Ab Initio Calculation of Exchange Interactions, Adiabatic Spin-Waves, and Curie Temperature of Itinerant Ferromagnets

Patrick Bruno

Max-Planck-Institut für Mikrostrukturphysik, Halle, Germany

People involved:

- Josef Kudrnovský (Prague and Halle)
- Vaclav Drchal (Prague)
- Ilja Turek (Brno)
- Marek Pajda (Halle)
- Leonid Sandratskii (Halle)

Summary:

- motivations
- adiabatic spin-waves
- the ``magnetic force theorem´´
- results: spin-wave energies and Curie temperature of bulk Fe and Ni
- results: 2D systems
- results: dilute magnetic semiconductors ($Ga_{1-x}Mn_xAs$)
- outlook

papers: M. Pajda *et al*., PRL 85, 5424 (2000)

M. Pajda *et al*.., PRB **64**, 174402 (2001)

P.B., cond-mat/0207592

L. Sandratskii and P.B., PRB (in press), cond-mat/0207050

(p)reprints: → www.mpi-halle.de

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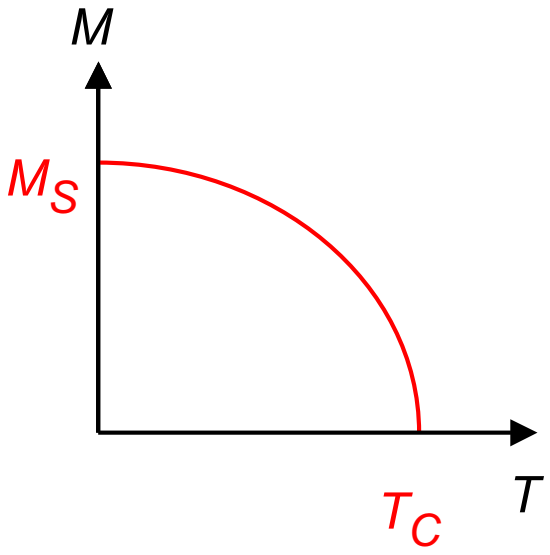
Motivations

? thermodynamics of itinerant ferromagnets ?

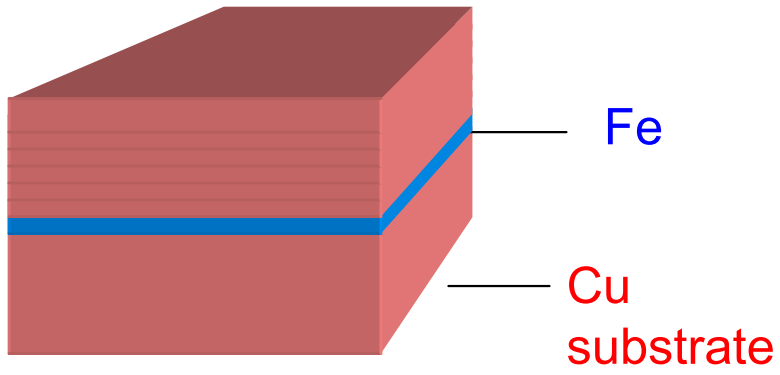
(temperature dependence of M_s , Curie temperature, ...)



- bulk transition metals
- low dimensional systems (ultrathin films, surfaces, ...)
- nanostructures (nanowires, clusters, ...)

influence of a metallic substrate

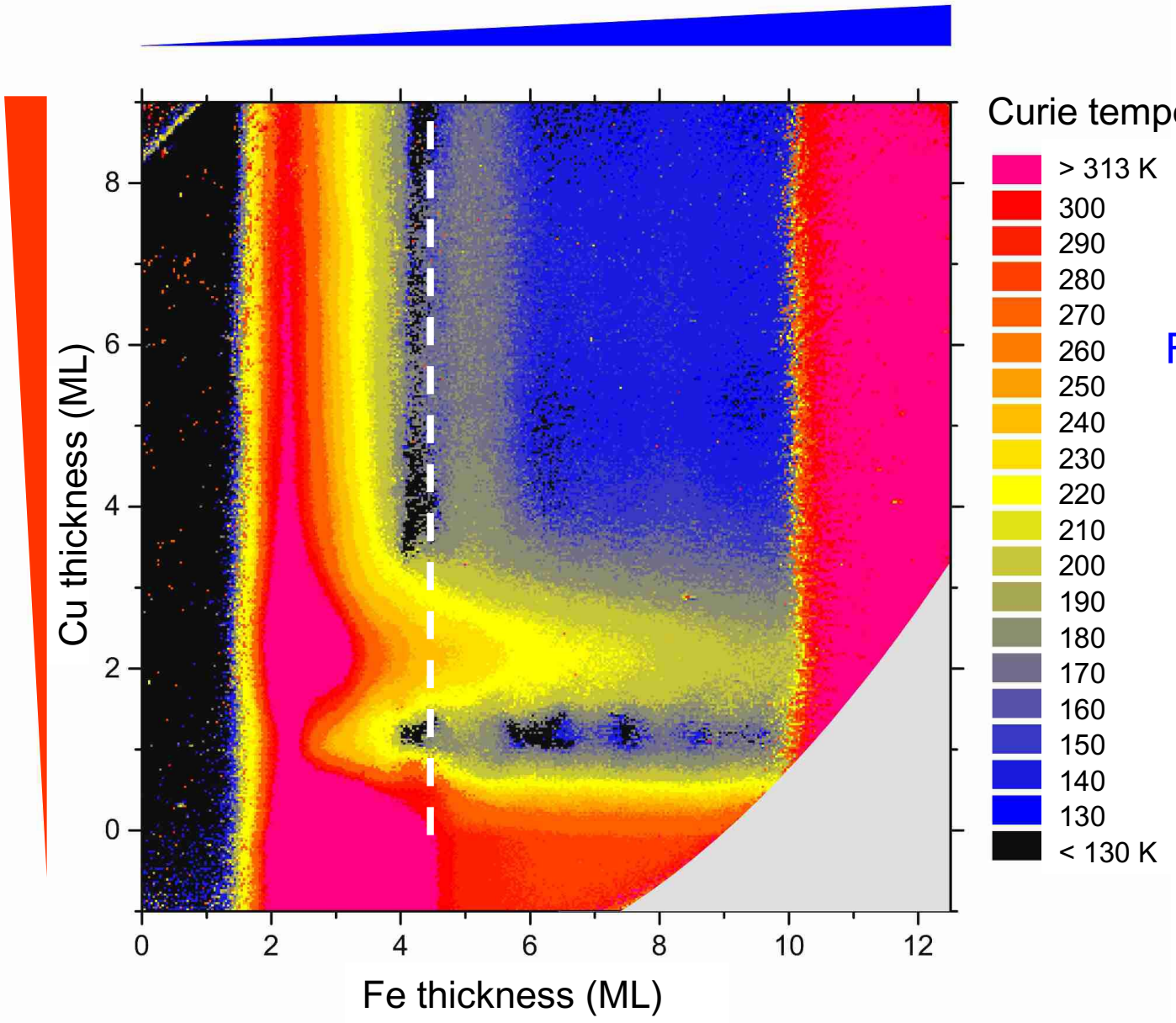
and of a capping layer

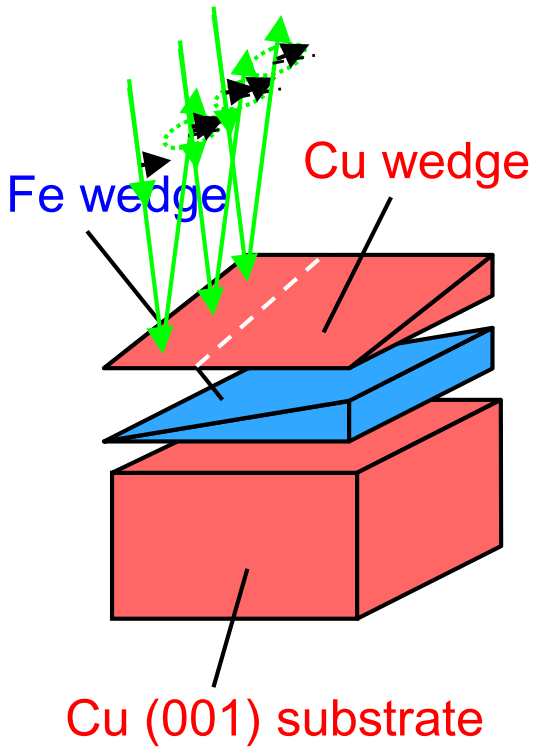


Experimental background

influence of a Cu coverage on the Curie temperature of an Fe film

R. Vollmer *et al.*, PRB **61**, 1303 (2000)

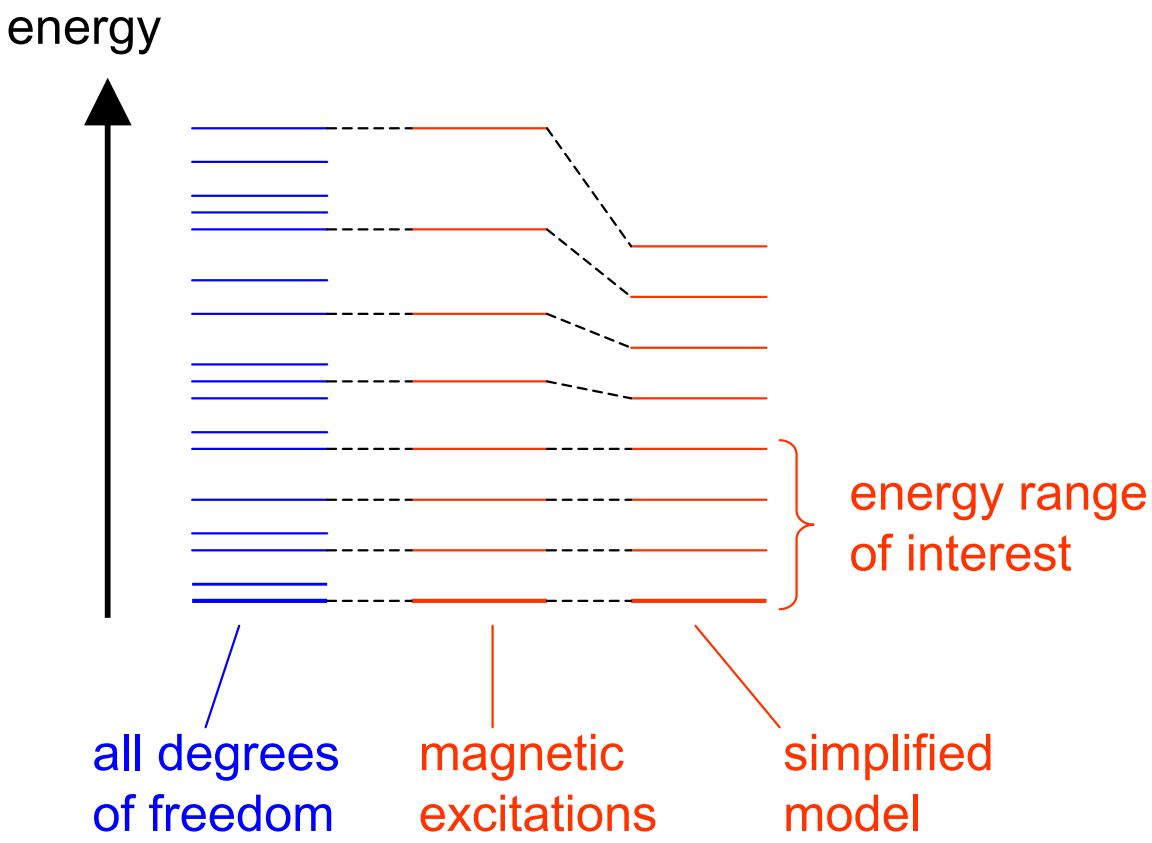




thermodynamics of magnetic systems

partition function: $Z = \sum_n \exp\left(-\varepsilon_n / k_B T\right)$

$$M = k_B T \frac{1}{Z} \frac{\partial Z}{\partial B}$$



Important constraints

Theory

- Nernst theorem: $S = 0$ at $T = 0$ $(\forall B)$ (for quantum systems)

$$\Rightarrow \left.\frac{\partial M}{\partial T}\right|_{T=0,B=0} = -\left.\frac{\partial^2 F}{\partial T\,\partial B}\right|_{T=0,B=0} = -\left.\frac{\partial^2 F}{\partial B\,\partial T}\right|_{T=0,B=0} = \left.\frac{\partial S}{\partial B}\right|_{T=0,B=0} = 0$$

- Goldstone theorem: $\lim_{q\to 0} \hbar\omega(\mathbf{q}) = 0$

- Mermin-Wagner theorem:

  no LRO at $T > 0$ for short-range, isotropic interactions, in 1D- or 2D-systems

  generalization for long-range oscillatory interactions (e.g., RKKY):
  (P.B., PRL **87**, 137203 (2001))

Experiment

- low temperature limit: $\Delta M(T) \sim T^{3/2}$ (Bloch law)
- Curie temperature, critical behavior (??)
- high-temperature limit: Curie-Weiss law (local moment)

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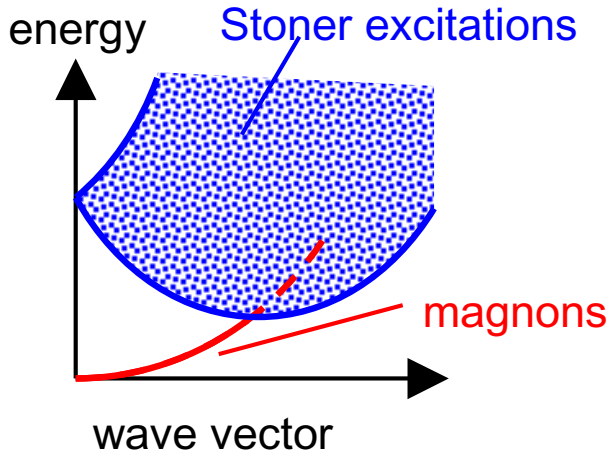
Relevant magnetic excitations

Stoner excitations

magnons !!!



→ **two-step approach**: 1. mapping onto a (classical) effective Heisenberg model

$$H_{\text{eff}} = - \sum_{i \neq j} J_{ij} \ \mathbf{u}_i \cdot \mathbf{u}_j$$

2. statistical physics calculation → $M_s(T)$, T_C

exchange interactions: J_{ij}

- essential parameter for understanding the Curie temperature
- metals → complicated oscillatory RKKY behavior

→ need of performing *ab initio* calculations

Calculation of thermodynamic properties

mean-field approximation: $k_B T_C^{MFA} = \frac{2}{3} \sum_{j \neq 0} J_{0j}$

- poor description of the low-temperature regime (no collective excitations)
- violation of the Mermin-Wagner theorem (2D)
- T_C is overestimated

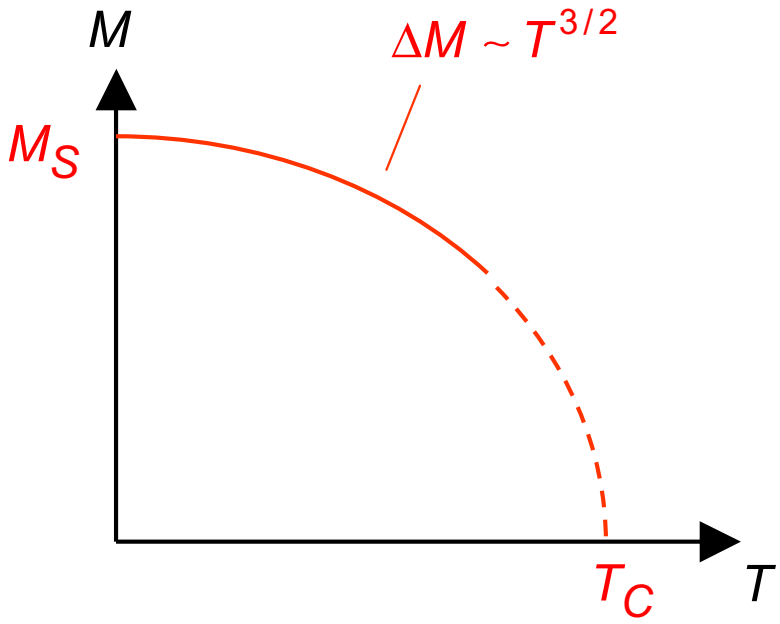
Green´s function method
+ random phase approximation (RPA, Tyablikov):

$$\frac{1}{k_B T_C^{RPA}} = \frac{6}{M_S} \frac{1}{N} \sum_{\mathbf{q}} \frac{1}{\hbar \omega(\mathbf{q})}$$

- spin-wave regime correctly described ($T << T_C$)
- reasonable extrapolation to T_C
- Mermin-Wagner theorem well satisfied

  ($\longrightarrow$ correct behavior for 2D systems)
- incorrect critical exponents

  (= spherical model)



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Alternative approaches

- disordered local moments (DLM) (Gyorffy, Staunton, 1985, 1992)
- coupled spin-fluctuations (Kübler, 1996)
- DMFT (Lichtenstein, Katsnelson, Kotliar, 2001)

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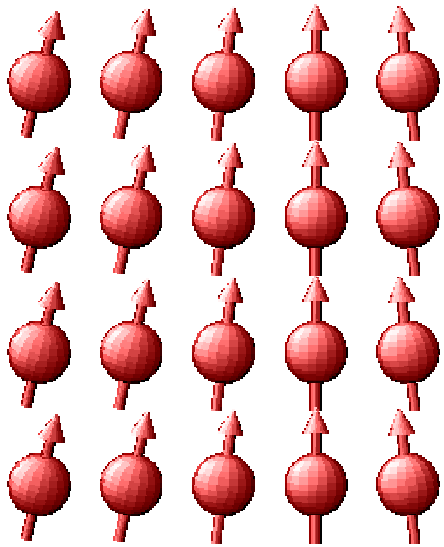
Adiabatic approximation

- neglect of the precession motion

  of the atomic magnetic moments

  → calculation of the energy of

  static spin-waves



adiabatic approximation valid if

spin-wave precession time >>

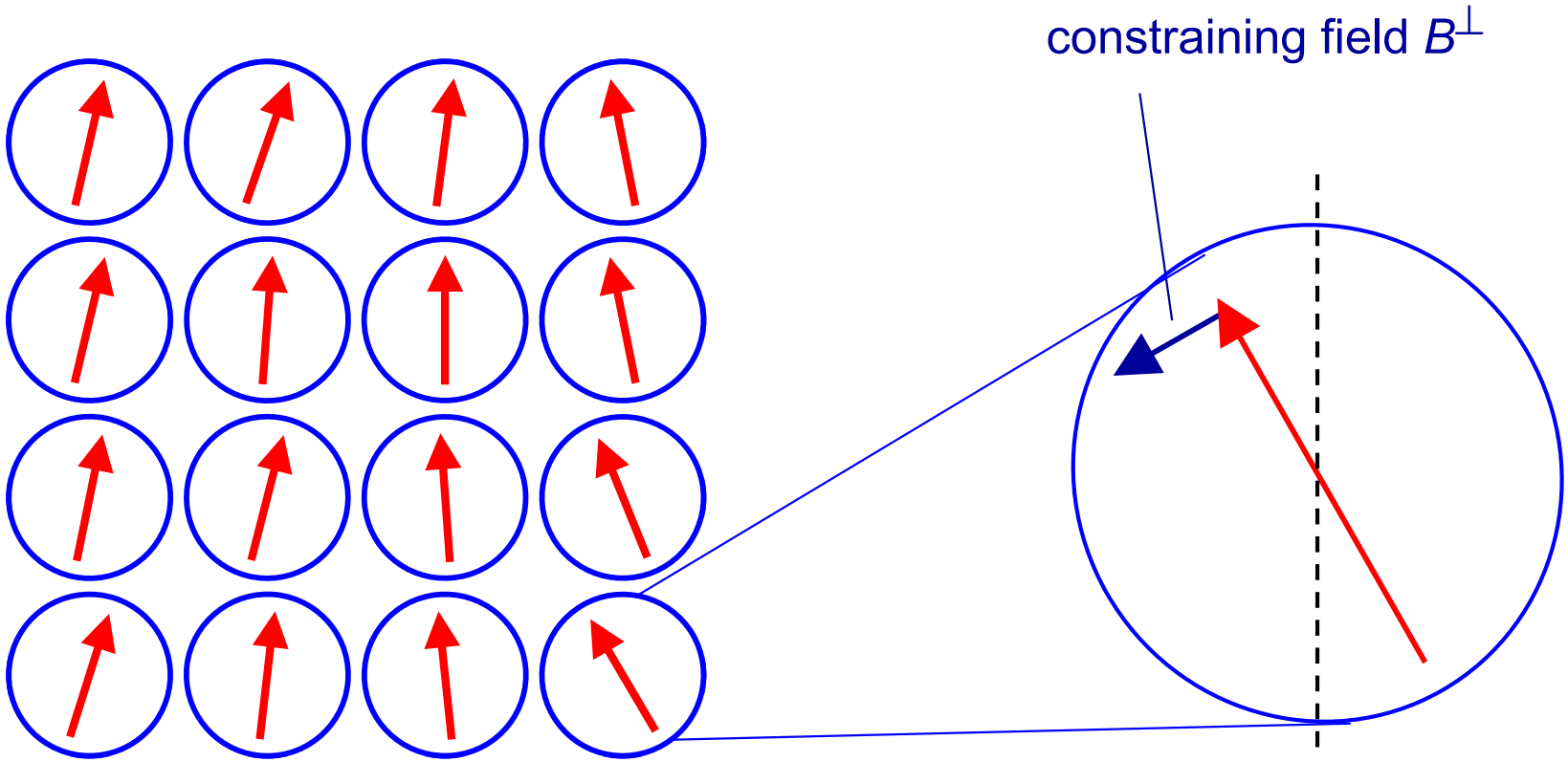
- spin-precession time in exchange field
- interatomic hopping time

i.e., if $\hbar\omega(\mathbf{q}) << \Delta,\ W$

Δ: exchange splitting

W: bandwidth

Constrained density functional theory



Hohenberg-Kohn energy functional: $\mathcal{E}_{\mathrm{HK}}[\rho] = \mathcal{T}_0[\rho] + \mathcal{E}_{\mathrm{ext}}[\rho] + \mathcal{E}_{\mathrm{H}}[\rho] + \mathcal{E}_{\mathrm{xc}}[\rho]$

constrained energy functional: $\mathcal{F}_{\mathrm{HK}}\left[\rho, \mathbf{B}^{\perp}\right] = \mathcal{E}_{\mathrm{HK}}[\rho] + \mathcal{E}_{\mathrm{cons}}\left[\rho, \mathbf{B}^{\perp}\right]$

$$\mathcal{E}_{\mathrm{cons}}\left[\rho, \mathbf{B}^{\perp}\right] \equiv -\sum_{\mathbf{R}} \mathbf{B}_{\mathbf{R}}^{\perp} \cdot \int_{\Omega_{\mathbf{R}}} \mathrm{d}\mathbf{r}\, \mathbf{m}(\mathbf{r})$$

Lagrange parameters

(Dederichs *et al.*, 1984)

Magnetic force theorem

(Liechtenstein *et al.*, 1987)

- idea: try to use the variational properties of the HK functional to avoid doing self-consistent total energy calculations
- problem: we cannot calculate $\mathcal{T}_0[\rho]$ for a charge- and spin-density $\rho \equiv (n, \mathbf{m})$ chosen *a priori*

  (requires the knowledge of the input potential $w_{\text{eff}} = V_{\text{eff}}\sigma_0 - \mathbf{B}_{\text{eff}} \cdot \boldsymbol{\sigma}$ which gives $\rho \equiv (n, \mathbf{m})$ as output)
- solution: use instead the Harris functional (Harris, 1985):

$$\mathcal{F}_{\text{Harris}}\left[\rho, \mathbf{B}^{\perp}\right] = \mathcal{T}'\left[\rho, \mathbf{B}^{\perp}\right] + \mathcal{E}_{\text{ext}}[\rho] + \mathcal{E}_{\text{H}}[\rho] + \mathcal{E}_{\text{xc}}[\rho] + \mathcal{E}_{\text{cons}}\left[\rho, \mathbf{B}^{\perp}\right]$$

$$\mathcal{T}'\left[\rho, \mathbf{B}^{\perp}\right] = \sum_{i=1}^{N_{\text{el}}} \varepsilon_{\text{i}}\left(w'_{\text{eff}}\left[\rho, \mathbf{B}^{\perp}\right]\right) - \int \mathrm{d}\mathbf{r}\left(n(\mathbf{r}) V'_{\text{eff}}(\mathbf{r}) - \mathbf{m}(\mathbf{r}) \cdot \mathbf{B}'_{\text{eff}}(\mathbf{r})\right)$$

$$w'_{\text{eff}}[\rho](\mathbf{r}) = V'_{\text{eff}}(\mathbf{r}) - \boldsymbol{\sigma} \cdot \mathbf{B}'_{\text{eff}}(\mathbf{r}) \equiv \frac{\delta\left(\mathcal{E}_{\text{ext}}[\rho] + \mathcal{E}_{\text{H}}[\rho] + \mathcal{E}_{\text{xc}}[\rho] + \mathcal{E}_{\text{cons}}\left[\rho, \mathbf{B}^{\perp}\right]\right)}{\delta\rho(\mathbf{r})}$$

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properties of the Harris functional:

- $\mathcal{F}_{\text{Harris}}\left[\rho^*, \mathbf{B}^{\perp *}\right] = \mathcal{F}_{\text{HK}}\left[\rho^*, \mathbf{B}^{\perp *}\right]$ for the values $\left(\rho^*, \mathbf{B}^{\perp *}\right)$ which minimize the HK functional

- the Harris functional is *stationary* (but necessarily *minimal*) with respect to ρ and $\mathbf{B}^{\perp}$ near the self-consistent solution $\left(\rho^*, \mathbf{B}^{\perp *}\right)$:

  $$\mathcal{F}_{\text{Harris}}\left[\rho, \mathbf{B}^{\perp}\right] = \mathcal{F}_{\text{HK}}\left[\rho^*, \mathbf{B}^{\perp *}\right] + \mathcal{O}_2\left(\delta\rho, \delta\mathbf{B}^{\perp}\right)$$

- the Harris functional has been found to provide a better approximation than the HK functional for the ground state energy

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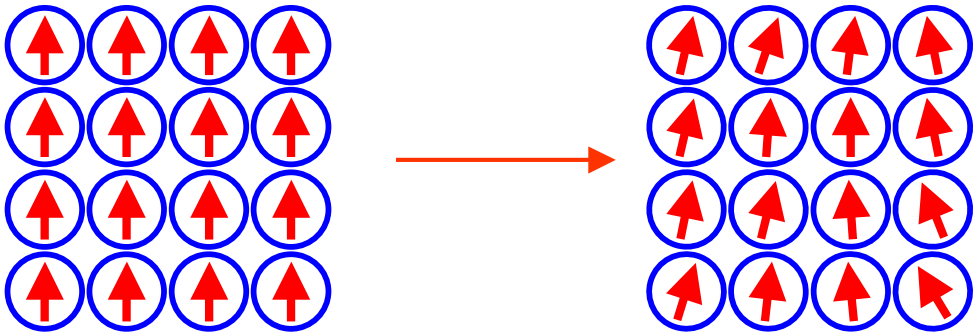
ferromagnetic configuration

$\mathbf{u}_{\mathbf{R}}^{0} = \mathbf{u}^{0} \quad (\forall \mathbf{R})$



constrained rotated configuration (infinitesimal rotation)

$\mathbf{u}_{\mathbf{R}} = \mathbf{u}^{0} + \delta\mathbf{u}_{\mathbf{R}}$

- we look for a second order expansion: $\Delta E(\delta\mathbf{u}) = \sum_{\mathbf{R}\mathbf{R}'} A_{\mathbf{R}\mathbf{R}'} \delta\mathbf{u}_{\mathbf{R}} \cdot \delta\mathbf{u}_{\mathbf{R}'} + \mathcal{O}_2(\delta\mathbf{u})$

with $A_{\mathbf{R}\mathbf{R}'} = -J_{\mathbf{R}\mathbf{R}'} + \delta_{\mathbf{R}\mathbf{R}'} \left(\sum_{\mathbf{R}''} J_{\mathbf{R}\mathbf{R}''} \right)$ and $\sum_{\mathbf{R}} A_{\mathbf{R}\mathbf{R}'} = \sum_{\mathbf{R}'} A_{\mathbf{R}\mathbf{R}'} = 0$

- approximate the total energy energy change by using the Harris functional with trial input $\left(\rho_0, \mathbf{B}^{\perp}\right)$
- within LDA, one obtains:

$$\Delta E(\delta\mathbf{u}) = \sum_{i=1}^{N_{\mathrm{el}}} \left\{ \varepsilon_i \left(w'_{\mathrm{eff}} \left[n_0, m_0(\mathbf{u}_0 + \delta\mathbf{u}), \mathbf{B}^{\perp} \right] \right) - \varepsilon_i \left(w'_{\mathrm{eff}} \left[n_0, m_0\mathbf{u}_0, 0 \right] \right) \right\} + \mathcal{O}_2\left(\delta n, \delta m, \delta\mathbf{B}^{\perp}\right)$$

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choice of Liechtenstein *et al.*:

input values of constraining field $\mathbf{B}^{\perp} = 0$

$$\rightarrow \qquad \Delta E(\delta\mathbf{u}) = \sum_{\mathbf{R}\mathbf{R}'} \tilde{A}_{\mathbf{R}\mathbf{R}'} \delta\mathbf{u}_{\mathbf{R}} \cdot \delta\mathbf{u}_{\mathbf{R}'} + \mathcal{O}_2\left(\delta n, \delta m, \mathbf{B}^{\perp *}\right)$$

with $$\tilde{A}_{\mathbf{R}\mathbf{R}'} = -\tilde{J}_{\mathbf{R}\mathbf{R}'} + \delta_{\mathbf{R}\mathbf{R}'}\left(\sum_{\mathbf{R}''} \tilde{J}_{\mathbf{R}\mathbf{R}''}\right)$$

and $$\tilde{J}_{\mathbf{R}\mathbf{R}'} = \frac{1}{\pi}\int_{-\infty}^{\varepsilon_F} \mathrm{d}\varepsilon\, \mathrm{Im\ Tr}\left(B^{\mathrm{xc}}_{\mathbf{R}} G^{\uparrow}_{\mathbf{R}\mathbf{R}'} B^{\mathrm{xc}}_{\mathbf{R}'} G^{\downarrow}_{\mathbf{R}'\mathbf{R}}\right)$$

estimation of the error:

$$\left.\begin{aligned} \delta n &= \mathcal{O}_2(\delta\mathbf{u}) \\ \delta m &= \mathcal{O}_2(\delta\mathbf{u}) \\ \mathbf{B}^{\perp *} &= \mathcal{O}_1(\delta\mathbf{u}) \end{aligned}\right\} \quad \mathcal{O}_2\left(\delta n, \delta m, \mathbf{B}^{\perp *}\right) = \mathcal{O}_2(\delta\mathbf{u})$$

therefore $\tilde{J}_{\mathbf{R}\mathbf{R}'} \neq J_{\mathbf{R}\mathbf{R}'}$

bare exchange parameters — *true* exchange parameters

improved choice:

input values of constraining field $\mathbf{B}^{\perp *}$ (= exact values)

$$\rightarrow \quad \Delta E(\delta\mathbf{u}) = \sum_{\mathbf{R}\mathbf{R}'} \Big[\tilde{A}_{\mathbf{R}\mathbf{R}'} \delta\mathbf{u}_{\mathbf{R}} \cdot \delta\mathbf{u}_{\mathbf{R}'} + \left(M_{\mathbf{R}} \delta_{\mathbf{R}\mathbf{R}'} - \tilde{K}_{\mathbf{R}\mathbf{R}'} \right) \mathbf{B}_{\mathbf{R}}^{\perp *} \cdot \delta\mathbf{u}_{\mathbf{R}'} - \tilde{\chi}_{\mathbf{R}\mathbf{R}'} \mathbf{B}_{\mathbf{R}}^{\perp *} \cdot \mathbf{B}_{\mathbf{R}'}^{\perp *} \Big] + \mathcal{O}_4(\delta\mathbf{u})$$

with $$\tilde{K}_{\mathbf{R}\mathbf{R}'} = \frac{1}{\pi} \int_{-\infty}^{\varepsilon_F} \mathrm{d}\varepsilon \, \mathrm{Im} \, \mathrm{Tr} \left(G^{\uparrow}_{\mathbf{R}\mathbf{R}'} B^{\mathrm{xc}}_{\mathbf{R}'} G^{\downarrow}_{\mathbf{R}'\mathbf{R}} + G^{\downarrow}_{\mathbf{R}\mathbf{R}'} B^{\mathrm{xc}}_{\mathbf{R}'} G^{\uparrow}_{\mathbf{R}'\mathbf{R}} \right)$$

and $$\tilde{\chi}_{\mathbf{R}\mathbf{R}'} = \frac{2}{\pi} \int_{-\infty}^{\varepsilon_F} \mathrm{d}\varepsilon \, \mathrm{Im} \, \mathrm{Tr} \left(G^{\uparrow}_{\mathbf{R}\mathbf{R}'} G^{\downarrow}_{\mathbf{R}'\mathbf{R}} \right)$$

calculation of the constraining fields: $$M_{\mathbf{R}} \delta\mathbf{u}_{\mathbf{R}} = \sum_{\mathbf{R}'} \left(\tilde{K}_{\mathbf{R}\mathbf{R}'} \delta\mathbf{u}_{\mathbf{R}'} + \tilde{\chi}_{\mathbf{R}\mathbf{R}'} \mathbf{B}_{\mathbf{R}'}^{\perp *} \right)$$

$$\rightarrow \quad \mathbf{B}_{\mathbf{R}}^{\perp *} = \sum_{\mathbf{R}'} \left[\tilde{\mathrm{X}}^{-1} \left(\mathrm{M} - \tilde{\mathrm{K}} \right) \right]_{\mathbf{R}\mathbf{R}'} \delta\mathbf{u}_{\mathbf{R}'} \qquad \text{with} \qquad M_{\mathbf{R}} = \sum_{\mathbf{R}'} \tilde{K}_{\mathbf{R}\mathbf{R}'}$$

final result: (using some matrix notations)

$$\mathrm{A} = \tilde{\mathrm{A}} + \frac{1}{2} \left(\mathrm{M} - \tilde{\mathrm{K}}^{\mathrm{T}} \right) \tilde{\mathrm{X}}^{-1} \left(\mathrm{M} - \tilde{\mathrm{K}} \right)$$

***renormalized* magnetic force theorem**

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Physical interpretation

define: $\Delta_{\mathbf{R}} \equiv \frac{4}{M_{\mathbf{R}}} \sum_{\mathbf{R}'} \tilde{J}_{\mathbf{R}\mathbf{R}'} = \frac{2}{M_{\mathbf{R}}} \int_{\Omega_{\mathbf{R}}} d\mathbf{r}\, B_{xc}(\mathbf{r}) m(\mathbf{r})$ $\approx$ exchange splitting on site **R**

simple approximation: $\tilde{K}_{\mathbf{R}\mathbf{R}'} \approx \frac{4\tilde{J}_{\mathbf{R}\mathbf{R}'}}{\Delta_{\mathbf{R}}}$ and $\tilde{\chi}_{\mathbf{R}\mathbf{R}'} \approx \frac{8\tilde{J}_{\mathbf{R}\mathbf{R}'}}{\Delta_{\mathbf{R}}\Delta_{\mathbf{R}'}}$ $\rightarrow$ $\tilde{A} = \frac{1}{2} M I_{xc} (1 - \tilde{\chi} I_{xc}) M$

$\rightarrow$

$$A = \tilde{A}\left(1 - 4M^{-1}\ {}^{-1}\tilde{A}\right)^{-1} = \left(\frac{M}{4} - \tilde{J}\right)\left(4M^{-1}\ {}^{-1}\tilde{J}\right)^{-1}$$

$$= \frac{1}{2} M I_{xc} (1 - \tilde{\chi} I_{xc}) (\tilde{\chi} I_{xc})^{-1} M = \frac{1}{2} M \left(\tilde{\chi}^{-1} - I_{xc}\right) M = \frac{1}{2} M \chi^{-1} M$$

equation of motion: $M_{\mathbf{R}} \frac{d\delta\mathbf{u}_{\mathbf{R}}}{dt} = 2 \frac{\partial \Delta E}{d\delta\mathbf{u}_{\mathbf{R}}} \times \mathbf{u}_0 = 4 \sum_{\mathbf{u}_{\mathbf{R}'}} A_{\mathbf{R}\mathbf{R}'} \delta\mathbf{u}_{\mathbf{R}'} \times \mathbf{u}_0$

spin-wave energies = eigenvalues of $\hbar\Omega \equiv 4M^{-1/2} A M^{-1/2}$

approximation $\rightarrow$ $\hbar\Omega = \hbar\tilde{\Omega}\left(1 - {}^{-1}\hbar\tilde{\Omega}\right)^{-1}$

for systems with one atom per cell $\rightarrow$ $\hbar\omega(\mathbf{q}) = \frac{\hbar\tilde{\omega}(\mathbf{q})}{1 - \hbar\tilde{\omega}(\mathbf{q})/\Delta}$

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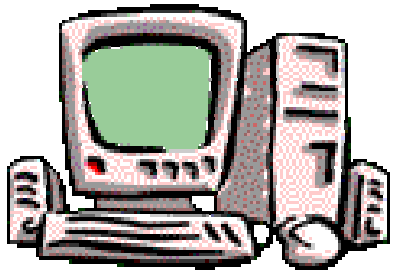
Technical details about calculations



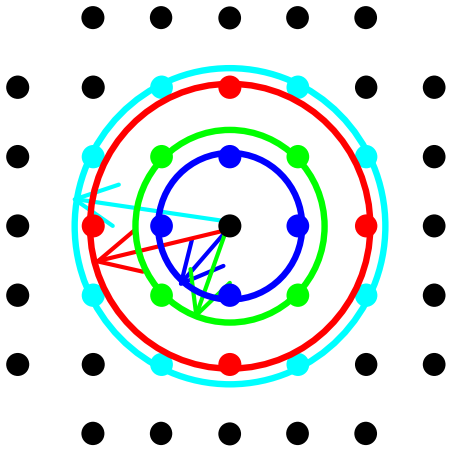
- density functional theory (local density approximation)
- tight-binding LMTO method
- utilisation of the ``magnetic force theorem´´

$$J_{i,j} \propto \mathrm{Im} \int \mathrm{d}\varepsilon\, f(\varepsilon)\, \mathrm{Tr}\left[\left(V_i^{\uparrow} - V_i^{\downarrow}\right) G_{ij}^{\uparrow}(\varepsilon + \mathrm{i}0^{+}) \left(V_j^{\uparrow} - V_j^{\downarrow}\right) G_{ji}^{\downarrow}(\varepsilon + \mathrm{i}0^{+})\right]$$

- integration over energies complex in the complex plane
- integration over the Brillouin zone using up to 5 x10^6 **k**-points
- calculation of $J_{i,j}$ for 172 (fcc),

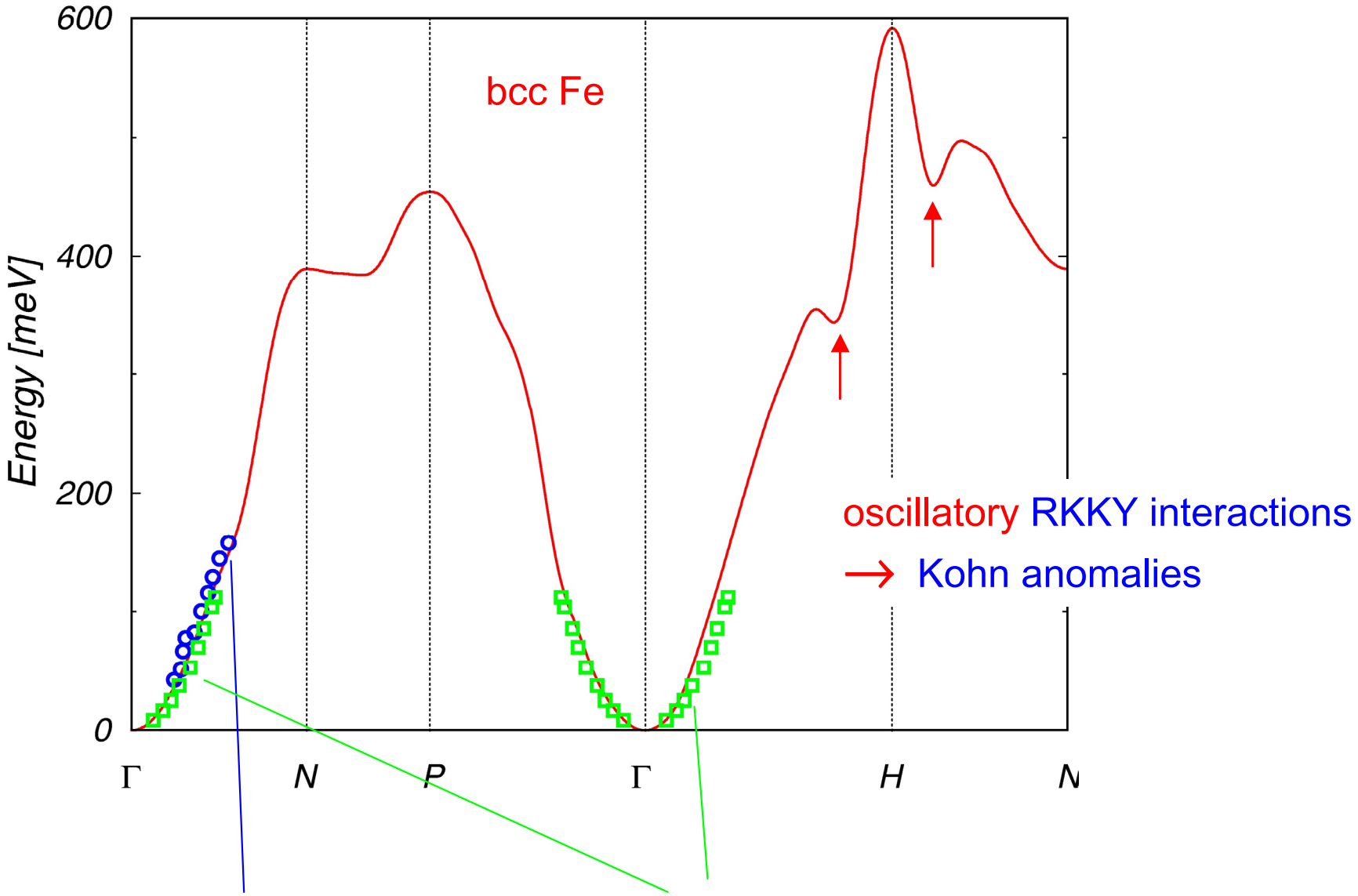
  195 (bcc) or 101 (2D) shells of neighbors

  (~ 7 to 10 x *a*) (*a* = lattice parameter)



Spin-wave dispersion of bulk bcc Fe

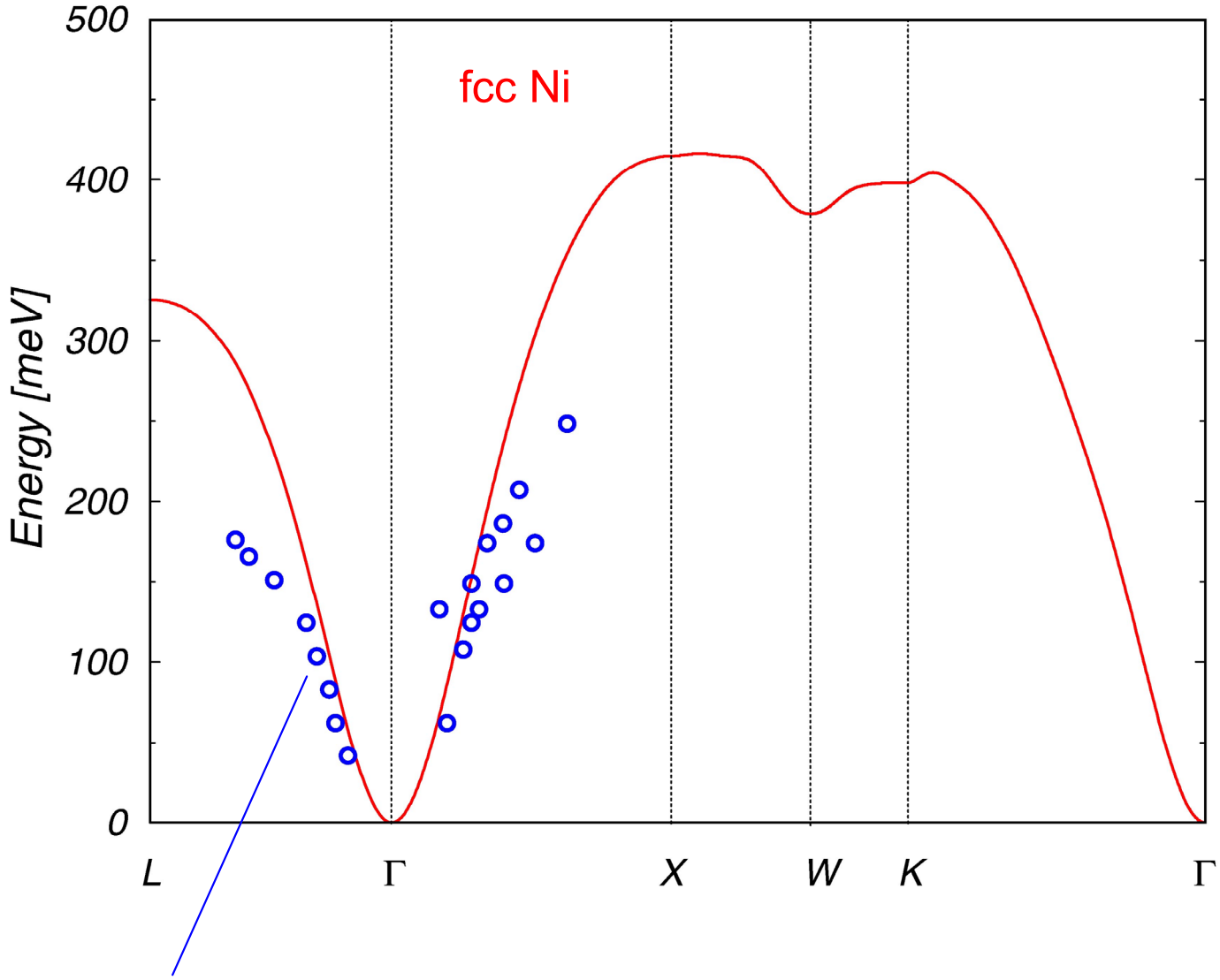
$$E(\mathbf{q}) = \frac{4\mu_B}{m_s} \sum_{j \neq 0} J_{0,j}\left(1 - \exp(i\mathbf{q}\cdot\mathbf{R}_{0,j})\right)$$



C.K. Loong *et al.*, JAP **55**, 1895 (1984)

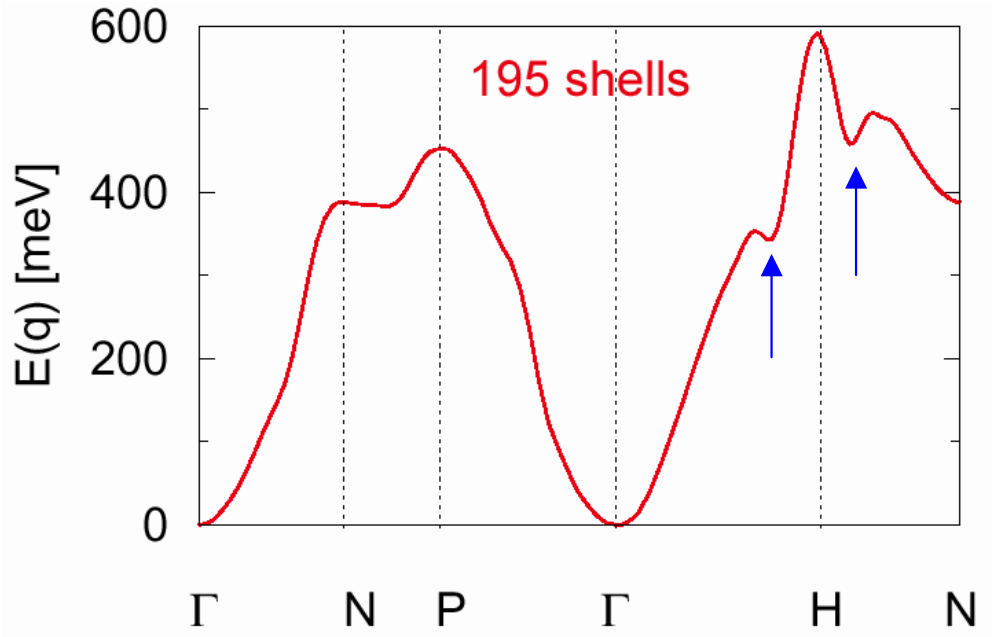
J.W. Lynn, PRB **11**, 2624 (1983)

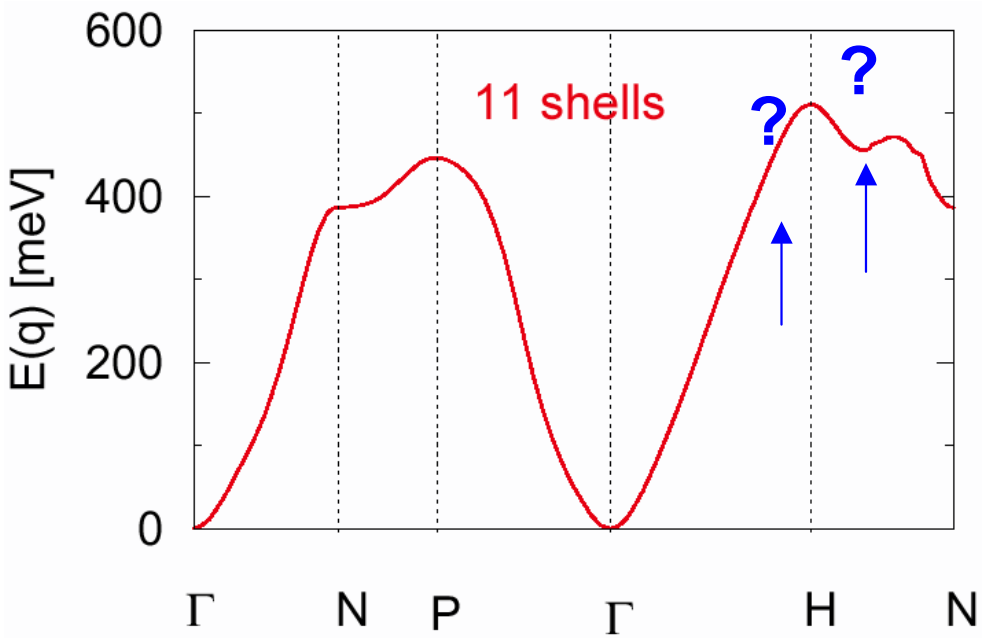
Spin-wave dispersion of bulk fcc Ni

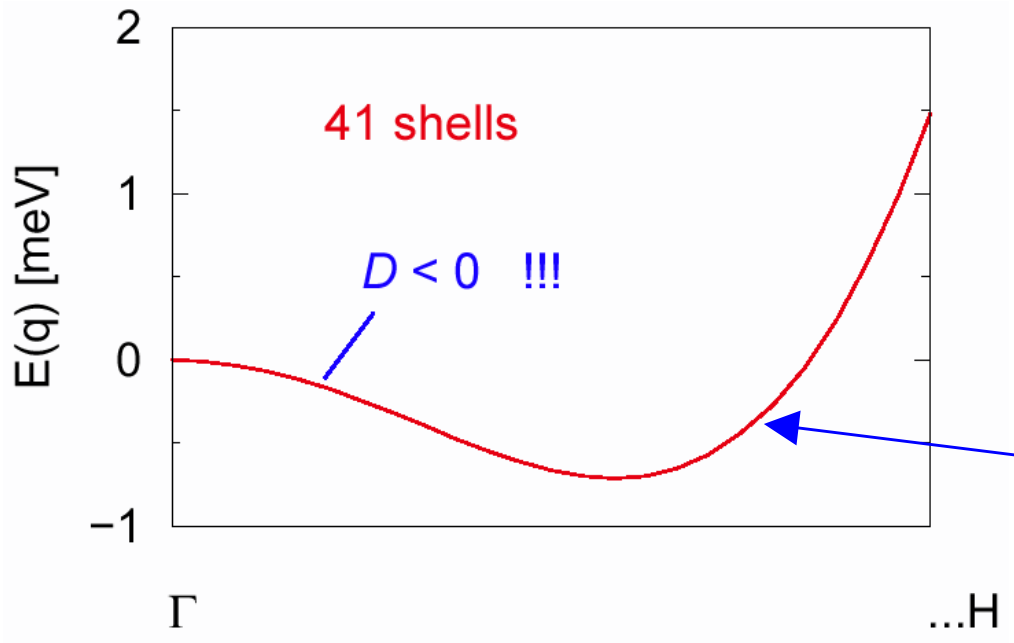


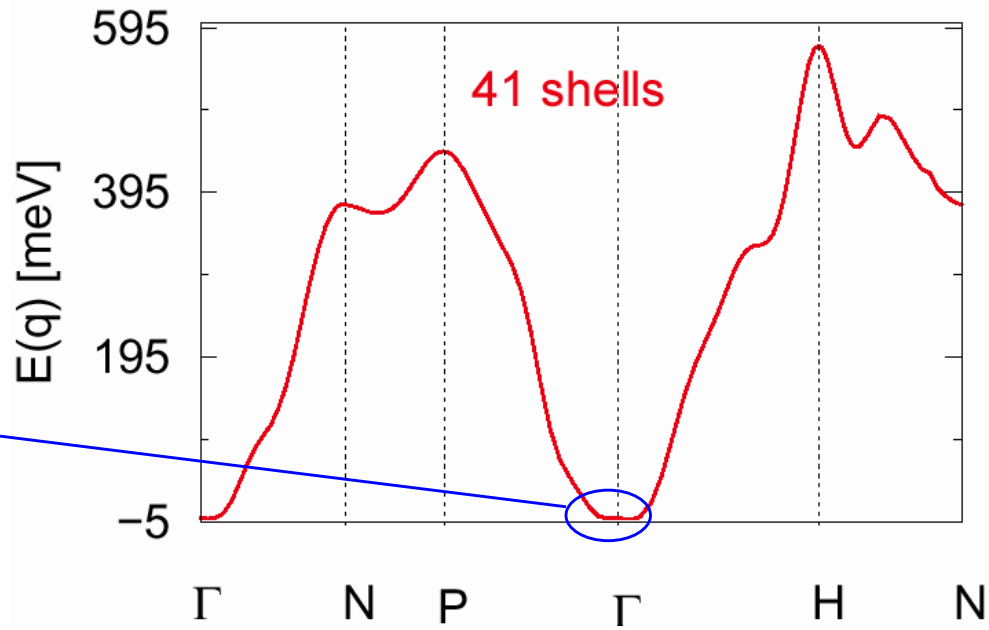
H.A. Mook and D.McK. Paul, PRL **54**, 227 (1985)

Convergence with number of shells (bcc Fe)









Calculation of the spin-wave stiffness constant

$$E(\mathbf{q}) = \frac{4\mu_B}{m_S} \sum_{j\neq 0} J_{0,j}\left(1-\exp(i\mathbf{q}\cdot\mathbf{R}_{0,j})\right) \quad \rightarrow \text{ for small } q \qquad E(\mathbf{q}) \approx D\, q^2$$

$$\text{with} \qquad D = \frac{2\mu_B}{3m_S} \sum_{R_{0,j}} J_{0,j}\, {R_{0,j}}^2$$

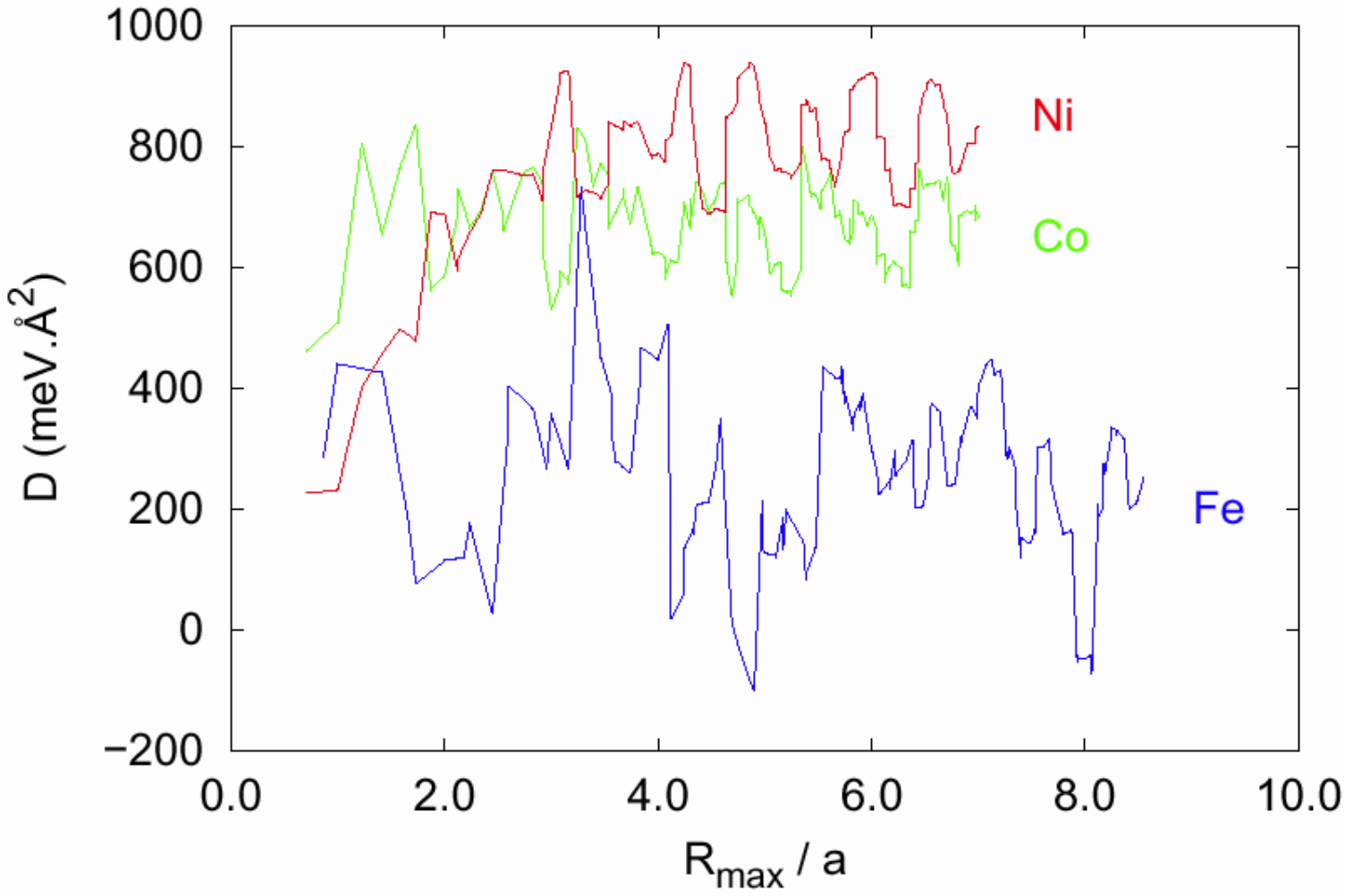
Problem: the calculation of D by using

$$D = \lim_{R_{max}\rightarrow\infty} \frac{2\mu_B}{3m_S} \sum_{0<R_{0,j}<R_{max}} J_{0,j}\, {R_{0,j}}^2$$

is non convergent due to the long-range of the oscillatory RKKY interaction

for a weak ferromagnet: $$J_{0,j} \propto \frac{\sin\left(\left(k_F^{\uparrow} + k_F^{\downarrow}\right)R_{0,j} + \phi\right)}{{R_{0,j}}^3}$$

$$D = \lim_{R_{max}\to\infty} \frac{2\mu_B}{3m_s} \sum_{0<R_{0,j}<R_{max}} J_{0,j}\, R_{0,j}^{\,2}$$



regularization of

$$D = \lim_{R_{\max}\to\infty} \frac{2\mu_B}{3m_S} \sum_{0<R_{0,j}<R_{\max}} J_{0,j}\, {R_{0,j}}^2$$

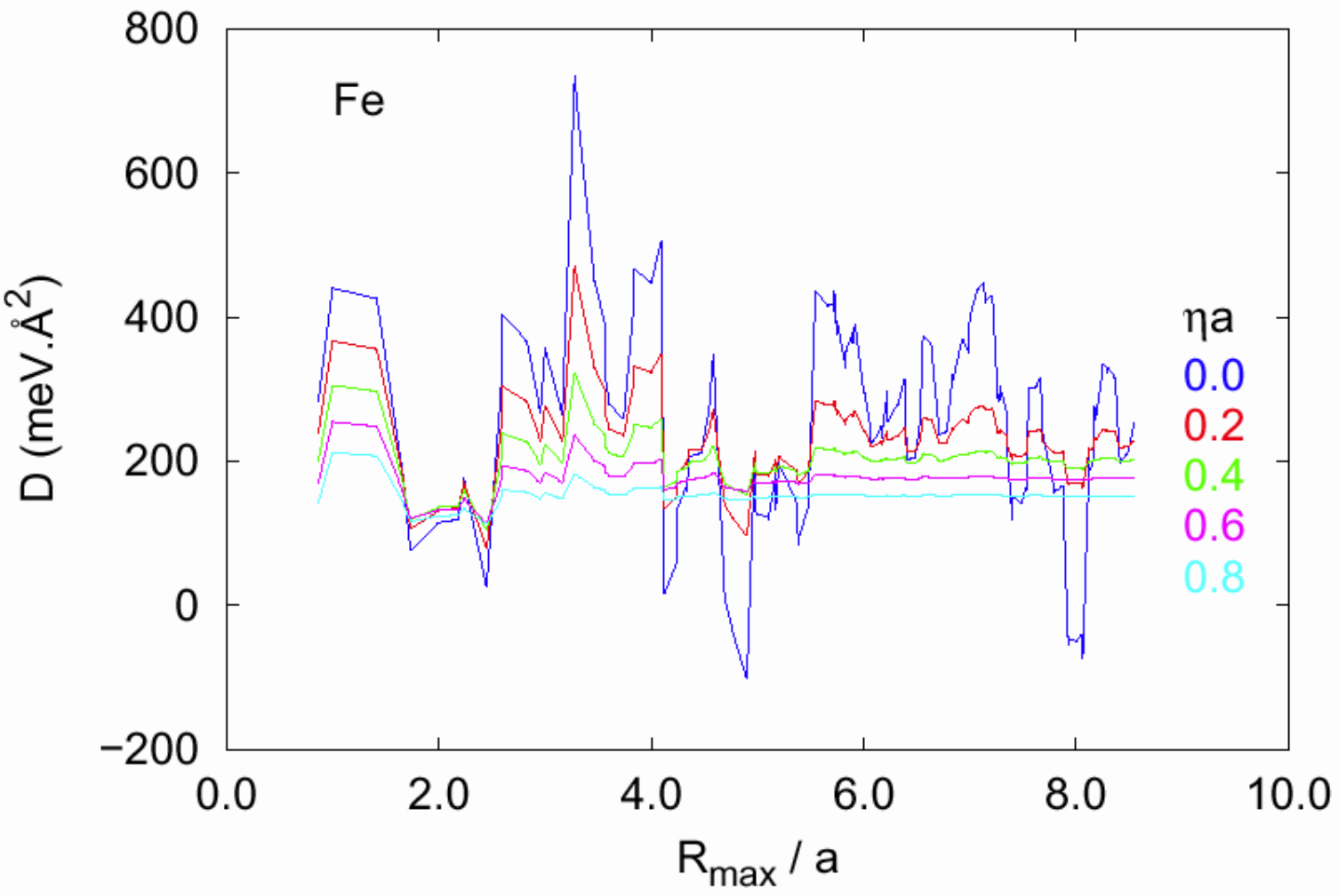
by substituing with the formally equivalent expression

$$D = \lim_{\eta\to 0} \lim_{R_{\max}\to\infty} \frac{2\mu_B}{3m_S} \sum_{0<R_{0,j}<R_{\max}} J_{0,j}\, {R_{0,j}}^2 \exp\left(-\eta R_{0,j}\right)$$

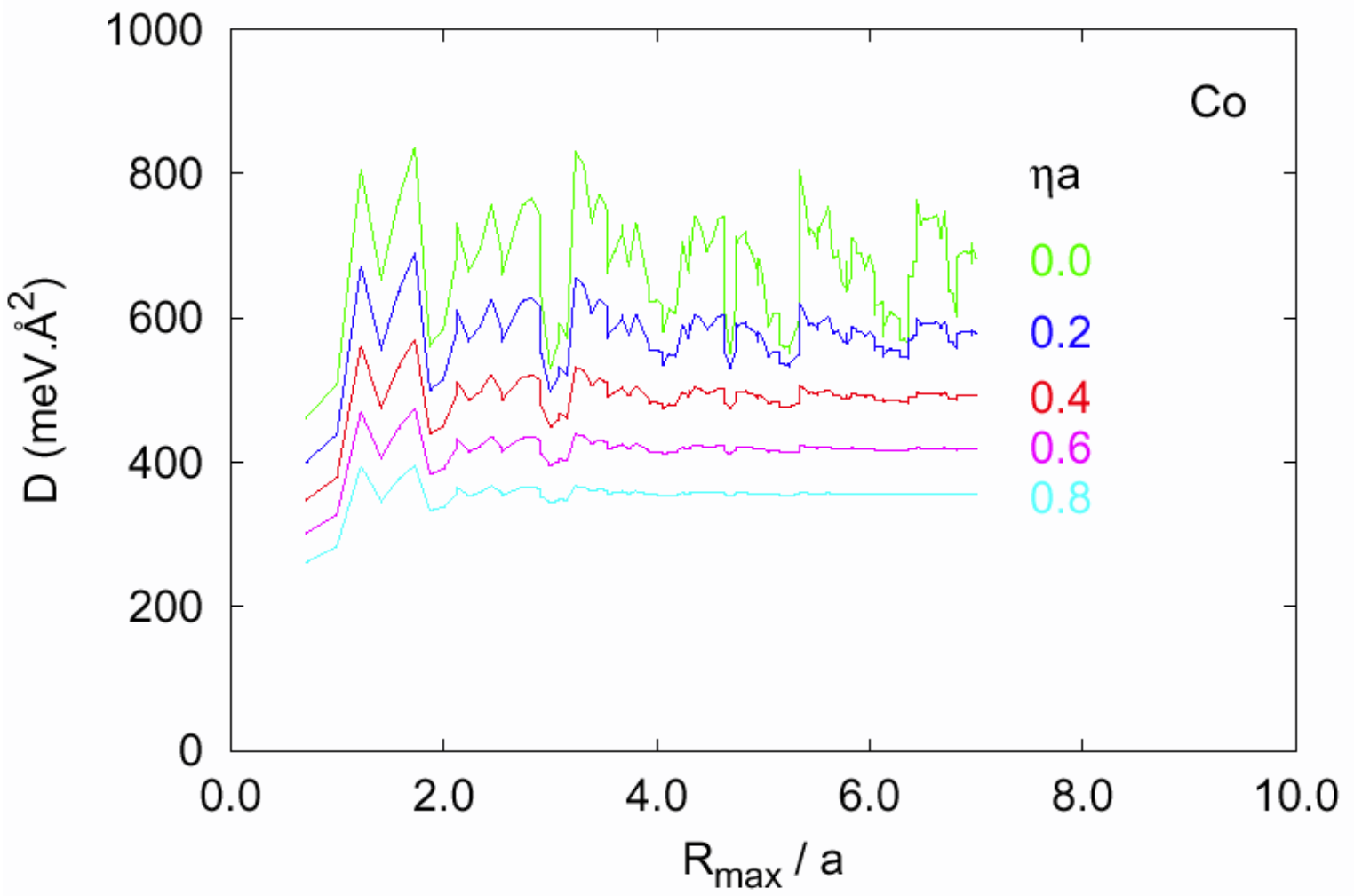
(numerically convergent)

~ Ewald summation technique

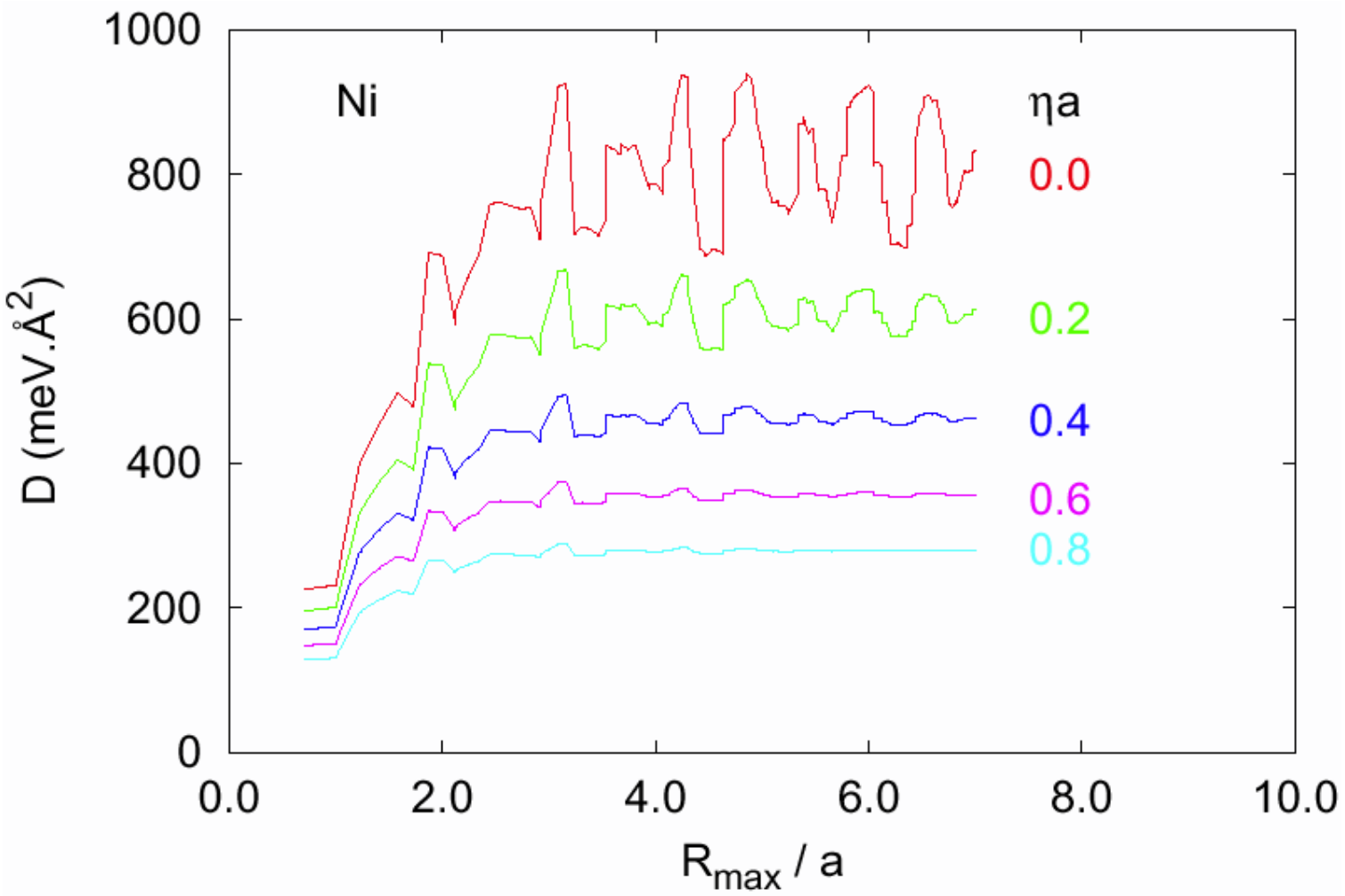
$$D = \lim_{\eta \to 0} \lim_{R_{max} \to \infty} \frac{2\mu_B}{3m_s} \sum_{0<R_{0,j}<R_{max}} J_{0,j} R_{0,j}^2 \exp(-\eta R_{0,j})$$



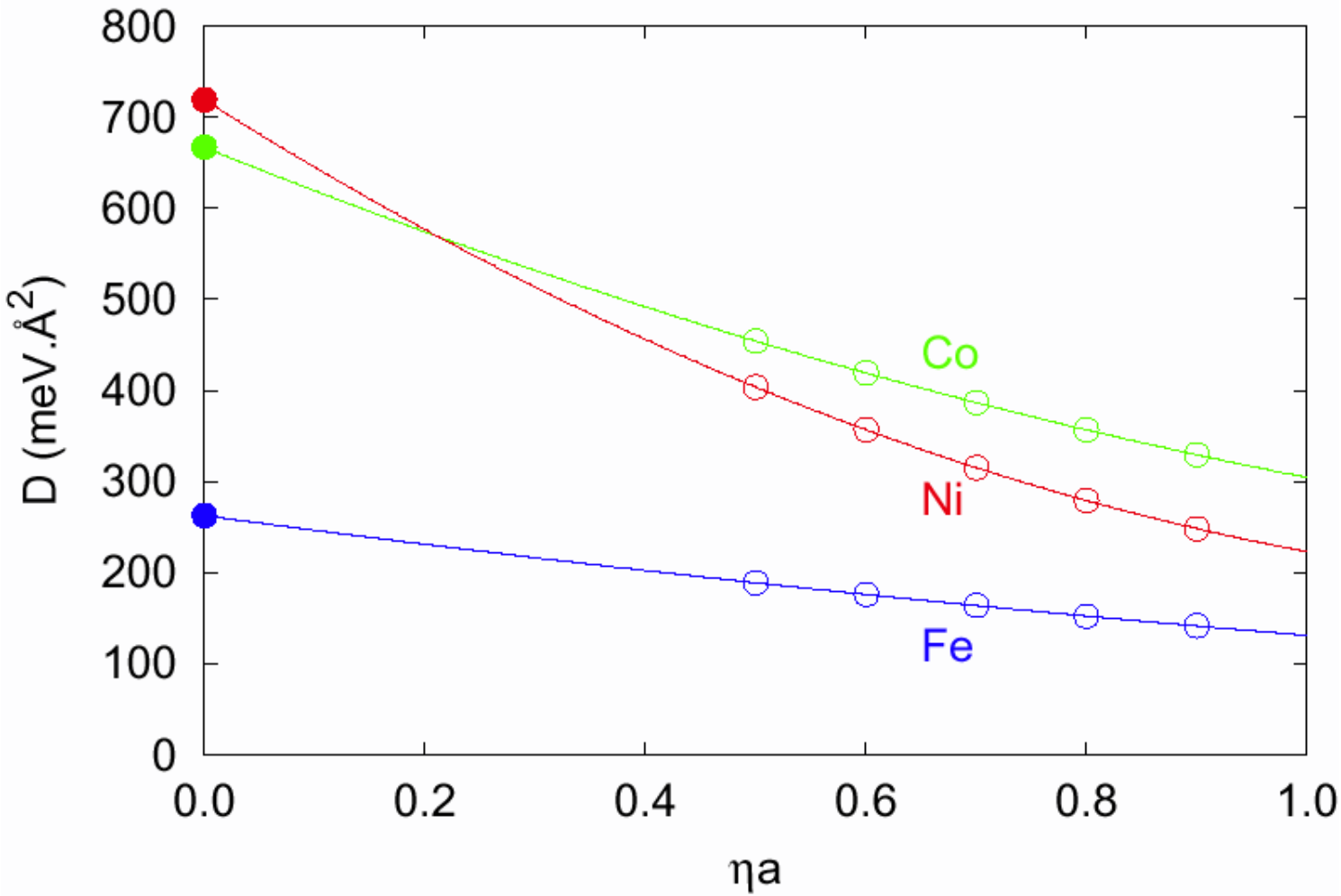
$$D = \lim_{\eta \to 0} \lim_{R_{max} \to \infty} \frac{2\mu_B}{3m_s} \sum_{0<R_{0,j}<R_{max}} J_{0,j} R_{0,j}^2 \exp(-\eta R_{0,j})$$



$$D = \lim_{\eta \to 0} \lim_{R_{max} \to \infty} \frac{2\mu_B}{3m_s} \sum_{0<R_{0,j}<R_{max}} J_{0,j} R_{0,j}^2 \exp(-\eta R_{0,j})$$



$$D = \lim_{\eta \to 0} \lim_{R_{max} \to \infty} \frac{2\mu_B}{3m_s} \sum_{0<R_{0,j}<R_{max}} J_{0,j} R_{0,j}^2 \exp(-\eta R_{0,j})$$



Results for bulk systems

≈ good agreement

metal	D_{th} (meV.Å^2)	D_{exp} (meV.Å^2)	T_C^{MFA} (K)	T_C^{RPA} (K)	T_C^{exp} (K)
Fe (bcc)	250	280, 330	1414	950	1045
Co (fcc)	663	580	1645	1131	1388 - 1398
Ni (fcc)	753	422, 555	397	350	624 - 631

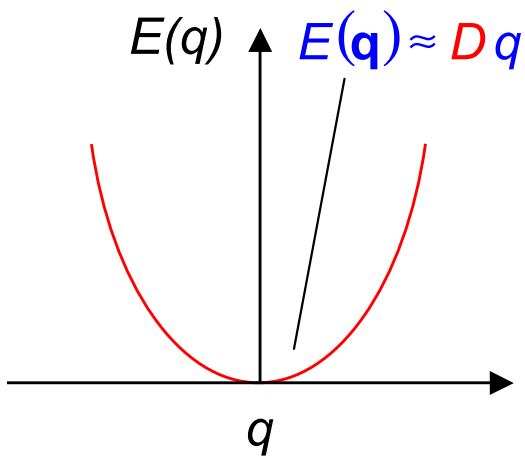
? origin of the strong disagreement for Ni ?

→ importance of Stoner excitations

→ adiabatic approximation rather poor

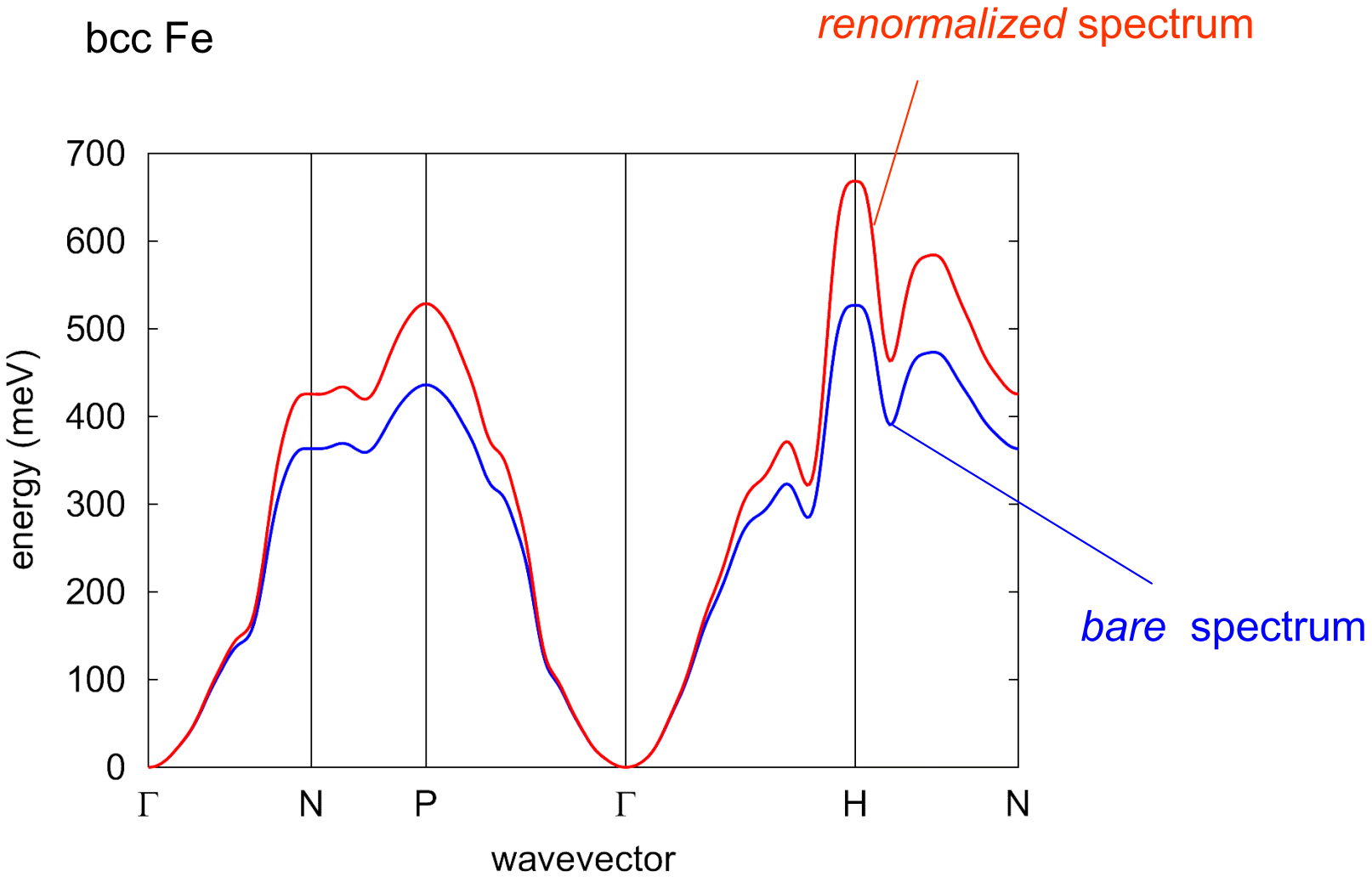
→ failure of the ``*bare* magnetic force theorem´´

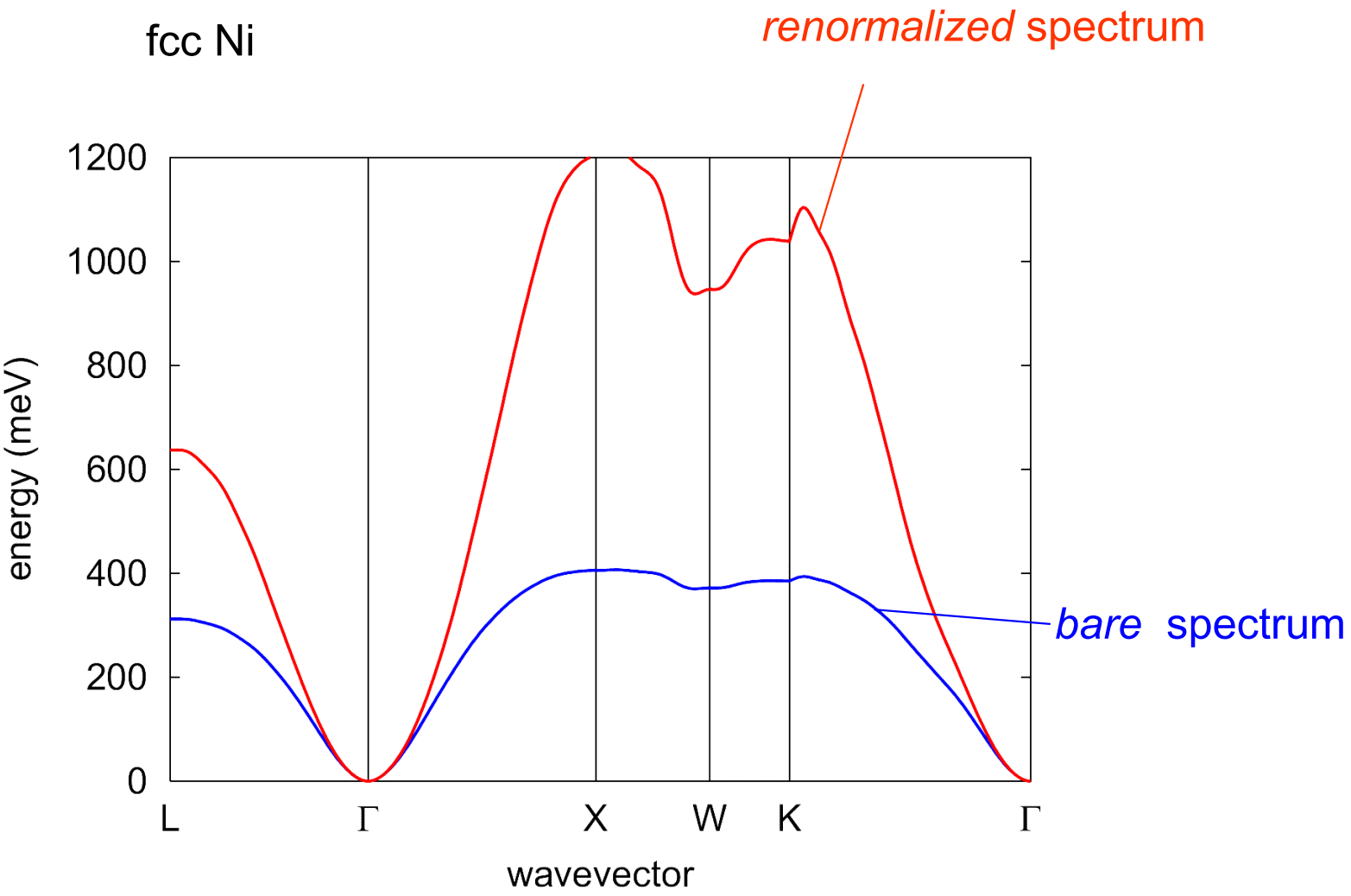
→ correlation effects ?



μΦ

Effect of renormalization of exchange interaction parameters




fcc Ni
renormalized spectrum
bare spectrum
energy (meV)
1200
1000
800
600
400
200
0
L
Γ
X
W
K
Γ
wavevector

Renormalized Curie temperature

Curie temperature obtained from the *renormalized* parameters J_{ij}

Curie temperature obtained from the *bare* parameters $\tilde{J}_{ij}$

metal	T_C^{exp} (K)	$\tilde{T}_C^{RPA}$ (K)	T_C^{RPA} (K)
Fe (bcc)	1045	950	1057
Ni (fcc)	624 - 631	350	634

$$\hbar\omega(\mathbf{q}) = \frac{\hbar\tilde{\omega}(\mathbf{q})}{1-\hbar\tilde{\omega}(\mathbf{q})/\Delta}$$

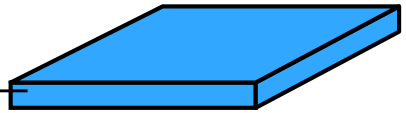
$$\frac{1}{k_B T_C^{RPA}} = \frac{6}{M_S}\,\frac{1}{N}\sum_{\mathbf{q}}\frac{1}{\hbar\omega(\mathbf{q})}$$

$$\rightarrow \quad k_B T_C^{RPA} = k_B \tilde{T}_C^{RPA}\left(1-\frac{6k_B\tilde{T}_C^{RPA}}{M\Delta}\right)^{-1}$$

$\mu\Phi$

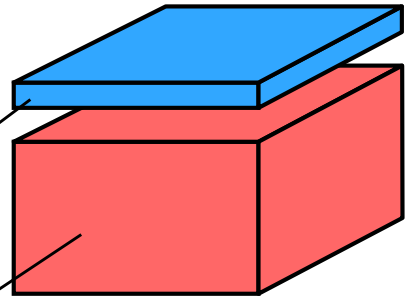
Two-dimensional systems

free standing
Fe or Co monolayer
(theorist's fantasy)



Fe or Co monolayer

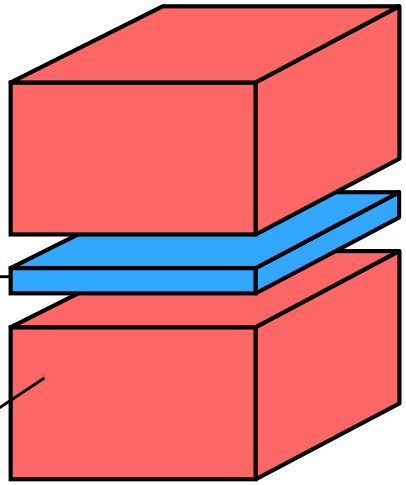
Cu substrate



Cu cap layer

embedded
Fe or Co monolayer

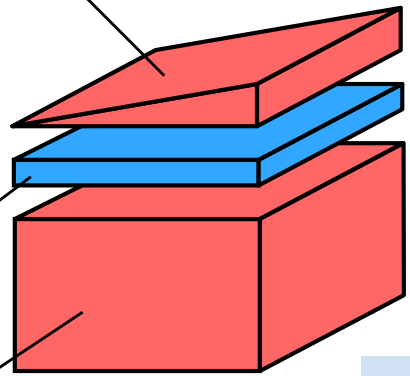
Cu substrate



Cu cap layer of variable thickness

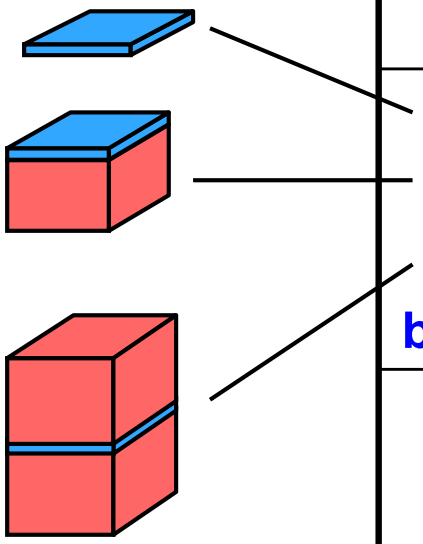
Fe or Co monolayer

Cu substrate



$\mu\Phi$

exchange interactions for an atomic layer



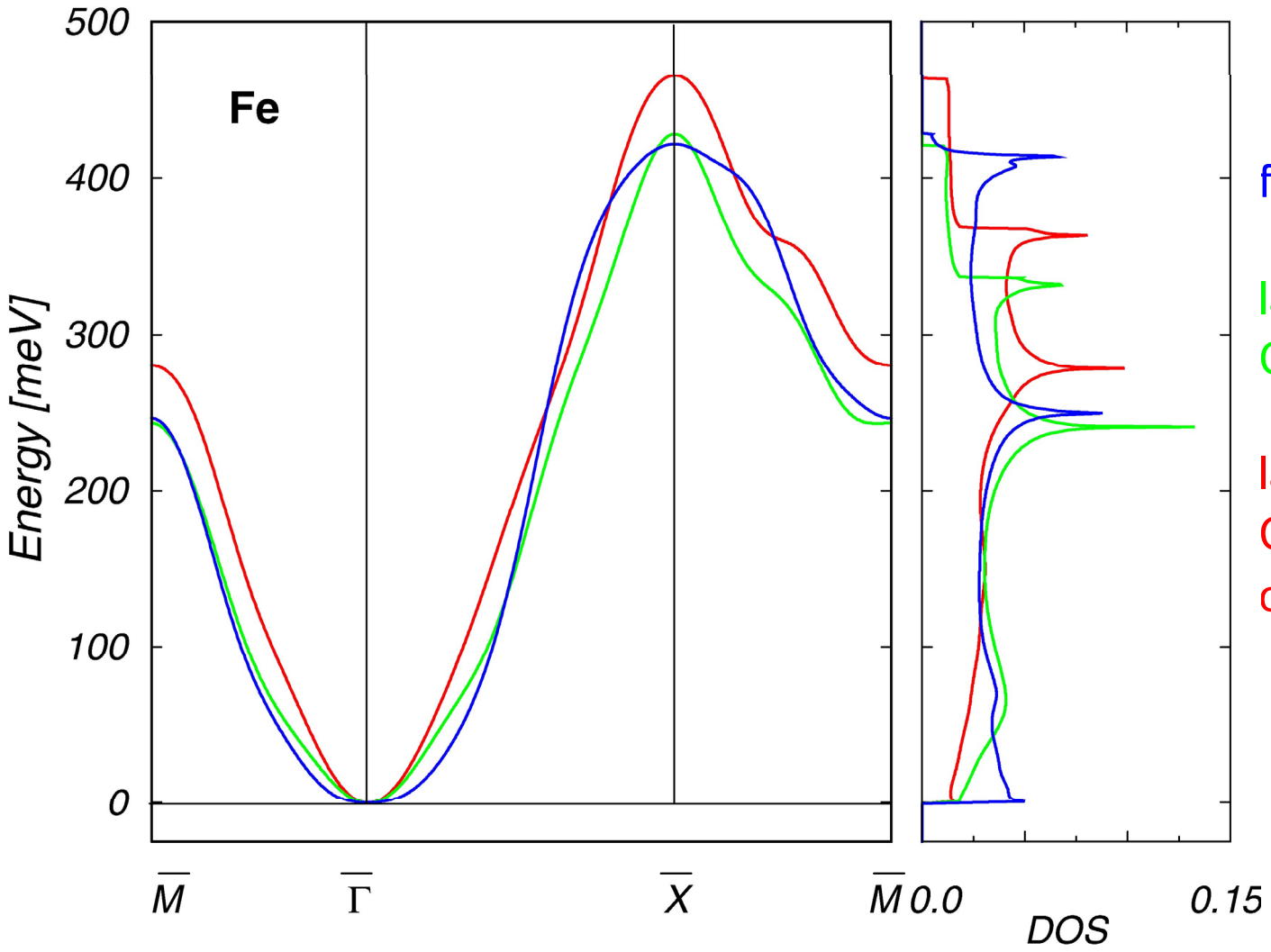
system	m_s (μ_B)	$J_{0,1}$ (meV)	T_C^{MFA} (K)
vac/**Fe**/vac	3.06	46.4	1265
vac/**Fe**/Cu	2.83	36.8	1068
Cu/**Fe**/Cu	2.59	35.9	1189
bulk Fe (bcc)	**2.26**	**19.6**	**1414**
vac/**Co**/vac	2.02	38.7	1300
vac/**Co**/Cu	1.79	31.9	1043
Cu/**Co**/Cu	1.58	27.4	797
bulk Co (fcc)	**1.61**	**14.7**	**1645**

exch. interactions much larger in 2D than in bulk

mean-field approx.
→ much too high T_C in 2D
→ need for better approx. (RPA)

$\mu\Phi$

spin-wave dispersion for an Fe atomic layer

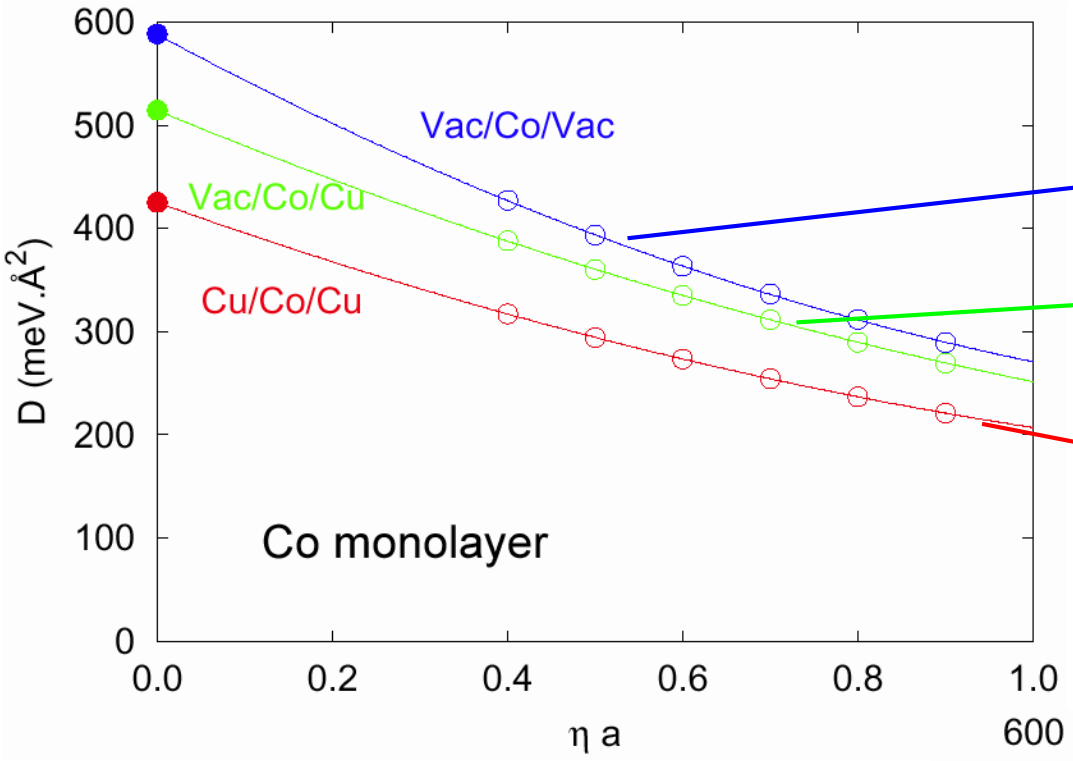


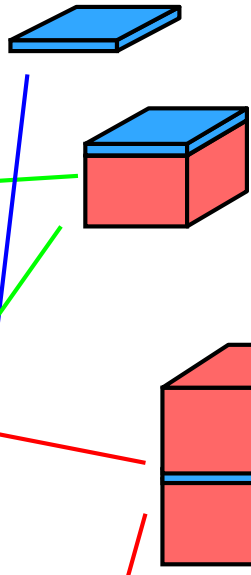
free-standing layer

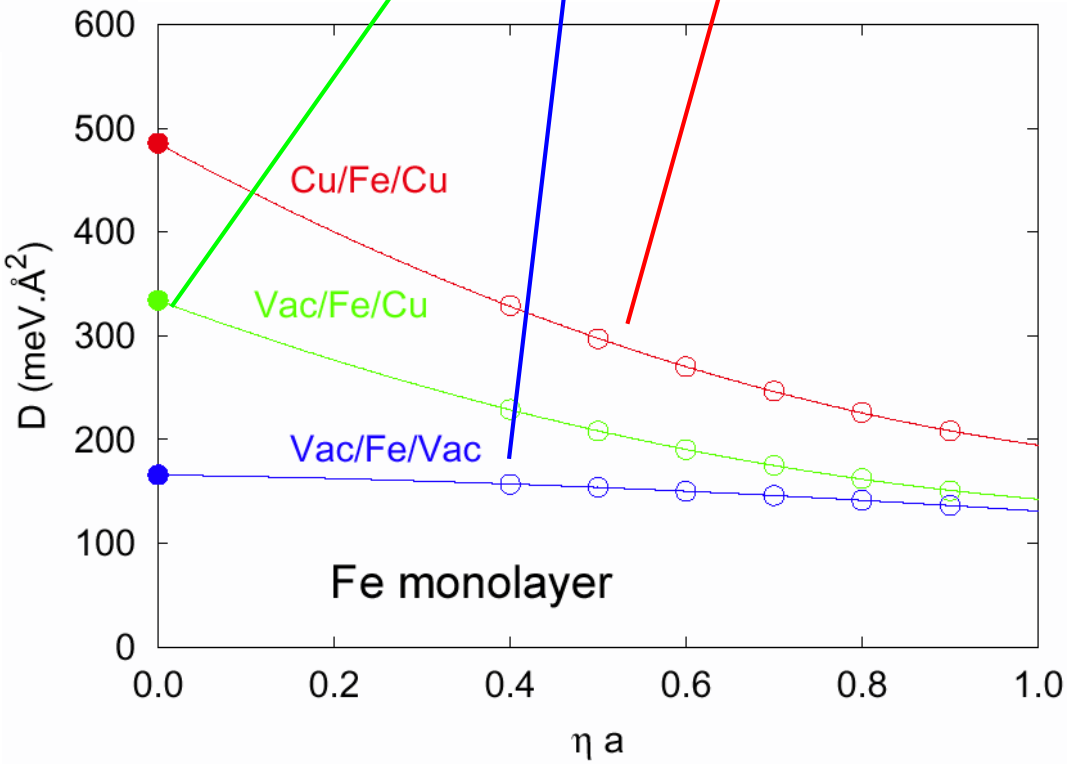
layer deposited on a Cu (001) substrate

layer deposited on a Cu (001) substrate and covered with a Cu cap

$\mu\Phi$







free-standing layer

layer deposited on a
Cu (001) substrate

layer deposited on a
Cu (001) substrate and
covered with a Cu cap

Curie temperature (RPA): $\frac{1}{k_B T_C^{RPA}} = \frac{6\mu_B}{m_s}\frac{1}{N}\sum_{\mathbf{q}}\frac{1}{E(\mathbf{q})}$

spin-wave energy: $E(\mathbf{q}) = \Delta_{\text{anis}} + \frac{4\mu_B}{m_s}\sum_{j\neq 0} J_{0,j}\left(1-\exp(i\mathbf{q}\cdot\mathbf{R}_{0,j})\right)$

$T_C^{2D} \propto \frac{J}{\ln(J/\Delta_{\text{anis}})}$

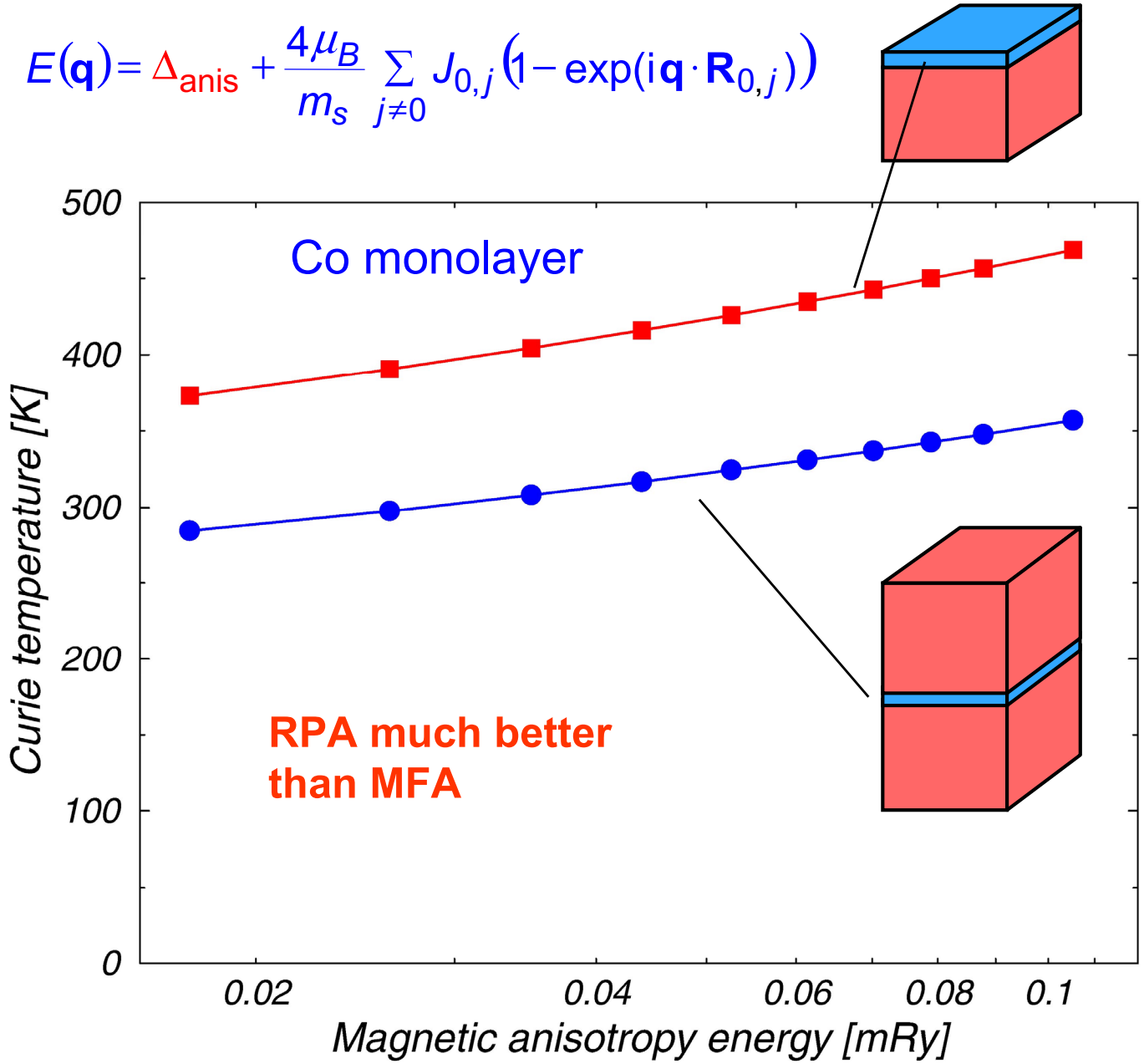
$T_C^{RPA} \rightarrow 0$

for $\Delta_{\text{anis}} \rightarrow 0$

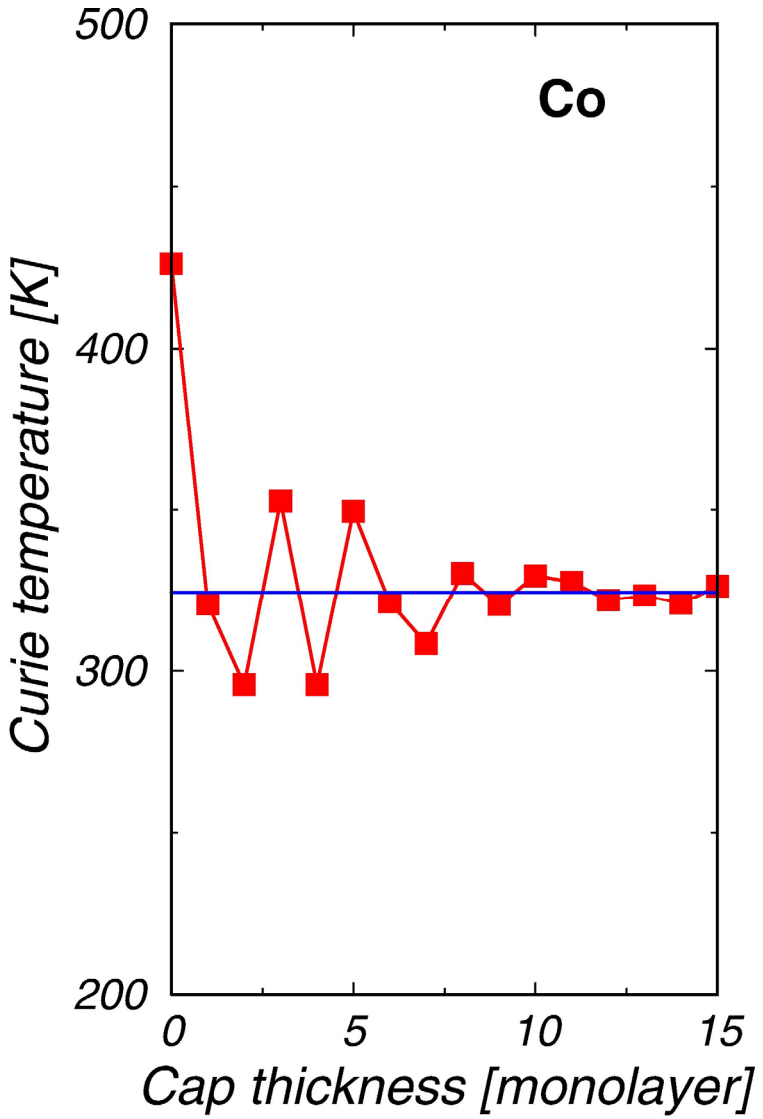
(Mermin-Wagner)

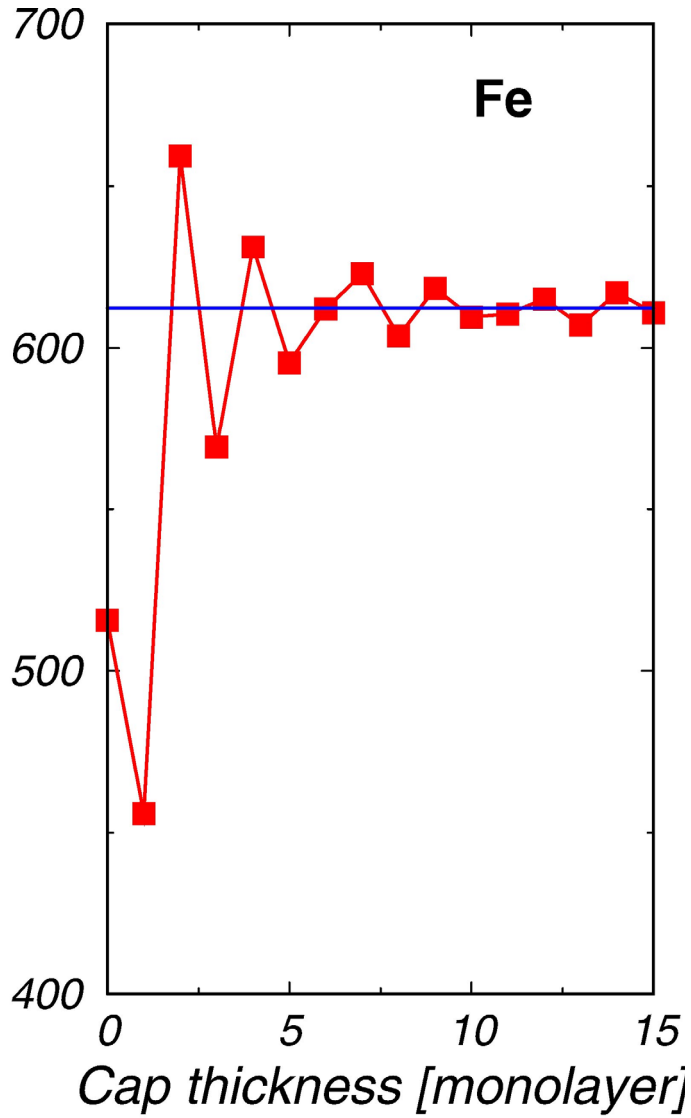
typical order of magnitude of the anisotropy:

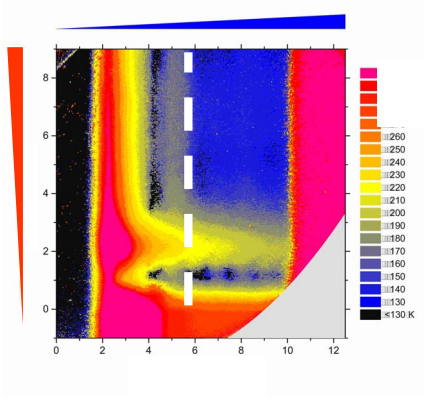
$\Delta_{\text{anis}} \approx \frac{2\pi M^2}{V}$

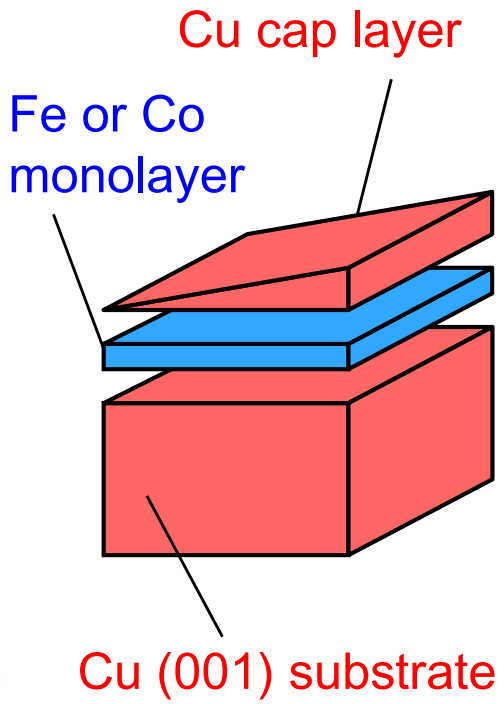


oscillation of the Curie temperature (RPA) as a function of the cap layer thickness







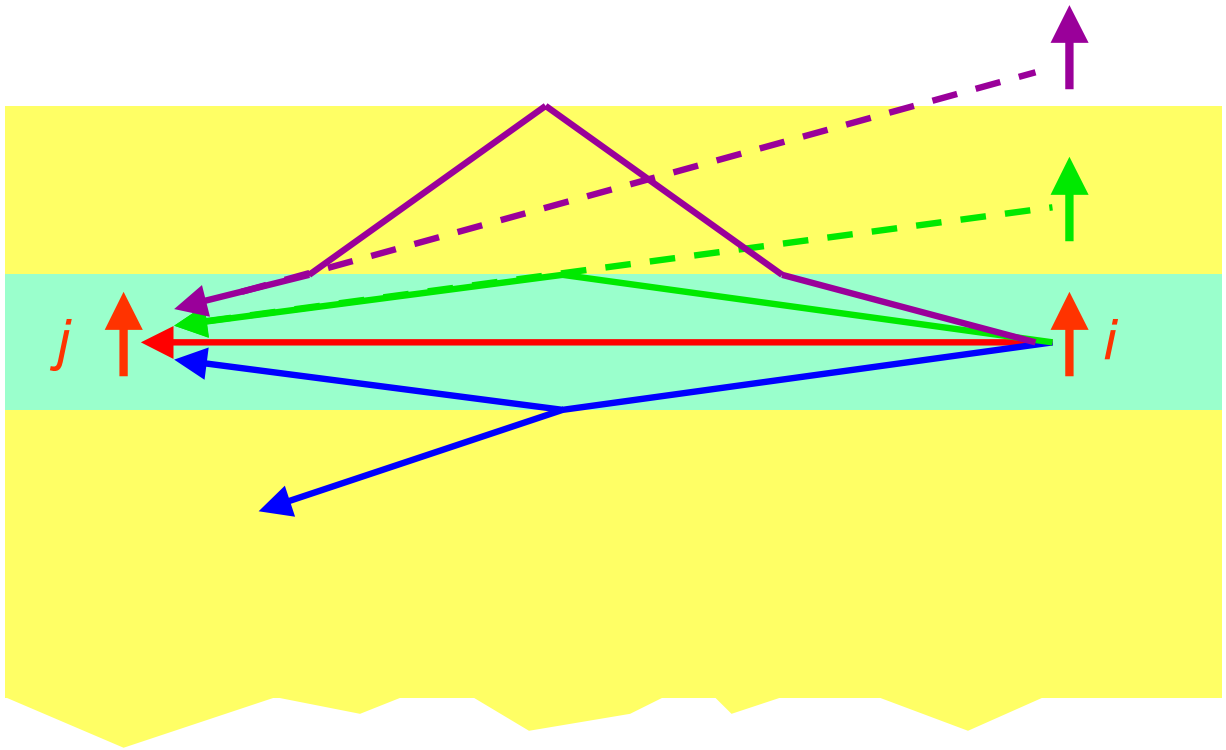


Qualitative interpretation of the results

$$J_{i,j} \propto \mathrm{Im} \int \mathrm{d}\varepsilon \, f(\varepsilon) \, \mathrm{Tr}\left[\left(V_i^{\uparrow} - V_i^{\downarrow}\right) G_{ij}^{\uparrow} \left(V_j^{\uparrow} - V_j^{\downarrow}\right) G_{ji}^{\downarrow}\right]$$

for bulk (3D):

$$G_{ij} \propto \frac{\exp\left(\mathrm{i}\, k R_{ij}\right)}{R_{ij}}$$

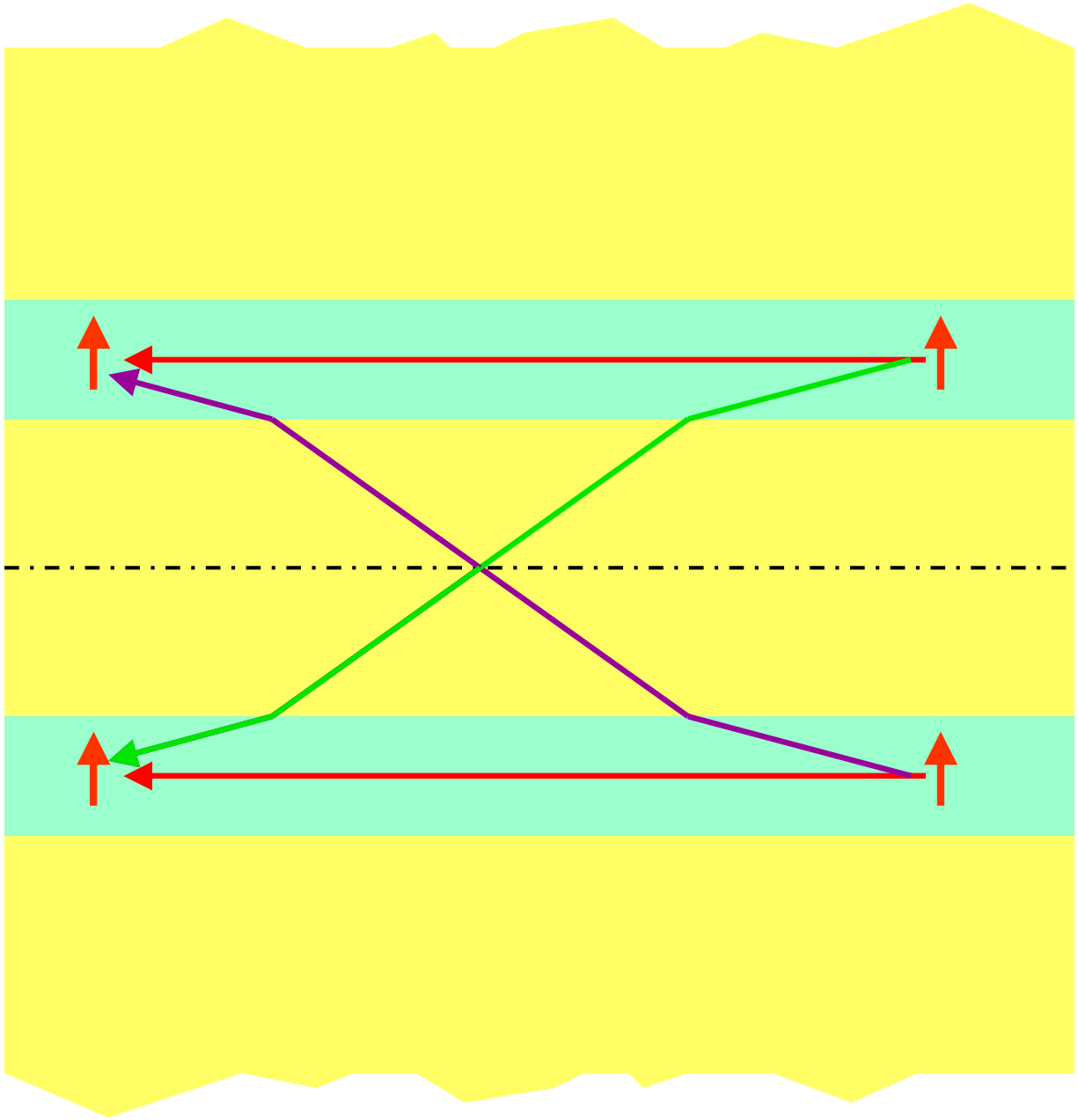


for 2D:

$$G_{ij} \propto \frac{\exp\left(\mathrm{i}\, k R_{ij}\right)}{\left(R_{ij}\right)^{1/2}}$$

$\mu\Phi$

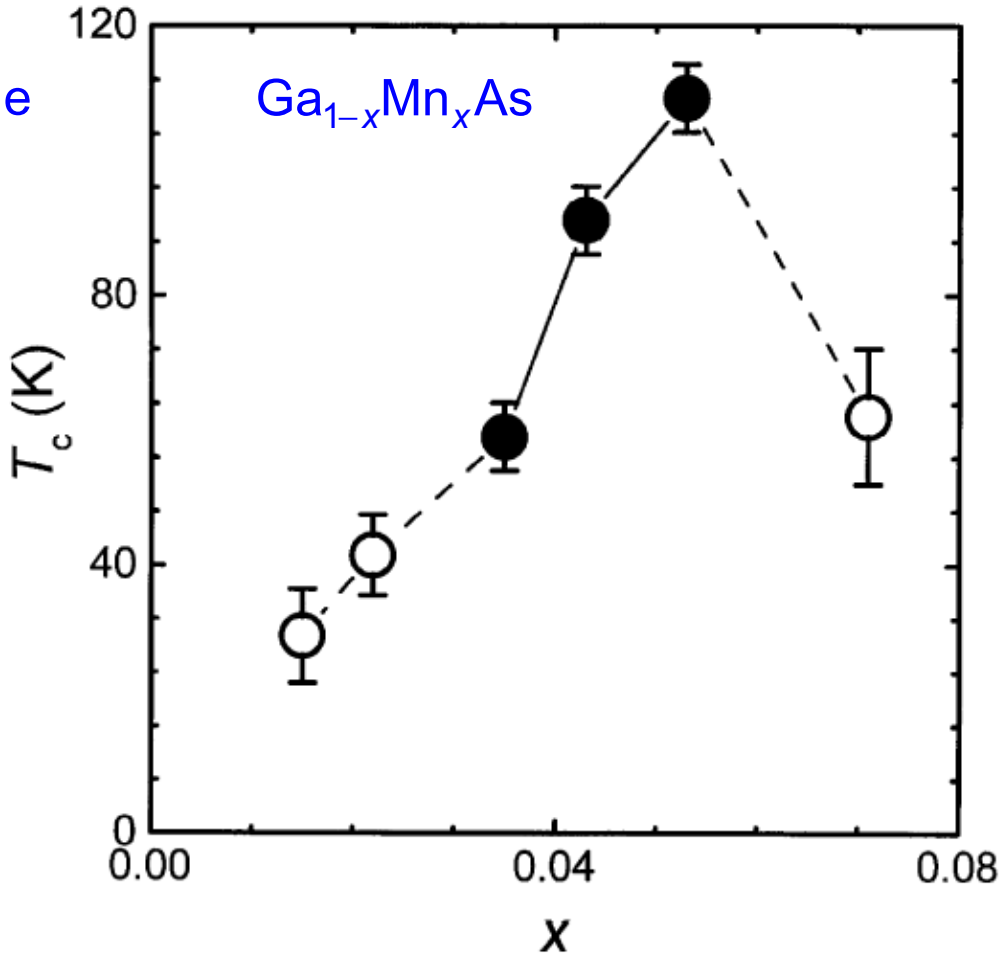
Analogy to oscillatory interlayer exchange coupling



Dilute magnetic semiconductors (DMS)

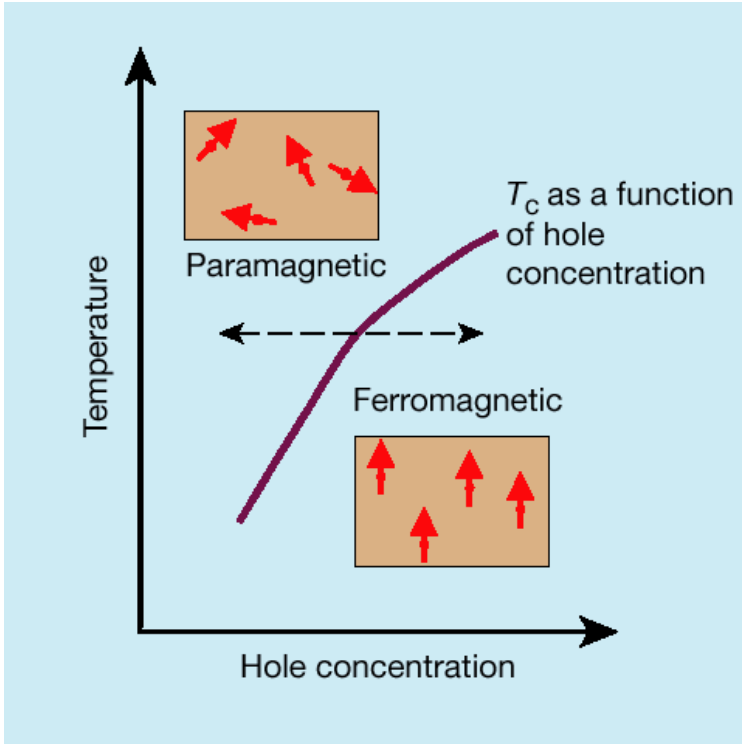
Mn $\rightarrow$ $S = 5/2$ + 1 hole

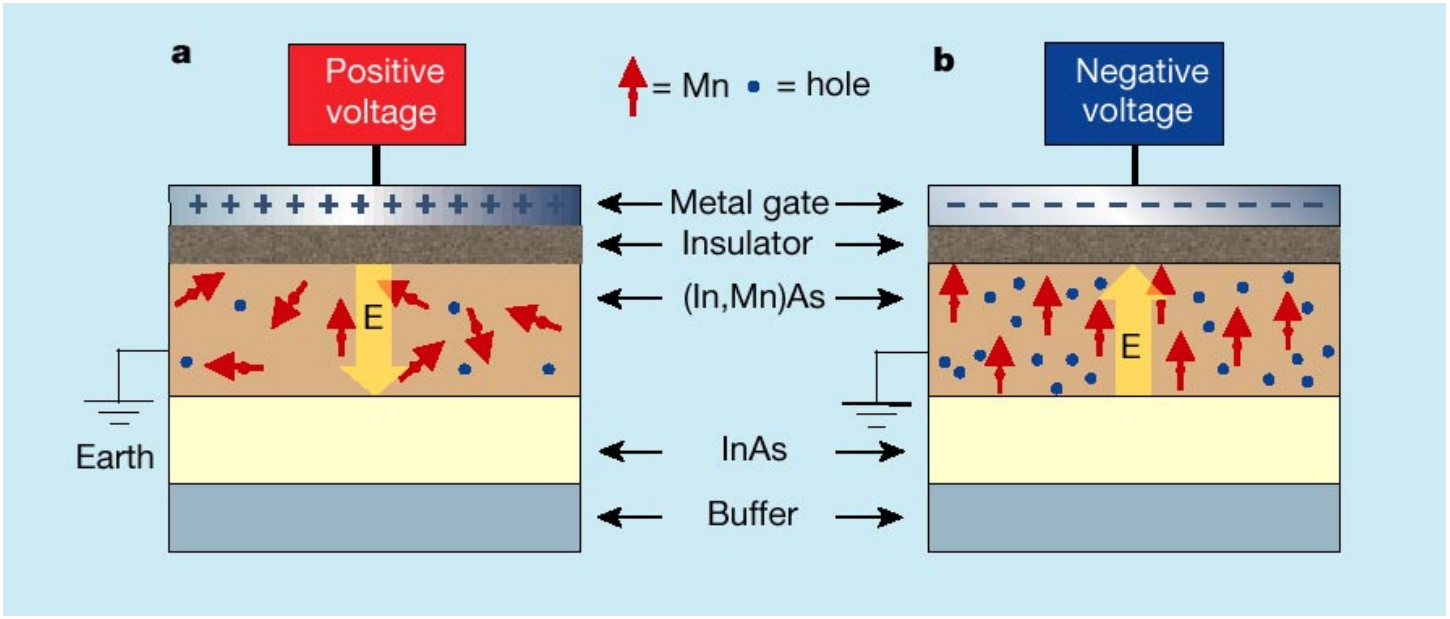
$J_{p\text{-}d} \approx -1.2$ eV (AF)

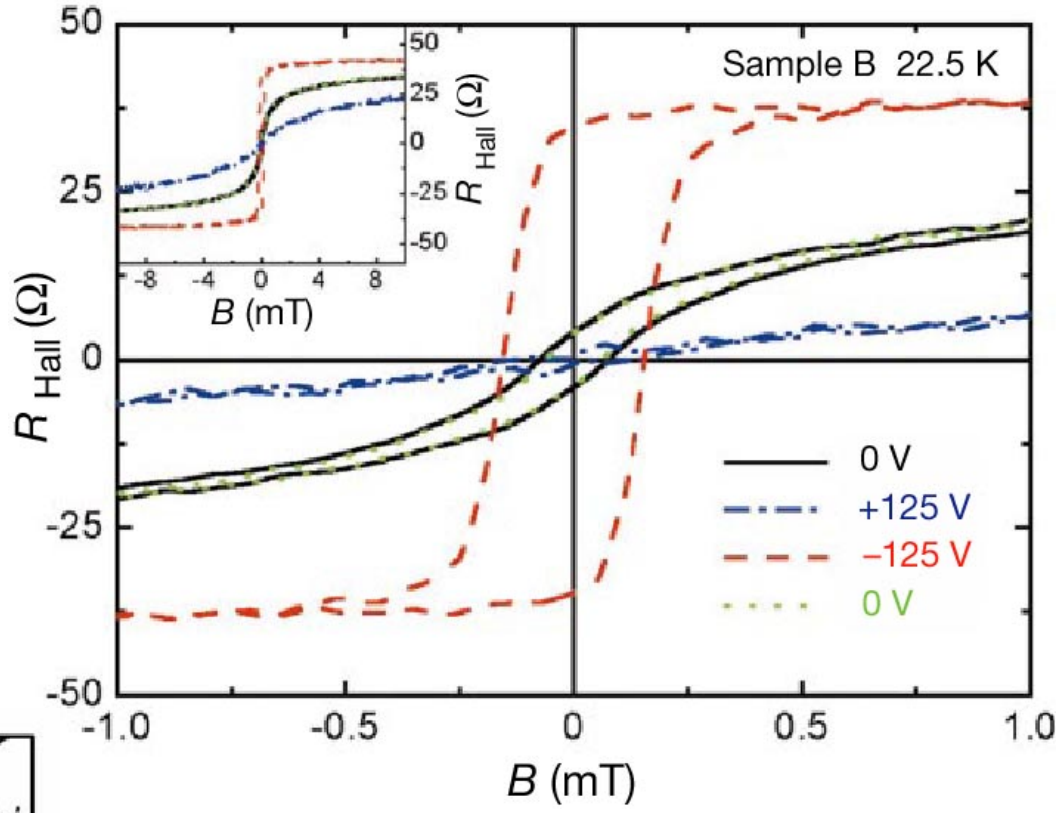


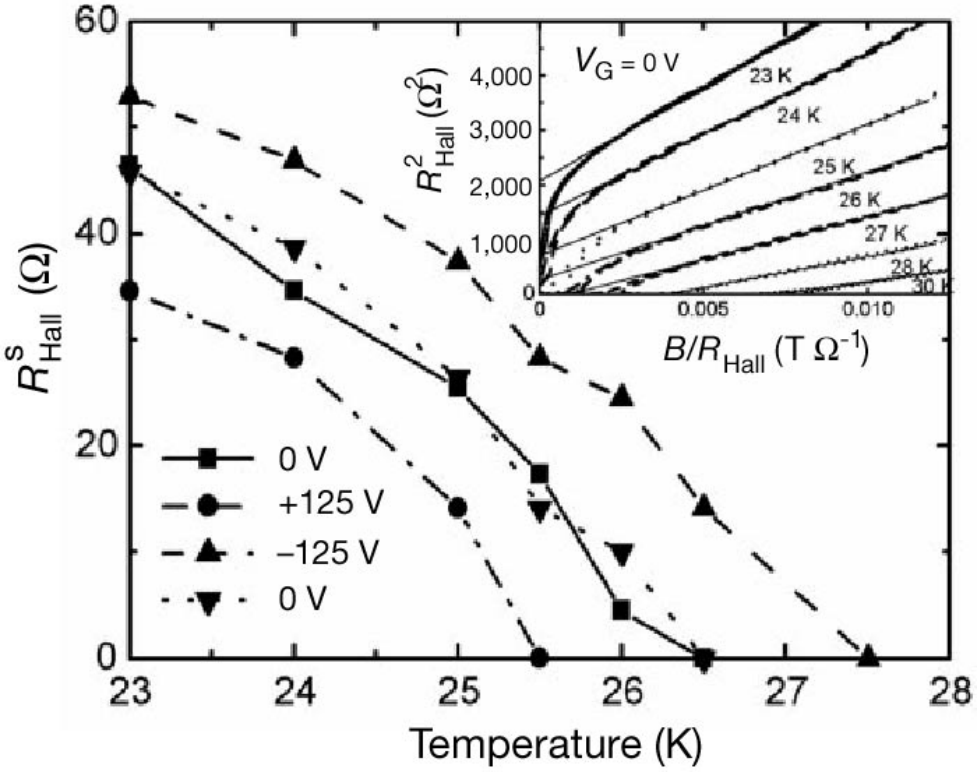
H. Ohno, Science **281**, 951 (1998)

$\mu\Phi$


Temperature
Paramagnetic
T_c as a function of hole concentration
Ferromagnetic
Hole concentration


a
Positive voltage
= Mn
= hole
b
Negative voltage
Metal gate
Insulator
(In,Mn)As
E
E
Earth
InAs
Buffer





H. Ohno *et al.*, Nature **408**, 944 (2000)

Calculation of exchange interactions and Curie temperature of DMS

$Ga_{1-x}Mn_xAs$

supercell approach (ordered compound)

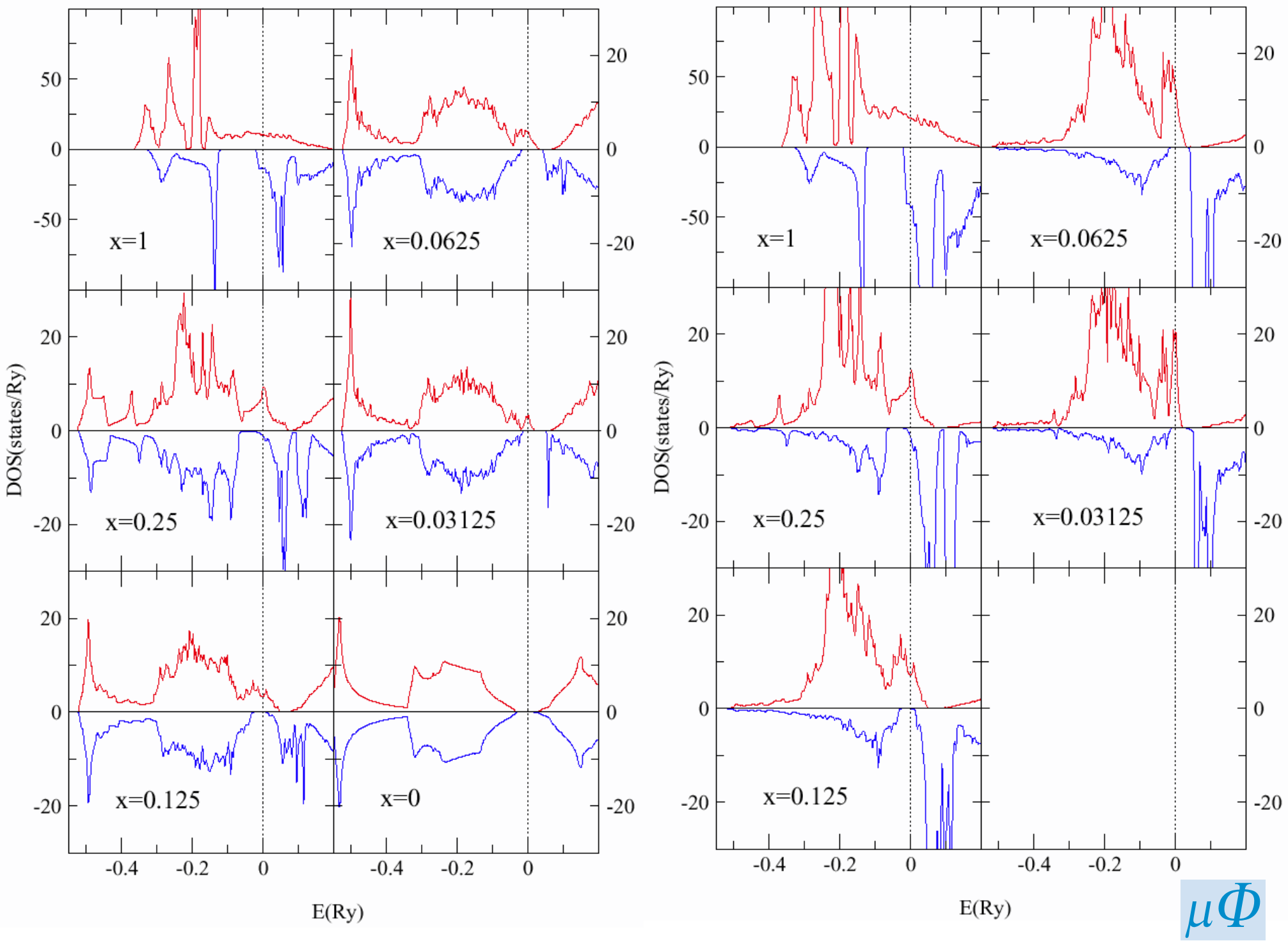
x = 0, 3.125%, 6.25%, 12.5%, 25%, 100%

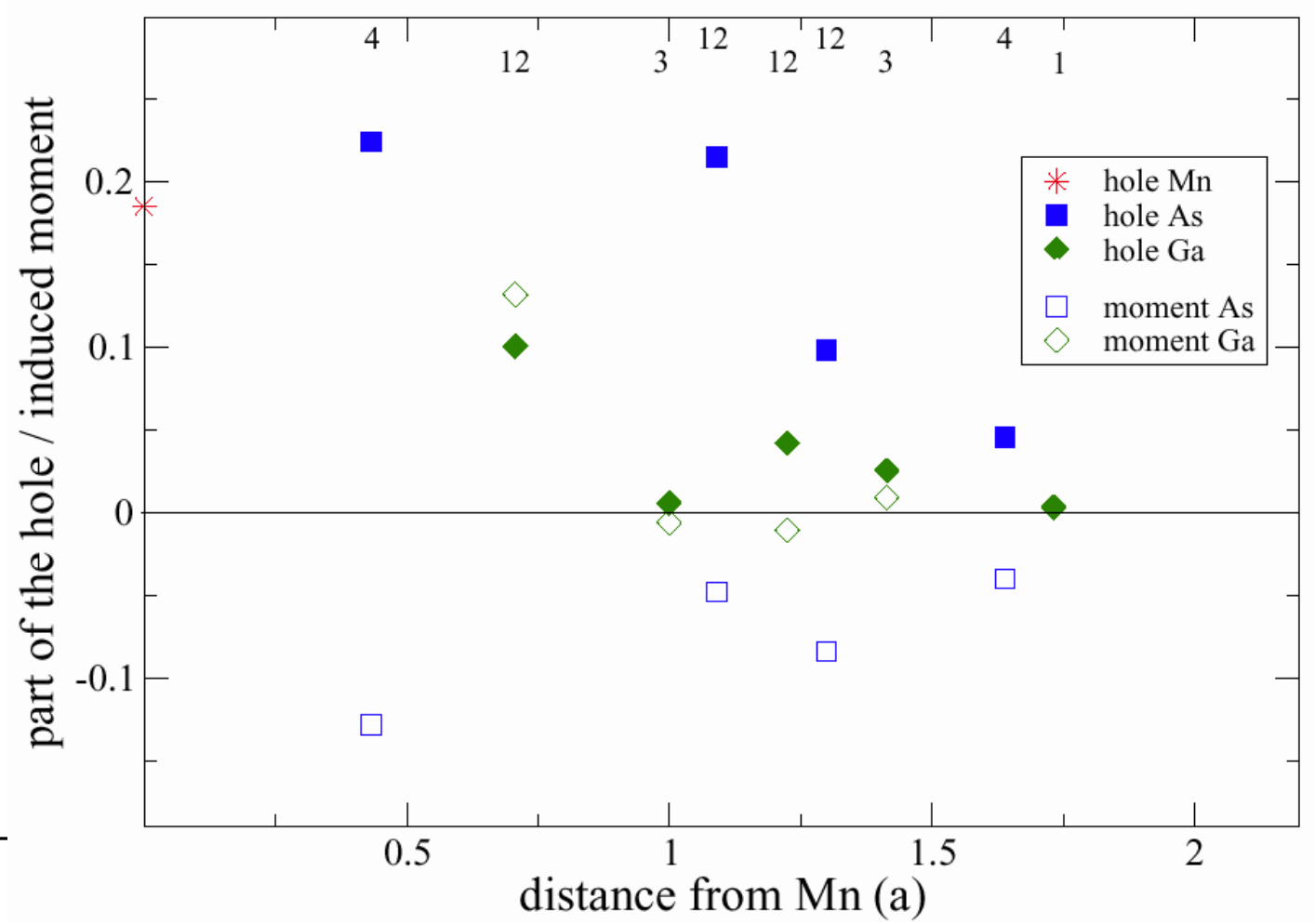
range accessible experimentally

augmented spherical waves (ASW) method

(calculations performed by Leonid Sandratskii)

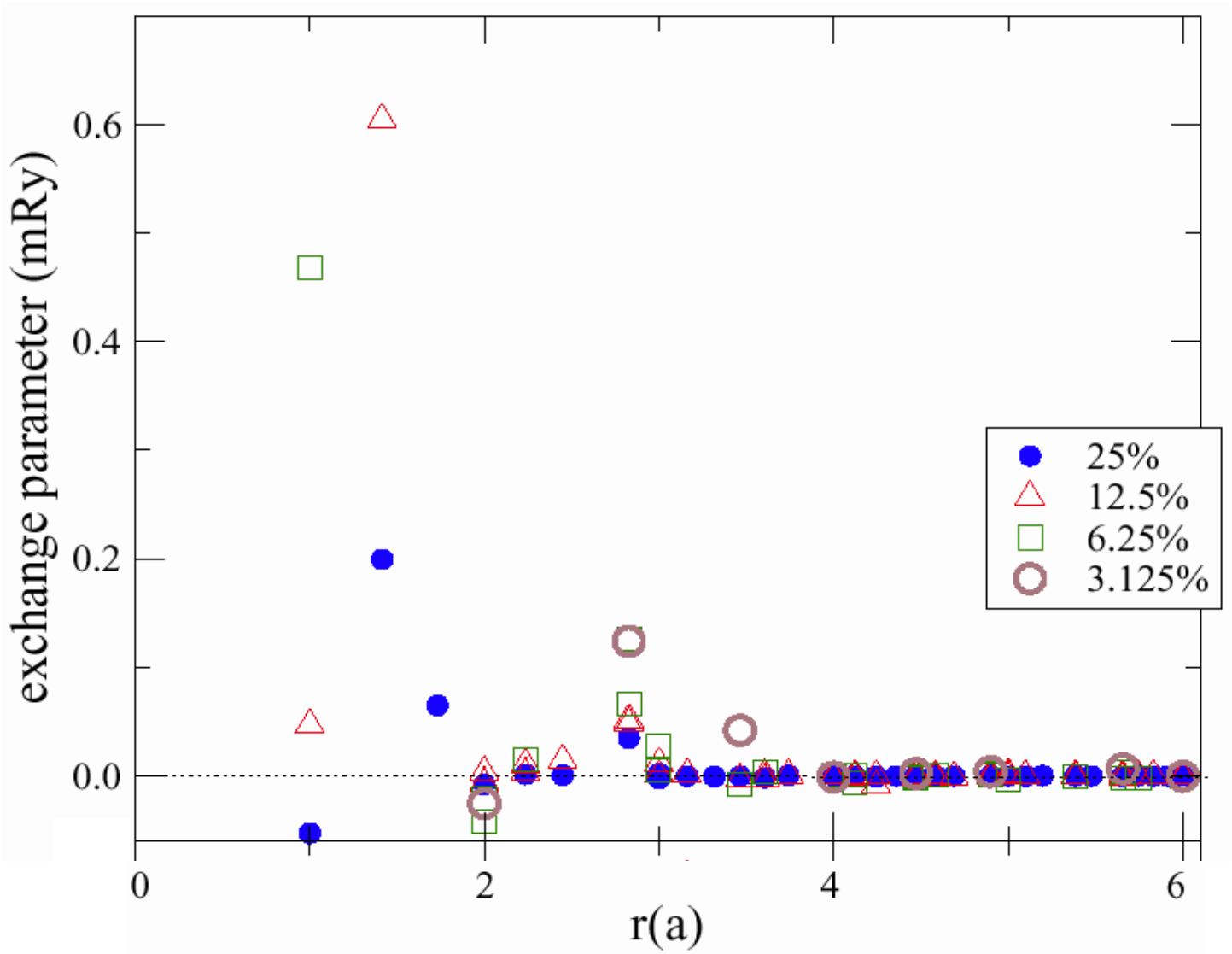
$\mu\Phi$

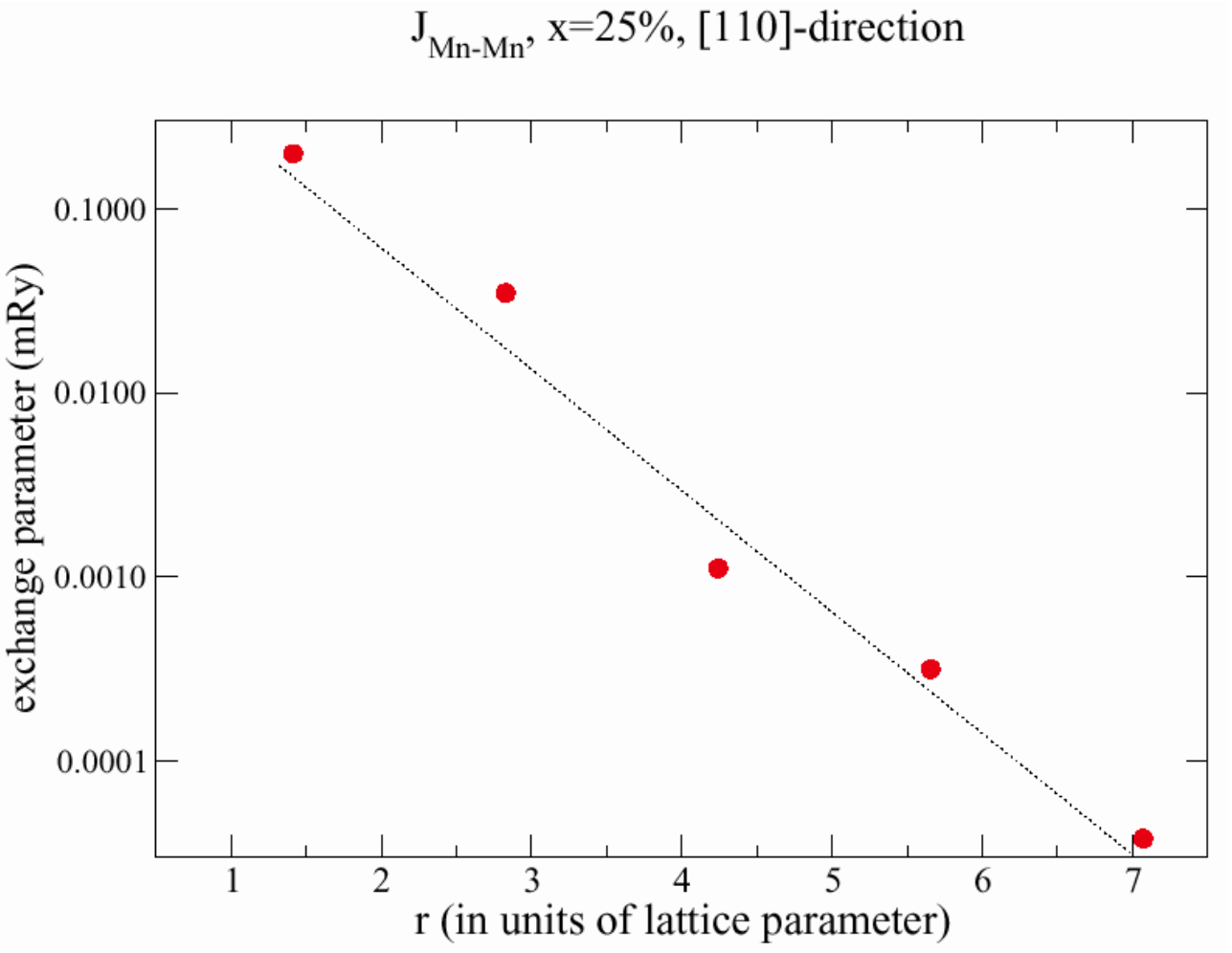

x=1
x=0.0625
x=0.25
x=0.03125
x=0.125
x=0
DOS(states/Ry)
E(Ry)



x	1	0.25	0.125	0.0625	0.03125
Mn	3.76	3.85	3.88	3.94	3.95
As	-0.18	-0.046	-0.046	-0.036	-0.032
cell	3.65	3.98	4.00	4.00	4.00

magnetic moment (in μ_B)

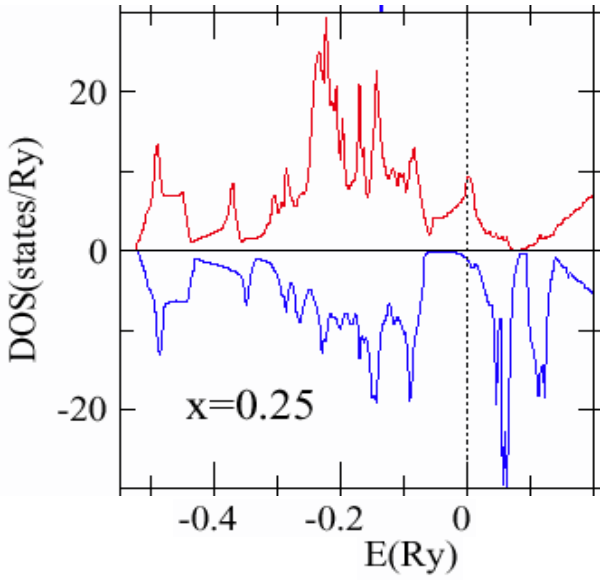

exchange parameter (mRy)
0.6
0.4
0.2
0.0
0
2
4
6
r(a)
25%
12.5%
6.25%
3.125%

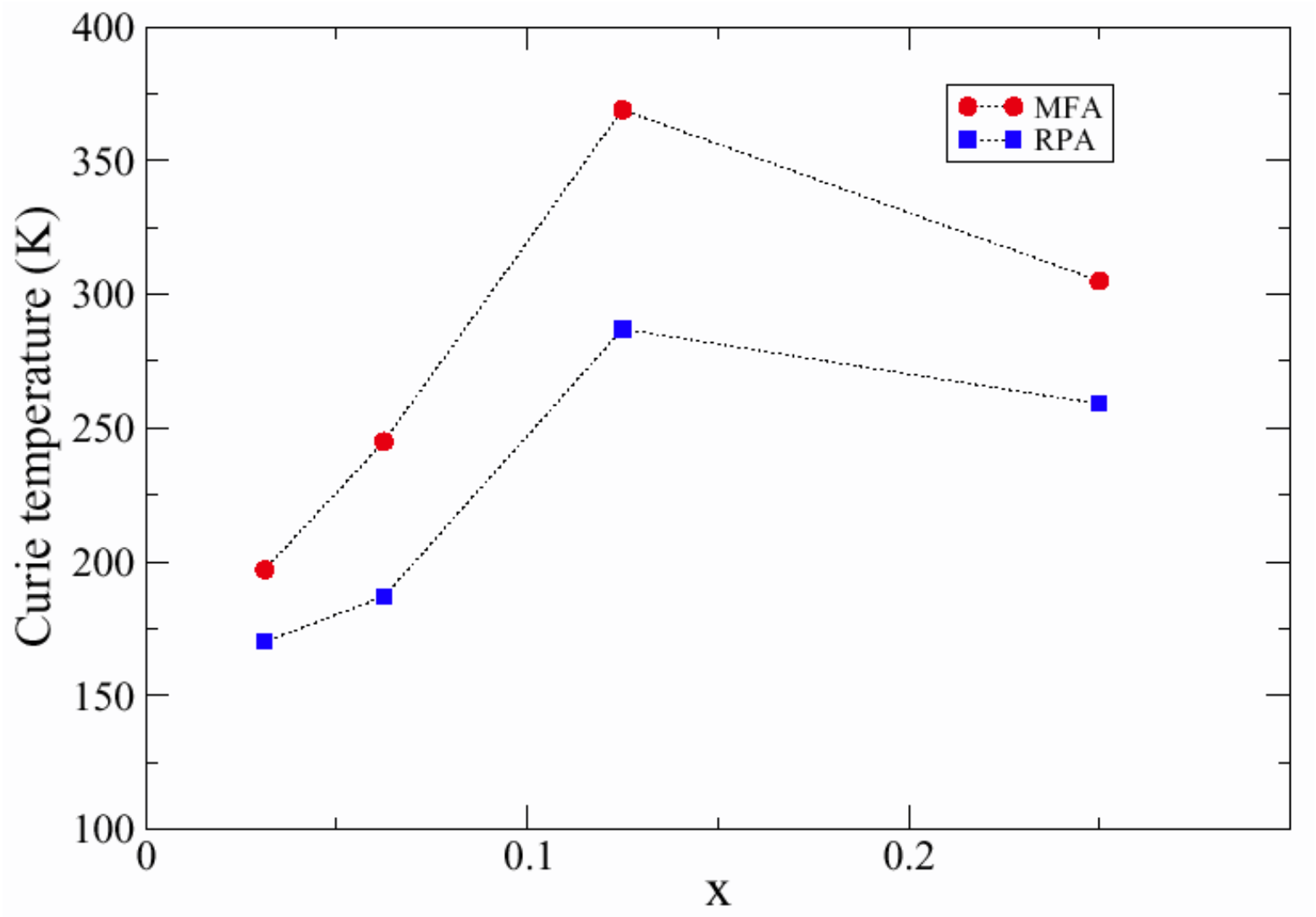


$$J_{\mathbf{RR}'} \sim \mathrm{Im}\left[G^{+}_{\uparrow}(\varepsilon_F)\, G^{+}_{\downarrow}(\varepsilon_F) \right]$$

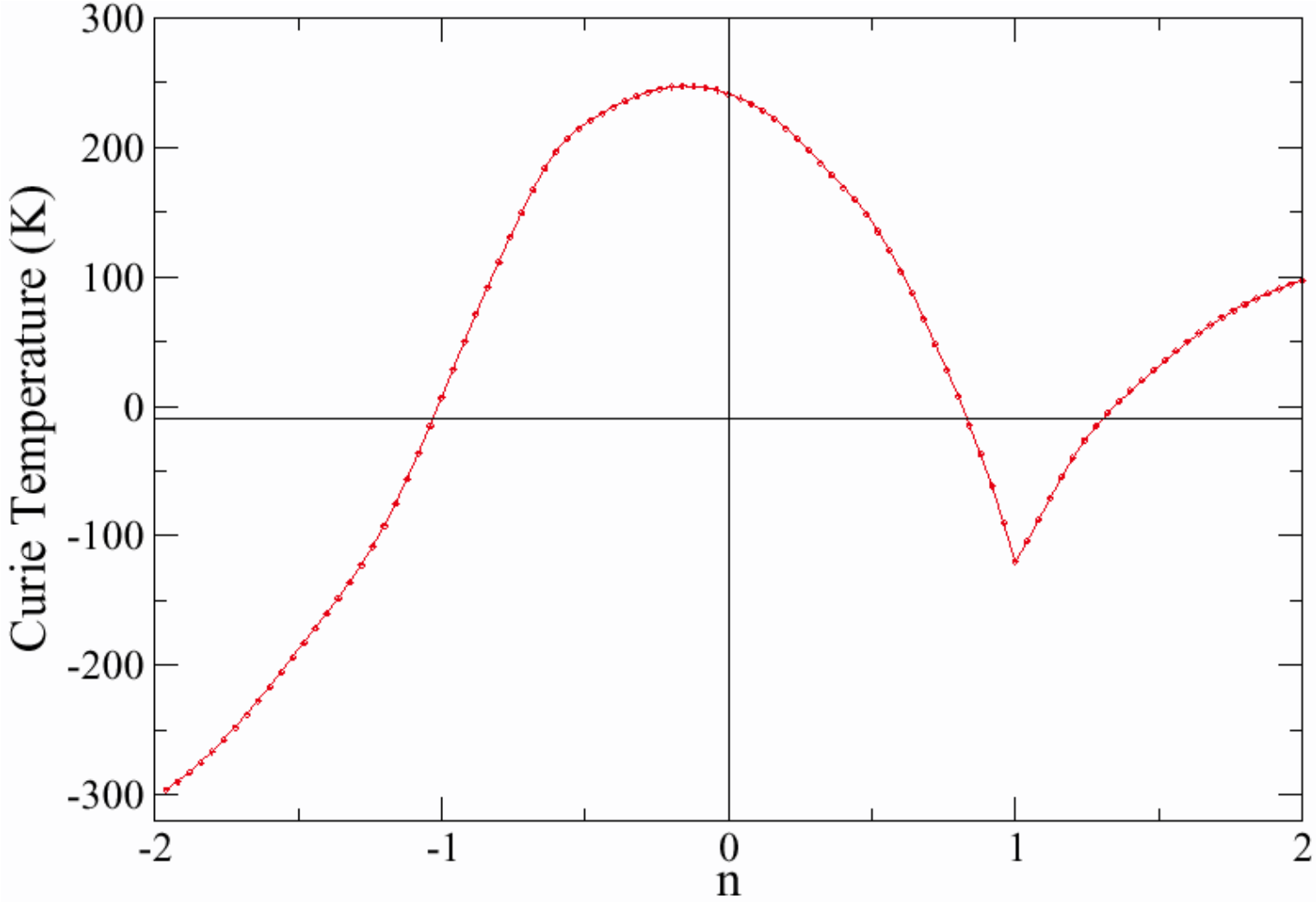
$$G^{+}_{\uparrow}(\varepsilon_F) \sim \frac{\exp\left(\mathrm{i} k_F^{\uparrow} \left| \mathbf{R} - \mathbf{R}' \right| \right)}{\left| \mathbf{R} - \mathbf{R}' \right|}$$

$$G^{+}_{\downarrow}(\varepsilon_F) \sim \frac{\exp\left(-\kappa_F^{\downarrow} \left| \mathbf{R} - \mathbf{R}' \right| \right)}{\left| \mathbf{R} - \mathbf{R}' \right|}$$




400
350
300
250
200
150
100
Curie temperature (K)
0
0.1
0.2
x
MFA
RPA

(mean-field) Curie temperature for x = 6.25%



excess of electrons per Mn atom

Conclusions and outlook

- ``*renormalized* magnetic force theorem´´ corrects the systematic error introduced by the ``*bare* magnetic force theorem´´
- no signficant additional computational effort → suitable for complex systems
- simple approximation yields simple renormalization of spin-wave energies and Curie temperature
- Curie temperature of Fe and Ni are in good agreement with experimental ones (?)
- application to: - disordered systems (e.g., permalloy, NiPd, ...)
  - films and surfaces
  - nanostructures
  - new materials (e.g., GaMnAs, ...)
- needed improvements: local moments at $T > T_C$
  quantum fluctuations
  ? combination with the DMFT approach ?

$\mu\Phi$

Acknowledgements

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Valeri Stepanyuk

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$\mu\Phi$