Interplay of Kondo effect and geometric frustration in quantum-critical CePdAl

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KITP Workshop on
Strong Correlations and Unconventional Superconductivity:
Towards a Conceptual Framework

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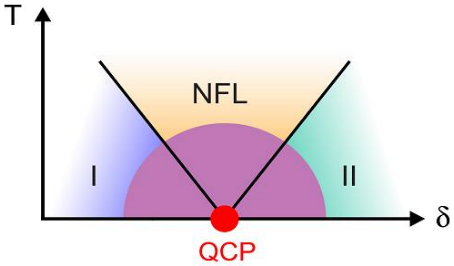

Kavli Institute for Theoretical Physics

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Outline

CePdAl: A partially frustrated heavy-fermion system

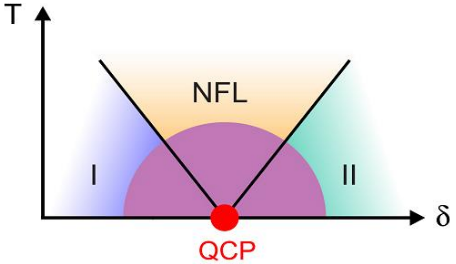
Approaching quantum criticality by Ni substitution

Possibility of a spin-liquid state in pure CePdAl ?

Magnetic (*B*,*T*) phase diagram probed by magnetostriction

IRONICS: Electronic correlations in AFe_2As_2 (*A* = K, Rb, Cs)

probed by quantum oscillations of the magnetostriction



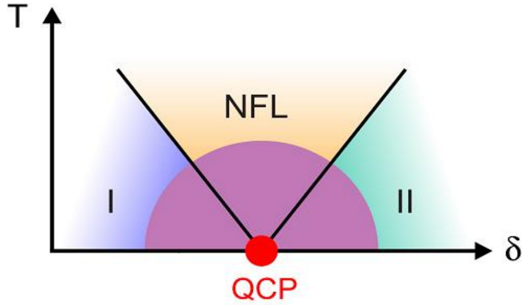
Who‘s done it and who paid for it

V. Fritsch, N. Bagrets, W. Kittler, C. Taubenheim, K. Grube — *CePdAl*

O. Stockert, S. Woitschach, Z. Hüskes — *MPI-CPfS Dresden*
neutron scattering: ILL, Munich

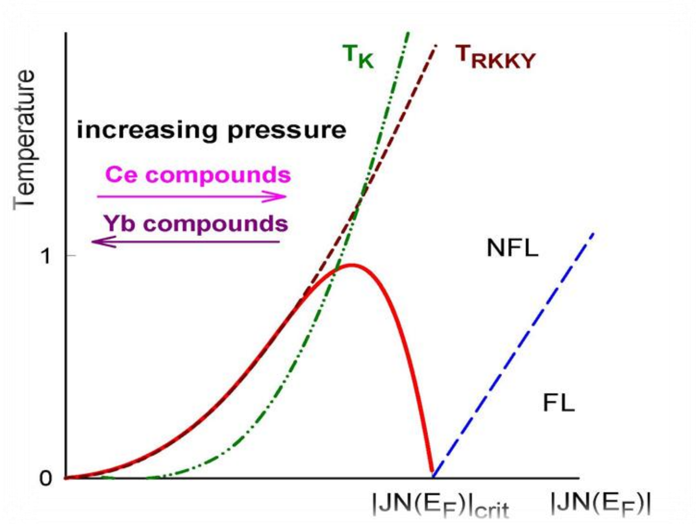
F. Eilers, D. Zocco, K. Grube, Th. Wolf — *AFe_2As_2*

Work supported by Deutsche Forschungsgemeinschaft
and Helmholtz Association of Research Centers



Competing interactions with the possibility of quantum phase transitions

Heavy-fermion metals: Kondo vs. RKKY interaction

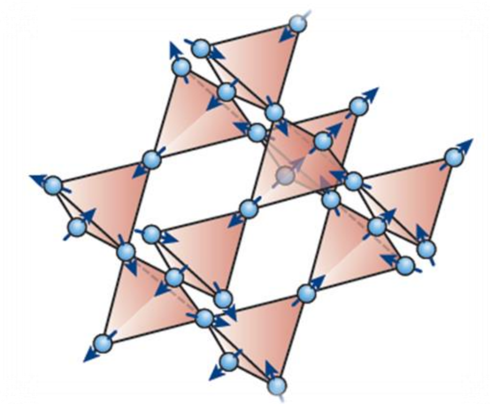


onsite – intersite competition

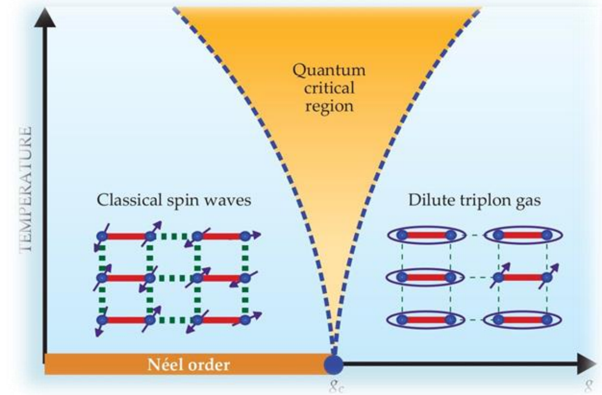
“Doniach phase diagram“

Competing nn or nn/nnn interactions in insulating magnets

Geometric frustration of nn interactions



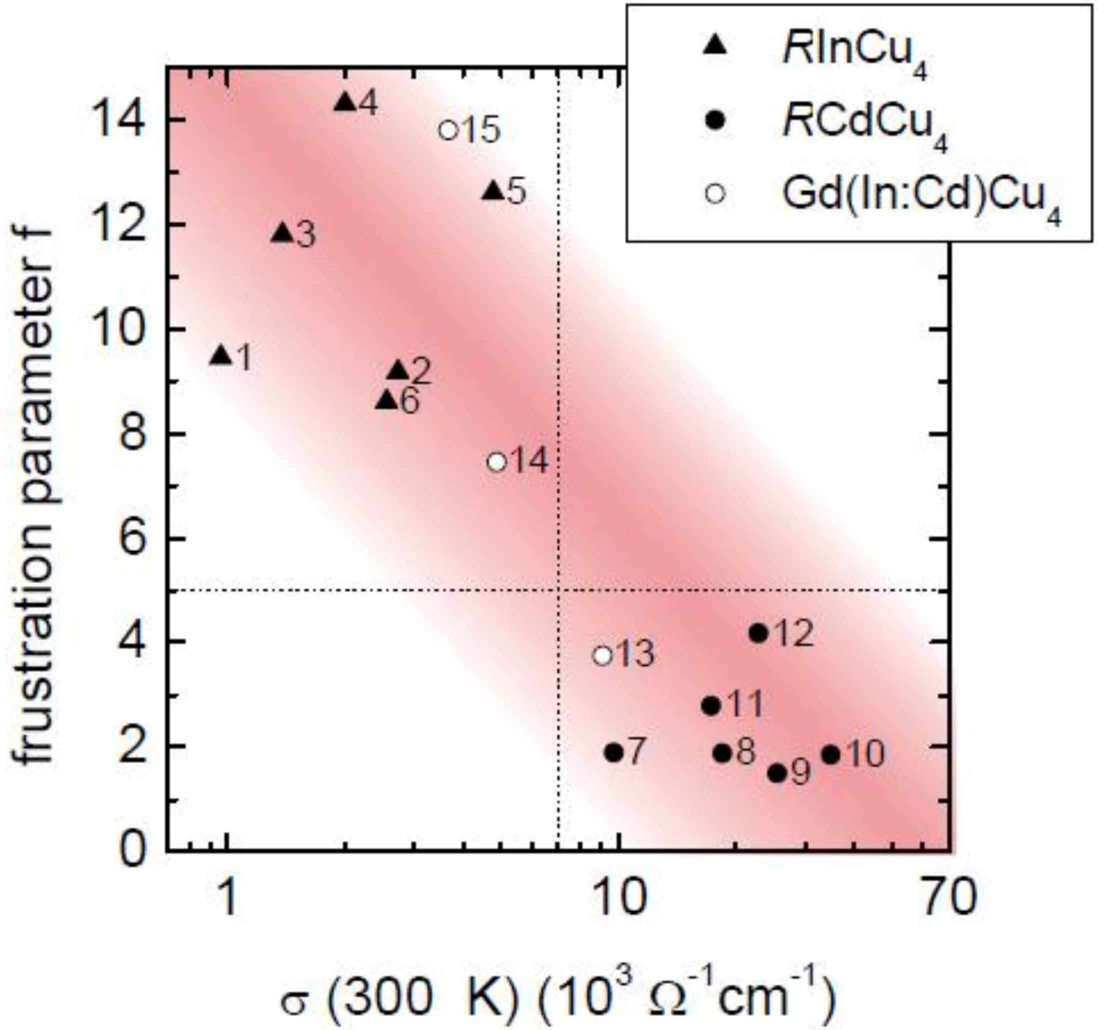
Competing nn and nnn interactions



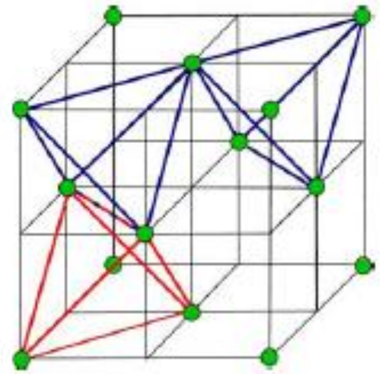
Sachdev and Keimer, Phys.Today 2011

Approaching a quantum critical point in partially frustrated $CePd_{1-x}Ni_xAl$

Frustration and conductivity



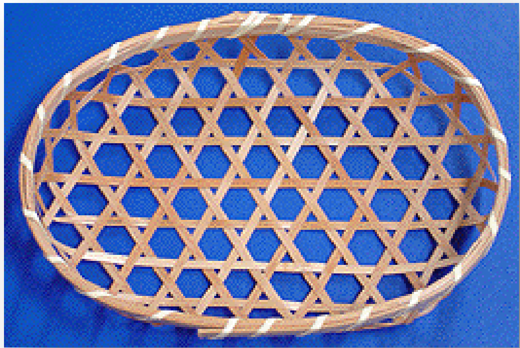
1: $GdInCu_4$
2: $TbInCu_4$
3: $DyInCu_4$
4: $HoInCu_4$
5: $ErInCu_4$
6: $TmInCu_4$
7: $GdCdCu_4$
8: $TbCdCu_4$
9: $DyCdCu_4$
10: $HoCdCu_4$
11: $ErCdCu_4$
12: $TmCdCu_4$
13: $GdIn_{0.25}Cd_{0.75}Cu_4$
14: $GdIn_{0.5}Cd_{0.5}Cu_4$
15: $GdIn_{0.75}Cd_{0.25}Cu_4$

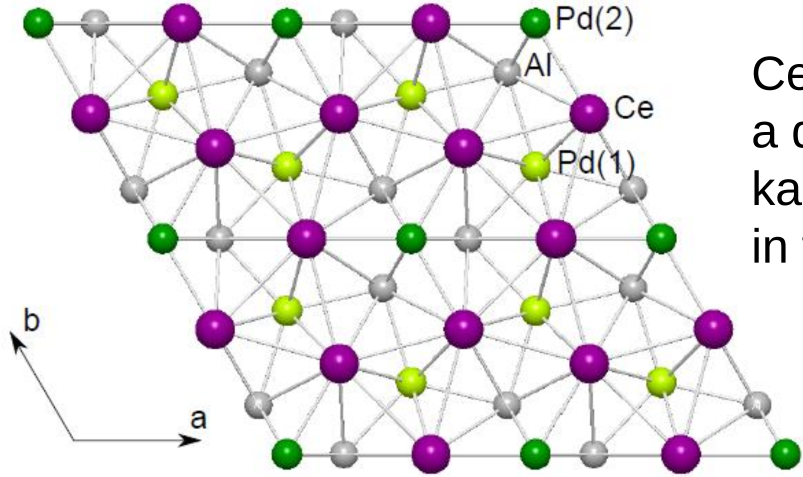


Frustration parameter $f = \Theta_{CW} / T_c$

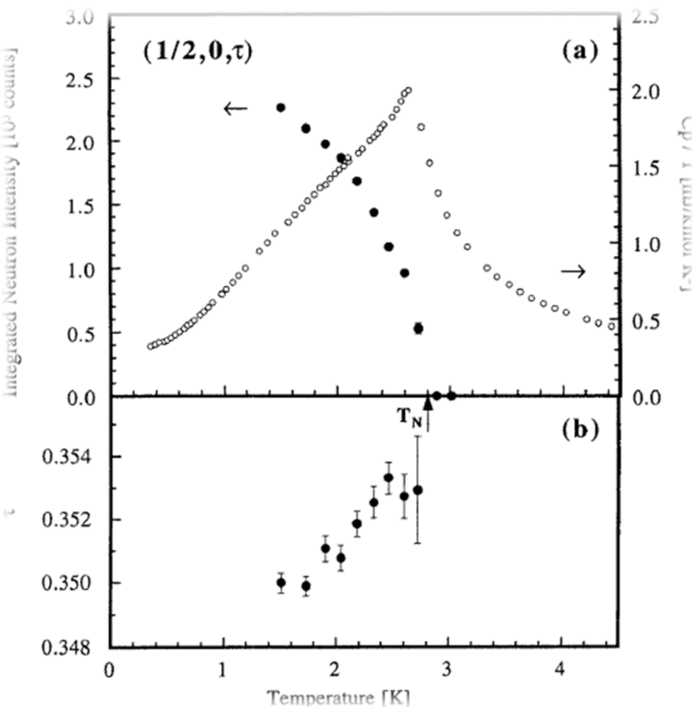
*V. Fritsch et al., PRB **72** (2006)*

CePdAl – a partially frustrated Ce-based compound





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



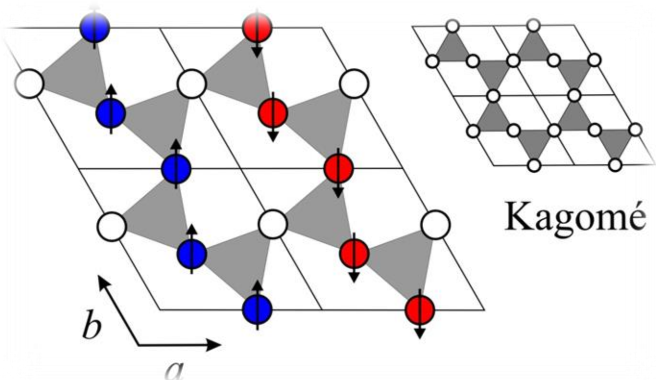
Kondo system, $T_K \approx 5$ K

Magnetic order below T_N = 2.7 K

Q = (½ 0 τ), τ ≈ 0.35

1/3 of Ce moments frustrated

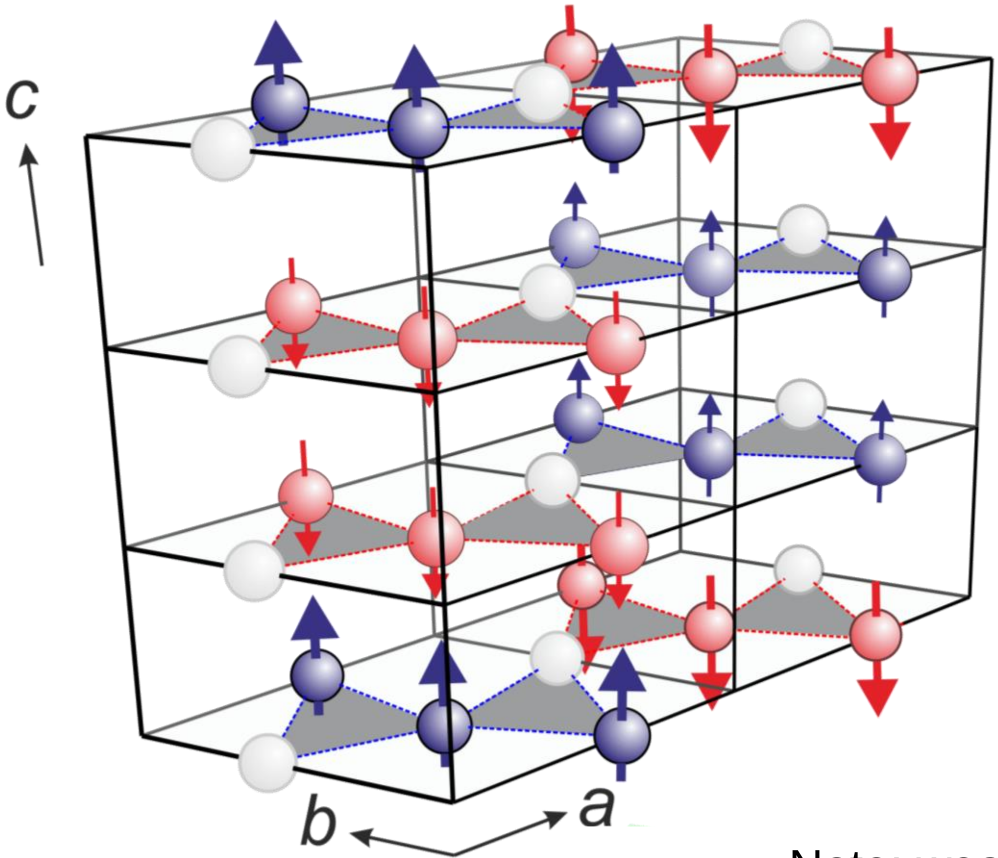
Magnetic structure



*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 wector

Q = (½ 0 τ), $\tau \approx 0.35$

1/3 of Ce moments frustrated

μ(Ce1) = 1.58 μ_B

Note: weak T-dependent incommensuration neglected in the picture

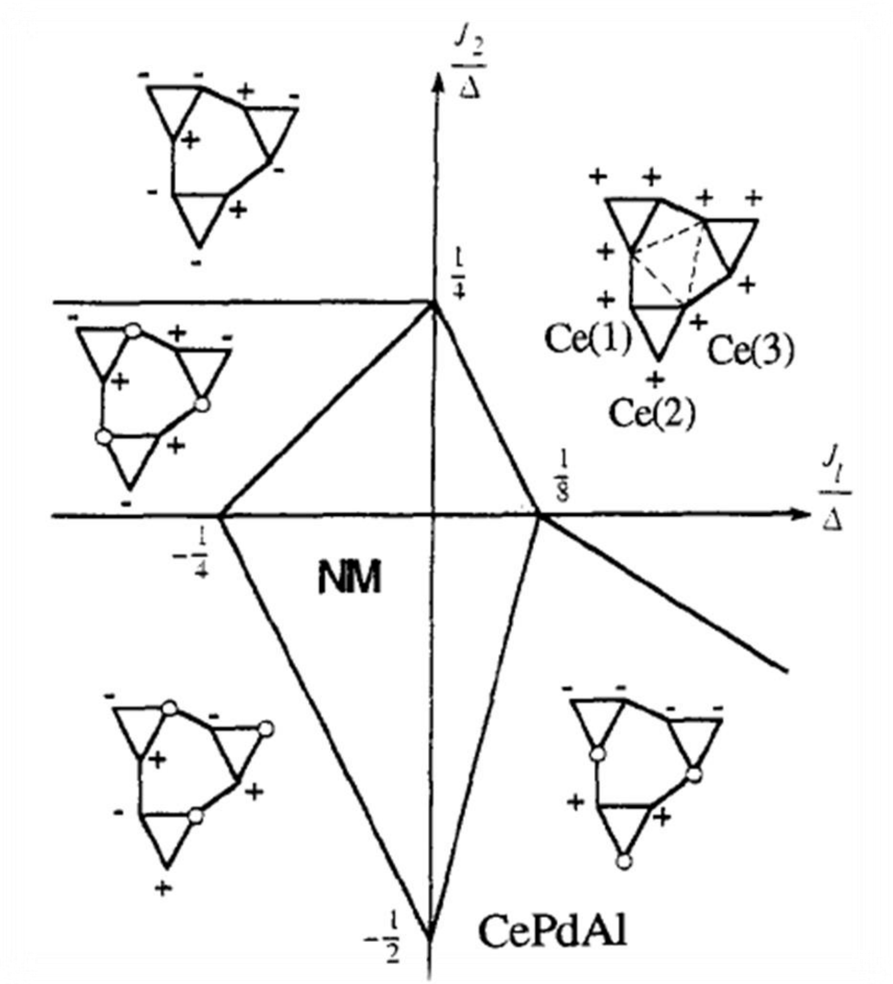
Model of frustrated kagomé-like planes

*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

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

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



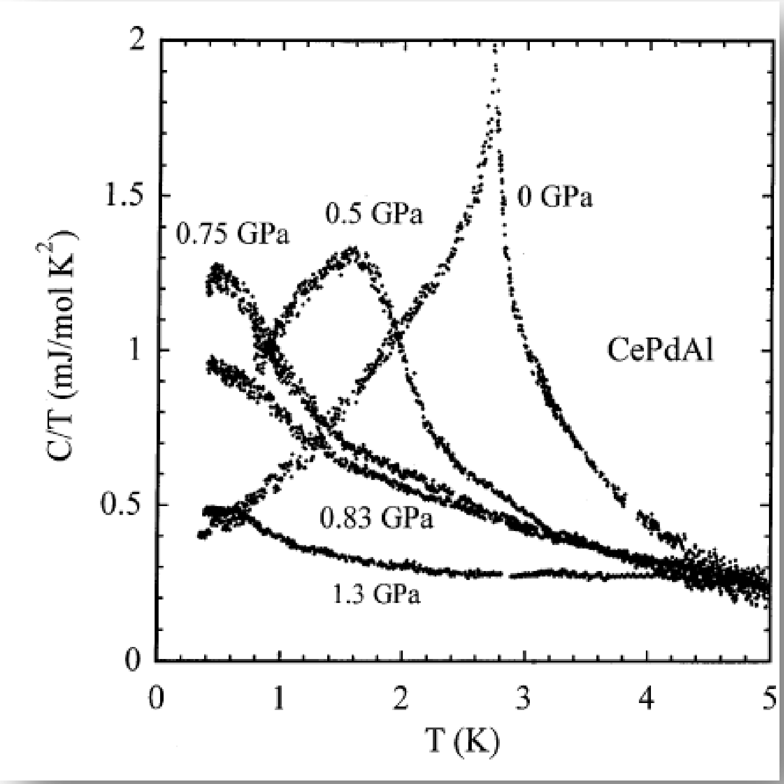
Mean-field phase diagram

confirmed by variational MC

*Motome et al., PRL **105**, 036403*

coupling between planes neglected!

CePdAl – a partially frustrated Ce-based compound



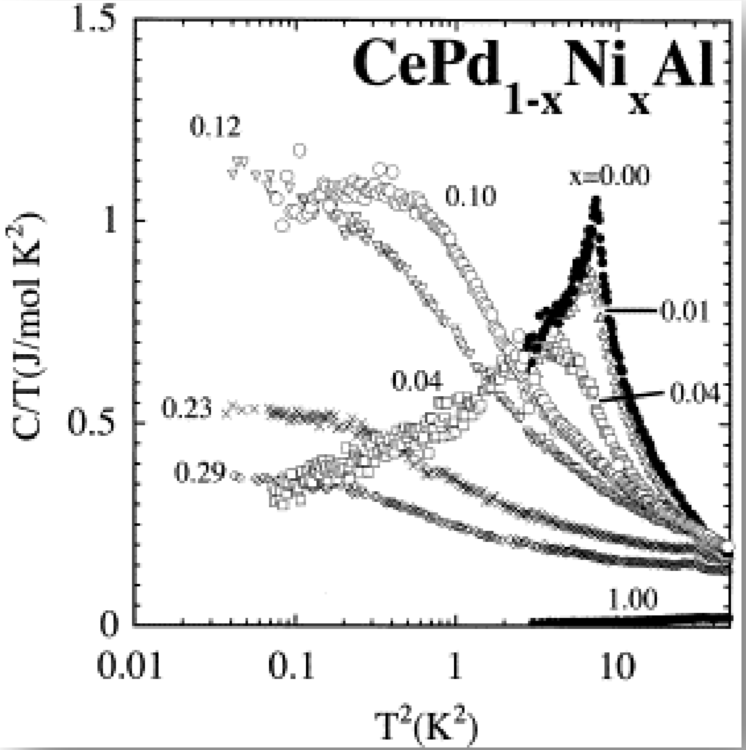
Suppression of T_N by hydrostatic pressure …

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

…

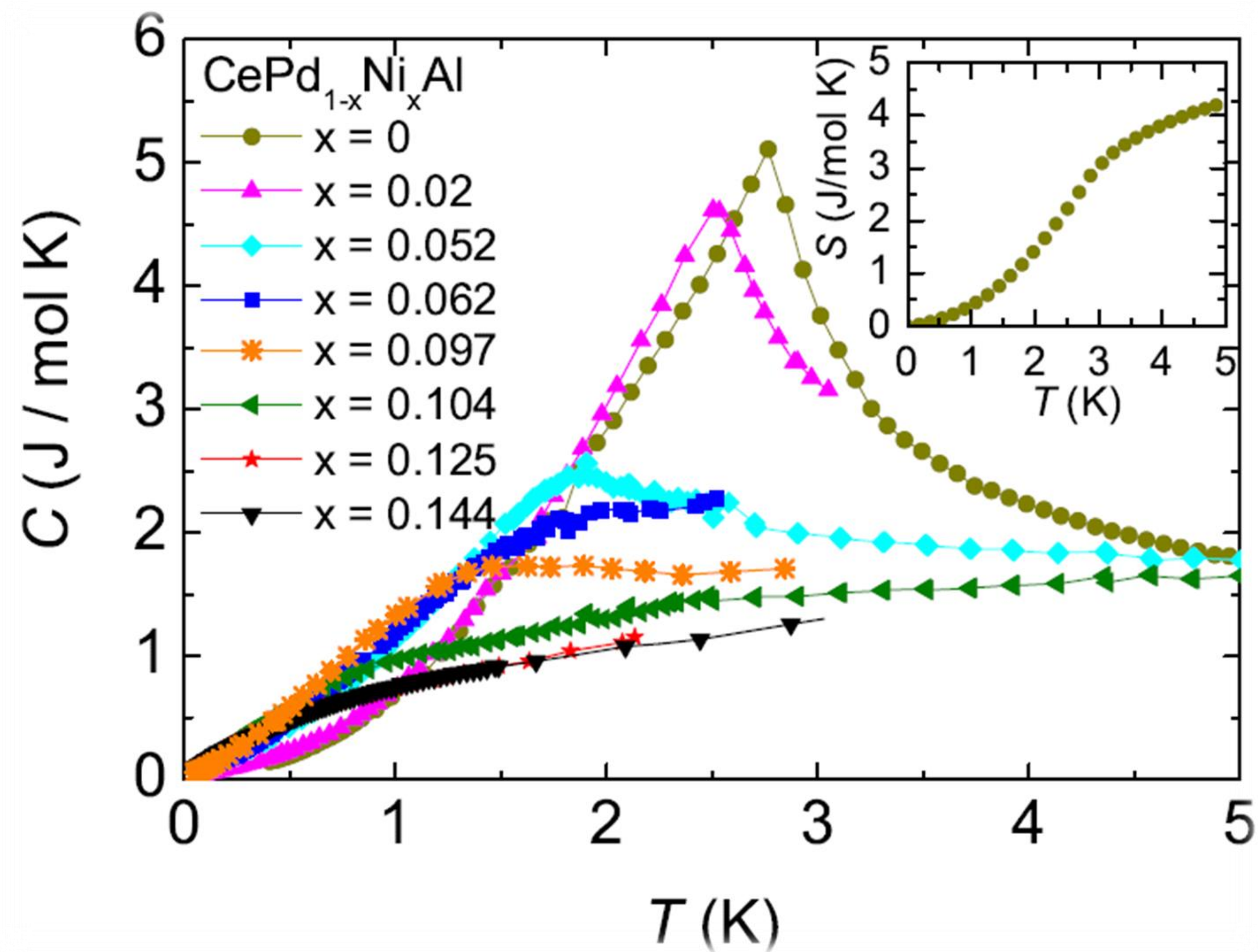
… or by isoelectronic Ni doping

Isikawa et al., Physica B ***281&282****, 36 (2000)*
Fritsch et al., PRB ***89****, 054416 (2014)*



Quantum critical point?

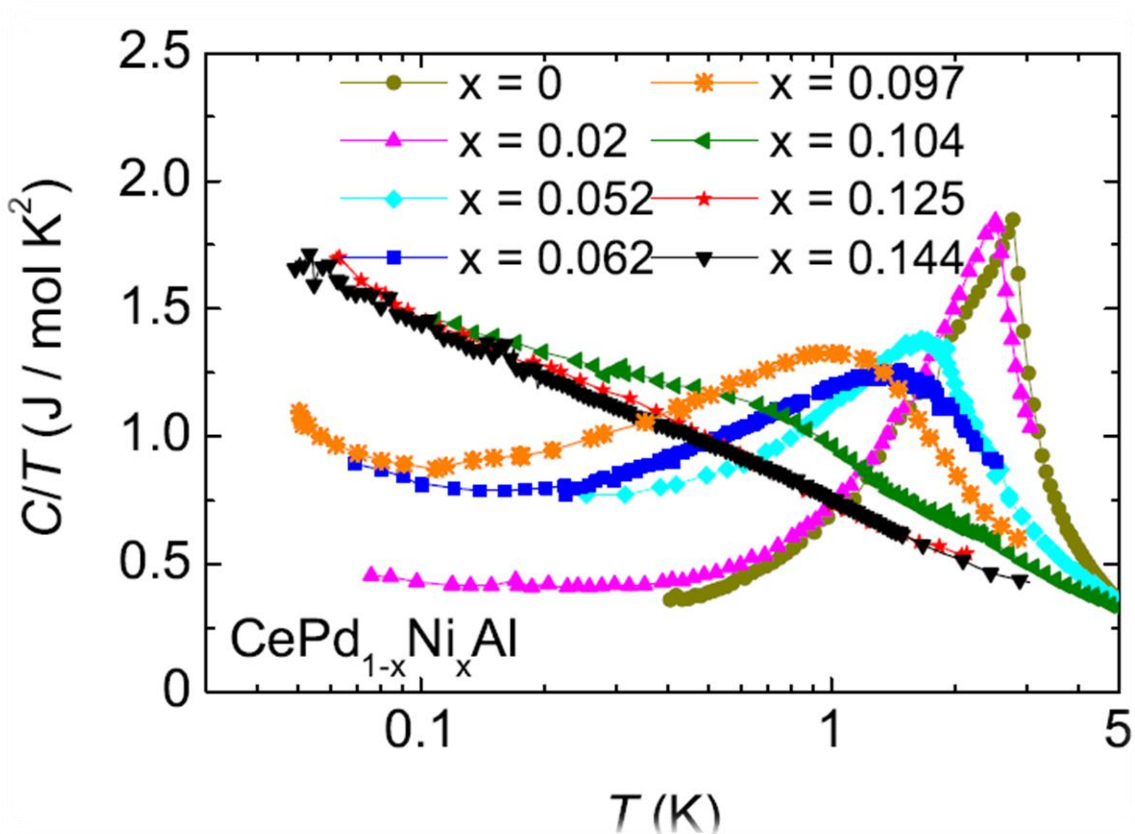
Specific heat of $CePd_{1-x}Ni_xAl$ polycrystals



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

Approaching quantum criticality of CePdAl by Ni substitution

Specific heat



V. Fritsch et al., PRB **89**, 054416 (2014)

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

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

2D quantum criticality or novel QCP?

cf. $\rho(T)$ of CePdAl

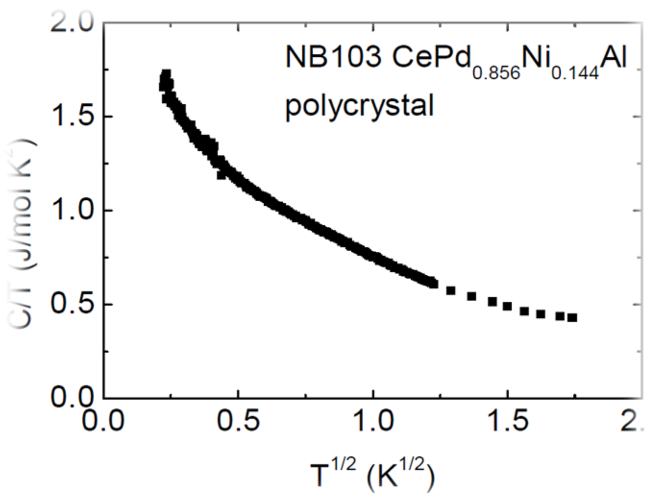
at p = 1 – 1.2 GPa:

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

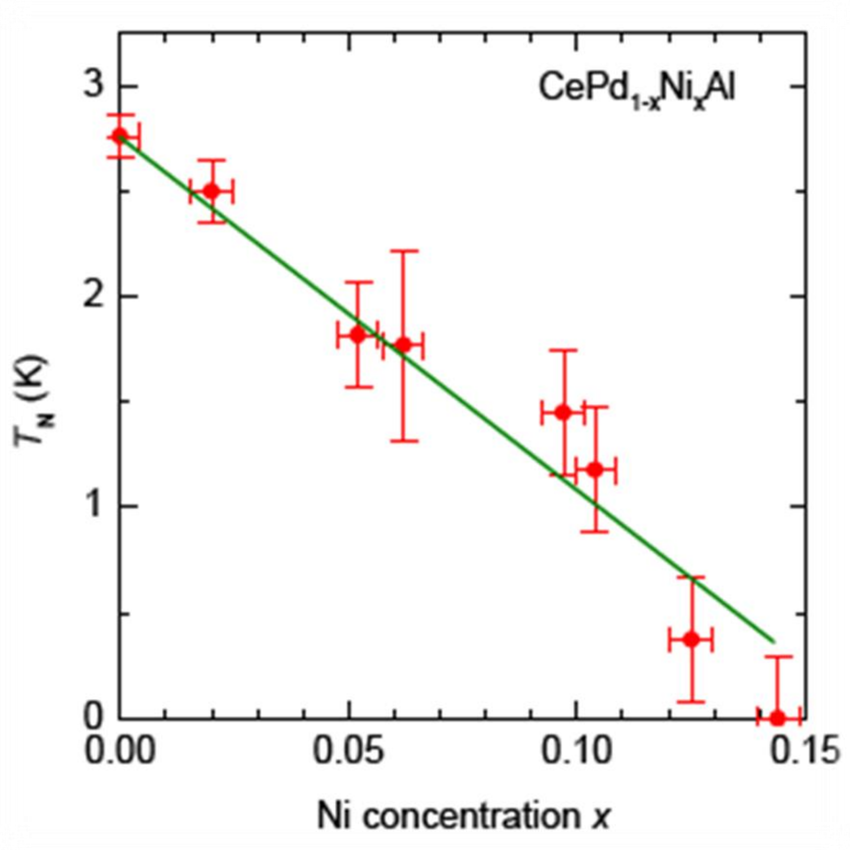
Goto et al., J. Phys. Chem. Sol. ***63****, 1159 (2002)*

Data do not follow

$C/T \sim 1 - \sqrt{T}$ (3D QCP)

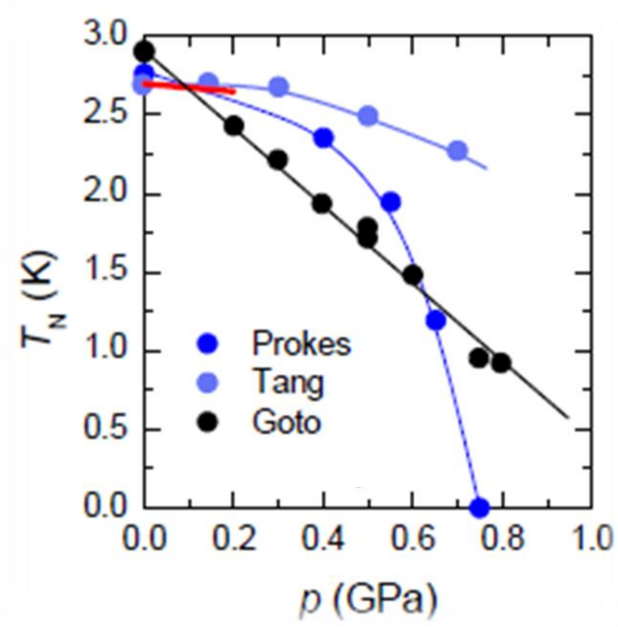


T_N (x) of $CePd_{1-x}Ni_xAl$ polycrystals



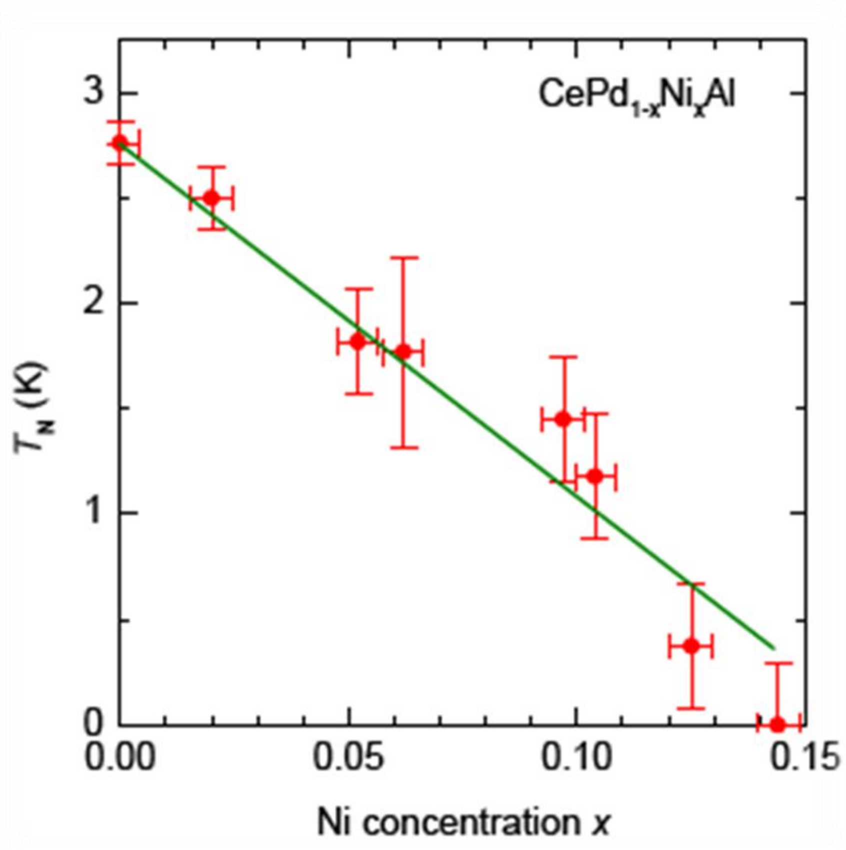
Comparison of pressure and Ni substitution: $T_N(V(x))$ and $T_N(V(p))$?
Experimental $T_N(p)$ data differ strongly!

*V. Fritsch et al., PRB **89**, 054416 (2014)*



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

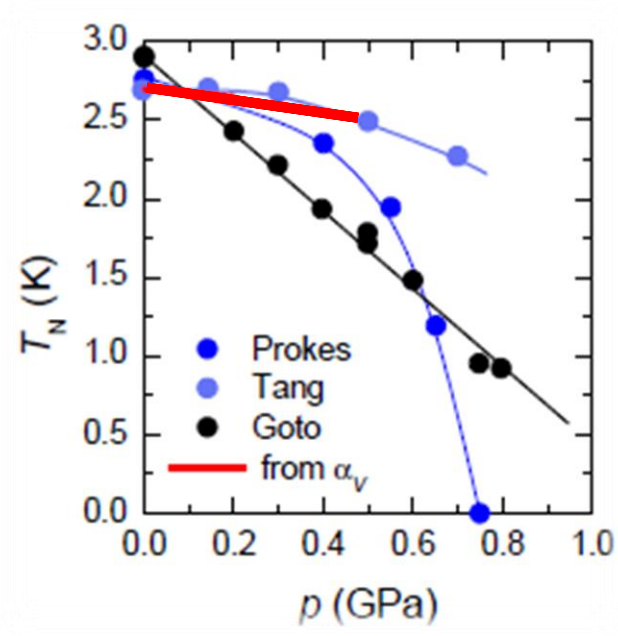
T_N (x) of $CePd_{1-x}Ni_xAl$ polycrystals



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 $dT_N/dp_c < 0$.

V. Fritsch et al., PRB **89**, 054416 (2014)



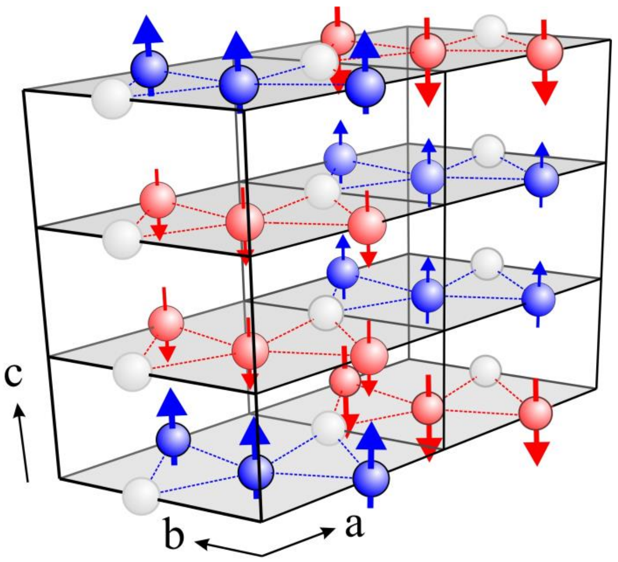
Best fit with linear T_N dependence on x, compatible with 2D HMM scenario, deviation for $x \rightarrow x_c$ (“order by disorder“?)

AF order and 2D quantum criticality in $CePd_{1-x}Ni_xAl$?

Interpretation within the Hertz-Millis-Moriya model (1):

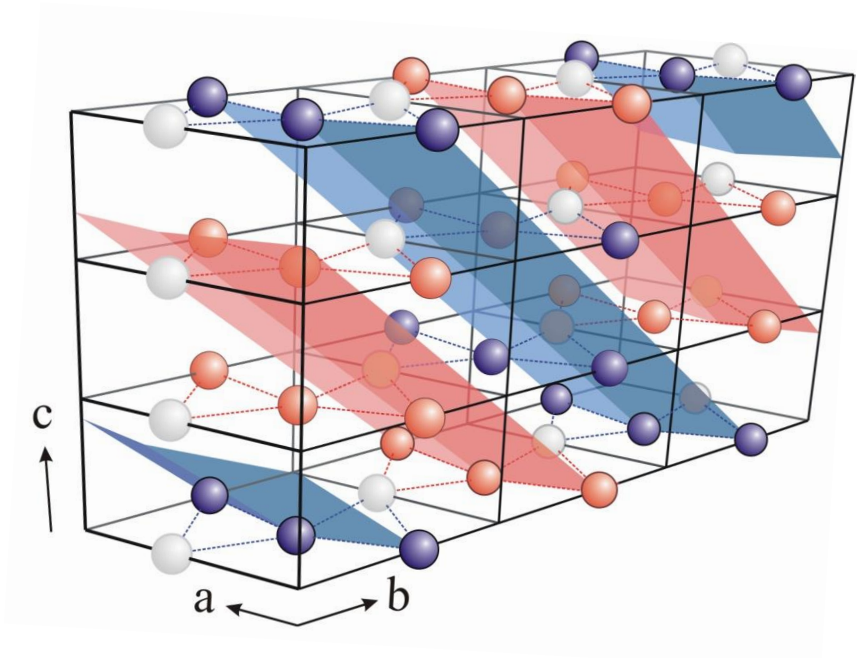
Candidates for planes with 2D?

kagomé *ab* planes



FM chains separated
by frustrated moments

planes $\mathbf{Q} = (\frac{1}{2}\ \ 0\ \ \tau)$, $\tau \approx \frac{1}{3}$



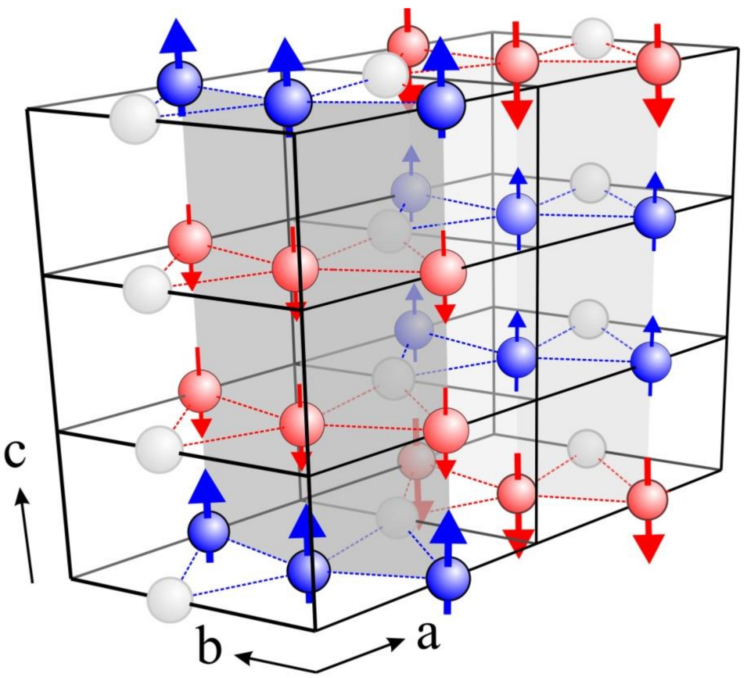
FM planes separated
by frustrated moments

AF order and 2D quantum criticality in $CePd_{1-x}Ni_xAl$?

Interpretation within the Hertz-Millis-Moriya model (2):

Candidates for planes with 2D?

planes ⊥ *ab*



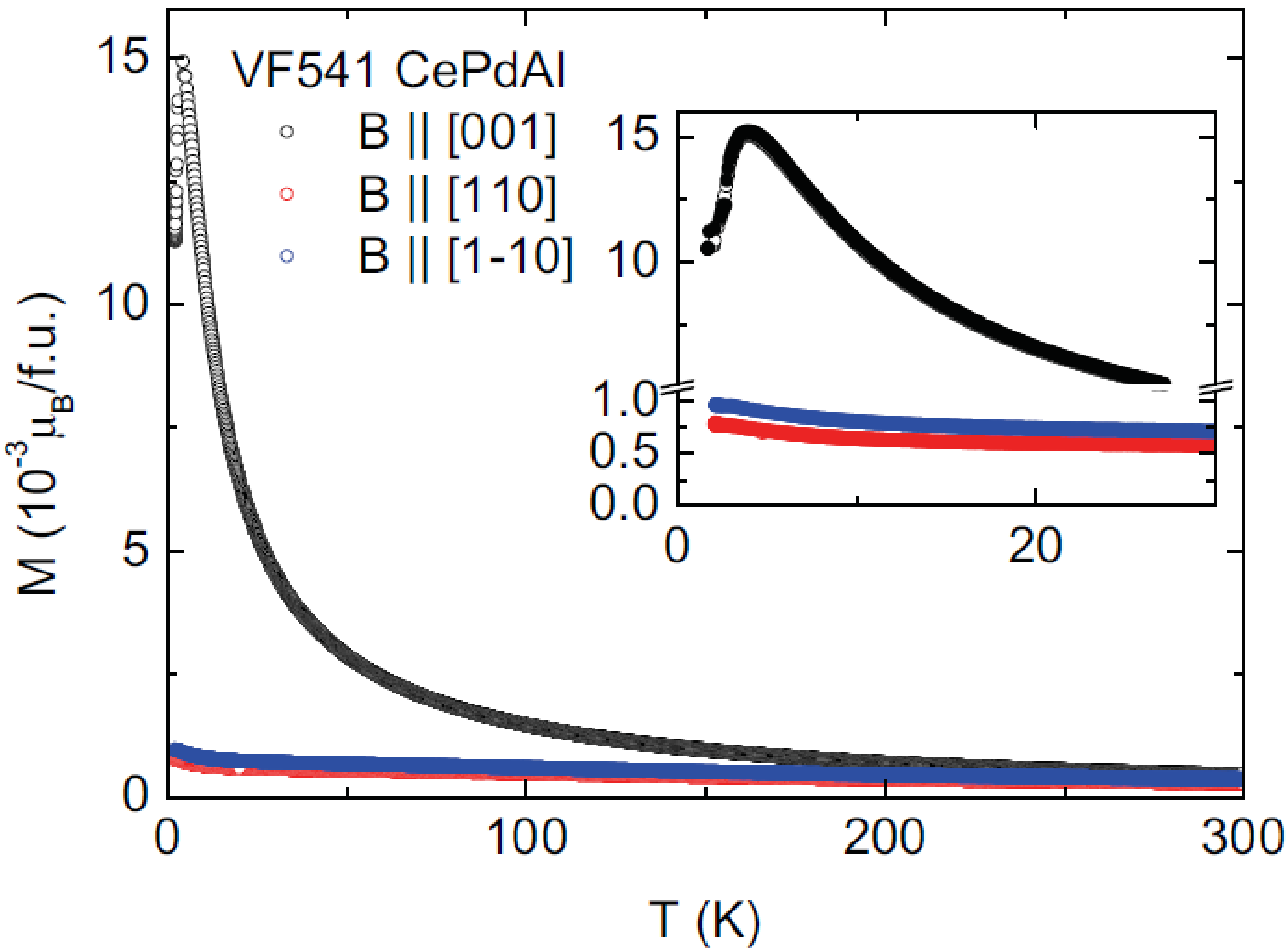
AF planes separated
by frustrated moments

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

Magnetic susceptibility of a CePdAl single crystal



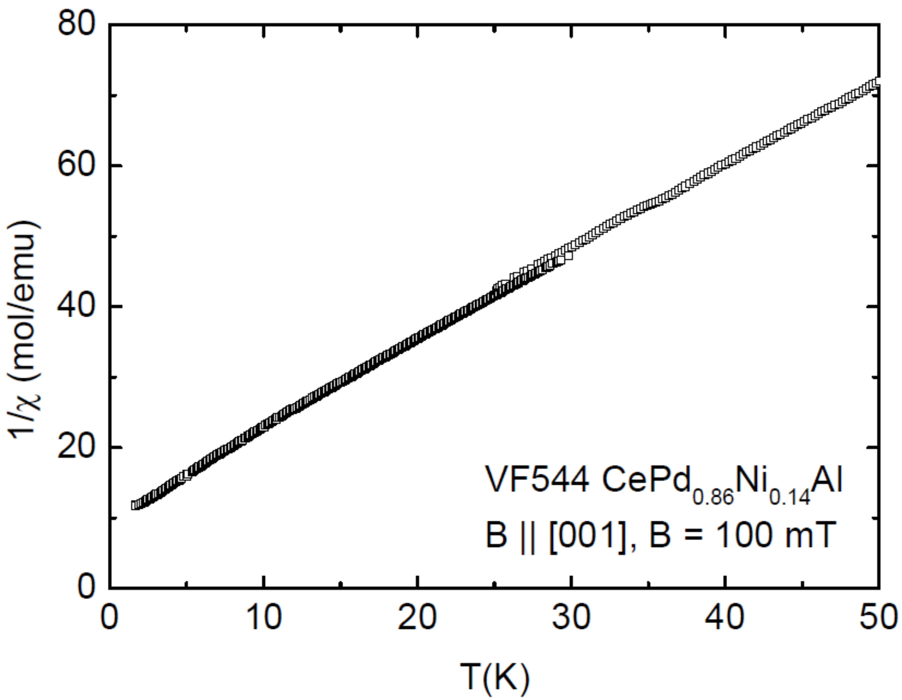
Strong Ising-like anisotropy due to single-ion crystal-field effects

Isikawa et al., J. Phys. Soc. Jpn. ***65,*** *Suppl. B, 117 (1996)*

First experiments on $CePd_{1-x}Ni_xAl$ single crystals

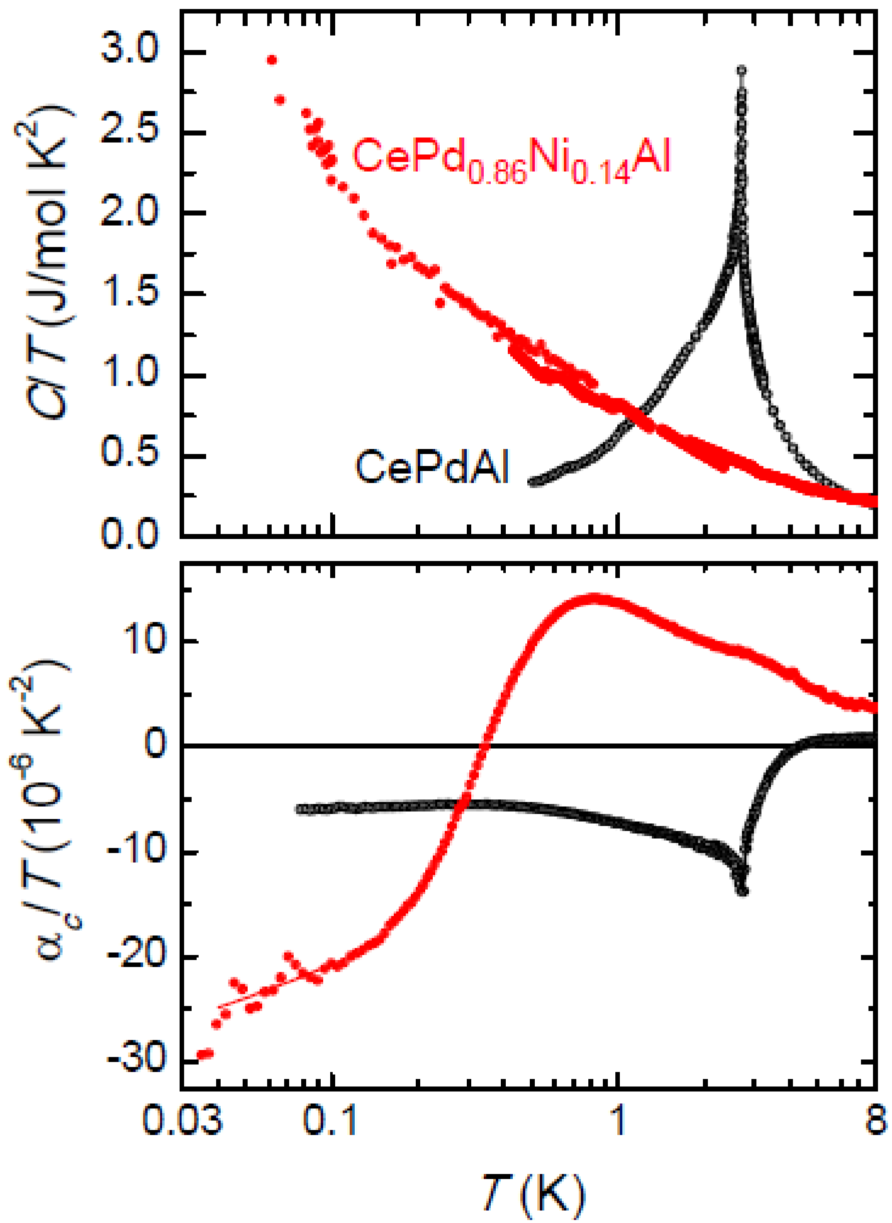
x = 0.14, close to QCP

Magnetic susceptibility



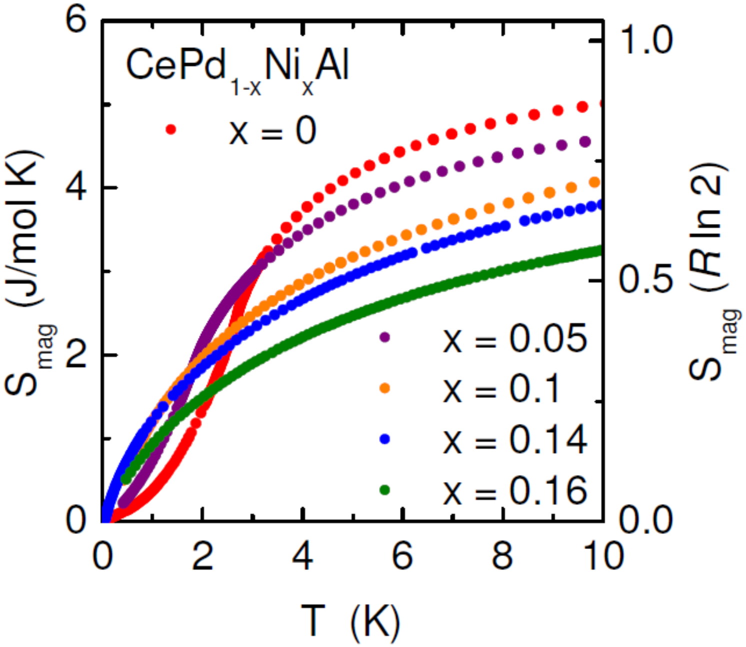
finite Θ_{CW} of $1/\chi$ vs. T

Specific heat and thermal expansion

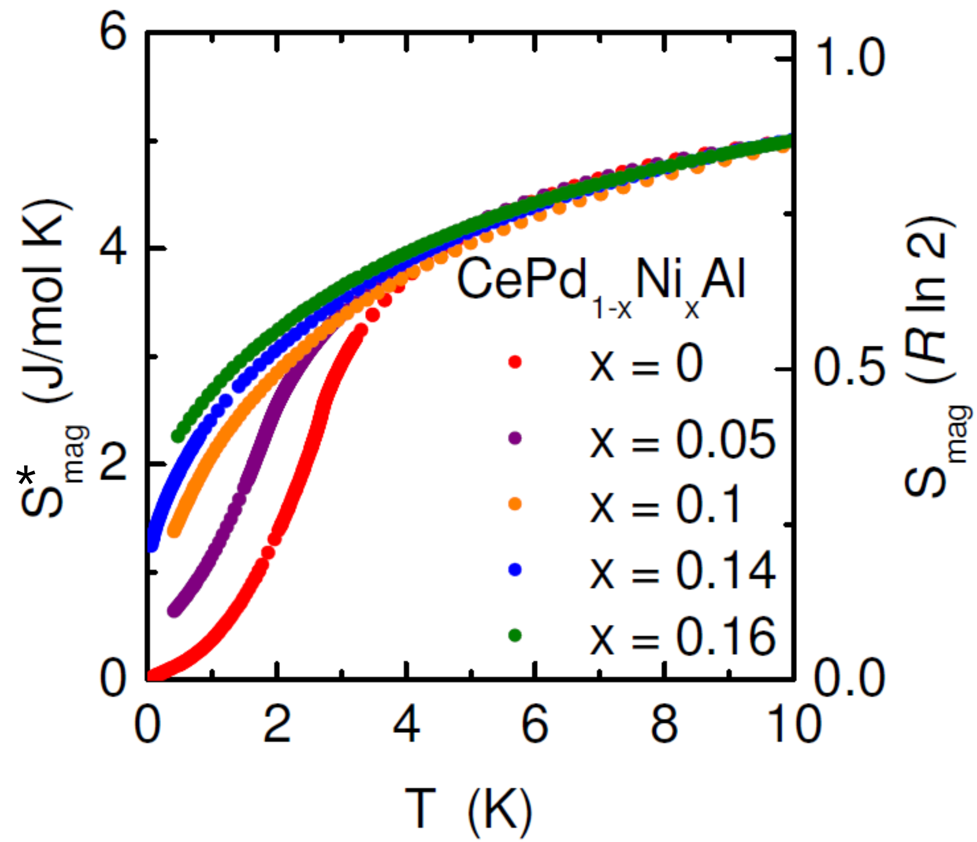


Magnetic ntropy of $CePd_{1-x}Ni_xAl$

$S_{mag}(T)$ as measured



$S^*_{mag}(T)$: data shifted vertically to coincide at T = 10 K



Systematic shift of $S^*_{mag}(T)$ to lower T upon approaching the quantum critical point

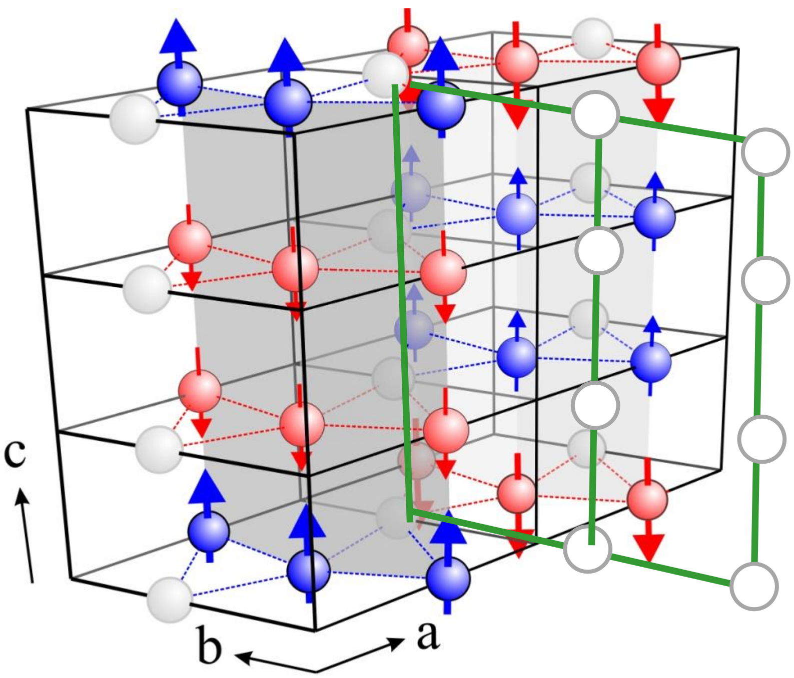
Frustrated Ce moments in CePdAl: spin liquid?

Spin liquid in CePdAl?

Metallic spin liquids are a rare species, one example:
Geometrically frustrated Kondo lattice $Pr_2Ir_2O_7$

*Nakatsuji et al., PRL **96**, 087204 (2006)*

CePdAl: planes ⊥ *ab*



Frustrated planes between AF planes form a rectangular 2D lattice:

2D Ising spin liquid?

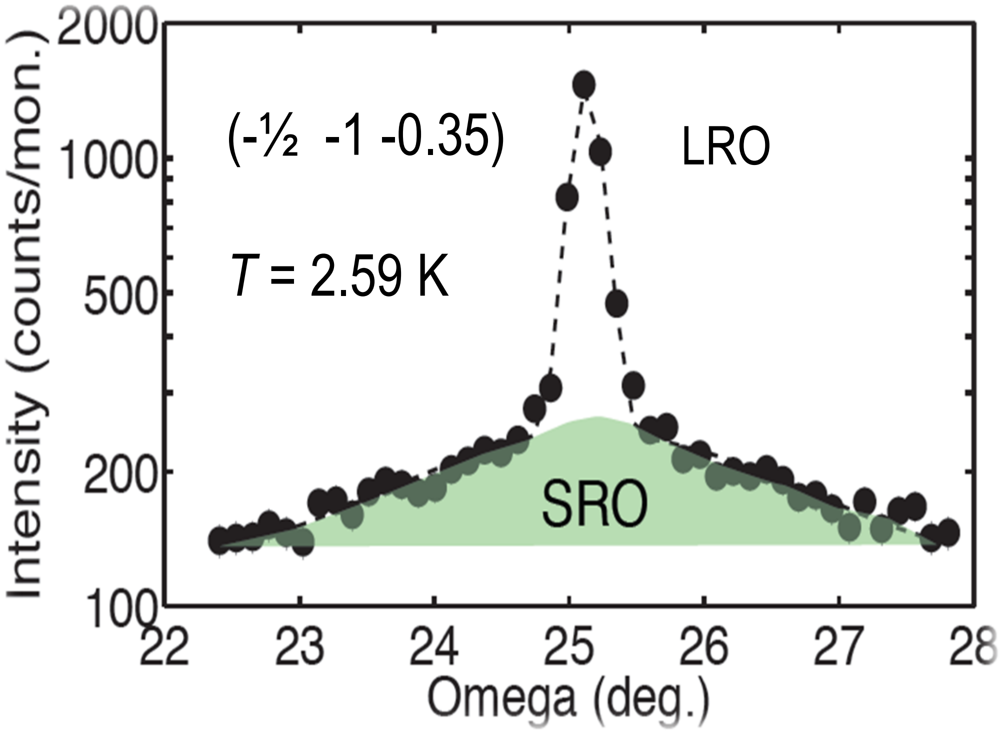
^{27}Al NMR measurements down to 30 mK
Dynamics of frustrated moments prevails down to very low T, with $T_1^{-1} \sim T$

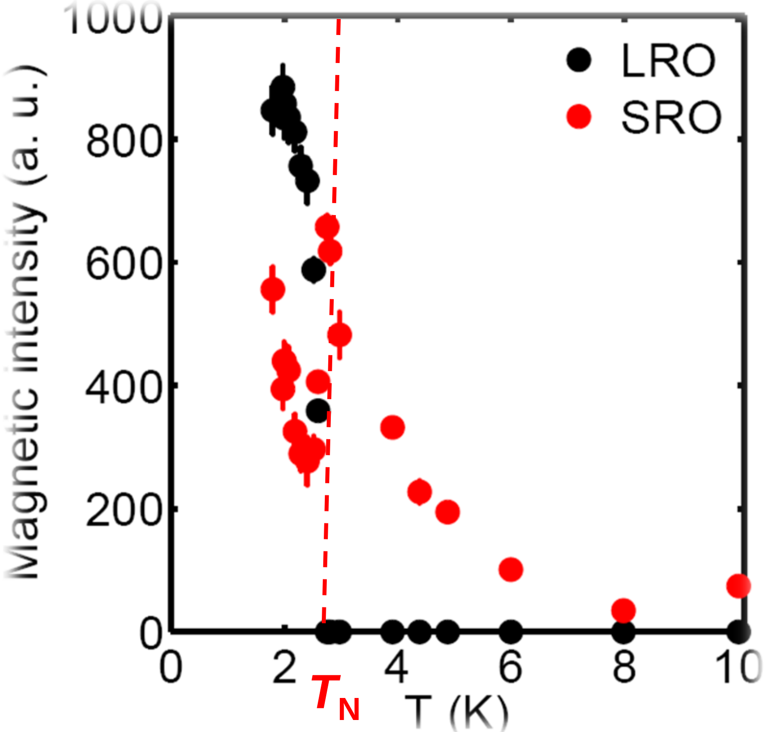
Oyamada et al.,
*Phys. Rev. B **77**, 064432 (2008).*

Long-range and short-range magnetic order in CePdAl

D20 ILL

O. Stockert et al., unpublished





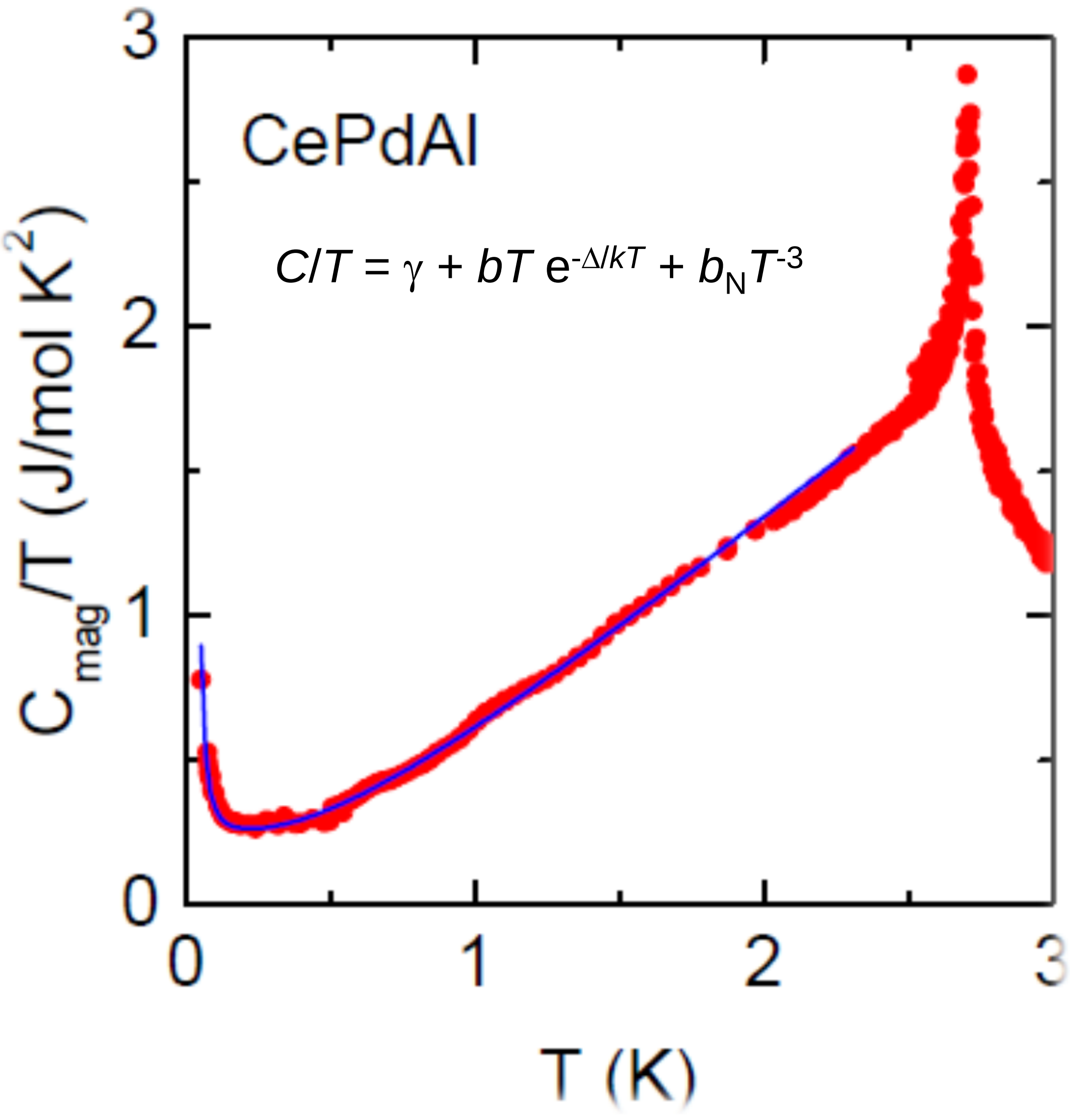
LRO/SRO intensity ratio of 2/1 below T_N:

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

$\rightarrow$ rationale for quasi-2D fluctations

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

Specific heat of CePdAl at low temperature



Several unusual features:

large γT term corresponding to

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

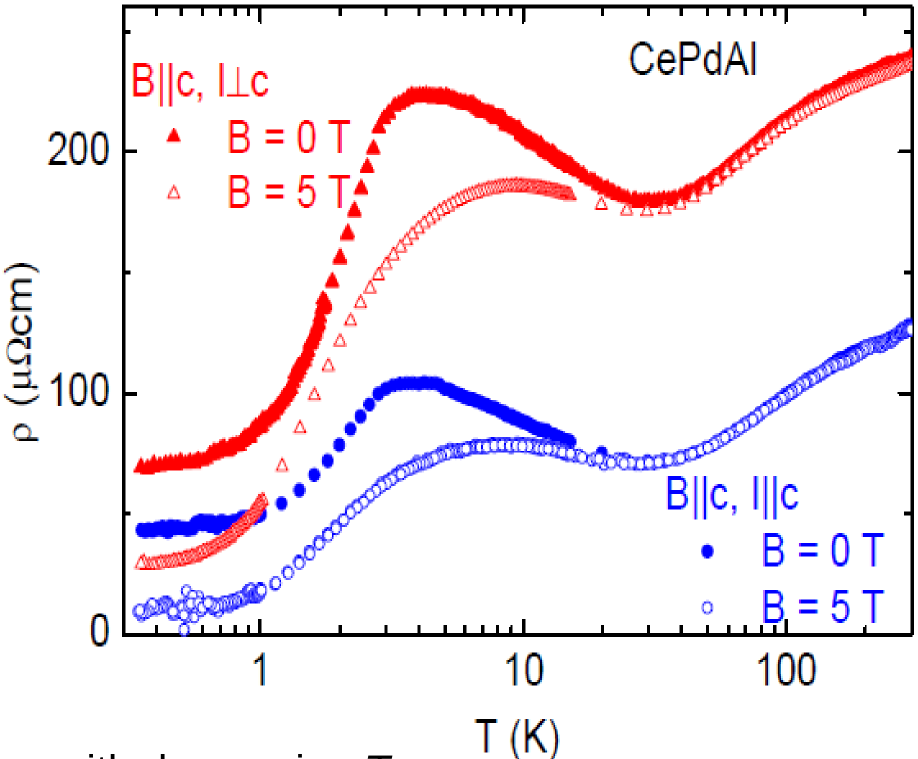
Term ~ T^2 setting in at 0.5 K

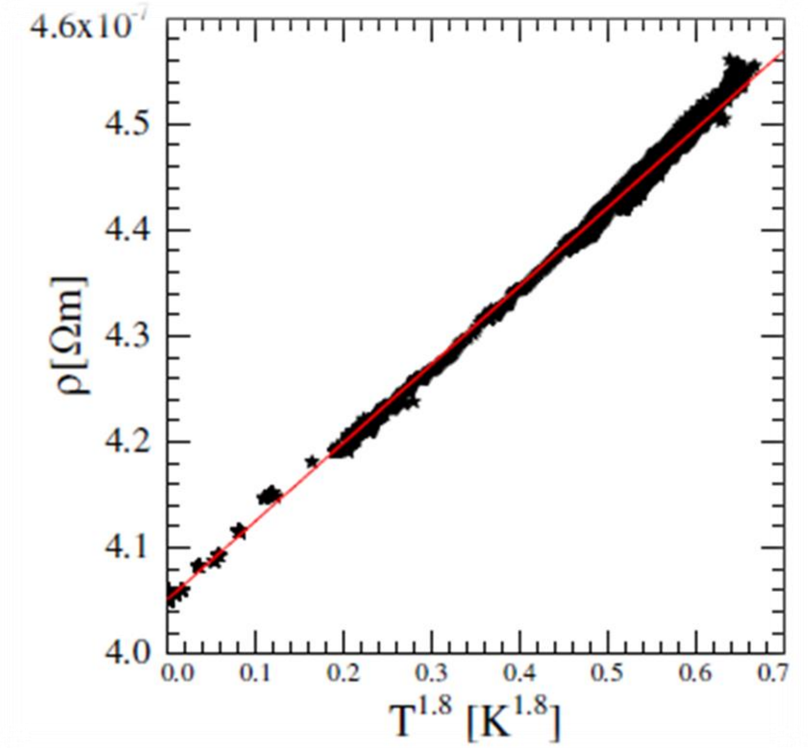
indication of a gap of corresponding excitations:

2D spin waves in an Ising system

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

Electrical resistivity of CePdAl single crystals





with decreasing T:

- Kondo increase
- coherence maximum
- drop to ρ_0

strong decrease of the residual resistivity ρ_0 in magnetic field above B_c:

$\Delta\rho_0/\rho_0$ strongest for ρ II c

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

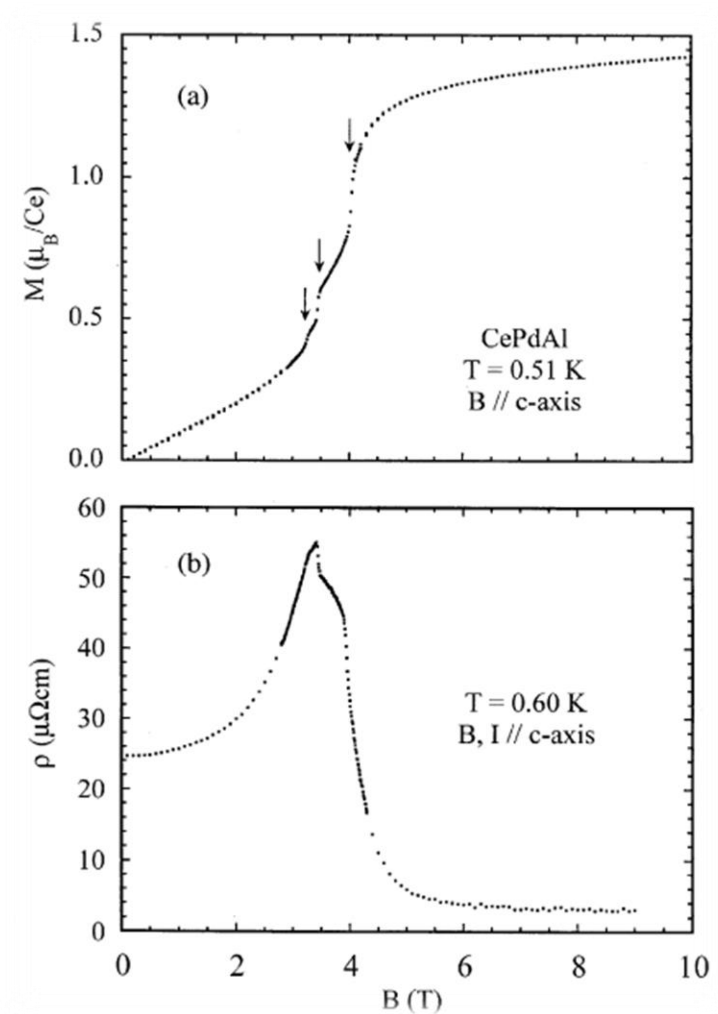
- no indication of Kondo effect by non-ordered frustrated Ce moments
- assuming T^2 resistivity:

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

  Spinon excitations?

Field-induced phases in CePdAl close to the critical field

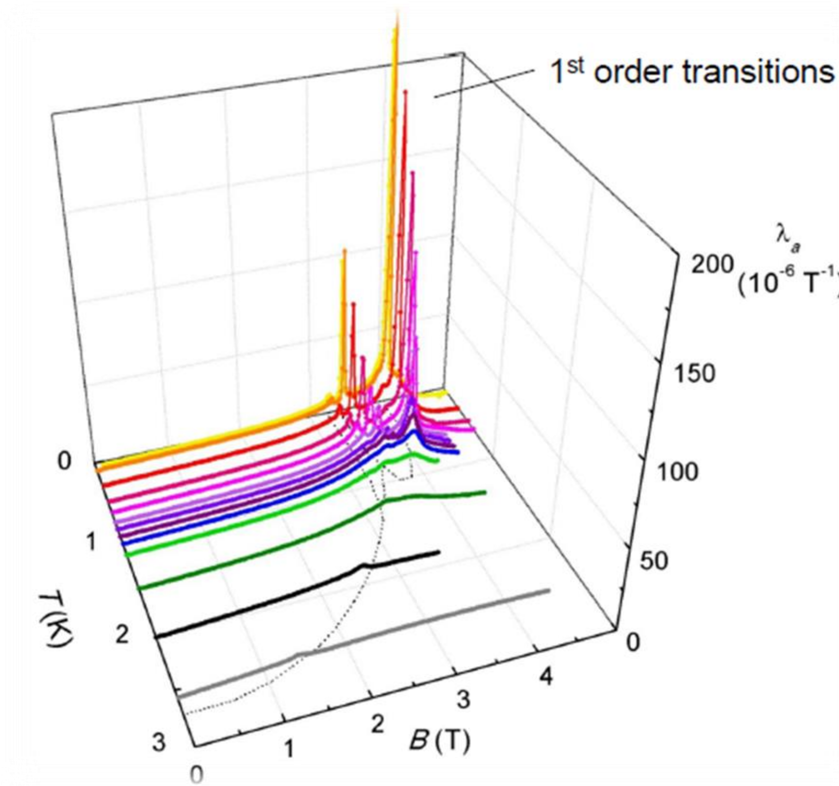
T. Goto et al.,
J. Phys. Chem. Sol. 63, 1159 (2002)



Features in magnetizattion *M*(*B*) and resistivity ρ(*T*) are suggestive of first-order transitions

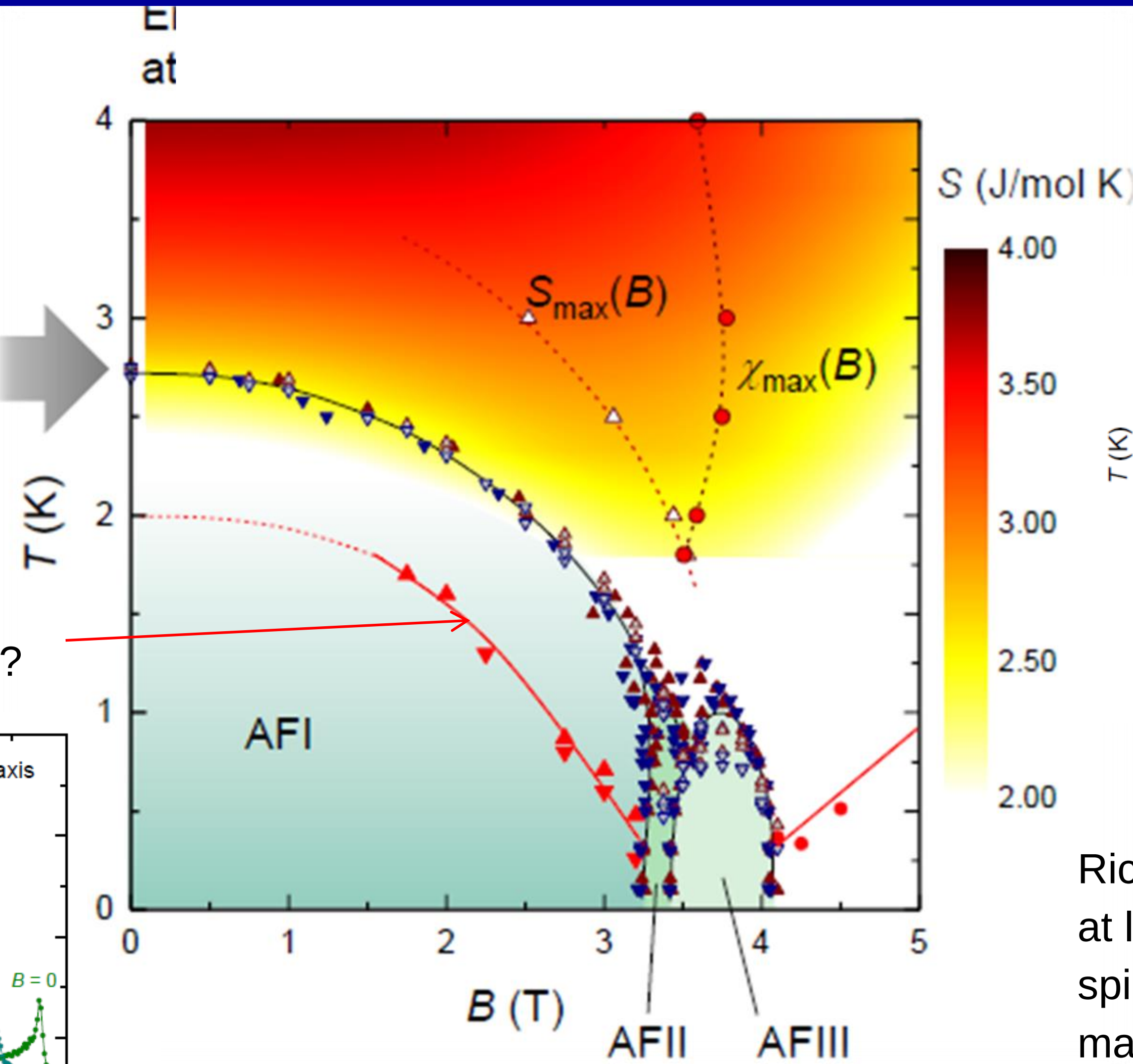
Possible origin: lifting of geometric frustration

Check with magnetostriction measurements

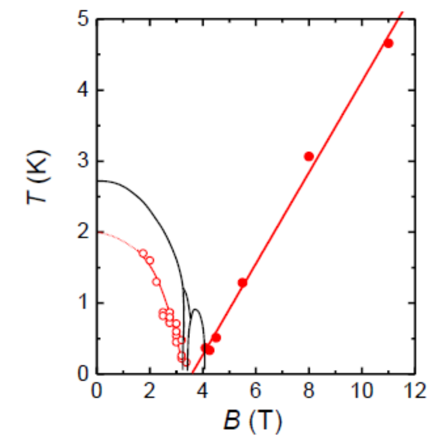


K. Grube et al., unpublished

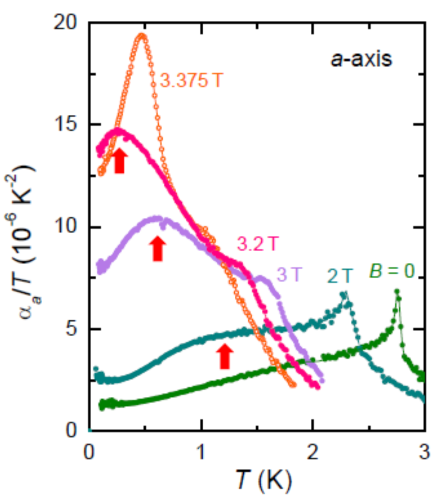
Magnetic phase diagram of CePdAl from thermal expansion and magnetostriction



High-field Schottky anomaly due to Zeeman splitting



Crossover to spin-liquid?



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

K. Grube et al., unpublished

Electronic correlations in $A\mathrm{Fe}_2\mathrm{As}_2$ (A = K, Rb, Cs) probed by quantum oscillations of the magnetostriction

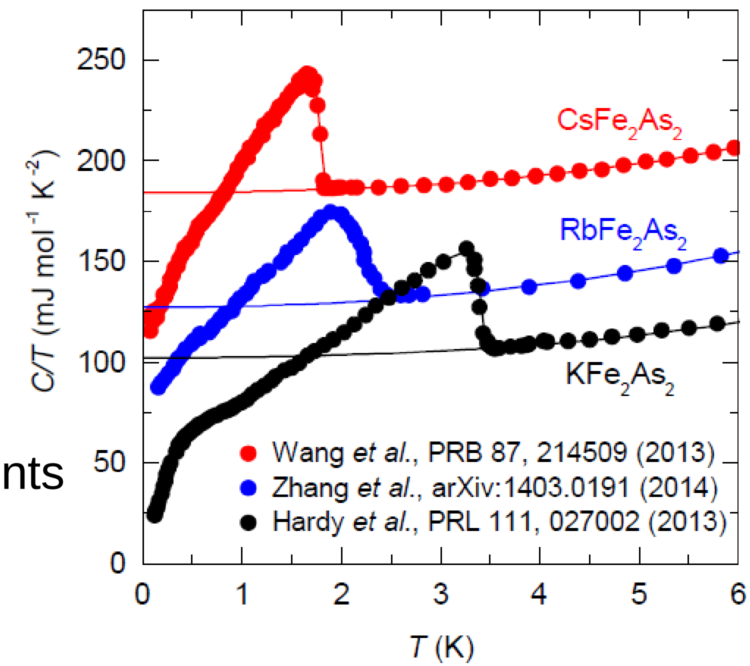
Strong correlations in AFe_2As_2 (A = K, Rb, Cs)

KFe_2As_2
Sommerfeld coefficient $\gamma = 94$ mJ / mol K^2

Kadowaki-Woods ratio

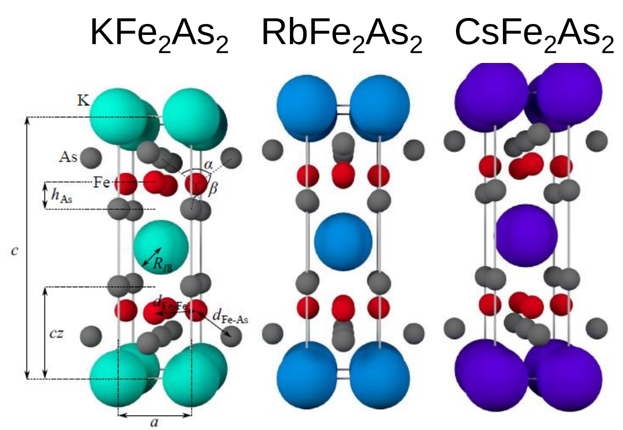
$$A/\gamma^2 = 2 \cdot 10^{-6}\ \mu\Omega\ \mathrm{cm}\ K^2\ \mathrm{mol}^2\ \mathrm{mJ}^{-2}$$

*F. Hardy et al., PRL **111**, 027002 (2013)*



Large γ in line with ARPES and dHvA measurements

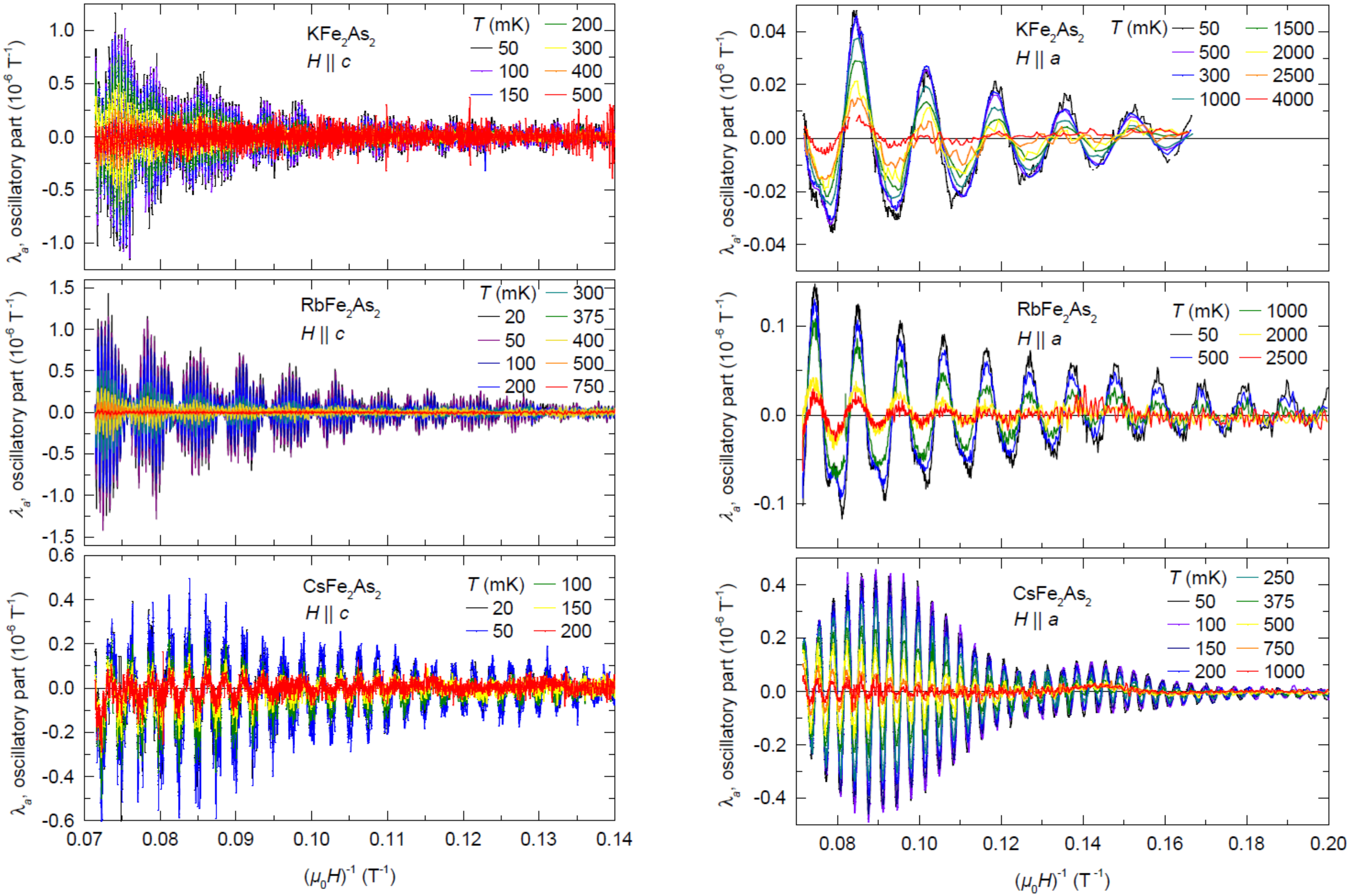
γ increases for $RbFe_2As_2$,
reaching 180 µJ / mole K^2 for $CsFe_2As_2$,



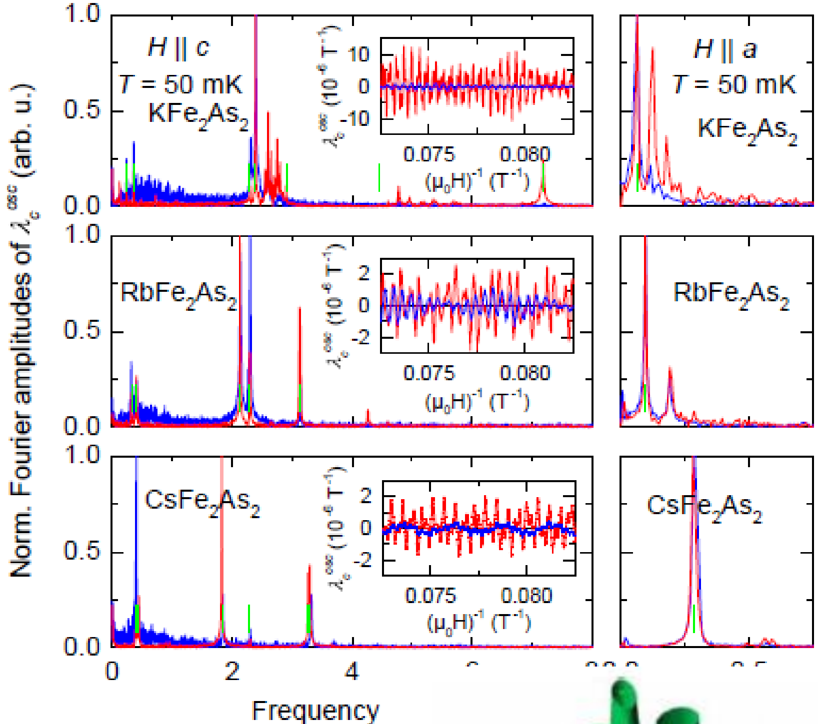
Increasing lattice constant and Fe-As distance along the K-Rb-Cs series: origin of strong correlations?

→ Quantum oscillations, here: magnetostriction

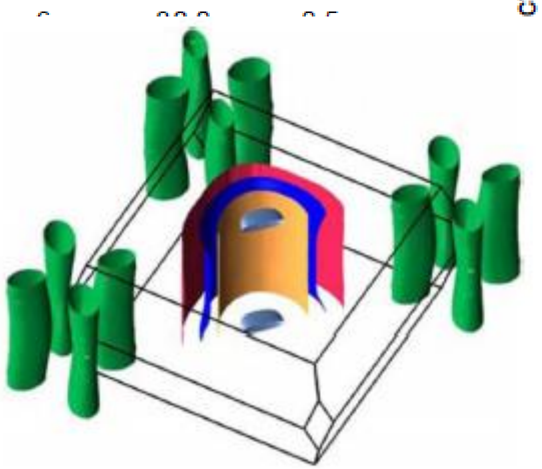
Quantum oscillations in the magnetostriction of AFe_2As_2 (A = K, Rb, Cs)

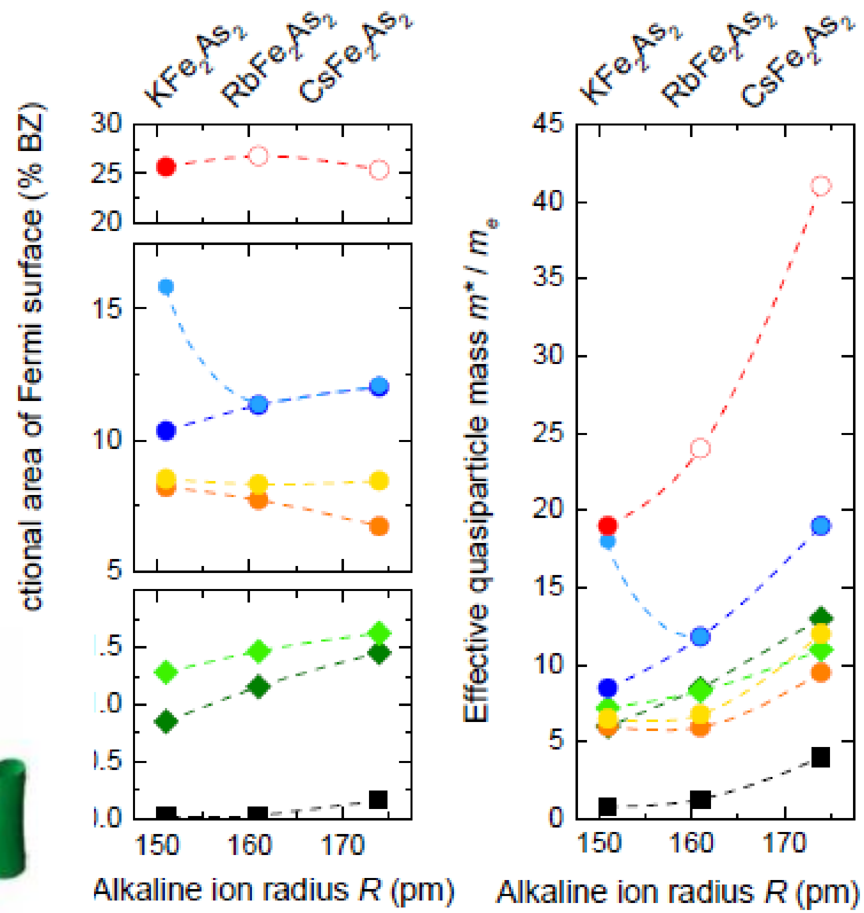


Effective masses of the series AFe_2As_2 (A = K, Rb, Cs)



Band structure of KFe_2As_2

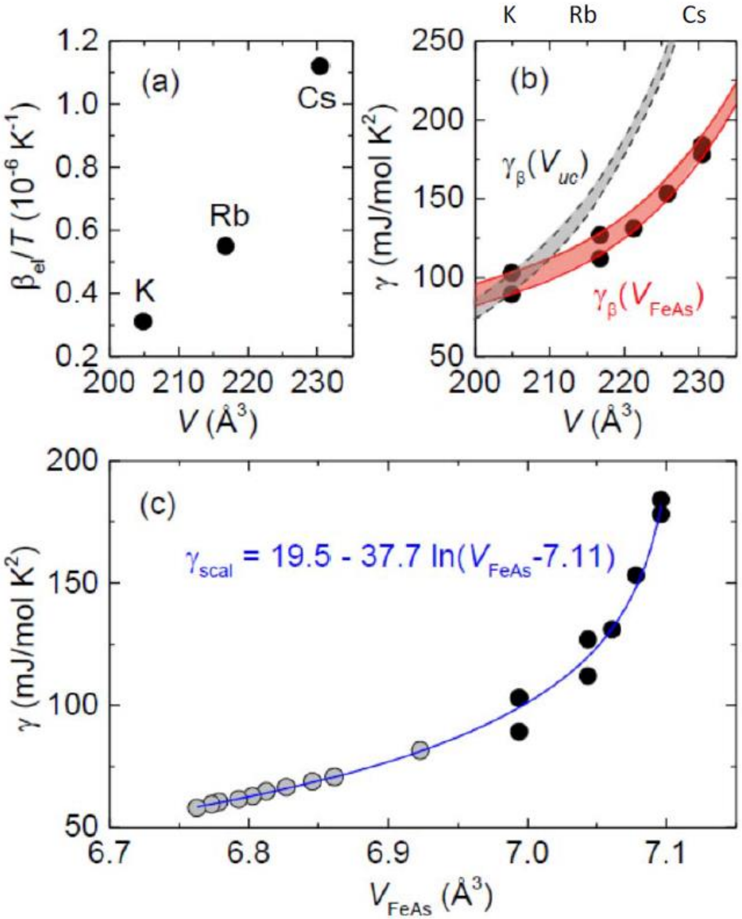




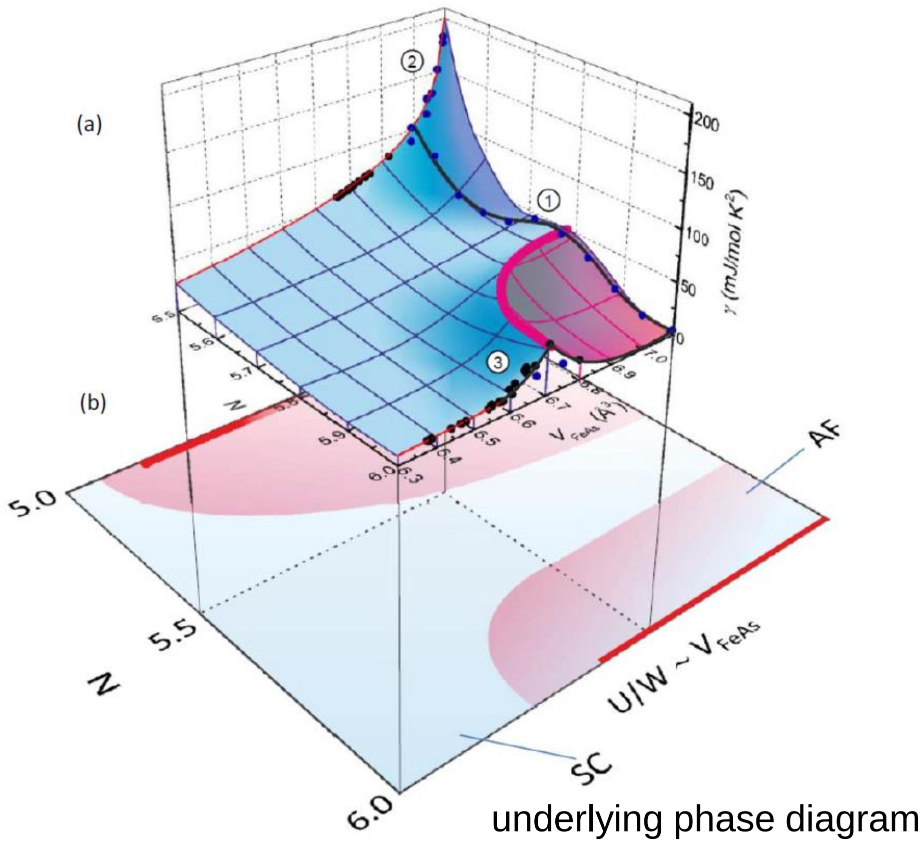
Hashamoto et al.,
PRB 82, 014526 (2010)

Evolution of the Sommerfeld coefficient γ

Volume dependence of γ



Yu et al., Curr. Opin. Sol. State Mat. Sci. ***17****, 65 (2013)*



underlying phase diagram
proposed by Yu et al.

(1) Substitution of Ba by K

(2) log divergence of γ with increasing V_{FeAs}

(3) Substitution of As by P. volume effect

Summary

CePdAl – a partially geometrically frustrated heavy-fermion metal

- Approach to QCP by Ni substitution:

  $C/T \sim -\log(T/T_0) \rightarrow$ 2D AF quantum critical fluctuations or novel QCP?

- Rationale für 2D fluctuations: AF planes decoupled by frustrated moments?
- No indication of a low-*T* Kondo effect of frustrated moments: 2D spin liquid ?
- Complex magnetic (*B*,*T*) phase diagram with several phases near B_c:

  lifting the frustration close to B_c

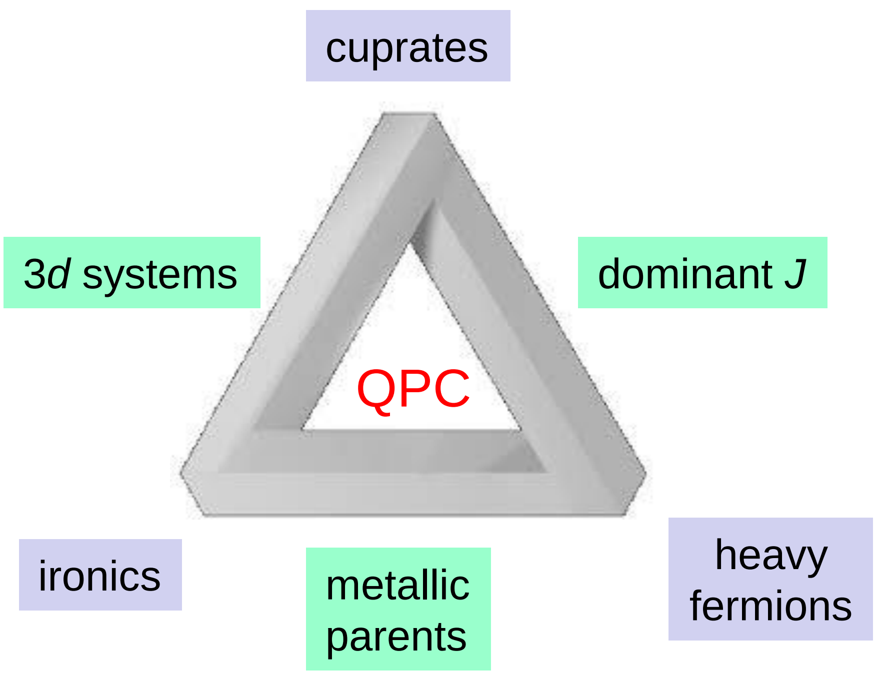
AFe_2As_2 – a route toward orbital selective Mott transition driven by volume expansion

General issues

- Universality classes of quantum phase transitions in metallic magnets?
- Electrons: spectators or activists?
- Spin liquid in the presence of frustrated magnetic moments?

The magic triangles of correlated systems

Materials



Phenomena