



The Abdus Salam  
International Centre for Theoretical Physics



2220-1

**15th International Workshop on Computational Physics and Materials  
Science: Total Energy and Force Methods**

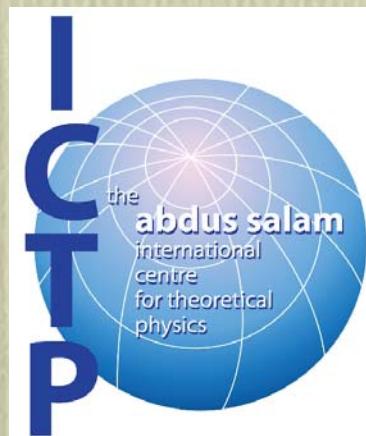
*13 - 15 January 2011*

**Self-consistent ab-initio lattice dynamics (SCAILD): Theory and numerical examples**

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# Self-consistent ab-initio lattice dynamics (SCAILD): Theory and numerical examples

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UPPSALA  
UNIVERSITET



International Workshop on Computational Physics  
and Materials Science: Total Energy and Force Calculations

Trieste

13-15, 2011

# Outline

- Standard methods for calculating phonons.
- The Self Consistent Ab Initio Lattice Dynamical (SCAILD) method
- Results
- Some Benchmarks
- The Future ?!

- Standard methods for calculating phonons.

Frozen phonon method

Direct method

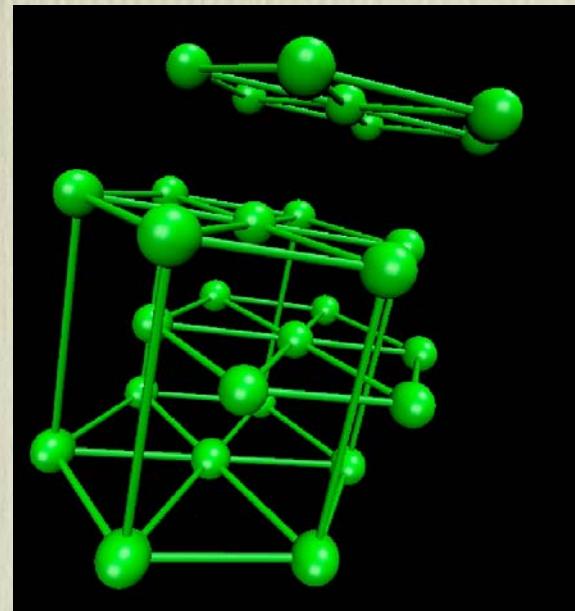
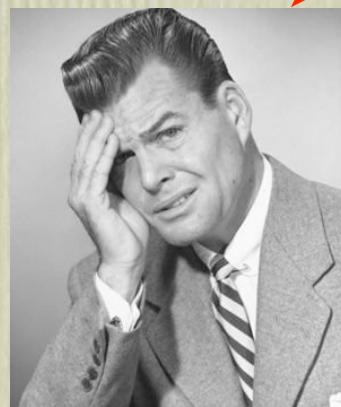
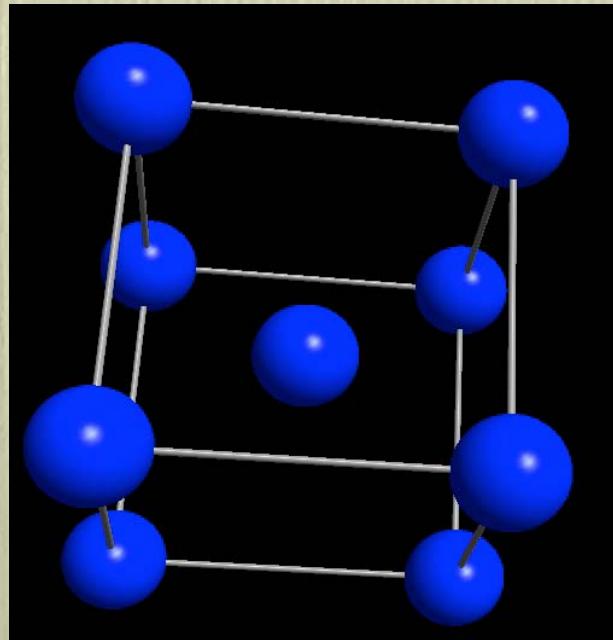
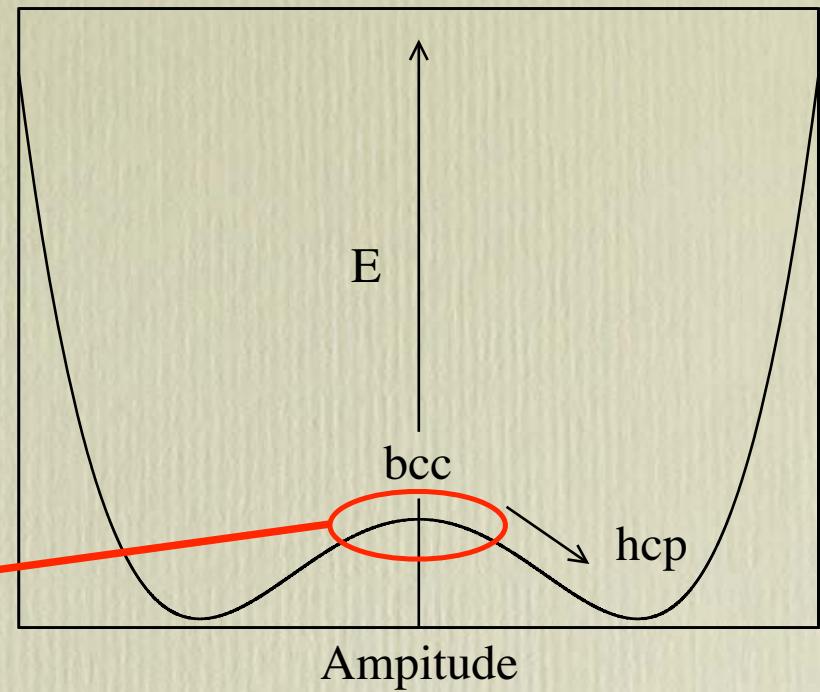
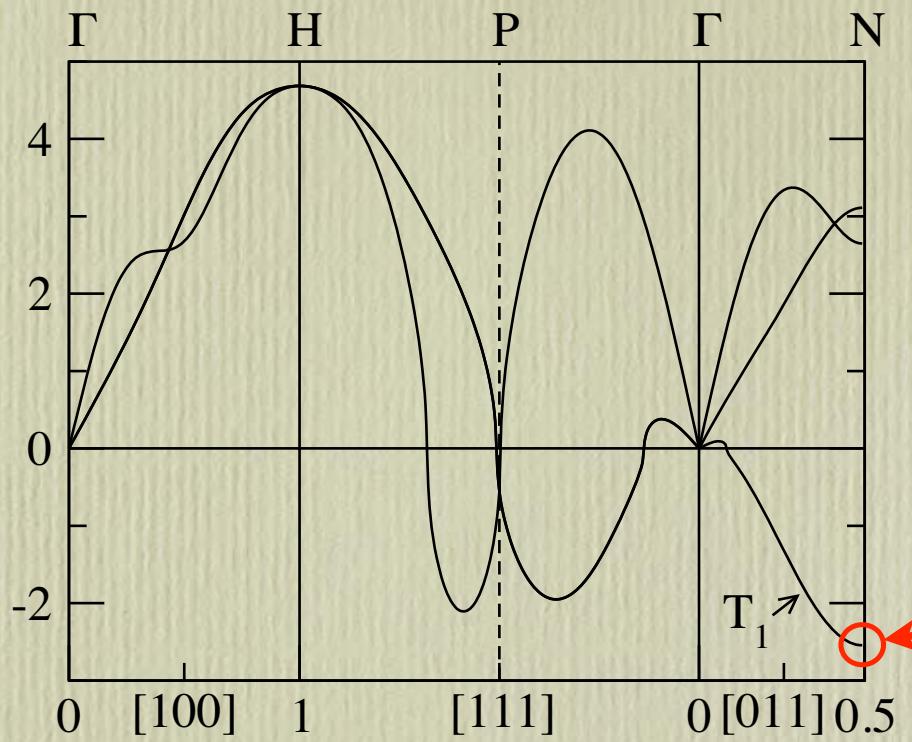
Linear response

All quasi harmonic methods !

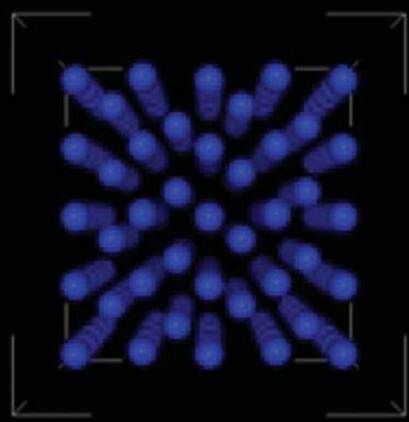
(quasi = only the effect of the thermal-expansion is taking into account when calculating the phonon frequencies  $\omega_{qs}$  )

# PROBLEMS !!

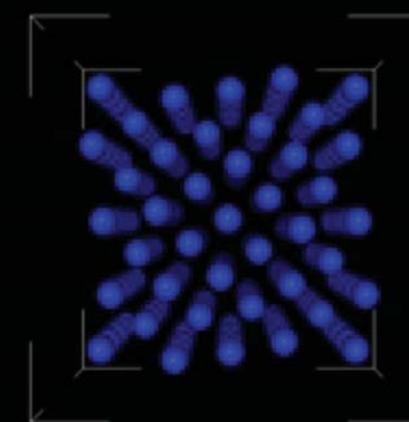
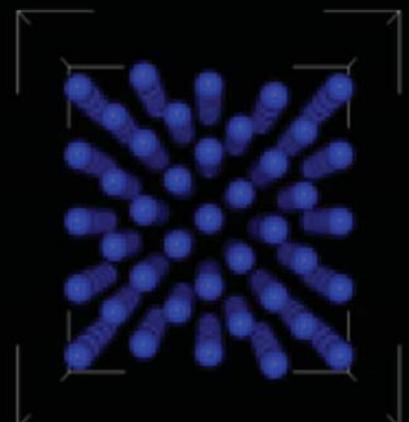
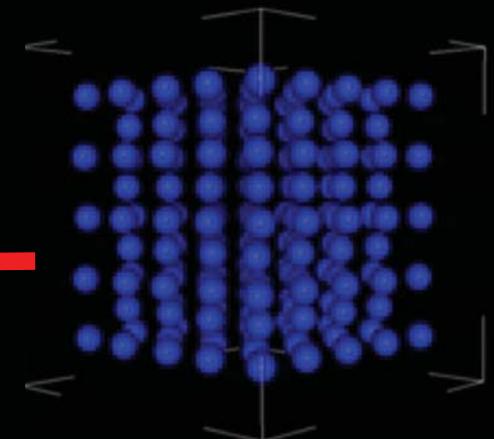
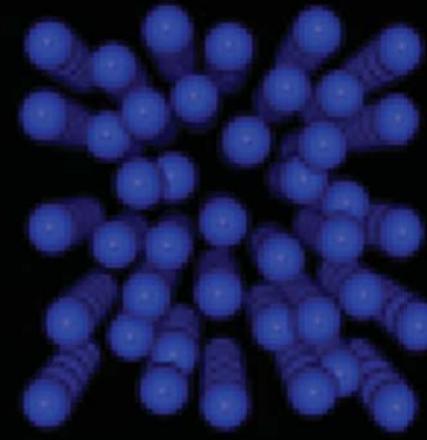
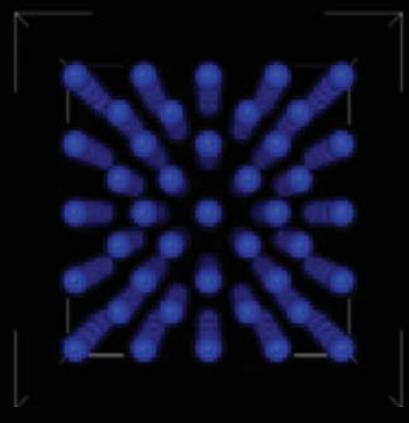
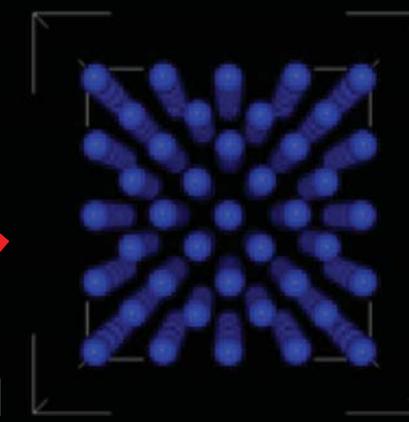




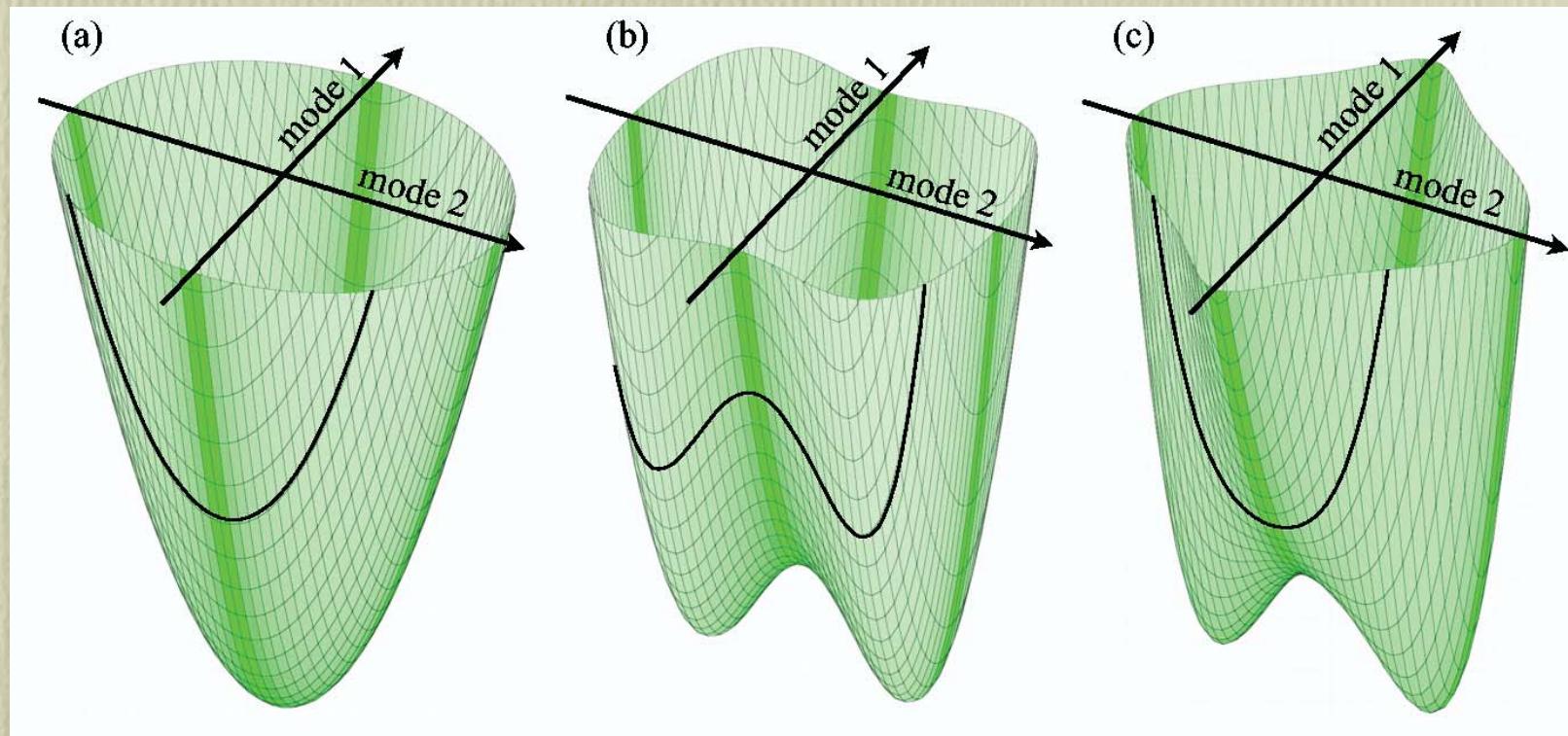
# The Self Consistent Ab Initio Lattice Dynamical (SCAILD) Method



$$U_R = \frac{1}{\sqrt{N}} \sum_{k_s} \epsilon_{k_s} R_{k_s} e^{iRk}$$



# Alternative Perspective: Dynamical Stabilization By Phonon-Phonon Interaction



## I) Calculate starting guess

$$\mathbf{R} = \mathbf{R}_0 + \mathbf{U} \Rightarrow \bar{\Phi}(\mathbf{R})$$

$$\Rightarrow \bar{D}(\mathbf{k}) = \sum_{\mathbf{R}} \bar{\Phi}(\mathbf{R}) e^{-i\mathbf{R}\mathbf{k}} \Rightarrow \omega_{\mathbf{k}s}^o$$

## 2) Calculate atomic displacements

$$\mathbf{U}_{\mathbf{R}} = \frac{1}{\sqrt{N}} \sum_{\mathbf{k}s} \epsilon_{\mathbf{k}s} \mathcal{R}_{\mathbf{k}s} e^{i\mathbf{R}\mathbf{k}}$$

$$\mathcal{R}_{\mathbf{k}s}^2 = \frac{\hbar}{M\omega_{\mathbf{k}s}} \left[ \frac{1}{2} + \bar{n} \left( \frac{\hbar\omega_{\mathbf{k}s}}{k_B T} \right) \right]$$

## 3) Calculate forces

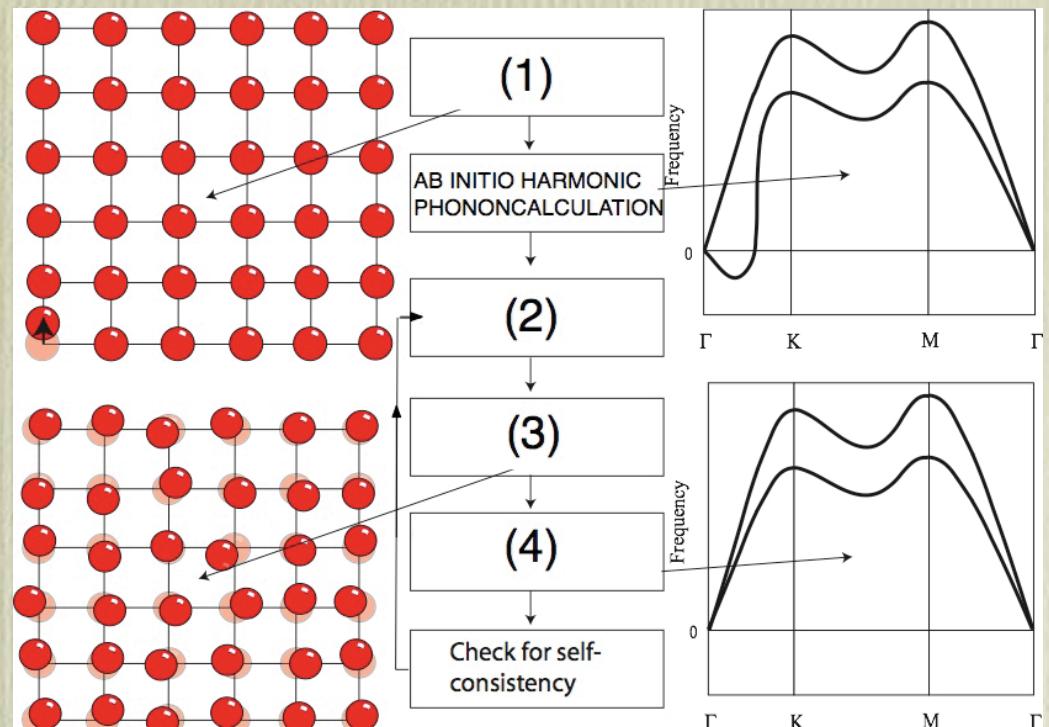
$$\mathbf{F}_{\mathbf{R}} = -\langle \Psi | \frac{\partial \mathcal{H}}{\partial \mathbf{R}} | \Psi \rangle$$

## 4) Calculate new frequencies

$$\bar{\omega}_{\mathbf{k}s}^2 = -\frac{1}{M} \frac{\epsilon_{\mathbf{k}s} \mathbf{F}_{\mathbf{k}}}{\mathcal{R}_{\mathbf{k}s}}$$

$$\Omega_{\mathbf{k}s}^2 = \frac{1}{m_{\mathbf{k}}} \sum_{S \in S(\mathbf{k})} \bar{\omega}_{S^{-1}\mathbf{k}s}^2 \Rightarrow \omega_{\mathbf{k}s}(N) = \frac{1}{N} \sum_{i=1}^N \Omega_{\mathbf{k}s}^2(i)$$

# THE SCAILD PROCEDURE



# The mean-field frequencies

$$\bar{\omega}_{\mathbf{k}s}^2 = -\frac{1}{M} \frac{\epsilon_{\mathbf{k}s} \mathbf{F}_{\mathbf{k}}}{\mathcal{R}_{\mathbf{k}s}}$$

$$\bar{\omega}_{\mathbf{k}s}^2 = \omega_{\mathbf{k}s}^2 \left( 1 + \frac{1}{2} \sum_{\mathbf{k}_1, \mathbf{k}_2} \sum_{s_1, s_2} \mathcal{A}(\mathbf{k}, \mathbf{k}_1, \mathbf{k}_2, s, s_1, s_2) \frac{\mathcal{R}_{\mathbf{k}_1 s_1} \mathcal{R}_{\mathbf{k}_2 s_2}}{\mathcal{R}_{\mathbf{k}s} \omega_{\mathbf{k}s}^2} + \dots \right)$$

$$\mathcal{R}_{\mathbf{k}s}^2 = \frac{\hbar}{M \omega_{\mathbf{k}s}} \left[ \frac{1}{2} + \bar{n} \left( \frac{\hbar \omega_{\mathbf{k}s}}{k_B T} \right) \right]$$

$$\mathcal{A}(\mathbf{k}, \mathbf{k}_1, \mathbf{k}_2, s, s_1, s_2) =$$

$$\frac{1}{(MN)^{3/2}} \sum_{\mathbf{R}, \mathbf{R}_1, \mathbf{R}_2} \sum_{\alpha, \beta, \gamma} \Phi_{\alpha\beta\gamma}(\mathbf{R}, \mathbf{R}_1, \mathbf{R}_2) \epsilon_{\mathbf{k}s\alpha} \epsilon_{\mathbf{k}_1 s_1 \beta} \epsilon_{\mathbf{k}_2 s_2 \gamma} e^{i(\mathbf{R}\mathbf{k} + \mathbf{R}_1\mathbf{k}_1 + \mathbf{R}_2\mathbf{k}_2)}$$

# Principal Results/Publications

(I) *Entropy driven stabilization of energetically unstable crystal structures explained from first principles theory.*

P. Souvatzis, O. Eriksson, M. I. Katsnelson and S. P. Rudin, Phys. Rev Lett. **100**, 095901 (2008)

(II) *Ab initio study of interacting lattice vibrations and stabilization of the  $\beta$ -phase in Ni-Ti shape-memory alloy*

P. Souvatzis, Dominik Legut, Olle Eriksson and M. I. Katsnelson, Phys. Rev. B **81**, 092201 (2010)

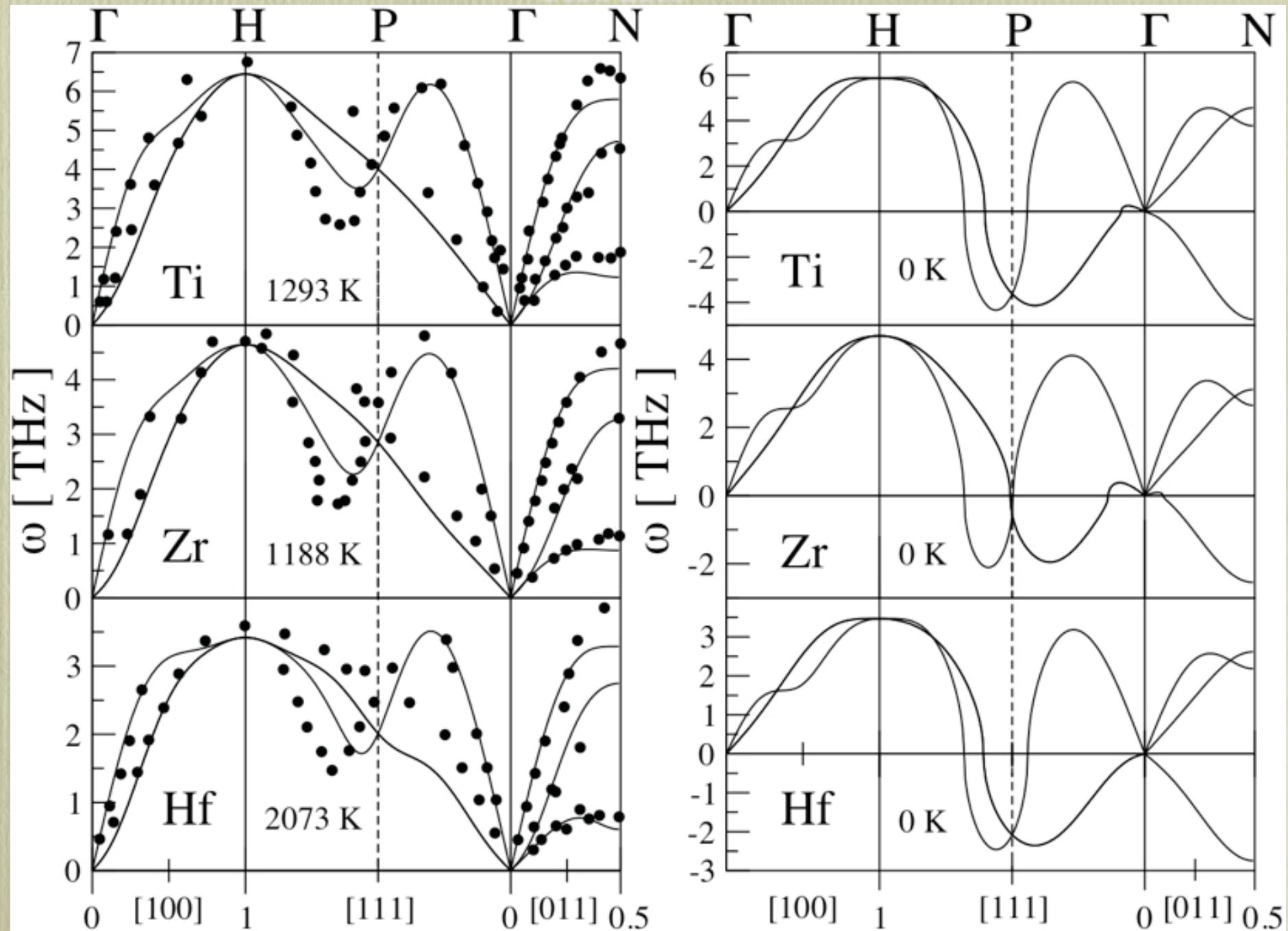
(III) *Dynamical stability of body center cubic iron at the Earths core conditions*

W. Luo, B. Johansson, O. Eriksson, S. Arpan, P. Souvatzis, M. I. Katsnelson and R. Ahuja, PNAS **107**, 9962-9964 (2010)

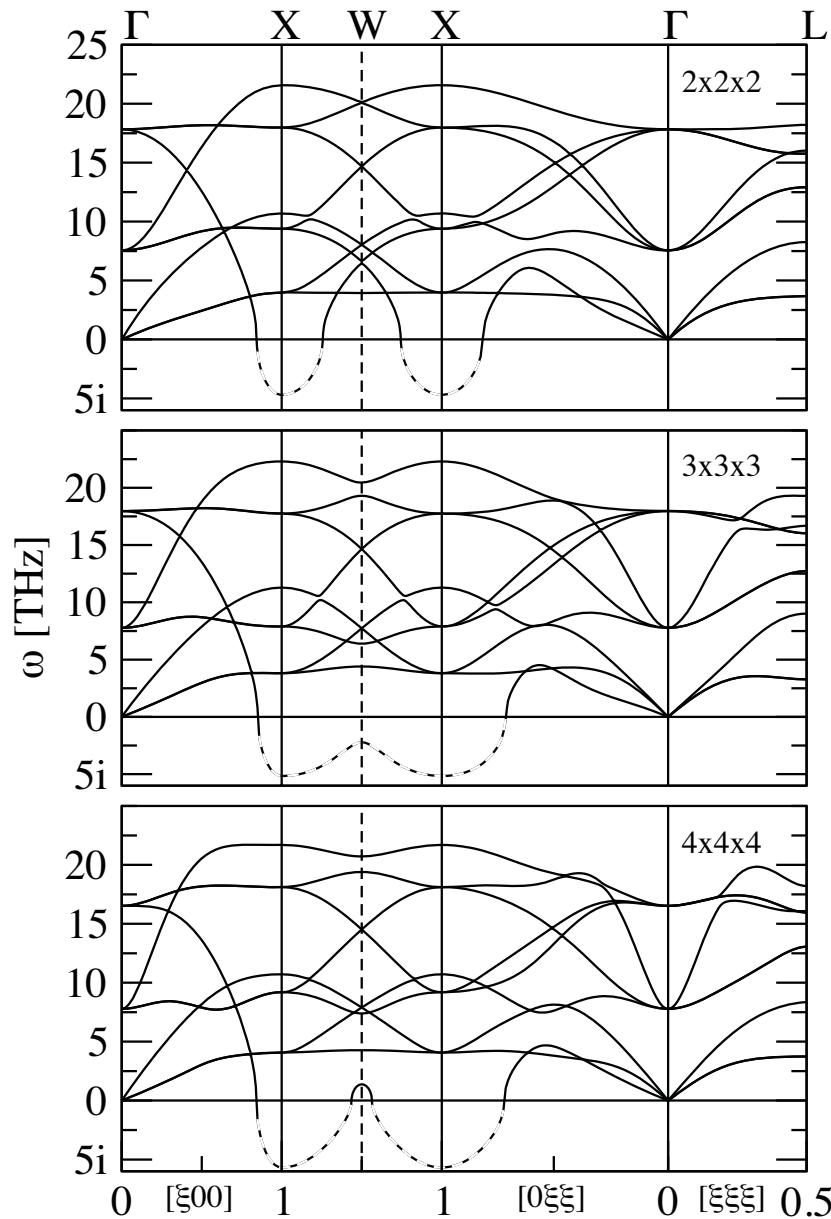
(IV) *Entropically Stabilized Local Dipole Formation in Lead Chalcogenides*

Emil S. Božin, Christos D. Malliakas, Petros Souvatzis, Thomas Proffen, Nicola A. Spaldin, Mercouri G. Kanatzidis, and Simon J. L. Billinge, Science, **330** 1660- 1663,(2010)

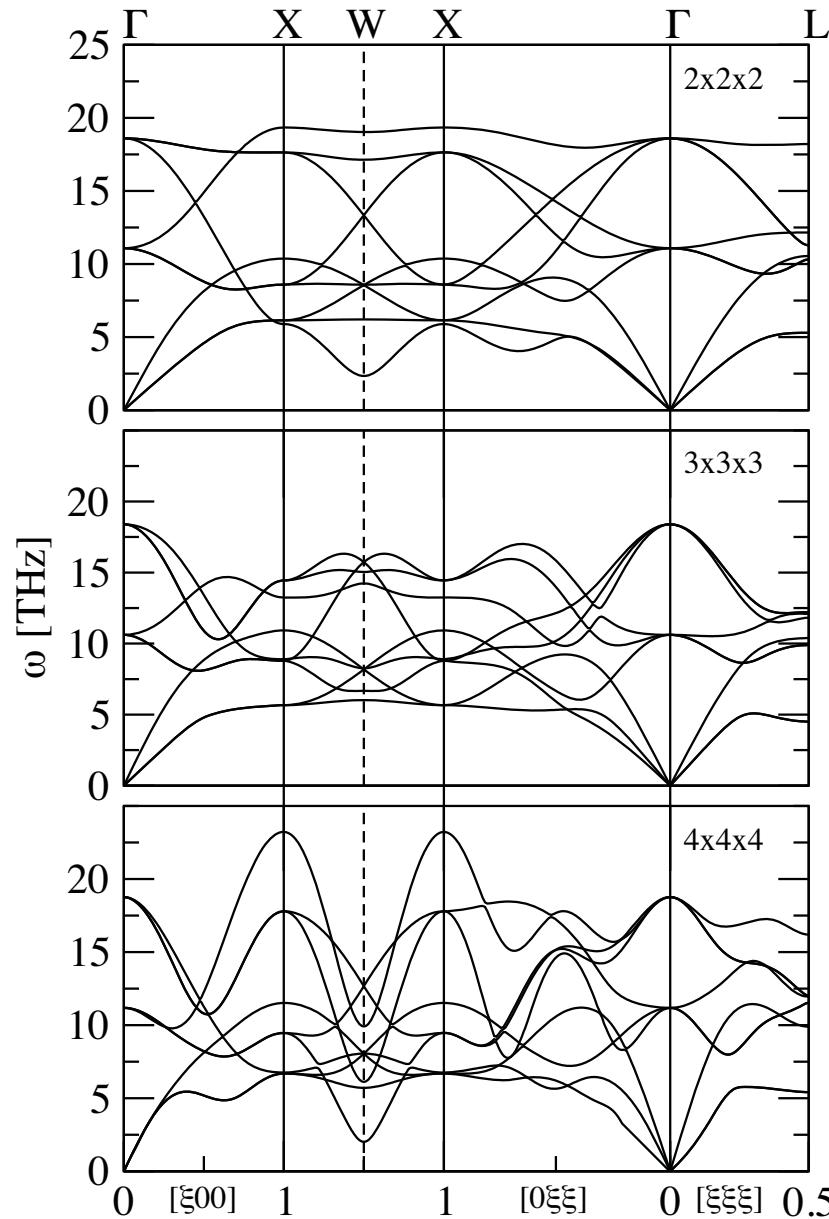
# Results

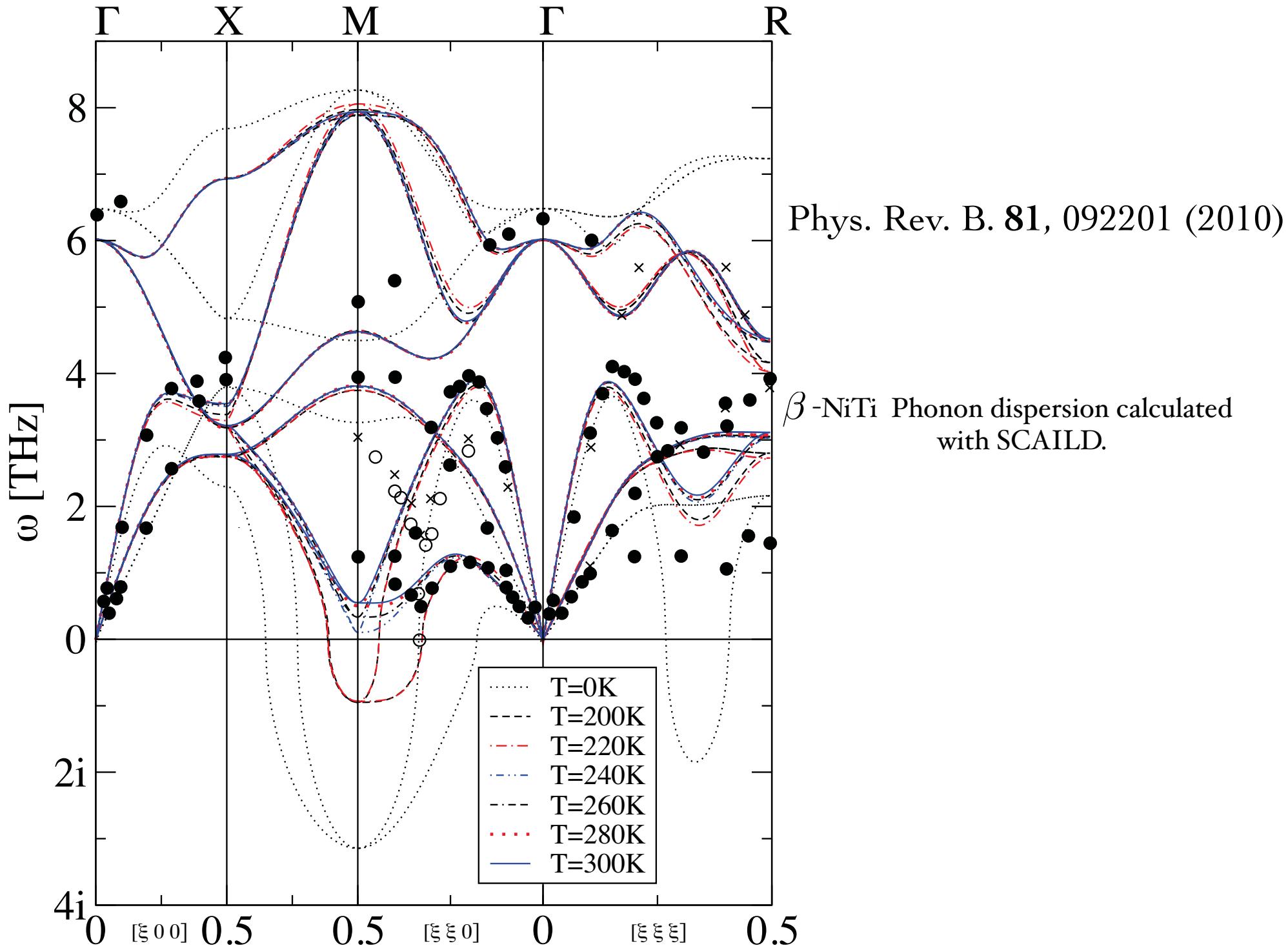


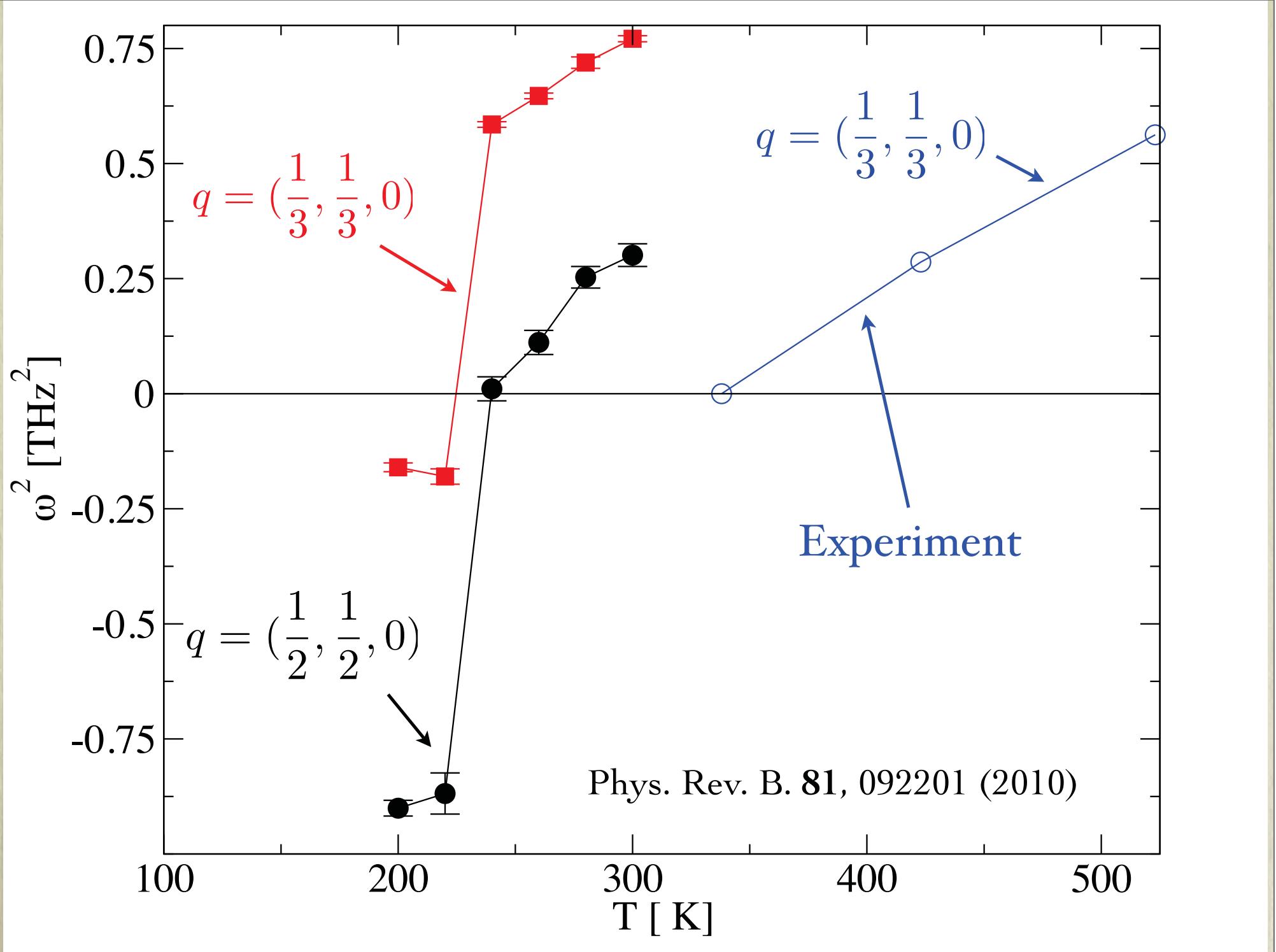
(a) quasiharmonic



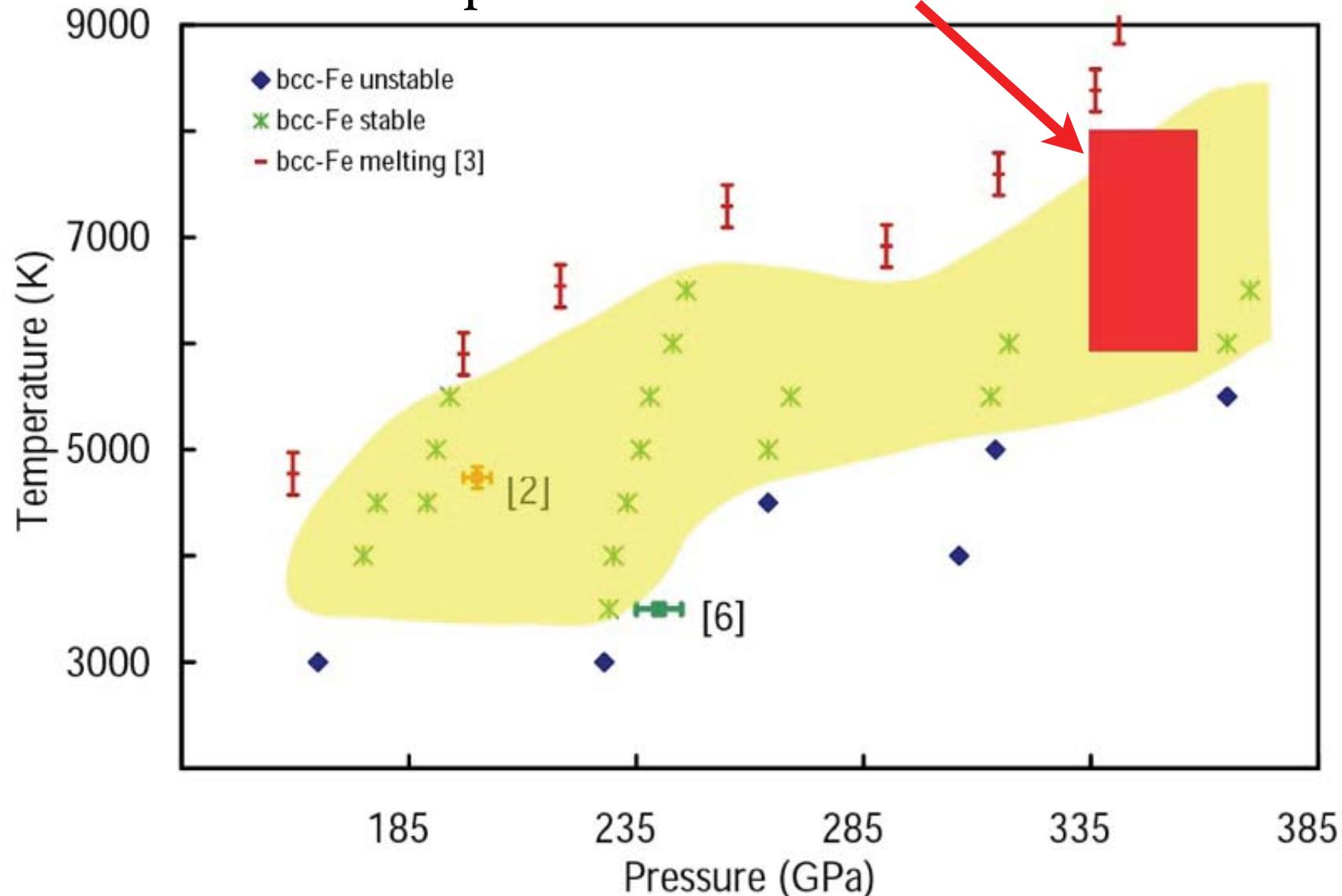
(b) 2570 K



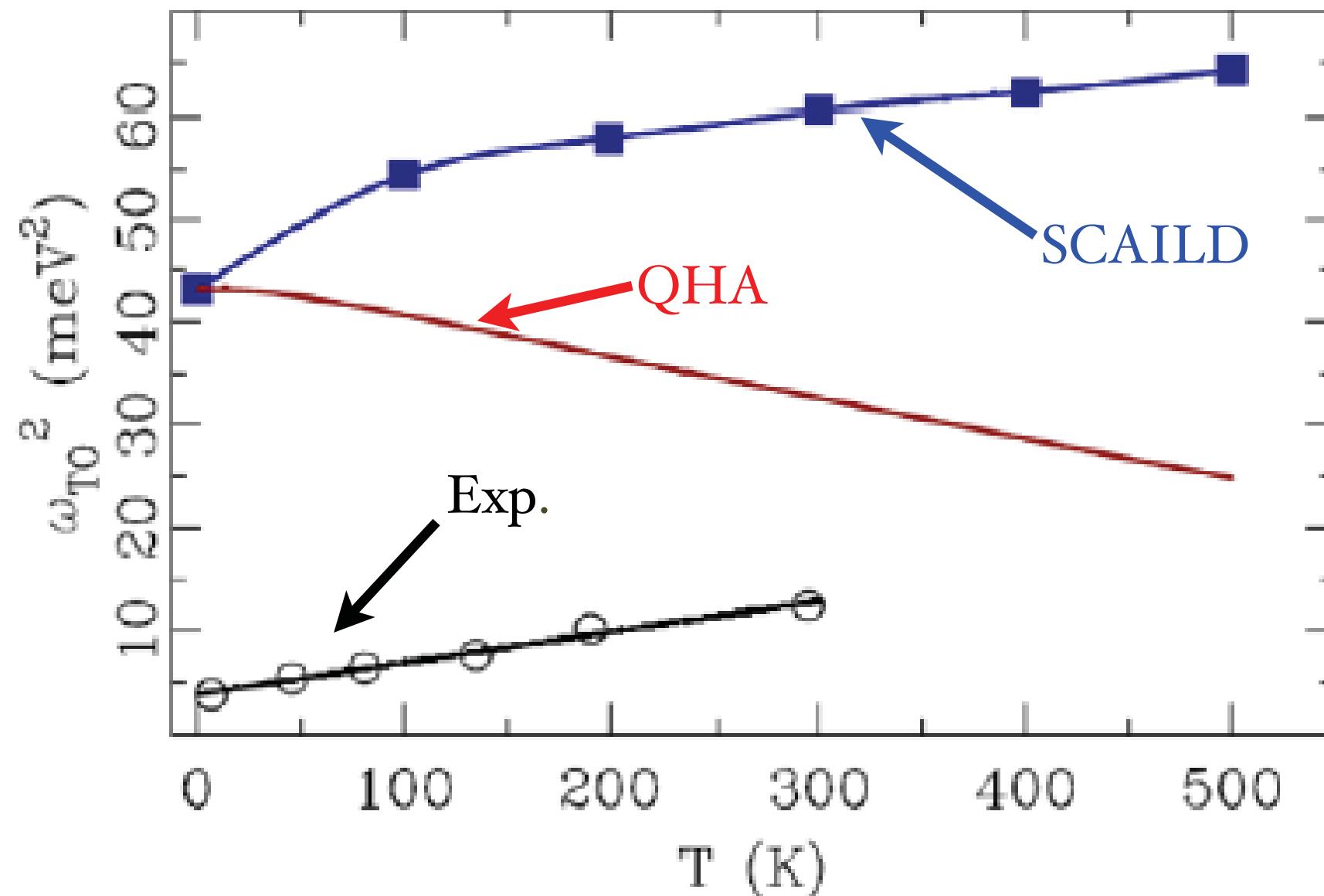




BCC Fe up to Earth Inner Core Conditions



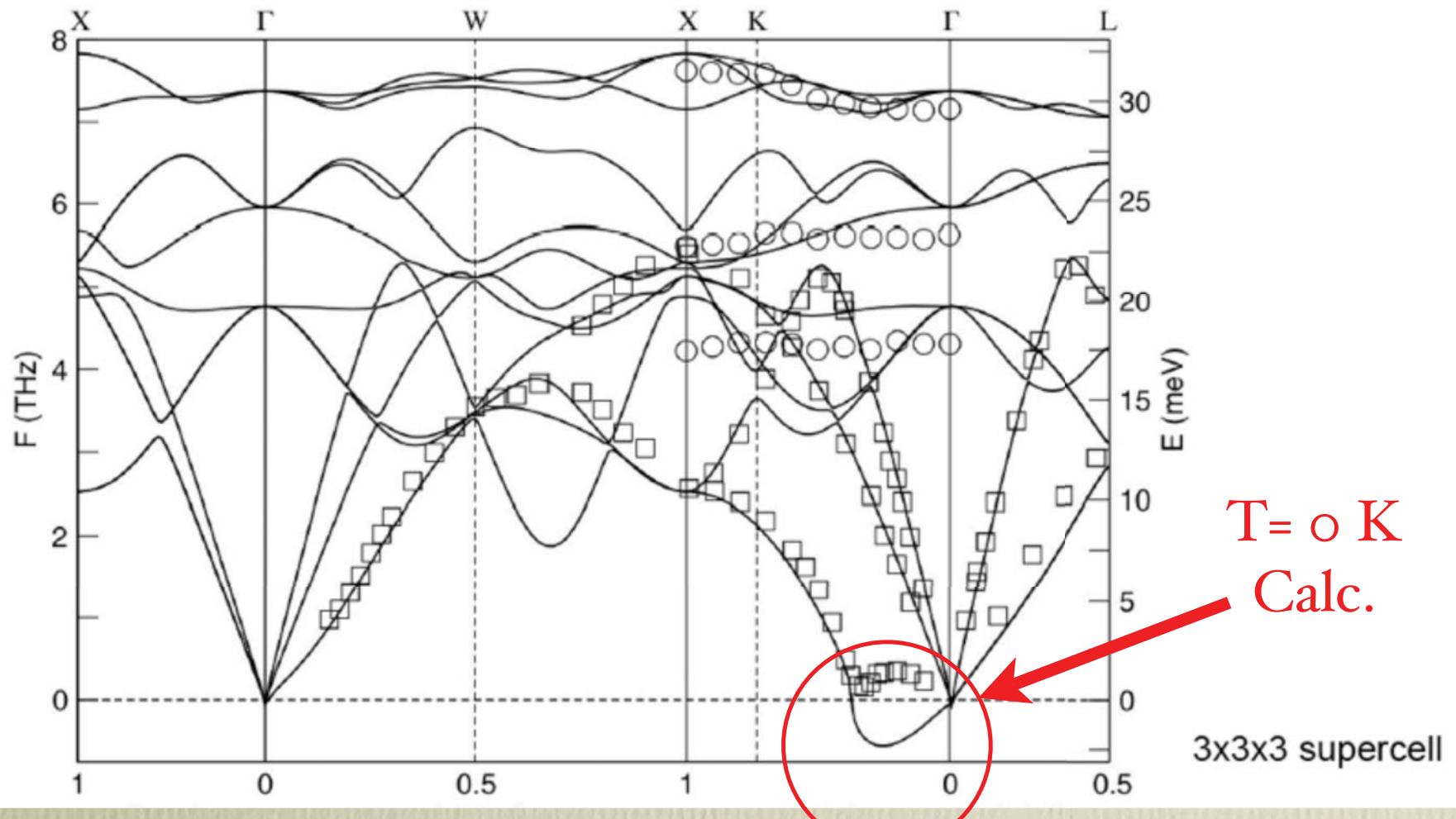
PbS



# Some Benchmark Calculations

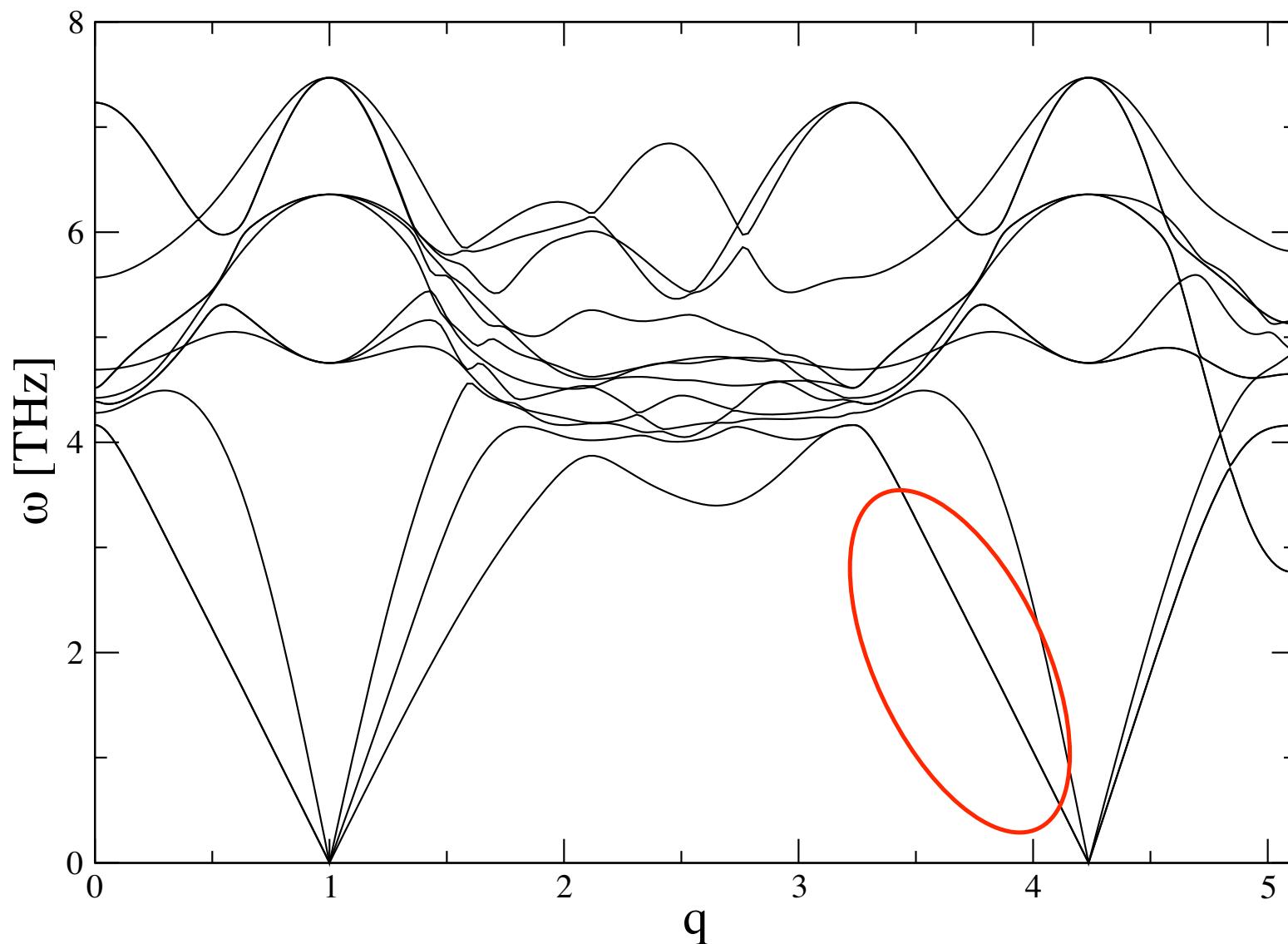


# Phonons of $\text{Ni}_2\text{MnGa}$



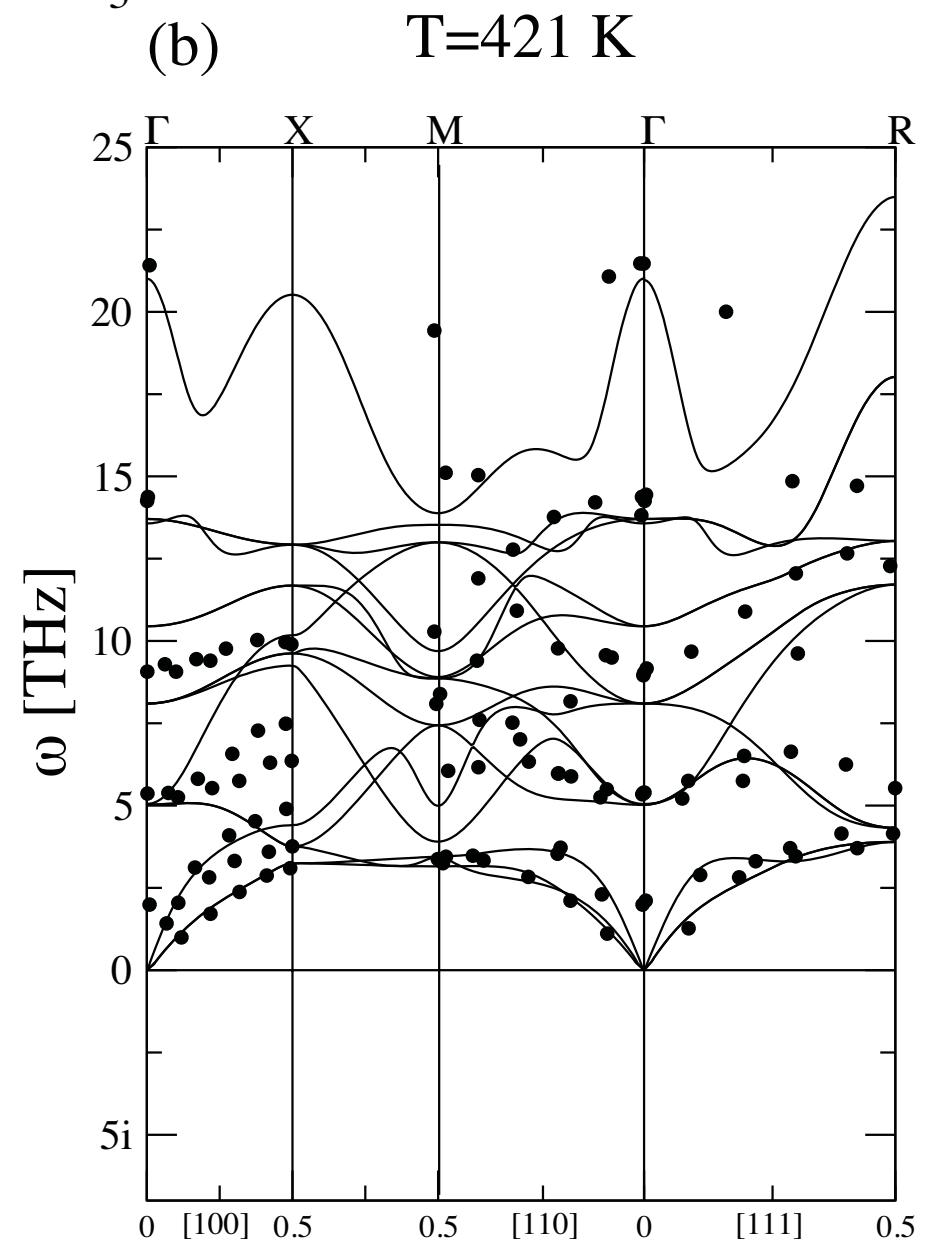
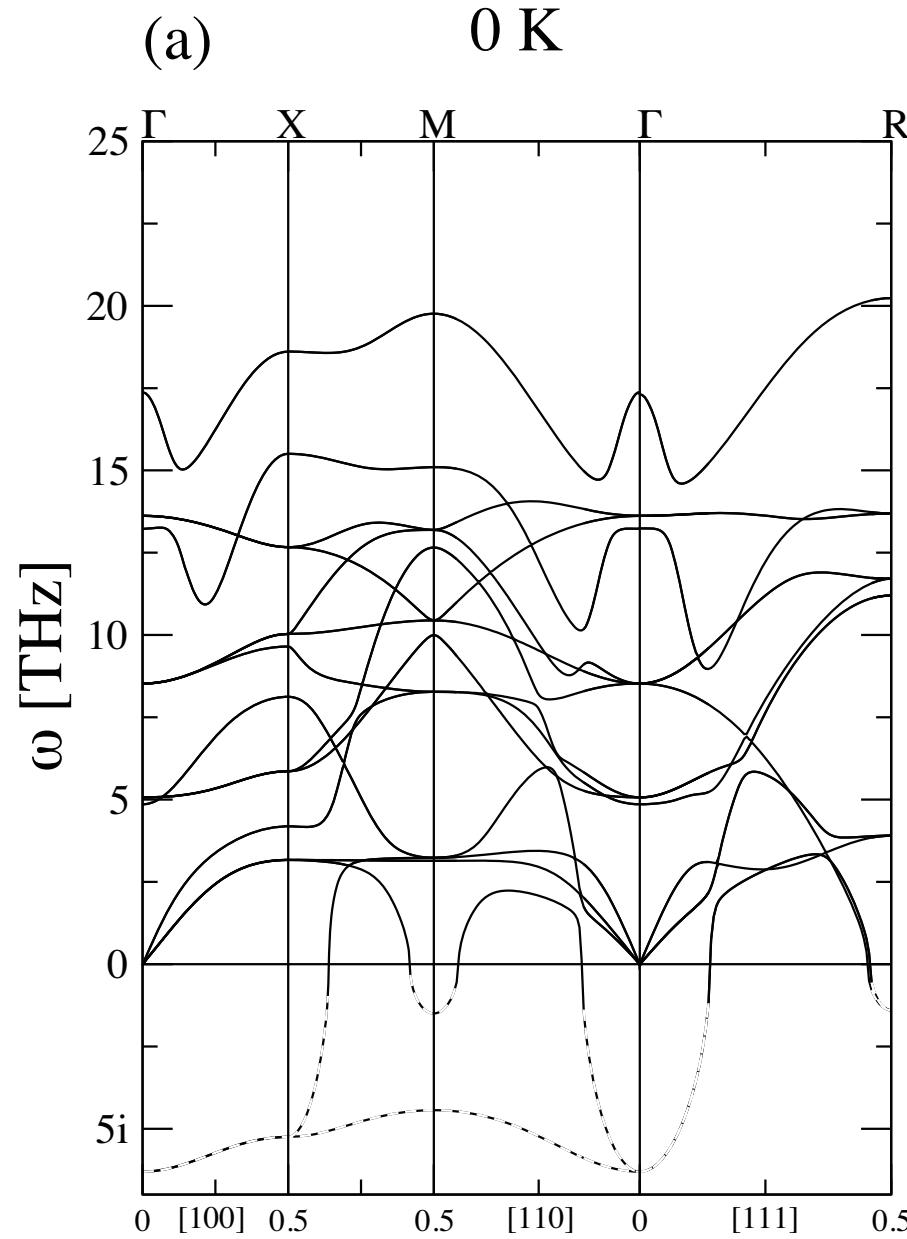
# $\text{Ni}_2\text{MnGa}$

T=300K, SCAILD Calc.



# Cubic

BaTiO<sub>3</sub>



# Future developments:

- Free energies, Calc. Running!
- Phonon lifetimes, Calc. Running!

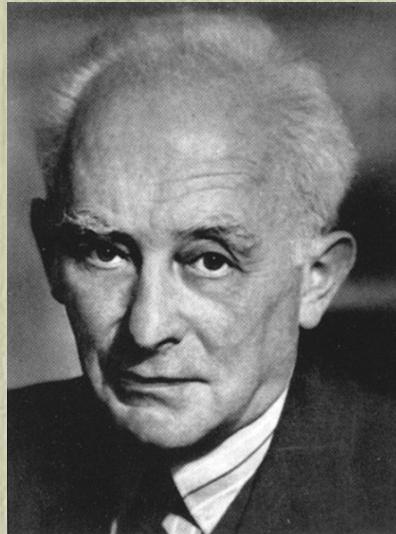
# How do I get access to the scaild code?

Send me an email ([petros.souvatjis@fysik.uu.se](mailto:petros.souvatjis@fysik.uu.se))  
requesting a password, then go to:

[http://web.mac.com/petros.souvatjis/Webbplats\\_2/SCAILD.html](http://web.mac.com/petros.souvatjis/Webbplats_2/SCAILD.html)

And download!

# THANK YOU!



*The Grandfather of Olivia Newton John (Max Born)*



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