Thermodynamic Properties of low-dimensional Spin Systems

Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln

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II. Physikalisches Institut
Universität zu Köln
Zülpicher Str. 77
50937 Köln

SFB 608

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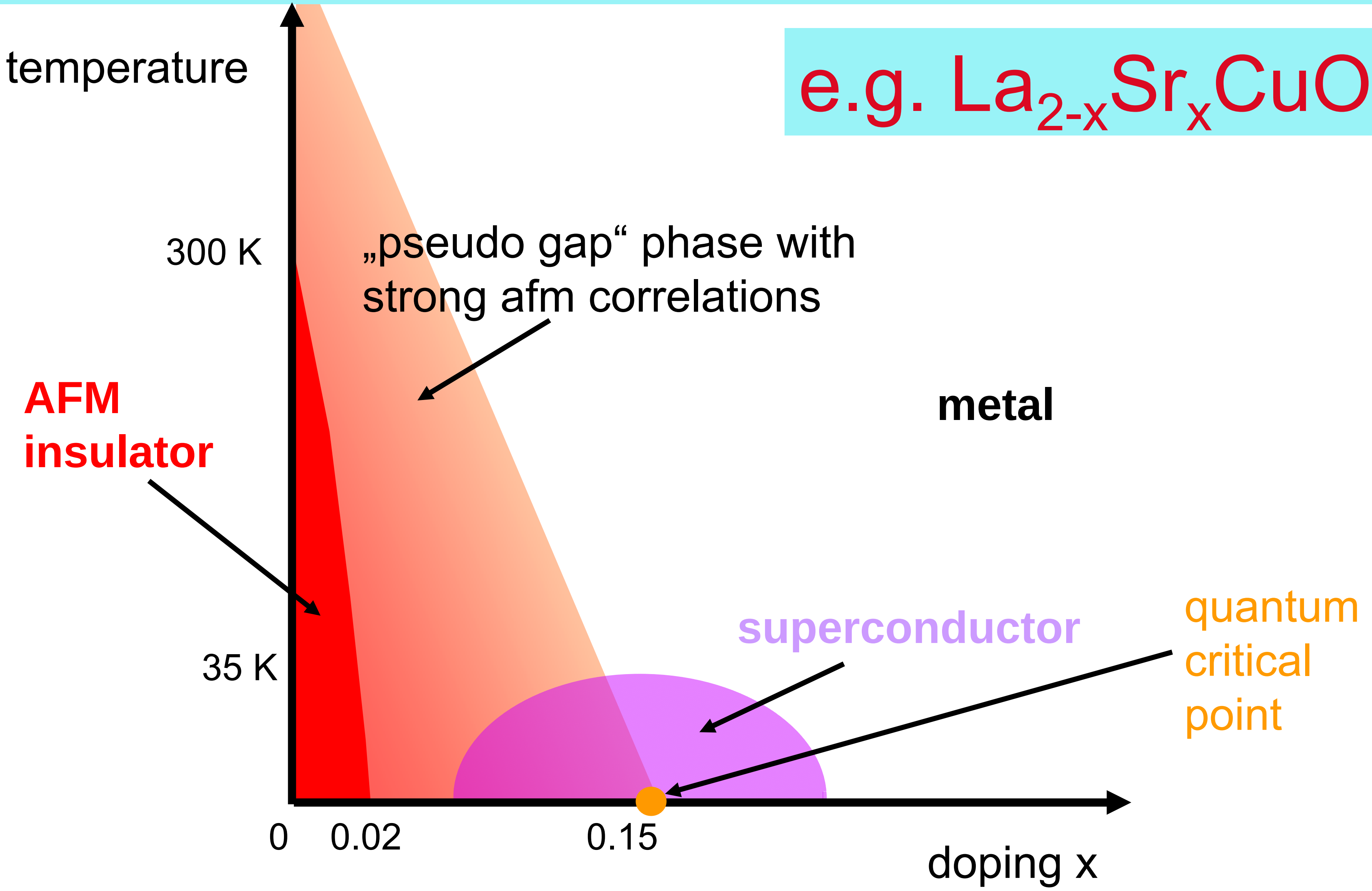
Thomas Lorenz, II. Physikalisches Institut, Universität zu Köln

- Motivation: Néel Order ⇔ Spin-liquid state
- Heisenberg Spin-½ chains & Spin-Peierls transition
- Thermodynamics of the Spin-Peierls compound $CuGeO_3$
- Quantum Phase transitions in Spin-½ chains and Spin-½ ladders
- Spin-½ chains „CuPzN“ & Spin-½ ladders „$HpipCuBr_4$“
- Beyond 1D Heisenberg Spin-½ systems

  higher dimension, anisotropic exchange , larger spin
- Summary

Phase diagram of High-Tc´s

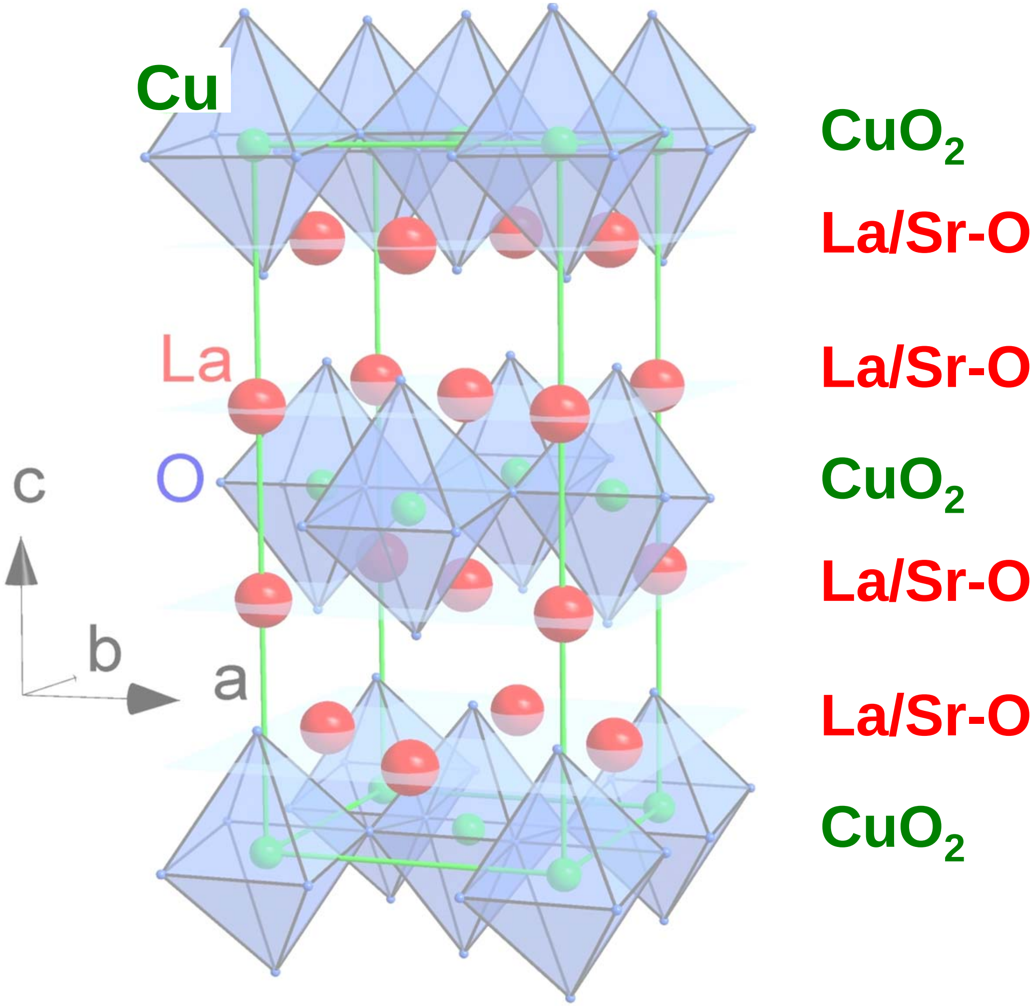
e.g. $La_{2-x}Sr_xCuO_4$

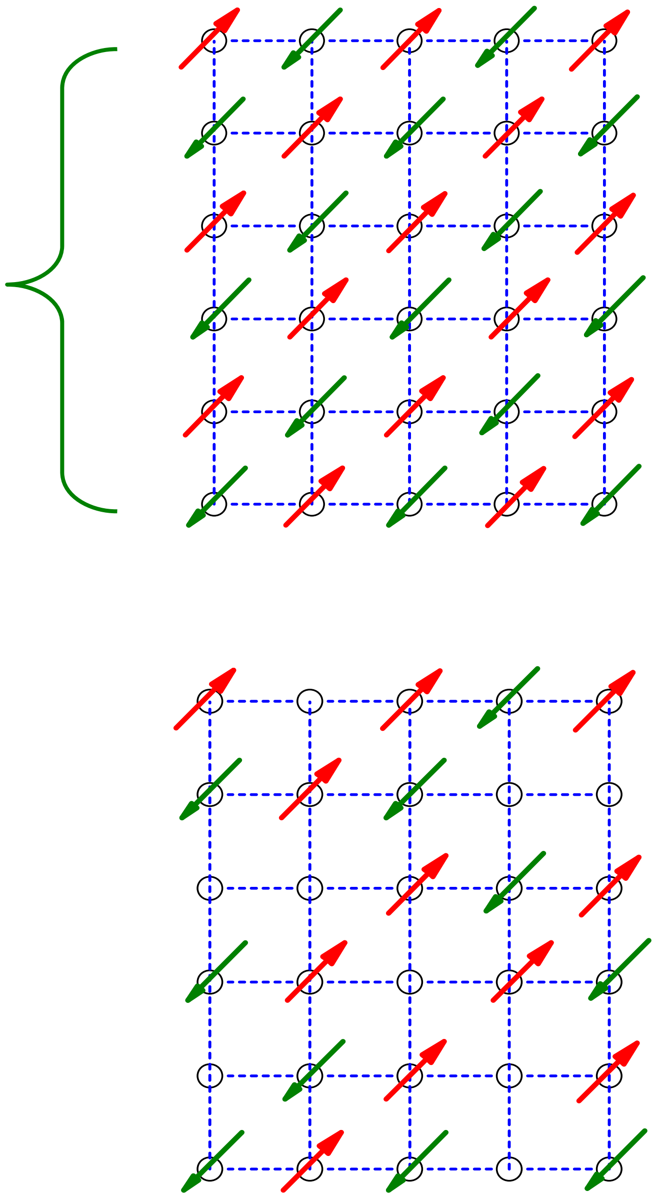


Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

Structure of High-Tc´s

e.g. $La_{2-x}Sr_xCuO_4$





Cu^{2+}

S=1/2

AFM insulator

„Spin solid“

x< 0.25

strongly cor-related metal

„Spin liquid“

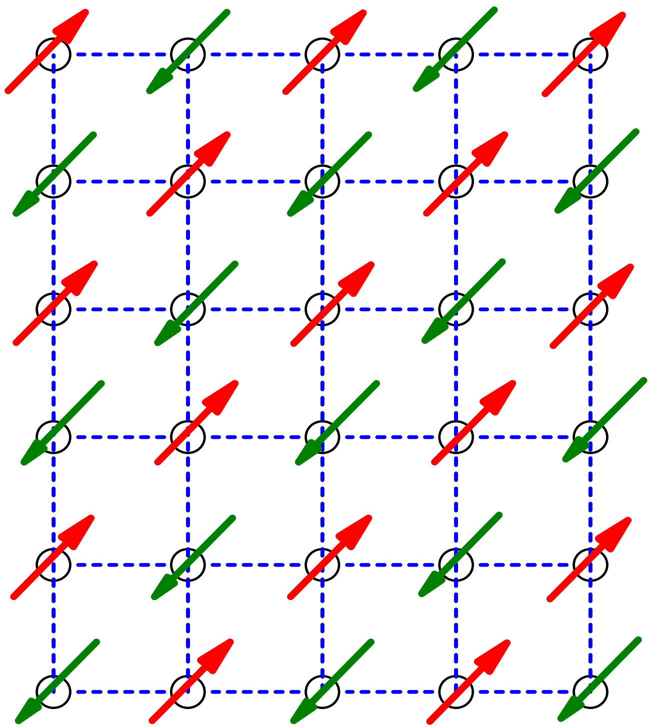
Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

Néel order vs. spin singlet state

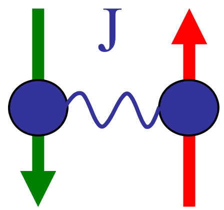
$$\mathrm{H} = J\sum_{\langle i,j\rangle} \vec{S}_i \vec{S}_j = J\sum_{\langle i,j\rangle} \frac{1}{2}\left(S_i^+ S_j^- + S_i^- S_j^+\right) + S_i^z S_j^z$$

$$S_i^{\pm} = S_i^x \pm i S_i^y$$

$$\mathrm{H} = J\sum_{\langle i,j\rangle} S_i^z S_j^z \quad \Rightarrow \quad \frac{E}{N} = -\frac{z}{2}\frac{J}{4}$$



$$|S=0\rangle = \frac{|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle}{\sqrt{2}} \quad \Rightarrow \quad E = -\frac{3}{4}J$$



$$|S=1\rangle = \begin{cases} |\uparrow\uparrow\rangle \\ \left(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle\right)/\sqrt{2} \\ |\downarrow\downarrow\rangle \end{cases}$$

Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

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Néel order vs. spin singlet state

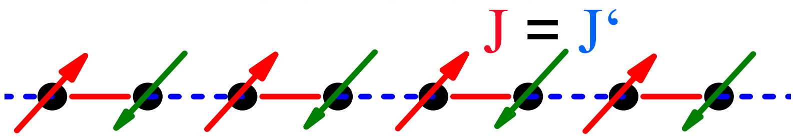
$$\mathrm{H} = J\sum_{\langle i,j\rangle} \vec{S}_i \vec{S}_j = J\sum_{\langle i,j\rangle} \frac{1}{2}\left(S_i^+ S_j^- + S_i^- S_j^+\right) + S_i^z S_j^z \qquad S_i^{\pm} = S_i^x \pm i S_i^y$$

$$\mathrm{H} = J\sum_{\langle i,j\rangle} S_i^z S_j^z \quad \Rightarrow \quad \frac{E}{N} = -\frac{z}{2}\frac{J}{4}$$

$$\left|S=0\right\rangle = \frac{\left|\uparrow\downarrow\right\rangle - \left|\downarrow\uparrow\right\rangle}{\sqrt{2}} \quad \Rightarrow \quad E = -\frac{3}{4}J$$

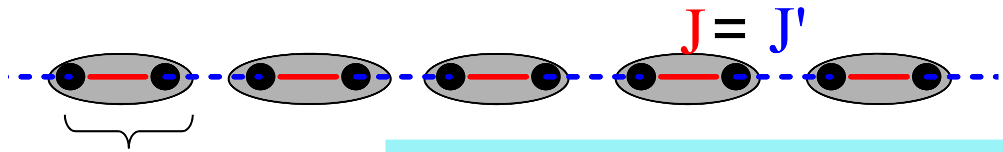
1 D Spin-1/2 chain

Néel state



$$E/N = -\frac{1}{4}J = -0.25\ J$$

Singlet-dimer state



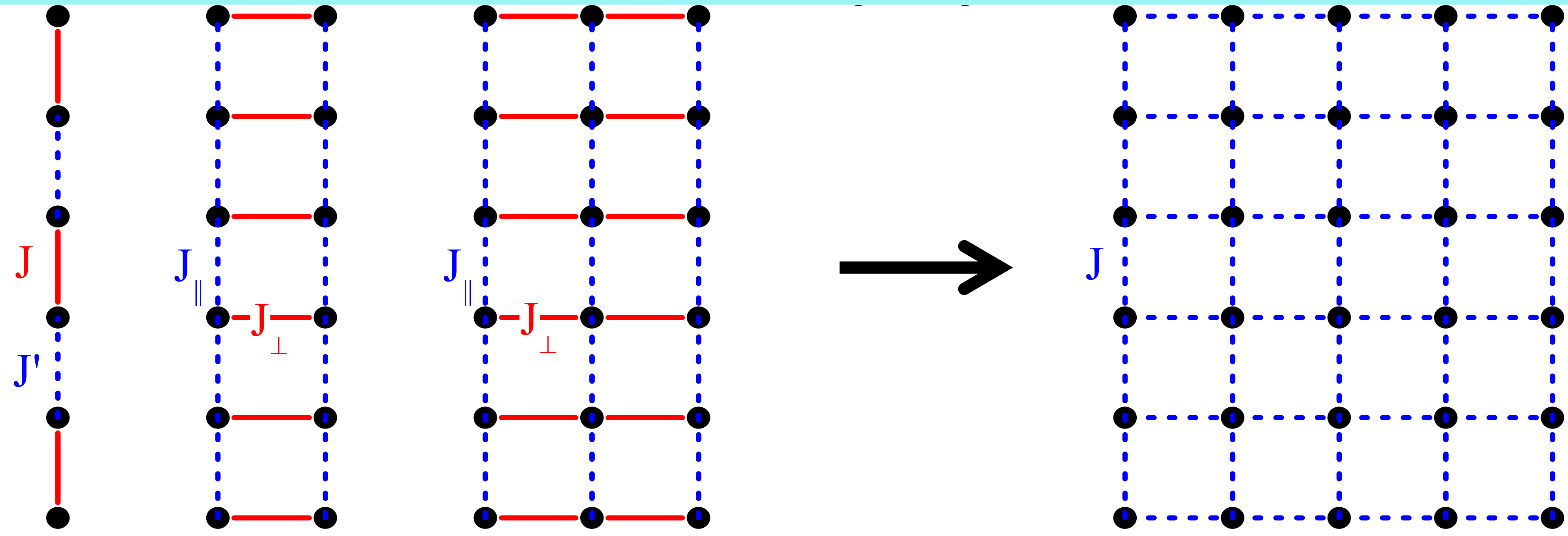
$$\frac{1}{\sqrt{2}}\left(\left|\uparrow\downarrow\right\rangle - \left|\uparrow\downarrow\right\rangle\right)$$

$$E/N = -\frac{1}{2}\frac{3}{4}J = -0.375\ J$$

Exact ground state $E/N \cong -0.44\,J$

Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

From 1D chains to 2D square lattice



Spin ½ chain: ~ dimer ground state

Spin ½ ladder: dimer ground state

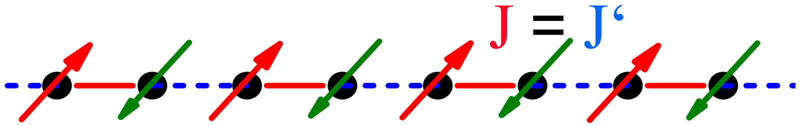
Three-leg spin ½ ladder: ~ spin ½ chain

spin ½ square lattice: ~ Néel state

Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

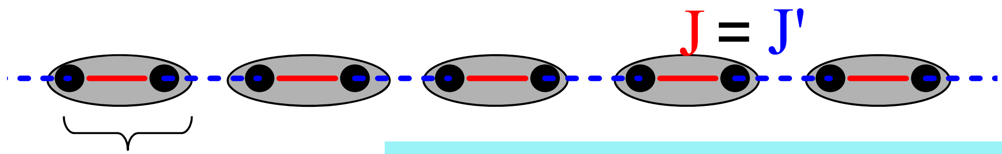
Heisenberg-Spin-1/2 chain

Néel state



$$E/N = -\frac{1}{4}J = -0.25\ J$$

Singlet-dimer state

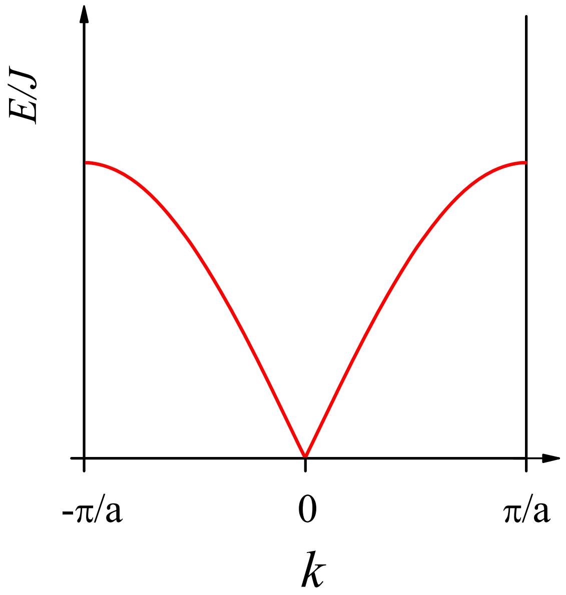


$$\frac{1}{\sqrt{2}}\left(|\uparrow\downarrow\rangle - |\uparrow\downarrow\rangle\right)$$

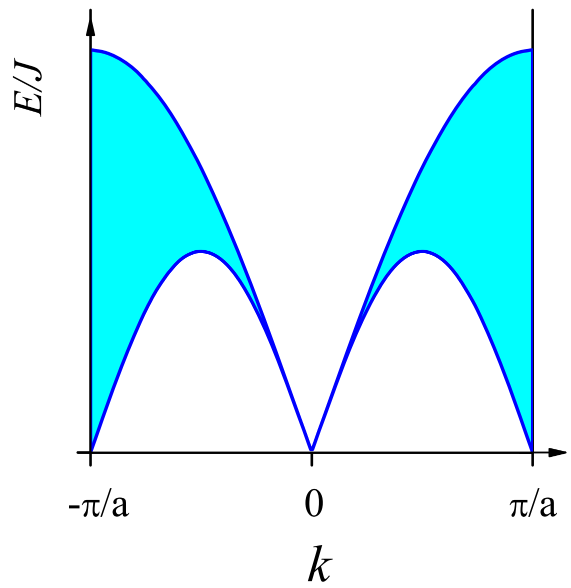
$$E/N = -\frac{1}{2}\frac{3}{4}J = -0.375\ J$$

Excitation spectra

Spin waves; magnons with S=1



2-spinon continuum with S=1/2



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II. Physikalisches Institut
Universität zu Köln
Germany

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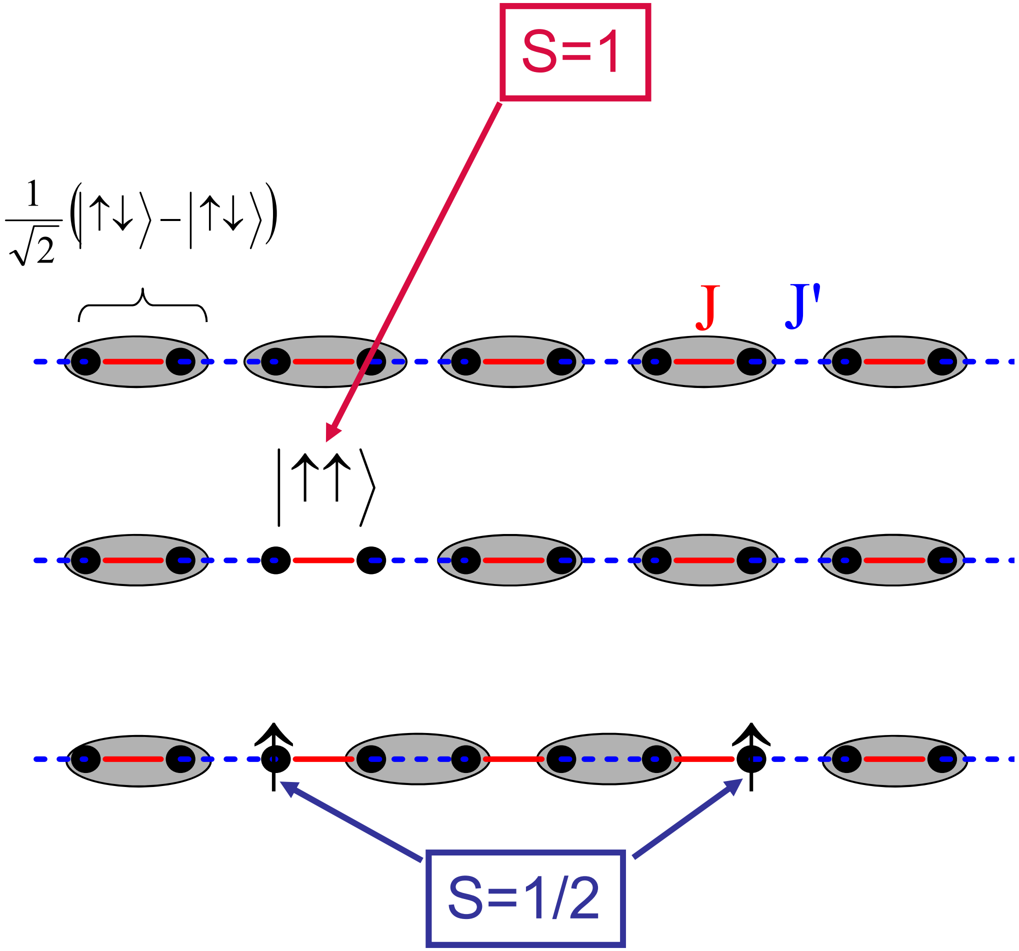
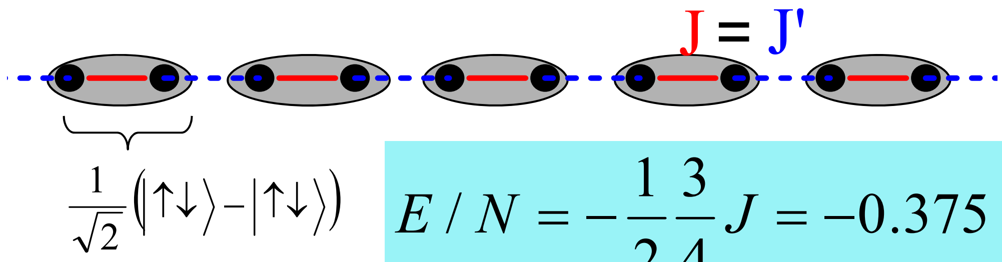
Heisenberg-Spin-1/2 chain

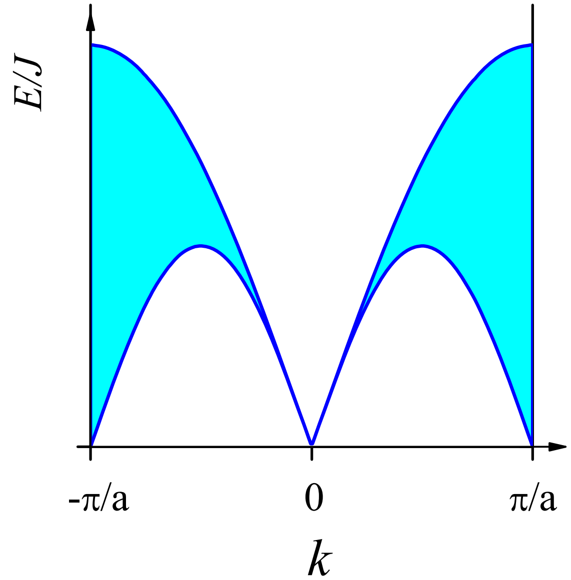


Singlet-dimer state



$$E / N = -\frac{1}{2}\frac{3}{4} J = -0.375\ J$$

2-spinon continuum with S=1/2



Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
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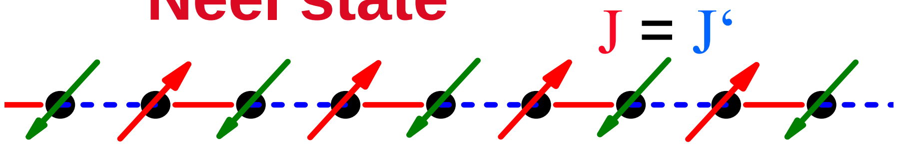
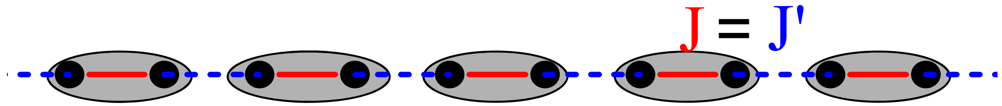
Thermodynamics of Spin-1/2 chains

Néel state



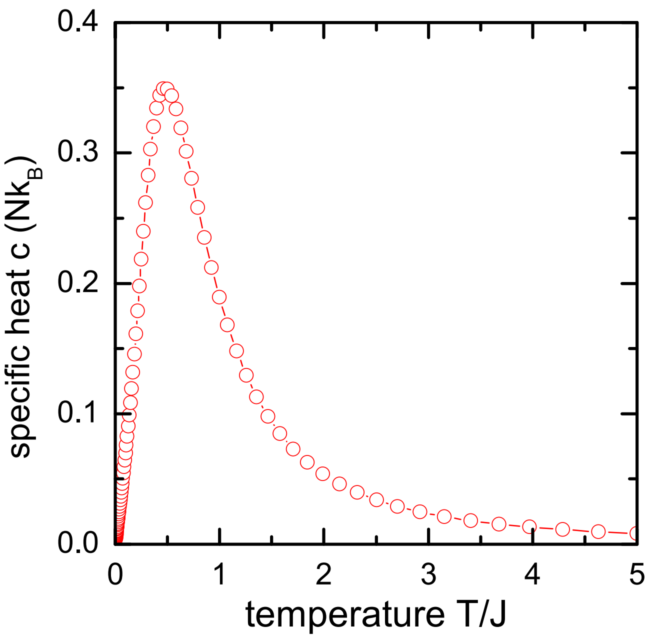
Singlet-dimer state

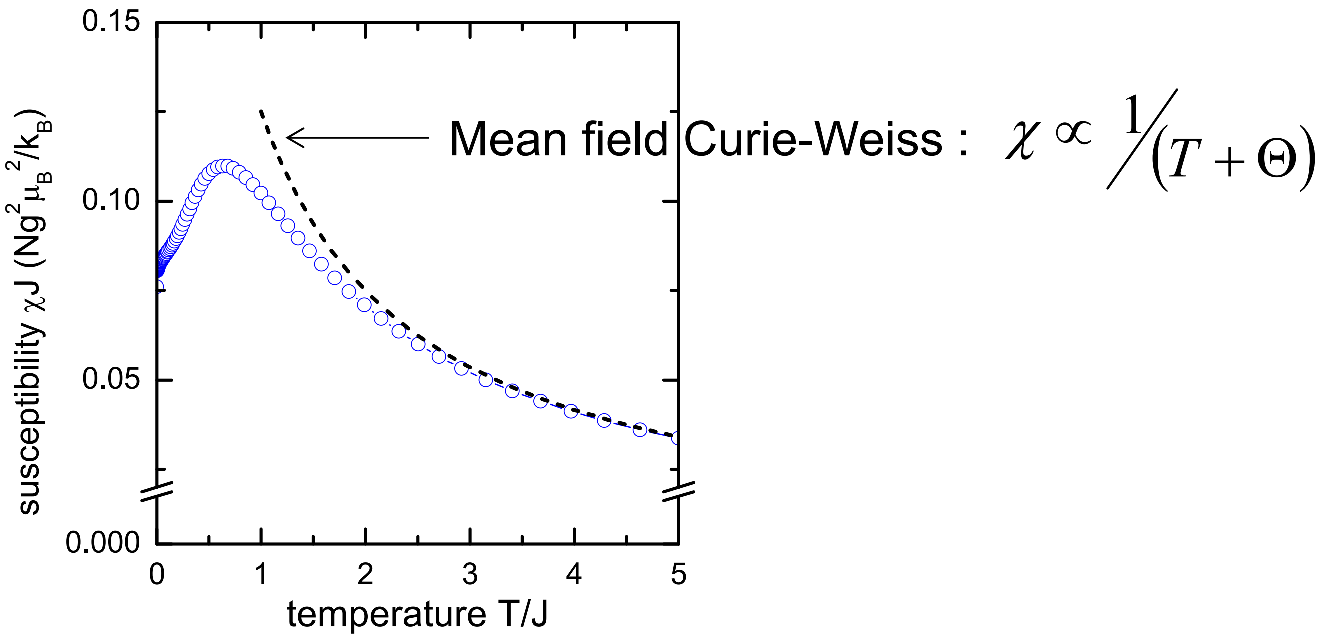


$$\mathrm{H} = J\sum_j \vec{S}_j \vec{S}_{j+1} = J\sum_j \frac{1}{2}\left(S_j^+ S_{j+1}^- + S_j^- S_{j+1}^+\right) + S_j^z S_{j+1}^z$$

⇒ solved exactly by Bethe ansatz A. Klümper, EPJ B**5**, 677 (1998)

⇒ free energy, entropy, specific heat, susceptibility, …





Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

Beyond NN Heisenberg Spin-1/2 chains

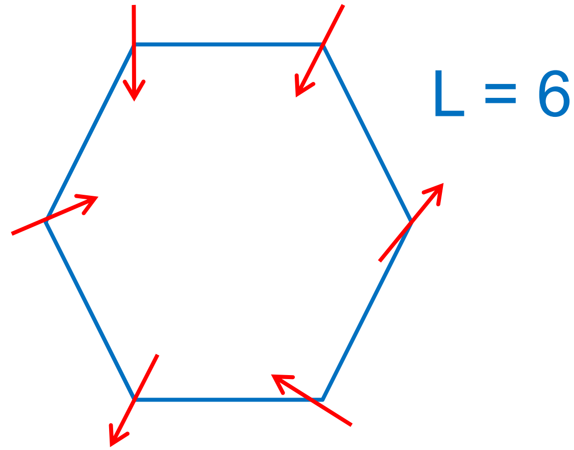
NN Heisenberg: $H = J\sum_j \vec{S}_j \vec{S}_{j+1}$

anisotropic exchange: $H = J\sum_j \left\{ S_j^x S_{j+1}^x + S_j^y S_{j+1}^y + \Delta\, S_j^z S_{j+1}^z \right\}$

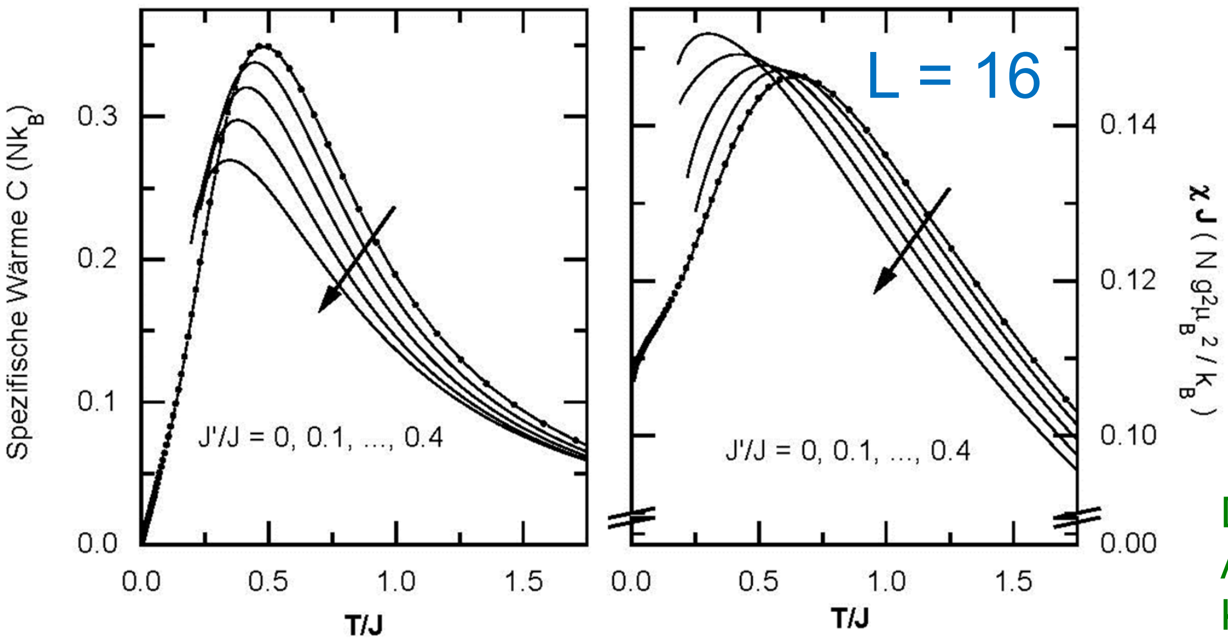
NN & NNN exchange: $H = \sum_j J\vec{S}_j \vec{S}_{j+1} + J'\,\vec{S}_j \vec{S}_{j+2}$

⇒ in general, no exact solution

⇒ numerical approximations, e.g., Quantum Monte Carlo, diagonalization of chains of finite length L, ...



Example: $J_{NN} = J$ & $J_{NNN} = J'$

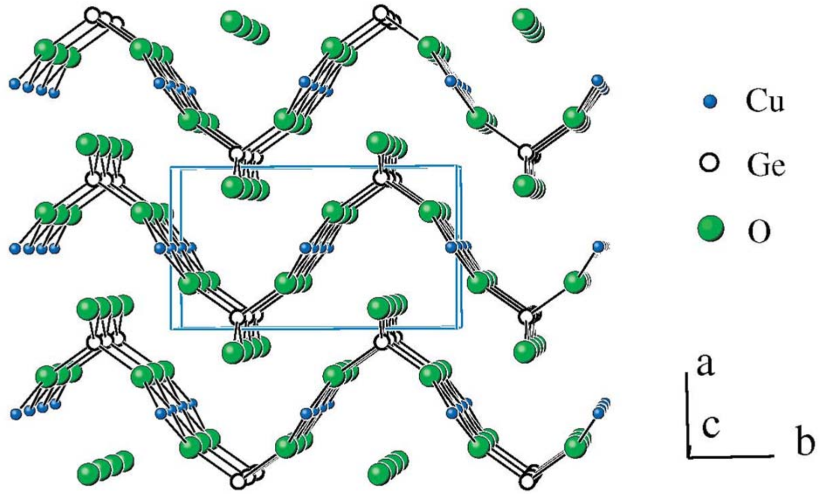


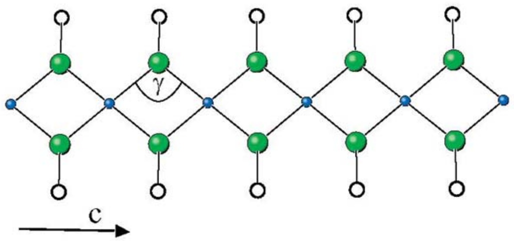
=> Finite size effects at low T

Data from:
A. Klümper, EPJB **5**, 677 (1998)
K. Fabricius et al. PRB **57**, 1102 (1998)

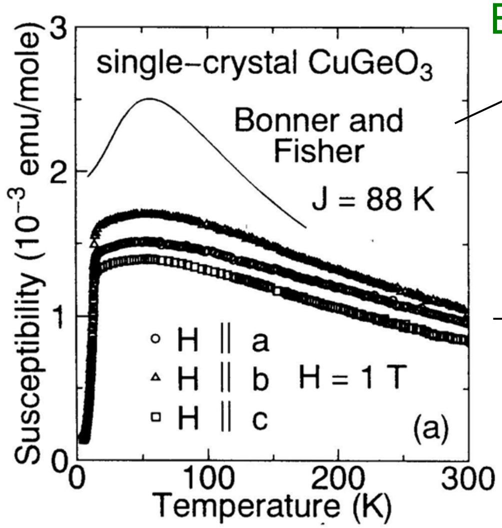
Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

Spin-1/2 chain material $CuGeO_3$





=> Cu^{2+} spin-1/2 chains || c axis



Bonner & Fisher PR **135**, A640(1964)

finite chains with L = 10

poor agreement with theoretical expectation

M. Hase et al. PRL **70**, 3651 (1993)

$$\chi(T \to 0) \to 0$$

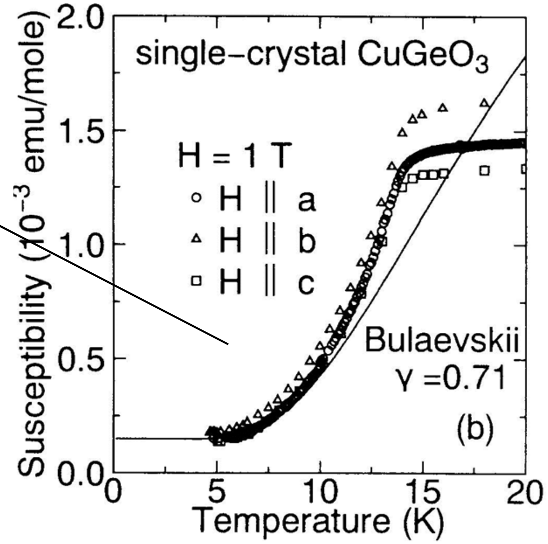
independent of field direction

⇒ no antiferromagnetic order

$$\chi_{\|}(T \to 0) \to 0 \quad \chi_{\|}(T < T_N) \approx const.$$

⇒ formation of a nonmagnetic spin-singlet ground state

⇒ “Spin-Peierls transition”



Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
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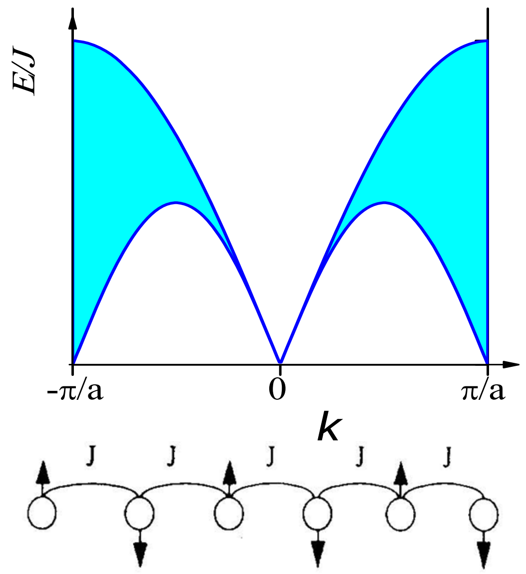
Spin-Peierls transition



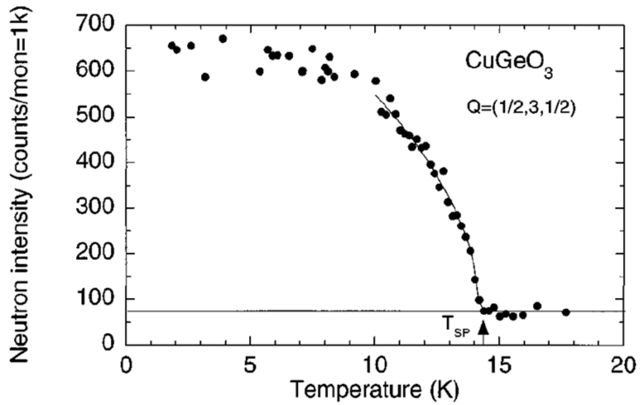
- uniform chain
- resonating dimers
- no spin gap
- continuous 2-spinon excitations (two S=1/2)

U => D : structural transition driven by a gain in magnetic energy:

$$\Delta E = +c_{elastic}\,\delta^2 - c_{magnetic}\,\delta^{4/3}$$

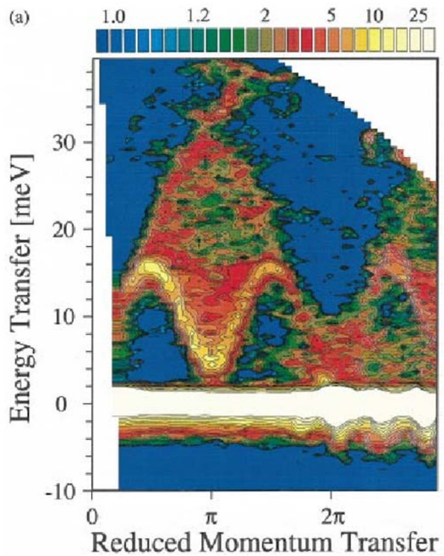
Cross & Fisher
PRB **19**, 402(1979)

Neutron scattering data



⇒ Doubling of unit cell

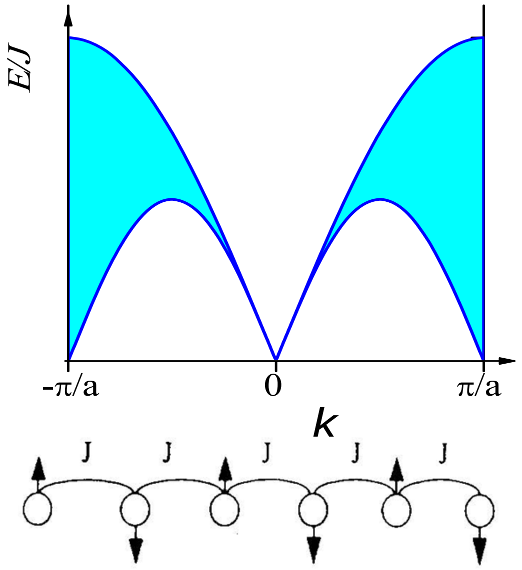
Regnault et al.
PRB **53**, 5579 (1995)

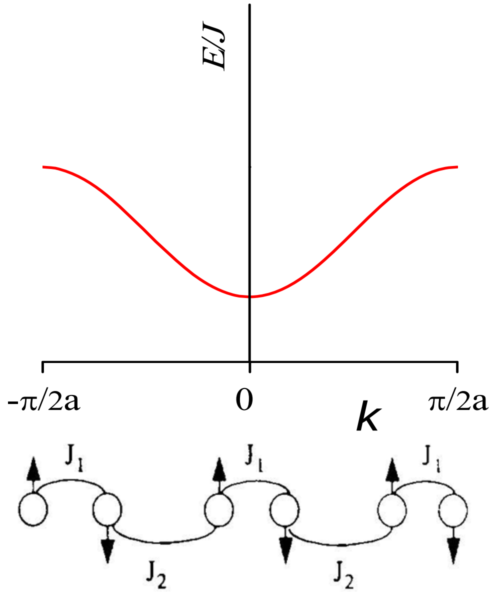


Arai et al.
PRL **77**, 3649 (1996)

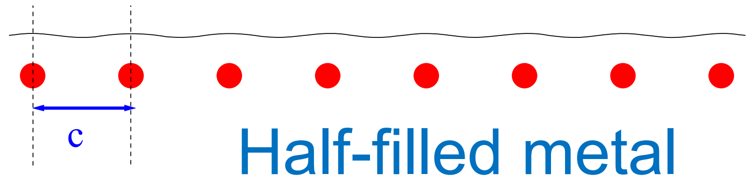
⇒ 2-spinon continuum

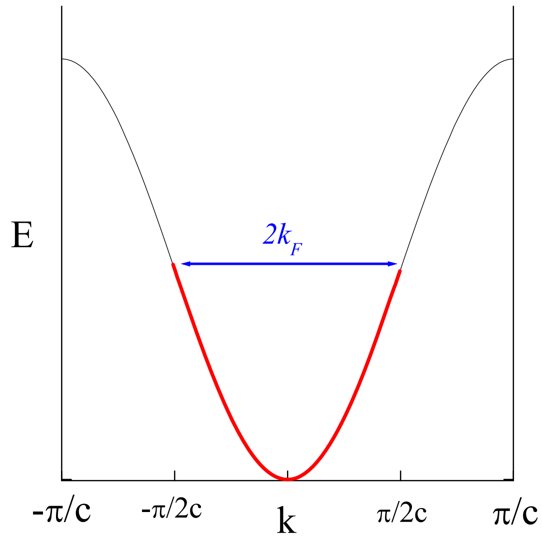
Spin-Peierls transition





Peierls transition in 1d metals





- uniform chain
- resonating dimers
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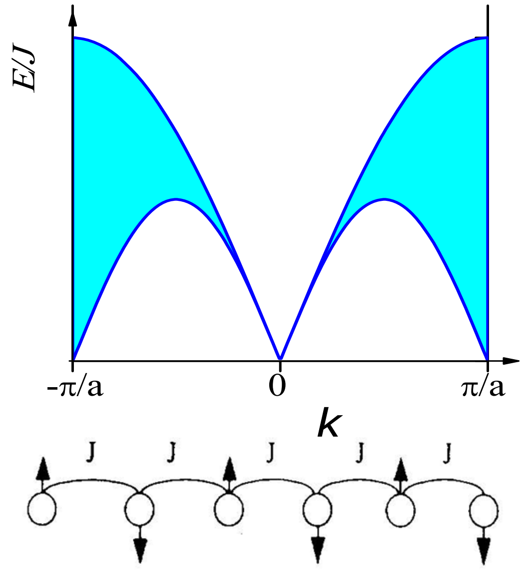
- dimerized chain $J_{1,2} = J(1 \pm \delta)$
- dimers on strong bonds
- finite spin gap
- triplet excitations (S=1)
- doubling of unit cell

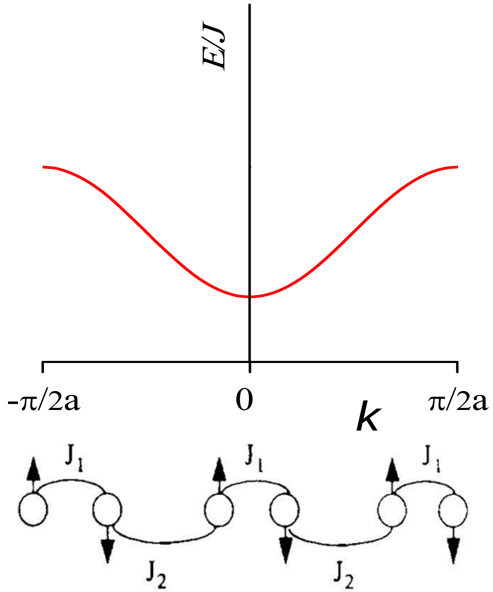
U => D : structural transition driven by a gain in magnetic energy:

$$\Delta E = +c_{elastic}\,\delta^2 - c_{elastic}\,\delta^{4/3}$$

Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

Spin-Peierls transition





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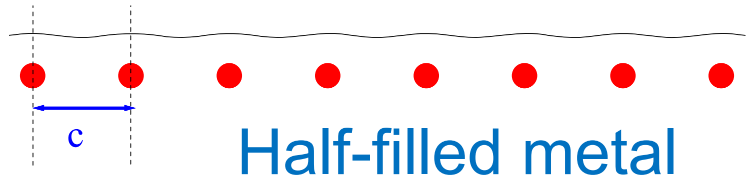
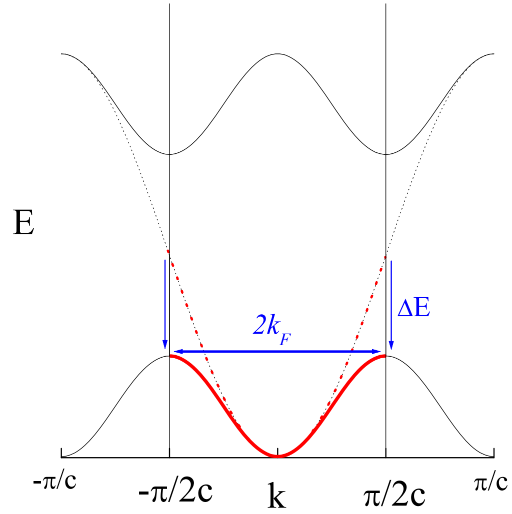
U => D : structural transition driven by a gain in magnetic energy:

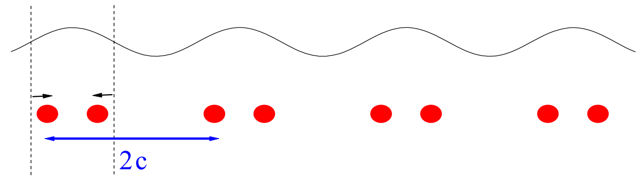
$$\Delta E = +c_{elastic}\,\delta^2 - c_{mag}\,\delta^{4/3}$$

Peierls transition in 1d metals



Half-filled metal



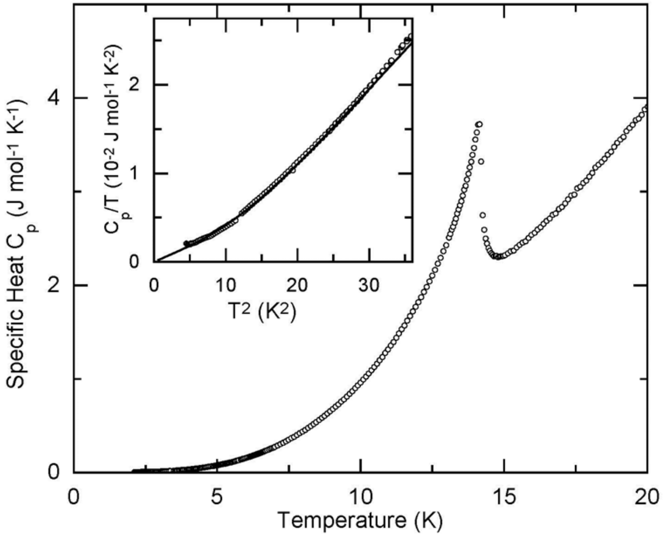


Dimerized, gapped insulator

Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

Spin-Peierls transition in $CuGeO_3$

Specific heat



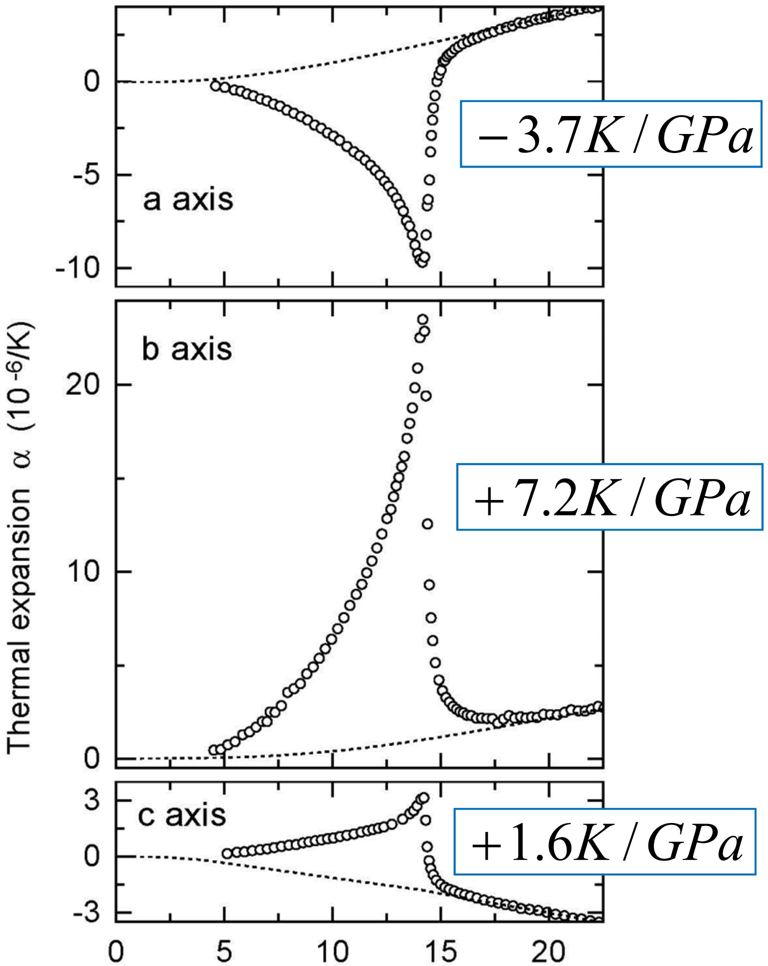
$$C_p = \beta T^3 + \Gamma \exp\left(\frac{-\Delta}{k_B T}\right) \rightarrow \Delta \cong 23 \text{ K}$$

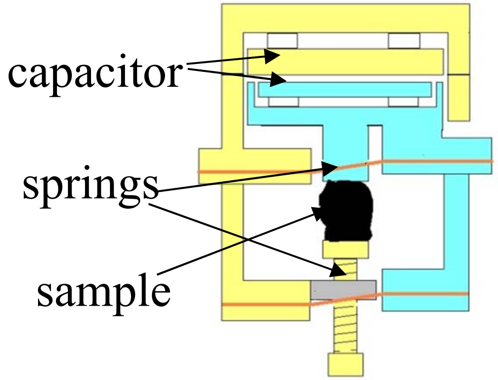
$$\alpha_i = \frac{1}{L_i}\frac{\partial L_i}{\partial T} \qquad i = a, b, c \text{ axis}$$

Ehrenfest relation:

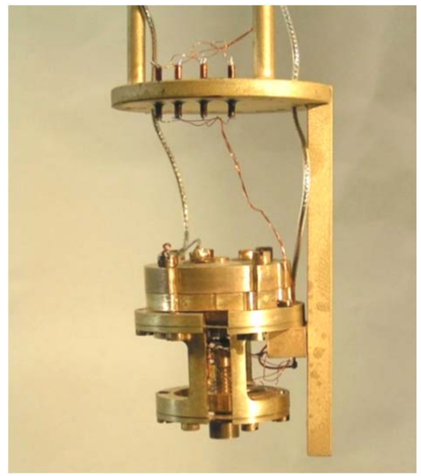
$$\frac{\partial T_c}{\partial p_i} = T_c V_{mol} \frac{\Delta \alpha_i}{\Delta C_p}$$

Thermal expansion





$$\Delta L_i \rightarrow \Delta d \rightarrow \Delta C$$



$$\Delta L_i / L_0 \approx 10^{-9}$$

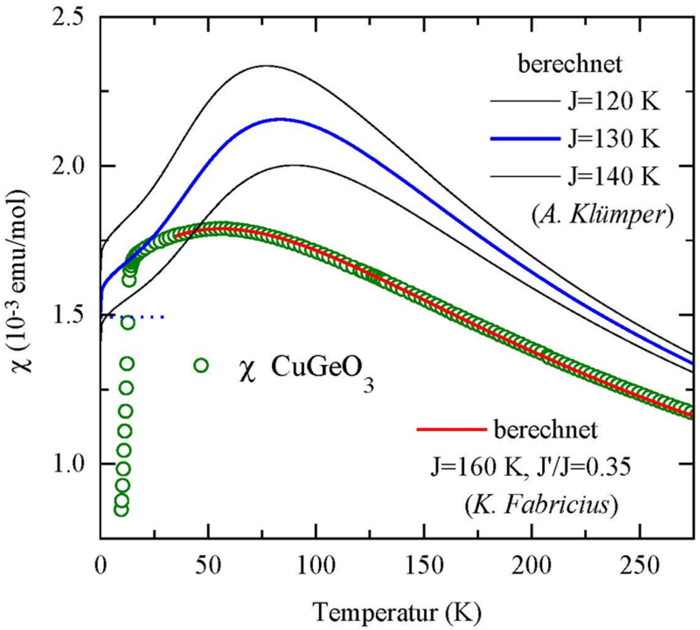
$\Rightarrow$ Unexpectedly large, highly anisotropic pressure dependencies of T_{SP}

Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

TL et al. PRB **54**, R15611 (1996)

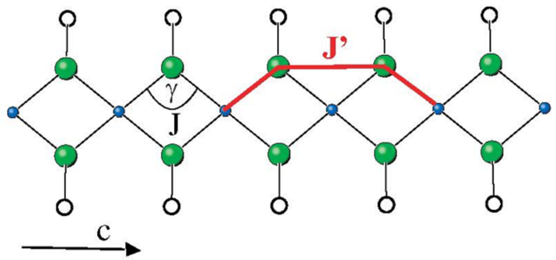
Uniform phase of $CuGeO_3$

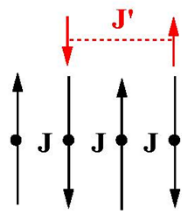
susceptibility



analy-
tically
exact
at all T

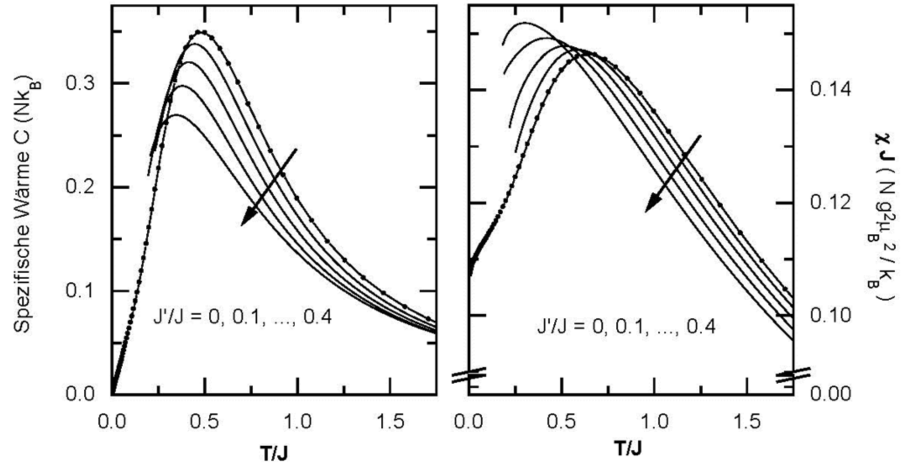
L = 28
T > 25K



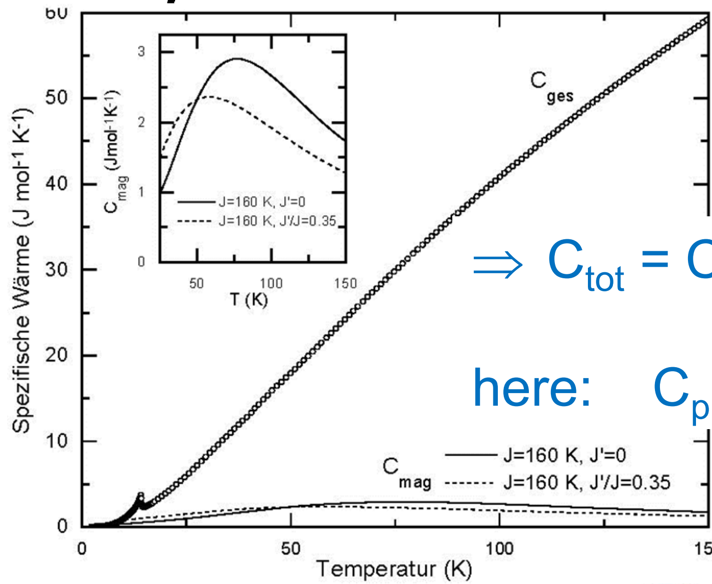


$\Rightarrow$ strongly frustrated spin chains due to competing NN and NNN exchange

calculated C(T,J,J') & χ (T,J,J')



Specific heat



$\Rightarrow$ $C_{tot} = C_{ph} + C_{mag}$

here: $C_{ph} >> C_{mag}$

Thomas Lorenz
II. Physikalisches Institut
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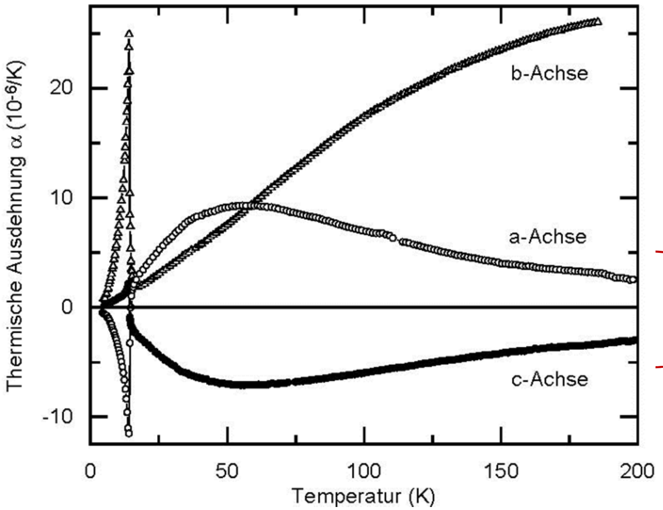
Germany

TL et al. PRB **54**, R15611 (1996)
Fabricius, …,TL, et al. PRB **57**, 1102 (1998)

Uniform phase of $CuGeO_3$

Thermal expansion



$\alpha_a \sim -2\,\alpha_c$

max (min) ~ 58 K

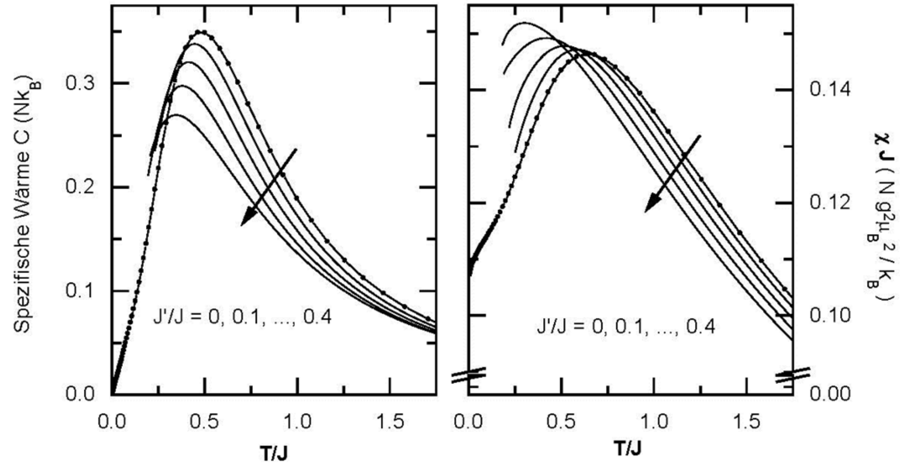
Maxwell relation :

$$\frac{\partial^2 G}{\partial T \partial p} = -\frac{\partial S}{\partial p} = \frac{\partial V}{\partial T} = V\alpha_{vol.}$$

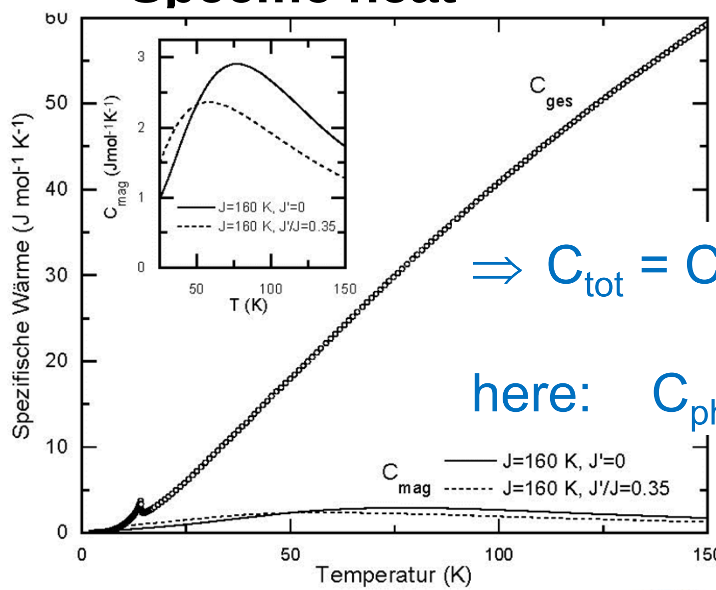
$$S = S\left(\frac{T}{J}, \frac{J'}{J}\right) \quad \Rightarrow \quad C_{mag} = \frac{T}{J}\partial_1 S\left(\frac{T}{J}, \frac{J'}{J}\right)$$

$$V\alpha_i^{mag} = \frac{\partial \ln J}{\partial p_i} C_{mag} - \partial_2 S\left(\frac{T}{J}, \frac{J'}{J}\right)\frac{\partial J'/J}{\partial p_i}$$

calculated C(T,J,J') & χ (T,J,J')



Specific heat



$\Rightarrow$ $C_{tot} = C_{ph} + C_{mag}$

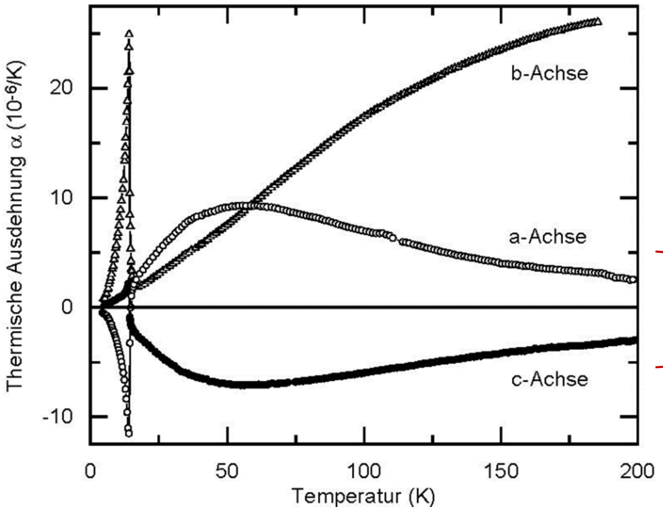
here: $C_{ph} \gg C_{mag}$

Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

TL et al. PRB **54**, R15611 (1996)
Fabricius, …,TL, et al. PRB **57**, 1102 (1998)

Uniform phase of $CuGeO_3$

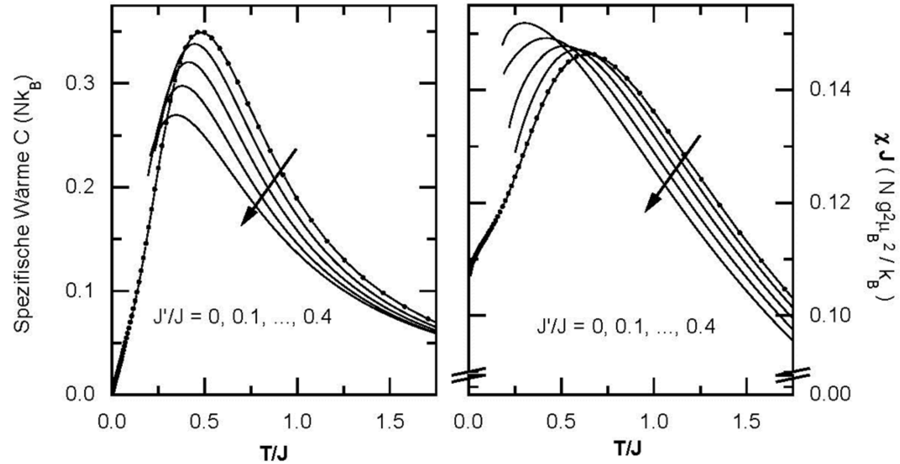
Thermal expansion



$\alpha_a \sim -2\ \alpha_c$

max (min) ~ 58 K

calculated C(T,J,J') & χ (T,J,J')



Maxwell relation :

$$\frac{\partial^2 G}{\partial T \partial p} = -\frac{\partial S}{\partial p} = \frac{\partial V}{\partial T} = V\alpha_{vol.}$$

$$S = S\left(\frac{T}{J}, \frac{J'}{J}\right) \quad \Rightarrow \quad C_{mag} = \frac{T}{J}\partial_1 S\left(\frac{T}{J}, \frac{J'}{J}\right)$$

$$V\alpha_i^{mag} = \frac{\partial \ln J}{\partial p_i} C_{mag} - \partial_2 S\left(\frac{T}{J}, \frac{J'}{J}\right) \frac{\partial J'/J}{\partial p_i}$$

$\Rightarrow$

$$\alpha_i^{mag} \approx \frac{1}{V} \frac{\partial \ln J}{\partial p_i} C_{mag} \qquad \text{"Grüneisen scaling"}$$

if $\partial_2 S\left(\frac{T}{J}, \frac{J'}{J}\right) \frac{\partial J'/J}{\partial p_i}$ is small.

Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

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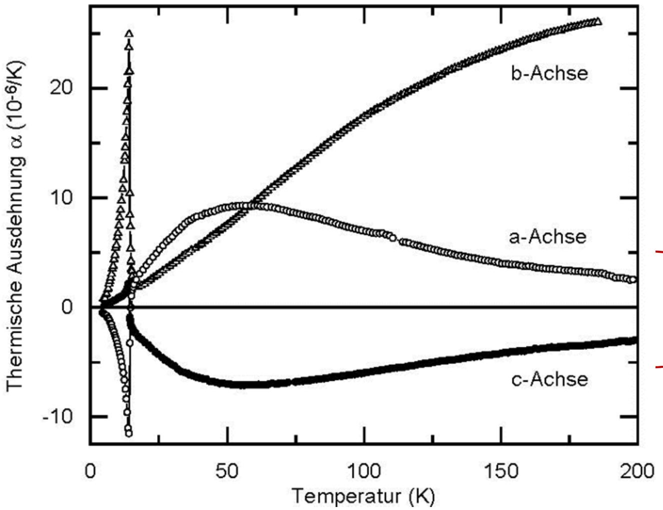
Uniform phase of $CuGeO_3$

Thermal expansion



$\alpha_a \sim -2\,\alpha_c$

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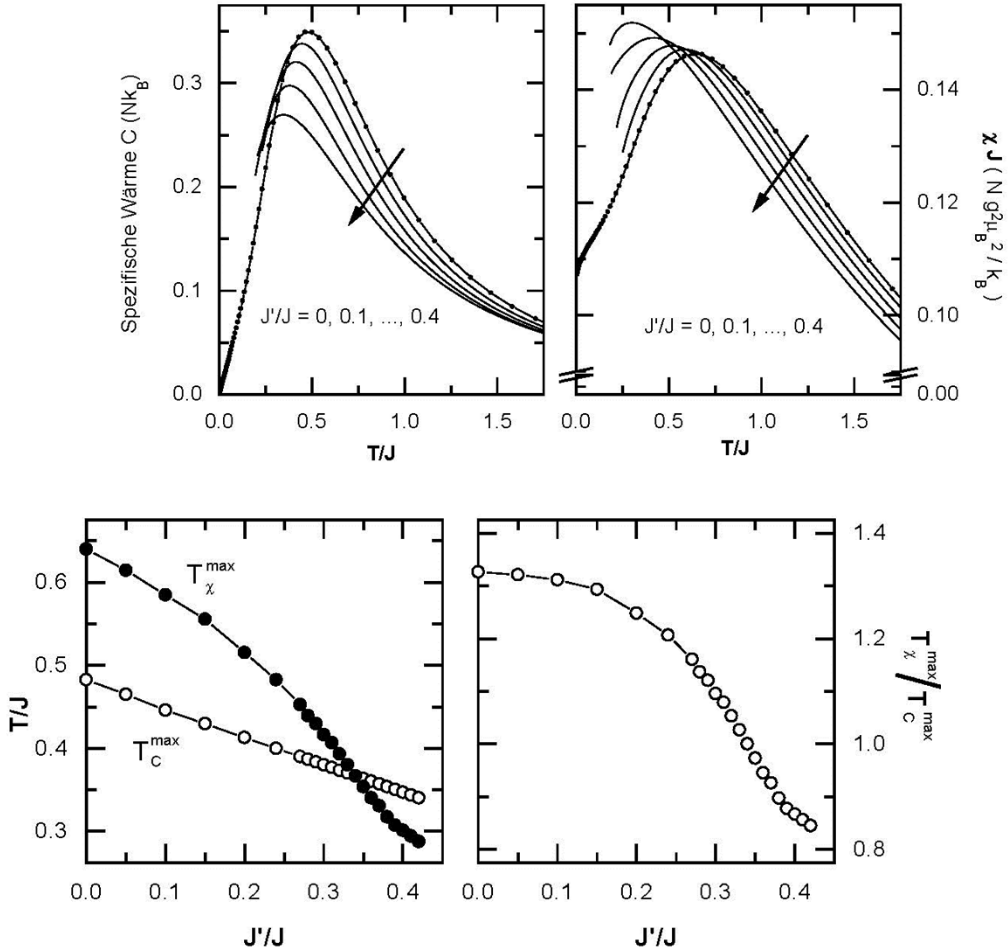
calculated C(T,J,J') & χ (T,J,J')



$$\alpha_i^{mag} \approx \frac{1}{V}\frac{\partial \ln J}{\partial p_i} C_{mag} \qquad \text{"Grüneisen scaling"}$$

$$\text{if} \quad \partial_2 S\left(\frac{T}{J},\frac{J'}{J}\right)\frac{\partial\, J'/J}{\partial p_i} \quad \text{is small.}$$

$$T_\chi^{\max} \approx 56\ \mathrm{K}$$

$$T_C^{\max} \approx 58\ \mathrm{K}$$

$$\Rightarrow \quad \frac{J'}{J} \approx 0.35$$

Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

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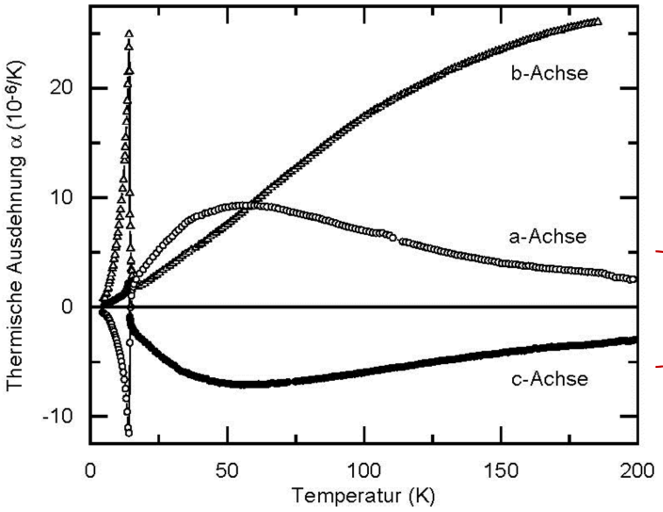
Fabricius, …,TL, et al. PRB **57**, 1102 (1998)

Uniform phase of $CuGeO_3$

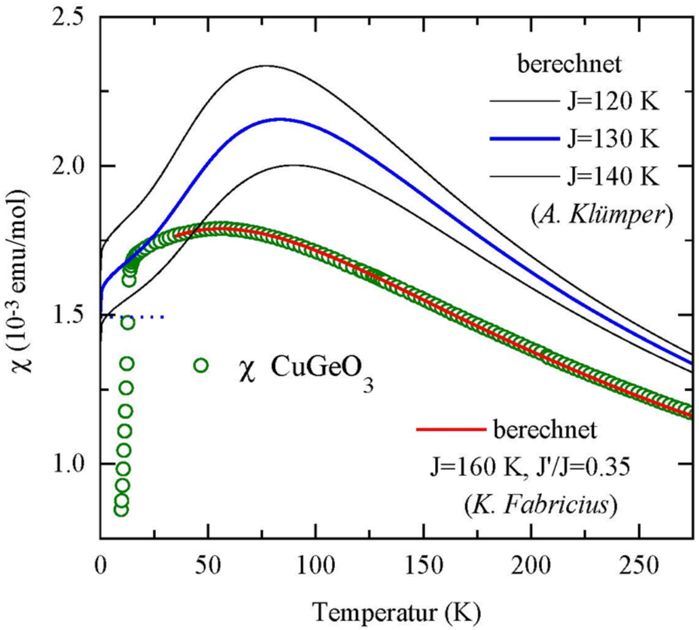
Thermal expansion



$\alpha_a \sim -2\,\alpha_c$

max (min) ~ 58 K

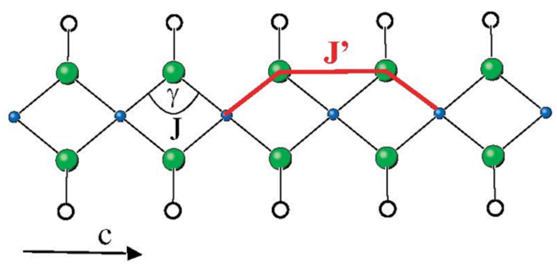
susceptibility

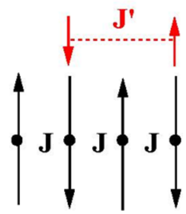


$J \approx 160\text{ K}$

$$\alpha_i^{mag} \approx \frac{1}{V}\frac{\partial \ln J}{\partial p_i} C_{mag} \qquad \text{"Grüneisen scaling"}$$

$$\text{if} \quad \partial_2 S\left(\frac{T}{J},\frac{J'}{J}\right)\frac{\partial J'/J}{\partial p_i} \quad \text{is small.}$$





$$T_\chi^{\max} \approx 56\text{ K}$$

$$T_C^{\max} \approx 58\text{ K}$$

$\Rightarrow$ $\frac{J'}{J} \approx 0.35$

Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

TL et al. PRB **54**, R15611 (1996)
Fabricius, …,TL, et al. PRB **57**, 1102 (1998)

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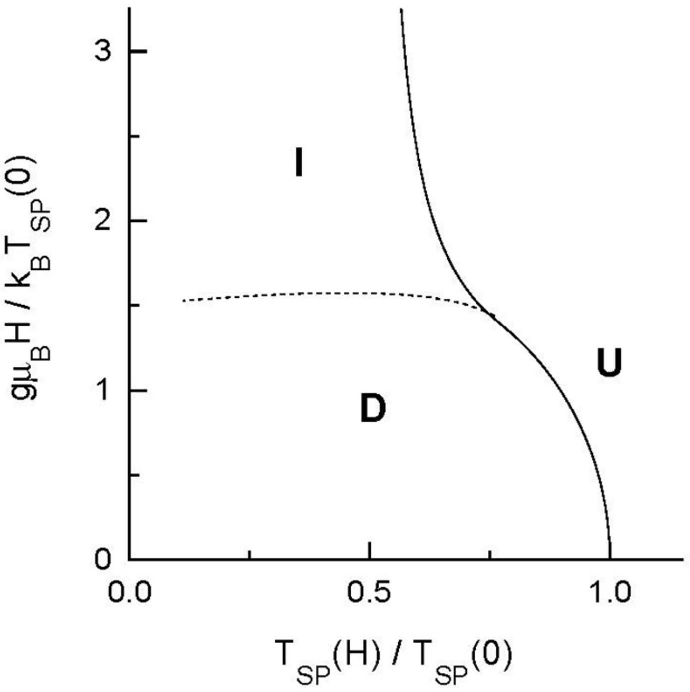
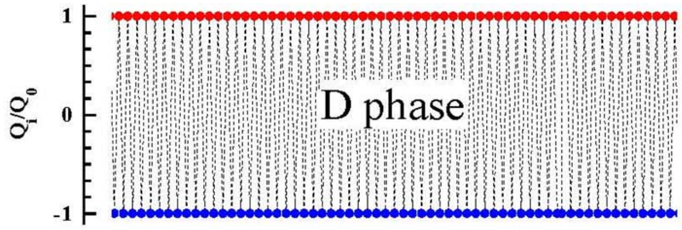
Spin-Peierls Phases in a Magnetic Field

Universal H–T–Phase Diagram



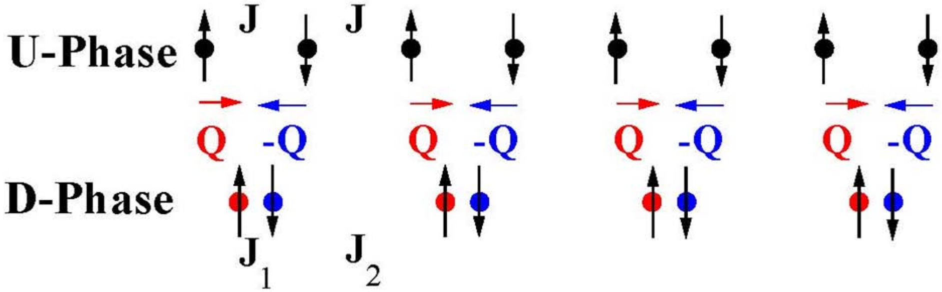
M.C. Cross PRB **20**, 4606 (1979)



D phase:

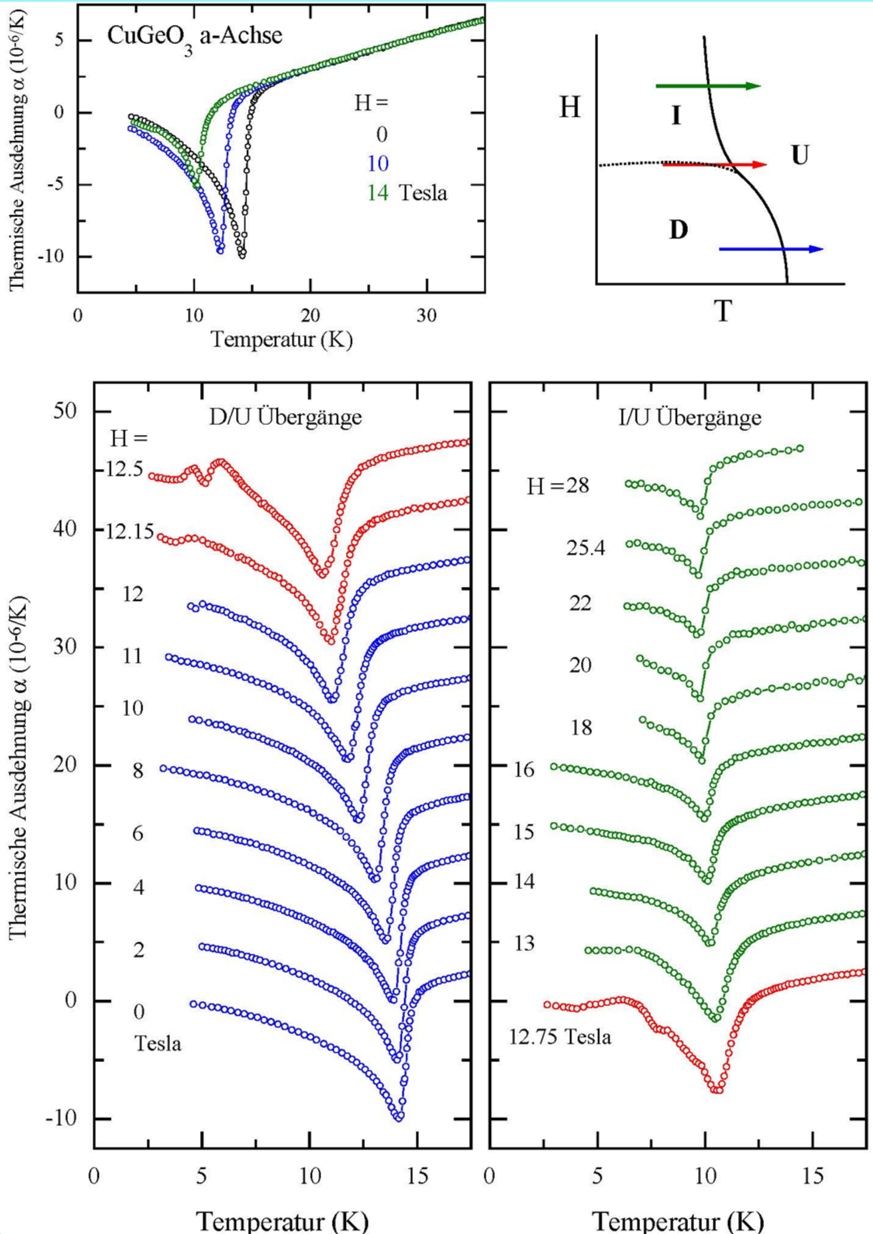
non–magnetic

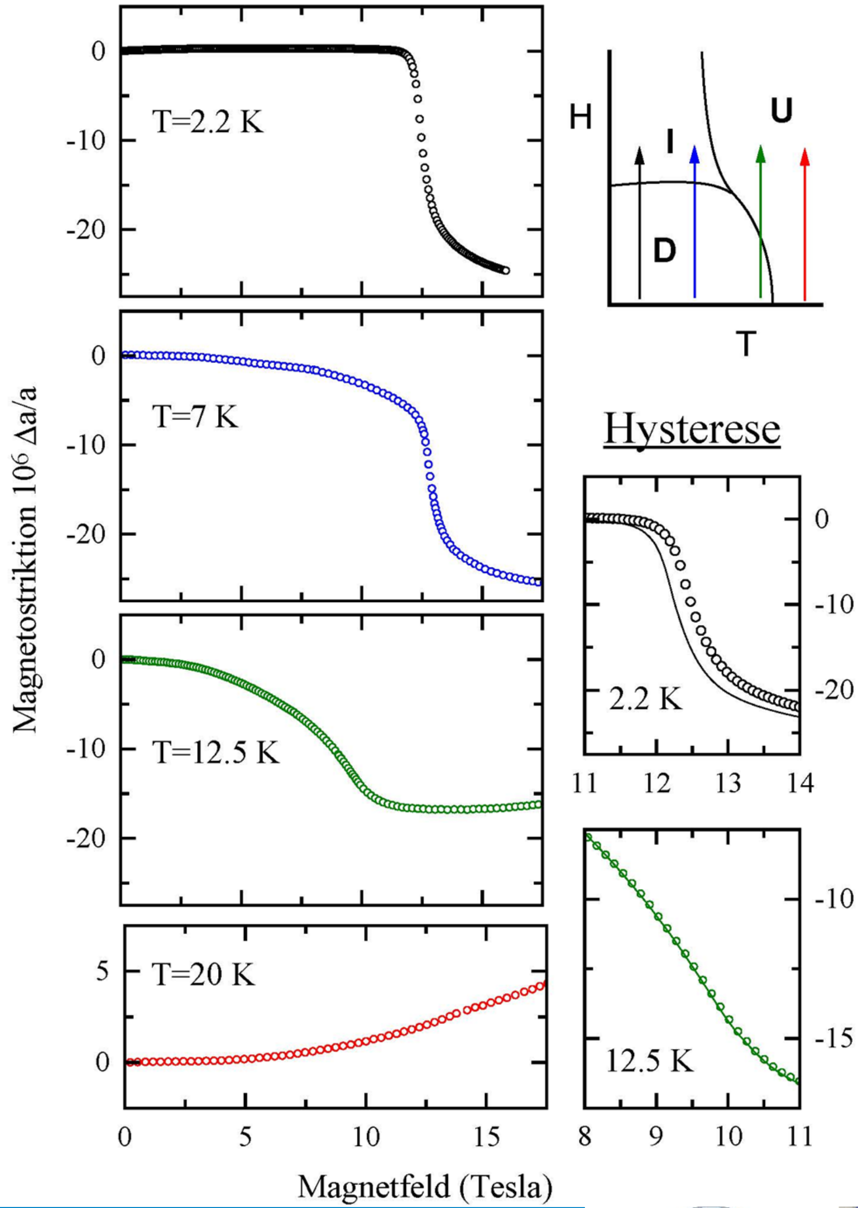
$Q_n = (-1)^n Q_0$



Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

$CuGeO_3$ in a Magnetic Field

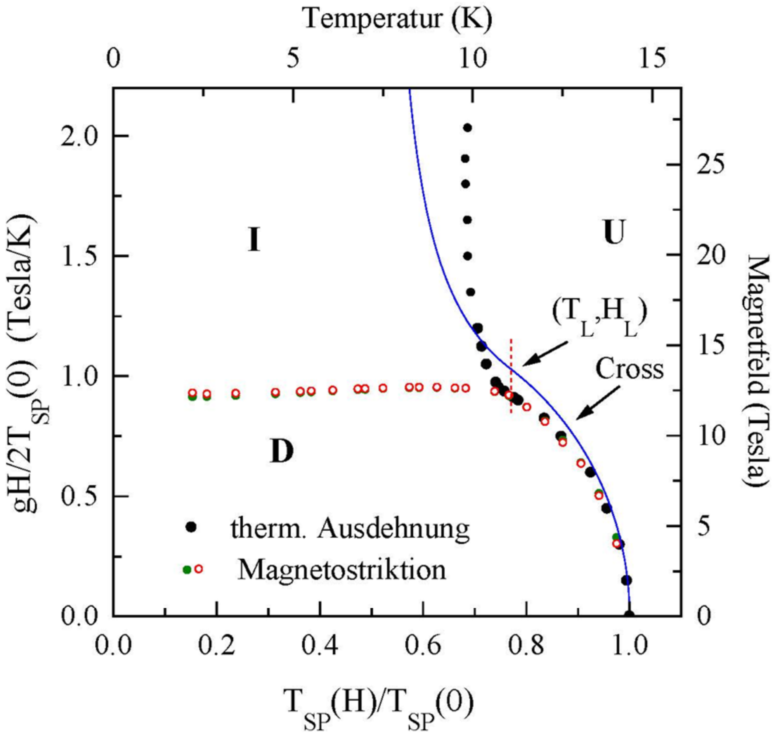


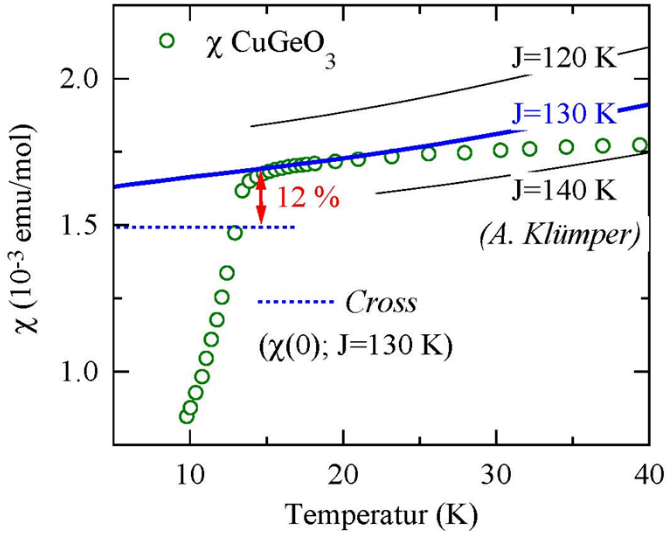


Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

TL et al. PRB **55**, 5914 (1997)
TL et al. PRL **81**, 148 (1998)

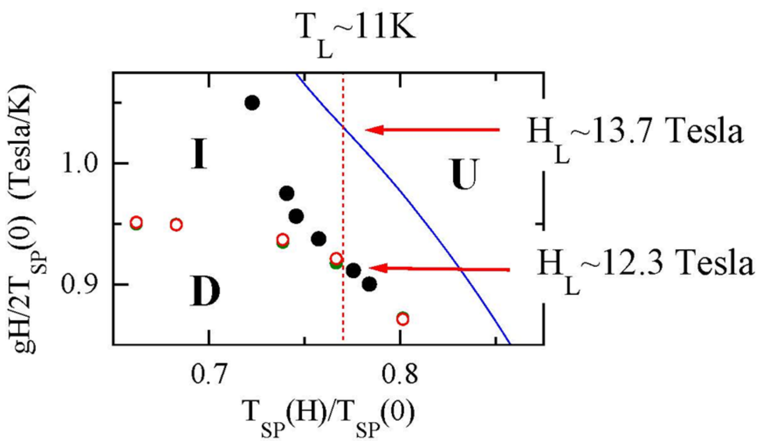
$CuGeO_3$ in a Magnetic Field



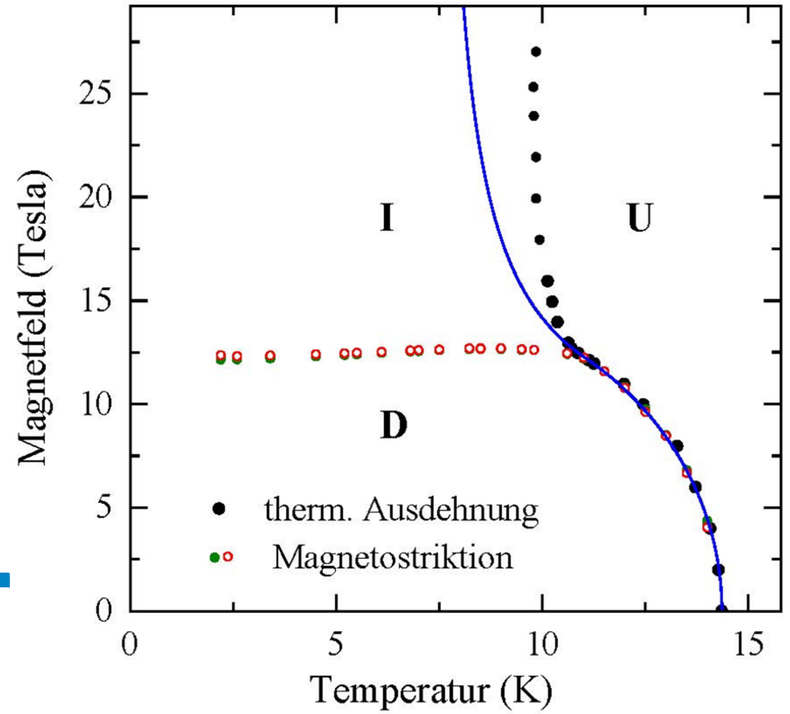


Field axis scales with χ ~ χ(0)

Cross PRB **20**, 4606 (1979)



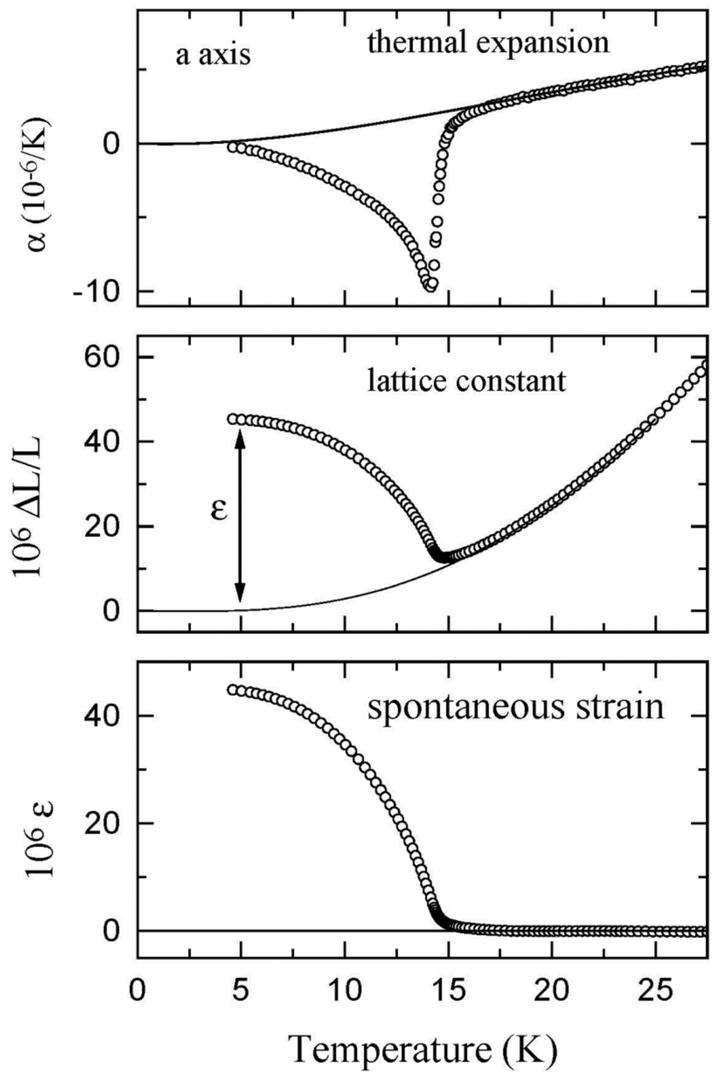
⇒ renormalize field axis by χ(T_{SP})



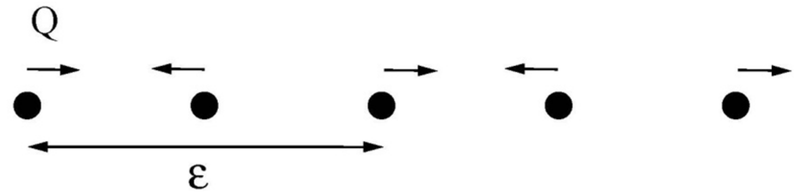
Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

TL et al. PRB **55**, 5914 (1997)
TL et al. PRL **81**, 148 (1998)

Spontaneous Strains & Order Parameter



TL et al. PRB **55**, 5914 (1997)

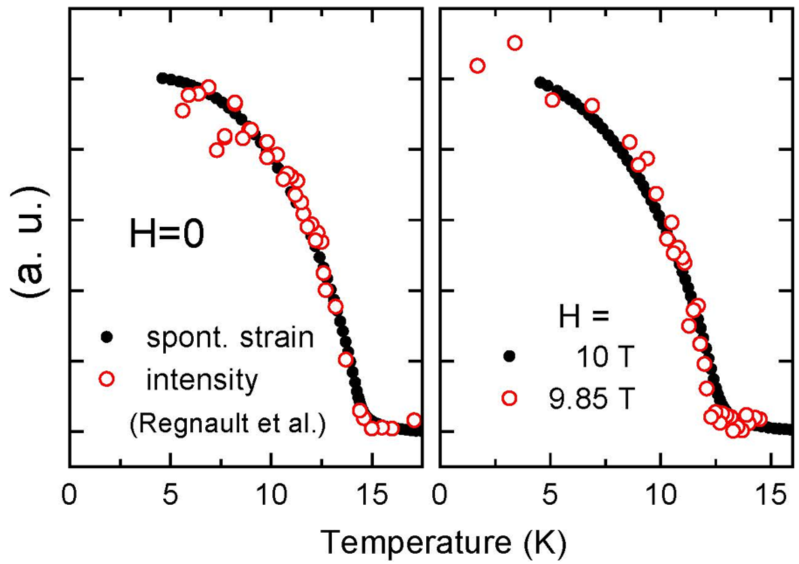


Landau–expansion:

$$f = f_0 + a\,Q^2 + b\,Q^4 + \ldots + \mu\,\epsilon\,Q^2 + \frac{1}{2}\,c\,\epsilon^2$$

$$\frac{\partial f}{\partial \epsilon} = 0 \quad \Rightarrow \quad \epsilon = -\frac{\mu}{c}\,Q^2$$

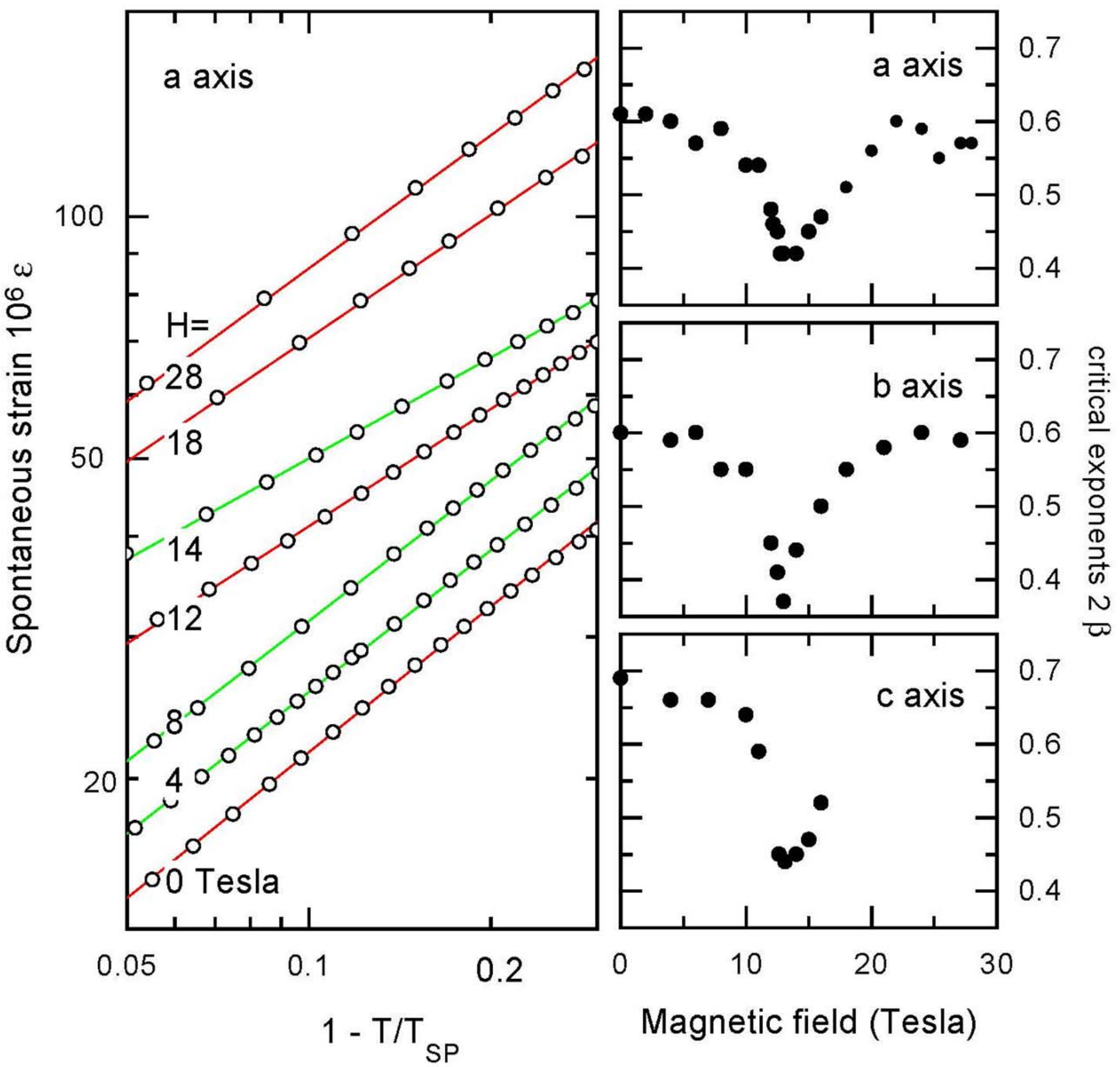
$\epsilon \propto Q^2 \propto$ intensity of superstructure reflections

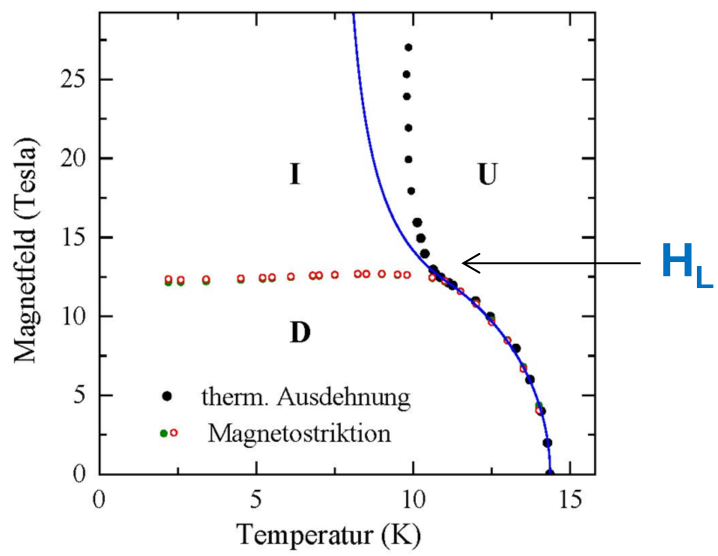


Regnault et al.
PRB 53, 5579 (1996)

Critical exponents of $CuGeO_3$

$$T \to T_{SP}: \quad Q \propto \left(1 - \frac{T}{T_{SP}(H)}\right)^{\beta} \quad \Rightarrow \quad \epsilon \propto Q^2 \propto \left(1 - \frac{T}{T_{SP}(H)}\right)^{2\beta}$$





D Phase: 3D Ising $\beta \sim 0.325$
(one-component OP: amplitude)

I Phase: 3D XY $\beta \sim 0.325$
(two-componet OP: amplitude & phase)

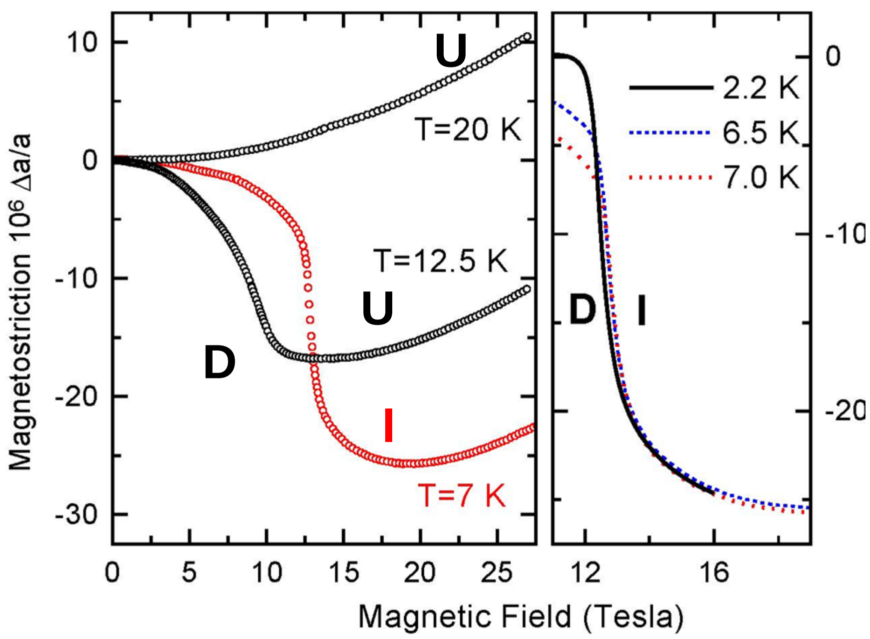
$H \sim H_L$: $\beta \sim 0.16$
(Lifshitz critical behavior)

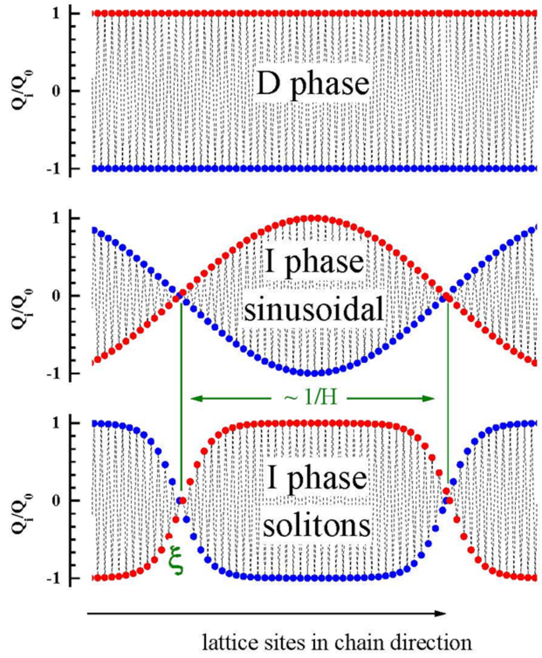
Vysochansky & Slivka
Sov. Phys. Usp. **35**, 123 (1992)

TL et al. PRB **55**, 5914 (1997)

Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

Incommensurate Phase of $CuGeO_3$





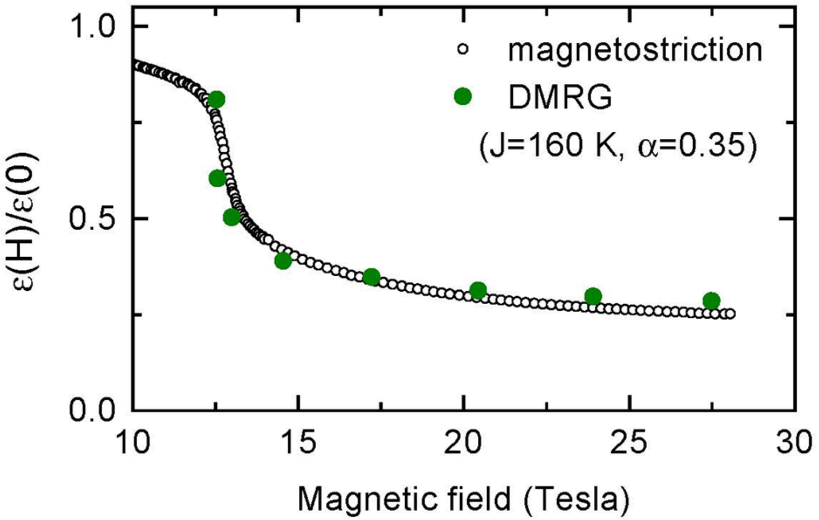
D phase:

$$\epsilon \propto Q_0^2$$

I phase:

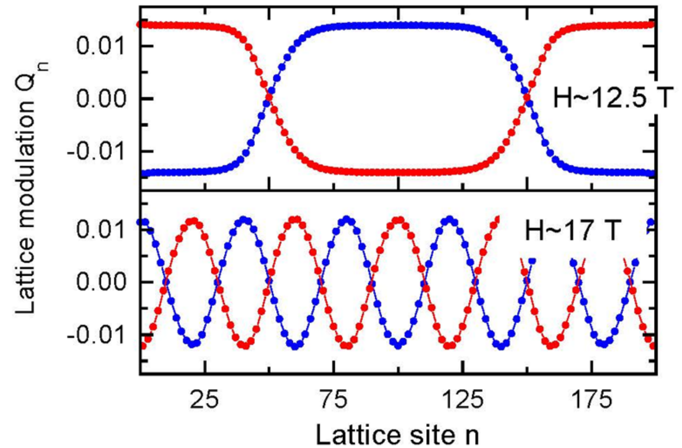
$$\epsilon \propto \langle Q_n^2 \rangle$$

$$\frac{\epsilon(H)}{\epsilon(0)} = \frac{\langle Q_n^2(H) \rangle}{Q_0^2}$$



Numerical results: (DMRG)

Parameters: J=160 K; $\frac{J'}{J} = 0.35$; $\Delta E = 23$ K



⇒ from a
soltion lattice
to a
sinusoidal modulation

Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

TL et al. PRL **81**, 148 (1998)

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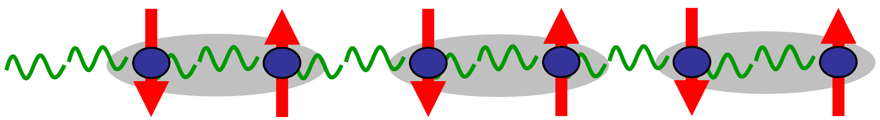
Summary I: Spin-Peierls System $CuGeO_3$

- $CuGeO_3$ was the 1st inorganic Spin-Peierls material (α-NaV_2O_5, TiOCl, …)
- strong magnetoelastic coupling & pronounced frustration J_{NN} *vs.* J_{NNN}
  (thermodynamics $J_{NNN} \sim 0.35\, J_{NN}$; Raman spectroscopy $J_{NNN} \sim 0.24\, J_{NN}$)
- universal H-T phase diagram
  (U/D agrees perfectly with theory, but systematic deviation for U/I boundary)
- incommensurate phase: soliton lattice => sinusoidal modulation
- (doping effects, e.g. (Cu,Zn)GeO_3 => coexisting Néel & Spin-Peierls order)

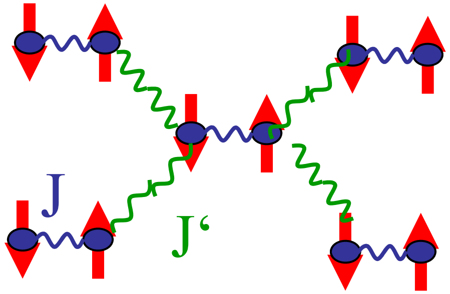
Remember: spin chains should be viewed as coupled singlet dimers !

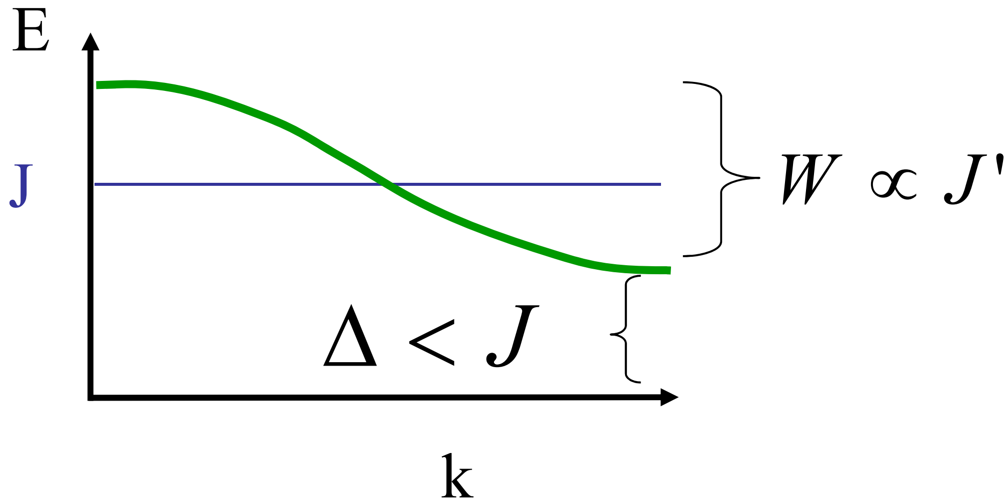


What about quantum phase transitions in high magnetic fields ?

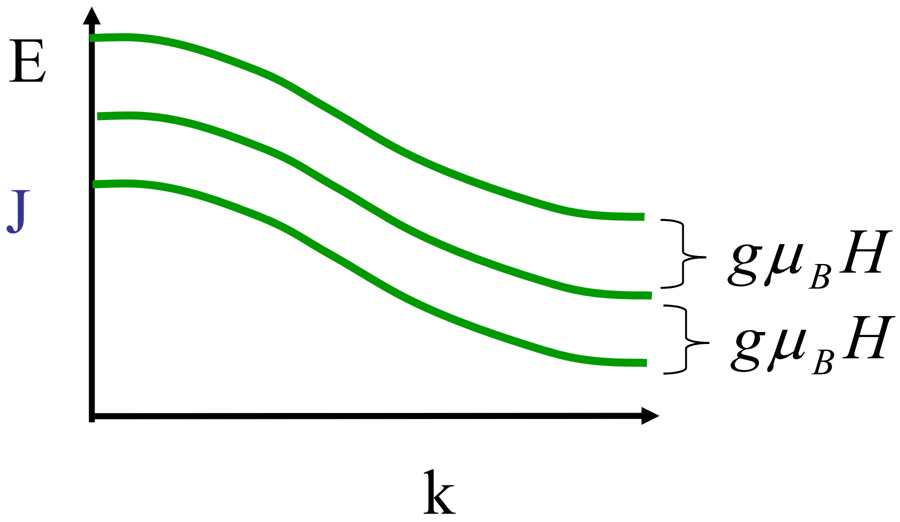
Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

Coupled spin-dimers in a magnetic field

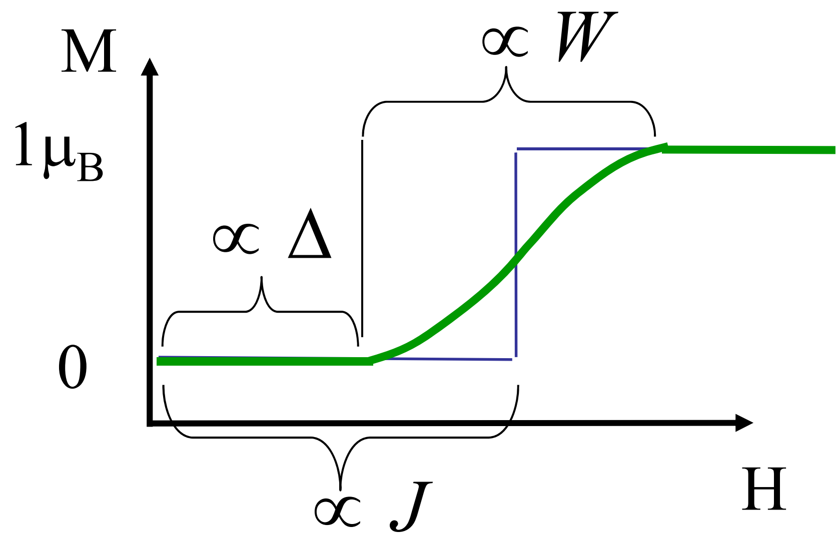




H > 0

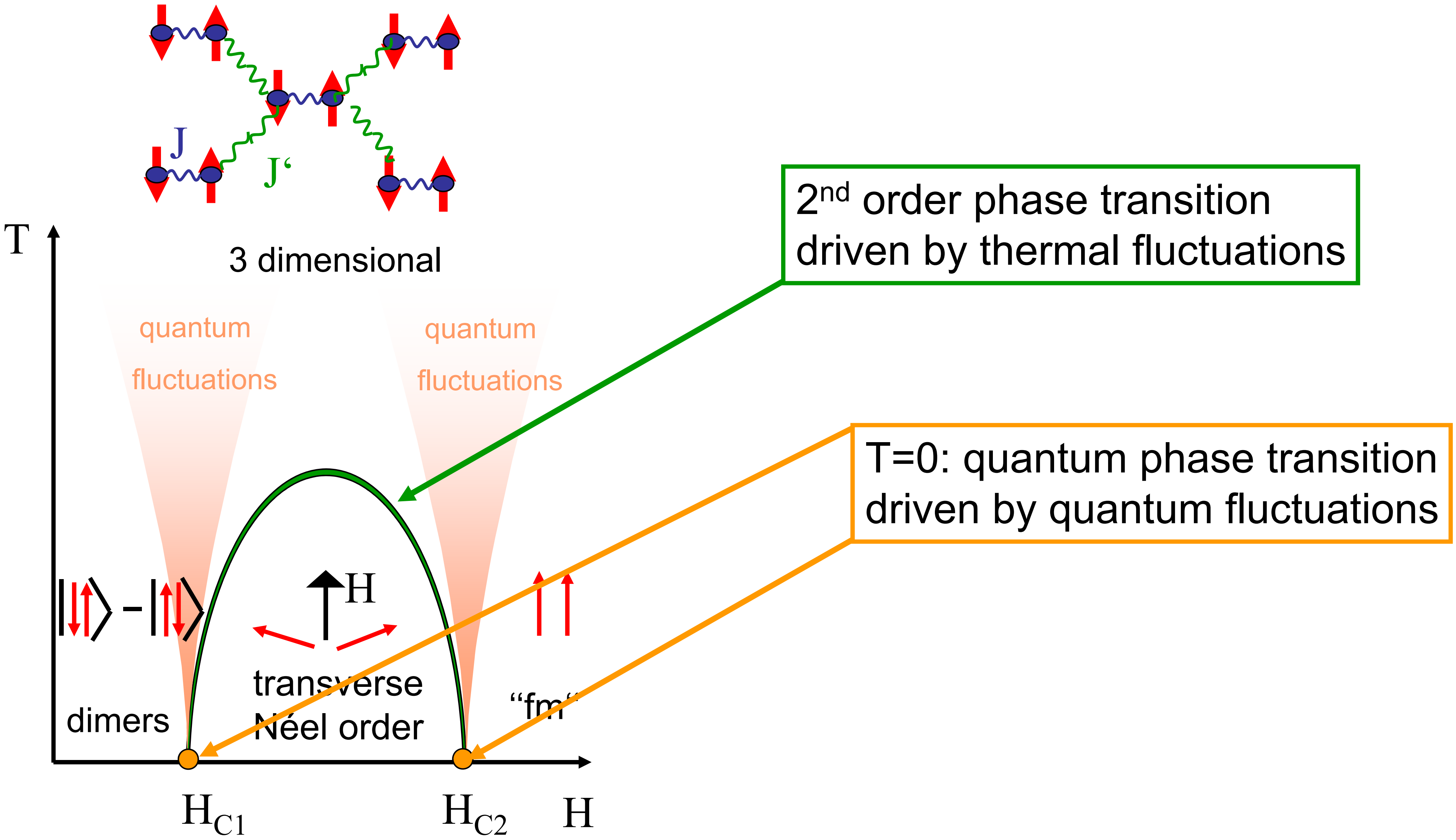


Magnetization at T -> 0



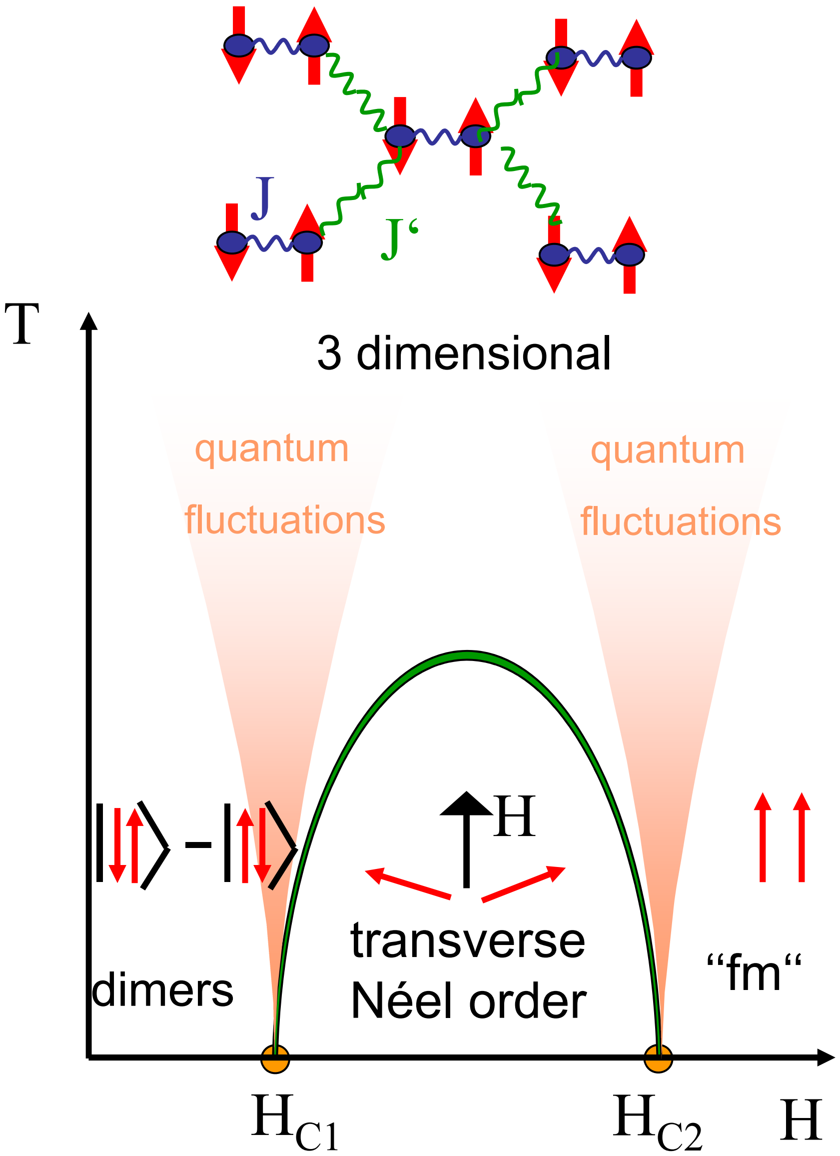
Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

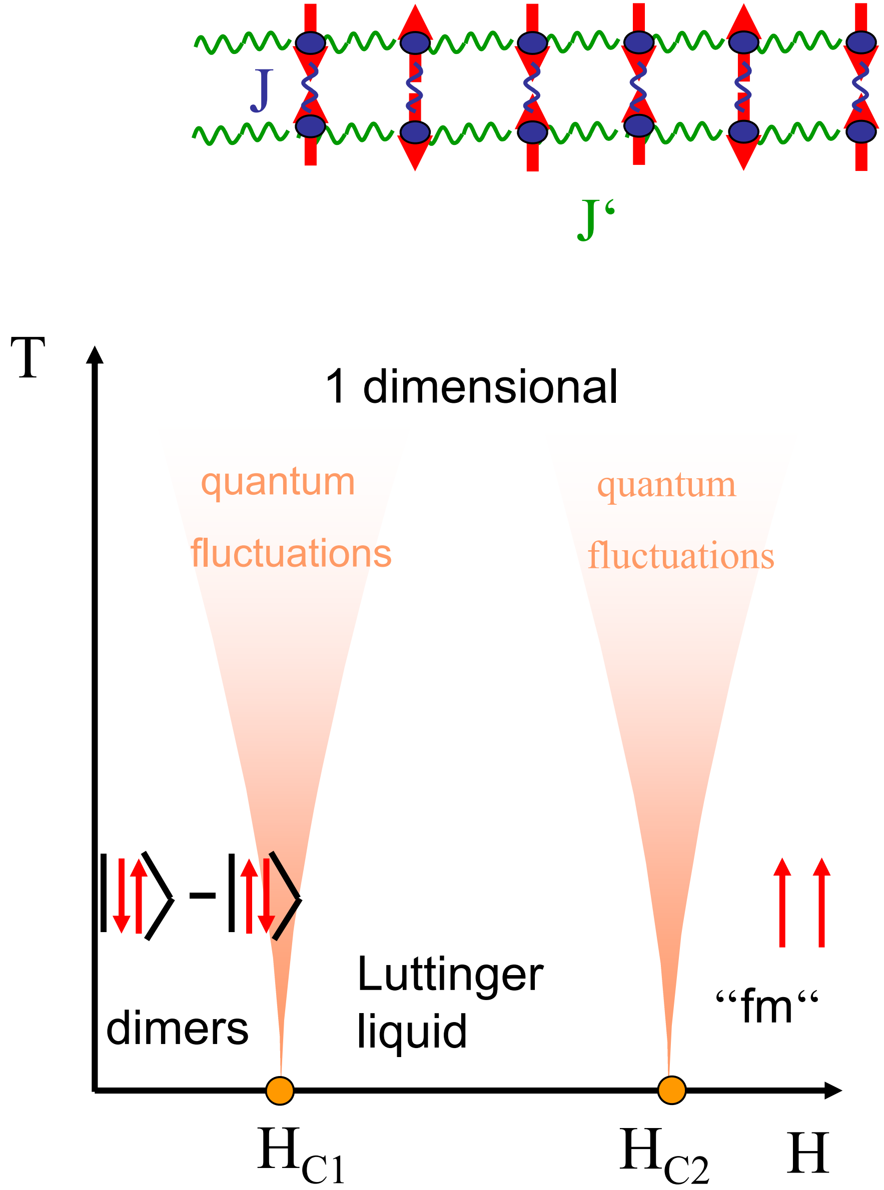
Low-dimensional coupled spin-dimer models



Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

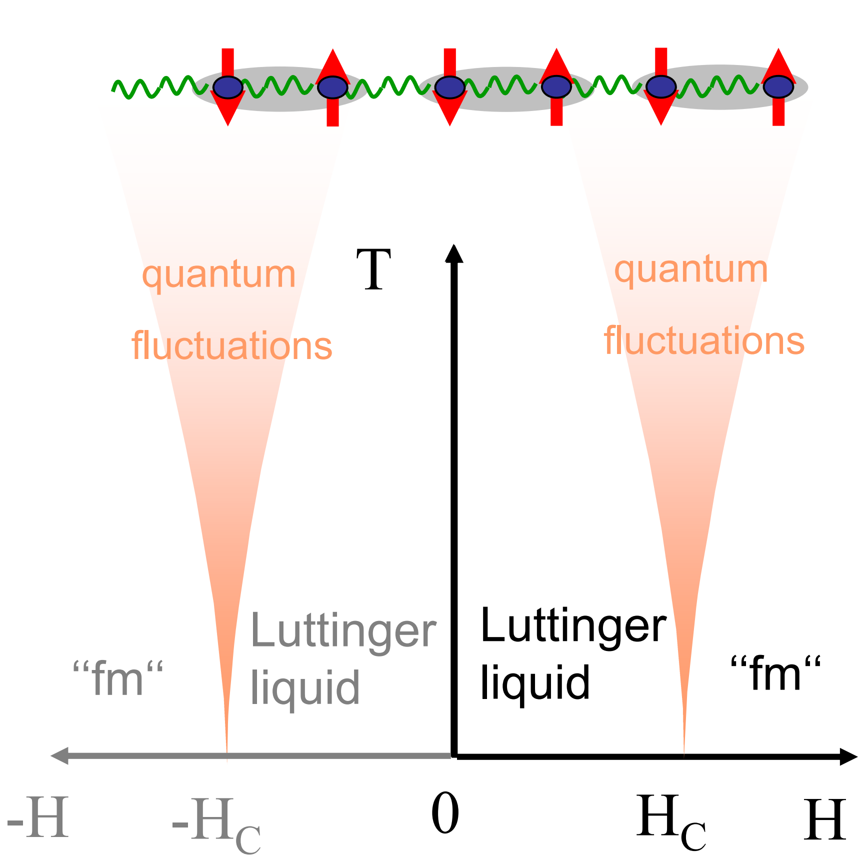
Low-dimensional coupled spin-dimer models

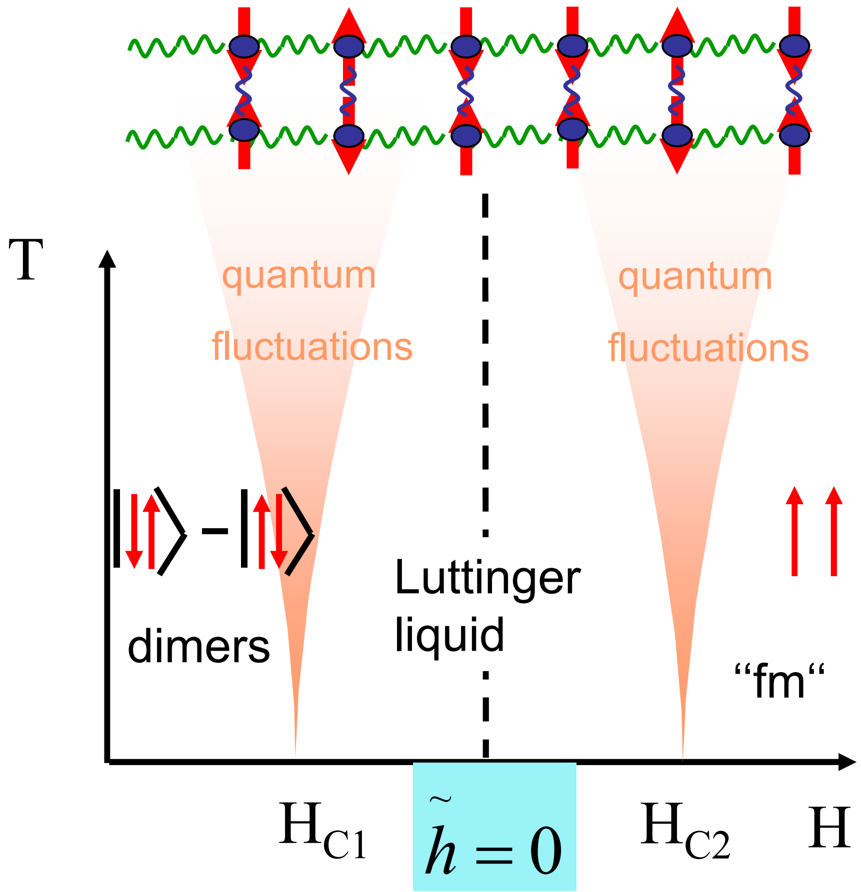




Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

Spin ½ chains vs. spin ½ ladders





$$H = J\sum_i \left(S_i^x S_{i+1}^x + S_i^y S_{i+1}^y + \lambda\, S_i^z S_{i+1}^z\right) - h\sum_i S_i^z$$

$$h = g\mu_B H$$

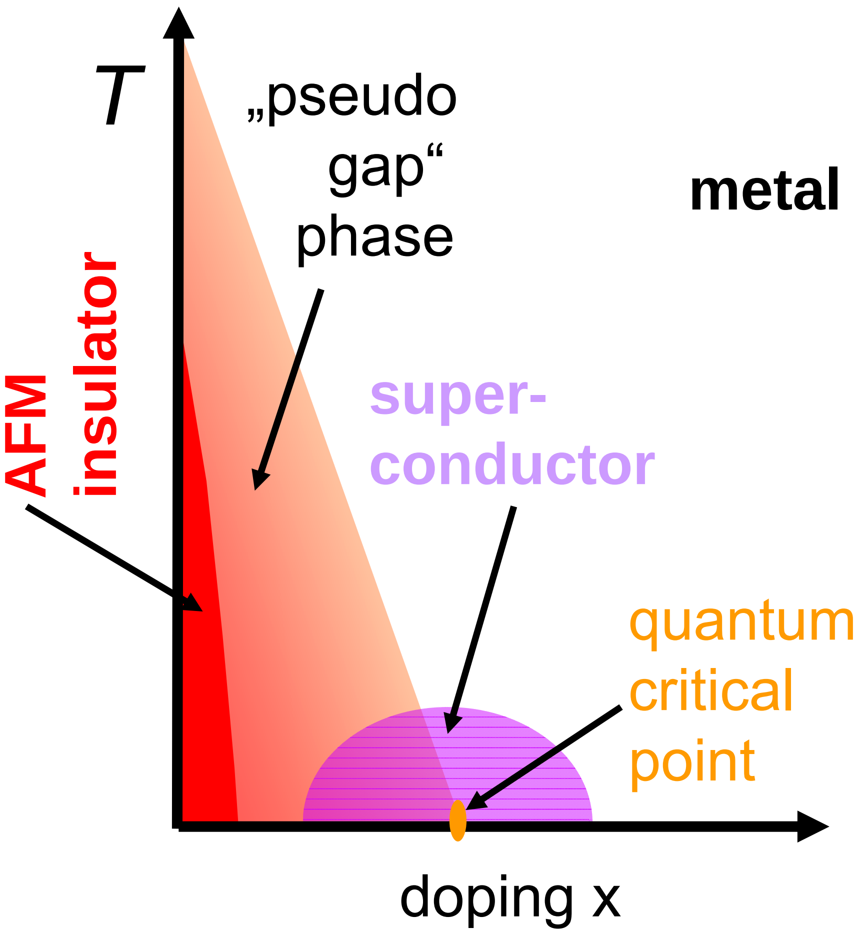
$$H = J_\parallel \sum_i \left(S_i^x S_{i+1}^x + S_i^y S_{i+1}^y + \frac{1}{2} S_i^z S_{i+1}^z\right) - \tilde{h}\sum_i S_i^z$$

$$\tilde{h} = g\mu_B H - \left(J_\parallel + J_\perp / 2\right) \qquad \lambda = 1/2$$

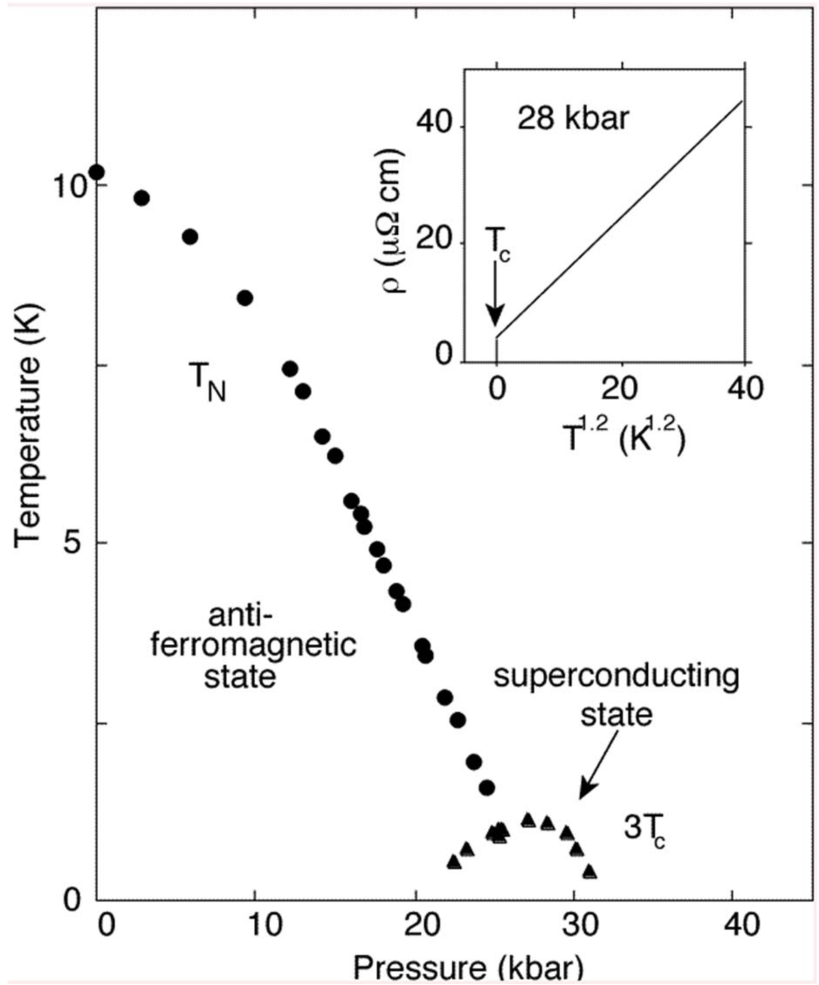
Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

Quantum Phase Transitions

High-Tc cuprates



Heavy fermions



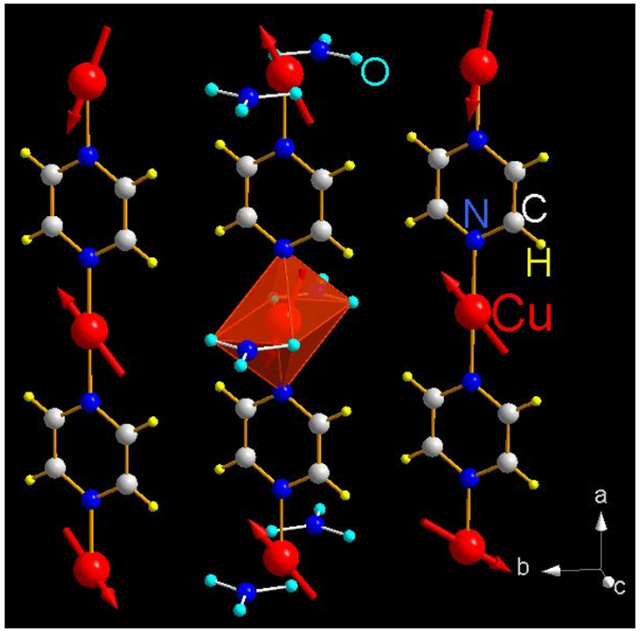
Mathur *et al*. Nature 394, 39 (1998)

Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

Copper pyrazine dinitrate „CuPzN“

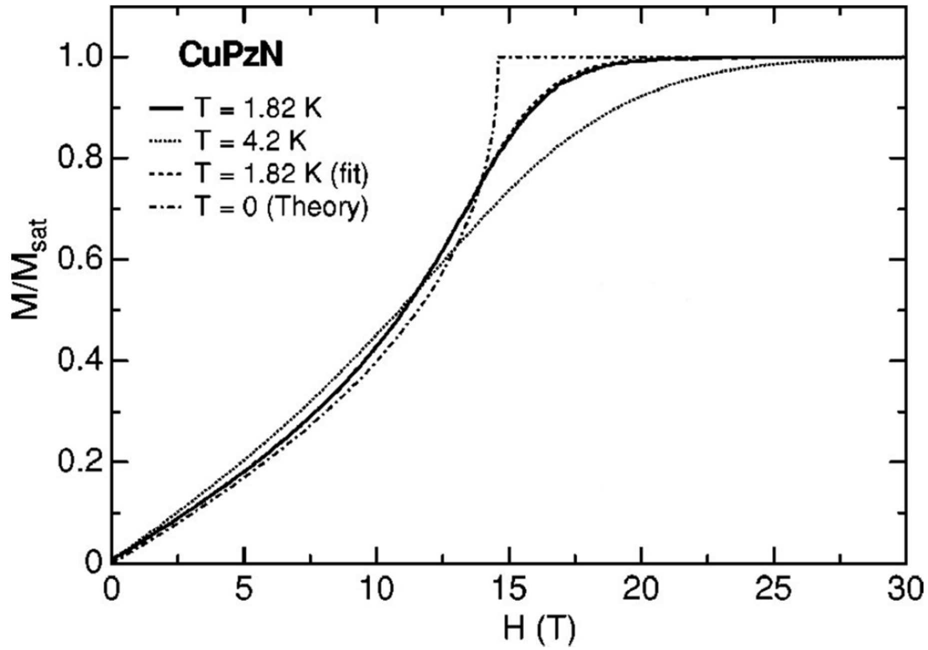
$Cu(C_4H_4N_2)(NO3)_2$

S = 1/2 chains || *a*



(picture from M. Valldor)

Magnetization



antiferromagnetic order at $T_N \approx 100$ mK
(measured by µSR)

saturation critical field: $H_c \approx 14$ T

$\Rightarrow$

$J = 10.3$ K
$J_\perp / J \sim 10^{-3}$

Hammar et al. PRB **59**, 1008 (1999)
Lancaster *et al.*, PRB **73**, 20410 (2006)

Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
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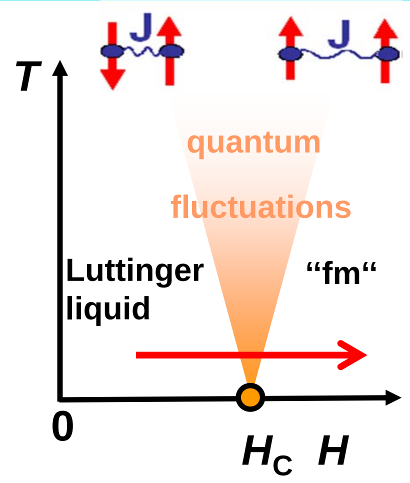
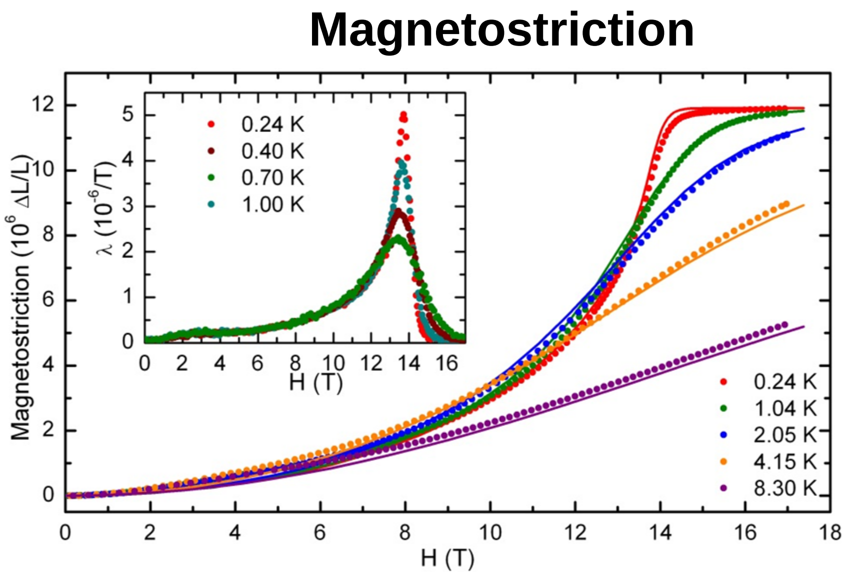
Copper pyrazine dinitrate „CuPzN“



⇒ no phase transitions above 100 mK !



$$\text{NN Heisenberg: } \mathrm{H} = J\sum_j \vec{S}_j \vec{S}_{j+1} \quad \Rightarrow \quad \text{entropy} \quad S = S\left(\frac{T}{J}\right) \quad \Rightarrow \quad \alpha_i^{mag} = \frac{1}{V}\frac{\partial \ln J}{\partial p_i} C_{mag} \quad \text{for } H = 0$$

$$\text{more general: } \quad \alpha_i^{mag} = \gamma_i \frac{\partial D(H,T)}{\partial T} \quad \& \quad \lambda_i = \frac{1}{L_i}\frac{\partial L_i}{\partial H} = \gamma_i \frac{\partial D(H,T)}{\partial H}$$

$$\text{with: } \quad \gamma_i = \frac{1}{V}\frac{\partial J}{\partial p_i} \quad \& \quad \text{spin-spin correlator } \; D(H,T) = \frac{1}{N}\sum_j \vec{S}_j \vec{S}_{j+1}$$

⇒ lines are calculation with

$$J = 10.3\,\mathrm{K} \quad \& \quad \frac{\partial \ln J}{\partial p_i} = 2.4\,\%/\mathrm{GPa}$$

here: $\alpha^{pho} << \alpha^{mag}$

Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

Anfuso, …,TL, et al. **77**, 235113 (2008)
Rohrkamp, …, TL. JPCS **200**, 012169 (2010)

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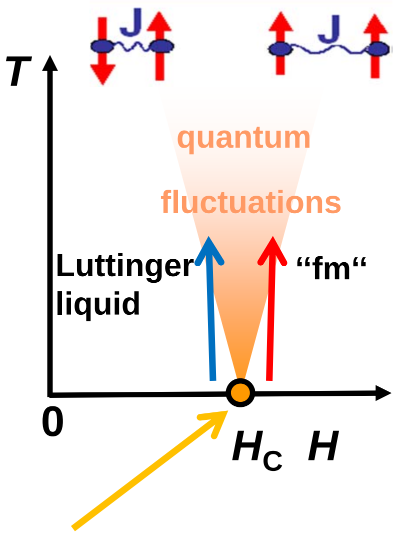
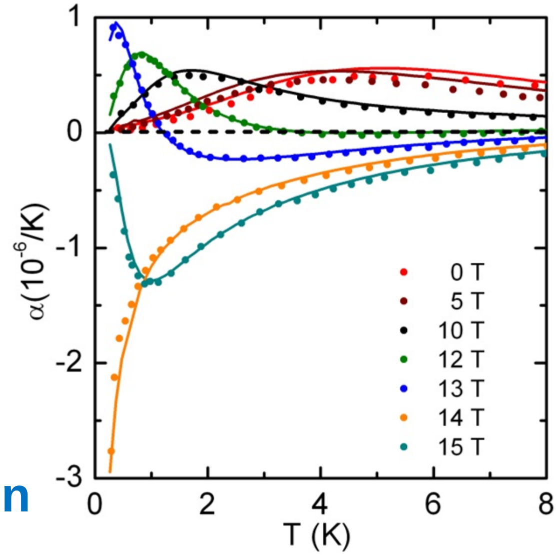
Copper pyrazine dinitrate „CuPzN“

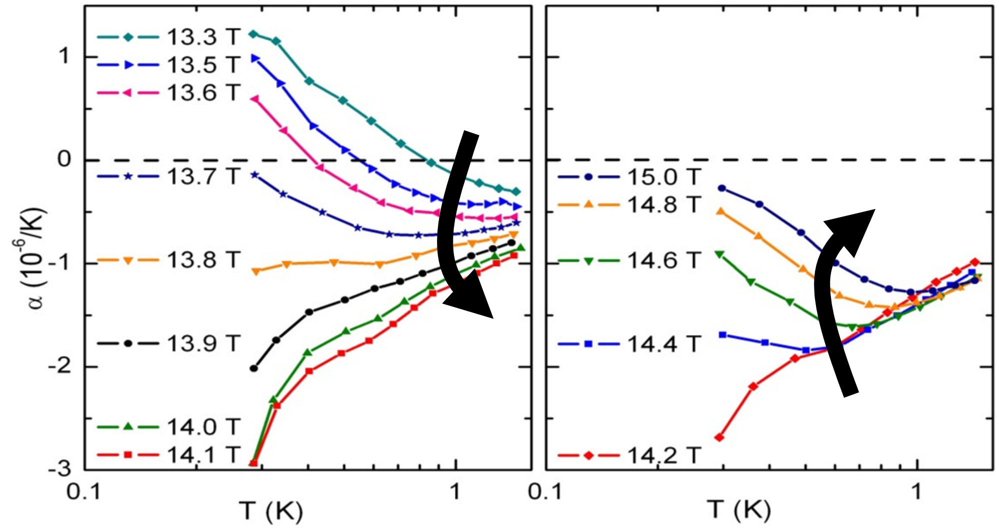


Quantum phase transition

thermal expansion



close to quantum phase transtion



Expected behavior:

In general:

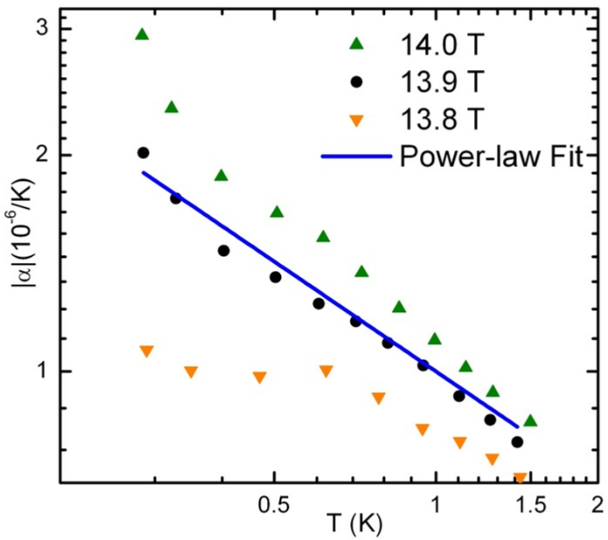
⇒ sign change of α and divergence of

Grüneisen parameter $\Gamma = \alpha / c$

In 1 D:

⇒ sign change and divergence of α

L. Zhu *et al.*, PRL **91**, 066404 (2003)



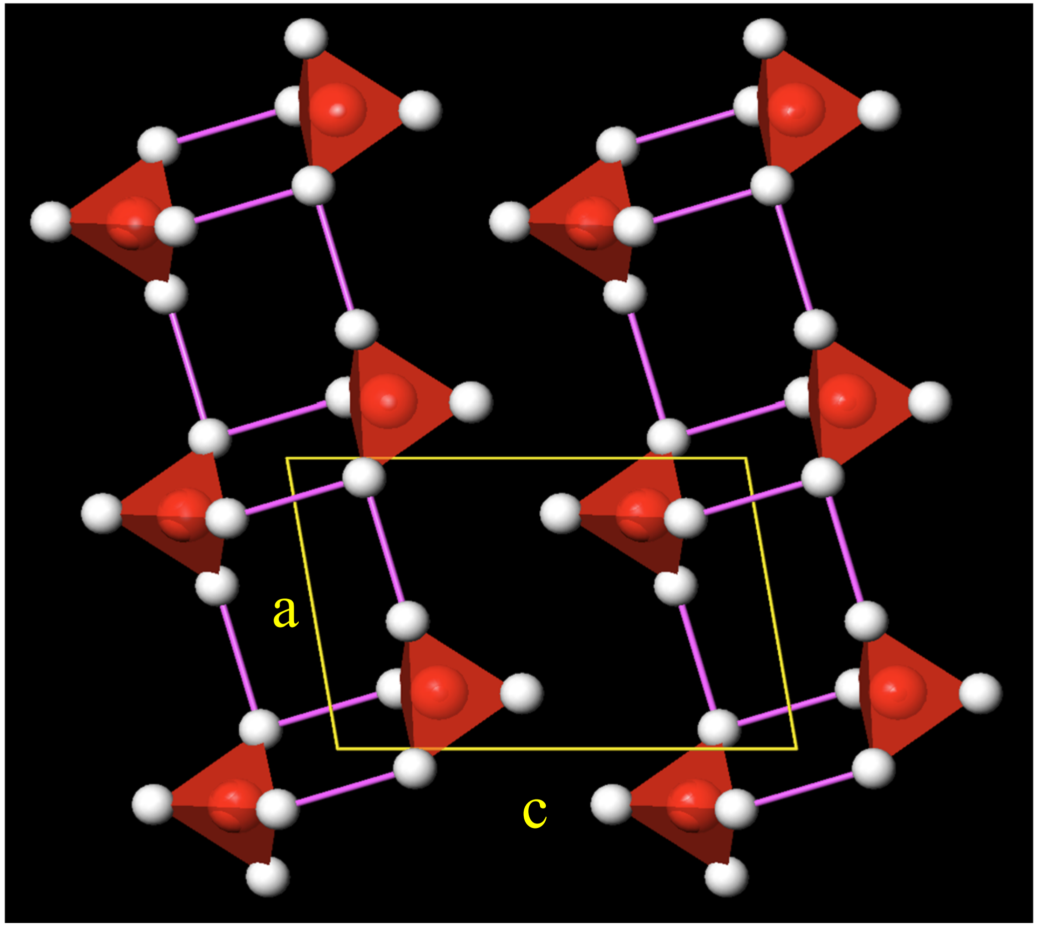
$$|\alpha| \propto 1/\sqrt{T}$$

Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

Anfuso, …,TL, *et al.* **77**, 235113 (2008)
Rohrkamp, …, TL. JPCS **200**, 012169 (2010)

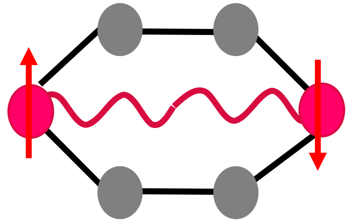
$(C_5H_{12}N)_2CuBr_4$: a spin-ladder system

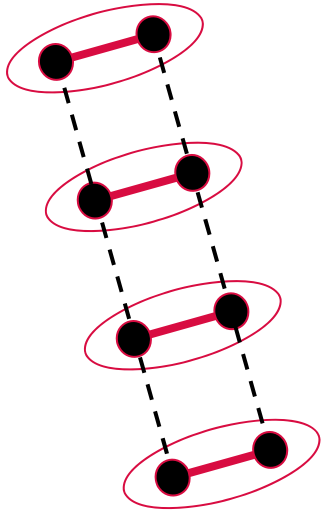
Bis(piperidinium)-$CuBr_4$ „BPCB“



(picture from C. Rüegg)

CuBr rungs



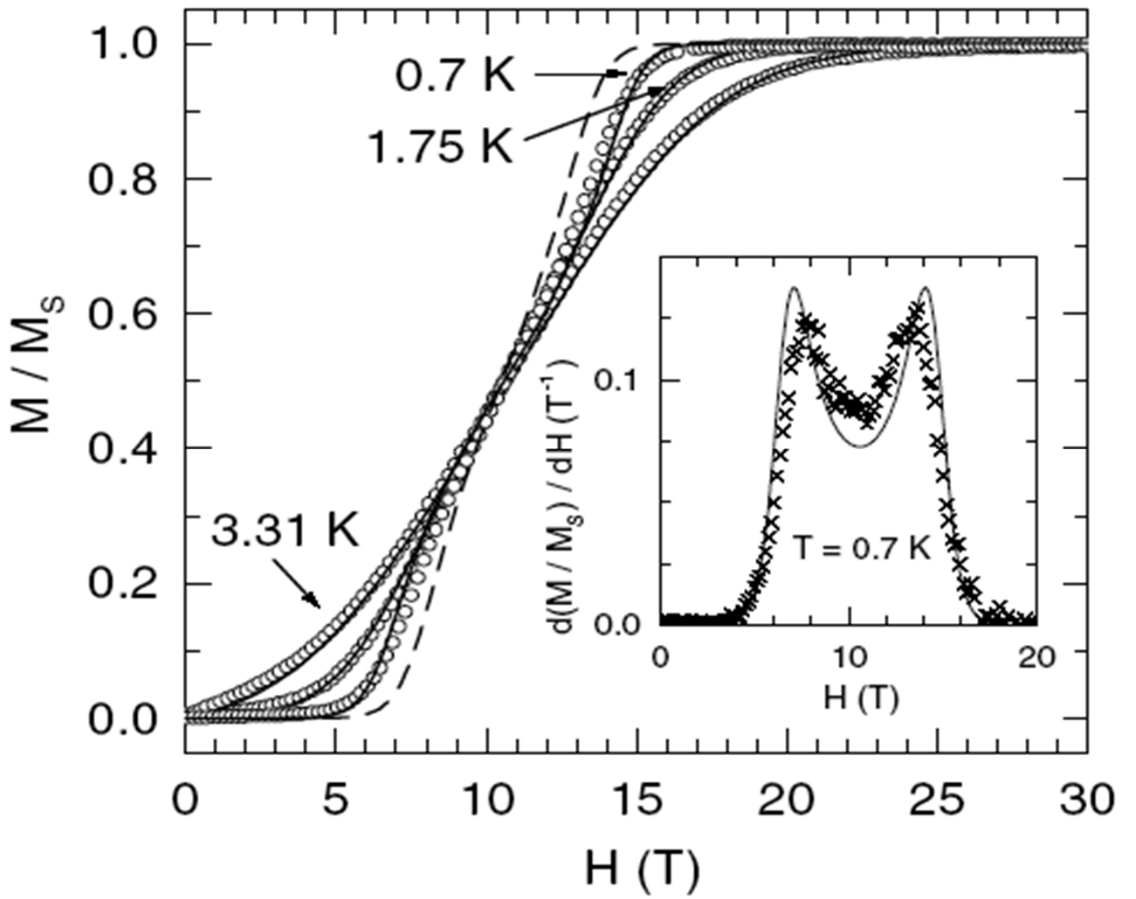


rung coupling J_r ~ 13 K
leg coupling J_l ~ 3.5 K

B.R. Patyal *et al.*, PRB **41**, 1657 (1990)
B.C. Watson *et al.*, PRL **86**, 5168 (2001)
C. Rüegg *et al.*, PRL **101,** 247202 (2008)
M. Klanjsek *et al.*, PRL **101**, 137207 (2008)
…

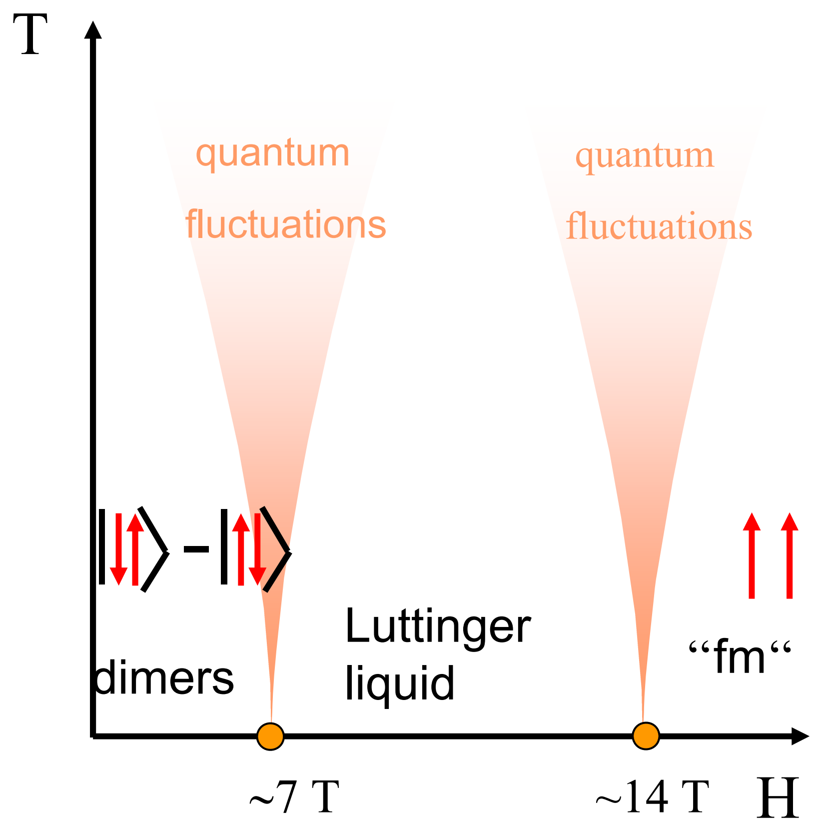
$(C_5H_{12}N)_2CuBr_4$: a spin-ladder system

Magnetization



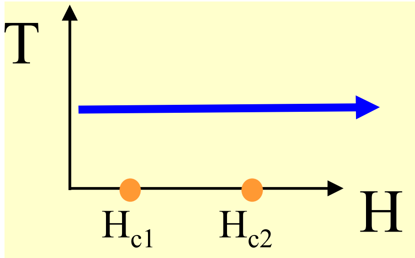
B.C. Watson *et al.*, PRL **86**, 5168 (2001)

Phase diagram

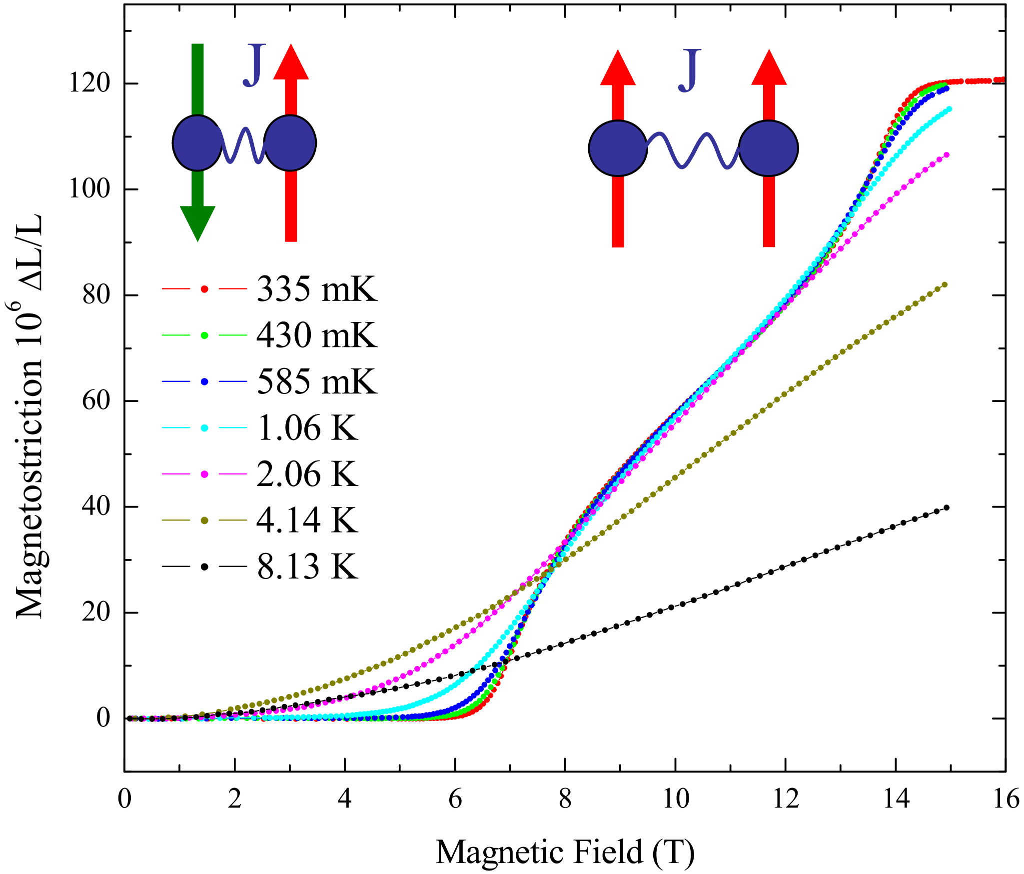


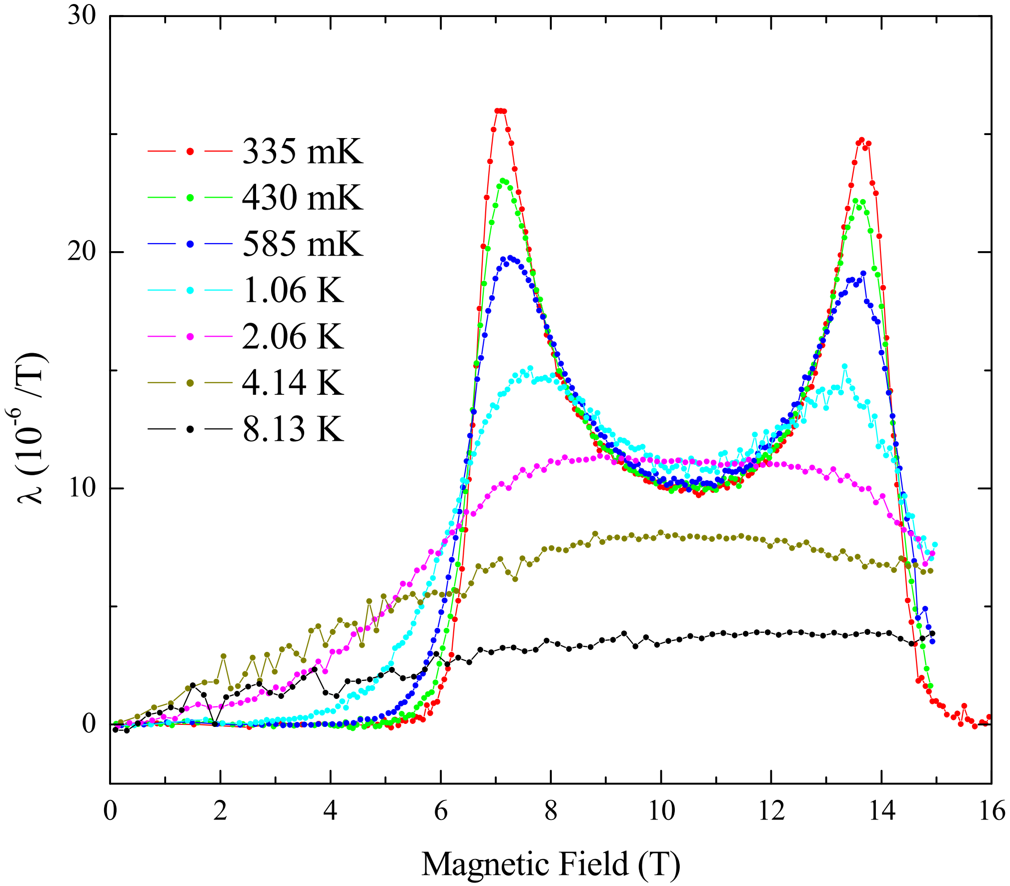
$(C_5H_{12}N)_2CuBr_4$: magnetostriction || rungs

$$\frac{\Delta L(H)}{L} = \frac{L(H) - L(0)}{L}$$



$$\lambda = \frac{1}{L}\frac{\partial \Delta L(H)}{\partial H}$$



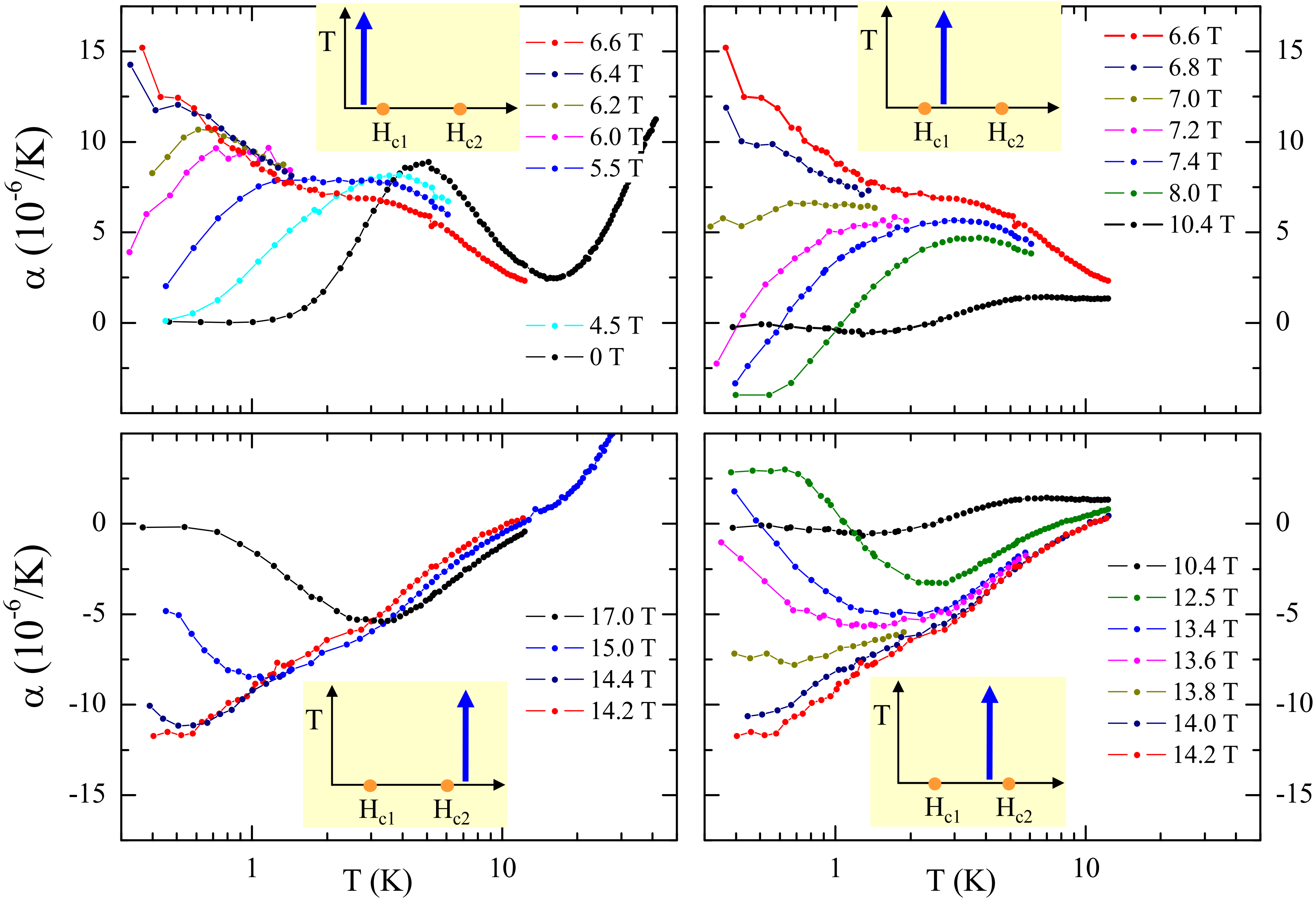


Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

TL *et al.* PRL **100,** 067208 (2008)

SFB 608

$(C_5H_{12}N)_2CuBr_4$: thermal expansion || rungs



TL *et al*. PRL **100,** 067208 (2008)

Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
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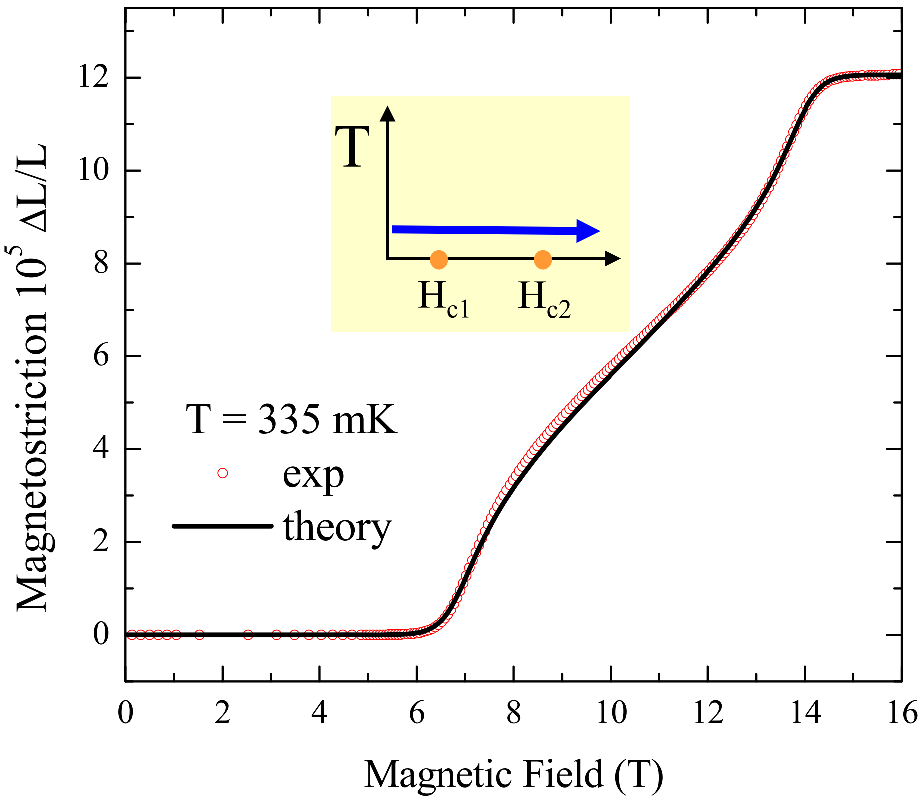
$(C_5H_{12}N)_2CuBr_4$: theory vs. experiment

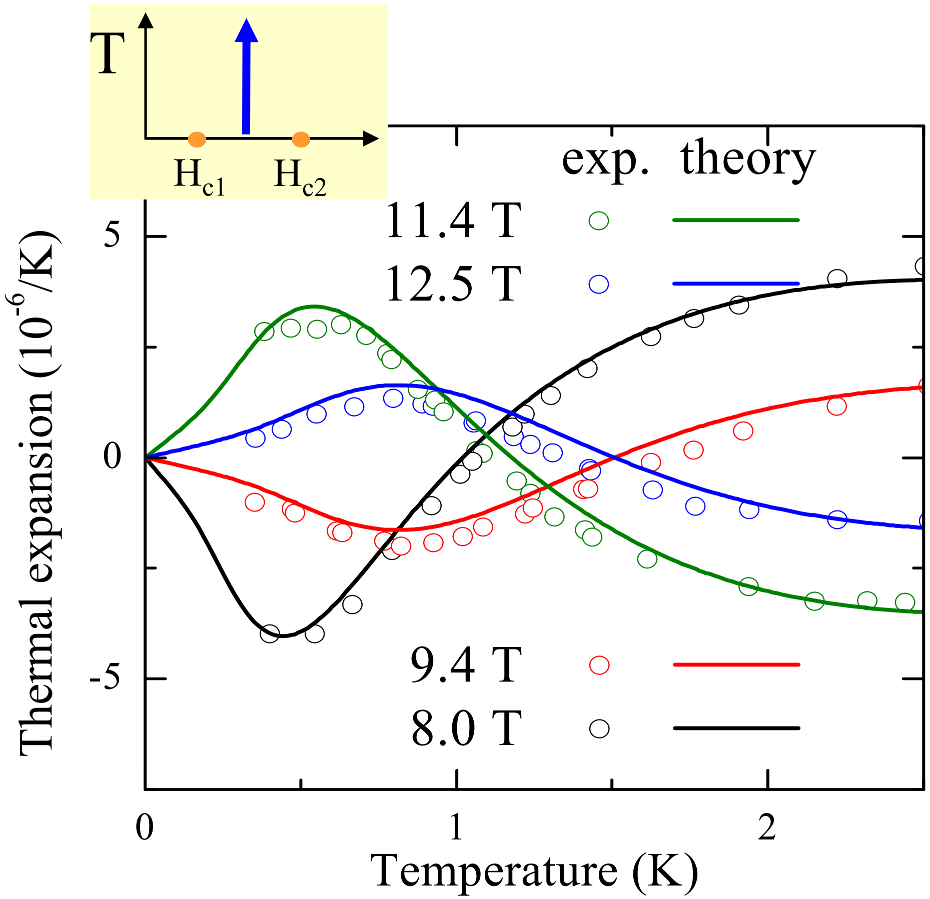
Simplified model: Jordan-Wigner fermions

$$H_{eff} = \int_{-\pi}^{\pi} \frac{dk}{2\pi} (2t \cos k - \mu)$$

=> 3 fit parameters: *t, position & prefactor*

=> calculate thermal expansion (**NO** additional parameter)

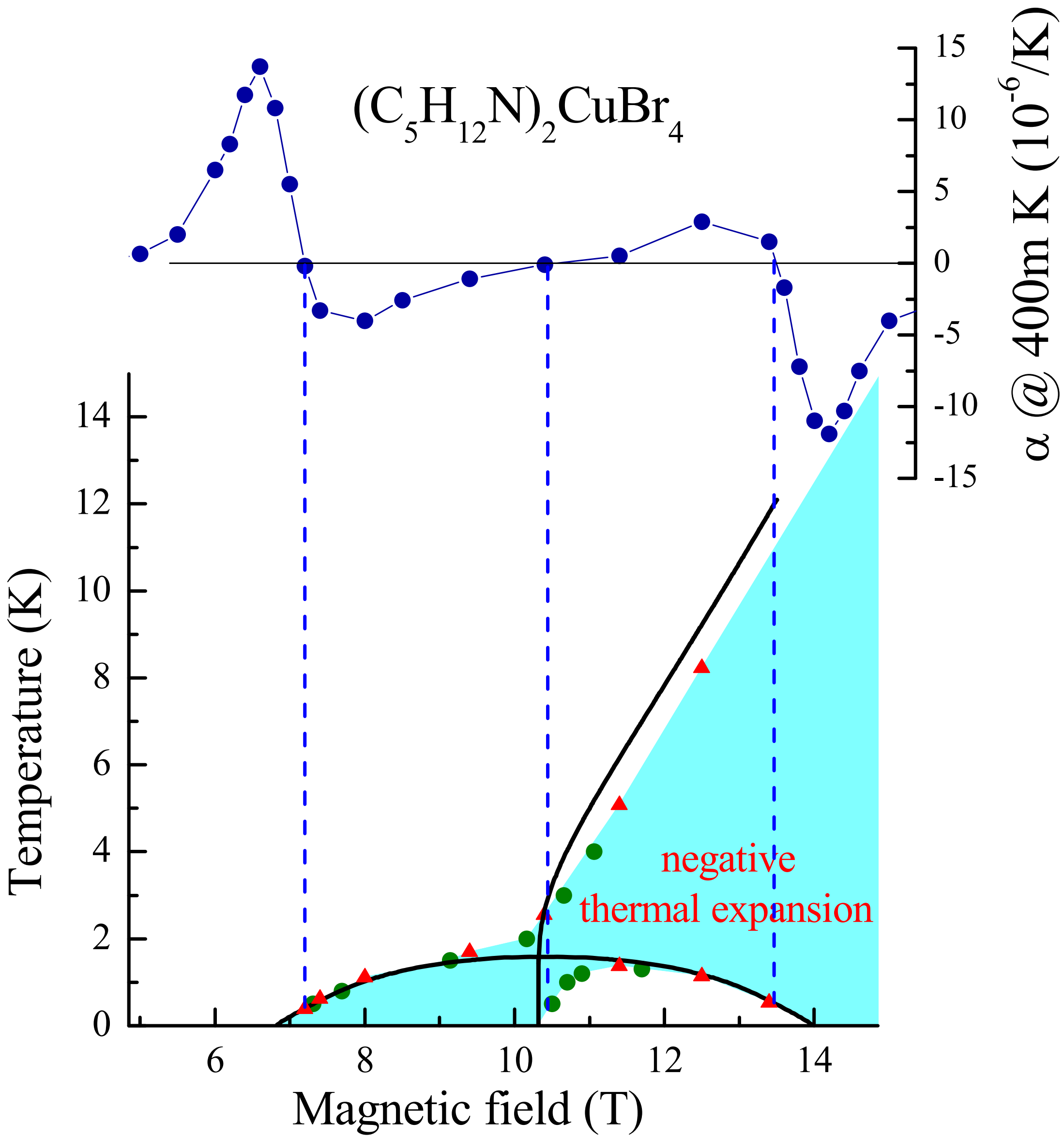


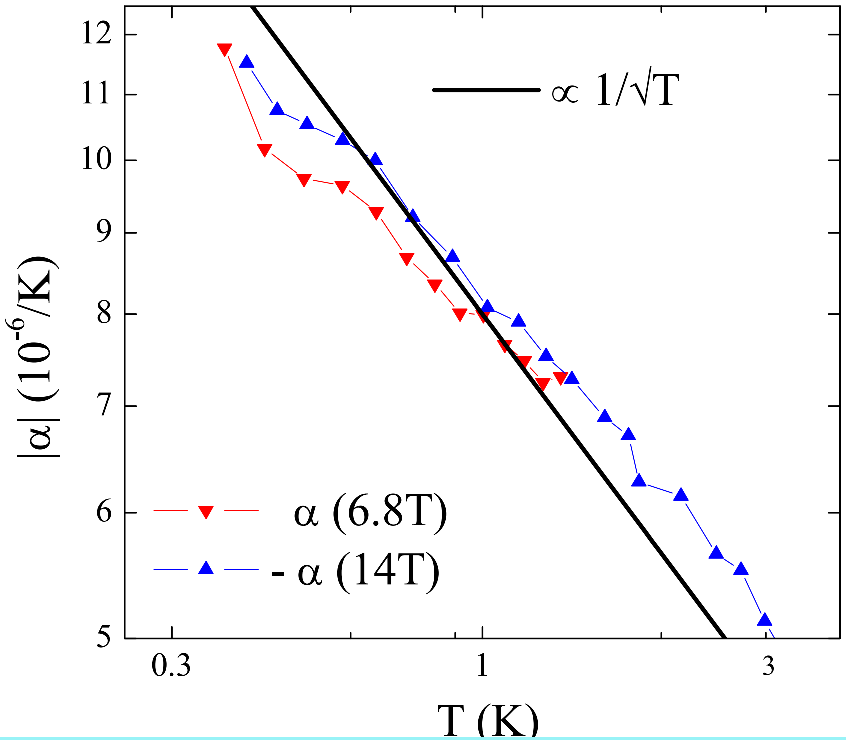


Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

TL *et al*. PRL **100,** 067208 (2008)

$(C_5H_{12}N)_2CuBr_4$: theory vs. experiment





⇒ All basic features of the experimental data are reproduced by the (too) simple model of free fermions !

Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

TL *et al*. PRL **100,** 067208 (2008)

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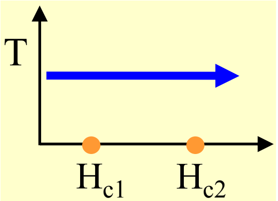
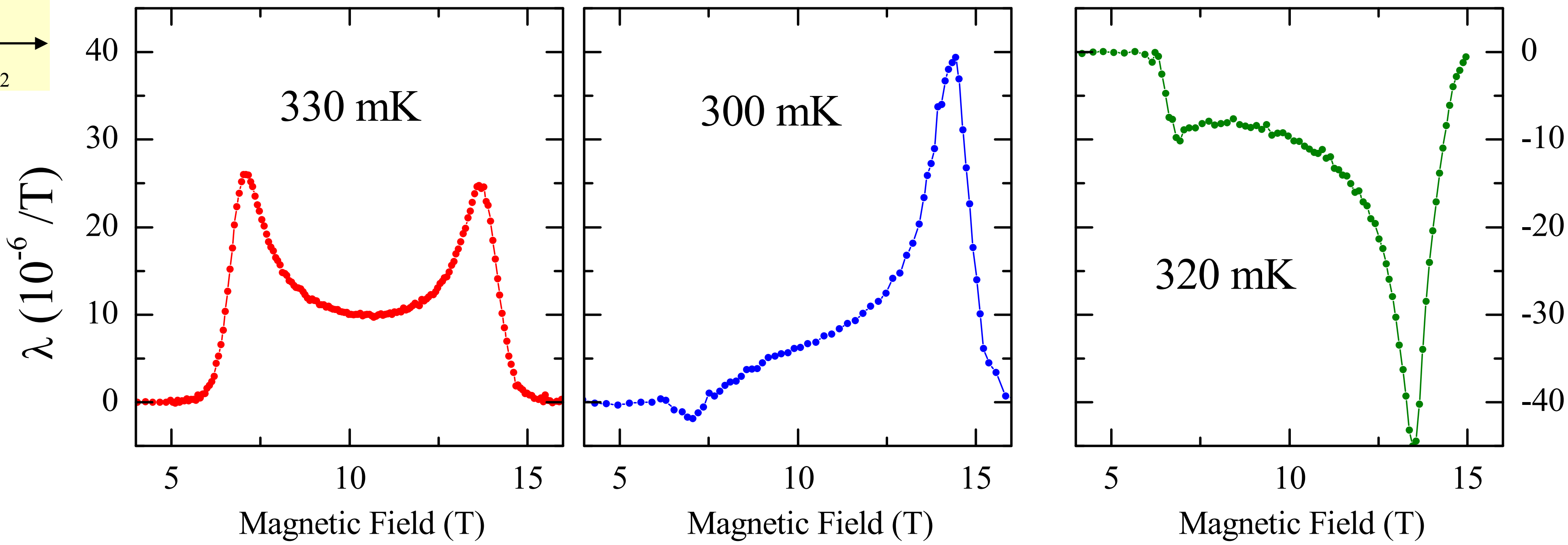
$(C_5H_{12}N)_2CuBr_4$: anisotropic magnetostriction



ΔL || rungs || legs ||b (perp. ladders)



$$H_{c1} = J_R - J_L$$
$$H_{c2} = J_R + 2J_L$$

$$\partial J_L / \partial p_R \approx 0 \qquad \partial J_R / \partial p_R > 0$$

$$\partial J_L / \partial p_L \approx \qquad \partial J_R / \partial p_L > 0$$

$$2 * \partial J_L / \partial p_b \approx \qquad \partial J_R / \partial p_b < 0$$

Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

TL *et al*. PRL **100,** 067208 (2008)
F. Anfuso, ..., TL, *et al*. PRB **77**, 235113 (2008)

SFB 608

Magnetostriction ⇔ spin-spin correlation

$$\lambda_n(T,H) = \gamma_n^R \frac{\partial D^R}{\partial H} + \gamma_n^L \frac{\partial D^L}{\partial H} \quad \text{with} \quad \gamma_n^{R,L} = \frac{1}{V}\frac{\partial J^{R,L}}{\partial p_n}$$

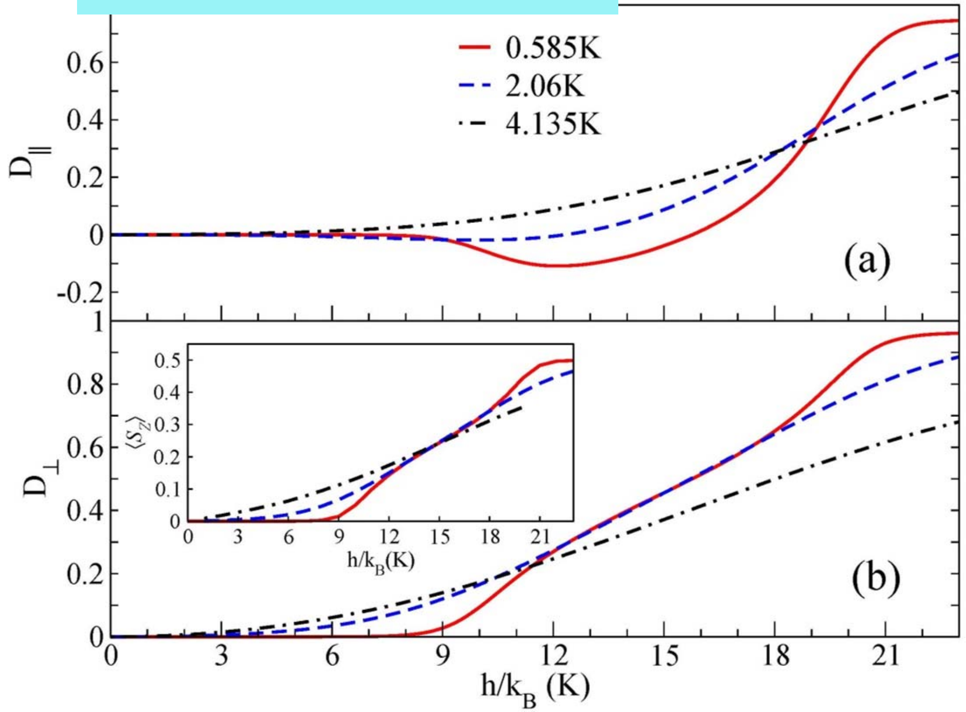
Leg correlator:

$$D^L(T,H) = \frac{1}{N}\sum_i^N \left\langle S_{i,1}S_{i+1,1} + S_{i,2}S_{i+1,2}\right\rangle$$

Rung correlator:

$$D^R(T,H) = \frac{1}{N}\sum_i^N \left\langle S_{i,1}S_{i,2}\right\rangle$$

QMC results

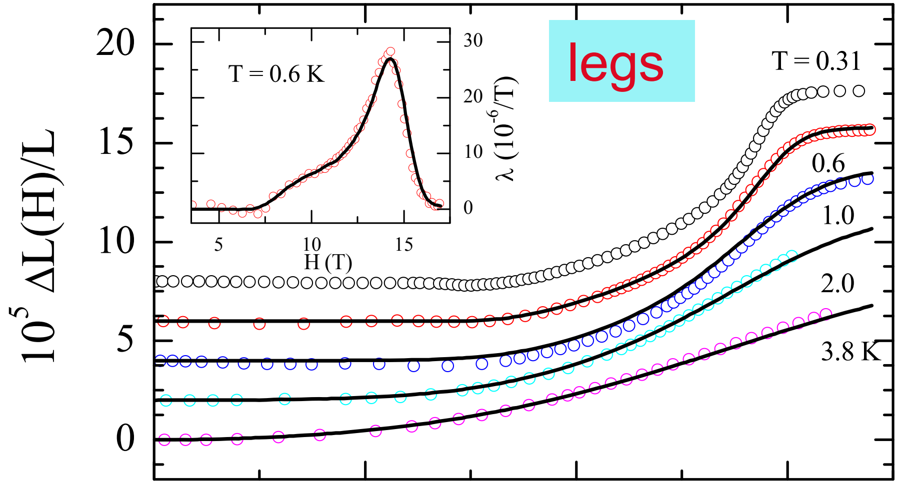


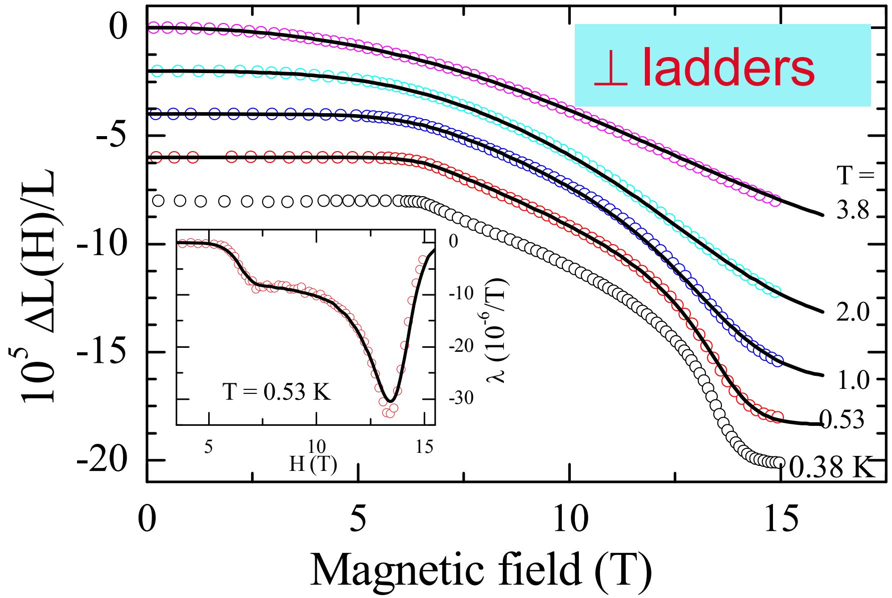
Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

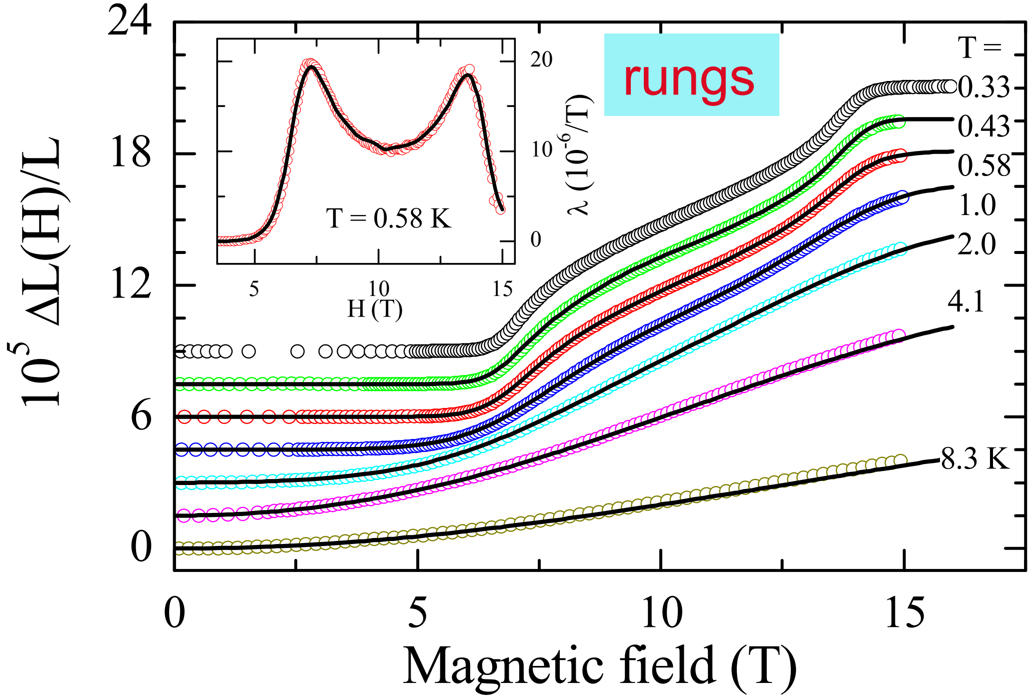
TL *et al*. PRL **100,** 067208 (2008)
F. Anfuso, ..., TL, *et al*. PRB **77**, 235113 (2008)

Magnetostriction ⇔ spin-spin correlation

fix $\lambda^{R,L}$ at 500 mK and calculate $\Delta L(H,T)/L$







⇒ lines are calculations

⇒ perfect agreement up to 8 K

Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

TL *et al*. PRL **100,** 067208 (2008)
F. Anfuso, ..., TL, *et al*. PRB **77**, 235113 (2008)

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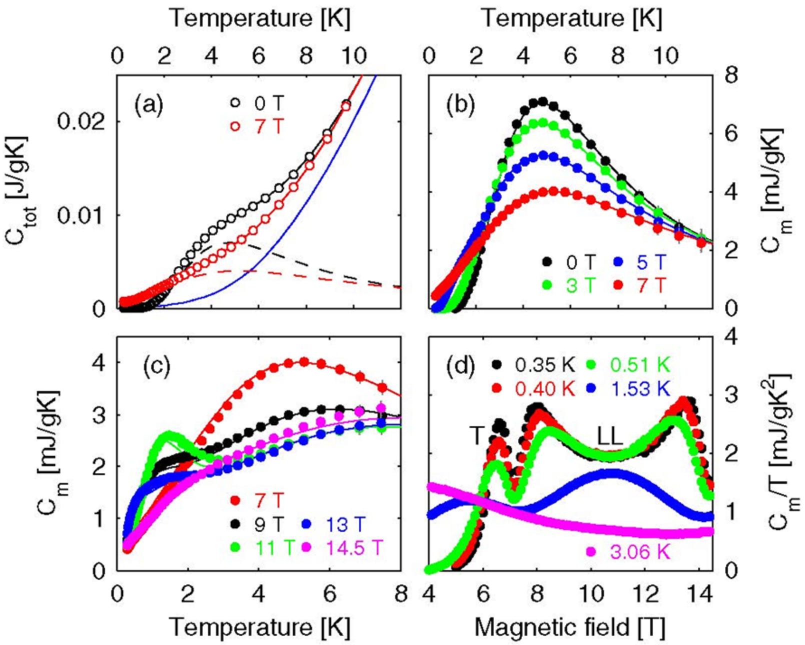
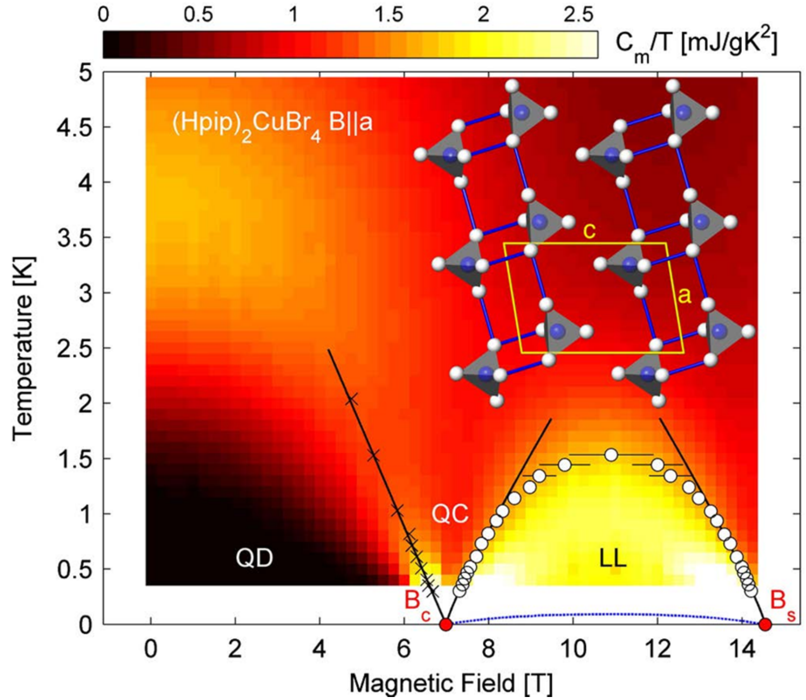
$(C_5H_{12}N)_2CuBr_4$: specific heat & magnetocaloric effect

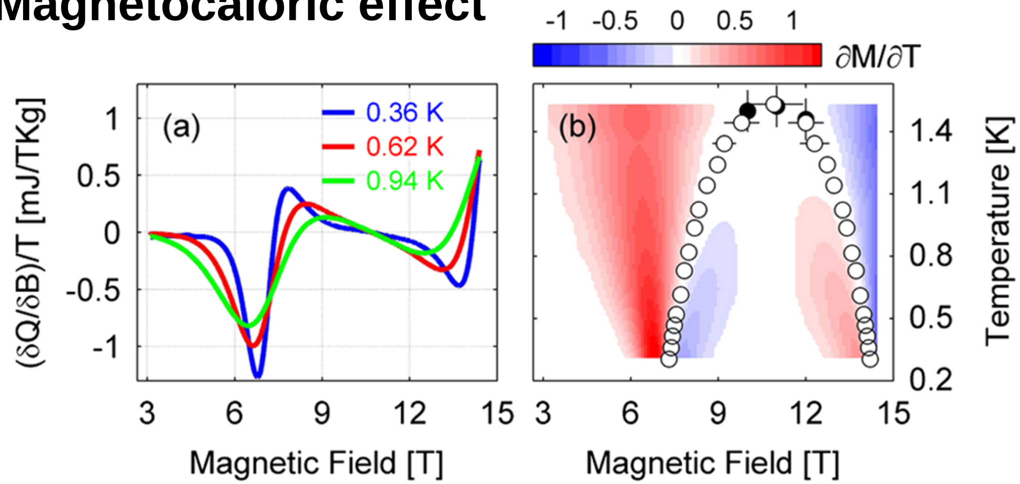


Specific heat

C. Rüegg *et al*. PRL **101,** 247202 (2008)



Magnetocaloric effect



Specific heat : $c\,(B,T>0)$

- 2 maxima below & above B_c
- minimum at $B_{min}(T \to 0) \to B_c$

Thermal expansion : $\alpha\,(B,T>0)$

- maximum below & minimum above B_c
- sign change at $B_{sc}(T \to 0) \to B_c$

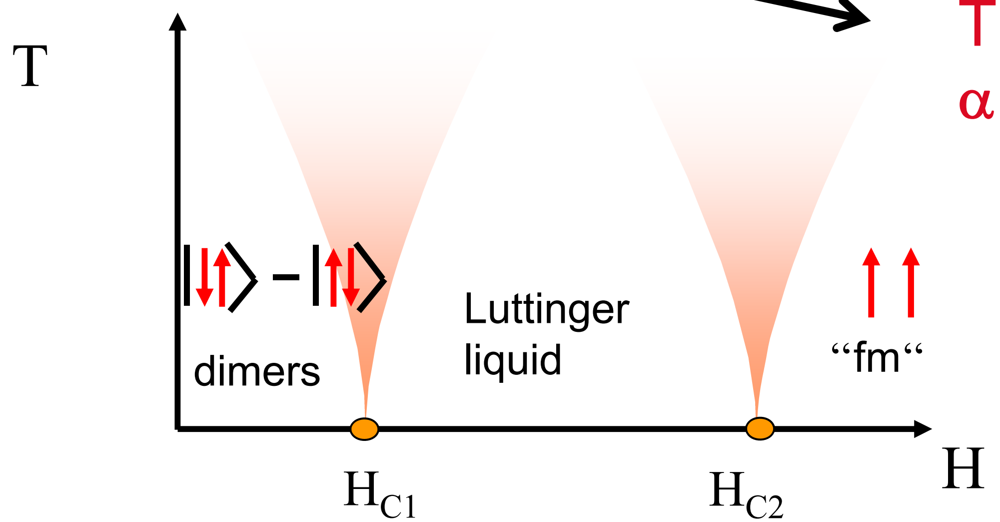
Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

Summary II: thermodynamics ⇔ quantum phase transitions

Spin-½ chains [CuPzN] and **spin-½ ladders** [BPCB] show highly anomalous low-temperature thermodynamics close to quantum phase transition(s)

- characteristic sign changes of $\alpha(T,H)$ and maxima of $c(T,H)$
- **$1/\sqrt{T}$ divergencies** of α for $T \to 0$ and $H = H_c$

?



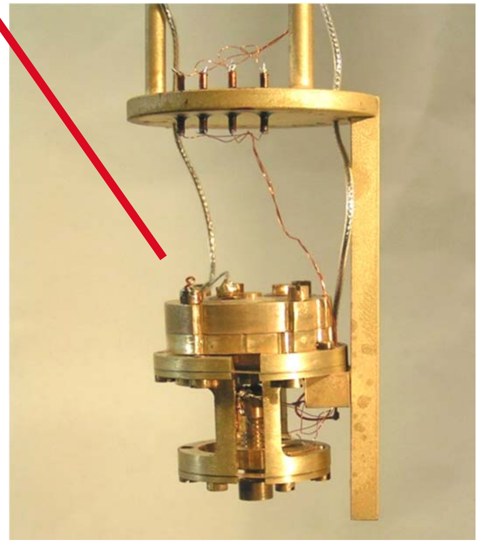
Thermodynamics:
$\alpha \to$ **0** **for T $\to$ 0**

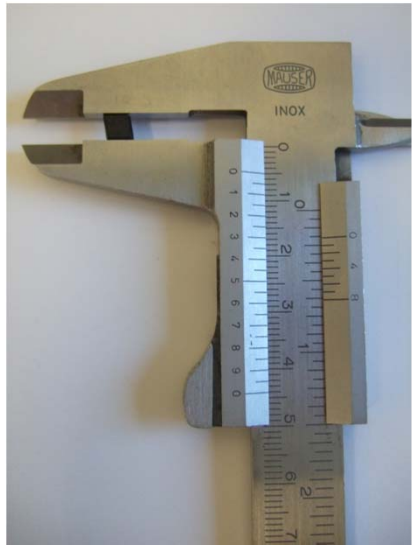
Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

Summary II: thermodynamics ⇔ quantum phase transitions

Spin-½ chains [CuPzN] and **spin-½ ladders** [BPCB] show highly anomalous low-temperature thermodynamics close to quantum phase transition(s)

- characteristic sign changes of $\alpha(T,H)$ and maxima of $c(T,H)$
- $1/\sqrt{T}$ divergencies of α for $T \rightarrow 0$ and $H = H_c$
- $\Delta L(T,H)$ measures spin-spin correlations





Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

Beyond 1D Heisenberg Spin-1/2 Systems

$$H = J \sum_{\langle i,j \rangle} \left\{ S_i^x S_j^x + S_i^y S_j^y + \Delta\, S_i^z S_j^z \right\} \quad \text{Heisenberg } (\Delta \approx 1) \ \text{ spin } S = \tfrac{1}{2}$$

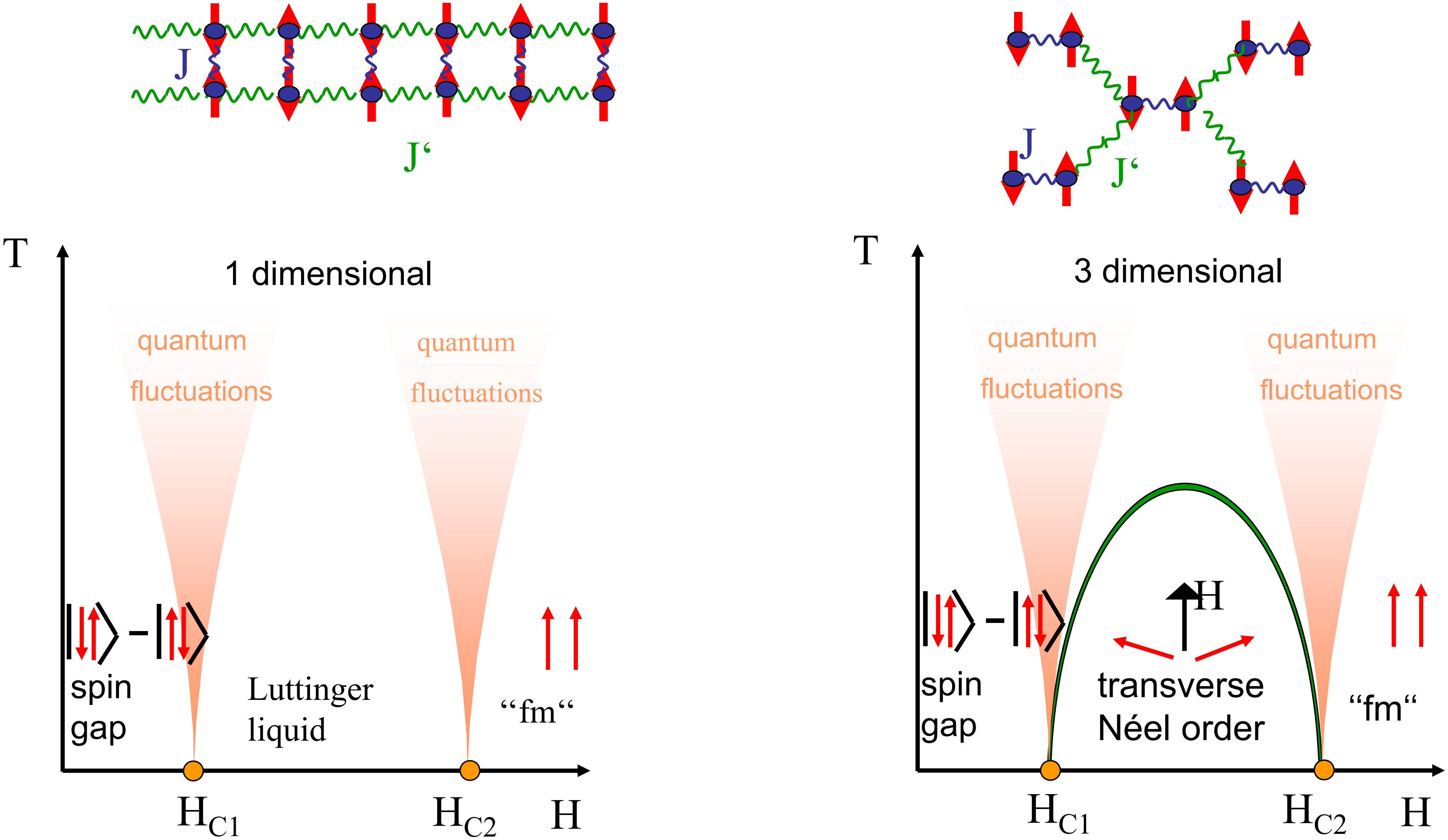
$\Rightarrow$ 3D-coupled dimers
e.g. **$TlCuCl_3$**

$\Rightarrow$ anisotropic exchange
Δ << 1 => XY spins
e.g. Cs_2CoCl_4
Δ >> 1 => Ising spins
e.g. $BaCo_2V_2O_8$ ← see poster S. Niesen
$CoNb_2O_6$

$\Rightarrow$ larger spin values
S >> 1 => S(S+1) ~ S^2
(„more classical"),
e.g. **$BaMn_2O_3$**,
$BaMn_2V_2O_8$ ← see poster S. Niesen

Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

1d spin ladder vs. 3d coupled spin dimers



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II. Physikalisches Institut
Universität zu Köln
Germany

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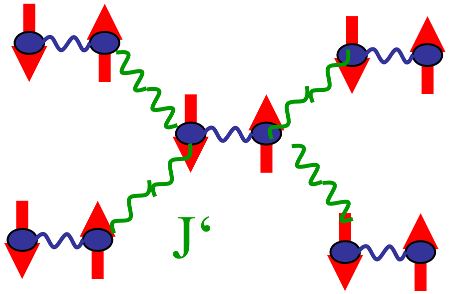
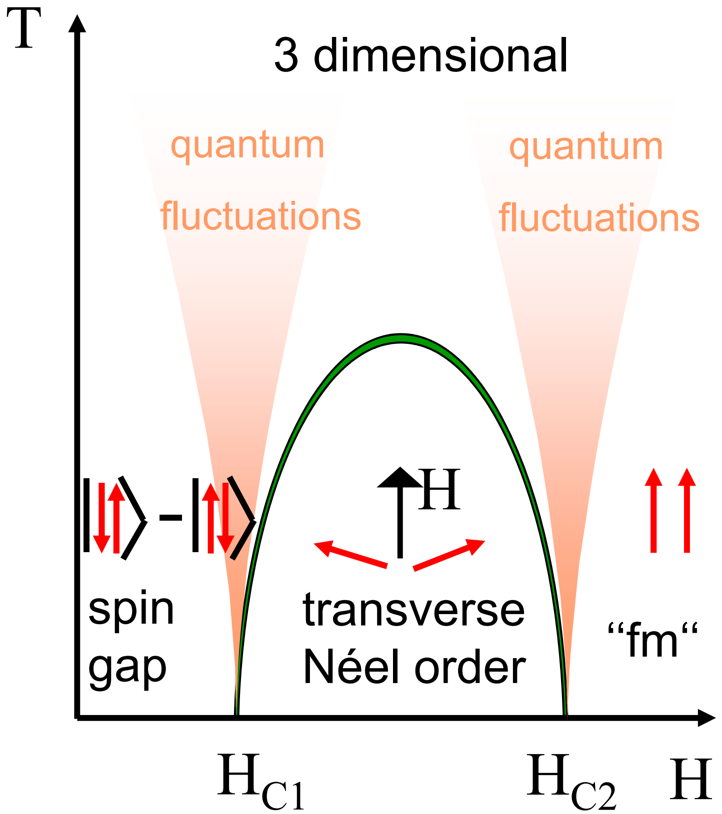
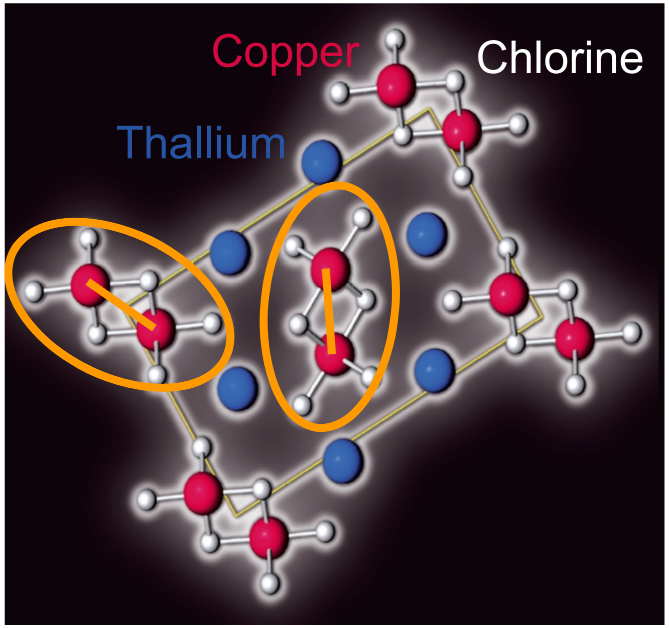
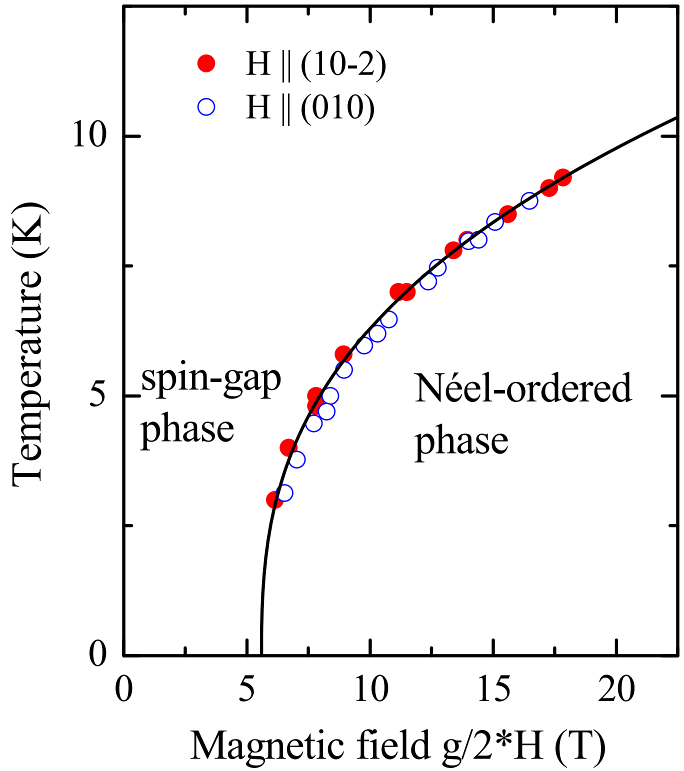
3d-coupled spin-dimer system $TlCuCl_3$







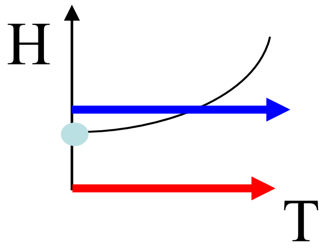
(picture from C. Rüegg)



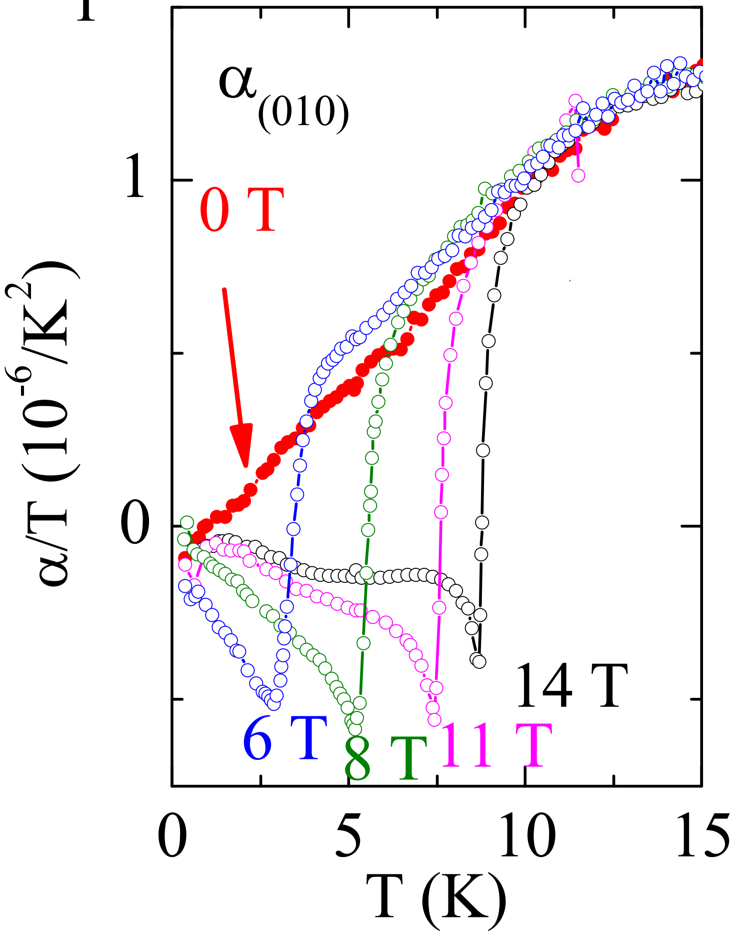
Rüegg et al., Nature **423,** 62 (2003)
Nikuni et al., PRL **84**, 5868 (2000)
Shindo et al. JPSJ **73**, 2642 (2004)
...

N. Johannsen, ... , TL.
PRL **95,** 017205 (2005)

3d-coupled spin-dimer system $TlCuCl_3$



$$\alpha = \frac{1}{L}\frac{\partial \Delta L(T)}{\partial T}$$



⇒ Quantum phase transition:

$H = H_{C0}$: - expected power laws

$$\frac{\alpha}{T} \propto \frac{1}{\sqrt{T}} \qquad \frac{C_p}{T} \propto \sqrt{T} \qquad \Gamma = \frac{\alpha}{C_p} \propto \frac{1}{T}$$

=> divergence and sign change of

$\Gamma(T\text{–>}0) = \alpha / C_p$

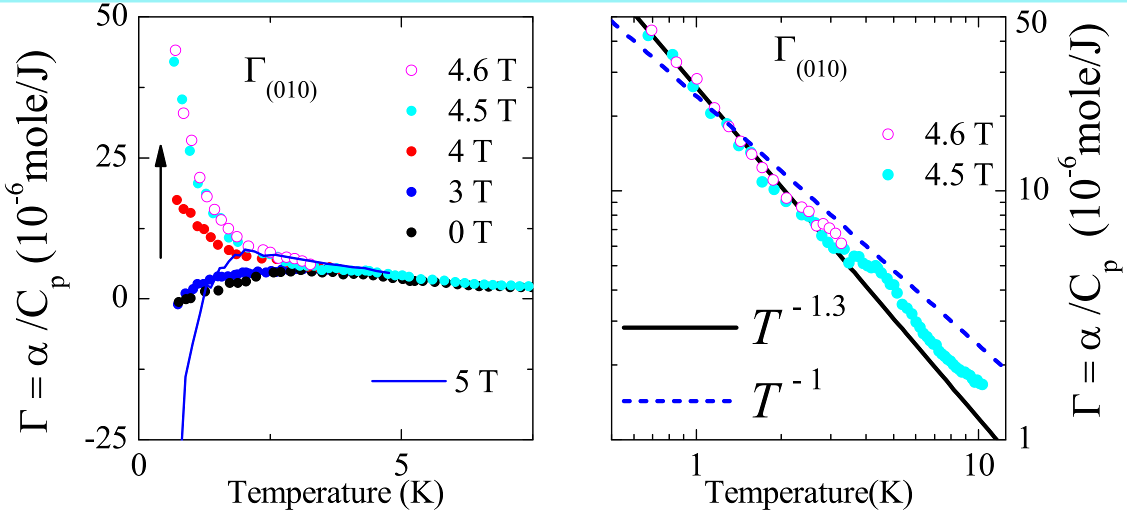
at H_{C0} as a generic feature of a QCP.

L. Zhu *et al*. PRL **95**, 066404 (2003);

M. Garst & A. Rosch. PRB **72**, 205129 (2005)

TL *et al*., JMMM **316**, 291 (2007).

$TlCuCl_3$: Grüneisen parameter



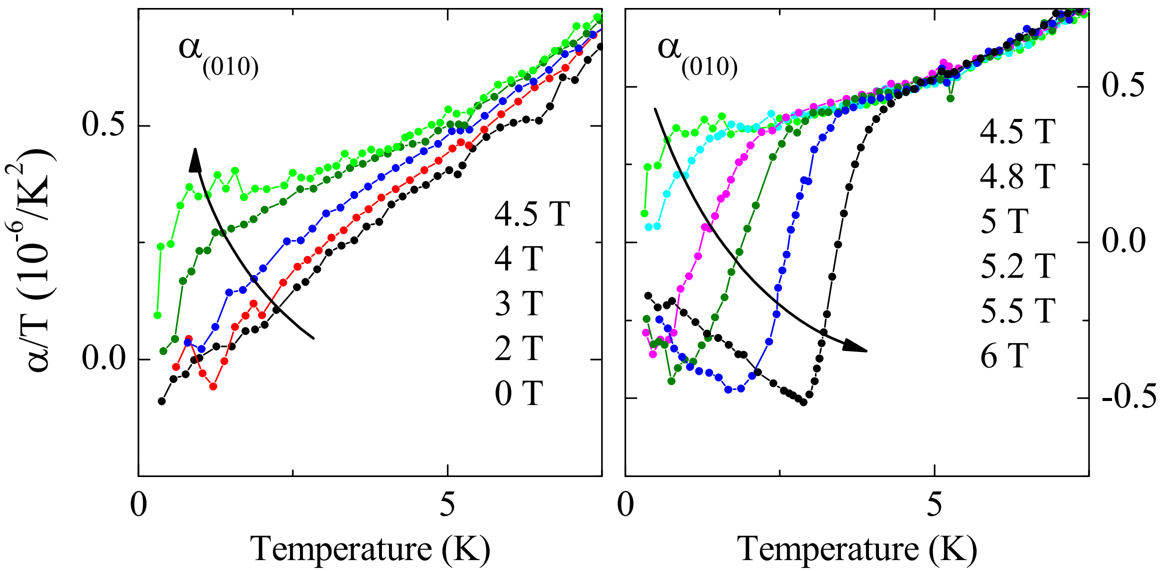
=> divergence and sign change of Γ for H-> H_{C0}

experiment: $\Gamma(T) \approx T^{-1.3}$

theory: $\Gamma(T) \approx T^{-1}$

Zhu et al. PRL 95 (2003)

Thermal expansion

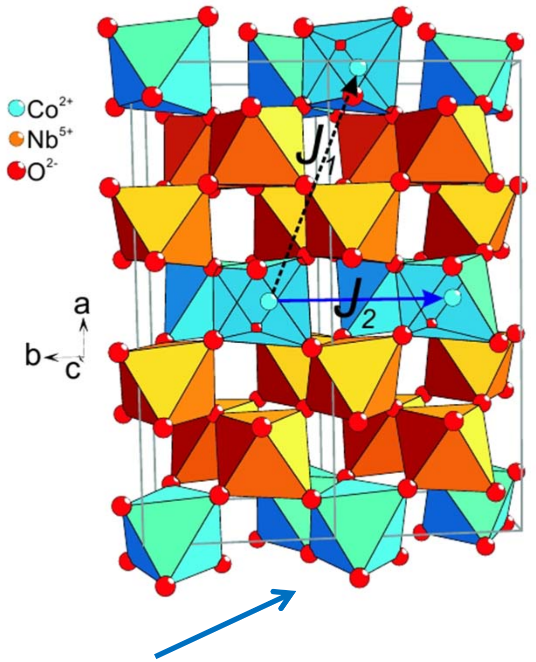


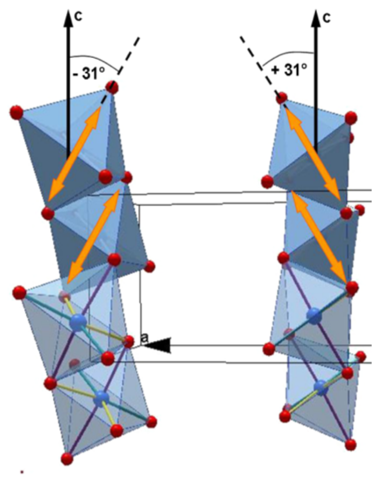
However, $\frac{\alpha}{T} \propto \frac{1}{\sqrt{T}}$ is not observed experimentally !

Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

TL *et al.*, JMMM **316**, 291 (2007).

Effective S = 1/2 Ising spin-chain system $CoNb_2O_6$

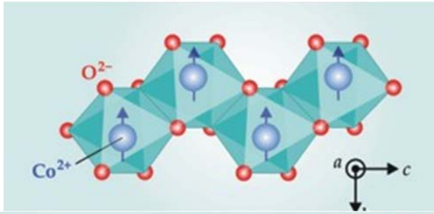




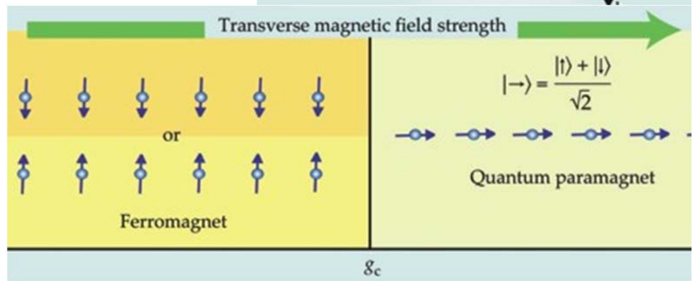
Chains of compressed CoO_6 octahedra along c axis

1D Ising spin ½ chain in transverse field

$$H = \sum_{i=1}^{N} - J S_i^z S_{i+1}^z + h S_i^x$$

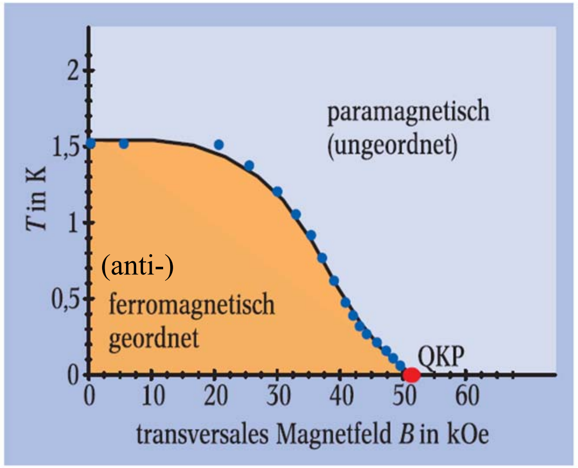


$$T = 0$$



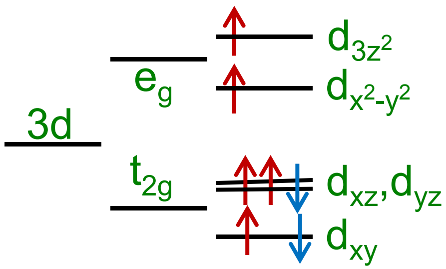
Keimer & Sachdev, Phys. Today **64**, 29 (2011)

interchain coupling => $T_{C,N} > 0$



M. Voijta, Phys. J. **1**, (2002)

Co^{2+} $3d^7$:



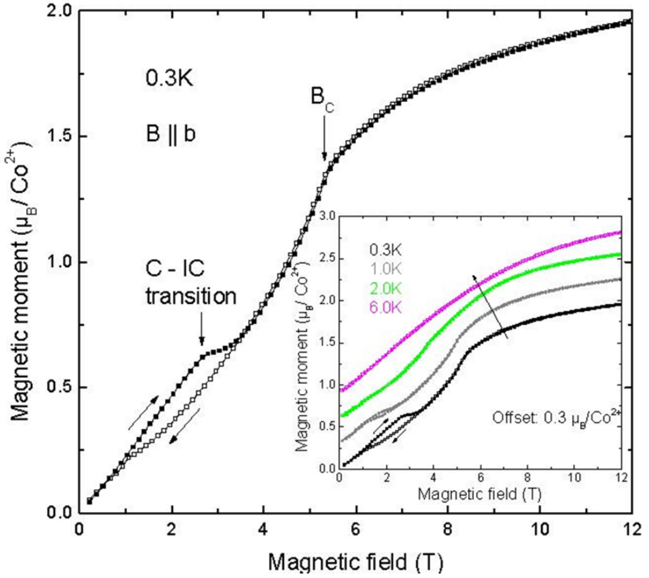
$\ell_z = \pm 1 \Rightarrow$ easy - axis anisotropy

effective Ising spins $S = 1/2$

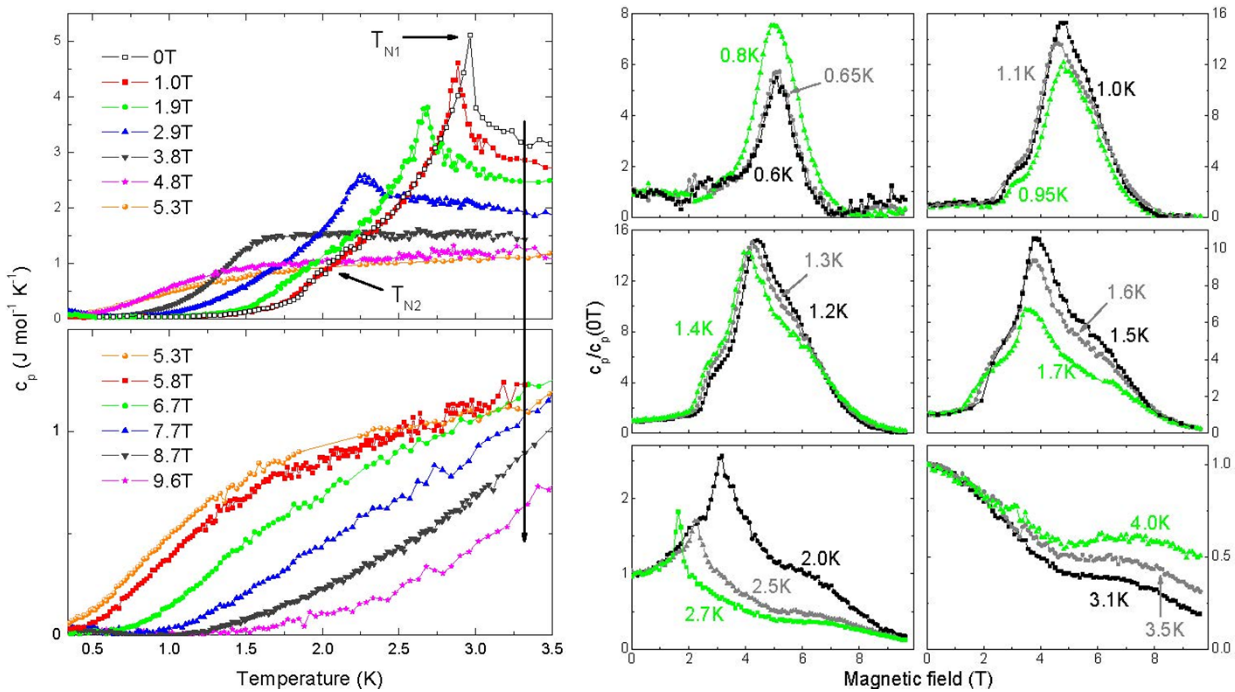
Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

Effective S = 1/2 Ising spin-chain system $CoNb_2O_6$

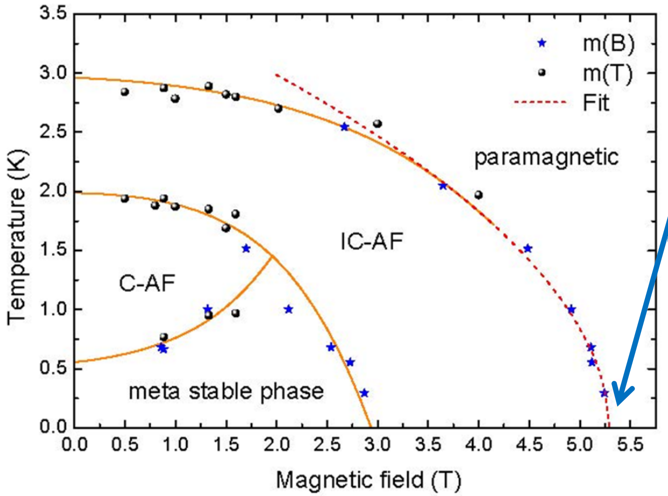
magnetization



Specific heat



phase diagram



Quantum phase transition

S = 1/2 Ising chain can be solved analytically

$$H = \sum_{i=1}^{N} -JS_i^z S_{i+1}^z + hS_i^x$$

(interchain coupling not included)

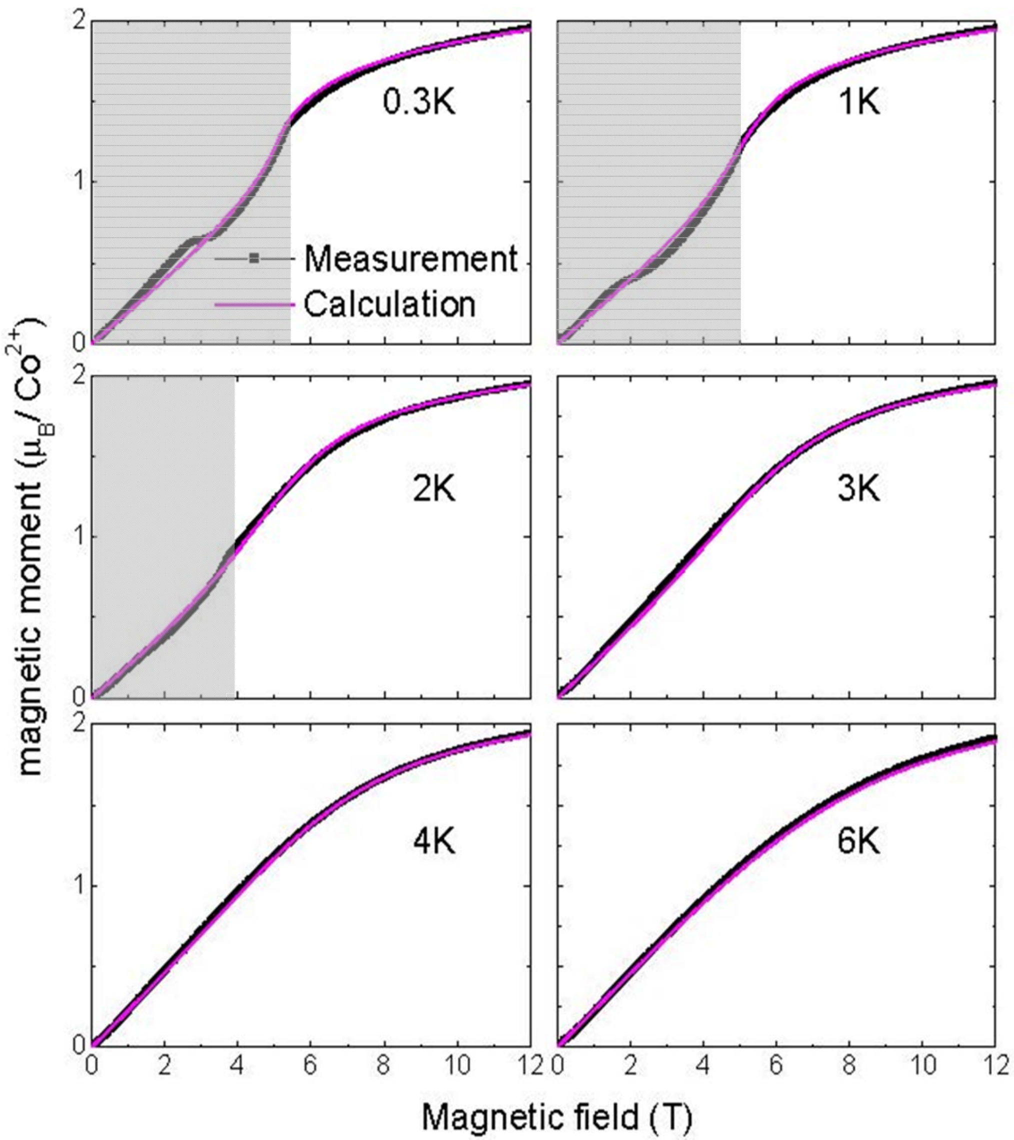
S. Sachdev, „*Quantum phase transitions*“ , Cambridge Uni. Press (2001)

S. Scharffe, diploma thesis, Uni. Köln (2012)

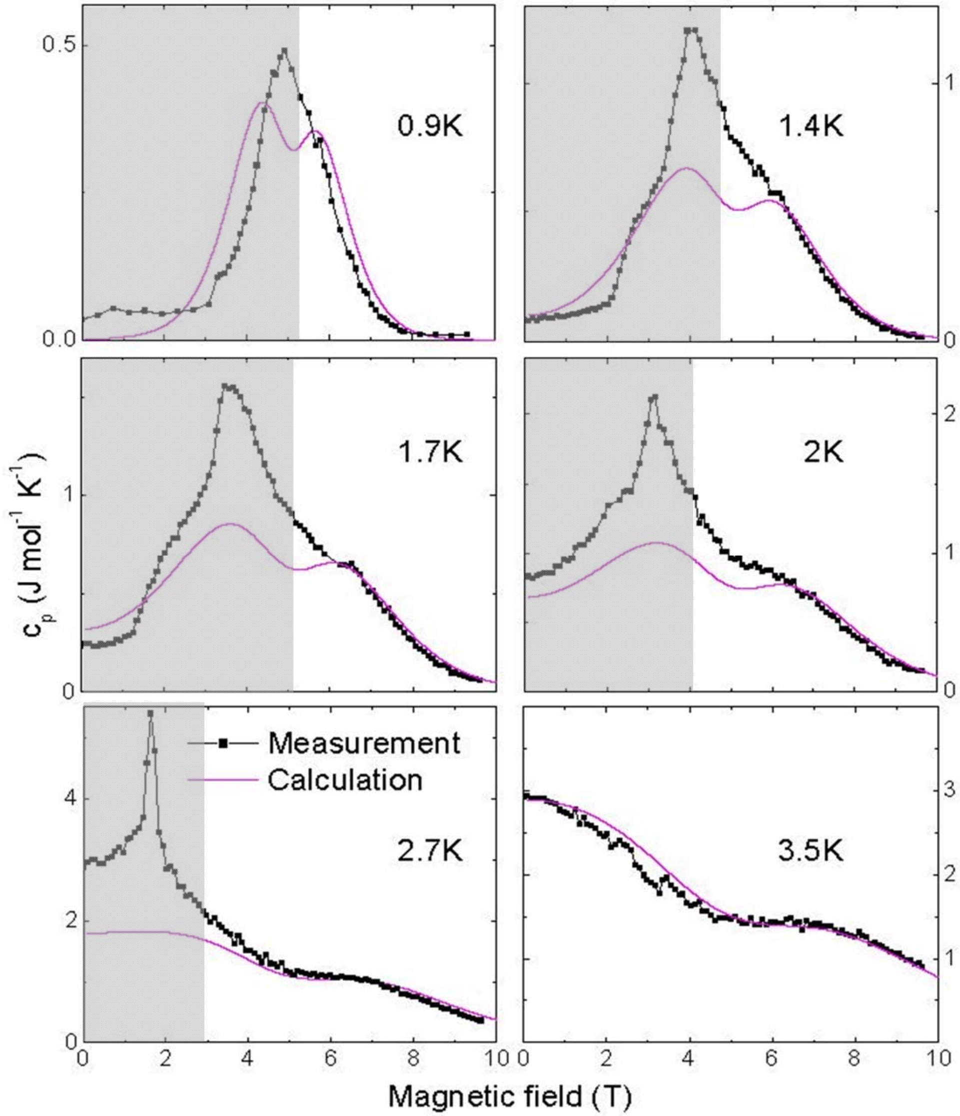
Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

Effective S = 1/2 Ising spin-chain system $CoNb_2O_6$

magnetization



Specific heat



Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

S. Scharffe, diploma thesis, Uni. Köln (2012)

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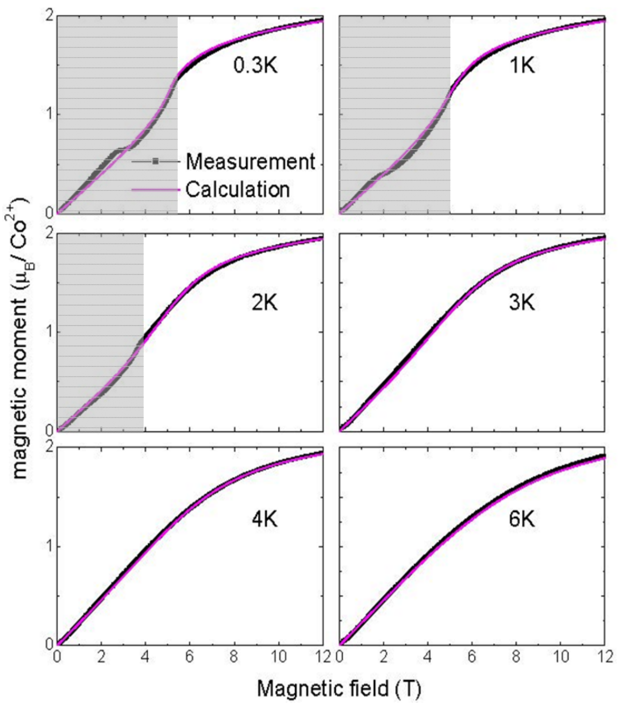
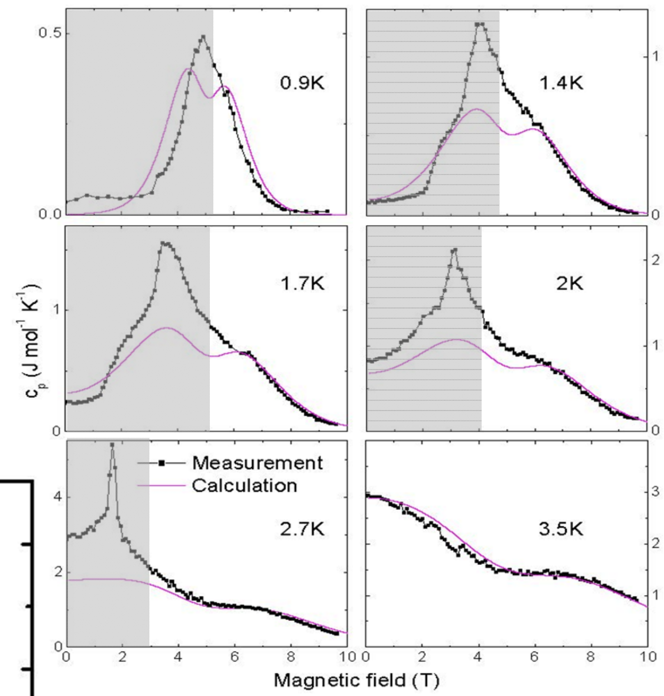
Effective S = 1/2 Ising spin-chain system $CoNb_2O_6$

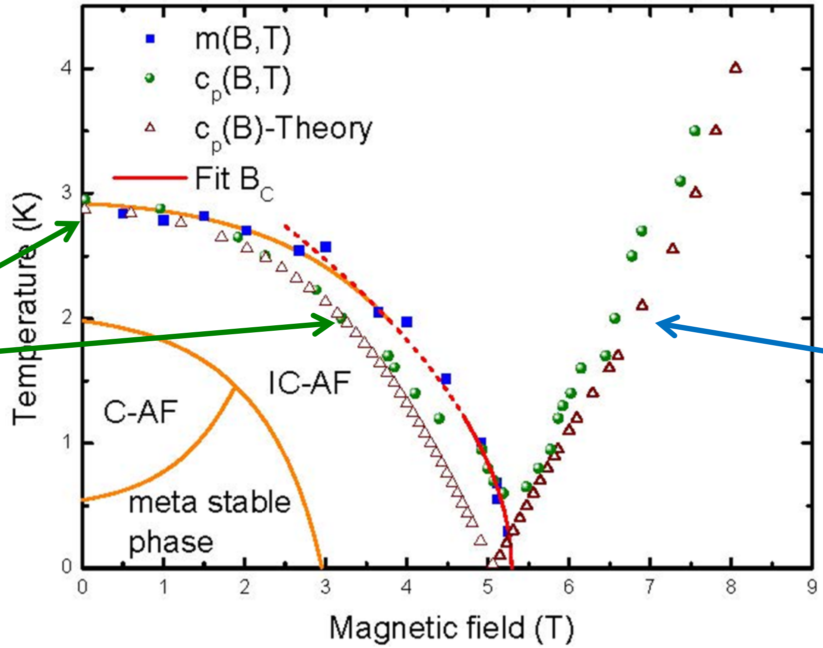
magnetization



Specific heat



Phase diagram



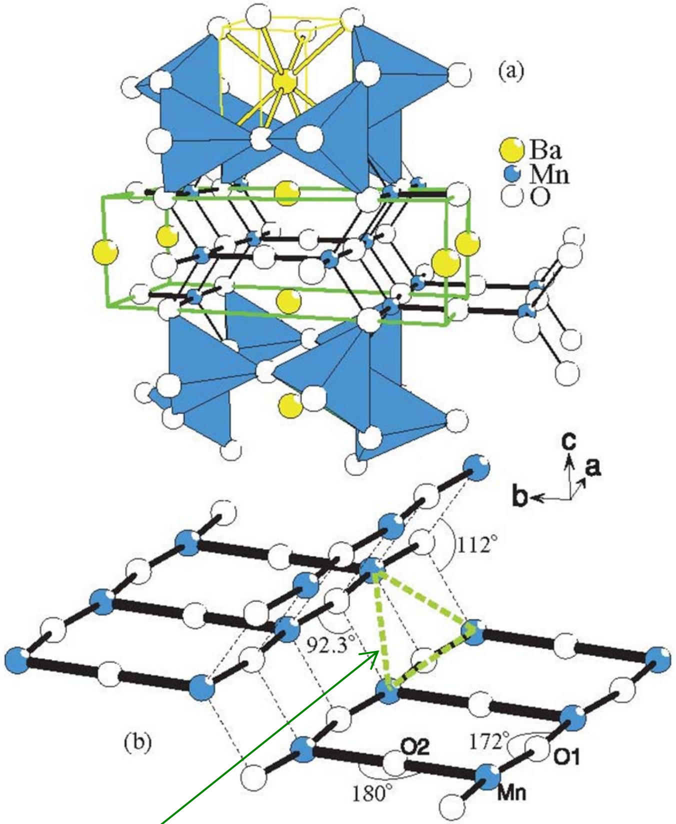
Maxima of c_p inside the 3D-ordered phase and even $T_N(0)$ might also be related to 1D physics ?

Maxima of c_p in the paramagnetic phase agree with theory.

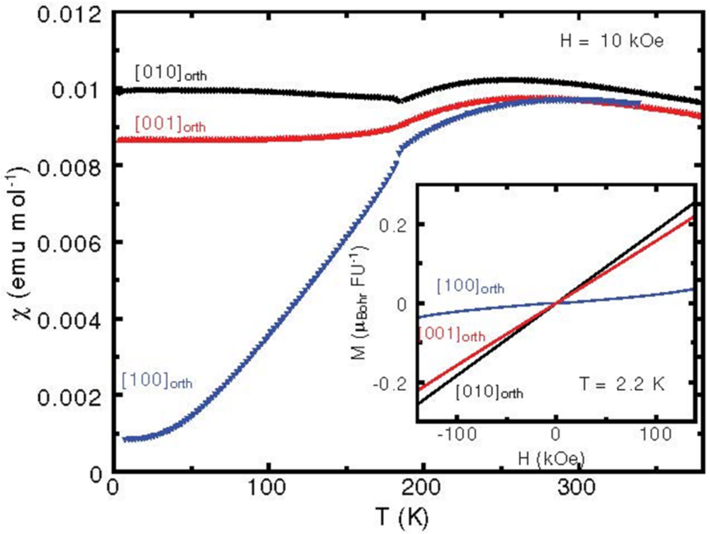
Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

S. Scharffe, diploma thesis, Uni. Köln (2012)

S = 5/2 spin-ladder system $BaMn_2O_3$



susceptibility

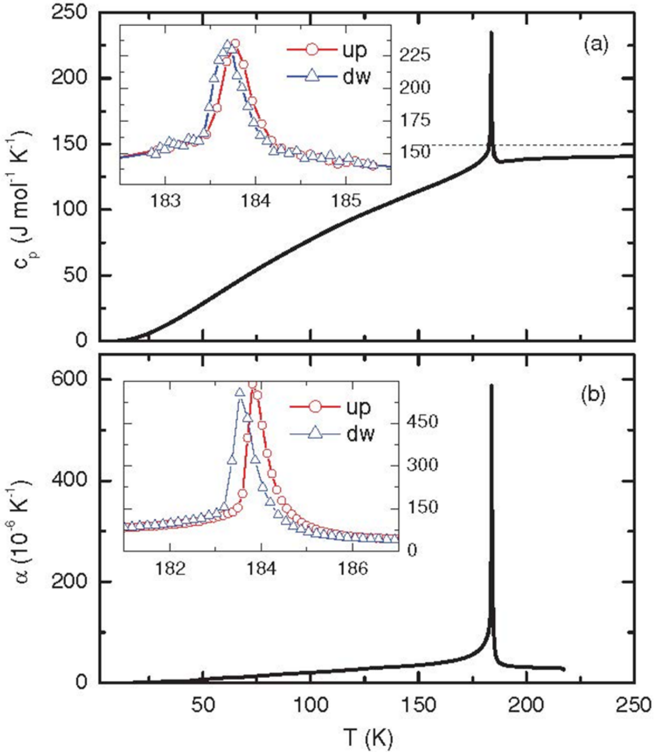


⇒ broad maximum of χ around 250 K

⇒ antiferromagnetic order at ~ 184 K

⇒ $J_{||,\perp}$ ~ 40 K => H_{sat} ~ 450 T

specific heat & thermal expansion

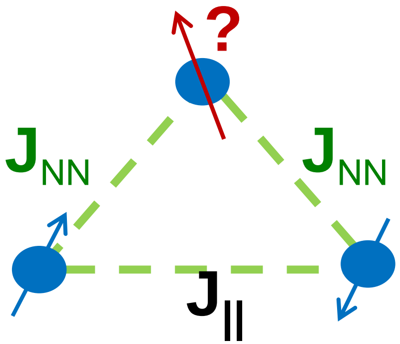


⇒ huge anomalies at T_N

⇒ 1st-order transition

⇒ structural change

NN ladders are Decoupled by geometrical frustration



Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

M. Valldor, ... , TL. PRB **83,** 024418 (2011)

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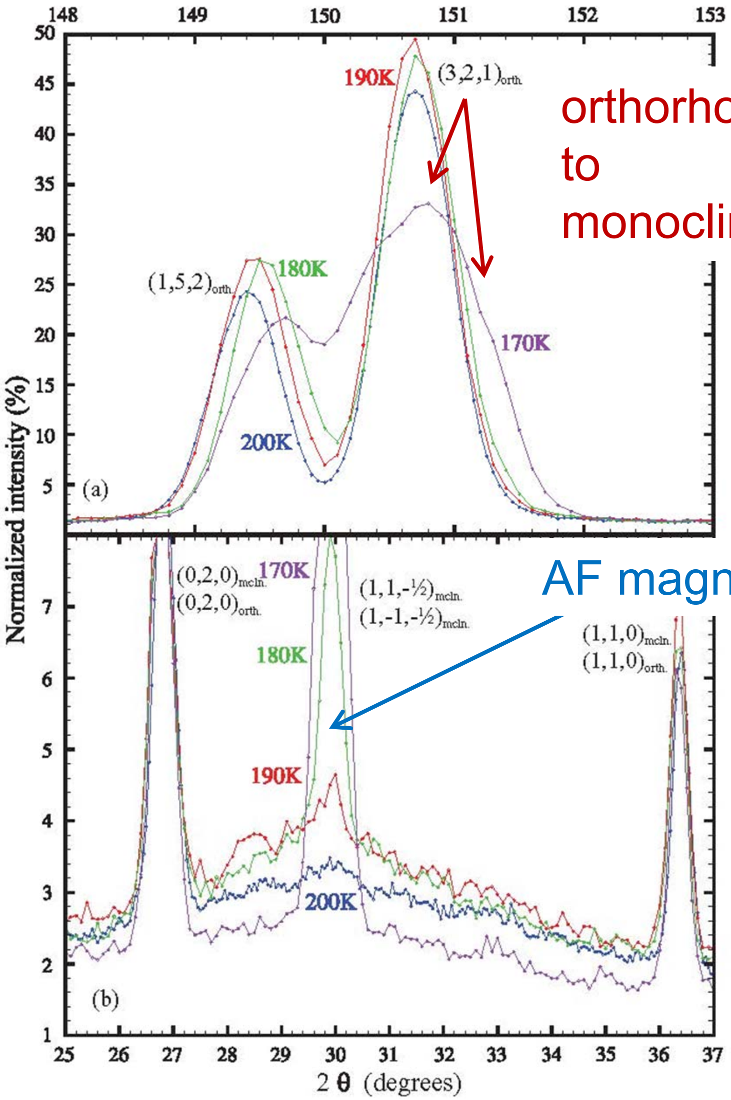
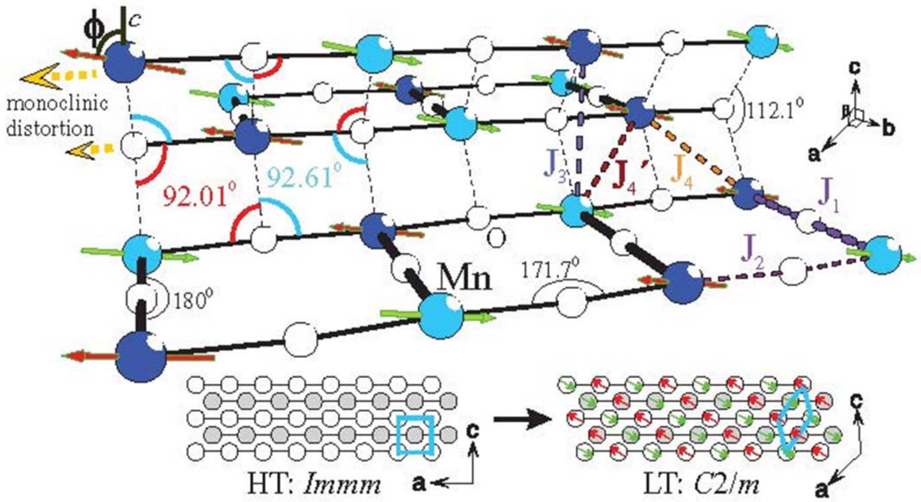
S = 5/2 spin-ladder system $BaMn_2O_3$

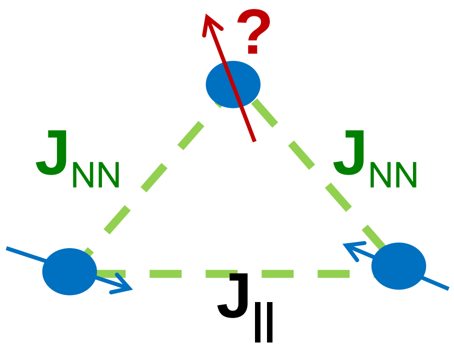
Neutron powder diffraction



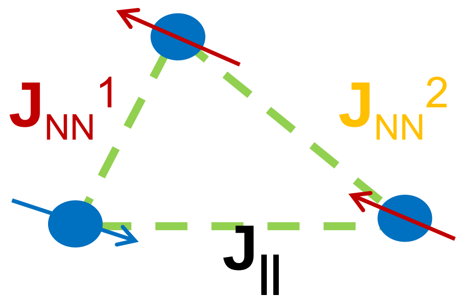
(magnetic) low-temperature structure



⇒ **magnetostrictive lifting of frustration**
⇒ **long-range AFM order**



T > T_N



T < T_N ($\Delta\gamma < 1°$)

Thomas Lorenz
II. Physikalisches Institut
Universität zu Köln
Germany

M. Valldor, ... , TL. PRB **83,** 024418 (2011)

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Summary

- Spin-½ Heisenberg chains & Spin-Peierls transition

structural & magnetic dimerization due to magnetic energy gain => spin gap

universal H-T phase diagram; incommensurate magnetic phase above H_c

$CuGeO_3$: in general well understood; strong magnetic frustration

- Field-induced Quantum Phase transitions in Spin-½ chains & Spin-½ ladders

characteristic sign changes of $\alpha(T,H)$ and double-maxima of c(T,H)

$1/\sqrt{T}$ divergencies of α for $T \rightarrow 0$ and $H = H_c$

$\Delta L(T,H)$ measures spin-spin correlations

- Beyond 1D Spin-½ Heisenberg systems

complex interplay of quantum of quantum & thermal criticality

dynamics of magnetic excitations => thermal transport, spin currents, …

influence of charge-carrier doping => metallic systems

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