Disorder and Frustration Effects on Ferromagnetism in Diluted, Magnetic Semiconductors

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Outline

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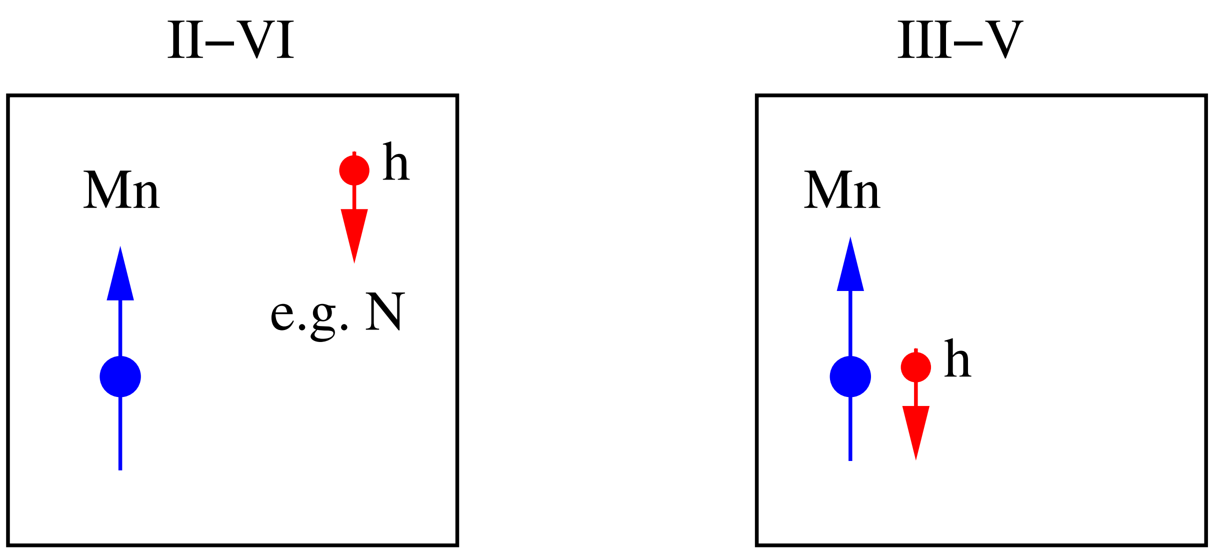
Introduction

DMS: Semiconductor doped with a magnetic ion, e.g. Mn, Fe

- II-VI semiconductors, e.g. (Cd,Mn)Te — typically Spin glasses
- III-V semiconductors, e.g. (Ga,Mn)As, (Ga,Mn)N — typically ferromagnets

Mn : half-filled $3d$ shell – spin-$\frac{5}{2}$ local moment

- II-VI: Mn substitutes for group II element, Mn^{2+} is same as group II element
  – need another dopant, e.g. P to introduce carriers.
- III-V: Mn substitutes for group III element and provides a hole – magnetic ion
  + acceptor.



$\mathbf{Ga_{1-x}Mn_xAs}$

Physical Properties :

- Ferromagnetic, $T_c = 173$ K for $x \simeq 0.08$ (Wang *et al.*, 2004)

- Large compensation – hole concentration less than Mn concentration.

- Grown with MBE – both random alloy and digital layers (also ion implantation)

Technological possibilities :

Spintronics – hopes of combining magnetic functionality with existing semiconductor technology, to use the spin as well as the charge of the electron

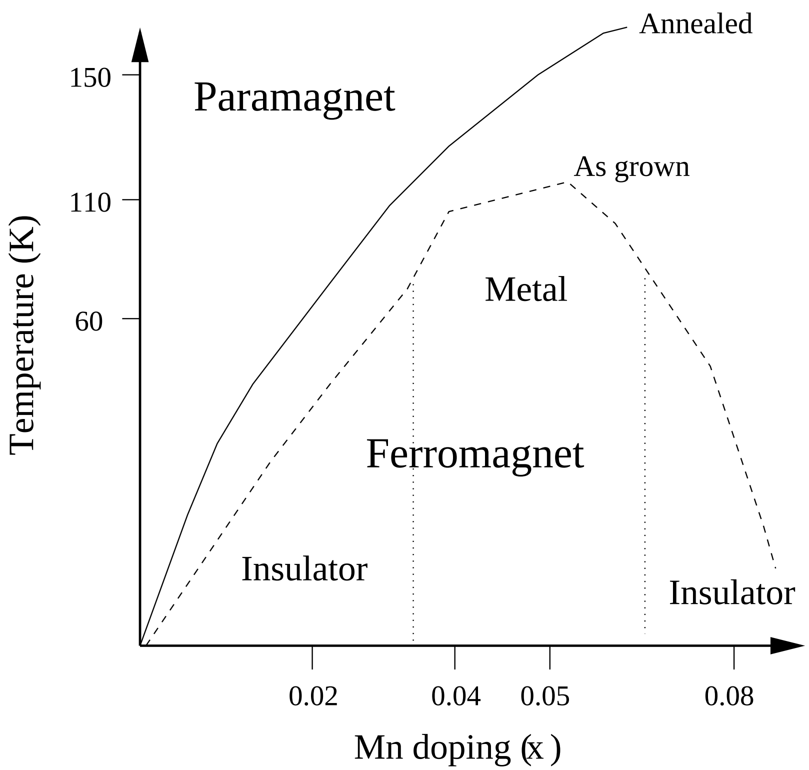
- Possibility of materials with tuned optical, magnetic and electronic properties

- Demonstrated applications – gate controlled ferromagnetism (Ohno, 2001)

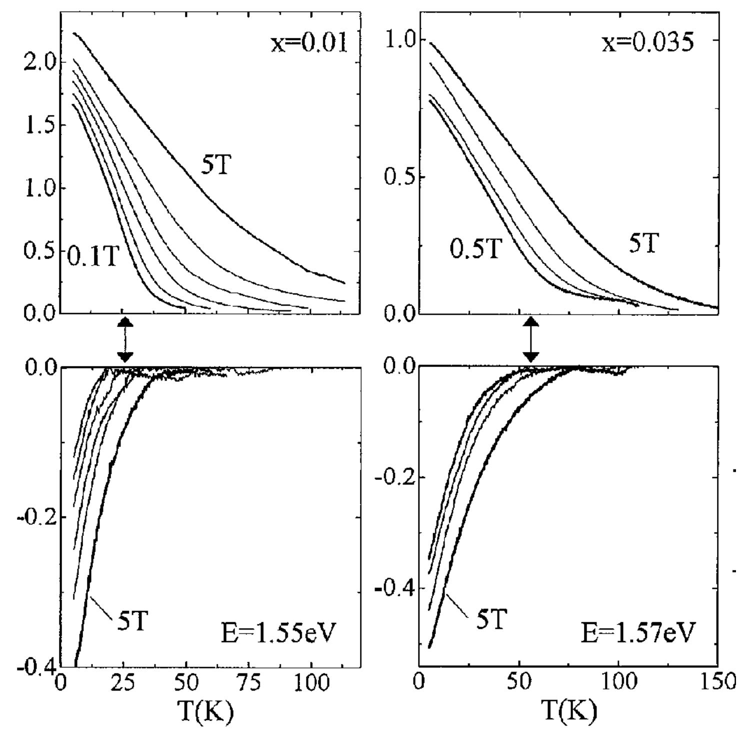
- Spin filters – materials to provide spin polarized currents

Experiments

Phase Diagram :
(low doping)



Unusual Magnetization curves

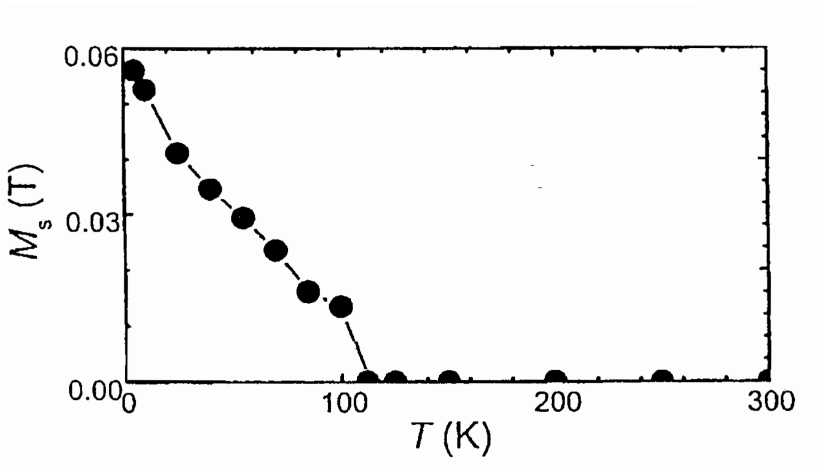


(Beschoten *et al.*, PRL **83**, 3073 (1999))

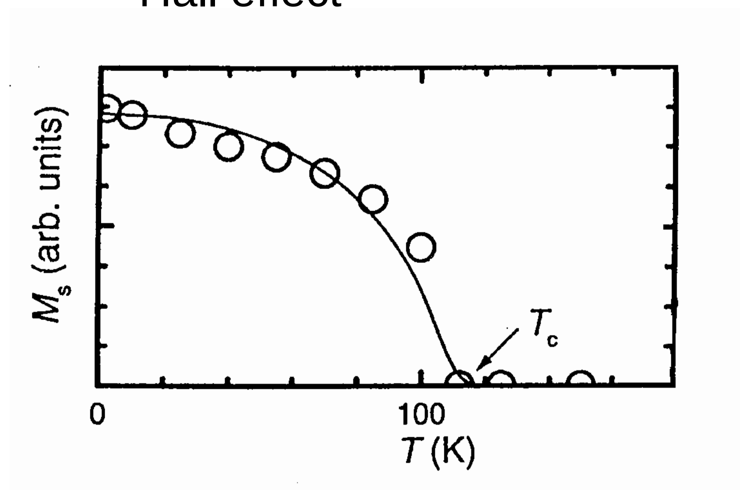
Experiments

Magnetization : Metallic Phase

SQUID measurement



Hall effect



Observation:

- Low temperature magnetization does not saturate in most samples

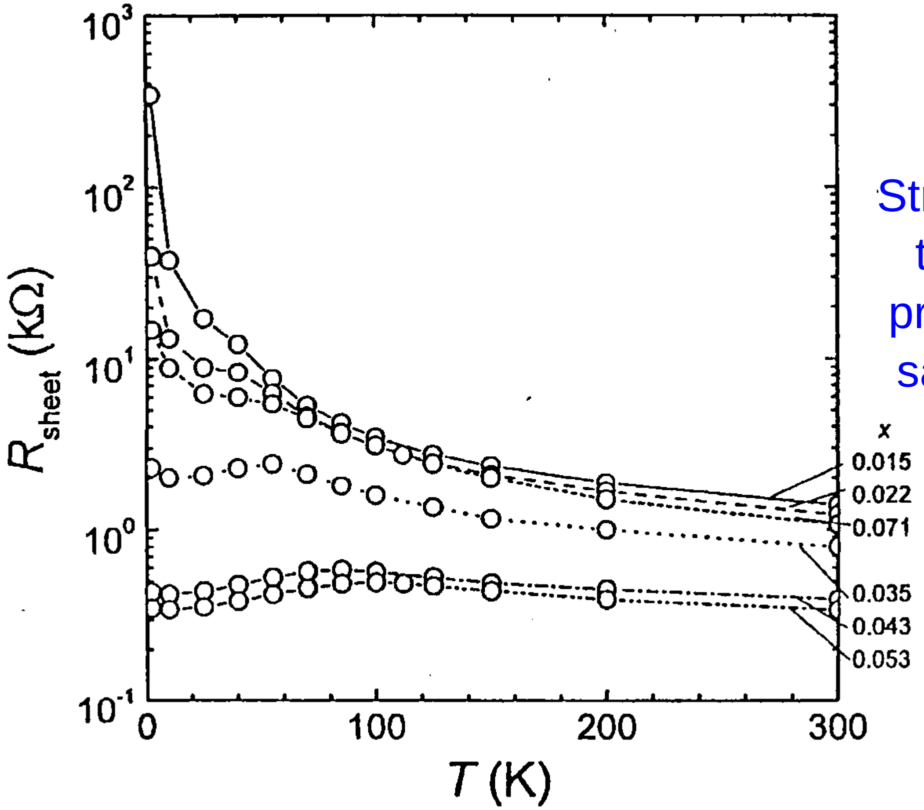
Possible Explanations:

- Paramagnetic/magnetically inert population of Mn spins (e.g. Mn interstitials)
- Anisotropic spin interactions
- Effective anti-ferromagnetic exchange interactions between Mn spins

Experiments

Resistivity

- Metal-Insulator transition with increasing Mn concentration



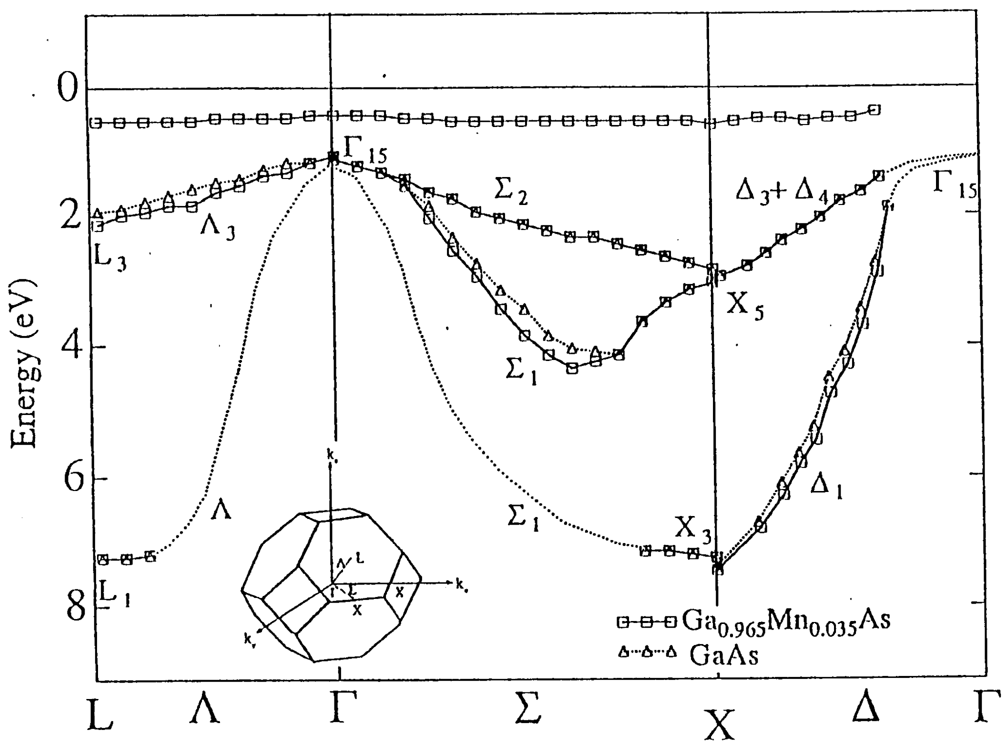
Strong correlation between transport and magnetic properties – most metallic samples have highest T_c.

(Ohno, J. Magn. Magn. Mat., **200**, 110 (1999))

Experiments

Impurity Band

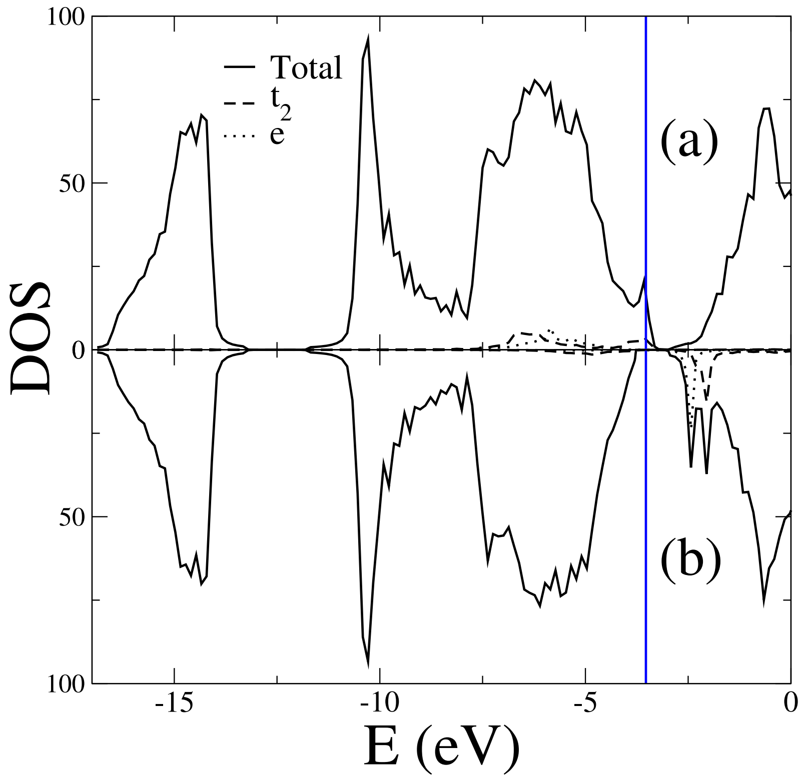
- Expectation of Mn impurity band for low Mn concentration
- Impurity band seen in ARPES, Photoconductivity experiments:



(Okabayashi *et al.*, Physica E **10**, 192 (2001))

Density Functional Theory

- Indication of impurity band near Fermi energy at $x = 0.03$
- (a) Majority spin DOS, (b) Minority spin DOS



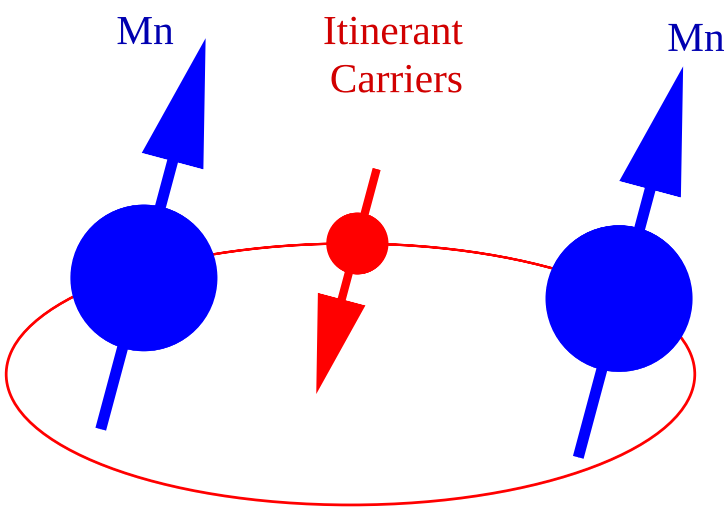
Question of transport mechanism – hopping of localized holes, or delocalized valence band holes, or somewhere in between?

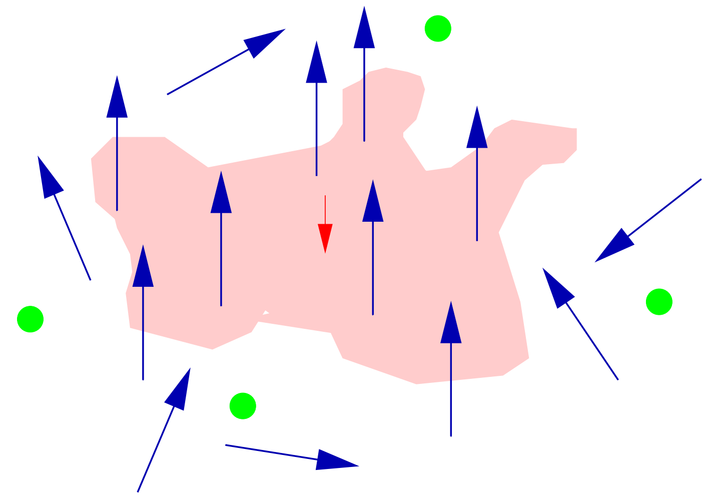
(Sanvito, Ordejon and Hill, Phys. Rev. B **63**, 165206 (2001))

Microscopic picture

Basic mechanism for ferromagnetism:

Hole -Mn antiferromagnetic exchange





Relevant Physics:

- Hole kinetic energy
- Hole -Mn antiferromagnetic exchange
- Hole - Mn coulomb interaction
- Positional disorder in Mn ions
- Hole-hole coulomb interaction
- Mn-Mn antiferromagnetic exchange
- Hole spin-orbit interaction
- Compensation Mechanisms:
  As antisites, Mn interstitials

Impurity band model

Hamiltonian:

$$\mathcal{H} = \Sigma_{ij} t_{ij} c^{\dagger}_{i\sigma} c_{j\sigma} + \frac{1}{2} \Sigma_{ij} J_{ij} \mathbf{S}_i \cdot (c^{\dagger}_{j\alpha} \boldsymbol{\sigma}_{\alpha\beta} c_{j\beta})$$

- Hopping between impurity sites
- Antiferromagnetic exchange between carriers and Mn spins
- Appropriate for insulating phase and around metal-insulator transition

Aim : Study effects of impurity potential + positional disorder of Mn spins

Approximations:

- Hydrogen-like 1s impurity orbitals ($\phi(r) \sim e^{-r/a_B}$) with $a_B = 7.8$ Å

$$t(r = |\mathbf{R}_i - \mathbf{R}_j|) \; = \; 2 \left(1 + \frac{r}{a_B}\right) e^{-r/a_B} \, \mathrm{Ry}$$
$$J(r = |\mathbf{R}_i - \mathbf{R}_j|) \; = \; J_0 \, e^{-2r/a_B}$$

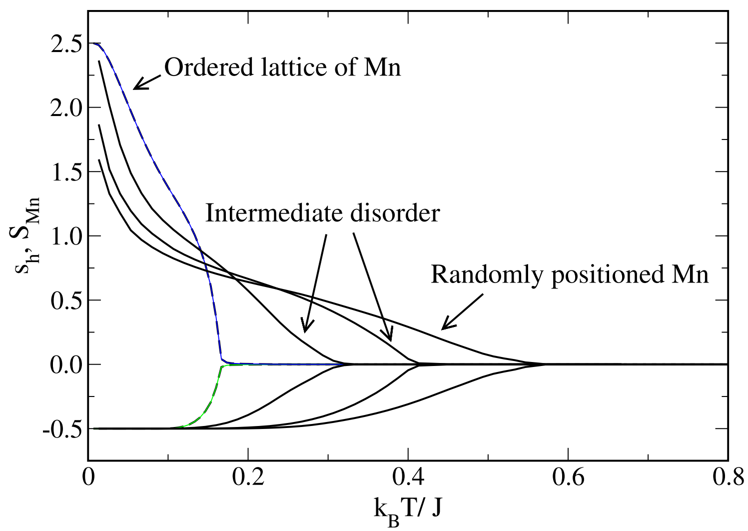
- Mn substitute on Ga FCC sublattice, localized Mn spins, classical spins
- No spin-orbit coupling – “electron” formalism, no on-site disorder
- neglect higher energy delocalized states in valence band
- $x = \frac{N_{Mn}}{4N^3}$ – Mn concentration, px – carrier concentration
- Exchange J_0 = 0.133 Ry (Bhattacharjee and Guillaume, 2000)

Mean Field Results

Mean field factorization: spins align in z direction – no temporal fluctuations

$$\mathbf{S}_i \cdot \mathbf{s}_j = \langle S_i^z \rangle s_j^z + S_i^z \langle s_j^z \rangle - \langle S_i^z \rangle \langle s_j^z \rangle$$

- Mean field study suggested that disorder can significantly enhance T_c



(Berciu and Bhatt, PRL **87**, 107203 (2001))

- Unusual magnetization curves (concave upward).
- Wide distribution of local charge densities
- Mean Field studies generally overestimate T_c – neglect temporal fluctuations.
- **How robust are the results of the mean field study?**

Monte Carlo simulations

Numerical Issues :

- Fermions coupled to classical spins – need to diagonalize the fermion problem for each spin flip
- Grand canonical ensemble for many different realizations of disorder (need to determine μ for each sample)

Numerical scheme :

- Perturbative Monte Carlo – make small spin rotations, use perturbation theory to update new values of carrier eigenvalues.
- Truncate Hilbert space

Sizes :

- $L \times L \times L$, with $L = 7$ to 15, 40 to 135 spins, 4 to 21 carriers
- 30 - 700 samples depending on temperature and x, p

$$x = 0.01, \begin{cases} p = 0.1 \\ p = 0.3 \end{cases}, \qquad x = 0.03, \begin{cases} p = 0.1 \\ p = 0.3 \end{cases}$$

Finite Size scaling : Binder Cumulant – independent of L at T_c

$$G(L) = \frac{1}{2}\left(5 - 3\frac{\langle M^4 \rangle}{\langle M^2 \rangle^2}\right)$$

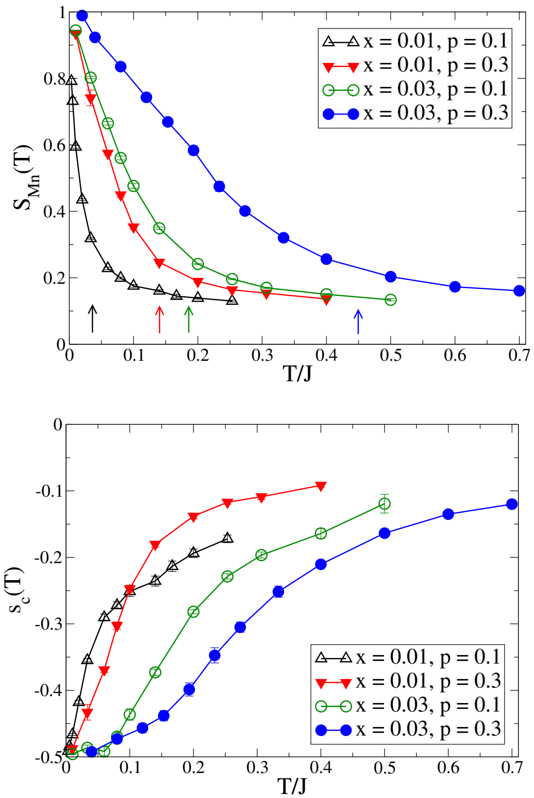
Magnetization

Impurity band model for (Ga,Mn)As (Berciu and Bhatt, 2001)

• Most appropriate for low Mn concentrations, high compensation:

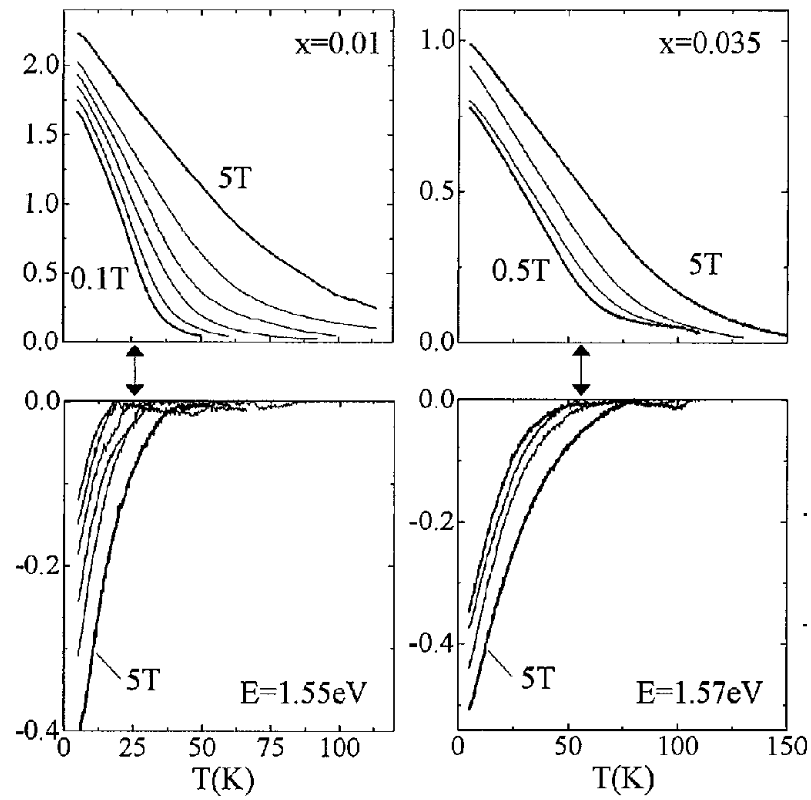
MC Simulations

(Kennett *et al.*, 2002)



Experiment

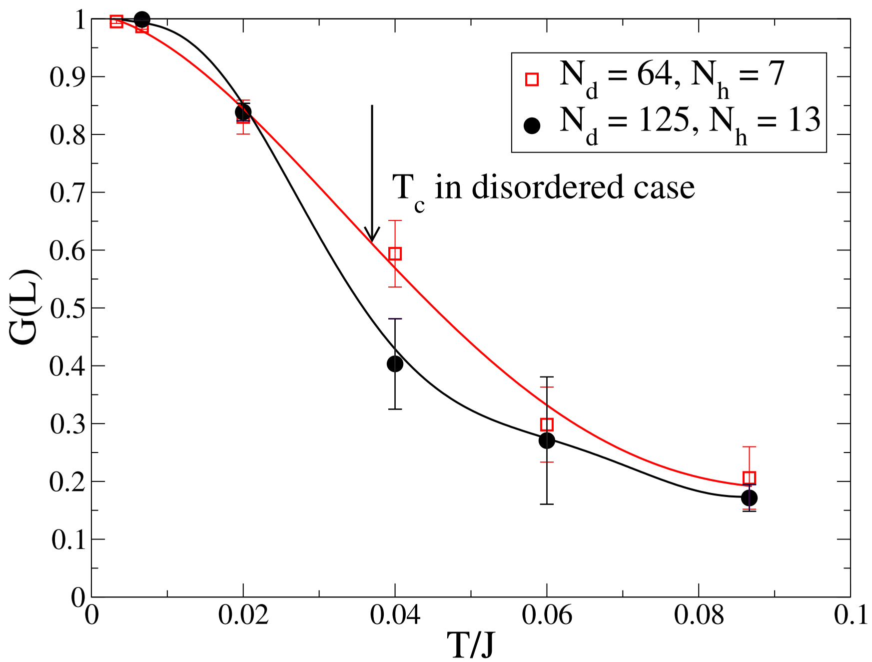
(Beschoten *et al.*, 1999)



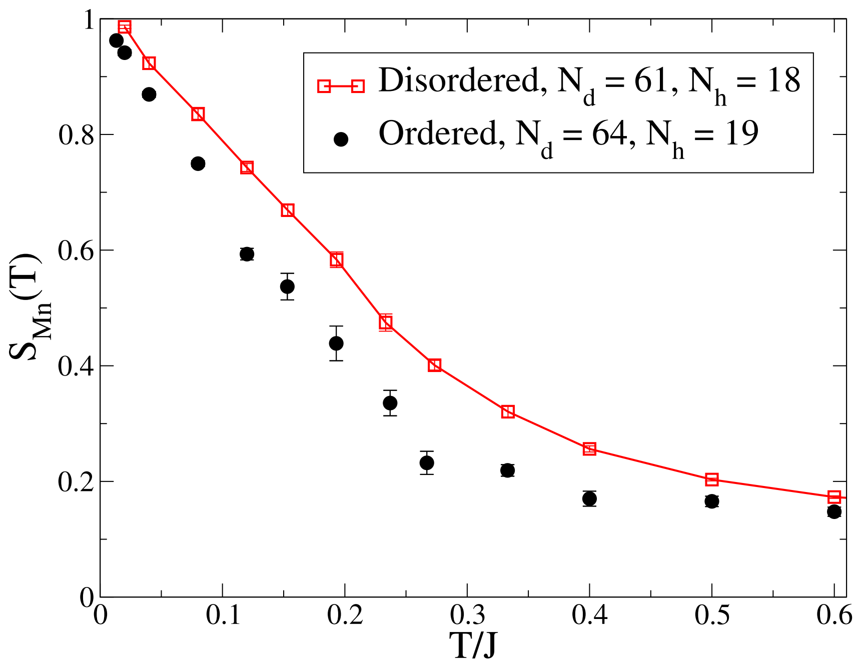
Mn and carrier magnetization

Order vs Disorder

Binder Cumulant :
$x = 0.01$, $p = 0.1$ Weak Disorder



Magnetization :
$x = 0.03$, $p = 0.3$

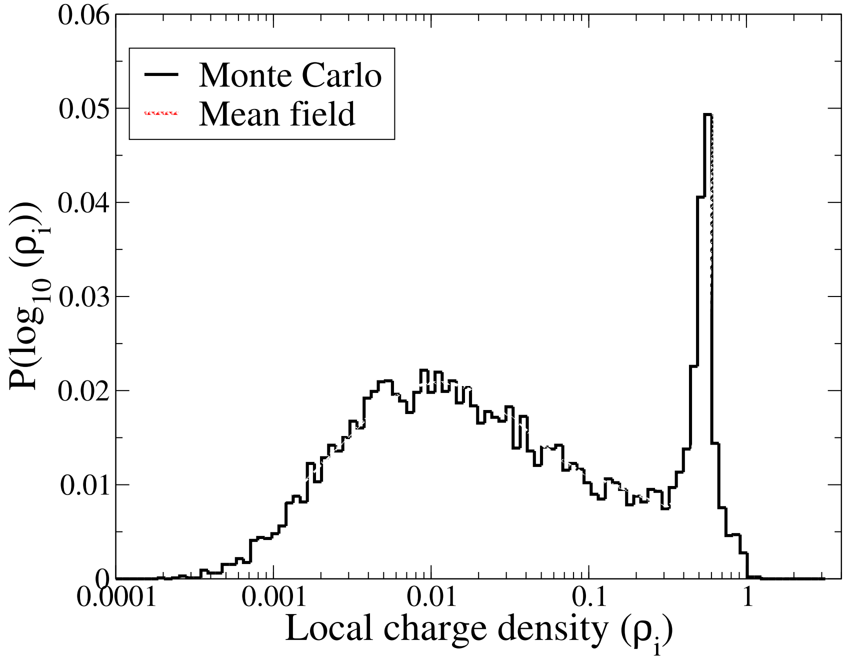


- Disorder enhances T_c even when fluctuations are included

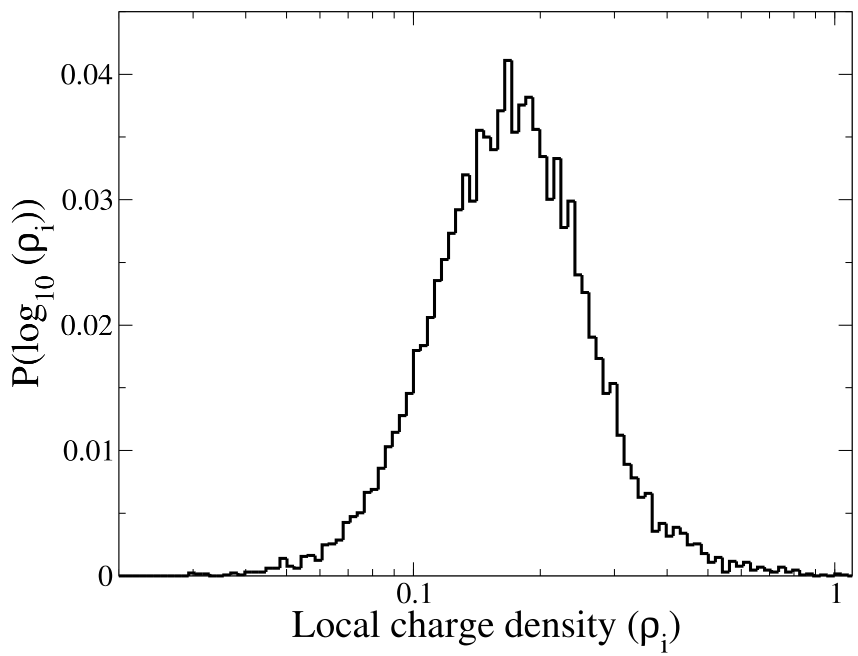
Local Charge density

Charge density: $\rho_i = \sum_{j,\sigma} |\phi_{ij}|^2 \left\langle c^\dagger_{j\sigma} c_{j\sigma} \right\rangle$

$x = 0.01$, $p = 0.1$ (Insulating)



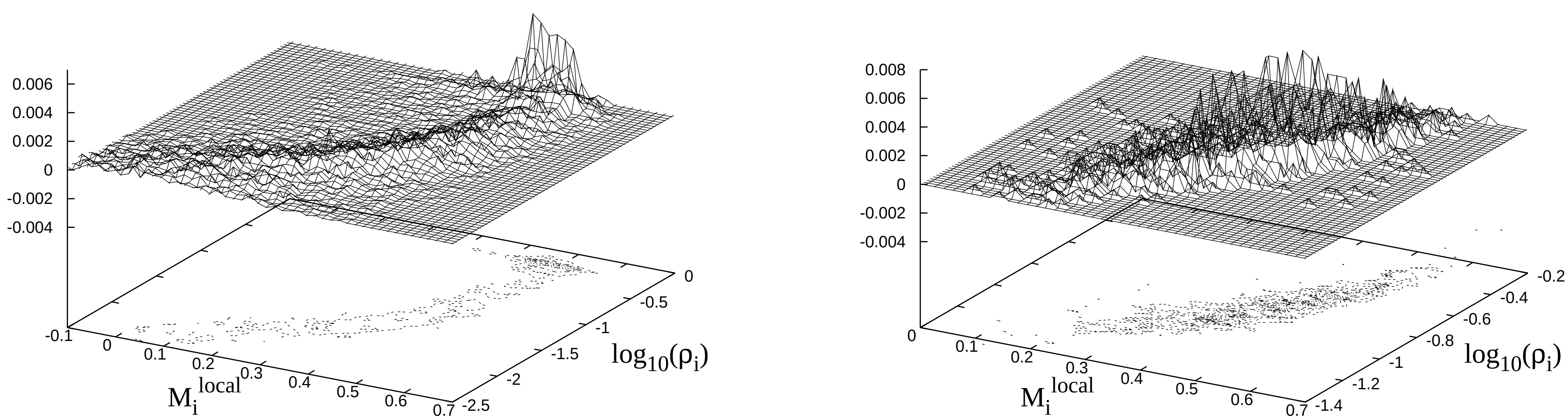
$x = 0.03$, $p = 0.1$ (near MIT)



Inhomogeneous Ferromagnetic state

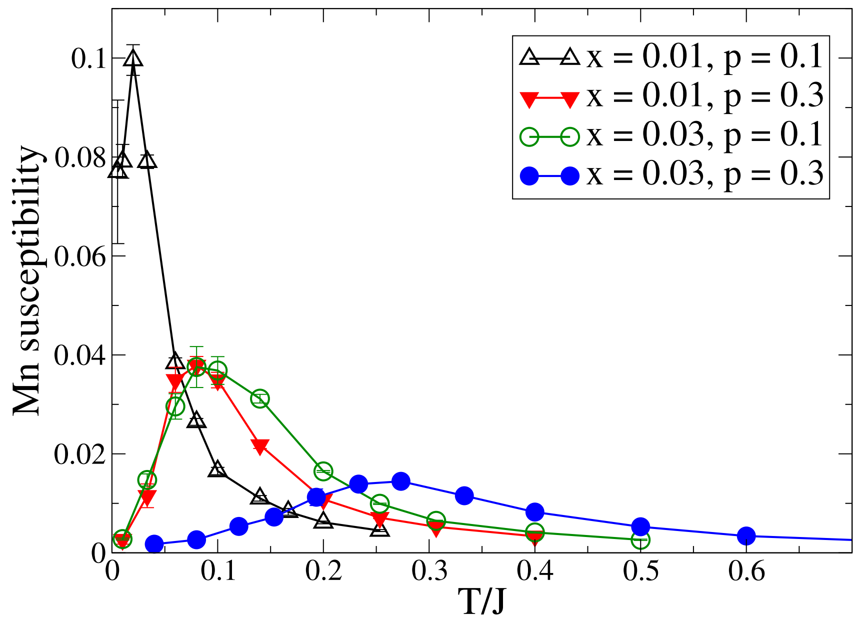
Strong correlation between local charge density and local magnetization:

$x = 0.01$, $p = 0.1$ (Insulating) $x = 0.03$, $p = 0.1$ (near MIT)

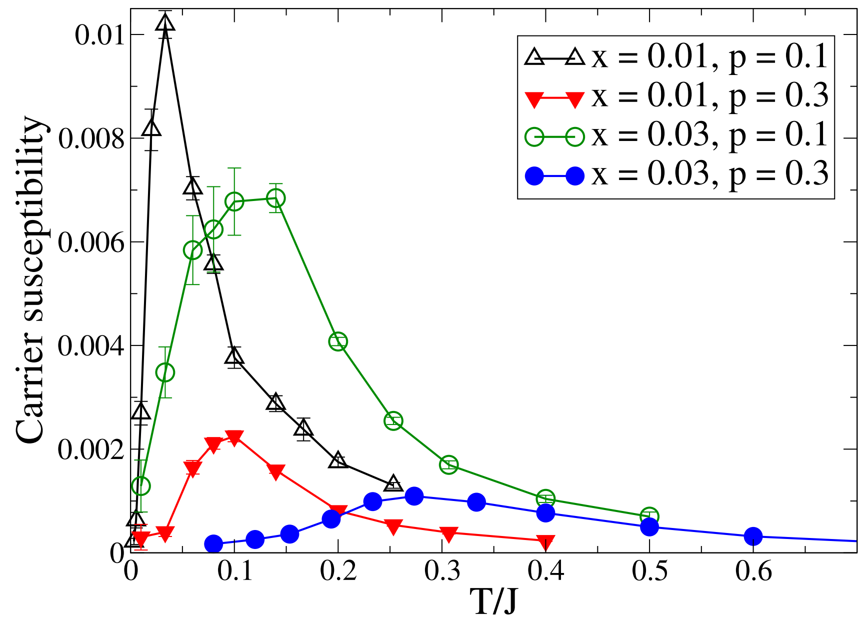


Susceptibility

Mn Susceptibility



Carrier Susceptibility



- Peak below T_c also observed in Monte Carlo studies of II-VI DMS (Wan and Bhatt) due to nearly free spins at low temperatures

High temperature susceptibilty

For $T \gg T_c$ and classical spins: $\mathbf{Curie\ Law}$

$$\frac{1}{\chi} \simeq \frac{3k_B T}{n(g\mu_B)^2 S^2}\left(1 - \frac{\theta}{T}\right),$$

(the factor of S^2 is replaced by $S(S+1)$ for quantum spins)

For 2 $T_c < T < 6T_c$:

$$S_{\text{eff}} \simeq 1.6 - 1.7S$$

For $T_c < T < 2T_c$:

$$S_{\text{eff}} \simeq 2S$$

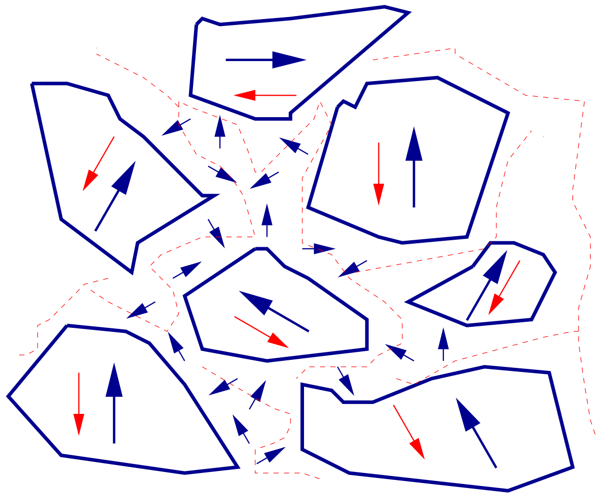
Enhancement may be evidence of magnetic polaron formation ?

- (Ga,Mn)P: experimental observation of $S_{\text{eff}} \simeq 4S$

Inhomogeneous Ferromagnetism

Large local carrier density $\Longleftrightarrow$ large local magnetization

- Magnetic polaron type picture



What has been left out ?

- Hole-hole coulomb interactions – at mean field level, on-site repulsion leads to slightly higher T_c
- On-site disorder – tends to counteract local clustering, at mean field level, T_c is slightly reduced

Qualitative features are likely to be robust

Phenomenological two component model

Can one parametrize experimental data using a simple mean-field model?

- Spin-only model: Mn spin $\mathbf{S}_i$, Carrier spin $\mathbf{s}_\alpha$

$$\begin{aligned} \mathcal{H} &= \sum_i \sum_\alpha J_{i\alpha}(\mathbf{R}_i)\mathbf{s}_\alpha \cdot \mathbf{S}_i, \\ J_{i\alpha}(\mathbf{R}_i) &= J_0 |\phi_\alpha(R_i)|^2 \end{aligned}$$

- Each Mn feels an effective field due to interactions with holes

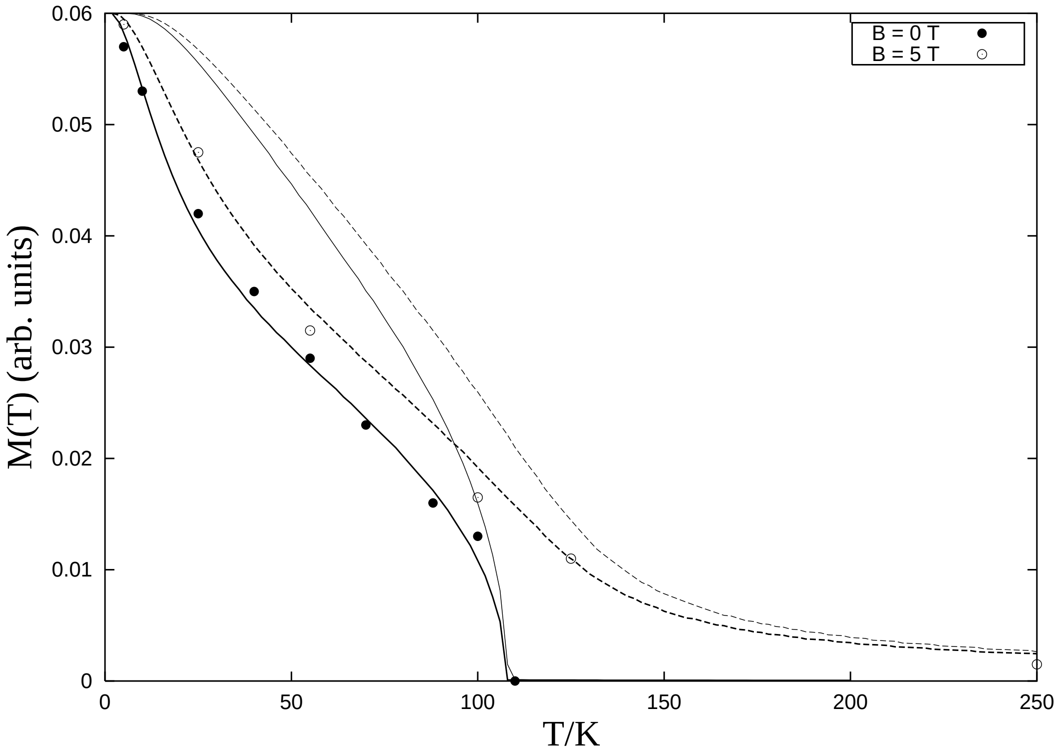
$$\mathbf{h}_i(\mathbf{R}_i) = \sum_\alpha J_{i\alpha}(\mathbf{R}_i)\mathbf{s}_\alpha$$

- Many more Mn than holes – large fluctuations in the local environments of different Mn spins.

- A single coupling $J_{ij} = J$ insufficient to capture behaviour from impurity band model, at least two different Js required J_1 and J_2 with concentrations n_1 and n_2

- Two populations of Mn spins, strongly and weakly coupled.

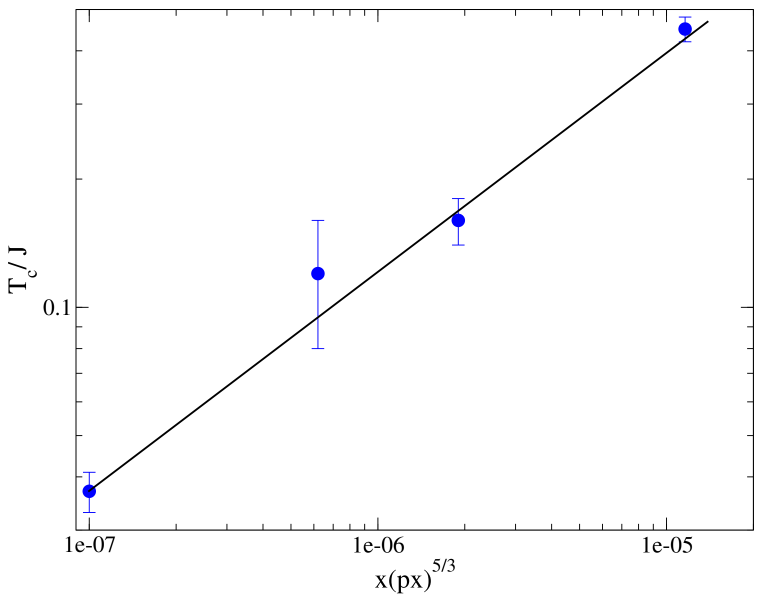
$$\mathcal{H} \simeq \sum_{i_1} \mathbf{h}_1 \cdot \mathbf{S}_{i_1} + \sum_{i_2} \mathbf{h}_2 \cdot \mathbf{S}_{i_2}$$

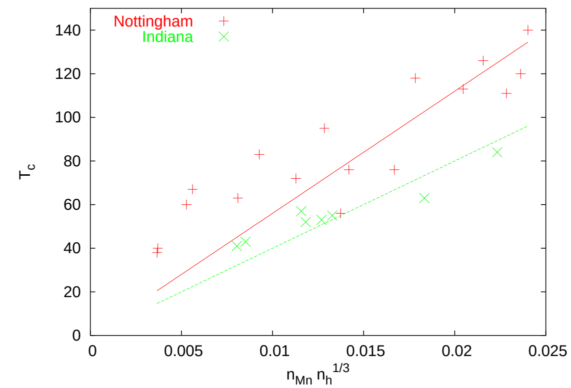


- Fit to data from Ohno ($T_c = 110$ K, $x = 0.053$)with $J_1 = 47.5$ K , $J_2 = 7.5$ K and $n_1 = 0.41\, n_{Mn}$ compared to $J = 31$ K

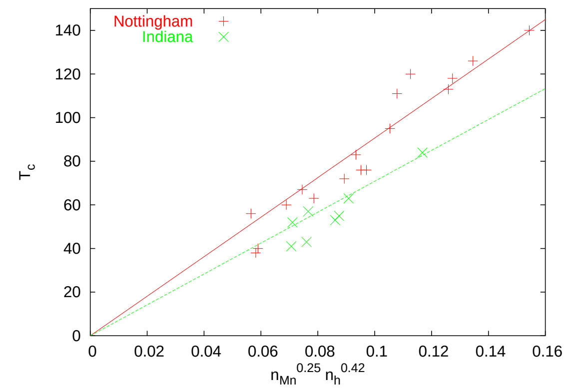
Critical Temperature

T_c as a function of x and p





Fit to expts. with Mean Field model



Find:

$$\alpha \simeq 0.5 \pm 0.15, \quad \beta \simeq 0.85 \pm 0.15$$

Fit to expts. with $\alpha = 0.25$, $\beta = 0.42$

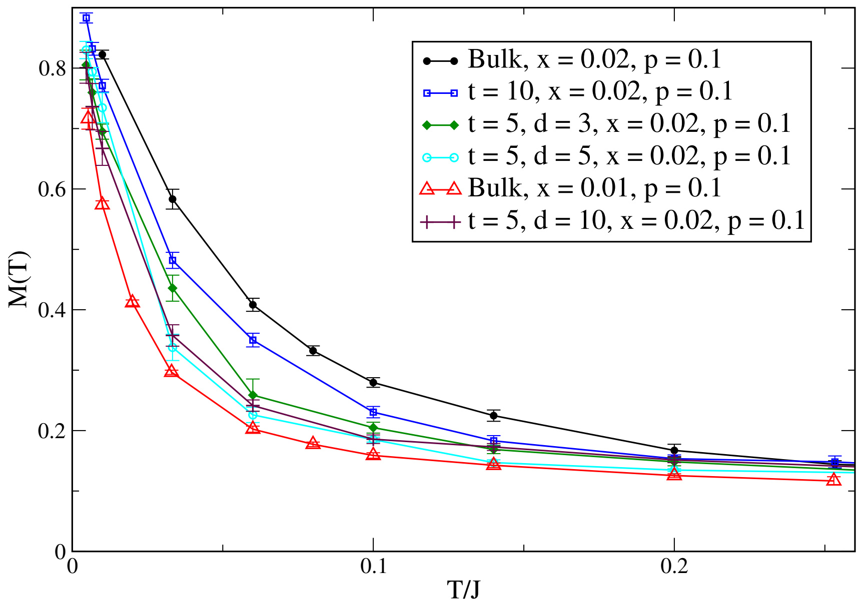
- Many theories give power law dependence of T_c on Mn, carrier concentration:

$$T_c \sim x^{\alpha}(px)^{\beta}$$

Layered DMS

- Many samples of $Ga_{1-x}Mn_xAs$ grown as digitally doped sructures – either as 1/2, 1/4, or 1/8 monolayers of MnAs in GaAs, or as layers of $Ga_{1-x}Mn_xAs$ – in practice Mn spins found to reside over 3-5 monolayers

- Performed calculations on layered DMS structures, investigating effects of layer thickness (t), layer spacing (d) and Mn, carrier concentrations.



For same overall concentration of Mn, magnetization is enhanced in layered structure

Anisotropy

Theory: Spin-orbit coupling + different masses for light and heavy hole bands leads to anisotropic exchange interactions between Mn spins (Zarand and Janko, 2002).

- Zarand and Janko found that for RKKY interactions, the saturation magnetization at low temperatures is reduced by 50%.

- Effects should be strongest at smaller values of Mn, carrier concentration

Hamiltonian

$$\mathcal{H} = -\sum_{ij} \sum_{\alpha\beta} J_{\alpha\beta}(\mathbf{R}_i - \mathbf{R}_j) S_i^\alpha S_j^\beta$$

- Classical spins (Mn: spin-5/2 is large) $\mathbf{S}_i$
- Spins are randomly placed on a fcc lattice

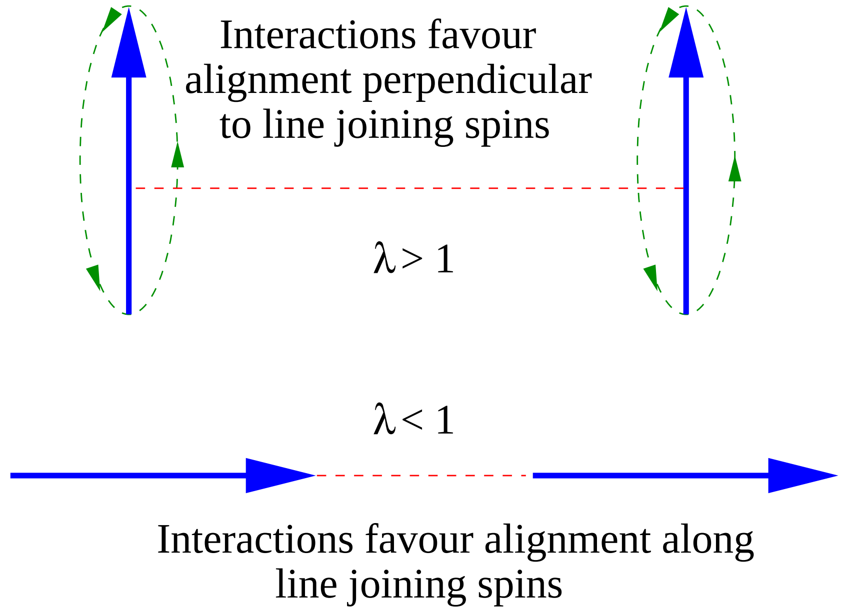
Anisotropic Exchange Interactions:

$$J_{\alpha\beta}(\mathbf{R}_i - \mathbf{R}_j) = J_{ij} \left(\lambda \delta_{\alpha\beta} + (1-\lambda) \hat{e}_{ij}^\alpha \hat{e}_{ij}^\beta \right) f(|\mathbf{R}_i - \mathbf{R}_j|)$$

Isotropic term and anisotropic term.

Anisotropic Exchange

Effects of Anisotropic spin interactions :



Types of model:

1) All exchanges are ferromagnetic (long and short range)

2) Some exchanges are anti-ferromagnetic (RKKY)

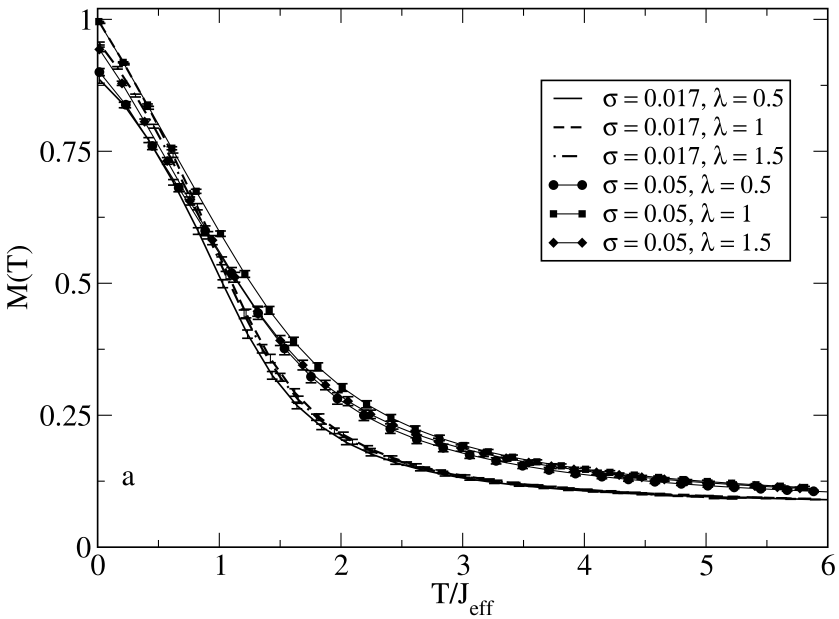
- Models also include some disorder in the couplings (J_{ij})
- Temperature rescaled to compare different anisotropy values
- Studied with Monte Carlo simulations.

Ferromagnetic Couplings: Magnetization

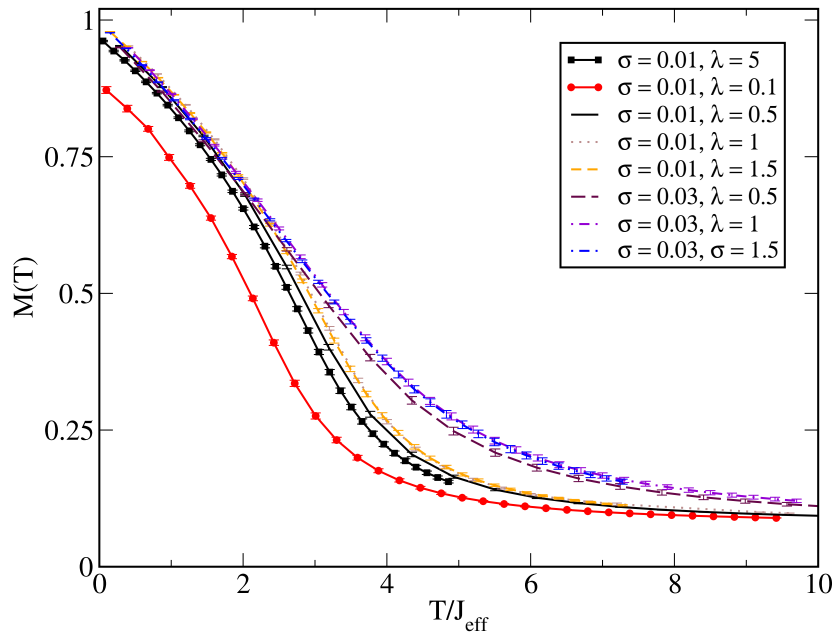
Relatively small reductions in saturation magnetization at low temperature even for large anisotropy

Short range interactions :

6 neighbours

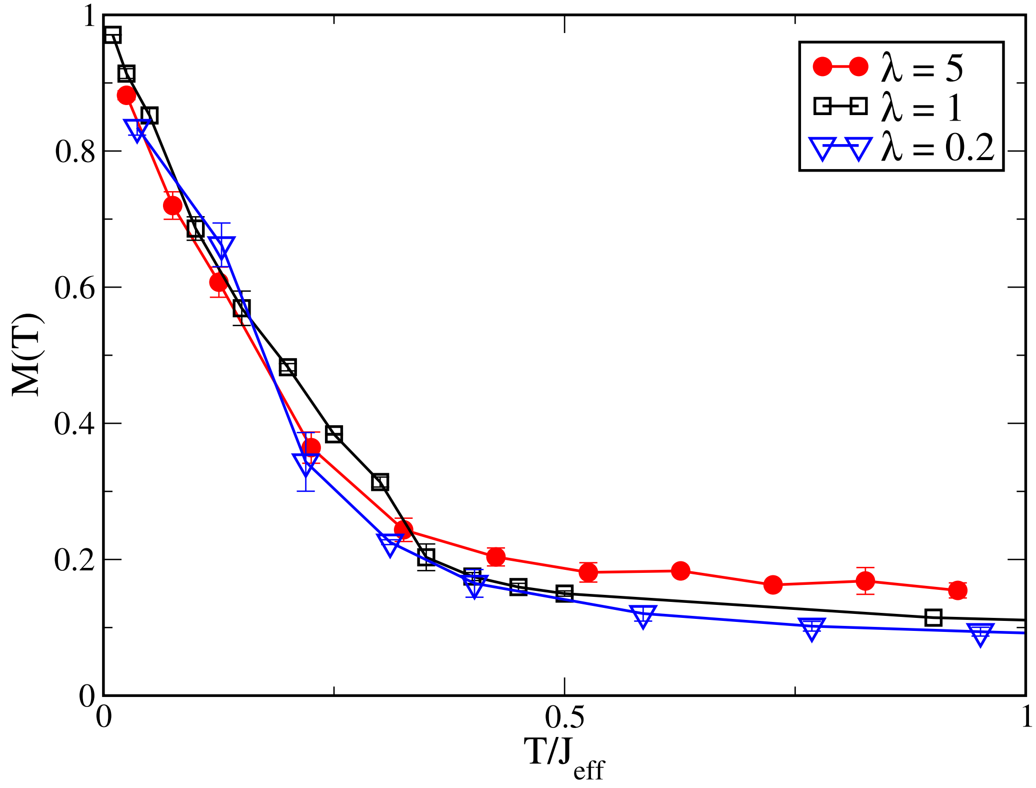


12 neighbours



Ferromagnetic Couplings: Magnetization

Long range interactions :



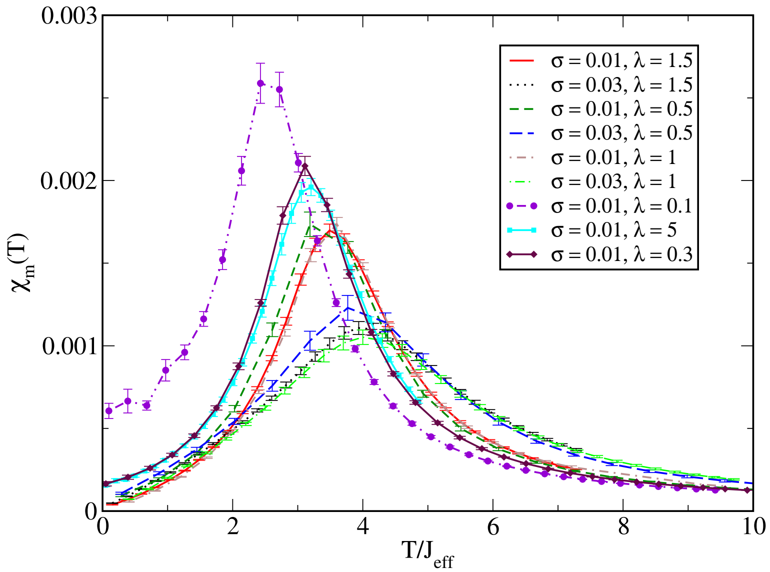
Results are apparently less sensitive to anisotropy for higher connectivity

Susceptibility

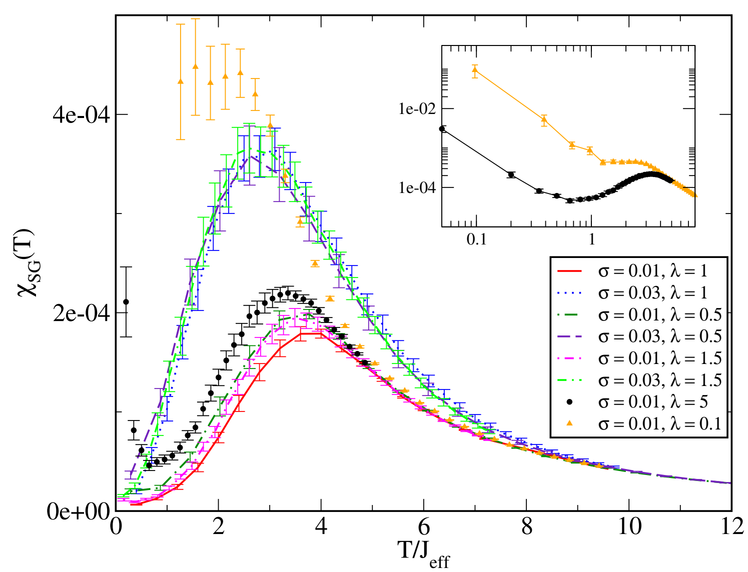
The spin glass and linear susceptibilty show deviations from isotropic behaviour only at large anisotropy

Linear Susceptibility:

12 neighbours model



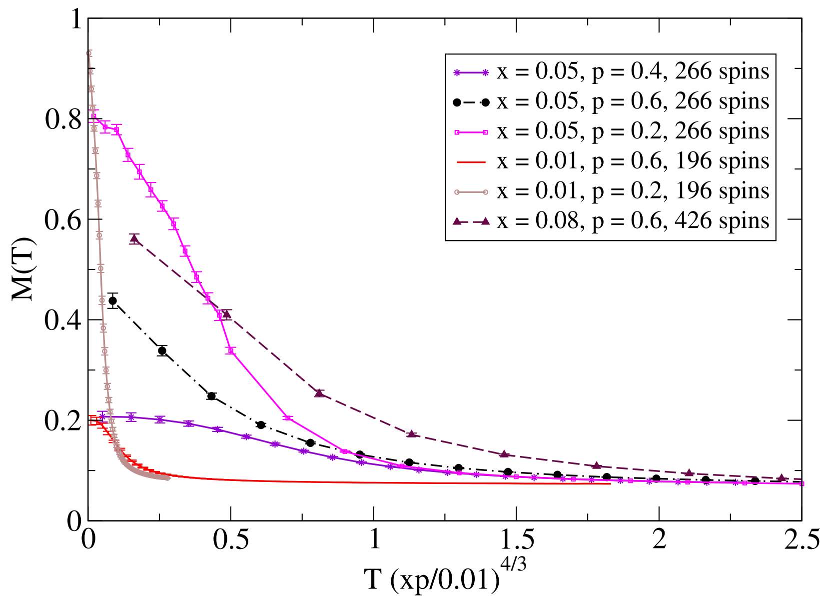
Spin Glass (non-linear) Susceptibility:

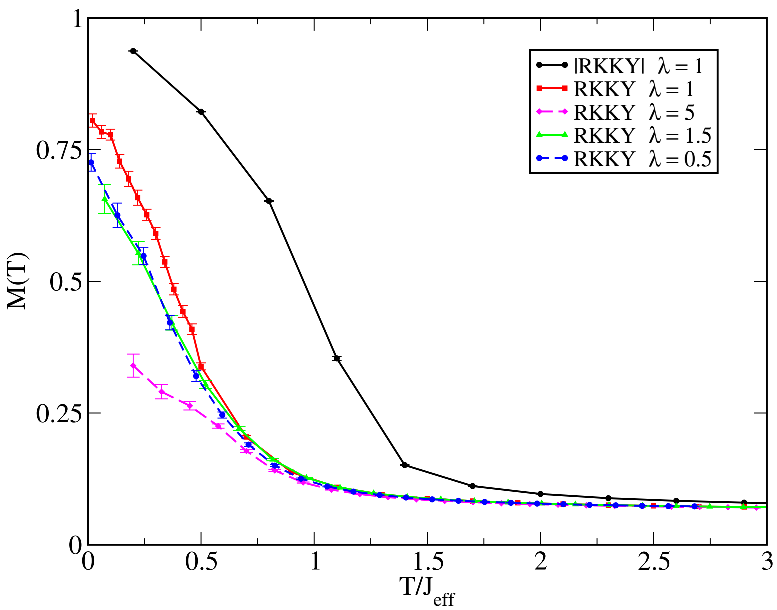


Spin freezing?

RKKY couplings + Anisotropy

Without anisotropy: With anisotropy:





Magnetization for RKKY model, at various x and p (corresponding to different choices of k_F).

Conclusions

- Have performed Monte Carlo simulations of an impurity band model for DMS and pure spin models of DMS

- Disorder leads to a large population of weakly coupled Mn spins and an inhomogenous ferromagnetic state, but does not induce frustration in the models studied

- Have investigated dependence of T_c on width and spacing of layers in layered structures

- Anisotropy only affects magnetic properties when it is large (the ratio of the parallel and perpendicular parts of the exchange is greater than about 5), and can lead to some decrease in the magnetization at low temperatures. Apparent divergence in non-linear susceptibility at low temperatures – Spin glass behaviour?

- Anti-ferromagnetic interactions can change the low temperature magnetization by large amounts. Significant effect of antiferromagnetic interactions needed for oscillatory exchange (e.g. RKKY) to lead to low saturation magnetization. Magnetically inactive Mn, such as Mn interstitials are also important.

Future Work

- Including role of As antisites, Mn interstitials
- Strengthening understanding of connection between magnetic and transport properties
- Studying other DMS, for which other types of models may be more appropriate
- Further study of hopping in DMS