# Characterization of matter in the Megabar regime using laser shock compression: applications to planetology

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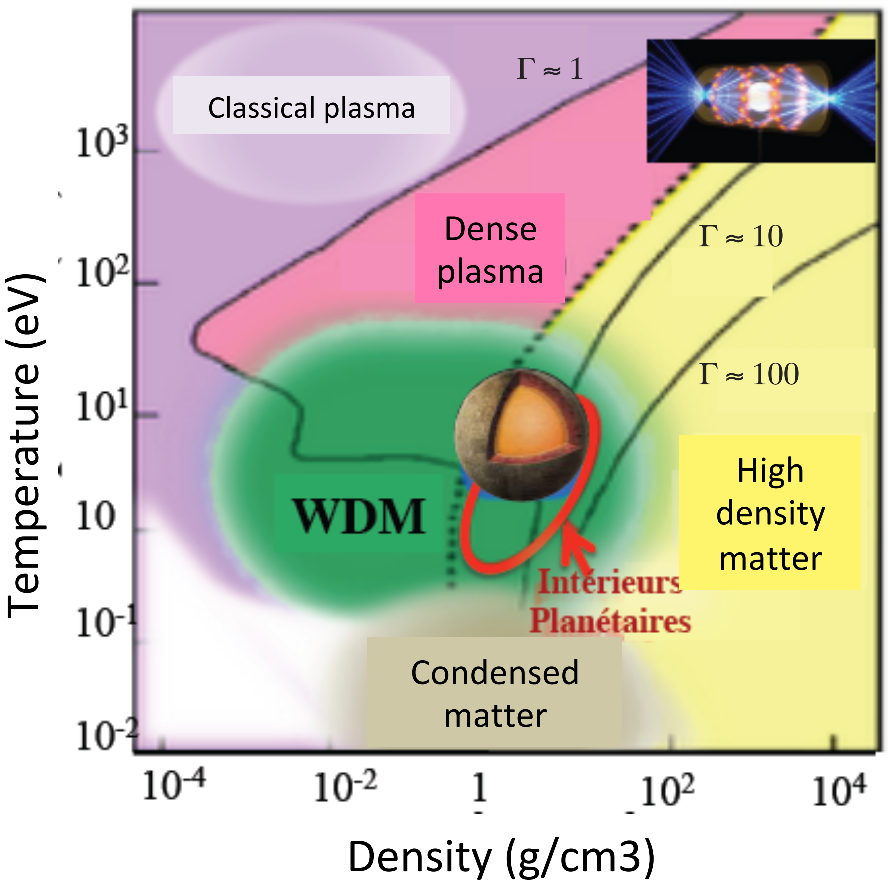
# OUTLINE

- ✓ Introduction: The Warm Dense Matter
  - Why its characterization is important ?
  - What is planetology in laboratory? How to do it?
  - What is a laser shock?
  - How to measure the equation of state?
- ✓ Recent developments: some examples on silicates
- ✓ Conclusions

# The Warm Dense Matter

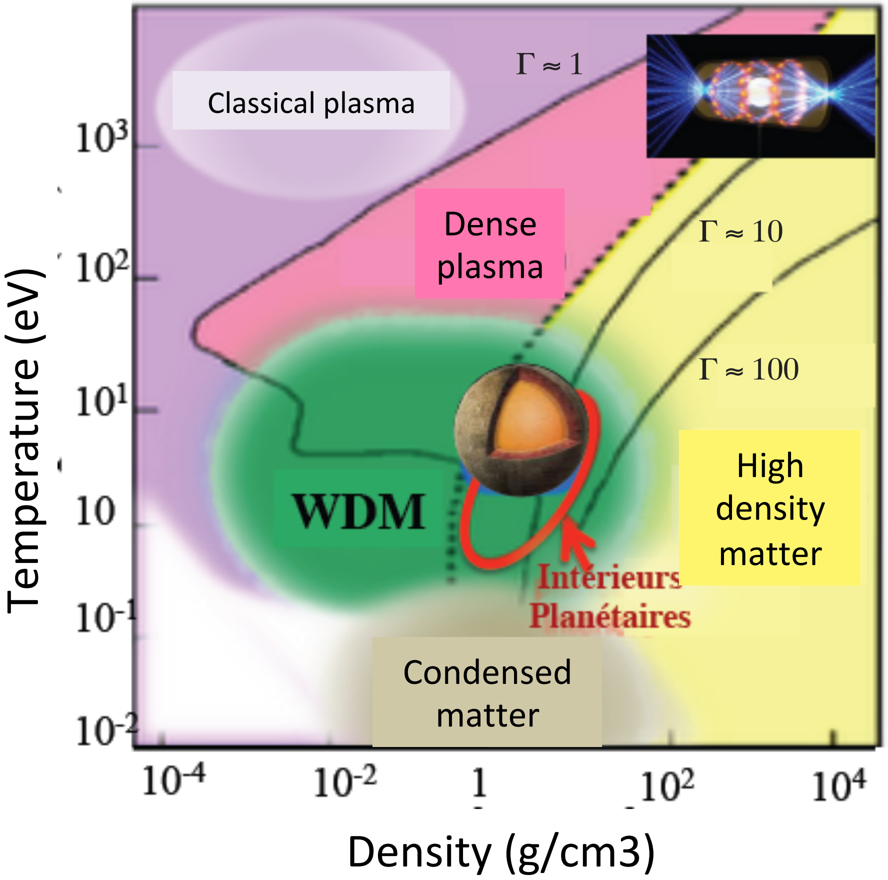


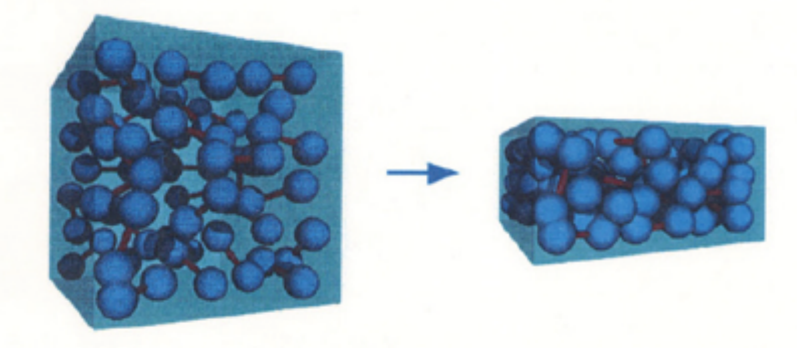
$0.\,1\ \rho_{solide} < \rho < 100\ \rho_{solide}$

$0.\,1\ eV < T < 100\ eV$

P > Mbar

# The Warm Dense Matter





**WDM is the state at the intersection between plasma physics and condensed matter physics.**

**WDM is correlated and degenerate**

Correlated $\Gamma = E_c / E_{th} > 1$

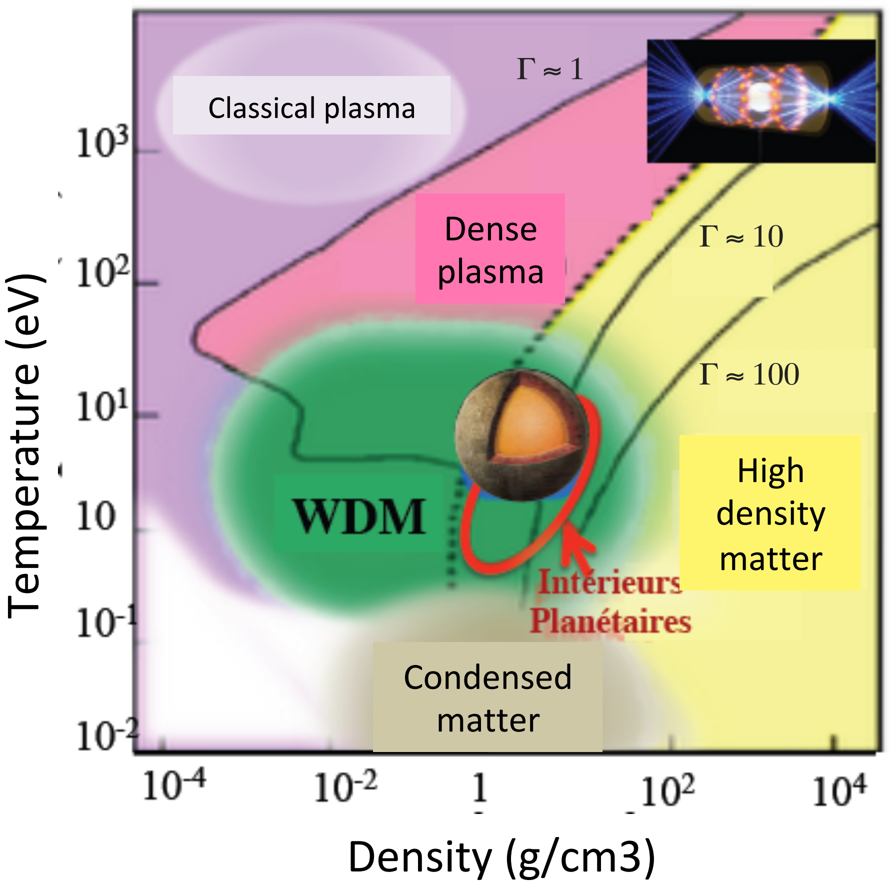
*Coulomb energy* $E_c$

*Thermal energy* $E_{th}$

Degenerate $T_F > T$

Difficult to simulate theoretically -> Quantum Molecular Dynamics calculations -> need of experimental data to validate them

# The Warm Dense Matter

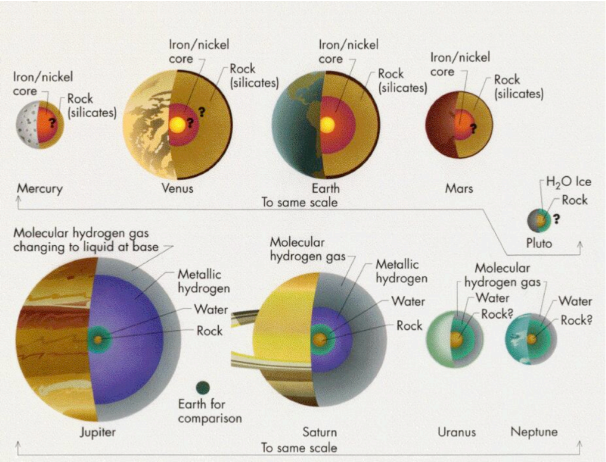


Conditions found in inertial confinement fusion path compression and inside planets

In the last tens years -> new topic "planetology in laboratory"

# An important motivation to study WDM : planetary science

- ✧ Study of Formation and evolution of planets
- ✧ Study of planet interiors

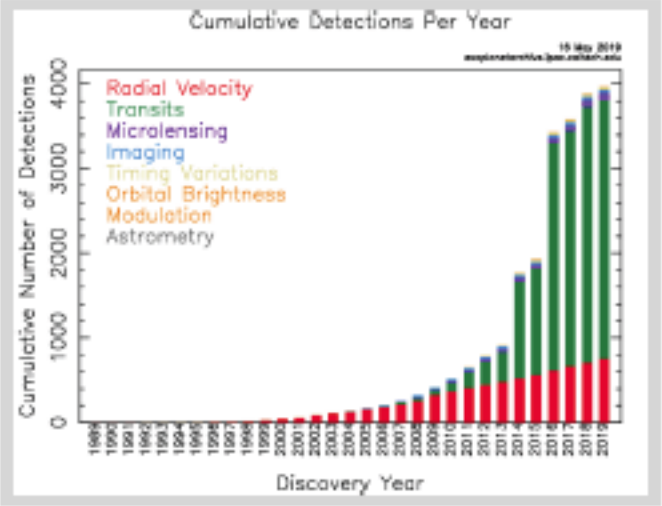


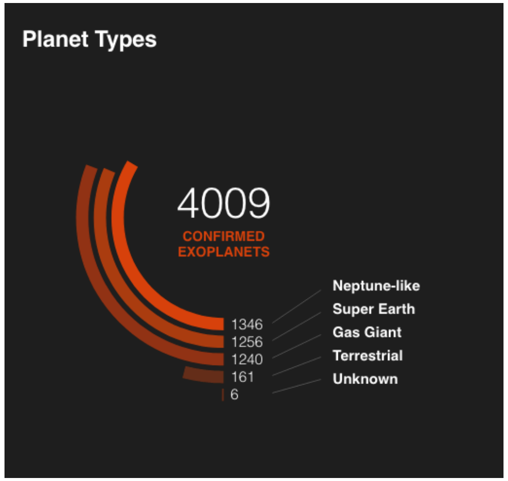
FAST GROWING SCIENCE DUE TO EXOPLANETS DISCOVERY

From 1988 to today

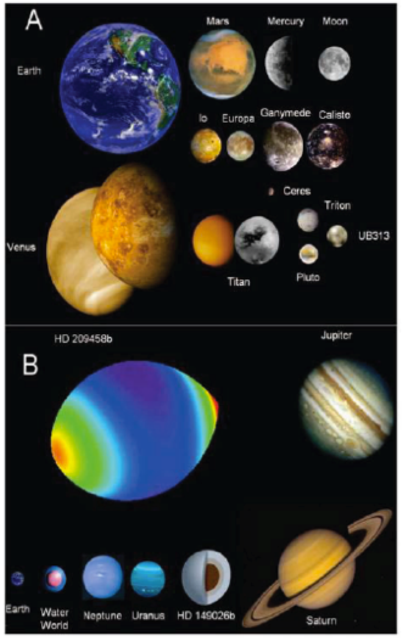
↓

4009 planets discovered





# Key questions about planets inside and outside the Solar System



- What is the nature of the iron core at the center of Earth and other terrestrial planets? -> Dynamo effect
- What is the interior structure of Jupiter and the other giant planets? Why is Saturn luminosity not compatible with its age?

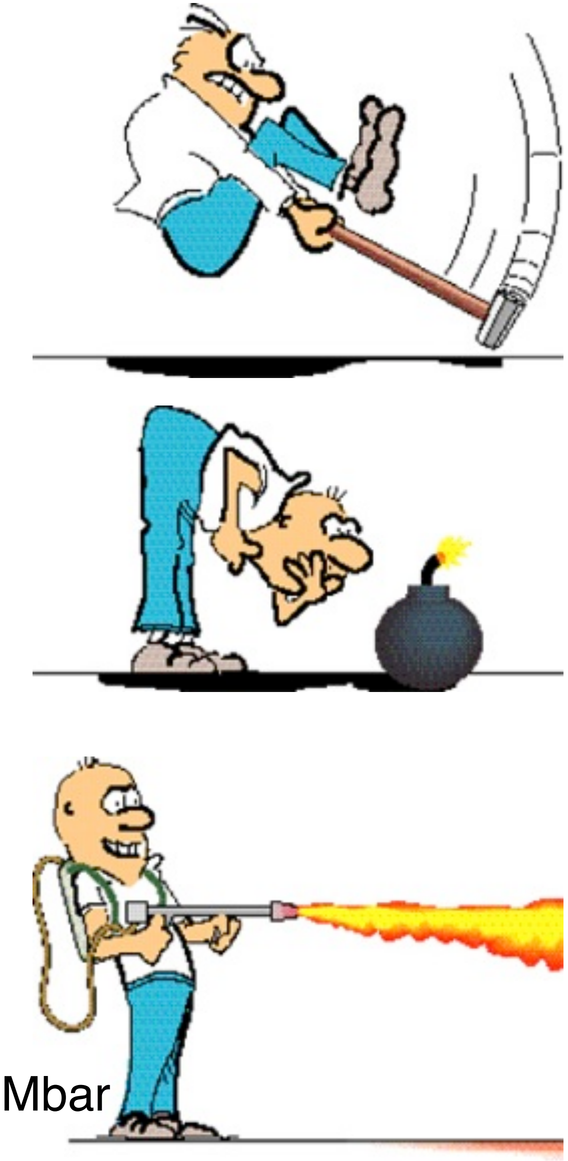
The internal structure is inferred indireclty from their global properties: M mass, the radius and gravitationnal moments (few observables)

For exoplanets -> only mass and radius

To progress in the planetology:

We have to know the equation of state, the conductivity, structural properties etc…of the matter existing in the planet interiors.

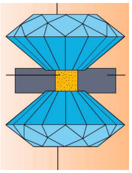
# How create this planetary matter in a laboratory?



**Static way**

**Diamond cell** → **Isothermal Compression**

P ≈ 0 - a few Mbar



**Dynamic way**

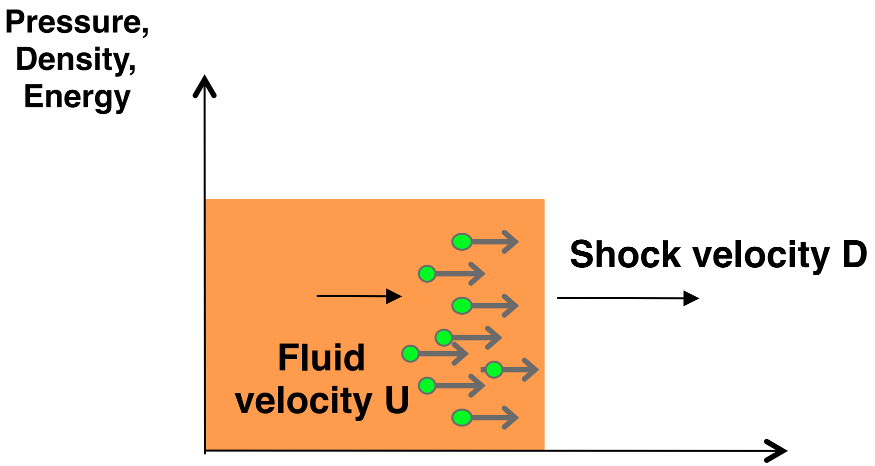
**Chemical explosions, gas guns, ions beams**

High power lasers

→ **Shock Compression**

P ≈ 0 – several tens of Mbar

# What is a shock ?

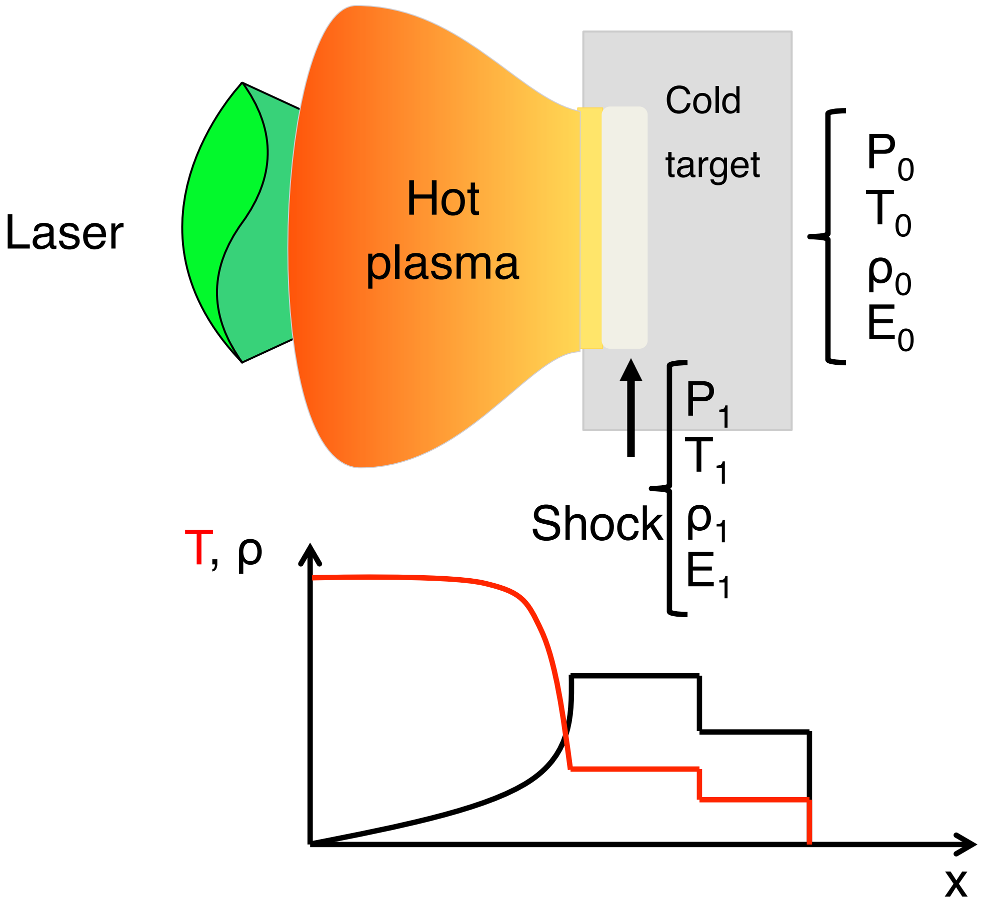


**Rankine Hugoniot relations**

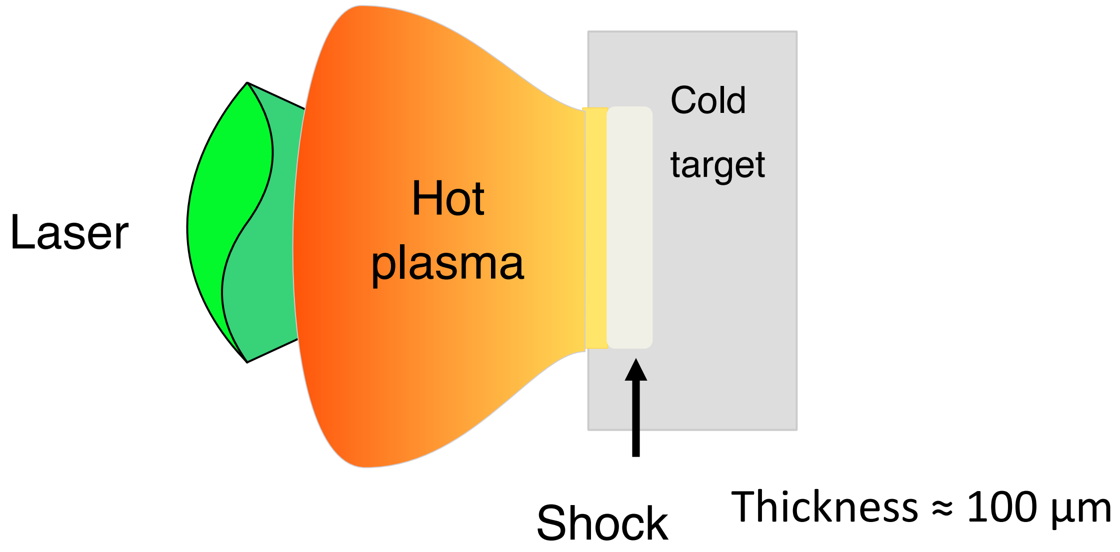
**Conservation of the**

- ⇒ **mass**
- ⇒ **momentum**
- ⇒ **energy**

# Laser driven shock



# Laser driven shock



**Ablation pressure**

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

**$I_L$ ($10^{14}$ W/cm$^2$)**
**λ (µm)**
**P (Mbar)**

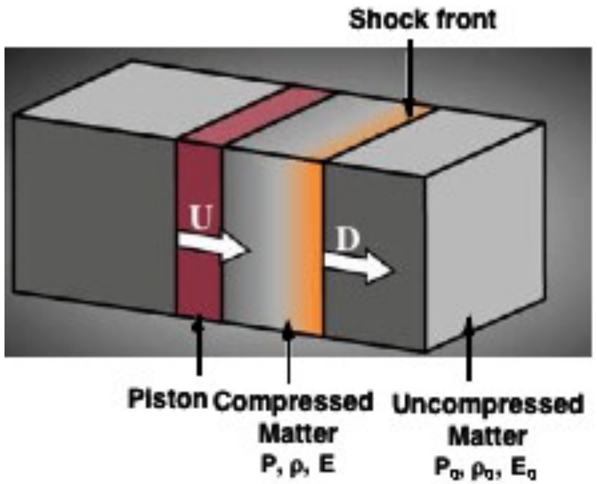
Typically E ≈ 100 -1000 J
Duration ≈ nanosecond
Focal spot ≈ 500-1000µm

Today ---> Pressures of several tens of Mbar

# EOS measurements using a shock

Rankine-Hugoniot

$$\begin{cases} \rho_0 D = \rho\,(D-U) & \text{mass} \\ \rho_0 D U = P - P_0 & \text{momentum} \\ \rho_0 D(E-E_0+U^2/2)=PU & \text{energy} \end{cases}$$



5 unknown parameters (D,U,P,E,$\rho$)

To have a point on f(P, E, $\rho$) = 0

✓2 parameters - *absolute measurement*
2 velocities D and U → time and distance

✓1 parameter - *relative measurement*
shock velocity D in 2 different materials (one used as reference)

# EOS measurements using a shock

Rankine-Hugoniot

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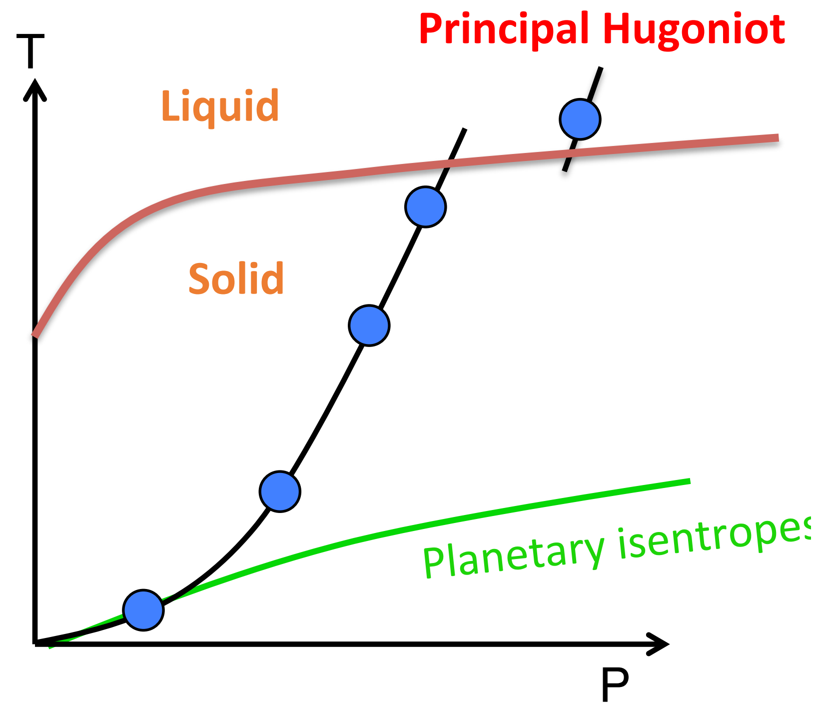


5 unknown parameters $(D,U,P,E,\rho)$

To have a point on $f(P, E, \rho) = 0$

✓2 parameters - *absolute measurement*

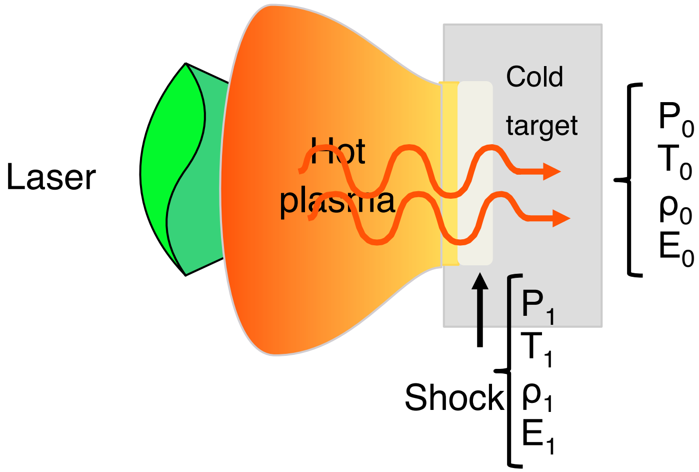
2 velocities D and U → time and distance

✓1 parameter - *relative measurement*

shock velocity D in 2 different materials (one used as reference)

# The requirements and challenges

The shock must be uniform, well controlled -> without preheating



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Complex multilayer targets

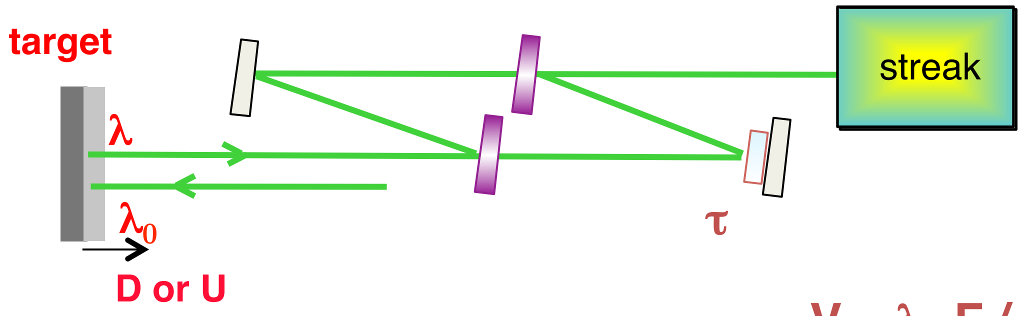
To measure parameters with required precision ( *≈ qq %* ) for a target < *1 mm* within ≈ *$10^{-9}$ s*

High resolution for the detectors
< *10 µm* et ≈ *$10^{-12}$ s*

# How do we measure shock parameters?

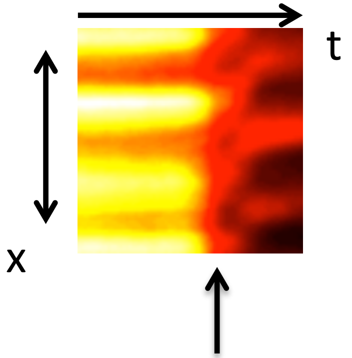
Usual diagnostics

**V**elocity **I**nterferometer **S**ystem for **A**ny **R**eflector (VISAR)



$V = \lambda_0 F / 2\tau$

Velocities (D or U) by
Doppler effect & reflectivity

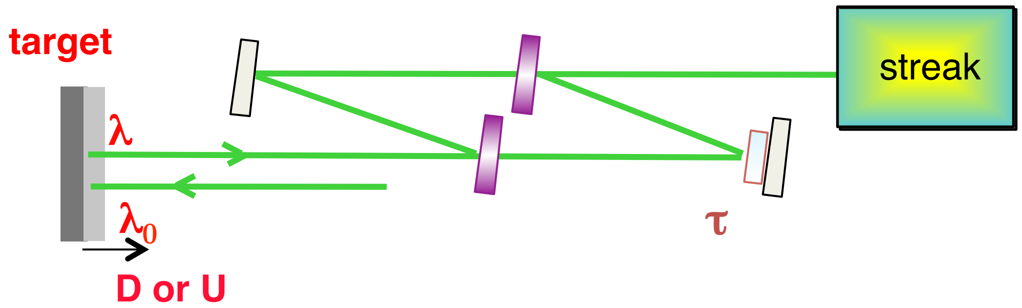


Shift of F fringes at t

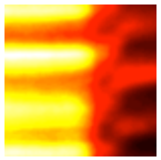
# How do we measure shock parameters?

## Usual diagnostics

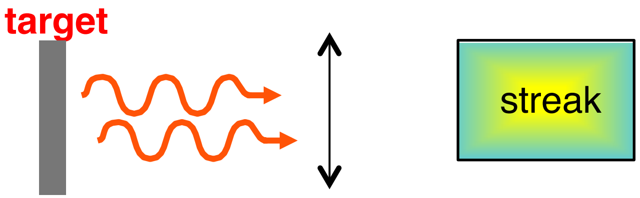
### Velocity Interferometer System for Any Reflector (VISAR)



Velocities (D or U) by Doppler effect & reflectivity

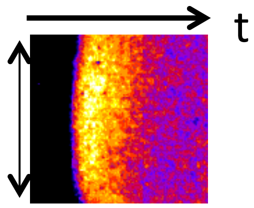


### Self-emission



Temperature T

**Kirchoff law: $I(T,\lambda) = \alpha(\lambda)\, I_P(T, \lambda)$**



**Planckian radiation**

**$\alpha(\lambda) = 1 - R(\lambda)$**

⇨ **T**

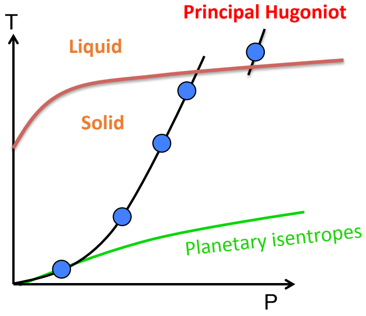
**We obtained reliable EOS data (method tested on standard materials)** Koenig et al. PRL 94

# Laser compression experiments for planetology at LULI

- Three main challenges

To perform microscopic study to progress in the knowledge of phase diagrams of the main planetary components

✓ Coupling X-ray diagnostics (X-ray diffraction, X-ray absorption near edge spectroscopy ...) with a laser shock



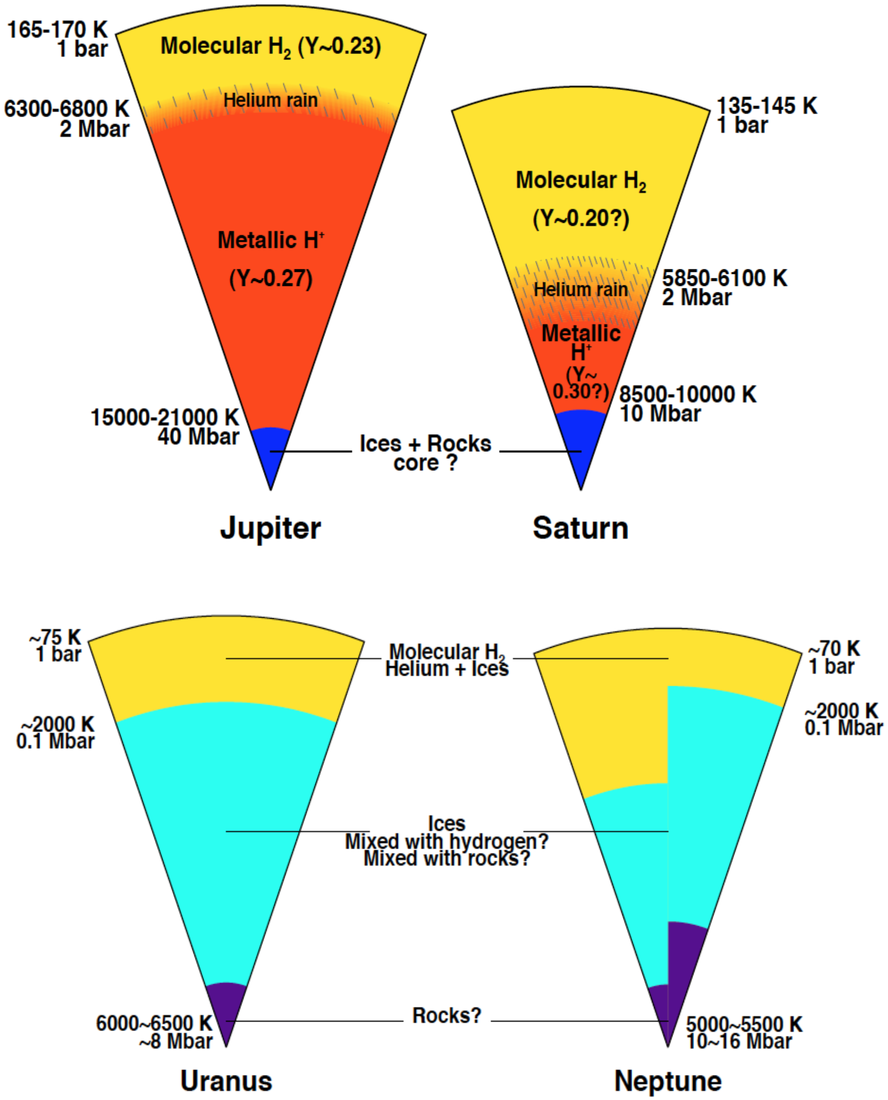
To achieve planetary conditions (dense and cold < 1eV)

Shock → Too hot

✓ Alternative compression techniques (quasi-isentropic compression, double choc, DAC+ shock)

To investigate complex compounds typical of planetary interiors

# Main planetary compounds



T. Guillot 2014

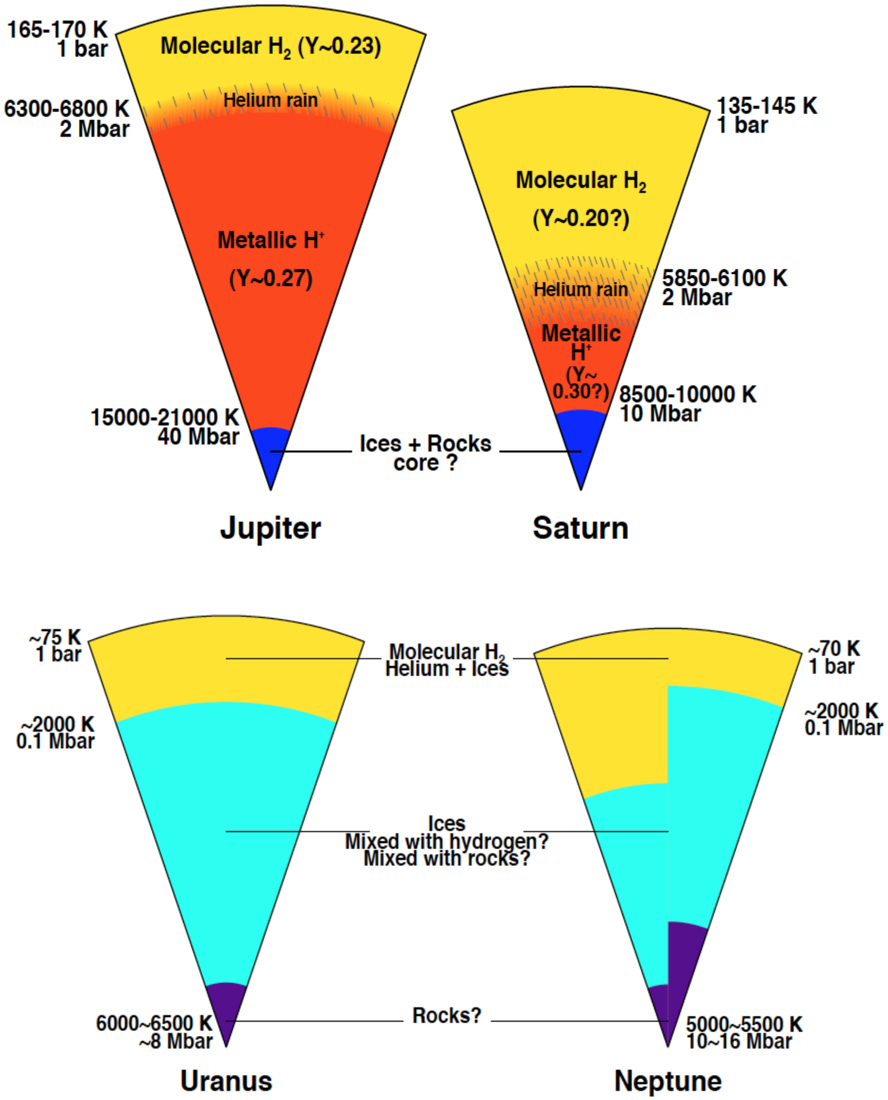
-hydrogen and helium

-mixtures of $H_2O$, $NH_3$, $CH_4$

-Silicates and oxides:
$(Mg,Fe)2SiO_4$, $(Mg,Fe)SiO_3$, $(Mg,Fe)O$

-Iron and iron alloys

# Main planetary compounds



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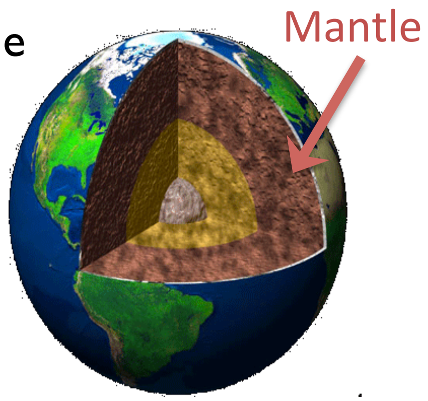
# The study of silicates : the context

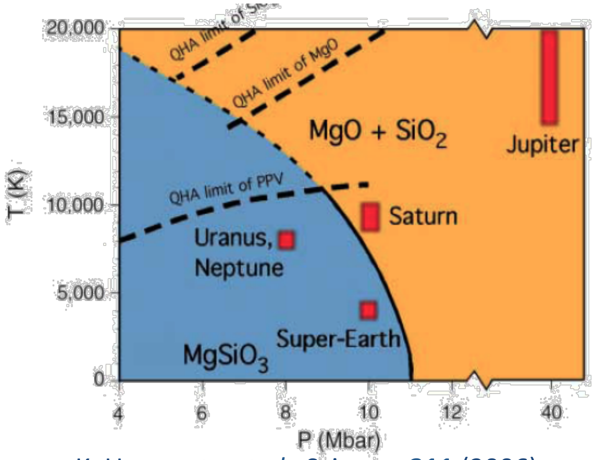


Silicates ($MgSiO_3$, $Mg_2SiO_4$, …) are expected to be inside

- cores of giant planets
- deep mantles of terrestrial ones

How do these compounds melt, dissociate or turn metallic?

→ Evolution and characteristics of these planets



K. Umemoto *et al.*, Science, **311** (2006)

Ab initio calculations predict the dissociation
of $MgSiO_3$ → ($SiO_2$ + MgO)

We need experimental data in high pressure regime to validate theory

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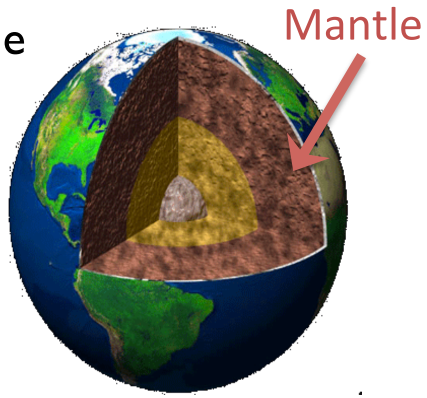

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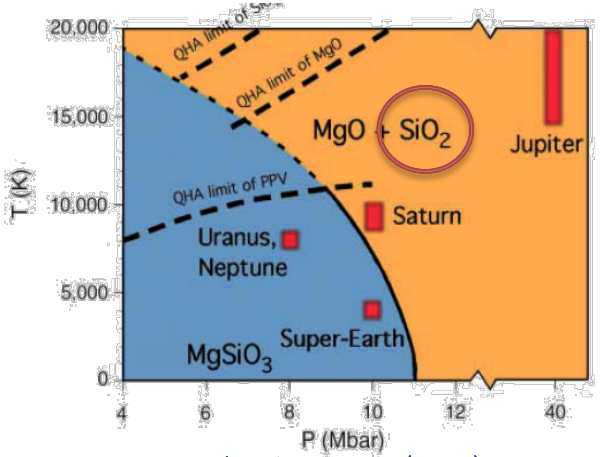

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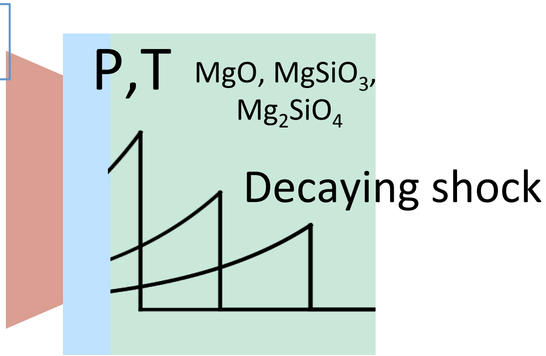
Different experimental campaigns:

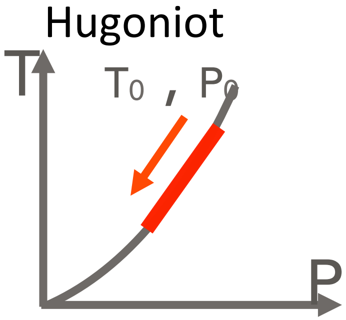
- Detection of phase transitions using decaying shock (LULI and GEKKO)
- Electronic and ionic structural changes using XANES (LULI)
- Phases transitions using X-ray diffraction (recent experiment at LCLS)

# Detection of phase transitions in MgO-$SiO_2$ systems using decaying shock

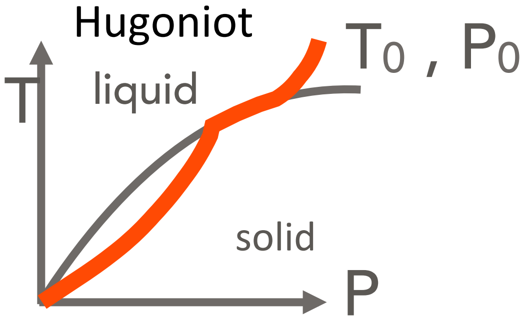
The principle

E ≈ 800 J
t = 1.5-2.5 ns
Φ ≈ 600 µm





No transition



Phase transition

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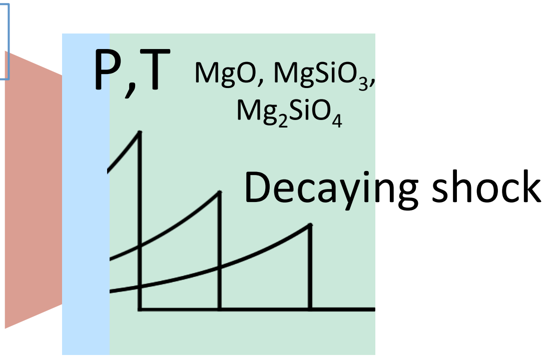

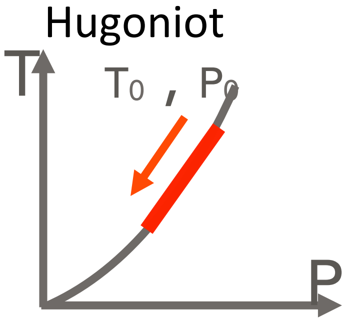

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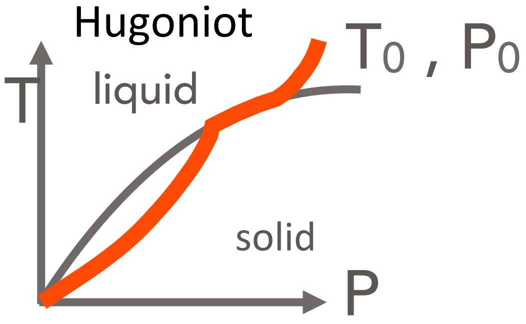
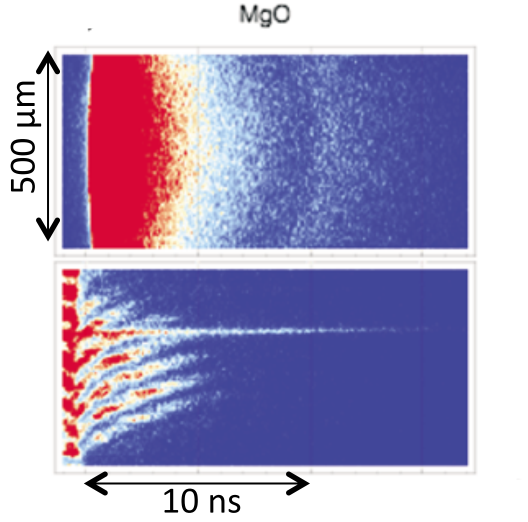


Phase transition



# Detection of phase transitions in MgO-$SiO_2$ systems

(LULI and GEKKO experiments)

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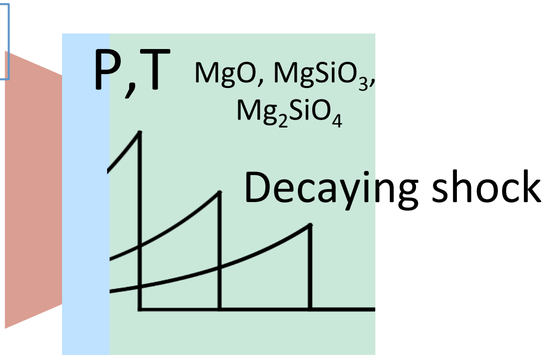
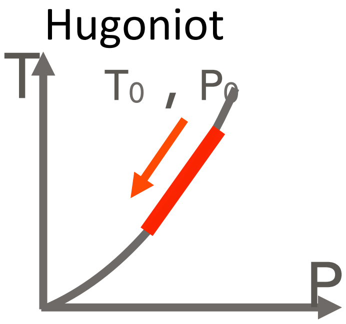


P,T

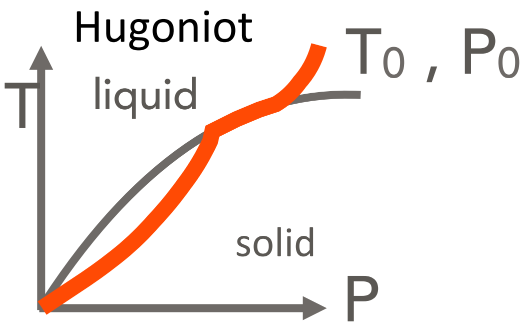
MgO, $MgSiO_3$, $Mg_2SiO_4$

Decaying shock



Hugoniot

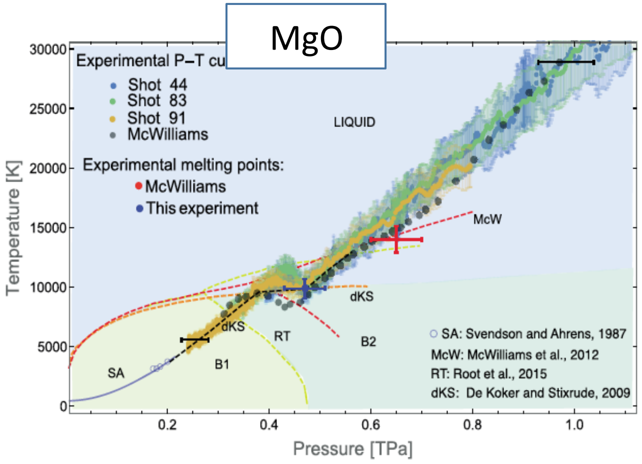
No transitions



Hugoniot

Phase transition

MgO



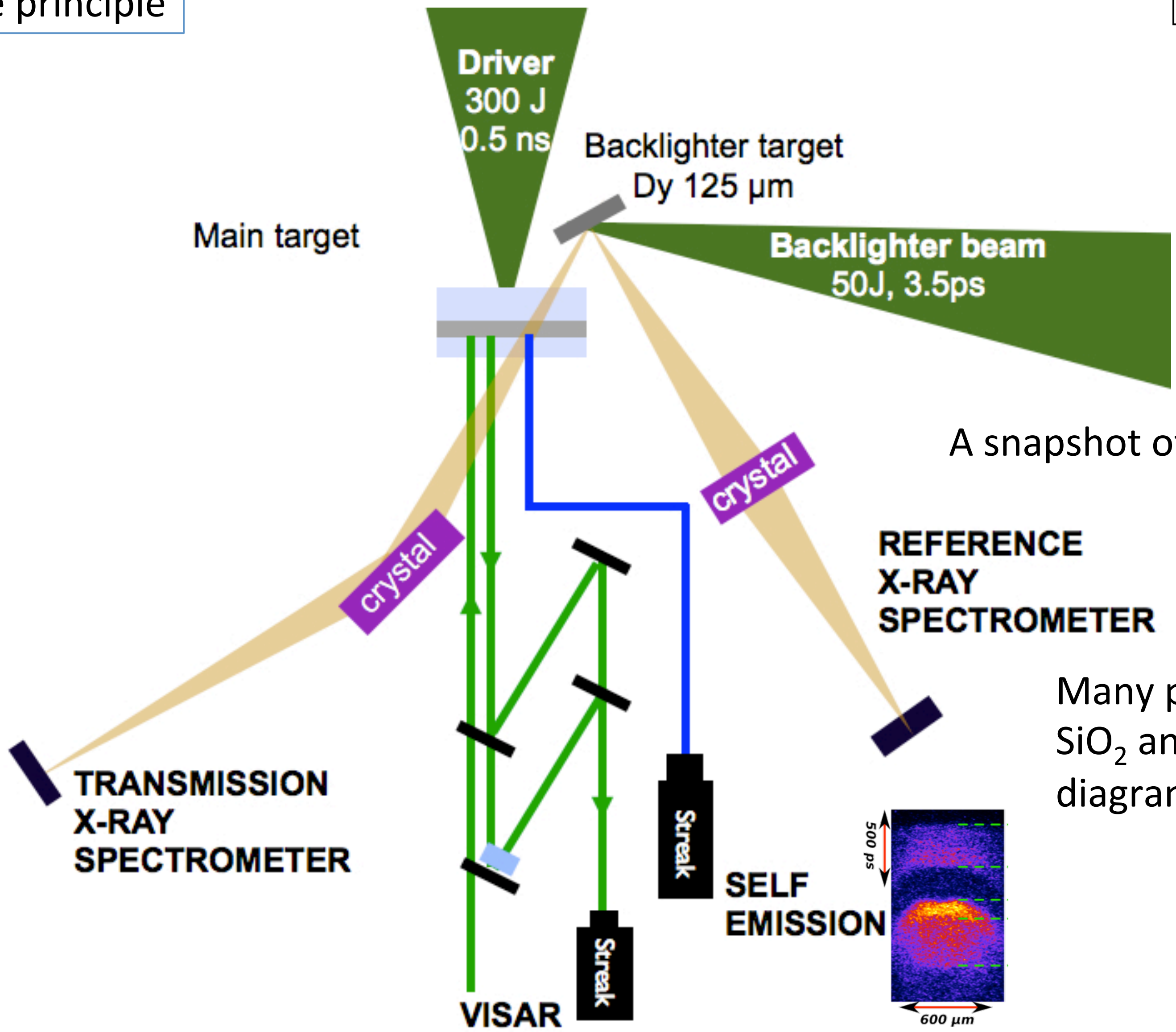
- Melting feature at 4.7 Mbar
- No other phase transitions
- Existence of a low conducting liquid layer at the mantle

Repercussions for super earth B field

R. Bolis et al. GRL 2016

# $SiO_2$ and MgO microscopic study using X-rays Absorption Near Edge Spectroscopy (experiments at LULI)

The principle

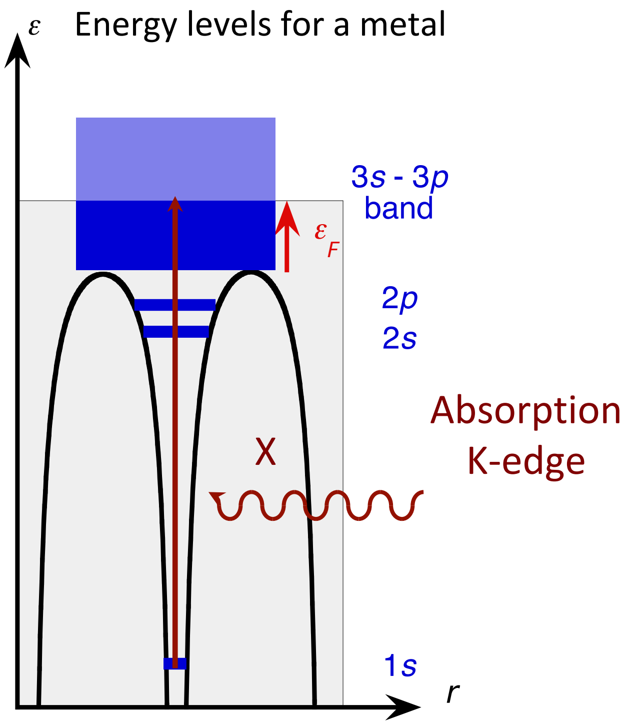


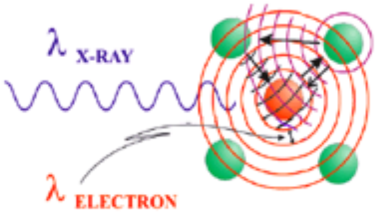
A snapshot of the plasma

Many probed conditions of $SiO_2$ and MgO phase diagram

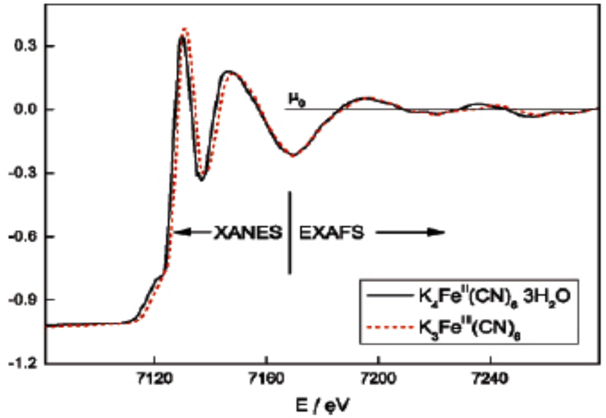
# $SiO_2$ and MgO microscopic study using X-rays Absorption Near Edge Spectroscopy (experiments at LULI)

## The principle





Multiple Scattering

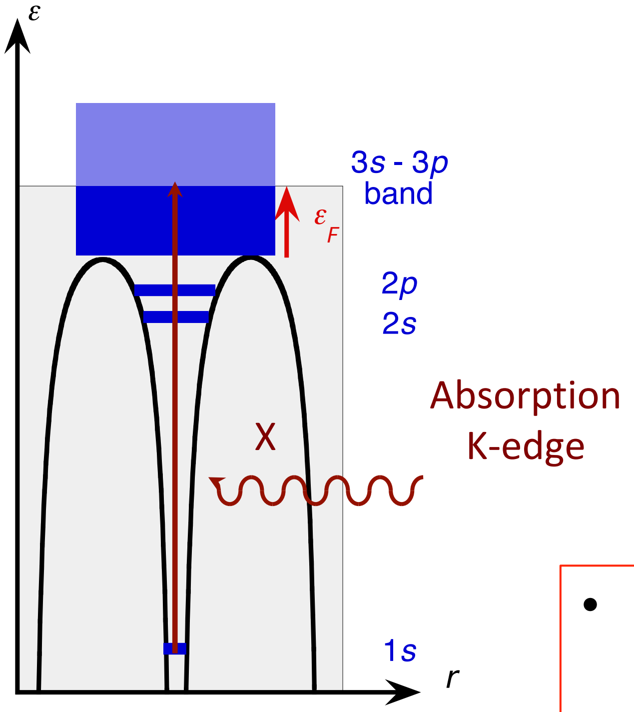


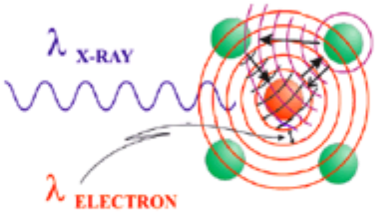
TYPICAL SPECTRUM CARACTERIZED by:

- Absorption edge corresponds to the transition of the photoelectron from the 1s level to the first unoccupied level above the Fermi energy
- Oscillations due to the scattering of the phtoelectron on the neighbouring atoms

# $SiO_2$ and MgO microscopic study using X-rays Absorption Near Edge Spectroscopy (experiments at LULI)

## The principle





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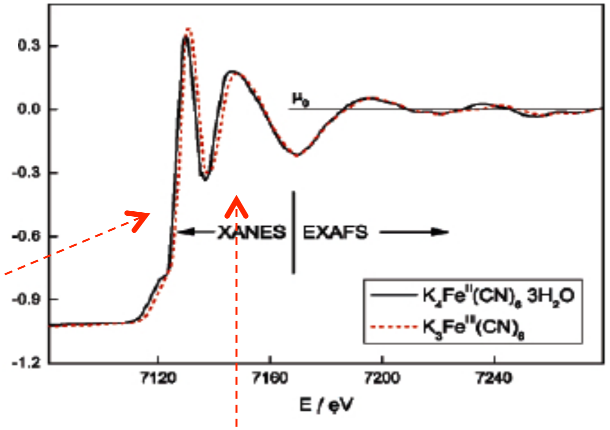


- Probe the valence electrons and the 1s level

- Probe the local atomic order

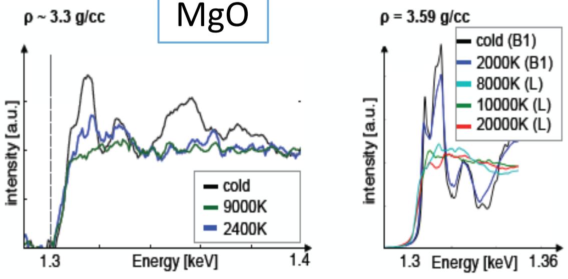
Powerful diagnostic to study both electronic and ionic structure changes, phase transitions and to test directly approximations used in theories

# SiO$_2$ and MgO microscopic study using X-rays Absorption Near Edge Spectroscopy (experiments at LULI)

Some data

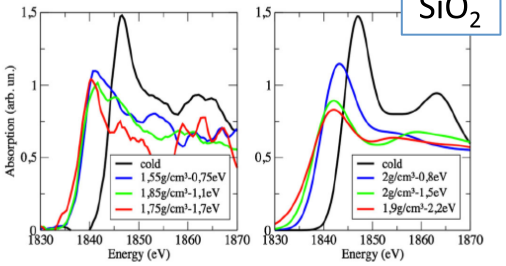
Experimental and ab initio spectra along isochores at $\rho \approx \rho_0$

MgO



Calculations are in agreement with experiment

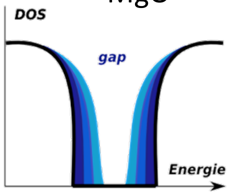
SiO$_2$

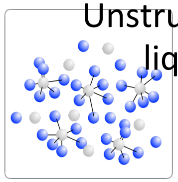


A. Denoeud et al PRL 2014, A. Denoeud et al. PRE 2016, Bolis et al submitted

✓ We were able to get information on the gap closure mechanism and on liquid structures

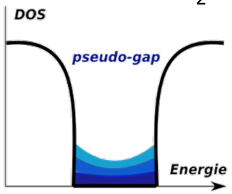
MgO

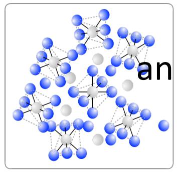




Unstructured liquid

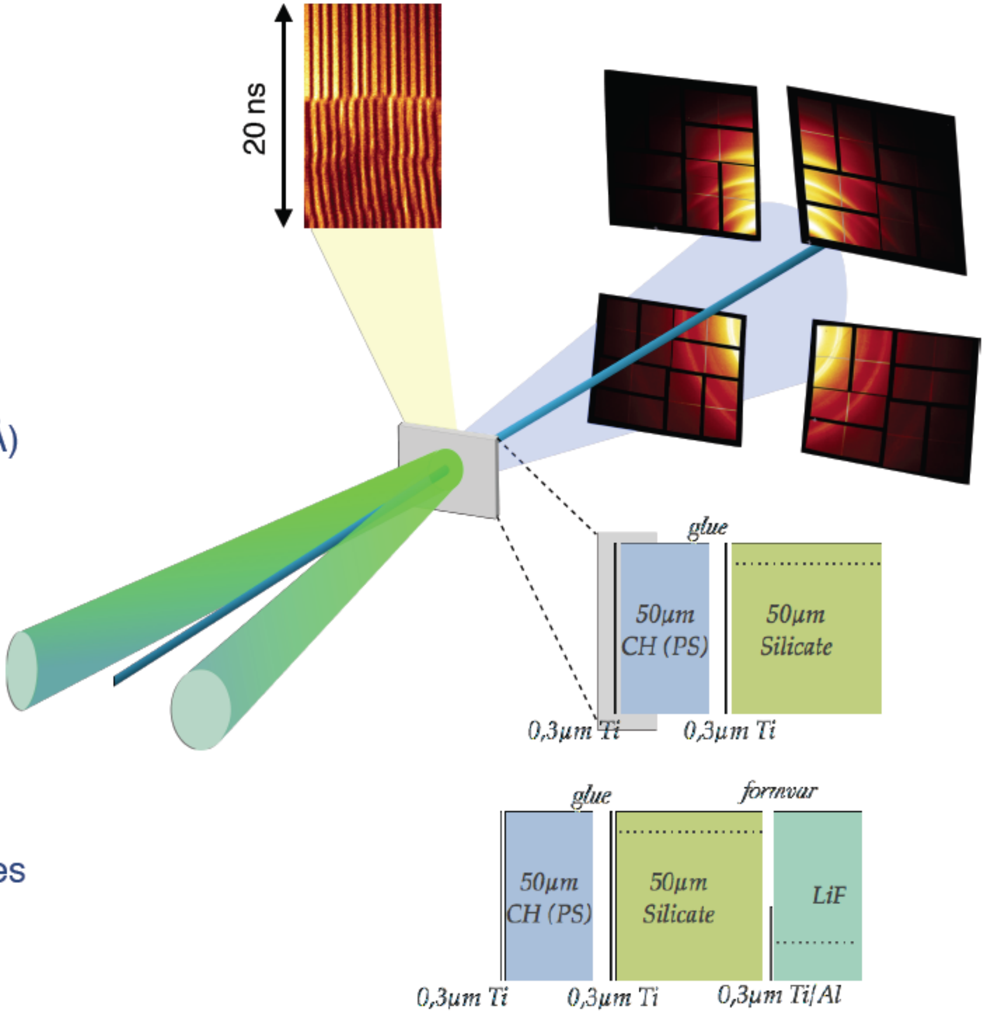
SiO$_2$





Liquid with an angular order

# X-ray diffraction at Linac Coherent Light Source



## LCLS free-electron laser

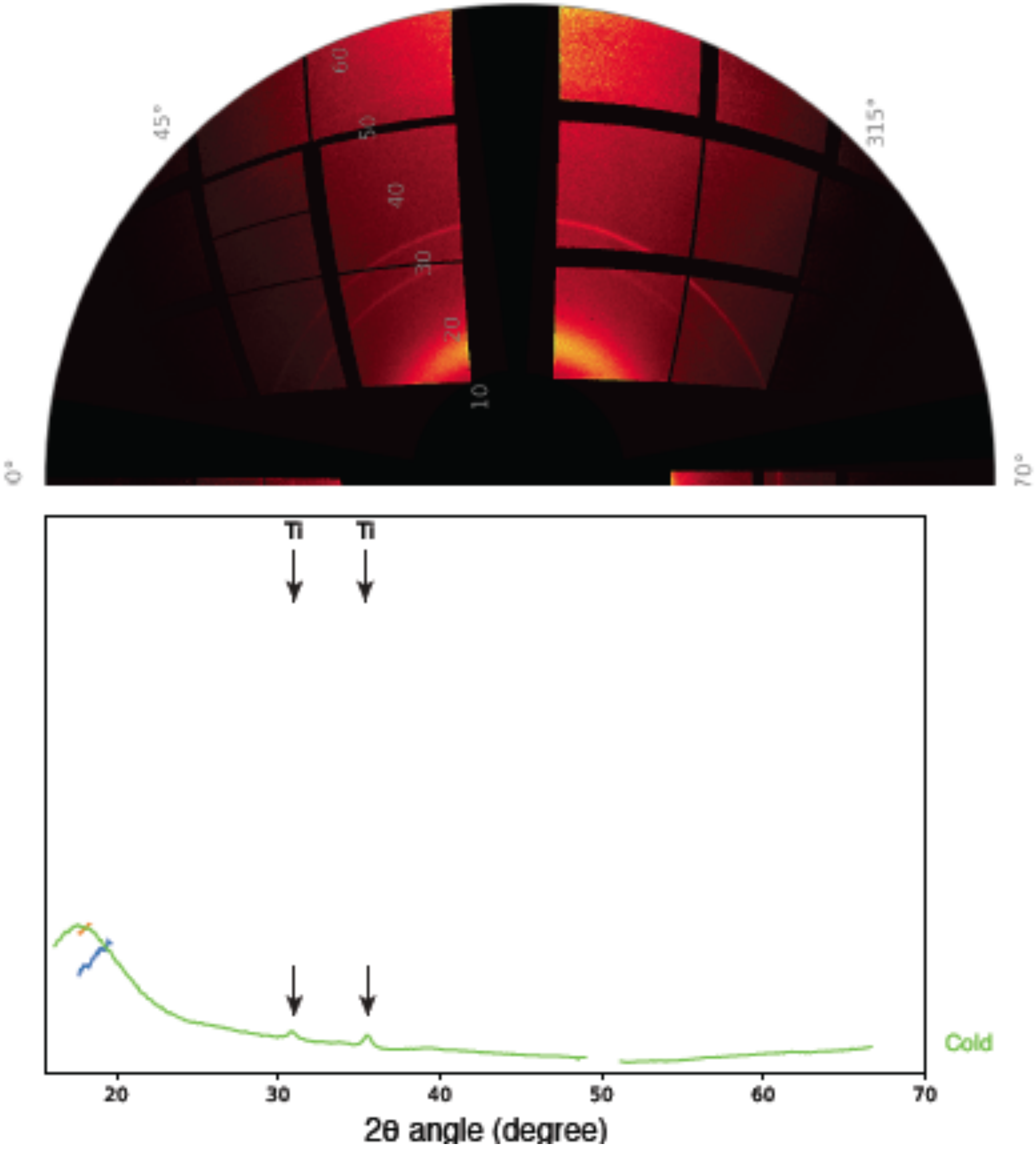
- hard X-rays at 9 keV (λ = 1.3753 Å)
- 50 µm focal spot
- 30 fs pulse

## Drive lasers

- 4 drive lasers combined in 2 arms
- Up to 70 J on target (λ = 532 nm)
- 350 µm focal spot with phase plates
- 10 ns pulses, temporal shaping
- 1 shot / 7 min / arm

# Results on fused silica $SiO_2$

## Fused-$SiO_2$ initially in 4-fold Si-O coordination



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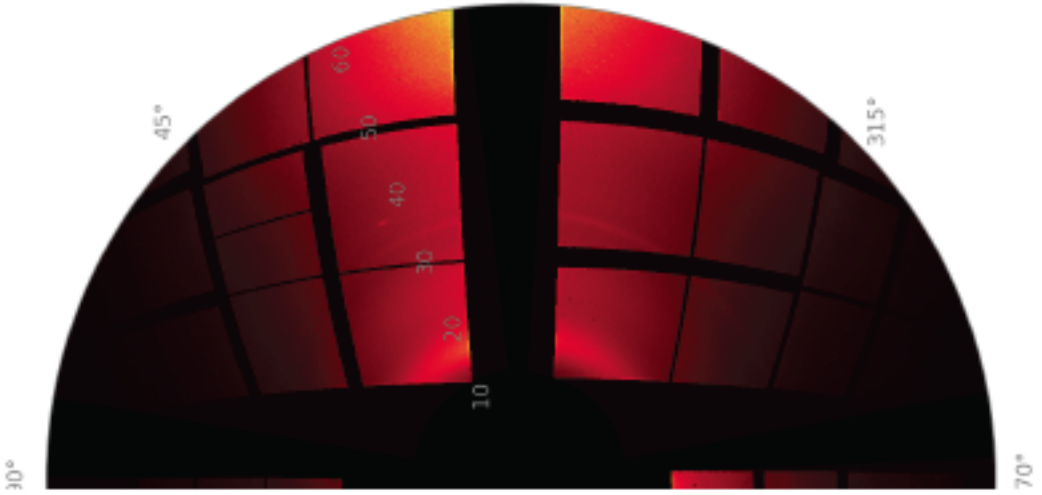
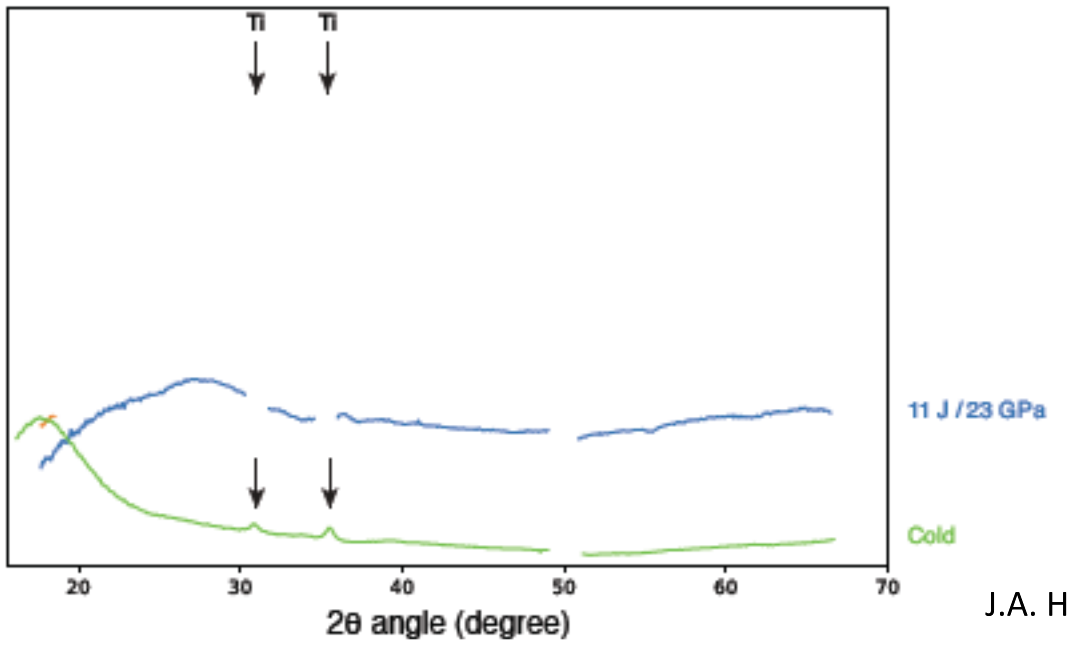
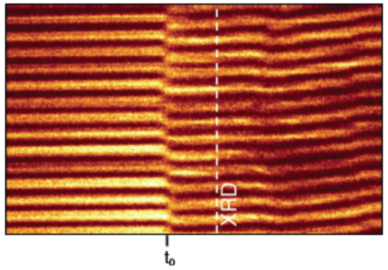
# Results on fused $SiO_2$

## Formation of amorphous phase with 6-fold coordination – 23 GPa – 3.3 ns

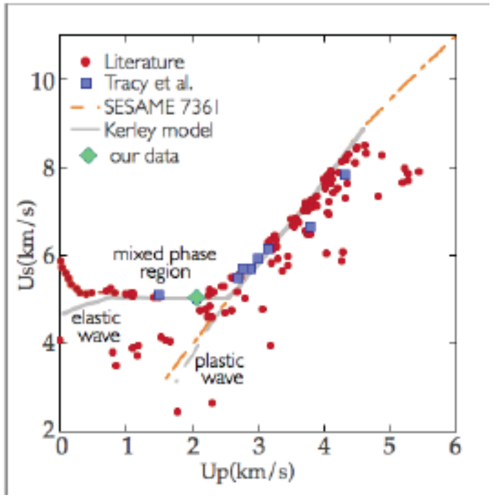




Up measurement from VISAR



Us from Us-Up relation



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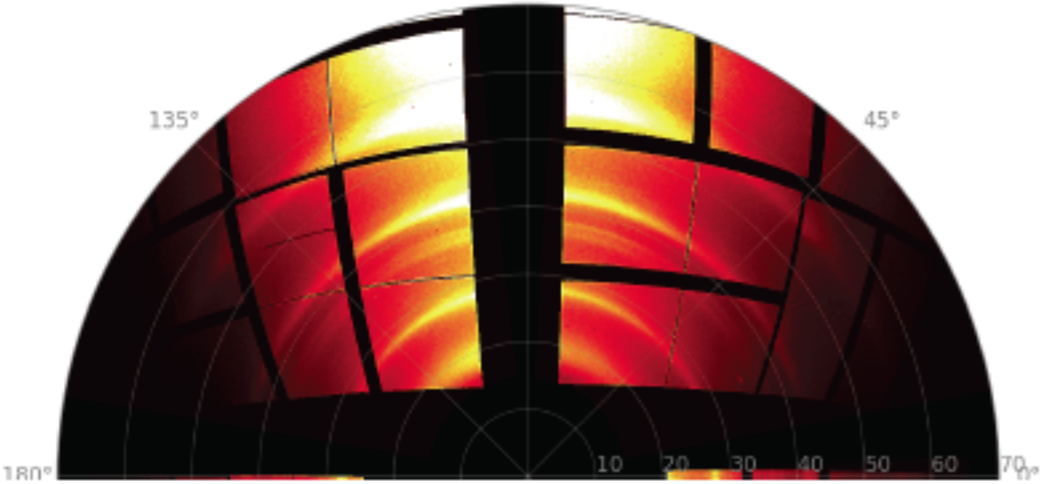
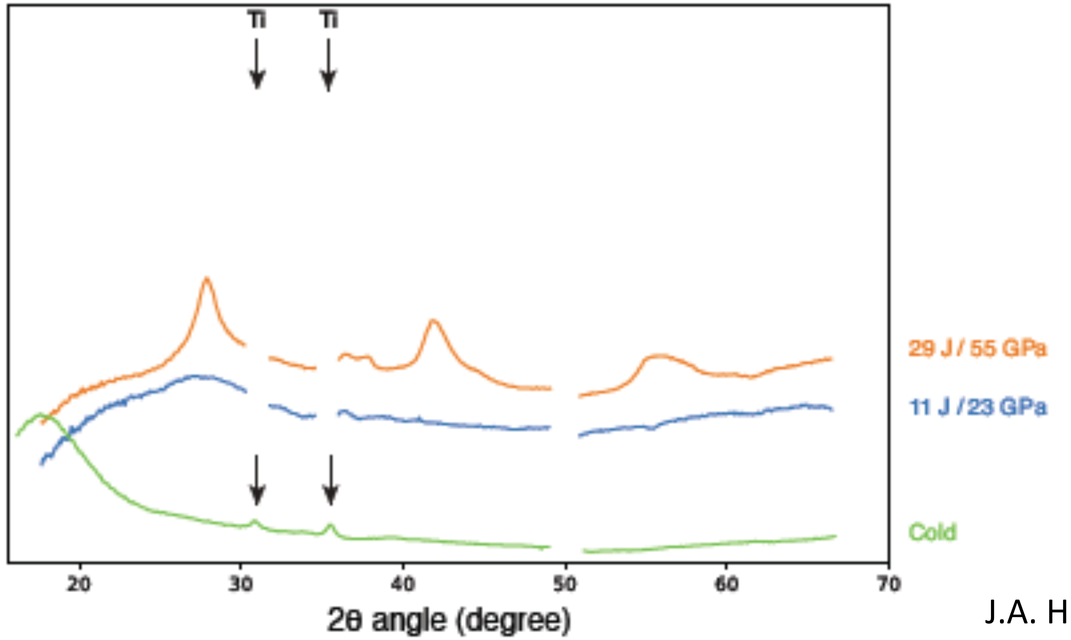
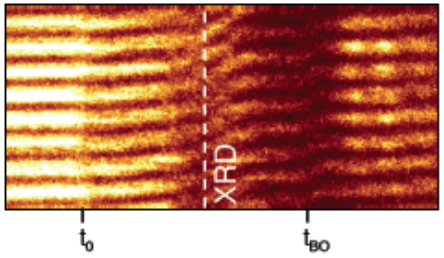
# Results on fused $SiO_2$

## Crystallization of stishovite ($TiO_2$-type structure) – 55 GPa – 4.6 ns

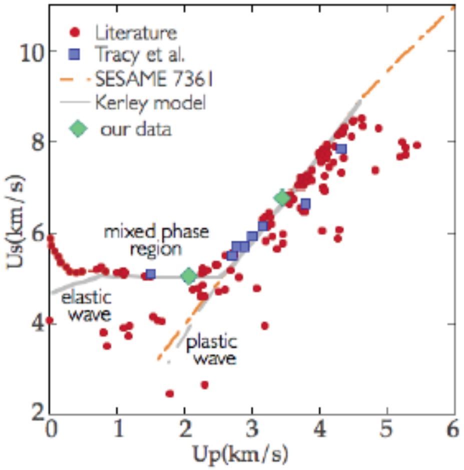




Up measurement from VISAR



Us from Us-Up relation



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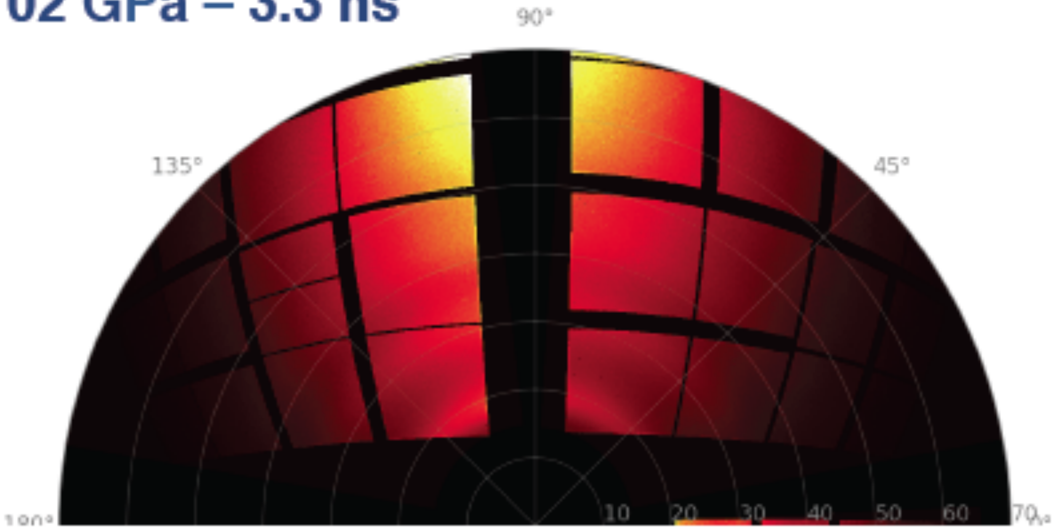
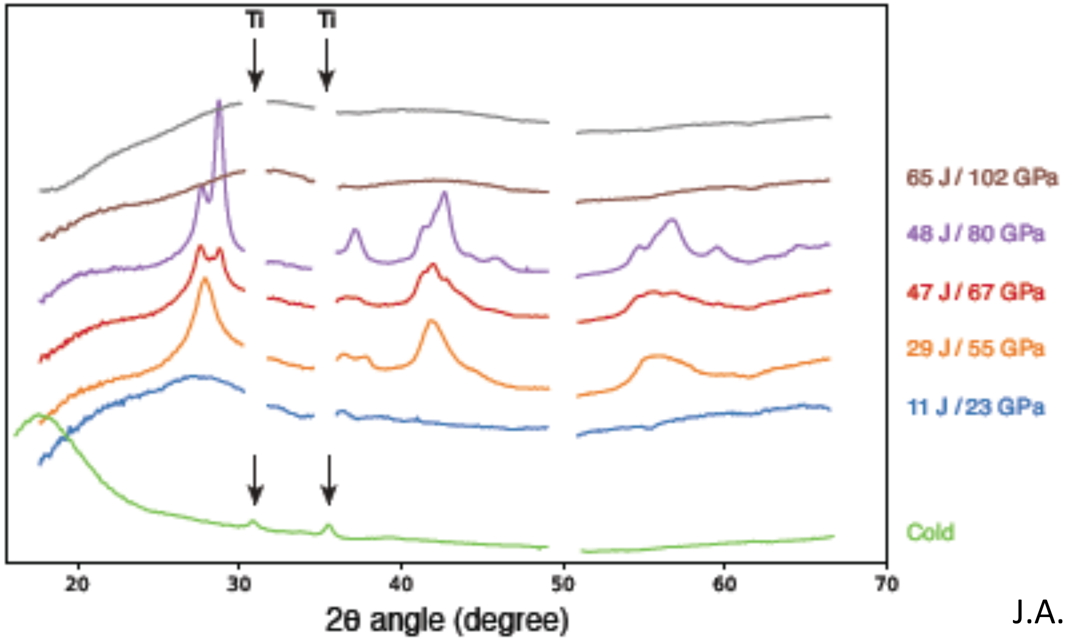
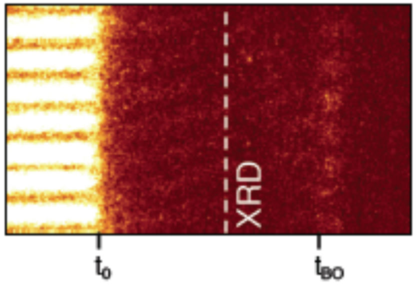
# Results on fused $SiO_2$

**Melt – 102 GPa – 3.3 ns**

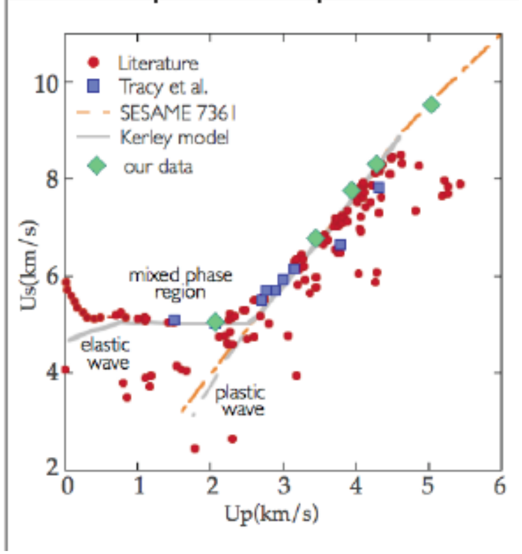




Us measurement from VISAR consistent with timing



Up from Us-Up relation



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# Conclusions

Reliable equation of state measurements using a laser shock

Studying phase transition is mandatory : Coupling with X-ray diagnostics -> complex, but very interesting results

Free electron lasers or synchrotron facilities coupled with a nanosecond laser  open an opportunity

# Collaborators

**LULI team: A. Ravasio, J. A. Hernandez, M. Guarguaglini, M. Koenig, A. Denoeud**

## Laser shock compression community laboratories

School/Graduate School of Engineering
Osaka University

cea

European XFEL

CELIA

Procédés et Ingénierie en Mécanique et Matériaux

SLAC
NATIONAL ACCELERATOR LABORATORY

INSTITUT P'
UPR 3346
Mécanique des Matériaux

ESRF
The European Synchrotron

## Geophysics and astrophysics community

IMPMC

MUSÉUM
NATIONAL D'HISTOIRE NATURELLE

HIROSHIMA UNIVERSITY

Stanford
University

l'Observatoire de Paris — LUTH
Laboratoire Univers et Théories

## Theoretical teams

cea

l'Observatoire de Paris — LUTH
Laboratoire Univers et Théories

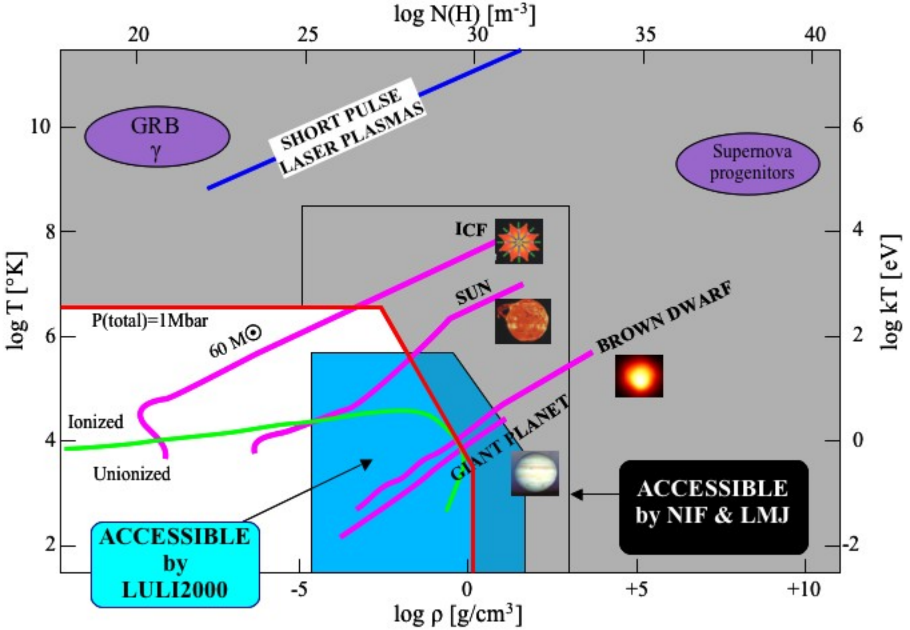
Universität
Rostock
Traditio et Innovatio

THANK YOU FOR ATTENTION

# ADDITIONAL SLIDES

# High energy density physics definition

Extreme matter states ⇔ HEDP ⇔ P > 1 Mbar (i.e $\rho/\rho_0 > 1$)



A wide range of physics area

HEDP ⇔ Energy density input ≈ energy density at room T

Typically: $10^9$ - $10^{11}$ J/m$^3$

volume involved in laser target ≈ $10^{-9}$ m$^3$

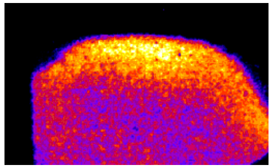
Easily accessible with a high laser power of a few hundreds J

# Experimental state of the art

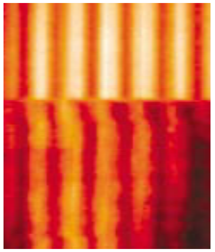
**1993** The pressure record of 0.75 Gbar *Cauble et al PRL (1993)*

**1994** High quality laser driven shocks

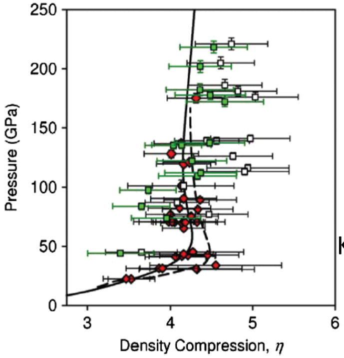
*Lower et al PRL (1994) Koenig et al PRE (1994)*



Koenig et al.
PRE (94)



Bradely et al.
PRL (2004)



Knudson et al.
PRL (2009)

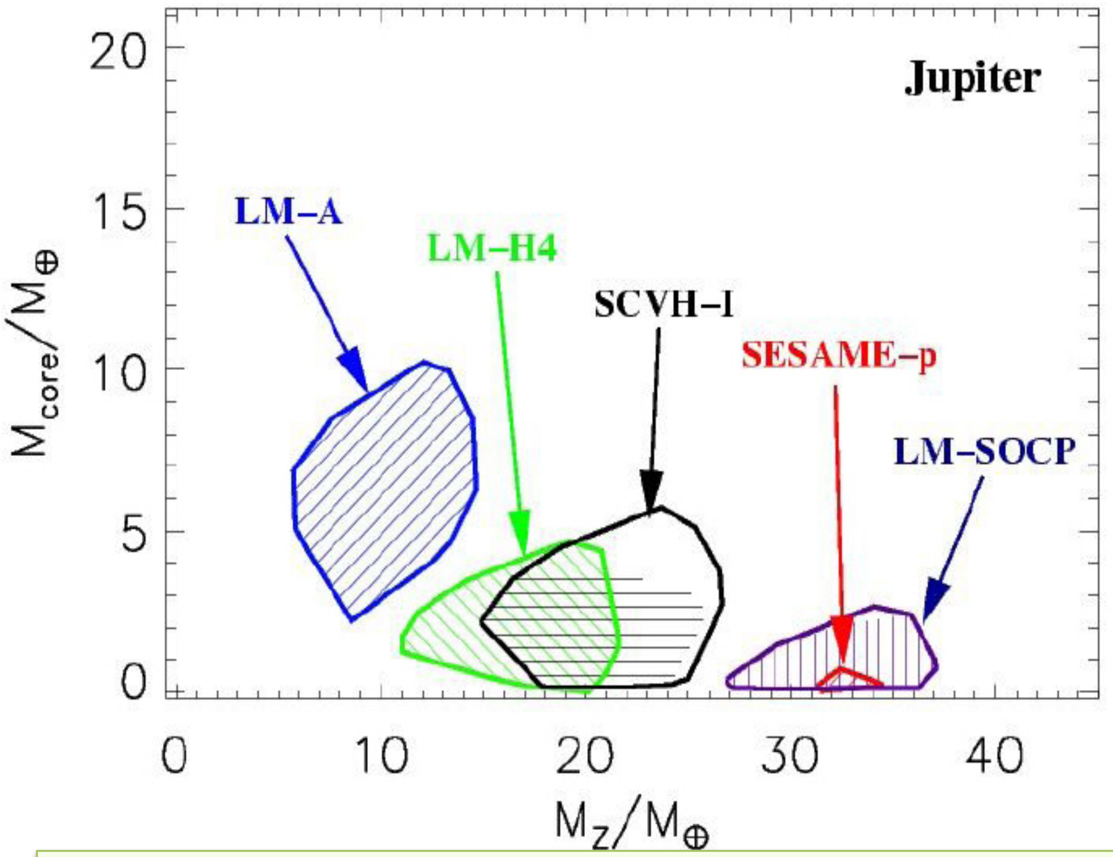
**1995** First Equation of State measurements

*Koenig et al PRL (1995) , Da Silva et al. PRL (1997)*

**1996 → today**

- Reliable Hugoniot data of interest for planetology
- Macroscopic characterization of WDM (EOS, reflectivity, conductivity, etc…)

# RESULTS FOR JUPITER



$M_+$ is earth mass, $M_Z$ envelop mass with heavy elements
Sophisticated EOS models -> ≠ answers

Core

- Accretion around solid mass

No core
Or very small

- gravitationnal collapse due to condensation

# DETECTION OF EXOPLANETS

1/ Radial velocity

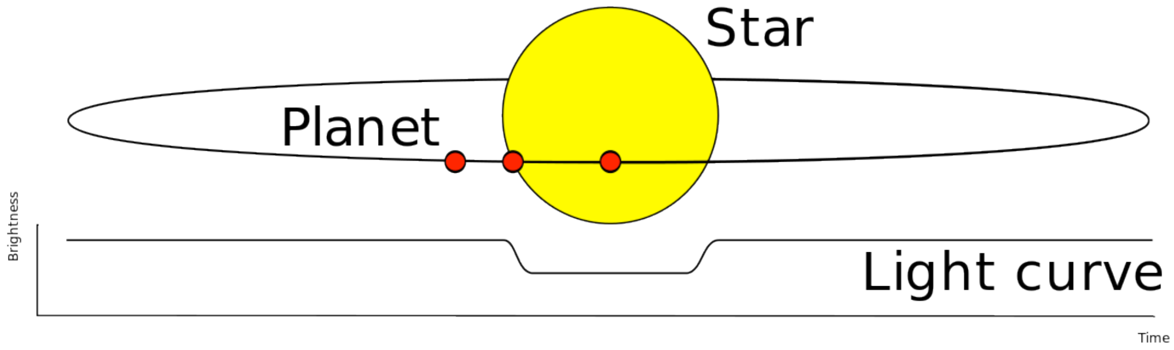
↓

Mass

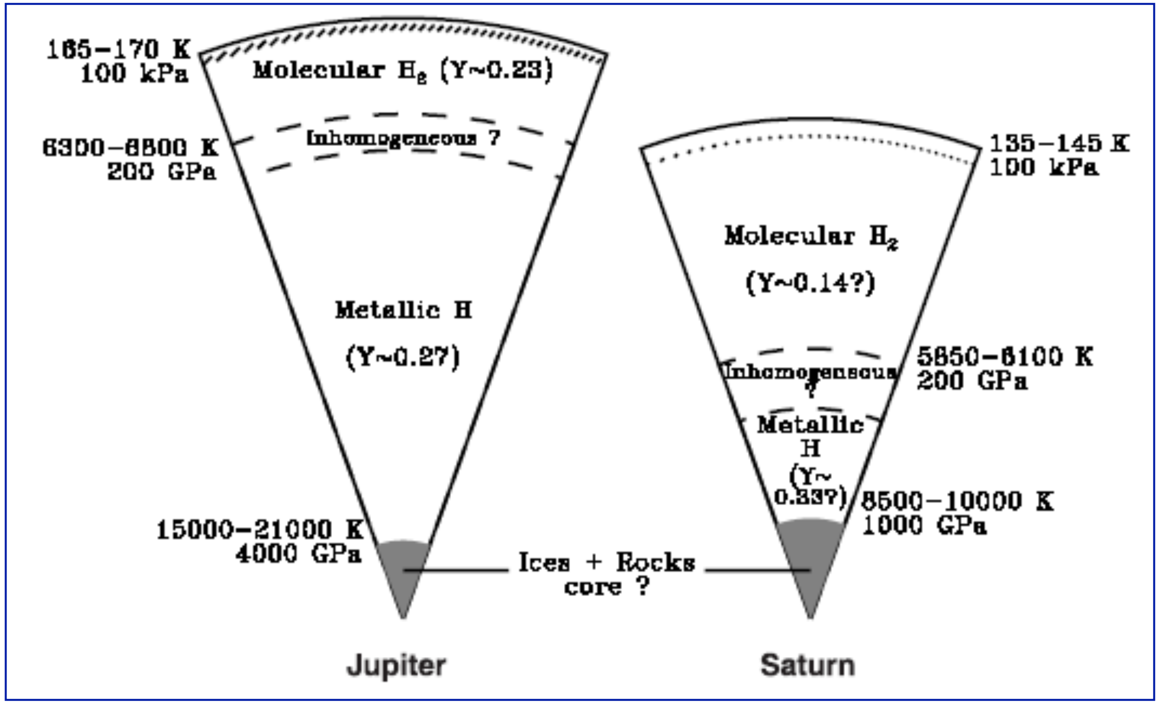
A star with a planet will move in its own small orbit in response to the planet's gravity. The radial velocity can be deduced from the displacement in the parent star's spectral lines due to the Doppler effect.

2/ Transit photometry

Radius



# Saturn is too bright



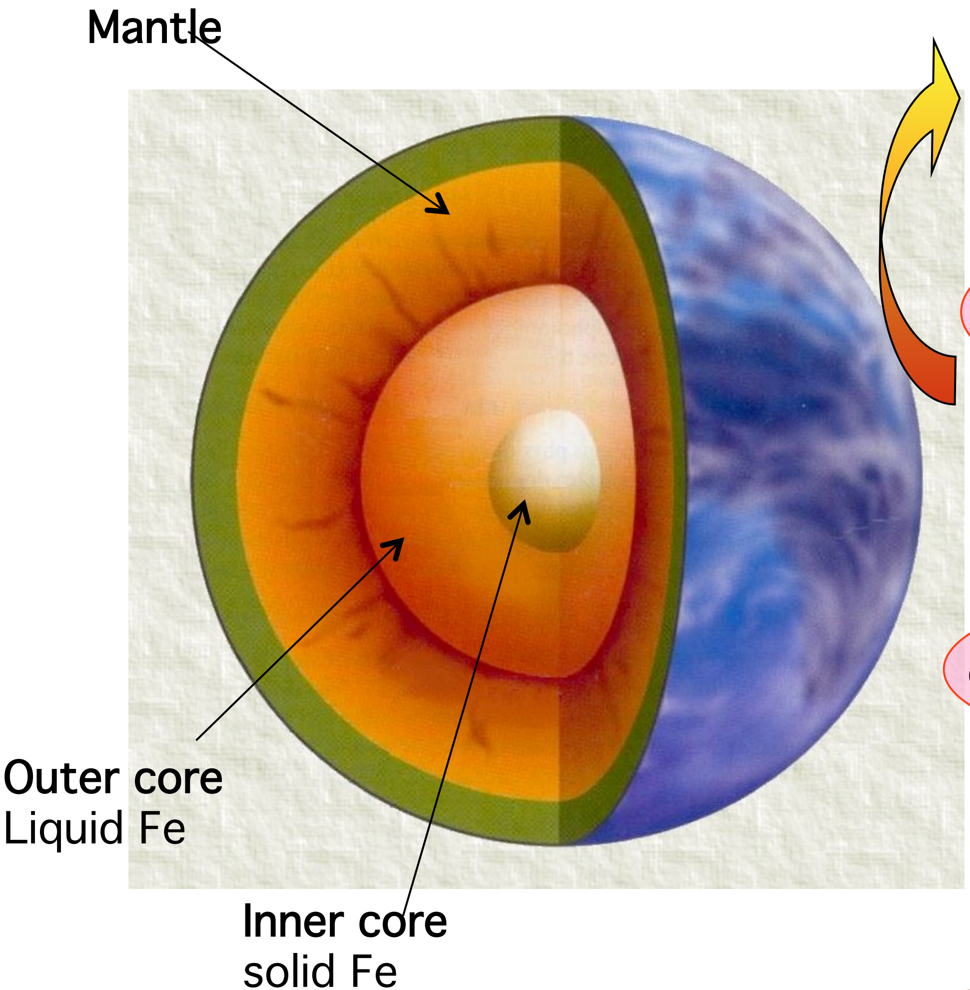
Age estimated using a homogenous model 2.4 MA

Solar system: 4.56 MA

- Problem of miscibility of He-H?

Saturn could generate some of its heat through the "raining out" of droplets of helium deep in its interior. As the droplets descend through the lower-density hydrogen, the process releases heat.

# Our comprehension of the Earth's interior depends upon the EOS of iron



Core’s thermodynamics

Emitted power by the inner core P ≈1- 4 TW

$$P = \frac{\Delta H_m \times M_{inner\ core}}{\tau}$$

Earth’s age τ ≈4.5 Gyr

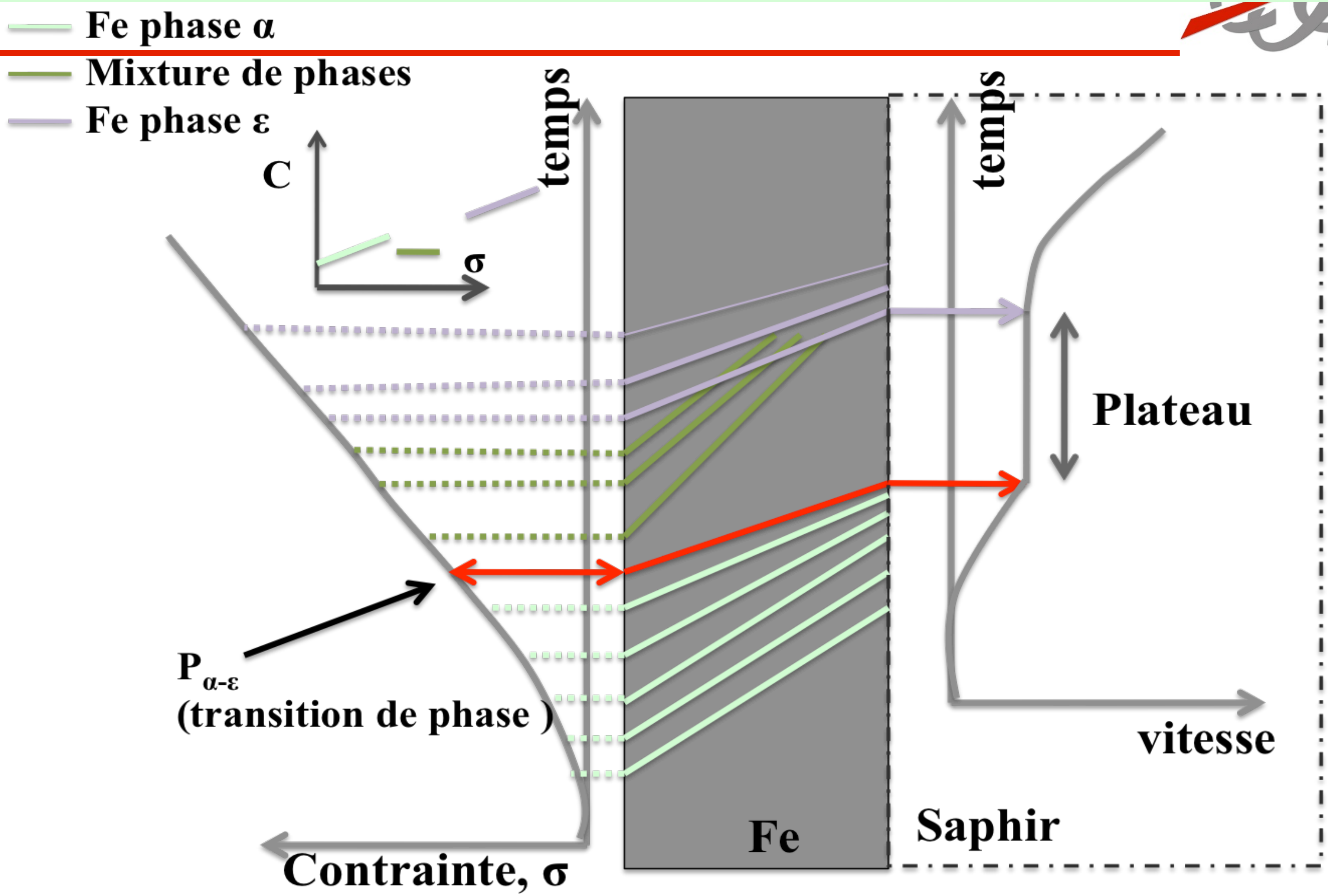
Latent heat of cristallisation ≈1- 2 MJ/ kg

$$\Delta H_m = \frac{T_m \Delta V_m}{dT_m / dP}$$

$$T_m(P)$$

Melting curve in the vicinity P = 3.3 Mbar

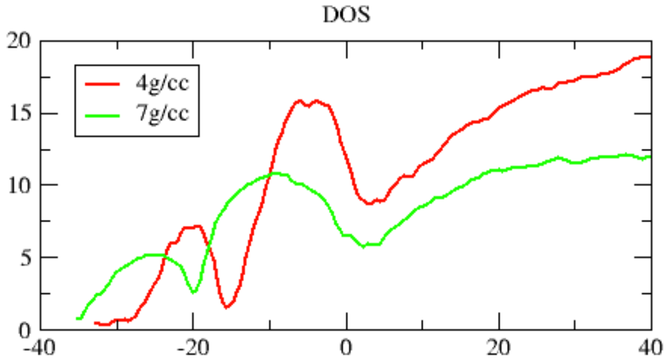
# Pourquoi la transition se manifeste t-elle par un plateau?



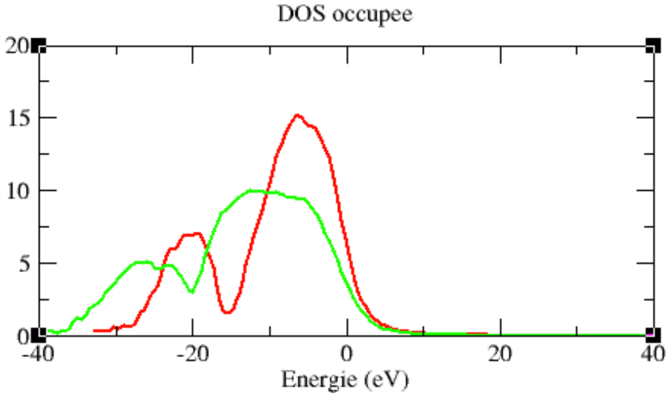
**Plateau => mixed phase**

# MODIFICATION Of THE DENSITY of STATES for the LIQUID SiO2 along the isotherm at 1 eV

Total DOS



Occupied DOS

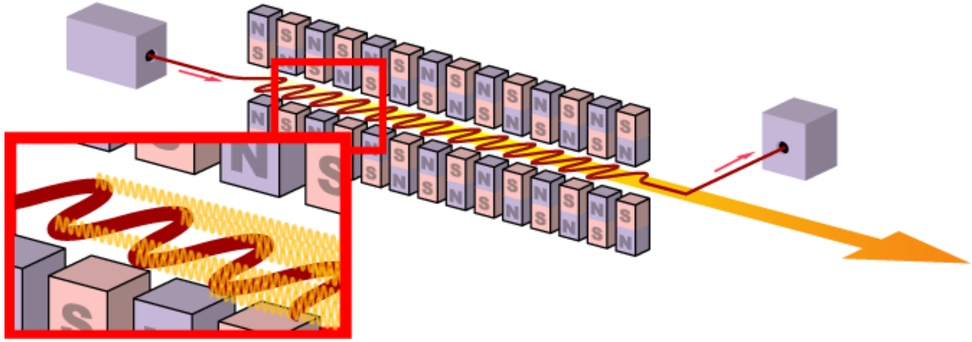


Relocalization of electron
around the Si-O units

More states with E < EF

Less free electrons → conductivity ↘

# XFEL principle

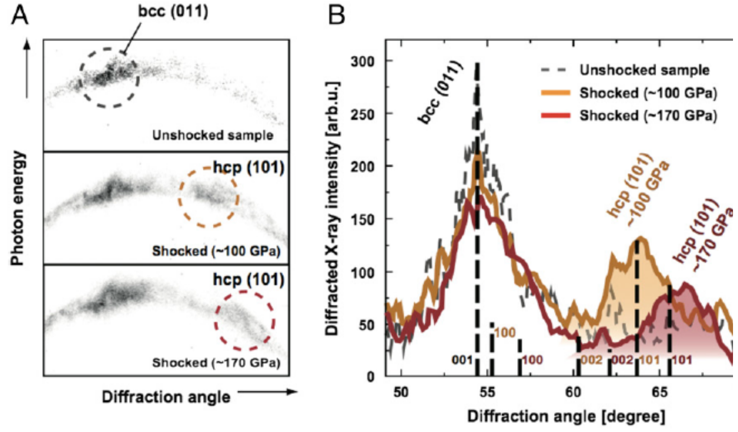


To create an FEL, a beam of electrons is accelerated to almost the speed of light. The beam passes through an undulator, a side to side magnetic field produced by a periodic arrangement of magnets with alternating poles across the beam path.

Self Amplified Spontaneous Emission → Stochastic process → spectral fluctuations

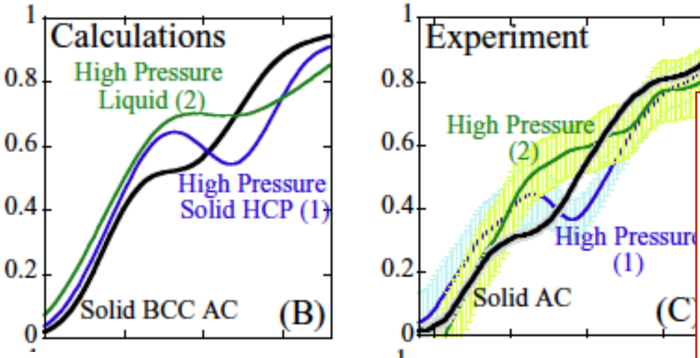
## Dynamic X-ray diffraction observation of shocked solid iron up to 170 Gpa

(Denoeud et al PNAS 2016)

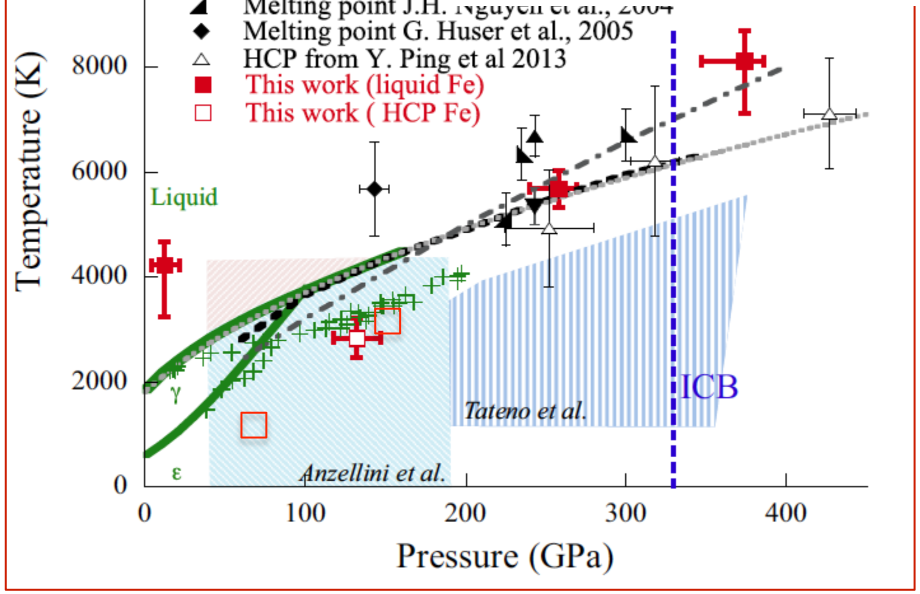


## Signature of melting at  using X-ray Absorption Near Edge Spectroscopy

(Harmand  et al PRB 2015)



*Experiment at LCLS*

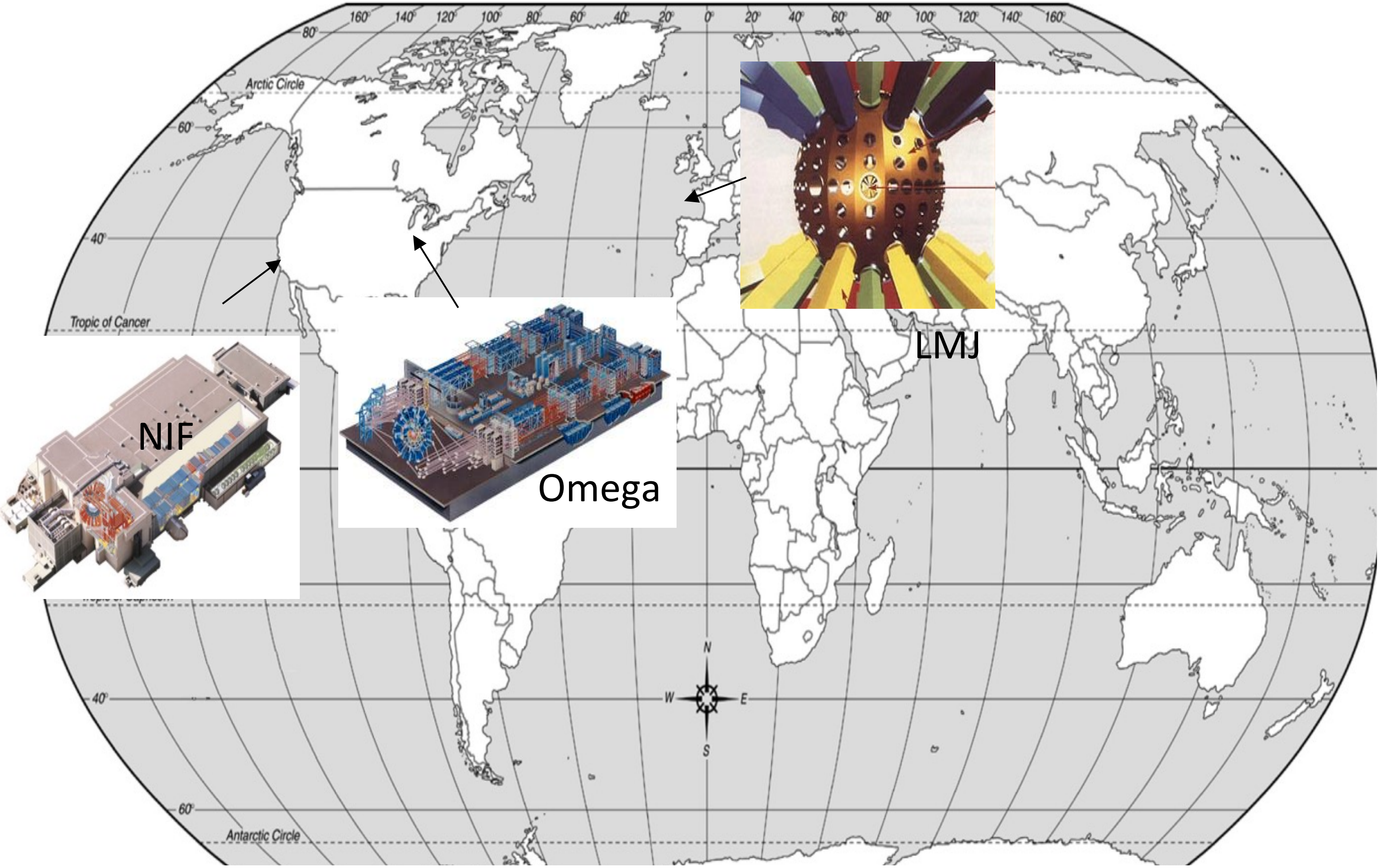


We were able to give a constrain on the melting curve

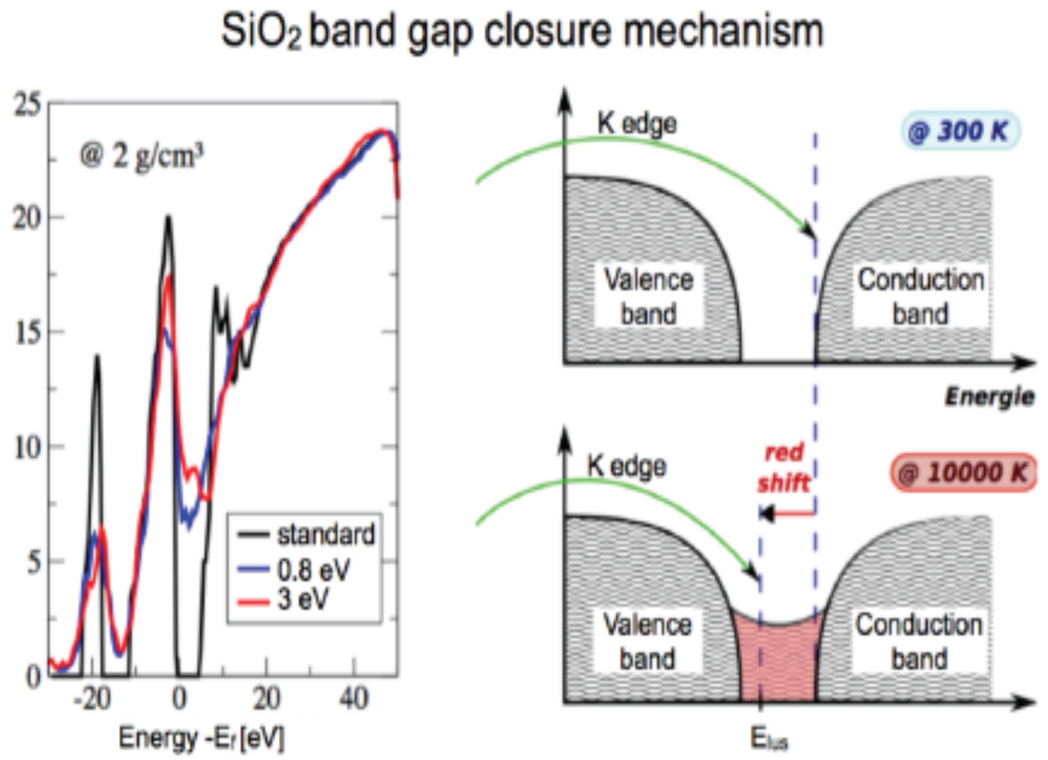
# 2 ACADEMIC HIGH POWER LASERS E≥2 kJ

Nanosecond pulses ($10^{-9}$ s)
Up to MJ energy

Short Pulse ≥ 1 to 4 kJ 1 ps



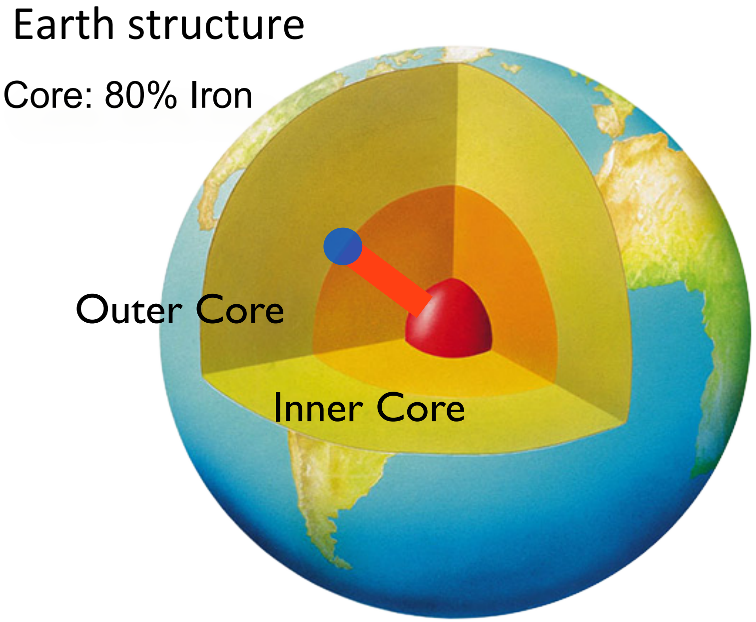
# SiO2



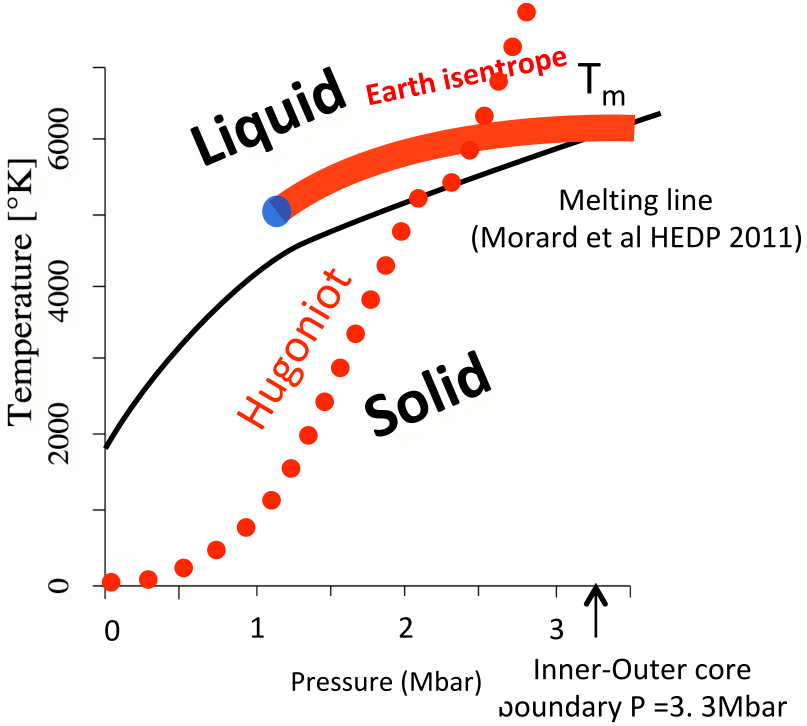
SiO₂ band gap closure mechanism

# Why do we need to develop quasi-isentropic compression?

**To get closer to planetary isentropes**



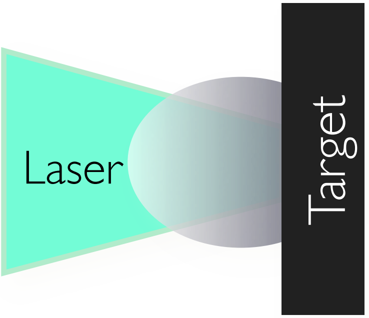
e.g. : the case of iron

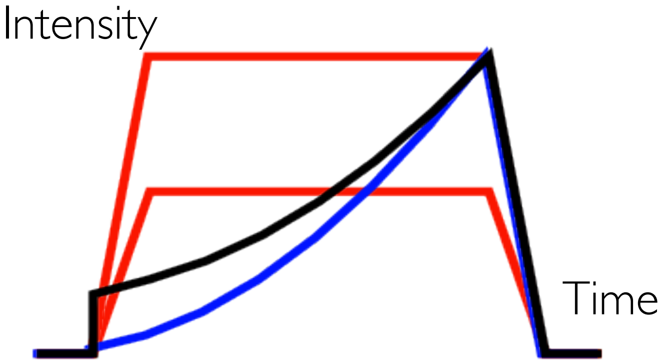


$T_m$ -----> important repercussions: convection, B….

The melting curve $T_m$ (P) has been recently calculated -----> Experimental validation

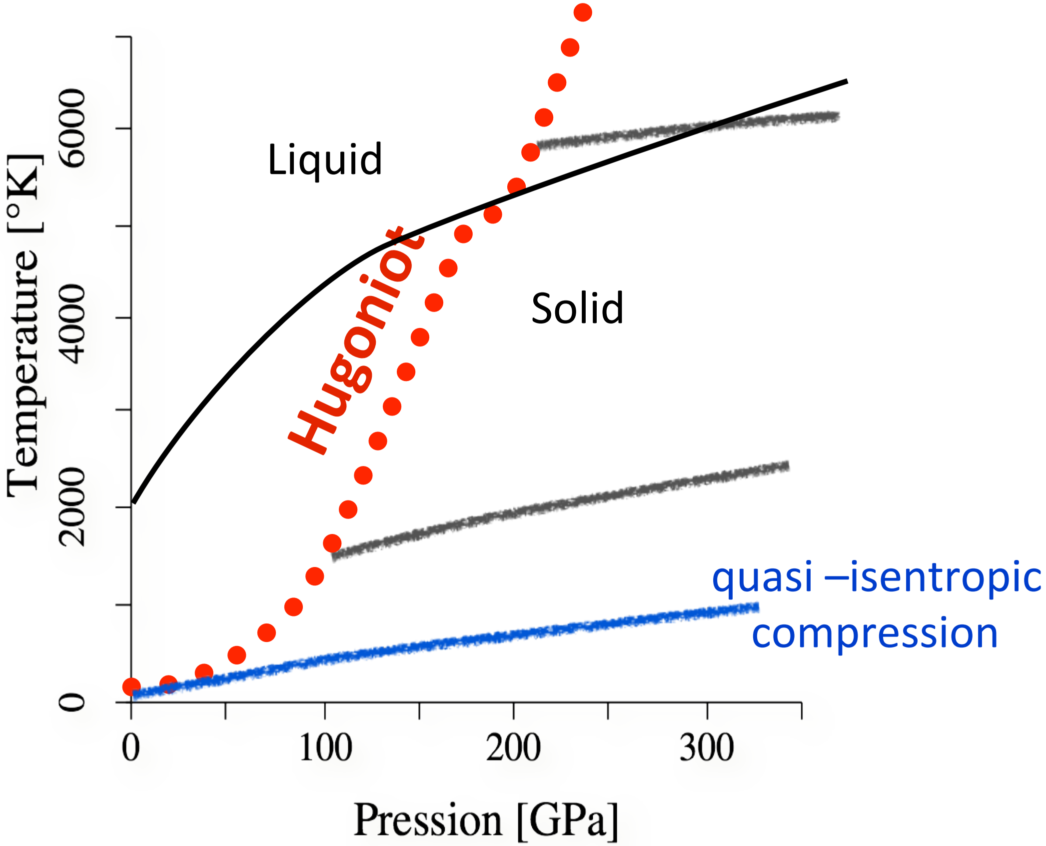
# Shock compression *versus* quasi isentropic compression





**SHOCK**

- **Irreversible processus => $dS \neq 0$ =>**
  - **the heating is strong**
- **Compression is limited (~ $4\rho_0$)**

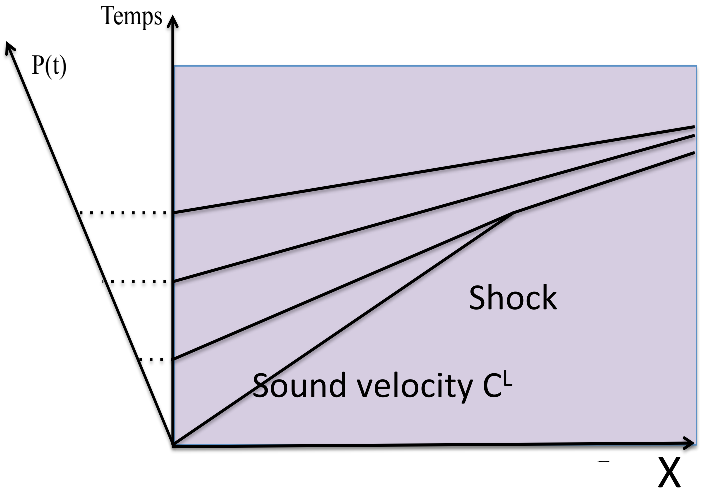


**QUASI-ISENTROPIC COMPRESSION**

- **Reversible processus =>**
  - **$dS \approx 0$ => low T**
- **High compression**

# The critical points for quasi-isentropic compression

Generation of a sequence of small compressions waves but multi Mbar pressures must be reached



↓

Very smooth and long laser profile

↓

Long compression time to maintain P

↓

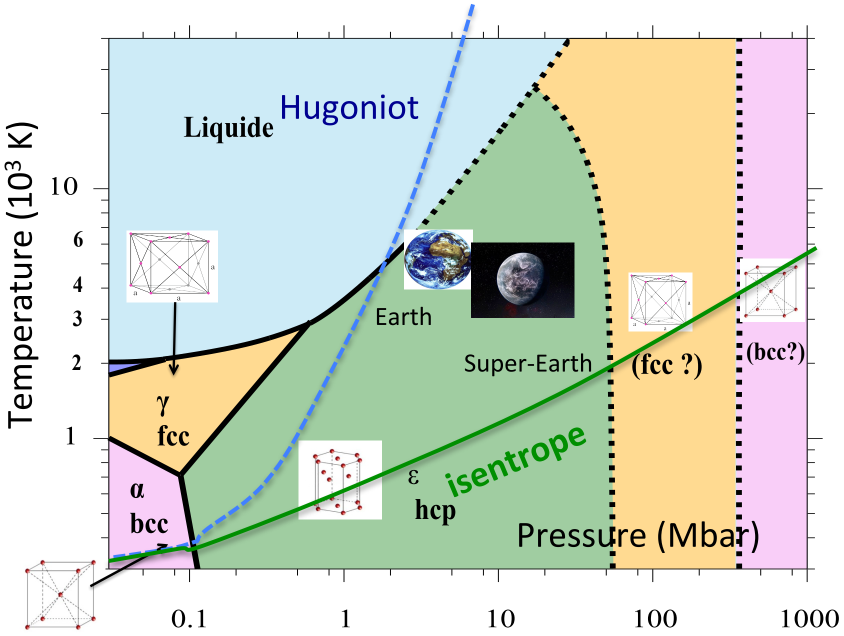
Thick sample, large focal spot

**A multi-kJ laser is necessary to achieve P > 10 Mbar**

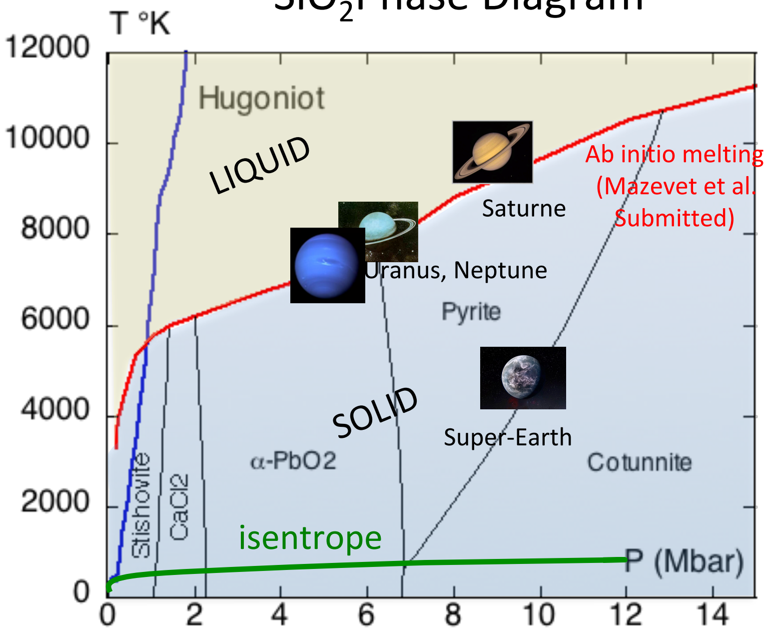
# Experiments at LIL in Bordeaux on iron and $SiO_2$

(the most energetic European laser)

Iron Phase Diagram



$SiO_2$Phase Diagram



Phase diagrams based on ab initio simulations (Morard 2011, Mazevet submitted, Sitxrude 2012)

The main goal: to reach Super-Earth and giant planets conditions and to investigate region around the melting

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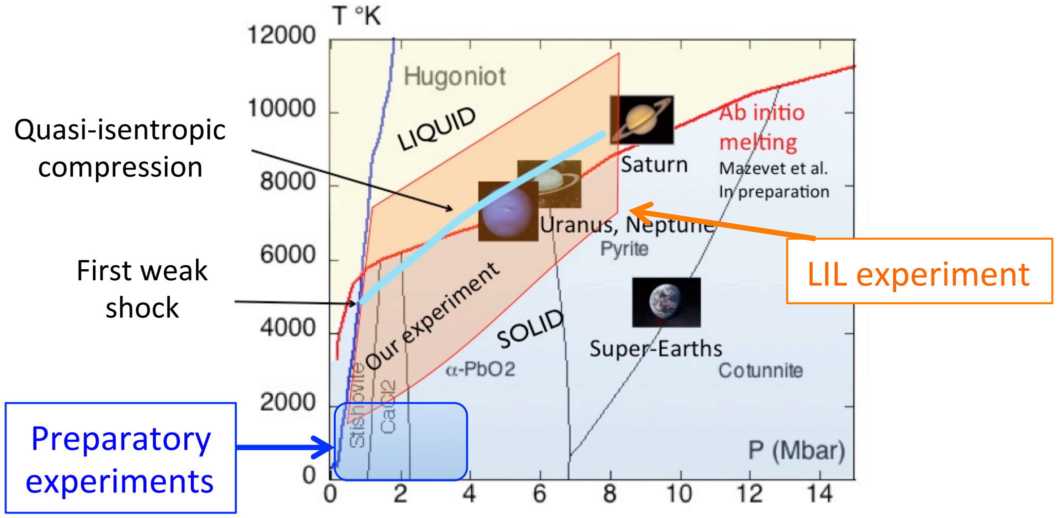
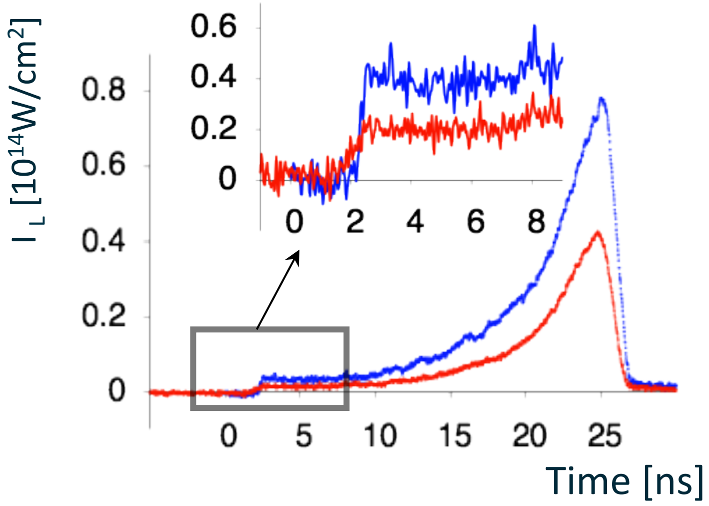
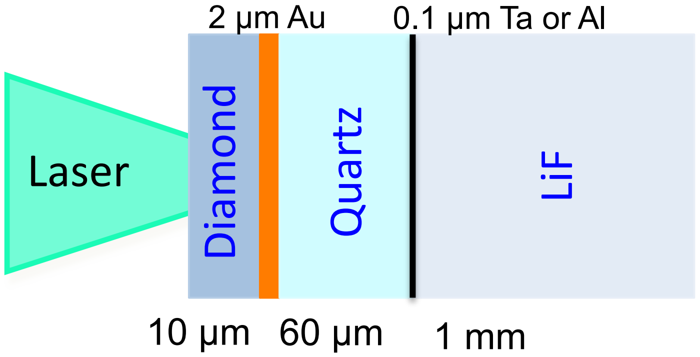
# Campaign at LIL: goal and principle

Novel technique coupling a *well-controlled* shock followed by a quasi-isentropic compression.



- ✓ Achieve planetary conditions and the melting region (Mazevet et al. submitted to PRL)





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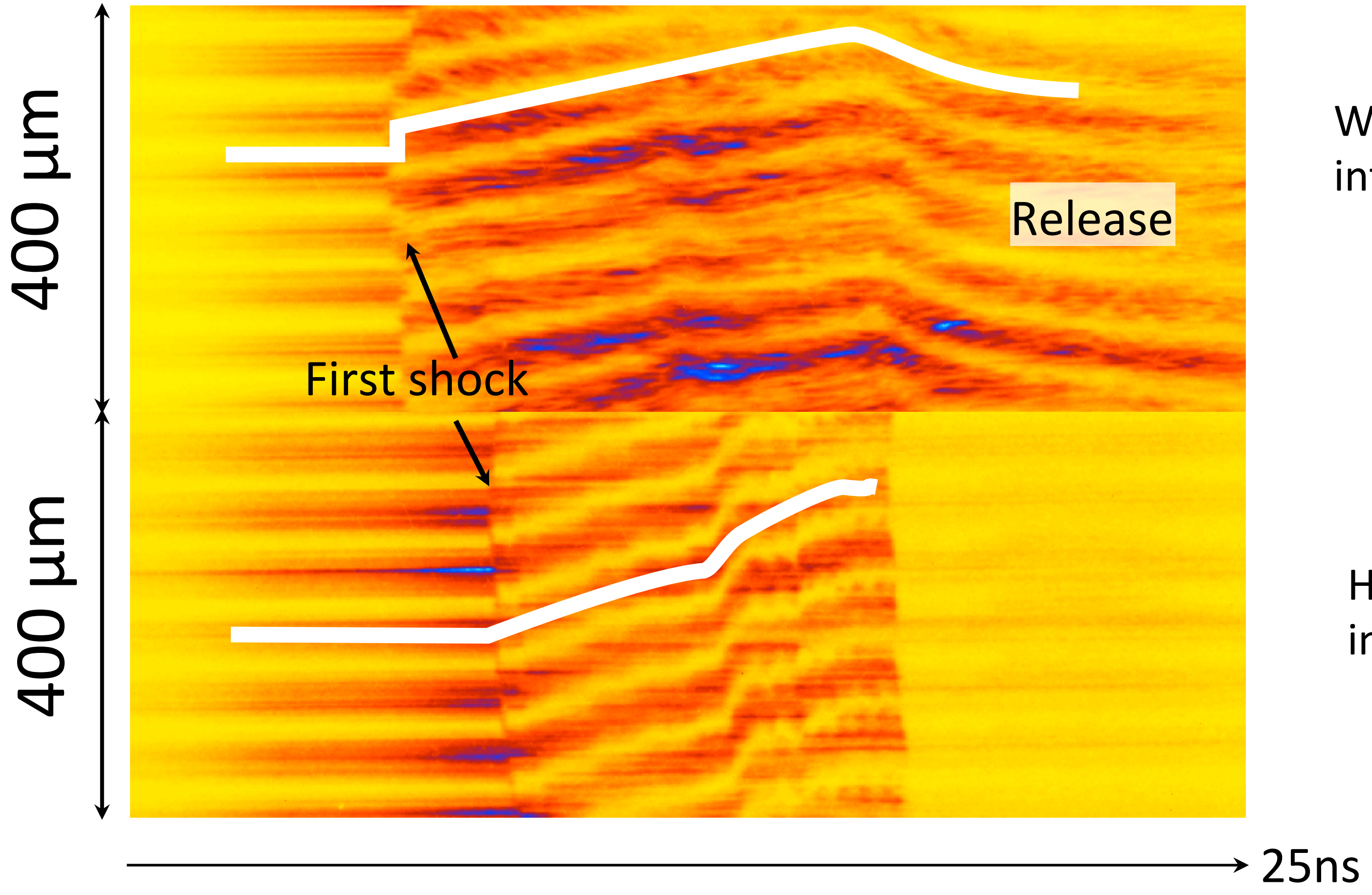
# VISAR images



Weak intensity

1 kJ

High intensity

4 kJ

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# Results: the thermodynamical path (P,T)

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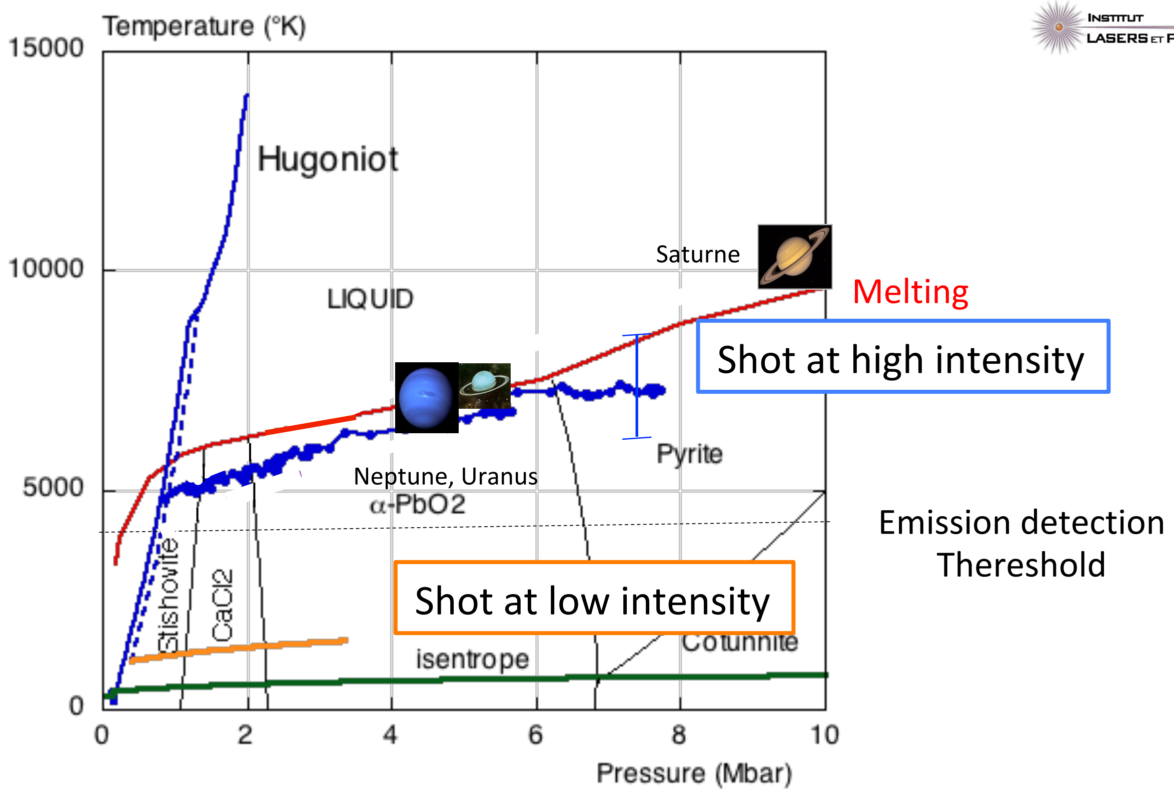
$U_p \longrightarrow$ **P**

Self-emission + R

↓

**T**



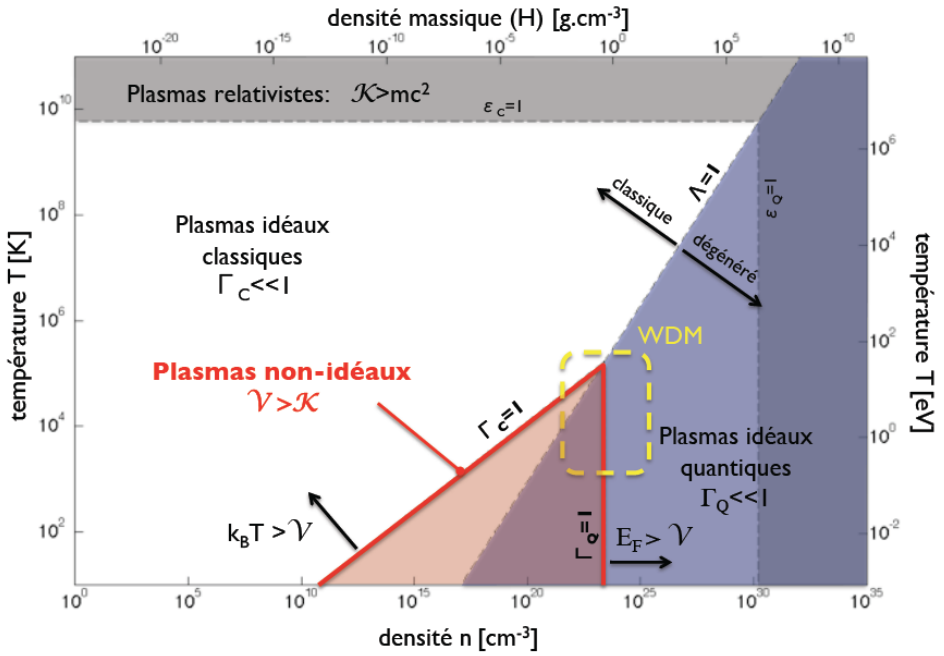
Melting

Shot at high intensity

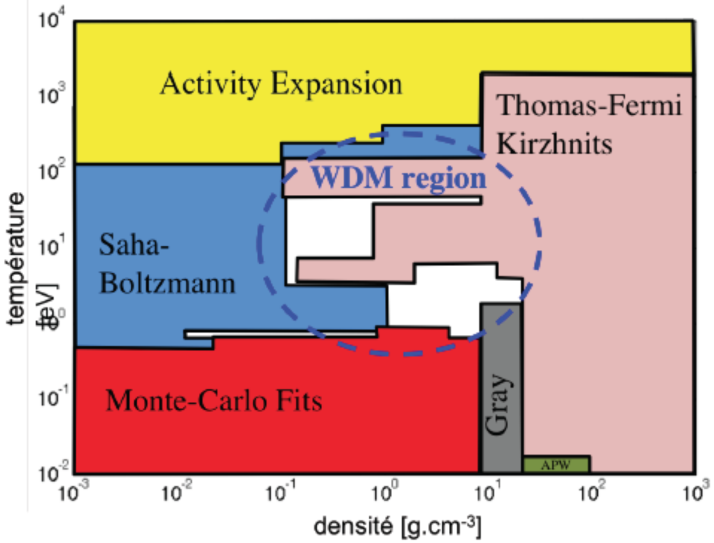
Shot at low intensity

Emission detection Thereshold

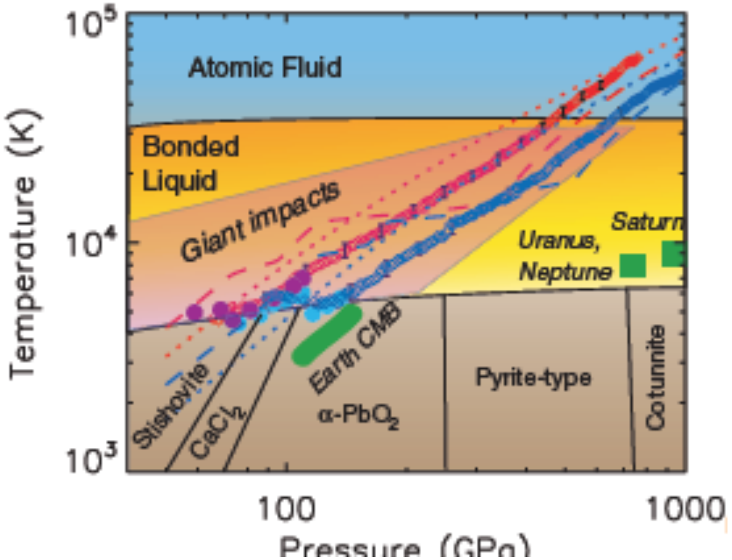
# Modèles théoriques



Electrons , comme système de fermions qui interagissent par un potentiel (champ moyen)



# A scenario for $SiO_2$ (Hicks et al. PRL 2006)



Specific heat capacity
deduced by the slope of Hugoniot T

$C_V$ increases above melting

$C_V$ shows a broad peak (yellow region)

↕

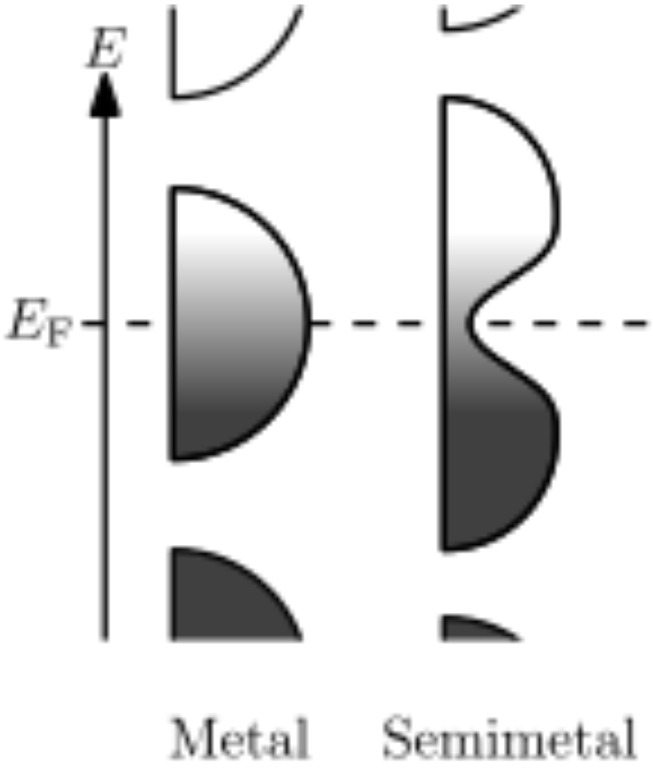
Energy required to break all chemical bonds

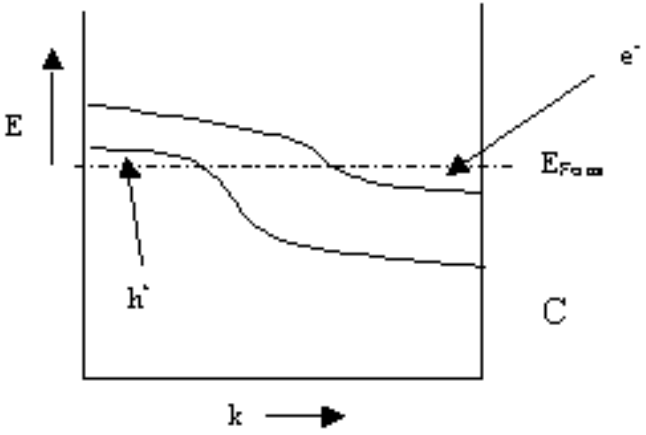
For higher T (blue region) : bond breaking is essentially complete

Yellow region: « bonded liquid »= liquid containing a mixture of variously sized polymerized structural units

Blue region: « atomic fluid » = no anymore chemical bonds

# CONDUCTION BAND STRUCTURE for a METAL and SEMIMETAL

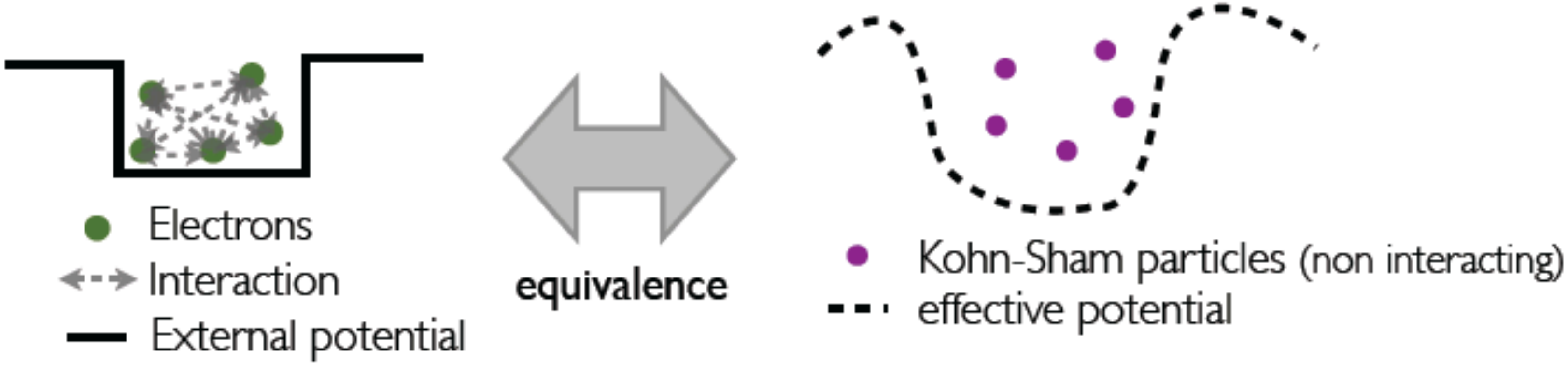




Semimetal band strucutre in k

# DENSITY FUNCTIONAL THEORY (DFT) & QUANTUM MOLECULAR DYNAMICS CALCULATIONS (QMD)

- **DFT** is a formally exact representation of the N electron Schrödinger Equation



● Electrons
⇠⇢ Interaction
— External potential

**equivalence**

● Kohn-Sham particles (non interacting)
- - effective potential

©Desjarlais

- QMD:
  - Born-Oppenheimer separation of electrons and ions
    - Electrons are treated using DFT (quantum or Ab initio)
    - Ions are advanced solving Newton equations and using the quantum forces created by the electrons (Molecular dynamics)
  - Periodic simulation **cell with N** *(162)* **atoms**
  - Constant volume and temperature
  - Use of pseudo-potentials (e.g. PAW)

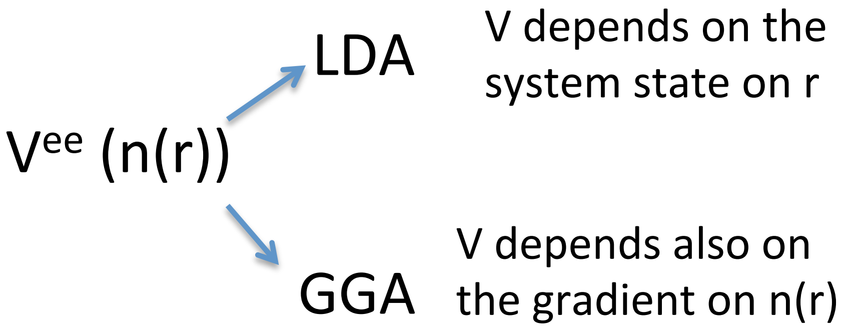
# Density Functional Theory

Any property of a system of many interacting particles can be viewed as a functional of the ground state density $n_0(r)$

## "Kohn-Sham" theory

It replaces the original many body problem by an auxiliary independent particle problem, which evolves in an external potential

External effectif potential



$V^{ee}(n(r))$

LDA

V depends on the system state on r

GGA

V depends also on the gradient on n(r)

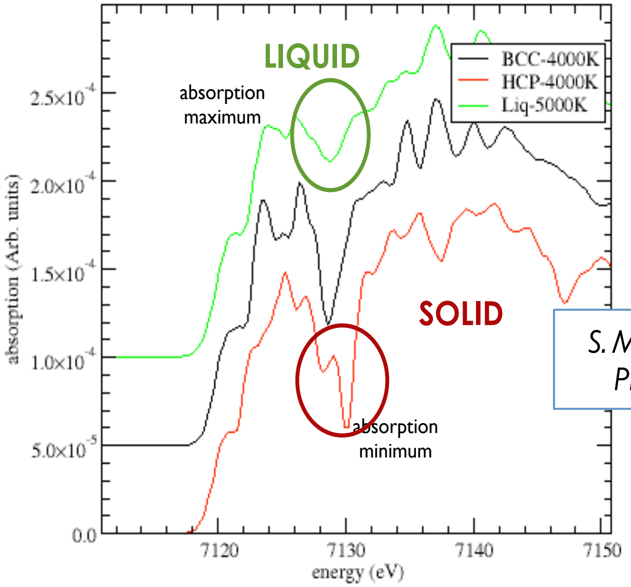
# XANES as a melting diagnostic for Fe

(experiment at LCLS)

**Context** Detection of iron melting at high pressures (geophysics)

ANR PLANETLAB & IRONFEL

- Ab initio calculations suggest XANES as a structural diagnostic for iron at planet cores conditions, including melting
- Disappearance of the deep absorption minimum

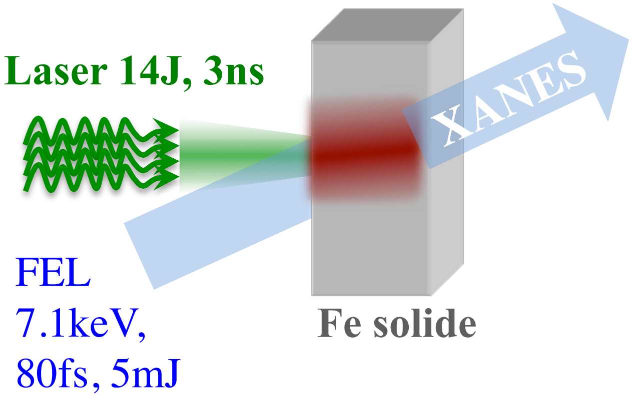


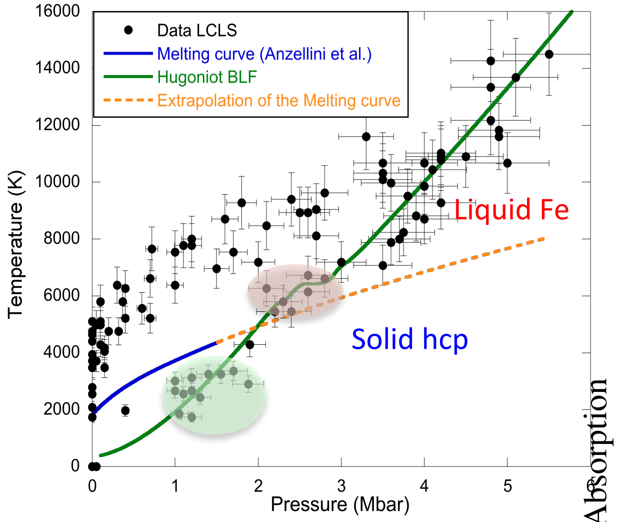
*S. Mazevet et al. PRB (2014)*

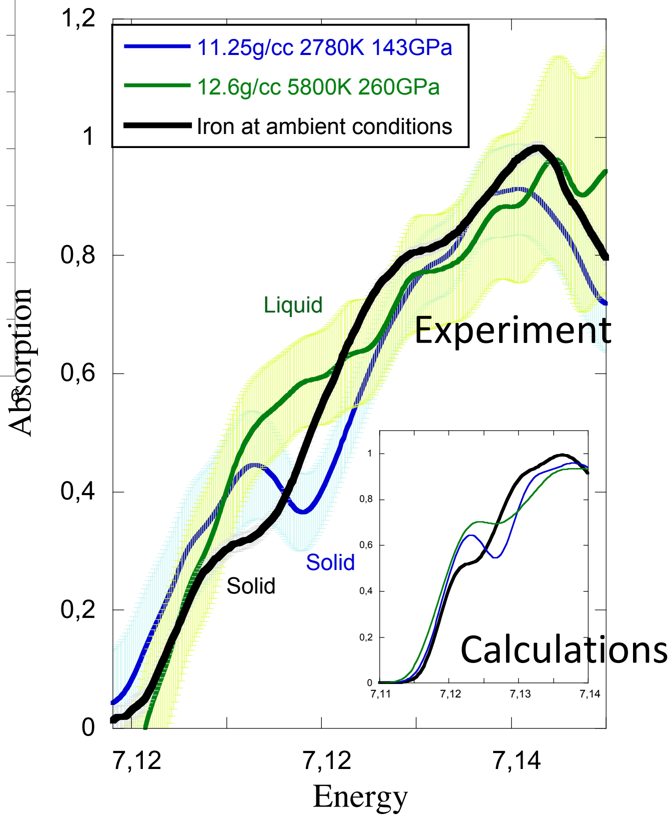
**Requirements** -> **Fe K-edge @ ~ 7.1 keV**

- Need a broad band, intense, short source ≈ 7 keV ⇒ not achieved with UHI lasers
- need a huge amount of energy to create the X-ray source (*Yaakobi et al. 2003, Ping et al. PRL 2013* ).
- Alternative approach ⇒ XFEL

# Some results







Good agreement between experiments and calculations for the evolution of the spectra (cold bcc/compressed hcp/compressed liquid)

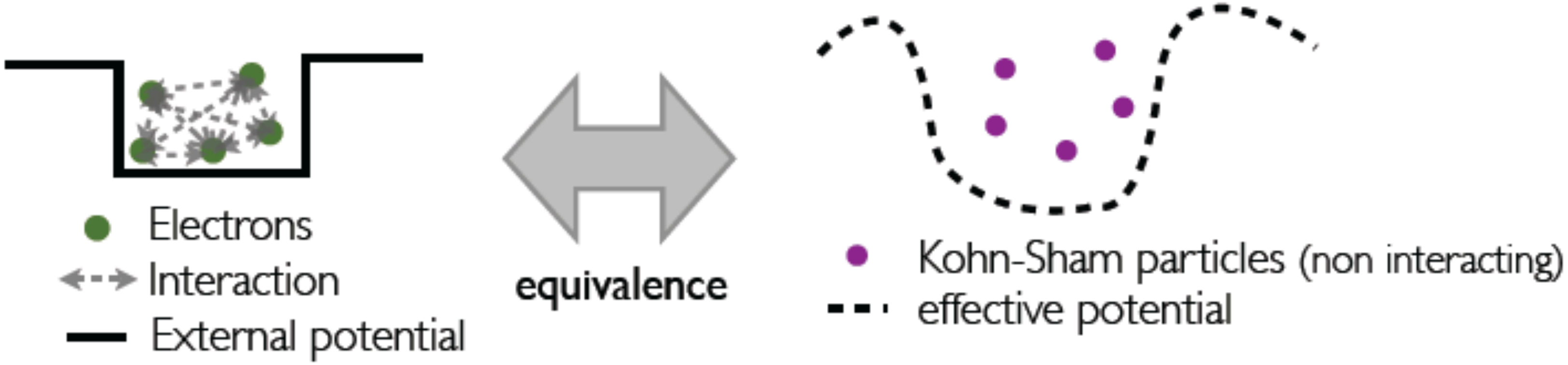
Data confirm that XANES is a good diagnostic to constrain the melting

(Harmand & Ravasio et al in preparation)

SUPPORTED BY ANR
PLANETLAB
&
IRONFEL

# DENSITY FUNCTIONAL THEORY (DFT) & QUANTUM MOLECULAR DYNAMICS CALCULATIONS (QMD)

- **DFT** is a formally exact representation of the N electron Schrödinger Equation



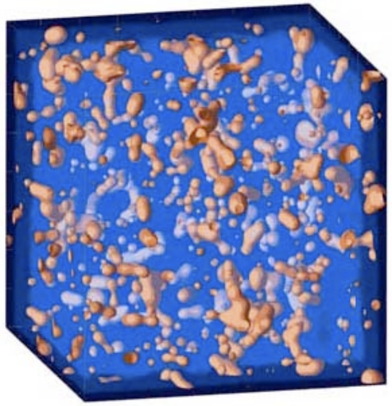
©Desjarlais

- **QMD**:
  - Born-Oppenheimer separation of electrons and ions
    - Electrons are treated using DFT (quantum or Ab initio)
    - Ions are advanced solving Newton equations and using the quantum forces created by the electrons (Molecular dynamics)
  - Periodic simulation **cell with N** *(162)* **atoms**
  - Constant volume and temperature
  - Use of pseudo-potentials (e.g. PAW)

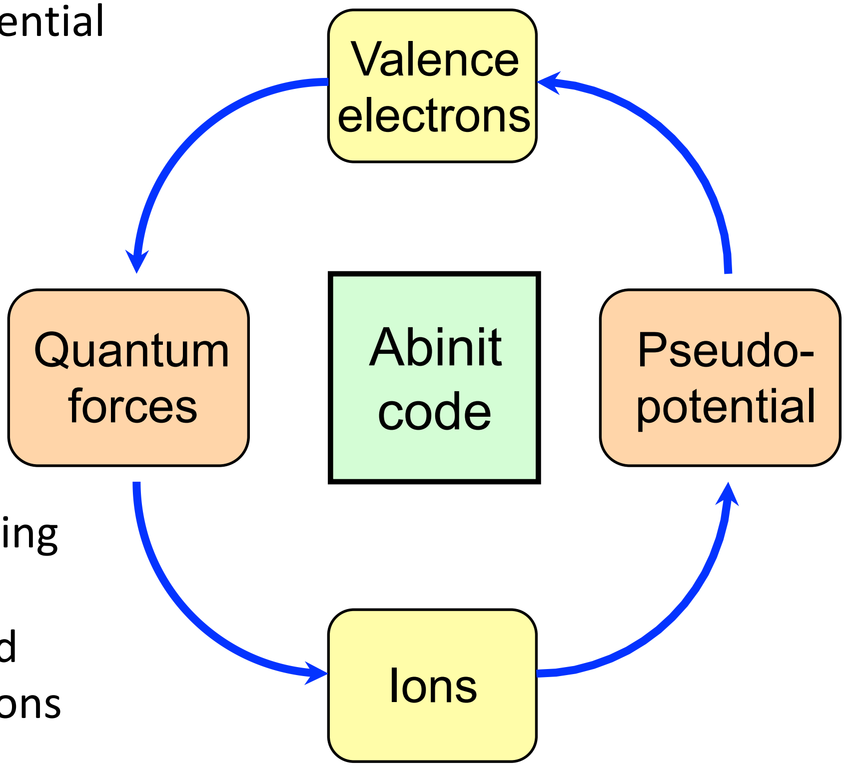
# Quantum Molecular Dynamics calculations

- Principle :
  - atoms are set in a cubic box with periodic limit conditions.

Given a ionic configuration → electronic structure which will induce a rearrangement of ions by quantum forces. The electrons interact with a pseudo potential



Electronic density of $D_2$ @ 1g/cc and 29.000 K



Valence electrons

Quantum forces

Abinit code

Pseudo-potential

Ions

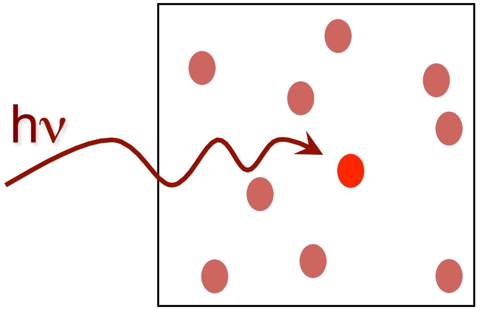
S. Mazevet et al., Phys. Rev Lett. **101** (2009) 155001

• Electrons are treated quatum mechanically using DFT → ***ab initio***

• Ions are moving solving Newton equations and using the quantum forces created by the electrons

→ **Molecular Dynamics**

# Quantum Molecular Dynamics calculations (2)



- Assumptions :
  - the number of atoms is fixed.
  - the simulation box size is calculated in order to reproduced the density.
  - the number of energy level calculated is fixed.

  - **electrons** are treated by **quantum mechanics** using the Density Functional Theory (DFT).
  - the **ions** motion is deduced from **Newton equations** using quantum forces created by electrons.

  - **finite temperature DFT** : occupations are given by Fermi-Dirac distribution.
  - **PAW (**Projector Augmented Wave) **formalism** : interaction between the valence electrons and core electrons + nucleus

  - absorption spectra are calculated applying the linear response theory(Kubo-Grenwhood formulation) and with the impurity model : the absorbing atom is in an excited configuration with 1 hole in the 1s level (the core hole has an infinite life time).

  Frozen core approximation: the core electrons are frozen

# PAW method

2 types of electrons :

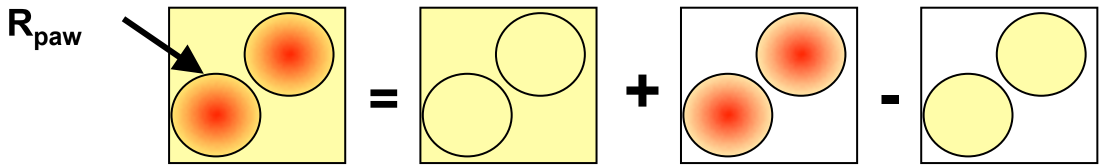
-**core electrons** → the core electronic wavefunctions $\phi_i(r)$ (isolated atomic wavefunction) are obtained using DFT -

**valence electrons** → the true wavefunctions are replaced by pseudo-wavefunctions for r < $R_{PAW}$

- the pseudo-wavefunction satisfy $\phi_i(r) = \tilde{\phi}_i(r)$ for r > $R_{PAW}$

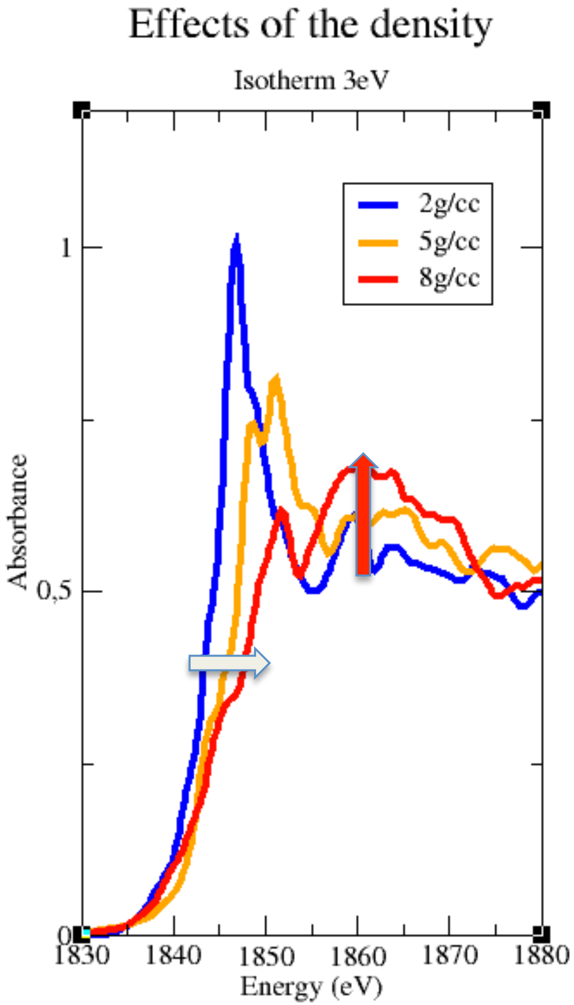
The **real valence wavefunction** is **known**, even in the core region, and related to the pseudo-wavefunction by a linear operator

$$\left|\psi_{i,k}\right\rangle = \left|\tilde{\psi}_{i,k}\right\rangle + \sum_{R,n}\left(\left|\phi_{R,n}\right\rangle - \left|\tilde{\phi}_{R,n}\right\rangle\right)\left\langle \tilde{p}_{R,n} \middle| \tilde{\psi}_{i,k}\right\rangle$$



→ **PAW calculations are equivalent to all-electrons calculations**

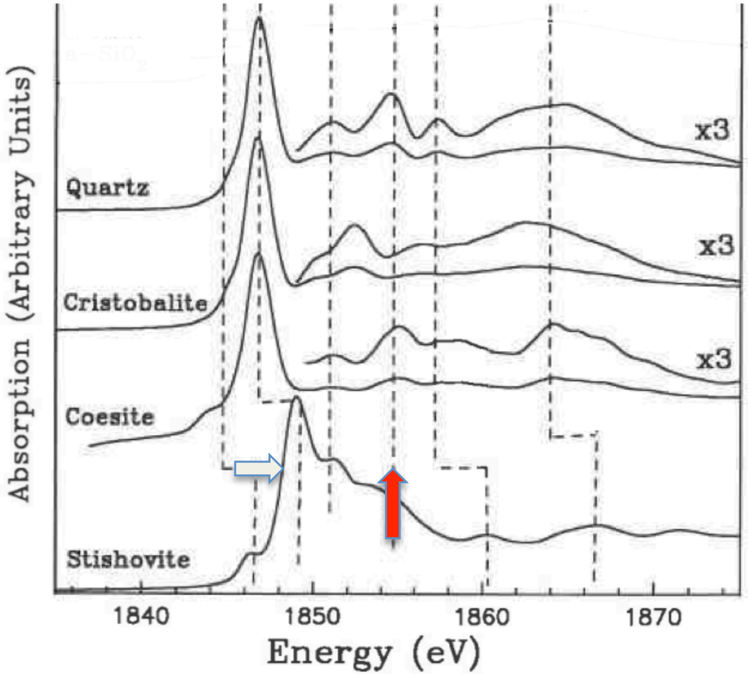
# Variations in density…



- *Blue shift with the density as in the experiment*
- *Like in the solid : blue shift with increasing coordination*

*The shift is a signature of the coordinance*

*Appearance of new 'structures' in XANES spectra*

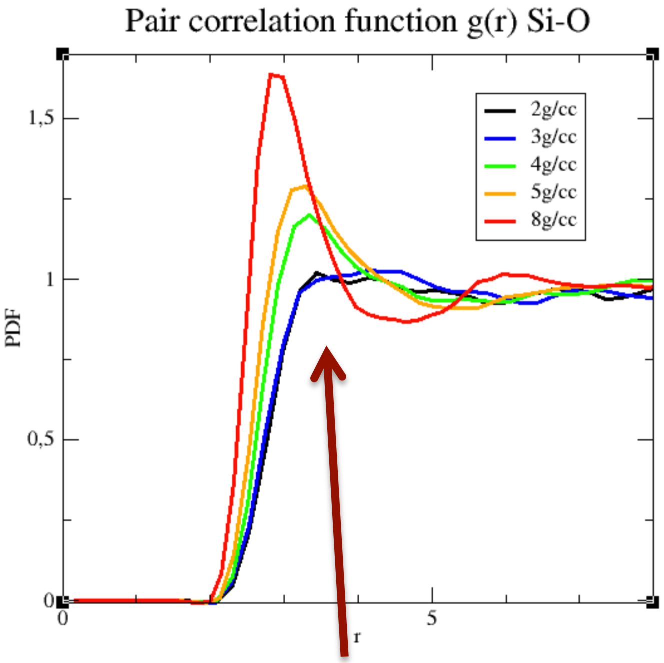


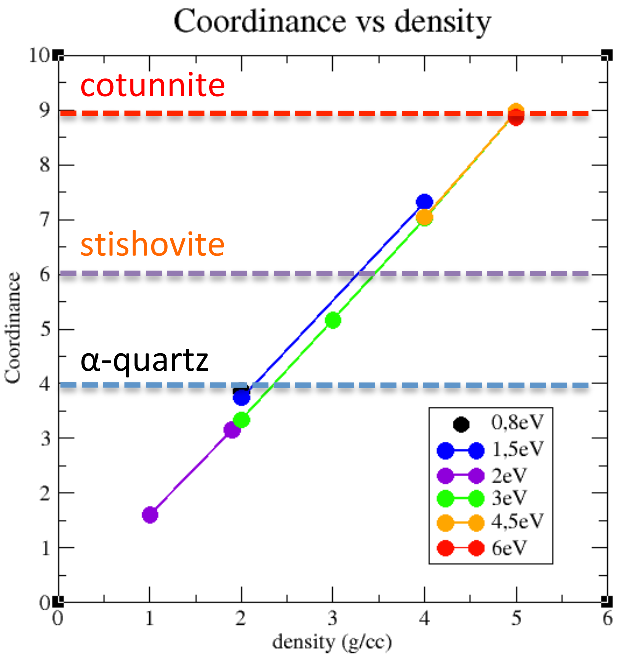
**Synchrotron data** *: Li et al, American Mineralogist 79, 622-632 (1994)*

# … as a signature of structuring

➢ *An explanation : the structuring of the liquid*

*Liquid seems to follow the coordinance of the solid phases*



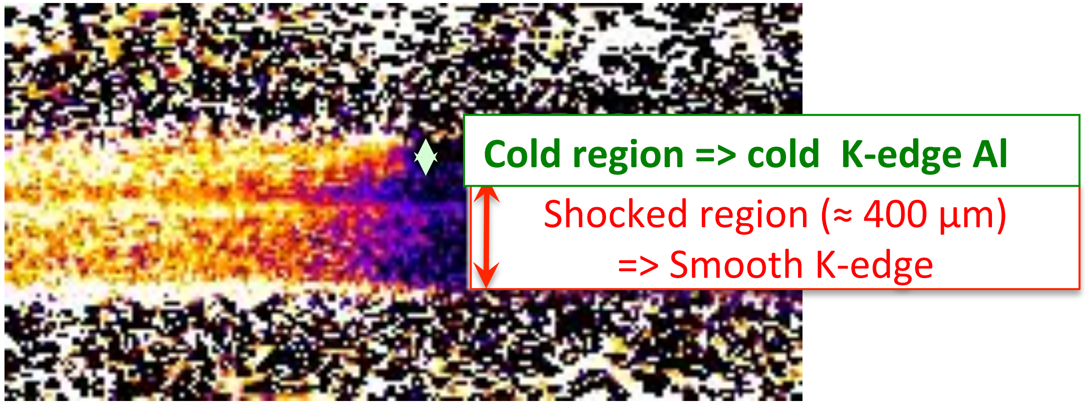


- *Pair correlation function shows a highly structured liquid for higher densities*

- *Coordination increases with density. Up to… highly structured liquid*

# Typical image and methodology

transmitted/reference spectrum



**Cold region => cold K-edge Al**

Shocked region (≈ 400 µm)
=> Smooth K-edge

Spatial resolution 35 µm

Spectral resolution 2.5 eV FWMH

$T_{tot} = I_T/I_{ref} = T_{Al}\, T_{CH}\, T_{Diam}$ ⇒ $T_{Al}$ is extracted ⇒ $T_{Al} = \exp(-\mu d\rho)$

measured

Tabulated

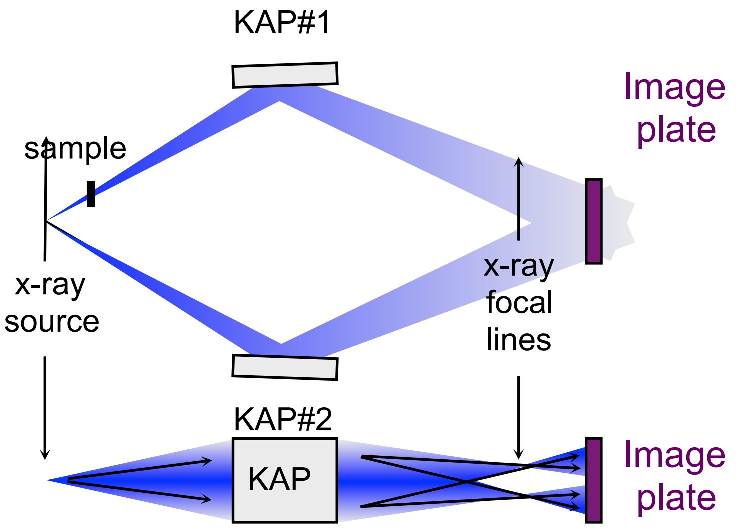
Absorption

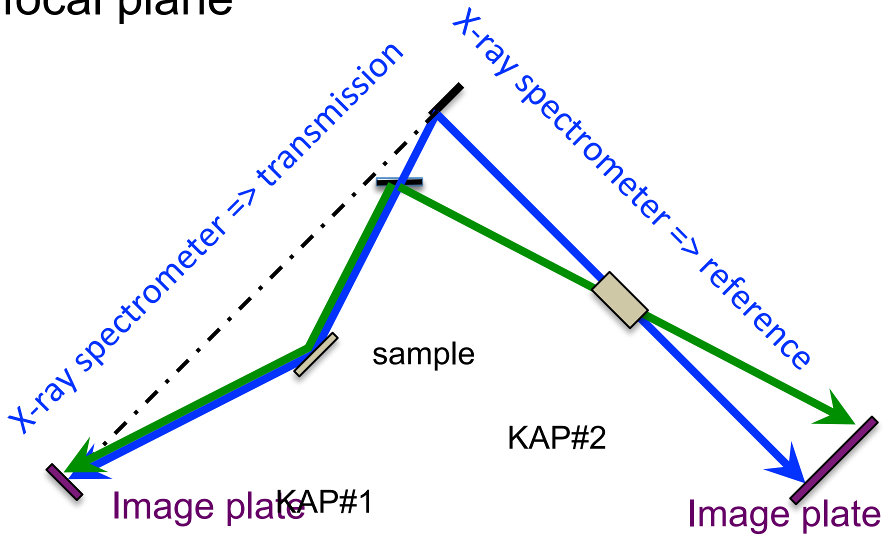
$\mu\,(E) = -1/d\rho \log(T_{Al})$

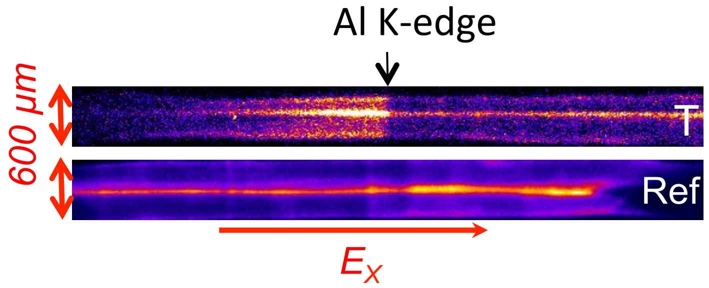
# Method for X-ray Absorption Spectroscopy

- Two X-ray spectrometers (KAP conical crystals)
  - Spectral & spatial resolution
  - Free of shot-to-shot X-ray fluctuations
  - Monitoring & correction of the emission from the ablator

Indeed we had spatial resolution by using a point projection geometry, by putting the Image plate away from the focal plane







The ratio of these images leads to the absorption spectra free of shot-to-shot fluctuations

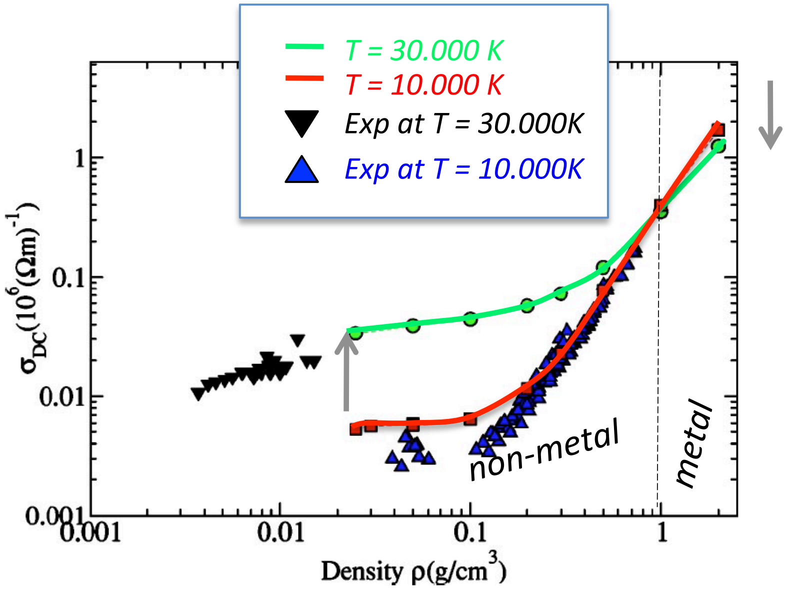
*A. Lévy, RSI (2010); F. Dorchies, APL (2008)*

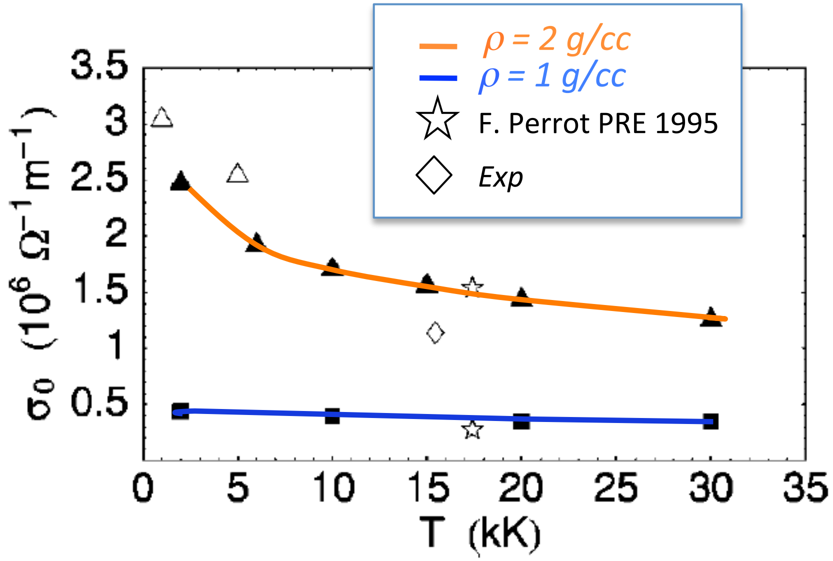
## What does this re-localization correspond to?

It corresponds to the onset of the metal/nonmetal transition

This is consistent with previous conductivity calculations

(Dejarlais et al. PRE (2002) & Mazevet et al. PRE (2005) )





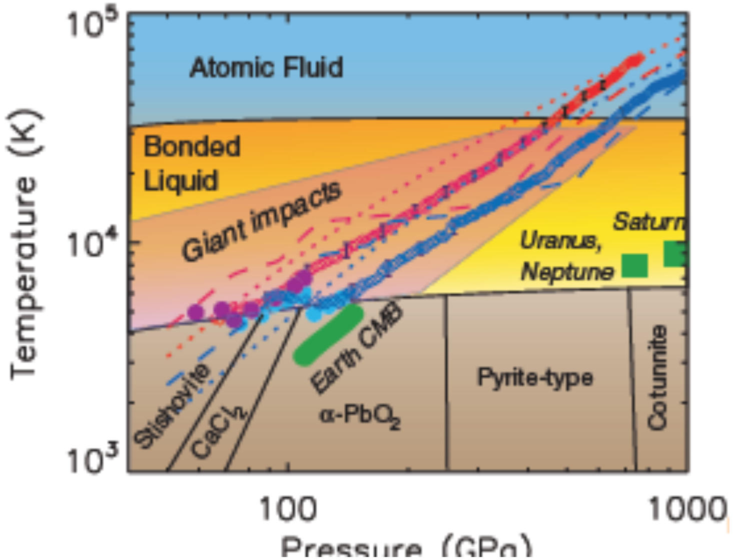
At ≈ 1 g/cc, the *ab initio* conductivity behavior switches from metal to non-metal.

For temperatures in the range up to 30.000 K

At ρ < 1 gr/cc conductivity increases with T

At ρ > 1 gr/cc conductivity decreases with T

# One scenario for $SiO_2$ (Hicks et al. PRL 2006)



Specific heat capacity deduced by the slope of Hugoniot T

$C_V$ increases above melting

$C_V$ shows a broad peak (yellow region)

↕

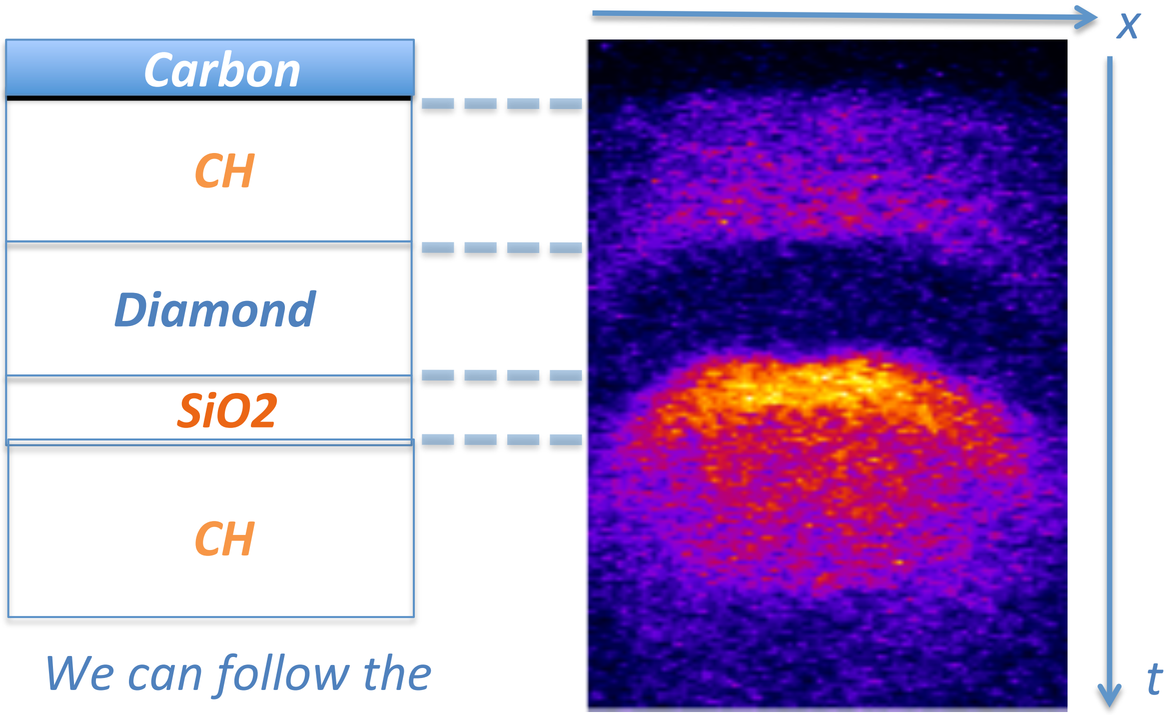
Energy required to break all chemical bonds

For higher T (blue region) : bond breaking is essentially complete

Yellow region: « bonded liquid »= liquid containing a mixture of variously sized polymerized structural units

Blue region: « atomic fluid » = no anymore chemical bonds

# Determination of the probed conditions



*We can follow the emission of the 4 layers*

*SOP image*

***shock timing***
***shock velocity***

**Hydrodynamic simulations**

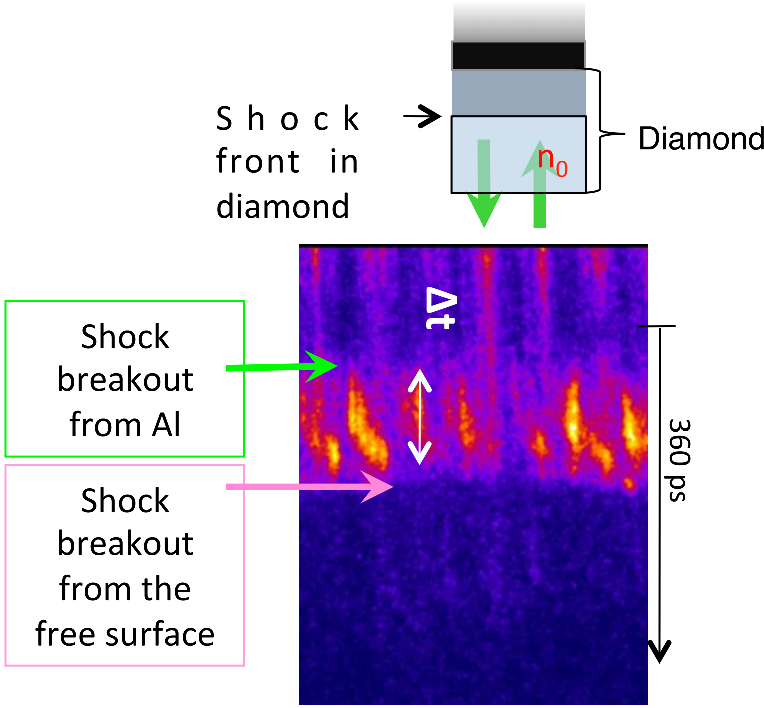
→

**Equations of state**

***Pressure***
***Density***
***Temperature***

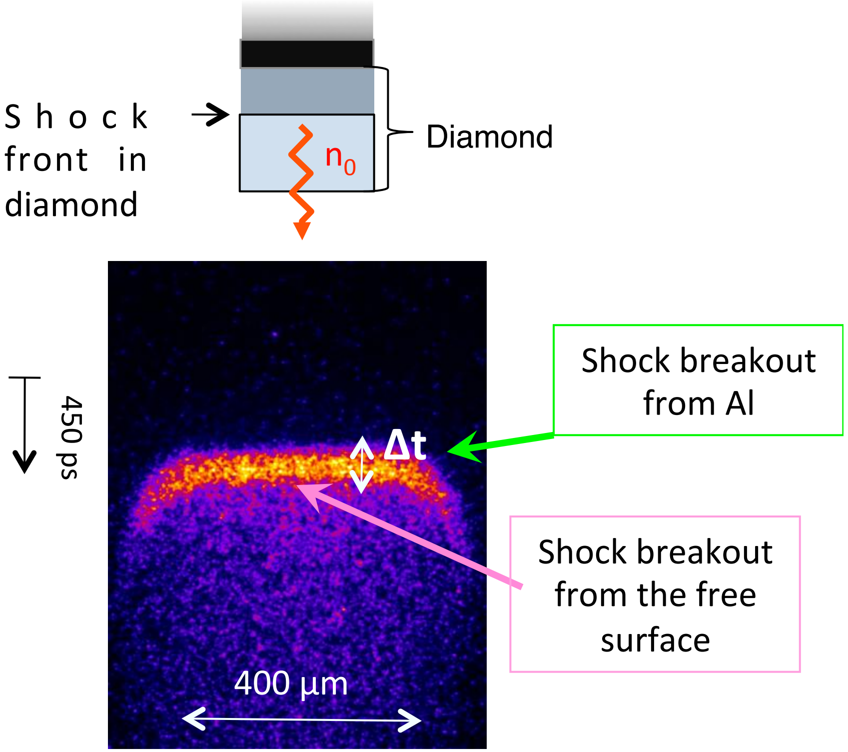
# Al plasma parameters deduced from rear side diagnostics

## VISAR diagnostic



## Emissivity diagnostic

**Very flat shock**



Fringe shift (Doppler effect) → **Shock velocity in Diamond $U_s$ (t)**

Δt → **Mean shock velocity in Diamond $U_s$**

→ **MULTI code** → **Al conditions are known**

*A. Benuzzi-Mounaix, submitted for publication*

# Generalities

X-ray scattering cross section → 3 contributions:

- Free e-
- Weak bound e-
- Thightly bound e-

Inelastic features broadened by thermal motions

Elastic feature (Rayleigh peak)

Two regimes

Scattering parameter $\alpha = \lambda_p/\lambda_s$

$$\lambda_p = \lambda_0/(2\sin(\theta/2))$$

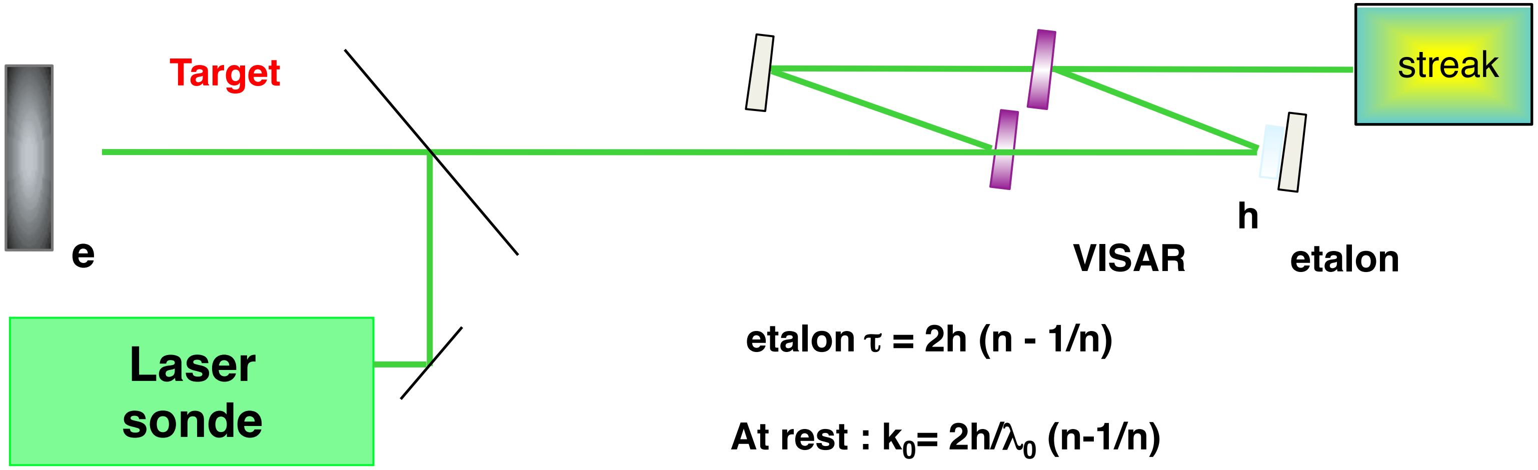
$\lambda_s$ is screening length

$\lambda_p$ is probe wavelength

If $\lambda_p$ is < $\lambda_s$ i.e. $\alpha < 1$ Non Collective regime Scattering on electrons ⇨ $T_e$

If $\lambda_p$ is > $\lambda_s$ i.e. $\alpha > 1$ Collective regime Also scattering on electronic wave ⇨ $n_e$

# Principle of the VISAR



**etalon $\tau = 2h\,(n - 1/n)$**

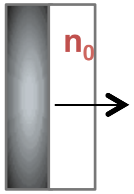
**At rest : $k_0 = 2h/\lambda_0\,(n-1/n)$**

**At a time t : $k = 2h/\lambda\,(n-1/n)$**

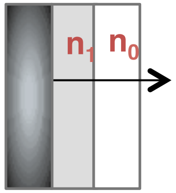
**Order of interference variation : $k-k_0=F$**

⇓

**$(\lambda_0-\lambda)/\lambda_0 = 2V/c$** ⇒ **$V = \lambda_0\,F / 2\tau$**



$$D = \frac{\lambda_0}{2\tau n_0}F$$



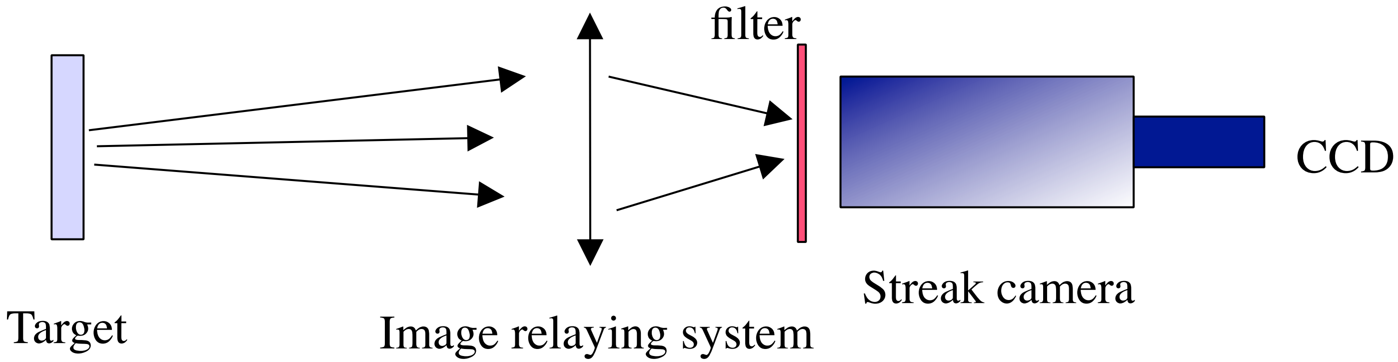
$$U = \frac{\lambda_0}{2\tau g}F$$

$$g = \left(n_2 - \frac{\rho_2}{\rho_2-\rho_1}(n_2-n_1)\right)$$

# T measurement

Temperature $T_b$ is is corrected to a grey body with the absorption coefficient
a = 1 – R, ( R measured at 532 nm).

$a\ (\lambda) = a\ (\lambda 0)\ (\lambda 0/\lambda)^{1/2}$



$$N = \frac{S\Omega\Delta t}{k}\int_{\Delta\lambda} B(\lambda,T_b)T(\lambda)r(\lambda)d\lambda$$

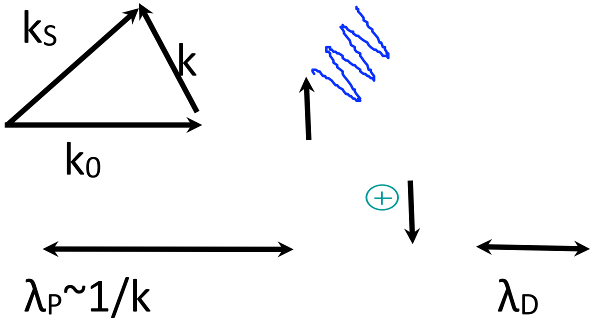
$\Omega$ = Solid angle
N=Counts on CCD
B = blackbody radiance
T= spectral reponse of the optycal system
r=spectral reponse of the photocatode
K(J/counts) = energy response (streak+CCD)
$\Delta\tau$ = pixell exposure time

We measure r,K and T by using a spectral Lamp (Optics Lab OL5500)

We know $N=f(T_b)$

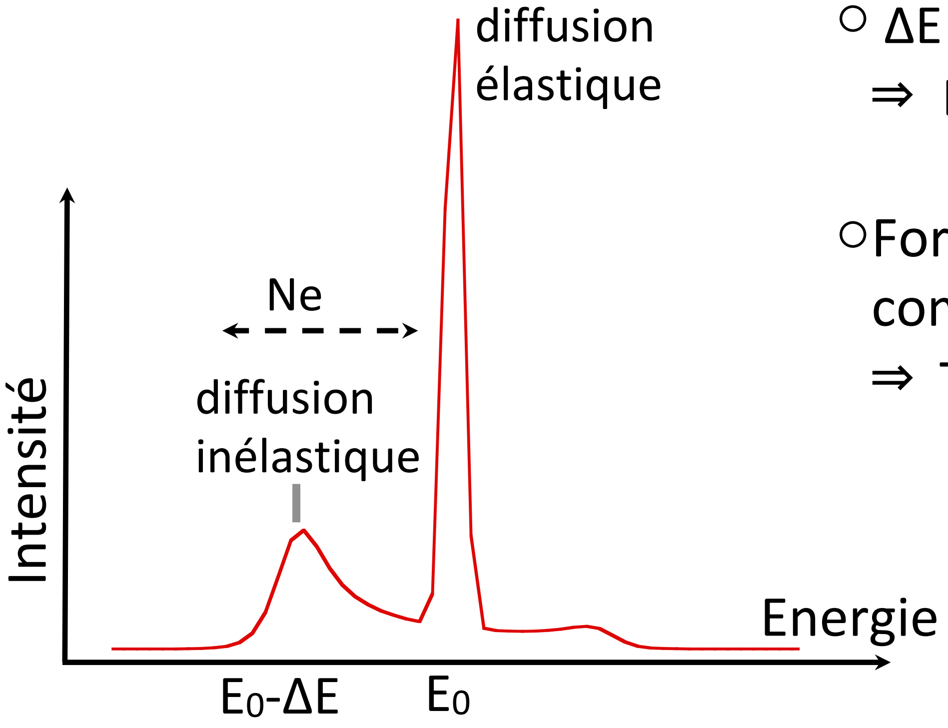
# Principe de la mesure

## Régime collectif: $\alpha=1/k\lambda_D\geq1$



Diffusion a lieu sur les ondes du plasma (plasmon)

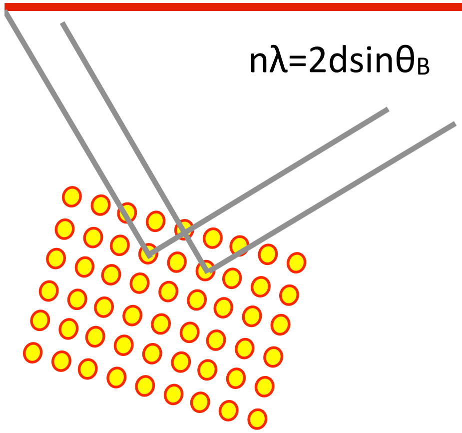
*spectre diffusé est représentatif des propriétés collectives du plasma*



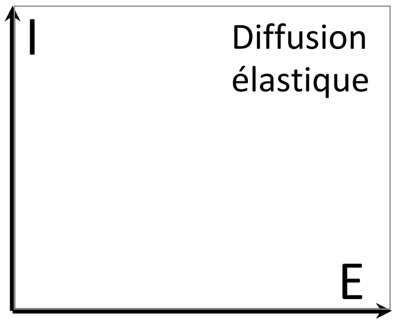
○ ΔE de la composante inélastique
⇒ Densité électronique

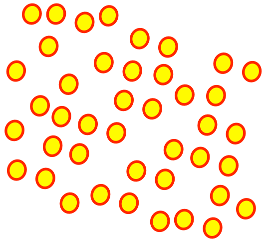
○ Forme et intensité de la composante inélastique
⇒ Température électronique

# Effets des corrélations

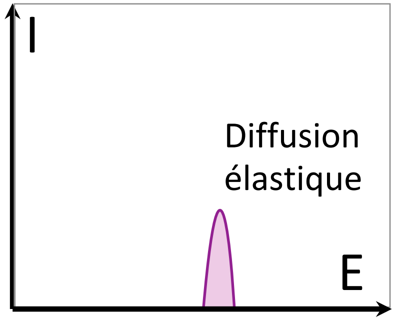


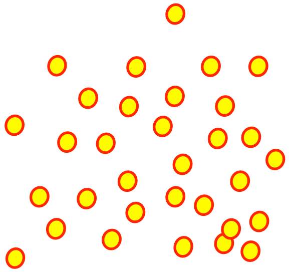
- le solide a une structure ionique bien définie (réseaux) $\Gamma_{ii}\to\infty$
  - *diffusion est déterminée par la loi de Bragg*
  - *il n'y a pas de diffusion en dehors de l'angle de Bragg*



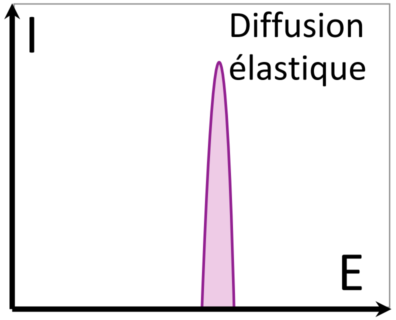


- un ordre à longue portée apparaît dans le plasma $\Gamma_{ii}>1$
  - *il y a diffusion en dehors de l'angle de Bragg mais moins intense*



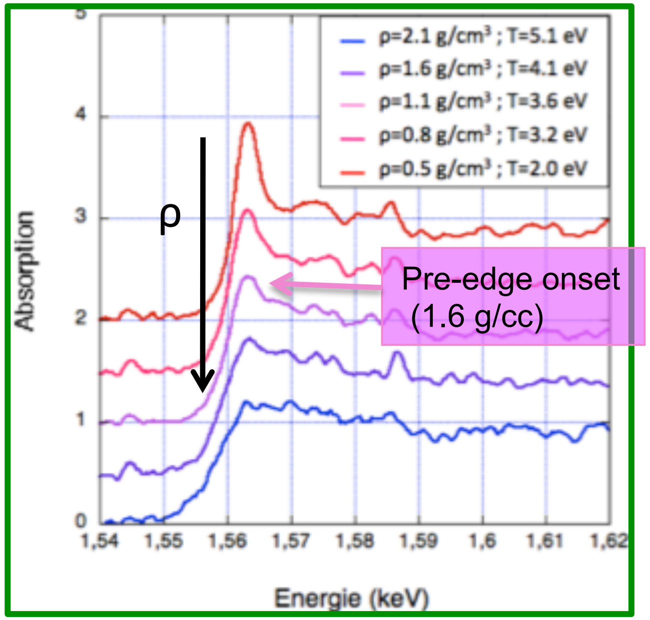


- plasma non corrélé n'a pas de structure définie $\Gamma_{ii}<1$
  - *diffusion est possible aussi en dehors de l'angle de Bragg*

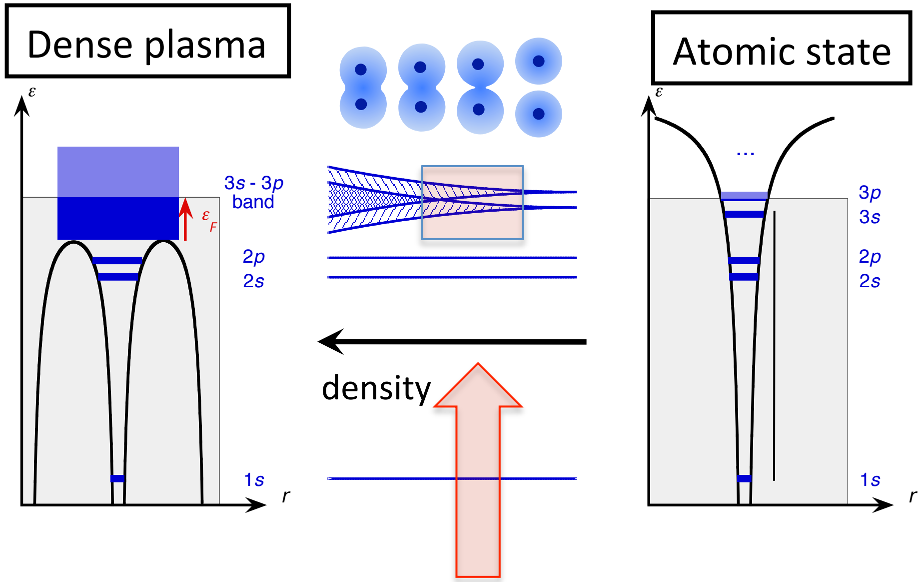


# Study of electronic structure changes in a large WDM domain using XANES

Low compression regime



Pre-edge onset (1.6 g/cc)



Dense plasma

Atomic state

density

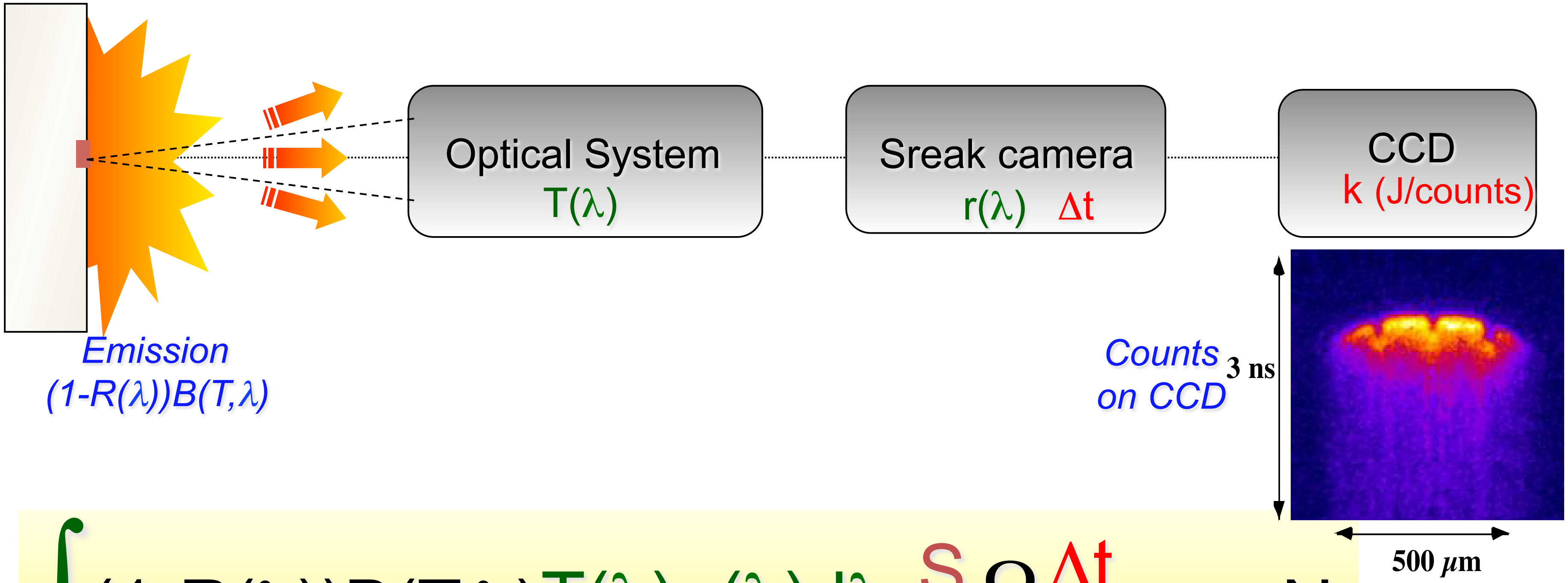
The pre-edge corresponds to the re-localization of the 3p state preceding the formation of the bound atomic states

It corresponds to the onset of the metal/nonmetal transition

Interpretation thanks to ab initio calculations

A. Levy et al PRL (2012)

# Self Emission



Optical System
$T(\lambda)$

Sreak camera
$r(\lambda)$ $\Delta t$

CCD
k (J/counts)

Emission
$(1-R(\lambda))B(T,\lambda)$

Counts on CCD

$$\int (1-R(\lambda))B(T,\lambda)T(\lambda)\ r(\lambda)d\lambda\ \frac{S\ \Omega\ \Delta t}{k} = N$$

*Full Calibration of the diagnostic leads to the shock temperature*

# Internal structure

Internal structure is govern by an equilibrium between pressure and gravity (hydrostatic) ( velocities due to matter movements are negligible)

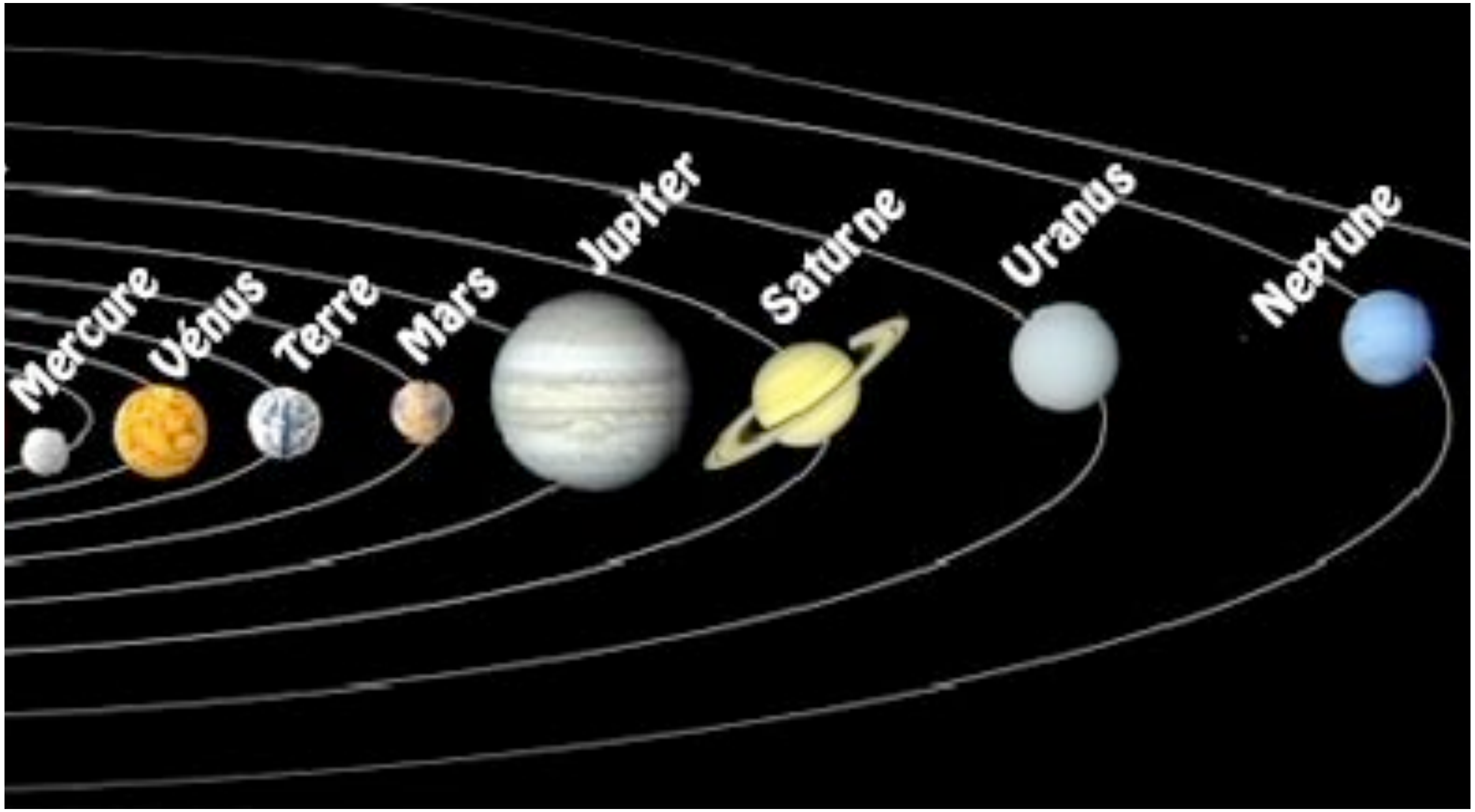
$$\nabla P = \rho \nabla (V + Q)$$

P is pressure and V & Q gravitational & centrifugal potentials
For giant planets Q ≈ 0.1 V. Rotation of Jupiter is 10 h (very high)

$$V(\vec{r}) = G \int \frac{\rho(\vec{r})}{\left|\vec{r} - \vec{r}'\right|} d^3\vec{r}$$

$$Q(\vec{r}) = \frac{1}{2} \omega^2 r^2 \sin^2 \theta$$

r is the radius with origin at the centre of the planet, $\theta$ the angle with respect to the rotation axis, & $\omega$ the rotation frequency at point r. if Q=0, we have the simple equation dP/dr=-$\rho$
Another equation is needed to get the temperature as a function of pressure


Mercure
Vénus
Terre
Mars
Jupiter
Saturne
Uranus
Neptune