Mechanisms of exchange interactions in dilute $Ga_{1-x}Mn_xN$ experiment and modelling

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magic spin
magnetic spin materials group

Sept. 10, 2012

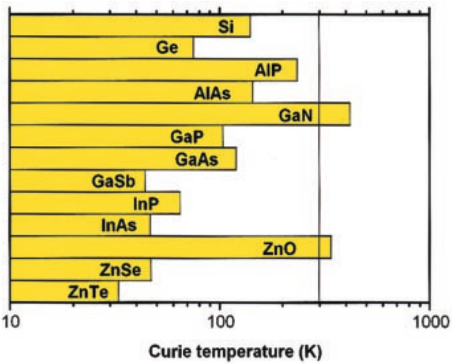
HFP INSTITUTE OF SEMICONDUCTOR AND SOLID STATE PHYSICS

SemiSpinNet erc MARIE CURIE ACTIONS IDEAS LIDER FWF

Outline

1. Introduction
2. Interaction of Mn 3*d*-states with GaN bands, $x_{\mathrm{Mn}} < 1\%$
3. Interaction between Mn ions, $x_{\mathrm{Mn}} > 1\%$
   - (Ga,Mn)N
   - (Ga,Mn)N:Si
4. Outlook

Motivation



[Dietl *et al.*, Science 2000]

(Ga,Mn)N

- Free holes: RT-FM?
  - Refined theory
  - Bound hole state?
  - *p*-*d* interaction strength?
- No holes:
  - Mn-Mn interaction?

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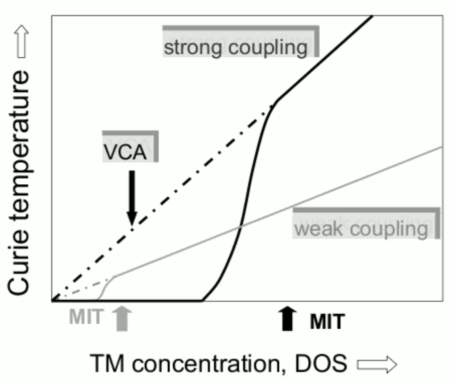

[Dietl, PRB 2008]

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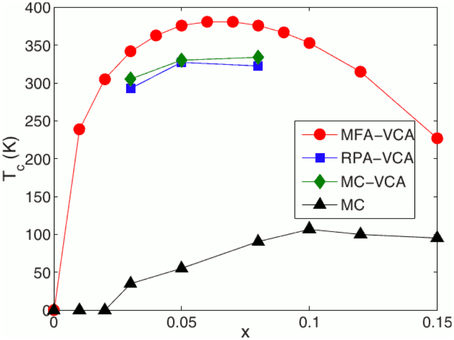

[Sato *et al.*, Rev. Mod. Phys. 2010]

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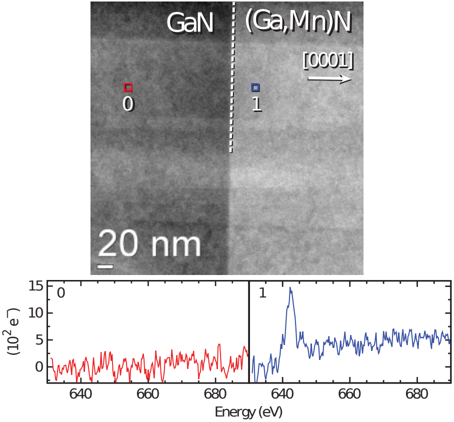
(Ga,Mn)N growth

- MOVPE
- $\mathrm{Al_2O_3}$ substrate
- 1 $\mu\mathrm{m}$ GaN buffer
- 0.5 $\mu\mathrm{m}$ (Ga,Mn)N
- Dopants available:
  - Silicon
  - Magnesium
  - Manganese
  - Iron
  - Indium
  - Aluminum

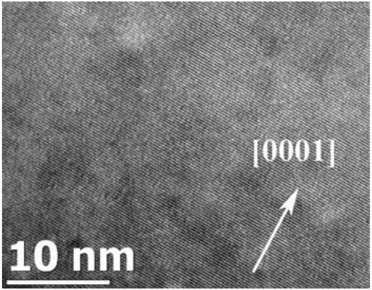
Details:
[Stefanowicz,..., AG,..., PRB 2010]

Search for secondary phases
Microscopy



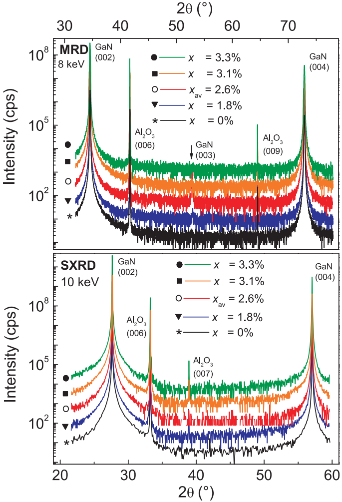
[Bonanni,..., AG,..., PRB 2011,
Stefanowicz,..., AG,..., PRB 2010]

- EELS
- HRTEM



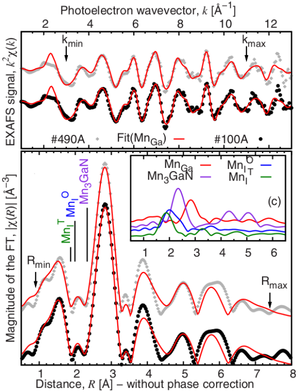
Search for secondary phases

Lab- and Synchrotron X-rays

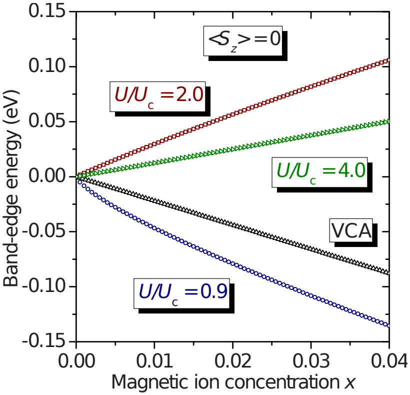


[Bonanni,..., AG,..., PRB 2011, Stefanowicz,..., AG,..., PRB 2010]

- High resolution XRD
- Synchrotron XRD
- Synchrotron XAS



Strong or weak coupling, effect on the band gap

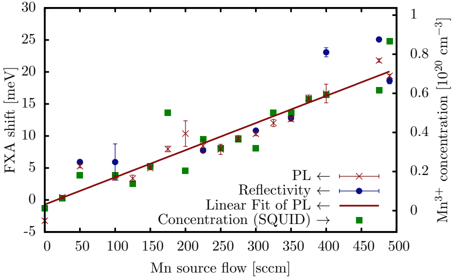


[Dietl, PRB 2008]

- Theory for $U > U_C$ (strong coupling):
  - Bound hole state
  - Doping: increase of E_{gap}
- PL as function of x_{Mn}
  - (Ga,Mn)N exciton energy
  - $E_{gap} \propto x_{Mn}$

strong coupling

Strong or weak coupling, effect on the band gap



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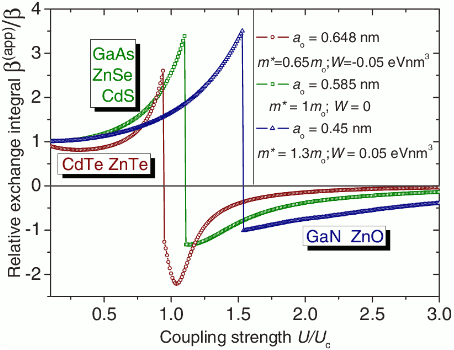
strong coupling

[Suffczyński, AG,..., PRB 2011]

Giant Zeeman Splitting



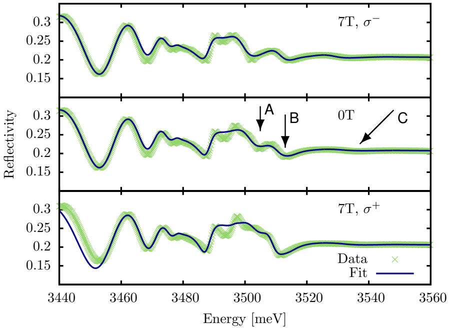
[Dietl, PRB 2008]

- [Dietl, PRB 2008]: Exchange: apparent constant β^{app}
- Reflectivity: $E_{\mathrm{Exciton}}(\vec{B})$
- Full Hamiltonian:
  - VB & CB exchange
  - VB CF splitting
  - Zeeman & Diamagn.
  - e-h interaction

Fit yields

- $N_0\beta^{\mathrm{app}} = 0.8 \pm 0.2$ eV
- $N_0\alpha^{\mathrm{app}} = 0.0 \pm 0.1$ eV

Giant Zeeman Splitting



[Suffczyński, AG,..., PRB 2011]

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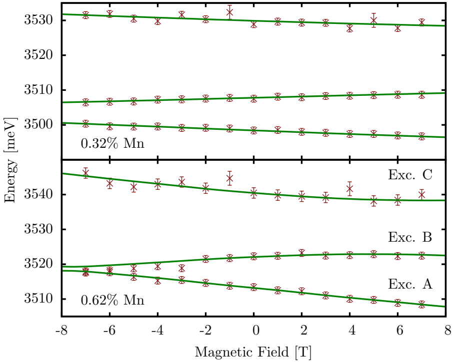

[Suffczyński, AG,..., PRB 2011]

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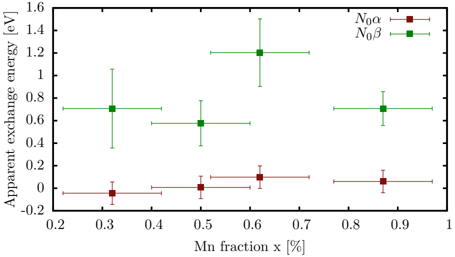

[Suffczyński, AG,..., PRB 2011]

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Conclusion

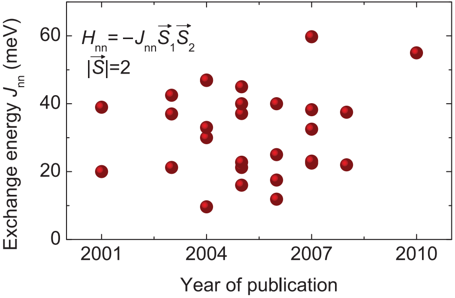
As predicted by theory in [Dietl, PRB 2008]:

- Band gap of (Ga,Mn)N increases with Mn concentration
- Apparent $p - d$ exchange integral β^{app} has nonstandard
  - sign
  - value

Indicates:

- (Ga,Mn)N: Strong coupling
- Bound hole state at Mn ions

Simulated exchange energies

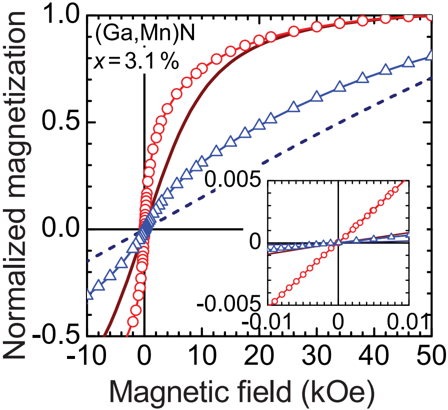


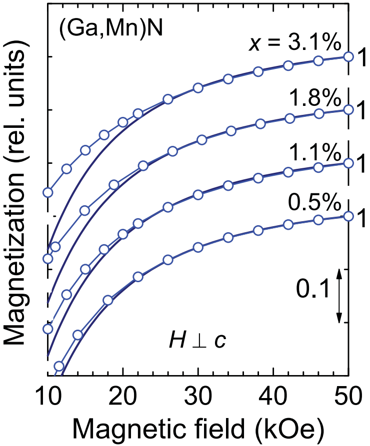
[Bonanni,..., AG,..., PRB 2011]

- Magn. Mn-Mn interaction without free carriers
- Numerous predictions:
  - short ranged
  - ferromagnetic

Lack of experimental values

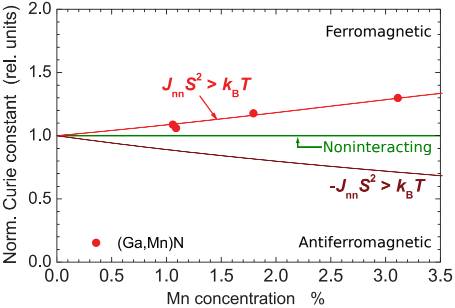
Magnetisation curves modified by Mn-Mn interaction





Circles: $\vec{H} \perp \vec{c}$
Triangles: $\vec{H} \parallel \vec{c}$
Lines: Noninteracting Model
[Bonanni,..., AG,..., PRB 2011]

Curie Constant

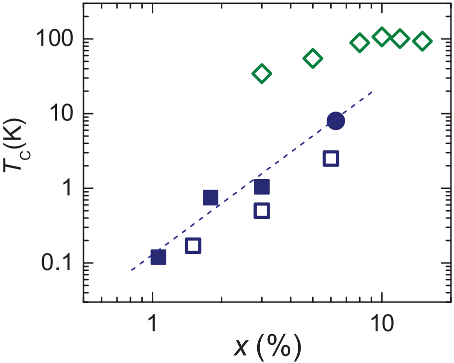


[Bonanni,..., AG,..., PRB 2011]

- $C_{\text{norm}} = \frac{C_{\text{measured}}}{C_{\text{noninteracting}}} > 1$
- $E_{\text{NN}} = -J_{\text{NN}} \vec{S}_1 \cdot \vec{S}_2$
- Comparison with theory: $J_{\text{NN}} > 10$ meV

Mn^{3+}: ferromagnetic interaction

Curie Temperature



[Sawicki,..., AG,..., PRB 2012]

Custom SQUID setup in Warsaw:

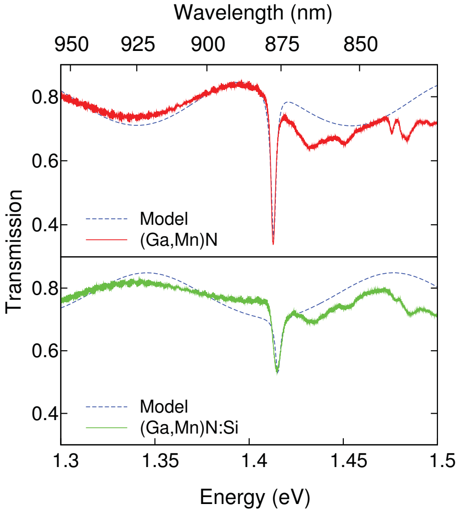
- ^{3}He-^{4}He dilution fridge
- Millikelvin range

Details

Sylwia Dobkowska
This room 12:00

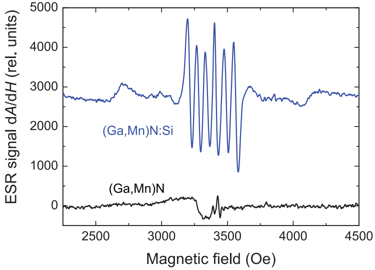
Full Squares: Our experimental values
Open Squres: Our Monte Carlo simulations
Circle: [Sarigiannidou *et al.*, PRB 2006], experimental
Diamonds: [Sato *et al.*, Rev. Mod. Phys. 2010], simulation

Codoping of (Ga,Mn)N with Si

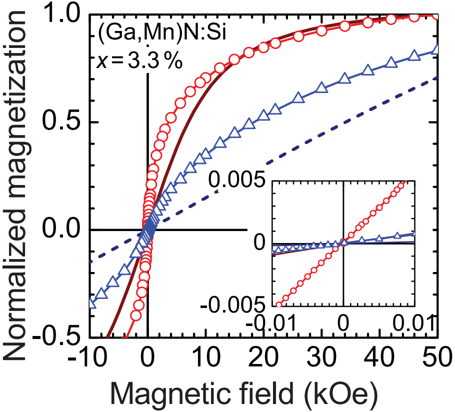


[Bonanni,..., AG,..., PRB 2011]

- Codoping modifies Mn valence
- Si codoping: $Mn^{3+} \rightarrow Mn^{2+}$
  - XANES
  - EPR
  - Intra-ion absorption



Magnetisation curves of (Ga,Mn)N:Si



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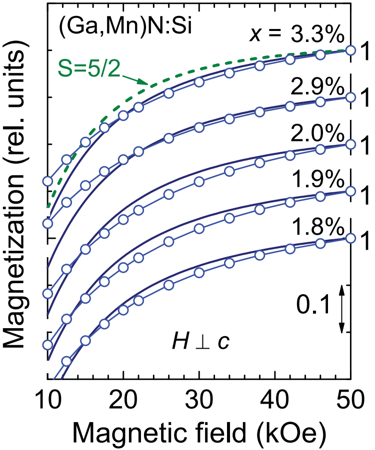

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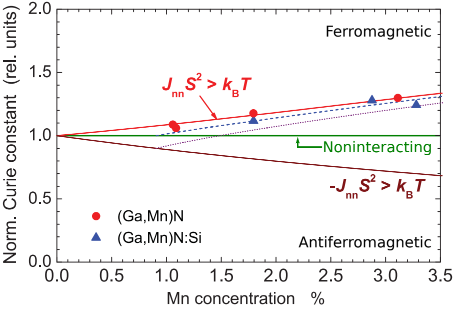


[Bonanni,..., AG,..., PRB 2011]

- Compared to Mn^{3+}:
  - Lower Magnetic anisotropy
  - Magnetisation curve differs
  - Lower Curie constant

Mn^{2+}: Antiferromagnetic coupling

Conclusion

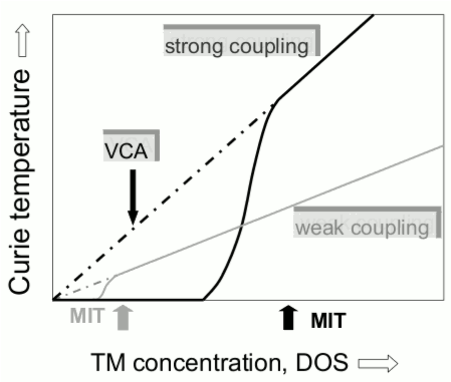
Mn^{3+}: Ferromagnetic superexchange

- Curie Constant higher than for noninteracting Mn
- Millikelvin SQUID: Hysteresis opens, T_{C} can be measured

Mn^{2+}: Antiferromagnetic superexchange

- indicated by saturation behaviour

Outlook



[Dietl, PRB 2008]

Strong Coupling

- Increase carrier concentration
- Modulation doping
- 2D hole gas

Interaction without carriers

- Knowledge of interactions
- Control FM/AF by codoping
- Detailed study of phase transitions

Thank you for your attention!

Bibliography

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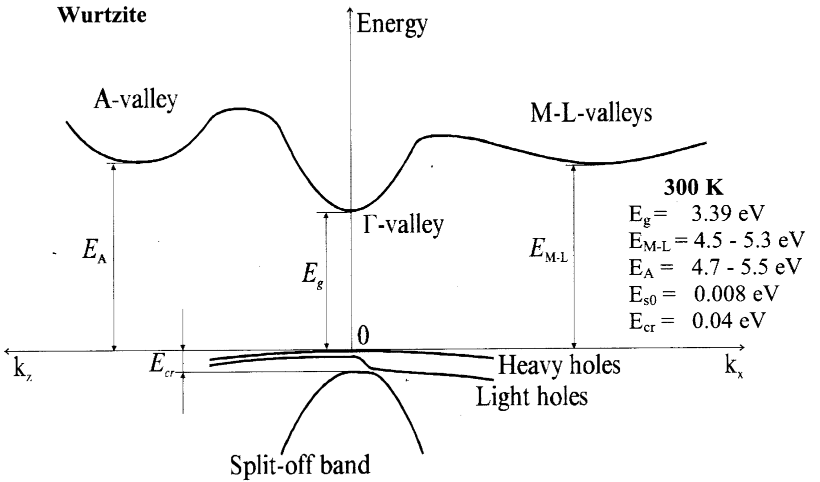
Effects of *s,p-d* and *s-p* exchange interactions probed by exciton magnetospectroscopy in (Ga,Mn)N.
Phys. Rev. B, 83(9):094421.

Giant Zeeman Splitting

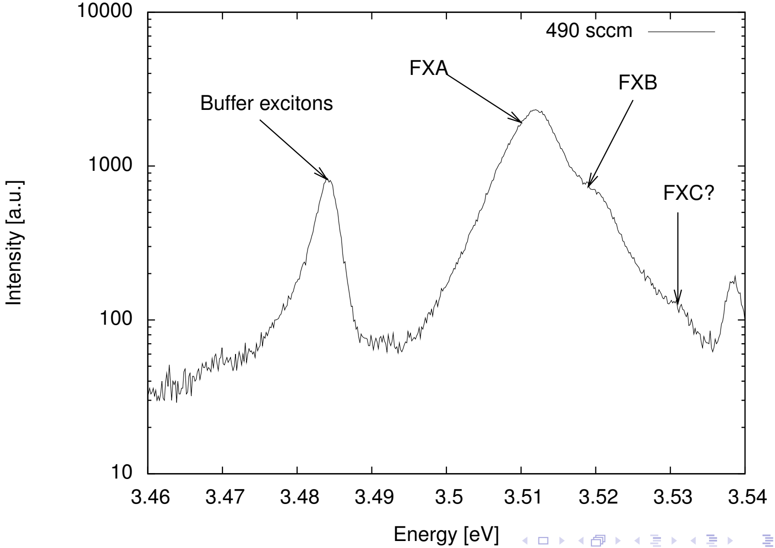
$$
\begin{aligned}
H_{exc} &= E_0 + H_{\mathrm{vb}} + H_{\mathrm{e-h}} + H_{\mathrm{Z,dia}} + H_{s,p-d} && (1)\\
H_{\mathrm{e-h}} &= -R^* + 2\gamma \vec{s}_e \cdot \vec{s}_h && (2)\\
H_{\mathrm{Z,dia}} &= \mu_B \vec{B} \left(g_e \vec{s}_e + 2\vec{s}_h - (3\tilde{\kappa} + 1)\, \vec{l}_h \right) + dB^2 && (3)\\
H_{s,p-d} &= -N_0 x_{eff} \left\langle \vec{S}\,(B) \right\rangle \cdot \left(\vec{s}_e \alpha_{app} + \vec{s}_h \beta_{app} \right) && (4)\\
H_{\mathrm{vb}} &= -\left(\tilde{\Delta}_1 \right) \left(l_{h_z}^2 - 1 \right) - 2\Delta_2 l_{h_z} s_{h_z} - 2\Delta_3 \left(l_{h_x} s_{h_x} + l_{h_y} s_{h_y} \right) && (5)
\end{aligned}
$$

Band Structure

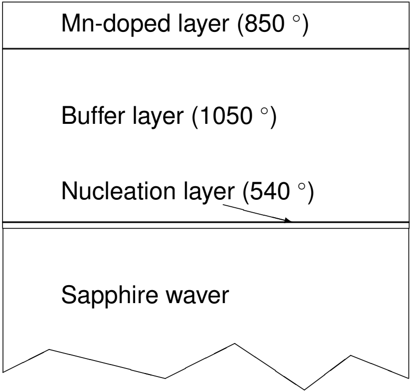


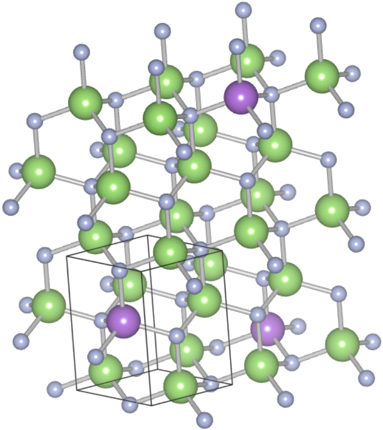
[http://www.matprop.ru/GaN_bandstr#Basic%20wurtzite]

Photoluminescence Spectra

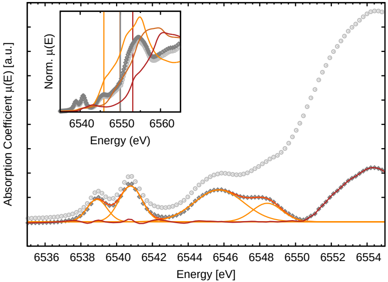


Sample Structure

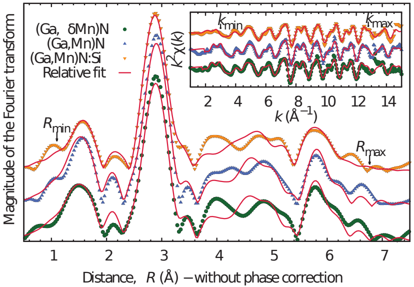




EXAFS/XANES data



Near-edge peak fit for x≈1%. First two are spin up and down t_2 states of a d^4 shell, second two are critical points in conduction bands. Inset: Comparison to MnO, Mn_2O_3, MnO_2 (d^5, d^4, d^3).



Mn k-edge EXAFS for x≈3%
Homogeneous sample has 0.2±0.4 Mn neighbours, no interstitial Mn is visible