Geophysical applications of nuclear resonant scattering

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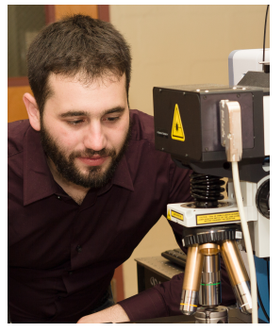
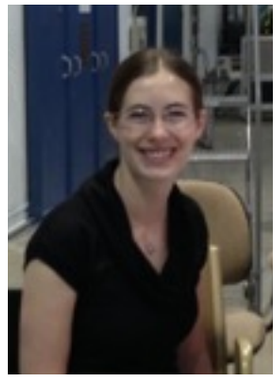
Seismological Laboratory
Geological & Planetary Sciences
Caltech

Workshop on Nuclear Resonant Scattering
Advanced Photon Source, Argonne National Laboratory
12 November 2016

Acknowledgements



Greg Finkelstein
SeismoLab Director's Postdoctoral Fellow



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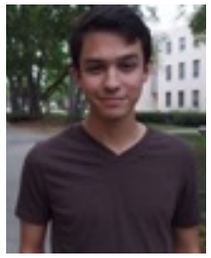
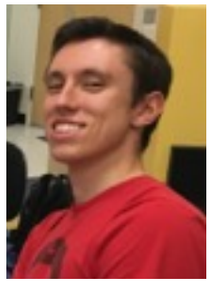


Tyler Perez
Ayya Alieva
Undergraduate Students



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Dan Bower: now Postdoctoral Fellow at ETH, Zurich
Bin Chen: now Asst. Professor, Univ. of Hawaii
Caitlin Murphy: now Congressional Fellow, Washington DC
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Aaron Wolf: now Asst. Research Scientist, Univ. of Michigan
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Advanced Photon Source
Ercan Alp, Tom Toellner, Jiyong Zhao, Wenli Bi, Michael Hu (NRS)
Mark Rivers, Vitali Prakapenka (GSECARS)
Guoyin Shen (HP-CAT)

COMPRES
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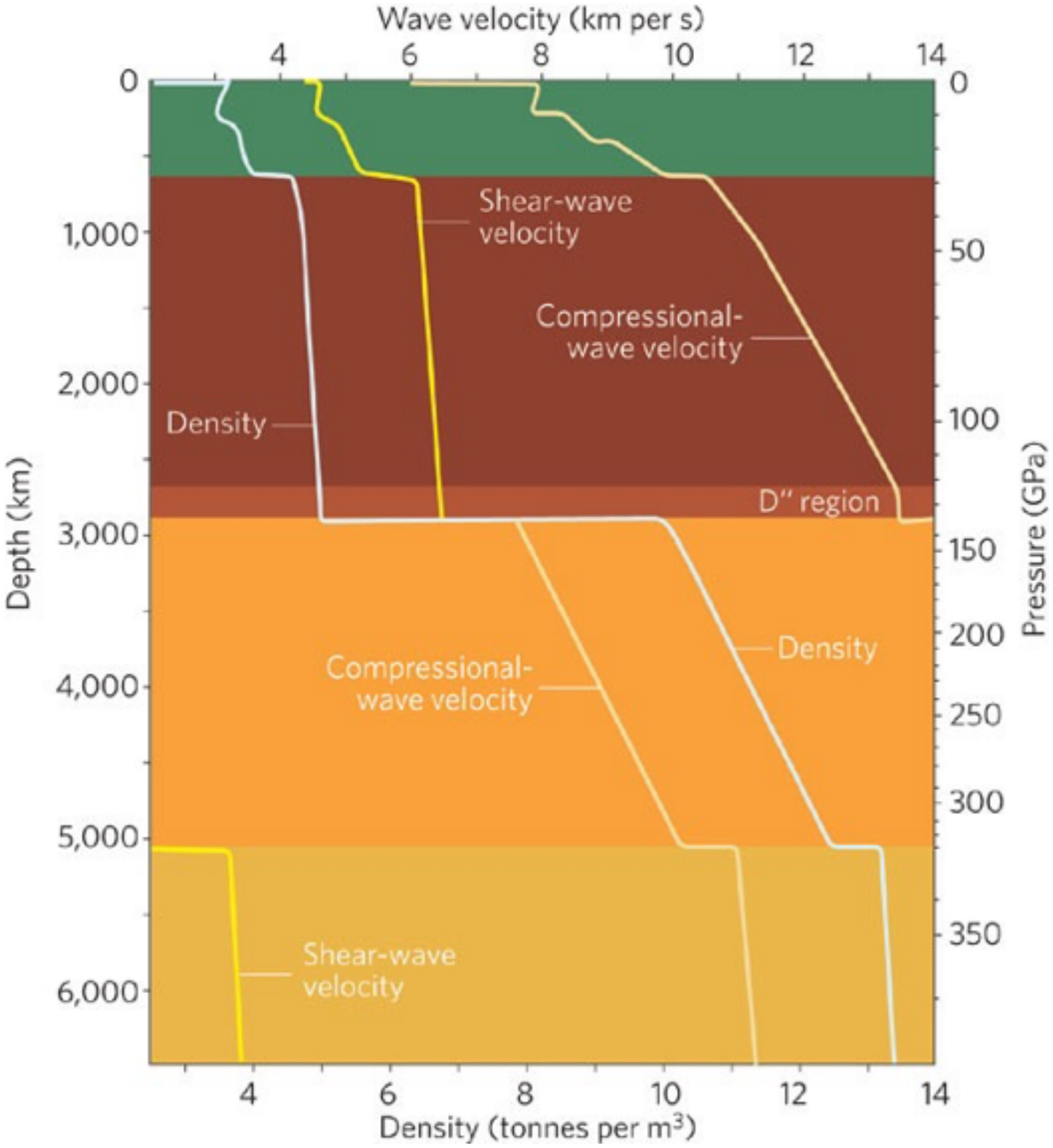
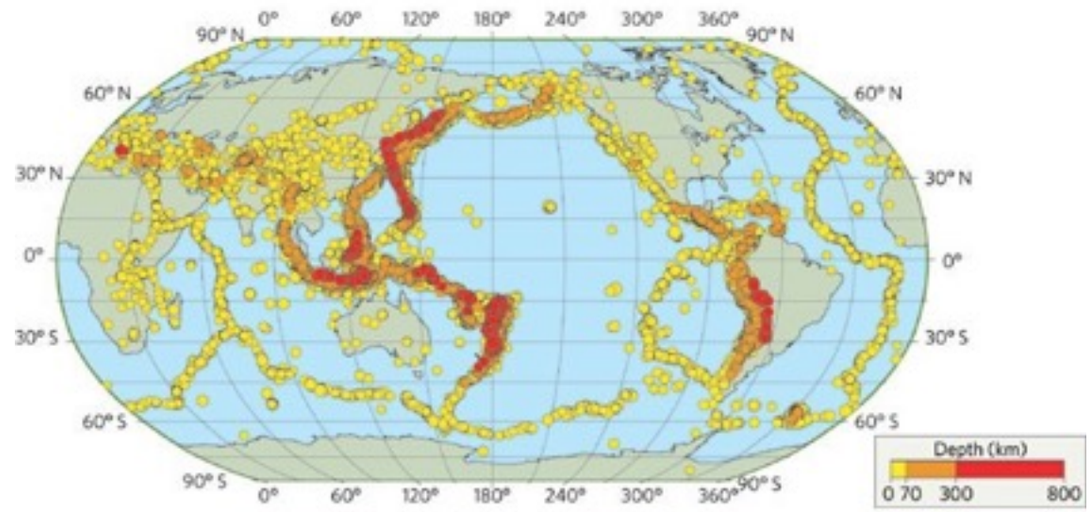
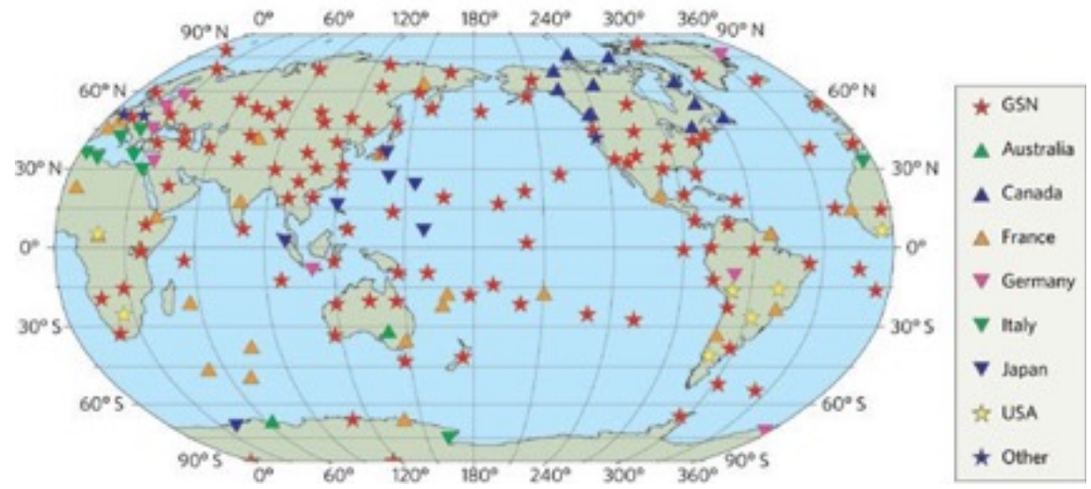
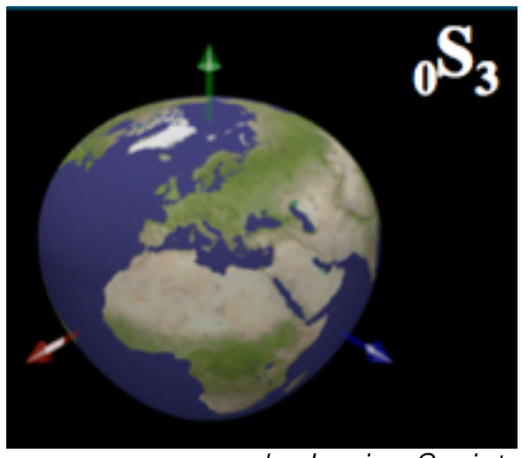
The first-order structure of Earth's interior





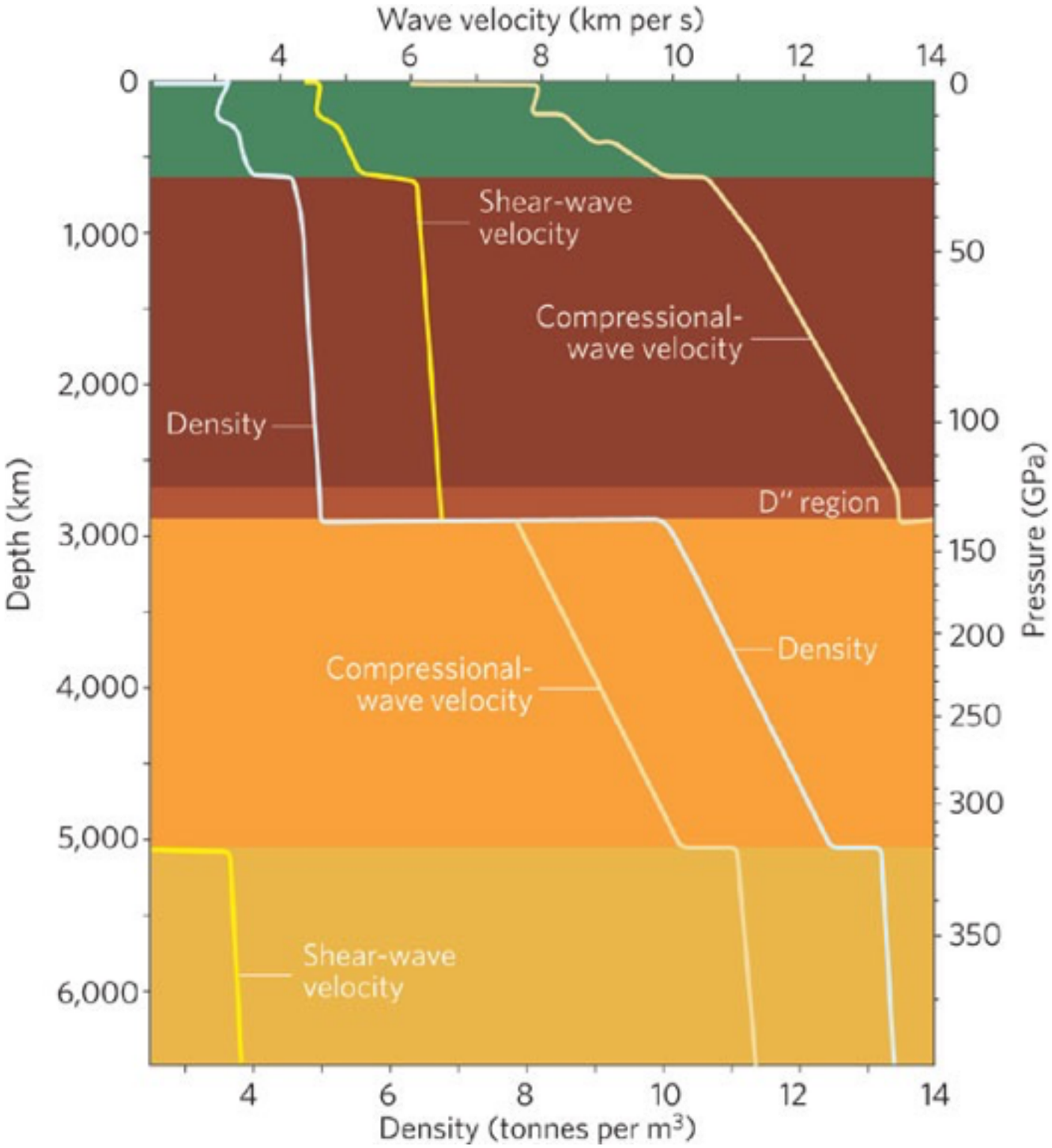


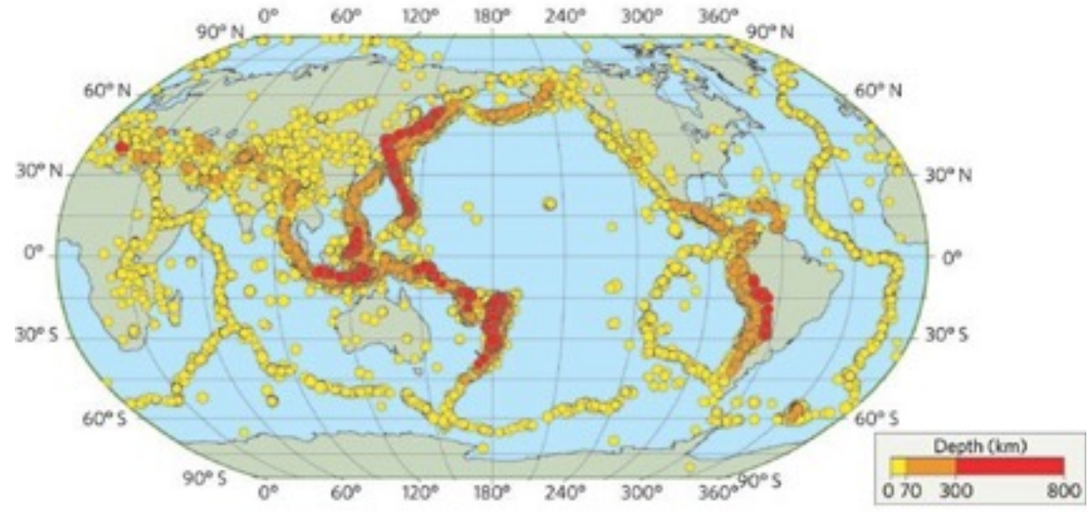


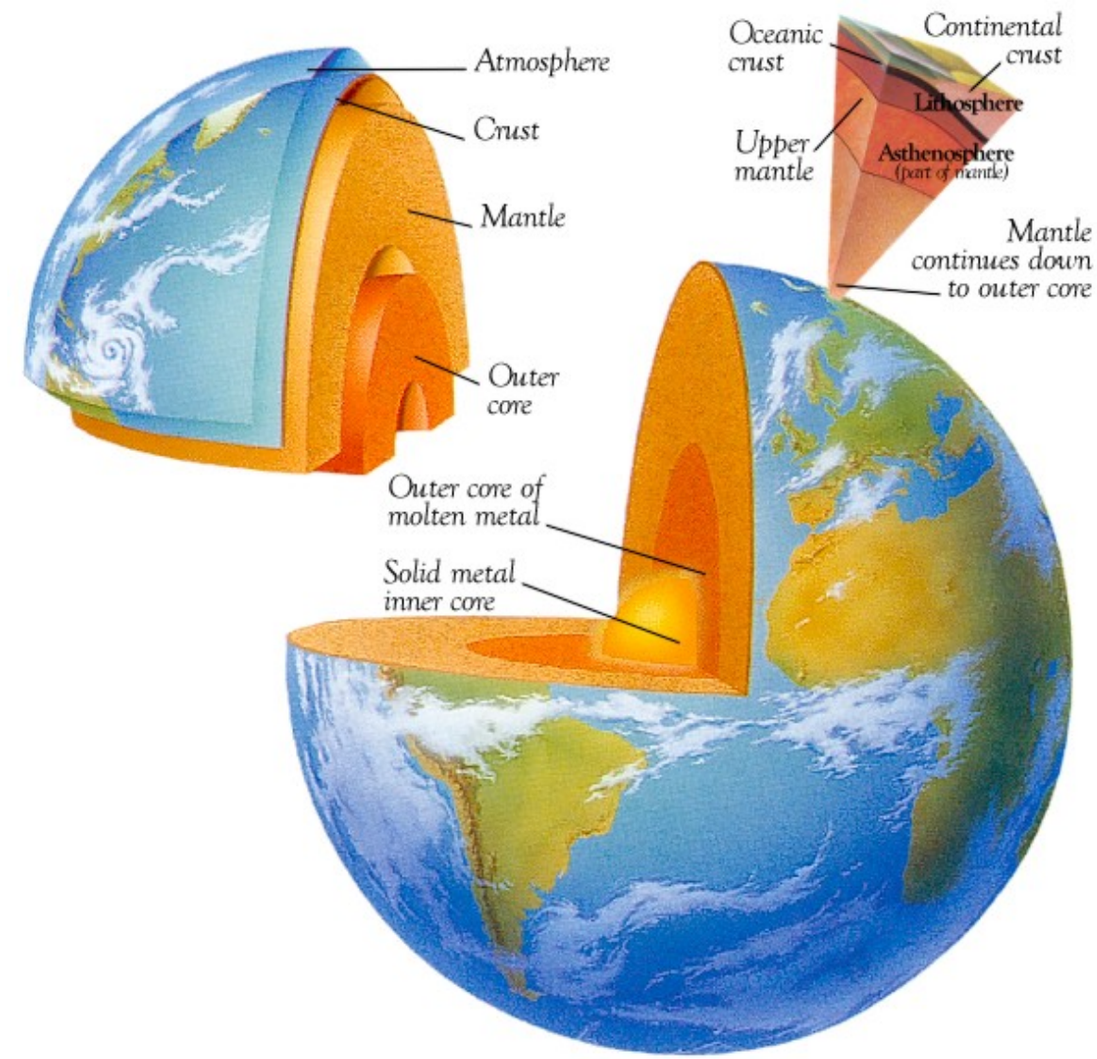
by Lucien Saviot

Preliminary Reference Earth Model (PREM): Dziewonski & Anderson (1981)
Romanowicz, Nature (2008), *Discovery Channel*

The first-order structure of Earth's interior

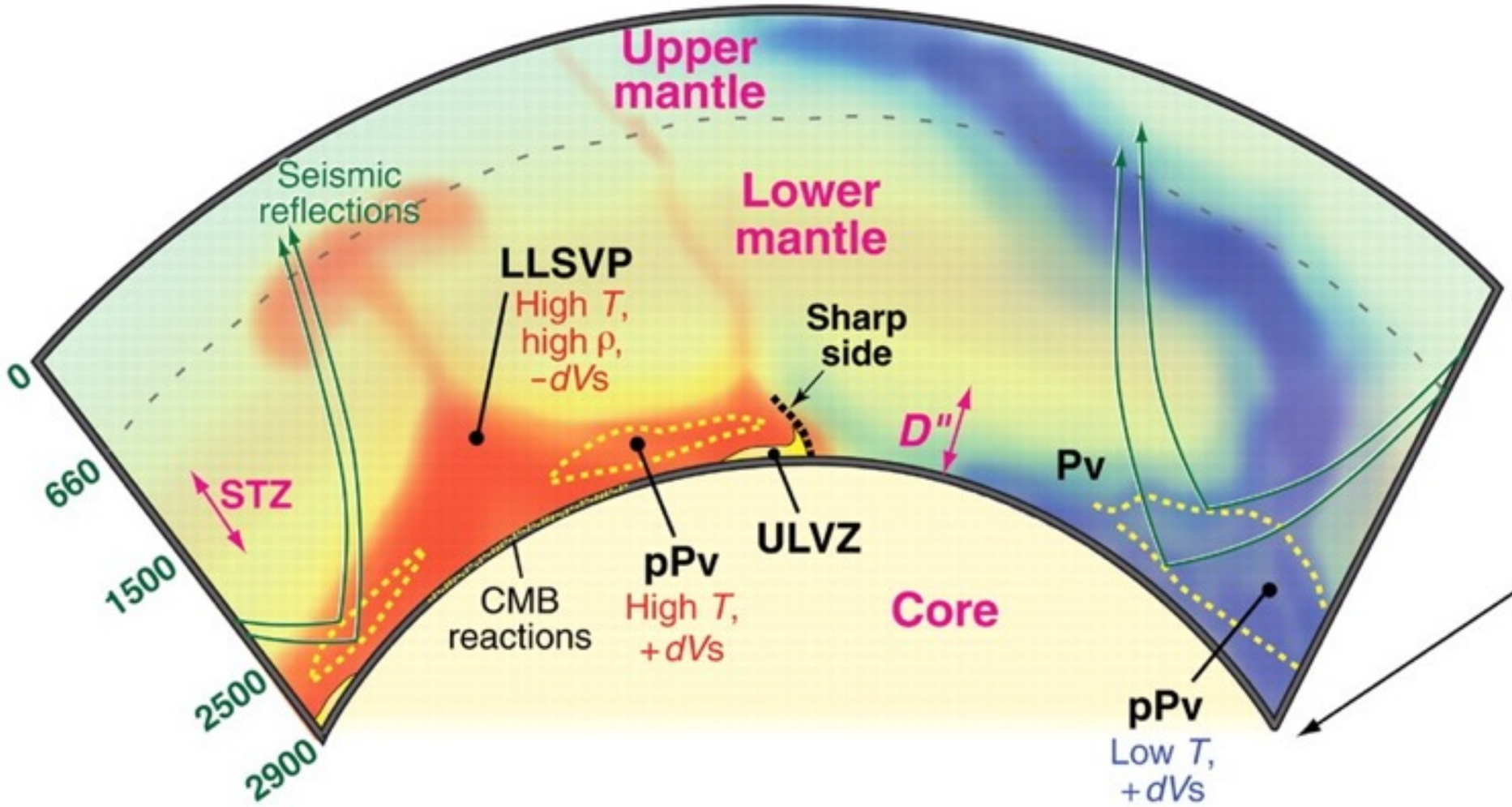


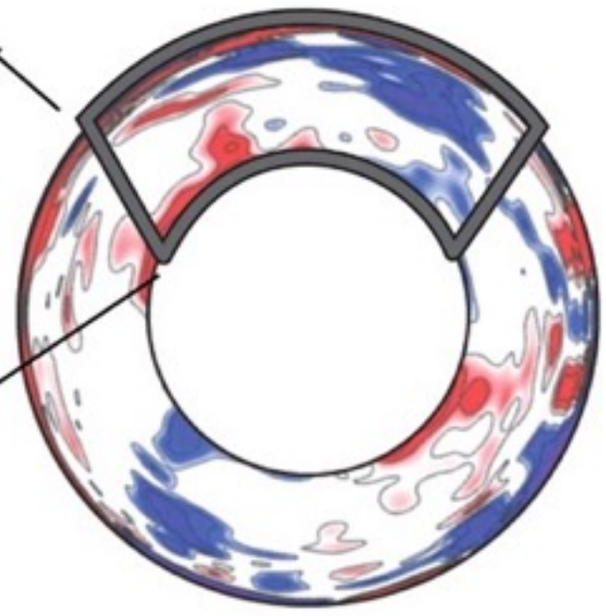




Preliminary Reference Earth Model (PREM): Dziewonski & Anderson (1981)
Romanowicz, Nature (2008), *Discovery Channel*

Perturbations from the 1D model reveal multi-scale features





Sound velocities
Thermal expansion
Thermal pressure
Grüneisen parameter

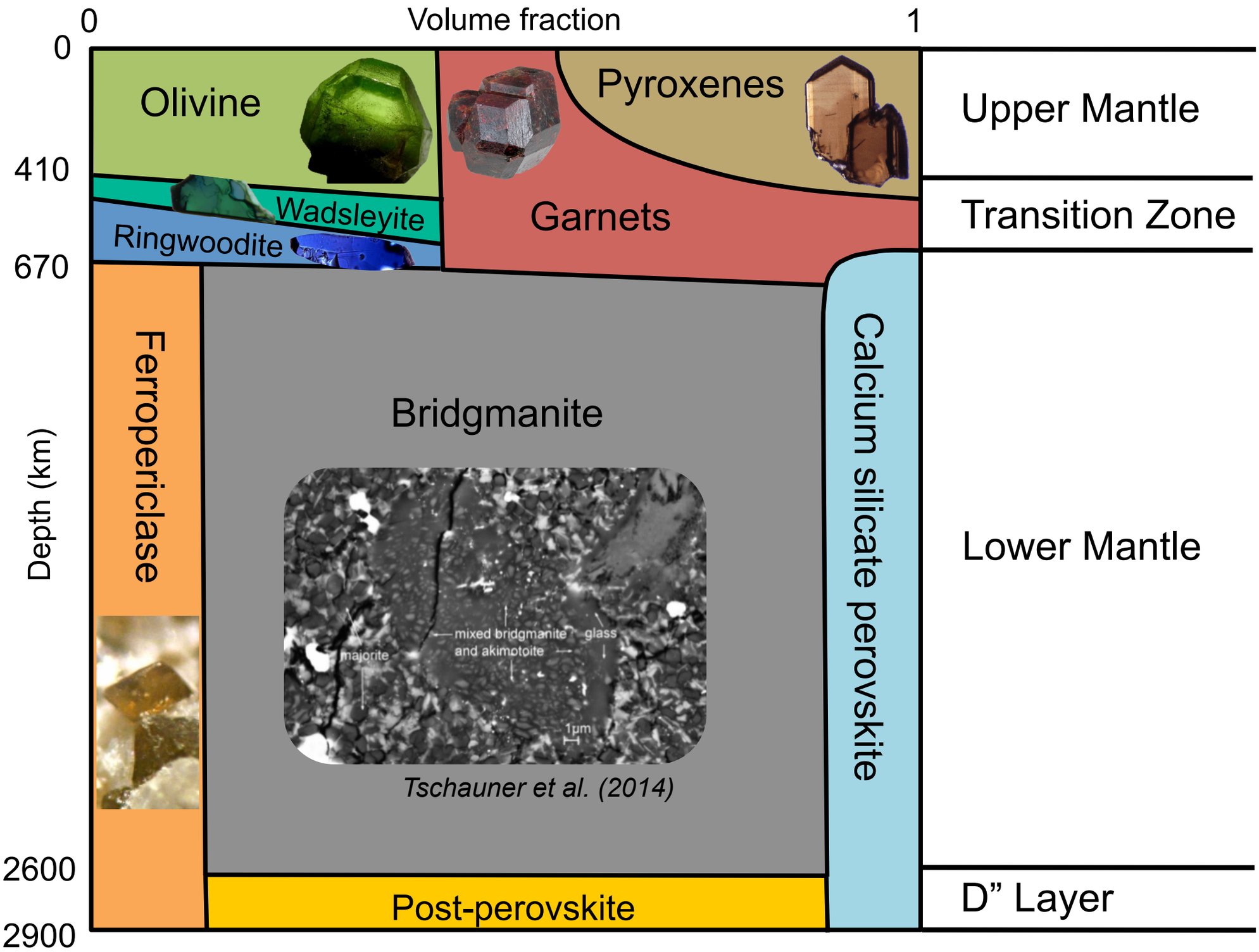
Nuclear resonant inelastic x-ray scattering

Oxidation and spin states of iron
Magnetic ordering
Melting

Synchrotron Mössbauer spectroscopy

Garnero & McNamara (2008), S20RTSb: *Ritsema et al.* (2004)

Minerals in a peridotitic mantle



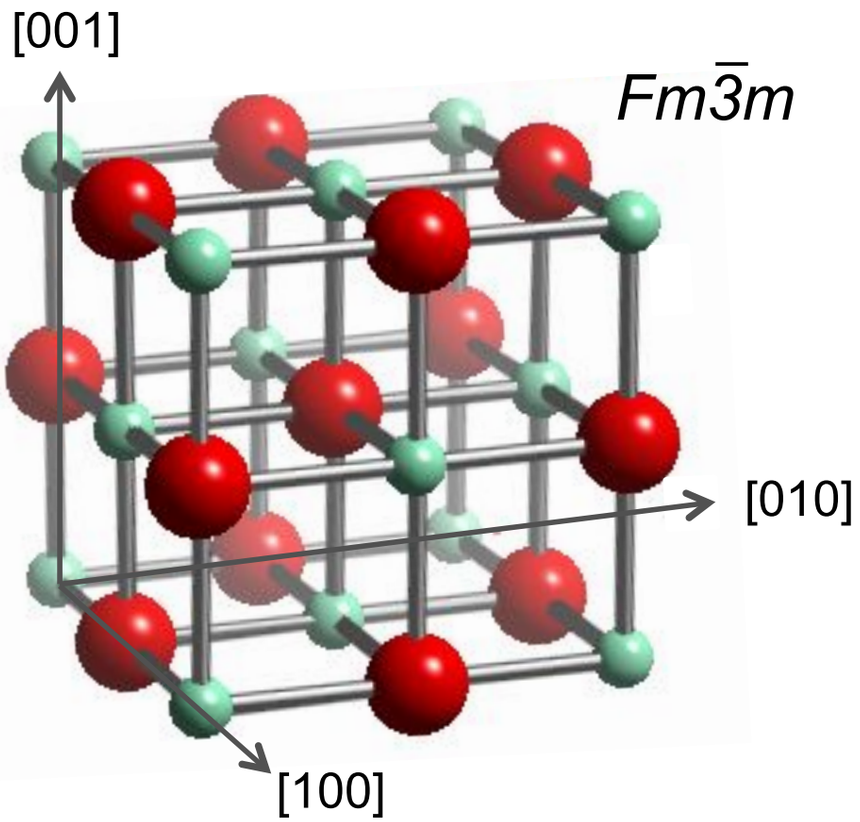
Tschauner et al. (2014)

Figure created by Natalia Solomatova. After J. Amodeo; OPX from minerals.gps.caltech.edu; olivine by Rob Lavinsky; wadsleyite by T. Kawazoe; ringwoodite by Joe Smyth

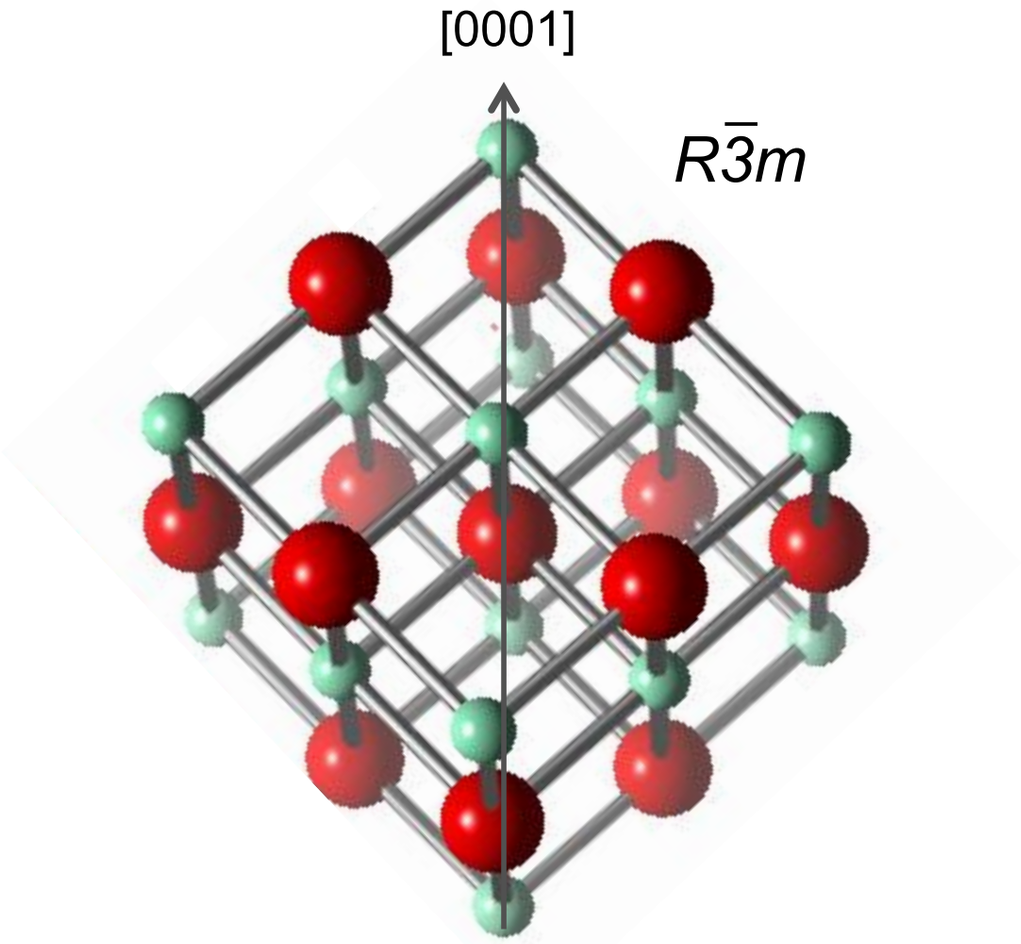
Periclase - Wüstite Solid Solution

MgO $(Mg_{0.5}Fe_{0.5})O$ FeO

"ferropericlase" "magnesiowüstite"



B1 phase



rhombohedral phase

Electronic spin pairing in octahedral ferrous iron

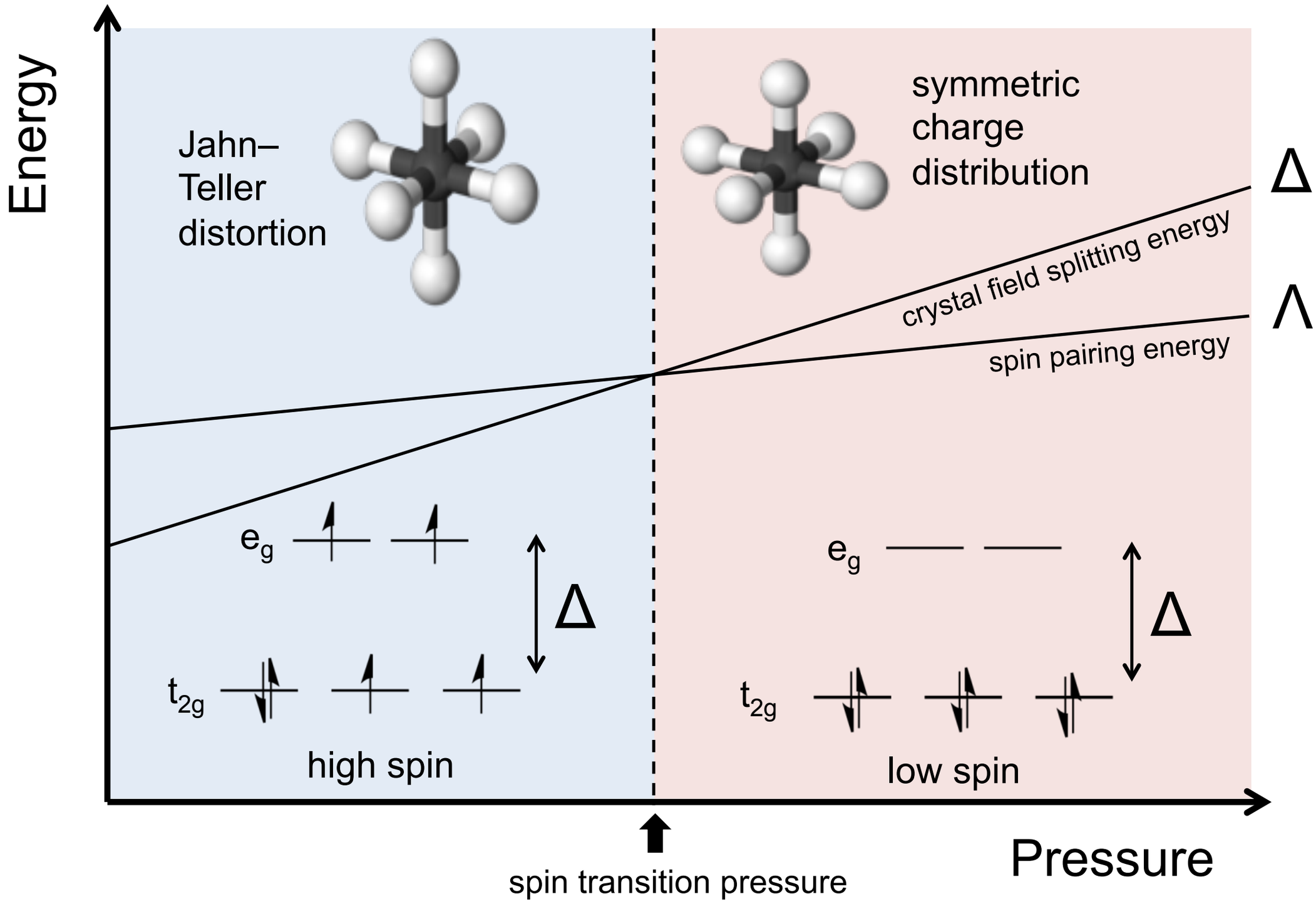


Figure prepared by Natalia Solomatova

Previous Experiments on (Mg,Fe)O

FeO %	Methods	Pressure Mediums	Pressure Scales	Spin Transition Pressure
MgO  FeO	○ Conventional Mössbauer ○ X-ray emission ○ Powdered x-ray diffraction ○ Single-crystal x-ray diffraction ○ Synchrotron Mössbauer ○ Optical absorption	○ None ○ Boron epoxy ○ NaCl ○ KCl ○ Ar ○ Alcohol mixtures ○ Ne ○ He	○ Ruby ○ NaCl ○ Pt ○ Au	○ 50% volume collapse ○ 50% low spin population ○ Spin pairing energy = crystal field splitting energy ○ Visual inspection of data

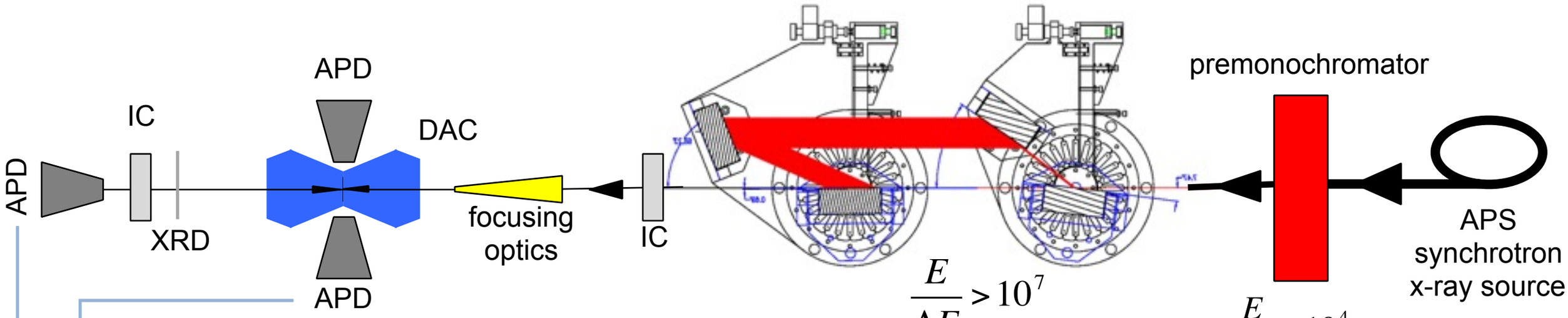
Fit a spin crossover equation of state to previous pressure-volume data sets for compositions ranging from 10-60 mol% FeO.

Compare resulting equation of state parameters.

Directly compare to results from Mössbauer spectroscopy, which is sensitive to iron's spin state.

$(Mg_{(1-x)}Fe_x)O$ with x=0.10 (Marquardt et al., 2009), x=0.17 (Lin et al., 2005), x=0.25 (Mao et al., 2011), x=0.35 (Chen et al., 2012), x=0.39 (Zhuravlev et al., 2010; Fei et al., 2007) and x=0.60 (Lin et al., 2005)

Experimental setup for nuclear resonant scattering: Sector 3-ID-B, Advanced Photon Source



$\frac{E}{\Delta E} > 10^7$

$\Delta E = 1.0$ meV

silicon
high-resolution
monochromator

$\frac{E}{\Delta E} = 10^4$

$E = 14.4125$ keV

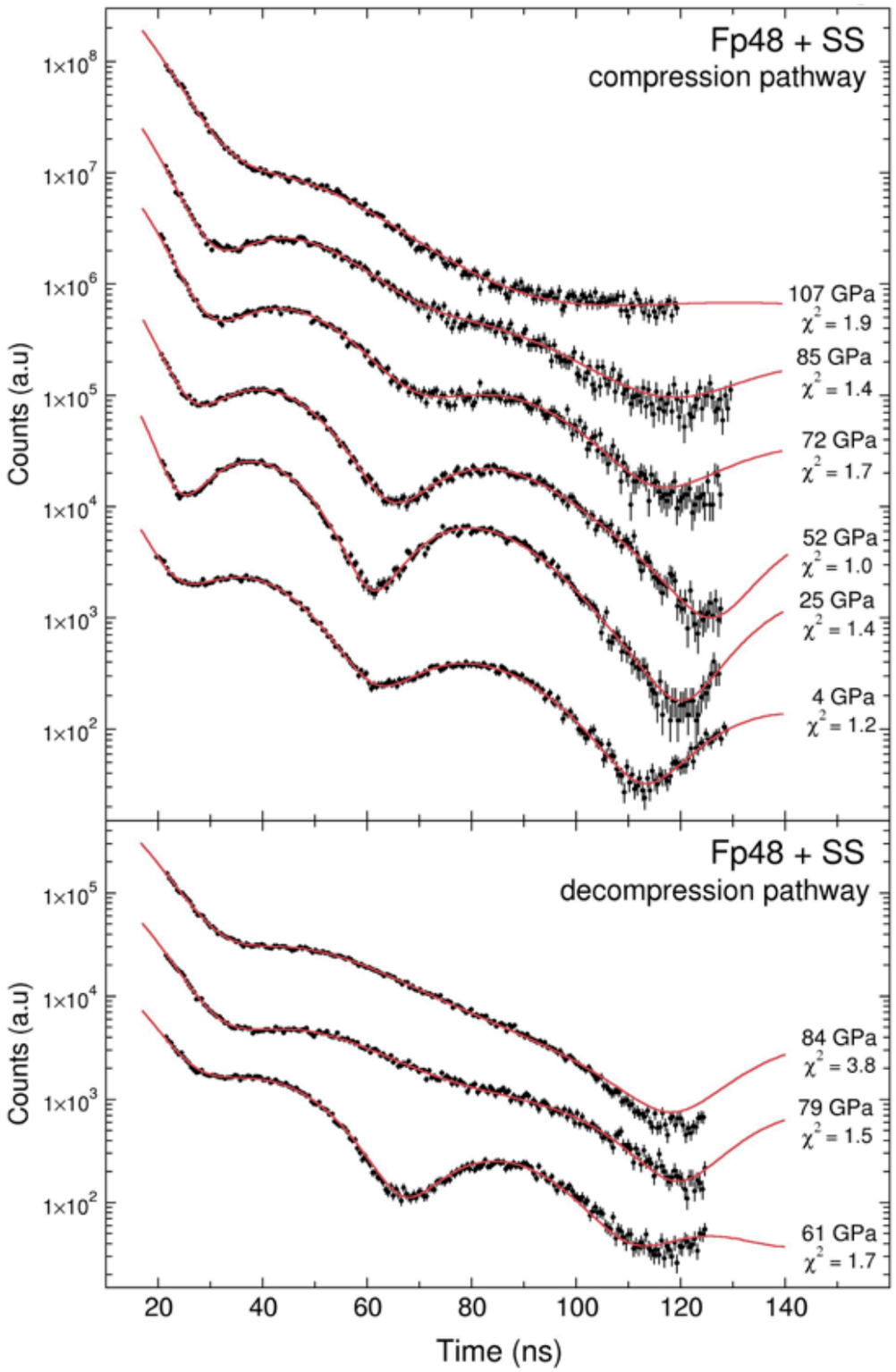
$\Delta E \approx 1$ eV

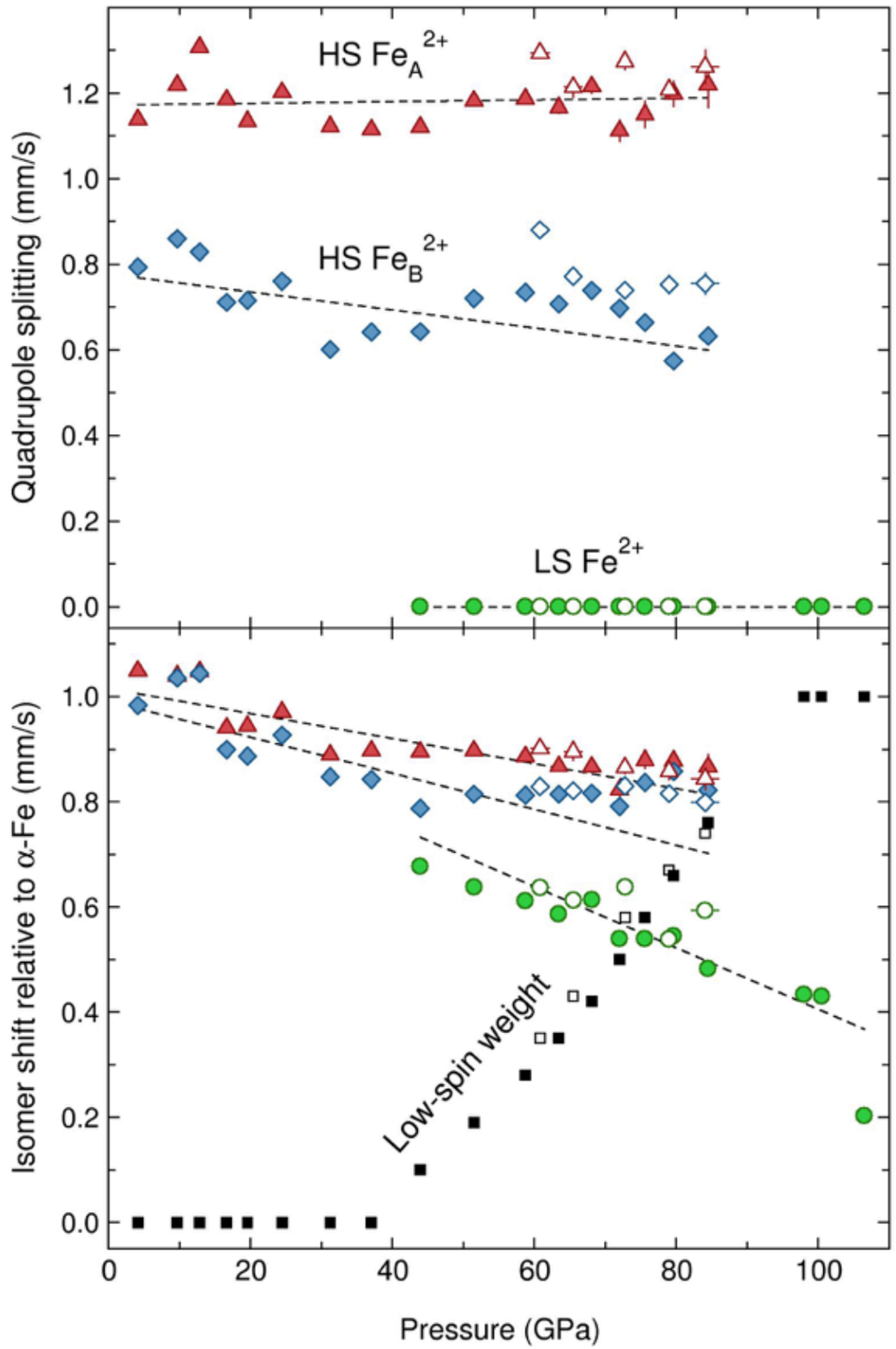
Nuclear resonant inelastic x-ray scattering
(Partial) projected phonon DOS:
thermodynamic properties (γ, P_{th} α), lnβ
sound velocities (V_D, V_P, V_S)

Nuclear resonant forward scattering
(Synchrotron Mössbauer spectroscopy):
valence, spin, magnetic ordering,
melting

e.g., Sturhahn (*JCPM* 2004); Toellner (*HI* 2000, *JSR* 2011);
Sturhahn & Jackson (*GSA* 2007); Jackson (*Springer* 2010)

Synchrotron Mössbauer spectroscopy study of (Mg,Fe)O





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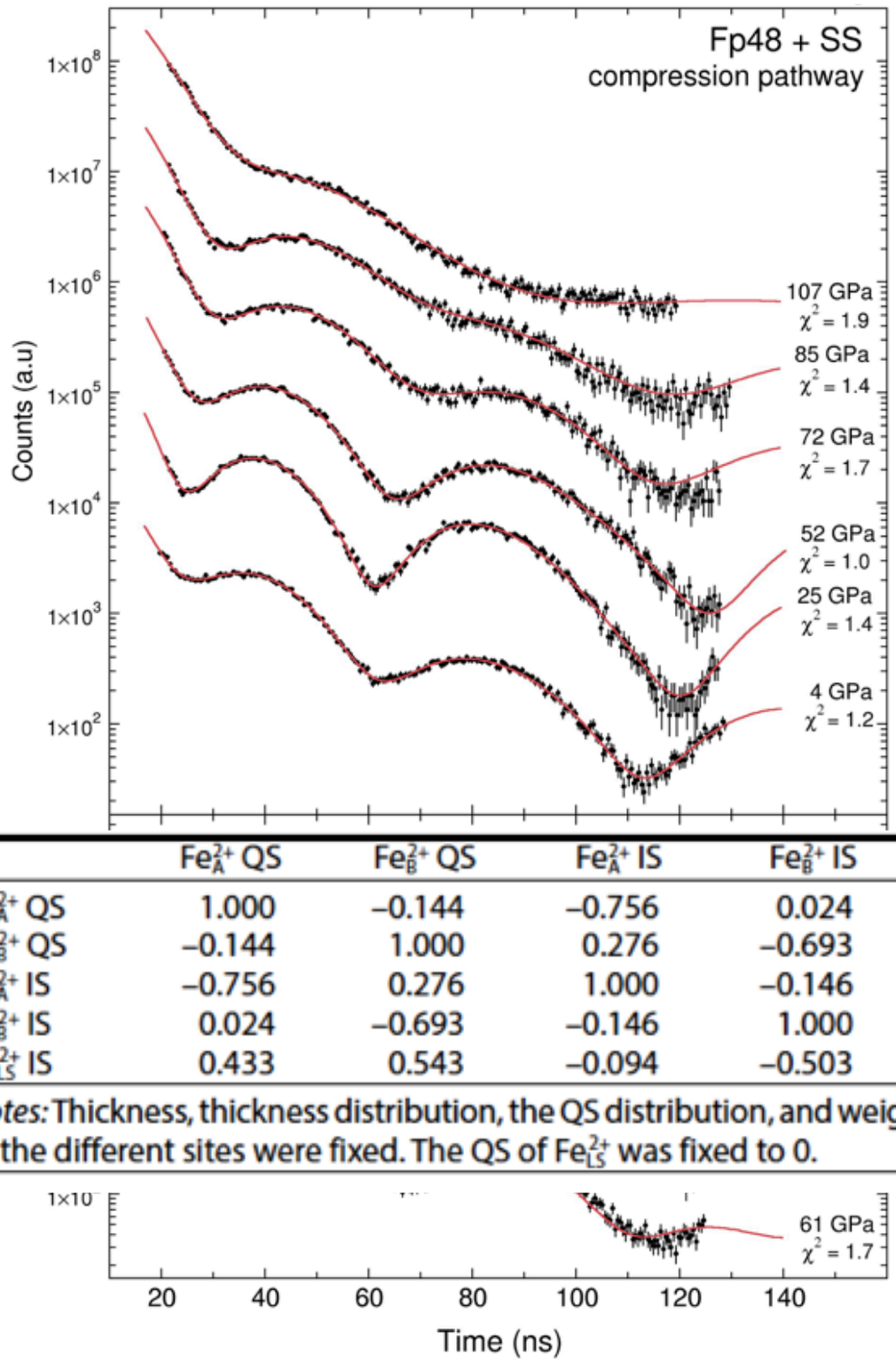
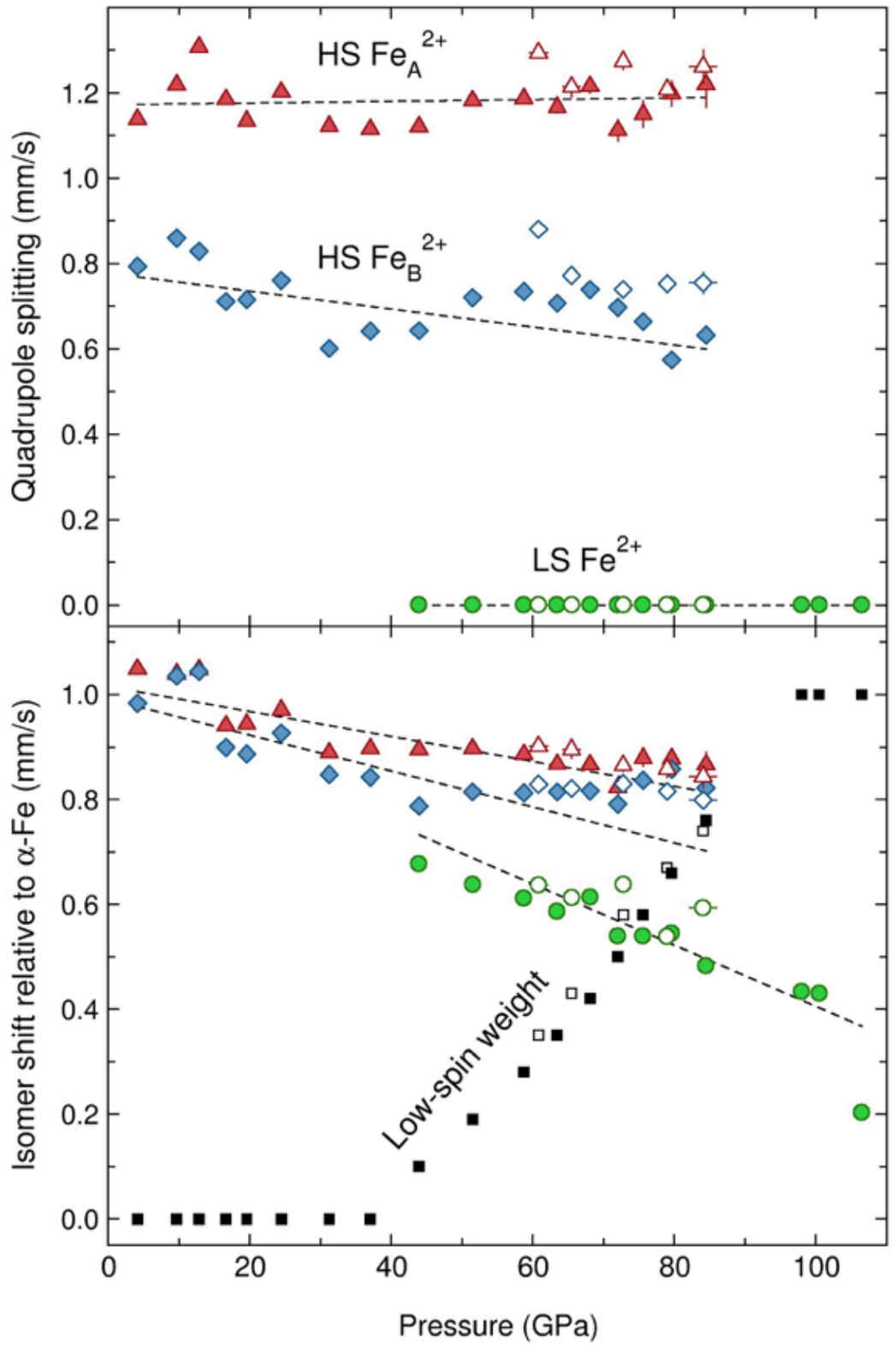


	Fe_A^{2+} QS	Fe_B^{2+} QS	Fe_A^{2+} IS	Fe_B^{2+} IS	Fe_{LS}^{2+} IS
Fe_A^{2+} QS	1.000	–0.144	–0.756	0.024	0.433
Fe_B^{2+} QS	–0.144	1.000	0.276	–0.693	0.543
Fe_A^{2+} IS	–0.756	0.276	1.000	–0.146	–0.094
Fe_B^{2+} IS	0.024	–0.693	–0.146	1.000	–0.503
Fe_{LS}^{2+} IS	0.433	0.543	–0.094	–0.503	1.000

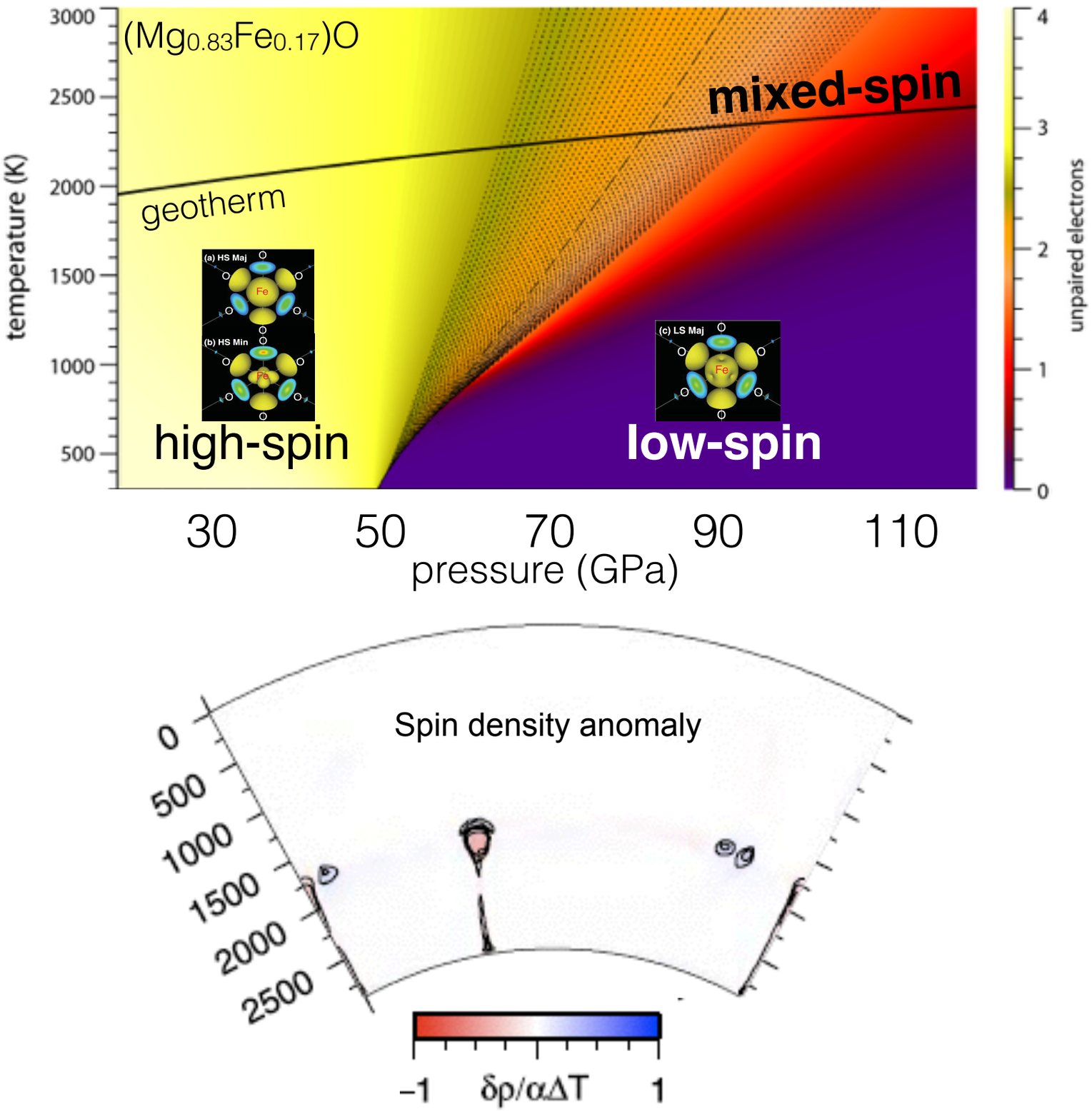
Notes: Thickness, thickness distribution, the QS distribution, and weight fractions of the different sites were fixed. The QS of Fe_{LS}^{2+} was fixed to 0.



CONUSS *Solomatova et al. Am. Min.* (2016)

Jennifer M. Jackson

Ferropericlase spin buoyancy

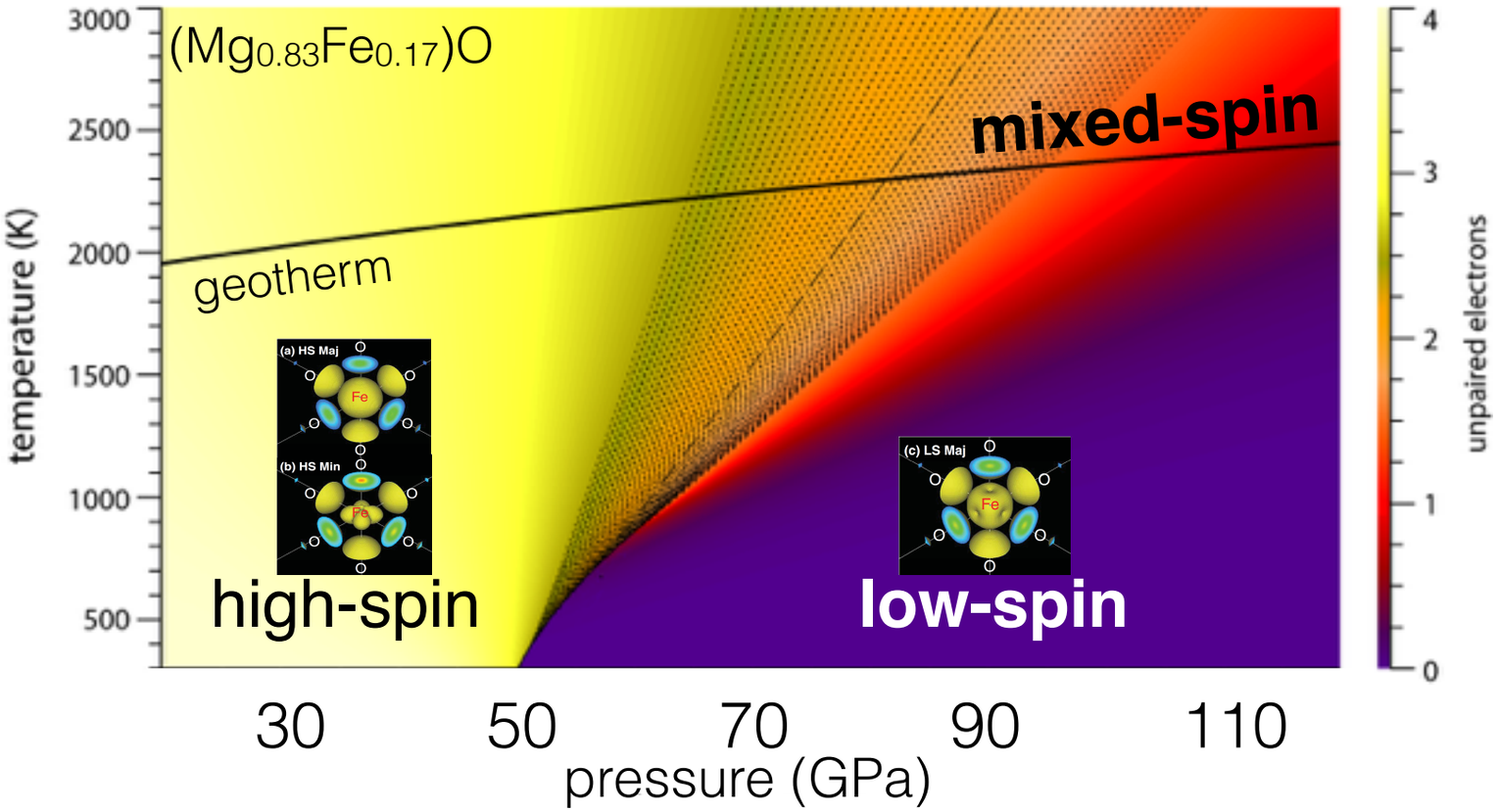


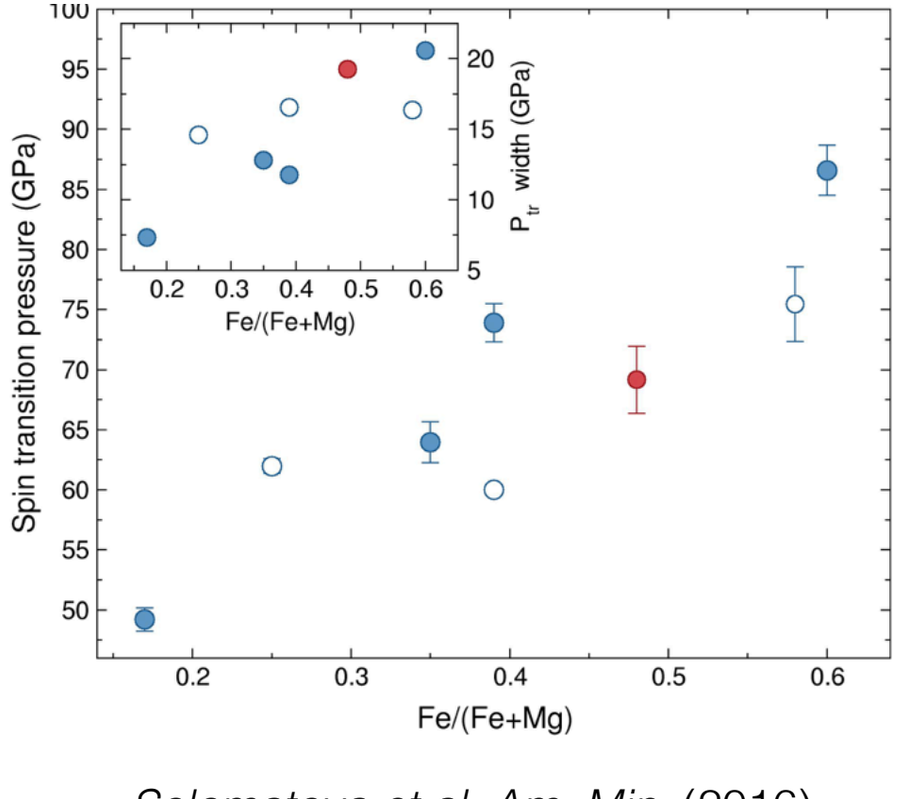
Solomatova et al. Am. Min. (2016)

Compare with first principles at 300 K:
Tsuchiya et al. (2006):
P_{spinTr} = 36 GPa
$Width_{spinTr}$ = 4 GPa

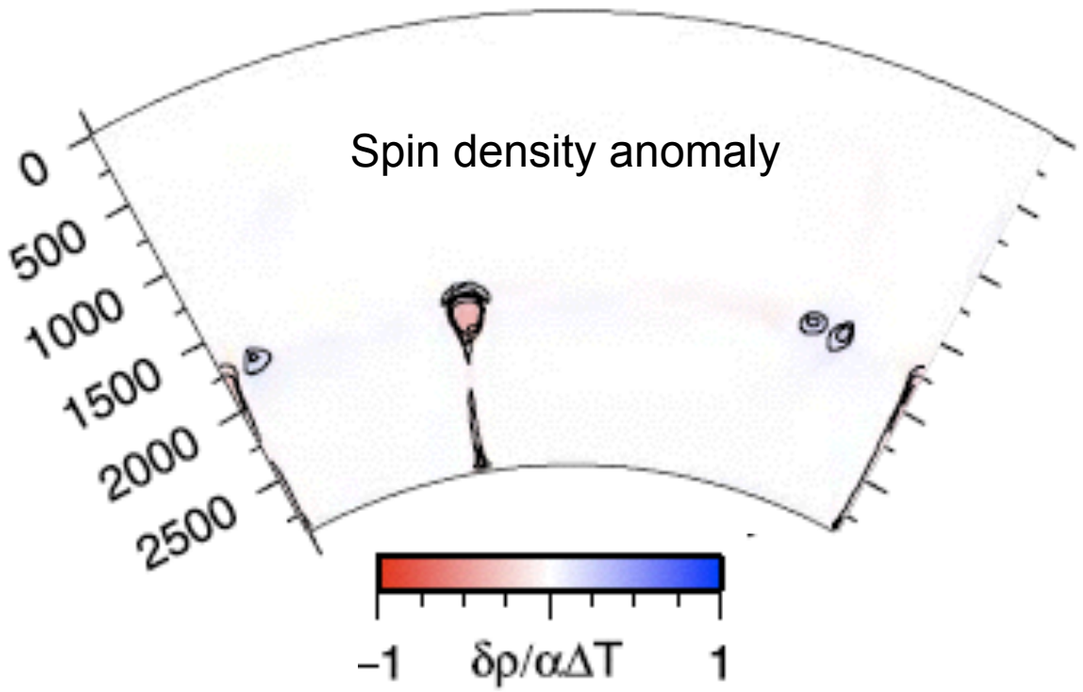
Holström & Stixrude (2015):
P_{spinTr} = 65 GPa
$Width_{spinTr}$ = 50 GPa

Ferropericlase spin buoyancy





Solomatova et al. Am. Min. (2016)



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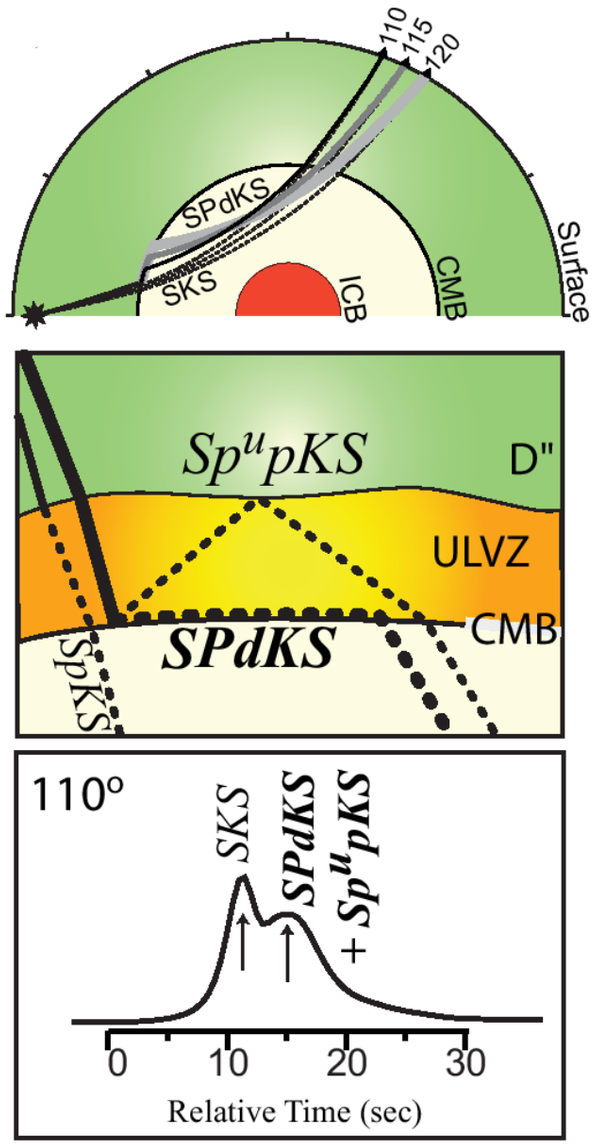
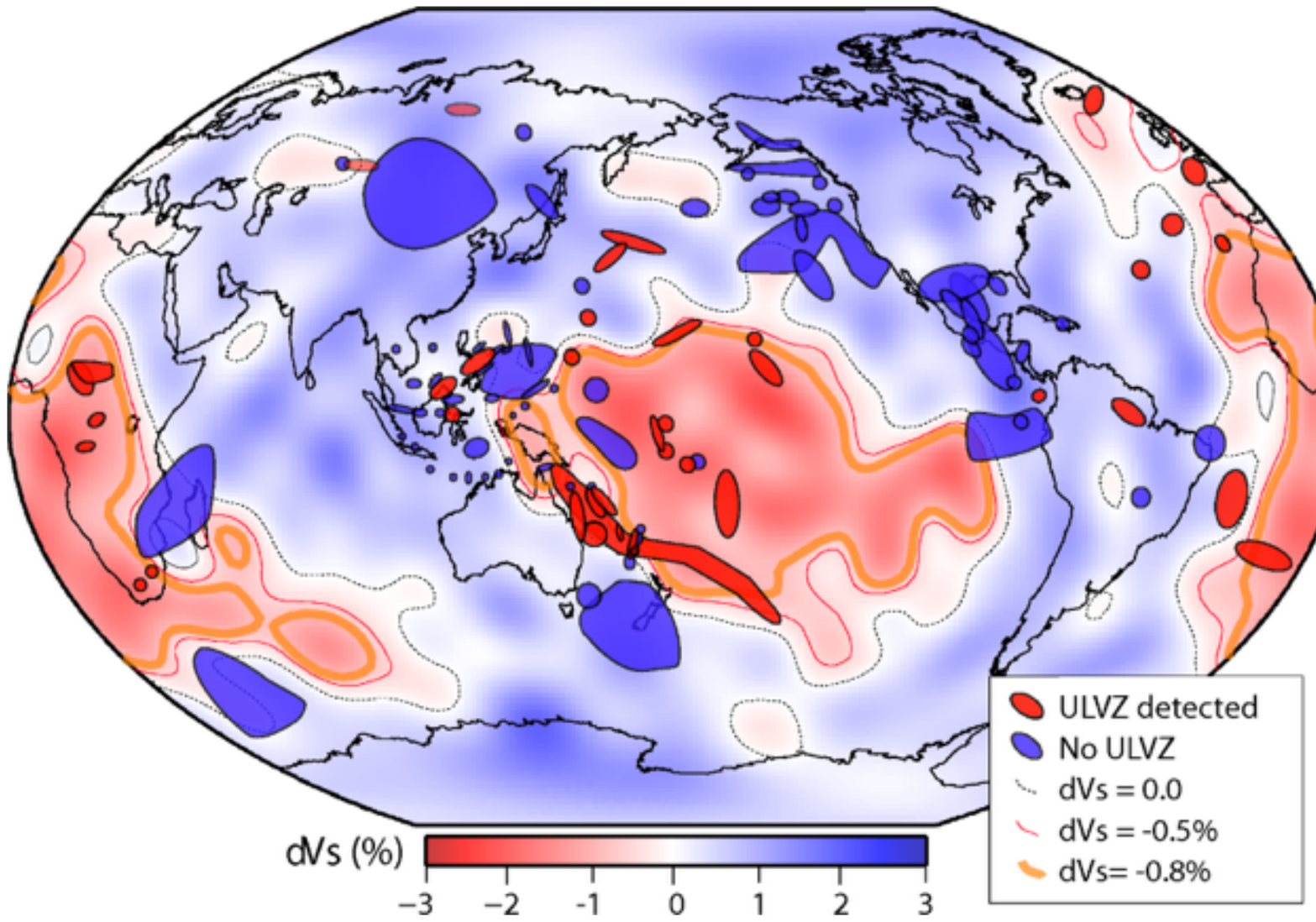
Distribution of low velocity zones at the base of the mantle





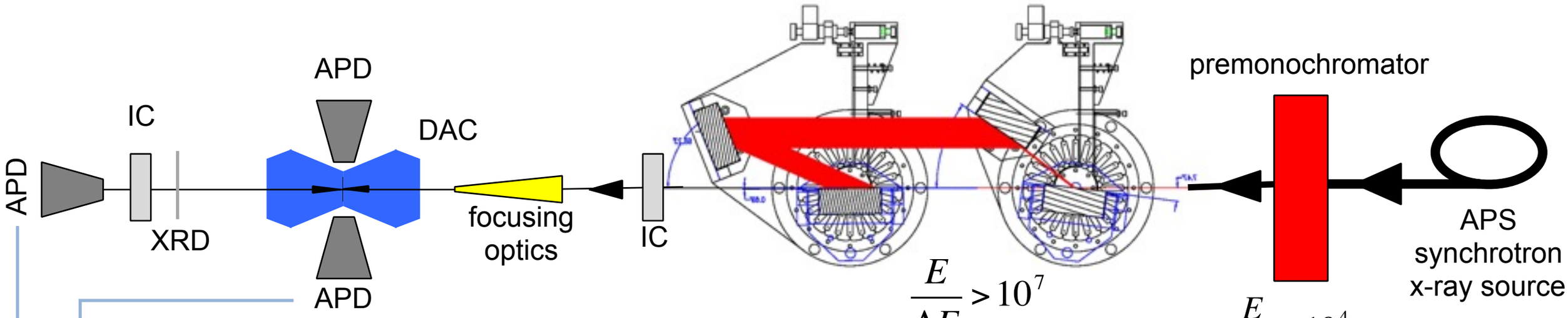
Slow
5 to 20% drop in V_P
10 to 30% drop in V_S

V_P/V_S
Variable
Not well-constrained

Dense
4 to 14% denser
Not well-constrained

McNamara et al. EPSL (2010)

Experimental setup for nuclear resonant inelastic scattering: Sector 3-ID-B, Advanced Photon Source



$\frac{E}{\Delta E} > 10^7$

$\Delta E = 1.0$ meV

silicon
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$\frac{E}{\Delta E} = 10^4$

$E = 14.4125$ keV

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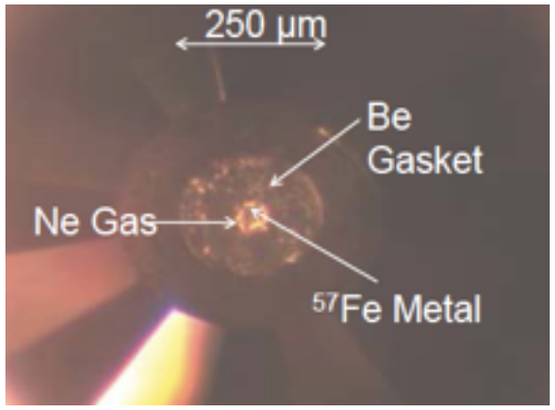
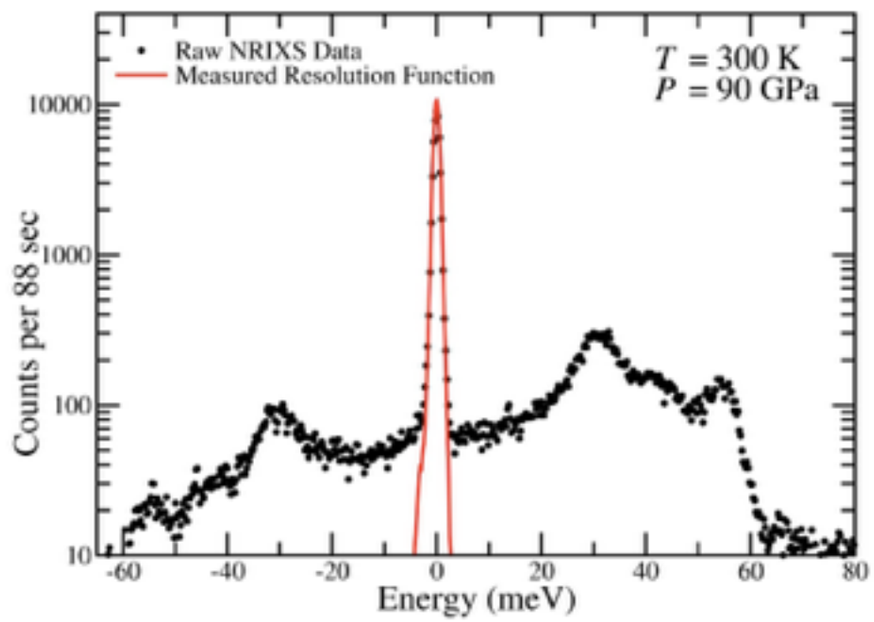
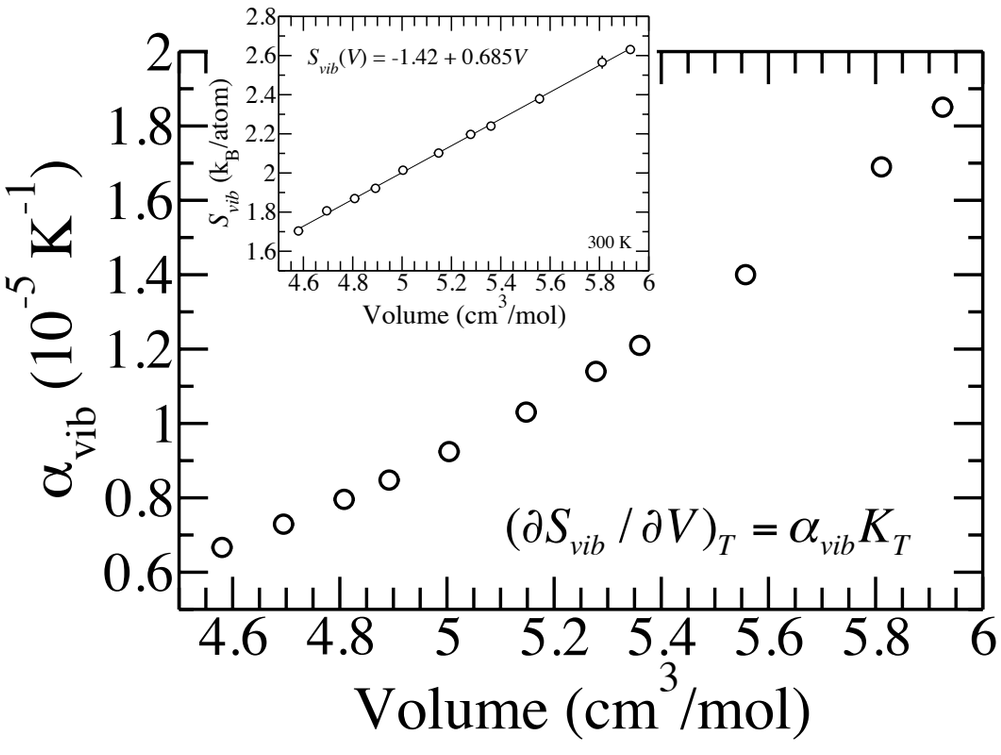
Nuclear resonant inelastic x-ray scattering
(Partial) projected phonon DOS:
thermodynamic properties (γ, P_{th} α), lnβ
sound velocities (V_D, V_P, V_S)

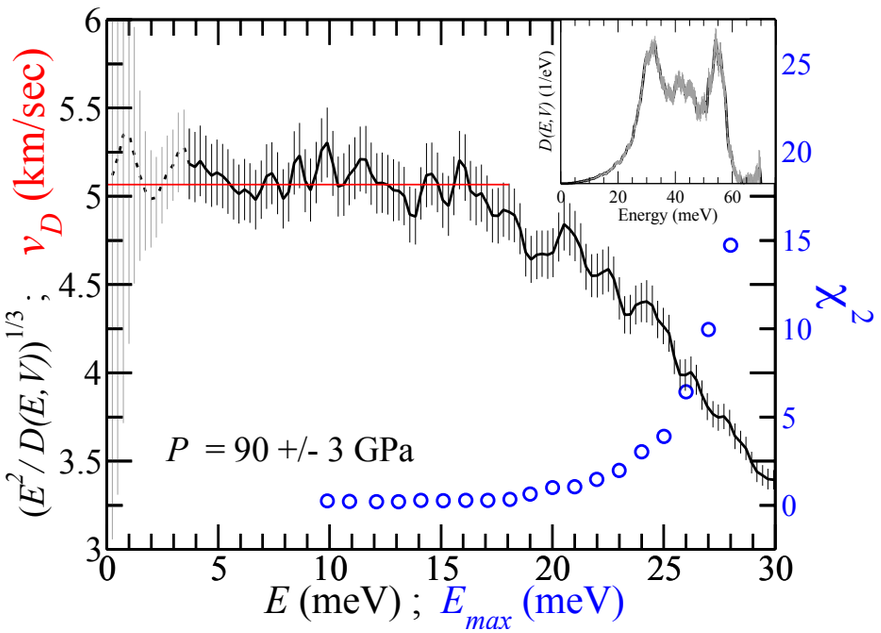
Nuclear resonant forward scattering
(Synchrotron Mössbauer spectroscopy):
Defines the resolution function

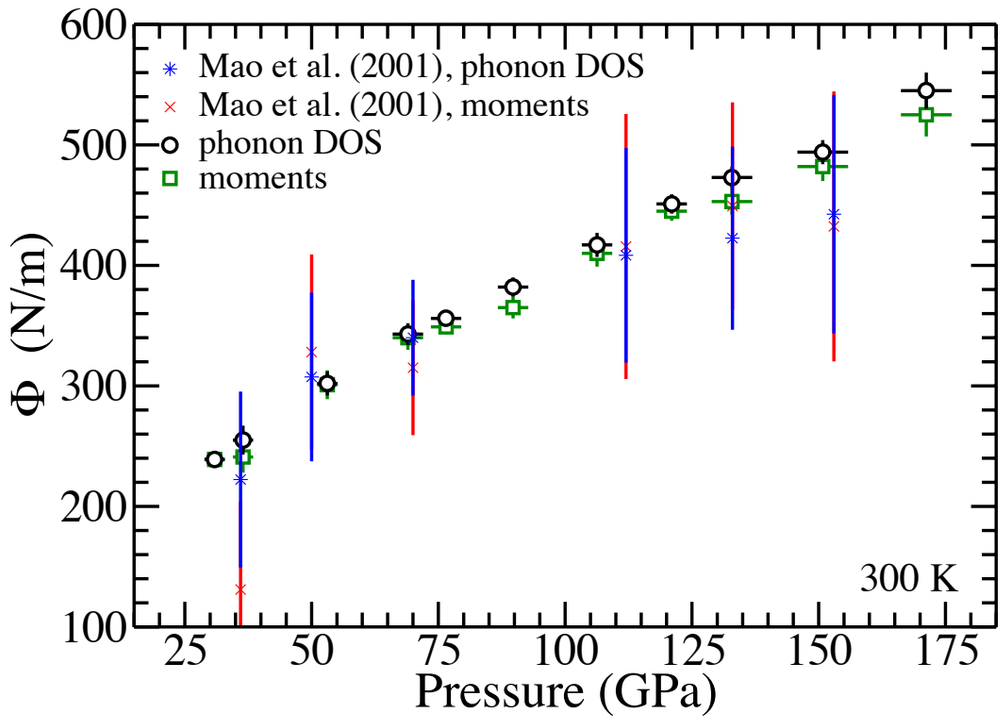
Volume dependence of the phonon DOS for *hcp*-iron to outer core pressures



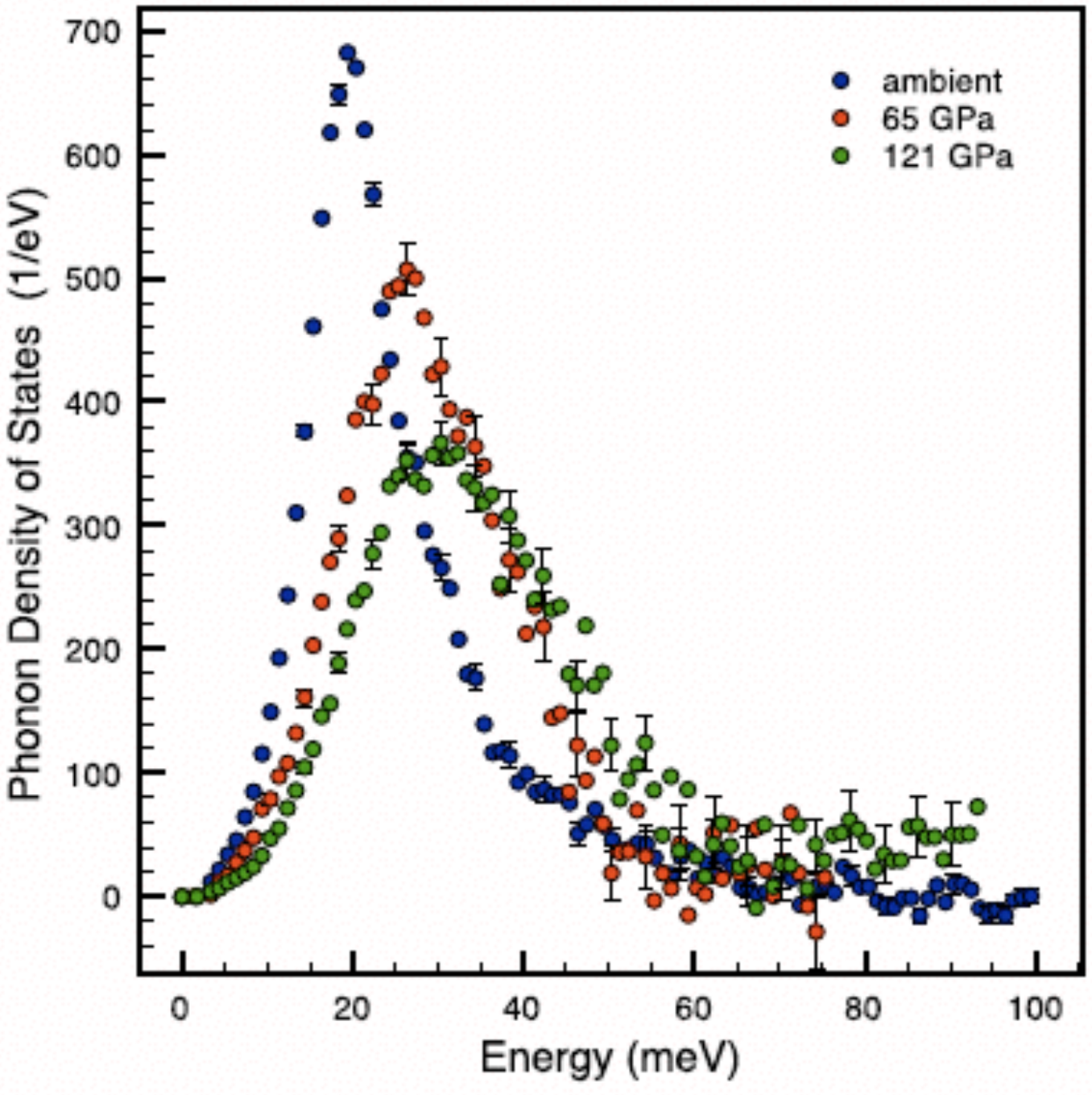


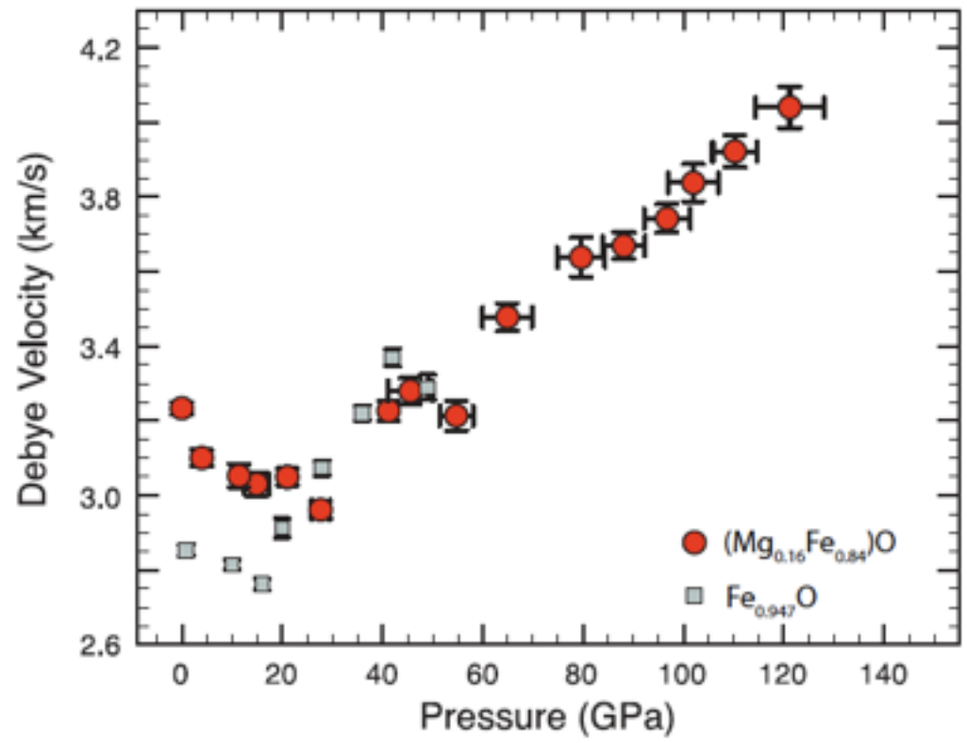




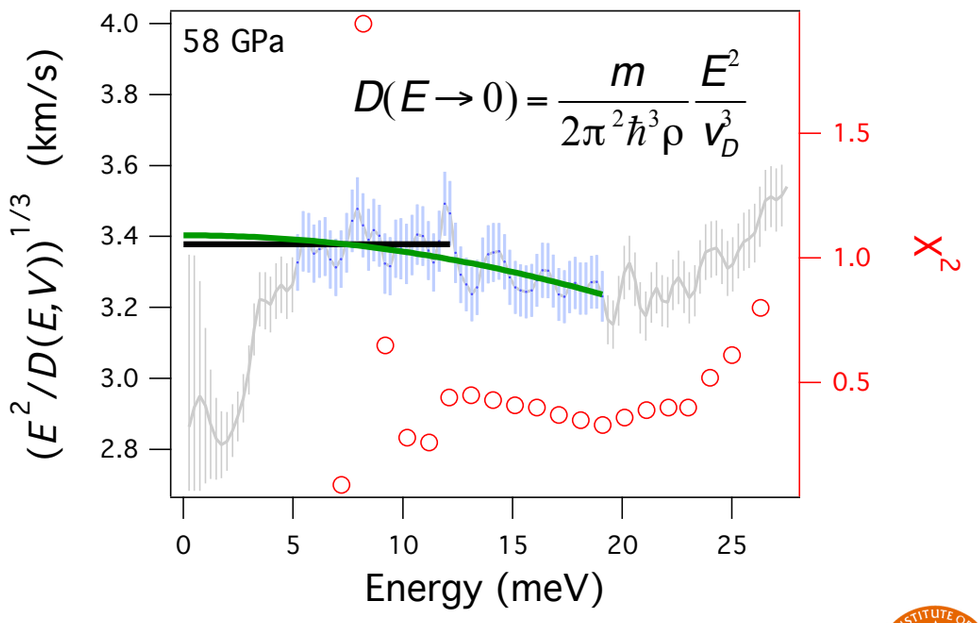


Partial phonon density of states of iron-rich (Mg,Fe)O magnesiowüstites





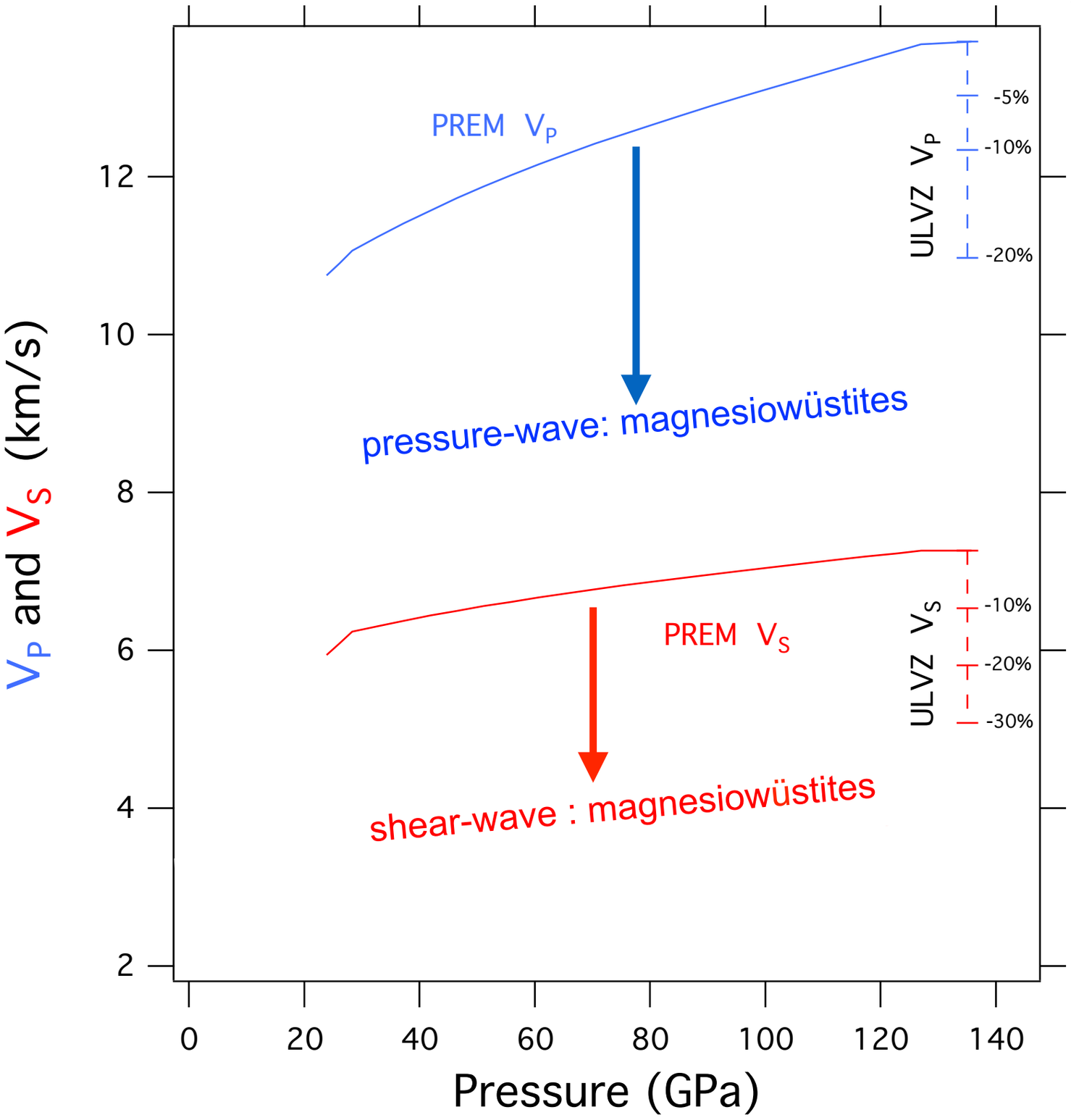
$(Mg_{0.06}Fe_{0.94})O$: Debye velocity, V_D



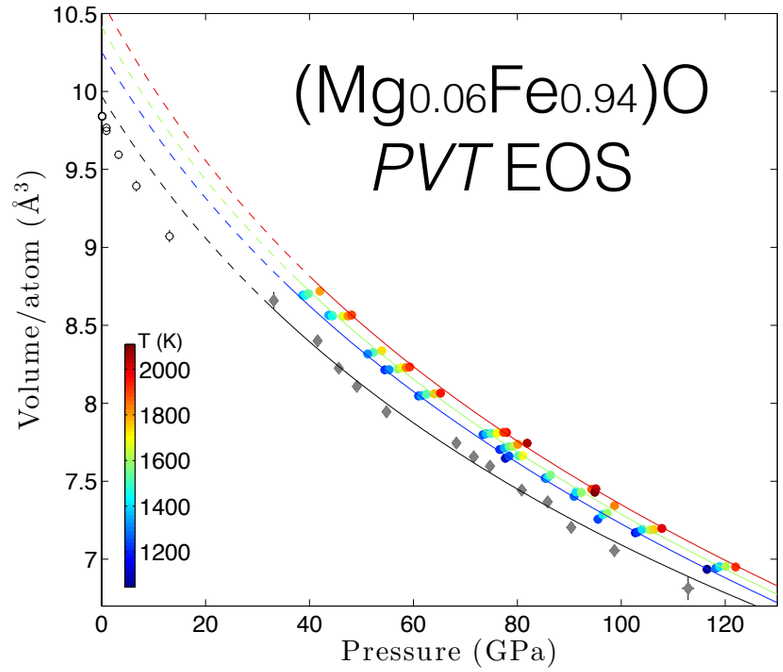
Wicks et al. GRL (2010), Wicks et al. PEPI (2015)
Wicks et al. (2016, under revision)

Ultralow-velocities in magnesiowüstites

$(Mg_{0.16}Fe_{0.84})O$, $(Mg_{0.06}Fe_{0.94})O$, FeO



V_P and V_S



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

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

Wicks et al. GRL (2010), Wicks et al. PEPI (2015)
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Ultralow-velocities in magnesiowüstites

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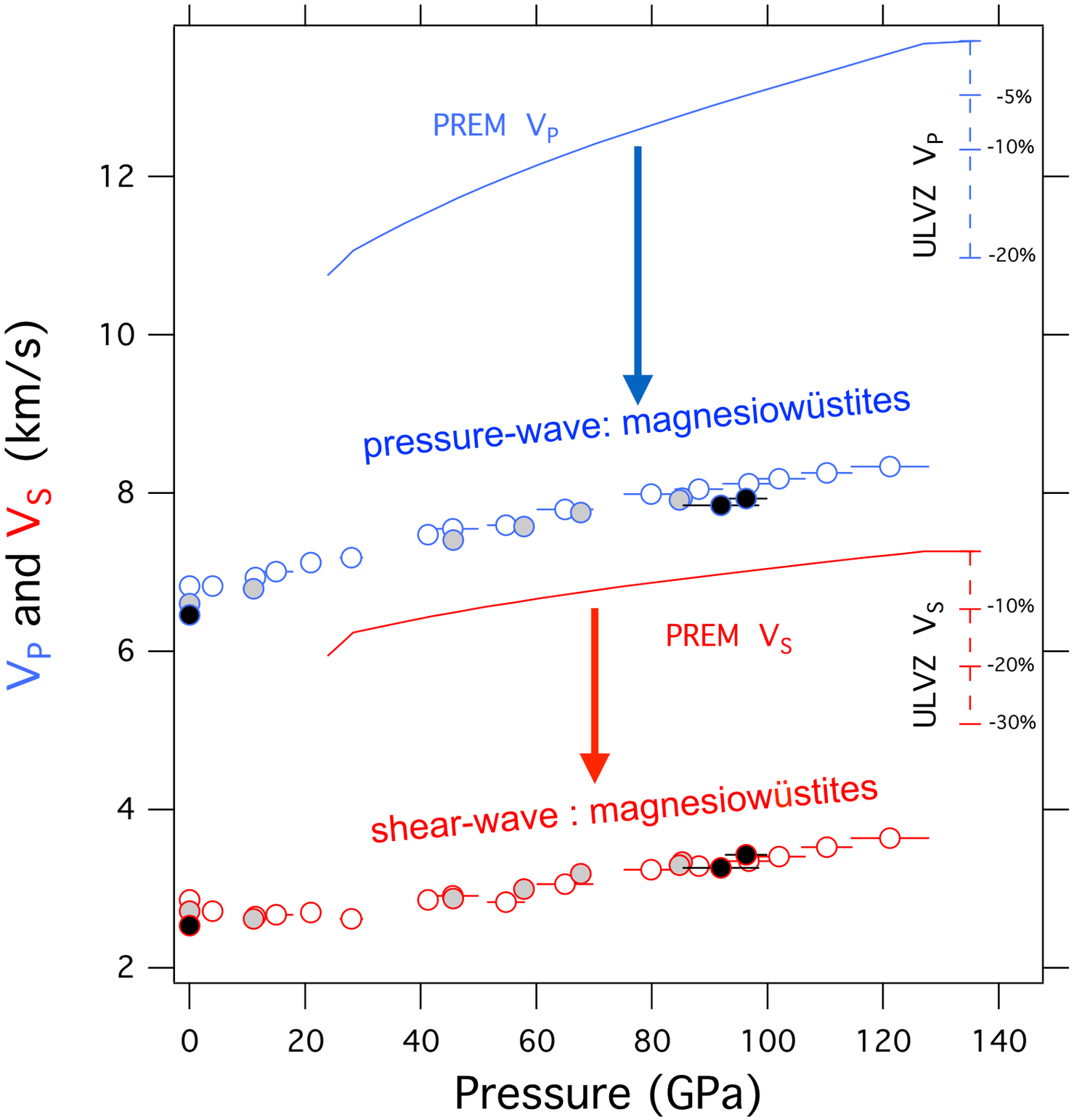
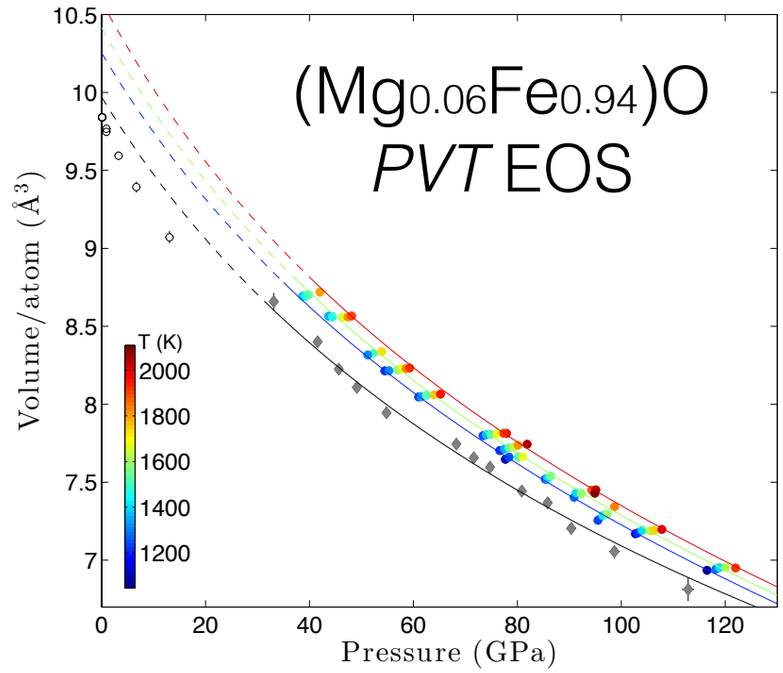


V_P and V_S



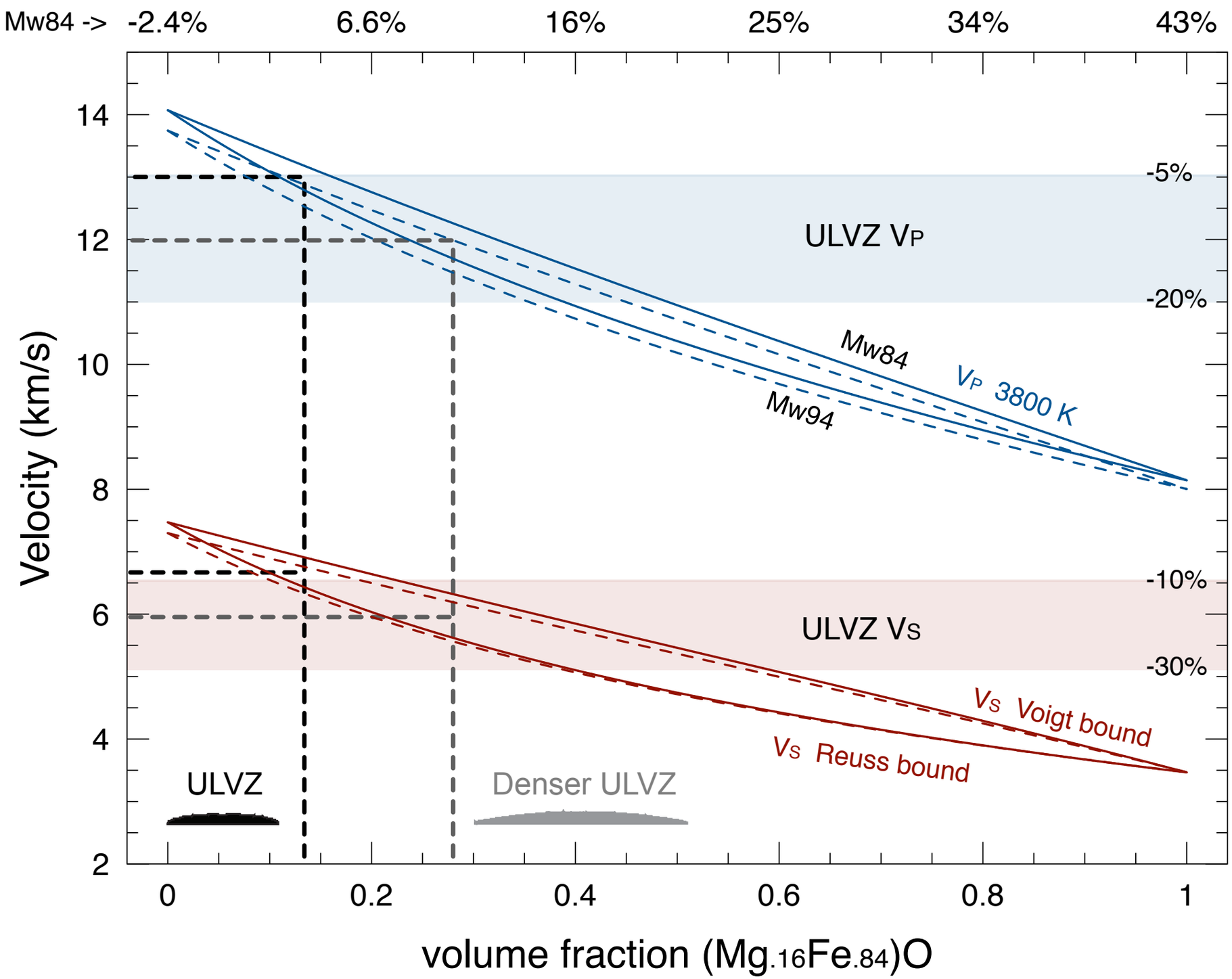
$$\frac{3}{V_D^3} = \frac{1}{V_P^3} + \frac{2}{V_S^3}$$

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

Self consistent model for ultralow-velocity zones:

Presence of magnesiowüstites

Δρ/ρ

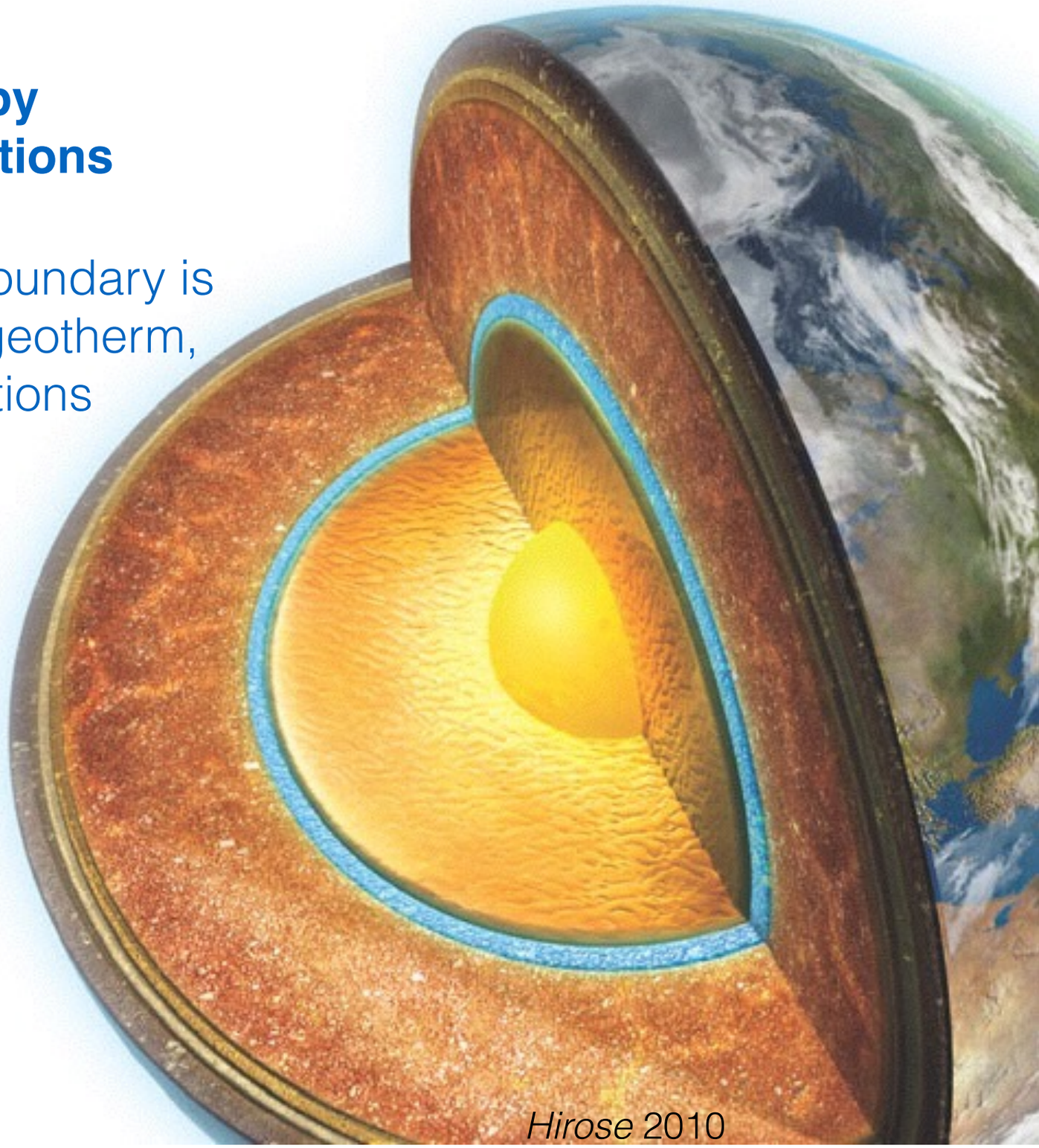


K_D: Tange et al. JGR (2009), $dV_{P,S}/dT$: Wentzcovitch et al. (2010)
EOS: Wicks et al. PEPI (2015), Wicks et al. (2016, under revision)

Constraining the temperature of Earth's core-mantle boundary

Phase stability dictated by pressure-temperature conditions

The temperature of the core-mantle boundary is determined from a plausible mantle geotherm, anchored by major phase transitions



T_{CMB} remains poorly constrained: 3400 to 4400 degrees K

Motivated to develop a technique to better constrain the thermal profile of the deep Earth

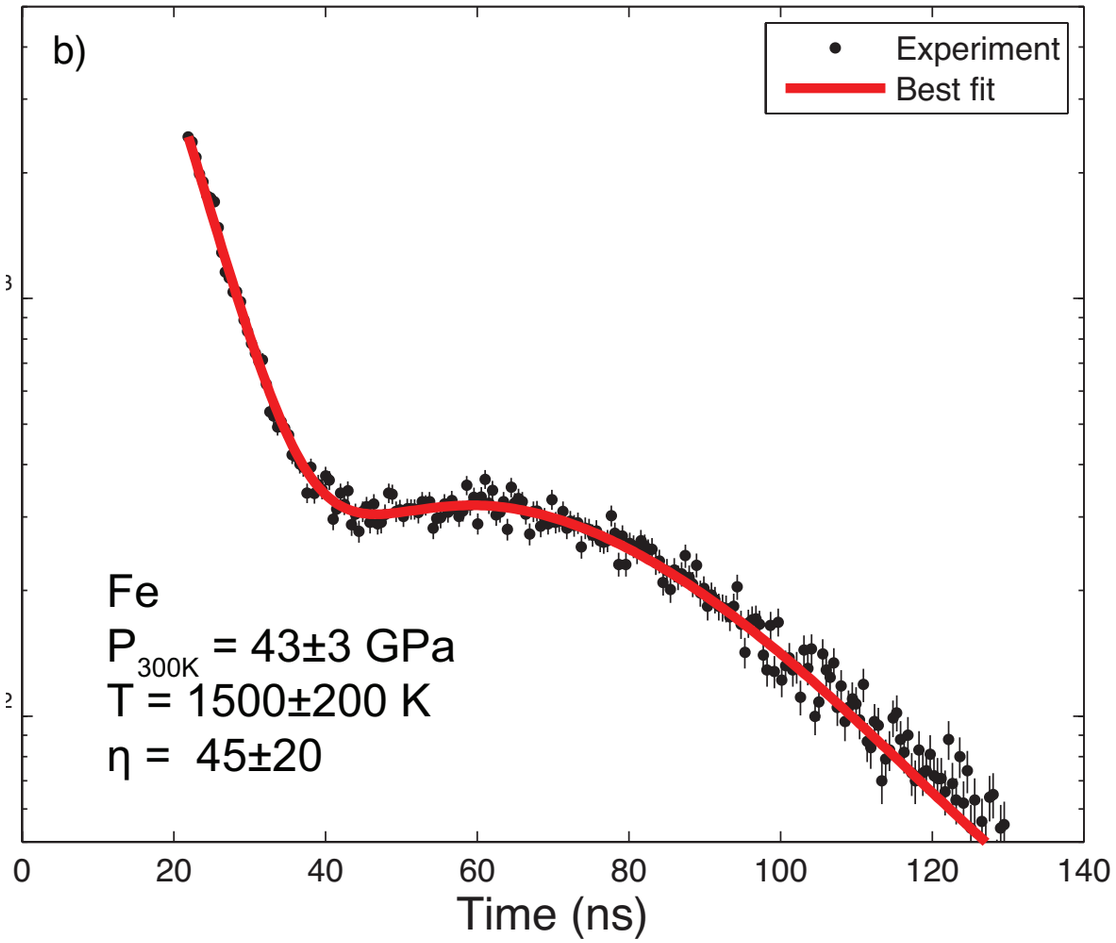
Detection of the solid – liquid boundary using synchrotron Mössbauer spectroscopy

$$f_{LM} = e^{-k^2\langle u^2\rangle} \Rightarrow 0$$

- When melting occurs: $f_{LM} = 0$
- Sensitive to the dynamics of the atoms
- Distinguishes a liquid from an amorphous state
- Near zero background
- Demonstrated for ^{119}Sn at ambient pressure using conventional Mössbauer spectroscopy
  - Boyle *et al. Proc. Phys. Soc.* 1960

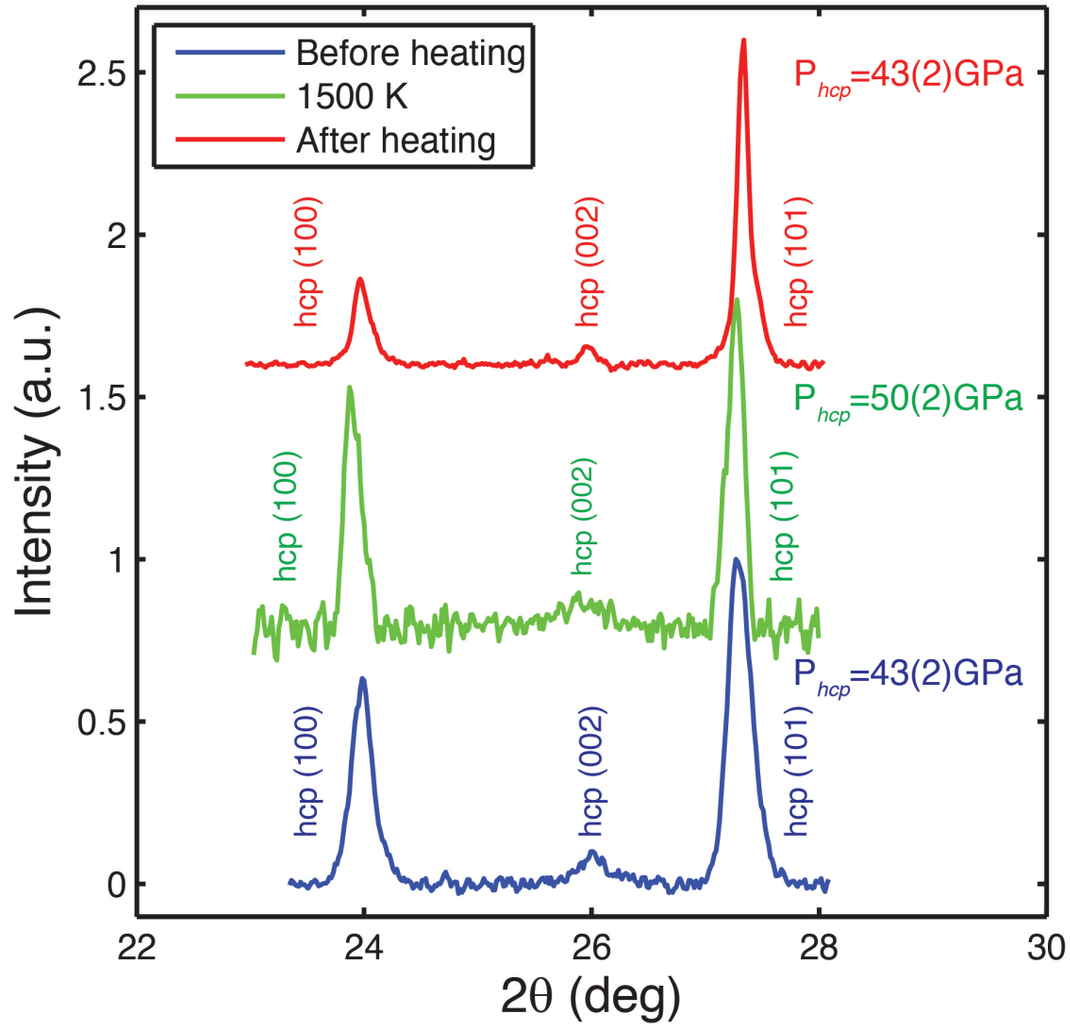
Mössbauer *Z. Phys.* 1958
Singwi & Sjölander *Phys. Rev.* 1960
Jackson *et al. EPSL* 2013

Synchrotron Mössbauer spectroscopy: iron-alloy melting measurements



Initial effective thickness of fcc-$Fe_{0.9}Ni_{0.1}$ constrained from high-PT

synchrotron Mössbauer spectroscopy

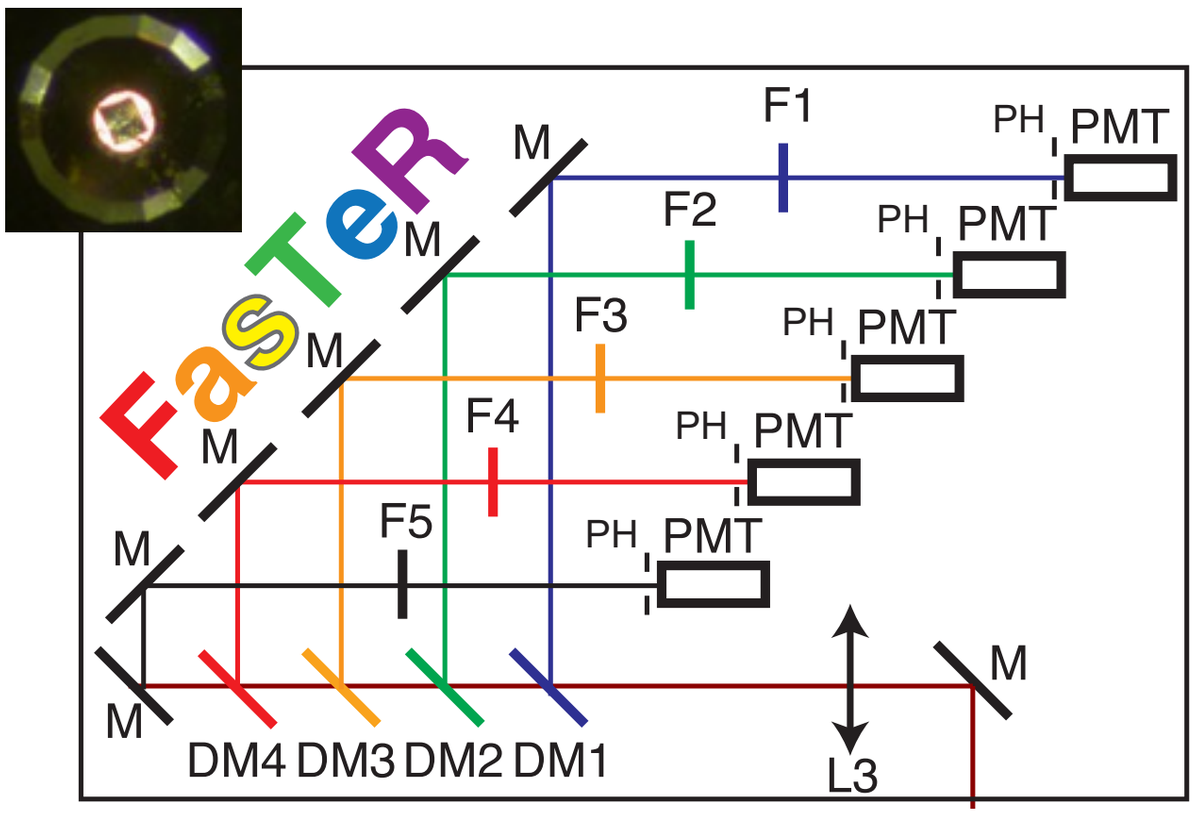


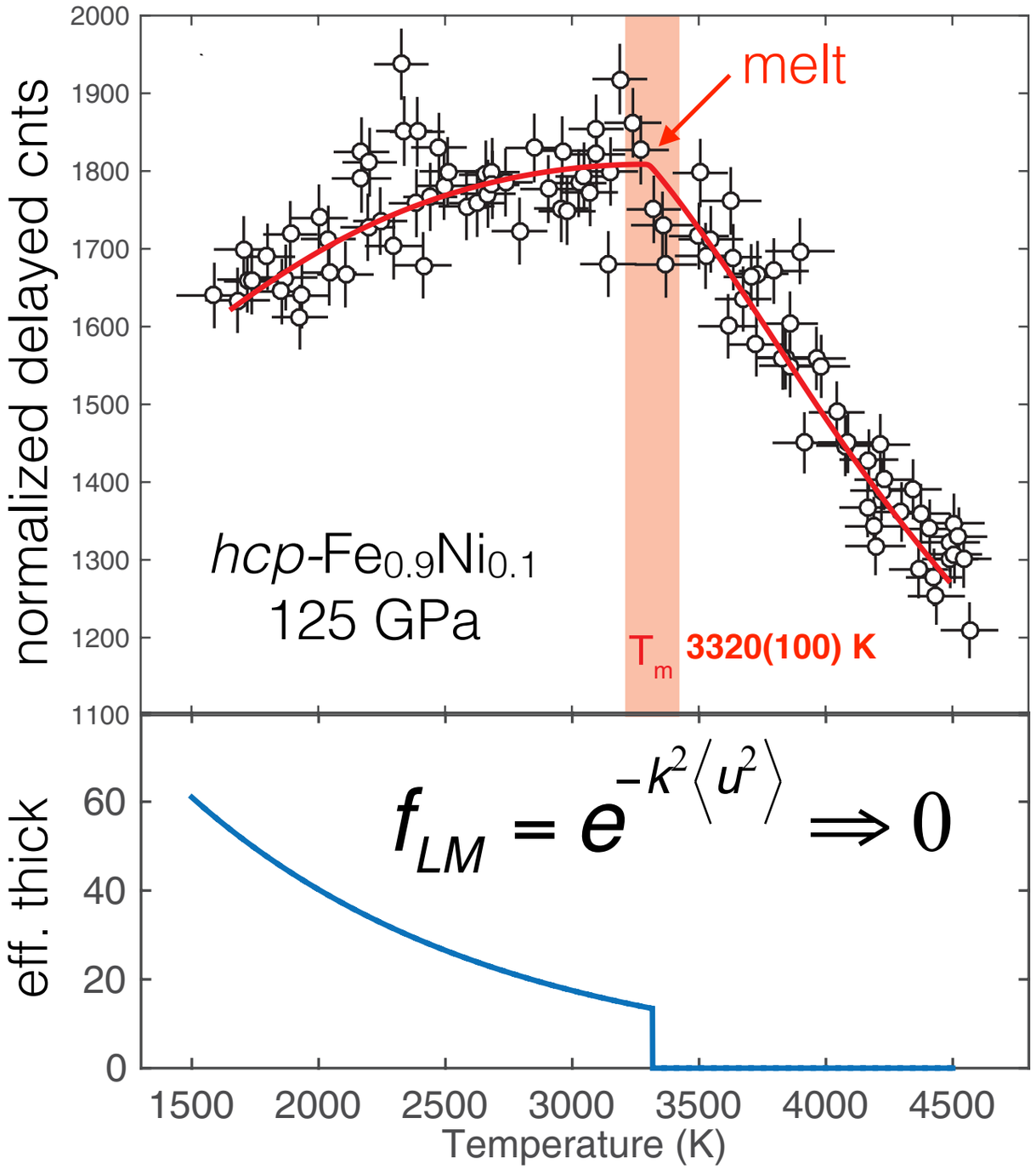
Thermal pressure of sample constrained from established equations of state and high-PT

x-ray diffraction

Synchrotron Mössbauer spectroscopy: melting measurements

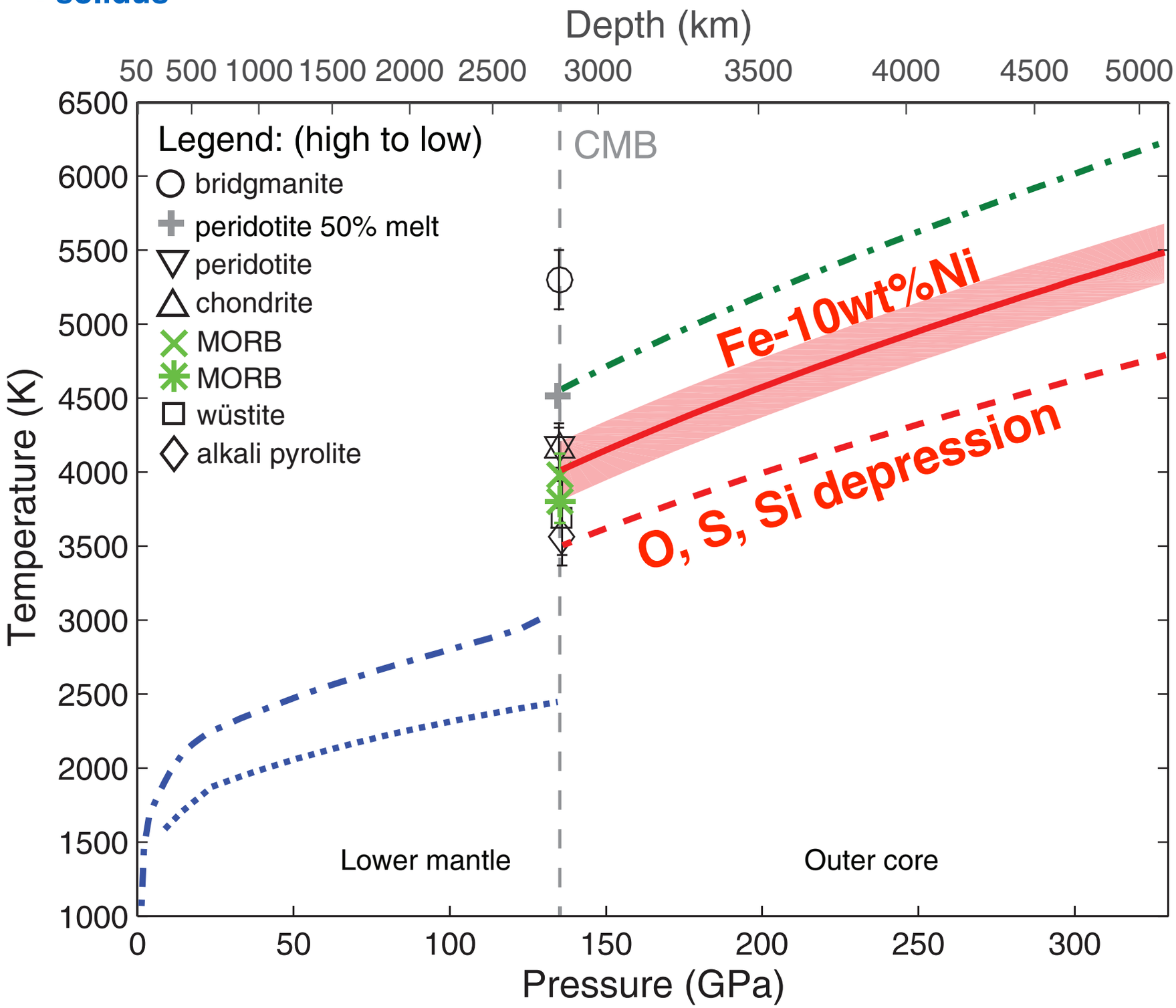
- Signal originates only from iron-bearing sample (no background)
- Monitoring atomic dynamics at high temperatures (melting) with high precision, using XRD to constrain thermal pressure
- Fast Temperature Readout system (FasTeR): microsecond readout, up to 400 Hz
- Stokes anti-Stokes temperature calibration





Melting and phase relations of deep mantle materials

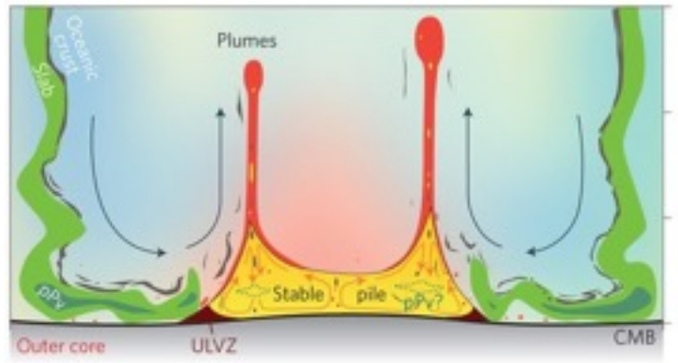
$T_{CMB} \sim T_{solidus}$



Zhang *et al. RSI* (2015), Zhang *et al. EPSL* (2016)

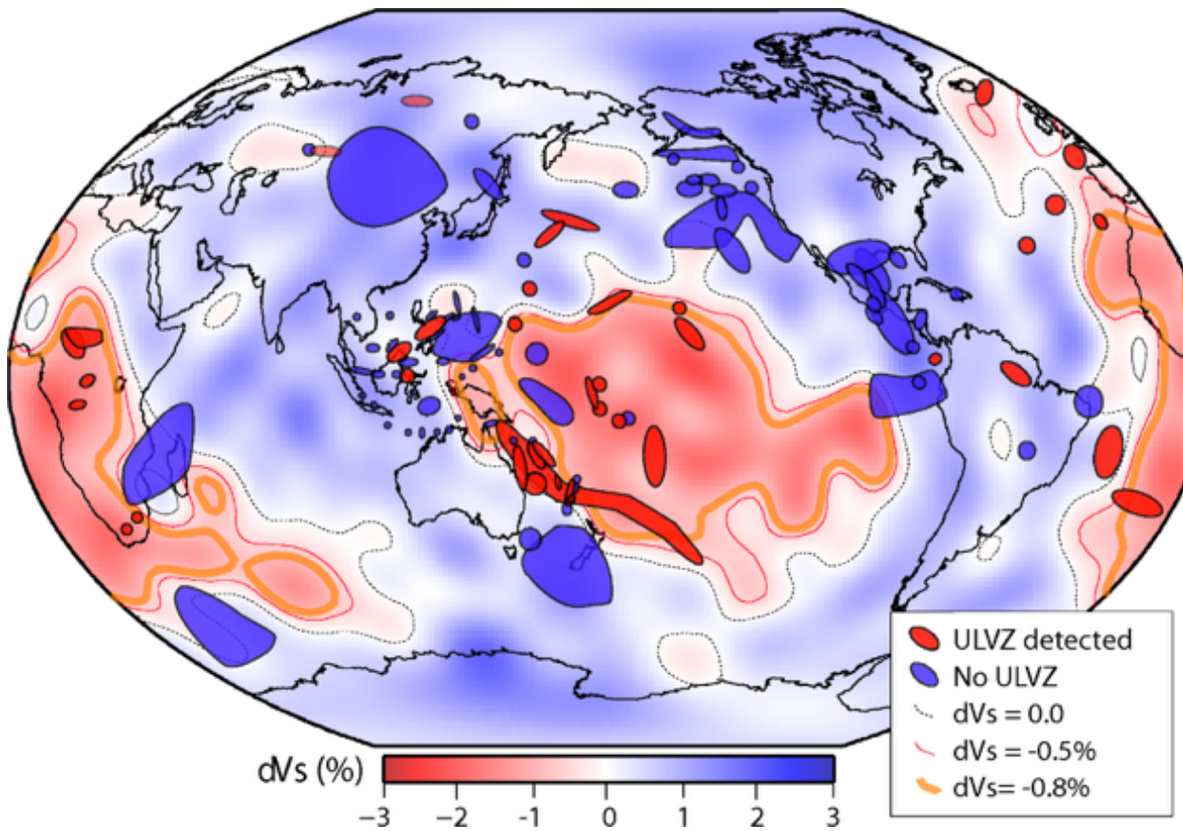
Concluding remarks

Nuclear resonant scattering techniques provide strong constraints on the vibrational thermodynamics (NRIXS), electronic environment (SMS), and melting (SMS) of Earth and planetary materials under extreme conditions.



ULVZs along the margins of thermochemical piles are likely compositional in origin.

- The **shear wave velocity** and density of magnesiowüstites permit a self-consistent model to explain these geophysically observed features.



(*McNamara et al. EPSL* 2010)

Further constraints on the **spin state** of candidate materials from SMS provides additional insights to the buoyancy of lower mantle assemblages.

The **melting curve** of $Fe_{0.9}Ni_{0.1}$ constrained by SMS, combined with plausible melting depressions, place CMB temperatures barely at or below the melting point of most deep Earth materials.

Thank you!