

**Identification of Auger electron heating  
irradiating solids with micrometer-focused XUV  
Free Electron Laser Radiation from FLASH  
at intensities larger than  $10^{16}$  W/cm<sup>2</sup>**

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# International Collaboration « Peak Brightness »

S.Bajt, L.F. Cao, J. Chalupsky, H. Chapman, J. Cihelka, H.-K. Chung, T. Döppner, S. Dusterer, T. Dzelzainis, M. Fajardo, C. Fortmann, E. Galtier, S.H. Glenzer, G. Gregori, P.V. Hajkova, P.A. Heimann, L. Juha, M. Jurek, R. Fäustlin, F. Khattak, M. Kozlova, J. Krzywinski, T. Laarmann, H.J. Lee, R. W. Lee, R. Levesque, P. Mercere, B. Nagler, A. Nelson, A. Przystawik, P. Radcliffe, H. Reinholz, D. Riley, G. Ropke, F.B. Rosmej, J. K. Seksl, R. Sobierajski, R. Thiele, T. Tiggesbaumker, S. Toleikis, N.X. Truong, T. Tschentscher, I. Uschmann, S. Vinko, J. Wark, T. Witcher, A. Wierling, E. Forster, R. Redmer, U. Zastra

**DESY (FLASH), UPMC, LULI, LLNL, SLAC, Berkeley, QUB, Oxford, Jena, Rostock.....**

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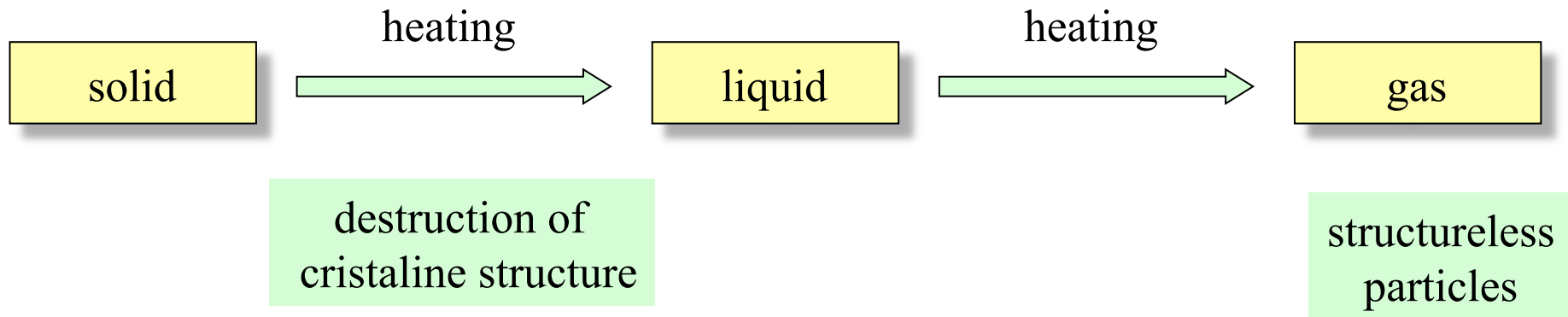
**II. Matter under extreme conditions: atomic physics**

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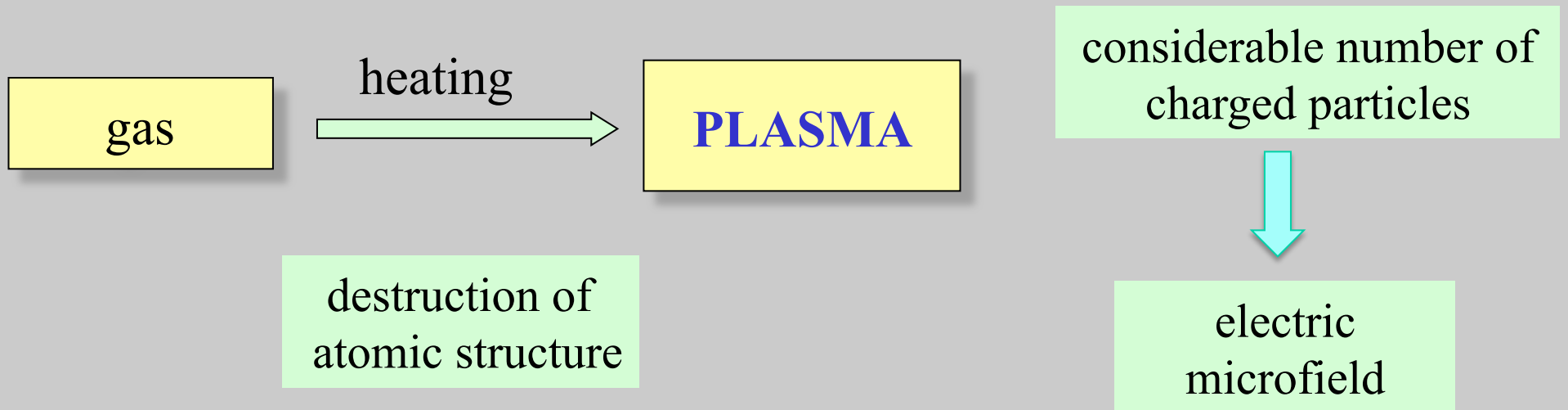
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# I. Introduction & Motivation

# Matter and Energy



*phase transitions = key physics*



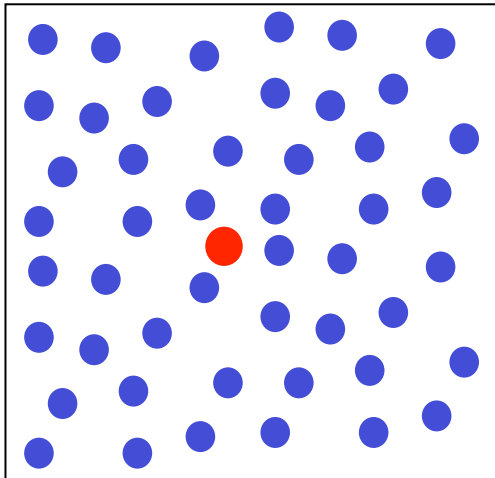
# Matter under extreme conditions

PLASMA

high pressure

Warm Dense Matter  
WDM:  $T \approx T_F$

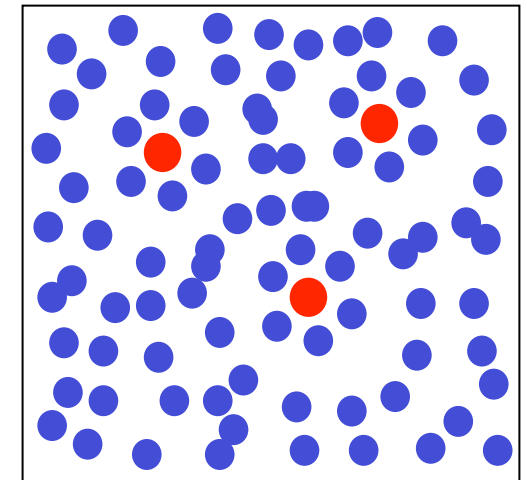
return of a certain cristaline structure



$$\frac{V_{Coulomb}}{V_{thermal}} = \Gamma \propto \frac{1}{rT} \propto \frac{\rho^{1/3}}{T}$$

decrease of temperature T

increase of density n



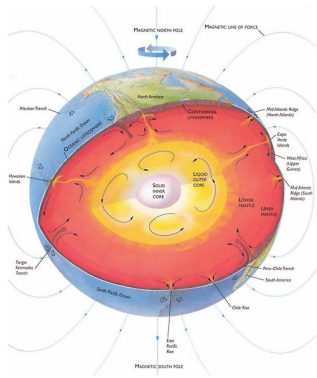
Partition function ?

Interesting experiment to encounter high  $\Gamma$

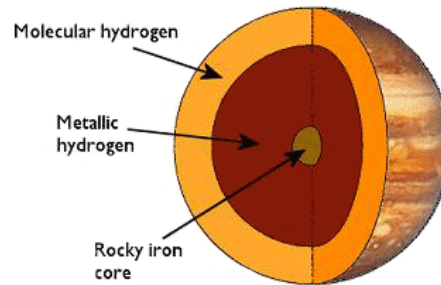
# Importance of Warm Dense Matter

## I. In the univers

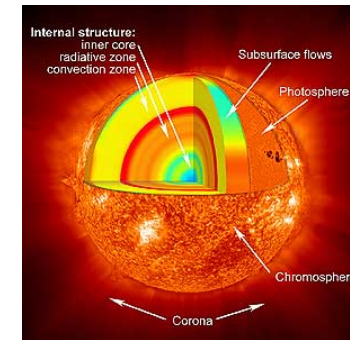
Earth:  
magnetic field



Jupiter:  
metallic hydrogen

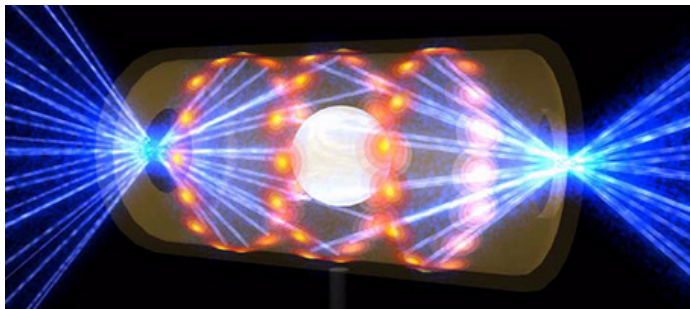


Sun:  
thermonuclear fusion

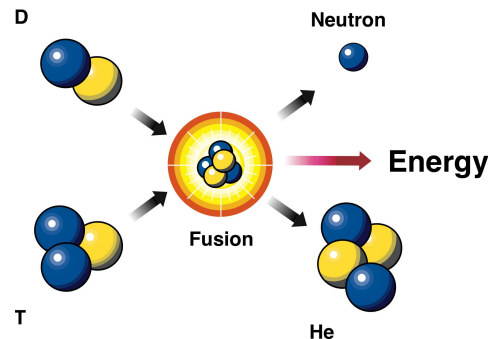


## II. Heating of matter

Laser/Heavy ion beams



Inertial fusion



Electric heating



## **II. Matter under extreme conditions:**

**-Atomic physics studies-**



# Atomic physics and derived quantities

Atomic Structure

Particle statistics

Atomic kinetics

## Atomic physics in dense plasmas

Atoms/Ions/Molecule/  
Cluster/....

Distribution of different  
ionisation stages

Distribution of ground  
and excited states

Emission

Scattering

Absorption/  
Opacity

Transmission

Conductivity

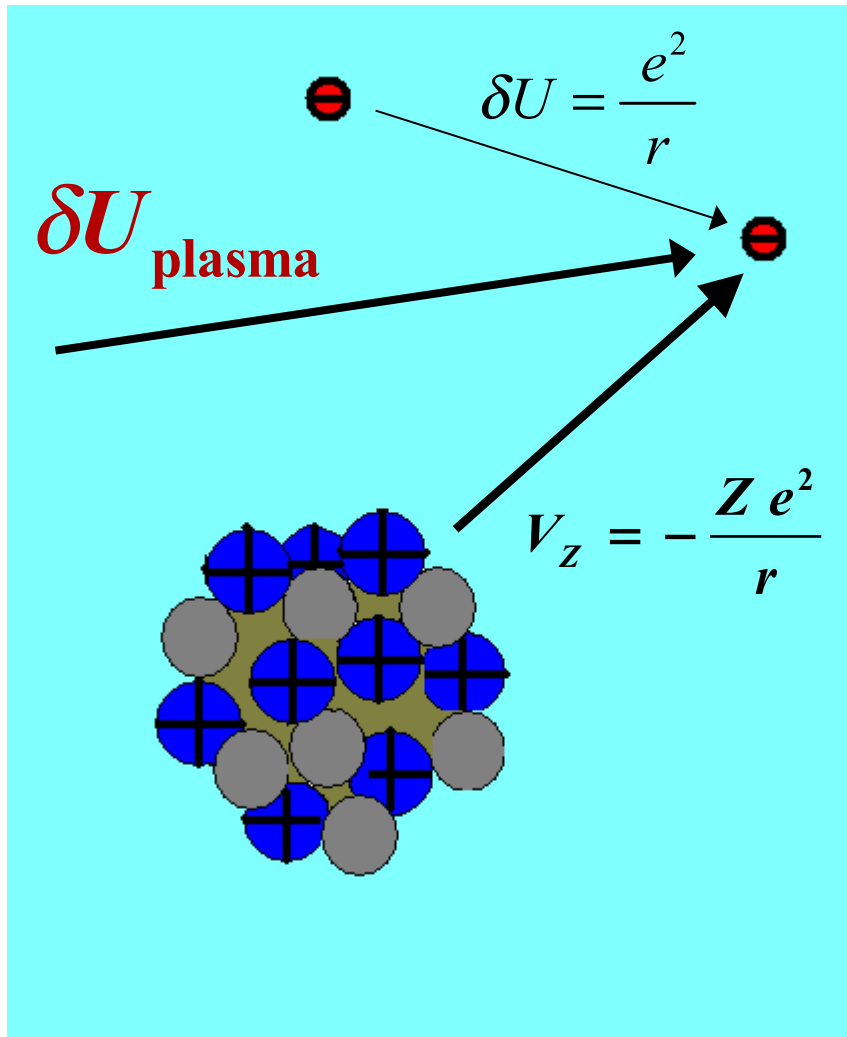
Compressibility

Phase transitions

•  
•  
•

Equation of state

# Atomic physics in dense plasmas



$$(L_r - \delta U_{\text{Plasma}} + E)(R^0 + \delta R) = 0$$

$$\delta R_{nl}(r) = \int_0^\infty dr' r'^2 g_{nl}(r, r') \delta U_{\text{Plasma}}(r') R_{nl}^0(r')$$

*General expression  
for arbitrary plasma potential*

F.B. Rosmej, K. Bennadji, V.S. Lisitsa, PRA 2011

- Arbitrary temperature
- Scaling laws
- Comparison with experiments



# LULI: e-e exchange energy shift in dense plasmas

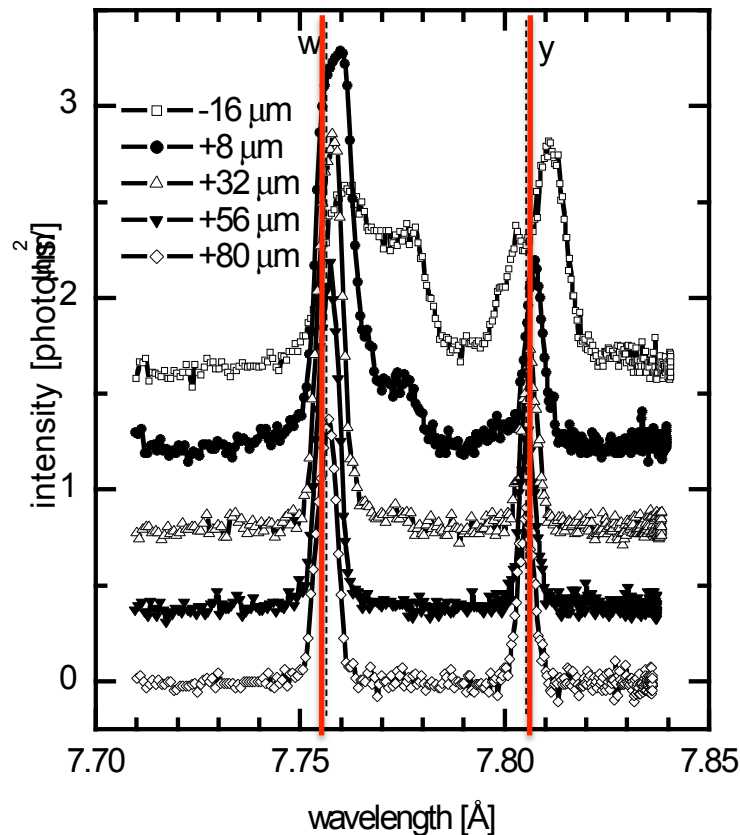
There exist one pair of suitable transitions in He-like ions:

Resonance line:  $1s^2 \ ^1S_0 - 1s2p \ ^1P_1$

Intercombination line:  $1s^2 \ ^1S_0 - 1s2p \ ^3P_1$

“A spin in a  
dense plasma”

$$\Delta E_x = (E(^1P_1) - E(^3P_1))_{vacuum} - (E(^1P_1) - E(^3P_1))_{plasma}$$



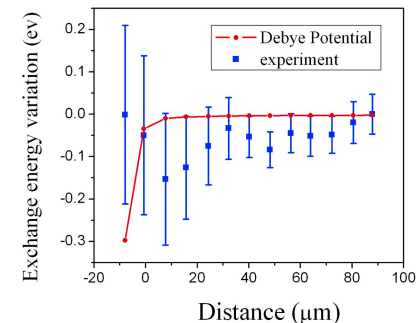
O. Renner, P. Adámek, P. Angelo, E. Dalimier, E. Förster, E. Krousky, F.B. Rosmej, R. Schott, JQSRT **99**, 523 (2006).

Finite temperature ion sphere model:

$$\Delta E_x \propto n_e \left( \frac{23}{Z^3} + \frac{11.6}{Z^2 \sqrt{T(eV)/2Ry}} \right)$$

Debye model:

$$\Delta E_x \propto \frac{n_e Z_{eff}}{ZT}$$



F.B. Rosmej, K. Bennadji (EMMI-fellow),  
V.S. Lisitsa, PRA 2011

Exp. Trend  $\Delta E_x(^1P_1) > \Delta E_x(^3P_1)$  ok F.B. Rosmej/UPMC/LULI

# Opacity measurements

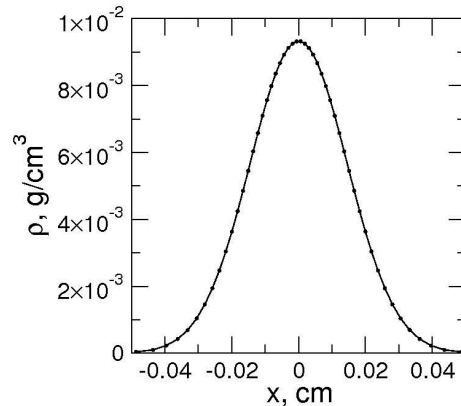
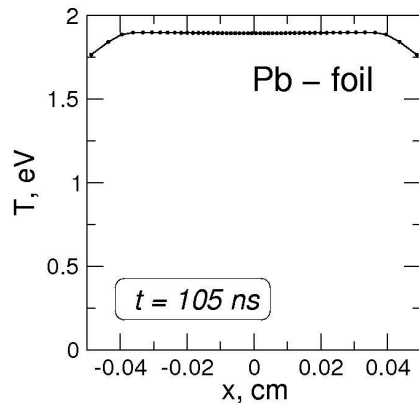
If  $T$  is too small for self-emission studies:  $\frac{V_{Coulomb}}{V_{thermal}} = \Gamma \propto \frac{1}{rT} \propto \frac{\rho^{1/3}}{T}$



Opacity measurements  
with heavy ion beams:

homogenous,  
macroscopic samples,  
known energy deposition

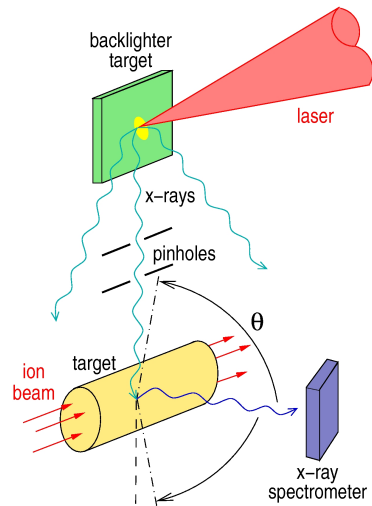
*Talk: Andreas Tauschwitz*



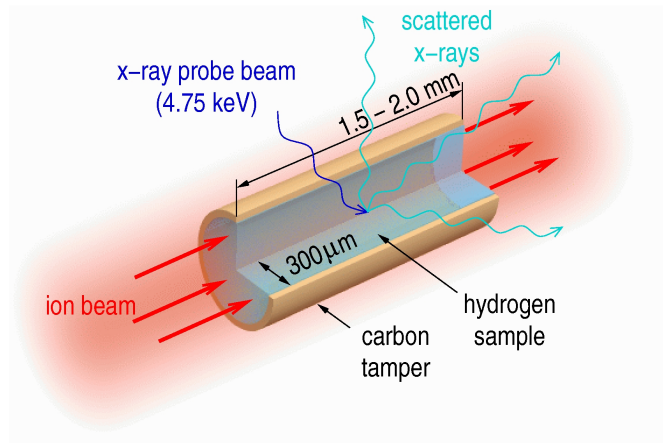
An. Tauschwitz, V.G. Novikov, A. Tauschwitz, F.B. Rosmej, J. Abdallah, E. Onkels, J. Jacoby, J.A. Maruhn: “Intense ion beams as a tool for opacity measurements in warm dense matter”, Rapid Comm. Appl. Physics B **95**, 13 (2009).

Absorption spectra – versus – At. Phys. simulations

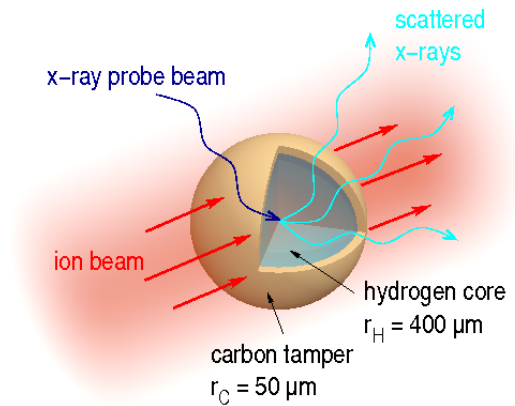
# WDM created that it can be **well** diagnosed



D. Riley & F.B. Rosmej 2003,  
GSI Annual report



An. Tauschwitz et al.,  
PRE **68**, 056406 (2003)



An. Tauschwitz et al.,  
HEDP **3**, 371 (2007)

*Talk: Paul Neumeyer – Backlighter/Scatter source developments*

# WDM

« Radiative properties of Warm Dense Matter  
produced by intense heavy ion beams »

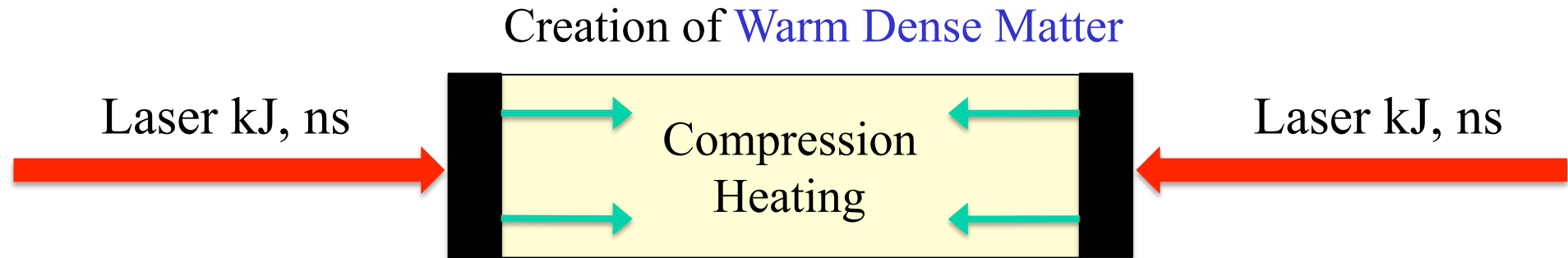
[www.gsi.de/forschung/phelix/Experiments/FAIR/WDM/index.html](http://www.gsi.de/forschung/phelix/Experiments/FAIR/WDM/index.html)

F.B. Rosmej/UPMC/LULI

# **III. Free Electron Lasers - FLASH**

**-Atomic physics studies-**

# Drawbacks of standard experimental approaches



**X-ray radiation** (diagnostic) necessary to **escape solid matter**,  
but creation impossible at small temperature



Creation of X-rays  $\Rightarrow$  high temperature

$$kT \approx E_X \approx keV$$

$\Rightarrow V_{\text{coulomb}} \ll V_{\text{therm}} \Rightarrow \Gamma \ll 1$ : the interesting regime is lost

ns-time scale: the direct access of transitions of phases is difficult



# Free Electron X-ray Lasers - XFEL



..... creation of K-, L-hole states



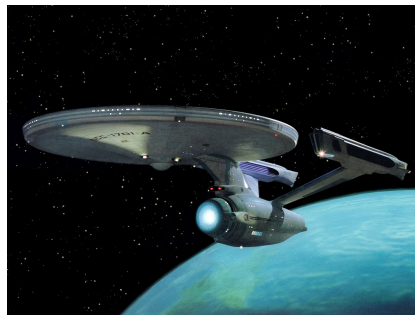
No temperature needed to excite X-rays !

*Synchrotron radiation* creates K-holes but does not heat the solid because of very low photon intensity ( $10^{12}$  photons/sec)

*XUV/X-ray Free Electron Lasers* have a brilliance about **10 orders** of magnitude higher to allow **photoionisation and heating**



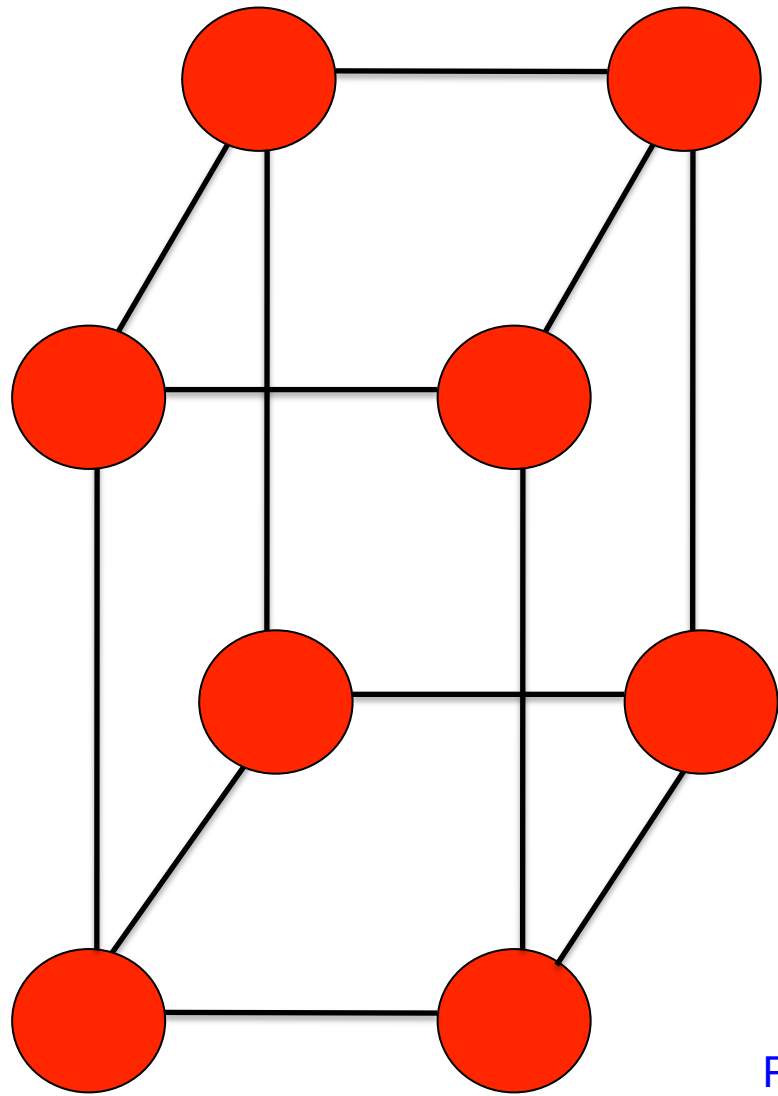
*10 orders of magnitude in velocity*



*Not just more quick ...but completely different*



# What is so different ?



**XFEL**



photoionisation  
of internal shells

**crystal**

10 fs

**hollow**

$I > 10^{16}$  W/cm<sup>2</sup>

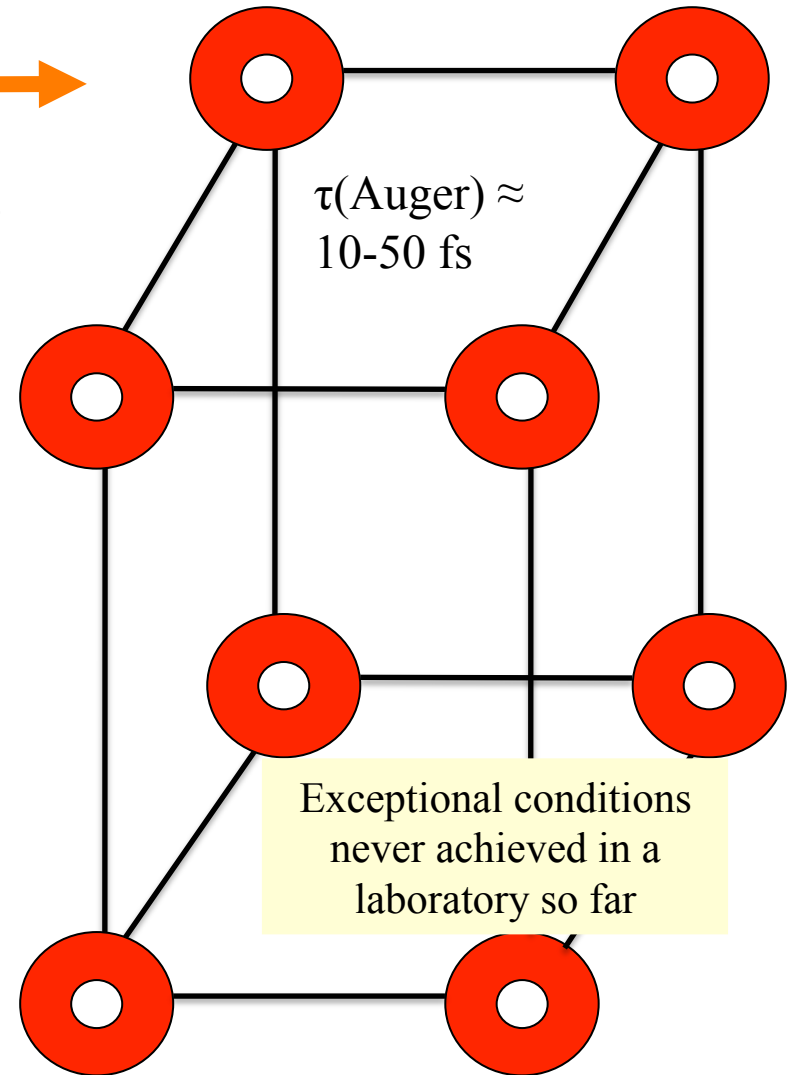
**Exotic  
Matter**

Conduction band

heating (Auger)

Phase transitions

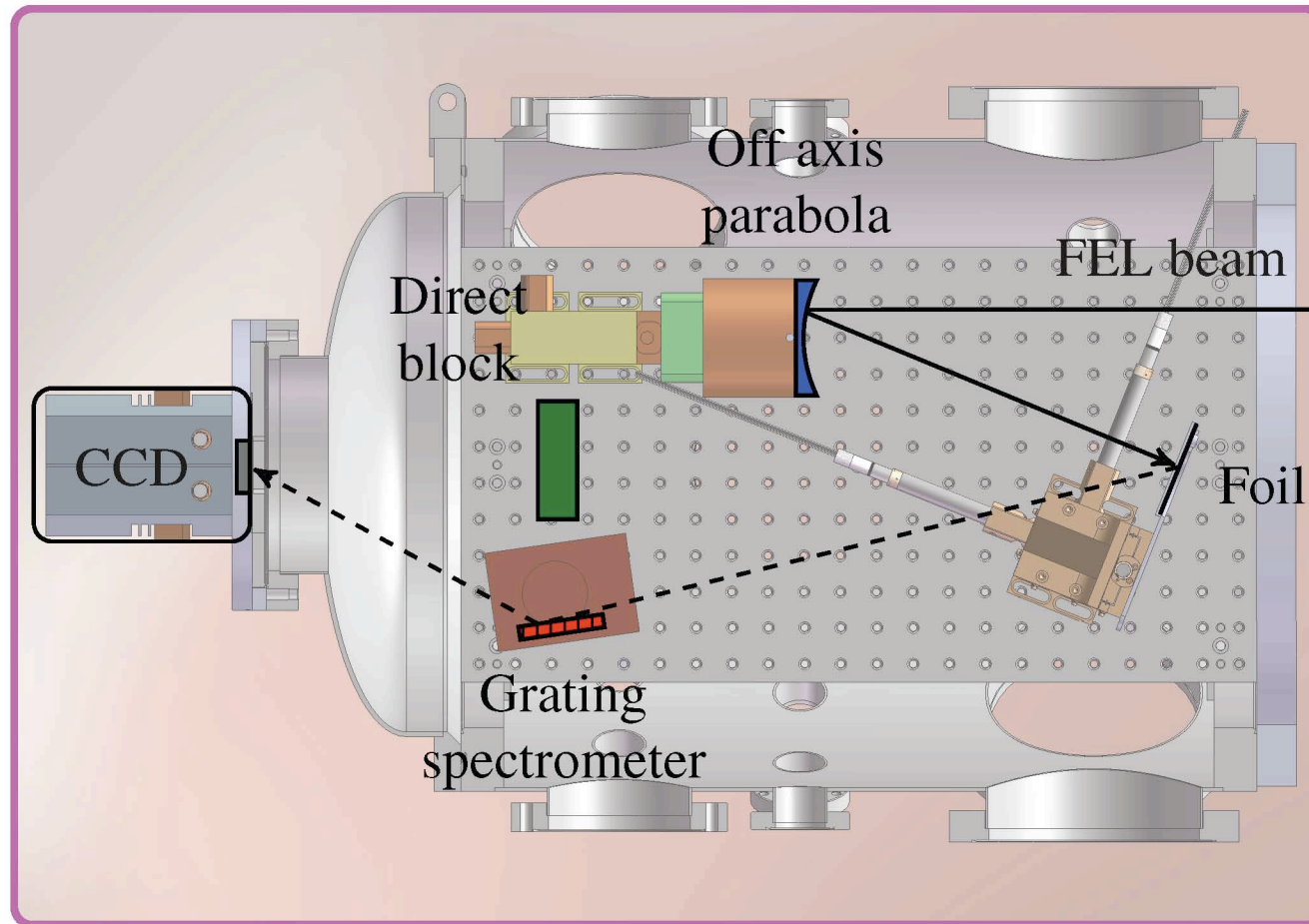
X-ray emission from exotic matter



$\tau(\text{Auger}) \approx$   
10-50 fs

Exceptional conditions  
never achieved in a  
laboratory so far

# Experimental setup



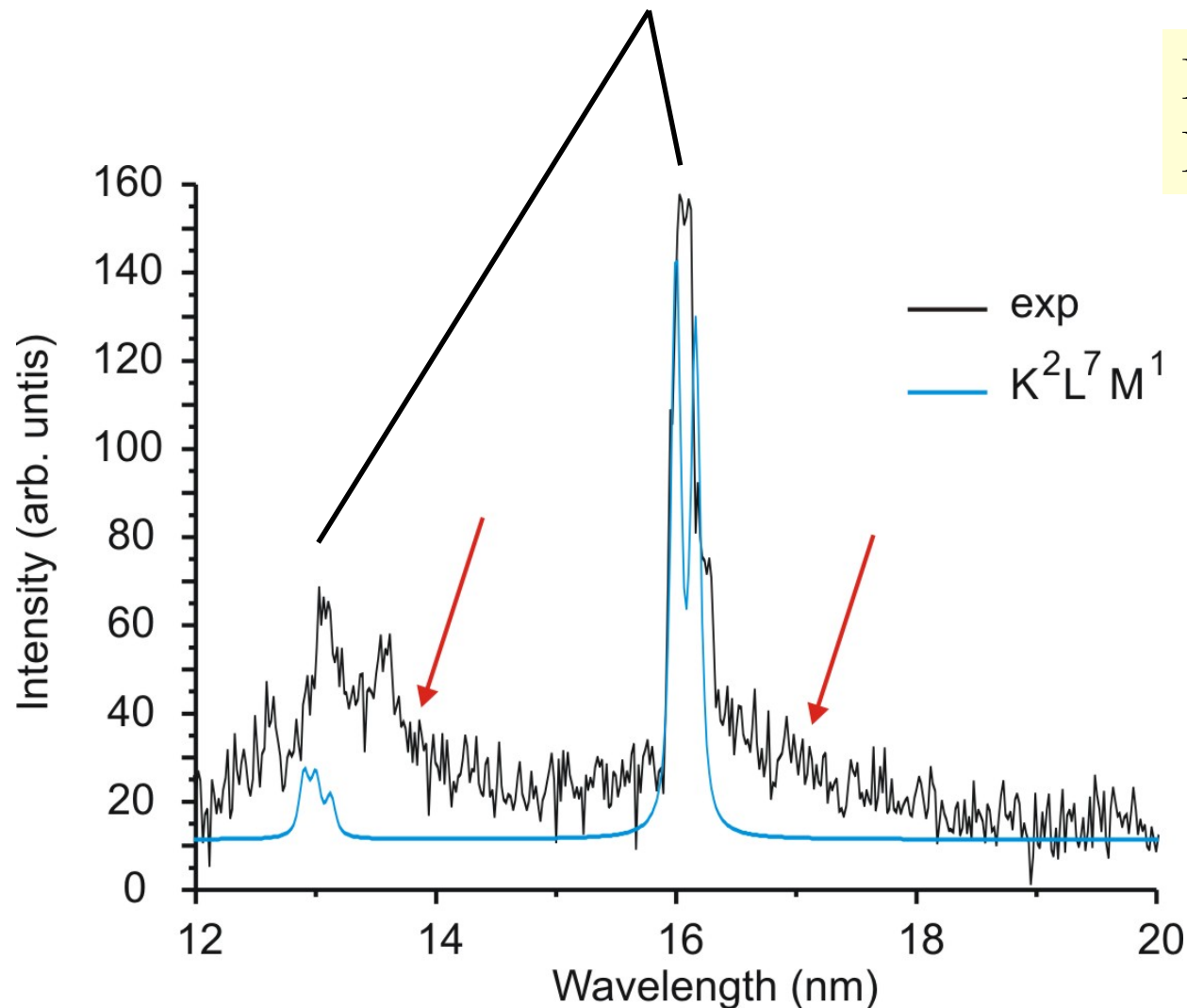
$\mu\text{m}$ -focussing:  $\phi < 1 \mu\text{m}$   
Intensities  $> 10^{16} \text{ W/cm}^2$

$\lambda = 13.5 \text{ nm}$  (92 eV),  $\tau = 15 \text{ fs}$   
 $E = 50 \mu\text{J}$ ,  $N = 10^{12}$  photons/pulse

# Al-case: Resonance lines do not explain the observations

Dominant spectral features: Al IV

Line emission:  
 $K^2L^7M^1 \rightarrow K^2L^8 + h\nu_{IV}$



# What is missing ?

Resonance line emission (13-16 nm):

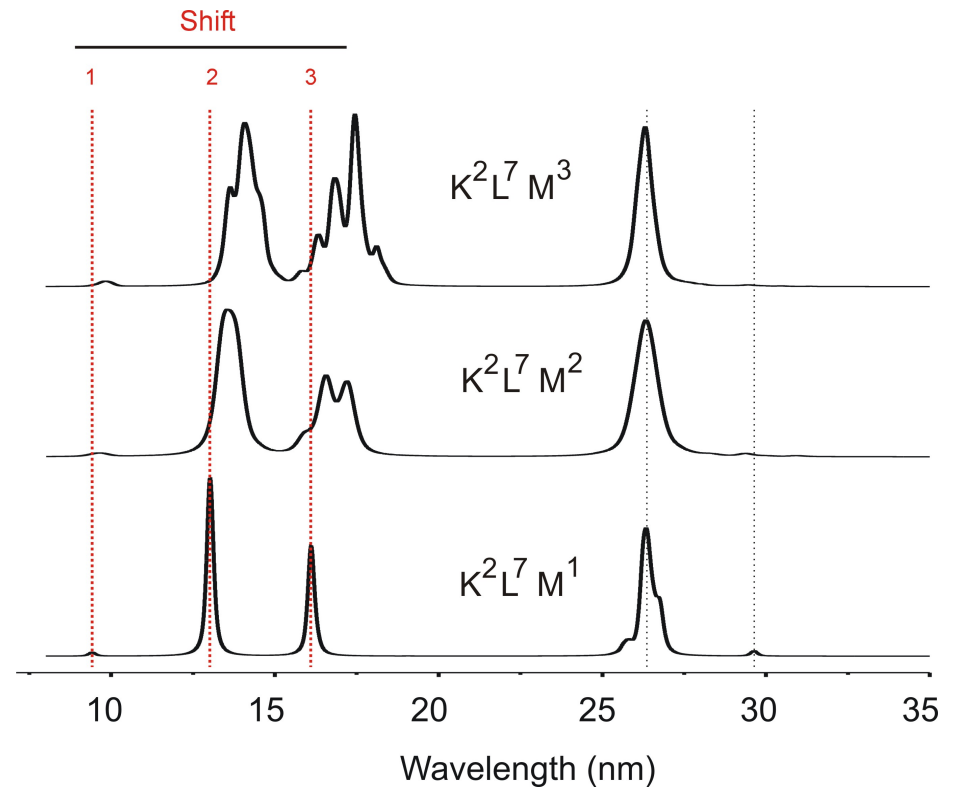
$$K^2L^7M^1 \rightarrow K^2L^8 + hc/\lambda$$

**Screened** resonance transitions:

$$K^2L^7M^2 \rightarrow K^2L^8M^1 + hc/(\lambda + \delta\lambda)$$

$$K^2L^7M^3 \rightarrow K^2L^8M^2 + hc/(\lambda + \delta\lambda')$$

**$\delta\lambda$  = red line wings**



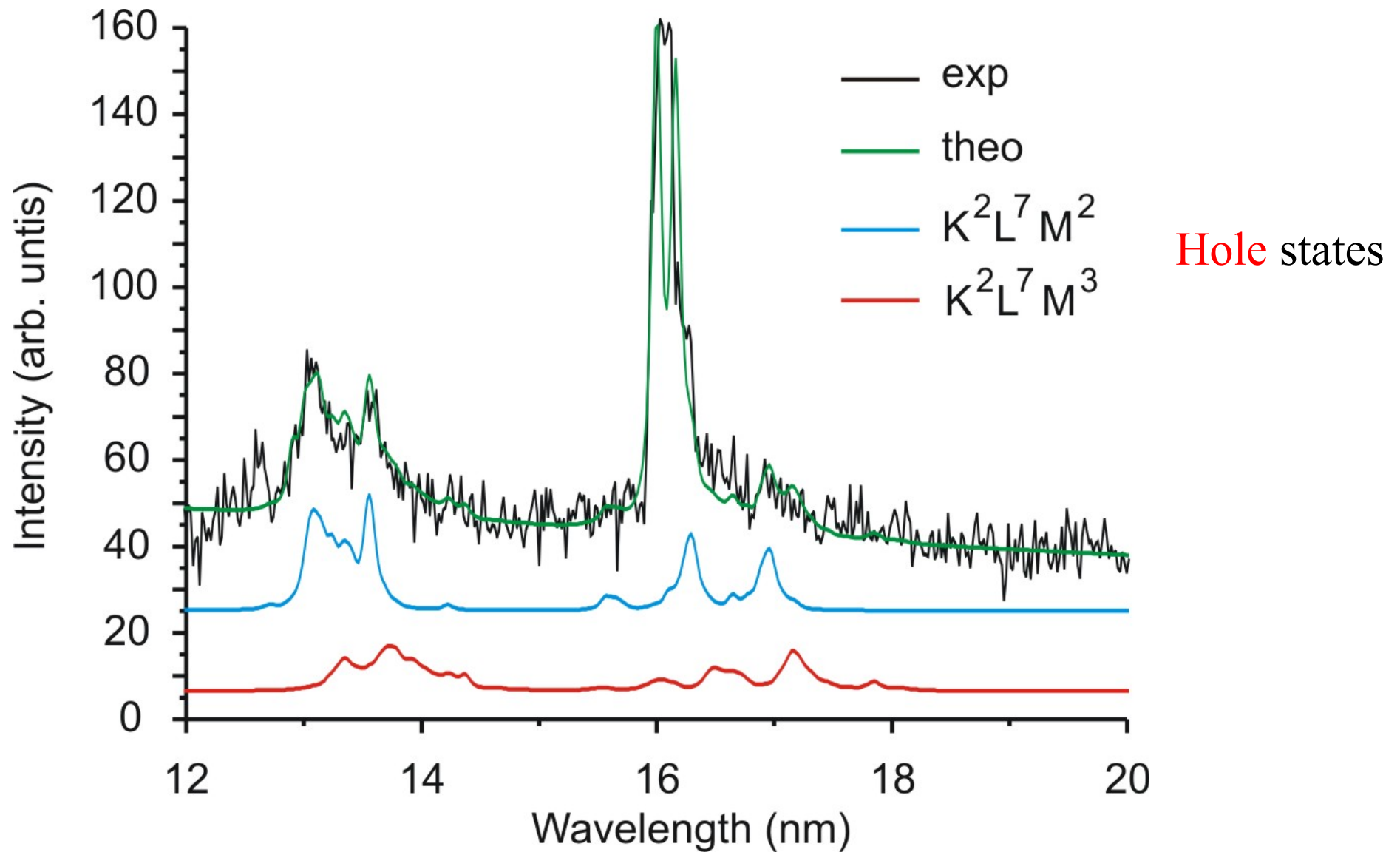
F.B. Rosmej et al., J. Phys. Conf. Series **244**, 042028 (2010)

**Screened** resonance transitions = Emission from hole states

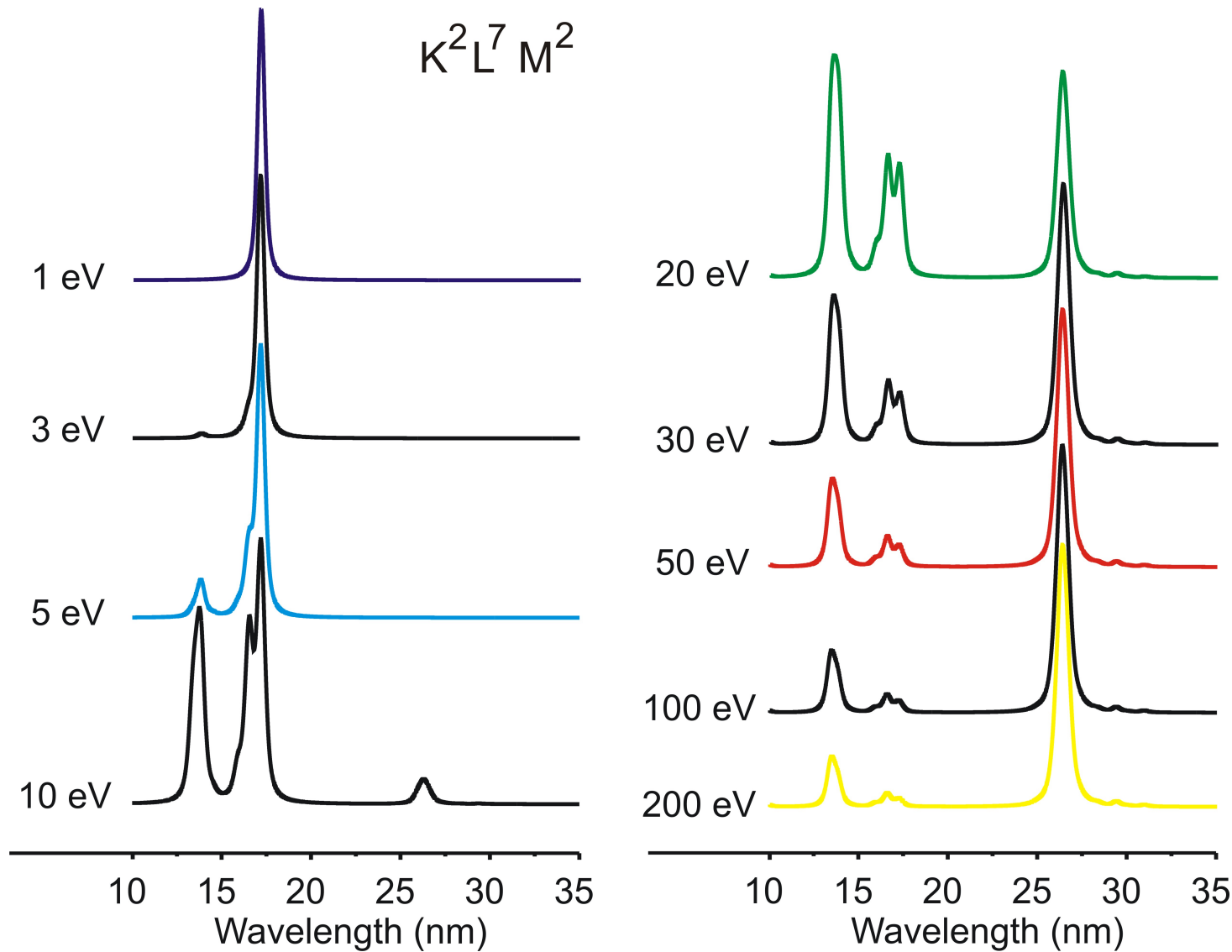
Hole states induced by photoioniation (FLASH: 92 eV):



# Inclusion of screened hole states matches the experiment



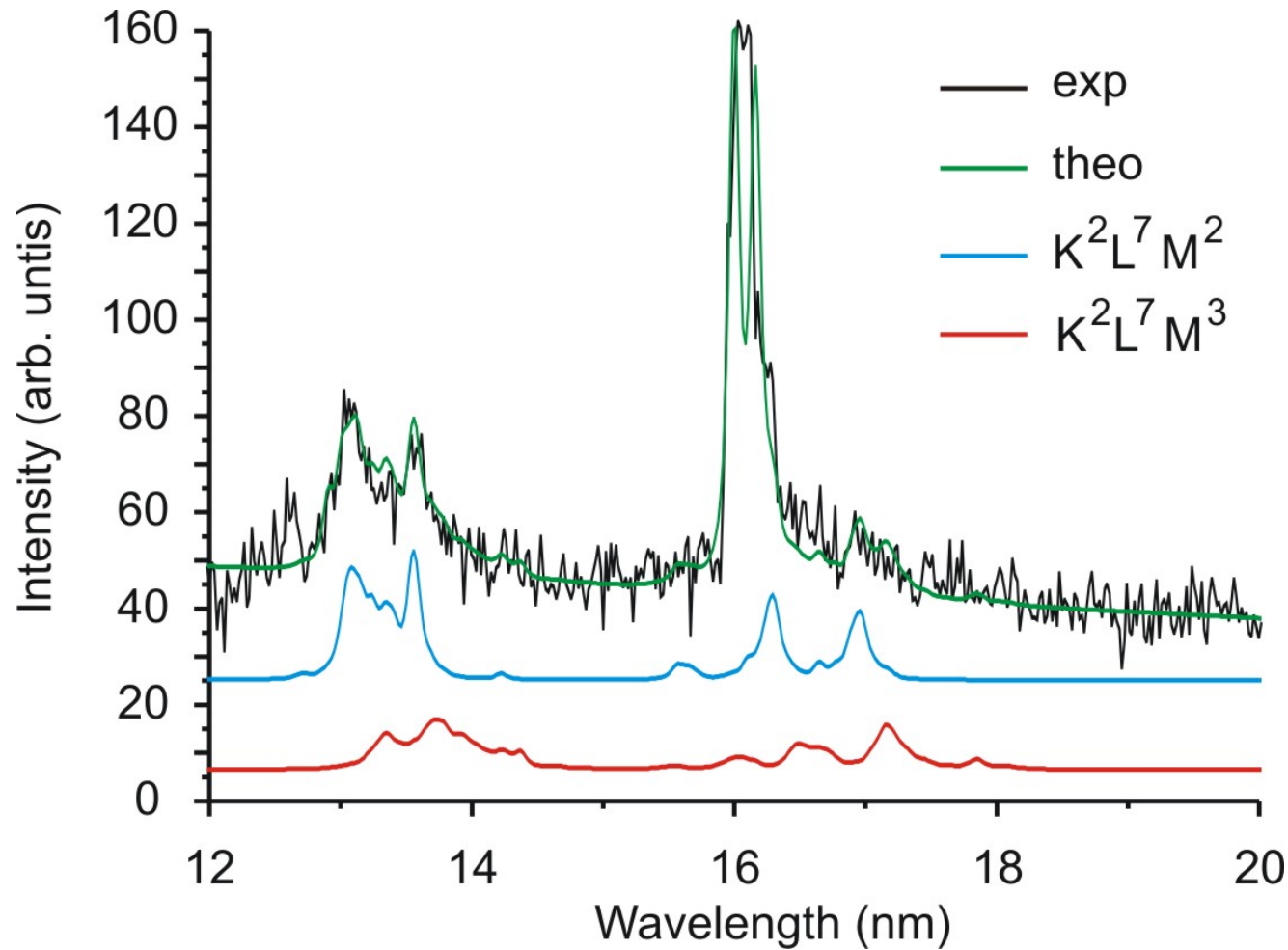
# Temperature dependence: $K^2L^7M^2$ -configurations



MARIA-simulations

\*MARIA-code (F.B. Rosmej):  
J. Phys. B Lett. **30**, L819 (1997)  
EPL **55**, 472 (2001);  
EPL **76**, 1081 (2006)

# Emission from hole states and temperature sensitivity



Hole states ( $K^2L^7M^2$ ,  $K^2L^7M^3$ )  $\rightarrow$   $kT_e \approx 25 \pm 10$  eV

## Do we understand a so high temperature ?



Distribution among 4 electrons in the M-shell:  $T \approx 8 \text{ eV}$

=> much lower than spectroscopic results (25 eV) !



### Relaxation of internal energy of « Hollow crystals »

Autoionization rate  $\approx$  Radiative decay rate  $\times (10^2-10^5)$

Relaxation by autoionisation:  $K^2L^7M^4 \rightarrow K^2L^8M^2 + e_{\text{Auger}}(70 \text{ eV})$

Equilibration in M-shell:  $2 \times 8\text{eV} + 1 \times 70 \text{ eV} \Rightarrow$  about 30 eV/electron



# Is the high $T = 30$ eV consistent with other observations ?

Principle of Microreversibility: inverse process must exist

**Time  
evolution !**

$$\text{Rate of inverse Auger effect} \propto \Gamma_{Auger} \frac{\exp(-E_{res} / kT_e)}{(kT_e)^{3/2}}$$

Observation of inverse Auger:  $K^2L^8M^2 + e \rightarrow K^2L^7M^3 \rightarrow K^2L^8M^2 + \mathbf{h\nu}_{target}$

XUV-FEL :  $K^2L^8M^3 + h\nu \rightarrow K^2L^7M^3 \rightarrow K^2L^8M^2 + \mathbf{h\nu}_{target}$

That is the **same emission** and can hardly be distinguished



## What else to observe ?

Detailed atomic physics of the L-shell:  $L^8 = 2s^2 2p^6$

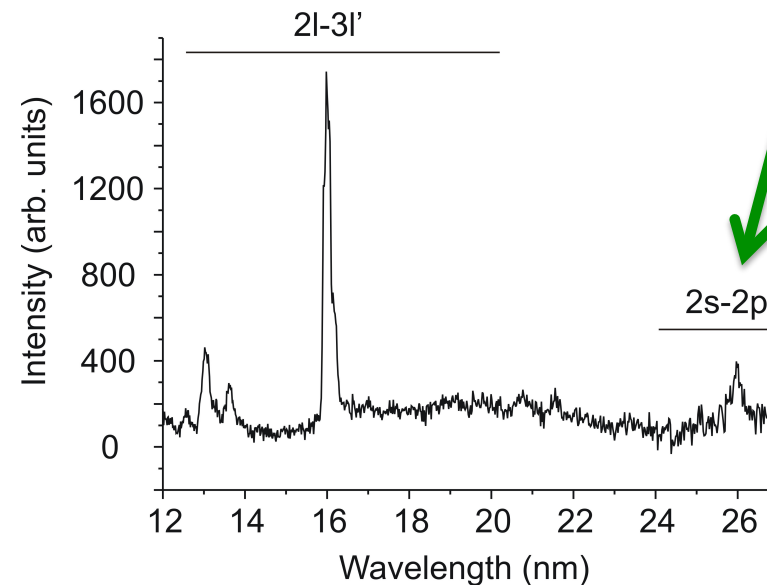
Photoionisation:  $K^2 2s^2 2p^6 M^3 + h\nu(92\text{eV}) \rightarrow K^2 2s^2 2p^5 M^2 + e$

$K^2 2s^2 2p^6 M^2 + h\nu(92\text{eV}) \rightarrow K^2 2s^1 2p^6 M^2 + e(-20\text{ eV})$

Inverse Auger:  $K^2 2s^2 2p^6 M^2 + e \rightarrow K^2 2s^2 2p^5 M^3$

$K^2 2s^2 2p^6 M^2 + e \rightarrow K^2 2s^1 2p^6 M^3 \rightarrow K^2 2s^2 2p^5 M^3 + h\nu_{2p-2s}$

=> The 2s-hole is from  
"Inverse Auger"  
=> 2p-2s intra-shell line  
comes from " $kT_e$ "



## Density estimate

Spectral distribution :

$$I(\omega) \approx \sum_k f_k \sum_{i,j} \frac{\hbar\omega_{k,ji}}{4\pi} g_{k,j} A_{k,ji} \exp(-\Delta E_{k,ji} / kT_e) \Phi_{k,ji}(\omega_{k,ji}, \omega)$$

$$\frac{\sum_{i,j \in M^2} \frac{\hbar\omega_{M^2,ji}}{4\pi} g_{M^2,j} A_{M^2,ji} \exp(-\Delta E_{M^2,ji} / kT_e)}{\sum_{i,j \in M^3} \frac{\hbar\omega_{M^3,ji}}{4\pi} g_{M^3,j} A_{M^3,ji} \exp(-\Delta E_{M^3,ji} / kT_e)} \approx \text{const.}$$

Fitted  $f_k$  parameters provide population ratio  $n(K^2L^7M^2)/n(K^2L^7M^3)$ :

Saha-Boltzmann



$$n_e \approx 3 \times 10^{22} \text{ cm}^{-3}$$

This is consistent :  $n_e C > A + \Gamma$

# X-FEL induced Hollow Ion Emission: One or two photons?

Inner-shell photoionization:

$$E_{\text{X-FEL}} = 3.1 \text{ keV} > E_i(1s^1)$$



Relaxation time:

$\tau(\text{autoionization}) \rightarrow \text{some } 10 \text{ fs}$



F.B. Rosmej, R.W. Lee, EPL 77, 24001 (2007)

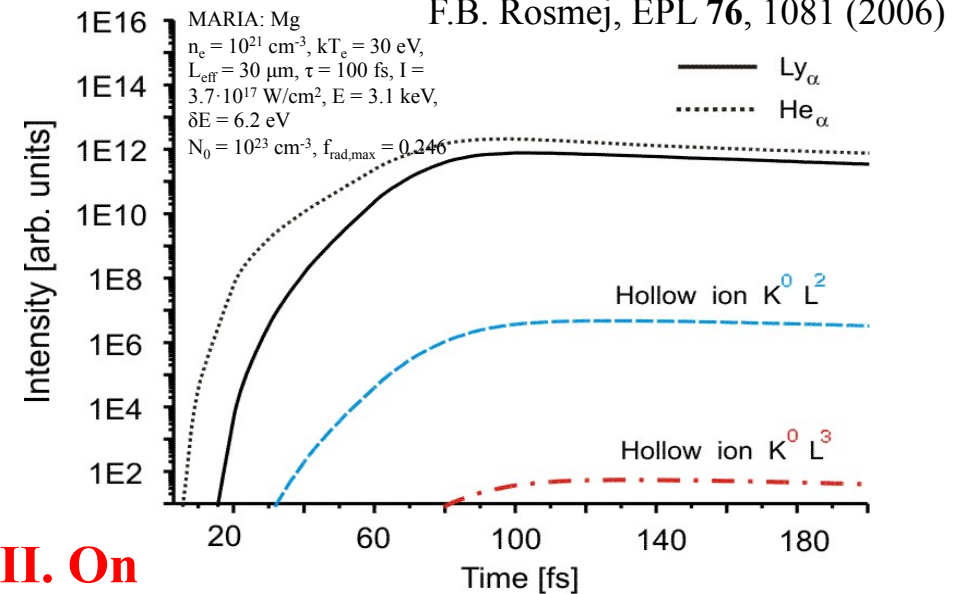
F.B. Rosmej et al., HEDP 3, 218 (2007)

**10 orders of magnitude will open up  
a new field of research**

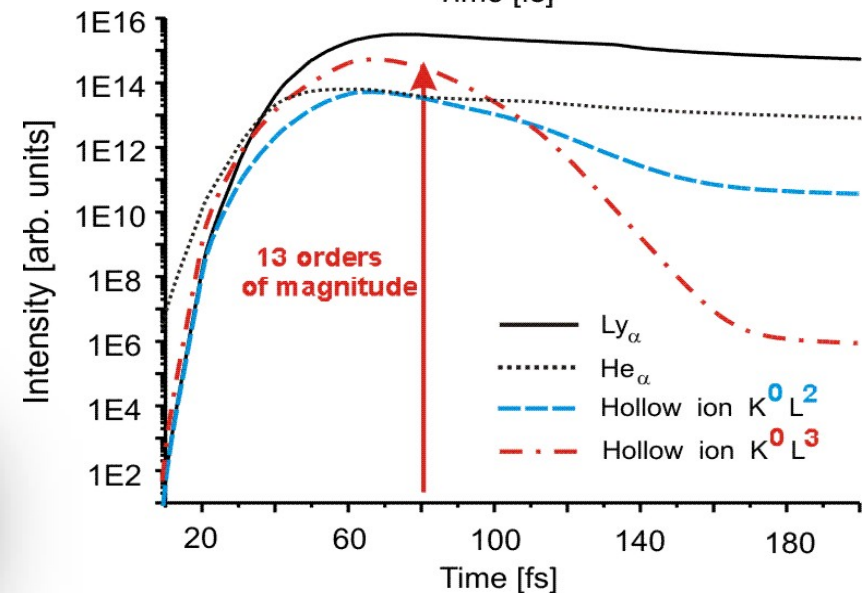
## II. Off

MARIA-code:

F.B. Rosmej, EPL 76, 1081 (2006)



## II. On



F.B. Rosmej/UPMC/LULI

# The temporal evolution

**XUV-spectroscopy of hole states provides proof of :**

- near solid density, high temperature
- **time evolution and heating mechanisms (Auger)**



10 fs Interaction FLASH-solid

fs Photoionisation  $K^2L^8M^3 + h\nu - K^2L^7M^3 + e$

Equilibration of photo-electrons in the conduction band:  $kT_e \approx 8 \text{ eV}$

50 fs Relaxation (potential energy) of hole states:  $K^2L^7M^3 - K^2L^8M^{1+} + e_{\text{Auger}}$

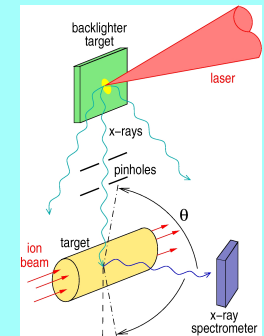
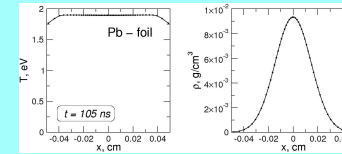
fs Auger electron energy is redistributed in the conduction band

ps Begin of crystal destruction and inverse Auger effect

ps XUV target emission from hole states

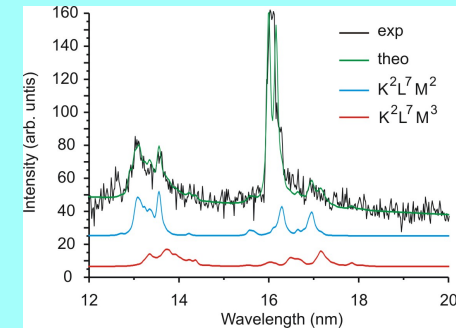
# IV. Conclusion and outlook

- XUV/X-ray FEL studies complementary to WDM produced by intense heavy ion beams:
  - opacity
  - radiative properties: scattering/emission



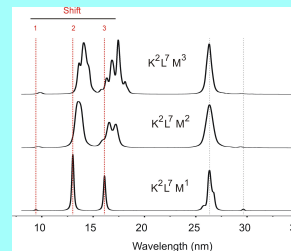
- L-shell spectroscopy and atomic physics identifies hole states:
  - Identification of Auger Effect
  - Signature of Auger heating

E. Galtier, F.B. Rosmej et al., PRL **106**, 164801 (2011)



- Time resolution via "atomic physics":
  - Radiation time scale is driven by Auger time scale (some 10 fs)

- Hollow ion X-ray emission:
  - 10 fs time scales
  - no opacity/transparency



**Spectroscopy is a non-interfering matter probe**

F.B. Rosmej, R.W. Lee: "Hollow ion emission driven by pulsed x-ray radiation fields", EPL **77**, 24001 (2007)

F.B. Rosmej/UPMC/LULI