ESRF

Phonon Density of States in $NdFeAsO_{1-x}F_x$

Mathieu Le Tacon, **Michael Krisch and Alexei Bosak**
Beamline ID28 "Inelastic X-ray Scattering II" - ESRF

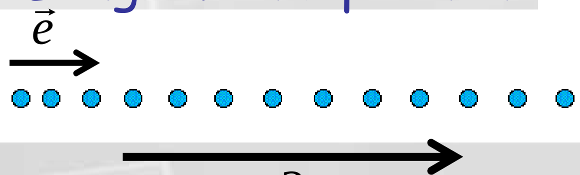
Jan-Willem Bos and Serena Margadonna
University of Edinburgh

Lattice dynamics Investigations

Phonons = Quantum Mechanical description of lattice vibrations

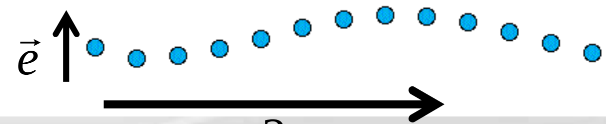
Ex. atomic chain

Longitudinal phonon



$\vec{q} = \frac{2\pi}{\lambda}\hat{x}$

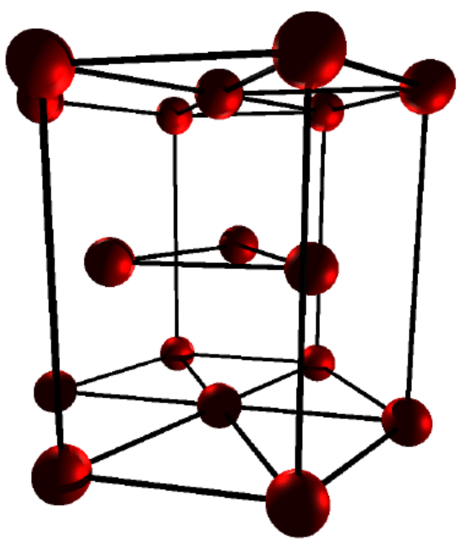
Transverse phonon

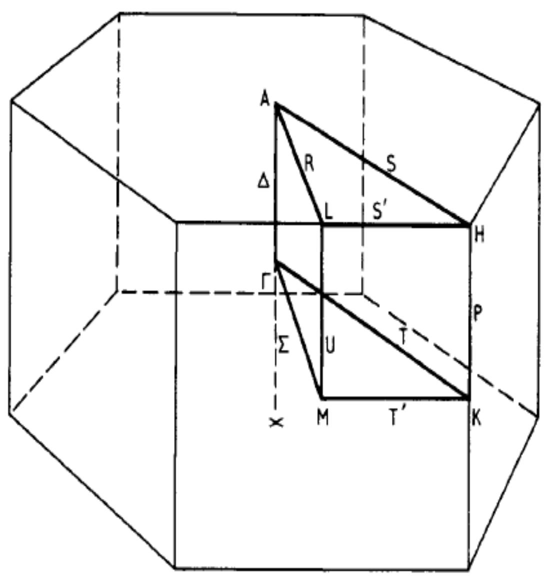


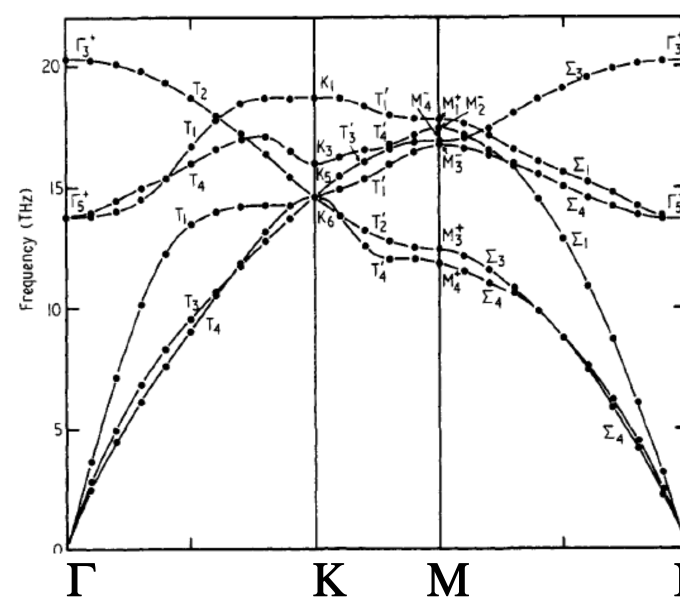
$\vec{q} = \frac{2\pi}{\lambda}\hat{x}$

Energy cost: $E = \omega(\vec{q})$

Ex: Case of Be (R. Stedman et al. J. Phys. F, 6 157 (1975))







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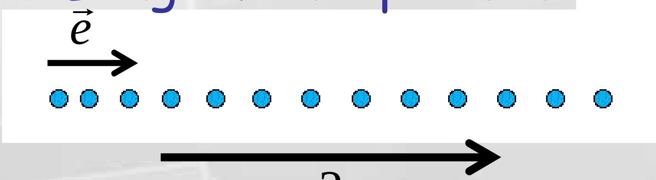

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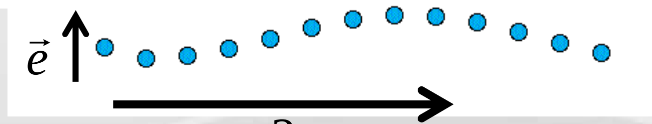


$\vec{q} = \frac{2\pi}{\lambda}\hat{x}$

Energy cost: $E = \omega(\vec{q})$

Why is it interesting ??

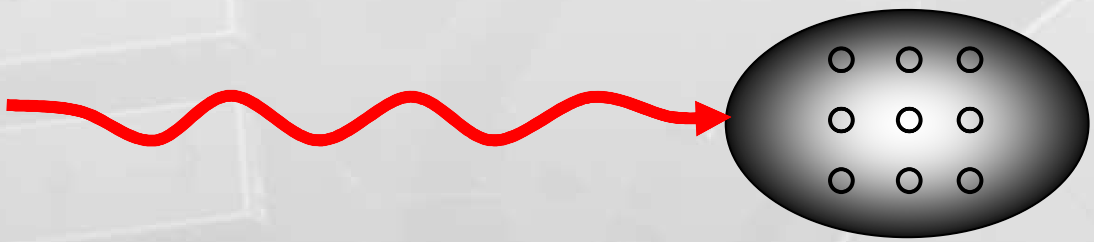
Contains many informations e.g.

- interatomic forces & elastic properties
- thermodynamics (thermal expansion, heat capacity, melting)
- electron phonon-coupling (superconductors, CDW systems)

Measuring the phonons

Typical energy ~ 10 meV

Infrared absorption

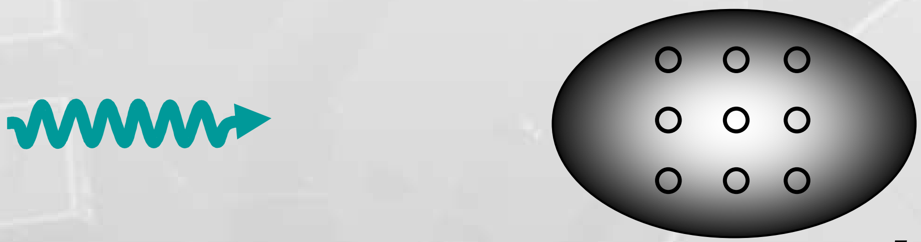


Measuring the phonons

Typical energy ~ 10 meV

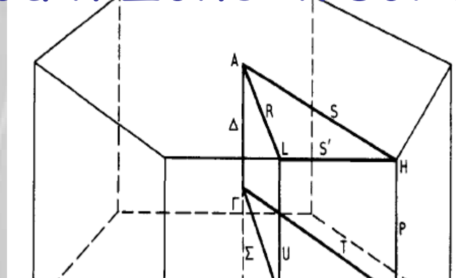
Infrared absorption

Inelastic scattering of visible light (2eV)



Typical wave vector of visible light: $k = E/\hbar c \approx 1.5\times 10^{-3}\, nm^{-1}$

Typical size of the Brillouin Zone in solids: $k_{ZB} = \pi / a \approx 10\, nm^{-1}$



Infrared and Visible light can only probe excitations close to the Γ point

Probing dispersive excitations with inelastic scattering

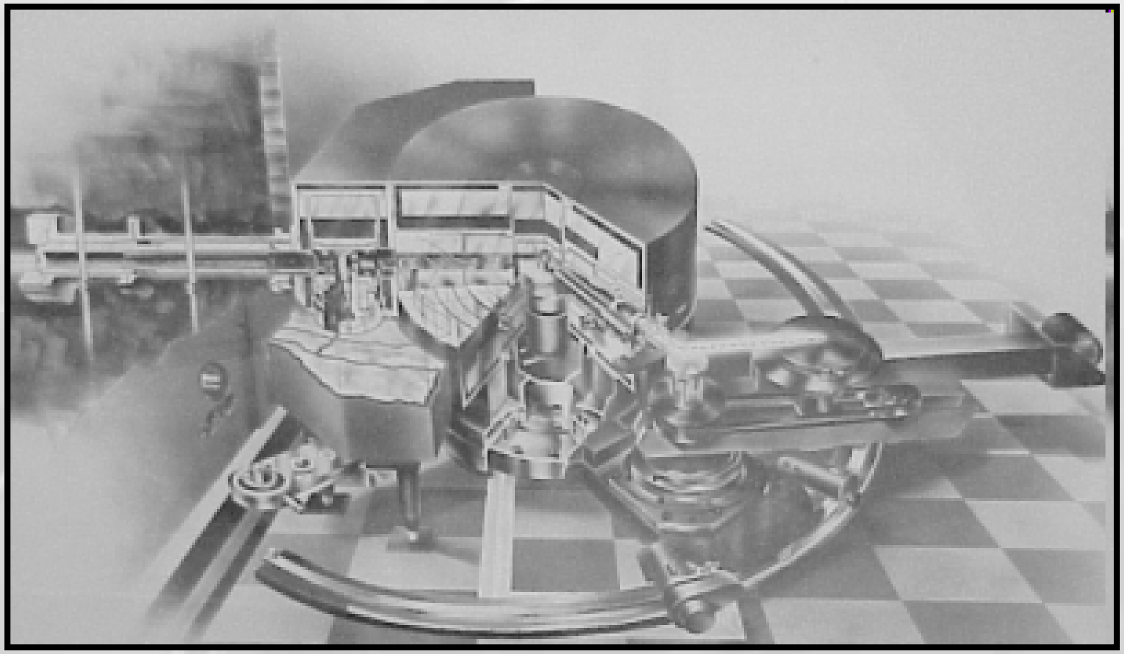
Transferred momentum Q needs at least to cover the entire BZ for the energy ranges of interest

Ideal probe: neutrons

Ex: thermal neutrons

$$E \approx 25\, meV$$

$$k = \sqrt{2EM/\hbar^2} \approx 10^{12}\, m^{-1}$$



The Triple Axis Neutron Spectrometer
(B.N. Brockhouse 1957, cf S. Petit's talk)

BUT intrinsic limitations:
- Weak cross section (Big Samples !)
- Strong coupling btw momentum energy

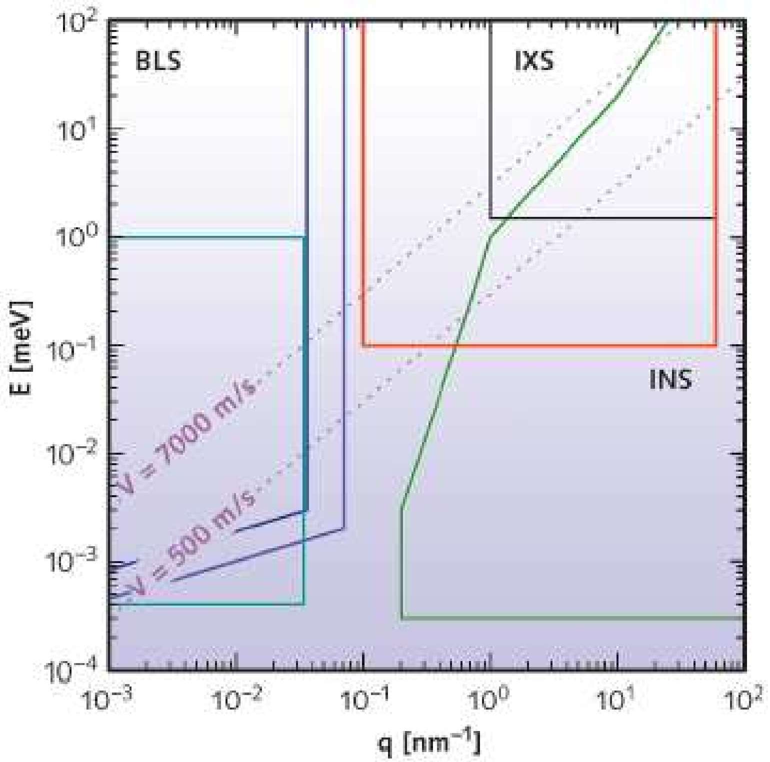
Solution: use X-rays

Inverse problem :

Need to use keV photons to have similar momentum than neutrons

Need for very high resolution power: e.g. ΔE =1.5meV @ E=21.7 keV

$$\frac{\Delta E}{E} \approx 10^{-8}$$



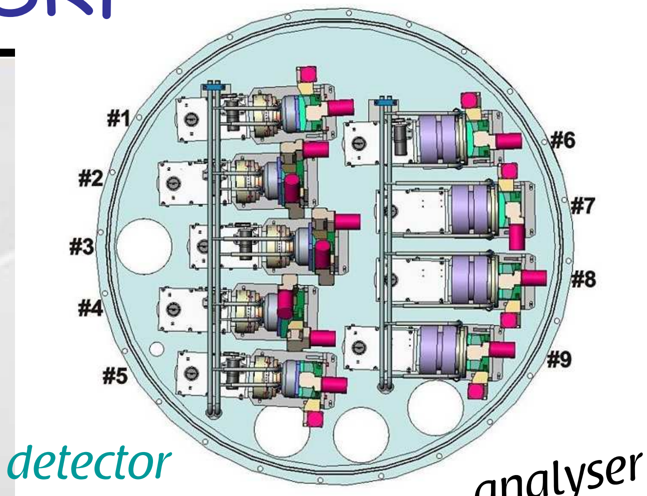
Possibility to explore collective dynamics in:

- crystalline materials, even for very small crystals/quantities
- disordered systems in which acoustic excitations have to be measured at low Q

Beamline ID28 at ESRF

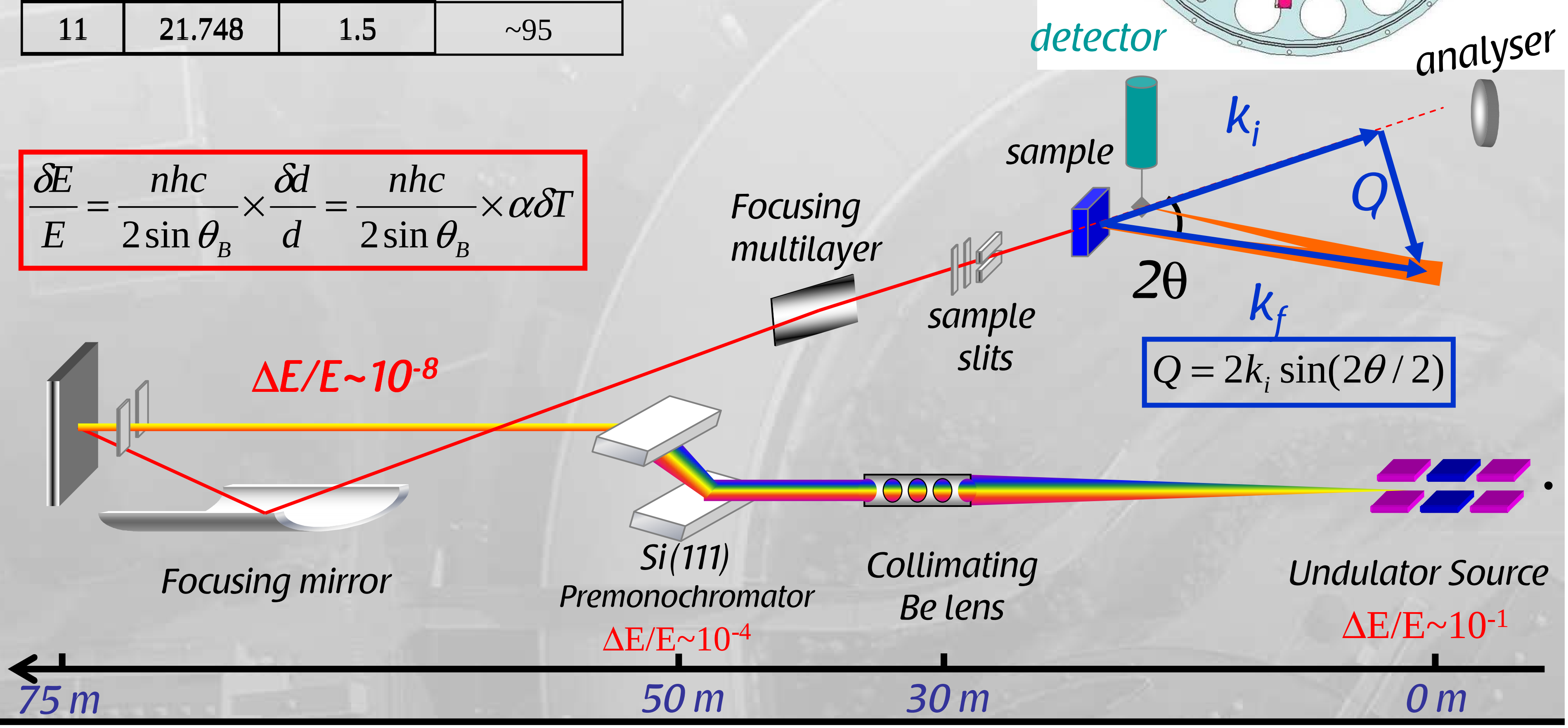
Backscattering (q_B = 89.98°)
High-Resolution Monochromator
Si(nnn):

n	E (keV)	ΔE (mev)	Q_{max} (nm^{-1})
8	15.816	5.4	~70
9	17.793	3.0	~80
11	21.748	1.5	~95

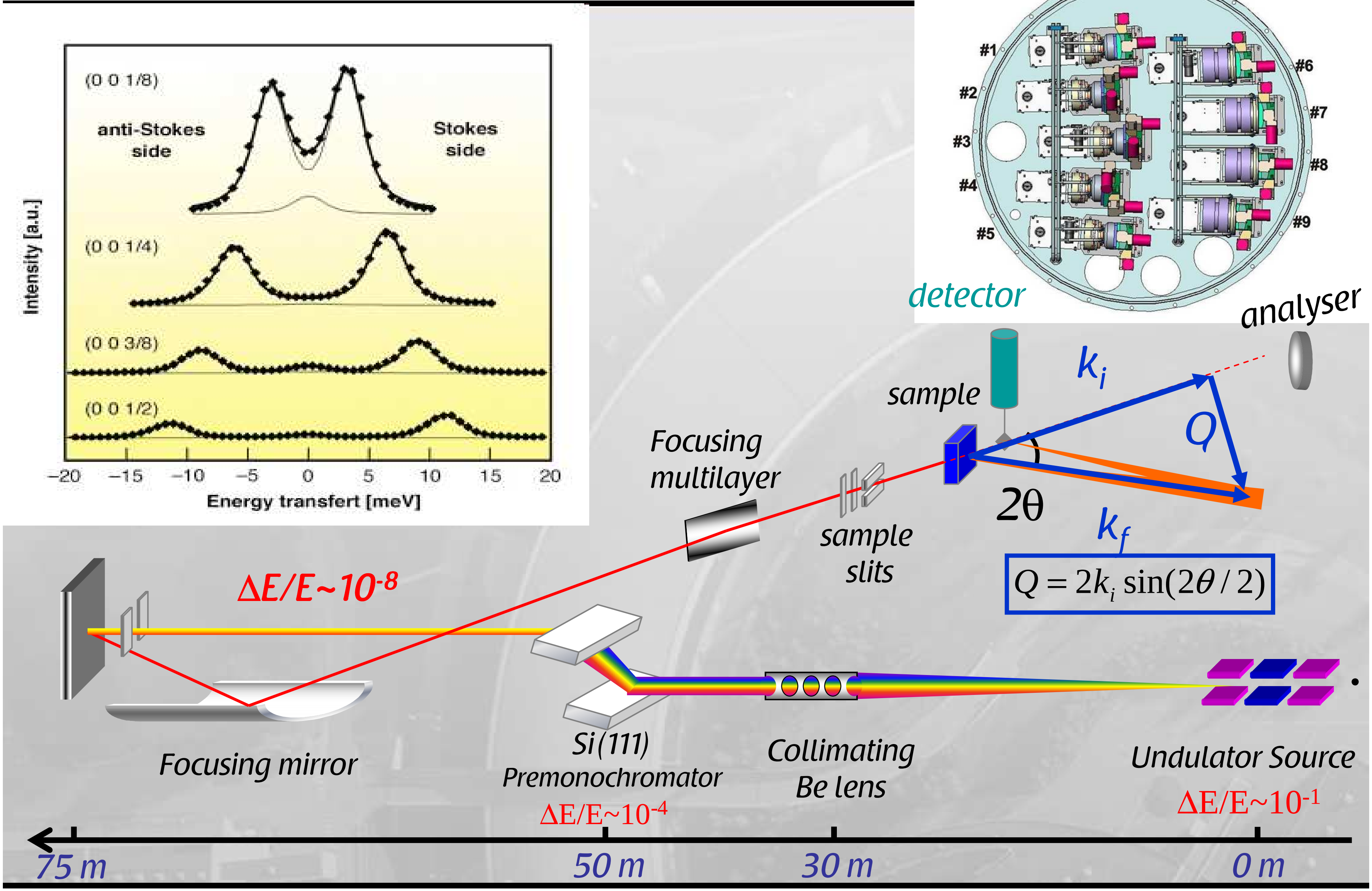


$$\frac{\delta E}{E} = \frac{nhc}{2\sin\theta_B} \times \frac{\delta d}{d} = \frac{nhc}{2\sin\theta_B} \times \alpha\delta T$$

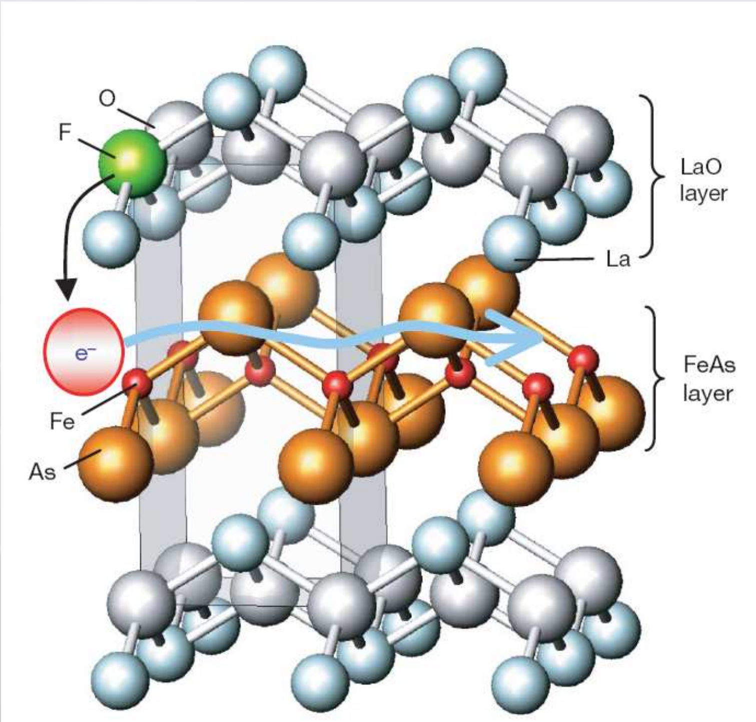
$$Q = 2k_i \sin(2\theta/2)$$



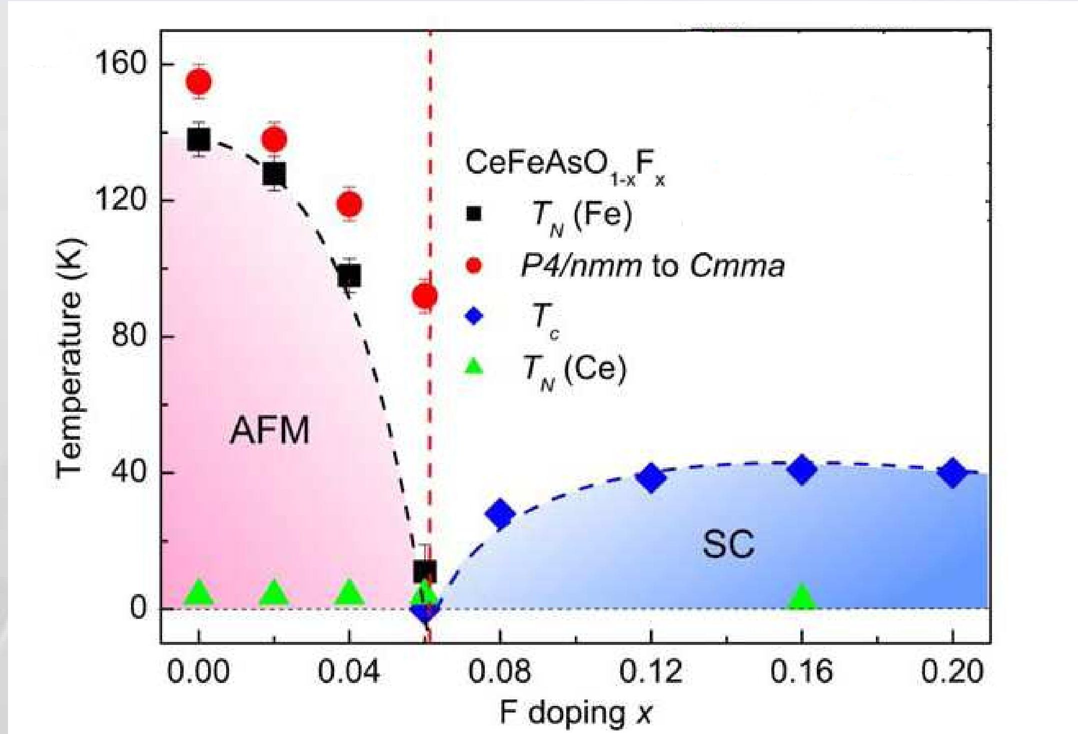
Beamline ID28 at ESRF



Iron based oxy-pnictides RE O Fe (As, P, N) [RE = La, Nd, Sm, Ce …]



Kamihara, Y., et al. J. Am. Chem. Soc. **130** 3296 (2008).
Takahashi, H., et al. Nature **453** 376 (2008)..



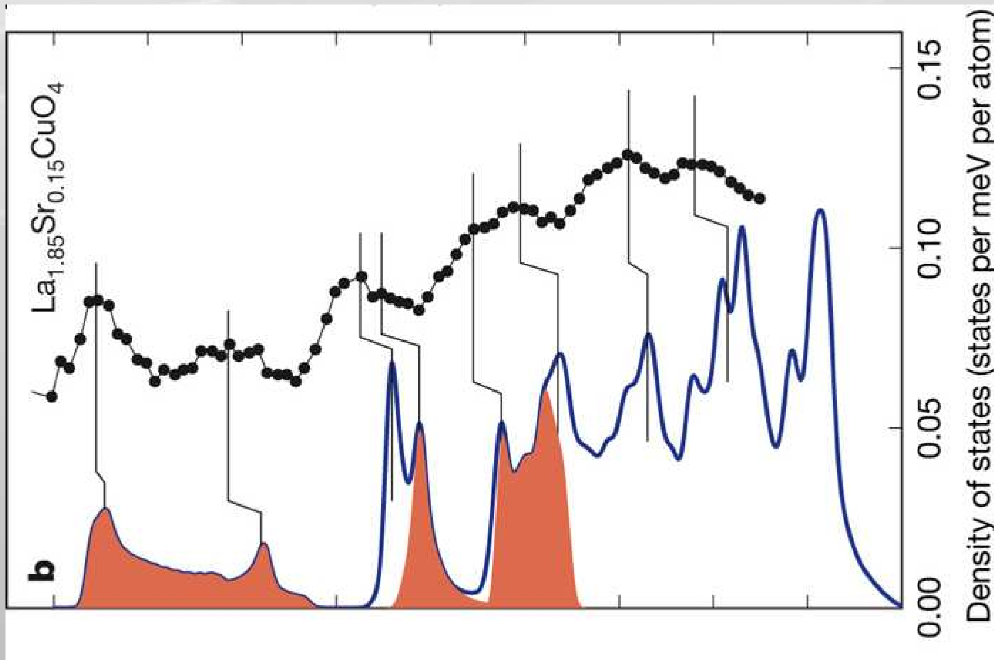
Chen, G. F., et al. Phys. Rev. Lett. **100** 247002 (2008).

Striking similarities with cuprates:

- Layered compounds
- Doping induce high-T_C superconductivity (T_C max ~55K)
- Strong electronic correlations
- proximity of SC and magnetic phases

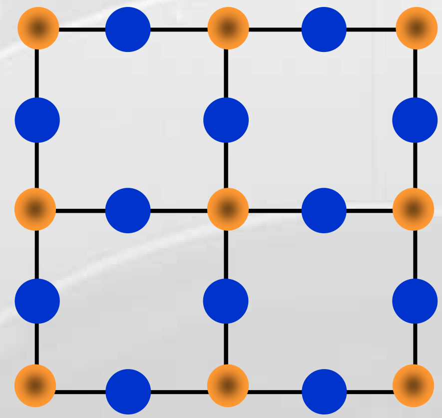
Lattice dynamics in cuprates

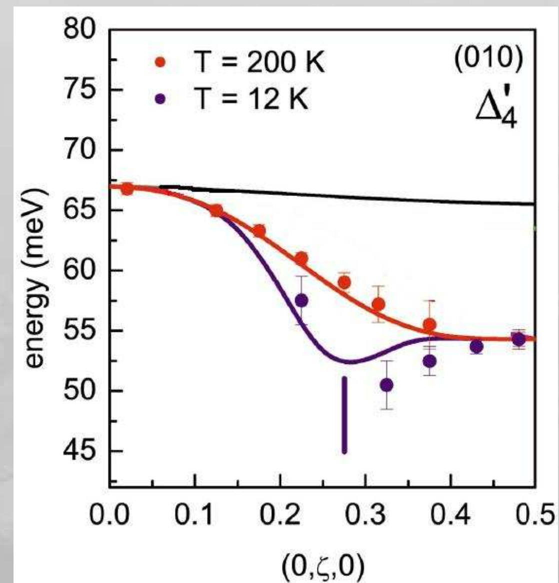
Strong correlations:
Hard comparison with LDA calculation



Giustino et al., Nature **452** 790 (2008)

Ex: half breathing mode softening and its relation to the kink/CO ?



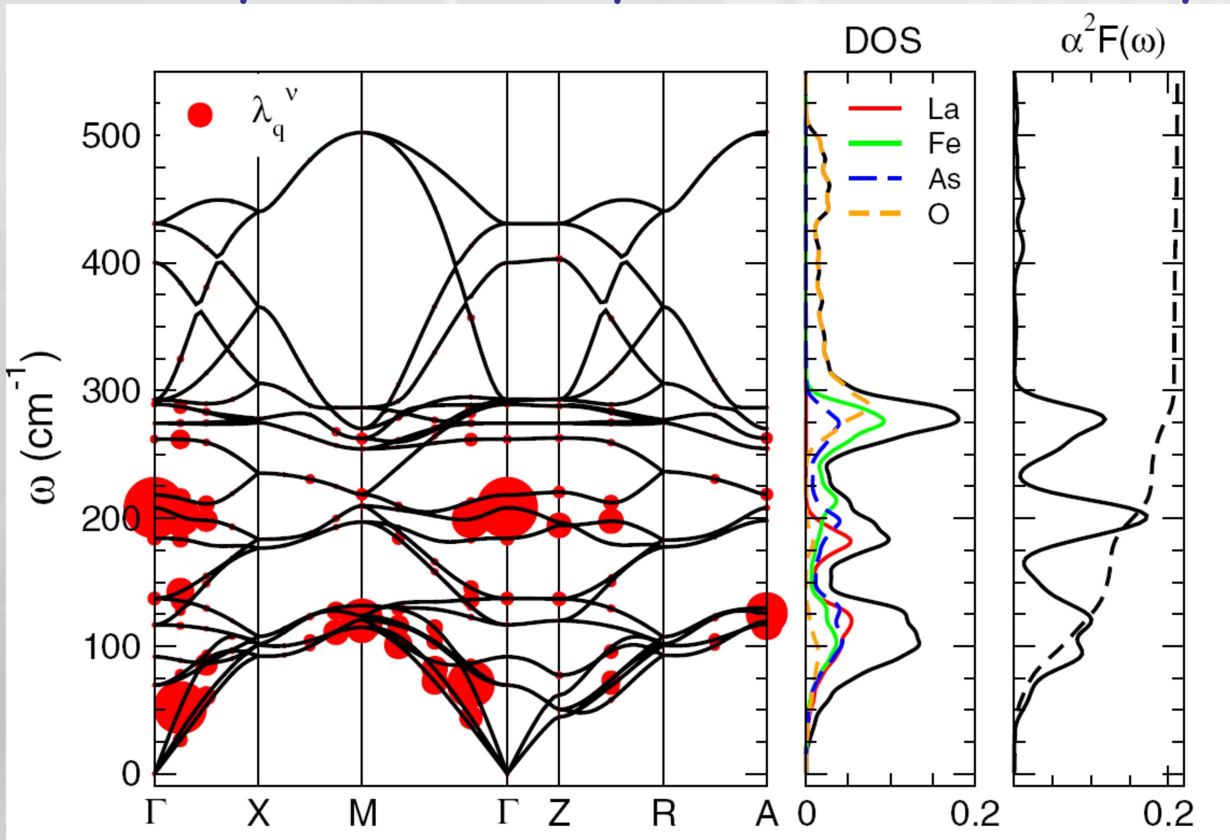


Pintschovius et al., PRB **69** 214506 (2008)

Lattice dynamics in oxy-pnictides

Are these compounds conventional e-ph superconductors ?

Density Functional perturbation theory



L. Boeri, et al. Phys. Rev. Lett. 101, 026403 (2008)

Electron-phonon interaction cannot explain T_c > 0.8 K

Measuring the Generalized Phonon Density of States

New compounds: few high quality single crystals availables

⇩

Powders

⇩

dispersion of phonon branches is not accessible

Averaging IXS measurements on powders over a large range of Q gives:

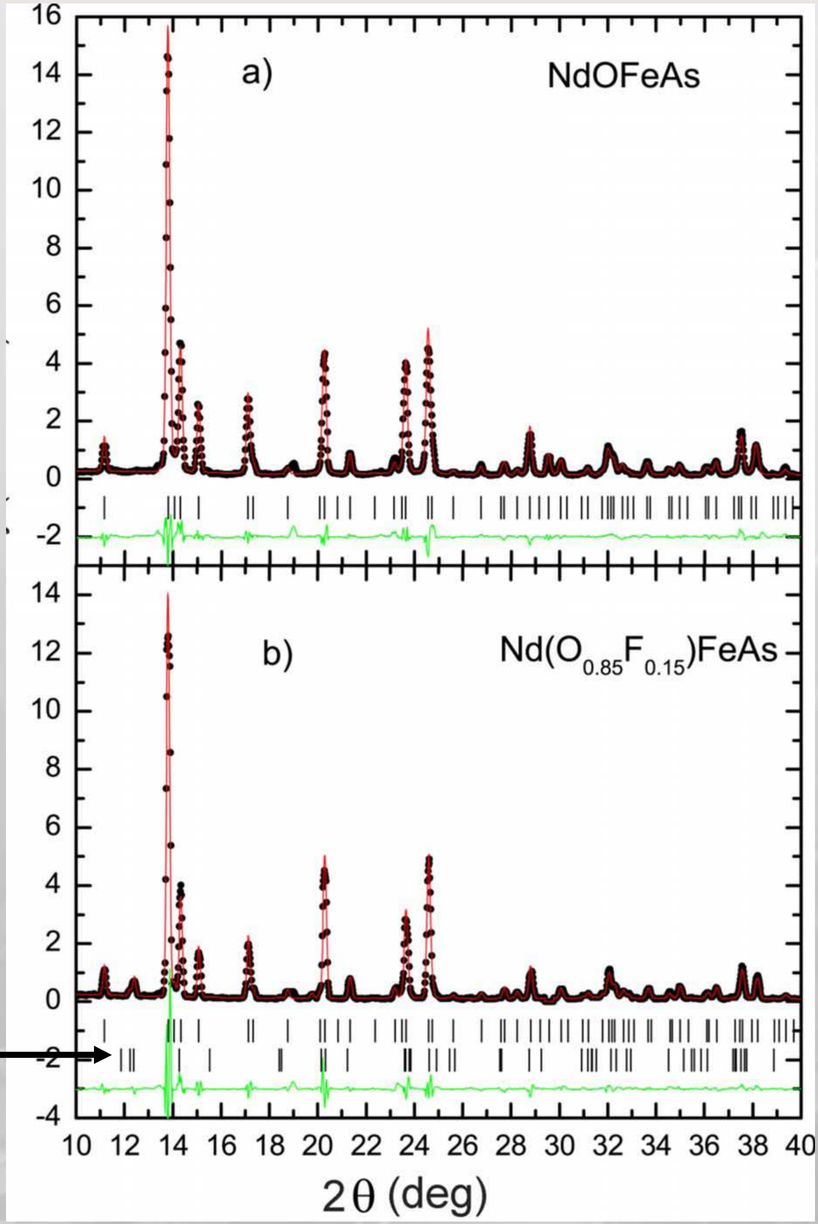
$$I(\omega) = A\langle Q^2 \rangle [1 + n(\omega, T)] \sum_n \frac{e^{-2W_n} f_n(\langle Q^2 \rangle)}{M_n \omega} G_n(\omega)$$

Partial phonon density-of-states

A. Bosak and M. Krisch, PRB 72, 224305 (2005)

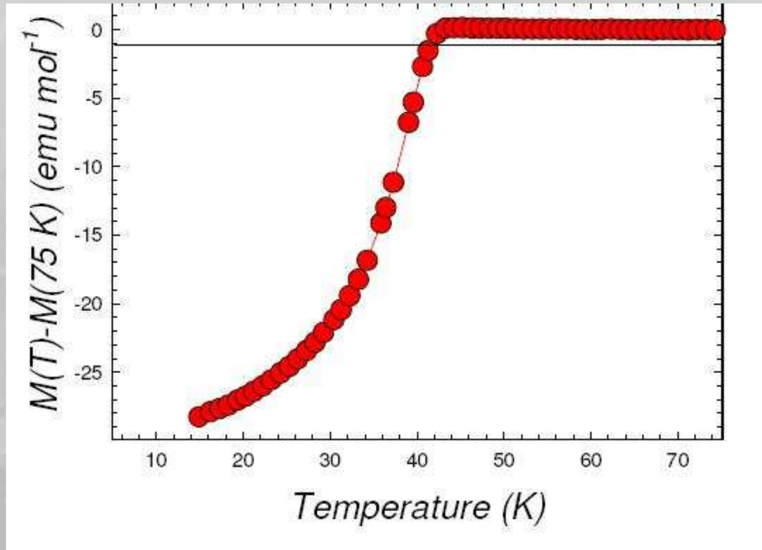
$NdFeAs(O_{1-x}F_x)$ Sample characterization

XRD pattern recorded on SNBL
$\lambda = 0.7$ Å



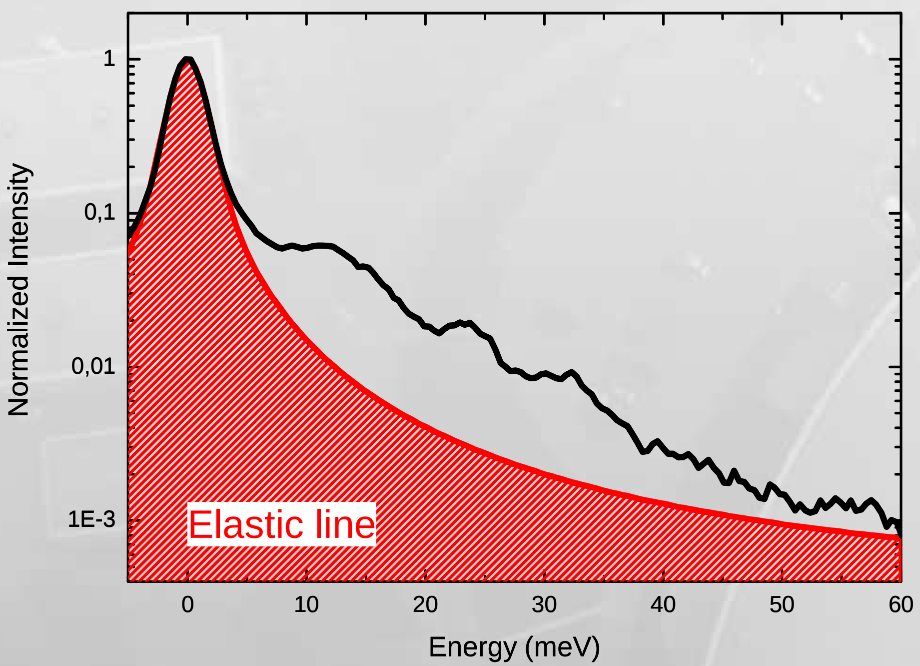
4% of NdOF

$NdFeAsO_{0.85}F_{0.15}$
$T_C = 42$ K



Generalized Phonon Density of States in $NdFeAs(O_{1-x}F_x)$

Q-sampling on NdFeAsO: 36 spectra between 53 nm^{-1} to 73 nm^{-1}



- Elastic line suppression
- Bose factor
- Multiphonon contribution

M. Le Tacon, et al. PRB 78, R140505 (2008)

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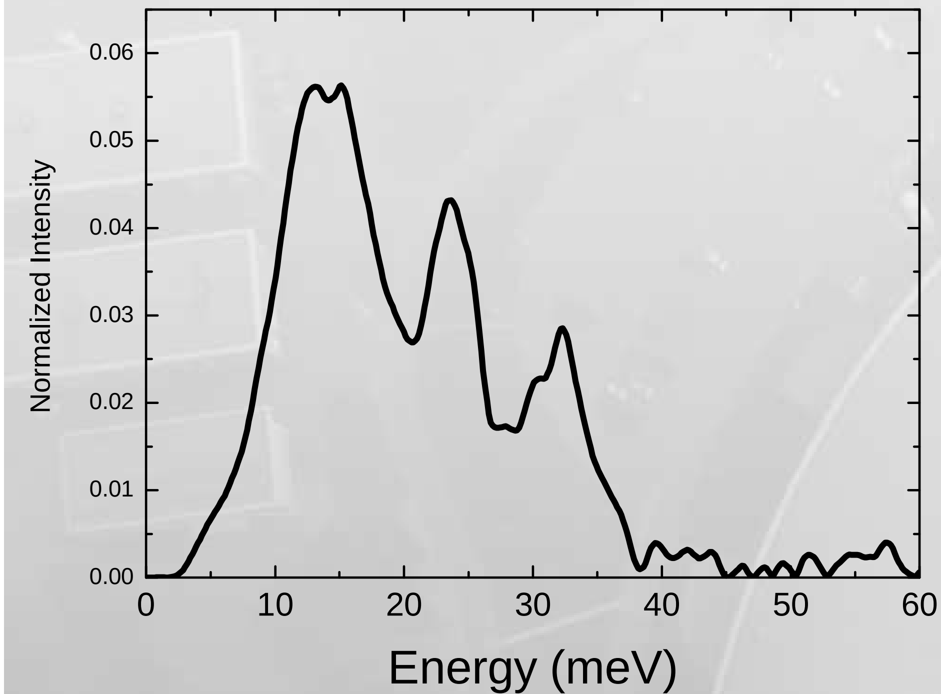

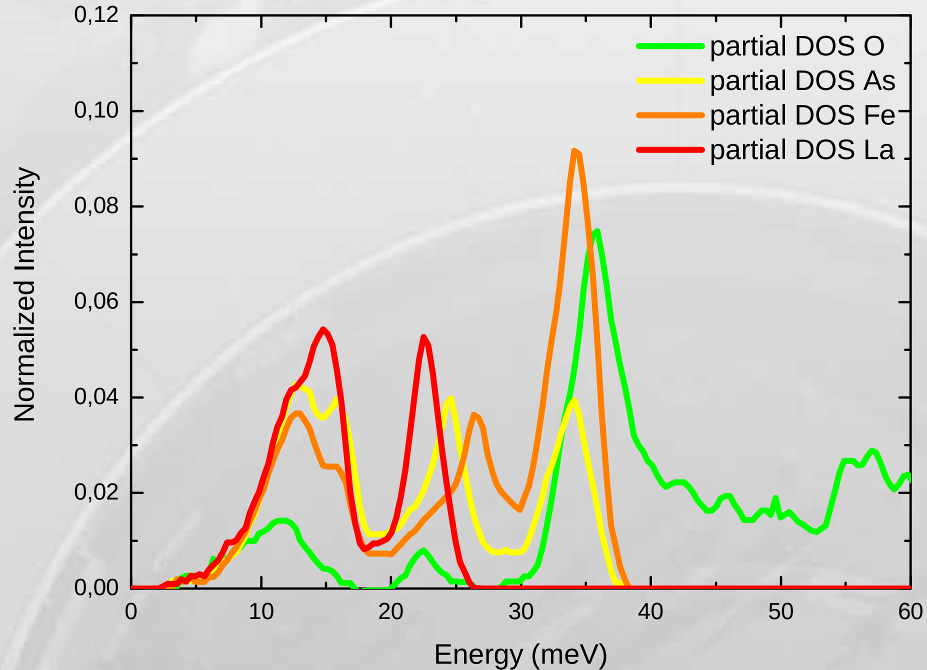

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- Weighting of the partial PDOS
- Convolution by the resolution function

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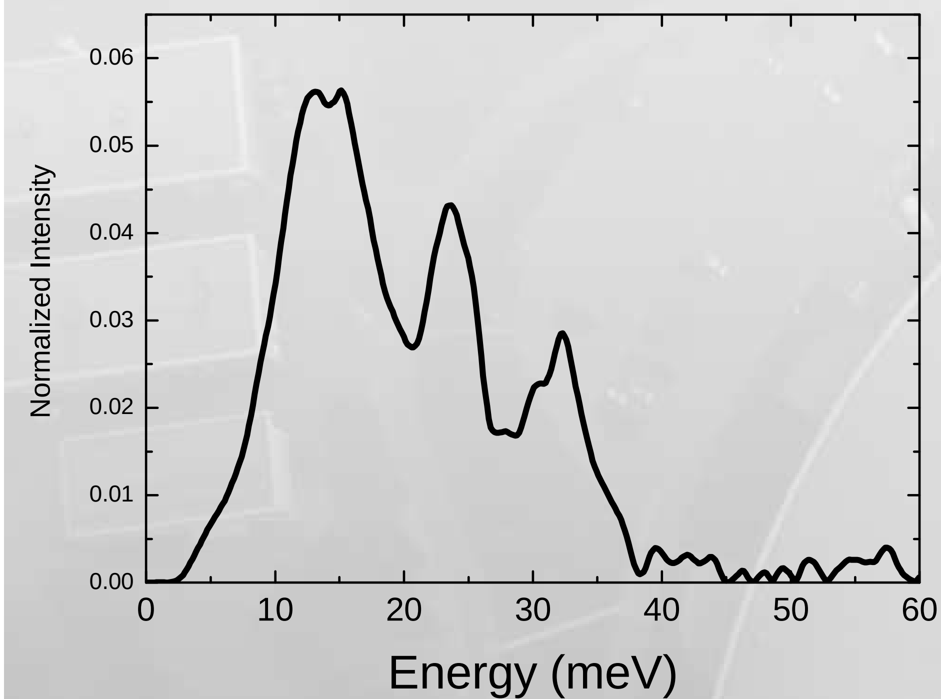

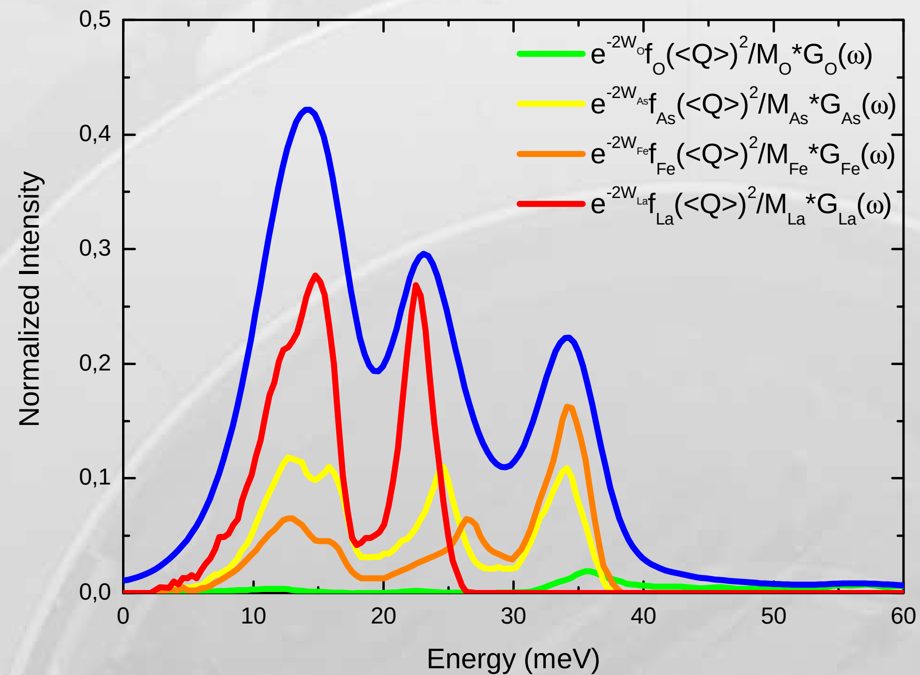

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Generalized Phonon Density of States in NdFeAs($O_{1-x}F_x$)

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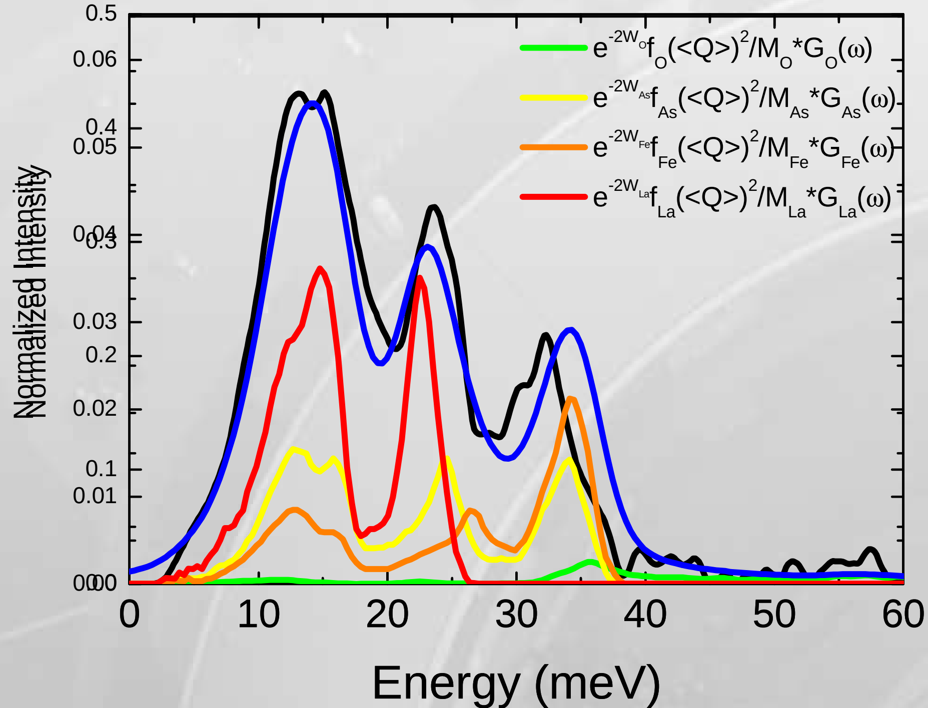


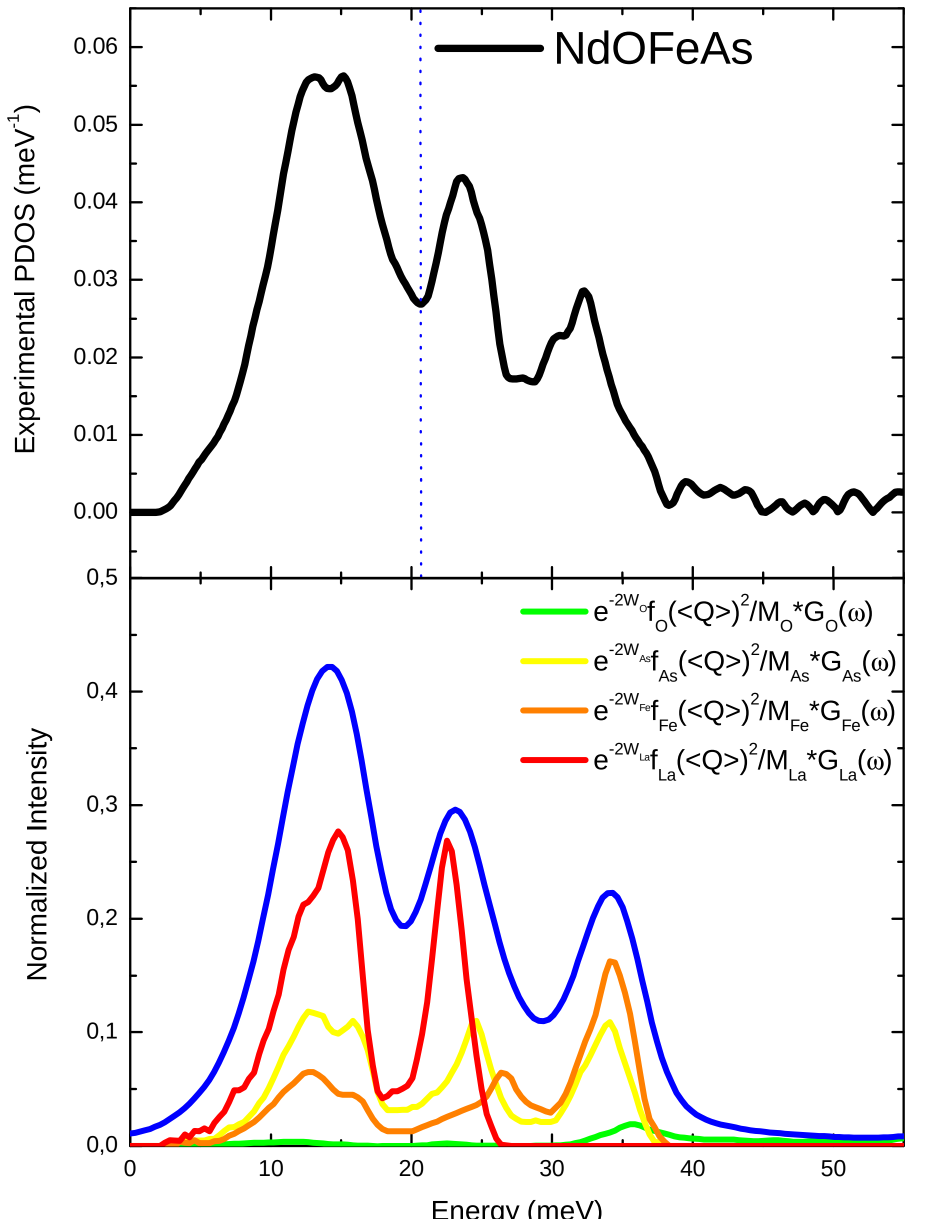
Surprising (?) overall good agreement,

third peak shifted by ~10%

(cf. Fukuda et al., JPCS 77 103715 (2008))

Softening by 30% of the Fe-As force constant

Generalized Phonon Density of States in $NdFeAs(O_{1-x}F_x)$



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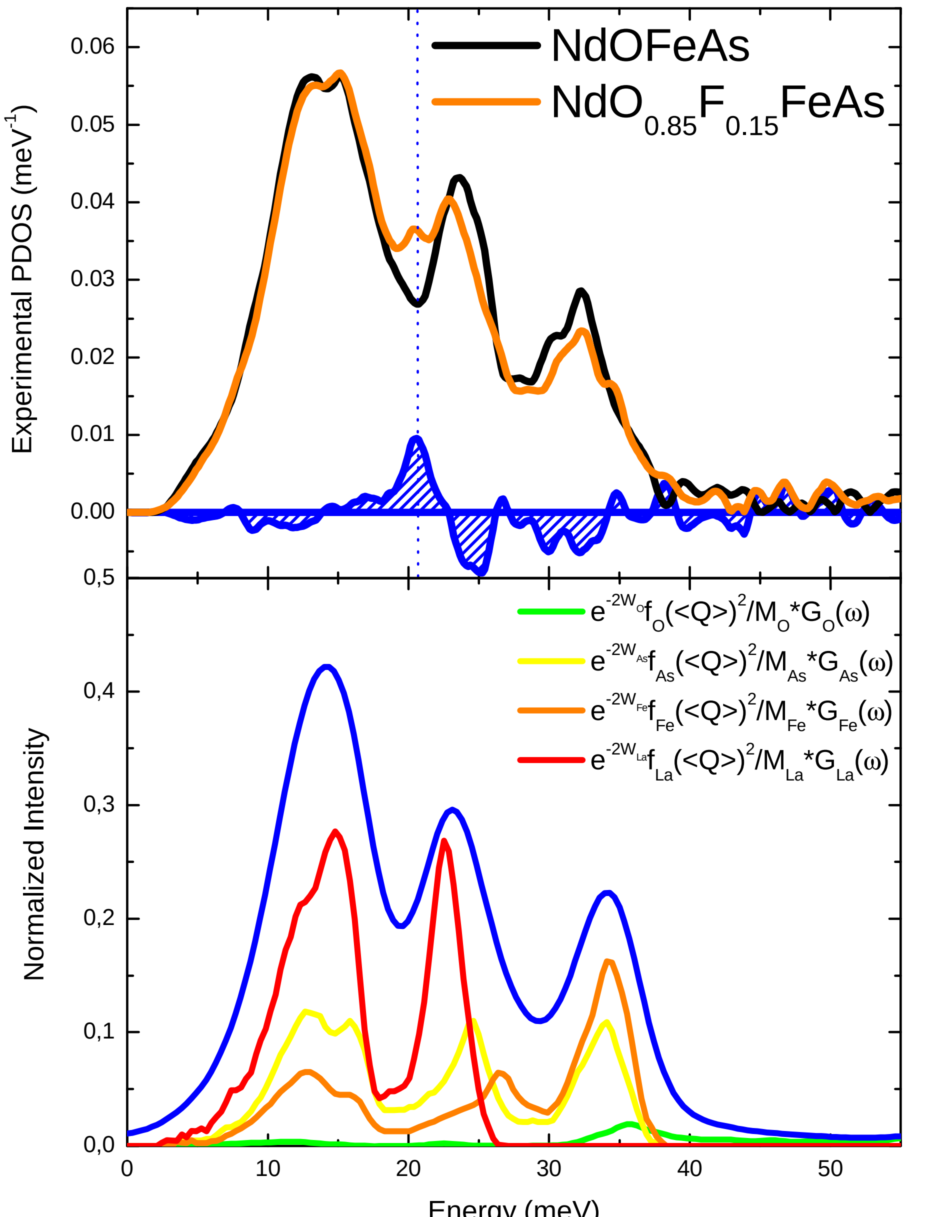

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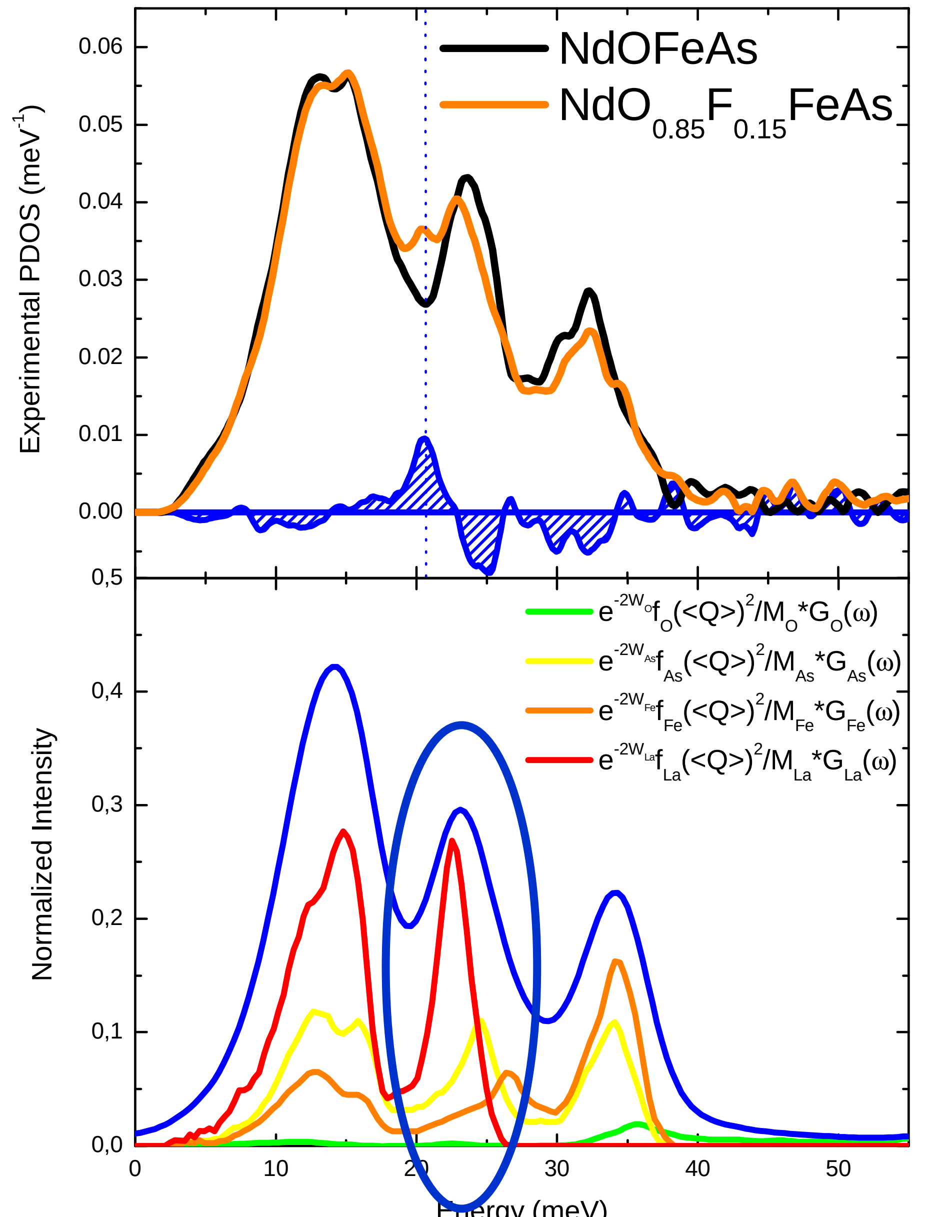
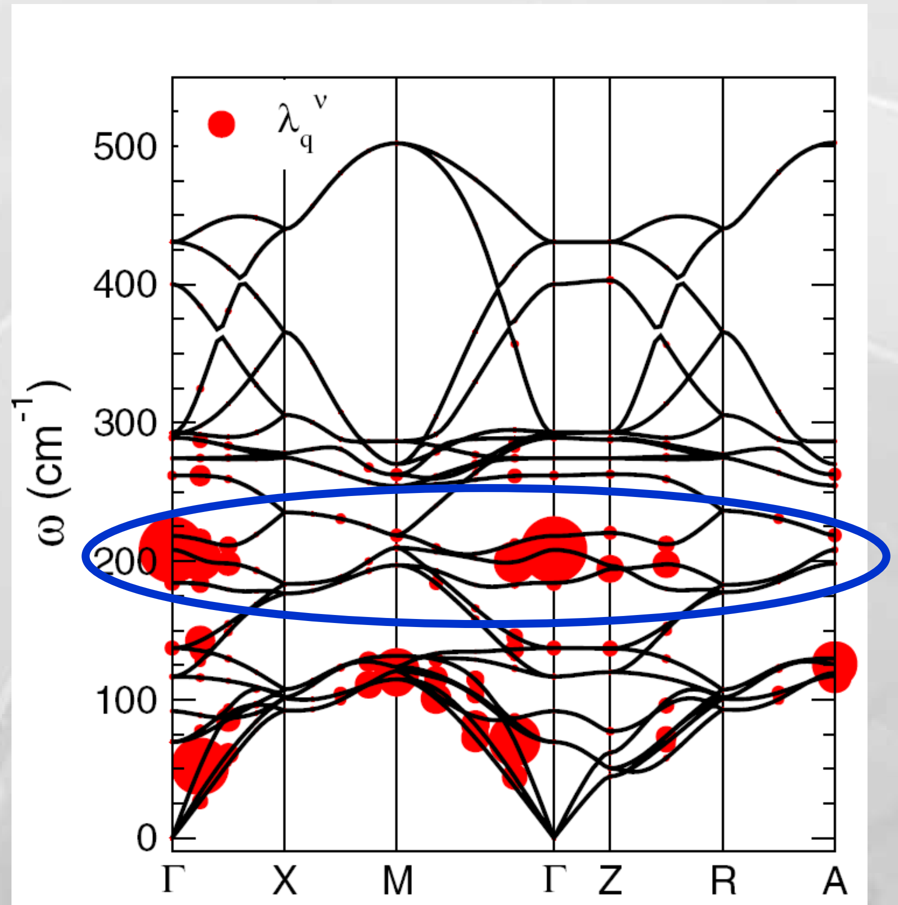
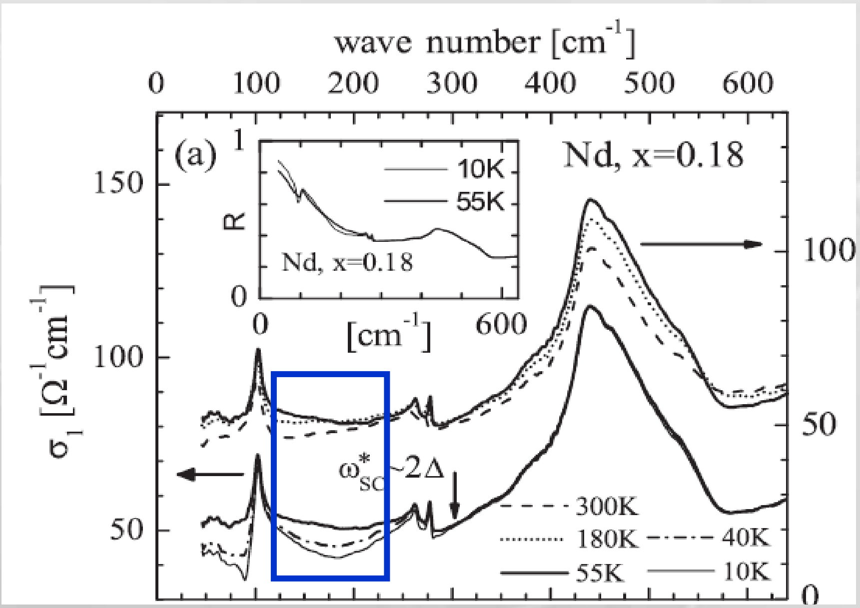


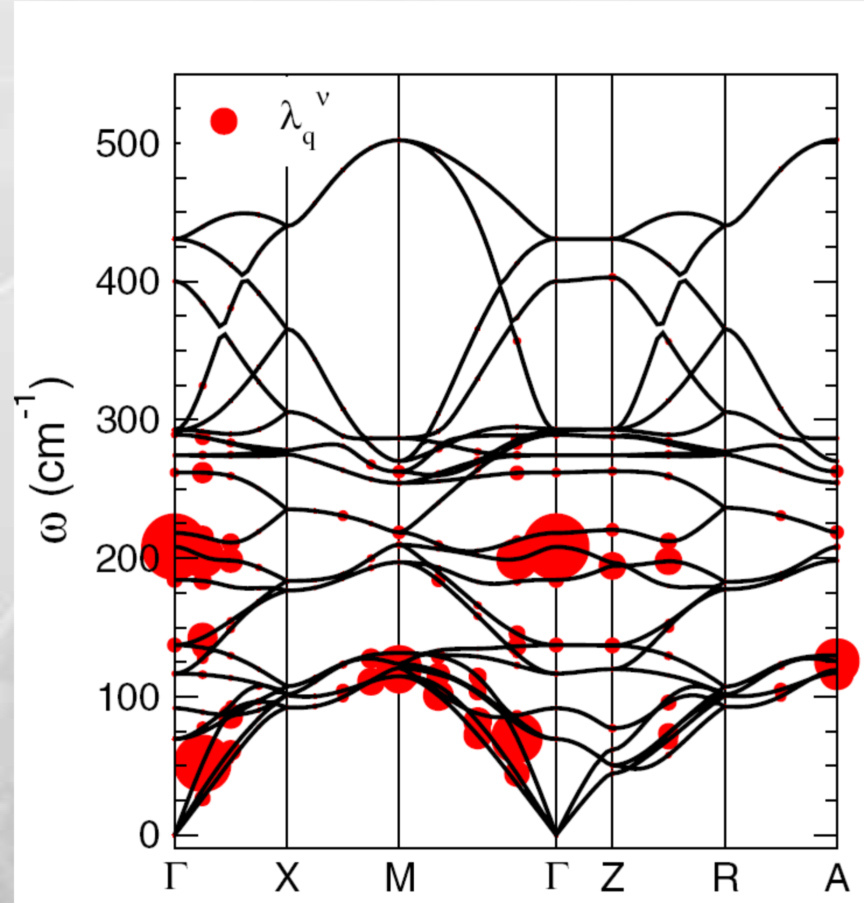


Softening of at least 3 meV of one of this 3 branches !!!

Generalized Phonon Density of States in $NdFeAs(O_{1-x}F_x)$

IR phonons in $NdO_{0.82}F_{0.18}FeAs$ (Dubroka et al, PRL 101, 097011)

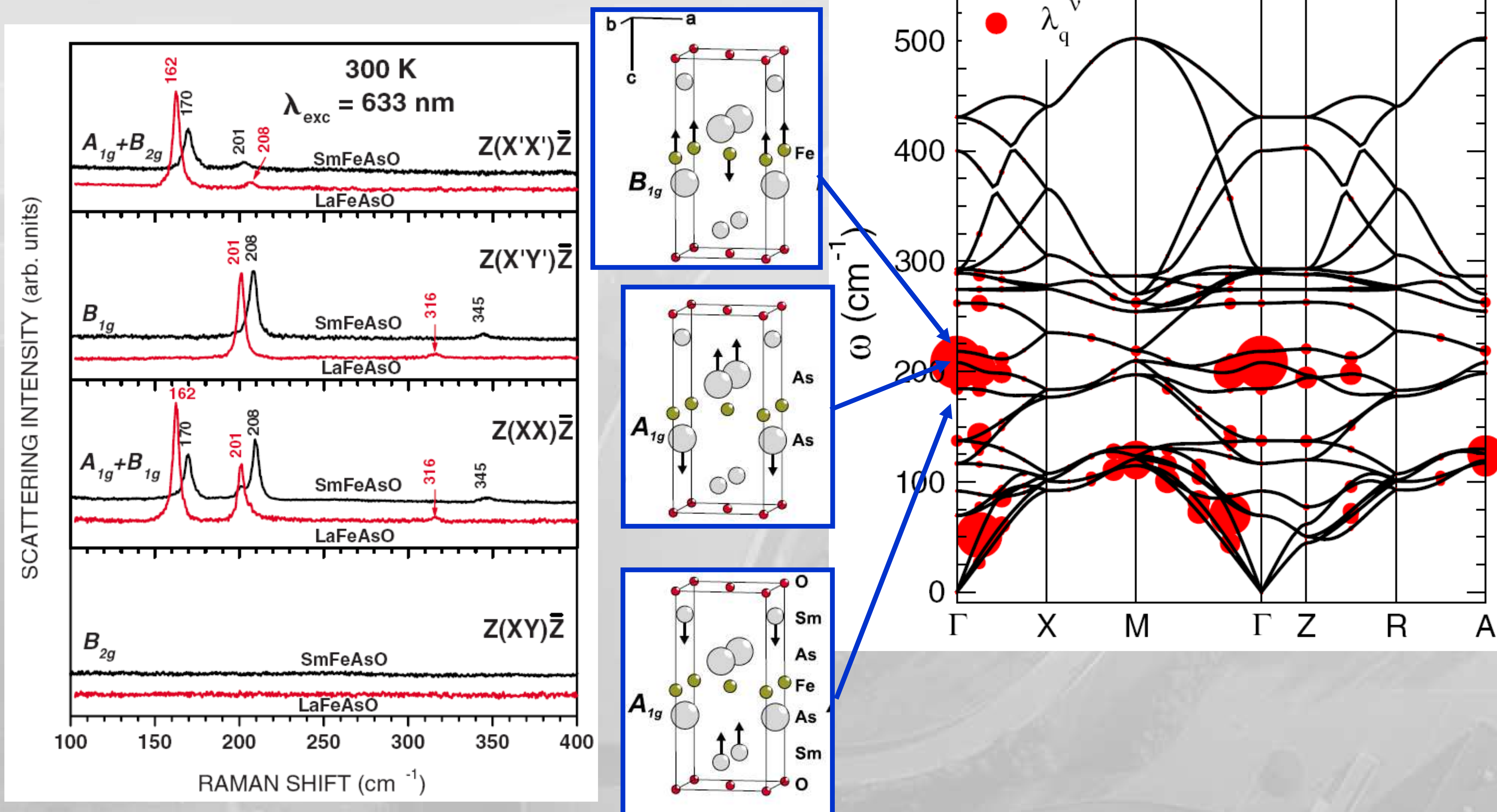




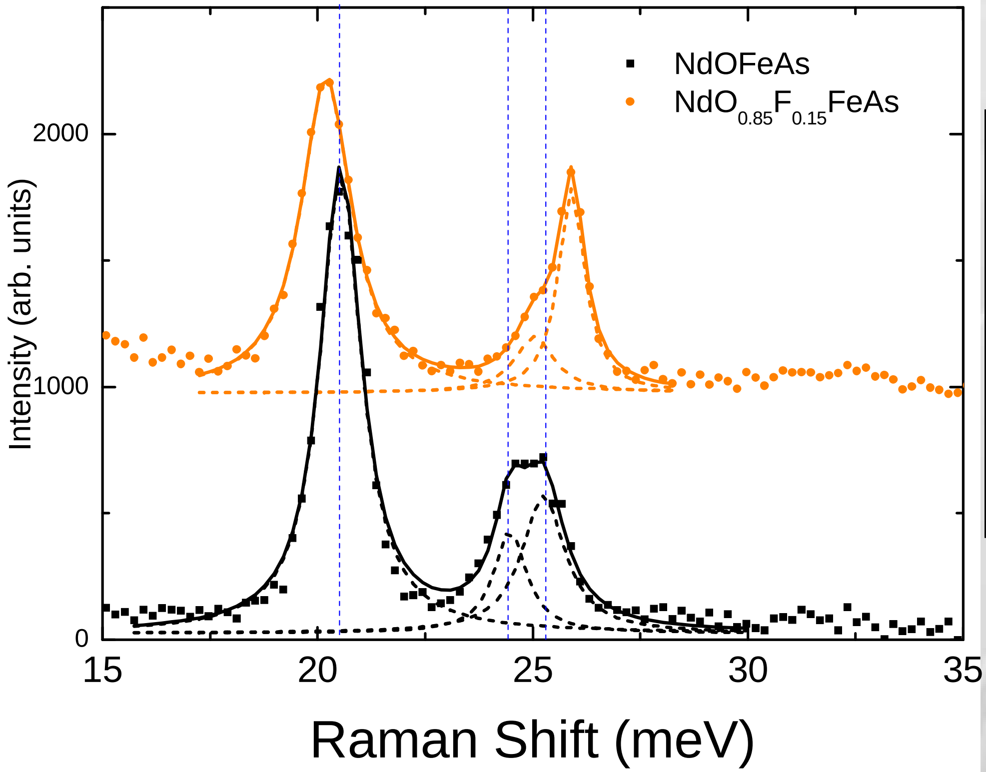
Generalized Phonon Density of States in $NdFeAs(O_{1-x}F_x)$

Raman scattering in LaFeAsO and SmFeAsO

(Hadjiev et al. PRB 77 R220505(2008))



Effect of doping on Raman Spectra



Raman phonon frequencies

phonon	NdOFeAs	$NdO_{0.85}F_{0.15}FeAs$
A_{1g} (Nd)	20.5 meV	20.2 meV
A_{1g} (As)	25.3 meV	25.9 meV
B_{1g} (Fe)	24.5 meV	25.1 meV

- Hardening of the Fe(B_{1g}) & As (A_{1g})branches
- Small softening of the Nd(A_{1g}) branch

Le Tacon et al., PRB 78, R140505 (2008)
Gallais et al., PRB 78 132509 (2008)

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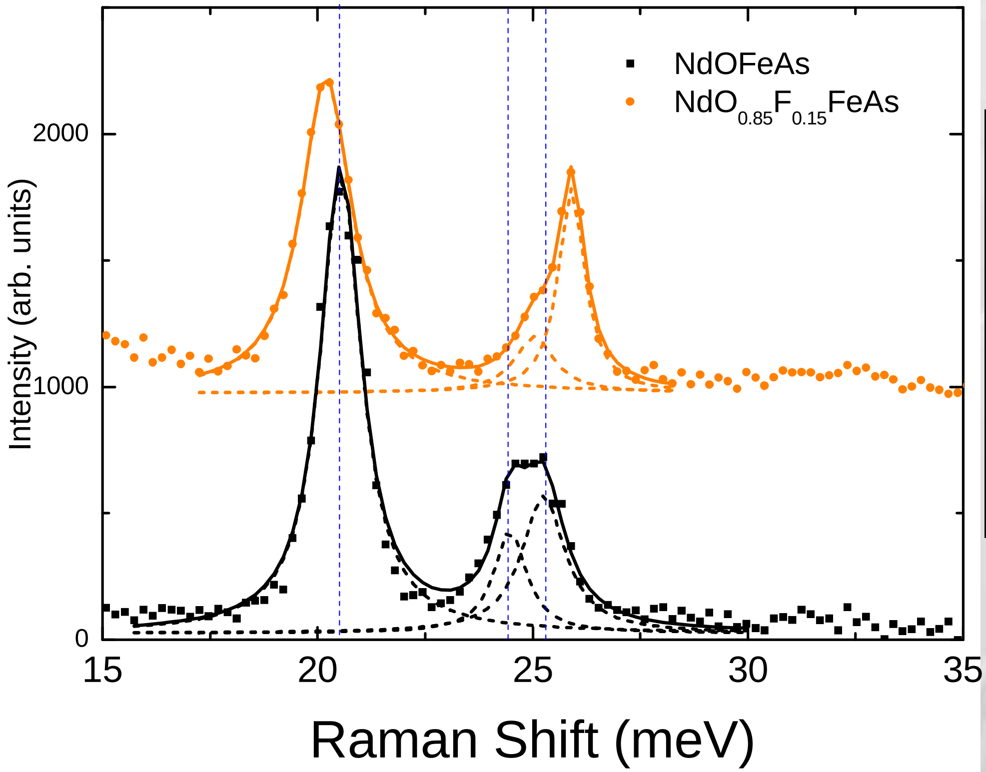

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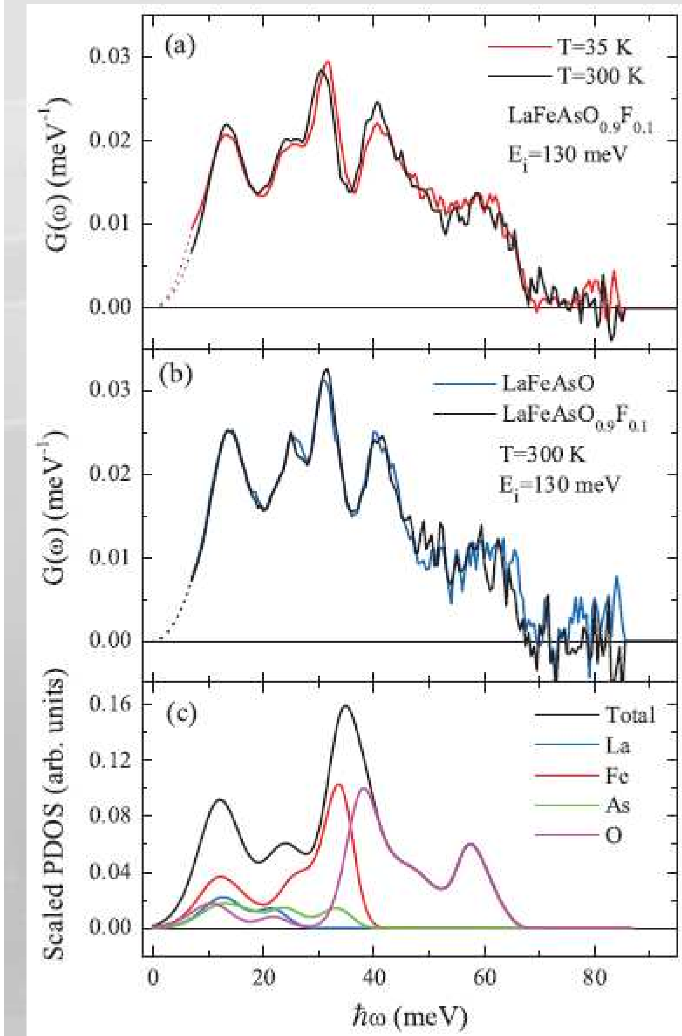
No zone center softening can explain the observed effect !

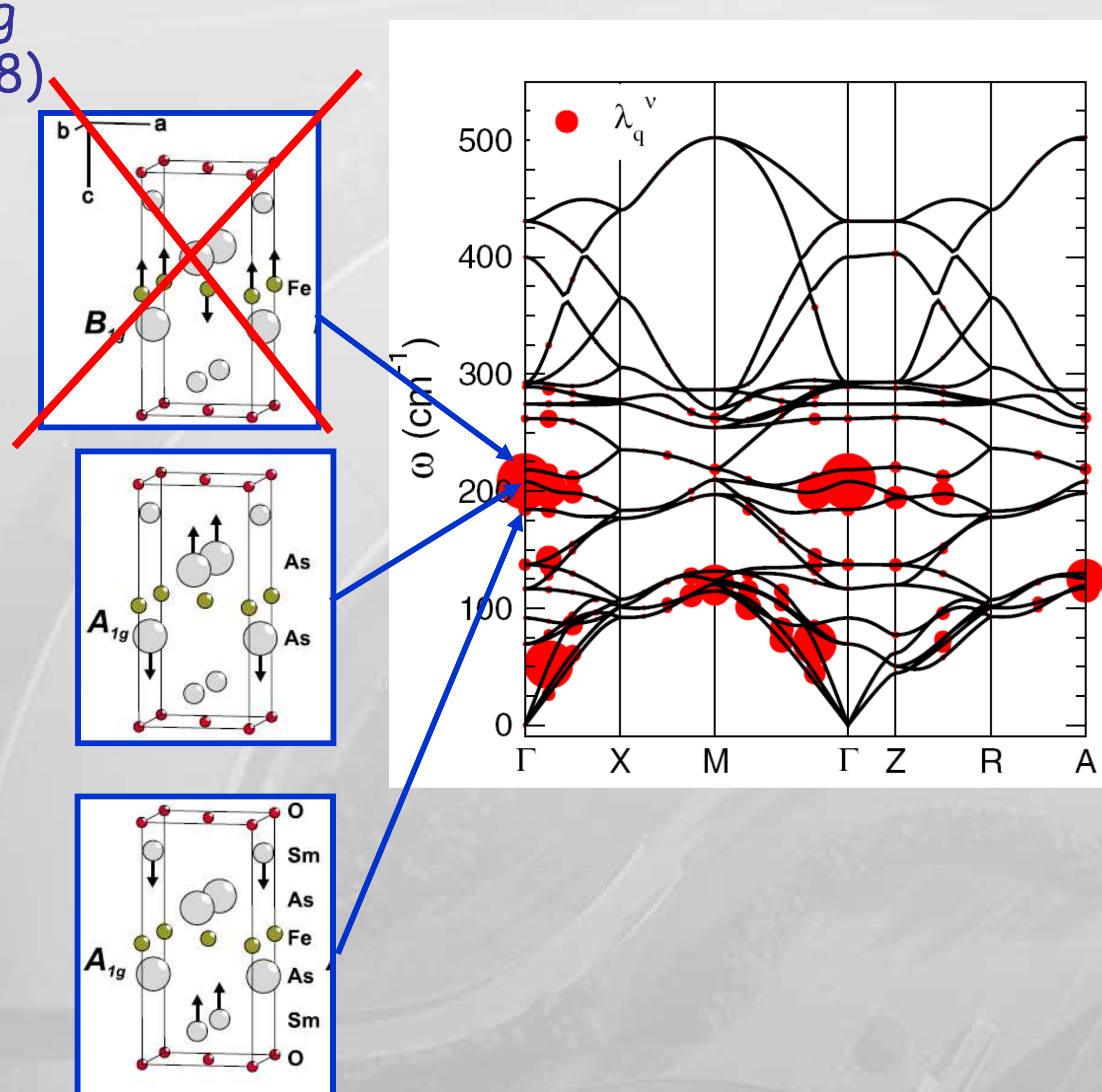
One has to search at finite Q in the dispersion.

Le Tacon et al., PRB 78, R140505 (2008)
Gallais et al., PRB 78 132509 (2008)

Any clue from other techniques ??

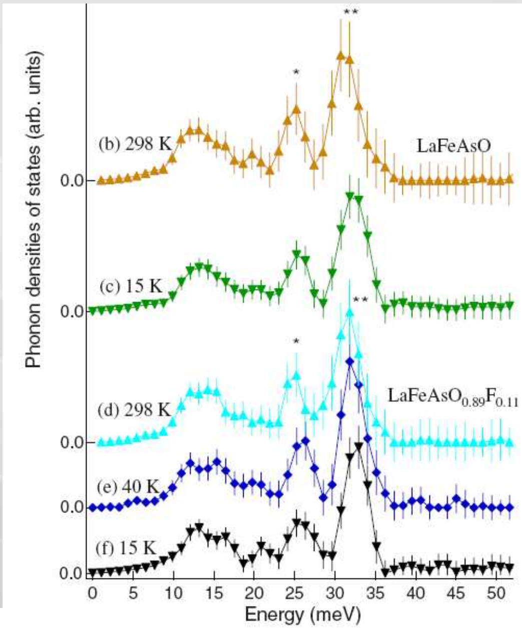
Inelastic Neutron Scattering
(Christianson et al., PRL 2008)

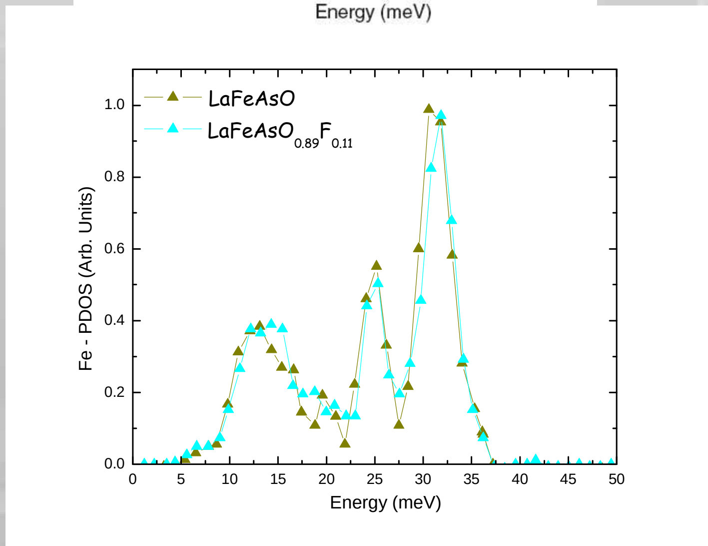


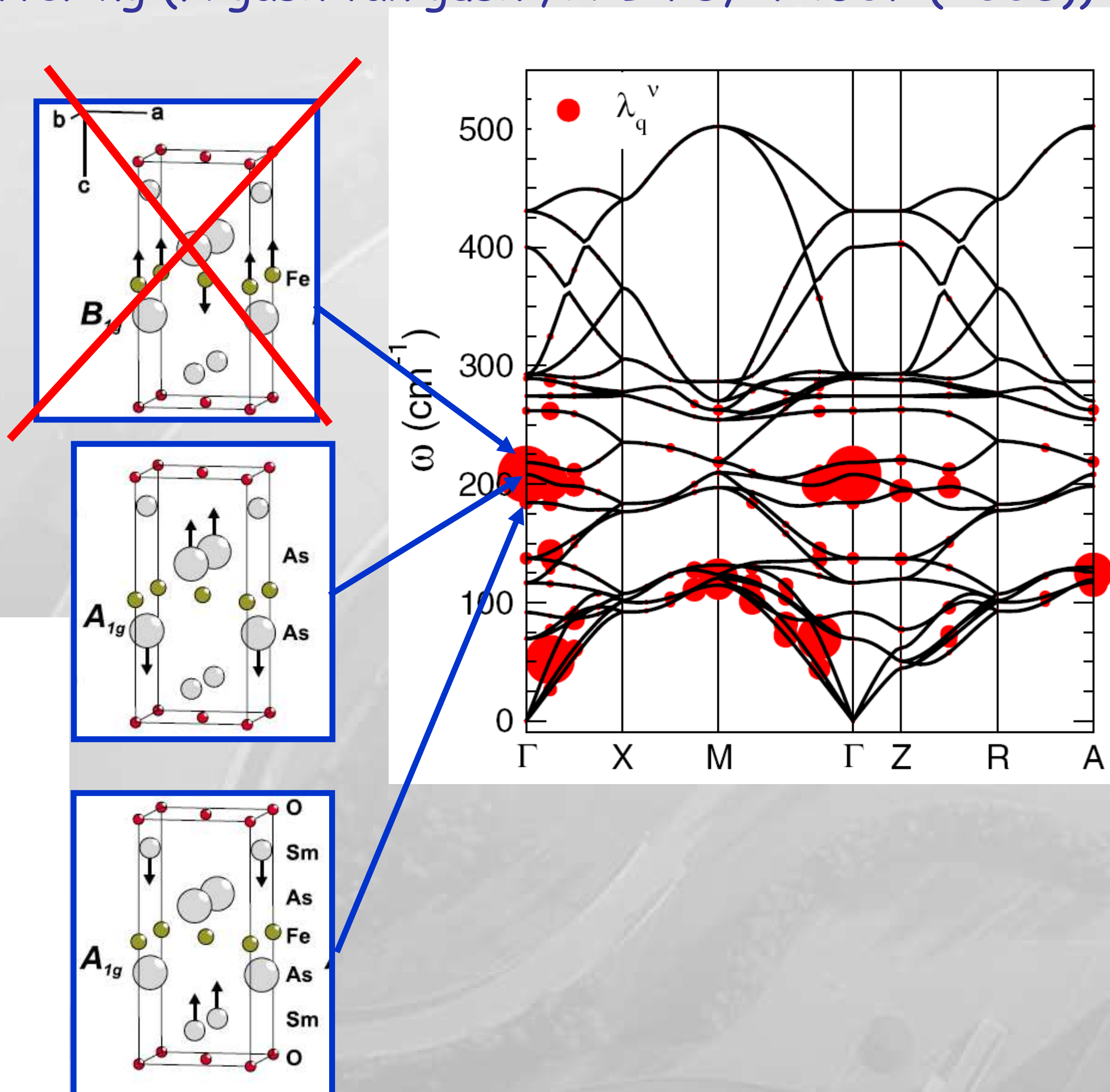


Any clue from other techniques ??

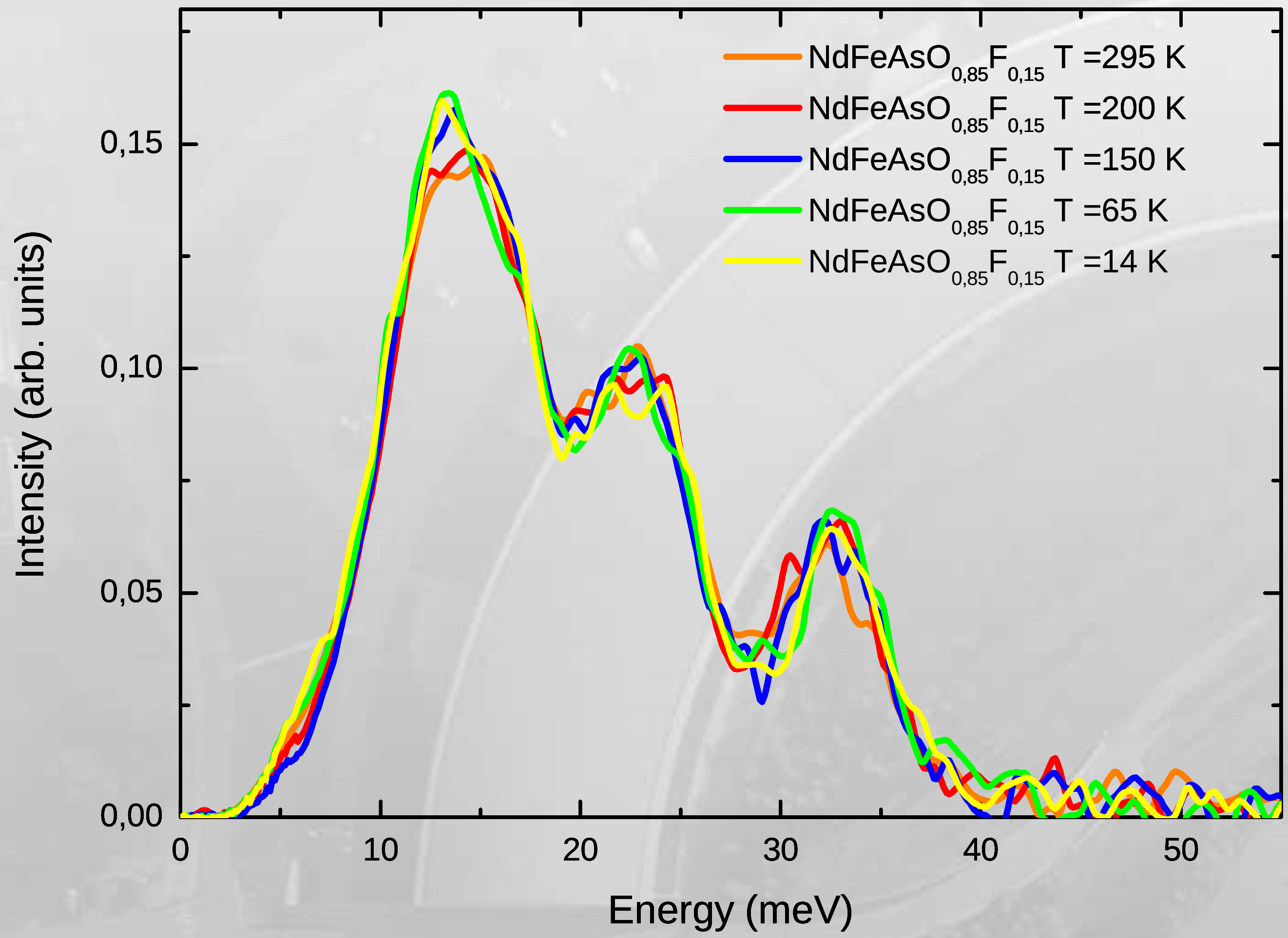
Nuclear Resonant Inelastic Scattering (Higashitanigushi, PRB 78, 174507 (2008))







T- dependence and relation to superconductivity ?



Conclusions

- Good agreement with simple LDA calculations:
strong correlations ??

- Lattice dynamics in iron-based oxypnictides is probably not as boring as first expected …

- Role of the rare earth ?

- Investigations in single crystals are now in progress …