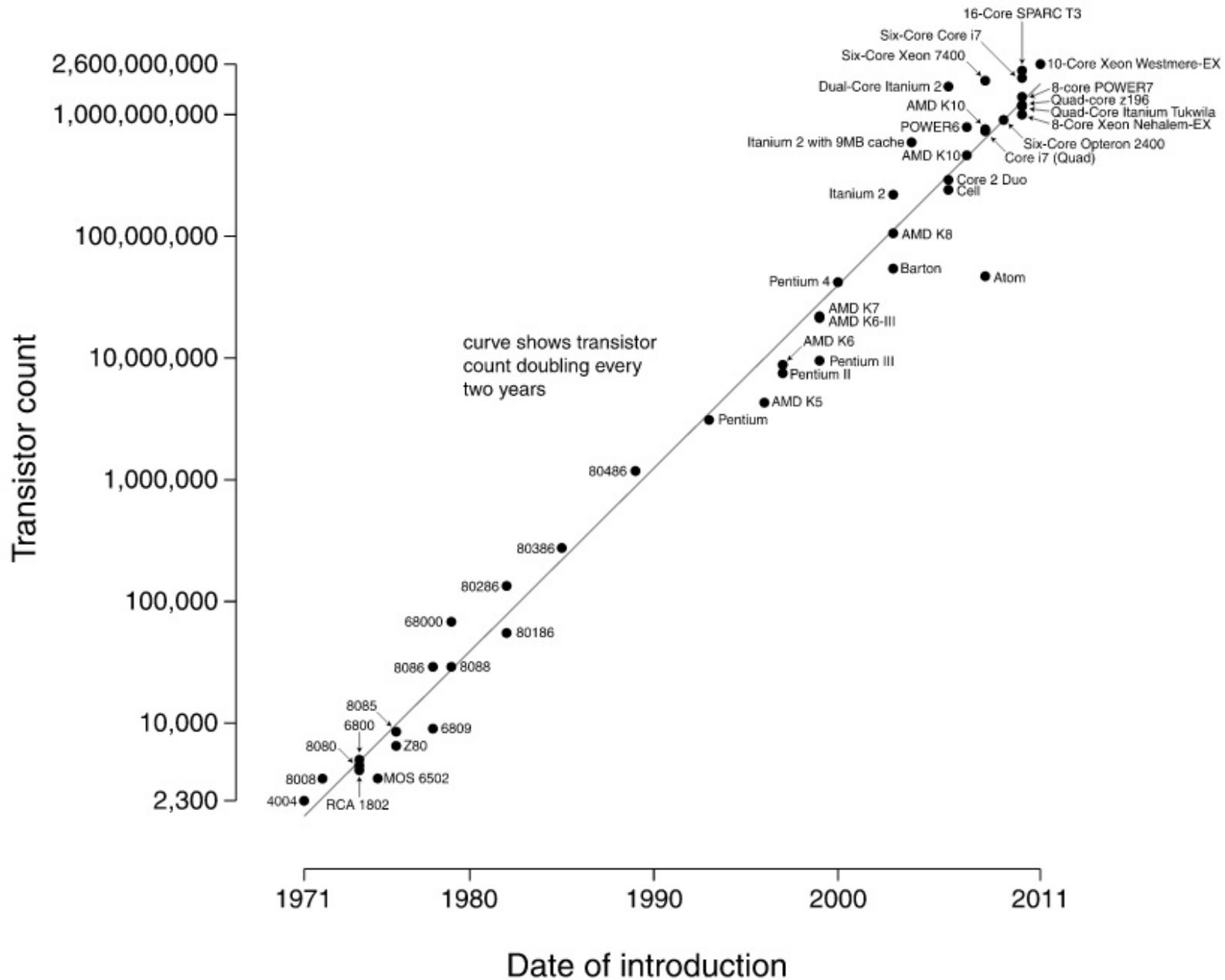
$$\hat{S}^2 Y_s^{m_s} = \hbar^2 s(s+1) Y_s^{m_s}$$

$$\hat{S}_z Y_s^{m_s} = \hbar m_s Y_s^{m_s}$$

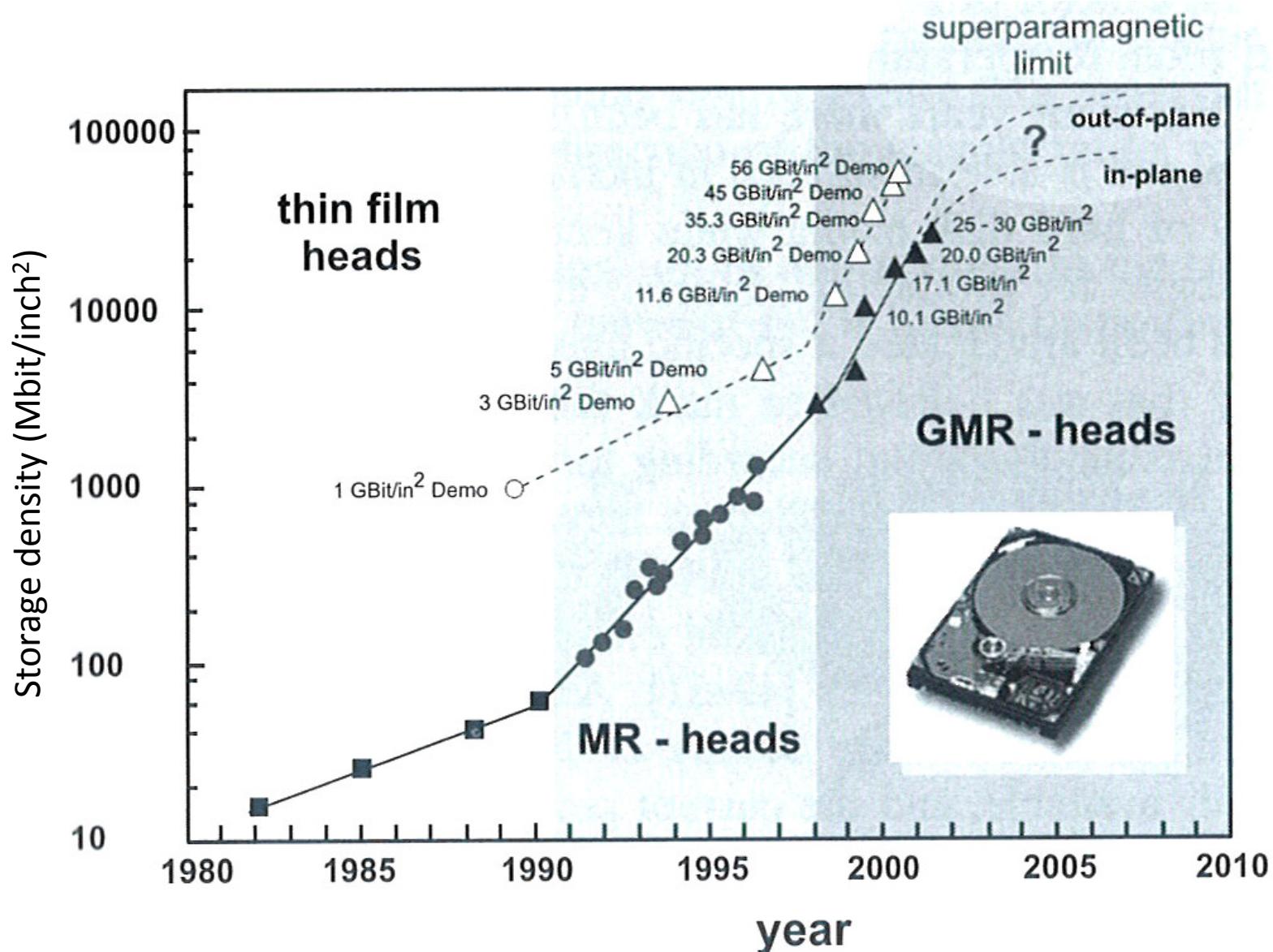




Microprocessor Transistor Counts 1971-2011 & Moore's Law



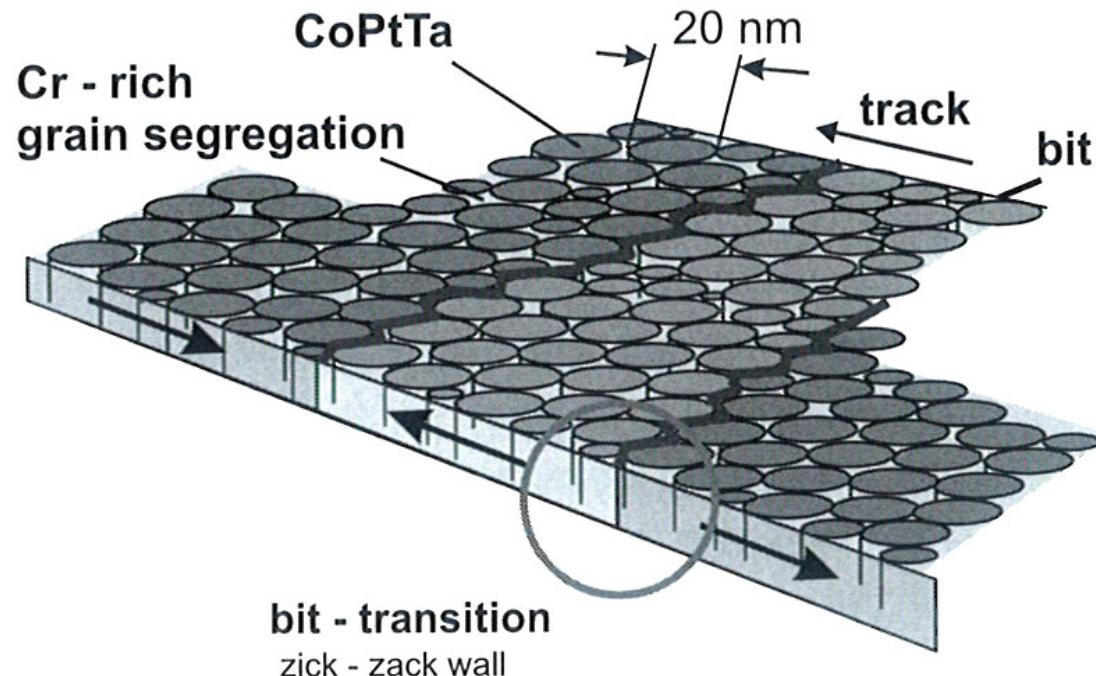
Moores law on data storage



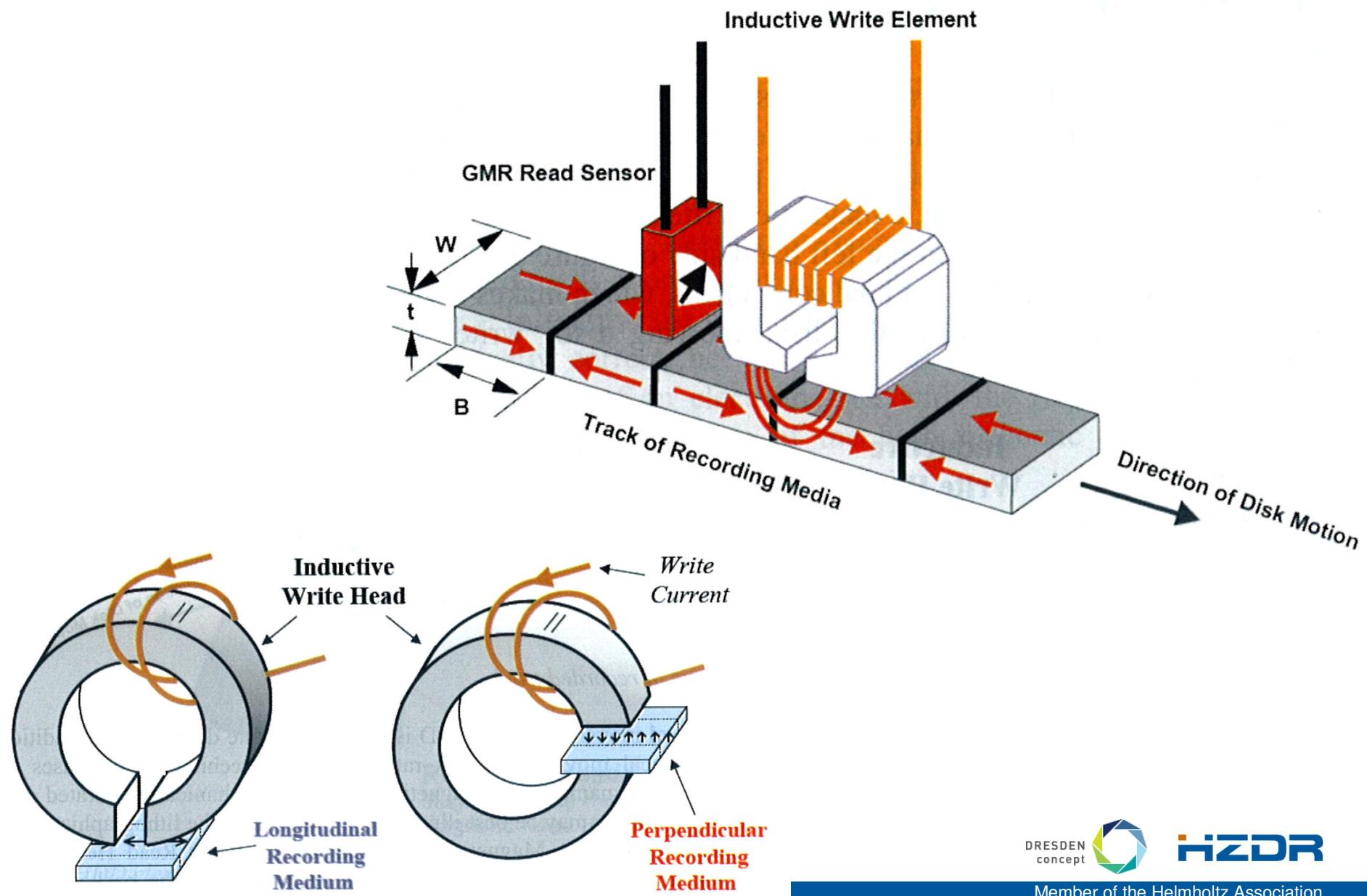
Traditional magnetic data storage

Technology NSIC:

1. Deposition of $\text{Co}_{70}\text{Pt}_8\text{Cr}_{22}$ + Ta on Cr/NiAl substrates
2. Annealing for phase separation between Cr-matrix and Co-Pt-Ta clusters
3. Result: Co-rich ferromagnetic clusters (10-15 nm) for data storage



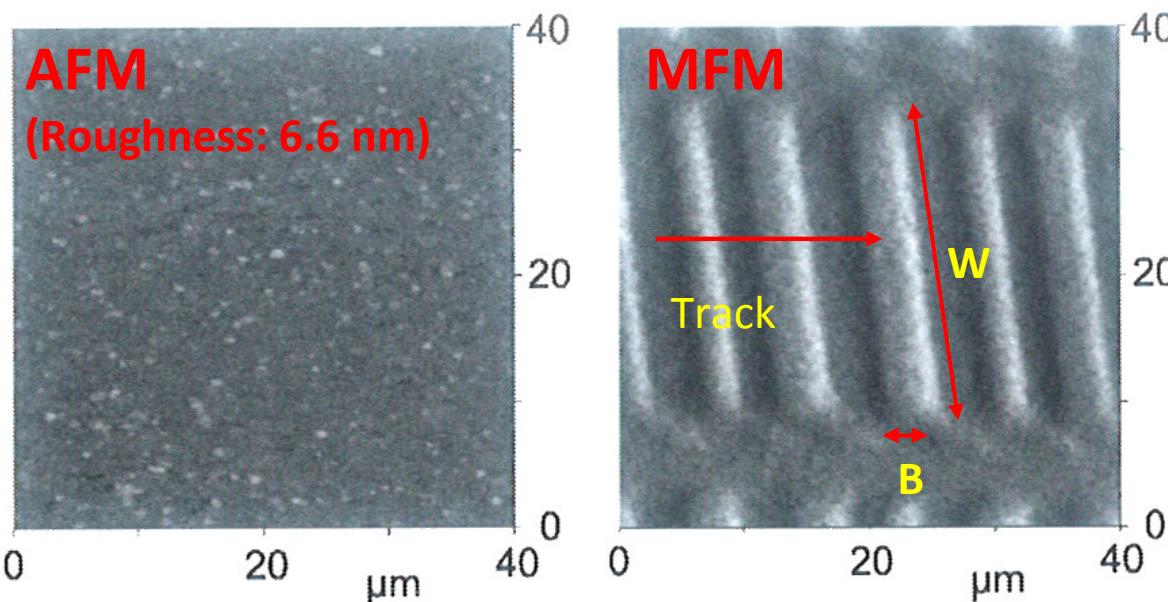
Traditional magnetic data storage



Stability

- Problems:

- Intergranular Exchange interaction good for perfect common alignment, but no Sharp switching at the borders
- No interaction → sharp switching
- Smaller clusters → Instability with respect to stray field and thermal instability (Superparamagnetic effect)
- Current criterion for stability: 10 years

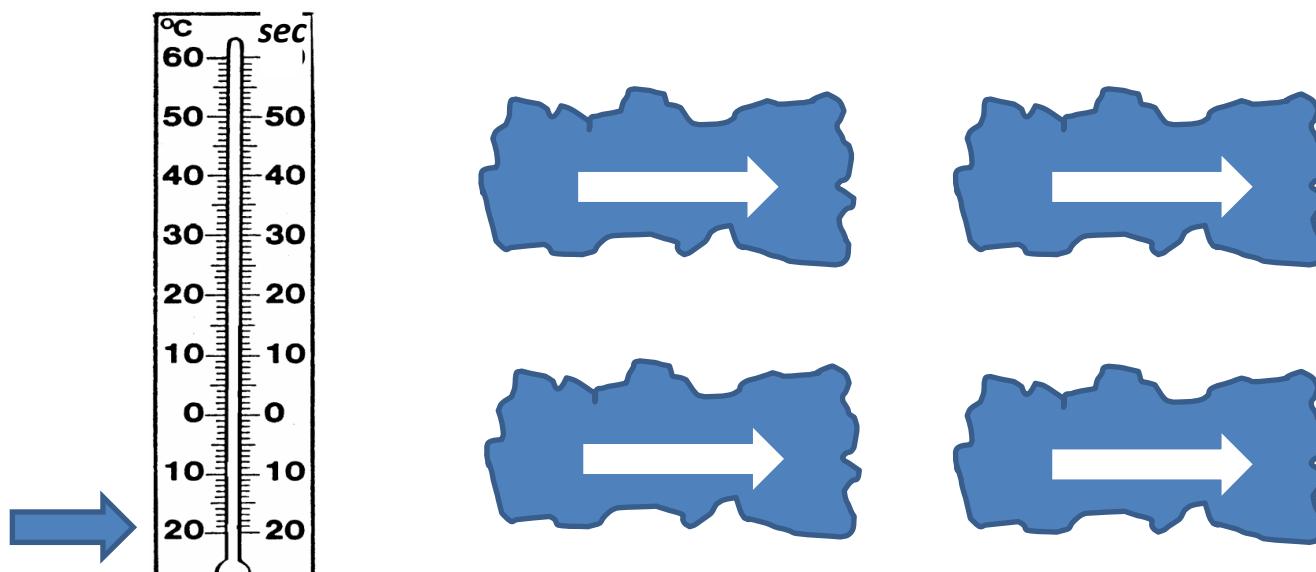
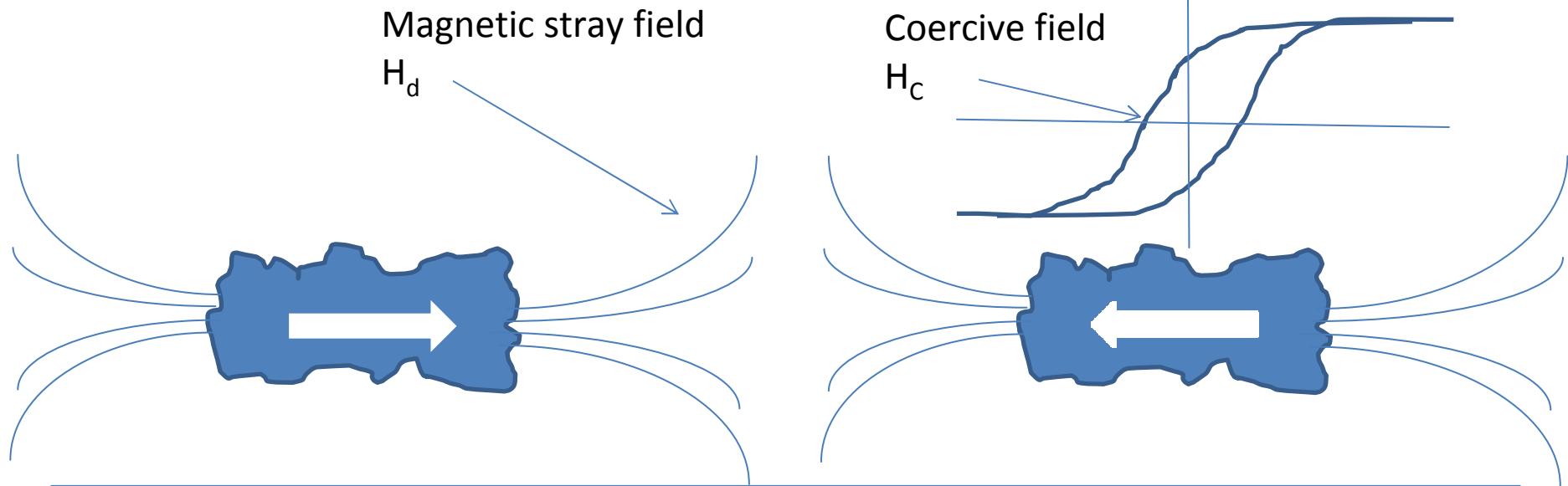


Ideal parameters for 20 Gbit/inch²

- Bitsize: 40x500 nm
- Density: 490 kbpi
- Remanence x film thickness:
 $M_r t = 0.4 \text{ memu/cm}^2, t \sim 12-15 \text{ nm}$



Stability



Stability

- Stray fields of neighbouring clusters

Coercivity H_c vs. Demagnetisation field H_d

$$H_c > H_d = \frac{8M_r t}{W} \left[\sqrt{1 + \left(\frac{2W}{B} \right)^2} - 1 \right]$$

Problems:

- Reduction of $H_d \rightarrow$ Lowering of read signal
- Increase of $H_c \rightarrow$ Large stray field at read head necessary (max. 5 kOe)

- Superparamagnetic effect

Magnetocrystalline anisotropy k_1 (spherical cluster):

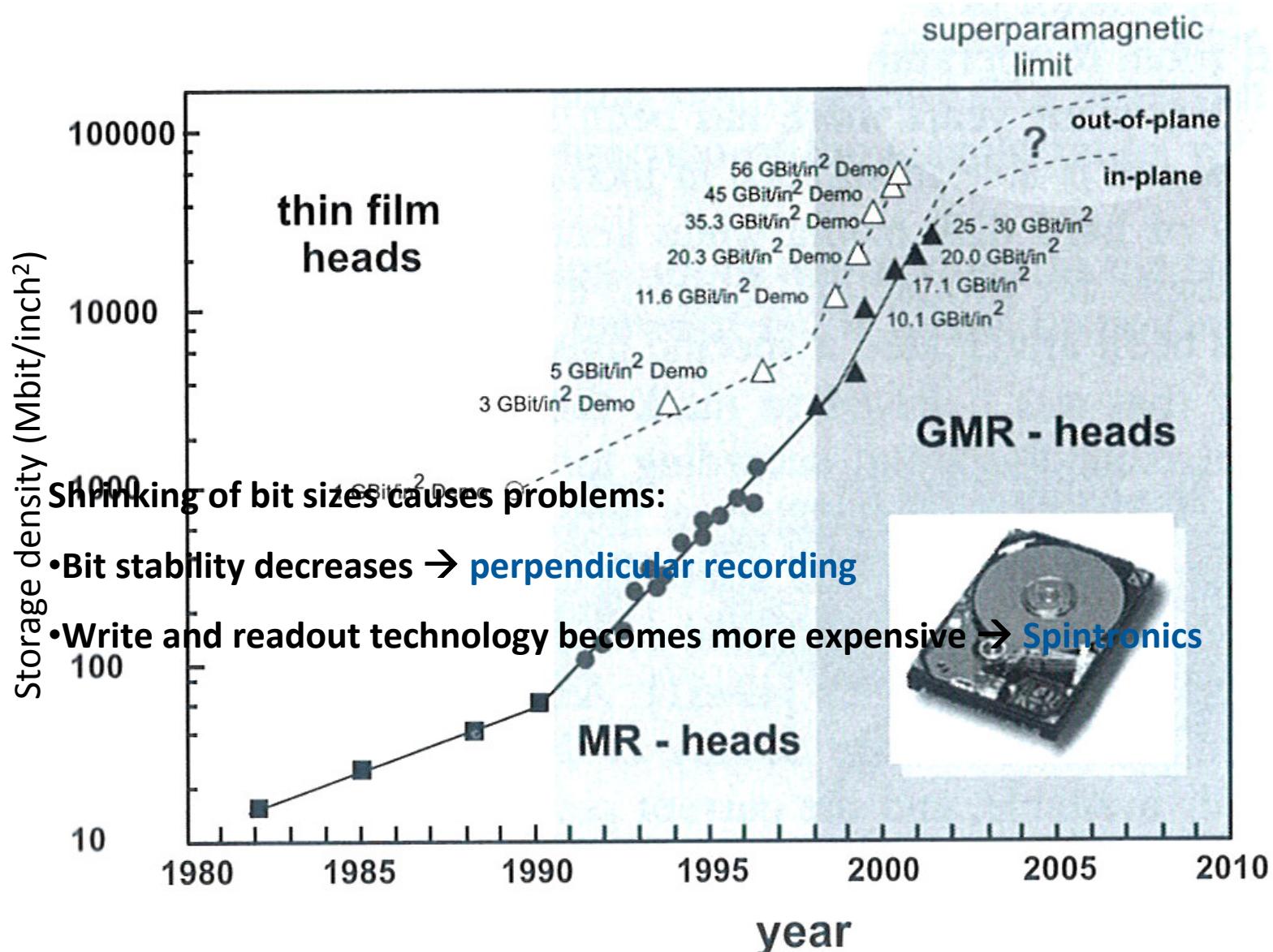
$$k_1 V > \ln \left[\frac{f_0 t}{0.67} \right] \cdot k_B T$$

Frequency Time

Parameters (300 K, 10 years):

- Minimum diameter for Co ($4.5 \times 10^5 \text{ J/m}^2$) = 8 nm
- Min. diameter for CoPt ($5.3 \times 10^6 \text{ J/m}^2$) = 3.5 nm

Moores law on data storage



Spintronics= Data processing and data storage with electron spins!

Ferromagnet

- Magnetisation (attracts other magnets)
- Spin polarised electric current (Spintronics)

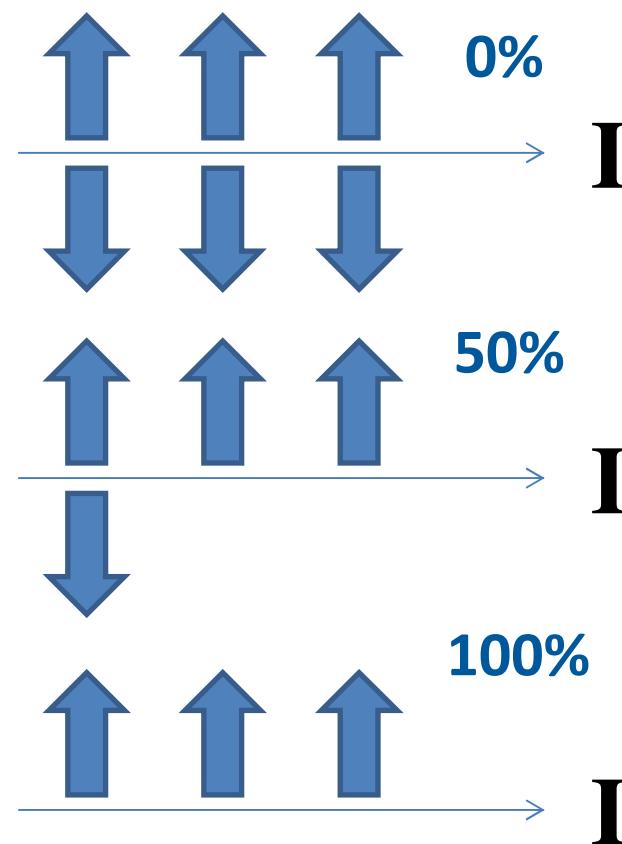




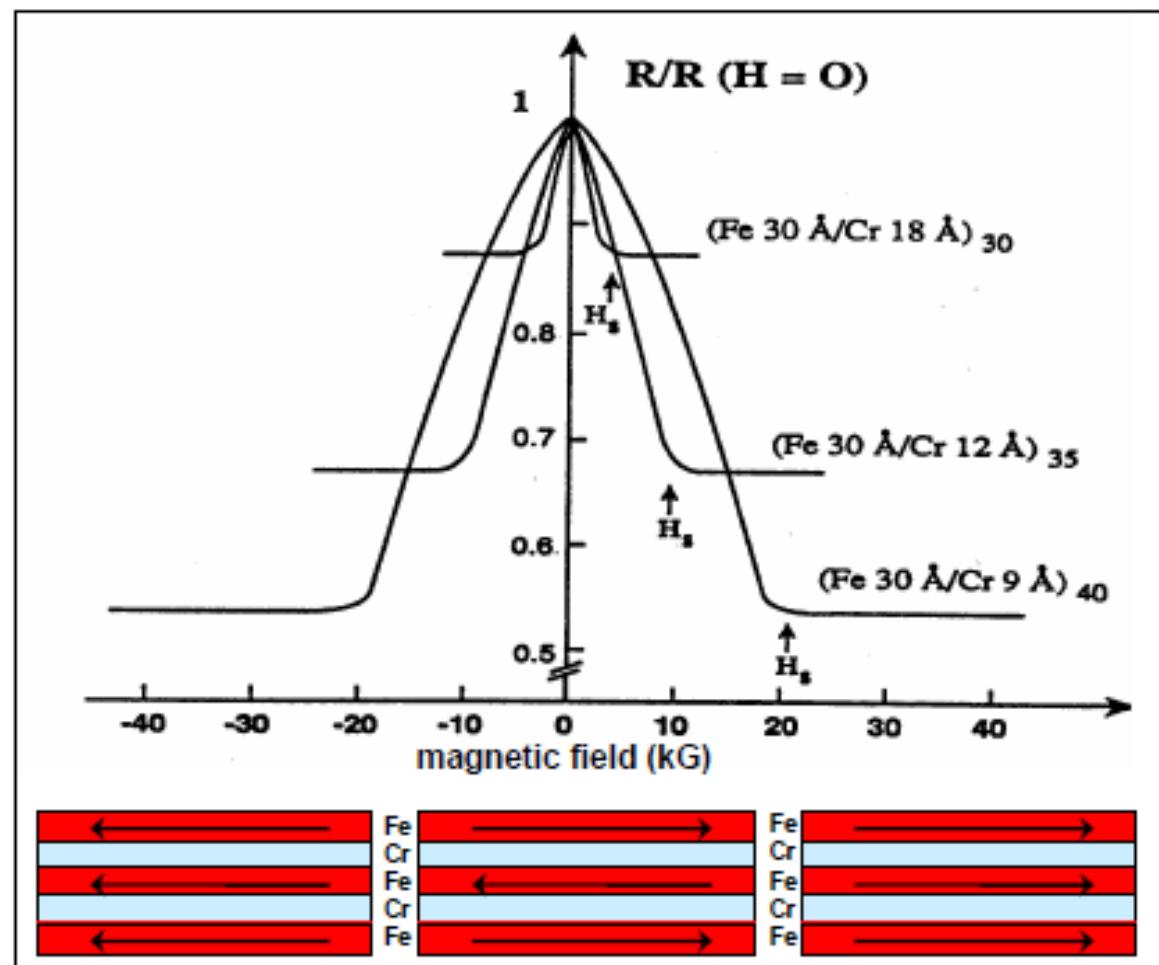
Spintronics

Requirement:

~ 100 % spin polarization
of electric current



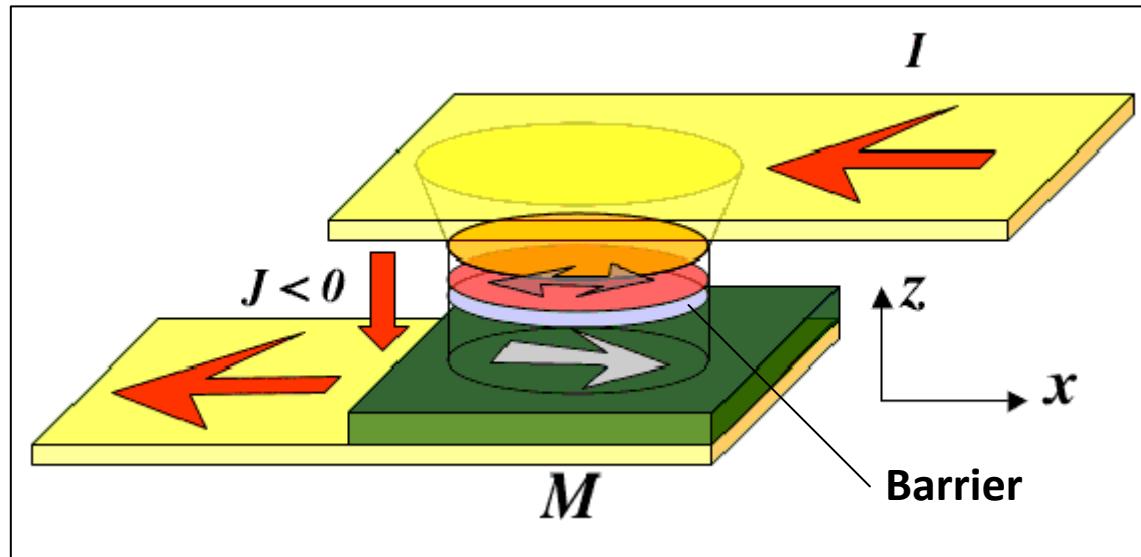
GMR effect (Nobel price 2007)



Spin transfer torque

Experiment: current through magnetic nanopillars

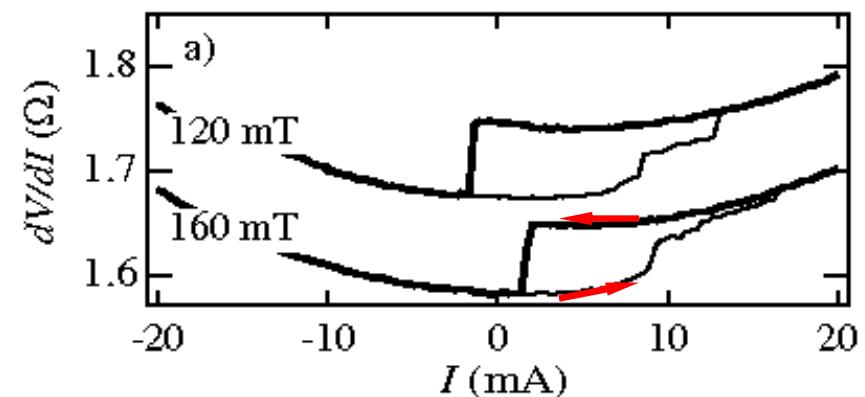
Figure:
courtesy of
J. Miltat



Katine et. al., *Phys. Rev. Lett.*, 84, 3149 (2000):

Main result: hysteresis observed for the differential resistance by the current sweep

Interpretation: magnetization switching caused by a spin-polarized current above a certain threshold





Spin transfer torque

Landau-Lifshitz-Gilbert-equation + STT

$$\frac{\partial \mathbf{m}}{\partial t} = -\gamma \mathbf{m} \times (\mathbf{H}_{\text{eff}} + \mathbf{h}_{\text{th}}) + \alpha \mathbf{m} \times \frac{\partial \mathbf{m}}{\partial t} + \frac{\gamma \hbar J \eta(\mathbf{m}, \mathbf{m}_p)}{2eM_s t_F} \mathbf{m} \times (\mathbf{m} \times \mathbf{m}_p)$$

Critical switching current:

$$J_{c0} = \frac{2e\alpha M_s t_F (H_K \pm H_{\text{ext}} + 2\pi M_s)}{\hbar \eta}$$

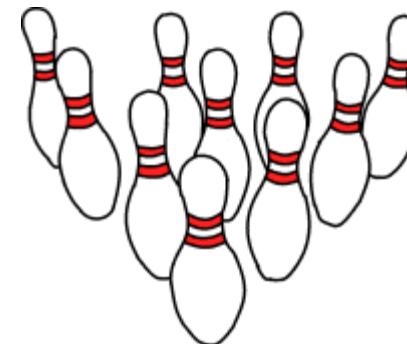
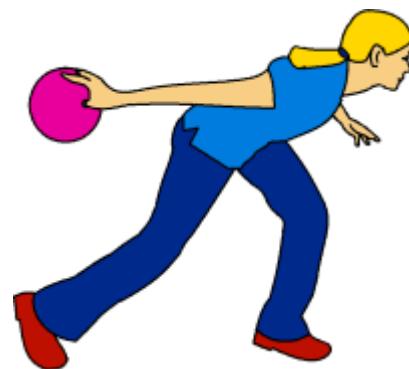
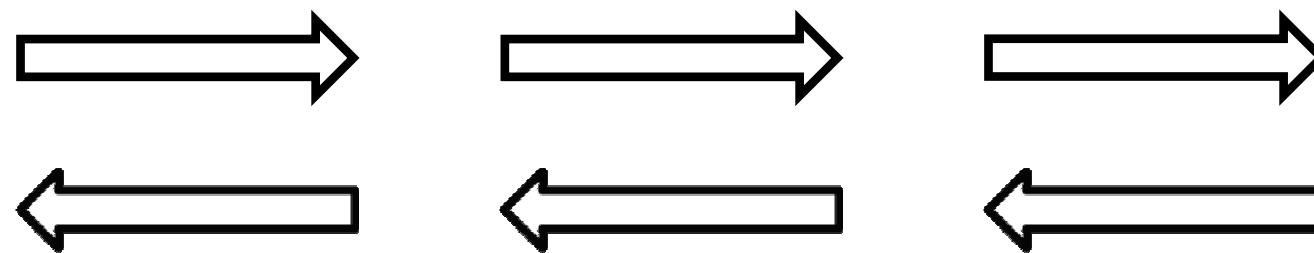
Film thickness Spin-Transfer parameter

$$J_c = J_{c0} \left(1 - \frac{k_B T}{K_F V} \ln \frac{t_p}{\tau_0} \right)$$



Spintronics

STT effect (Noble price ???)

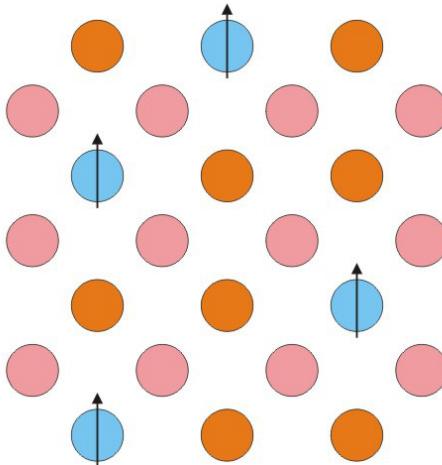


Diluted magnetic semiconductors (DMS)



Spintronics – Diluted Magnetic Semiconductors

DMS



Dispersed TM ions
(~ 5%) on
uniform lattice sites

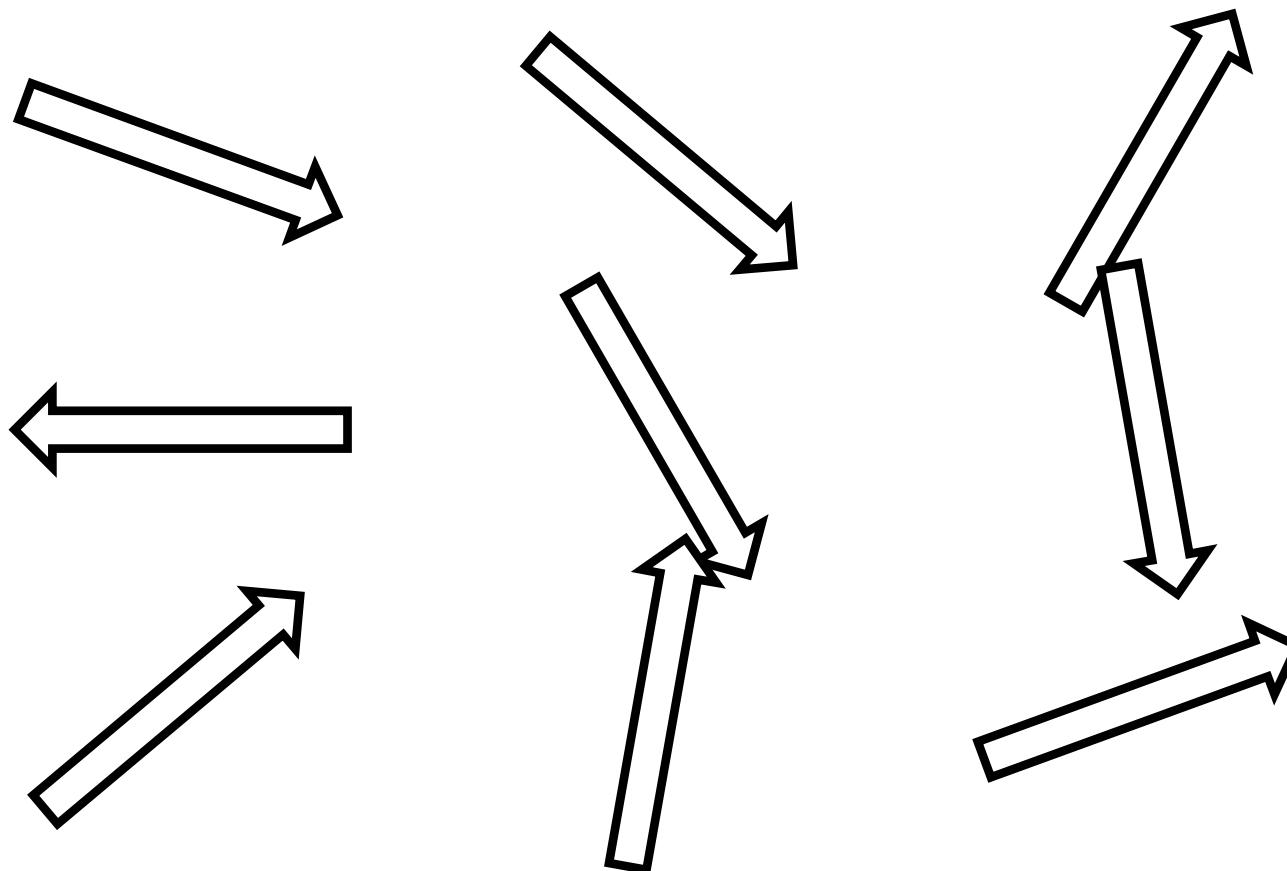
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2. **Paramagnetic DMS, giant Zeeman splitting and bound magnetic polarons**
3. Magnetic coupling
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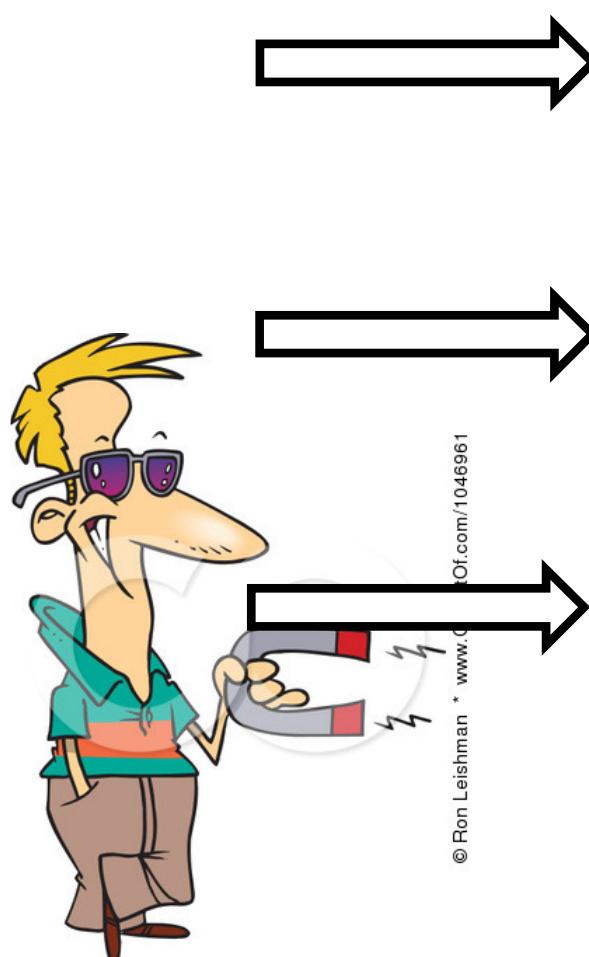
Magnetic moments in external fields

Magnetic field off

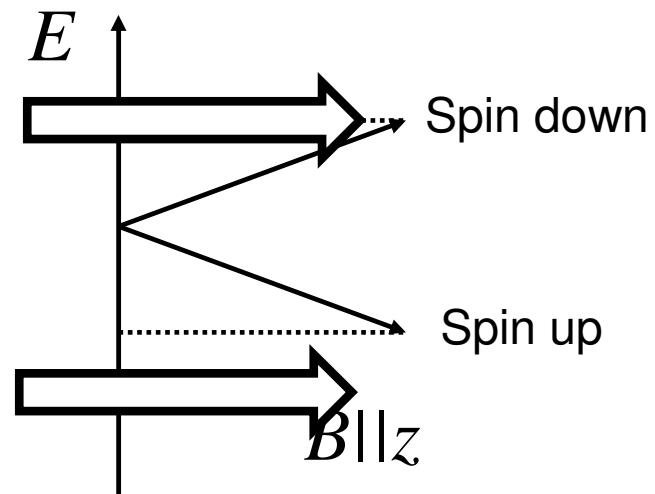


Magnetic moments in external fields

Magnetic field on



$$E = -\mu B \cos \theta$$
$$\theta = \pm \pi$$

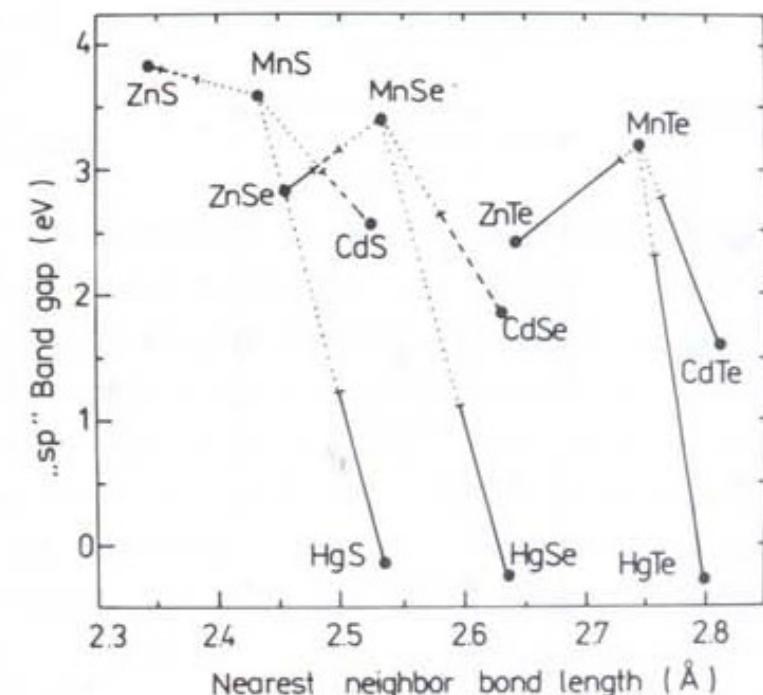


Paramagnetic DMS

J. K. Furdyna, JAP 64, 1988

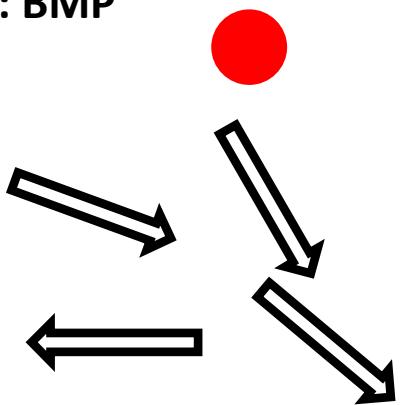
- Mainly II-VI doped with Mn - paramagnetic
- Giant Zeeman splitting of the conduction band
- Spin polarization of charge carriers
- Bound magnetic polaron at zero field
- Spin-glass freezing for high Mn concentration

**Classical DMS: Ideal spin aligner
but no ferromagnetic order at RT!**



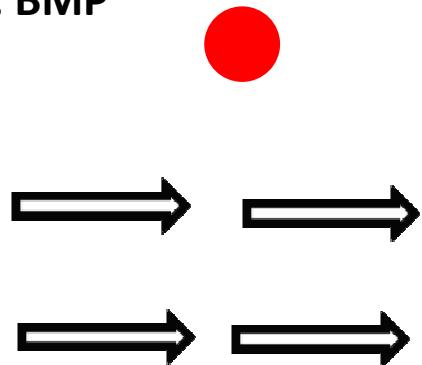
Paramagnetic DMS

Example: BMP



Paramagnetic DMS

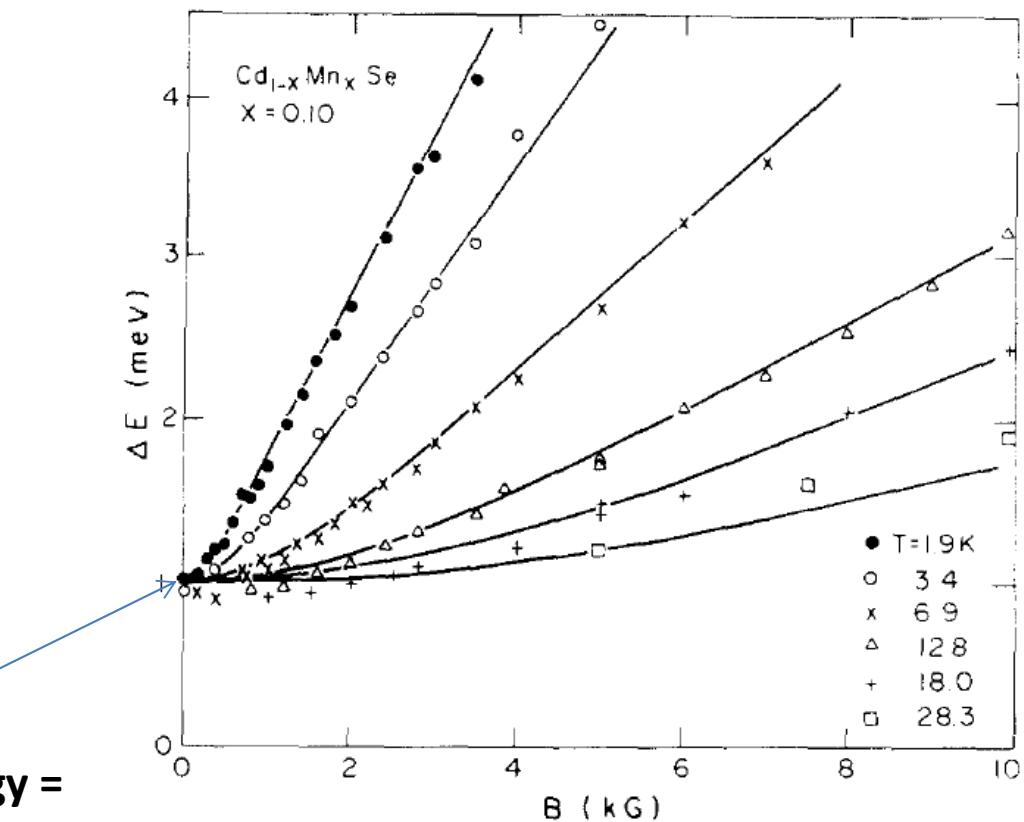
Example: BMP



Origin: *sp-d exchange interaction
Method: *Spin Flip Raman Scattering

Zero field
Spin flip energy =
sign for BMP

Spin-flip energies for CdMnSe
at different temperatures:



P. A. Wolff and J. Warnock

J. Appl. Phys. 55, 2300 (1984)

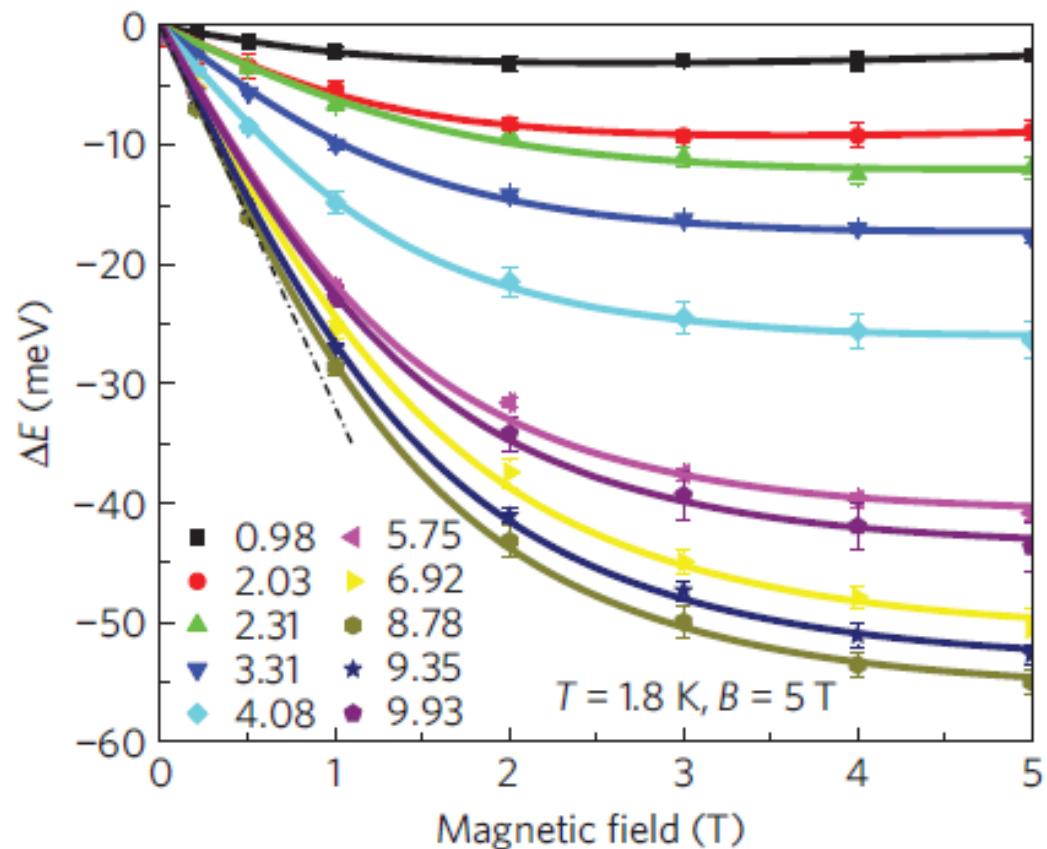


hZDR

Paramagnetic DMS

Example: Giant Zeeman splitting in
CdSe:Mn²⁺
(J. H. Yu et al., Nature Materials 2009)

Method: *MCD
Origin: *sp-d exchange interaction
*negative s-d interaction due to
quantum confinement



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Magnetic coupling

How magnetic moments arrange themselves?



Finding the energy minimum:

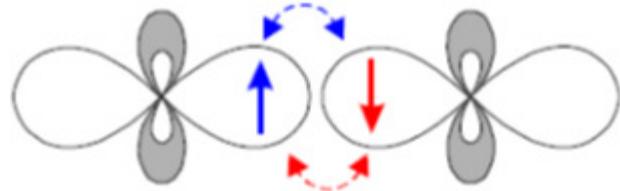
- System gains energy from quantum mechanical exchange interaction (delocalization)
- System avoids too high Coulomb repulsion
- System avoids too large kinetic energies
- Pauli principle must be fulfilled
- Hunds rules must be fulfilled

$$\hat{H}_t = -t \sum_{\langle ij \rangle, \sigma} (c_{i\sigma}^+ c_{j\sigma} + c_{j\sigma}^+ c_{i\sigma}).$$

$$\hat{H}_U = U \sum_i n_{i\uparrow} n_{i\downarrow}$$

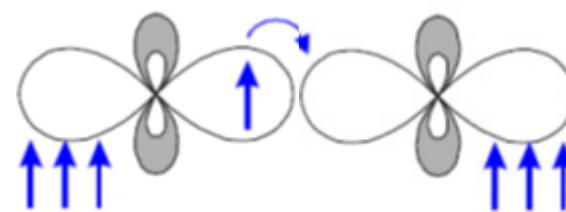
Magnetic coupling

Virtual hopping related



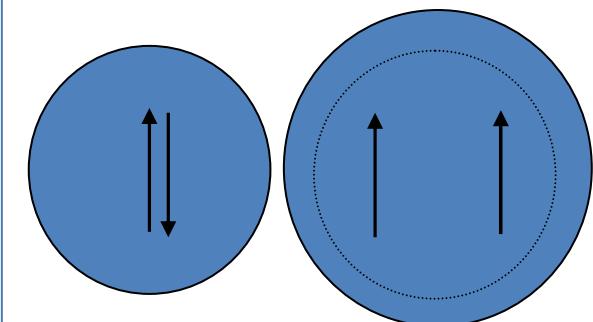
$$\Delta E = 2 \sum_{i>j} \frac{2t^2}{U} S_i S_j$$

Electronic transport related



$$\Delta E \sim -zt \cos \frac{\theta}{2}$$

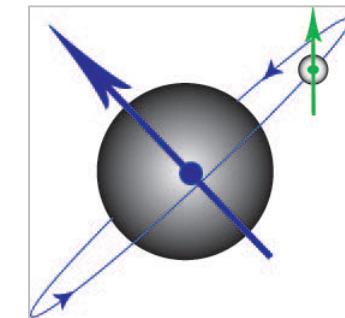
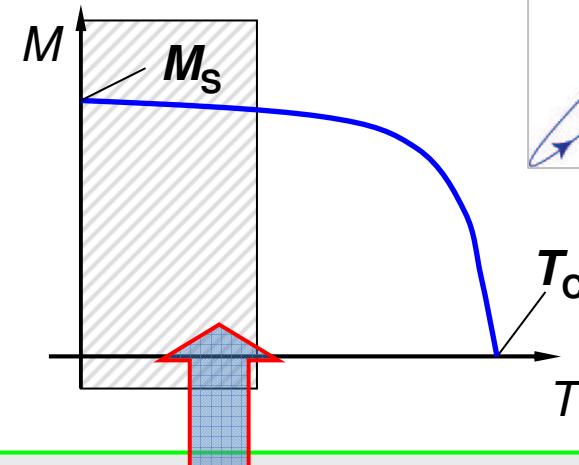
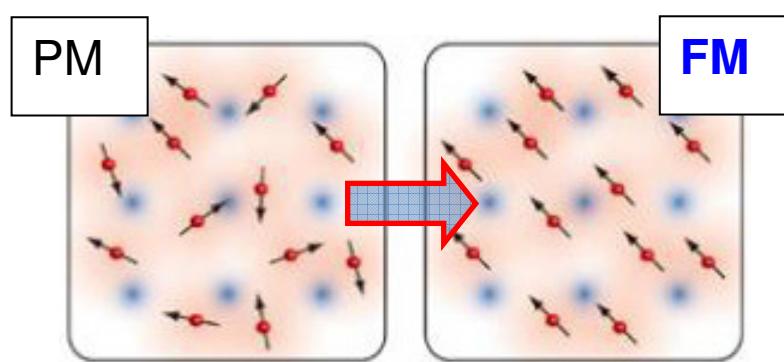
Itinerant



$$\Delta E \sim [1 - U g(E_F)]$$

Magnetic coupling

FM materials possess a **spontaneous magnetization** (magnetization without an external field) due to the **very strong exchange interaction** between atomic magnetic moments (which are due to the own angular moments of electrons called spins)



$$|\mathbf{M}| = \text{Const} = M_s$$

$$\mathbf{m} = \mathbf{M} / M_s$$

Unit vector field $\mathbf{m(r)}$ and the **saturation magnetization** M_s completely characterize the magnetization state of a FM body well below T_c

$$E_{\text{FM}} = F(\{\mathbf{m(r)}\}, M_s)$$

Spintronics – Diluted Magnetic Semiconductors

Magnetic coupling: Lets drive the mobility screw!

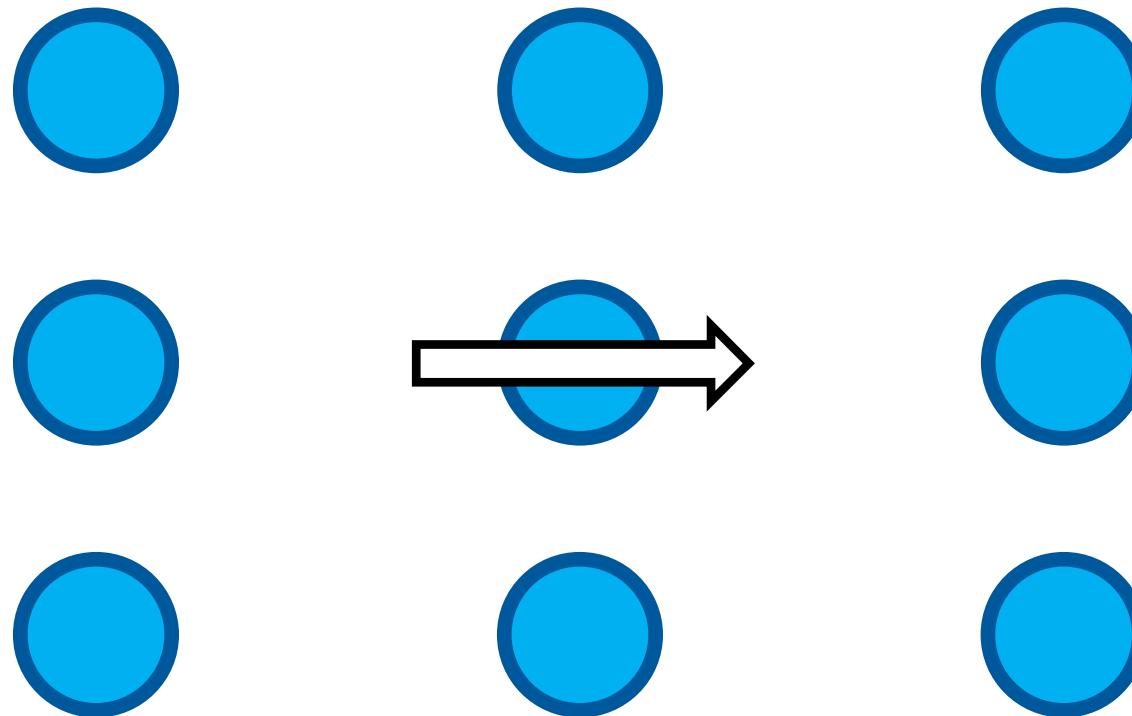
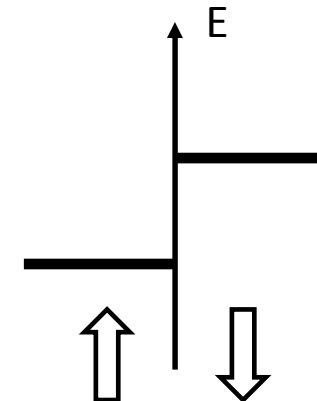




Spintronics – Diluted Magnetic Semiconductors

Driving the screw

- Paramagnet



Local picture

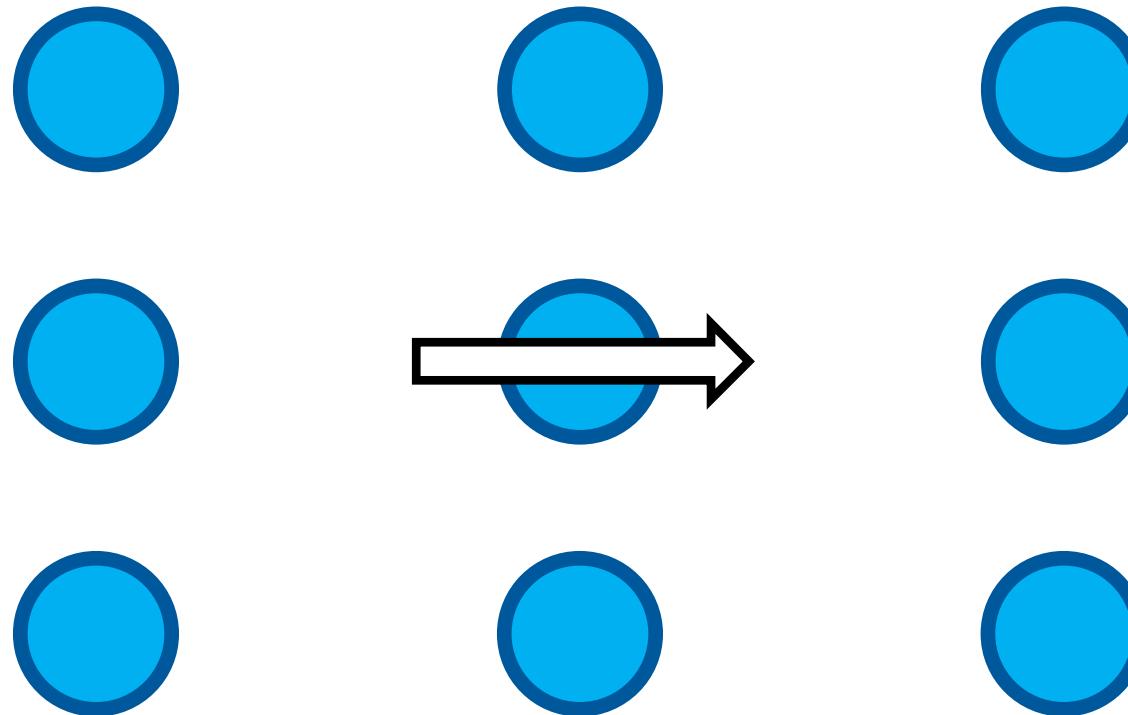
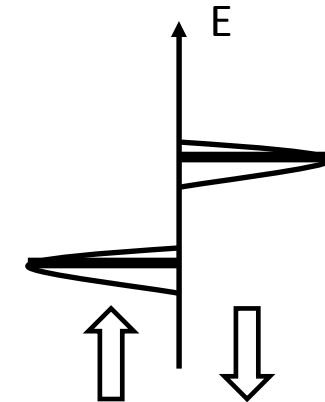
page 31



Spintronics – Diluted Magnetic Semiconductors

Driving the screw

- Paramagnet



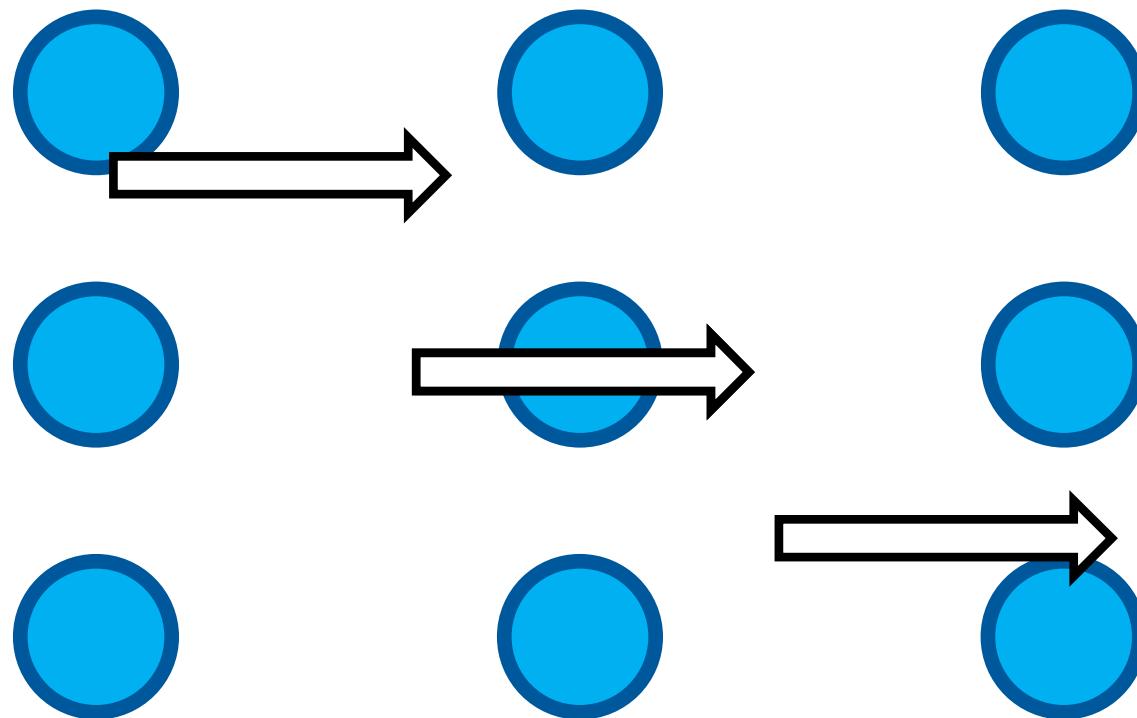
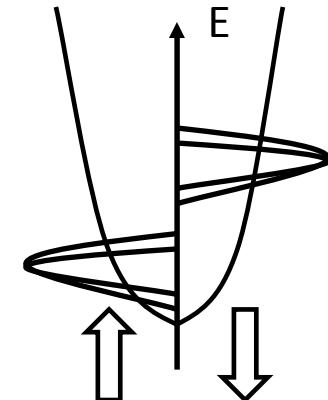
Local picture

page 32

Spintronics – Diluted Magnetic Semiconductors

Driving the screw

- Driving magnet

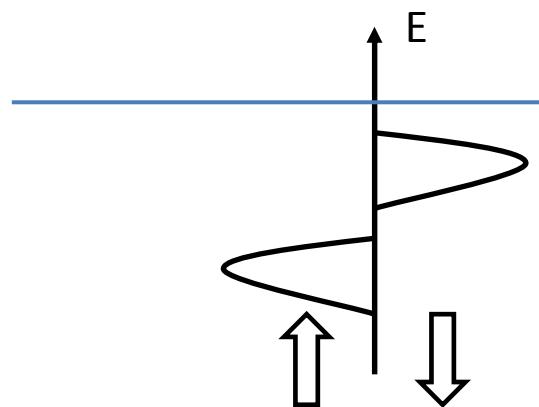


Spintronics – Diluted Magnetic Semiconductors

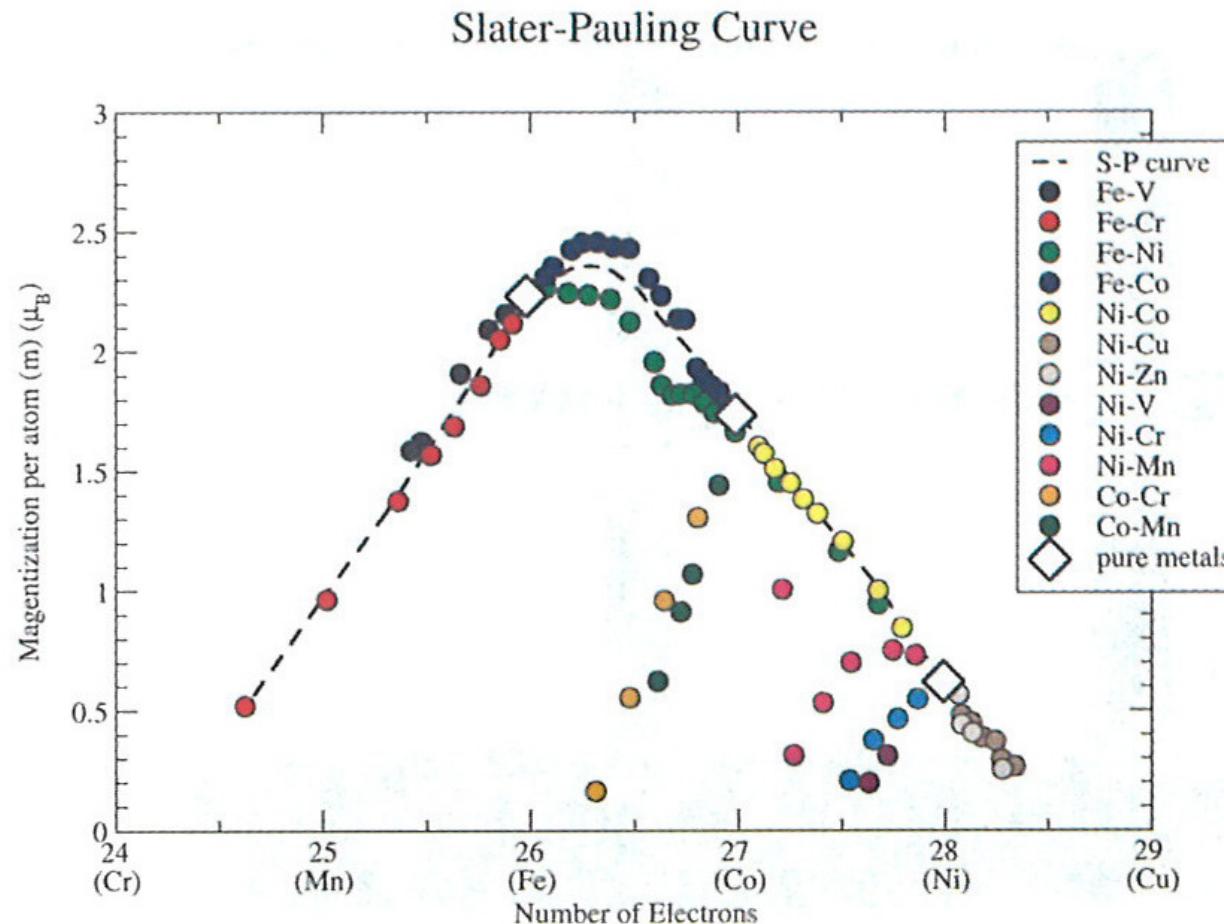
- Ferromagnet

The half-metallic state:

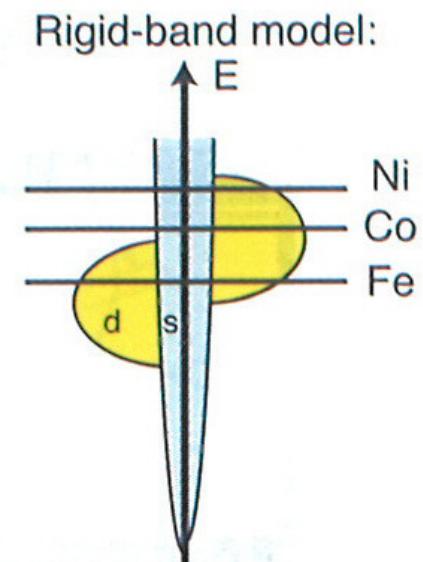
- Magnetic metal
- Magnetic semiconductor
- 100 % Spinpolarization



Metallic ferromagnets



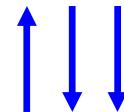
data taken from Bozorth, PR 79, 887 (1950)



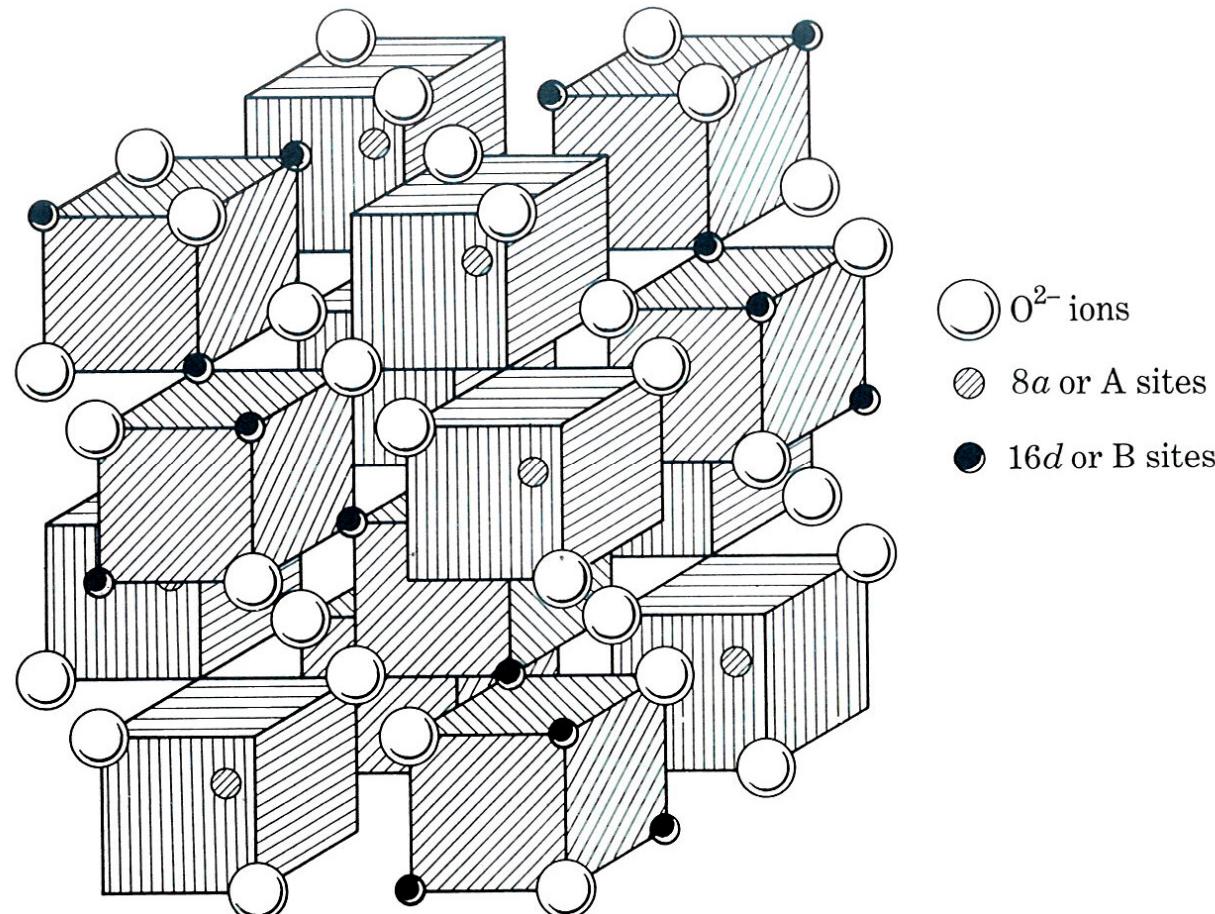
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Ferrimagnetic spinels



Normal spinel structure



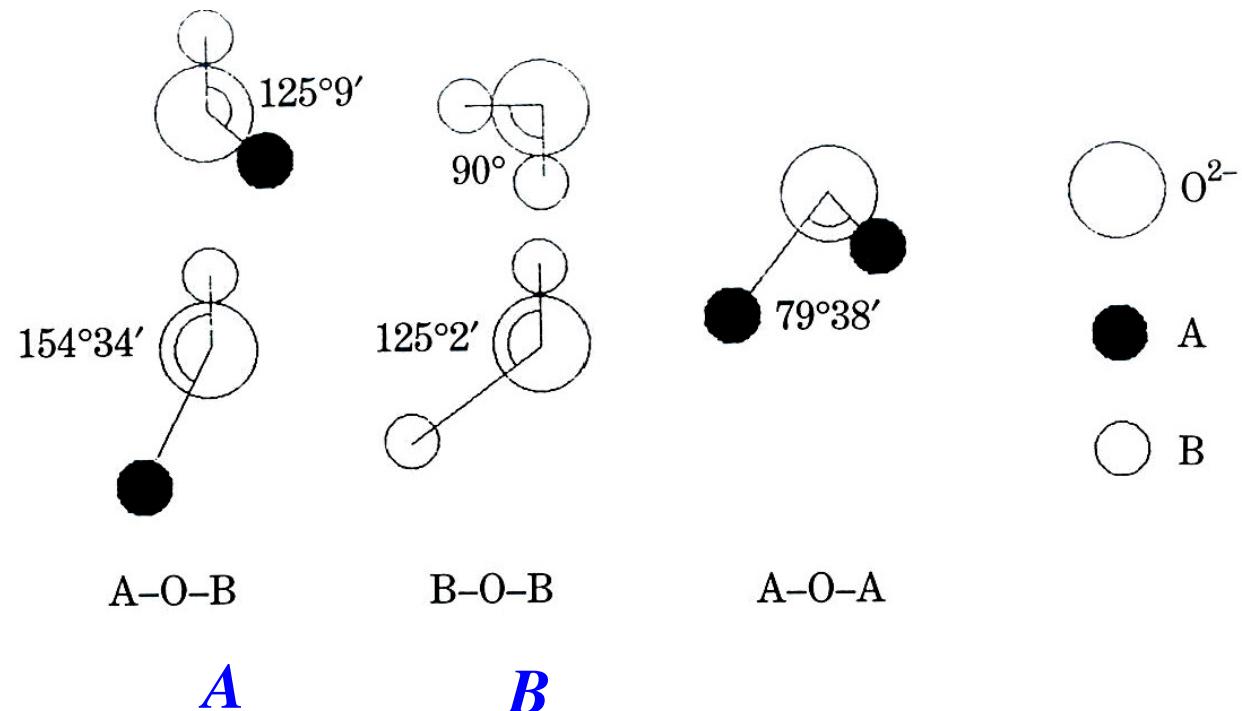
Ferrimagnetic spinels

Magnetic Structure:

A-O-B: Antiferro

A-O-A: Ferro

B-O-B: Ferro



General Spinell:

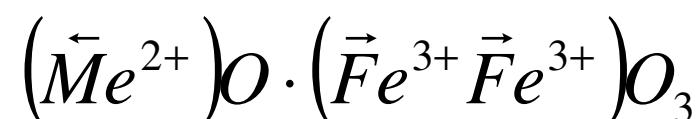


Zn: $\delta=1$

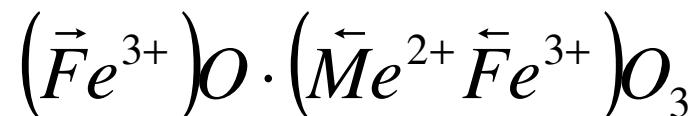
Mn: $\delta=0.8$

Fe: $\delta=0$

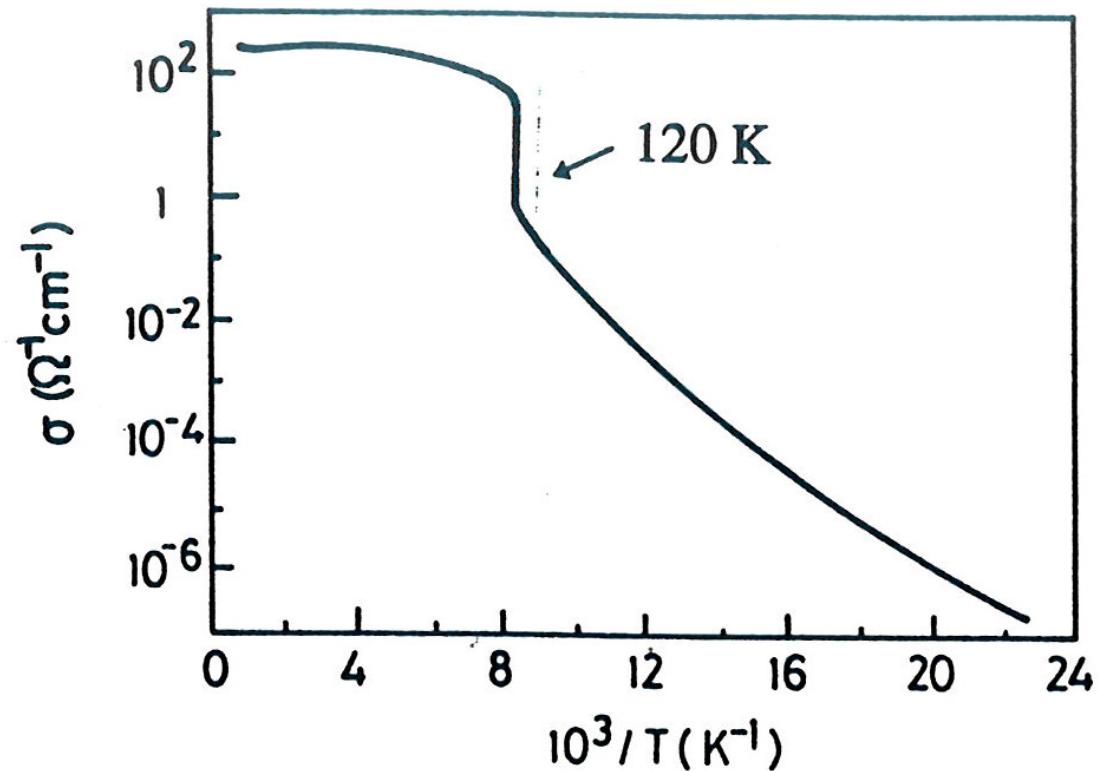
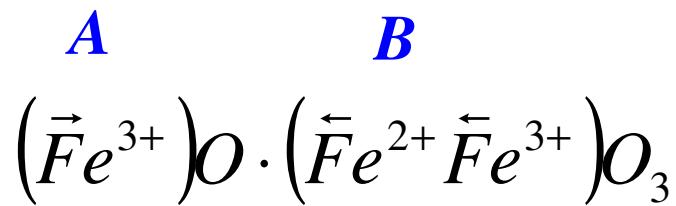
Normal Spinell ($\delta=1$):



Inverse Spinell ($\delta=0$):



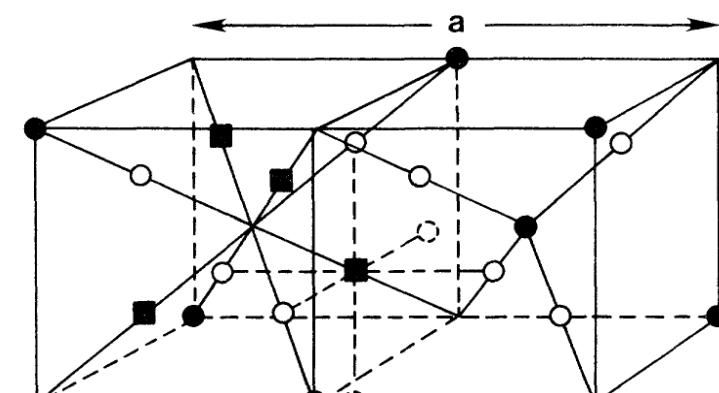
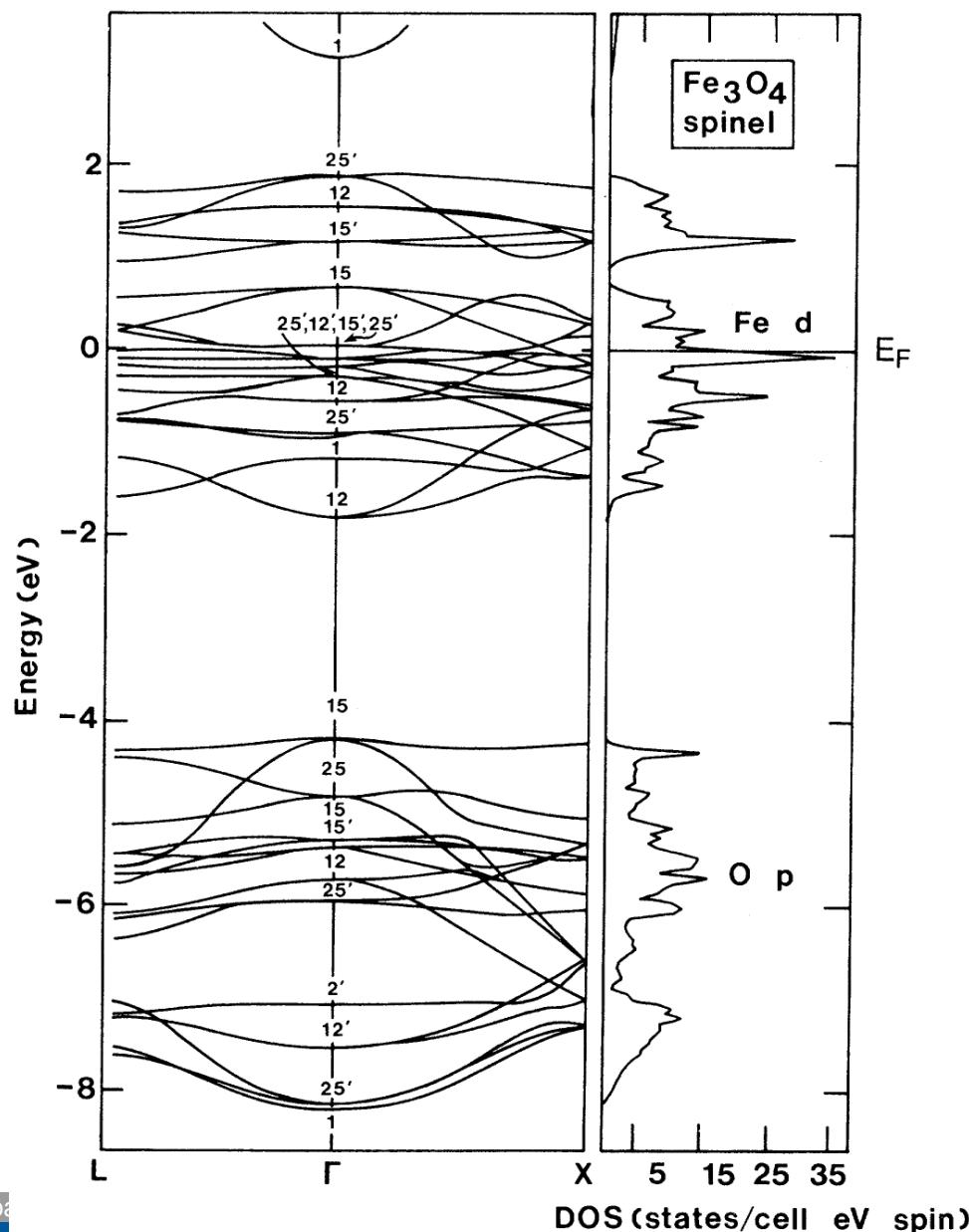
Ferrimagnetic spinels: Magnetite



**Fe³⁺ and Fe²⁺ coexist on B-sites → e⁻-hopping is possible
→ High conductivity and spin polarized electrons**



Ferrimagnetic spinels: Magnetite



Cubic spinel structure

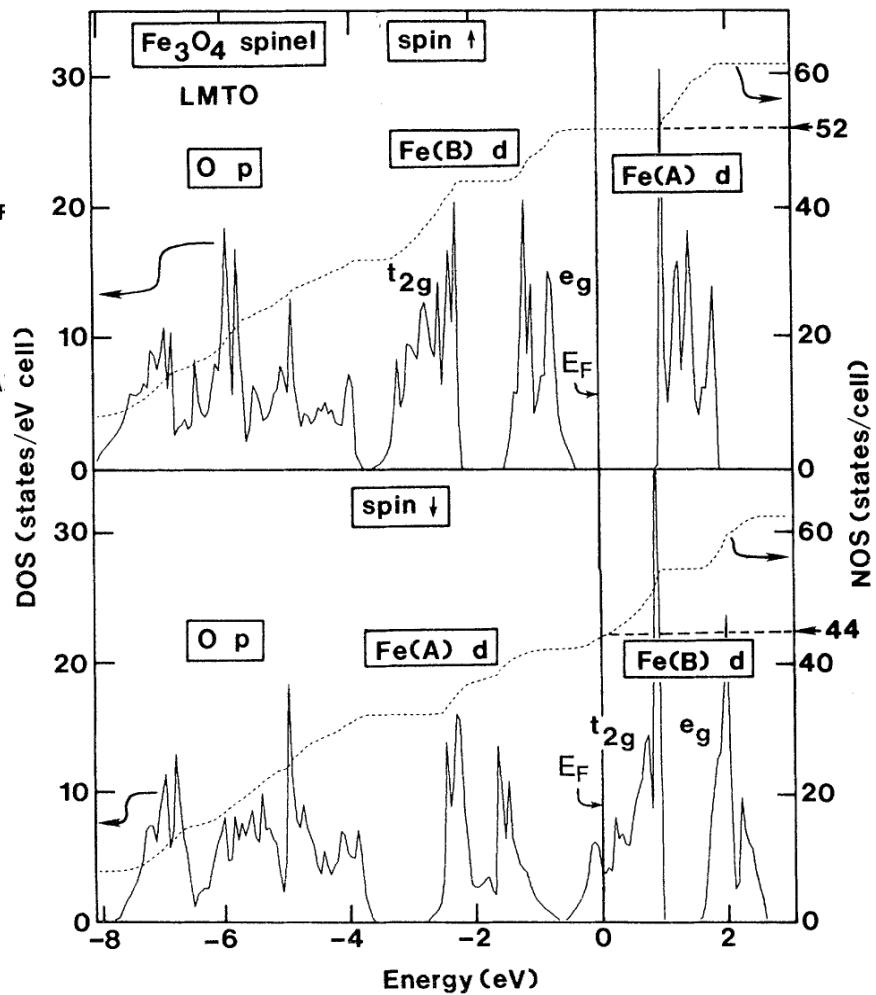
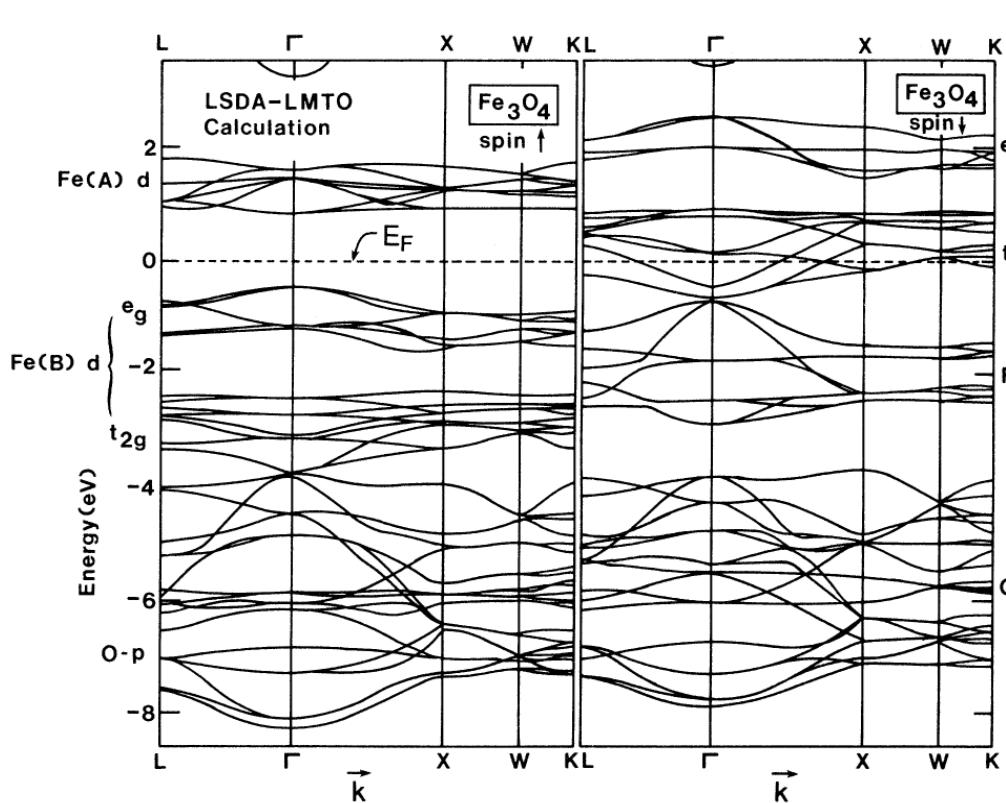
$\text{Fe(A)} \bullet \quad \text{Fe(B)} \blacksquare \quad \circ \text{O}$

(a)

Fe(A) Fe(B)

$5\mu_B$ $5\mu_B$ $4\mu_B$

Ferrimagnetic spinels: Magnetite



- Mixed valency Fe^{2+3+}
- Moment = $4\mu_B$ per Fe_3O_4 formula unit
- ↑ semiconducting, $E_g \sim 1.5$ eV

Ze Zhang, PRB 1991



Magnetic semiconductors: Eu chalcogenides

EuO: $T_c = 69.33 \text{ K}$ $\Delta E = 1.12 \text{ eV}$ $a = 5.141 \text{ \AA}$

EuS: $T_c = 16.57 \text{ K}$ $\Delta E = 1.65 \text{ eV}$ $a = 5.986 \text{ \AA}$

EuSe: $T_M = 4.6 \text{ K}$ $\Delta E = 1.8 \text{ eV}$ $a = 6.195 \text{ \AA}$

EuTe: $T_N = 9.58 \text{ K}$ $\Delta E = 2.0 \text{ eV}$ $a = 6.598 \text{ \AA}$

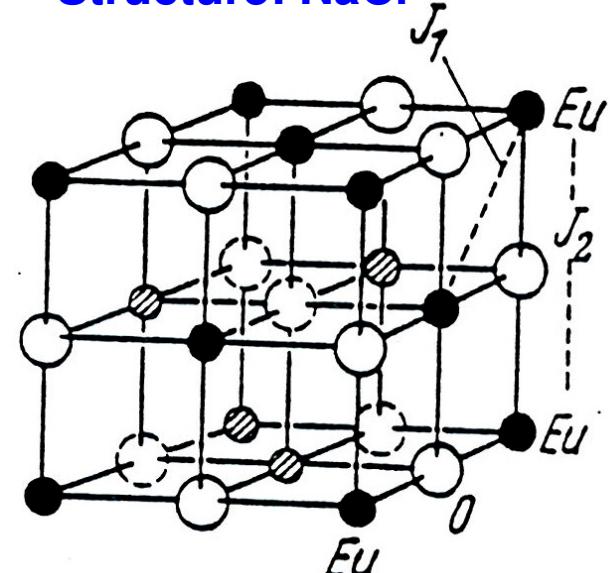
Heisenberg-exchange

Interaction between nearest (nN) and
Next nearest neighbors (nnN):

$$\hat{H} = -\sum_{ij} \tilde{J}_{ij} \vec{S}_i \vec{S}_j$$

$$\hat{H} = -\tilde{J}_1 \sum_{nN} \vec{S}_0 \vec{S}_{nN} - \tilde{J}_2 \sum_{nnN} \vec{S}_0 \vec{S}_{nnN}$$

Structure: NaCl



$J_1 > 0$, 4f-5d-4f-exchange (5d=empty orbital) - FM

$J_2 < 0$, 4f-2p-4f-Superexchange (180°) - AFM

Magnetic semiconductors: Eu chalcogenides

IFF Ferienkurs Jülich 1998

$$\hat{H} = -\tilde{J}_1 \sum_{nN} \vec{S}_0 \vec{S}_{nN} - \tilde{J}_2 \sum_{nnN} \vec{S}_0 \vec{S}_{nnN}$$

Molekular field approximation:

$$T_C = \frac{2}{3} S(S+1)(z_1 \tilde{J}_1 + z_2 \tilde{J}_2)$$

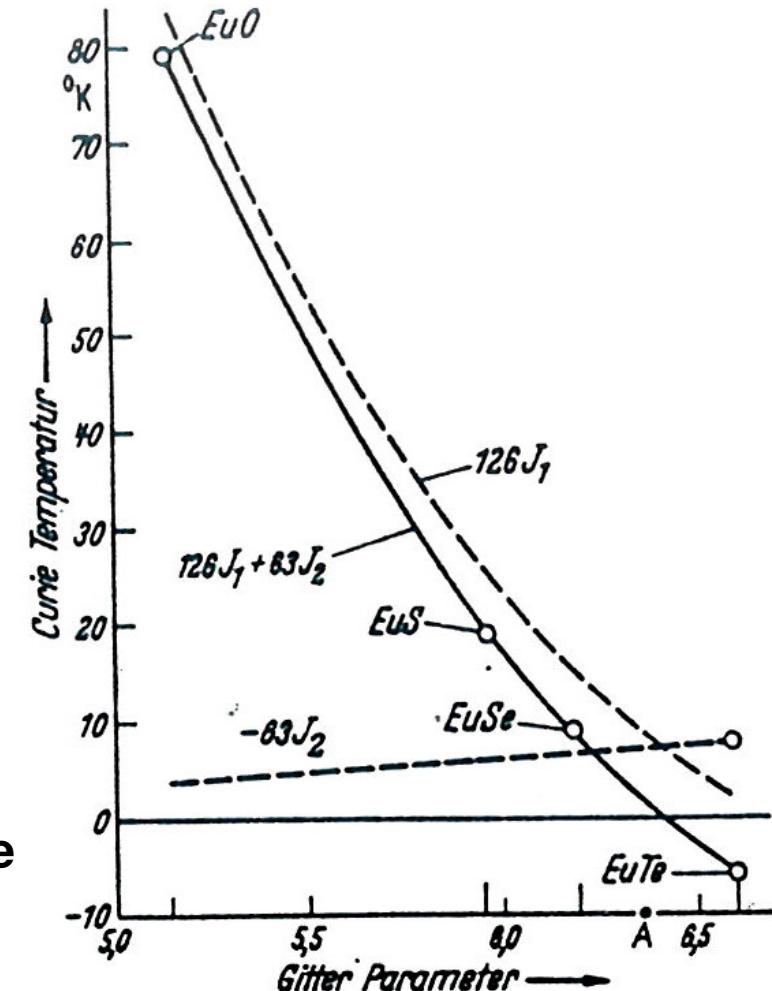
$$T_N = \frac{2}{3} S(S+1)(-z_2 \tilde{J}_2)$$

nN (12) nnN (6)

$$T_C = 126J_1 + 63J_2$$

$$T_N = -63J_2$$

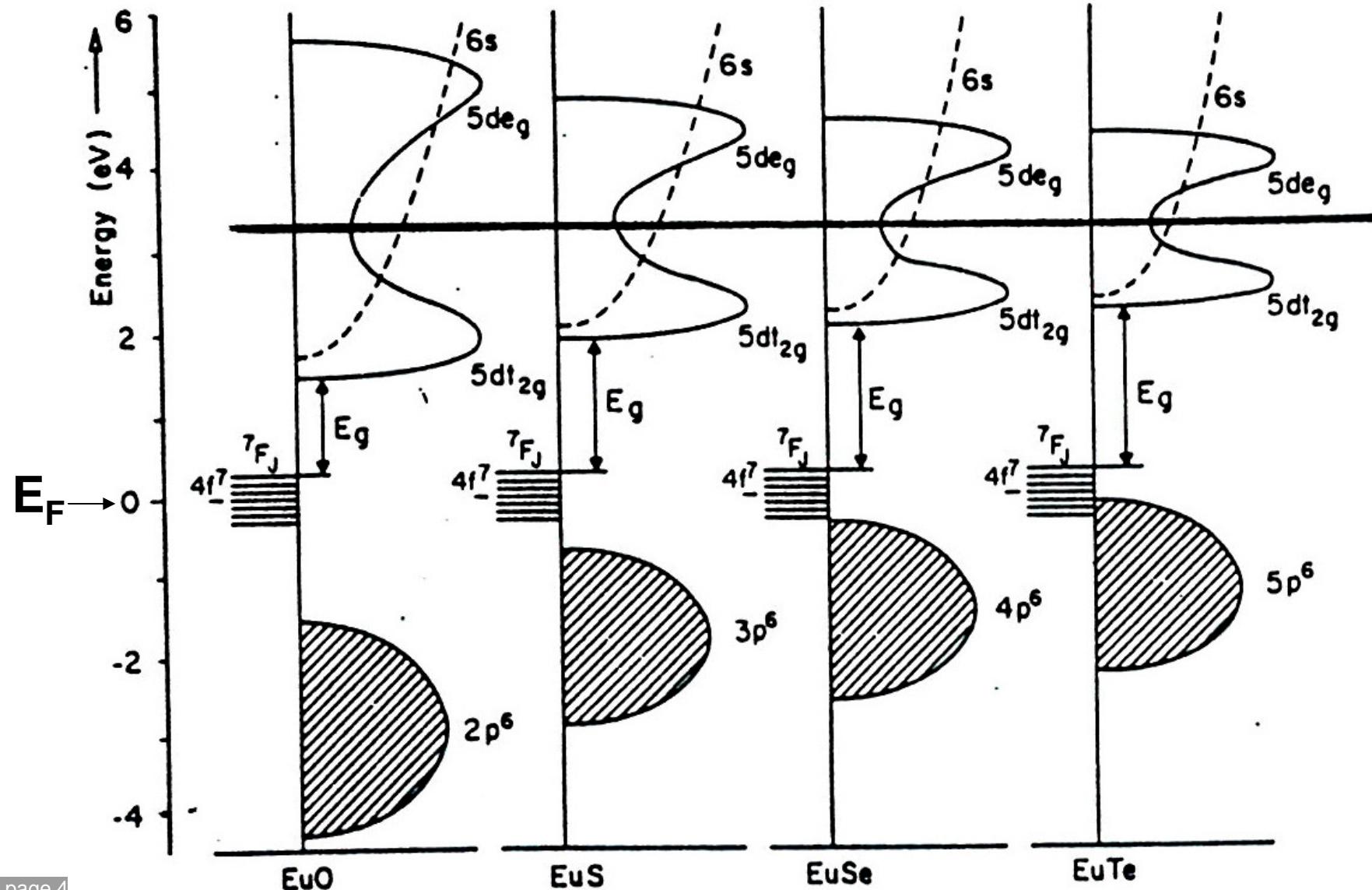
Changes with lattice constant



Magnetic semiconductors: Eu chalcogenides

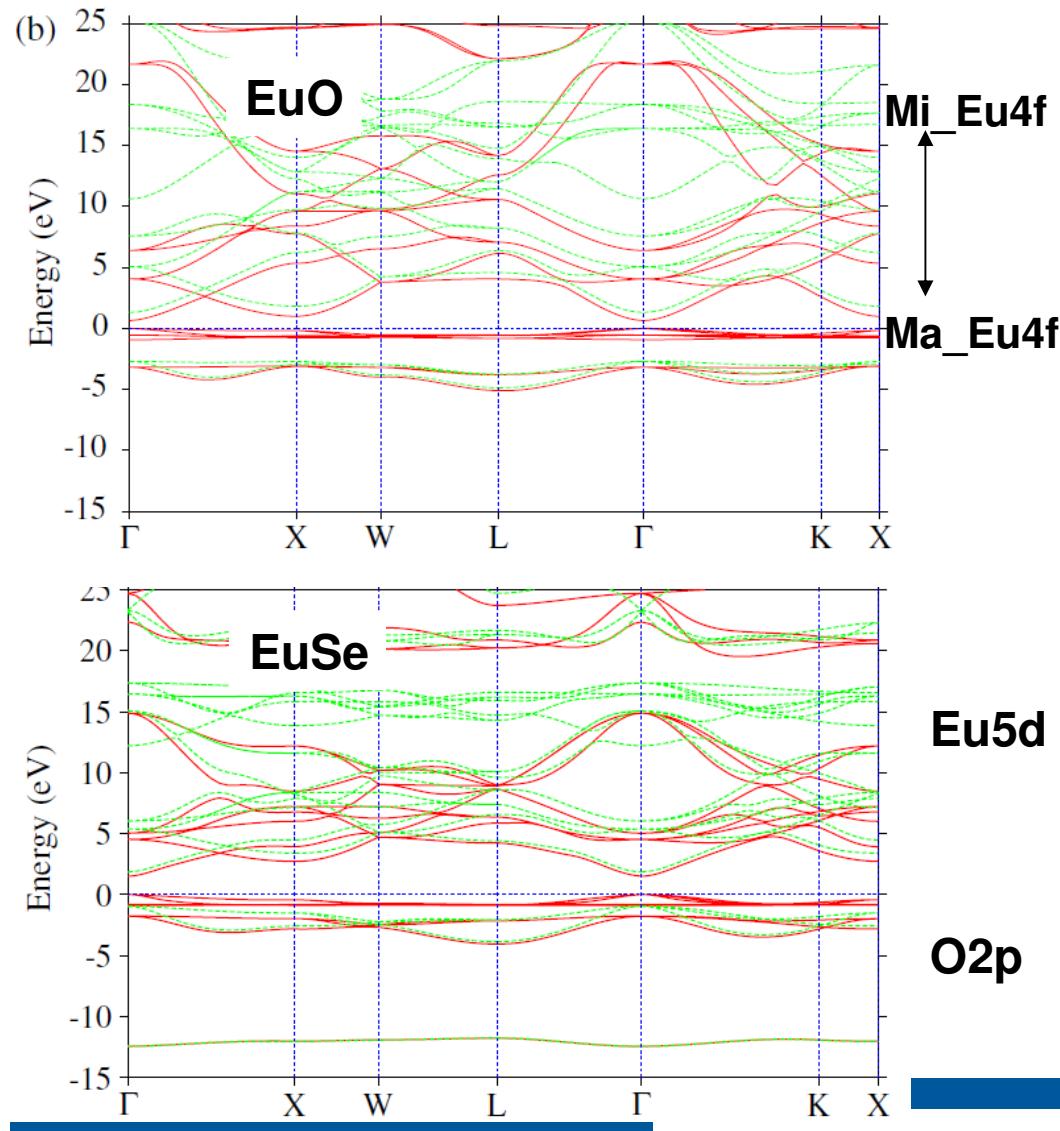
Paramagnetic DOS

$4f^7Eu$

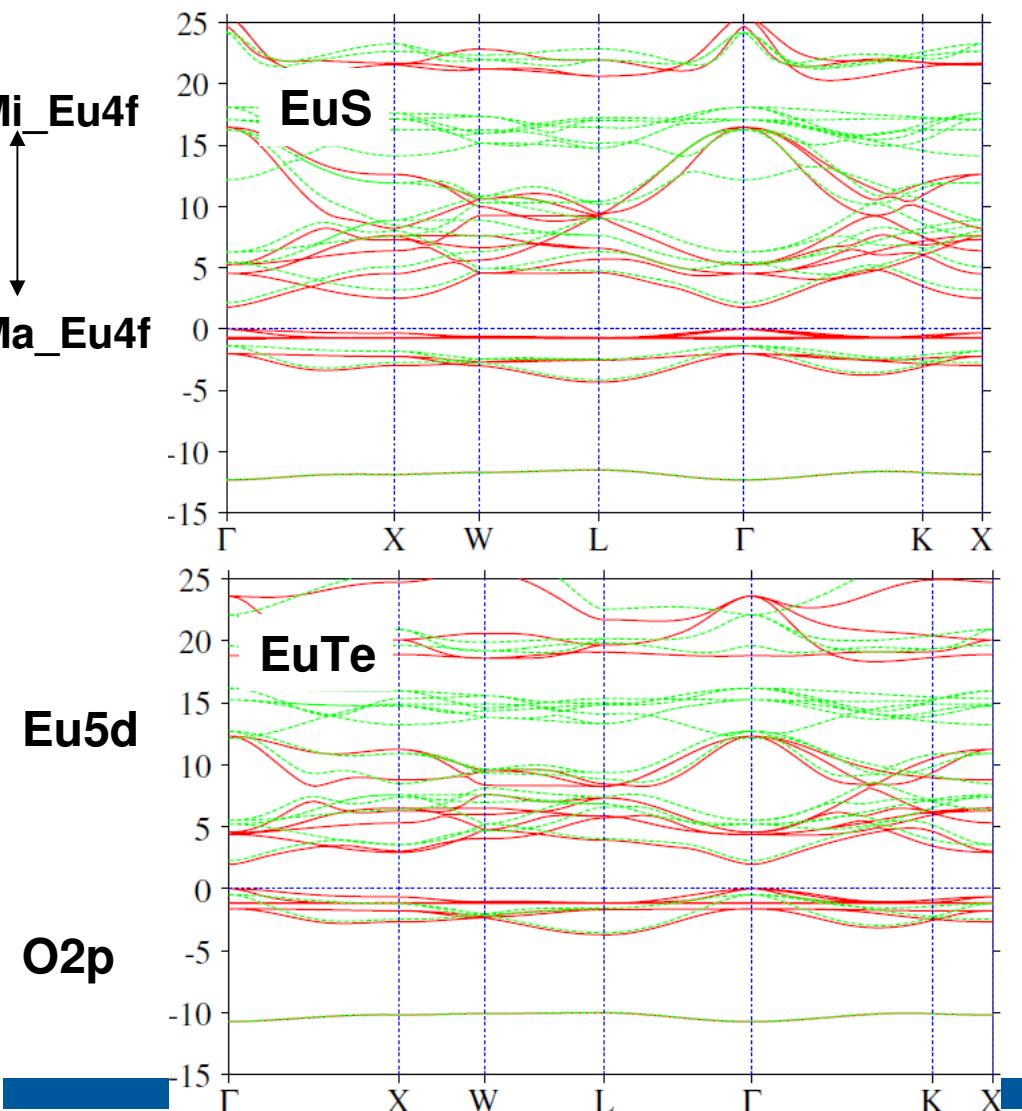


Magnetic semiconductors: Eu chalcogenides

DOS for Eu-Chalkogenides (FM)



Red=Majority (Ma)
Green=Minority (Mi)



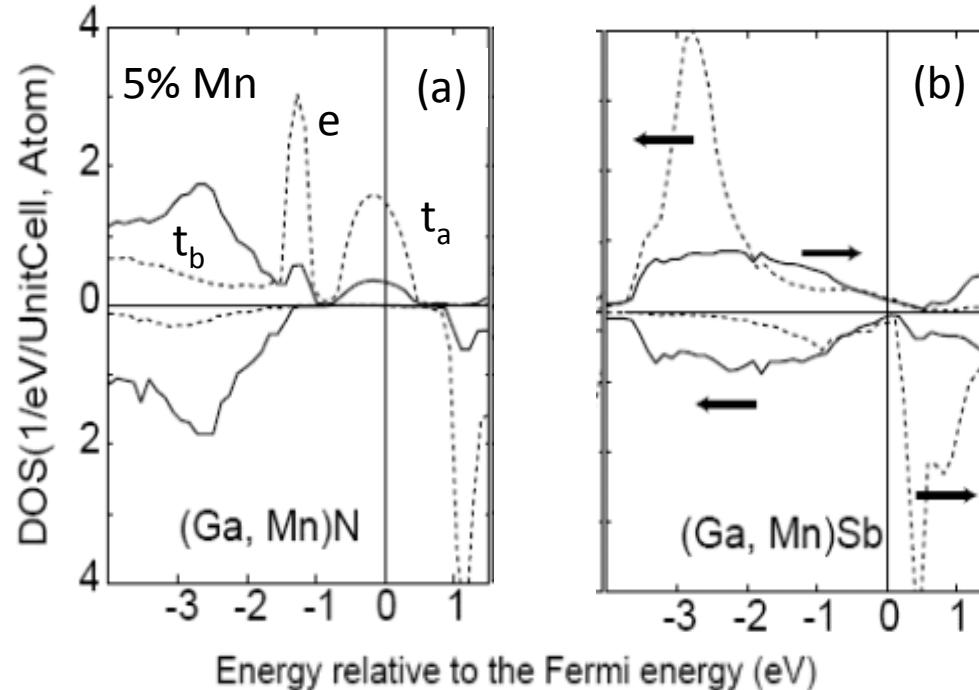
Outline

1. Principles of spintronics
2. Paramagnetic DMS, giant Zeeman splitting and bound magnetic polarons
3. Magnetic coupling
4. Half metallic materials
- 5. Magnetism and Transport in GaMnAs**
6. Oxide based DMS by ion implantation

Spintronics – Diluted Magnetic Semiconductors

Theory

P. H. Dederichs,
IFF Ferienkurs
Jülich, 2005

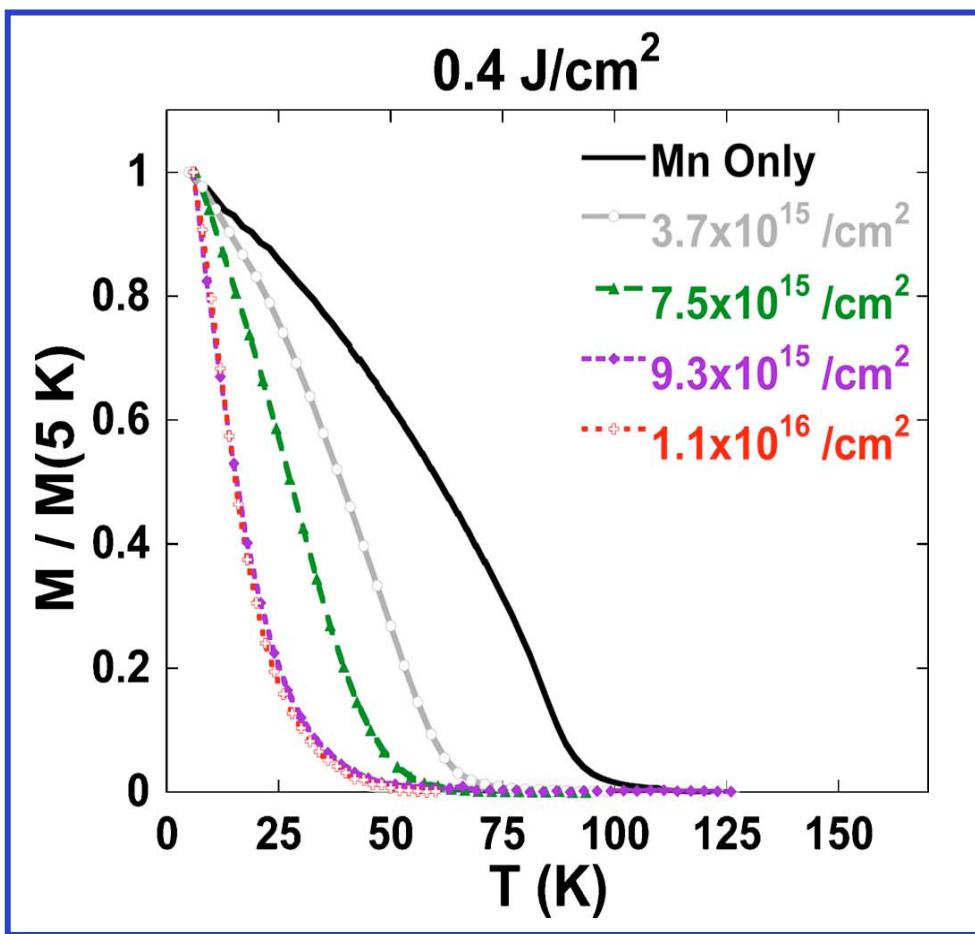


(a) – d-d Zener exchange - $T_C \sim \sqrt{c}$

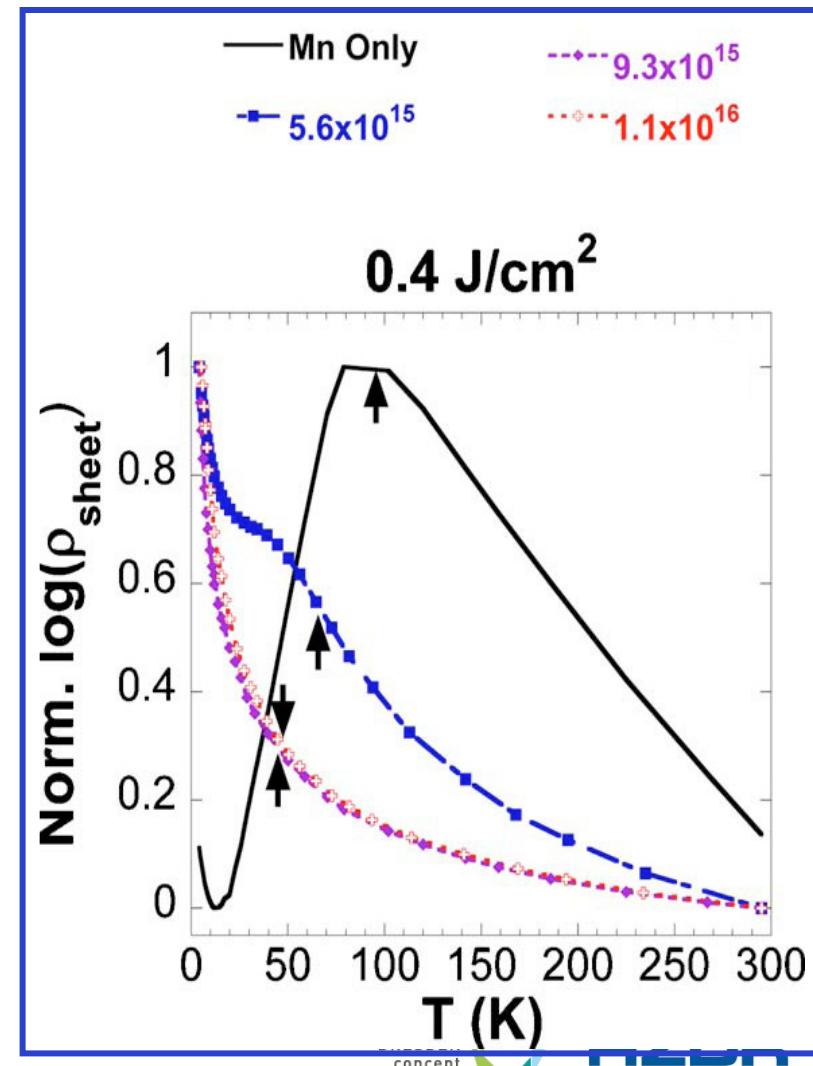
(b) – p-d Zener exchange - $T_C \sim c$

Spintronics – Diluted Magnetic Semiconductors

GaAs + Mn + Te Implantation + laser-melting:
FM for metallic and localized case



Scarpulla et al. JAP 103, 123906, (2008)



concept

Spintronics – Diluted Magnetic Semiconductors

- Reproducible ferromagnetic DMS: GaMnAs (T. Dietl, Science, 2000)
- Curie temperature record (2011) = -88 °C

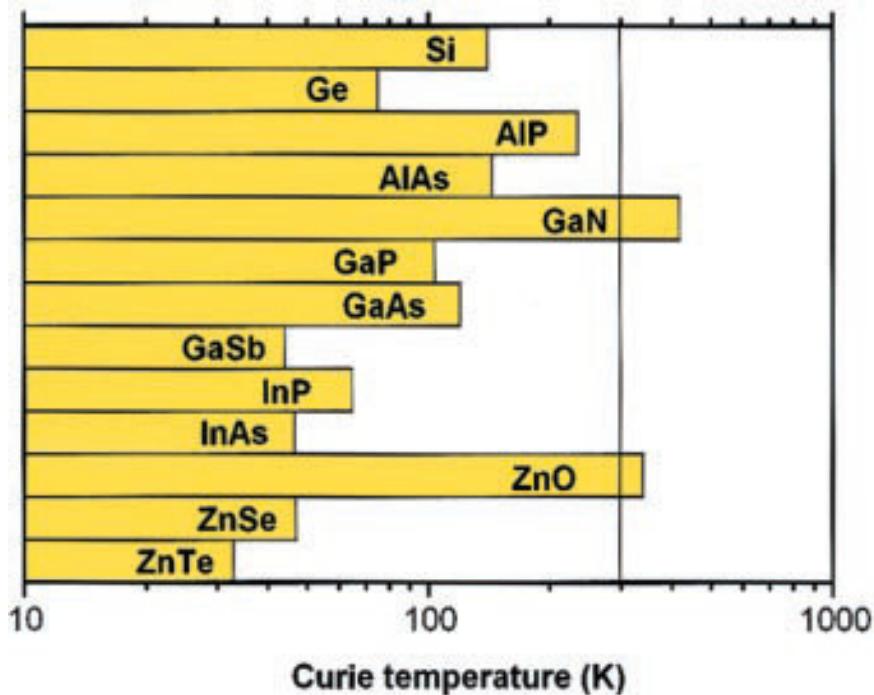


Outline

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Magnetic doping of oxides

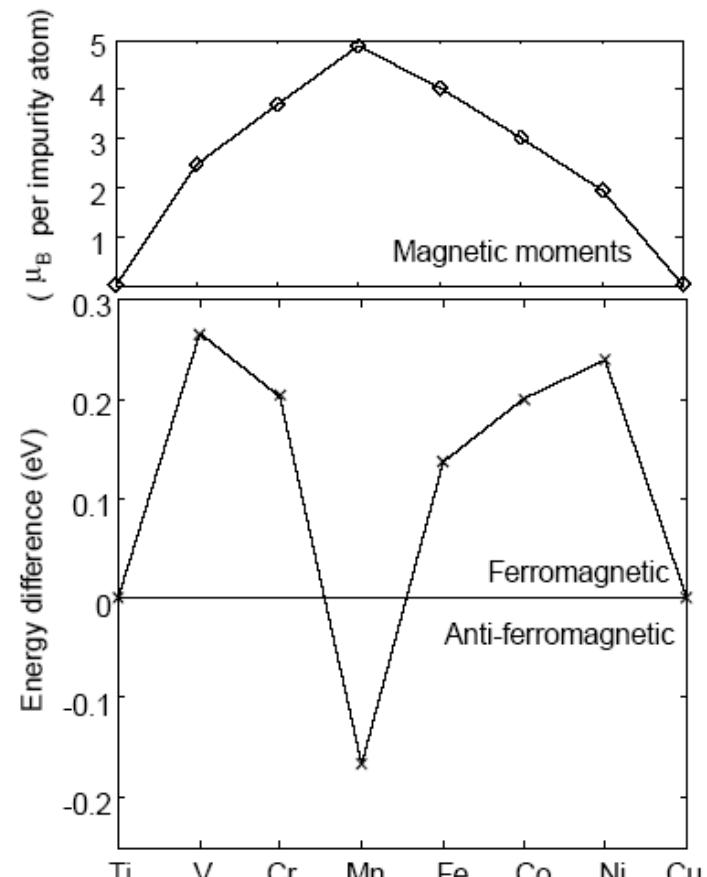
TM doped semiconductors and oxides



5% Mn and p-carriers

$3.5 \times 10^{20}/\text{cm}^3$

T. Dietl, Science, 287 (2000) 1091



25% TM in ZnO, without any additional carrier doping

K. Sato, JJAP, 39 L555

Magnetic doping of oxides

Co or V ions

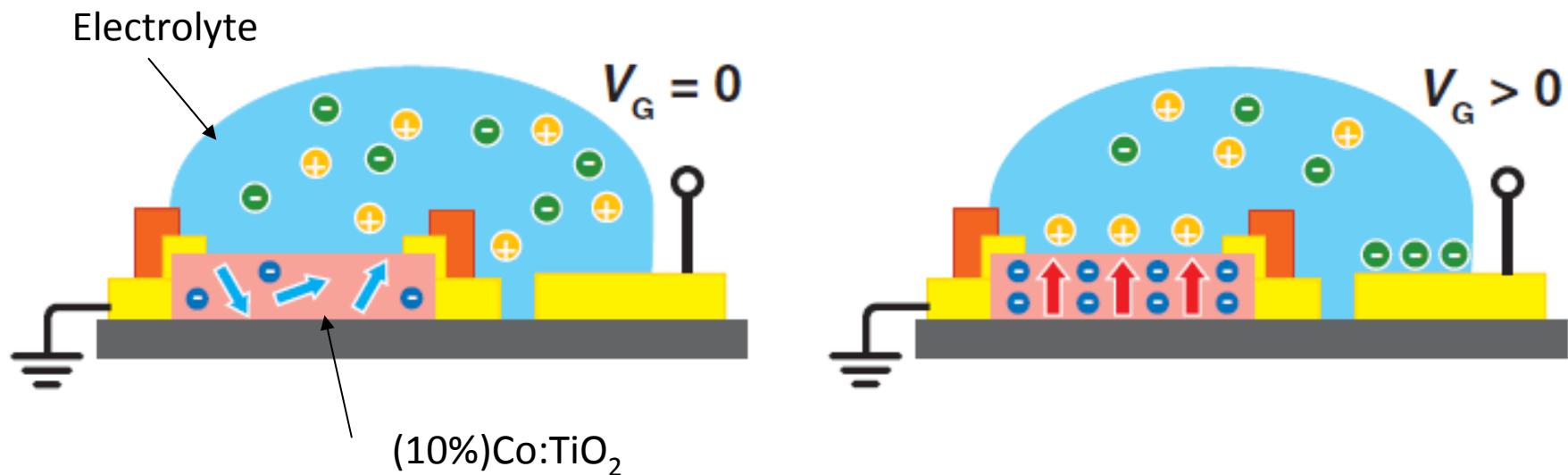


Oxide (here TiO₂)

Magnetic doping of oxides

Candidate with electrically induced magnetization switching:

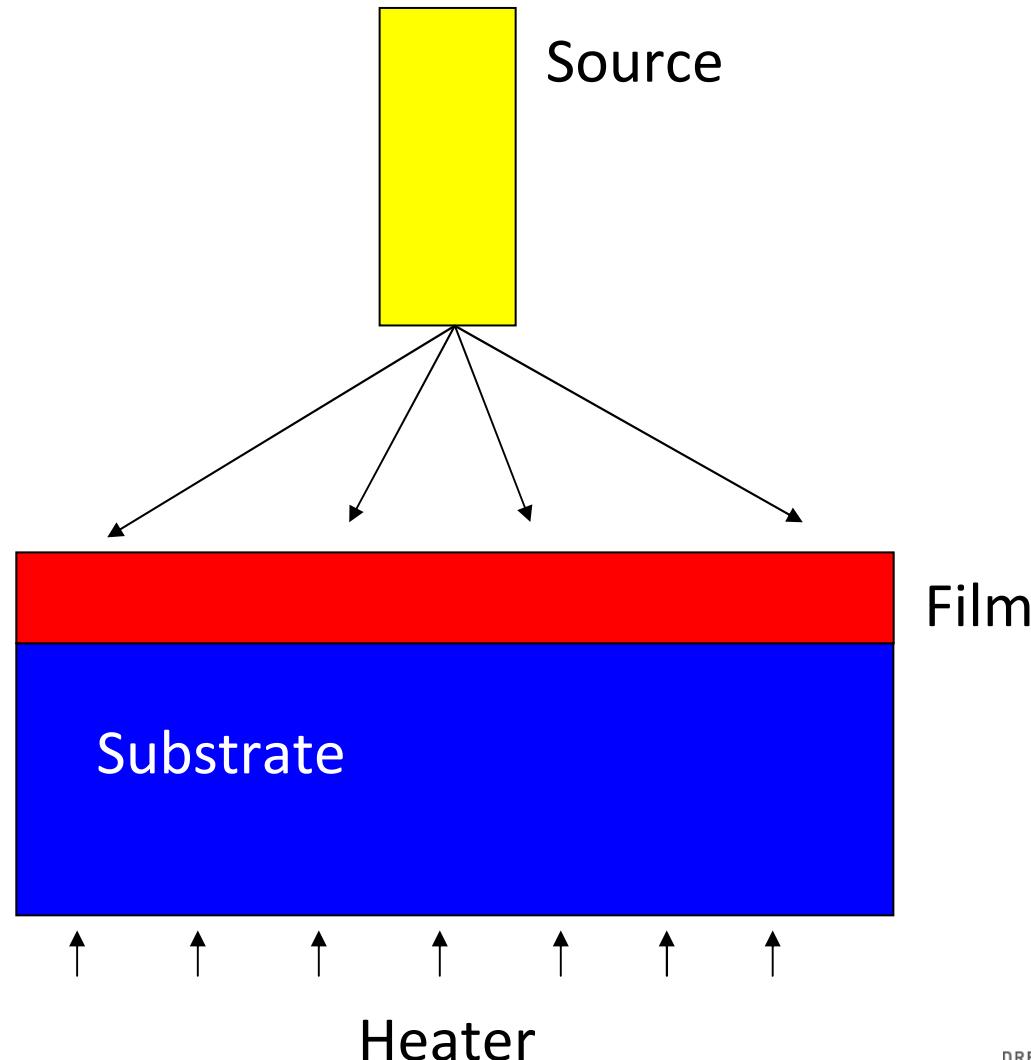
- TiO_2 doped with Cobalt (e.g., Y. Yamada, Science 2011)





Deposition methods

- Physical vapor deposition





Thin film growth

Physisorption/Chemisorption: Weak/Strong interaction between substrate and deposited film

Physisorption

Van der Waals between the atoms of
The film and their image charge:
Lenard-Jones-Potential

$$U(r) = -\frac{\alpha}{r^6} + \frac{\beta}{r^{12}}$$

Flat potential minimum of 10-100 meV
at 0.3-1 nm above the surface

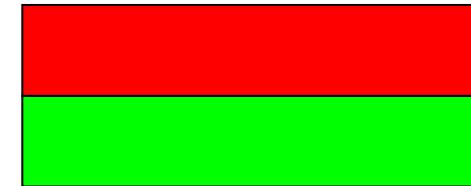
Chemisorption

Change of the electronic structure
Of the surface. Described by Morse
Potentials:

Deep potential minimum ~eV
at 0.1-0.3 nm above the surface

Thermodynamic considerations

Assumption of a thermodynamic equilibrium:
Surface diffusion must be faster than particle flux.



Energies for dominating surface energies γ :

$$\gamma_{Substr} < \gamma_{Adsorbat} + \gamma_{Interface}$$

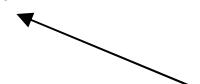
→ Island growth (Old surface+
droplets preferred)

$$\gamma_{Substr} \geq \gamma_{Adsorbat} + \gamma_{Interface}$$

→ Layer growth (Formation of new
surface preferred)

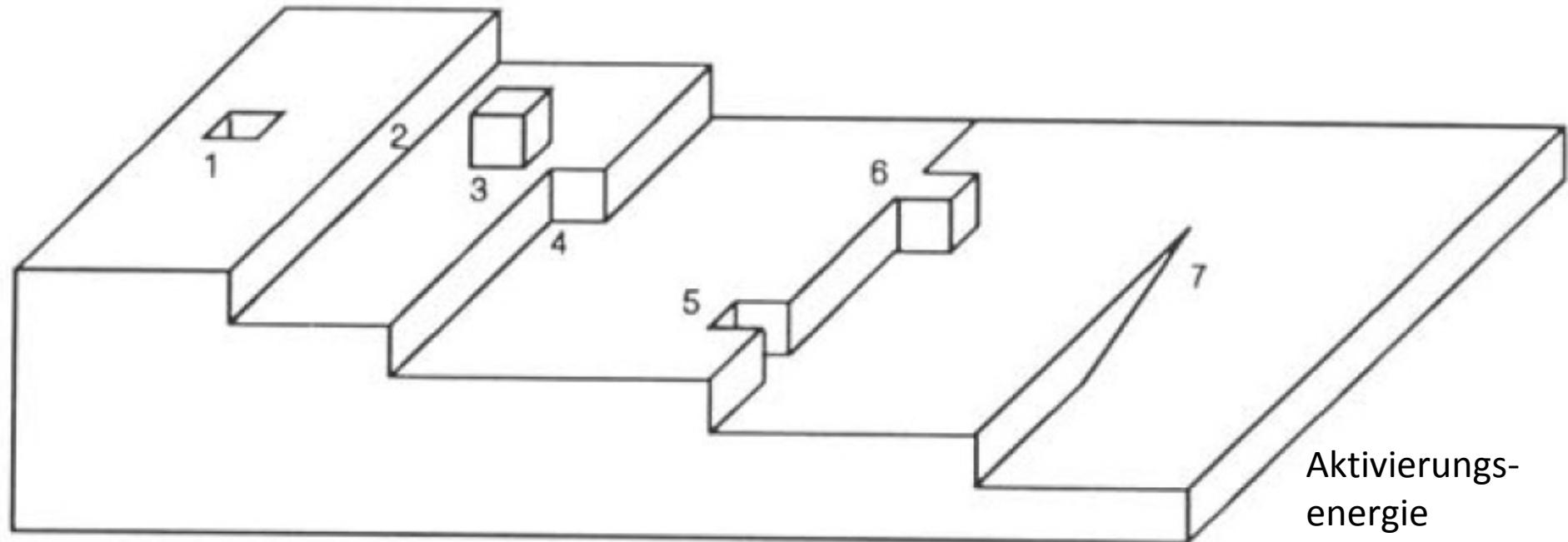
$$\gamma_{Interface} = 0$$

→ Homoepitaxie



Deviation of the interface energy from that
of two substrate layers

Kinetic/microscopic model



- (1) Vacancy
- (2) At step
- (3) Adsorbate (single atom)
- (4) Kink
- (5) In step
- (6) Adsorbate at step
- (7) Screw dislocation

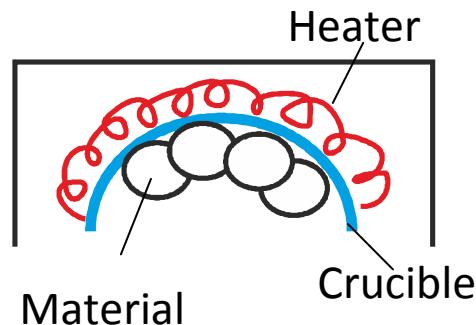
Duration of stay: $\tau = \frac{1}{\nu} e^{\frac{E_{desorption,diffusion}}{k_B T}}$

- Surface defects are preferential sites for nucleation

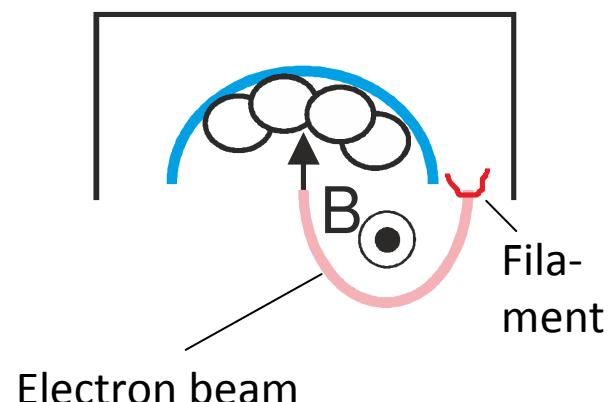
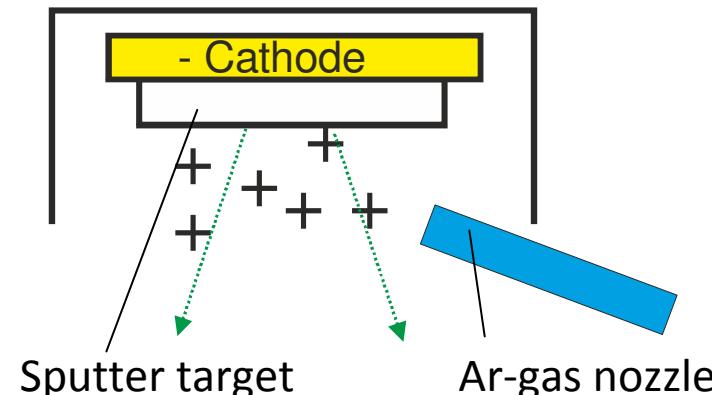


Methods for deposition

Molecular beam epitaxy
(MBE)



Sputter deposition



Pulsed laser deposition
(PLD)

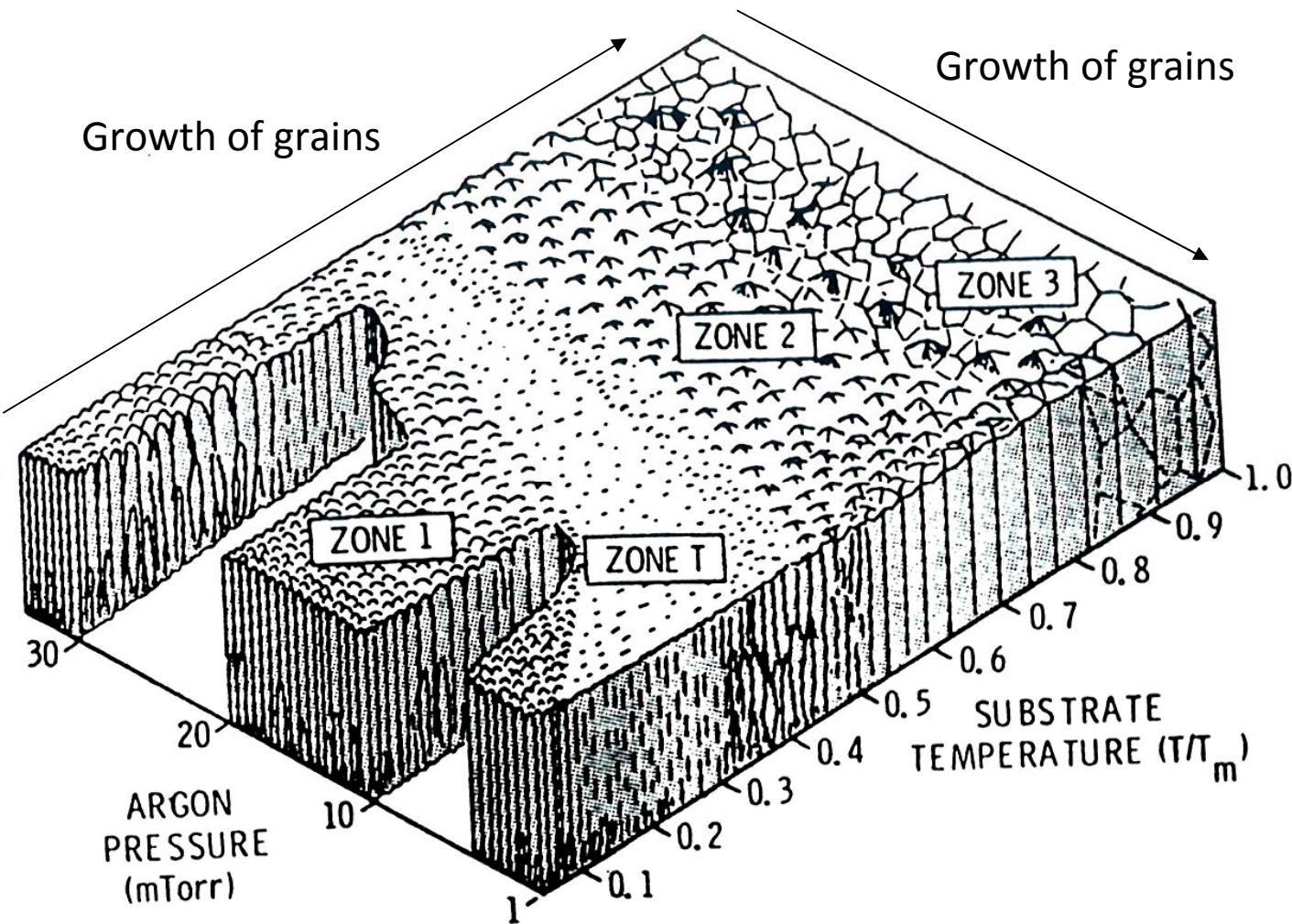
Methods for deposition

	Physical Vapor Deposition		
	Evaporation	Sputtering	PLD
Mechanism of production of depositing species	Thermal energy	Momentum transfer	Thermal energy
Deposition rate	High Up to 750,000 Å/min	Low, except for pure metals (Cu:10000 Å/min)	Moderate
Deposition species	Atoms and ions	Atoms and ions	Atoms, ions and clusters
Throwing power for a) Complex shaped Object b) Into blind hole	Poor, line of Sight Coverage Poor	Nonuniform thickness Poor	Poor Poor
Metal deposition Alloy deposition Refractory compound deposition	Yes Yes Yes	Yes Yes Yes	Yes Yes Yes
Energy of deposit-species	Low 0.1 to 0.5 eV	Can be high 1- 100 eV	Low to high
Substrate heating (external)	Normally yes	Not generally	Yes



Thornton-Zone-Diagram

Morphology of the as grown film:



Vacuum generation

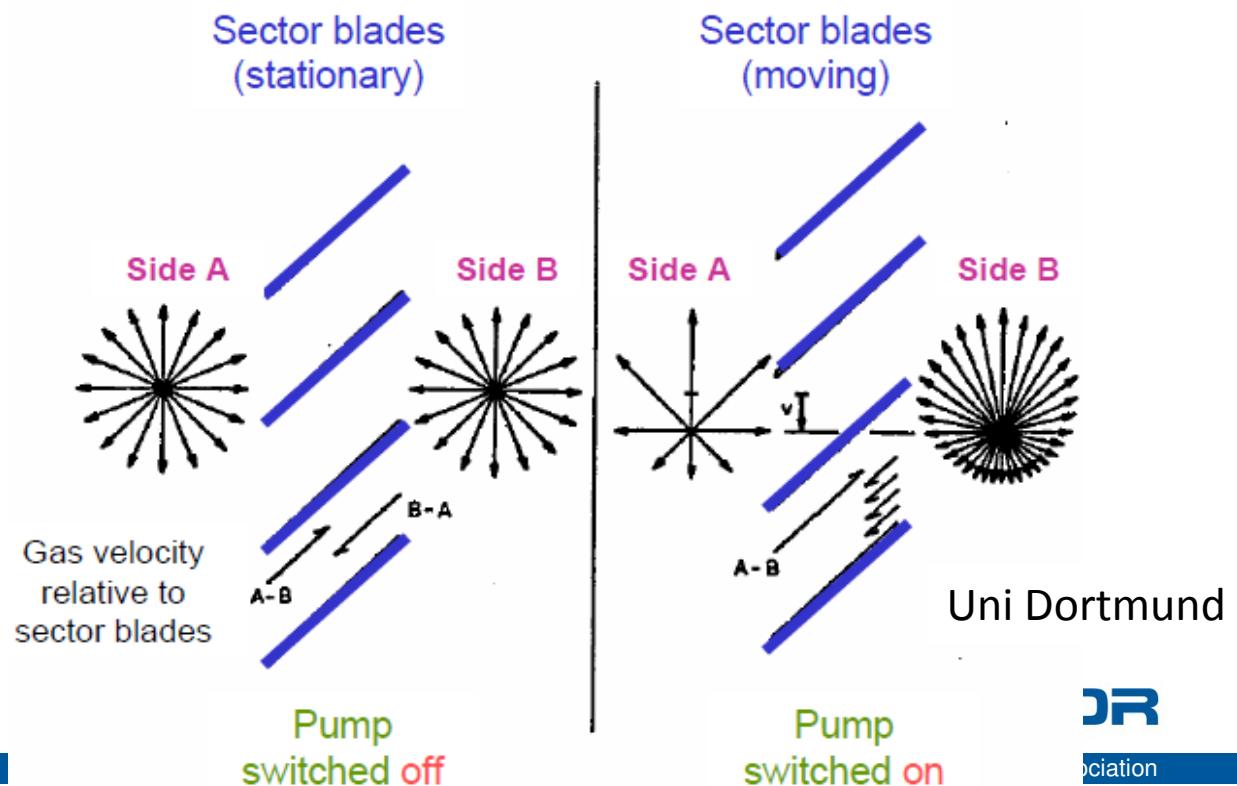
For low deposition rates, ultra high vacuum is required!

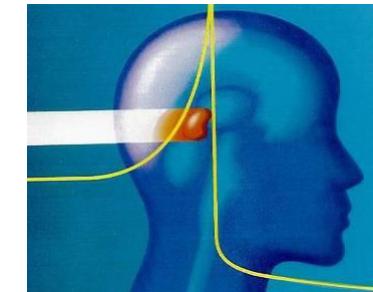
Pressure, Torr	Mean free path, cm (between collisions)	Collisions /sec (between molecules)	Molecules/(cm ² sec) (striking surface)	Monolayers /sec *
10 ⁻³	5.1	9 10 ³	3.8 10 ¹⁷	440
10 ⁻⁵	510	90	3.8 10 ¹⁵	4.4
10 ⁻⁷	5.1 10 ⁴	0.9	3.8 10 ¹³	4.4 10 ⁻²
10 ⁻⁹	5.1 10 ⁶	0.009	3.8 10 ¹¹	4.4 10 ⁻⁴

* Assuming the condensation coefficient is unity

Fore-pumps:

- Roughing-
- Membrane-
- Scroll-
- **High vacuum pumps**
- Turbomolecular-
- Ion getter-
- Titanium evaporator-
- Kryo-



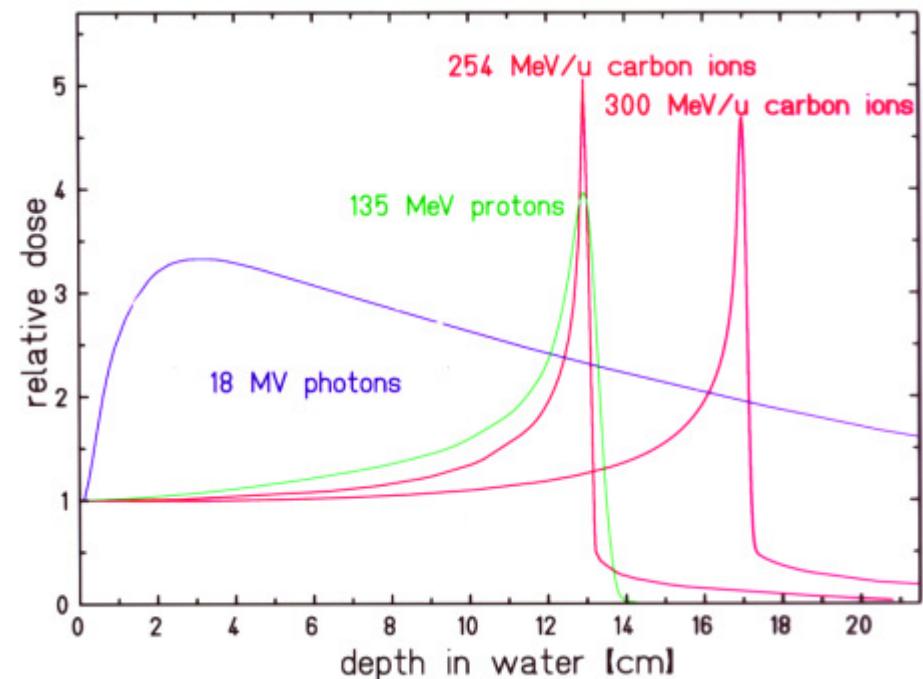
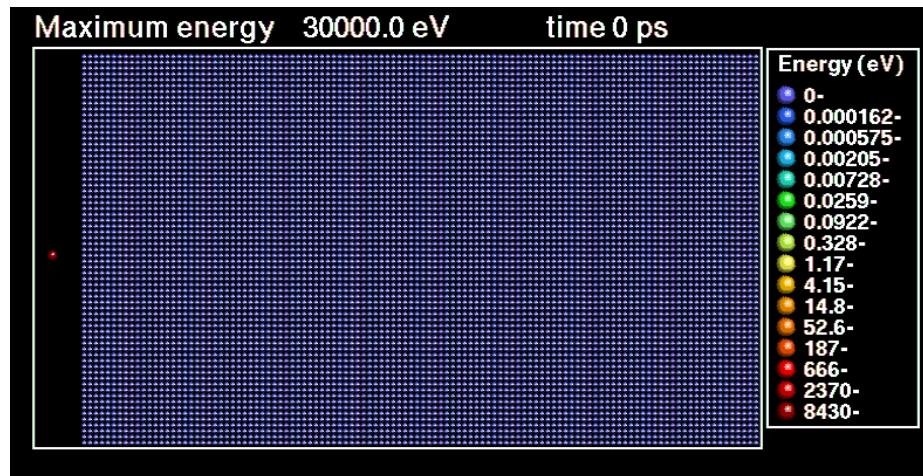


Courtesy: GSI

Ion implantation

- Doping (e.g. semiconductors)
- Synthesis of nanoparticles and buried layers
- Defect generation, amorphization and interface mixing
- Sputtering and nano-patterning
- Phase transformation and annealing
- Bio application (destruction of tumors)

30 keV Xe in Au (courtesy K. Nordlund)



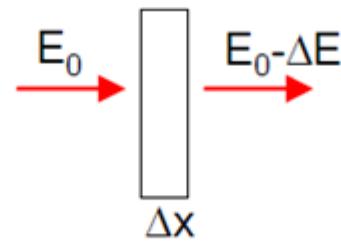


Ion-solid- interaction

Loss of energy per unit length

Stopping power:

$$B(E) = \left(\frac{dE}{dx} \right) \quad \left[\frac{eV}{nm} \right]$$



$$E_0 = \sum \Delta E = \sum \left(\frac{dE}{dx} \right) \Delta x$$

Stopping cross section: $S(E) = \frac{1}{n} \left(\frac{dE}{dx} \right) \quad [eV \cdot nm^2]$

Nuclear (low energies, high ion mass):

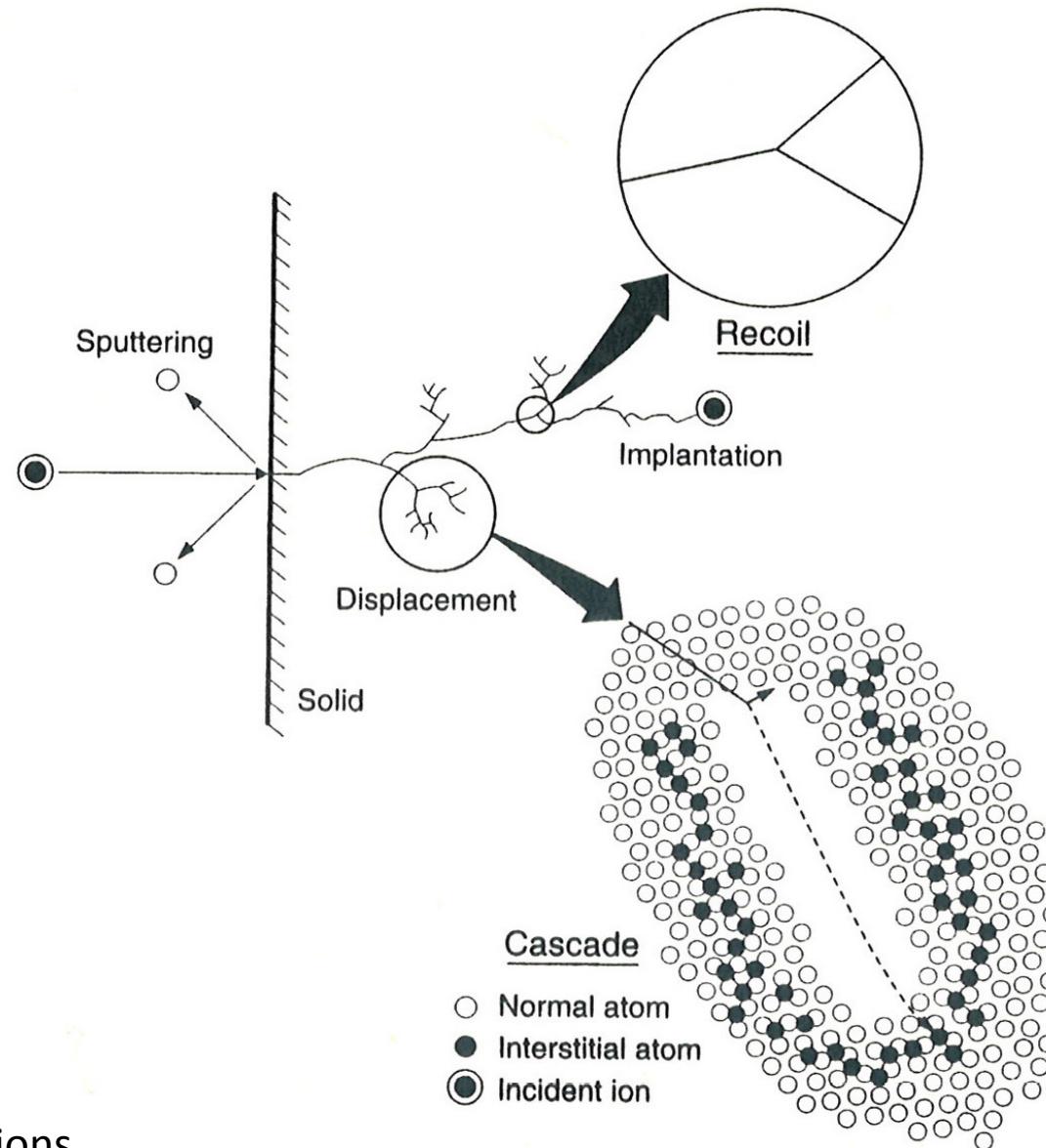
- Massive collisions
- High energy loss and angular deviation per collision
- Lattice damage (Vacancy, Interstitial)

Electronic (high energies, low ion mass):

- Negligible collision
- Low energy loss and angular deviation per collision
- Low lattice damage
- Ionisation



Ion-solid- interaction

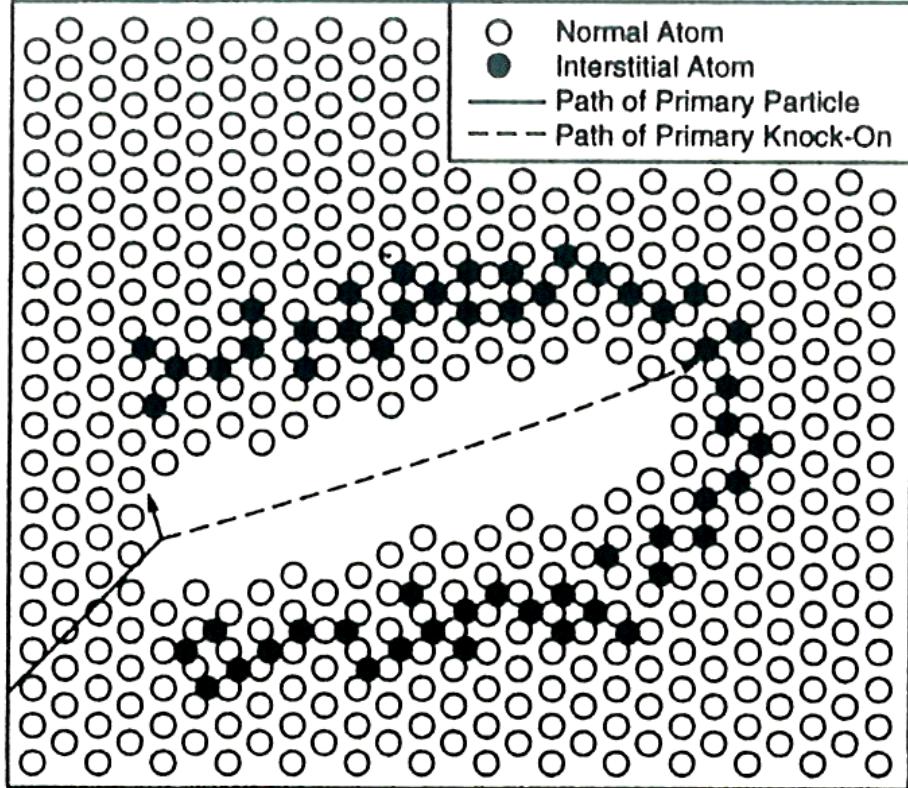


M. Nastasi
Ion-Solid Interactions
2004

Spikes

Displacement-Spike

High density cascade of moving atoms
Inside a certain volume



Spikes: Mean free path l_d between the collisions
Reaches the lattice constant

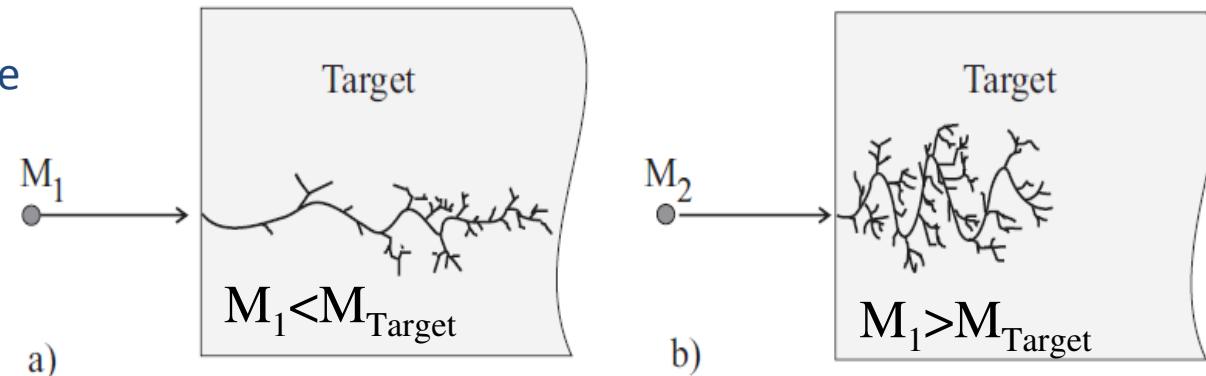
Thermal Spike

At the end of the displacement spike → Atoms have too low energy to generate
Displacements and dissipate the residual energy via phonons (heat). $t_{\text{thermal}} \sim 10^{-12} \text{ s}$

Defects

(aus Diss. S. Eichler, Halle 1997)

Displacement cascade

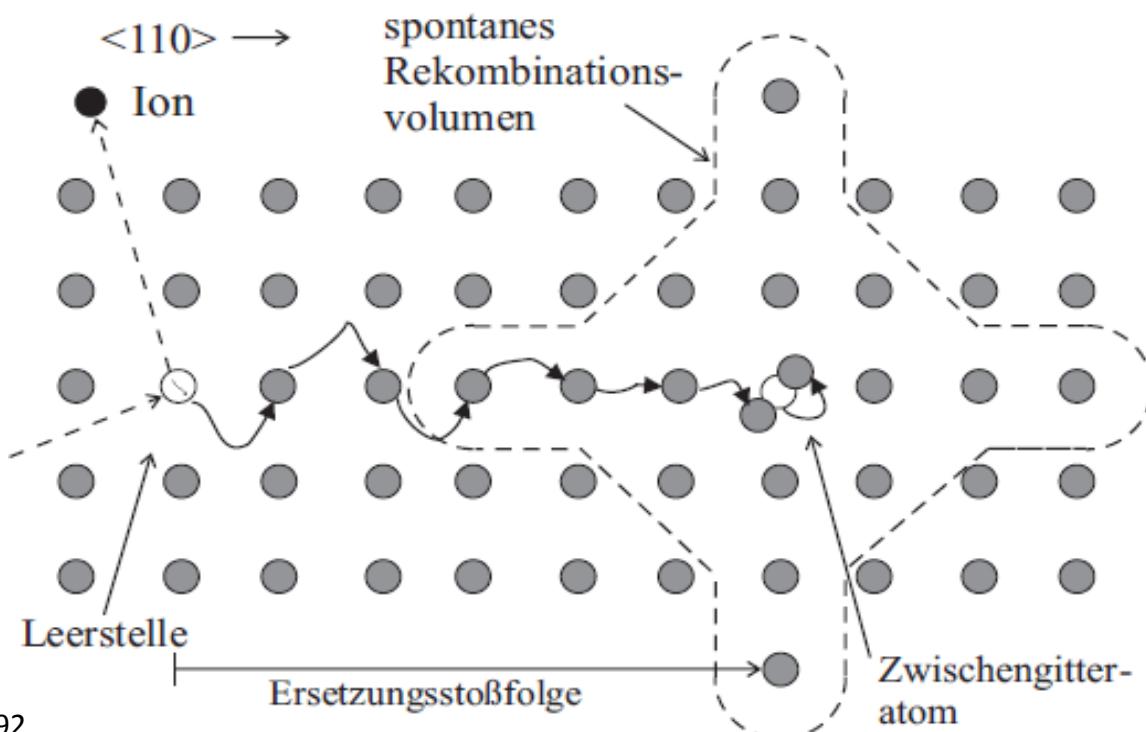


H. Ryssel, 1978

Pointdefects

- Vakancies
- Interstitial atoms
- = Frenkel-Pairs

Can be energetically
Instable and transform into
defect complexes



Bergmann/Schäfer 1992

Defects

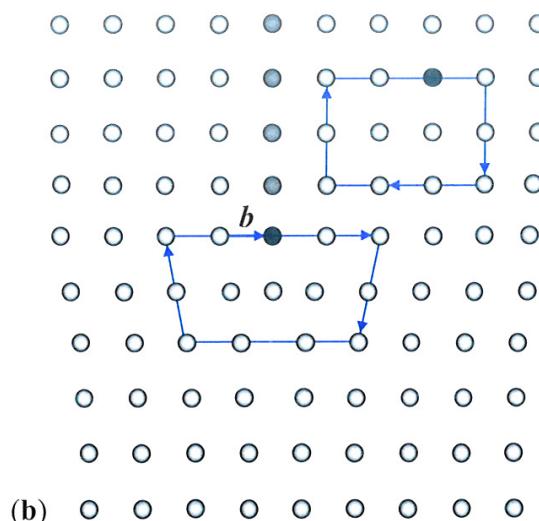
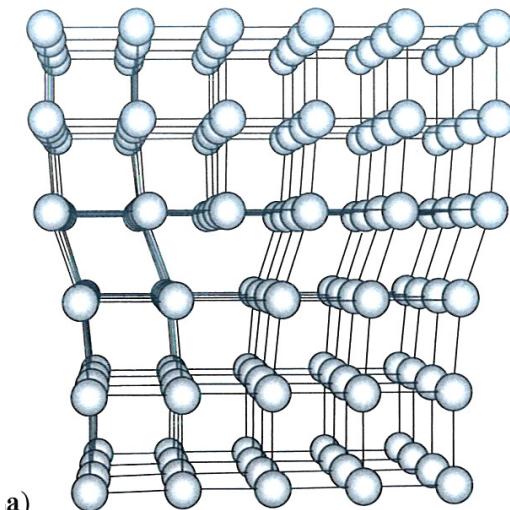
Defect complex

- Point defect + Point defect
- Point defect + Target atom

Example: Oxygen in Si →

Extended defects

- Dislocations
- Grain boundaries



Defekt	Ausheiltemperatur (isochron)
VO	570-670 K
VO ₂	770 K
V ₂ O	620 K
V ₃ O	670 K
V ₂ O ₂	670 K
V ₃ O ₃	870 K

e.g. added lattice plane
b=Burgers-vector

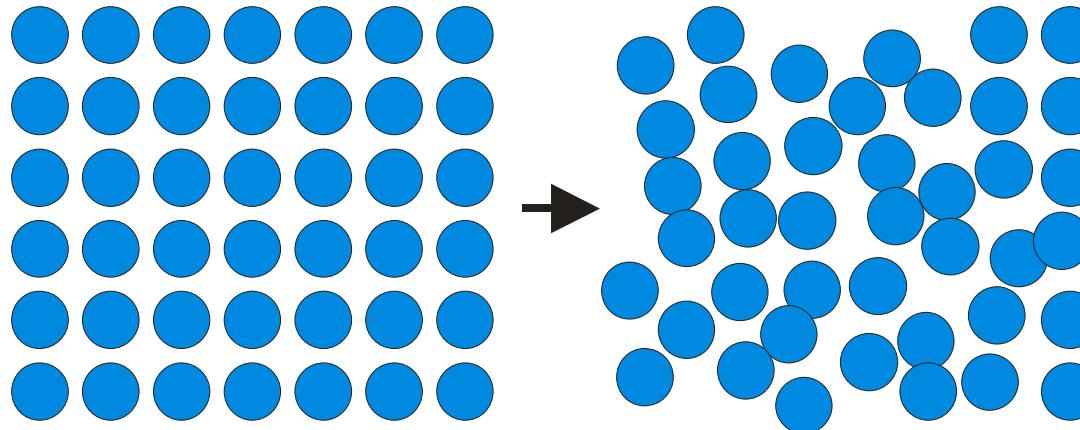
S. Hunklinger,
"Festkörperphysik", 2007





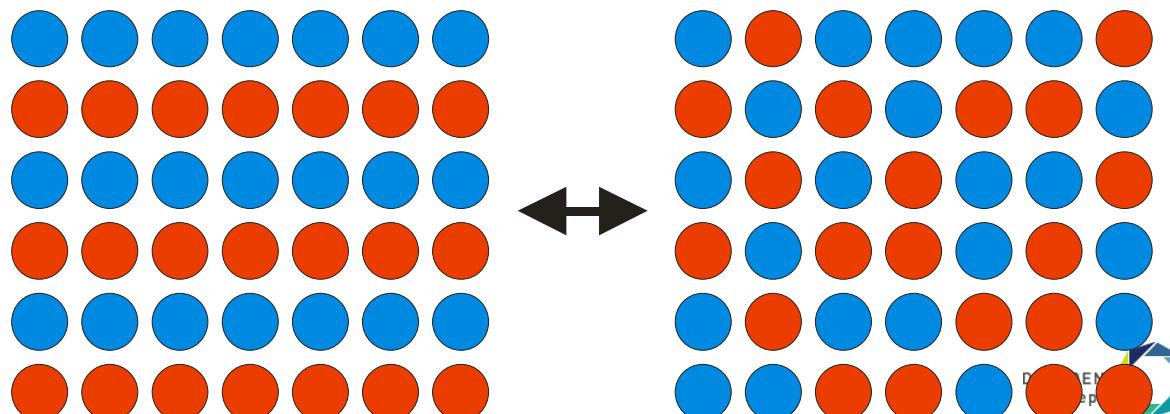
Defects

Amorphisation



Amorphous materials have dangling bonds
→ Passivation of Si with H

Chemical disorder
(Multi layers, alloys)



e.g. ferro/para FeAl, FeRh, fcc-fct FePt

Doping by ion implantation

Implant doping (in semiconductor industries)

- **Step1: Implantation**
- **Step2: Annealing for defect removal**

Problems for DMS:

- **Cluster buildup due to supersaturation**
- **Defects might be helpful for coupling**

Spintronics – Diluted Magnetic Semiconductors

Diluted magnetic semiconductors: Lets drive the DEFECT screw!

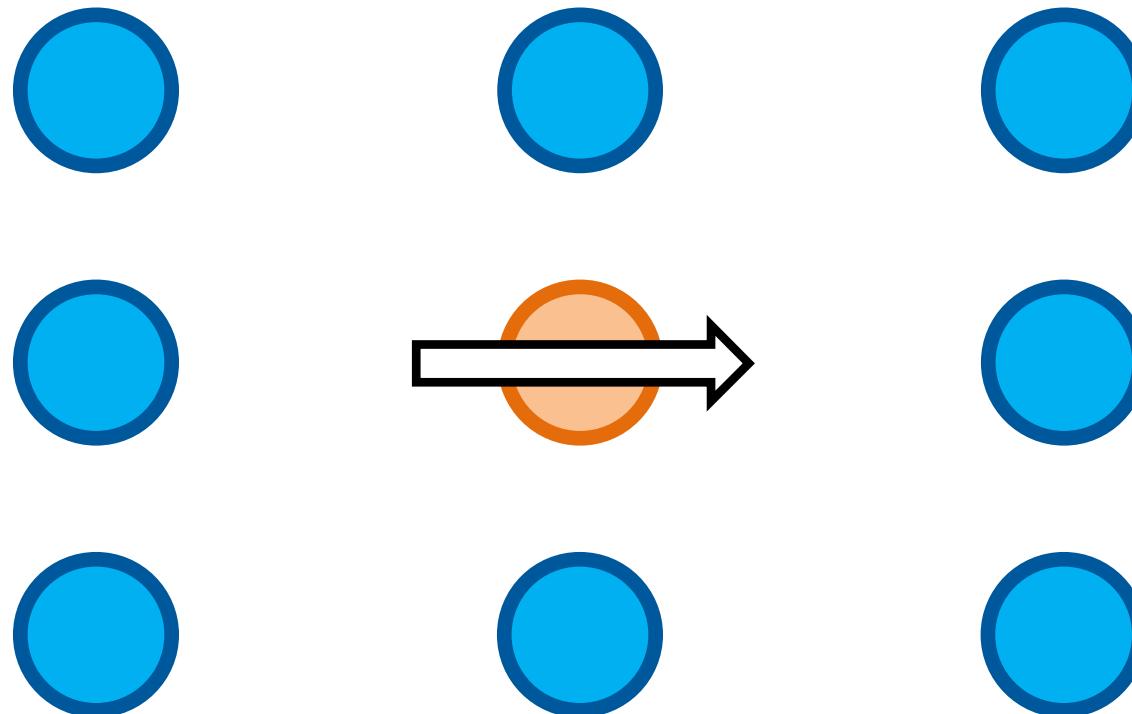
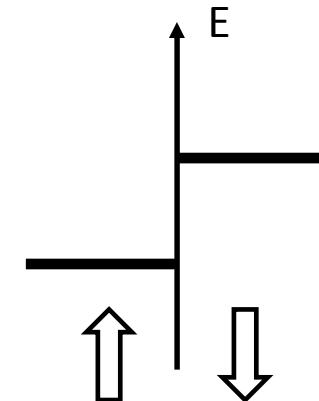




Spintronics – Diluted Magnetic Semiconductors

Driving the screw

- Paramagnet



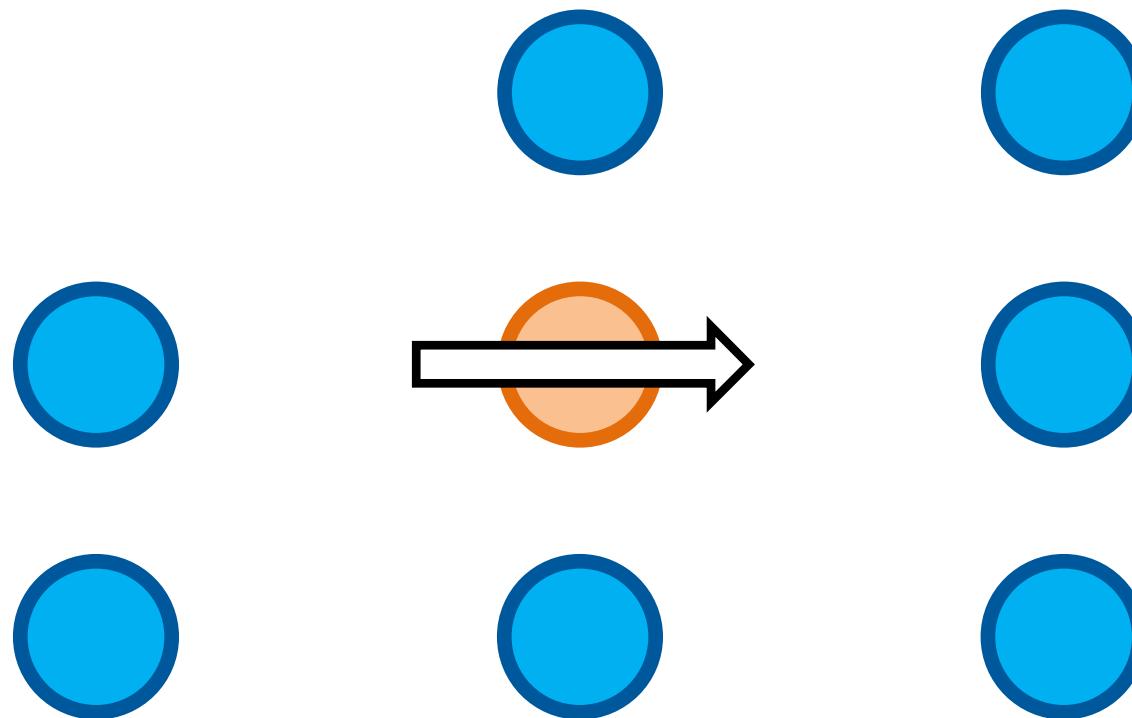
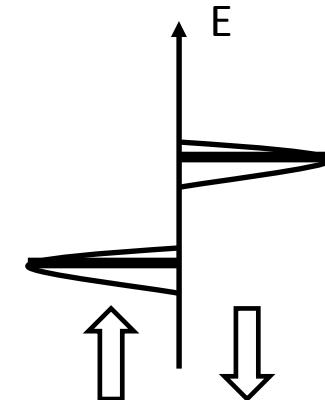
Local picture

page 71

Spintronics – Diluted Magnetic Semiconductors

Driving the screw

- Paramagnet



Local picture

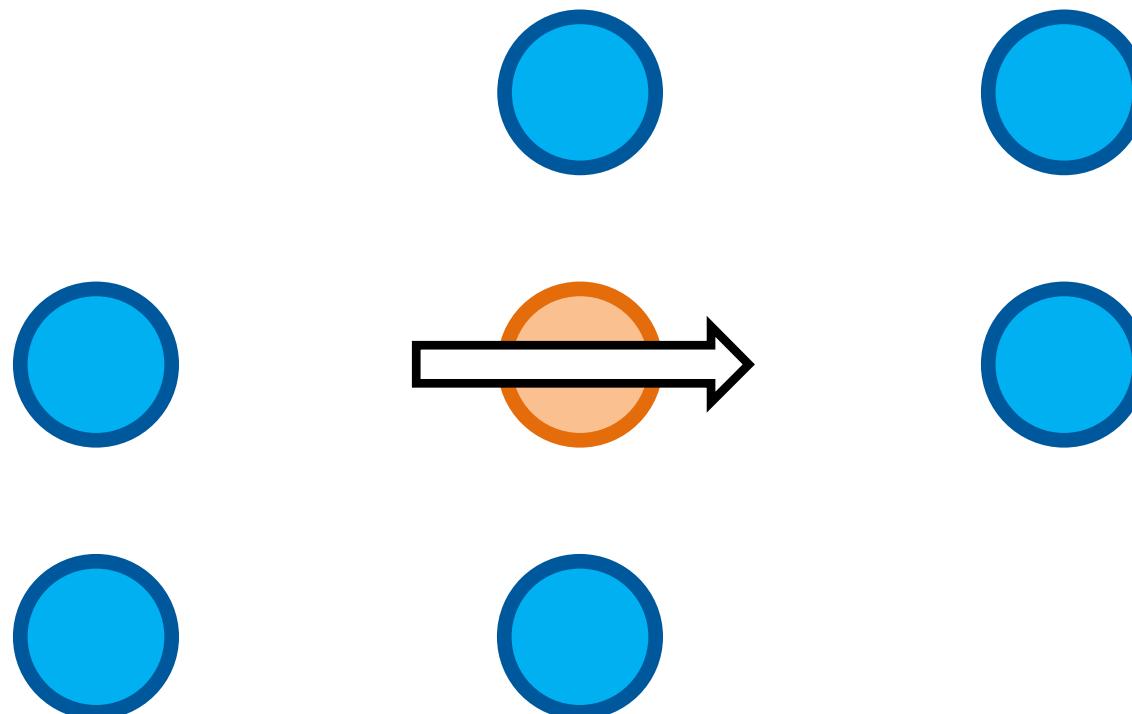
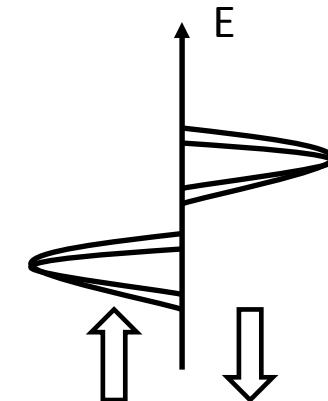
page 72



Spintronics – Diluted Magnetic Semiconductors

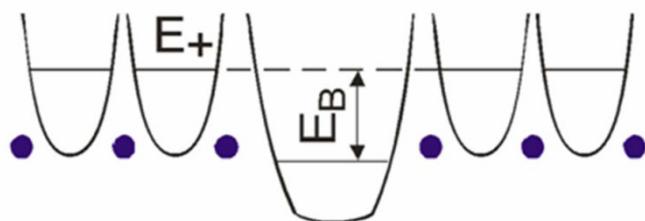
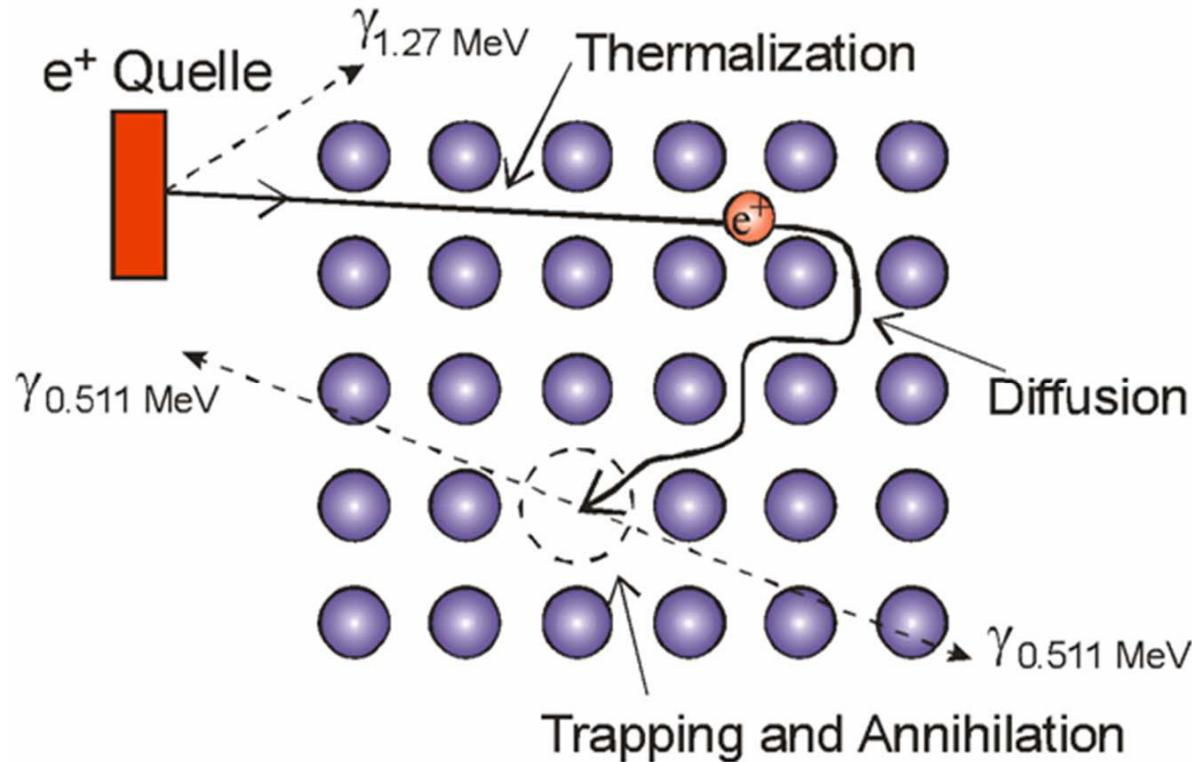
Driving the screw

- Ferromagnet



Defects in TiO₂

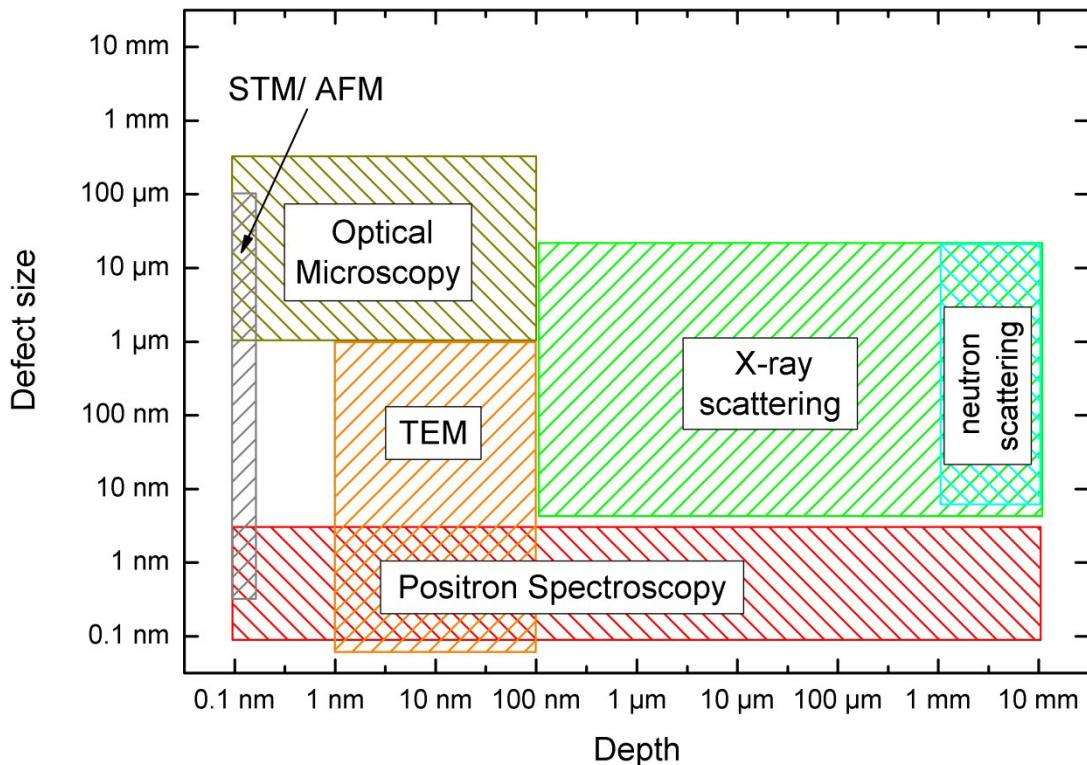
Why Positrons?



Courtesy R. Krause-Rehberg

- Positron @ the attractive defect potential
→ positron lifetime increases in a vacancy
 - Doppler broadening decreases in open volume defect
 - Coincidence spectrum depends on local chemistry
- Defect identification and quantification possible

Defects in TiO_2



Positron Spectroscopy:

non-destructive method for material investigation and characterization of atomic defects

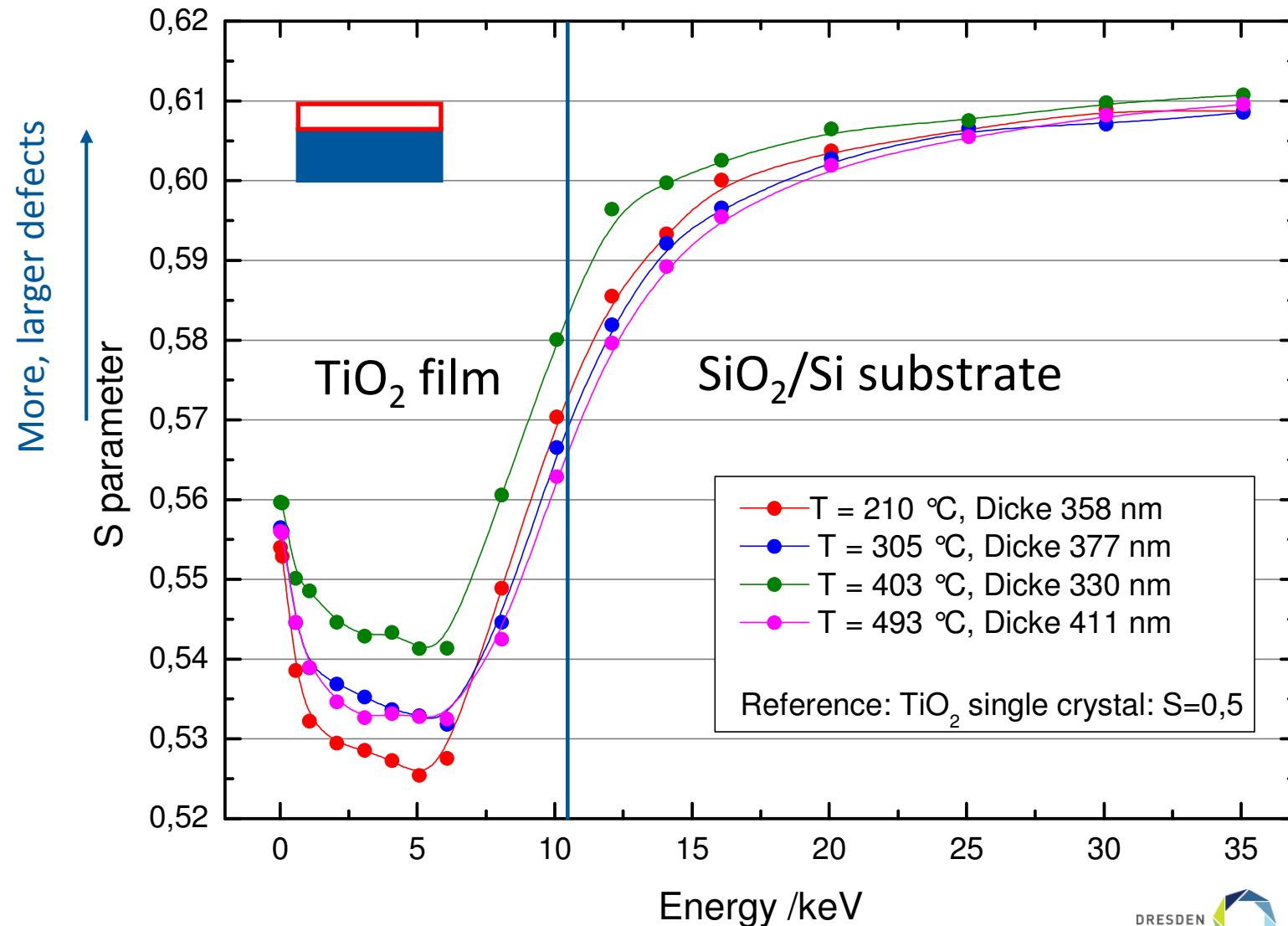
sensitivity is much higher for small defect sizes than standard techniques

STM ... Scanning Tunneling Microscope, AFM ... Atomic Force Microscope, TEM ... Transmission Electron Microscope



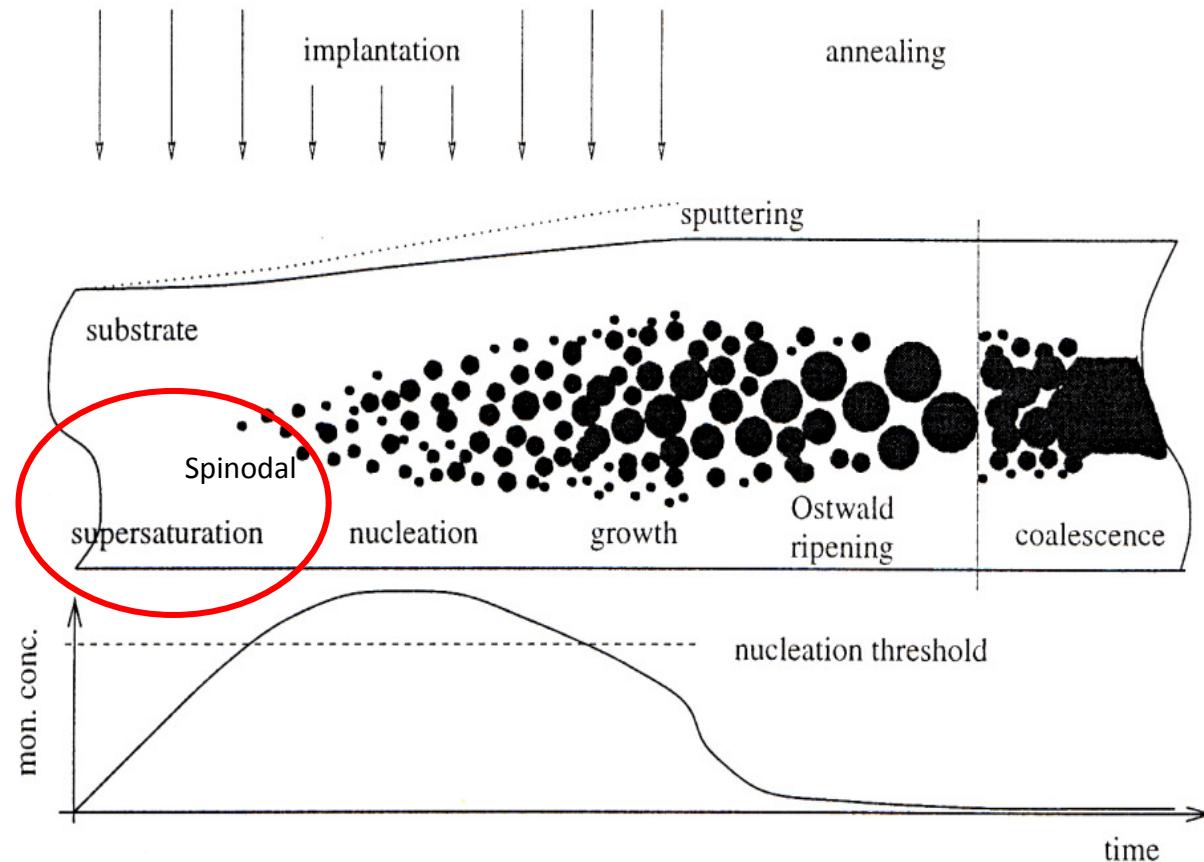
Defects in TiO_2

Positron annihilation spectroscopy



Example Supersaturation by Fe ion implantation

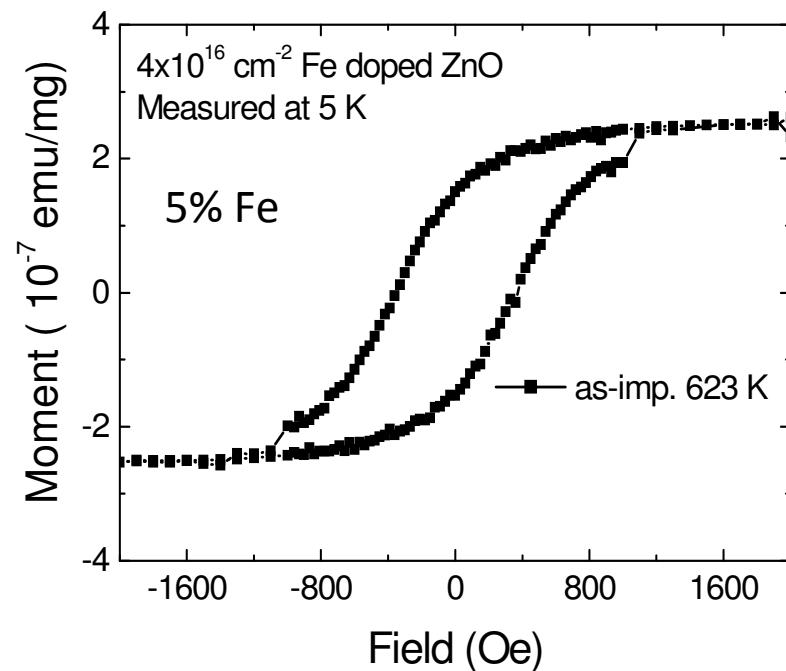
Problem: Cluster buildup due to ion implantation



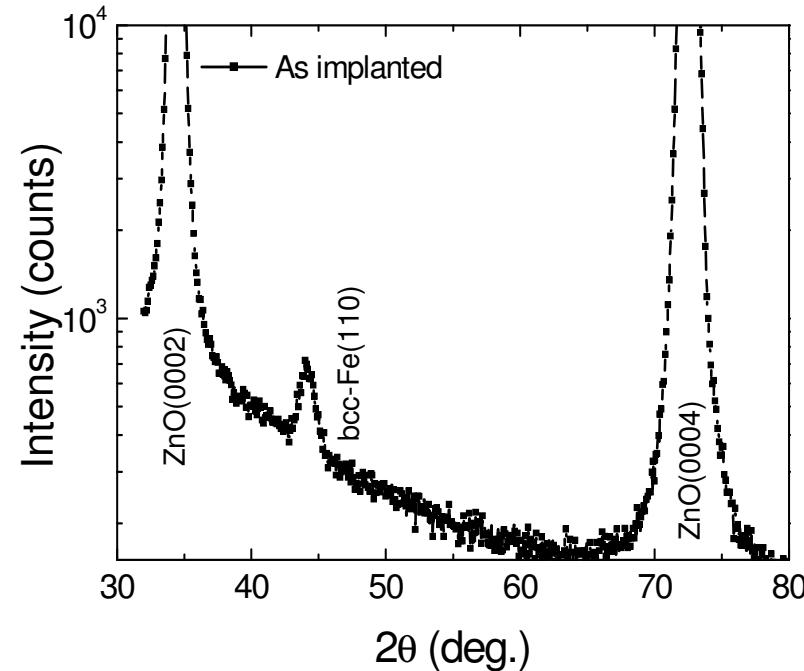


Cluster formation upon implantation

SQUID

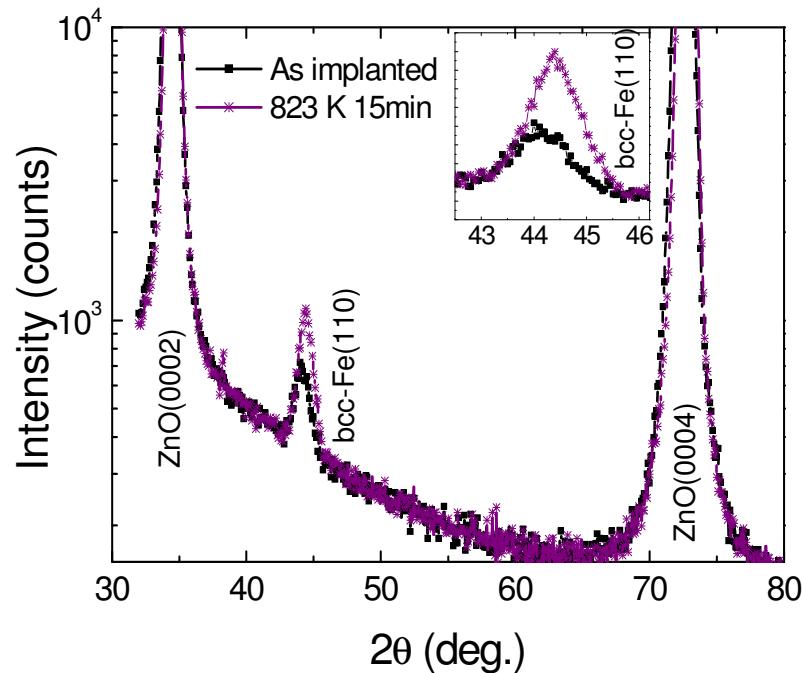
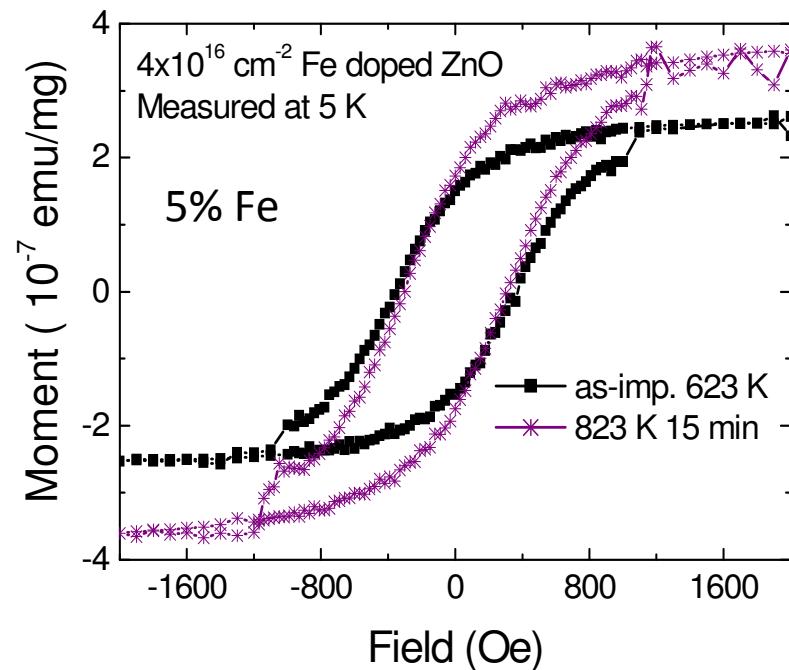


Synchrotron-XRD (ROBL)



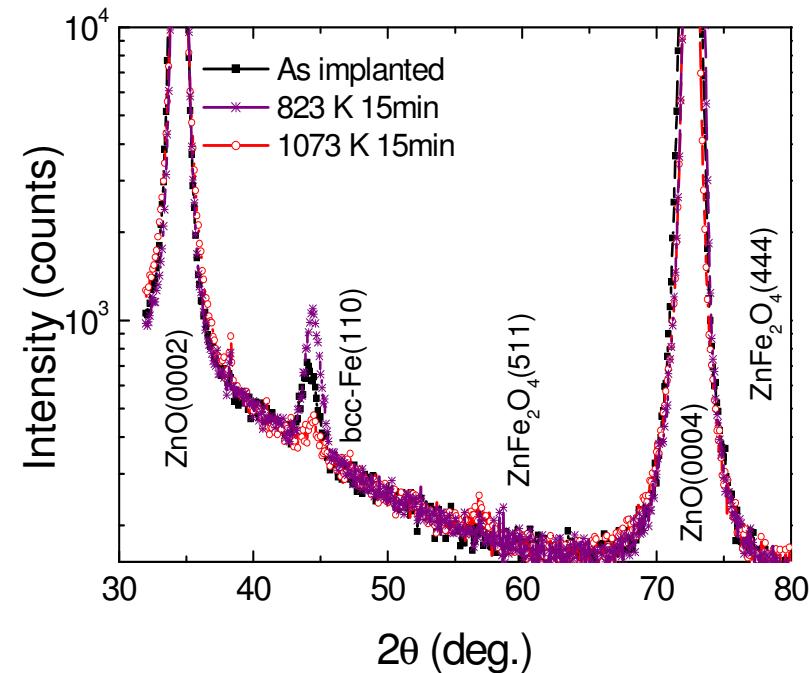
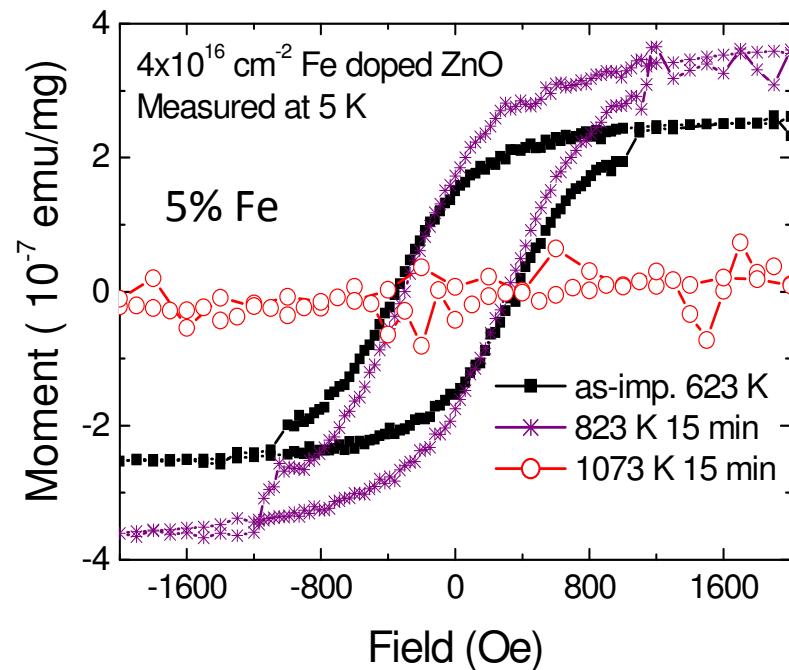


Cluster formation upon implantation



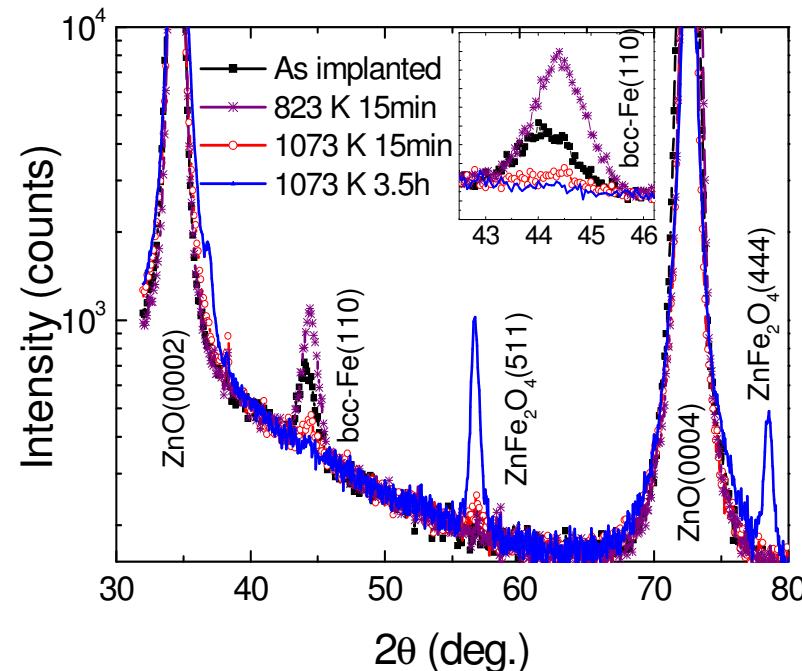
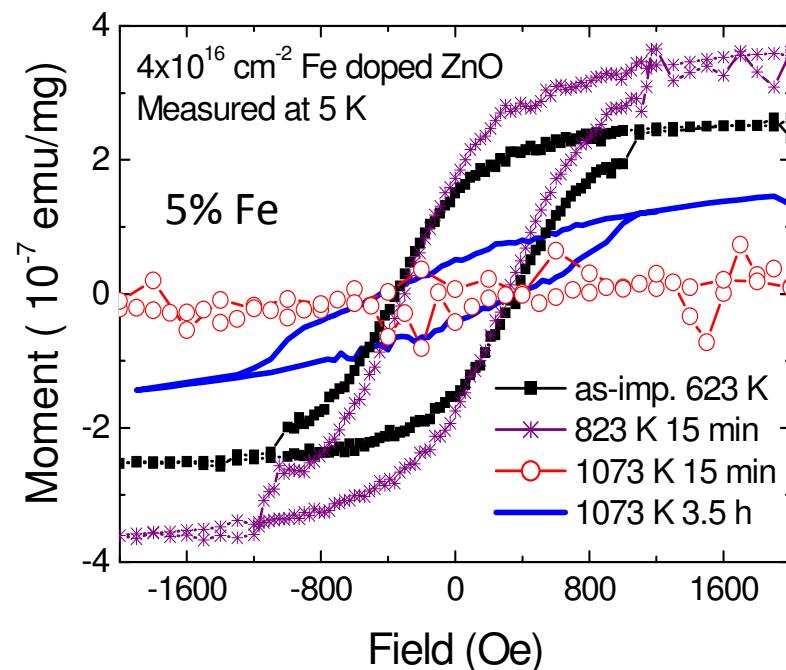


Cluster formation upon implantation





Cluster formation upon implantation



- Low annealing temperature T_A : bcc Fe
- High T_A and short time : no crystalline secondary phases detected
- High T_A and long time : ZnFe₂O₄

J. Phys. D-Appl. Phys. 40, 964, 2007

The DETI.2 project

Alevtina Smekhova
Spokesperson in Moscow



Kay Potzger
Spokesperson in Dresden



Thank you!

Master students welcome!