Near Edge X-ray Absorption Spectroscopy (XANES) as a diagnostic to study Warm Dense Matter

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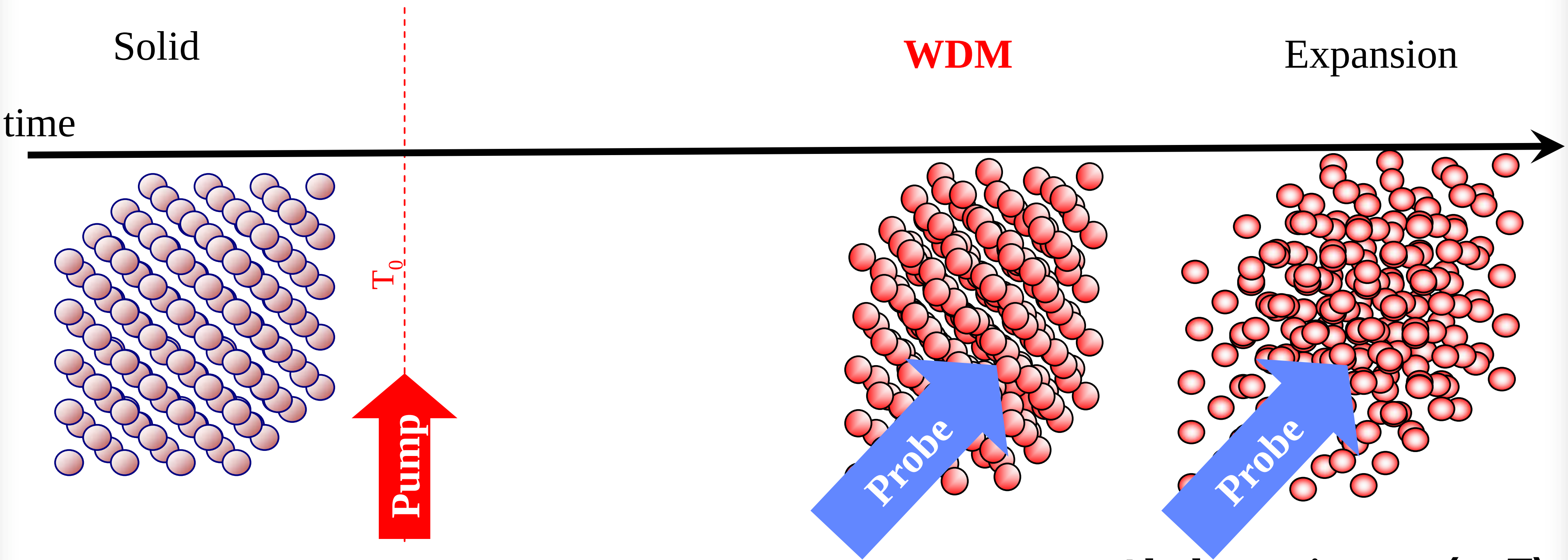
Pump – probe experiments for WDM studies

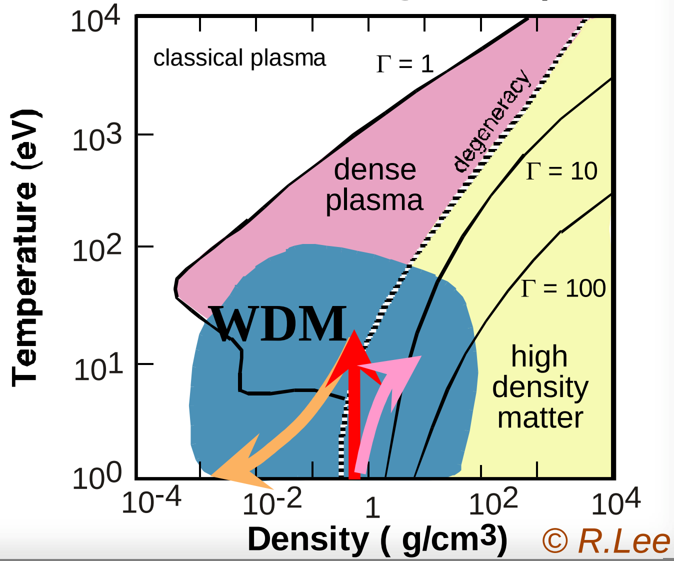


- Between solid and plasma state:
  - strongly correlated ions
  - degenerated electrons $T_e \sim T_F$
- Need experimental data

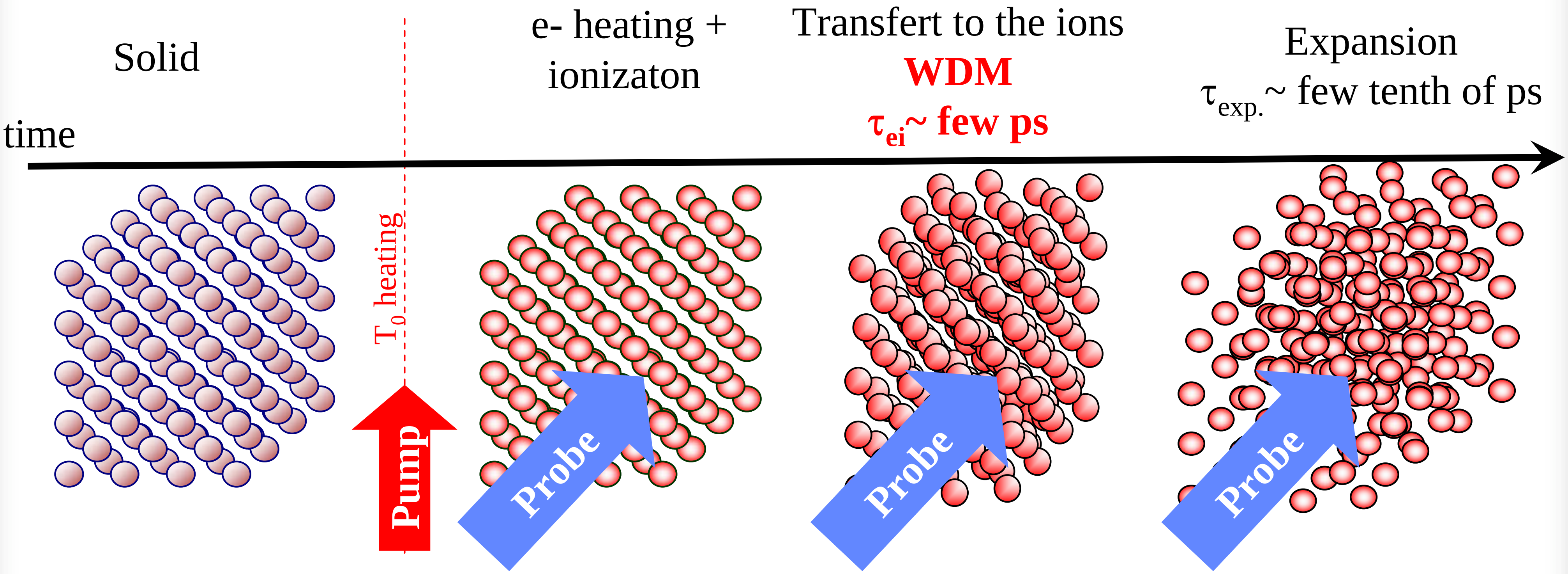
Isochoric heating by laser, by protons or by photons + ***isentropic expansion***

Shocks

Al Phase Diagram (ρ, *T*)



Isochoric heating: Time scale



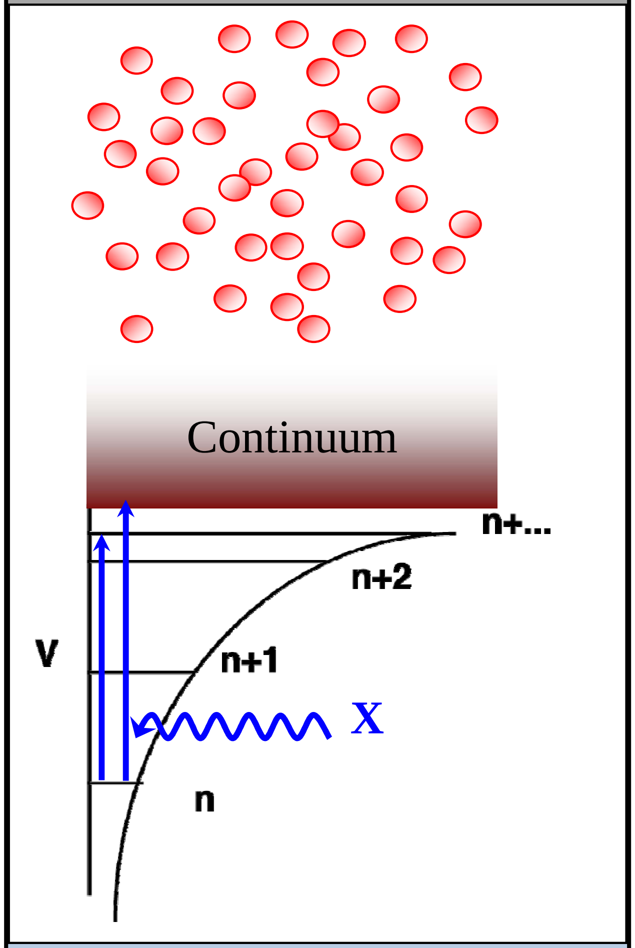
- Homogeaneous heating (T, ρ): $\tau_{heating} < \tau_{exp.}$
- Probe $\tau_{probe} < \tau_{exp}$ ➡ Probe < 10ps)
- Probe the deep local order

➡ Ultrafast pump – probe x-ray absorption spectroscopy experiment to study a disordered state
Access to the ion-ion correlations

X-Ray Absorption Spectroscopy of WDM

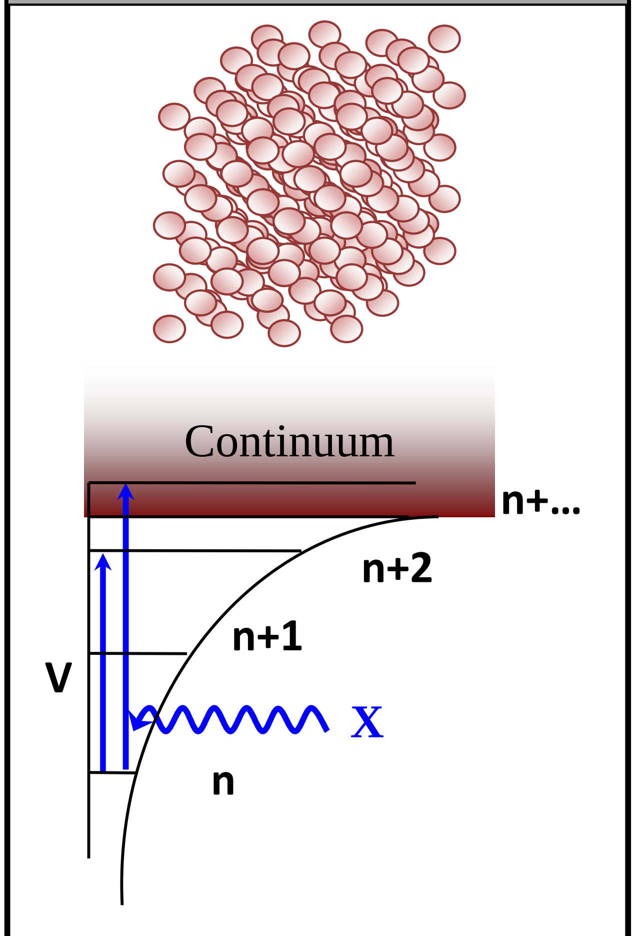
Probe unoccupied density of state

Hot Plasmas



Absorption edge + lines

Warm Dense Matter



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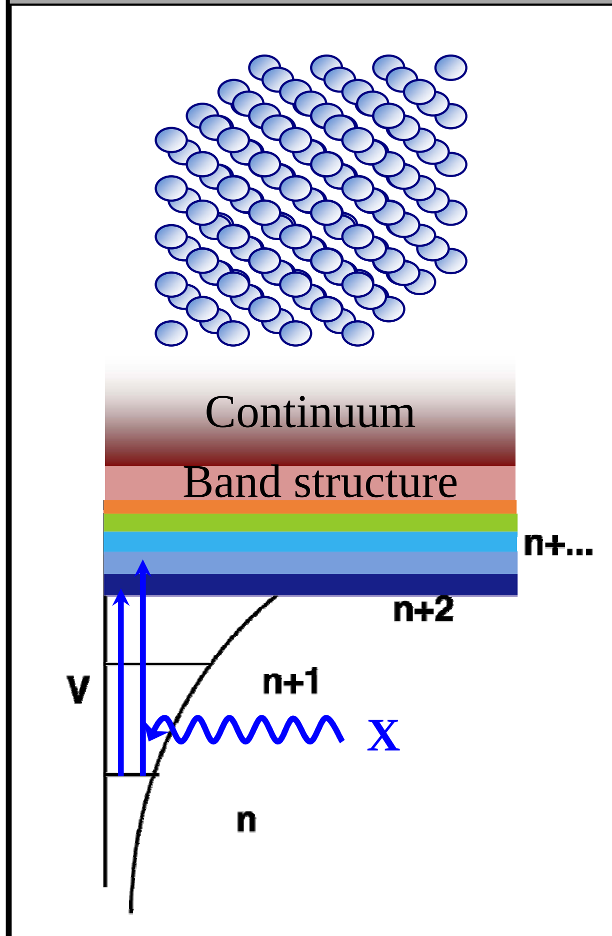
Solid



Absorption edge

Near edge x-ray absorption spectroscopy (1)

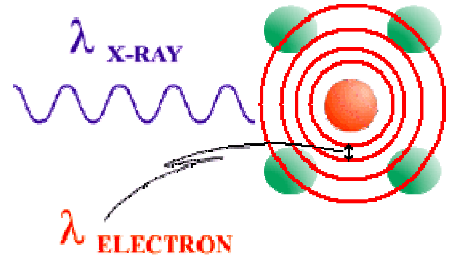
Temperature and density probe

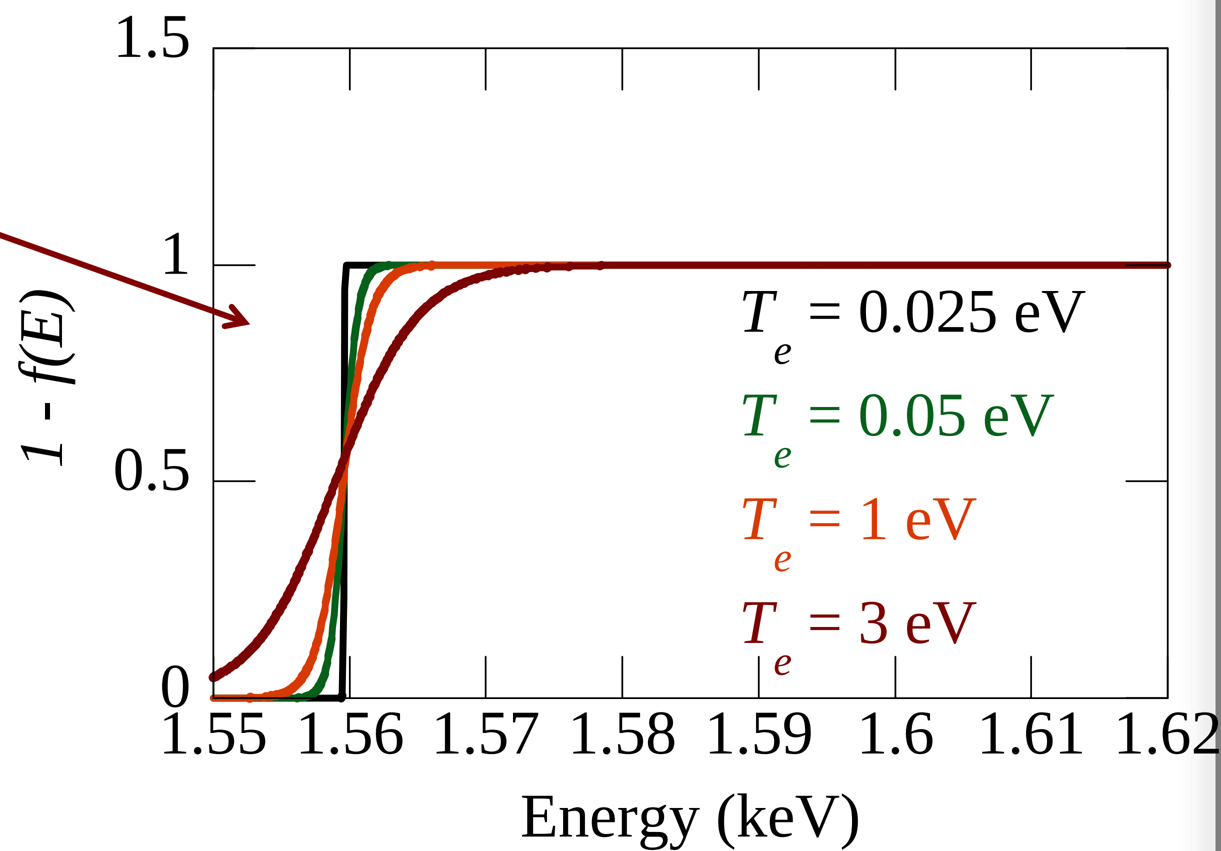
Fermi Golden rule :

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

Fermi Dirac Function :

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





Absorption edge position & slope :

→ T_e for an isochoric heating

→ generally T_e & ρ

Near edge x-ray absorption spectroscopy (2)

Probe the local order

Fermi Golden rule :

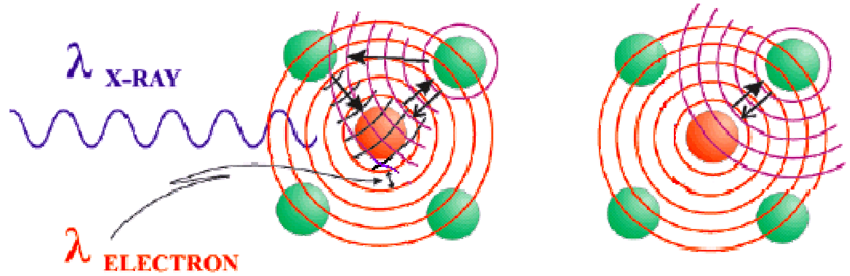
$$\sigma_{if}(\nu) \propto h\nu \left| \langle \phi_f | R | \phi_i \rangle \right|^2 (1 - f(E))$$

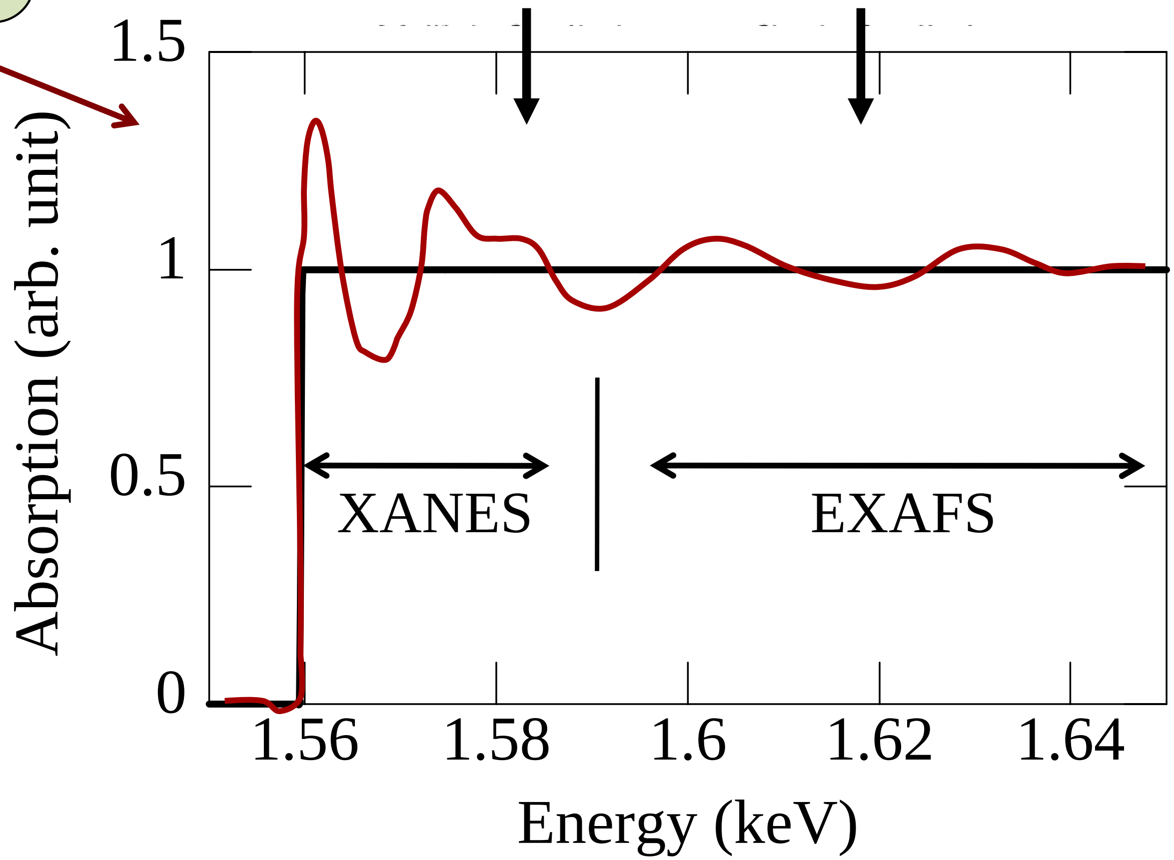
$$\text{with } |\phi_f\rangle = |\phi_{free}\rangle + |\phi_{scattering}\rangle$$

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

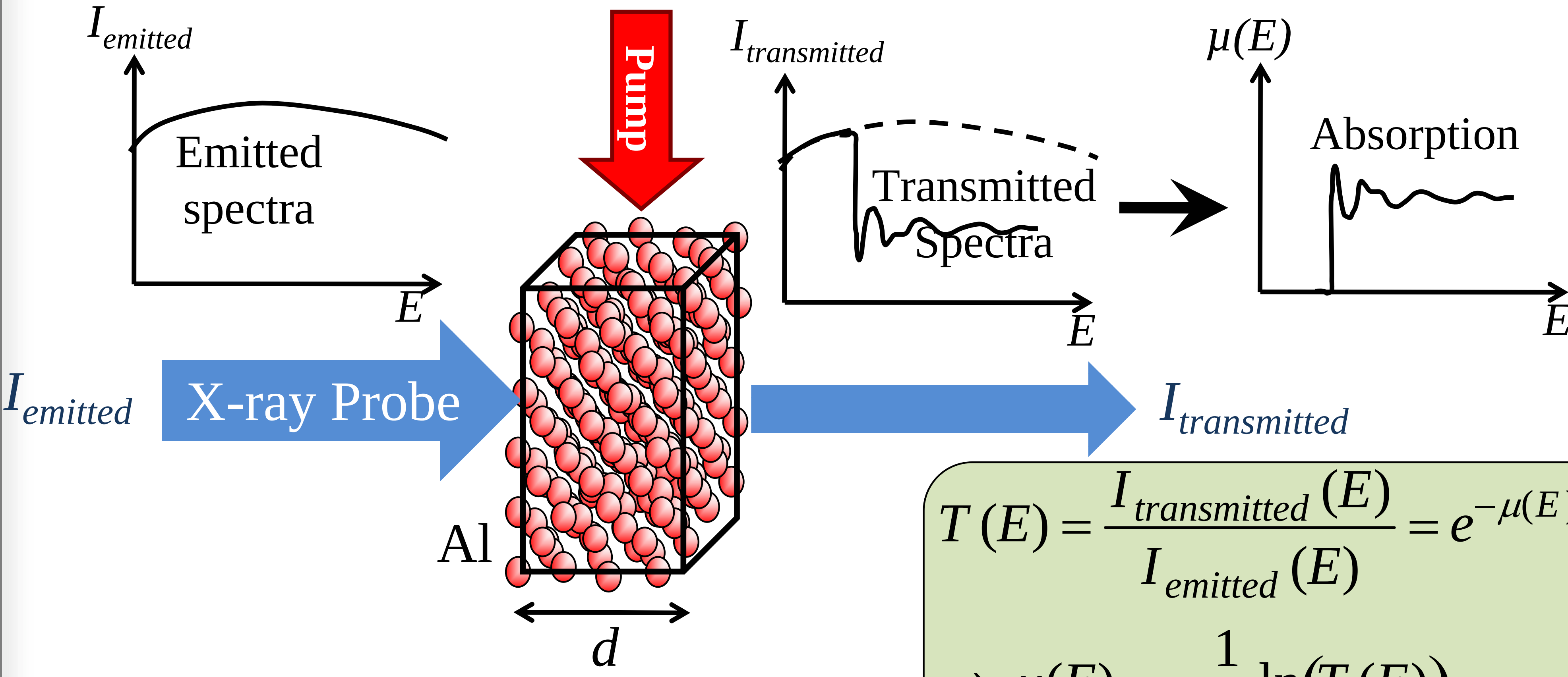
- **EXAFS** Extended X-ray Absorption Fine Structure

→ **local order** (correlation function…)





X-ray absorption experiments for WDM studies

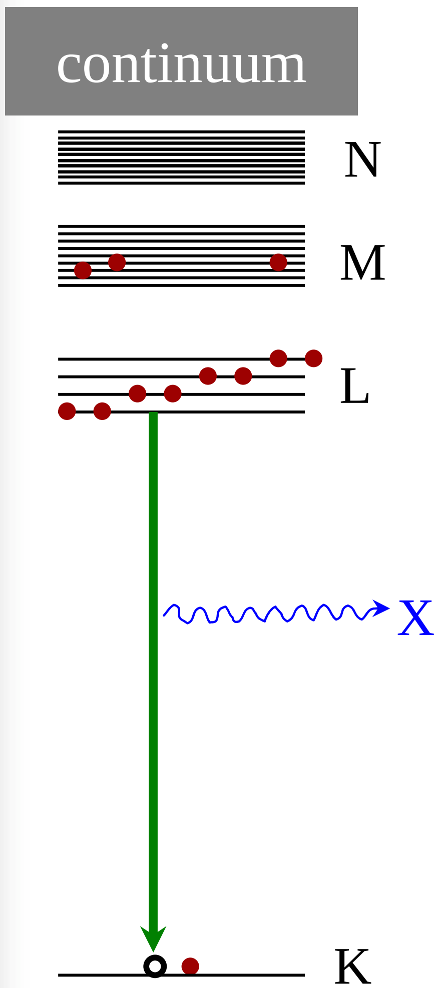


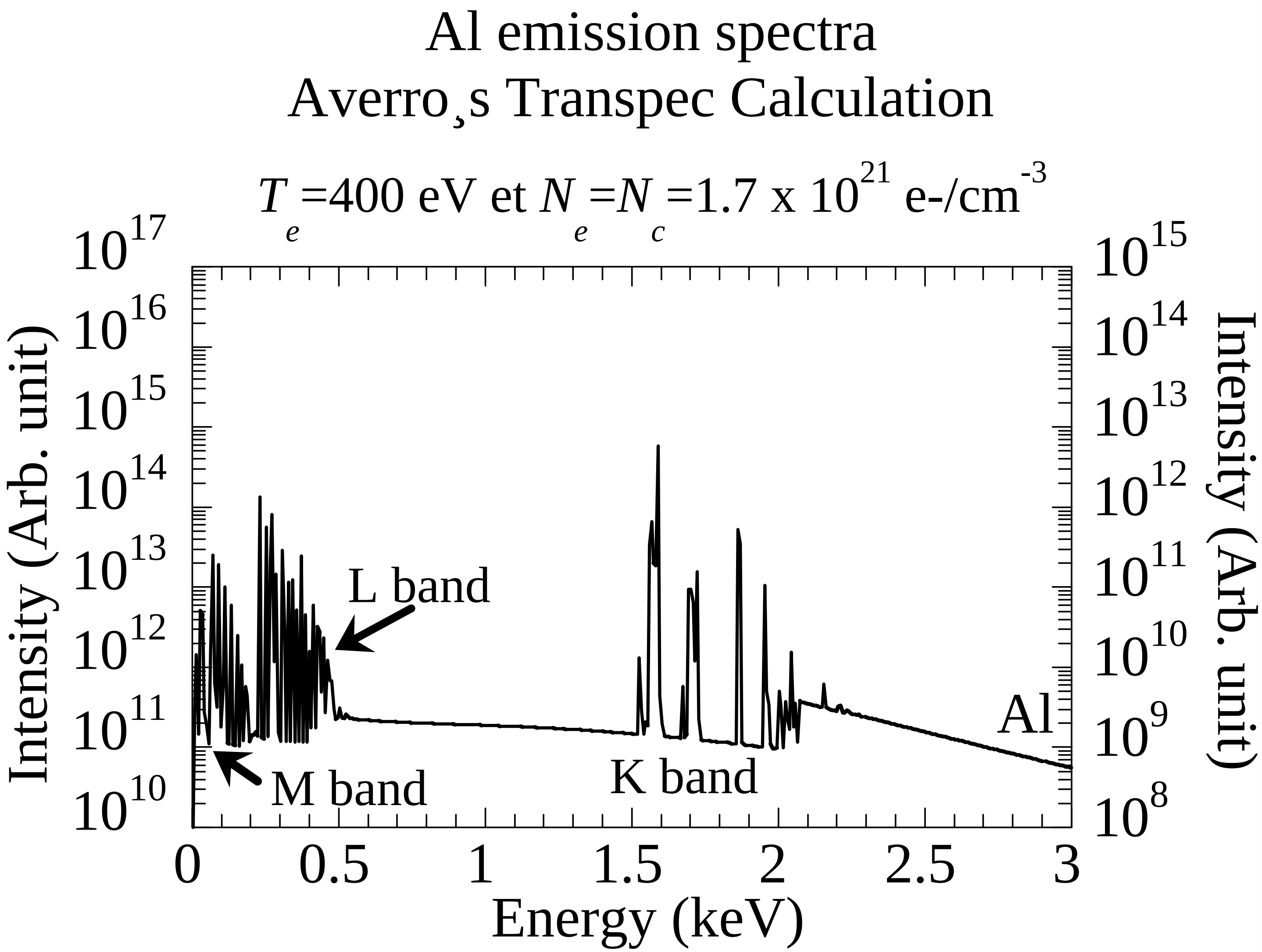
$$T(E) = \frac{I_{transmitted}(E)}{I_{emitted}(E)} = e^{-\mu(E).d}$$

$$\Rightarrow \mu(E) = -\frac{1}{d}\ln\left(T(E)\right)$$

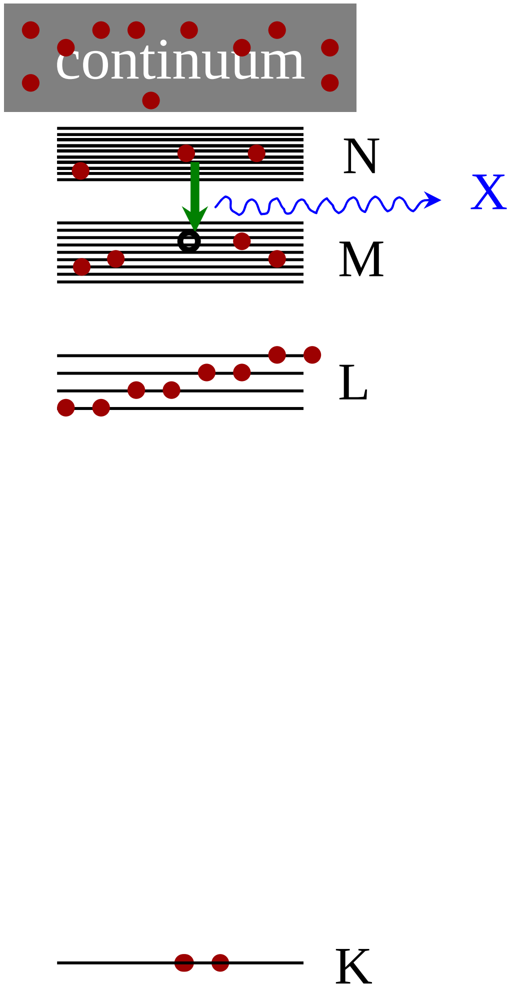
- **Specific x-ray absorption spectrometer**
  Emitted & transmitted spectra measured at the same time
- **Well adapted x-ray source**: broad band, around 1.56 keV (Al K-edge), intense & ultrafast

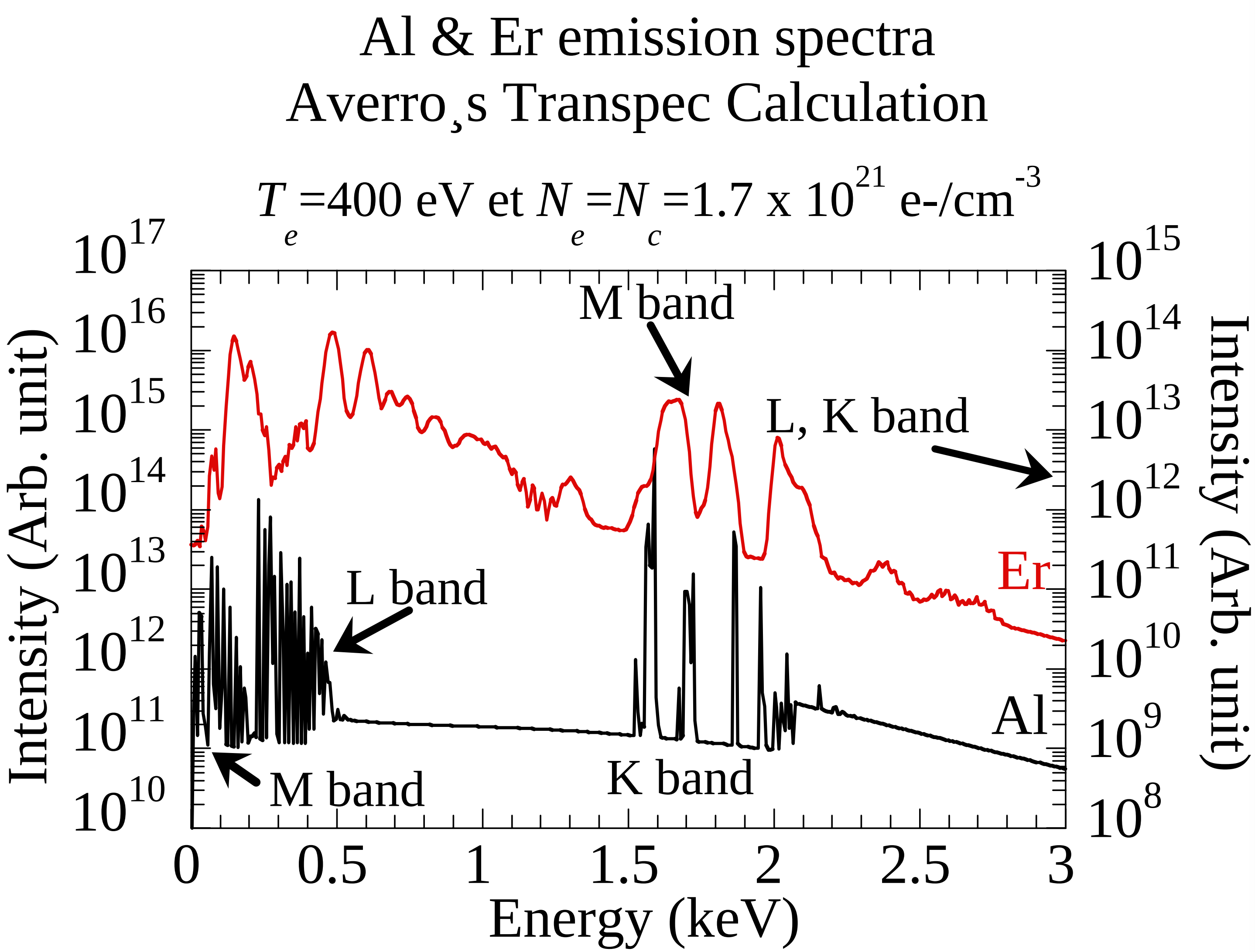
Laser-plasma X-ray sources from high-Z elements





Laser-plasma X-ray sources from high-Z elements

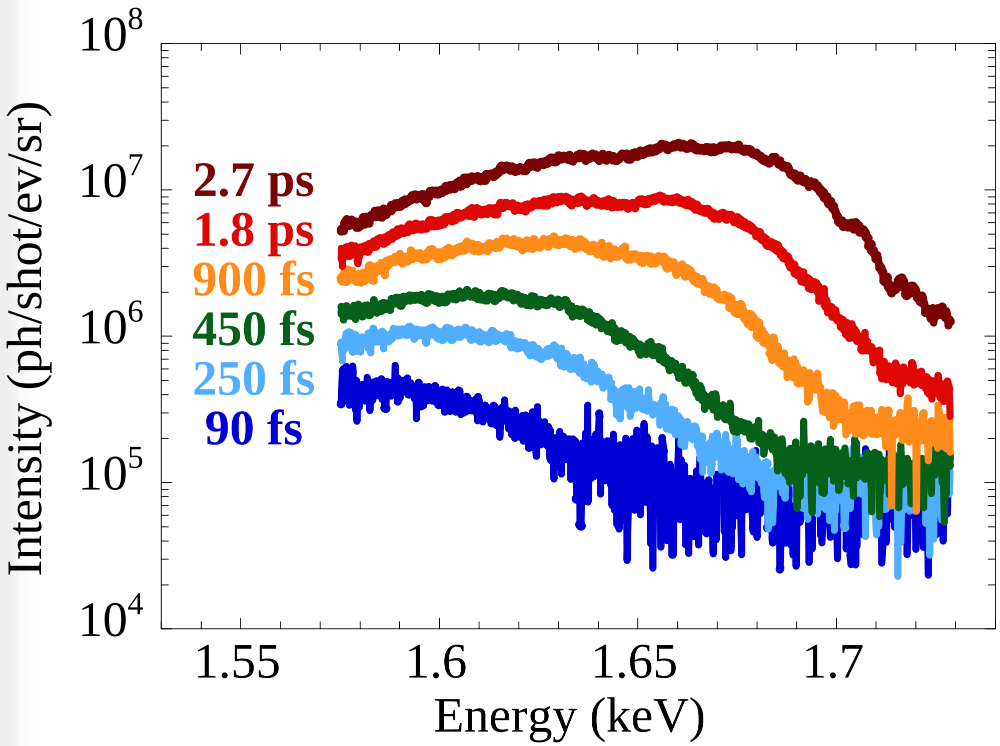




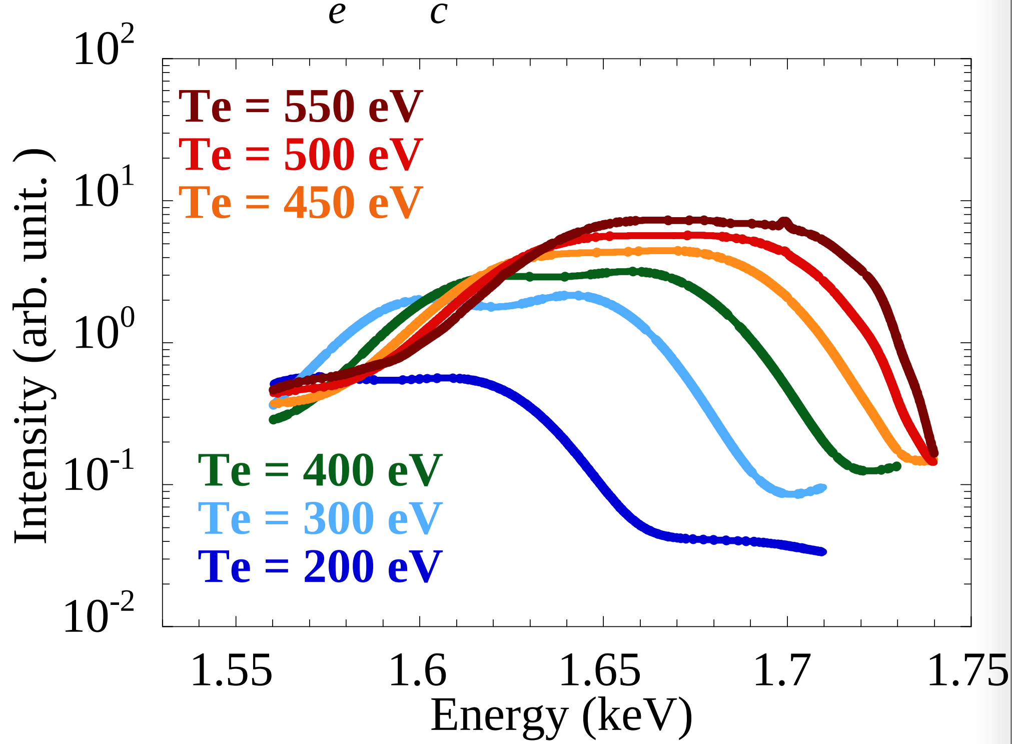
Adaptable, Broad band & **multi-keV** x-ray source

X-ray source optimization (1/2)

Variation of the laser duration
Er irradiated with E_{las} = 5 mJ

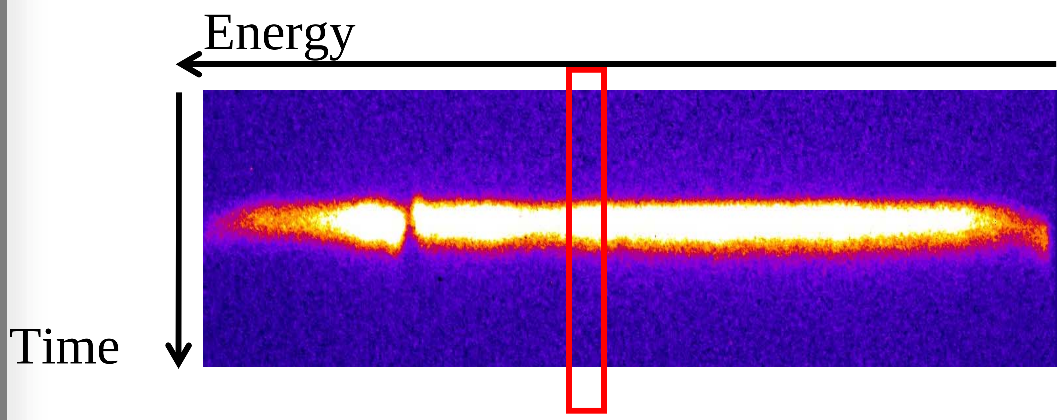


Averroes -Transpec Simulation
$N_e = N_c = 1.7*10^{21}$ cm^{-3}

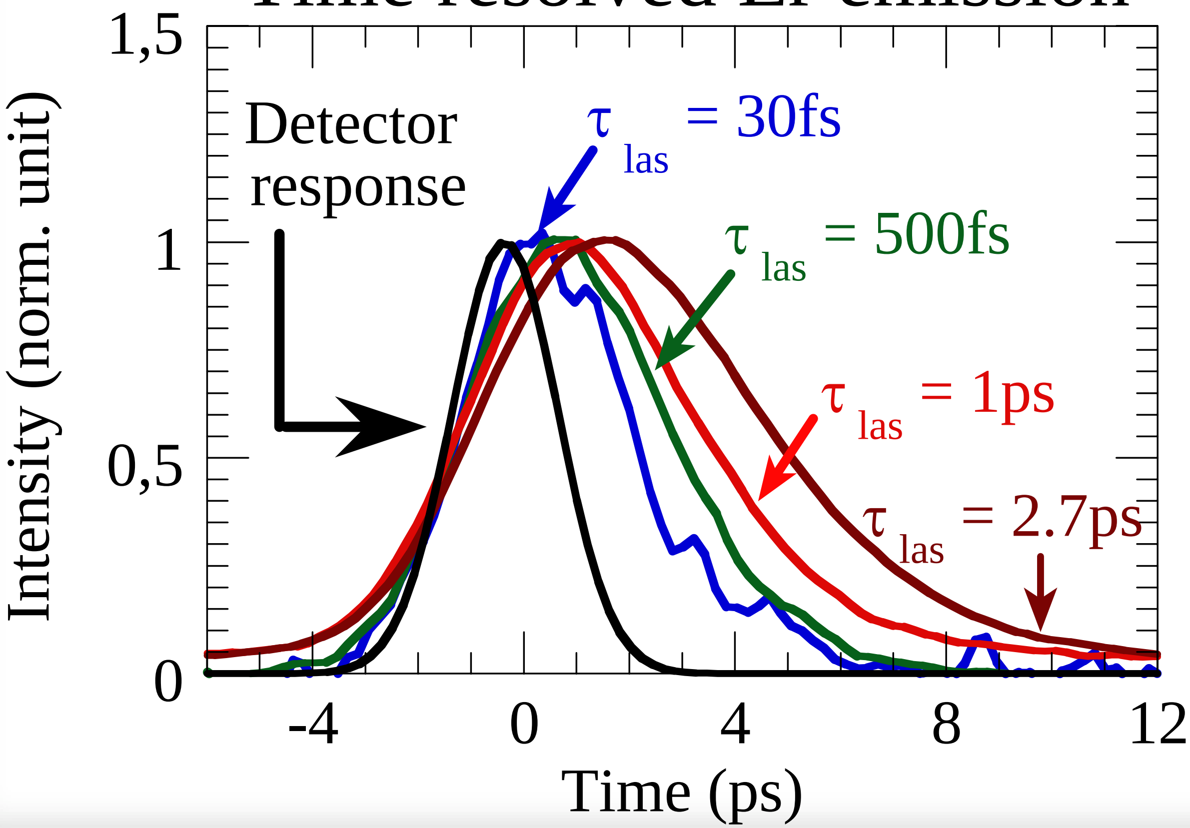


→ T_e increase with E_{las} & τ_{las}

X-ray source optimization (2/2)



Time resolved Er emission



τ_{las}	τ_{Er} (ps rms)
45 fs	1.7 ± 0.4
500 fs	2.2 ± 0.3
1 ps	2.8 ± 0.3
2.8 ps	3.7 ± 0.3
4.7 ps	4.2 ± 0.4

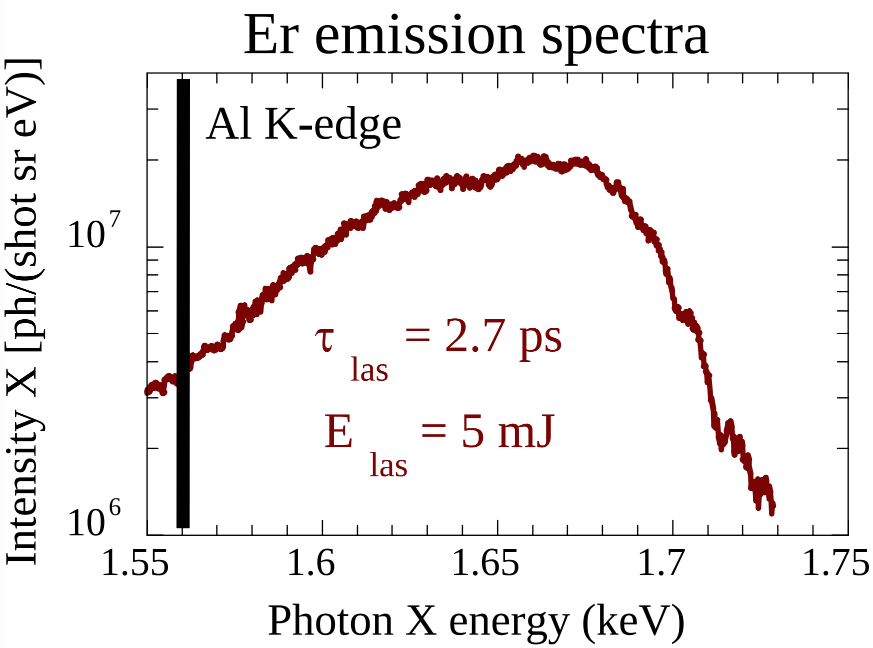
Time resolution : ~1.5 ps rms

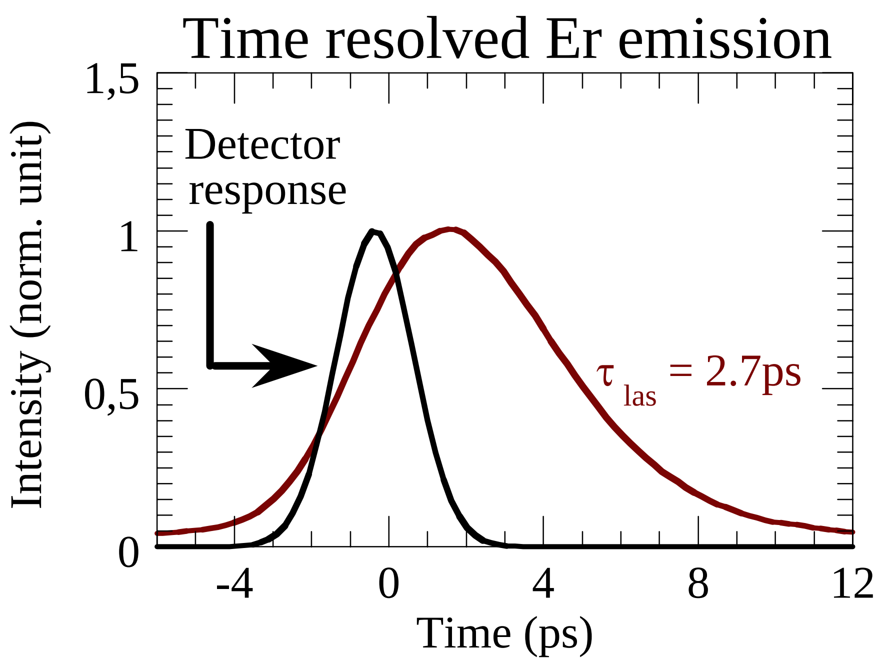
Broad band x-ray source **≤ 10 ps**

Intense, short (ps) and broad band X-ray source

- **Adaptable** spectral range
- **"Tabletop"** laser facilities (5 mJ, kHz - 100 mJ, 10 Hz)

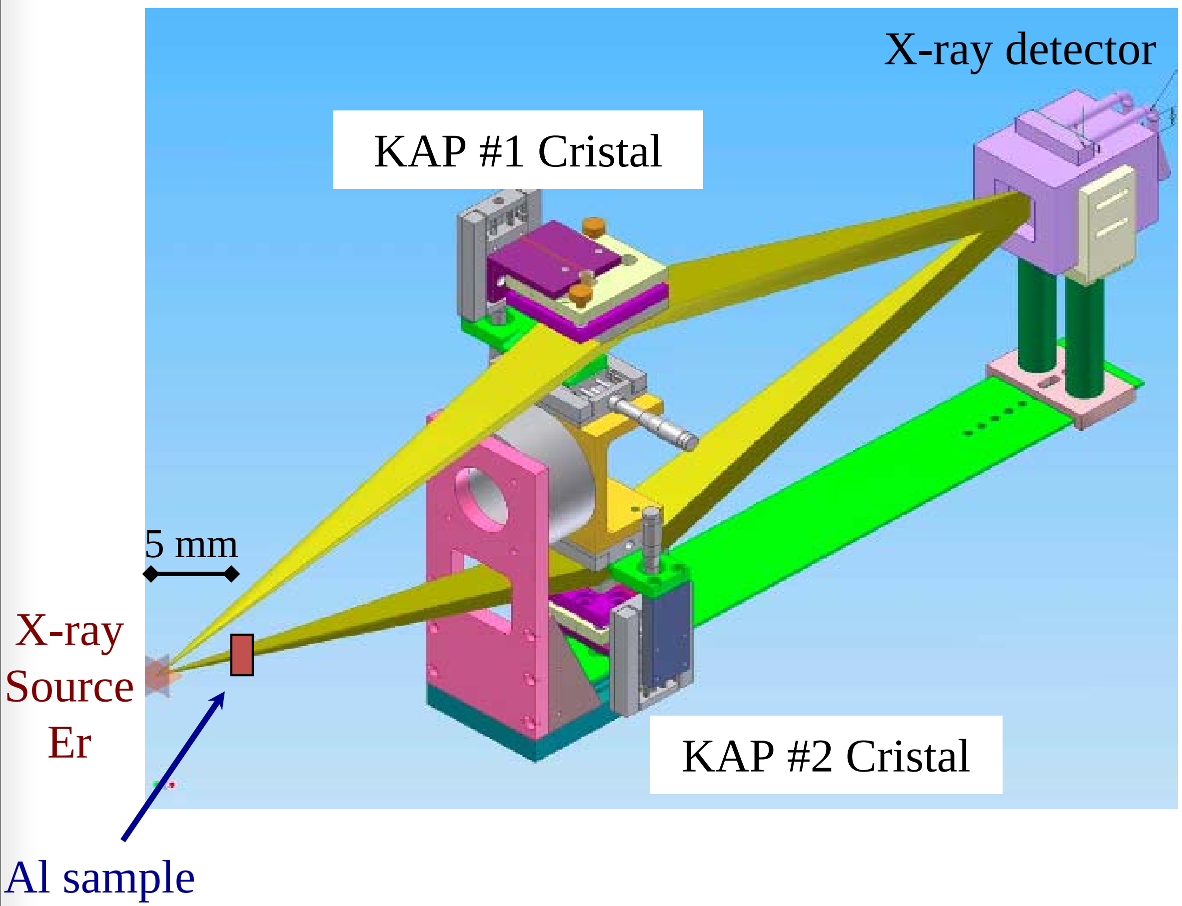
- Intense **~ 10^7 ph/shot/eV/sr** ↔ 1‰ E_{las} (integr. over 200 eV - 2π sr).
- Broad band spectral range over **~ 150 eV** on the Al K-edge.
- X-ray duration **~ 1 to 4 ps rms.**

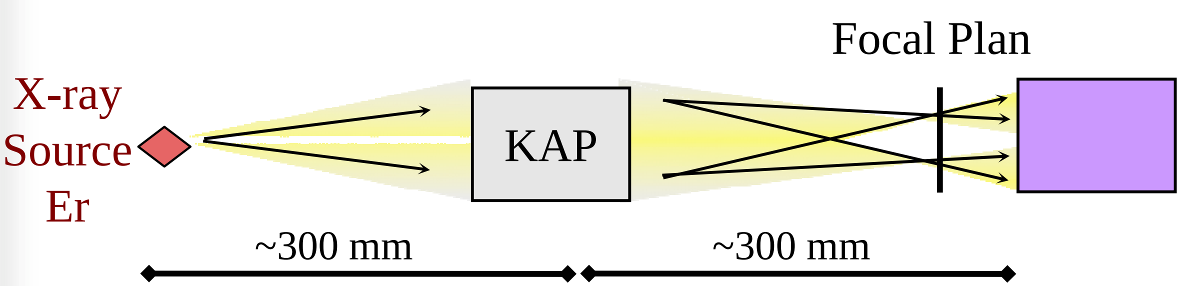


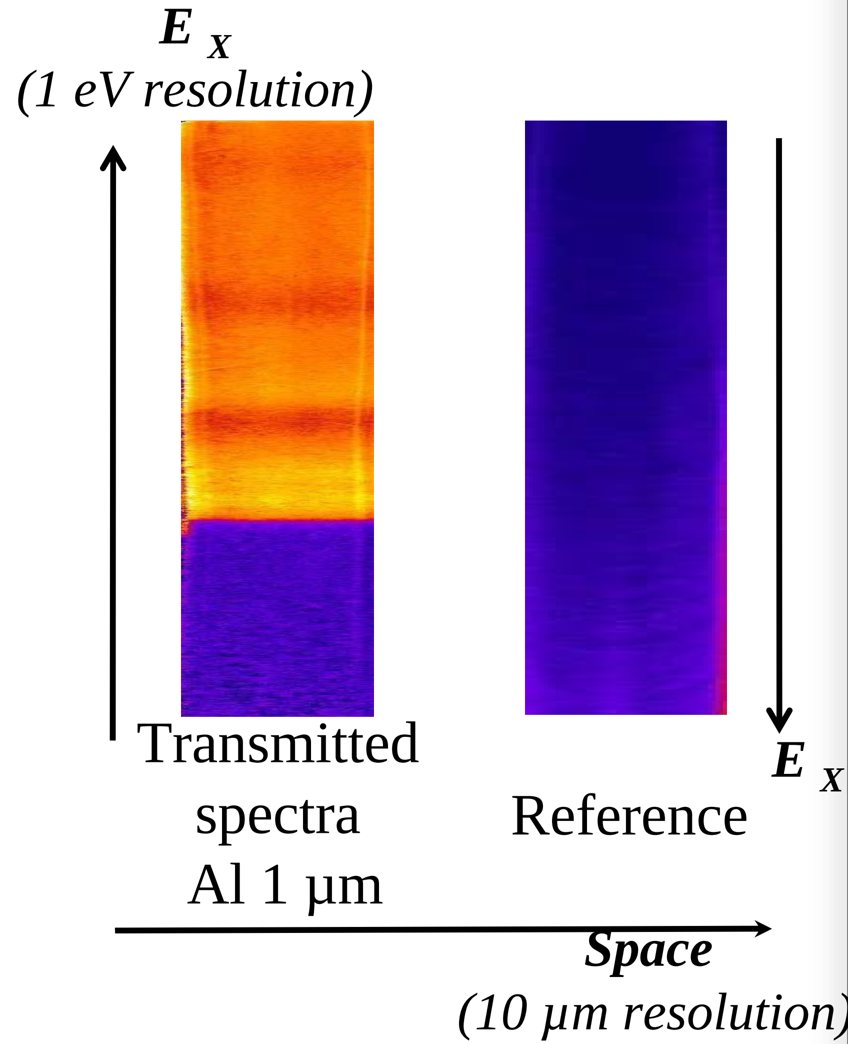


* *M.Harmand et al., Phys. of Plasma, 16 (2009) 063301*

X-ray absorption spectrometer



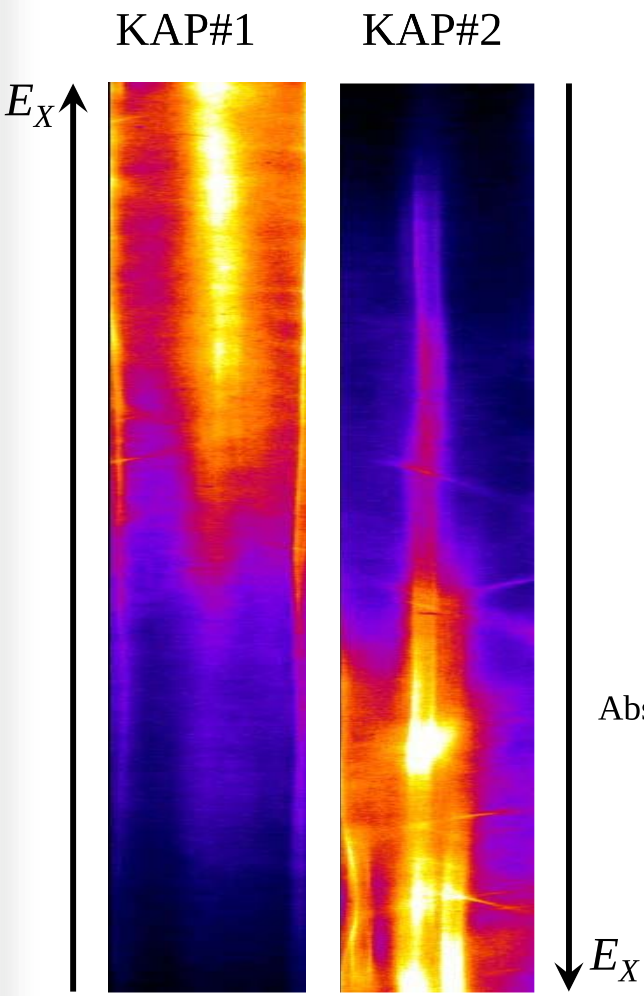




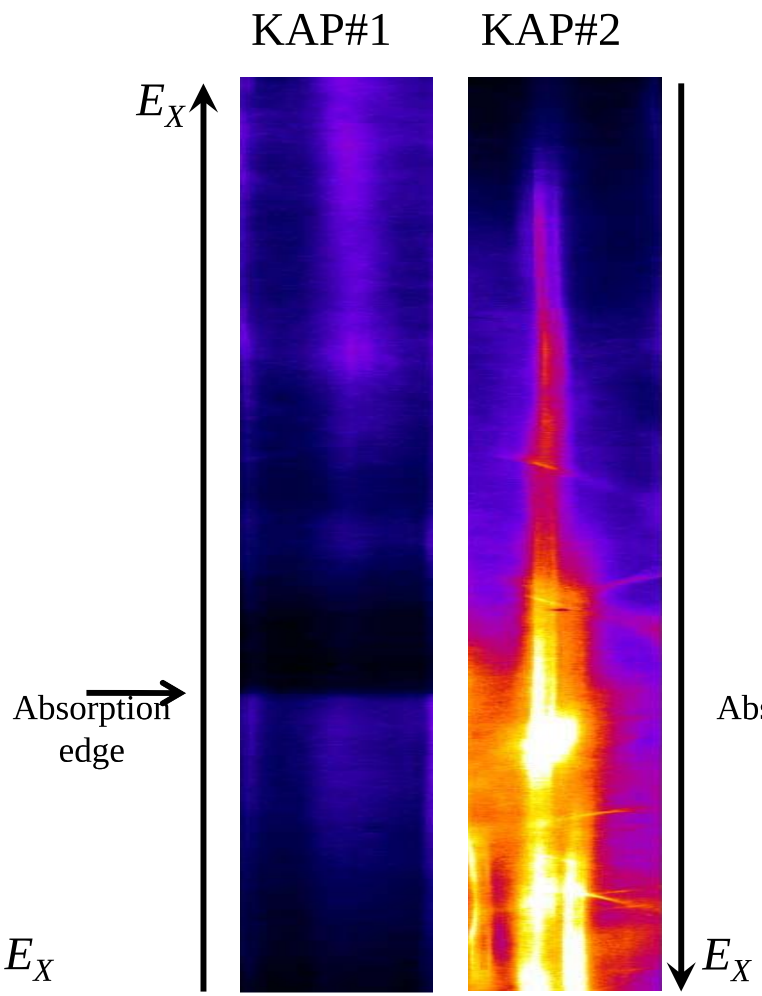
- Transmitted spectra & emitted spectra measured simultaneously.

- Spectral & Spatial resolution.

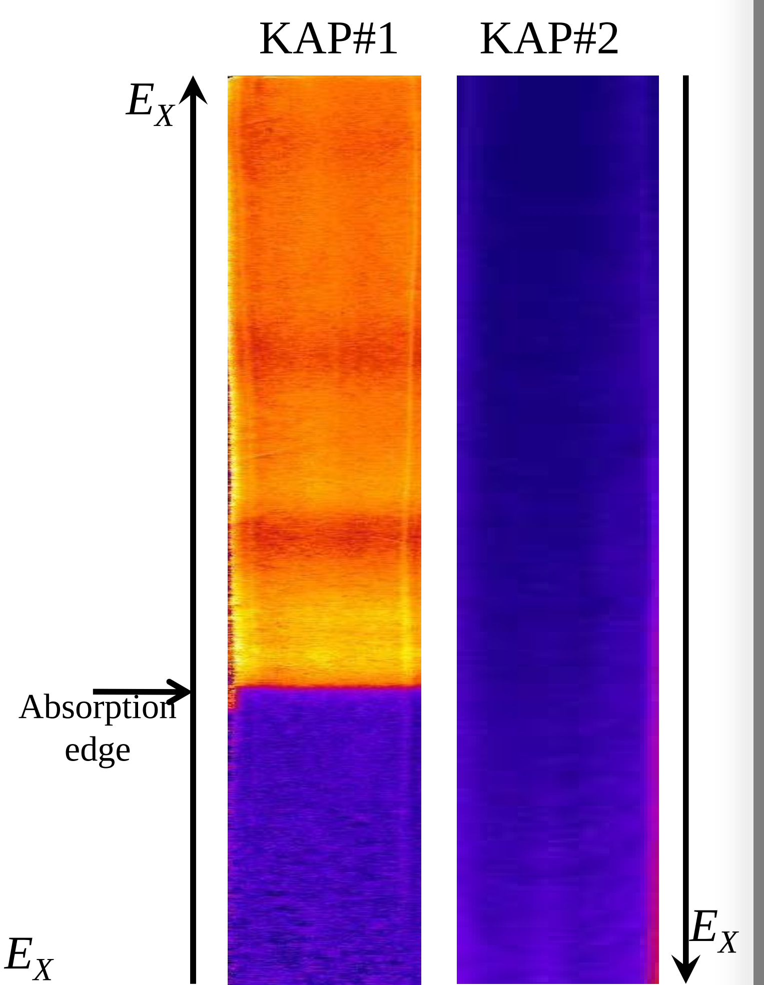
X-ray absorption spectra extraction



(1) Without Sample



(2) 1 µm Al on KAP#1



(3) Transmission T_{mes} corrected from defects

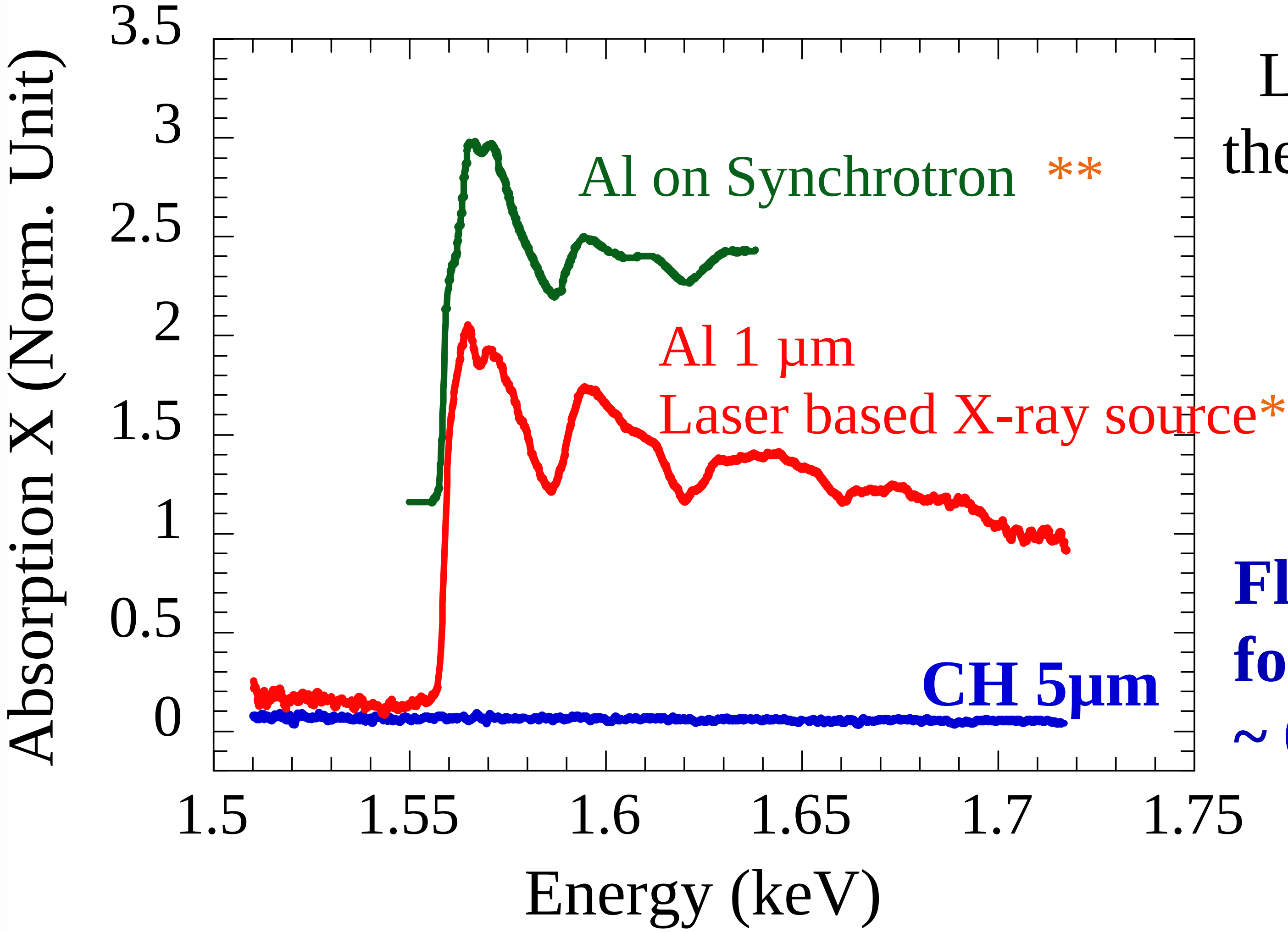
$$T_{mes}(E) = \frac{I_{transmis}}{I_{émis}}$$

→

$$\mu(E) = -\frac{1}{d}\ln\left(T_{mes}(E)\right)$$

High quality XANES spectra

Accumulation of 30s @ 1kHz – cold sample @ 300K)



Laser parameters for the X-ray backlighter:

E_{las} ~ 5 mJ

τ_{las} ~2.7 ps

1 kHz

Fluctuation for CH 5 µm ~ 0.3 - 0.6%

** A. Levy et al. Soumis à Rev. Sci. Instr. ; F. Dorchies et al. Appl. Phys. Lett. 93, 121113 (2008)*

*** J. Wong et al. Solid State Commun. 92 (1994) 559.*

XANES Calculation: QMD simulation

Ionic population

3D Pseudo-potentiel

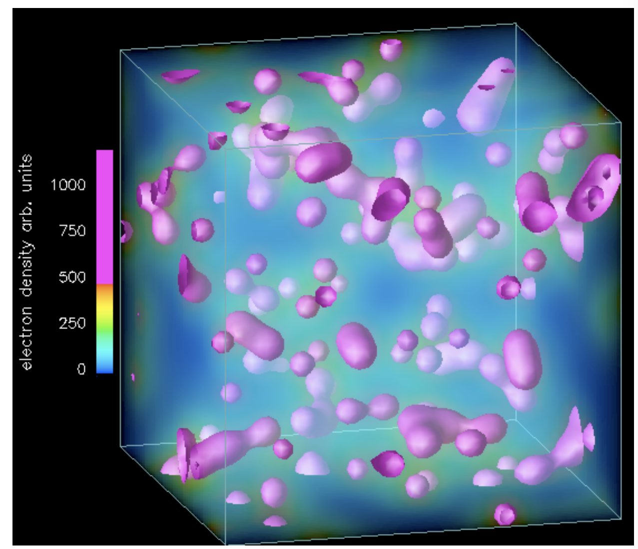
Abinit
Code*

Quantum forces on ions

T_e constant
V constant

Valence electron wave functions

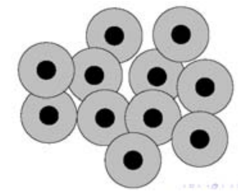
Calculation of the absorption **



** X. Gonze, Compt. Mater. Sci. 25 (2002) 478*
*** V. Recoules et al., Phys. Rev. B 80 (2009) 604110*
S. Mazevet et al., Phys. Rev. Lett. 101 (2008) 155001

XANES Calculation: WDM-FD simulation

DFT-NPA Equations → NPA (single ion + screening cloud) electron density; Pair interaction



↓

HNC Equations → Correlation function *g(r)* (ion – ion)

↓

g(r) sampling → N atom configurations

↓↓↓↓↓↓↓↓↓↓↓↓↓↓↓↓↓

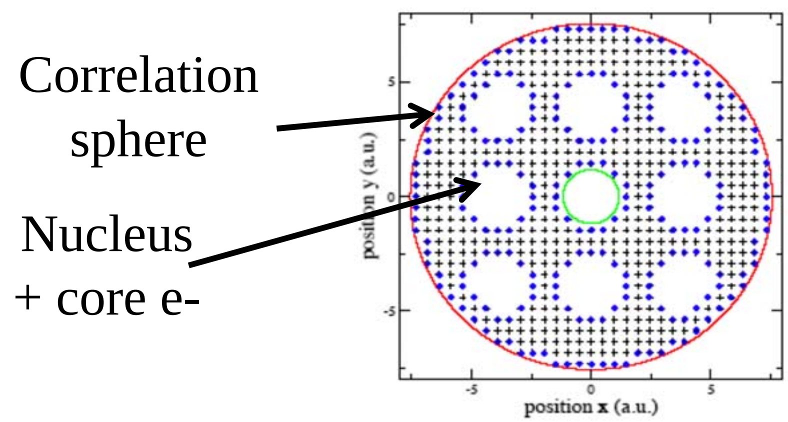
3D Finite Difference calculation → Cross section

↓

Average

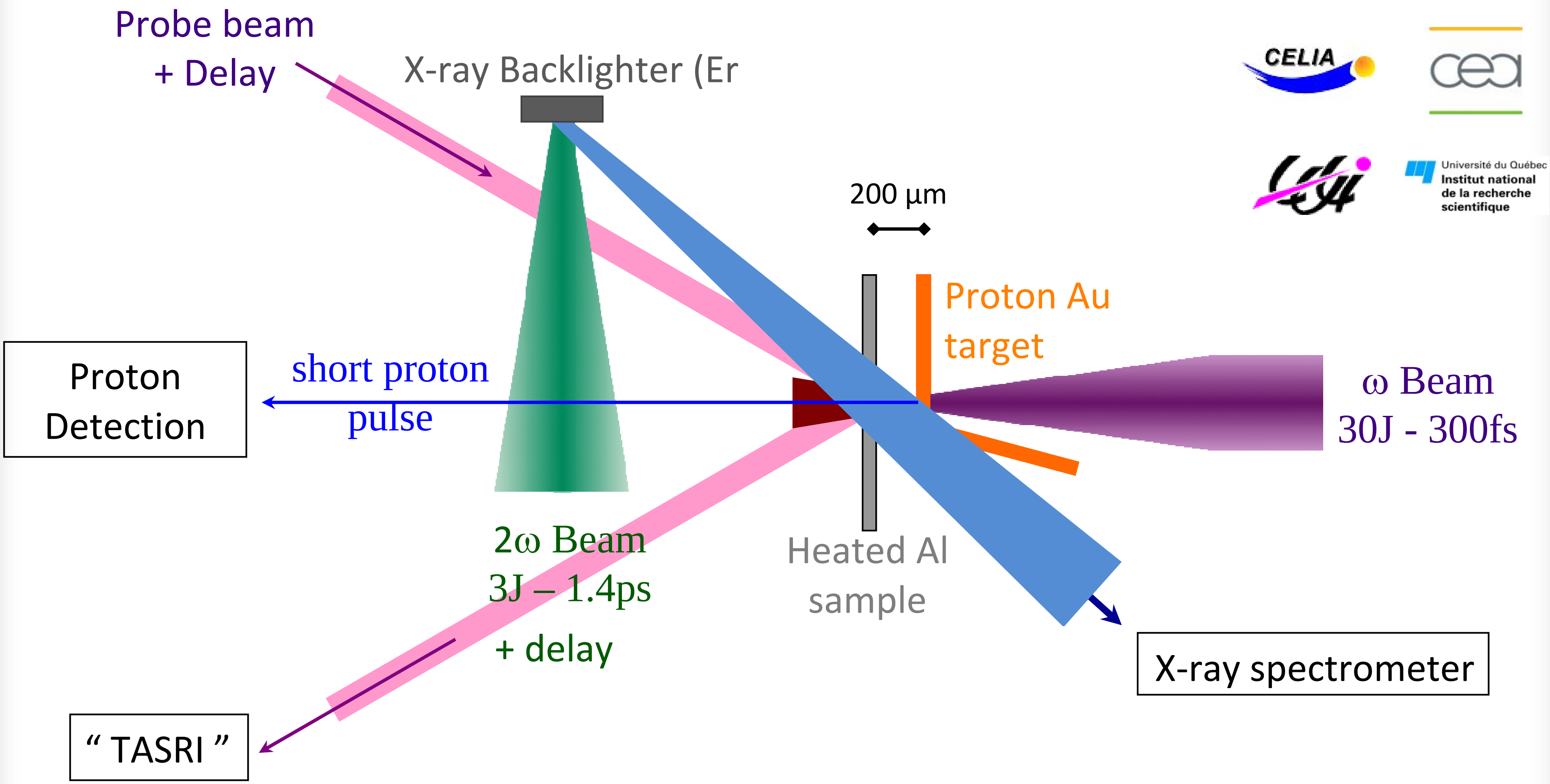
↓

XANES spectra



O. Peyrusse., J. Phys. Condens. Matter, 20 (2008) 195211

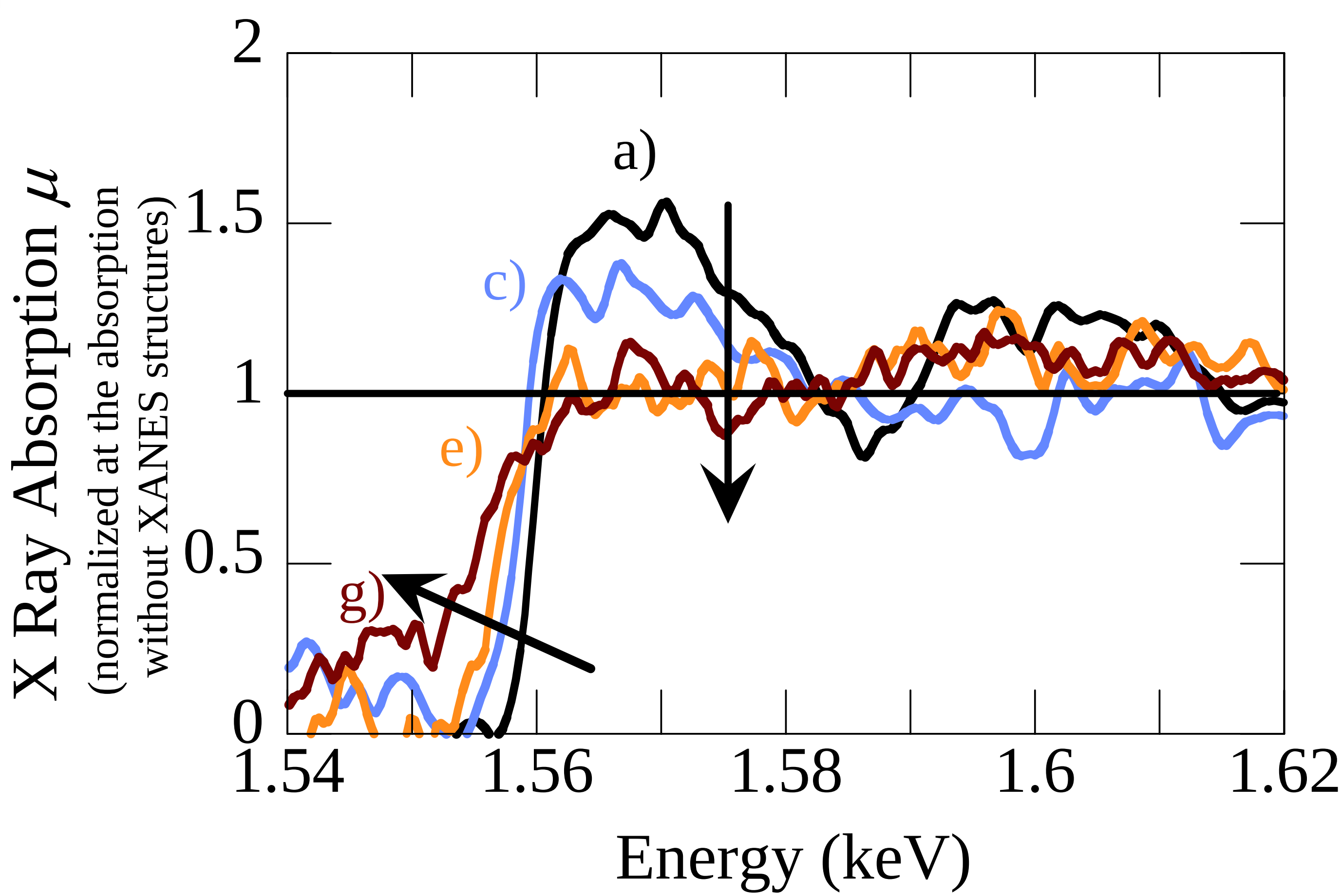
Isochoric Proton Heating Experiment (@ LULI 100TW)



Isochoric heating* of an Al target at solid density

*A. Mancic et al., Phys. Rev. Lett. 104 (2010) 03500 ; A. Levy et al., PPCF 51 (2009) 124021

Isochoric Proton Heating Experiment (@ LULI 100TW)



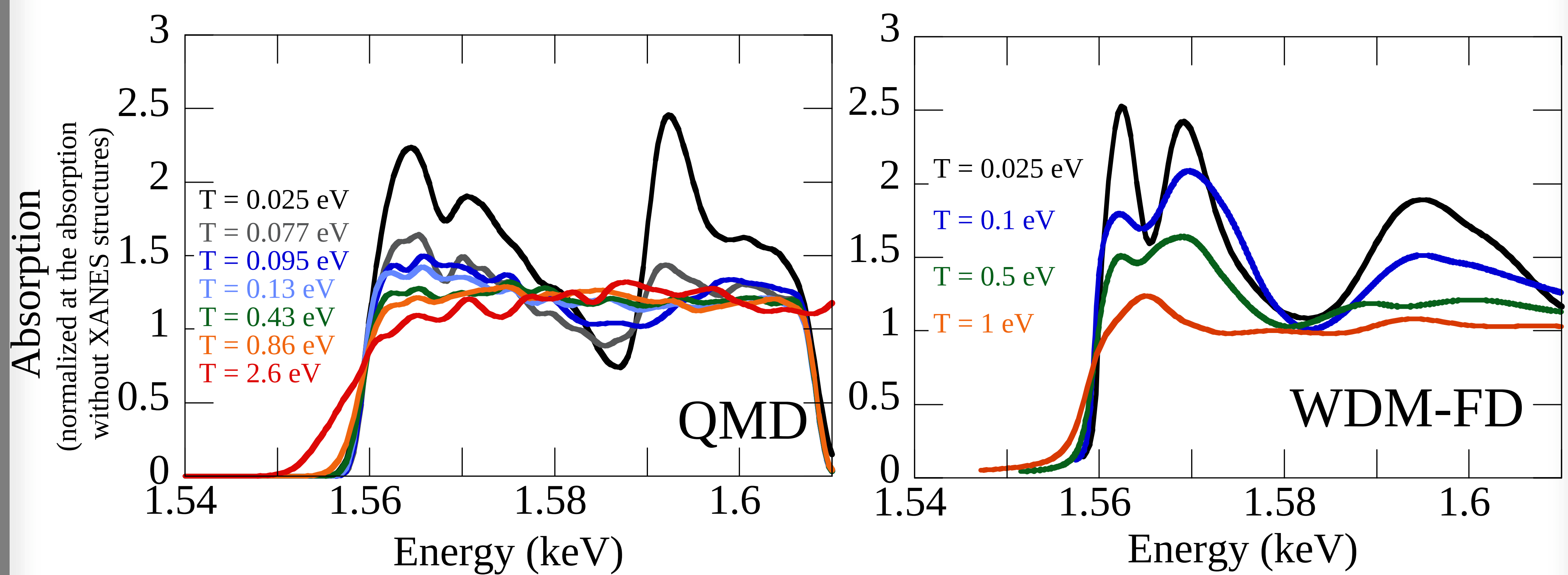
Temperatures deduced from Fermi – Dirac function

a) "Cold" Al
c) 0.27 ± 0.15 eV
e) 1.40 ± 0.20 eV
g) 2.74 ± 0.25 eV

- The absorption edge slope is used to measure *T*
- XANES structures are vanishing when *T* increases.

Absorption Spectra Interpretation

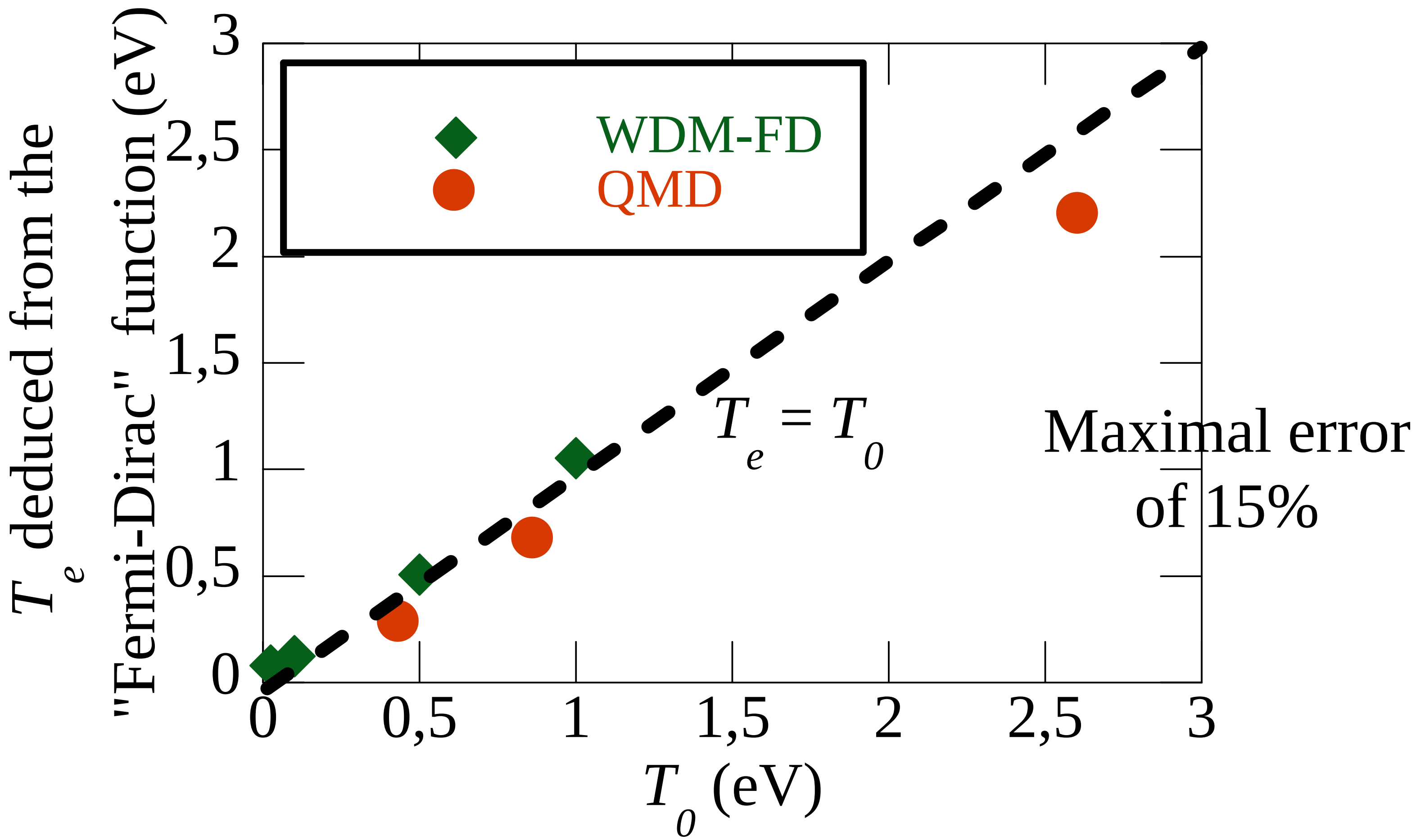
QMD & WDM-FD calculation in function of *T*



- The slope decreases when the temperature increases
- XANES structure vanishing

Validation of the T_e extraction

using the Fermi-Dirac function

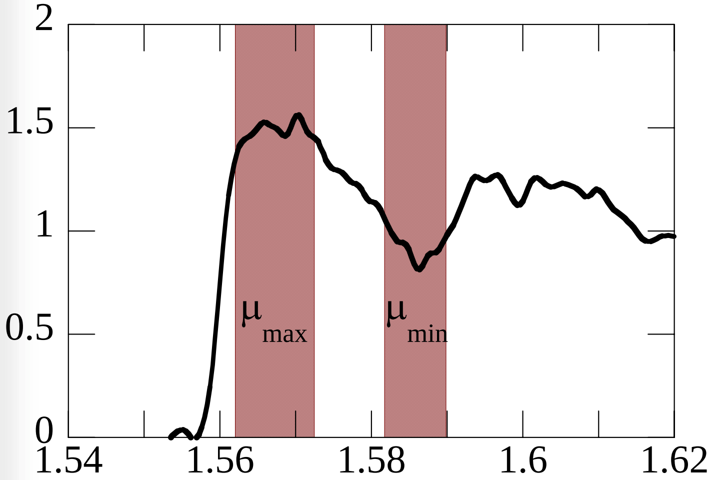


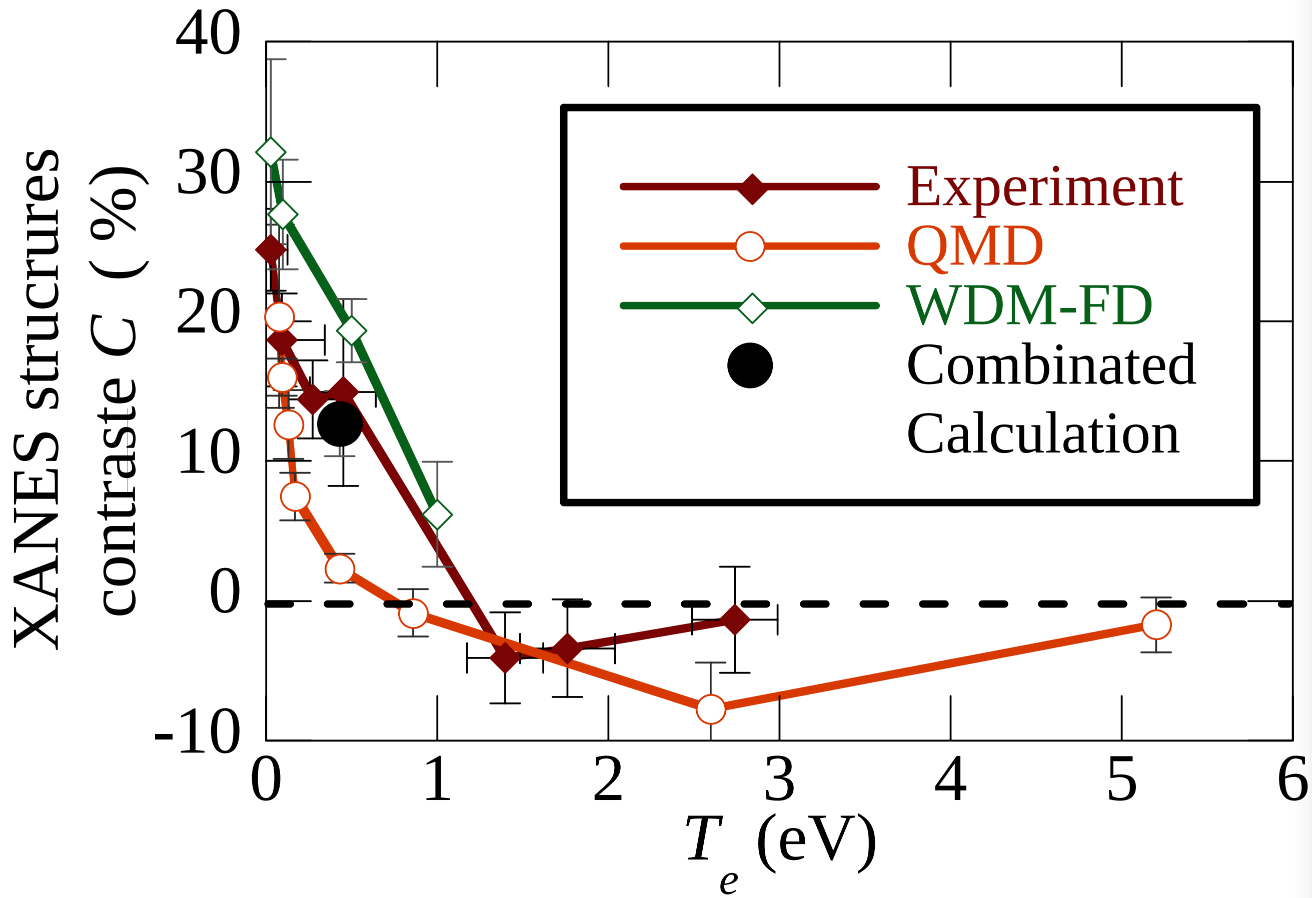
➡ Until ~ 3 eV at Aluminum solid density

XANES structures vanishing

Variations of the XANES Contrast C

$$C = \frac{\mu_{\max} - \mu_{\min}}{\mu_{\max} + \mu_{\min}}$$



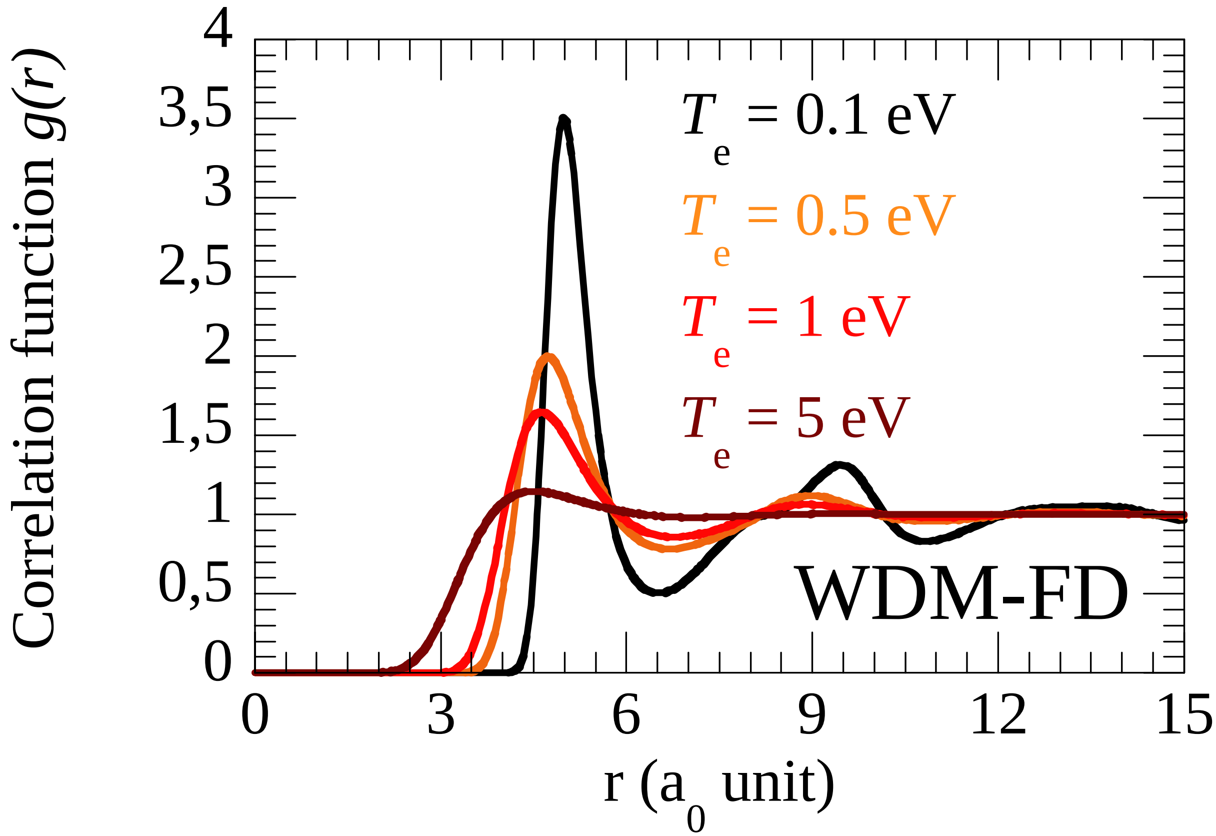


- XANES structure are vanishing more quickly with the QMD calculation.

- Combinated calculation : 3D spatial configuration obtained with QMD + XANES spectra obtained with WDM-FD

Interpretation of the XANES structure vanishing

Ion – ion correlation function *g(r)*



→ Loss of ion - ion correlations implies XANES contraste vanishing.

Interpretation :

Loss of ions – ions correlations

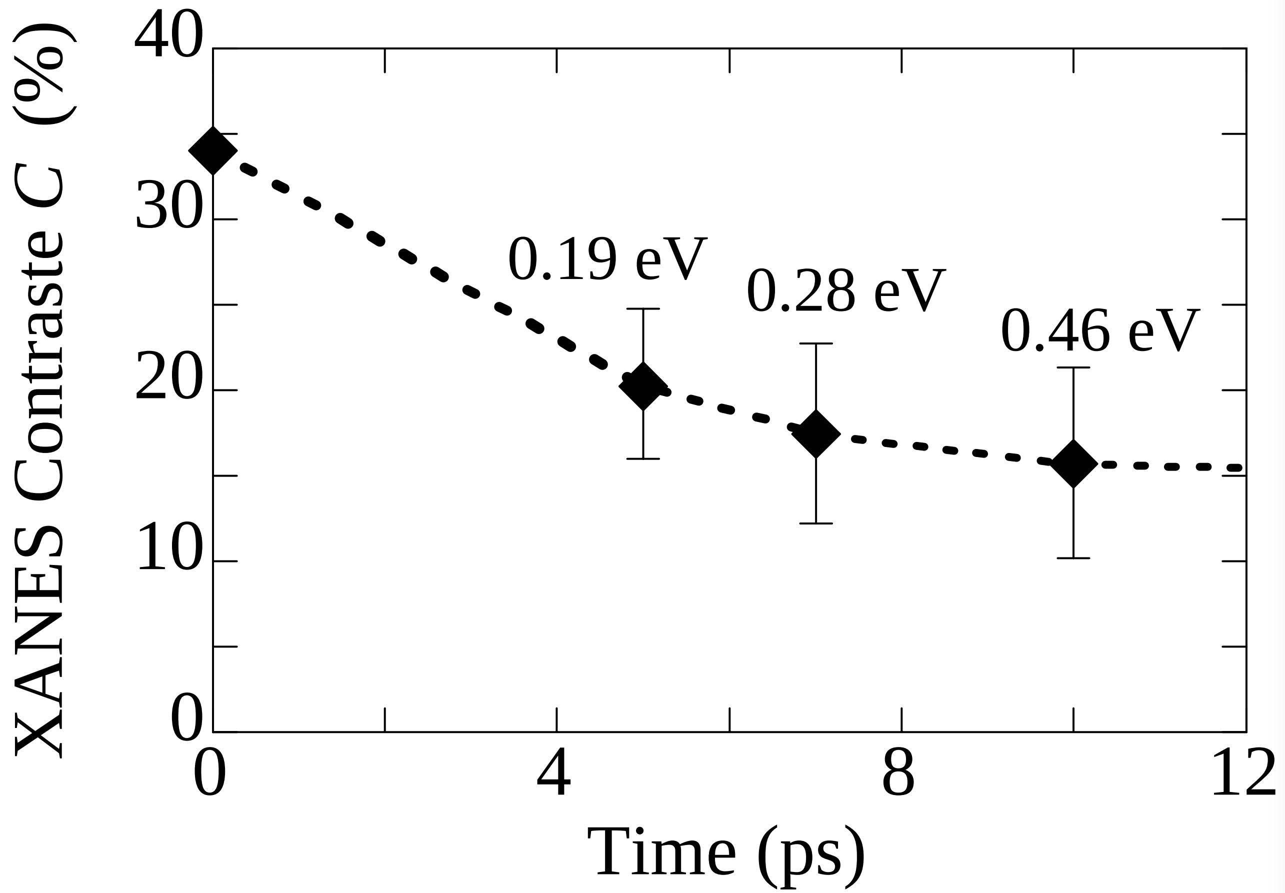
Hydrodynamic code proton energy deposition (ESTHER)

↓

T_e in function of the time

↓

DMQ calculation for different T_e



➡ Estimation of the upper bound at ~10 ps for the loss of the short range ordering (limitated by the proton heating time)

Summary & perspectives

- "Tabletop" X-ray sources from M-band emission of high-Z elements
  - Well controlled & well understood X-ray source
  - M-band Emission → maximal spectral range @ 5 keV
  - Minimal duration ps, sub-ps for clusters

- X-ray absorption Spectrometer with fluctuations < 1%

➔T, local order (ion - ion correlations), ρ

- Isochoric proton heating:

➔ Observation of a significant loss of WDM correlation around 1eV within 10 ps (upper limit)

Summary & perspectives

- Explore higher densities: laser shock (see A. Benuzzi-Mounaix Talk)

- Explore WDM dynamic:
  - by ultrafast laser heating (CELIA on « Eclipse » laser, 100 mJ, 10 Hz, 40 fs)
  - by FEL heating

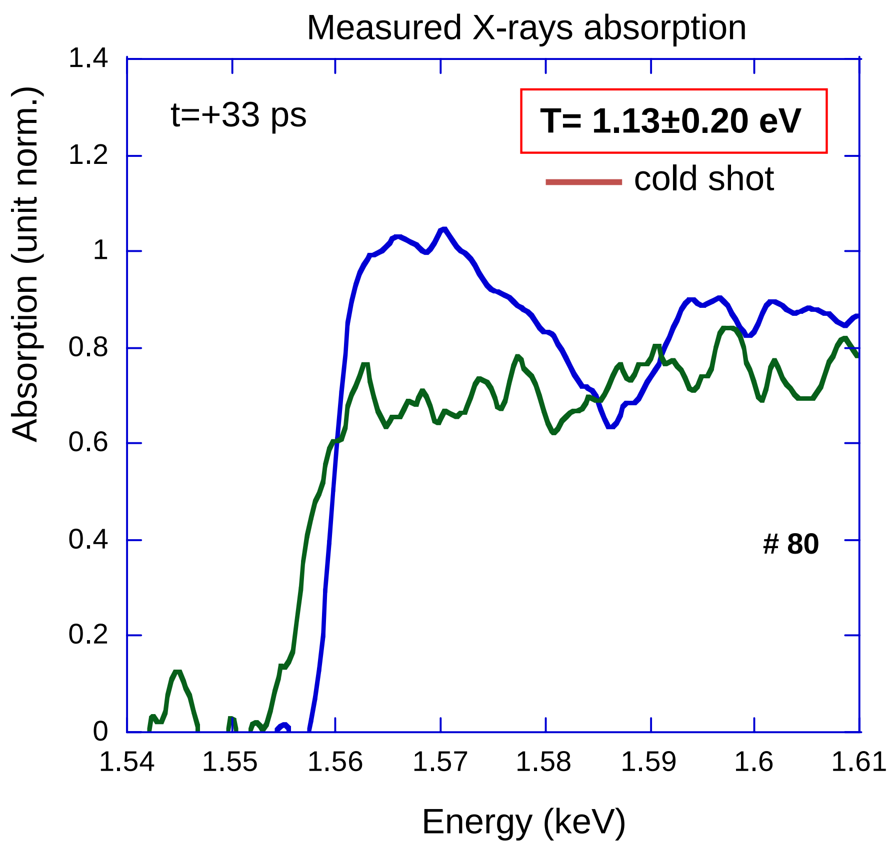
- Explore new elements: (ex: Fe K-edge @ 7.112 keV)
  - Adapted X-ray source (Betatron sources, Thomson Scatt sources, Bremsstralhung emission…)
  - Improve calculations…

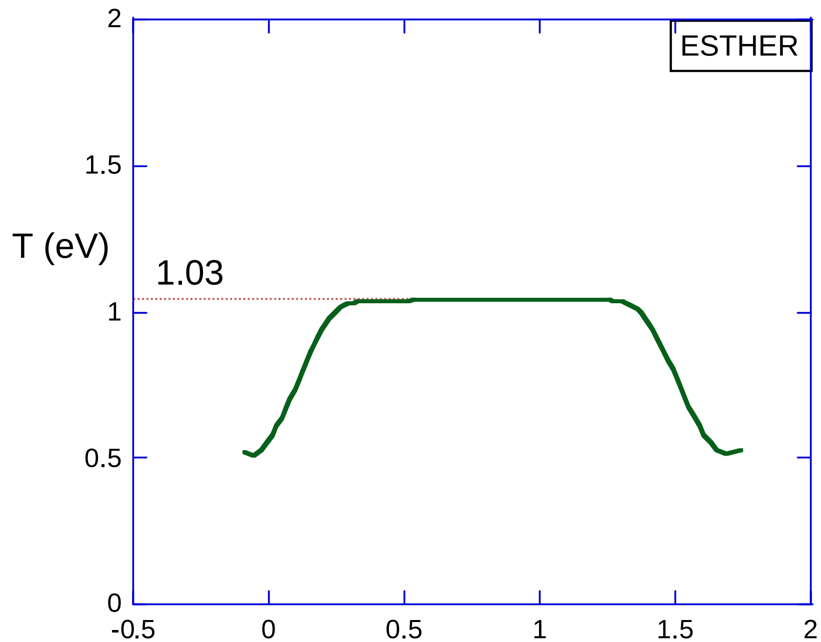
Thank you for your attention

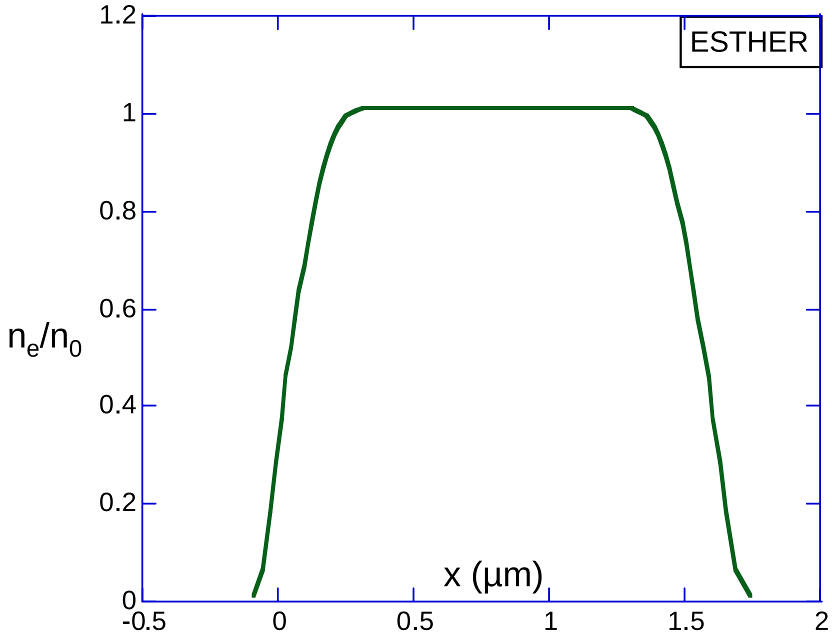
Isochoric proton heating

Heated target: **Al 1.6 µm**

TASRI: no data

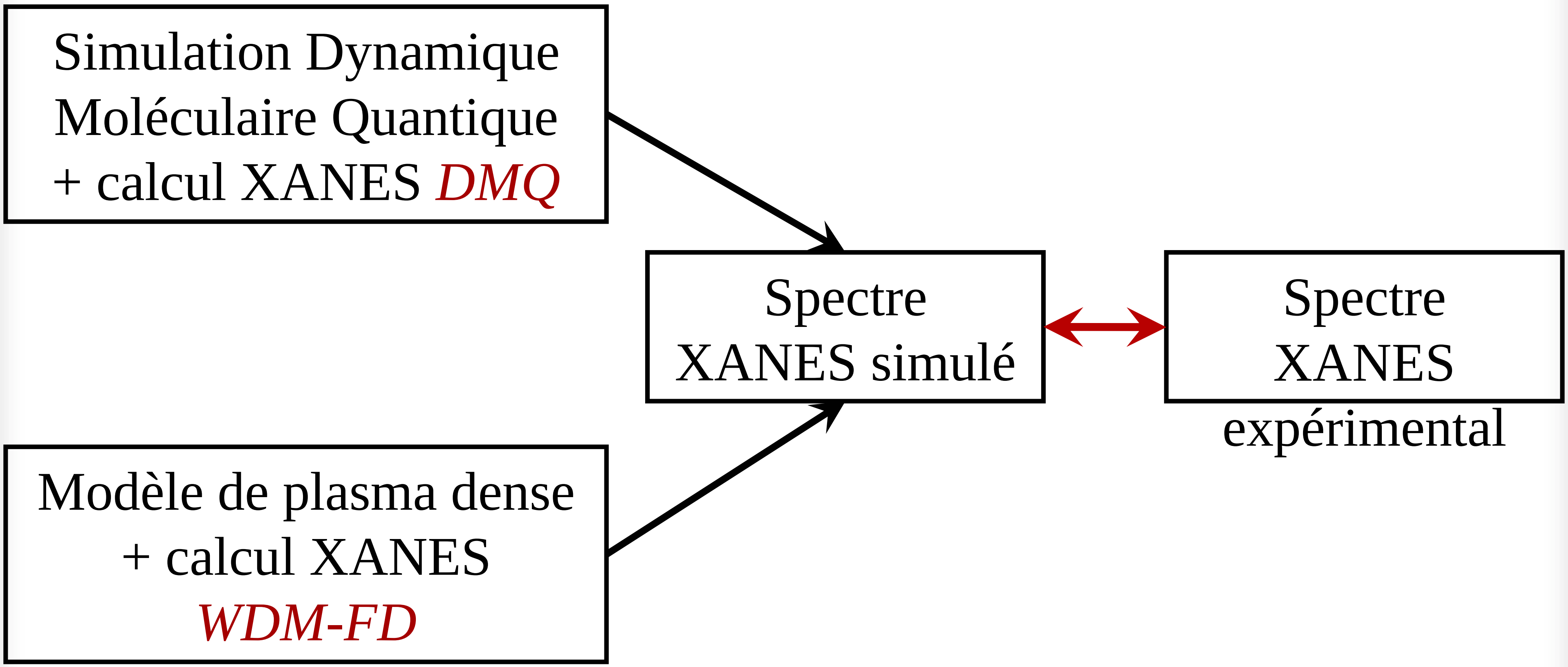






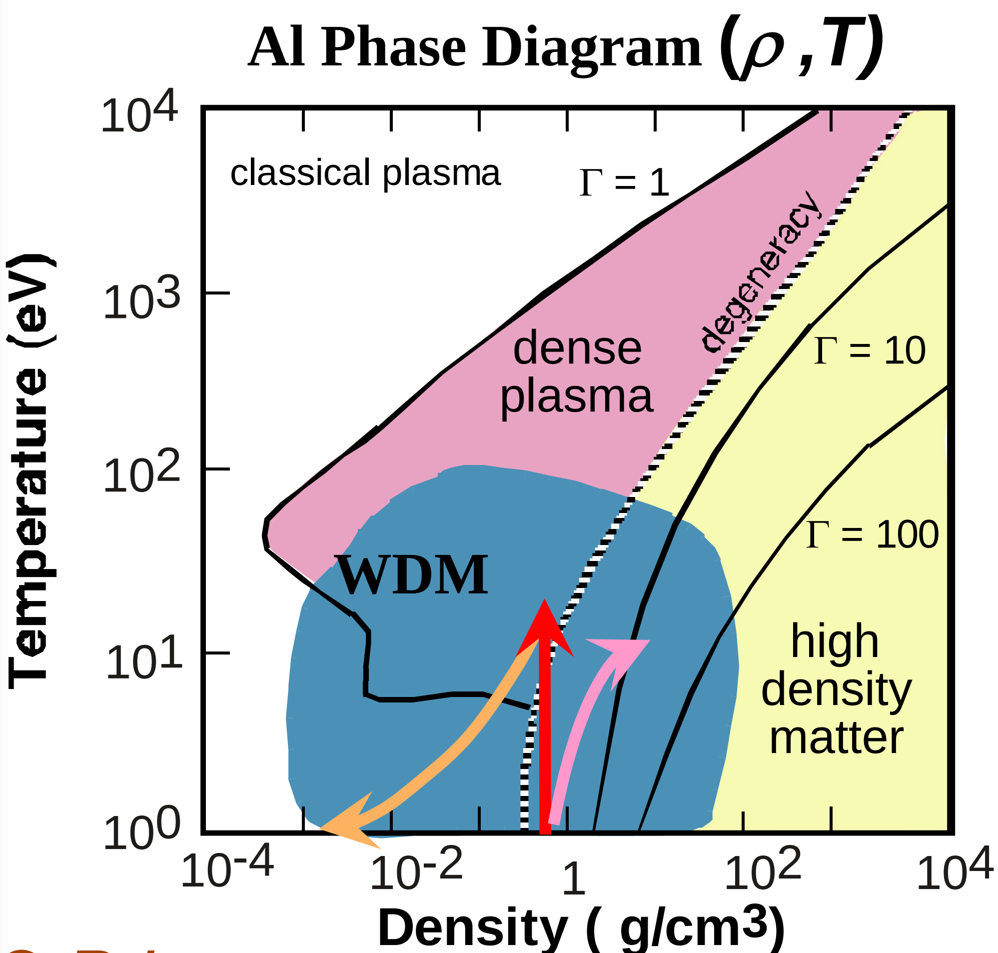
Analyse des spectres d'absorption X

Comparaison à des calculs de spectres XANES



Les informations pertinentes (fonction de corrélation…) sont calculées par les codes DMQ et WDM-FD.

WDM Studies



- Between solid and plasma state:
  - strongly correlated ions
  - degenerated electrons $T_e \sim T_F$

- Need experimental data

Isochoric heating by laser, by protons or by photons + ***isentropic expansion***

Shocks

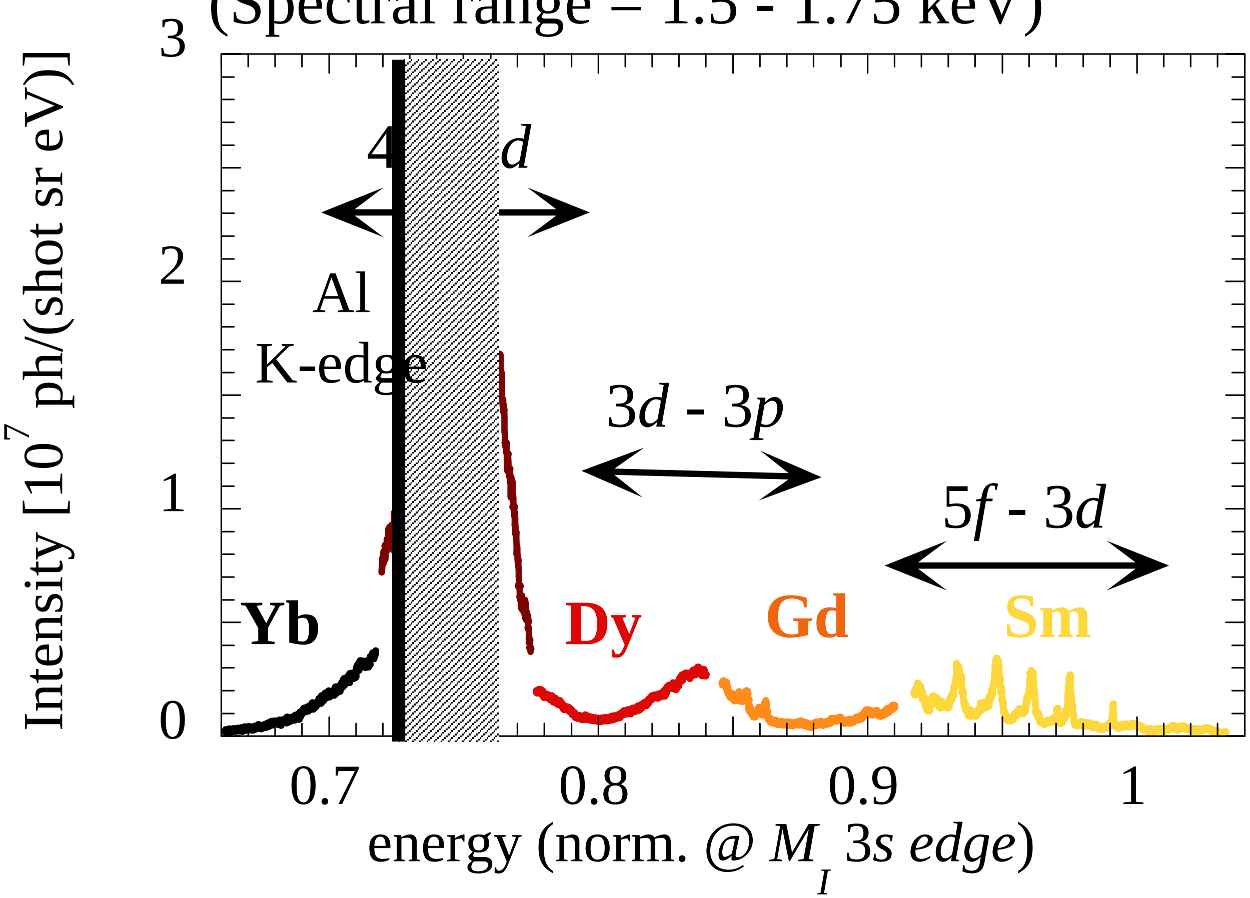
© R.Lee

→ Experimental technique to study a disordered state giving access to the ion-ion correlations and the e- density of states

Broad M-band emission from a high-Z elements

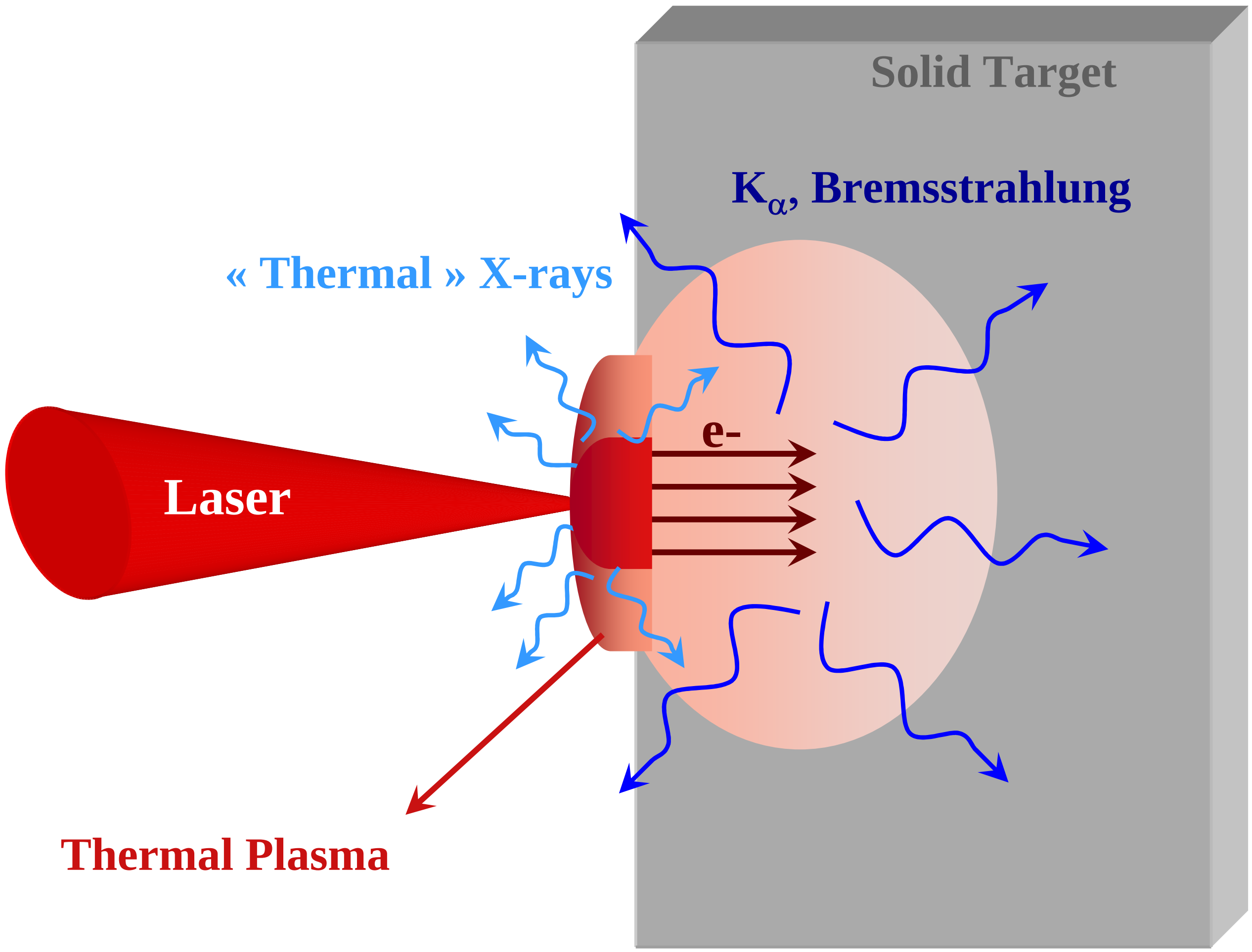
High-Z emission spectra (E_{las}=5mJ, τ_{las}=2.7ps)

(Spectral range = 1.5 - 1.75 keV)



→ *4f – 3d* transition array: intense & broad band X-ray emission

Laser-plasma X-ray sources



➡ X-ray source **intense** and **ultra-short** (~ few ps) due to a fast plasma expansion