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Geophysics Studies with High-resolution X-ray Spectroscopy

Wolfgang Sturhahn
(sturhahn@anl.gov)

U.S. Department of Energy

UChicago Argonne LLC

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Inelastic x-ray and nuclear resonant scattering group:

Ahmet Alatas
E. Ercan Alp
Svetlana Kharlamova
Michael Lerche *(now at HPsynC, Carnegie Institution of Washington)*
Bogdan Leu
Thomas S. Toellner
Jiyong Zhao

California Institute of Technology:

Jennifer M. Jackson

Lawrence Livermore National Laboratory:

Jung-Fu Lin

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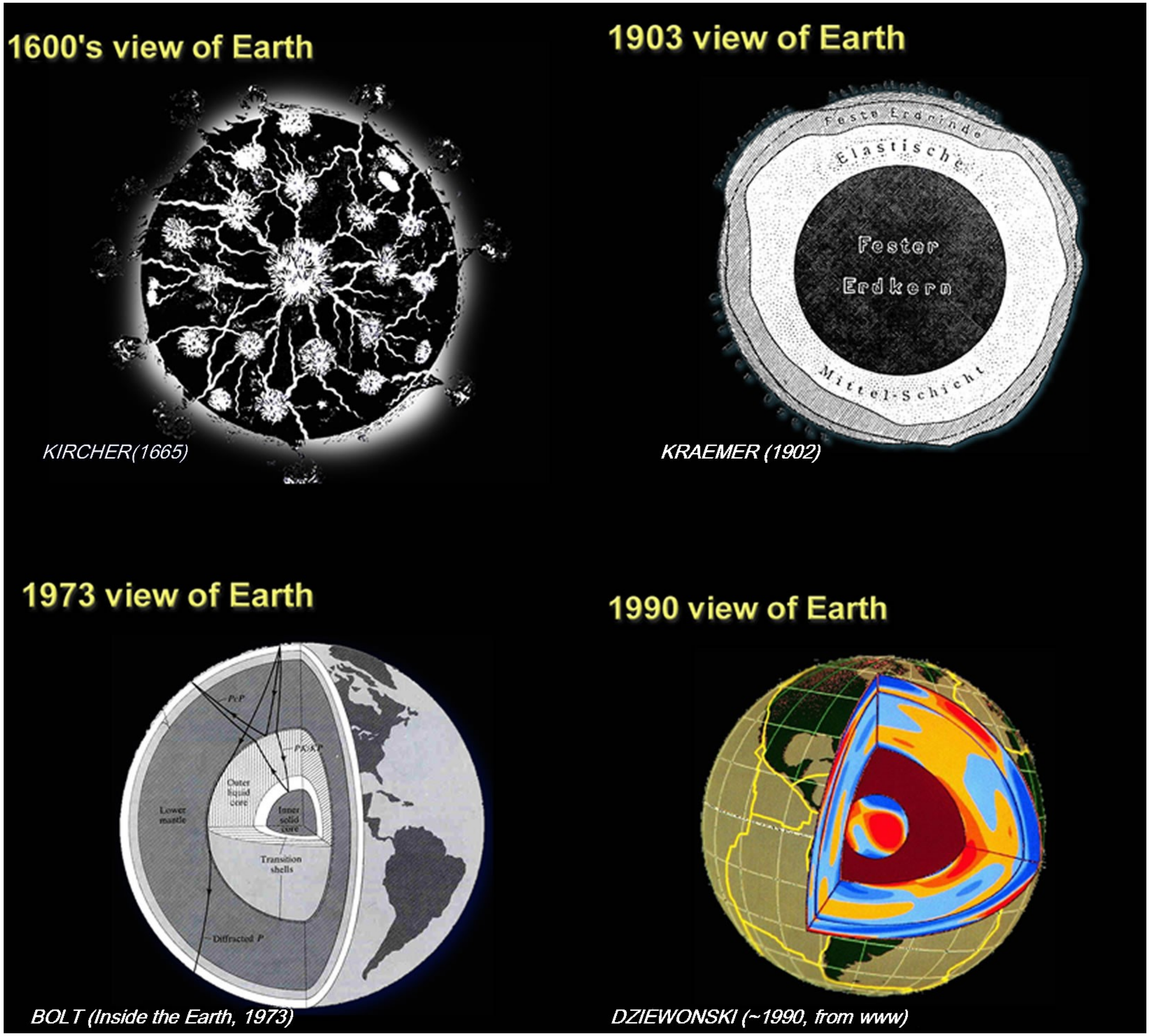
Models of Earth's interior:



- ✫ seismic studies
- ✫ gravity and magnetic fields
- ✫ cosmo-chemical models
- ✫ geodynamical modeling

- ✫ material properties, e.g.,
  - sound velocities & elasticity
  - density
  - viscosity

IXS techniques:

Technique		probed excitation	resolution	APS location
HERIX	momentum-resolved high-resolution IXS	phonon dispersion	1-5 meV	3-ID, 30-ID
MERIX	momentum-resolved medium-resolution IXS	electrons near Fermi surface	100-300 meV	9-ID, 30-ID
NRIXS	momentum-integrated nuclear resonant IXS	phonon density of states	1 meV	3-ID, 16-ID
XRS	x-ray Raman spectroscopy	core electron release in low-Z elements	0.1-1 eV	9-ID, 13-ID, 16-ID, 17-ID, 20-ID, 30-ID
XES	x-ray emission spectroscopy	core-valence electron transitions	0.5-1 eV	10-ID, 13-ID, 16-ID
PEEM	photo-electron emission microscopy	electron energy levels	5 meV	4-ID
CS	Compton scattering	electron energy levels	1 eV	
MCS	magnetic Compton scattering	spin-polarized electron energy levels	100 eV	

High-resolution IXS methods:

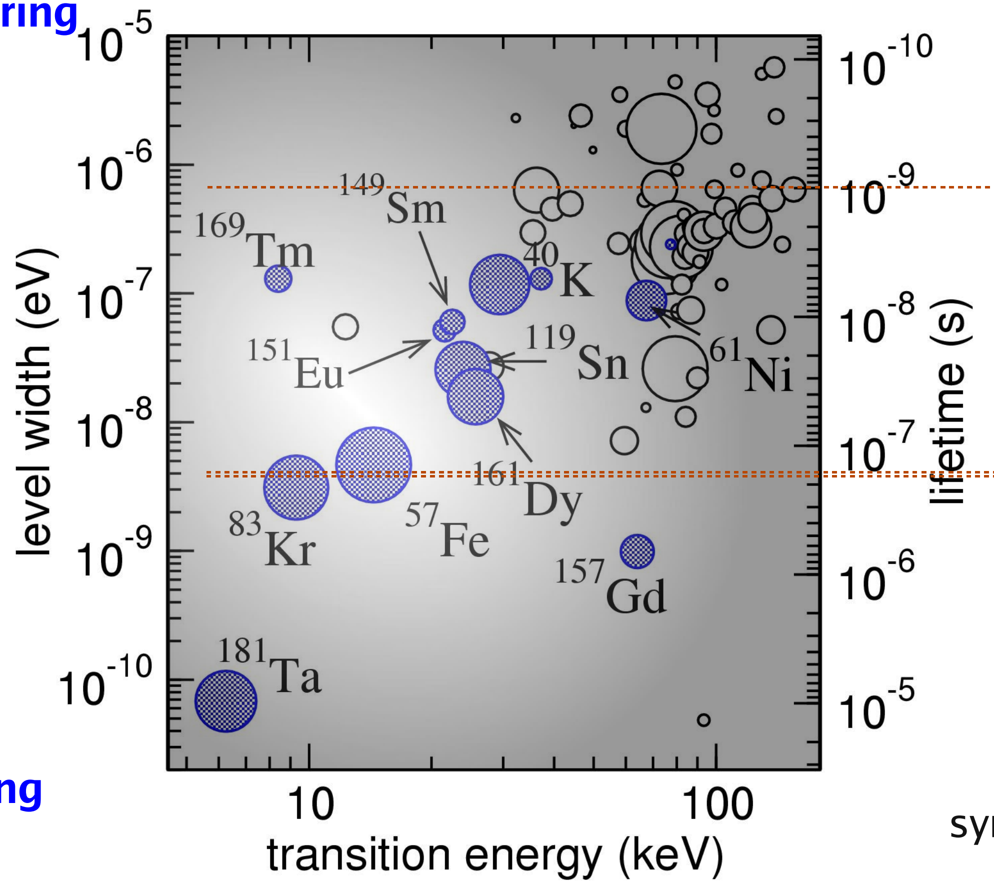
- **HERIX – High-Energy Resolution Inelastic X-ray scattering**
  - ✰ phonon dispersion relations
  - ✰ applications include determination of sound velocities and dynamics in liquids

- **NRIXS – Nuclear Resonant Inelastic X-ray Scattering (a.k.a. NRVS and NIS)**
  - ✰ local vibrational density of states
  - ✰ applications include determination of sound velocities and thermodynamic properties

- **SMS – Synchrotron Mössbauer Spectroscopy (a.k.a. NFS)**
  - ✰ internal magnetic fields, electric field gradients, isomer shifts
  - ✰ applications include magnetic phase transitions, determination of spin & valence states, and melting studies

Isotopes for nuclear resonant scattering:

larger scattering strength



detector resolution

APS, ESRF bunch separation

easier timing

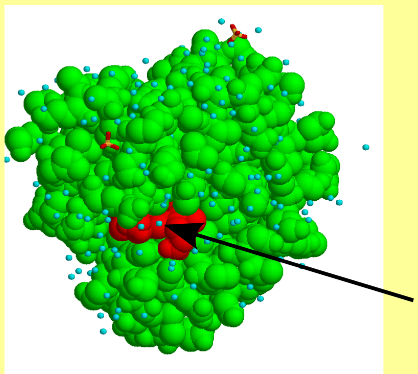
symbol area is proportional to the nuclear resonant cross section

more absorption

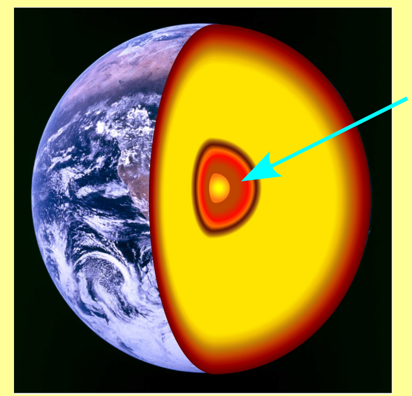
less intensity

NRS Target applications:

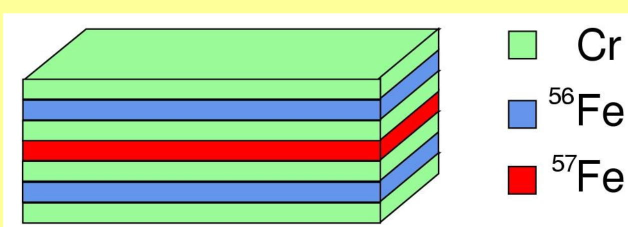
- **perfect isotope selectivity & complete suppression of non-resonant signals**
- **excellent sensitivity -- 10^{12} nuclei in the focused beam (~150pg iron metal)**



☆ proteins and other large molecules



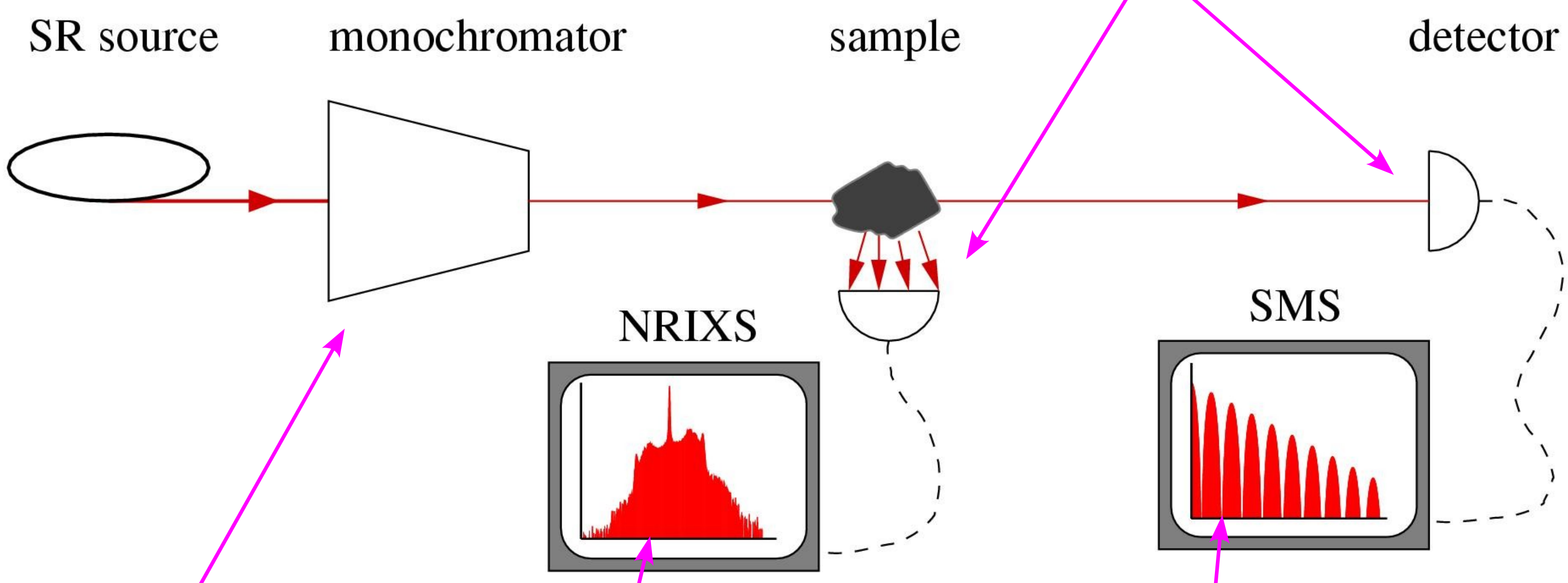
☆ materials under high pressure



☆ nano-structures

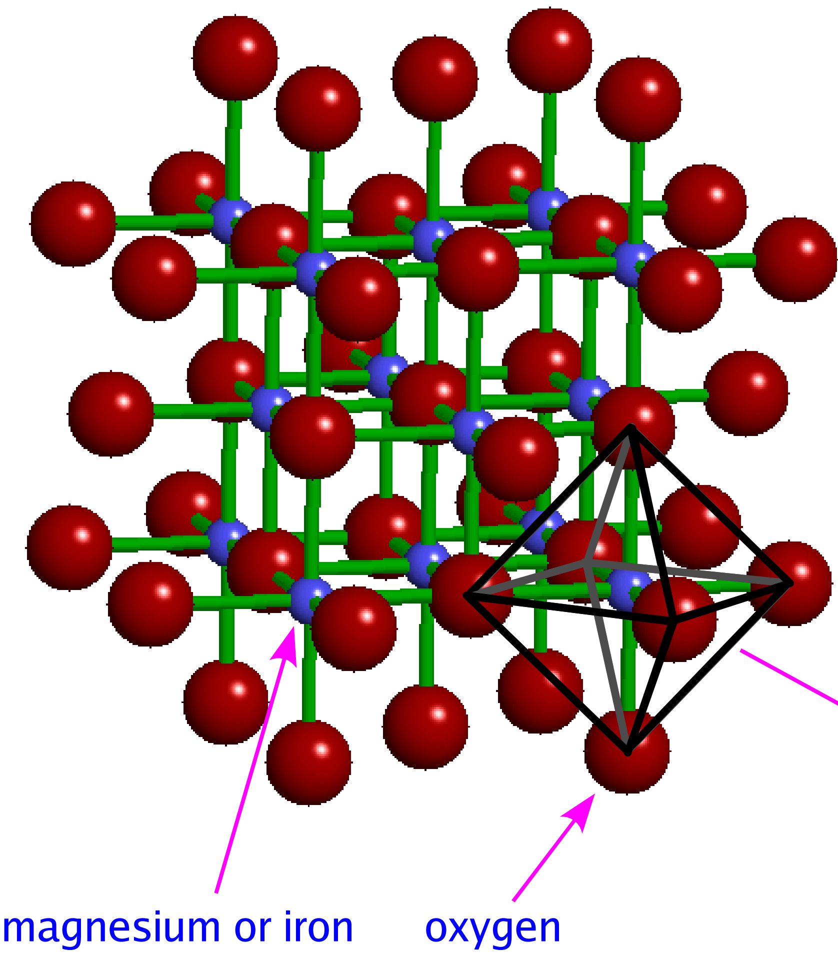
Experimental setup for NRS:

- x-ray pulses must be sufficiently separated in time
- detectors must have good time resolution and excellent dynamic range

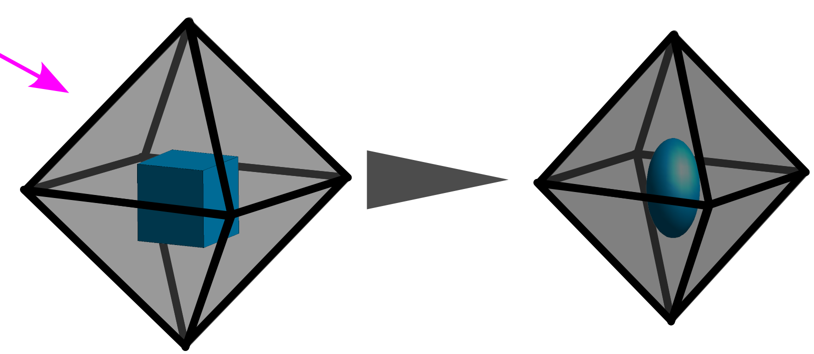


- phonon spectrum
- time spectrum
- monochromatization to meV-level required
- energy is tuned around nuclear transition

Structure of Periclase:

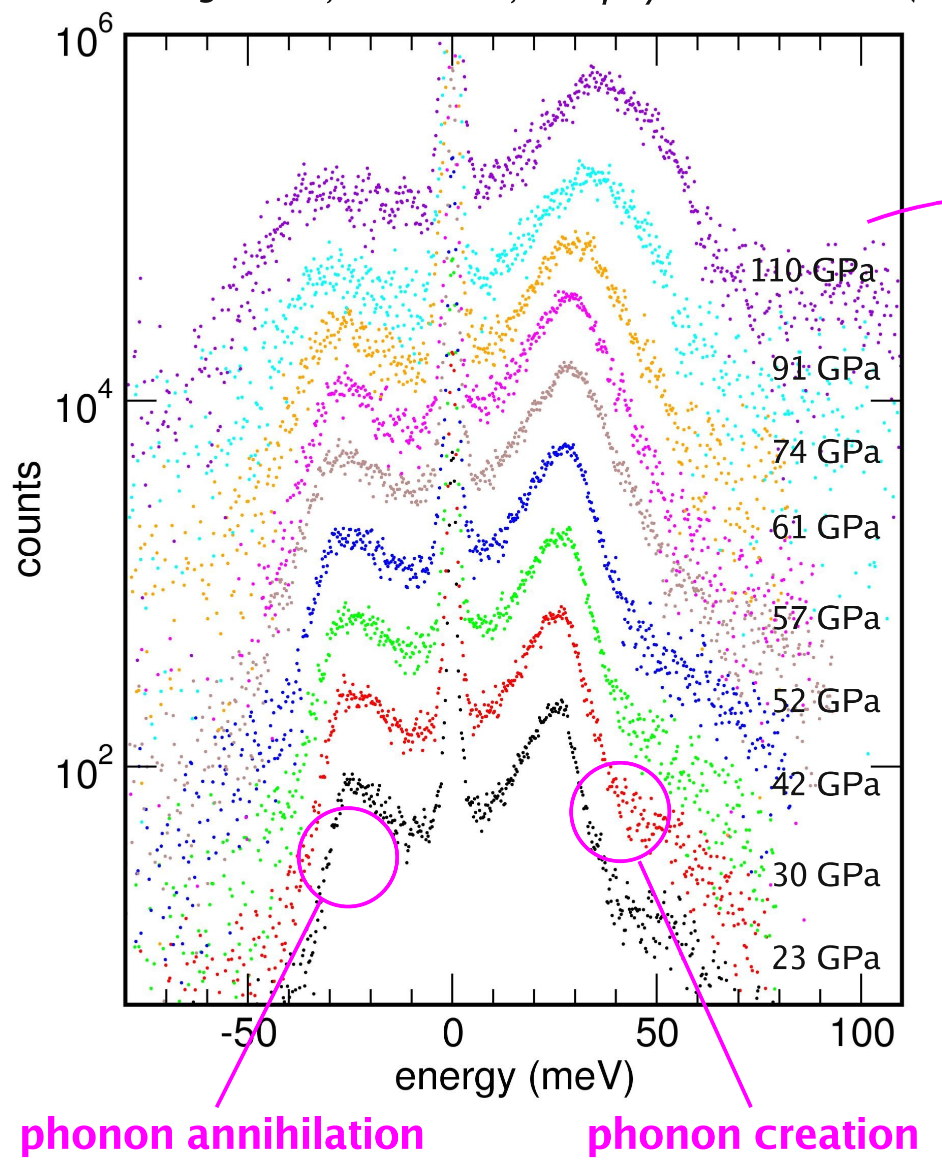


- NaCl structure; cubic unit cell
- MgO and FeO form a solid solution
- Mg and Fe atoms are surrounded by six oxygen atoms forming a slightly distorted octahedron
- upon compression the localized (non-binding) 3d electrons of Fe can change configuration



NRIXS on $(Mg_{0.75}Fe_{0.25})O$:

J.-F. Lin, S.D. Jacobson, W. Sturhahn, J.M. Jackson, J. Zhao, C.-S. Yoo, Geophys.Res.Lett. 33 (2006)

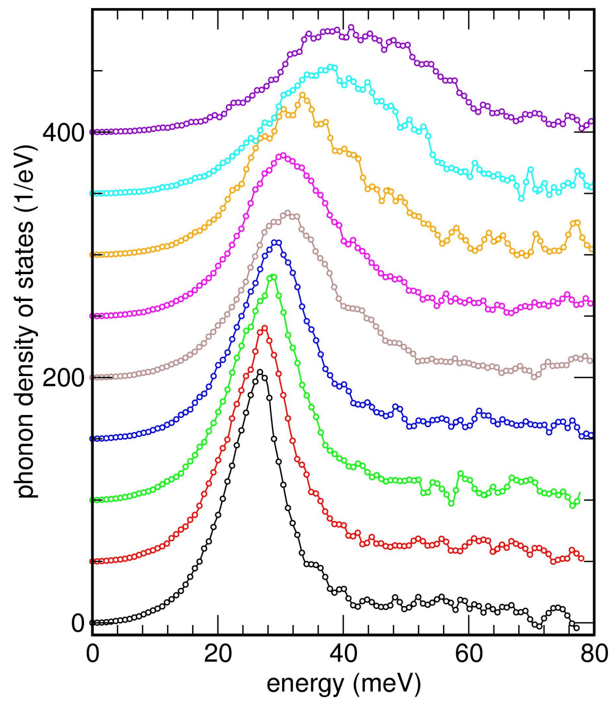


☆ **the partial phonon DOS is extracted from the spectrum**

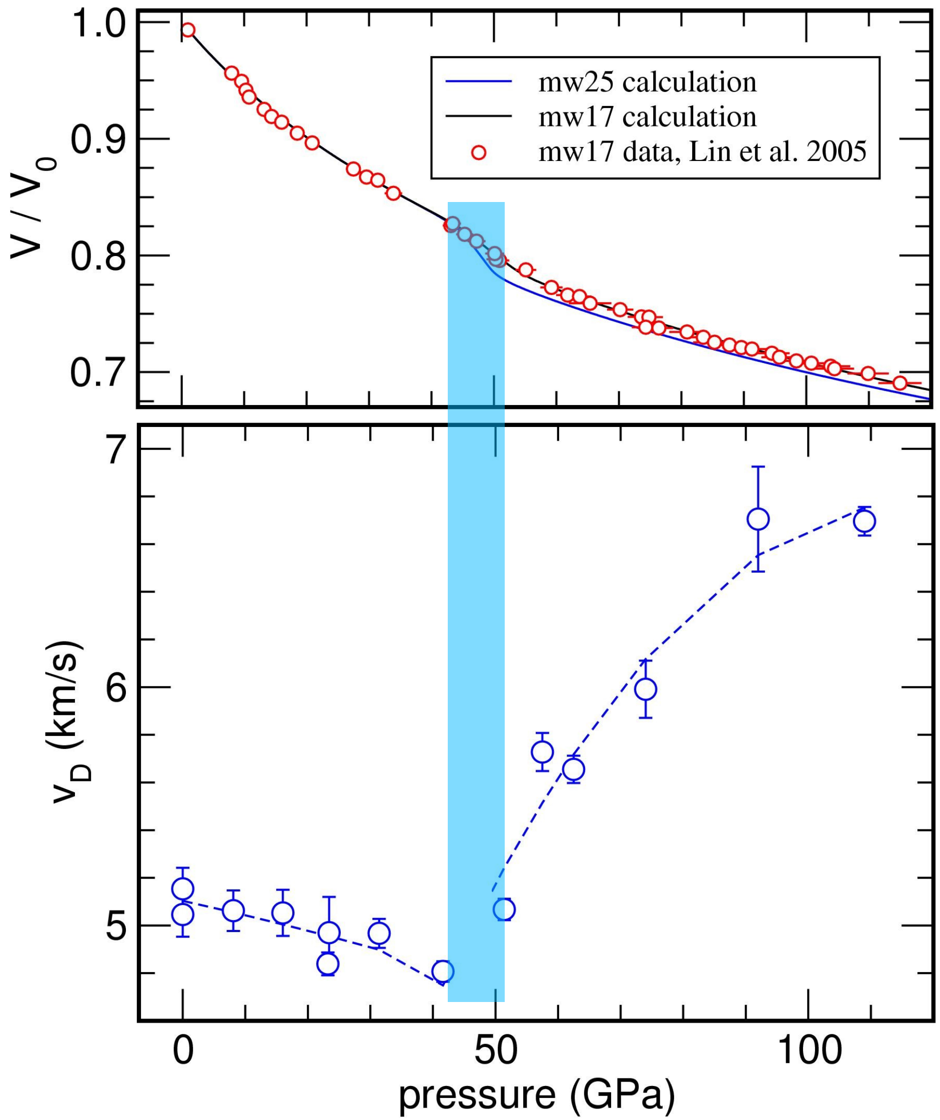
V.G.Kohn et al., Phys.Rev. B 58 (1998)

M.Hu et al., Nucl.Instrum.Methods A 428 (1999)

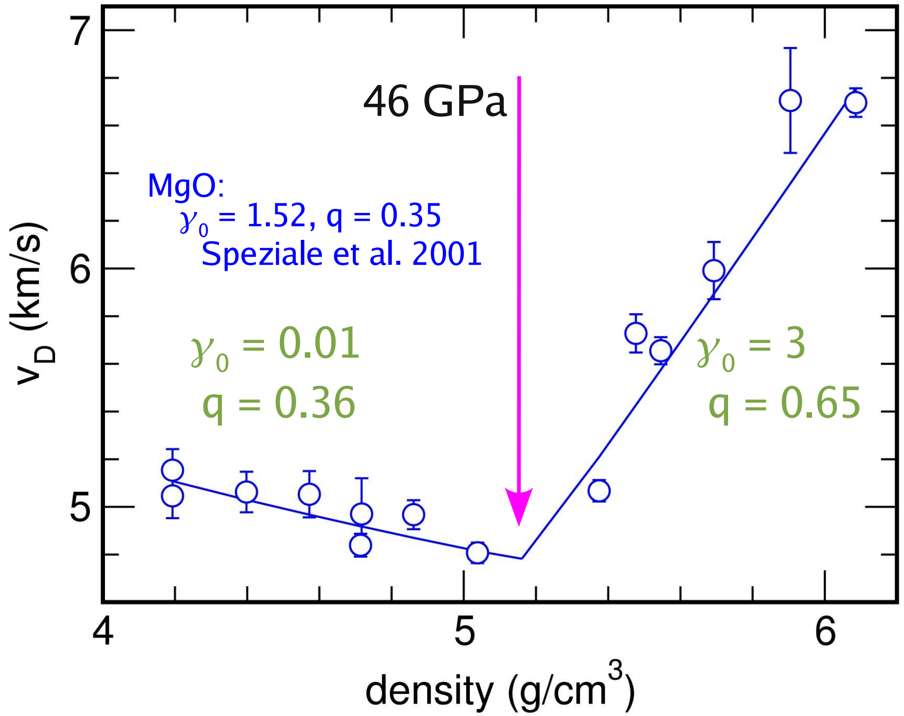
W.Sturhahn, Hyperfine Interact. 125 (2000)



Debye sound velocities of $(Mg_{0.75}Fe_{0.25})O$:

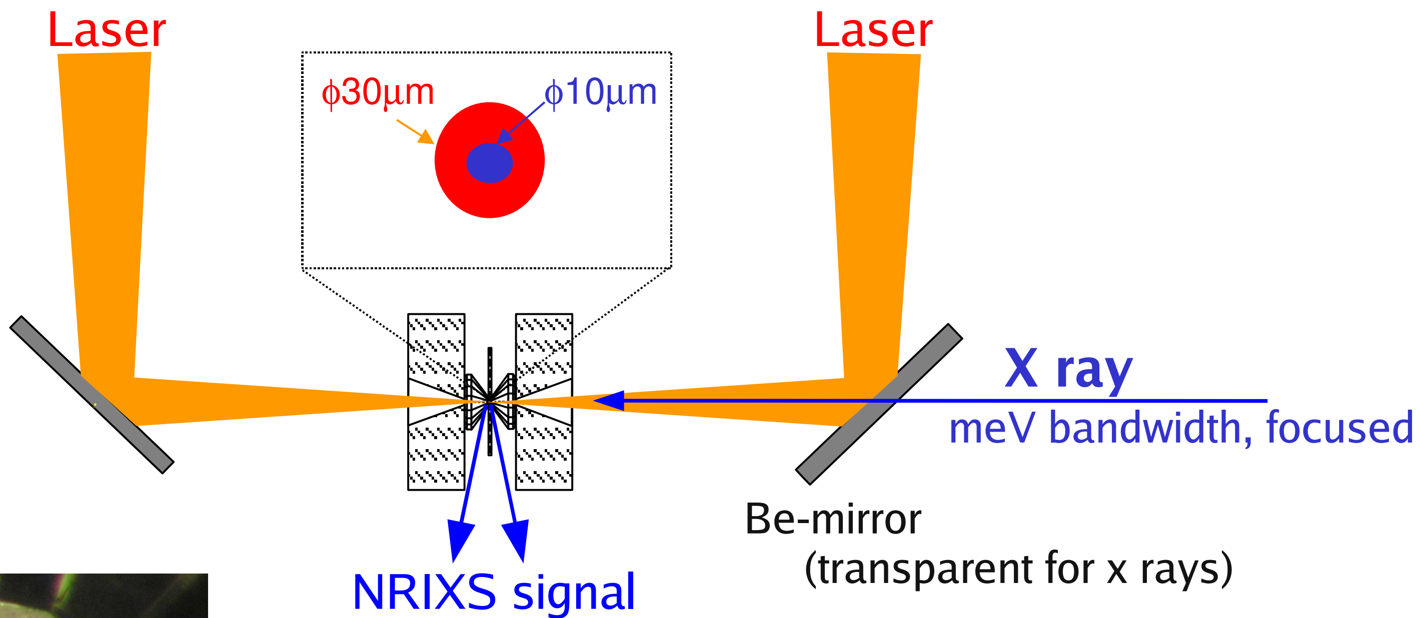


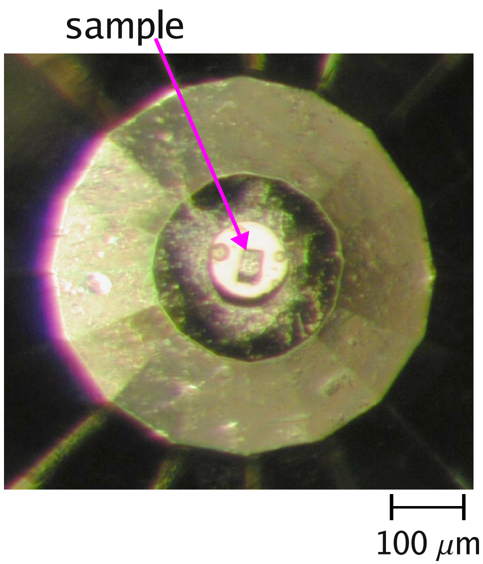
$$v_D = v_{D0}\left(\frac{\rho_0}{\rho}\right)^{1/3}\exp\left[\frac{\gamma_0}{q}\left(1-\left(\frac{\rho_0}{\rho}\right)^{q}\right)\right]$$



☆ we observe a pronounced anomaly of the sound velocity near the spin crossover

NRIXS in the DAC with Laser heating:



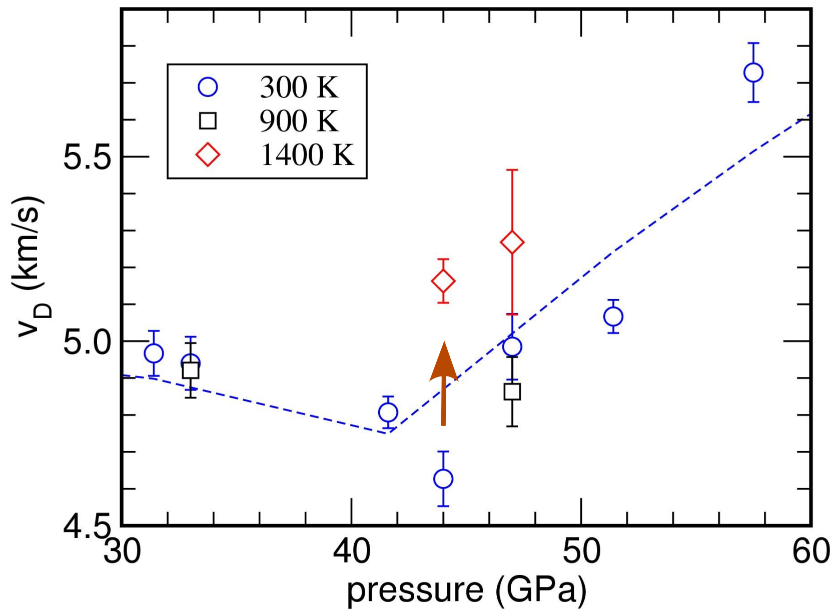


➢ **challenges**

- ☆ stability during data collection time (6-12h)
- ☆ chemical reactions
- ☆ quality of thermal insulator surrounding the sample

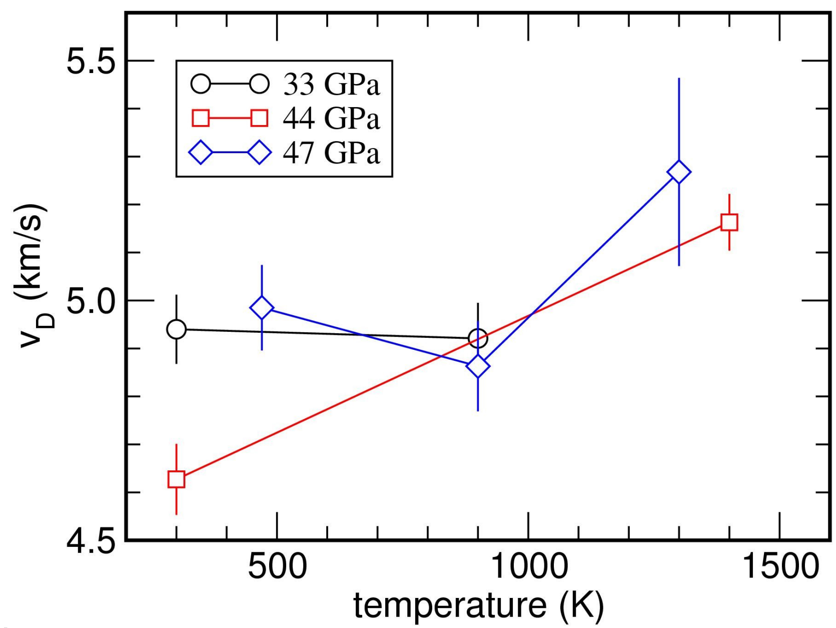
J.Zhao et al., High Press. Res. 24 (2004)

First evidence for anomalous temperature effects:



- NRIXS data from $(Mg_{0.75}Fe_{0.25})O$ in the Laser-heated diamond anvil cell provide temperature dependent Debye sound velocities.

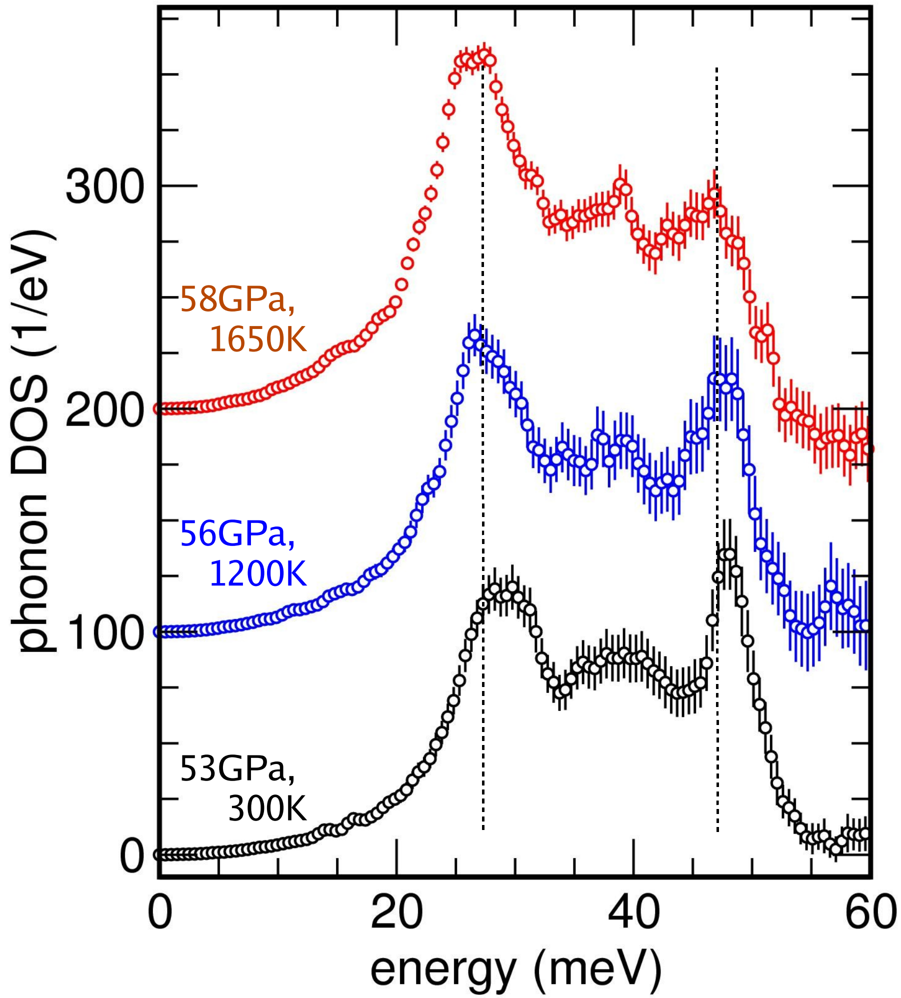
- the data indicate a positive temperature derivative of the Debye sound velocity in the spin crossover region.



in collaboration with:

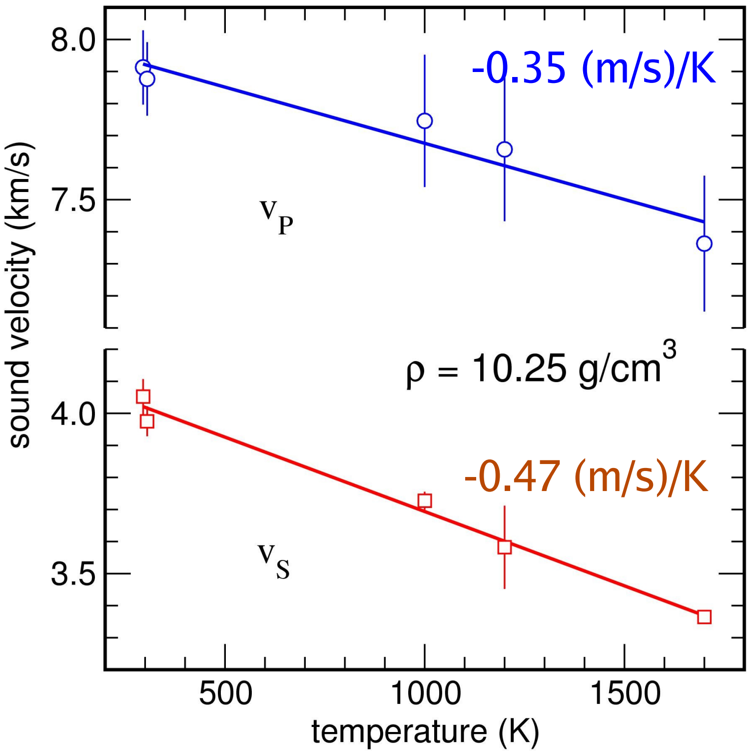
Jung-Fu Lin, Lawrence Livermore National Laboratory

Phonon DOS of hot, dense iron:

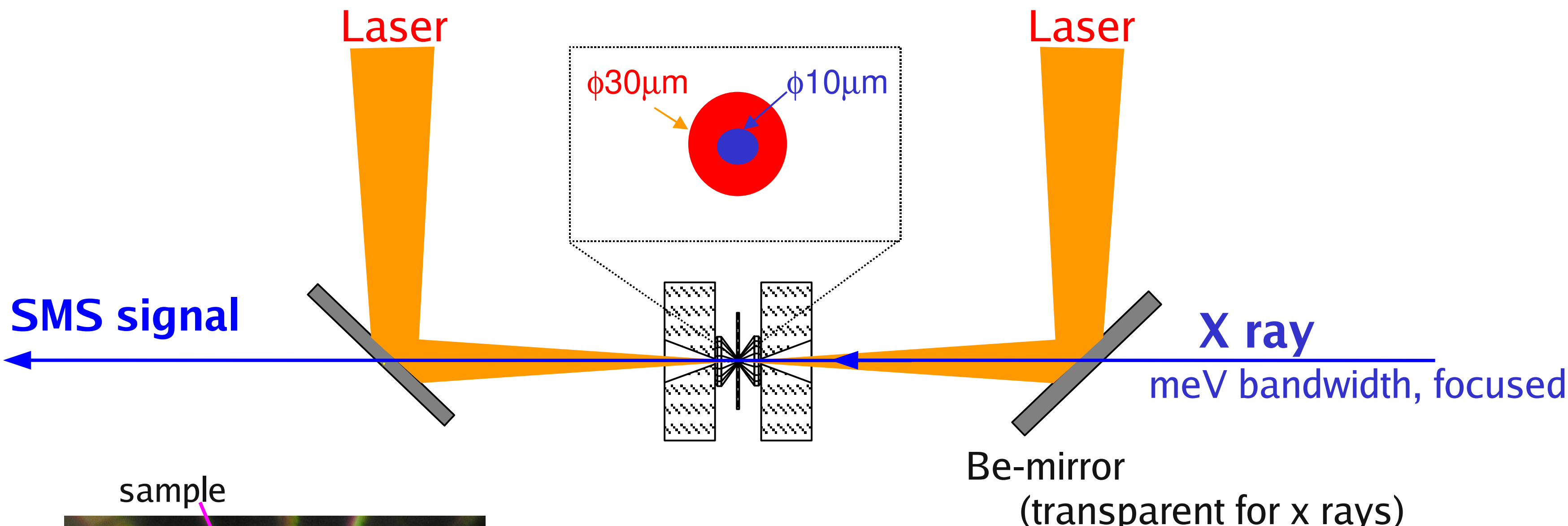


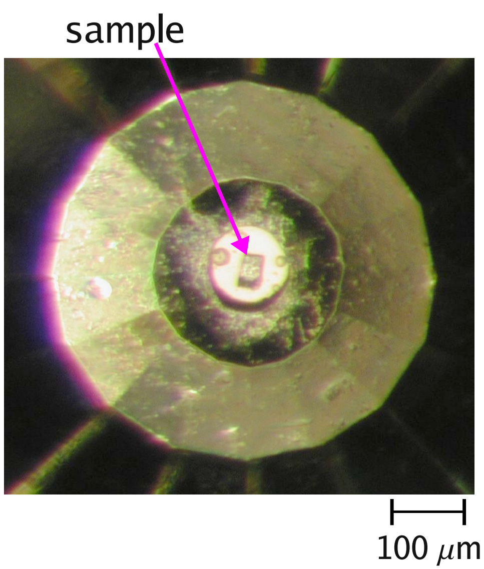
J.-F. Lin, W. Sturhahn, J. Zhao, G. Shen, H.-K. Mao, R.J. Hemley, Science 308 (2005)

- ✰ the phonon density of states was derived for compressed and Laser-heated hcp-iron
- ✰ a softening of the lower-energy phonon modes is observed



SMS in the DAC with Laser heating:

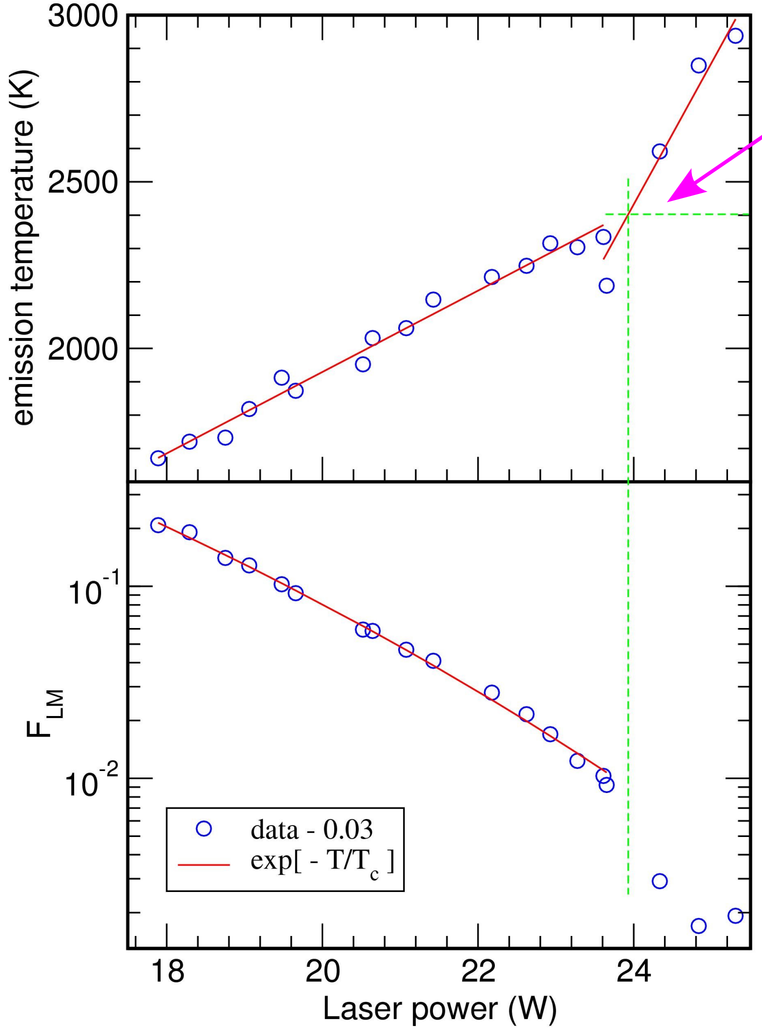




- **challenges**
  - ☆ stability during data collection time (15-30min)
  - ☆ chemical reactions
  - ☆ quality of thermal insulator surrounding the sample

J.M. Jackson et al. (paper in preparation)

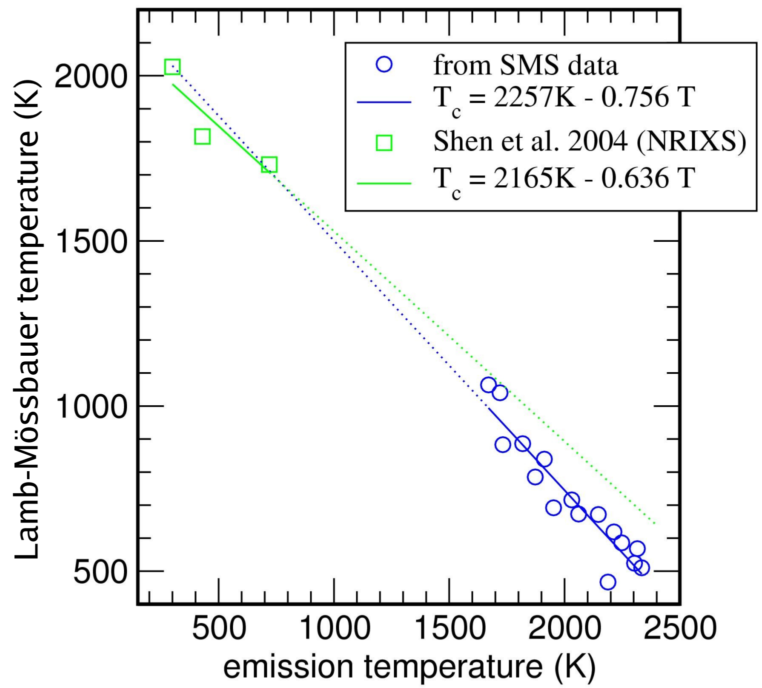
Melting under high pressure:



2400K ⇨ melting of fcc-iron metal (32GPa)

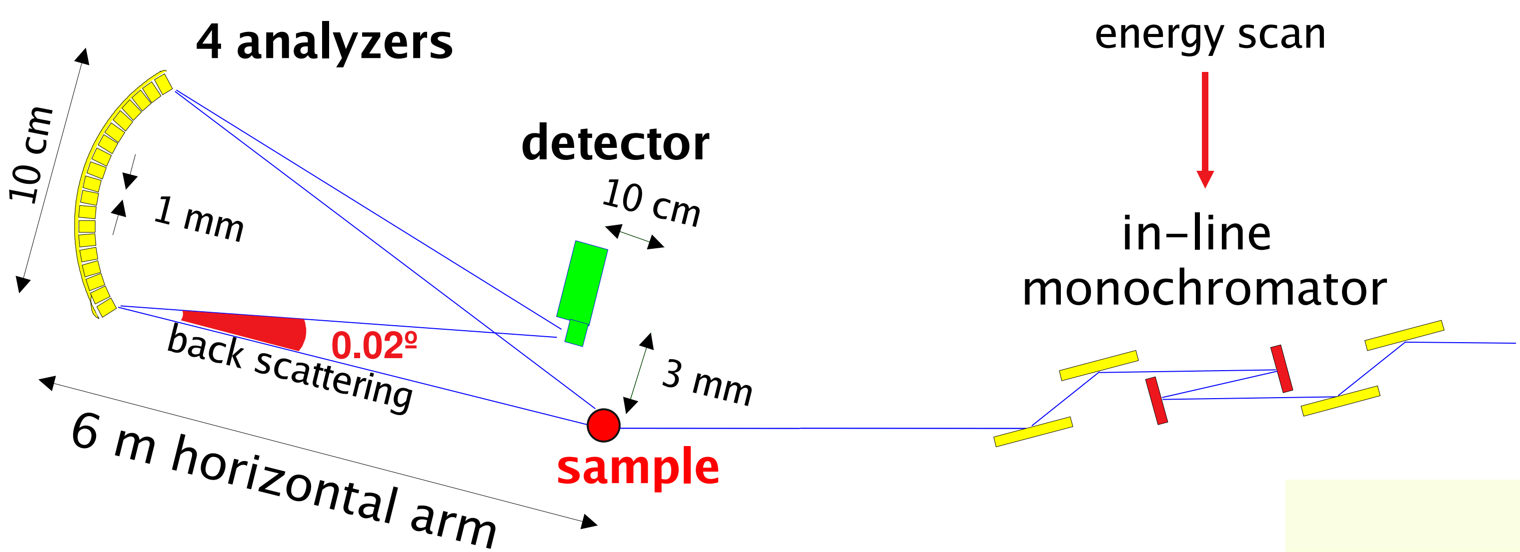
$$F_{LM}(T \to \infty) = \exp\left[-\frac{T}{T_{LM}}\right]$$

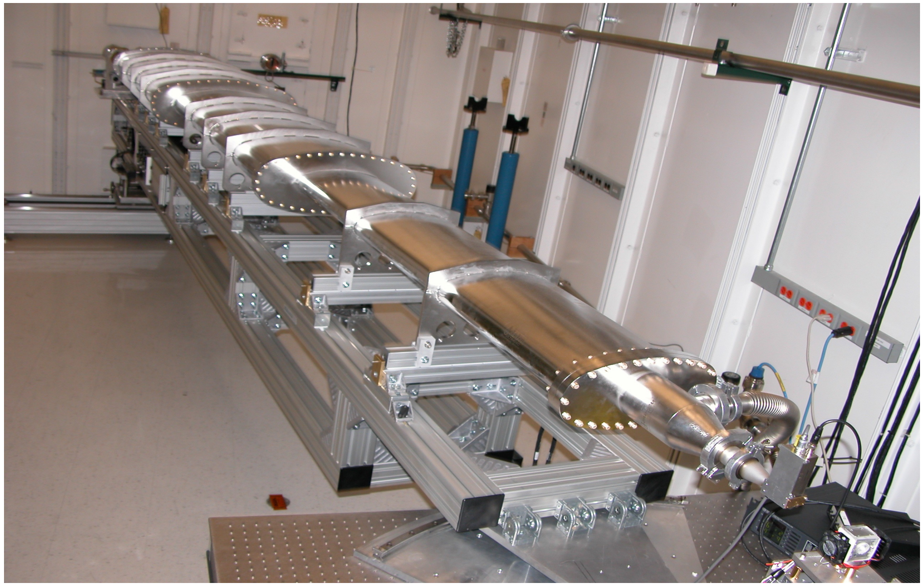
$$\frac{1}{T_{LM}} = \int \frac{E_R}{E^2}\mathcal{D}(E)dE$$



J.M. Jackson et al. (California Institute of Technology, paper in preparation)
in collaboration with J.D. Bass and S.V. Sinogeikin,
University of Illinois at Urbana-Champaign

HERIX spectrometer at sector 3-ID:

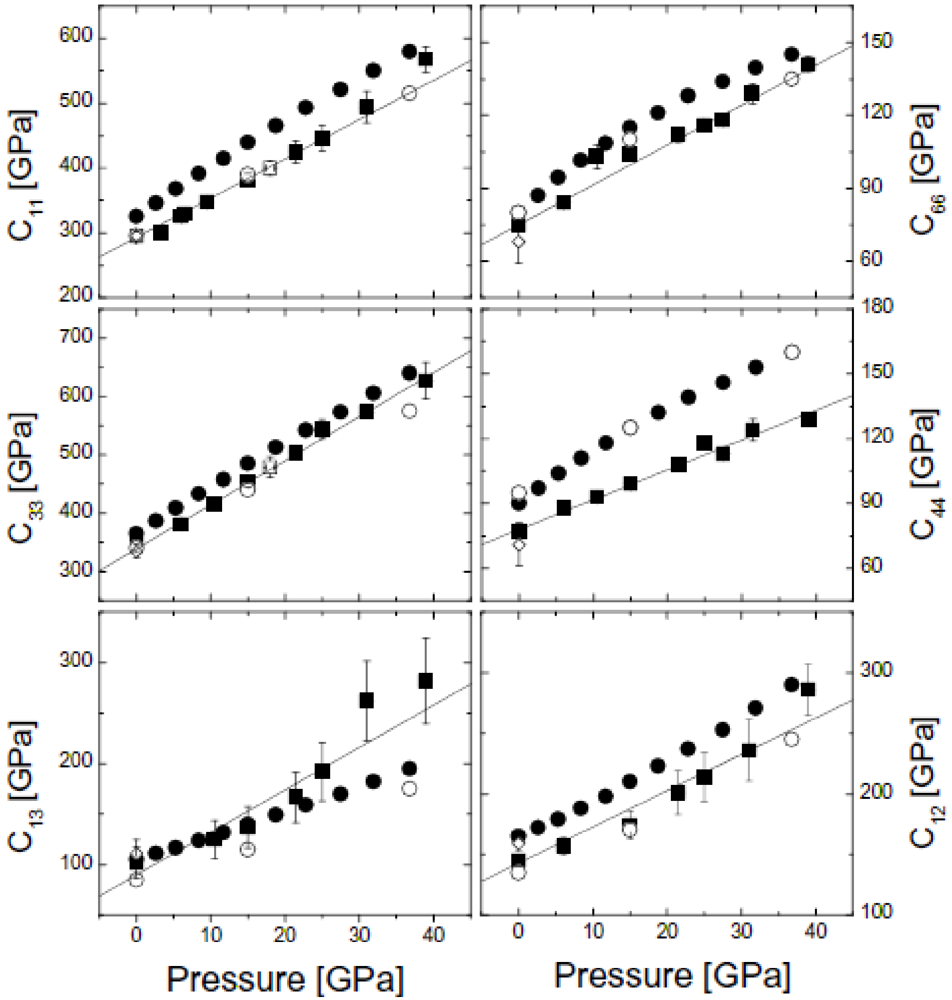




Energy (keV)	21.657
Reflection	18 6 0
ΔE_{total} (meV)	2.2
Q-range (nm^{-1})	32
ΔQ (nm^{-1})	1.8 (0.7 used)
# of analyzers	4
Flux (ph/s)	4×10^9
Beam size (μm^2)	150 x 200

Elastic tensor from IXS:

(data taken at ESRF ID28)



- hcp-Co single crystal
- squares: from IXS data
- circles: ab-initio

- elastic anisotropy of materials under high pressure

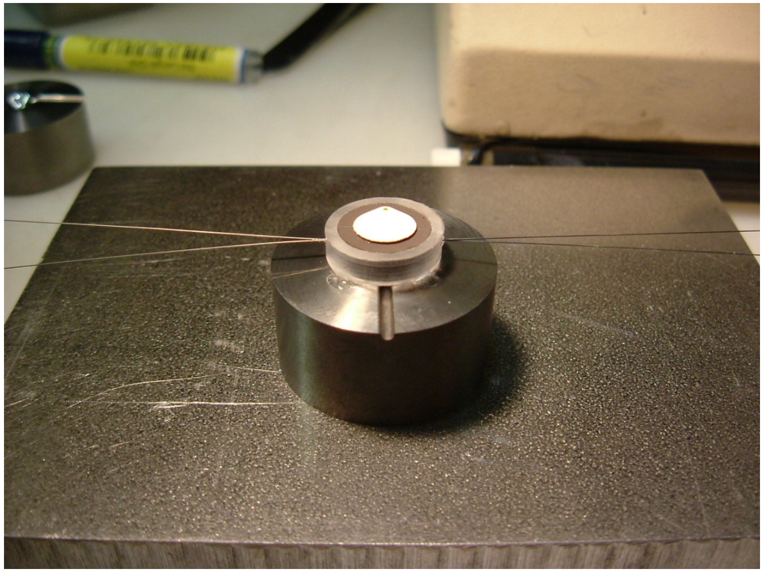
D. Antonangeli, M. Krisch, G. Fiquet, D.L. Farber, C.M. Aracne, J. Badro, F. Occelli, H. Requardt, Phys. Rev. Lett. ***93*** *(2004)*

Diamond anvil cells or Large-volume devices?

(wide angle diamond anvil cell)

- ✫ lightweight device
- ✫ heating & cooling possible
- ✫ extended pressure range (several 100 GPa)
- ✫ very small sample volume
- ✫ low x-ray absorption

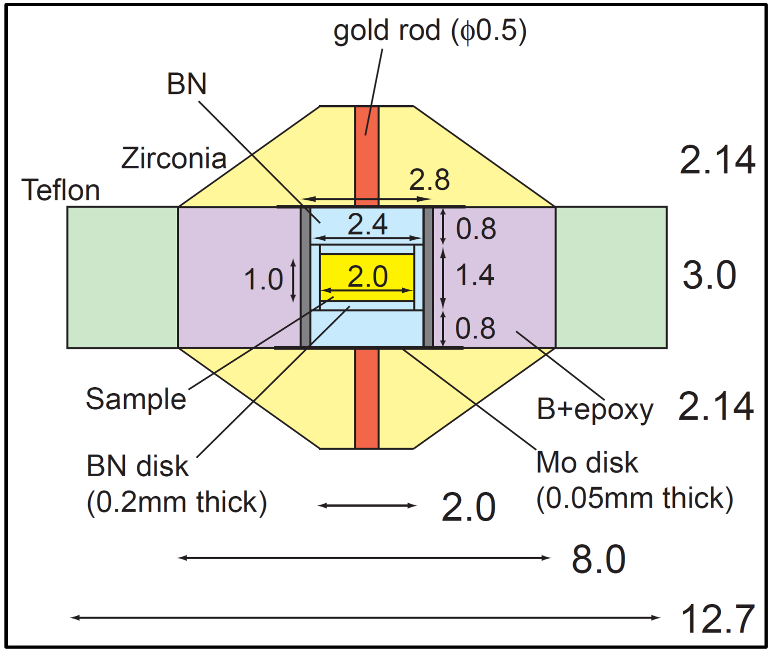
- ✫ large sample volume
- ✫ stable heating conditions
- ✫ limited pressure range (10-15 GPa)
- ✫ high x-ray absorption



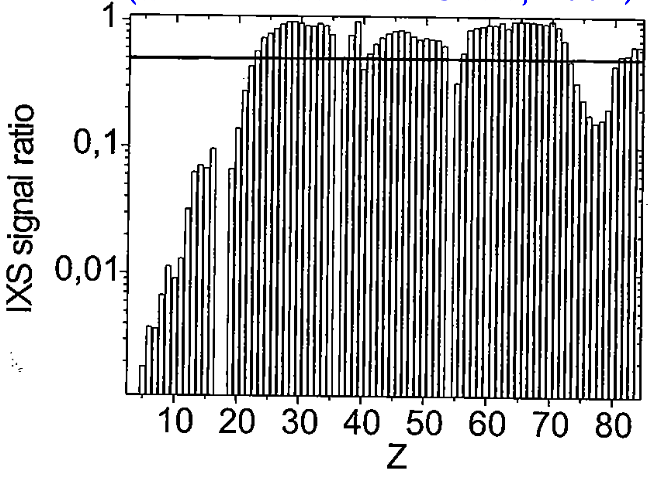
(gasket assembly for Paris-Edinburgh cell)

Paris-Edinburgh cell:

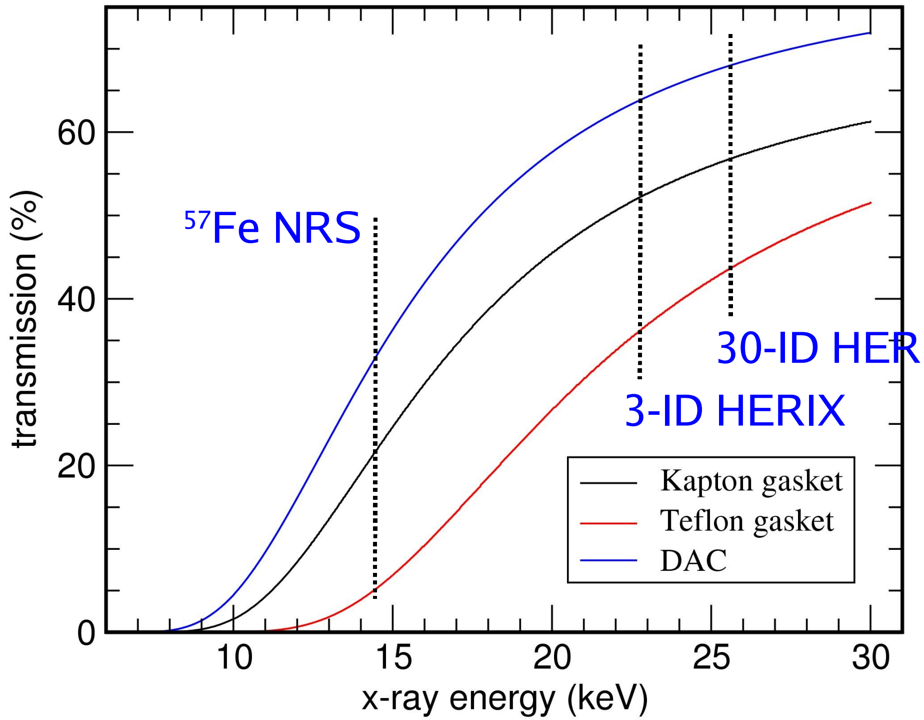
(gasket assembly for Paris-Edinburgh cell)



(after: Krisch and Sette, 2007)



20μm sample
17.8 keV energy



- ✰ 2 mm sample size usable in HERIX instrument geometry
- ✰ ten-fold intensity gain for light element samples

Where to go in the next 5 to 10 years?

- **technically**
  - ✰ improve counting rates by a factor of ten
  - ✰ new IXS capabilities, e.g., beam line 30-ID at the APS
  - ✰ combine IXS with various high-pressure devices
- **methodologically**
  - ✰ synergies of experimental techniques, e.g., integration of NRS and diffraction
  - ✰ IXS and NRS under extreme conditions, e.g., high pressure
  - ✰ develop alternate NRS avenues, e.g., reconstructive SMS
- **scientifically**
  - ✰ explore cross-disciplinary applications of NRS and IXS
  - ✰ Earth materials under high pressure and temperature
  - ✰ local vibrational dynamics of enzymes and proteins
  - ✰ magnetism and vibrations of artificial nanostructures
  - ✰ pump-probe experiments
  - ✰ ....