Can Ferromagnetism in Mn doped GaN be Carrier Mediated?

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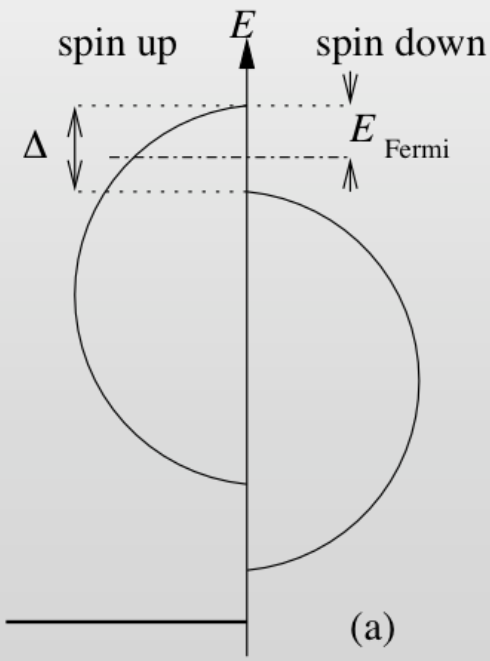
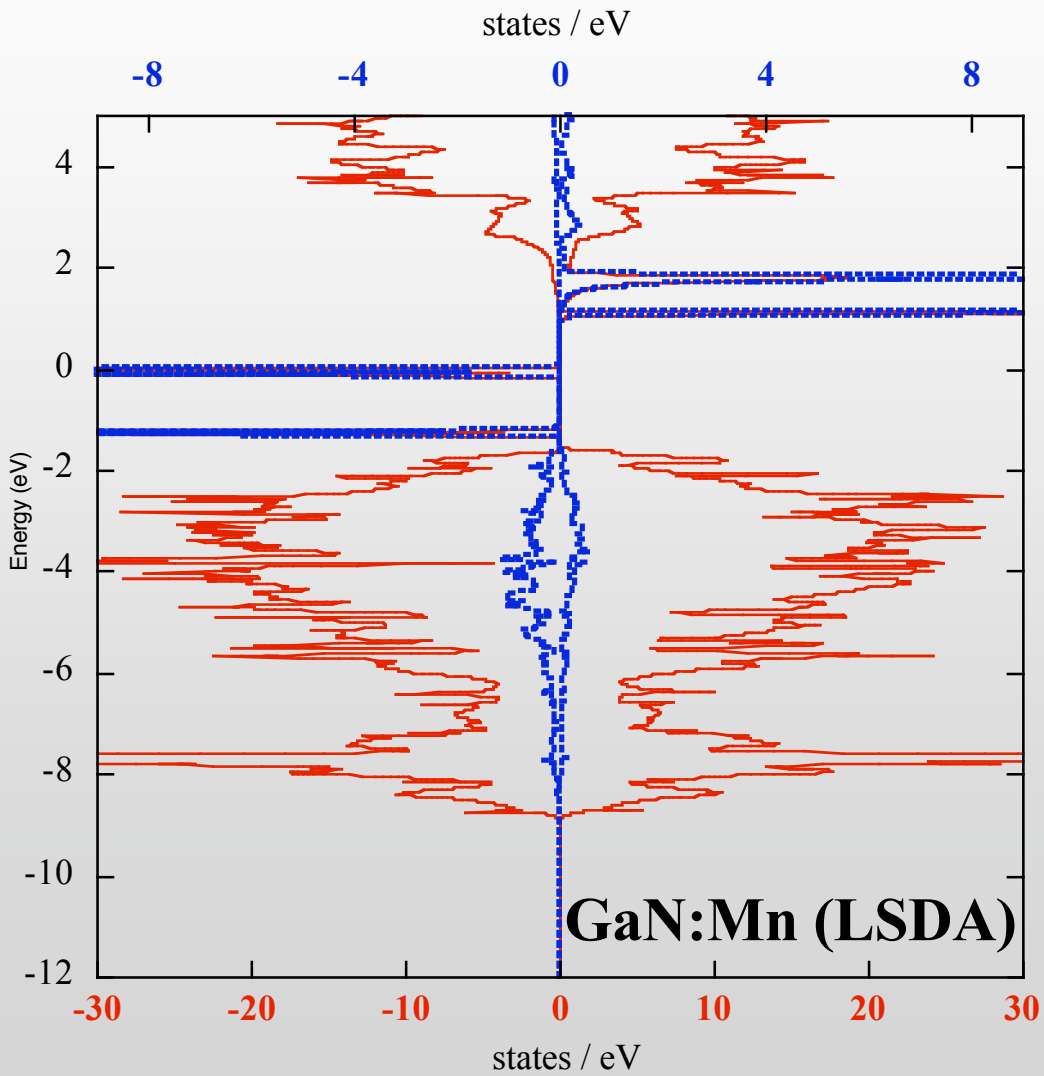
The Desired Electronic Structure

- Mn dope Ga based III-V: GaN, GaP, GaAs, GaSb
  - Mn ($3d^5 4s^2$) substitutes Ga ($3d^{10} 4s^2 4p$)
- Ideally: all Mn-*d* orbitals stay localized (atom-like)
  - (GaMn)N: Mn(d^5) + spin polarized hole
  - Zener model description of FM
- Is that really so?
  - why not d^4, no hole in valence band
  - or the hole strongly coupled (local)
  - or Mn-*d* states are not atom-like at all



Ab Initio Calculations: LSDA



- three t_{2g} bands @ E_F
  - d-band metal
- double exchange
  - solved by ground state calculation?

LSDA & Spurious Self-Interactions

ionic potential

kinetic energy

$$T_s[\rho] = \sum_i^N \langle \psi_i | -\frac{\hbar}{2m}\nabla^2 | \psi_i \rangle$$

$$E_v[\rho] = \int \rho(\xi) v(\xi) d\xi + T_s[\rho] + J[\rho] + E_{xc}[\rho]$$

Hartree term - classical coulomb interaction

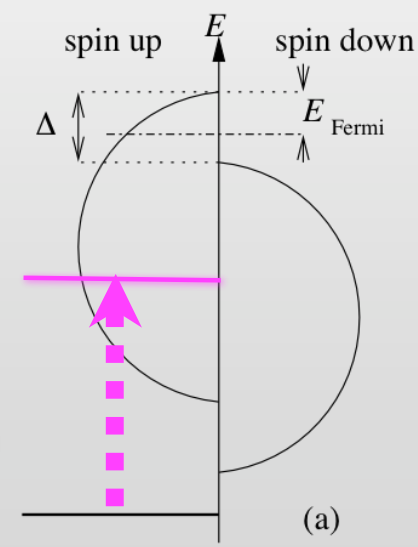
$$J[\rho] = \int \rho(\xi)\rho(\xi') w(\xi,\xi') d\xi d\xi'$$

charge density

$$\rho = \sum_i^N |\psi_i(\xi)|^2 = \sum_i^N \rho_i(\alpha, \vec{r})$$

self-interaction term cancelled by exact exchang correlation

$$J[\rho_i(\alpha)] = \int \rho_i(\alpha,\vec{r})\rho_i(\alpha,\vec{r}') w(\vec{r},\vec{r}') d\vec{r} d\vec{r}'$$



effect of self interaction: shift *d-levels* up in energy

LSDA: introduces "spurious" self-interactions

$$E_{xc}[\rho] \approx E_{xc}^{LSDA}[\rho]$$

Self-Interaction Corrected LSD

$$E^{SIC-LSD} = E^{LSDA} - \sum_i \delta_{i,\alpha}$$

(Perdew & Zunger, PRB 1981)

Self-interaction for particular orbital: $\delta_{i,\alpha} = J[\rho_i(\alpha)] + E_{xc}^{LSD}[\rho_i(\alpha)]$

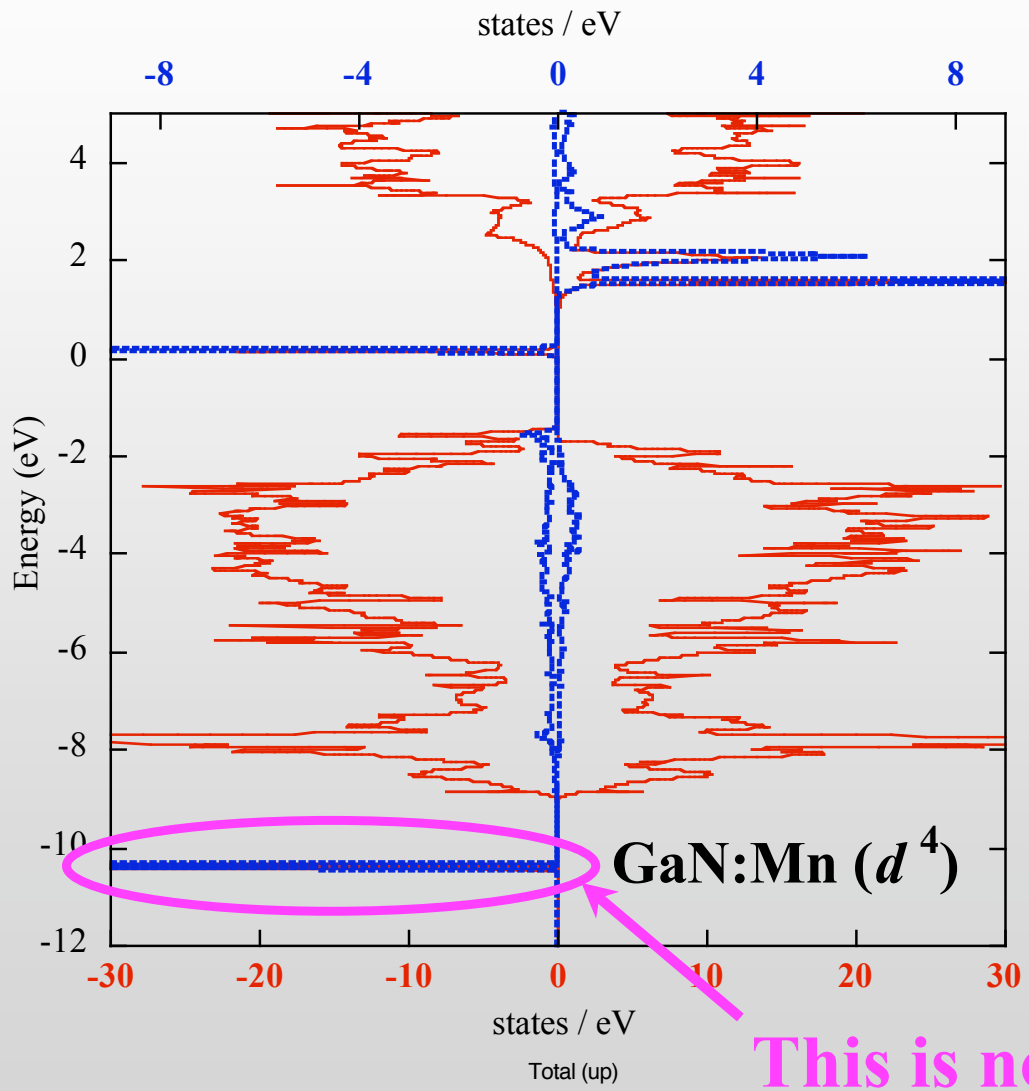
Properties:

1. self-interaction vanish for extended orbitals (LSDA is contained in SIC-LSD)

$$\delta_{i,\alpha} = 0 \Rightarrow E^{SIC-LSD} \equiv E^{LSDA}$$

2. sum rules for exchange correlation hole are satisfied
3. functional of orbitals - very hard to minimize
   - several successful implementation for atoms
   - only two correct implementations for in solids
     (Svane 1988, Temmerman et al. 1993)
   - capable of describing Mott-Heisenberg and charge-transfer insulators
     (note, all LSDA can describe is a Slater transition)

SIC-LSD Results for GaN



valence defined by number of SI-correcting states

- valence = $Z - Z^{core} - Z^{SIC}$
- LSDA = Mn^{7+} or d^0

ground state: Mn d^4

- all but one t_{2g} localize

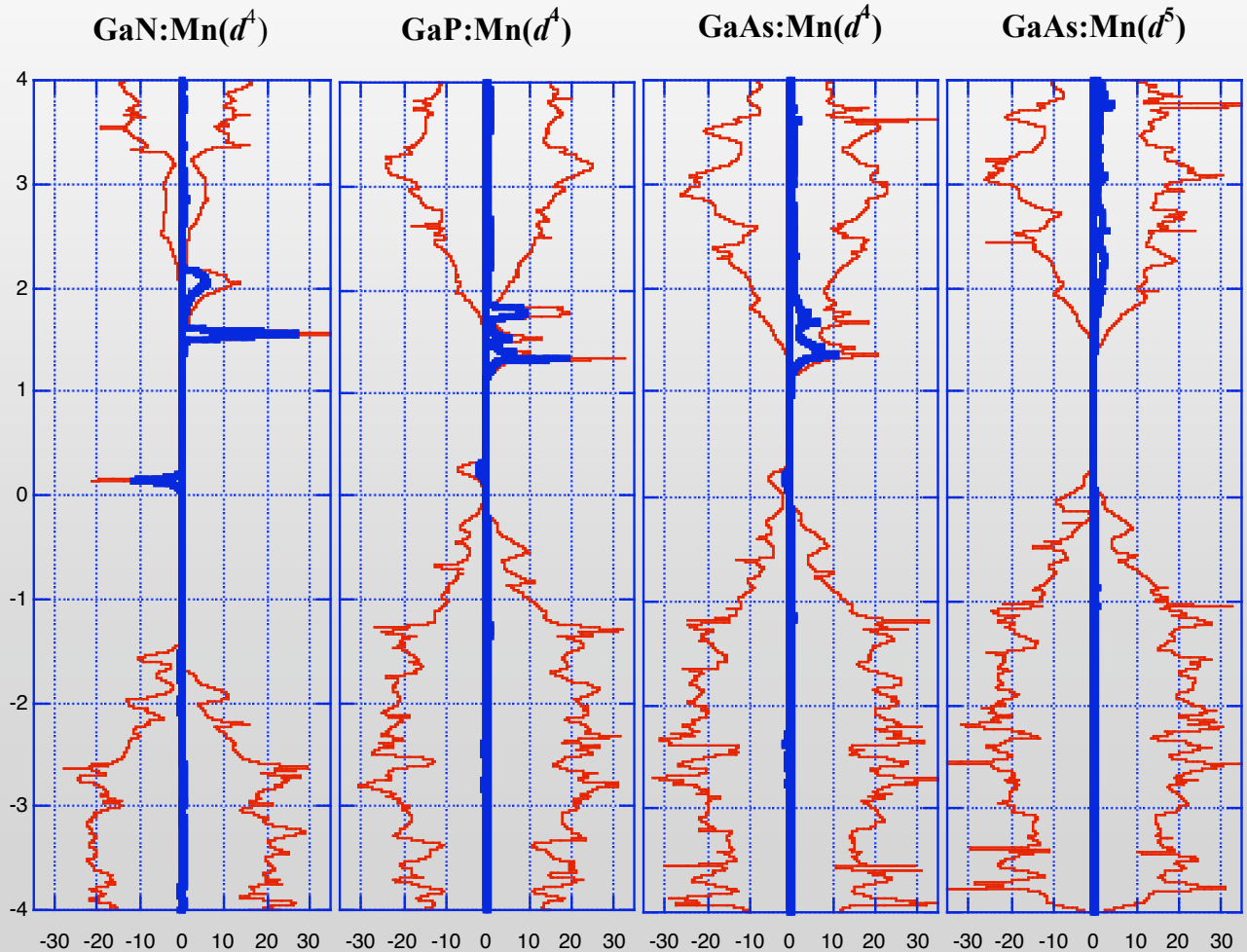
$E(d^5+h)-E(d^4) = 1.28$ eV

$E(\text{LSDA})-E(d^4) = 1.30$ eV

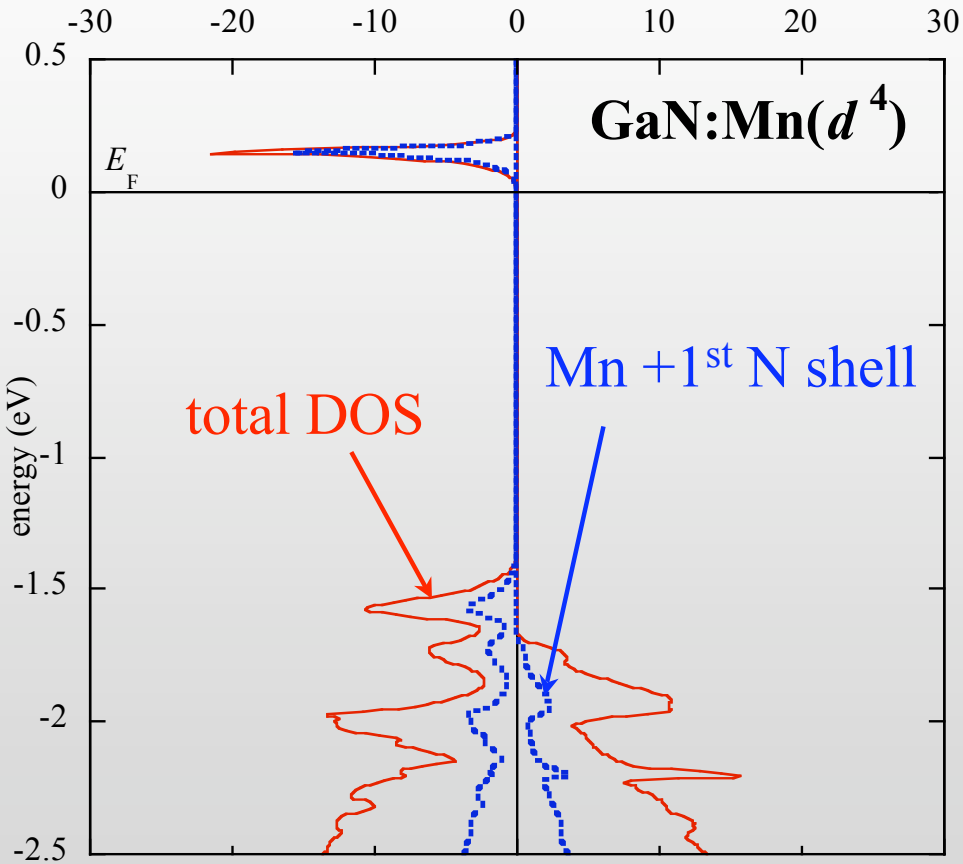
This is not where the Mn-d levels will appear in photoemission!

Mn doped GaN, GaP, and GaAs

Ground state	GaN:Mn(d^4)	GaP:Mn(d^4)	GaAs:Mn(d^5+h)
$E(d^4)$-$E(d^5+h)$	-1.28 meV	-0.04 meV	0.18 meV
Mn moment	3.61 μ_B	3.91 μ_B	4.50 μ_B



Zooming in on (GaMn)N with d^4



mid-gap impurity level:

- localized on Mn & first N neighbor shell

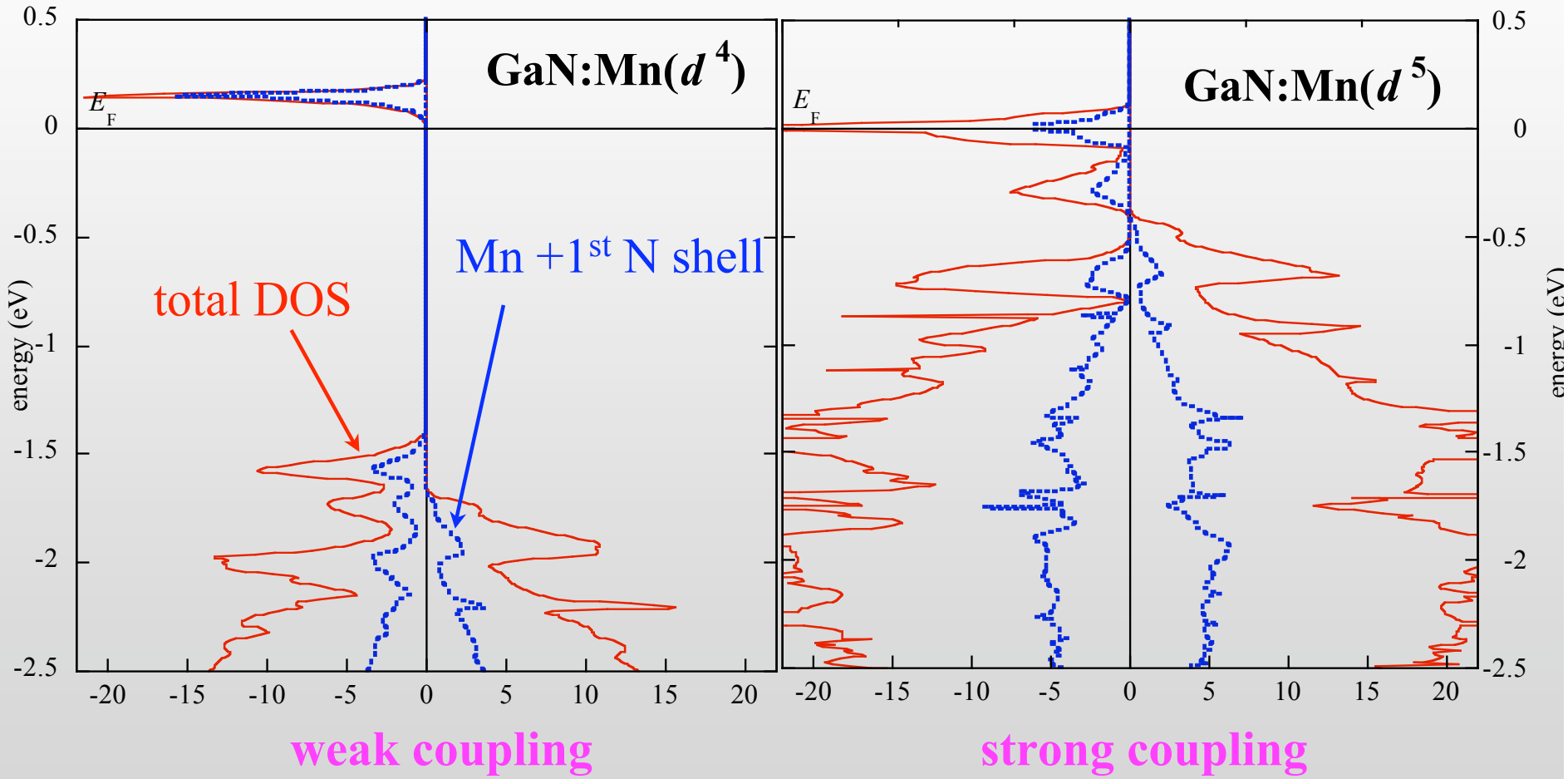
Top of valence band:

- spin polarized
- split doublet merged with valence band (weak coupling)
- localized on ~ three N neighbor shells

Hypothesis: carrier mediated ferromagnetism *p*-doped (GaMn)N

If (GaMn)N was in (d^5+h) ...

Remember: $E(d^4)$-$E(d^5+h)$ = -1.28 eV

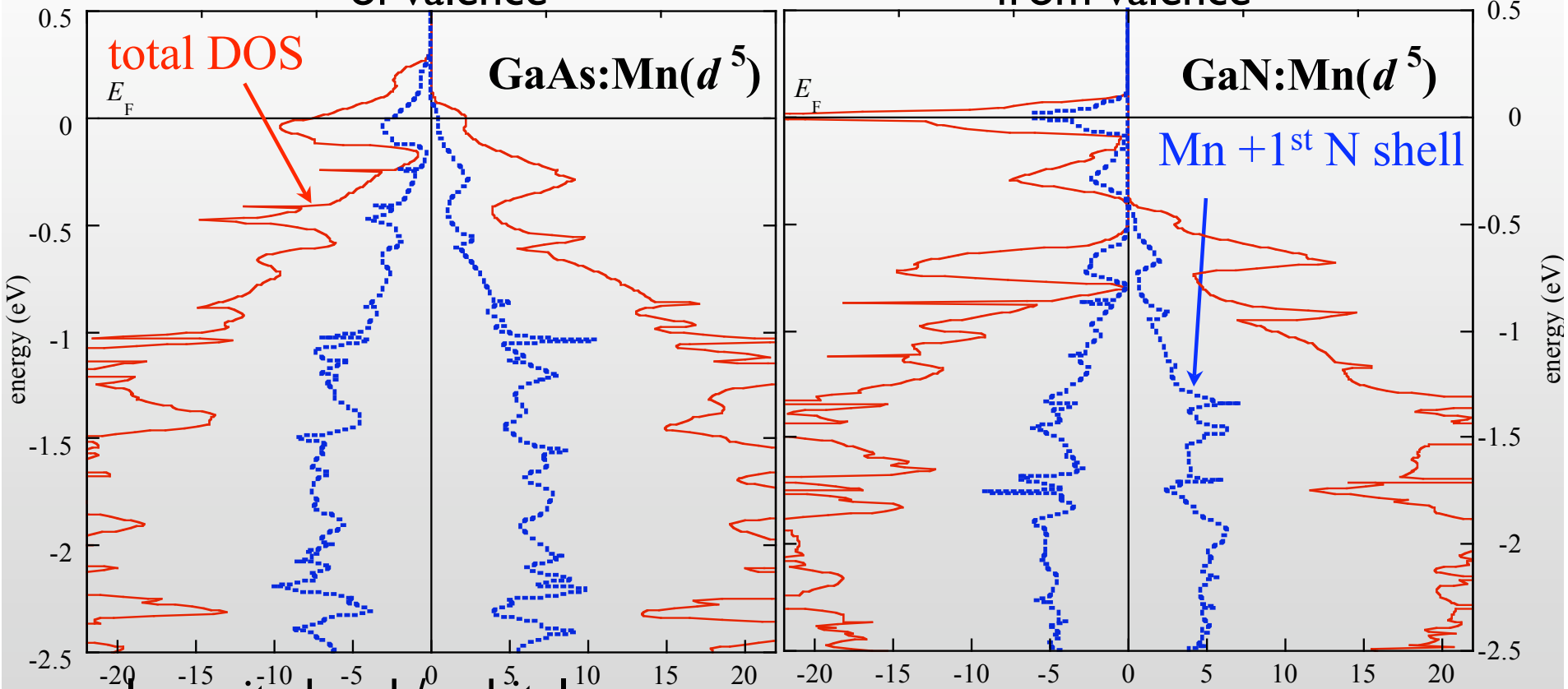


not included in SIC-LSd: possibility to form a Zhang-Rice singlet which may lower energy of (d^5+h)

Compare (d^5+h) in GaAs and GaN

Impurity band part of valence

Impurity band split from valence



Impurity band / orbital:

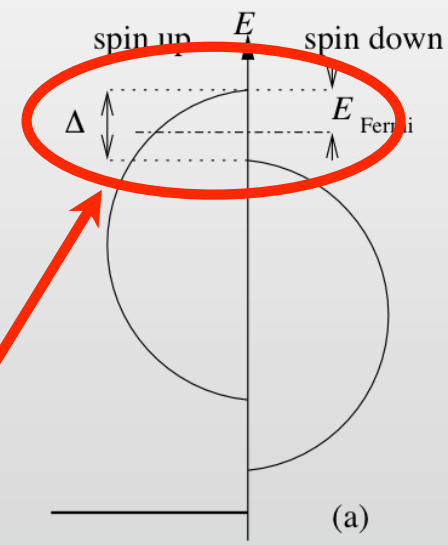
- merged with valence
- extends beyond 4 As shells

Impurity band / orbital:

- split from valence
- localized within 4 N shells

Conclusions / Suggestions

- SIC-LSD, a generalization of LSDA for systems with localized orbitals (such as Mn doped III-V)
- Mn ground state configuration is
  - d^4 in GaN and GaP
  - d^5 in GaAs
- Simple picture is too simple but not wrong
  - VBM is spin polarized
  - shallow impurity states are Mn centered
  - weak coupling for GaN:Mn(d^4) and GaAs:Mn(d^5)
  - strong coupling in hypothetical GaN:Mn(d^5)



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