Femtosecond structural probing of warm dense matter with Betatron laser-based x-ray source

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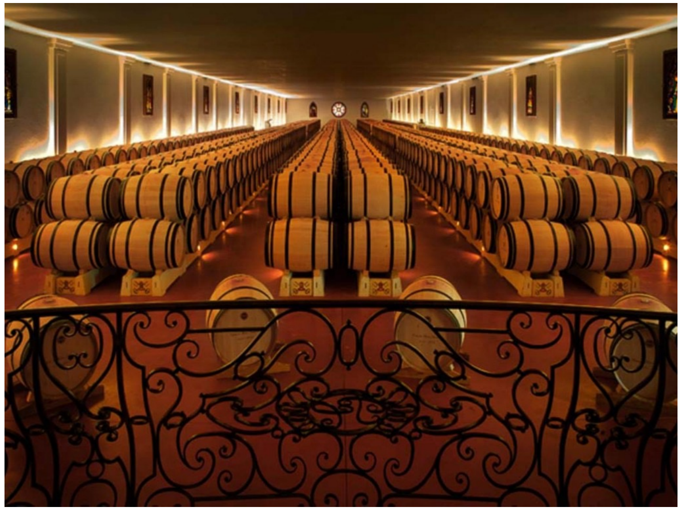

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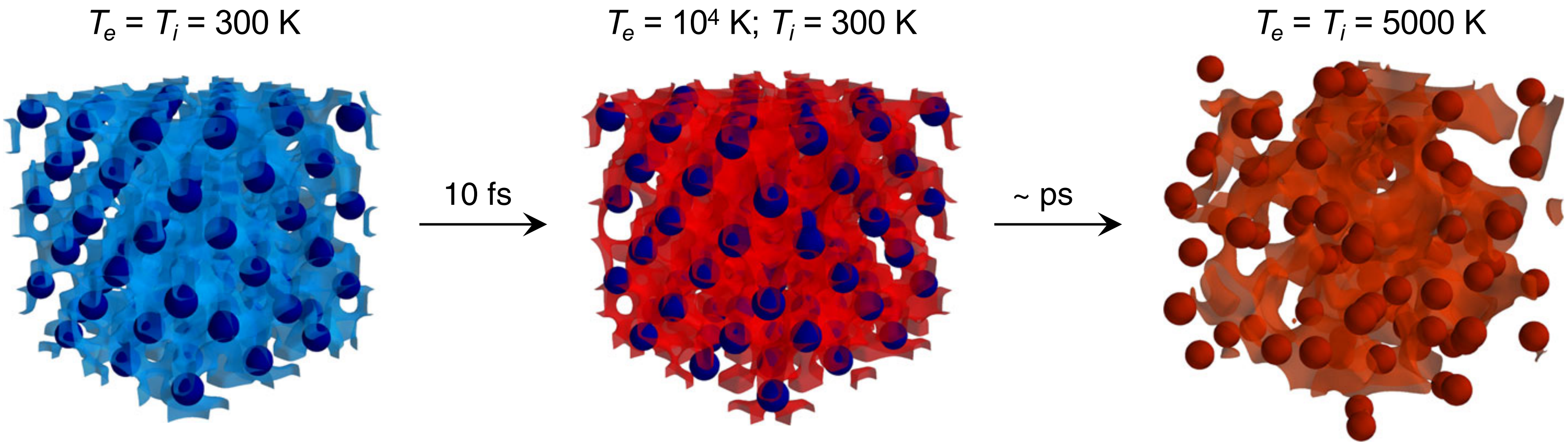
Non-equilibrium Warm Dense Matter

- One way to achieve WDM is to use femtosecond laser isochoric heating
  - Ultrafast and selective excitation of the electrons
    - ✓ Electron – lattice/ions thermal equilibration
    - ✓ Electron energy transport
    - ✓ Non-equilibrium phase transitions *(bond softening, bond hardening, …)*
  - Access to the electron –nuclei dynamic interplay and more stringent simulation testing



K. Widmann et al., *Phys. Rev. Lett.* ***92*** *125002 (2004)*

A macroscopic approach : two-temperature model *(TTM)*

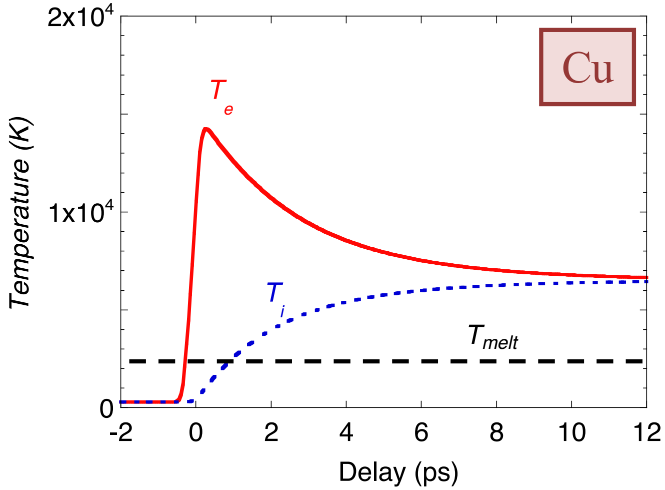
- A crude and simplified model
  - Macroscopic approach
  - Electrons are just described by T_e
  - Lattice / ions by T_i

- Prediction of the electron / ion dynamic
  - Electron heating (C_e)
  - Electron energy transport (κ_e)
  - Electron – ion thermal equilibration $(C_e,\ G_{e\text{-}i},\ C_i)$
  - Role of the expansion *(TTM integrated in hydro code)* **
  - Estimation of the “thermal” melting time $(T_i \geq T_{melt})$

- Physics is carried over into the appropriate choice of coefficients

Two-temperature model *

$$C_e \frac{dT_e}{dt} = -G_{e-i}(T_e - T_i) + \nabla(\kappa_e \nabla T_e) + S(t)$$
$$C_i \frac{dT_i}{dt} = G_{e-i}(T_e - T_i) + \nabla(\kappa_i \nabla T_i)$$



* *S. I. Anisimov* et al., *JETP **39**, 375 (1974)*
** Esther code in *J.-P. Colombier* et al., *Phys. Rev. B **71**, 165406 (2005)*

Experimental investigation of non-equilibrium WDM

- Need for time-resolved diagnostics *(ps down to fs)*
  - Optical probing *(reflectivity, transmission, phase, … dielectric function)*

  ⇒ Indirect observable that relies on models to be connected with the atomic scale

  - X-ray probing

  ⇒ Provides, in principle, a "direct" access to the atomic scale

    ✓ Diffraction (XRD) *(including electron diffraction)*

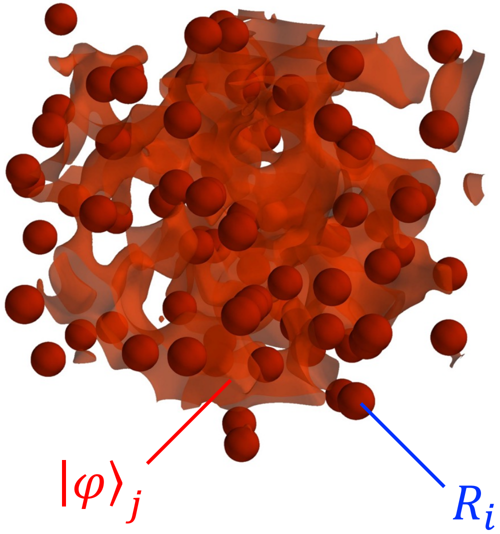
    ⇒ Structure factor

    ✓ Scattering (XRTS)

    ⇒ Structure factor *(elastic)*, T_e, n_e, Z^* *(inelastic)*

    ✓ Absorption spectroscopy (XAFS)

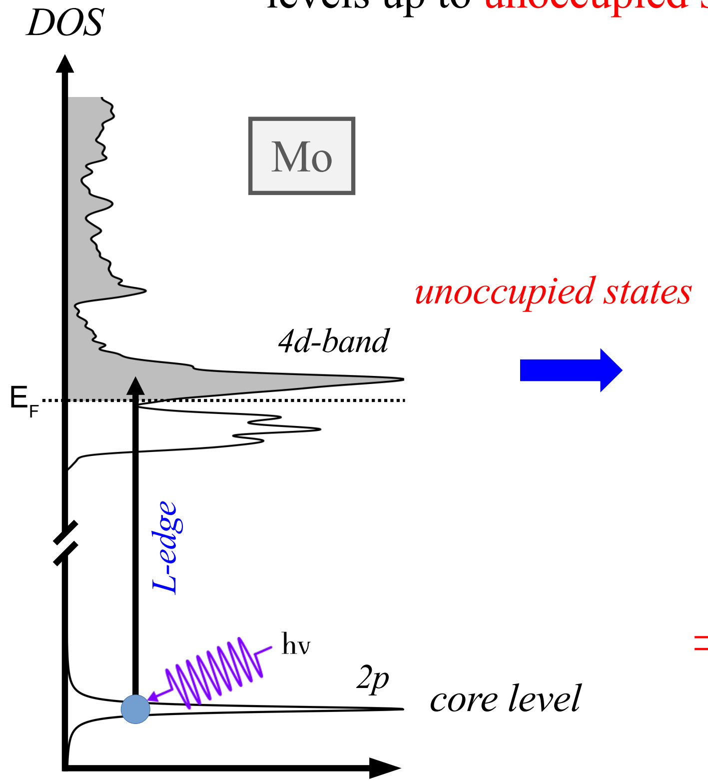
    ⇒ Electron structure & atomic order *(short-range or periodicity)*

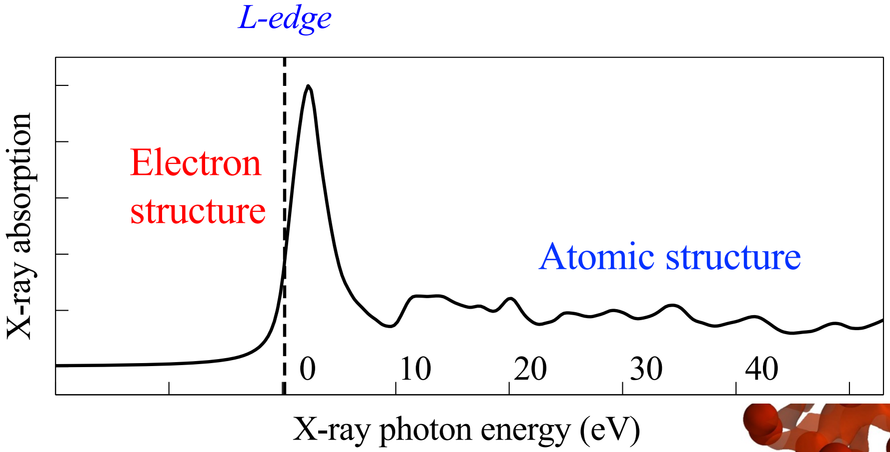


A. Rousse et al., *Nature (London)* ***410*** *65 (2001)*
R. Ernstorfer et al., *Science* ***323*** *1033 (2009)*
G. Gregori et al., *Phys. Rev. E* ***67*** *026412 (2003)*
B. I. Cho et al., *Phys. Rev. Lett.* ***106*** *167601 (2011)*

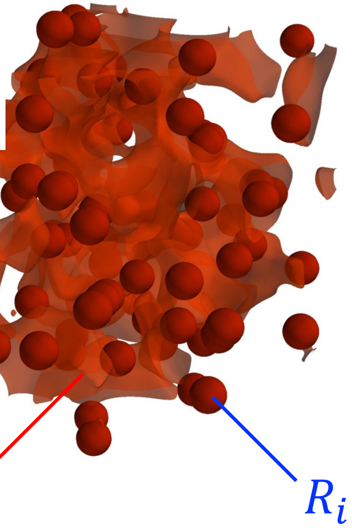
Principle of X-ray Absorption Near-Edge Spectroscopy *(XANES)*

- X-ray absorption spectroscopy near a given edge *(K, L, M => element selective)*
  - Dominated by the photoelectric effect and associated electron transitions from core levels up to unoccupied states near the continuum





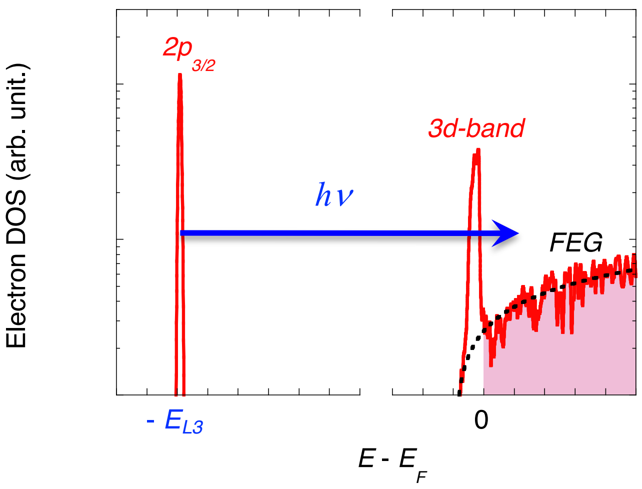
⇒ Need to transpose
the knowledge in interpretation
from solid-state physics to WDM

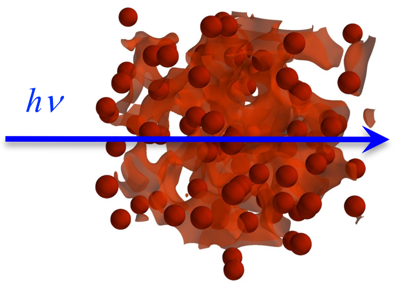


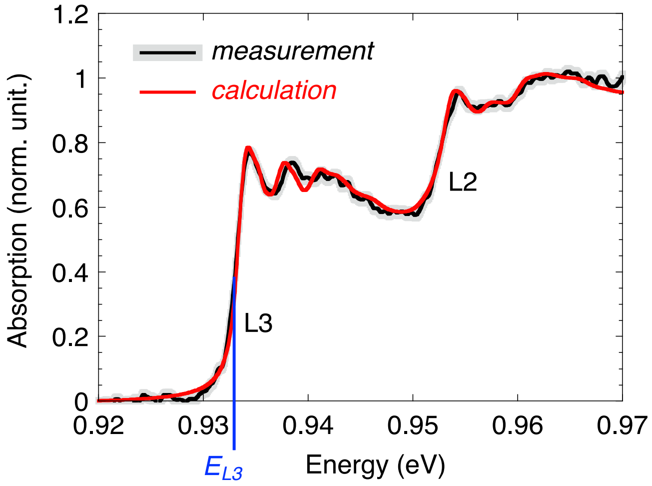
X-ray Absorption: Principles, Applications, Techniques of […] XANES (Wiley, New York, 1988)

L3-edge XANES calculations in warm dense copper *(1/3)*

- XANES spectra near the L3-edge probe 3*d*-states from $2p_{3/2}$ *(selection rule)*
  - *Ab initio* DFT + linear response theory to calculate X-ray absorption
  - Very good agreement in cold solid copper calculation vs experiment *(fcc)*







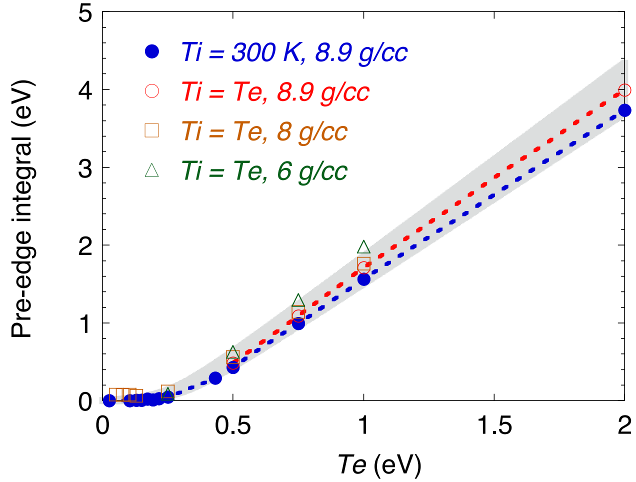
⇒ Interpretation of the different spectral features in terms of electron and atomic structures

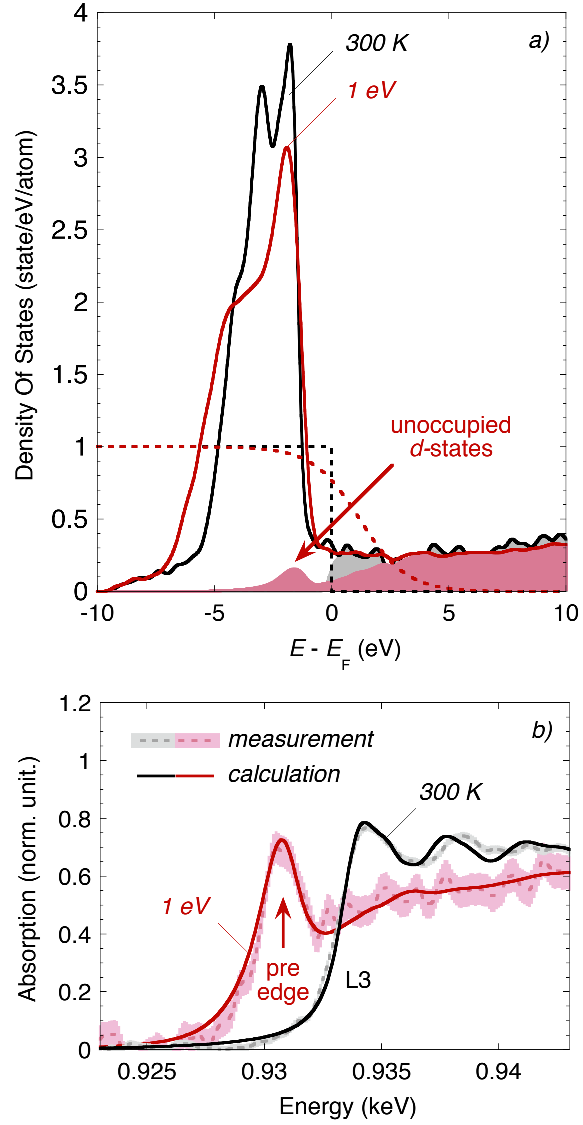
N. Jourdain et al., *Phys. Rev. B* ***101****, 125127 (2020)*

F. Dorchies and V. Recoules, Physics Reports ***657****, 1 (2016)*

L3-edge XANES calculations in warm dense copper *(2/3)*

- Observation of a pre-edge when $T_e \geq 0.25$ eV
  - Some *3d* electrons are promoted above the Fermi energy
    - ✓ Similar trend at thermal equilibrium or non-equilibrium
    - ✓ From solid density down to expanded liquid one





⇒ Fair diagnostic of the electron temperature T_e

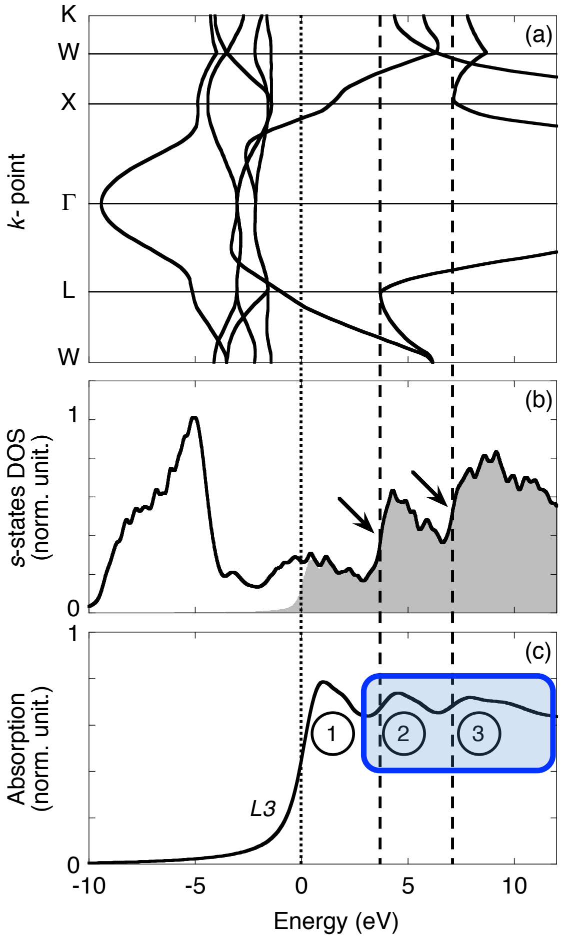
⇒ Previously observed in the B. I. Cho *et al.* experiment *

** B. I. Cho* et al.*, Phys. Rev. Lett.* ***106****, 167601 (2011)*
N. Jourdain et al.*, Phys. Rev. B* ***101****, 125127 (2020)*

L3-edge XANES calculations in warm dense copper *(3/3)*

- Observation of post-edge modulations
  - Van Hove singularities in the electron Density of State
    - ✓ Results from the fcc electronic band structure *(dE/dk = 0)*
    - ✓ Associated characteristic peaks in the XANES spectra
  - These modulations are sensitive to the lattice periodicity
  - They disappear in the liquid phase

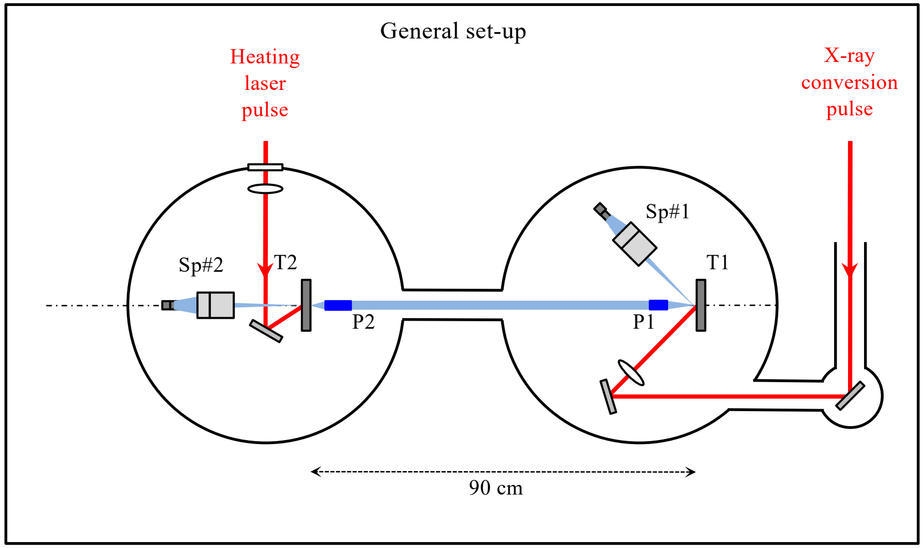
⇒ Diagnostic of the crystalline structure



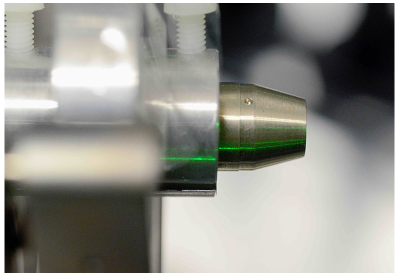
N. Jourdain et al., *Phys. Rev. B* ***101****, 125127 (2020)*

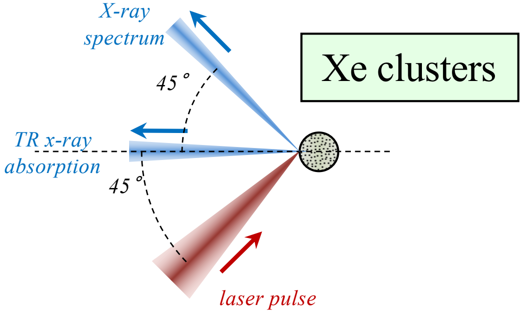
Picosecond-time-resolved XANES measurements *(1/2)*

- Time-resolved XANES station *(CELIA)*
  - Ti:Sa laser, 100 mJ focused on target *(T1)* => "thermal" laser-plasma-based X-ray source
    - ✓ Laser duration adjustable from ≤ 100 fs to ~ 10 ps
    - ✓ 10 Hz laser, but effective repetition rate of 1 Hz *(motions of target T1 and sample T2)*



X-ray transport through polycapillaries





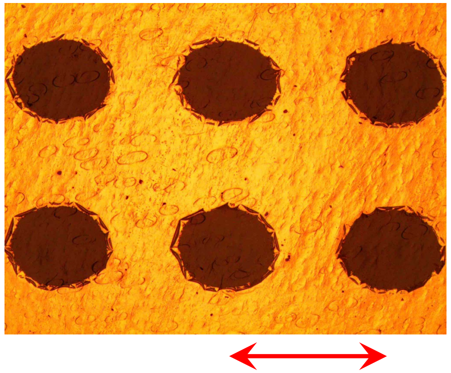
⇒ 1.2 ± 0.2 ps rms time resolution *(from following measurements)*

F. Dorchies et al., *Rev. Sci. Instrum.* ***86****, 073106 (2015)*

Picosecond-time-resolved XANES measurements *(2/2)*

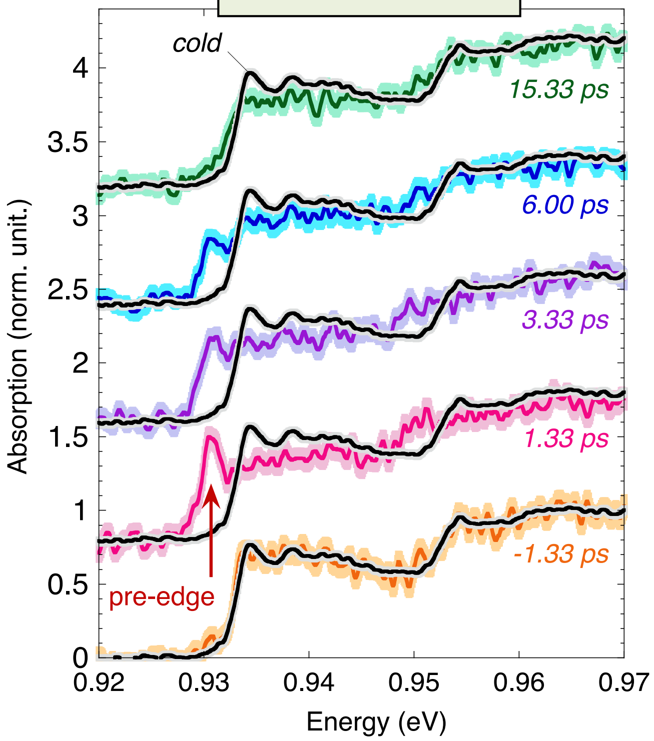
- Pump *(optical laser)* – probe *(X-ray)* experiment
  - About 300 shots to build each absorption spectrum *(~ 5 mn)*
  - Observation and time-resolution of the pre-edge => $T_e(t)$

80 nm
Cu sample



~ 1 mm

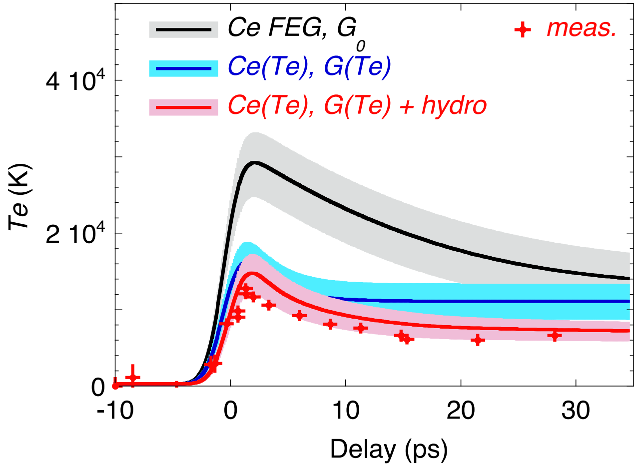
0.4 ± 0.1 J/cm^2

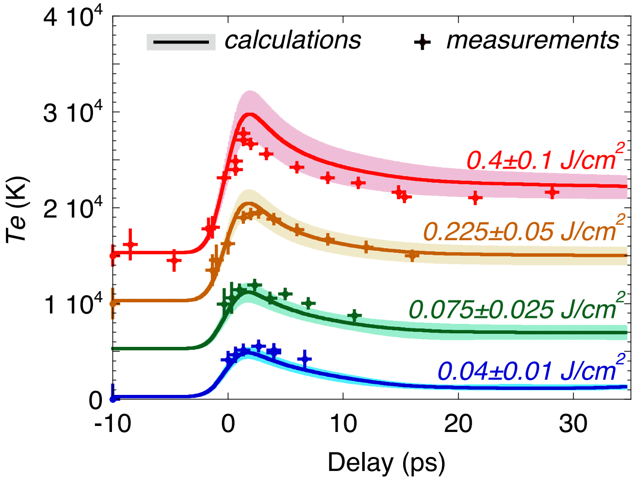


N. Jourdain et al., *Phys. Rev. B* ***97****, 075148 (2018)*

Electron-ion thermal equilibration dynamics

- Electron-ion thermal equilibration over ~ 10 ps
  - Well reproduced by Two-Temperature Model if:
    - ✓ T_e-dependent coefficients are considered *
    - ✓ Hydrodynamic expansion is considered





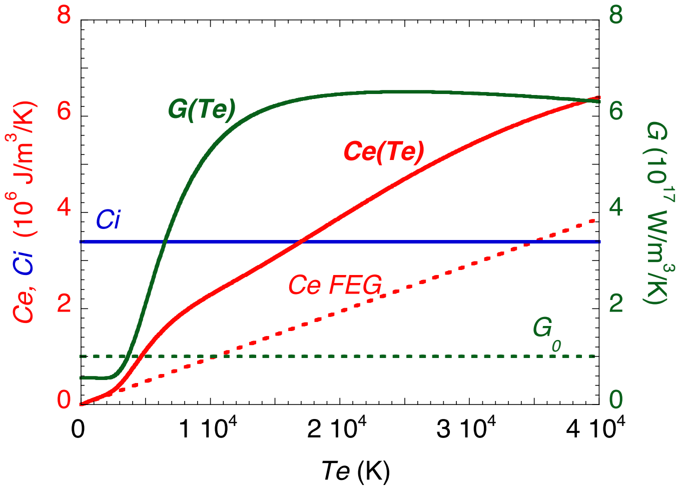
TTM calculations are convoluted by experimental time-resolution

Two-temperature model

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

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

⇒ Validated up to ~ 2 eV

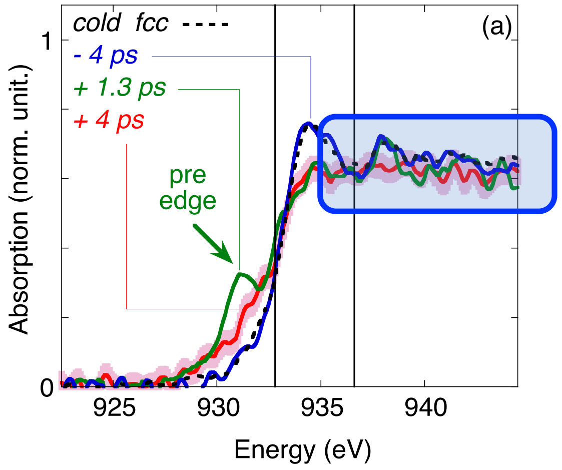


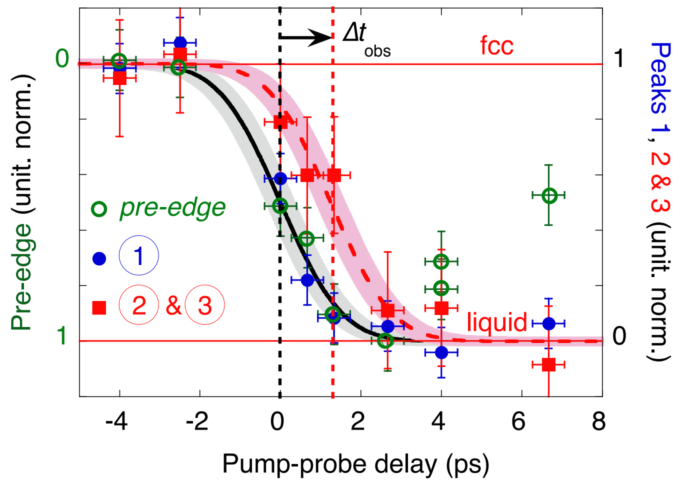
N. Jourdain et al., *Phys. Rev. B* ***97****, 075148 (2018)*

* *Z. Lin* et al., *Phys. Rev. B 77, 075133 (2008)*

Solid-liquid transition dynamics *(1/2)*

- About 2000 shots are required to resolve post-edge modulations *(~ 30 mn)*
  - A delay is observed between:
    - ✓ pre-edge increase *(electron heating* T_e*)*
    - ✓ and modulations decrease *(melting)*



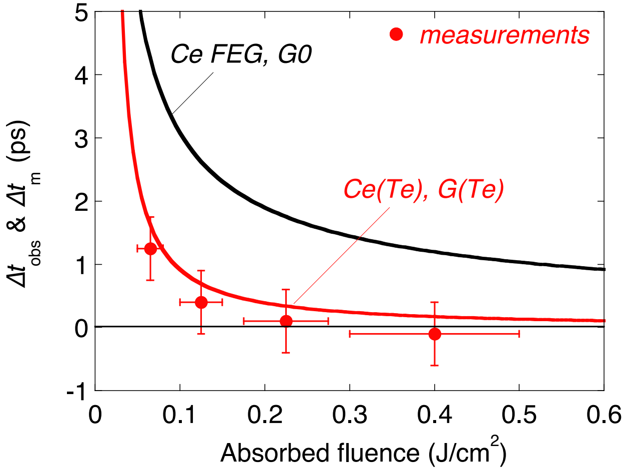


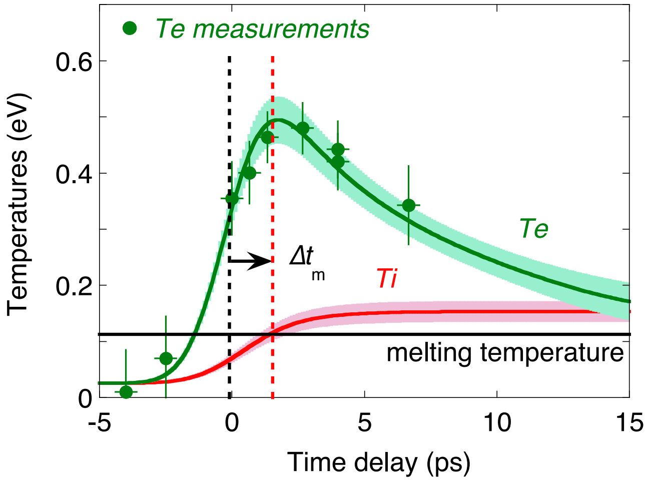
⇒ Melting time measurement

N. Jourdain et al., *Phys. Rev. Lett.* ***126****, 065001 (2021)*

Solid-liquid transition dynamics *(2/2)*

- Liquid phase observed in ~ 1 ps, compatible with a thermal melting
  - TTM independently controlled with $T_e(t)$
    - ✓ In good agreement with theory *(no bond hardening is expected in this fluence regime)*
    - ✓ In contradiction with electron diffraction measurements *





⇒ Bond hardening evidence needs for higher level of heating and fs time-resolution

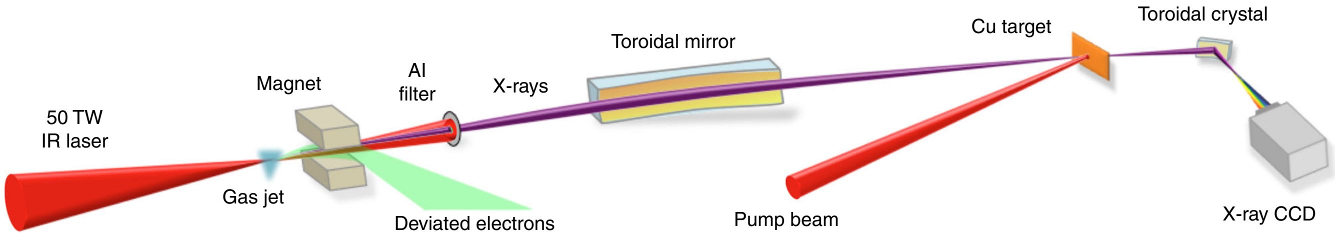
N. Jourdain et al., *Phys. Rev. Lett.* ***126***, *065001 (2021)*

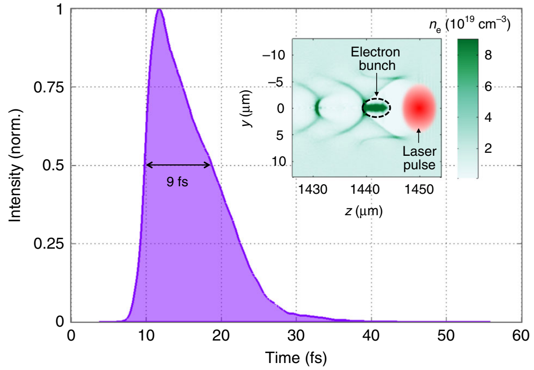
* *R. Ernstorfer* et al., *Science* ***323***, *5917 (2009)*
* *M. Z. Mo* et al., *Science* ***360***, *1451 (2018)*

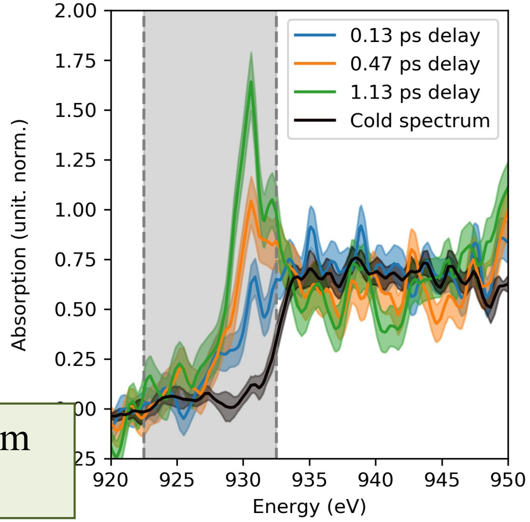
Demonstration of femtosecond XANES

See Kim Ta Phuoc's talk

- Betatron beamline at LOA
  - Ideal characteristics for femtosecond XANES *(broadband, ~ 10 fs duration)*







⇒ 75 ± 25 fs rms time resolution

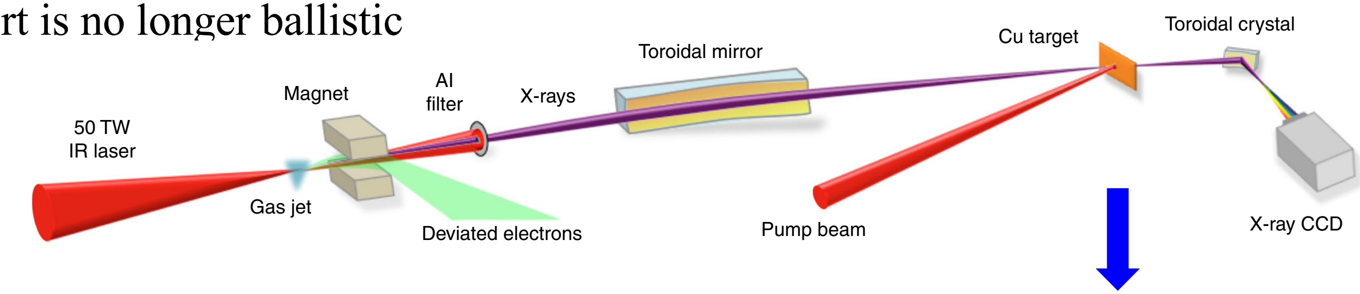
~ 100 shots (~ 15 min) / spectrum
~ 10 % noise on transmission

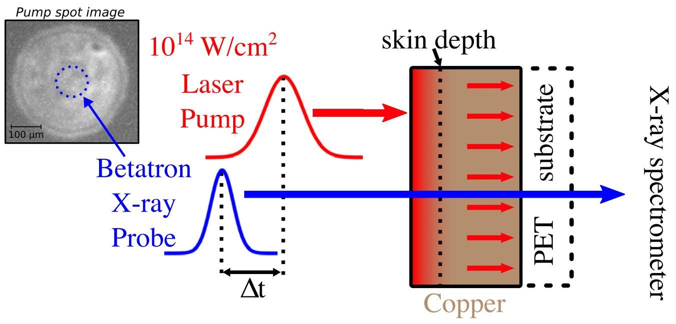
A. Rousse et al., *Phys. Rev. Lett.* ***93****, 13 (2004)*

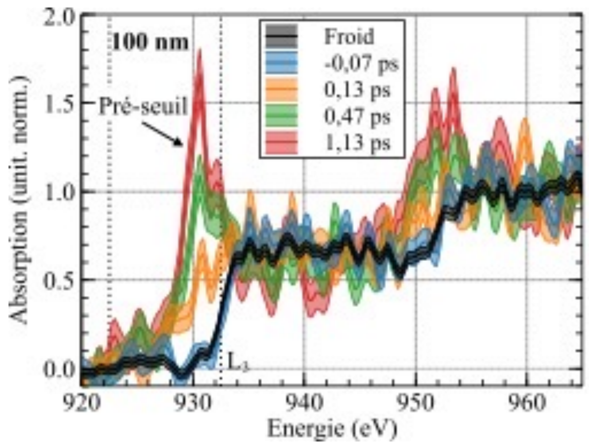
B. Mahieu et al., *Nature Communications* ***9****, 3276 (2018)*

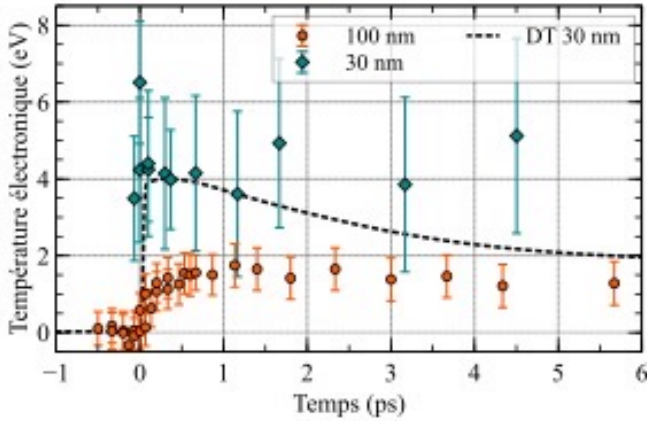
Investigation of the electron transport dynamics *(1/2)*

- At high flux, the T_e rise time is slowed down $(2.5x10^{13} > Q_{sat} = 1.5x10^{13}\ W/cm^2)$
  - Within the fist picosecond
  - ⇒ Electron transport is no longer ballistic









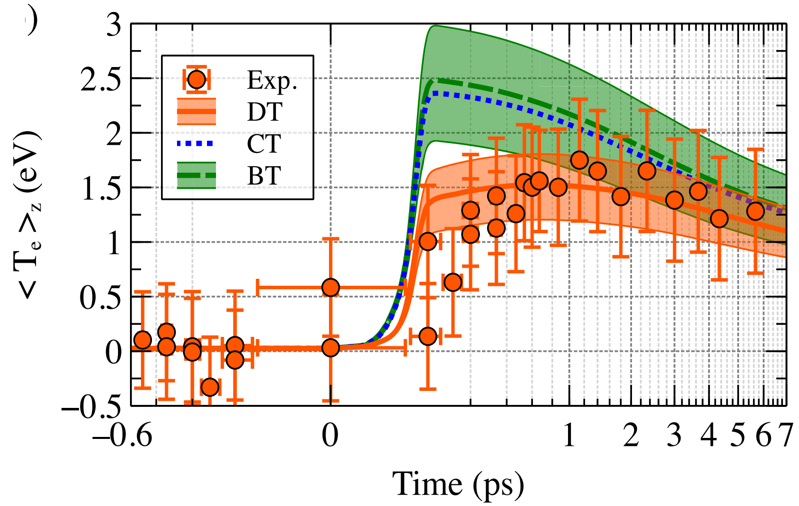
A. Grolleau et al., *Phys. Rev. Lett.* ***127****, 275901 (2021)*

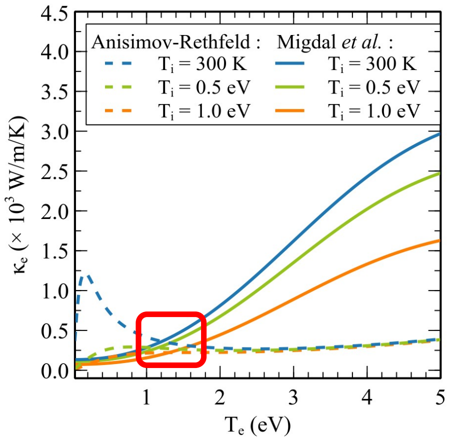
Investigation of the electron transport dynamics *(2/2)*

- Data are best reproduced with pure Diffusive electron Transport
  - DT = Diffusive Transport
  - BT = Ballistic Transport
  - CT = Composite Transport
    - ✓ Saturated Ballistic *(60 %)*
    - ✓ Then Diffusive *(40 %)*

⇒ Suggests a reduction of the electron mean free path

⇒ A tool to test κ_e models





Two-temperature model

$$C_e \frac{dT_e}{dt} = -G_{e-i}(T_e - T_i) + \nabla(\boxed{\kappa_e}\nabla T_e) + S(t)$$
$$C_i \frac{dT_i}{dt} = G_{e-i}(T_e - T_i) + \nabla(\kappa_i \nabla T_i)$$

A. Grolleau et al., *Phys. Rev. Lett.* ***127****, 275901 (2021)*

Conclusion

- Time-resolved X-ray measurements of non-equilibrium WDM
  - Non-equilibrium situations shed light on the electron – ion dynamic interplay
  - X-ray diagnostics test DFT simulations at their suitable atomic scale
  - XANES probes electron and atomic structures

- Time-resolved XANES measurements on warm dense copper *(ps down to fs)*
  - Electron-ion thermal equilibration *(~ 10 ps)*
  - "Thermal" melting observed *(~ 1 ps)*
  - Diffusive electron transport dominant at higher flux *(~ 1 ps over 100 nm)*

⇒ Two-Temperature Model successfully tested, coupled to hydro code ESTHER *(≤ 3 eV)*

Perspectives

Cu

- Bond hardening on copper ?
  - Requires higher flux, then thin sample and fs resolution with high signal-to-noise ratio

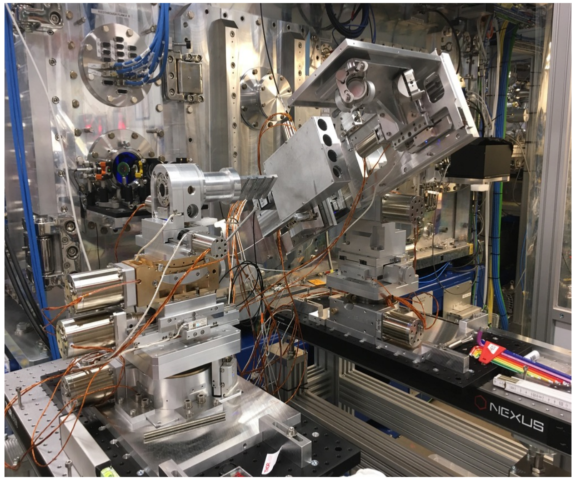
FemTraXS project

Mo

Si_3N_4

- Other materials
  - Molybdenum as a transition metal prototype => L3-edge *(2.52 keV)*
  - Silicon nitride *(insulator)* => K-edge *(1.84 keV)*
  - ⇒ Require specific studies to interpret the XANES patterns in the WDM regime

- Other facilities *(XANES demonstrated on XFEL)*
  - X-FEL provides fs resolution and high X-ray flux
  - Dedicated XANES device implemented on Eu-XFEL / HED
  - ⇒ Couple time-resolved XANES / XRD



J. Gaudin et al., *Scientific Report **4** 4724 (2014)*
M. Harmand et al., *Phys. Rev. B **92**, 024108 (2015)*
F. Dorchies et al., *Phys. Rev. B **92**, 144201 (2015)*

Thank you for your attention

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