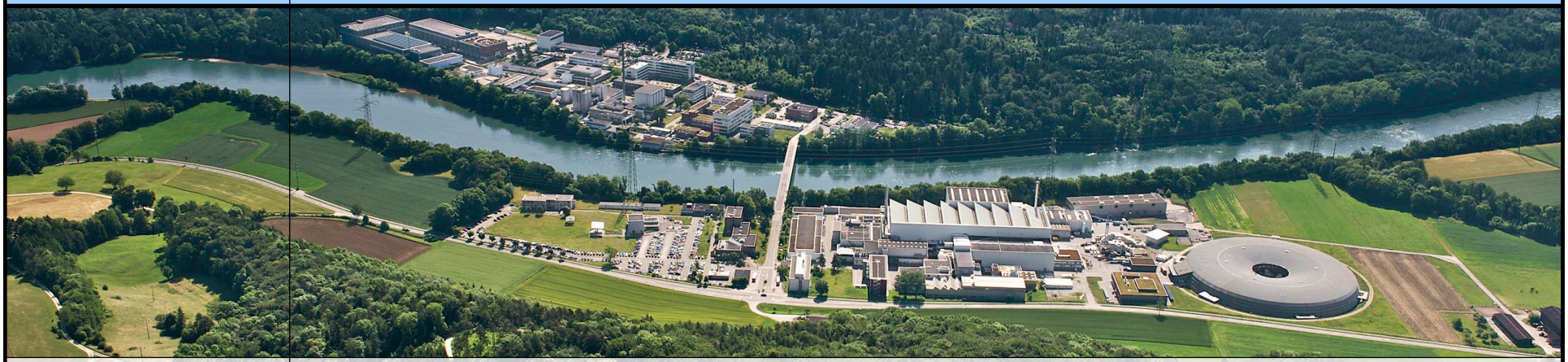
PAUL SCHERRER INSTITUT



Wir schaffen Wissen – heute für morgen

Quantum Critical Points in Systems with Effective Singlet Ground-States

Christian Rüegg

Laboratory for Neutron Scattering and Imaging
Paul Scherrer Institute and University of Geneva

PSI-China Meeting, Brugg, 4 May 2015

Outline

Quantum Critical Points in Systems with Effective Singlet Ground-States

I: **Introduction:** Quantum Critical Points in quantum magnets

II: **Neutron Scattering at SINQ:** News and High pressure studies

III: **Pressure-controlled QCP in $TlCuCl_3$:** Evolution of elementary excitations

IV: **Quantum vs. Thermal melting:** Critical behaviour near the QCP

V: **Conclusions**

Publications:

- P. Merchant, B. Normand, Ch. Rüegg *et al.*, Nat. Phys. **10**, 373 (2014).
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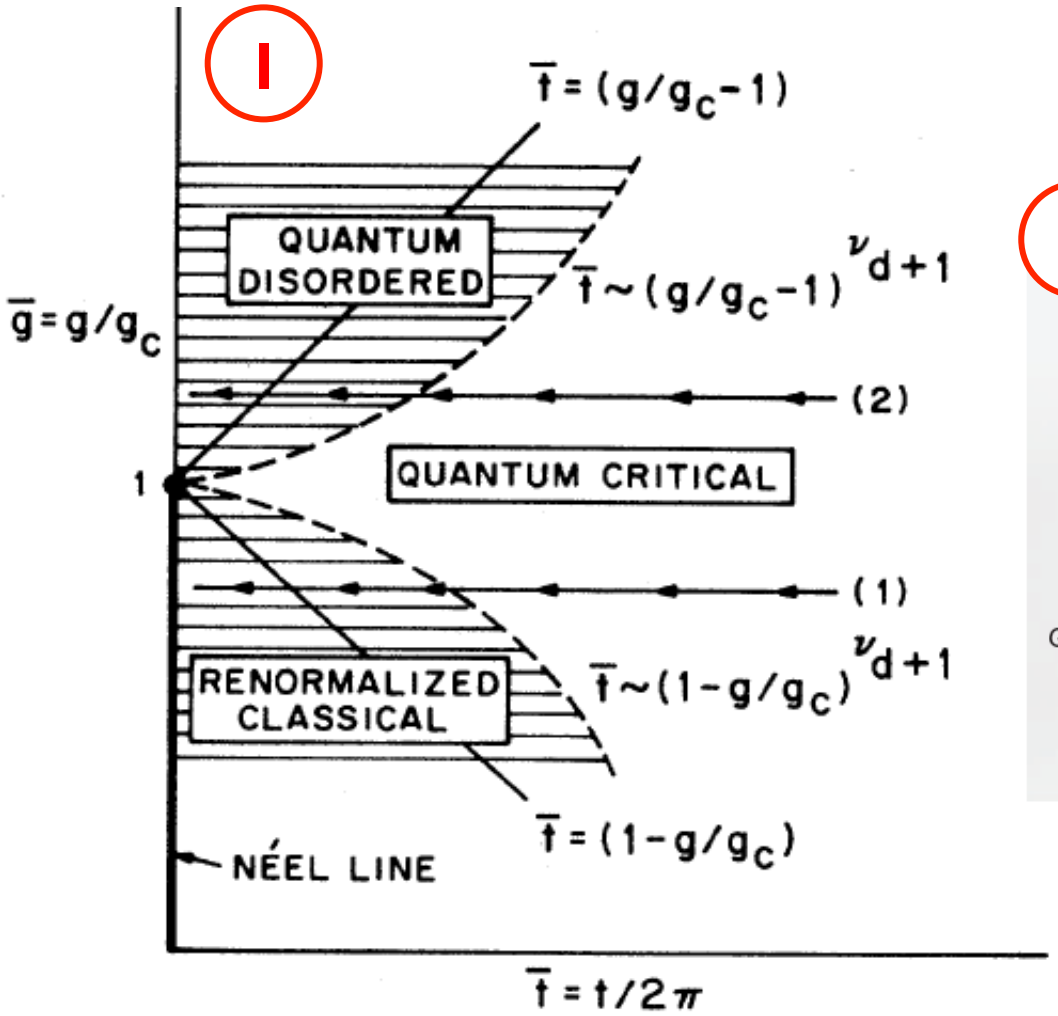
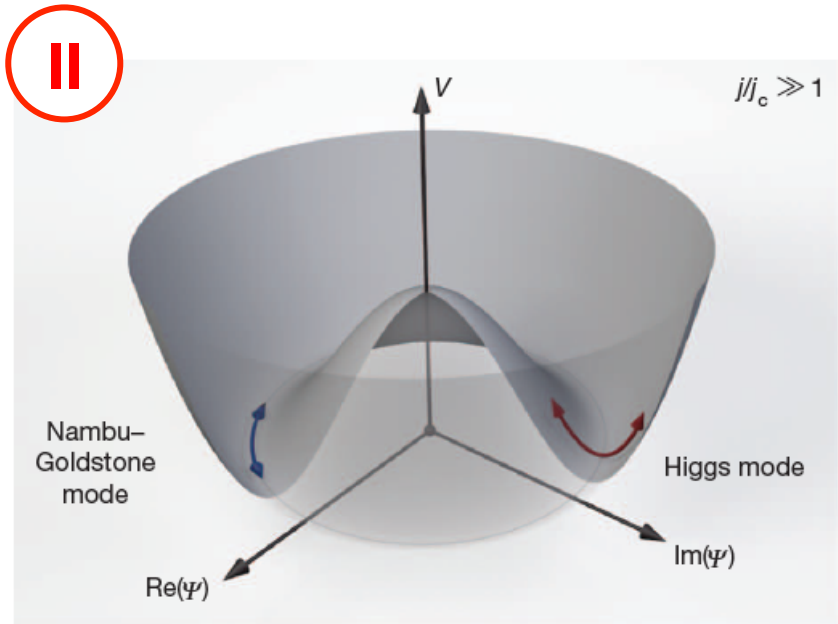
Quantum Critical Points in Magnets

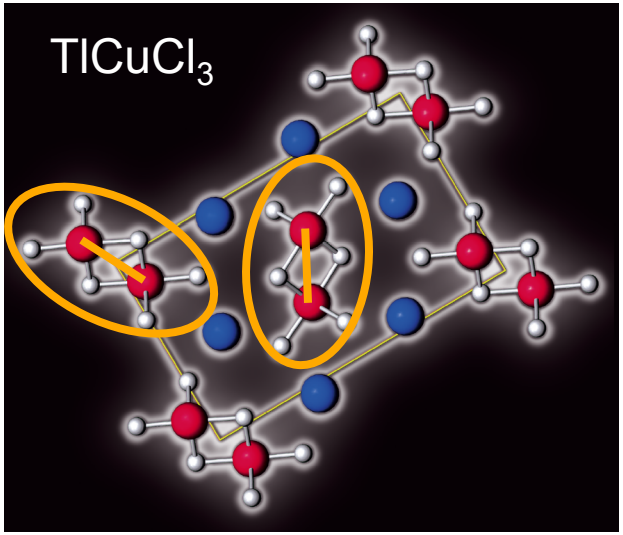


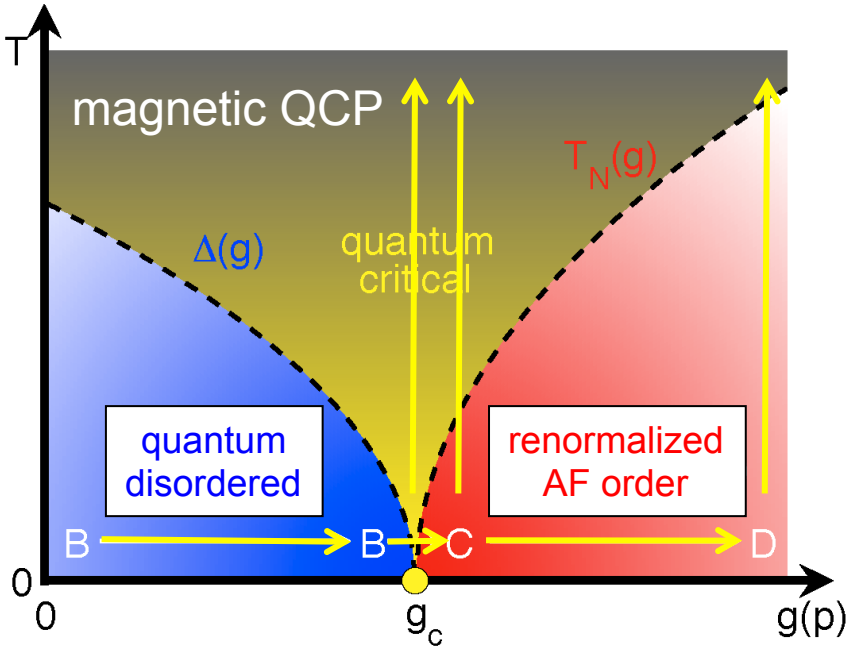


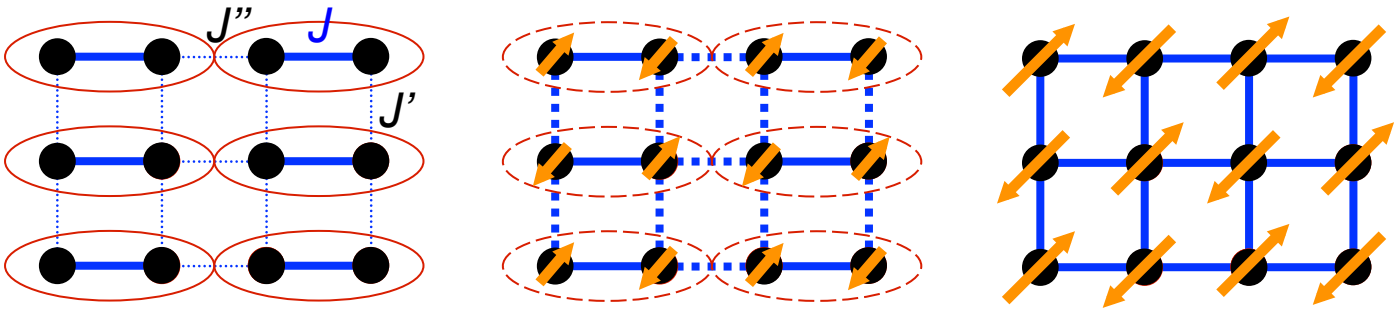
M. Endres *et al.*, Nature (2012).

• S. Chakravarty, B. I. Halperin, and D. R. Nelson, Phys. Rev. B **39**, 2344 (1989).

Pressure-Tuned Quantum Critical Point







dimers → critical magnet → ordered AFM

pressure →

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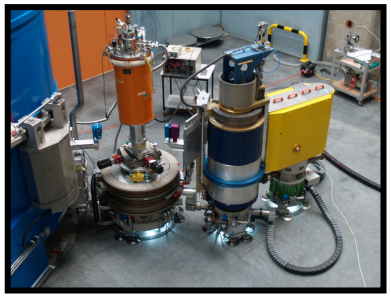
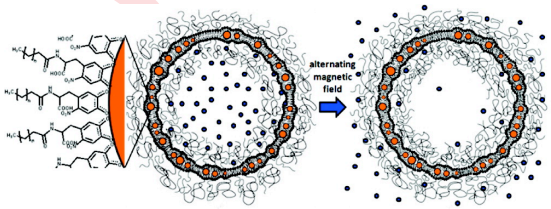
Laboratory for Neutron Scattering and Imaging

STAFF:
26 Scientists (Head: Prof. Ch. Rüegg, PSI/UniGE)
12 Postdocs
14 Ph.D. students

INSTRUMENTS AT SINQ:
13 instruments at SINQ with 400 experiments per year



SCIENCE:
Fundamental, materials-driven research with neutrons
Key topics: atomic and magnet structures, magnetism, electronic correlations, soft matter and biology, energy storage and conversion, health care
150 publications per year (40% in high-impact journals)



EDUCATION:
Ph.D. program, student practicals (EPFL, ETH, Uni Basel), lectures, schools

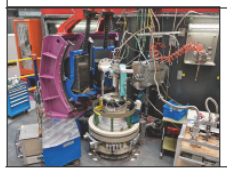
FUNDING:
PSI, SNF, SBFI, EU, national and international partners (EPFL, UniGE, DANSCATT)

Swiss Spallation Neutron Source SINQ

Instruments at SINQ

Diffraction | Spectroscopy | SANS + Ref. | Imaging

TriCS



HRPT

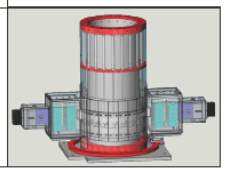
NEUTRA

POLDI

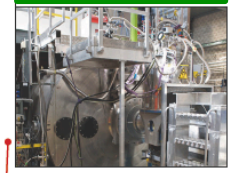
MORPHEUS

AMOR

MuPAD

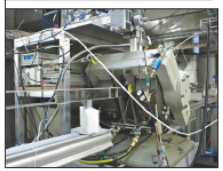


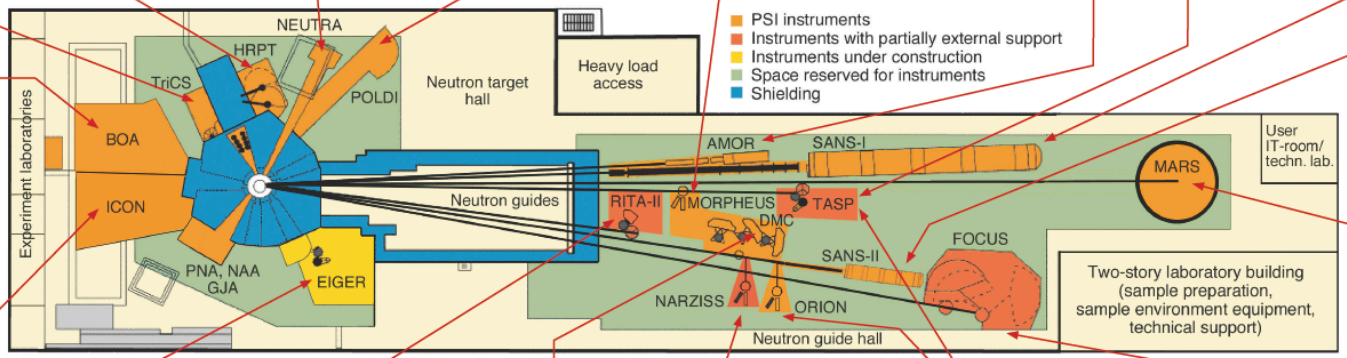
SANS-I



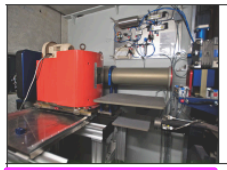
SANS-II

BOA





MARS



ICON

EIGER

RITA-II

DMC

NARZISS

ORION

TASP

FOCUS

New Instruments CAMEA and ZEBRA at SINQ

Design Goals

Small samples of new emergent materials, thin films, heterostructures

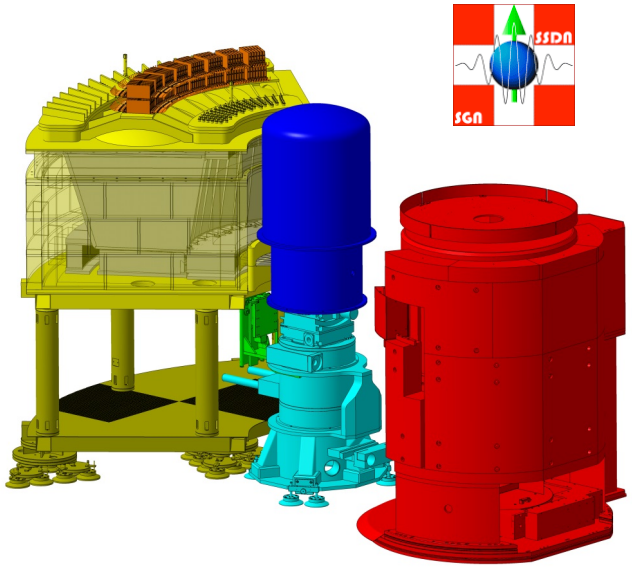
Multi-extreme conditions (temperature, pressure, magnetic and electric fields)

CAMEA

Upgrade of cold TAS RITA-II at SINQ

Collaboration: PSI-EPF Lausanne

Funding: SNF R‘equip, EPFL, and PSI

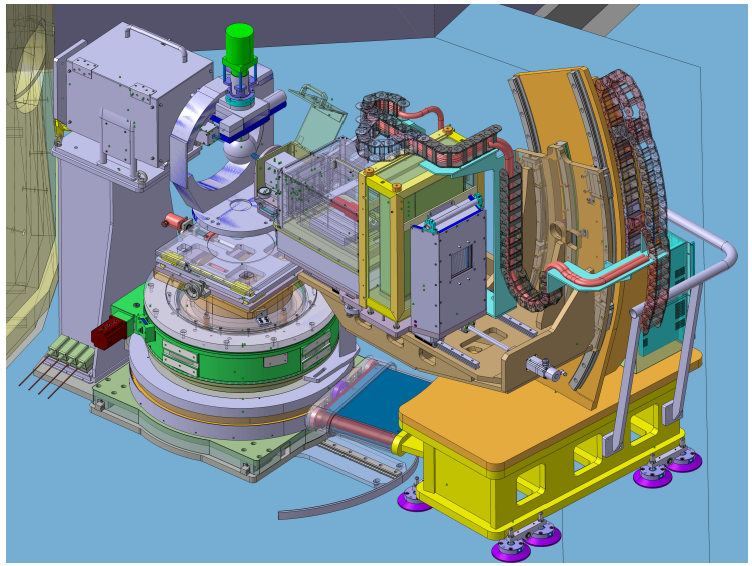


ZEBRA

Upgrade of diffractometer TriCS at SINQ

Collaboration: PSI-University of Fribourg

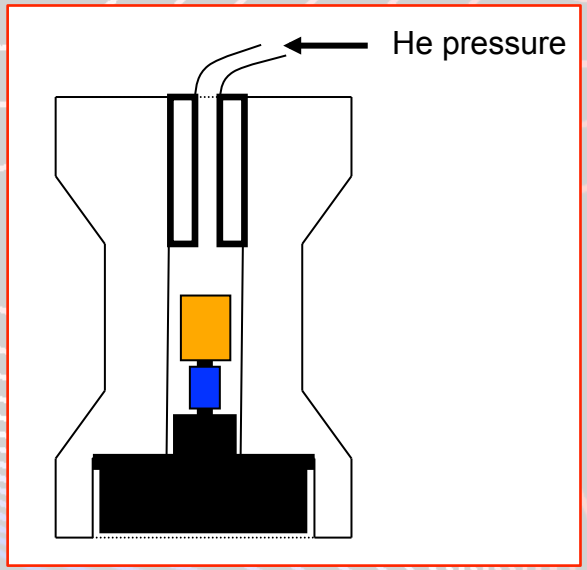
Funding: SNF R‘equip and PSI



Neutron Scattering Experiments under Pressure

Sample environment:	Sample volume:	Signal:
cryostat 1.5 K	>3 cm^3	1
Pressure Cells:		
5 kbar (gas, 1.5 K)	2 cm^3	0.3
16 kbar (clamp, 1.5 K)	1 cm^3	0.1
30 kbar (McWhan, 1.5 K)	0.2 cm^3	0.02
200 kbar (Paris-Edinb. 4 K)	0.03 cm^3	0.005

Contact: Jonathan White, PSI



Gas: L. Melesi *et al.* Clamp: R. Sadykov *et al.* P-E: S. Klotz *et al.*

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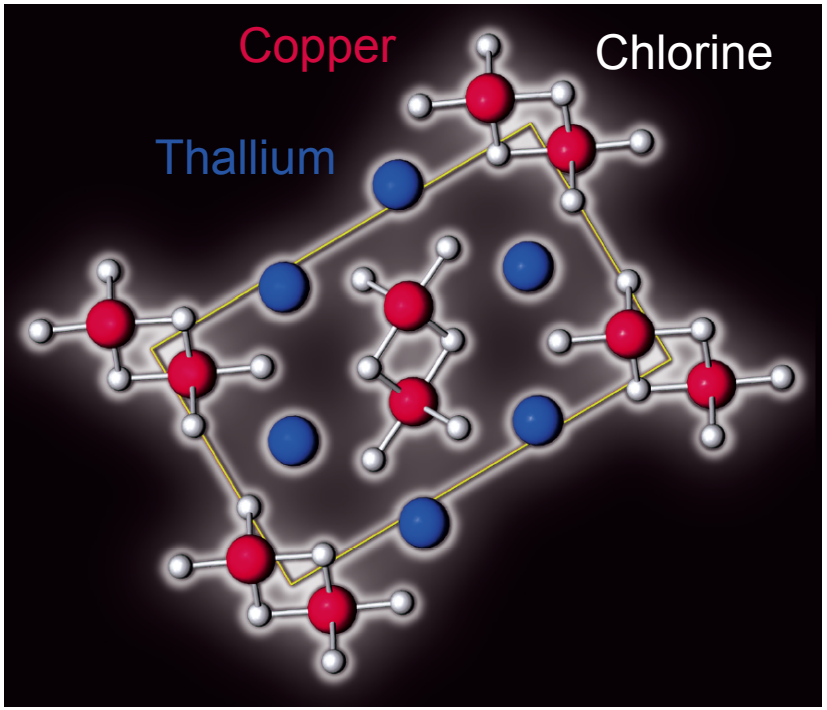
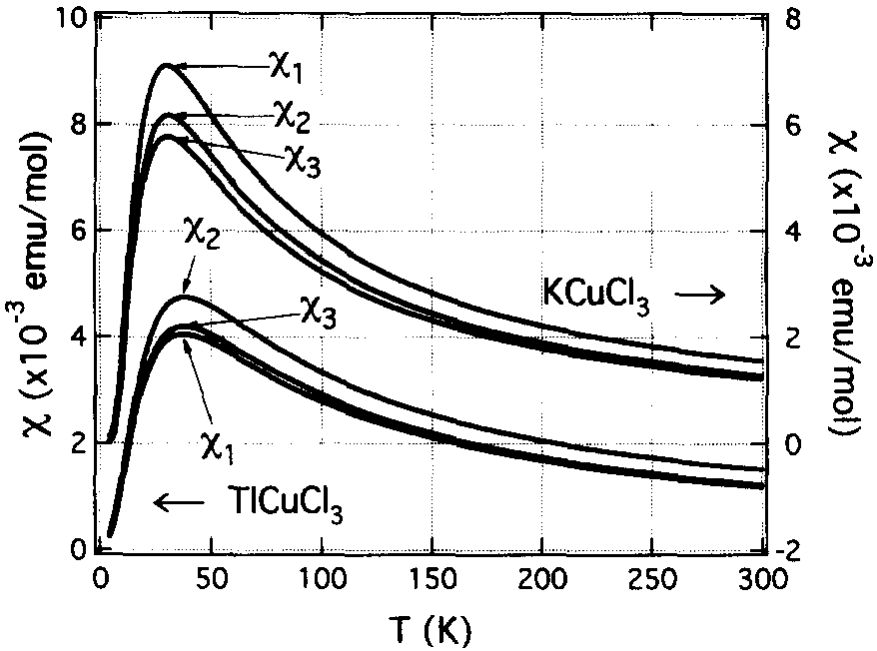
$ACuCl_3$ (A=K, Tl, NH_4)



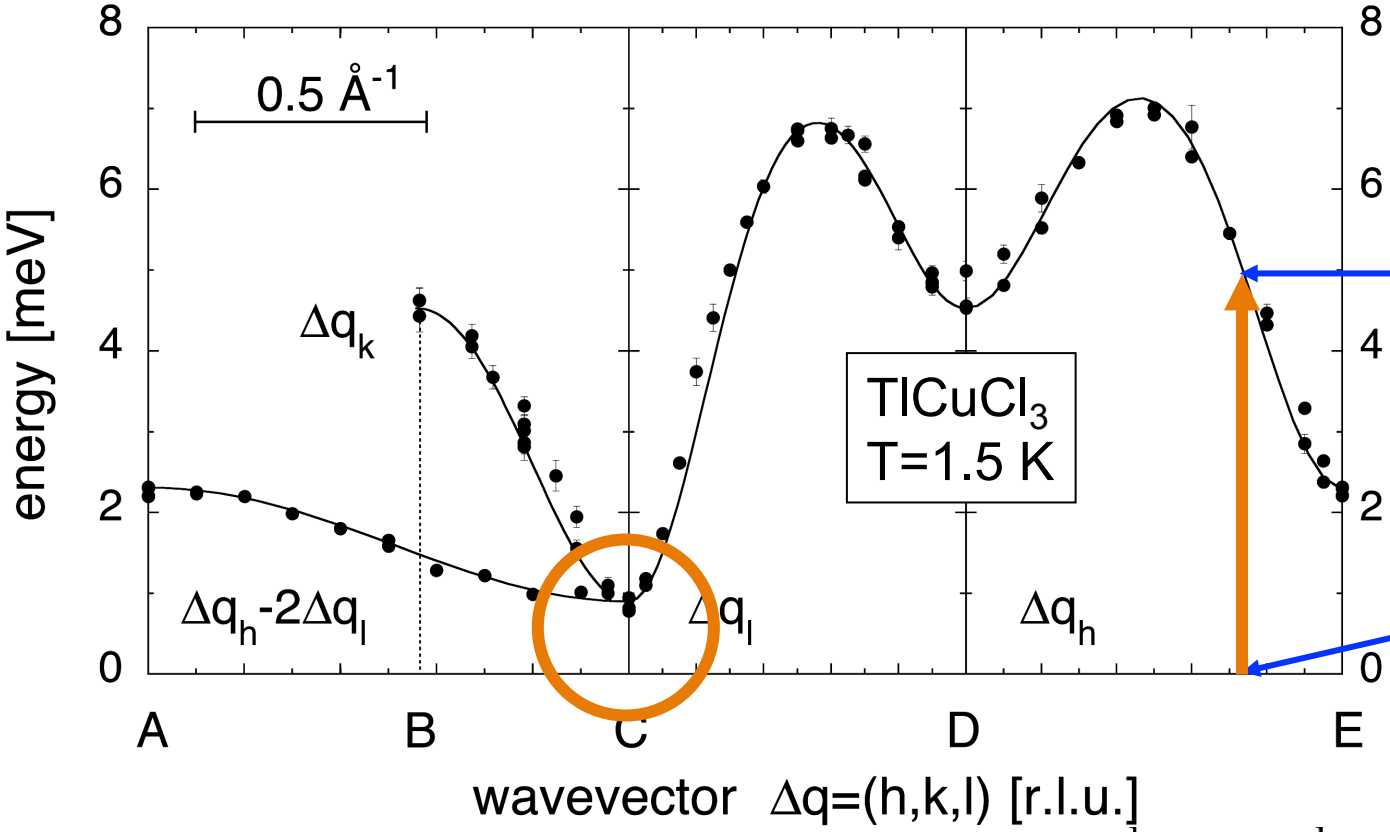


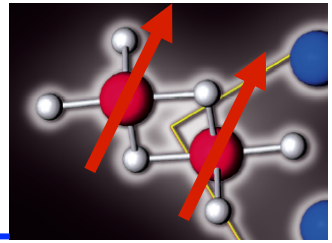
- $TlCuCl_3$ gapped,singlet ground state
- QCP at H_c = 5.7 T to magnon BEC
- continuous increase of magnetization

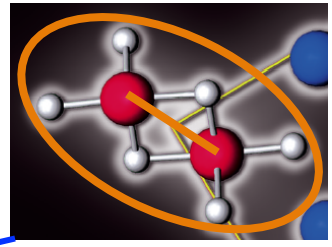
- monoclinic space group $P2_1/c$, Z=4
- Cu^{2+} ions with S = 1/2, two sublattices
- lattice parameters $a \approx 4$ Å, $b \approx 14$ Å, $c \approx 9$ Å, $\beta \approx 96°$

- A. Oosawa *et al.*, J. Phys.: Condens. Matter **11**, 265 (1999).
- H. Tanaka et al., Physica B **237-238**, 120 (1997).
- A. Oosawa *et al.*, Phys. Rev. B **66**, 104405 (2002).

Dispersion of Singlet-Triplet Excitations

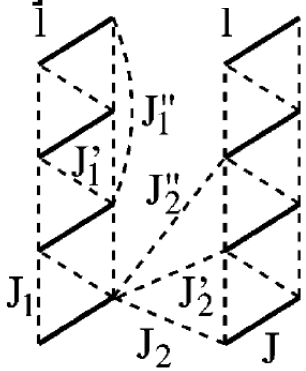


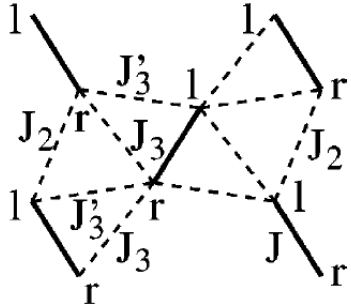




3D coupled dimers (bond operators):

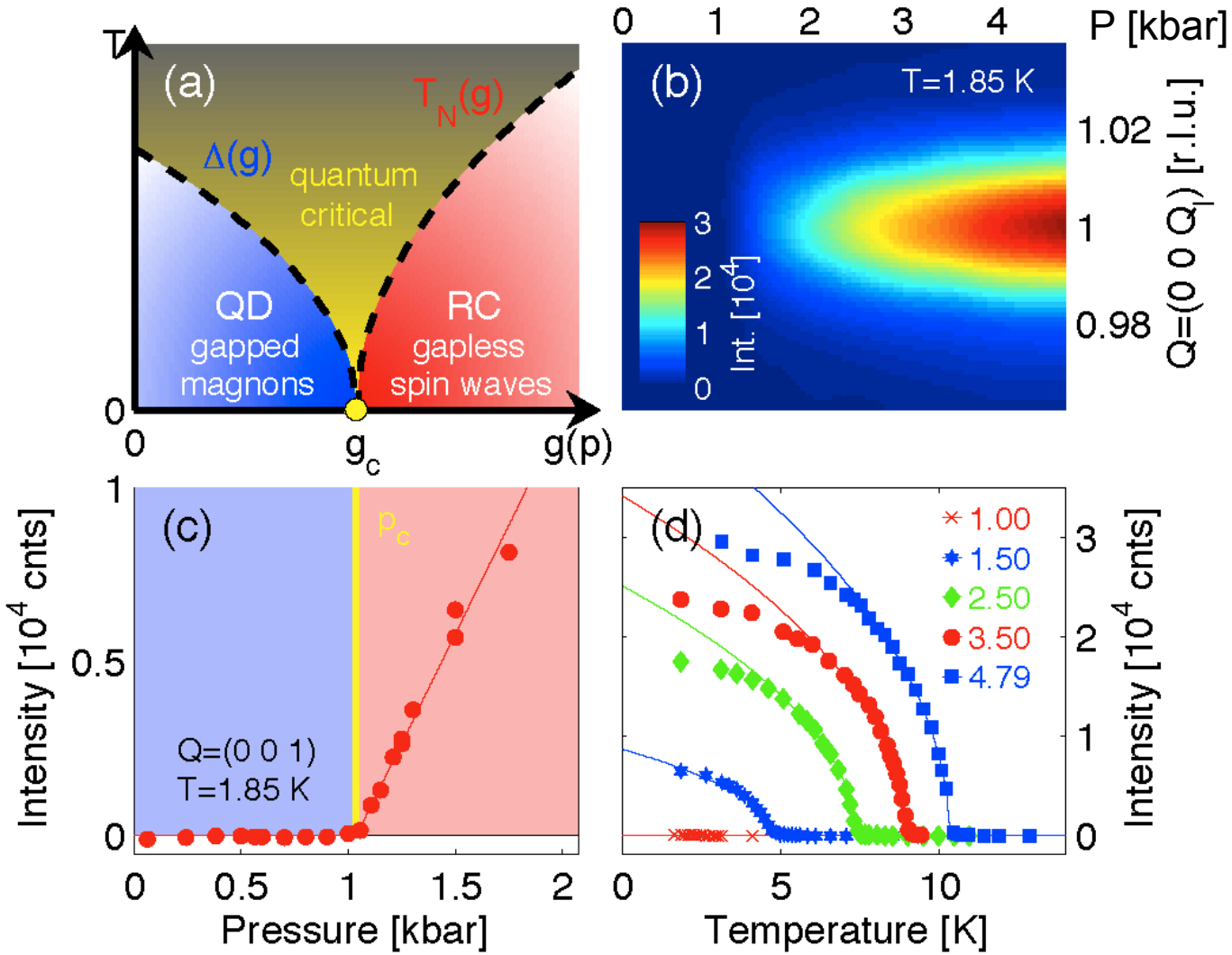
J=5.50, J_{1t}=-0.43, J_2"=3.16, J_{3t}=0.91 [meV]





- N. Cavadini *et al.*, Phys. Rev. B 63, 172414 (2001).
- N. Cavadini *et al.*, J. Phys.: Condens. Matter 12, 5463 (2000).
- M. Matsumoto *et al.*, Phys. Rev. B 69, 054423 (2004).

Pressure-tuned Quantum Critical Point



- Ch. Rüegg *et al.*, Phys. Rev. Lett. **100**, 205701 (2008).
- A. Oosawa *et al.*, JPSJ. **73**, 1446 (2004).

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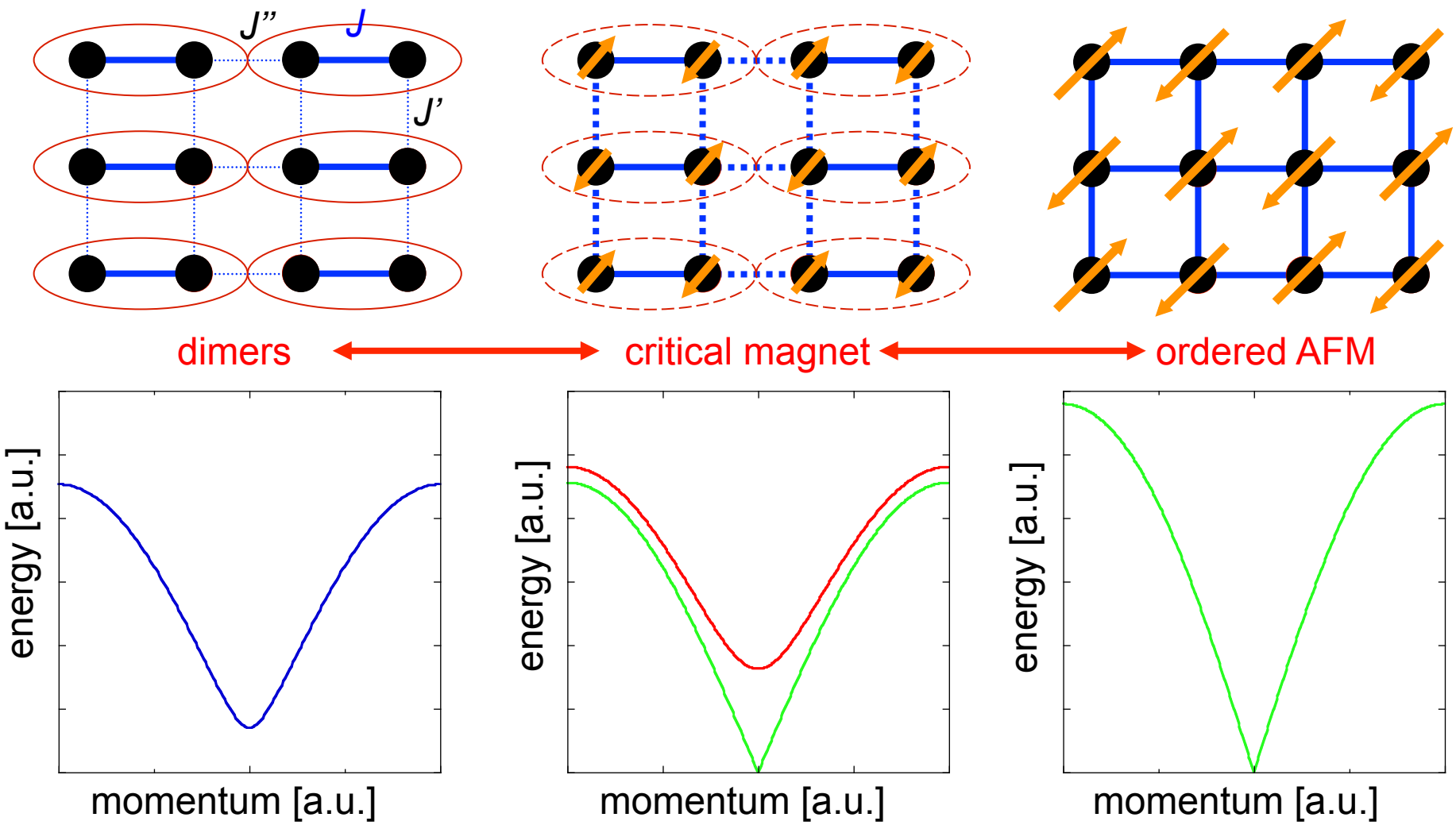
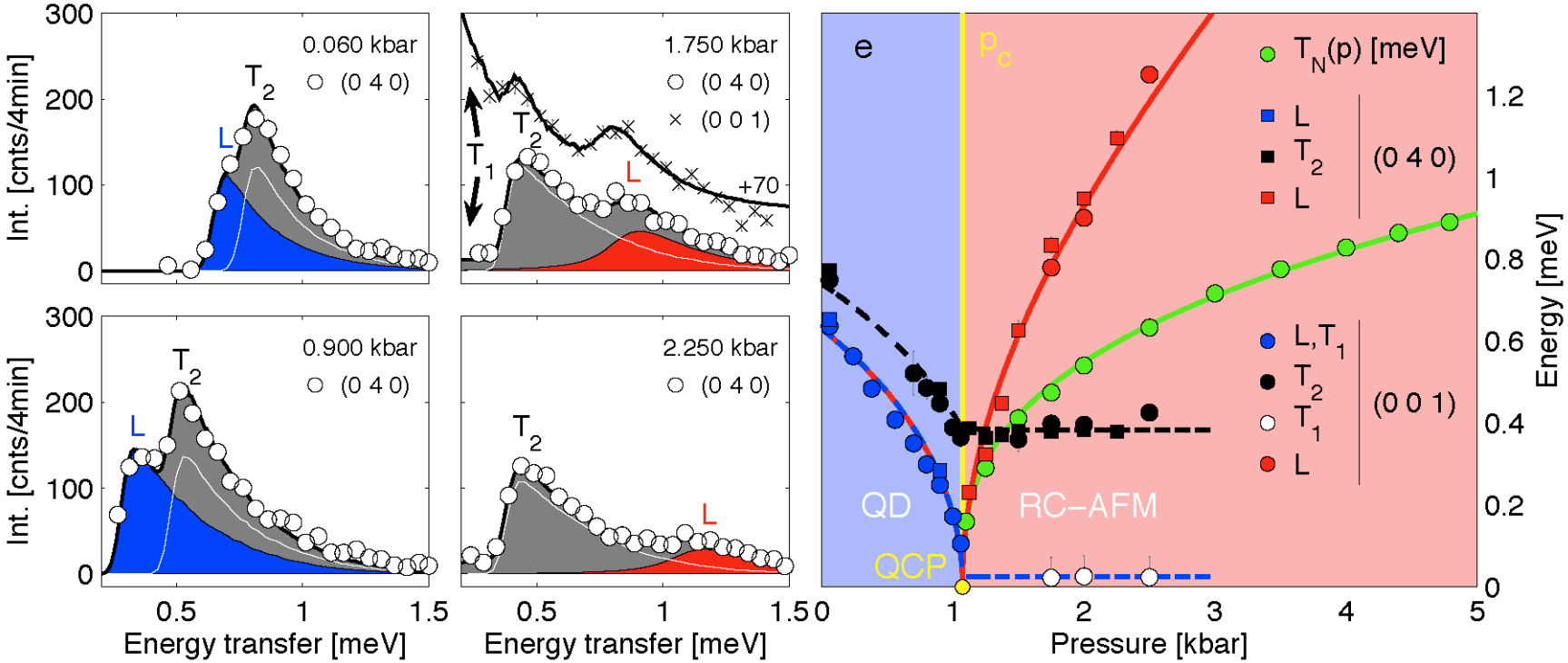


S = 1 triplet excitations

AFM spin waves, 'Higgs'

AFM spin waves

Pressure-control of Elementary Excitations



$T_{1,2}$: transverse spin wave modes
L: longitudinal amplitude mode

QD: Quantum Disordered
RC-AFM: Renormalized-Classical Antiferromagnet

$$\mathcal{H} = \sum_i J(p)\mathbf{S}_{i,l}\cdot\mathbf{S}_{i,r} + \sum_{ij;m,m'=l,r} J_{ij}(p)\mathbf{S}_{i,m}\cdot\mathbf{S}_{j,m'}$$

- P. Merchant *et al.*, Nat. Phys. **10**, 373 (2014).
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- linear pressure dependence with small quadratic correction
- J increasing, J_2 decreasing (A. Oosawa, $KCuCl_3$)

$$J_2(p) = J_2(1 + A_2 p + B_2 p^2)$$

- exchange anisotropy (0.8, 0.002)%

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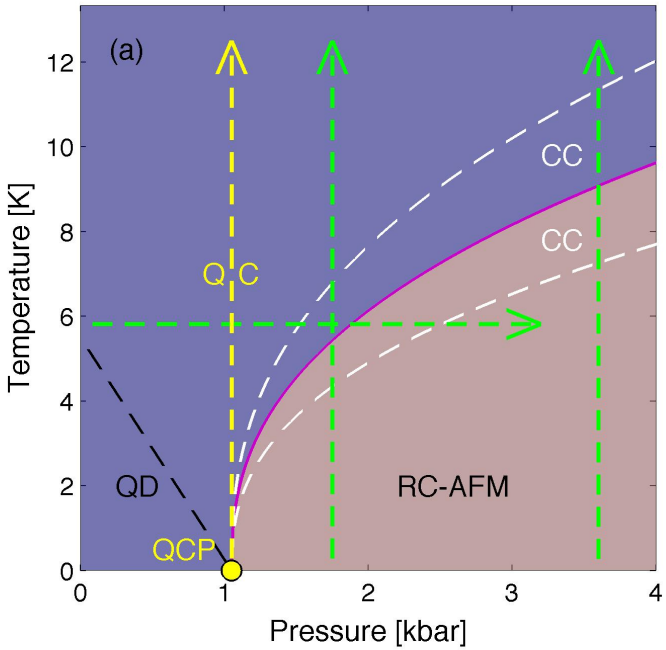
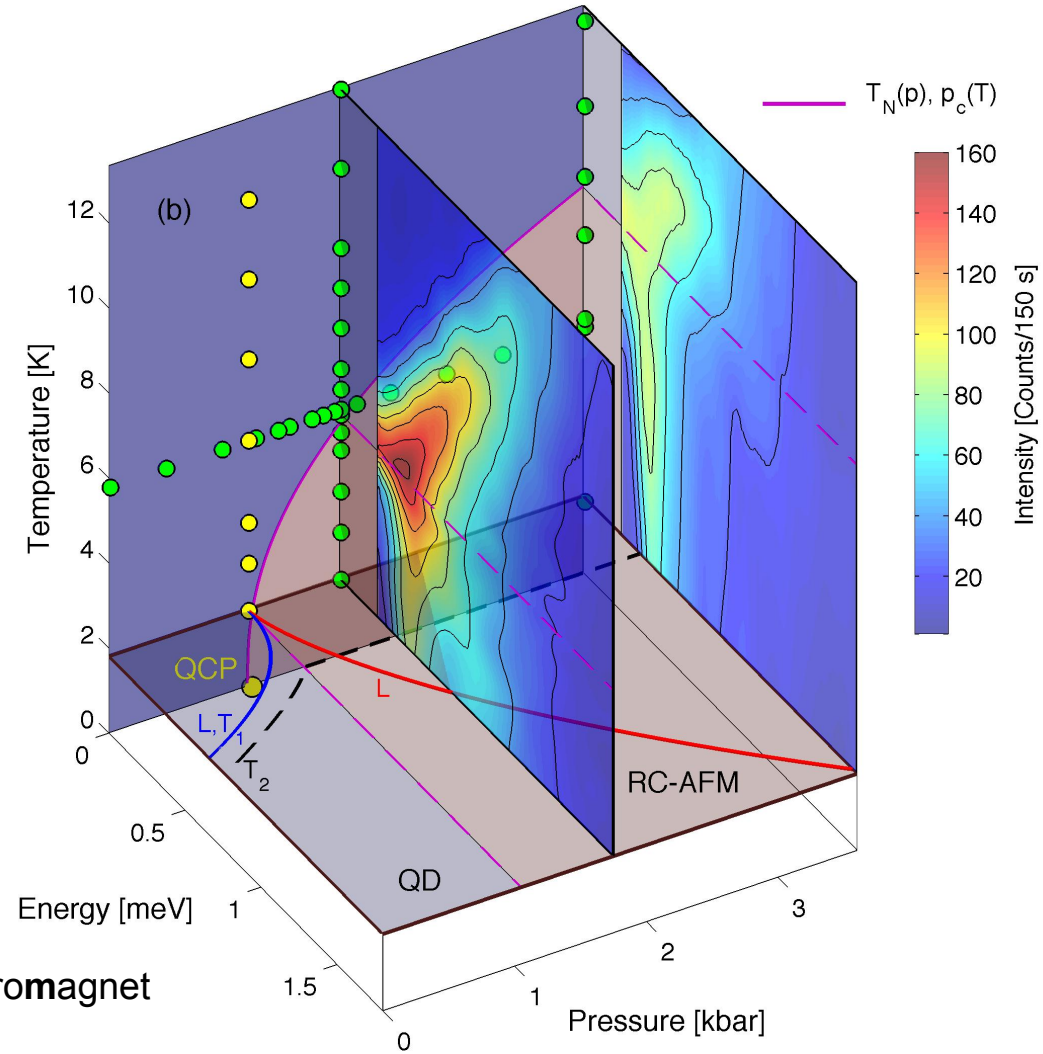
Quantum vs. Thermal Fluctuations





QCP: **Q**uantum **C**ritical **P**oint
QD: **Q**uantum **D**isordered
RC-AFM: **R**enormalized-**C**lassical **A**ntiferro**m**agnet
CC: **C**lassical (Thermal) **C**ritical

- P. Merchant, B. Normand, K. W. Krämer, M. Boehm, D.F. McMorrow, and Ch. Rüegg, Nat. Phys. **10**, 373 (2014).

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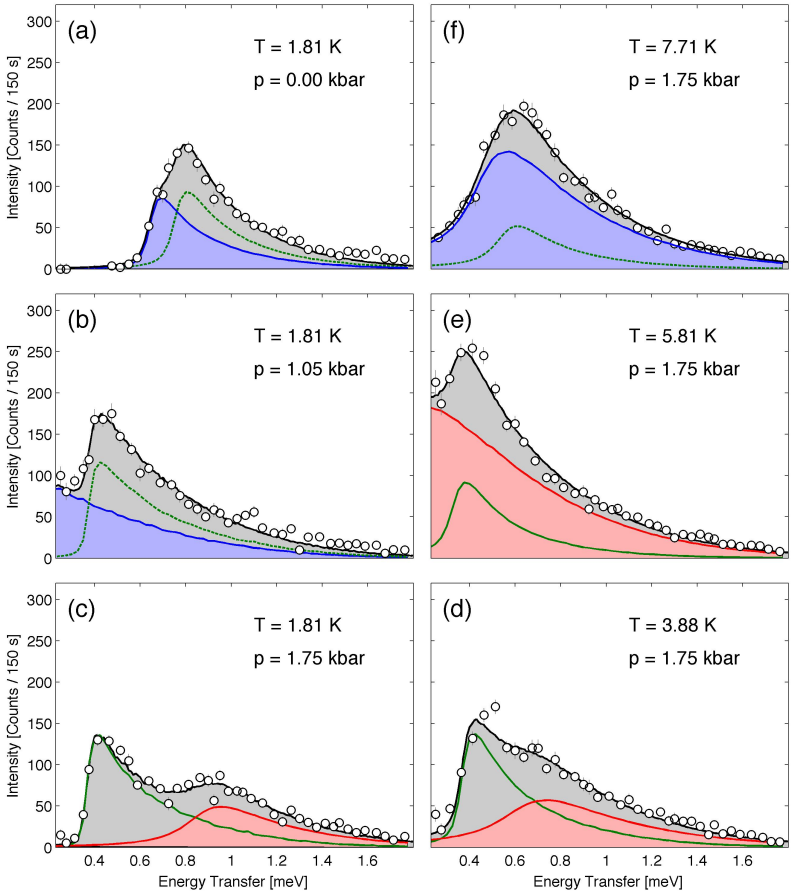

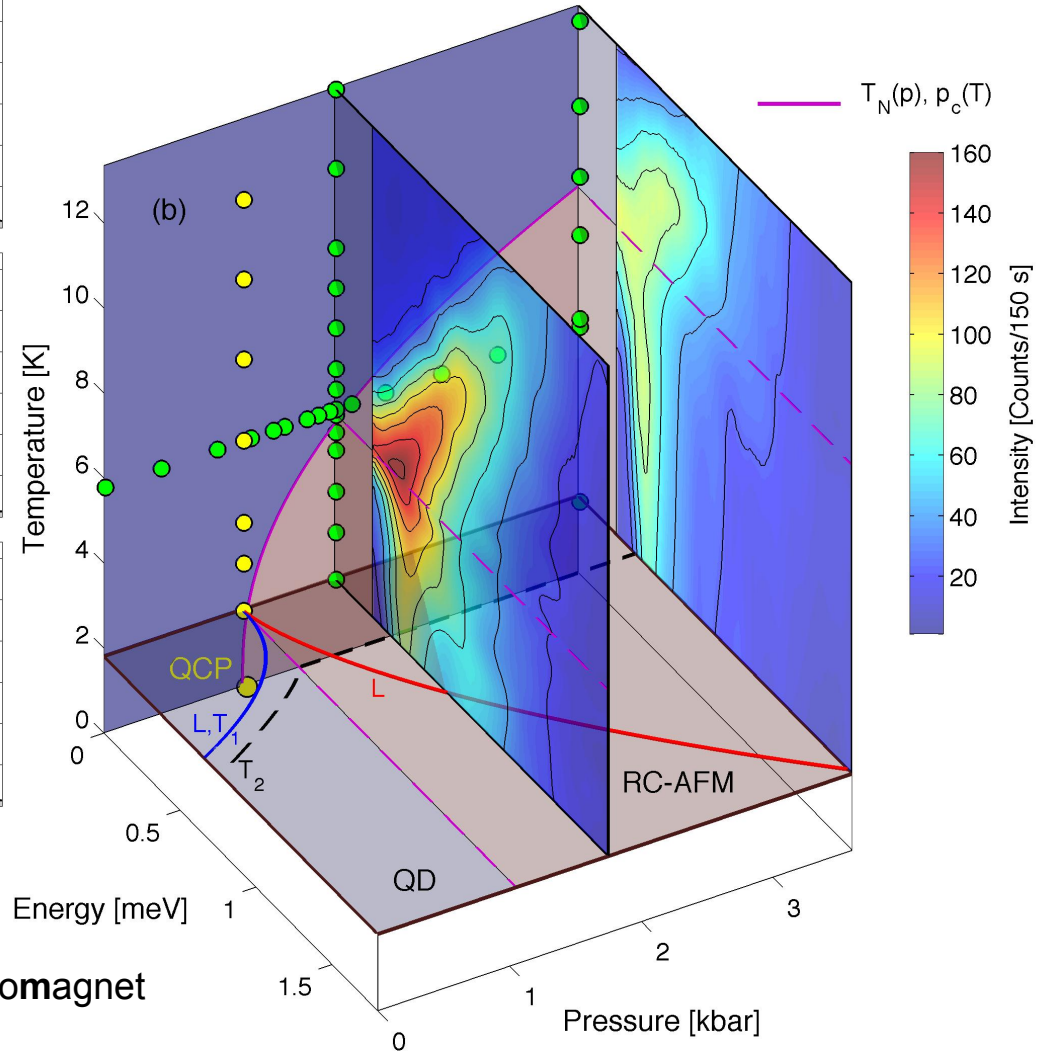

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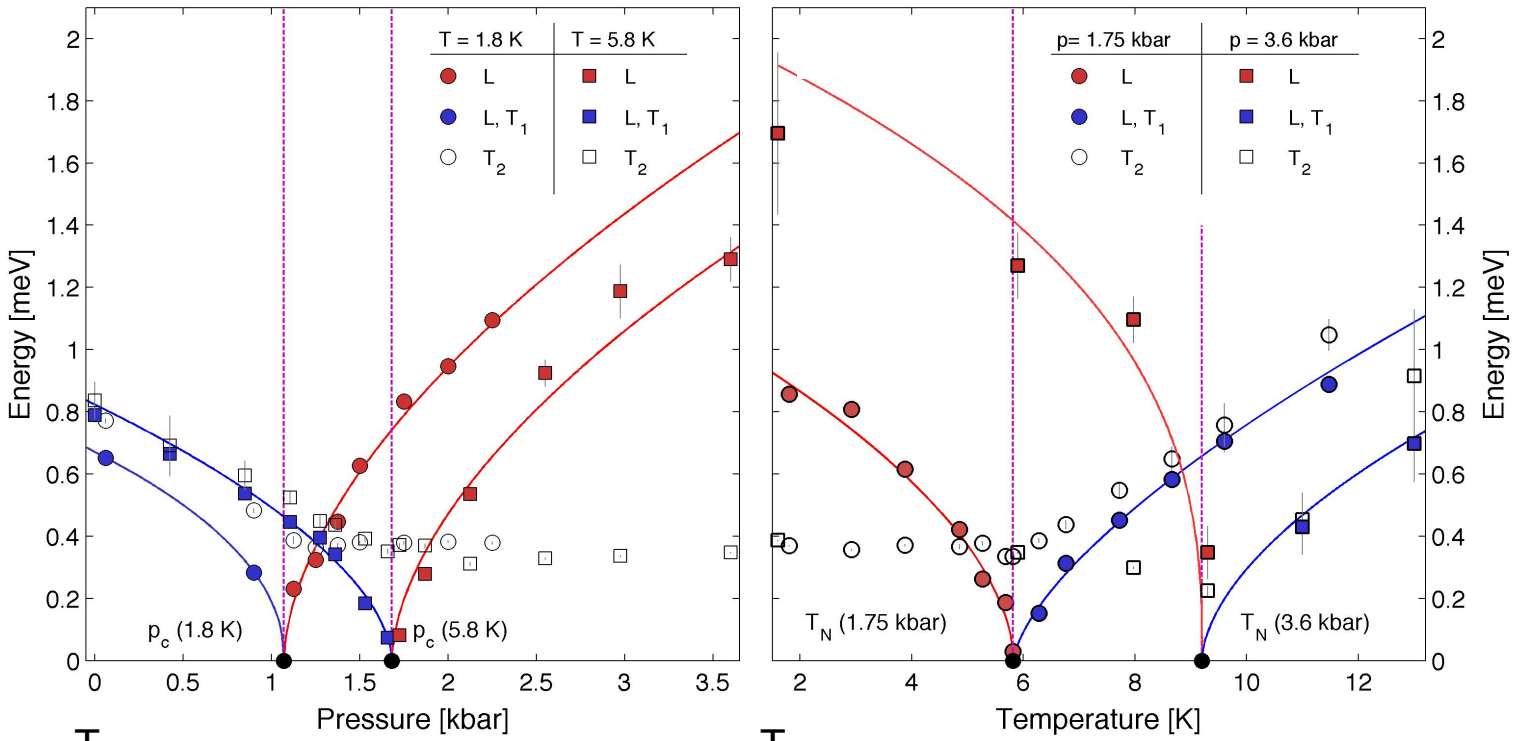

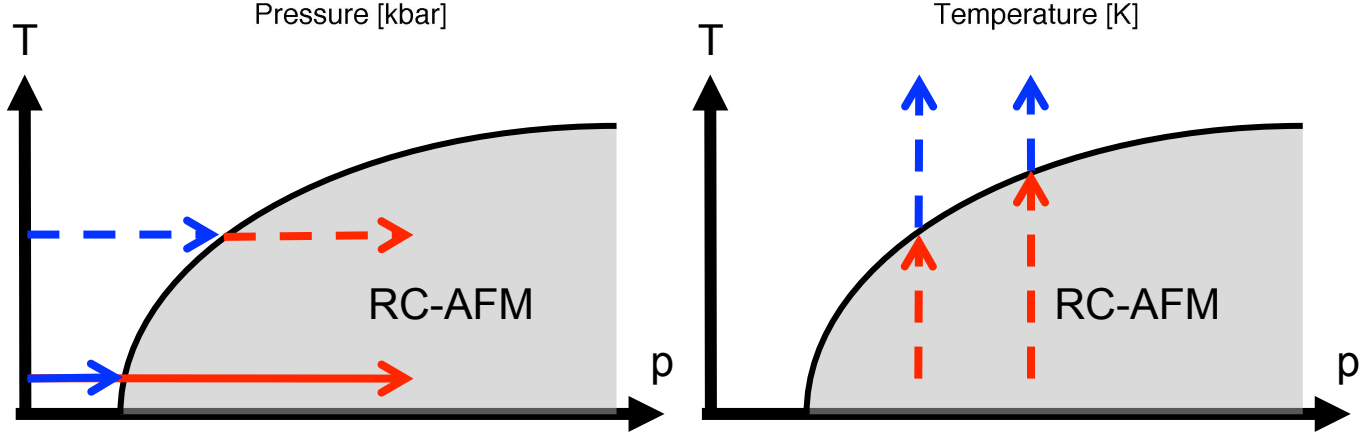

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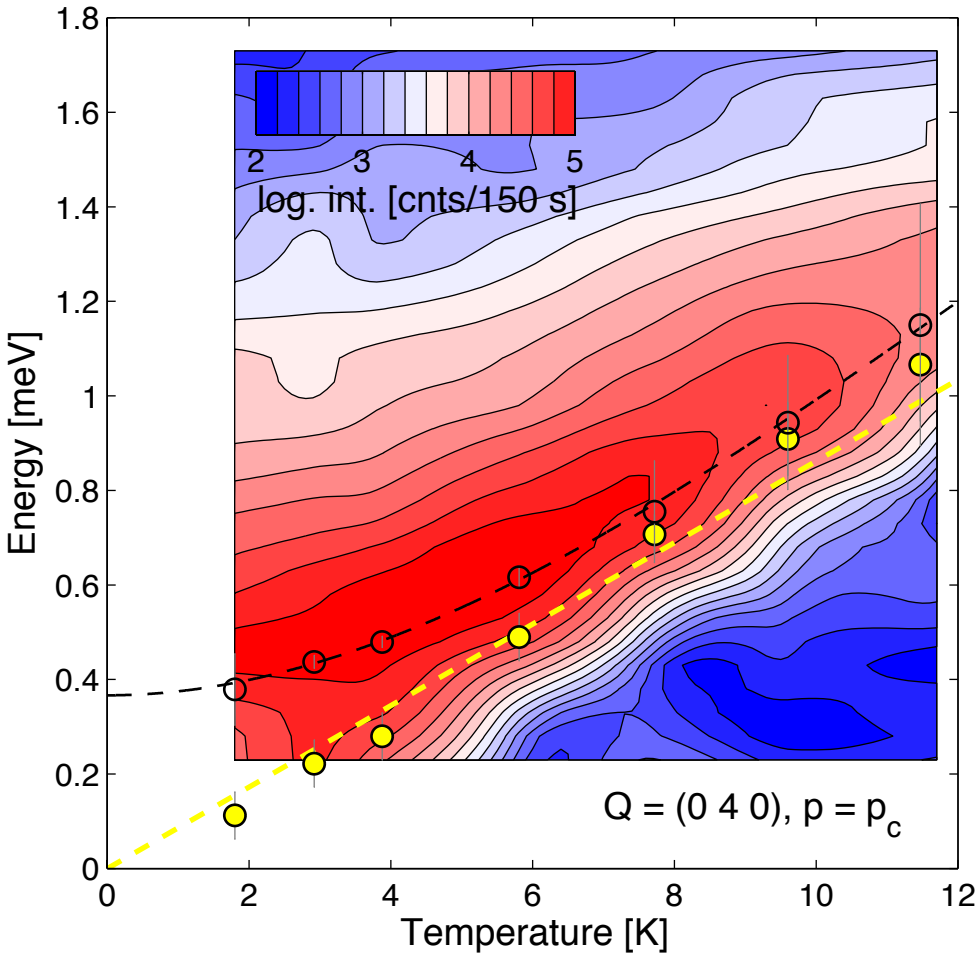
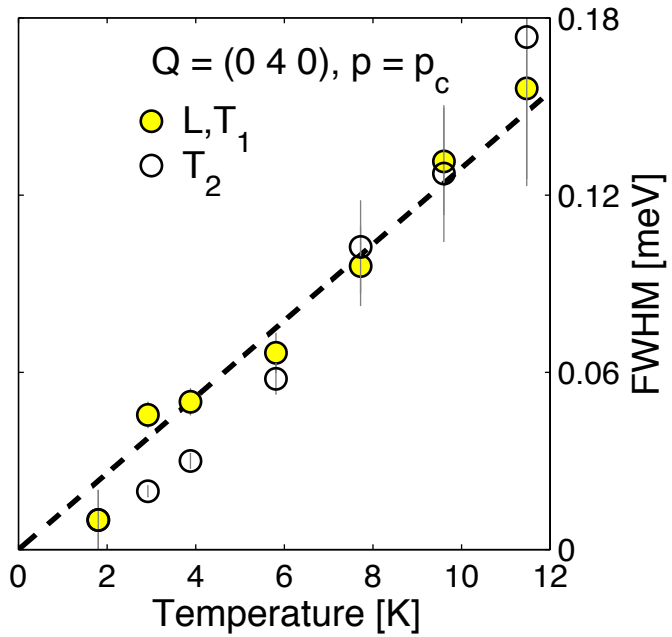




QUANTUM CRITICALITY

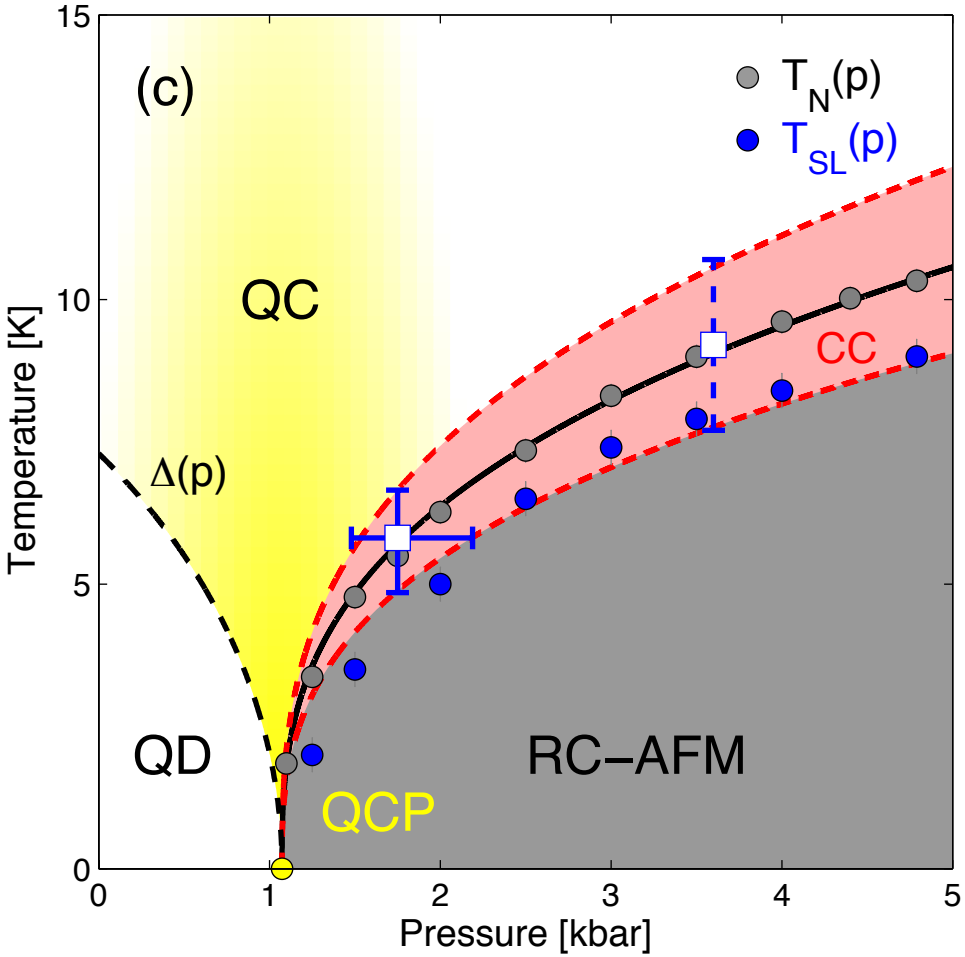
1) Lorentzian lineshape with 'gap'

2) ω/T scaling of excitation energy

3) ω/T scaling of FWHM

- P. Merchant *et al.*, Nat. Phys. **10**, 373 (2014).

Quantum vs. Thermal Fluctuations



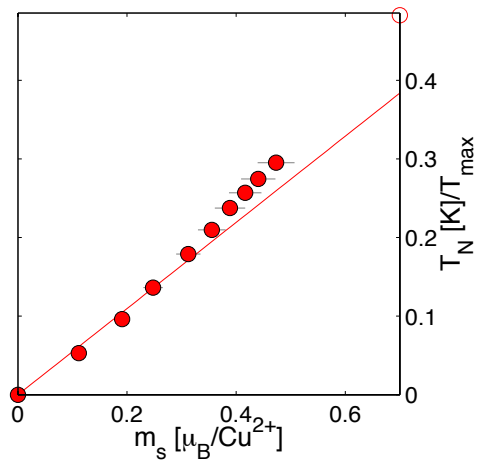
QUANTUM vs. THERMAL MELTING

1) Similar effect on magnetic fluctuations

2) Narrow classical critical (CC) regime
   - dynamics (long. mode)
   - static order (critical exp.)

3) Universal proportionality of T_N and m_s

4) Independent quantum and thermal fluctuations near this QCP



- P. Merchant *et al.*, Nat. Phys. **10**, 373 (2014).
- S. Jin and A.W. Sandvik, PRB **85**, 020409 (2012).
- Qin, Normand, Sandvik, Meng (2015).

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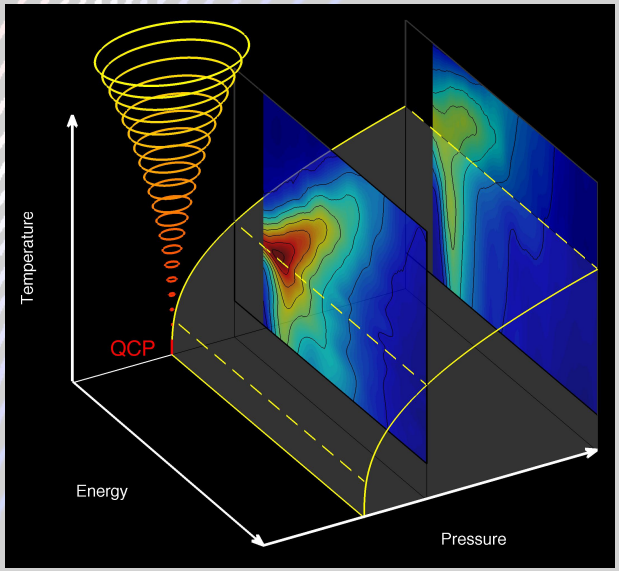
Conclusions

Neutron High-Pressure Experiments

- wide pressure and temperature range now available (150 kbar/4 K, 30 kbar/0.3 K)
- limited by sample volume (no DACs, but can do spectroscopy)

Quantum and thermal melting of order in Quantum Magnets

- direct control of exchange interactions and ground state by pressure
- new elementary excitation (amplitude *Anderson-Higgs* mode) emerges at QCP
- test of field theory predictions
- similarity of quantum and thermal melting
- QC regime has excitation ‘gap’
- QC scaling form is Lorentzian
- universal proportionality of T_N and m_s



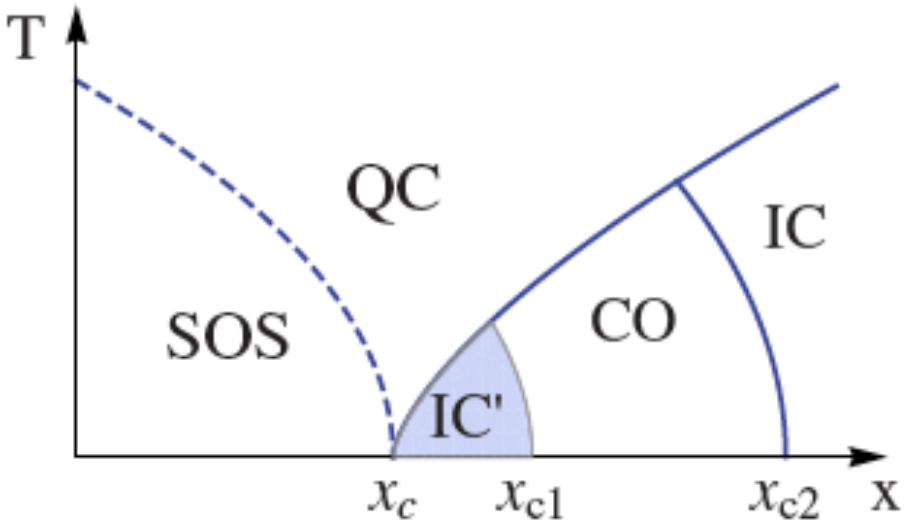
P. Merchant *et al.*, Nat. Phys. **10**, 373 (2014).

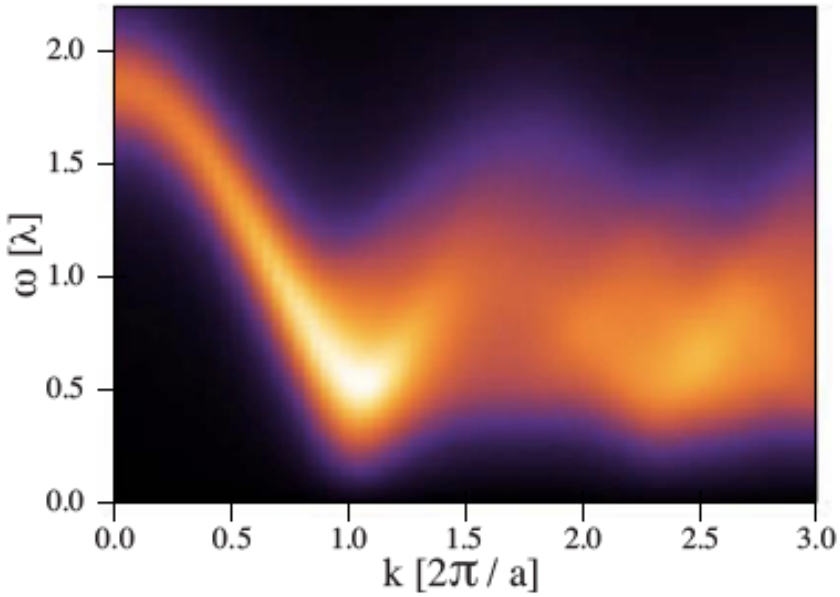
Systems with Effective Singlet Ground-States

PRL **102,** 096406 (2009) PHYSICAL REVIEW LETTERS week ending 6 MARCH 2009

Spin-Orbital Singlet and Quantum Critical Point on the Diamond Lattice: $FeSc_2S_4$

Gang Chen,[1] Leon Balents,[2] and Andreas P. Schnyder[2]
[1]*Physics Department, University of California, Santa Barbara, California 93106, USA*
[2]*Kavli Institute for Theoretical Physics, University of California, Santa Barbara, California 93106, USA*
(Received 3 October 2008; published 5 March 2009)





• A. Biffin, O. Zaharko, R. Coldea, Ch. Rüegg, *et al.*

Systems with Effective Singlet Ground-States

PRL **111,** 197201 (2013) PHYSICAL REVIEW LETTERS week ending 8 NOVEMBER 2013

Excitonic Magnetism in Van Vleck–type d^4 Mott Insulators

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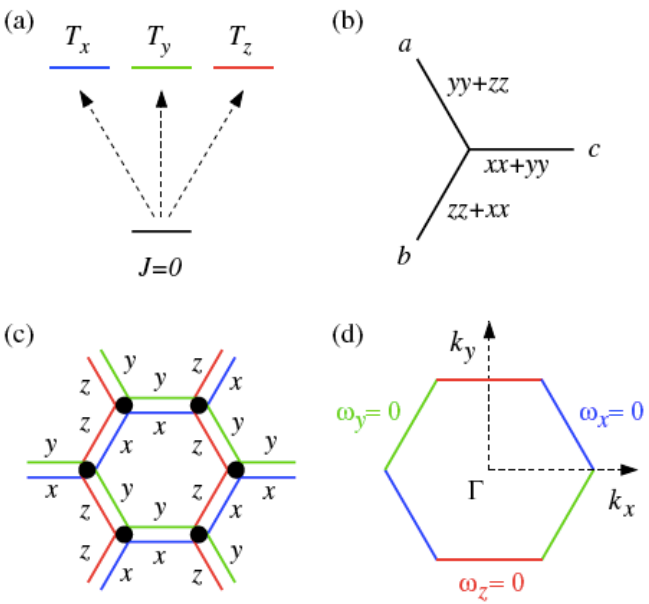
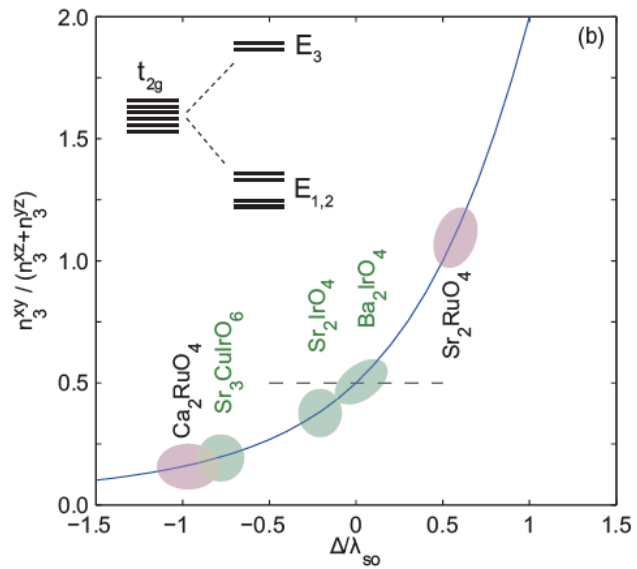
In Mott insulators with the t_{2g}^4 electronic configuration such as of Re^{3+}, Ru^{4+}, Os^{4+}, and Ir^{5+} ions, spin-orbit coupling dictates a Van Vleck–type nonmagnetic ground state with an angular momentum $J = 0$, and the magnetic response is governed by gapped singlet-triplet excitations. We derive the

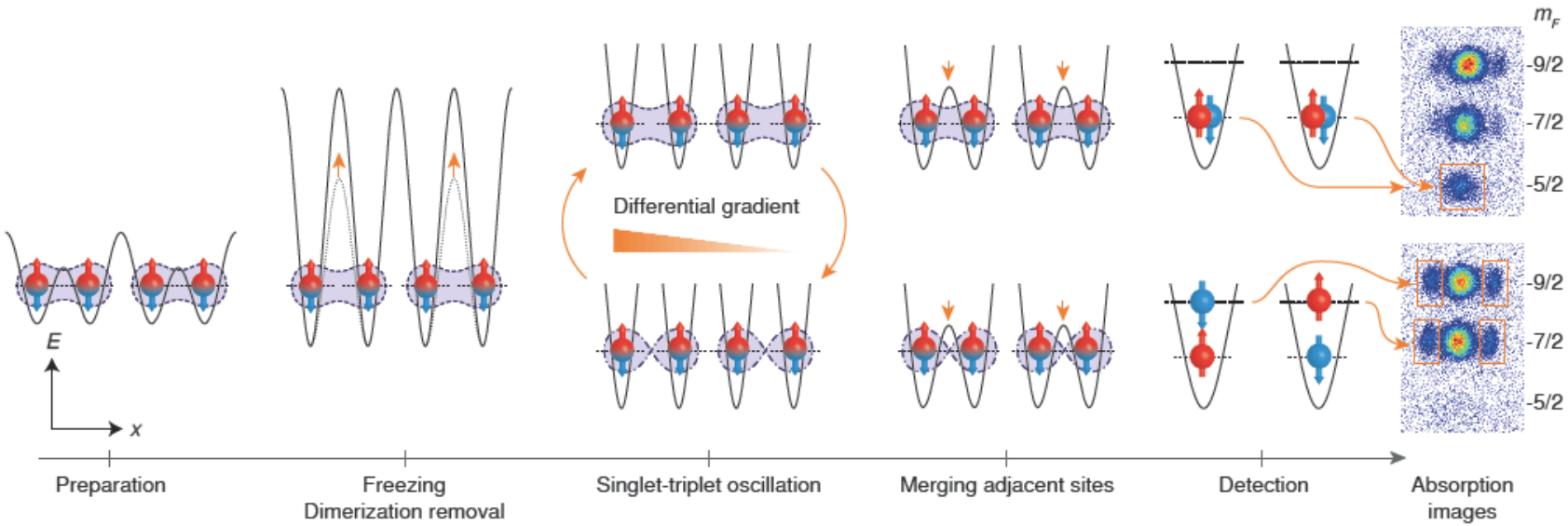




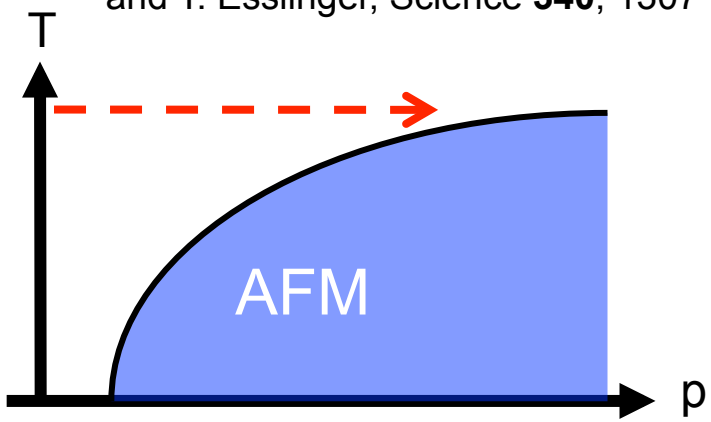
• C. Fatuzzo, *et al.* PRB **81**, 155104 (2015)

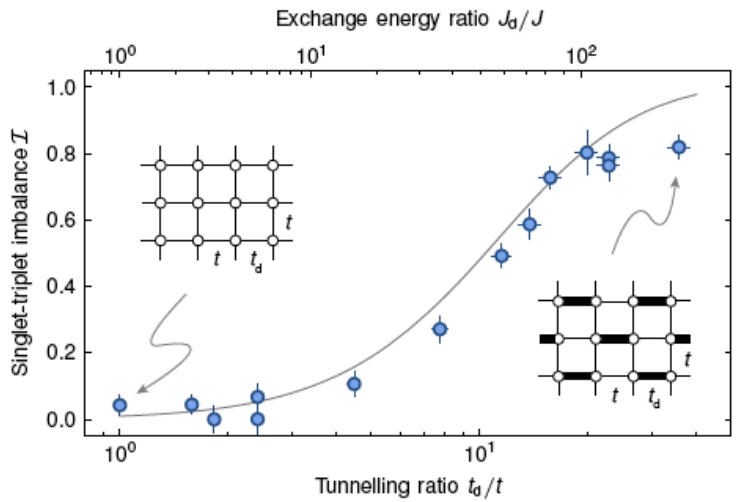
Quantum Dimerized to AFM with Cold Atoms

Quantum magnetism of ultracold fermions in an optical lattice



D. Greif, T. Uehlinger, G. Jotzu, L. Tarruell, and T. Esslinger, Science **340**, 1307 (2013).





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  A. Sandvik (Boston)
  T. Esslinger (ETH Zurich)

- **Neutron Scattering Centers**
  SINQ (B. Roessli) and ILL (M. Boehm)
  FRM-II (T. Keller)

- **Samples**
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