



DE LA RECHERCHE À L'INDUSTRIE

Energy Transport Dynamics in Warm Dense Copper using femtosecond time-resolved XANES spectroscopy

Ludovic LECHERBOURG

3rd workshop on Studies of Dynamically Compressed Matter with X-rays | 2021 January 14-15



Ludovic Lecherbourg
Patrick Renaudin
Vanina Recoules

CEA, DAM, DIF, Arpajon, France



Fabien Dorchies
CELIA, Talence, France



Kim Ta Phuoc
Julien Gautier

LOA, Palaiseau, France

Noémie Jourdain
Adrian Grolleau
Thesis CEA/CELIA

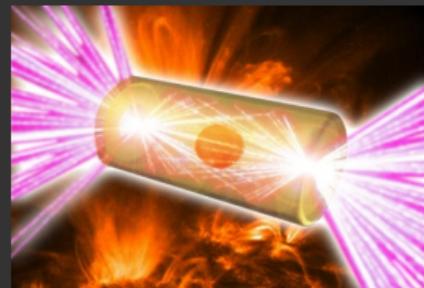
Warm Dense Matter (WDM) is involved in different fields of physics

Astrophysics



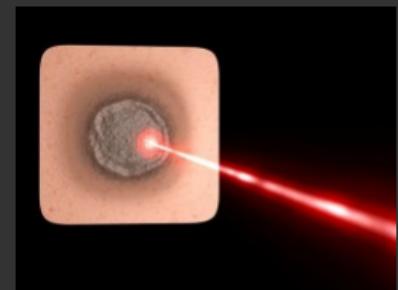
- Planetary science (cores, mantle, etc...)
- Stars (brown dwarves structure, neutron stars surface)

Inertial Confinement Fusion



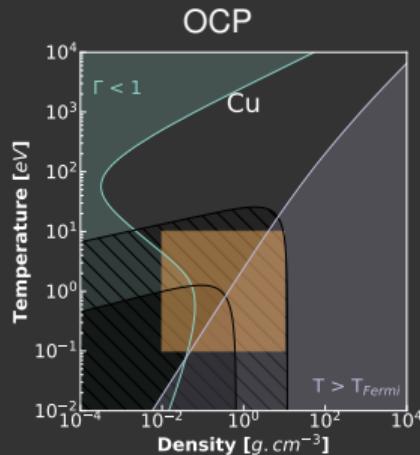
- First moments of the laser-holraum interaction (Au)
- Compression of the ablator (CH)
- Compression of the fuel (D_2)

Laser processing



- Precision laser ablation
- Laser engraving for nano-electronics components

Between plasma physics and solid state physics
Warm : $\lesssim 10 \text{ eV}$ and **Dense** : $\sim \rho_0$

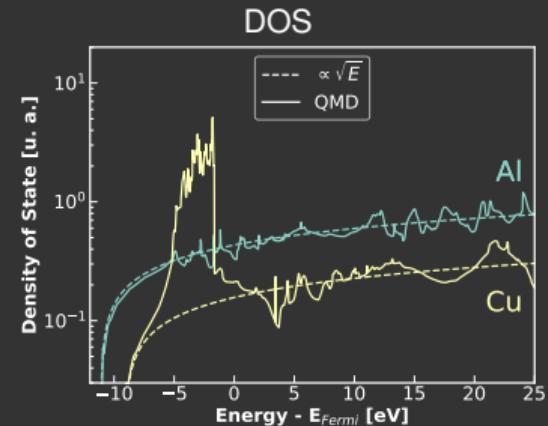


Atoms remain correlated

Coupling parameter
 $\Gamma \equiv \frac{\text{Potential energy}}{\text{Kinetic energy}} \gtrsim 1$

Electrons are degenerated

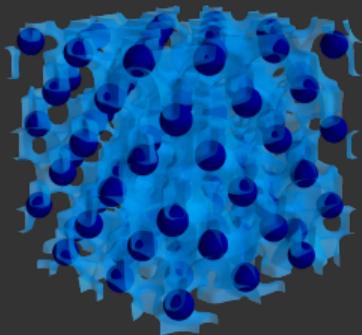
Degeneracy parameter
 $\eta \propto \sqrt{\frac{\text{Fermi energy}}{\text{Kinetic energy}}} \sim 1$



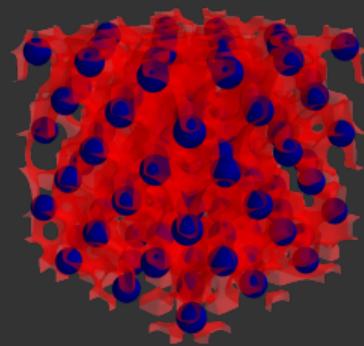
Complex theoretical description of this regime

Laser produced Warm Dense Matter : solid → plasma transition regime

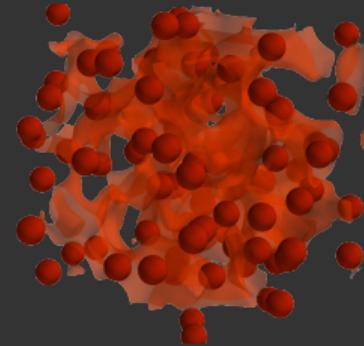
Step 1 : Cold solid

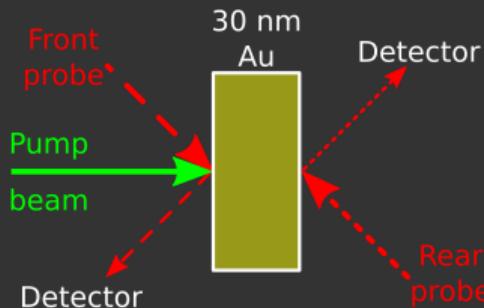
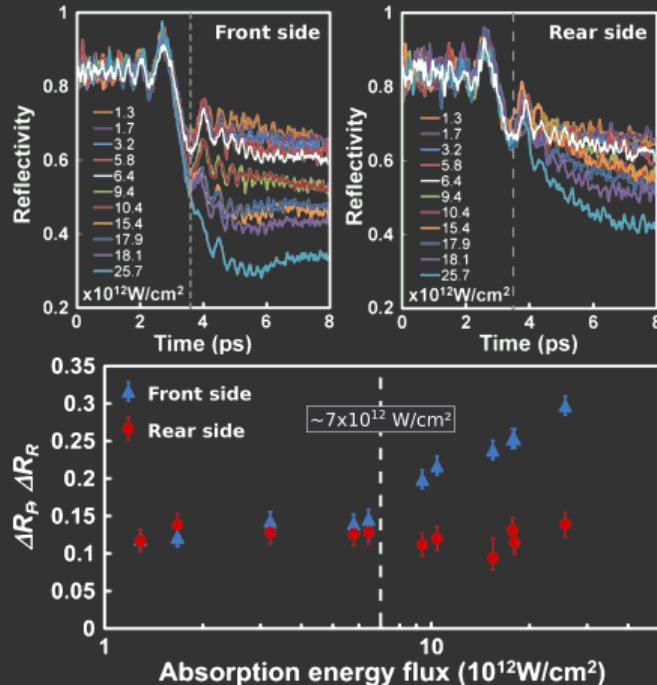
 $T_e = T_i = 300 \text{ K}$ $t = 0 \text{ s}$

Step 2 : HETL WDM

 $T_i < T_{fusion}$
 $T_e \sim \text{few eV}$
 $t \lesssim 1 - 10 \text{ ps}$

Step 3 : ETL WDM

 $T_i \approx T_e \gtrsim 1 \text{ eV}$ $t \gtrsim 10 \text{ ps}$



Chen *et al.* measured in 2012 surface reflectivity changes of gold after laser heating (400 nm, 45 fs FWHM) using chirped optical probe (800 nm).

They highlighted a threshold in the relative reflectivity changes, depending on the absorbed flux.

Results suggest two different regimes for the energy transport.

Figures are from Chen *et al.*, Phys. Rev. Letters **108**, 165001 (2012).

$I < \text{threshold}$

Pump
beam

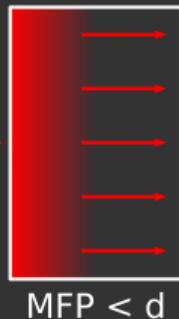


Electrons are excited within the laser skin-depth

- #1
- Energy is transported by ballistics electrons in the sample over their mean free path (MFP).
 - They thermalise through electrons-electrons collisions and heat the sample in few tens of fs.
 - ⇒ homogeneous heating if sample thickness $\lesssim \delta_\lambda + \text{MFP}$

$I > \text{threshold}$

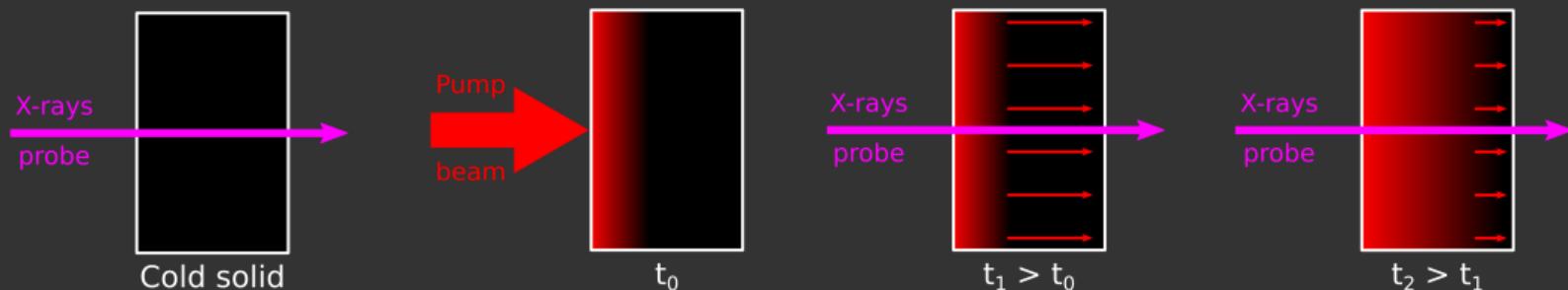
Pump
beam



- #2
- Electrons heat up more and thermalize much faster, in few fs after heating, therefore within the laser skin-depth
Mueller *et al.*, Phys. Rev. B 87, 3 (2013)
Fourment *et al.*, Phys. Rev. B 89, 161110 (2014)
 - Energy reservoir on the front side heats the whole sample by thermal conduction
 - ⇒ inhomogeneous heating lead by thermal conduction

Performing XANES spectroscopy using a betatron source

XANES → $\left\{ \begin{array}{l} \text{■ Direct measurement of } T_e \\ \text{■ In-depth probing} \\ \text{■ Betatron femtosecond time resolution} \end{array} \right\} \Rightarrow \text{Follow in detail the dynamics of } T_e$





Local coupled differential equations describing both electrons and ions temperatures dynamics :

$$C_e \frac{dT_e}{dt} = -G_{ei}(T_e - T_i) + \nabla(\kappa_e \nabla T_e) + S(t)$$

$$C_i \frac{dT_i}{dt} = G_{ei}(T_e - T_i) + \nabla(\kappa_i \nabla T_i)$$

The physics lies in the coefficients :

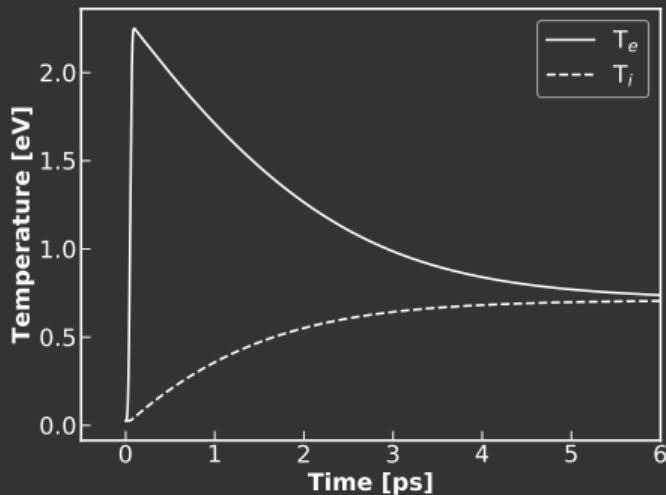
C_e, C_i heat capacities

G_{ei} coupling parameter

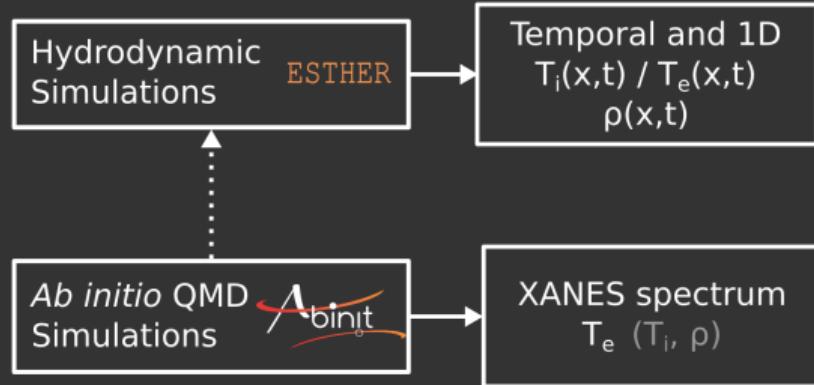
κ_e, κ_i conductances

C_e, G_{ei} and κ_e can be computed from various models, in particular from *ab-initio* QMD simulations.

Lin *et al.*, Phys. Rev. B **77** (2008)

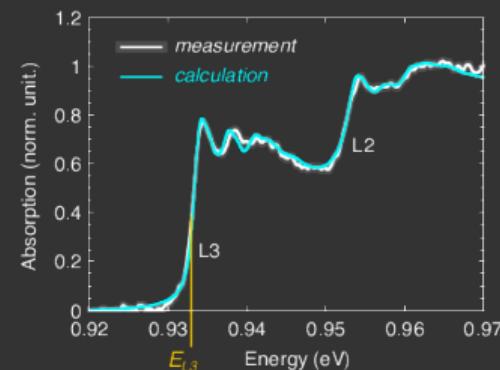
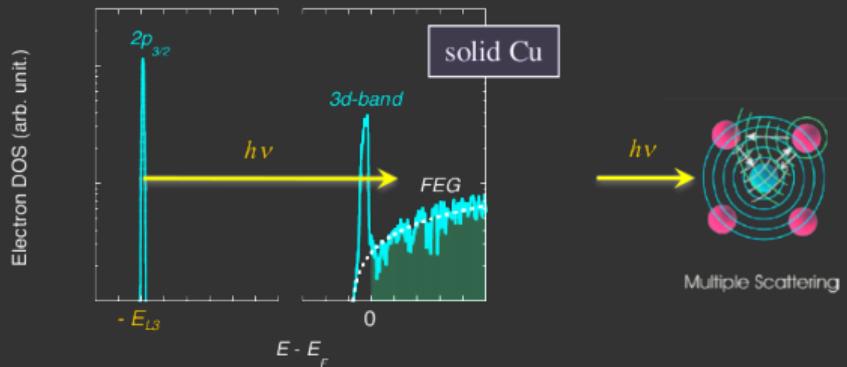


Illustrative 1D Hydrodynamics TTM simulations using ESTHER code.



XANES = X-ray Absorption Near-Edge Spectroscopy

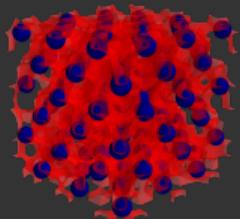
- Probes the (unoccupied) electron DOS from a core level
- Sensitive to the spatial organization of neighboring ions



⇒ **Very proven technique since the 70's and 80's, in condensed and diluted media, but needs to be revisited in the extreme conditions of WDM**

Generation of a thermodynamic configuration

~50 h on 500 CPU



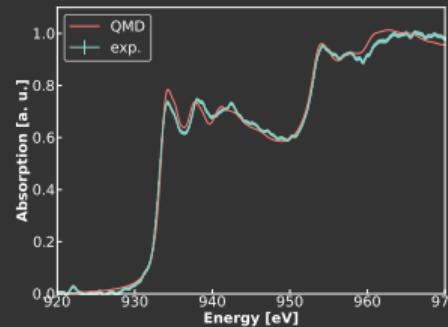
- Density Functional Theory
- Local Density Approximation exchange and correlation
- Projected Augmented Wave scheme
- 108 atoms
- 3x3x3 grid

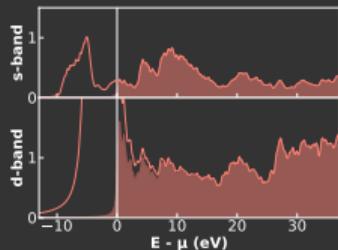
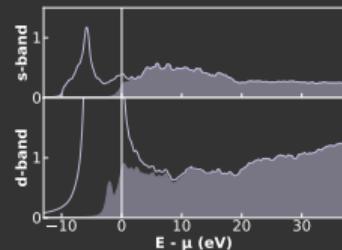
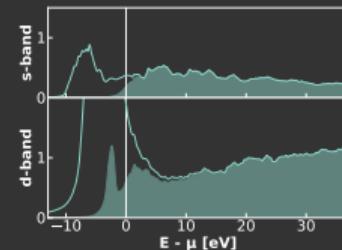
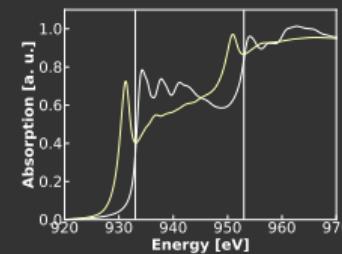
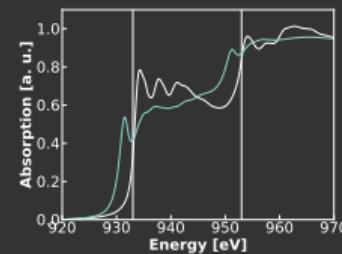
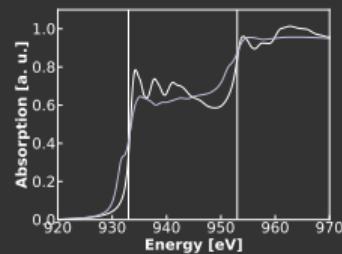
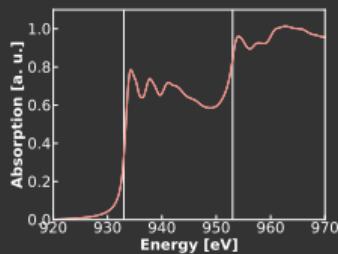
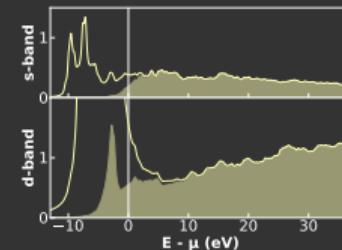
Calculation of photoabsorption cross section

~10 h on 500 CPU

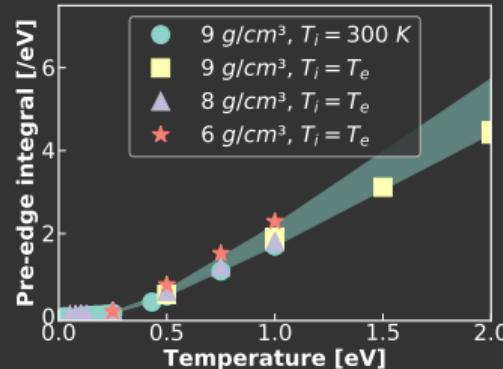
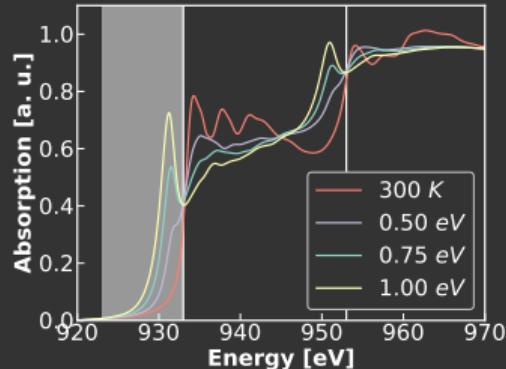
$$\sigma_k(h\nu) = 4\pi^2 h\nu \sum_{i=1}^{n_{\text{orbitales}}} (1 - f(\epsilon_{f,k})) |\langle \phi_{f,k} | \nabla | \phi_{2p,k} \rangle|^2 \delta(\epsilon_{2p} - \epsilon_f - h\nu)$$

Fermi-Dirac occupations	Transition matrix elements	KS eigenvalues
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$T_e = 300 \text{ K}$  $T_e = 0.5 \text{ eV}$  $T_e = 0.75 \text{ eV}$  $T_e = 1.0 \text{ eV}$ 

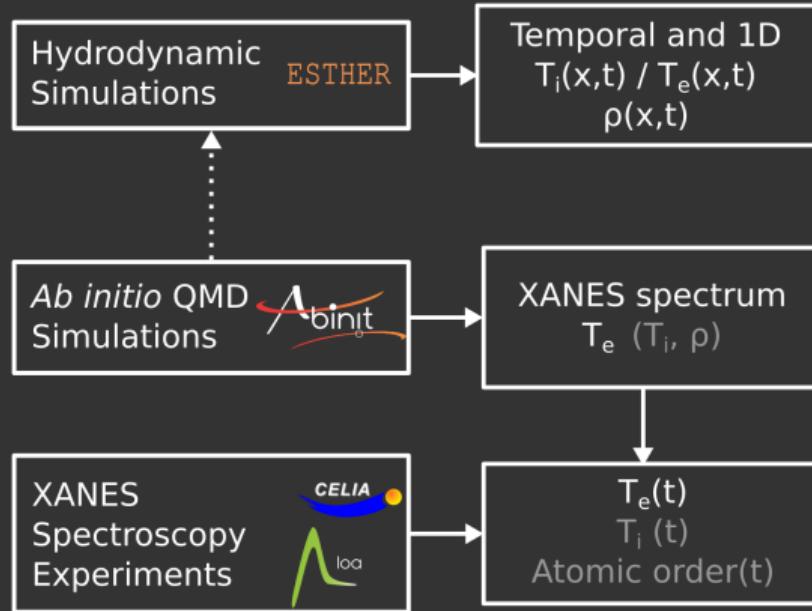
When T_e increases, available levels appear in the d-band, which increases the absorption before the Fermi level.



The electronic temperature T_e can be measured with the integral of the pre-edge.

A function to attribute a T_e to our experimental data have been deduced.

N. Jourdain *et al.*, Phys. Rev. B **101**, 125127 (2020)



Working regime

- Based on **Chen et al.** work

Copper deposition

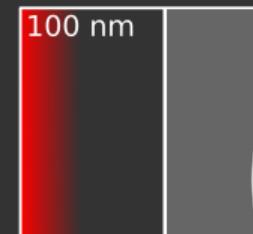
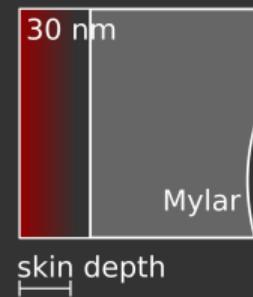
- Realized by evaporation
- Substrat : 1 μm of PET "Mylar"
- Laser skin depth @ 800 nm \sim 13 nm
- 1 eV electrons ballistic range \sim 70 nm

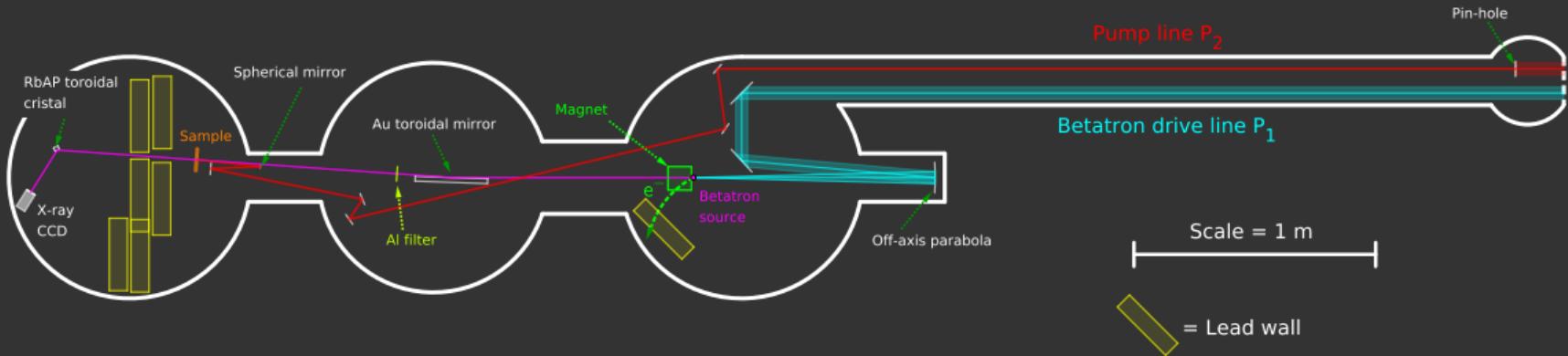
Nguyen-Truong, J. Phys. : Condens. Matter **29** (2017)



Two heating regimes investigated

- 30 nm : quasi homogeneous energy deposition
- 100 nm : inhomogeneous energy deposition





Pump beam (on sample)

- $\phi = [380 \pm 10] \mu\text{m}$ - top-hat
- Incidence $_{\perp} = [2.2 \pm 0.2]^\circ$
- Pulse duration FWHM = 30 fs
- $[1.1 \pm 0.3] \times 10^{14} \text{ W/cm}^2$

Betatron drive beam

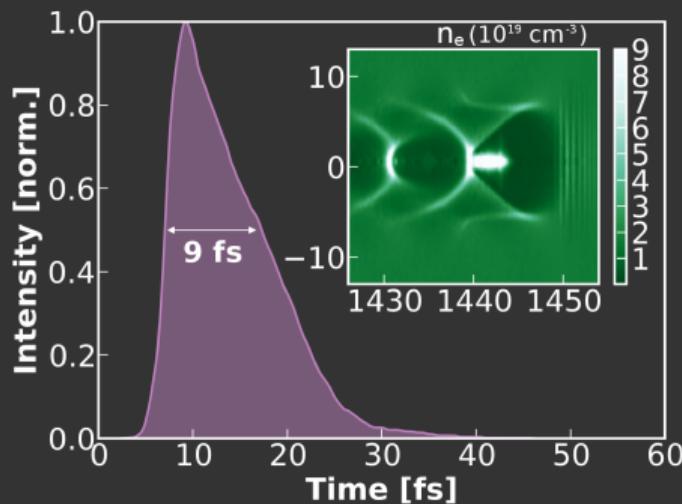
- $\sim 50 \text{ TW}$
- On gas jet $\sim 3 \times 10^{19} \text{ W/cm}^2$
- Gas jet : 99%He / 1%N, ~ 20 bars

Betatron X-ray beam

- Low divergence $\sim 10 \text{ mrad}$
- $\phi \approx 150 \mu\text{m}$ on sample
- Normal incidence on sample
- $\lesssim 6.5 \times 10^4 \text{ ph / eV / shot}$
@ $\sim 930 \text{ eV}$

Betatron pulse duration estimated from PIC simulations

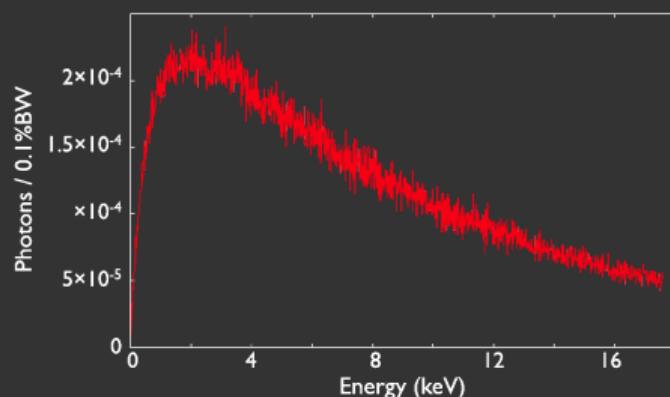
Mahieu *et al.*, Nature Comm. 9(1), 2018



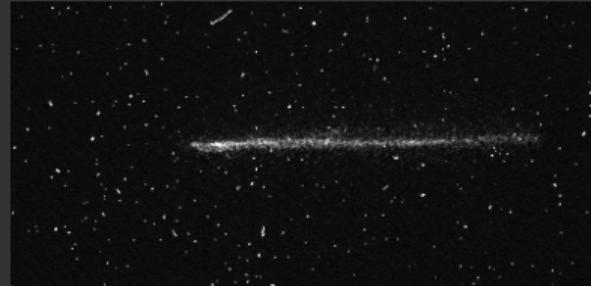
Temporal resolution is then limited by pump-probe angle of 2.2° and X-ray spot size of $\sim 150 \mu\text{m}$ leading to $\sim 20 \text{ fs resolution}$.

Betatron emission spectrum for 10^{19} W/cm^2 800 nm laser.

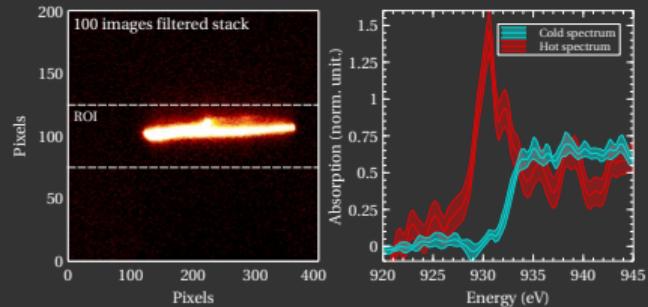
Figure from Corde *et al.*, Rev. Mod. Phys. 85 :1-48, 2013



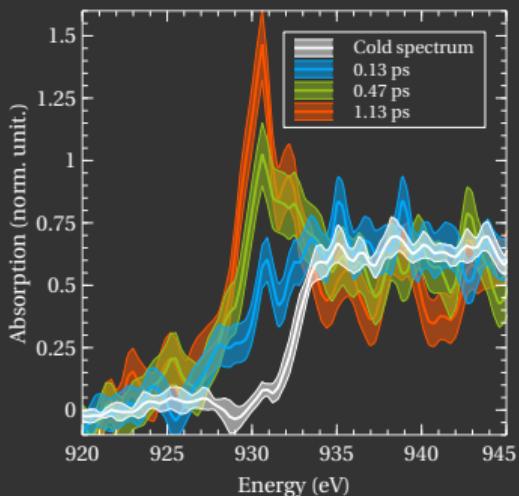
- Accumulation required : betatron spectrum stability over ~ 100 shots/series.
- Hot spots filtration (3-axis filtering).
- A hole is created by the pump beam at each shot : the sample is automatically moved to a clean area between two shots (0.3 Hz).
- Routine to minimize errors during XANES spectra extraction :
 - one reference serie "*R*" without sample ;
 - one **cold** serie "*C*" at ambient temperature ;
 - one **hot** serie "*H*" with the pump beam.



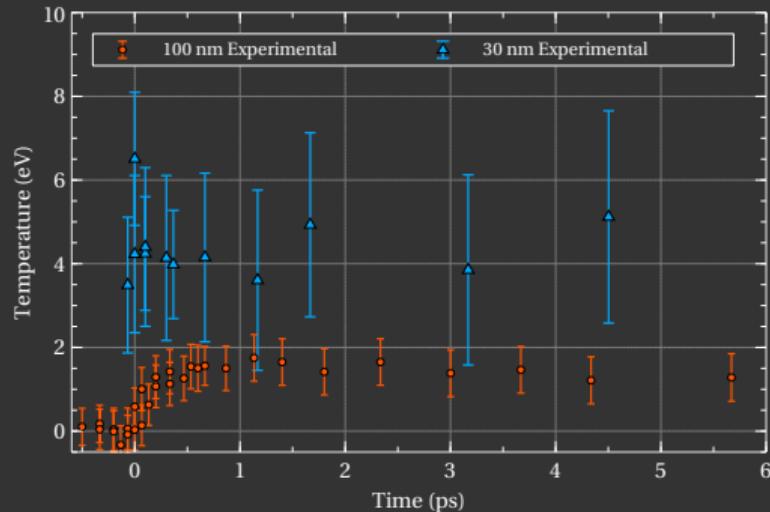
Shot to shot stability over 100 images



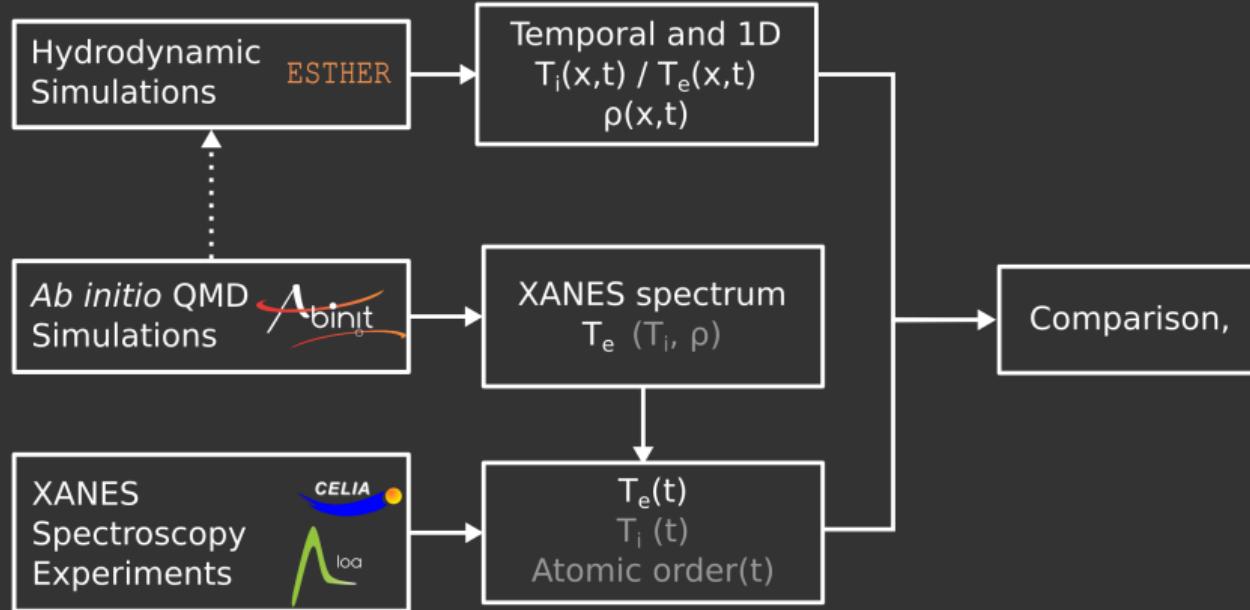
- Different delays probed to infer the dynamics of T_e (~ 25 min / delay).
- Pre-edge absorption increases as sample is heated.
- Uncertainties : photons counting statistics (colored areas).



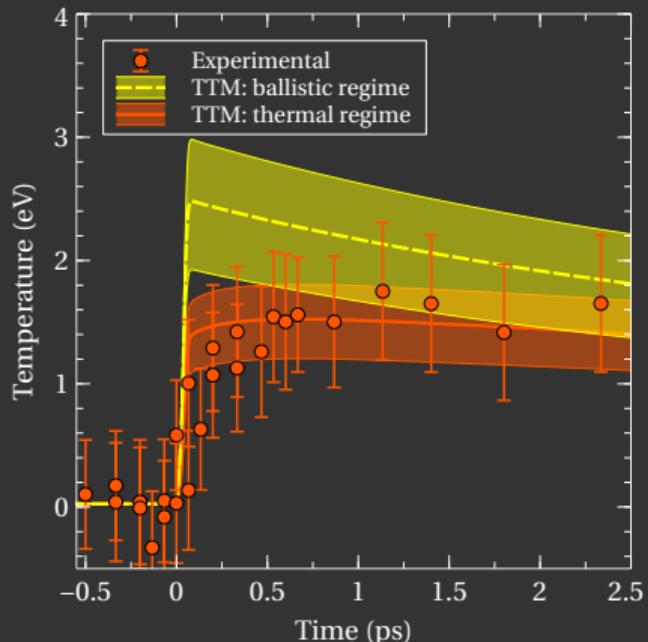
- Pre-edge integral is transformed into electrons temperature using the model from *ab initio* simulations.
- Almost no energy is lost in the substrate for 30 nm samples during the heating, as T_e rises 3 times higher than for the 100 nm samples \Rightarrow energy deposition < 30 nm.



Data presented has required $\sim 10'000$ shots, acquired during 3 days plus 3 weeks for preparation.



- Experimental data show a slow increase to ~ 1.5 eV in ~ 1 ps
- TTM Simulations (ESTHER code) :
 - absorbed flux based on energy and absorption coefficient measurements
 - uncertainties on absorbed energy is represented by colored areas
- Ballistic regime : instantaneous heating, then electron-ions collisions \Rightarrow do not reproduce T_e dynamics during the first ps
- Thermal regime : slower heating, T_e rises to 1.5 eV in 0.7 fs
 - ⌚ Experimental data unveils the conduction phase inside the sample, below the first picosecond
 - ⌚ Errorbars are still too high to differentiate conduction models
- Long time T_e for [100 nm] simulation is consistent with experimental data, in agreement with previous work
Jourdain *et al.* Phys. Rev. B **97**, 075148 (2018)



- Copper (30 nm and 100 nm) samples were heated using femtosecond laser at $\sim 1 \times 10^{14} \text{ W/cm}^2$.
- Almost no energy is lost in the substrate for 30 nm samples during the heating, as T_e rises 3 times higher than for the 100 nm samples \Rightarrow energy deposition $< 30 \text{ nm}$.
- Electrons temperature dynamics is inferred using femtosecond TR-XANES, which highlight 3 thermodynamics steps :
 1. electronic temperature rise quickly in the skin depth during laser pulse ;
 2. the energy reservoir created heat the whole sample by conduction for $\sim 1 \text{ ps}$;
 3. electrons heat the lattice by electron-ion collisions.

Grolleau *et al.* in preparation

- Improve the signal to noise ratio (better hot electrons filtration, higher betatron source flux) \Rightarrow possibility to differentiate conduction models ?
- Other materials (study on molybdenum is on-going)



Thank you for your attention

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Commissariat à l'énergie atomique et aux énergies alternatives

CEA, DAM, DIF | F-91297 ARPAJON

T. +33 (0)1 69 26 40 00

Direction des Applications Militaires

Établissement public à caractère industriel et commercial | RCS Paris B 775 685 019

Fields

Nuclear physics, plasma physics, atomic physics, molecular physics.

Description - Missions

- Design, conduction and analysis of plasma experiments on Laser MégaJoule (LMJ) and partners facilities.
- Collaboration with plasma diagnostics development service and analysis programs developpers.
- Collaboration with simulations codes developpers.
- International collaborations.

Location

26 Rue de la Piquetterie, 91680 Bruyères-le-Châtel, France (30 km south-west from Paris).

Requirements

- PhD in Physics, good knowledge in plasma physics, programming and signal processing.
- Good french and english speaking.

Availability

September, 2021

Contact

berenice.loupias@cea.fr
stephanie.brygoo@cea.fr