NRIXS and SMS with laser-heated diamond anvil cells

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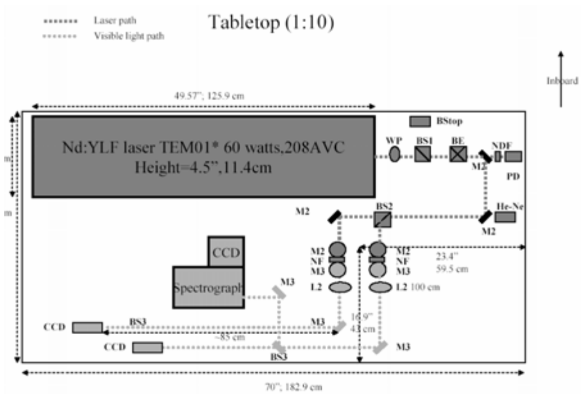
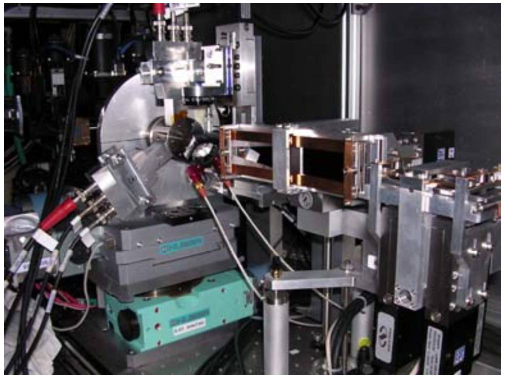


Just started, June 4th, 2002



System setup, December 2002

Collaborators :

Argonne National Lab: Wolfgang Sturhahn, Jiyong Zhao, Ercan Alp
Geophysical Lab: Ho-kwang Mao, Russell Hemley
The University of Chicago: Guoyin Shen

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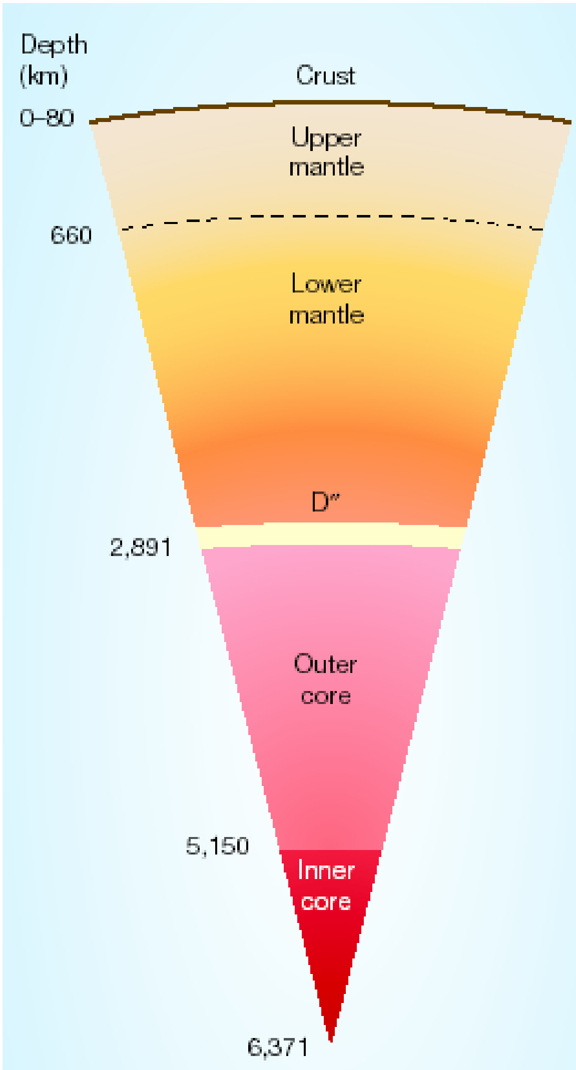
Outline

- **Motivation: study of the Earth's core and mantle by NRIXS and SMS**
- **NRIXS study of hcp-Fe in a LHDAC**
- **Absolute temperature determination: detailed balance principle**
- **Sound velocities of hcp-Fe at high PT: temperature effect**
- **SMS study of Fe_2O_3 in a LHDAC**
- **Conclusions and future challenges**

Iron in the Earth's mantle and core



Duffy, Nature, 2004

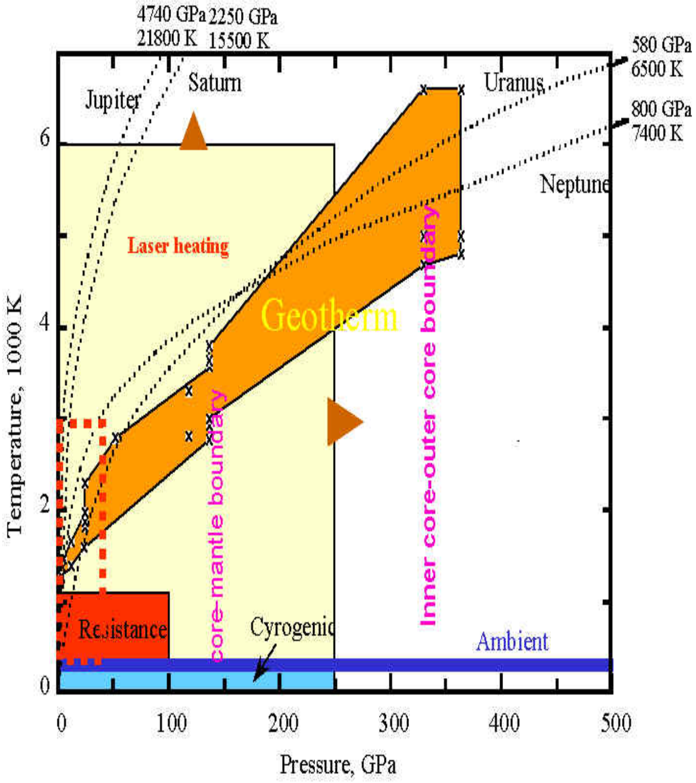
Major component	By who, when, and what
Magnesiowustite [(Mg,Fe)O] and silicate perovskite [(Mg,Fe)SiO3]	Liu et al, 1975 X-ray diffraction after heating
Post silicate perovskite [(Mg,Fe)SiO3] and [(Mg,Fe)O]	Murakami et al, 2004 *In situ* X-ray in a LHDAC
Liquid Fe-Ni alloy and light elements	Birch, 1952 Shock wave
Solid Fe-Ni alloy and a small amount of light elements	

Iron, a transition metal, exists in all major components of the Earth's interior. The unique physical properties of iron can be unveiled by NRIXS, SMS, XES, and other techniques and help us understand the Earth's interior.

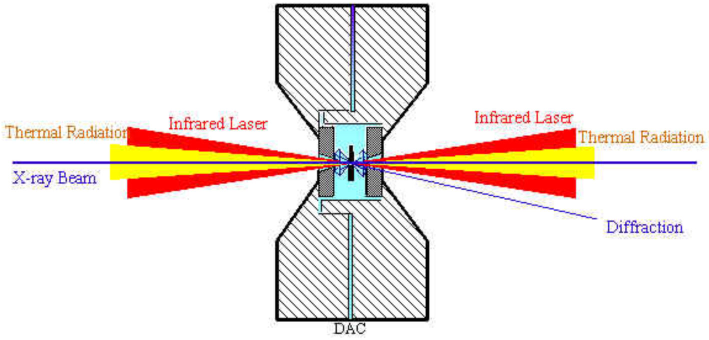
Laser-heated diamond anvil cell

A new window to study planetary interiors

Pressure-temperature range



Laser-heated Diamond cell



Pressure (P), temperature (T), and composition (X) are three most important thermodynamic parameters. The pressure and temperature range of a LHDAC is best suited for studying mineral physics of planetary interiors.

Mineral physics in a laser-heated diamond cell

Earth's mantle and core materials have been extensively studied for their crystal structures. However, other physical properties remain not well understood.

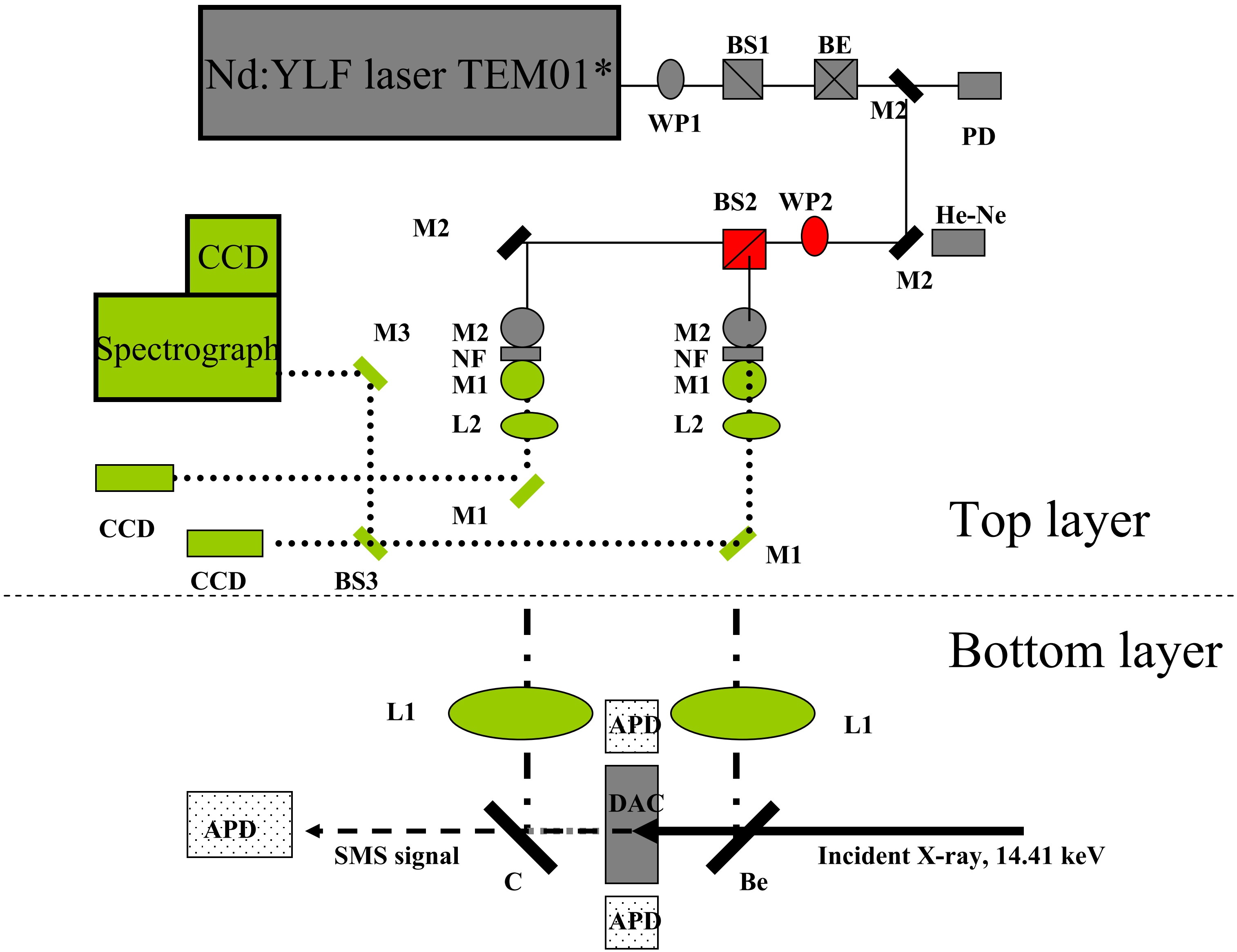
- **Quenched experiments**
- **X-ray diffraction**
- **NRIXS and SMS**
- **Raman and IR**
- **X-ray emission spectroscopy**
- **Single-crystal zone diffraction**
- **Inelastic X-ray Scattering**
- **Brillouin spectroscopy**
- **Neutron diffraction**
- **Other techniques….**

- **Chemical compositions, structure**
- **Crystal structure, P-V-T EOS**
- **DOS, V_D, Vp, Vs, G, T,…**
- **Optical modes**
- **Spin states of transition metals (Fe)**
- **Structure refinement, phonon dispersion**
- **Phonon dispersion (Vp)**
- **Acoustic modes (Vs, Vp)**
- **Structure of low-Z elements…**
- **Other properties…….**

A double-sided laser-heating system has been built to study NRIXS and SMS of Fe-containing materials under high pressures and temperatures.

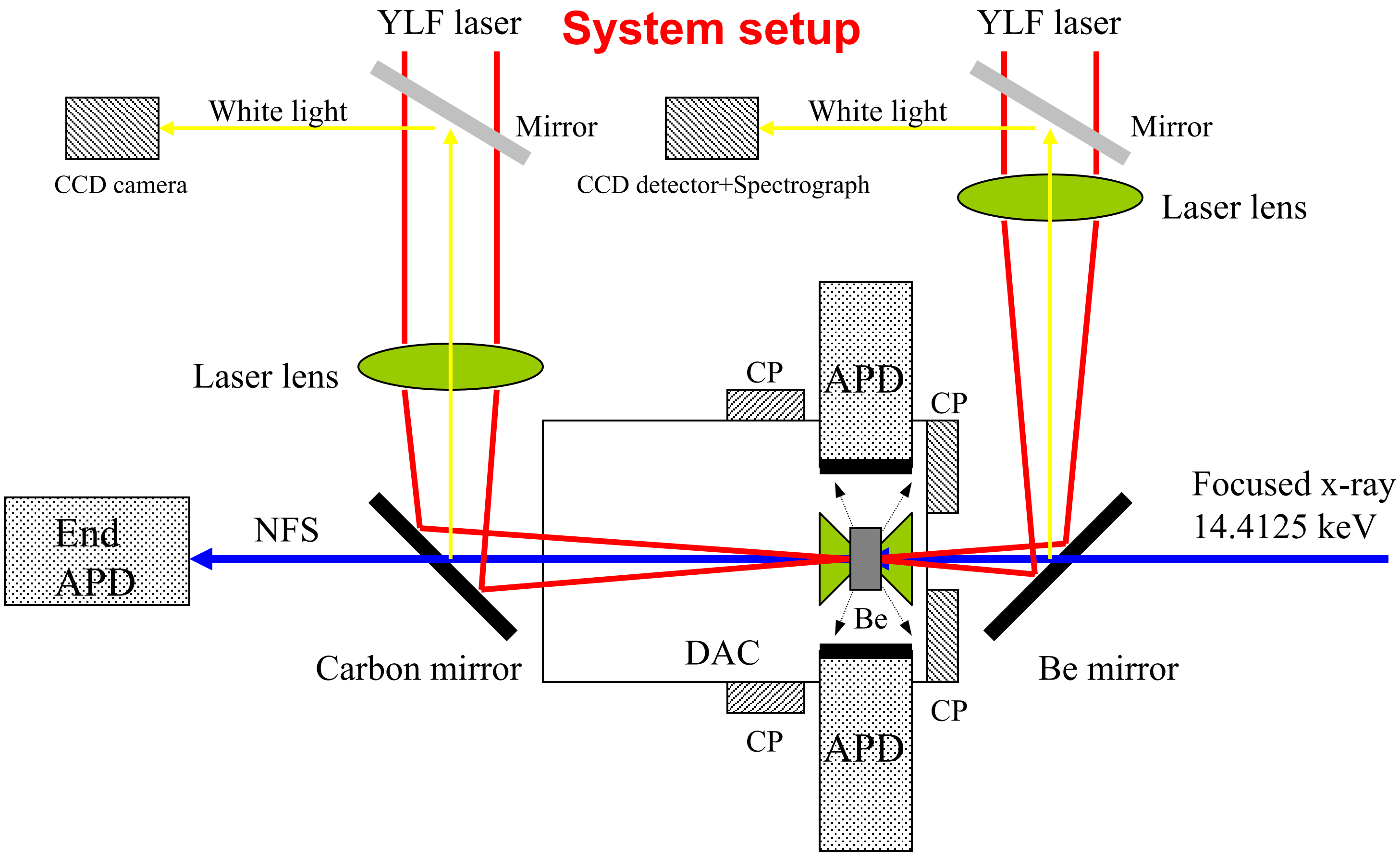
Lin et al., GRL, 2004; Lin et al., RSI, 2004

NRIXS and SMS with a laser-heated diamond cell

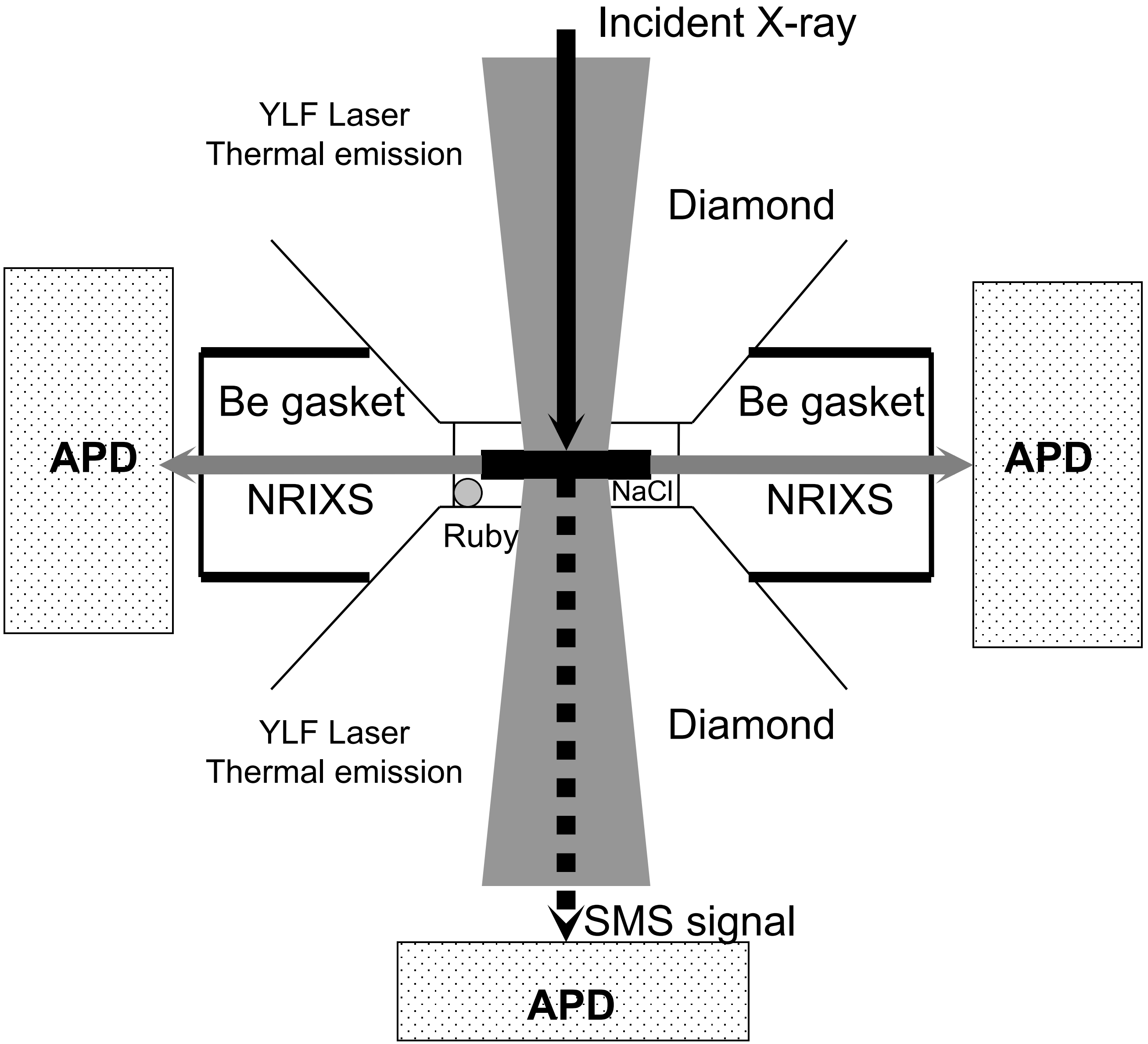


Lin et al., 2005 (in press)

Nuclear resonant in a laser-heated diamond cell

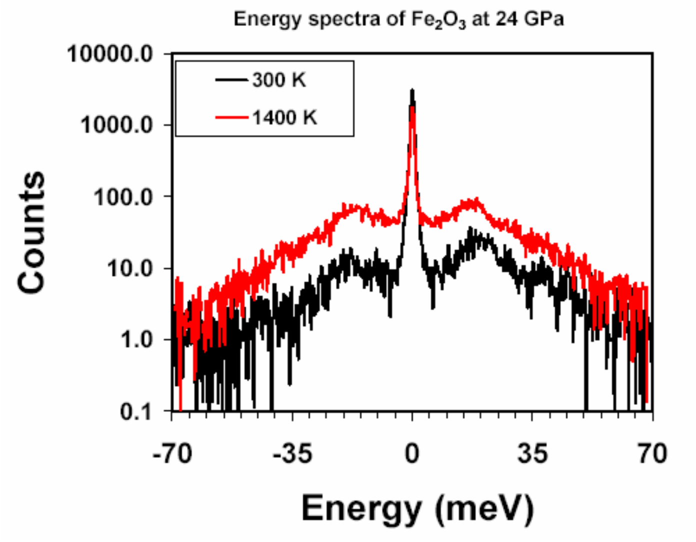


X-ray, laser, and sample



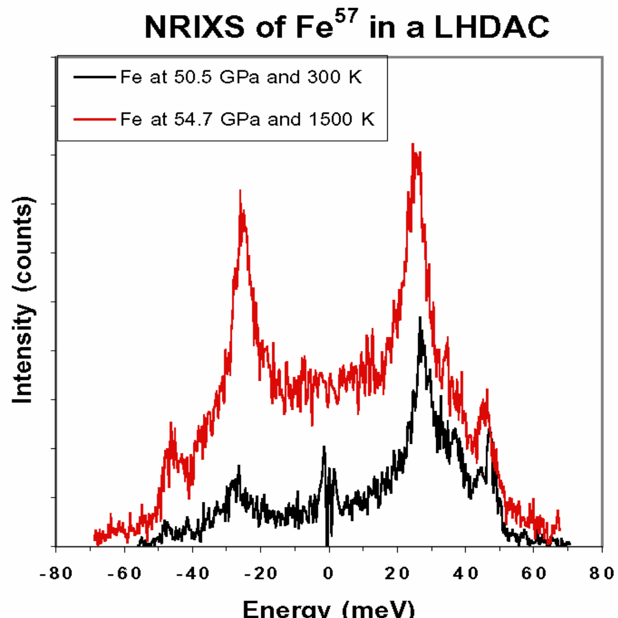
Some Technical Problems:

1. Mechanical stability of the LHDAC
Problems: sample movement and pressure change during heating
Solution: cooling plates to remove heat away from the DAC
2. Be gasket has lower shear strength than Re, B, BN gaskets
Problems: thinner sample/lower signals, deformation of the sample chamber
Solution: B, BN, or diamond gaskets (Lin et al., RSI, 2003)
3. Thickness of the sample decreases during long term heating
Problems: counts in phonon creation and annihilation vary with time
Solution: X-ray absorption monitor to correct for the intensity/thickness factor



Temperature determination

Energy spectra of hcp-Fe



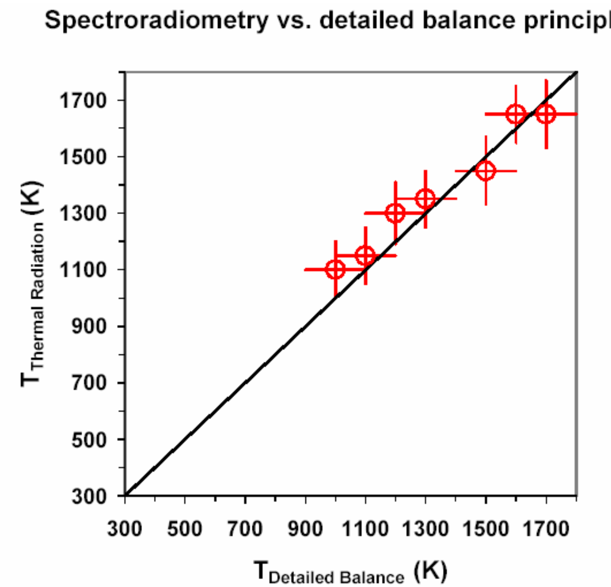
The asymmetry is independent of sample properties other than temperature and is given by the Boltzmann factor:

$I(E)/I(-E) = exp(E/k_BT)$

Temperature value is then given by:

$T = E/(k_B ln(I(E)/I(-E))$

Temperature in a LHDAC



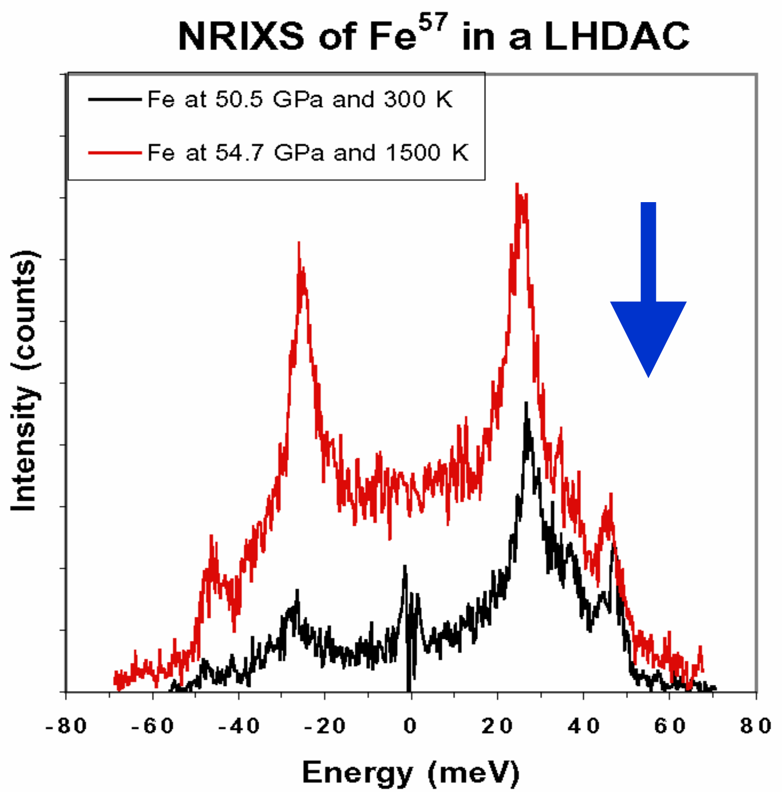
These temperatures are in very good agreement with values determined from the thermal radiation spectra fitted to the Planck radiation function up to 1700 K.

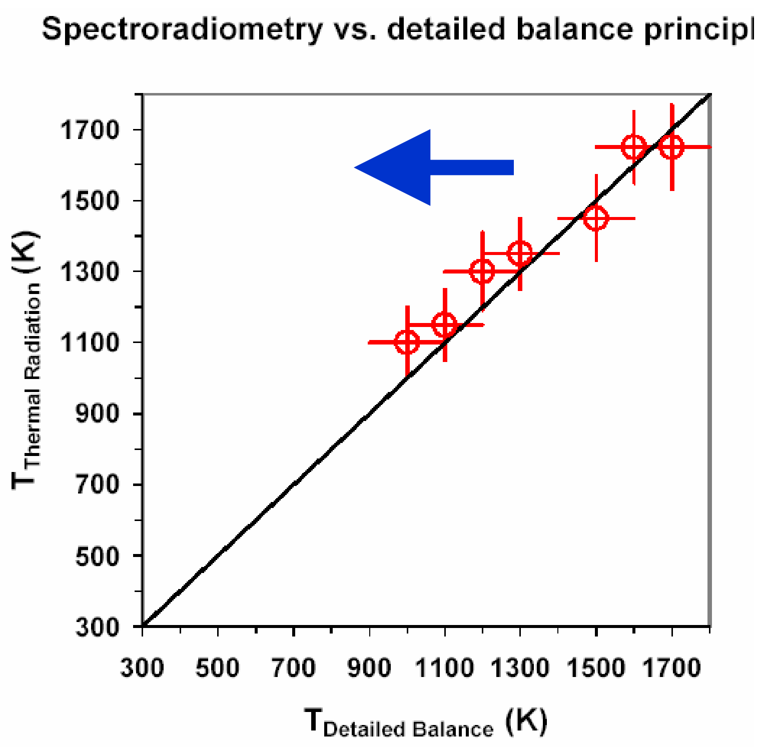
This is the first independent confirmation of the validity of temperatures determined from the Planck radiation law in the LHDAC experiments.

Lin et al., GRL, 2004, Shen et al., PCM, 2004

Absolute temperature determination?

Thinner sample during heating **Misalignment**





A well prepared sample with a stable heating is essential to the success of the LHDAC experiments.

Reliability of the derivation of V_D, Vp, Vs, G

$$\frac{K_S}{\rho} = V_P^2 - \frac{4}{3} V_S^2$$

$$\frac{G}{\rho} = V_S^2$$

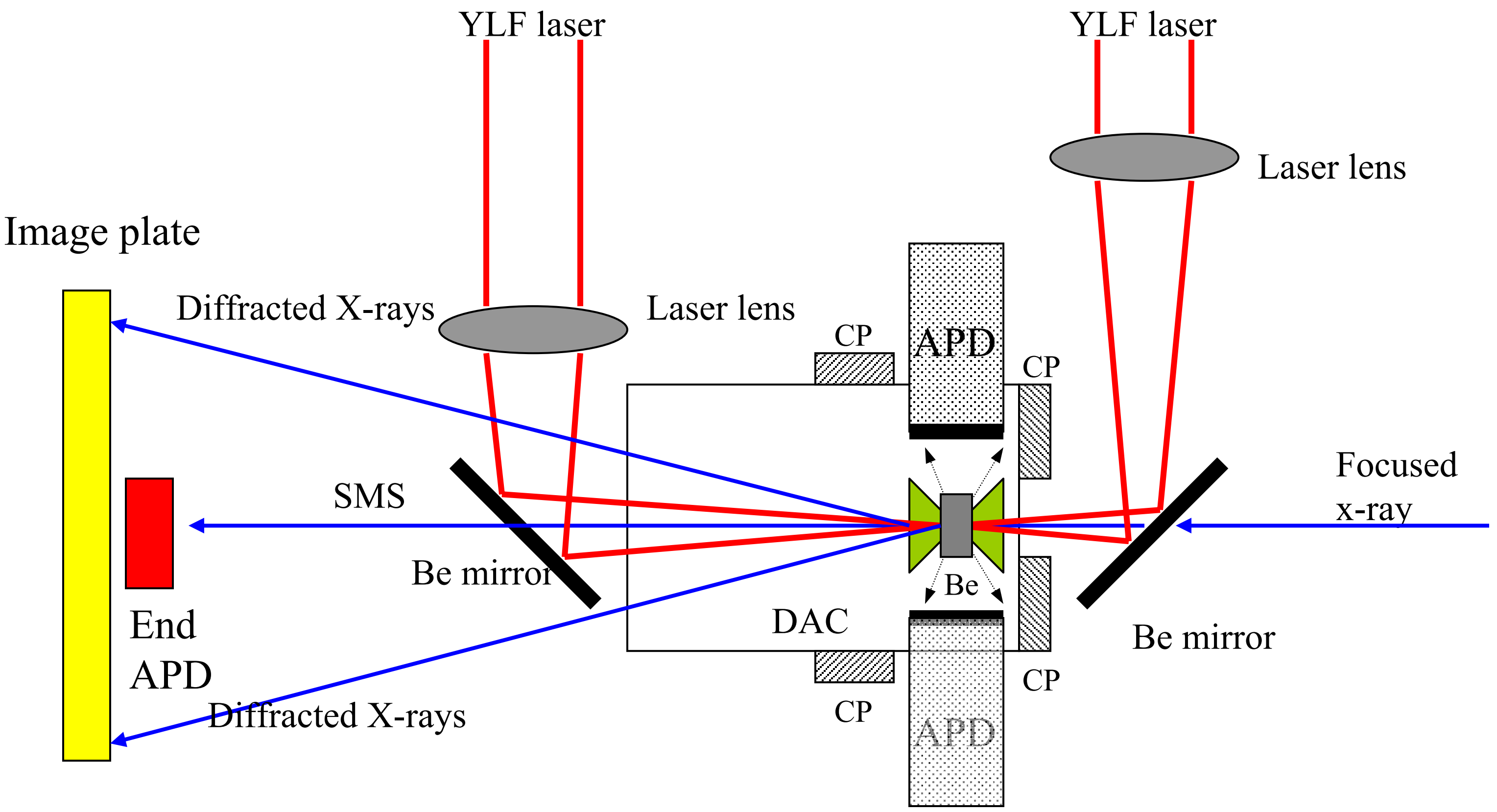
$$\frac{3}{V_D^3} = \frac{1}{V_P^3} + \frac{2}{V_S^3}$$

The derivation of the V_S is relatively insensitive to the differences in the EOS data (Ks and ρ). Therefore, the NRIXS technique is particular suited at constraining V_S with a precise measurement of V_D.
Ks and ρ are calculated from thermal EOS provided by previous X-ray diffraction study, causing uncertainties in deriving Vp, Vs, and G.

Estimation of thermal pressure in a LHDAC:
Constant pressure condition: no thermal pressure
Constant volume condition: total thermal pressure (αKT)
Since NaCl insulating layers are relatively soft, the pressure increase due to heating is in between these bounds.

NRIXS-SMS and diffraction

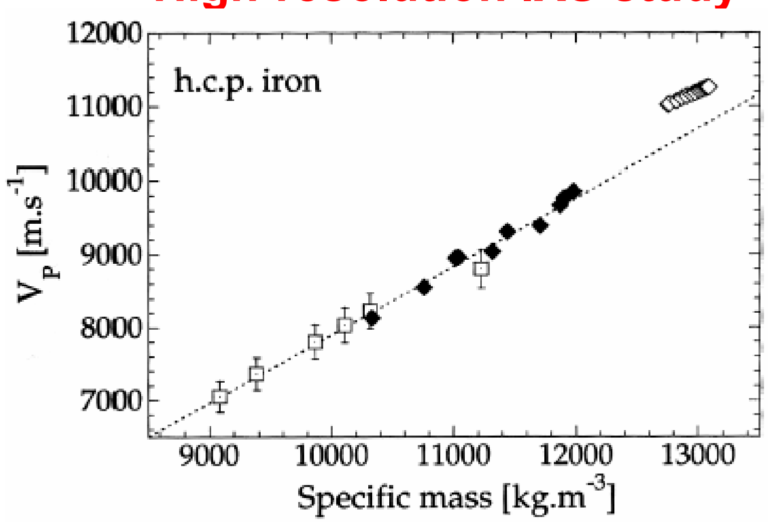
In situ **X-ray diffraction, NRIXS, and SMS studies in a LHDAC provide structural (density), magnetic, elastic, vibrational, and thermodynamic information of the sample. This is also a powerful tool to detect melting.**



Sound velocities of dense hcp-Fe

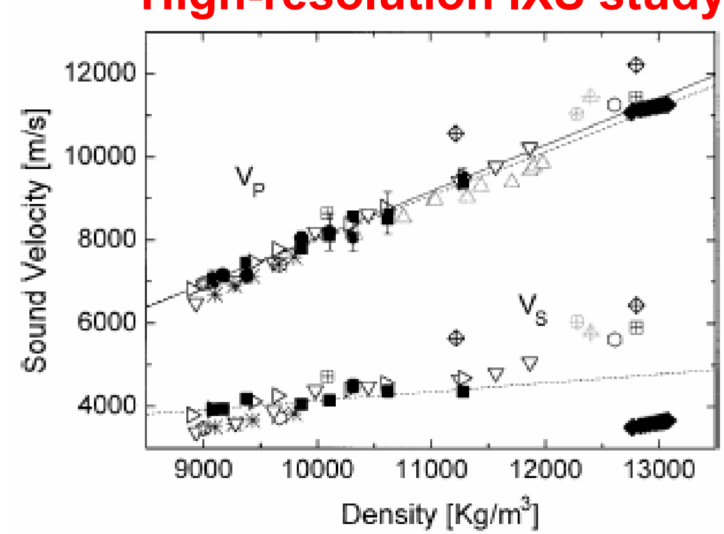
Birch's Law: sound velocity-density linear relation

High-resolution IXS study



Fiquet et al., Science, 2001

High-resolution IXS study



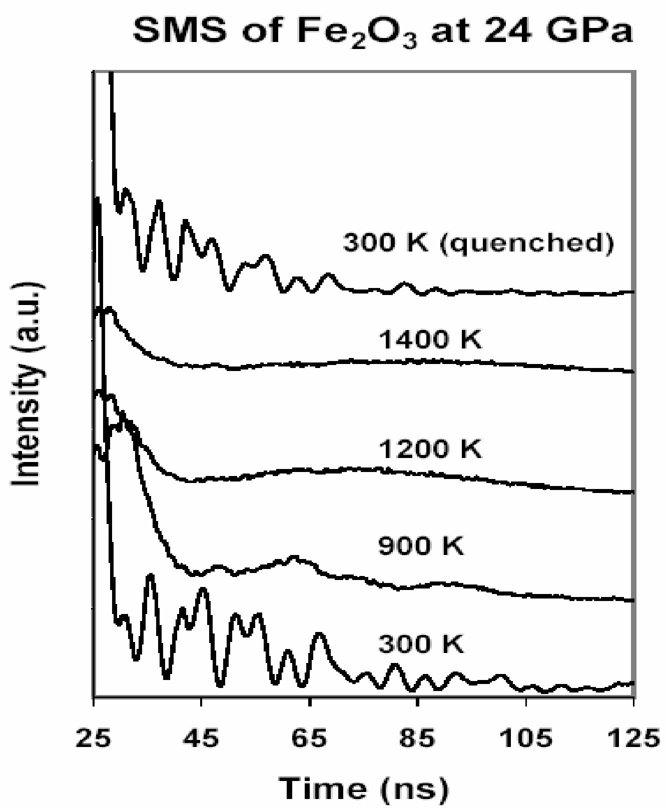
Antonangeli et al., EPSL, 2004

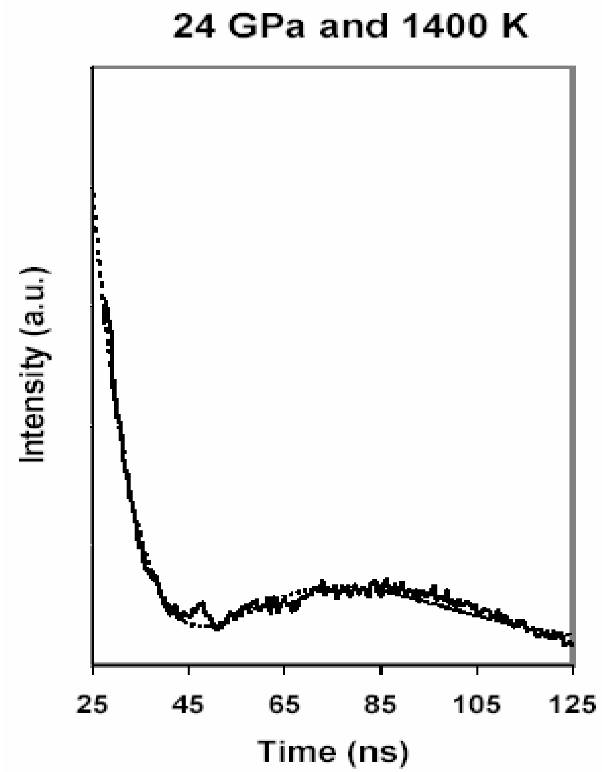
Fiquet et al. (2001) suggested an inner core that is 4-5% lighter than hcp-Fe. Recent studies by Antonangeli et al., however, show that the extrapolated Vp values are now in fair agreement with PREM model whereas the extrapolated Vs values are 30% higher than the PREM model.

Despite numerous studies on the sound velocities of hcp-Fe, inconsistencies in the extrapolated Vp and Vs remain to be resolved.

!!Temperature effect on the sound velocities need to be considered.!!

SMS study under high pressures and temperatures
Fe_2O_3 as an example



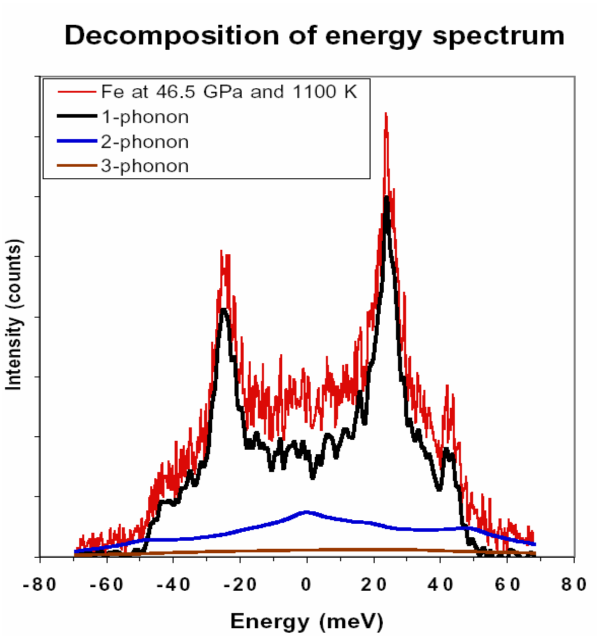


Oscillations in the time spectra are observed that originate from the nuclear-level splitting, whereas the flat feature indicates nonmagnetic state. The SMS spectra are evaluated by CONUSS programs to permit derivation of magnetic hyperfine parameters.

Lin et al., 2005 (in press)

Future experimental and theoretical challenges

Quasiharmonic vs. anharmonic model



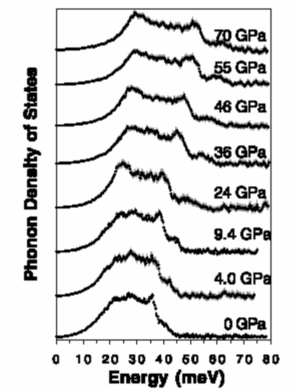
Quasiharmonic model: the atomic motions are harmonic under the given conditions of pressure, temperature, and other parameters. In this approximation, we allow for thermal effects like expansion and change of force constants with atomic distances but still assume that the vibrations occur in a harmonic potential. This model works for our data at modest temperatures. **Anharmonic model for deriving DOS of hcp-Fe at inner core P-T conditions. First-principles theoretical calculations.**

Study dilute systems, magnesiowustite and silicate perosvkite, under high PT

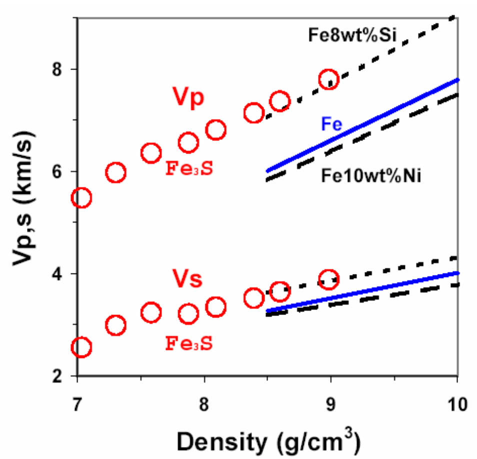
Normally, it takes ~12 hours to collect a series of reasonable energy spectra for hot dense hcp-Fe. The collecting time for mw and pv will be much longer (days) for mw and pv. An increase of a factor of 10 in incident X-ray intensity can solve many technical problems.

NRIXS and SMS of Fe compounds in a LHDAC?

300 K studies

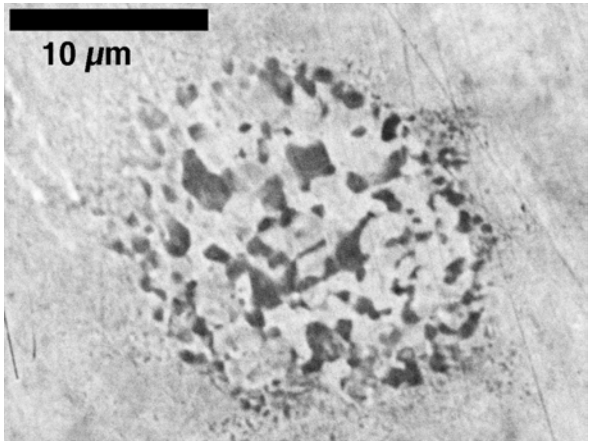


PDOS of $Fe_{0.85}Si_{0.15}$ alloy



Sound velocities of iron compounds
Lin et al., GRL, 2003; EPSL, 2004.

High-temperature studies?



BSE image of a quenched Fe-Si alloy showed two phases coexisted at ~31 GPa and 1976 K. (Lin et al., 2002)

The existence of two phases can not be distinguished in the energy spectra. The diffusion of light elements away from the laser heated spot changes the chemical composition of the starting materials.

Conclusions

1. We have built a laser heating system to study the PDOS of iron-containing materials with NRIXS and SMS in a LHDAC.
2. The detailed balance principle applied to the NRIXS spectra provides absolute temperatures of the laser-heated sample.
3. The compressional (V_P) and shear wave velocities (V_S) of hcp-Fe decrease with increasing temperature under moderate high pressures.
4. Time spectra of the synchrotron Mössbauer spectroscopy at 10 GPa and 24 GPa upon laser heating reveal that Fe_2O_3 undergoes a magnetic to nonmagnetic transition.
5. Although this study demonstrates a new arsenal of *in situ* probes to study magnetic, vibrational, elastic, and thermodynamic properties of 57Fe-containing materials, there remain technical and theoretical problems.
6. An increase of a factor of 10 in incident X-ray intensity can solve many technical problems—time, sample quality, stability, and higher P-T,