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Why are the Co-based 115 compounds different?: The case study of $GdMIn_5$ (M=Co,Rh,Ir)

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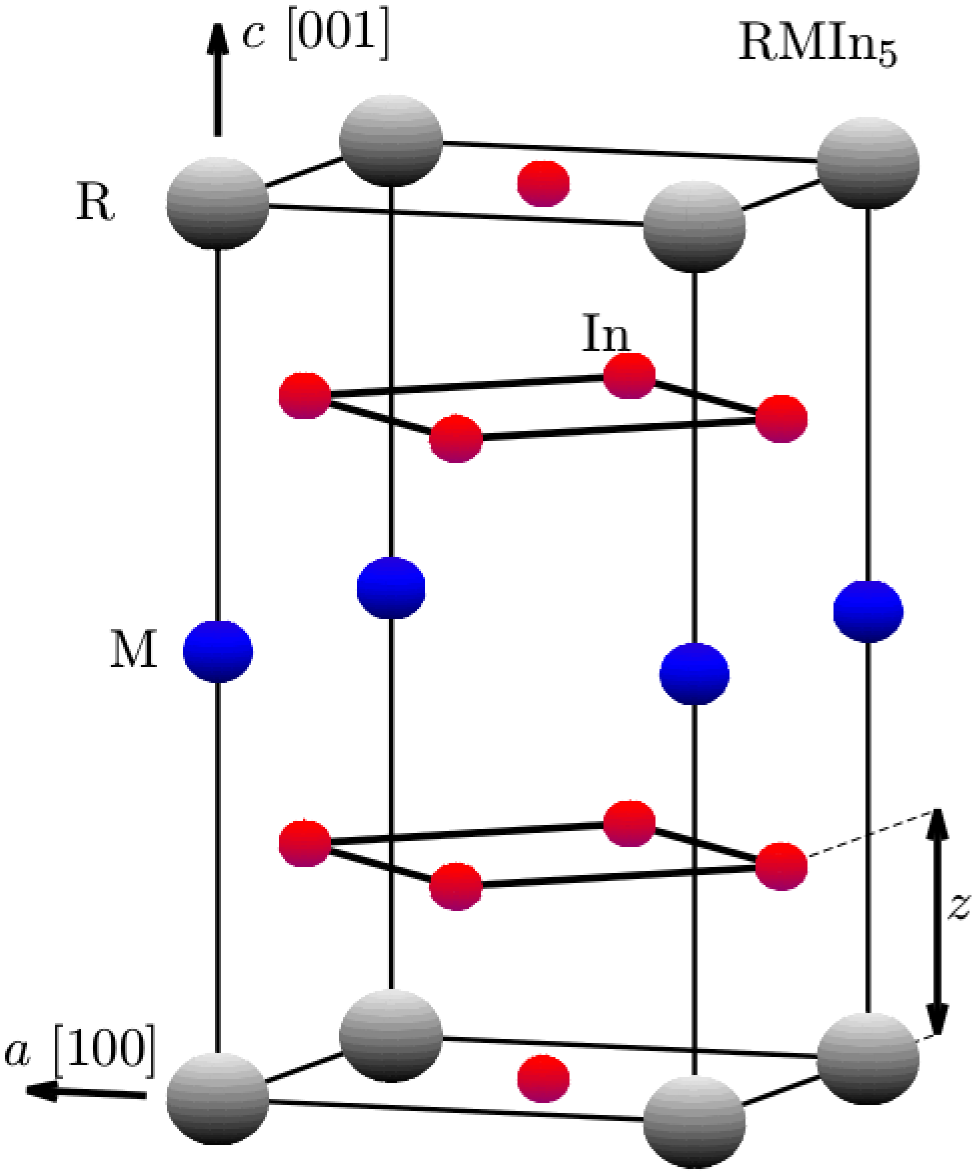
Colaborators

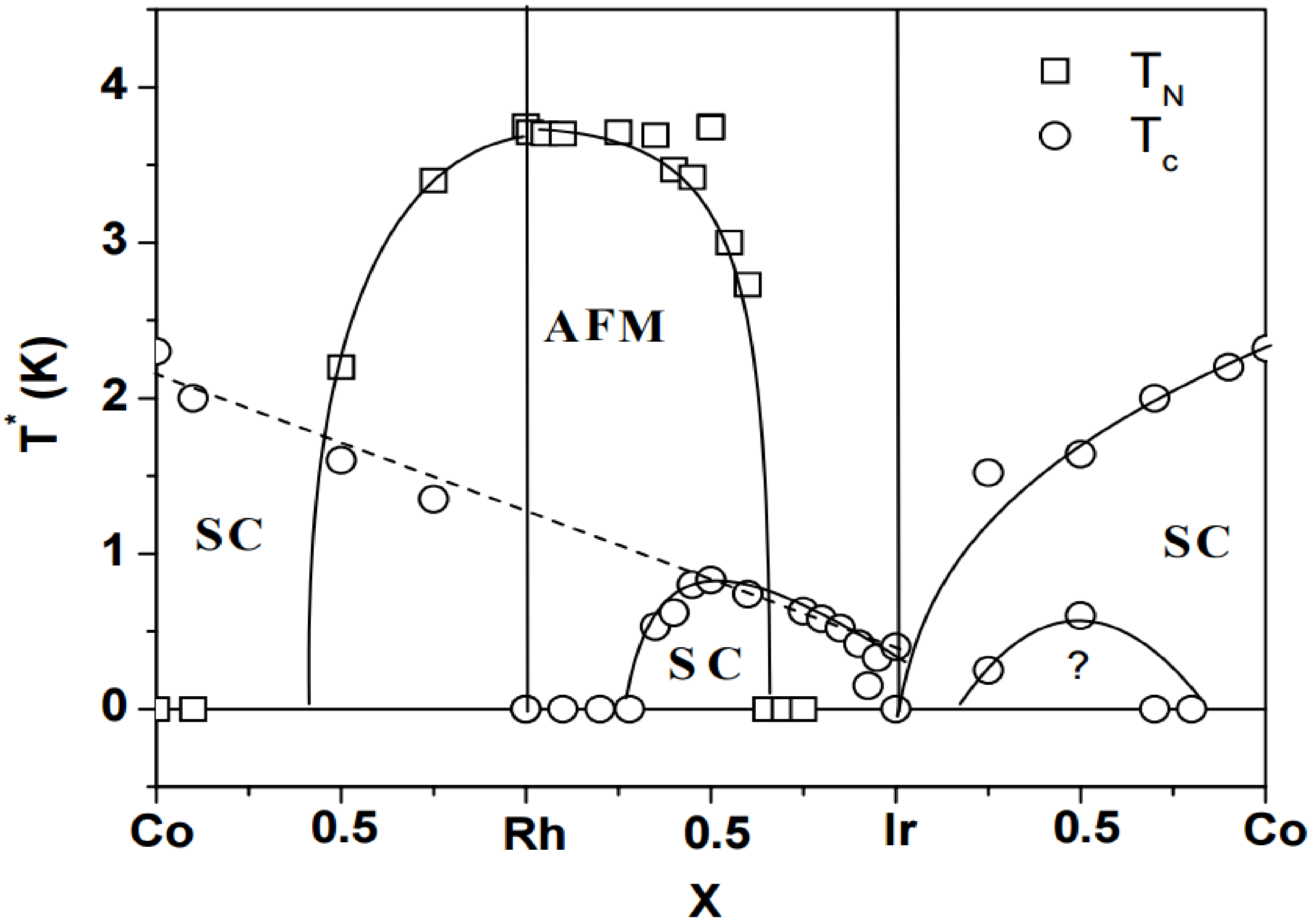
- Diana Betancourth, Pablo Pedrazzini, Víctor Correa (**Low Temperature Physics Laboratory, Bariloche**)
- Jorge Facio, Daniel García (**Condensed Matter Theory Group, Bariloche**)
- Verónica Vildosola (**Condensed Matter Theory Group, Buenos Aires**)
- We also benefited from the interaction with Eduardo Granado, Pascoal Pagliuso & Raimundo Lora Serrano (**Campinas**)

The 115 compounds



- RIn_3, MIn_2 planes.
- Interesting physics dominated by 4f electrons.
- Heavy fermion behavior (up to x1000 mass enhancement).
- Unconventional superconductivity.
- Complex magnetic states.
- While Co, Rh and Ir are isovalent, they produce very different ground states in the R=Ce compounds.

Properties of R=Ce 115 compounds

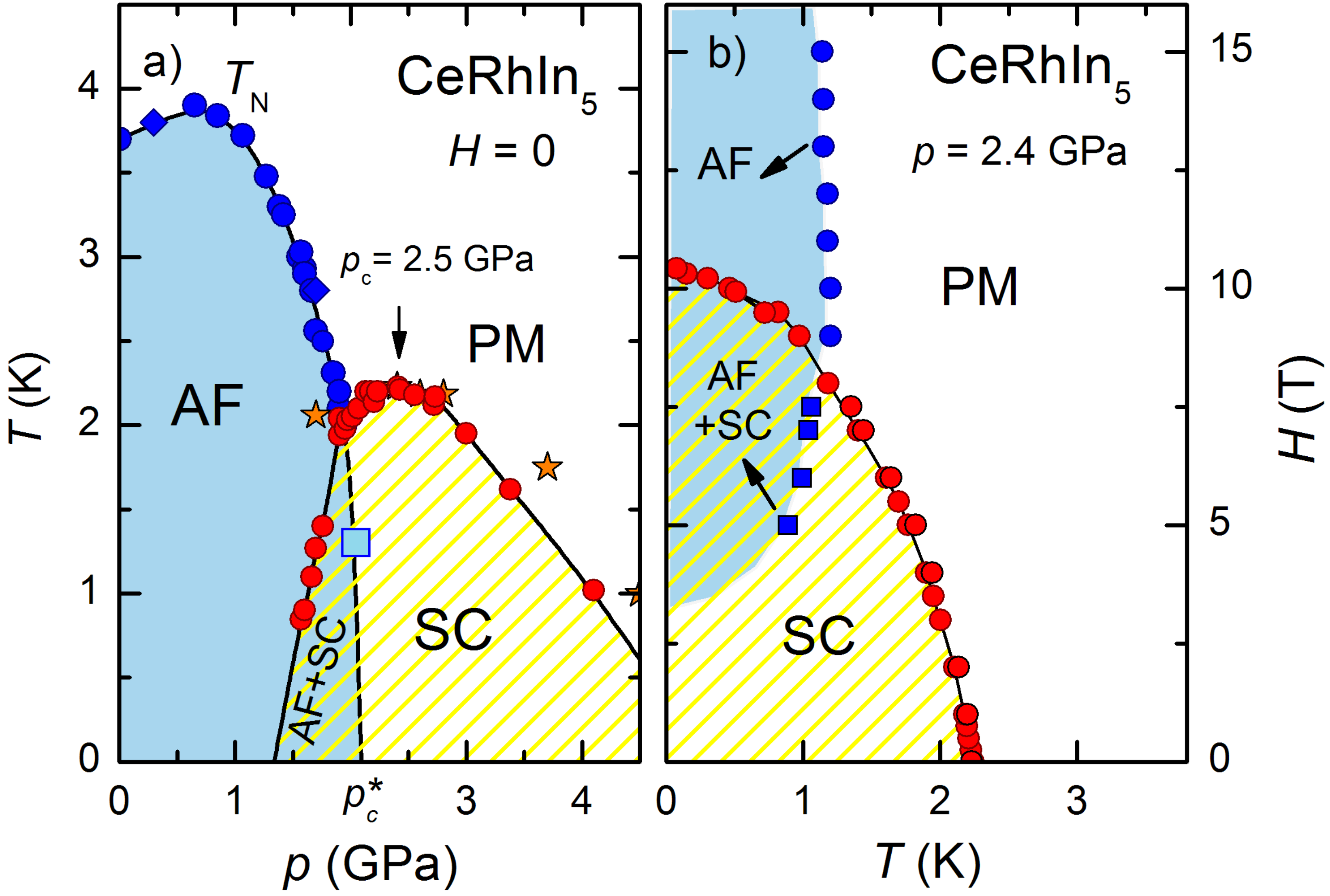


Phase diagram of Ce(Co,Rh,Ir)In_5

- $\mathrm{CeCoIn_5}$ T_C=2.3K
- $\mathrm{CeRhIn_5}$ T_N=3.6K
- $\mathrm{CeIrIn_5}$ T_C=0.4K

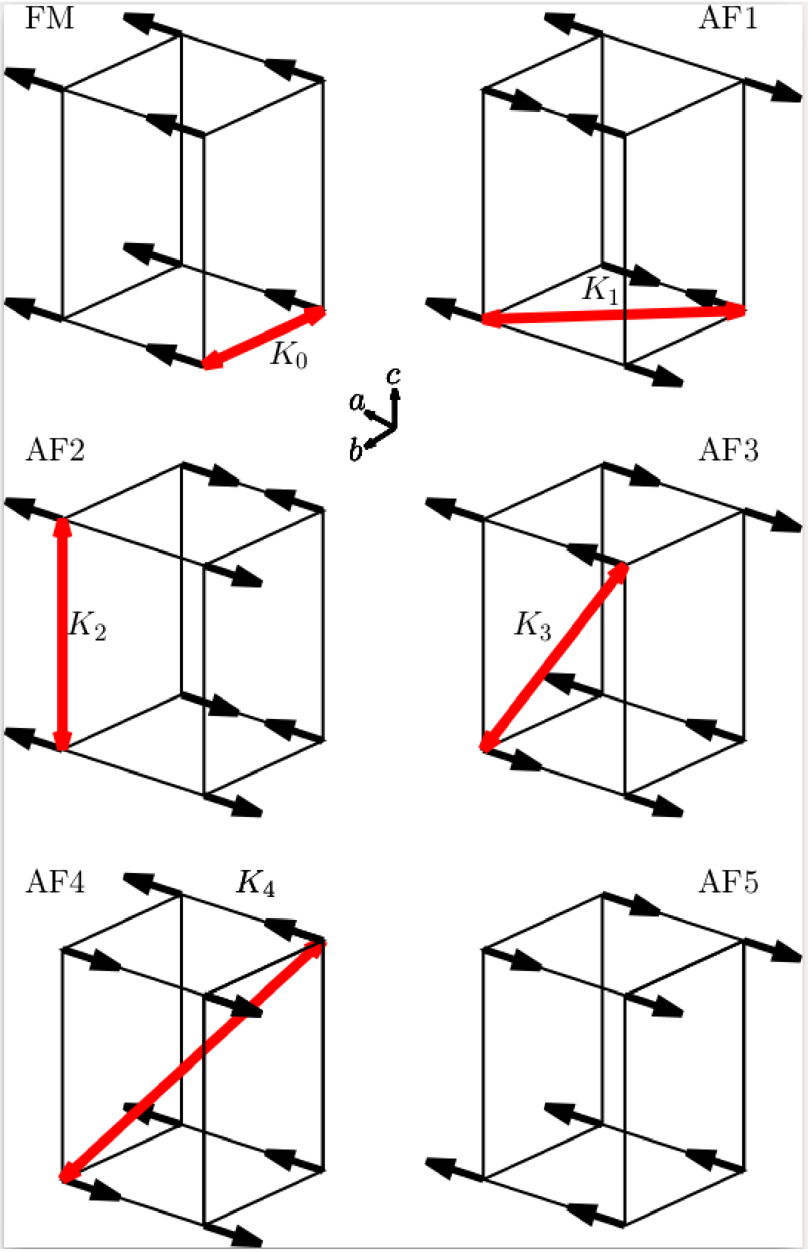
P. G. Pagliuso *et al.*, Physica B **312** 129 (2002)

Magnetism and superconductivity in $CeRhIn_5$



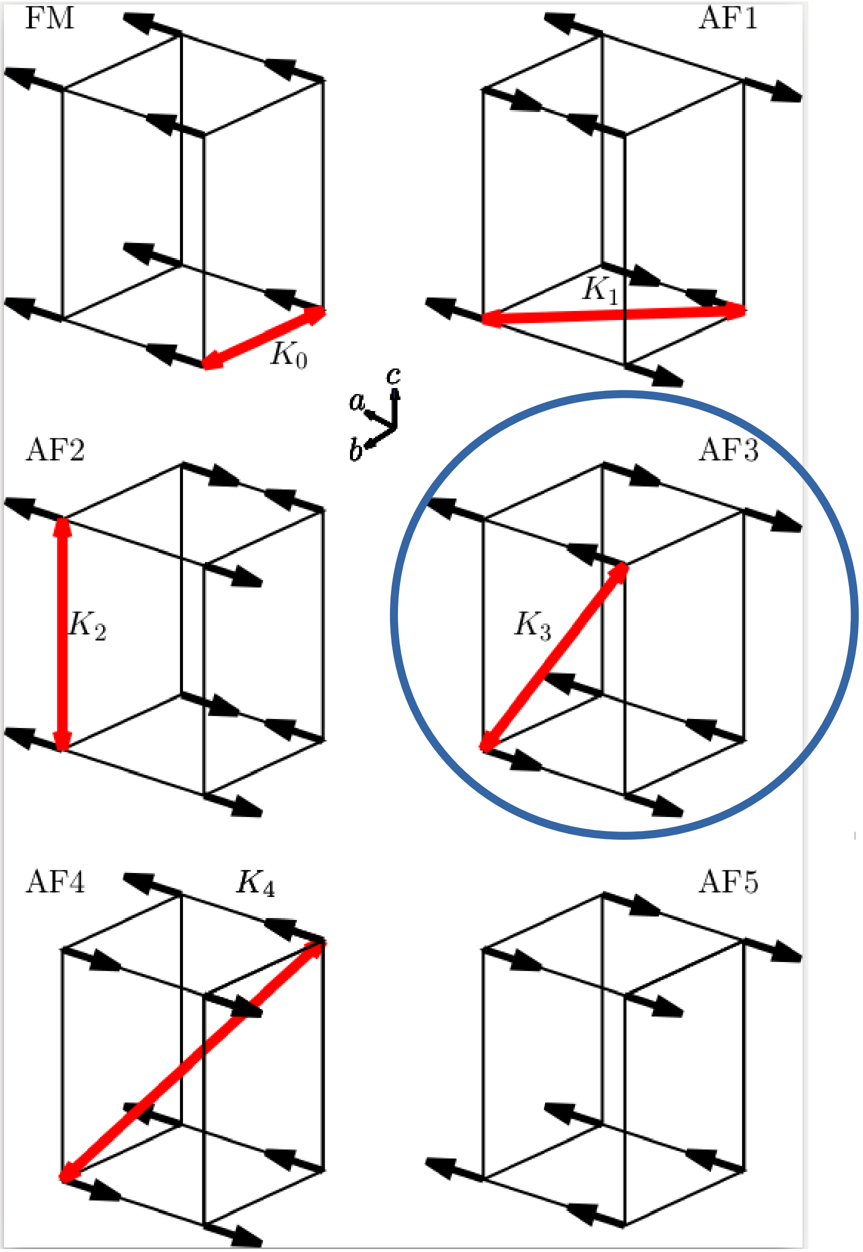
G. Knebel *et al.*, Phys Rev B **74** 020501 (2006)

Magnetic 115 compounds



- Magnetic moments in the 4f levels of the Rare Earth.
- Exchange interactions mediated by conduction electrons.
- C-type antiferromagnet:
  - DyRhIn5 (magnetization),
  - HoRhIn5 (magnetization),
  - NdRhIn5 (neutron diffraction)
  - GdRhIn5 (resonant x-ray diffraction)
- Competition between antiferromagnetic couplings K_0 & K_1

Magnetic 115 compounds



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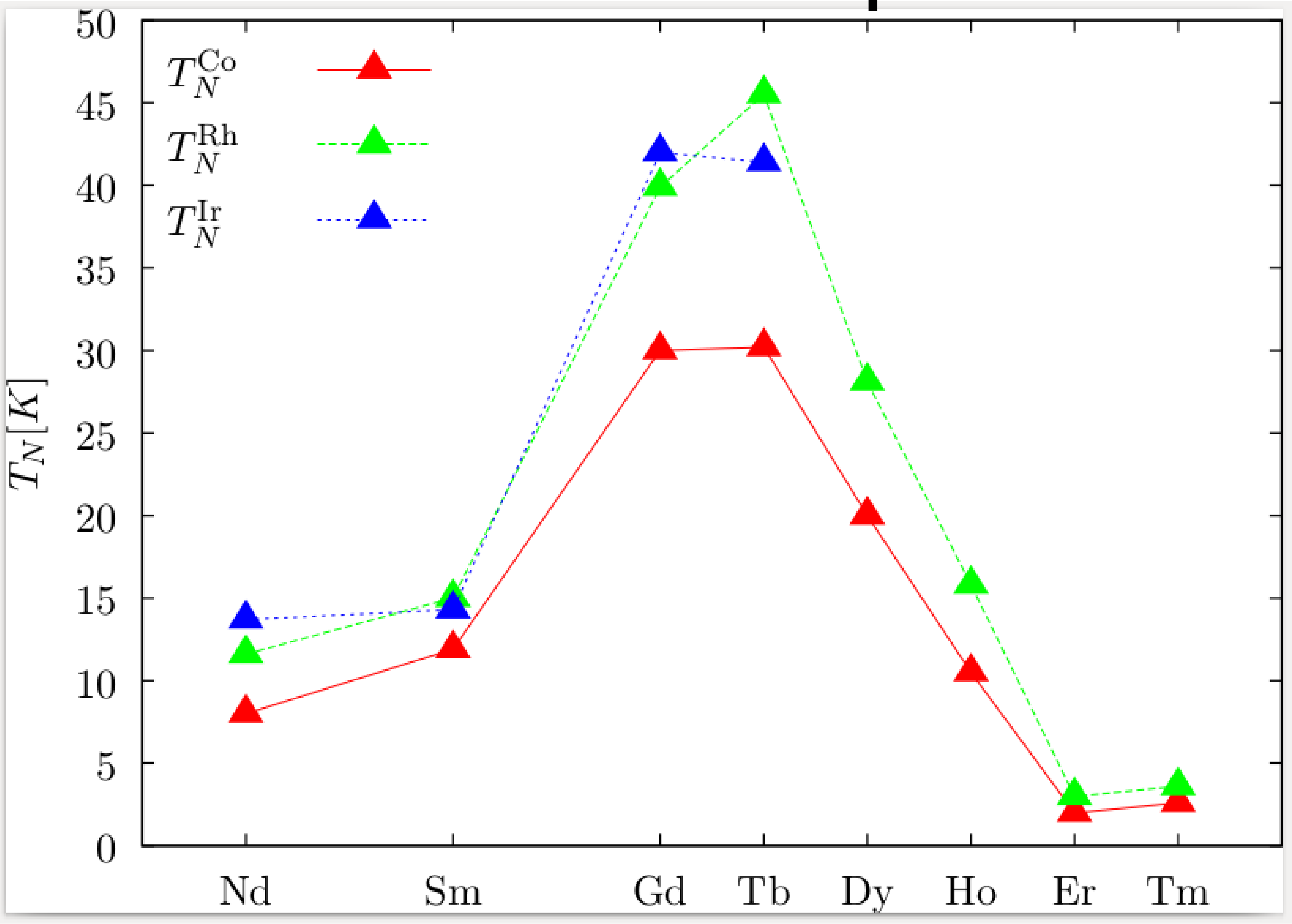
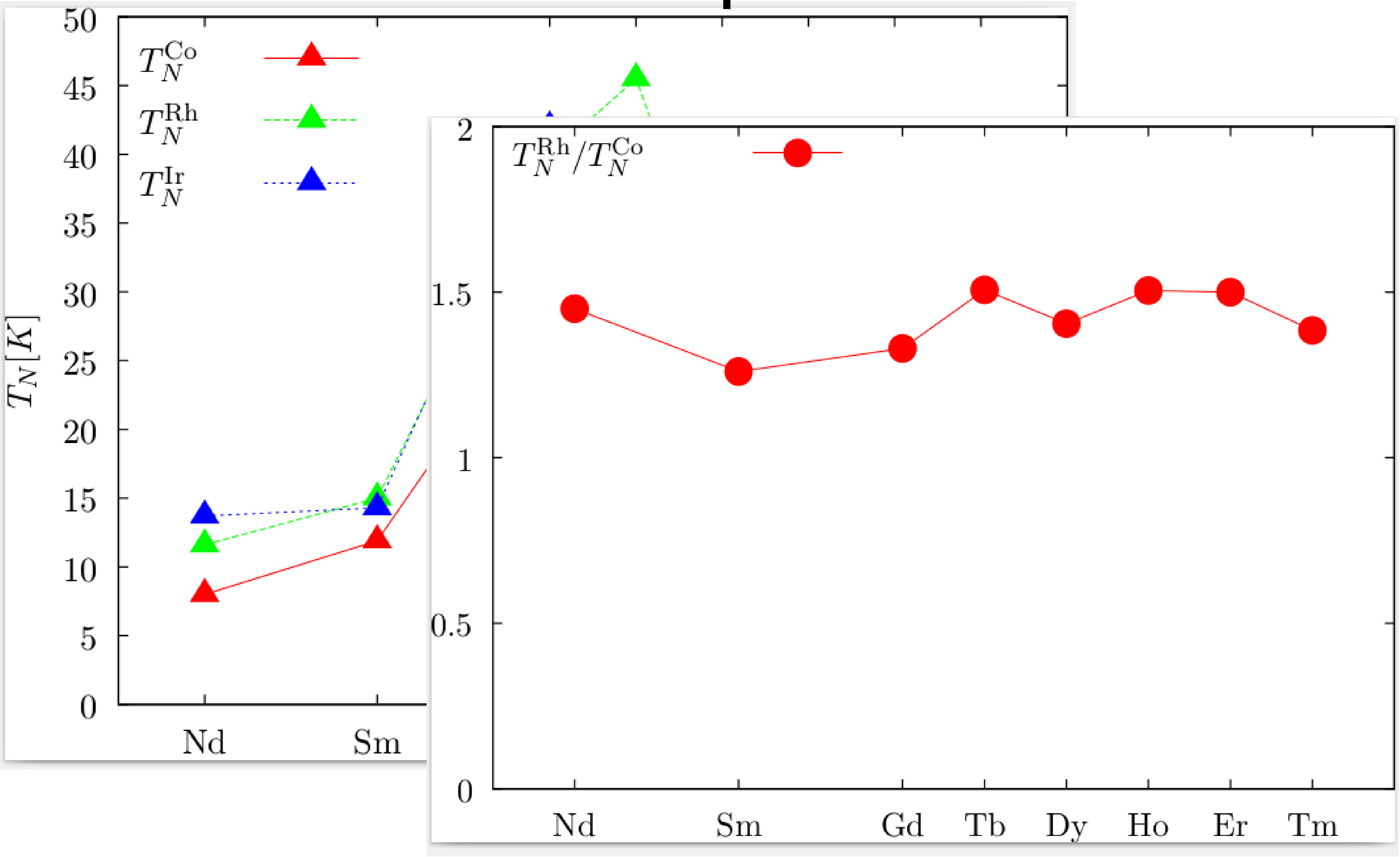
Néel temperatures



Néel temperatures



Variation of T_N dominated by the de Gennes factor

$GdMIn_5$

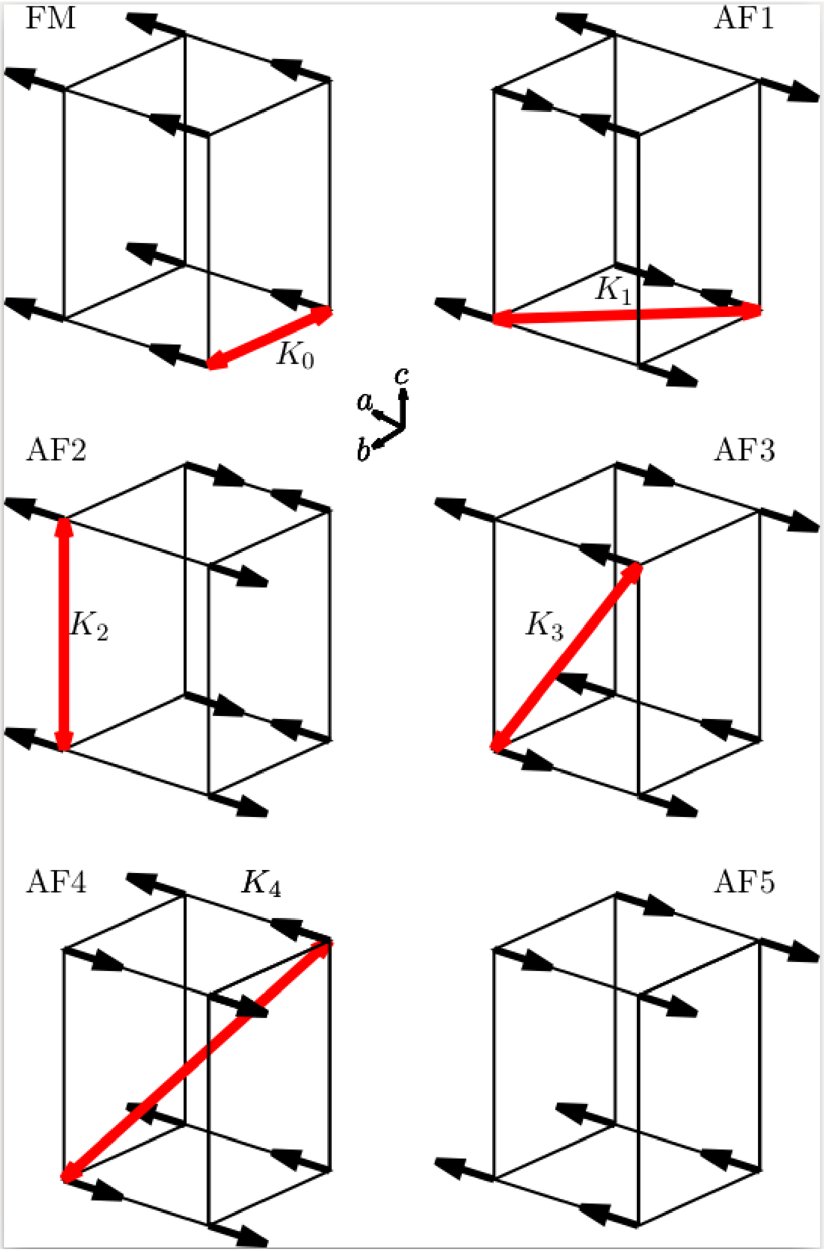
- Appealing compound to study the role of the transition metal M:
  - Gd^{3+} ions → S=7/2, L=0, J=7/2
  - We can neglect spin-orbit coupling effects.
  - Large magnetic moment takes the magnetic energy contribution to the total energy above the DFT (GGA+U) energy resolution.
  - No heavy fermion or superconducting behavior.

$$\mathcal{H} = \sum_{n.n.} K_0 J_i \cdot J_j + \sum_{n.n.n.} K_1 J_i \cdot J_j + \cdots$$

Relative energies (in K)

	$GdCoIn_5$	$GdRhIn_5$	$GdIrIn_5$
FM	126	145	149
AF1	62	65	56
AF2	59	95	74
AF3	0	0	0
AF4	23	50	44
AF5	125	133	128

$$\begin{aligned}
E^m_{FM}/J^2 &= 2K_0 + 2K_1 + K_2 + 4K_3 + 4K_4 \\
E^m_{AF1}/J^2 &= -2K_0 + 2K_1 - K_2 + 4K_3 - 4K_4 \\
E^m_{AF2}/J^2 &= -2K_0 + 2K_1 + K_2 - 4K_3 + 4K_4 \\
E^m_{AF3}/J^2 &= -2K_1 - K_2 + 4K_4 \\
E^m_{AF4}/J^2 &= -2K_1 + K_2 - 4K_4 \\
E^m_{AF5}/J^2 &= 2K_0 + 2K_1 - K_2 - 4K_3 - 4K_4
\end{aligned}$$



Mean field

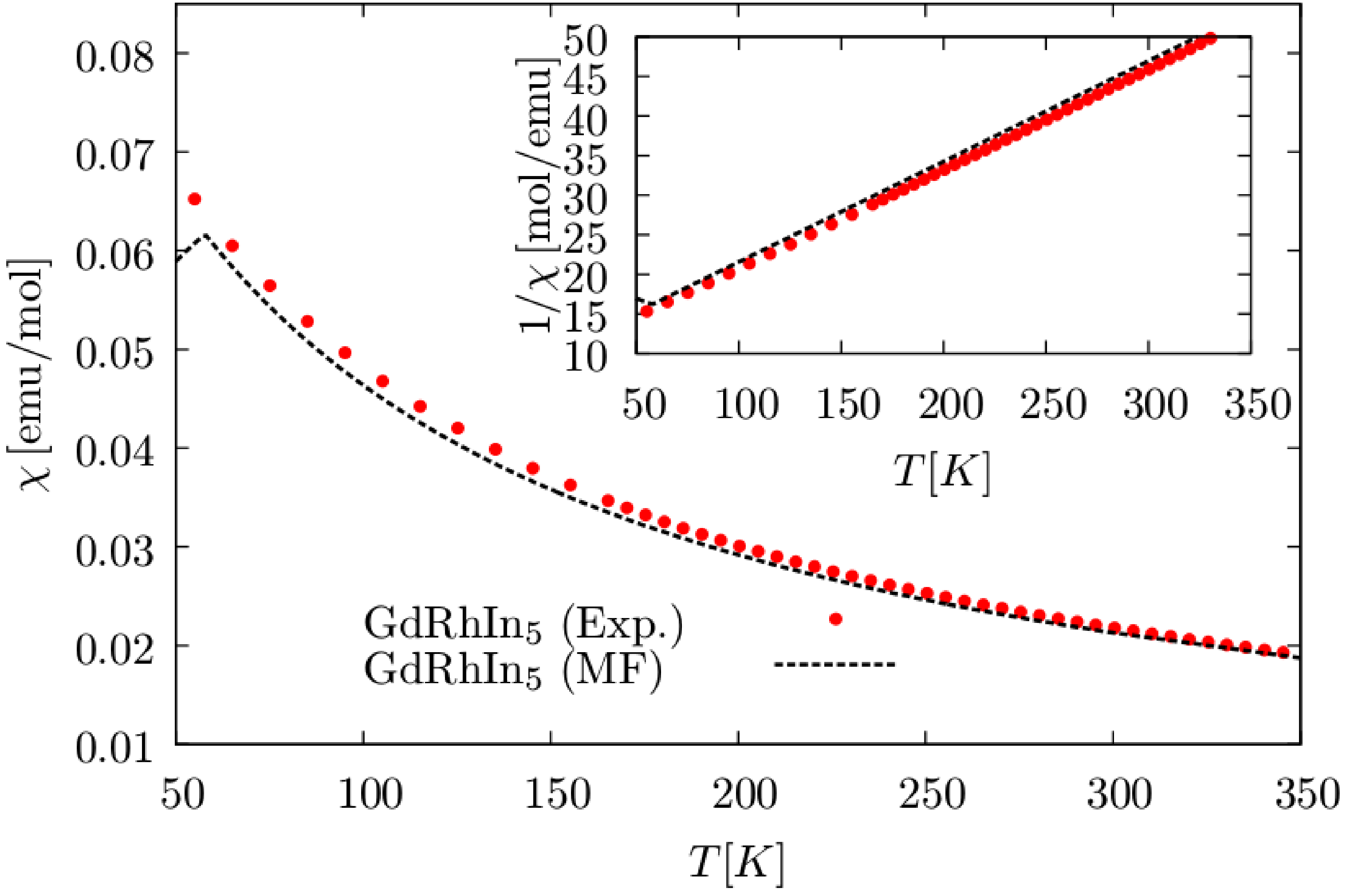
$$\mathcal{H} = \sum_{n.n.} K_0 J_i \cdot J_j + \sum_{n.n.n.} K_1 J_i \cdot J_j + \cdots$$

	$GdCoIn_5$	$GdRhIn_5$	$GdIrIn_5$
K_0	1.28	1.21	1.51
K_1	1.64	1.74	1.63
K_2	**0.49**	**1.43**	**1.30**
K_3	0.04	−0.01	0.02
K_4	−0.11	−0.15	−0.12

$$T_N^{MF} = \frac{J(J+1)}{3}(4K_1 + 2K_2 - 8K_4)$$

	$GdCoIn_5$	$GdRhIn_5$	$GdIrIn_5$
T_N^{MF}	44.4	57.6	52.9
T_N^{exp}	30	39	40

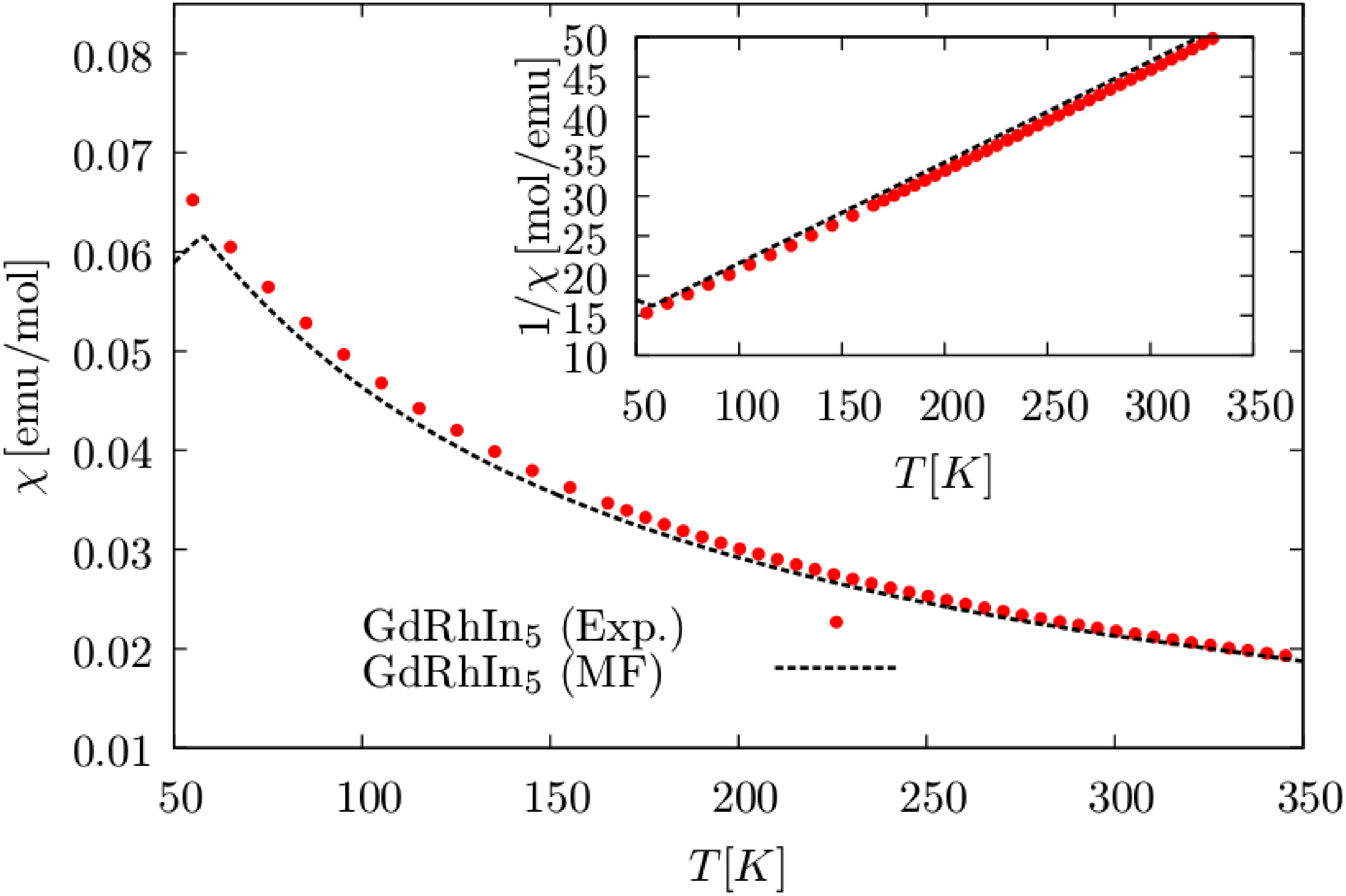
Mean field



$$\chi \sim \frac{C}{T-\theta}$$

$$\theta = \frac{J(J+1)}{3}\left(4K_0 + 4K_1 + 2K_2 + 8K_3 + 8K_4\right)$$

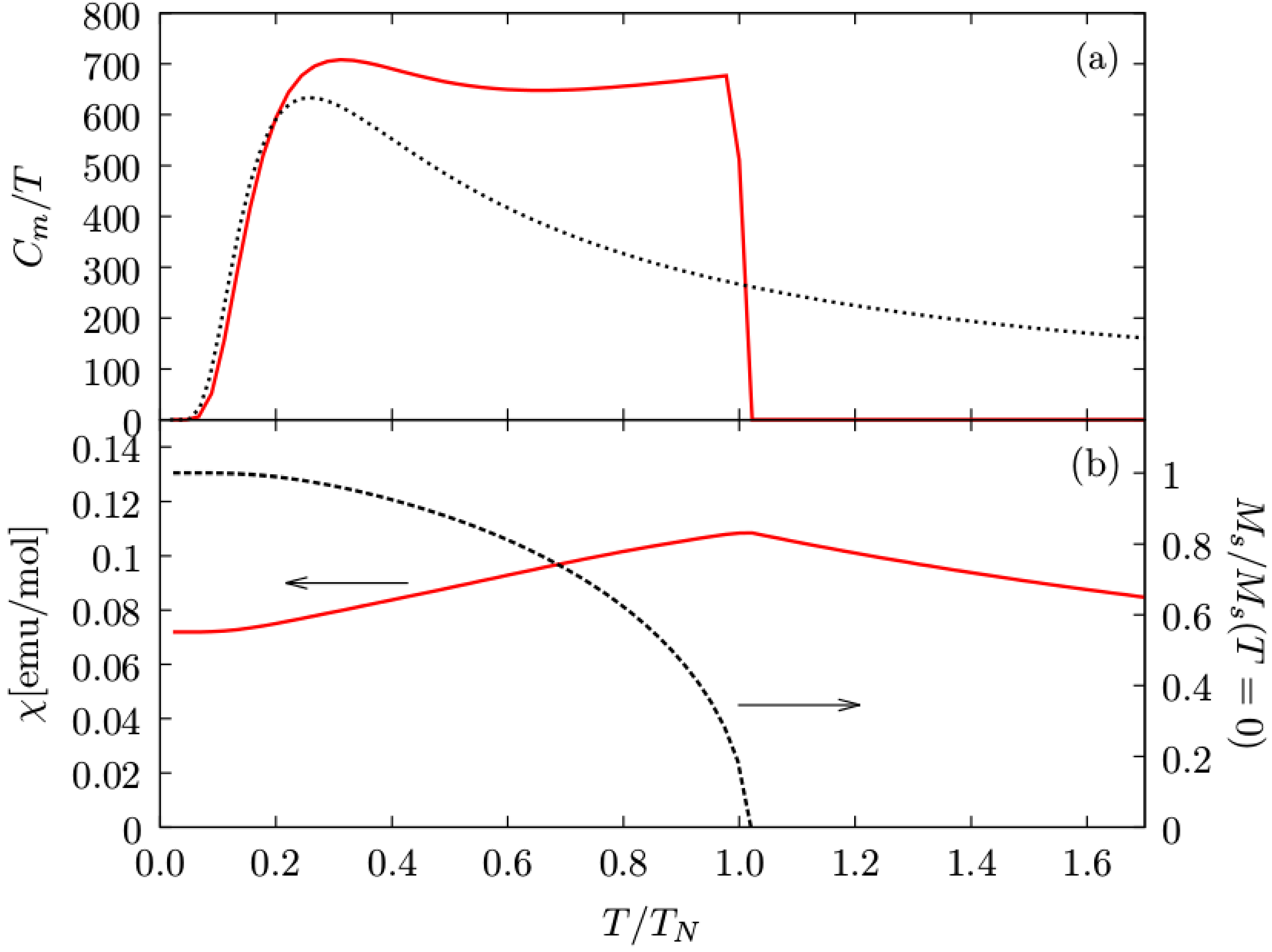
Mean field



$$\chi \sim \frac{C}{T-\theta}$$

	$GdCoIn_5$	$GdRhIn_5$	$GdIrIn_5$
θ θ^{exp}	63.3 ~ 50	66.5 69^a, 63.8^b	75.2 64^a

Mean field


(a)
(b)
C_m/T
800
700
600
500
400
300
200
100
0
χ[emu/mol]
0.14
0.12
0.1
0.08
0.06
0.04
0.02
0
$M_s/M_s(T=0)$
1
0.8
0.6
0.4
0.2
0
0.0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
1.6
T/T_N

Quantum Monte Carlo

- Sign problems for the full magnetic Hamiltonian.
- Let's Include quantum fluctuations to the mean field solution.

$$T_N^{MF} = \frac{J(J+1)}{3} \underbrace{(4K_1 + 2K_2 - 8K_4)}_{zK_{eff}}$$

- We perform the Quantum Monte Carlo calculations in a cubic lattice with an effective nearest neighbor coupling K_{eff}.
- Systems with up to 30^3 sites.

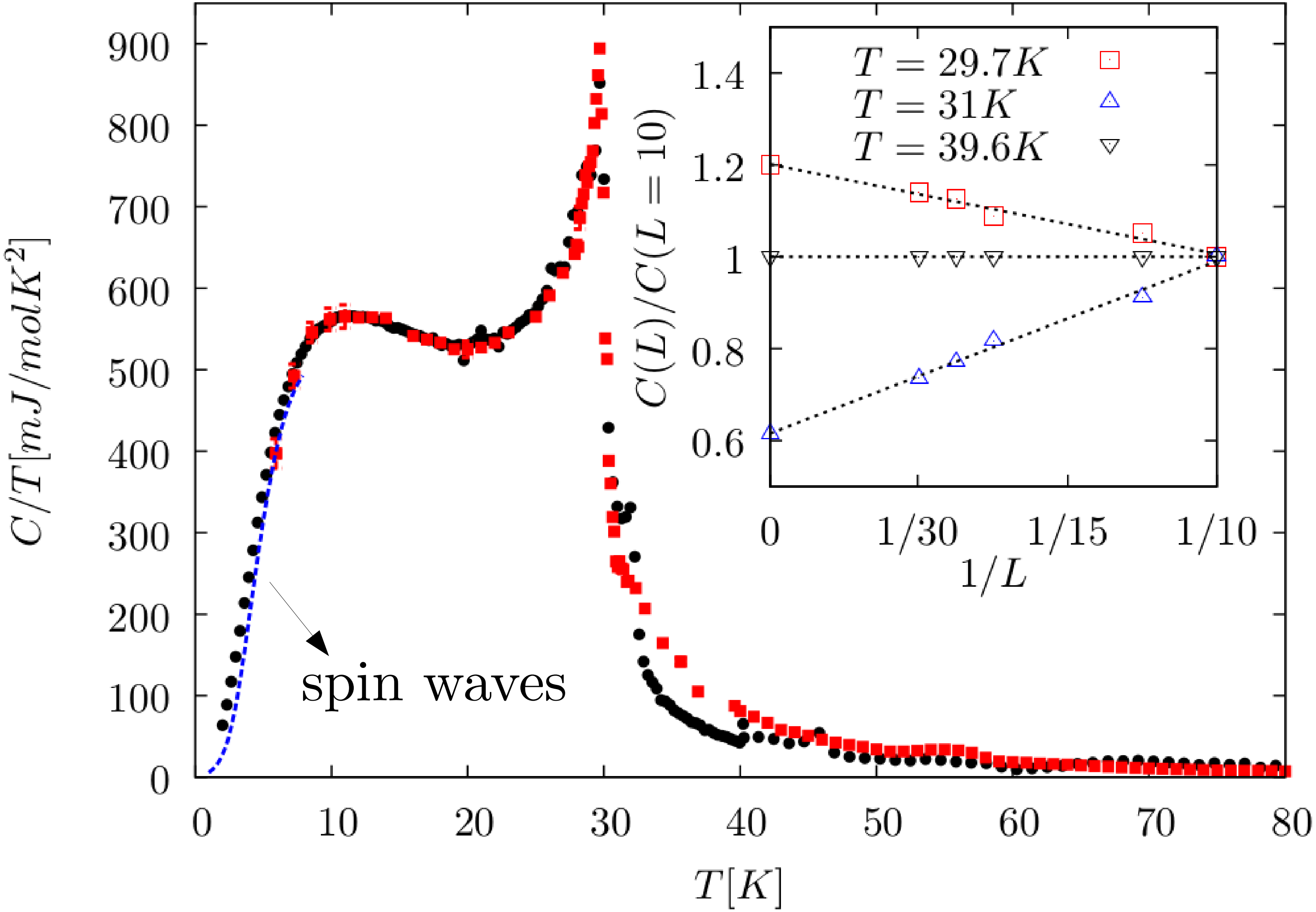
ALPS Library, Anders W. Sandvik, Phys. Rev. B 59, R14157(R) (1999)

QMC results

- Quantum fluctuations reduce the Néel temperature.
- A better agreement with the experimental results is obtained:

	$\mathrm{GdCoIn_5}$	$\mathrm{GdRhIn_5}$	$\mathrm{GdIrIn_5}$
T_N^{MF}	44.4	57.6	52.9
T_N^{QMC}	32.3	41.9	38.4
T_N^{exp}	30	39	40

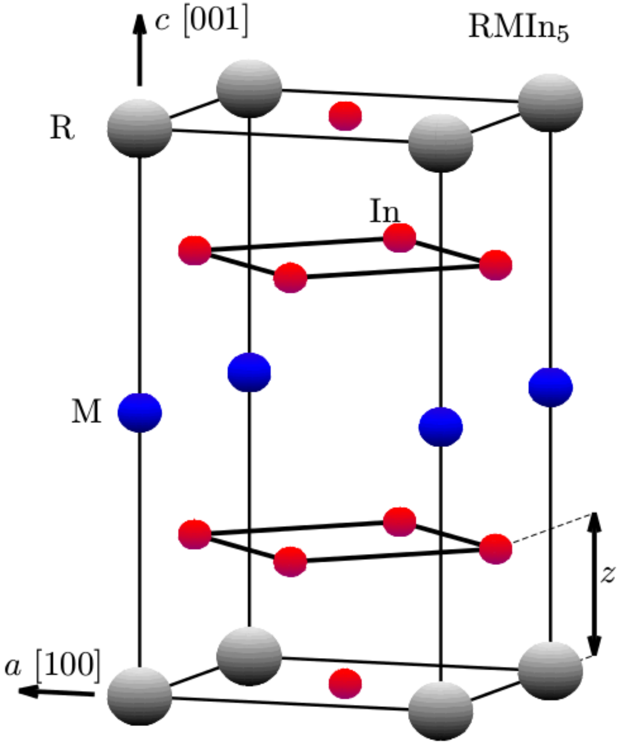
QMC results



$C/T[mJ/molK^2]$
900
800
700
600
500
400
300
200
100
0
0
10
20
30
40
50
60
70
80
$T[K]$
spin waves
$C(L)/C(L=10)$
1.4
1.2
1
0.8
0.6
0
1/30
1/15
1/10
$1/L$
$T = 29.7K$
$T = 31K$
$T = 39.6K$

Toy model for the K_2 coupling

- Gd 4f orbitals not hybridized with the conduction electrons.
- Gd 5d bands almost unoccupied.
- M d bands partially filled (~4 electrons).
- Wannier orbital analysis:Largest hybridization between Gd 5d orbitals and M d orbitals.



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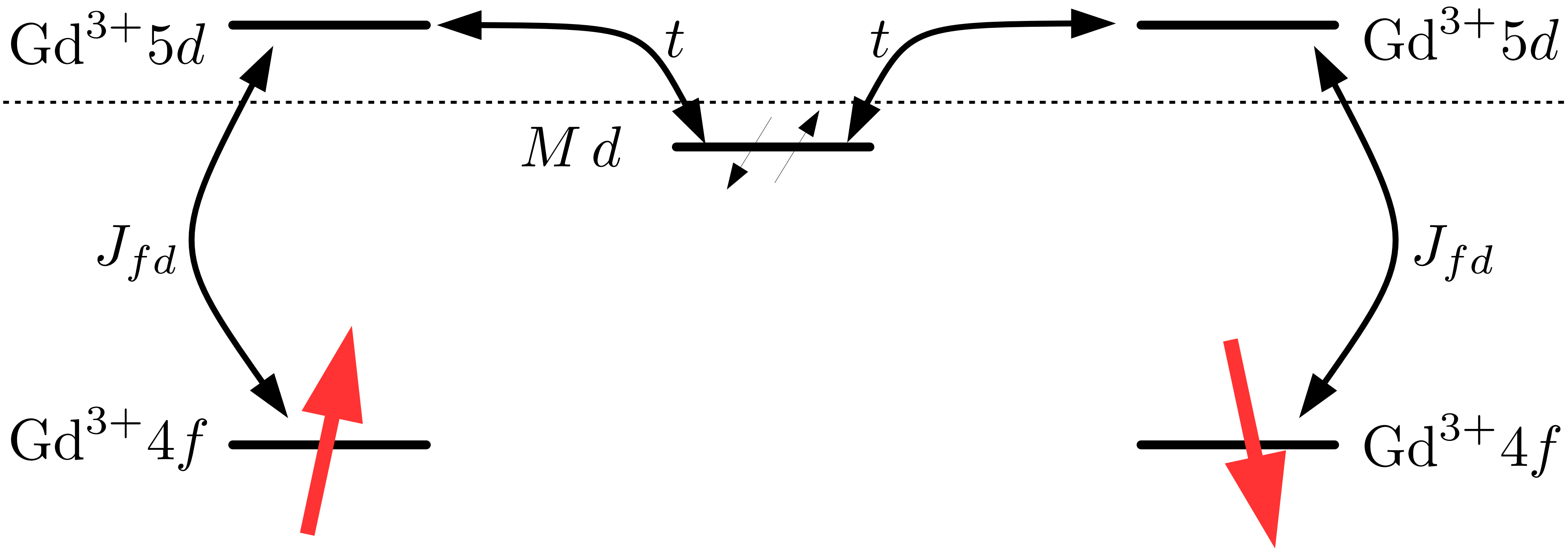
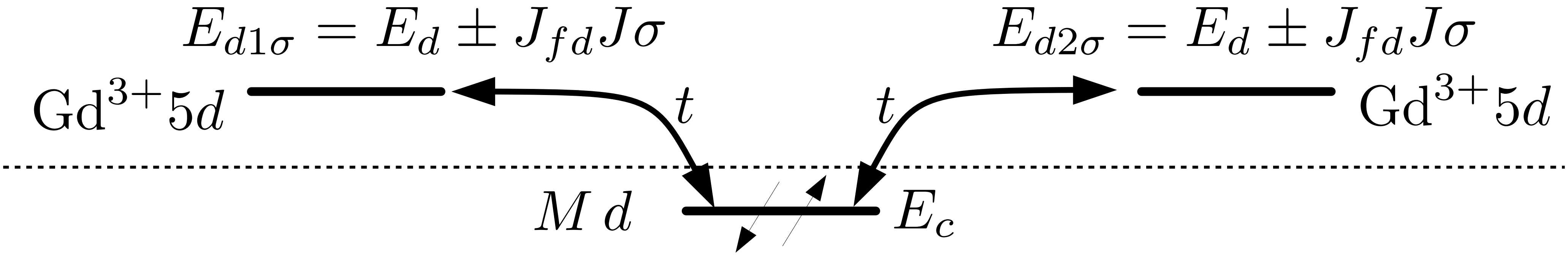


Toy model for the K_2 coupling

- Consider 4f local magnetic moments as static



$E_{d1\sigma} = E_d \pm J_{fd} J \sigma$

$E_{d2\sigma} = E_d \pm J_{fd} J \sigma$

$\mathrm{Gd}^{3+} 5d$

t

t

$\mathrm{Gd}^{3+} 5d$

$M\, d$

E_c

$$H_2 = \sum_{i=1,2} \sum_{\sigma} E_{di\sigma} d^{\dagger}_{i\sigma} d_{i\sigma} + E_c \sum_{\sigma} c^{\dagger}_{\sigma} c_{\sigma} + t \sum_{i} \sum_{\sigma} (d^{\dagger}_{i\sigma} c_{\sigma} + H.c.)$$

Toy model for the K_2 coupling

$$H_2 = \sum_{i=1,2} \sum_{\sigma} E_{di\sigma} d^\dagger_{i\sigma} d_{i\sigma} + E_c \sum_{\sigma} c^\dagger_\sigma c_\sigma + t \sum_{i} \sum_{\sigma} (d^\dagger_{i\sigma} c_\sigma + H.c.)$$

$$K_2 \sim E(\uparrow, \downarrow) - E(\uparrow, \uparrow)/2J^2$$

$$K_2 \sim \frac{2J_{fd}^2 t^4}{(E_c - E_d)^5} \quad \text{to lowest order in } t$$

$$E_d \sim 3eV \qquad t^{Co} \sim 0.6t^{Rh,Ir}$$

$$E_c^{Rh} \sim E_c^{Ir} \sim -2.5eV \qquad E_d^{Co} \sim -1.1eV$$

Toy model for the K_2 coupling

$$H_2 = \sum_{i=1,2} \sum_{\sigma} E_{di\sigma} d^{\dagger}_{i\sigma} d_{i\sigma} + E_c \sum_{\sigma} c^{\dagger}_{\sigma} c_{\sigma} + t \sum_{i} \sum_{\sigma} (d^{\dagger}_{i\sigma} c_{\sigma} + H.c.)$$

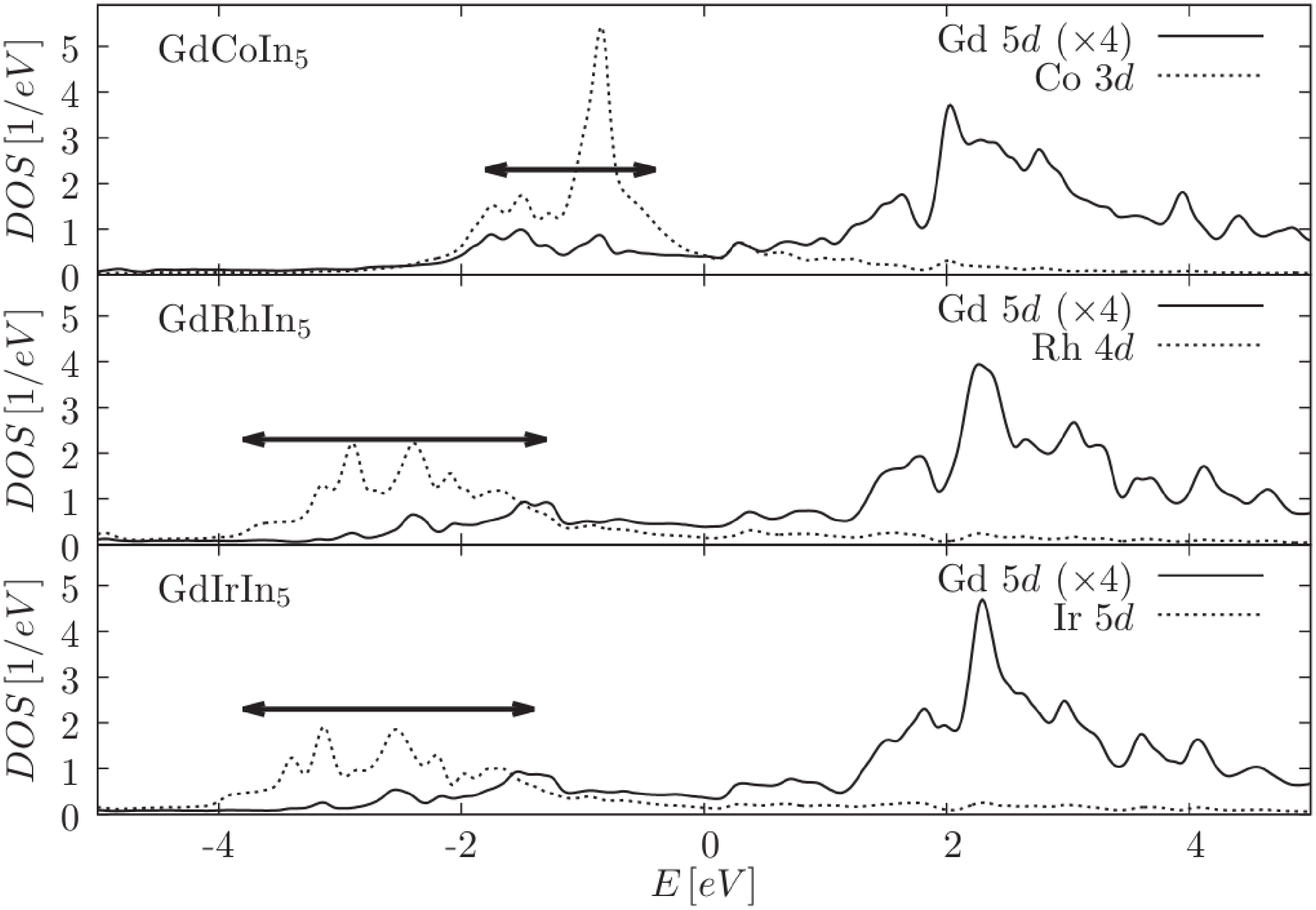
$$K_2 \sim E(\uparrow,\downarrow) - E(\uparrow,\uparrow)/2J^2$$

$$K_2 \sim \frac{2J^2_{fd} t^4}{(E_c - E_d)^5} \quad \text{to lowest order in } t$$

$$E_d \sim 3eV \qquad \boxed{t^{Co} \sim 0.6 t^{Rh,Ir}}$$

$$E^{Rh}_c \sim E^{Ir}_c \sim -2.5eV \qquad E^{Co}_d \sim -1.1eV$$

Parameter estimation

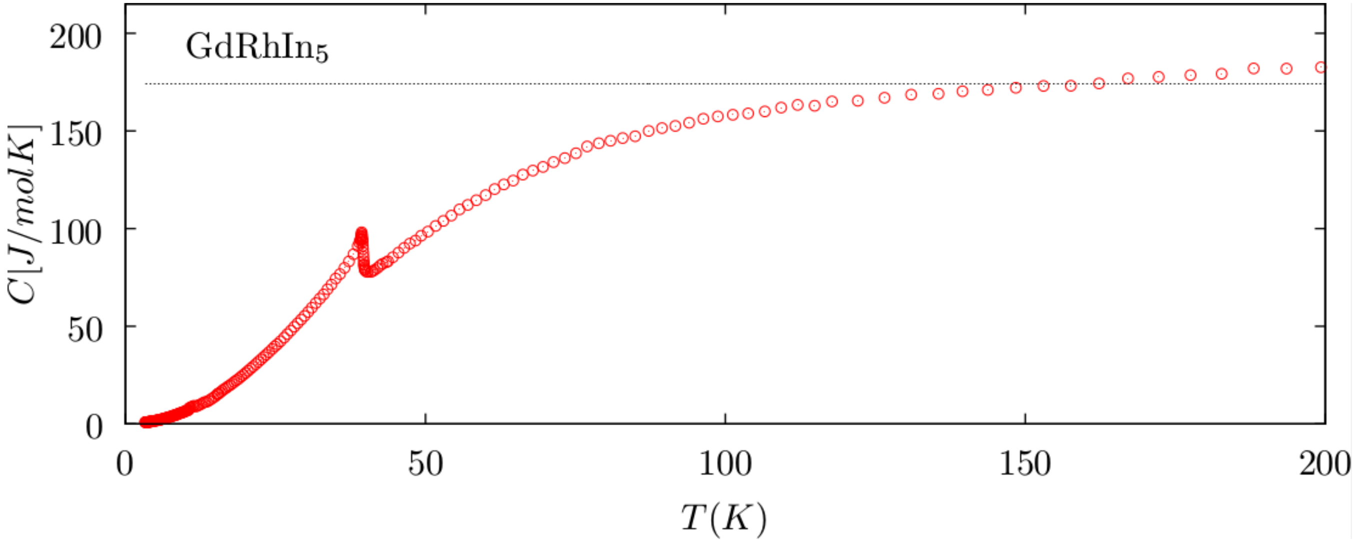


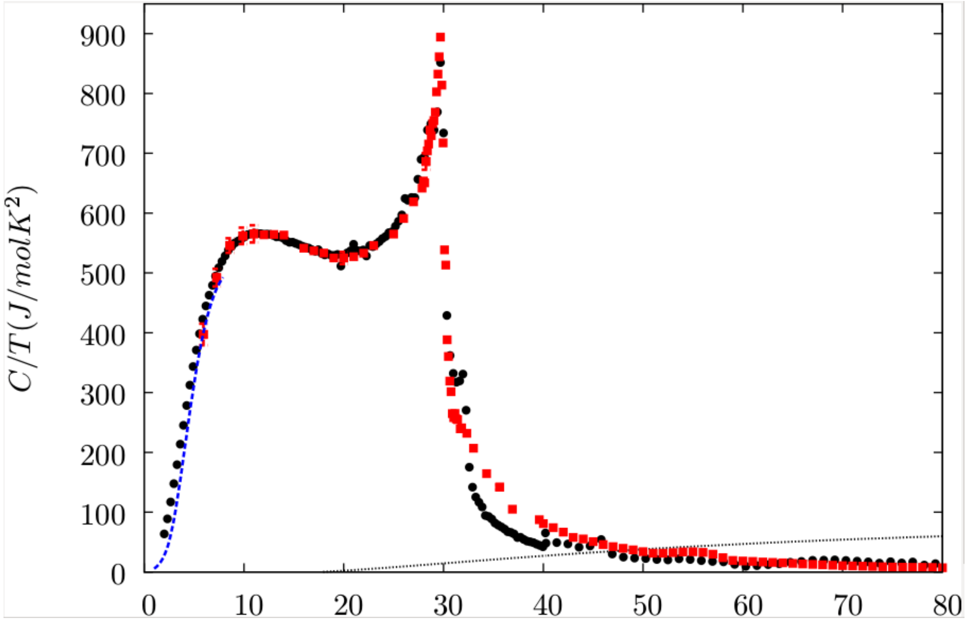
Magnetic contribution to the specific heat (phonon specific heat subtraction)

- We calculated the phonon contribution to the specific heat using density functional theory (GGA+U) within a frozen phonon approximation.
- Experiments performed at constant pressure and finite temperatures.
- Calculations done at zero temperature and fixed volume.
- Anharmonic effects?
- Using thermodynamic relations and some simplifying assumptions we get (see e.g. Wallace book):

$$C_p \sim C_{vH}(1 + cT)$$

Phonon contribution subtraction and anharmonicity



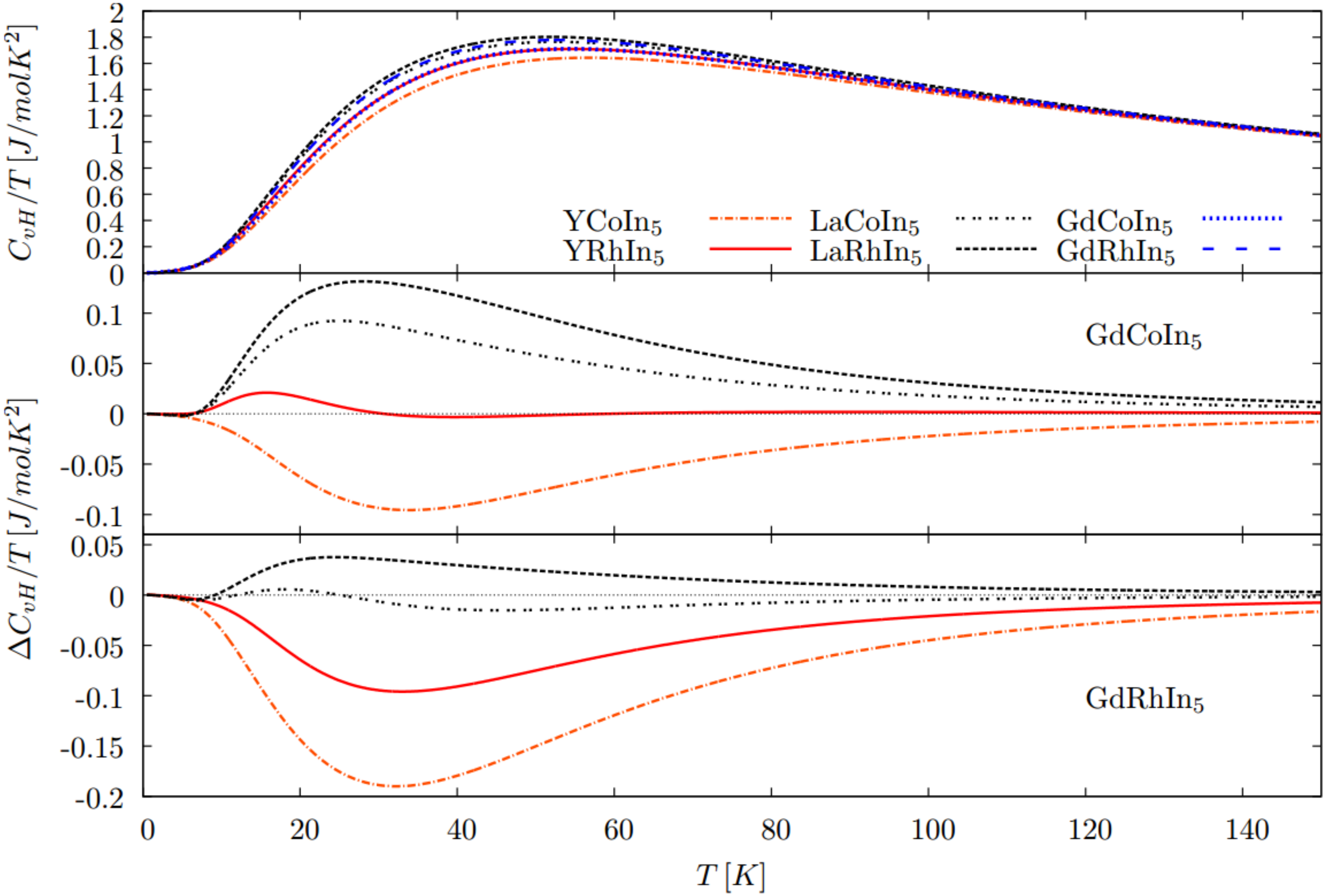


$$C_p \sim C_{vH}(1 + cT)$$

Magnetic contribution to the specific heat (phonon specific heat subtraction)

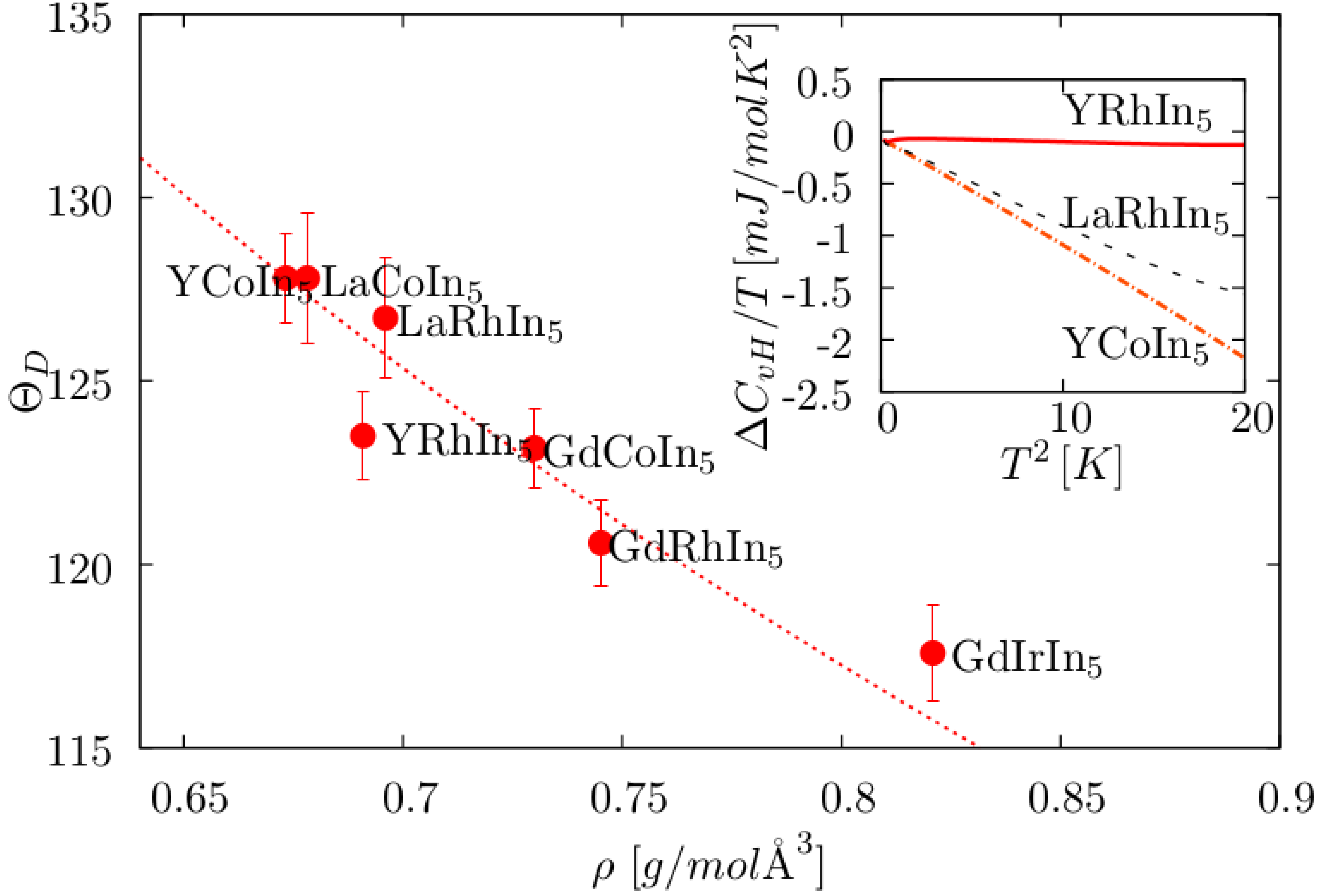
- Experimentally: subtraction of the specific heat of an isostructural non-magnetic compound.
- Usual non-magnetic analogues: $YMIn_5$ and $LaMIn_5$.
- Why should this subtraction work?
- We calculated the phonon contribution to the specific heat of a variety of compounds using density functional theory (GGA+U) within a frozen phonon approximation.

Phonon contribution to the specific heat

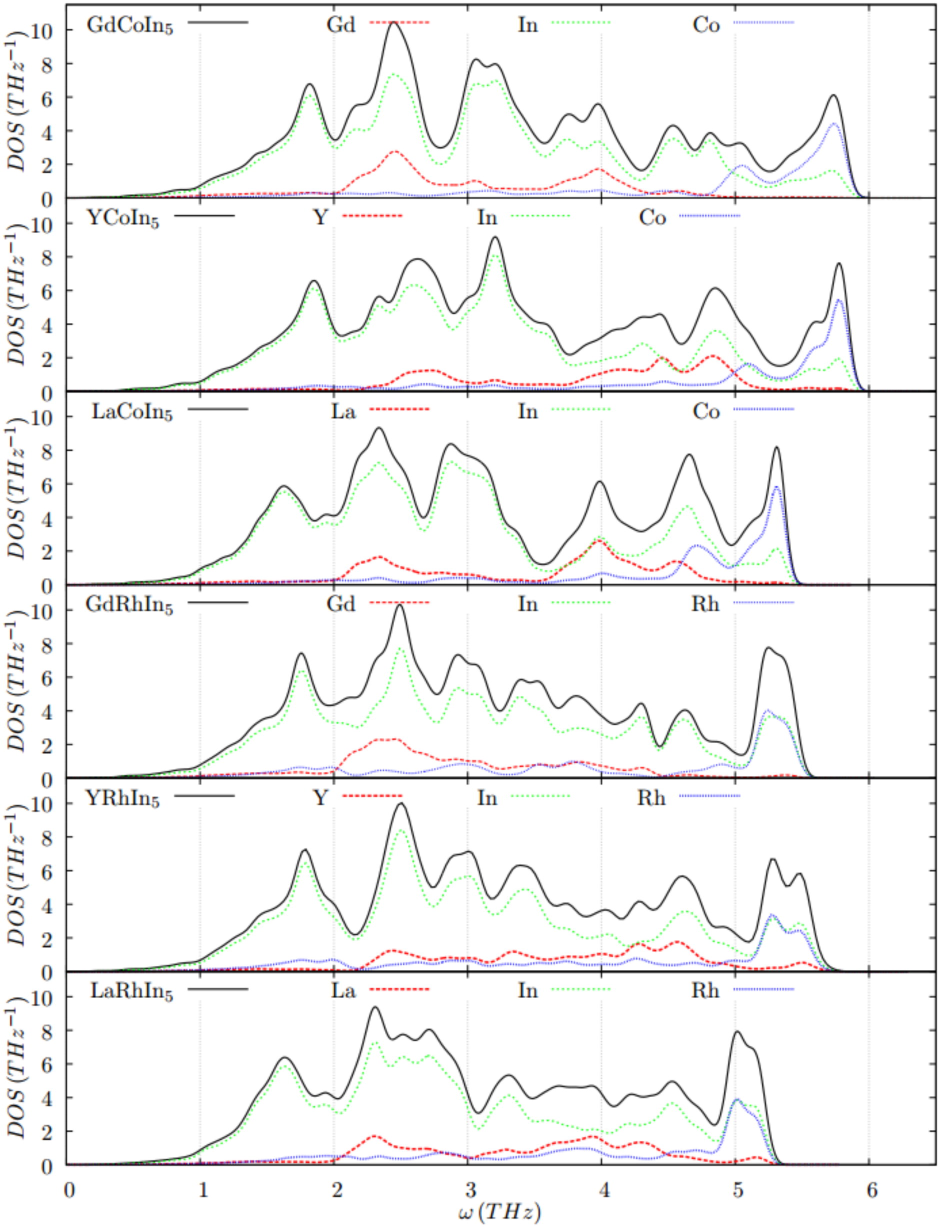


Jorge I. Facio *et al.*, J. Magn. Magn. Mater. **407**, 406 (2016)

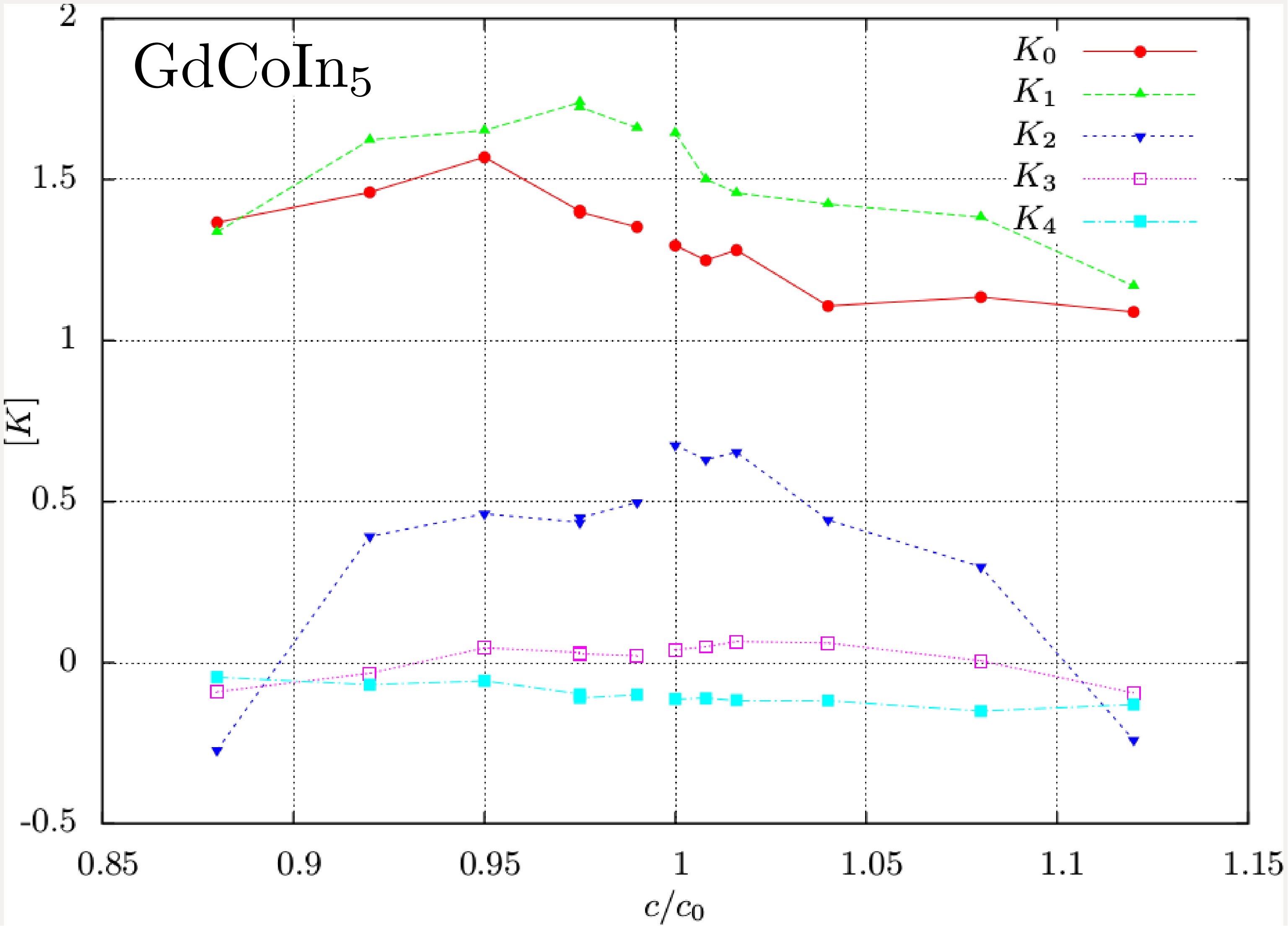
Debye temperatures

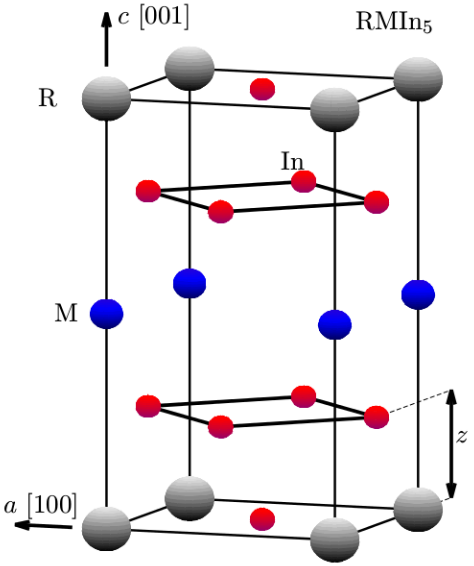


Phonon spectral density



c-axis compression and expansion





Work in progress....

Conclusions

- Reduced interplane magnetic coupling K_2 dominates reduction of Néel temperature in Co compounds compared to Rh and Ir compounds.
- Reduction of K_2 is mainly due to a reduced hybridization between Gd 5d and Co 3d orbitals.
- More 2D magnetic behavior in Co compounds but no clear signature of a more 2D behavior observed in the *electronic structure.*
- Best non-magnetic compound to extract phonon specific heat in Gd compounds depends on the transition metal.
- Future work: include spin-orbit coupling effects (Tb compounds)

D. Betancourth *et al.*, J. Magn. Magn. Mater. **374**, 744 (2015)
Jorge I. Facio *et al.*, Phys. Rev. B **91**, 014409 (2015)
Jorge I. Facio *et al.*, J. Magn. Magn. Mater. **407**, 406 (2016)