'PRINCESS'

Raman Scattering study of URu_2Si_2

J. Buhot, M.-A. Méasson, M. Cazayous, Y. Gallais, A. Sacuto
Laboratoire Matériaux et Phénomènes Quantiques, UMR 7162 CNRS, Université Paris Diderot - Paris 7, France

Collaborations:

R. Lobo
LPEM, ESPCI-ParisTech, CNRS, UPMC, F-75231 Paris Cedex 5, France

P. Piekarz
Institute of Nuclear Physics, Polish Academy of Sciences, 31-342 Kraków, Poland

F. Bourdarot, S. Raymond, G. Lapertot, D. Aoki
SPSMS, UMR-E CEA / UJF-Grenoble 1, INAC, 38054 Grenoble, France

Outline

- ✓ The heavy fermion compound URu_2Si_2 and its mysterious Hidden order
- ✓ Introduction to Raman Scattering
- ✓ Raman scattering study of lattice dynamic of URu_2Si_2
- ✓ Raman Electronic signal in URu_2Si_2 compound
  - Look for a gap opening
  - CEF excitation at high energy
- ✓ Conclusion and outlook

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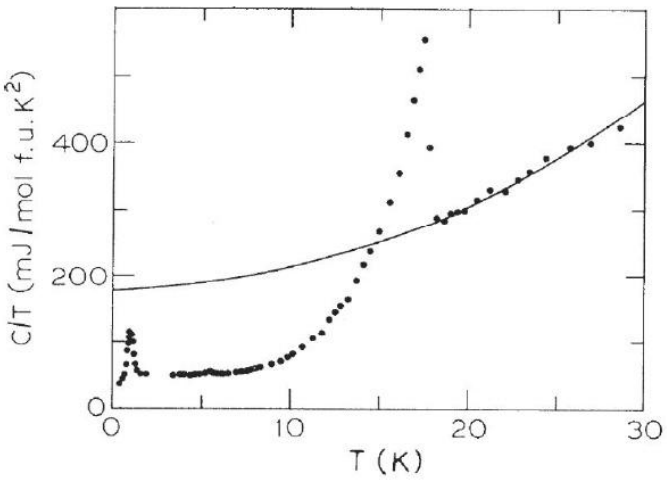
First measurements in 1985



Palstra *et al.* PRL (1985)

Two transitions:

- ✓ At 17.5K (2nde order) → Hidden Order (HO)
  Large entropy vararіation (ΔS = 0.3Rln2)

- ✓ At 1.5K → SC

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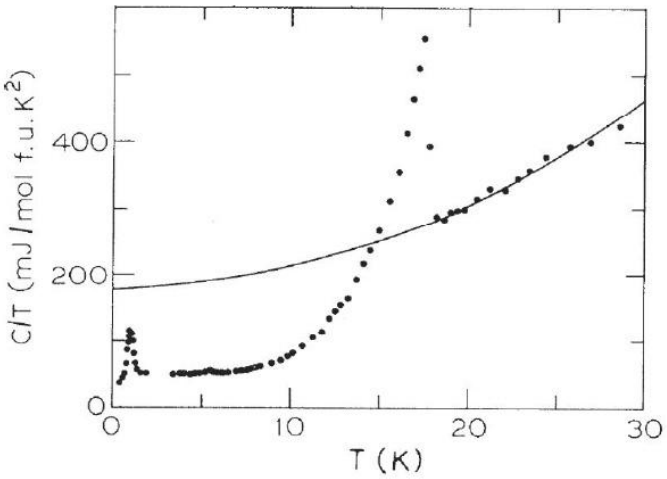

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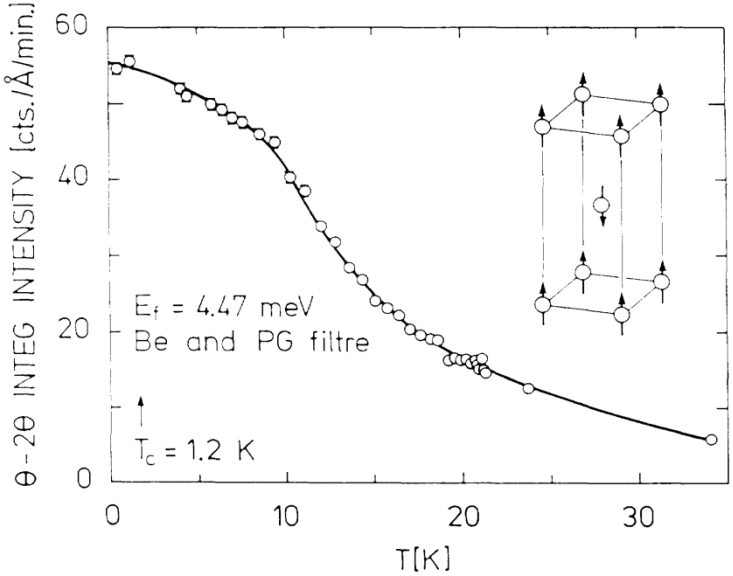
Broholm *et al.* PRL (1987)

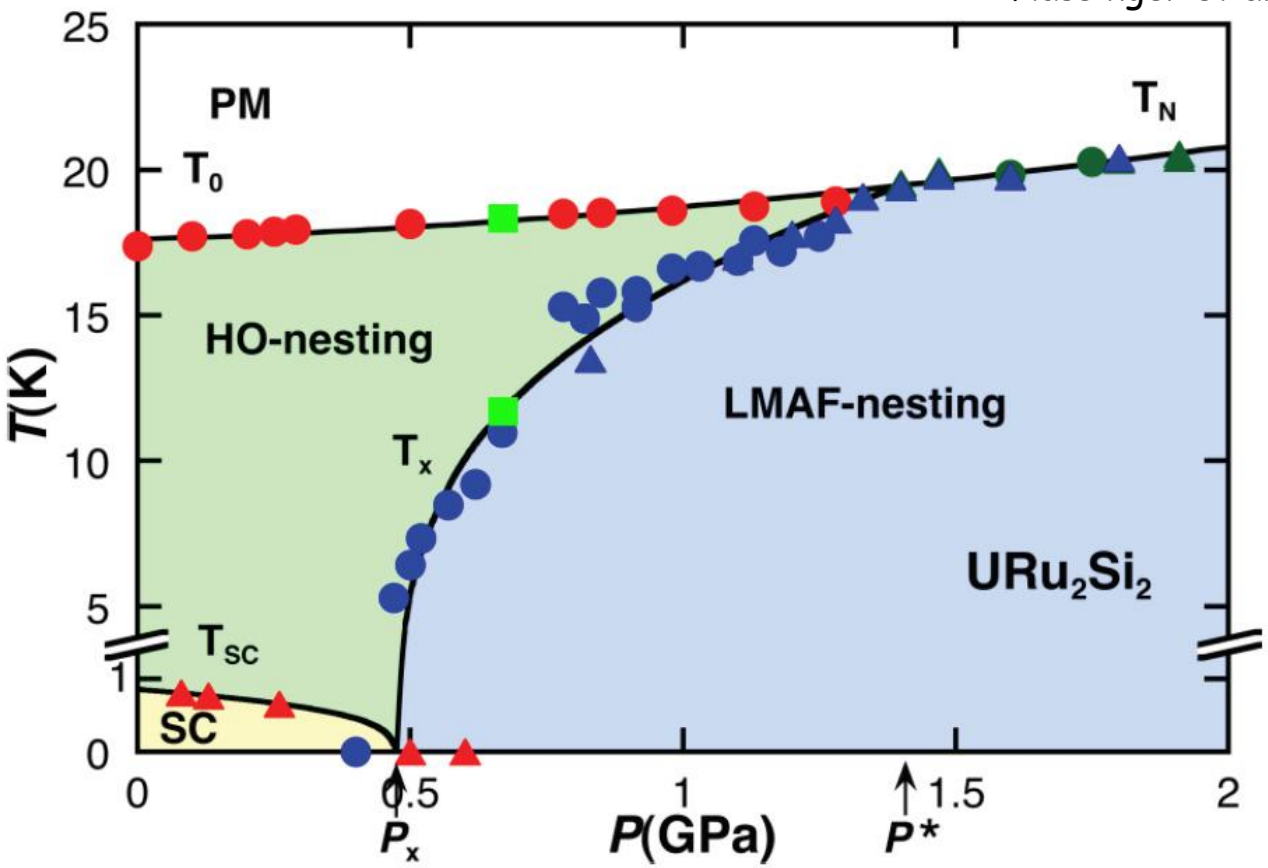


$m_0 \approx 0.03\ \mu_B/U$

- ✓ Too small to be order parameter
- ✓ Defects or stacking faults → not intrinsic!

P-T phase diagram of URu_2Si_2 compound

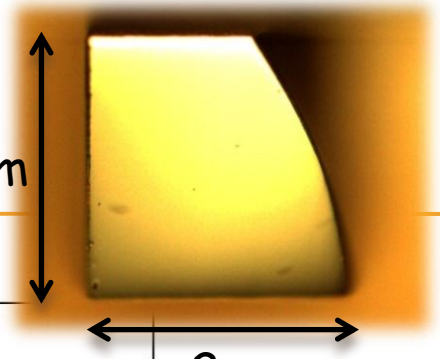
Hassinger *et al.* PRB (2008)

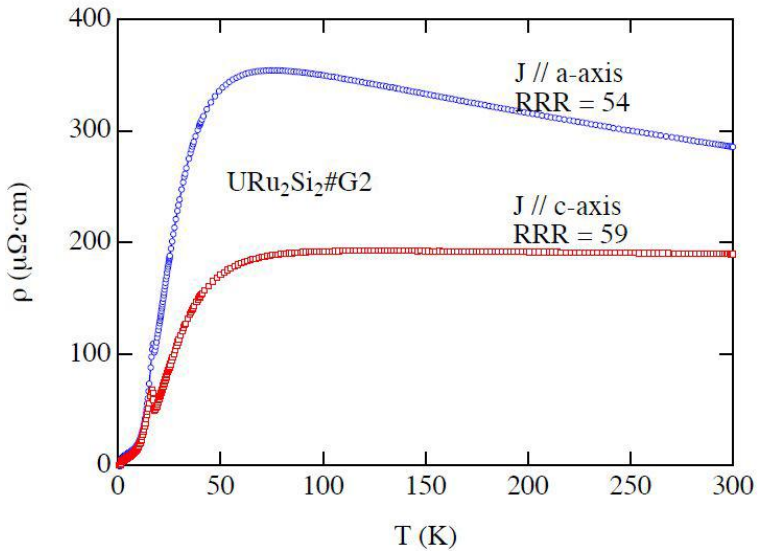


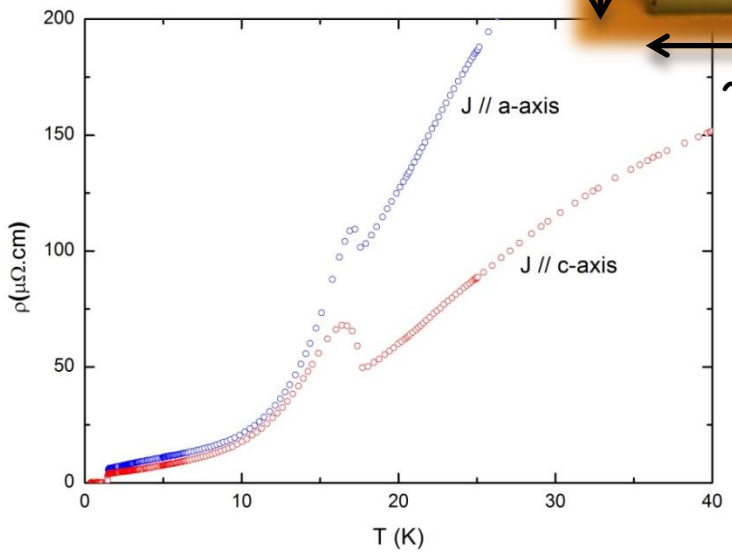
✓ Several theoretical survey:

Localized 5f electrons vs itinerant 5f electrons

Resistivity measurement: sample quality?





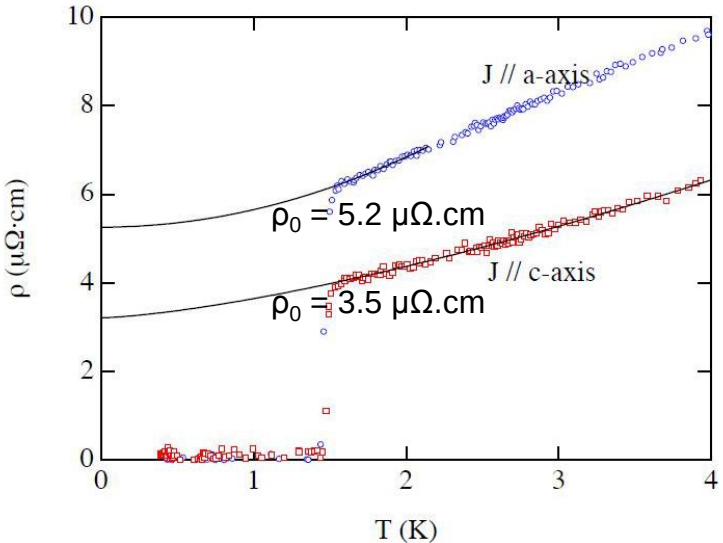


- ✓ <u>Several energy scales:</u>

T_0 = 17.5K, T_C = 1.5K and T_{KL} = 70K

- ✓ Low residual resistivity

High quality sample



Optical spectroscopy

Raman scattering studies

For 25 years, only two studies…

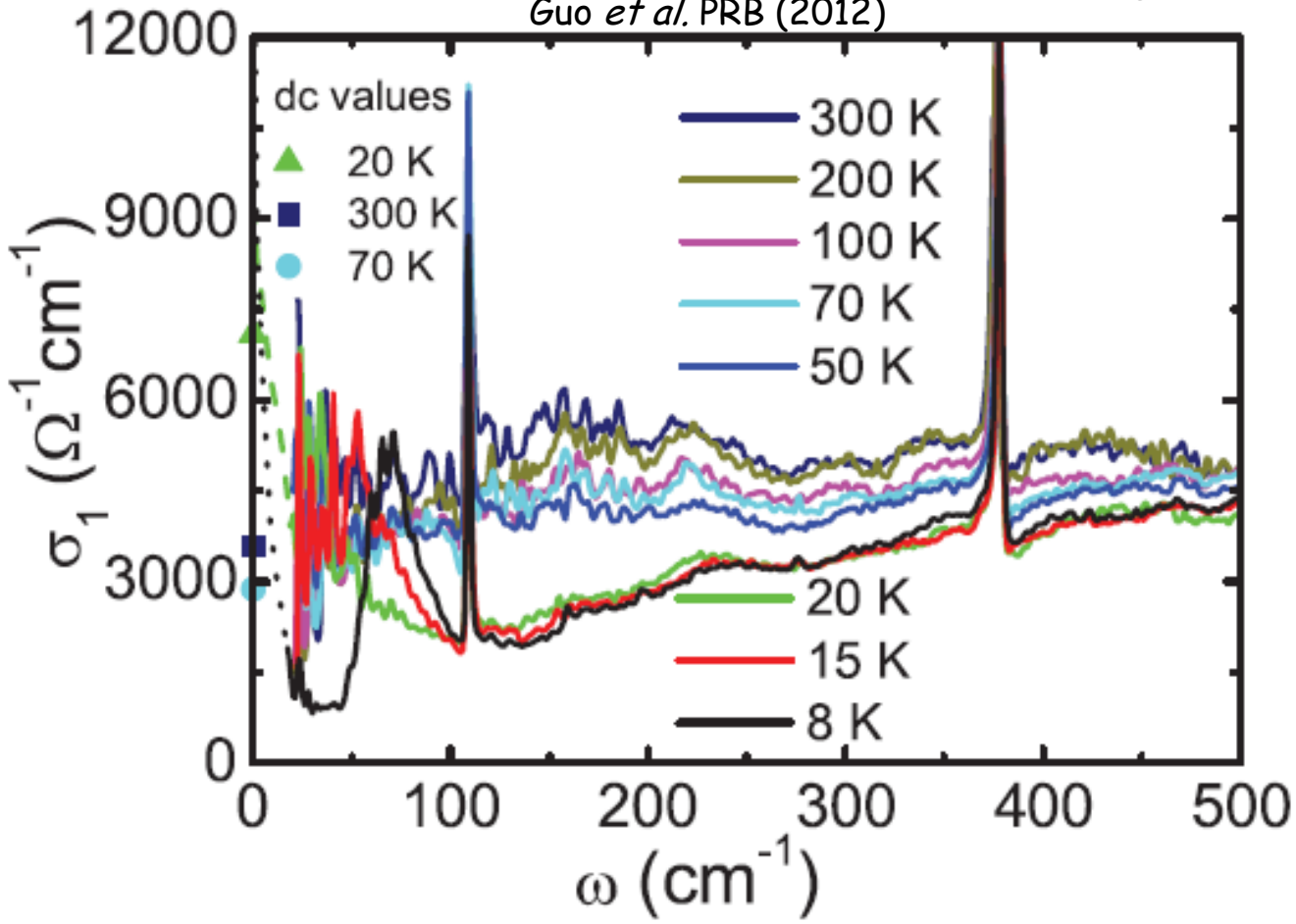
Cooper *et al.* PRB (1987)

Lampakis *et al.* Physica B (2006)

- ✓ Never studied at very high energy (>600cm^{-1})
- ✓ No complete study of the electronic Raman signal in all symmetries

Optical conductivity

Guo *et al.* PRB (2012)



See also: Bonn *et al.* PRL (1988)
Levallois *et al.* PRB (2011)
Nagel U *et al.* PNAS (2012)

✓ <u>Two gaps:</u> → Below 400 cm^{-1}, **below ~50K (T_{KL})** → hybridization-gap
→ At ~40 cm^{-1} , **below T_0**

Motivations of our study by Raman spectroscopy

- ✓ Investigate all symmetries for a wide energy range (up to 4000cm^{-1})
- ✓ Signature of the hidden order transition in Raman scattering?
- ✓ Opening of a gap in Raman spectroscopy? Symmetry?
- ✓ Look for CEF excitations (theory: multipolar order)

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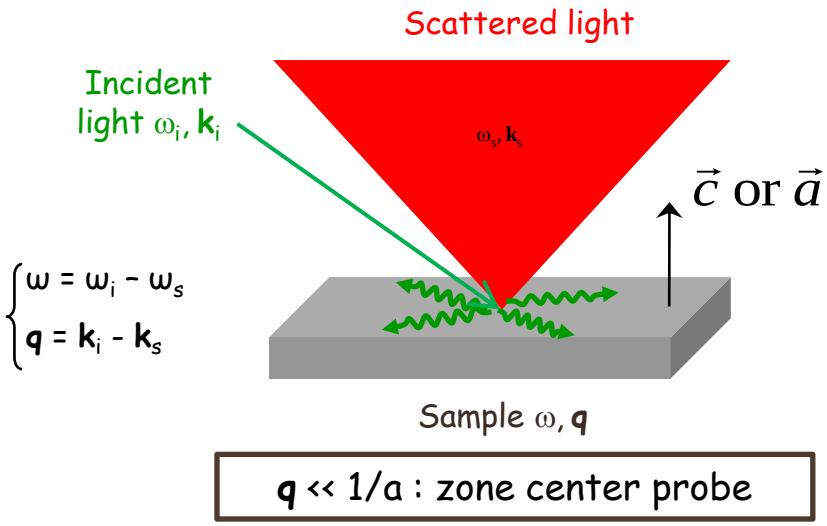
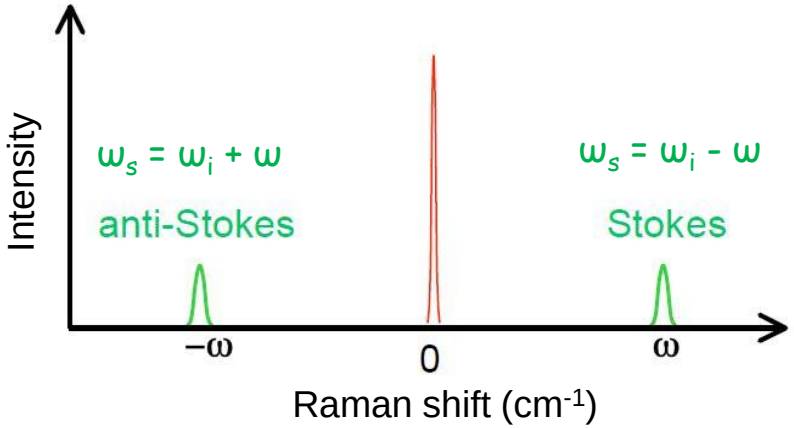
Inelastic scattering of monochromatic light





NB: 8 cm^{-1} = 1 meV

Raman intensity measured:

$$I_s \propto \frac{d^2\sigma}{d\Omega d\omega_S} = \frac{V\omega_I\omega_S^3}{(4\pi)^2c^4}\left|\hat{\mathbf{e}}_S \cdot \frac{d\chi(\omega)}{d\xi} \cdot \hat{\mathbf{e}}_I\right|^2 \langle\xi(\mathbf{q})\xi(\mathbf{q})^*\rangle_\omega$$

Differential cross-section

Raman Tensor → selection rules (Group Theory)

Correlation function of observable (phonons, magnons,…)

Selection rules for URu_2Si_2

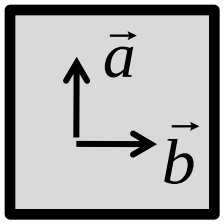
URu_2Si_2 → Space group: I4/mmm
Point group: D4h

Raman active Tensors:

$$A_{1g} \oplus B_{1g} \oplus 2E_g \oplus B_{2g} \oplus A_{2g}$$

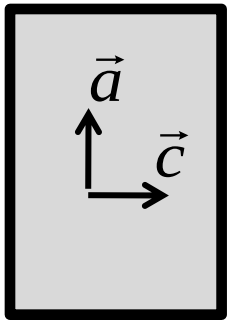
Active phonons

Characteristic of magnetic scattering



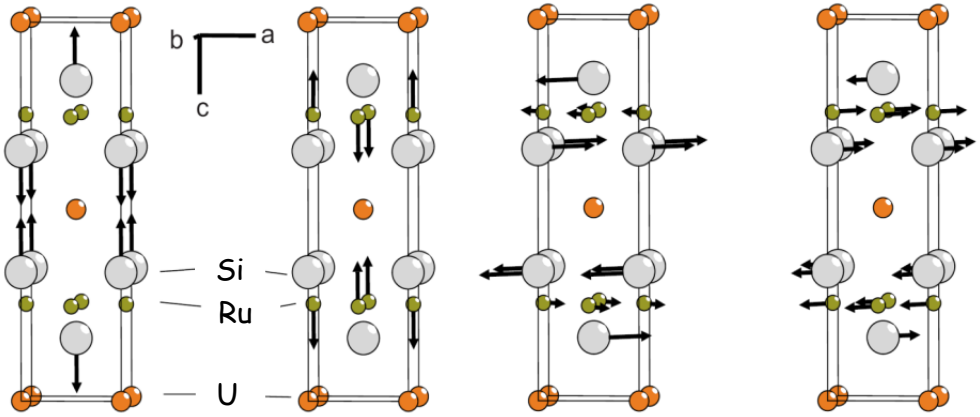
c(aa)c : A_{1g} + B_{1g}

c(ab/ab)c : A_{2g} + B_{1g}



b(aa)b : A_{1g} + B_{1g}

b(ac)b : E_g



A_{1g} B_{1g} E_g E_g

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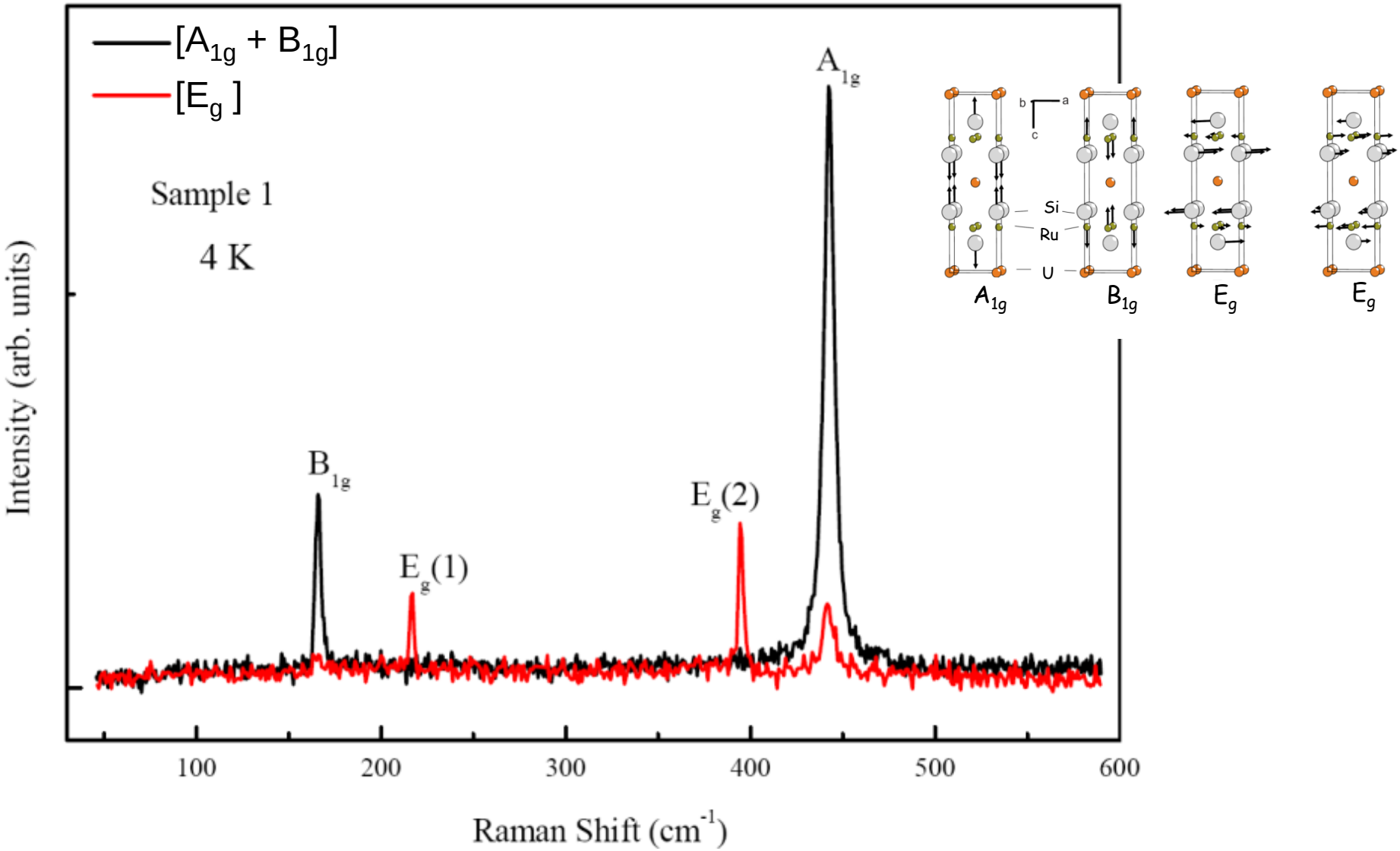
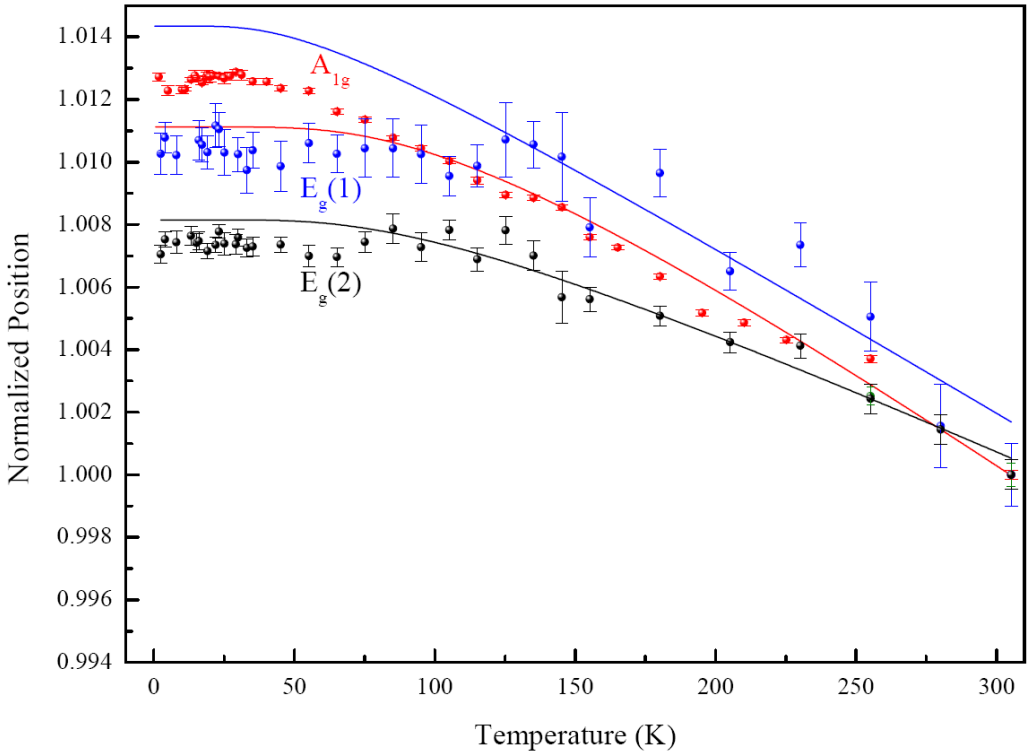
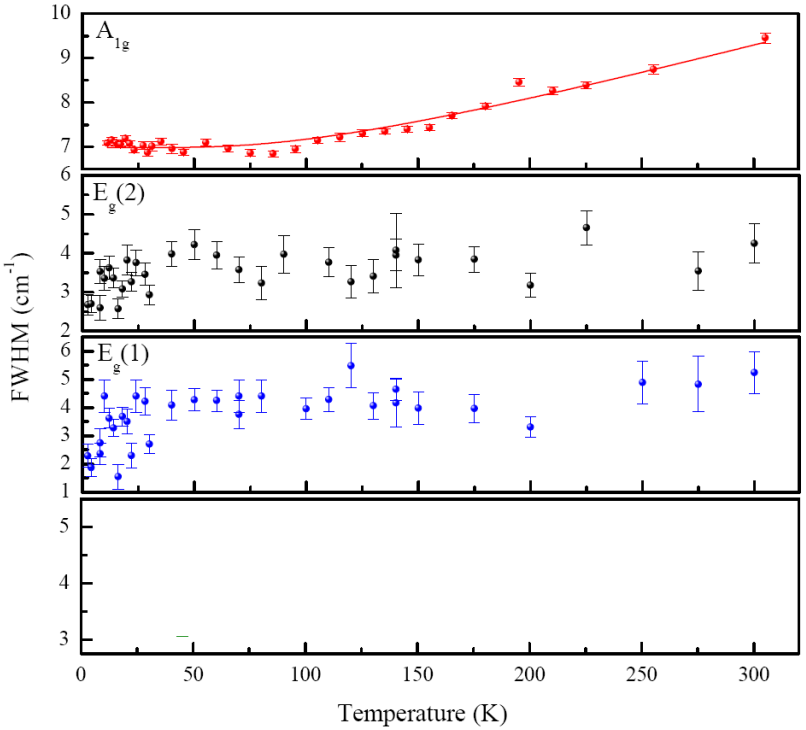
Lattice Dynamic



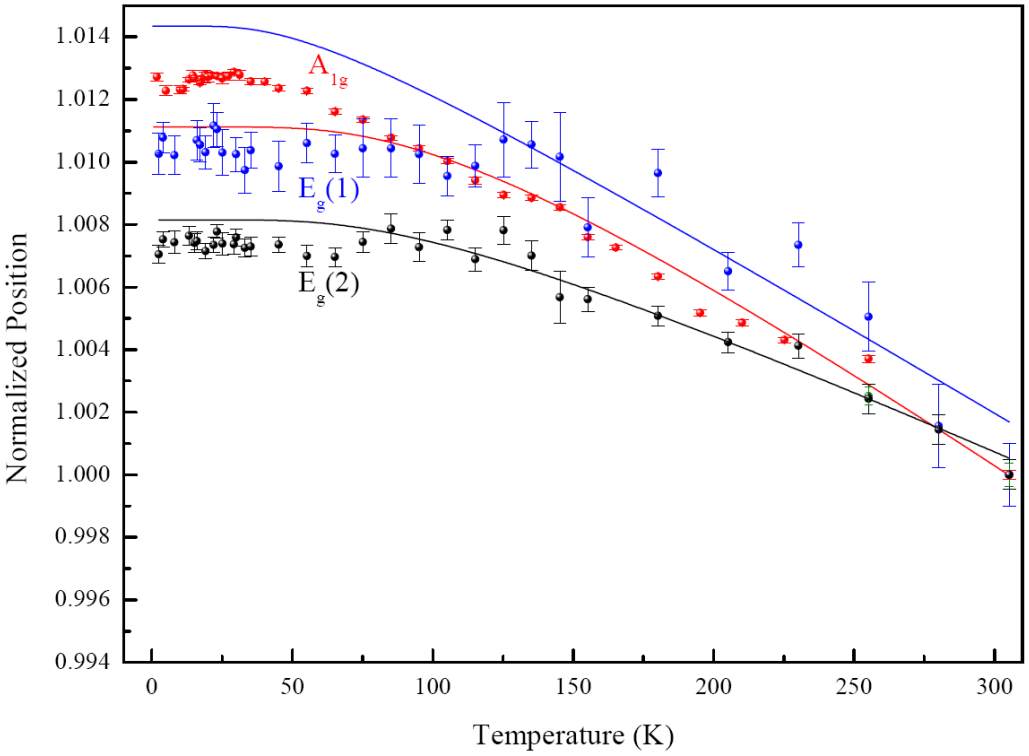
Temperature dependence of phonon modes

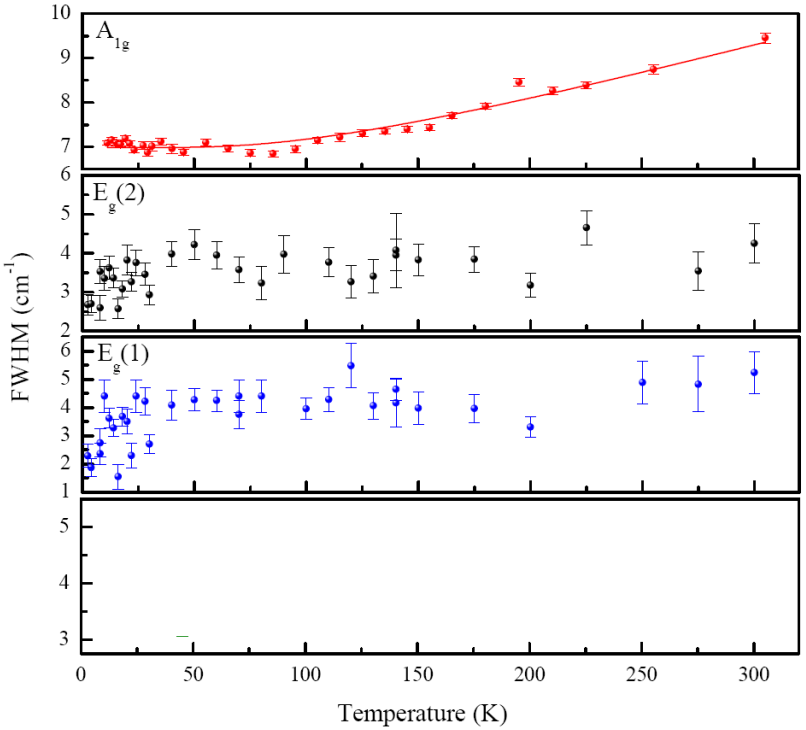




- ✓ No anomaly at T_0 for A_{1g} and E_g modes
- ✓ Position of A_{1g} and E_g modes → Classical behavior vs Temperature.

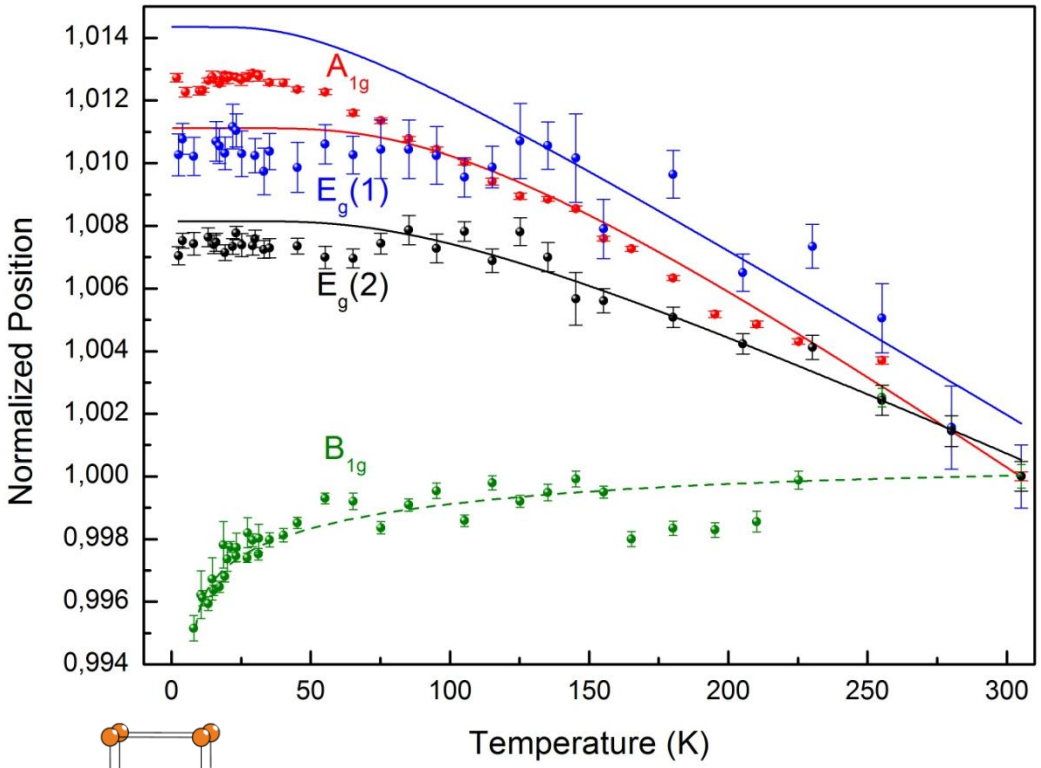
Temperature dependence of phonon modes

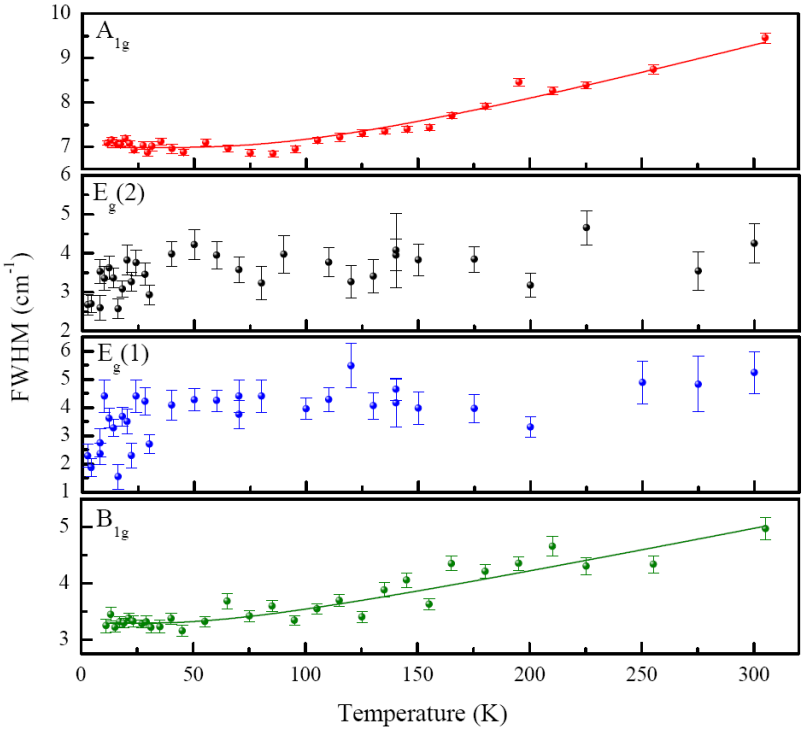


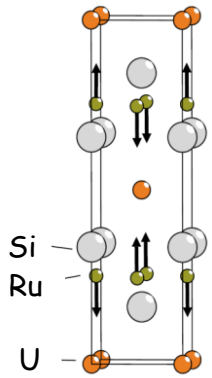


- ✓ No anomaly at T_0 for A_{1g} and E_g modes
- ✓ Position of A_{1g} and E_g modes → Classical behavior vs Temperature.
- ✓ FWHM of E_g: Slight decrease below ~30K → Electron-phonon coupling below T_{KL}?
  Hybridization-gap seen by optical conductivity?

Temperature dependence of phonon modes







- ✓ **Position of B_{1g} mode unusual: almost constant and softens below ~50K (T_{KL})**
- ✓ **No effect on FWHM**

Origin of the B_{1g} mode softening

- ✓ Electron-phonon coupling ?
- ✓ Magneto-elestic coupling ?
- ✓ Anharmonic effects ?

Origin of the B_{1g} mode softening

- ✓ Electron-phonon coupling ? → **No effect on FWHM**
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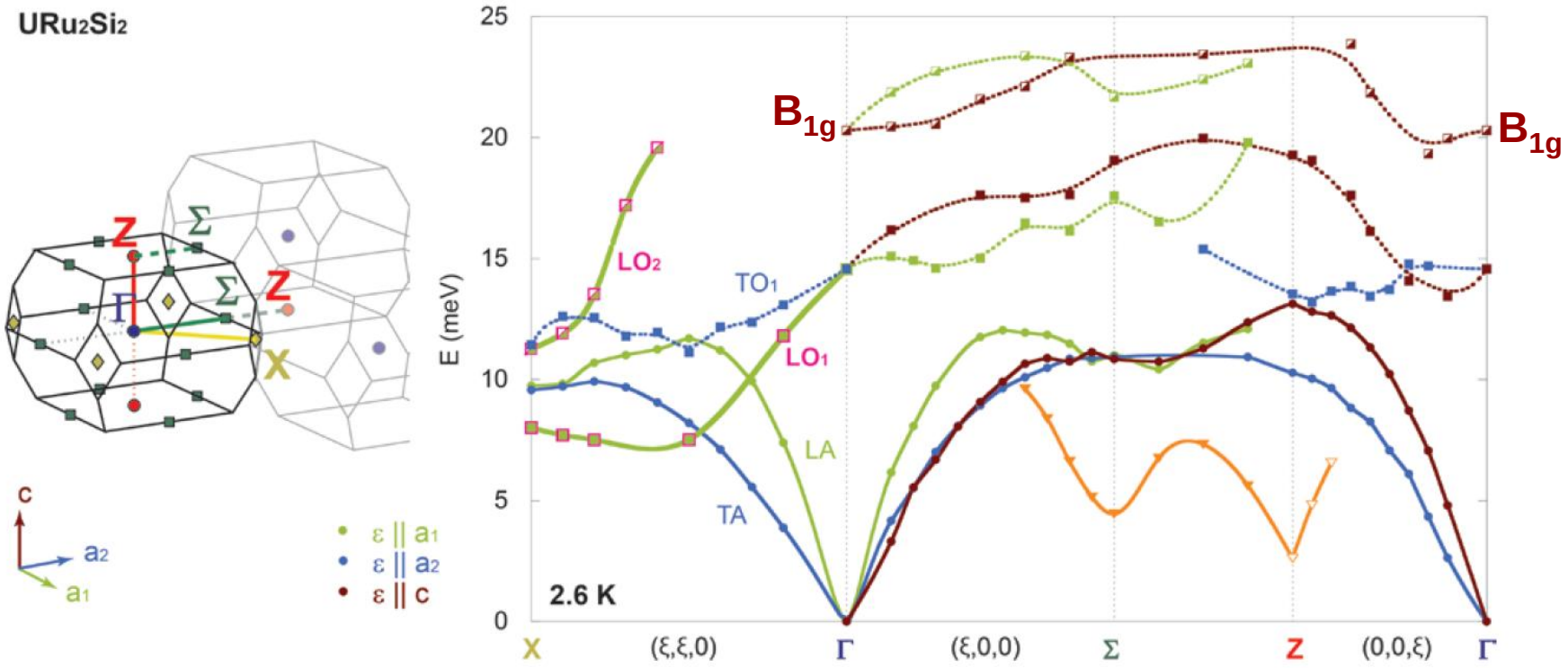
Soft lattice dynamics

Butch *et al.* arXiv:1212.6238 (26 Dec 2012)



- ✓ LO modes softening in the [110] direction in HO phase
- ✓ Not originate from simple magneto-elastic coupling

Origin of the B_{1g} mode softening

- ✓ Electron-phonon coupling ? → **No effect on FWHM**
- ✓ Magneto-elestic coupling ? → **Magnetic excitations do not affect phonon branch**
- ✓ Anharmonic effects ?

Comparison with theoretical calculation and neutron scattering

✓ Phonons dispersion curves calculated (P. Piekarz)

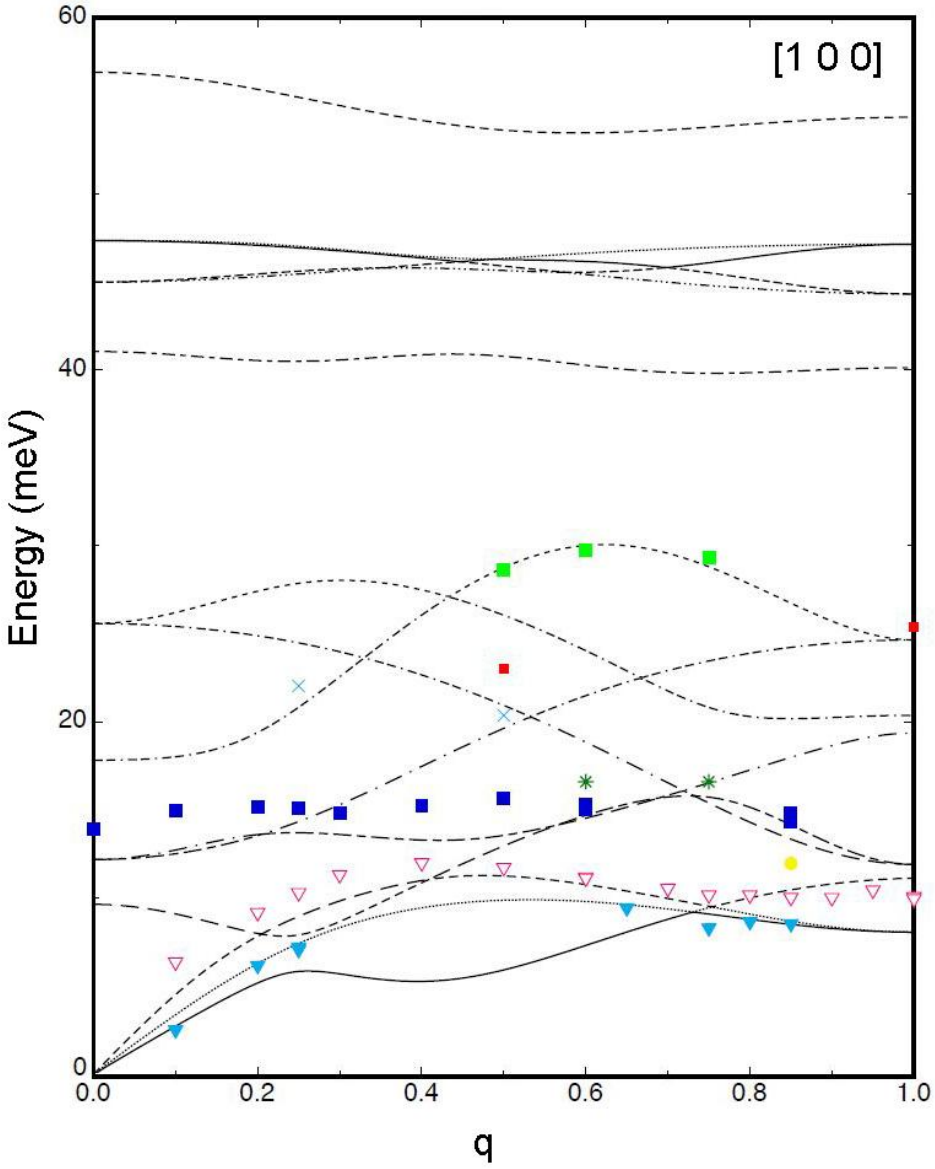
DFT + Direct method

K. Parlinski Collection SFN 12 161 (2011)

✓**Harmonic approximation**

✓ T = 0 K

✓ Neutron measurement (IN8 ILL Grenoble, F. Bourdarot, S. Raymond and J. Buhot)



Raman vs theoretical calculation

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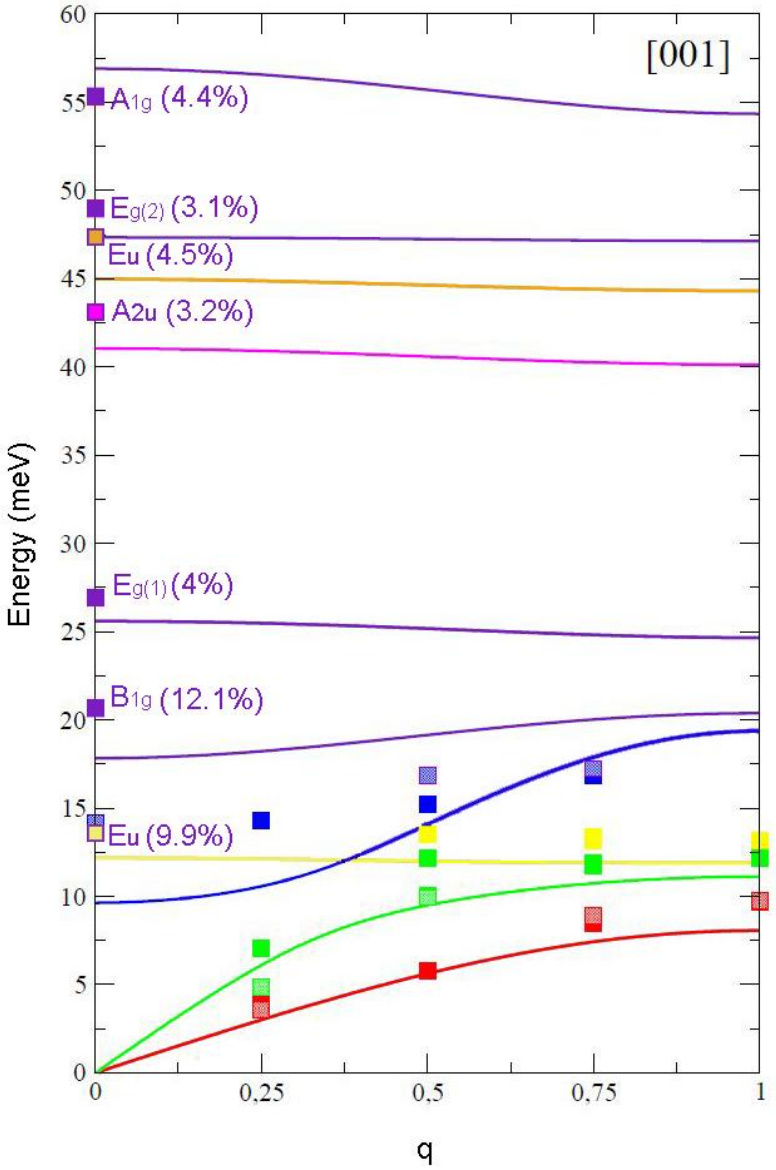

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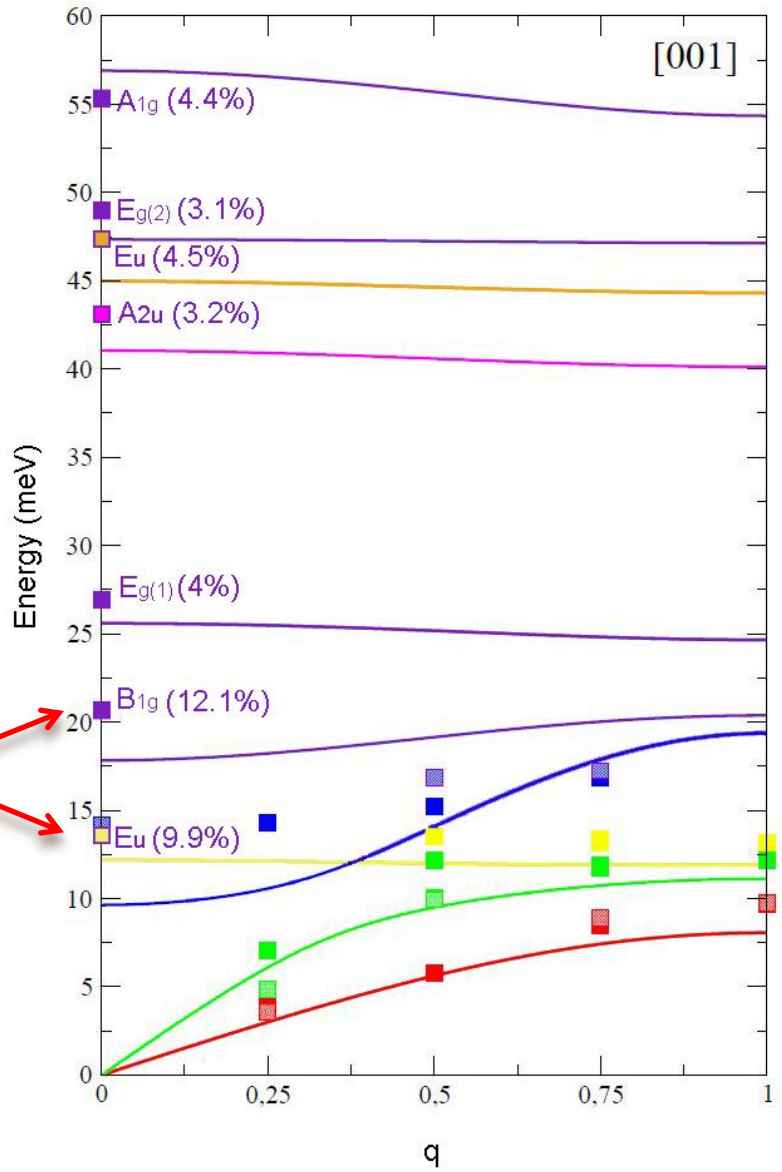
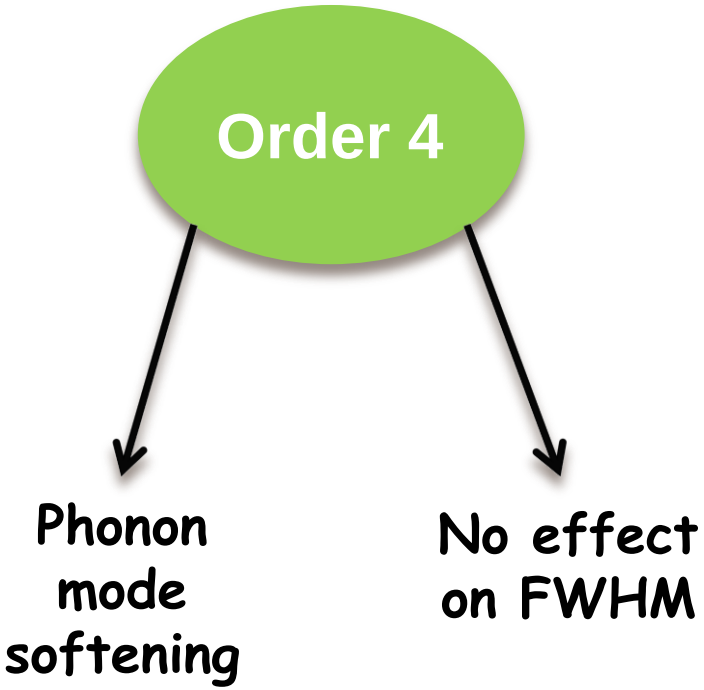


✓ Difference with calculation are largest for low energy optical modes.

Anharmonicity and Lattice parameter

For example:



✓ Repeating calculations at Γ point **including anharmonic effects**

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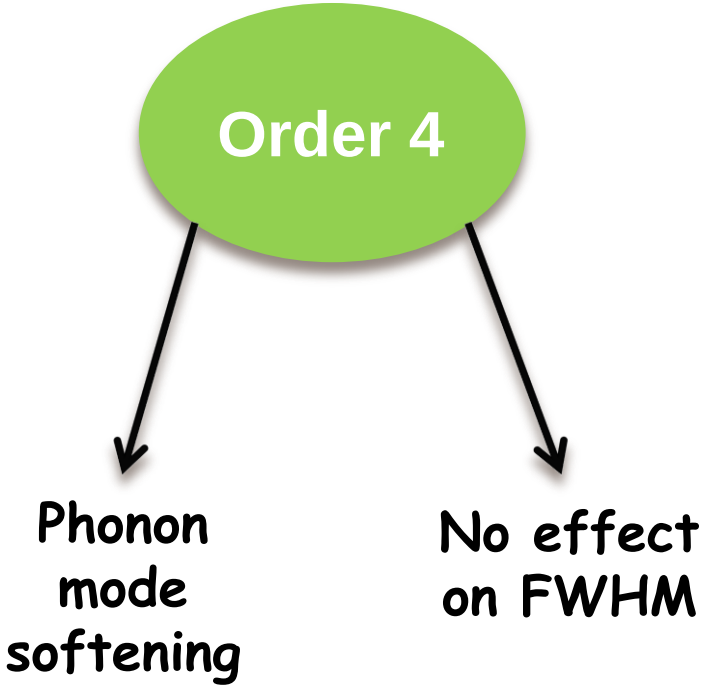
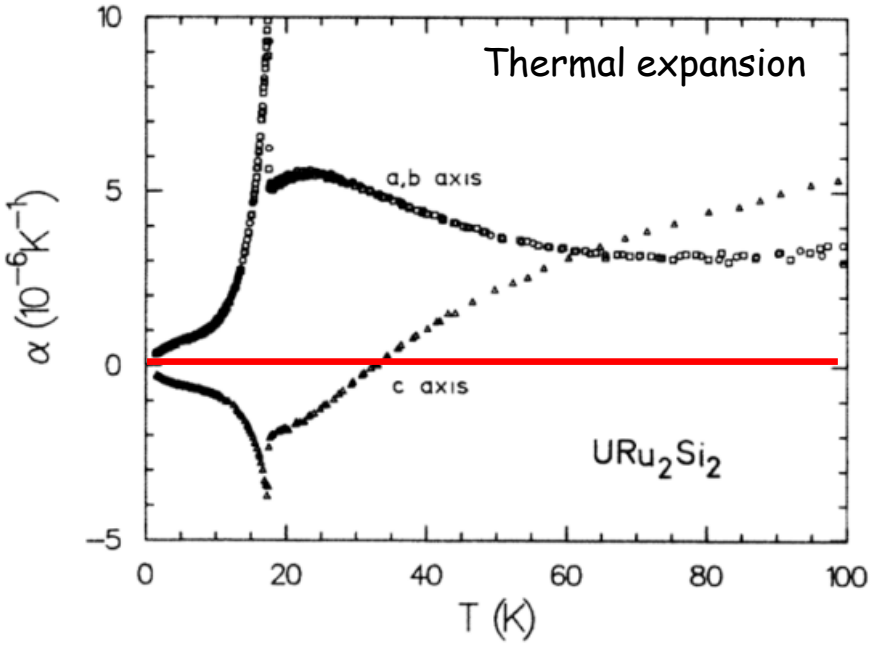


De Visser *et al.* PRB 1986



✓ Atomic position required (Amitsuka)

✓ Repeating calculations at Γ point **including anharmonic effects**

Origin of the B_{1g} mode softening

- ✓ Electron-phonon coupling ? → **No effect on FWHM**
- ✓ Magneto-elestic coupling ? → **Magnetic excitations do not affect phonon branch**
- ✓ Anharmonic effects ? → **To be checked with calculation**

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- ✓ Anharmonic effects ? → **To be checked with calculation**
- ✓ Coupling with another degree of freedom ?

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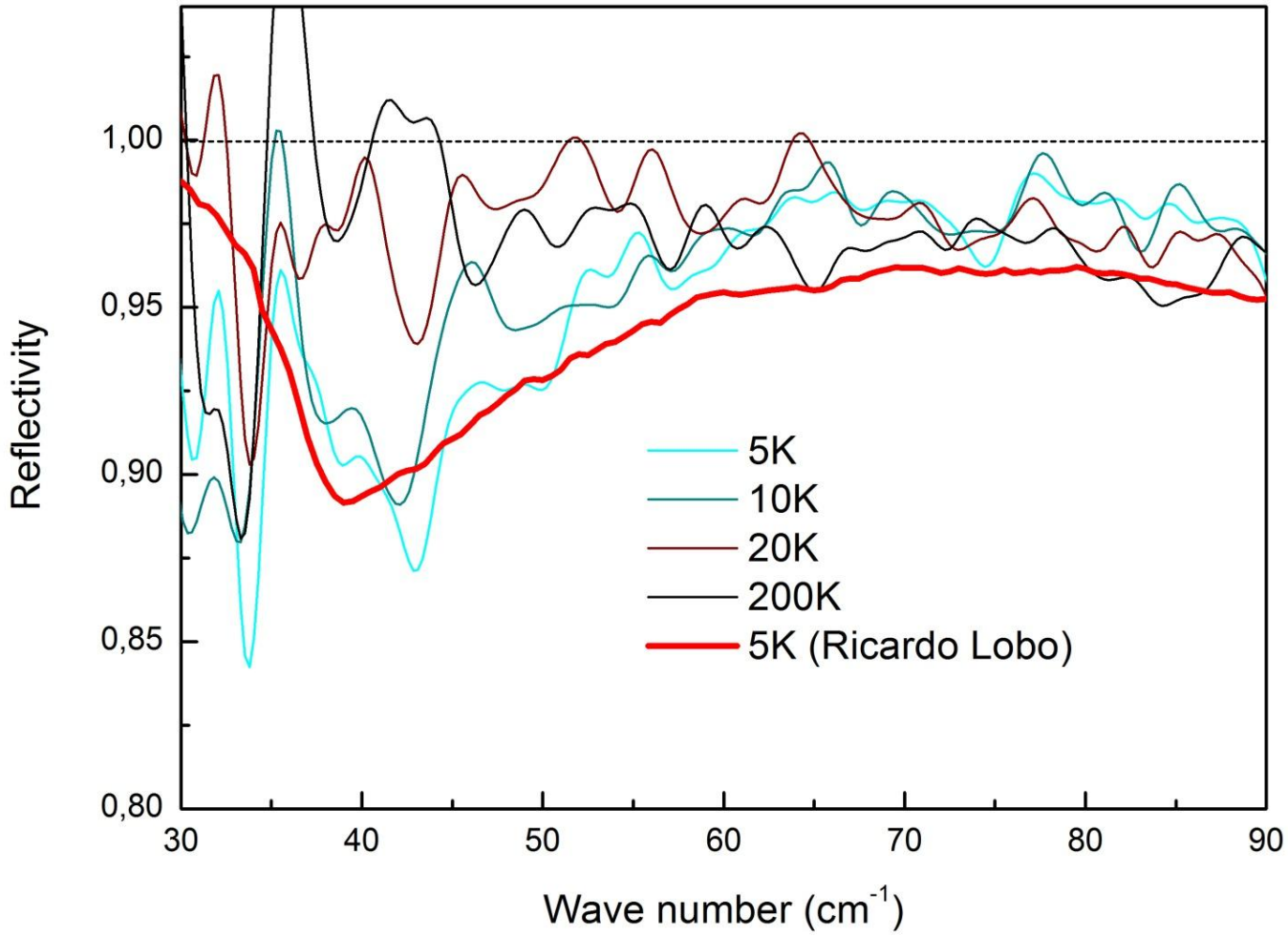
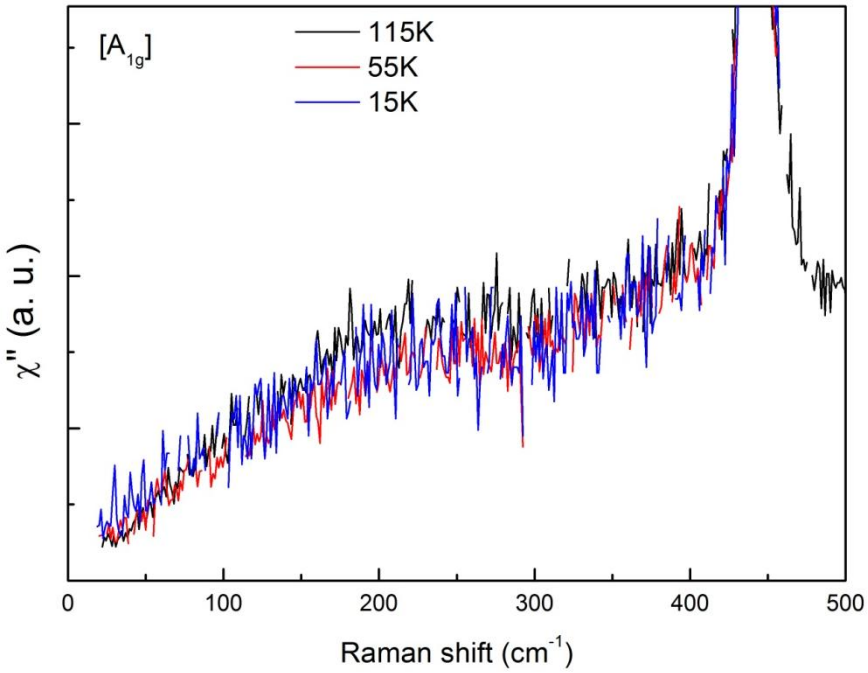
Optical far-infrared reflectivity of our sample

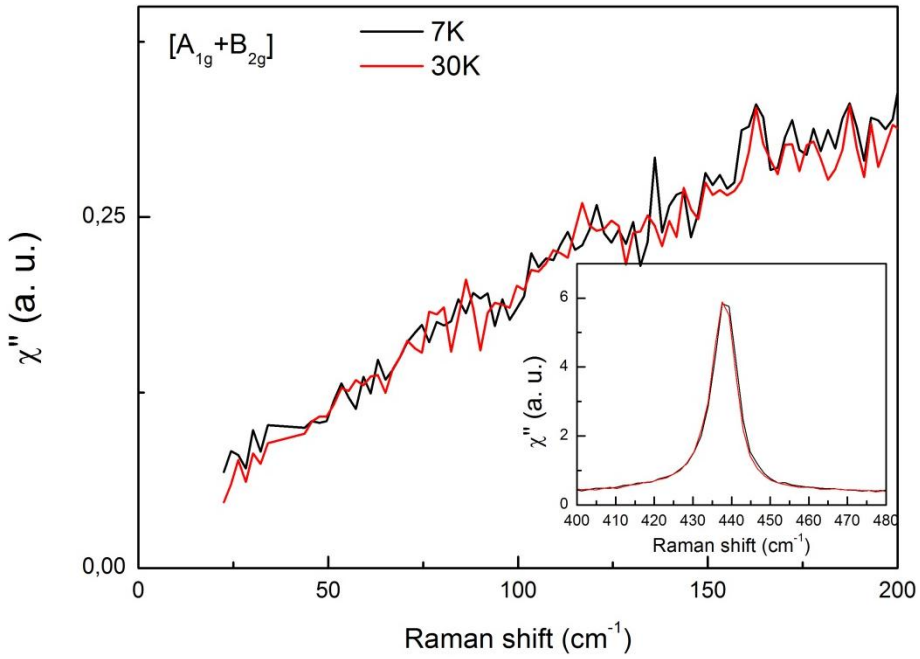
✓ Performed at ESPCI (R. Lobo, J. Buhot)



Look for a gap opening

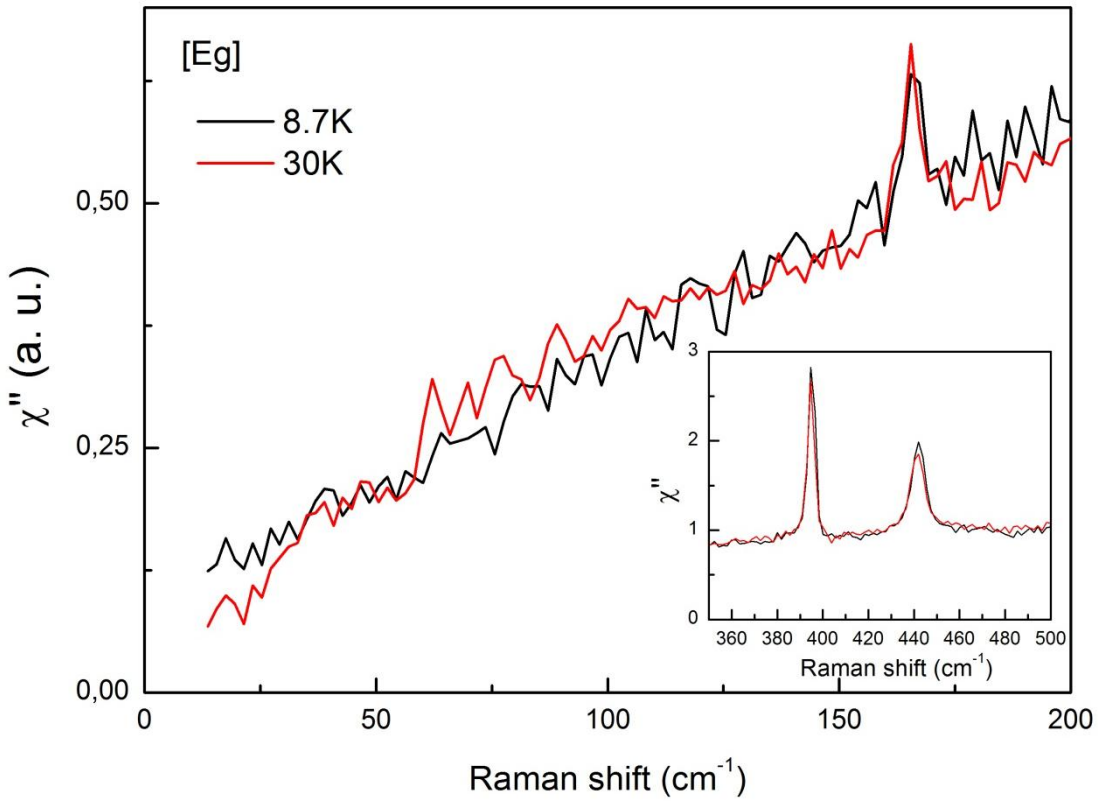
The case of A_{1g} symmetry…





✓ **For all symmetry**, no gap opening below 50K and below T_0

Look for a gap opening in E_g symmetry



- ✓ No clear gap opening below T_0
- ✓Slight **increase at low frequency** below 30K, to be checked

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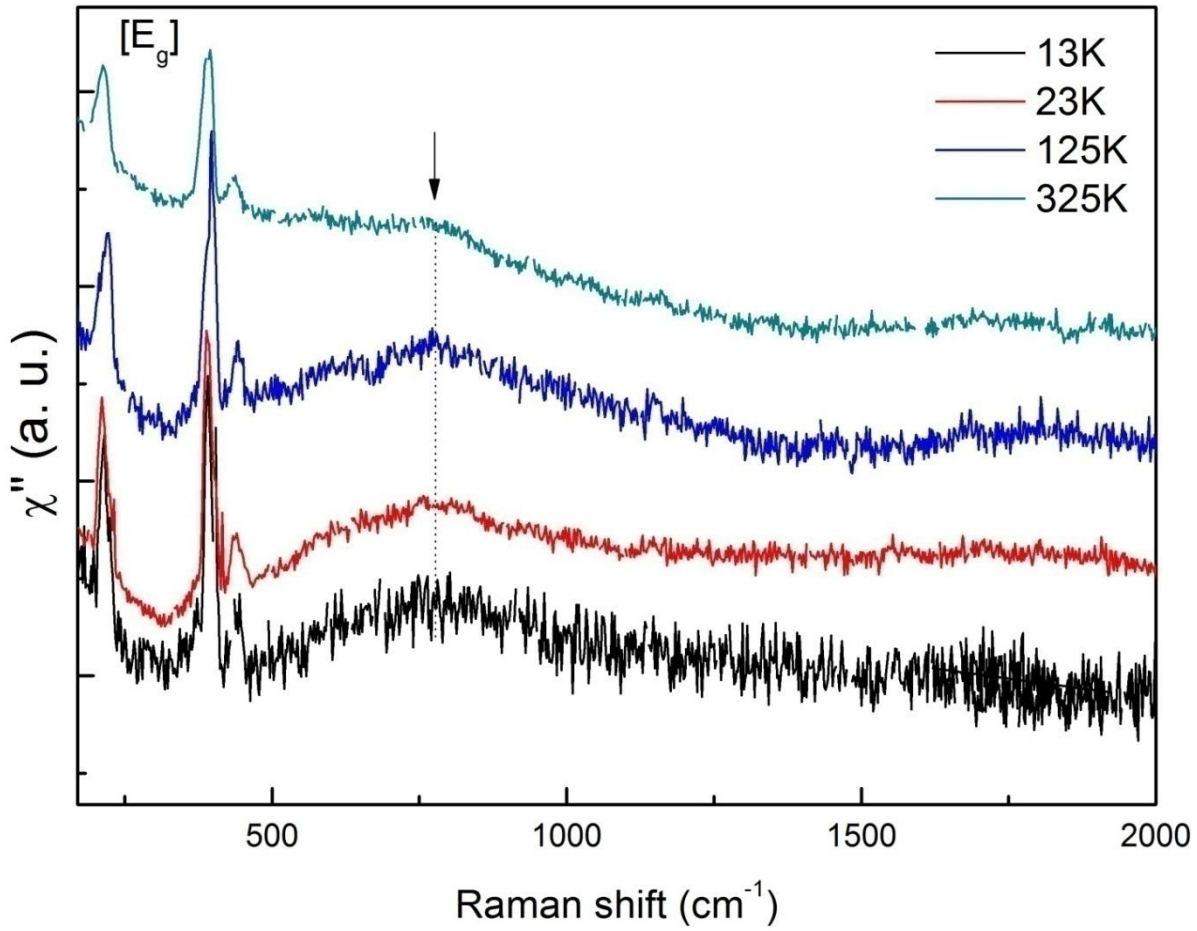
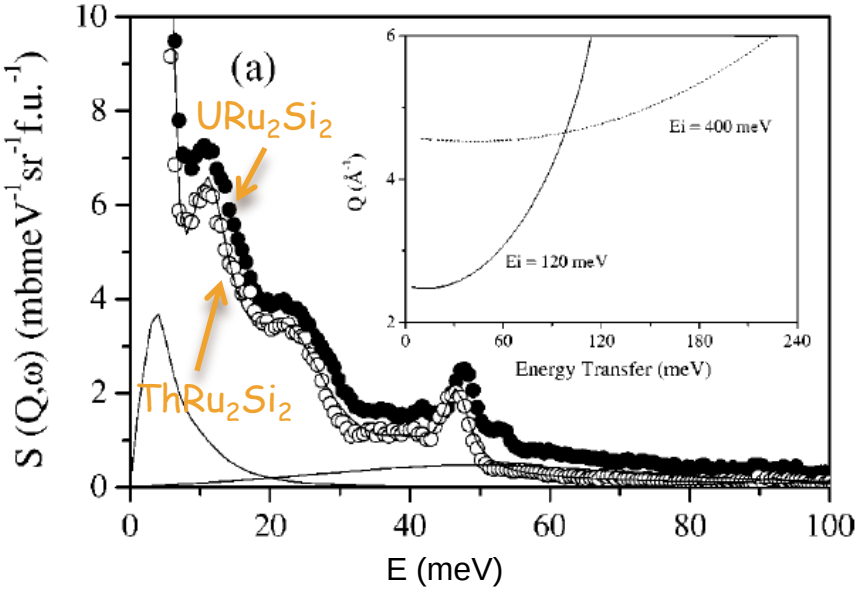
CEF excitation in E_g symmetry

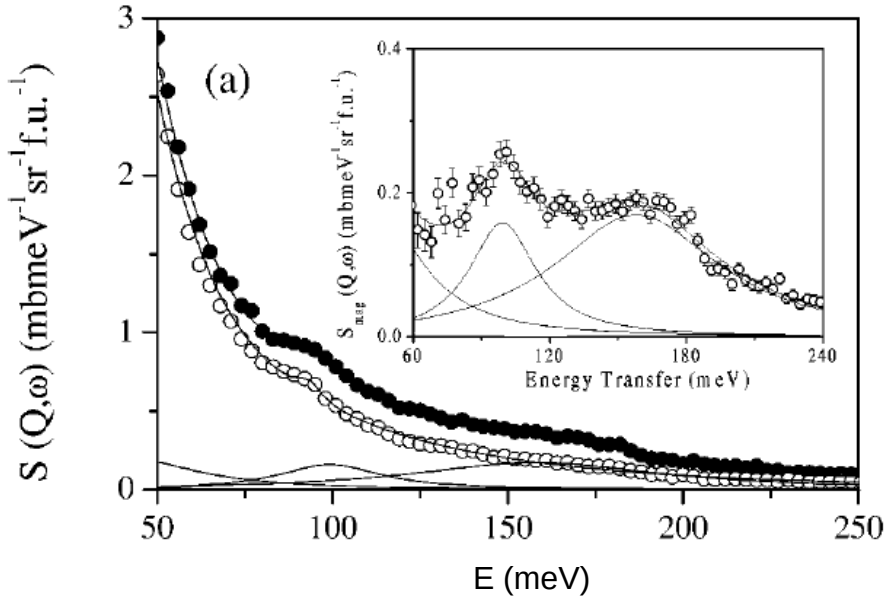
- ✓ New broad peak **at 770 cm^{-1}** (FWHM ~300-700 cm^{-1})
- ✓ Too large to be connected at a double phonon process



Comparison with neutron measurement

Park *et al.* PRB 66, 094502 (2002)





Energy (cm^{-1})	FWHM (cm^{-1})	CEF transition
395 ± 8	516 ± 16	$\Gamma_3 \rightarrow \Gamma_5^{(1)}$
798 ± 8	**290 ± 16**	$\Gamma_3 \rightarrow \Gamma_4$
1274 ± 8	717 ± 16	$\Gamma_3 \rightarrow \Gamma_5^{(2)}$

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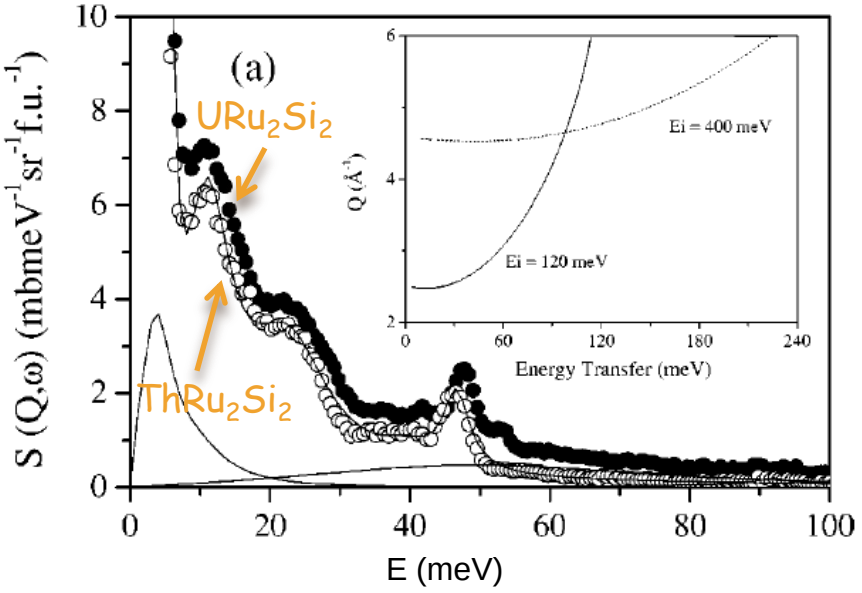

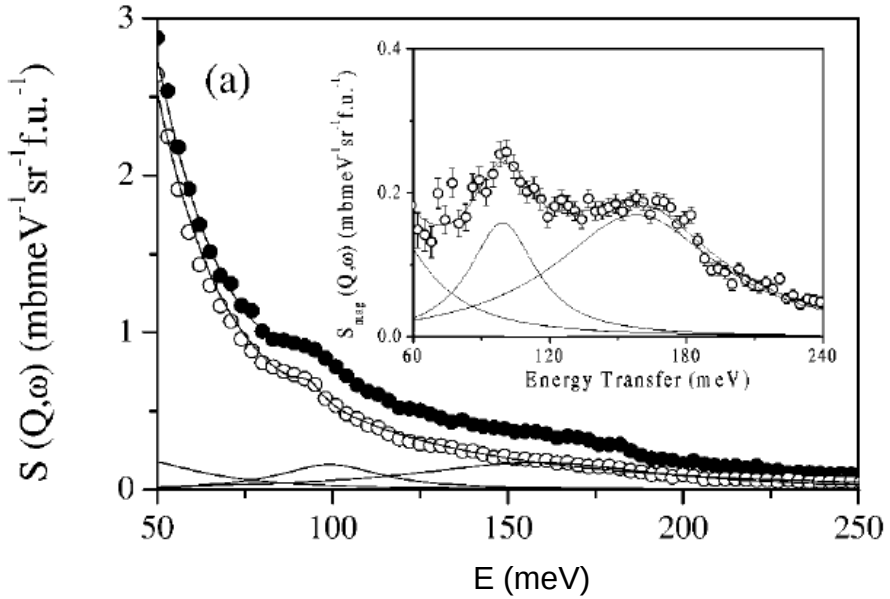

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1274 ± 8	717 ± 16	$\Gamma_3 \rightarrow \Gamma_5^{(2)}$

Selection rules in Raman scattering for CEF transitions

- ✓ CEF visible in E_g symmetry:

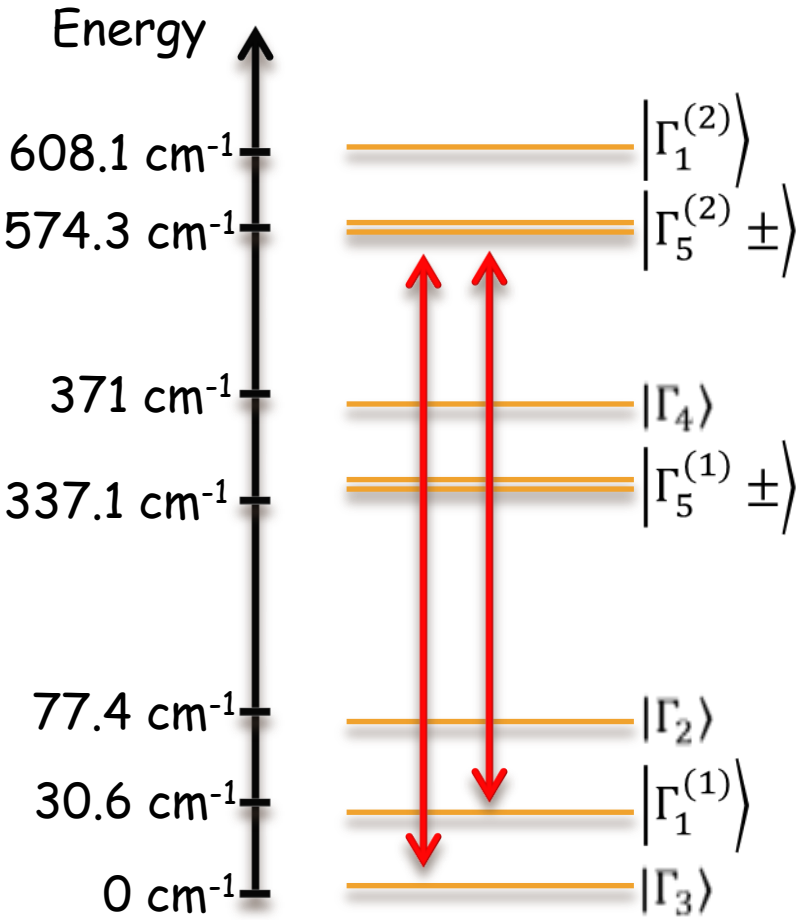
$$\left|\Gamma_5^{(1)} \pm\right\rangle \leftrightarrow \left|\Gamma_1^{(1)}\right\rangle \leftrightarrow \left|\Gamma_1^{(2)}\right\rangle \leftrightarrow |\Gamma_2\rangle \leftrightarrow |\Gamma_3\rangle \leftrightarrow |\Gamma_4\rangle$$

$$\left|\Gamma_5^{(2)} \pm\right\rangle \leftrightarrow \left|\Gamma_1^{(1)}\right\rangle \leftrightarrow \left|\Gamma_1^{(2)}\right\rangle \leftrightarrow |\Gamma_2\rangle \leftrightarrow |\Gamma_3\rangle \leftrightarrow |\Gamma_4\rangle$$

- ✓ **First direct measurement** of CEF transition
- ✓ New constrain for theoretical schemes (localized model)

Comparison with a complete theoretical CEF scheme

→ Few complete schemes with all CEF levels, particularly at high energy



Santini *et al.* PRL 73, 7 (1994)

✓ Two visible transitions in E_g symmetry can correspond to observed excitation at high energy

Conclusion

For lattice dynamic…

- ✓ **No anomaly at T_0** for A_{1g}, B_{1g} and E_g phonons modes
- ✓ Temperature dependence of the **B_{1g} phonon mode unusual**:

Anharmonic effect ?
Coupling with another degree of freedom (multipolar)?
…

Importance of the lattice dynamic!

For electronic signal…

- ✓ **No clear gap opening** below T_{KL} and T_0
- ✓ **CEF signature** at high energy: **new constrain for future theoretical schemes**

Outlook

- ✓ Repeating **calculations of phonon energies** at Γ point including anharmonic effects
- ✓ Study under **magnetic field (10 T):** splitting of CEF doublets
- ✓ **High Pressure** study through the HO to AF transition