Electron correlations in diluted magnetic semiconductors

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- motivation: experimental data show importance of correlation effects in DMS

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- Curie temperatures
- percolation in diluted magnetic system
- conclusions and open problems

DILUTED MAGNETIC SEMICONDUCTORS

III-V semiconductors (e.g. GaAs, GaN) doped with Mn

Mn can substitute Ga – single acceptor (adds 1 hole)

interstitial Mn – double donor (adds 2 electrons)

As-antisite – double donor (adds 2 electrons)

structure: GaAs zincblende, GaN wurtzite

ferromagnetism mostly in p-type samples

half-metals

Curie temperatures: Ga-Mn-As $\approx$ 170 K

Ga-Mn-N 0 – 940 K (controversial)

Mn in GaN, GaP, GaAs, and GaSb

Sato et al., Physica B **340-342** (2004), 863.

host	a [Å]	hybr.	Mn level	Δ[eV]^a	exch.	range
GaN (2p)	4.49	strong	in gap	0.10	double	short
GaP (3p)	5.45	interm.	in VB	0.30	p-d	long
GaAs (4p)	5.65	interm.	in VB	0.39	p-d	long
GaSb (5p)	6.10	weak	in VB		p-d	long

^{a} Mahadevan and Zunger, PRB **69** (2004), 115211.

PHOTOEMISSION SPECTRA

Mn-3d peak in different position than predicted by LSDA

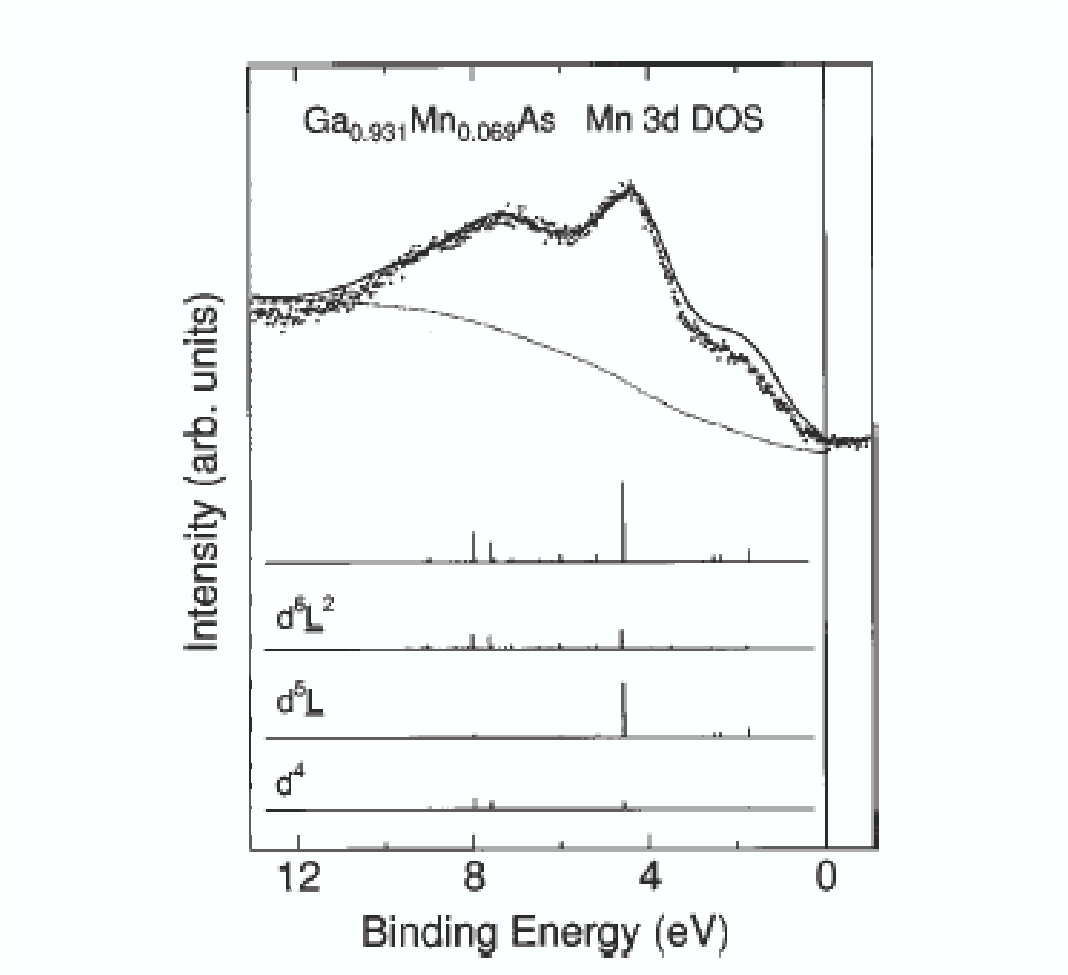
at approx. 4 eV below E_{F}

J. Okabayashi et al., PRB **59**, R2486 (1999)

C. Carbone (private communication)

H. Åsklund et al., PRB **66**, 115319 (2002)

PHOTOEMISSION SPECTRA



J. Okabayashi et al., PRB **59**, R2486 (1999), Fig. 3

ELECTRONIC STRUCTURE

FP-LAPW applied to supercells

TB-LMTO-CPA applied to random alloys

both methods yield basically the same electronic structure within the LSDA

electron correlations included approximately within the LSDA+U method

Park et al., Physica B **281-282**, 703 (2000)

in most of calculations $U = 4$ eV, $J = 0.8$ eV

again, both methods yield basically the same electronic structure within the LSDA+U

LDA+U

$$
\begin{aligned}
E_{tot} &= E_{LSDA} + E_{ee} - E_{dc} \\
|\gamma\rangle &= |lm\sigma\rangle \ldots \text{spinorbital} \\
v_{\gamma_1,\gamma_2,\gamma_3,\gamma_4} &= \langle\gamma_1,\gamma_3|V^{ee}|\gamma_2,\gamma_4\rangle - \langle\gamma_1,\gamma_3|V^{ee}|\gamma_4,\gamma_2\rangle
\end{aligned}
$$

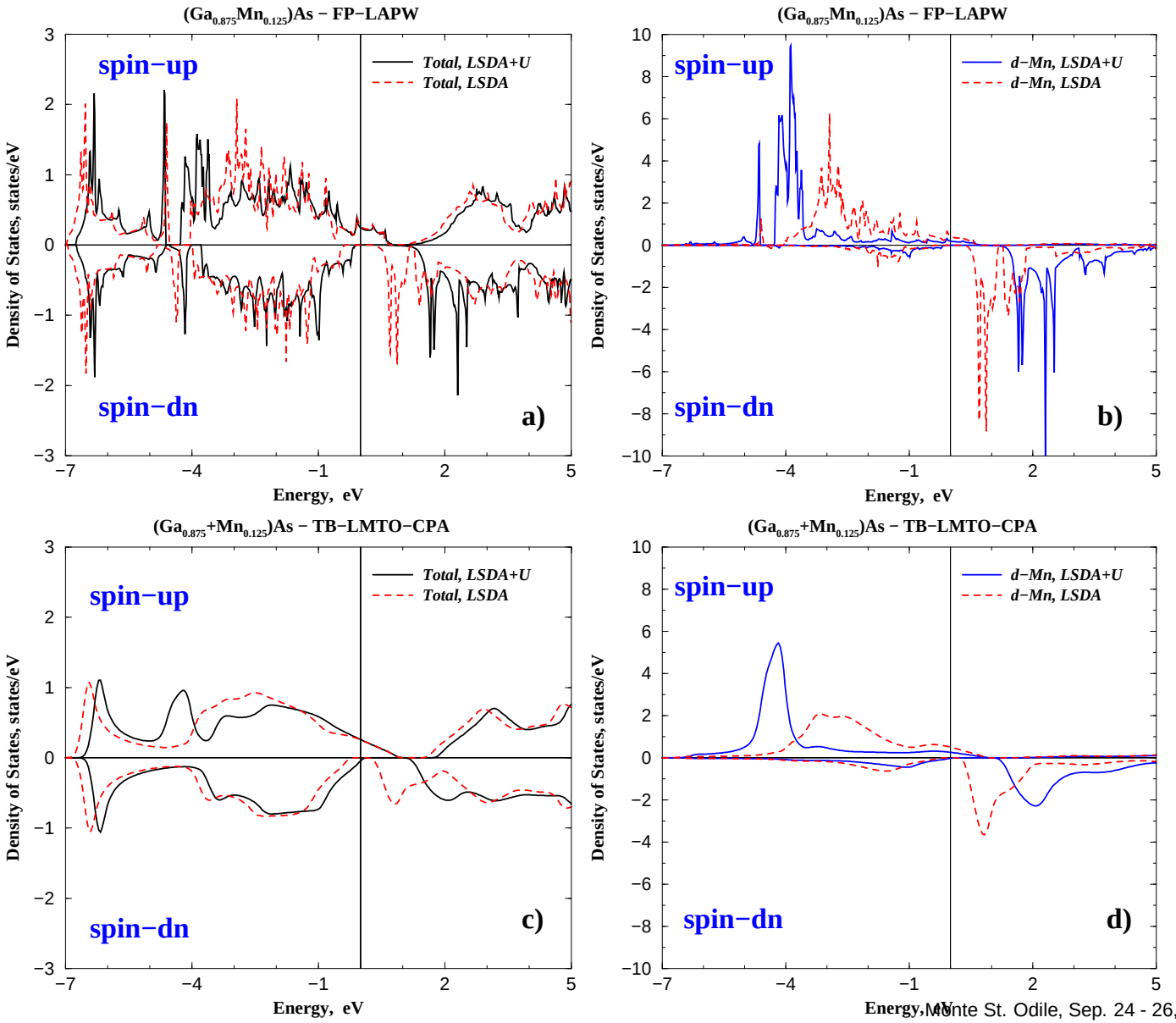
LDA+U: fully-localized limit (FLL)

$$E_{ee}^{\mathrm{FLL}} = \frac{1}{2} \sum_{\gamma_1,\gamma_2,\gamma_3,\gamma_4} v_{\gamma_1,\gamma_2,\gamma_3,\gamma_4}\, n_{\gamma_1,\gamma_2} n_{\gamma_3,\gamma_4}$$

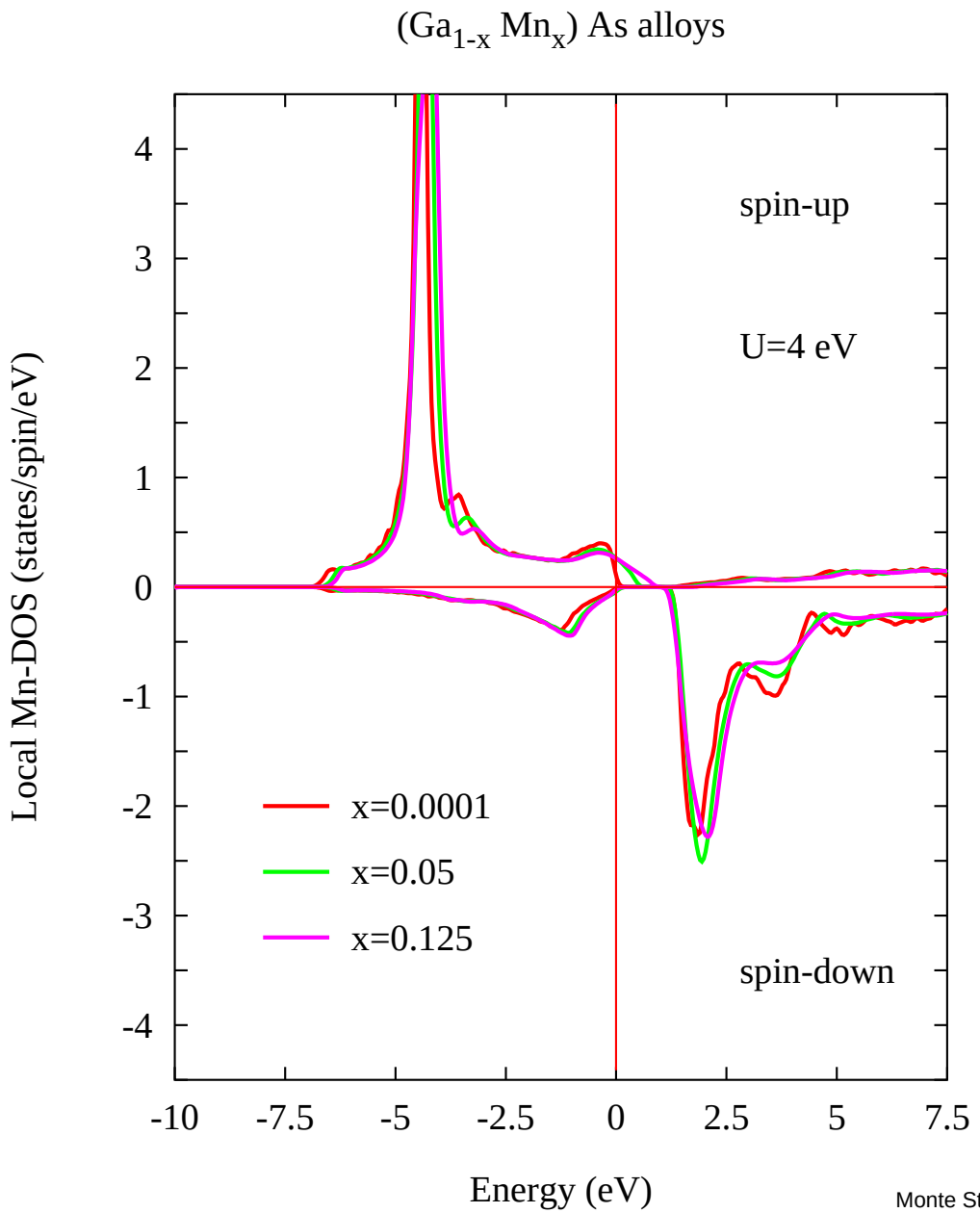
$$E_{dc}^{\mathrm{FLL}} = \frac{U}{2} n(n-1) - \frac{J}{2} \sum_{\sigma} n^{\sigma}(n^{\sigma}-1)$$

$$n^{\sigma} = \frac{1}{2l+1} \sum_{m=-l}^{l} n_{m\sigma,m\sigma}\,, \qquad n = \sum_{\sigma} n^{\sigma}$$

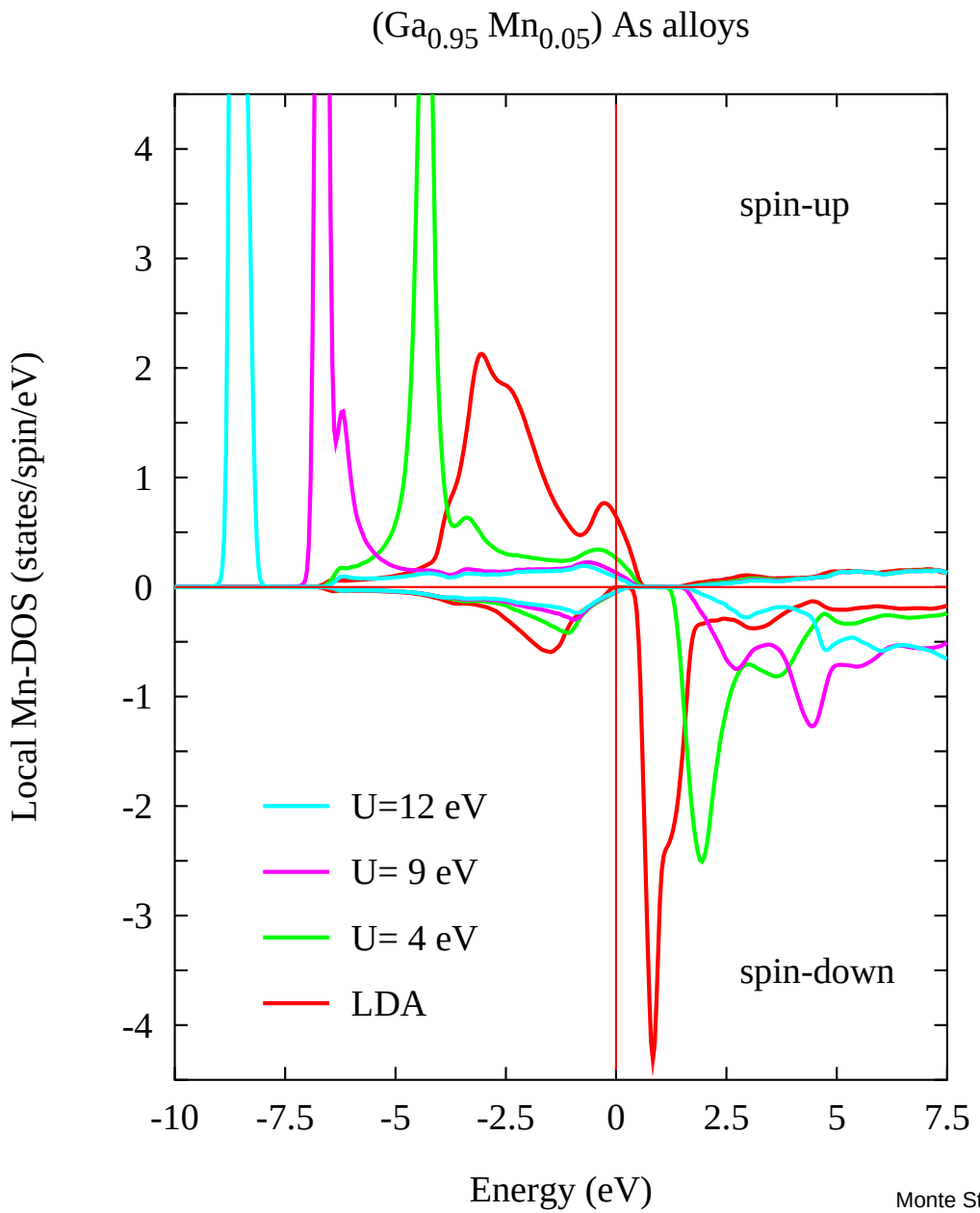
FP-LAPW and TB-LMTO-CPA DOS



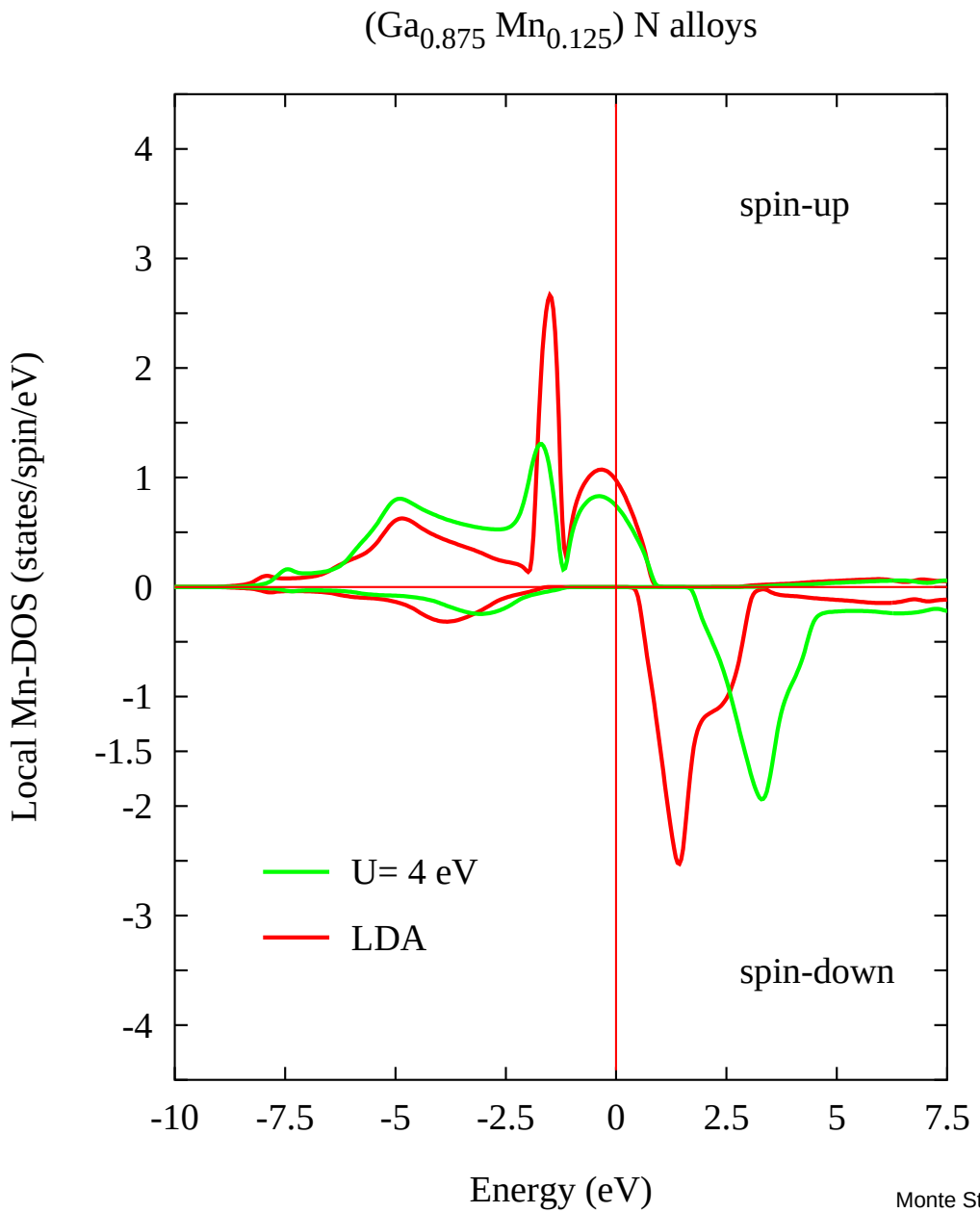
TB-LMTO-CPA d-Mn DOS: varying Mn concentration



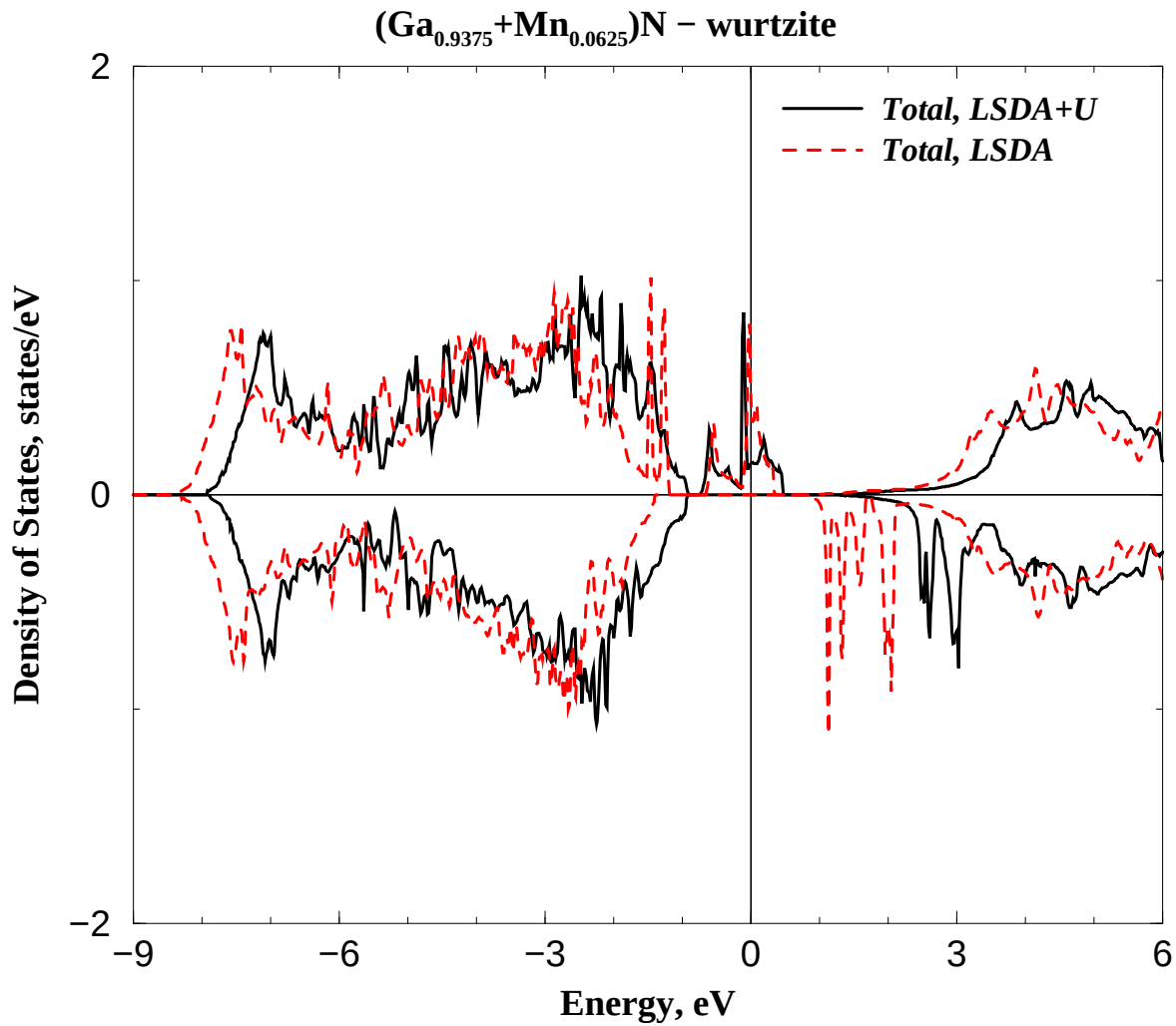
TB-LMTO-CPA d-Mn DOS: varying U



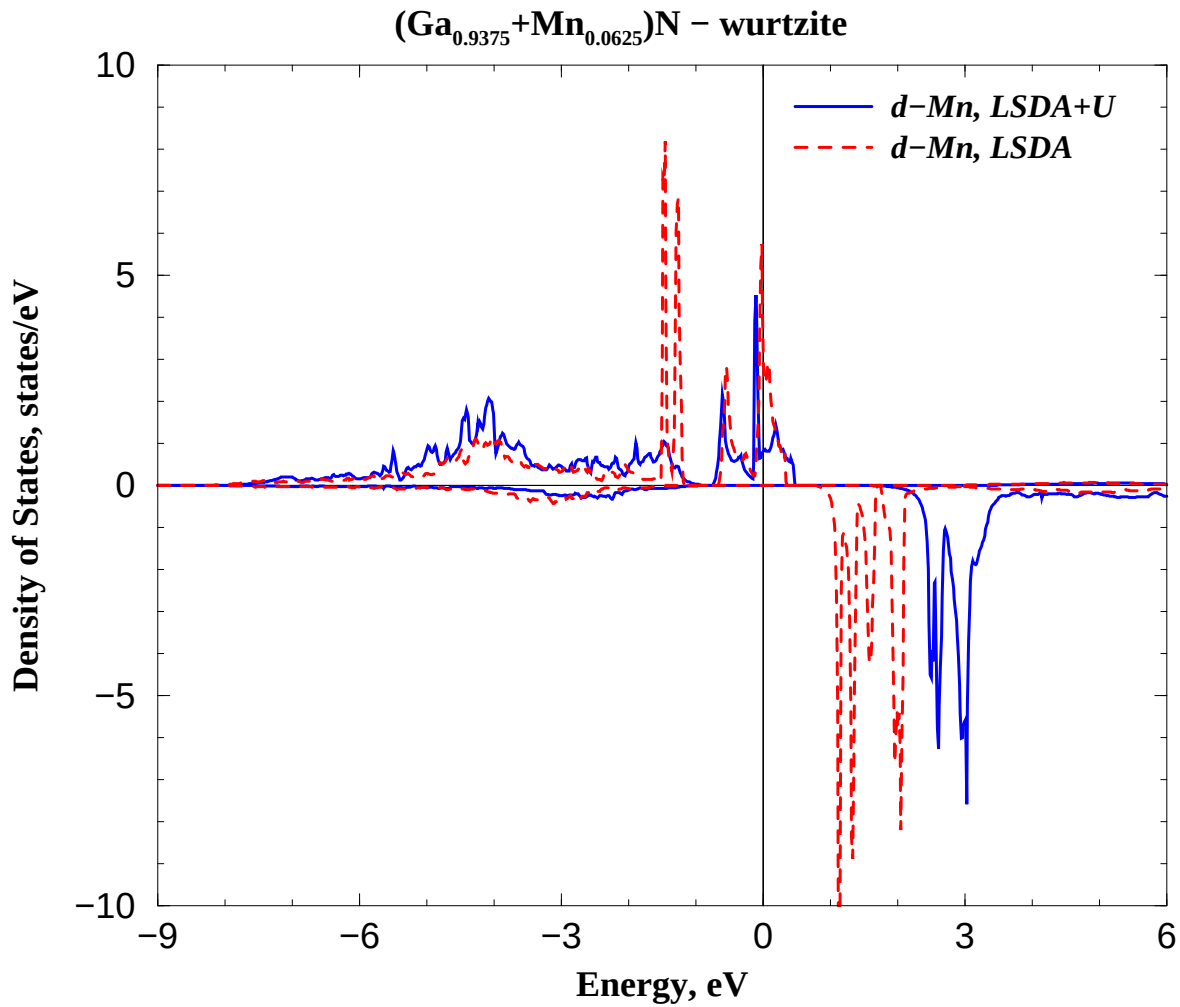
TB-LMTO-CPA d-Mn DOS: GaN



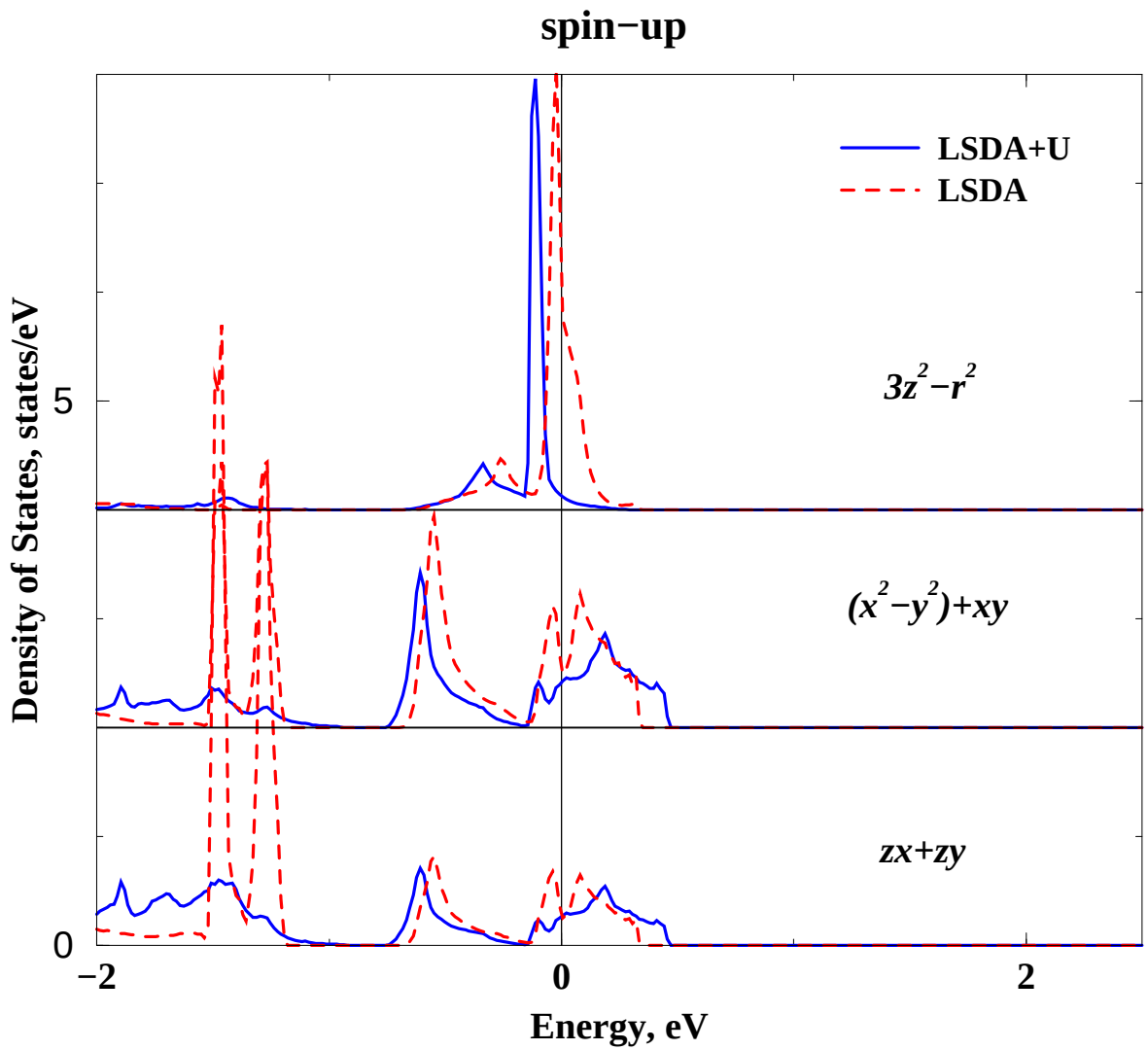
FP-LAPW total DOS: GaN wurtzite



FP-LAPW 3d-Mn DOS: GaN wurtzite



orbital-resolved 3d-Mn DOS



electronic structure: summary

Mn is a shallow acceptor in GaAs

Mn is a deep acceptor in GaN (due to strong hybridization of d-Mn and p-N states)

GaMnAs: d-Mn peak is shifted to its experimentally observed position

GaMnN: d-Mn (e_g) peak on the top of the valence band is moved inside the valence band, while the peak in the gap (t_{2g}) remains almost unchanged

classical Heisenberg Hamiltonian

$$H_{\mathrm{Heisenberg}} = -\sum_{i,j} J_{ij}\, \mathbf{e}_i \cdot \mathbf{e}_j$$

$\mathbf{e}_i$ = unit vector parallel to magnetic moment at site i

J_{ij} = exchange interaction

$J > 0$ ... FM

$J < 0$ ... AFM

exchange interactions

method of infinitesimal rotations

A.I. Liechtenstein et al., JMMM **67**, 65 (1987), one-particle theory

M.I. Katsnelson and A.I. Liechtenstein, PRB **61**, 8906 (2000), interacting systems

$$J_{ij} = \frac{1}{4\pi}\mathrm{Im}\int_{-\infty}^{E_\mathrm{F}} \mathrm{d}E\,\mathrm{tr}_L\left[\delta_i(E)g_{ij}^{\uparrow}(E+)\delta_j(E)g_{ji}^{\downarrow}(E+)\right]$$

$\delta_i(E) = P_i^{\uparrow}(E) - P_i^{\downarrow}(E)$ ... exchange splitting

$g_{ij}(z)$ = auxiliary GF

valid for robust magnetic moments

describes well transversal excitations (spin waves)

longitudinal (Stoner) excitations neglected

applications to DMS

J. Kudrnovský et al., PRB **69** 115208 (2004) LDA only

A.B. Shick et al., PRB **69** 125207 (2004) LDA+U

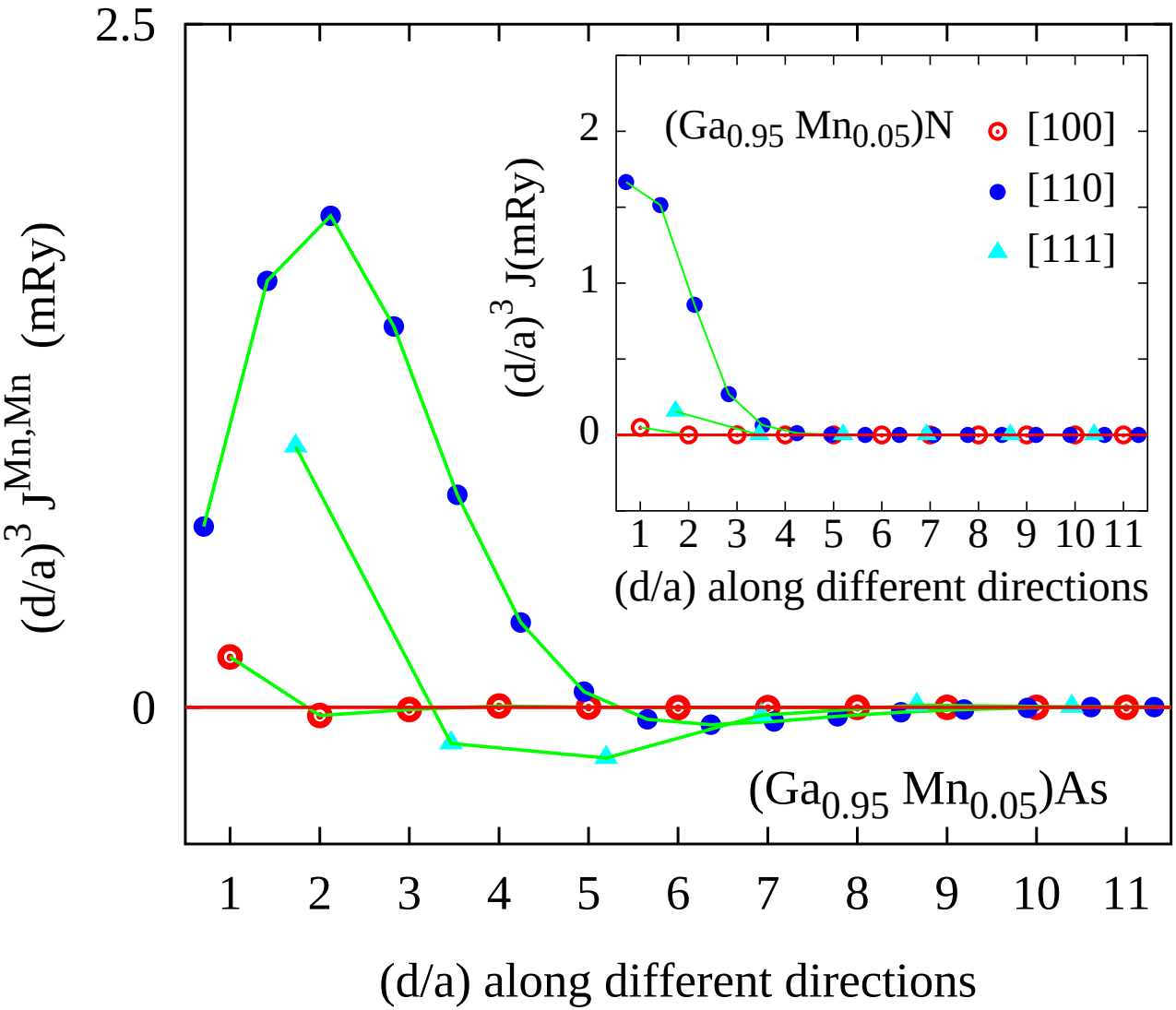
I. Turek et al., Psi-k Highlight of the month, Sep. 2003 review

Sato et al., Europhys. Lett. **61** 403 (2003), KKR

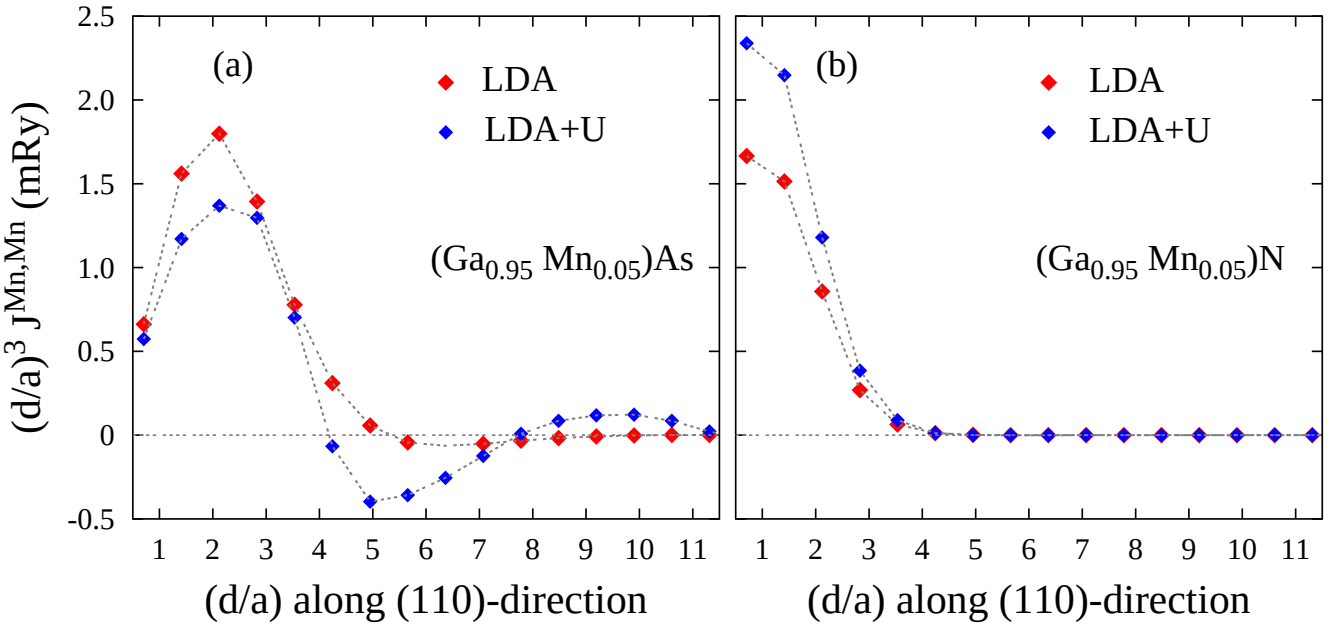
Sandratskii and Bruno, PRB **66** 134435 (2002), frozen magnons

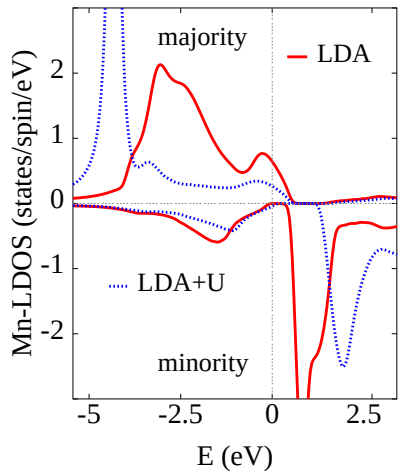
Sandratskii et al., PRB **69** 195203 (2004), LDA+U, frozen magnons

anisotropy of exchange interactions



exchange interactions: LSDA vs. LSDA+U





Curie temperature

- mean-field approximation (MFA)

$$k_{\mathrm{B}} T_{\mathrm{C}}^{\mathrm{MFA}} = \frac{2x}{3} \sum_{i \neq 0} J_{0i}$$

- random-phase approximation (RPA)

$$\left(k_{\mathrm{B}} T_{\mathrm{C}}^{\mathrm{RPA}} \right)^{-1} = \frac{3}{2x} \, \frac{1}{N} \sum_{\mathbf{q}} \left[J(\mathbf{0}) - J(\mathbf{q}) \right]^{-1} ,$$

$$\text{where} \quad J(\mathbf{q}) = \sum_{i (i \neq 0)} J_{0i} \cdot \mathrm{e}^{i \mathbf{q} \cdot \mathbf{R}_{0i}}$$

- Monte Carlo simulations yield very accurate value of T_{C}

diluted random magnetic systems

two problems:

(1) random distribution of impurities on the lattice

(2) large fluctuations of local fields

- mean-field approximation: fails
- RPA in real space: works rather well

  G. Bouzerar et al., cond-mat 0405322

- Monte Carlo simulations yield very accurate value of T_{C}

  L. Bergqvist, O. Eriksson, J. Kudrnovský, V. Drchal, P. Korzhavyi, and I. Turek, PRL **93** (2004), 137202

  numerically highly demanding – up to 16 neighbors

PERCOLATION

nearest-neighbor interactions: $x_{\rm crit}(fcc) \approx 0.19$

asymptotic behavior of J_{ij}

$$J_{\mathbf{d}} \propto \ \mathrm{e}^{-\,\boldsymbol{\lambda}^{\uparrow}\cdot\mathbf{d}}\,.\,\mathrm{e}^{-\,\boldsymbol{\kappa}_{\rm F}^{\downarrow}\cdot\mathbf{d}}\ \frac{\sin(\mathbf{k}_{\rm F}^{\uparrow}\cdot\mathbf{d}+\Phi^{\uparrow}+\Phi^{\downarrow})}{|\mathbf{d}|^{3}}$$

J_{ij} highly anisotropic

J_{ij} oscillatory

very delicate problem: no analytic or exact solution

brute force: L. Bergqvist Monte Carlo simulations

numerical experiment

isotropic interactions: $J_{ij} = \frac{1}{R_{ij}^3}$

interactions always positive: $J_{ij} > 0$

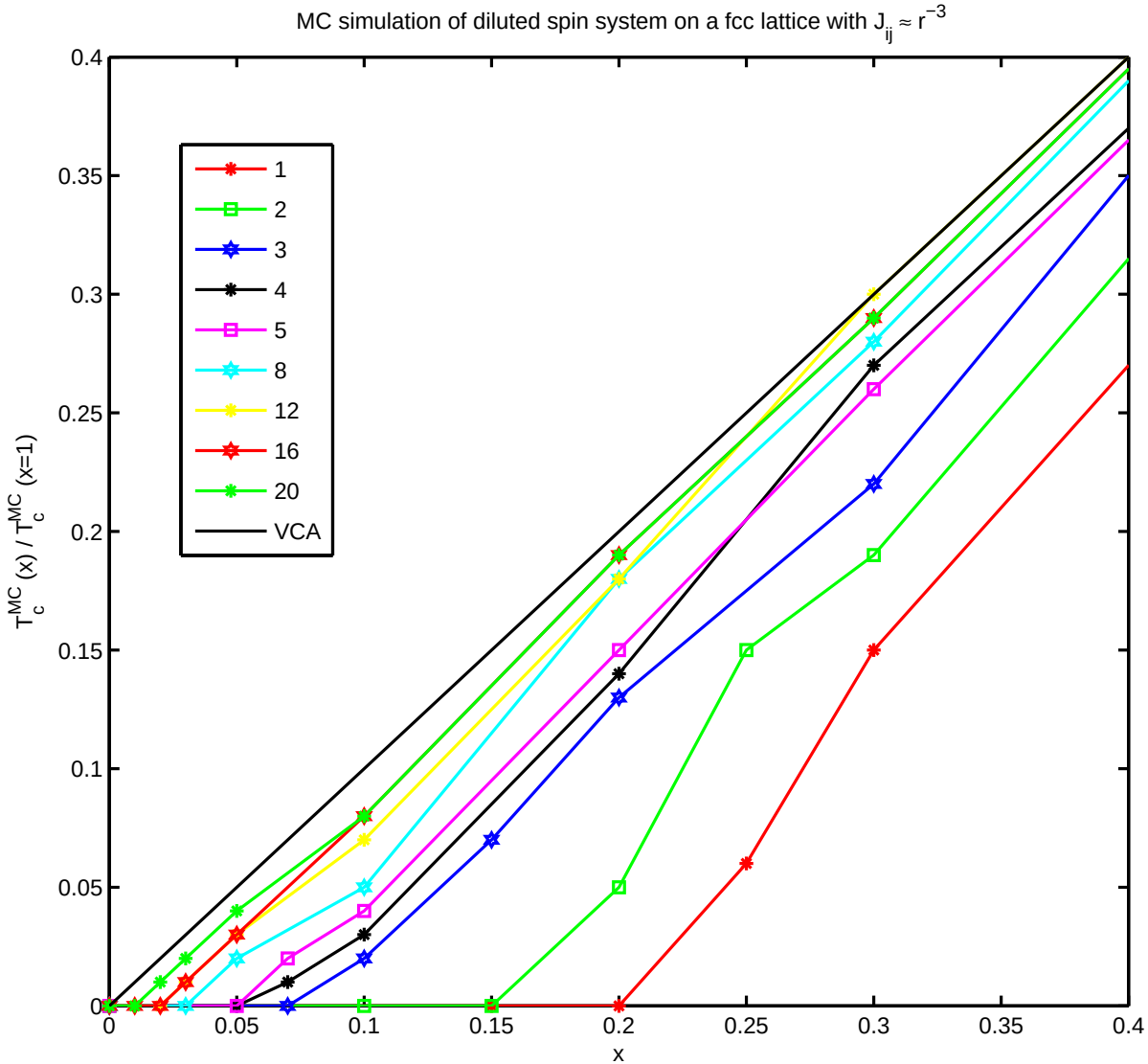
fcc lattice randomly occupied

occupied fraction of lattice points x

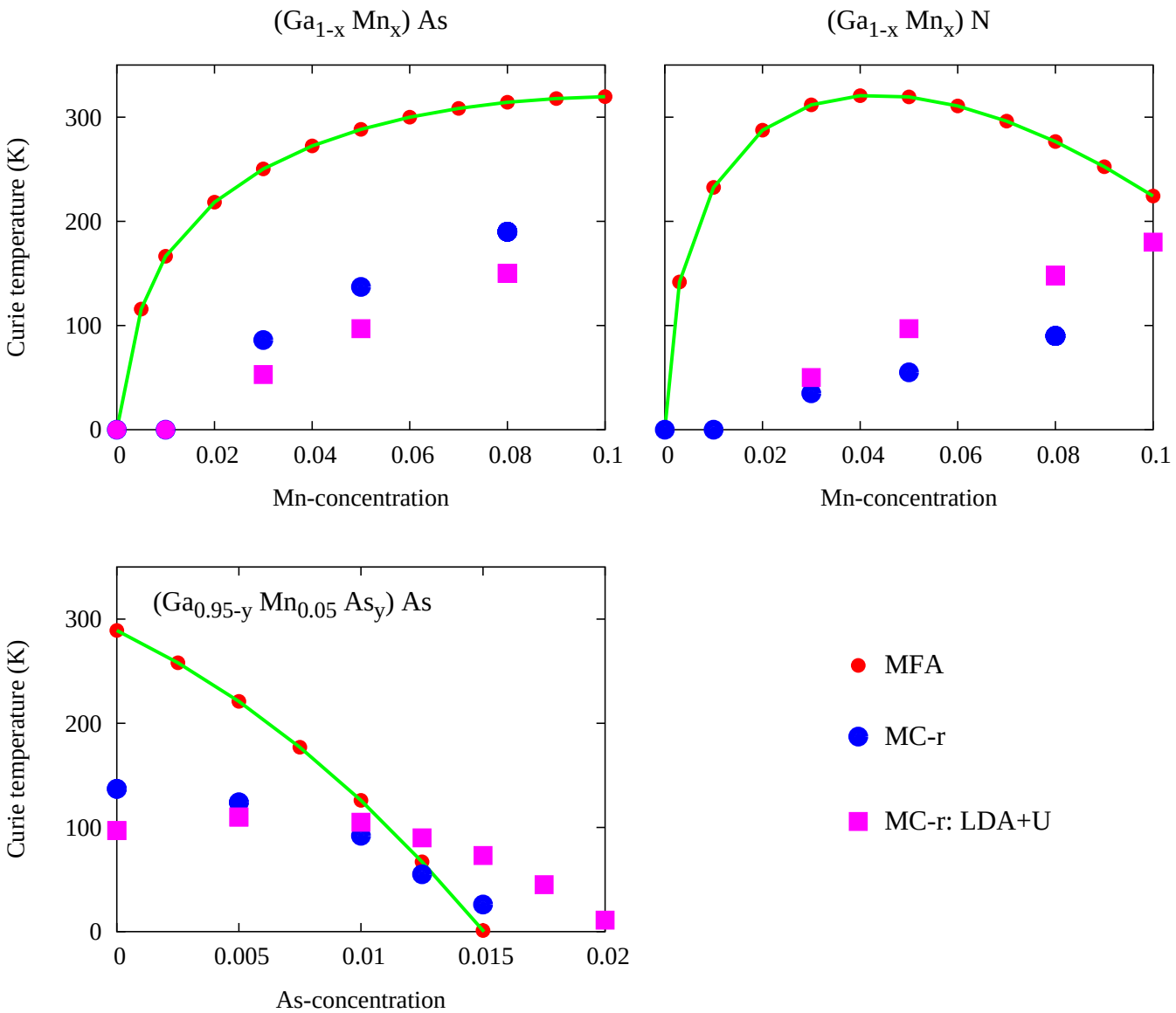
first n neighbors included

reduced Curie temperature $\frac{T_{\mathrm{C}}(x)}{T_{\mathrm{C}}(1)}$

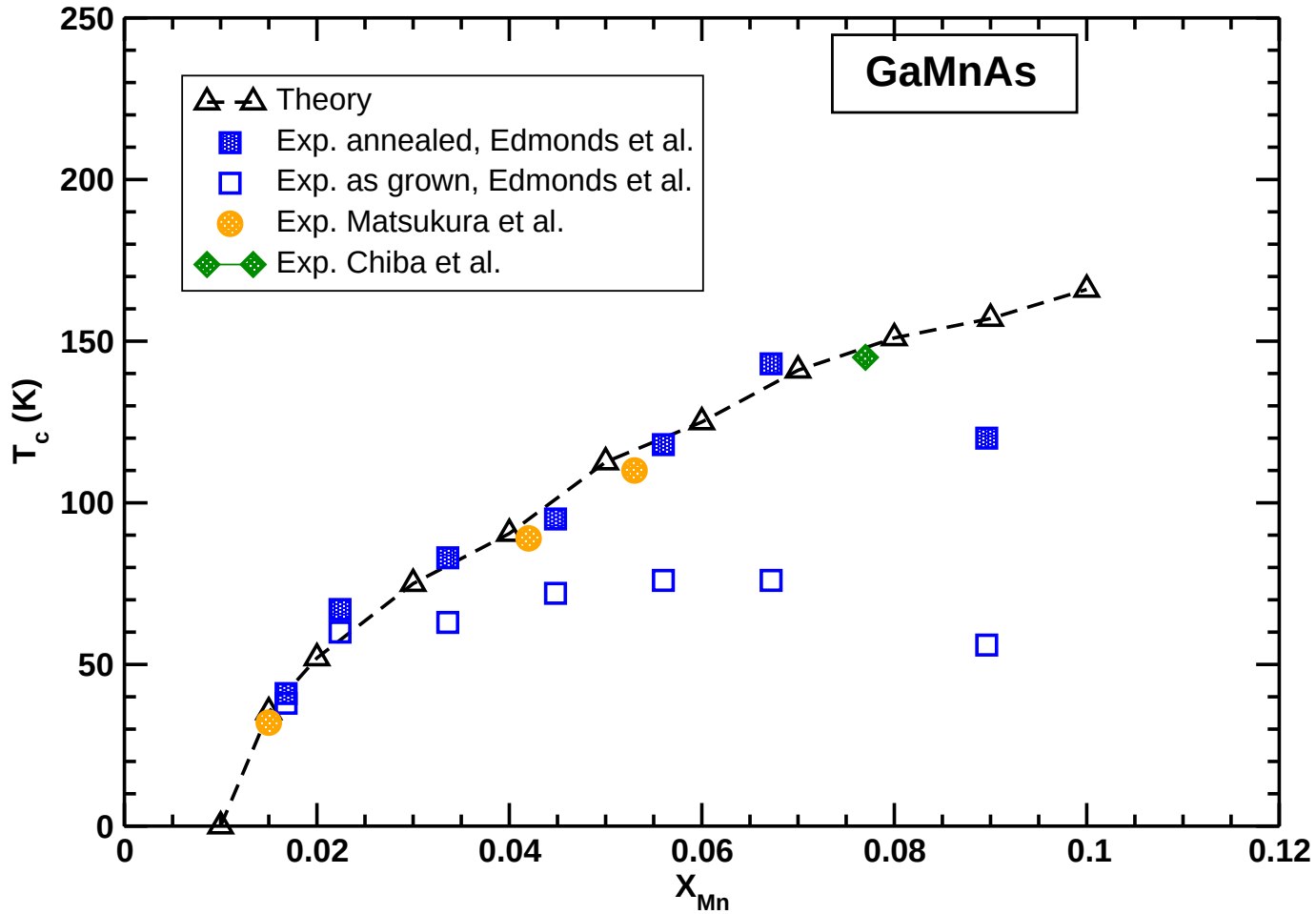
magnetic percolation



Curie temperatures

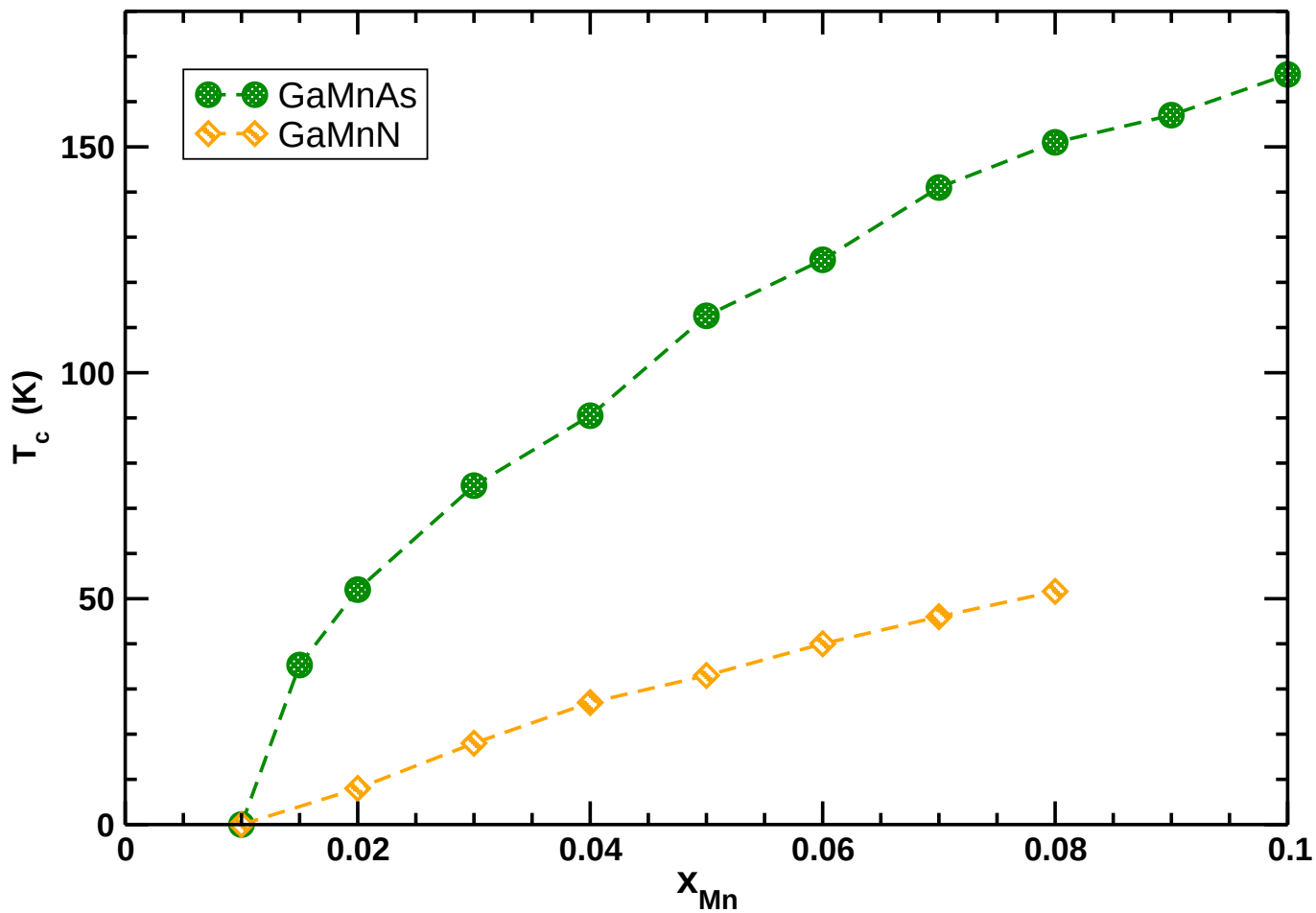


Curie temperature in GaMnAs



G. Bouzerar et al., cond-mat 0405322, Fig. 1

Curie temperature: GaMnAs vs. GaMnN



G. Bouzerar et al., cond-mat 0405322, Fig. 4

CONCLUSIONS

- LSDA+U gives photoemission spectra of DMS in agreement with experiment
- electron correlations modify exchange interactions in DMS
- percolation effects highly important
- Monte Carlo and real-space RPA yield Curie temperature in accordance with experiment