

Electronic, magnetic, and transport properties of halfmetallic semi-Heusler (Cu,Ni)MnSb alloys

Václav Drchal and Josef Kudrnovský

Institute of Physics ASCR, Praha, Czech Republic



Grant Agency of ASCR: project IAA100100616

in collaboration with

Ilja Turek and Peter Weinberger

MOTIVATION

- interesting physics of $\text{Cu}_x\text{Ni}_{1-x}\text{MnSb}$ alloys
 - NiMnSb ferromagnet
 - CuMnSb antiferromagnet
 - $\text{Cu}_x\text{Ni}_{1-x}\text{MnSb}$ crossover from FM to AFM at $x \approx 0.7$
 - Curie temperature is a smooth function of x around $x \approx 0.7$
 - magnetization and resistivity change dramatically at $x \approx 0.7$
- potential materials for spintronics
 - halfmetals
 - match lattice constants of semiconductors
 - Curie temperature is above the room temperature

OUTLINE

- structure and experimental data
- models of magnetic (dis)order
- electronic structure: TB-LMTO method
- effective Heisenberg Hamiltonian
- magnetic properties
- residual resistivity
- conclusions and outlook

C_{1b} STRUCTURE

fcc lattice

$$\mathbf{A}_1 = \frac{a}{2}(0, 1, 1)$$

$$\mathbf{A}_2 = \frac{a}{2}(1, 0, 1)$$

$$\mathbf{A}_3 = \frac{a}{2}(1, 1, 0)$$

basis

$$\boldsymbol{\tau}_1 = \frac{a}{2}(0, 0, 0) \dots \text{Cu, Ni}$$

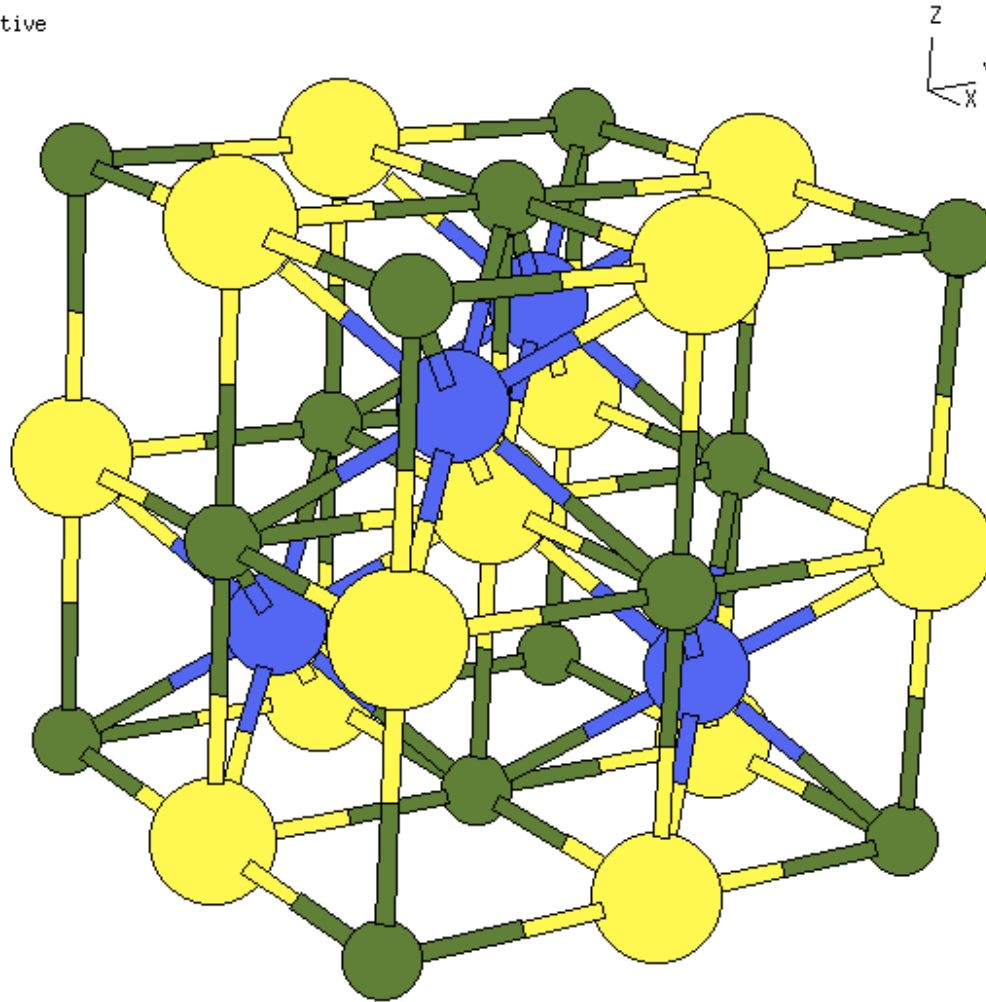
$$\boldsymbol{\tau}_2 = \frac{a}{2}\left(\frac{1}{4}, \frac{1}{4}, \frac{1}{4}\right) \dots \text{Mn}$$

$$\boldsymbol{\tau}_3 = \frac{a}{2}\left(\frac{1}{2}, \frac{1}{2}, \frac{1}{2}\right) \dots \text{empty}$$

$$\boldsymbol{\tau}_4 = \frac{a}{2}\left(\frac{3}{4}, \frac{3}{4}, \frac{3}{4}\right) \dots \text{Sb}$$

STRUCTURE

Active



EXPERIMENTAL DATA AND AB INITIO CALCULATIONS

CuMnSb

R.H. Foster et al. J. Phys. Chem. Solids **29** (1968), 855. (neutron diffraction, atomic and magnetic structure: AFM(111))

J. Boeuf et al. Phys. Rev. B **74** (2006) 024428. (magnetization, resistivity, specific heat)

T. Jeong et al. Phys. Rev. B **71** (2005) 184103. (ab initio)

(Cu_xNi_{1-x})MnSb

S.K. Ren et al. J. Alloys & Compounds **387** (2005) 32; S.K. Ren et al. J. Mag. Magn. Mater. **288** (2005) 267. (magnetization, Curie temperature, resistivity, susceptibility)

I. Galanakis et al. Phys. Rev. B **77** (2008) 214417. (ab initio)

J. Kudrnovský et al. Phys. Rev. B **78** (2008) 054441. (ab initio)

MODELS OF MAGNETIC (DIS)ORDER

$0 \leq x_{Cu} \leq 0.7$ FM state

MODEL I

$0.7 \leq x_{Cu} < 1.0$ uncompensated DLM state on the Mn sublattice

for $x_{Cu} = 0.7$: $x(Mn^\uparrow) = 1, x(Mn^\downarrow) = 0$

for $x_{Cu} = 1.0$: $x(Mn^\uparrow) = x(Mn^\downarrow) = 0.5$

linear interpolation inbetween

MODEL II

for $x_{Cu} = 1.0$: AFM(100) or AFM(111) state on the Mn sublattice

for $x_{Cu} = 0.7$: FM(100) state on the Mn sublattice

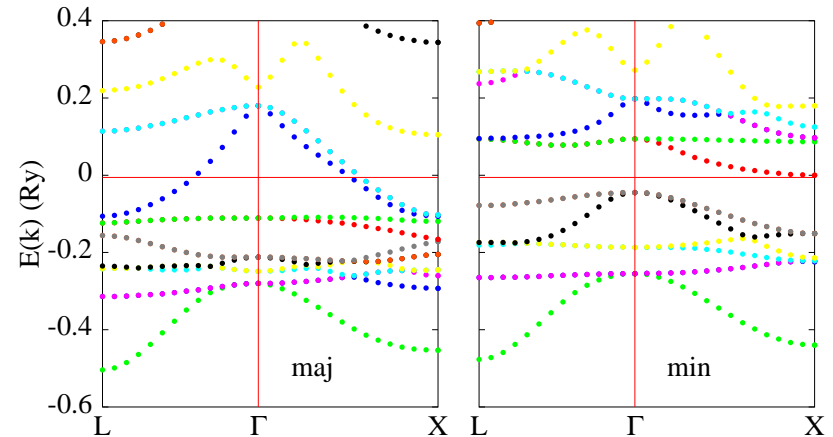
for $0.7 < x_{Cu} < 1.0$ fraction r of moments on one of the AFM lattices is reversed, fraction r is a linear function of x_{Cu}

ELECTRONIC STRUCTURE

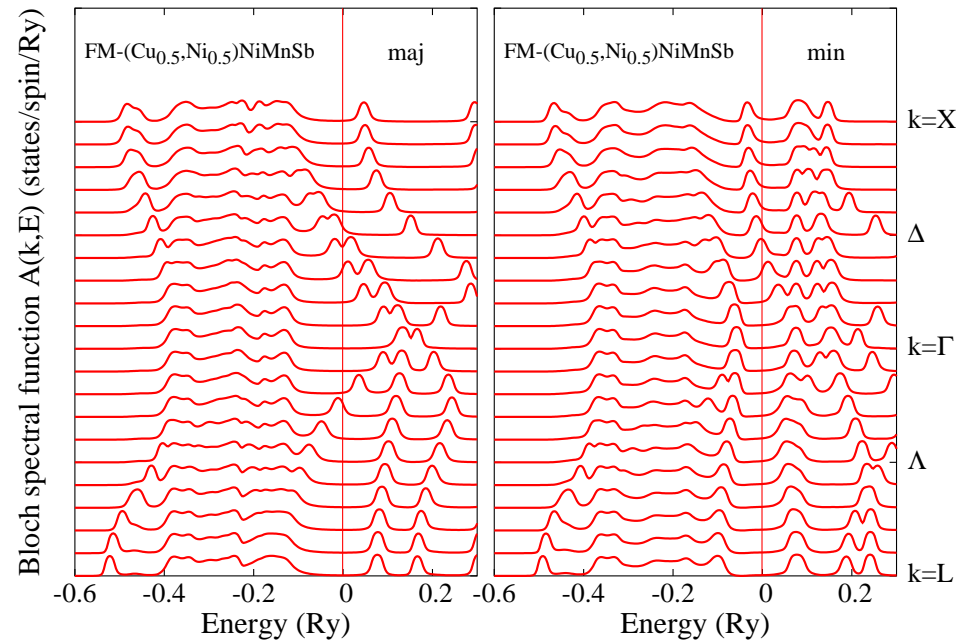
- TB-LMTO-CPA
- LSDA Vosko-Wilk-Nusair
- different magnetic structures
 - ferromagnet
 - disordered local moment (DLM)
 - AFM (100)
 - AFM (111)

NiMnSb and $\text{Cu}_{0.5}\text{Ni}_{0.5}\text{MnSb}$

bandstructure of ferromagnetic NiMnSb



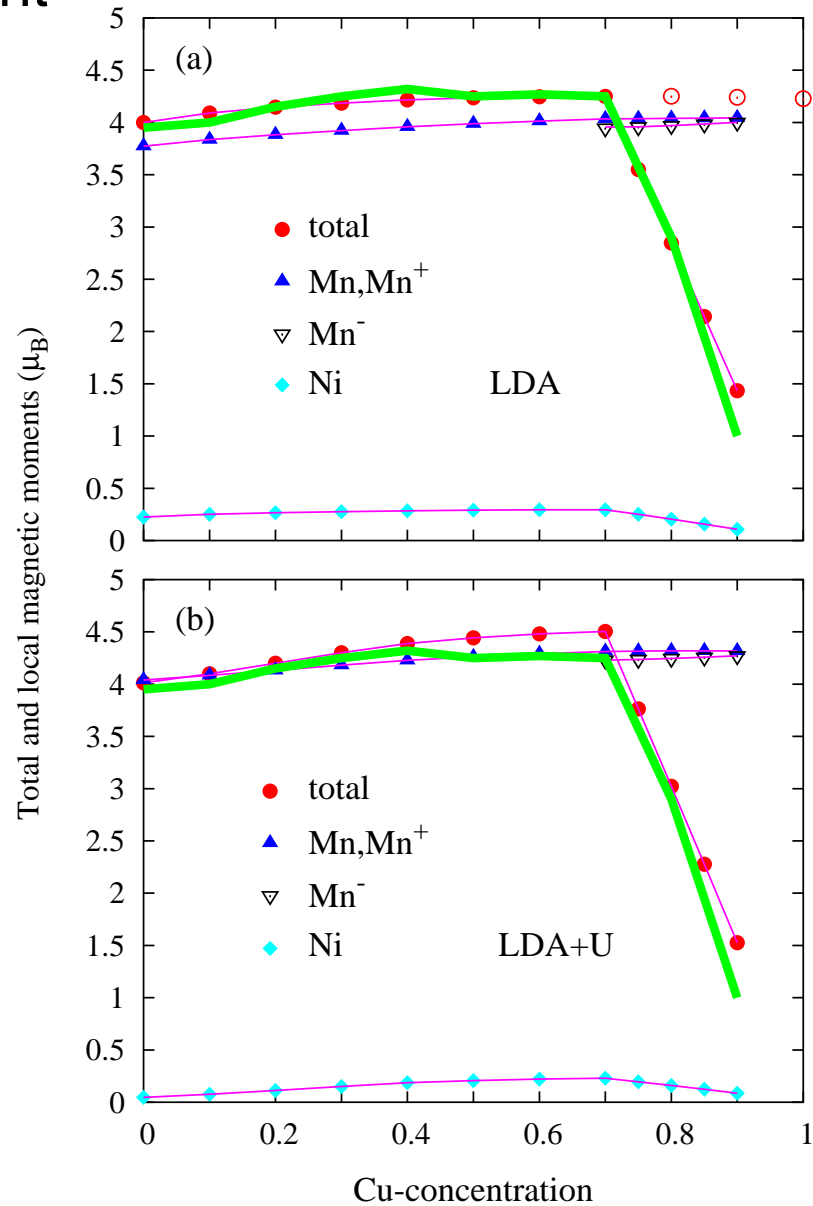
Bloch spectral function of $\text{Cu}_{0.5}\text{Ni}_{0.5}\text{MnSb}$
 weak disorder in Mn states above E_F
 strong chemical disorder in occupied states



magnetic moments $Cu_xNi_{1-x}MnSb$

in good agreement with experiment

(S.K. Ren et al. J. Alloys & Compounds, **387**
(2005),32)



HEISENBERG HAMILTONIAN

$$H = - \sum_{ij} J_{ij} \mathbf{e}_i \cdot \mathbf{e}_j$$

exchange interactions ... J_{ij} ... 62 neighbors
Curie temperature

$$k_B T_C^{MFA} = \frac{2}{3} \sum_j J_{0j}^{Mn, Mn}$$

$$k_B T_C^{RPA} = \frac{2}{3} \left\{ \frac{1}{N} \sum_{\mathbf{q}} \left[J(0)^{Mn, Mn} - J(\mathbf{q})^{Mn, Mn} \right] \right\}^{-1}$$

HEISENBERG HAMILTONIAN

Lichtenstein formula

$$J_{\mathbf{R},\mathbf{R}'}^{M,M'} = \frac{1}{4\pi} \text{Im} \int_C \text{tr}_L \left[\delta_{\mathbf{R}}^M(z) \bar{g}_{\mathbf{R},\mathbf{R}'}^{M,M'\uparrow}(z) \delta_{\mathbf{R}'}^{M'}(z) \bar{g}_{\mathbf{R}',\mathbf{R}}^{M',M\downarrow}(z) \right] dz$$

$$\delta_{\mathbf{R}}^M(z) = P_{\mathbf{R}}^{M,\uparrow}(z) - P_{\mathbf{R}}^{M,\downarrow}(z)$$

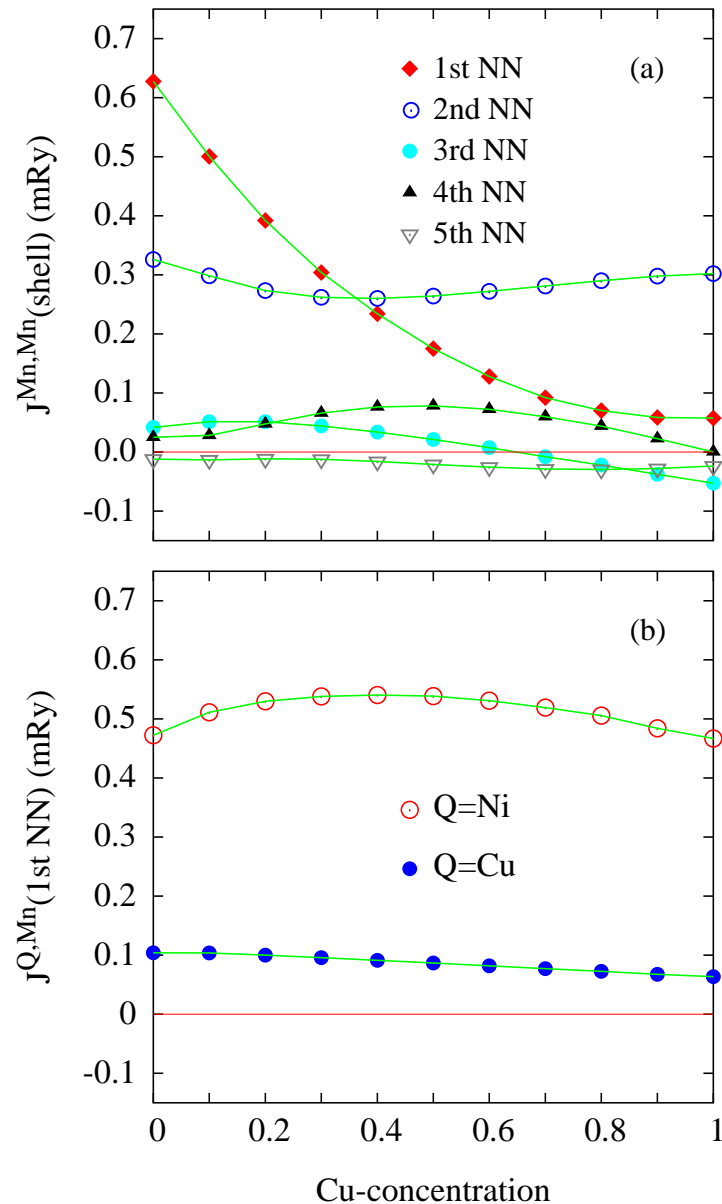
$\bar{g}_{\mathbf{R},\mathbf{R}'}^{M,M'\sigma}(z)$... conditionally averaged Green function

non-random Mn sublattice, only indirect effect of Cu-Ni disorder

exchange interactions in $Cu_x Ni_{1-x} MnSb$

exchange interactions

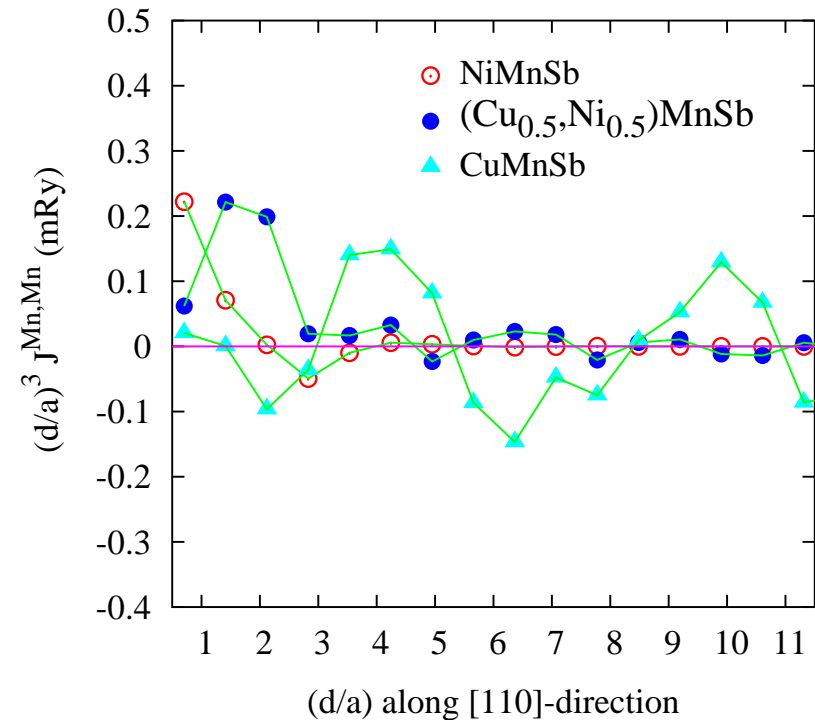
- Mn-Mn important
- mixed Mn-Ni and Mn-Cu beyond first NN very small
- mixed Mn-Ni and Mn-Cu for nearest-neighbors are neglected (tests by L. Sandratskii)
- mixed Ni-Ni, Ni-Cu, Cu-Cu negligible



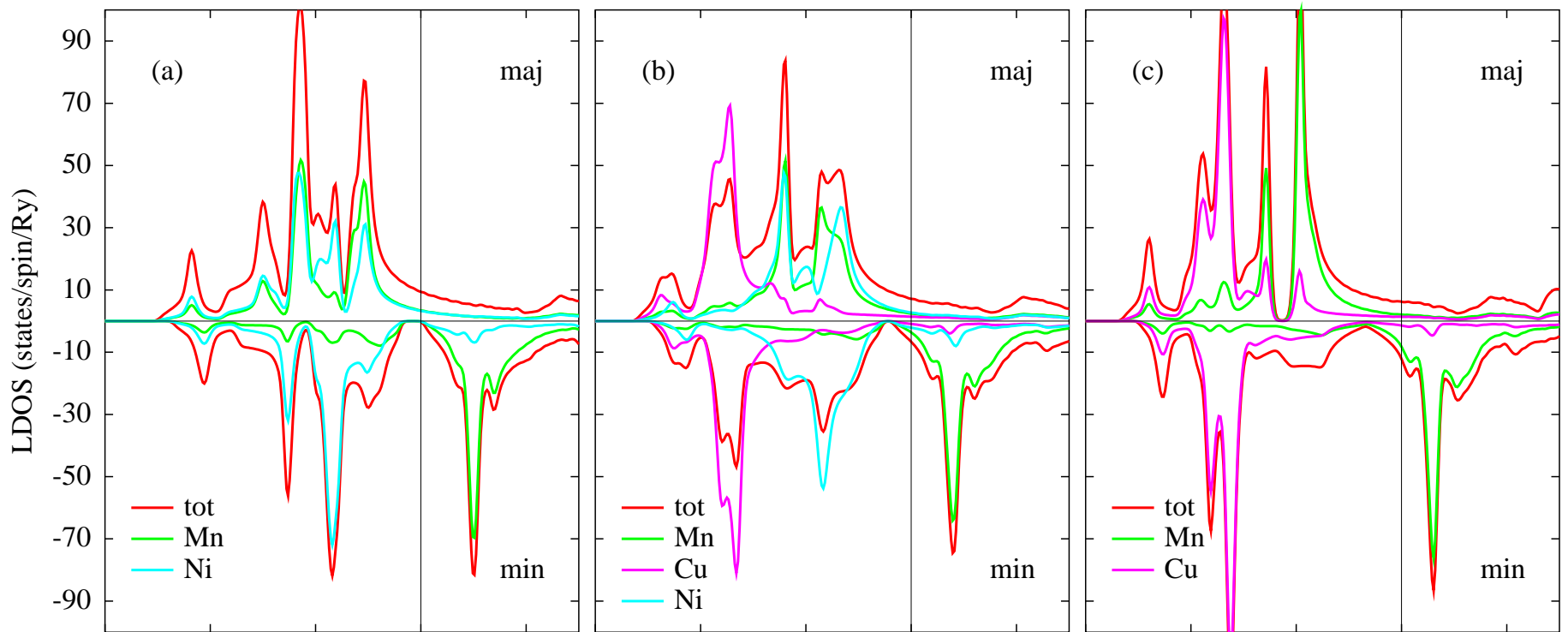
exchange interactions along [1 10] direction

prominent exchange interactions are in [1 10] direction

- NiMnSb: gap in minority states: exponential damping
- CuMnSb: metal ... RKKY-like oscillations $J \propto d^{-3}$
- (Cu_{0.5}Ni_{0.5})MnSb intermediate case

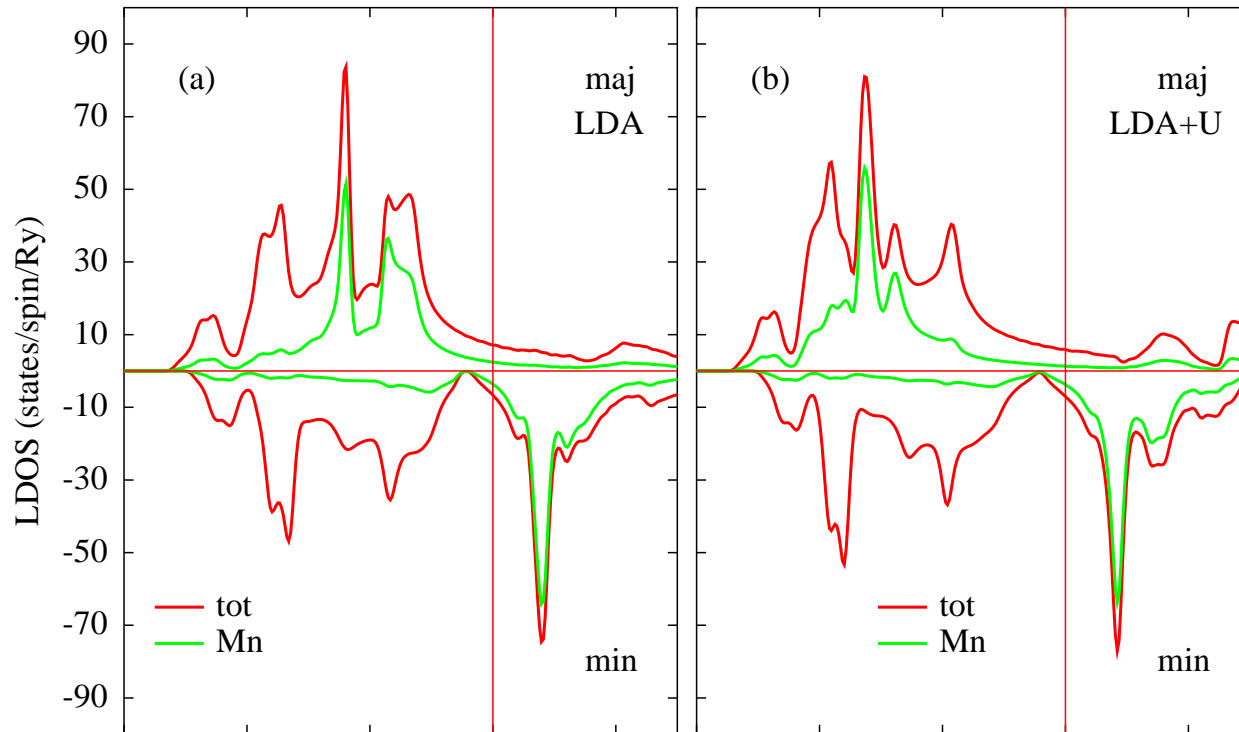


DOS NiMnSb, $\text{Cu}_{0.5}\text{Ni}_{0.5}\text{MnSb}$, CuMnSb



E_F moves towards unoccupied Mn states: superexchange increases
strong disorder on Cu-Ni sublattice for $E \in (-0.4, -0.15)$ Ry

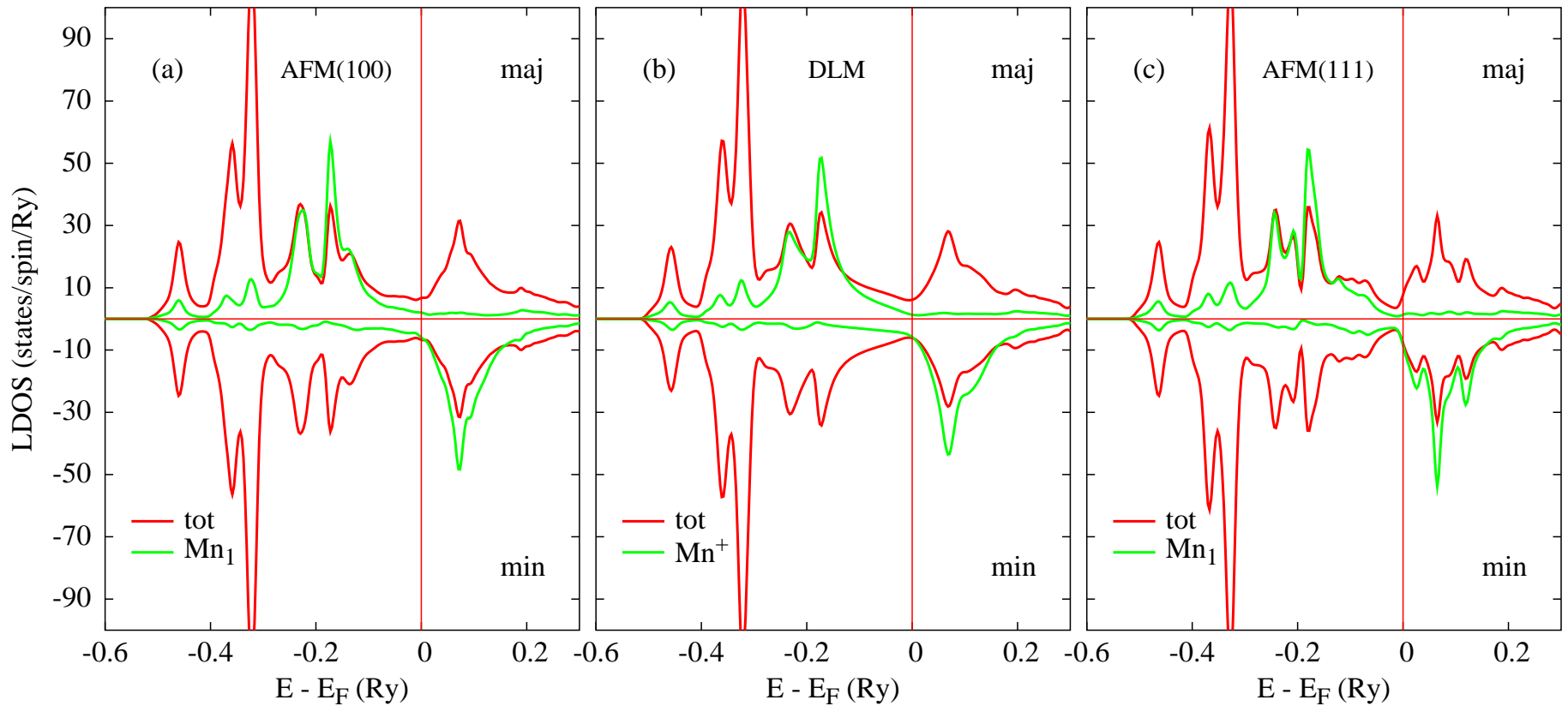
DOS $Cu_{0.5}Ni_{0.5}MnSb$ (LDA, LDA+U)



LDA+U: $U = 0.13$ Ry

Mn majority states moved slightly downwards

DOS $Cu_{0.5}Ni_{0.5}MnSb$ AFM (100), DLM, AFM (111)

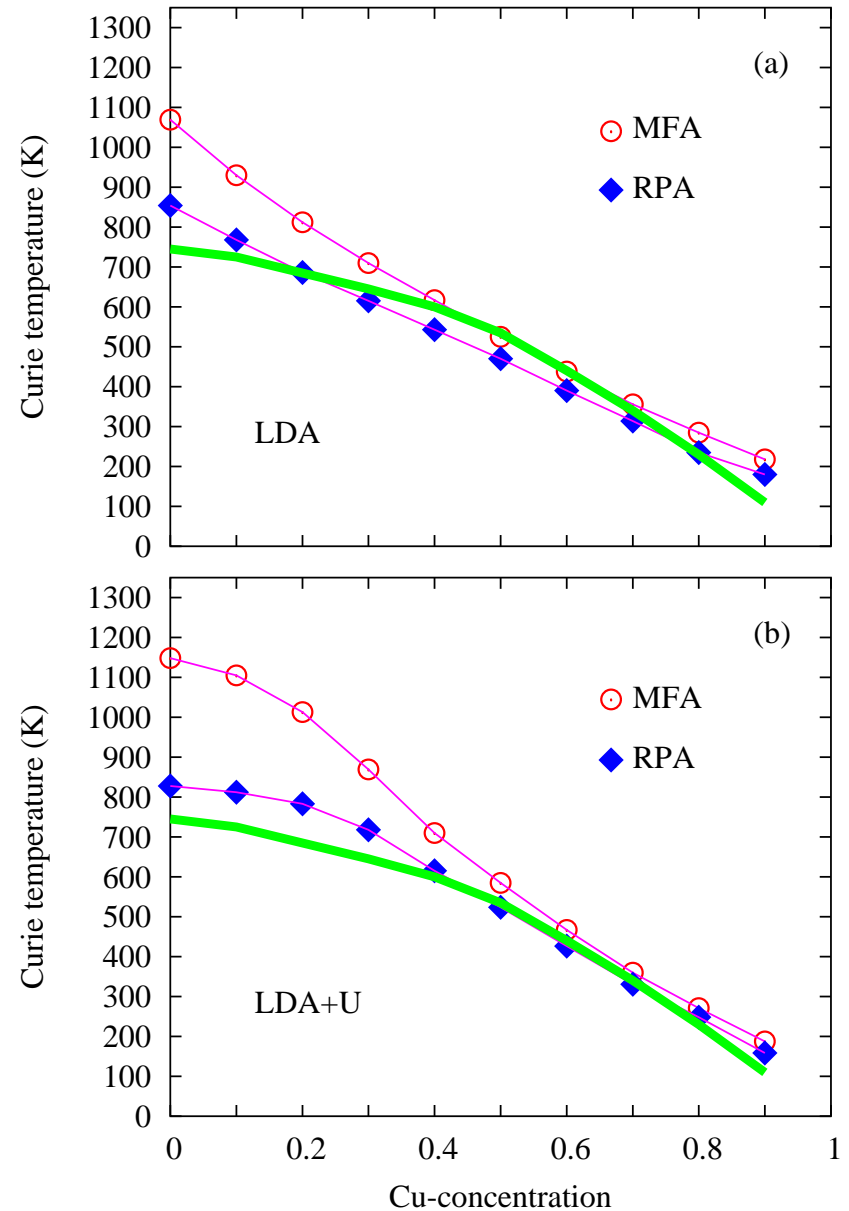


very similar LDOS \implies very similar values of magnetic moments and of exchange integrals

$Cu_xNi_{1-x}MnSb$: Curie temperatures

Curie temperatures

- good agreement of RPA with experiment
- J 's calculated from DLM state yield higher Curie temperature, but induced moments vanish



$Cu_xNi_{1-x}MnSb$: resistivities

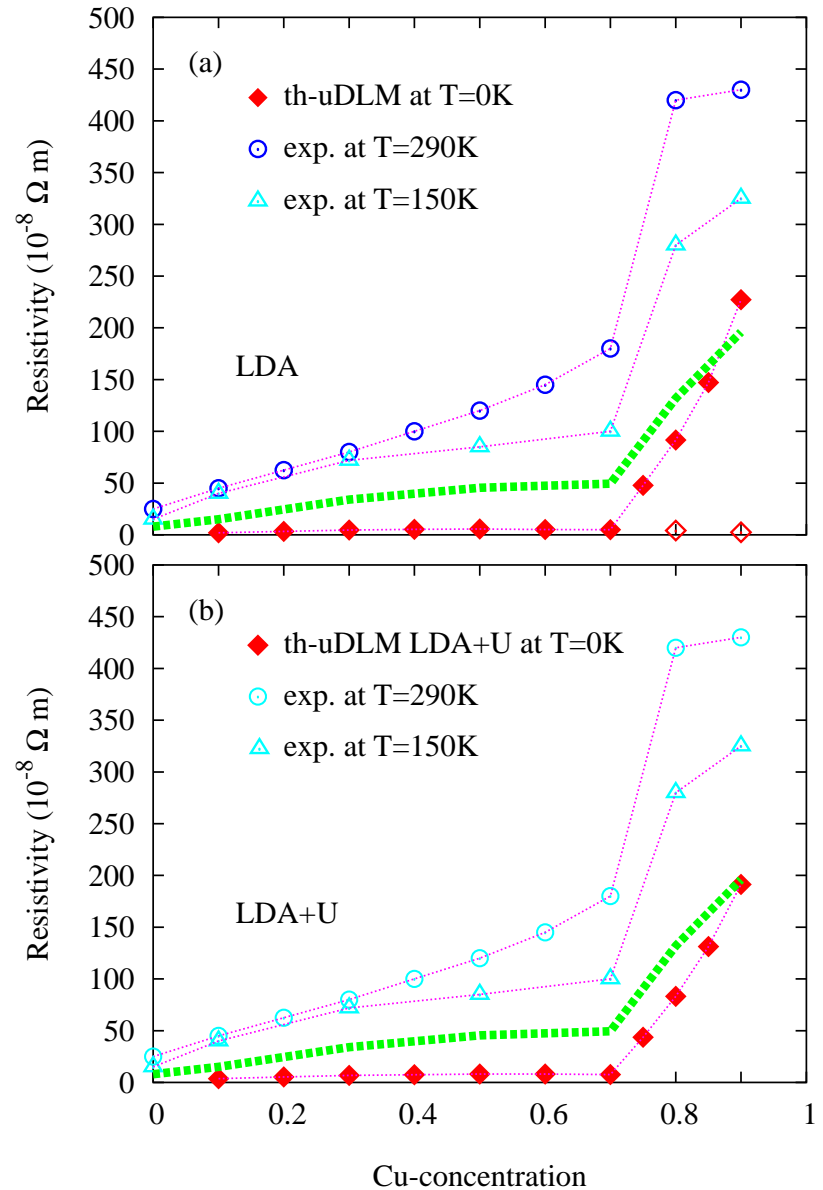
- residual resistivity - scattering on chemical and magnetic disorder
- electron-phonon scattering
- electron-magnon scattering

CPA with vertex corrections (important for magnetic disorder)

e-ph and e-m scatterings neglected

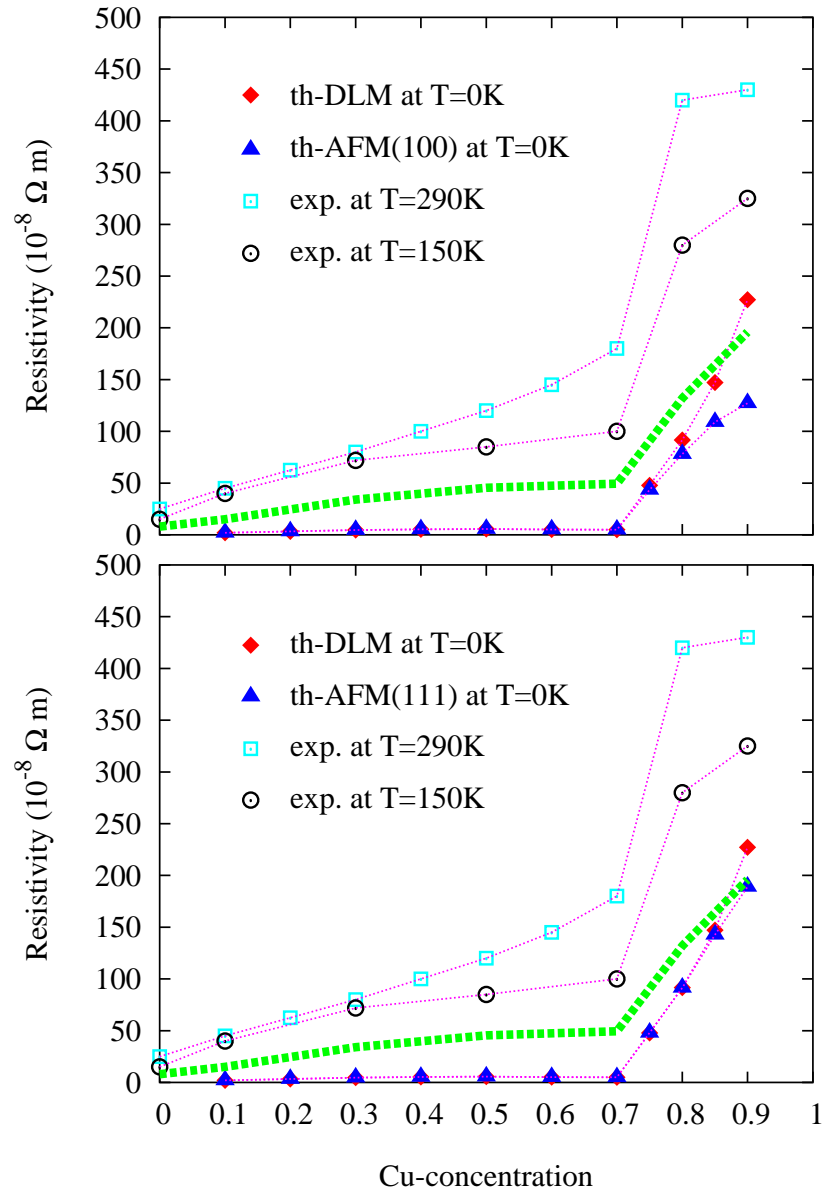
experimental values (S.K. Ren et al. J. Alloys & Compounds, **387** (2005),32) extrapolated to 0 K

resistivities for model I (DLM)



$Cu_xNi_{1-x}MnSb$: resistivities

resistivities for model II (AFM)

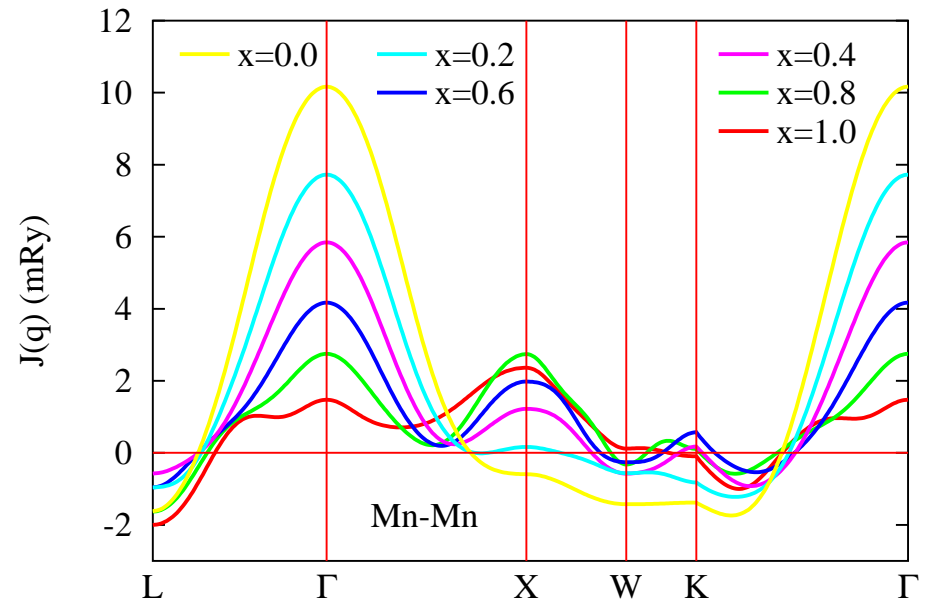


$Cu_x Ni_{1-x} MnSb$: Fourier transform of exchange interaction

exchange interactions

$$J^{Mn,Mn}(\mathbf{q}) = \sum_{\mathbf{R}} e^{i\mathbf{q}\cdot\mathbf{R}} J_{0,\mathbf{R}}^{Mn,Mn}$$

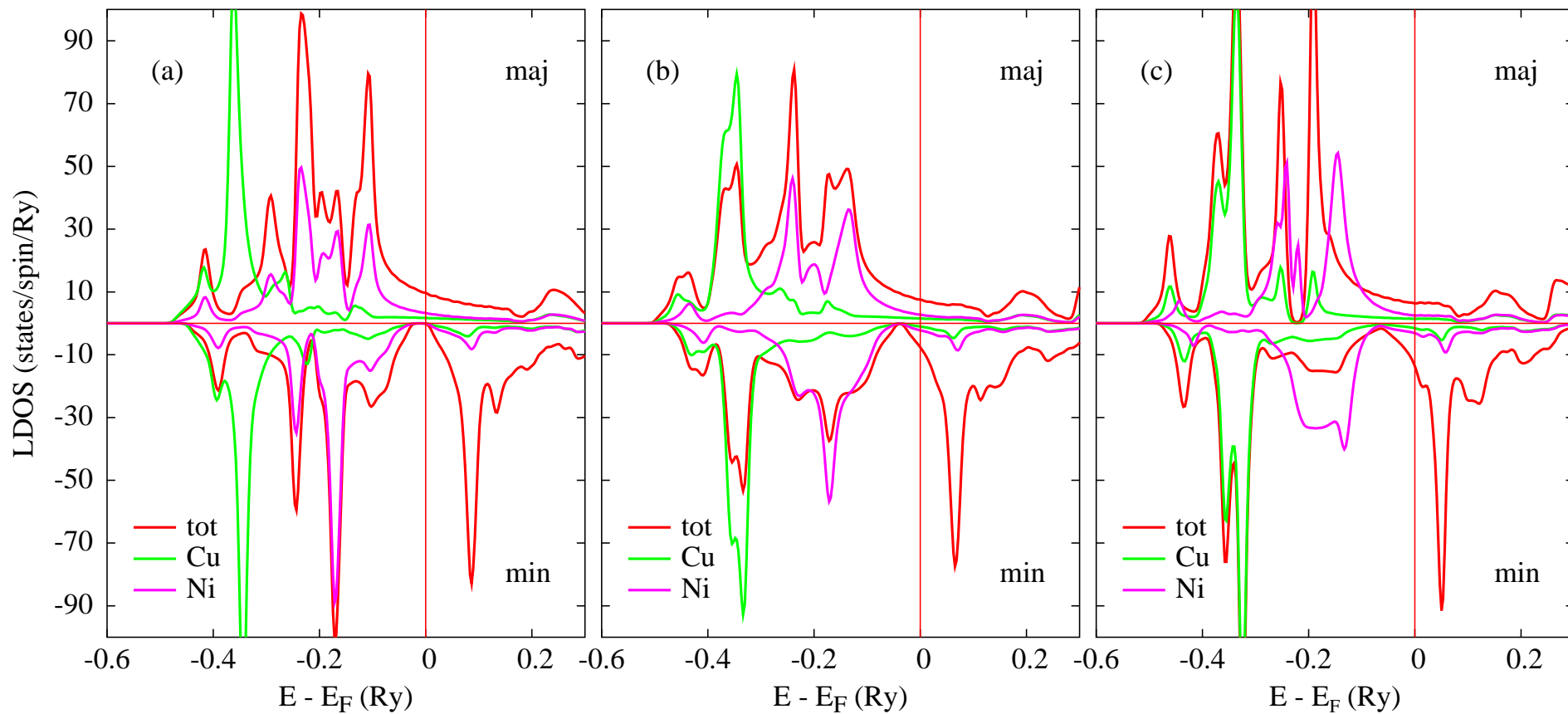
max $J(\mathbf{q})$... minimum of energy



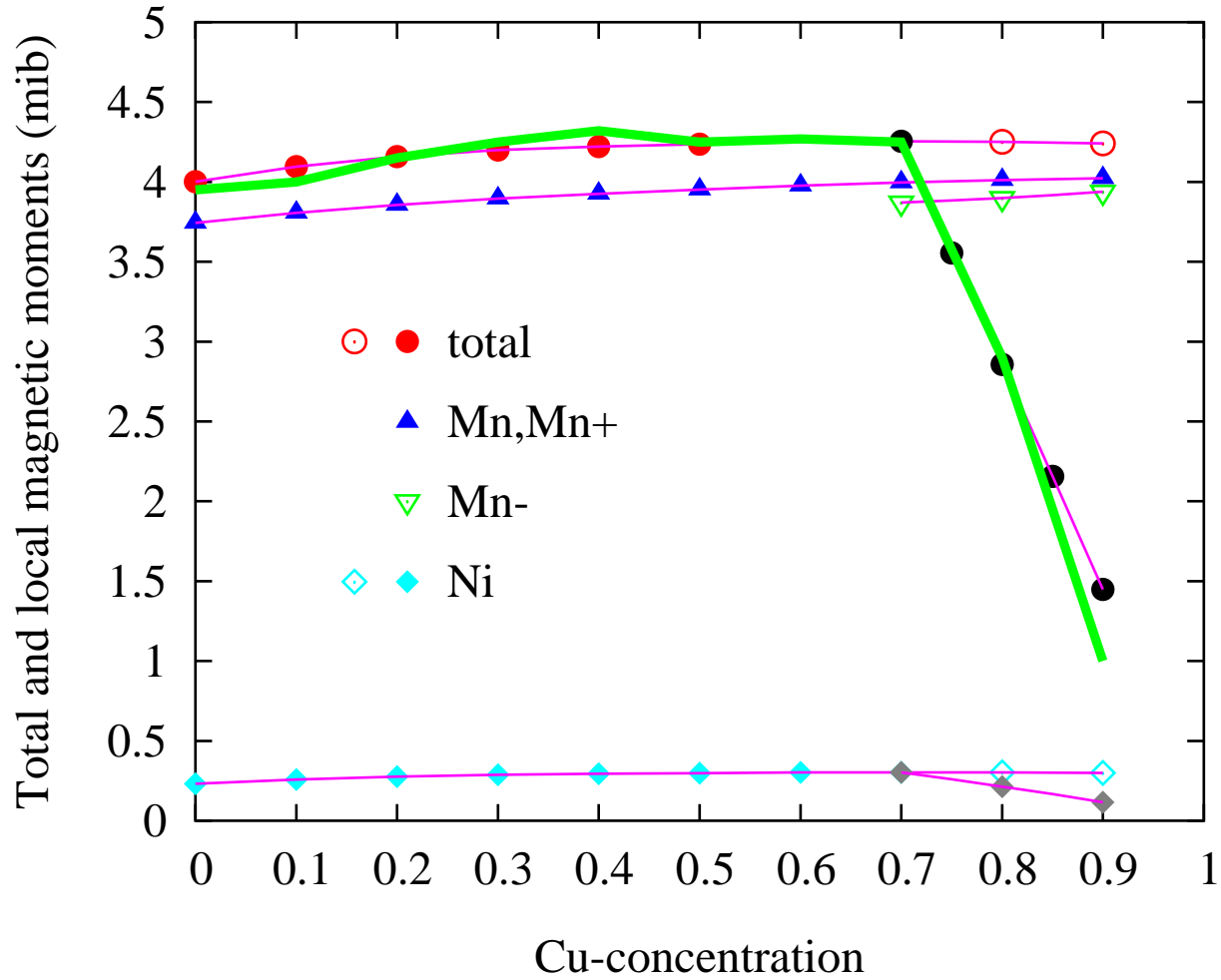
CONCLUSIONS AND OUTLOOK

- simple model of magnetic disorder successfully explains properties of (Cu,Ni)MnSb semi-Heusler alloys
 - sudden decrease of magnetization for $x_{Cu} > 0.7$
 - sudden increase of resistivity for $x_{Cu} > 0.7$
 - smooth and monotonic decrease of T_C with increasing x_{Cu}
- RPA gives T_C in a good agreement with experiment
- results are insensitive to a particular model of magnetic disorder
- open problem: ground state of CuMnSb (theory AFM(100), experiment AFM(111))

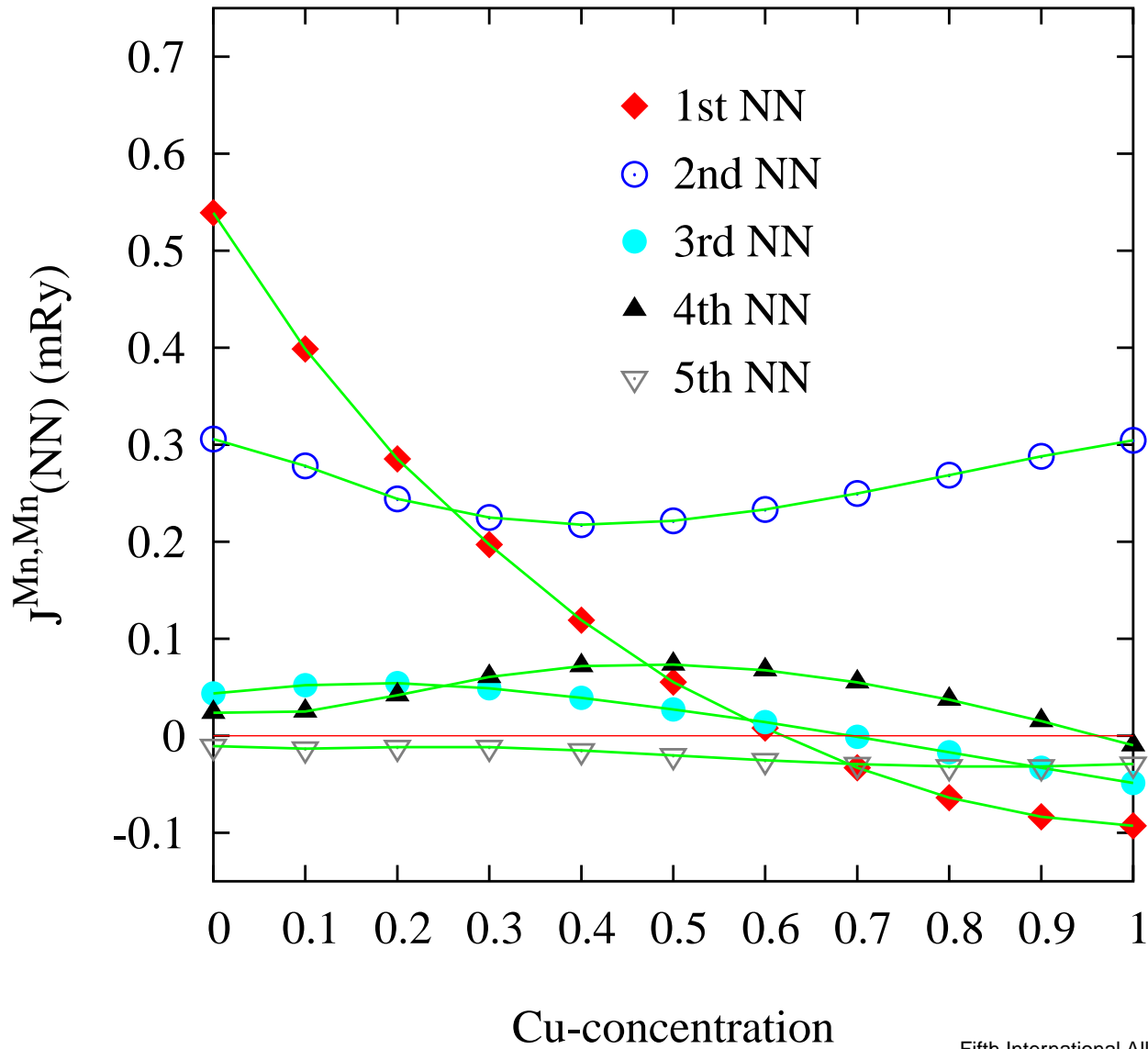
SPDF DOS for NiMnSb, Cu_{0.5}Ni_{0.5}MnSb, CuMnSb



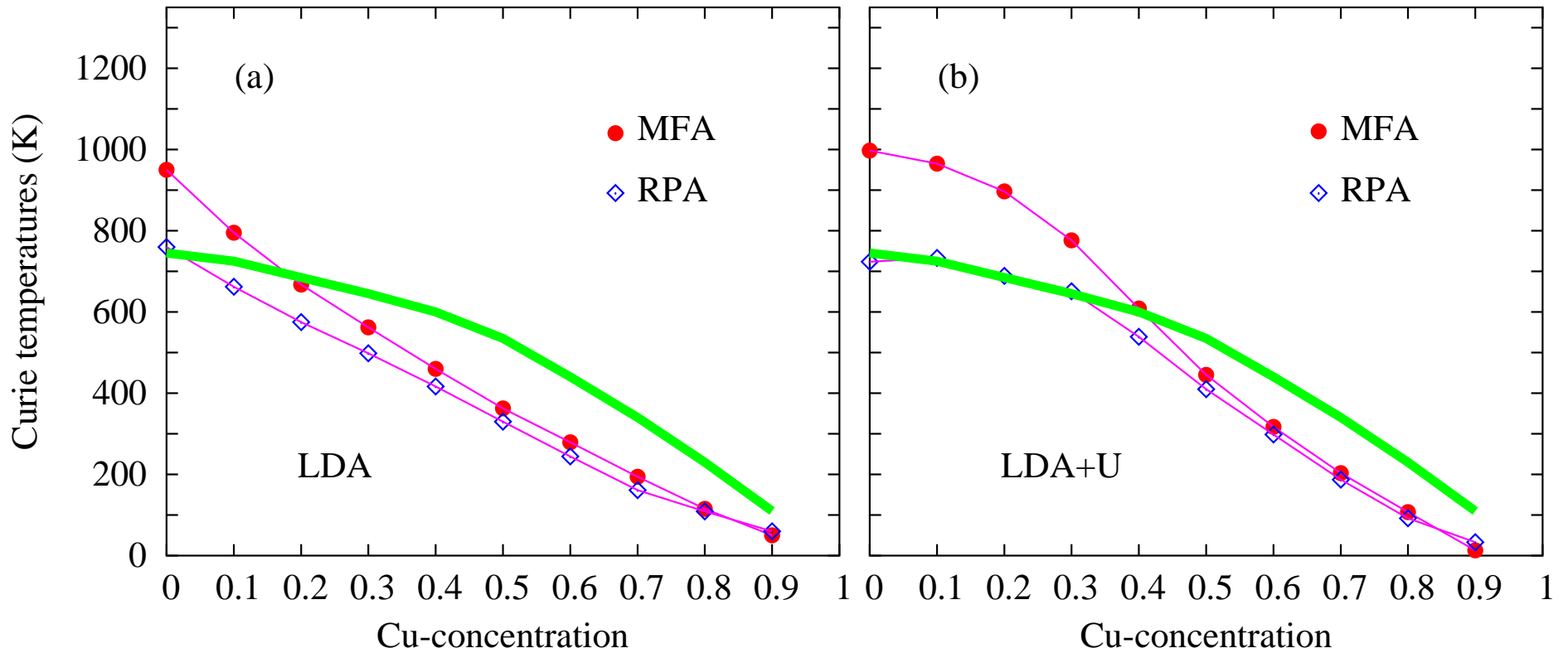
SPDF magnetic moments



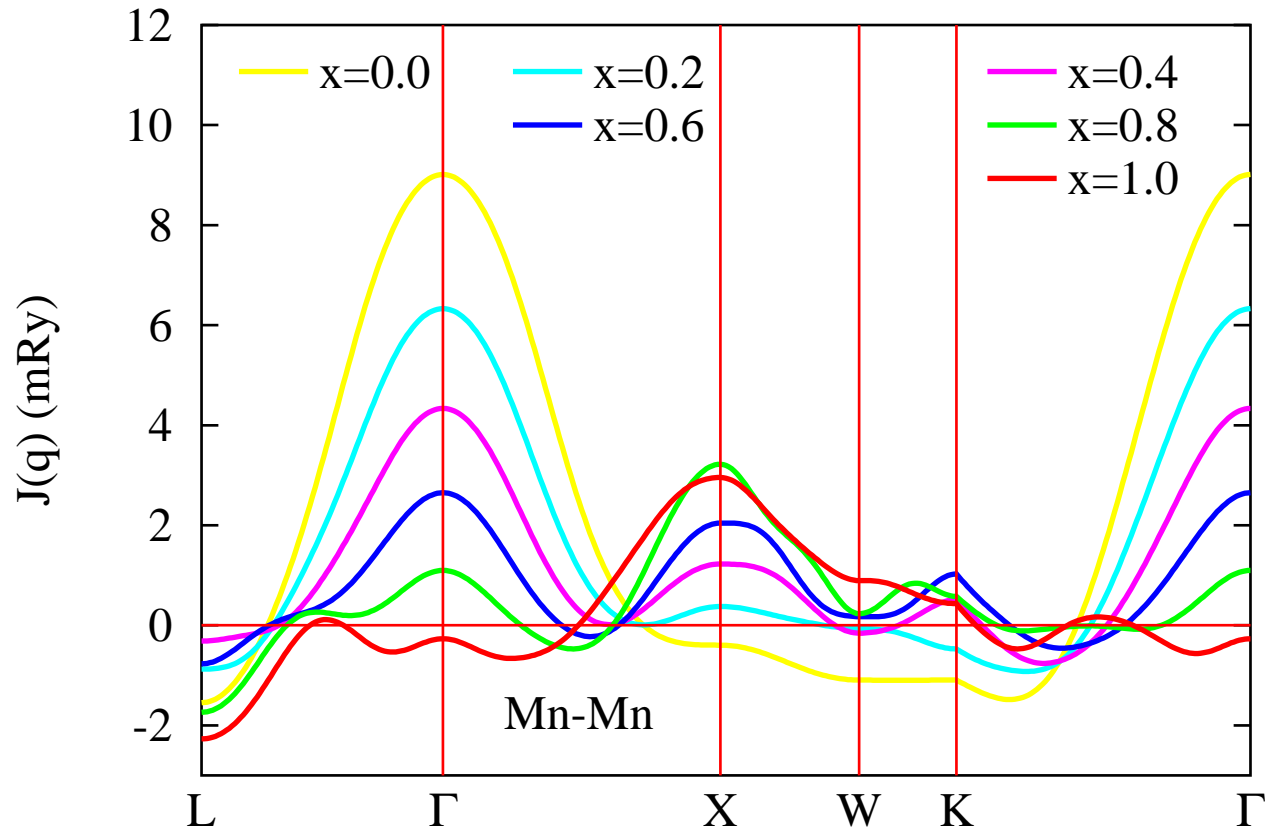
SPDF exchange interactions



SPDF Curie temperatures

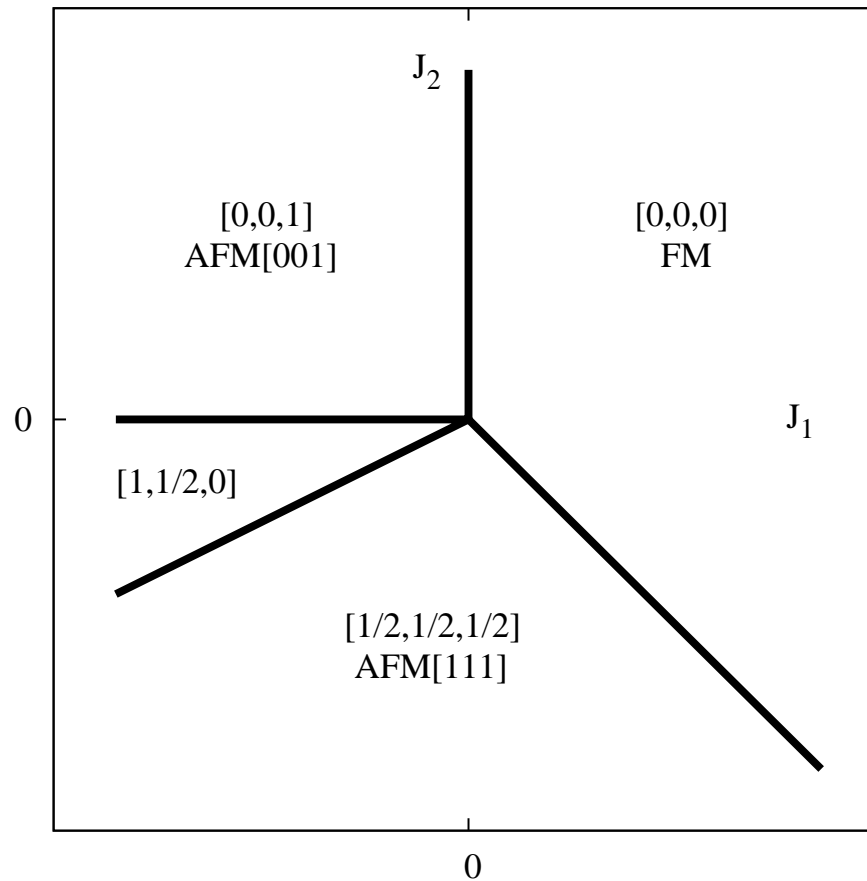


SPDF Fourier transform of exchange interactions



Phase diagram of Ising model

Ising model with 2 NN on fcc-lattice: phase diagram



*