Institute of Physics, Chinese Academy of Sciences

Heavy Fermion Physics: Perspective and Outlook

Quantum Phase Transition in a Partially Frustrated System: CePdAl

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Strongly correlated electron systems: electrons at the verge of localization

Electrons localized: magnetism (unless electron shells are completely occupied) due to exchange (= Coulomb interaction + Pauli principle)

Electrons delocalized: superconductivity is the rule rather than the exception, but one needs a "glue" to overcome electron repulsion:

electron phonon coupling for "conventional" superconductors

"magnetic" coupling via magnons, spin fluctuations, magnetic excitons in "unconventional" superconductors…

Strongly correlated electron systems: electrons at the verge between localization and itinerancy

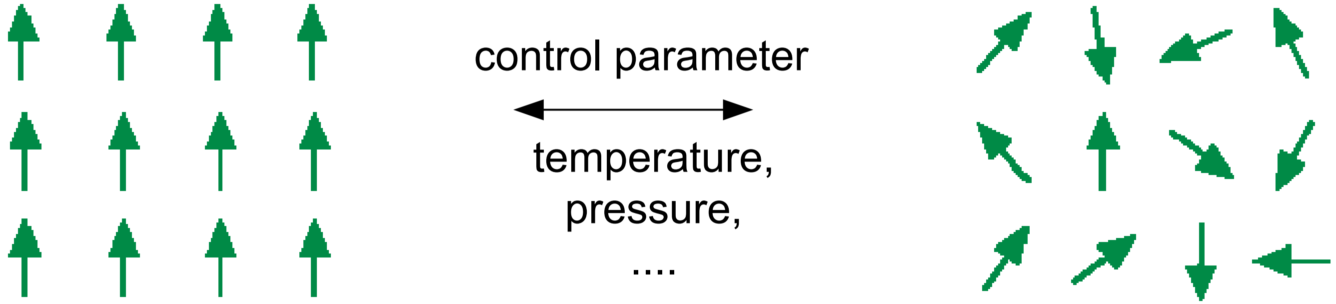
Superconductivity and magnetism are often found in close proximity to each other, unlike weakly correlated metals where weak magnetic fields and/or magnetic impurities suppress superconductivity

Electron interactions depend on inter-electron distance: hence strong coupling between electrons and lattice dynamics

Magnetic instabilities in metals: how does a magnetic solid "melt"?

magnetic order

disordered magnetic moments

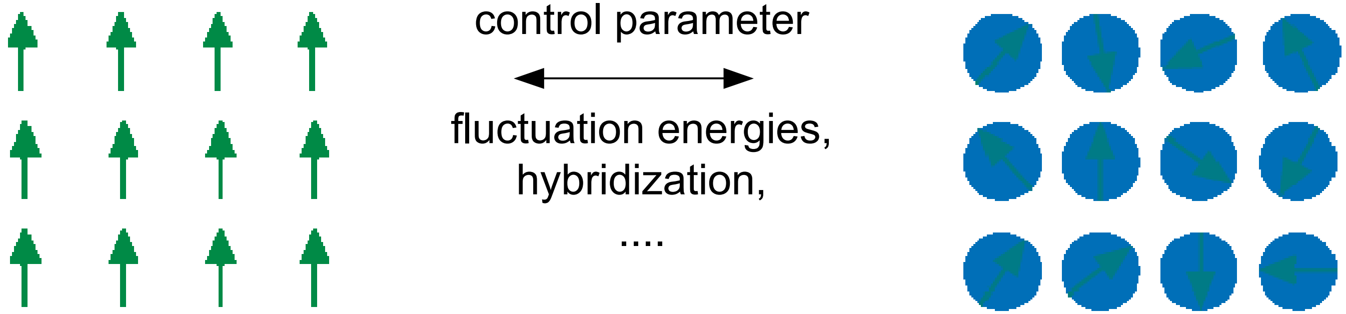


control parameter

temperature,
pressure,
....

magnetic order

magnetic moments „lost“



control parameter

fluctuation energies,
hybridization,
....

scenarios:

charge fluctuations, Kondo effect, itinerant magnetism

Outline

Introduction: Quantum phase transitions - General

Quantum phase transitions in heavy-fermion metals, e.g., $CeCu_{6-x}Au_x$

$CeCoIn_5$: line of quantum critical points hidden by the superconducting dome

CePdAl – a partially frustrated heavy-fermion system

- magnetic phase diagram
- Approach to quantum criticality by Ni doping

Partial order in MnSi under pressure for $T_C \rightarrow 0$ and for $p = 0$ at T_C

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M. Garst, A. Rosch, M. Vojta, P. Wölfle, P. Coleman, Q. Si, J. Kroha

Territory of theory

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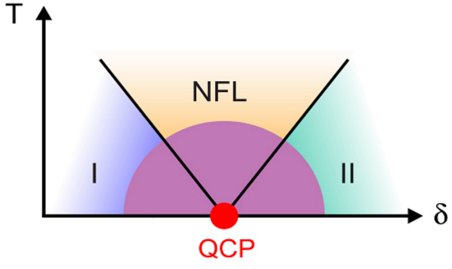
and Helmholtz Association of Research Centers



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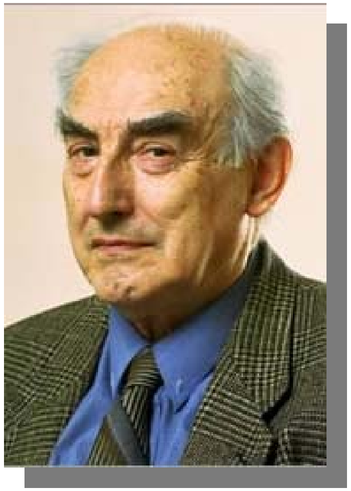
Quantum phase transitions in heavy-fermion metals

The Standard Model of phase transitions: Ginzburg-Landau-Wilson theory

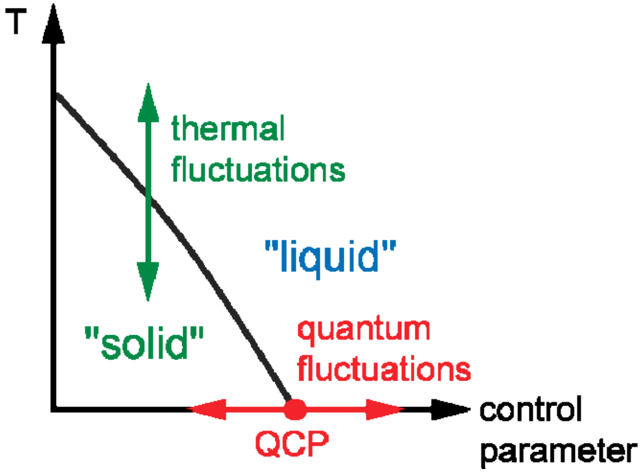


V. Ginzburg

Universality:

critical behavior (exponents α, β, γ, ν, ...) depend on spatial dimension and symmetry of the order parameter only because correlation length diverges at T_c, $\xi \sim |T - T_c|^{-\nu}$

correlation time: $\tau \sim \xi^z$ (“critical slowing down“)



G. K. Wilson

J. A. Hertz

$T_c \rightarrow 0$: energy of fluctuations $\hbar/\tau$ important:
temperature sets the system size
in the time direction: $d \rightarrow d + z$

Problem: low-energy fermions

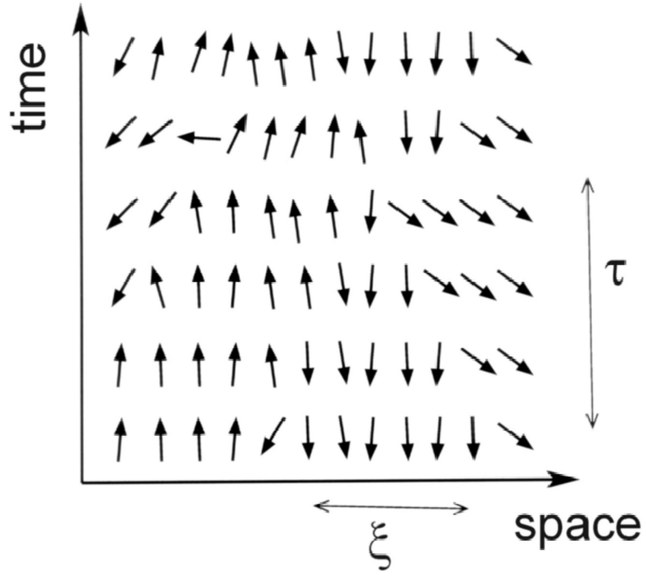
Quantum phase transitions (2^{nd} order)

Energy scale of quantum effects: $\frac{\hbar}{\tau}$

classical case: $\frac{\hbar}{\tau} \ll k_B T \approx k_B T_c$, quantum fluctuations negligible

quantum case: $\frac{\hbar}{\tau} \geq k_B T$, possible if $T_c \rightarrow 0$

spatial *and* temporal fluctuations determine dynamics, $\tau \sim \xi^z$



effective dimension $d_{eff} = d + z$

new universality classes

role of temperature

control paramter

thermal excitations via $k_B T$

finite system size $\tau \leq \hbar / k_B T$

The Standard Model of metals: Landau Fermi-liquid theory

L. D. Landau

1:1 correspondence between excitations of interacting and noninteracting systems:

"Fermi liquid"

Electron-electron interactions parametrized by few parameters m^*, F_0^a, F_0^s, ...

$$C = \gamma T = \frac{m^*}{m_0} \gamma_0 T; \quad \chi = \frac{m^*}{m_0} \frac{1}{1+F_0^a} \chi_0$$

$$\Delta \rho \sim T^2$$

Since ~ 1990: many systems show deviations:

"non-Fermi liquids"

NFL behavior can arise from distinctly different physical origins:

Multichannel Kondo effect
Distribution of Kondo temperatures
Quantum phase transitions

Origin of heavy masses $m \approx 100\ m_0$ in Ce- and Yb-based rare-earth alloys

Two “ingredients“:

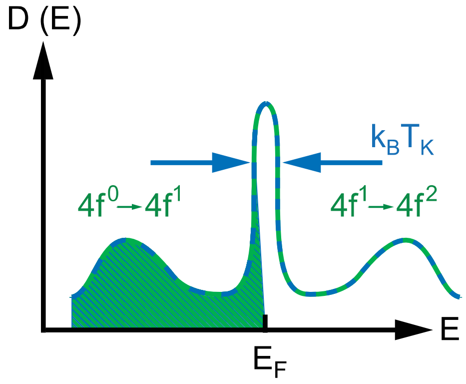
- hybridization of 4*f* and conduction electrons
- strong on-site electron repulsion in 4*f* state

Singly occupied lowest 4*f* state will be screened by conduction electrons:

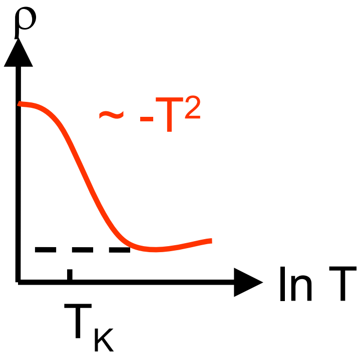
singlet formation

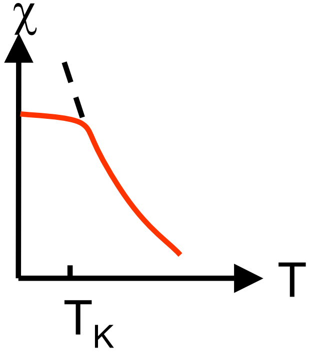
Resonance at E_F due to virtual excitions from 4*f* state to E_F

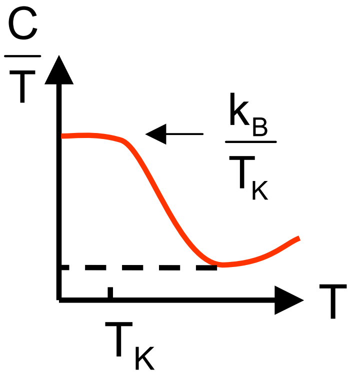
Kondo resonance



“Kondo anomalies“ at low *T*



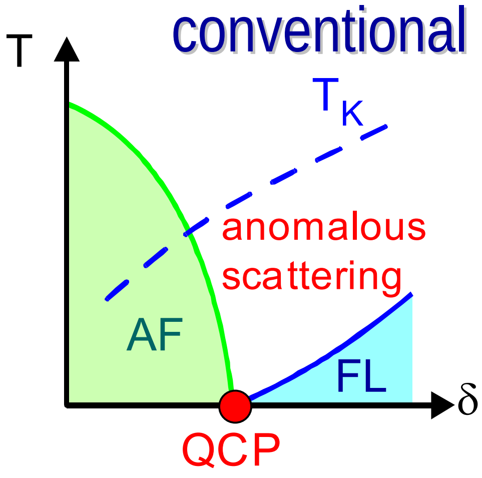


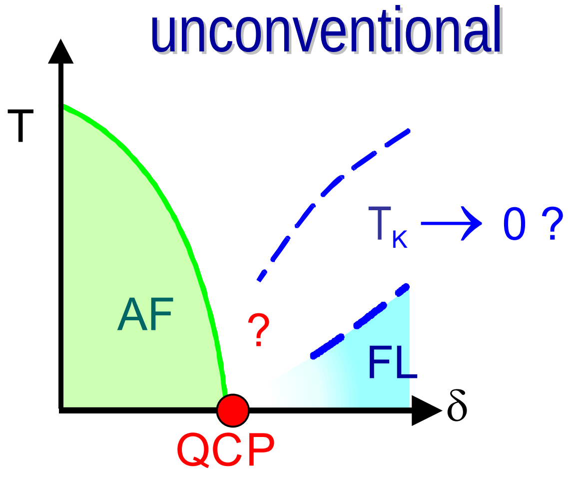


local Fermi liquid

Heavy-fermion system: lattice-coherent superposition of Kondo anomalies

Quantum criticality: “good guys vs. bad guys“





Scattering of heavy quasiparticles by spin fluctuations: diverging *m** for 3D FM and 2D AF

Unbinding of heavy composite quasiparticles: change of Fermi volume

$CeNi_2Ge_2$

$CeCu_2Si_2$

$CeCoIn_5$

$Ce_{1-x}La_xRu_2Si_2$

$CePd_2Si_2$

$CeRhIn_5$

$CeCu_{6-x}Au_x$

$YbRh_2Si_2$

$UCu_{5-x}Pd_x$

Multiple energy scales?
Dimensionality ?
Disorder effects ?

Hertz, Millis, Moriya, Rosch et al.

Coleman, Si, Pepin et al.

Magnetic order in $CeCu_{6-x}Au_x$

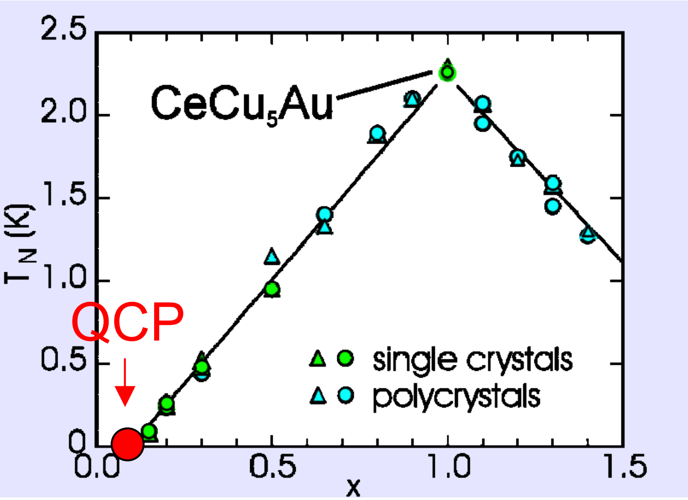
$CeCu_6$: heavy fermions with $\gamma = 1.6$ J/molK^2

non-magnetic groundstate

Ōnuki et al., Amato et al.

short lived AF correlations

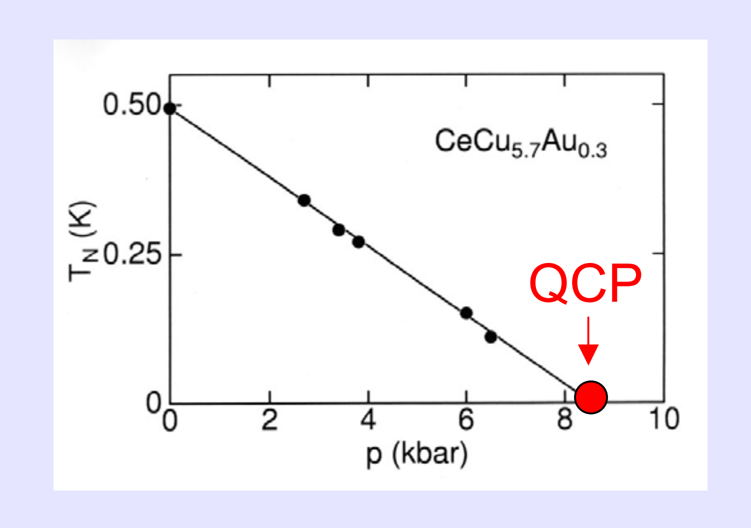
Aeppli et al., Rossat-Mignod et al.



Alloying with Au: long-range AF order

"negative lattice pressure" explains $T_N(x)$ for $x < 1$

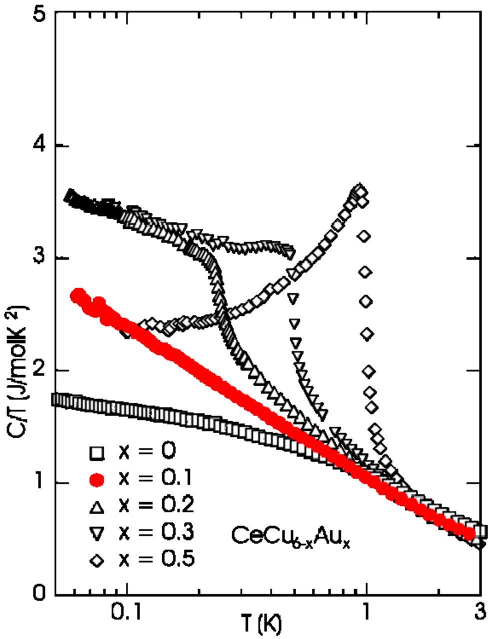
Direct proof: Néel temperature T_N vanishes under hydrostatic pressure



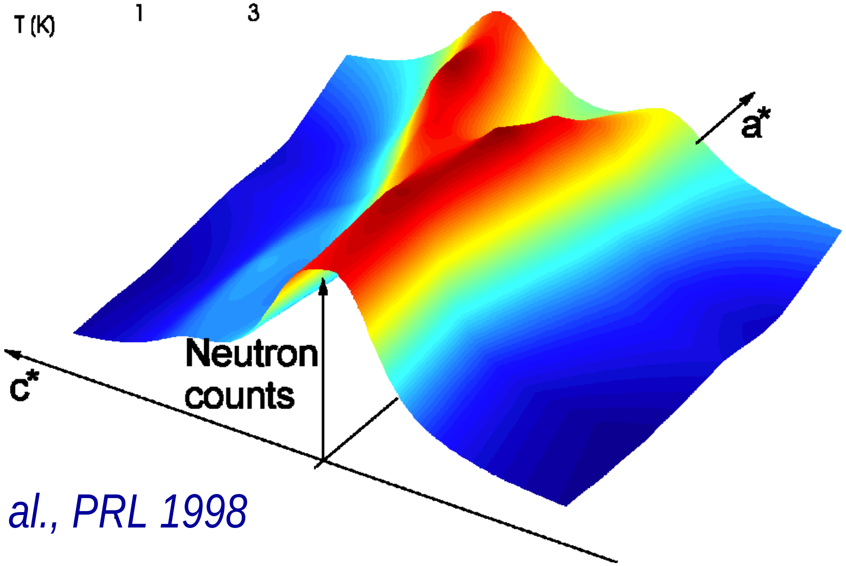
$x = 0.1$: Quantum critical point with "non-Fermi liquid" behavior

What‘s so special about $CeCu_{6-x}Au_x$?

Specific heat

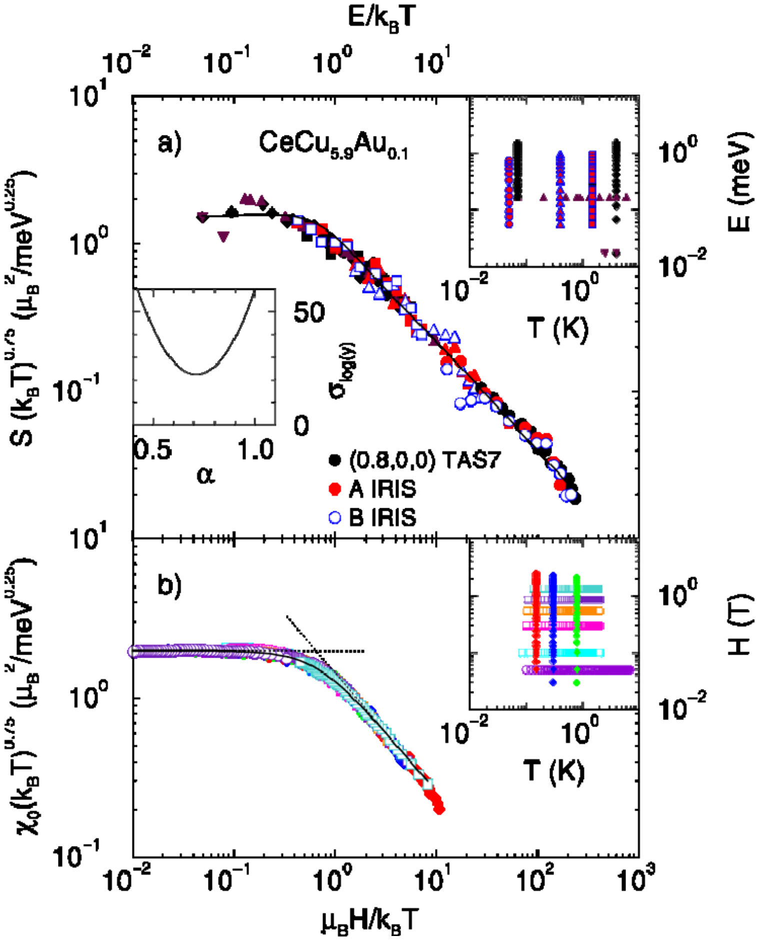


Anomalous *q*-dependence of critical fluctuations in a wide *x* range



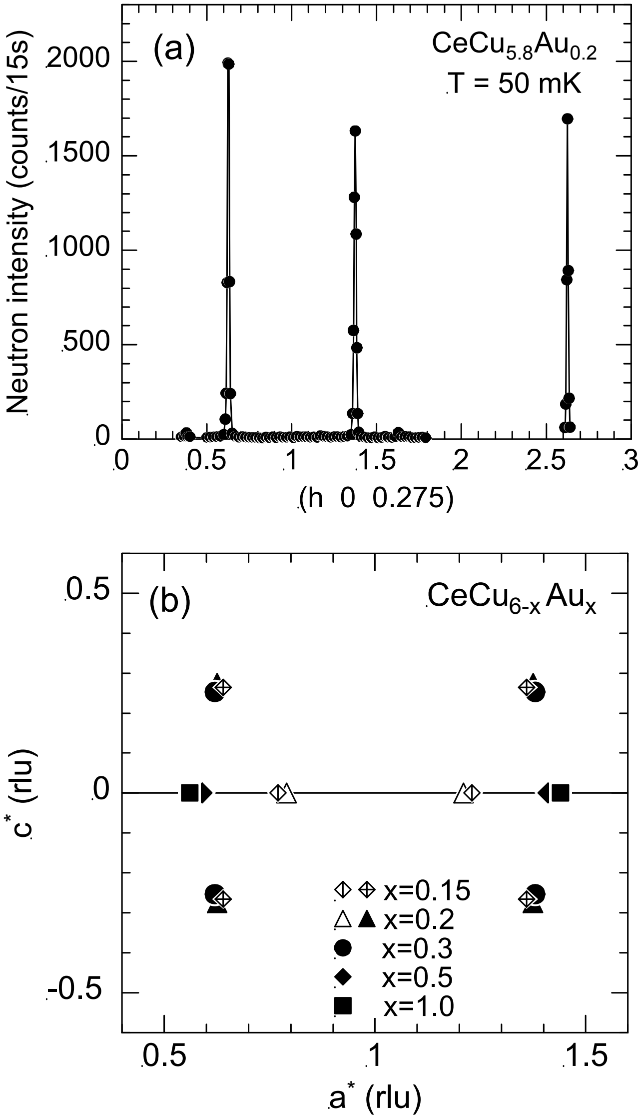
O.Stockert et al., PRL 1998

ω/*T* scaling of critical fluctuations for *x* = 0.1, independent of *q*



Schröder et al., PRL 1998; Nature 2000

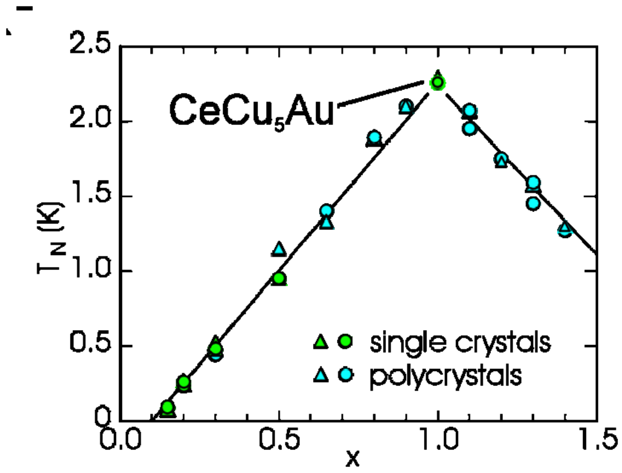
Incommensurate magnetic order of $CeCu_{6-x}Au_x$

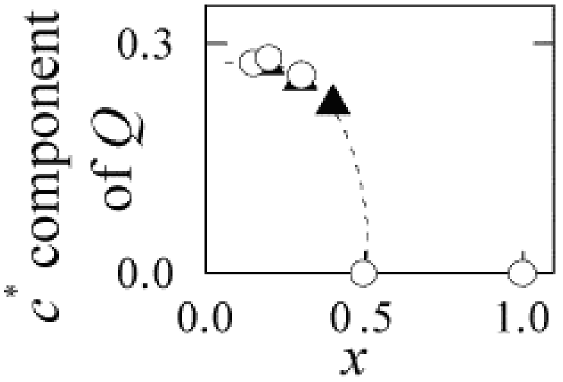


Three-dimensional magnetic order, **Q** vector confined to the *a*c** plane

$CeCu_{6-x}Au_x$ is almost Ising like:
$M_c : M_a : M_b = 10:2:1$

“Jump“ of ordering wave vector to the *a** axis between $x = 0.3$ and $x = 0.5$ while $T_N(x)$ varies smoothly

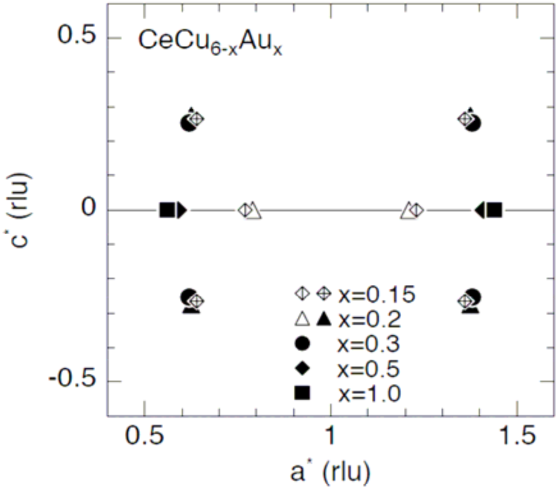


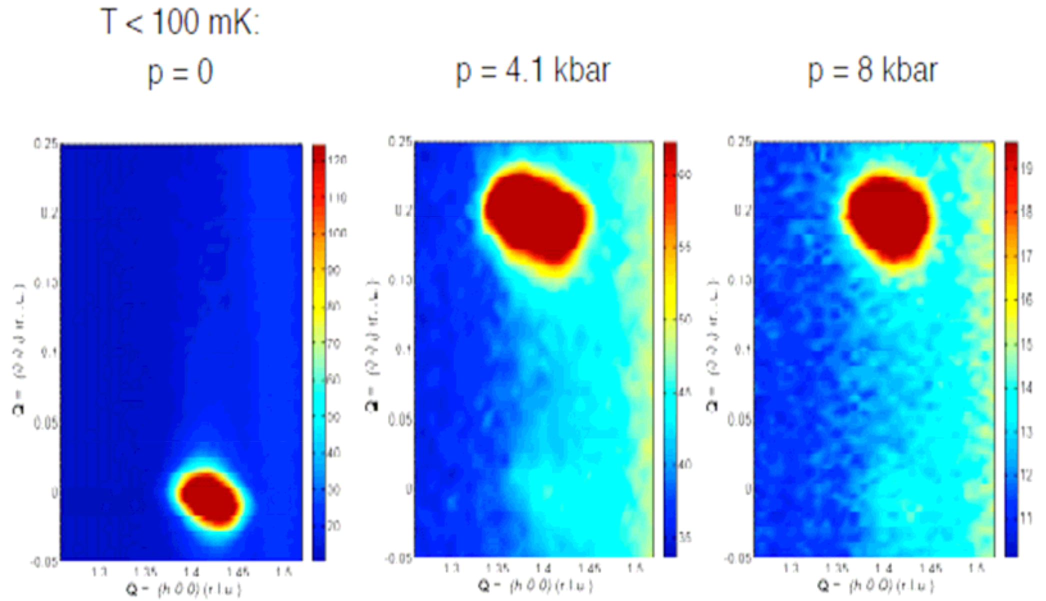


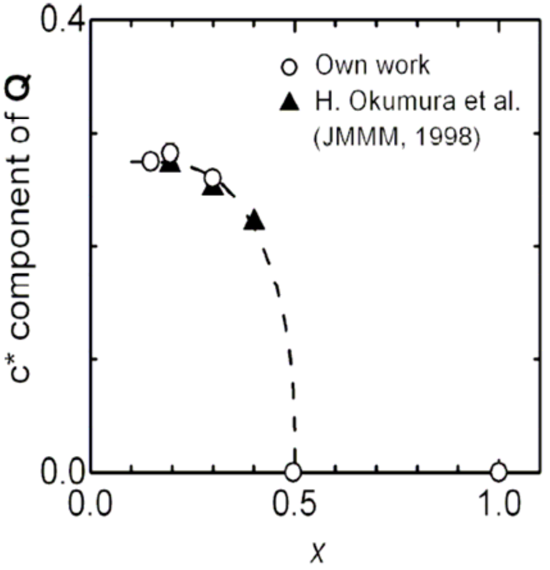
We know that we can tune quantum criticality by concentration or hydrostatic pressure.
How about the magnetic order under pressure?

Evolution of the magnetic structure of $CeCu_{5.5}Au_{0.5}$ under hydrostatic pressure

A. Hamann, D. Reznik, O. Stockert, V. Fritsch

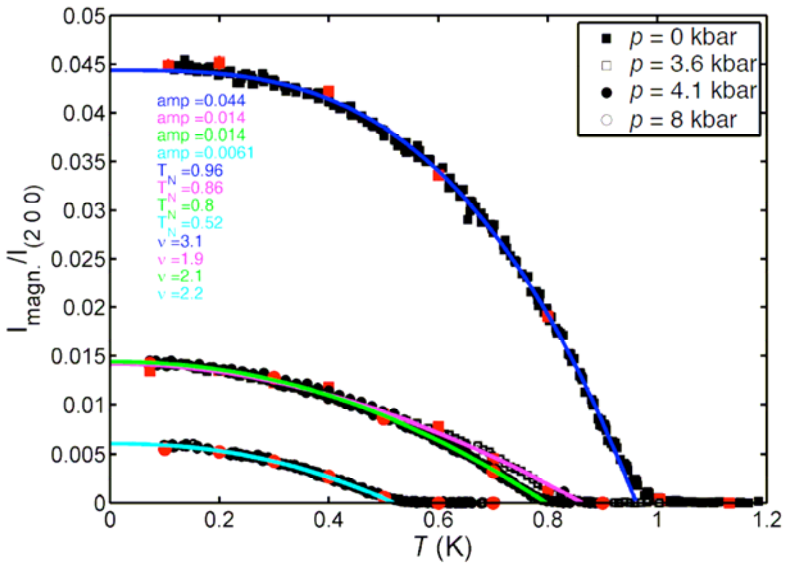




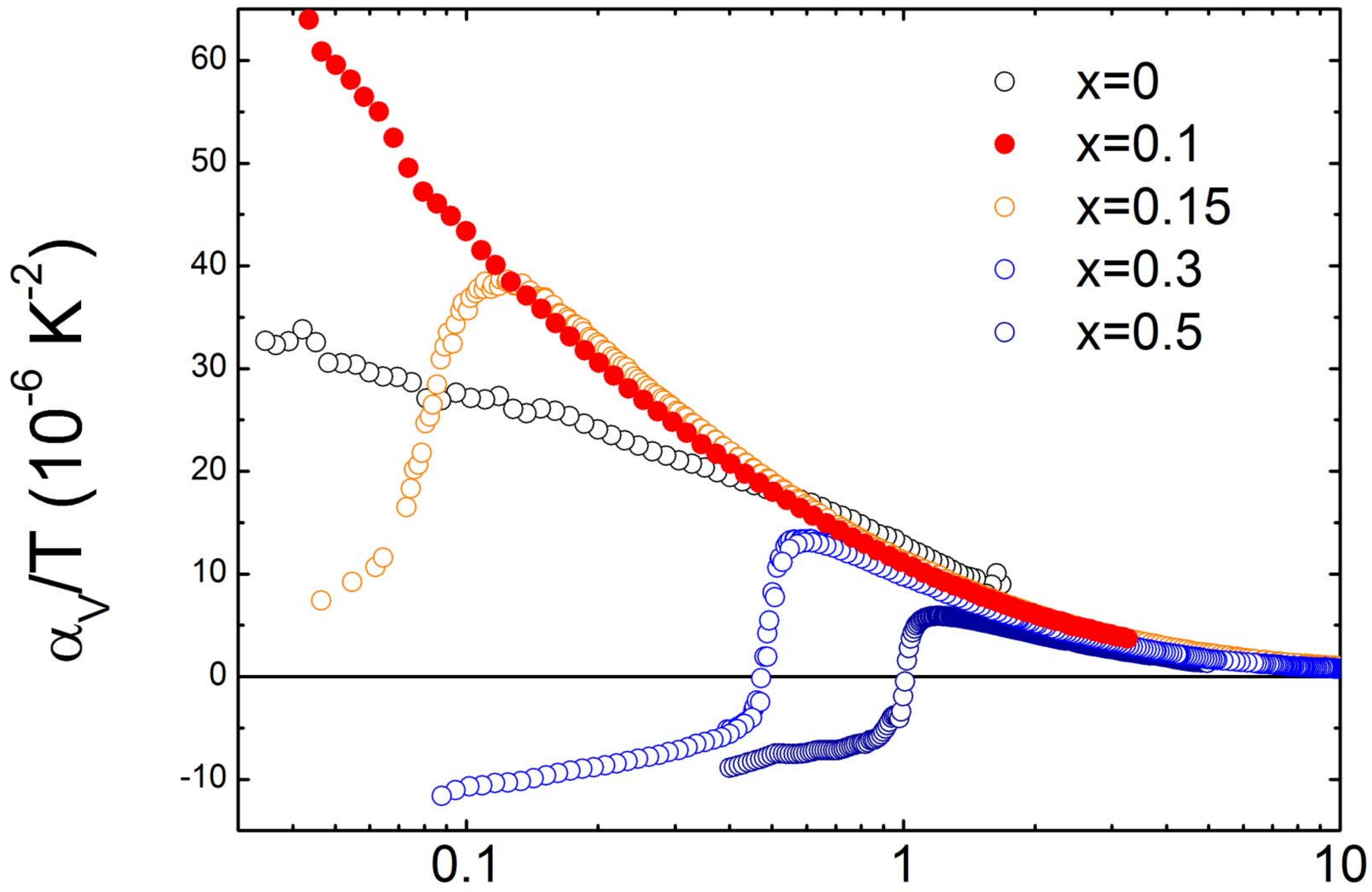


Change of T_N, ordering wave vector, and ordered moment with pressure ($p = 0 \rightarrow 8$ kbar) and Au content ($x = 0.5 \rightarrow 0.3$) is nearly identical!

Strong change of **Q** is in marked contrast to smooth $T_N(x)$ dependence



Scaling of the volume expansivity $CeCu_{6-x}Au_x$

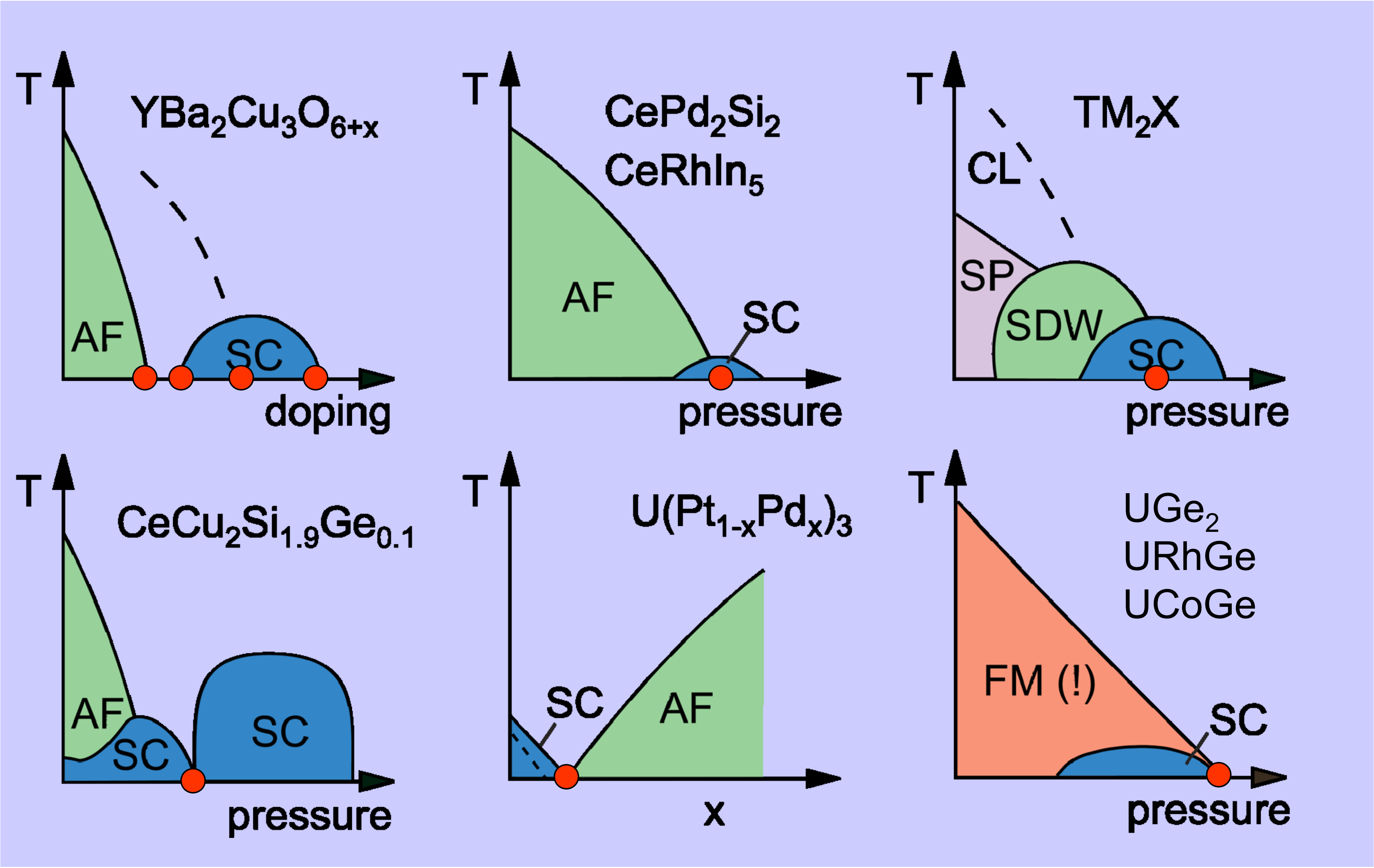


Change in magnetic ordering wave vector between $x = 0.3$ and $x = 0.5$: associated with sign change of jumps in $\Delta\alpha_i$ at T_N.

Remarkable scaling of $\alpha_V(T)$ despite the different α_i's. : Robustness of QCP

Towards establishing a $(p,B.T)$ phase diagram of quantum criticality near B_{c2} in $CeCoIn_5$

Interplay of superconductivity and magnetism near quantum critical points



The “hidden“ QCP in $CeCoIn_5$

Non-Fermi liquid behavior near B_{c2},
e. g., $C/T \sim \ln (T_o/T)$ (2D AF Hertz-Millis)

A. Bianchi et al., Phys. Rev. Lett. ***91****, 257001 (2003)*

No signature of AF order near B_{c2} in $CeCoIn_5$

Additional phase within the SC region: “Q phase“

M. Kenzelmann et al., Science ***321****, 1652 (2008)*

Cd doping induces AF order

Y. Tokoiwa et al., Phys. Rev. Lett. ***101****, 037001 (2008)*
S. Nair et al., PNAS ***107****, 9537 (2010))*

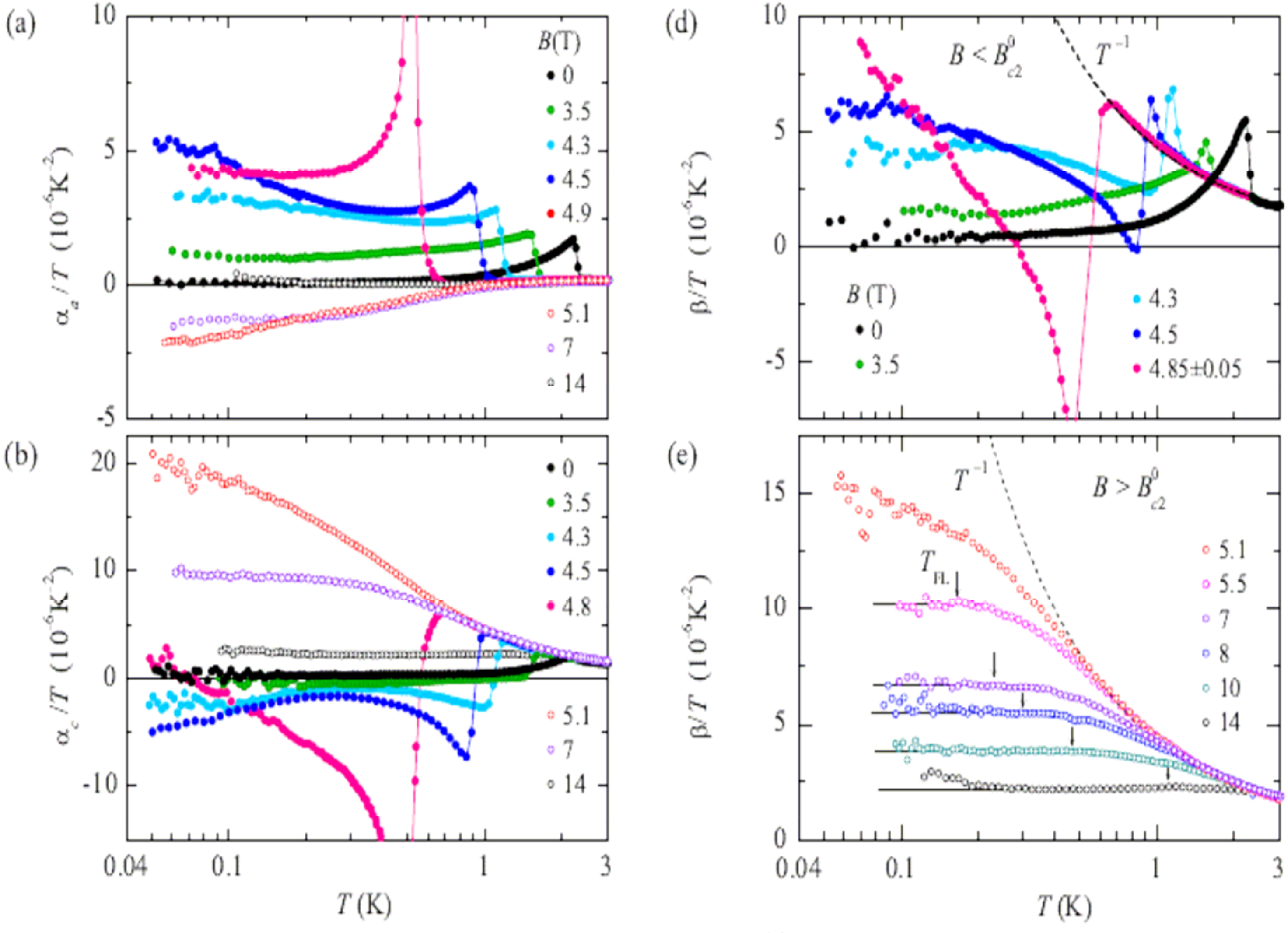
2D-3D crossover from previous thermal-expansion measurements

J. G. Donath et al., Phys. Rev. Lett. ***100****, 136401 (2008)*

Thermal expansion of $CeCoIn_5$ for $B \parallel c$

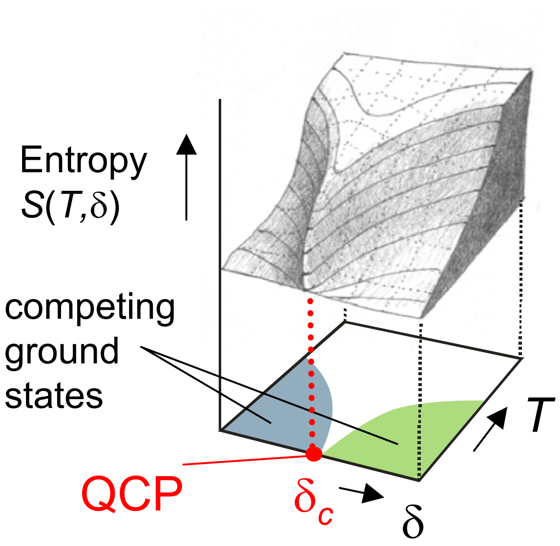
Linear expansion $\alpha \parallel a$ and c

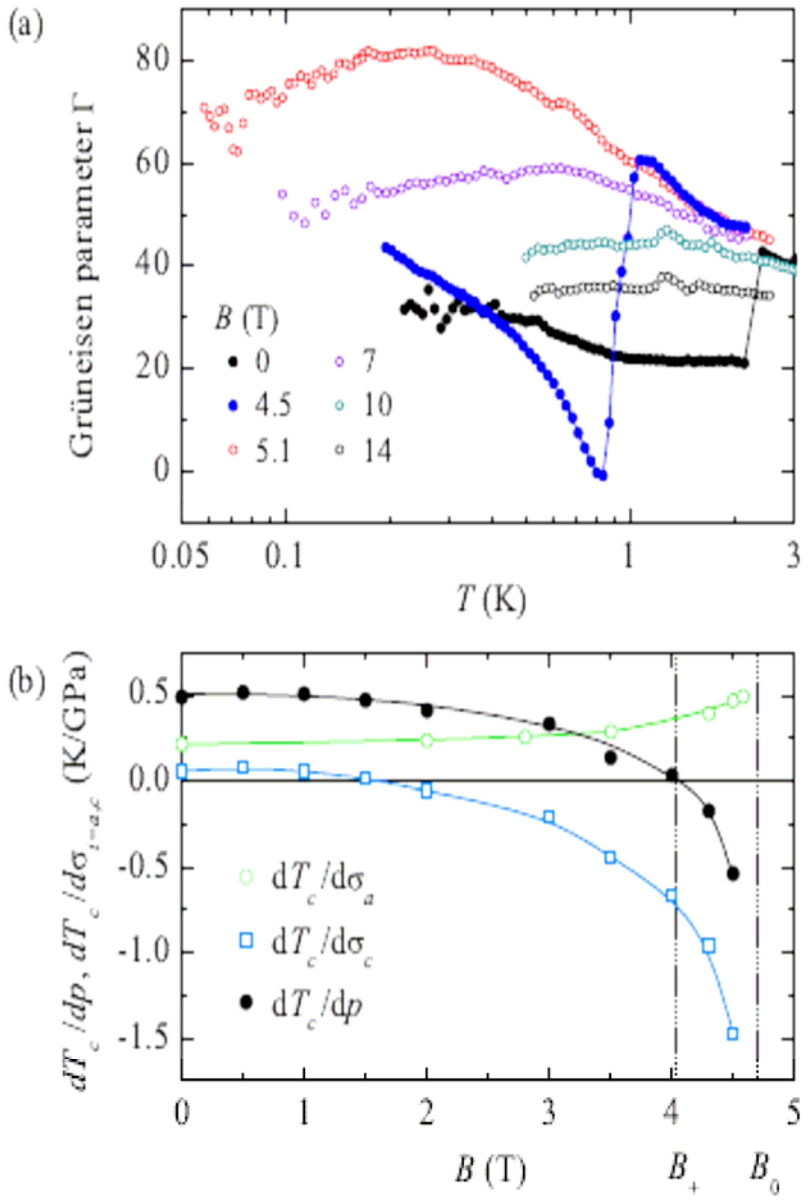
Volume expansion β



Grüneisenparameter as a test for quantum criticality

*Garst, Rosch PRB **72**, 205129 (2005)*



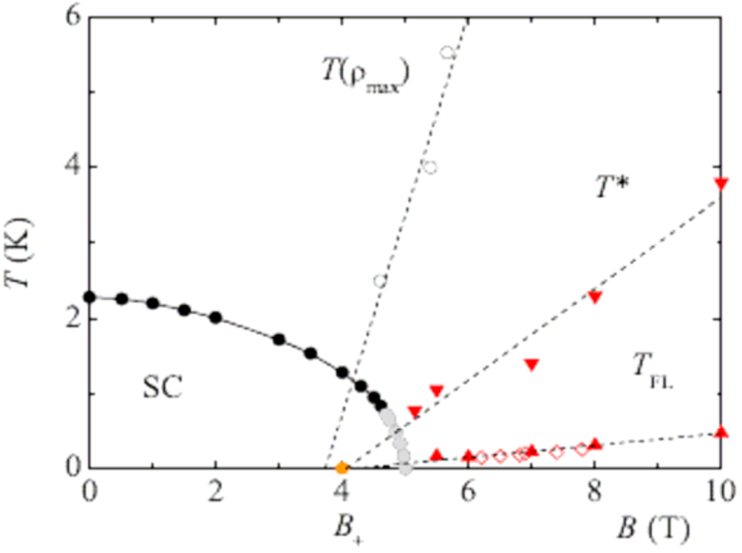


Grüneisen parameter remains positive throughout investigated field range: no QCP at $p = 0$

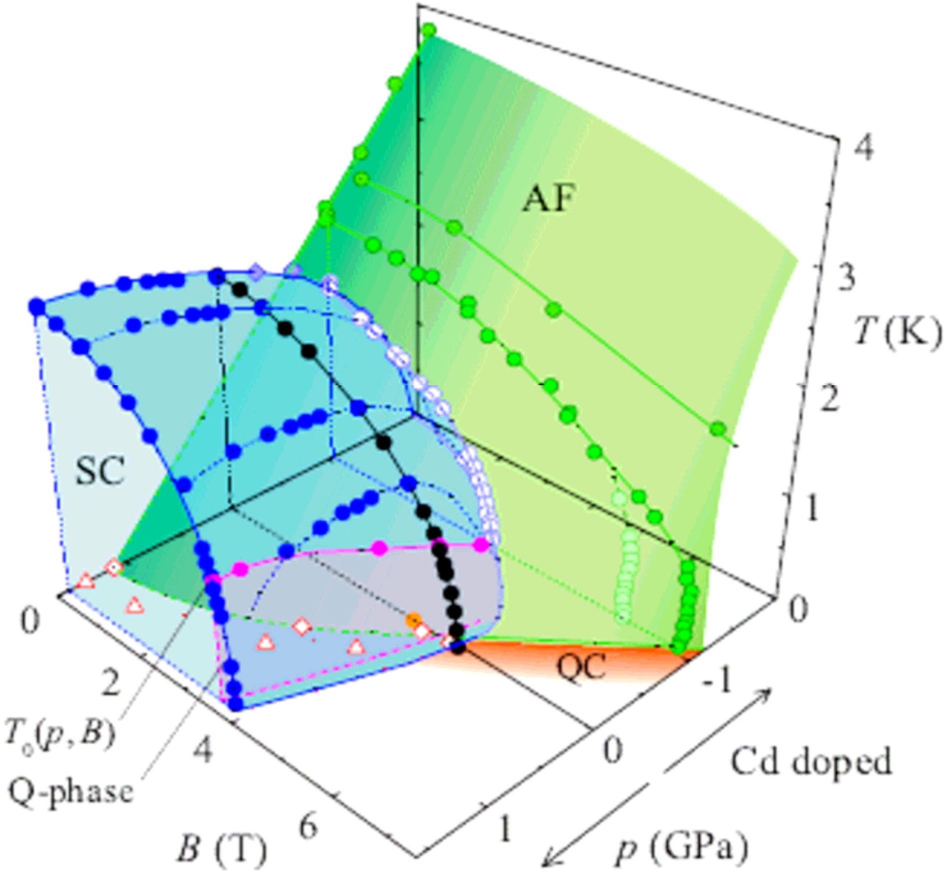
Sign change of dT_c/dp at $B_+ = 4.1$ T: maximum $T_c(p)$ of superconducting dome

Possibility of a line of quantum critical points in the (p,B) plane of $CeCoIn_5$

S. Zaum et al., PRL 2011



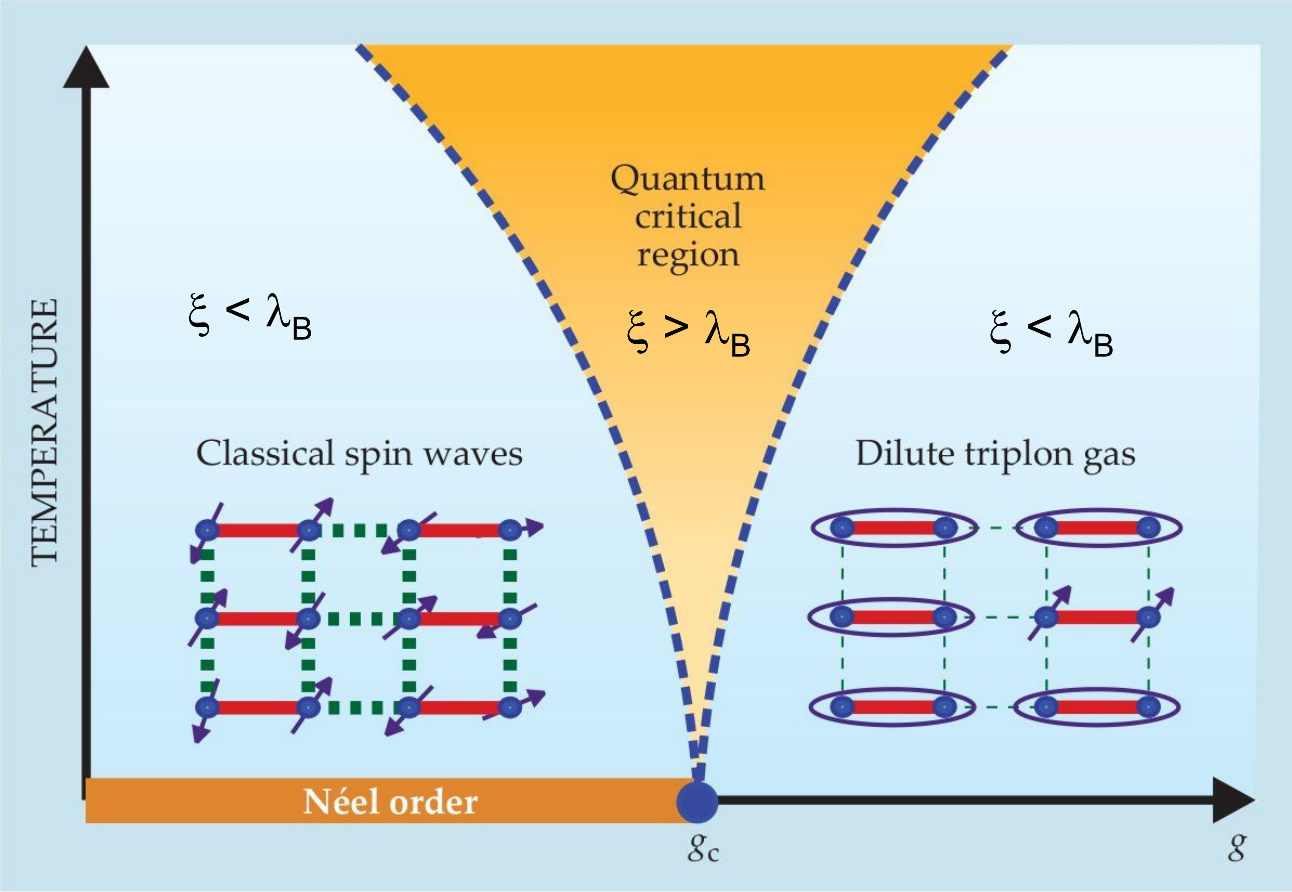
$T(\rho_{max})$:	*Paglione et al., PRL 2003*
$T_{FL,Hall}$	*Singh et al., PRL 2007*



$T_{c,max}(p)$	*Ronning et al., PRB 2003*
	Tayama et al., JPSJ 2005
	Knebel et al., Phys. Stat. Sol. 2010
$T_{c,max}(p)$	*Lengyel 2008*
Cd doping	*Pham et al., PRL 2007*
	Donath et al., Phys. B 2008
	Tokiwa et al., PRL 101
	Nair et al., PNAS 2010

Frustration as a tuning parameter for QPT in metals?

Phase diagram of S = ½ 2D insulating magnet



S. Sachdev and B. Keimer, Phys.Today, Feb. 2011

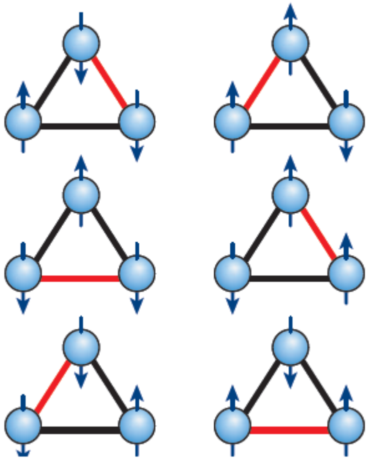
Coherence length ξ and de-Boglie wavelength $\lambda_B = \hbar c/k_B T$ of the excitations

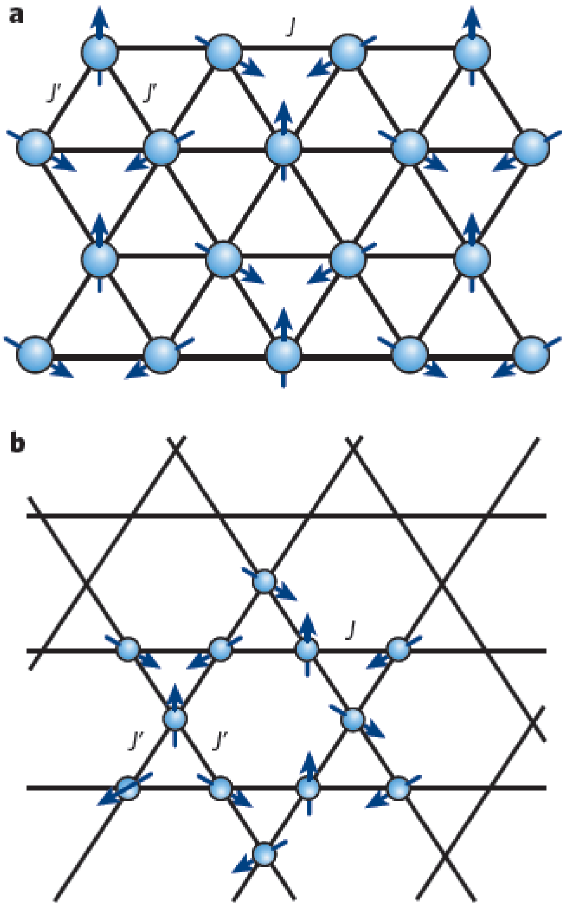
Quantum critical range: $\xi > \lambda_B$, temperature is the only relevant energy scale

Frustration as a route to quantum criticality

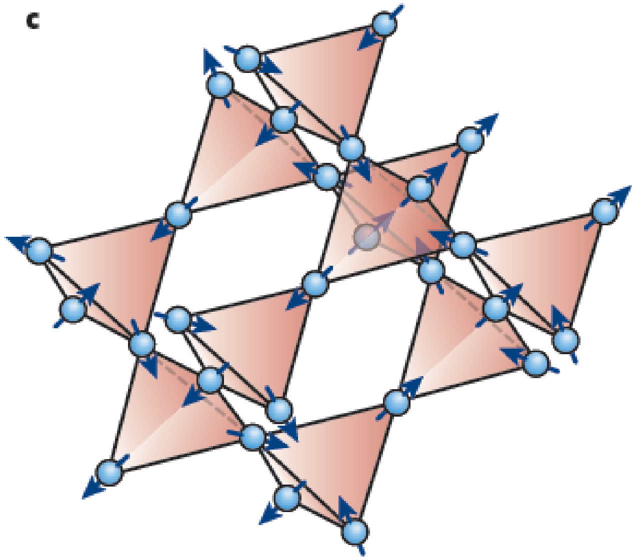
2D triangular and kagome lattices

Six-fold degeneracy
of a triangular
Ising placquette



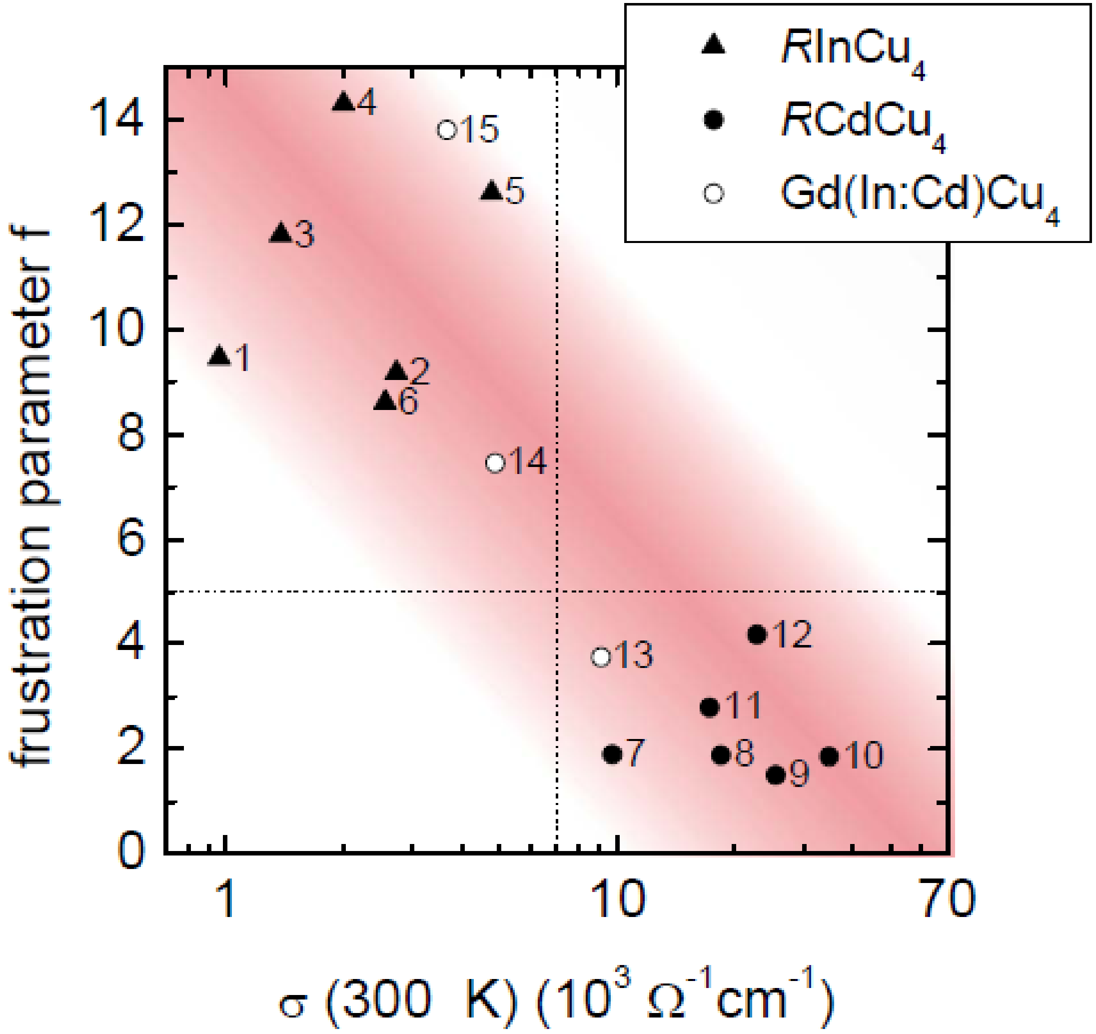


3D kagome lattice

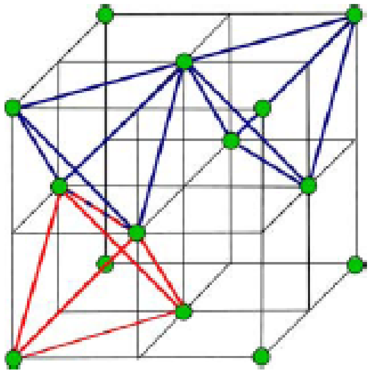


Large degree of degeneracy leads to low-lying fluctuations
and thus to suppression of magnetic order

Frustration and conductivity



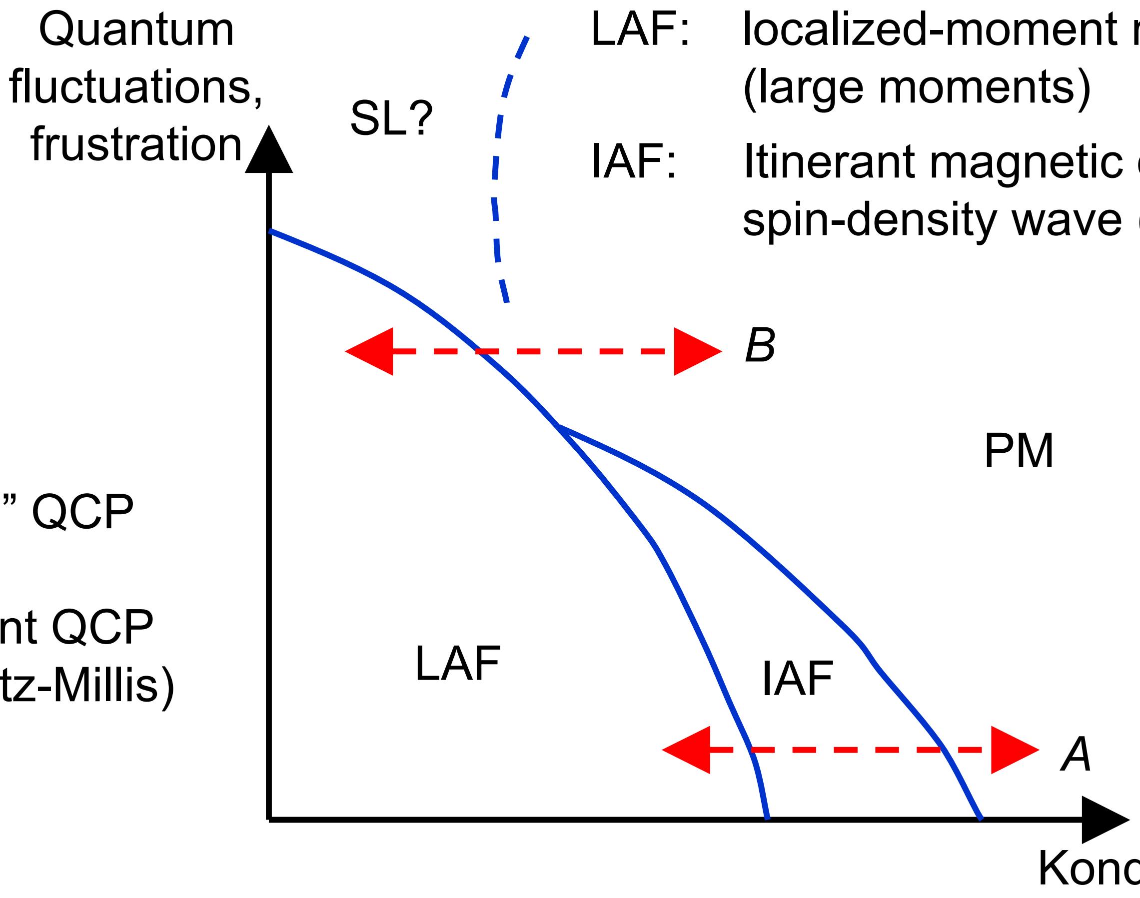
1: GdInCu$_4$
2: TbInCu$_4$
3: DyInCu$_4$
4: HoInCu$_4$
5: ErInCu$_4$
6: TmInCu$_4$
7: GdCdCu$_4$
8: TbCdCu$_4$
9: DyCdCu$_4$
10: HoCdCu$_4$
11: ErCdCu$_4$
12: TmCdCu$_4$
13: GdIn$_{0.25}$Cd$_{0.75}$Cu$_4$
14: GdIn$_{0.5}$Cd$_{0.5}$Cu$_4$
15: GdIn$_{0.75}$Cd$_{0.25}$Cu$_4$



Frustration Parameter f = Θ_{CW}/T_c

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

Possible additional phase line at *T* = 0: LMM vs. HMM



LAF: localized-moment magnetic order (large moments)

IAF: Itinerant magnetic order spin-density wave (small-moments)

Q. Si et al.
P. Coleman

B: “2D local” QCP

A: 3D itinerant QCP (Hertz-Millis)

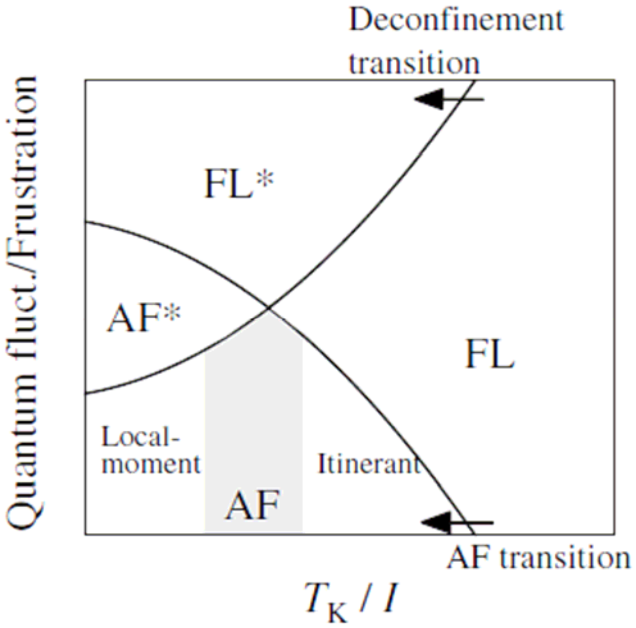
Internal consistency for $\mathrm{CeCu}_{6-x}\mathrm{Au}_x$

Lowering the effective dimensionality leads to an increase of quantum fluctuations, and thus to the local QCP. Magnetic field restores 3D and hence yields Hertz-Millis

cf. experiments on Co- and Ir-doped $\mathrm{YbRh_2Si_2}$ *S. Friedemann et al,, Nature Phys. 2009*

Possible continuous evolution from local-moment to itinerant antiferromagnetism in Kondo systems

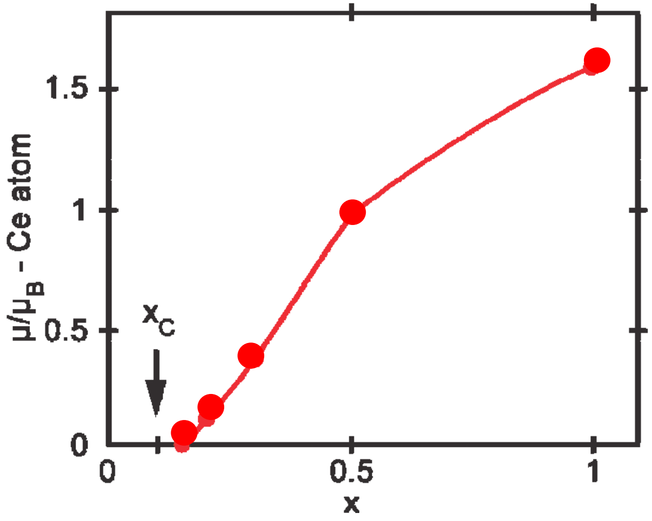
M. Vojta, PRB 78, 125109 (2008)
See also T. Senthil et al., PRL 90, 216403 (2003)



LAF - IAF transition may be gradual

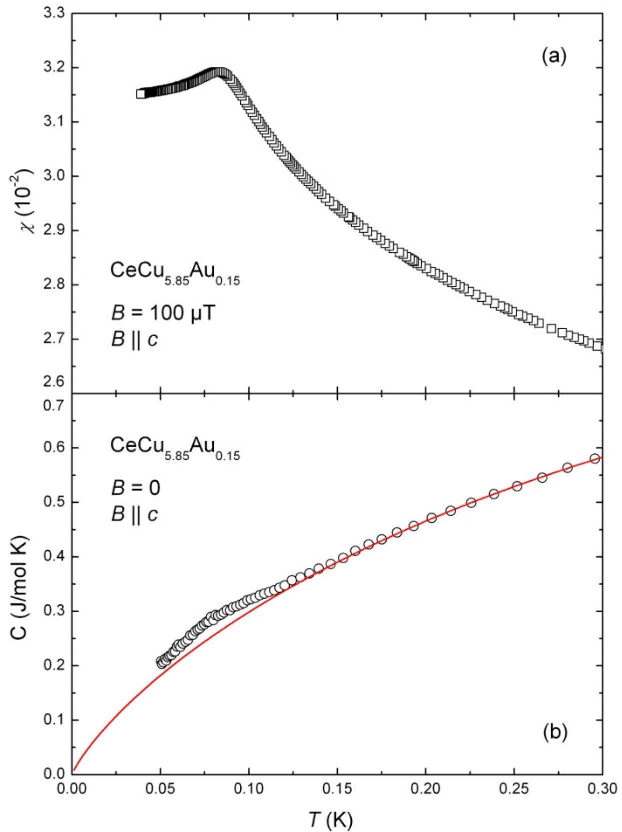
How to experimentally “control” the vertical axis?

What is the effect of magnetic field in this plot?

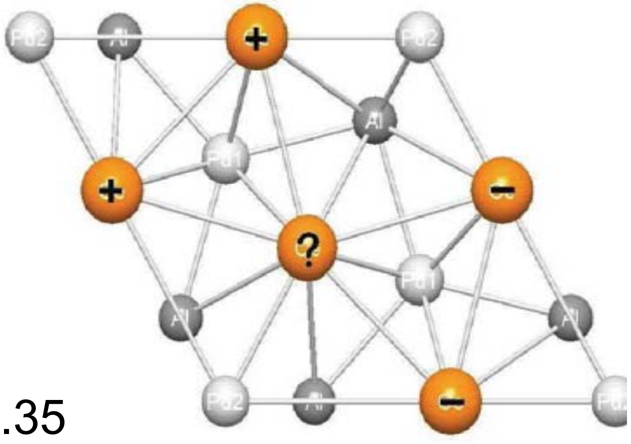


$CeCu_{1-x}Au_x$: gradual evolution of ordered magnetic moments (from ENS)

Tiny specific-heat anomaly at T_N on top of a large “non-Fermi-liquid” background



CePdAl – a partially frustrated Ce-based compound



Magnetic order
with T_N = 2.7.K

Q = (½ 0 τ), τ = 0.35

1/3 of the Ce moments frustrated

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

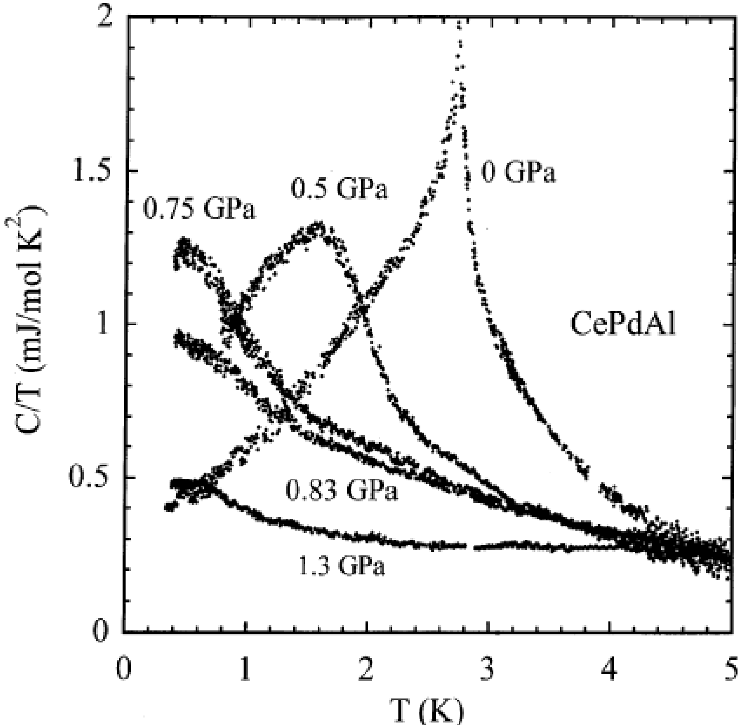
Suppression of T_N by hydrostatic pressure …

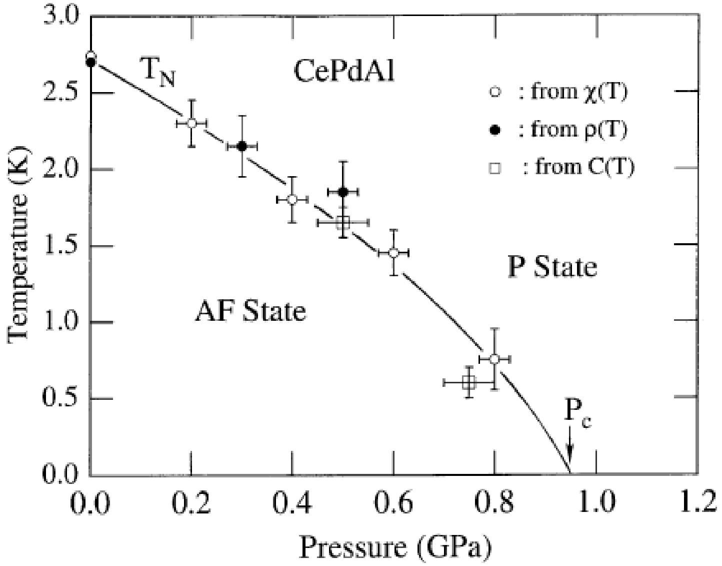
Goto et al., J. Phys. Chem: Sol. ***63****, 1159, (2001)*

or by isoelectronic Ni doping

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

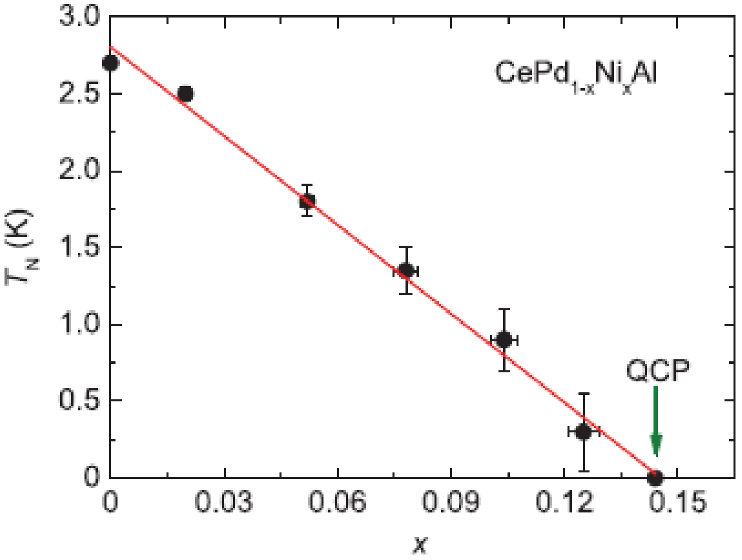
Quantum critical point?

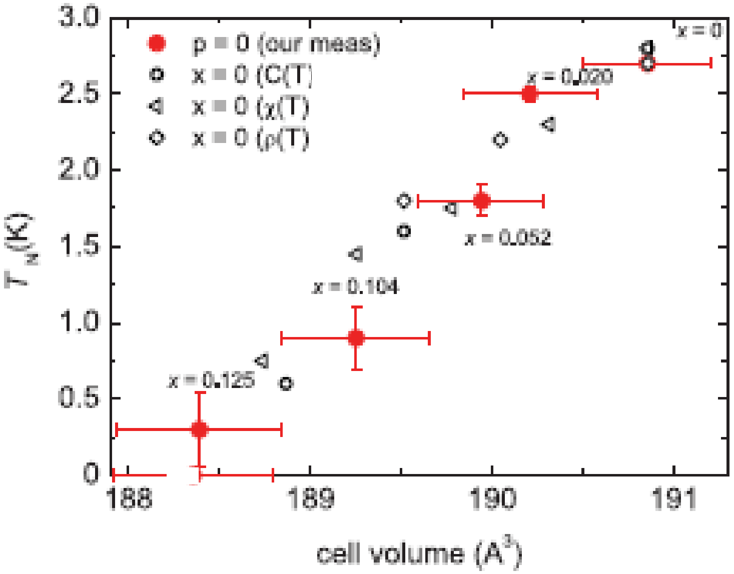




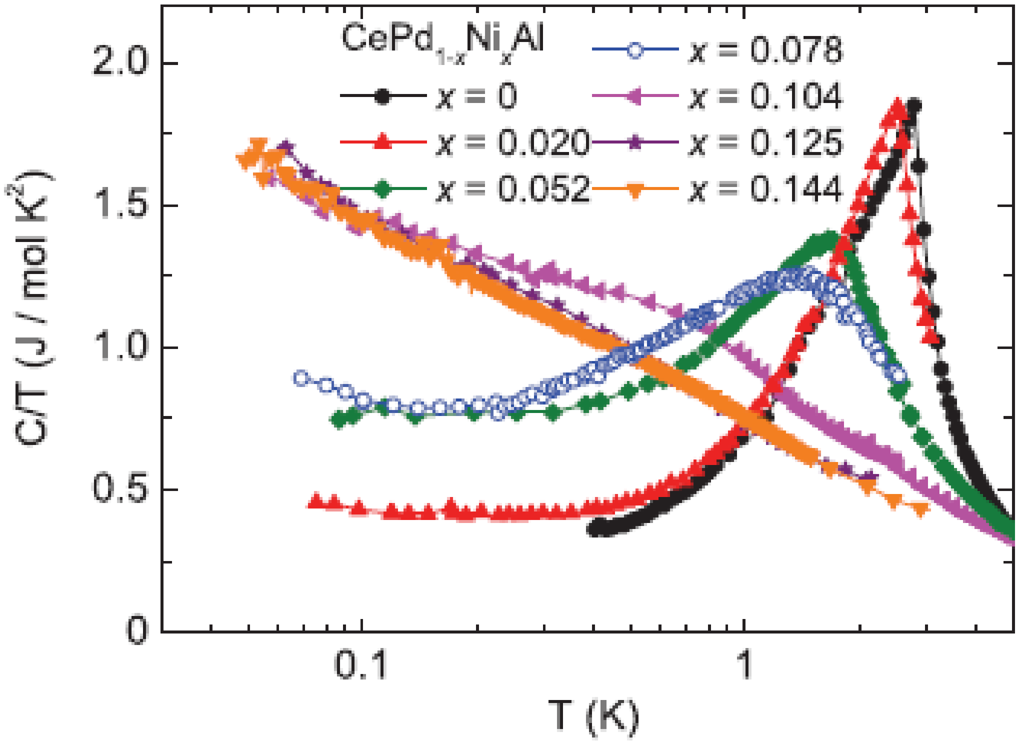
Suppression of magnetic order in CePdAl by Ni doping

Neél temperature





Specific heat



Equivalence of pressure and Ni doping when T_N is plotted against unit-cell volume

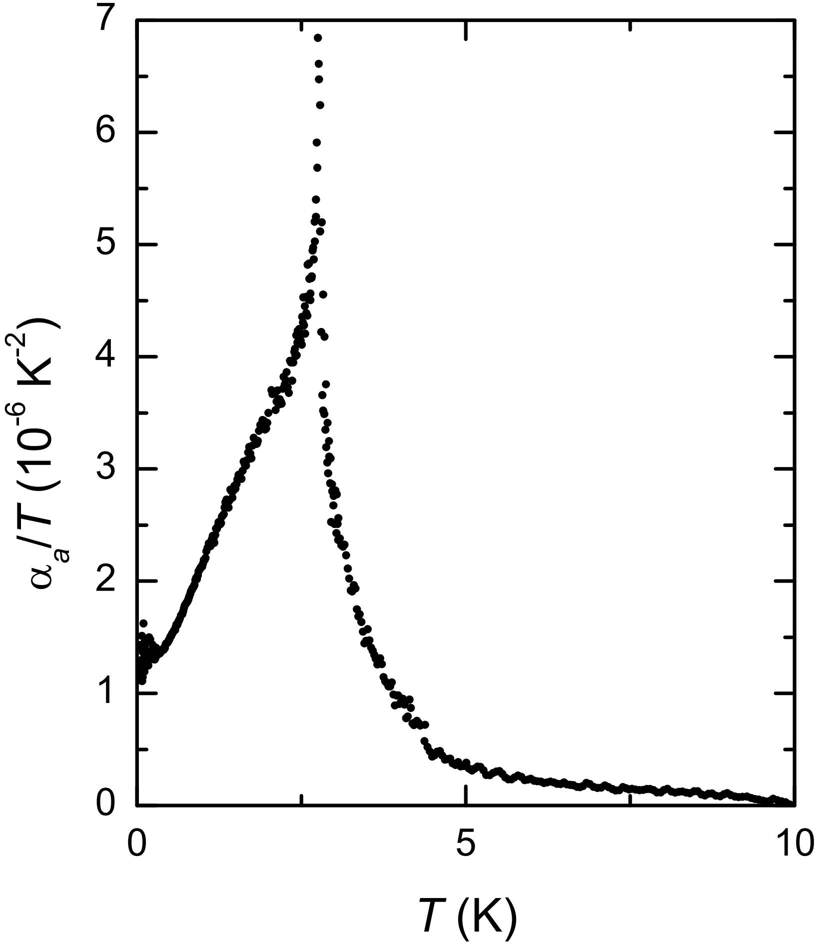
$T_N \rightarrow 0$ for $x \approx 0.14$: $C/T \sim -\log(T/T_0)$

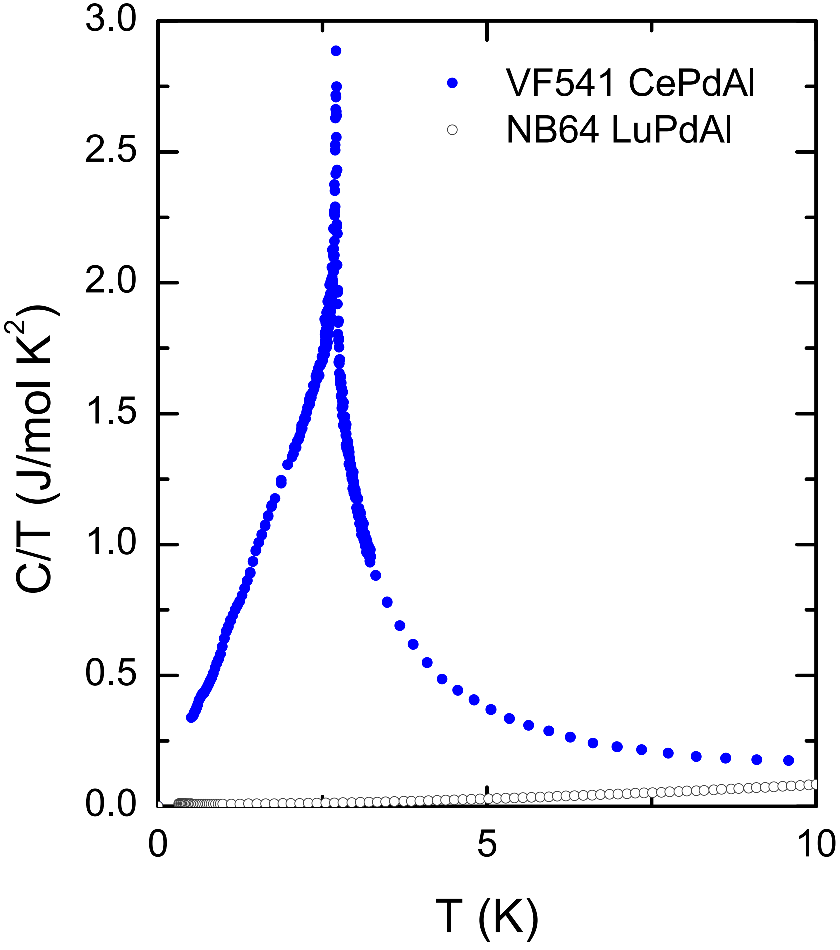
or for $p = 1 – 1.2$ GPa: $\rho(T) \sim \rho_0 + AT^n$

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

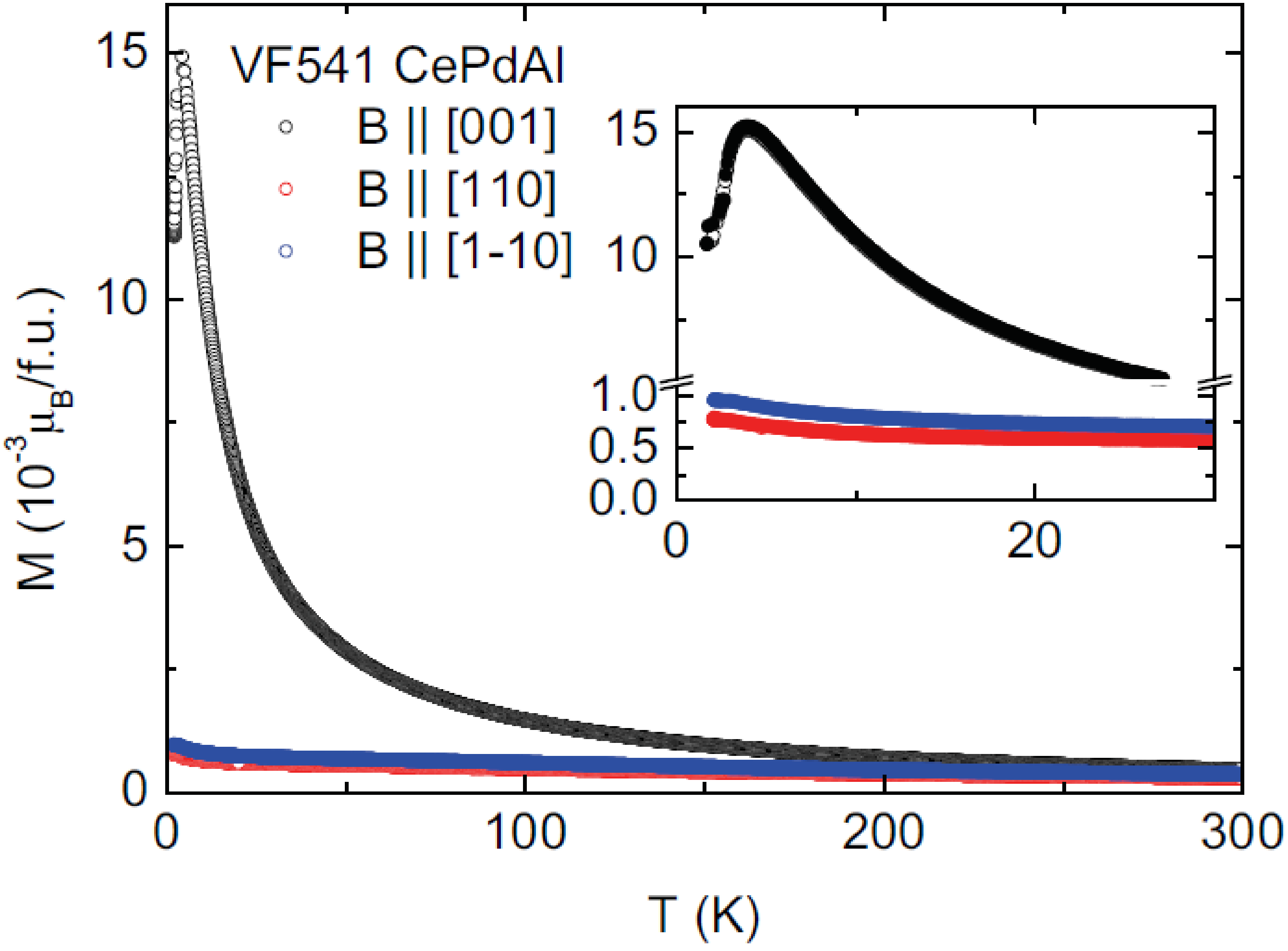
Two-dimensional criticality or novel QCP?

Specific heat and thermal expansion of single-crystalline CePdAl





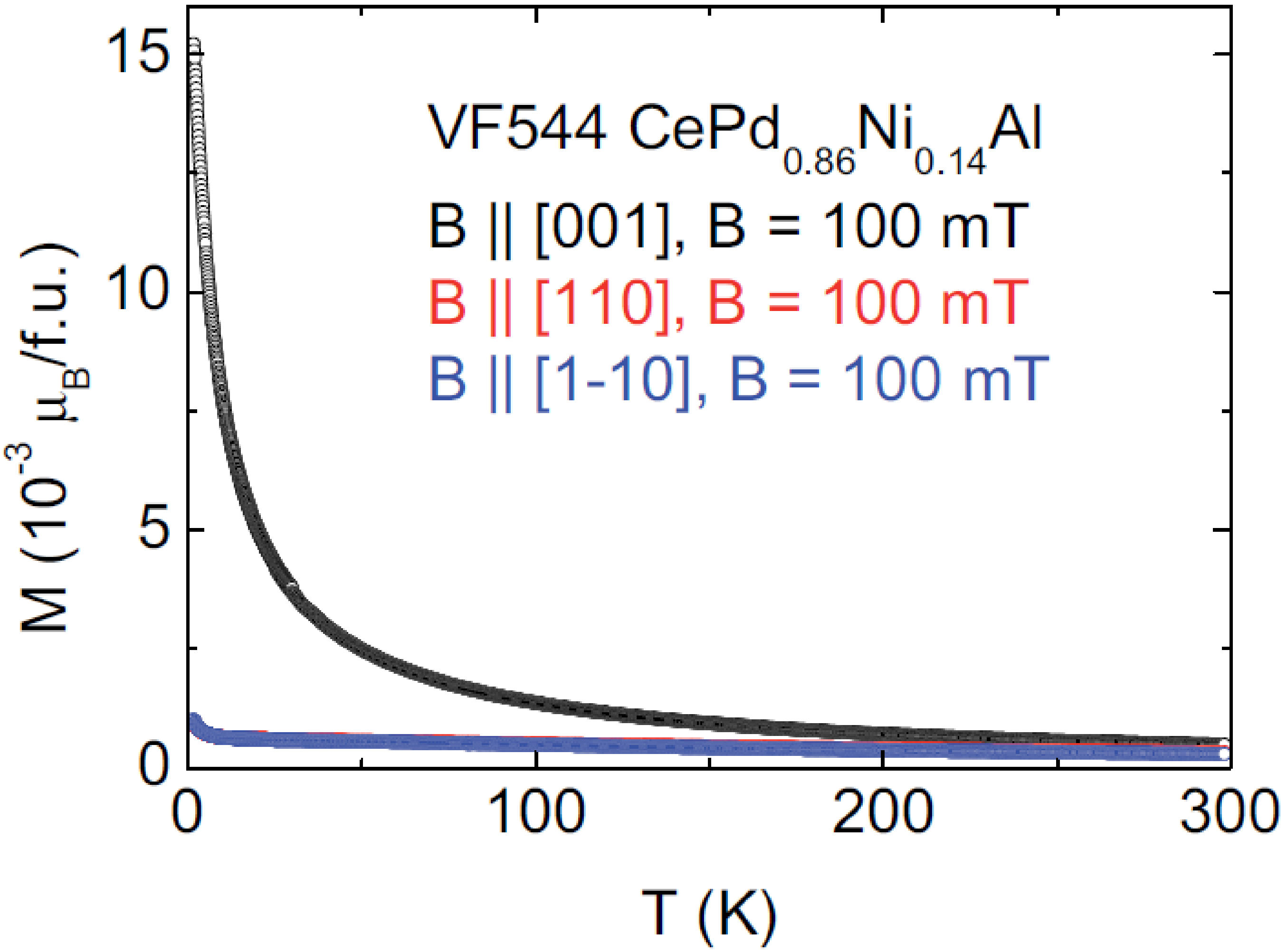
Magnetic susceptibility of a CePdAl single crystal



Note: strong Ising-like anisotropy due to single-ion crystal-field effects

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

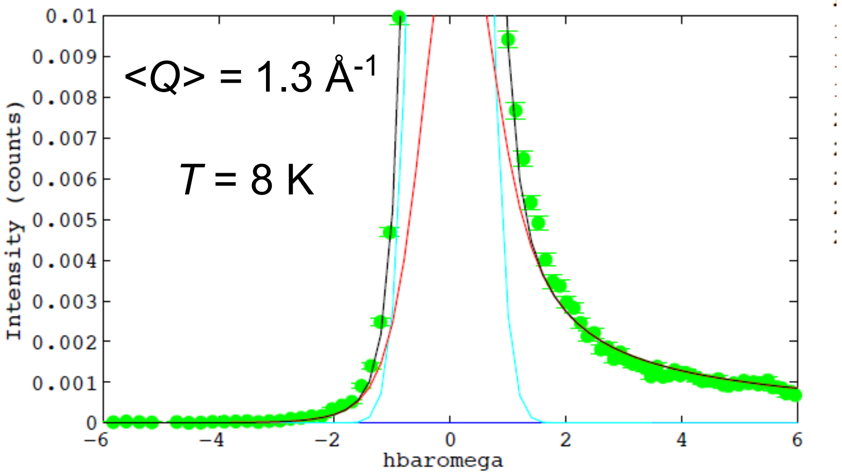
Magnetic susceptibility of $CePd_{1-x}Ni_xAl$ with $x = 0.14$



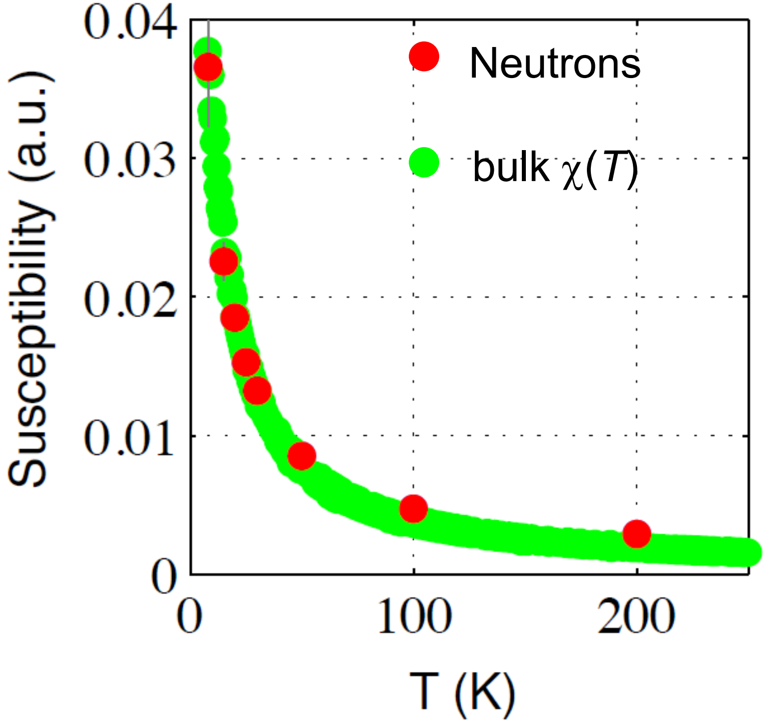
Strong anisotropy remains upon approaching the quantum critical point

Dynamic susceptibility of CePdAl from quasielastic neutron scattering

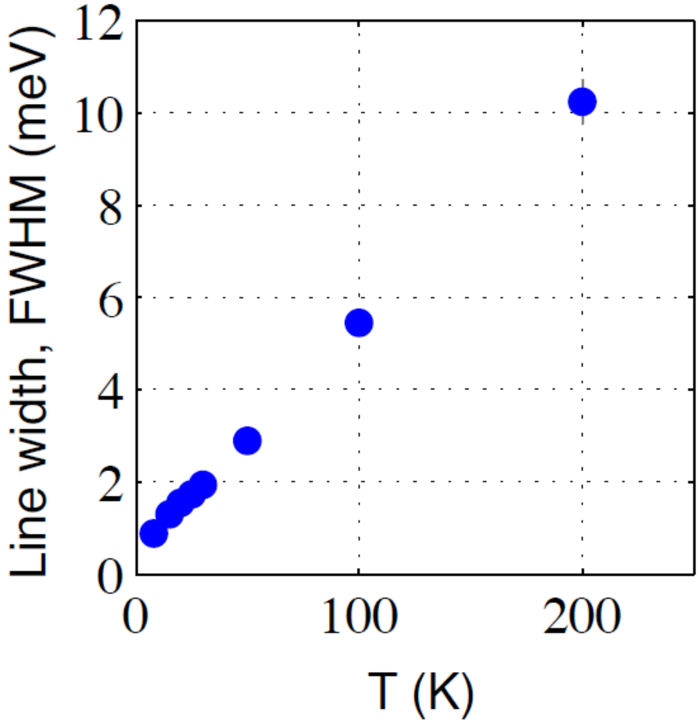
IN4 ILL (TOF)
powder measurements



Integrated scattering intensity
scales with bulk susceptibility

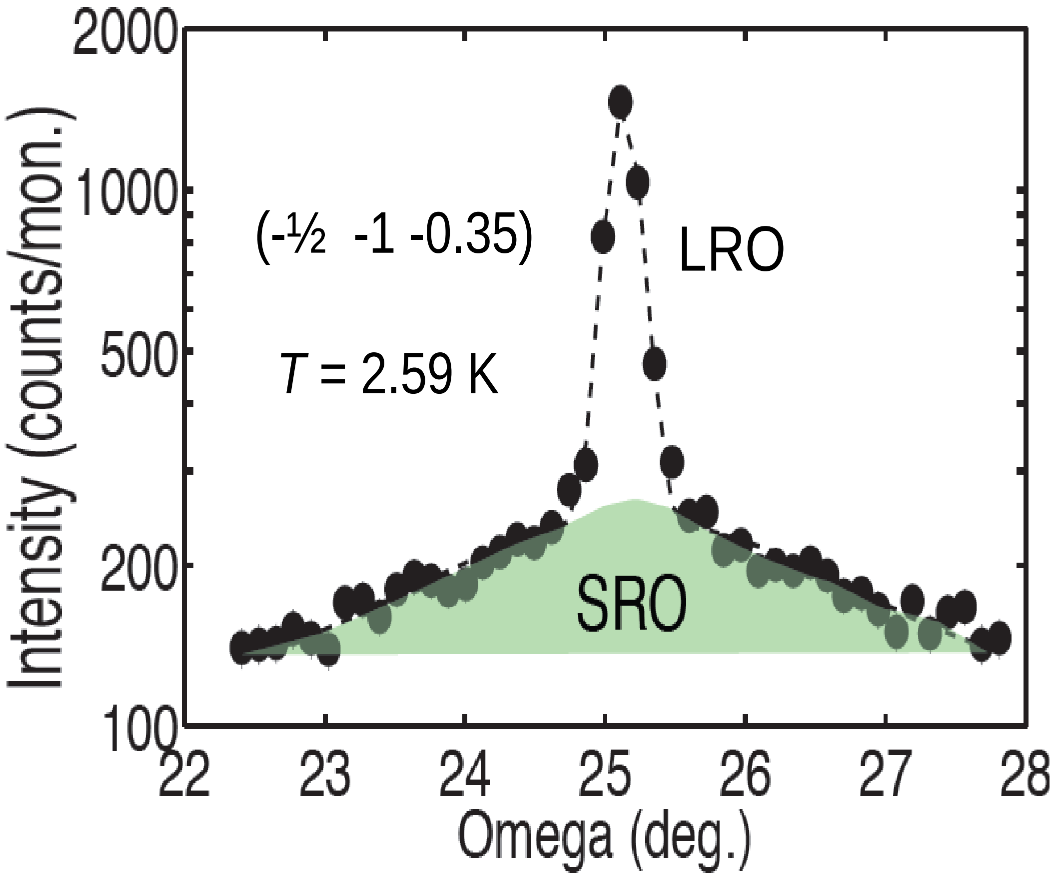


Residual linewidth: Kondo effect
Unusual dependence $\Gamma(T) \sim T$

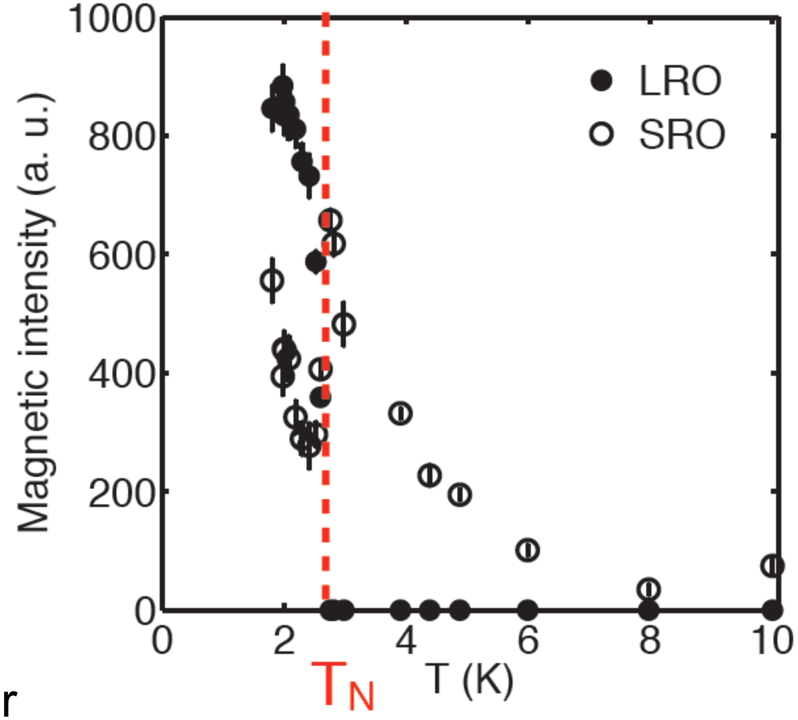


Magnetic order in CePdAl

D20 ILL

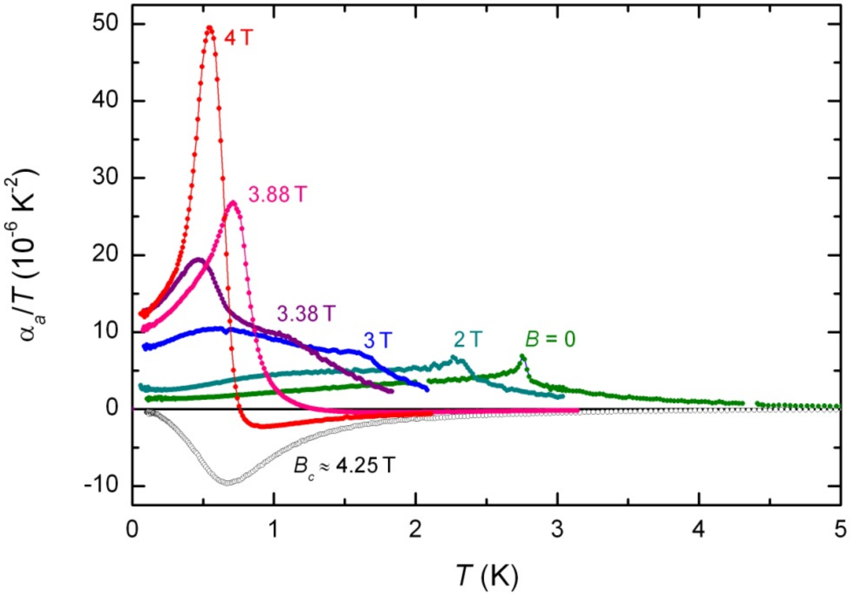


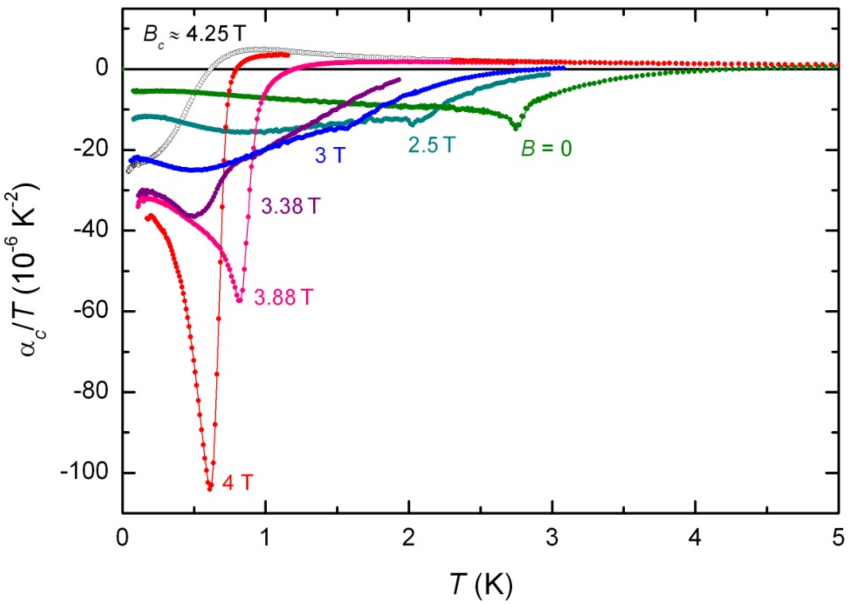
Observation of both
long-range *and*
short-range order below T_N:

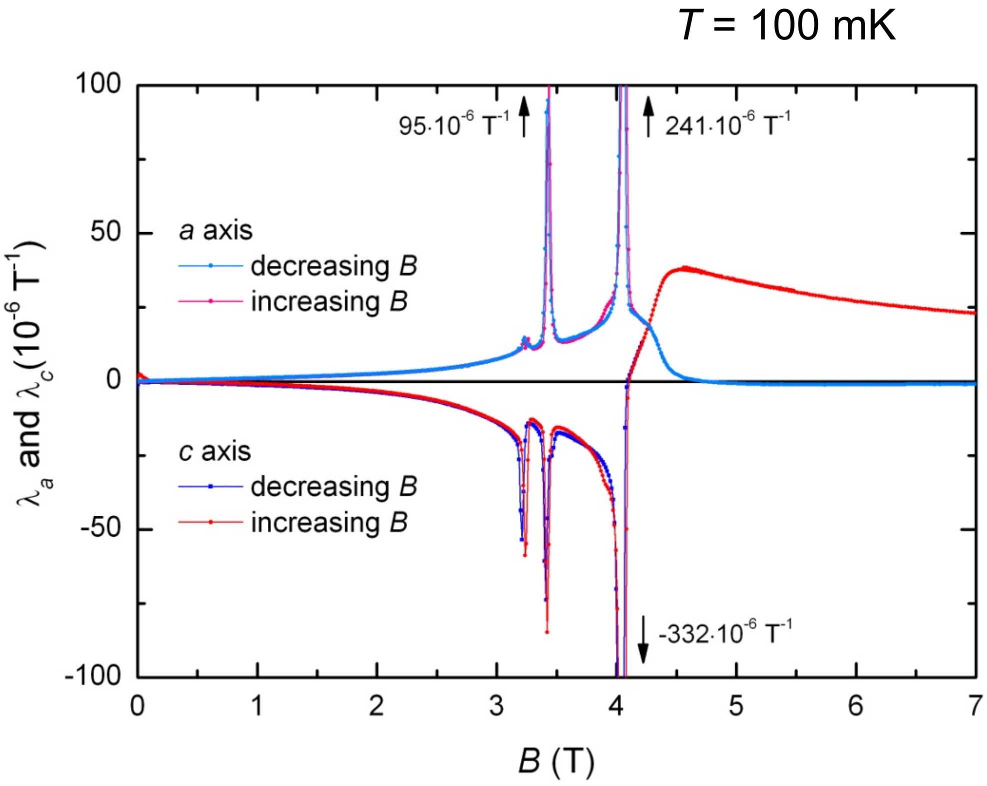


LRO/SRO intensity ratio of 2/1
Is compatible with the assumption that
the frustrated moments believed not to order
are in fact short-range ordered!

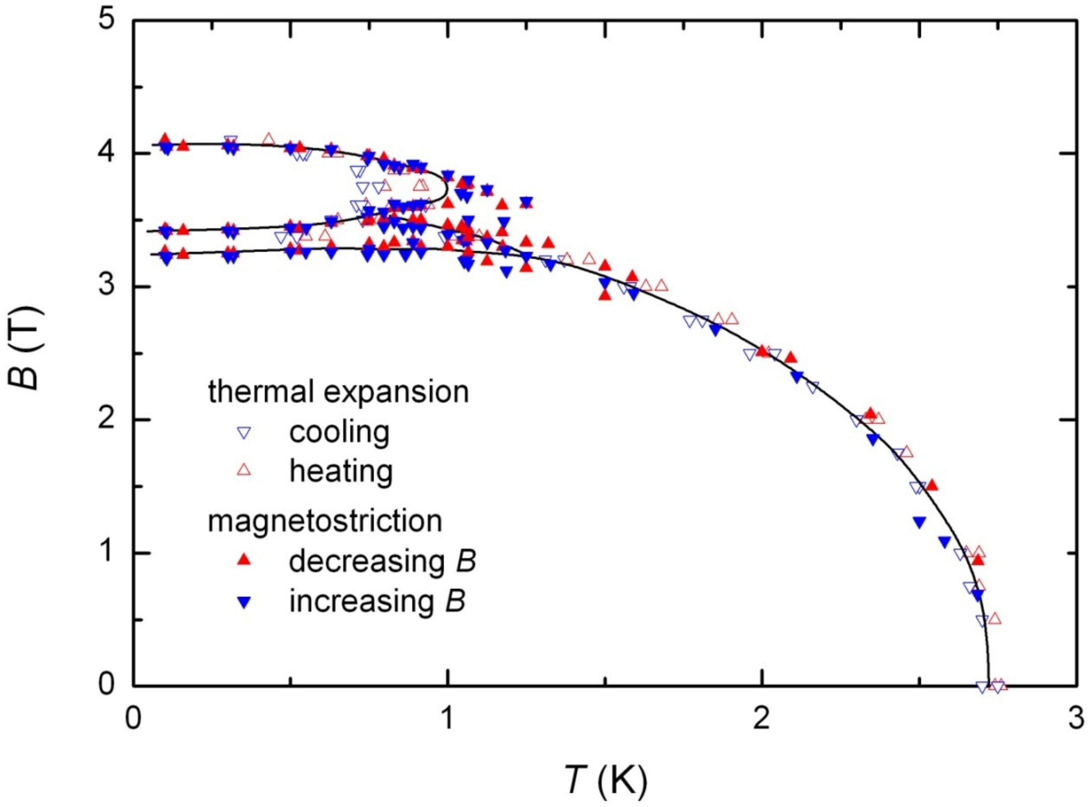
Thermal expansion and magnetostriction of CePdAl





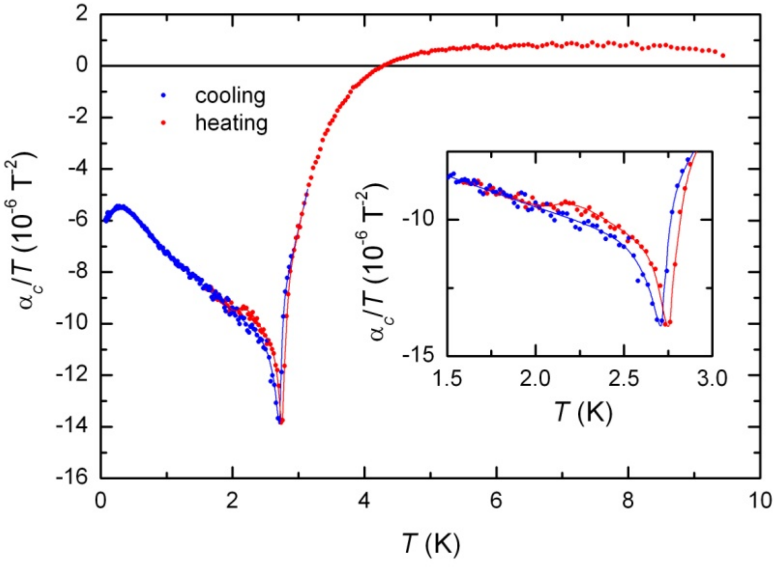


Magnetic phase diagram of CePdAl from thermal expansion and magnetostriction

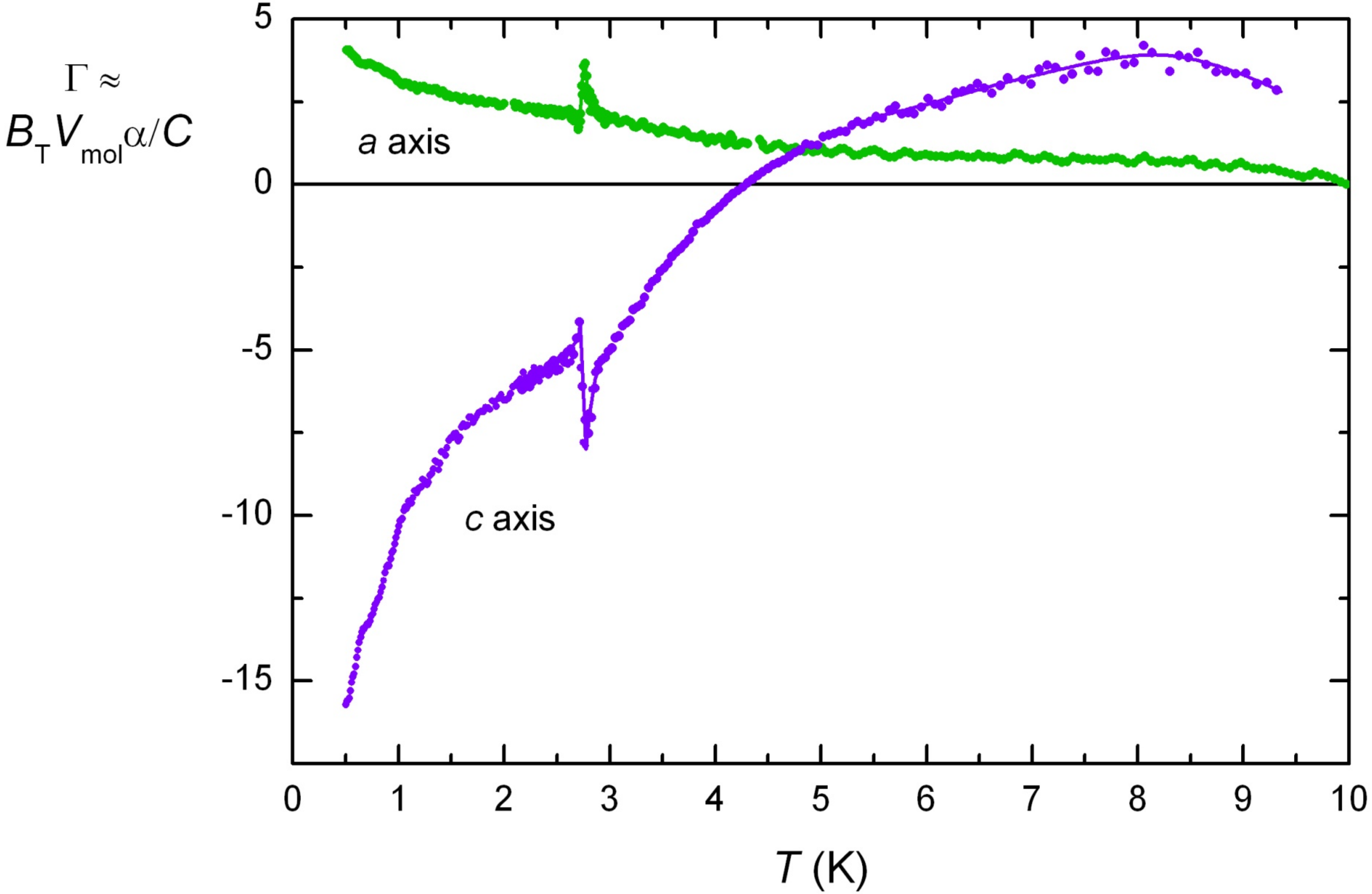


Complicated sequence of phases close to B_c

Note: transition in zero field might be first order: weak hysteresis

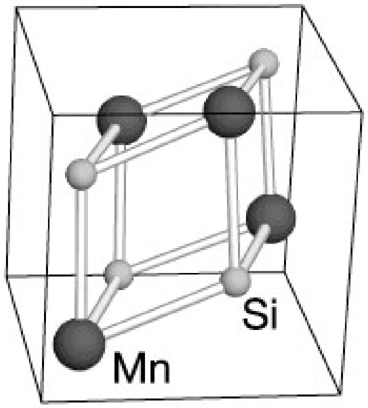


Strong *T* dependence of the Grüneisen parameter for $T \to 0$



Partial order in MnSi

The weak itinerant ferromagnet MnSi



Representative of weak itinerant magnets:

$ZrZn_2$, Sc_3In, Ni_3Al, YNi_3, CoS_2, ...

cubic, B20 structure, no inversion symmetry

ferromagnetic: $T_c = 29.5$ K, $\mu = 0.4\ \mu_B$

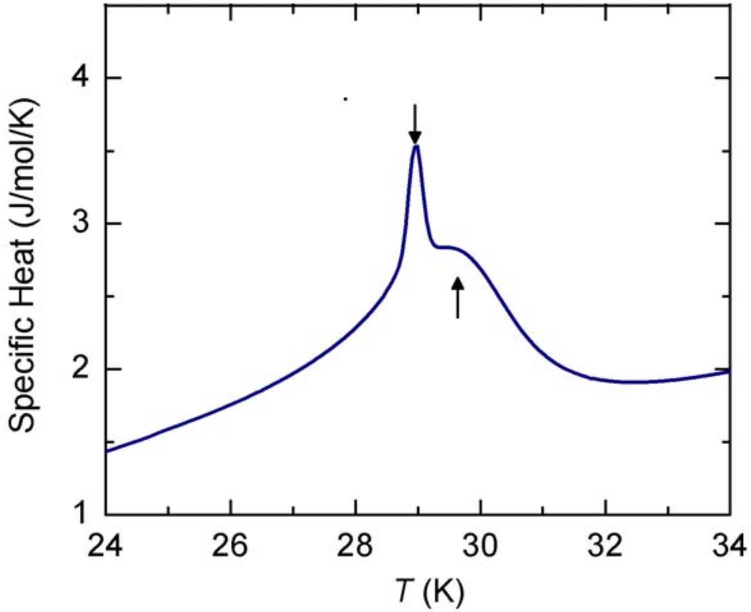
Specific heat: small entropie change at T_c

at low T: $\gamma \approx 38$ mJ/mol K^2

Spin-orbit coupling leads to a helical twist of the magnetization

$\lambda = 175$ Å

Magnetic superlattice reflections along <111> close to Bragg peaks



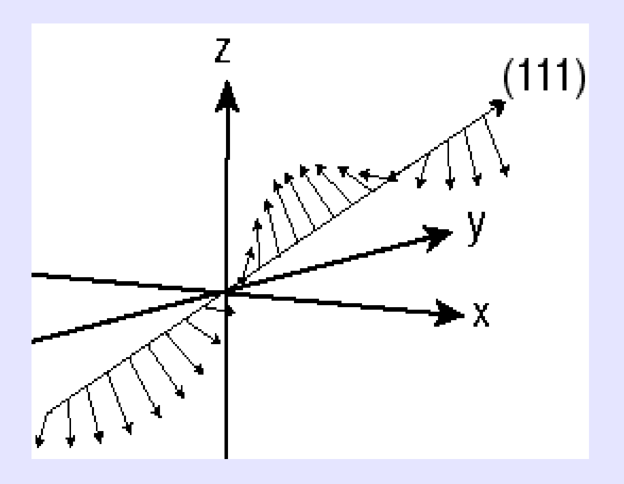
Characteristic energy scales in MnSi

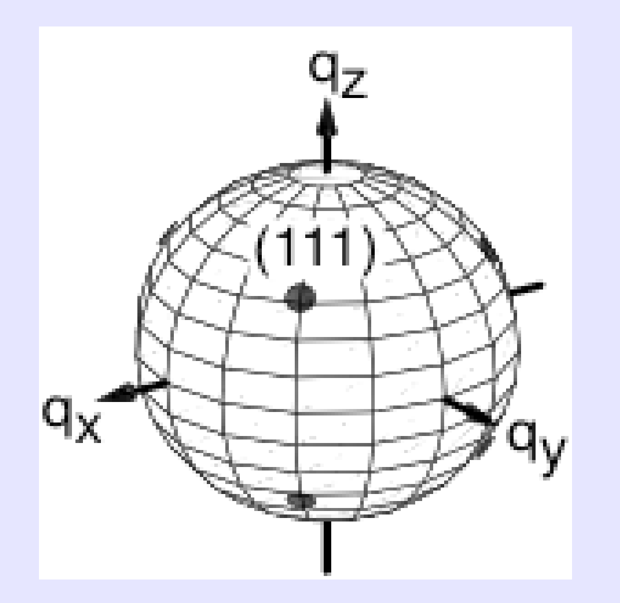
- ferromagnetic exchange

- spin-orbit coupling:
  Dzyaloshinskii-Moriya interaction
  $\mathbf{s} \cdot (\nabla \times \mathbf{s})$
  leads to long-wavelength spiral structure
  $\lambda \approx 175$ Å (cf. a = 4.558 Å)

- crystal field potential ($P2_13$):
  helix locked at <111> or <100>, not <110>

  ⇒ sharp satellite reflections
  at <111> positions
  around nuclear Bragg peaks



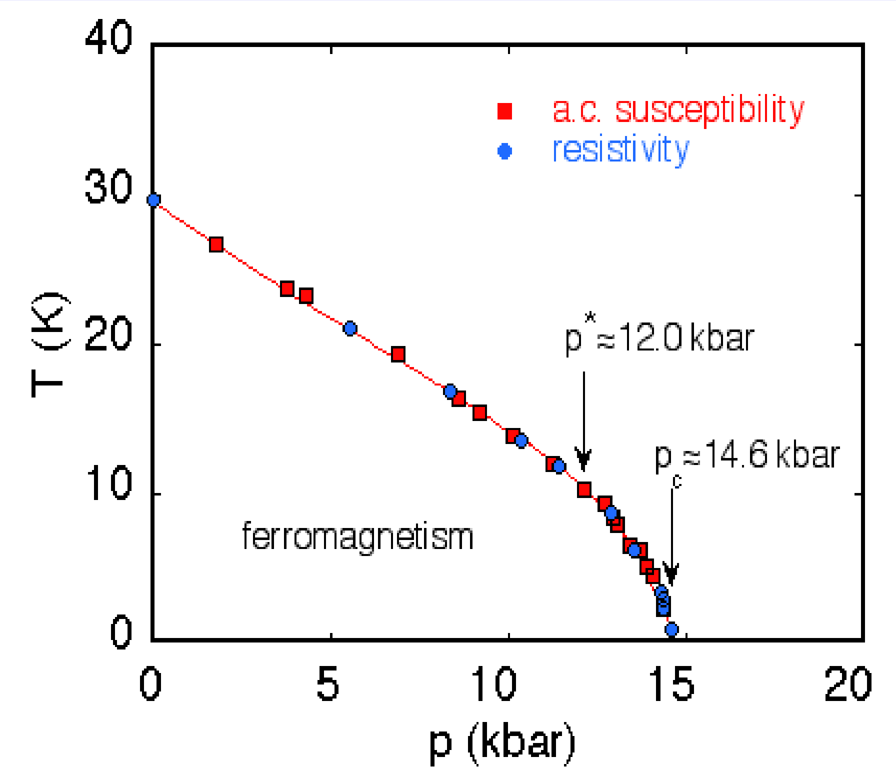


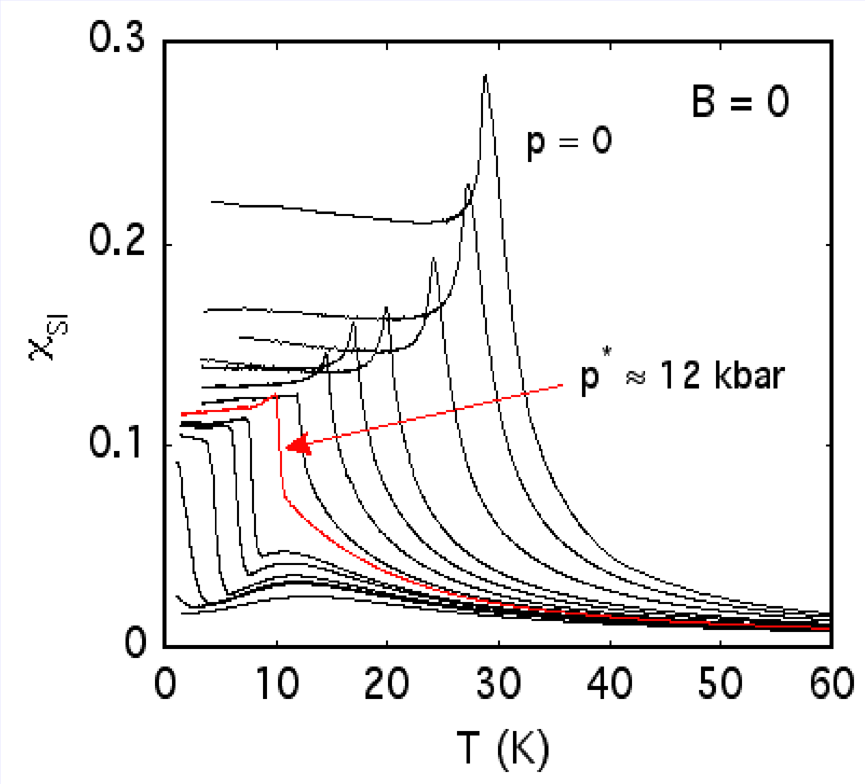
Phase diagram of MnSi under pressure

C. Pfleiderer et al. 1997, 2003

Pressure dependence of the Curie temperature

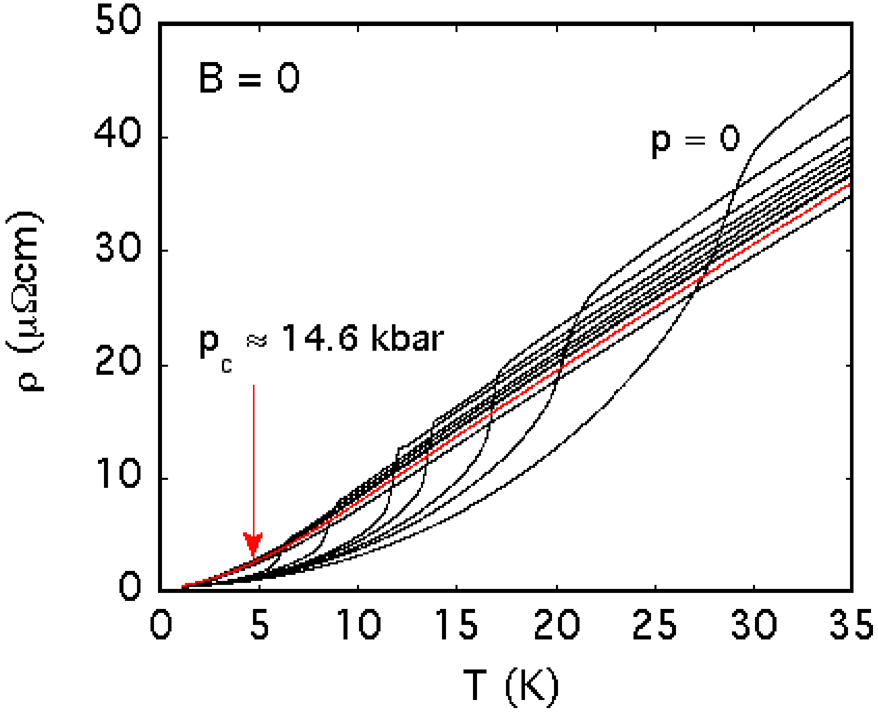
Magnetic susceptibility under pressure

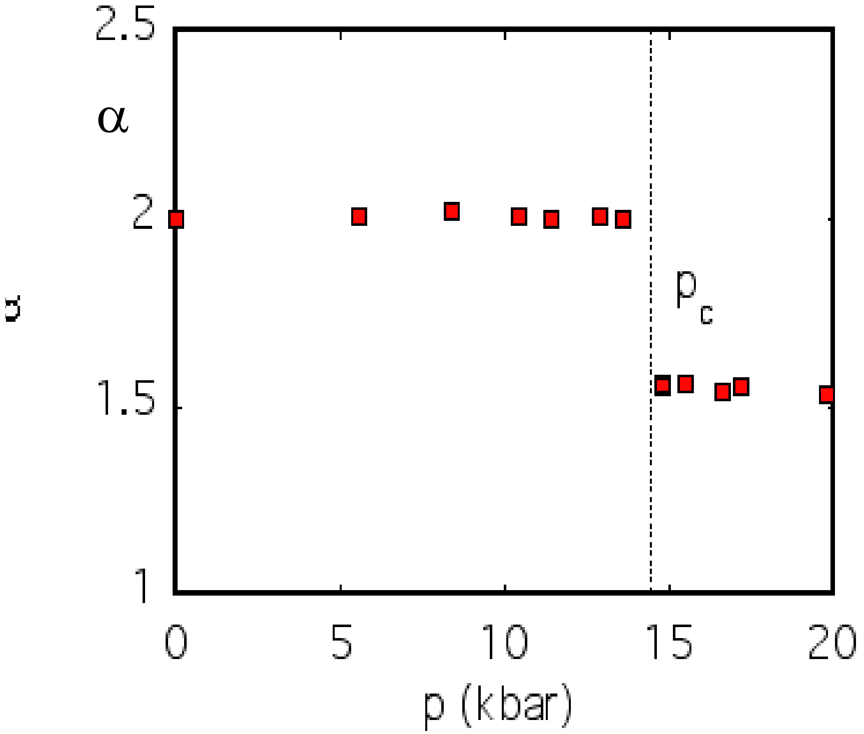




Electrical resistivity of MnSi under high pressure

C. Pfleiderer et al. 1997, 2001; N. Doiron-Leyaud et al. 2003





Fermi-liquid *T* dependence

$\rho(T) = \rho_0 + AT^{\alpha}$, $\alpha = 2$

observed for $p < p_c$, $T < T_c$ only

Non-Fermi-liquid behavior

$\alpha = 3/2$

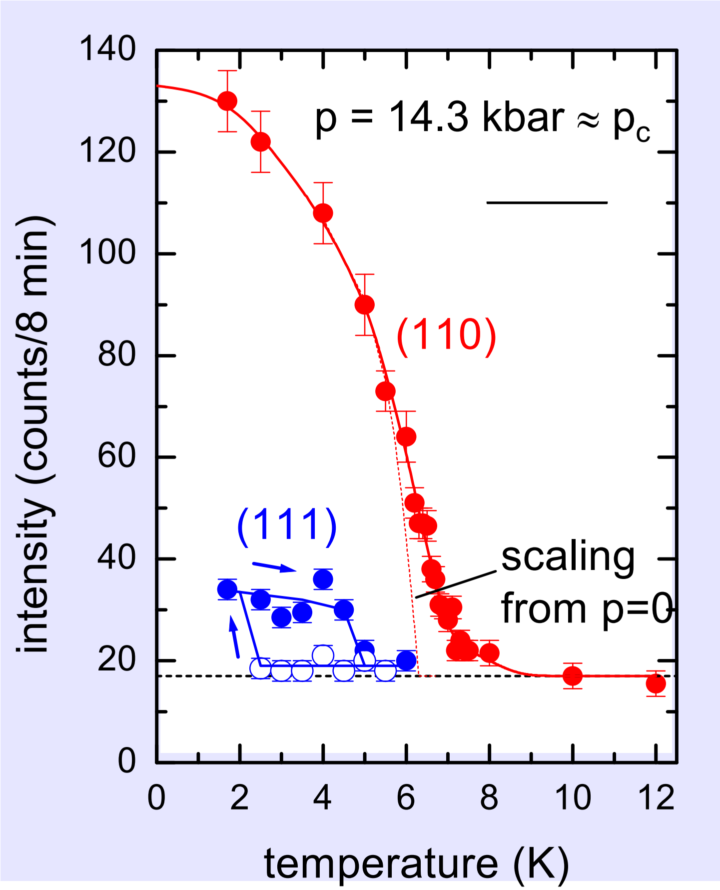
for $p > p_c$ over large p, T range

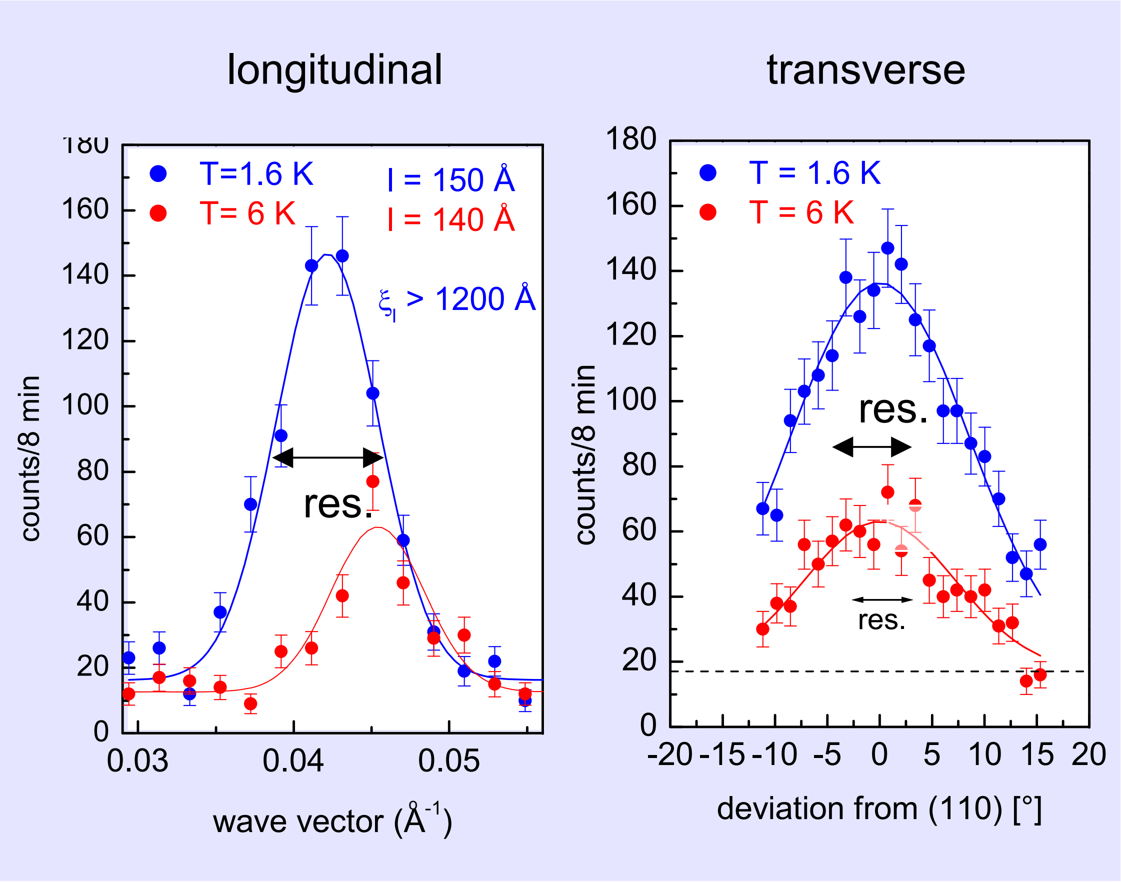
Elastic neutron scattering at $p \approx p_c$

C. Pfleiderer et al., Nature 2004

Temperature dependence

Q scans near the satellite reflections





strong intensity shift
from (111) to (110)

long-range order *along* the
helical direction (resolution limited)

wide *angular* distribution
of helical direction

Strange magnetic state of partial order

Observed around and even above p_c, with sluggish onset

Order remains helical with little change of periodicity and total intensity, compared to $p = 0$

Long-range (> 2000 Å) order along propagation direction of the helix

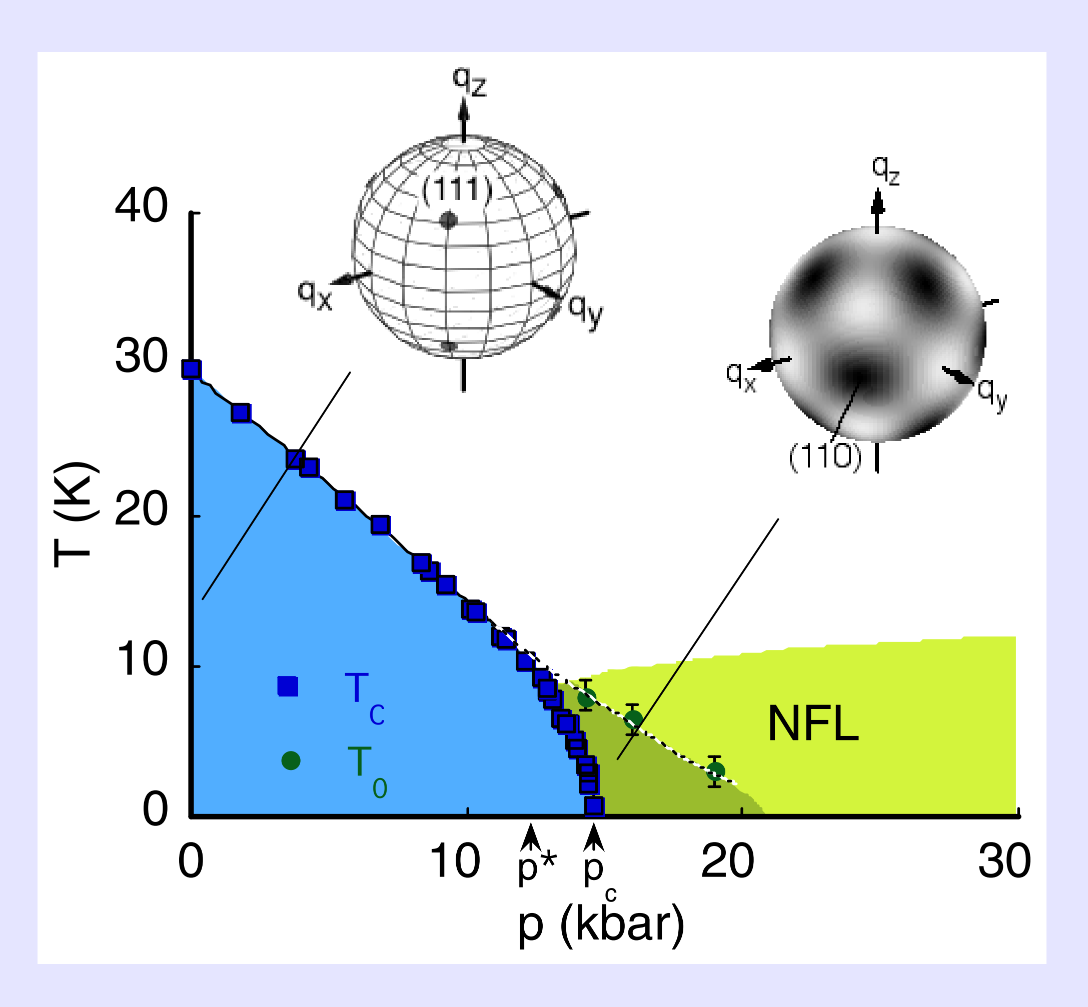
Propagation directions are distributed over a very wide angular range: “partial order“, analogous to partial order in certain types of liquid crystals

µSR: partial order is dynamic

T. Uemura, Nature Phys. 2007

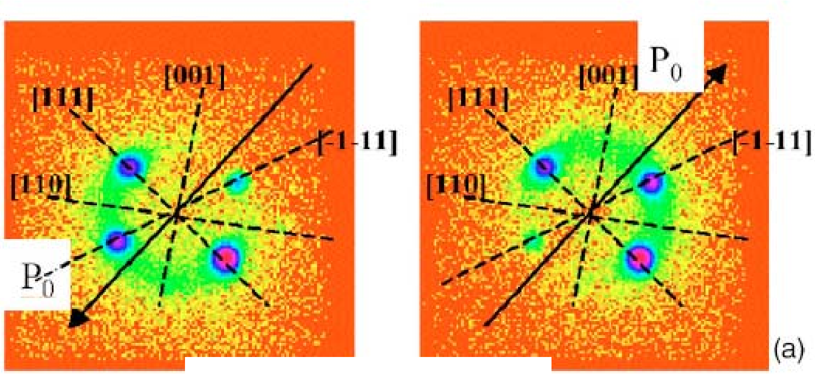
Partial order also seen in NMR experiments

W. Yu et al. PRL 2004

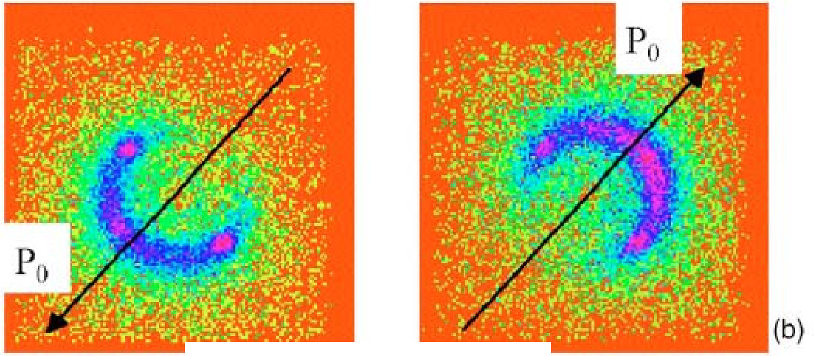


Fate of the helical order above T_c at $p = 0$

S. V. Grigoriev, P. Böni et al. 2005

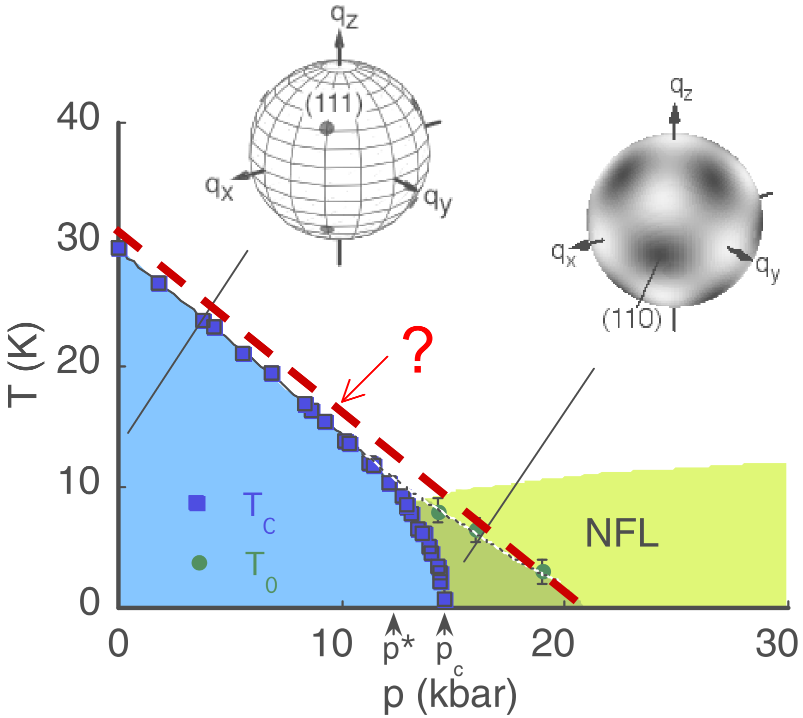


T = T_c – 0.1 K



T = T_c + 0.2 K

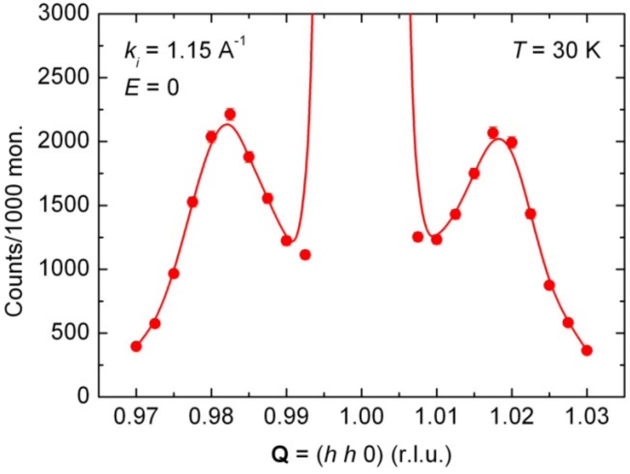
Small-angle polarized neutron scattering at FRG-1 Geesthacht



Existence of orientationally disordered helical structure above T_c

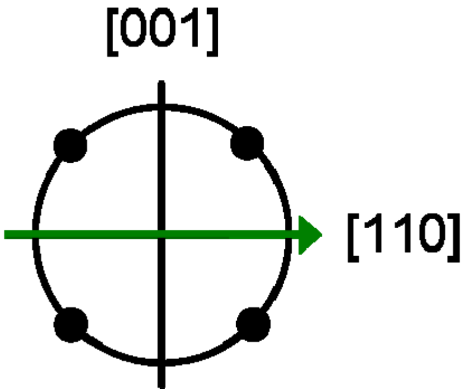
Relation to partial order for $T_c \to 0$?

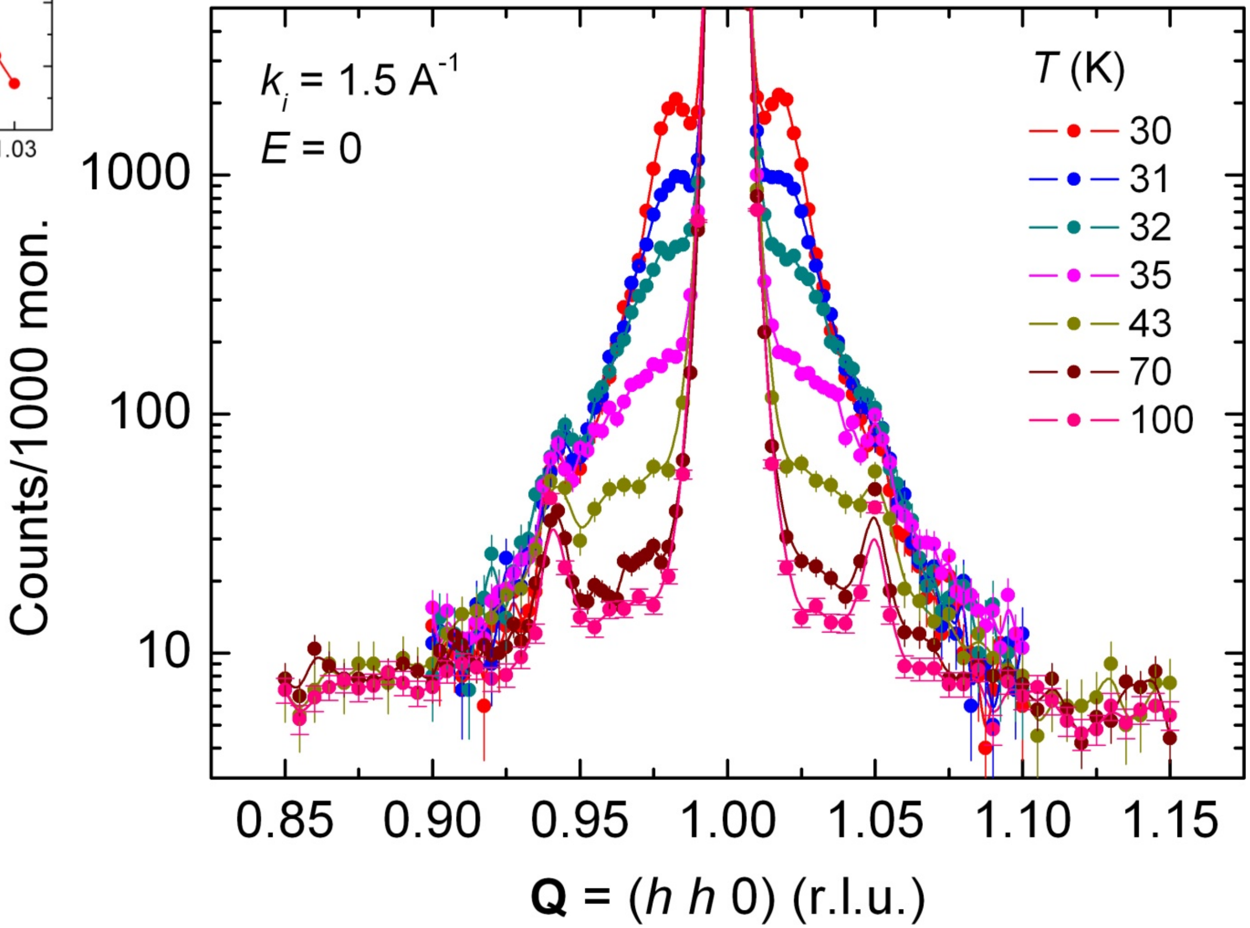
q-dependent elastic scatteringin MnSi above T_c



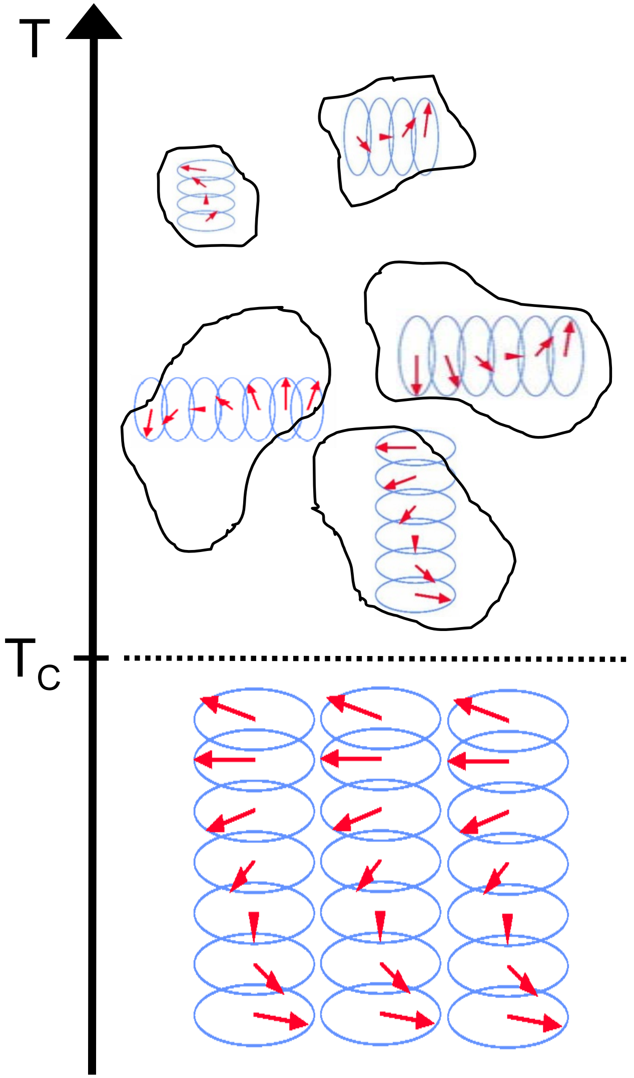
*A. Hamann et al., PRL **107**, 037207 (2011)*

Elastic scans through surface of sphere of superlattice reflections



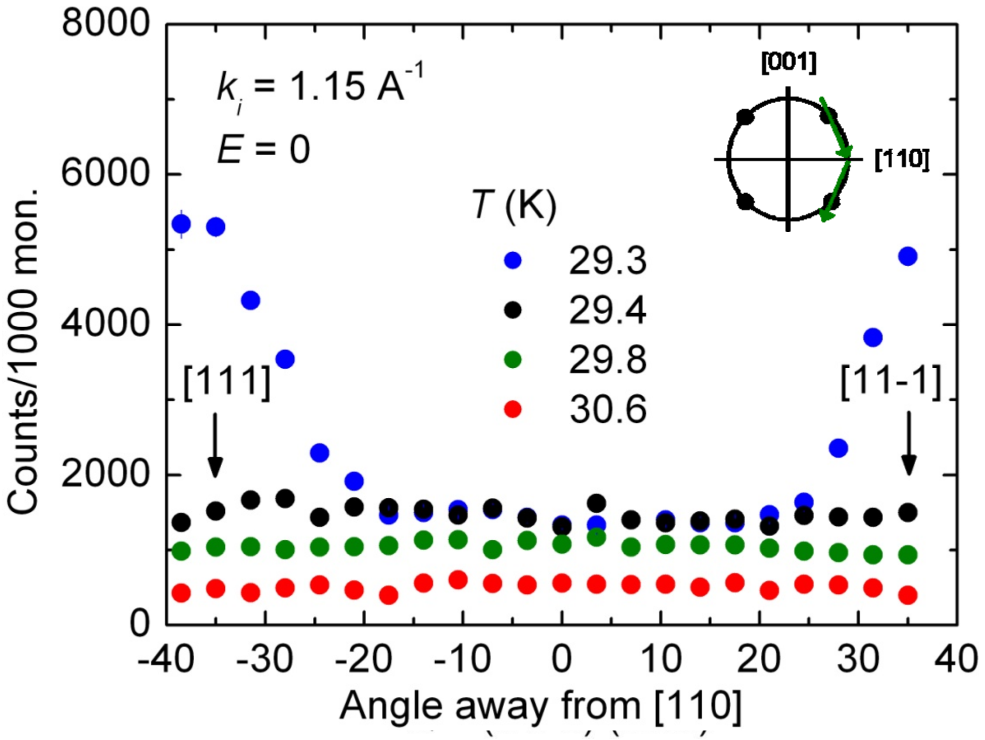


Spin topology above *T*$_C$

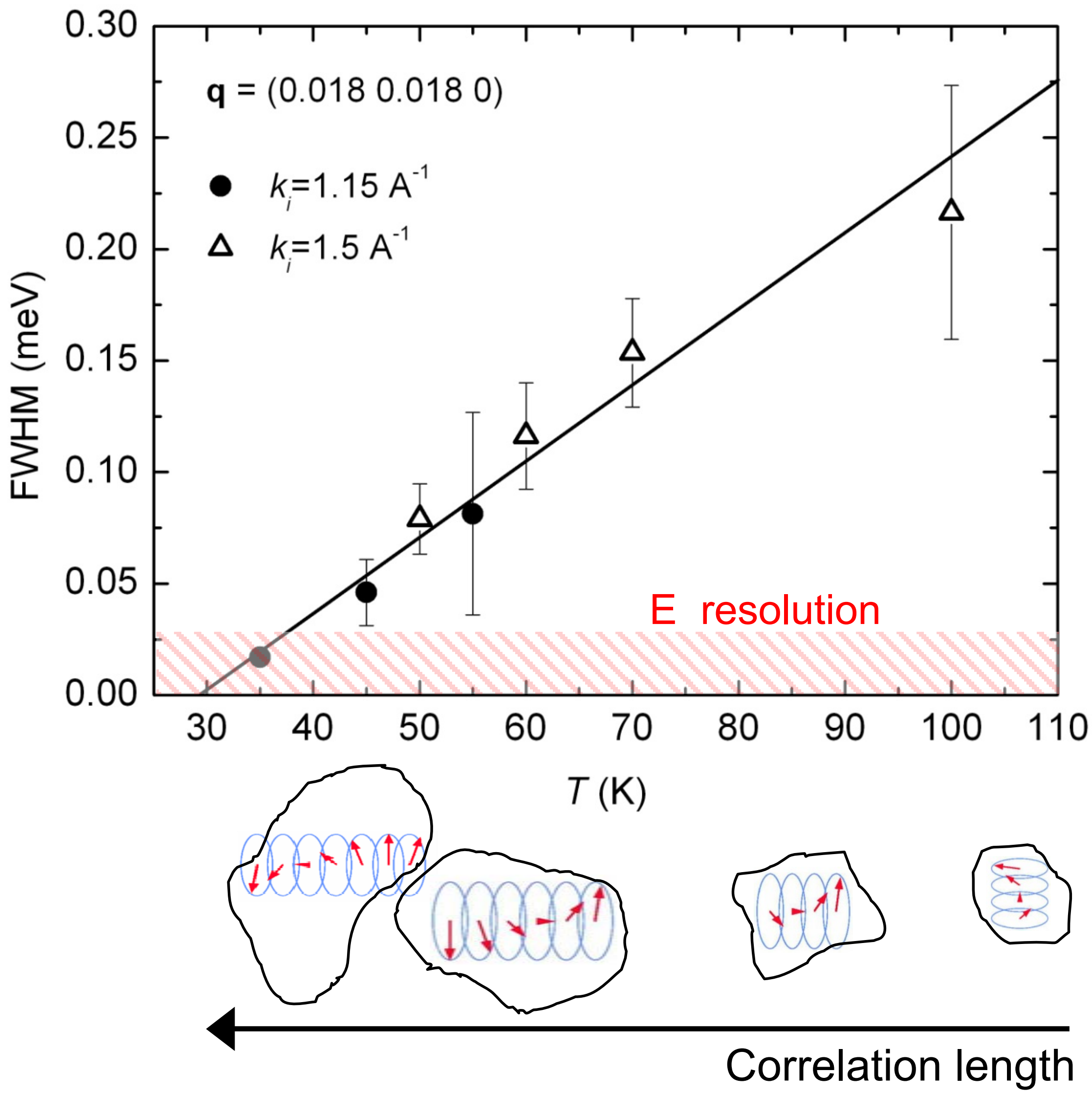


Local helical correlations

- Pitch conserved from low-*T* phase
- Correlation length decreases with higher *T*
- Helices point in any direction



Intrinsic energy width above T_C



Smaller number and size of the clusters towards increasing *T*

⬇

Faster diffusive motion, i.e., shorter correlation time

$$\mathrm{FWHM} \sim \tau^{-1} \sim T$$

“simple“ *T* dependence

Spin-cluster calculations

$$\mathcal{H} = -\frac{1}{2N}\sum_{i=1}^{N}\left(\sum_{j(i)}\left(J\mathbf{s}_i\cdot\mathbf{s}_j + \mathbf{D}_{i,j}\cdot(\mathbf{s}_i\times\mathbf{s}_j)\right)\right)$$ + crystal potential

one order of magnitude smaller

with $|\mathbf{D}_{i,j}| = D$ and $\mathbf{D}_{i,j} = -\mathbf{D}_{j,i}$.

Helix is unlikely to be the ground state for 2D and 3D:

Moments in planes ⊥ [111] are frustrated with respect to DM interaction

Model assumptions:

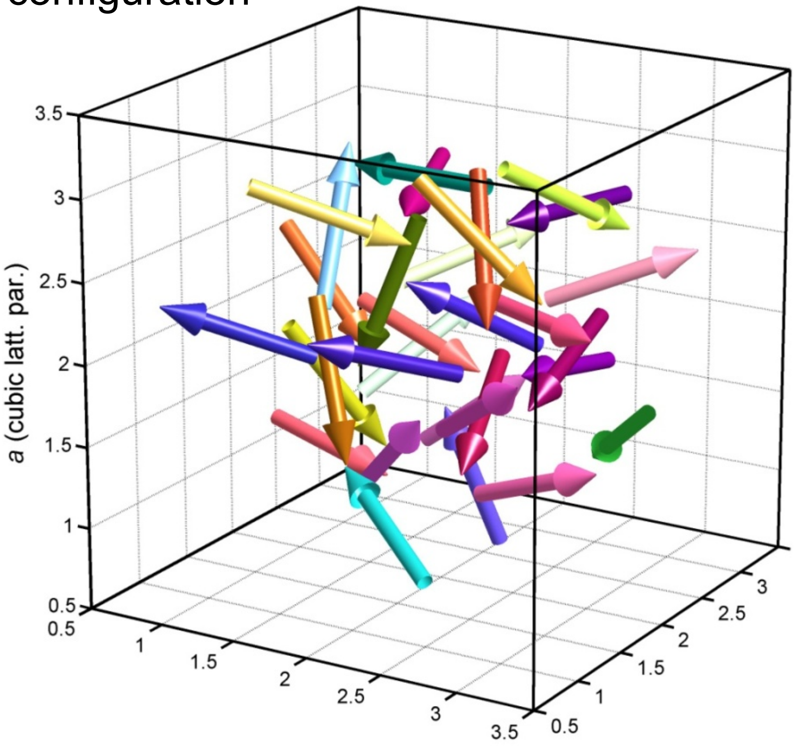
(1) Spins localized at Mn sites in the B20 MnSi structure, neglecting Si atoms

(2) Spins interact with their 6 nearest neighbors only

(3) Orientation optimization performed for individual spins one-by-one in random order (fixed magnitude)

(4) Different *D*/*J*

Random starting configuration

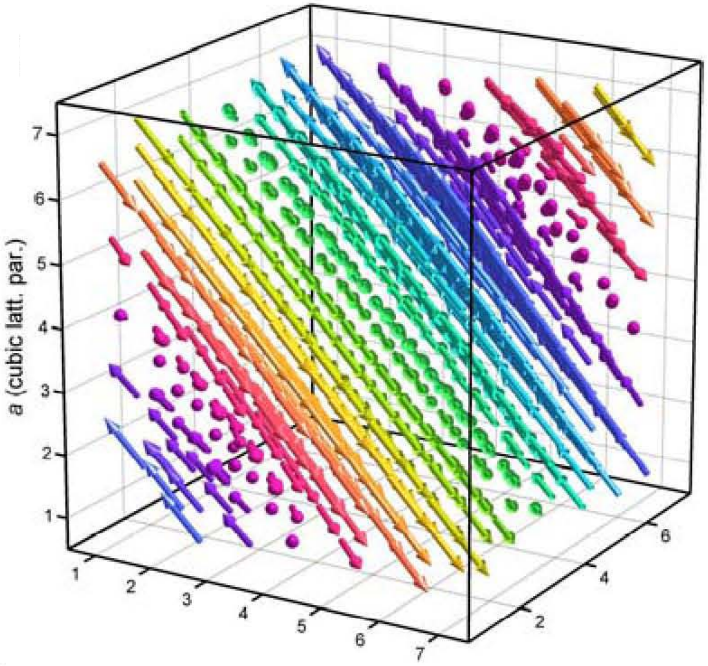


Color code indicates spin direction

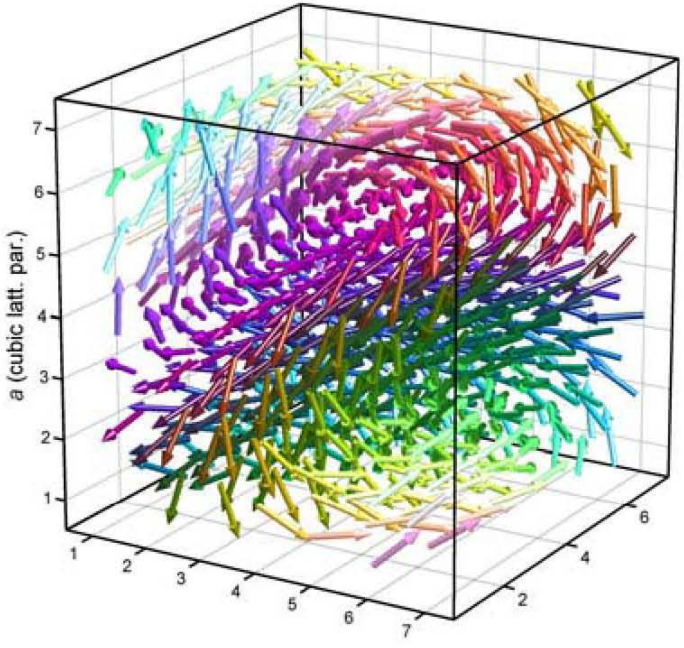
Ground states of finite clusters with exchange and DM interactions

*A. Hamann et al., PRL **107**, 037207 (2011)*

$$\mathcal{H} = -\frac{1}{2N}\sum_{i=1}^{N}\left(\sum_{j(i)}\left(J\mathbf{s}_i\cdot\mathbf{s}_j + \mathbf{D}_{i,j}\cdot(\mathbf{s}_i\times\mathbf{s}_j)\right)\right), \quad \text{with}\,|\mathbf{D}_{i,j}| = D \text{ and } \mathbf{D}_{i,j} = -\mathbf{D}_{j,i}\ .$$

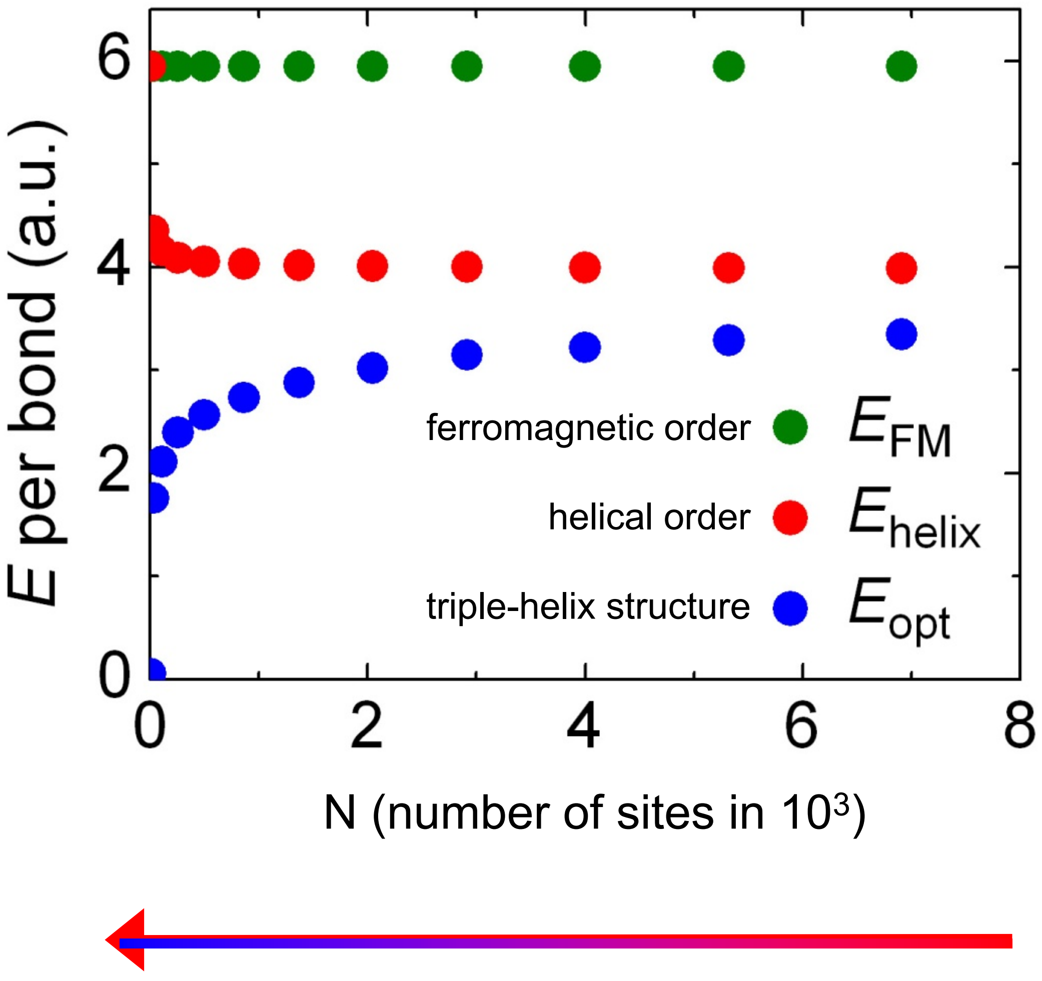


Helical order along [111] direction
locked by crystal potential



“triple helix” is the groundstate
in the absence of crystal potential

Qualitative energy considerations



- Triple-helix clusters destabilize with increasing size
- Triple-helix structure cannot be locked onto crystal anisotropy
- Single helix can be locked

→ Crystal potential makes the single helix favorable at low T

Transition at T_C should be of first order:

- Topologically distinct phases
- No continuous crossover

Conclusion and questions

- How does a metallic “spin solid“, notably in a heavy-fermion system, melt at a QCP?
  Break up of three-dimensionality?
  What happens to the Fermi surface?
  Prospects of spin-liquid phases (cf. $YbRh_2Si_2$).

- Robust quantum-critical concentration range in $CeCu_{6-x}Au_x$: sample with $x = 0.5$ can under hydrostatic pressure be driven to magnetic ordering wave vector of $x = 0.3$. Likewise, scaling of the volume thermal expansivity is observed up to $x = 0.5$.

- What is the origin of the anomalous QCP in $CeCu_{6-x}Au_x$ and $YbRh_2Si_2$ as opposed to $CeCu_2Si_2$ and other systems following the standard Hertz-Millis-Moriya scenario.

- Thermal expansion of $CeCoIn_5$ suggests a quantum-critical line in the $(B,p,T=0)$ plane emanating from a QCP at $p \approx -1$ kbar for $B = 0$ and passing through $B \approx 4$ T for $p = 0$.

- Approach to QCP in Ni-doped $CePd_{1-x}Ni_xAl$ for $x \approx 0.14$: $C/T \sim \log(T_0/T)$.

- MnSi: competing energies (exchange, DM, ...) lead to new topological phases and unusual phases retaining helical pitch (triple-helix-structure).