The lattice dynamics of Cerium across the γ -> α transition

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Outline

- Motivation
- Main results (experiment, modeling, theory)
- Perspectives

Acknowledgments

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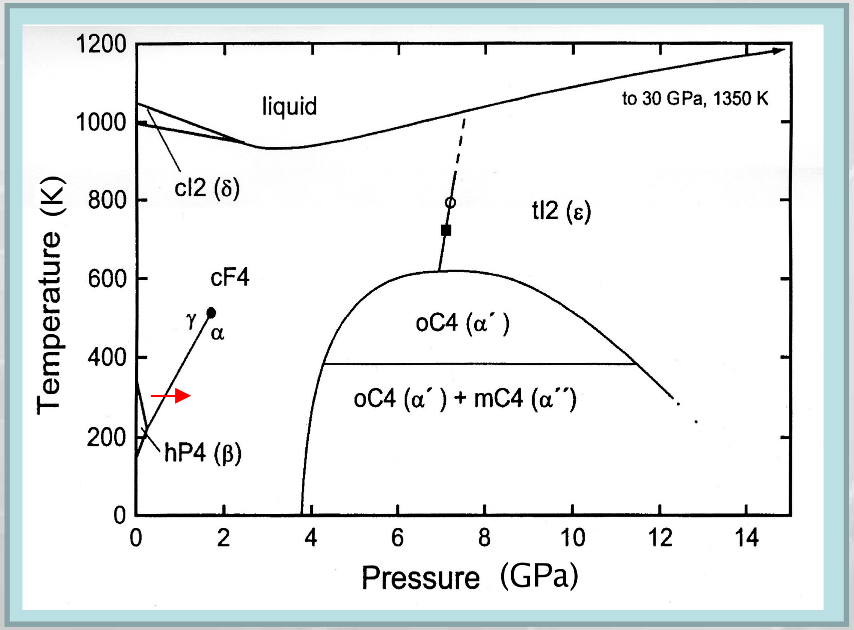
The γ -> α transition in metallic cerium



fcc – fcc transition

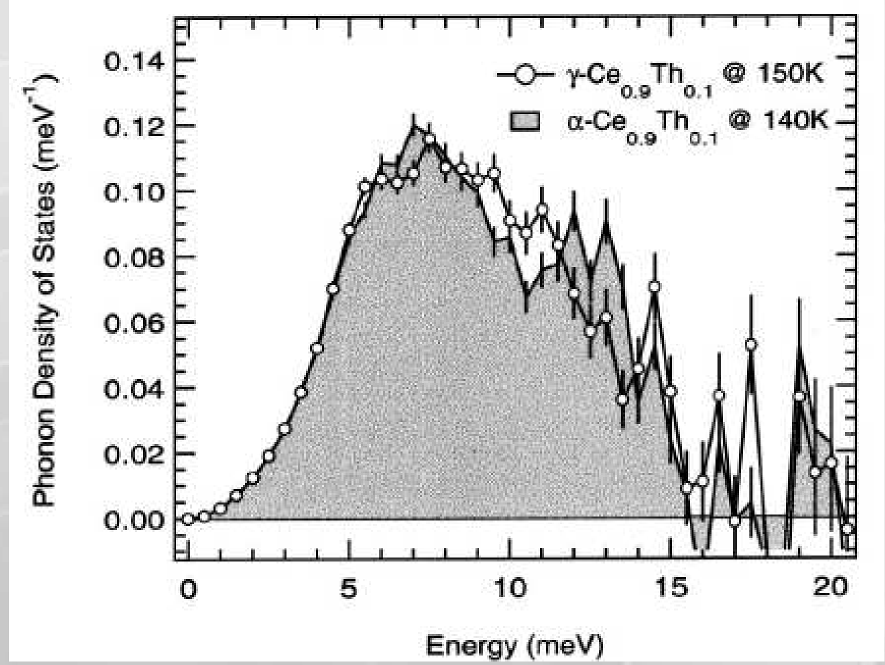
P_{trans} = 7.5 kbar at 300 K

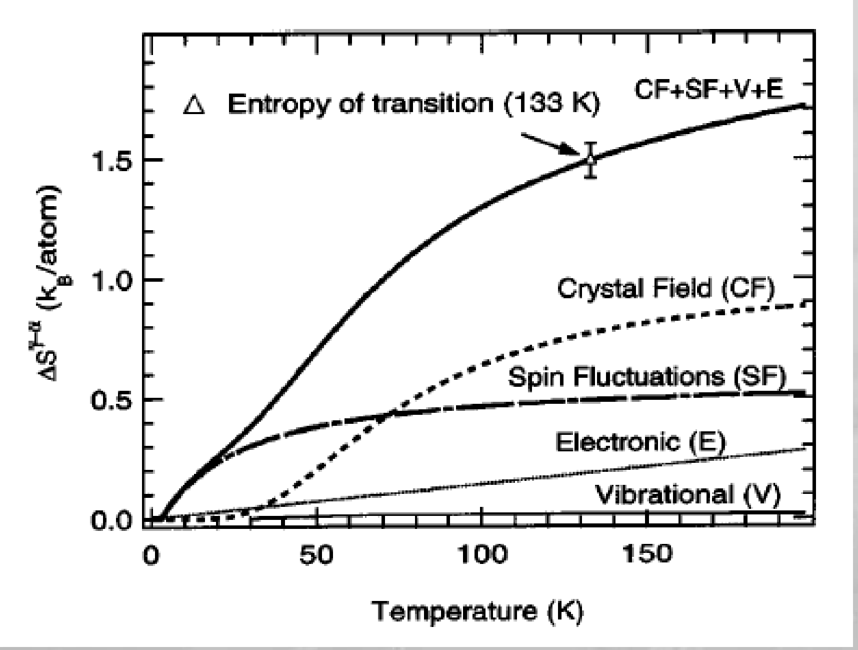
15% volume collapse

degree of localisation and correlation of the 4f electrons:
Mott transition scenario <-> Kondo volume collapse model

What about the lattice contribution?

Inelastic Neutron Scattering (INS)

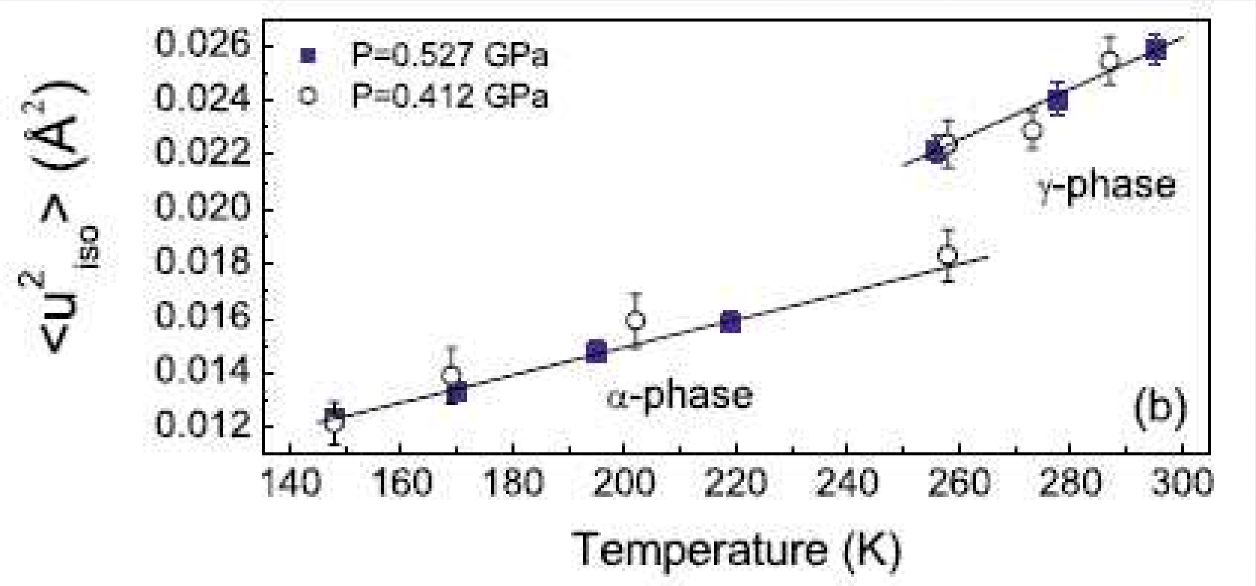




ΔS_{vib} negligible

M.E.Manley et al.; Phys. Rev. B (2003)

Neutron and x-ray diffraction



$$\Delta S_{vib}^{\gamma-\alpha} = \frac{3}{2} k_B \ln \frac{u_\gamma^2}{u_\alpha^2} = 0.7 k_B$$

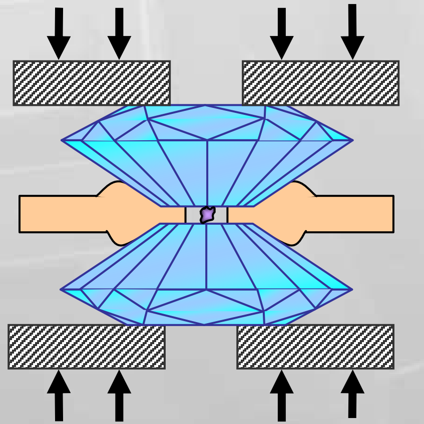
$$\Delta S_{tot} = 1.5 k_B$$

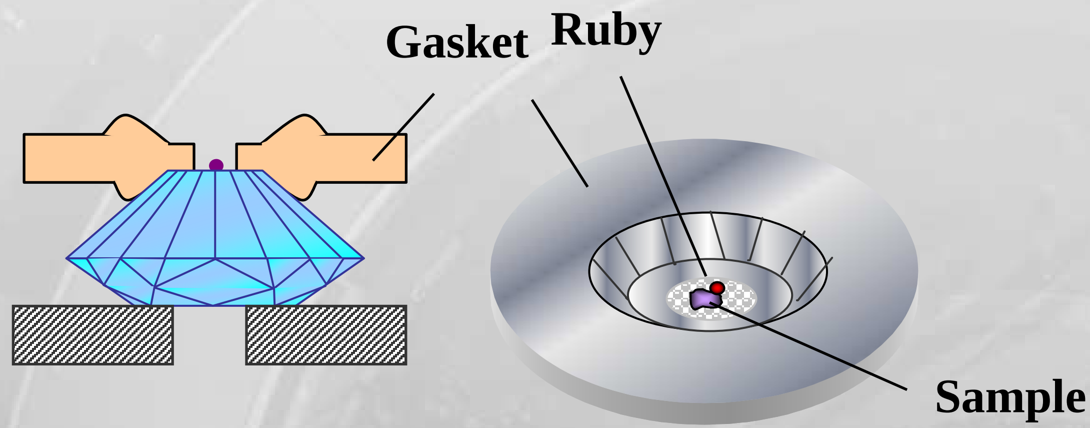
- Further evidence from high P – high T x-ray diffraction: M.J. Lipp et al.; Phys. Rev. Lett. (2008)

I.-K. Jeong et al.; Phys. Rev. Lett. (2004)

Inelastic x-ray scattering at high-pressure

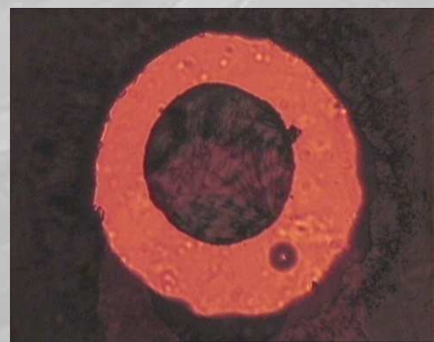
- Single crystal => momentum resolved phonon dispersion
- Small sample volume: good crystal quality





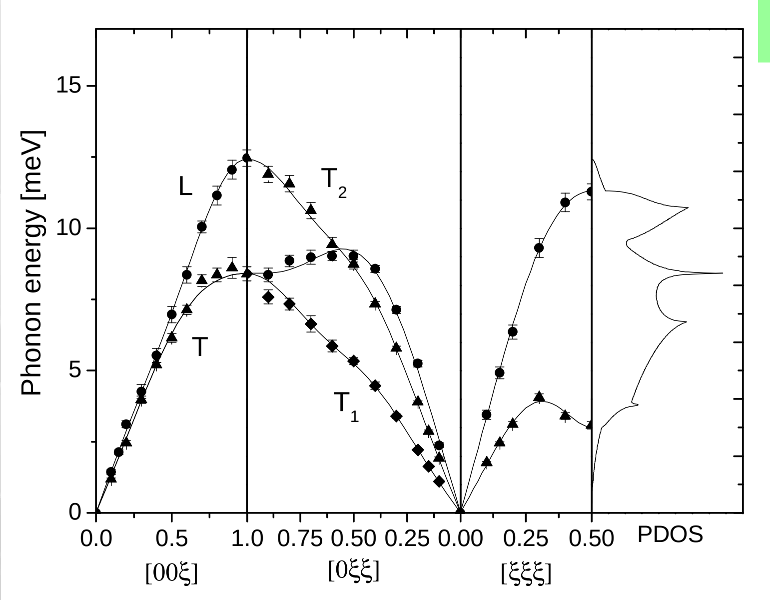
Samples:
60 μm diameter, 20 μm thickness

ID28 @ ESRF:
ΔE = 3 meV @ 17794 eV

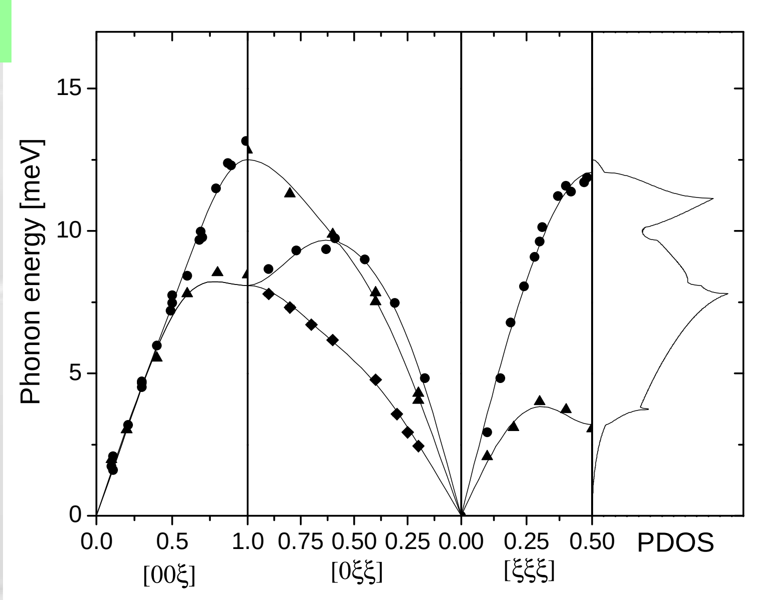


γ-phase

P ambient (INS)

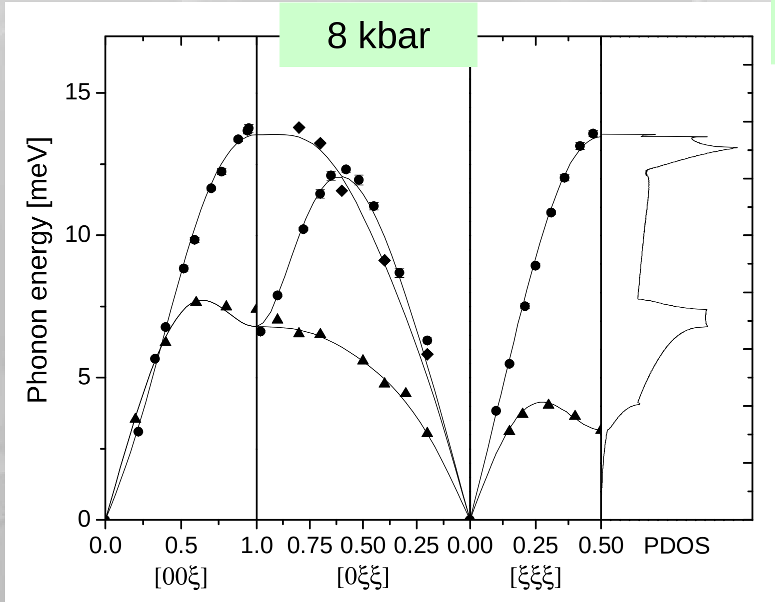


4 kbar

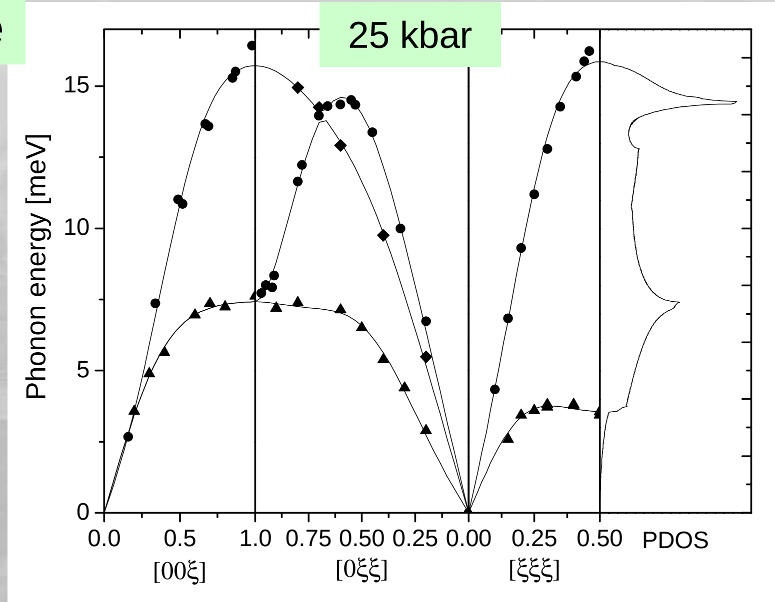


α-phase

8 kbar

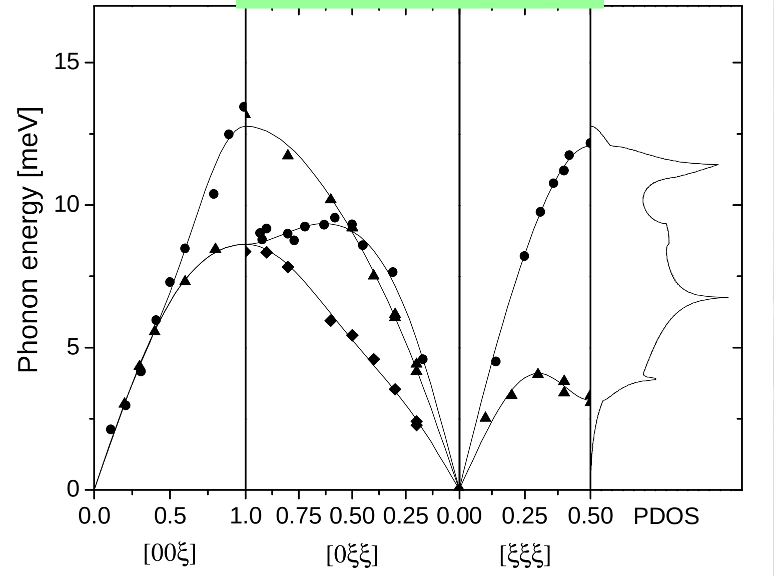


25 kbar

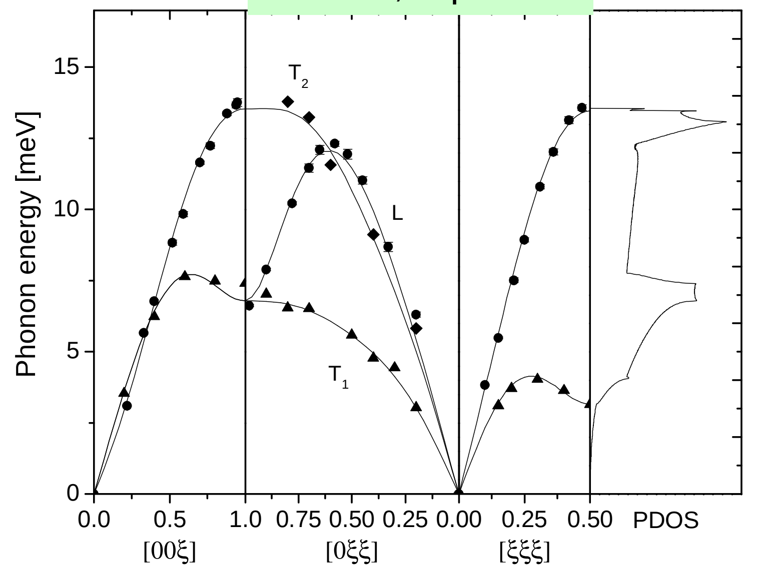


INS: C. Stassis et al.; Phys. Rev. B, 1979

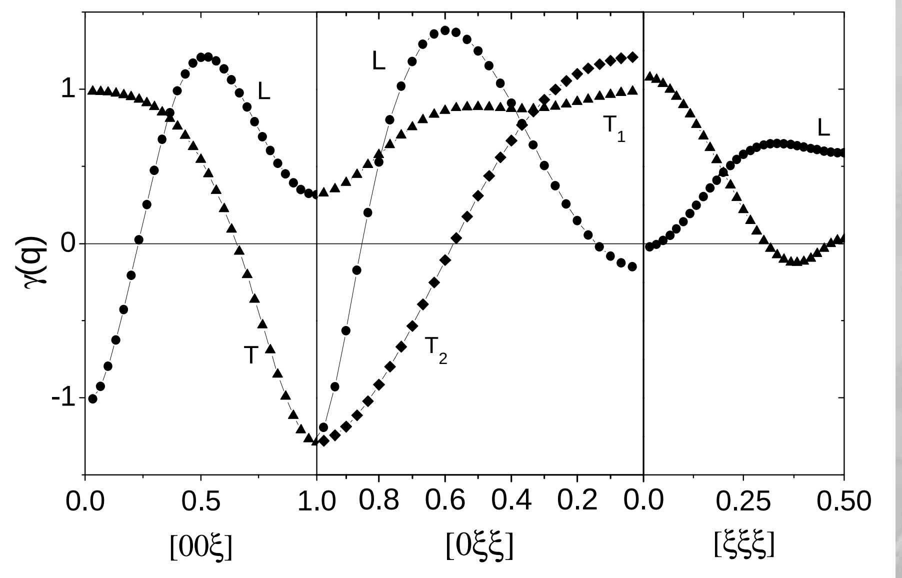
6 kbar, γ-phase



8 kbar, α-phase



$$\gamma_q = -\frac{\partial(\ln E(q))}{\partial(\ln V)}$$



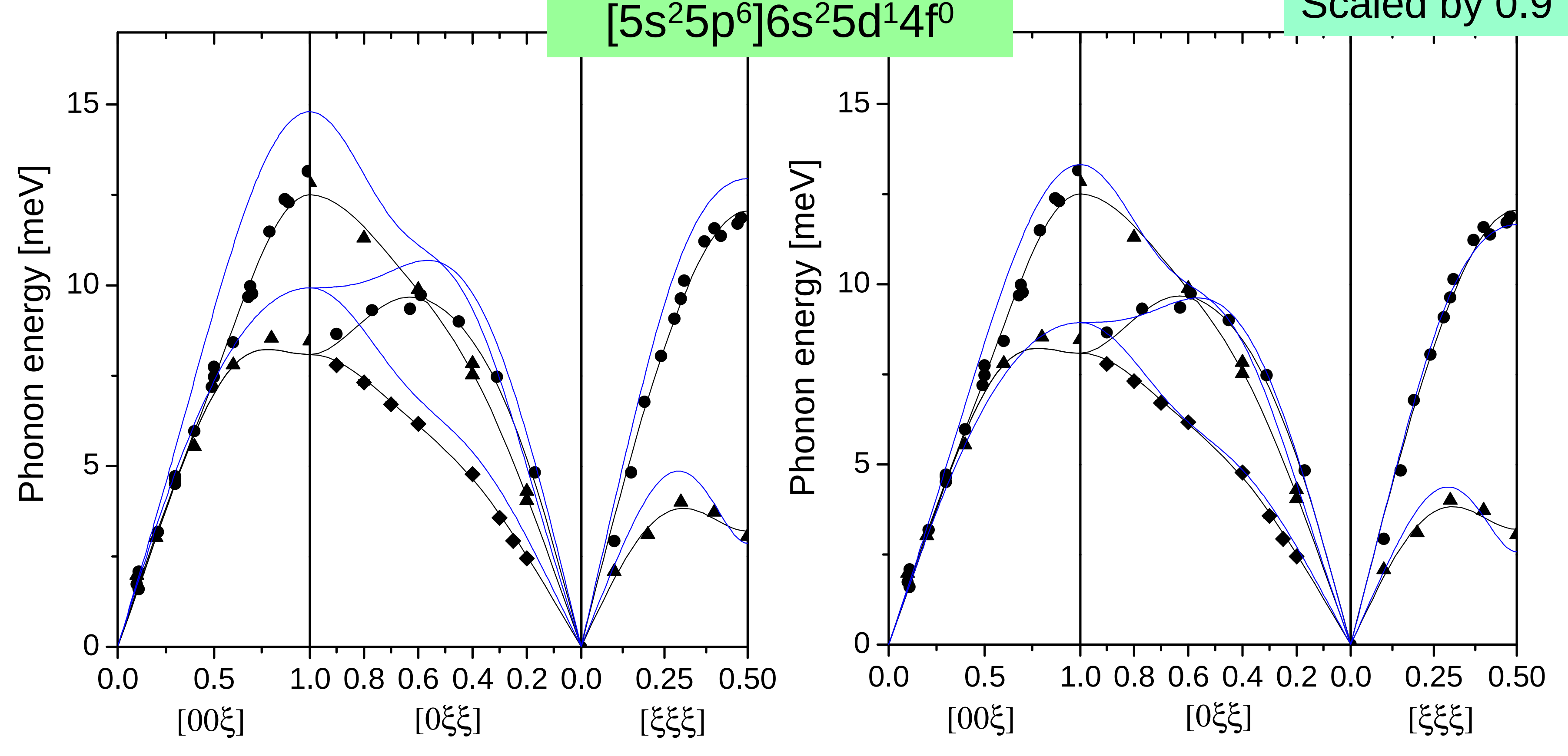
Experiment versus *ab initio* GGA calculation

(VASP, PBE, PAW, PHONON 4.22)

4 kbar, γ-phase

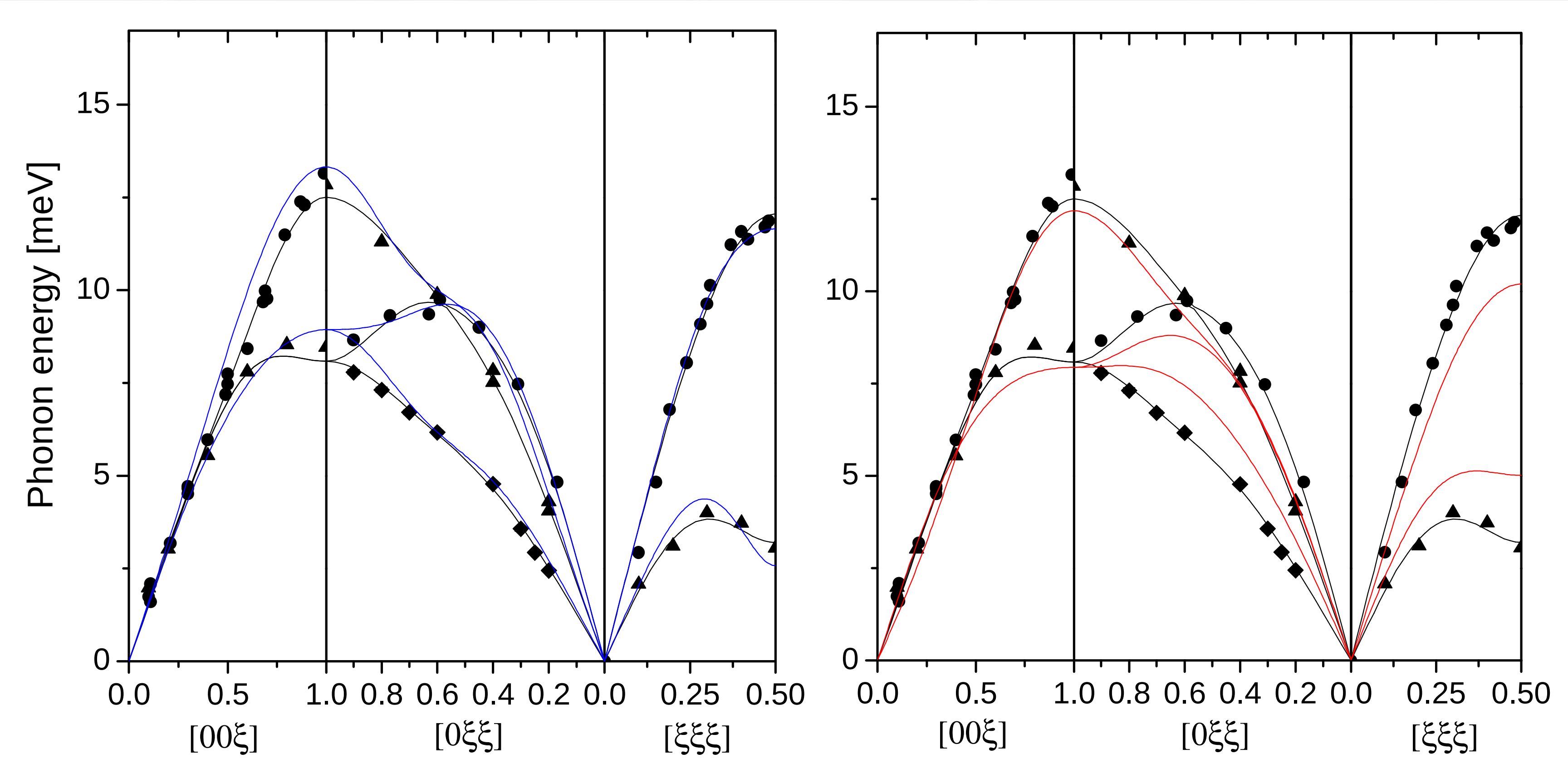
$[5s^2 5p^6] 6s^2 5d^1 4f^0$

Scaled by 0.9



4 kbar, γ-phase, scaled
$[5s^2 5p^6] 6s^2 5d^1 4f^0$

4 kbar, γ-phase
$[5s^2 5p^6] 6s^2 5d^1 4f^1$

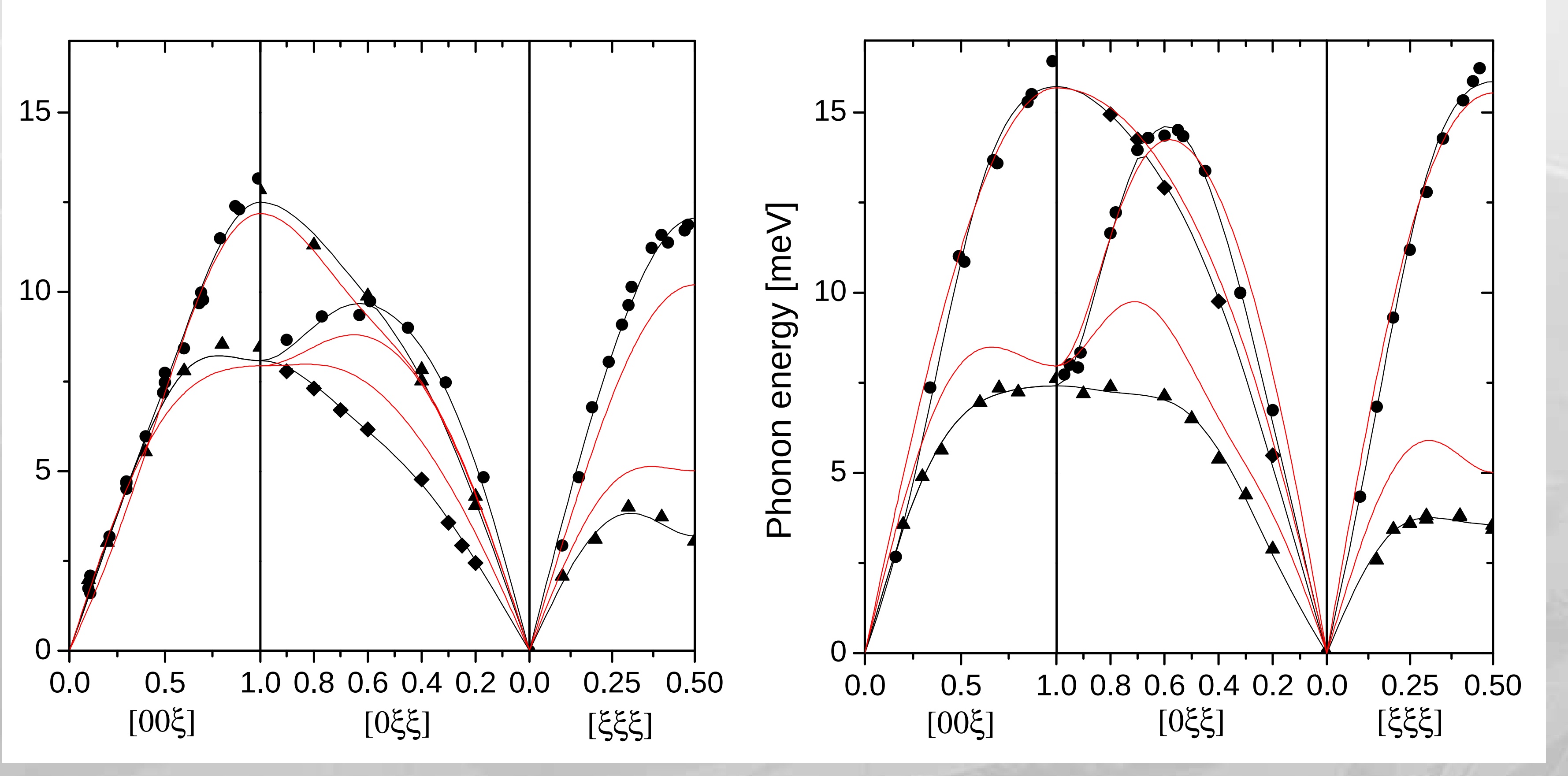


trivalent potential

tetravalent potential

4 kbar, γ-phase
[5s^{2}5p^6]6s^{2}5d^{1}4f^1

25 kbar, α-phase
[5s^{2}5p^6]6s^{2}5d^{1}4f^1



Trivalent potential yields negative phonon energies

Conclusions

- $\Delta S_{vib} = 0.3\ k_B$
  - amazing agreement with ultrasonic results by Voronov et al. 1960: $0.32\ k_B$
  - factor 2 difference w/r to recent X/N diffraction results

- Softening of C_{11} ($\gamma(q) < 0$ at low q for LA (001), consistent with observed softening of bulk modulus

- Most pronounced phonon anomalies around the X-point => significant changes in the electronic structure

- "Standard" ab initio methods capture the essentials, but do not reproduce (low energy) TA branches.

F.F. Voronov et al.; Soviet Physics Doklady (1960)