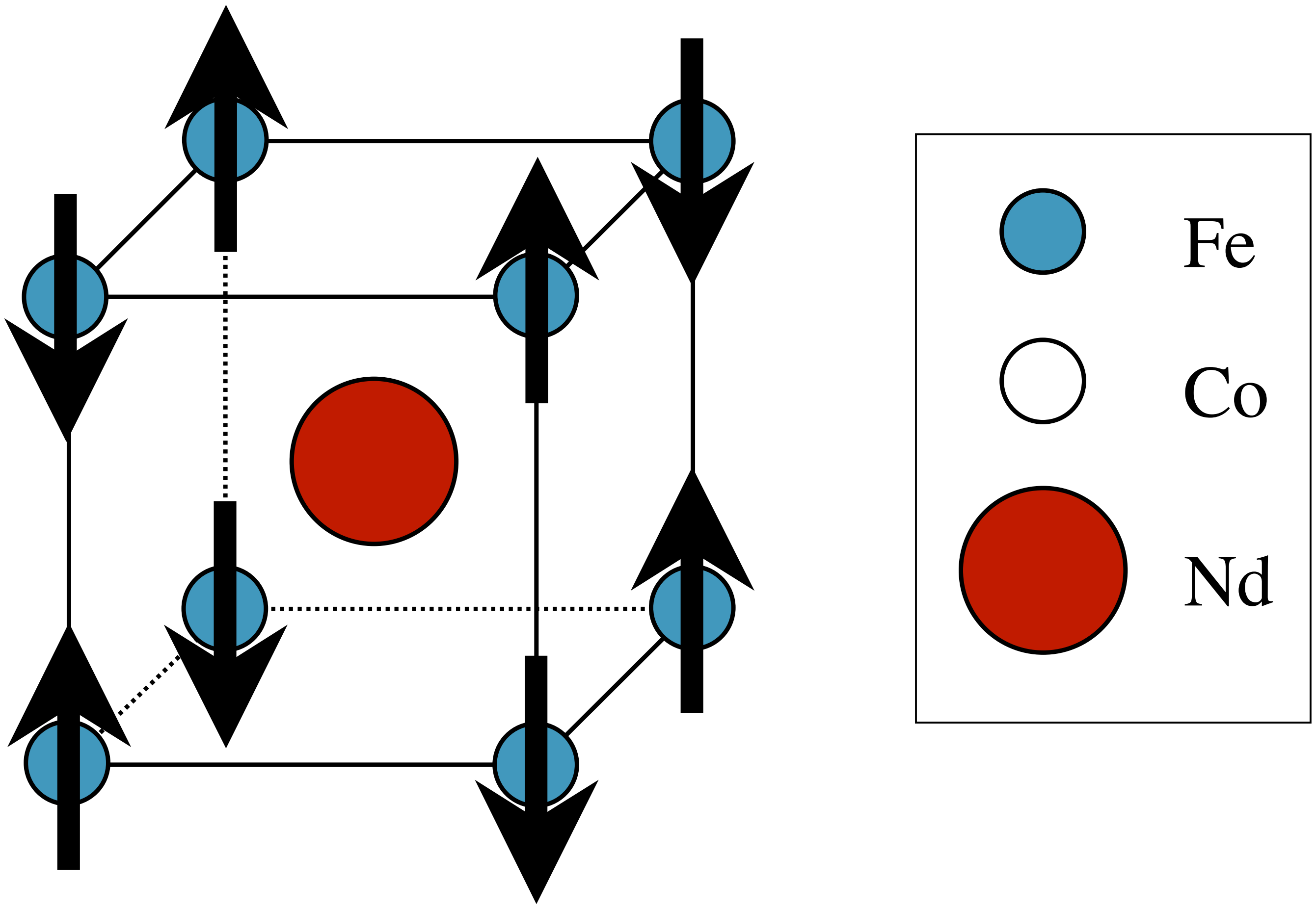
x > 0 : distribution of internal fields

Relevant parameter: G = level of uncompensation

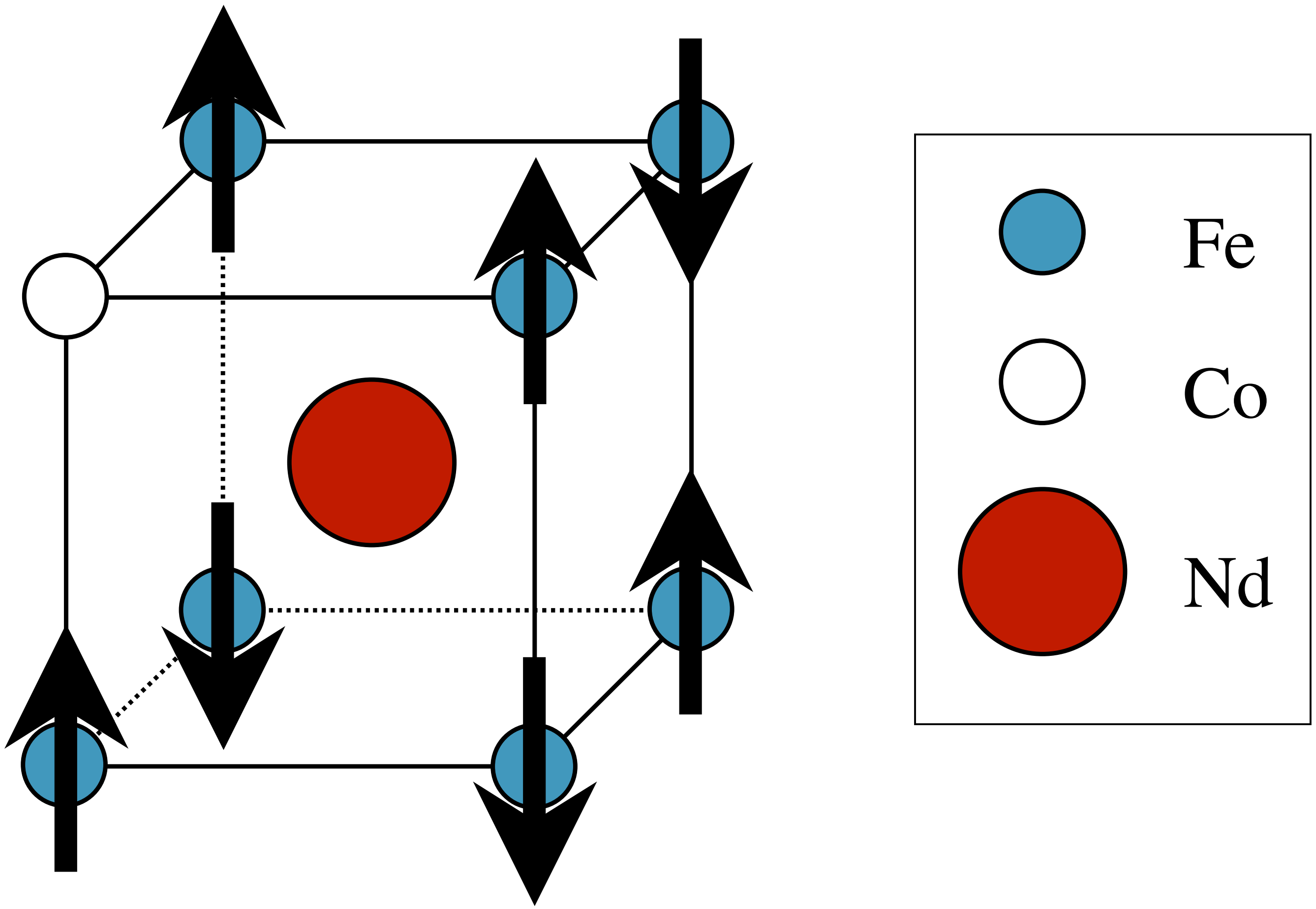
G = 0



x > 0 : distribution of internal fields

Relevant parameter: G = level of uncompensation

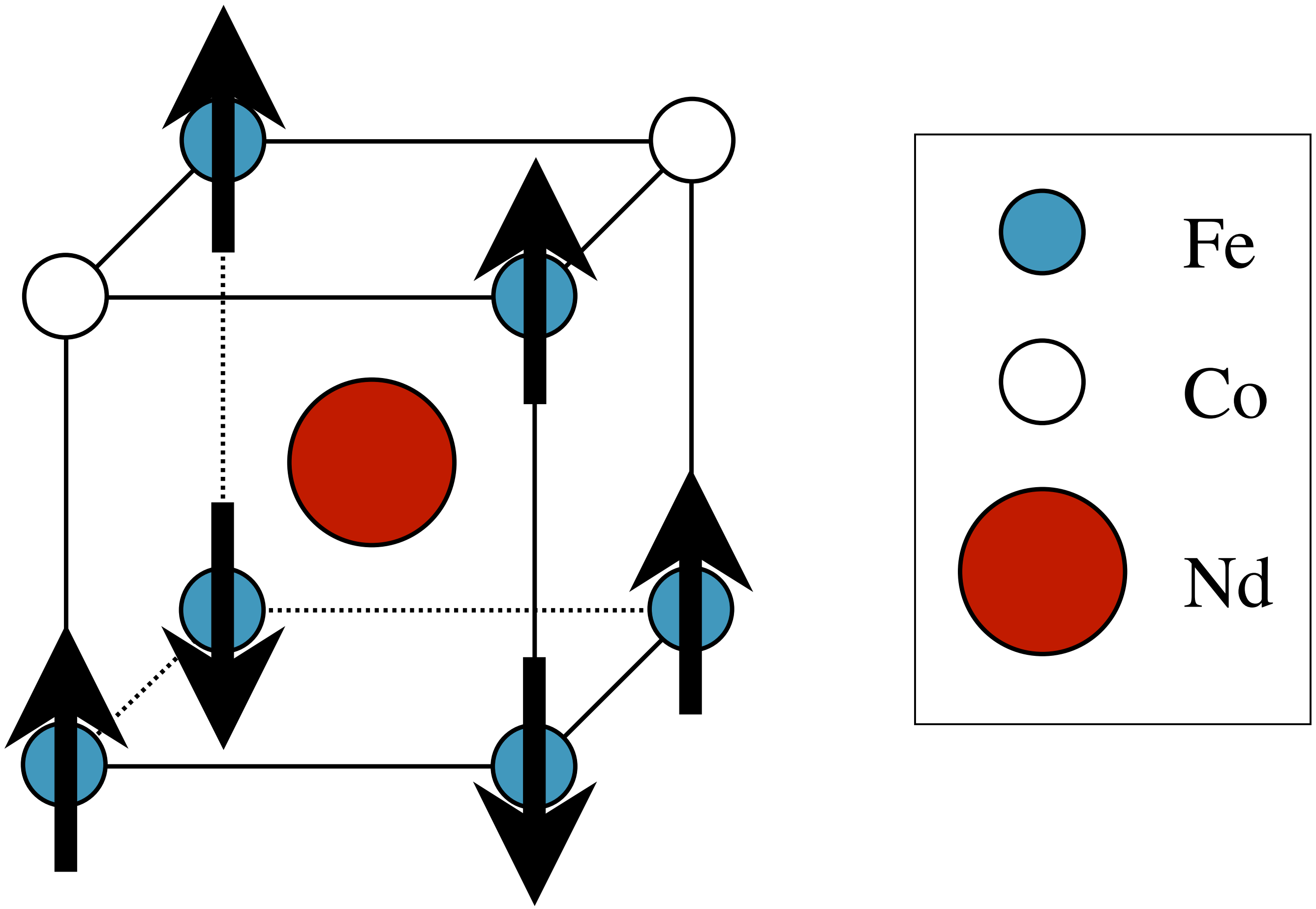
G = 1



x > 0 : distribution of internal fields

Relevant parameter: G = level of uncompensation

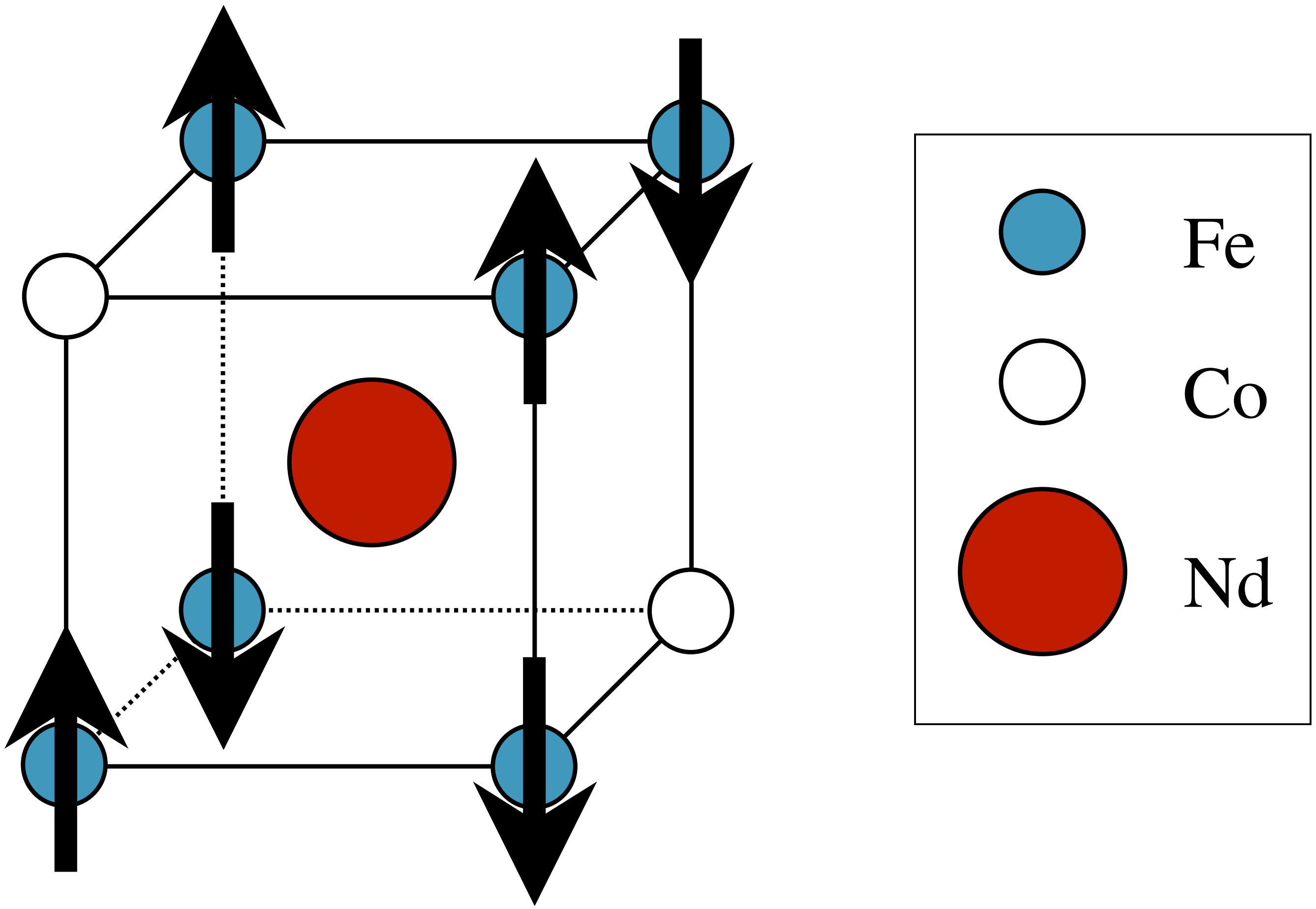
G = 2



x > 0 : distribution of internal fields

Relevant parameter: G = level of uncompensation

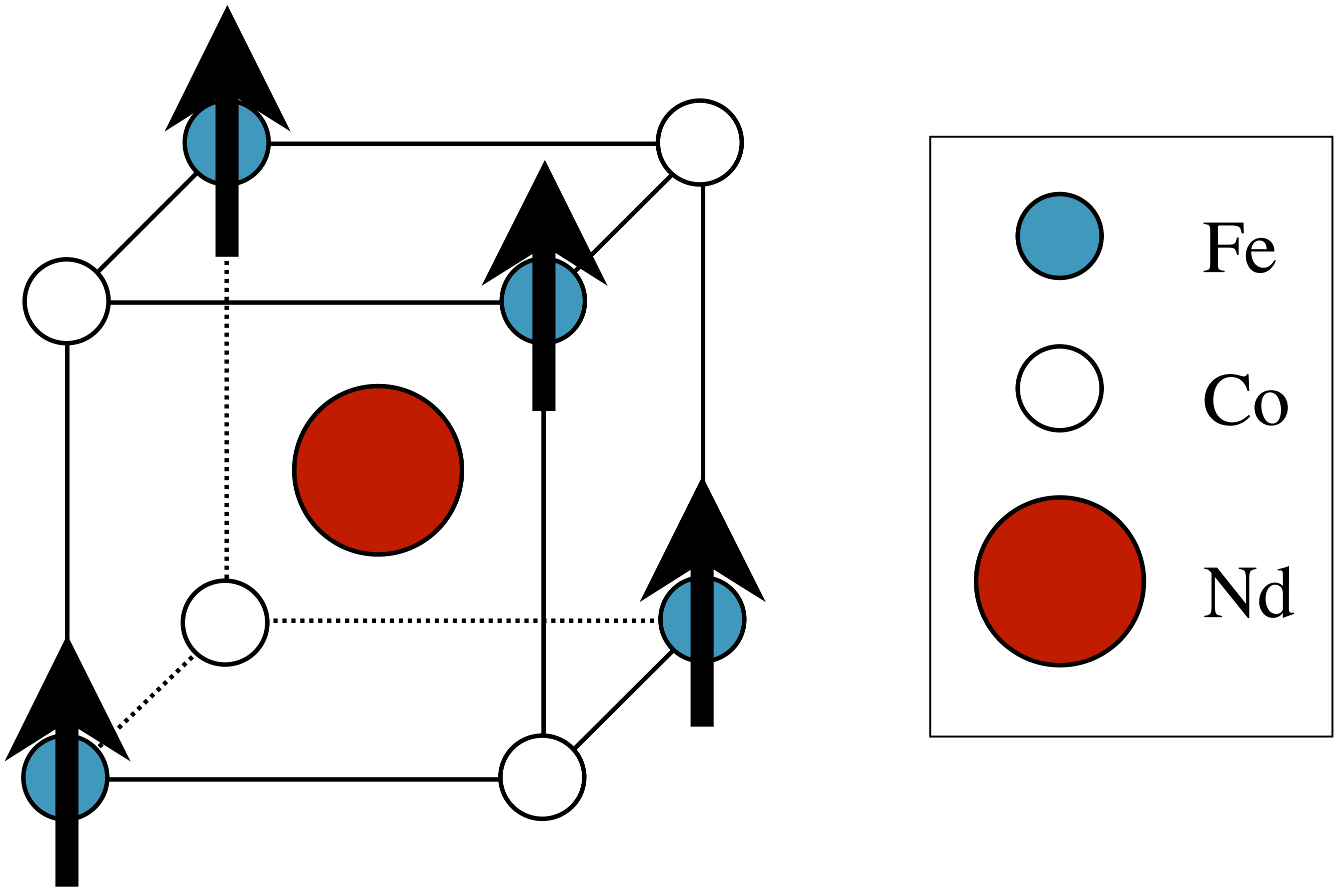
G = 0



x > 0 : distribution of internal fields

Relevant parameter: G = level of uncompensation

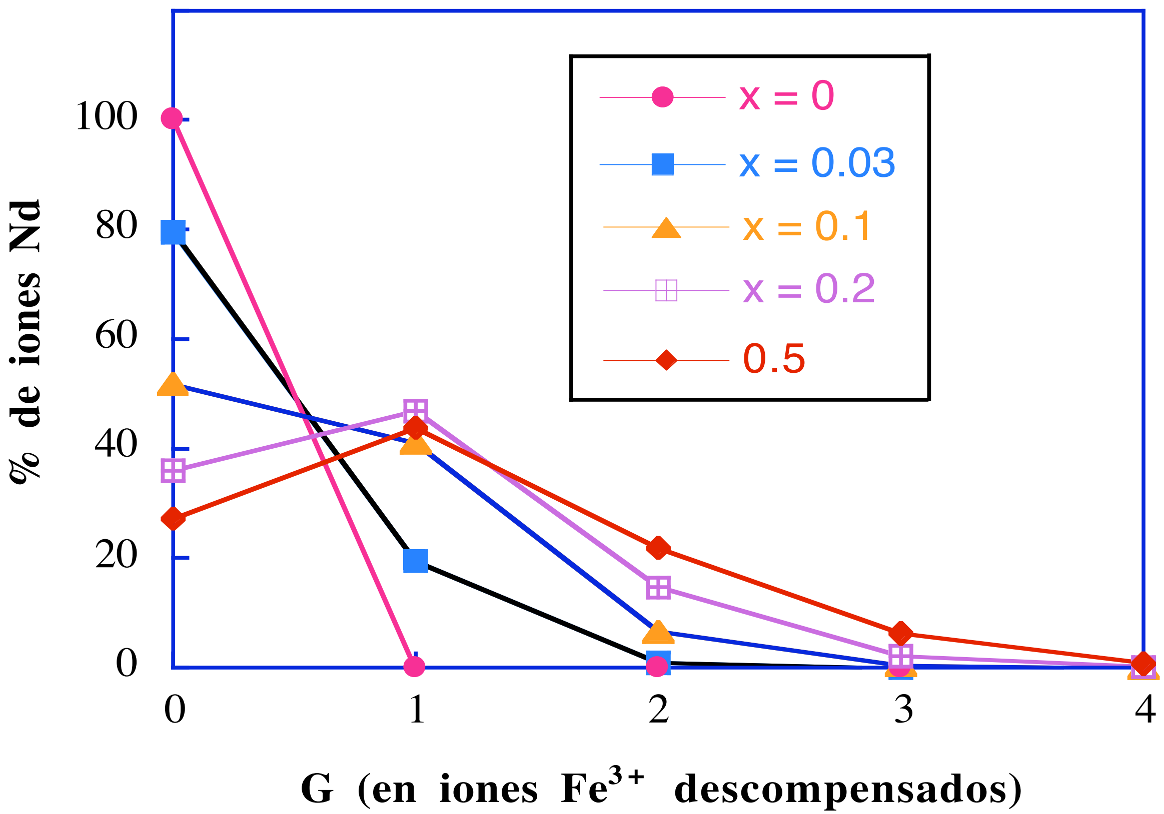
G = 4



x > 0 : distribution of internal fields

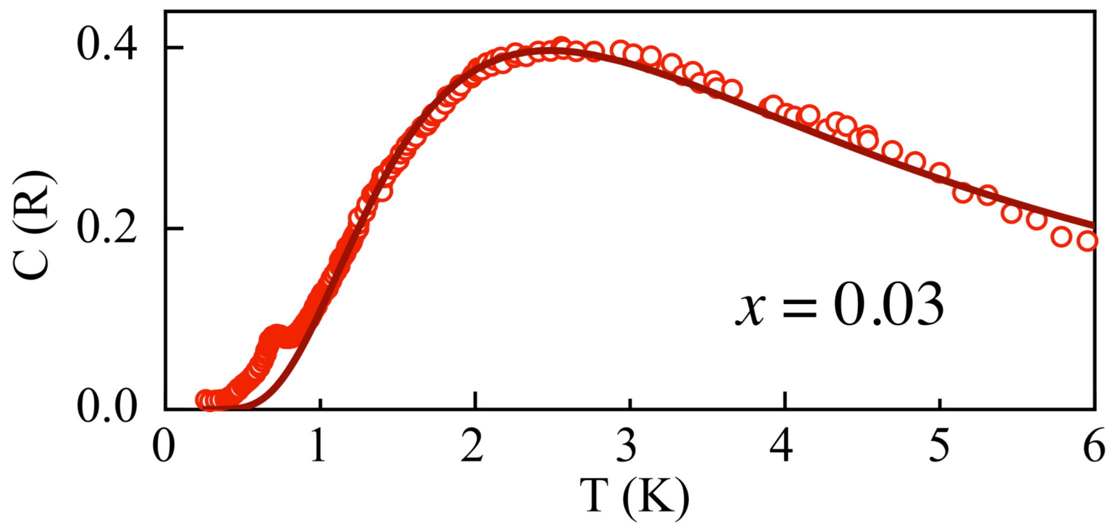
Relevant parameter: level of uncompensation

$$C_p = \sum_{G=0}^{4} p_G \, \mathrm{Sch}(\Delta_G)$$



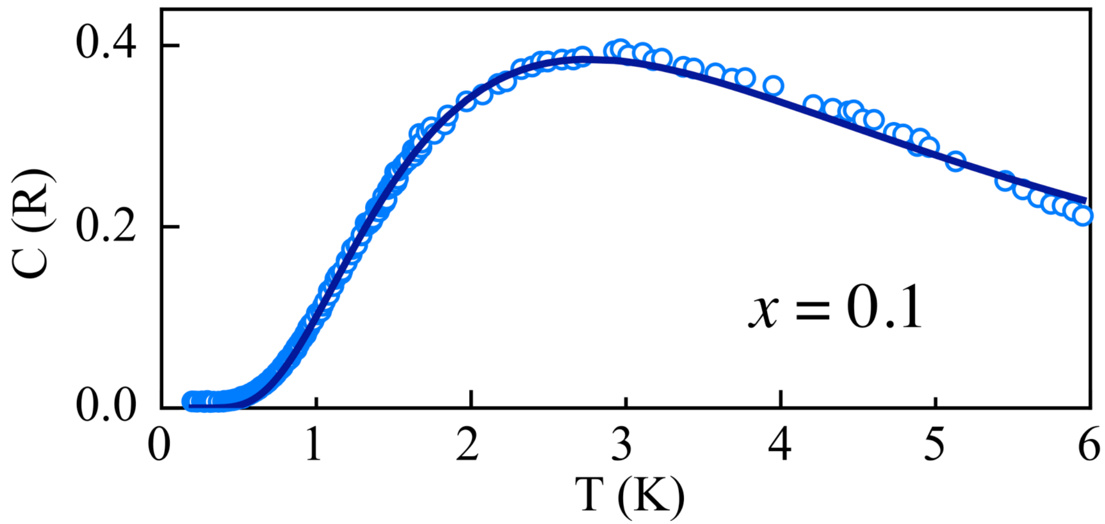
x > 0 : sum of Schottky contributions

E/k_B (K)	$x = 0.03$	$x = 0.1$	$x = 0.25$	$x = 0.5$
Δ_0	5.31(1)	4.93(6)	4.05(4)	3.32(3)
Δ_1	10.4(1)	8.15(7)	7.65(5)	7.19(4)
Δ_2		12.1(5)	10.2(3)	15.6 (2)
Δ_3				34.6(1.2)



x > 0 : sum of Schottky contributions

E/k_B (K)	$x = 0.03$	$x = 0.1$	$x = 0.25$	$x = 0.5$
Δ_0	5.31(1)	4.93(6)	4.05(4)	3.32(3)
Δ_1	10.4(1)	8.15(7)	7.65(5)	7.19(4)
Δ_2		12.1(5)	10.2(3)	15.6 (2)
Δ_3				34.6(1.2)



x > 0 : sum of Schottky contributions

E/k_B (K)	$x = 0.03$	$x = 0.1$	$x = 0.25$	$x = 0.5$
Δ_0	5.31(1)	4.93(6)	4.05(4)	3.32(3)
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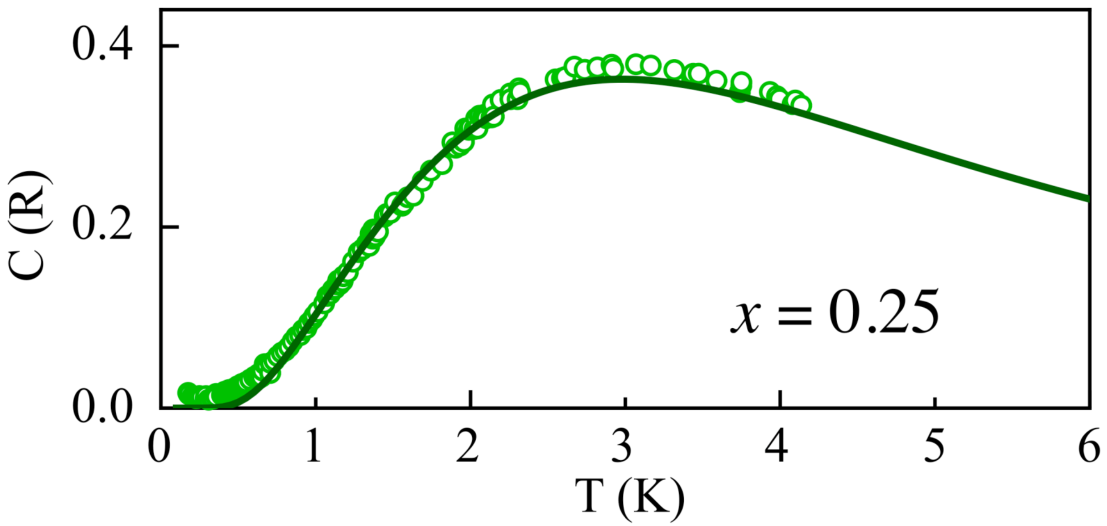
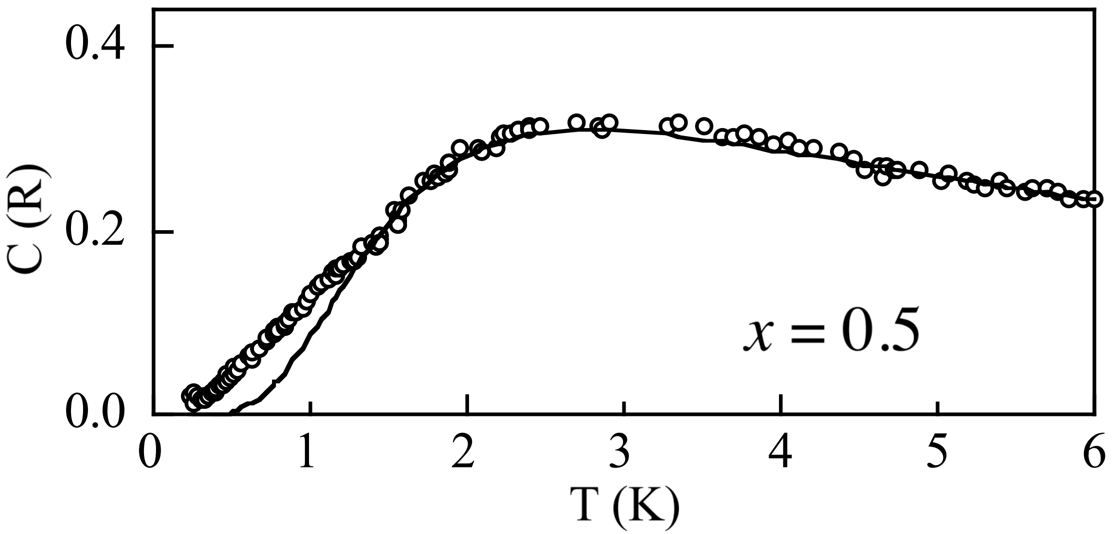
Δ_2		12.1(5)	10.2(3)	15.6 (2)
Δ_3				34.6(1.2)



x > 0 : sum of Schottky contributions

E/k_B (K)	$x = 0.03$	$x = 0.1$	$x = 0.25$	$x = 0.5$
Δ_0	5.31(1)	4.93(6)	4.05(4)	3.32(3)
Δ_1	10.4(1)	8.15(7)	7.65(5)	7.19(4)
Δ_2		12.1(5)	10.2(3)	15.6 (2)
Δ_3				34.6(1.2)

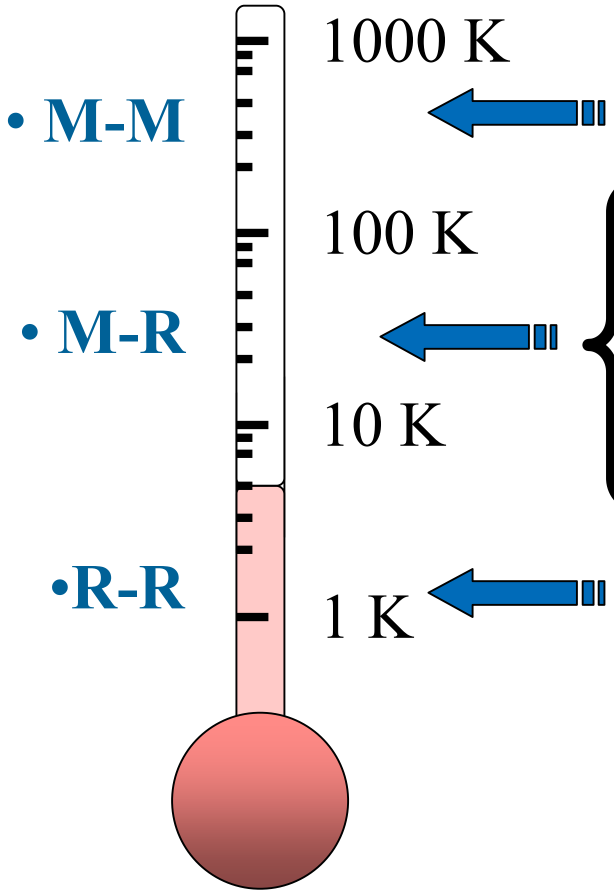


Conclusions

- The dilution of LS Co^{3+} in $NdFe_{1-x}Co_xO_3$ is equivalent to the inclusion of magnetic vacancies in the system
- The vacancies uncompensate the AF Fe ordered sublattice, increasing the Fe-Nd interaction and inhibiting Nd cooperative order for $x \geq 0.1$
- A mean field model for the parent compound has been modified to take account for low doping $NdFe_{1-x}Co_xO_3$ ($x \leq 0.1$)
- It is appealing to suggest that this is the mechanism for negative magnetization in manganites (isntead of ferrimagnetism)

RMO_3 : Model systems

M-M, M-R, R-R interactions.



M antiferro order (Cr, Fe, Ni, ...)

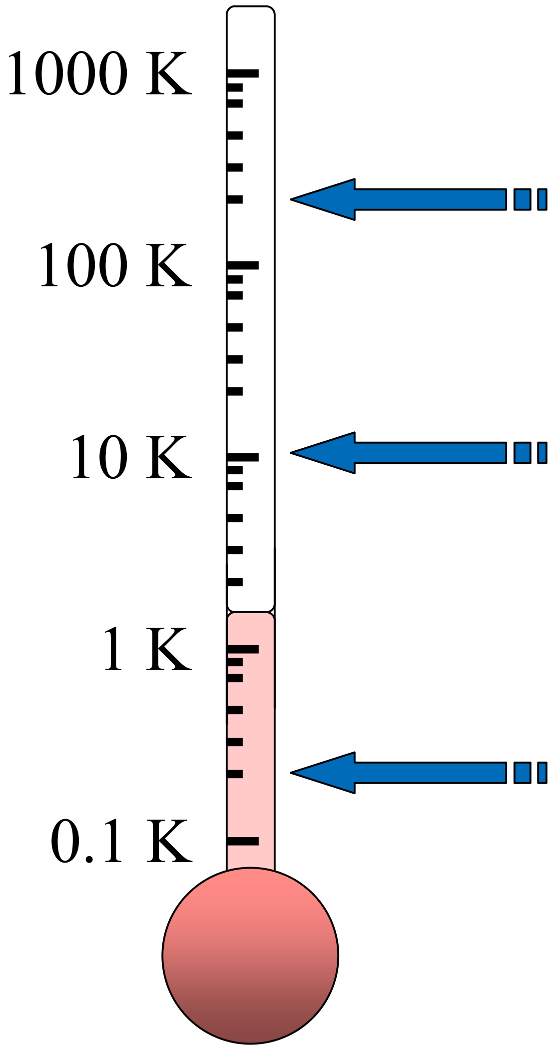
M SRT's, ...

R induced order: M-R

R cooperative order: R-R

(Nd, Sm, Gd, Er, ...)

$NdNiO_3$: Peculiar system



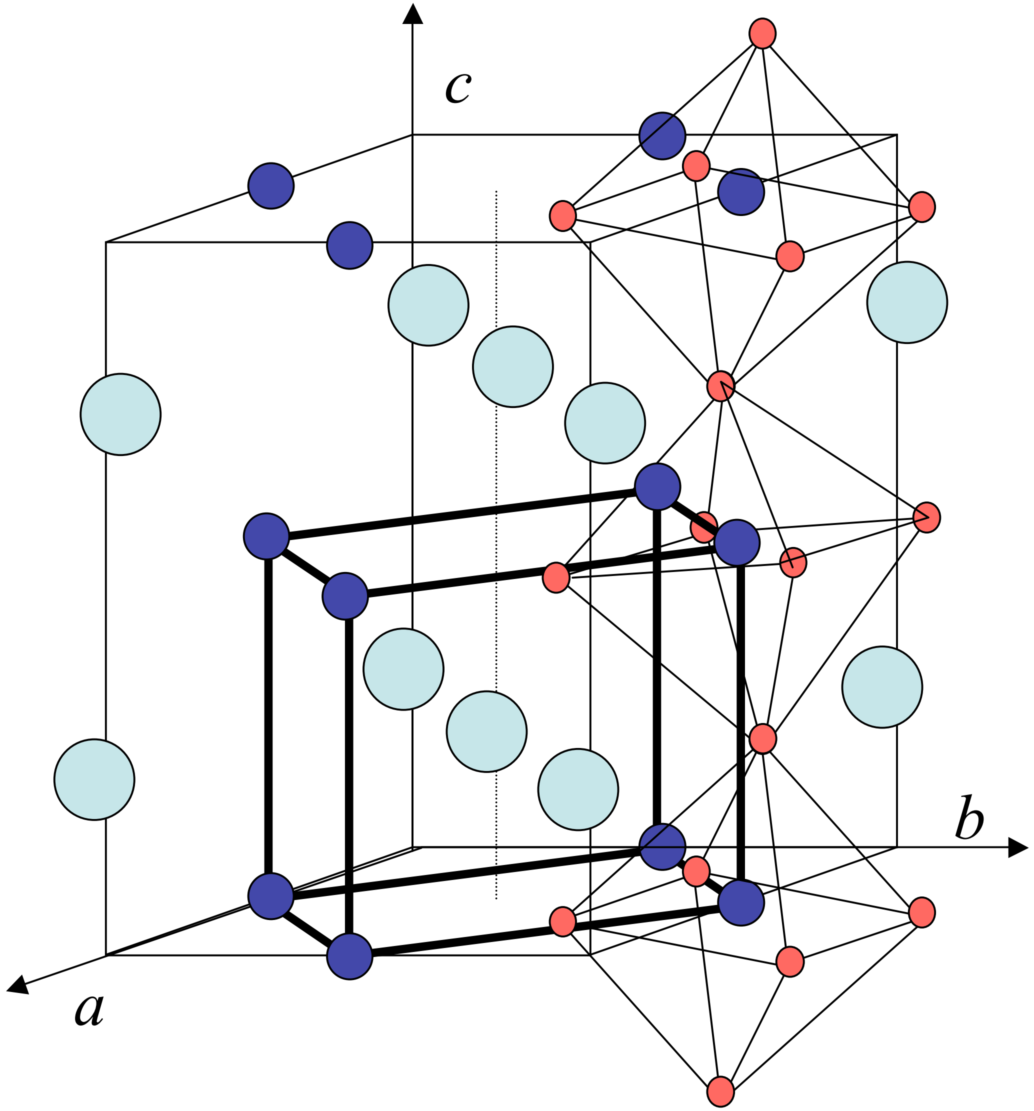
$T_N = T_{M\text{-}I} = 200$ K → **Ni AF order**
(a very unusual one!)
M-I transition &
Orbital order

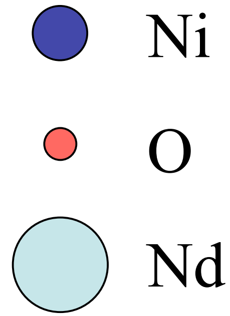
$T_{Nd} = 20$ K → **Nd induced order**
(very unusual one, too!)

$T = 0.2$ K → **Enhancement of the diffracted neutron intensities (unclear origin)**

for a review, see M. Medarde, J. Phys.:Condens Martter ***9****, 1679 (1997)*
J.L. García Muñoz et al., Phys. Rev. B ***50****, 978 (1994)*
S. Rosenkranz, Ph.D. Thesis ETH Zürich, 11853 (1996)

$NdNiO_3$: crystal structure



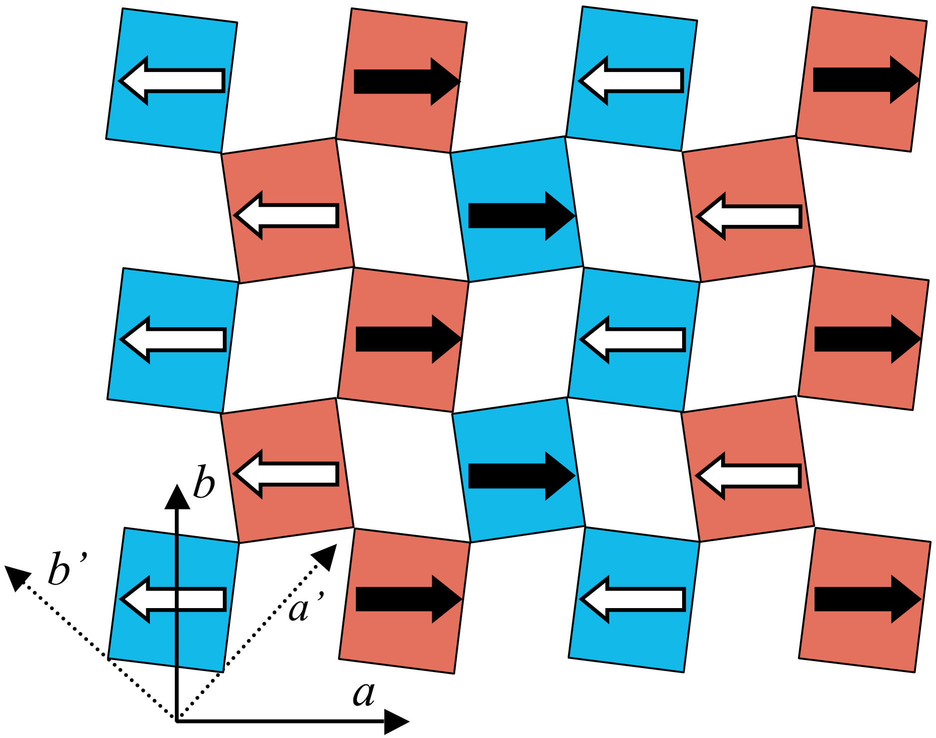


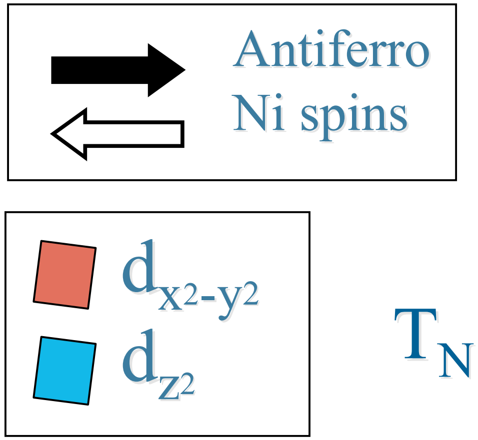
- *Pbnm* orthorhombic perovskite

Each Nd ion is placed at the center of a Ni cube

Ni occupies *ab* planes : A planes

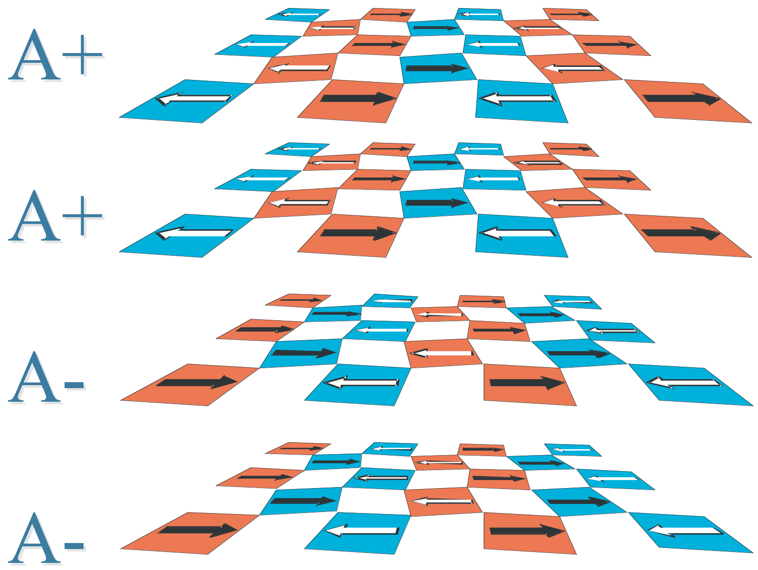
Ni magnetic order in $NdNiO_3$: "Model 1"





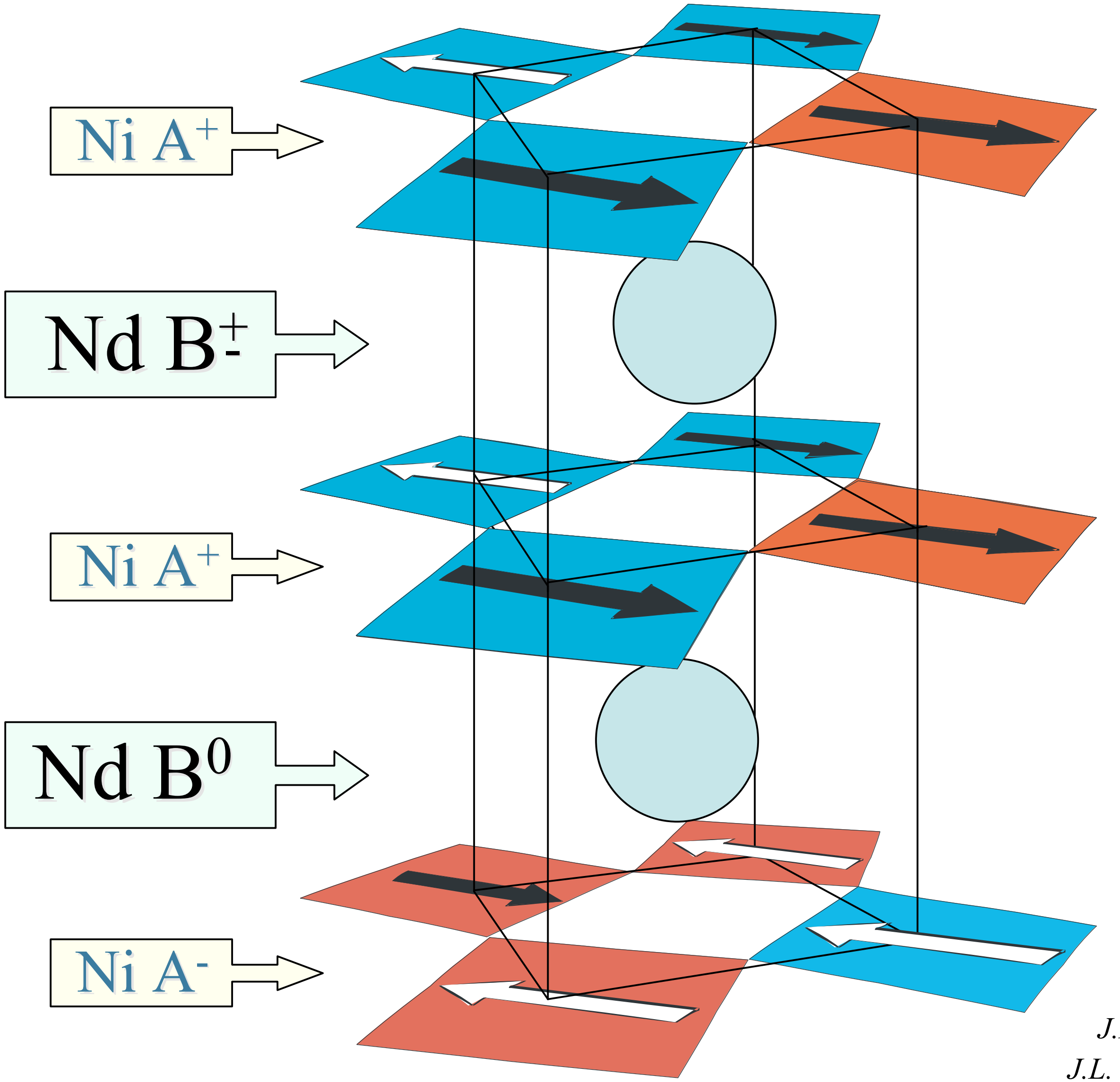
$T_N = 200$ K

Stacking of alternating *ab* layers
$A^+ A^- A^- A^+ A^+ A^- A^- A^+ A^+ ...$



*J.L. García Muñoz et al., Phys. Rev. B **50**, 978 (1994)*
*J.L. García Muñoz et al., Phys. Rev. B **51**, 15197 (1995)*

“Model 1”: Two kind of magnetic Nd ions



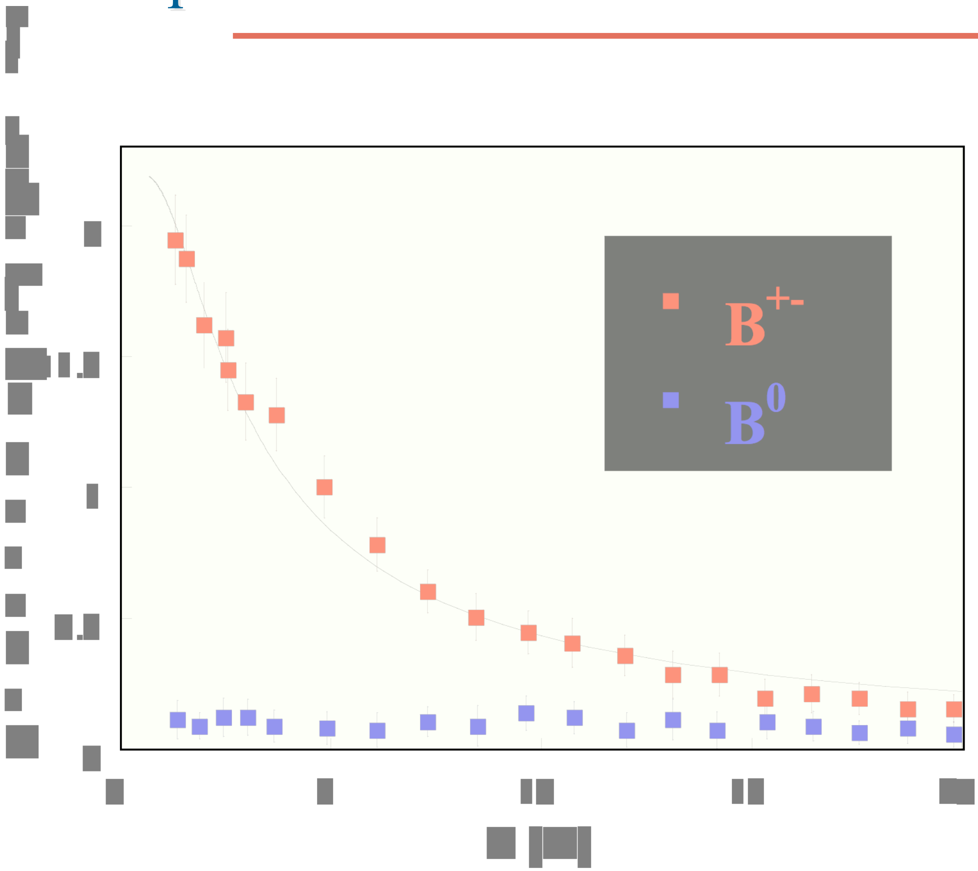
Model 1 hypothesis

Uncompensated environment:
Strong polarization

Compensated environment:
Zero polarization

J.L. García Muñoz et al., Phys. Rev. B ***50****, 978 (1994)*
J.L. García Muñoz et al., Phys. Rev. B ***51****, 15197 (1995)*

“Model 1”: Diffracted intensity from polarized Nd



However...

Nd compensated environments :

non-zero Zeeman splittings

$NdCrO_3$: $\Delta = 27$ K ;

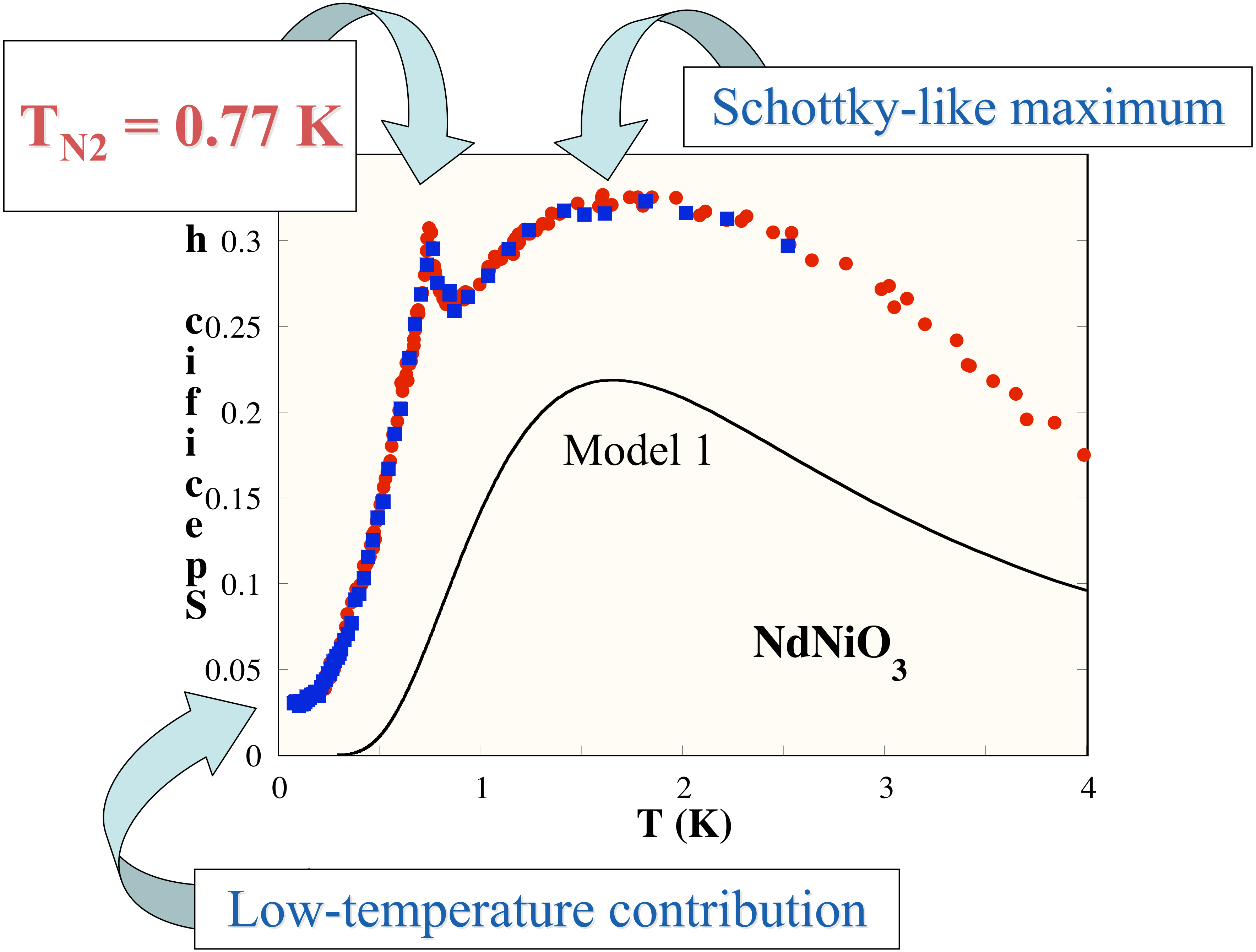
$NdFeO_3$: $\Delta = 5.7$ K

$NdMnO_3$: $\Delta \sim 25$ K

Macroscopic Experimental test of “Model 1”:

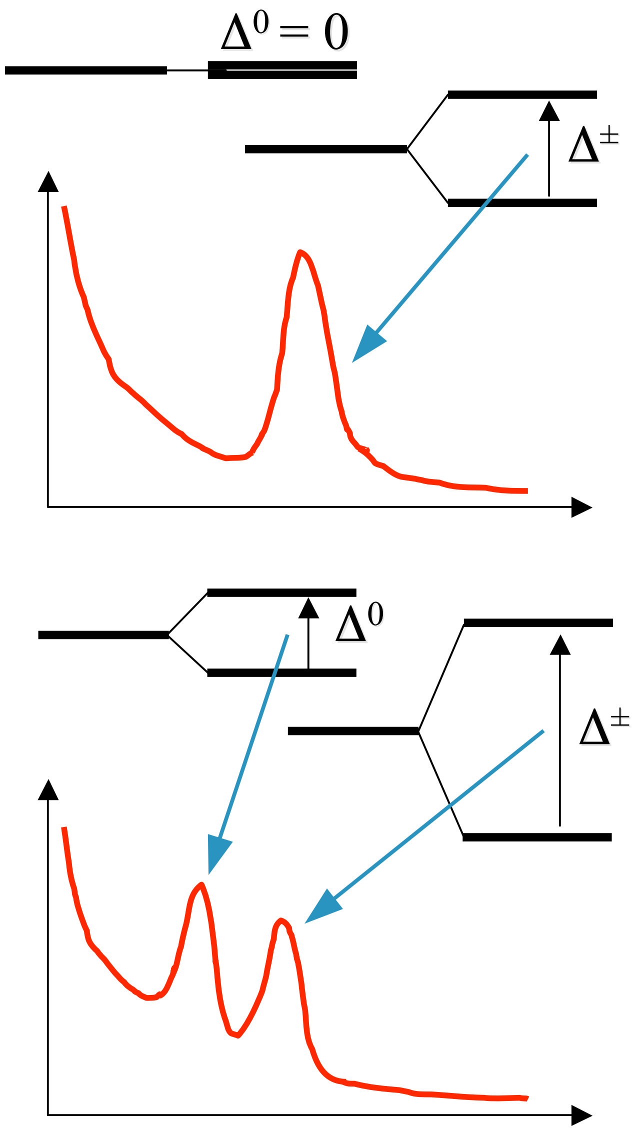
Low-temperature specific heat

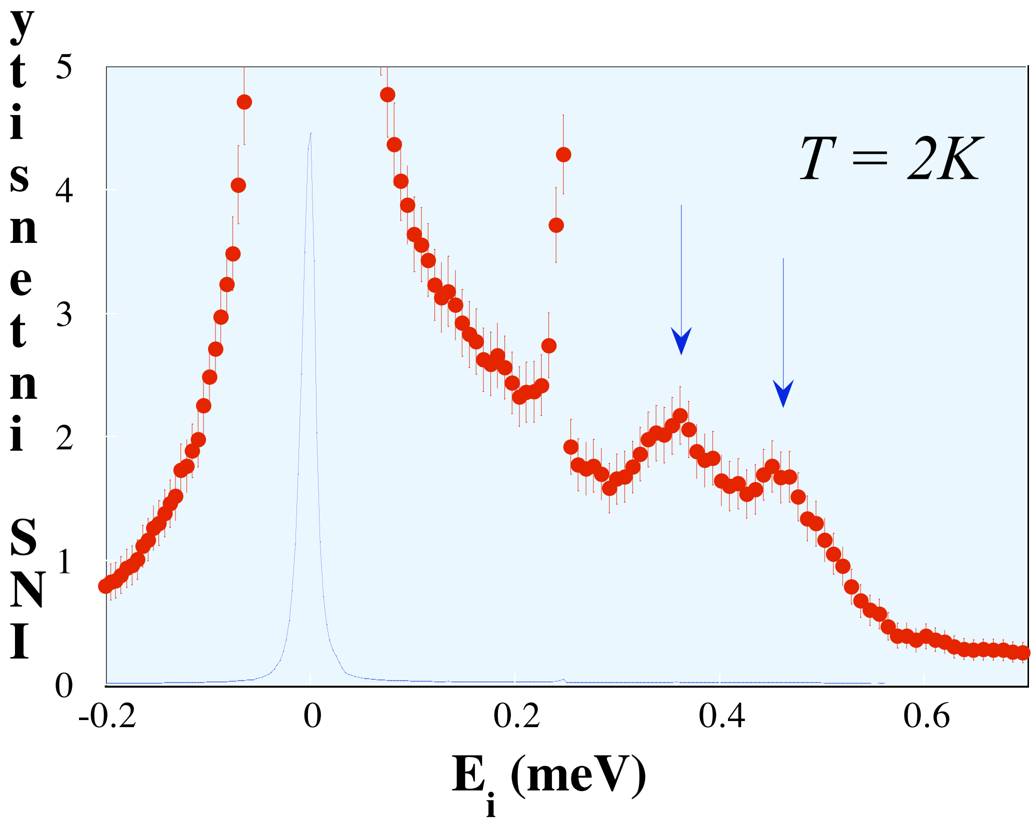
J.L. García Muñoz et al., Phys. Rev. B ***50****, 978 (1994)*



T_N2 = 0.77 K
Schottky-like maximum
Specific h
0.3
0.25
0.2
0.15
0.1
0.05
0
0
1
2
3
4
T (K)
Model 1
NdNiO3
Low-temperature contribution

A microscopic probe:
high resolution Inelastic Neutron Scattering

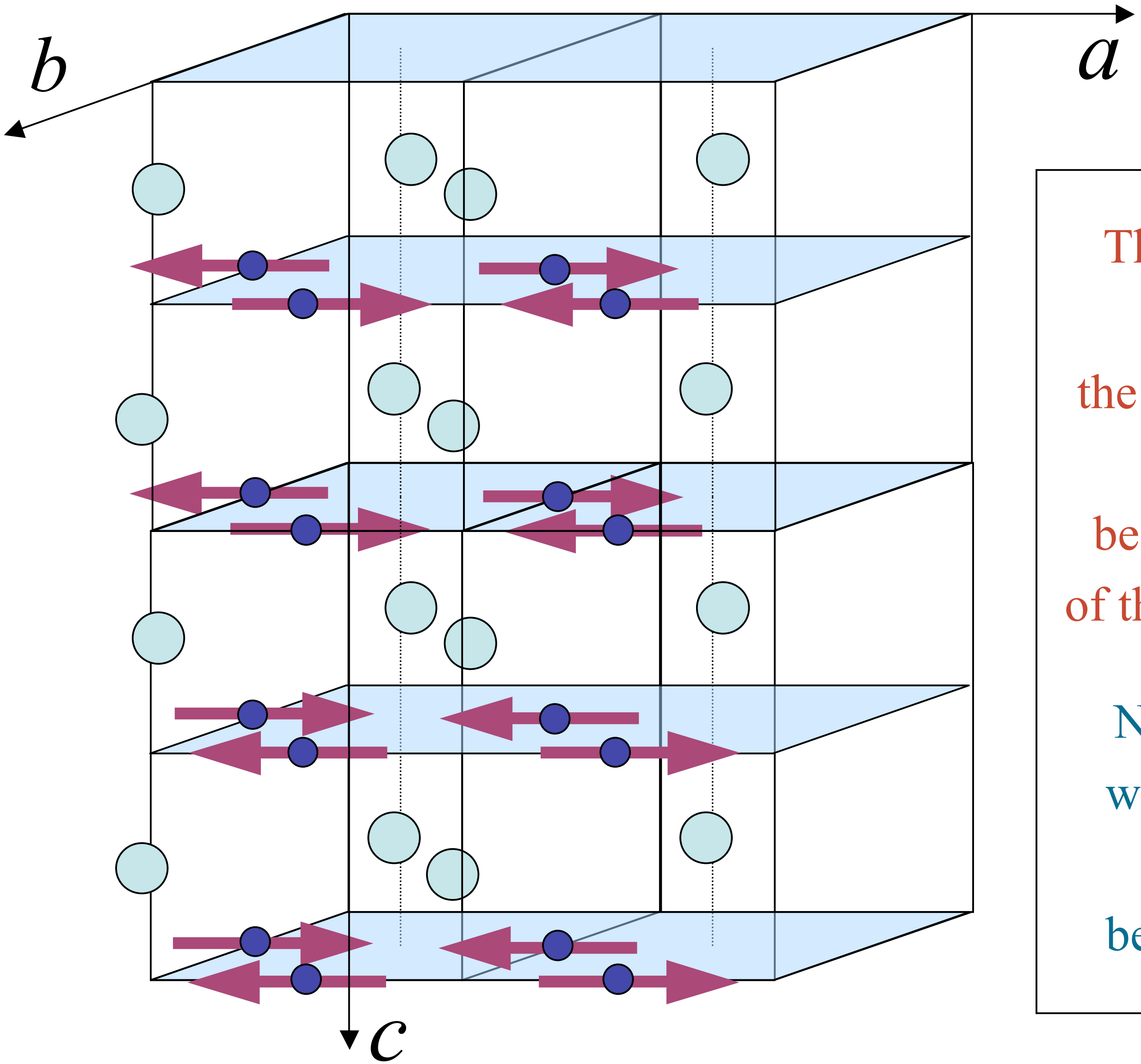




Two channels evidenced by HR-INS:

$\Delta^0 = 4.1\ \mathrm{K}\ ;\ \ \Delta^{\pm} = 5.2\ \mathrm{K}$

Quadruple Magnetic unit cell, $T_N = 200$ K



The Ni N_x magnetic mode
POLARIZES
the Nd subsystem in a mode
(let's call it $\mathbf{n_x}$)
belonging to the same *irep*
of the magnetic group than N_x

Nd-Nd cooperative order
will favour another mode
(let's call it **r**)
belonging to another *irep*

Mean field model: *specific heat*

Spin Hamiltonian for the Nd system:

$$\mathcal{H} = -2\theta_c \rho \hat{\mathbf{r}} - 2\theta_p \nu \hat{\mathbf{n}}_x - g_x \mu_B H_{exc} \hat{\mathbf{n}}_x - 2\theta_c \rho^2 - 2\theta_p \nu^2$$

Self interaction terms

Ni-Nd Zeeman term (eff. exchange field)

Nd-Nd exchange of the polarized component

Nd-Nd exchange in cooperative mode

where...

$\rho = -\frac{1}{2}\langle \hat{\mathbf{r}} \rangle$ and $\nu = -\frac{1}{2}\langle \hat{\mathbf{n}}_x \rangle$ are mean field order parameters for the cooperative and induced order modes

θ_c and θ_p are the Nd-Nd exchange constants in cooperative and polarized modes

Free energy :

$$F^{\text{á}} = \sum_{\text{á}=\pm,0} \frac{1}{2}\theta_{\mathrm{c}}\rho^2 + \frac{1}{2}\theta_{\mathrm{p}}\nu^2 - T\ln\left\{2\cosh\left(\frac{\Delta^{\text{á}}}{2T}\right)\right\}$$

with

$$\Delta^{\text{á}} = \sqrt{(\theta_{\mathrm{c}}\rho)^2 + \left(g_x\mu_B H_{exc}^{\text{á}} + \theta_{\mathrm{p}}\nu\right)^2}$$

being the exchange splittings of the Nd^{3+} (±, 0) ground doublets.

By minimizing F with respect to ρ and ν, we get the characteristic equations of the system, which gives the ordered net Néel moments ρ and ν **as a function of *T*.**

Ecuaciones características :

$$\rho = \rho \frac{2\theta_{\mathrm{c}}}{\Delta^{á}} \tanh\left(\frac{\Delta^{á}}{2T}\right)$$

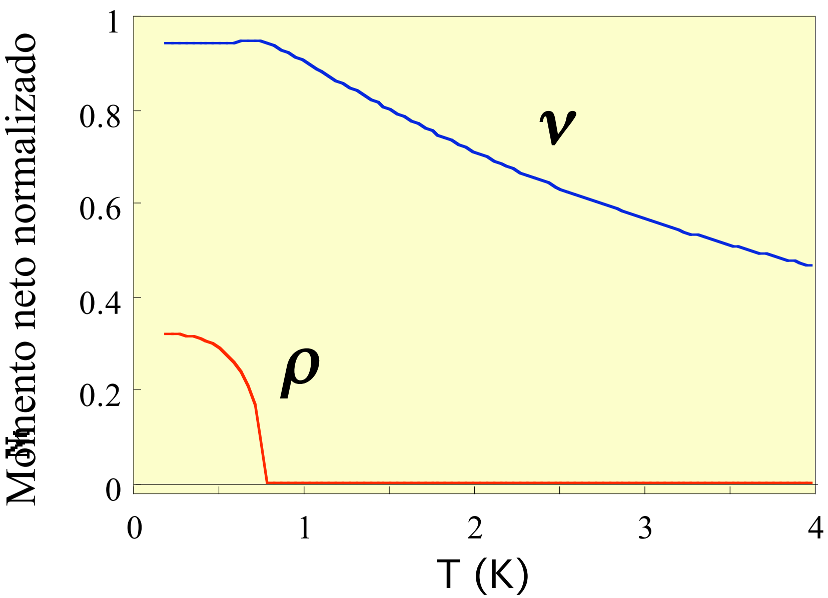
$$\nu = \frac{g_x \mu_B H^{á}_{exc} + 2\theta_{\mathrm{p}}\nu}{\Delta^{á}} \tanh\left(\frac{\Delta^{á}}{2T}\right)$$

$$\rho = 0$$

Fase paramagnética con orden inducido

$$\frac{\Delta^{á}}{2\theta_{\mathrm{c}}} = \tanh\left(\frac{\Delta^{á}}{2T}\right)$$

Fase ordenada cooperativamente

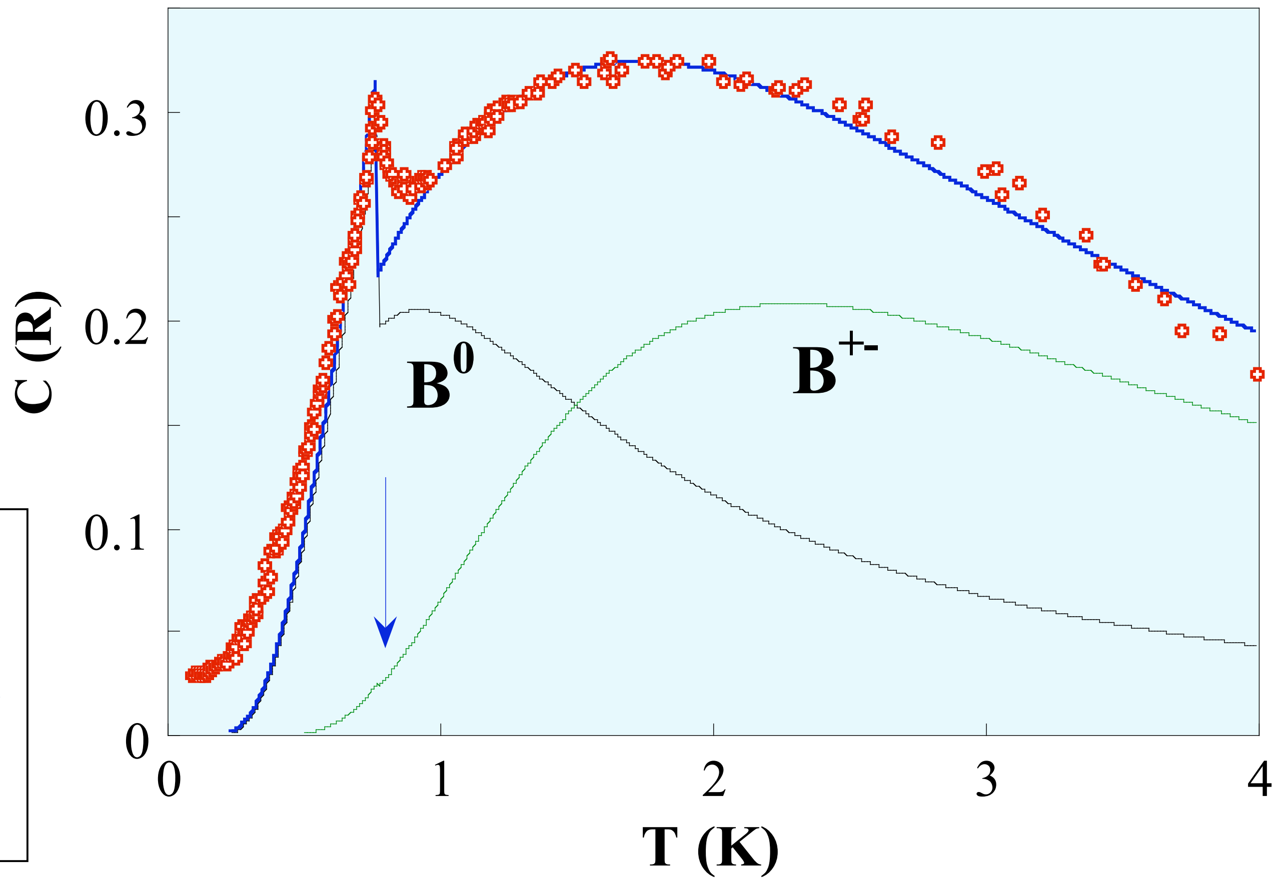


Transición de Fase

Entropy...

$$S = \ln 2 + \sum_{á\,=\pm,0} \ln\left[\cosh\left(\frac{\Delta^{á}}{2T}\right)\right] - \frac{\Delta^{á}}{2T}\tanh\left(\frac{\Delta^{á}}{2T}\right)$$

and specific heat :

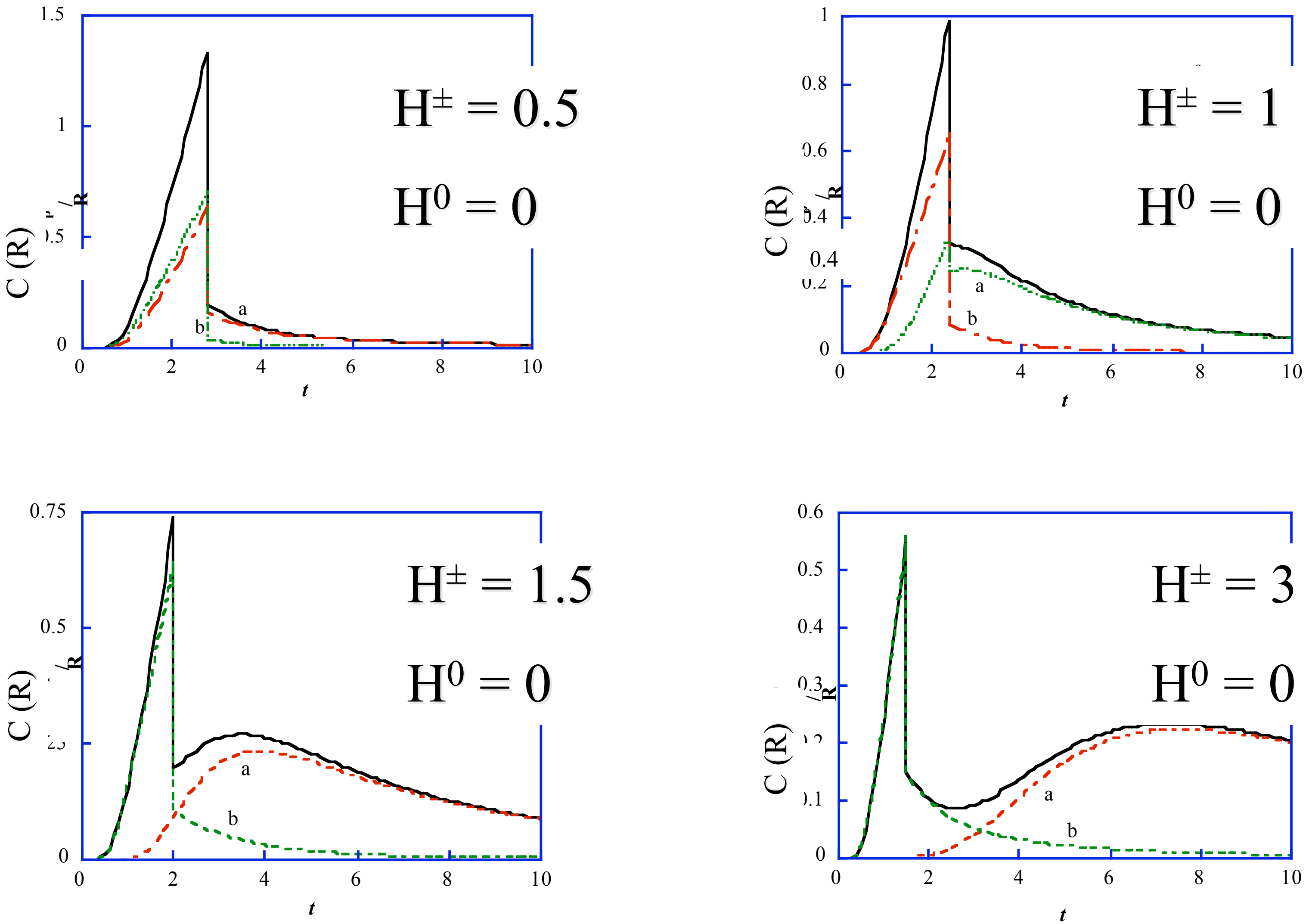


$\Delta^0{}_t = 2.6$ K ≅ $\Delta^0{}_e = 4.1$ K

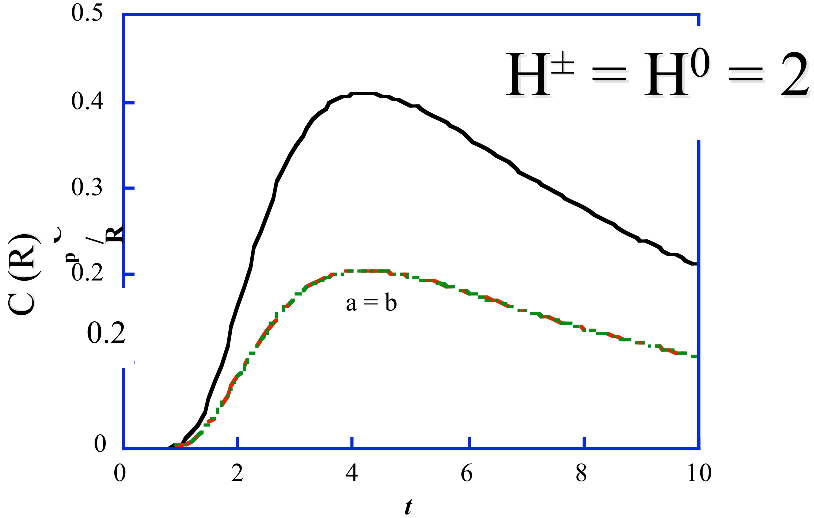
$\Delta^{\pm}{}_t = 5.3$ K Z $\Delta^{\pm}{}_e = 5.2$ K

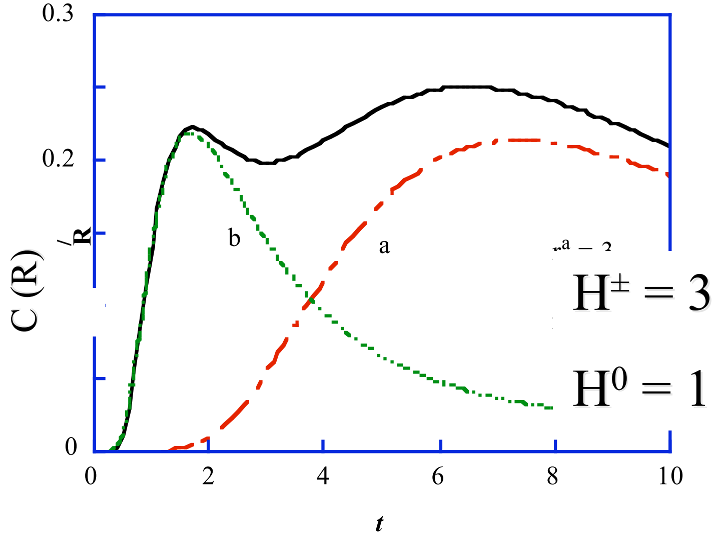
$T_{Nt} = T_{Ne} = 0.77$ K

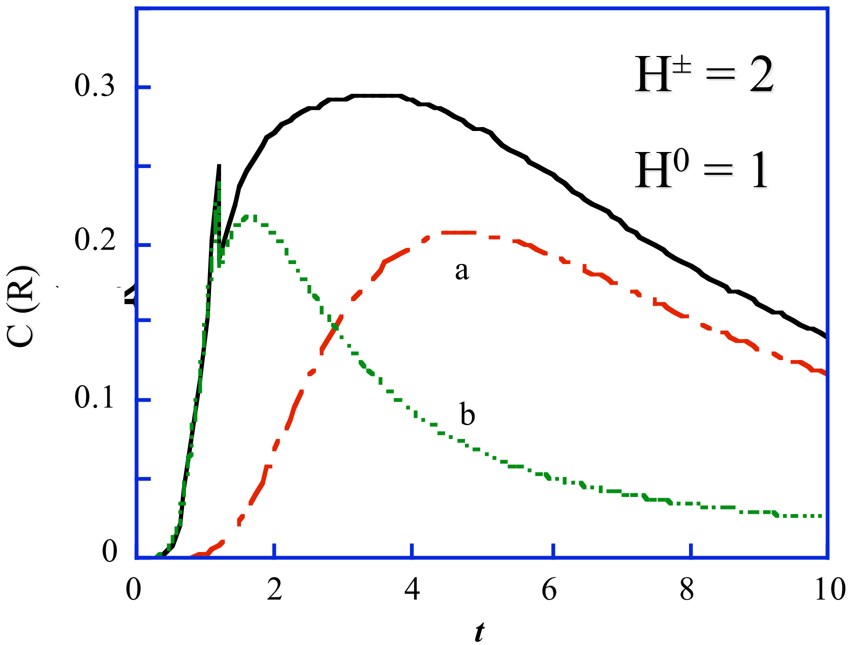
Results for $H^0_{exc} = 0$:



Results for $H^0_{exc} > 0$:

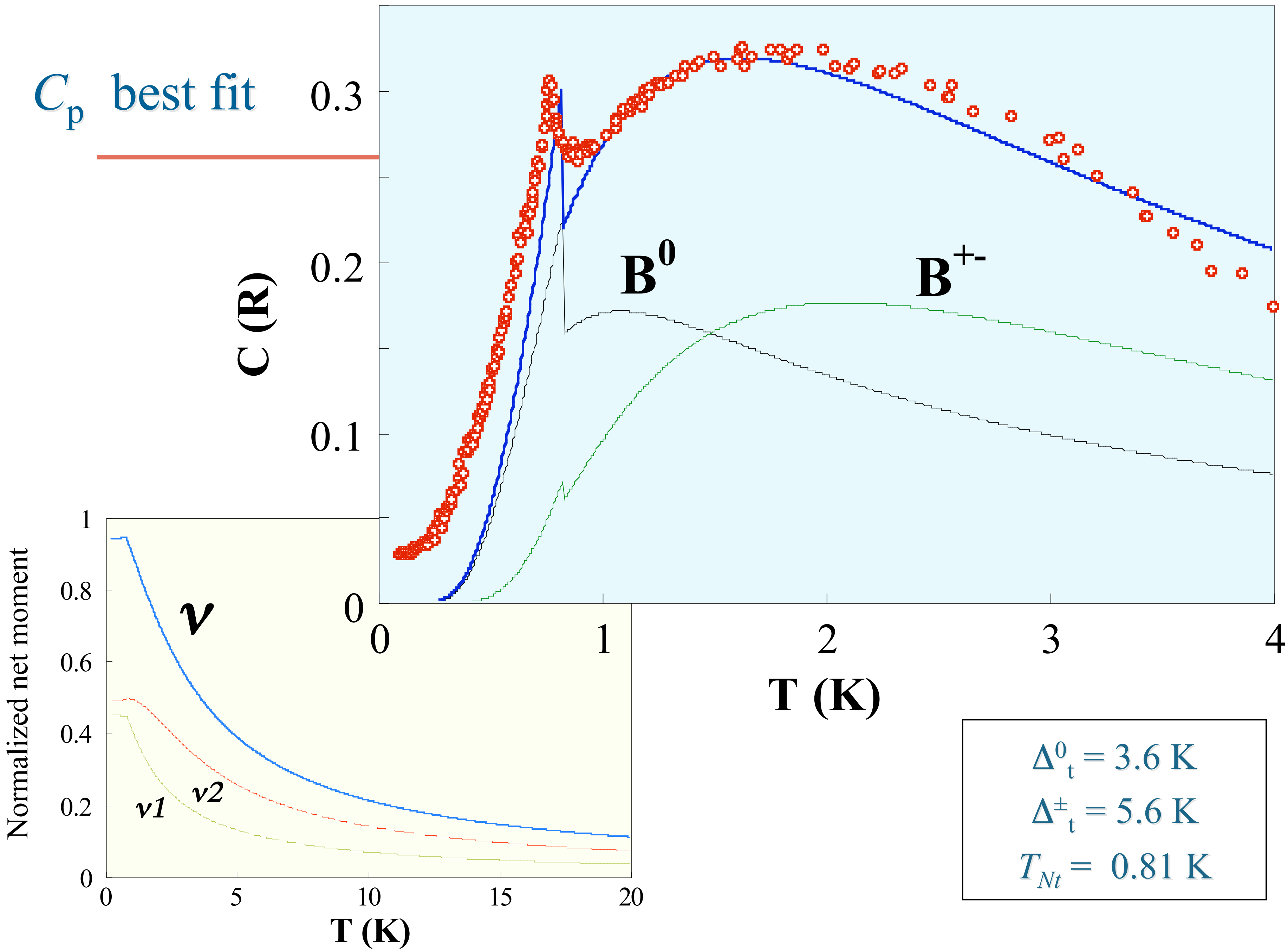






Only intermediate (though different) values for both, $H^{\pm}_{exc}$ and H^0_{exc} reproduce the experimental results

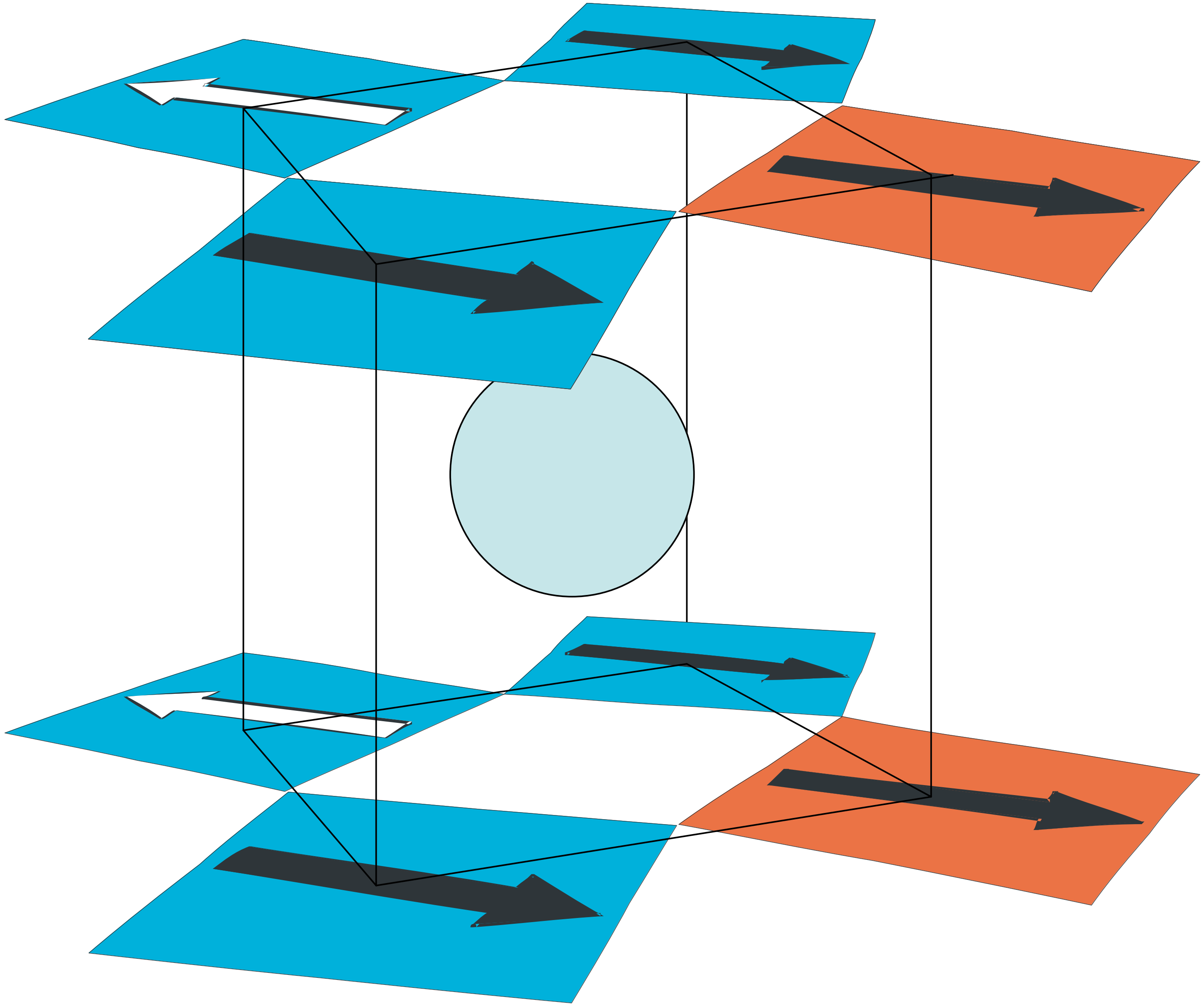
C_p best fit



$\Delta^0_t = 3.6$ K

$\Delta^{\pm}_t = 5.6$ K

$T_{Nt} = 0.81$ K



Glassy behaviour of the Nd sublattice induced by Fe doping in $NdFe_xGa_{1-x}O_3$

F. Bartolomé [a,*], M. Parra-Borderías [a], J. Blasco [a], J. Bartolomé [a]

[a] *Instituto de Ciencia de Materiales de Aragón, CSIC - Universidad de Zaragoza. Pedro Cerbuna 12, 50009 Zaragoza. Spain*

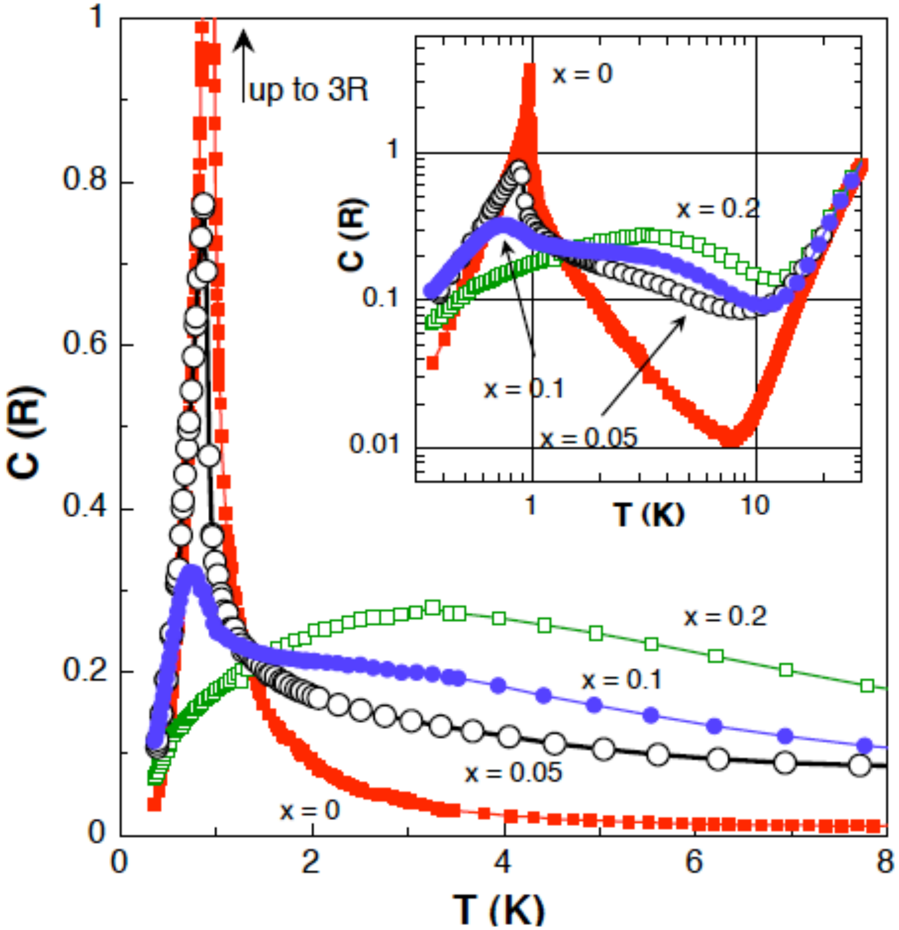
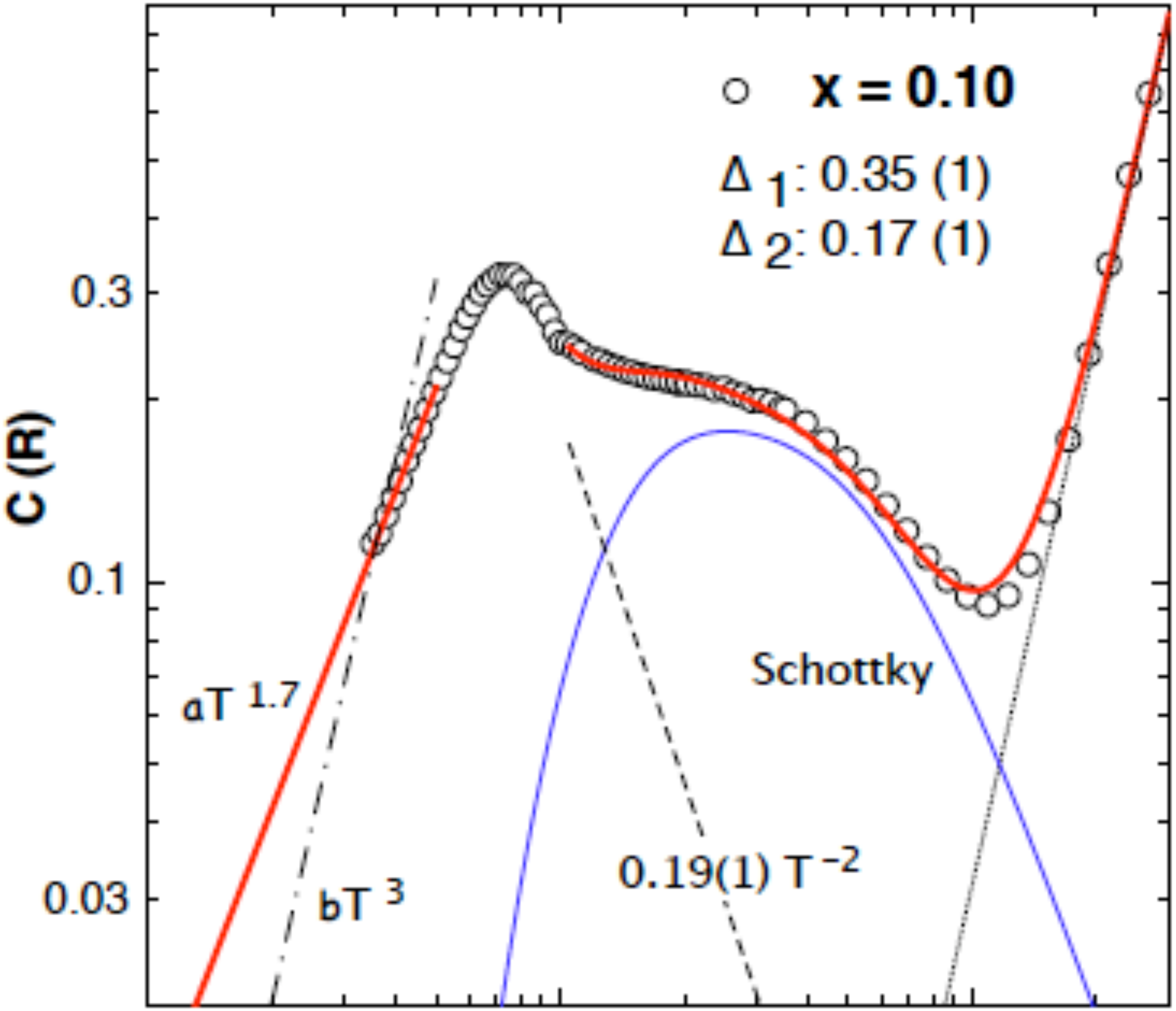


Fig. 1. Low-temperature specific heat measurements on the $NdFe_xGa_{1-x}O_3$ system for x =0 (■), 0.05 (○), 0.1 (●) and 0.2 (□). The inset shows the data in a double-log scale, up to 30 K to evidence the lattice contributions.


x = 0.10
Δ 1: 0.35 (1)
Δ 2: 0.17 (1)
0.3
0.1
0.03
C (R)
aT 1.7
bT 3
0.19(1) T −2
Schottky

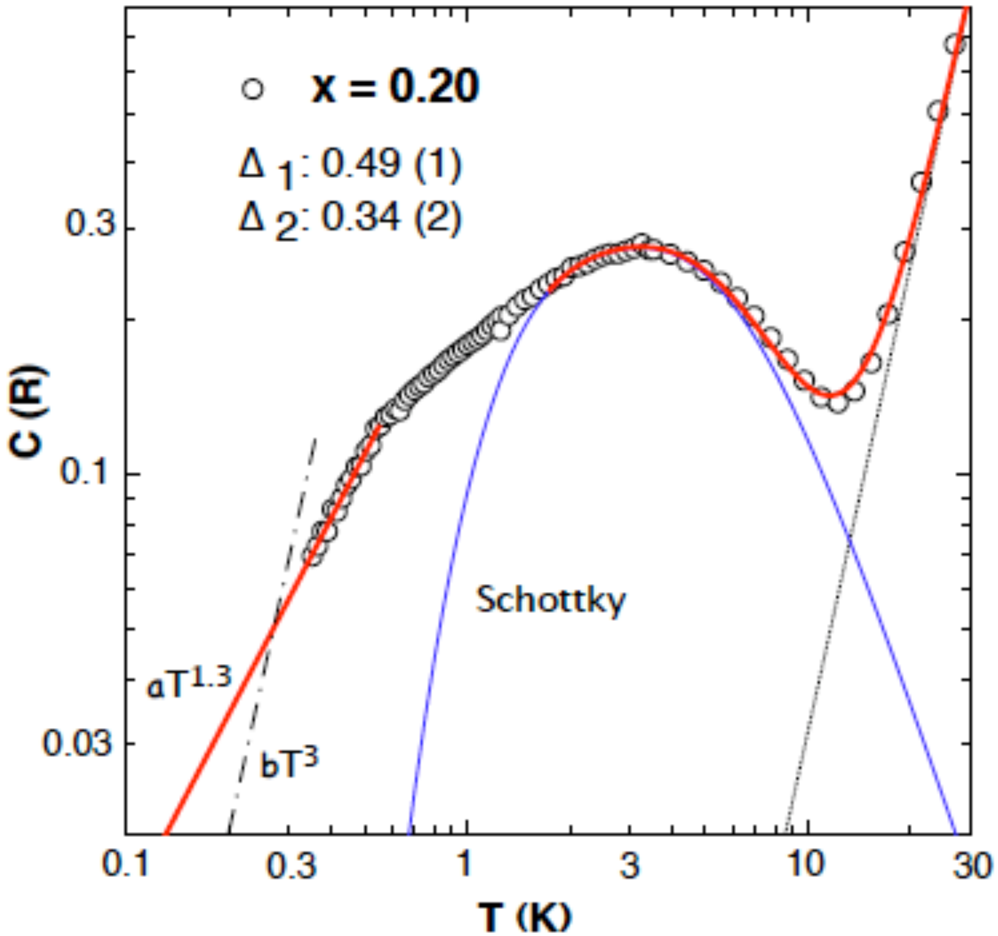
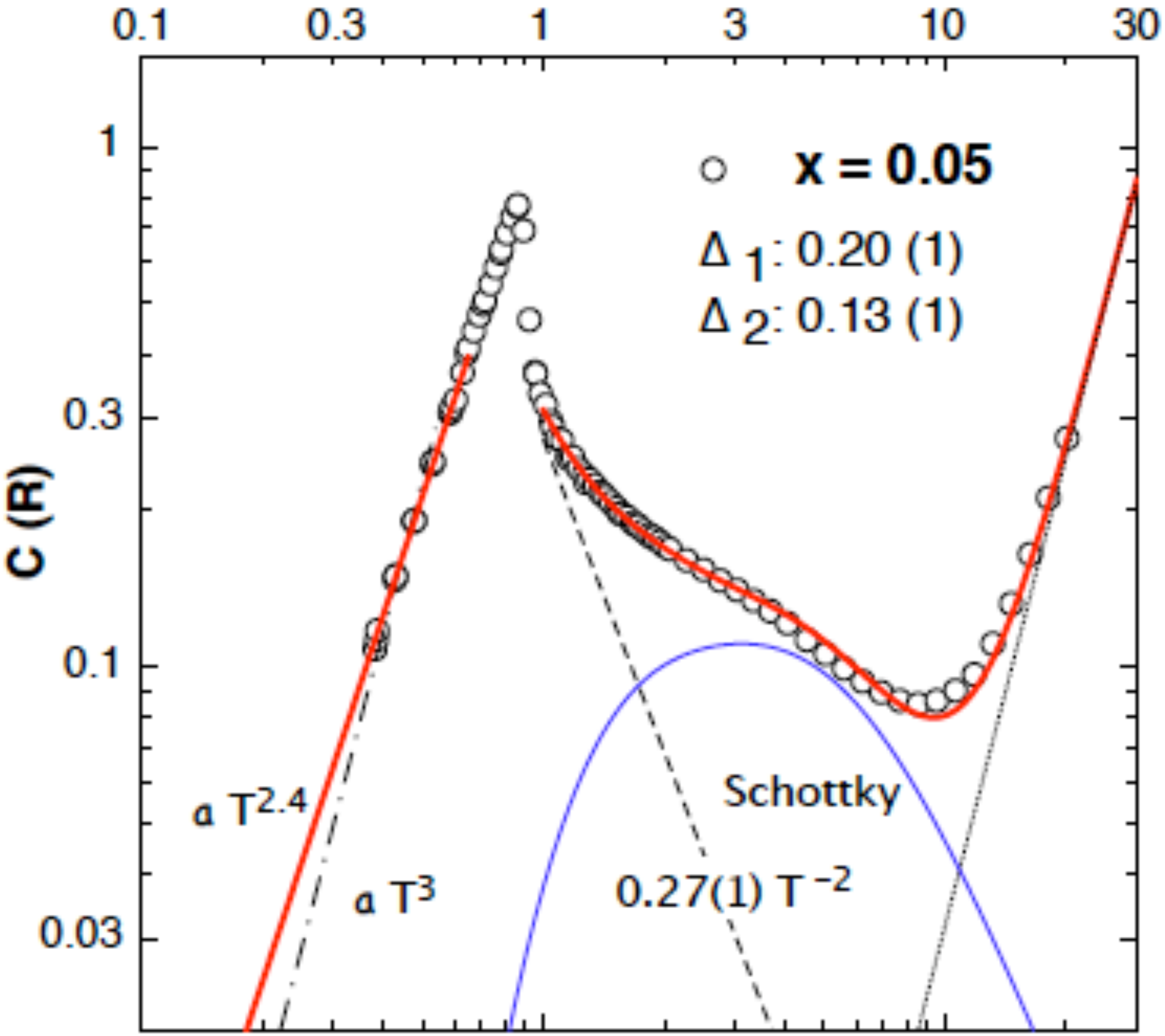


Fig. 2. Analysis of the low-temperature specific heat data (○) for $x = 0.05$ (top), $x = 0.1$ (middle), and $x = 0.2$ (bottom). See the text for details.


0.1
0.3
1
3
10
30
1
0.3
0.1
0.03
C (R)
x = 0.05
Δ 1: 0.20 (1)
Δ 2: 0.13 (1)
Schottky
a T2.4
a T3
0.27(1) T −2