Dynamical properties investigated by (neutrons) and synchrotron X-rays: Applications in Earth and Planetary Science

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The plot

- Earth's and telluric planetary interiors: what do we know, how we know it

- Few (of the many) things we would like to know

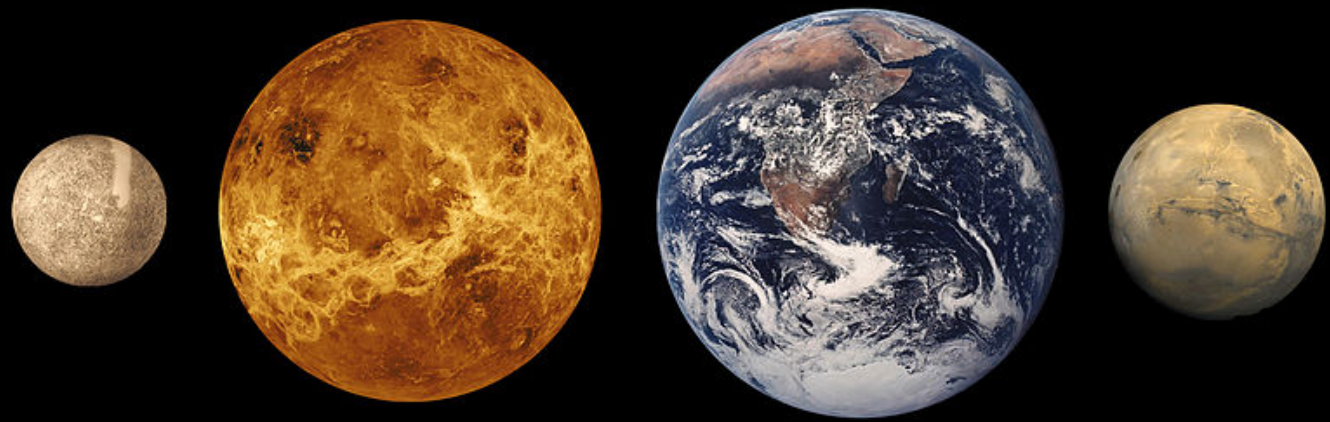
- Ongoing efforts in high-pressure high-temperature experimentation

  - Silicates and oxides → Lower mantle

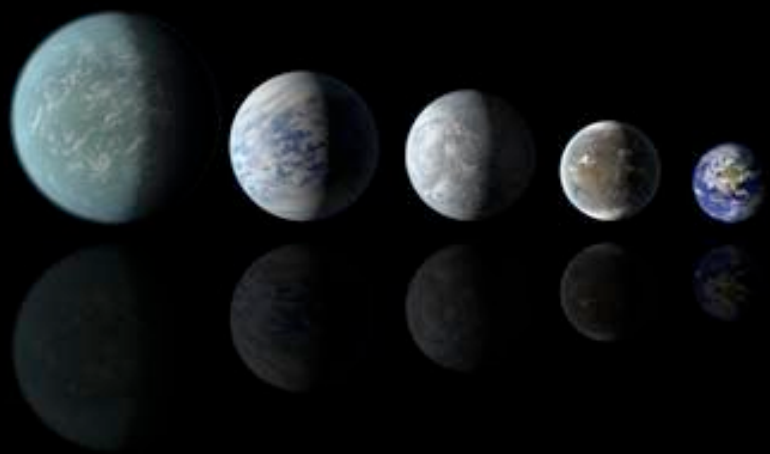
  - Iron and iron alloys → Core

The Earth: a telluric planet

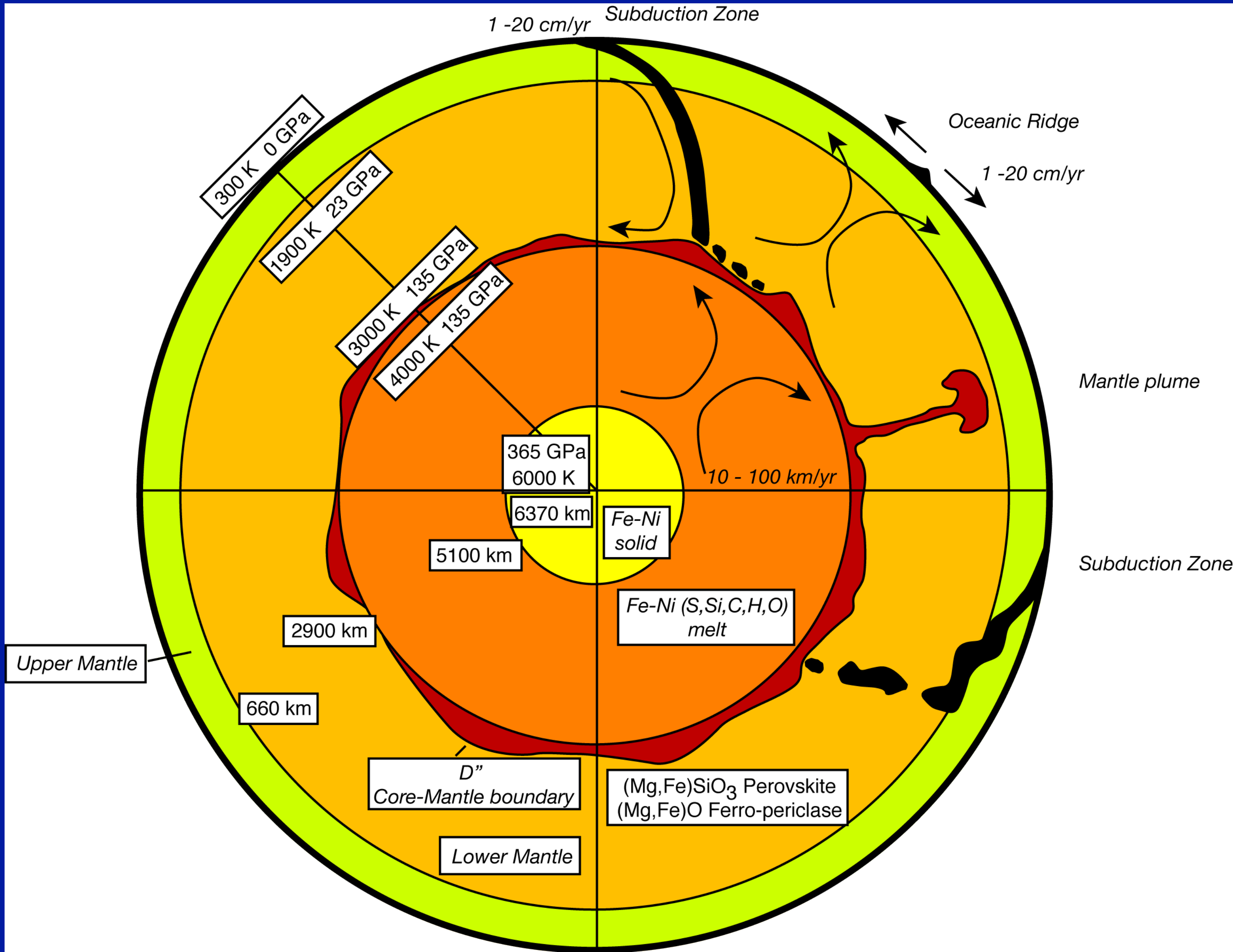
amongst others in the Solar system…



amongst others in the Universe…



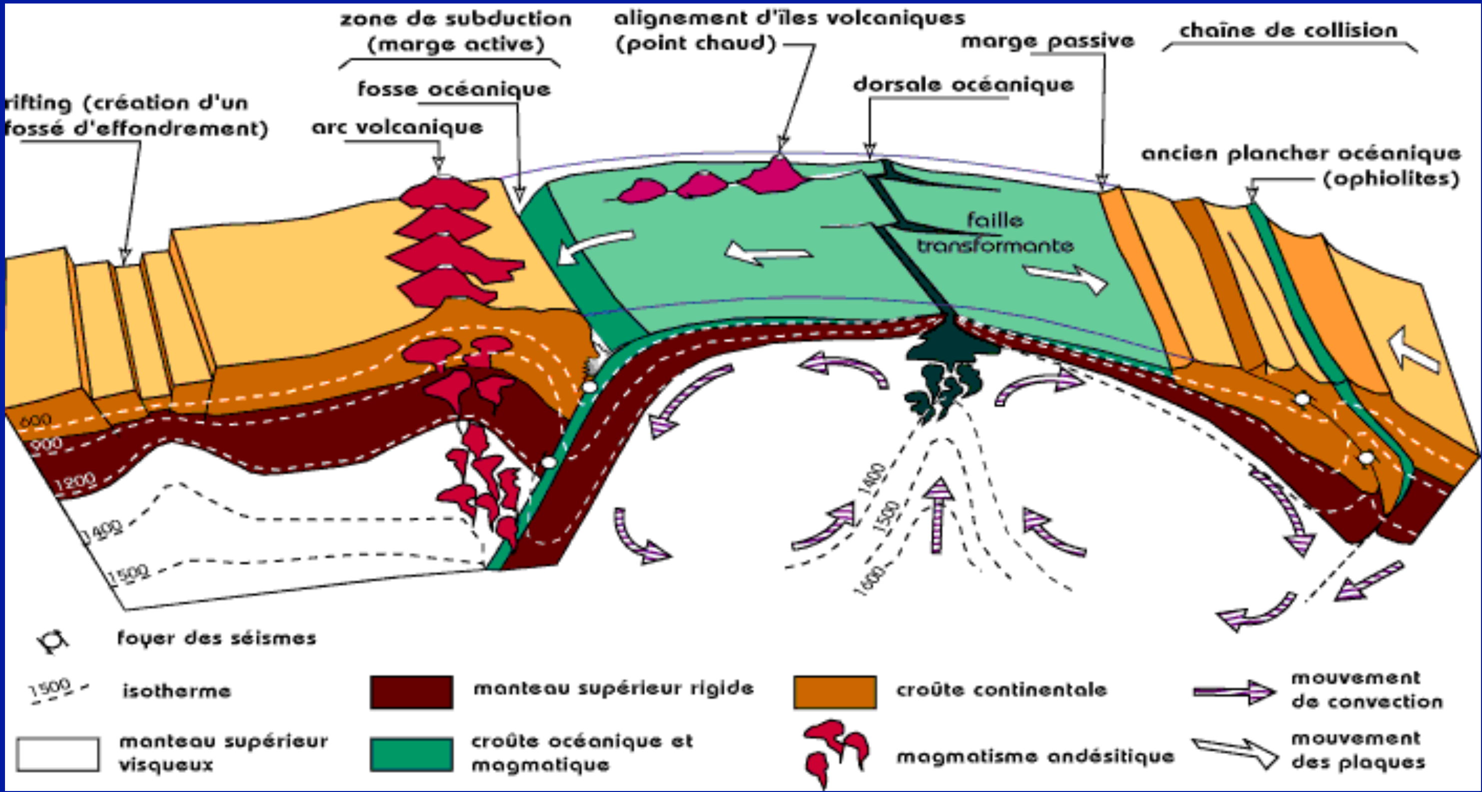
How does the Earth's interior look like?

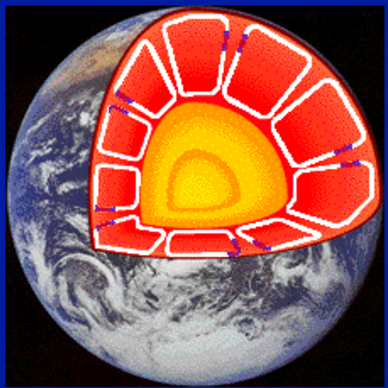


Can we drill?

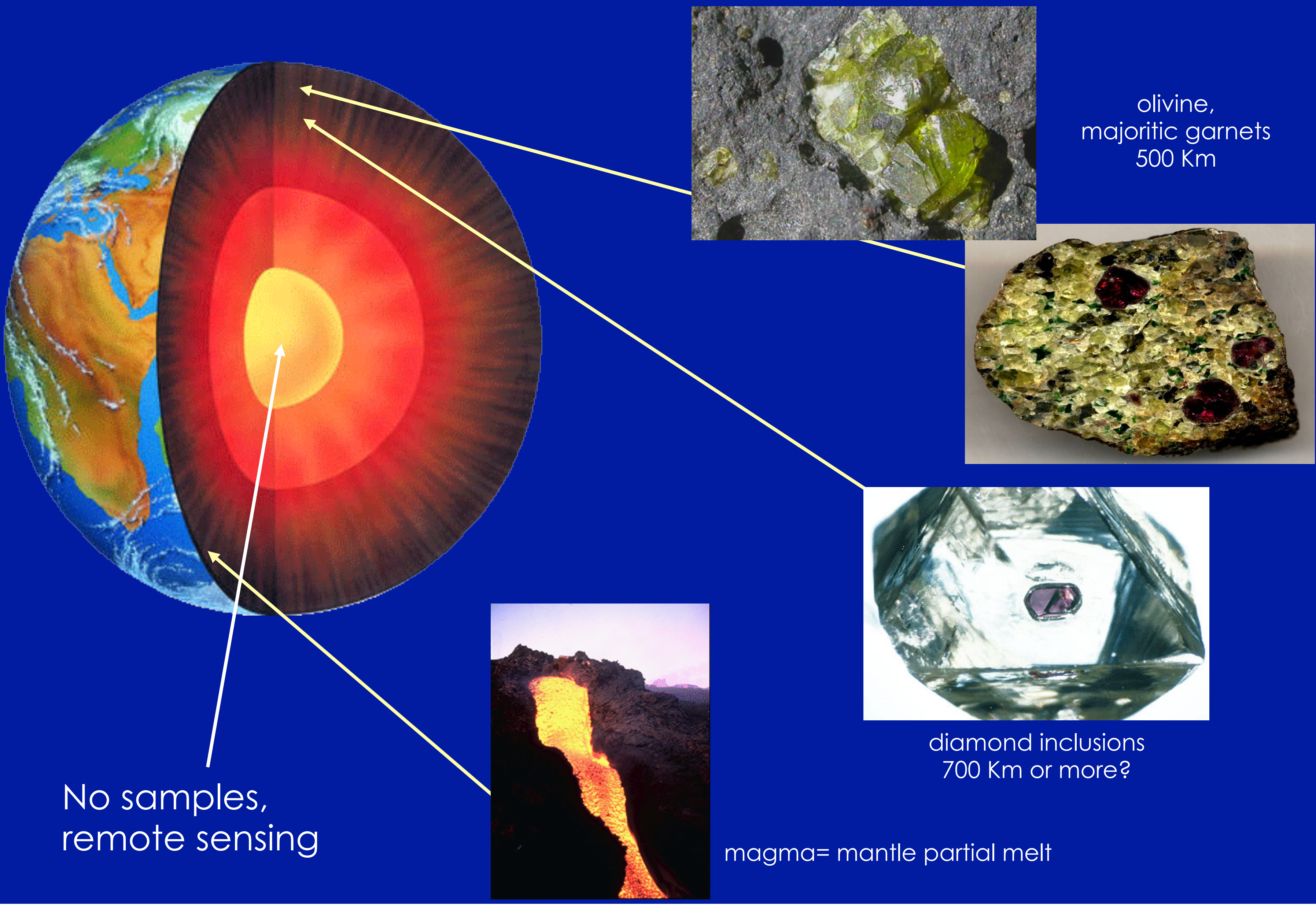
Deepest drilling S3G –Kola peninsula (Russia) – 12262 m

But things move: heat flow and convection

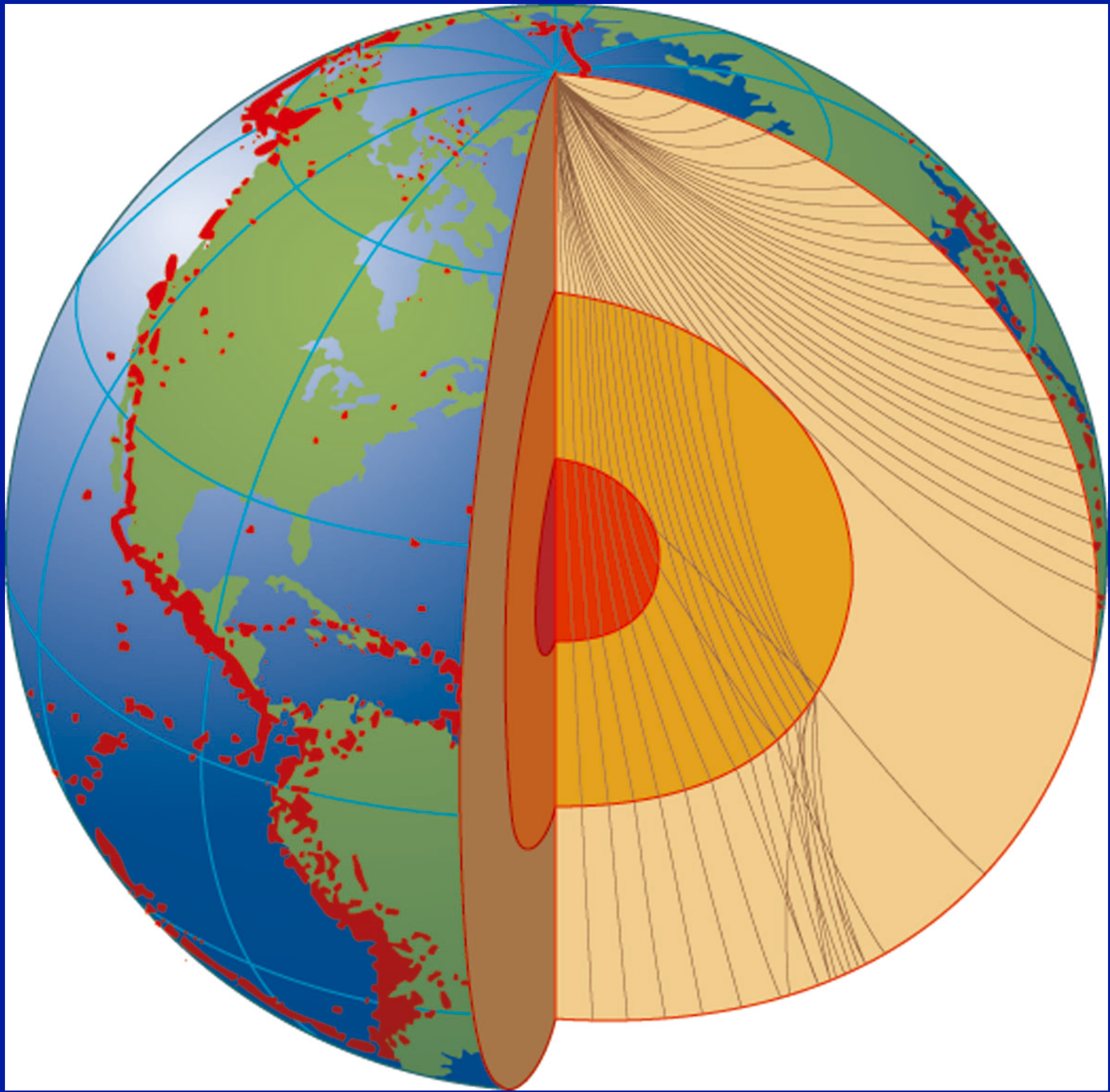




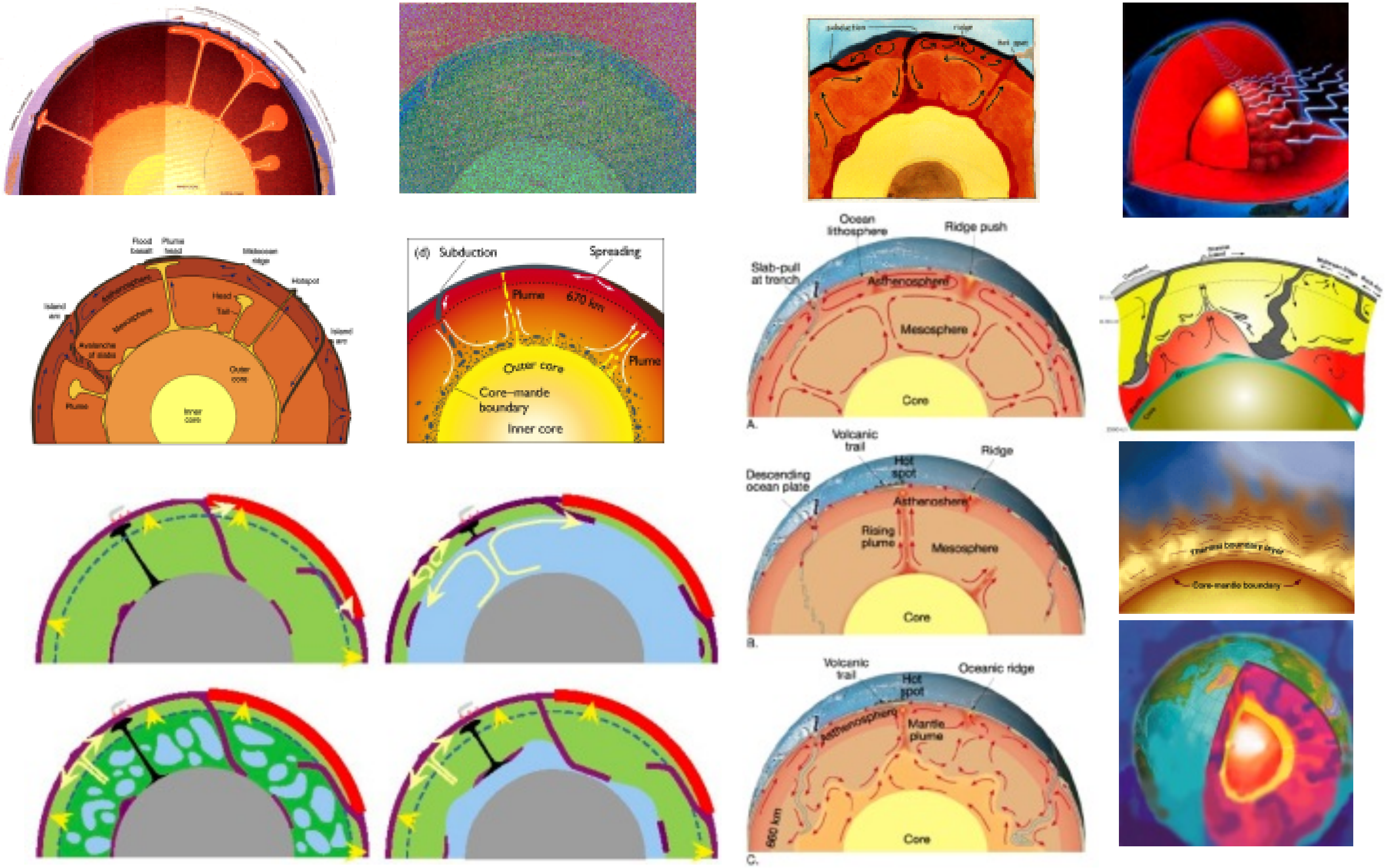
Some samples from the mantle



Seismic waves within the Earth
(and free oscillation)

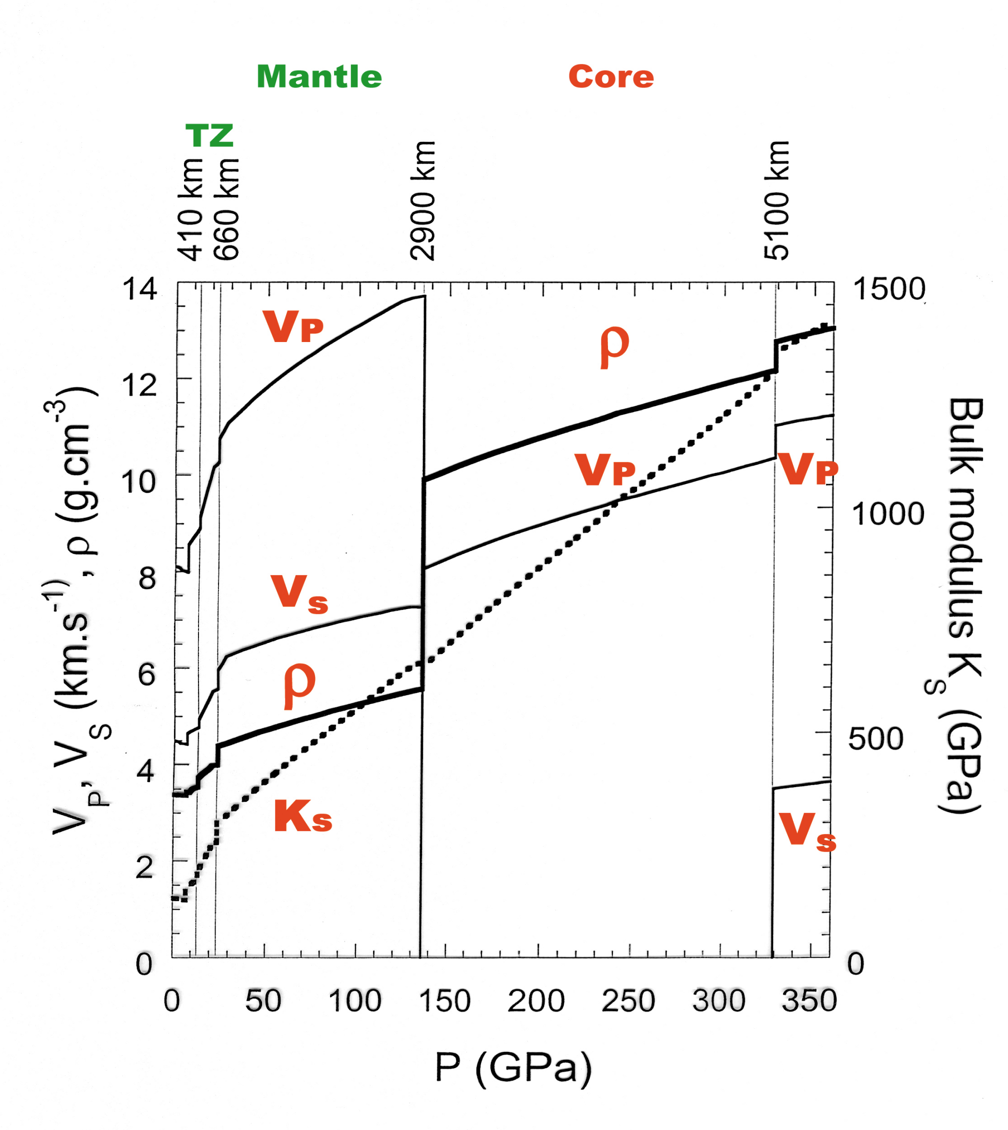


Imagining Earth's Interior



diagrams courtesy of E. Garnero

1-D seismic profiles ←→ Elasticity of geo-materials



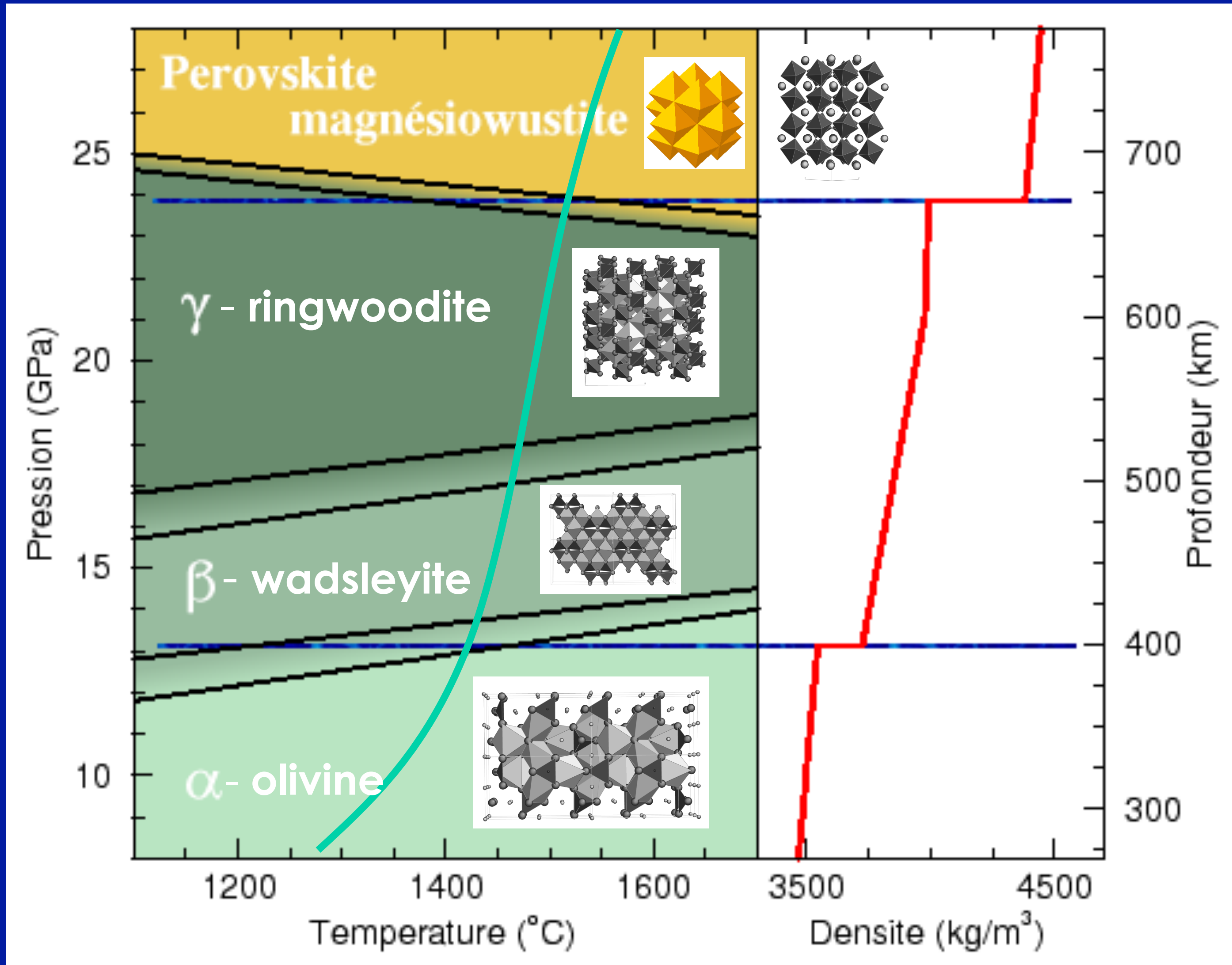
"What materials may have the elastic properties demonstrated by the seismic waves under the conditions of the interior?"

F. Birch, 1952

→ Inelastic x-ray scattering

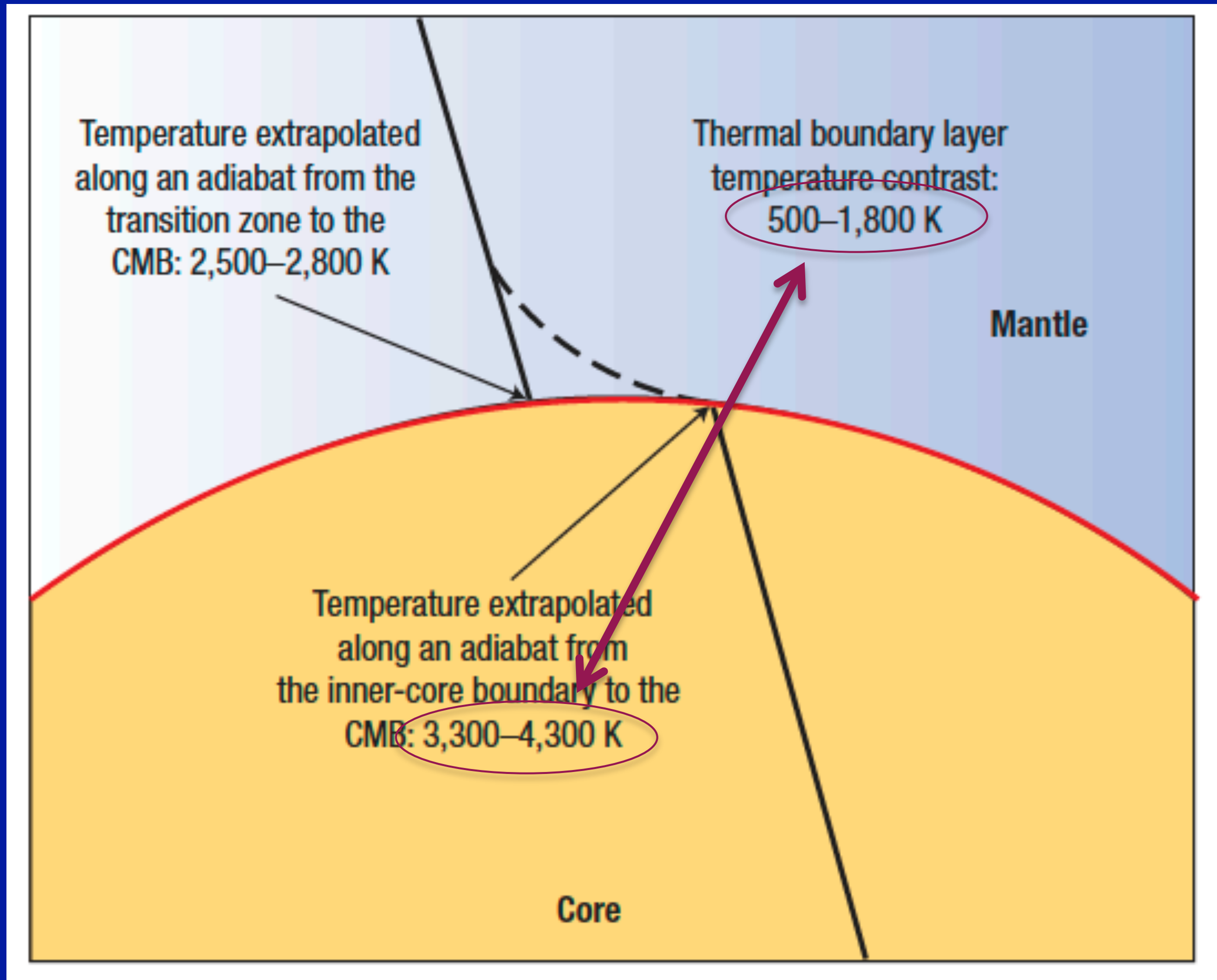
→ Nuclear resonant inelastic x-ray scattering

Phase transitions



$(Mg,Fe)_2SiO_4$-olivine phase diagram and seismic discontinuities

Temperatures at core-mantle boundary

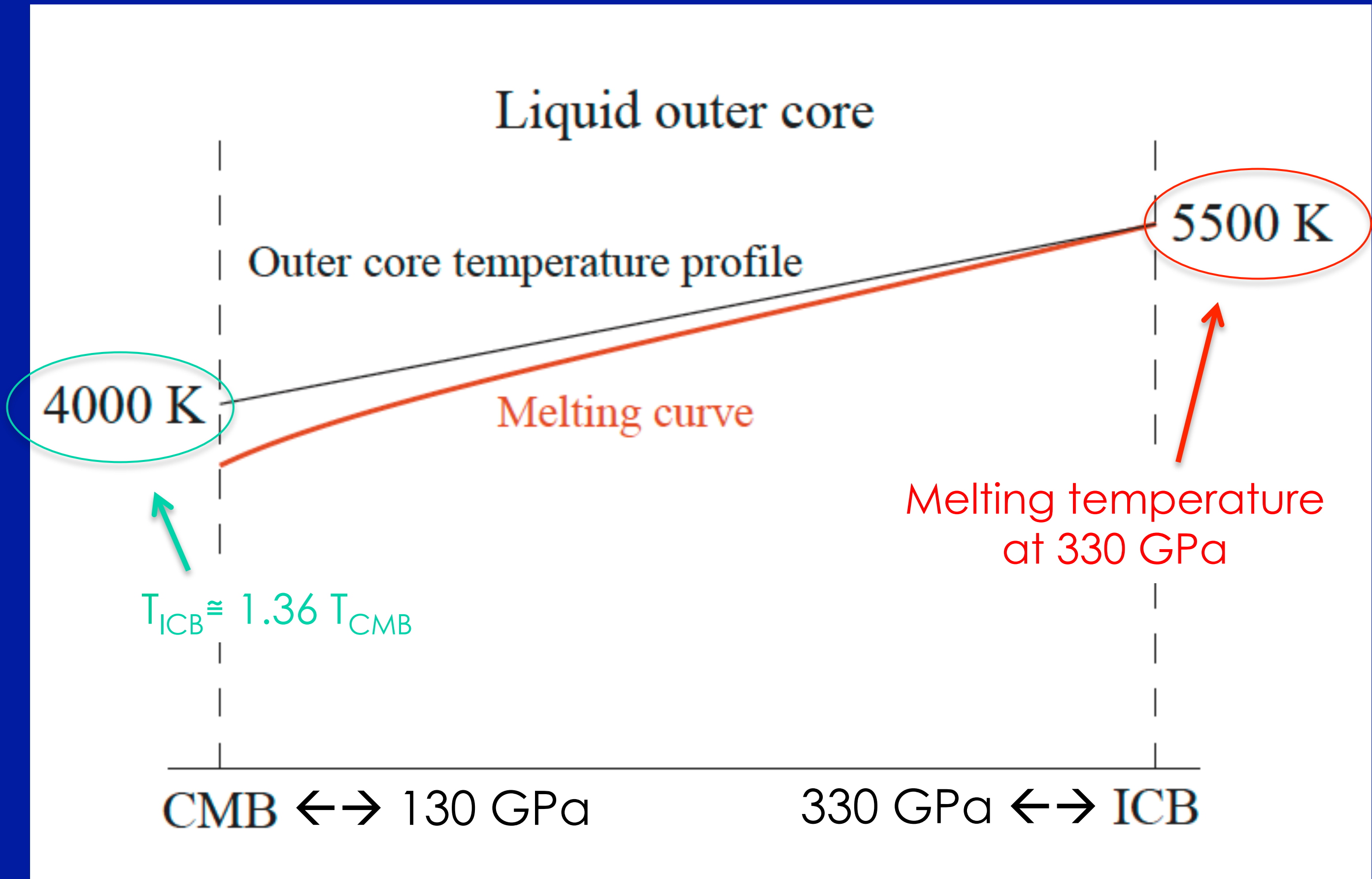


Lay et al., Nature Geoscience, 2008

Large uncertainties

→ geotherm, heat flux, thermal history, geodynamo, mantle convection

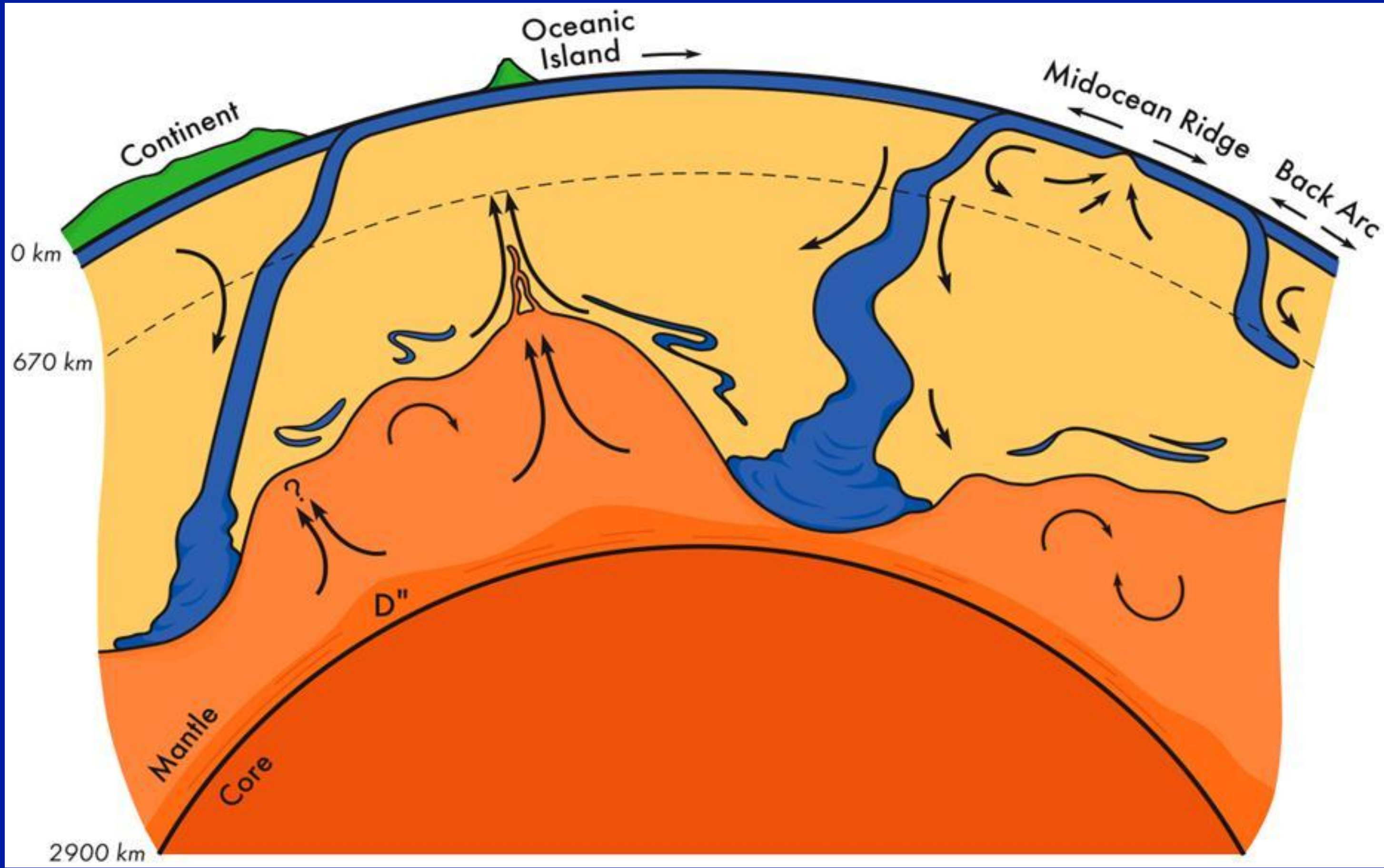
Melting point at inner core – outer core boundary as anchor point for geotherm



after Hernlund & Labrosse, GRL 2007

→ Nuclear resonant inelastic x-ray scattering
→ Synchrotron Mössbauer spectroscopy

Lower mantle complex dynamics

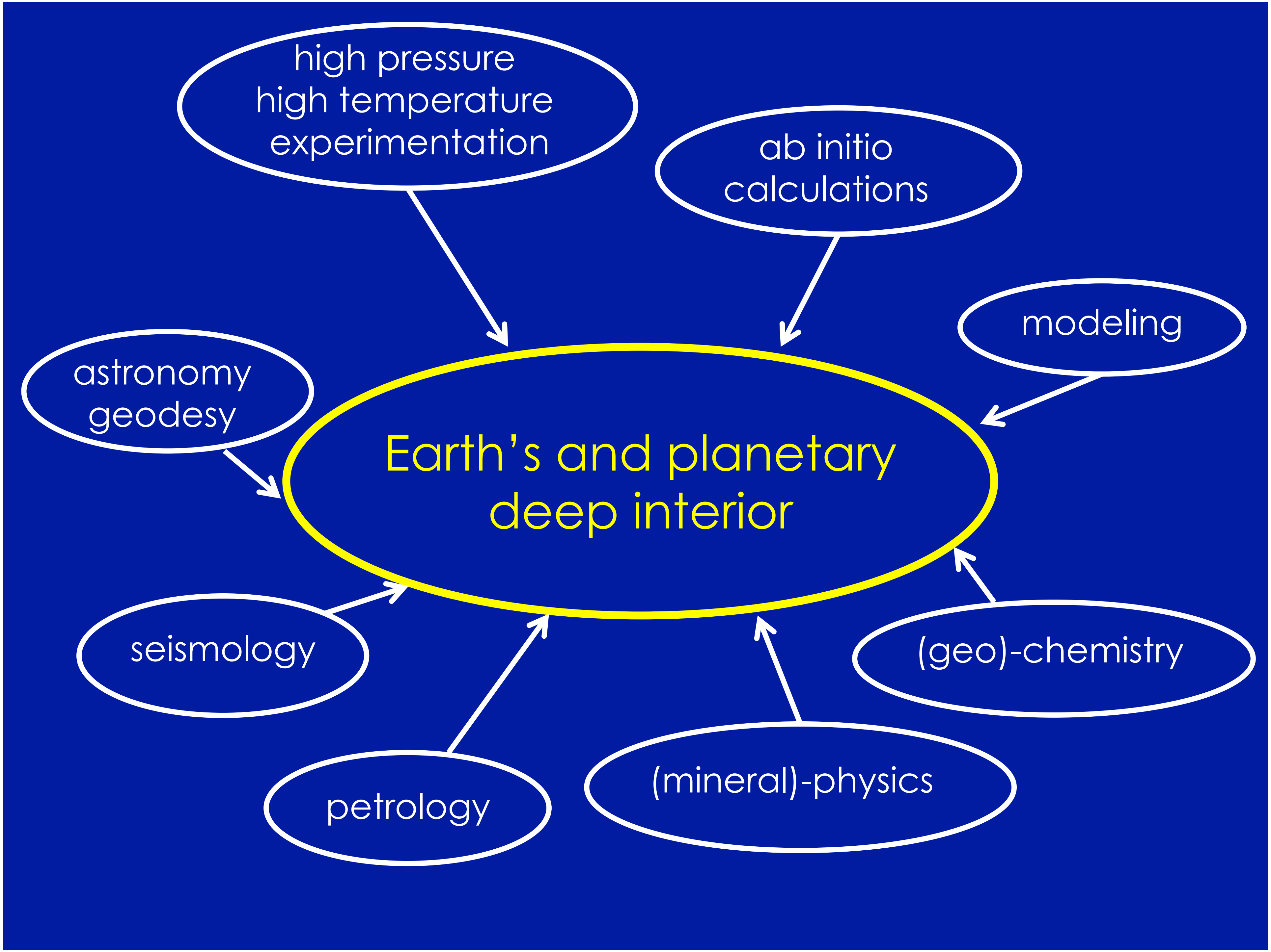


after Kellog, Science1999

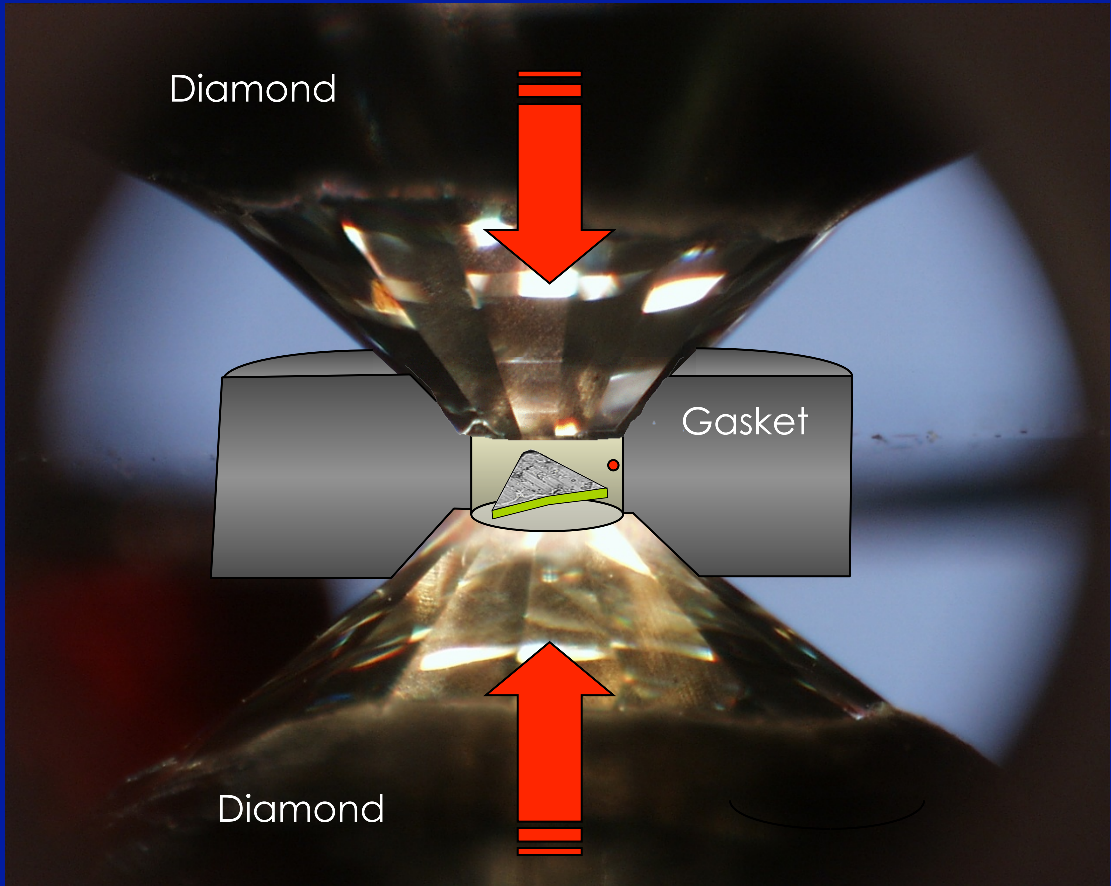
→ mineralogy & composition

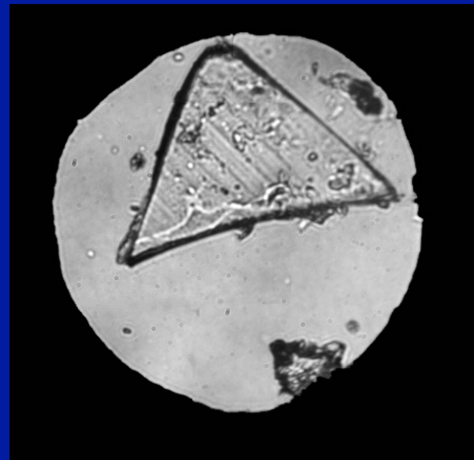
→ density & viscosity

→ heat transport mechanisms

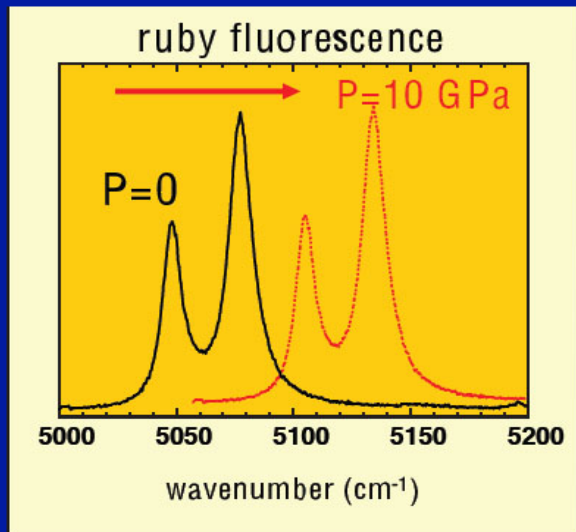

high pressure
high temperature
experimentation
ab initio
calculations
modeling
astronomy
geodesy
Earth's and planetary
deep interior
seismology
petrology
(mineral)-physics
(geo)-chemistry

Diamond anvil cell (DAC)

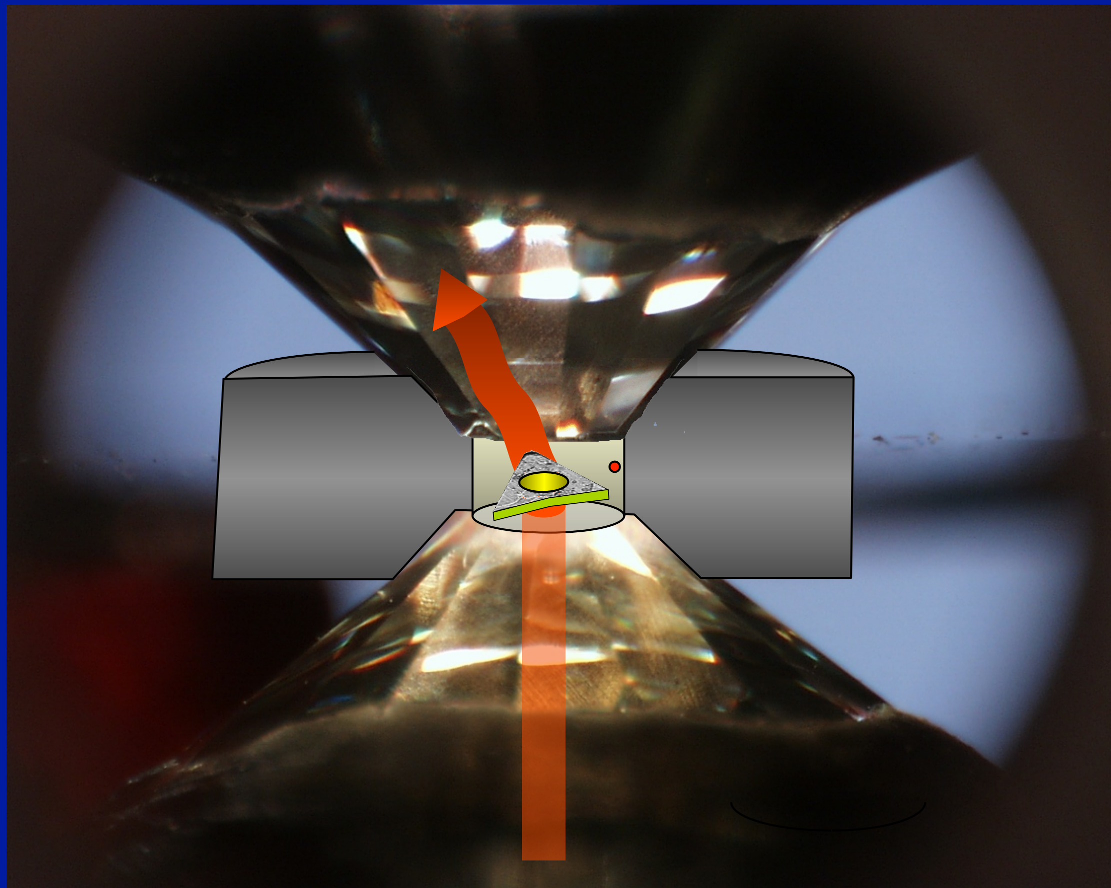


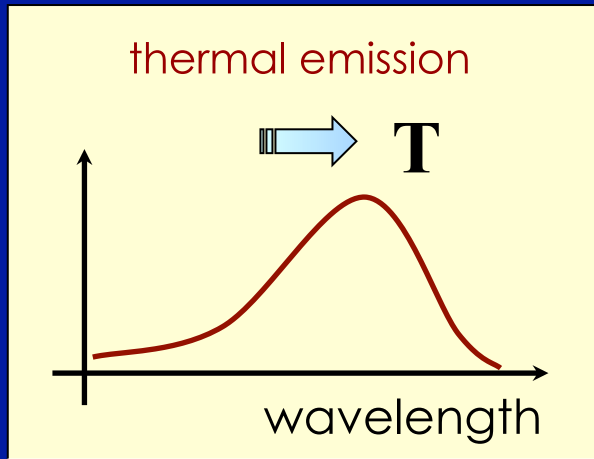


20 µm

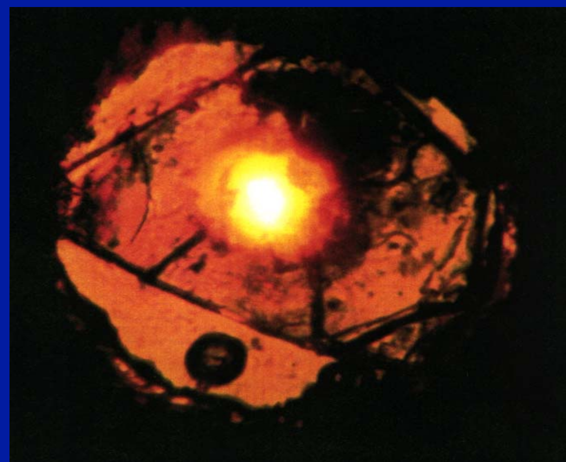


Diamond anvil cell (DAC)

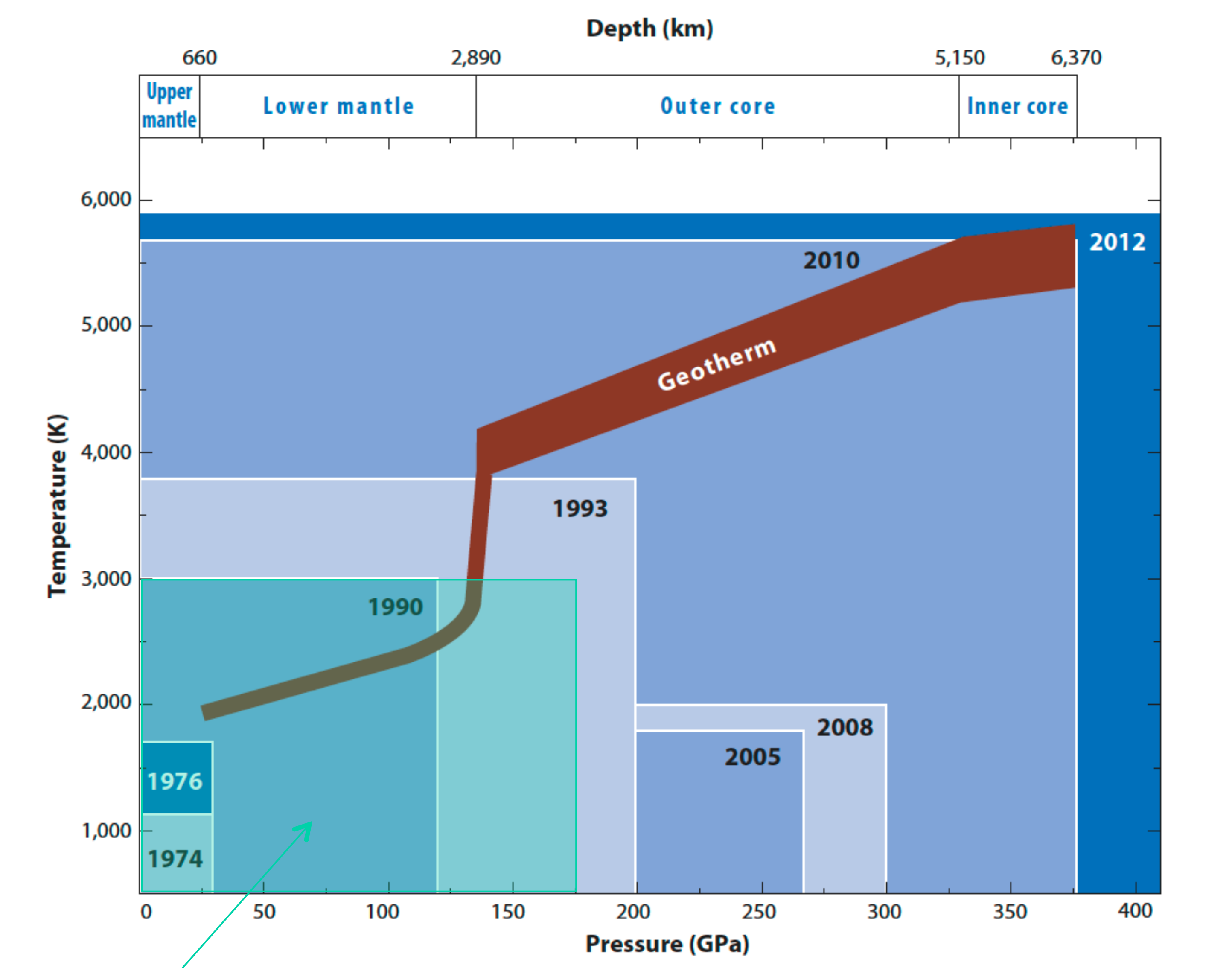




$$B_{\mathrm{Planck}}(\lambda, T) = \frac{2hc^2}{\lambda^5} \frac{1}{e^{hc/\lambda kT} - 1}$$



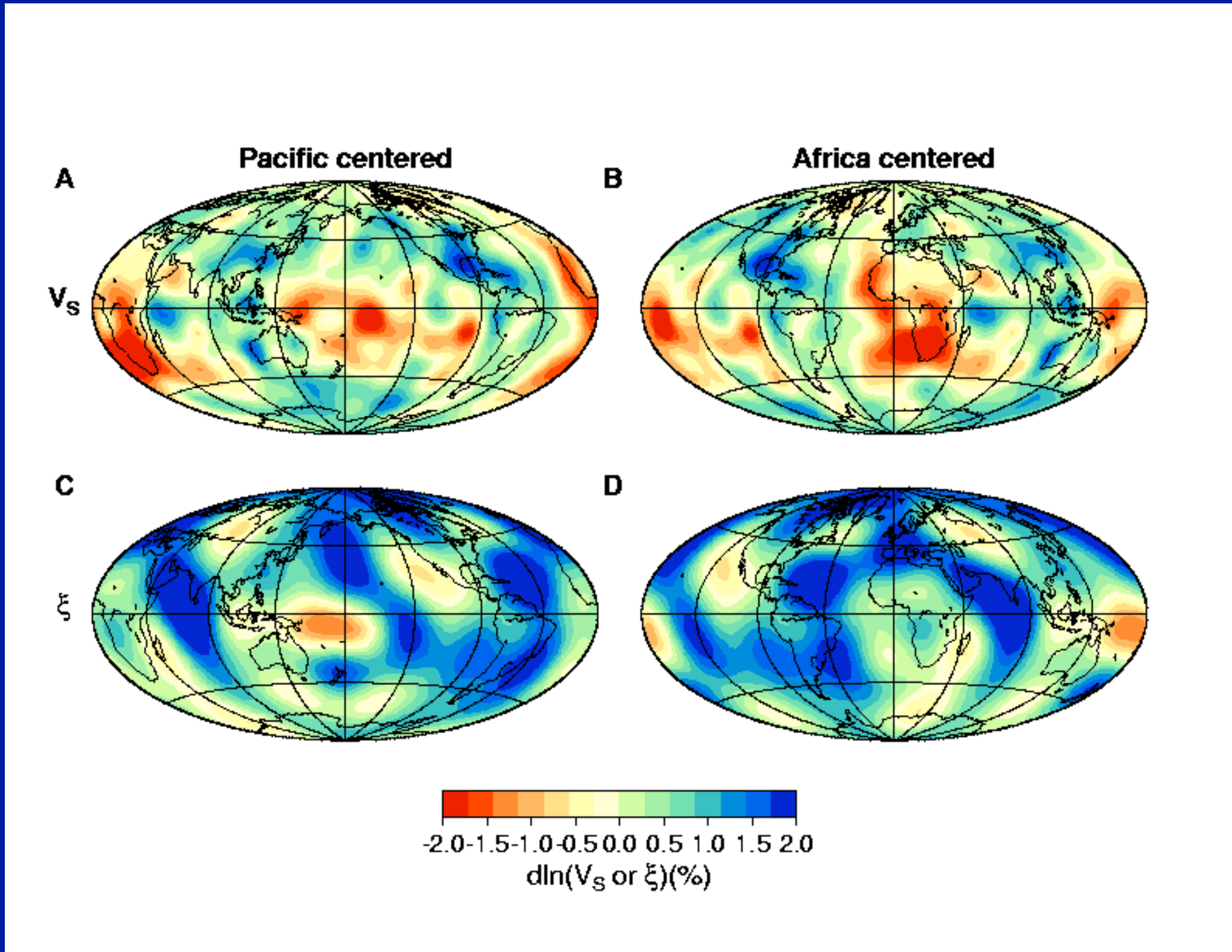
Progress in P-T range by laser-heated DAC



after Hirose et al., Annu. Rev. Earth Planet. Sci., 2013

Techniques other than x-ray diffraction

Lower mantle heterogeneities



Panning and Romanowicz, Science 2004

lateral variations

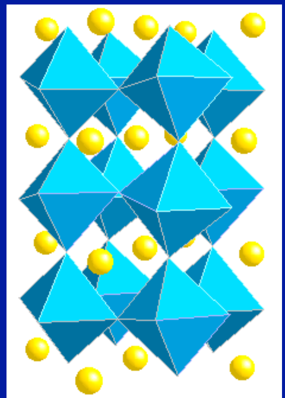
shear anisotropy

→ Things look complex, source of complexity?

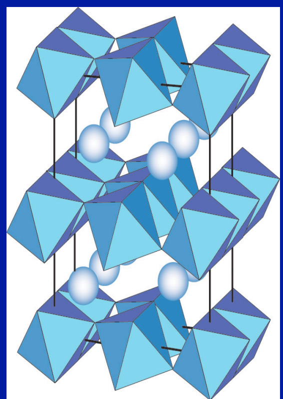
Lower mantle mineralogy
(23 GPa <P<135 GPa)

magnesium silicate perovskite

$(Mg_{1-x-z},Fe_x,Al_z)(Si_{1-y},Al_y)O_3$

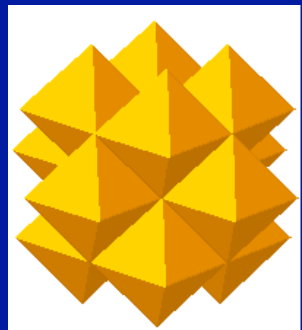


post-perovskite



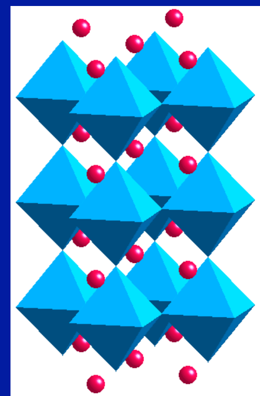
ferripericlase

$(Mg_x,Fe_{1-x})O$



calcium silicate perovskite

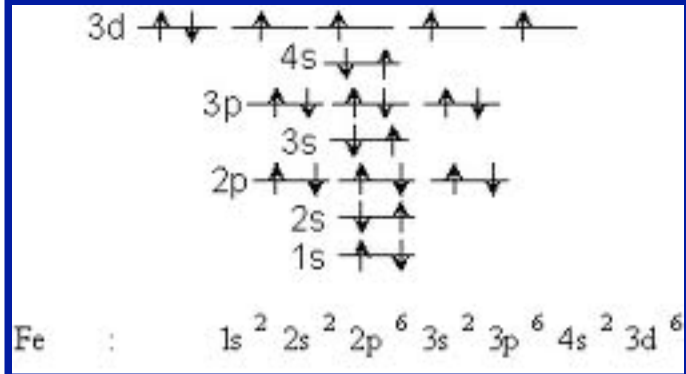
$CaSiO_3$



Electronic structure of iron

Isolated atom

Crystalline
Field

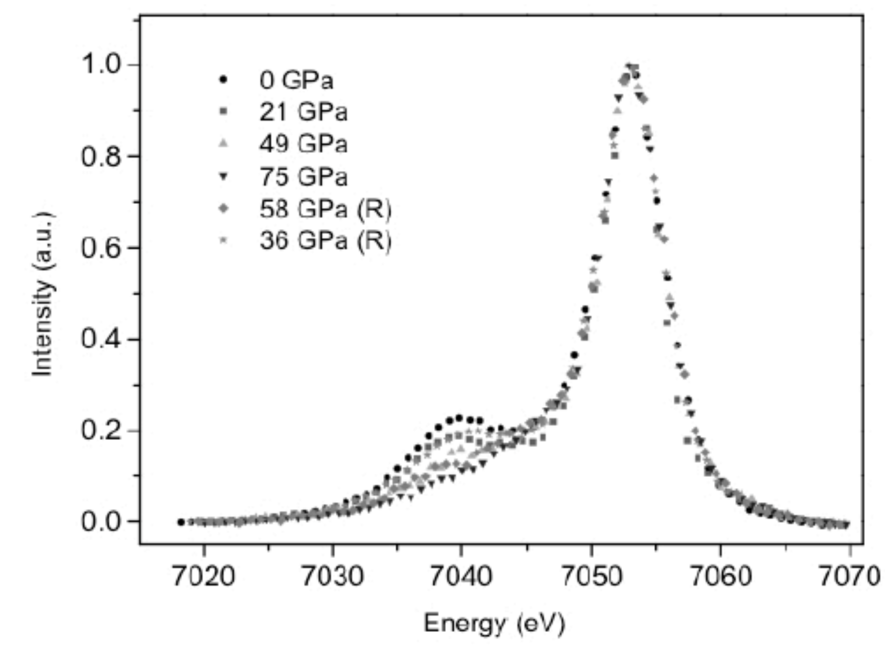


Pressure induced HS-LS transitions?

K_β emission spectroscopy on lower mantle minerals

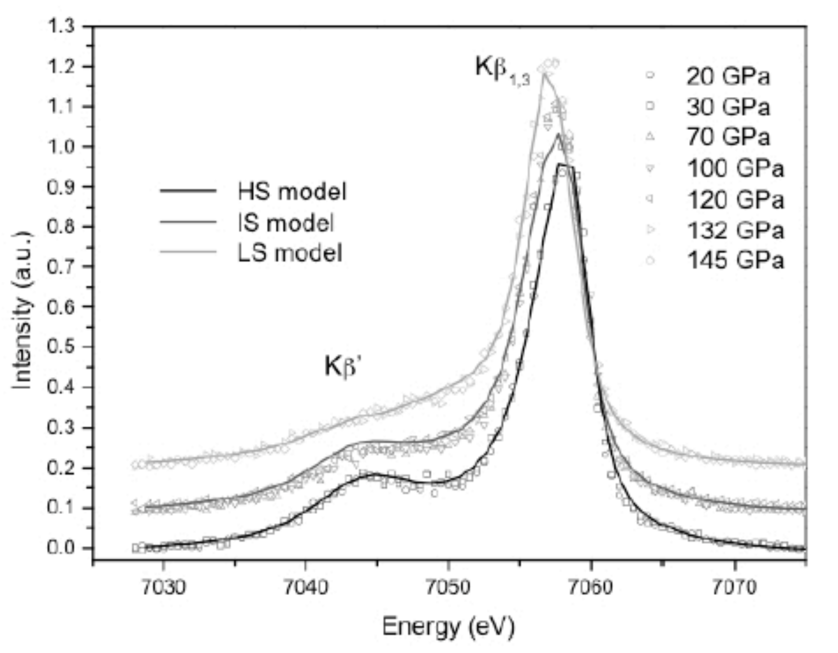
3d transition metals: the spectral shape of K_β emission line is dominated by the final state interaction between the 3p core hole and the partially filled 3d shell

(Mg,Fe)O-ferropericlase



Badro et al., Science 2003

(Mg,Fe)SiO_3-perovskite



Badro et al., Science 2004

→ pressure-induced spin pairing transition in lower mantle minerals

10 years of fine tuning
(iron content, valence state, chemical composition, high temperature)

Key techniques:

- K_β photemission spectroscopy

  → direct probe of the macroscopically averaged spin of specific atomic species (Fe)

- Synchrotron Mössbauer spectroscopy / Nuclear forward scattering

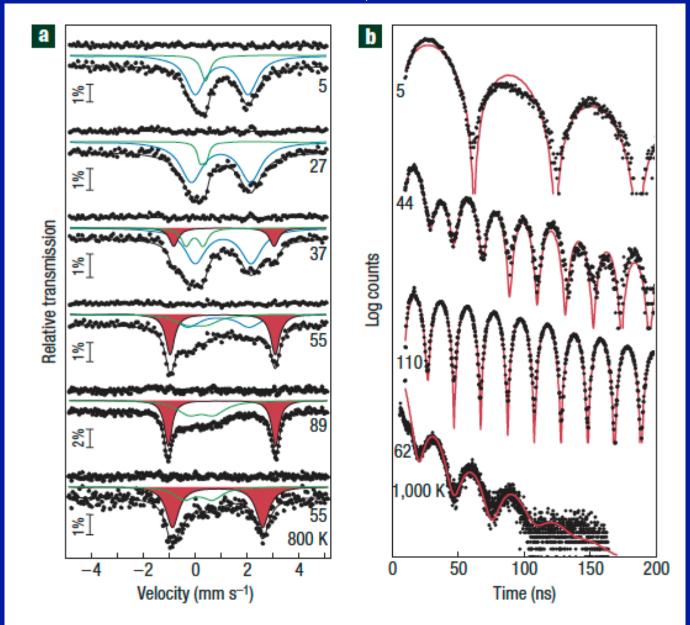
  → redox-selective (Fe^{2+} vs. Fe^{3+}) probe of magnetic state (time domain)

- Synchrotron Mössbauer source

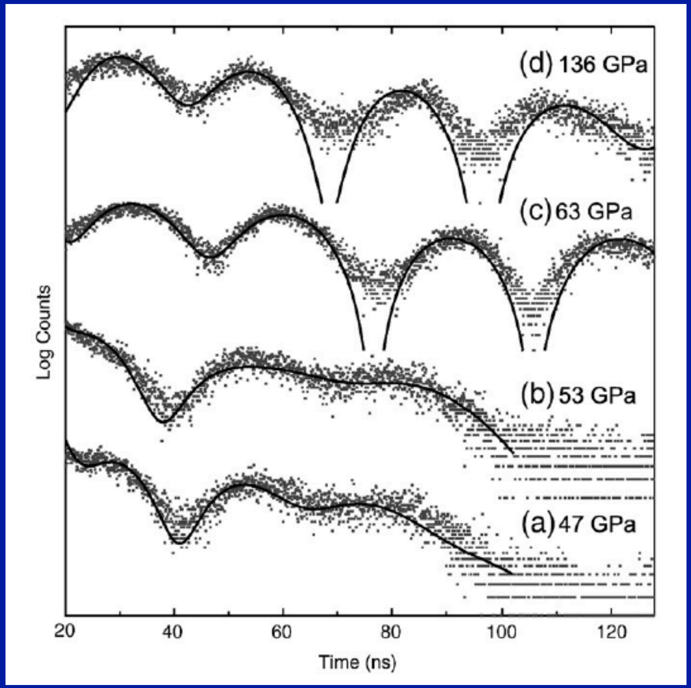
  → redox-selective (Fe^{3+} vs. Fe^{3+}) probe of magnetic state (energy domain)

Examples

$(Mg,Fe)SiO_2$-perovskite

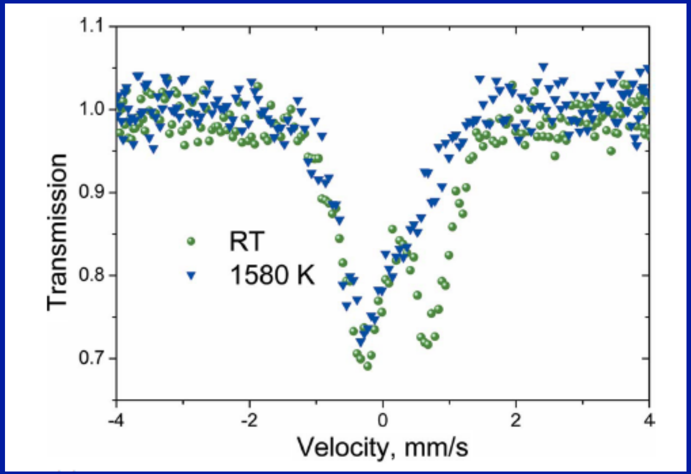


McCammon et al., Nature Geo sci. 2008



Catalli et al., Am. Min. 2010

(Mg,Fe)O-ferroericlase

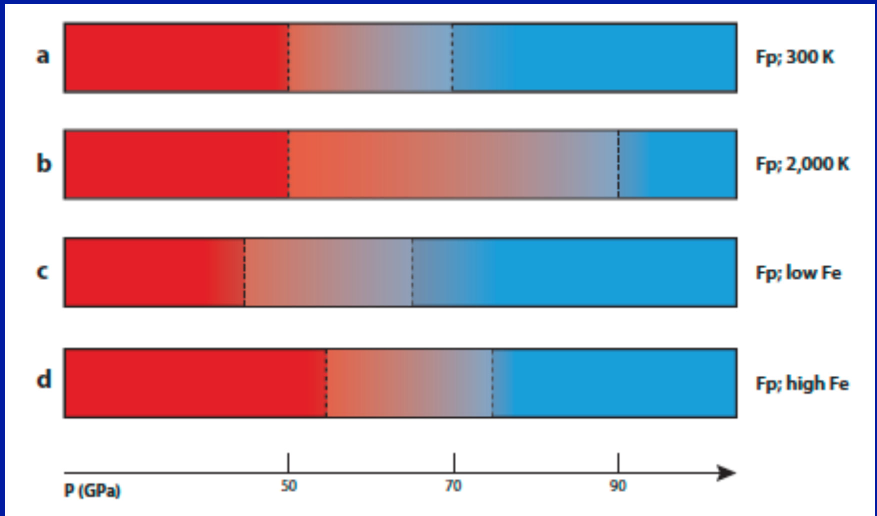


Potapkin et al., J. Synchrotron Rad. 2012

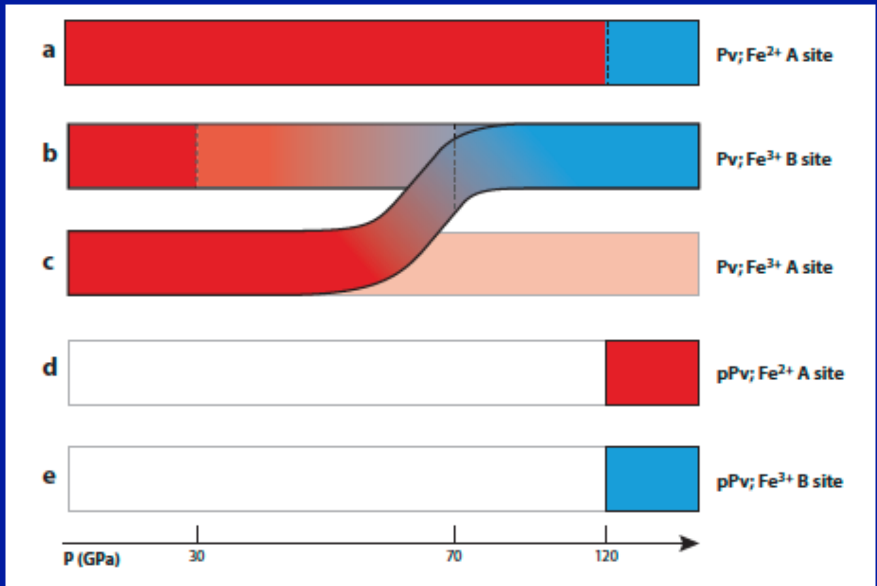
and today

Spin state evolution with pressure

(Mg,Fe)O-ferroericlase



(Mg,Fe)SiO_2-perovskite



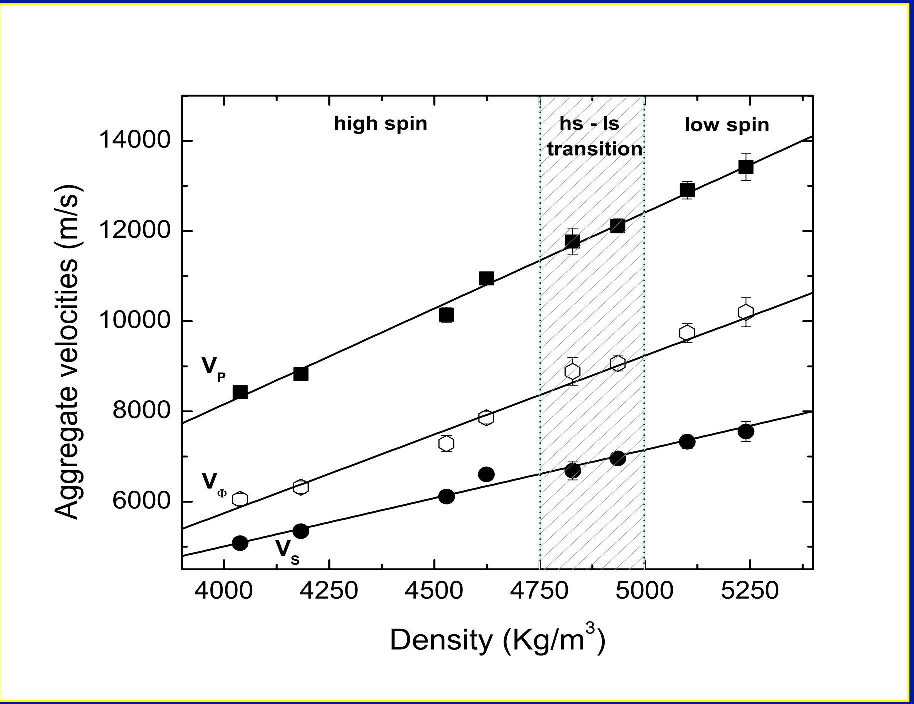
Badro Annu Rev. Earth Planet. Sci. 2014

Geophysical implications?

No detectable effects on 1D seismic profiles

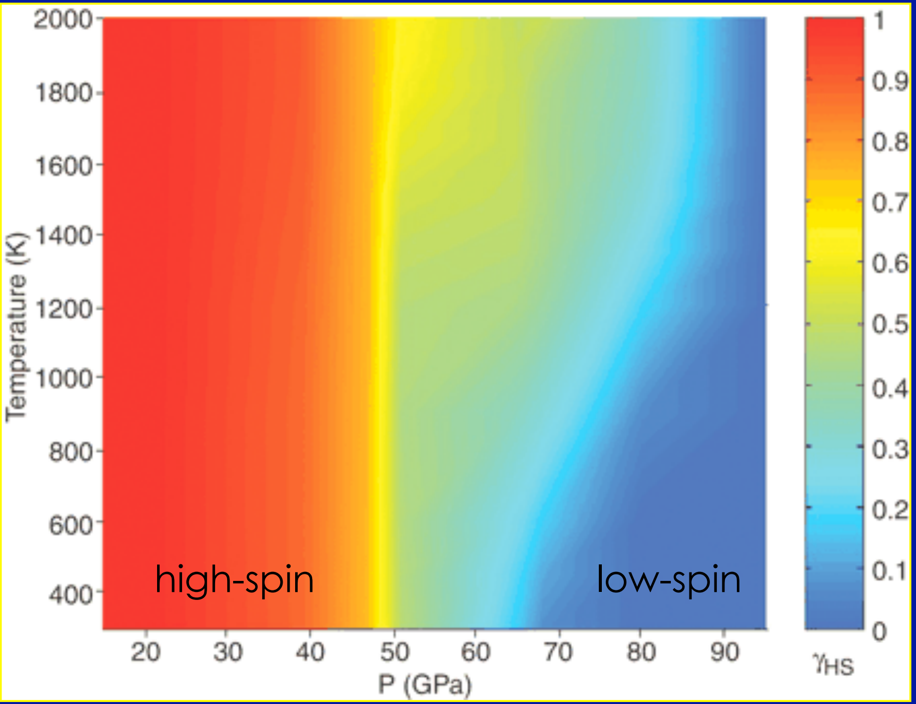
$(Mg_{1-x},Fe_x)O$-ferropericlase

IXS on single crystal Fp at HP → no effects on aggregate velocities



Antonangeli et al., Science 2011

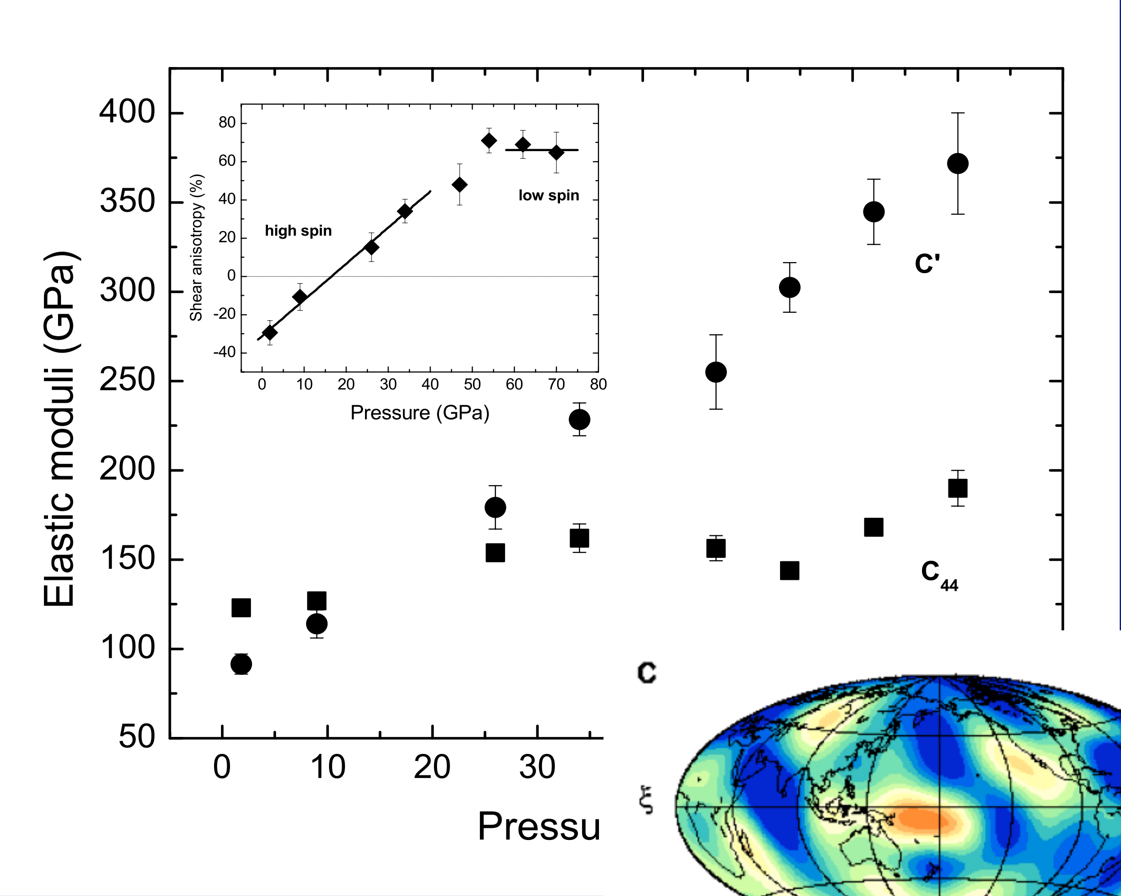
“gradual” spin transition over large P-T range

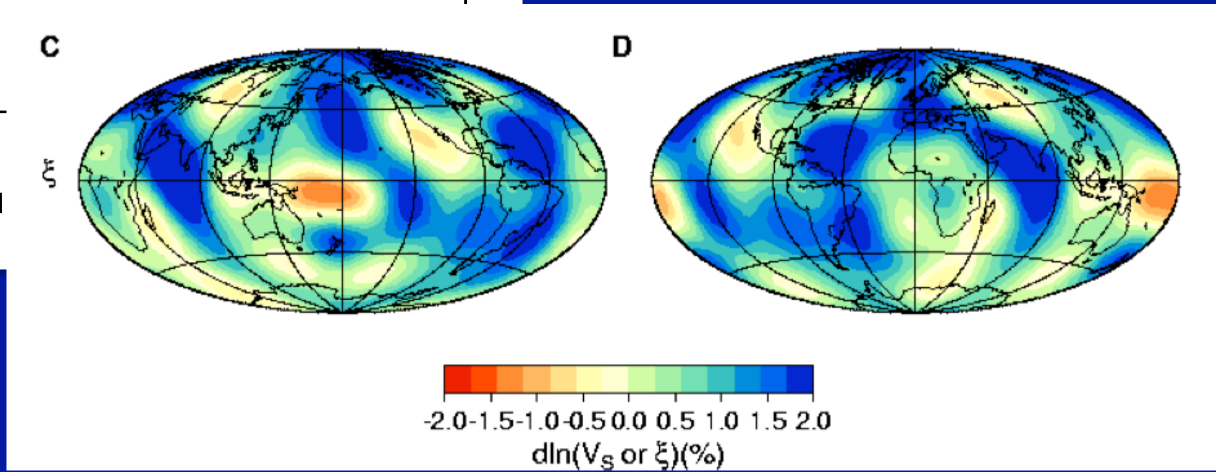


Lin et al., Science 2007

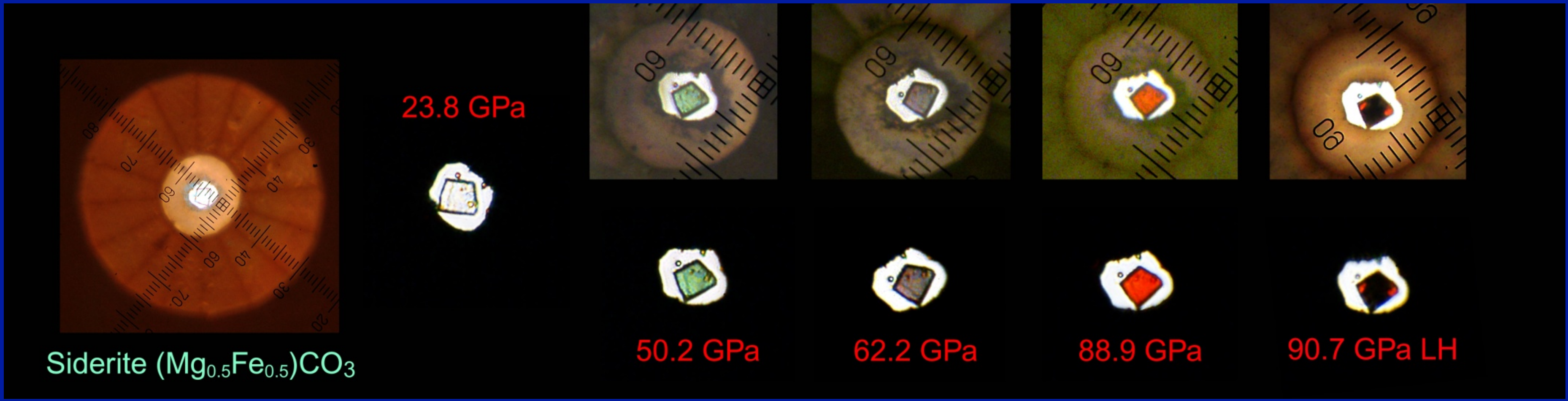
Seismologically transparent transition?
Shear anisotropy in ferropericlase

$A=2(C'-C_{44})/(C'+C_{44})$



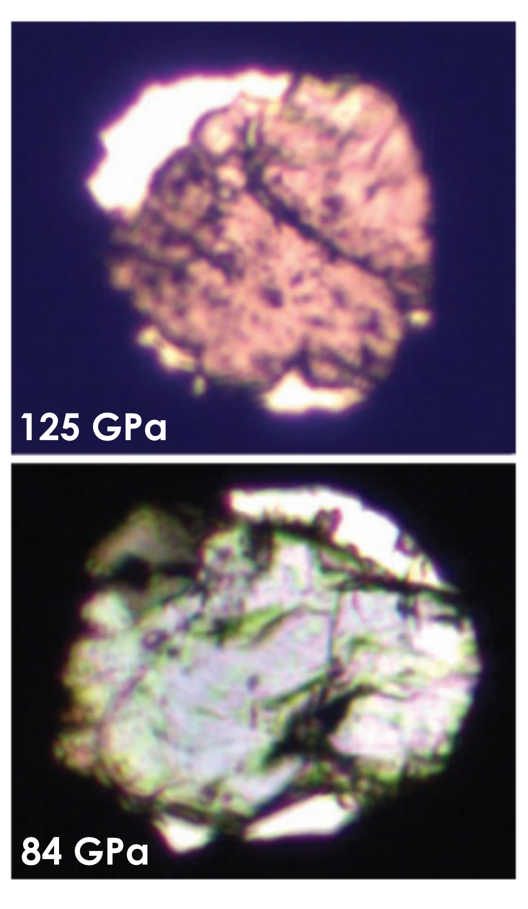


Spin transition and optical properties

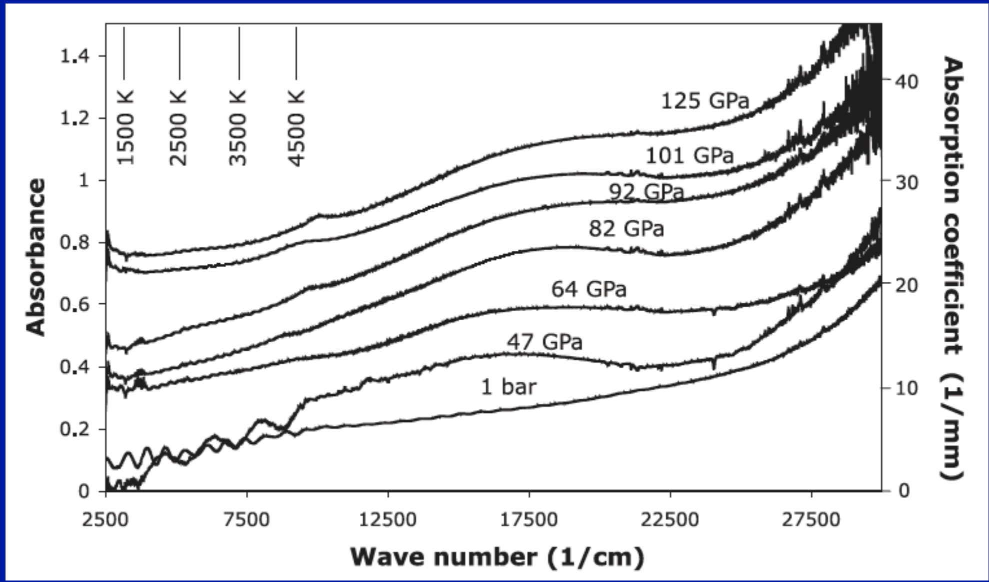


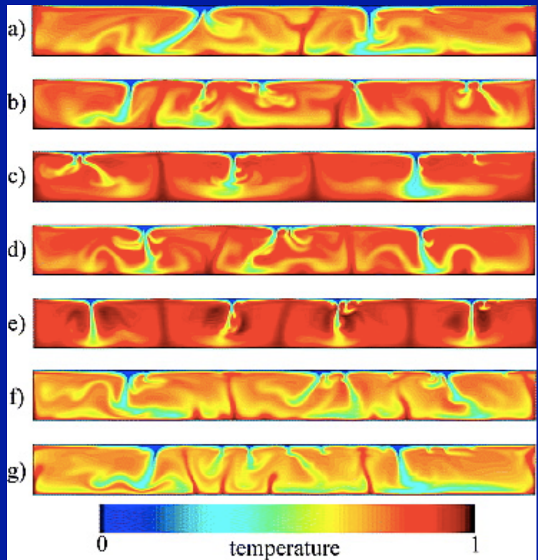
Spin transition and radiative heat transfer

Near IR and visible absorption spectra of $(Mg,Fe)SiO_3$



Keppler et al., Science 2008



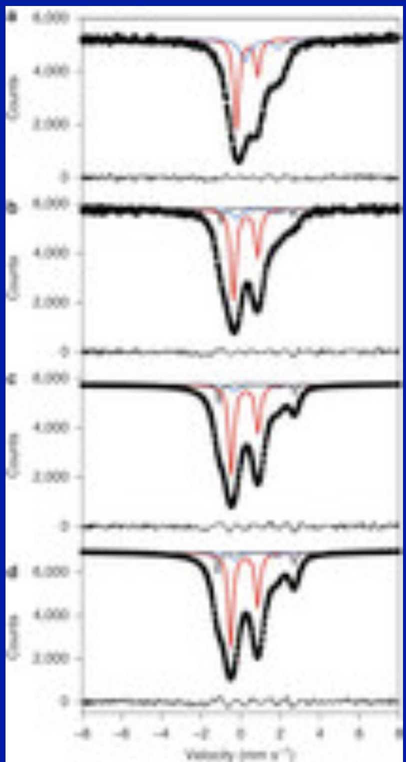


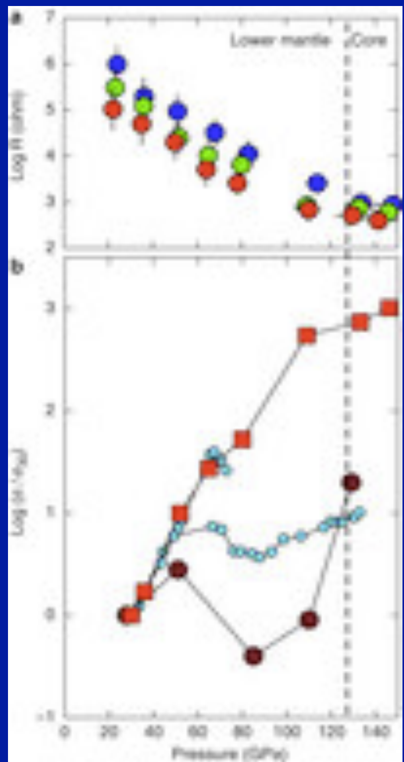
Naliboff et al., GRL 2006

Spin transition and electrical conductivity

SMS experiments on Al-bearing $(Mg,Fe)SiO_3$-perovskite
→ Fe^{3+} remains in HS state throughout lower mantle P-T conditions

Electrical conductivity measurements
→ No drops in electrical conductivity on samples with only Fe^{3+}
→ Drops in conductivity for HS-LS transition of Fe^{2+}



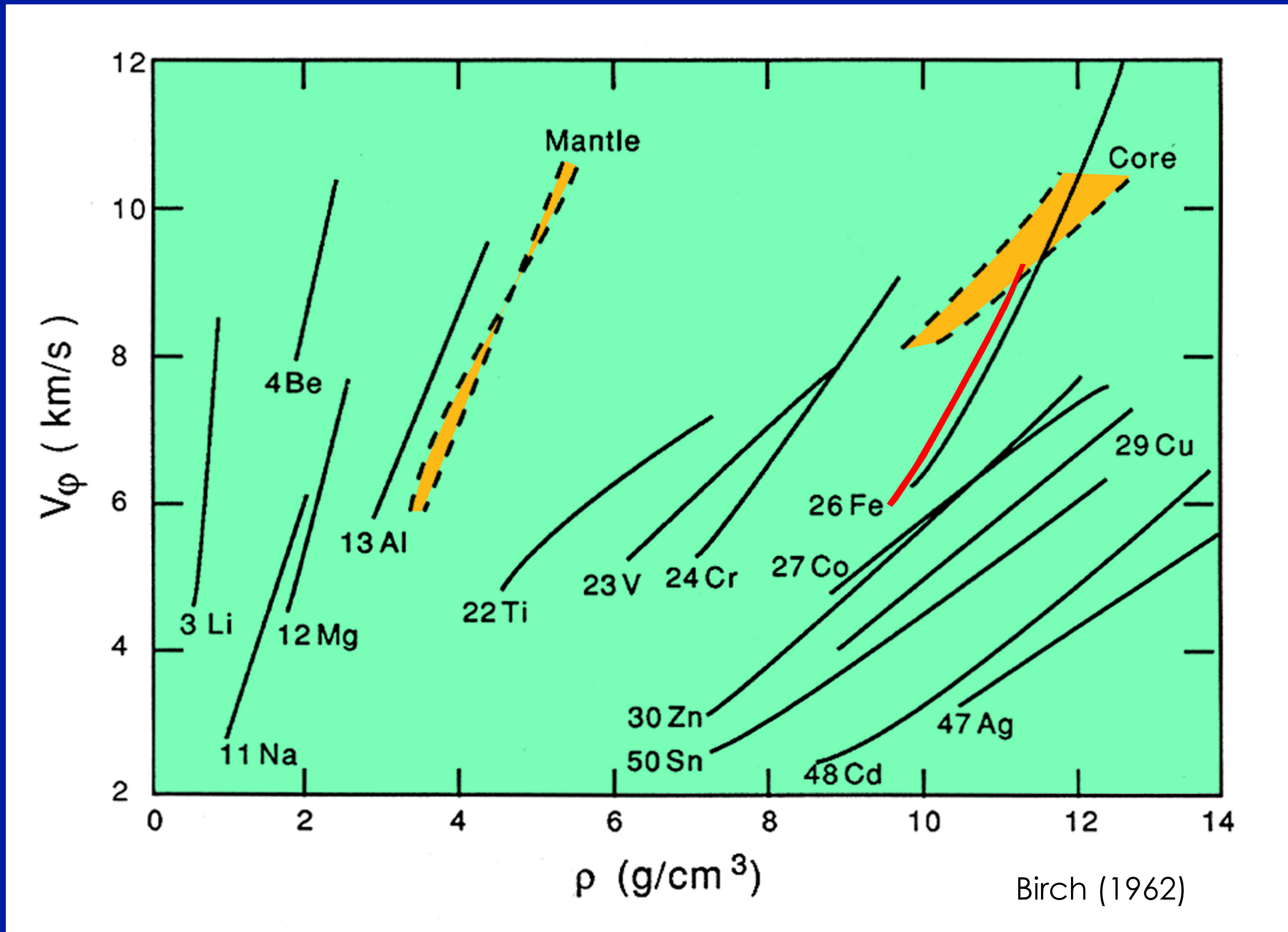


Potapkin et al., Nature Comm. 2013

Spin transitions
←→
Heterogeneities in lower mantle electrical conductivity

The only way out is in…

Core: velocity vs density systematics

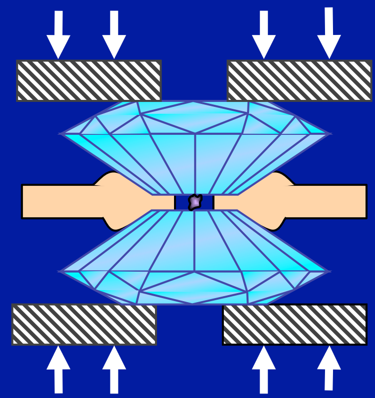


Fe (+Ni) main constituent of Earth's core

3^{rd} generation synchrotron sources + diamond anvil cell

Sample volume < 10^{-5} mm^3

Beam size < 100 μm (<10 μm)



(Non-resonant, momentum resolved) inelastic x-ray scattering

IXS

Nuclear resonant inelastic x-ray scattering

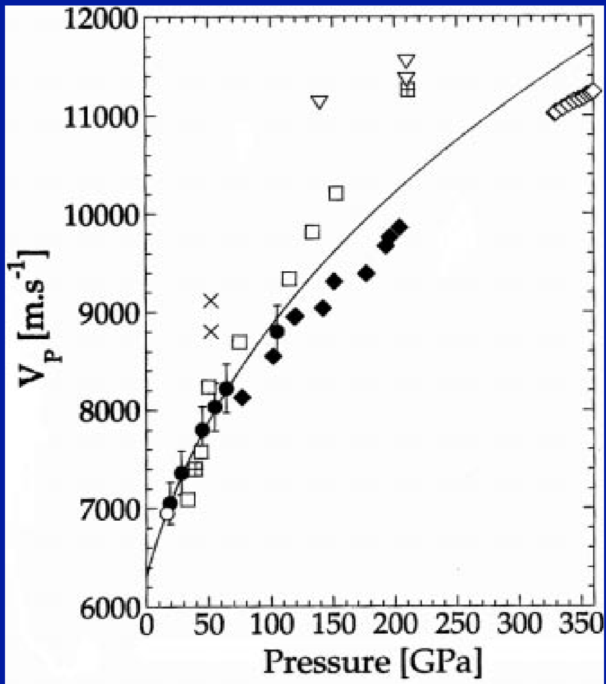
NRIXS

Pioneering experimental studies on Fe…

Sound Velocities in Iron to 110 Gigapascals

Guillaume Fiquet,[1*] James Badro,[1] François Guyot,[1] Herwig Requardt,[2] Michael Krisch[2]

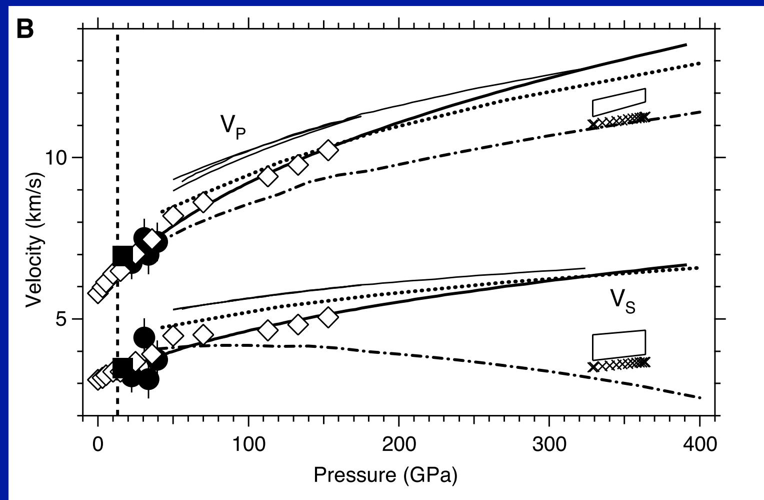
The dispersion of longitudinal acoustic phonons was measured by inelastic x-ray scattering in the hexagonal closed-packed (hcp) structure of iron from 19 to 110 gigapascals. Phonon dispersion curves were recorded on polycrystalline iron compressed in a diamond anvil cell, revealing an increase of the longitudinal wave velocity (V_P) from 7000 to 8800 meters per second. We show that hcp iron follows a Birch law for V_P, which is used to extrapolate velocities to inner core conditions. Extrapolated longitudinal acoustic wave velocities compared with seismic data suggest an inner core that is 4 to 5% lighter than hcp iron.



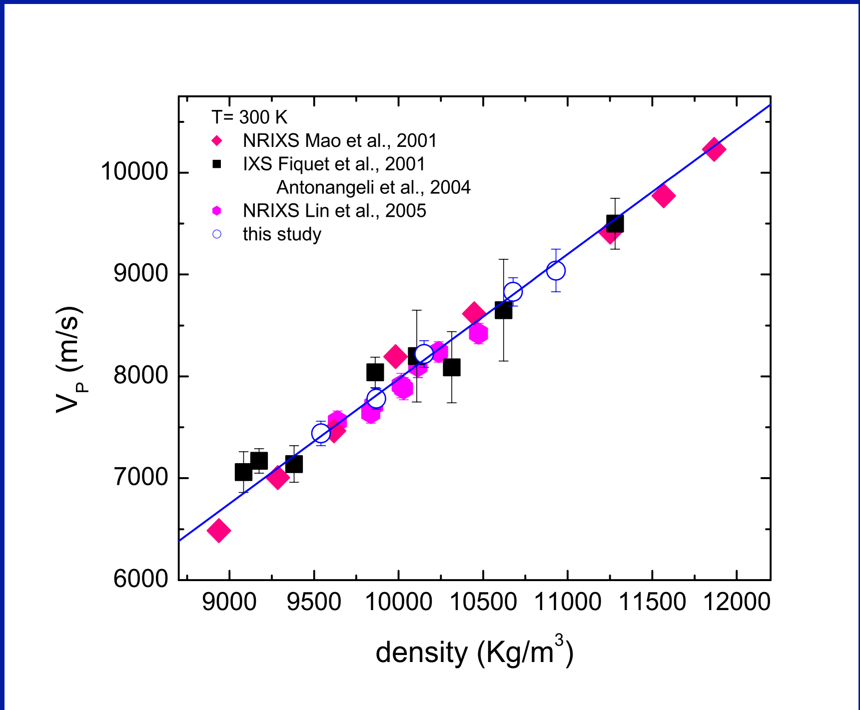
Phonon Density of States of Iron up to 153 Gigapascals

H. K. Mao,[1] J. Xu,[1] V. V. Struzhkin,[1] J. Shu,[1] R. J. Hemley,[1] W. Sturhahn,[2] M. Y. Hu,[2] E. E. Alp,[2] L. Vocadlo,[3] D. Alfè,[3] G. D. Price,[3] M. J. Gillan,[3] M. Schwoerer-Böhning,[4] D. Häusermann,[4] P. Eng,[5] G. Shen,[5] H. Giefers,[6] R. Lübbers,[6] G. Wortmann[6]

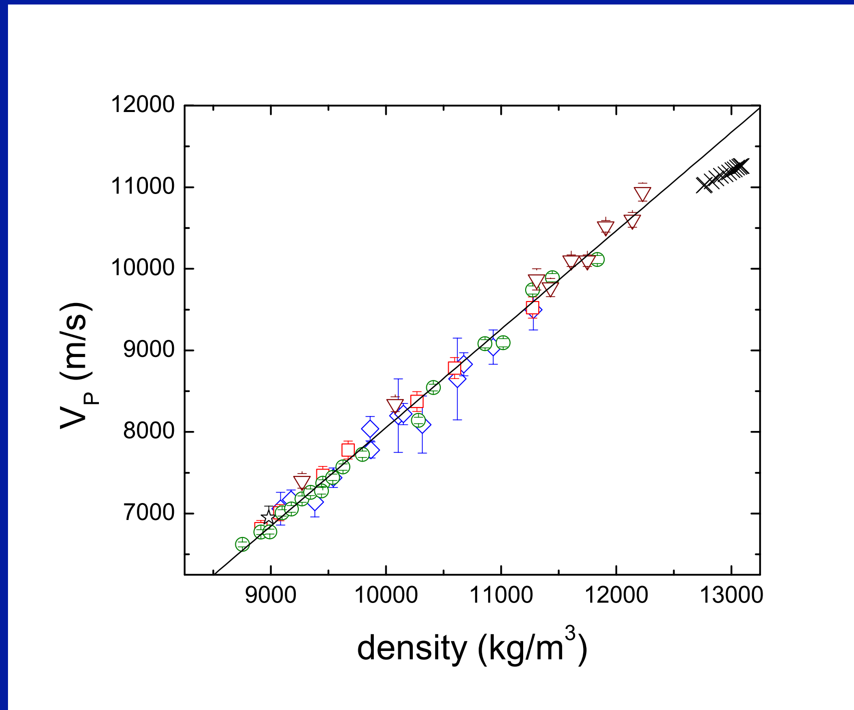
We report phonon densities of states (DOS) of iron measured by nuclear resonant inelastic x-ray scattering to 153 gigapascals and calculated from ab initio theory. Qualitatively, they are in agreement, but the theory predicts density at higher energies. From the DOS, we derive elastic and thermodynamic parameters of iron, including shear modulus, compressional and shear velocities, heat capacity, entropy, kinetic energy, zero-point energy, and Debye temperature. In comparison to the compressional and shear velocities from the preliminary reference Earth model (PREM) seismic model, our results suggest that Earth's inner core has a mean atomic number equal to or higher than pure iron, which is consistent with an iron-nickel alloy.



… converged later on



Antonangeli et al., EPSL 2012

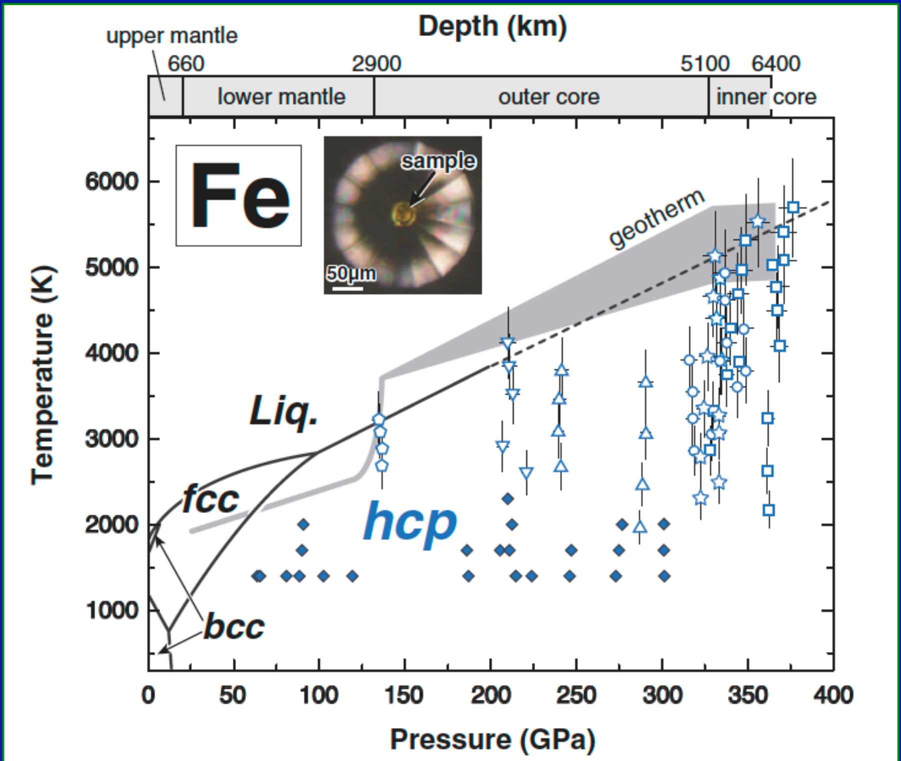


Antonangeli et al., submitted

And in overall agreement with laboratory-based measurements (utrasonics, ISLS, picosecond acoustics)

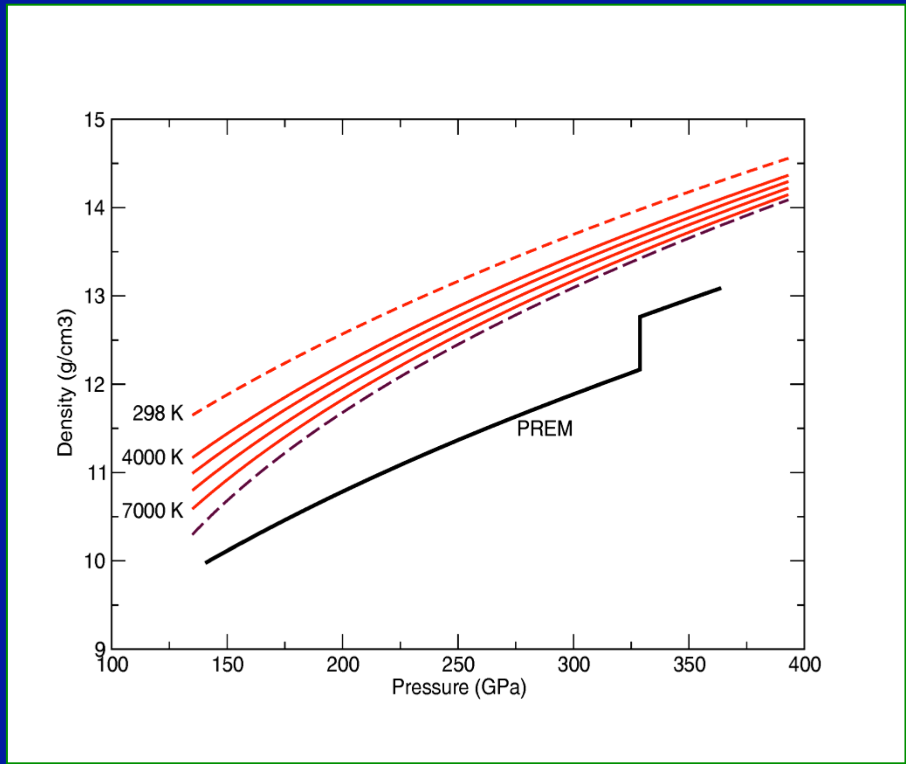
→ Baseline upon which build on complexity
(high temperature, effect of inclusions, visco-elastic relaxations…)

hcp-Fe stable at inner core conditions



Tateno et al., Science 2010

EOS of hcp-Fe vs Earth's models



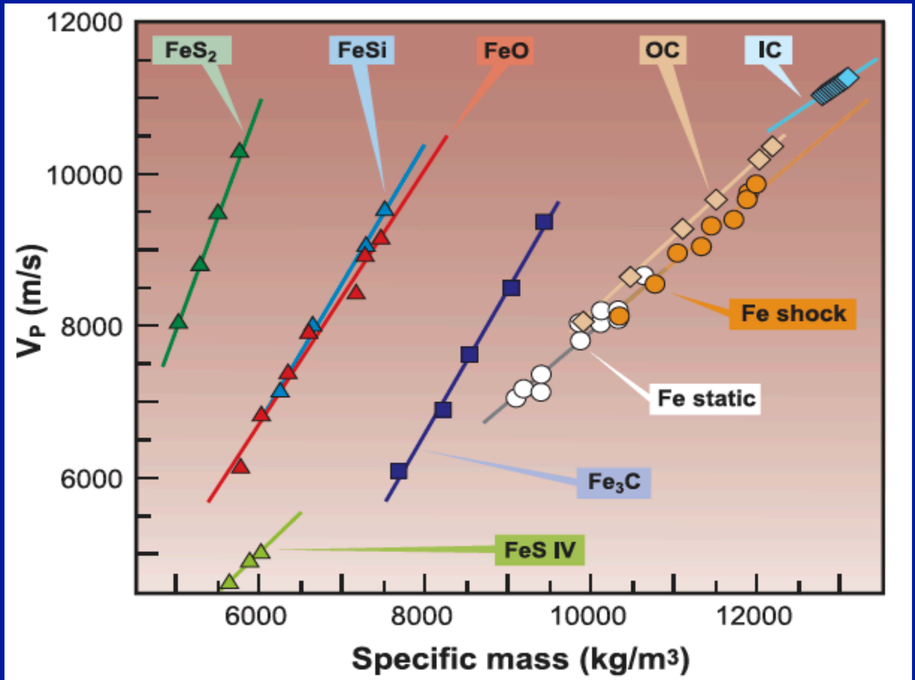
Fiquet et al., in prep

light elements in the core (Si, S, O, C, H …)

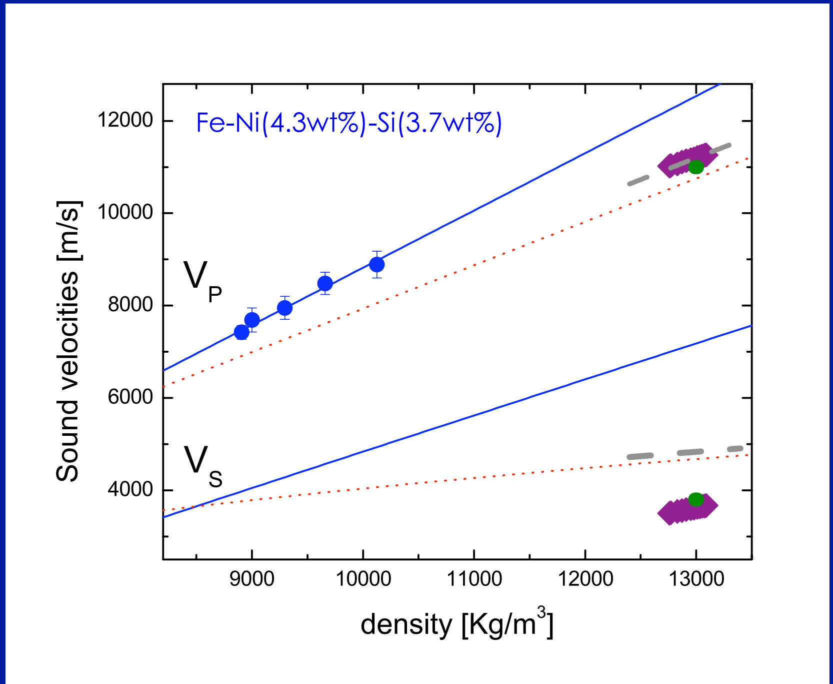
Which effects on the physical properties of pure Fe?

→ Phase stability, phase diagram, melting curves, sound velocity…

Sound velocities (by IXS) and density (by XRD) in solid Fe and Fe-compounds at high P, ambient T



after Badro et al., EPSL 2007; Fiquet et al., PEPI 2009

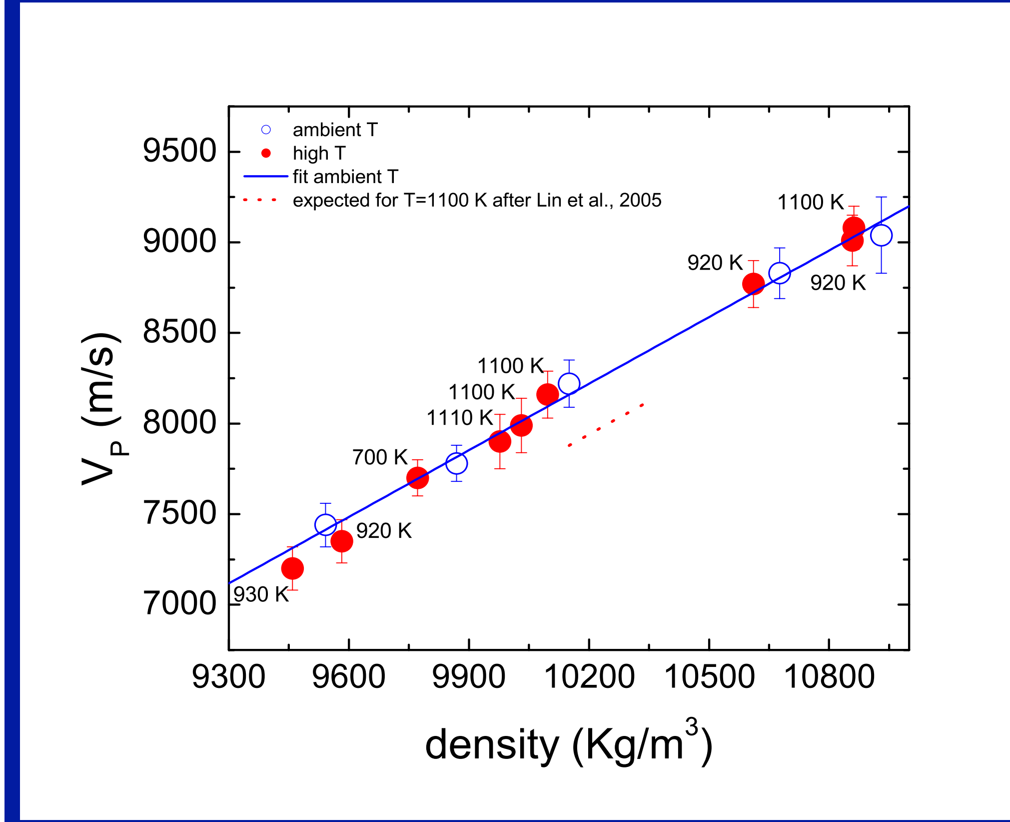


Antonangeli et al. EPSL 2010

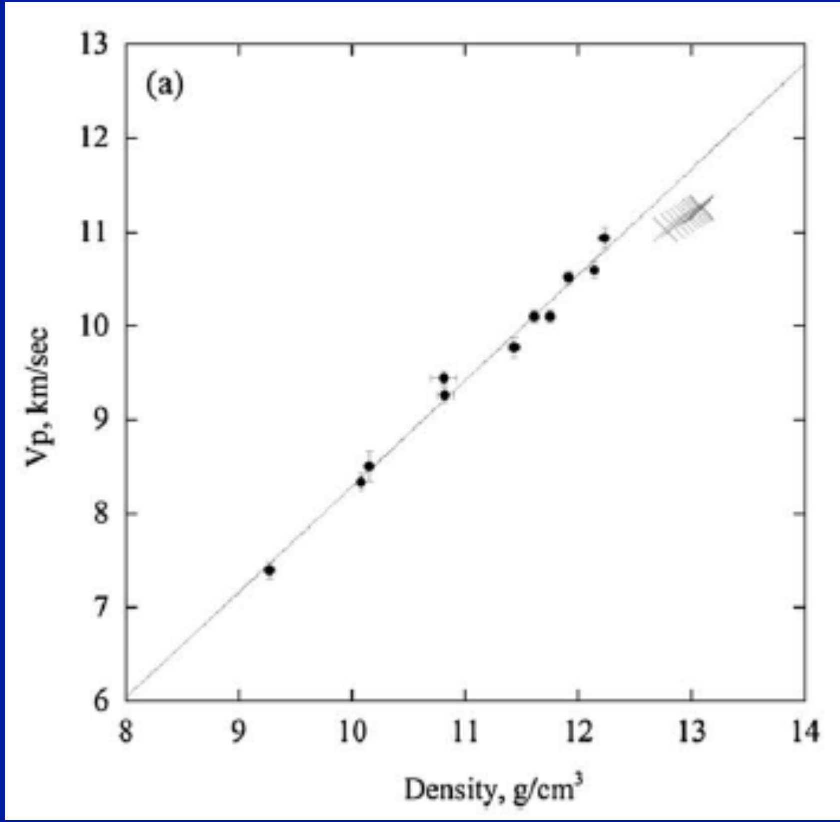
Mixing models →~3 wt.% Si in the inner core; ~5 wt.% O in outer core

but still large pressure extrapolation
and temperature correction from calculations

IXS on pure Fe: no temperature effect up to 1100 K

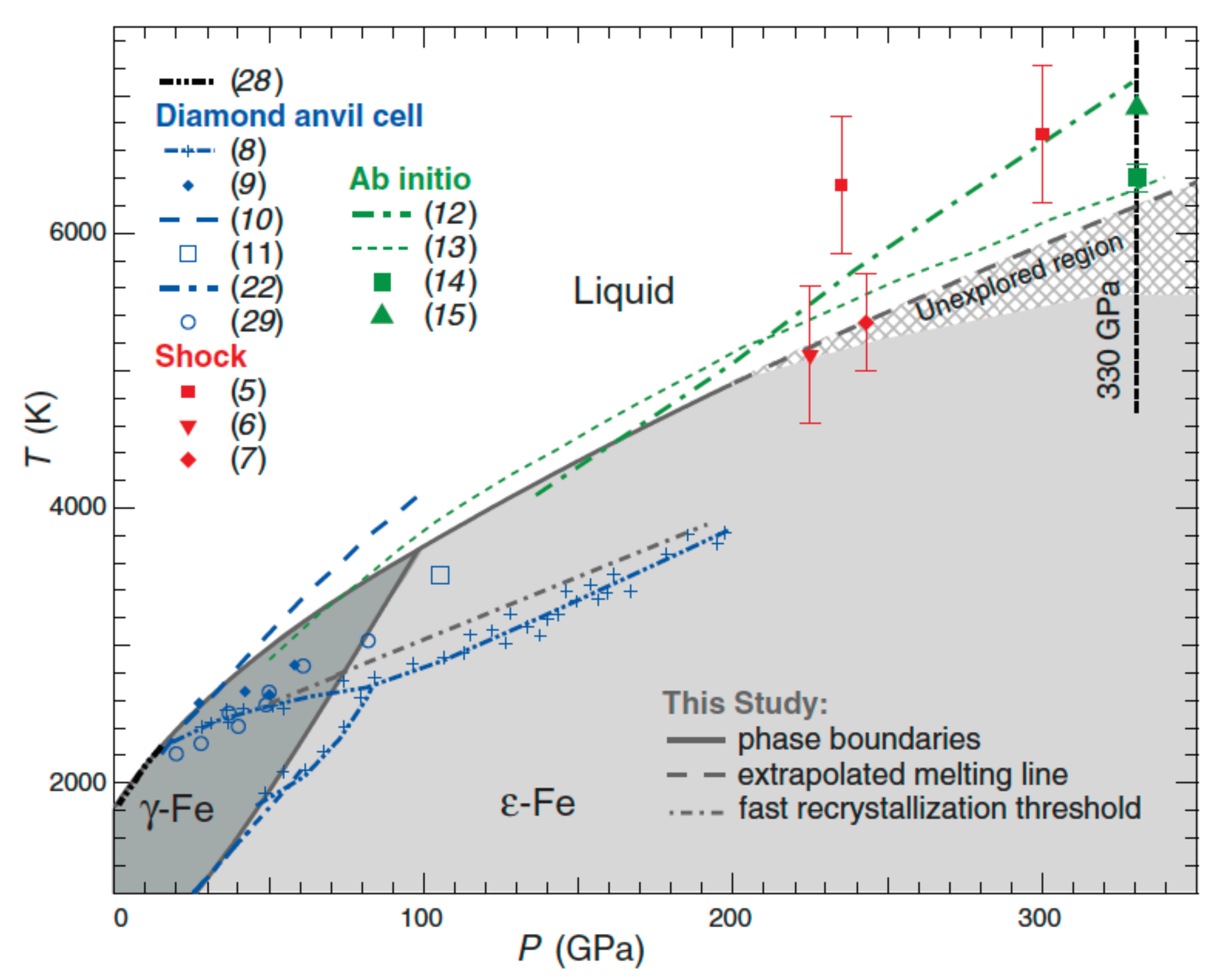


Antonangeli et al., EPSL 2012



Ohtanii et al., GRL 2013

Melting curve of Fe based on fast x-ray diffraction

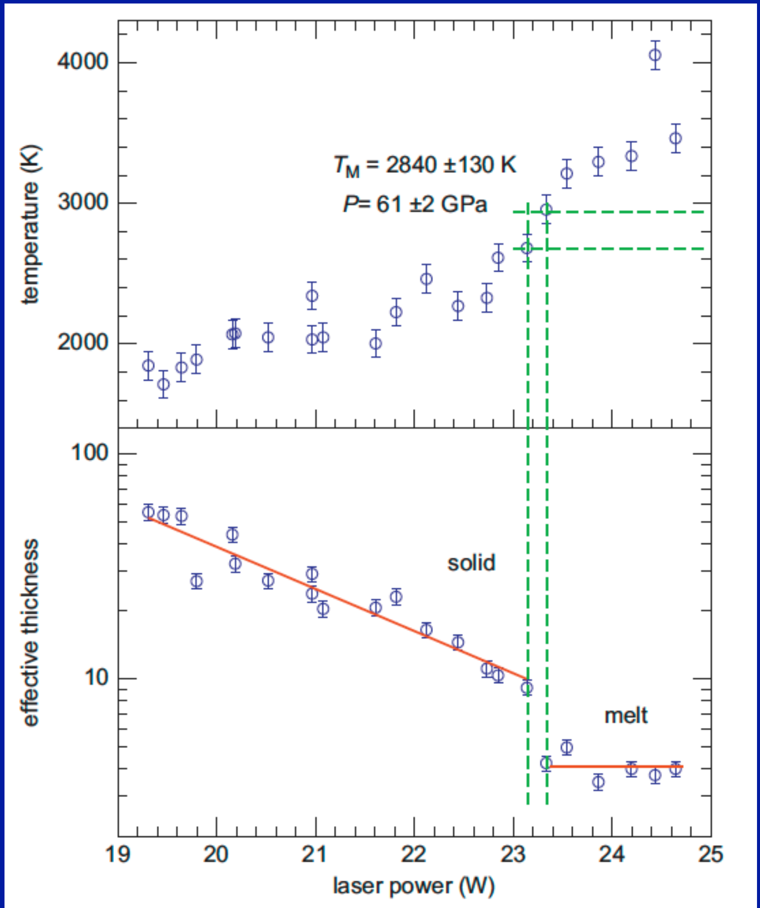


Anzellini et al., Science 2013

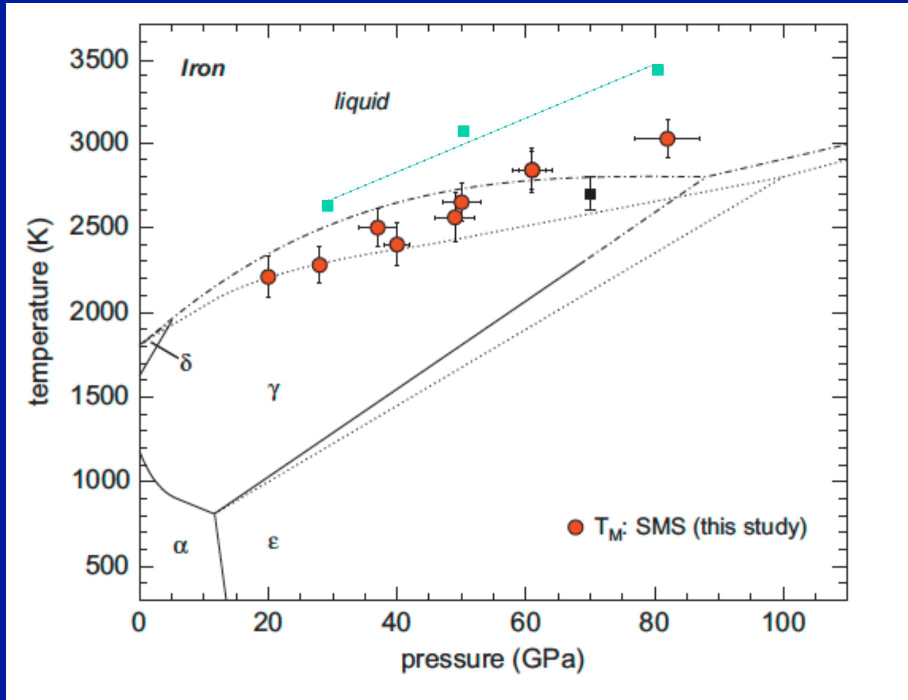
Melting curve of Fe based on atomic dynamics

Idea: monitoring mean-square displacement of atoms (solid ≠ liquid) by measuring the Lamb-Mössbauer factor (goes to 0 for liquid)

Nuclear forward scattering



Jackson et al., EPSL 2013



Earth's and planetary interior
What do we need? Synergetic approach

- Complementarity between static and dynamic experiments
- Complementarity between experiments and calculations

  → Central role of dynamical properties

- Complementarity between synchrotron, neutron and laboratory-based techniques
- Complementarity in backgrounds (physics, chemistry, Earth science, planetary science…)