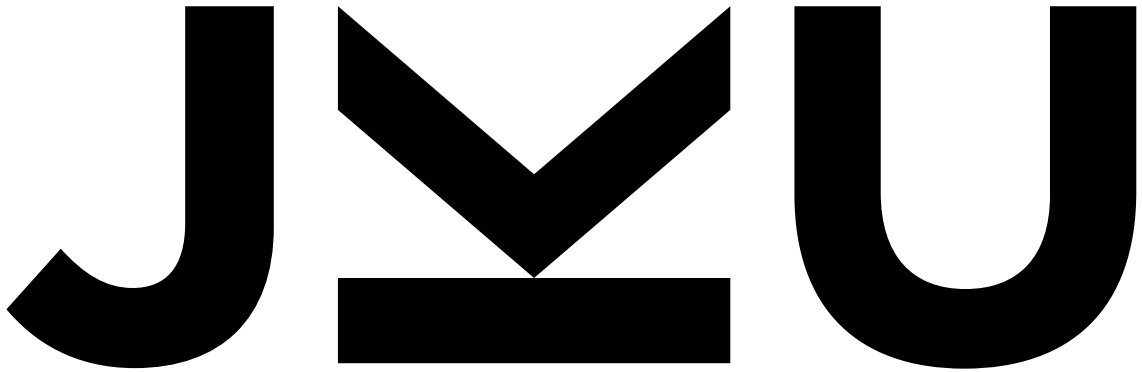

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Charge states of transition metal ions and local magnetic structure of dilute magnetic semiconductor (Ga,Fe)N:Mn – an emission Mössbauer spectroscopy study

Proposal: INTC-P-692

Rajdeep Adhikari[1], Haraldur Pall Gunnlaugsson, Hilary Masenda, Arthur Ernst, Bogdan Faina, Juliana Schell, Alberta Bonanni and the Mössbauer Collaboration ISOLDE/CERN

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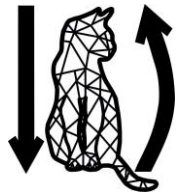
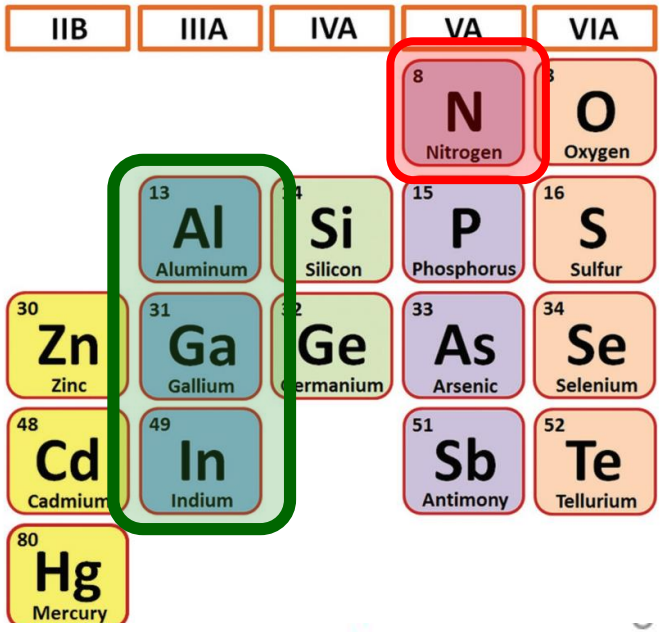
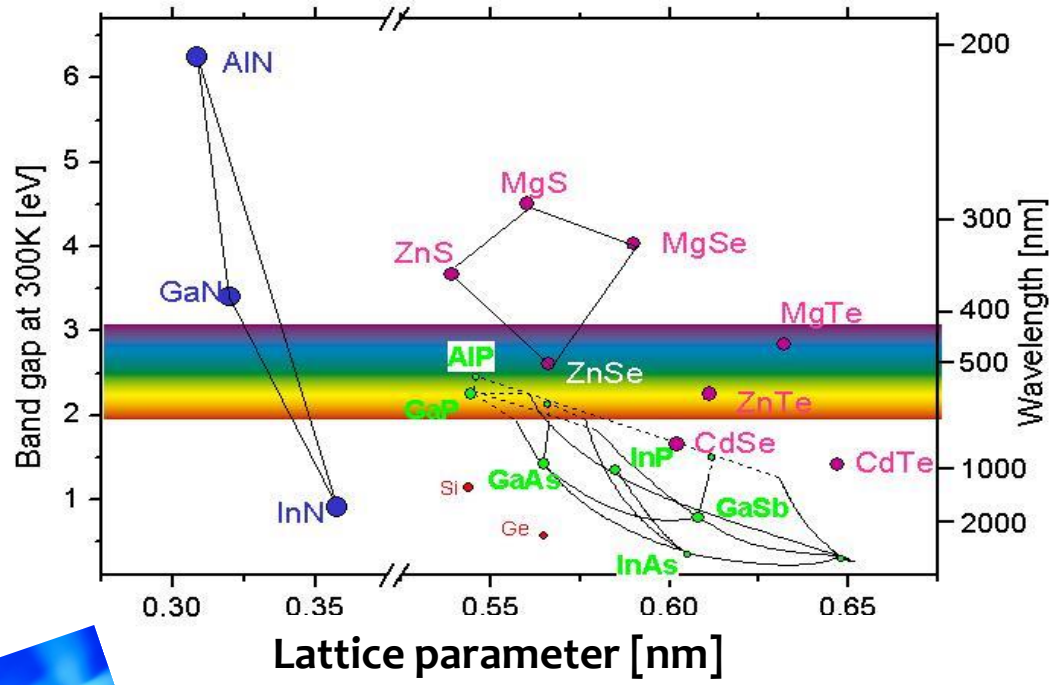
HFP | Institut für Halbleiter- und Festkörperphysik

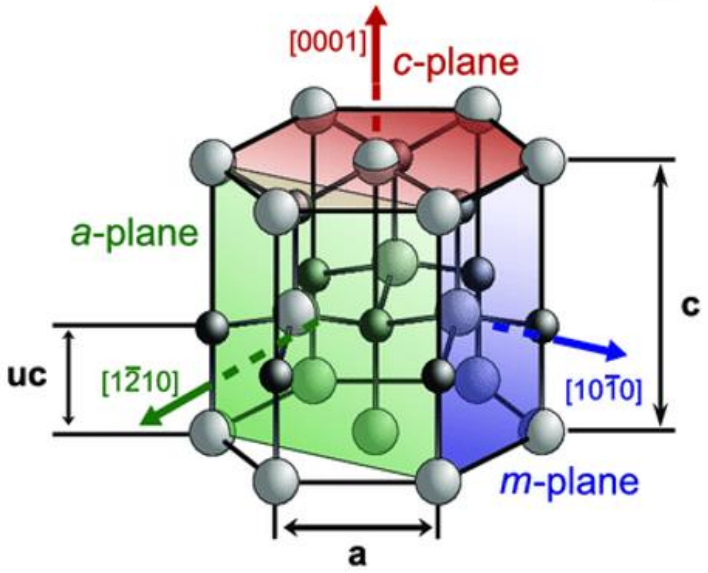
Collaborations and funding

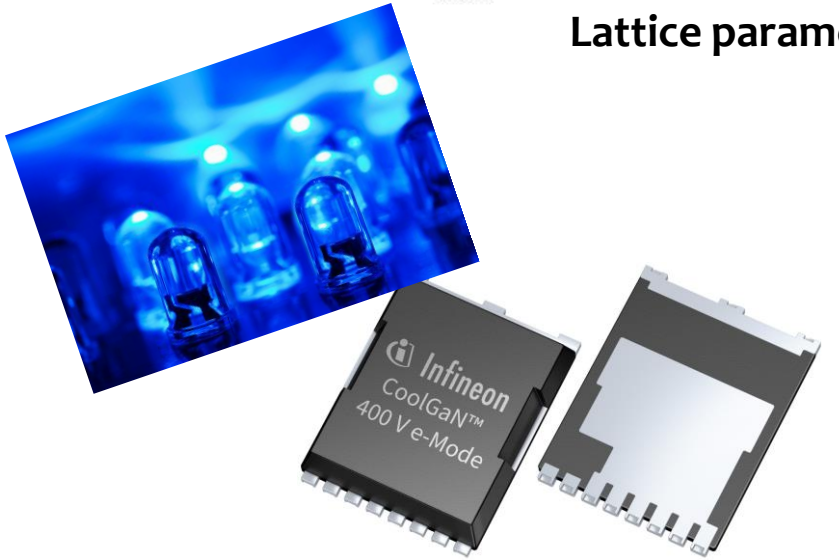


III-nitride compounds





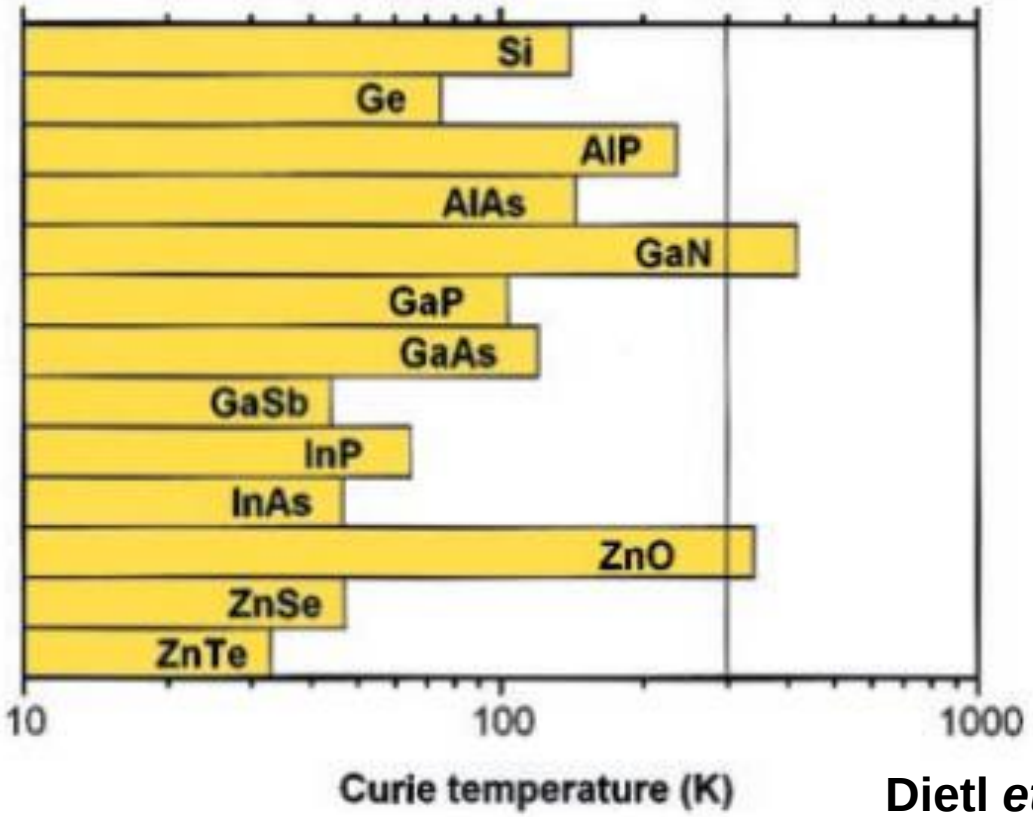




Doping of III-nitride semiconductors: Dilute magnetic semiconductors (DMS)

Zener Model Description of Ferromagnetism in Zinc-Blende Magnetic Semiconductors

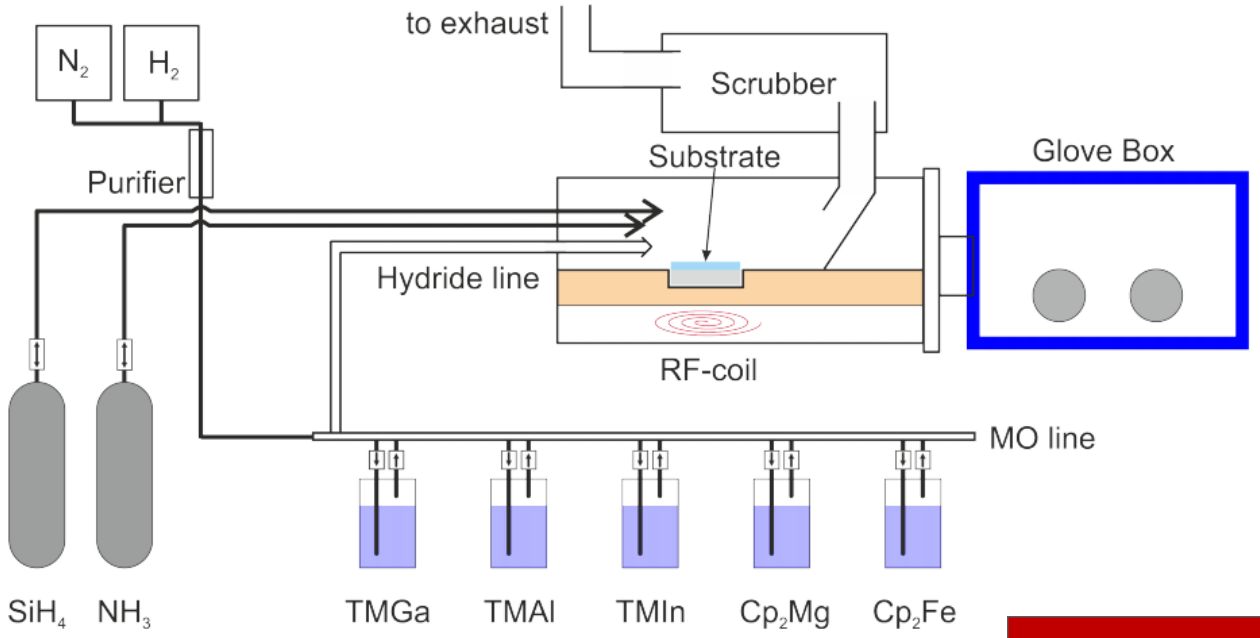
T. Dietl,[1,2]* H. Ohno,[1]* F. Matsukura,[1] J. Cibert,[3] D. Ferrand[3]

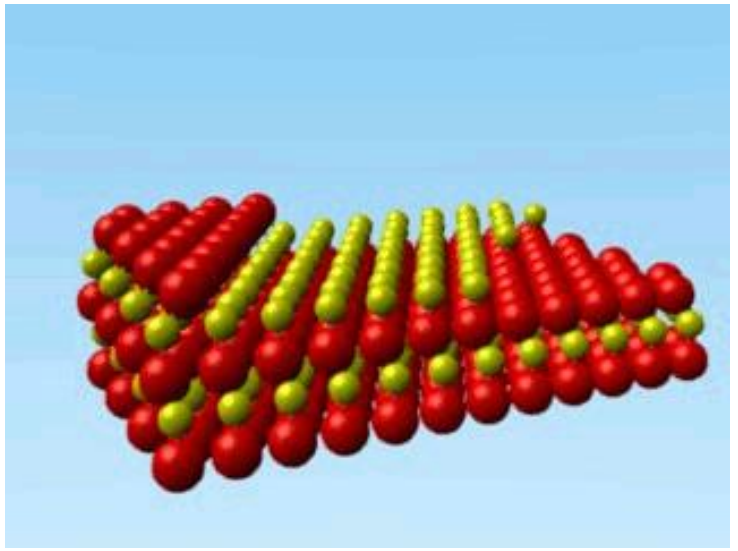


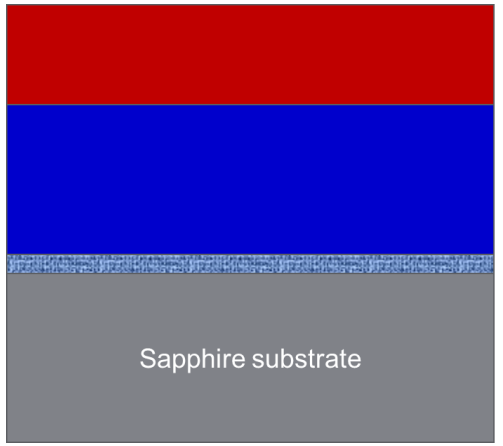
- Carrier mediated ferromagnetism
- Free holes

Dietl *et* al. Science (2000)

Growth: Metal organic vapor phase epitaxy (MOVPE)





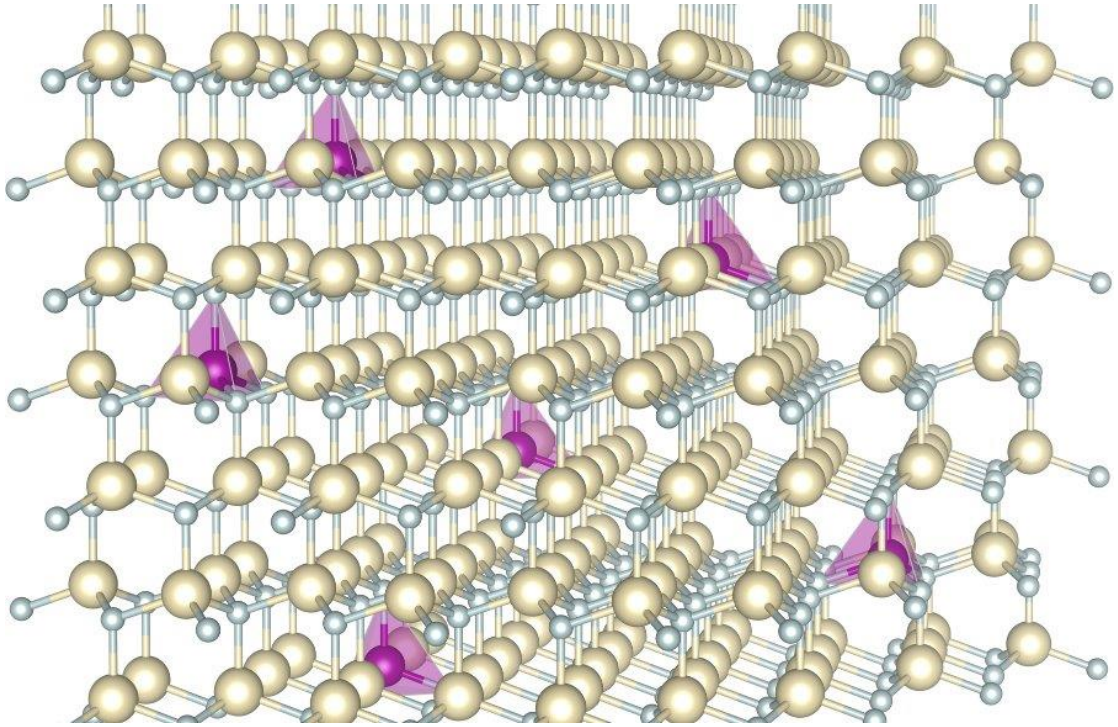


Active layer

Buffer layer (1040°C for GaN/1150°C for AlGaN)

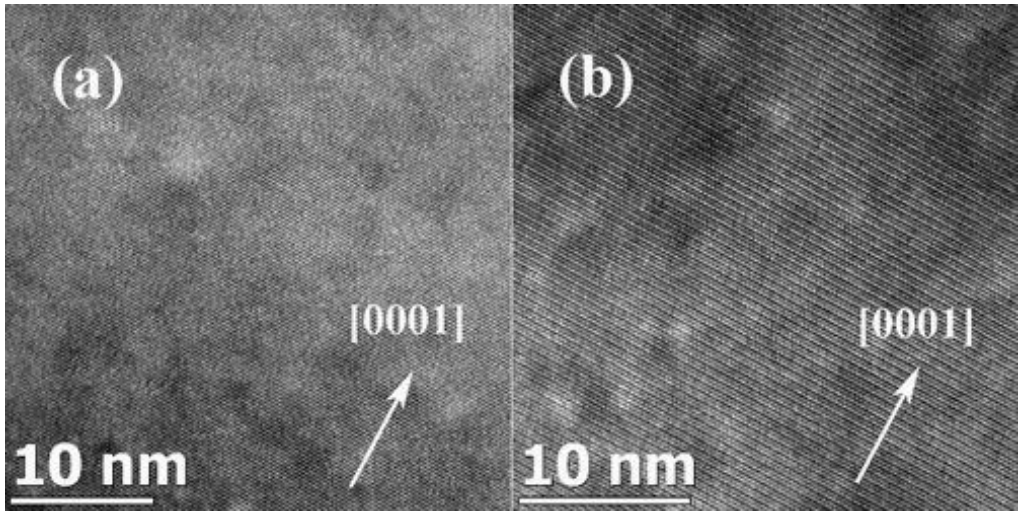
Low temperature nucleation layer
(540°C for GaN/800°C for AlN)

Doping of III-nitride semiconductors: Magnetic



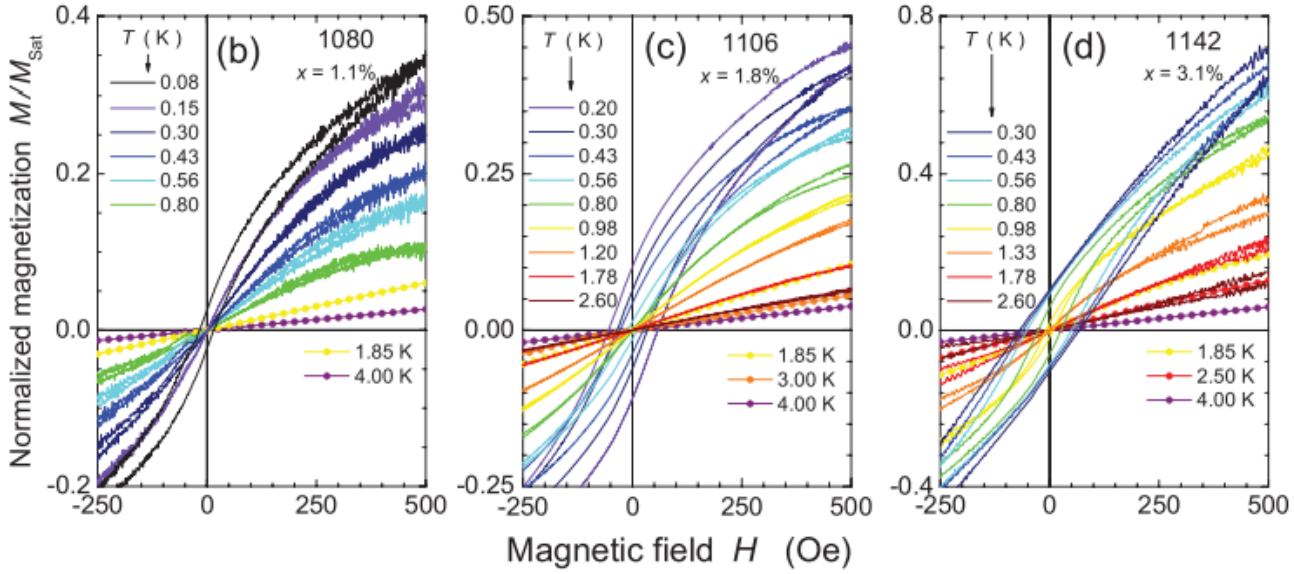
(Ga,Mn)N or (Ga,Fe)N
Dilute magnetic semiconductor (DMS)

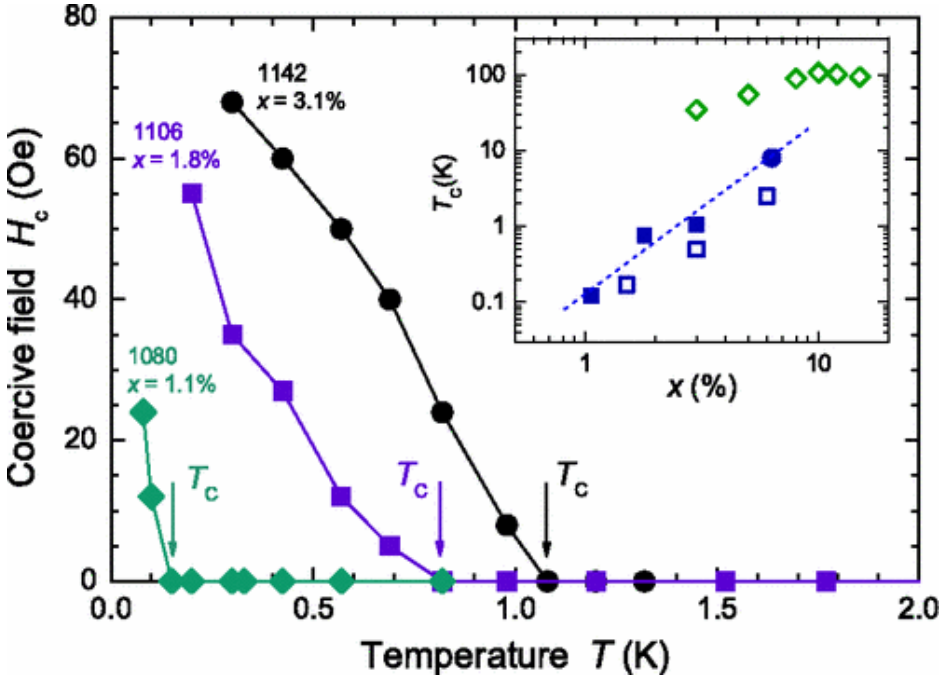
- Growth mode: homogeneous doping
- Substitutional
- Upto 3.1% Mn doping
- Upto 0.6% Fe doping



No free holes !!

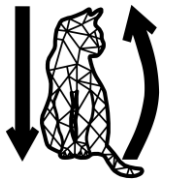
(Ga,Mn)N



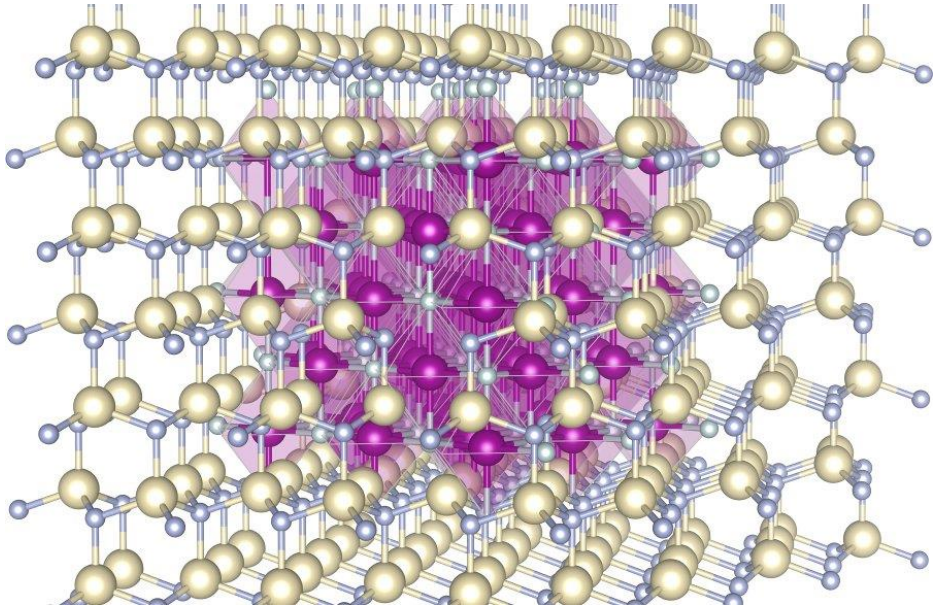


- Ferromagnetism in 3.1% (Ga,Mn)N ~1.2 K
- Homogeneous distribution – upto 95% substitutional Mn in Ga sites
- Charge state: Mn^{3+}
- Superexchange mechanism for ferromagnetism
- No free holes

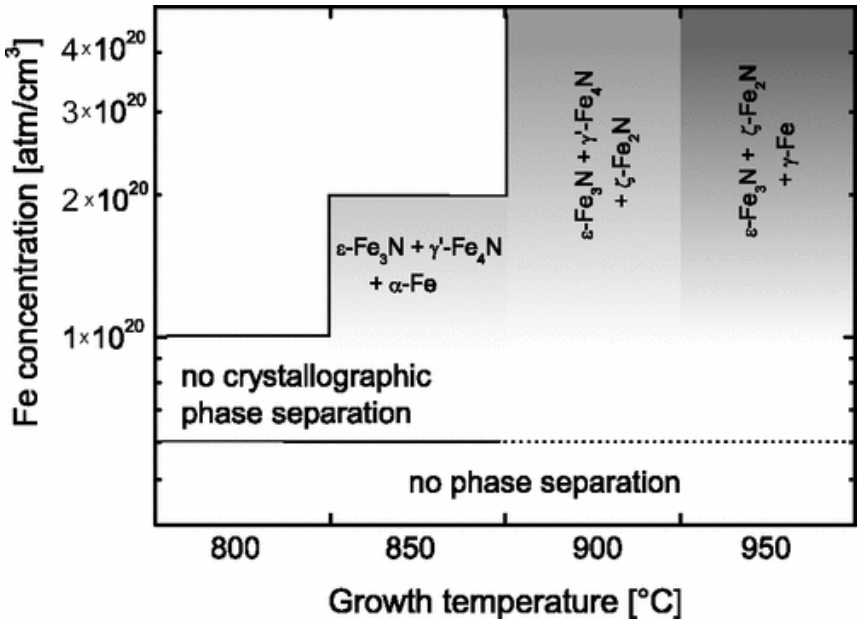
M. Sawicki *et al*. PRB (2012)



(Ga,Fe)N

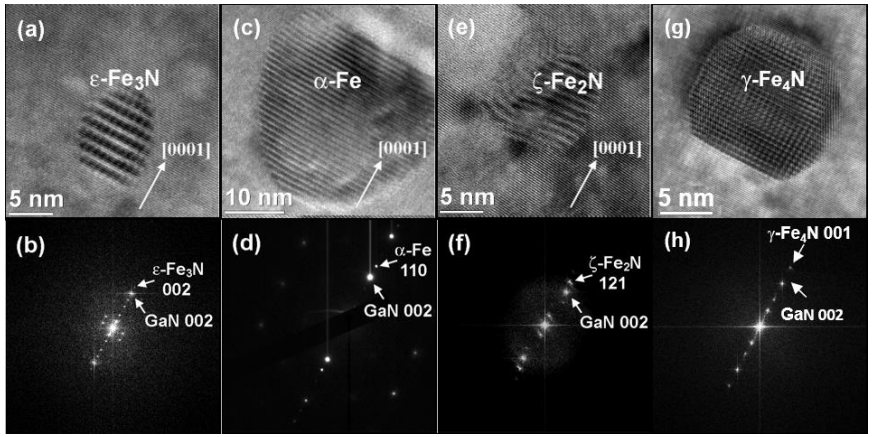


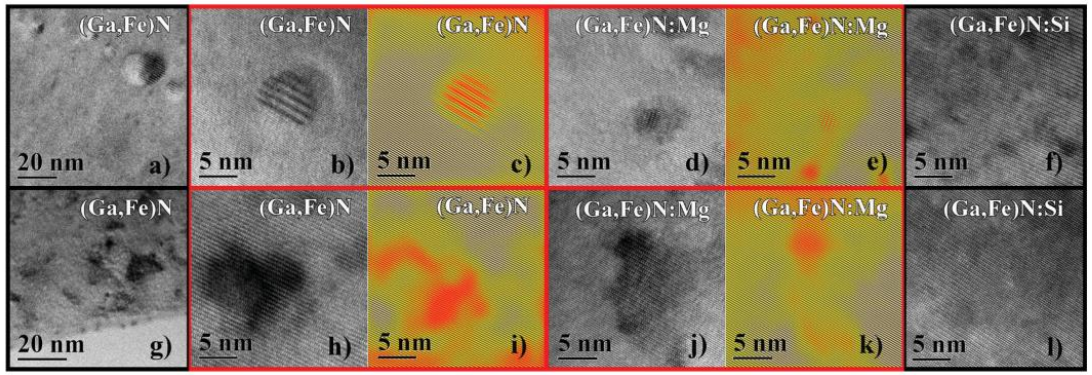
GaN:Fe – Fe_4N nanocrystals
Condensed magnetic semiconductor (CMS)

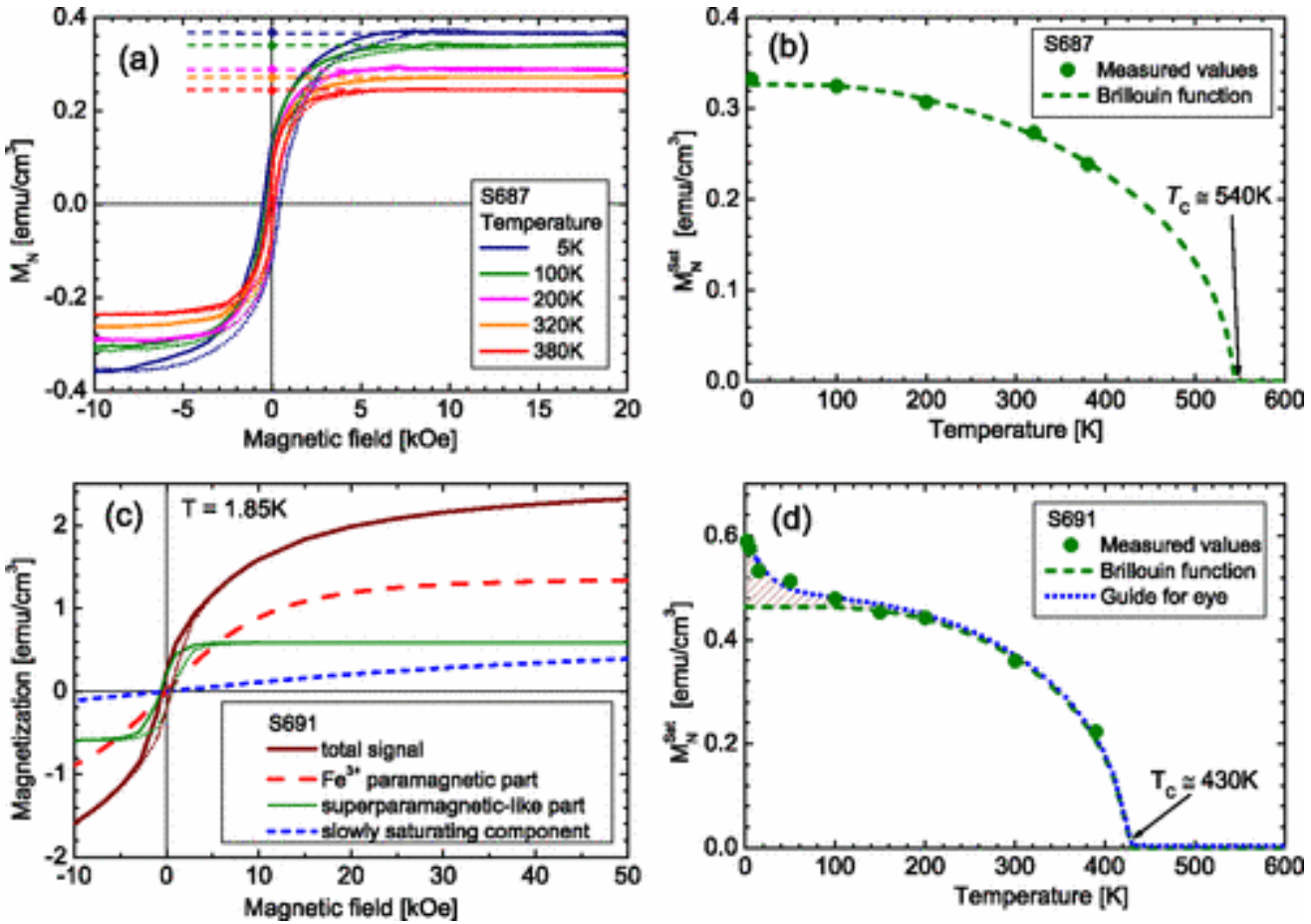


- Growth mode: Homogeneous/Delta doping
- Precipitation of secondary phases
- Above 0.6% Fe doping
- Growth temperature
- No precipitation for Mn doping

No free holes !!







Bonanni *et* al. PRL (2008)
A. Navarro-Quezada *et al.* PRB (2010)

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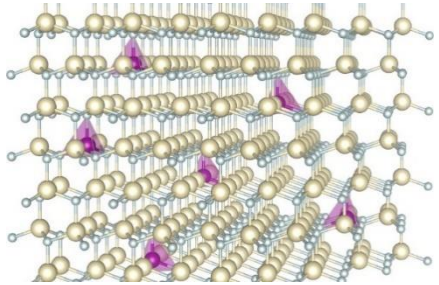
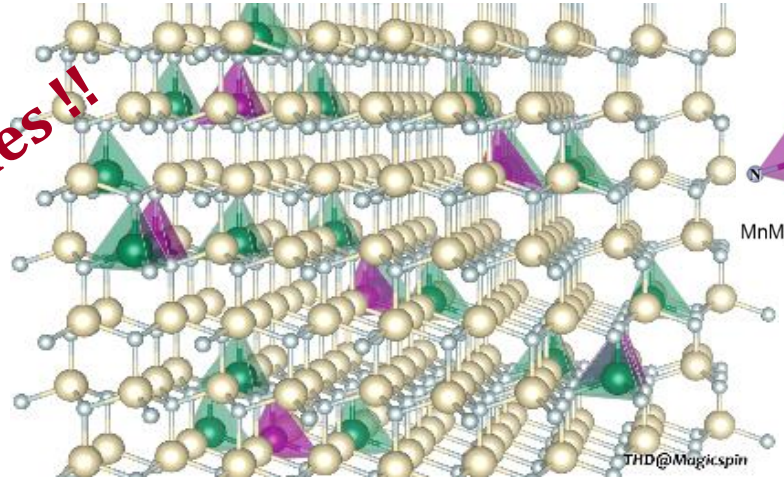
Co-doping: Dilute magnetic semiconductors (DMS)



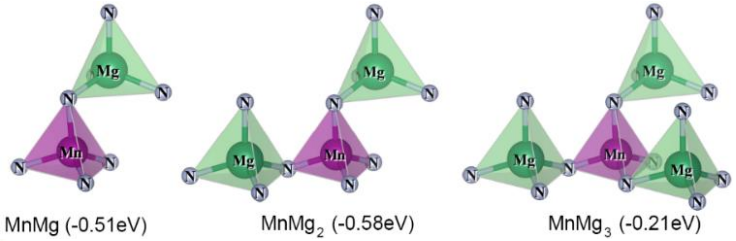
GaN:Mn

GaN:Mg

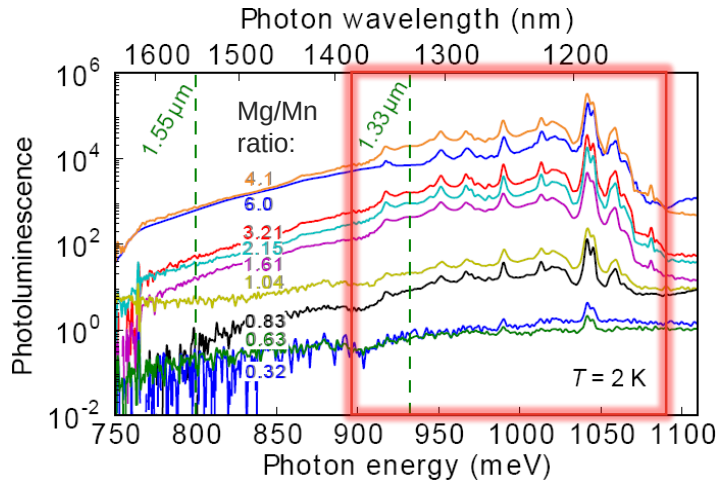
Still no free holes !!

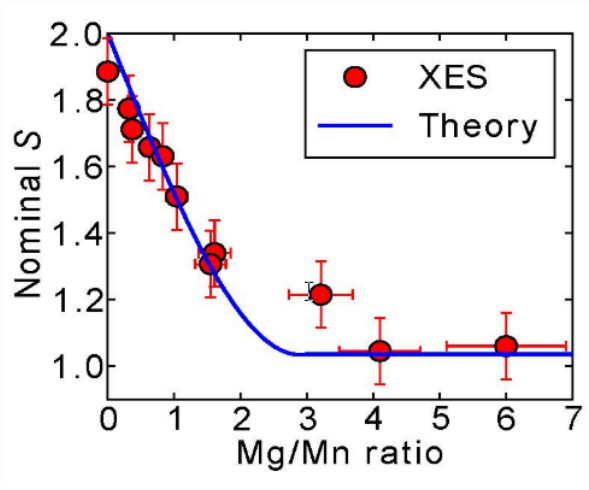


GaN:(Mn,Mg)



Complexes

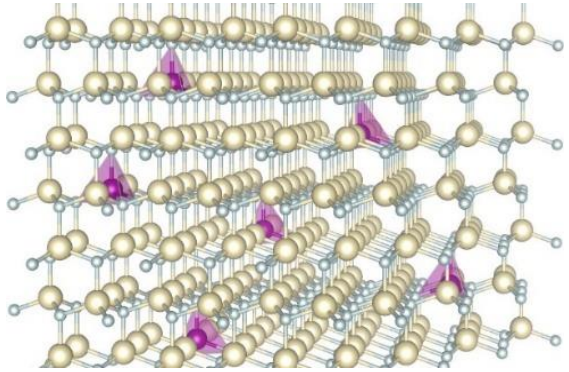




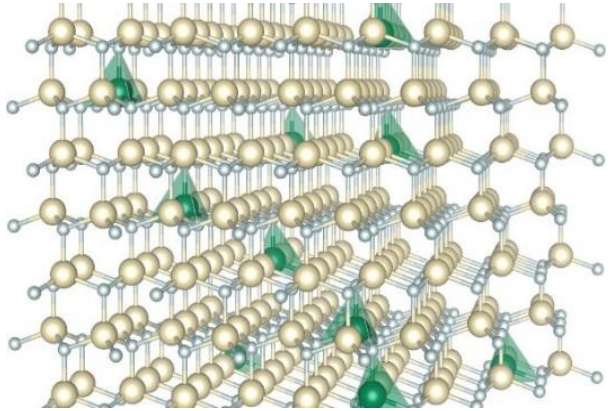
Devillers *et* al. Sci. Rep (2012)

Charge state of Mn

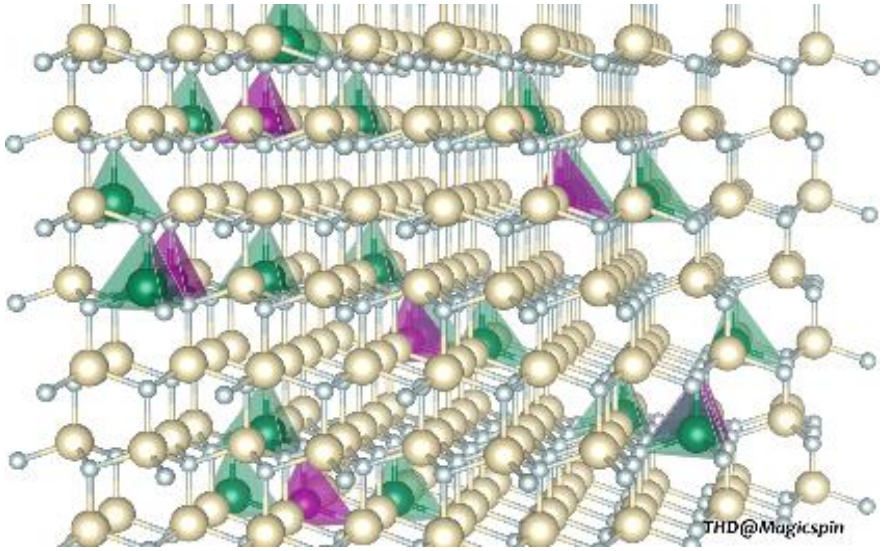
- GaN:Mn → $S = 2$, Mn^{3+}
- GaN:(Mn,Mg), Mn-Mg_1 → $S = 3/2$, Mn^{4+}
- GaN:(Mn,Mg), Mn-Mg_k, $k > 1$ → $S = 1$, Mn^{5+}



GaN:Mn

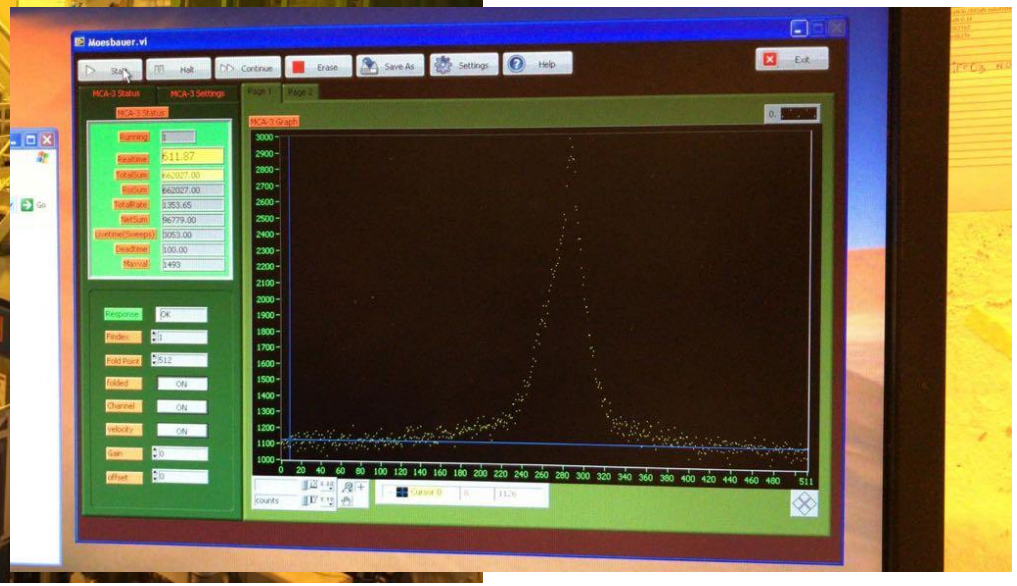


GaN:Fe



GaN:(Mn,Fe)

Co-doping of GaN with Mn and Fe – largely missing !!



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Emission Mössbauer spectroscopy: What we learned from IS-630 and IS-576 ?

IOP Publishing New J. Phys. 24 (2022) 103007 https://doi.org/10.1088/1367-2630/ac9499

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PAPER

Unusual charge states and lattice sites of Fe in $Al_xGa_{1-x}N$:Mn

OPEN ACCESS

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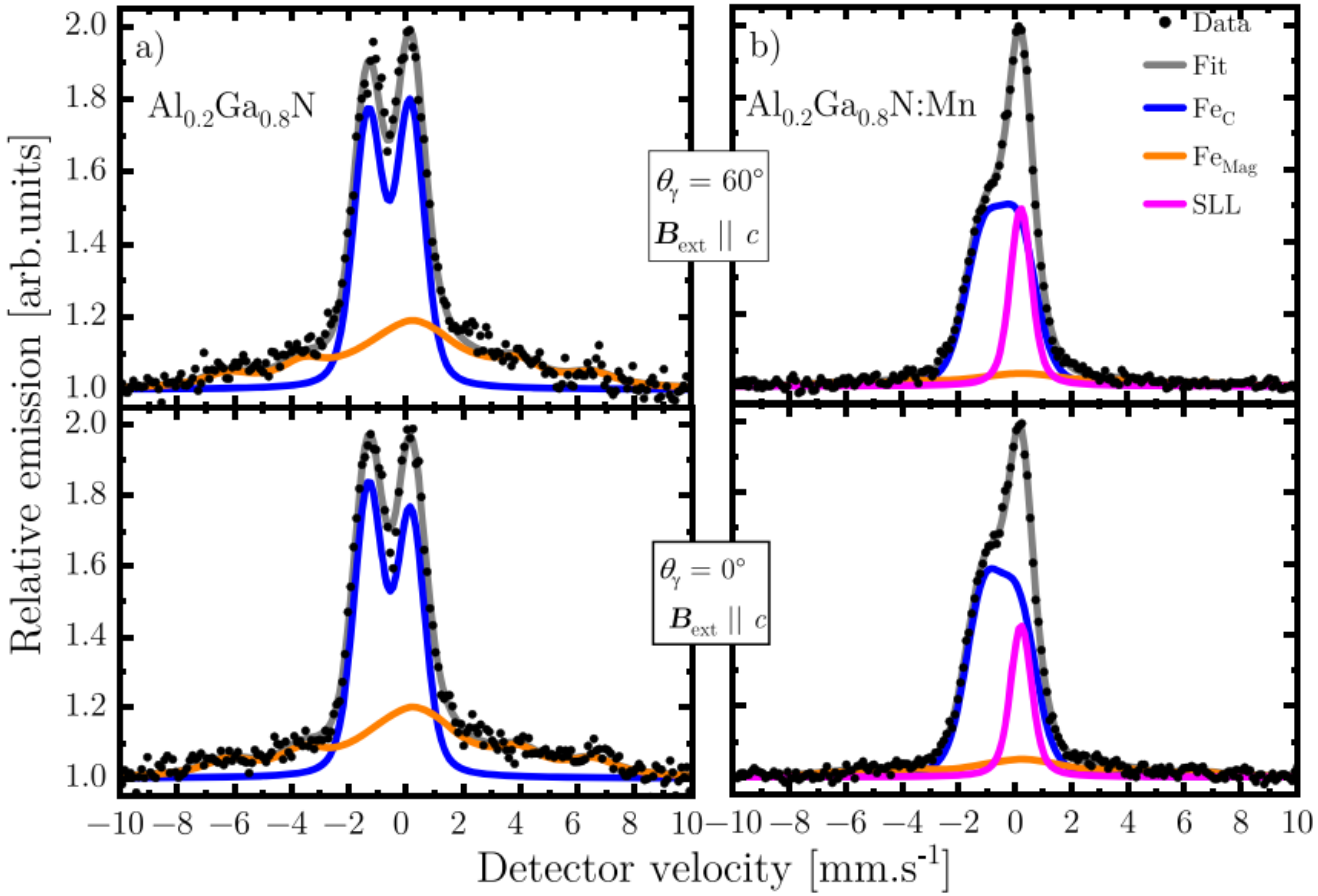
Hilary Masenda[1,2,*], Haraldur Páll Gunnlaugsson[3], Rajdeep Adhikari[4], Krish Bharuth-Ram[5,6], Deena Naidoo[1], Aitana Tarazaga Martín-Luengo[4], Iraultza Unzueta[7], Roberto Mantovan[8], Torben Esmann Mølholt[9], Karl Johnston[9], Juliana Schell[9,10], Adeleh Mokhles Gerami[11], Petko Krastev[12], Bingcui Qi[3], Sveinn Ólafsson[3], Haflidi Pétur Gíslason[3], Arthur Ernst[13,14] and Alberta Bonanni[4]



- Substitutional Fe^{3+} state
- Fe^{2+} state associated with V_N
- In Mn doped AlGaN: Unusual charge state of Fe

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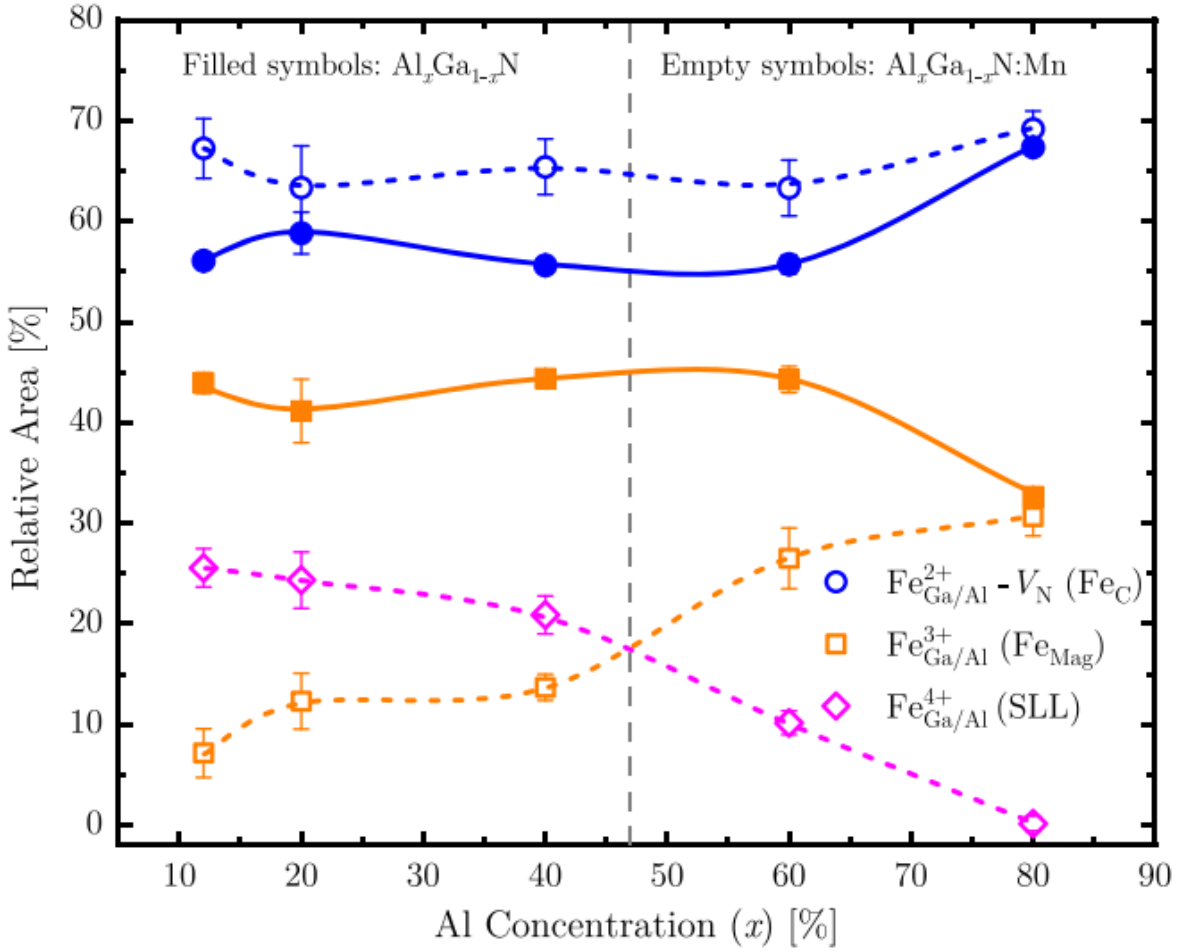

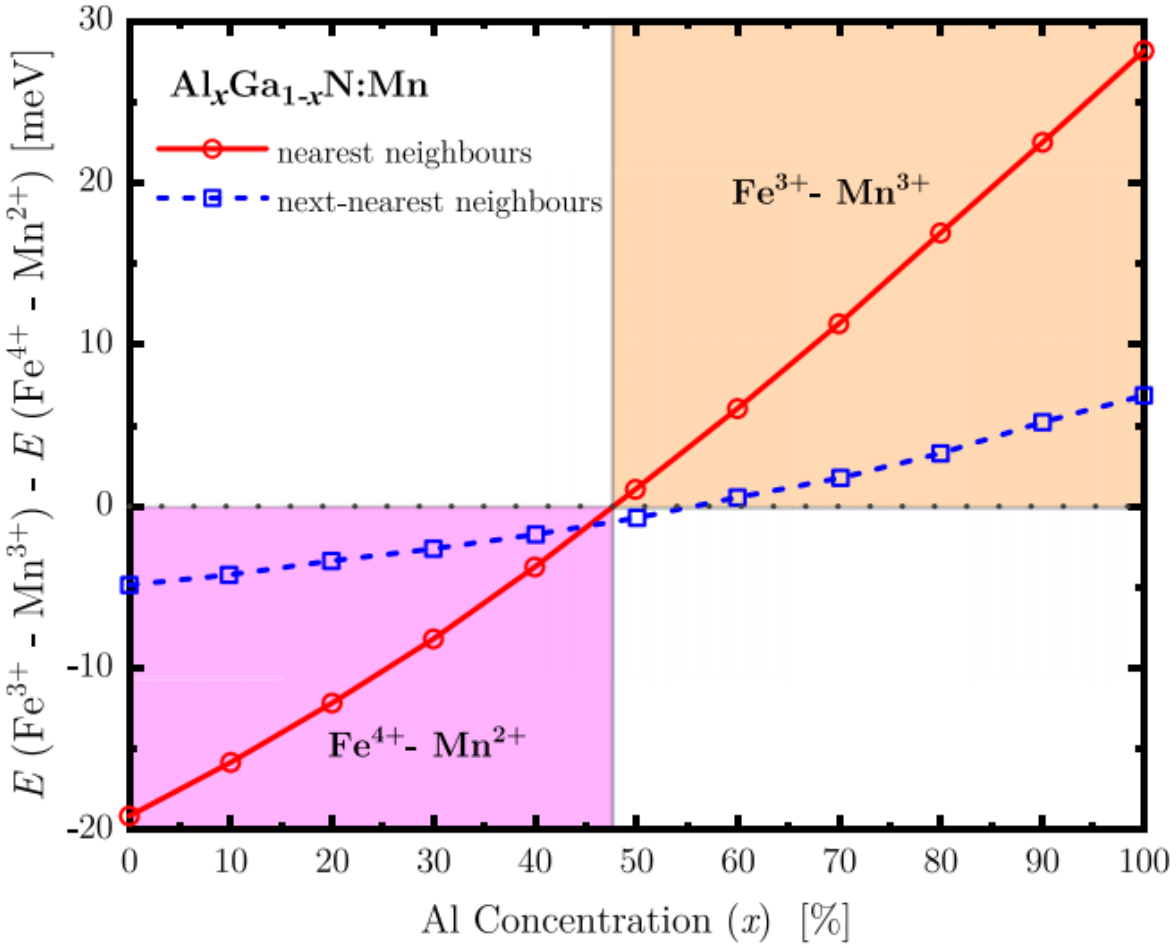

Result: co-doping promotes unusual charge states Fe^{4+}-Mn^{2+} which is pronounced in low AlGaN:Mn with low Al (<50%)

Motivation for the proposal P-692

- Investigation of magnetic properties in co-doped (Ga,Fe)N:Mn
- Fe^{4+}-Mn^{2+} mixed charge states: route to double exchange mechanism in the DMS
- Enhanced strength of magnetic interaction: increased T_c ??
- Would co-doping promote precipitation or Mn-Fe complexes?
- to elucidate the effect of Mn on the suppression of segregation of Fe_xN NC phases in (Ga,Fe)N with Fe > 0.5%

Sample series	x_{Fe} (at. %)	x_{Mn} (at. %)
S1	0.1%	0.5; 1.0; 3.0
S2	0.4%	0.5; 1.0; 3.0
S3	1.0%	0.5; 1.0; 3.0
S4	2.0%	0.5; 1.0; 3.0

Proposed experiments and shifts

			Emission Mössbauer Spectroscopy				
Required isotope	Implanted beam	Type of experiment	Approx. intensity (at/μC)	Target/ion source	Reqd. atoms/sample	Comments	No. of requested shifts
^{57}Mn	^{57}Mn	eMS (M1; M2)	10^{8}	UC_x; RILIS	1 x 10^{12}	Measurements at 100 K ≤ T ≤ 600 K Also as a function of magnetic field achieved using a permanent magnet	12

M1: eMS spectra of the (Ga,Fe)N:Mn samples implanted with ^{57}Mn range (100-600) K

M2: At every *T*, magnetic field and angular dependent eMS measurements will be conducted

Proposed experiments and shifts

Sample series	Type of experiment	No. of requested shifts	Comments
S1	eMS (M1, M2)	3	Hot runs, angular dependences and cold runs
S2	eMS (M1, M2)	3	
S3	eMS (M1, M2)	3	
S4	eMS (M1, M2)	3	

Complementary research

- Structural characterization: XRD, HRTEM
- SQUID magnetometry
- EPR, FMR
- Optical characterization
- X-ray spectroscopy at synchrotron facilities (SOLEIL and Elettra)
- DFT calculations – complementing the eMS experimental data

Expected outcomes and outlook

- Determination of lattice site occupancy, the charge and the spin states of Mn and Fe ions in dilute (Ga,Fe)N:Mn;
- Nature of magnetic interaction in dilute (Ga,Fe)N:Mn – magnetic co-doping as a viable route towards DMS;
- to identify the signatures of Fe_m-Mn_k magnetic complexes, if any, are formed for Mn, Fe co-doped GaN epitaxial layers;
- Mixed charge states – beyond superexchange in GaN DMS?
- Further co-doping with Mg (holes) and/Si (electrons)

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