

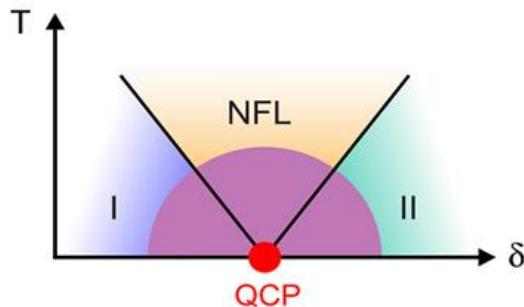
# Beyond Landau Fermi liquid and BCS superconductivity near quantum criticality

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# Lecture 4

## Examples of quantum critical *d*- and *f*-electron systems

Contents of Lecture 4

CePdAl – a partially frustrated heavy-fermion system

Approaching quantum criticality of CePdAl with Ni substitution

Frustrated Ce moments in CePdAl: a two-dimensional spin liquid?

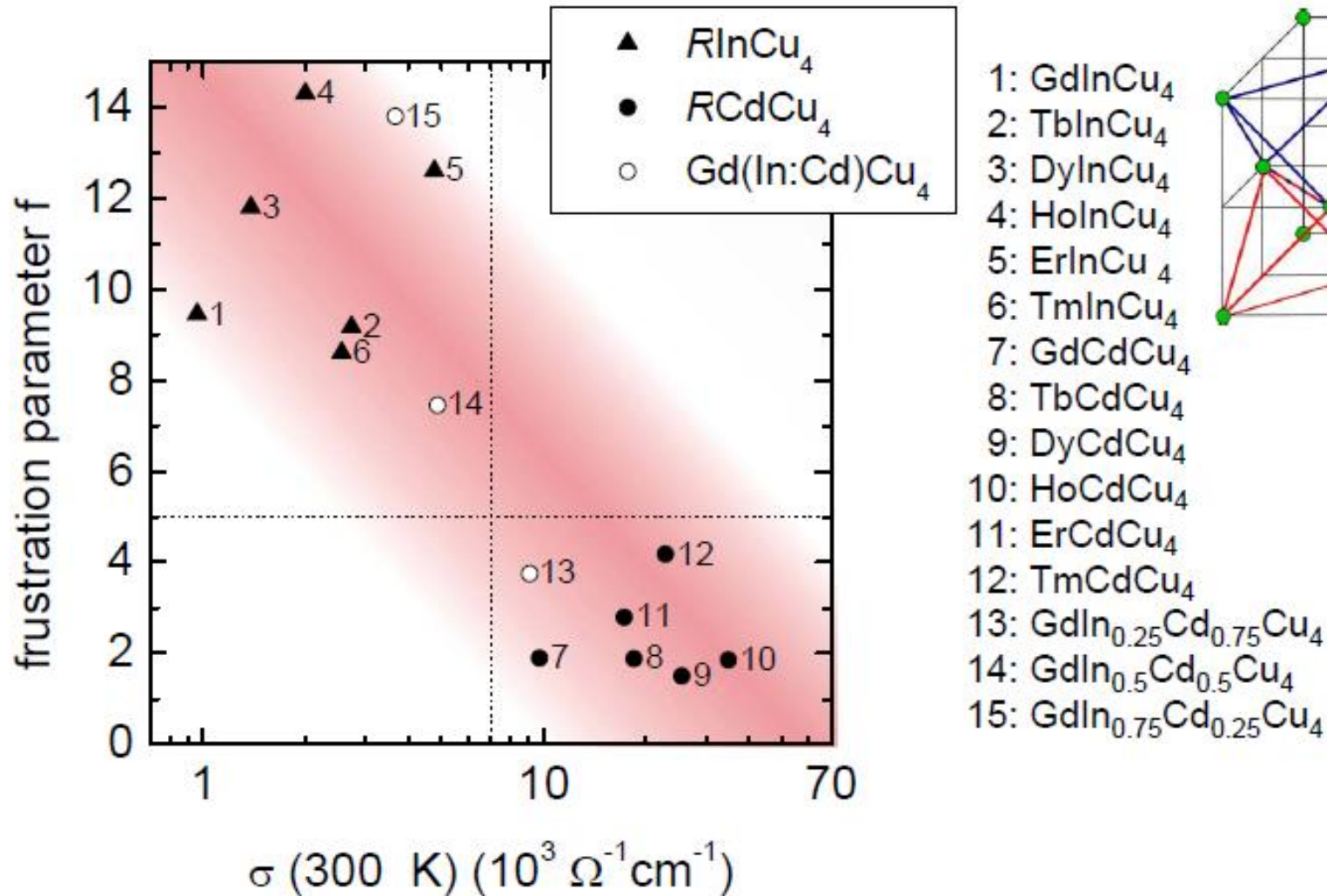
Classical and quantum phase transitions in the itinerant  
ferromagnet  $\text{Sr}_{1-x}\text{Ca}_x\text{RuO}_3$

Evolution of critical exponents in  $\text{Sr}_{1-x}\text{Ca}_x\text{RuO}_3$  with  $x$

Unusually slow the quantum critical dynamics

CePdAl – a partially frustrated  
heavy-fermion system

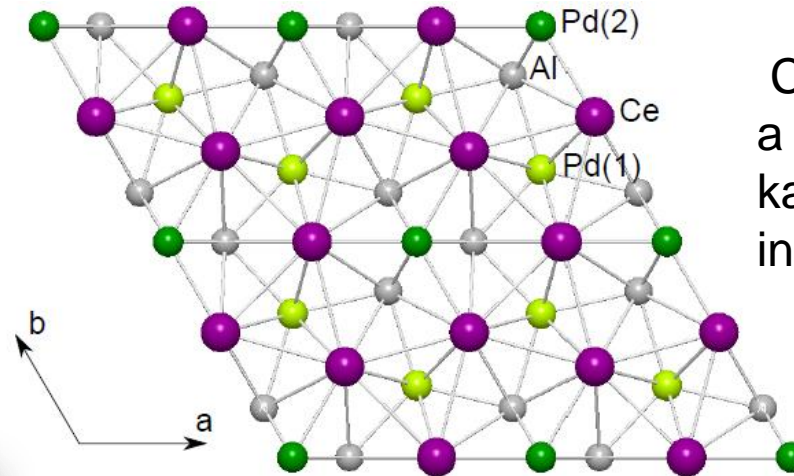
# Frustration and conductivity



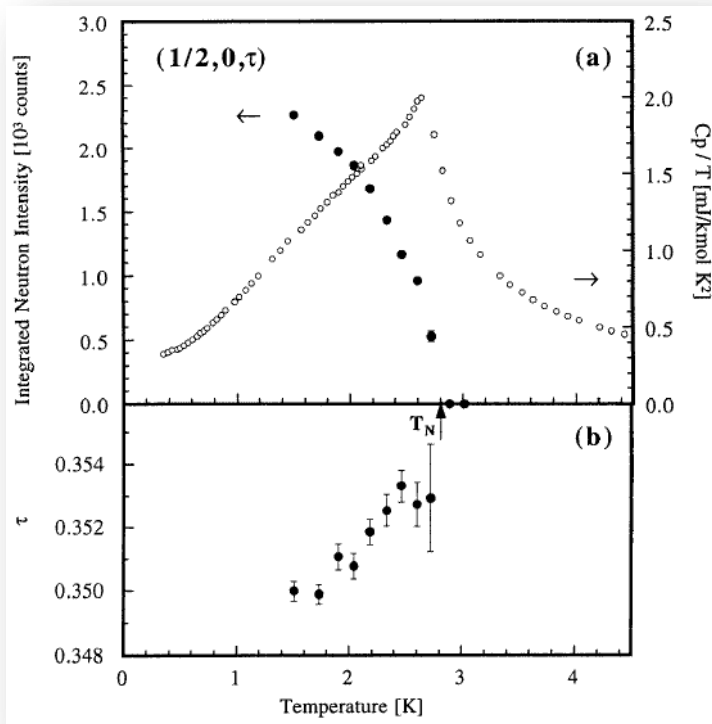
Frustration parameter  $f = \Theta_{\text{CW}}/T_c$

*V. Fritsch et al., PRB 72 (2006)*

# CePdAl – a partially frustrated Ce-based compound



Ce atoms form a distorted kagomé lattice in the  $ab$  plane

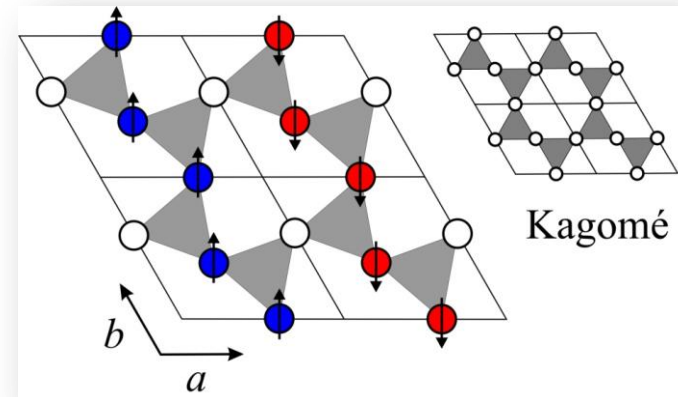


Magnetic order below  $T_N = 2.7$ .K

$$\mathbf{Q} = (\frac{1}{2} \ 0 \ \tau), \ \tau \approx 0.35$$

1/3 of Ce moments frustrated

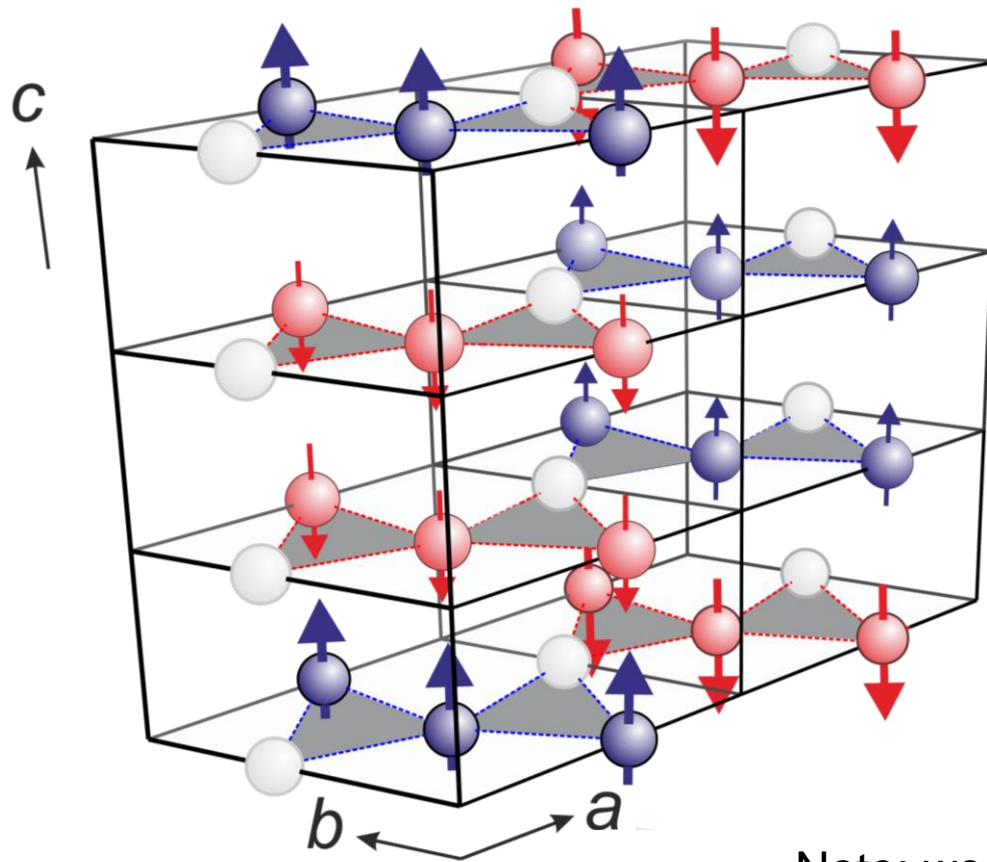
Magnetic structure



Kitazawa et al., *Physica B* **199&200**, 28, (1994)  
 Dönni et al., *J. Phys.: Cond. Matt.* **8**, 11213 (1996)

# Three-dimensional magnetic structure of CePdAl

Dönni et al.,  
*J. Phys.: Cond. Matt.* **8**, 11213 (1996)



Magnetic ordering  
vector

$$\mathbf{Q} = (\frac{1}{2} \ 0 \ \tau), \tau \approx 0.35$$

1/3 of Ce moments  
frustrated

$$\mu(\text{Ce1}) = 1.58 \mu_B$$

Note: weak  $T$ -dependent  
incommensuration  
neglected in the picture



# Model of frustrated kagomé-like planes

*Nunez-Regueiro and Lacroix, Physica C 282-287, 1885 (1997)*

nn interaction  $J_1$  (FM) and nnn  $J_2$  (AF)  
in the  $ab$  (kagome) planes,  
neglect of interplane coupling  $J_3$

$$H = \sum_i \Delta_i(T) |\mu_i|^2 - \frac{1}{2} \sum_{i \neq j} J_{ij} \vec{\mu}_i \cdot \vec{\mu}_j$$

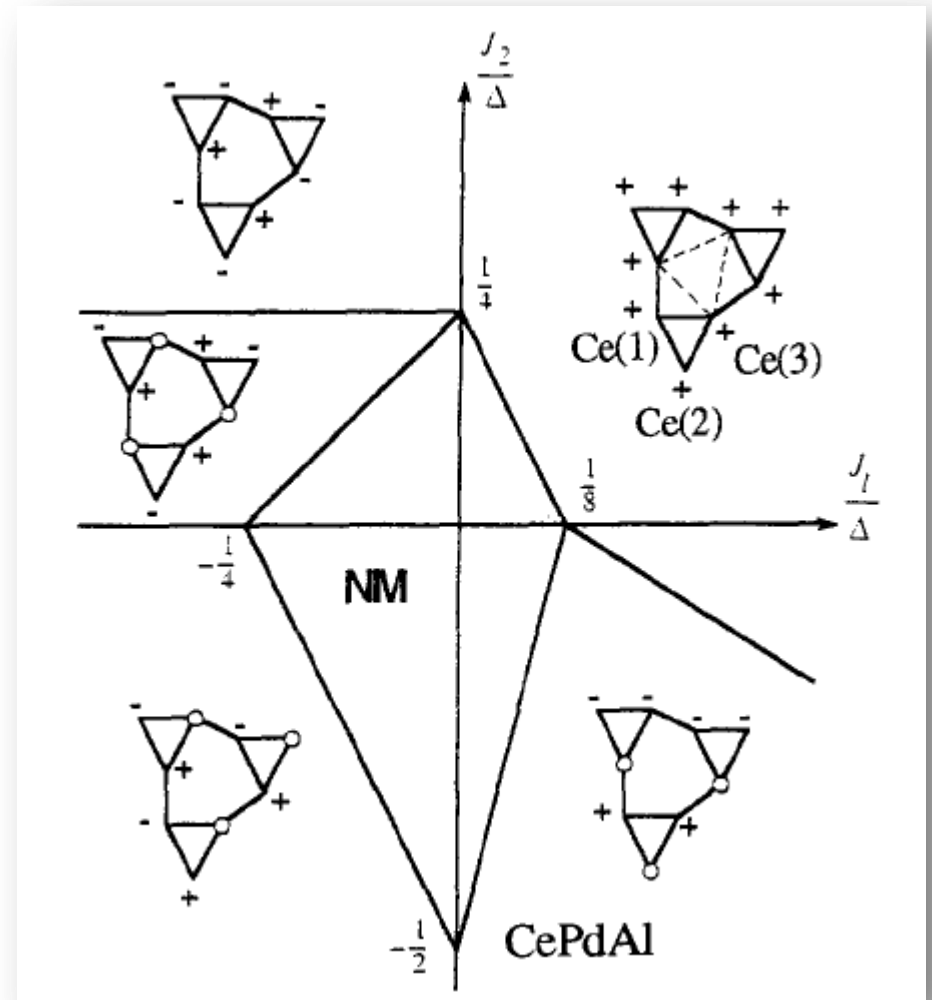
Kondo effect modelled by the energy  
difference  $\Delta_i(T)$  between Ce  
nonmagnetic Kondo state  $\mu_i = 0$  and  
magnetic state  $\mu_i \neq 0$ .

Mean-field phase diagram

confirmed by variational MC

*Motome et al., PRL 105, 036403*

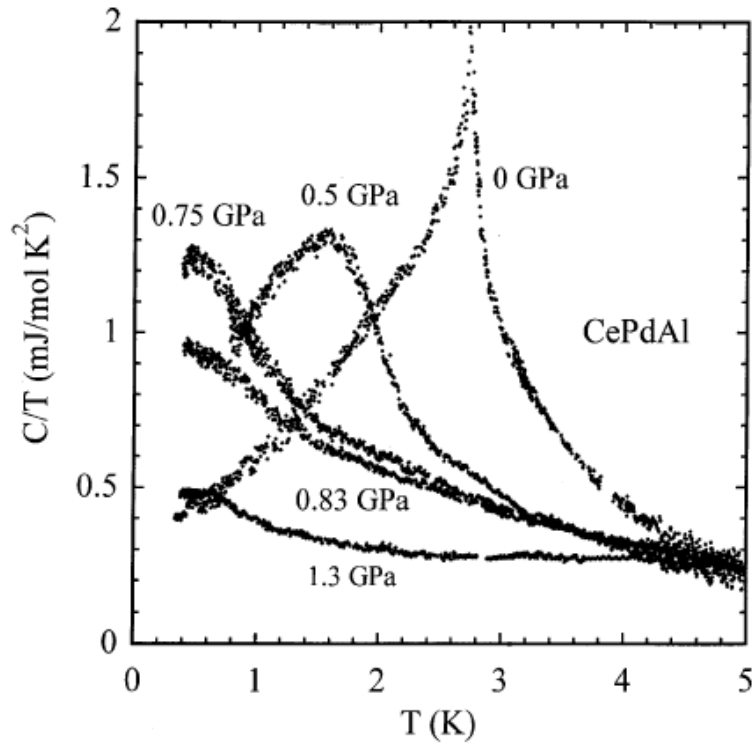
coupling between planes neglected!



Approaching quantum criticality  
of CePdAl  
with Ni substitution



# CePdAl – a partially frustrated Ce-based compound



Suppression of  $T_N$  by hydrostatic pressure ...

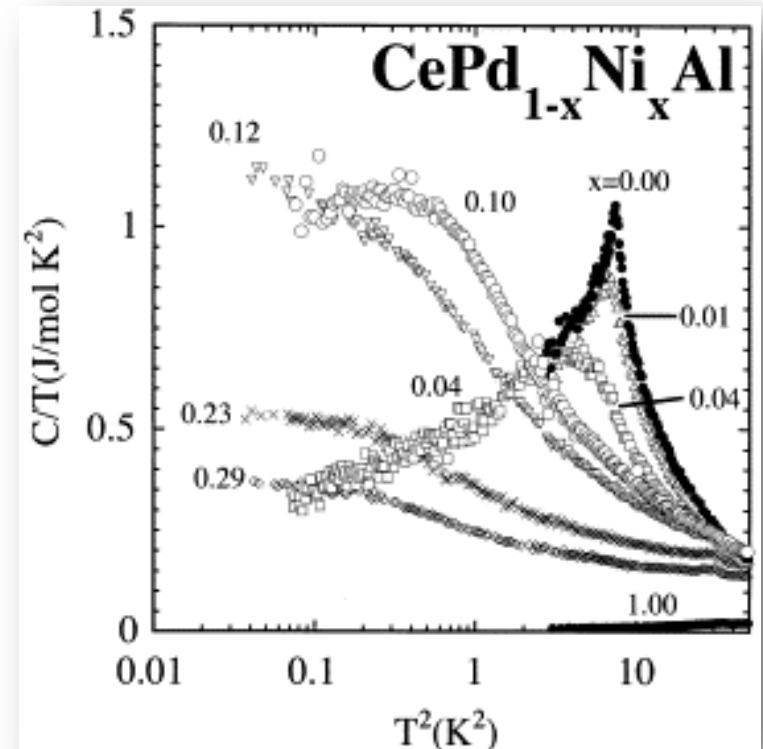
*Goto et al., J. Phys. Chem: Sol. 63, 1159 (2001)*

...

... or by isoelectronic Ni doping

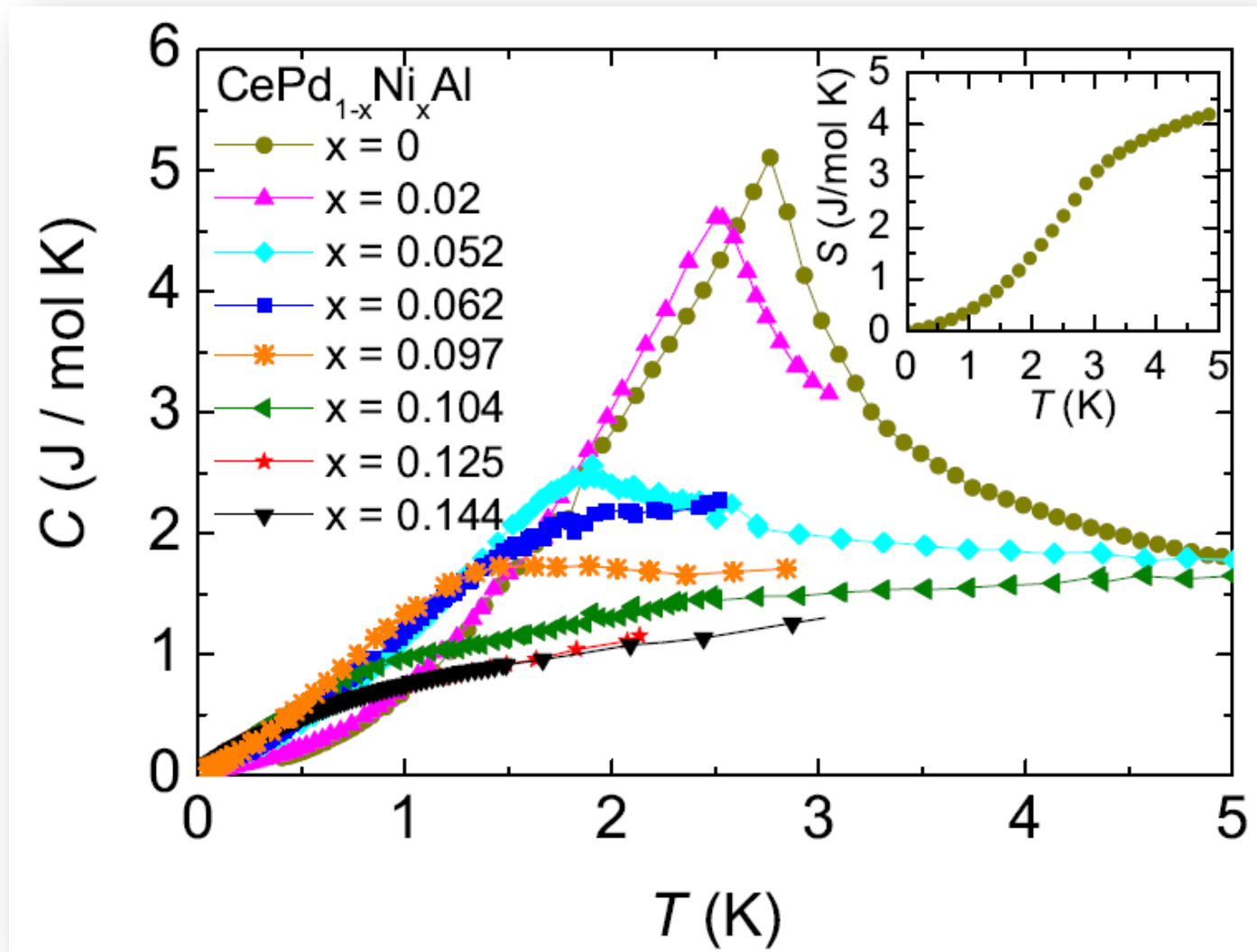
*Isikawa et al., Physica B 281&282, 36 (2000)*

*Bagrets, Fritsch et al., unpublished*



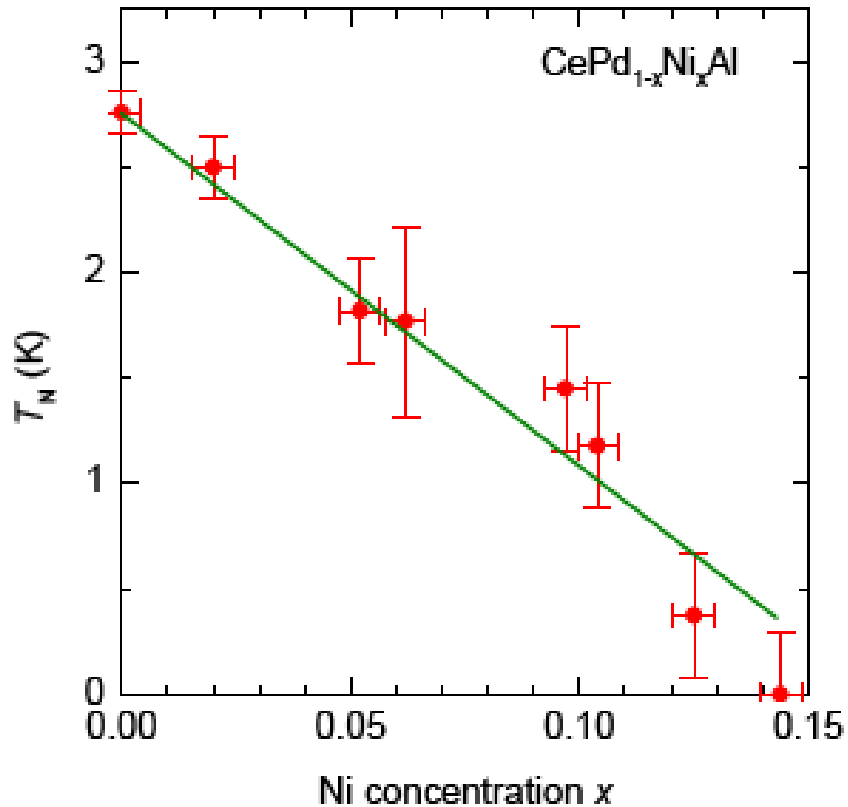
Quantum critical point?

# Specific heat of $\text{CePd}_{1-x}\text{Ni}_x\text{Al}$ polycrystals



Specific-heat anomaly at  $T_N$  broadens and is completely suppressed around  $x = 0.14$

# $T_N(x)$ of $\text{CePd}_{1-x}\text{Ni}_x\text{Al}$ polycrystals

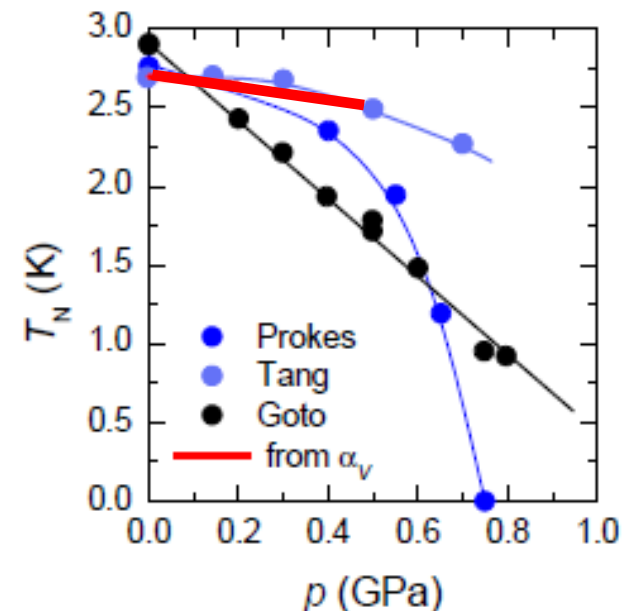


*V. Fritsch et al., unpublished*

Best fit with linear  $T_N$  dependence on  $x$ , compatible with 2D HMM scenario, deviation for  $x \rightarrow x_c$  (“order by disorder“?)

Comparison of pressure and Ni substitution:  $T_N(V(x))$  and  $T_N(V(p))$ ?  
Experimental  $T_N(p)$  data differ strongly!

Likely reason: non-hydrasticity of  $p$ .  
Thermal expansion:  $\alpha \parallel c < 0$ ,  $\alpha \perp c > 0$   
 $\rightarrow dT_N/dp_a > 0$  and  $dT_N/dp_c < 0$ .



# Approaching quantum criticality of CePdAl by Ni substitution

Specific heat

$T_N \rightarrow 0$  for  $x \approx 0.14$

$C/T \sim -\log(T/T_0)$

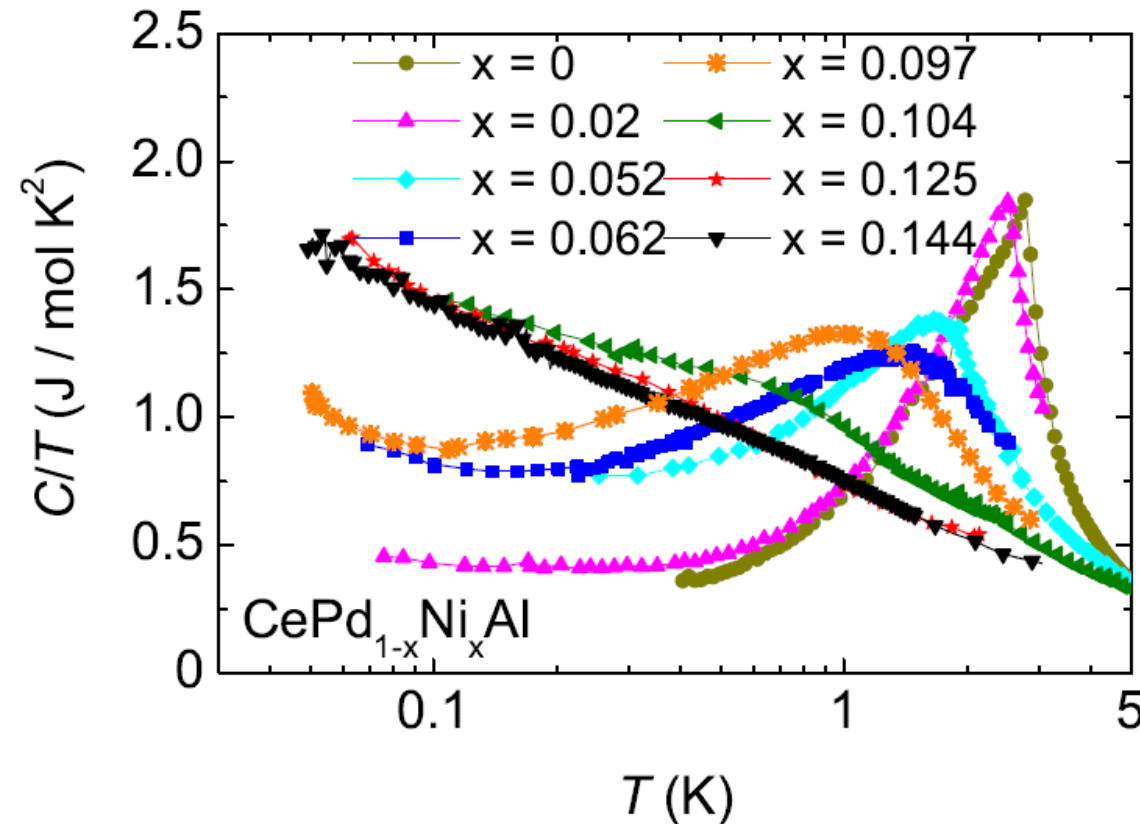
2D quantum criticality  
or novel QCP?

cf.  $\rho(T)$  of CePdAl

at  $p = 1 - 1.2$  GPa:

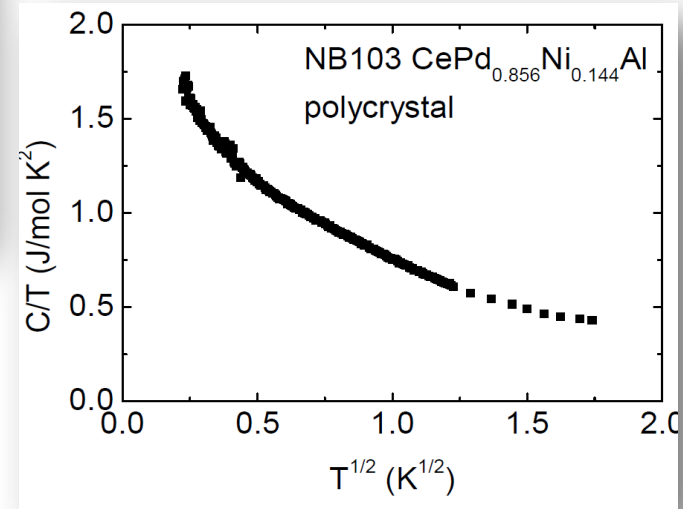
$\rho(T) \sim \rho_0 + AT^n$

*Goto et al., J. Phys. Chem. Sol. 63, 1159 (2002)*

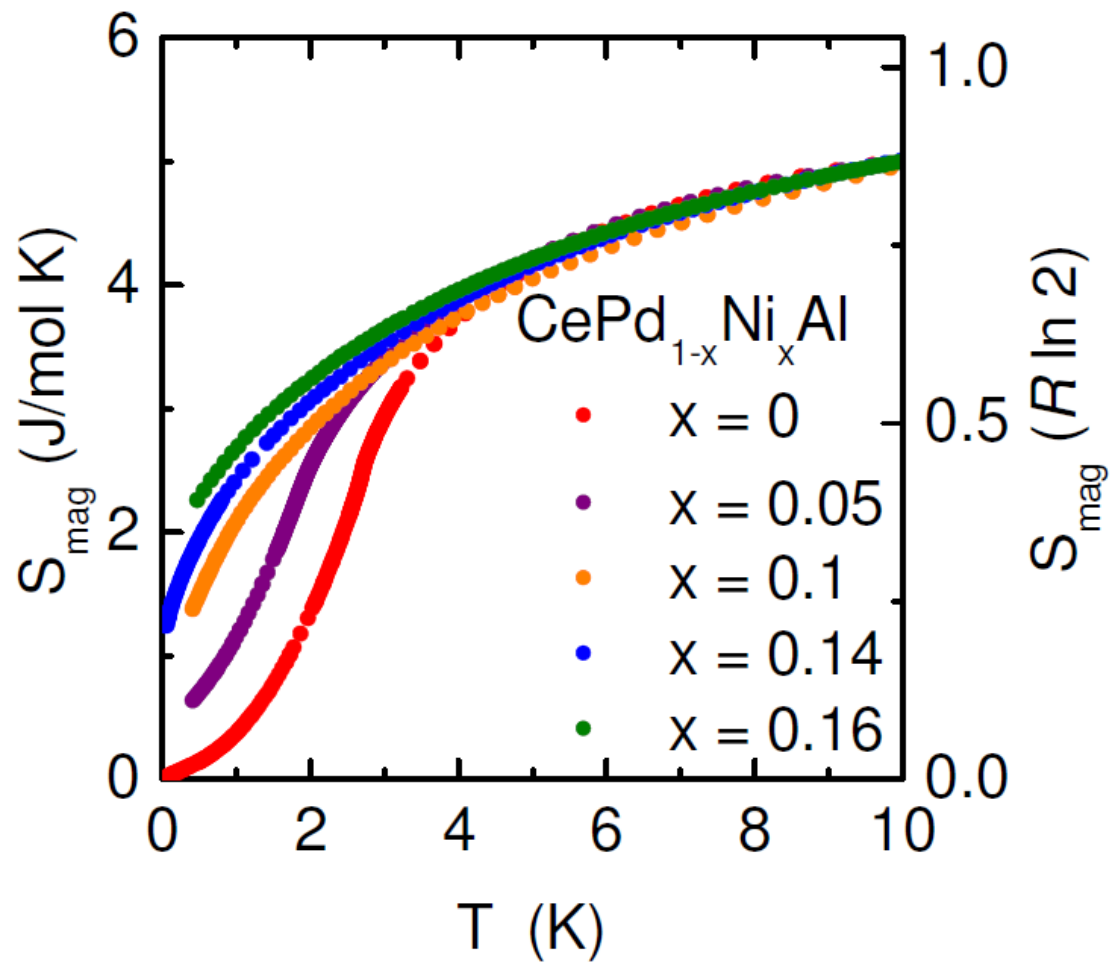
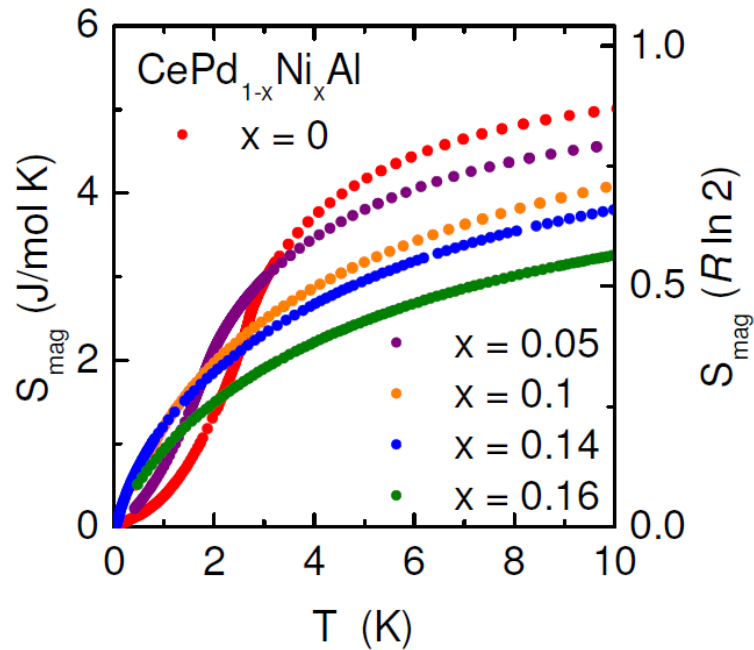


*V. Fritsch et al., PRB 2014*

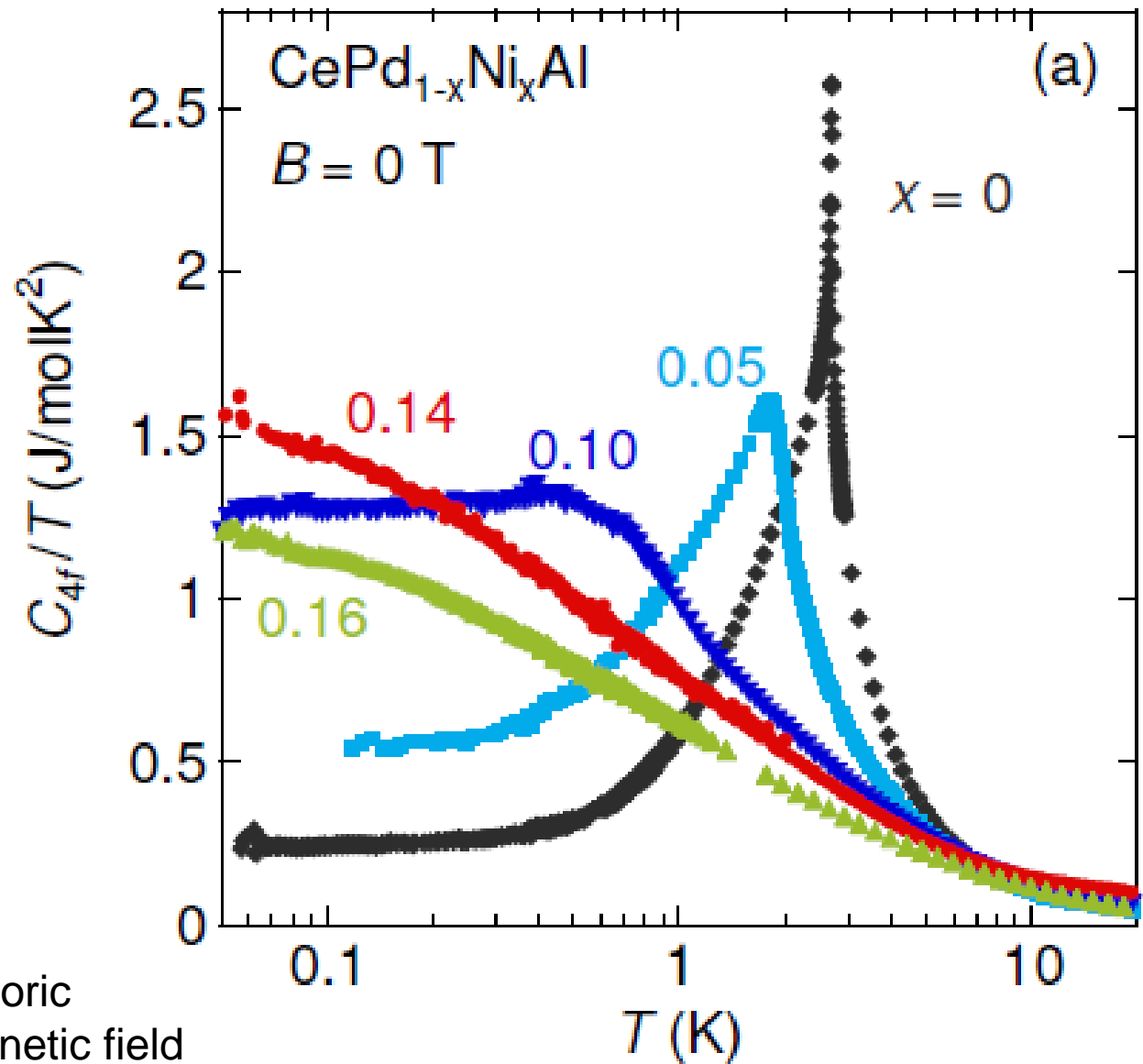
Data do not follow  $C/T \sim 1 - \sqrt{T}$  (3D QCP)



# Entropy of $\text{CePd}_{1-x}\text{Ni}_x\text{Al}$

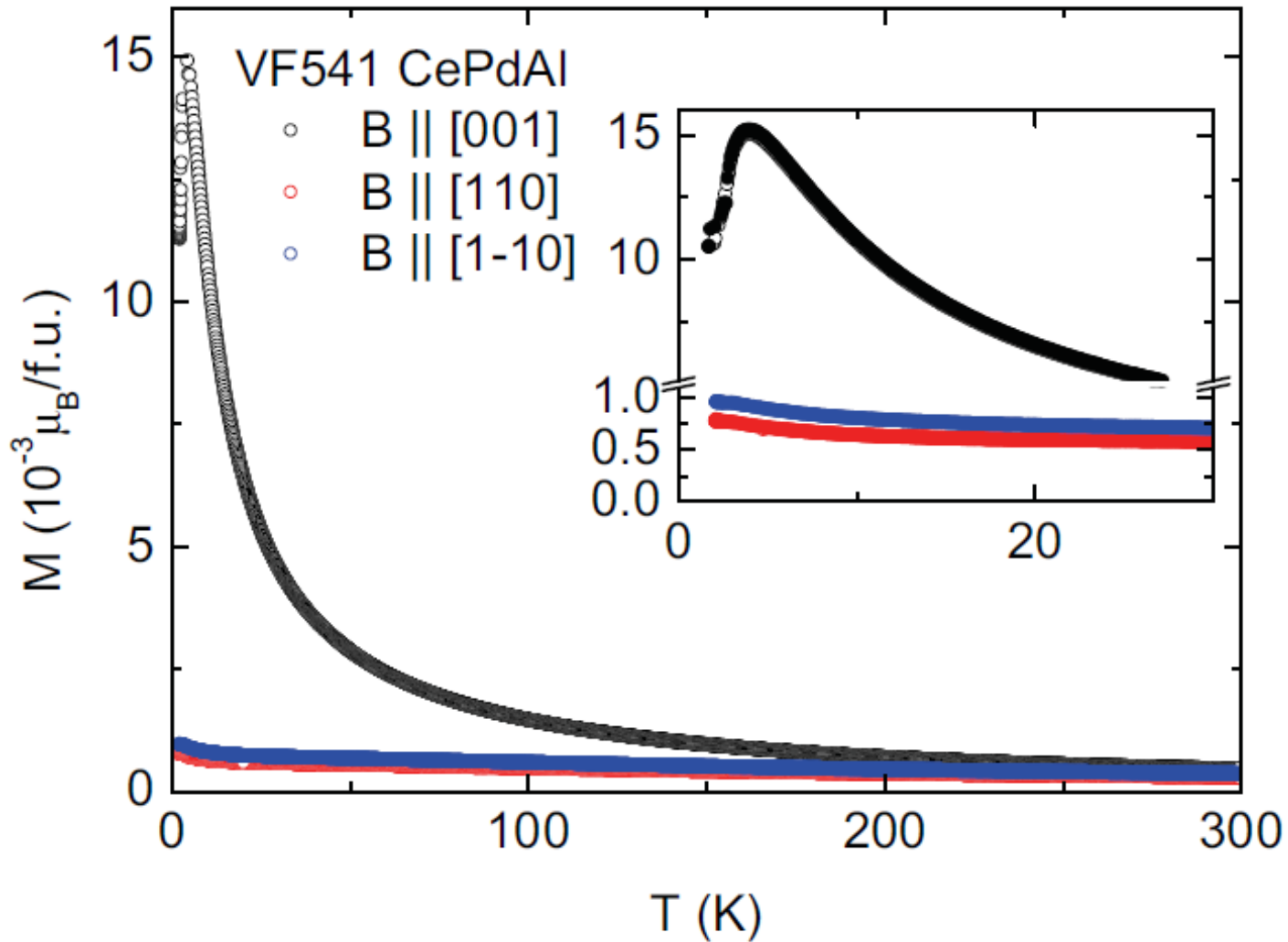


# First experiments on $\text{CePd}_{1-x}\text{Ni}_x\text{Al}$ single crystals



Planned: magnetocaloric  
measurements in magnetic field

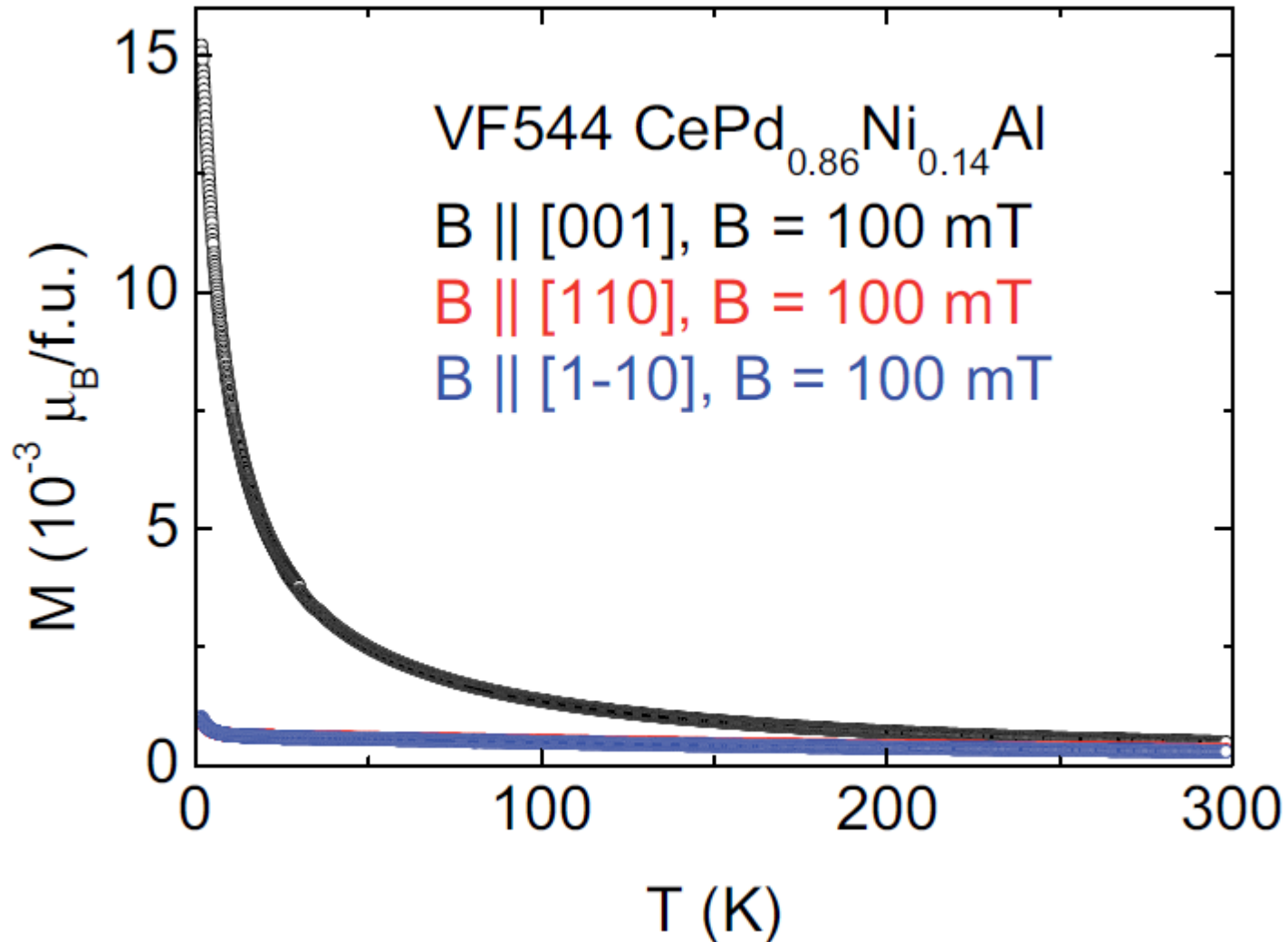
# Magnetic susceptibility of a CePdAl single crystal



Strong Ising-like anisotropy due to single-ion crystal-field effects



# Magnetic susceptibility of $\text{CePd}_{1-x}\text{Ni}_x\text{Al}$ with $x = 0.14$

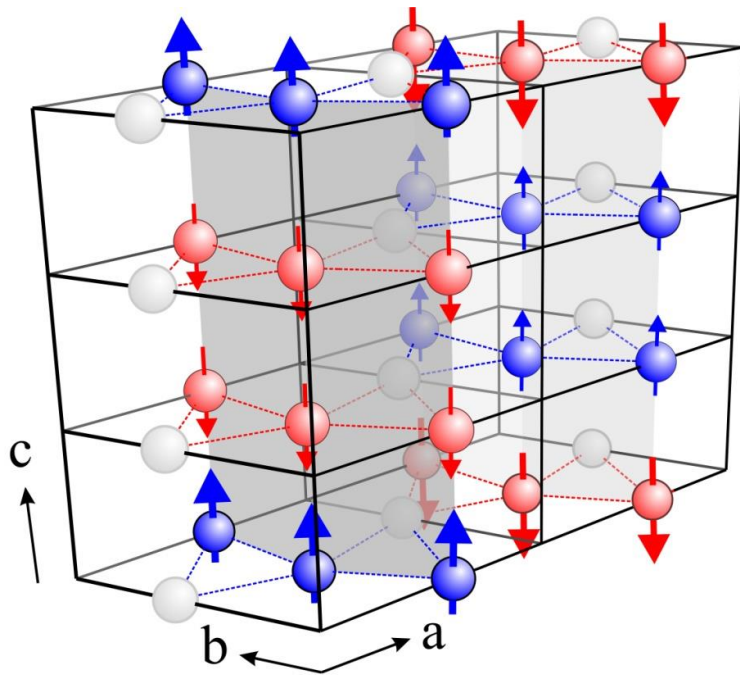


Ising-like anisotropy survives

# AF order and 2D quantum criticality in $\text{CePd}_{1-x}\text{Ni}_x\text{Al}$ ?

Interpretation within the Hertz-Millis-Moriya model:  
candidate for planes with 2D fluctuations?

planes perpendicular  $ab$



AF planes separated  
by frustrated moments

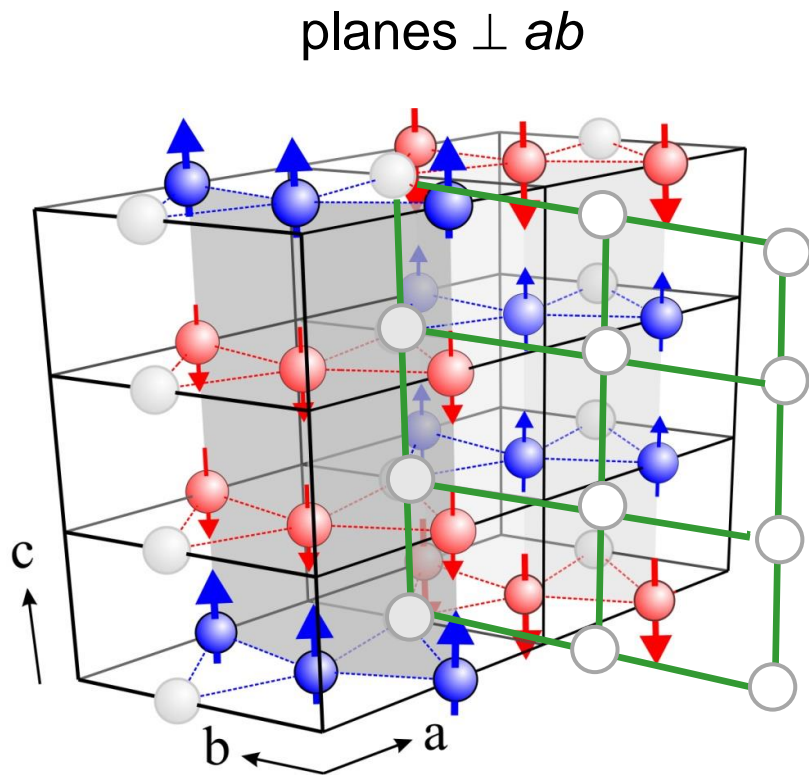
Proposition needs to be checked  
by inelastic neutron scattering

In this scenario, frustrated moments  
play a key role and provide  
a rationale for 2D fluctuations

However, frustrated moments  
may lead to additional fluctuations  
not contained in the HMM model

Frustrated Ce moments  
in CePdAl:  
a two-dimensional spin liquid?

# Spin liquid in CePdAl?



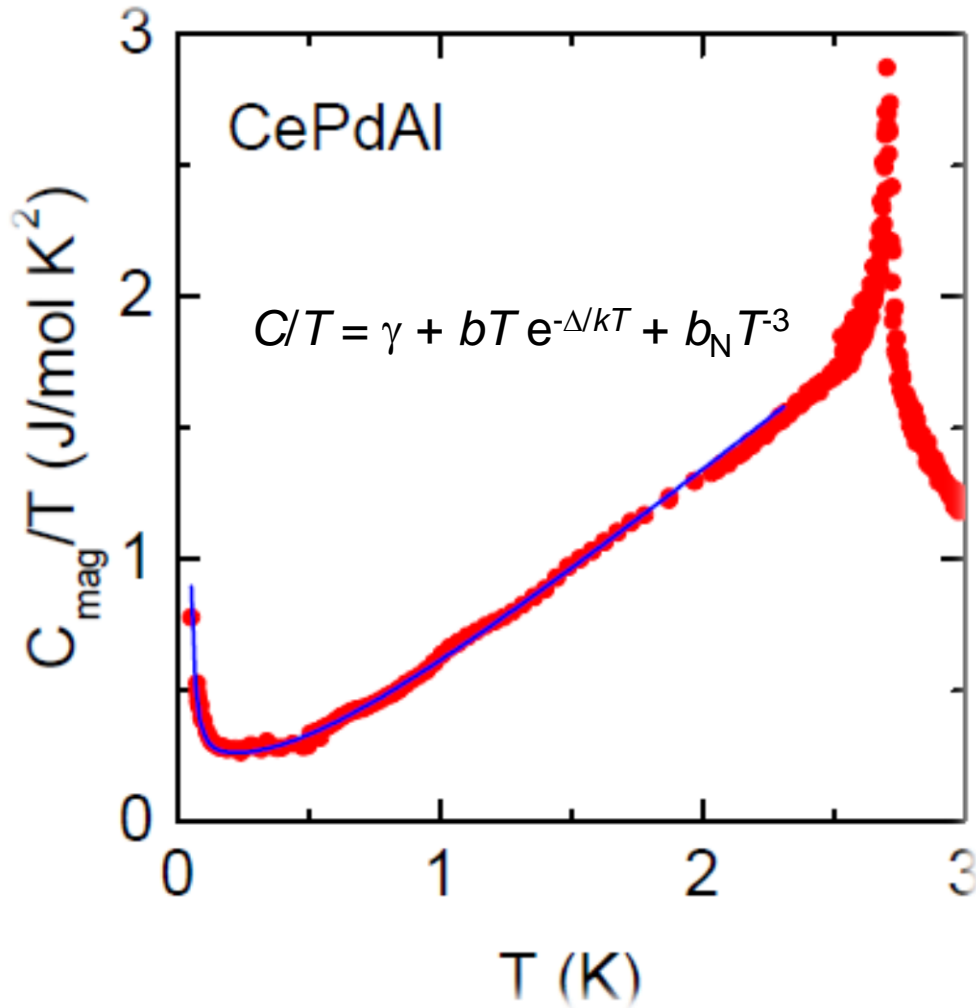
AF planes separated  
by frustrated moments

Frustrated planes between AF planes form  
a rectangular 2D lattice:  
2D Ising spin liquid?

$^{27}\text{Al}$  NMR measurements down to 30 mK:  
dynamics of frustrated moments prevails  
down to very low  $T$ , with  $T_1^{-1} \sim T$

*Oyamada et al., Phys. Rev. B 77, 064432 (2008).*

# Specific heat of CePdAl at low temperature



Several unusual features

large  $\gamma T$  term

$$\gamma \sim 0.8 \text{ J/mole}_{\text{Ce-no}} \text{K}^2$$

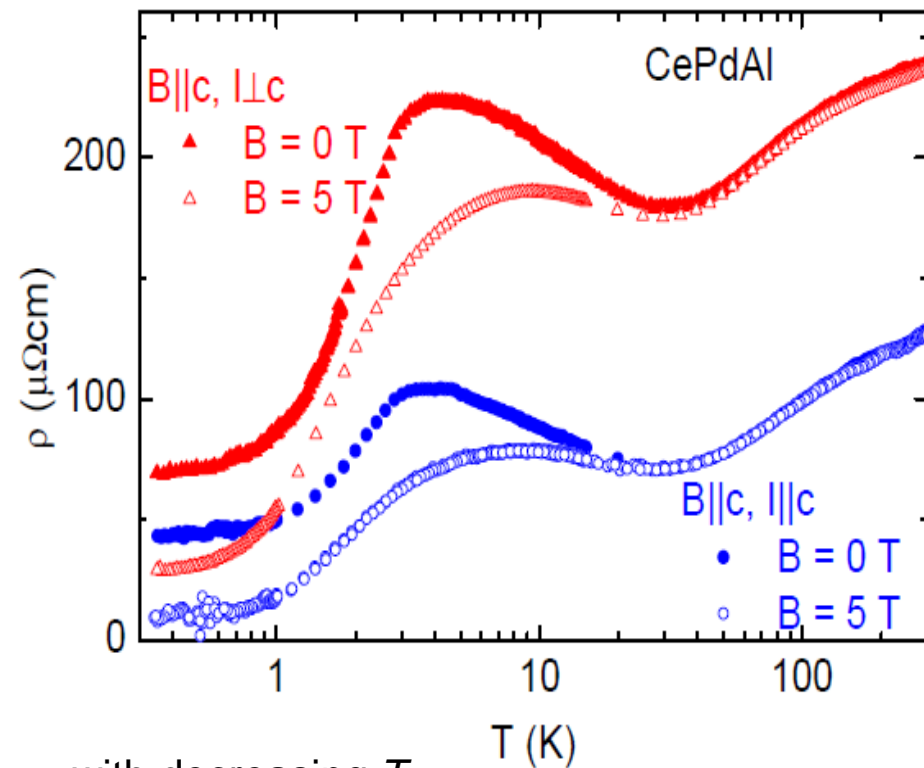
$T^2$  term  $\sim T^2$  setting in at 0.5 K

excitation gap:  $\Delta / k_B \approx 0.9 \text{ K}$

2D spin waves in AF planes

$T^{-2}$  contribution at low  $T$  presumably  
due to nuclear hyperfine splitting

# Electrical resistivity of CePdAl single crystals

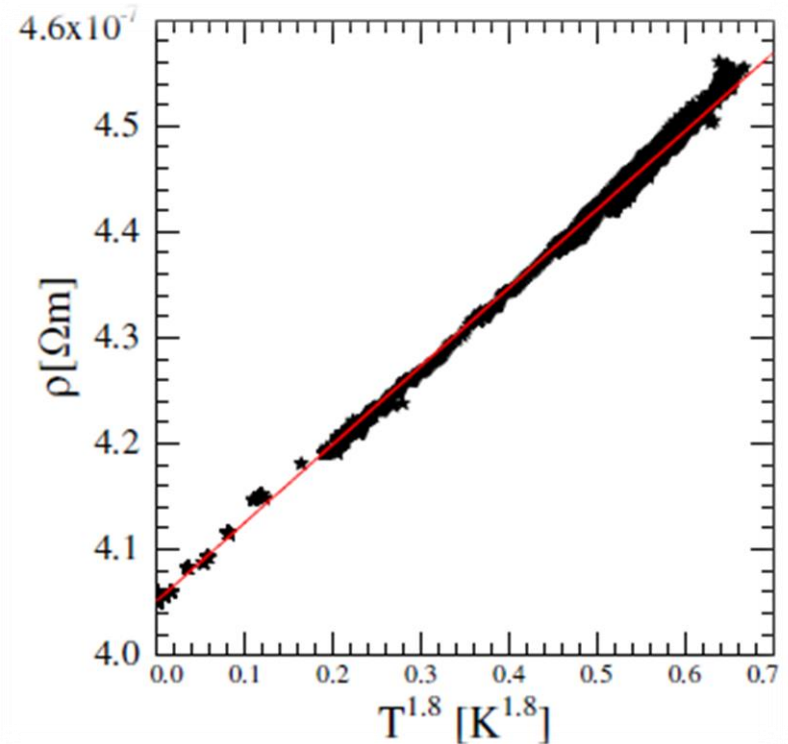


with decreasing  $T$ :

- Kondo increase
- coherence maximum
- drop to  $\rho_0$

strong decrease of the residual resistivity  $\rho_0$   
in magnetic field above  $B_c$ :

$$\Delta\rho_0/\rho_0 \text{ strongest for } \rho \parallel c$$



at lowest temperature:  $\rho(T) = \rho_0 + AT^{1.8}$

- no indication of Kondo effect  
by non-ordered frustrated Ce moments
- assuming  $T^2$  resistivity:

$$A/\gamma^2 \sim 13 a_{\text{KW}}$$

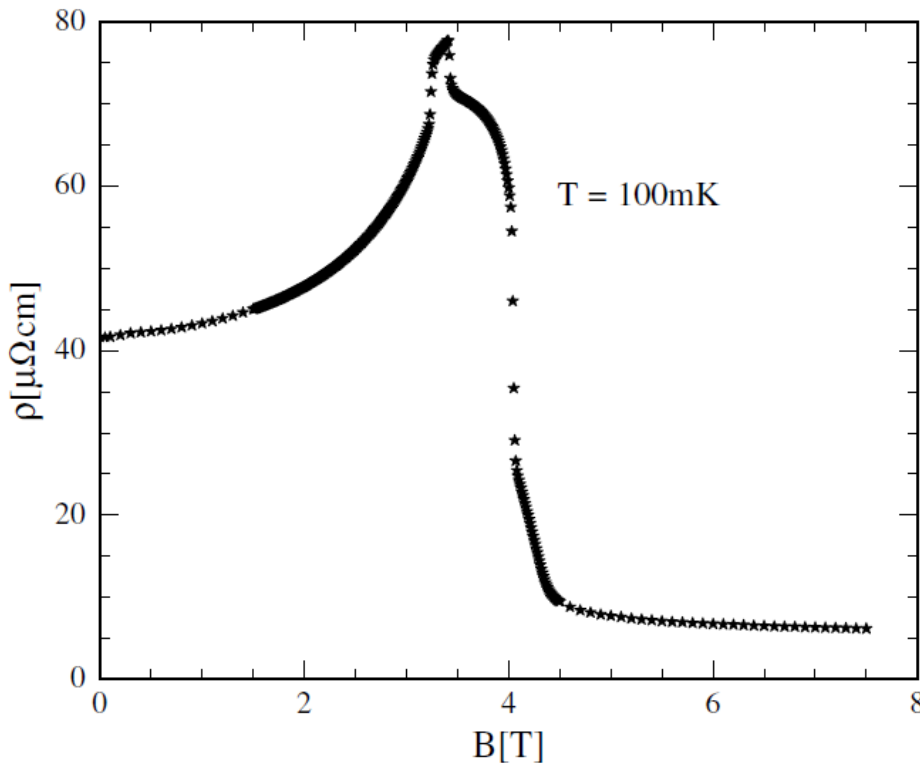
Spinon excitations?

# Field-induced phases in CePdAl close to the critical field

Features in magnetization  $M(B)$  and resistivity  $\rho(T)$  are suggestive of first-order transitions

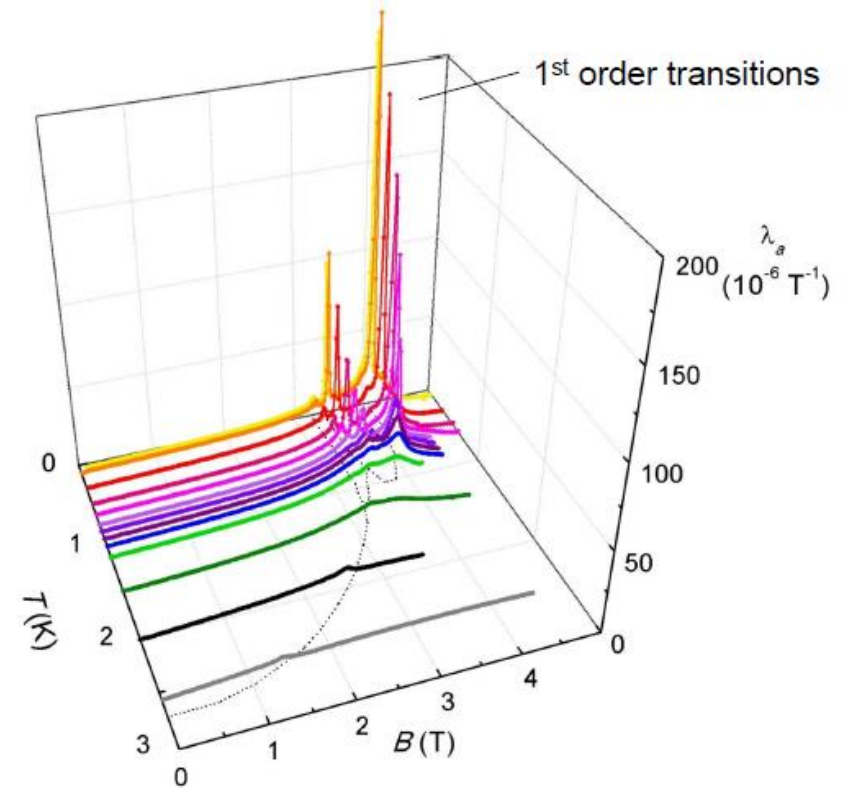
*T. Goto et al., J. Phys. Chem. Sol. 63, 1159 (2002)*

Resistivity at 100 mK



*C. Taubenheim et al.*

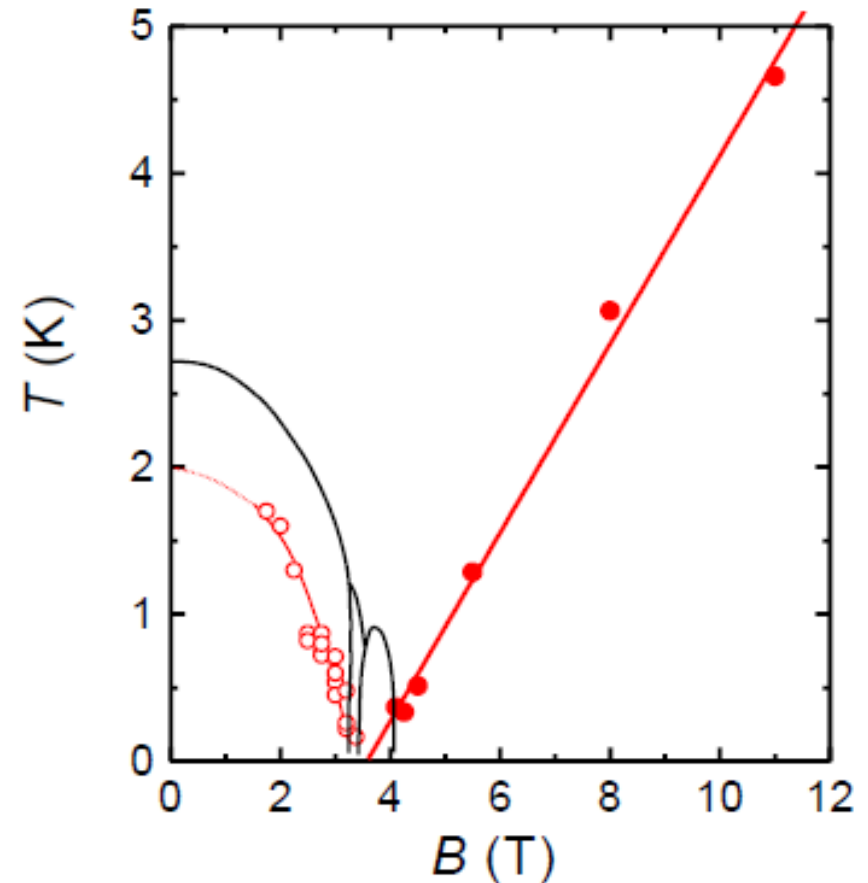
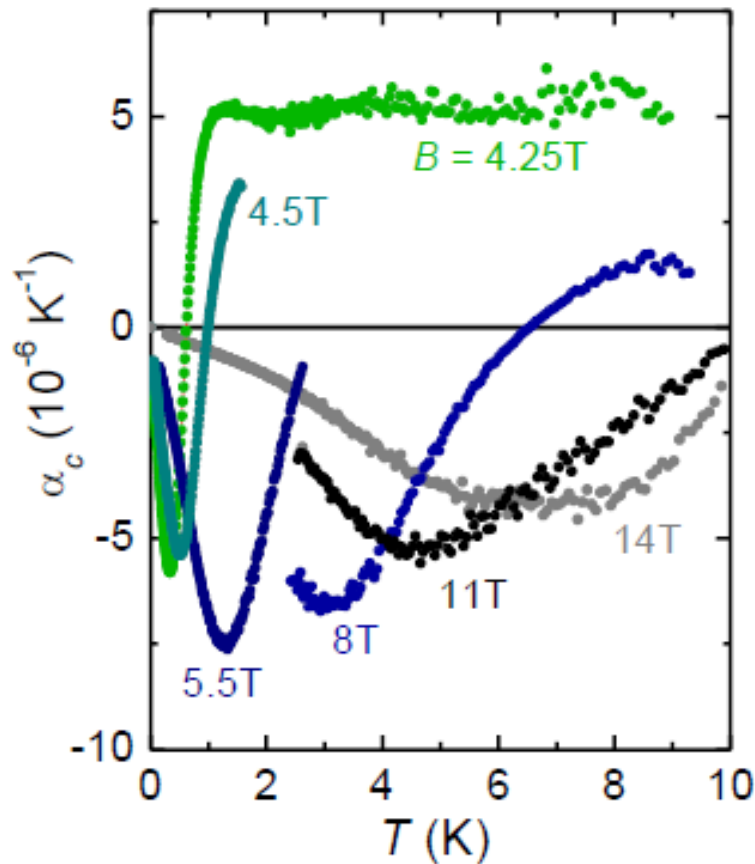
Magnetostriction



*K.Grube et al.*



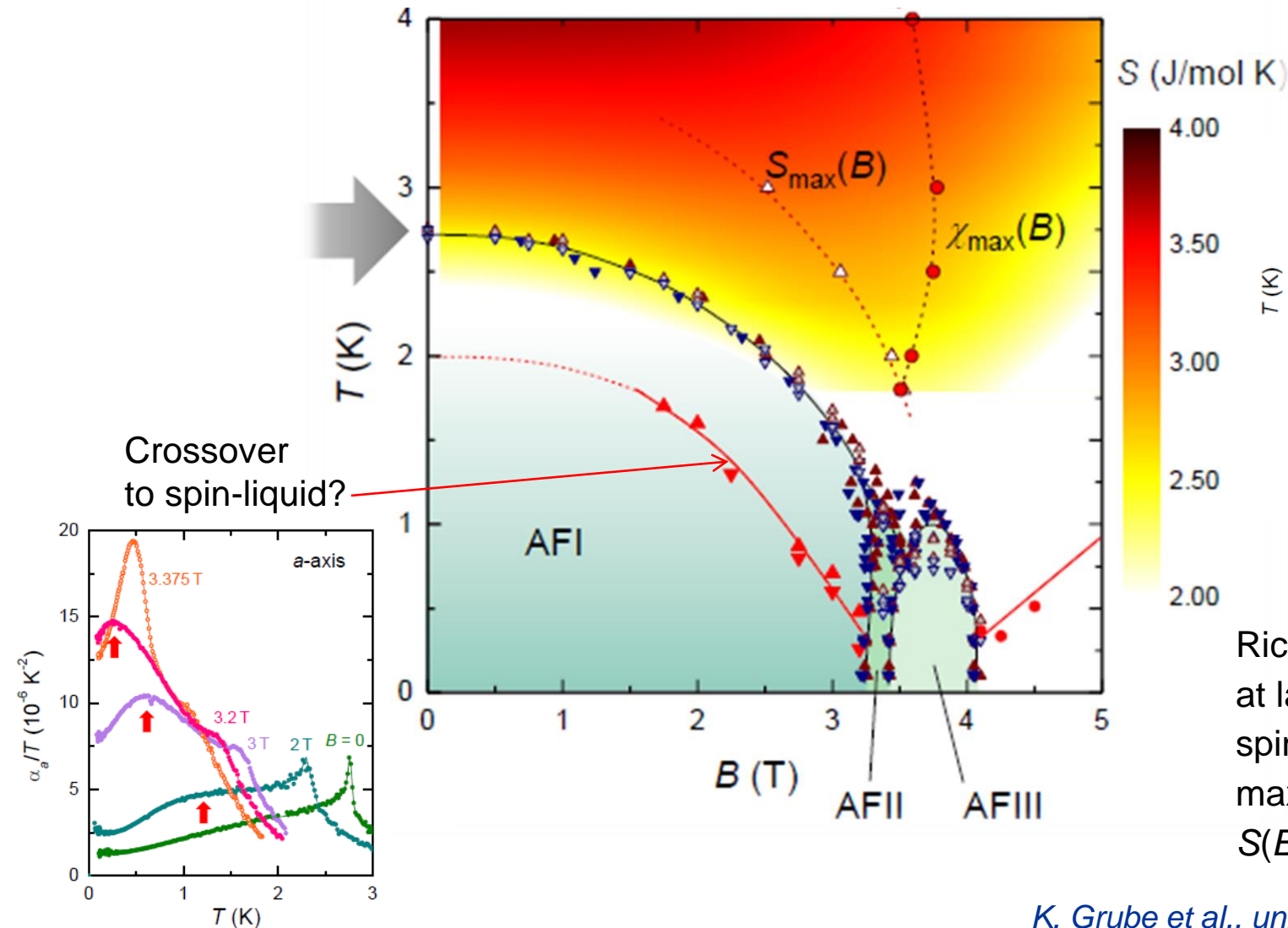
# High-field thermal expansion of CePdAl



Extrema in  $\alpha_c$  above  $B_c$ : due to Zeeman splitting of the lowest CEF doublet?

$$k_B T_{max} \sim g \mu (B - B_c)$$

# Magnetic phase diagram of CePdAl from thermal expansion and magnetostriction

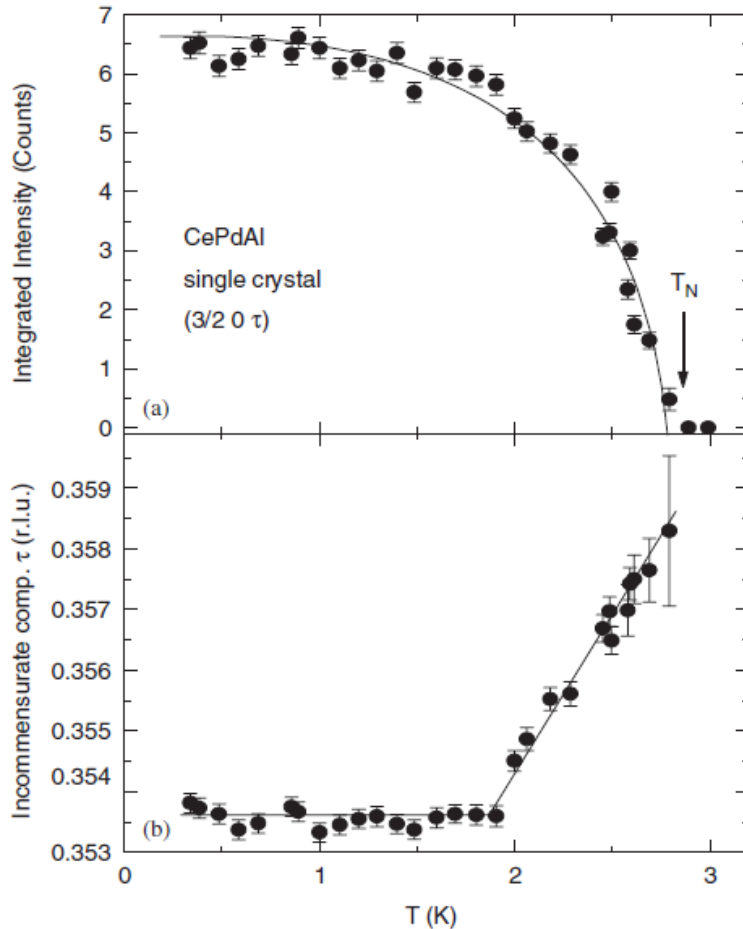


High-field Schottky anomaly due to Zeeman splitting

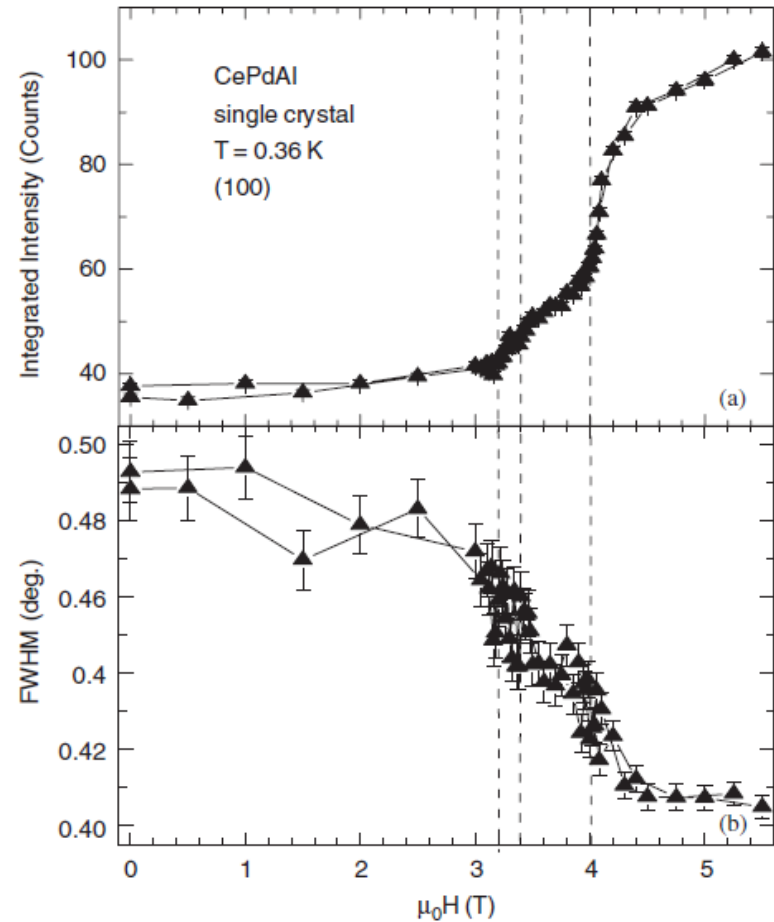
Rich phase diagram at large  $B$  and low  $T$ : spin liquid freezes out, maximum of  $S(B, T = \text{const})$

# Towards a $(B, T)$ phase diagram from elastic neutron scattering

Lock-in of the  $c$  component  $\tau$  of the magnetic propagation vector at 1.9 K: relation to feature in  $\alpha_a(T)$  ?



Evolution of ferromagnetic component with magnetic field



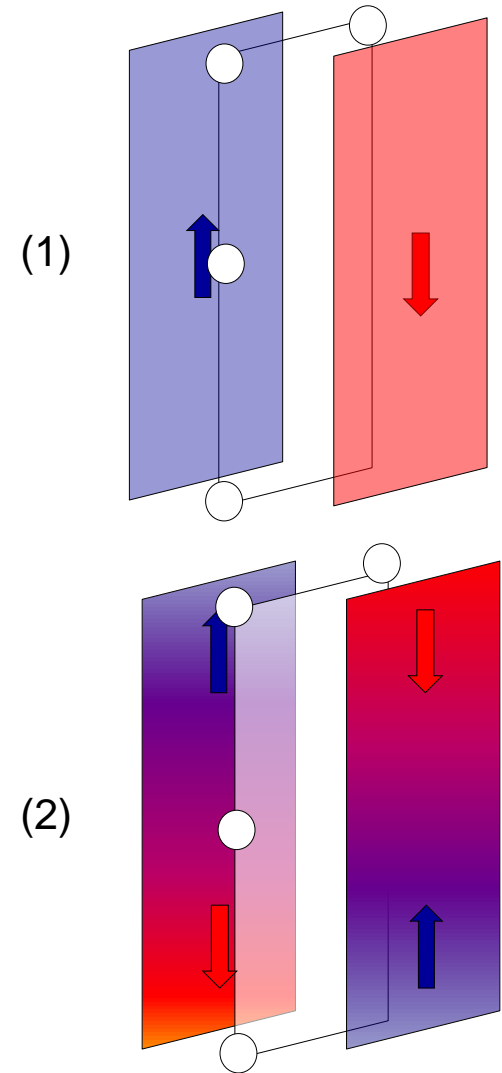
# Fate of the Kondo effect in the presence of frustration?

If we now naively generalize this single-impurity model to the lattice, we will find that the  $T=0$  ground state always has Kondo screening. It is only upon including frustrating intermoment exchange interactions—equivalent to having “dispersing” spinons—that it is possible to break down Kondo screening and reach a state in which the slave boson is not condensed.

*Senthil, Vojta, Sachdev, PRB 69, 135111 (2004)*

A pedestrian's approach to the problem

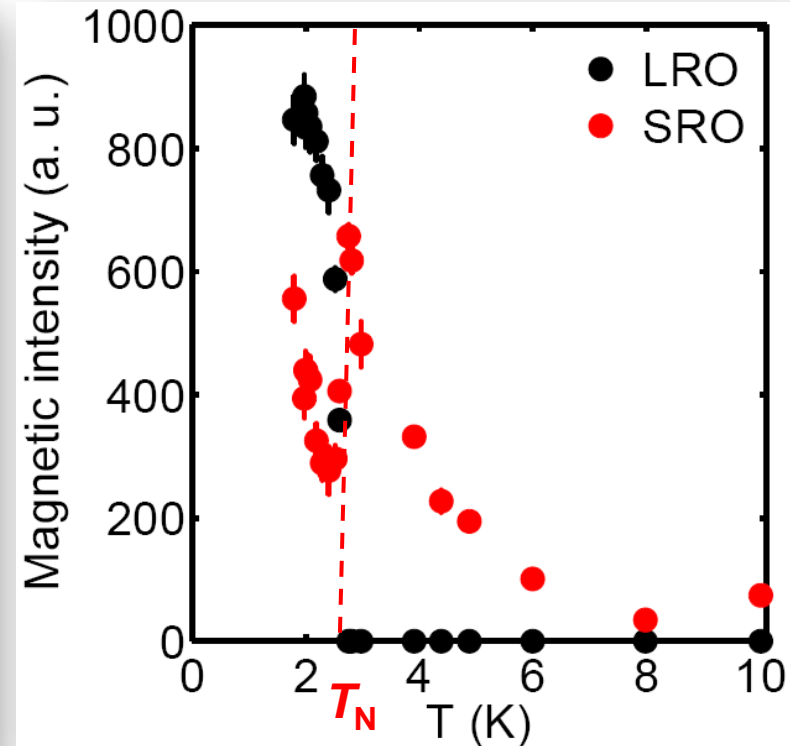
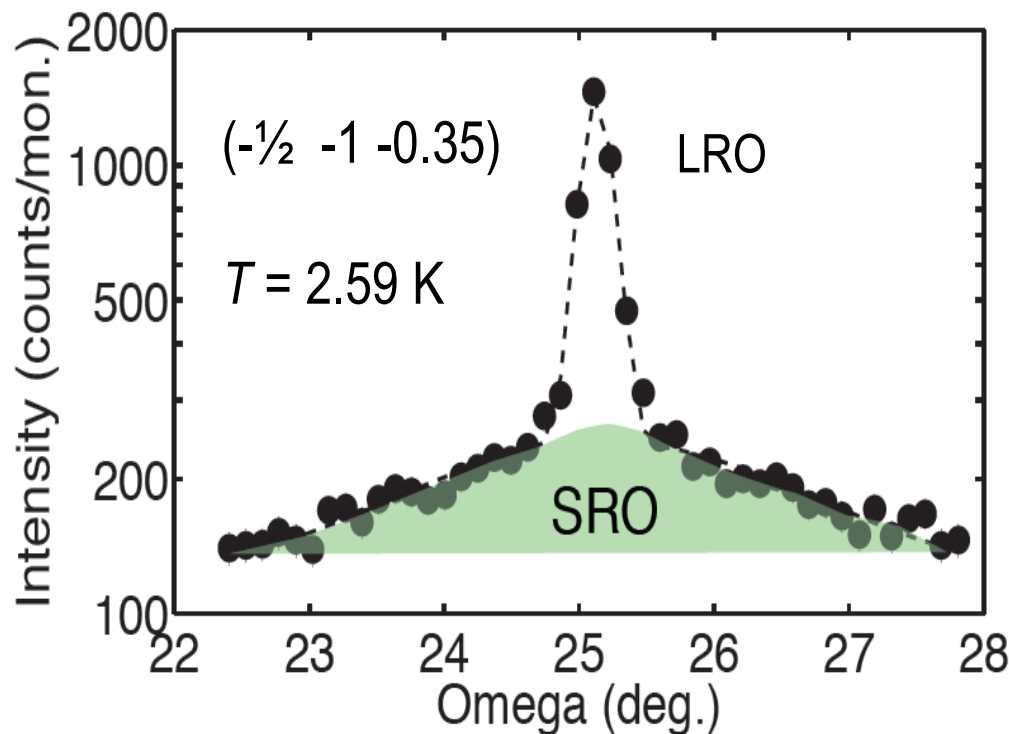
- (1) 2D rectangular Kondo lattice exposed to frustrating molecular field within an  $ab$  layer?
- (2) Include staggered magnetization.
- (3) Additional complication:  $6s^25d^1$  electrons.



# Long-range and short-range magnetic order in CePdAl

D20 ILL

*O. Stockert et al., unpublished*



LRO/SRO intensity ratio of 2/1 below  $T_N$ :

compatible with short-range (dynamic?) order of frustrated moments

→ rationale for quasi-2D fluctuations

cf. NMR measurements

*Oyamada et al., Phys. Rev. B 77, 064432 (2008).*

Classical and quantum  
phase transitions in the  
itinerant ferromagnet



# A brief history of $\text{Sr}_{1-x}\text{Ca}_x\text{RuO}_3$

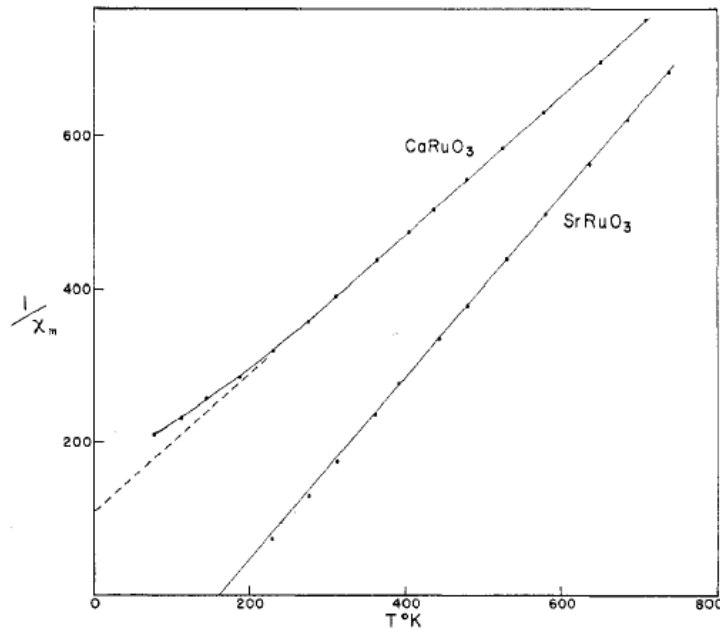
$\text{SrRuO}_3$  is the  $n = \infty$  member of the Ruddlesden-Popper series  $\text{Sr}_{n+1}\text{Ru}_n\text{O}_{3n+1}$

Most prominent are the  $n = 1$  and  $n = 2$  materials

$\text{Sr}_2\text{RuO}_4$  odd-parity superconductor

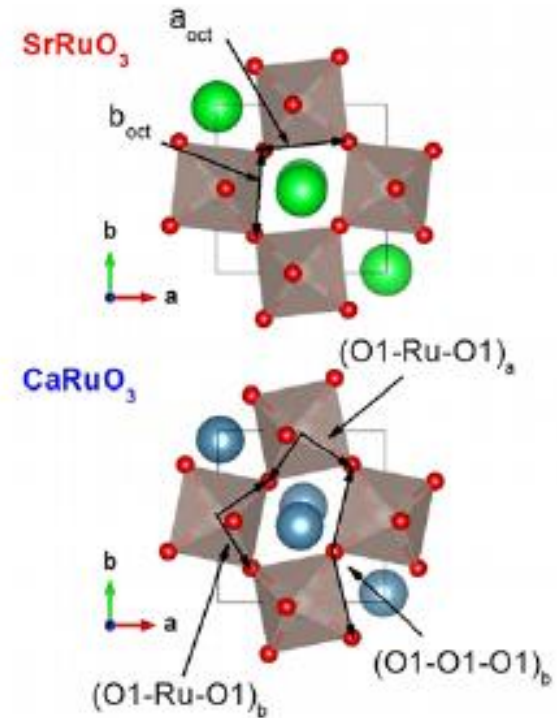
$\text{Sr}_3\text{Ru}_2\text{O}_7$  anomalous quantum criticality

## Discovery of ferromagnetism



A. Callaghan et al., *Inorg. Chem.* **5**, 1572 (1966)

Structural differences between  $\text{SrRuO}_3$  and  $\text{CaRuO}_3$



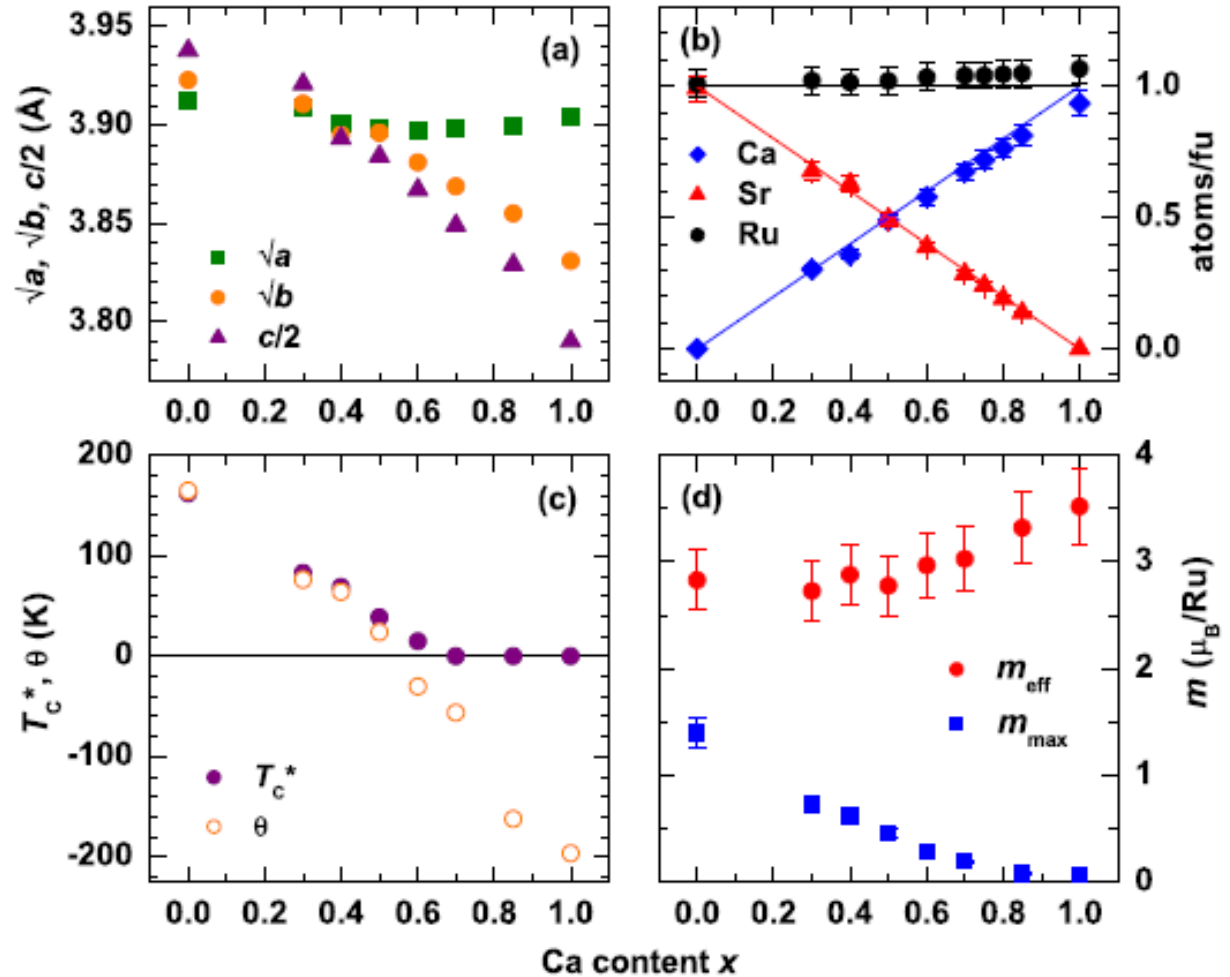
S. Lee et al., *J. Phys. Cond. Matt.* **25**, 465601 (2013)



# Structural and magnetic data of $\text{Sr}_{1-x}\text{Ca}_x\text{RuO}_3$

Polycrystalline samples:

- Sintering of  $\text{SrCO}_3$ ,  $\text{CaCO}_3$ , and  $\text{RuO}_2$  powders
- Calcination at  $900^\circ\text{C}$ , 10 h
- Milling and pelleting and sintering at  $1370^\circ\text{C}$ , 30 h



Evolution of critical exponents  
in  $\text{Sr}_{1-x}\text{Ca}_x\text{RuO}_3$  with  $x$

# Critical exponents: single crystals vs. polycrystals

Two cases

- (1) Magnetic easy axis varies on a length scale  $< \xi$ : random anisotropy changes the universality class of the transition
- (2) Crystallites ( $\sim$  several  $\mu\text{m}$ )  $\gg \xi$ : individual crystallites obey scaling relations. Each crystallite is exposed to an effective field

$$H_{\text{eff}} = H \times f(\theta, \varphi)$$

leading to a magnetization density  $M(\theta, \varphi)$ .

Averaging over crystallites with  $p(\theta, \varphi)$  gives the total magnetization density

$$M = \int \sin \theta d\theta d\varphi p(\theta, \varphi) M(\theta, \varphi) = t^\beta \tilde{\phi}(H/t^{\beta\delta}),$$

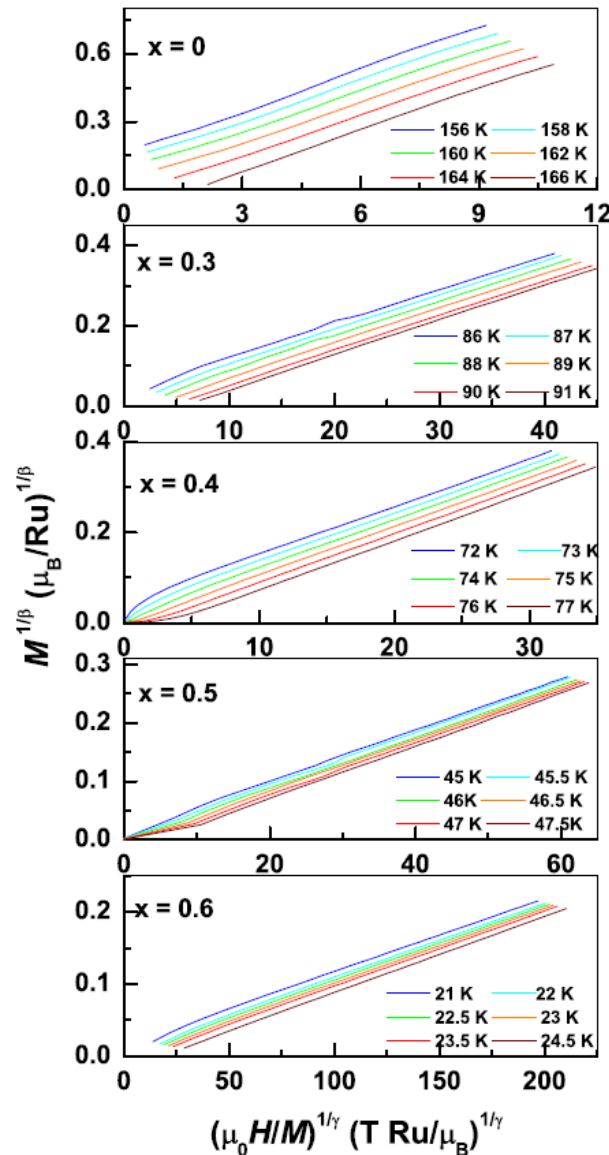
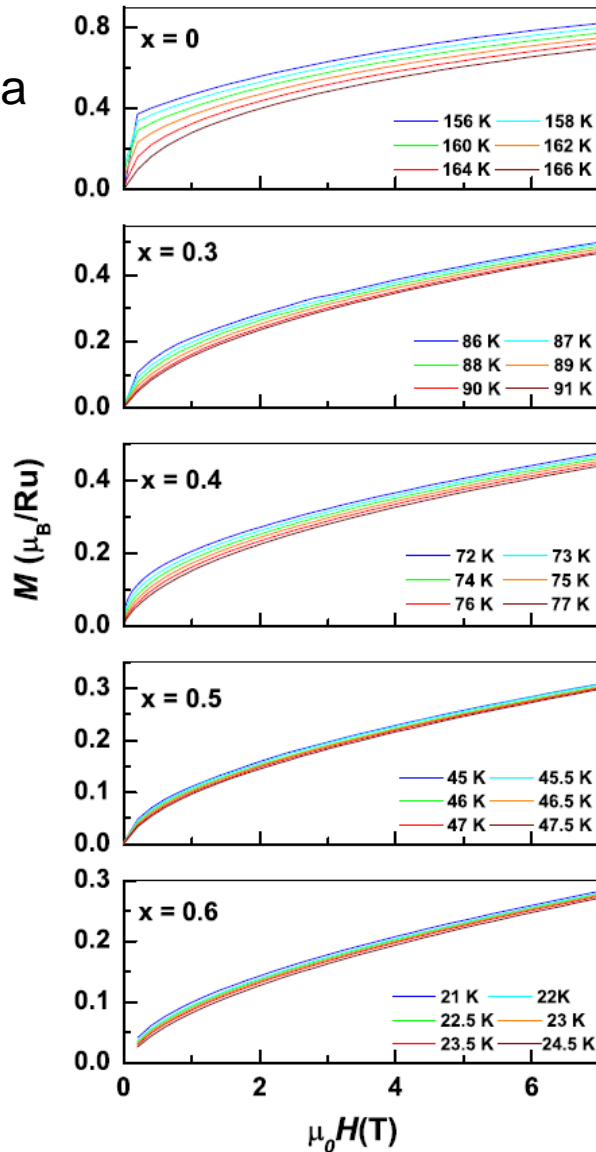
where

$$\tilde{\phi}(x) = \int \sin \theta d\theta d\varphi p(\theta, \varphi) \phi(f(\theta, \varphi), x).$$

This is the same scaling form with modified scaling function (except for pathological  $p(\theta, \varphi)$  distributions). In particular, the limits for small and large arguments, corresponding to Arrott-Noakes plots, are the same.

# Magnetization of ferromagnetic $\text{Sr}_{1-x}\text{Ca}_x\text{RuO}_3$

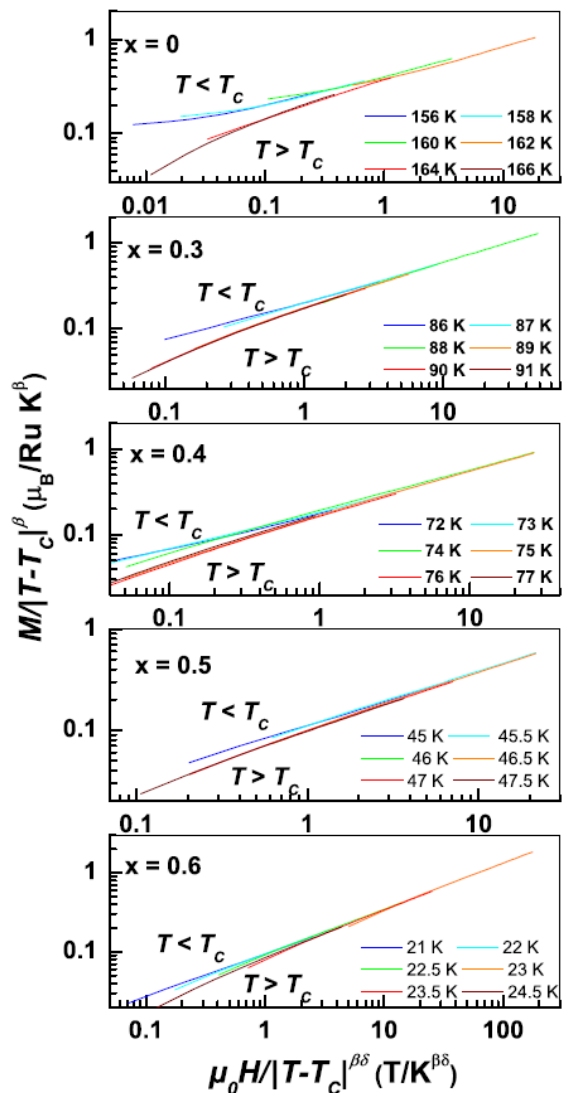
$M(H)$  data



Determination of critical exponents  $\beta$  and  $\gamma$  from modified Arrott plots

*D. Fuchs et al.,  
PRB 89, 147405 (2014)*

# Scaling analysis of the finite- $T$ transitions in $\text{Sr}_{1-x}\text{Ca}_x\text{RuO}_3$



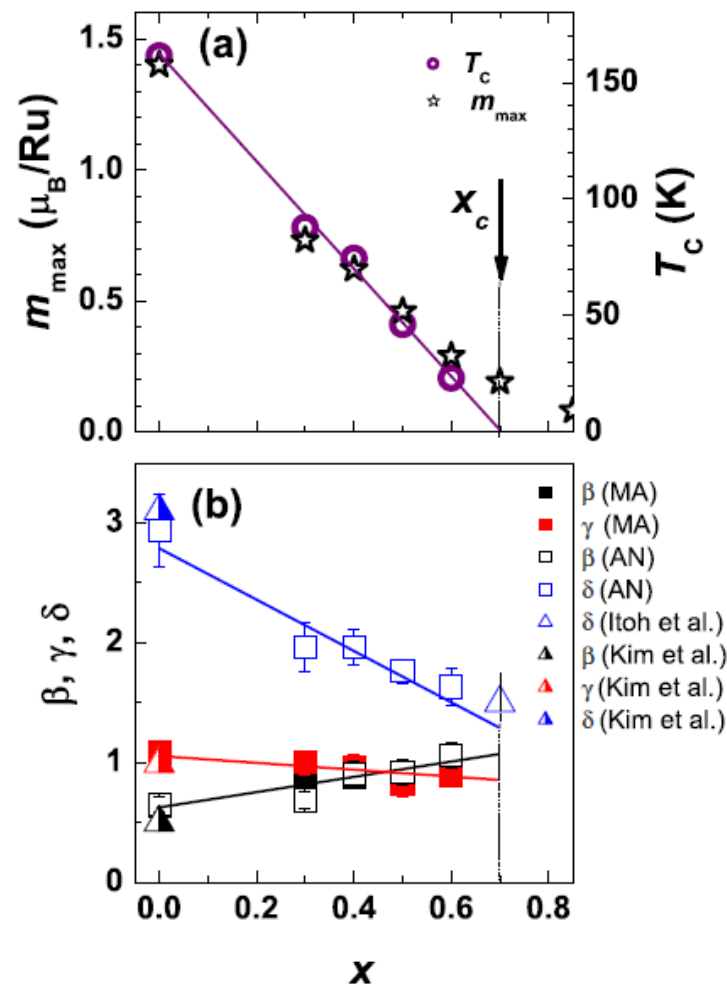
$$M(t, H) = t^\beta \phi(H/t^{\beta\delta})$$

$x = 0$   
 $\beta \approx 0.5, \gamma \approx 1, \delta \approx 3$

$x > 0$   
 continuous change  
 of  $\beta, \gamma$ , and  $\delta$

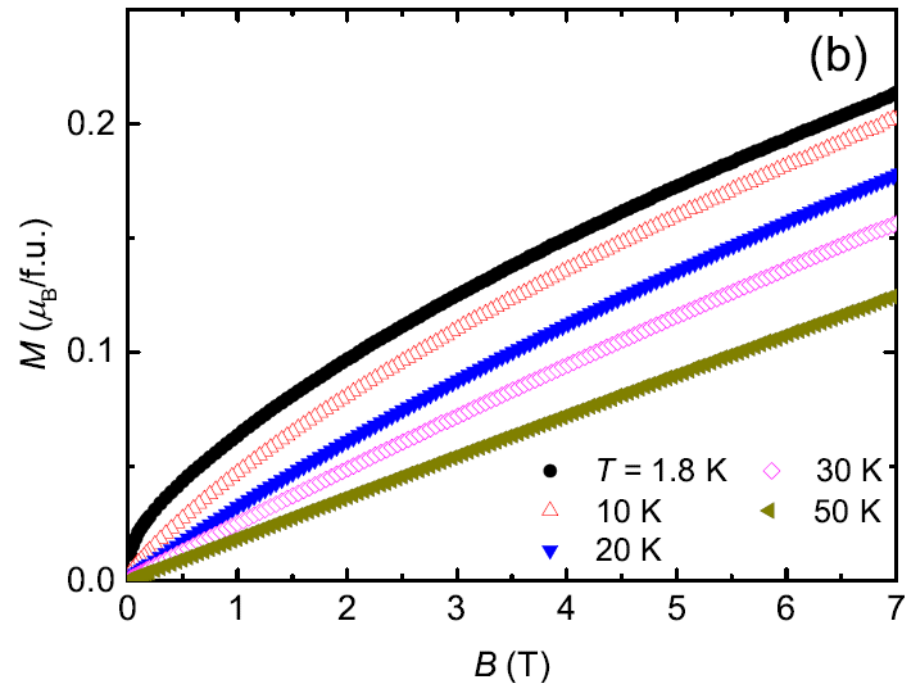
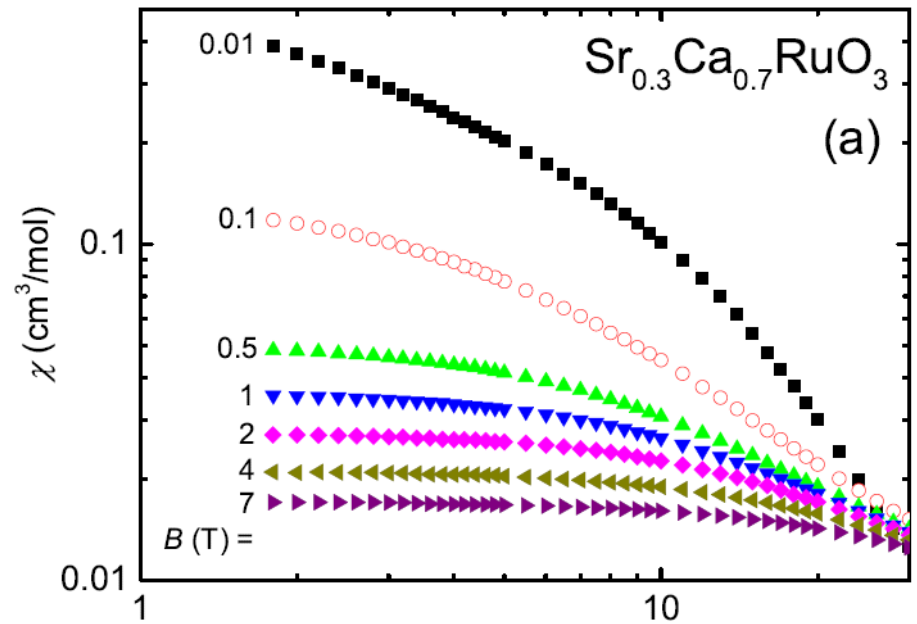
Widom relation  
 $\gamma/\beta = \delta - 1$   
 approximately obeyed  
 for all  $x \leq 0.6$

*D. Fuchs et al.,  
 PRB 89, 147405 (2014)*



# Unusually slow quantum critical dynamics

# Magnetization and susceptibility of $\text{Sr}_{0.3}\text{Ca}_{0.7}\text{RuO}_3$





# Scaling relations near a quantum critical point

Scaling law of the free energy at a QCP

$$\mathcal{F}(T, B) = b^{-(d+z)} \mathcal{F}(b^z T, b^y B),$$

yields for the magnetization

$$M(T, B) = -\frac{\partial \mathcal{F}}{\partial B} = b^{y-(d+z)} M(b^z T, b^y B).$$

Setting the scale factor  $b^z T = 1$ , or  $b^y B = 1$  for  $T \rightarrow 0$  gives

$$M(T, B) = T^{\frac{d+z-y}{z}} \Phi\left(\frac{B}{T^{y/z}}\right), \text{ or } M(B) \propto B^{\frac{d+z-y}{y}} \propto B^{1/\delta}$$

The correlation length exponent  $\nu$  can be obtained by determining the magnetization as function of  $r = x_c - x$  because  $\xi \sim |r|^{-\nu}$ :

$$M(r) = r^{\nu(d+z-y)} M(r=0) \sim |r|^\beta$$

Hence  $\beta = \nu(d+z-y)$ . With  $\delta$  we obtain

$$y = \delta\beta/\nu.$$

The critical part of the specific heat scales as

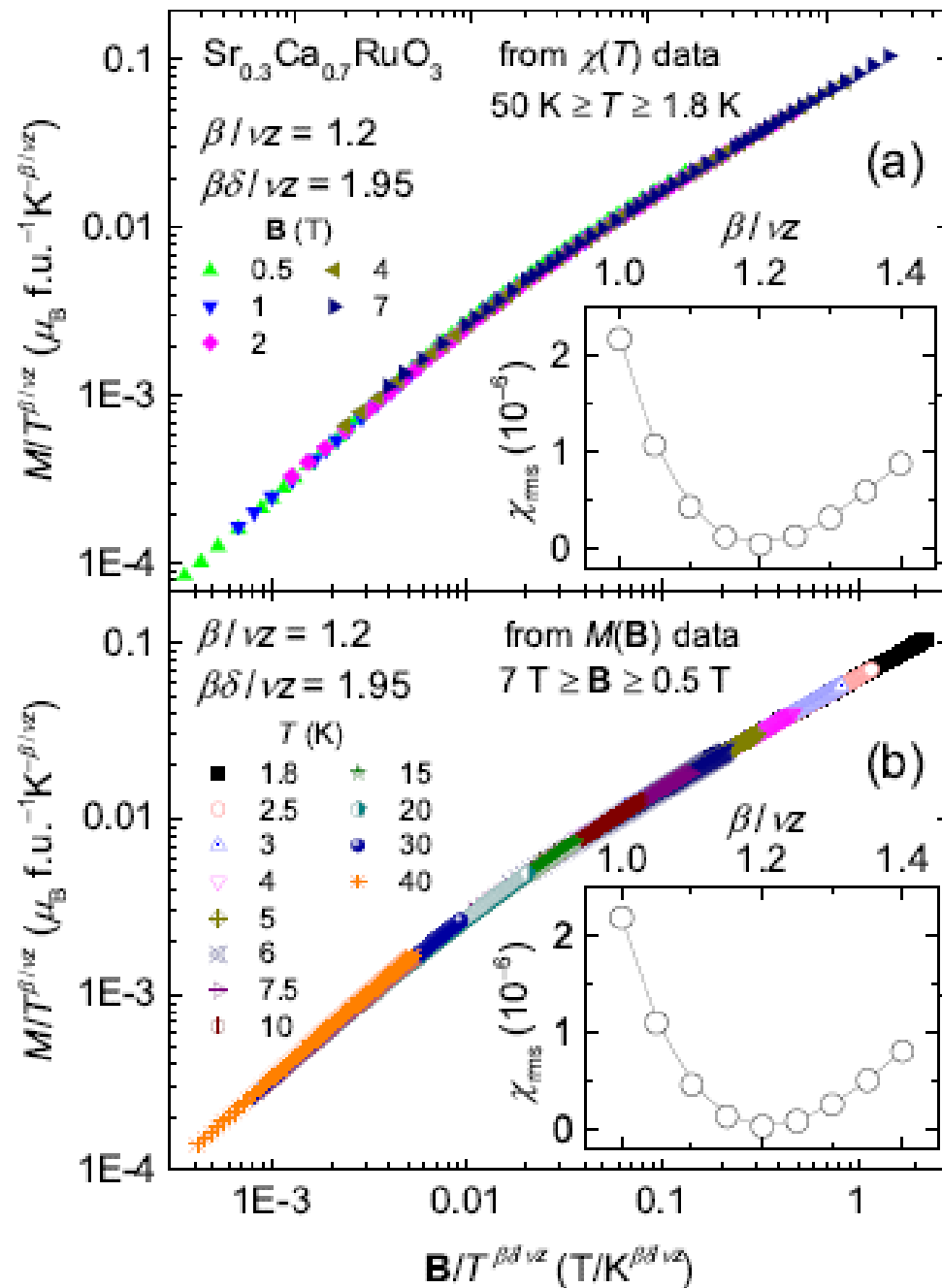
$$C_{cr}(T, B) = T^{\frac{d}{z}} \Psi\left(B/T^{\frac{\beta\delta}{\nu z}}\right).$$

# Scaling of the susceptibility and magnetization

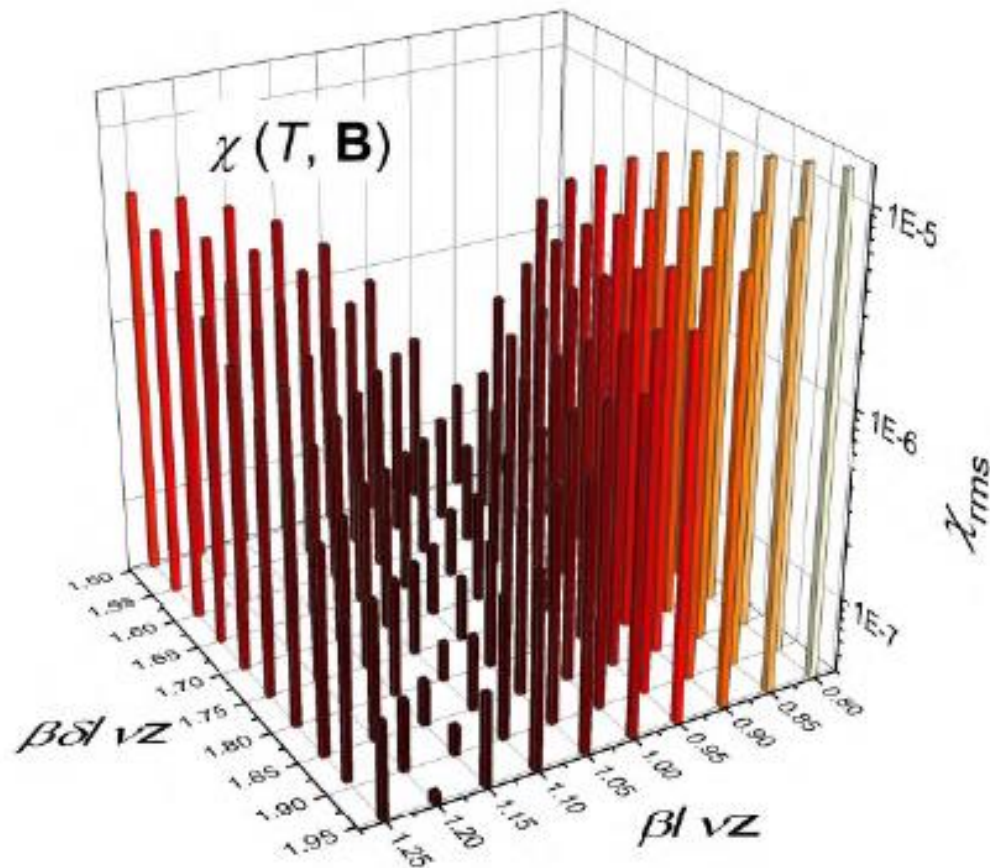
best data collapse observed with exponents

$$\frac{\beta}{vz} \approx 1, \quad \frac{\delta\beta}{vz} \approx 1.7$$

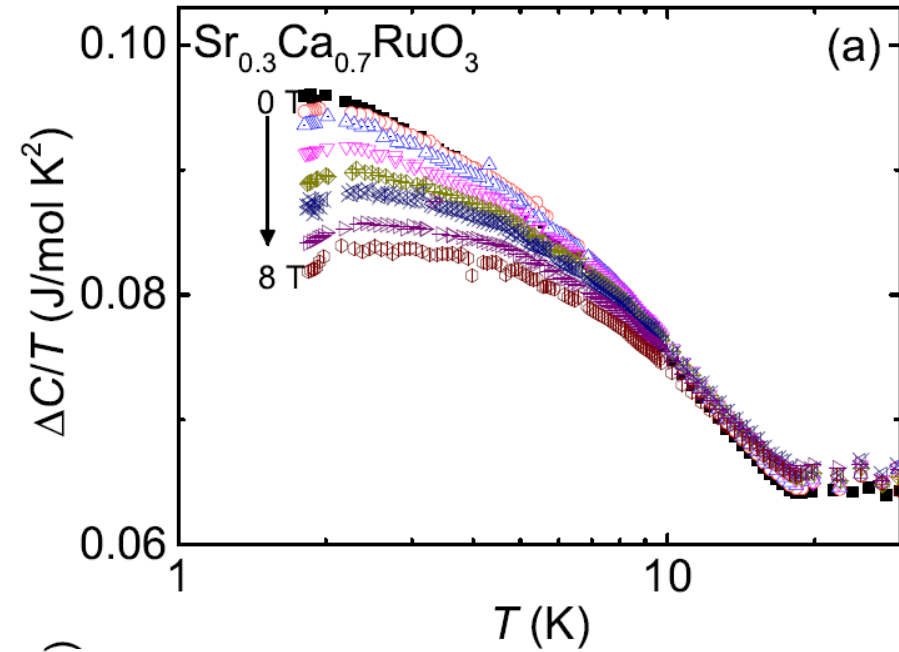
for both  $\chi(T,B)$  and  $M(T,B)$ .



# Determining the least-squares fit



# Scaling of the field dependence of the specific heat

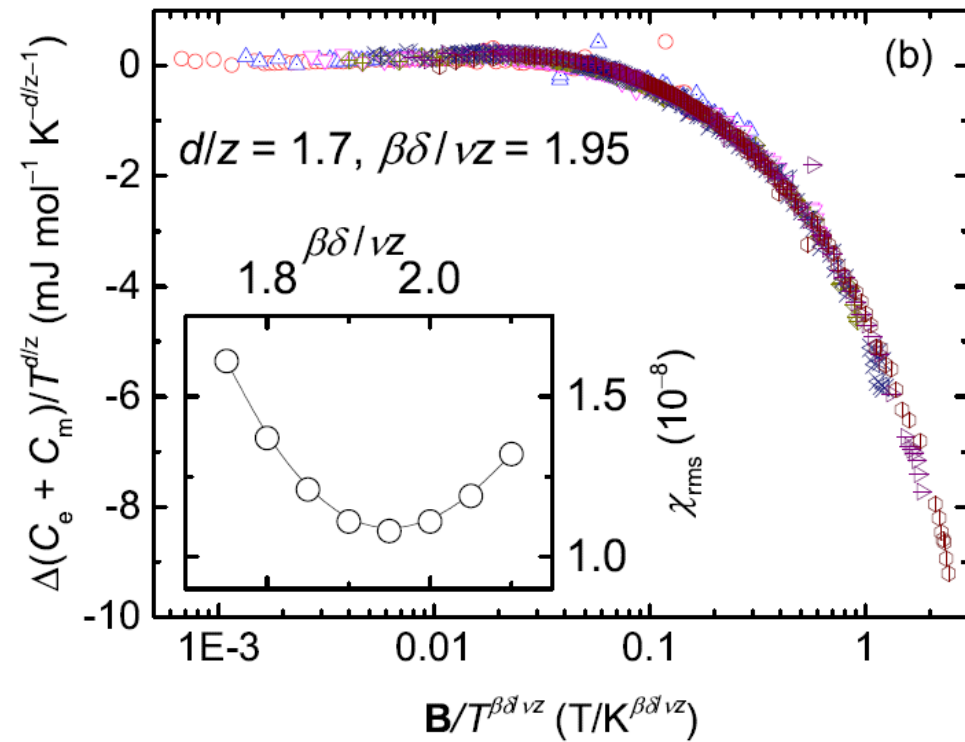


Strikingly,  $d/z = 1.7$  yields with  $d = 3$ :  
 $z \approx 1.8$ ,  
 much smaller  
 than in typical ferromagnets

$$\Delta C(B, T) - \Delta C(0, T)$$

(phonon contribution subtracted)

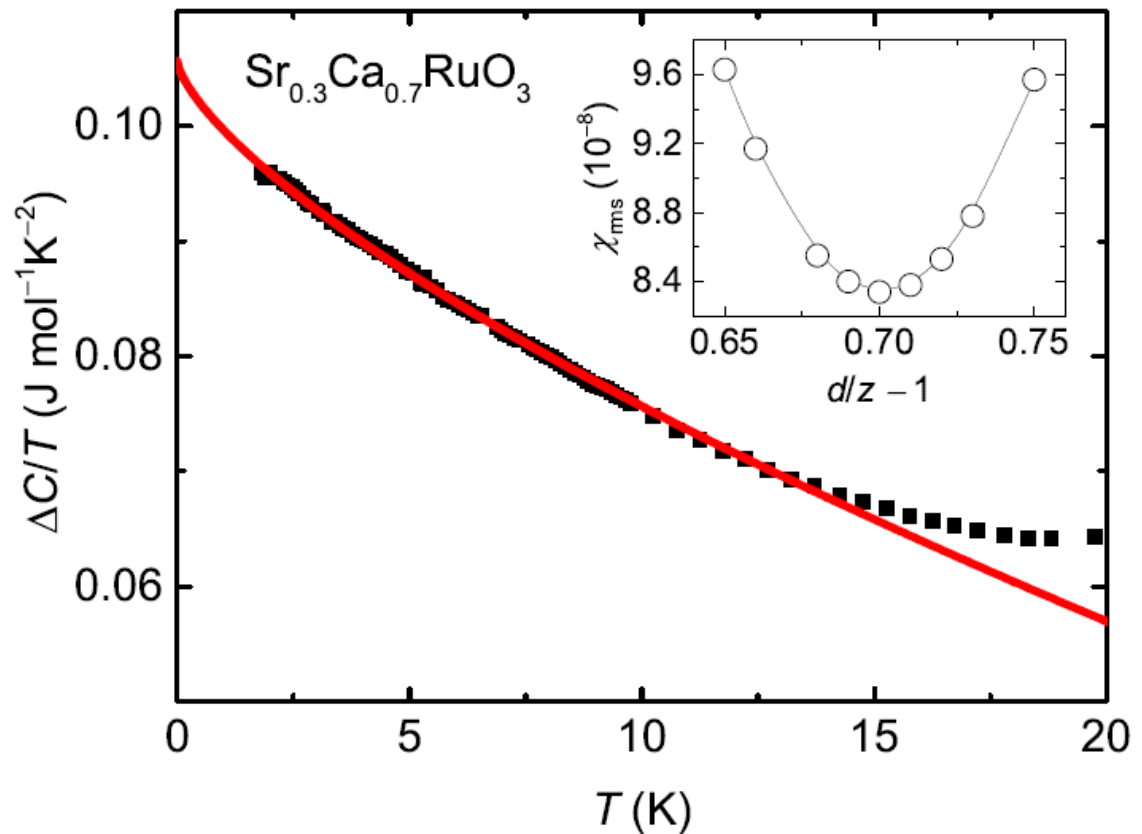
best data collapse observed with  $\frac{\delta\beta}{vz} \approx 1.7$   
 in agreement with  $\chi(T, B)$  and  $M(T, B)$



# Zero-field specific heat of $\text{Sr}_{0.3}\text{Ca}_{0.7}\text{RuO}_3$

Consequence of  $d > z \approx 1.8$ : critical contribution subleading to electronic contribution, resulting in a finite  $\Delta C/T$  at  $T = 0$

Power-law dependence  
 $\Delta C/T \sim -T^{0.7}$   
in perfect agreement  
with  $d/z - 1 = 0.7$



# Qualitative discussion on the anomalous dynamic exponent $z$ in $\text{Sr}_{0.3}\text{Ca}_{0.7}\text{RuO}_3$

Original prediction by Hertz 1976

$z = 3$  for a clean ferrromagnet

$z = 4$  for a disordered ferromagnet

Generally, disorder tends to enhance  $z$

Complication because additional dynamics of fermions might lead to a first-order transition

*Belitz and Kirkpatrick*

Alternative scenario Griffiths phase in ferromagnet

example  $\text{Ni}_{1-x}\text{V}_x$

*Ubaid-Kassis et al., PRL 104, 066402 (20910)*

Strong coupling between critical fluctuations and incoherent quasiparticles:  
local fluctuations may lead to a local gap in the Stoner continuum,  
leading to less damping of fluctuations, i. e., smaller  $z$

