NATIONAL HIGH MAGNETIC FIELD LABORATORY

National Nuclear Security Administration

FLORIDA STATE UNIVERSITY 1851


NHMFL/FSU/TCC
A. Grockowiak
J. Burke
L. Riner
J. Krehl
F. de Torres
A. Yannakopoulos
L. Rodenbach
A. Estry
A.Williams
**M. Woods**

**W. Coniglio**
D. Graf
T.P. Murphy
J-H. Park
P. Schlottmann
Robert Schwartz
J. Whalen/A. Chown


# Extreme conditions for strongly correlated electronic systems


Audrey GROCKOWIAK, Stanley TOZER
W.Coniglio, D.Graf, J-H. Park, T.Murphy

grockowiak@magnet.fsu.edu
tozer@magnet.fsu.edu

LANL
J.-X. Zhu
J.C. Cooley
J.C. Lashley
R.D. McDonald
A. Migliori
C. Mielke
F. Freibert
J. Mitchell
P. Tobash

BNL
C. Petrovic



This work was funded by the DOE/NNSA under DE-NA0001979 and was performed at, and funded by, the National High Magnetic Field Laboratory, which is supported by NSF Cooperative Agreement No. DMR-1157490 and by the State of Florida.

# Mission

- Educating high school, undergraduates, graduate students and postdocs

(high pressure, electronics, cryogenics, probe preparation, crystal growth…)
Past members :
Ryan Stillwell LLNL postdoc in Jason Jeffries group, spring 2014, Q clearance, in the process of being converted
Michael Woods U growth & preparation UVC Nuclear Engineering GS
Ashley Bernheisel Electronics Boeing & Electrical Engineering GS
Michael Bernstein Materials-U preparation ASU Physics GS

# 2015 activities

## Scientific mission :

- Physics of actinides, 4f and 5f electron systems with complex phenomena : charge density waves, volume collapse, superconductivity, quantum critical point…

- Apply external parameters (temperature, magnetic field, **pressure**) coupled with experimental techniques (Tunnel Diode Oscillation, dilatometry…) to understand fundamental properties of these materials.

- ➢ Fermiology of actinides and lanthanides : Tunnel Diode Oscillations studies under high pressure, high magnetic fields and low temperatures (**$CeCu_2Ge_2$**, Be, Ce, Cr, dU, $NiS_2$, $SmB_6$, …) (8 weeks of magnet time at NHMFL FSU and LANL, and HZDR)

- ➢ Dr. Tozer's eternal quest to understand Uranium : **high pressure XRD at ESRF**

- ➢ High pressure Fiber Bragg Grating Dilatometry for thermal expansion and magnetostriction (dU, Ce, $ScF_3$, **$CeSb_2$**, $CeCu_2Ge_2$,…)

- ✓ Developing techniques for these studies that can be transferred to DOE laboratories and NHMFL user program

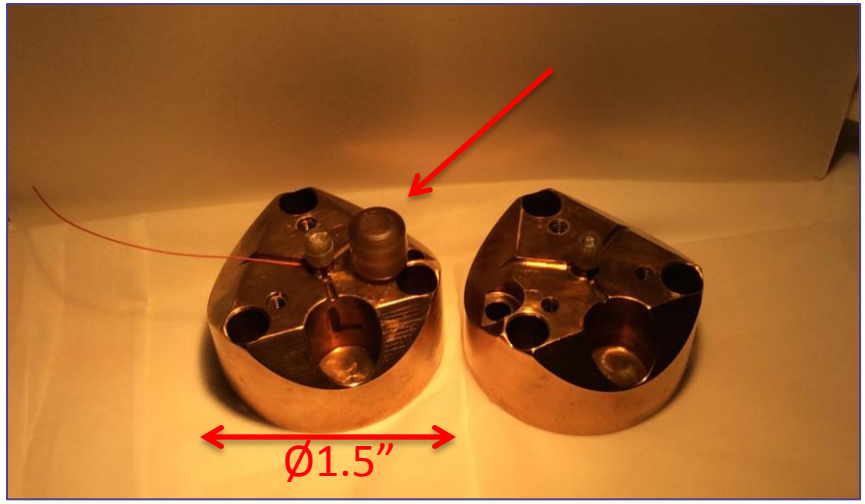
# Example : Instrumentation

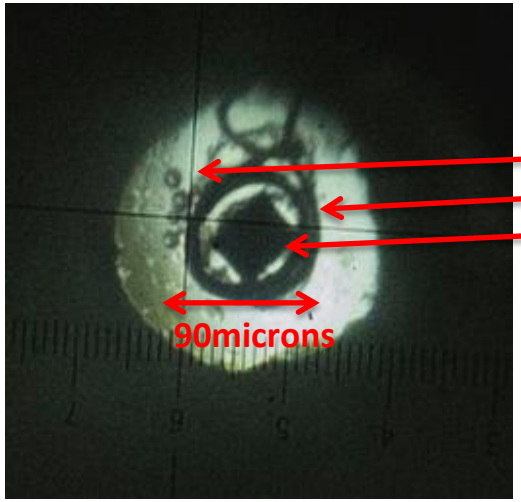
Tunnel Diode Oscillations (TDO) (William Coniglio)

- Contactless measurement => stress free, easy to load the samples, in part. radioactive ones.
- RF tank circuit driven by TD to measure penetration depth in metals and magnetization in insulators.

Non-metallic Diamond Anvil Cells (Stan Tozer and Audrey Grockowiak)

No heating or vibrations during pulsed fields, can be rotated in DC and pulsed magnets at cryogenic T




Ruby spheres
TDO coil
$SmB_6$ sample

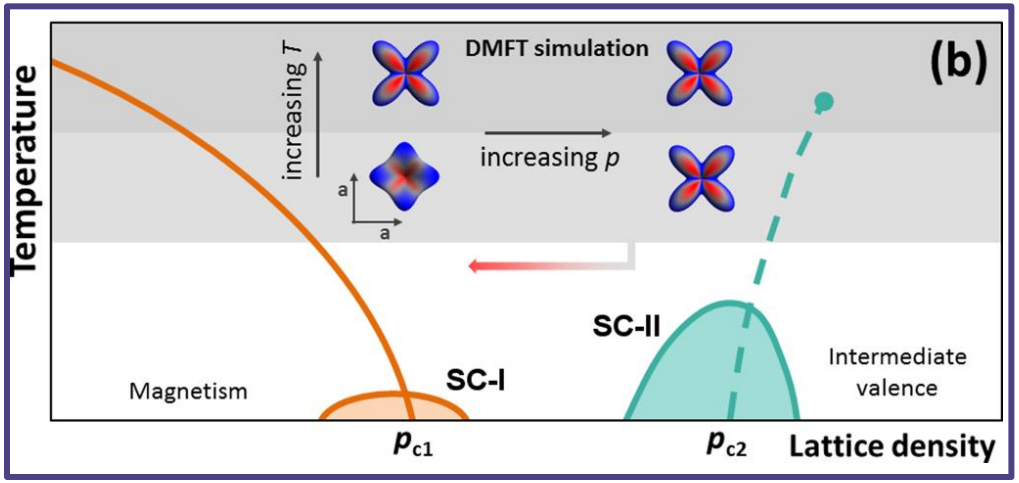
⇒ **Extreme conditions : 70 T and $^3$He temperatures**

⇒ **Current record : 115 kbar !**

# Example : $CeCu_2Ge_2$

- High pressure Tunnel Diode Oscillation and $CeCu_2Ge_2$

$CeCu_2Si_2$ : archetypal heavy Fermion system with 2 superconducting domes




PHYSICAL REVIEW B 91, 201108(R) (2015)
Science, vol32, 2003

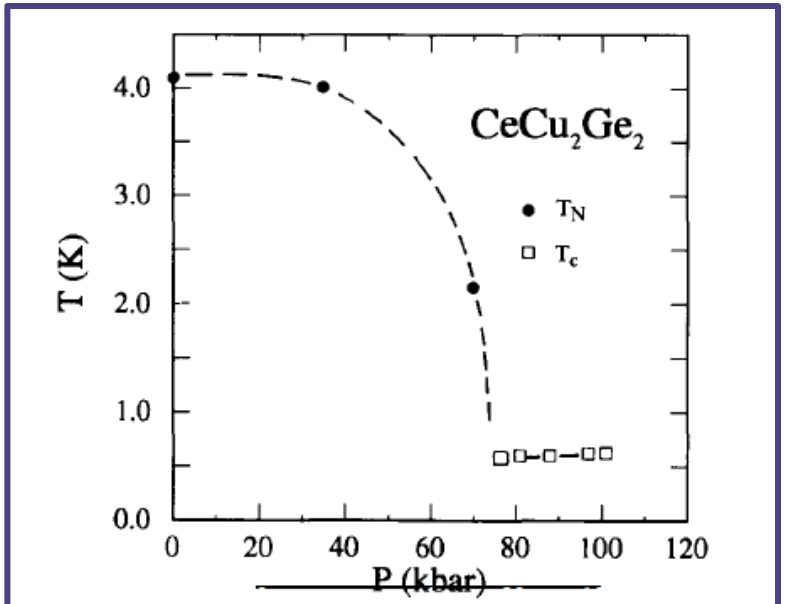



Fig. 6. *P–T* diagram of magnetic and superconducting phases of $CeCu_2Ge_2$.

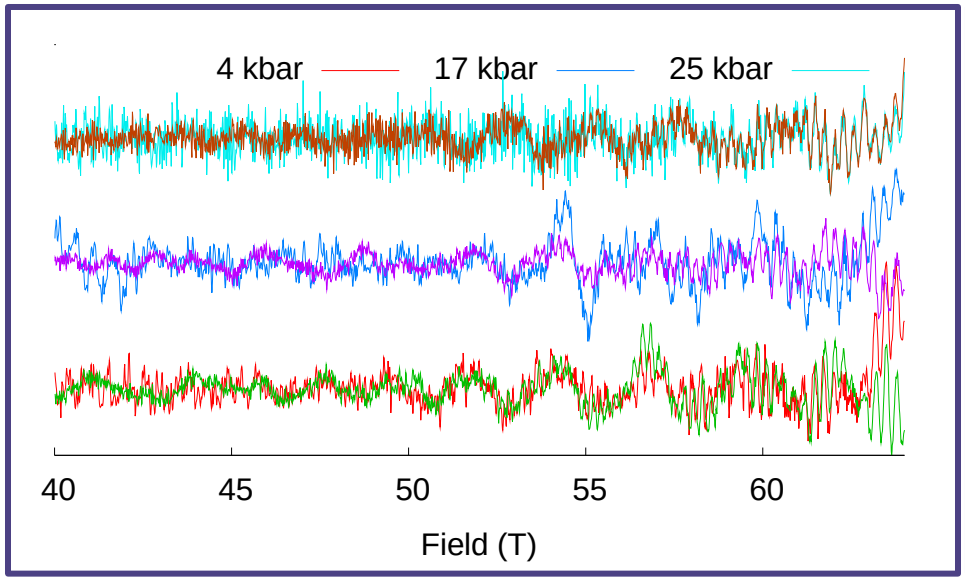
Jaccard et al., PhysicsLettersA 163 (1992) 475—480

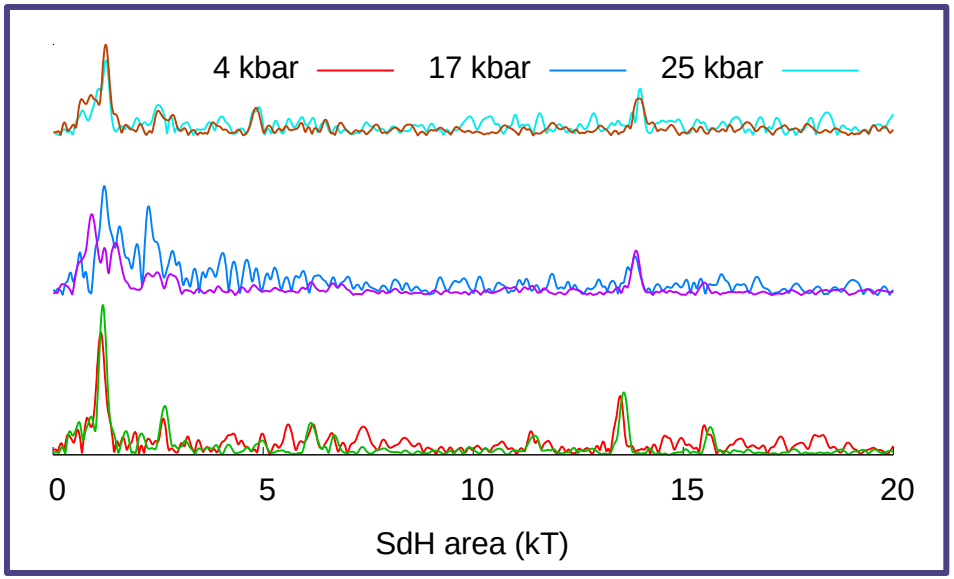
$CeCu_2Ge_2$ : parent compound, ~ Si compound at -12GPa
1 SC dome close to AFM transition
2 Quantum Critical Points

Study the Fermiology as a function of pressure, and measure SC in CeCu2Ge2

# Example : $CeCu_2Ge_2$

- High pressure Tunnel Diode Oscillation and $CeCu_2Ge_2$




Amplitude of the QO detected by the TDO as a function of field (LANL) for various pressures

Fourier Transform of the QO reveal different orbits of the Fermi surface, shifting with pressure.

Data taken at $^3$He temperature, field orthogonal to the (a,b) plane
Currently mapping the P,B,T and angle phase space to get complete picture of the Fermi surface of $CeCu_2Ge_2$ (3 weeks of magnet time in HZDR Dresden, April 2016)
Can use dHvA signal from Cu to calibrate magnets :
Poster **MAT41** by W. Coniglio : LANL : 101.17+/-008 T
"High magnetic field calibration using de Haas-van Alphen oscillations in polycrystalline copper"

# Example : HP XRD of dU

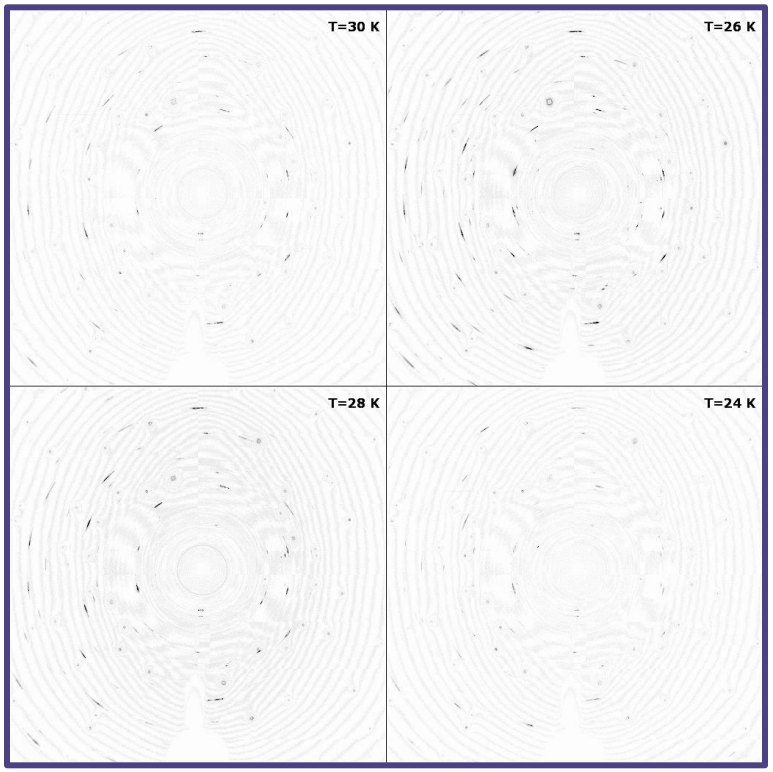
- Depleted Uranium :

Attempt to study crystal structure under pressure and at low temperatures

3 Charge Density Waves : 43 K, 37 K, 23 K

ESRF, Grenoble : only facility to allow low temperature and high pressure on single crystals

ID27 (Mohammed Mezouar and Gaston Gabarini) + Prof. McMahon (Edinburgh)




XRay diffraction patterns at various temperatures

Ambient pressure structural transition between 20 and 30 K

Stable structure below 24 K and above 30 K

Intermediate temperatures : transition region many peaks change intensity dramatically

=> Next run : July 2016

-plastic DAC are transparent to XRD (moderate background) ! (There is a great future in plastics…)

Allows :

-complete rotation of the DAC (best limit : 90°)

-perform XRD in pulsed fields (currently 30T)

-simultaneous Fermiology measurement with our TDO set up

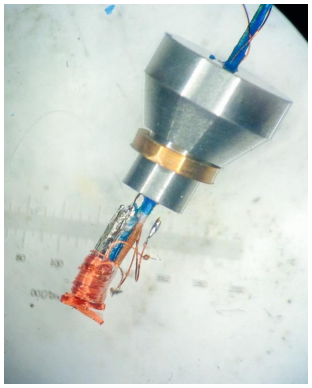
# Example : ΔL/L of $CeSb_2$

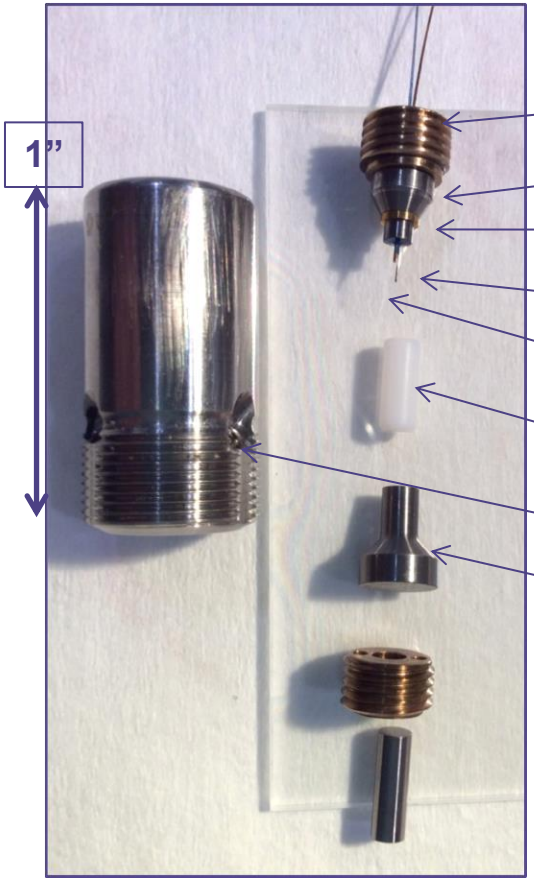
- High pressure thermal expansion and magnetostriction

- **How does it work ?**
  - Fiber Bragg Grating Dilatometry : POSTER **MAT42**
- **Why is this important ?**
  - Thermodynamic quantity (proportional to Free Energy)

Permits to probe changes in Free Energy,
Shown to detect phase transitions, superconducting transitions, quantum oscillations…
(Remember Clausius-Clapeyron equation and Ehrenfest relations)

- **Next steps :**
  - "μ-me" to fit in 70T magnet bore at HZDR (.354")
  - Coupling with TDO setup for simultaneous fermiology and dilatometry studies.






**Enable coupling with optical fibers**
**P max : 30 kbar = 3 GPa**

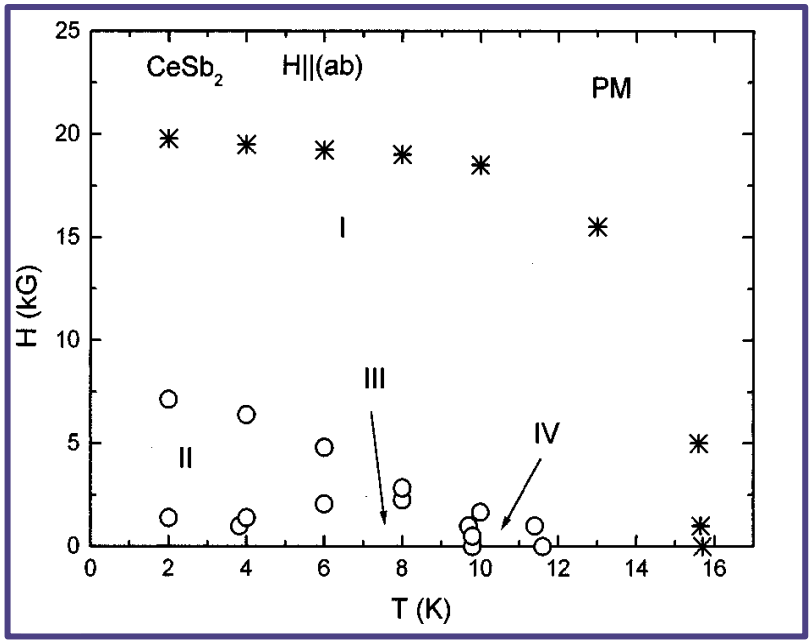
# Example : ΔL/L of $CeSb_2$

- Study of $CeSb_2$ – Collaborator : Pr. M.Grosche, Cavendish Lab

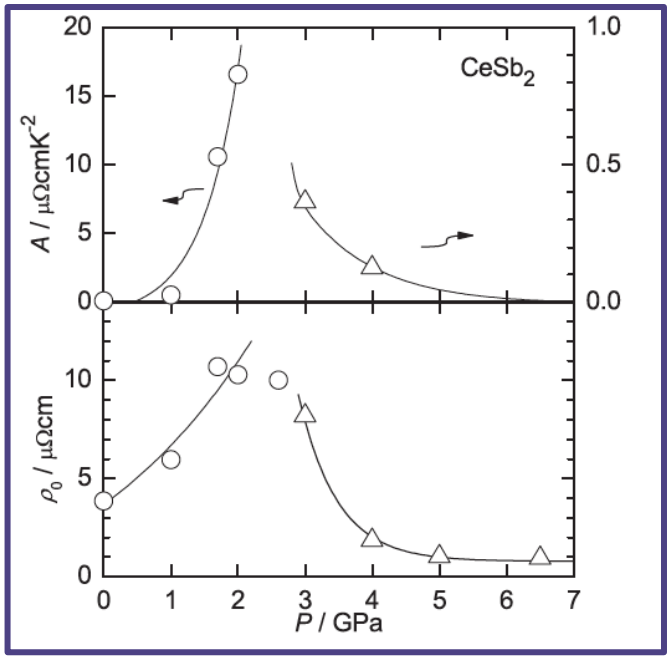
$CeSb_2$ : orthorhombic heavy fermion ferromagnet with Kondo lattice

3 low-T magnetic transitions : $T_c$ ~15.5 K, 11.7 K and 9.3 K

Likely candidate for field or pressure tuned quantum critical point around 26 kbar



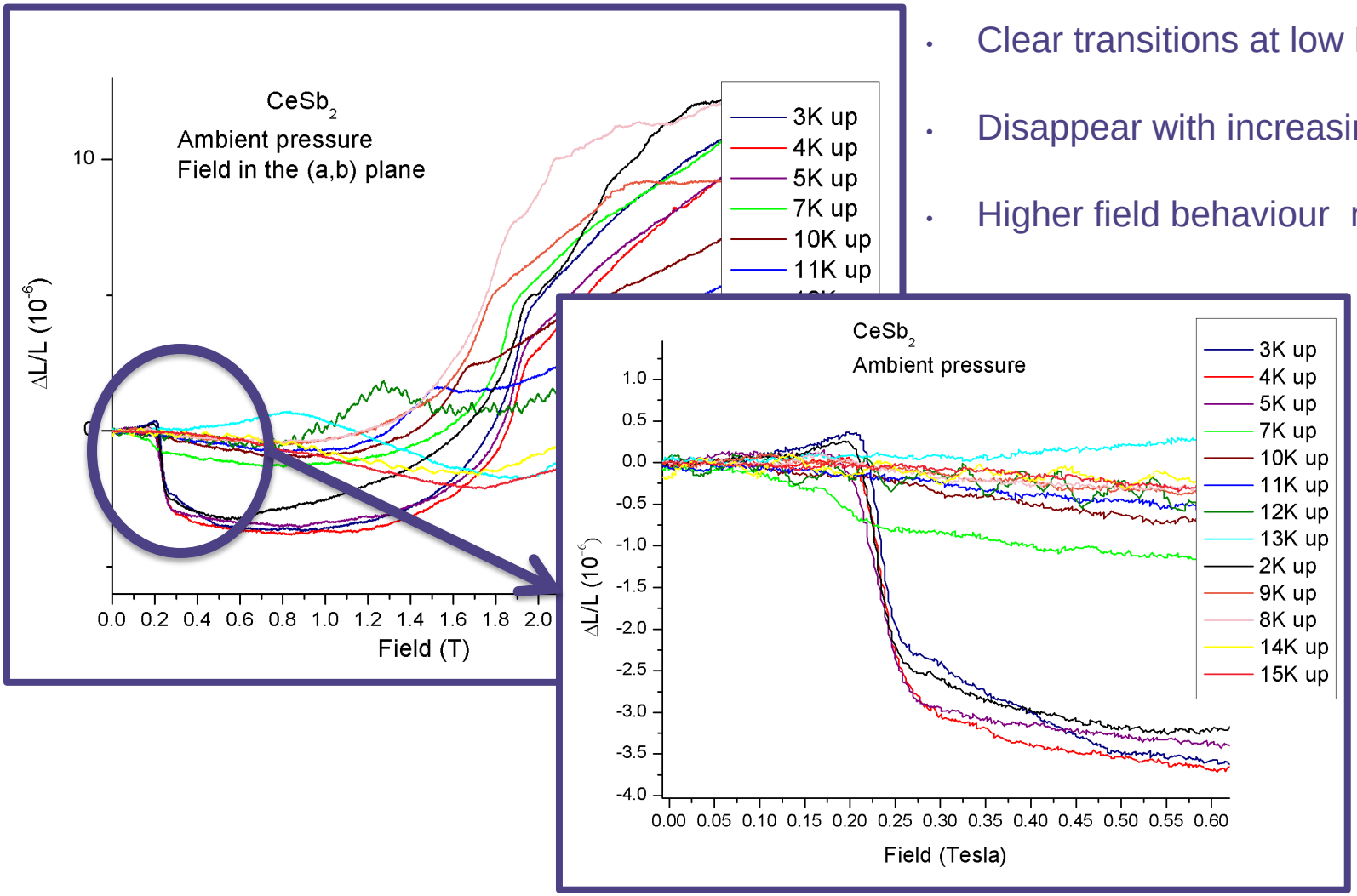

Tentative H-T phase diagram from magnetization and magnetoresistance measurements, ambient pressure
(Canfield et al., PRB57, 21 (1998))




The coefficient A of $T^2$ dependence of ρ, and the residual resistivity as a function of pressure suggests a QCP at 26 kbar
T. Kagayama et al. / Physica B 359– 361 (2005) 320–322

# Example : ΔL/L of $CeSb_2$

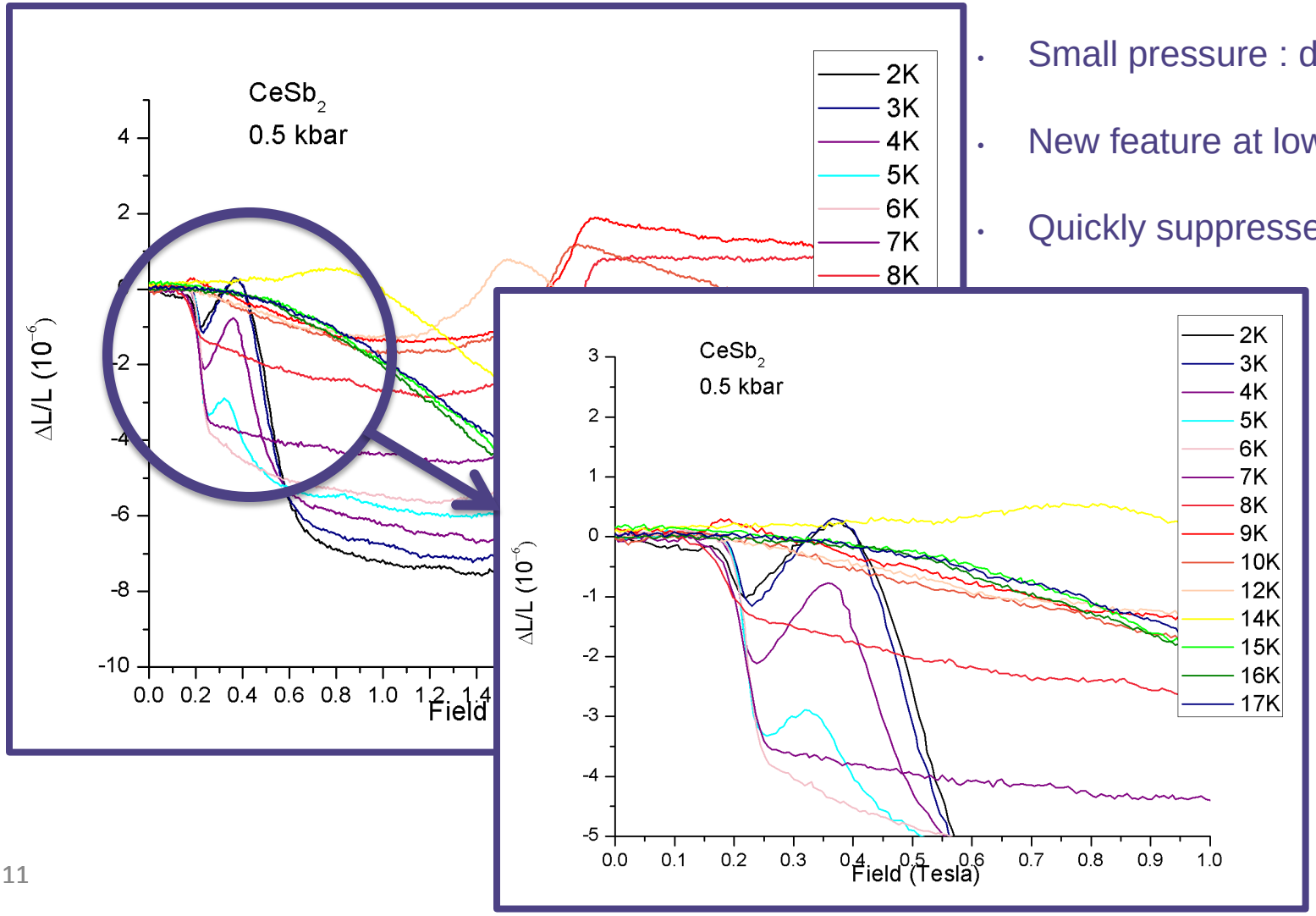
- Magnetostriction of $CeSb_2$ : ambient pressure




- Clear transitions at low H
- Disappear with increasing T
- Higher field behaviour not trivial

# Example : ΔL/L of $CeSb_2$

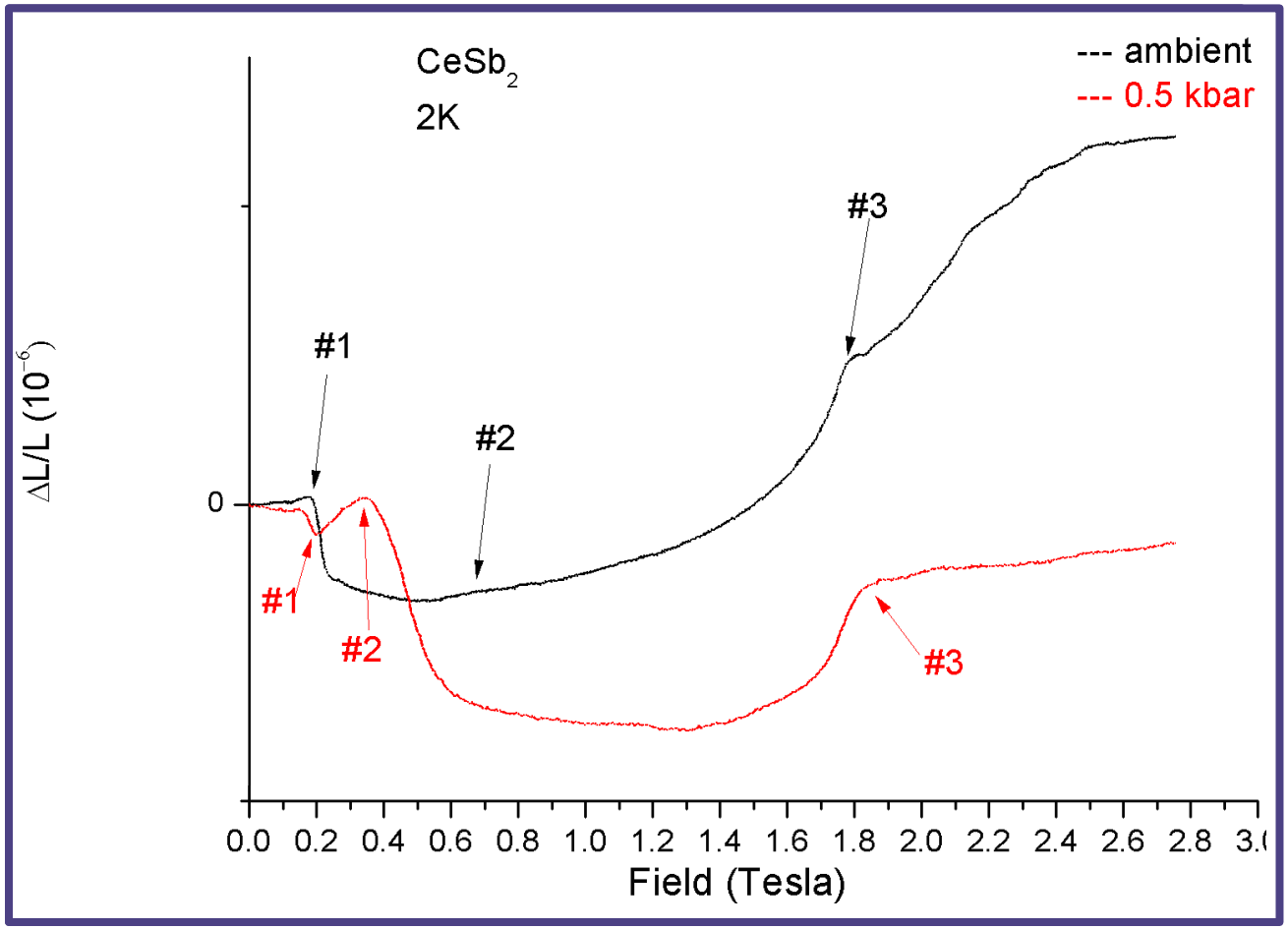
- Magnetostriction of $CeSb_2$ : 0.5 kbar pressure




- Small pressure : drastic change
- New feature at low field
- Quickly suppressed by 5K

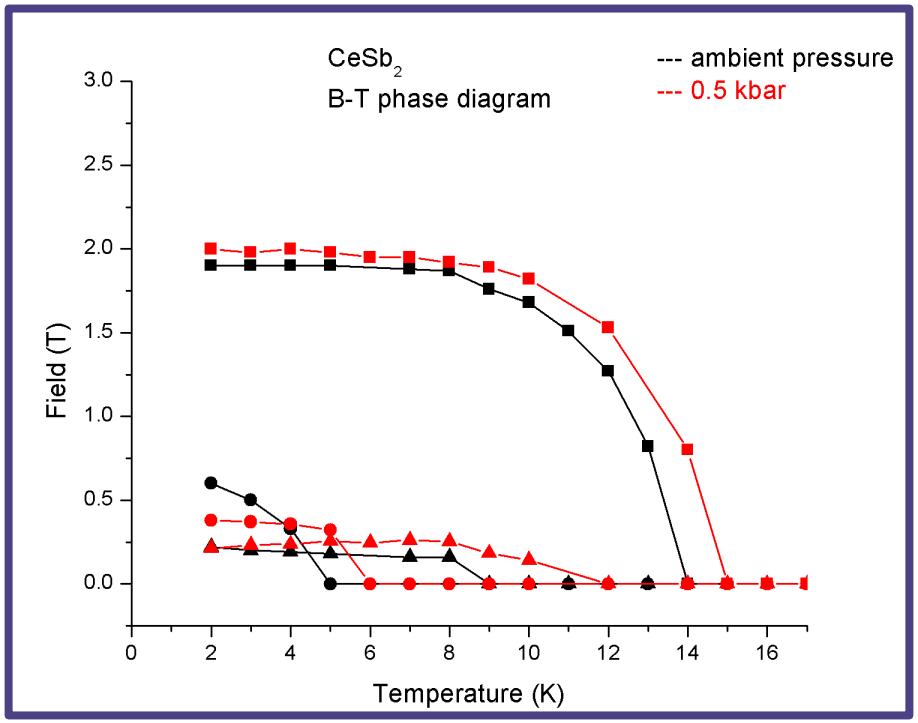
# Example : ΔL/L of $CeSb_2$

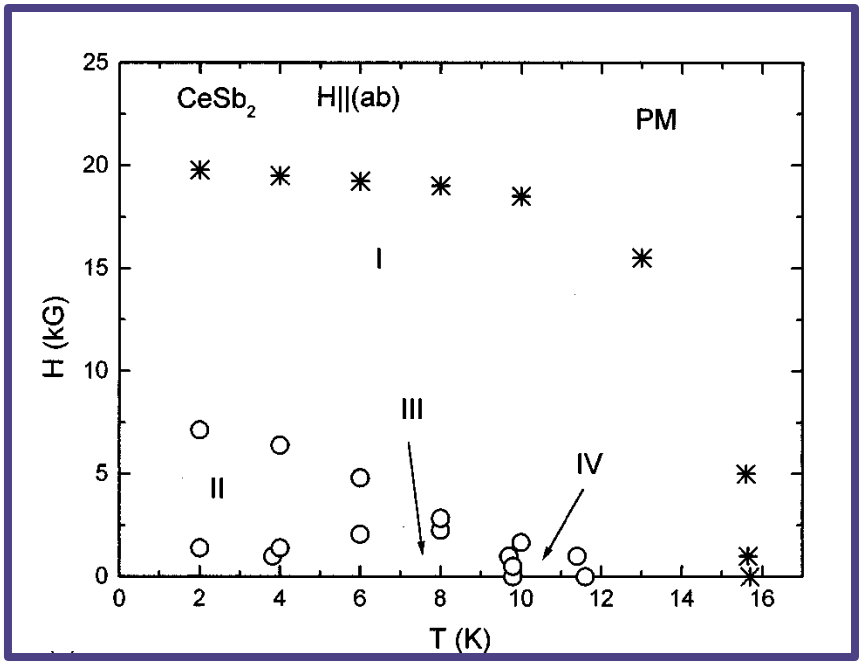
- Magnetostriction of $CeSb_2$ – Defining the low H transitions



# Example : ΔL/L of $CeSb_2$

- Tentative B-T phase diagram of $CeSb_2$



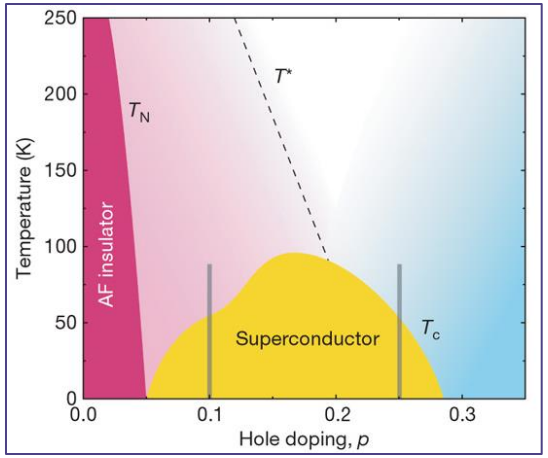




- Good agreement with previous experiment
- Transitions are enhanced with small pressure (as also reported in Physica B 359–361 (2005) 320–322)
- ➢ Currently increasing the pressure in order to detect the QCP

# Prospectives

- $CeCu_2Ge_2$ : finish high pressure fermiology
- Diamond Anvil Cell + Electronic transport with 4 contacts for resistivity and Hall Effect measurements
- Improve hydrostaticity condition in our DACs in order to observe fermiology above 100 kbar:
  oil or alcohol => Helium (M. Somayazulu and M.Ahart, Carnegie Institution, Geophysical Lab, Bethesda. PI : R.Hemley)
- High Pressure Fiber Bragg Grating Dilatometry in pulsed field : next April at HZDR-Dresden
- High pressure Ultrasound (HZDR, C.Ruegg, Sergei Zherlitsyn) next April
- Collaboration at LNCMI Toulouse, pulsed field facility : next March and July
- High Pressure X Ray Diffraction at ESRF on dU : next July
- **>100 kbar in small plastic DACs opens new possibilities :**

  - Cuprates : collaboration with Louis Taillefer's group, Sherbrooke
    study YBCO pseudo-gap region by modulating doping on one sample
  - $SmB_6$ : collaboration with P.Goddard ( University of Warwick), and J.Cooley (LANL).
    Look for metallic state.
  - Fisher's group (Stanford) : rare-earth tellurides
  - Analytis' group (Berkeley) : Iridates, Pnictides




*Nature* **447**, 565-568(31 May 2007)

# Diversity and education

Postdoc

- Audrey Grockowiak **applying for research faculty and assistant professor positions**

Undergraduates

- Ameila Estry Materials U alloys (worked with J. Cooley (LANL) summer 2014, REU with P.Canfield summer 2016)
- Lauren Riner Cryogenics
- Jessey. Krehl Electronics
- Fernando de Torres Electronics
- Anna Yannakopoulos Band structure calculations
- Linsey Rodenbach Materials-U and Cr

High School

- Allan Williams Band structure calculations
- Elisabeth Wackes Cryogenics

Past undergraduate and graduate students :

- Ryan Stillwell LLNL postdoc in Jason Jeffries group, spring 2014, Q clearance, in the process of being converted ?
- Michael Woods U growth & preparation UVC Nuclear Engineering GS
- Ashley Bernheisel Electronics Boeing & Electrical Engineering GS
- Michael Bernstein Materials-U preparation ASU Physics GS

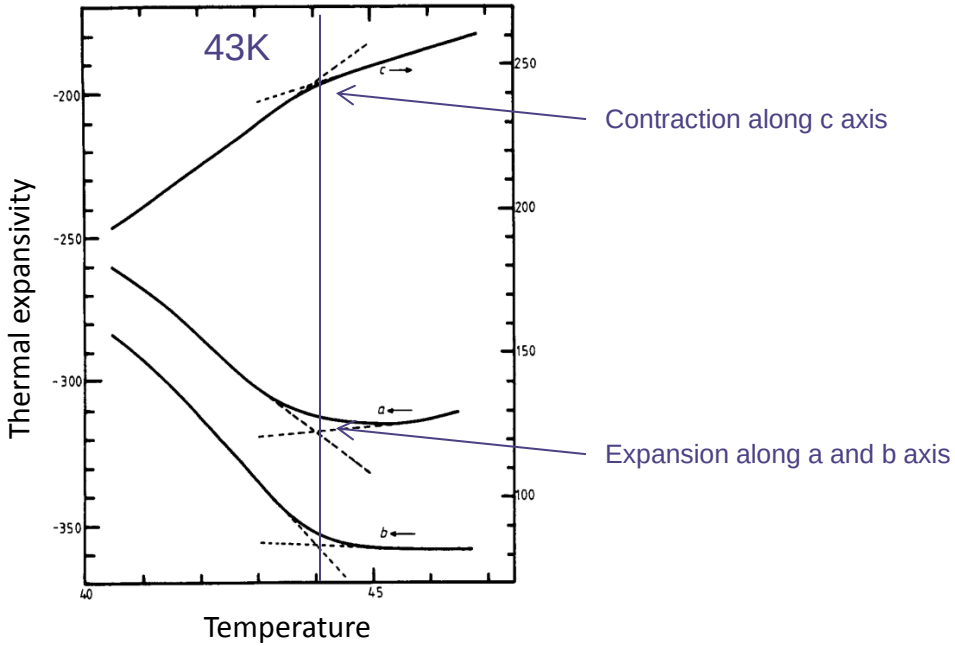
# Research motivation

- Physics of actinides, 4f and 5f electron systems with complex phenomena : charge density waves, volume collapse, superconductivity, quantum critical point…

- Apply external parameters (temperature, magnetic field, pressure) coupled with experimental techniques (TDO, dilatometry…) to understand fundamental properties of these materials.

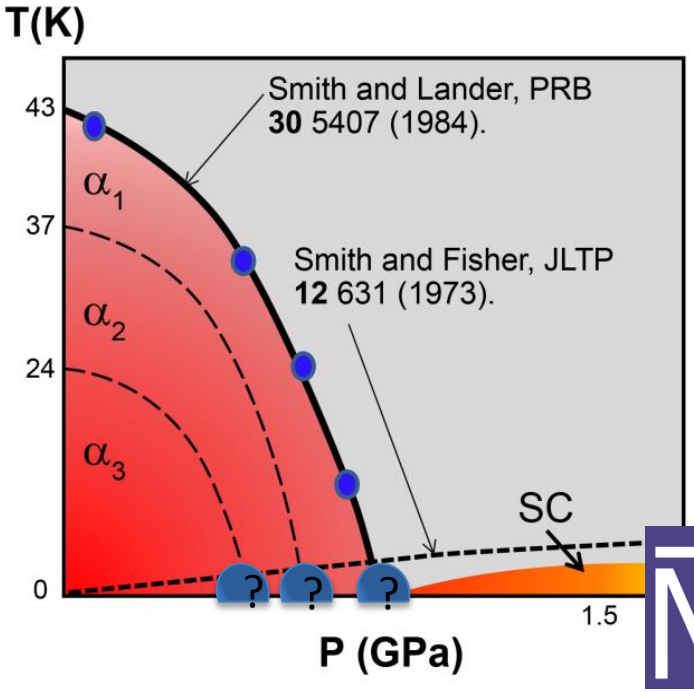
Uranium 238 : touchable actinide to advance studies on Ce and Pu

3 CDW : 43K, 37K, 23K associated with structural change




Tindall et al., J. Phys. F: Met. Phys. **14** (1984) 113 1-1 136

Superconductivity at high pressure coexisting with CDW



# Introduction to dilatometry

- Measures changes in length of a material under external parameters
  - Temperature => thermal expansion
  - Magnetic field => magnetostriction
  - Pressure => compressibility

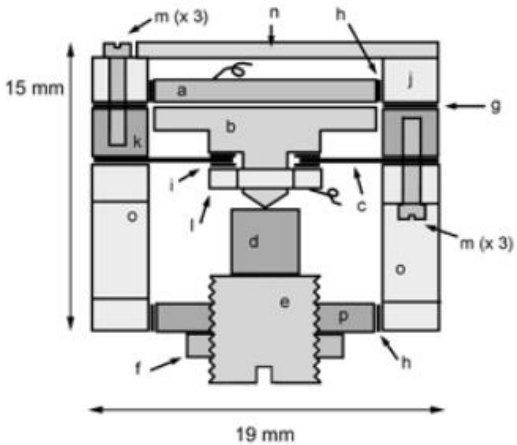
- Thermodynamic quantity sensitive to phase transitions :

=> Another probe that provides understanding of fundamental properties of materials. Can detect, for example, superconducting transitions, and quantum oscillations.

# Introduction to dilatometry

- Dilatometry techniques

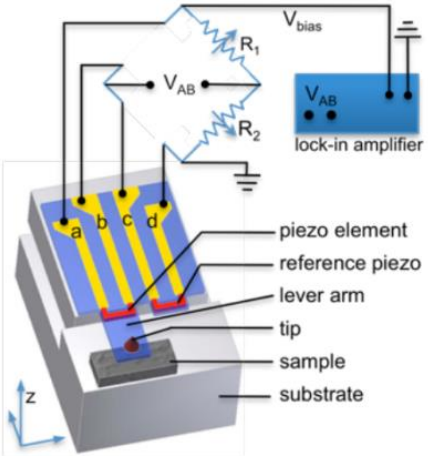
  - Capacitance dilatometry




Sample attached to mobile capacitance plate
Variation of capacitance => sample expansion
Resolution : 0.1A at low T

G. M. Schmiedeshoff et al., RSI. 77, 123907 (2006)

  - Piezo Cantilever



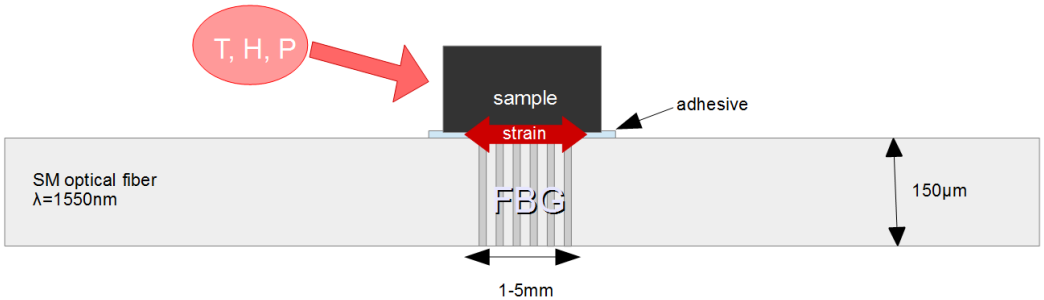

Atomic force microscope tip on sample
Resistance change in piezo element =>sample expansion
Resolution : 0.1-1A at low T

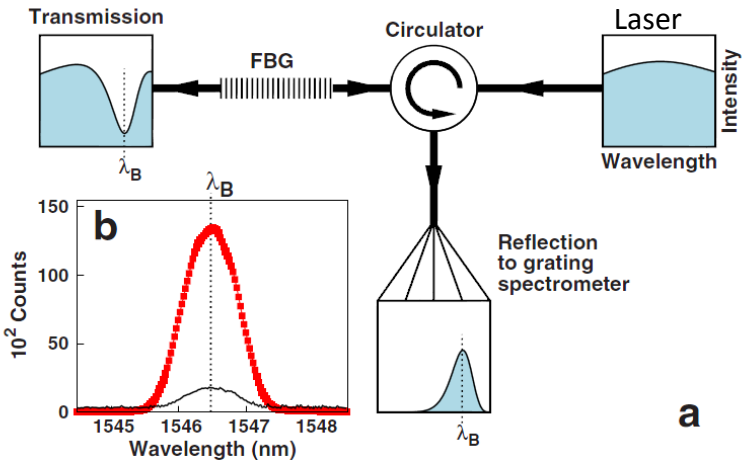
Park, J.-H., et al., Rev. Sci. Instrum. 80, 116101 (2009)

=> Cannot be coupled to pressure cells !

# High pressure technique

- Fiber Bragg Grating dilatometry




R.Daou et al., Rev. Sci. Instrum. 81, 033909 (2010)

Periodic n-index modulation of the fiber : grating

Refracts one Bragg wavelength $\lambda_B$

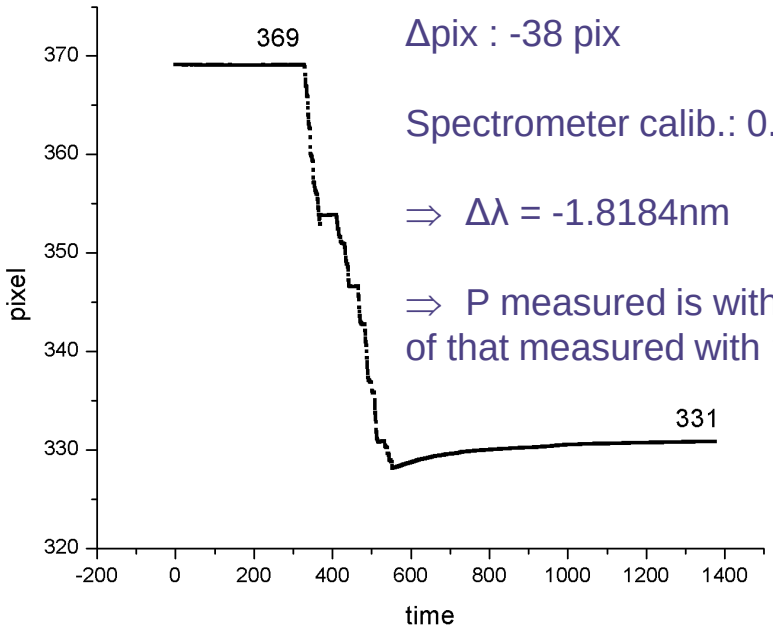
**Sample expansion along fiber => strain fiber => Shift in $\lambda_B$**

Improved resolution : $\Delta L/L \approx 10E-7$!!

# High pressure technique

- Coupling FBG with a pressure cell : Guertin Piston Cylinder

FBG can also be used to measure pressure




Δpix : -38 pix

Spectrometer calib.: 0.0673nm/pix

⇒ Δλ = -1.8184nm

⇒ P measured is within 0.1% of that measured with ruby

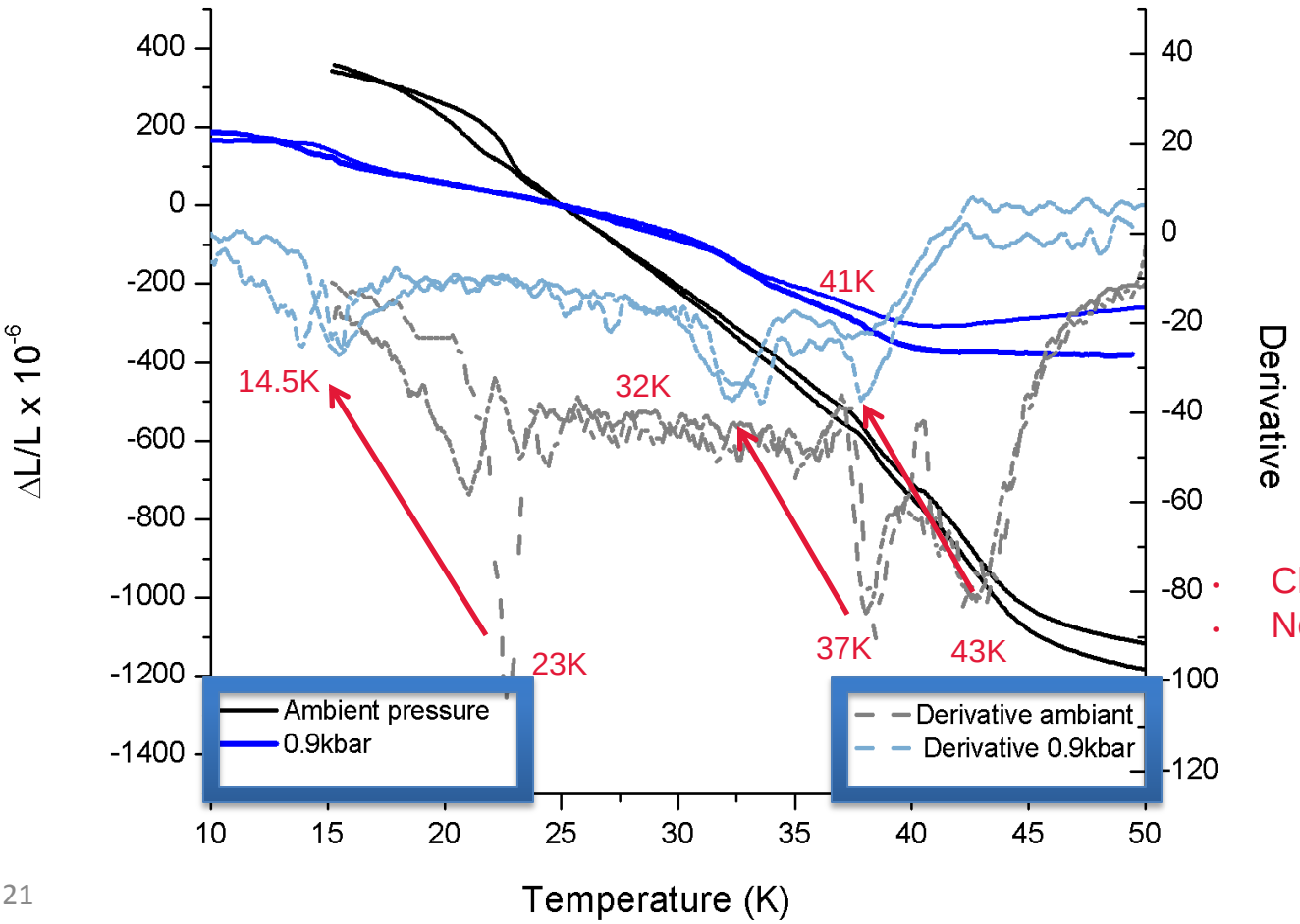


A. Grockowiak

# Preliminary results : U-238

- Dilatometry under pressure

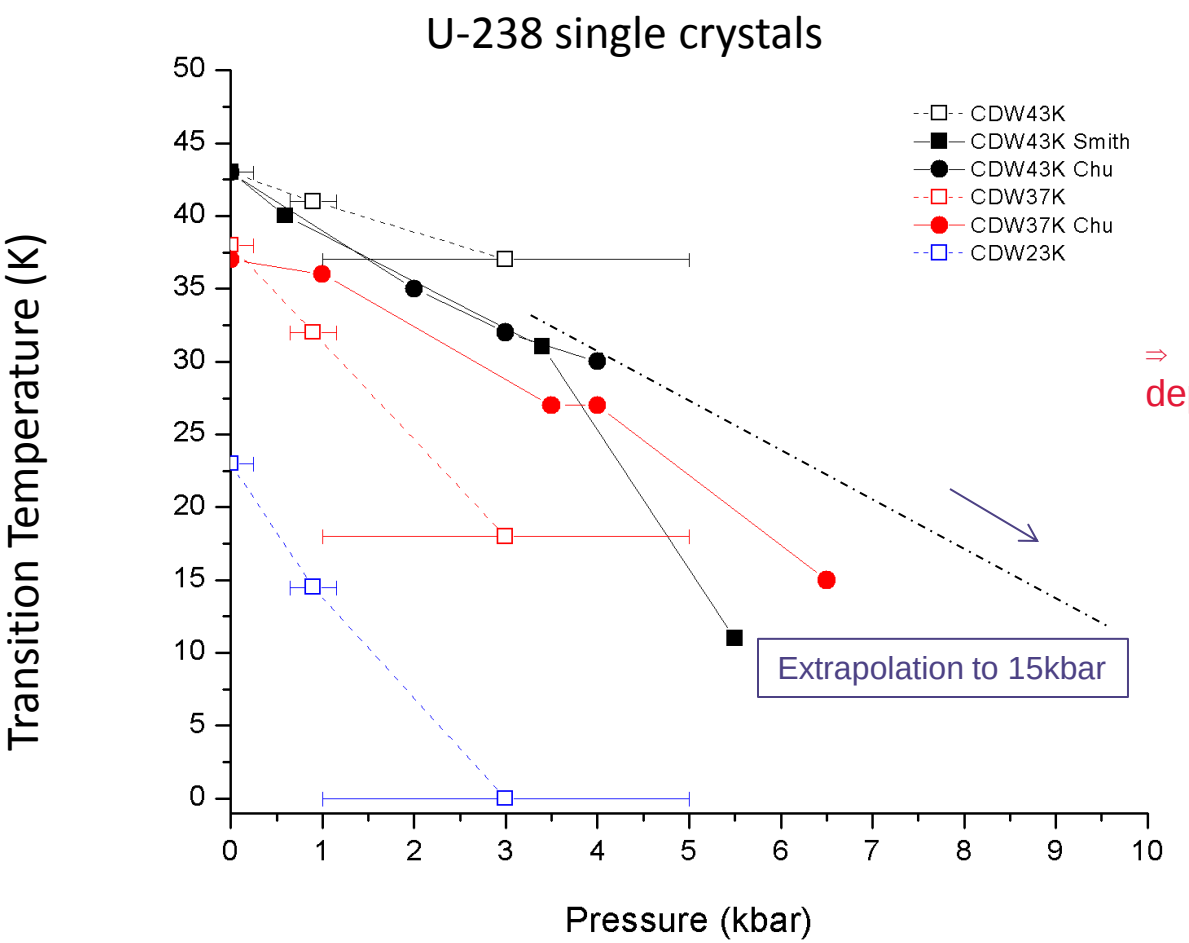
Samples : -J.Willit (ANL)
-M.Woods (MagLab)




Clear shift of the 3 CDW to lower T
No magnetic field dependence

# Preliminary results : U-238

- Dilatometry under pressure

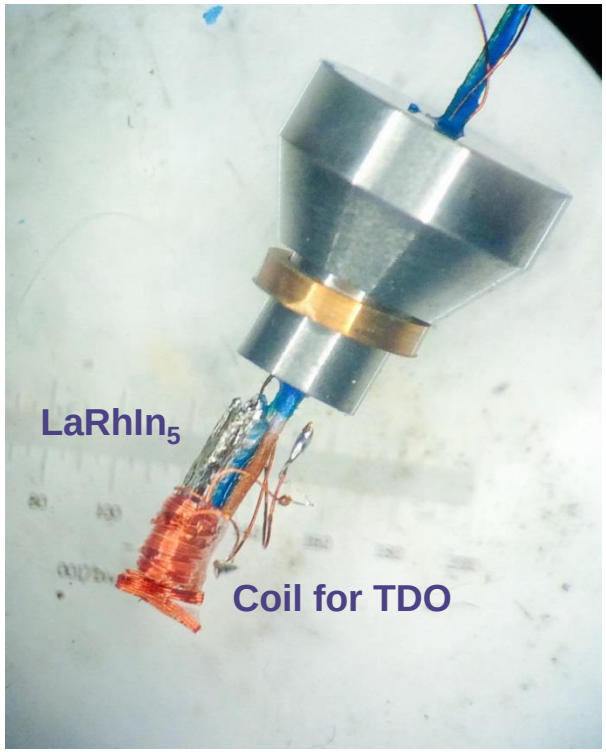



Smith : neutron scattering (PRB 30, 10 (1984)
Chu : ac calorimetry (Physics Letter 46A 1 (1973))
Open symbols-this study

⇒ Clear discrepancies in the pressure dependence of the transition temperatures !

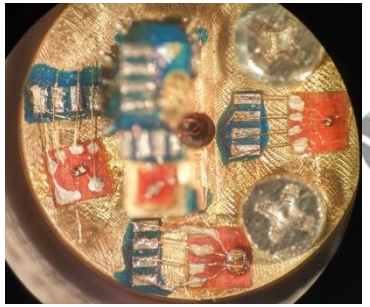
# Work in progress

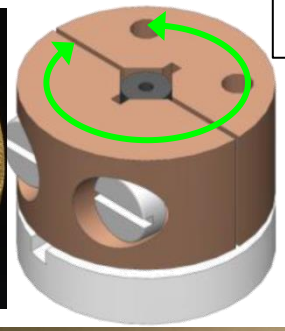
- FBG high pressure dilatometry achieved
- Usable in pulsed and DC magnetic fields, and low T
- Complement current high P techniques, to help investigate complex phases of actinides
- Applicable to other material



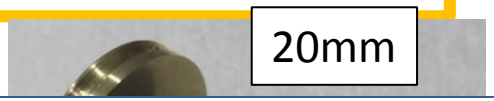
# Instrumentation – Two-axis rotation

- Using the Attocube rotation systems to add a $\phi$ component to rotation
- Labview software has been used to make a user interface for aligning samples and automating measurements (work in progress).

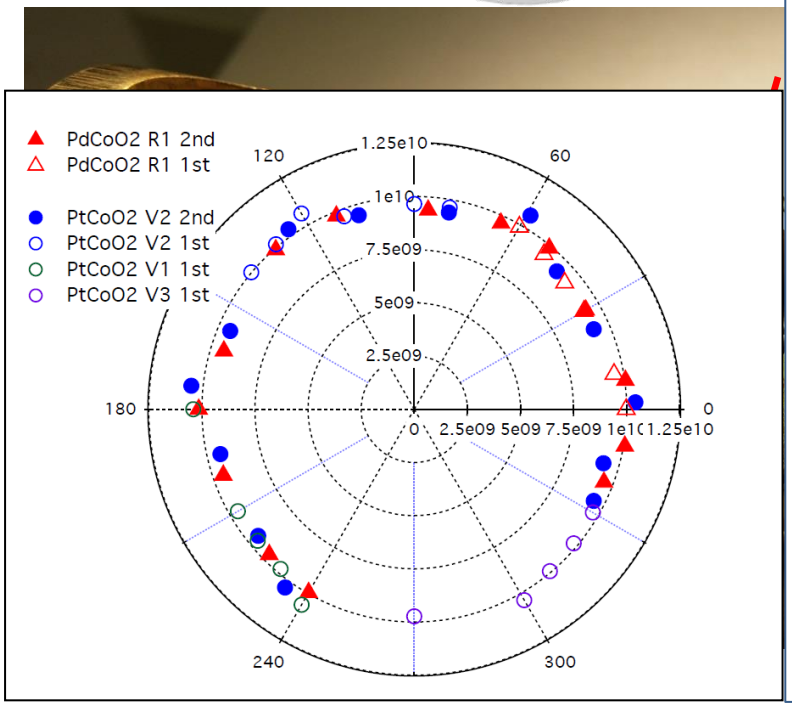


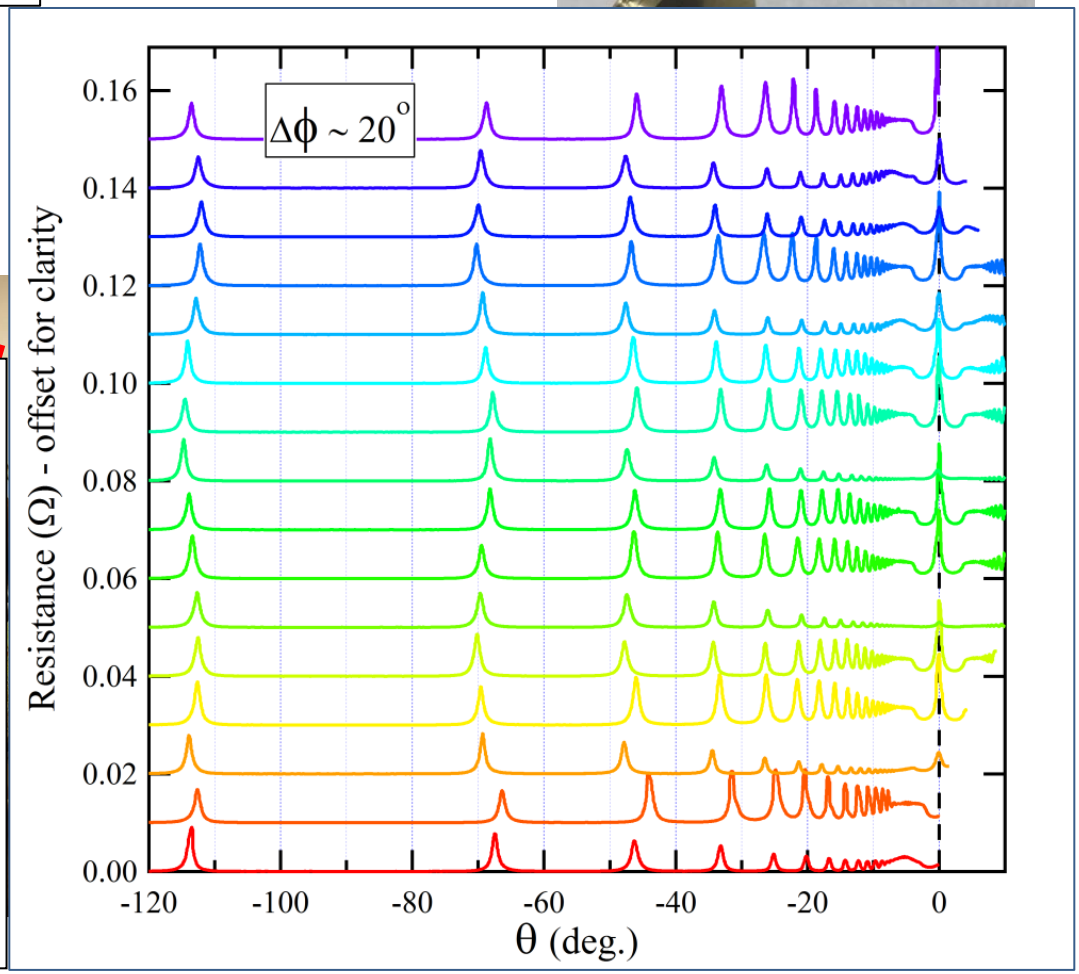


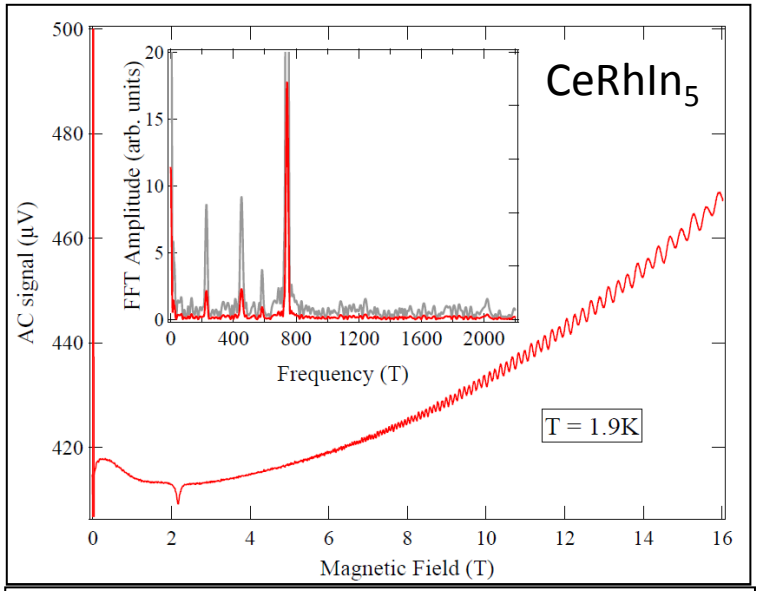
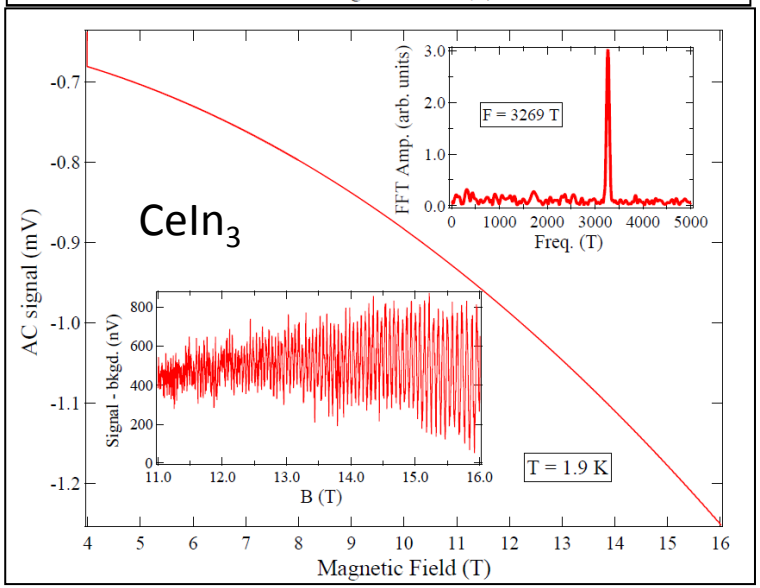
10mm



20mm



# Instrumentation - AC susceptibility

- NHMFL users (i.e. H. Yuan - $CeRhIn_5$) have asked for this capability under high pressure.
- Initial results are shown for the test cases of $CeIn_3$ and $CeRhIn_5$ at ambient pressure.