X-ray scattering at high pressures and low temperatures: Squeezing cool electrons

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TECHNISCHE UNIVERSITÄT DRESDEN

People and funding

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G. Garbarino, M. Hanfland,
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ESRF

M. v. Zimmermann, S. Francoual,
J. Sears

DESY

and many more….

SFB 1143

GRK 1621

ct.qmat
Complexity and Topology in Quantum Matter

Outline

- Why pressure and low temperatures?!!
- Typical experimental setups
- Scientific examples
  - Charge density waves and superconductivity
  - Pressure-driven covalency and magnetism
  - Strong covalency: Towards room temperature SC

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The theory of “everything”

$$\hat{H} = -\frac{\hbar^2}{2}\sum_i \frac{\nabla^2_{\vec{R}_i}}{M_i} - \frac{\hbar^2}{2}\sum_i \frac{\nabla^2_{\vec{r}_i}}{m_e} - \frac{1}{4\pi\epsilon_0}\sum_{i,j}\frac{e^2\, Z_i}{|\vec{R}_i - \vec{r}_j|} + \frac{1}{8\pi\epsilon_0}\sum_{i\neq j}\frac{e^2}{|\vec{r}_i - \vec{r}_j|} + \frac{1}{8\pi\epsilon_0}\sum_{i\neq j}\frac{e^2\, Z_i Z_j}{|\vec{R}_i - \vec{R}_j|}$$

The fundamental rules can be easily written down, but the resulting behavior is very hard to predict
(if not impossible)

➡ **Emergent properties**
Unconventional Superconductivity,
self-organization of electrons, spin liquids…

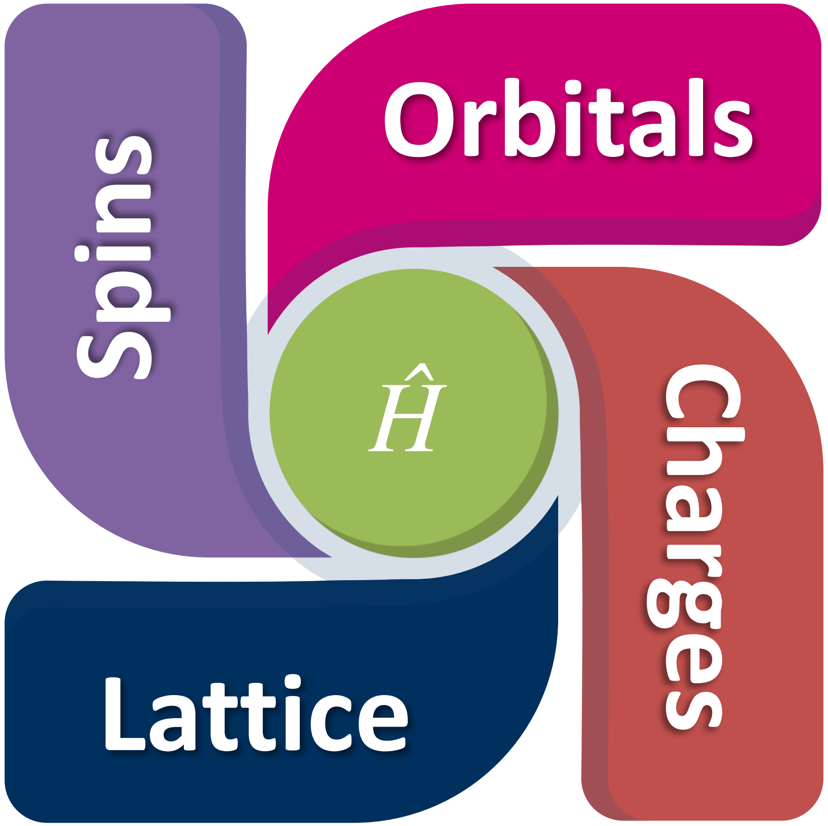
Coupled degrees of freedom

SPINS

Crystal field splitting
Charge fluctuations
Orbital occupation and exchange

ORBITALS

Local lattice distortions
Overlap integrals and charge transport
Magnetic coupling (GKA-rules)



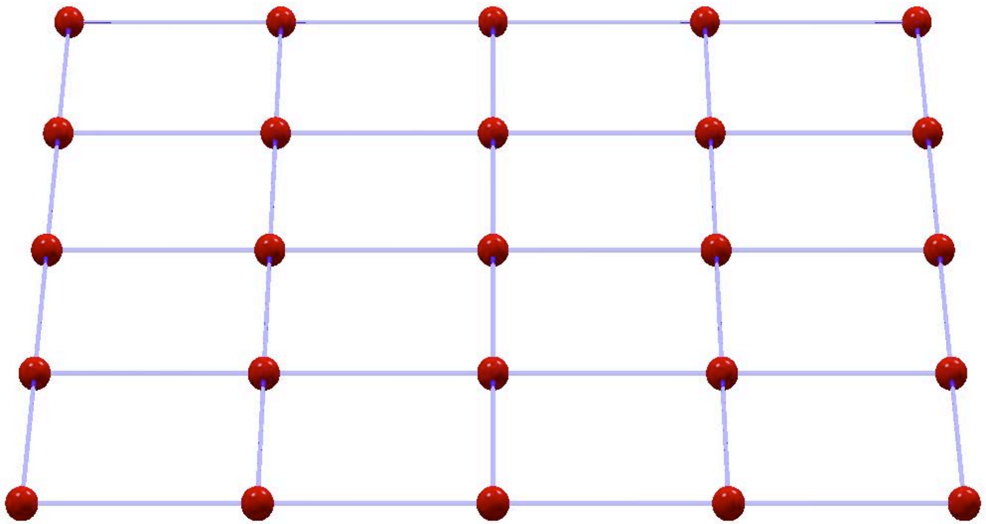
LATTICE

Polaronic charge carriers
Crystal field splitting and orbitals
Magnetic coupling (bond distances angle)

CHARGES

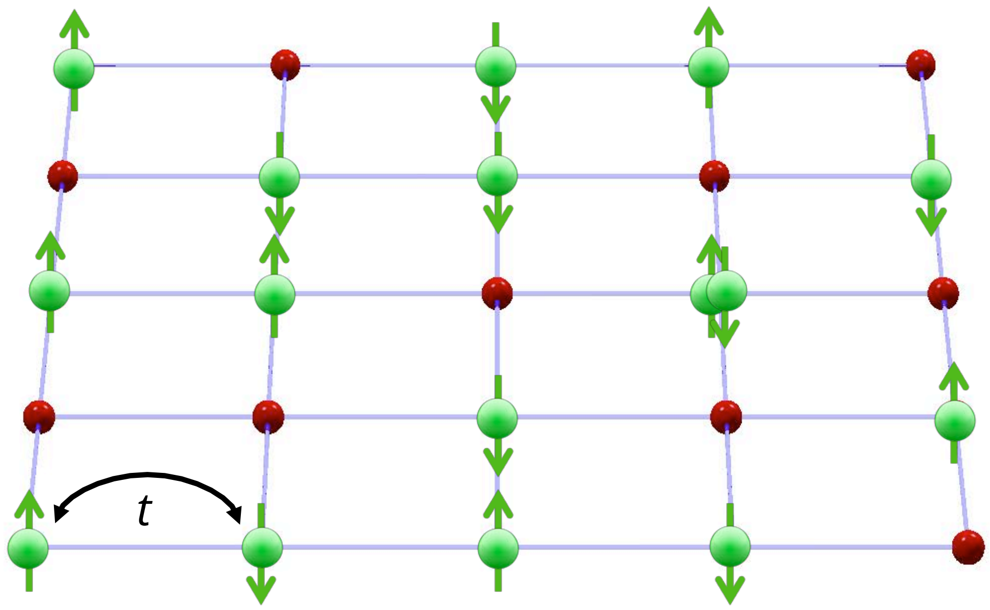
Electron-phonon coupling
Spin fluctuations
Orbital occupation

Independent particles



Independent particles

A simple model 2 states per site ↑↓

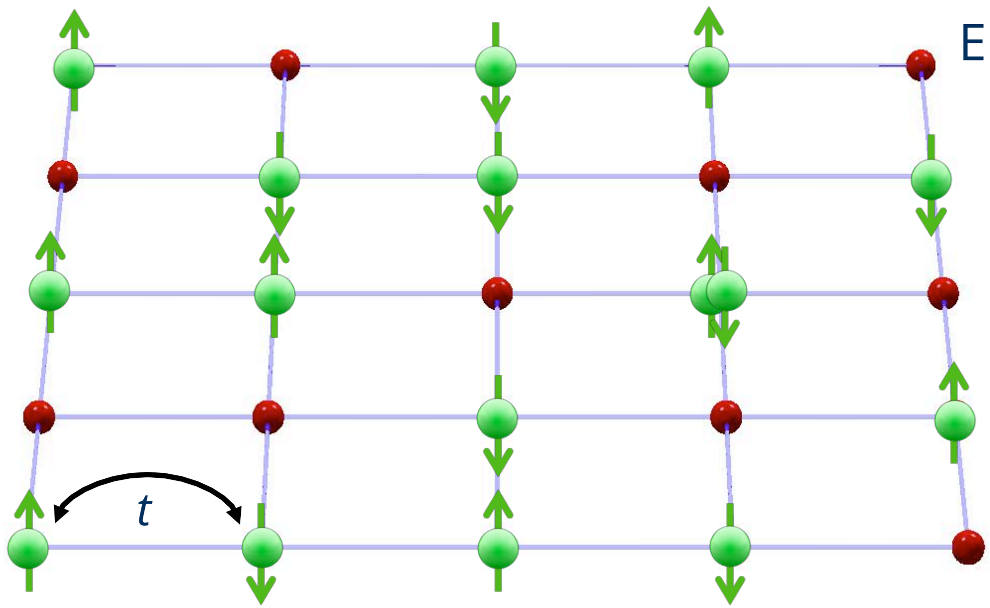


- t : kinetic energy
- Hopping only restricted by Pauli
- Otherwise **no** interactions

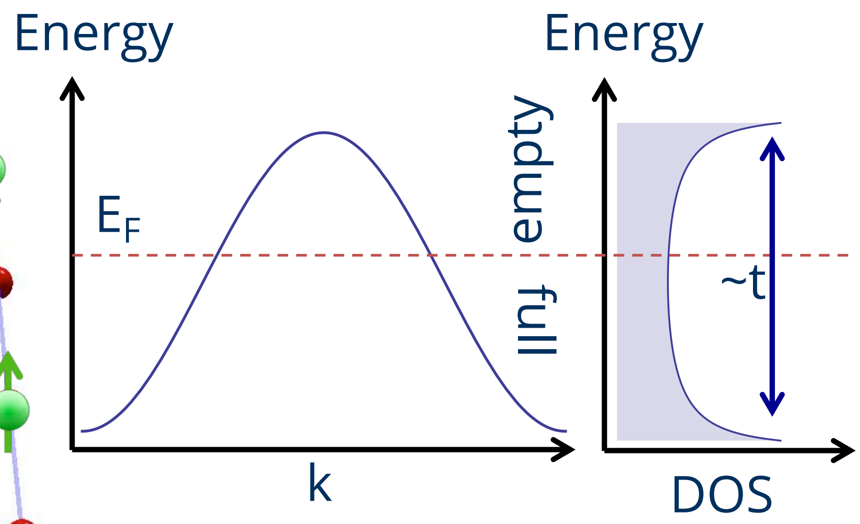
$$\mathcal{H} = -t \sum_{\langle ij \rangle, \sigma} (c^{\dagger}_{i\sigma} c_{j\sigma} + \text{h.c.})$$

Independent particles

A simple model 2 states per site ↑↓



Quasiparticle band structure



$$\mathcal{H} = -t \sum_{\langle ij \rangle, \sigma} (c^{\dagger}_{i\sigma} c_{j\sigma} + \mathrm{h.c.})$$

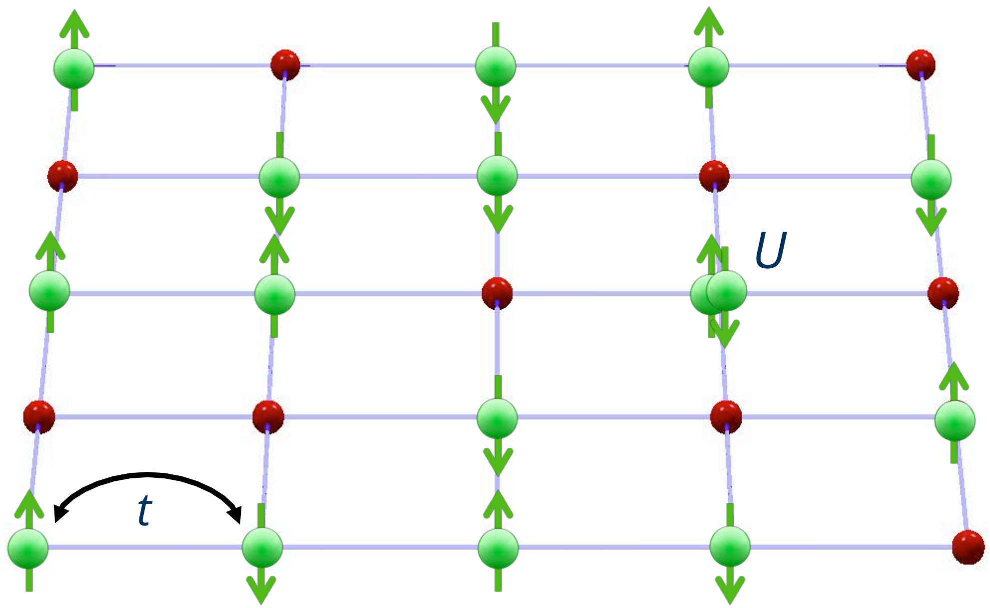
Band theory:

all states occupied: **insulator**

otherwise: **metal**

Correlated electrons

Mott system for t<U

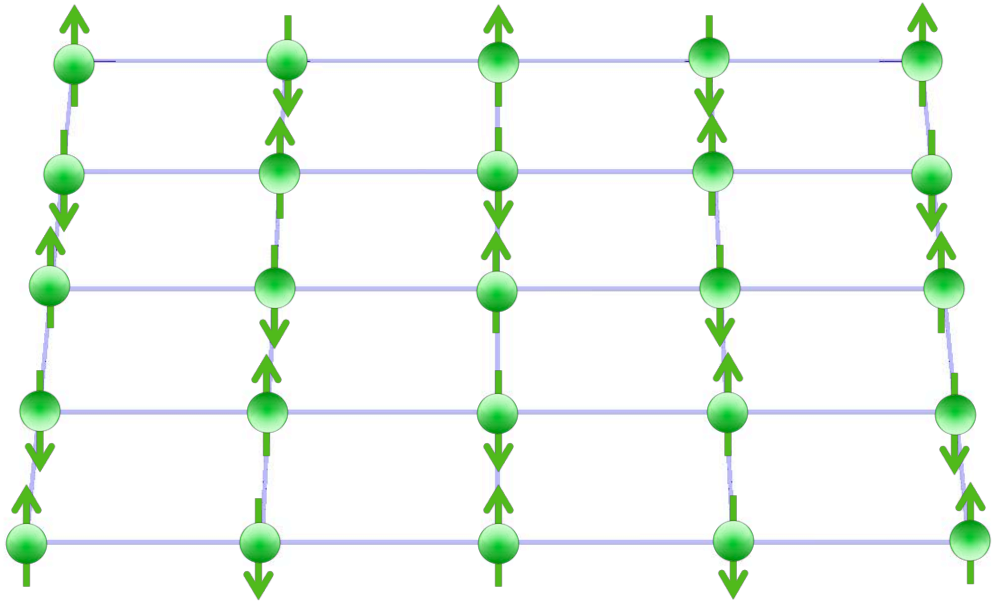


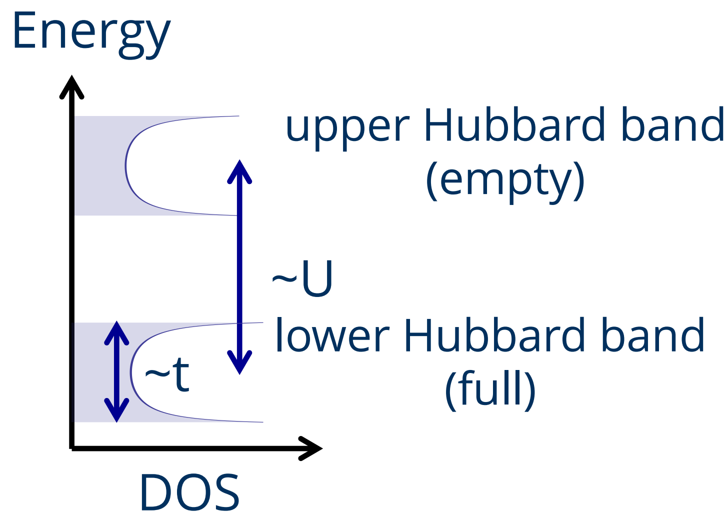
- *t* : kinetic energy
- ***U*: Electron-electron interaction**
- *U* creates additional correlations
- ***U* competes with *t***

$$\mathcal{H} = -t \sum_{\langle ij \rangle, \sigma} (c^\dagger_{i\sigma} c_{j\sigma} + \text{h.c.}) + U \sum_i n_{i\uparrow} n_{i\downarrow}$$

Correlated electrons

Electronic order at half filling





$$\mathcal{H} = -t \sum_{\langle ij \rangle, \sigma} (c_{i\sigma}^{\dagger} c_{j\sigma} + \text{h.c.}) + U \sum_{i} n_{i\uparrow} n_{i\downarrow}$$

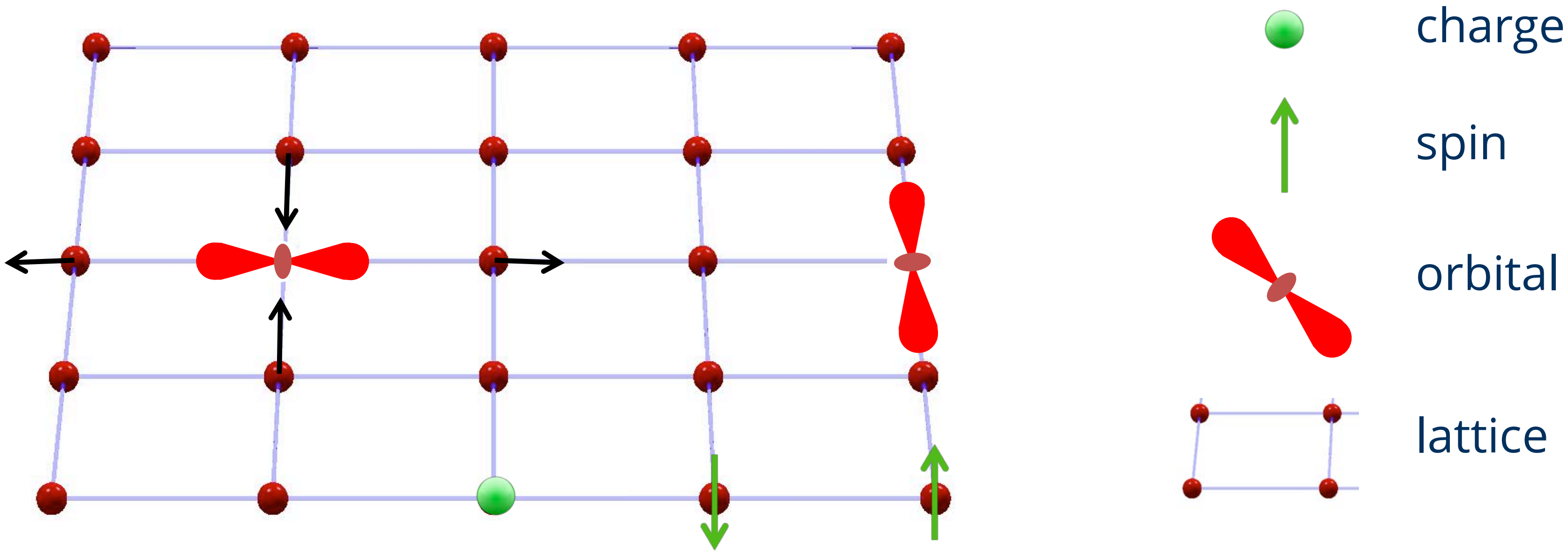
Half filling

Insulating state!

AFM ground state!

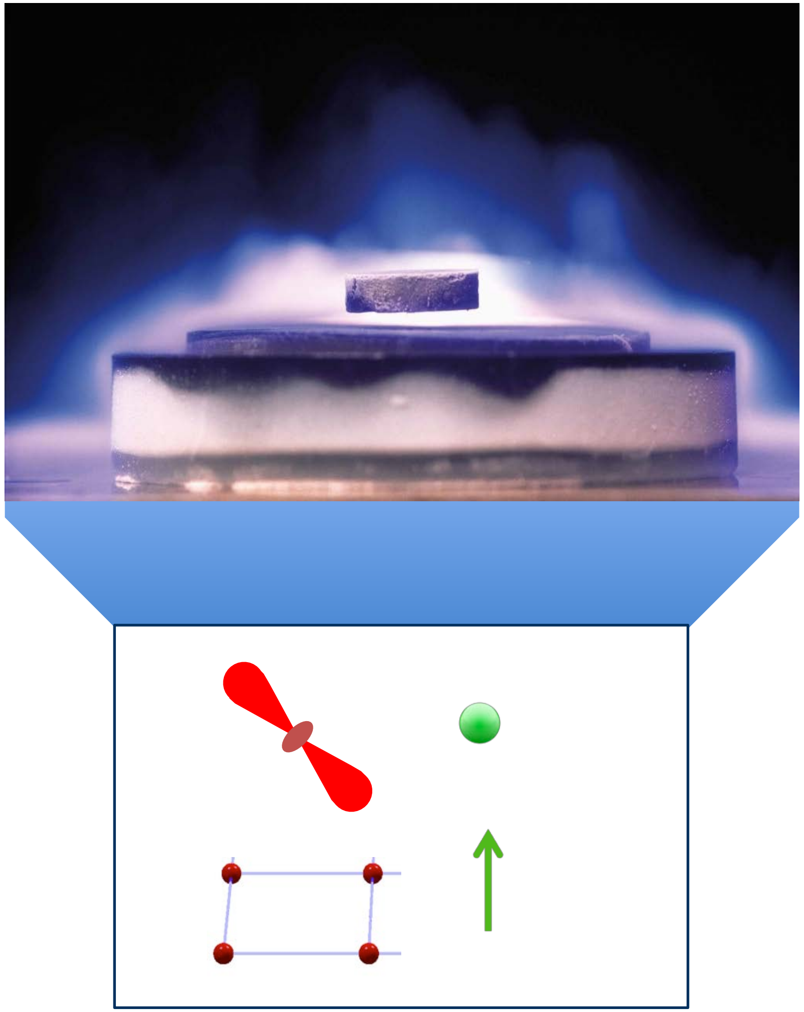
Complex electron systems

Various interacting degrees of freedom

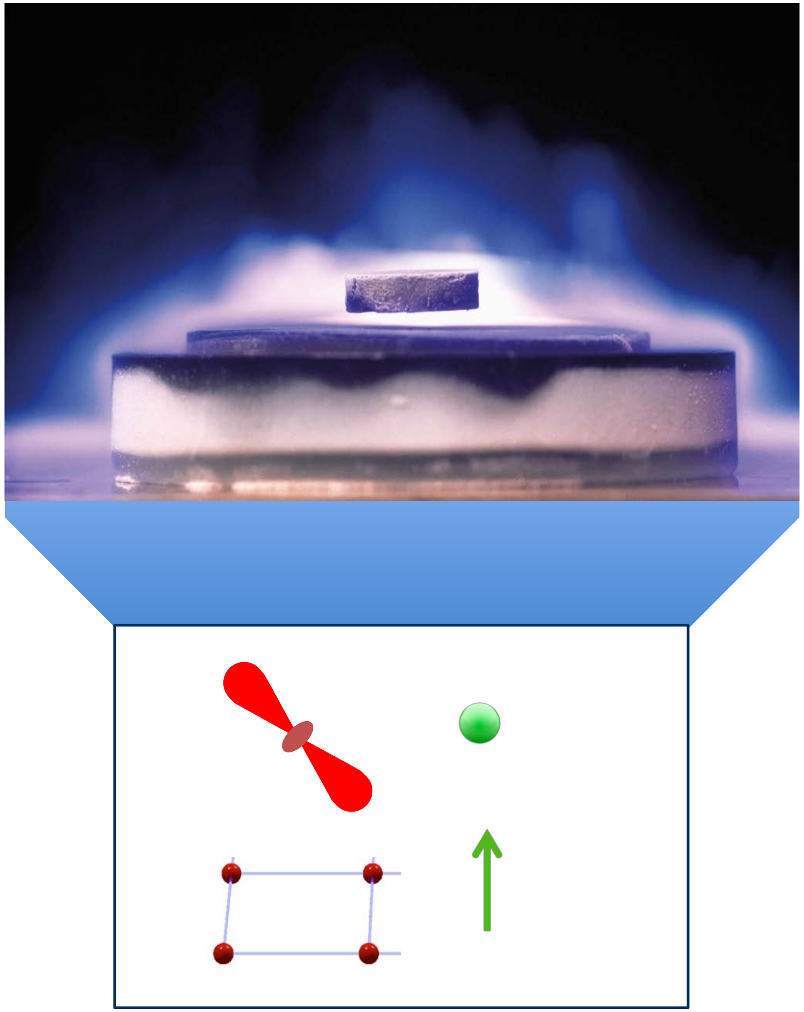


- Breakdown of independent particle picture
- Rich physics: HTSC, CMR, multiferroic order, phase competition and coexistence…(technological potential)

More is different



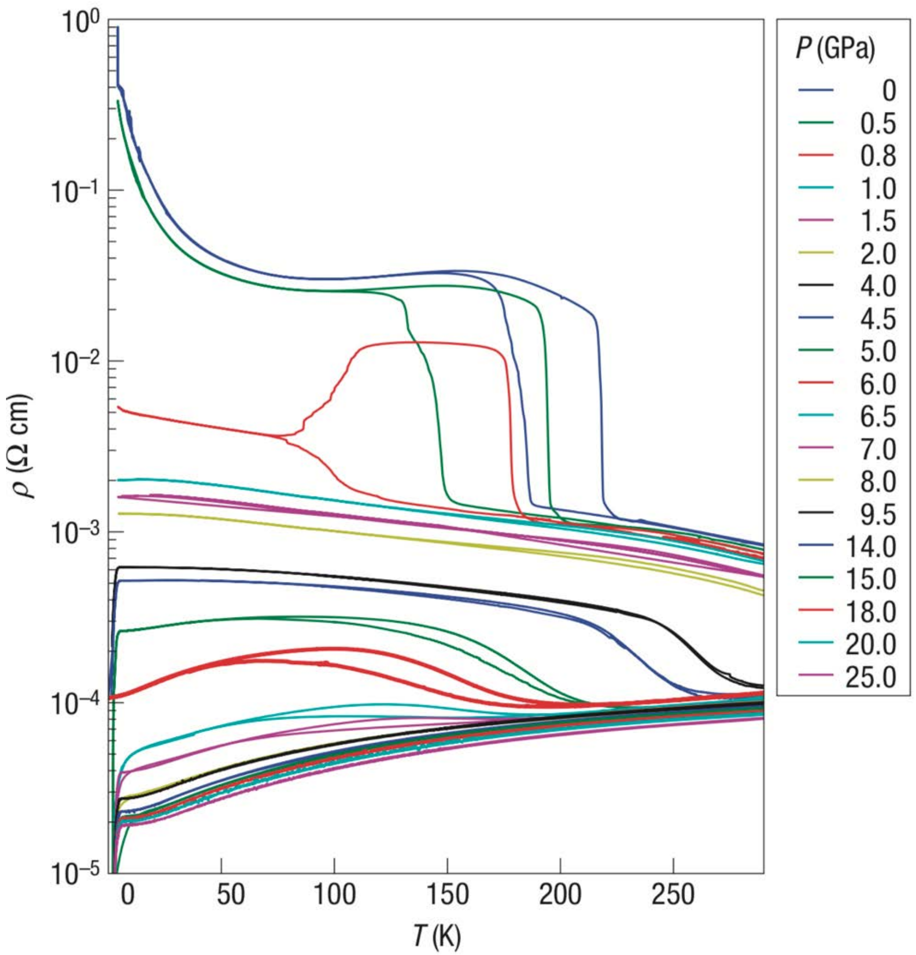
More is different



Collective quantum phases in condensed matter:

- Superconductivity, electronic order, spin liquids …
- Formidable theoretical challenge
  → Experiments!
- Studies of ground state
  → low temperature!
- Controlled manipulation lattice without external symmetry breaking
  → hydrostatic pressure!

Examples: Pressure-induced SC




ARTICLES

From Mott state to superconductivity in $1T\text{-}TaS_2$

B. SIPOS[1]*, A. F. KUSMARTSEVA[1]*, A. AKRAP[1], H. BERGER[1], L. FORRÓ[1] AND E. TUTIŠ[2]
[1]Ecole Polytechnique Fédérale de Lausanne, IPMC, CH-1015 Lausanne, Switzerland
[2]Institute of Physics, Bijenička c. 46, Zagreb, Croatia
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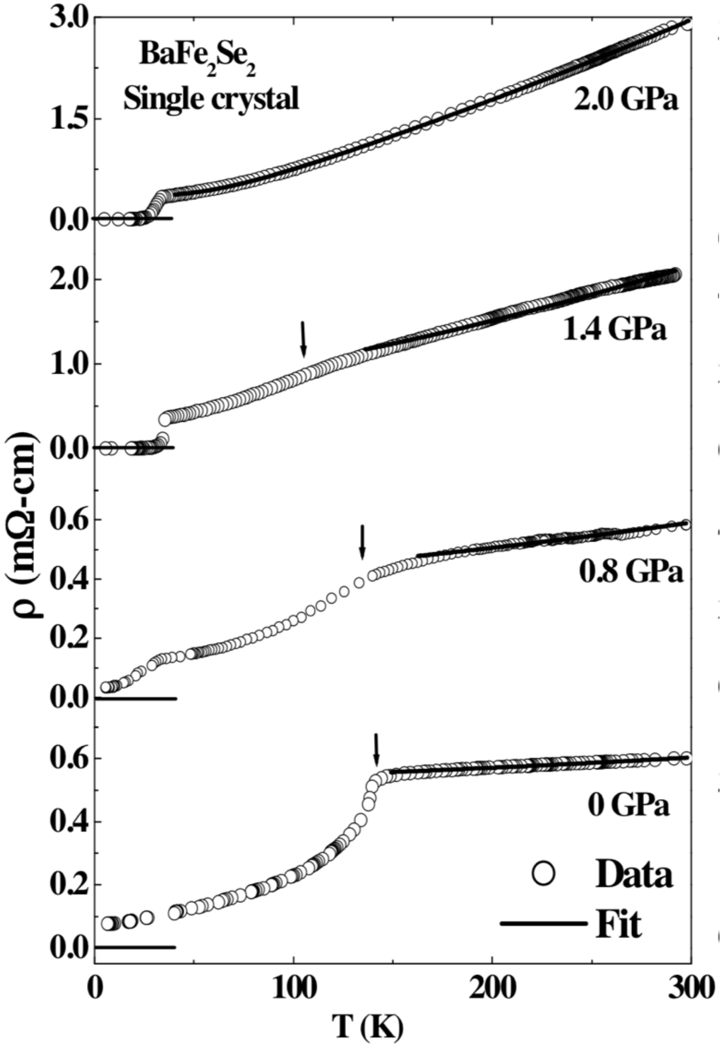
Published online: 9 November 2008; doi:10.1038/nmat2318

- What happens at the atomic level?
- Structural changes?
- Electronic order in real space?

Examples: Pressure-induced SC



epl A LETTERS JOURNAL EXPLORING THE FRONTIERS OF PHYSICS

July 2009

EPL, **87** (2009) 17004 www.epljournal.org
doi: 10.1209/0295-5075/87/17004

Pressure-induced superconductivity in $BaFe_2As_2$ single crystal

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Materials Science Group, Indira Gandhi Centre for Atomic Research - Kalpakkam, 603102, Tamilnadu, India

- What happens at the atomic level?
- Structural changes?
- Electronic order in real space?

Examples: Pressure-induced SC

npj | Quantum Materials

www.nature.com/npjquantmats

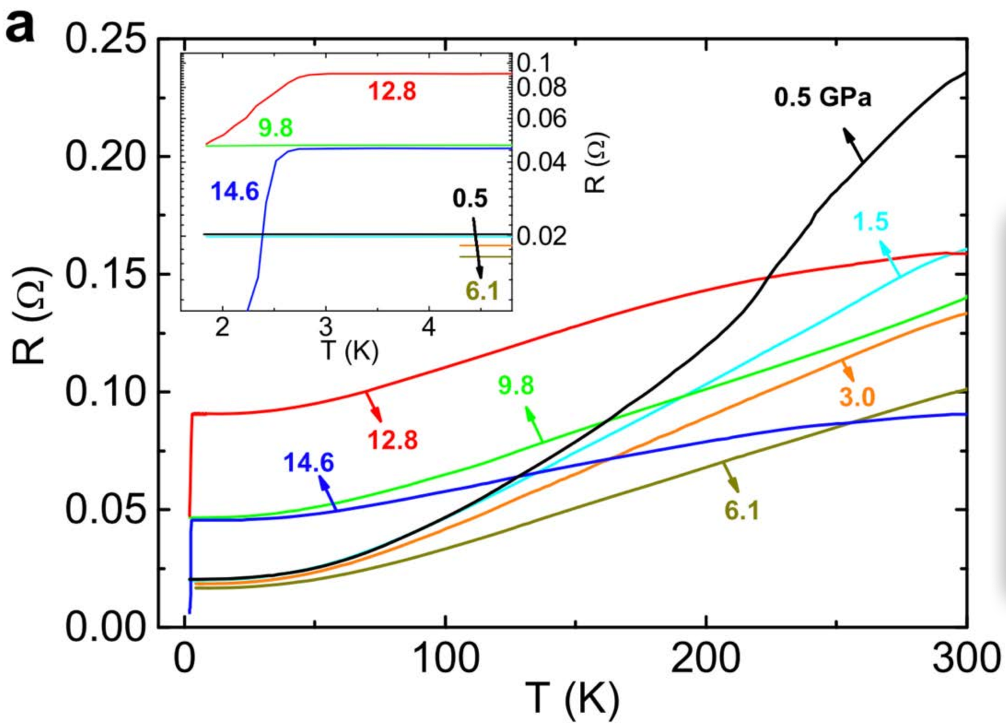
ARTICLE

Pressure- emimetal

$NbAs_2$

X-ray scattering experiments can help!

Yupeng Li[1], Chao An[2], Chenqiang Hua[1], Xuliang Chen[2], Yonghui Zhou[2], Ying Zhou[2], Ranran Zhang[2], Changyong Park[3], Zhen Wang[1,4], Yunhao Lu[4], Yi Zheng[1,5], Zhaorong Yang[2,5] and Zhu-An Xu[1,4,5]



- What happens at the atomic level?
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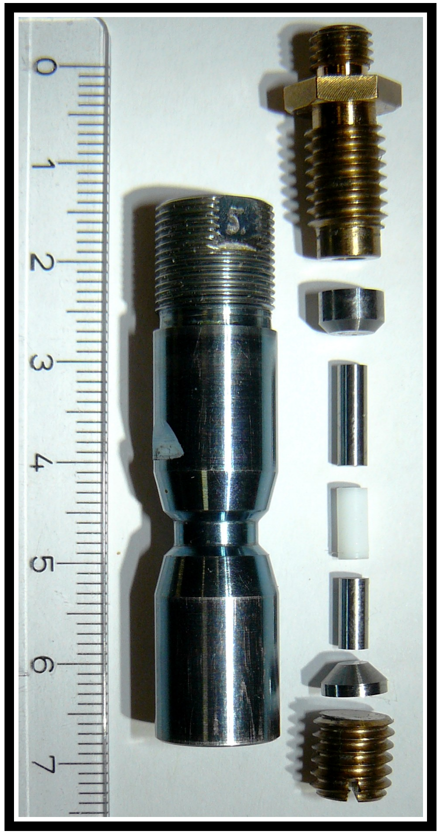
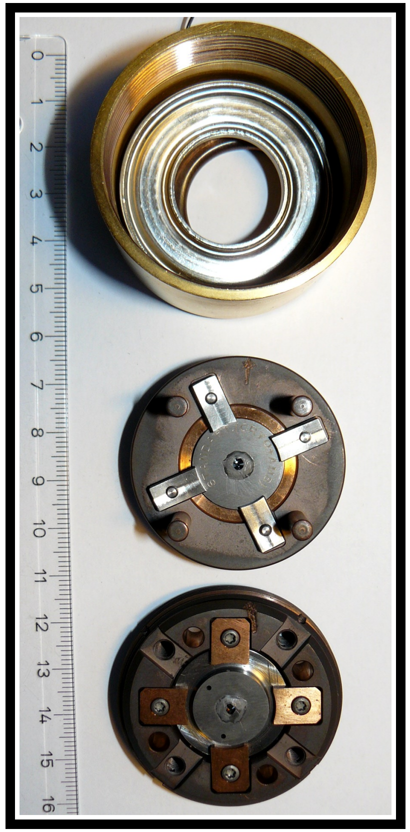
Pressure cells



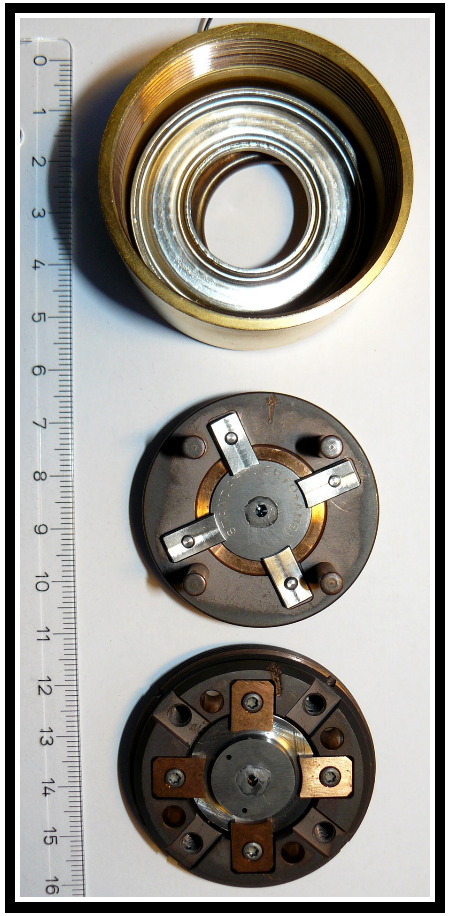
Clamp type cell
(up to 1.5 GPa)

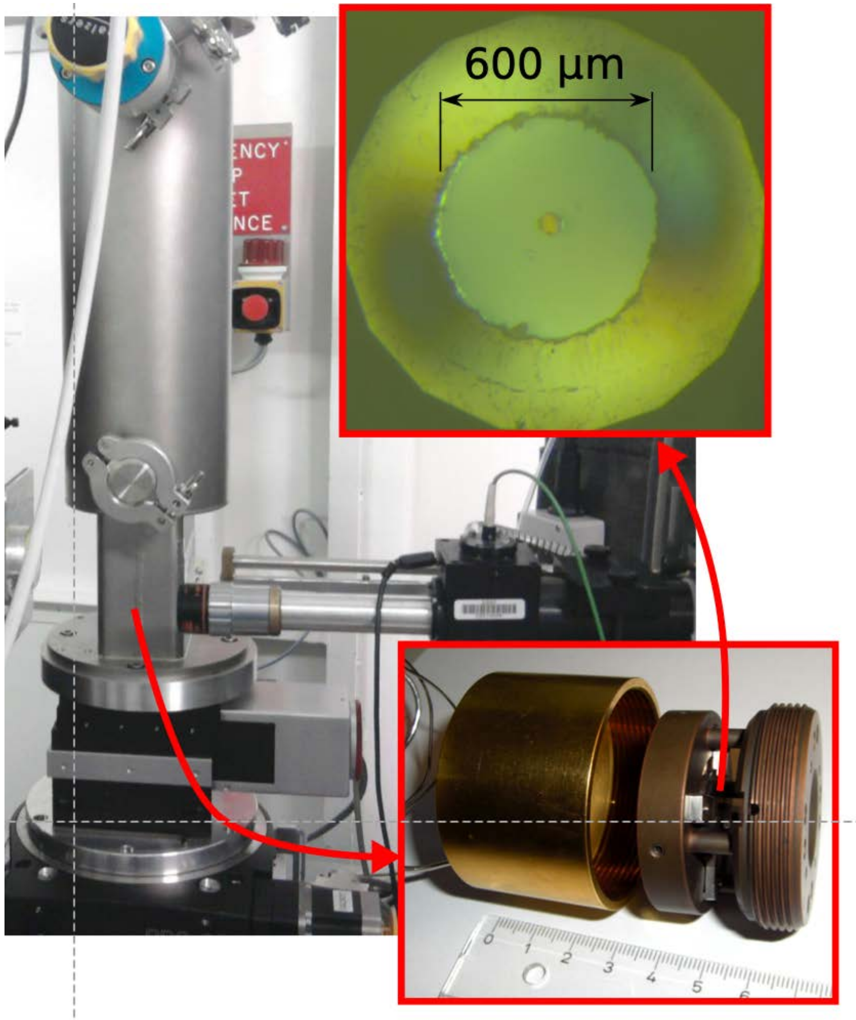
SiC anvil cell
(up to 6 GPa)



diamond anvil cell
(up to 200 GPa)

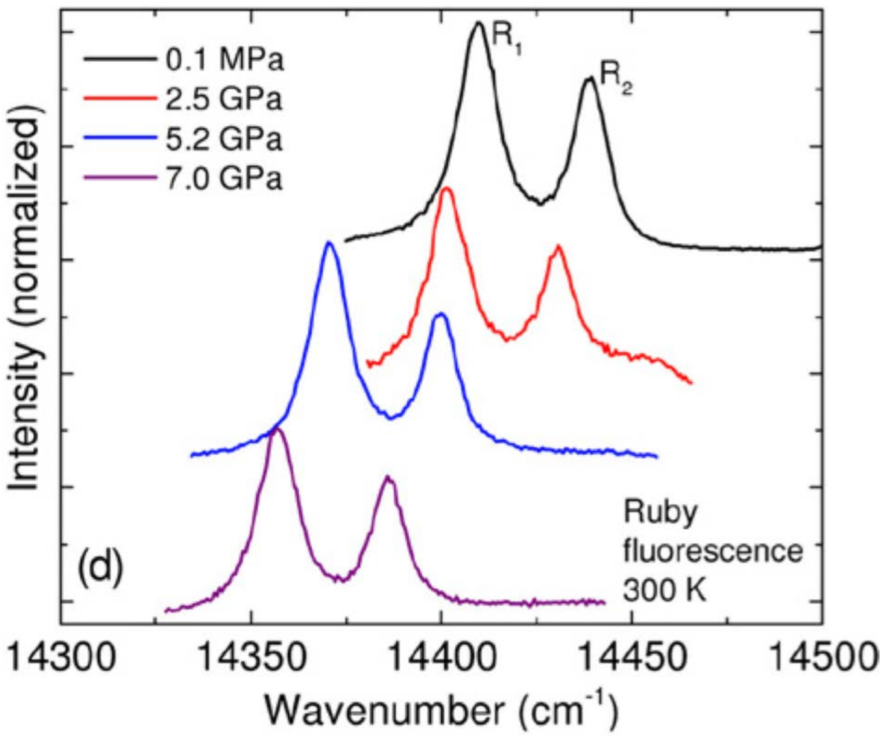
Membrane-driven DAC



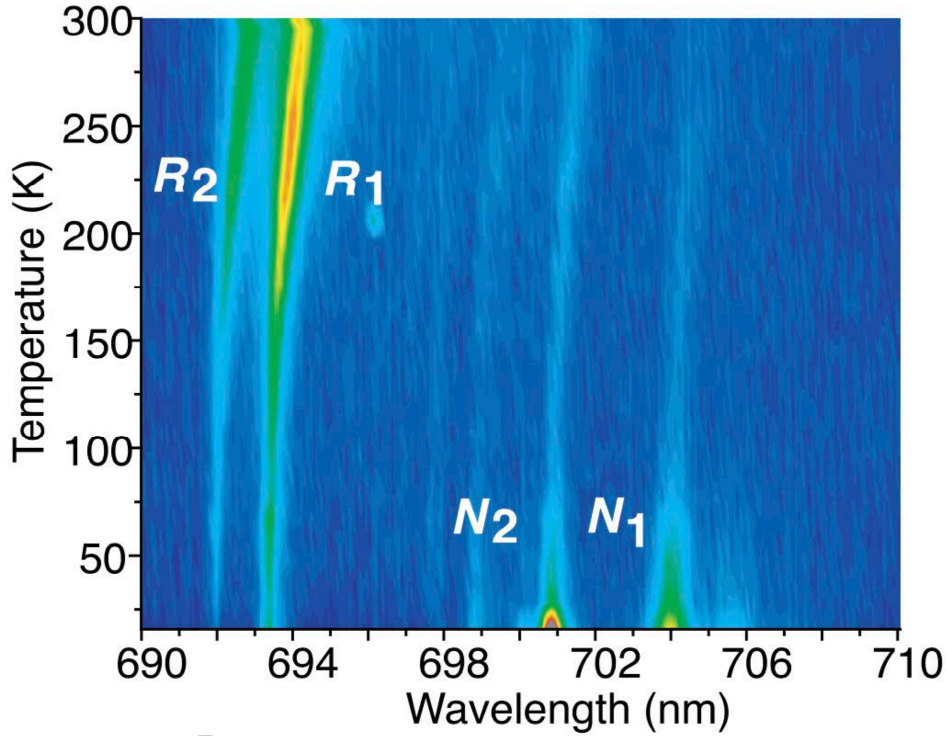


Pressure monitoring

M. Knaapila *et al.*, Rev. Prog. Phys. 2016

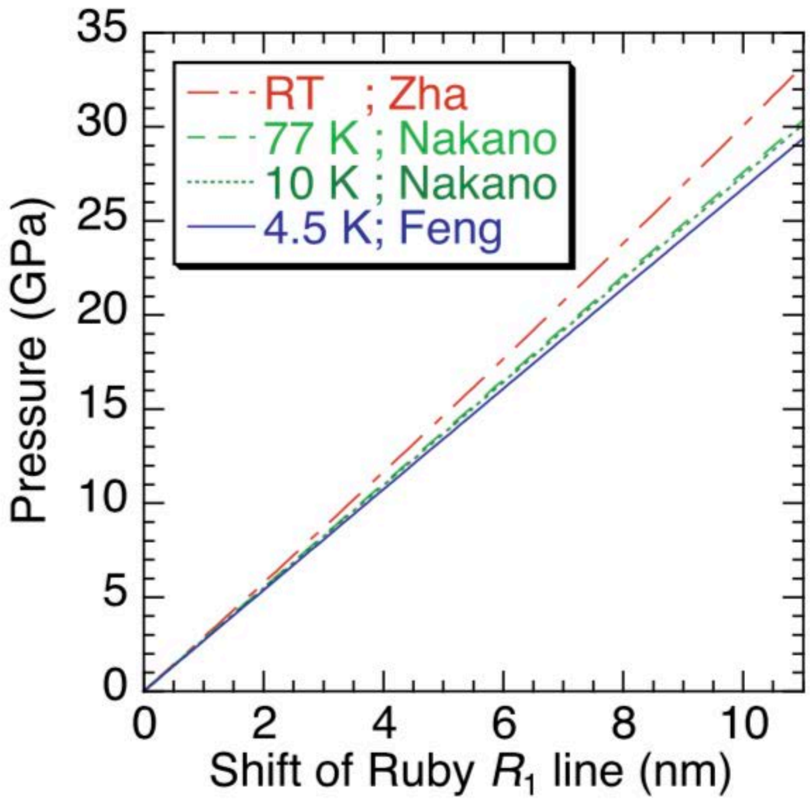


H. Yamaoka *et al.*, J. Appl. Phys. 2012

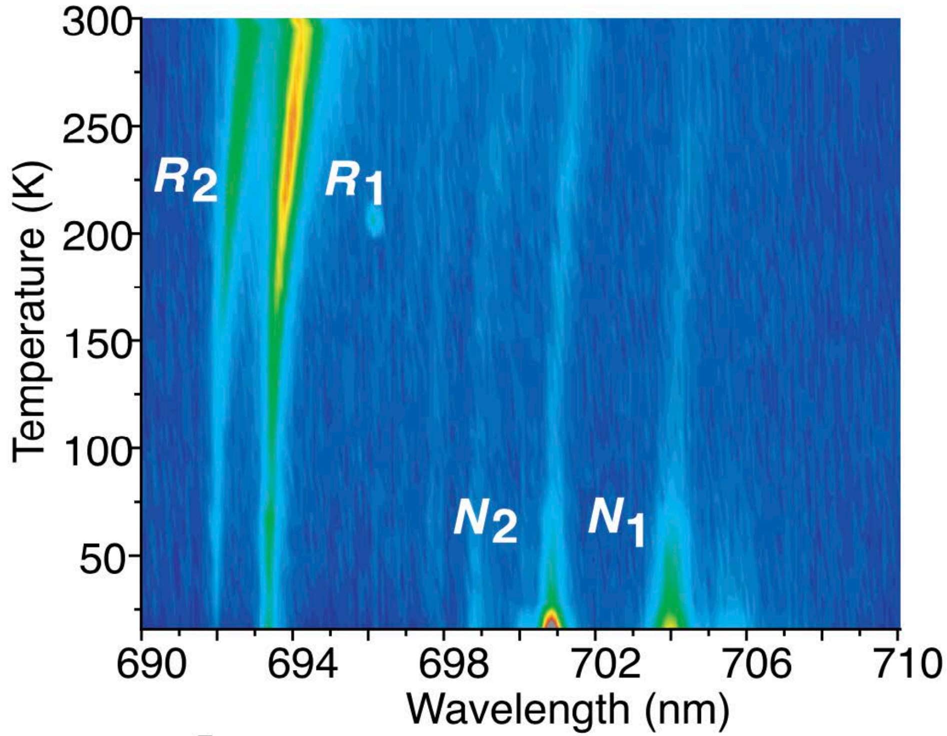


Cr^{3+} in Al_2O_3: $^2E_g \rightarrow {}^4A_{2g}$

Pressure monitoring

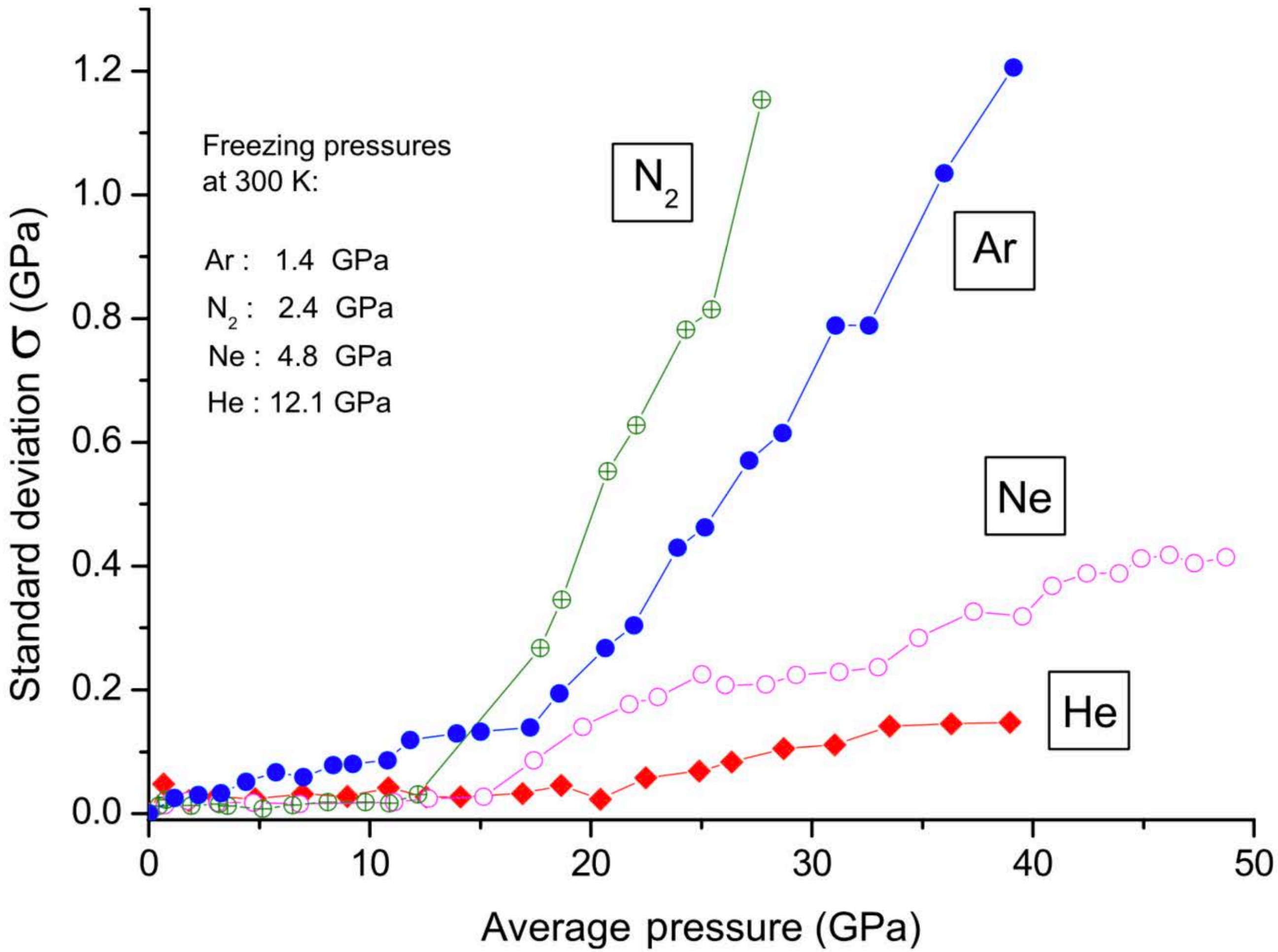


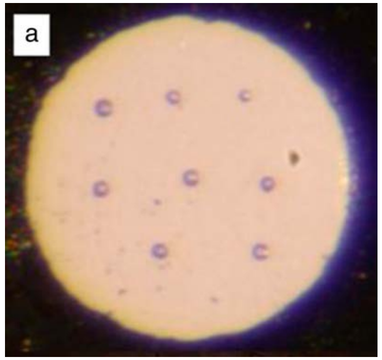
H. Yamaoka *et al.*, J. Appl. Phys. 2012



Pressure transmitting media

Pressure variation in sample space:

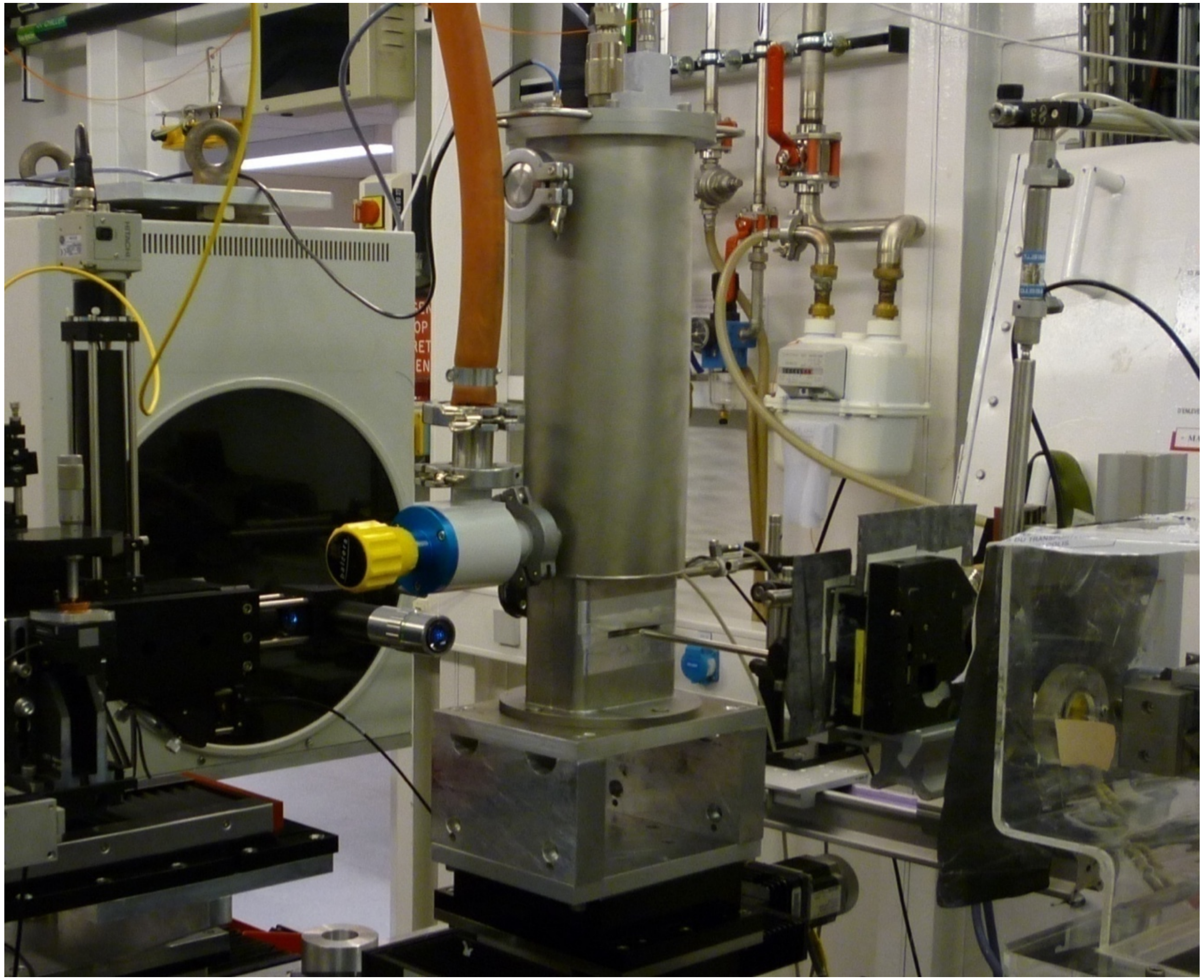




Liquids:
- methanol-ethanol mixtures
- daphne 7474
- silicone oil

S. Klotz, J. Phys. D: Appl. Phys. **42** , 075413 (2009)

LHe-flow cryostat



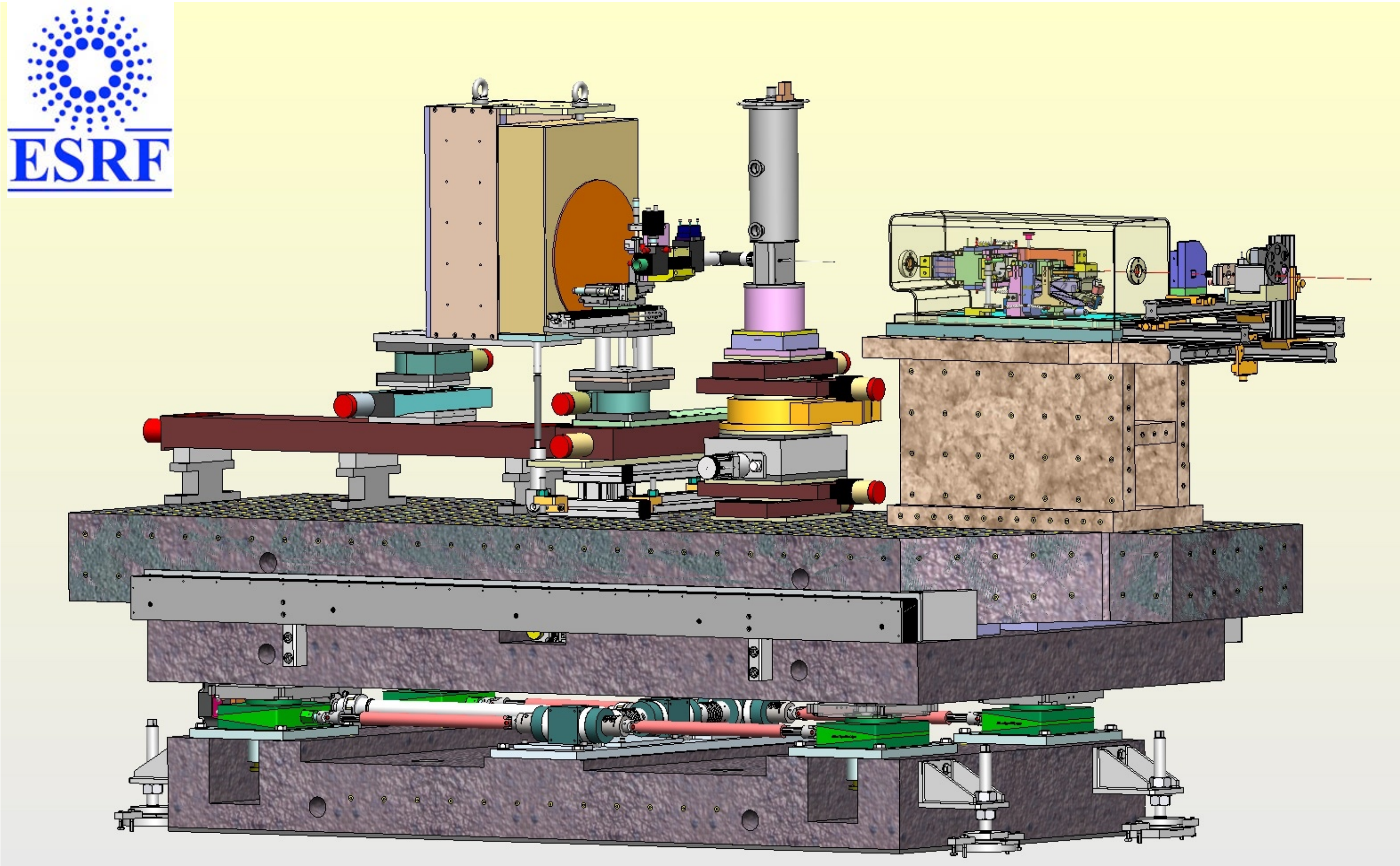
Helium flow cryostat

- T_{min}=3 K
- Membrane-driven DAC P_{max}=200 GPa
- High resolution XRD on powder and single crystal samples
- Ruby fluorescence, Raman, transport, magnetic measurements

available on ID15b, ID12, ID18, ID28, ID24

ID27 of the ESRF

Low-temperature and high-pressure XRD



ID27 before EBS

- 20 keV – 60 keV
- $\phi_{max} \simeq 1 \cdot 10^{11}$ ph/s at 33 keV in 0. 1%BW
- 0.100 mrad (H)
  0.005 mrad (V)
  with KB-mirrors
- 3x3 μm^2 beam

ID27 new

- 25 keV – 60 keV
- $\phi_{max} \simeq 8 \cdot 10^{13}$ ph/s at 33 keV in 0. 1%BW
- 0.0070 mrad (H)
  0.0045 mrad (V)
  with KB-mirrors
- 0.3 x 0.3 μm^2 beam

Comparison

In-house experiment versus ID27

ID27 new

- 25 keV – 60 keV
- $\phi_{max} \simeq 8 \cdot 10^{13}$ ph/s
  at 33 keV in 0. 1%BW
- 0.007 mrad (H)
  0.005 mrad (V)
  with KB-mirrors
- 0.3 x 0.3 μm^2 beam

ID27 before EBS

- 20 keV – 60 keV
- $\phi_{max} \simeq 1 \cdot 10^{11}$ ph/s
  at 33 keV in 0. 1%BW
- 0.100 mrad (H)
  0.005 mrad (V)
  with KB-mirrors
- 3x3 μm^2 beam

Sealed tube

- 17.5 keV and 19.6 keV
- $\phi_{max} \simeq 2 \cdot 10^{7}$ ph/s
  at 17.5 keV
- 1 mrad – 3 mrad
  2D focusing optics
- 80x80 μm^2 beam

ESRF-EBS

smaller samples
higher pressures
high resolution
higher sensitivity

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- Why pressure and low temperatures?!!
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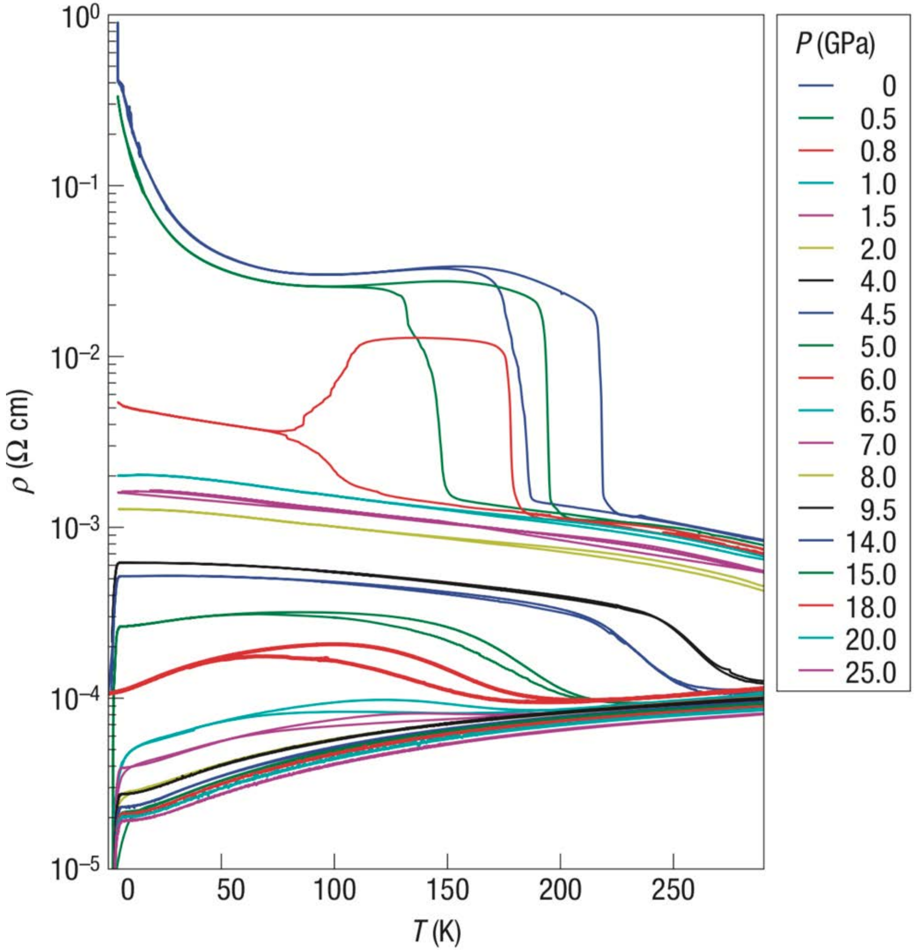
  – Strong covalency: Towards room temperature SC

Examples: Pressure-induced SC




ARTICLES

From Mott state to superconductivity in 1T-TaS_2

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[1]Ecole Polytechnique Fédérale de Lausanne, IPMC, CH-1015 Lausanne, Switzerland
[2]Institute of Physics, Bijenička c. 46, Zagreb, Croatia
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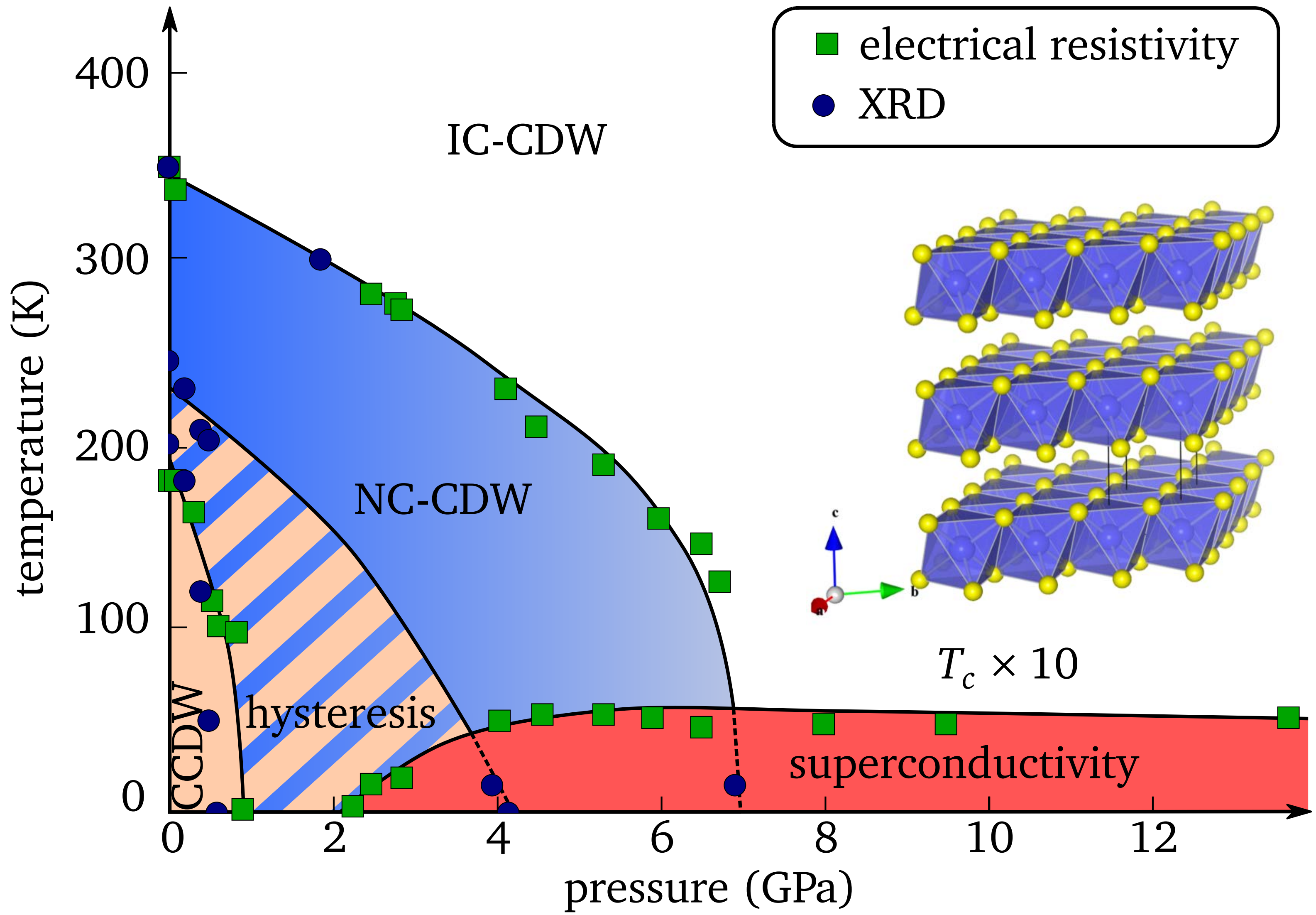
Published online: 9 November 2008; doi:10.1038/nmat2318

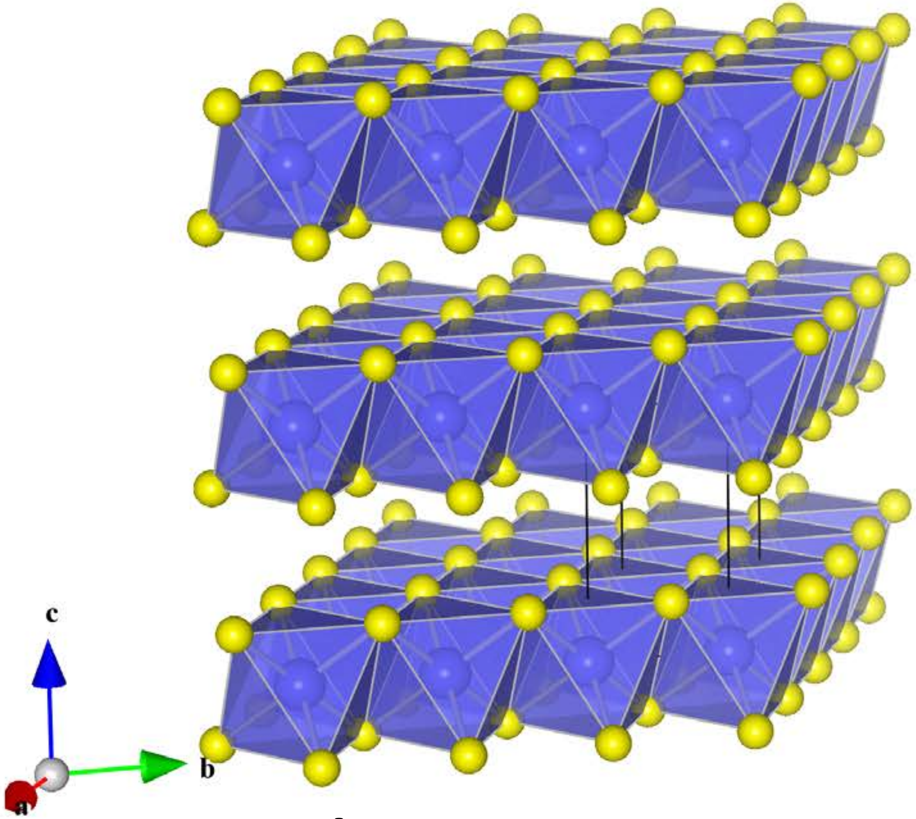
- What happens at the atomic level?
- Structural changes?
- Electronic order in real space?

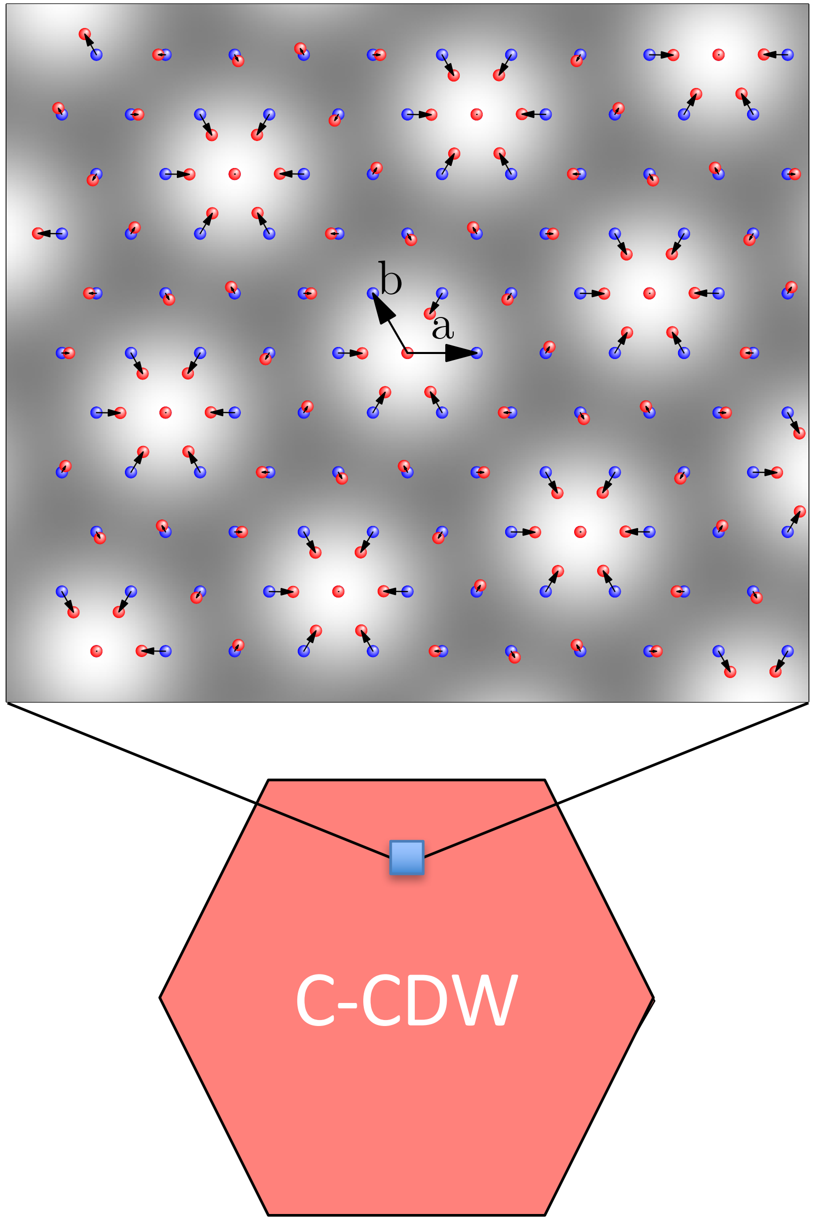
p-T phase diagram

Resistivity by Spios et al., Nature Materials (2008)



CDWs in 1T-TaS_2





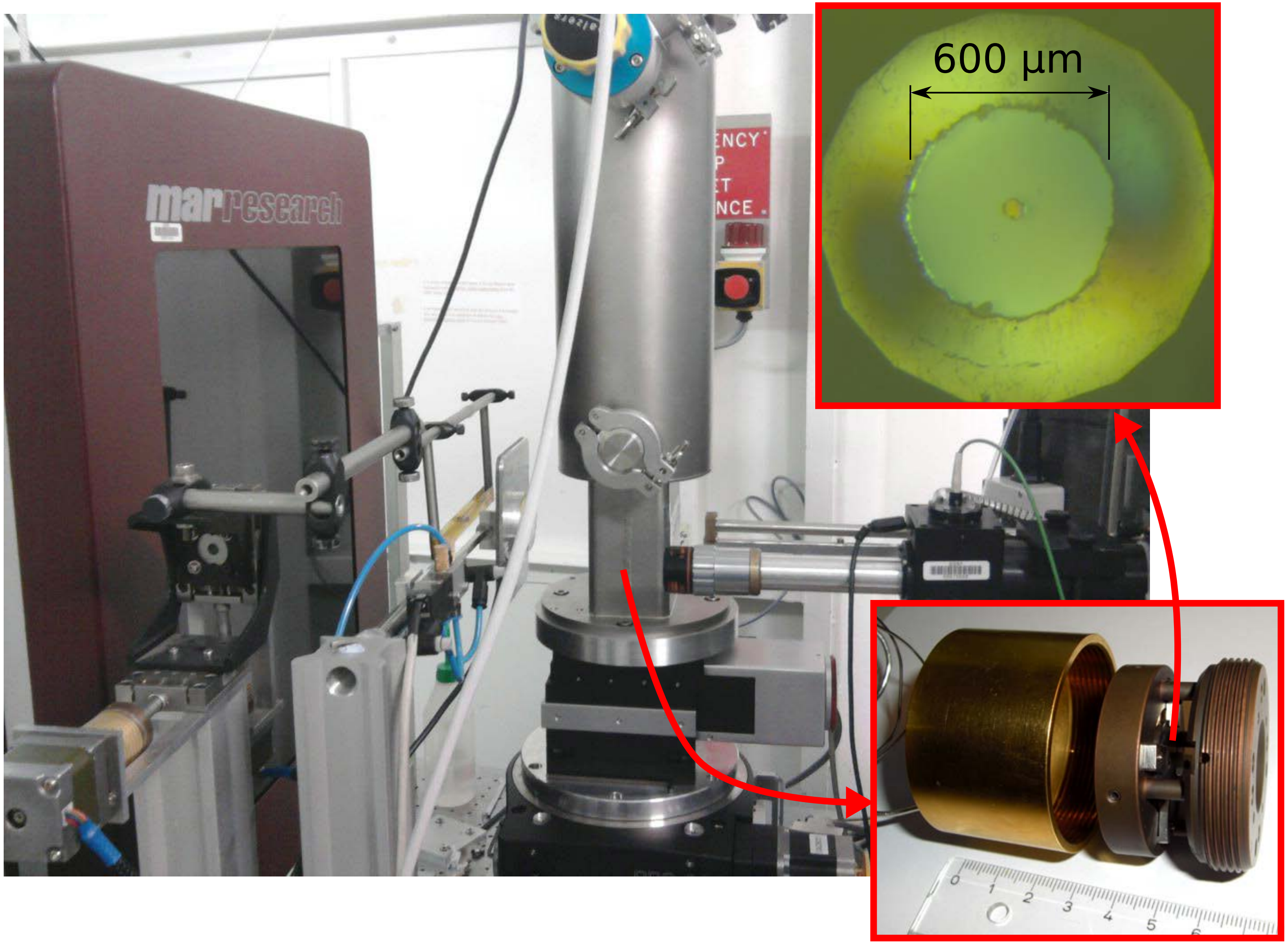
Nearly commensurate CDW:

- ordered CDW-defects
- stabilized by P at low T
- becomes SC at low T

(Sipos *et al.,* Nature Mat. 2008)

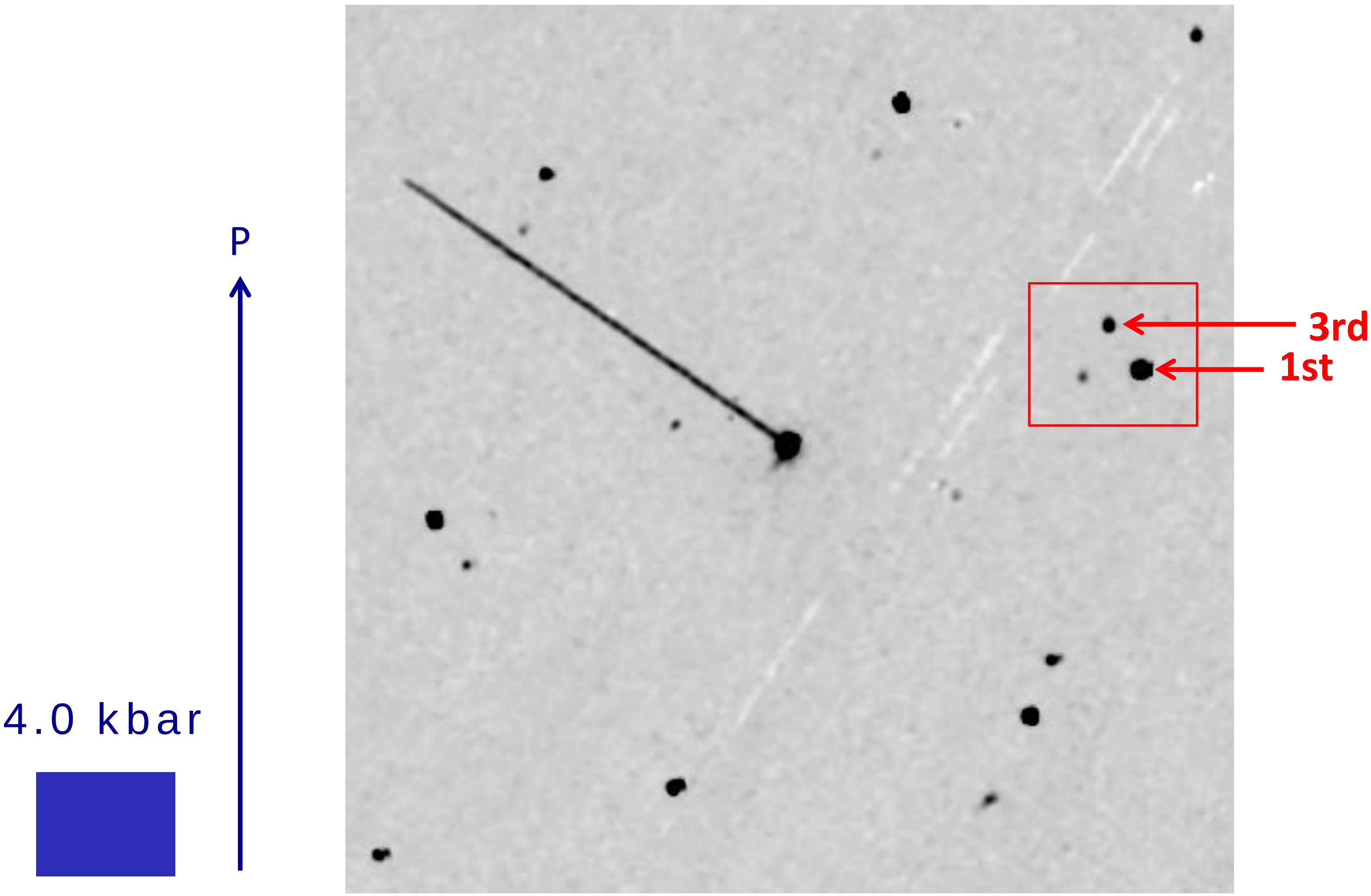
Experiment at ID09

ESRF



Nearly commensurate CDW

Intensity map in reciprocal space (HK0-plane)



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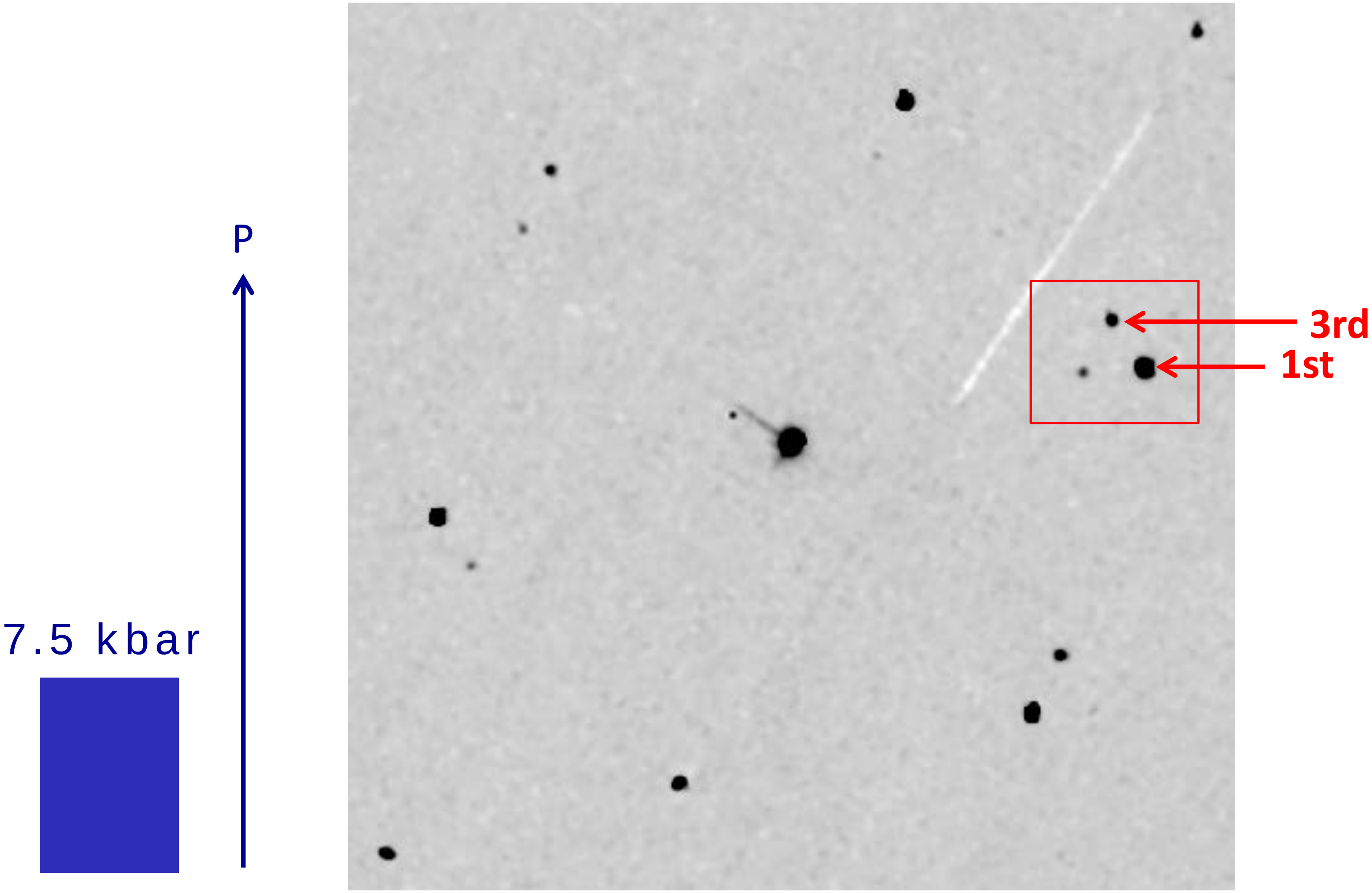

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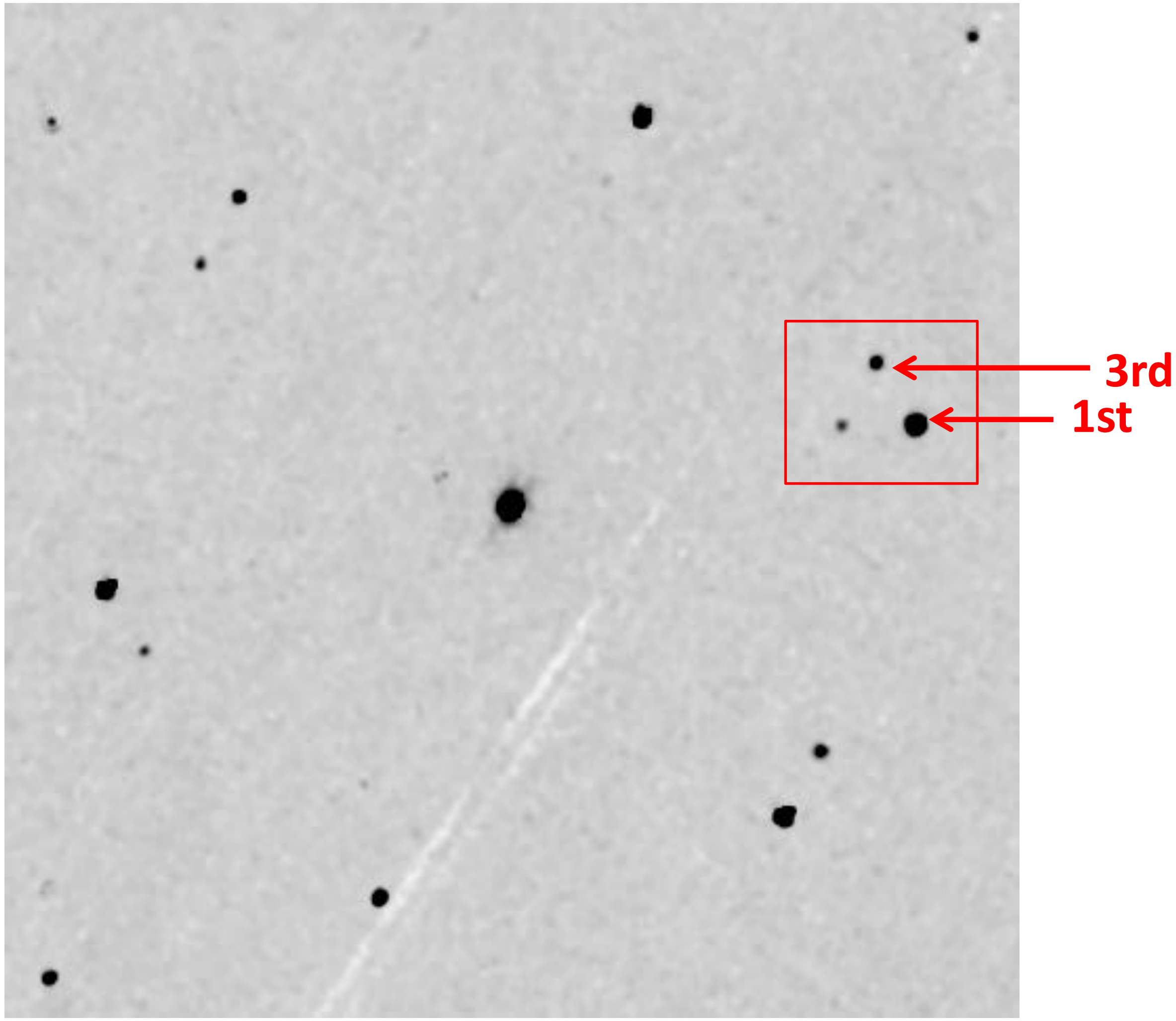

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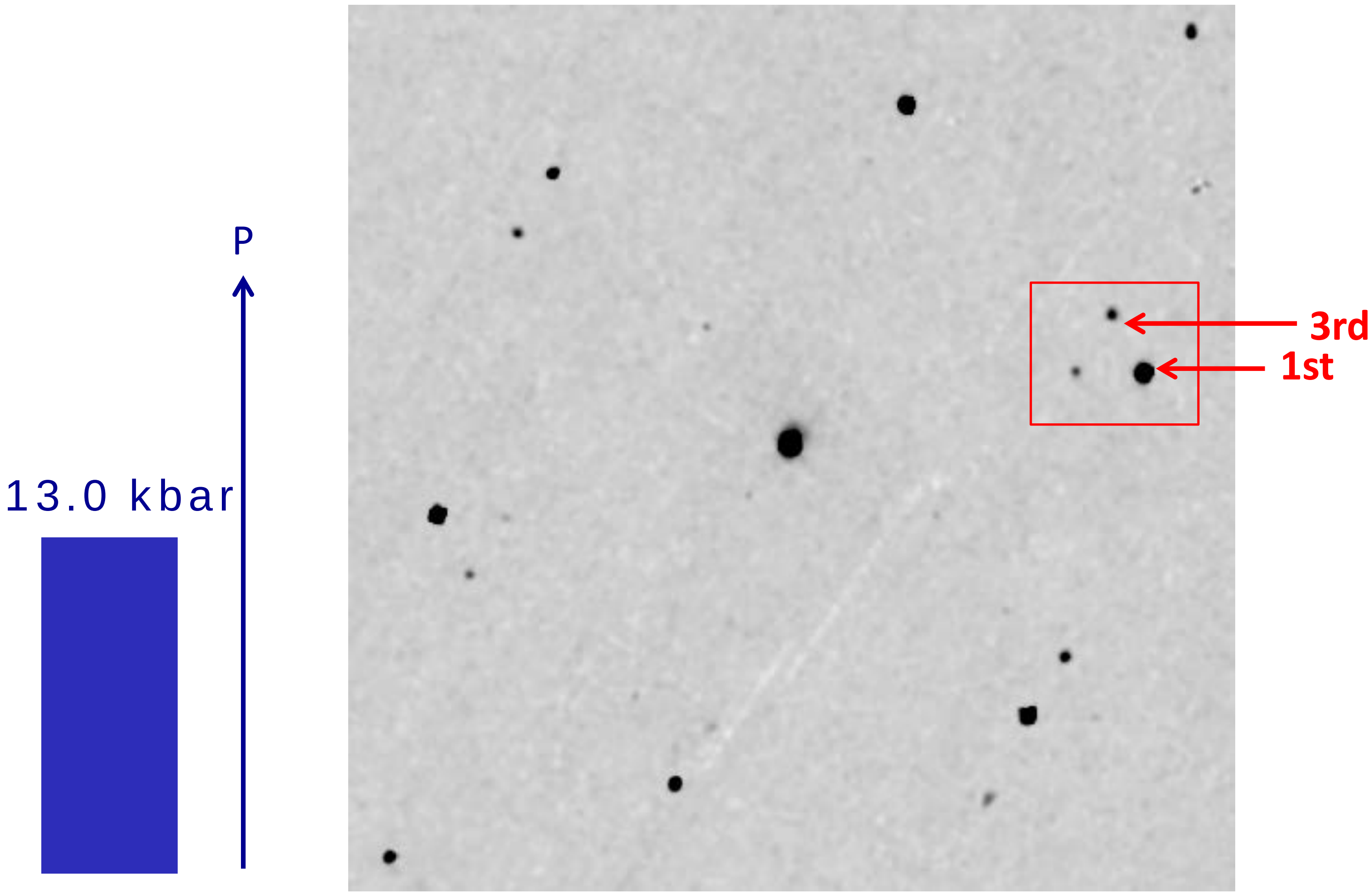

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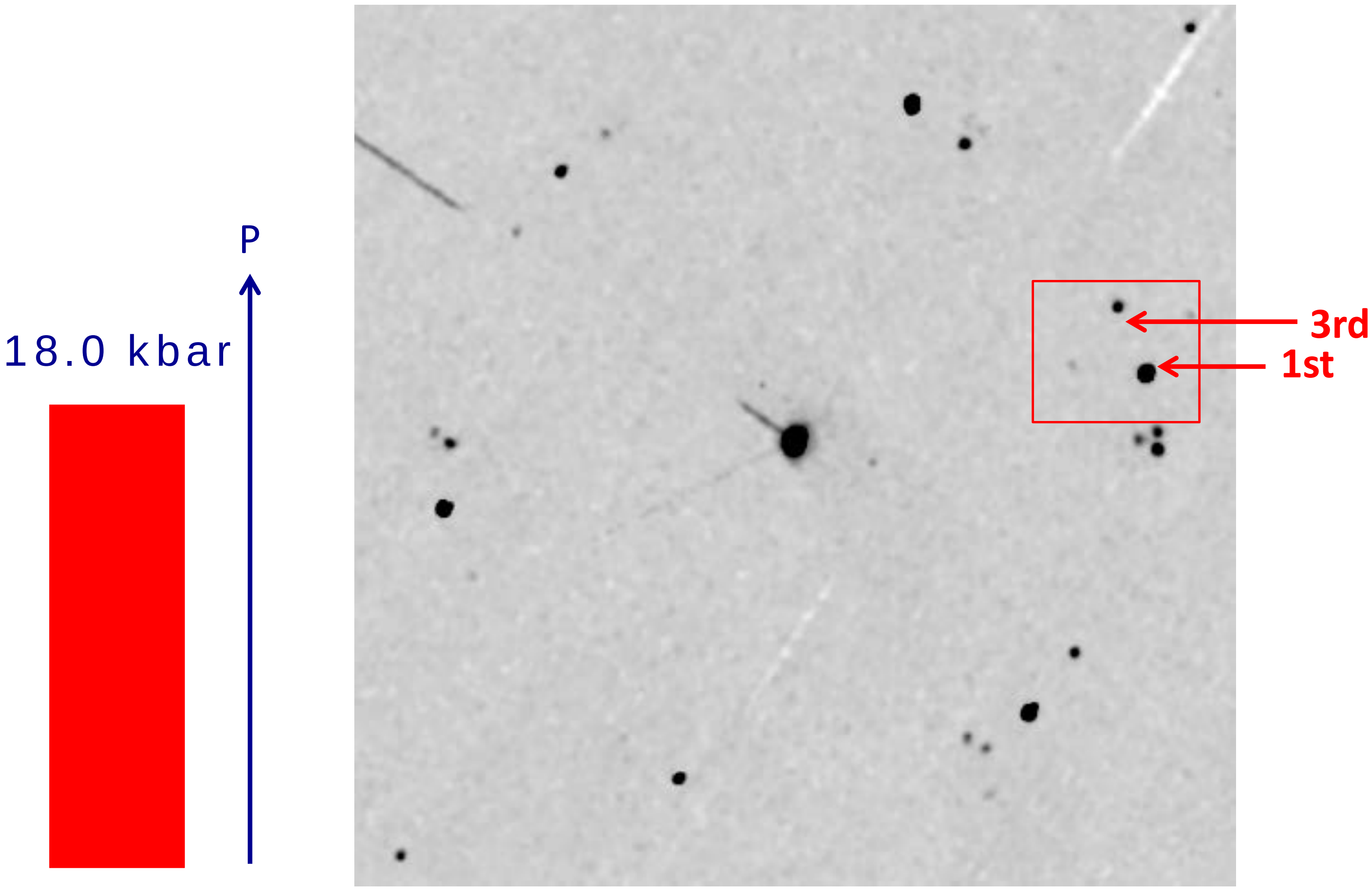
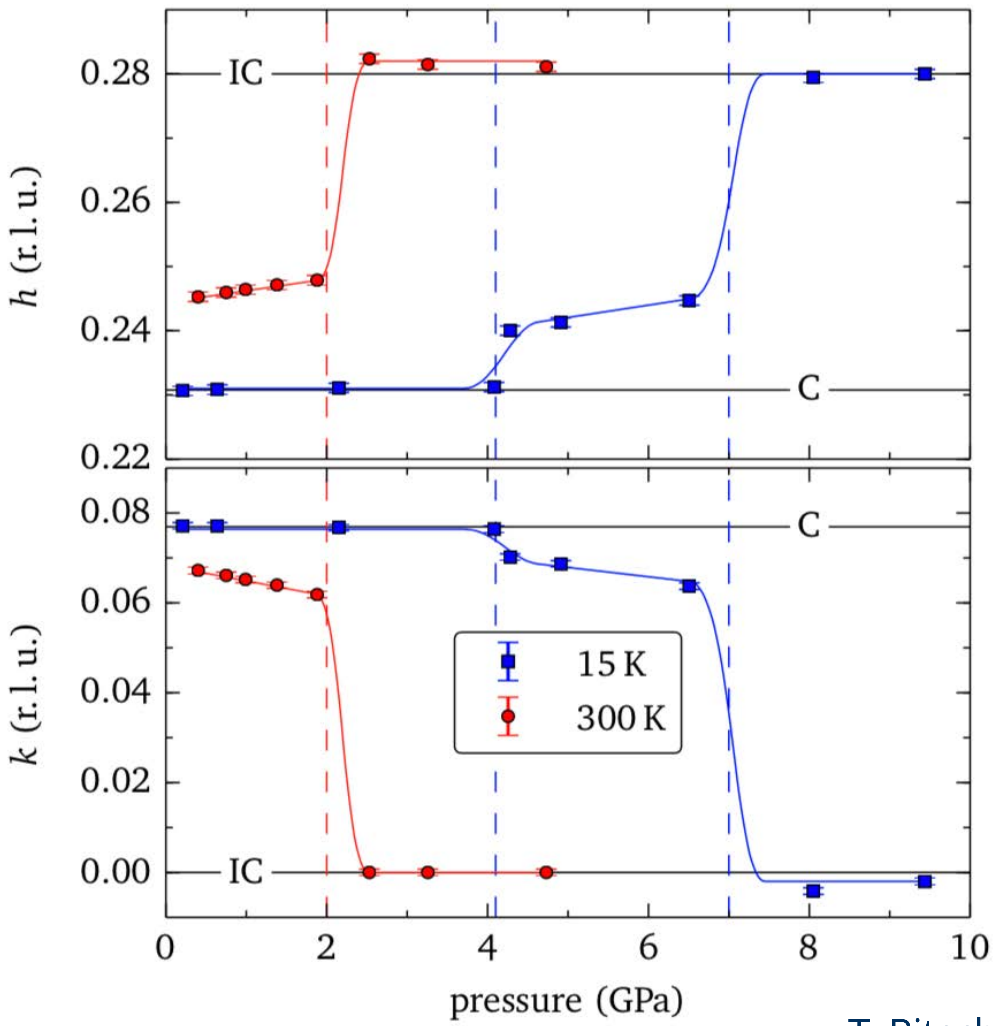


q versus P

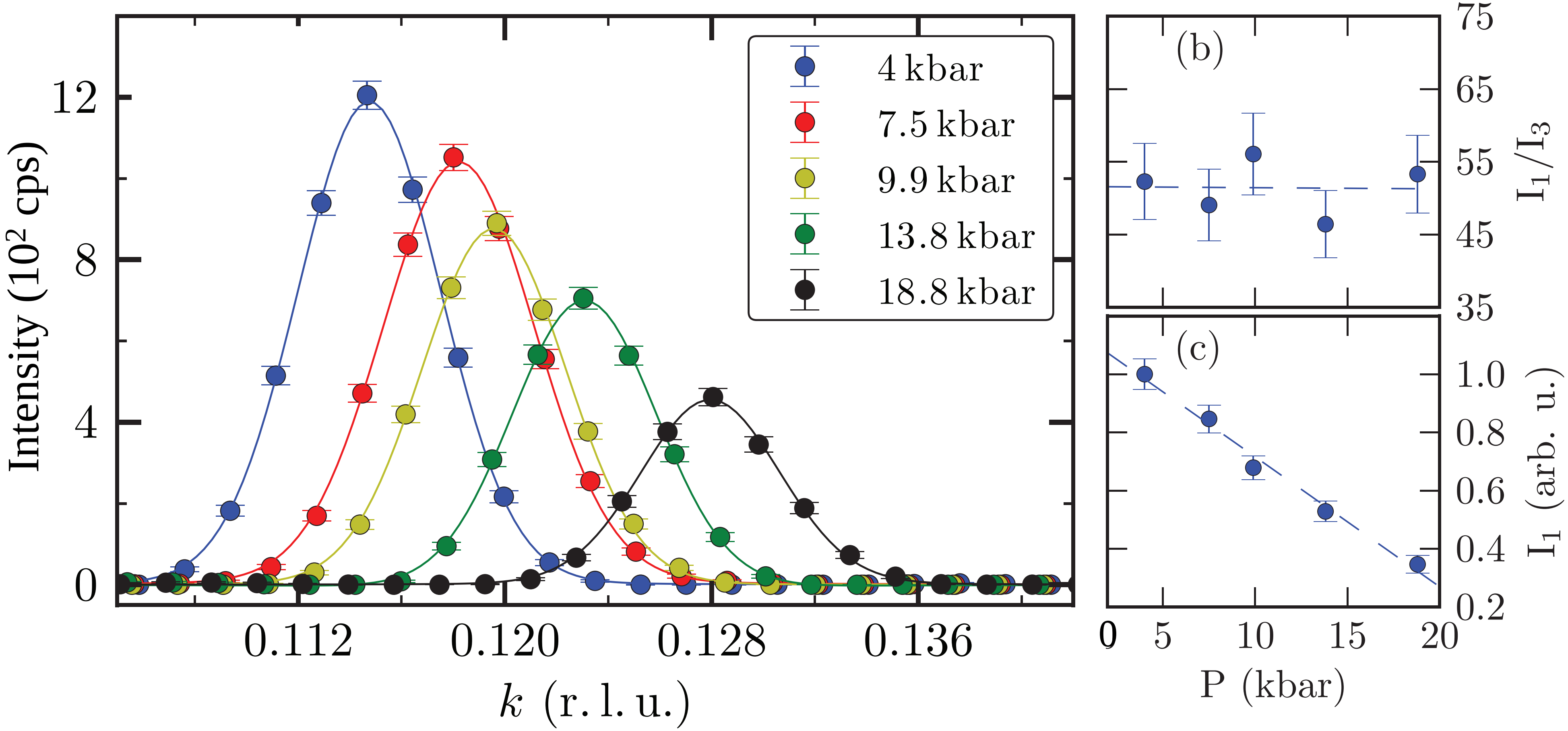


T. Ritschel *et al.*, PRB **87** (2013)

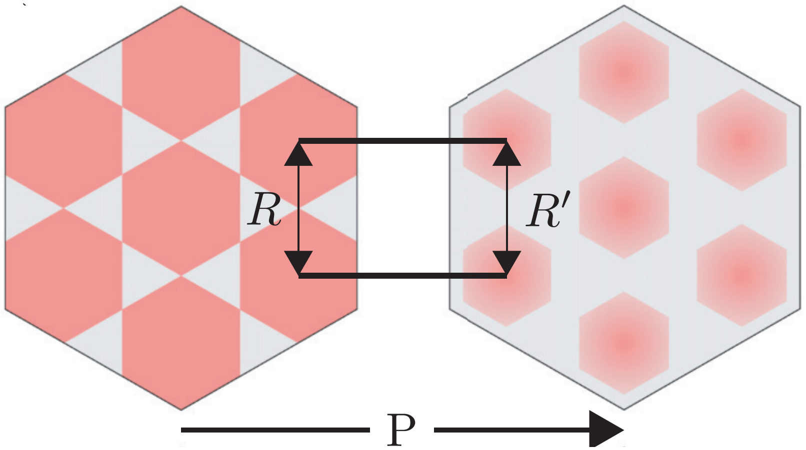
q versus P

Nearly commensurate CDW

T. Ritschel *et al.*, PRB **87** (2013)



Superconducting CDW



- R≈R′ → q constant
- Defects widen → intensity ratios change

Expectations:

q constant

intensity ratios change

$$R = \frac{8\pi}{3\sqrt{13} \cdot |\mathbf{q} - \mathbf{q}_C|}$$

XRD experiment:

- q changes with P
- Constant intensity ratios

  ➔ Internal structure of defects unchanged

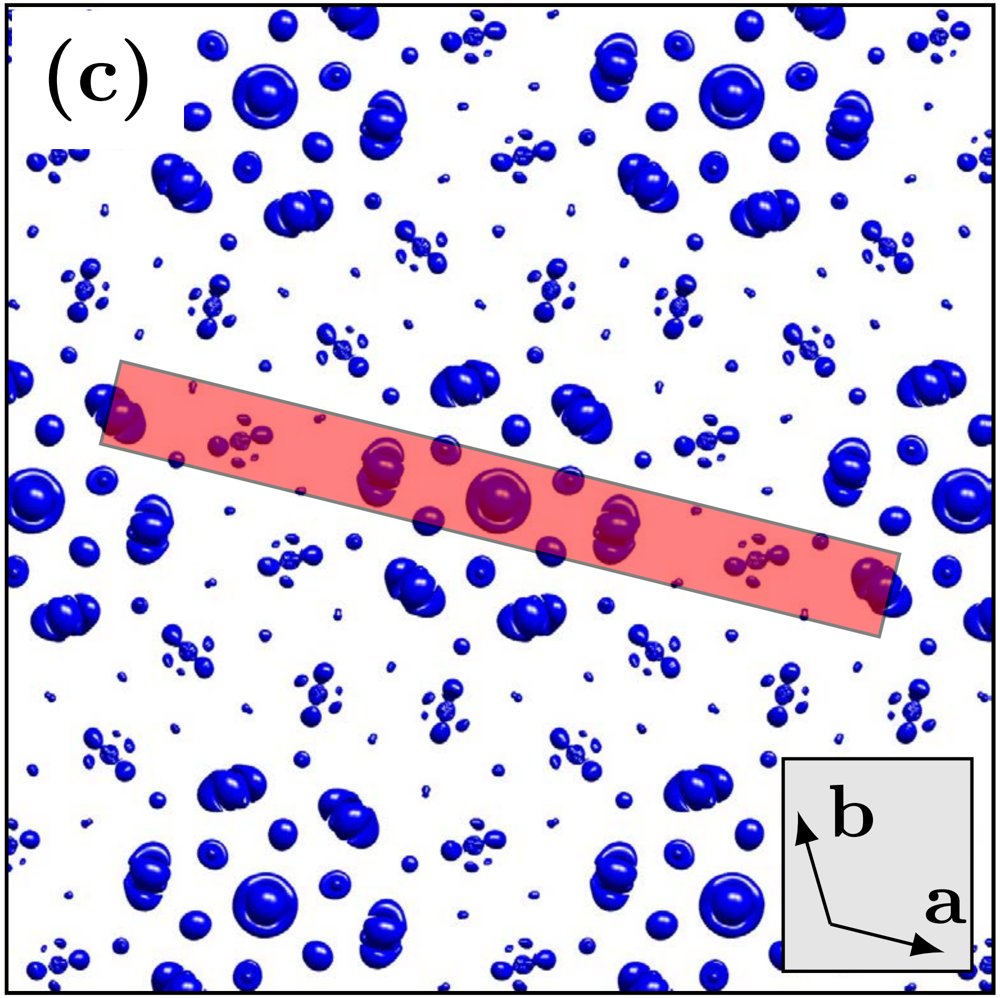
  ➔ **Defect lattice compressed!**

Whole CDW-defect lattice is SC

T. Ritschel *et al.*; PRB **87**, 125135 (2013)

What drives P-dependence?!

Quasi-molecular orbitals: orbital-lattice coupling



T. Ritschel *et al.*,
Nature Phys. (2015)

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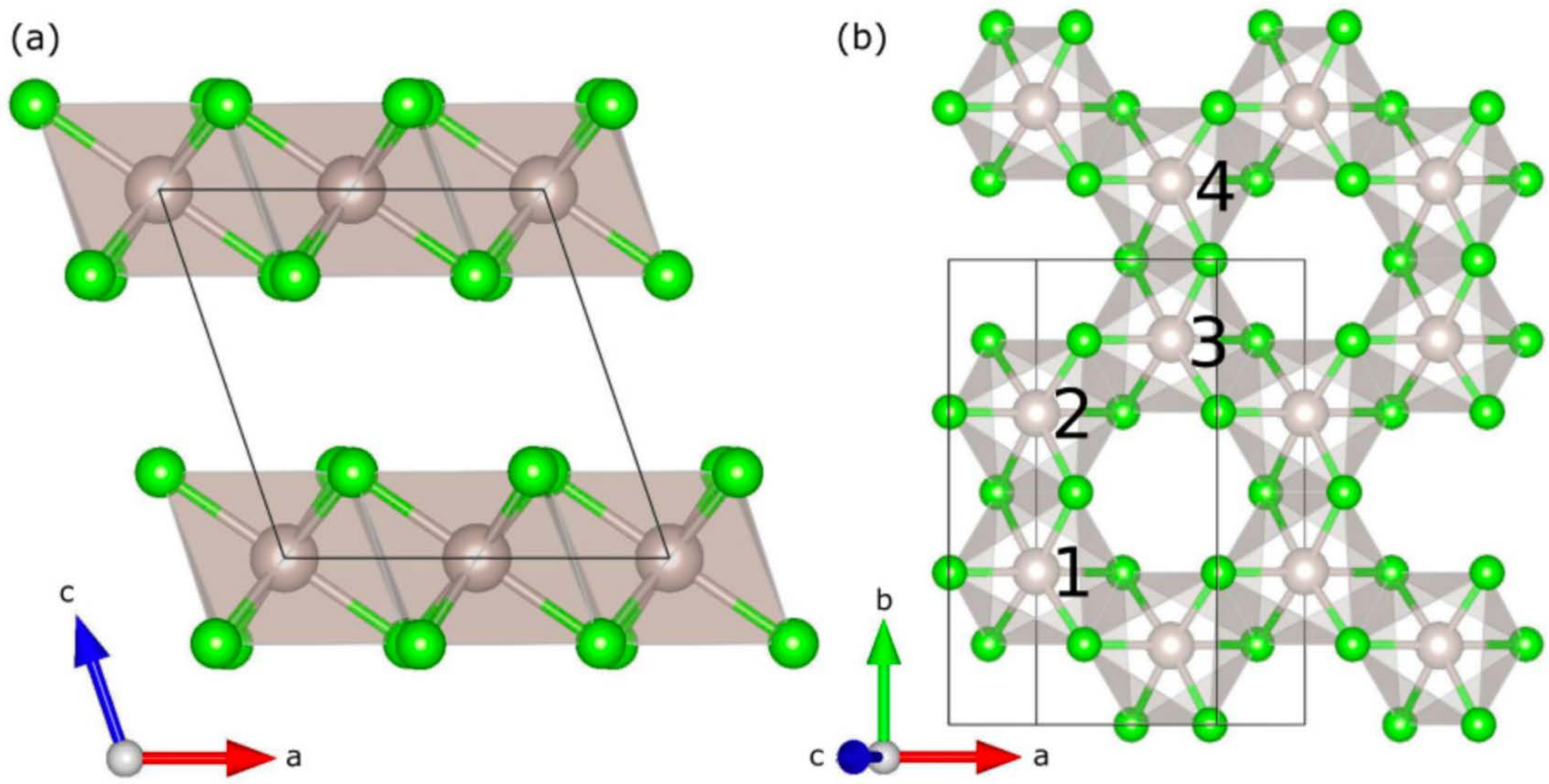
α-$RuCl_3$: candidate for Kitaev spin liquid

Monoclinic lattice structure (ambient)

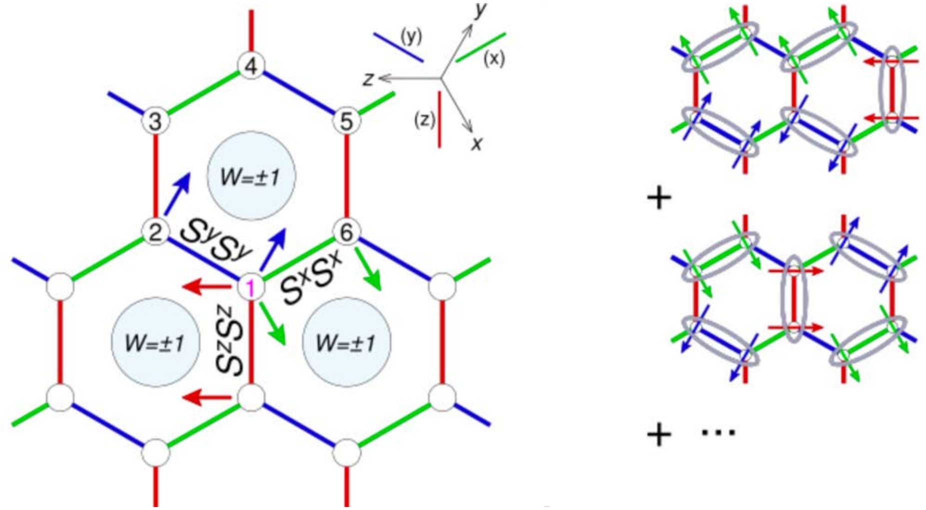


R.D. Johnson *et al.*, PRB **92** (2015)

α-RuCl$_3$: candidate for Kitaev spin liquid

Kitaev magnetism

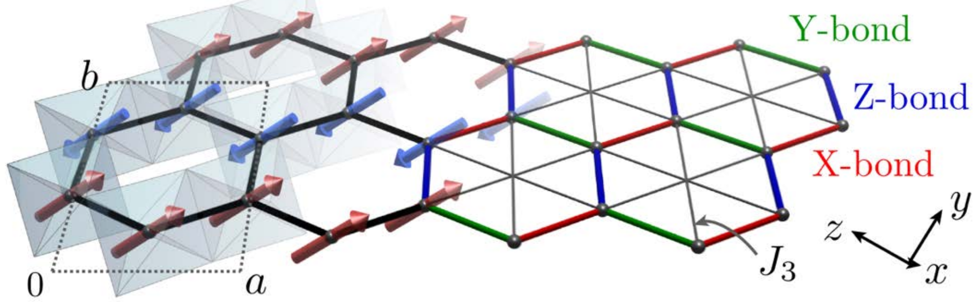
H. Takagi *et al.*, Nature Rev. (2019)



- Exactly solvable model
- Quantum spin liquid

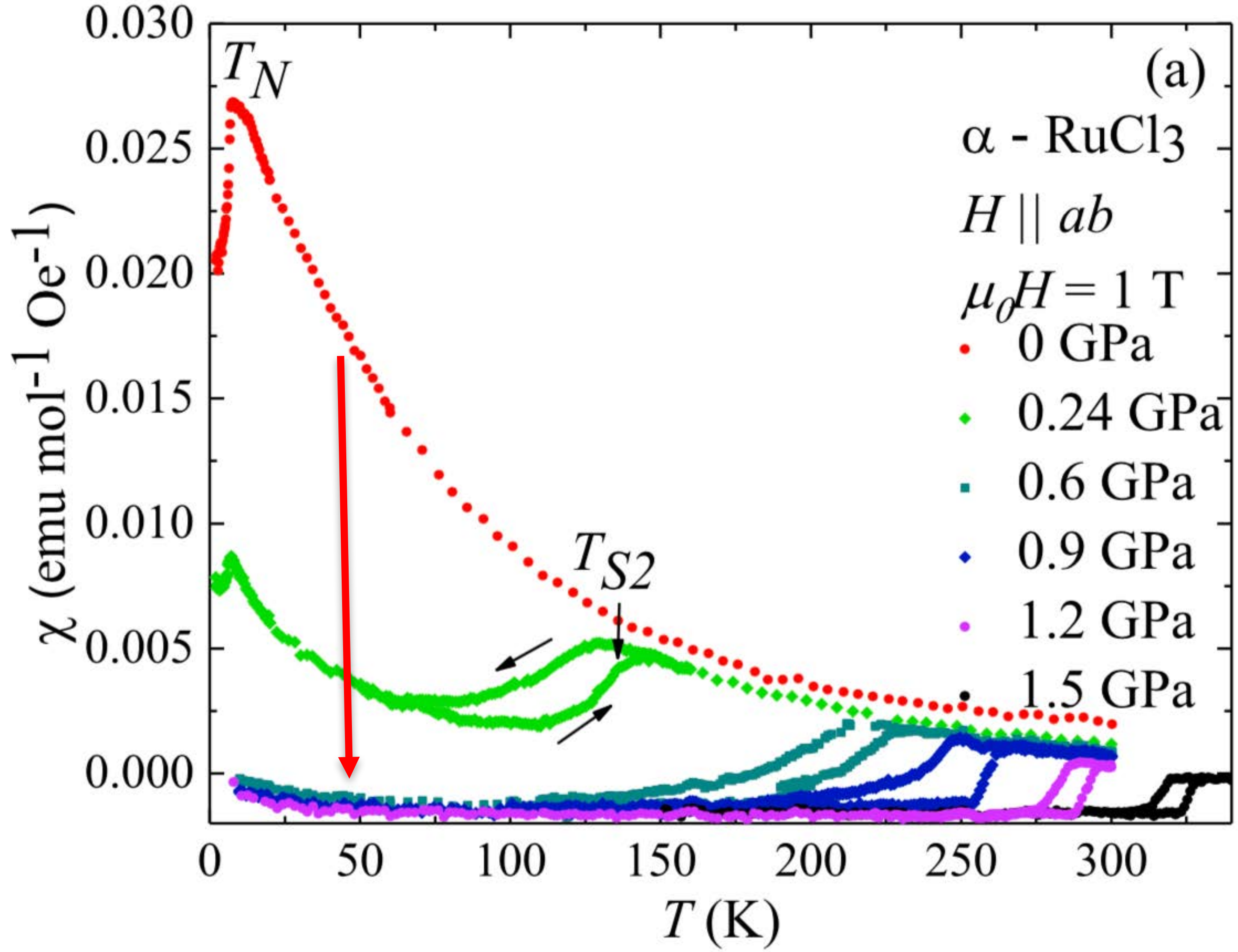
Realization in α-RuCl$_3$

R. Valenti *et al.*, Univ. Frankfurt



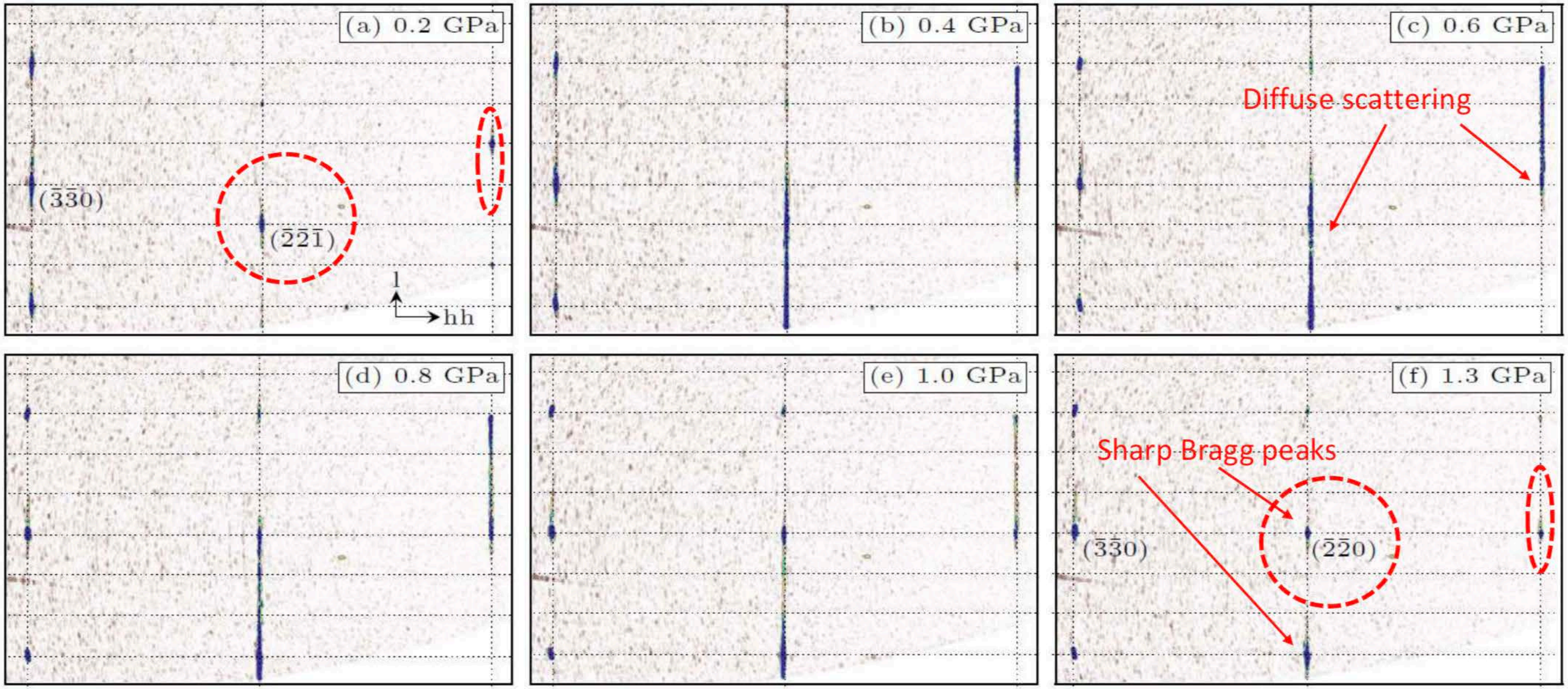
- But: AFM below 7 K!
- **Hydrostatic P**?!! (need to avoid symmetry breaking!!)

Pressure dependent magnetism

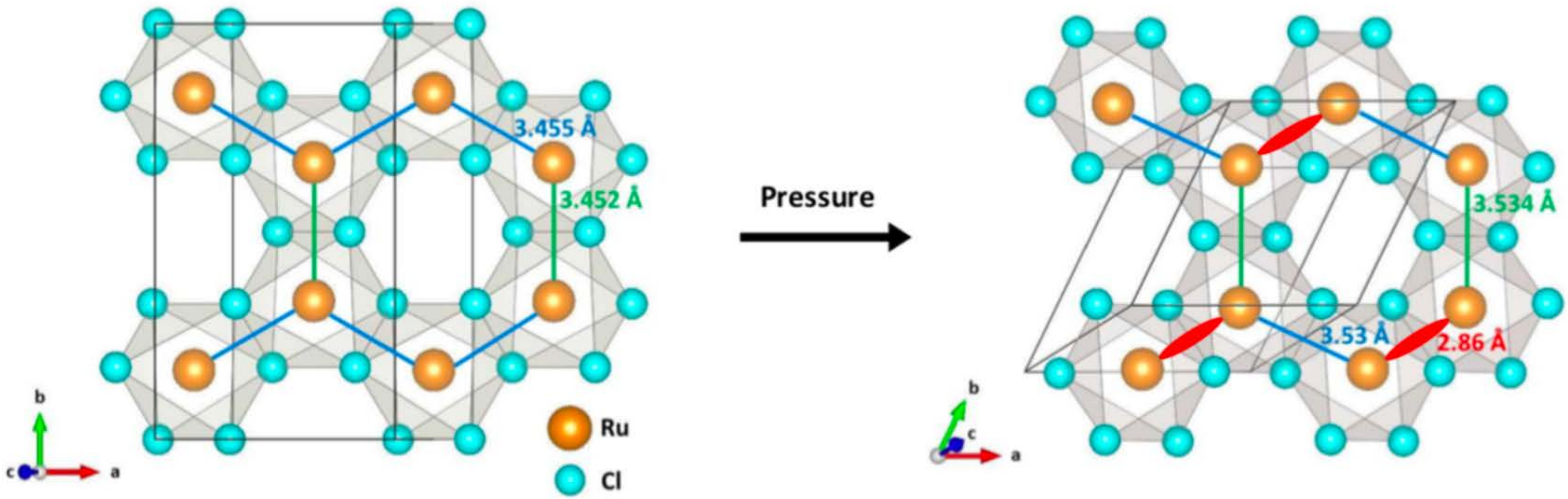


G. Bastien *et al.*, PRB **97** (2018)

Pressure-driven structural transition



XRD: Structure refinement



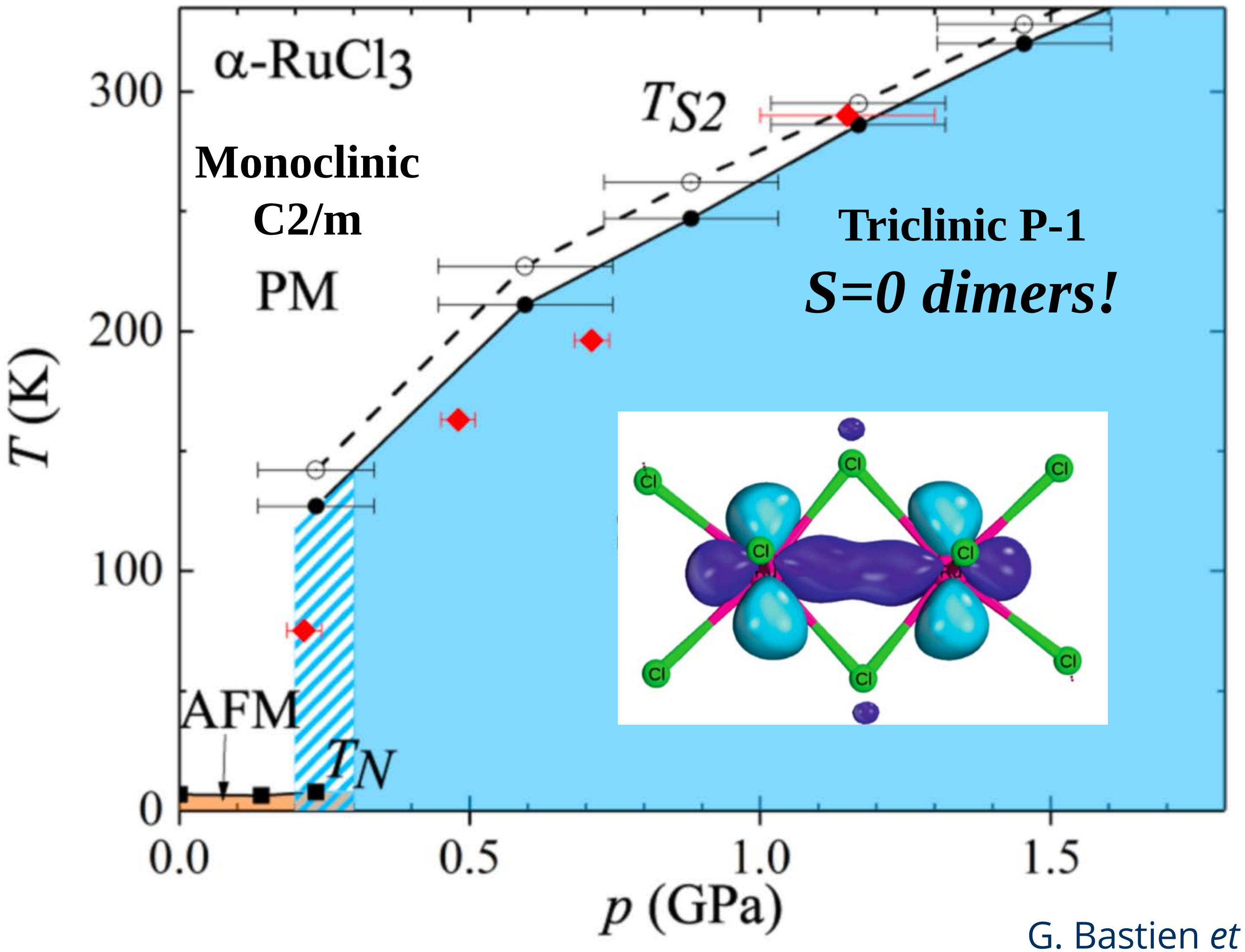
Monoclinic
C2/m

Triclinic
P-1

G. Bastien *et al.*, PRB **97** (2018)

Pressure-induced dimerization

Quasi-molecular orbitals: orbital-lattice coupling



G. Bastien *et al.*, PRB **97** (2018)

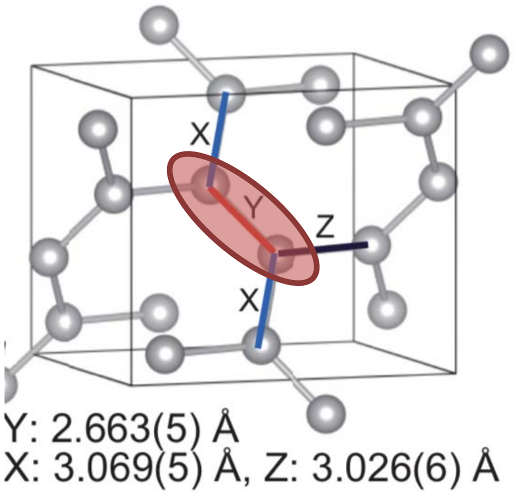
Other example: Li_2IrO_3

Pressure-induced dimerization: Wide spread in 4d- 5d TMOs!

PHYSICAL REVIEW B **99**, 125127 (2019)

Pressure-induced collapse of the spin-orbital Mott state in the hyperhoneycomb iridate β-I

T. Takayama,[1,2] A. Krajewska,[1,2] A. S. Gibbs,[3] A. N. Yaresko,[1] H. Ishii,[4] H. Yamaoka,[5] K. Ishii,[6] N. Hiraoka,[4] N. P. Funnell,[3] C. L. Bull,[3] and H. Takagi[1,2,7]

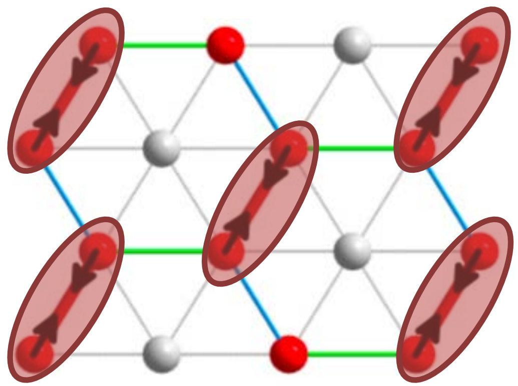


PHYSICAL REVIEW B **97**, 020104(R) (2018)

Rapid Communications

Competition between spin-orbit coupling, magnetism, and dimerization in the honeycomb iridates: α-Li_2IrO_3 under pressure

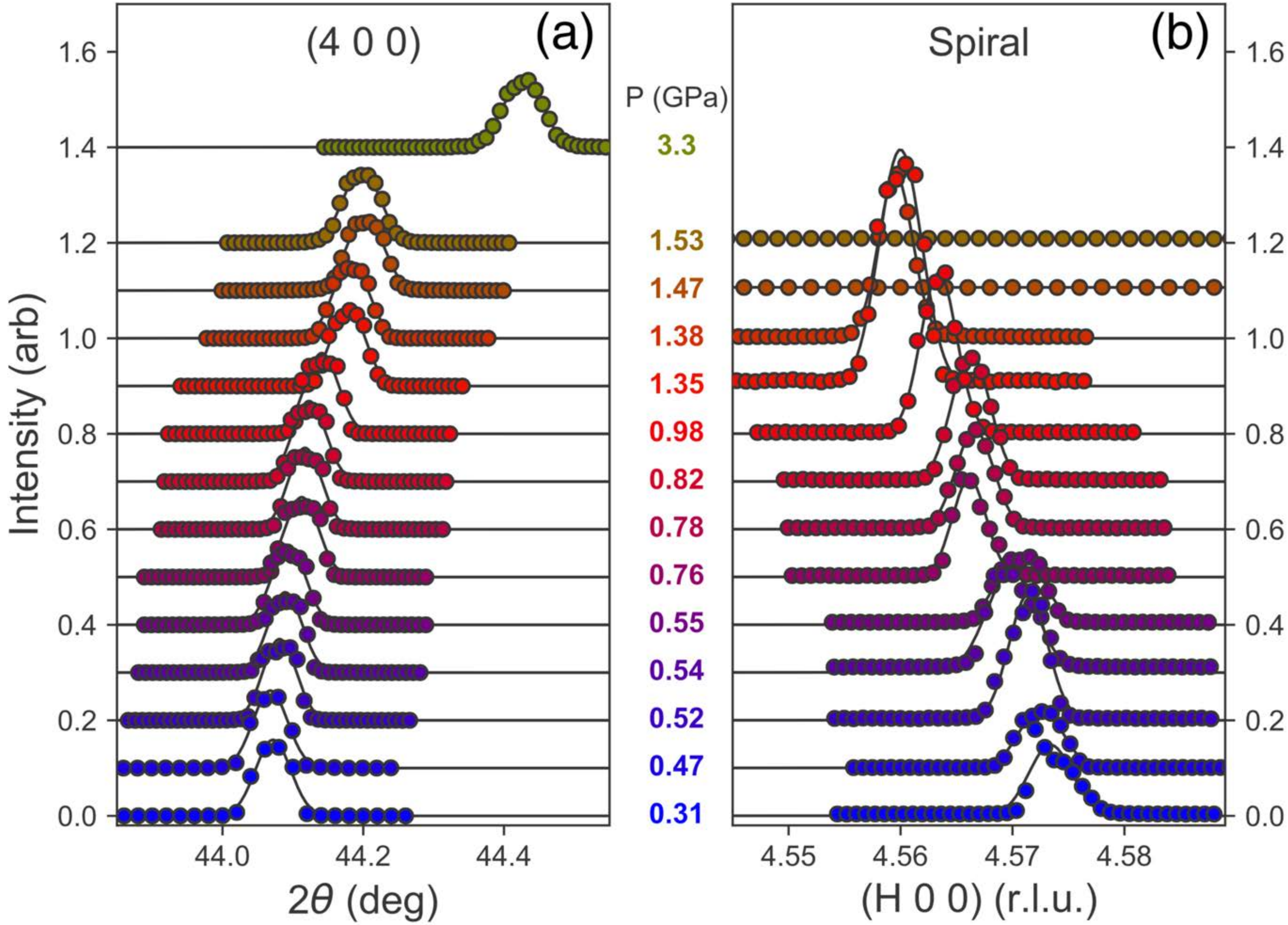
V. Hermann,[1] M. Altmeyer,[2] J. Ebad-Allah,[1,3] F. Freund,[4] A. Jesche,[4] A. A. Tsirlin,[4] M. Hanfland,[5] P. Gegenwart,[4] I. I. Mazin,[6] D. I. Khomskii,[7] R. Valentí,[2,*] and C. A. Kuntscher[1,†]



γ-Li_2IrO_3

Resonant **magnetic** x-ray scattering at the Ir L_3-edge

T= 4.7 K



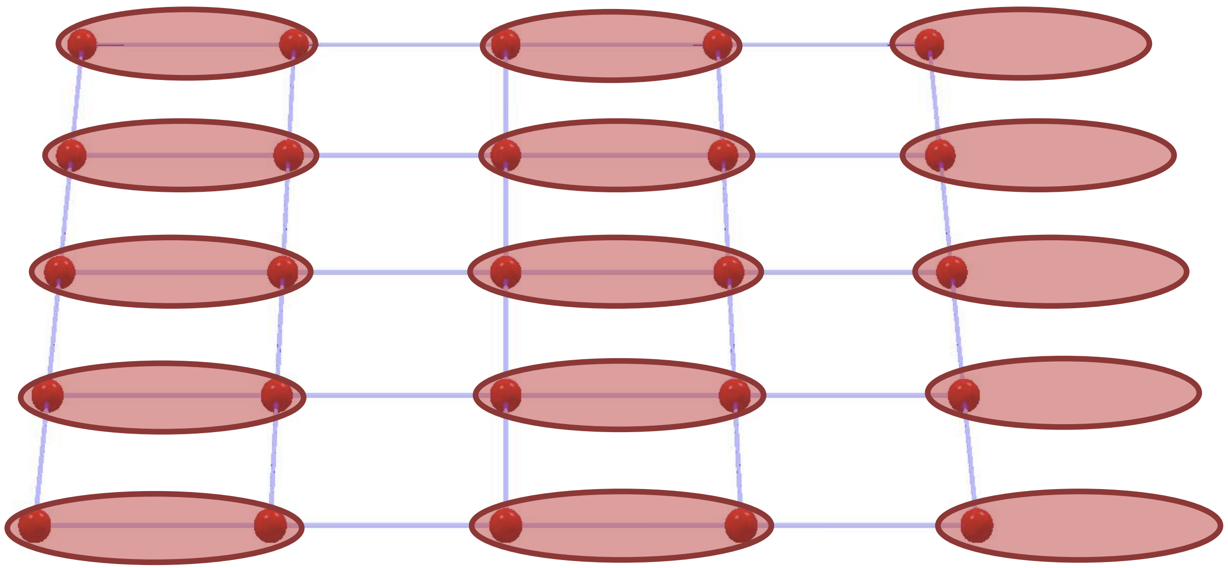
N.P. Breznay *et al.*, PRB 2017

Outline

- Why pressure and low temperatures?!!
- Typical experimental setups
- Scientific examples
  - Charge density waves and superconductivity
  - Pressure-driven covalency and magnetism
  - Strong covalency: Towards room temperature SC

H_2 under pressure

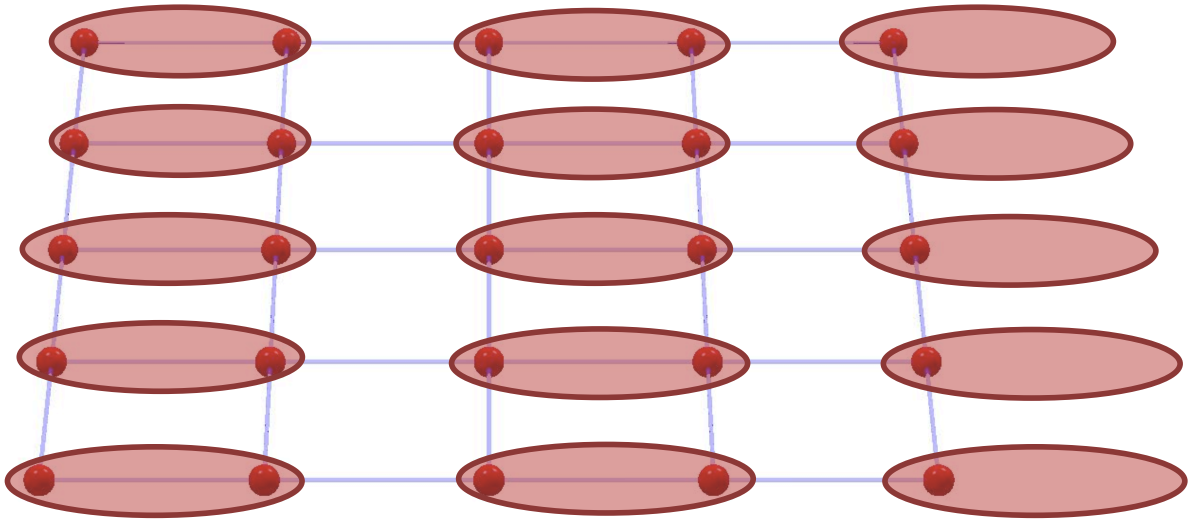
Cartoon! (We start from molecular orbitals)



- Lattice of weakly interacting H_2-molecules
- Electrons localized in covalent bonds
- Insulator

H_2 under pressure

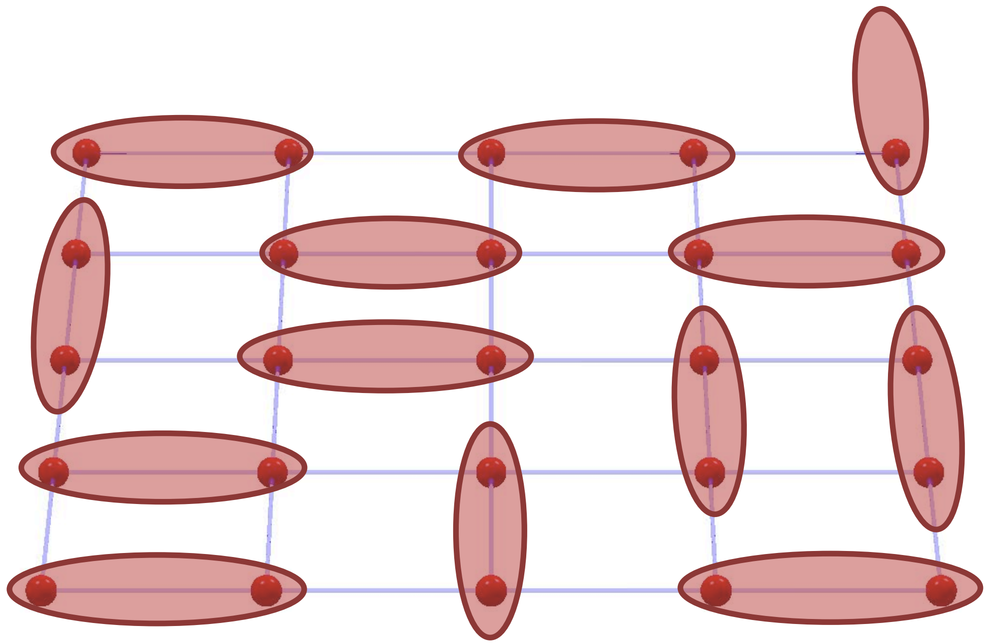
Cartoon!



- Pressure: weak bonds become stronger

H_2 under pressure

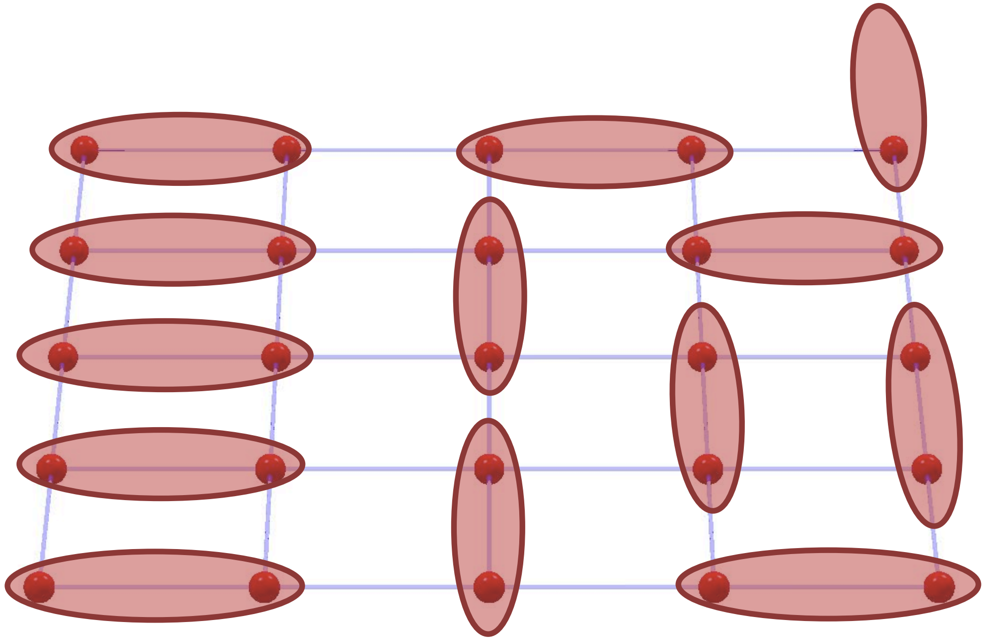
Cartoon!



- Pressure: weak bonds become stronger
- Various bond configurations possible
- Delocalization!

H_2 under pressure

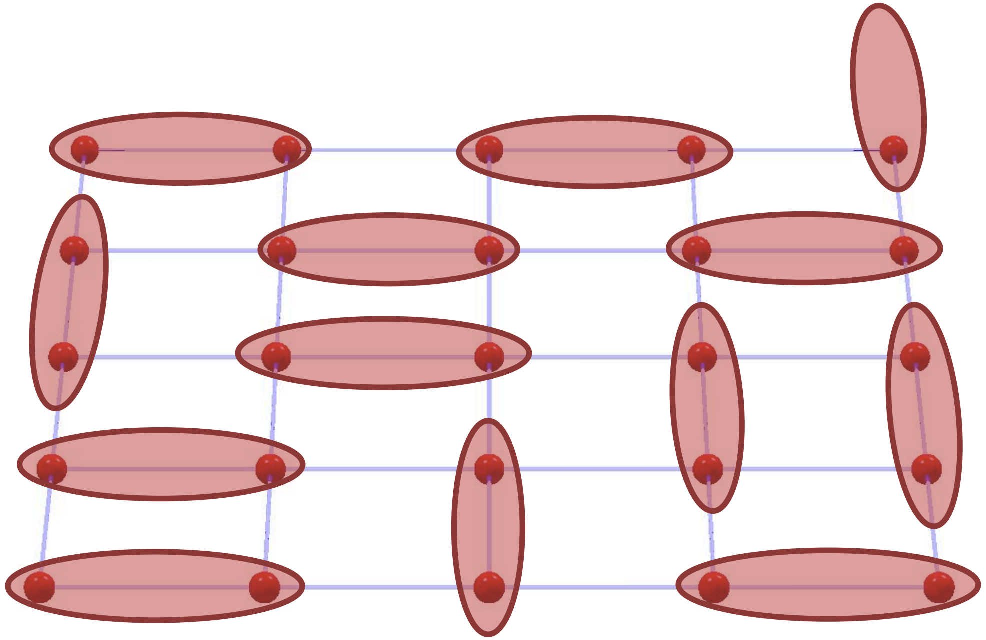
Cartoon!



- Pressure: weak bonds become stronger
- Various bond configurations possible
- **Delocalization!**

H_2 under pressure

Hydrogen is predicted to be a conventional HTSC!



High!
Light H-sites

Large!
Covalent bonds involved

$$T_c = \frac{\omega_{\log}}{1.2k_B} \exp\left[-\frac{1.04(1+\lambda)}{\lambda - \mu^*(1+0.62\lambda)}\right]$$

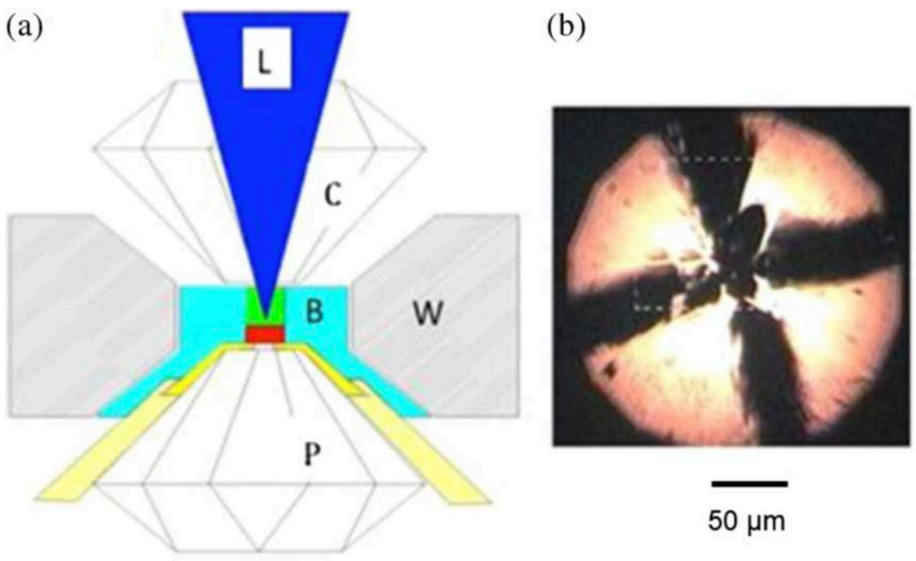
Conventional SC close to RT?!

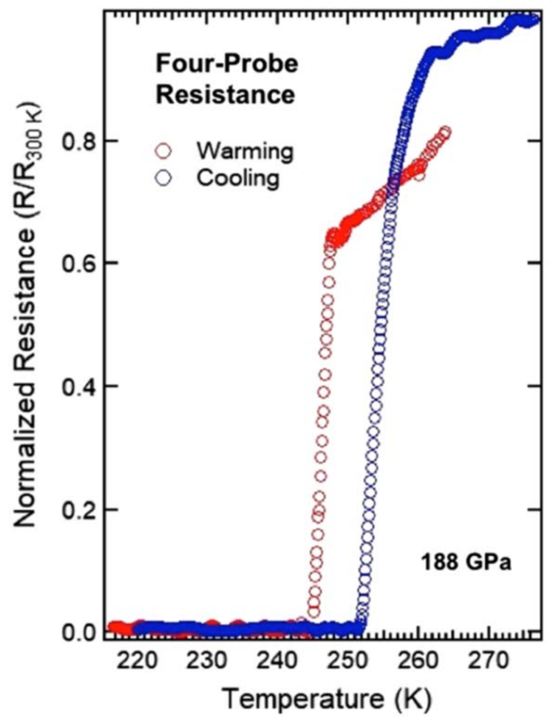
PHYSICAL REVIEW LETTERS **122,** 027001 (2019)

Editors' Suggestion | Featured in Physics

Evidence for Superconductivity above 260 K in Lanthanum Superhydride at Megabar Pressures

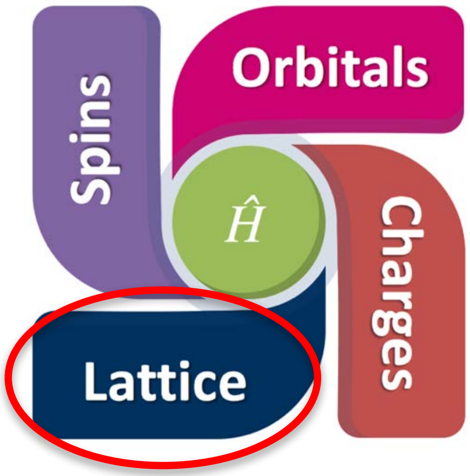
Maddury Somayazulu,[1,*] Muhtar Ahart,[1] Ajay K. Mishra,[2,‡] Zachary M. Geballe,[2] Maria Baldini,[2,§] Yue Meng,[3] Viktor V. Struzhkin,[2] and Russell J. Hemley[1,†]





Conclusion

- Interacting electron systems: fascinating, often puzzling phenomena
- Experiments needed!
  → Low T: Ground state properties
  → High P: tune lattice without external symmetry breaking



- Tweak balance between different interactions
- Here: intersite covalency & quasi-moelcular orbitals
- ESRF-EBS Enables to explore uncharted regions of the phase diagrams!

Thank you very much!!