Optical investigation of excitons in GaN:Mn

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Aim of the research

- Determination of GaN:Mn energy gap variation with Mn concentration
- Quantitative observation of giant Zeeman splitting of A, B **and C** excitons in GaN:Mn

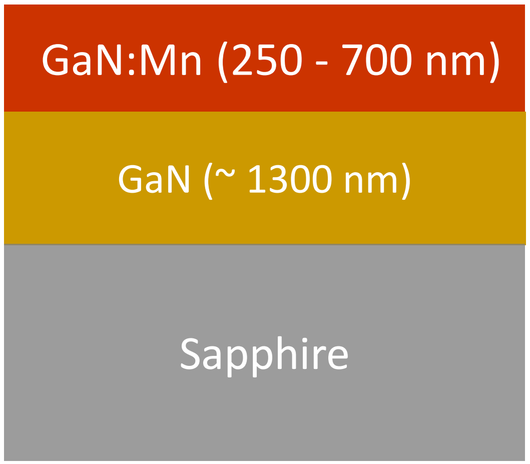
Compare to : Pacuski,

PRB'07

- Determination of both exchange constants $N_0\alpha$ and $N_0\beta$ of Mn^{3+} ion interaction with band carriers in GaN:Mn

Compare to GaAs: Szczytko, PRB'99; Krebs, PRB'09;

Samples



Grown by MOVPE on GaN buffer

x_{Mn} = 0.14 % to 0.8 % determined by SQUID and SIMS

Samples from the same series

High res. transmission electron microscopy
& Synchrotron x-ray diffraction
& Extended x-ray absorption fine structure

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No crystallographic phase separation

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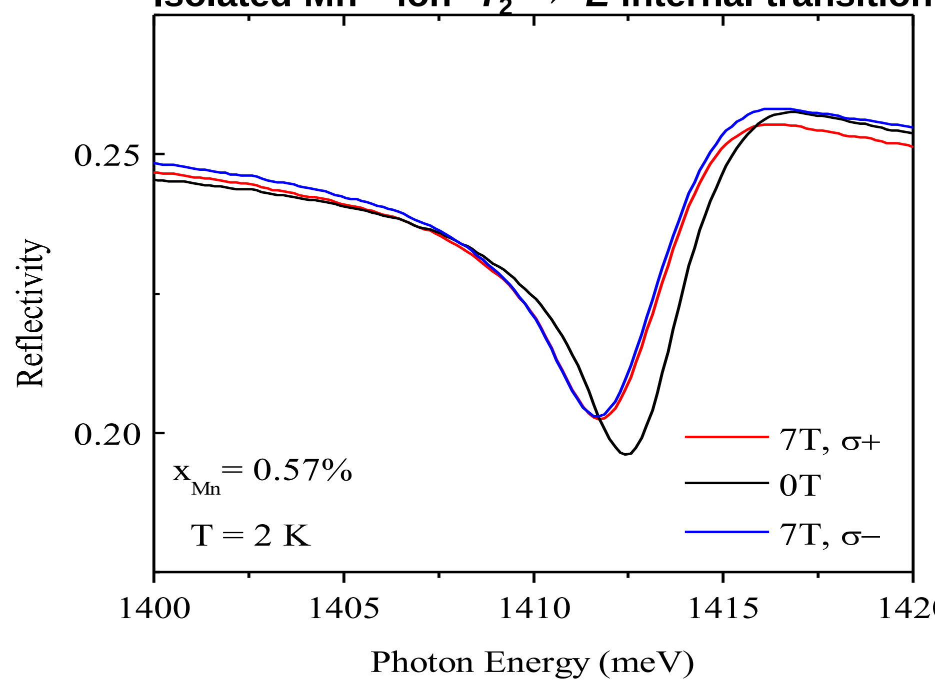
Mn ions occupy Ga-substitutional sites

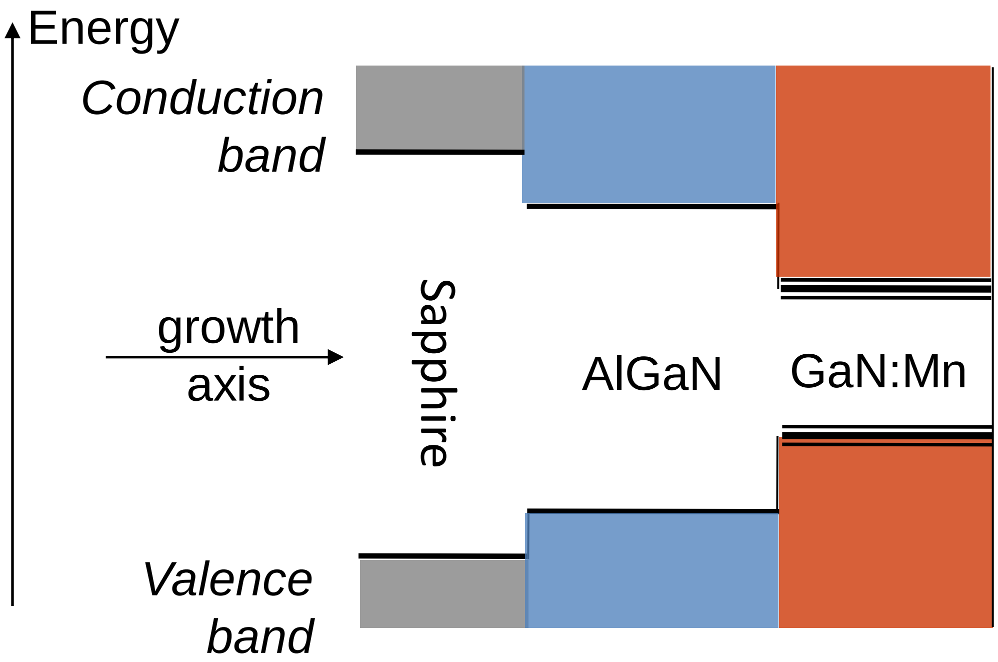
Isolated Mn^{3+} ion $^5T_2 \rightarrow {}^5E$ internal transition

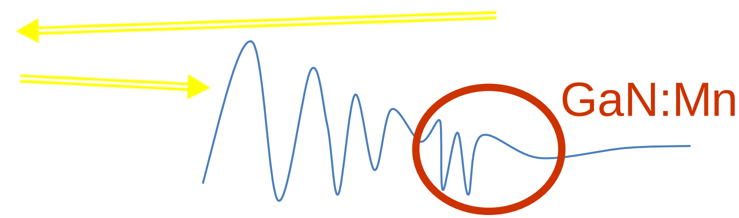


→ Presence of isolated Mn^{3+} ions confirmed

Compare to : Wołoś, PRB’04, Marcet, PRB’06

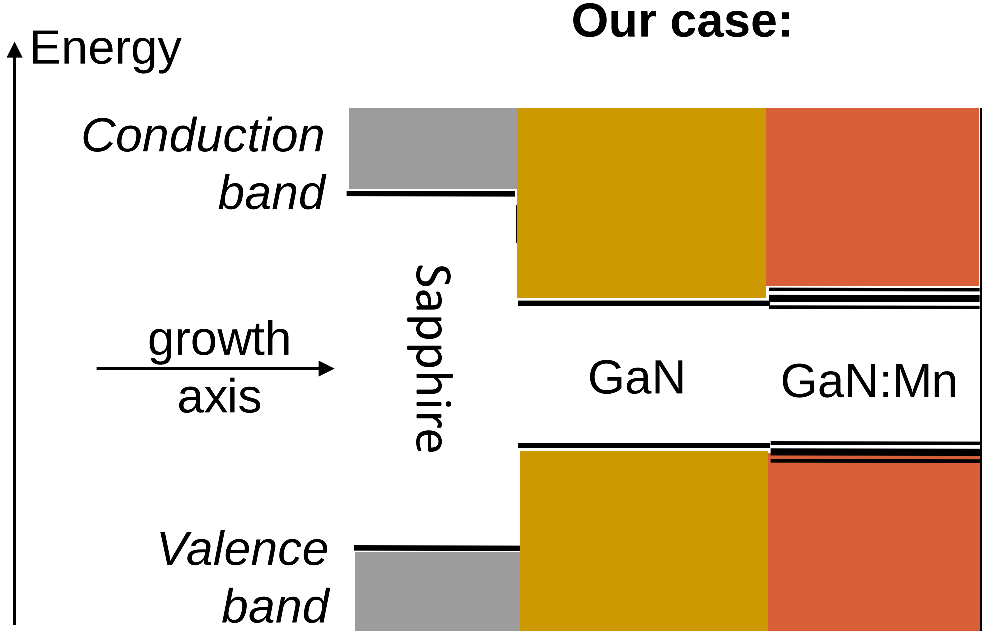
Samples

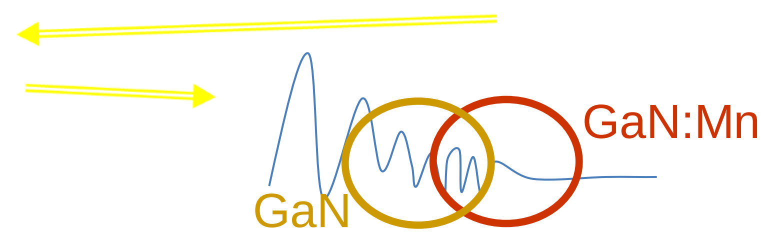




Excitons in GaN:Mn and AlGaN separated spectrally but structure strained

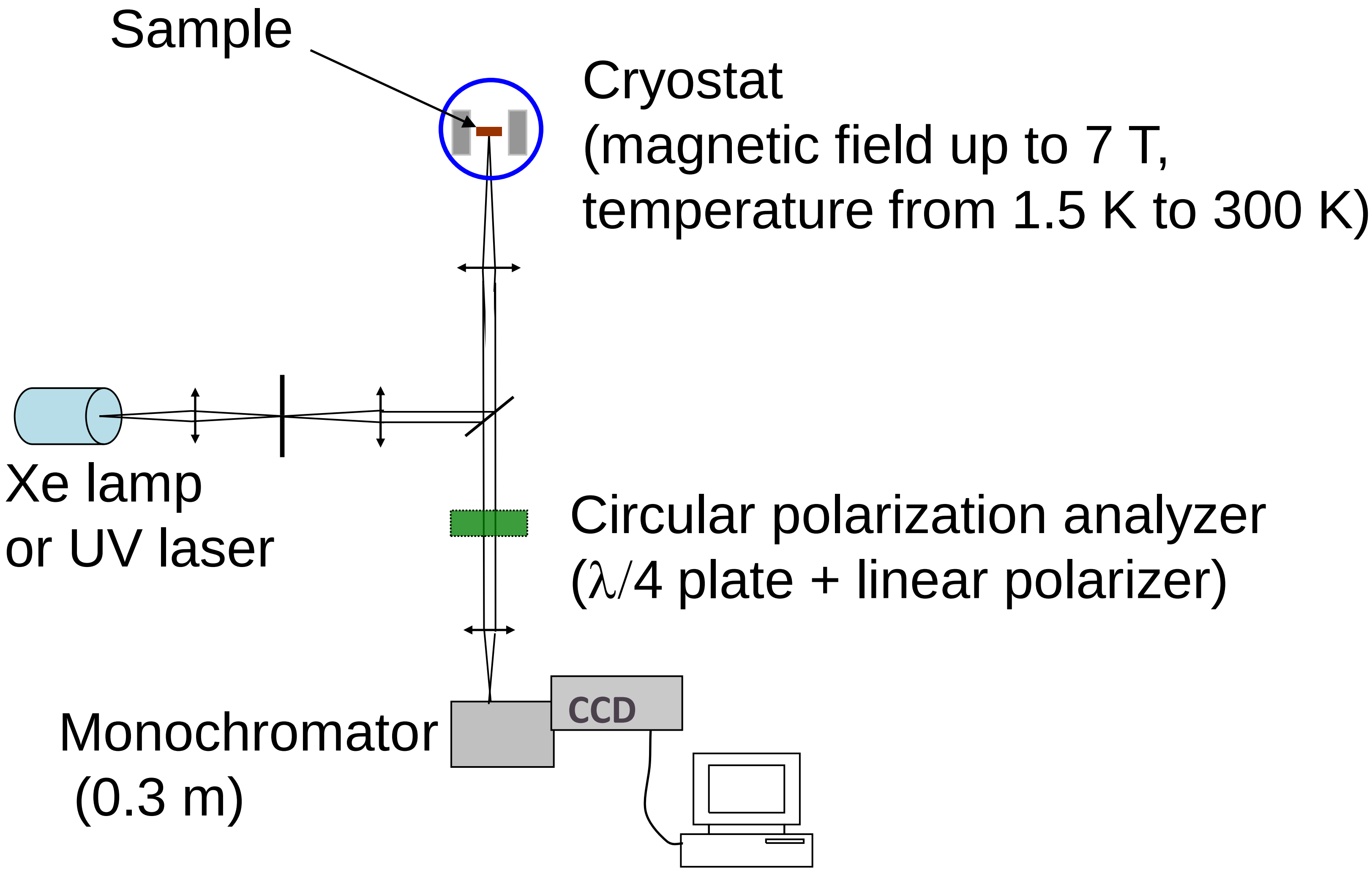
Our case:



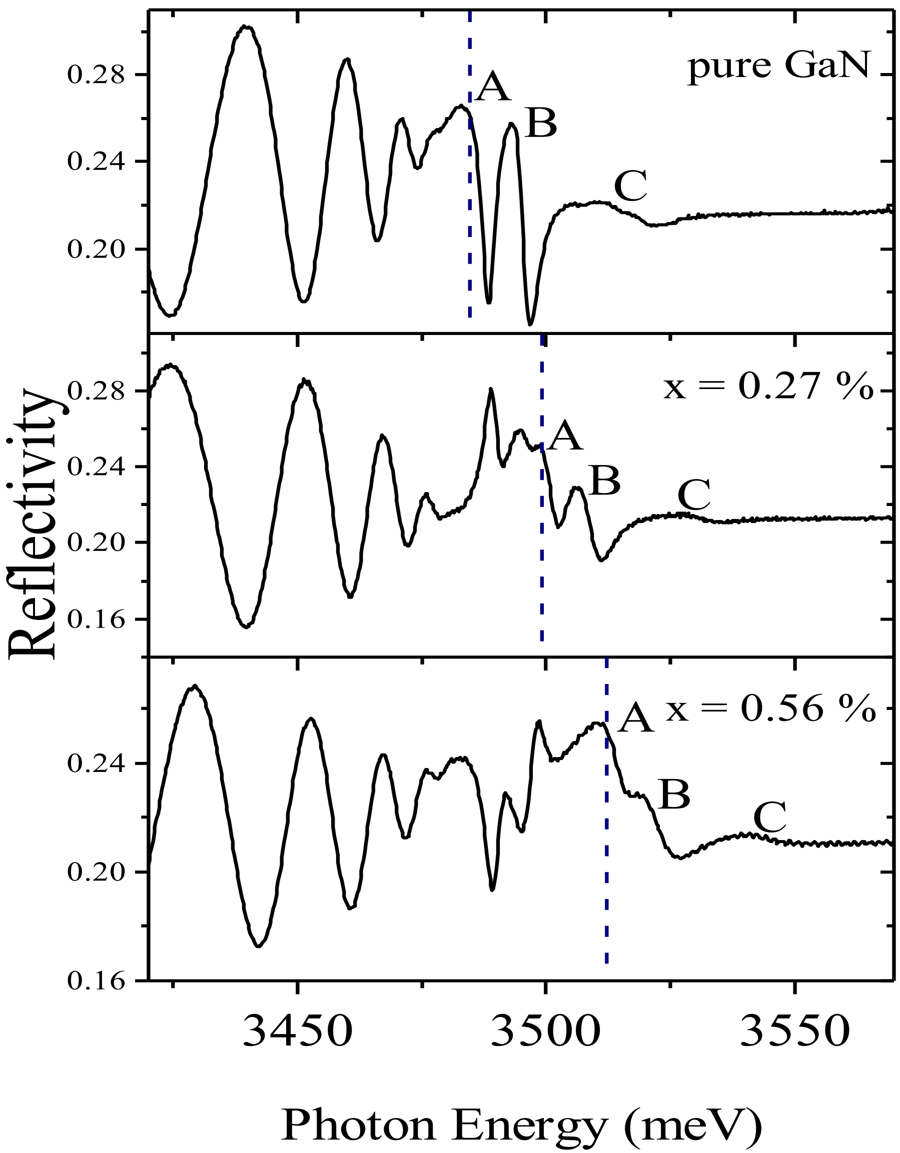


Excitons in GaN:Mn and GaN close spectrally but no strain in the structure

Experimental Setup



Results – zero field reflectivity

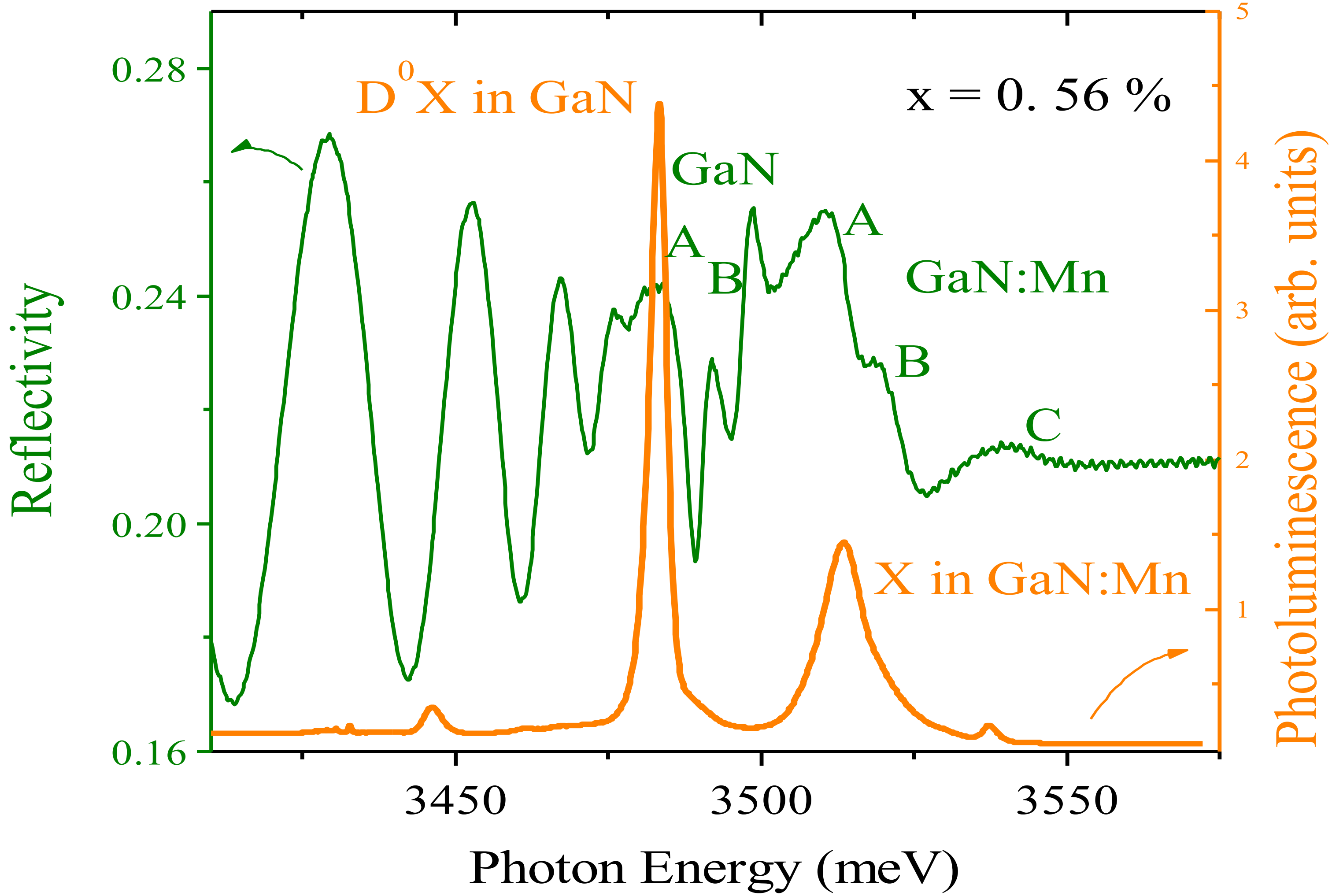


→ Contributions from GaN and GaN:Mn close spectrally and strong influence of interferences

→ Well resolved A, B **and C** excitons in GaN and GaN:Mn

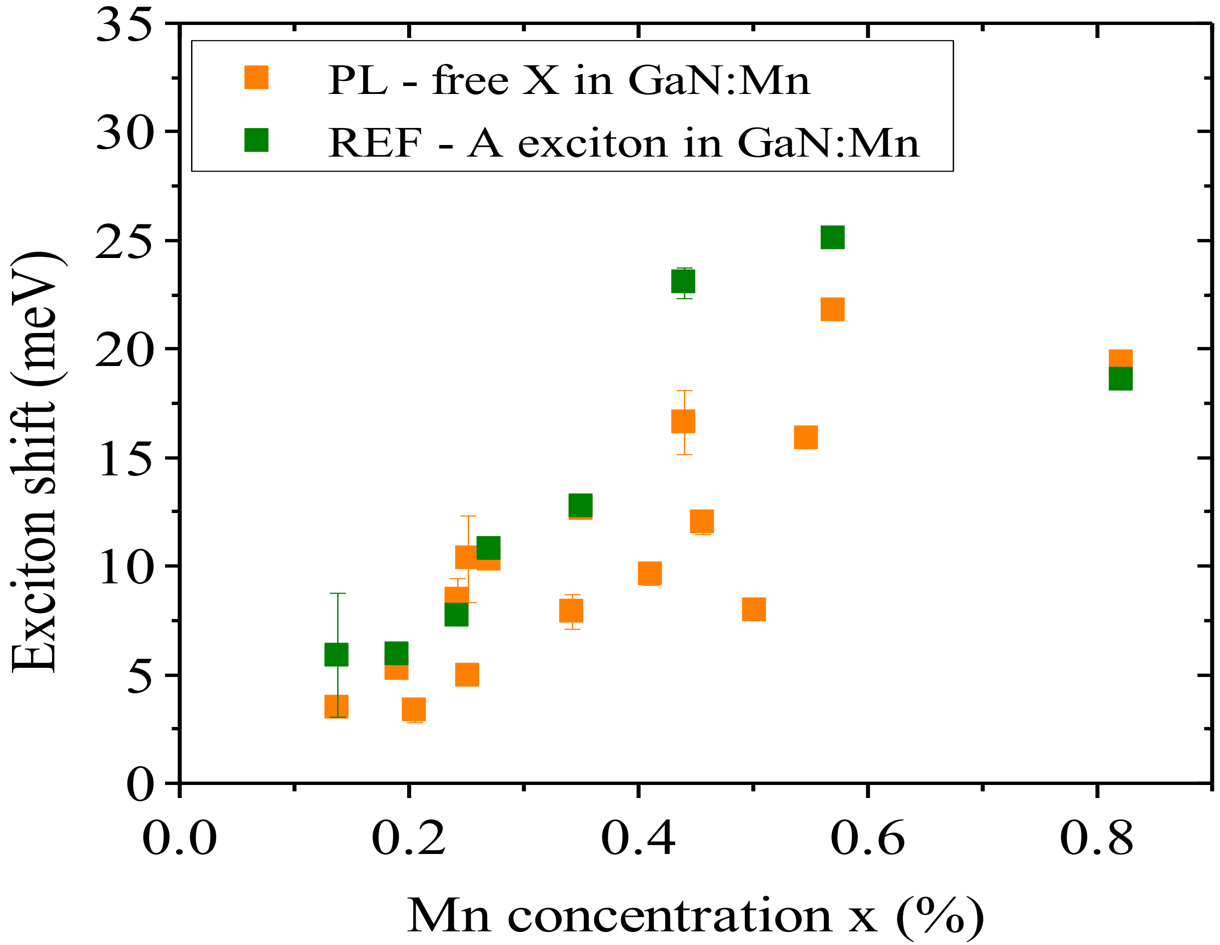
→ Excitons shift towards highier energy when Mn concentration increased

Results – zero field photoluminescence



→Exciton bound to neutral donor in GaN and free excitons in GaN:Mn transitions dominate PL spectra

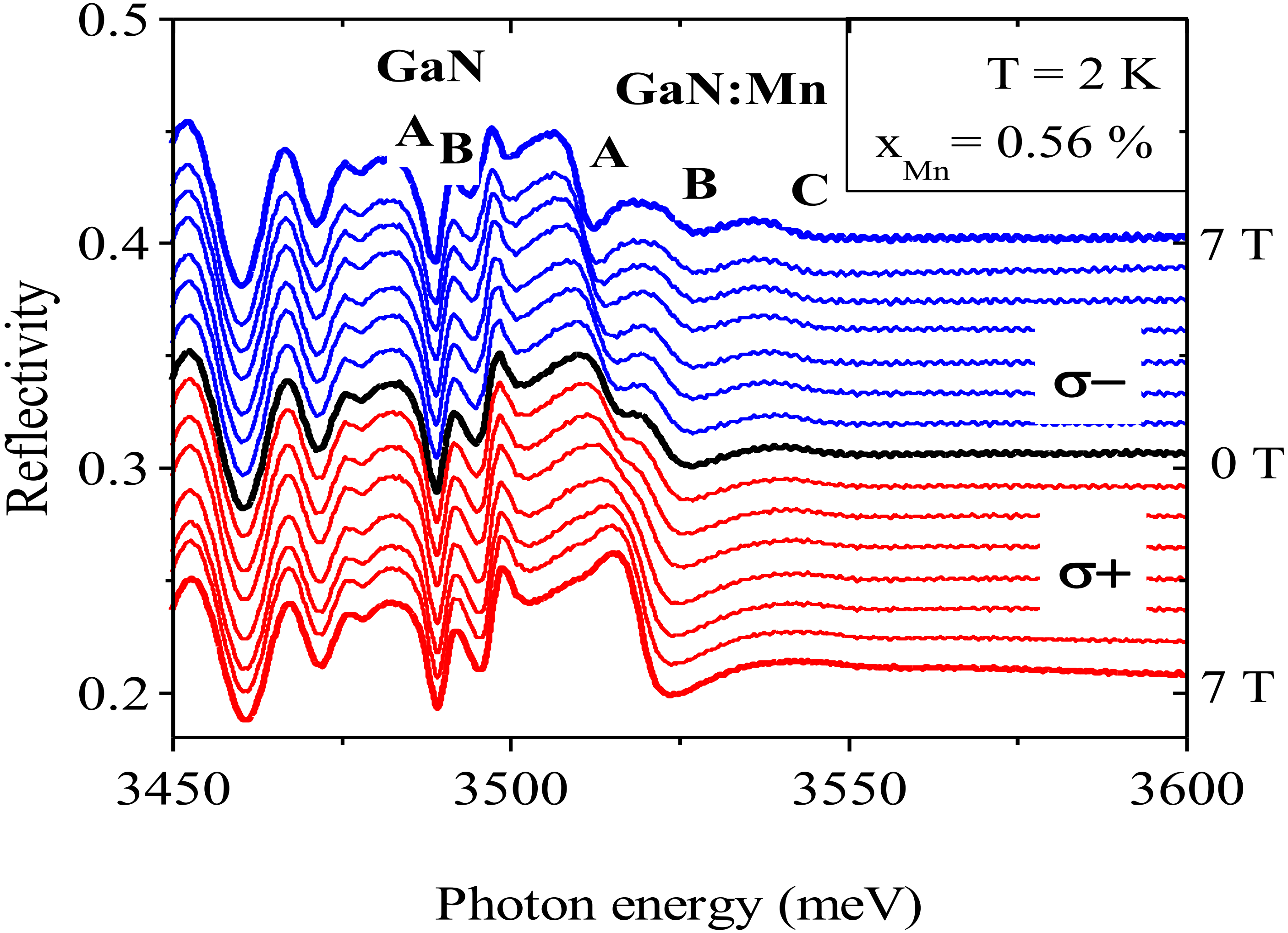
GaN:Mn energy gap vs Mn concentration



→Increase of the GaN:Mn band gap with increasing Mn concentration

→Indication for the strong coupling between holes and Mn spins predicted by theory (Dietl, PRB 2008)

Results - magnetoreflectivity



→ Reflectivity in magnetic field confirms identification of excitonic transitions

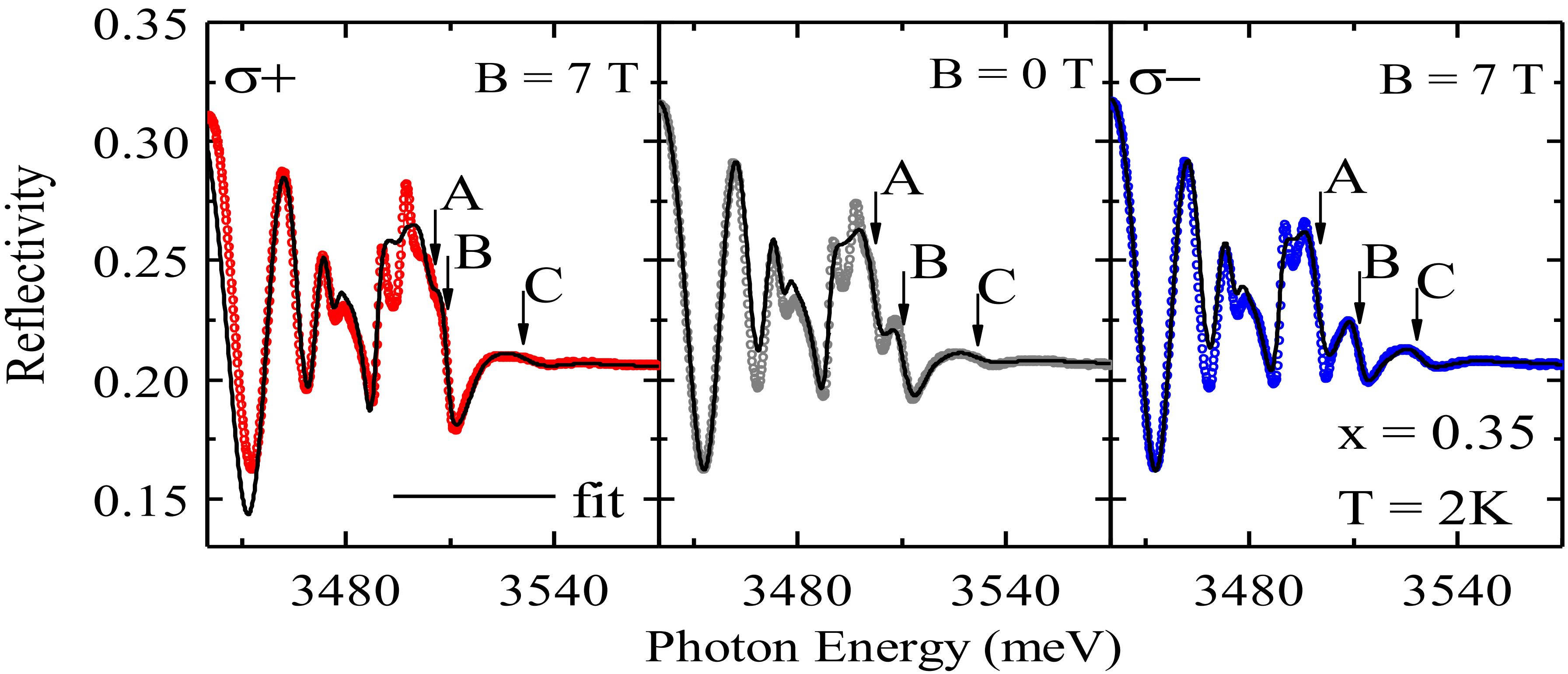
Modelling of the Reflectivity Spectra

Dielectric function for GaN and GaN:Mn layers:

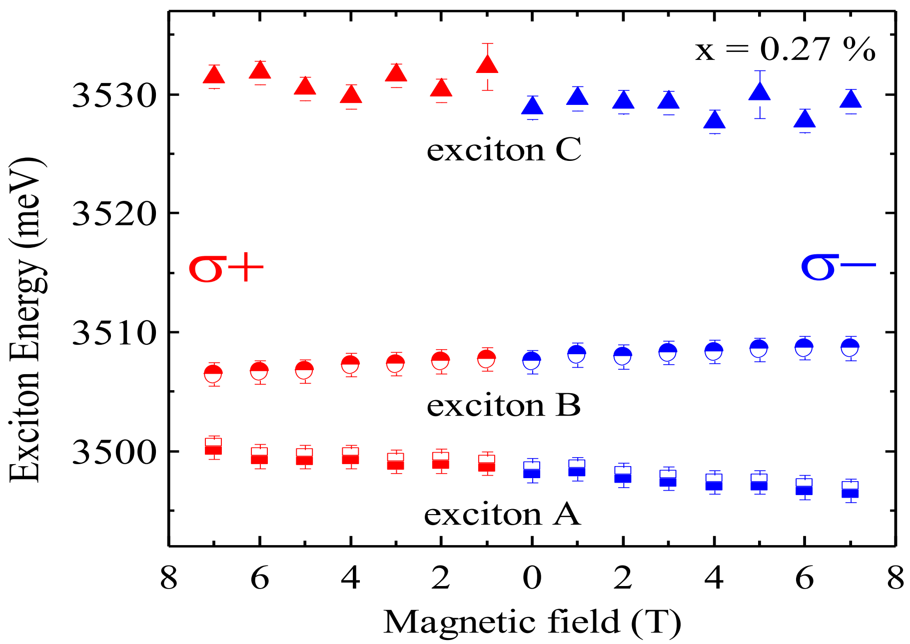
$$\varepsilon_j(E) = \varepsilon_0 + \frac{4\pi \cdot \alpha_{Aj} \cdot E_{Aj}^2}{(E_{Aj} - E)^2 - i \cdot E \cdot \Gamma_{Aj}} + \frac{4\pi \cdot \alpha_{Bj} \cdot E_{Bj}^2}{(E_{Bj} - E)^2 - i \cdot E \cdot \Gamma_{Bj}} + \frac{4\pi \cdot \alpha_{Cj} \cdot E_{Cj}^2}{(E_{Cj} - E)^2 - i \cdot E \cdot \Gamma_{Cj}} +$$

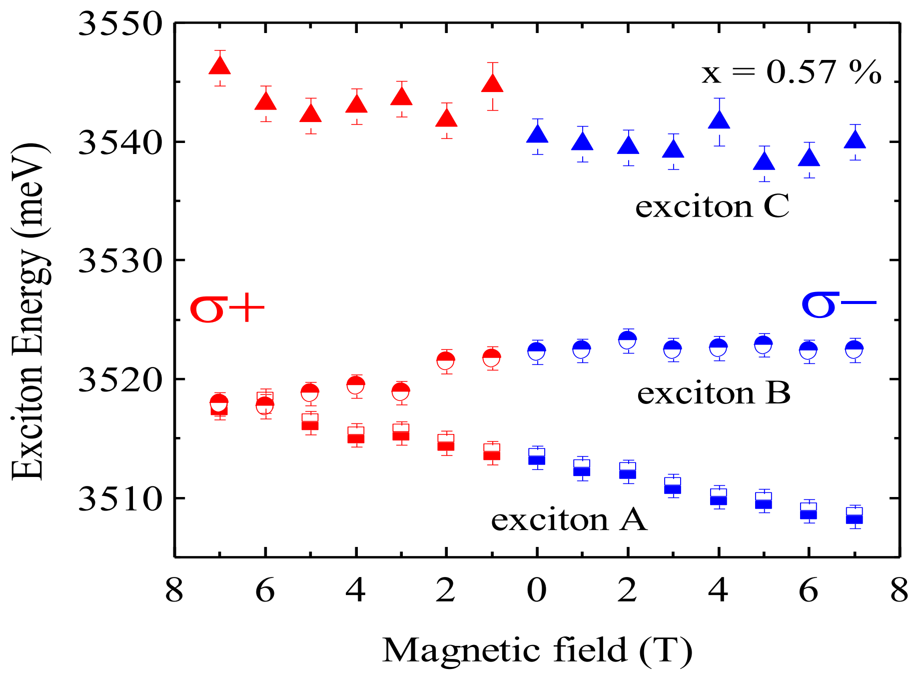
$$+ \textit{excitonic excited states} + \textit{continuum of unbound states}$$

Fitting parameters: energies, widths and polarizabilites of excitons A, B, C



Excitonic splitting in magnetic field





→ Observation of giant Zeeman splitting on A, B and C excitons in GaN:Mn

→Due to anticrossings between excitons, excitonic spliting not proportional to magnetization

Modelling of the of the excitonic positions in magnetic field

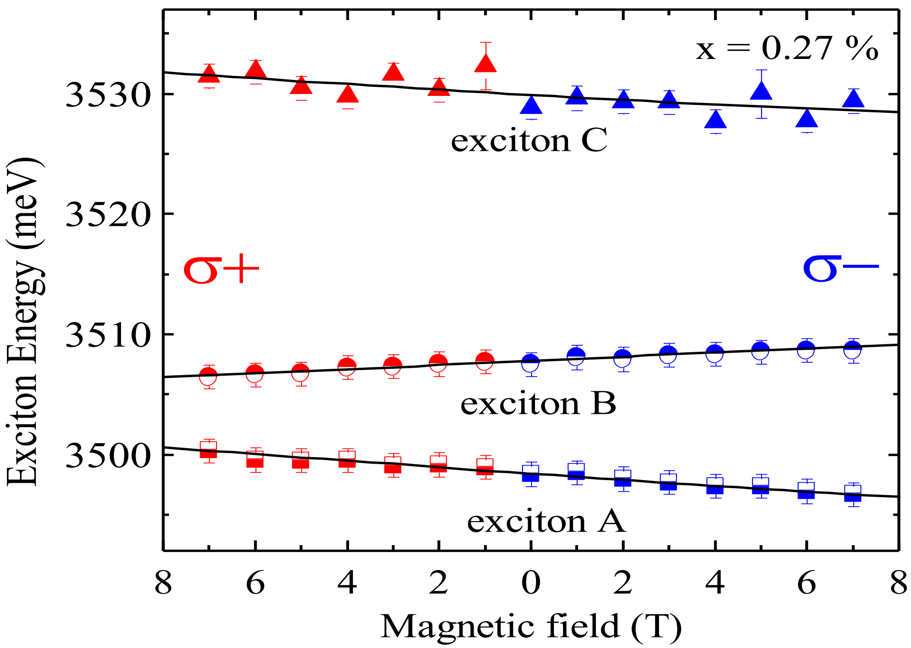
Effective Hamiltonian:

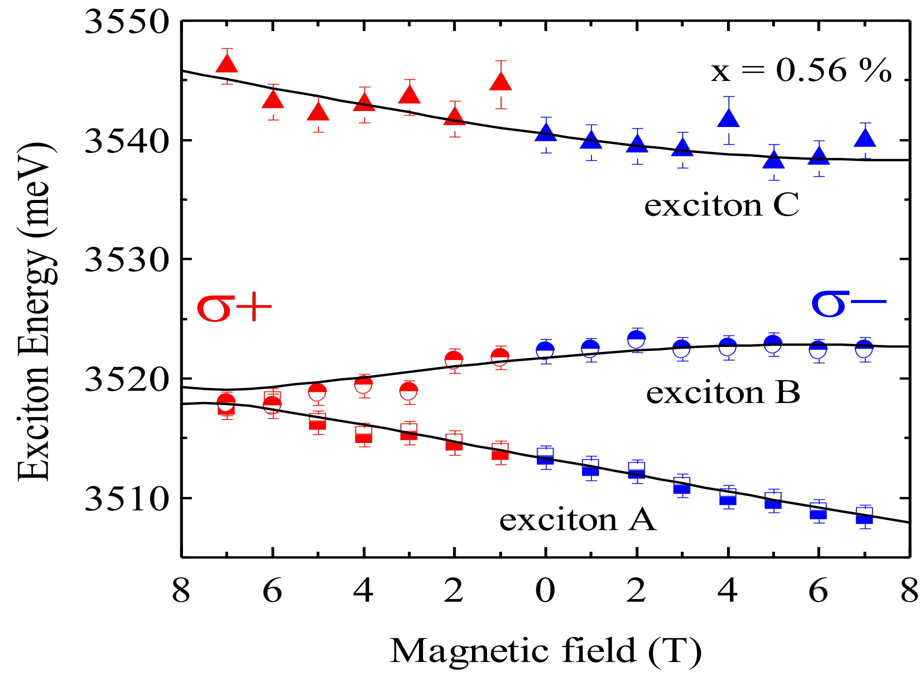
$$H = E_0 + H_V + H_{e-h} + H_{s,p-d} + H_{Zeeman} + H_{dia}$$

Hamiltonian of exchange interaction between Mn^{3+} ions and free carriers:

$$H^{\sigma\pm}_{s,p-d} = \pm\frac{1}{2}N_0 x_{Mn}\langle -S_Z\rangle \begin{pmatrix} \beta-\alpha & 0 & 0 \\ 0 & \alpha-\beta & 0 \\ 0 & 0 & \alpha+\beta \end{pmatrix}$$

Excitonic splitting in magnetic field

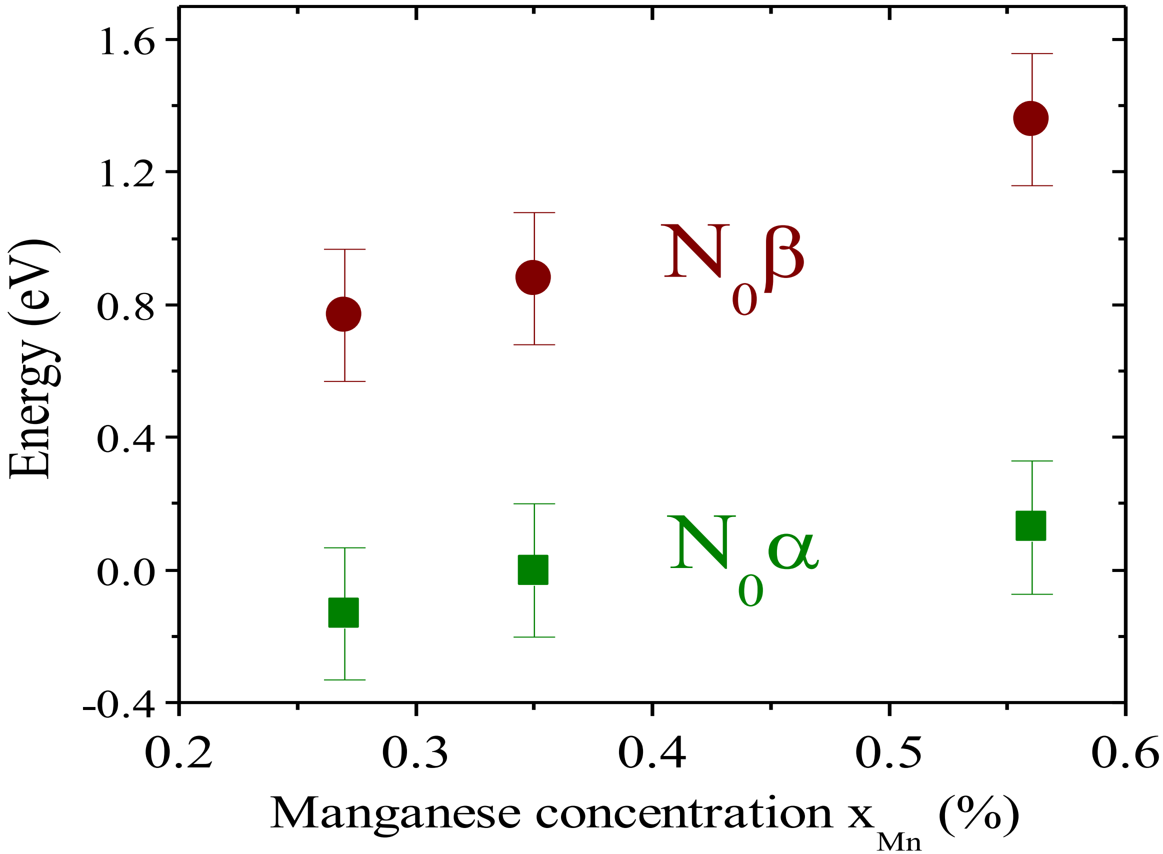




→Free parameters of the fit: $N_0\alpha$, $N_0\beta$, band gap energy, Δ_1, Δ_2

→Quantitative description of excitonic shifts in magnetic field

Apparent exchange constants



$N_0(\beta-\alpha)$=1.2 eV, Pacuski, PRB'07

Apparent:

- $N_0\beta$ positive → delocalized hole-Mn^{3+} ion exchange interaction ferromagnetic
- $N_0\alpha$ small → small effective strength of exchange interaction electron-Mn^{3+} ion

→ localizing potential of the Mn^{3+} ion (Dietl, PRB'08)

Summary

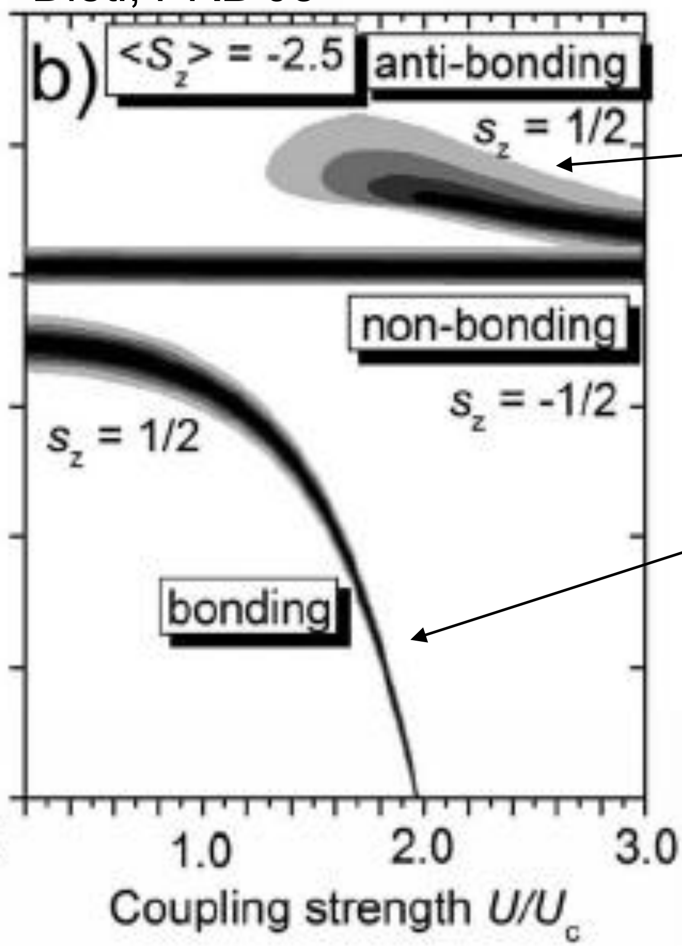
- Increase of the GaN:Mn band gap with increasing Mn content
- Mn^{3+} – band hole exchange interaction ferromagnetic

→ Strong coupling between hole and Mn^{3+} ion

- Observation of giant Zeeman splitting of A, B **and C** excitons in GaN:Mn

Strong coupling regime

Dietl, PRB'08



$N_0\beta$ positive – ferromagnetic interaction

$N_0\beta$ negative – antiferromagnetic interaction

Hv

Valence band at *k* = 0:

$$H_v = \begin{pmatrix} -\Delta_2 & 0 & 0 \\ 0 & \Delta_2 & -\sqrt{2}\Delta_3 \\ 0 & -\sqrt{2}\Delta_3 & \Delta_1 \end{pmatrix}$$

Δ_1 – trigonal component of crystal field and biaxial strain
Δ_2 , Δ_3 – anisotropic spin-orbit interactions