

# EFFECTIVE MAGNETIC HAMILTONIANS FROM FIRST PRINCIPLES

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# MOTIVATION

- magnetic structures are usually determined from total energy calculations:
  - only a few configurations can be investigated
  - approach is limited to the ground state ( $T = 0$ )
- alternatively, the total energy is mapped onto classical Heisenberg Hamiltonian and statistical mechanics are used to study magnetic structure at  $T > 0$ 
  - excited states are accessible
  - all configurations can be investigated
  - temperature dependence of the magnetic structure can be studied
  - magnon spectra, critical temperatures, spin stiffness, etc.
- besides many successes this approach has certain problems:
  - valid only for robust (constant size) moments
  - induced moments (contribution to energy, ghost magnon bands)
  - orientation of magnetic moments with respect to crystallographic axes is not determined

# OUTLINE

- Local (on-site) exchange
- Isotropic exchange (classical Heisenberg Hamiltonian)
- Anisotropic exchange
- Application 1: effective magnetic Hamiltonian for  $3d$  and  $4d$  metals
- Application 2: magnetic structure of magnetic monolayers on non-magnetic substrates
- Application 3: spin-disorder resistivity
- Conclusions and outlook

# PRESENT APPROACH

we wish to treat on equal footing

- local exchange  $H^{\text{LE}}$
- interatomic isotropic exchange  $H^{\text{Heis}}$
- interatomic anisotropic exchange  $H^{\text{aniso}}$

$$H = H^{\text{LE}} + H^{\text{Heis}} + H^{\text{aniso}}$$

## Methods

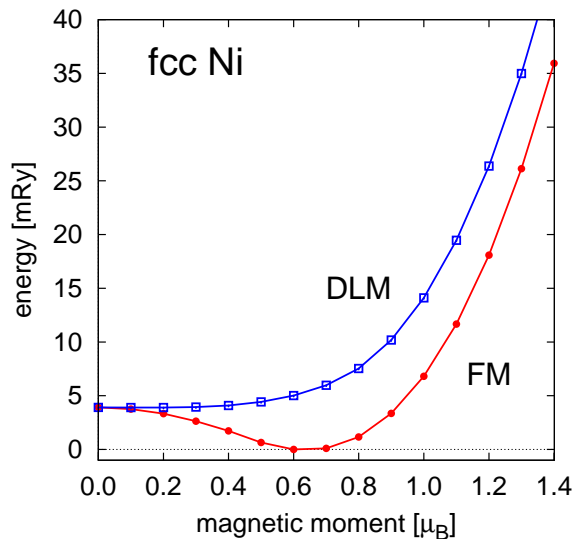
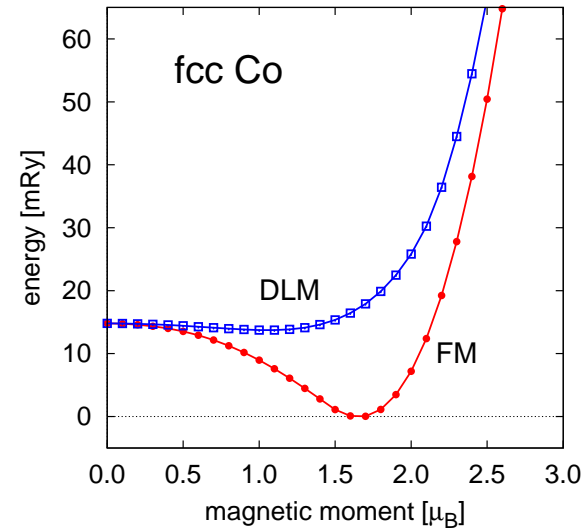
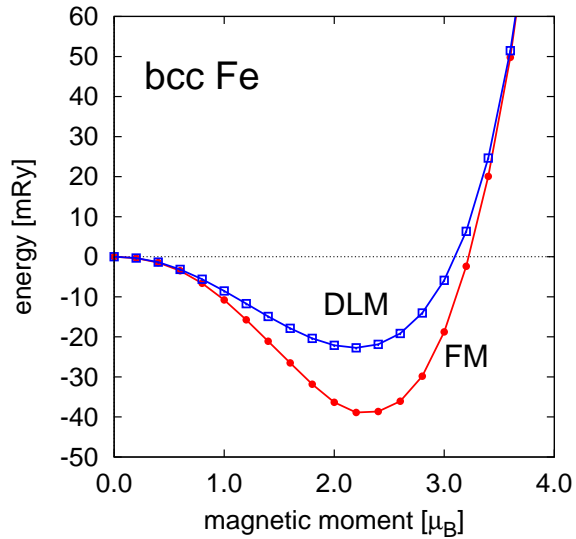
- electronic structure from first principles (in our case usually TB-LMTO-CPA)
- mapping of the total energy onto an effective Hamiltonian  $H^{\text{eff}}$
- find the ground state of  $H^{\text{eff}}$
- use the methods of statistical mechanics to determine magnetic properties of the system at  $T > 0$  such as  $M(T)$ , phase transitions, susceptibilities, ...

# LOCAL EXCHANGE 1

- local exchange together with exchange field from other atoms is responsible for appearance of magnetic moments
- information on the local exchange can be found from constrained LSDA calculations (fixed spin moment method)  
Schwartz, Mohn J. Phys. F **14** L129 (1984), Moruzzi et al. PRB **34** 1784 (1984), Dederichs et al. PRL **53** 2512 (1984)
- fixed spin moment method can be extended to disordered local moments (DLM) state

# LOCAL EXCHANGE 2

## ferromagnetic $3d$ metals

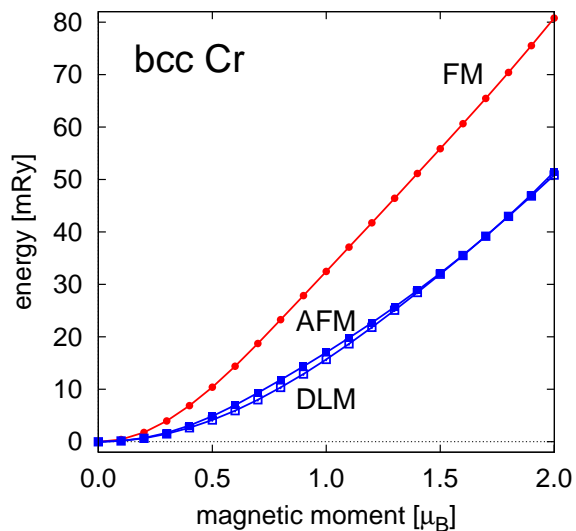
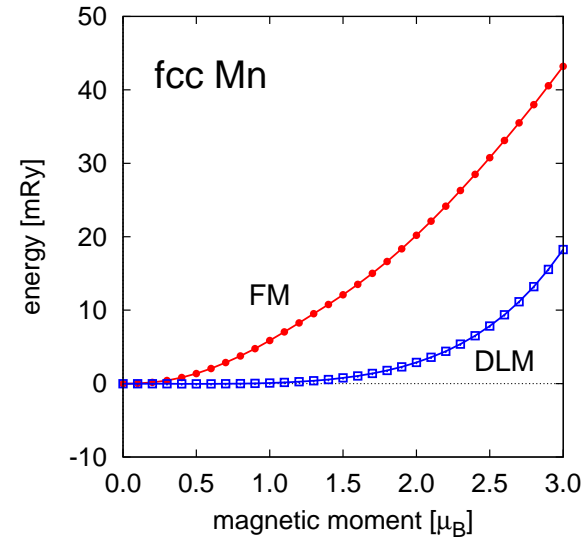
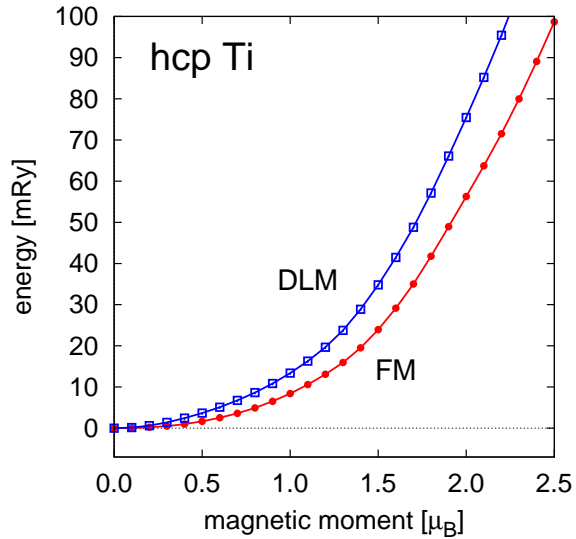


the energy is described with a good accuracy by the 4th degree polynomial

$$H^{\text{LE}} = - \sum_i [A_i + B_i M_i^2 + C_i M_i^4]$$

# LOCAL EXCHANGE 3

## other $3d$ metals



usually for small  $M_i$  quadratic polynomial is sufficient

$$H^{\text{LE}} = - \sum_i B_i M_i^2$$

but not always (e.g. DLM Mn)

# LOCAL EXCHANGE 4

$$E(M) \approx \frac{1}{2}D(M - M_0)^2$$

system	$M_0[\mu_B]$	$B$ [mRy/ $\mu_B^2$ ]	$C$ [mRy/ $\mu_B^4$ ]	$D$ [mRy/ $\mu_B^2$ ]
bcc-Fe FM	2.280	-16.07	1.53	33.85
fcc-Co FM	1.656	-15.69	3.35	108.31
fcc-Ni FM	0.639	-3.60	12.71	108.66
bcc-Fe DLM	2.193	-10.90	1.15	18.14
fcc-Co DLM	1.056	-8.20	2.63	9.83
fcc-Ni DLM	0.118	-1.88	12.13	1.59
bcc-Cr FM	0.000	13.58	-0.30	92.79
bcc-Cr DLM	0.000	16.83	-1.17	36.40
fcc-Mn FM	0.000	5.28	-0.06	15.86
fcc-Mn DLM	0.526	-0.34	0.26	0.54
fcc-Cu FM	0.000	109.93	5.26	405.24



# EXCHANGE ENERGY

DLM state:

$$E_{\text{tot}}^{\text{DLM}} \approx E_{\text{intra}}^{\text{exch}}$$

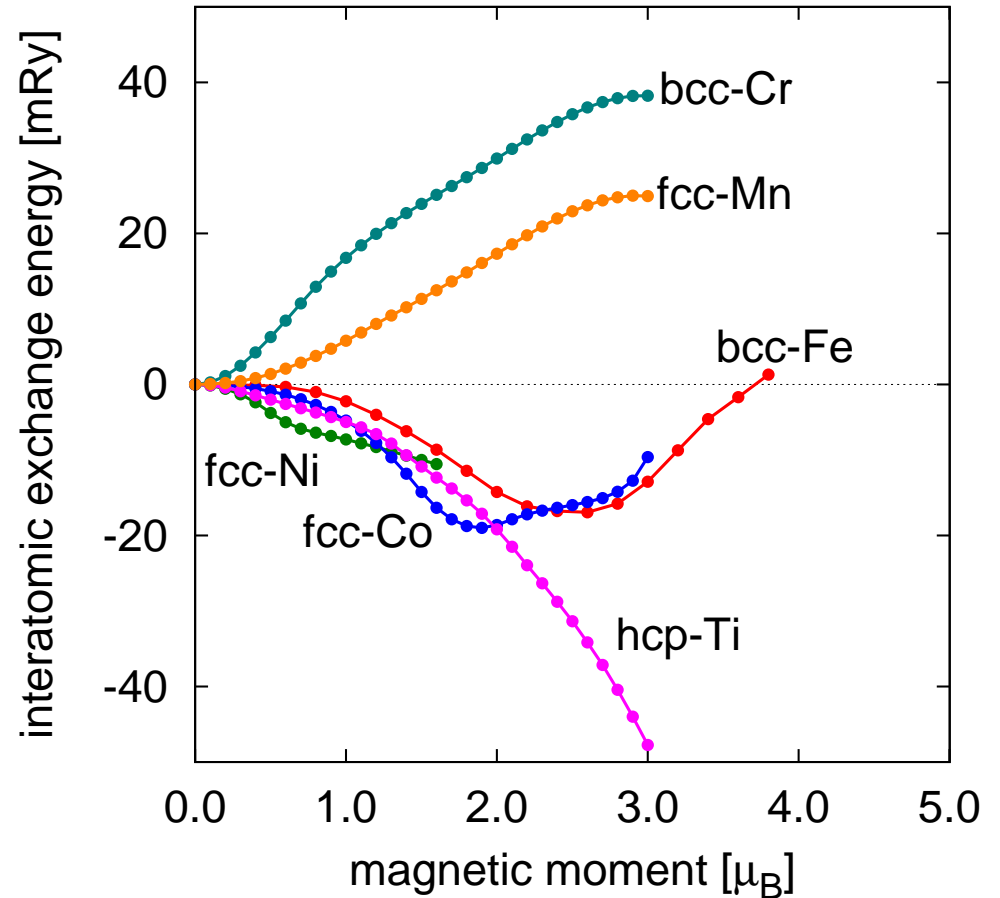
... intraatomic exchange energy

FM state:

$$E_{\text{tot}}^{\text{FM}} \approx E_{\text{inter}}^{\text{exch}} + E_{\text{intra}}^{\text{exch}}$$

... sum of interatomic and intraatomic exchange energy

$$E_{\text{inter}}^{\text{exch}} = E_{\text{tot}}^{\text{FM}} - E_{\text{tot}}^{\text{DLM}}$$



- negative interatomic exchange energy: tendency to FM ordering
- positive interatomic exchange energy: tendency to AFM ordering
- Ti: strong exchange field, but atomic polarization requires large energy

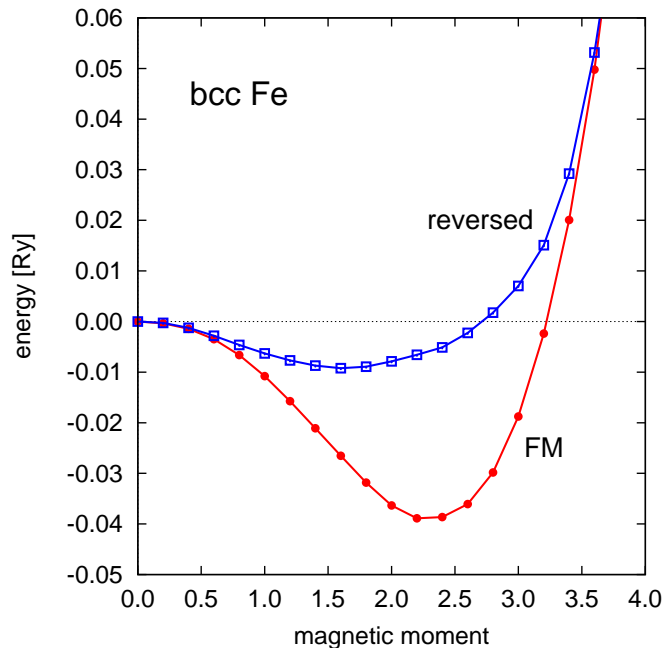
# REVERSED MOMENTS

consider one reversed moment in a ferromagnetic material, the interatomic exchange acting on this atom is reversed and we expect that the energy of such atom is changed to

$$E^{\text{reversed}} = E_{\text{intra}}^{\text{exch}} - E_{\text{inter}}^{\text{exch}} = E_{\text{tot}}^{\text{DLM}} - E_{\text{inter}}^{\text{exch}} = 2 E_{\text{tot}}^{\text{DLM}} - E_{\text{tot}}^{\text{FM}}$$

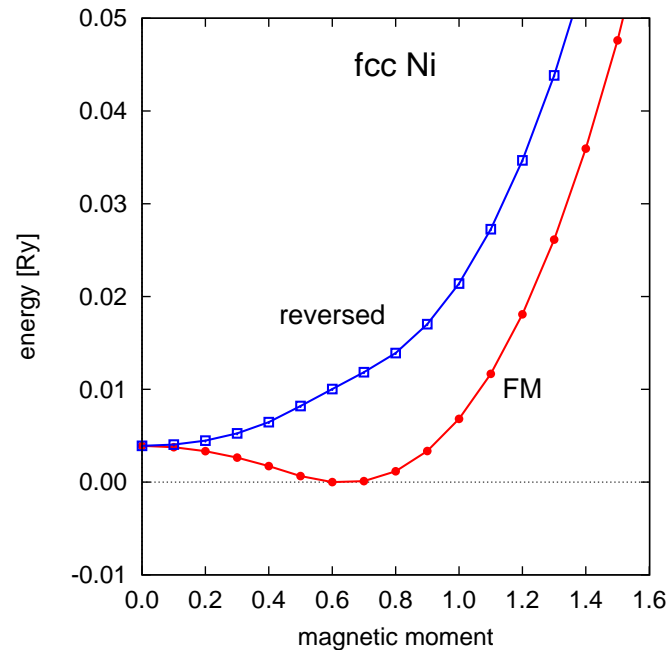
## Iron

energy is increased  
moment is diminished



## Nickel

moment disappears



# ISOTROPIC (HEISENBERG) EXCHANGE 1

$$H^{\text{Heis}} = - \sum_{ij} J_{ij} \mathbf{e}_i \cdot \mathbf{e}_j, \quad \mathbf{e}_i = \frac{\mathbf{M}_i}{|\mathbf{M}_i|} \dots \text{unit vectors}$$

exchange interactions ...  $J_{ij}$

$J_{ij} > 0$  – ferromagnetic coupling

$J_{ij} < 0$  – antiferromagnetic coupling

magnetic force theorem: Liechtenstein formula: Liechtenstein et al. JMMM **67** 65 (1987),  
Turek et al. Phil. Mag. **86** 1713 (2006)

$$J_{ij} = \frac{1}{4\pi} \text{Im} \int_C \text{tr}_L \left[ \Delta_i(z) \bar{g}_{ij}^{\uparrow}(z) \Delta_j(z) \bar{g}_{ji}^{\downarrow}(z) \right] dz$$

$$\Delta_i(z) = P_i^{\uparrow}(z) - P_i^{\downarrow}(z)$$

$\bar{g}_{ij}^{\sigma}(z)$  ... intersite block of the Green function

- responsible for mutual orientation of moments, but the orientation of moments with respect to crystallographic axes is indetermined
- higher order interactions, e.g.,  $J_{ijkl}(\mathbf{e}_i \cdot \mathbf{e}_j)(\mathbf{e}_k \cdot \mathbf{e}_l)$  might be important

# ANISOTROPIC EXCHANGE INTERACTIONS

- responsible for orientation of moments with respect to crystallographic axes
- origin: relativistic terms (spin-orbit coupling) and dipole-dipole interactions

$$H^{\text{anisotropy}} = \sum_i K(\mathbf{e}_i) - \sum_{i,j} \mathbf{e}_i^T \mathbb{J}_{ij}^S \mathbf{e}_i - \sum_{i,j} \mathbf{D}_{ij} [\mathbf{e}_i \times \mathbf{e}_j]$$

$K(\mathbf{e}_i)$  ... on-site anisotropy energy

$\mathbb{J}_{ij}^S$  ... symmetric part of interatomic exchange,  $\text{Tr } \mathbb{J}_{ij}^S = 0$ :

contains crystal field effects and dipole-dipole interaction

$\mathbf{D}_{ij}$  ... Dzyaloshinski-Moriya vector (antisymmetric part of interatomic exchange)

L. Udvardi et al. PRB **68** 104436 (2003) ... KKR expressions

## dipole-dipole interaction

$$E^{\text{dip}} = k \frac{\mathbf{M}_{\mathbf{R}} \cdot \mathbf{M}_{\mathbf{R}'} - 3 (\mathbf{M}_{\mathbf{R}} \cdot \mathbf{n}) (\mathbf{M}_{\mathbf{R}'} \cdot \mathbf{n})}{|\mathbf{R} - \mathbf{R}'|^3},$$

$\mathbf{n} = (\mathbf{R} - \mathbf{R}')/|\mathbf{R} - \mathbf{R}'|$  is the unit vector in the direction  $\mathbf{R} - \mathbf{R}'$ , and  $\mathbf{M}_{\mathbf{R}}$  is the total magnetic moment at site  $\mathbf{R}$ ,  $k = 2.66 \times 10^{-5}$  in atomic Rydberg units

# GROUND STATE (T=0)

## systems with one sublattice

isotropic case: the ground state is given by a single  $\mathbf{q}$ -vector wave

$$\mathbf{e}_i(\mathbf{q}) = \left( \cos(\mathbf{q} \cdot \mathbf{R}_i), \sin(\mathbf{q} \cdot \mathbf{R}_i), 0 \right)$$

which corresponds to the maximum of the Fourier transform of exchange interactions:

$$J(\mathbf{q}) = \sum_j e^{i\mathbf{q} \cdot \mathbf{R}_j} J_{0,j}, \quad E(\mathbf{q}) = -J(\mathbf{q})$$

anisotropic case: the energy of the ground state is given by the **largest eigenvalue** of the  $3 \times 3$  matrix  $\mathbb{J}(\mathbf{q})$  and the orientation of moments follows from the corresponding eigenvector

## systems with two or more sublattices

theory is rather complicated, see Alexander PR **127** 420 (1962)

# FINITE TEMPERATURES

methods of statistical mechanics:

- mean field: low accuracy, overestimates ordering temperature
- random-phase approximation (RPA)
- Monte Carlo simulations: high accuracy, high demands on computer resources

Remark:

fixed-size moments :  $Z_0 = \int_{|\mathbf{M}|=M} d^2 M e^{\beta \mathbf{M} \cdot \mathbf{H}}$  give Langevin function

variable-size moments :  $Z_0 = \int d^3 M e^{\beta \mathbf{M} \cdot \mathbf{H} - \beta(A + BM^2 + CM^4)}$

are analytically intractable (problem of Mexican hat in an external field)

# Fe MONOLAYER ON Ir(001) SURFACE 1

## experiment

- high-quality layers without clustering
- negligible intermixing of Fe and Ir
- geometry known from LEED: reduced interlayer distance between Fe and Ir layers
- MOKE: no magnetic signal from a monolayer

## ab initio theory

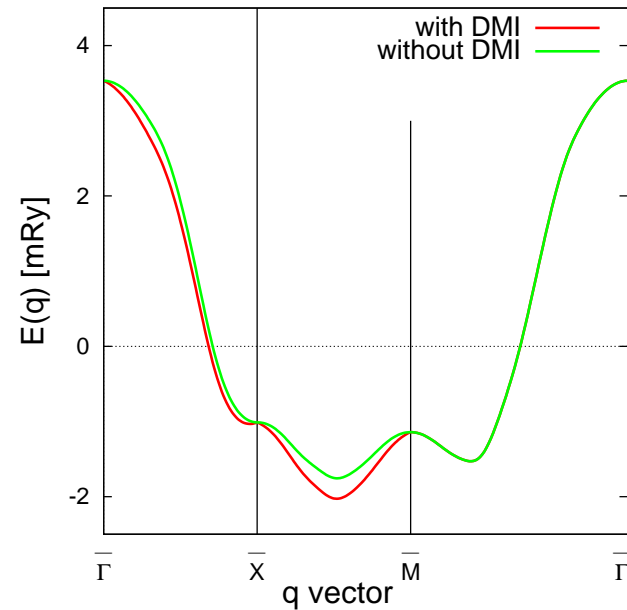
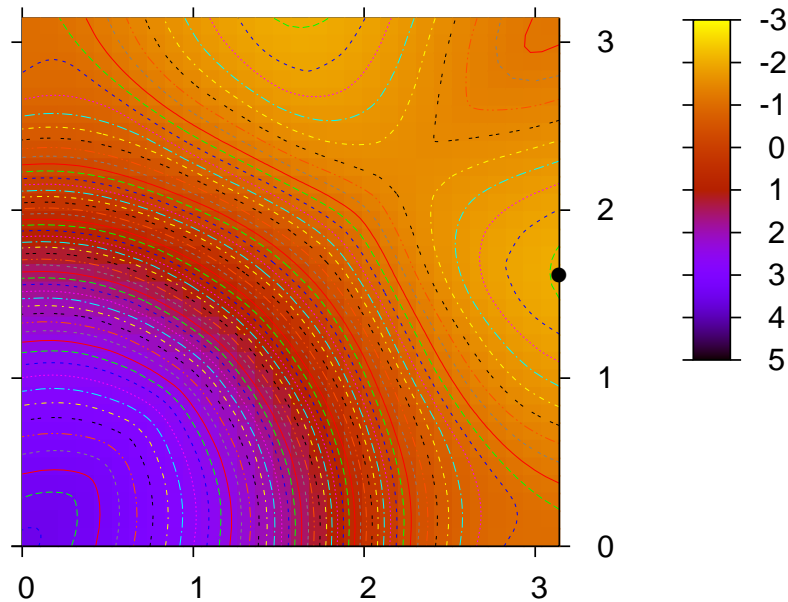
Kudrnovský et al.: PRB **80** (2009), 064405 (VASP, WIEN2k, LMTO)

- isotropic Heisenberg Hamiltonian
- reduced interlayer distance (-12 %) is of vital importance for correct description:
  - without relaxation: FM
  - with relaxation: tendency to AFM ordering

Deák et al.: PRB **84** (2011), 224413 (TB-KKR)

- isotropic Heisenberg Hamiltonian
- Dzyaloshinskii-Moriya interactions (DMI)
- biquadratic interactions
- extensive simulations

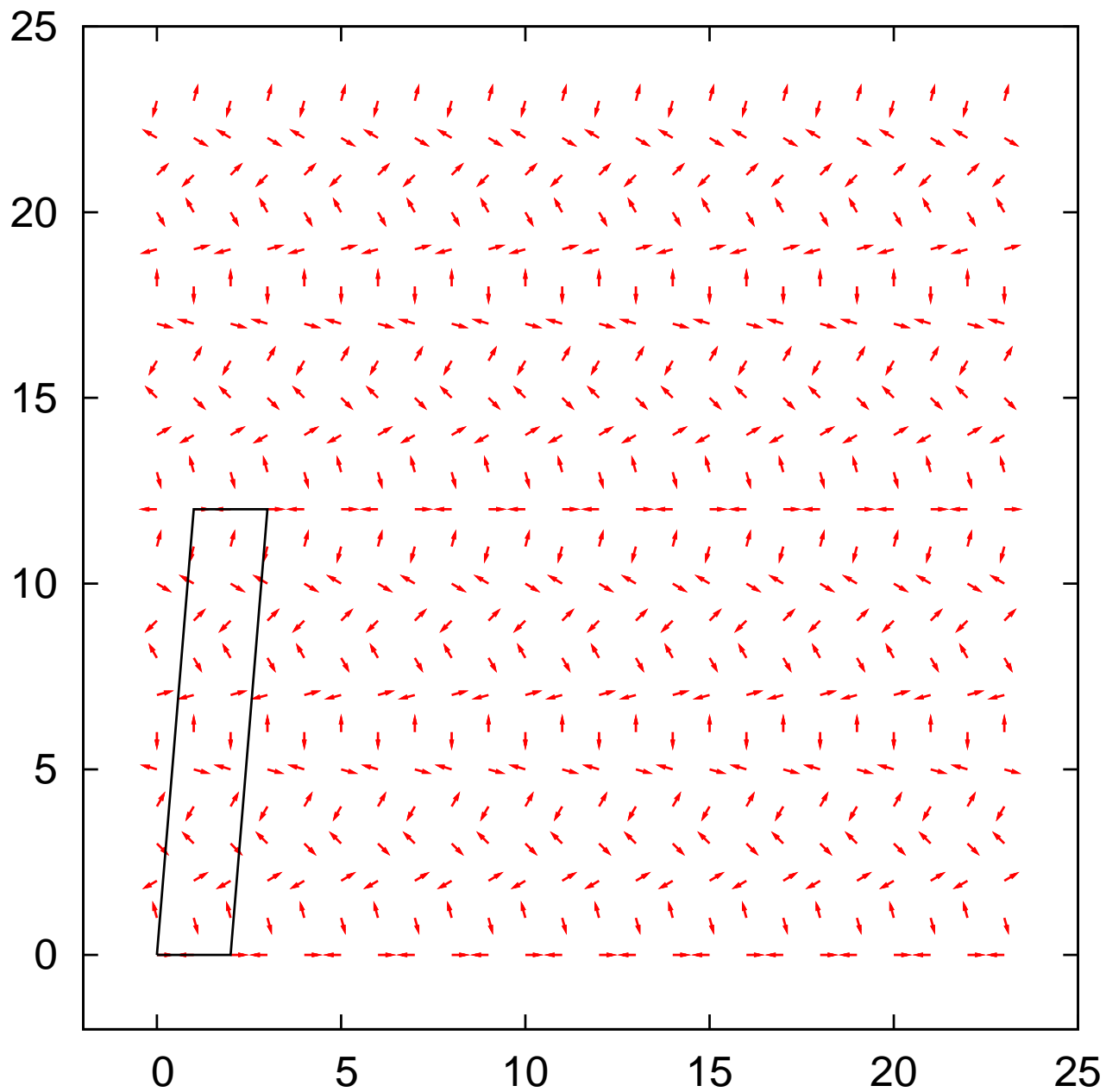
# Fe MONOLAYER ON Ir(001) SURFACE 2



- anisotropic interactions change the energy of the ground state
- anisotropic interactions change very little the position  $\mathbf{q}_0$  of energy minimum
- we find  $\mathbf{q}_0 \approx \frac{\pi}{a}(1, \frac{7}{12})$
- elementary cell:  $\mathbf{A}_1 = a(2, 0)$      $\mathbf{A}_2 = a(1, 12)$
- basis: 24 Fe atoms, directions of moments given by angle  $\phi = x.120^\circ + y.105^\circ$ , where  $x$  and  $y$  are coordinates of atom in square lattice
- all moments lie in one plane ( $xz$ )



# Fe MONOLAYER ON Ir(001) SURFACE 3



# SPIN DISORDER RESISTIVITY 1

Weiss, Marotta: J. Phys. Chem. Solids **9** (1959), 302

Wysocki et al.: PRB **80** (2009), 224423

Wysocki et al. APS March Meeting (2011), L19.1

scattering on spin disorder above  $T_C$  is simulated by disordered local moments  
good agreement with experimental data for Fe, but for Ni theoretical  $\rho_{\text{mag}}$  is approx.  
twice larger than the experimental value

**remedy:** consider moments in Nickel reduced to 0.3 - 0.4  $\mu_B$

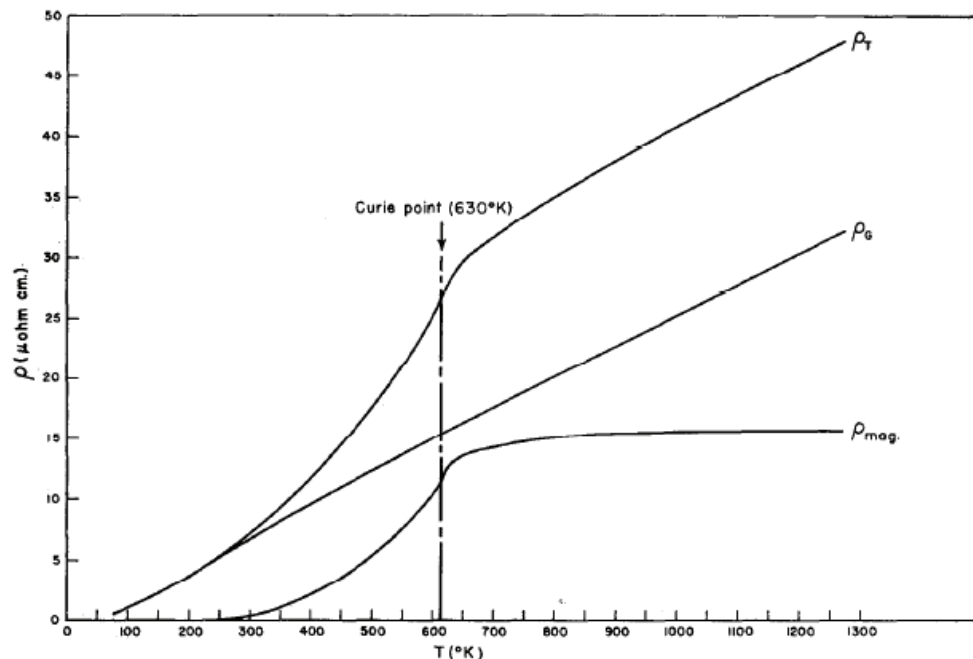


FIG. 2.  $\rho_T$ , the total,  $\rho_{\text{MAG}}$ , the magnetic and,  $\rho_B$ , the lattice components of resistivity for nickel as a function of temperature.

# SPIN DISORDER RESISTIVITY 2

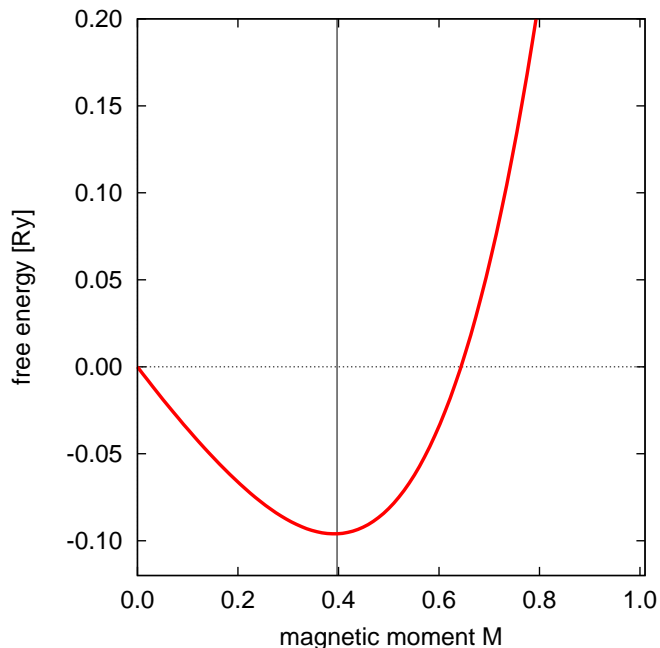
how large are the moments on Ni atoms at Curie temperature?

rough estimate:

- average exchange field is zero in the DLM state
- energy per atom is  $A + BM^2 + CM^4$
- entropy per atom is  $\log(2J + 1)$ , where  $M = g\mu_B J$

minimize free energy at  $T = T_C$  w.r.t.  $M$ :

$$F(M) = A + BM^2 + CM^4 - k_B T_C \log(2J + 1)$$



$$M = 0.397\mu_B$$

this is in agreement with neutron scattering data ( $0.4 \mu_B$ ) of Acet et al.: Europhys. Lett. **40** (1997) 93 and theory ( $0.42 \mu_B$ ) of Ruban et al.: PRB **75** (2007) 054402.

# CONCLUSIONS AND OUTLOOK

- versatile tool for determination of magnetic structure
- yields a deeper understanding of the formation and ordering of magnetic moments
- open problems:
  - statistical mechanics of variable–size moments
  - dependence of  $\mathbb{J}(\mathbf{q})$  on size of moments
  - ground state of systems with several sublattices
  - reliable determination of anisotropic and higher-order interactions from first principles