

Warm Dense Matter (WDM) studies on FEL facilities

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Summary :

1 / Warm Dense Matter (WDM)

- Fundamentals
- Create WDM in laboratory
- Challenges

2/ Ultrafast X-ray diagnostics for WDM

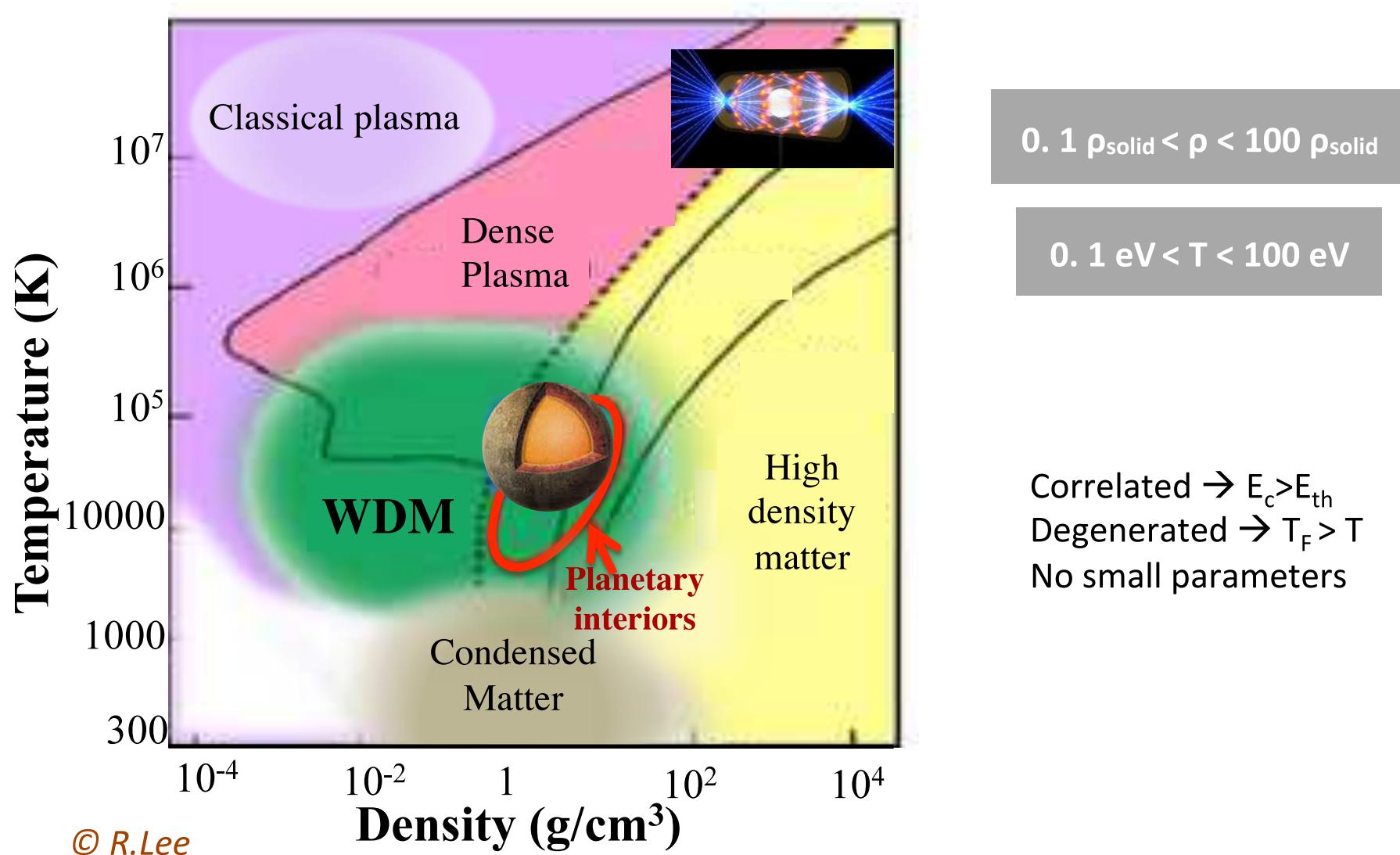
- X-ray Scattering
- X-ray absorption

3/ WDM through its time scales

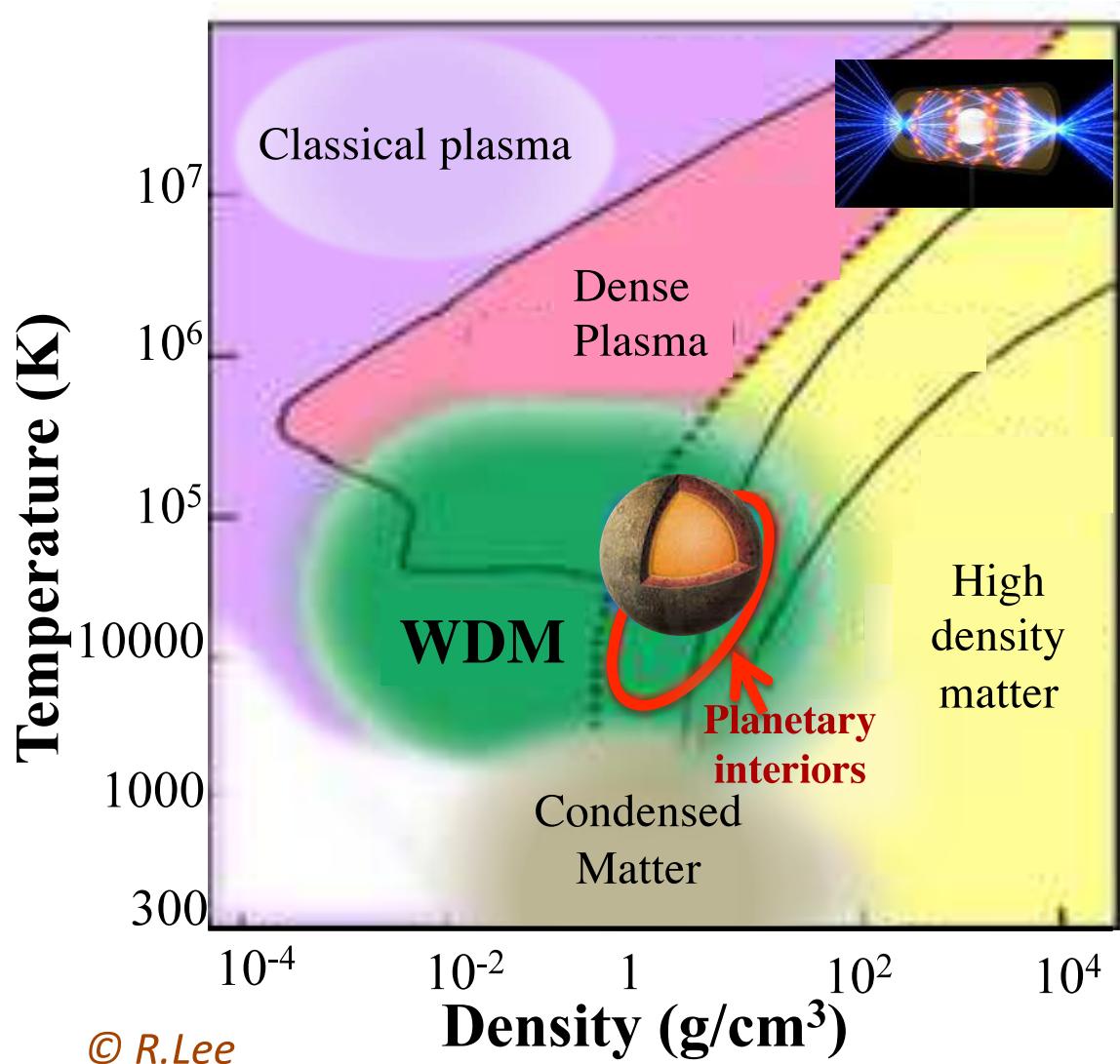
- Ultrafast heating
- Thermal equilibration
- Phase transitions and phase diagrams
- Shock deformation

4/ Conclusion & Perspectives

Warm Dense Matter (WDM): complex and almost unexplored regime



Warm Dense Matter (WDM): complex and almost unexplored regime



Modelisation are difficults
Perturbation theory invalid
Perfect gaz does not apply

- Approximations of plasma physic?
- Approximation from solid physics?

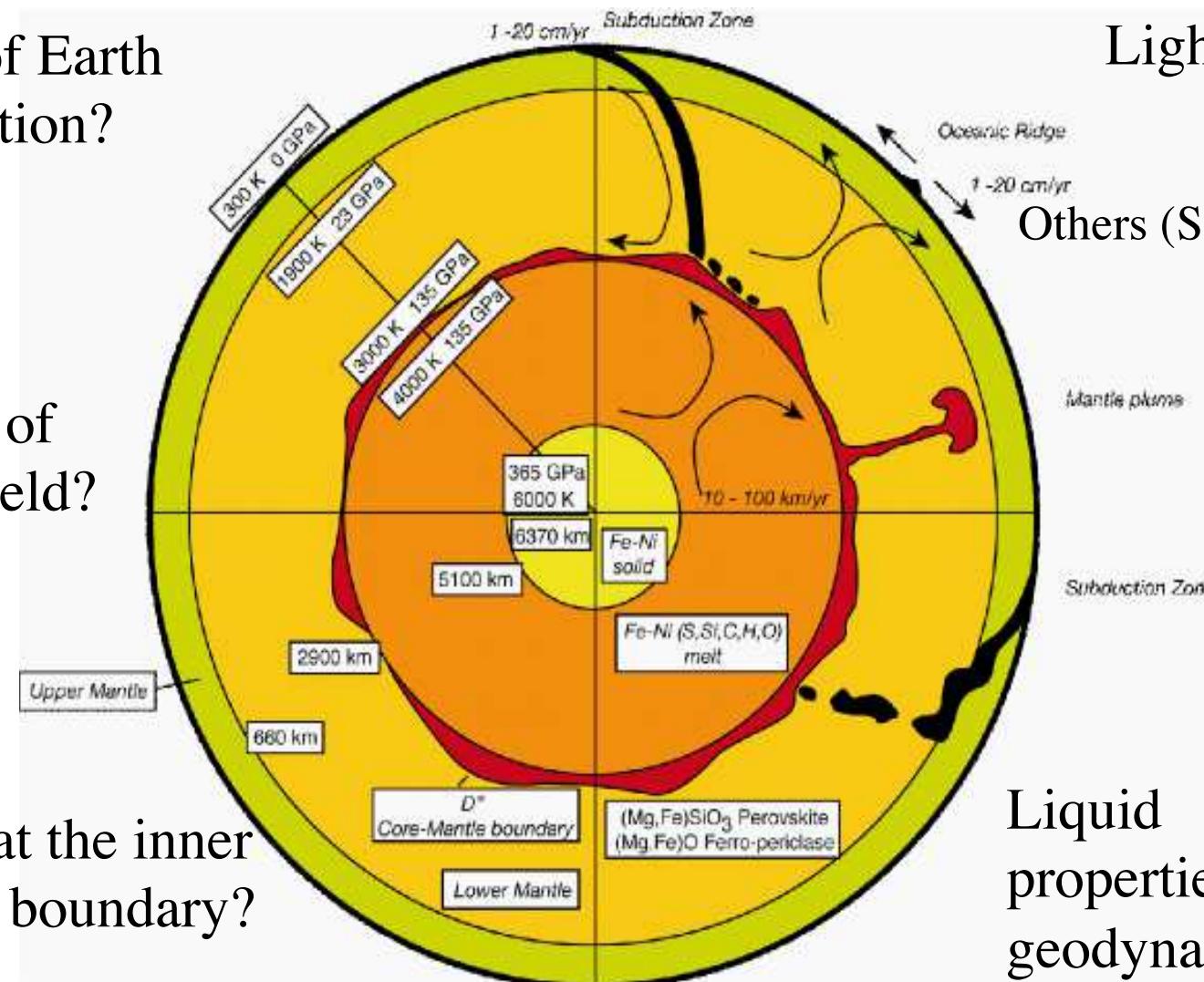
Inner structure of the Earth

Scenario of Earth differentiation?

Generation of Magnetic field?

Heat flux at the inner outer core boundary?

100GPa = 0.1TPa, 1Mbar

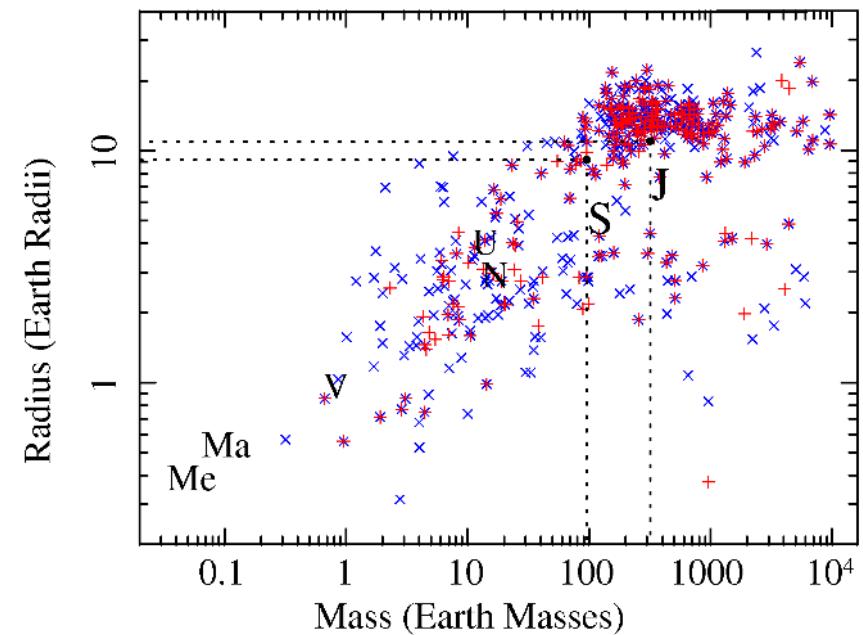


Liquid properties affect geodynamo?

Exoplanets

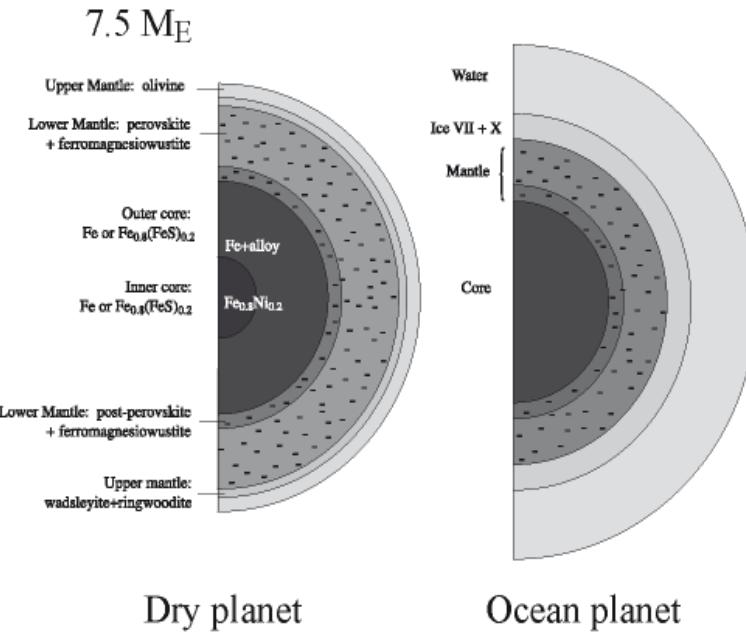


3727 confirmed exoplanets (Kepler) (10.01.2017)
1 discovery / 2 days
> 70 habitable planets (Dec 2017)
Internal pressures up to 10s TPa



2014, Yaqoob et al

Exoplanets Interiors and properties



Core-Mantle Boundary
 $P = 1100 \text{ GPa}$
 $T = 5000 \text{ K}$

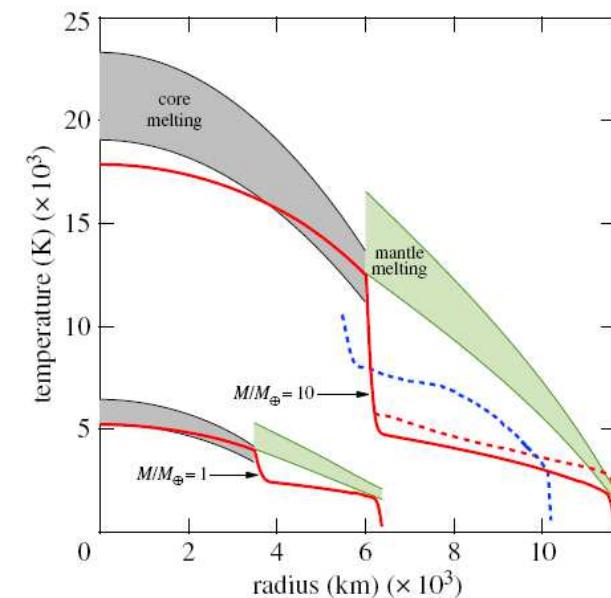
Center of the planet
 $P = 3400 \text{ GPa}$
 $T = 7000 \text{ K}$



Stixrude, 2014

Valencia 2007

Big inner core, entirely solid?
Tectonic?
Geodynamo?
Liquid core?
...



Hyper-velocity impacts on Earth



Meteor Crater (USA)
50 000 years
1200m diameter
Meteorite of 50m 300 000 tons
10-20km/s



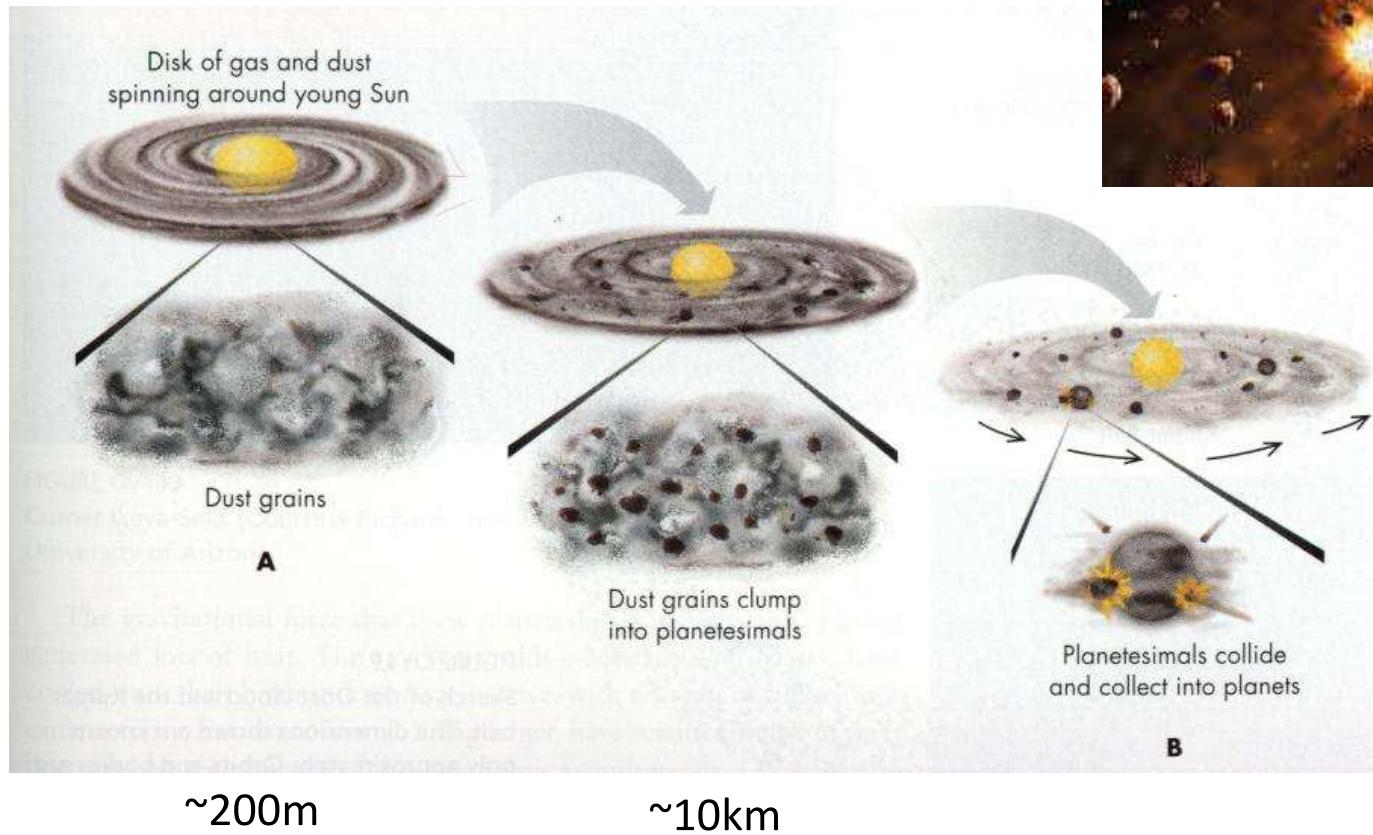
Popigai Crater (Russia) converted
to a Diamond mine
35 millions years



Cheyabinsky
Meteorite
10000Tons, > 15m,
~19km/s (Russia)

Understanding crater
shapes and projectile
left overs?
Formation of diamond,
stichovite?
Preventing impacts?

Planet formation and accretion process



What are the properties of the initial materials?
Presence of water?

Moon formation – Giant Impacts

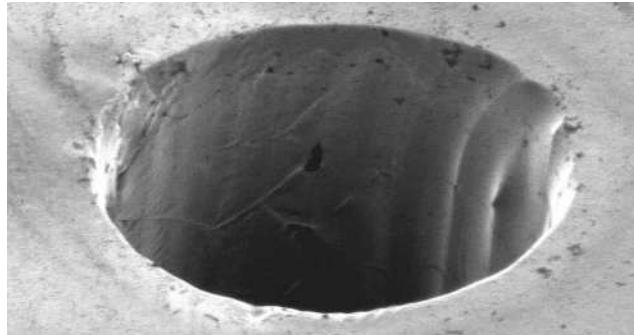
Collision between a proto-Earth and a Mars size body, 4.5Gy ago

Many debris from the collision and accretion to form the Moon ?

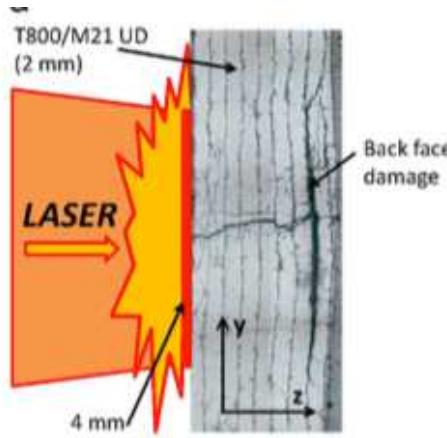
How much is left from the external body in the Earth?



Laser process

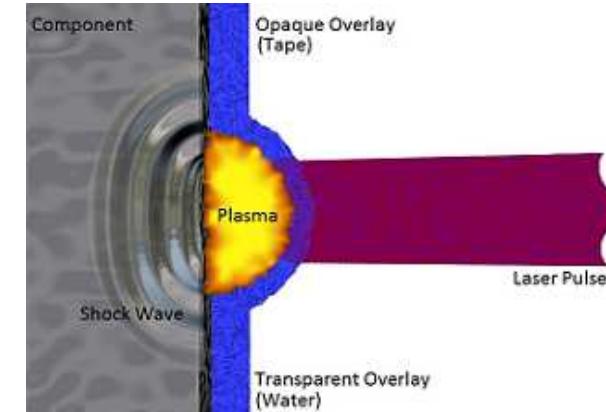


Laser drilling

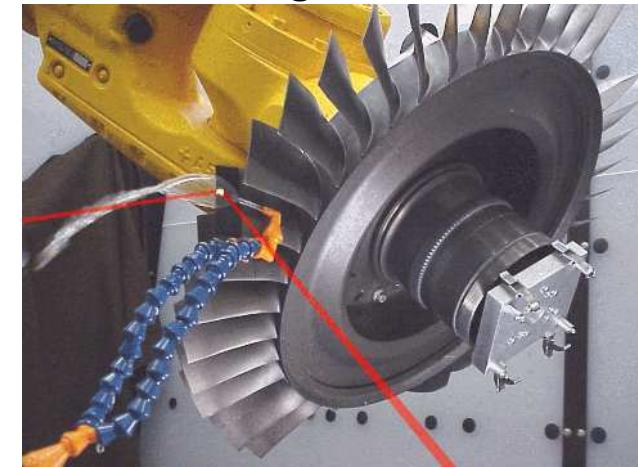


Laser shock adhesion Test

Laser welding



Laser Peening



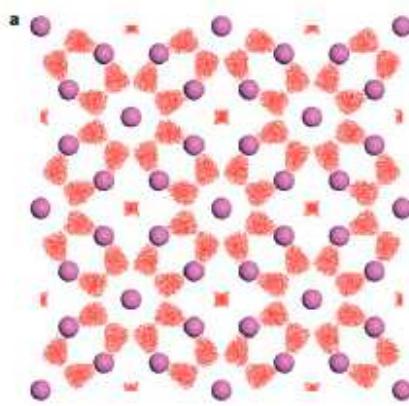
Ehrhart et al 2014

Shock deformations and
microstructures?
Change of absorption
during drilling and welding

Fundamental and unknown physics

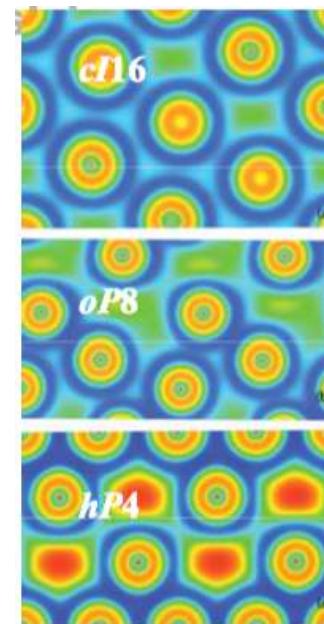
At high pressures and high temperature, electron ion interaction can play a major role

- Orbital hybridization
- Interstitial, localized electron bonding
- Gap changes...
- bond hardening

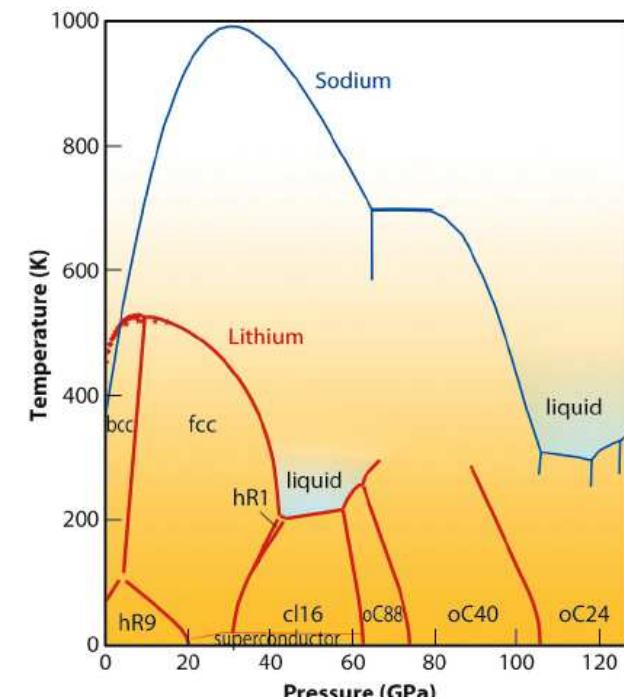


Host guest structures
in Al at Tpa pressures

Pickard, *Nature Materials* 9, 2019



Electron localized function of Na
Marques et al, *PRB* 2009



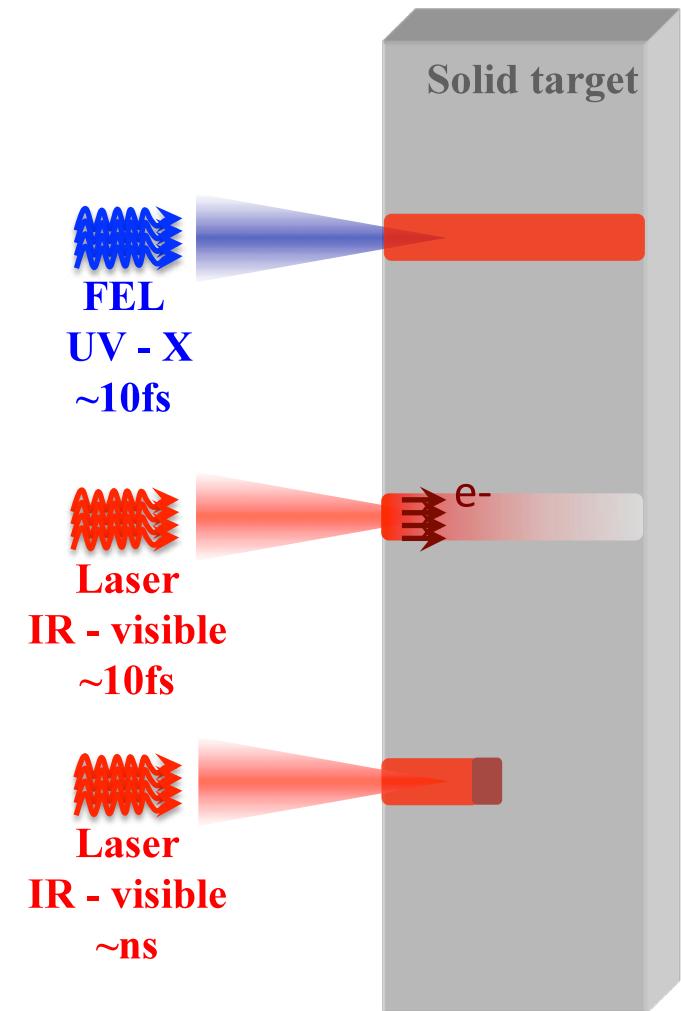
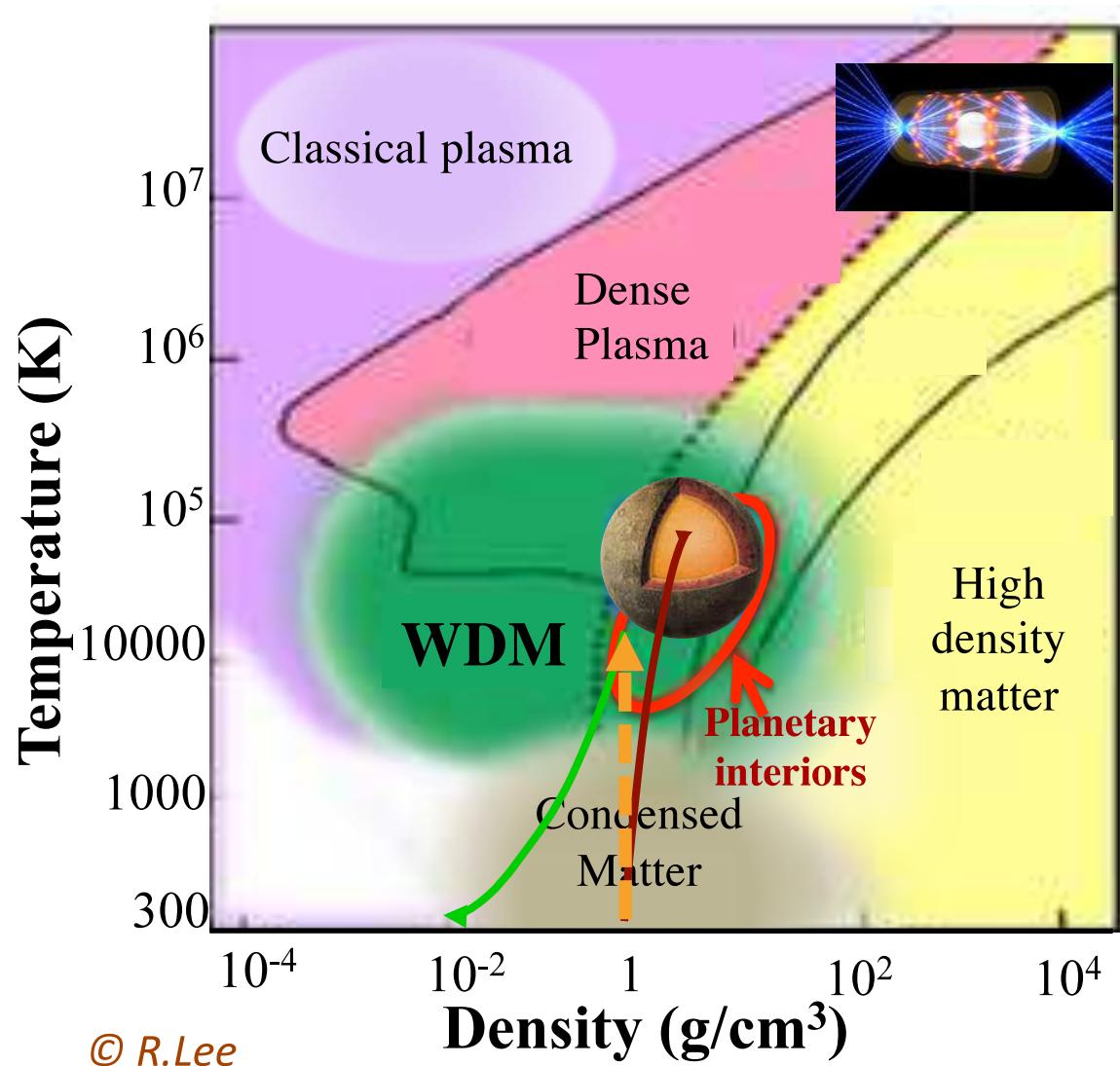
Effect on the melting line
Gregoryanz et al., *PRL* 2005

Few important challenges for WDM

- Phase diagrams and EOS at extreme pressures and temperatures (Inner structure of Earth and Exoplanets)
- Fundamental physical properties at extreme conditions (Liquid densities, metalisation, conductivity for planet modeling...)
- Phase transition during hyper-veloce impact (origin of water on Earth)
- Crater formation and debris production in space
- Exotic state of matter at high pressure
- Laser absorption and optimisation of laser cutting
- Deformation mechanisms under high strain rates (Laser peening)

Needs experimental measures in
laboratory

How to produce WDM in laboratory



Isochoric heating

Ultrafast heating produces non thermal equilibrium $T_e \neq T_i$

Laser heating only on the skin depth ($<100\text{nm}$)
X-ray heating in volume but produces energetic photoelectrons

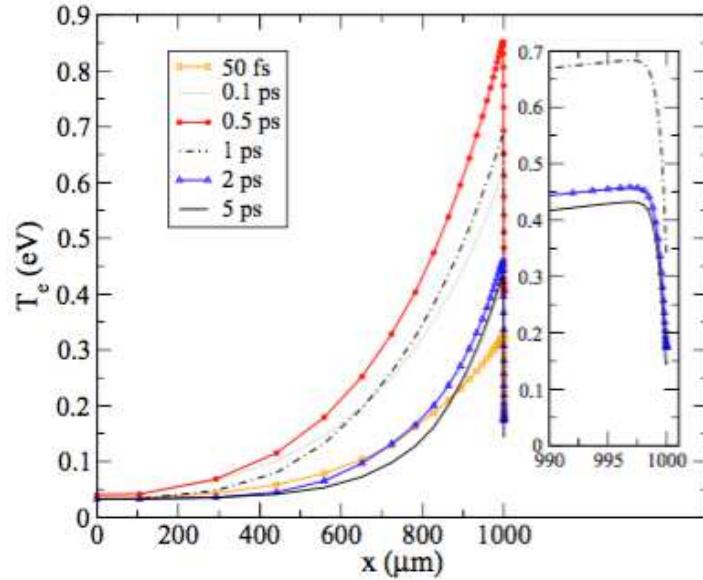


FIG. 9. Calculated electron temperature profiles in a 1-mm-thick bulk Si target *with* transport of energetic photoelectrons. Irradiation conditions of Fig. 8.

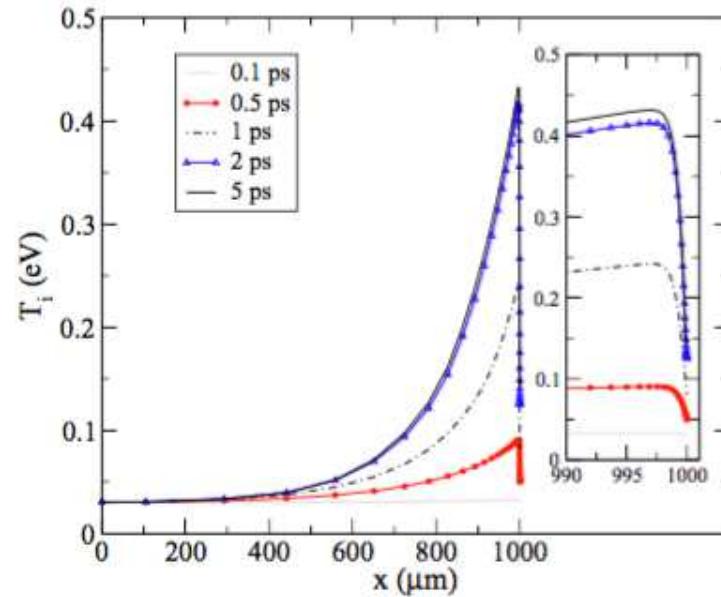
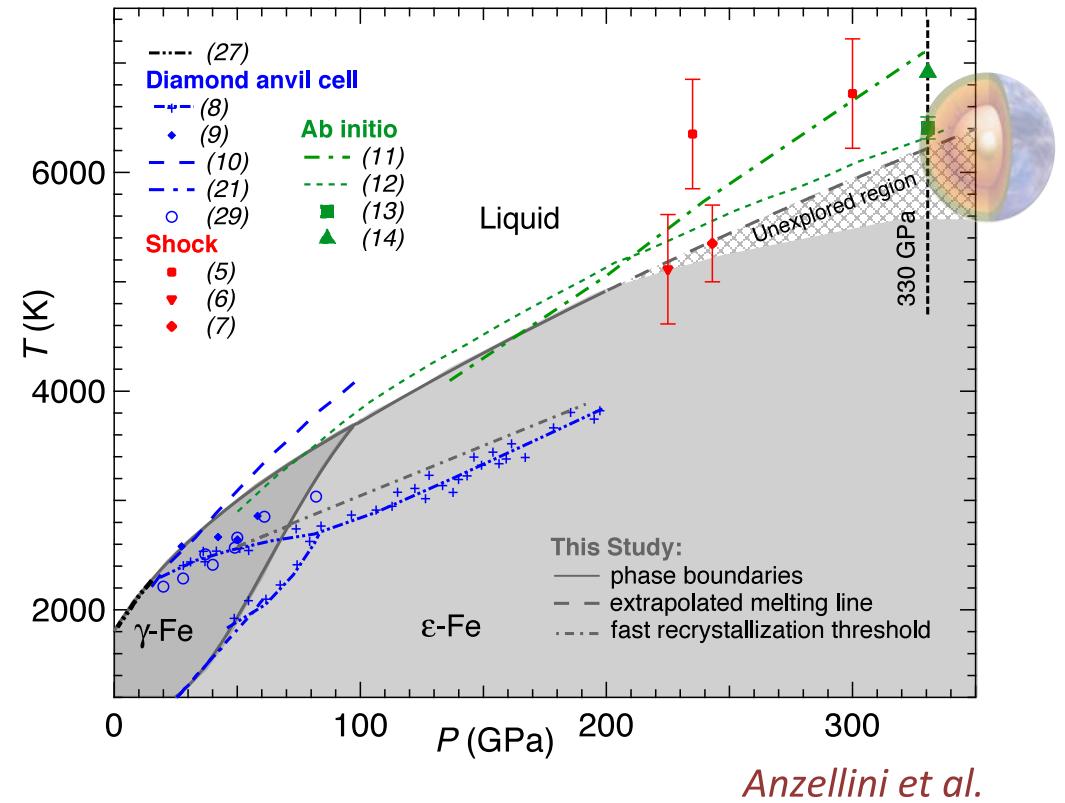
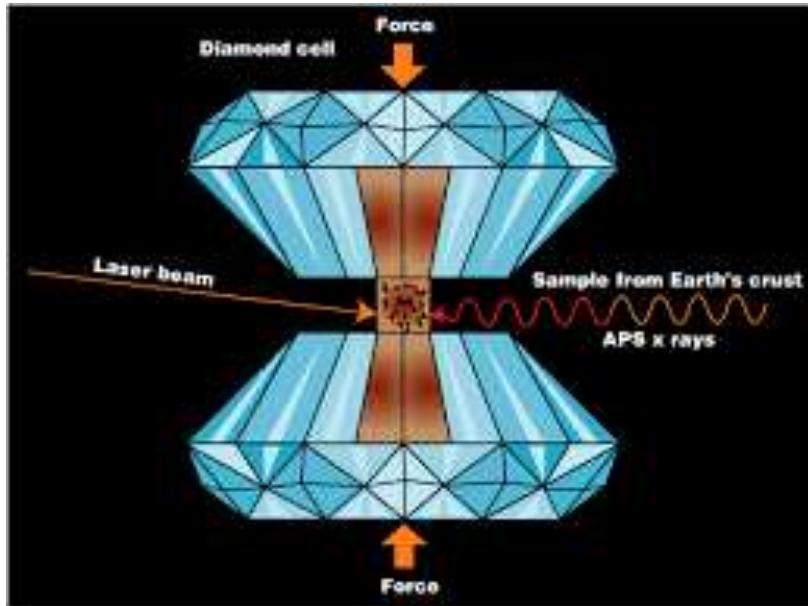


FIG. 8. Calculated ion temperature profiles in a 1-mm-thick bulk Si target *with* transport of energetic photoelectrons. The intensity is of $3.1 \times 10^{15} \text{ W/cm}^2$, the pulse (Gaussian) duration is 20 fs (FWHM), and the photon energy is 10 keV. XFEL radiation comes from the right at normal incidence.

Static compression (LH-DAC)



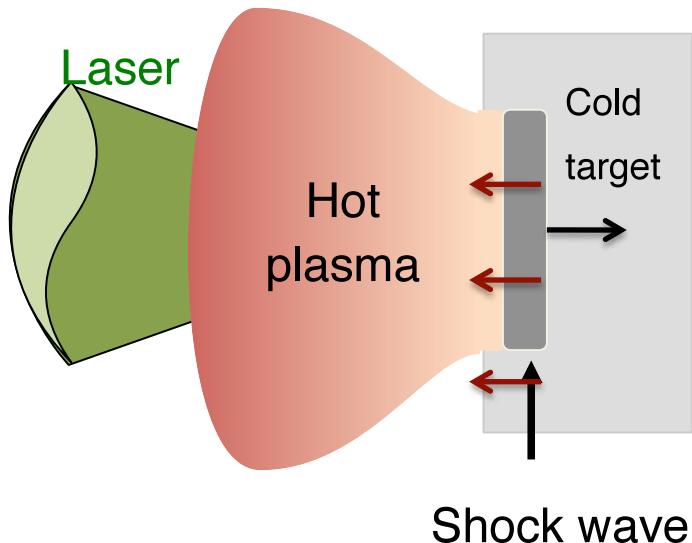
Wide range of P,T conditions

Hydrostatic conditions

Contamination problems (mostly for liquids measurements)

Anzellini et al.

Dynamic compression



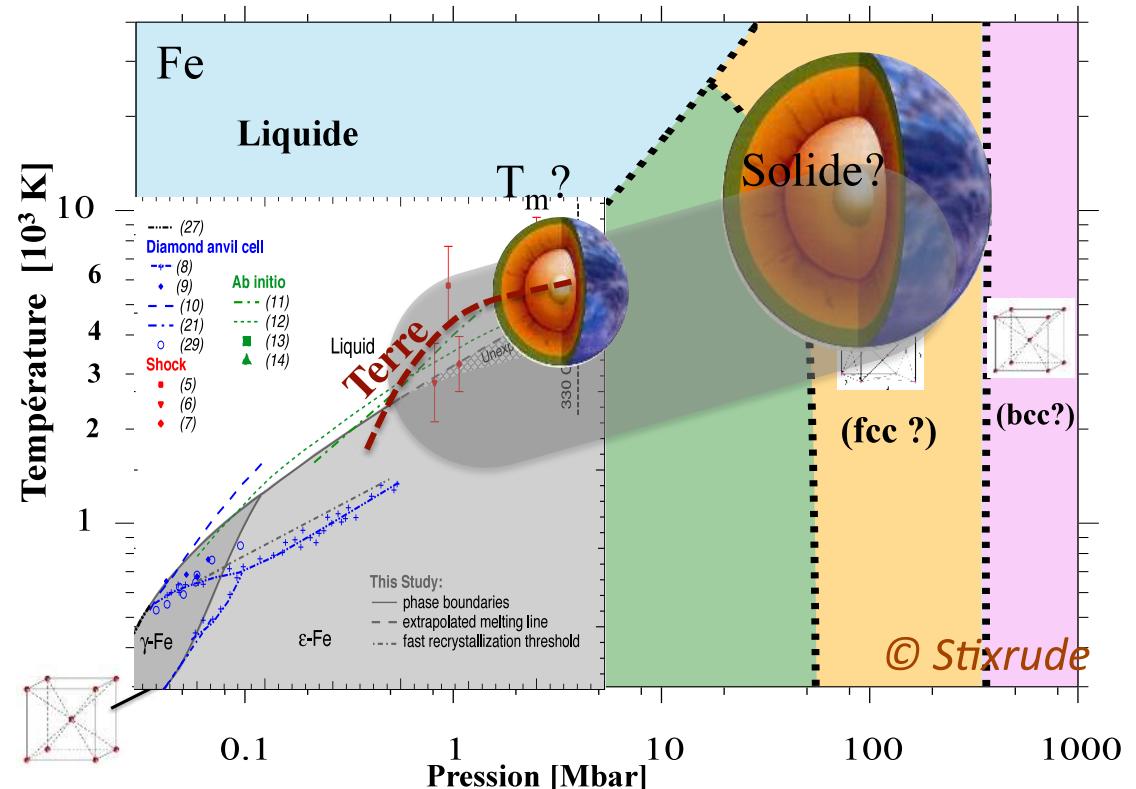
Ablation pressure

$$P \approx 12(I_L / \lambda)^{2/3}$$

$$I_L \rightarrow 10^{14} \text{ W/cm}^2$$

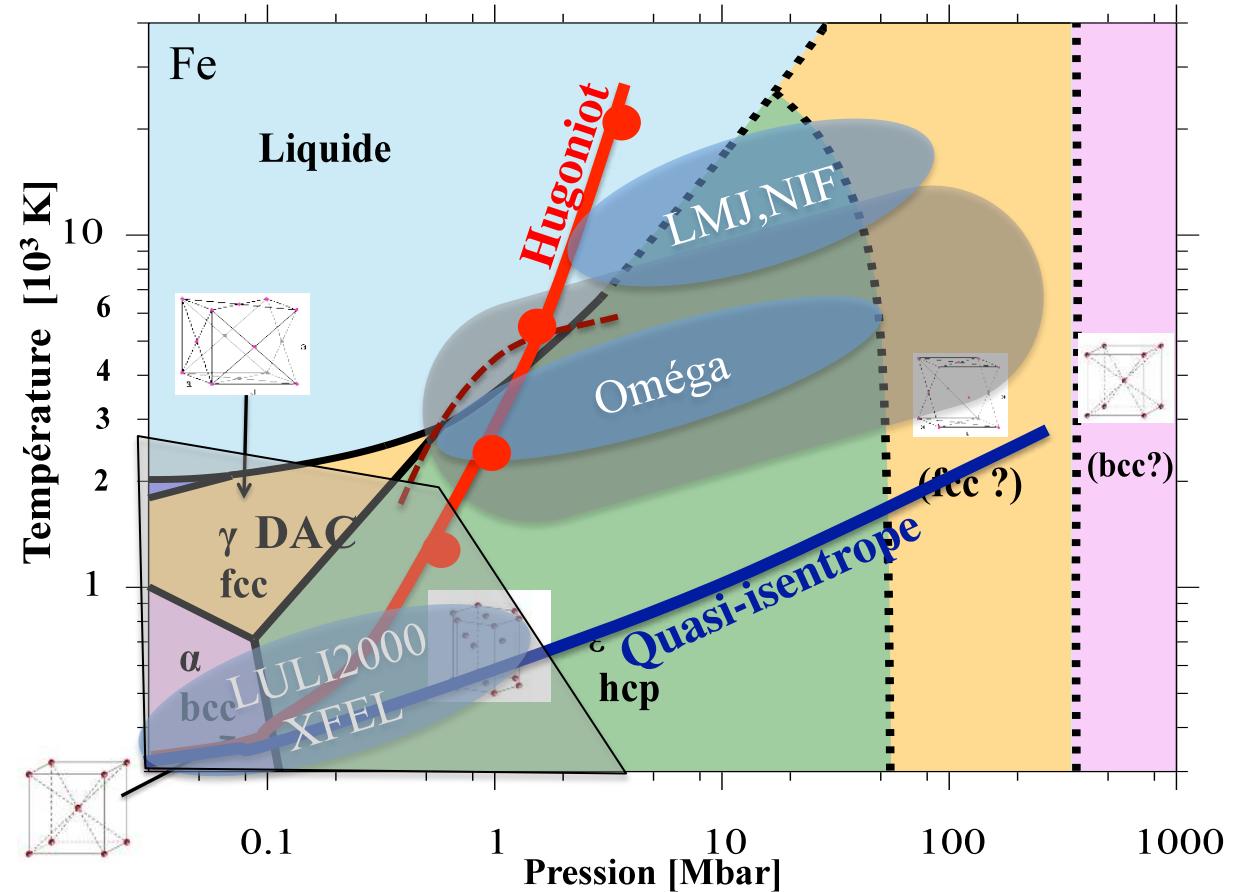
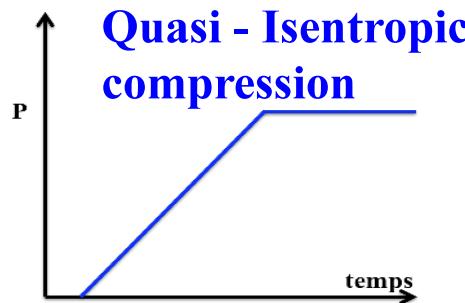
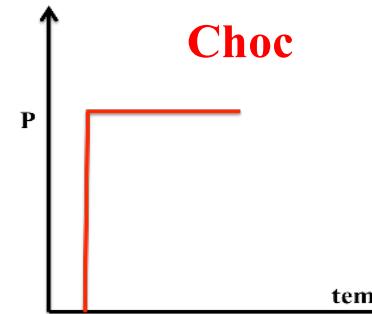
$$\lambda \rightarrow \mu\text{m}$$

$$P \rightarrow \text{Mbar}$$



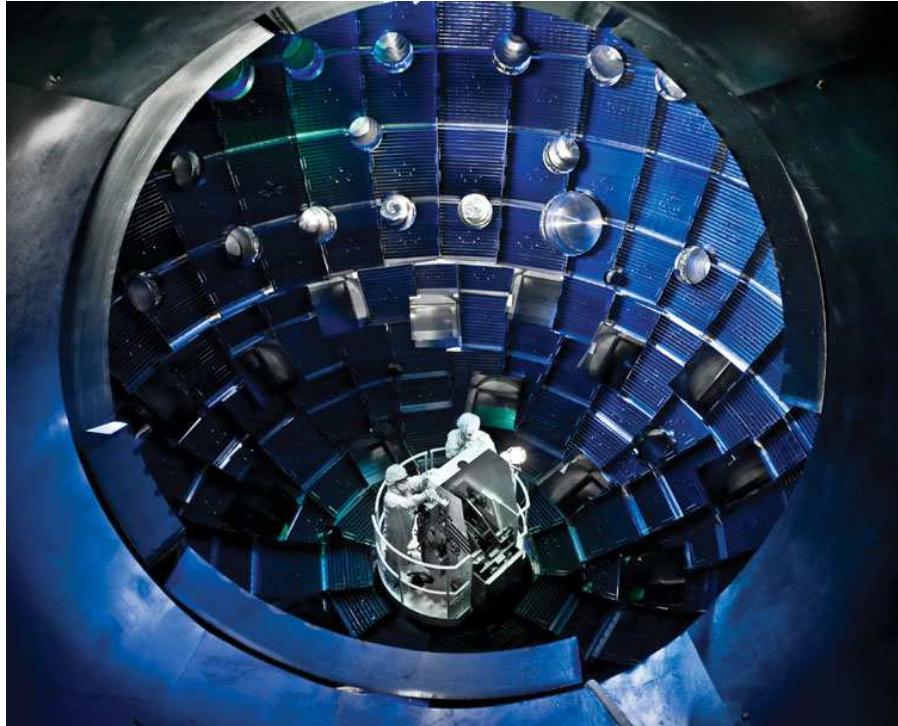
Earth	Fe	3.5Mbar	6000K
Super-Earth	Fe	40Mbar	8000K
Jupiter	H-He	40Mbar	20000K

Dynamic compression

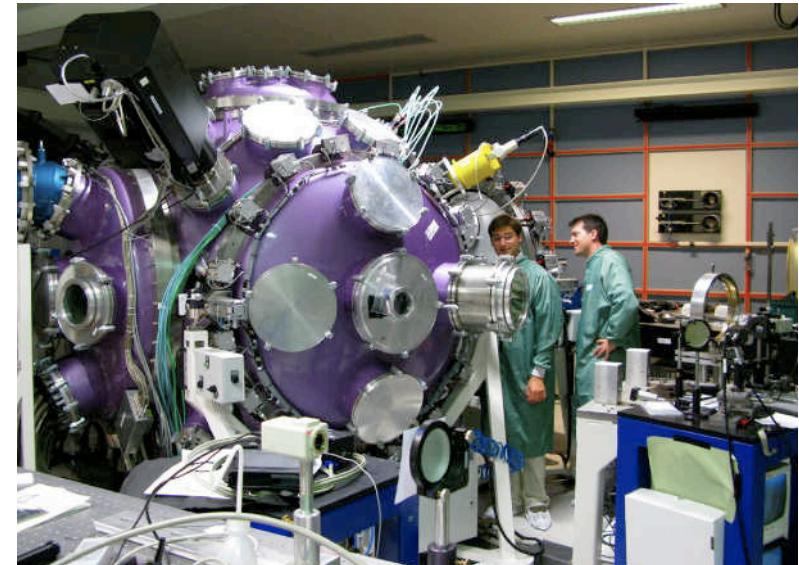


Alternative dynamic compressions are allowing to reach extreme (ρ, T) conditions equivalent to those found in the Earth's core and exoplanets.

High Energy Lasers



NIF (USA) - MJ

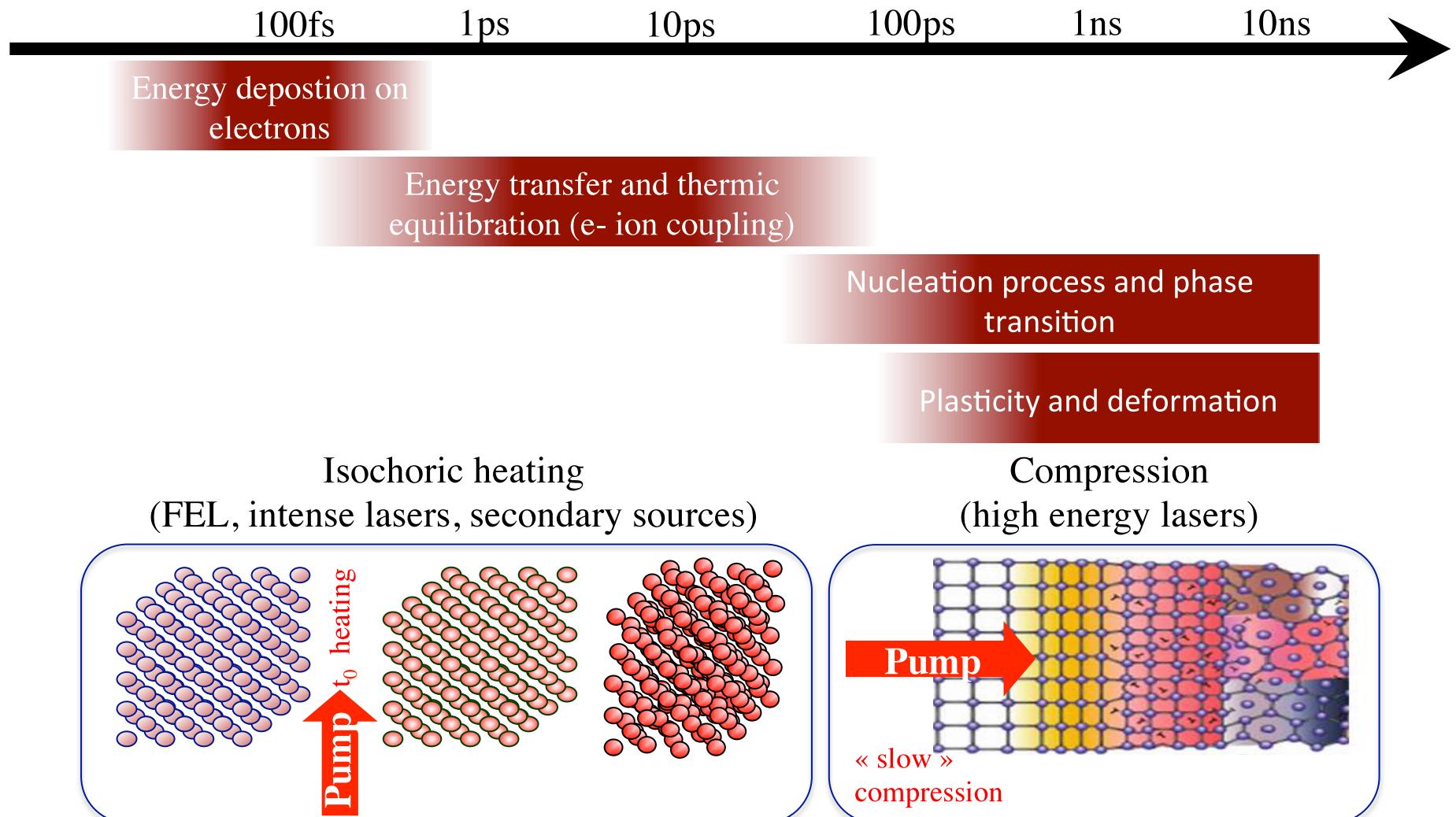


LULI (Paris France) - kJ

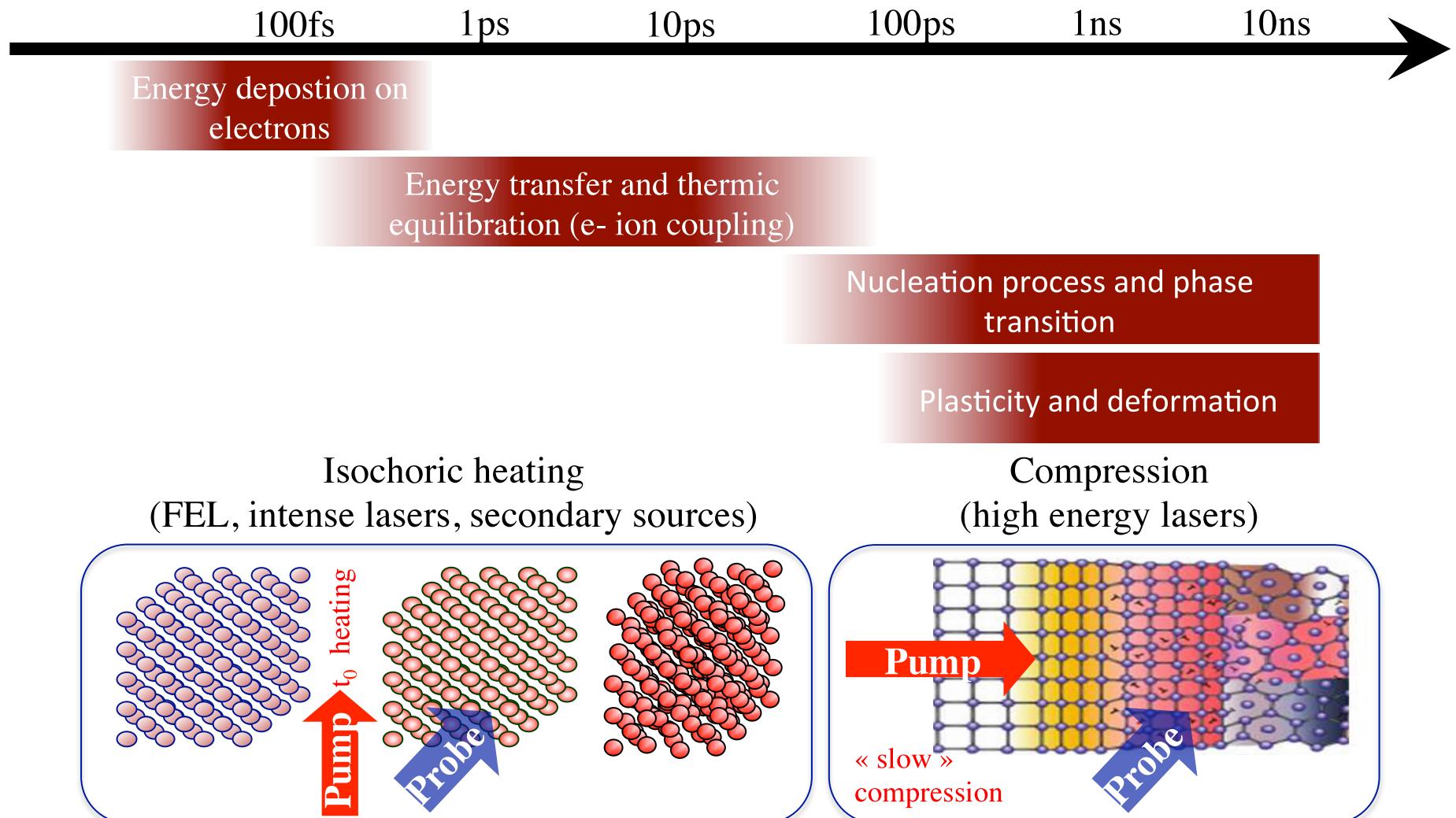


MEC LCLS (USA) – 50J

WDM in laboratory : Time scales



WDM in laboratory : Time scales



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2/ Ultrafast X-ray diagnostics for WDM

- **X-ray Scattering**
- **X-ray absorption**

3/ WDM through its time scales

- Ultrafast heating
- Thermal equilibration
- Phase transitions and phase diagrams
- Shock deformation

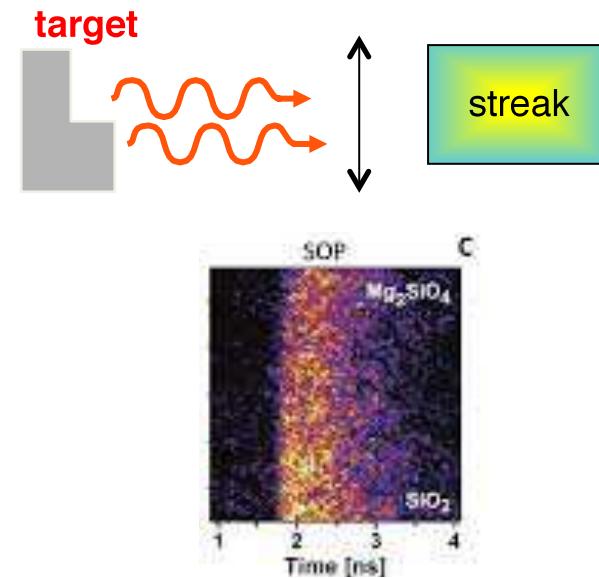
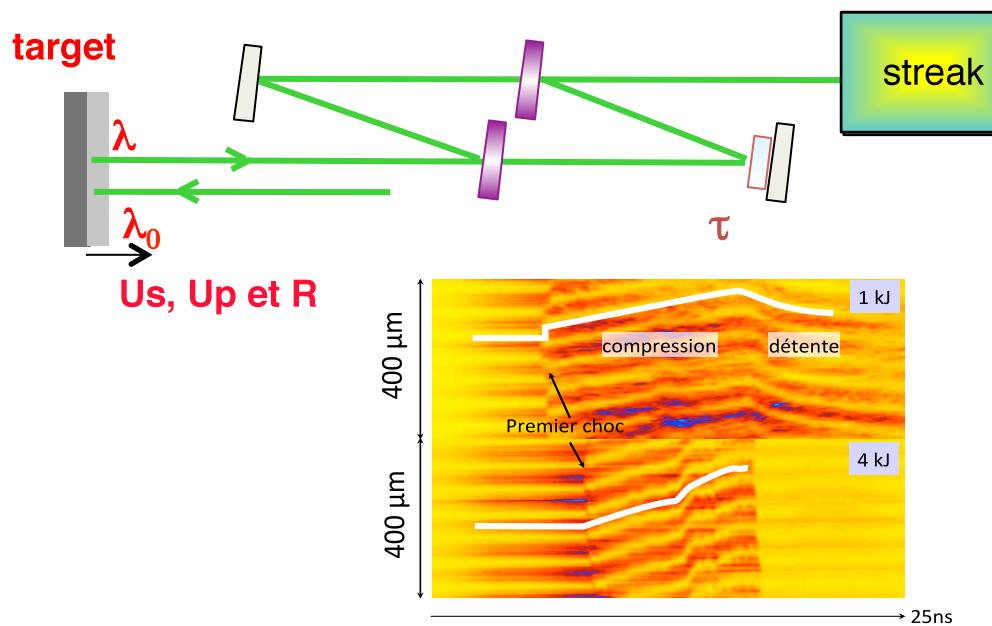
4/ Conclusion & Perspectives

« Traditional » diagnostics

Traditional diagnostics for WDM

- Pyrometry (SOP)
- Surface movements (VISAR and FDI)
- Reflectivity
- Arrival times (velocities)
- Recovery
- ...

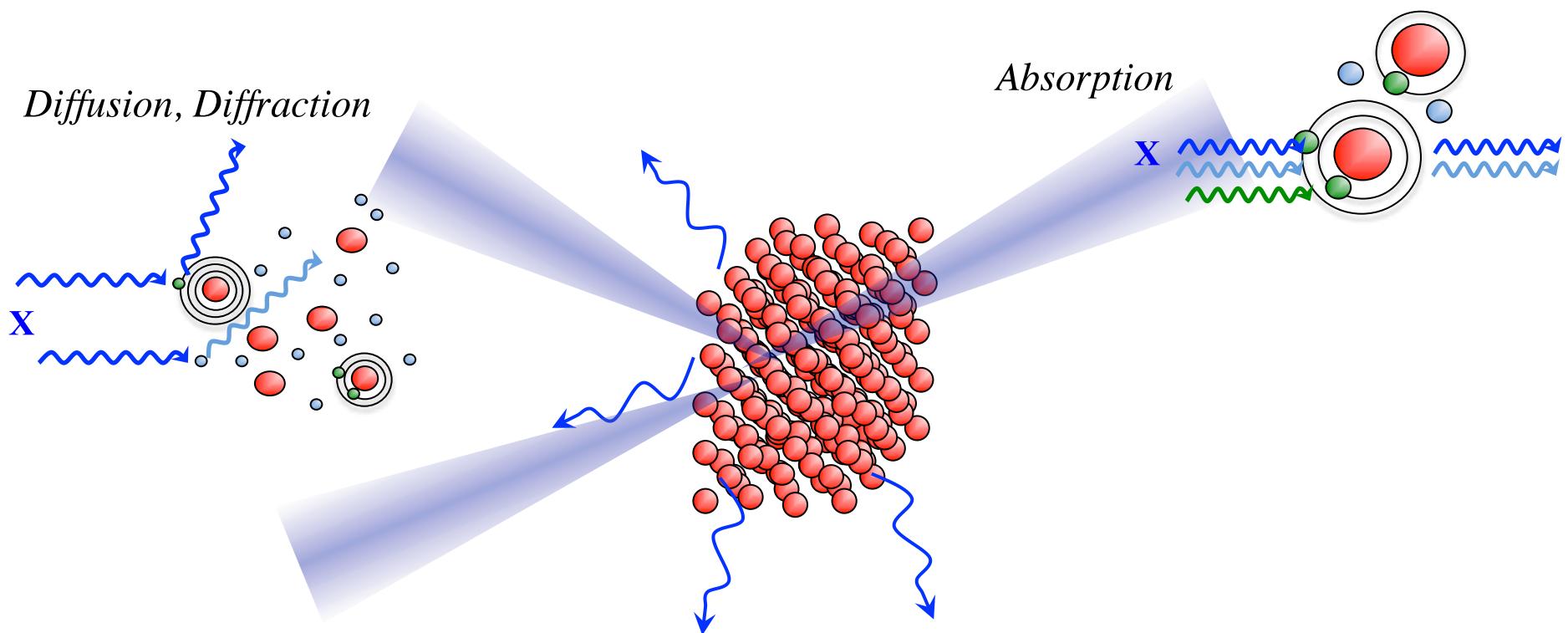
Optical diagnostics are limited to **surface only and do not give structural informations**
→ Last decades developments with plasma based X-ray sources and particules



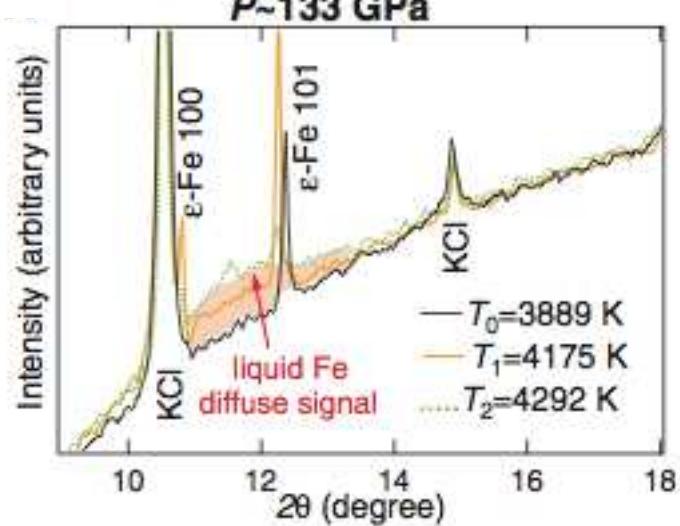
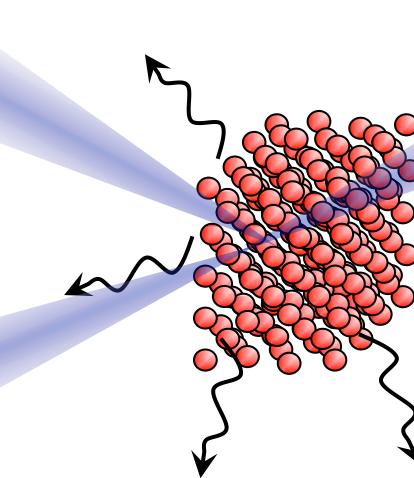
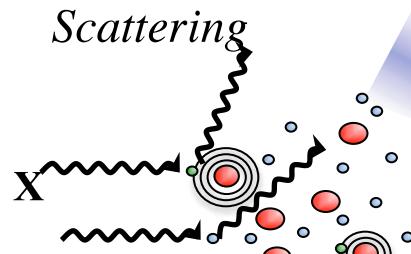
X-rays to probe WDM

X-ray diagnostics to probe in **the bulk volume**

- Access to equation of states (density, temperature)
- Access to microscopic structural informations
- Access to electronic and atomic populations

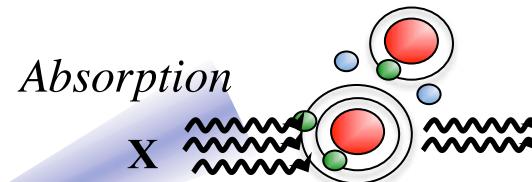


X-ray diagnostics for WDM

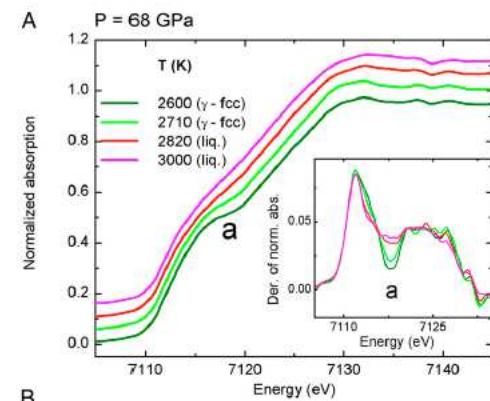


Anzellini et al. Science 2014

Single – shot

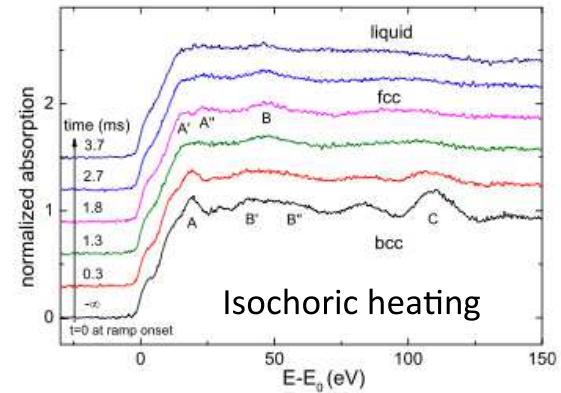


Aquilanti et al. PNAS, 2016

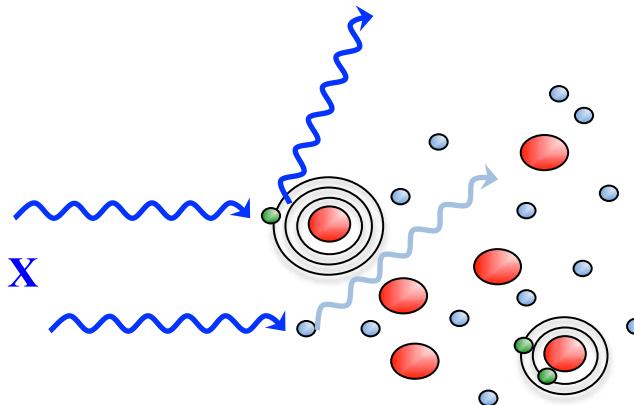


R

Marini et al. JAP, 2014

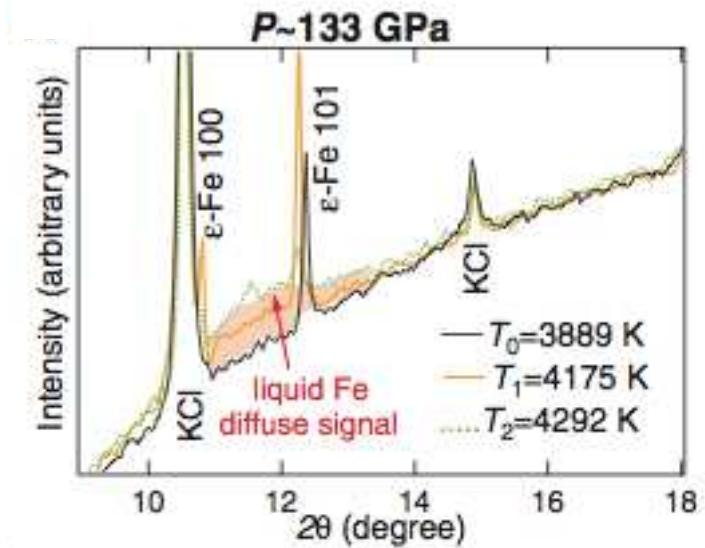
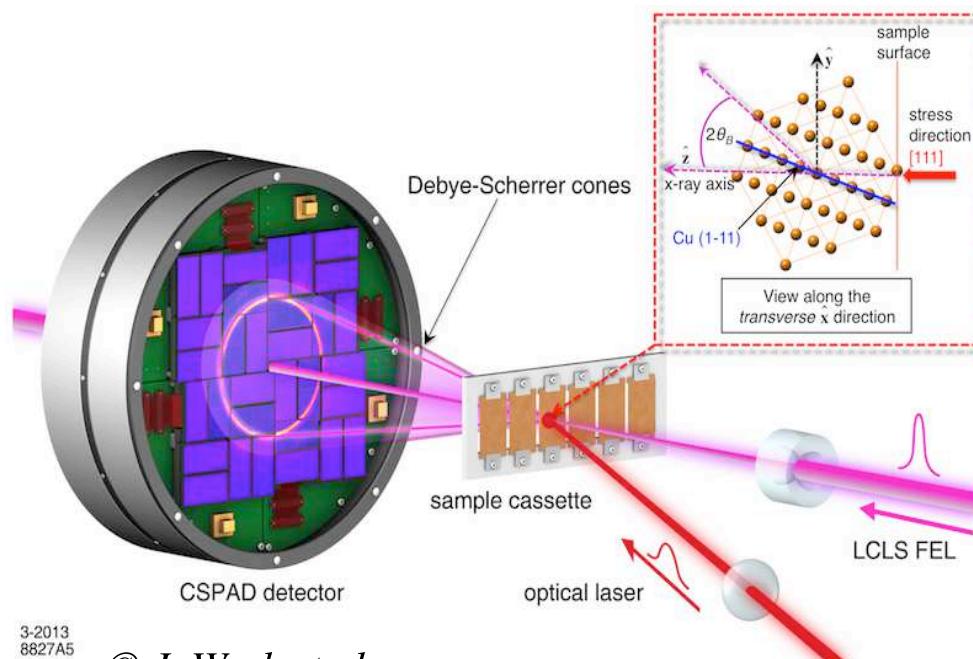


X-ray Scattering



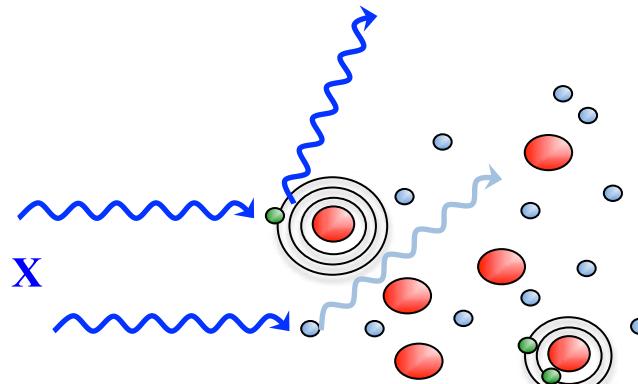
X-ray diffraction resolved in angle
→ structure factor, phases, density,
liquid onset,

$$\sigma_{Scatt}(k, w) = \sigma_{Scatt\ individual\ e-} S(k, w)$$



Anzellini et al. Science 2014

X-ray Scattering

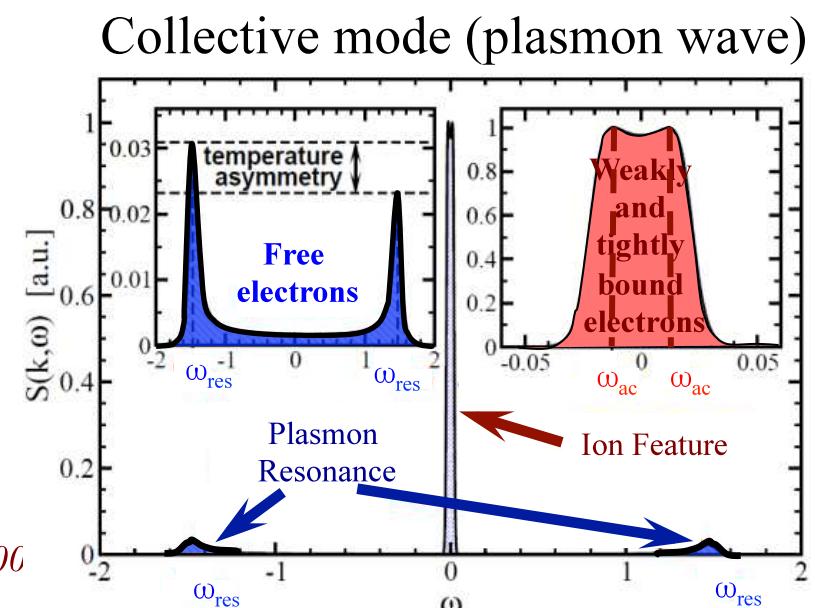


X-ray diffusion resolved spectrally
 - Plasmon \leftrightarrow doppler effect \rightarrow Ne,Te

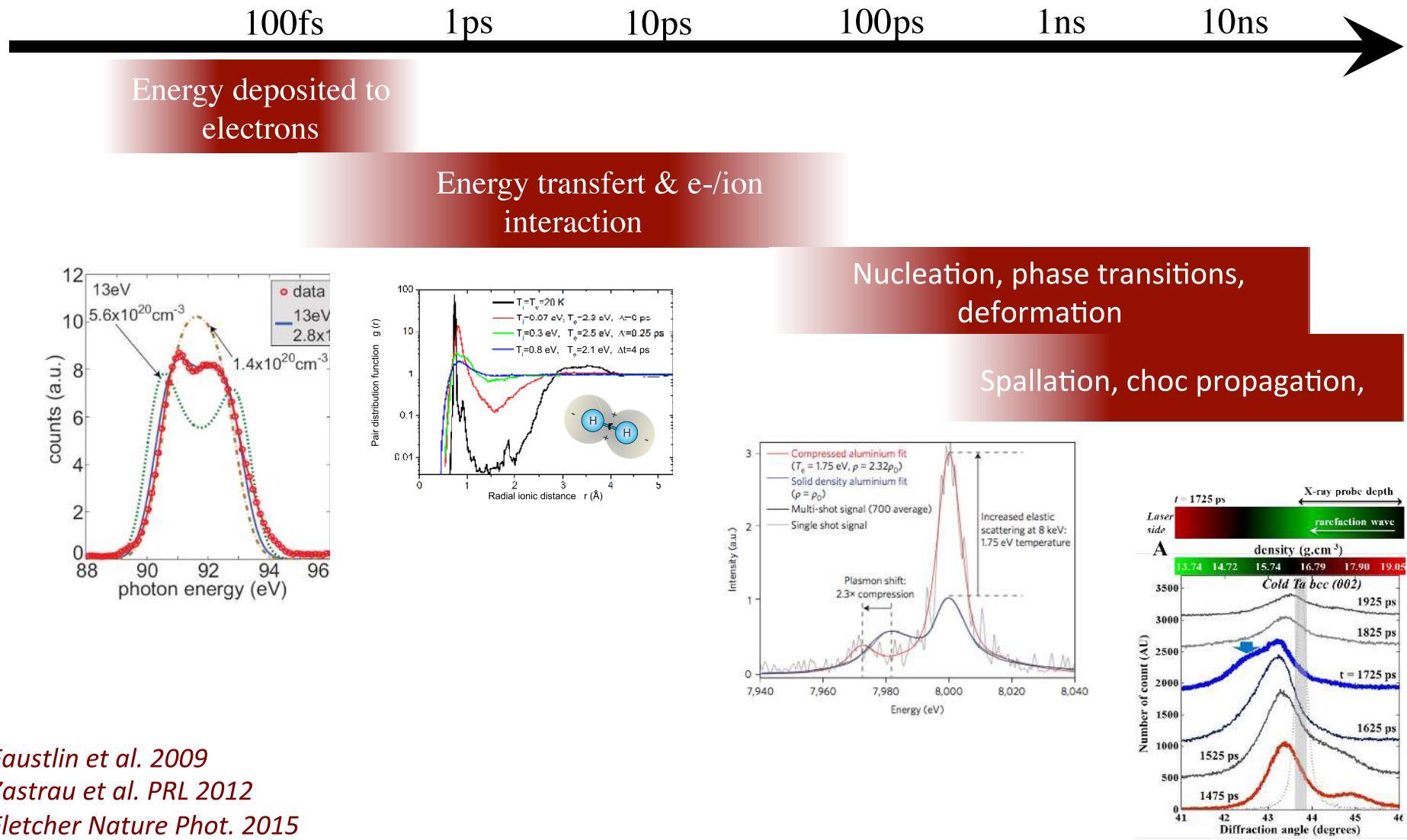
$$S(k, \omega) = Z_f S_{ee}^0(k, \omega) + |f_I(k) + q(k)|^2 S_{ii}(k, \omega) + Z_c \int S_c(k, \omega - \omega') S_S(k, \omega') d\omega'$$

e- feature Ion feature Bound - free

Low scattering cross section
 → Brilliant sources : 10^{12-13} ph/pulse
 Small spectral shift
 → fine Bandwith : <0.5% for seeded FEL



X-ray Scattering at ultrafast time scales



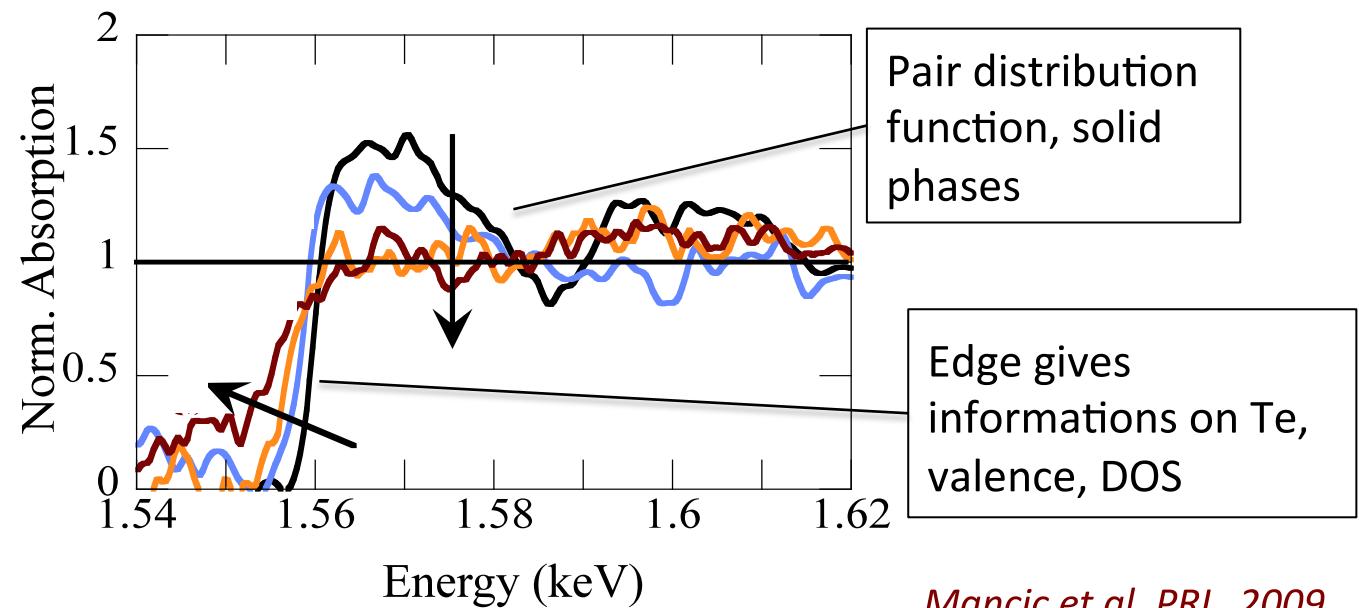
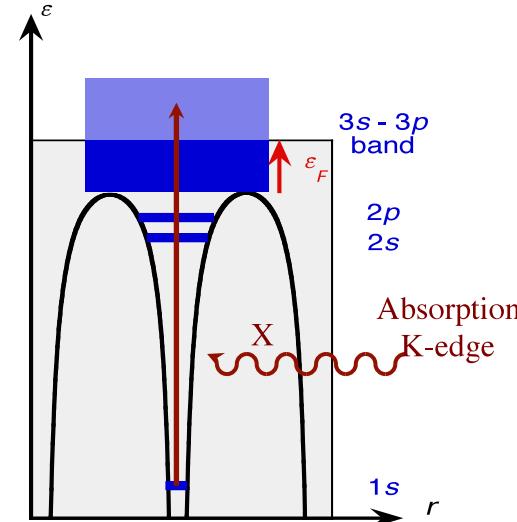
XAS for WDM

XANES-EXAFS gives access to:

- Crystallographic phases
- Melting diagnostic
- Unoccupied DOS (valence e-)
- Band structures
- Hybridation processes
- Temperature (Te, Ti)
- Screening effects ...

Needs simulations:

- Ab-initio
- FEFF



Mancic et al. PRL, 2009

XANES to measure the temperature

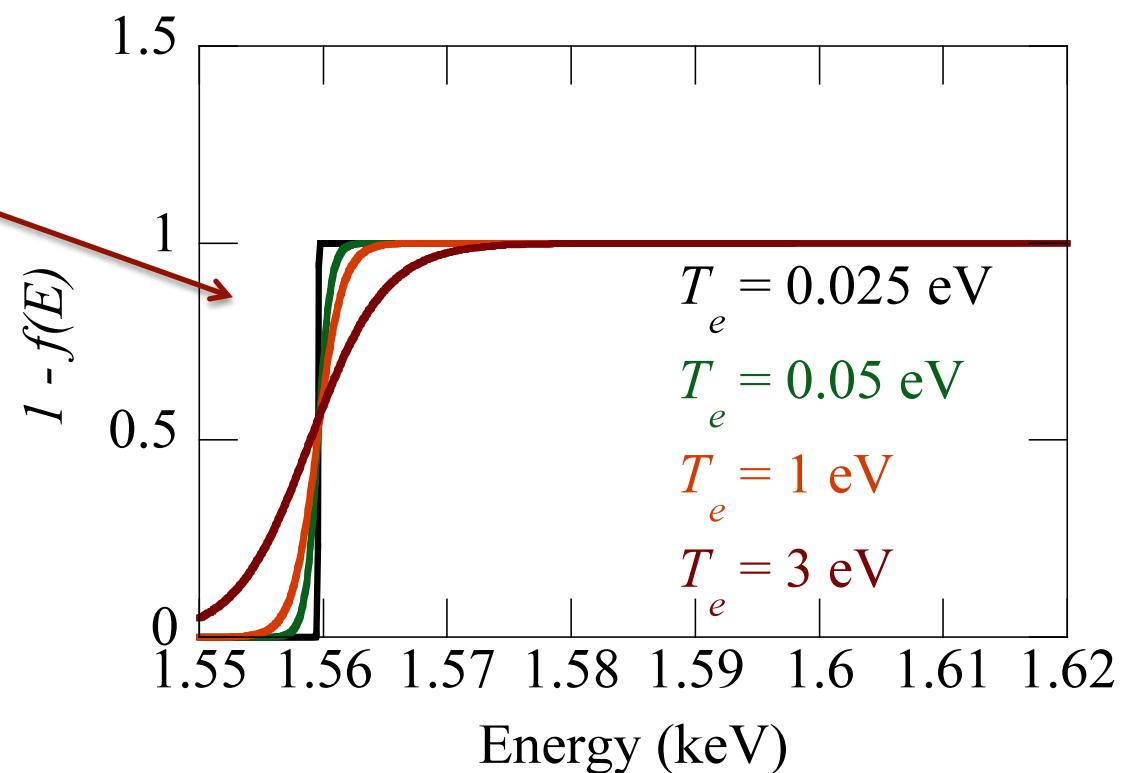
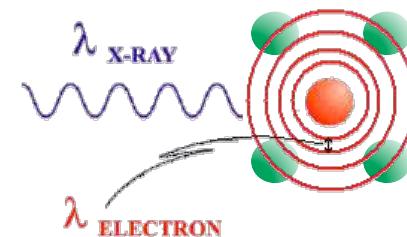
Fermi Golden rule :

$$\sigma_{if}(v) \propto h\nu \left| \langle \phi_f | R | \phi_i \rangle \right|^2 (1 - f(E))$$

Fermi Dirac Function :

$$f(E) = \frac{1}{1 + e^{(E - \mu(T_e))/k_B T_e}}$$

Absorption edge position & slope → generally **T** & **ρ**

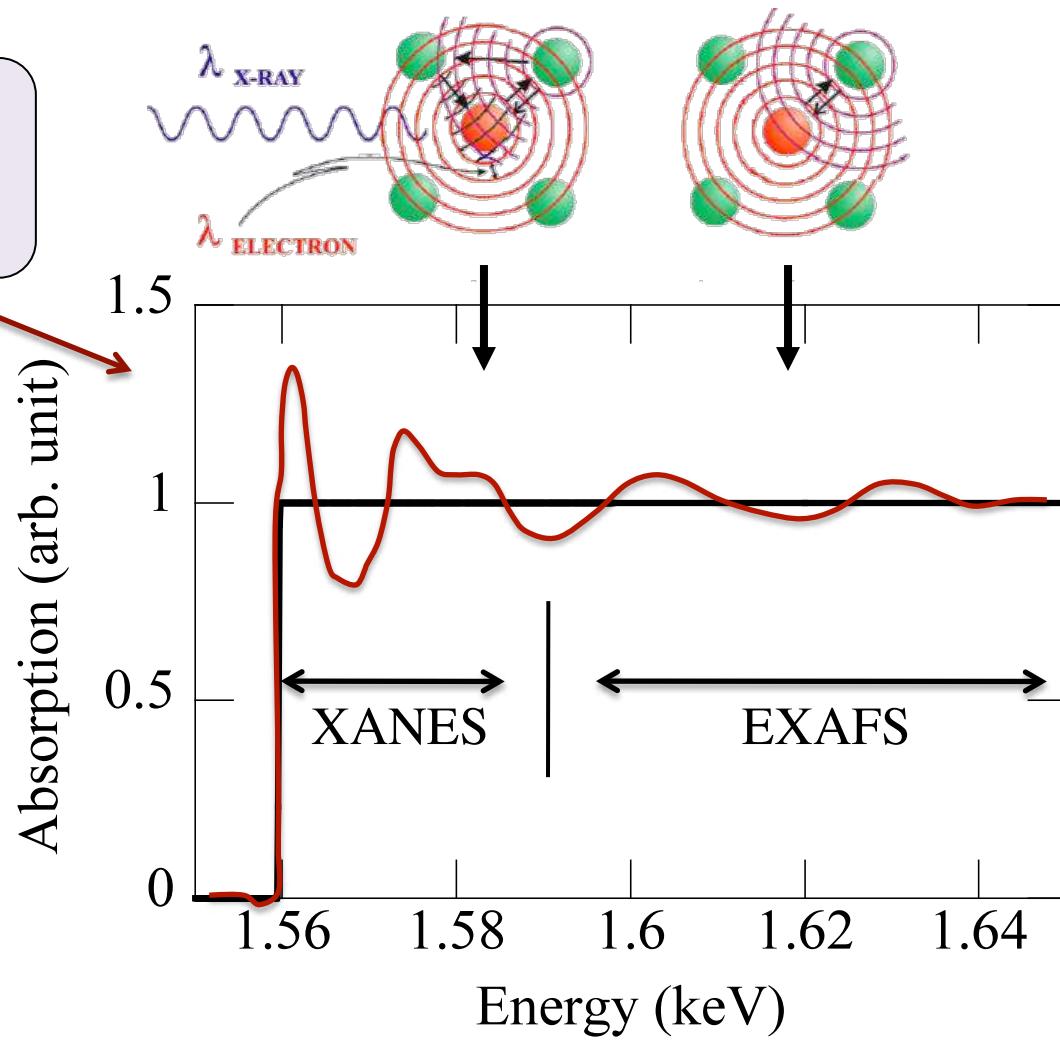


XANES and EXAFS to access DOS and crystallographic phases

Fermi Golden rule :

$$\sigma_{if}(v) \propto h\nu \left| \langle \phi_f | R | \phi_i \rangle \right|^2 (1 - f(E))$$

with $|\phi_f\rangle = |\phi_{free}\rangle + |\phi_{scattering}\rangle$



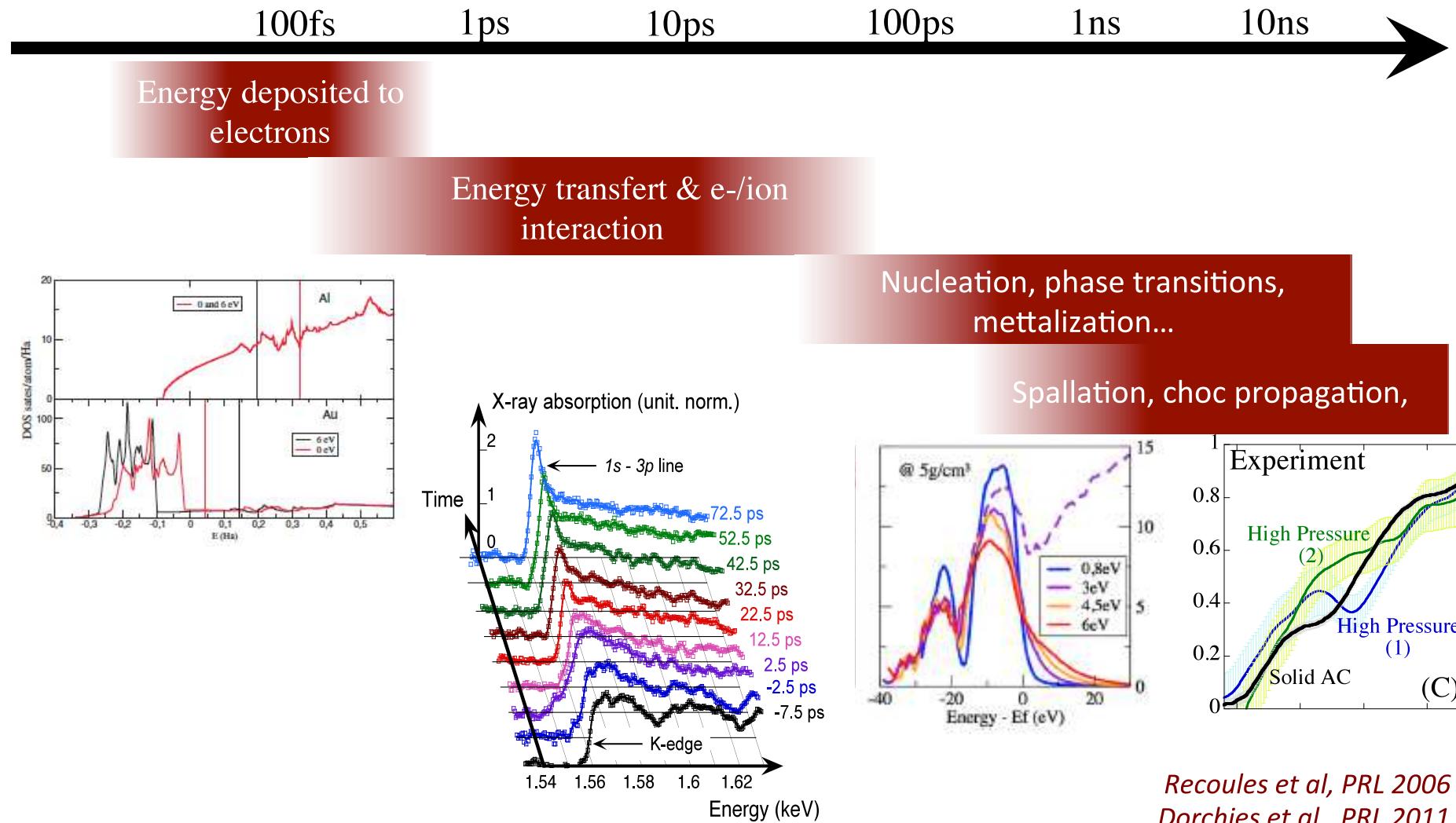
- **XANES** X-ray Absorption Near Edge Spectroscopy

- **EXAFS** Extended X-ray Absorption Fine Structure

→ **local order** (solid phases correlation function...)

→ **valence, DOS**

XAS at ultrafast time scales



*Recoules et al., PRL 2006
Dorchies et al., PRL 2011
Denoeud et al. PRL 2014
Harmand et al. PRB 2015*

Facilities

Lasers

(LULI, CELIA, Apollon, ELI...)

- Isochoric heating
- Compression
- Secondary sources (X, particles)

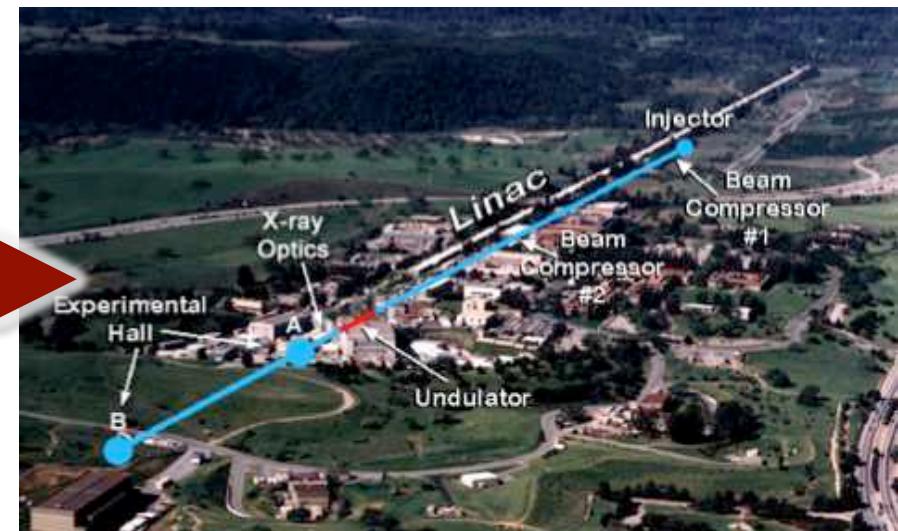
FEL

(LCLS, SACLÀ, PSI, Eu-XFEL...)

- Isochoric Ultrafast heating
- Ultrafast brilliant X-ray or XUV probe



MEC @ LCLS
EH5 @ SACLÀ
HED @ Eu-XFEL
+ PSI, FLASH, FERMI



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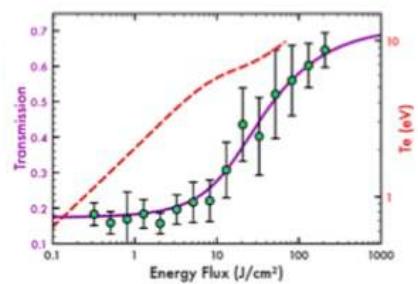
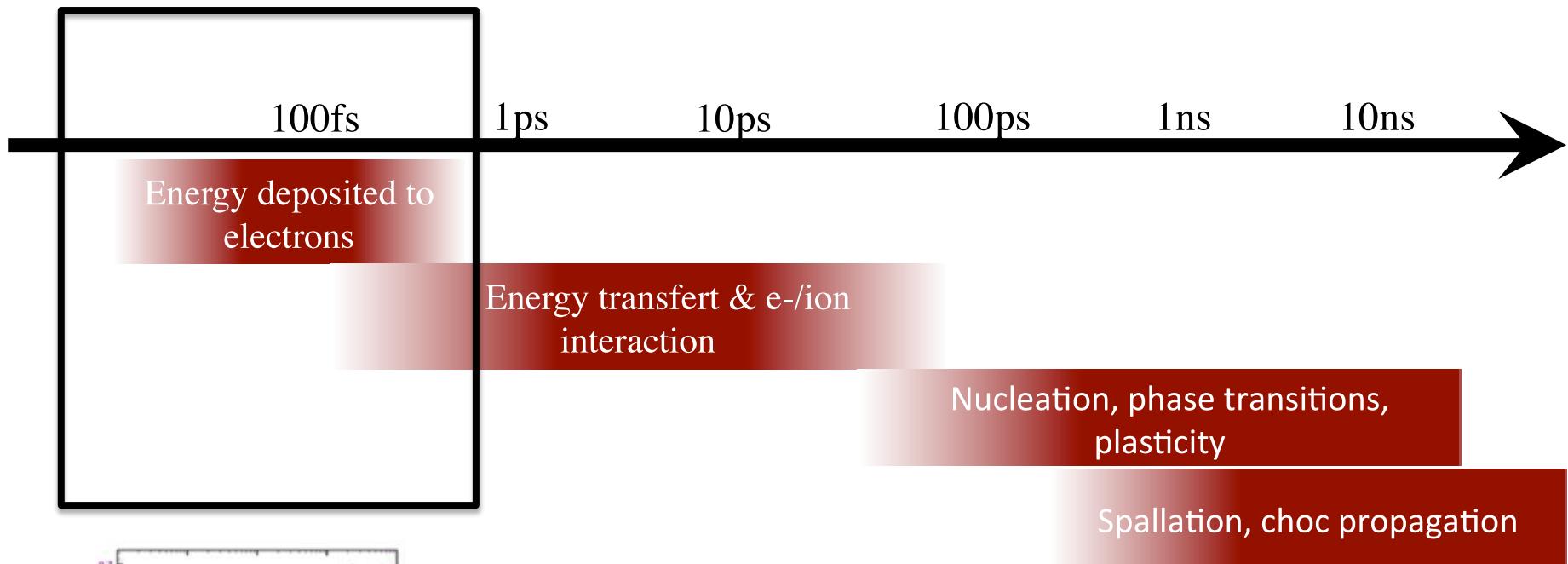
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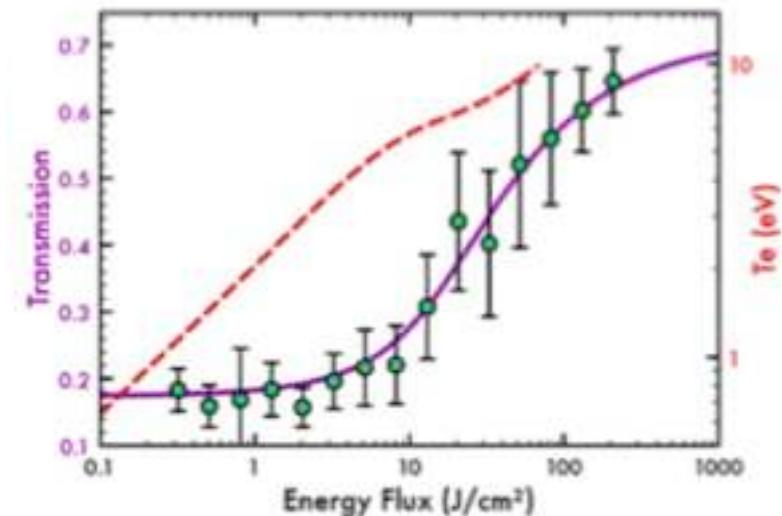
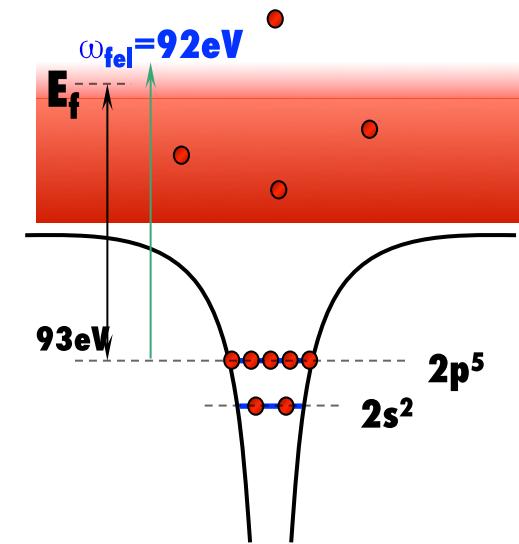
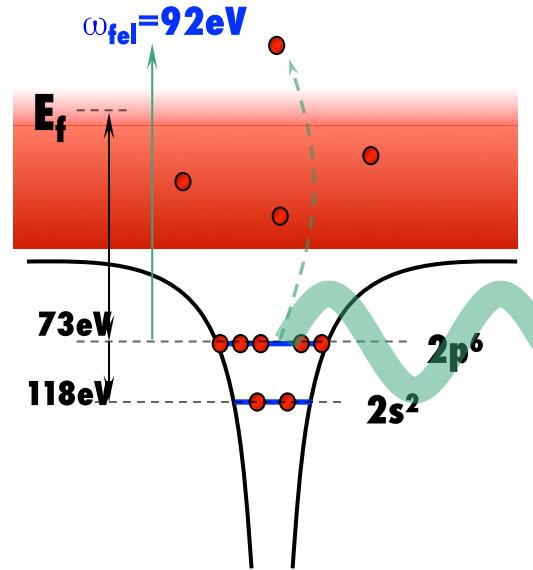
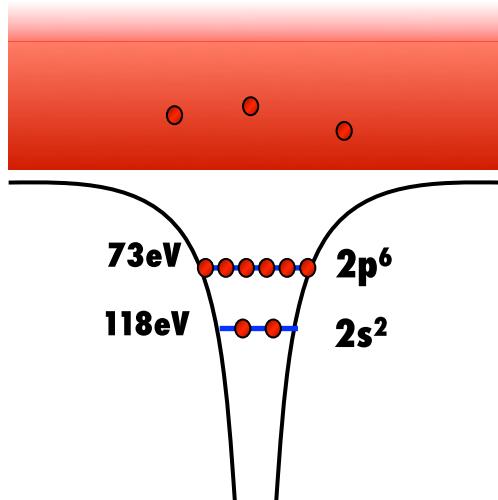
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- Ultrafast heating**
- Thermal equilibration**
- Phase transitions and phase diagrams**
- Shock deformation**

4/ Conclusion & Perspectives



FEL heating and Al saturation

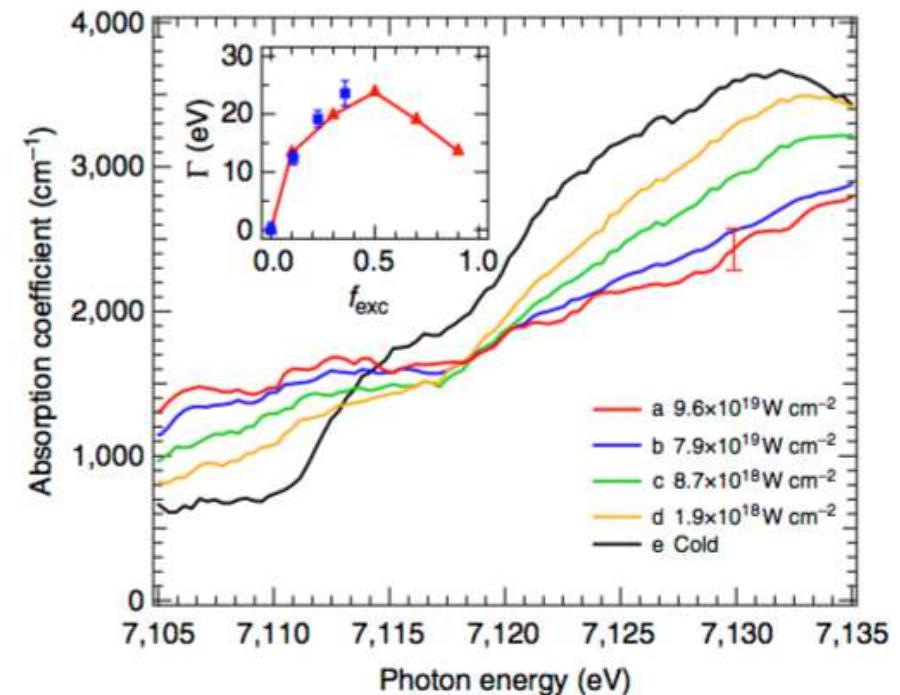
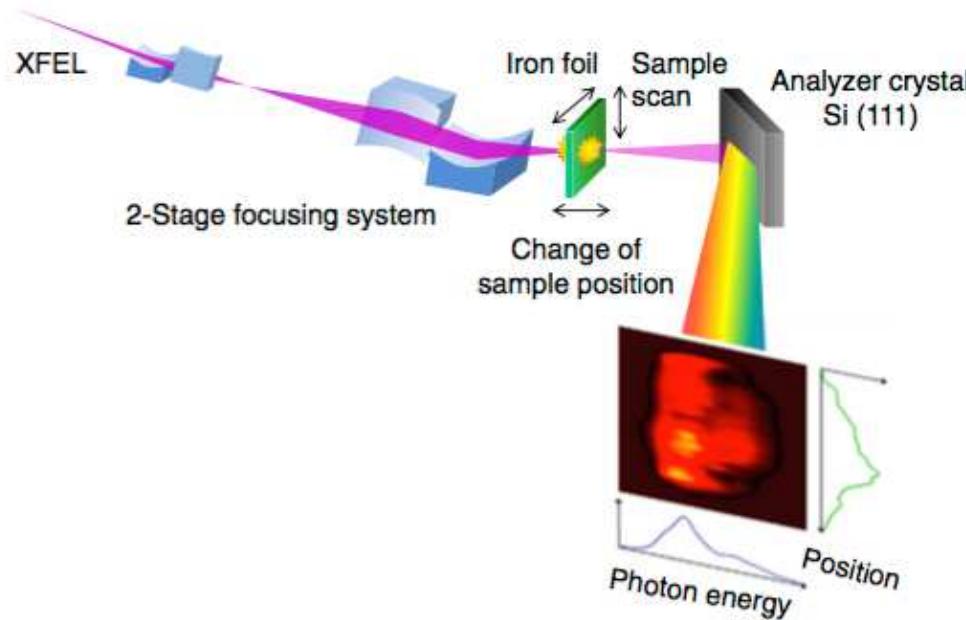


“Transparent Aluminium” (pulse < 50fs)

- Temperature T_e Increases
- Shift of the L-edge
- DOS modification due to L-shell core holes

B. Nagler et al., Nature Phys. 5, 693 (2009).

Saturable absorption of Iron

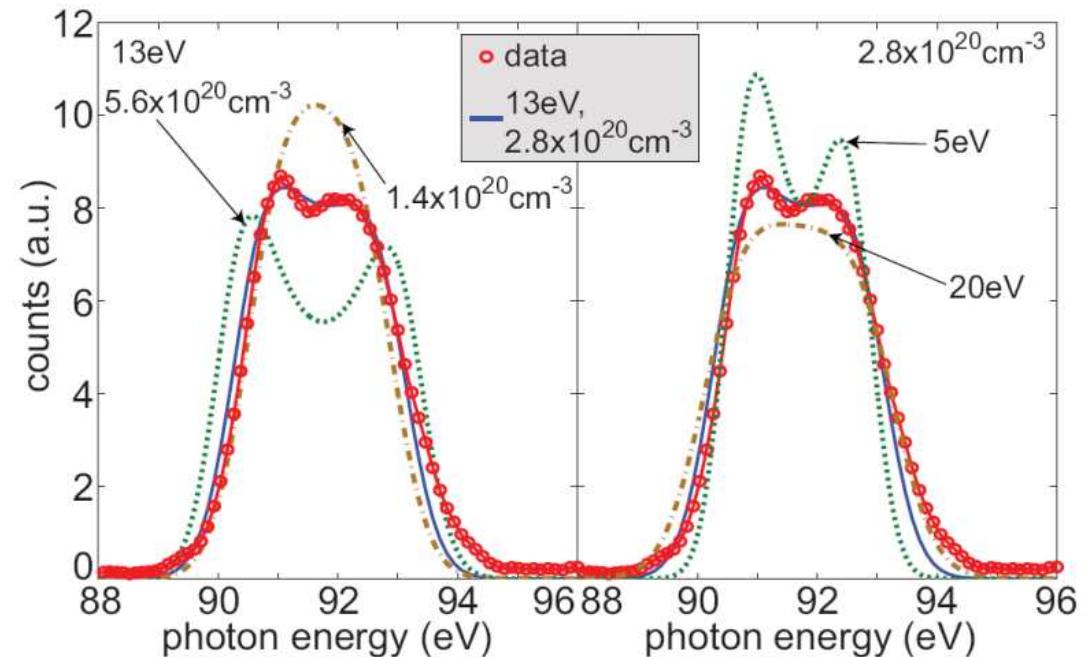
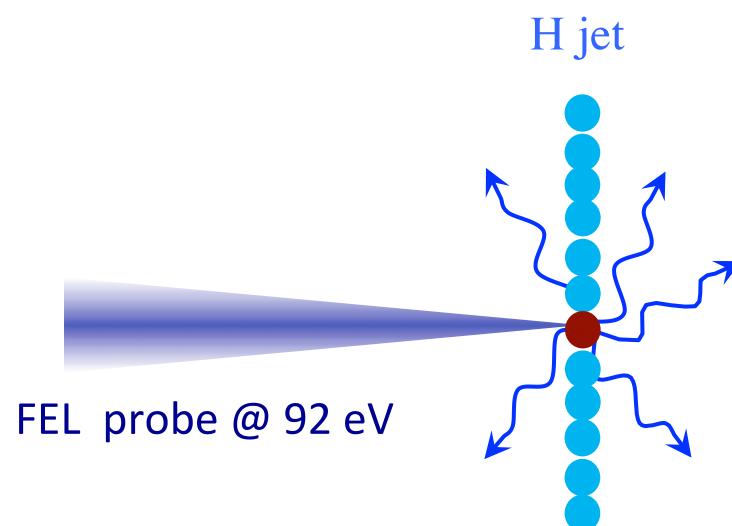


XFEL self-heating et SACLA
(1000 shots accum.)

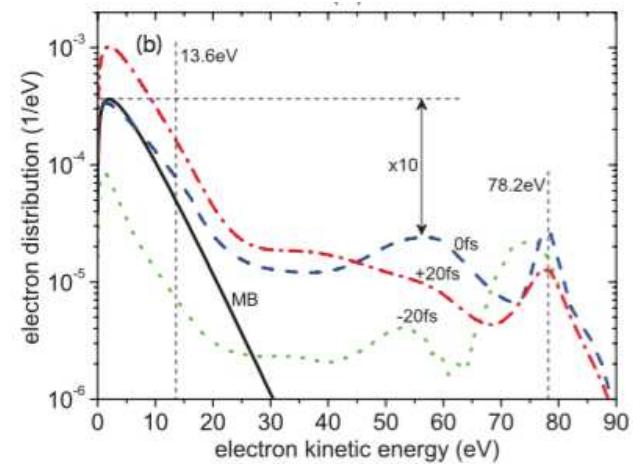
Temperature increases with XFEL
intensity

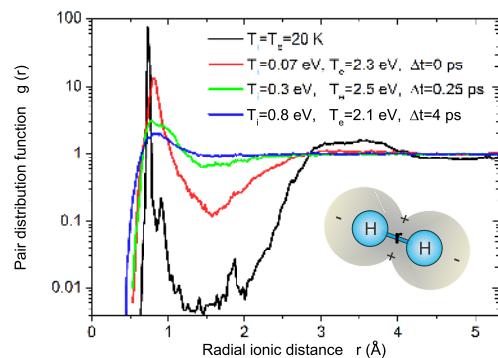
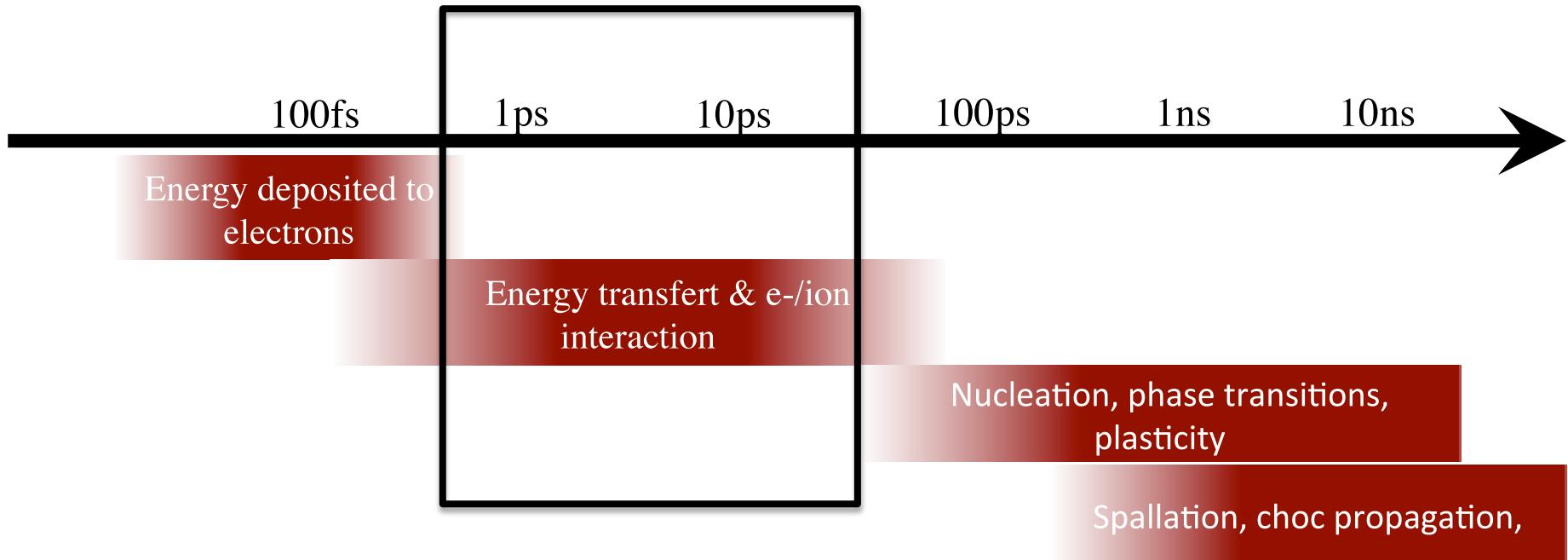
H. Yoneda et al. *Nat. Commun.* 5:5080 (2014).

Self Heating FEL at FLASH

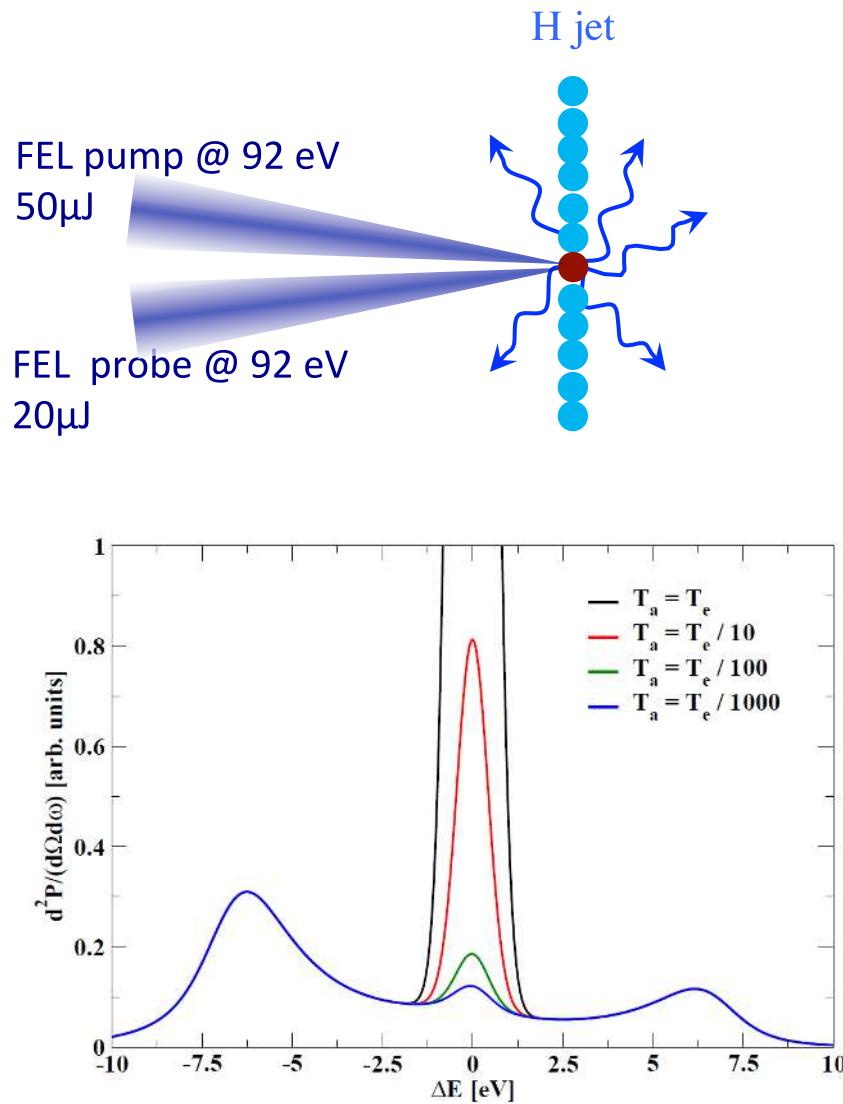


- The FEL pulse heats and probe during its own pulse duration (<100fs)
- Plasmon measurements to extract the Temperature

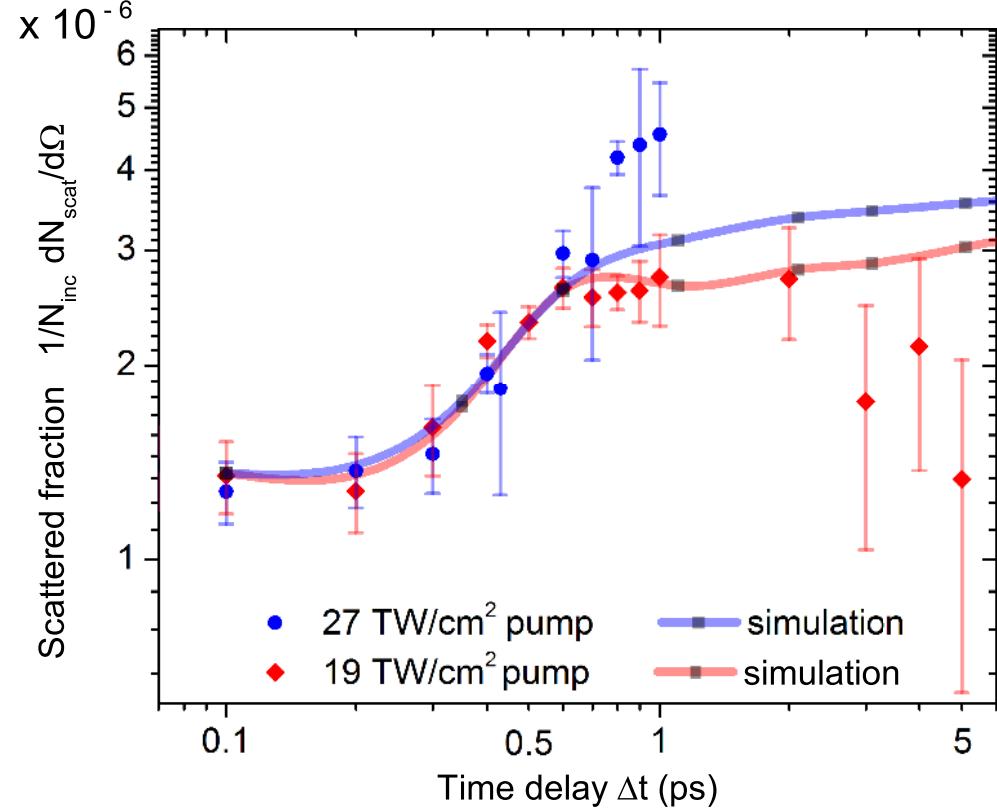




Electron – ion coupling

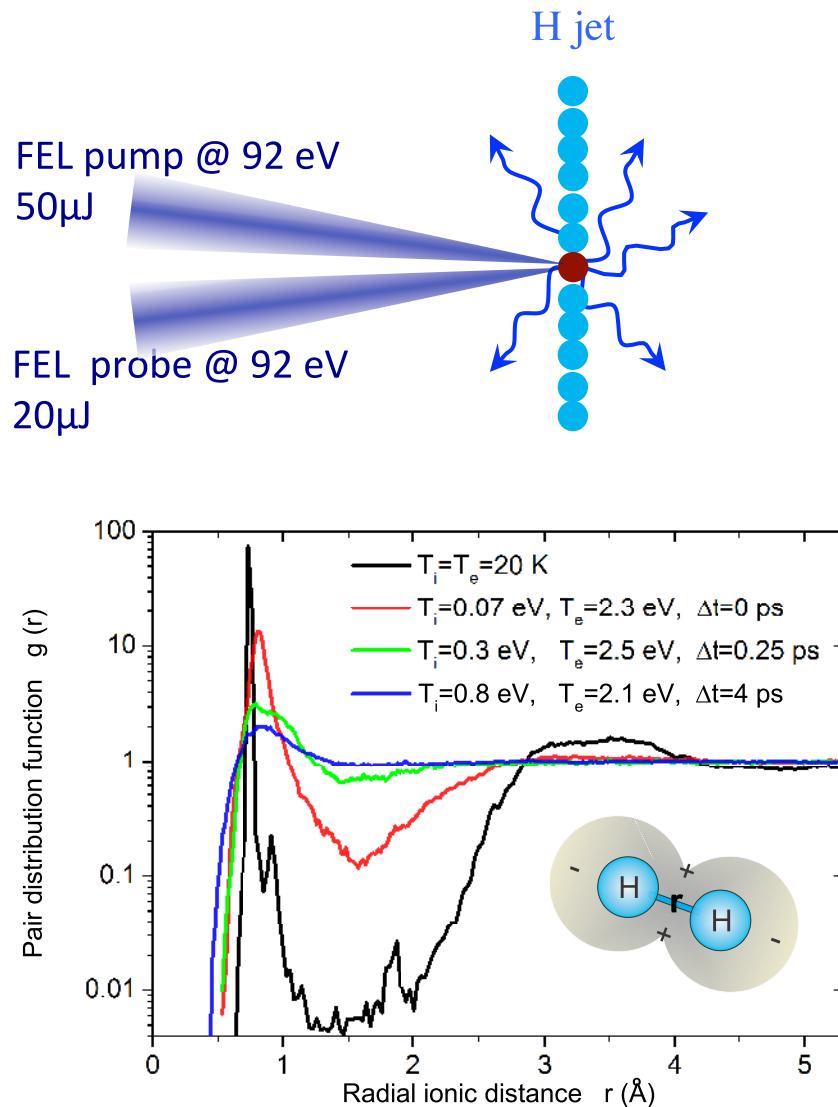


U. Zastrau, P. Sperling, M. Harmand et al, PRL (2014)

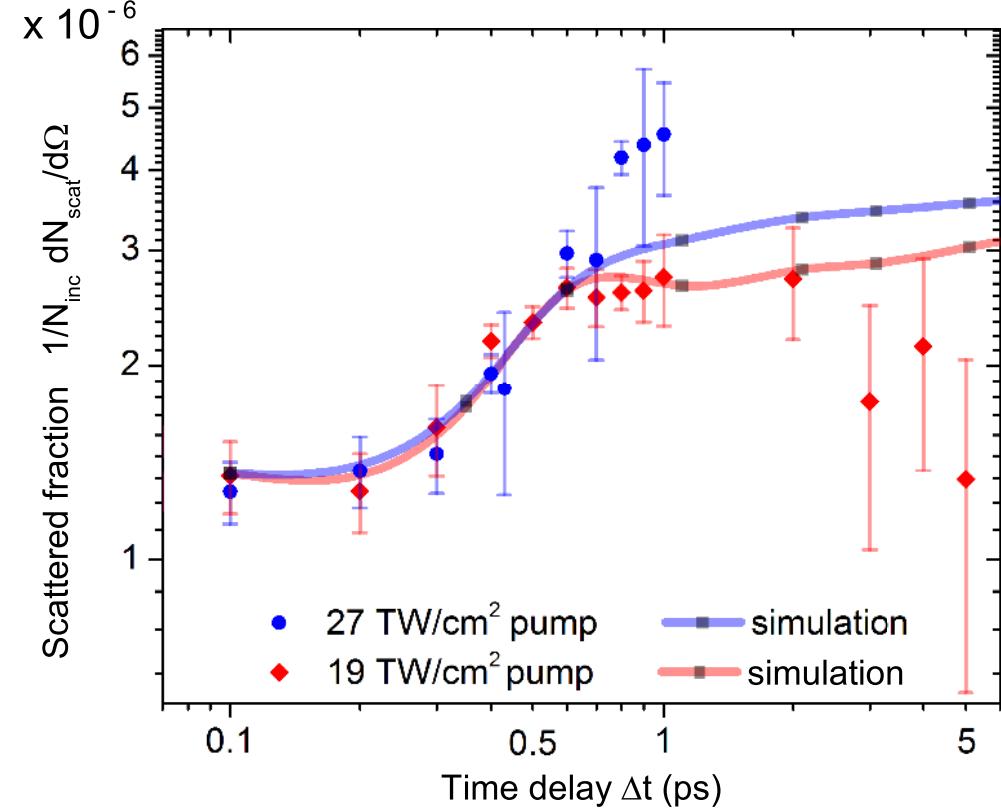


- Hydrodynamic simulations show homogeneous heating ($\sim 11 \mu m$ attenuation length for 92 eV photons)
- Scattering factor S_{ii} increases with T_i

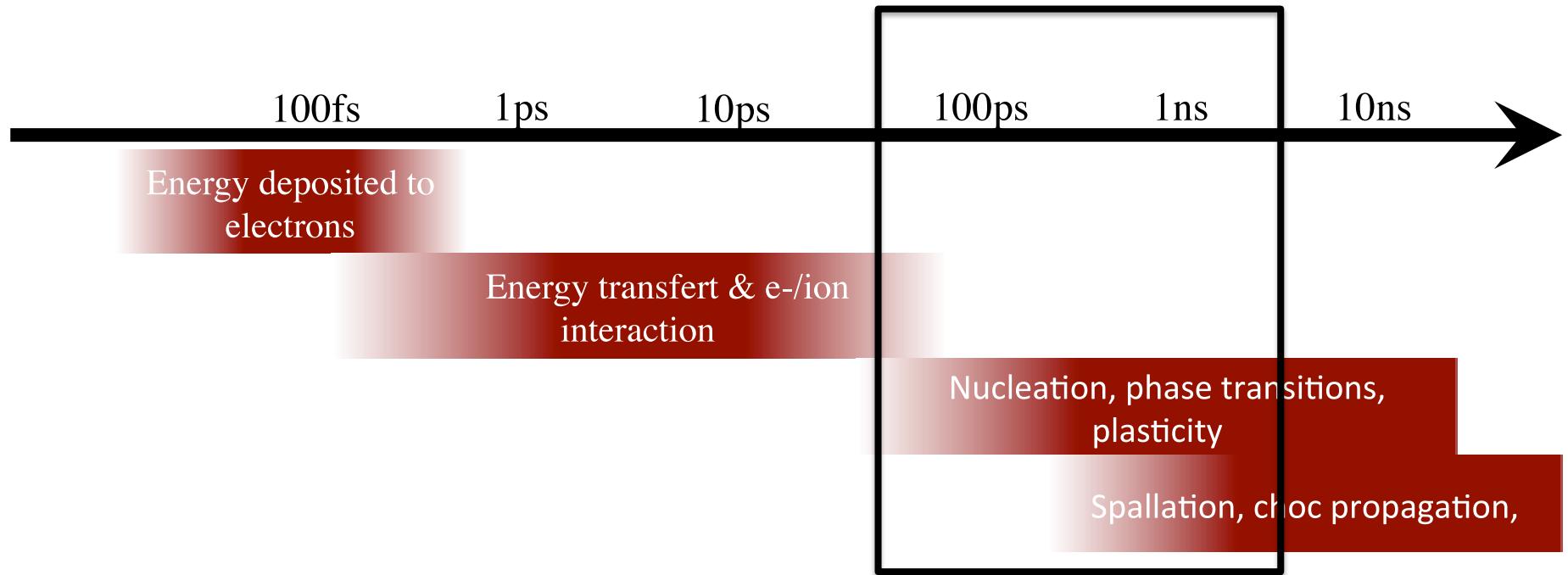
Electron – ion coupling



U. Zastrau, P. Sperling, M. Harmand et al, PRL (2014)

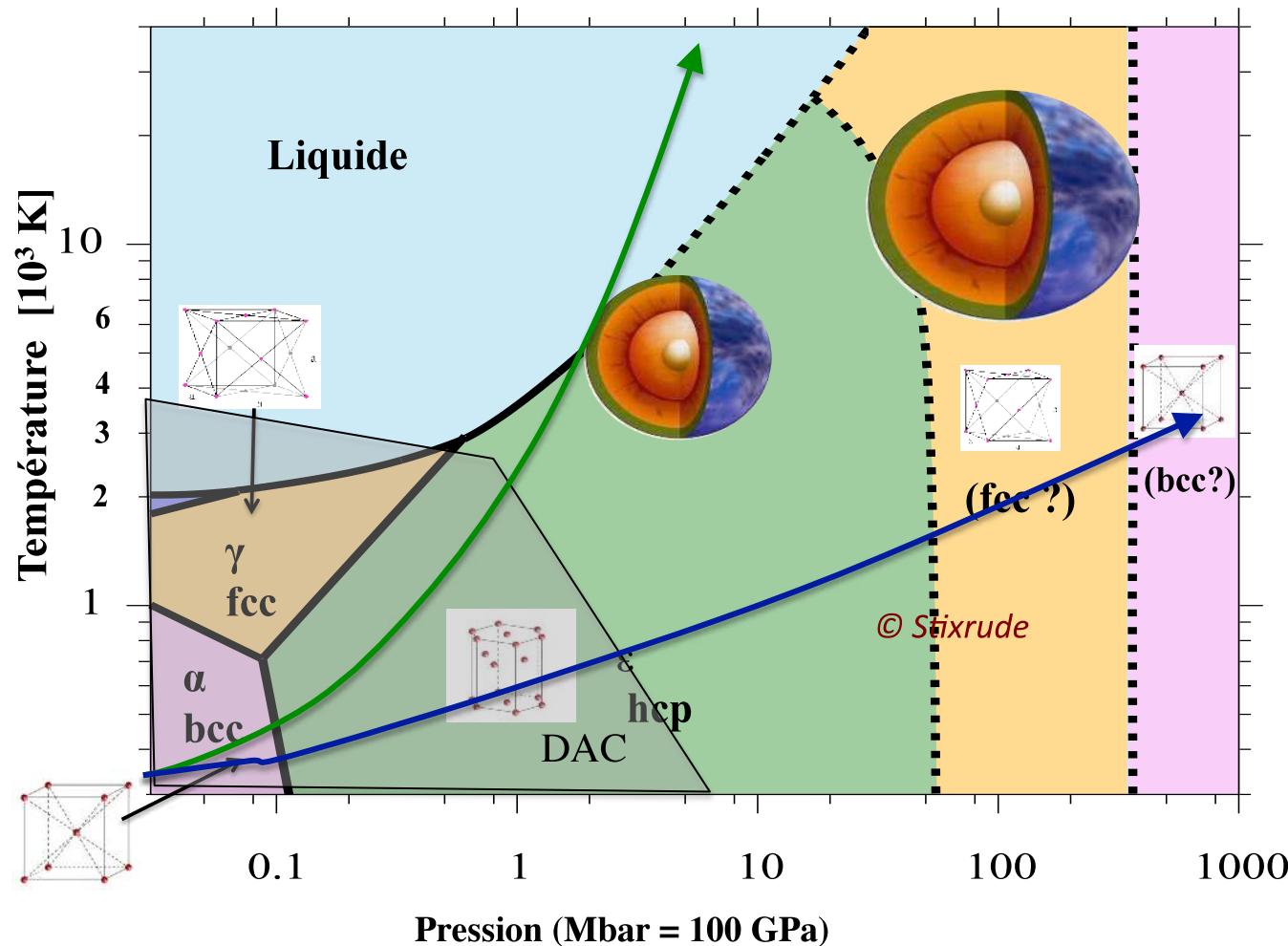


- Energy relaxation from electron to ions in 1ps
- Dissociation of H₂
- Allows to constrain the conductivity of Hydrogen



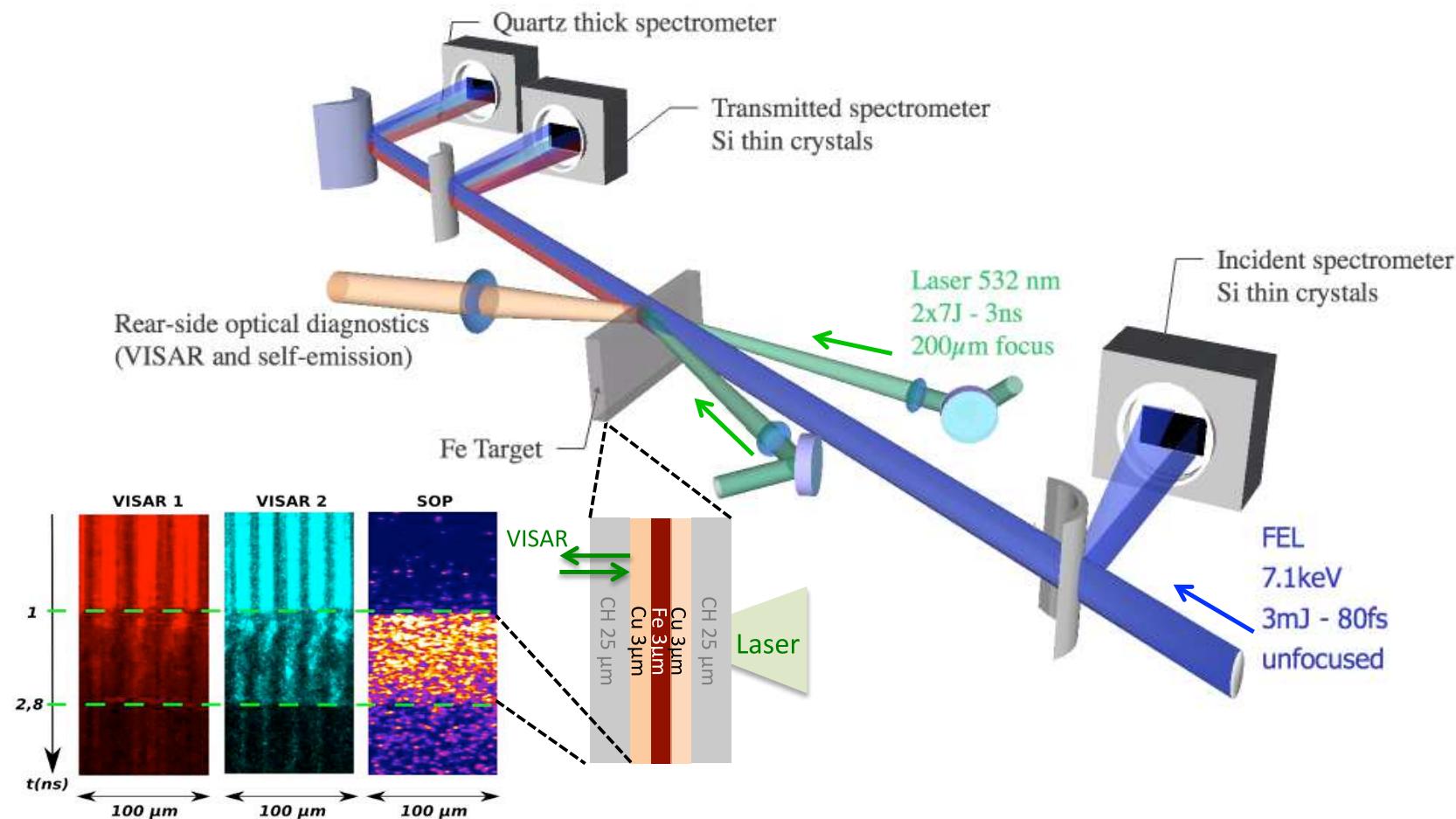
Case of Iron Phase diagram

Fe Phase diagram

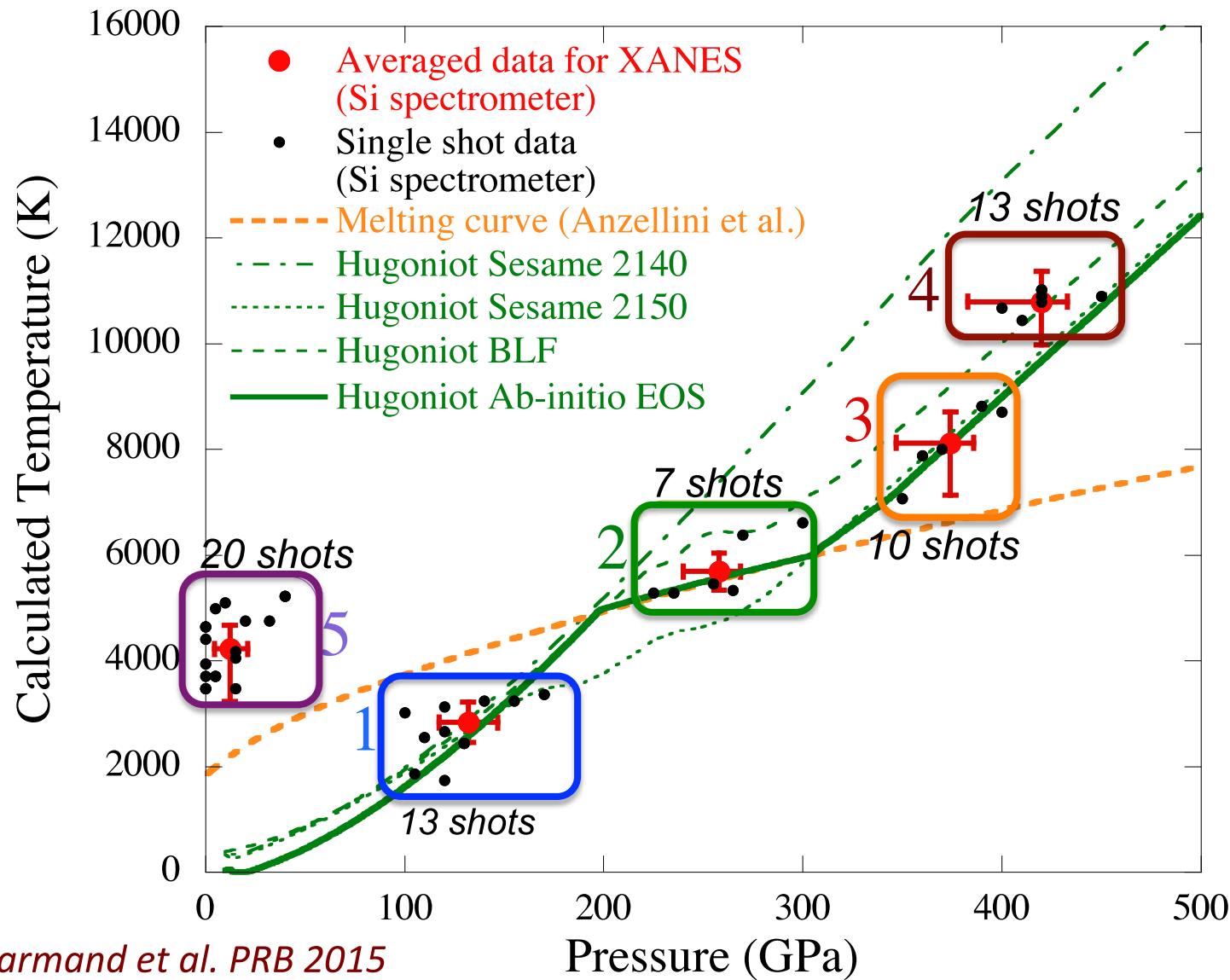


Melting curve of Iron at
far extreme pressures?
Effect of light elements?

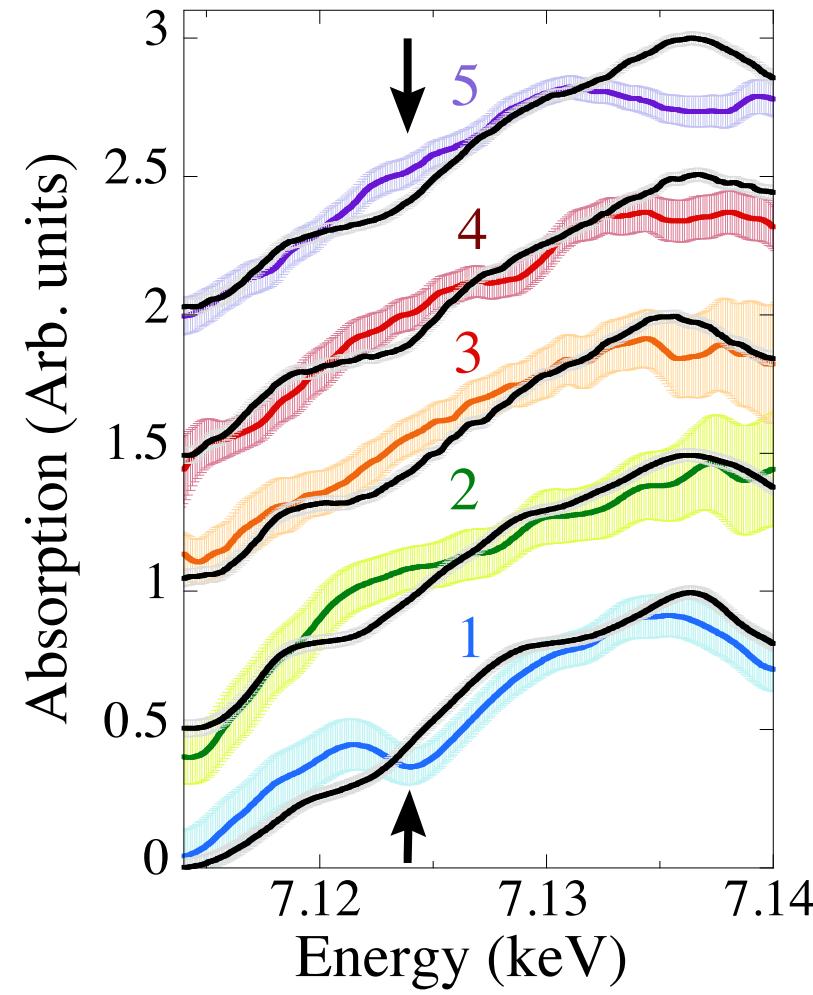
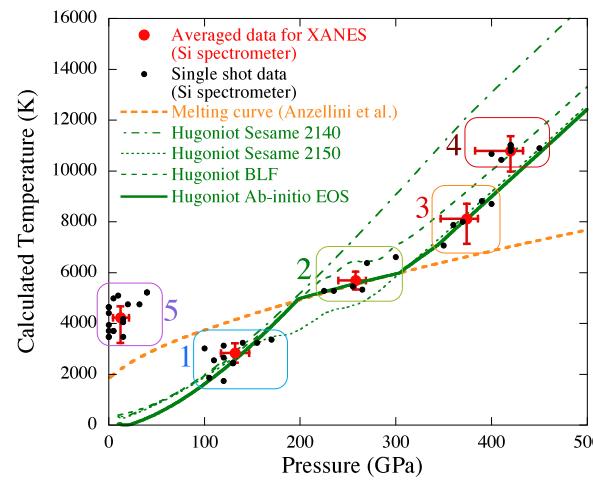
XANES to detect melting at LCLS



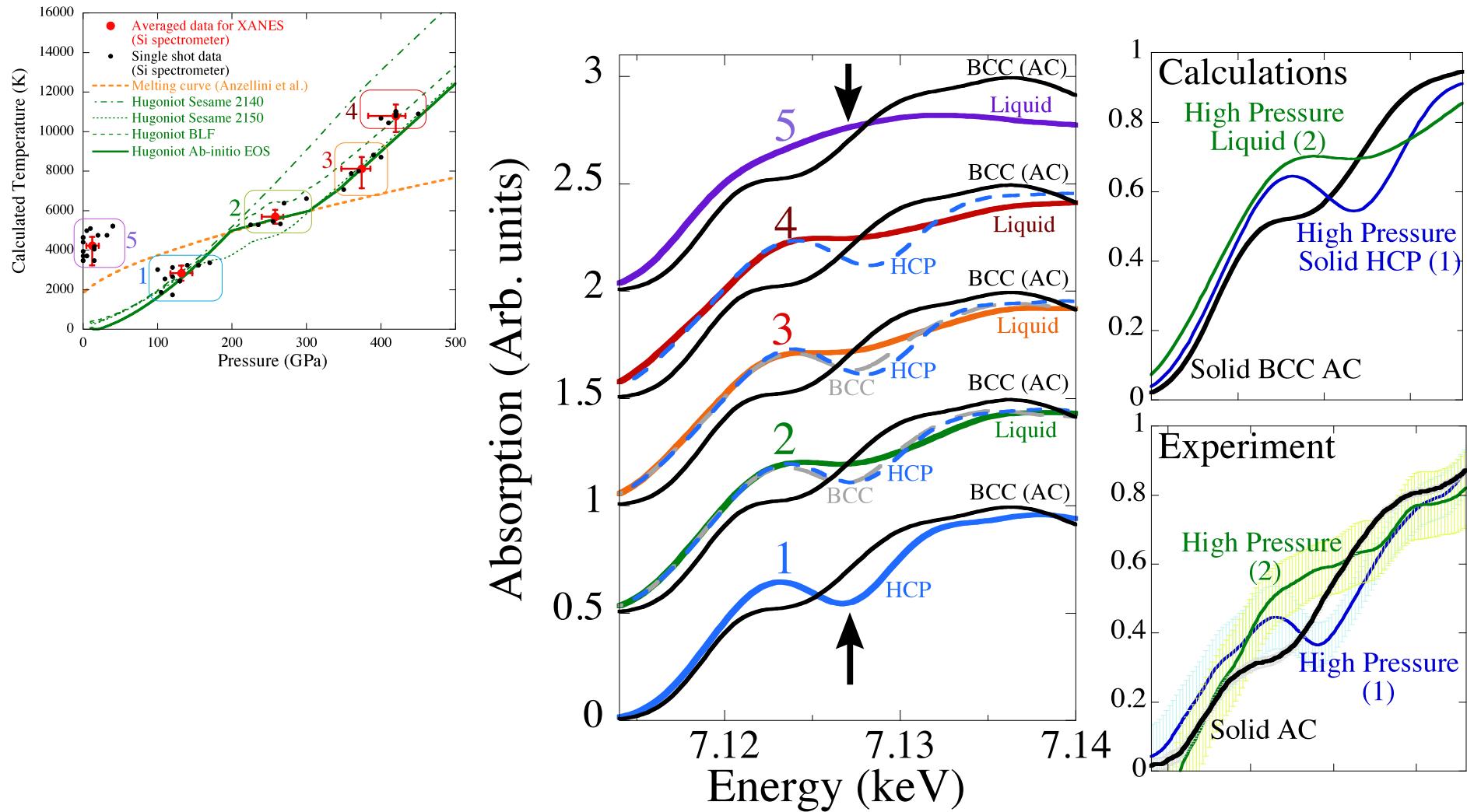
Probed conditions from optical diagnostics (VISAR & SOP)



XANES spectra at 5 P-T conditions



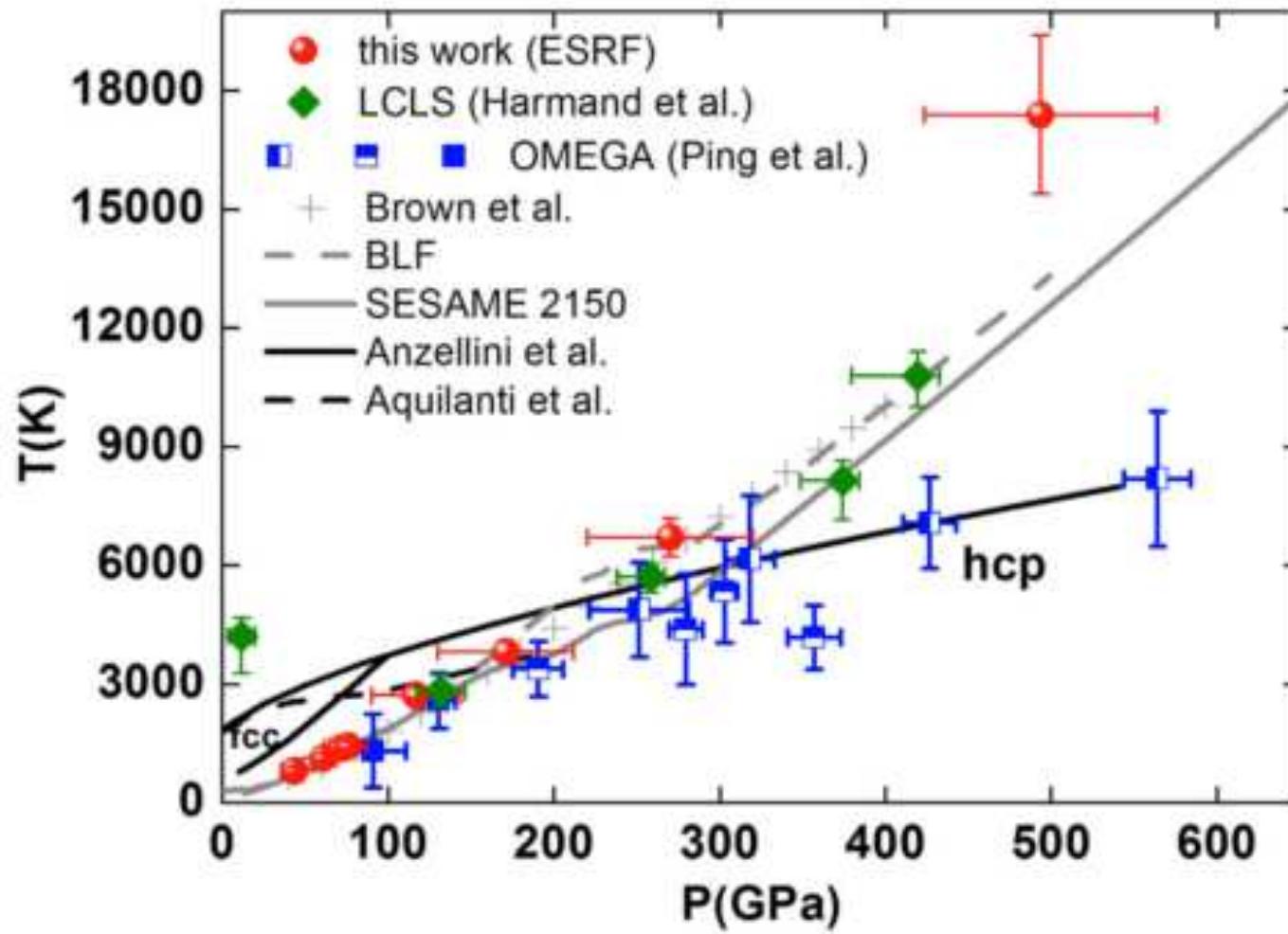
XANES spectra in good agreement with ab-initio simulations



S. Mazevet et al., PRB 2014

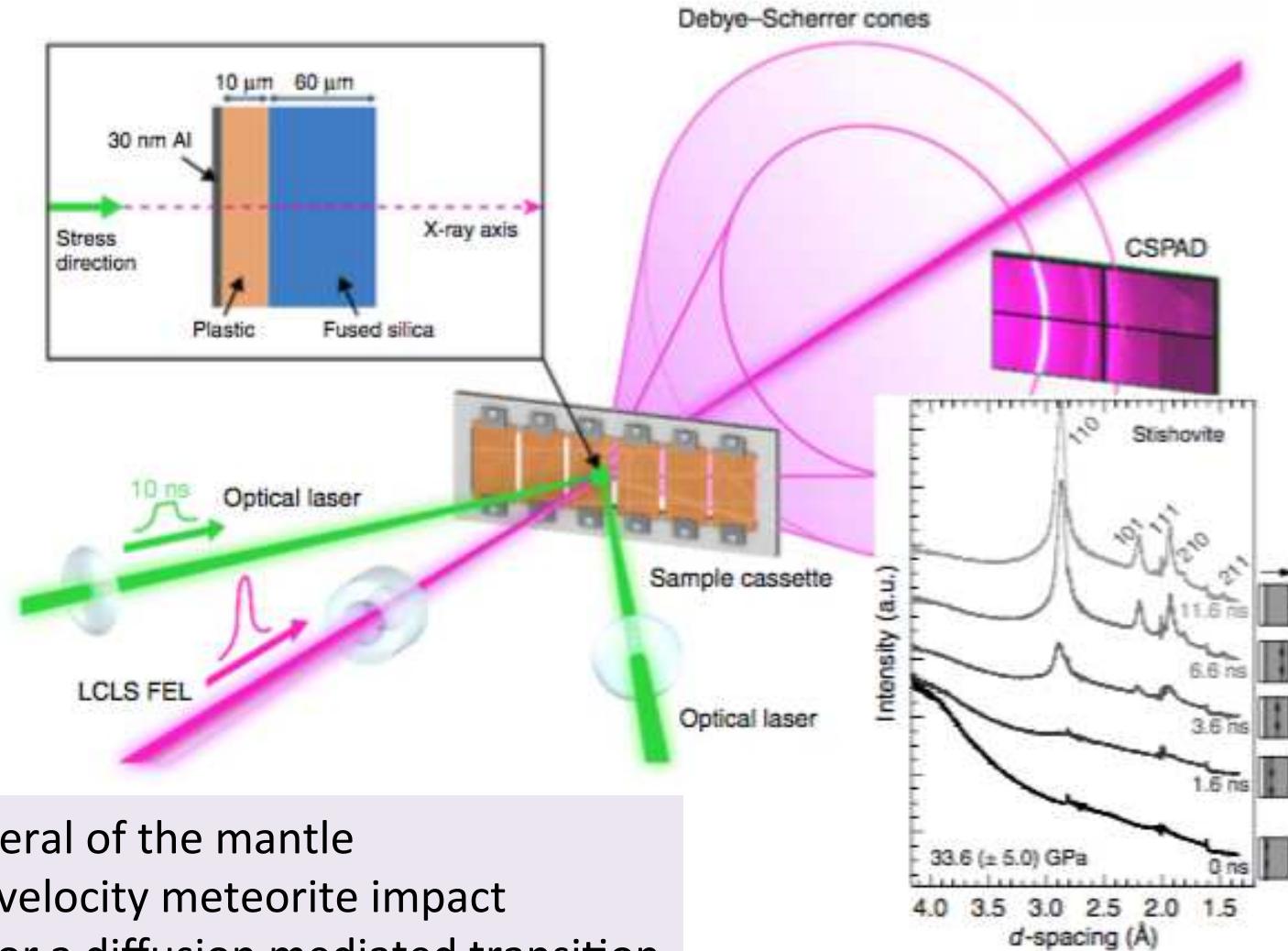
M. Harmand et al. PRB 2015

Summary of the most recent melting experiments



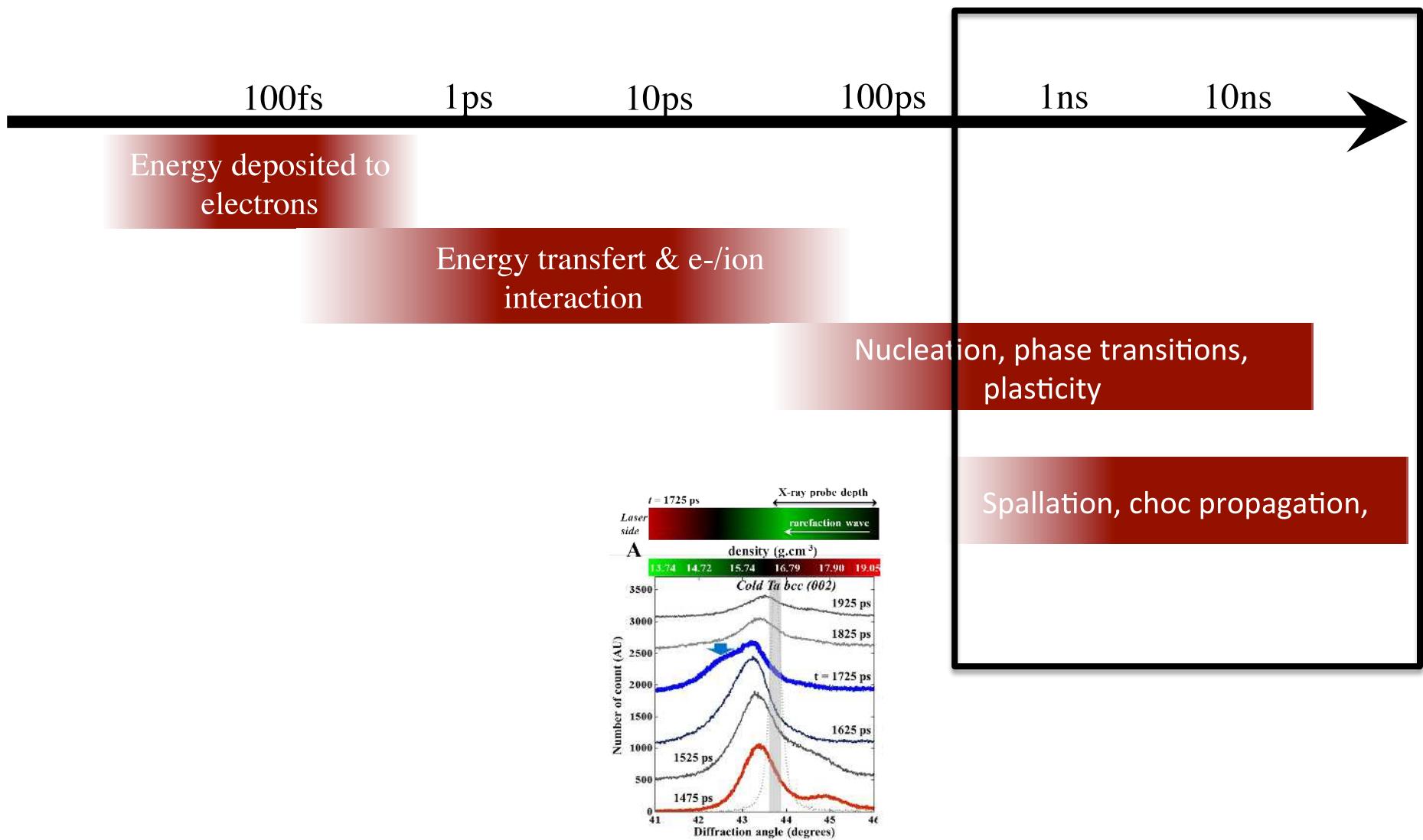
Are we probing in the release?
When do we start to see the melting?

Production of nm-size stichovite under shock as example of an ultrafast reconstructive PT

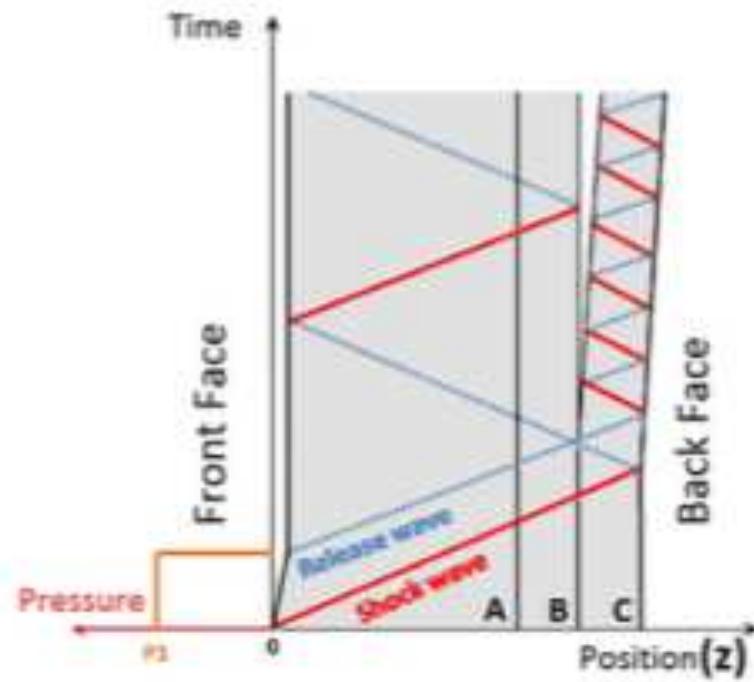
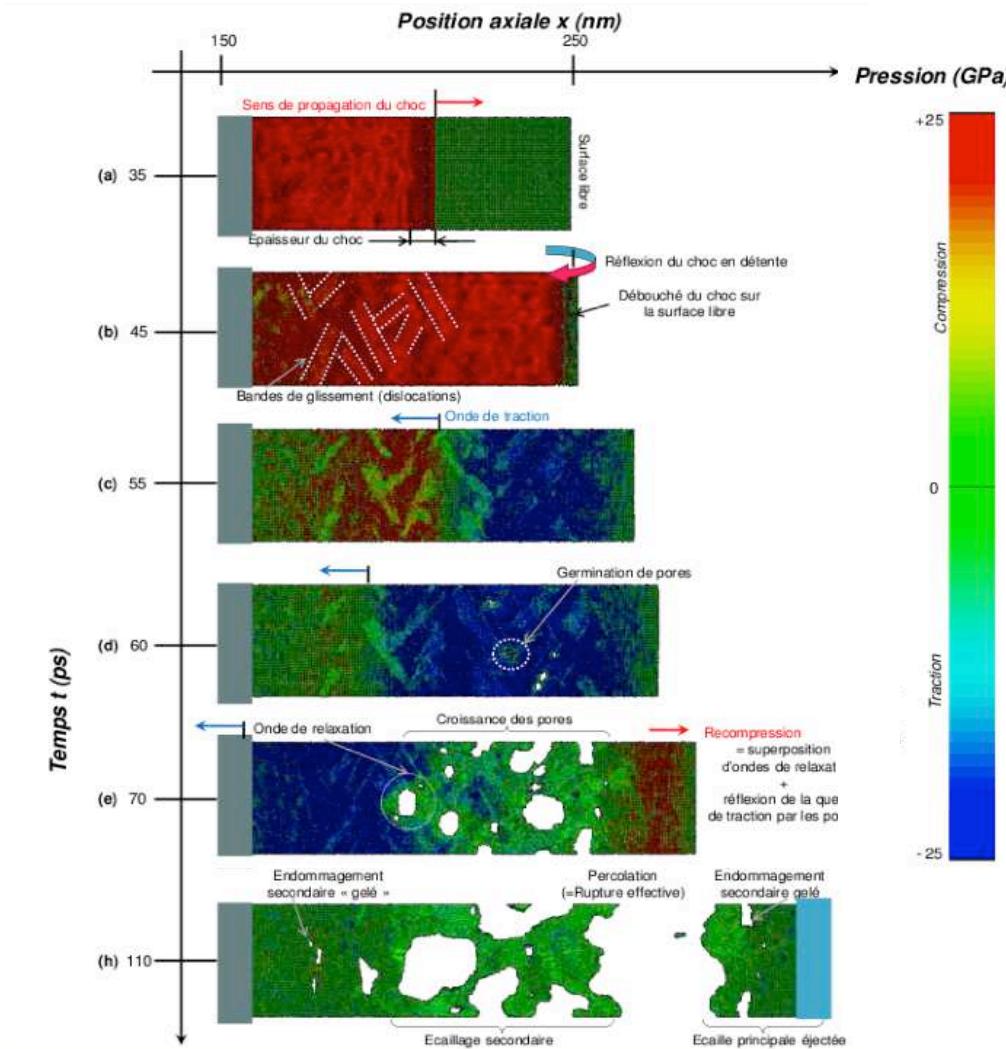


One of the major mineral of the mantle
Only found on hyper-velocity meteorite impact
→ Very fast process for a diffusion mediated transition
→ Was expected to require an amorphous PT

Gleason et al. *Nature Com.* 2015

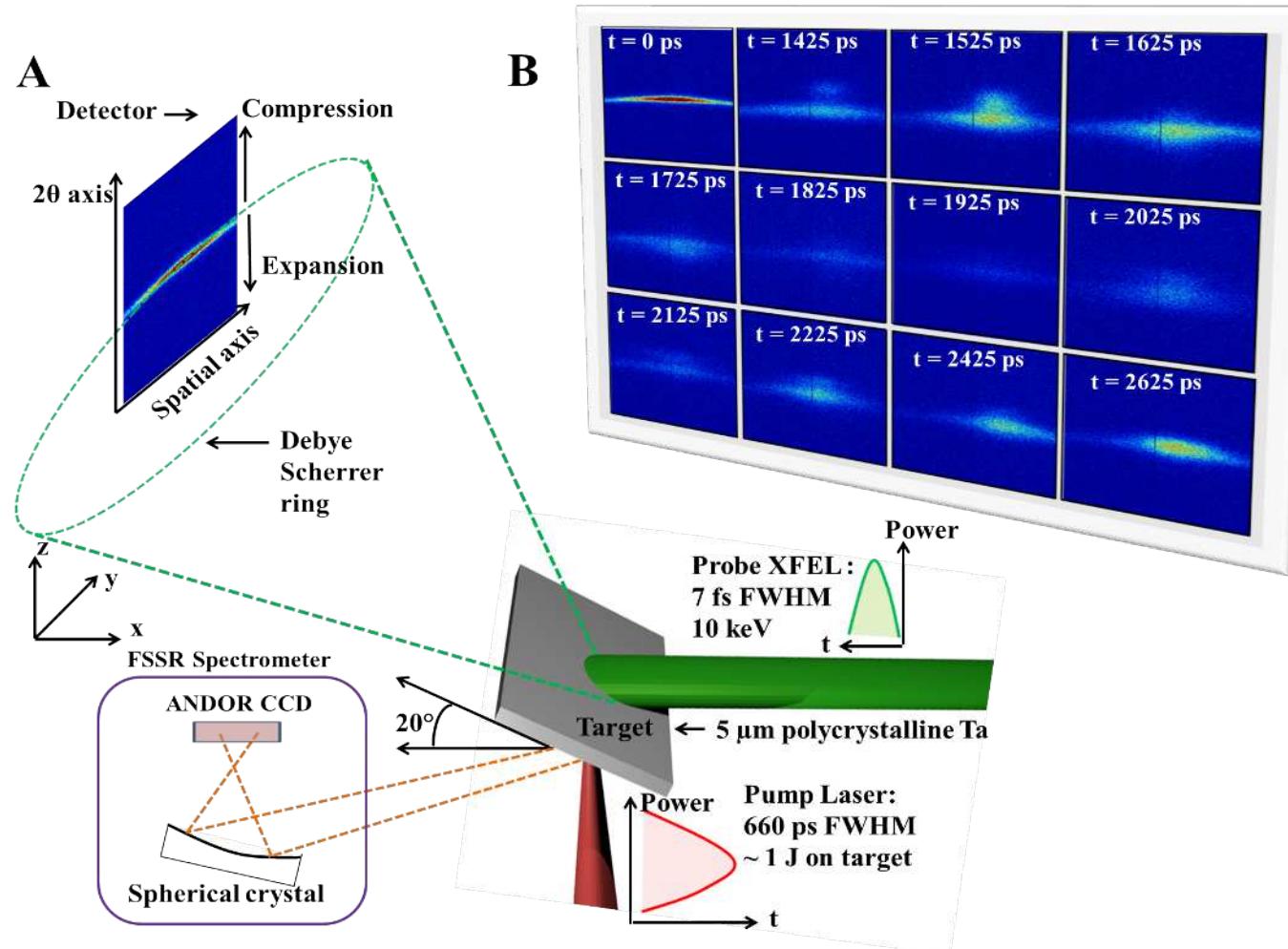


Spallation process under shock

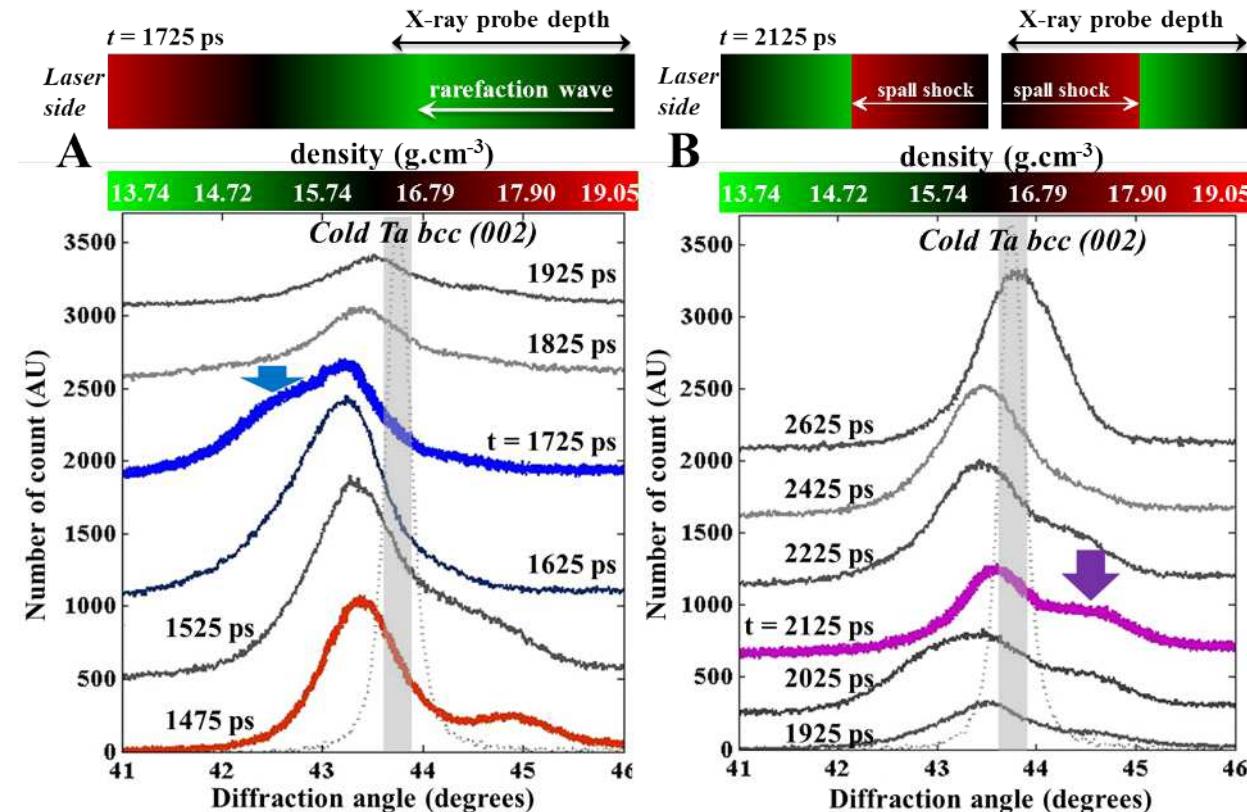


Successive compressive and tensile loading at interfaces

Spallation process under shock



Spallation process under shock



In situ tracking of the compression and release waves
Good agreement with MD simulations

Conclusion and Perspectives

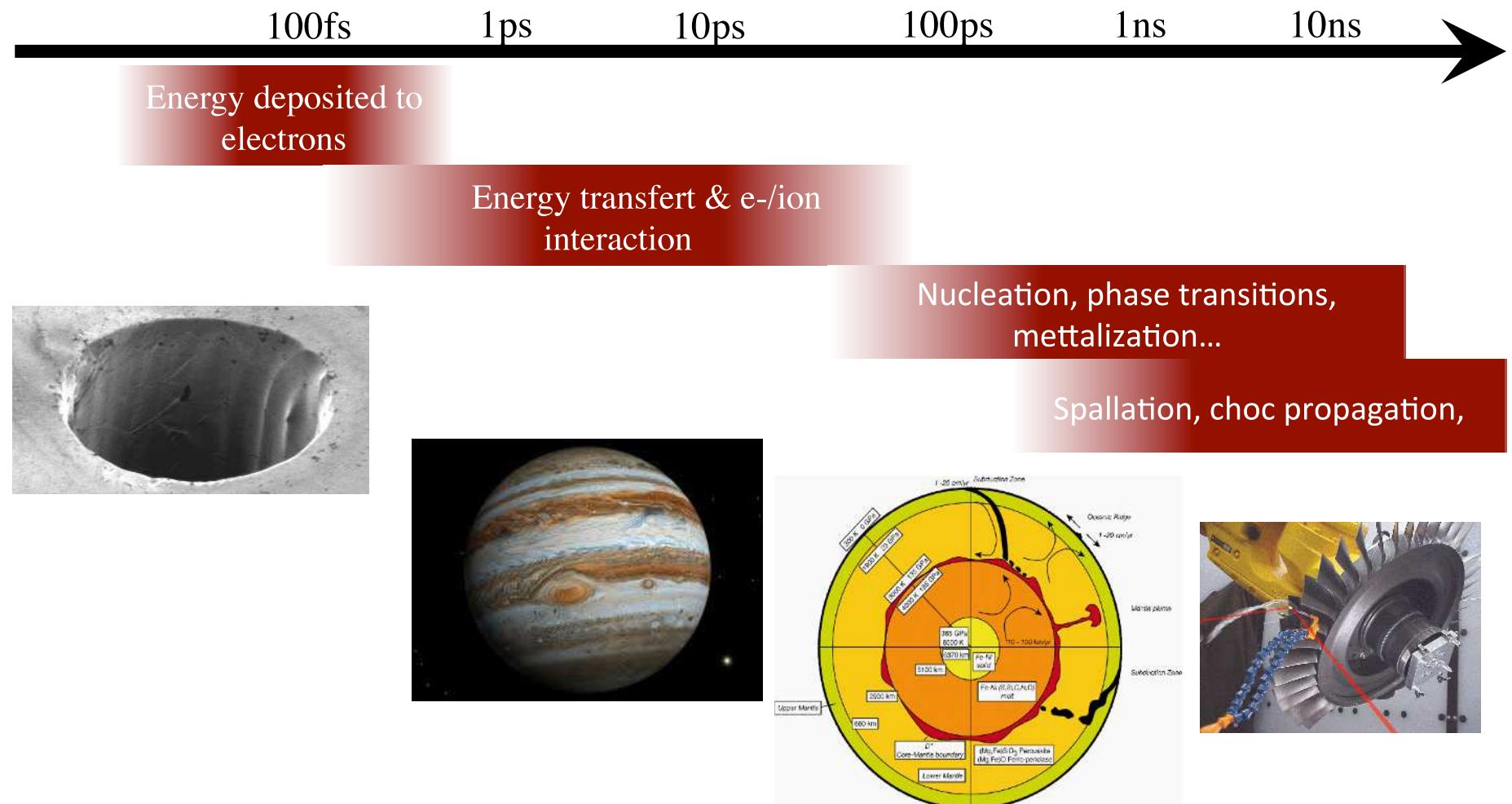
FEL are offering unique new diagnostics and approaches to understand WDM

- Access to structural diagnostics, in the bulk of the material
- Access to a full understanding of the dynamic processes from fs to ns

The applications for WDM studies in geoscience, laser processes, fundamental understanding still need developments

- Close collaboration with theory groups for modeling
- Instrumental developments for multiple diagnostics and for high precision measurements
- Effort towards the application has to be pursued

Conclusion and Perspectives



Collaboration

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