

Quantum Criticality and Superconductivity in Spin and Charge Systems

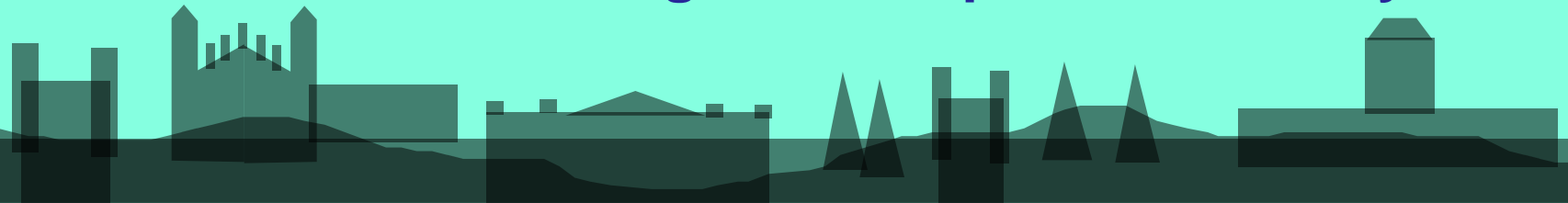
Siddharth S Saxena

Shoenberg Laboratory for Quantum Matter , Cavendish Laboratory, University of Cambridge, UK

Cavendish Laboratory

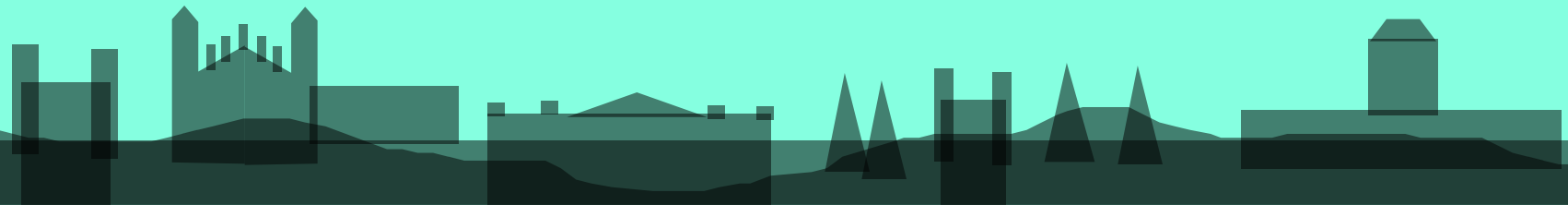


Research and Teaching in the Department of Physics



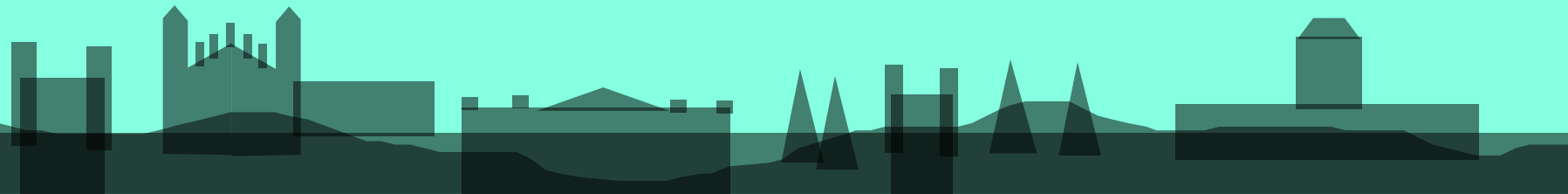
Snapshot

- 100 – 120 MSci (4-year) undergraduates annually
- 60 – 70 PhD's annually
- ~ 250 postdocs and research staff
- ~ 80 research support staff
- 75 teaching faculty + 33 staff level fellows
- £14M annual research grant expenditure
- Institute of Astronomy – ~20 faculty, 50 graduate students, 50 postdocs
- 37 Noble Prizes



The Role of Physics

- Practical Application of Physics Research for the Benefit of Society
- The Intellectual Underpinning of Physics and all Cognate Sciences
- The Development of Individuals with the Ability to Relate Phenomena to Mathematical Structures
- Physics for Its Own Sake – Insights into the Nature of Our Physical Universe



The Balance of the Programme

- **The core of the Laboratory's programme is experimental physics, supported by theory.**
- **The policy of the Department is to maintain a very powerful core of fundamental physics in all its diversity.**
- **The Department encourages applied and collaborative research in response to the evolution of the basic disciplines and the interests of the staff.**

The Cavendish Laboratory – Research organisation

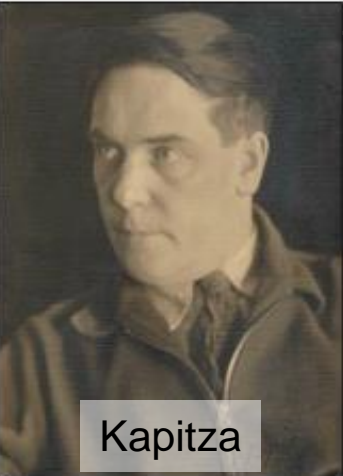
- **High Energy Physics**
- **Astrophysics**
- **Biological and Soft Systems**
- **Semiconductor Physics**
- **Optoelectronics**
- **Microelectronics Research Centre**
- **Quantum Matter**
- **Physics and Chemistry of Solids**
- **Theory of Condensed Matter**
- The policy of the Laboratory is to delegate as much of the responsibility for the research programme to the research groups as possible. They each have their own administrative staffs.
- The central administration aims to allow the research groups to concentrate upon research and teaching.

***Shoenberg Laboratory for Quantum
Matter***

Cavendish Laboratory,

University of Cambridge, UK

The QM



Kapitza



Allen



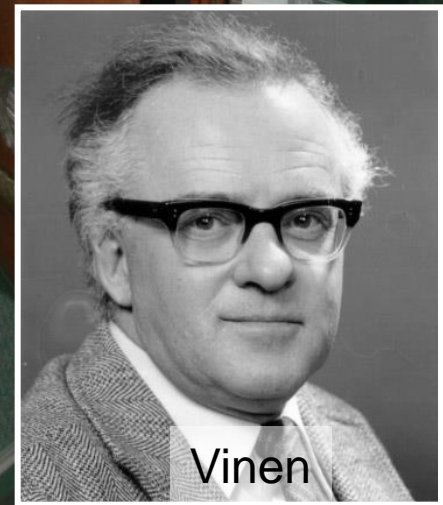
Misener

1938 - Superfluidity in BEC ^4He
(Nobel prize Kapitza 1978)



Mond Laboratory

1956 - Quantised vortices in ^4He (Hall, Vinen)

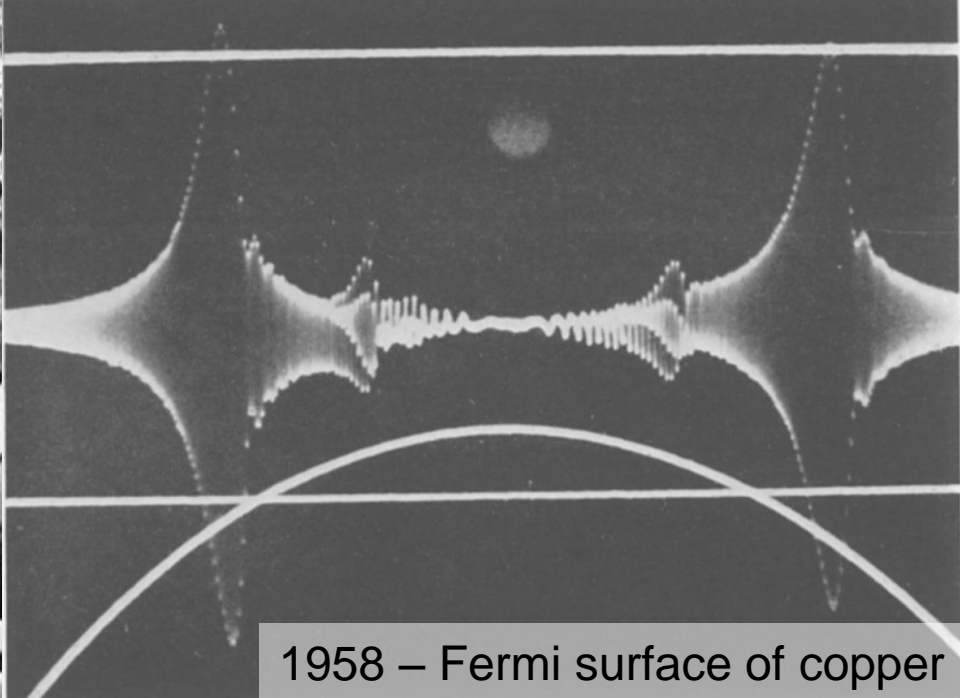


Vinen

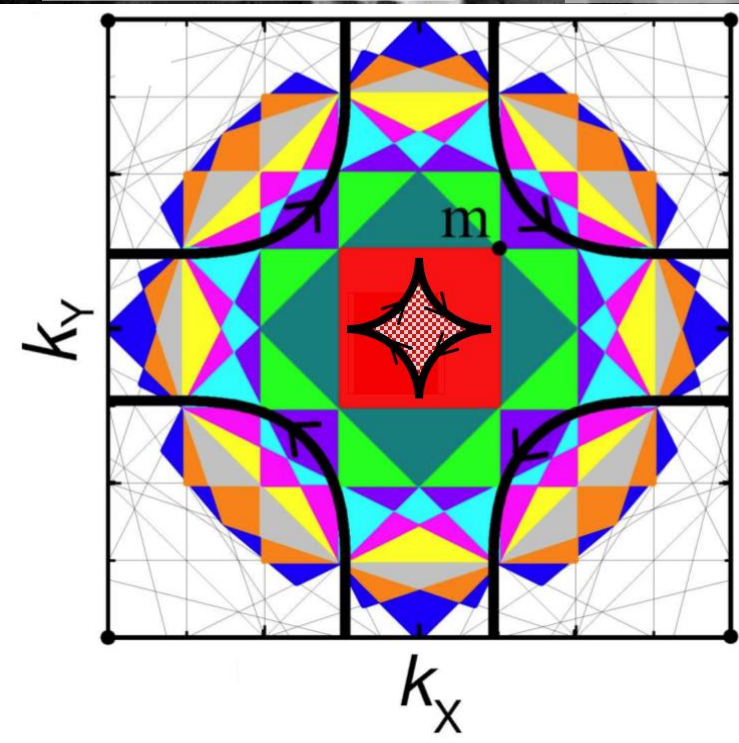


David Shoenberg

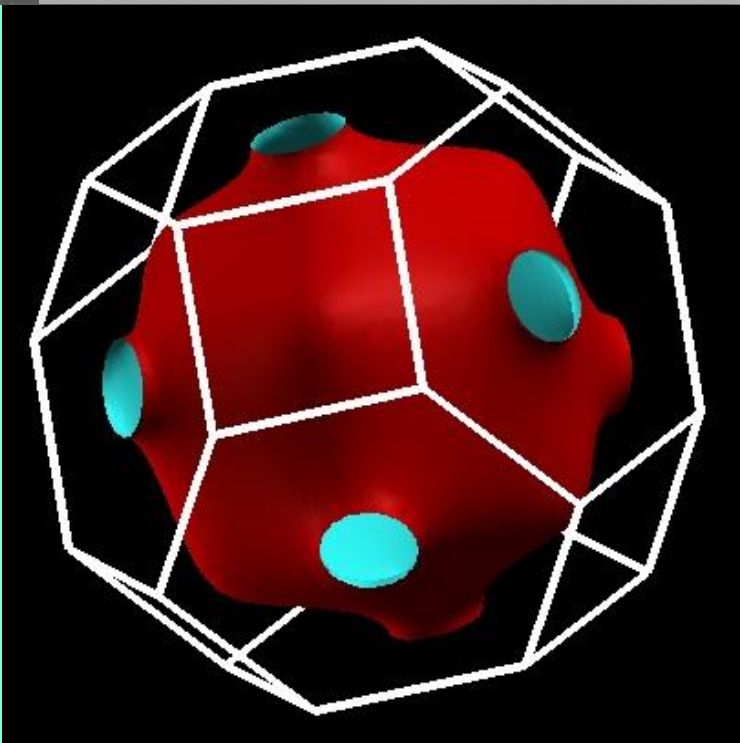
Brian Pippard



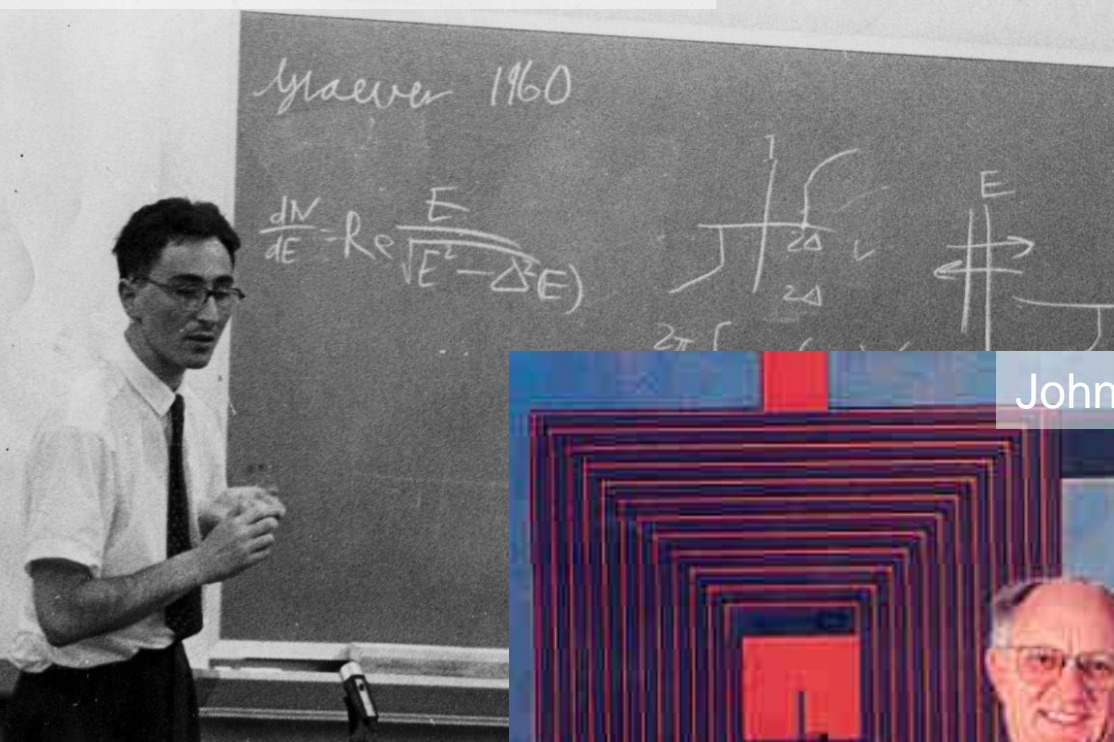
1958 - Fermi surface of copper



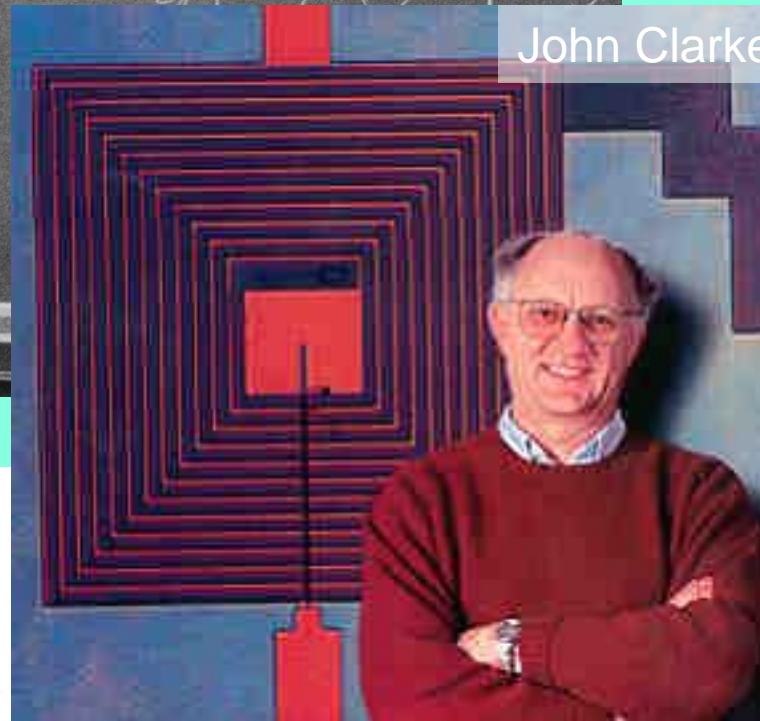
Copper
Copper oxide
($\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$)
[Sebastian *et al.* 2012]



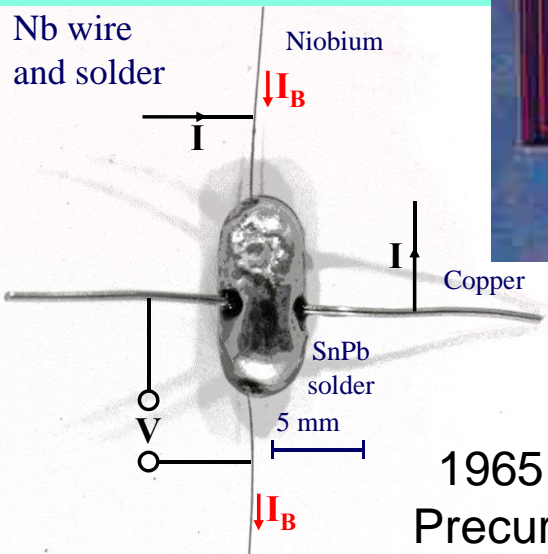
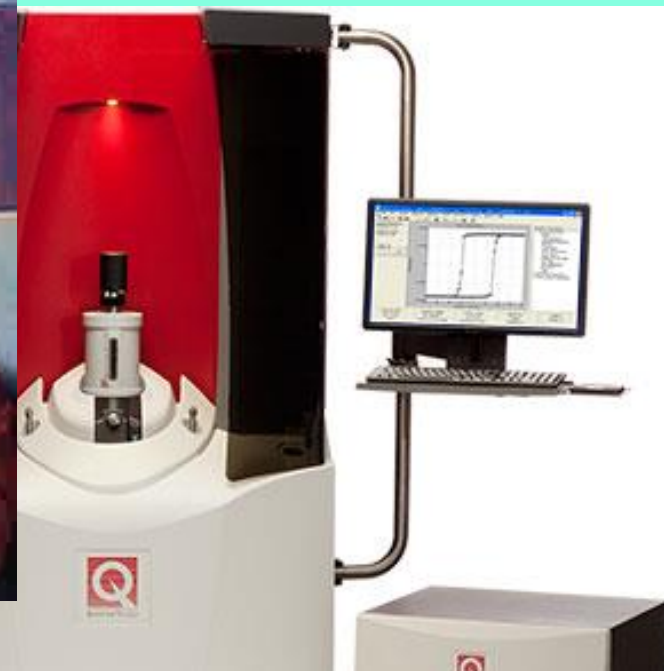
Brian Josephson, Nobel prize 1973



Brian Pippard:
"John, how would you like a voltmeter with a resolution of 2×10^{-15} V in one second?"

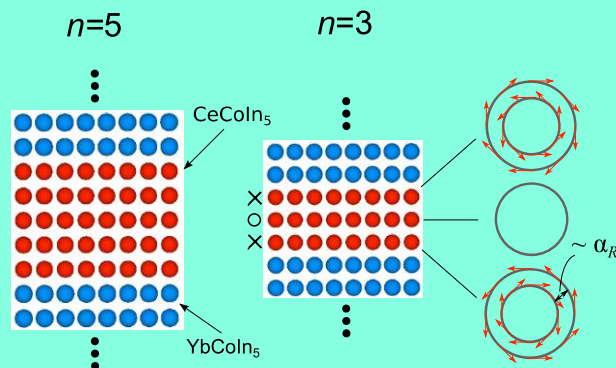


John Clarke

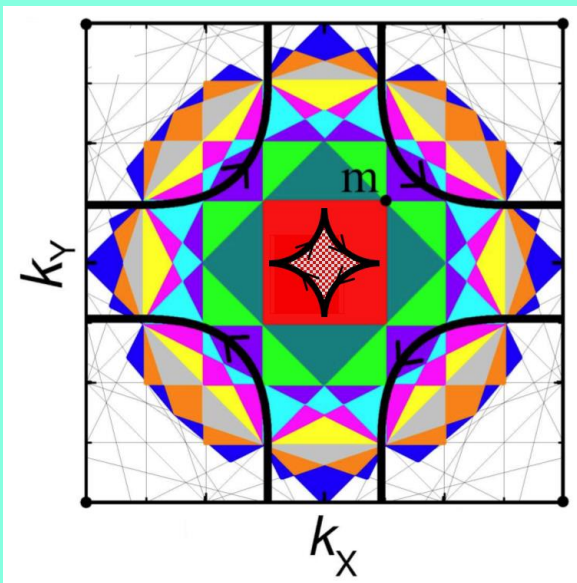


1965 - 'SLUGS':
Precursor SQUIDS

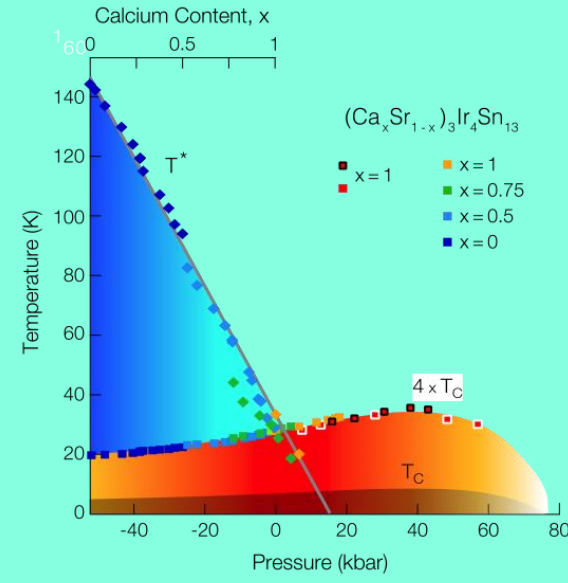
Magnetic Property
Measurement System (MPMS® 3)
The next generation of advanced SQUID magnetometry.



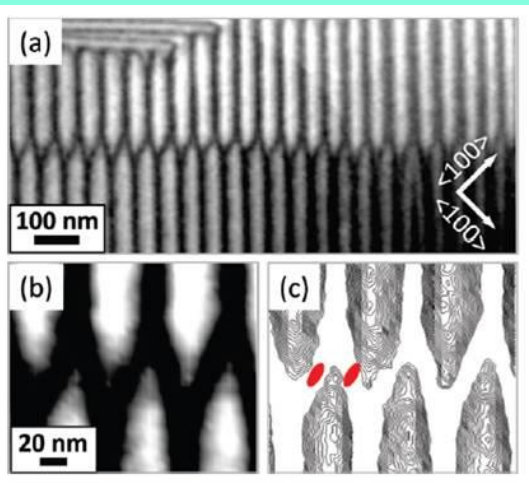
f-metal superconducting heterostructures (PRL 2012)



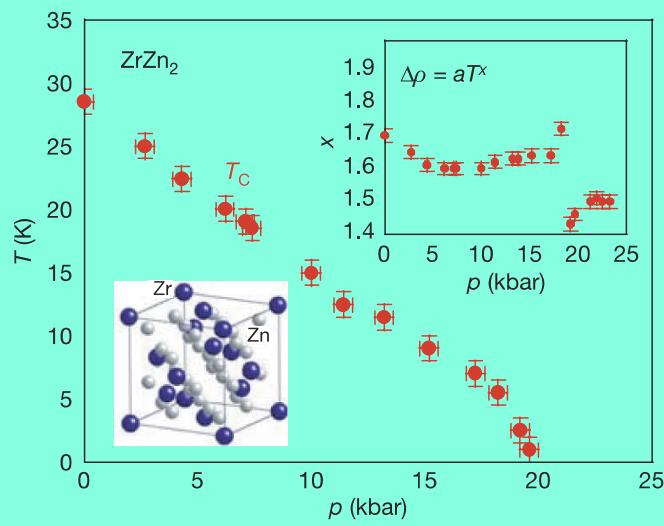
Fermi surface of cuprate superconductors (multiple Nature, PRL, PNAS ...)



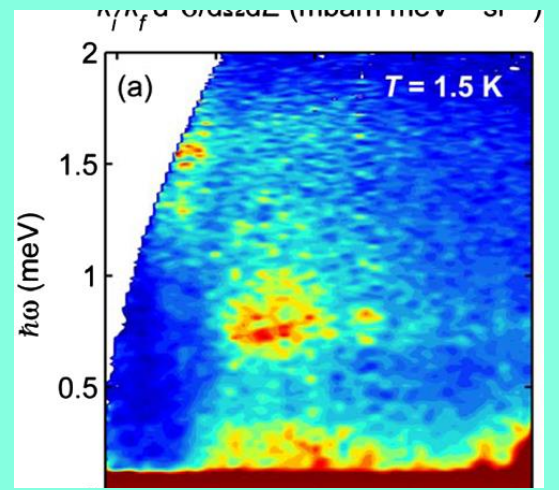
Structural quantum critical point (PRL 2012)



Ferroelectric closure domains (Nature Comm. 2013)



Marginal Fermi liquid (Nature 2008)



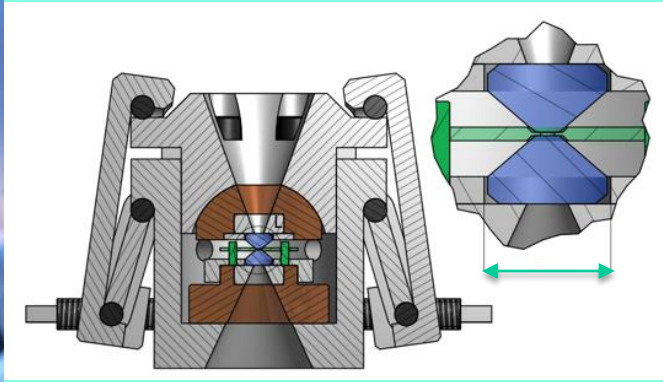
Quantum spin liquid in LiCuSbO4 (PRL 2012)



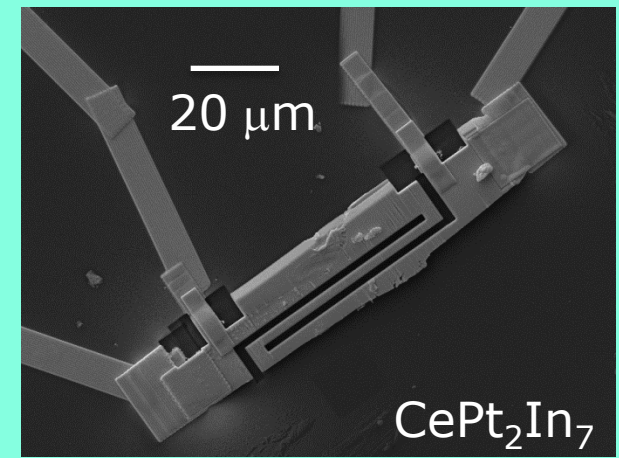
18 Tesla/1 milli-Kelvin
cryomagnet
for Fermi surface studies



Adiabatic
demagnetisation refrigerators
(spin-offs CMR, Camcool)



High pressure devices
(collaboration Diamond)



Sample handling on
the microscale
(spin-offs Camcool)

Collaborators

Cambridge

S.Rowley, L. Spalek, Christos Panagopoulos, S. Haines, P. Nahai-Williamson, M. Dean, R. Smith, A. Kusmartseva, C. Liu, Sam Brown, P. Aireza, H.J. Kim, M. Sutherland, G. Lonzarich

G. Csanyi, P. Littlewood, A. Nevidomskyy, C. Pickard and B. Simons

University College London

M. Ellerby, C. Howard, T. Weller, N. Skipper, A. Waters, K. Rahnejat, N. Shuttleworth, D. McMorrow

Lausanne

A Akrap and Laszlo Forro

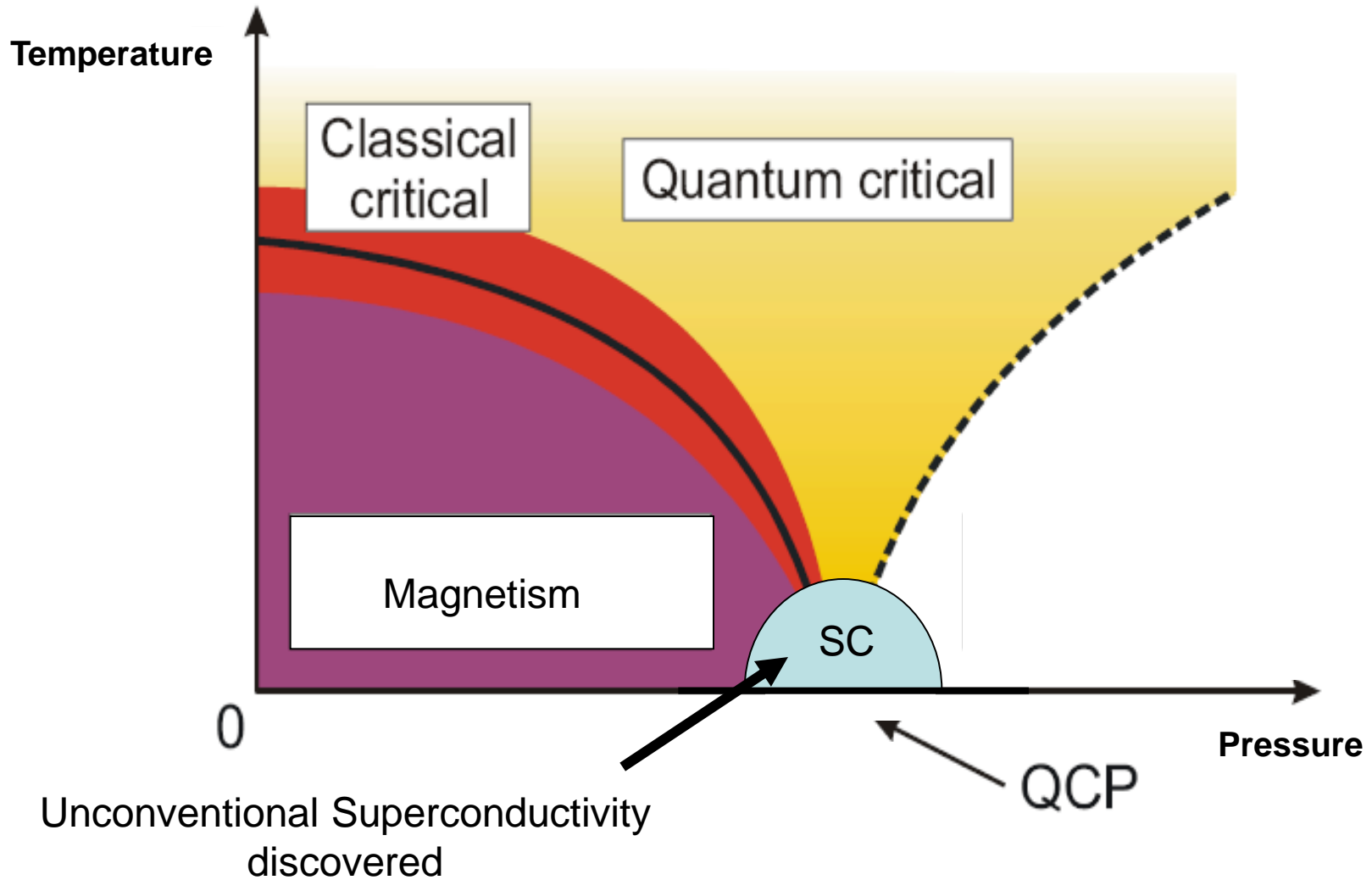
Paris and Milan

Matteo d'Astuto, Claudia Dallera

Sherbrooke

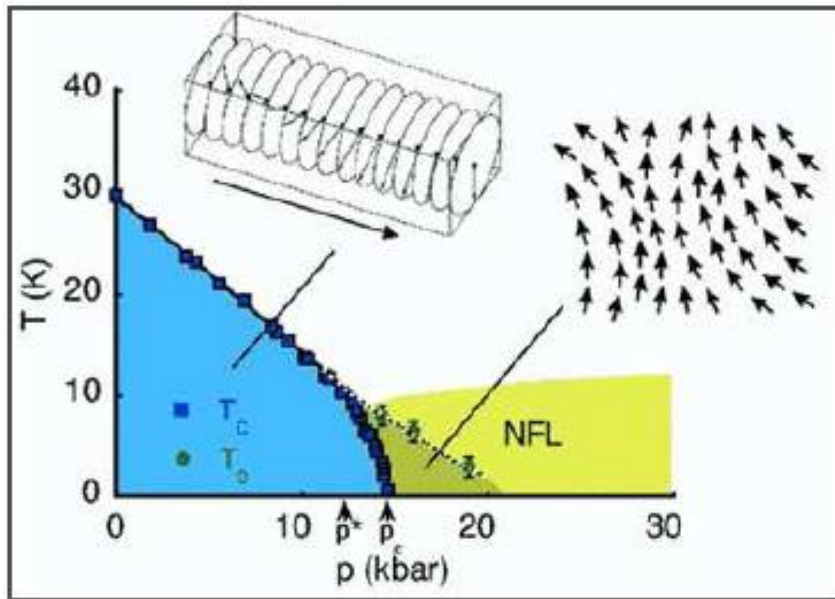
Nicolas Doiron-Leyraud, Louis Taillefer

Quantum Phase Transitions



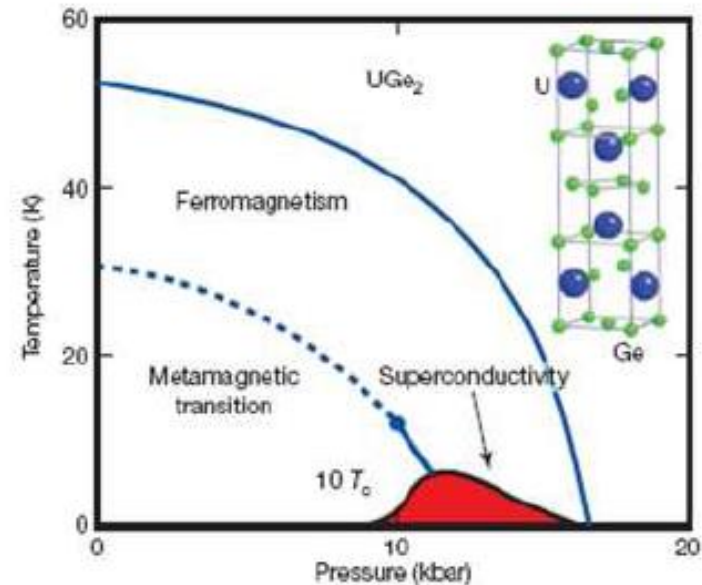
N.D. Mathur et al, Nature, Vol. 394, 39, (1998)
S.S. Saxena et al, Nature, Vol. 406, 587, (2000)

Neighbourhood of quantum phase transitions in metals – emergent states



d-electron metal

Spin skyrmion state in MnSi
on border of ferromagnetism



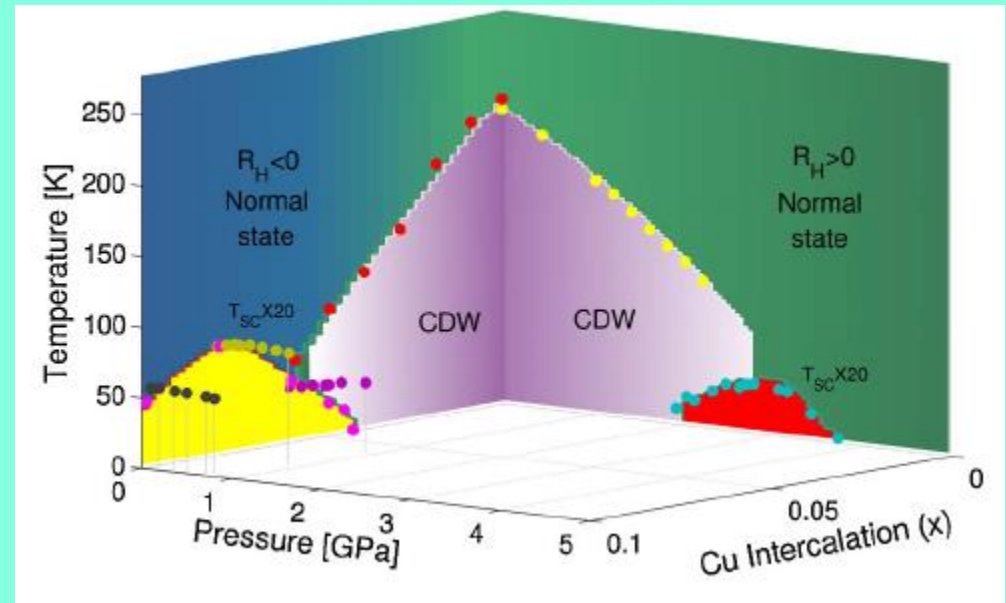
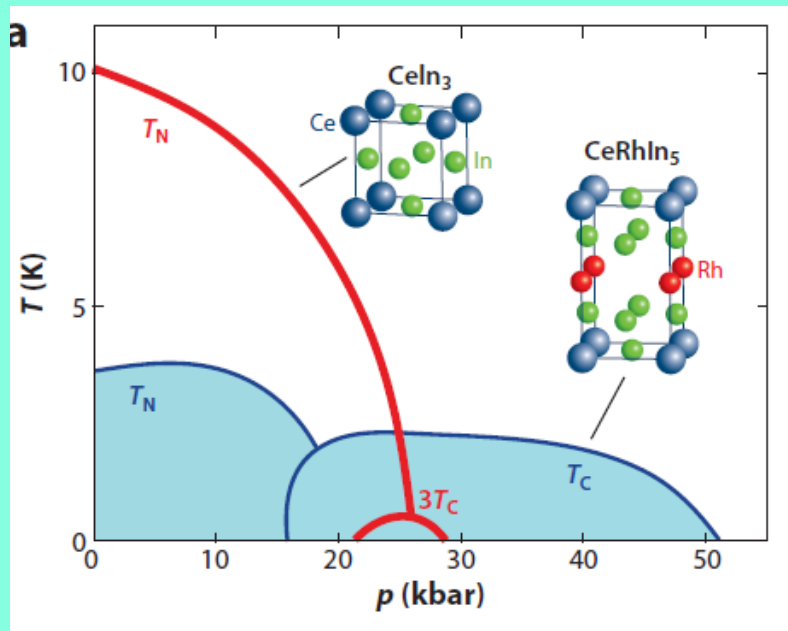
f-electron metal

p-wave superconductor in
heavy fermion UGe₂ on border
of ferromagnetism. Also
superconducting Sr₂RuO₄

Other metallic itinerant-ferromagnets which become superconducting

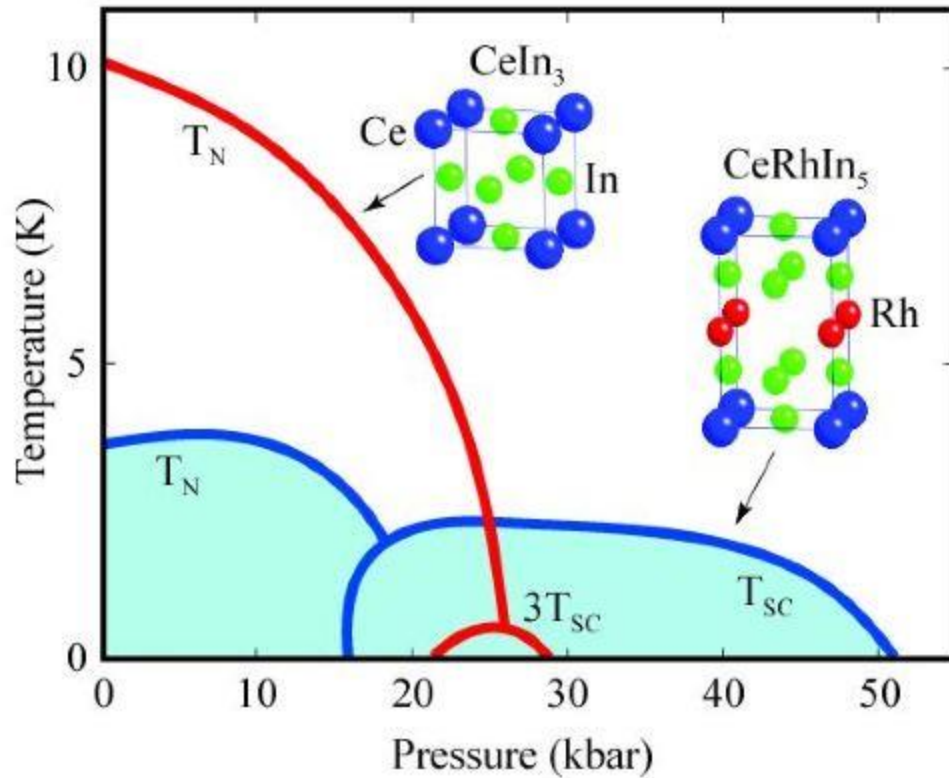
- URhGe, $T_c = 250 \text{ mK}$ (Aoki et al. Nature 2001)
- Fe, $T_{c\text{max}} = 2\text{K}$ (Shimizu et al. Nature 2001)
- UIr, $T_c = 140 \text{ mK}$ (Akazawa et al. JPCM 2004)
- UCoGe (N.T. Huy, et. al. PRL 2007)
- Possibly New Pnictides, Chalcogenides etc

Neighbourhood of quantum phase transitions in metals – emergent states



f-electron metal
d-wave superconductivity on
border of heavy fermion
antiferromagnetism

Superconductivity on border
of charge density wave
transition in TiSe_2



$$\underline{\text{CeIn}_3 : \rho \sim T^{3/2}}$$

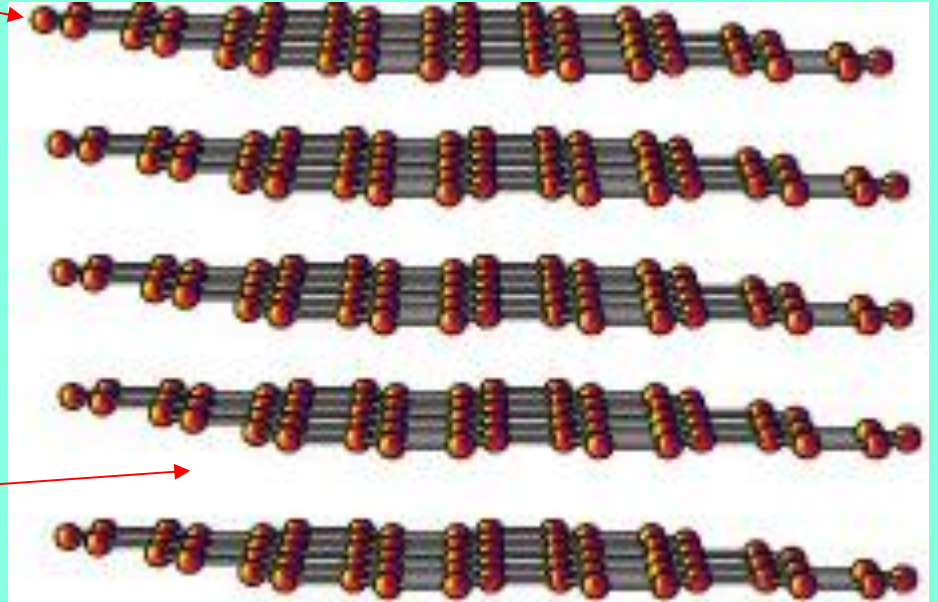
Walker et al. (1997), ...

$$\underline{\text{CeMIn}_5, \text{M} = \text{Co, Rh, Ir} : \rho \sim T}$$

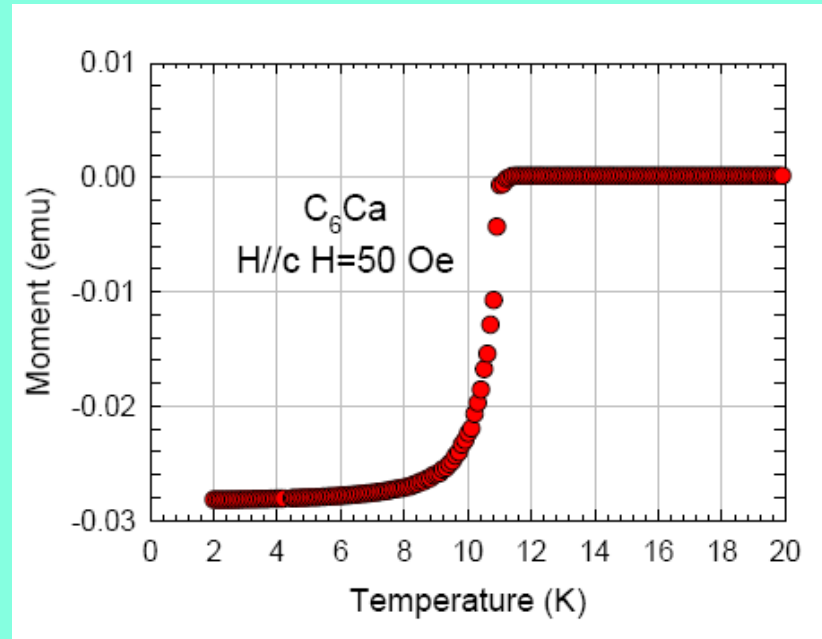
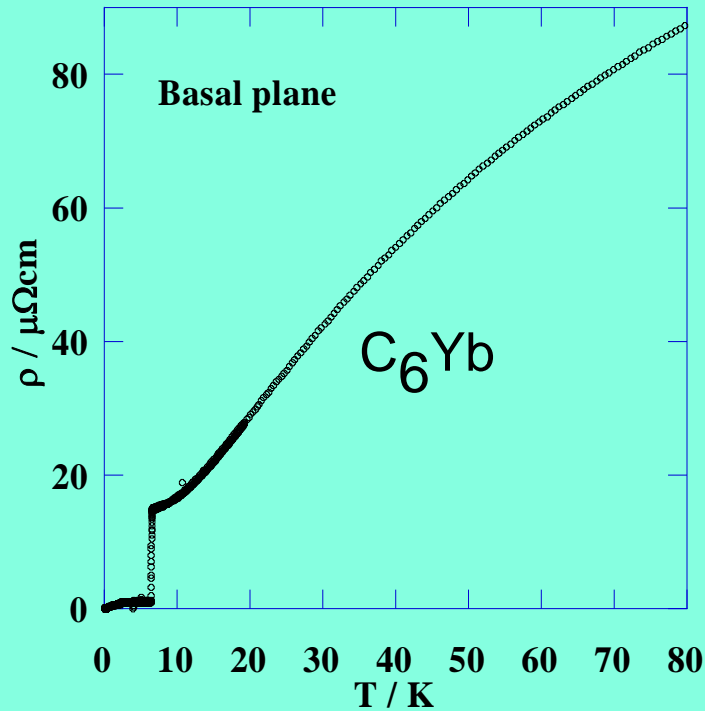
Sarrao et al. (2001), Hegger et al. (2000), ...

Graphite Intercalates

- Two dimensional hexagonal sheets of Carbon are held together by van der Waals forces.
- Can introduce metal atoms in between the sheets.
- This process can produce superconductivity. For example C_8K superconducts at 0.15K.
- Reports on superconductivity in Graphite Sulphur Composites and Nanotubes



Superconductivity in C_6Yb and C_6Ca

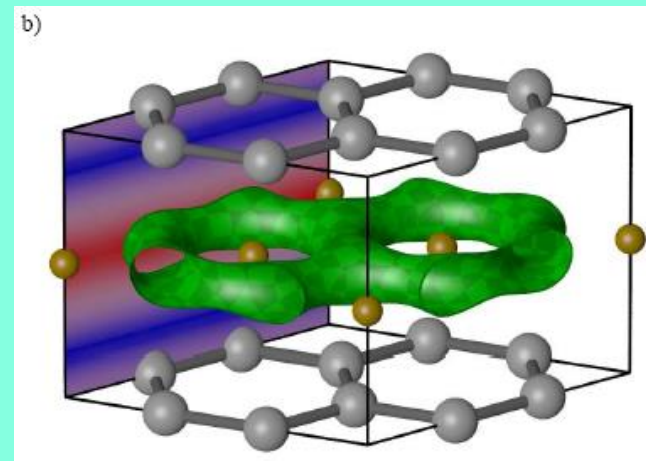
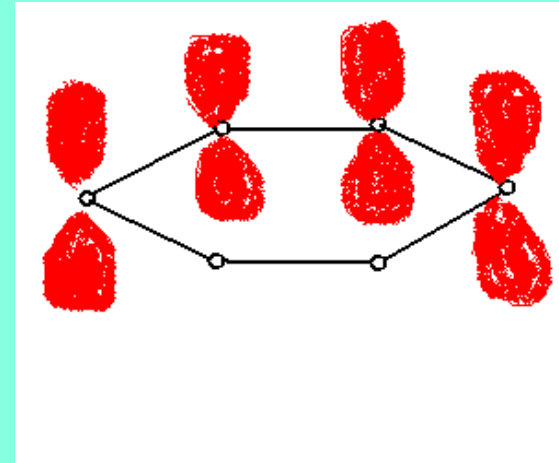
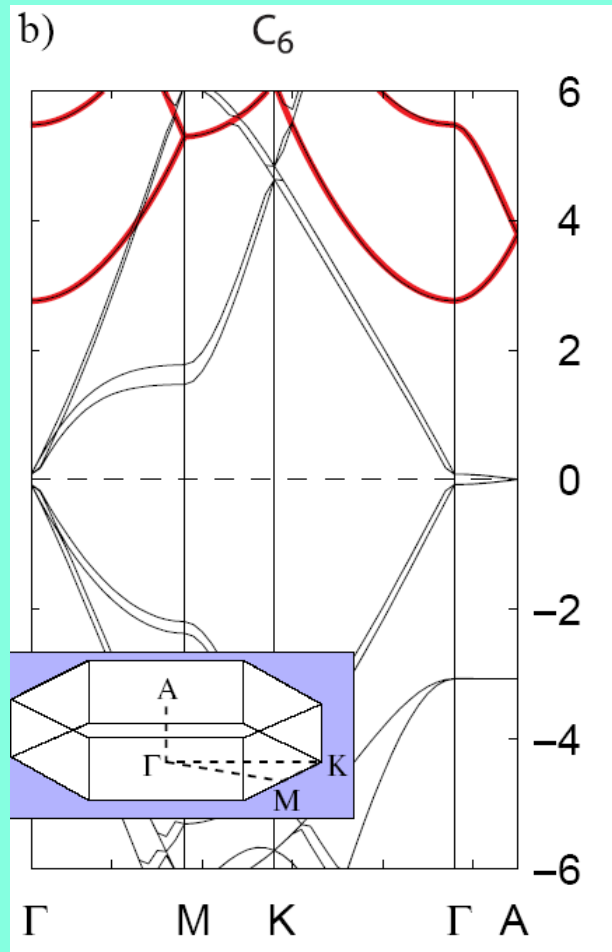


Weller, T., Ellerby, M., Saxena, S. S., Smith, R. & Skipper, N. *Nature Phys.* 1, 39–41 (2005)

Why is this interesting?

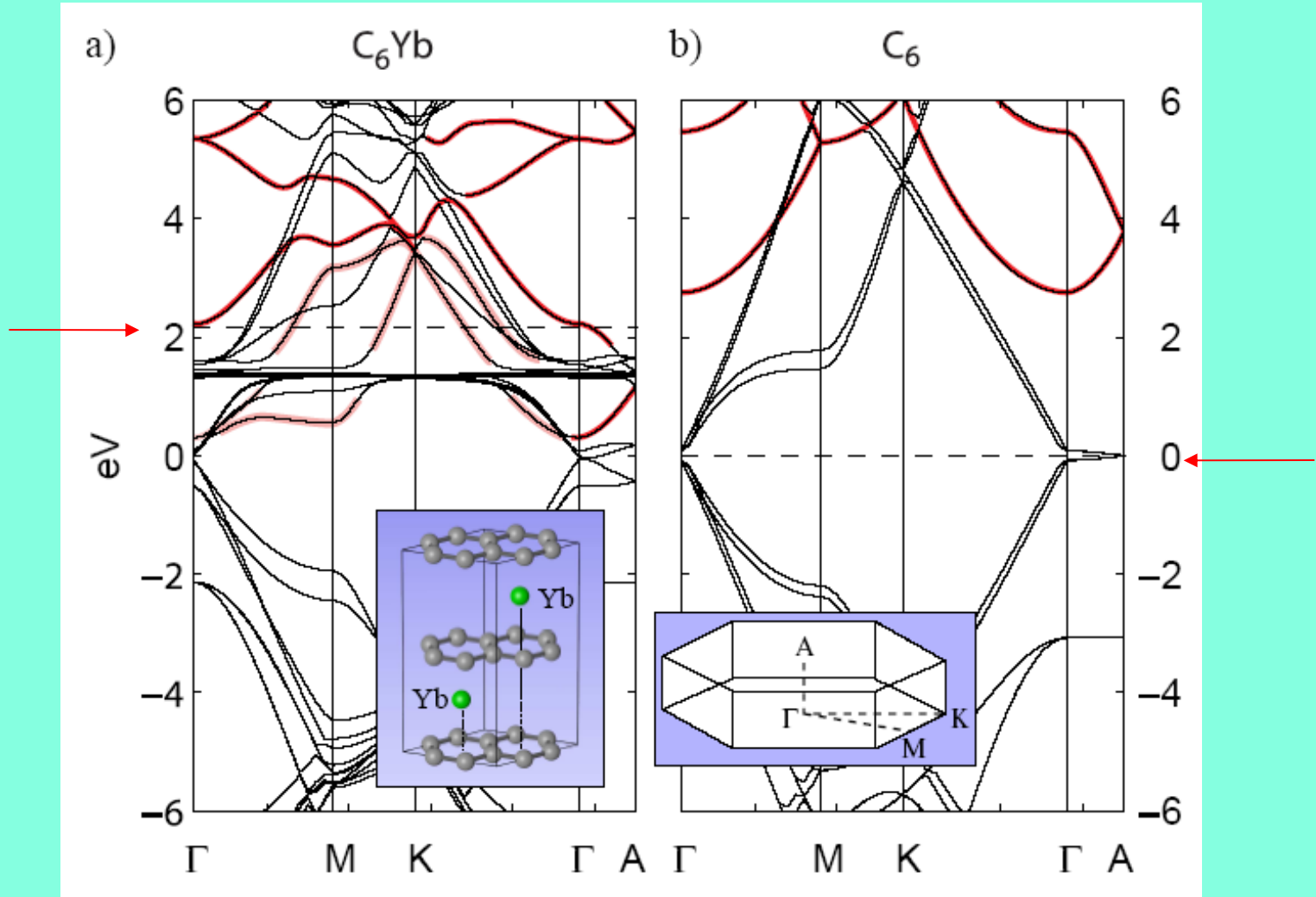
Compound	Electron doping	T_c
C_8K	1/8	0.15K
C_6Li	1/6	-
C_6Ca	1/3	11.5K
C_6Yb	1/3	6.5K
C_3Li	1/3	-
C_2Li	1/2	1.9K

Graphite band structure

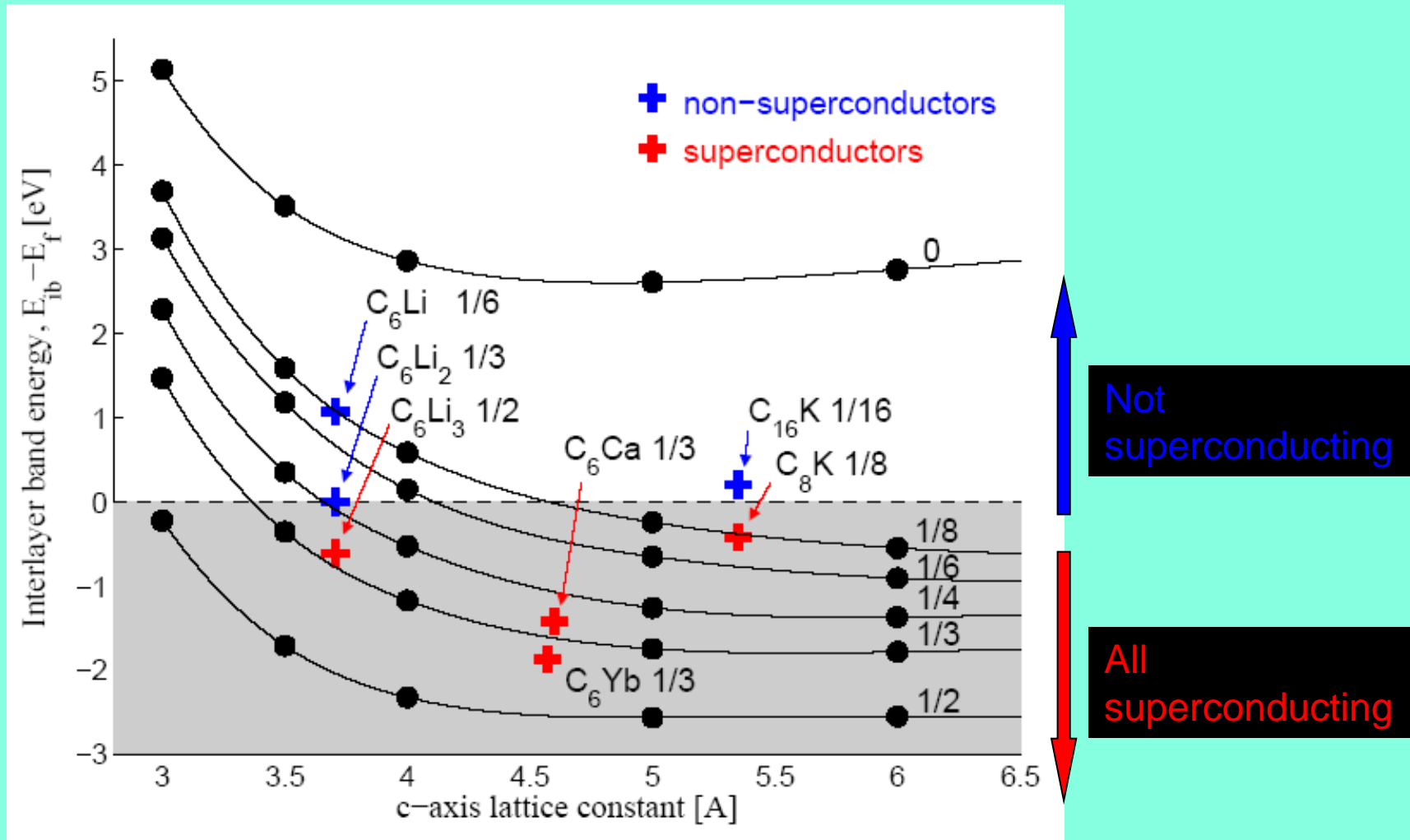


Band Structure – interlayer band

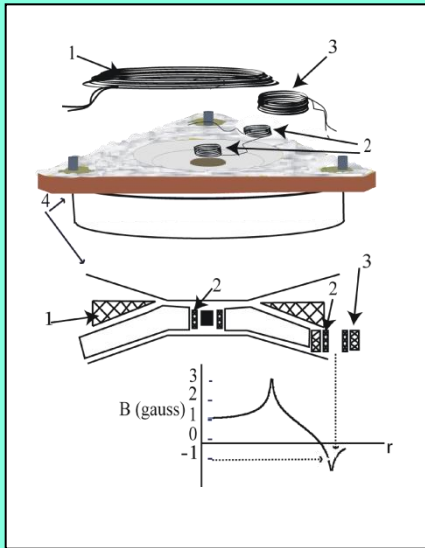
Csányi, G., Littlewood, P. B., Nevidomsky, A. H., Pickard, C. J. & Simons, B. D. Nature Phys. 1, 42–45 (2005).



Position of interlayer band

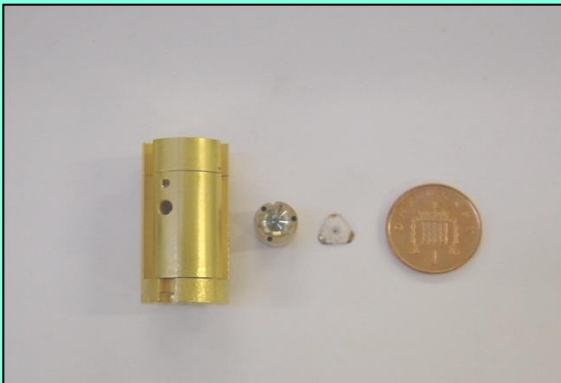


Experimental Techniques

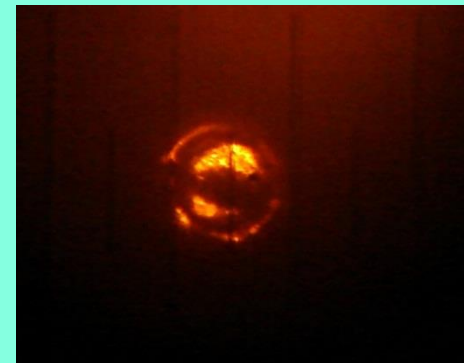
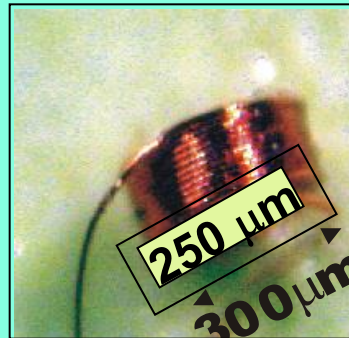


Susceptibility measurements in an anvil cell

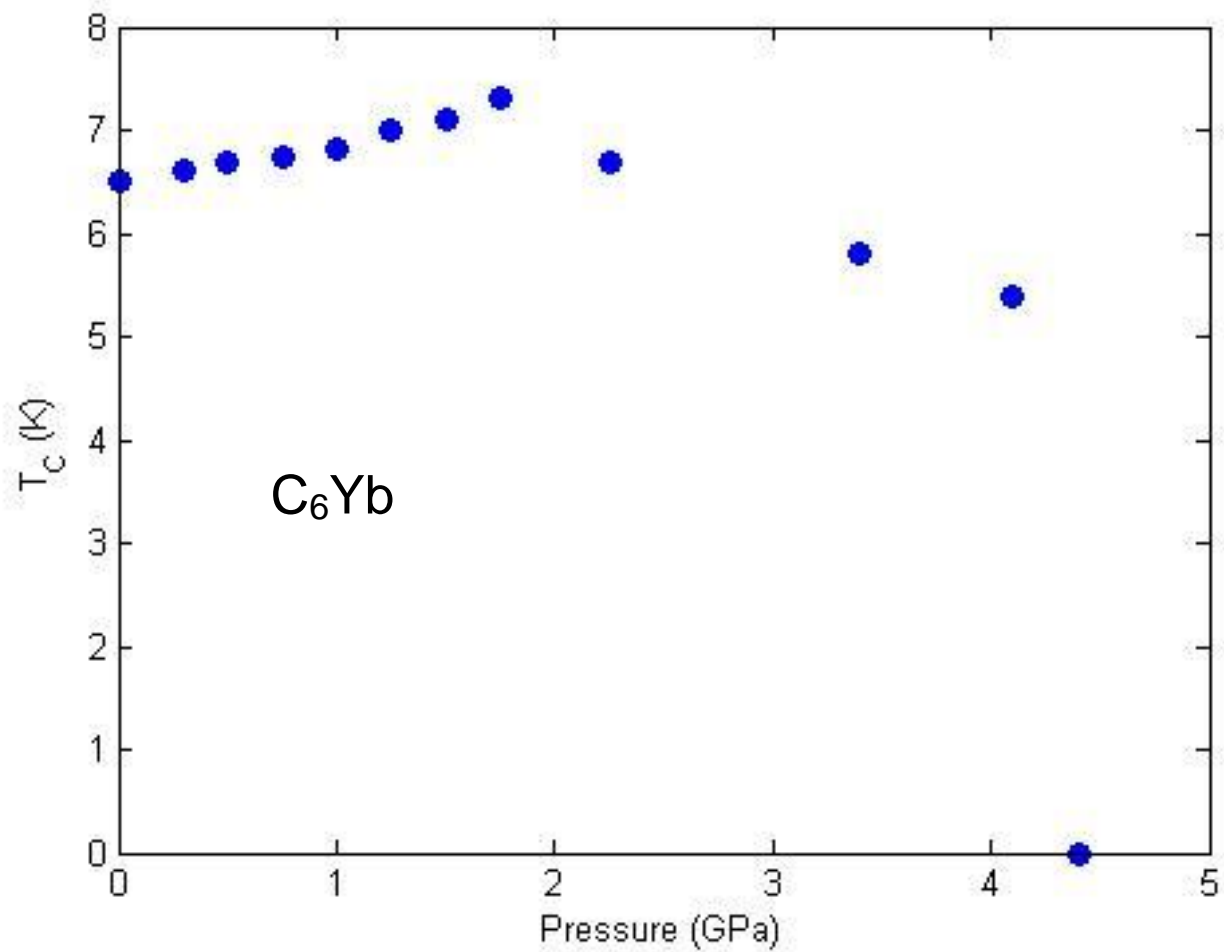
- Low Temperature (down to $T \sim 30$ mK)
- * High magnetic fields (up to 18 T)
- * Pressures up to ~ 120 Kbar

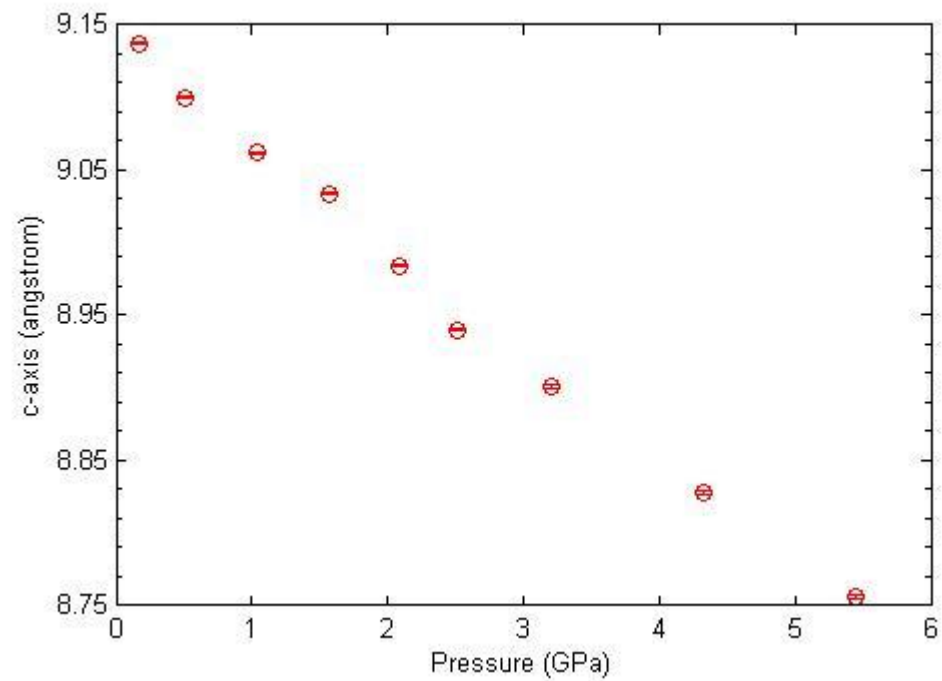
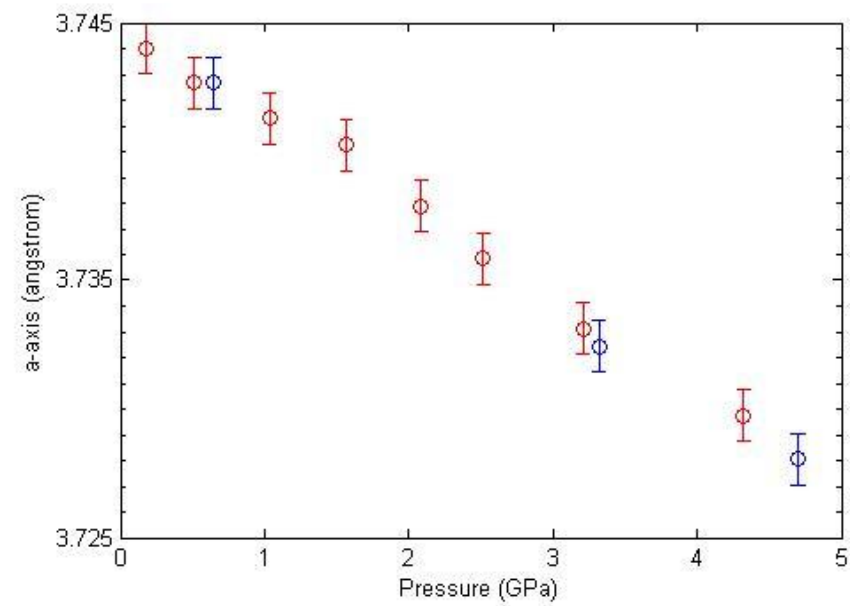


Miniature anvil cell

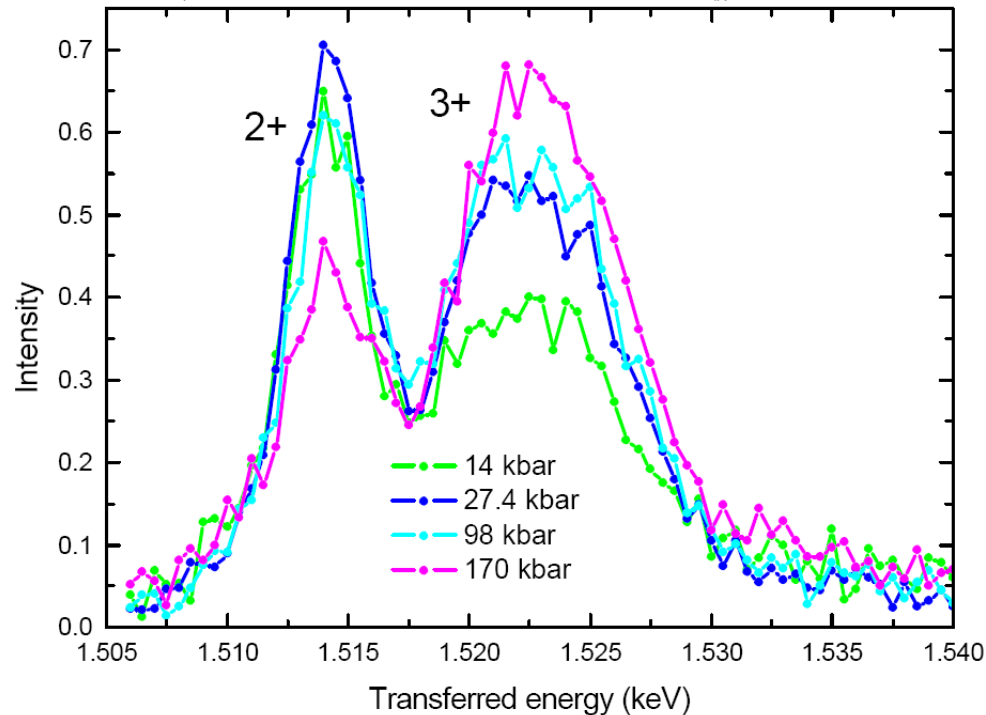


A good filling factor allows
Measurements of small samples
With small magnetic moments



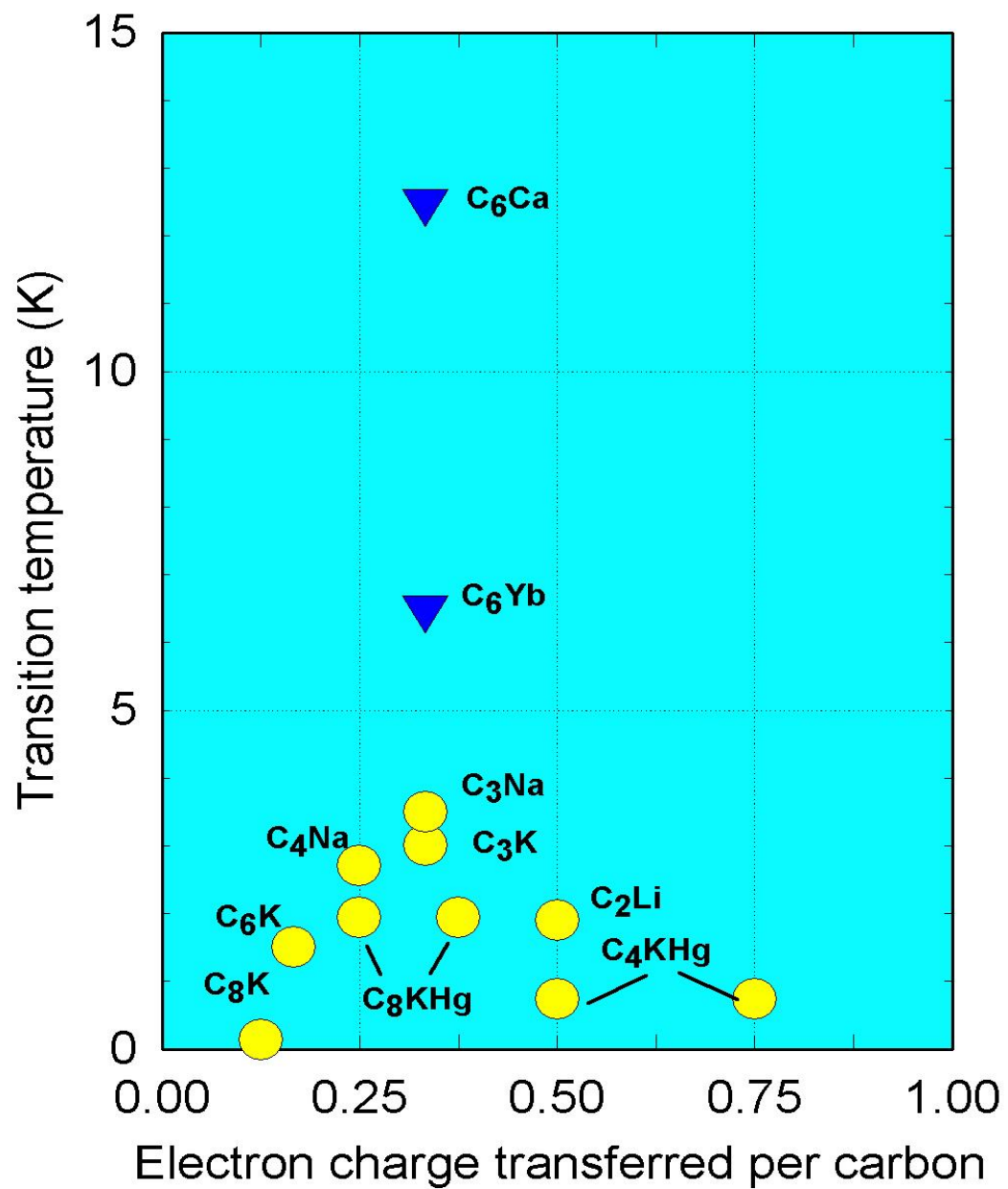


C_6Yb Sample 3, in pressure cell loaded in Argon glovebox,
 $L\alpha_1$ Resonant Inelastic Scattering excited at $h\nu_{IN} = 8.9325$ keV

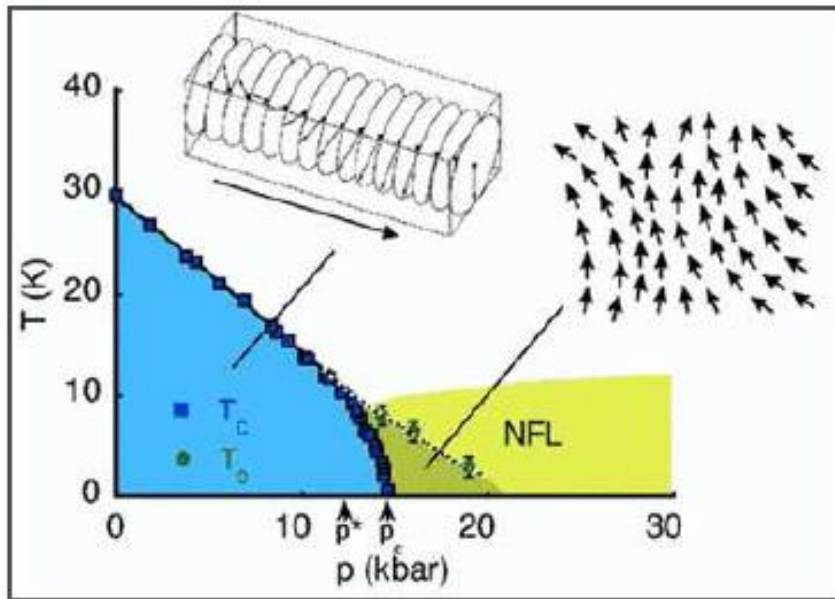


ESRF beamline ID26, December 2005

Transition in C_6Yb may be attributable to a shift in the charge state as observed in other Yb compounds. Tentative Resonant X-ray scattering by Claudia Dalleria and Matteo d'Astuto reveals a shift in the charge state between 1.4 and 2.7 GPa from 2+ to 3+.

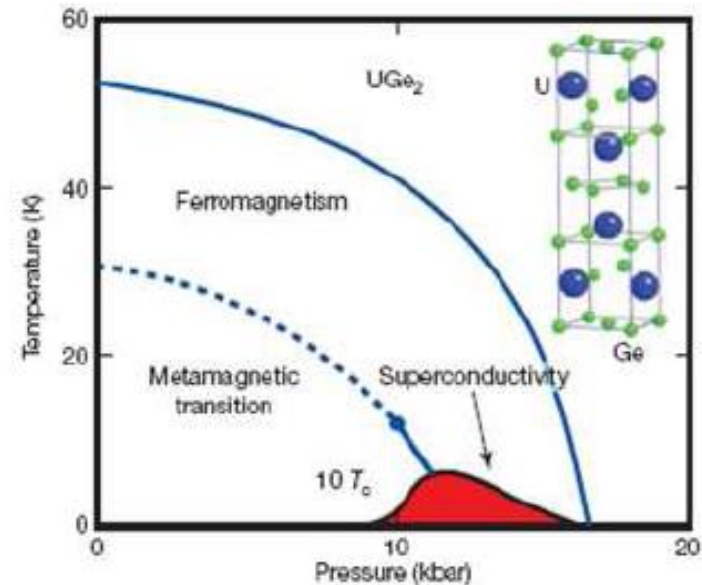


Neighbourhood of quantum phase transitions in metals – emergent states



d-electron metal

Spin skyrmion state in MnSi on border of ferromagnetism

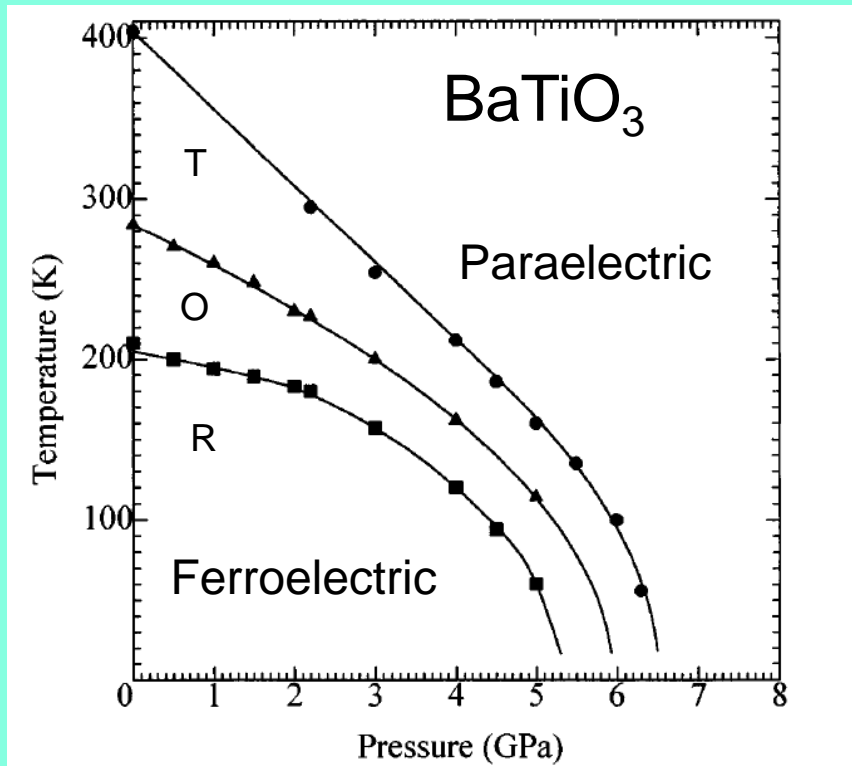


f-electron metal

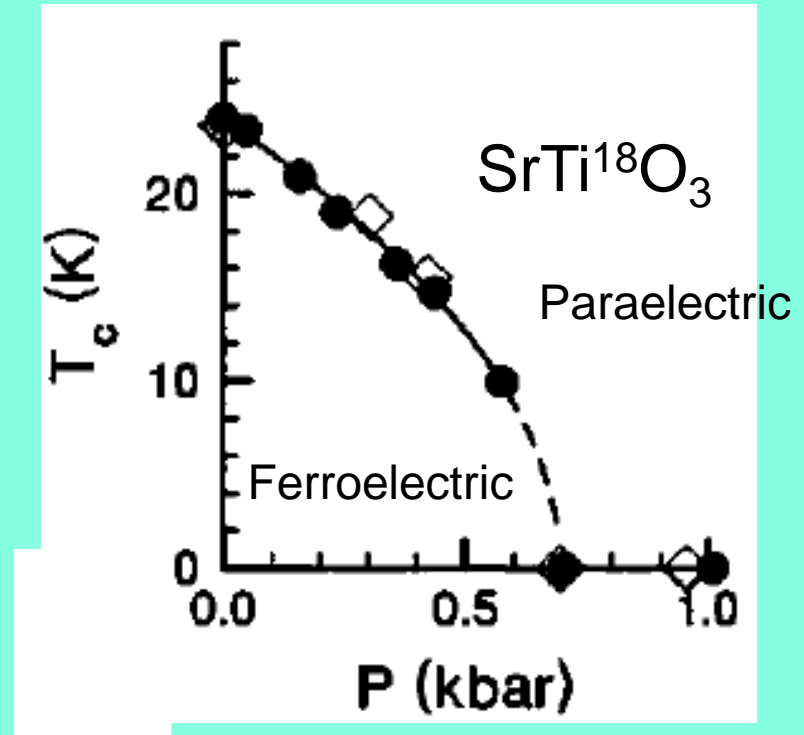
p-wave superconductor in heavy fermion UGe₂ on border of ferromagnetism. Also superconducting Sr₂RuO₄

Examples of Quantum Phase Transitions in Ferroelectric Materials

Displacive Type (cf. Itinerant magnetism)



T. Ishidate and S. Abe, H. Takahashi and N. Môri (1997)



Venturi et al. (2004)

What's different / interesting about the Ferroelectric QCP?

- Quantum criticality from the lattice
- Quantum critical fluctuations observed up to 50K!
- Simplest cases provide a Text-book paradigm of QC in the solid-state
- Emergence of new low temperature phases near QCP?

Review of metallic itinerant ferromagnets

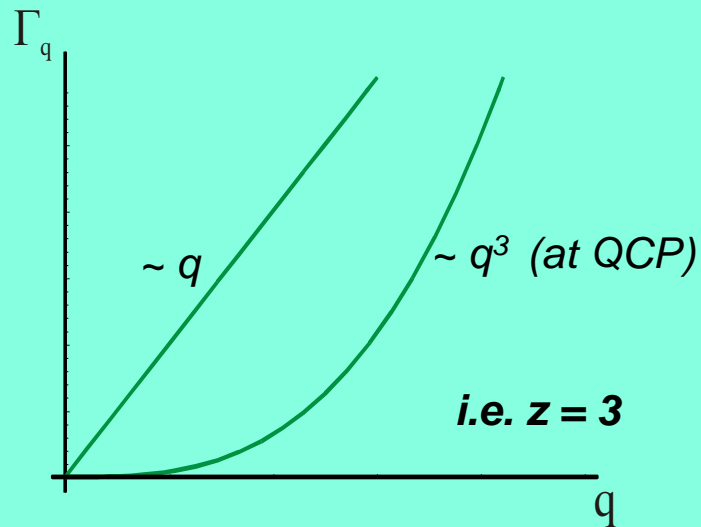
3D Itinerant Ferromagnet

- Dissipative Modes (Paramagnons)

$$\chi_{\mathbf{q}\omega}^{-1} = \chi_{\mathbf{q}}^{-1} \left(1 - i \frac{\omega}{\Gamma_{\mathbf{q}}} \right)$$

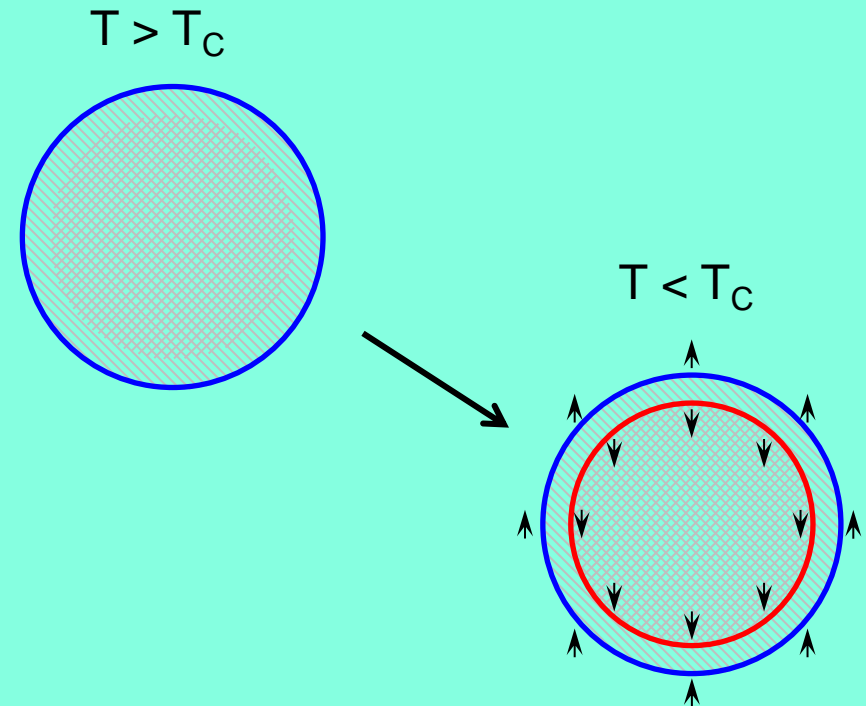
- Relaxation Rate

$$\Gamma_{\mathbf{q}} = \gamma q (\chi^{-1} + cq^2)$$



$$F = \frac{a}{2} M^2 + \frac{b}{4} M^4 + c(\nabla M)^2$$

$$M = M(r, t)$$



Mean-Field Predictions for Ferroelectric Charge Fluctuations

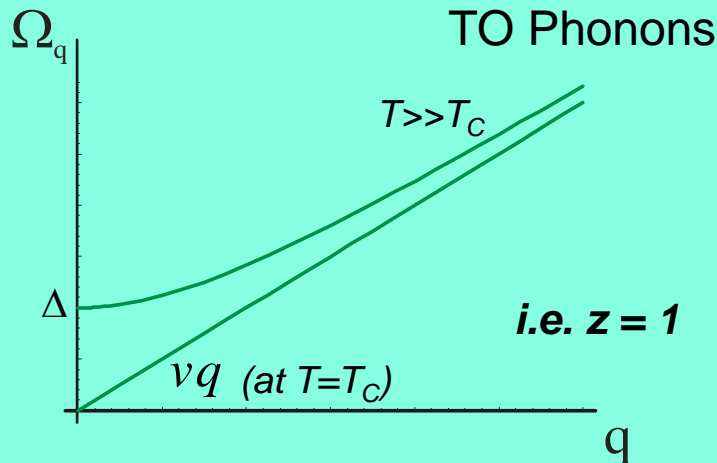
3D Cubic Ferroelectric

- Propagating Modes (Soft-Optic Phonons)

$$\chi_{q\omega}^{-1} = \chi_q^{-1} \left(1 - \frac{\omega^2}{\Omega_q^2} \right)$$

- Dispersion

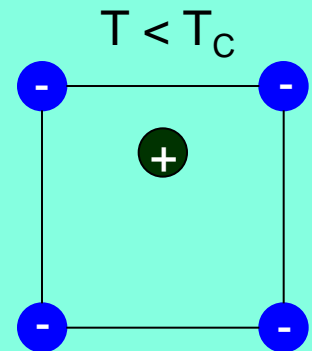
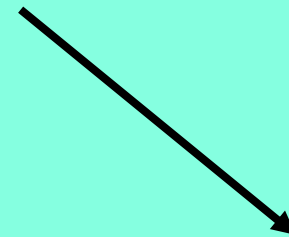
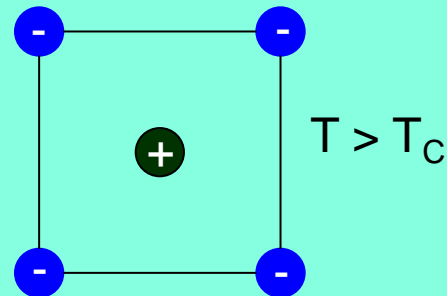
$$\Omega_q = \sqrt{\Delta^2 + v^2 q^2}$$



$$F = \frac{a}{2} P^2 + \frac{b}{4} P^4 + c(\nabla P)^2$$

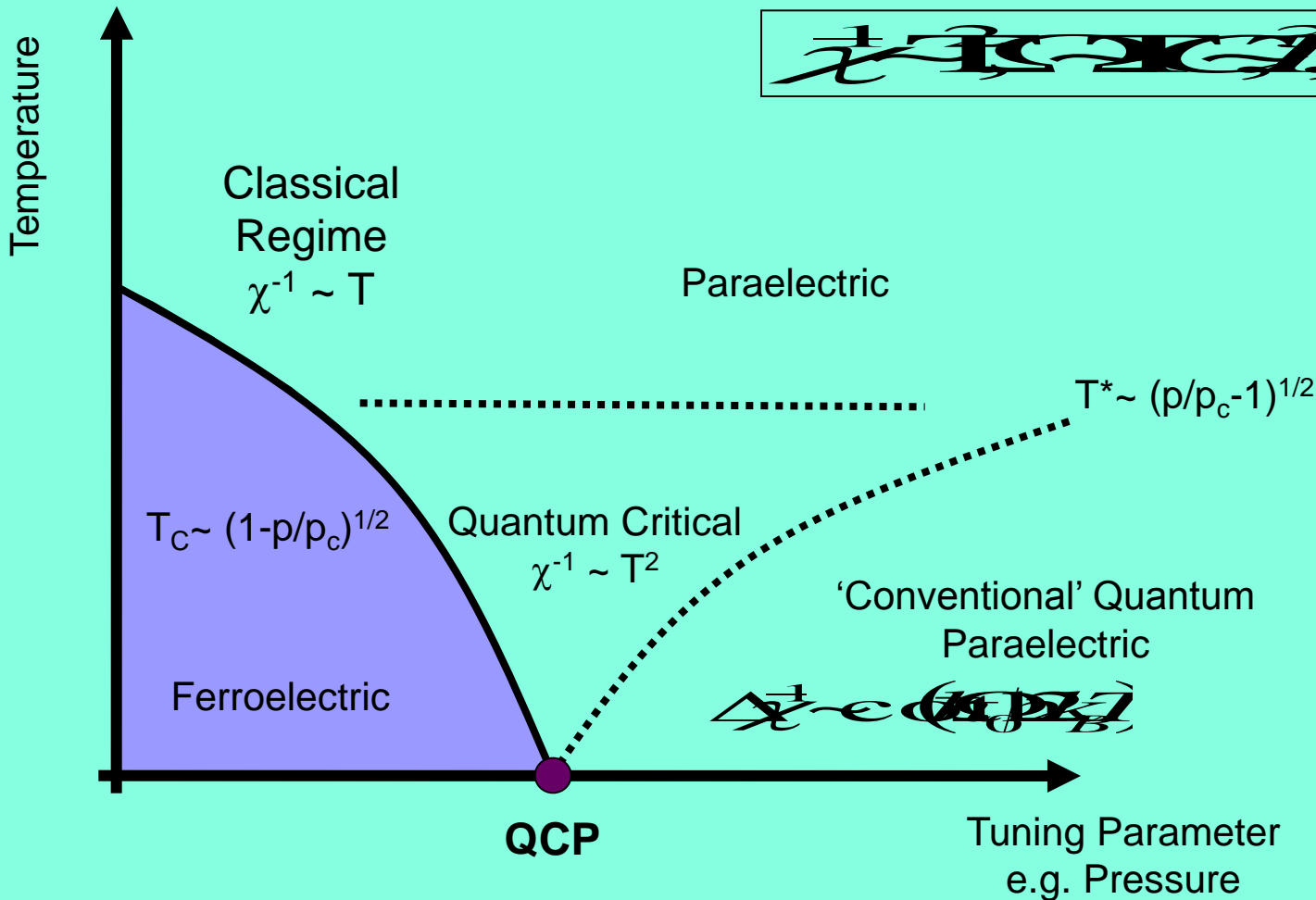
+ dipole _ interactions

$$P = P(r, t)$$



Predicted Phase Diagram for Low Temperature Ferroelectrics

Quantum Fluctuations of charge polarization in four dimensions.
 Simplest ferroelectrics - Text-Book paradigm of QCP in the solid-state



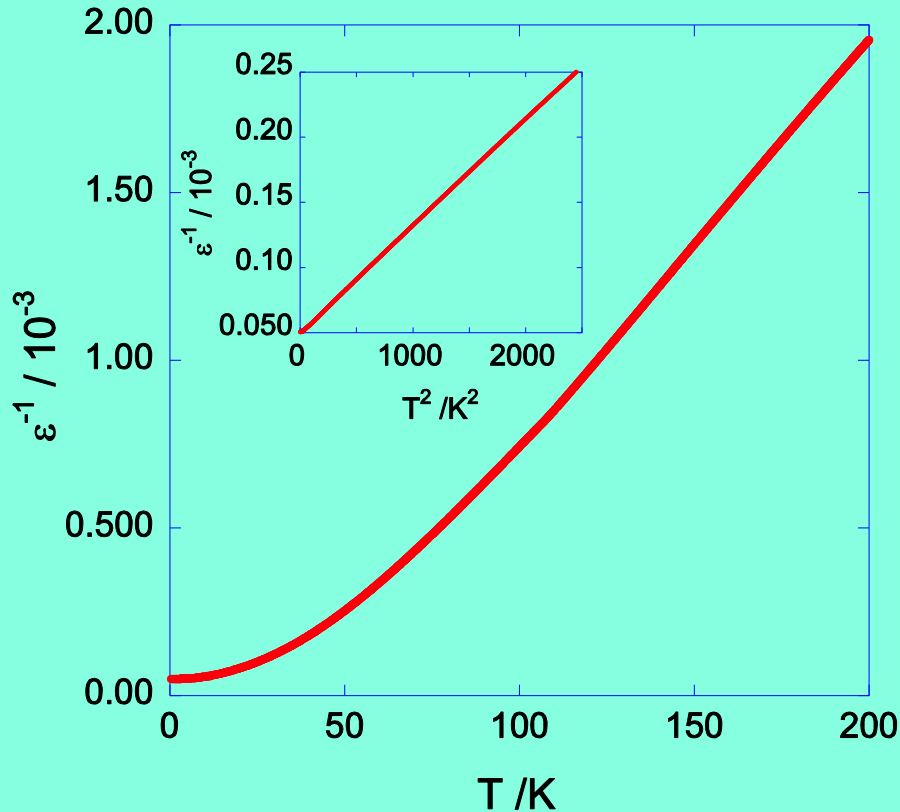
Measurements

- 45mK adiabatic demagnetisation fridge fitted with miniature coaxial cables connected to high precision capacitance bridge
- Sensitivity 10^{-18}F , frequencies up to a few MHz
- Dielectric constant, polarization, pyroelectric current
- Large electric fields $\sim 50 \text{ kVcm}^{-1}$
- 30 kbar pressure cell available with coaxial cables into high pressure region (accuracy 5×10^{-2} kbar)

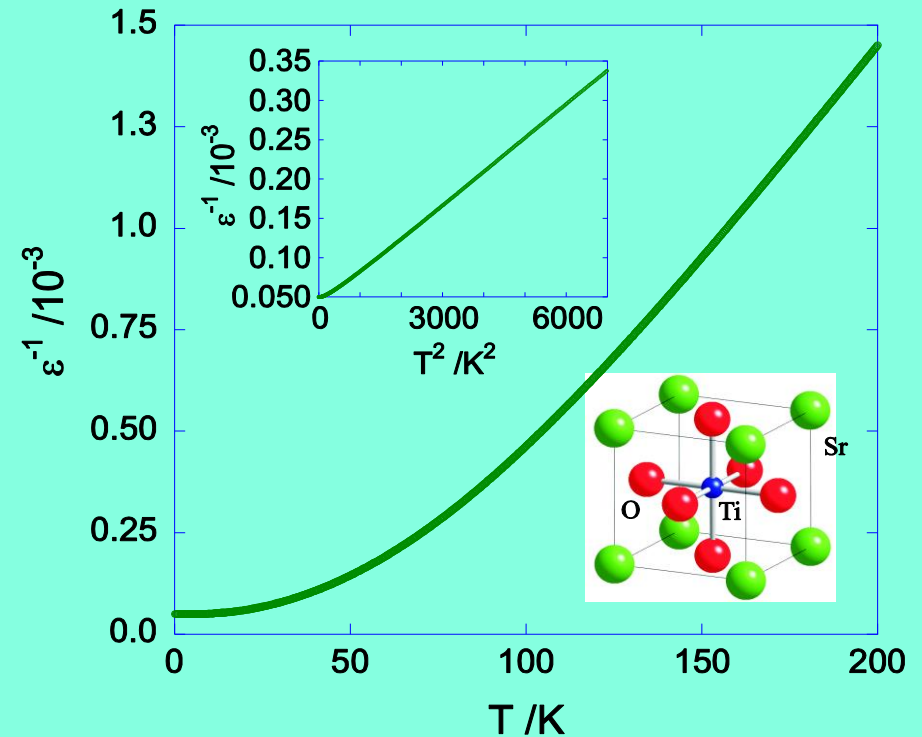


SrTiO₃

Experiment



Self-consistent phonon model



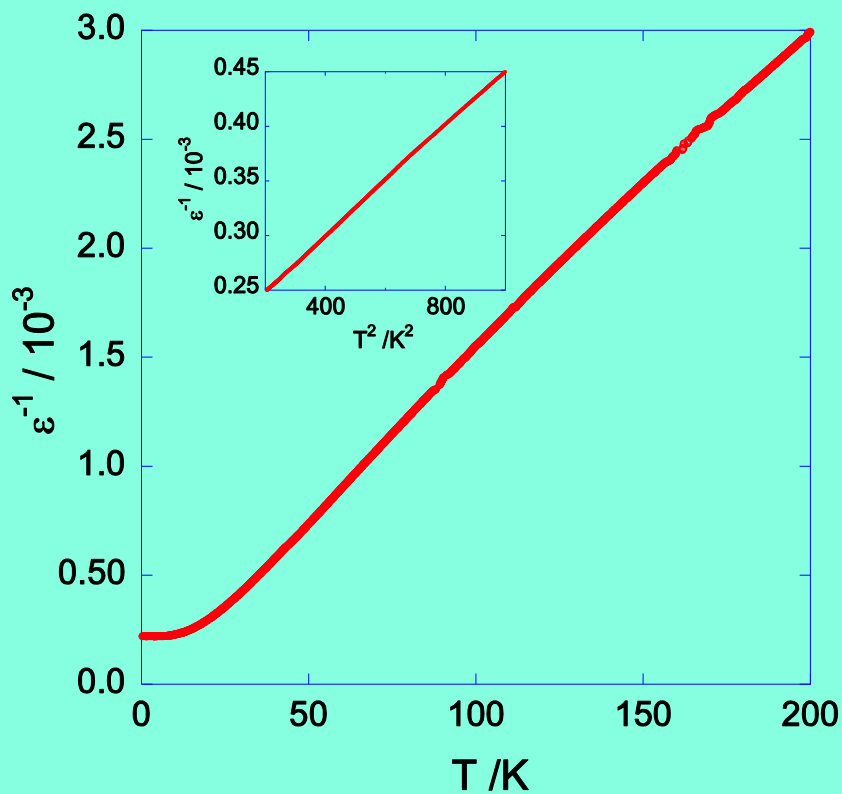
Close qualitative AND quantitative agreement with simple model for quantum critical point without freely adjustable parameters.

Closed form equation for low temperature limit near QCP:

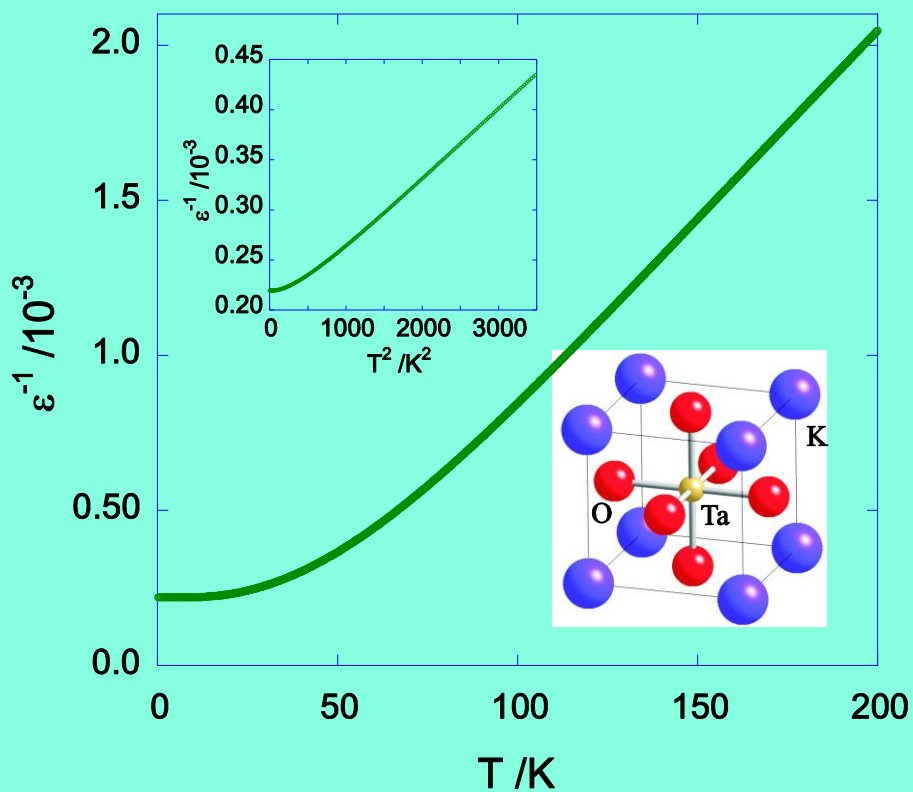
$$\chi^{-1} = a + (10/3) (\epsilon_0 k_B^2 b \Delta^2 / 12 \hbar a v^2) T^2$$

KTaO₃

Experiment



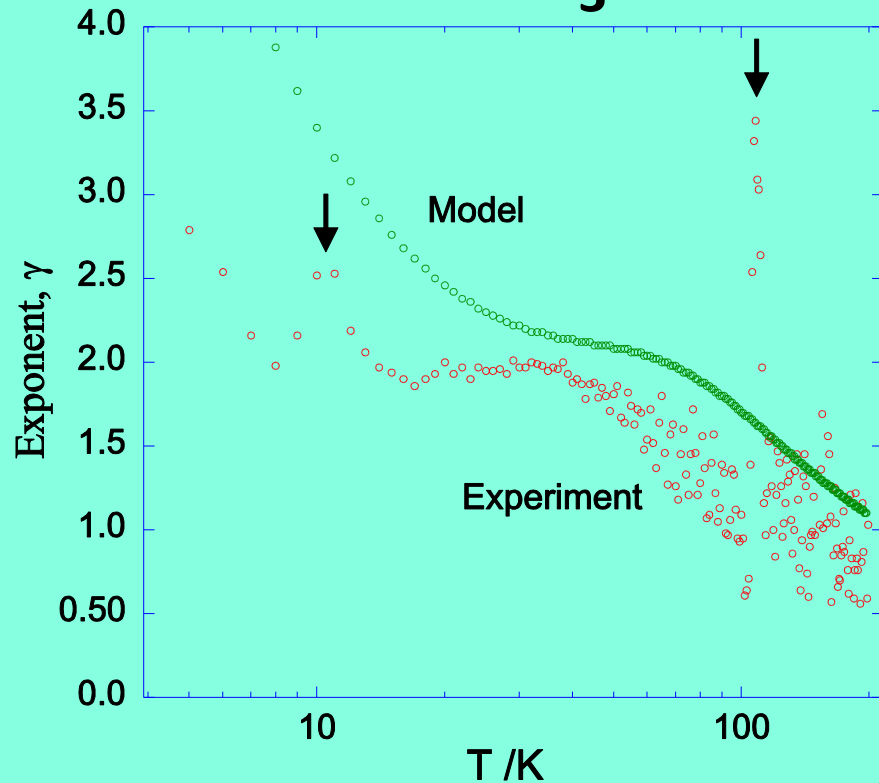
Self-consistent phonon model



Close quantitative agreement with simple model for quantum critical point without any freely adjustable parameters

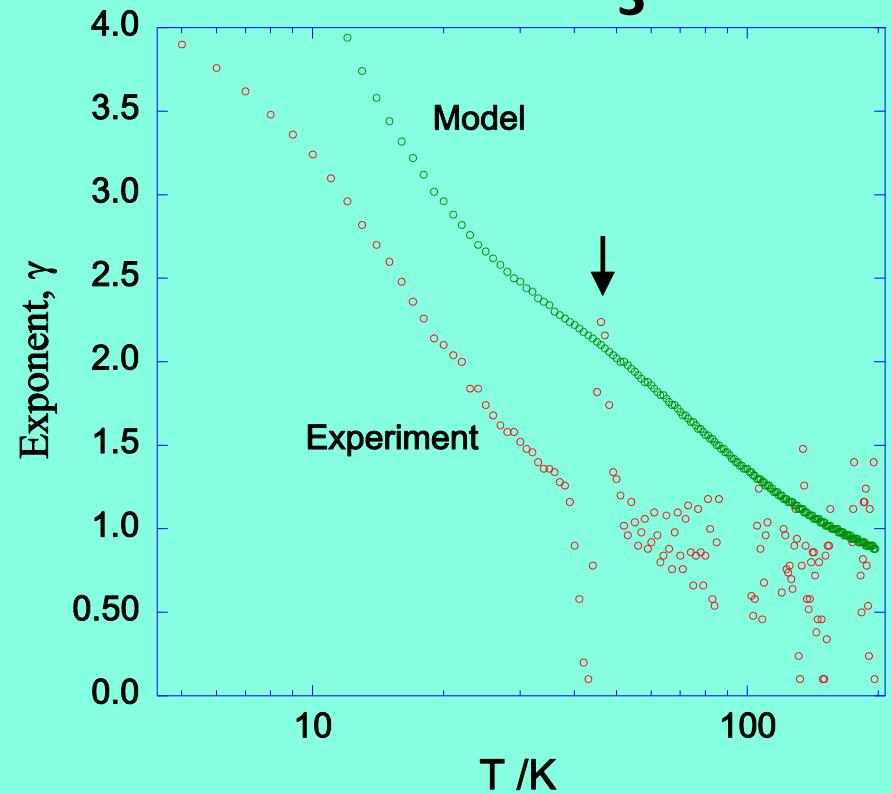
Unconventional Power Law

SrTiO₃



T^2 power law observed in quantitative agreement with model without freely adjustable parameters
Quantum Critical Fluctuations up to ~ 50 K

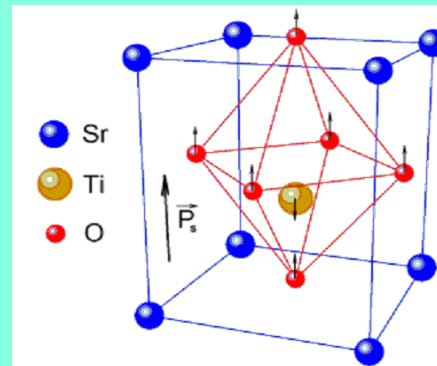
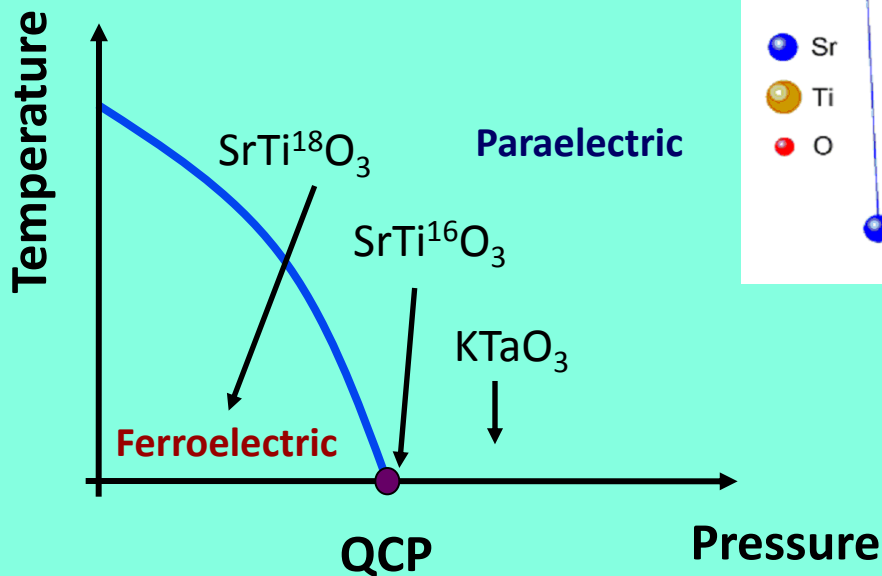
KTaO₃



No extended region of T^2 behaviour since further from QCP. Higher power indicative of exponential dependence

SrTiO₃ and KTaO₃

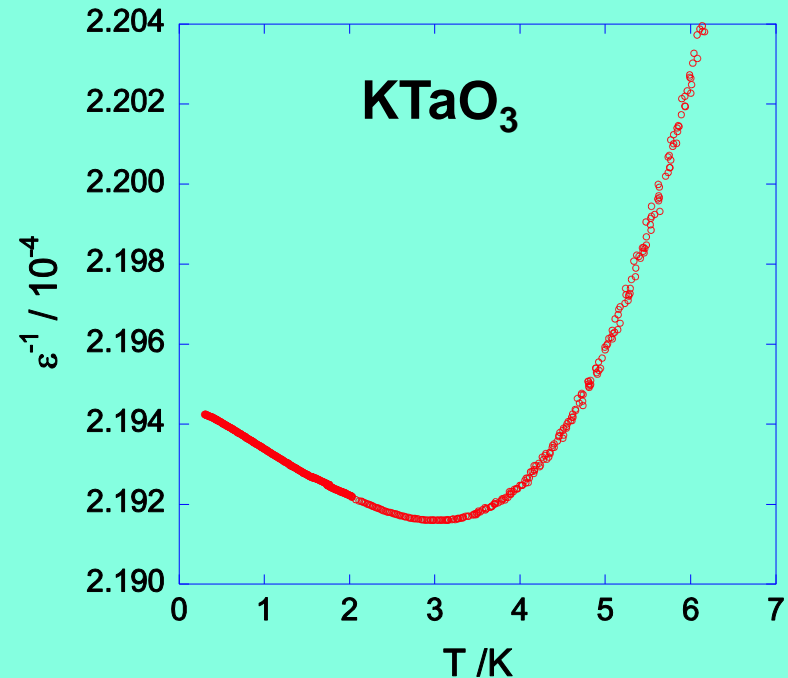
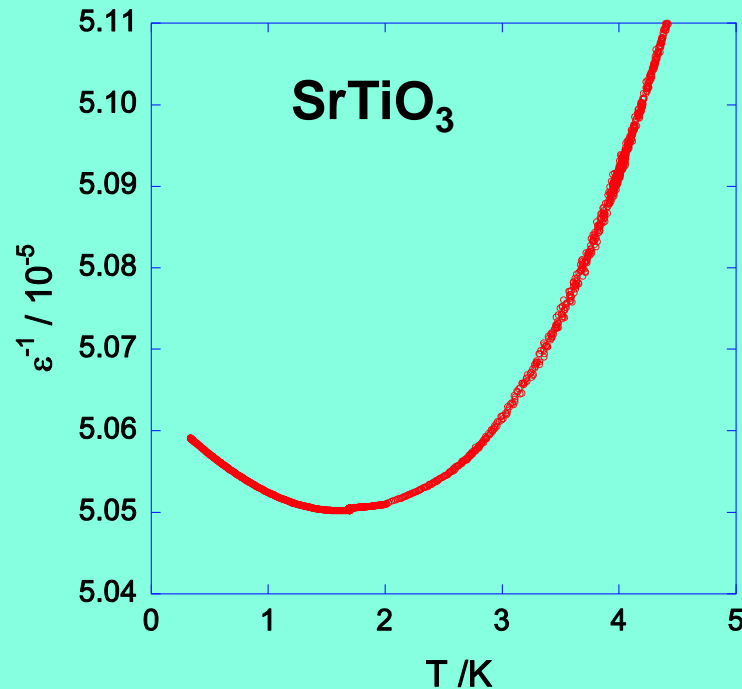
- Simple cubic Perovskite structures at room temperature
- Transition metal oxide insulators
- Clean and well studied materials close to displacive instability
- Naturally exist close to quantum critical point (QCP) without the need for pressure tuning



Strontium Titanate crystal pairs

Discovery of New Low Temperature Regime in SrTiO₃ and KTaO₃

Connected with QCP



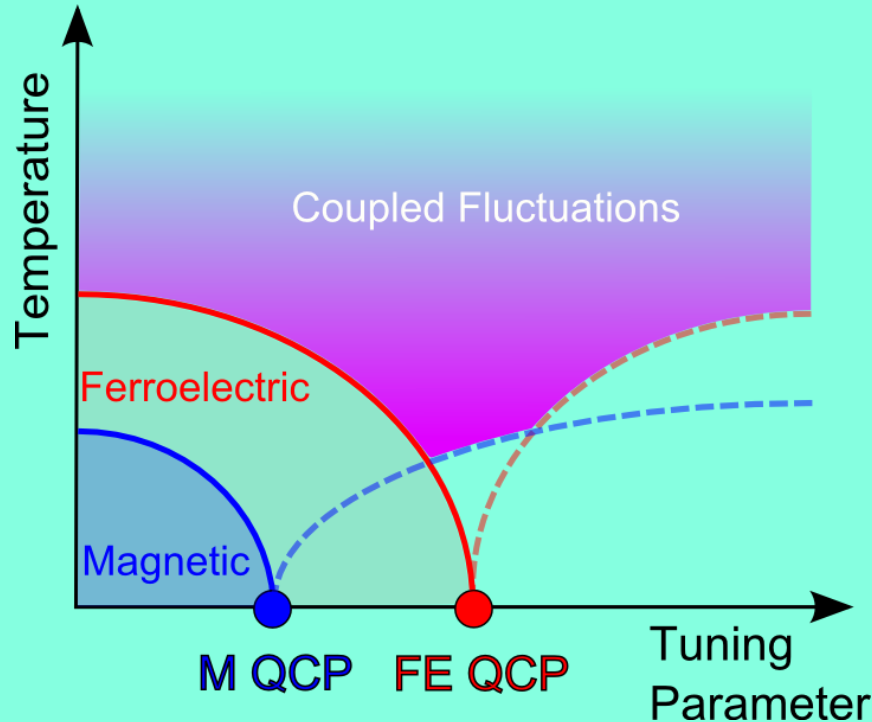
- Possible Explanation involves **coupling to acoustic phonons near QCP** when $T \ll \Delta$

$$\Delta F = (\nabla \phi) P^3$$

- Predicted by D.E. Khmel'nitskii and V.L. Shneerson, Sov. Phys. – Solid State, 13, 687 (1971) !

- See also Palova, Chandra and P. Coleman (2008)

Outlook: Multiferroic Quantum Criticality



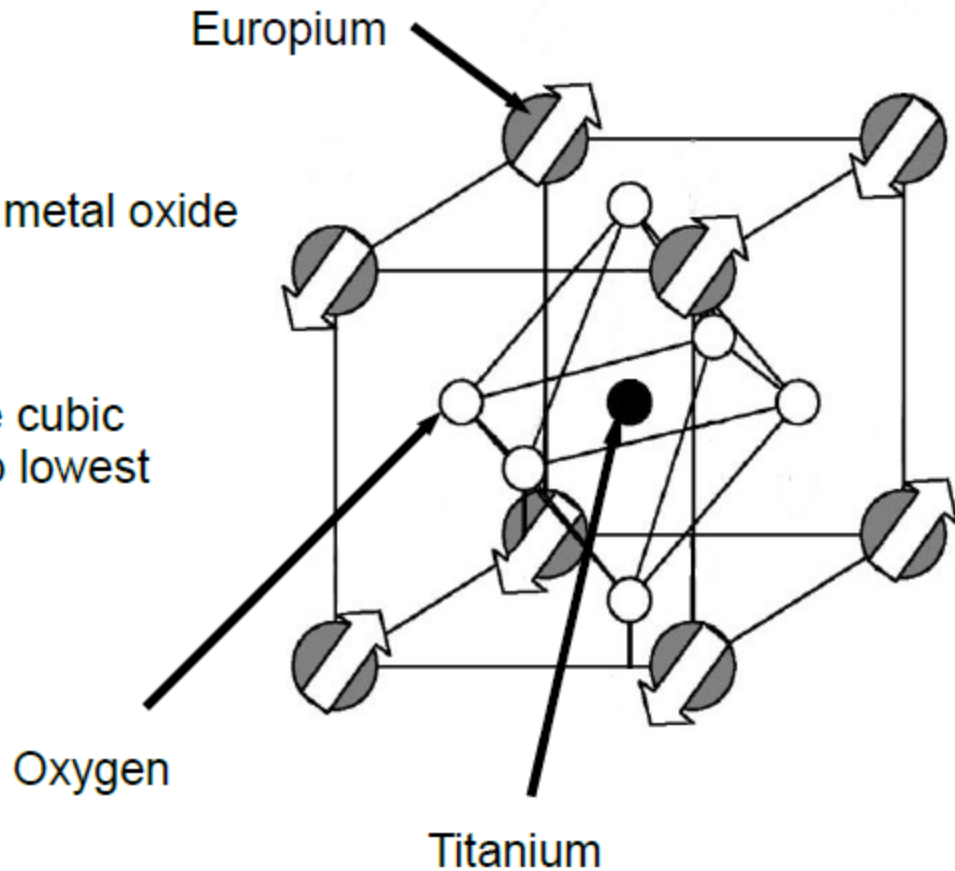
Multiferroics (eg. EuTiO_3) are ideal systems in which to study the Quantum Critical Fluctuations of coupled spin and charge order parameters by tuning the respective transitions to absolute zero.

**Leszek Spalek, S. Rowley, M. Shimuta, T. Katsufuji, C. Panagapoulos,
S. Saxena**

(A major collaboration with Heraklion)

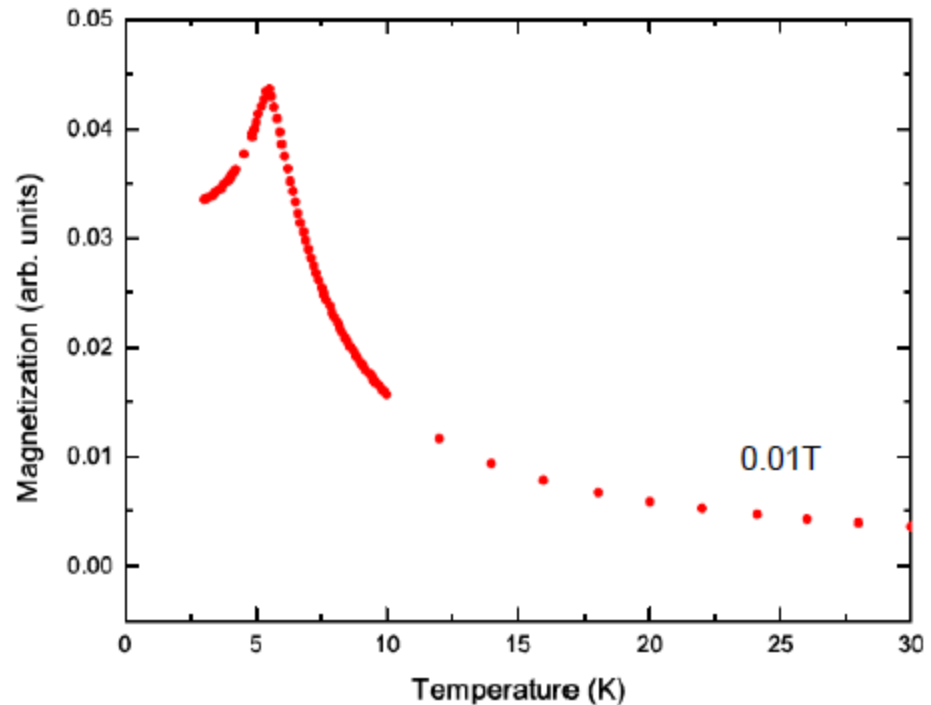
EuTiO_3 – the basics

- Rare-Earth Transition metal oxide
- Magnetic insulator
- Reported to be simple cubic perovskite structure to lowest temperatures

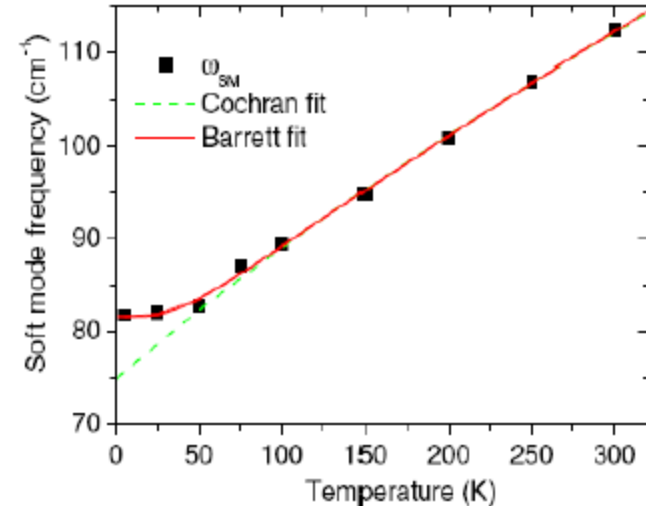
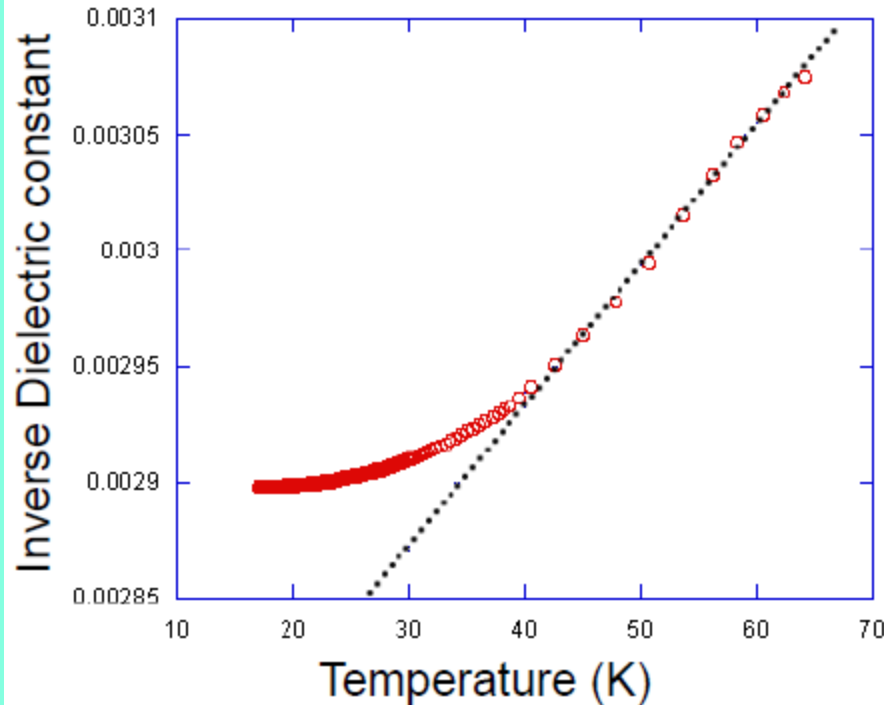


$\text{Eu}^{2+}\text{Ti}^{4+}\text{O}_3^{2-}$ - An Antiferromagnet

- Magnetic $\text{Eu}^{2+}(4f^7)$ atoms with $S=7/2$ ($7\mu_B$)
- Antiferromagnetic ordering below $T_N = 5.5\text{K}$
- Nearest neighbor coupling: $J_1 = -0.07\text{K}$



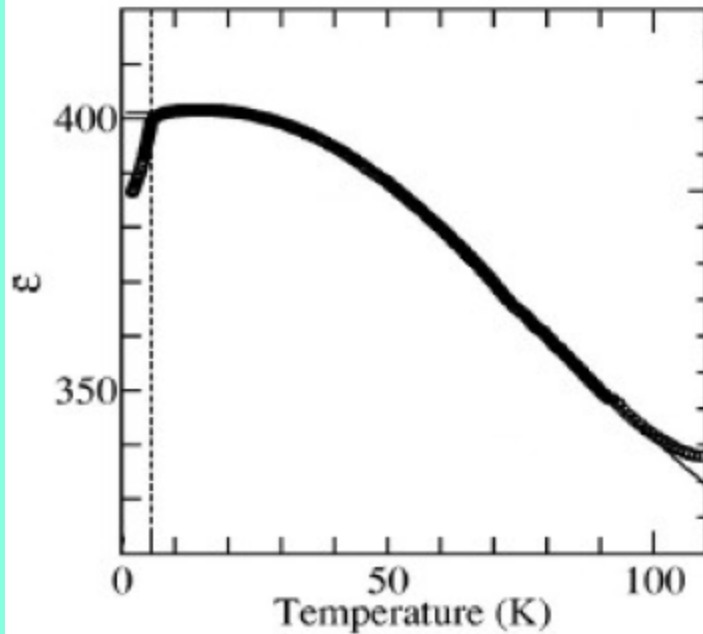
A Quantum Paraelectric



S. Kamba et al., Eur. Phys. Lett. (2007)

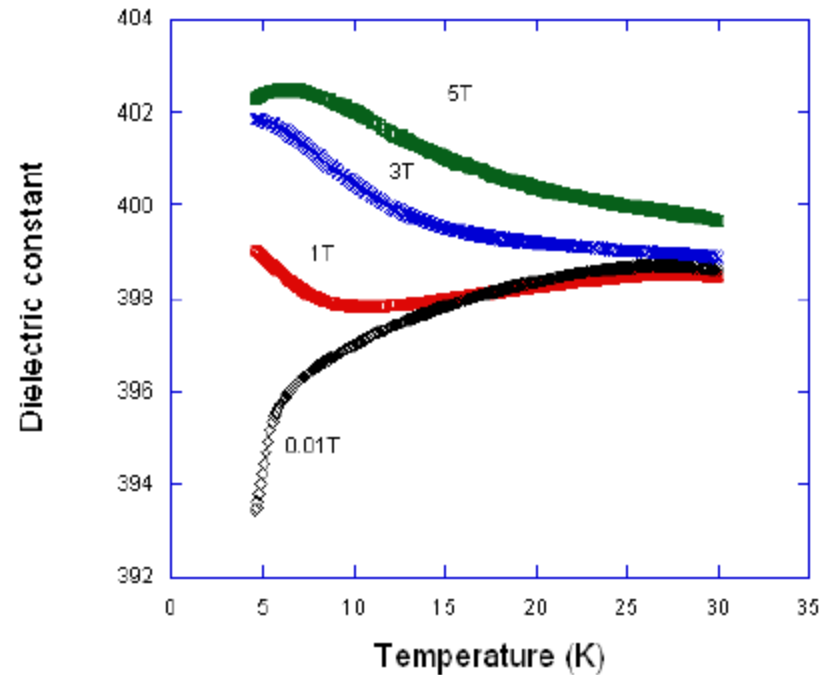
- Breakdown of classical behavior at low temperatures
- Expected transition avoided
- No ordering observed similar to SrTiO_3

A Magnetolectric



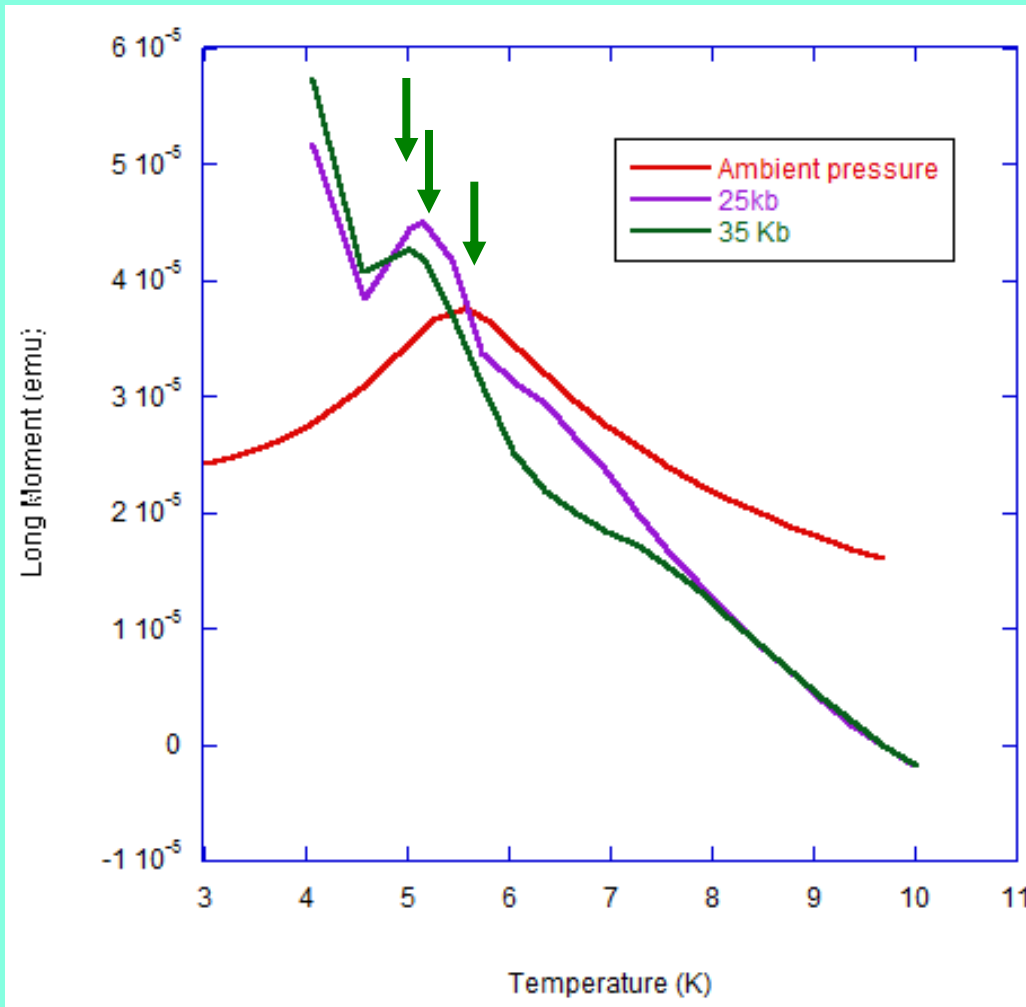
T.Katsufuji and H.Takagi, PRB (2001)

- Coupling observed between the electric and magnetic subsystems



- Dielectric constant strongly enhanced with magnetic field

EuTiO₃ Pressure Tuning



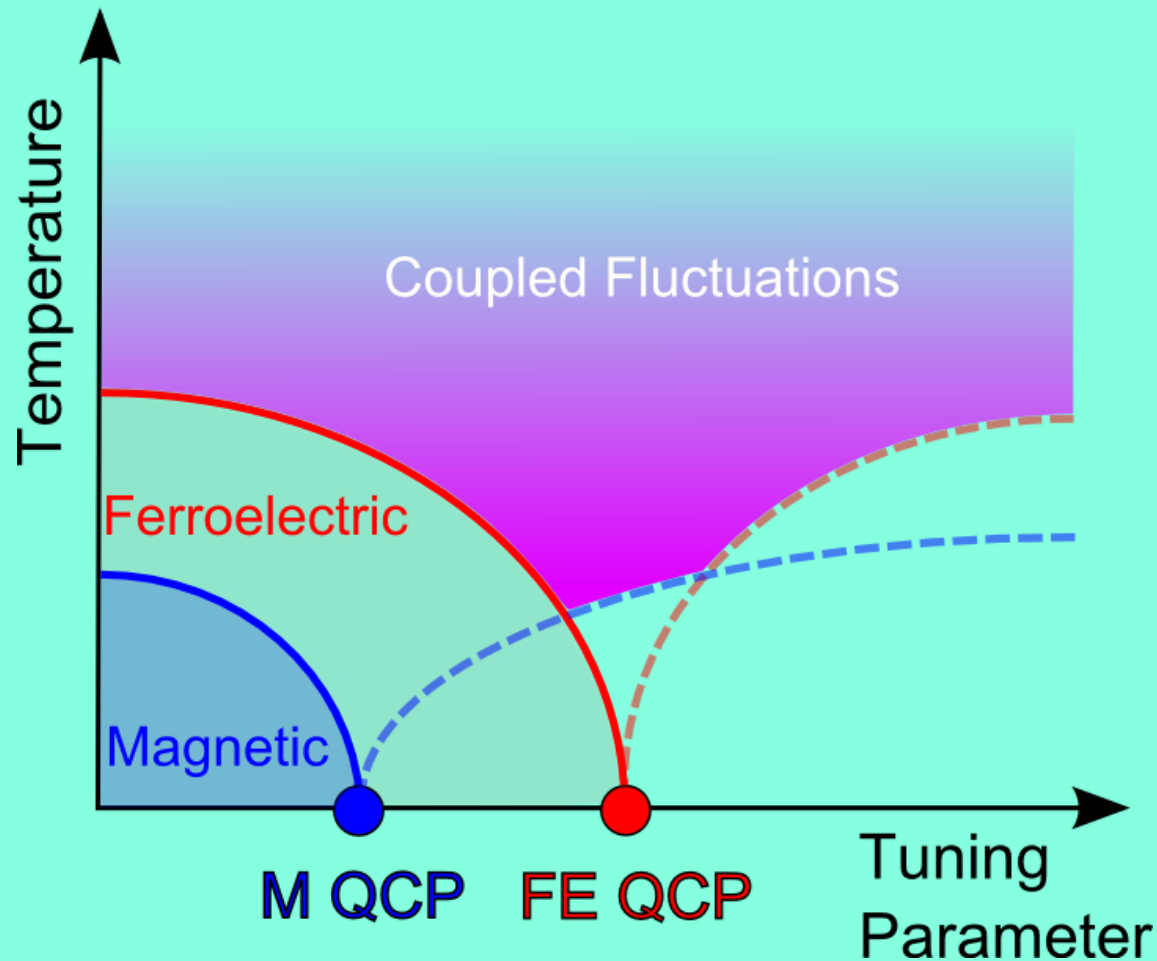
$$T_N(p=0)=5.58\text{K}$$

$$T_N(p=25\text{kbar})=5.14\text{K}$$

$$T_N(p=35\text{kbar})=5.0\text{K}$$

A gradual suppression of the Neel temperature

Outlook: Multiferroic Quantum Criticality

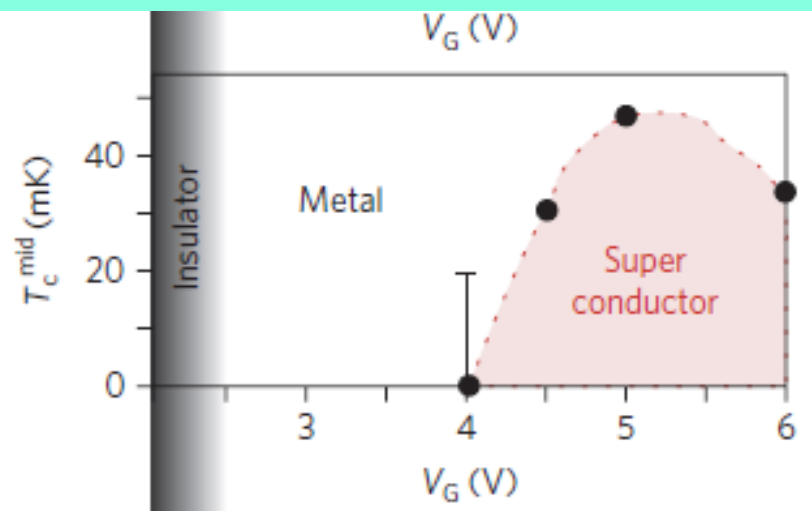
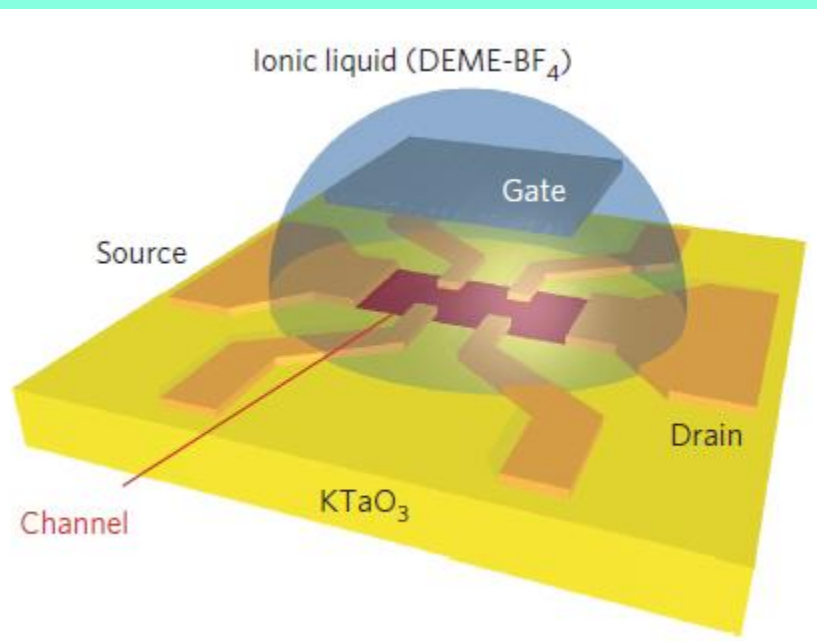


Reintroducing charge carriers - ferroelectric metals

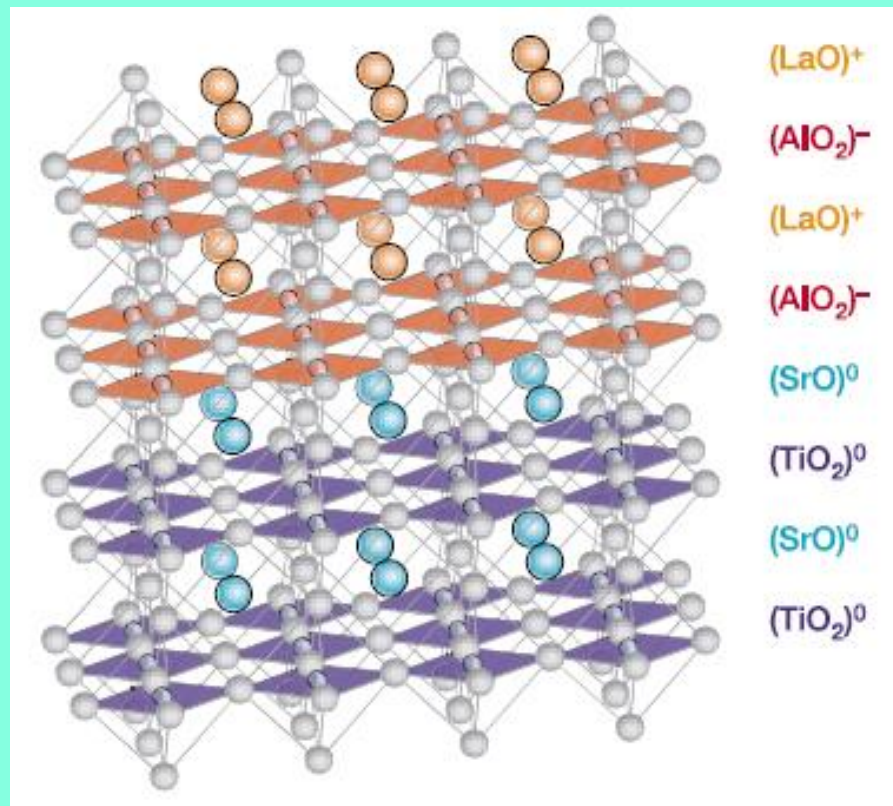
- Looking to the future we imagine that quantum ferroelectric fluctuations may mediate effective electron-electron interactions in crystals supporting mobile charge carriers.
- By adding carriers into incipient or weakly ferroelectric materials, superconductivity is commonly observed.
- For example **Nb doped or oxygen reduced SrTiO₃**
- Superconductivity in 2D layer at SrTiO₃ / LaAlO₃ **oxide interface**
- Superconductivity in SrTiO₃ when **exposed to UV light**
- **Ionic liquid gated SrTiO₃ and KTaO₃**

Others including the ferroelectric semiconductors -> GeTe, SnTe

Gated KTaO₃ and SrTiO₃



SrTiO₃ / LaAlO₃ oxide interface



Discovery of metastable
phase transition in
Pressurized Cs_2CuCl_4

Collaborators

Cavendish Laboratory, University of Cambridge:

Dr. H.J. Kim, D.Y. Kim

Seoul National University:

Saehwan Chun, Prof. Kee Hoon Kim

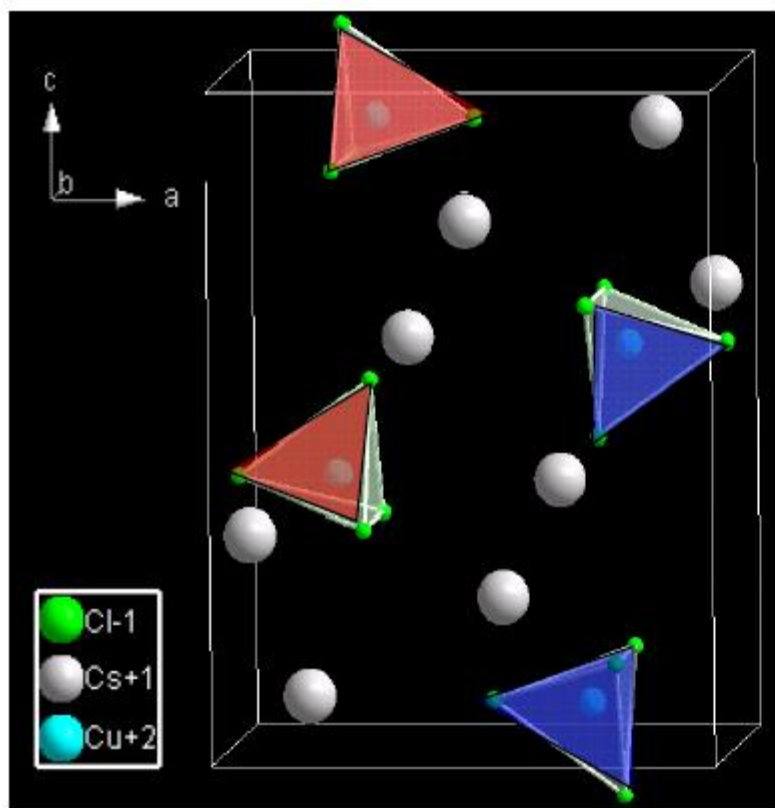
Rutgers University:

H. T. Yi, Prof. Sang-Wook Chung.

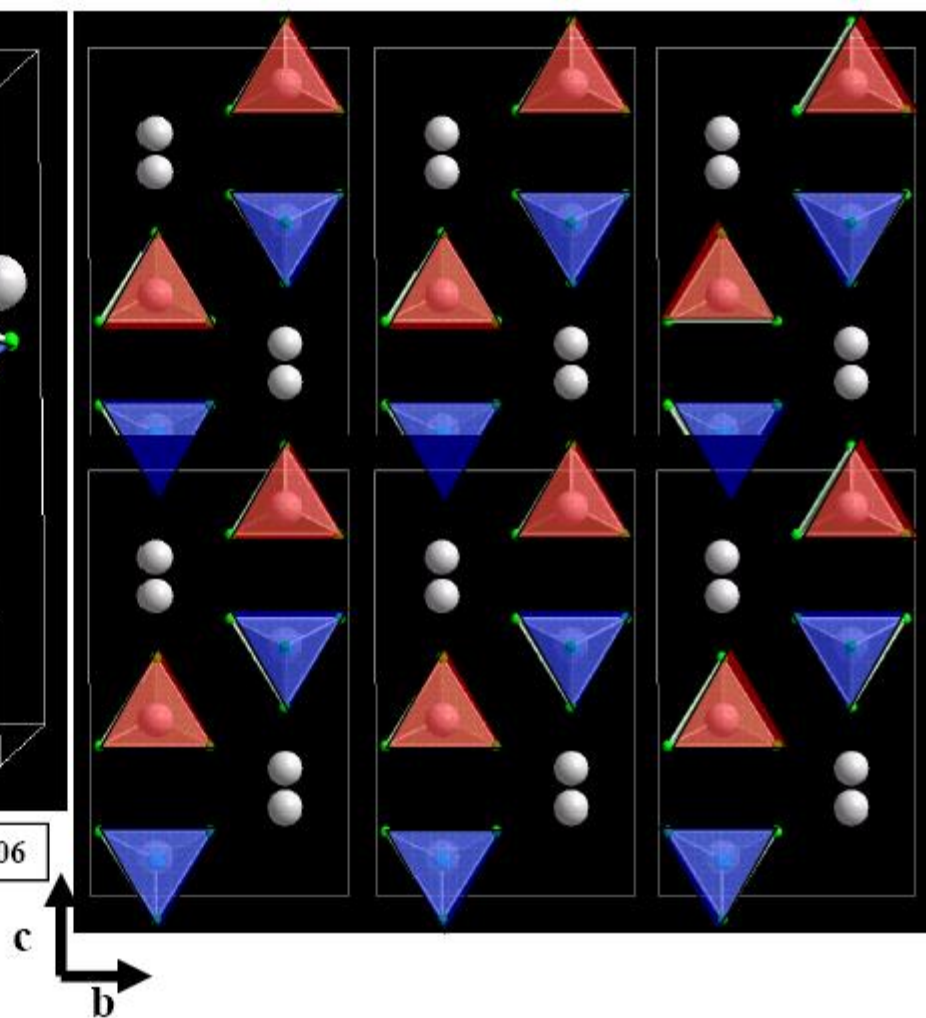
Depart. Of Chemistry, University of Cambridge:

Dr. J. E. Davies.

Structure of Cs_2CuCl_4

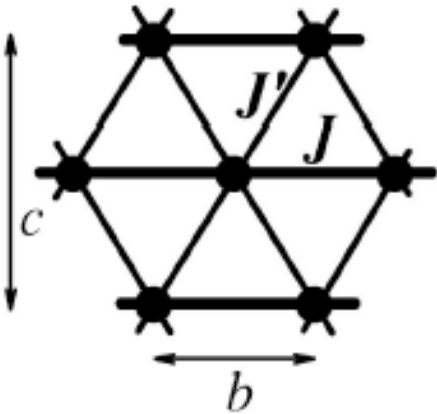


J.A. McGinney, *J. Am. Chem. Soc.* 29 (1972) 8406

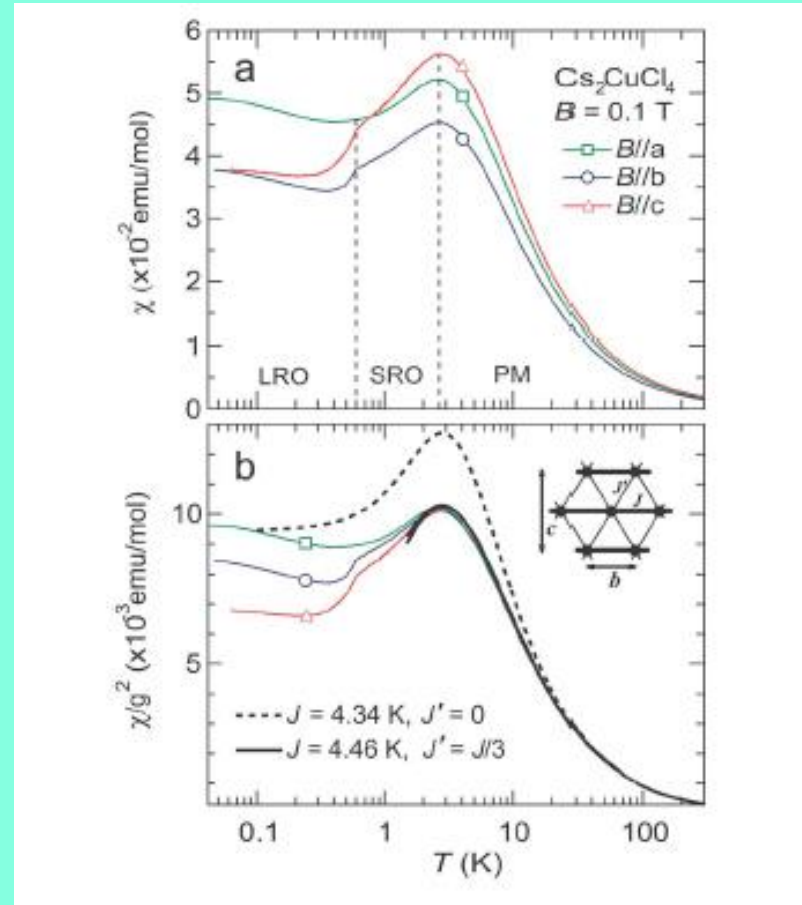


Magnetic structure of Cs_2CuCl_4

2-dimensional Frustrated antiferromagnetic spin liquid phase

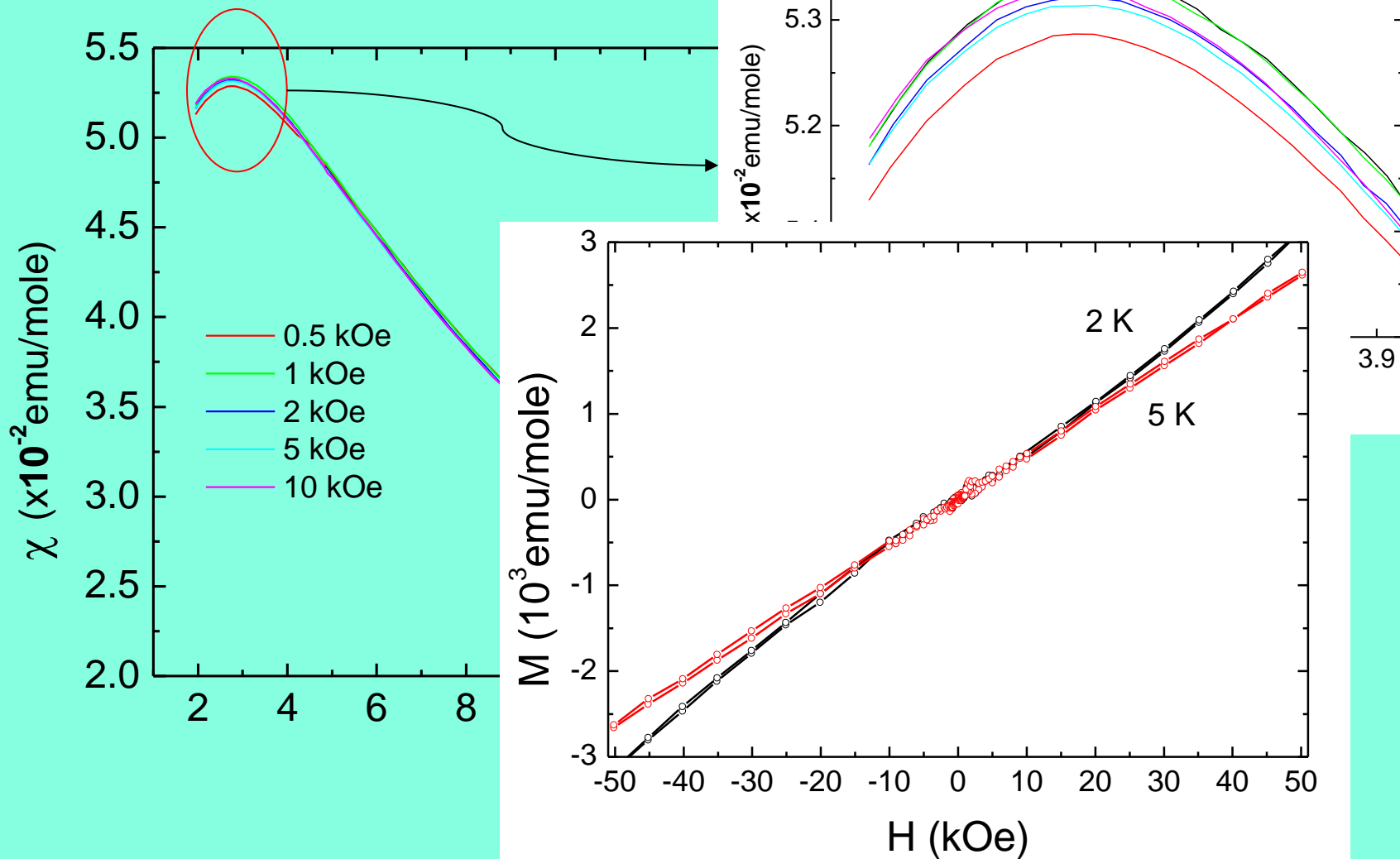


The ratio of $J'/J = 0.34$



Tokiwa *et al.* PRB **73**, 134414 (2006)

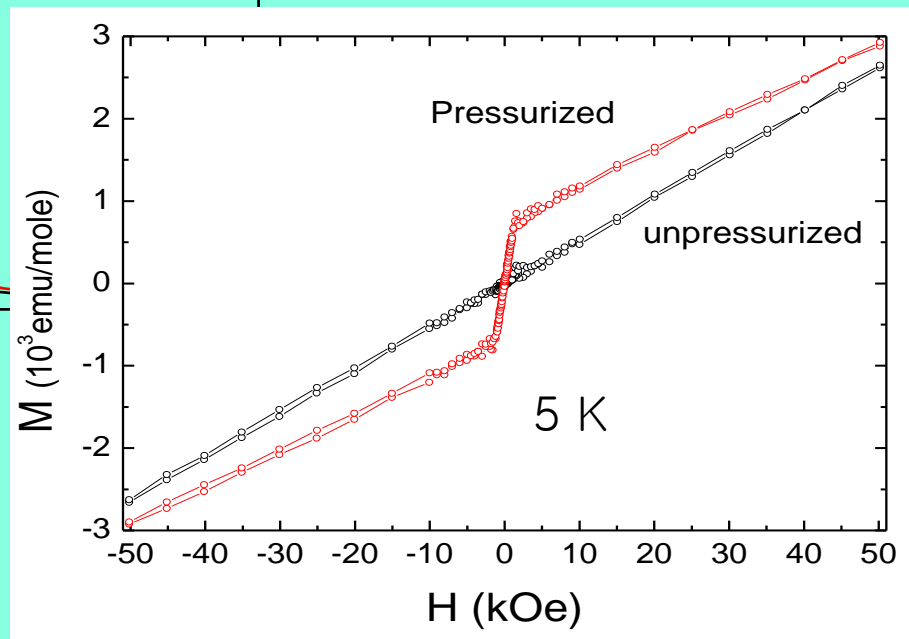
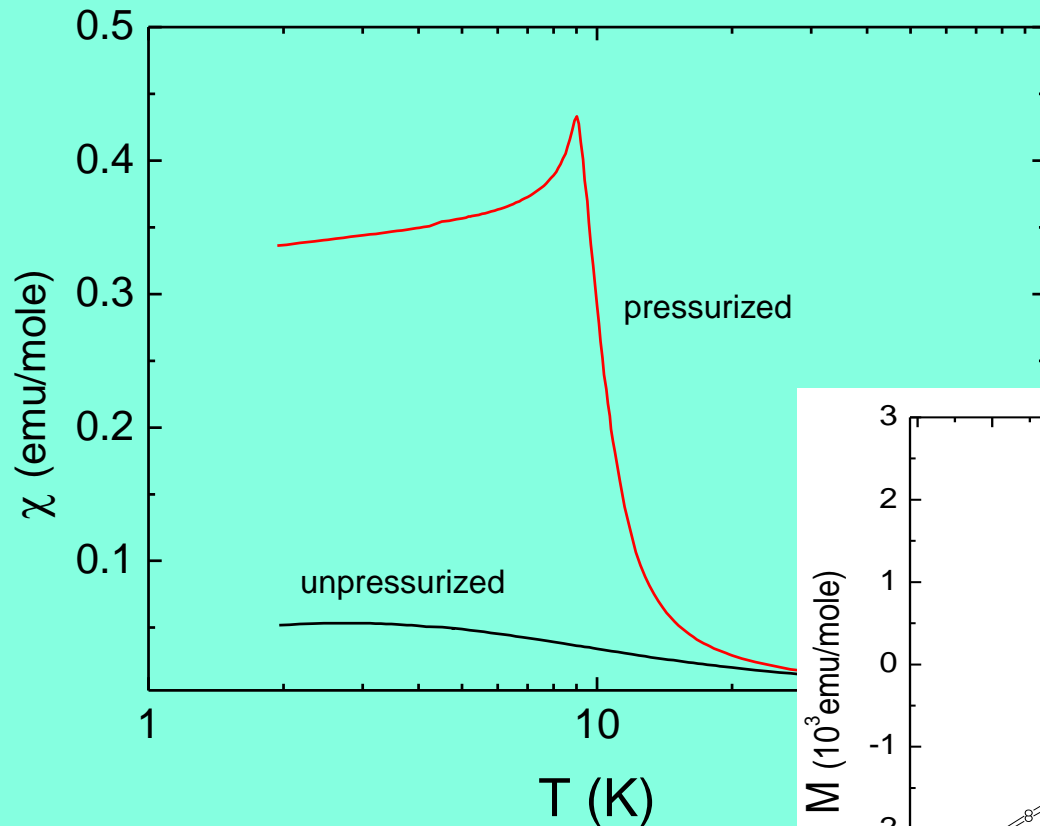
Magnetization of unpressurized Cs_2CuCl_4



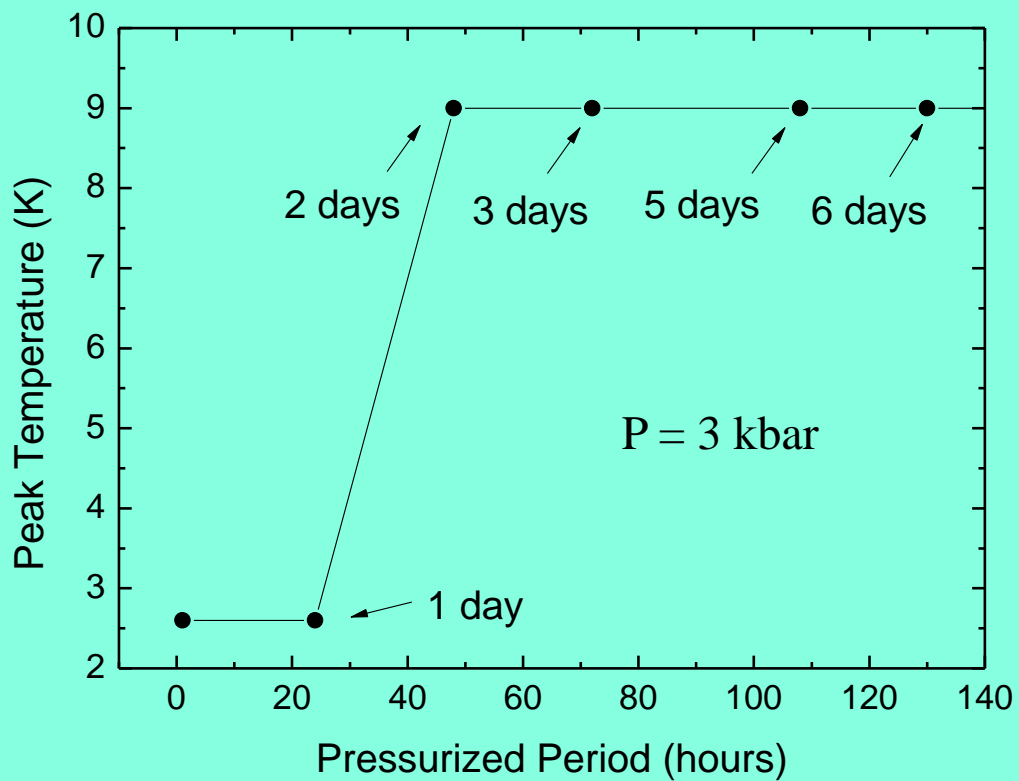
Pressurized process

1. Cs_2CuCl_4 was put into the hydrostatic pressure cell and pressurized above a 3 kbar.
2. Sample was kept in the pressure cell for a few days at room temperature.
3. The sample was taken out from the pressure cell.
4. Magnetization of the pressurized sample was measured using SQUID magnetometer under ambient pressure ($P = 0$ kbar) .

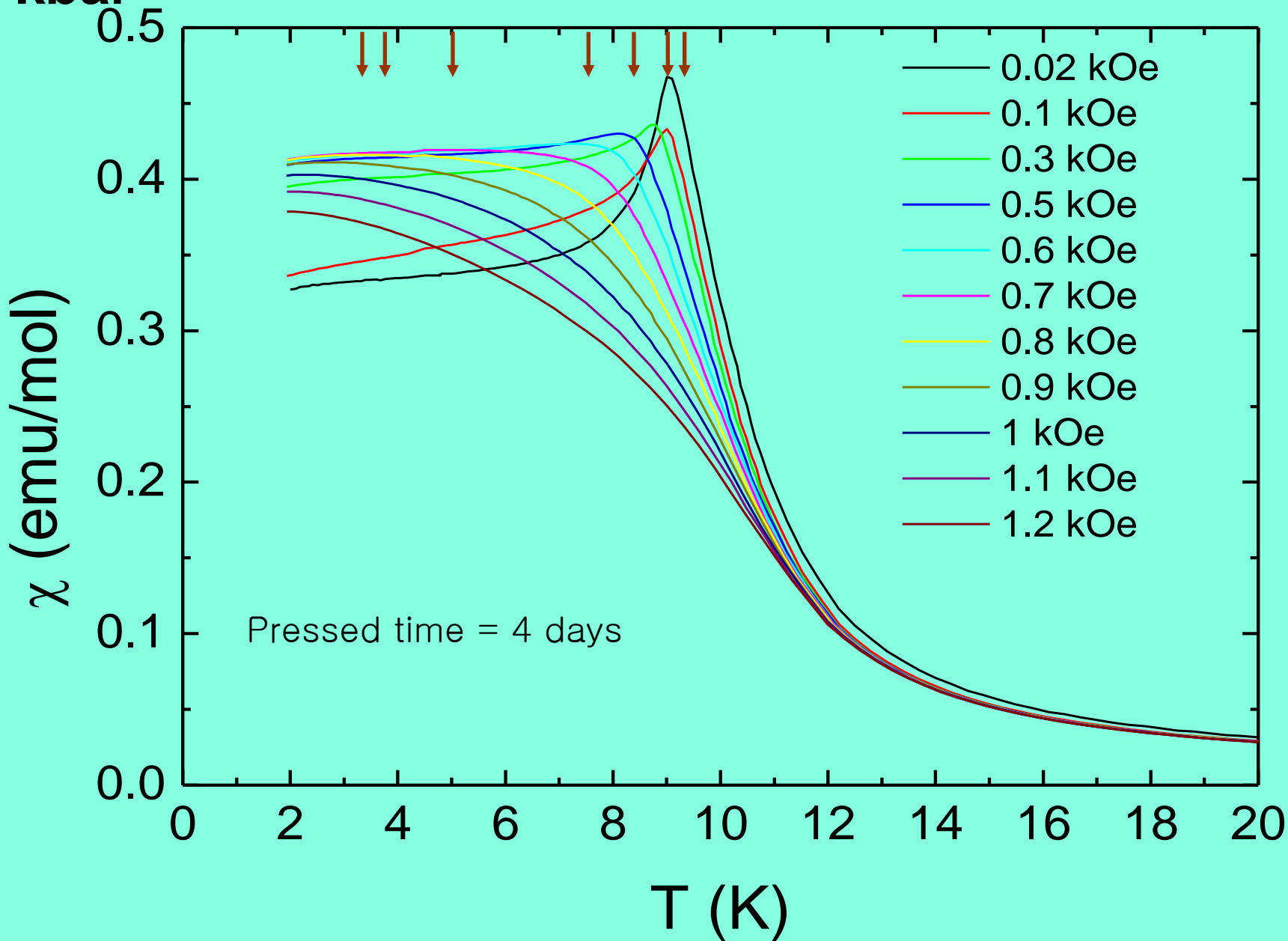
Comparison between Unpressurized and Pressurized Cs_2CuCl_4



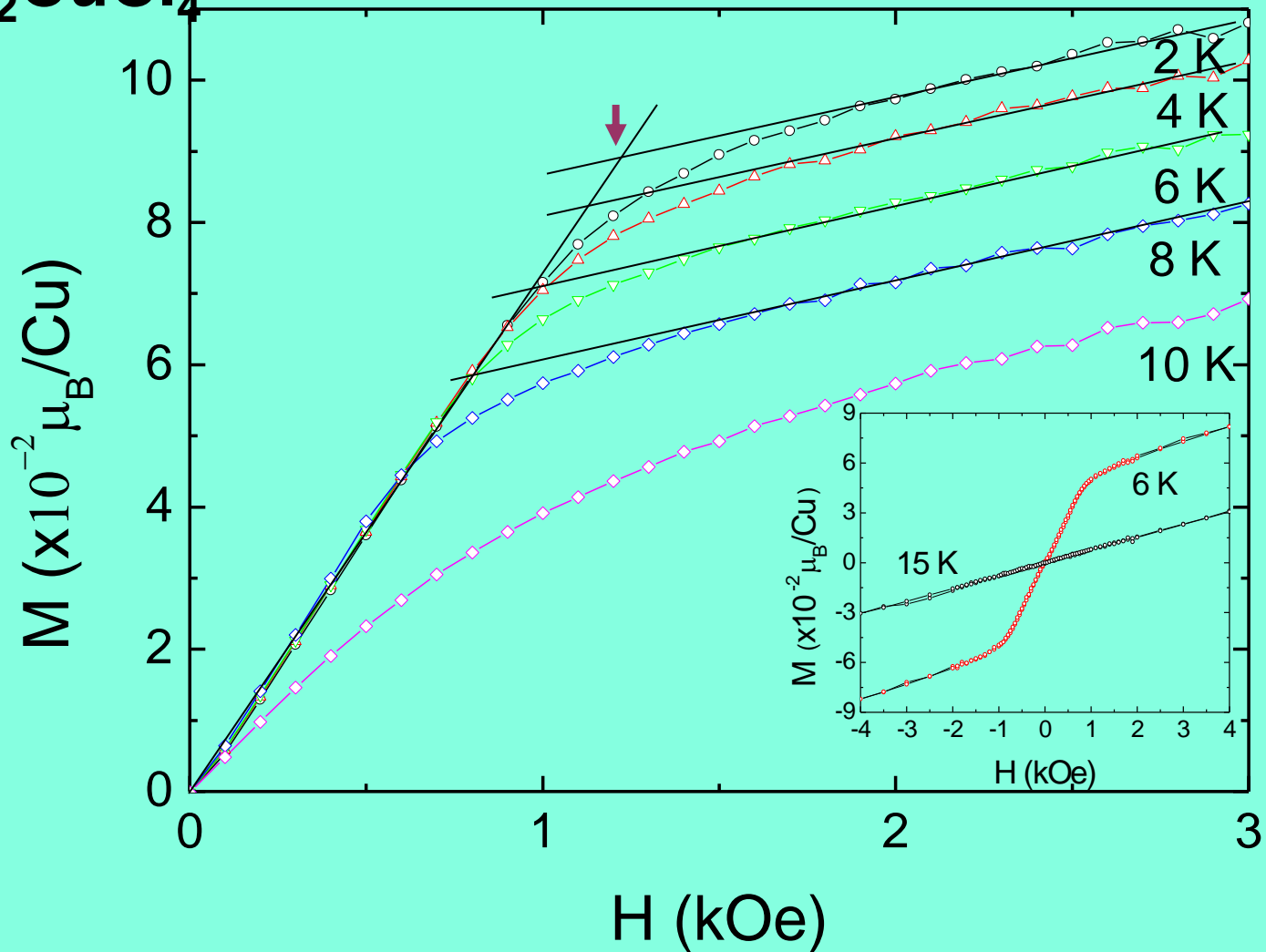
Pressing time dependence of phase transition



Magnetization of Pressurized Cs_2CuCl_4 under $P = 0$ kbar



Magnetic curves at each T for pressurized Cs_2CuCl_4



Similar behavior of Helimagnet magnetization

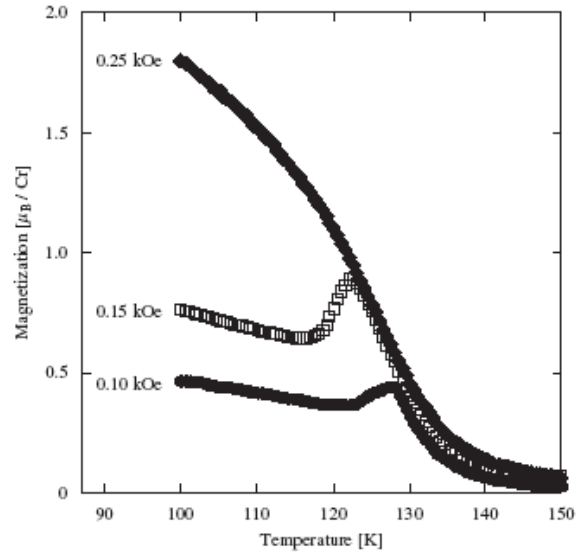


Fig. 1. Temperature dependence of the magnetization under the magnetic field along the hexagonal a - b plane in $\text{Cr}_{1/3}\text{NbS}_2$.

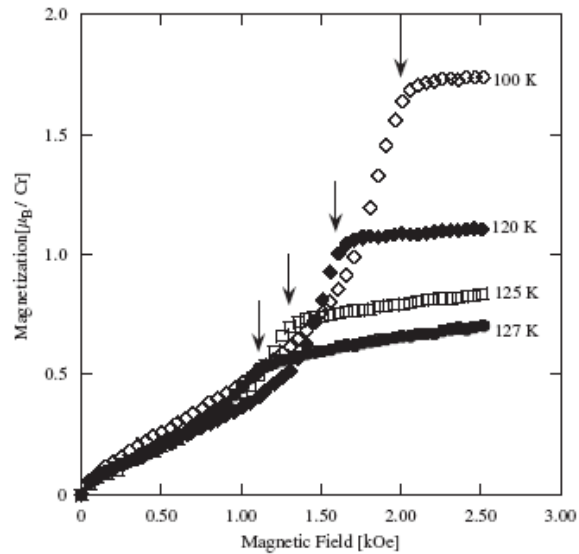


Fig. 3. Magnetization process under the magnetic field along the hexagonal a - b plane in $\text{Cr}_{1/3}\text{NbS}_2$.

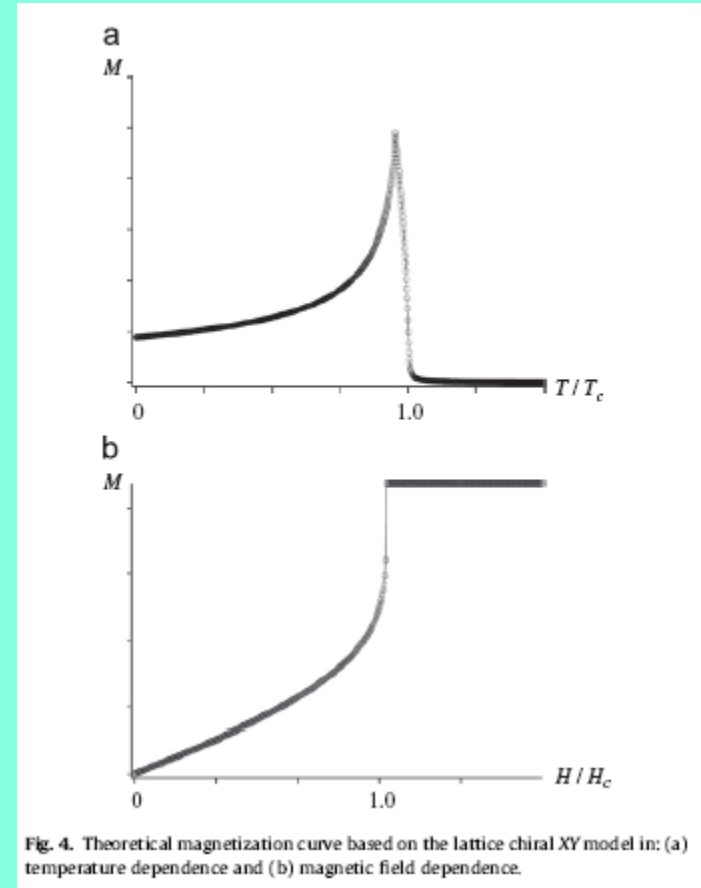


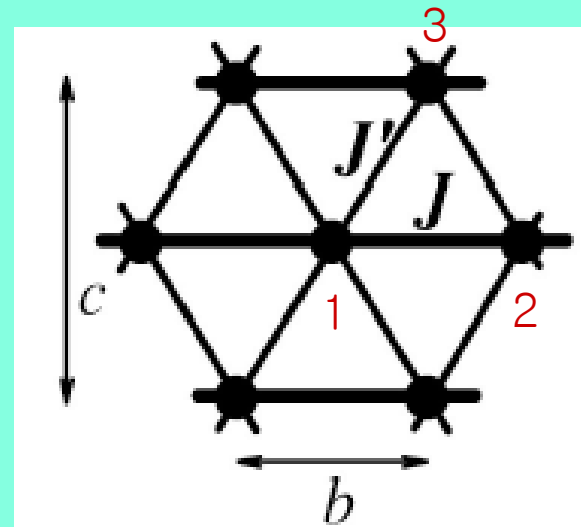
Fig. 4. Theoretical magnetization curve based on the lattice chiral XY model in: (a) temperature dependence and (b) magnetic field dependence.

Y. Kousaka *et al.* (2009)

X-ray diffraction results at RT under $P = 0$

kbar

	Unpressurized Cs_2CuCl_4 (i)	Pressurized Cs_2CuCl_4 (ii)	(ii) - (i)
a (Å)	9.7652(4)	9.7663(4)	0.0011(4)
b (Å)	7.6338(3)	7.6094(3)	-0.0244(3)
c (Å)	12.3858(5)	12.4146(5)	0.0288(5)
V (Å ³)	923.31	922.6	-0.71



c-axis ↑

b-axis ↓

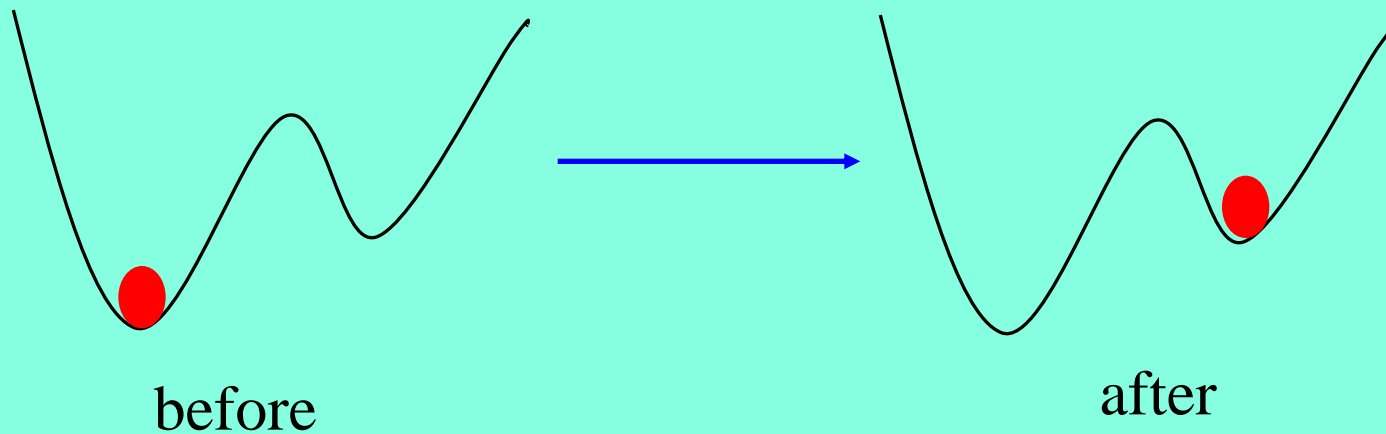
distance	Unpressurized Cs_2CuCl_4 (i)	Pressurized Cs_2CuCl_4 (ii)	(ii) - (i)
1-2	7.6338	7.6094	-0.0244(3)
1-3	7.2747	7.2805	0.0058(6)

J coupling ↑

J' coupling ↓

J'/J ↓ < 0.34

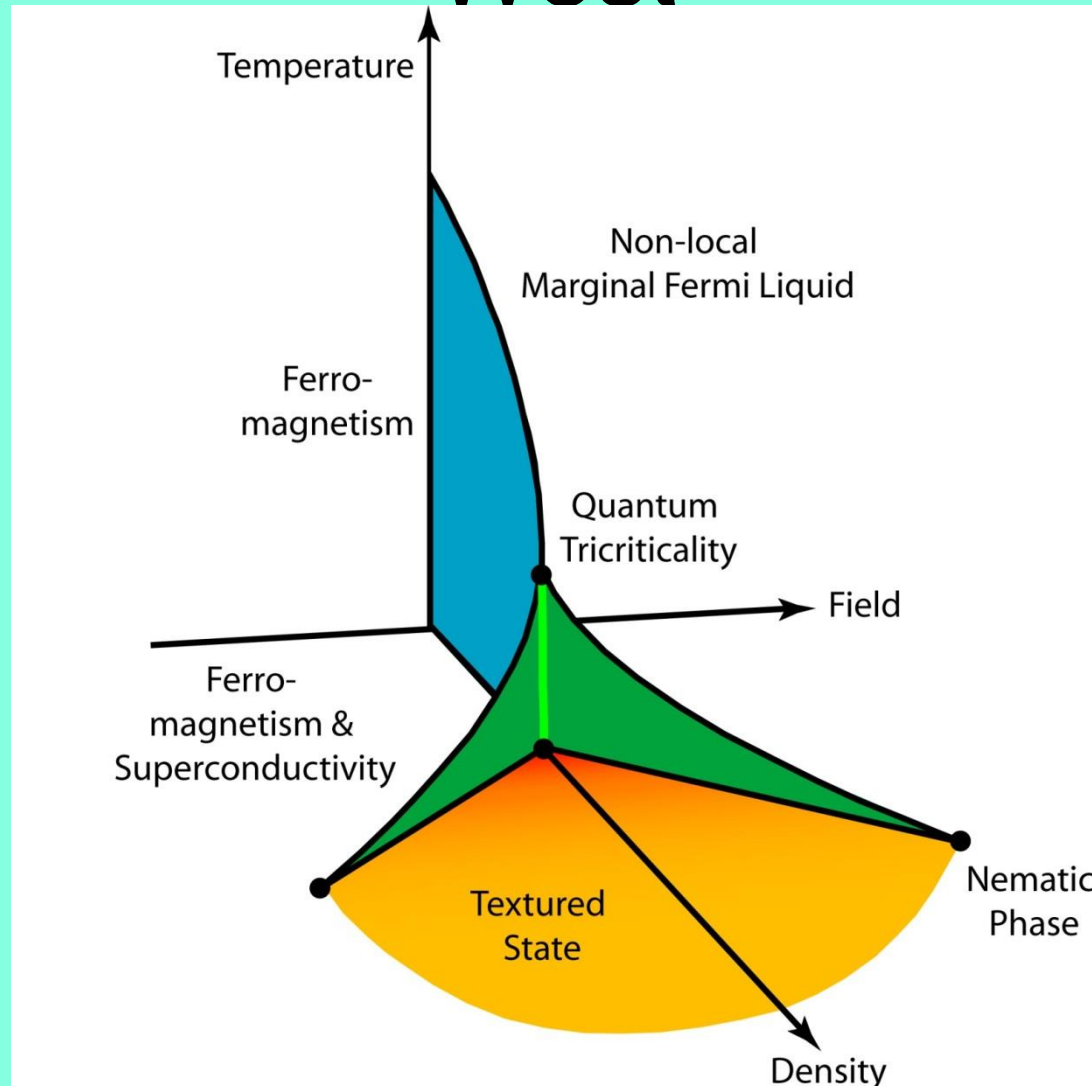
- Structure of pressurized Cs_2CuCl_4 is slightly distorted.
- Magnetic interaction of pressurized Cs_2CuCl_4 may transit into metastable magnetic structure. (J'/J is changed).



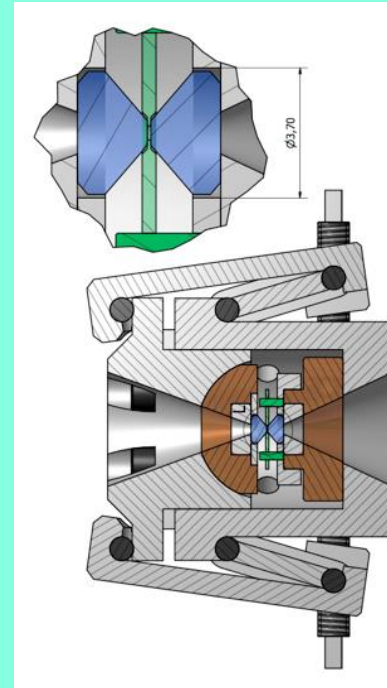
Summary

- Observation of new metastable magnetic phase in pressurized frustrated antiferromagnet Cs_2CuCl_4 driven by low pressure.
- Crystal structures does not changed but the lengths of b - and c -axis are slightly changed.
- It is considered to be due to the change of coupling competition between J and J' .

Border of Order: A Quantum Wild West



Automated Temperature-Field-Pressure Tuning

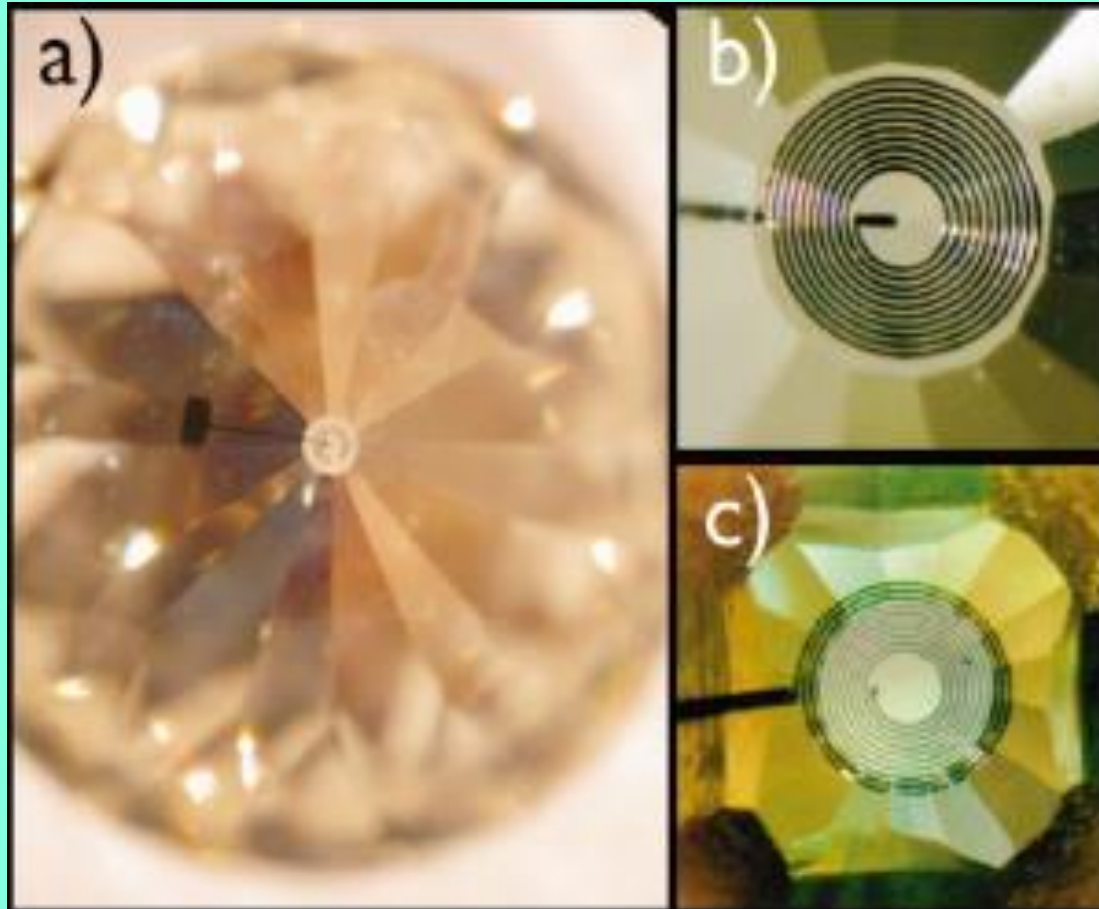


Cryogen-Free Pulse Tube with Magnetic Refrigerator
& Variable Pressure System

Cavendish Laboratory, CamCool Ltd
IHT Kazakhstan, CHT Uzbekistan

Diamond Anvil with Sensing Microcoil Protected by Diamond Overcoat

(a)
Designer
Diamond
Anvil



(b)
Expanded
View of
0.3 mm
Culet
with
Sensing
Microcoil

(c)
After
Encasing
with
Diamond
Layer

DD Jackson, *et. al.*, Phys. Rev. B (2005)
184416

Technology Focus

Creating a research and innovation platform

88 Noble Prize Winners on one hand

6.5 Billion Dollars of Venture Capital on the other hand

Development Contextualisation

From Technology Transfer → Technology development

Cambridge Innovation Vision

Nurture, both, organic and induced technology development pathways

Support innovation at both industrial and individual scale

Nurture talent, both locally and internationally

Human Capital Development, Technological Advancement and Environmental Sustainability must operate from the same platform

Eurasia Sci-Tech Platform Initiative

- The primary objective of the CIS Sci-Tech Platform initiative is to establish the basis for the technological advancement and sustainable long-term growth of the Central Asian economies
- The initiative looks at the three key determinants of economic growth: *Labour, Capital and Technology* and conducts in-depth review of the current state of these determinants to guide establishment of technology clusters
- Examples are The Tashkent Centre for High Technology, Partnership with Kazakh National Wealth Fund companies. New Projects in Kyrgyzstan and Azerbaijan.

Tashkent Centre for High Technologies

A unique centre offering the latest technical services, training and innovation advise

A human capital development platform for training Uzbek scientist, engineers and technology entrepreneurs

A high-end research hub for physical, chemical, biological and engineering sciences

Cambridge & UK academics, business persons and entrepreneurs will provide training and advise

First step towards creating a innovation eco system

Tashkent Centre for High Technologies

A Company called Cantabrigia Advisors Ltd. (www.cantabrigia-advisors.com) was launched to interface between Cambridge Enterprise, Cambridge University Technical Services, Cambridge Central Asia Forum and Uzbekistan Academy of Sciences, Ministry of Education to deliver the CHT

To initiate the project the Uzbek government has committed more than 25 million euros for the latest scientific equipment and a further amount for creating building and management infrastructure and more than a million euros for training and education of scientific staff

Presidential Decree in October 2011 gives us a full go ahead!

Tashkent Centre for High Technologies



Asymmetry in Global Science and Technology

On one side the Western/Northern systems of education are purported to be the Best!

While on the other side it fails to produce both good enough and enough skilled labour for the business and institutions in The Western countries themselves

This discrepancy is largely overcome by importing both the students and the researchers from emerging economies of Asia and Eurasia

This generates a philosophical and practical anomaly in the form of the highly trained supply is coming from places marked as inferior in delivery of desired education and training!

Summary

- Quantum Critical Regime offers frame work for new discoveries.
- Led to the case of magnetically mediated superconductivity and first example of itinerant electron ferromagnetic superconductor is discovered.
- Magnetism and Low Dimensionality are found as physical attributes which aid superconductivity at elevated temperatures.
- This road map has led us to the very different case of Graphite Intercalates and Quantum Critical Behaviour of Ferroics and Multiferroics
- Non invasive tuning parameter like high-pressure not only makes new physics tractable, but can also serve as a tool for new material design.

Key Publications

Superconductivity in graphite intercalated compound C_6Yb at 6.5K and C_6Ca at 12K
(Highest of any graphite and Ce, Yb or U based compound)

T. Weller et al. *Nature Physics*, Vol. 1, pp 39-41

Superconductivity in itinerant electron ferromagnet UGe_2
(First example of superconducting pairing of electrons in an itinerant ferromagnet)

S.S. Saxena et al. *Nature*, Vol. 406, pp. 587-92

Prediction and possible explanation of superconductivity in high pressure phase of Iron
(Anisotropic pairing as only very pure specimen superconduct. High magnetic susceptibility)

S.S. Saxena, P.B. Littlewood, *Nature*, Vol. 412, pp. 290-291

Possible explanation of superconductivity in non centro-symmetric magnet $CePt_3Si$
(Magnetic non centro-symmetric materials possibly have exotic mixed-state pairing)

S.S. Saxena, P. Monthoux, *Nature*, Vol. 427 pp 799-799

Marginal Breakdown of the Fermi Liquid State on the Border of Metallic Ferromagnetism
(Observation of a Singular Fermi Liquid)

R. P. Smith et al. *Nature*, Vol. 450, pp 1220-1223