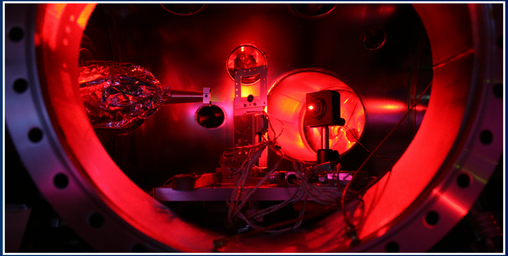

UNIVERSITY OF OXFORD

The Physics of Solid Density Plasmas Created by Intense X-Ray Free Electron Lasers

Justin Wark

Department of Physics
Clarendon Laboratory
&
Trinity College

University of Oxford, UK

justin.wark@physics.ox.ac.uk

Nobel Symposium on Free Electron Laser Research June 14-18, 2015

Outline

- Motivation: creating ‘warm dense matter’.
    - The choice of aluminium.
- First Experiments on FLASH at 92 eV.
    - Making transparent aluminium (saturable absorption in the XUV).
    - The electronic structure of warm dense matter.
    - The energy budget.
- Experiments on LCLS at 1400 - 1800 eV.
    - Measuring Ionisation Potential Depression (Al, Mg, Si)
    - DFT Theory of IPD
    - Saturable absorption at > 1500 eV.
    - First measurements of collisional ionisation rates at solid densities.
- Creation of uniform, optically-thin solid-density plasmas ~120 atoms across.
- Historical context: re-climbing Moseley’s ladder.

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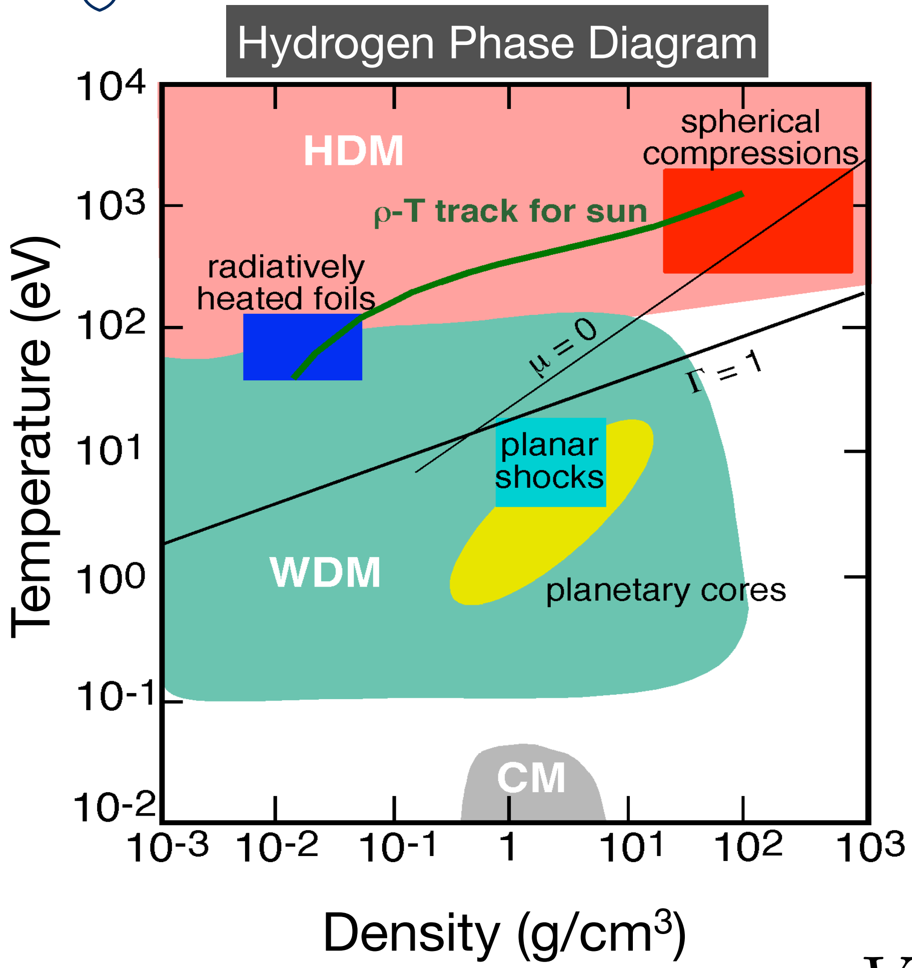
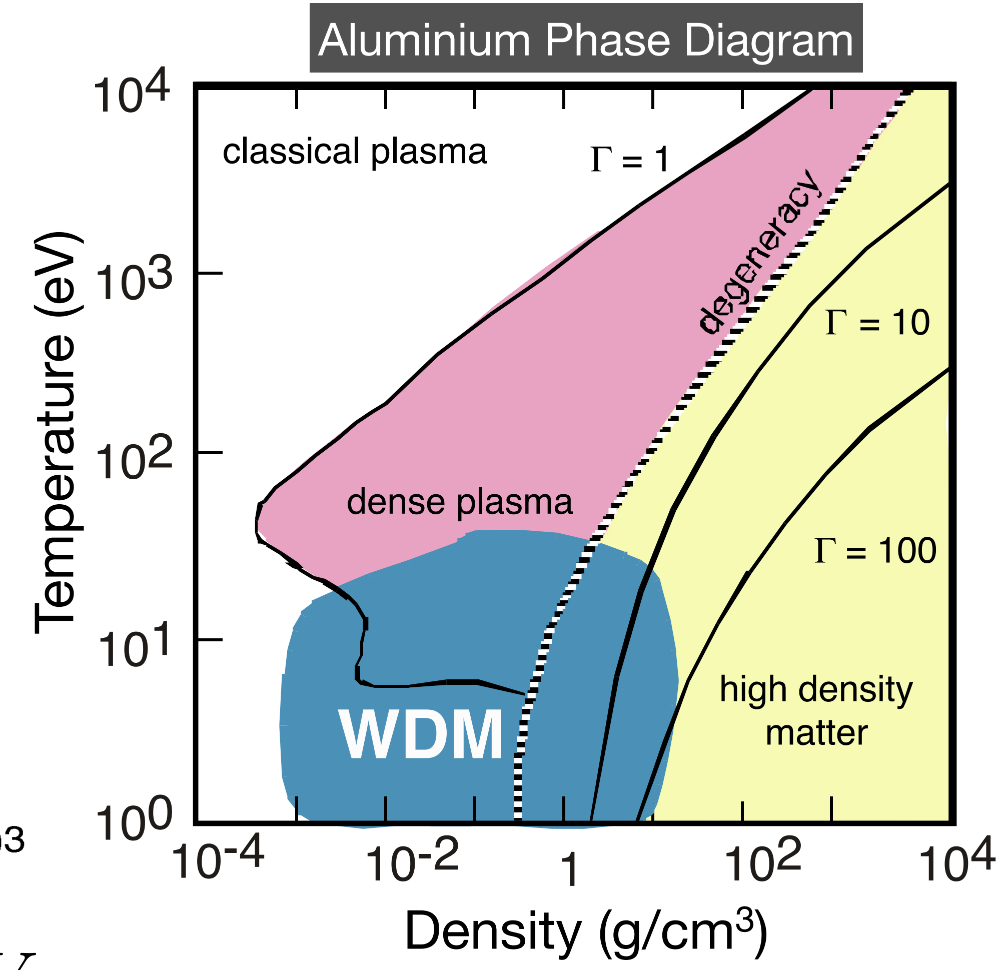
High energy-density systems: Warm and Hot Dense Matter





$$\Gamma = \frac{V_{\text{Coulomb}}}{E_{\text{Kinetic}}} \approx 1$$

X-ray lasers can isochorically heat matter - no hydrodynamic expansion

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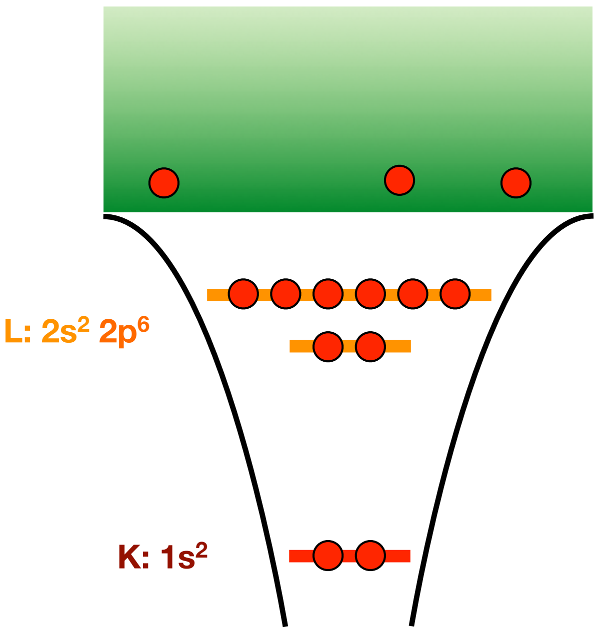
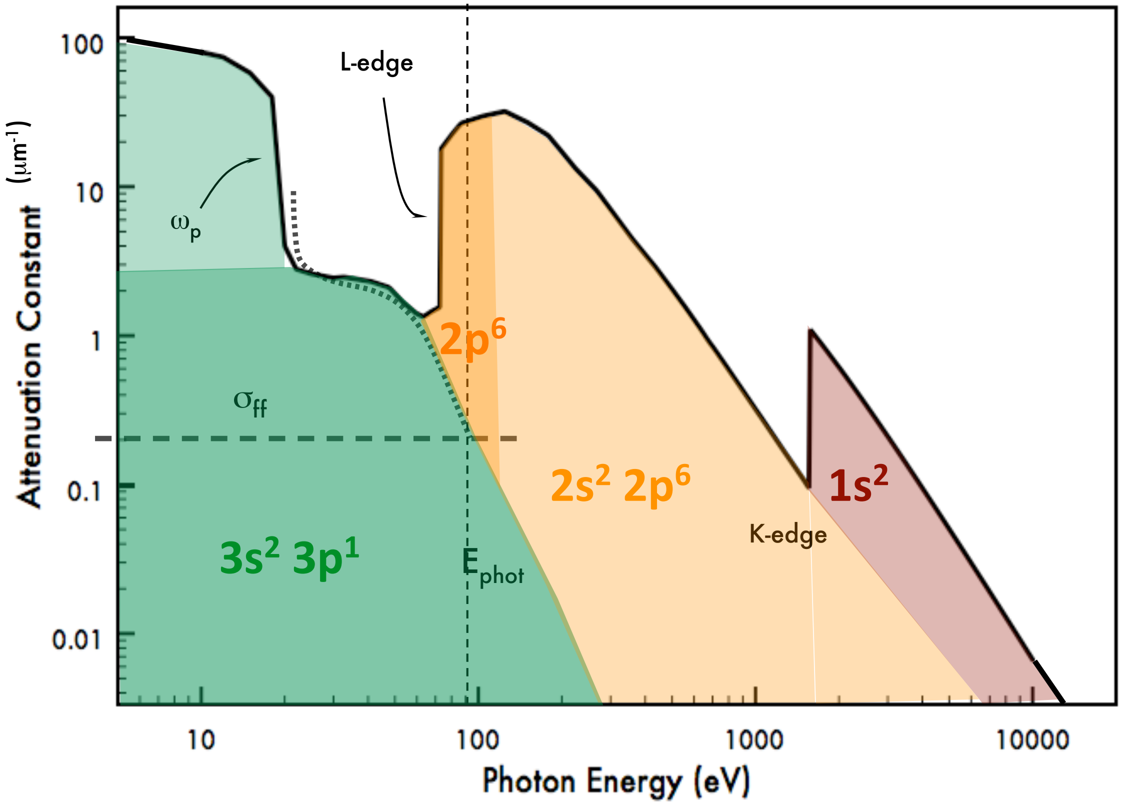
Electronic structure of Aluminium (Z=13)

Neutral Al





Aluminium has a relatively simple Neon-like core, but already displays 'plasma' conducting behaviour as a trivalent prototypical metal. The atomic physics is simple enough to be tractable, yet complex enough to be interesting. As such, it is the element of study for many warm dense matter experiments.

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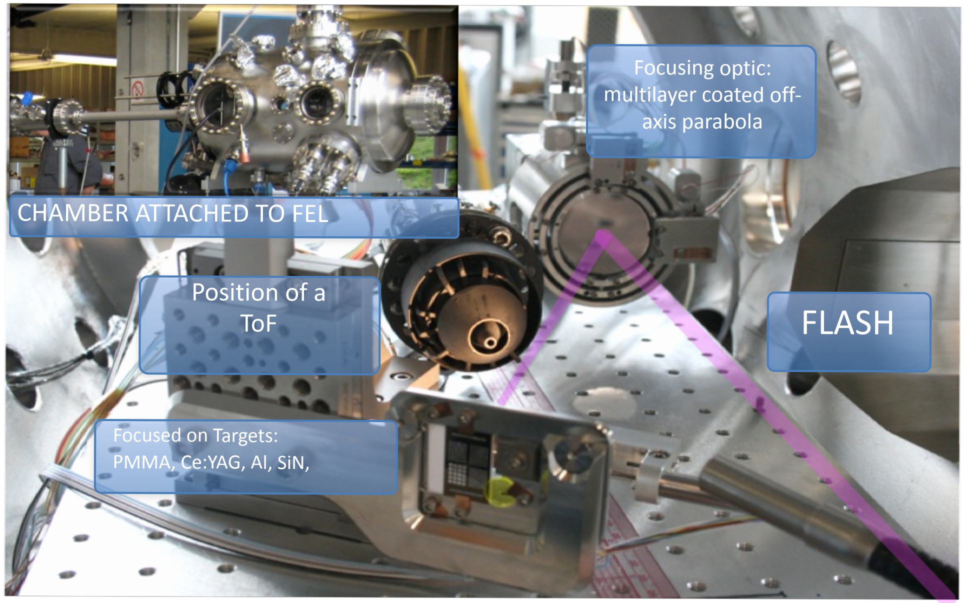
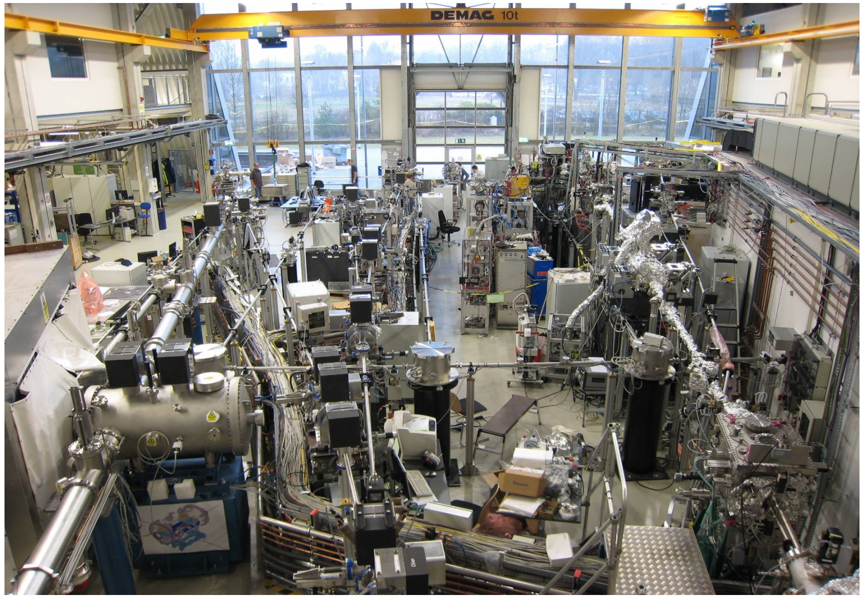
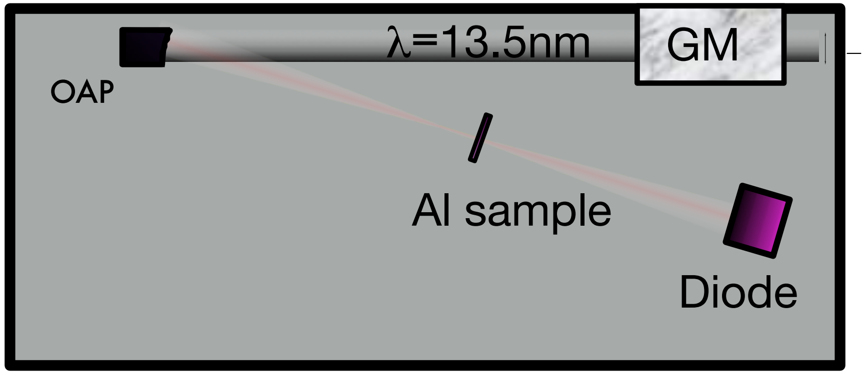
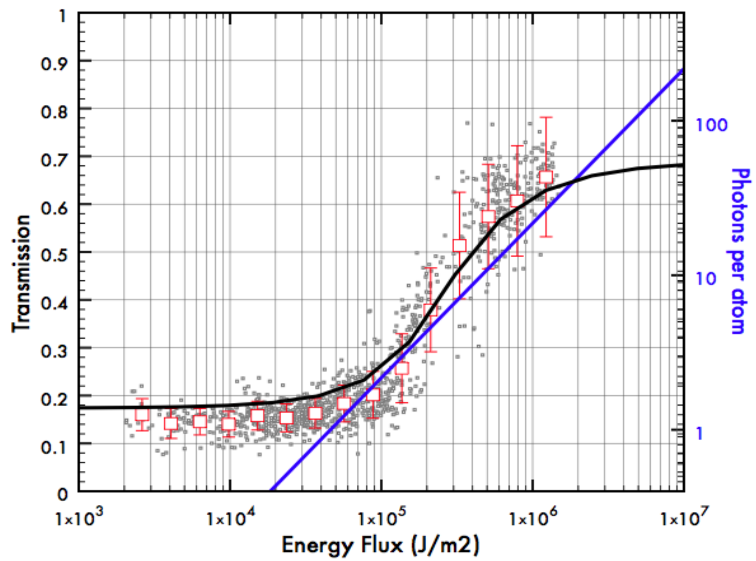
Irradiating thin (50-nm) Al with intense (> 10^{16}Wcm^{-2}) XUV (92eV) light









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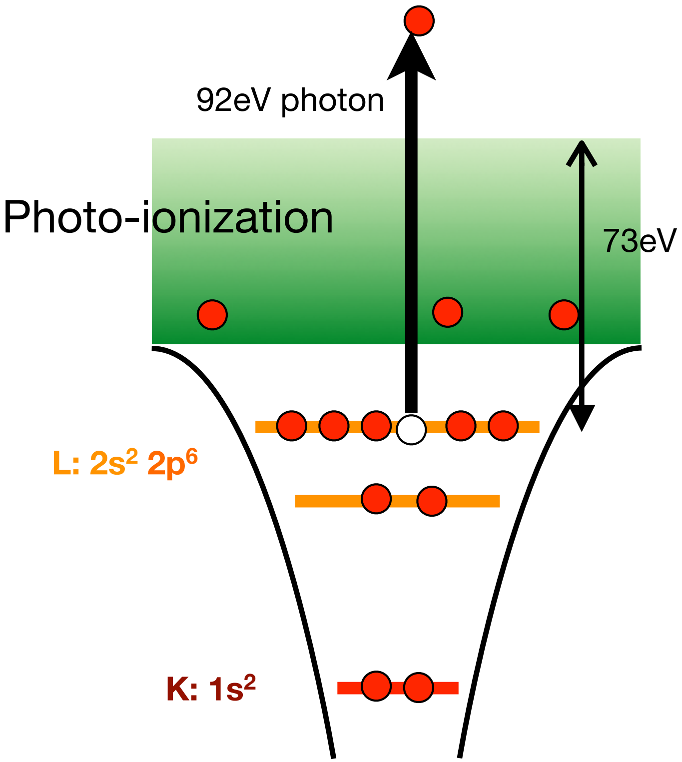
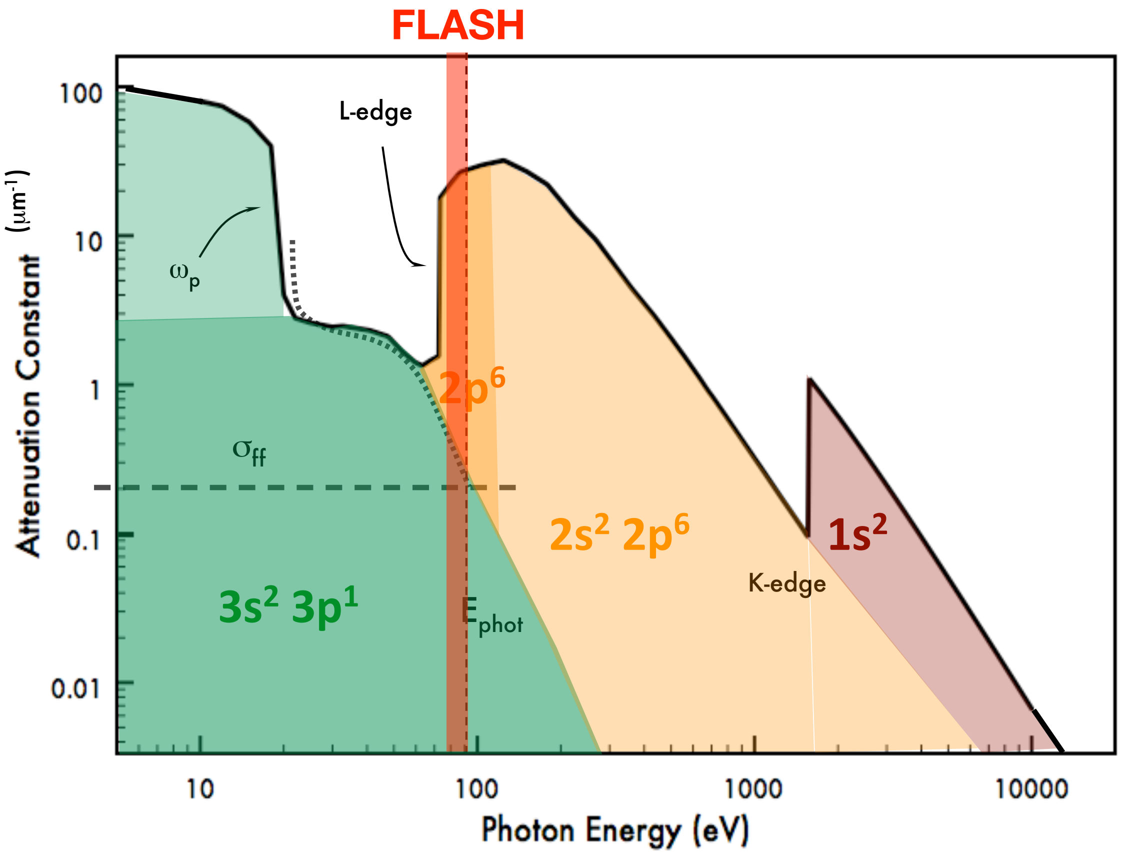
Electronic structure of Aluminium

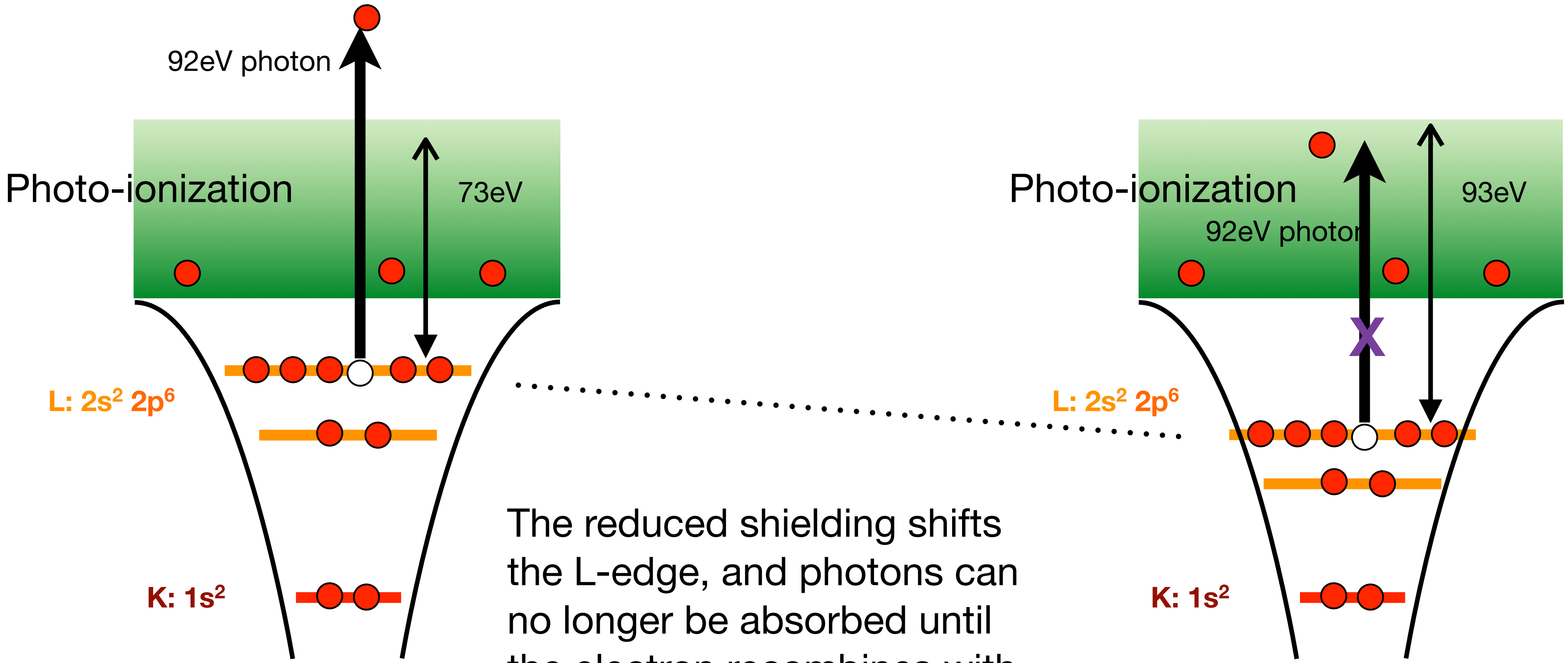
Neutral Al





L=shell lifetime ~40 fs

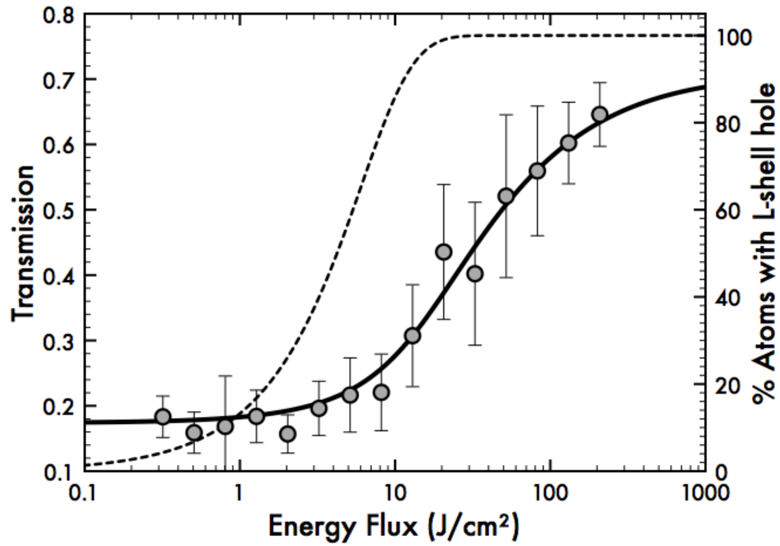
Ejection of the L shell electron shifts the L-edge



The reduced shielding shifts the L-edge, and photons can no longer be absorbed until the electron recombines with a lifetime of 40-fs, but this is longer than the 15-fs pulse length.

Meanwhile, the electron starts to thermalise with the Fermi sea, heating it up.

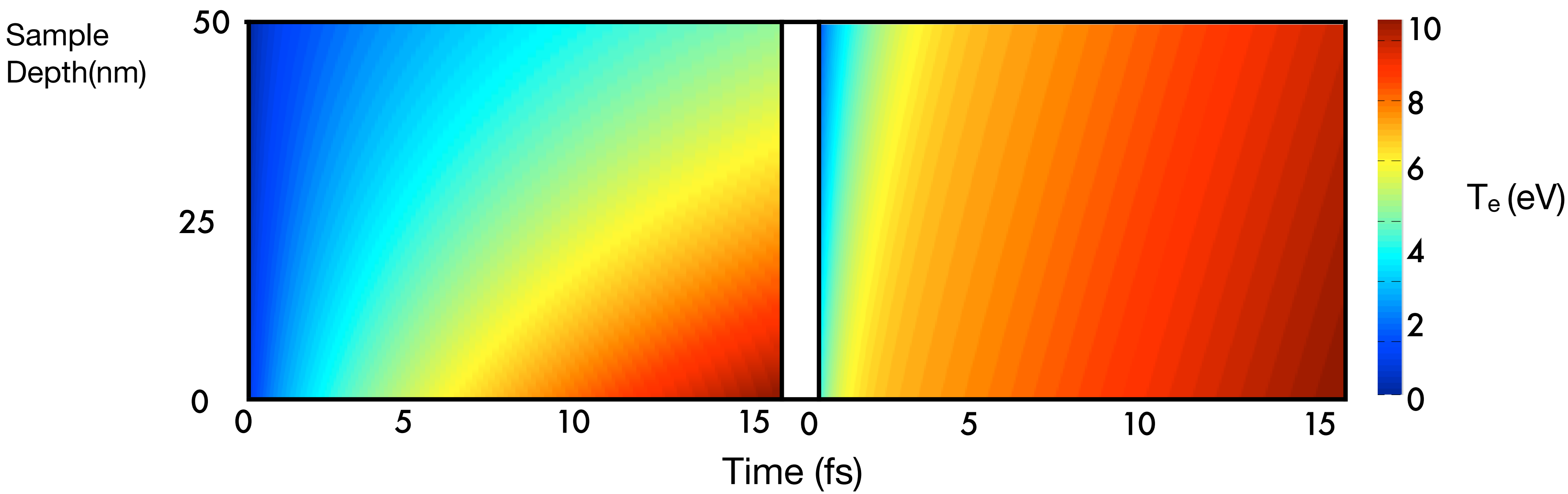
Saturation of L-shell holes, and homogeneous *isochoric* heating



The plot shows the percentage of atoms with an L-shell hole at the end of the 15-fsec pulse.

Note 100% is reached at fairly low fluences - but the transmission is still not very high at such a fluence, because we are plotting the transmission integrated in time, and there is always relatively high absorption at the start of the pulse.

The saturable absorption leads to much more uniform energy deposition within the fold



Nature Physics ***5****, 693 (2009)*

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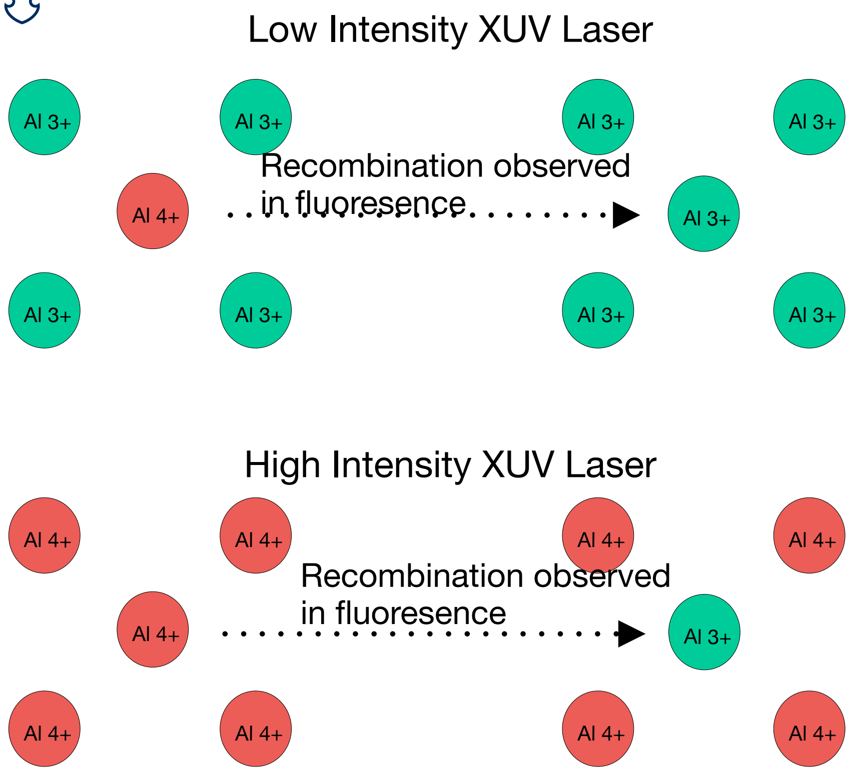
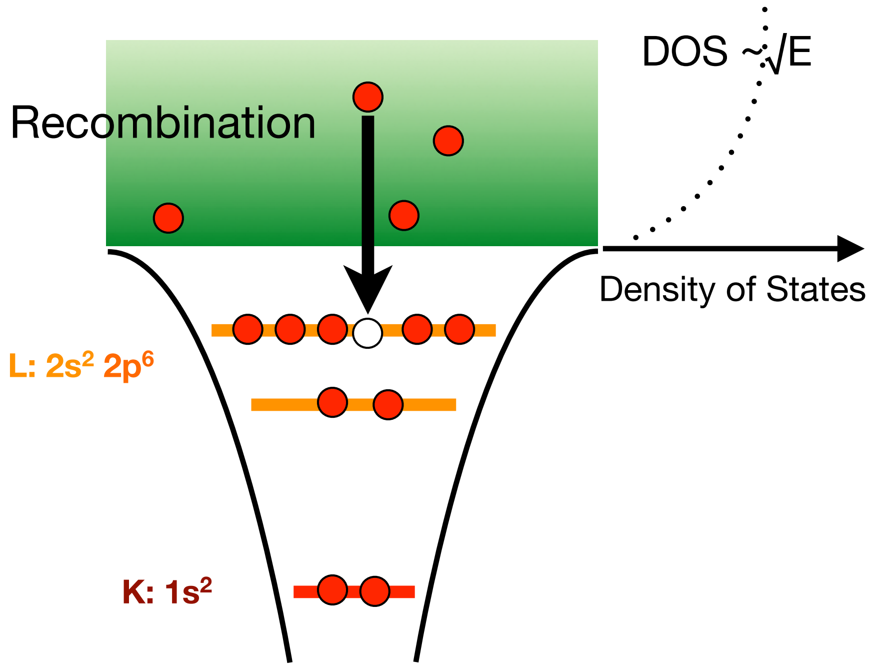
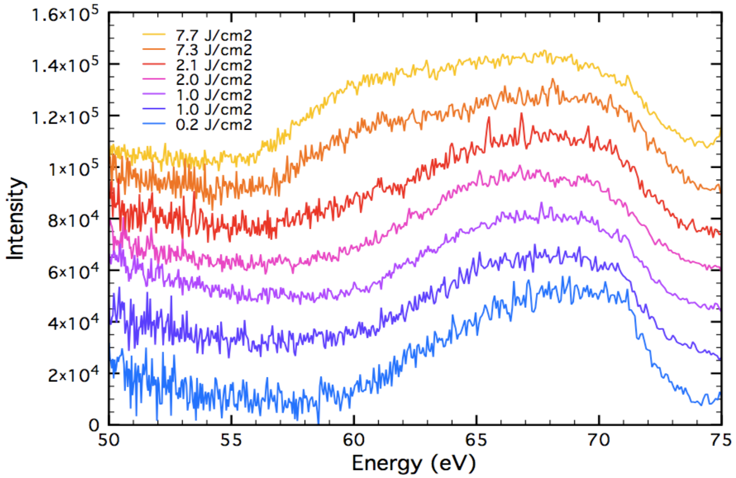
Probing the electronic structure of a solid density plasma

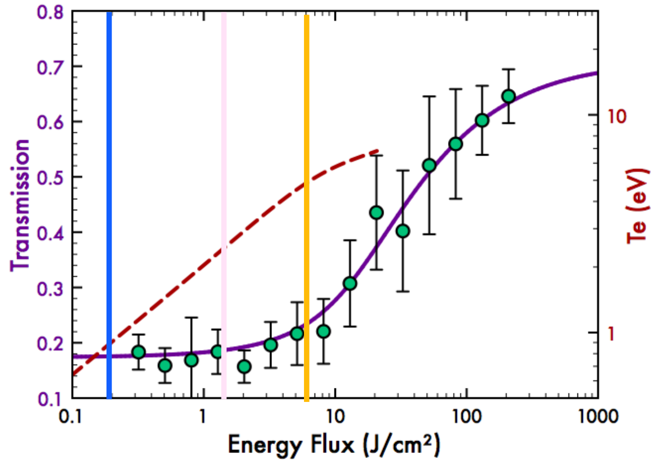


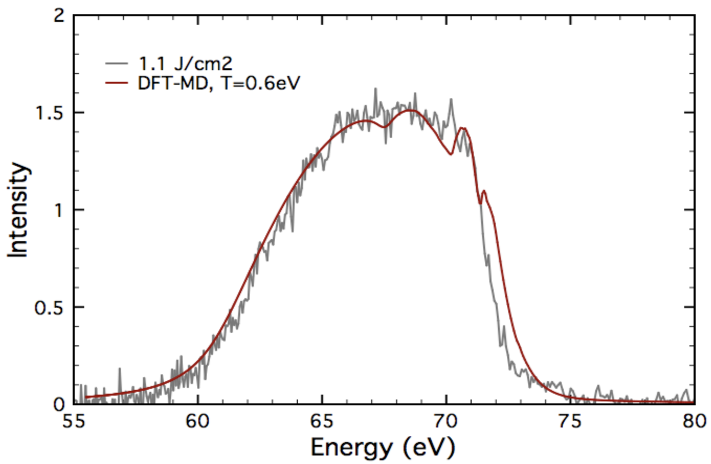


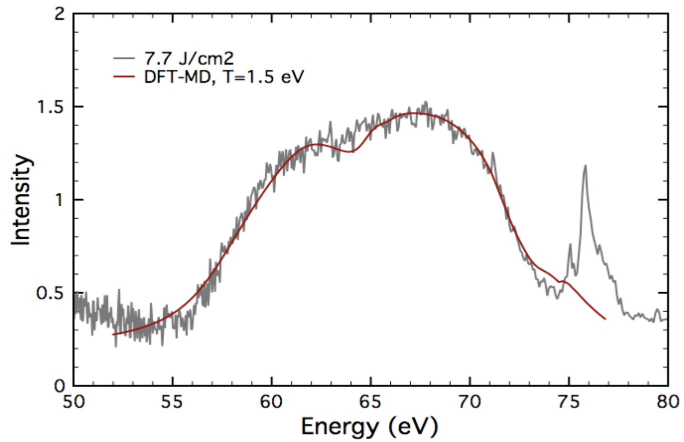
The environment surrounding the recombining ion changes with increasing saturation, and the average free electron density changes from 3+ to 4+. This will alter the electronic structure (the density of states) and therefore the observed XUV emission spectrum, where we see fluorescence as electrons recombine from the continuum.

Probing the electronic structure of a solid density plasma









The emission spectra show the shape of the density of states of the ionised plasma. The inferred temperatures will be below those predicted to exist at the end of the pulse, as the continuum electrons are heated by the Auger effect as recombination occurs.

There is good agreement between experiment and ab initio Density Functional Theory calculations of the density of states.

Phys. Rev. Lett, ***104***, *225001 (2010)*

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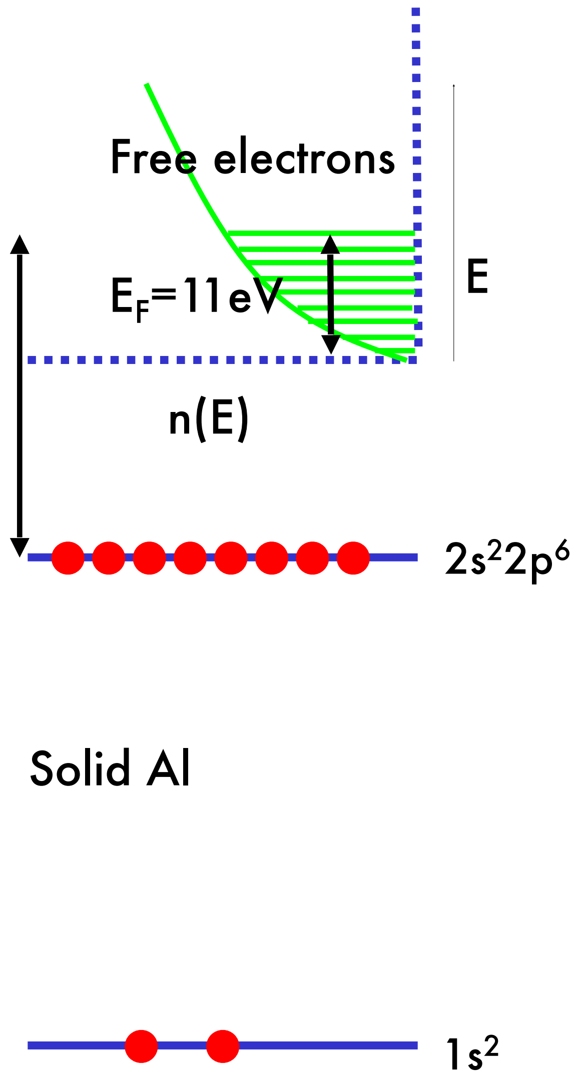
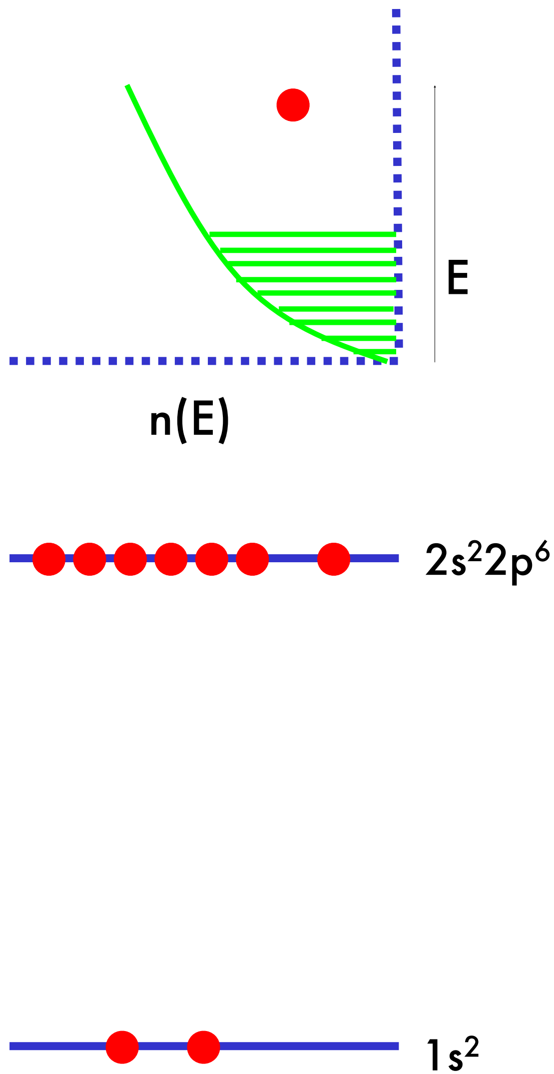
Energy Budget: Highest Intensities



+ 92eV =



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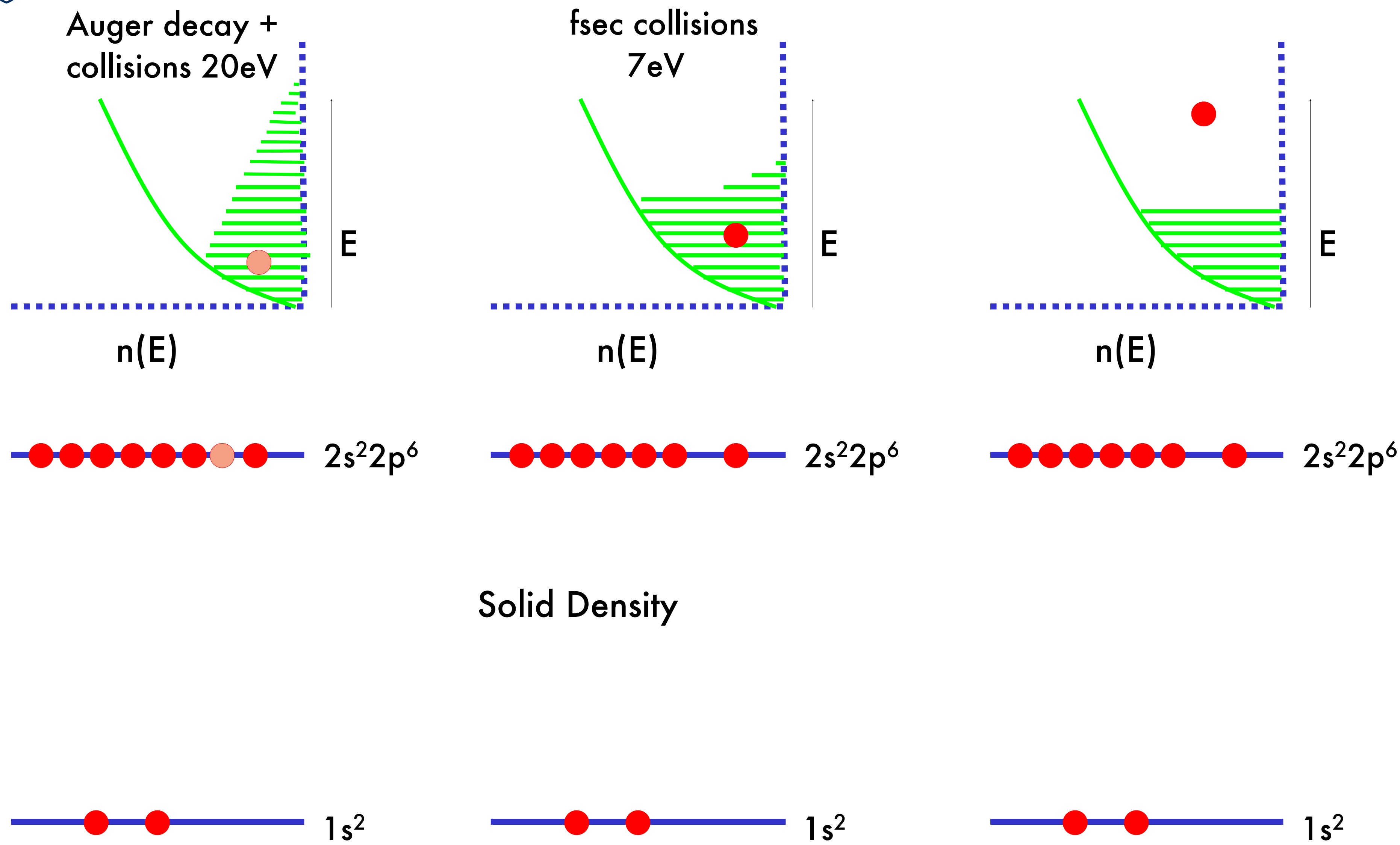

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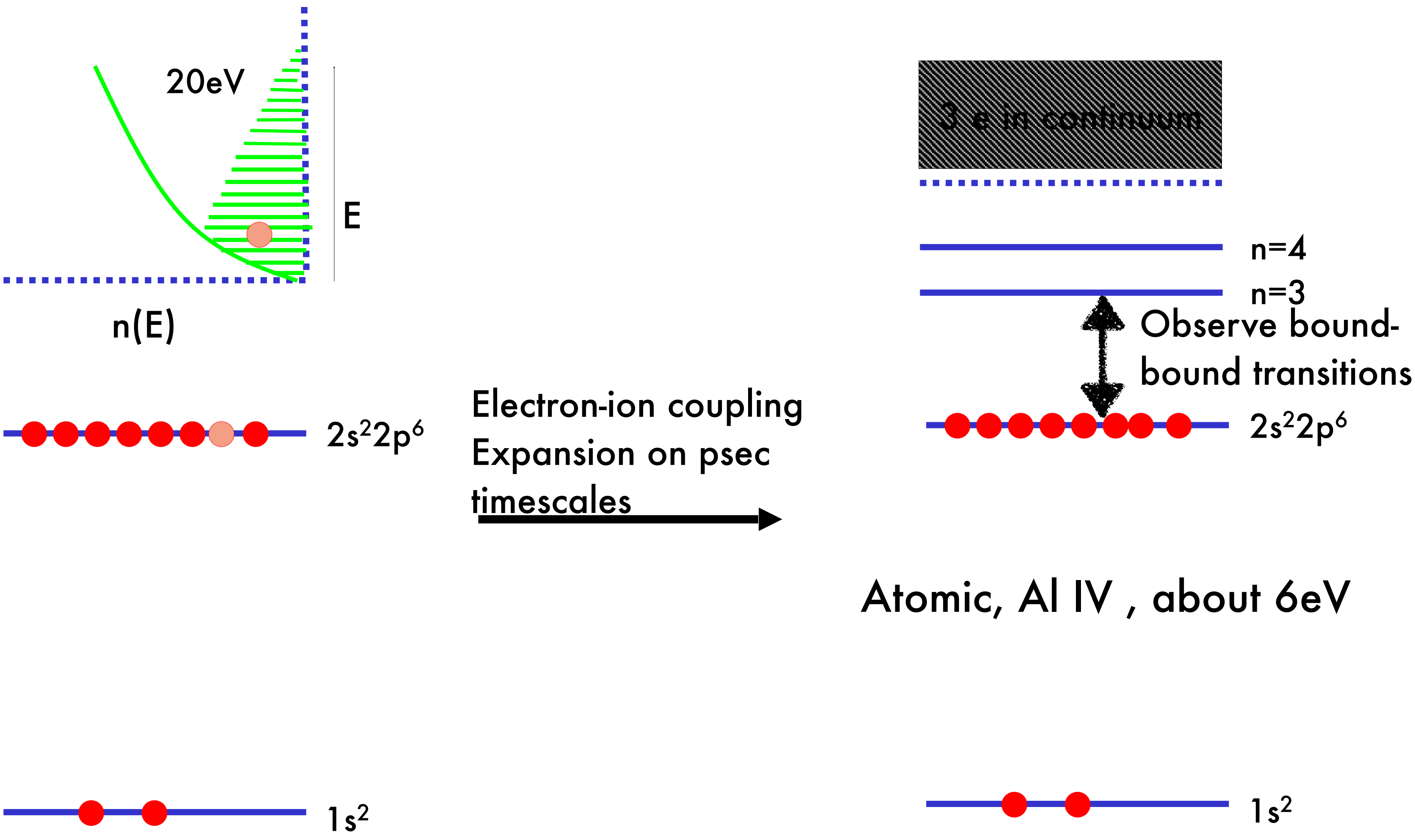
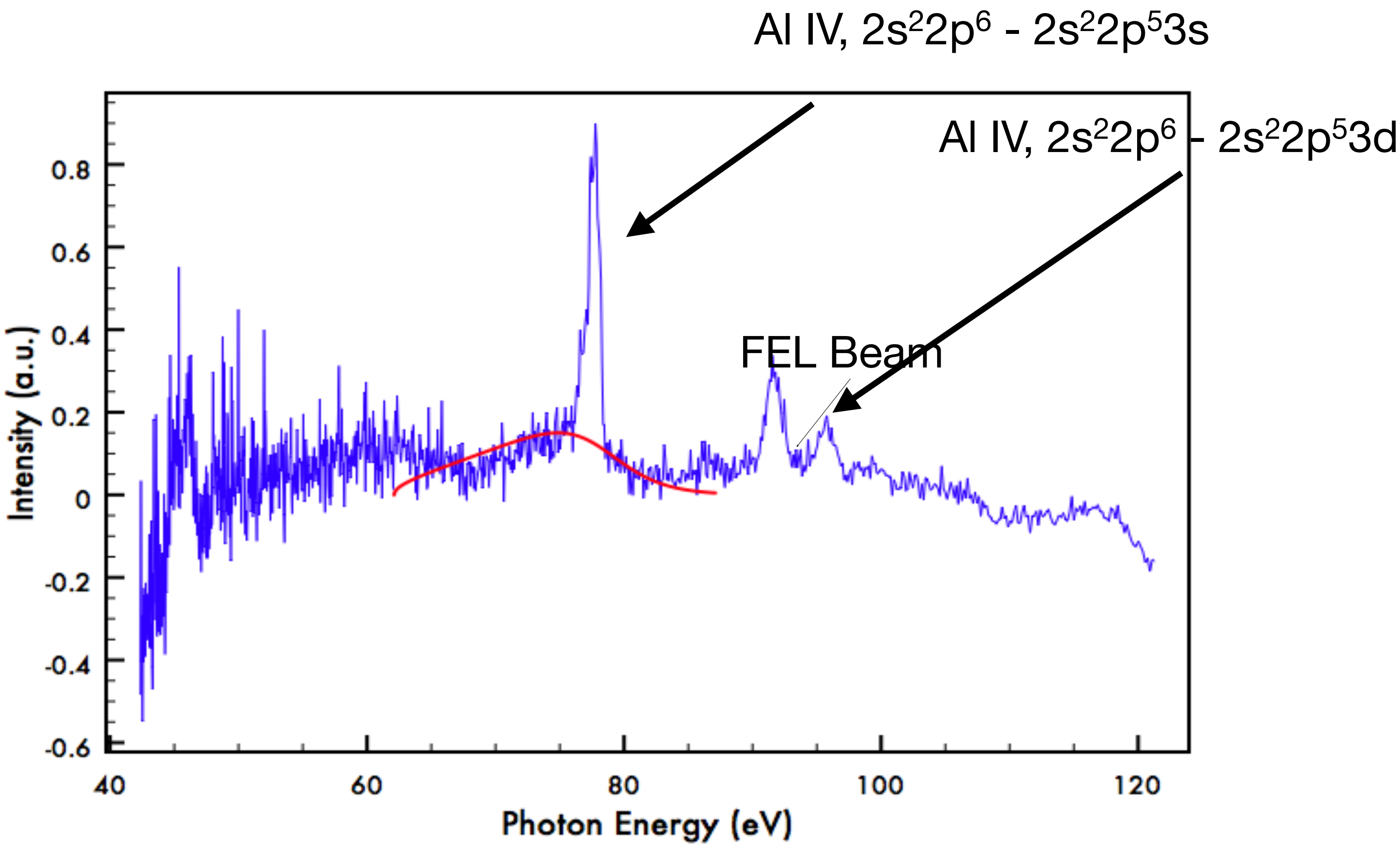


Energy Budget: Highest Intensities

- After several picoseconds we expect the target to disassemble, and see atomic spectra. We only see Al IV lines, consistent with assumed final temperatures of order 6-eV. Little evidence of Al V.



Phys. Rev. Lett, ***106***, *164801 (2011)*

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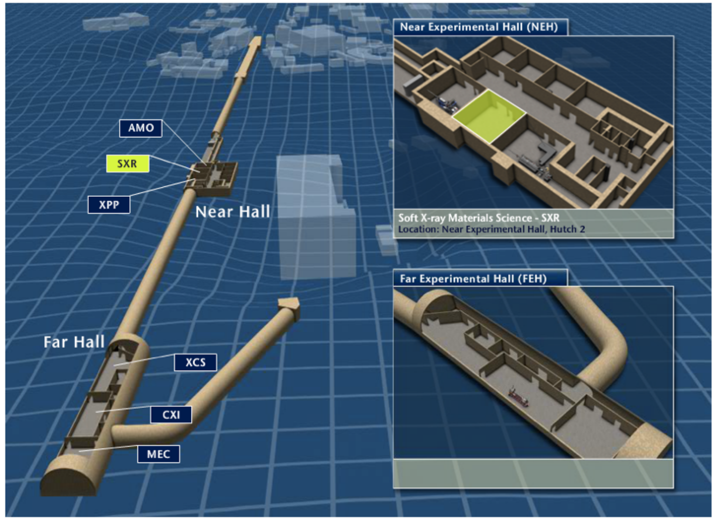
The experiment at the Linac Coherent Light Source X-ray Free-Electron Laser



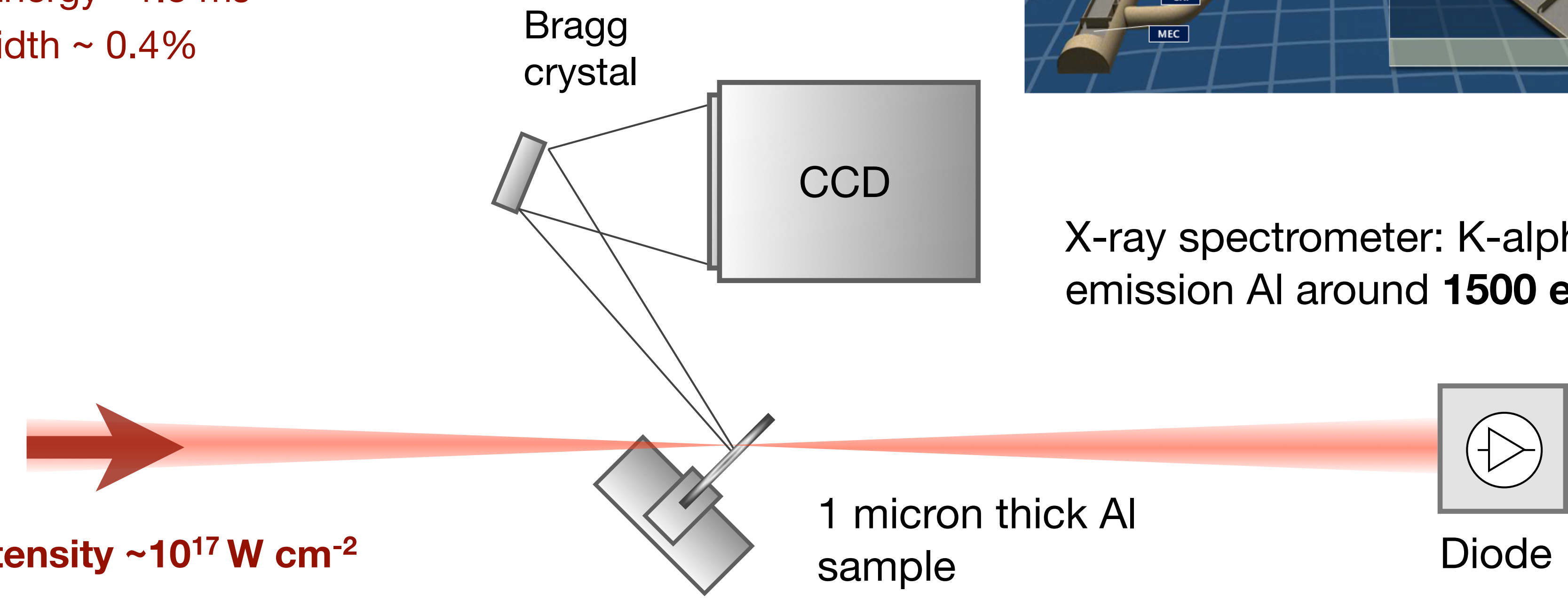
LCLS pulse

Photon energy: 1460–1830 eV

Pulse length < 60 fs

Pulse Energy ~1.5 mJ

Bandwidth ~ 0.4%



X-ray spectrometer: K-alpha emission Al around **1500 eV**

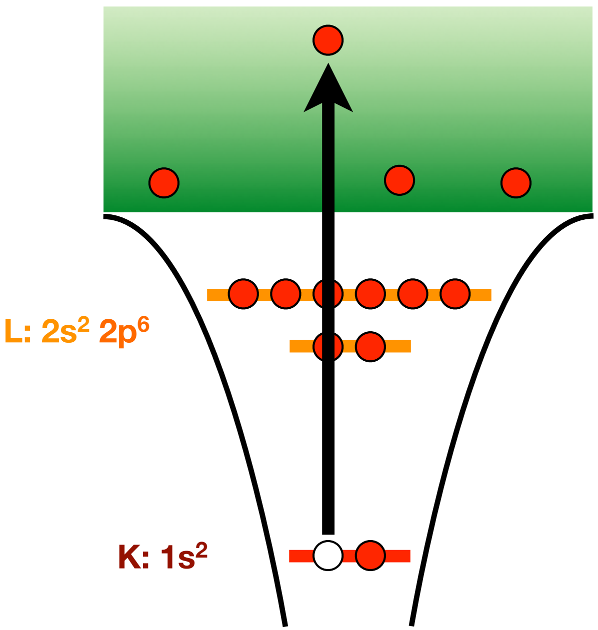
Peak Intensity ~10^{17} W cm^{-2}

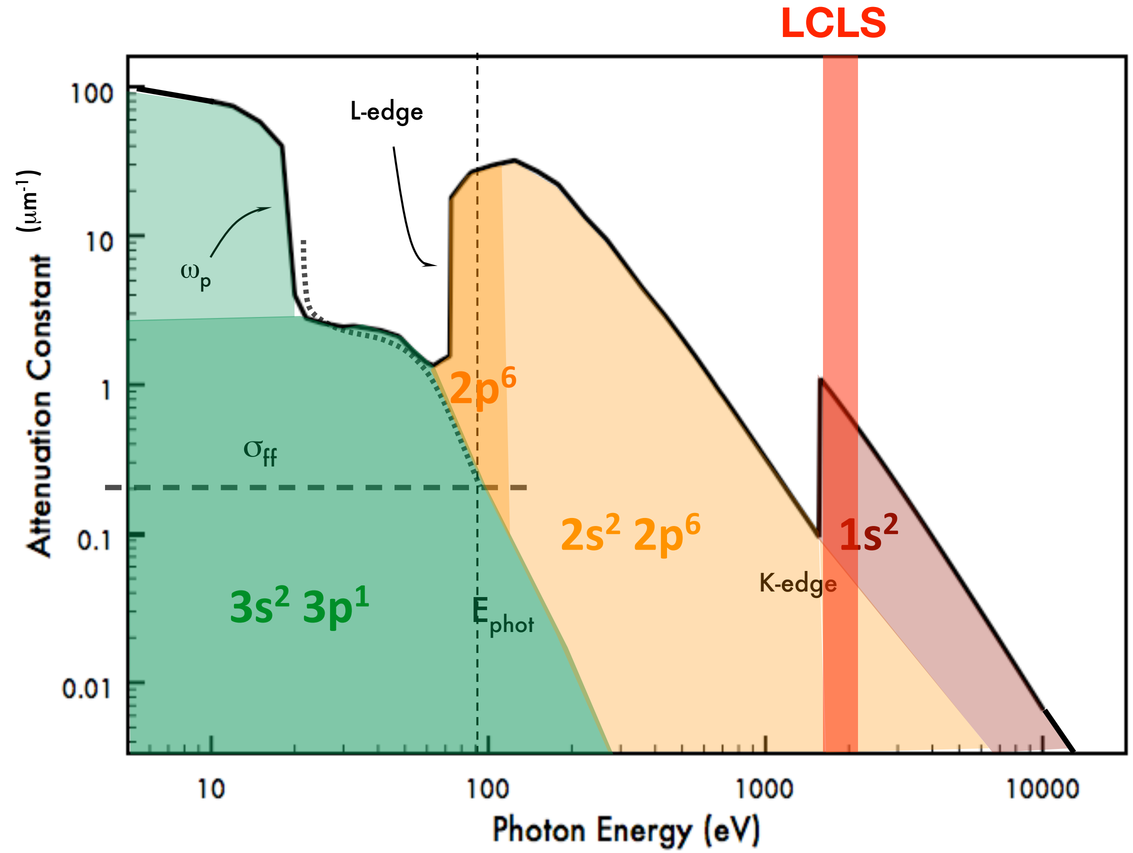
Vinko et al., Nature **482**, 59 (2012)
Ciricosta et al., PRL **109**, 065002 (2012)

Electronic structure of Aluminium

Neutral Al

Photo-ionization



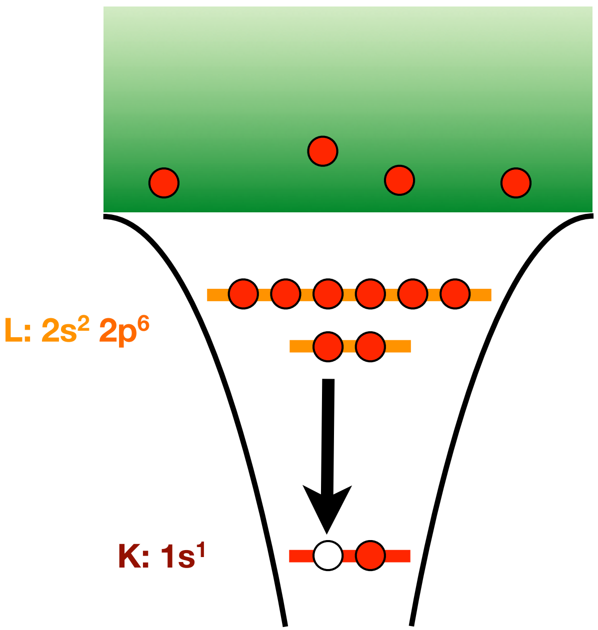


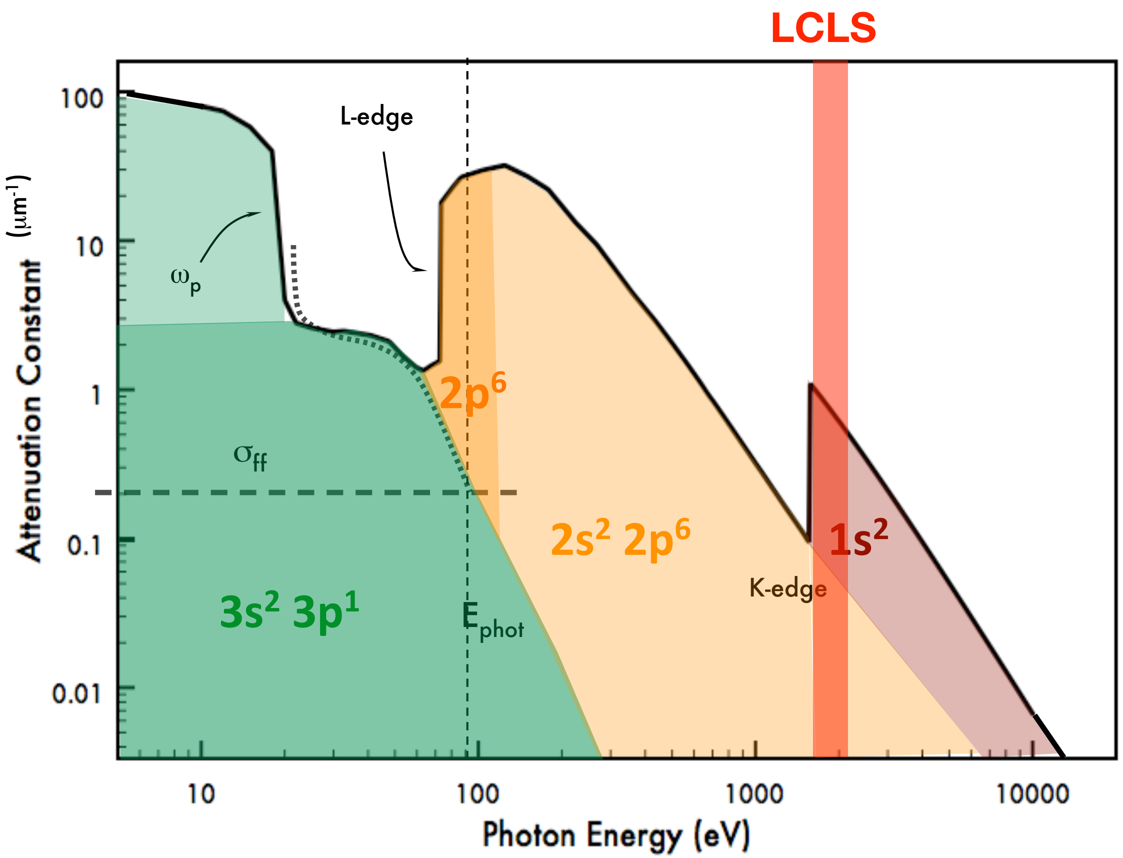
Core-hole lifetime ~1fs

Electronic structure of Aluminium

Neutral Al

K-alpha emission

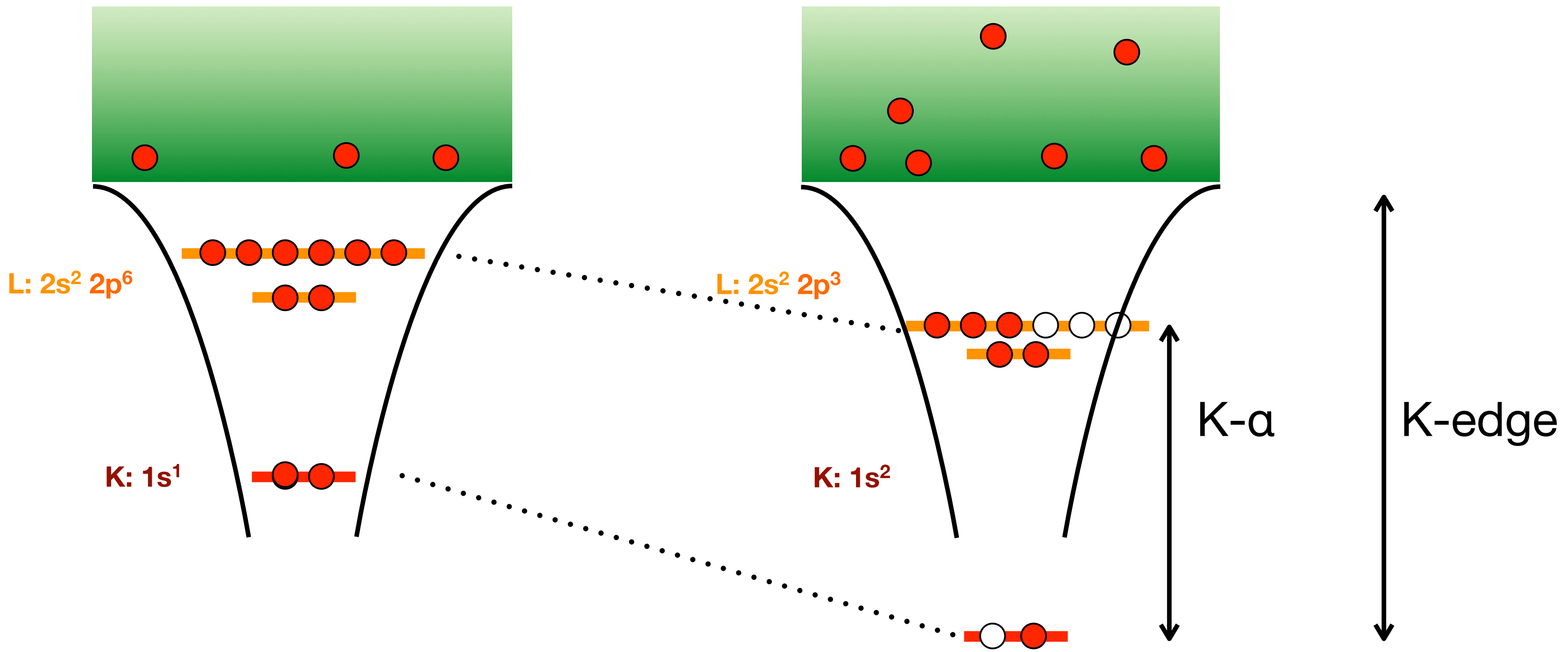




Core-hole lifetime ~1fs

Electronic structure of Aluminium

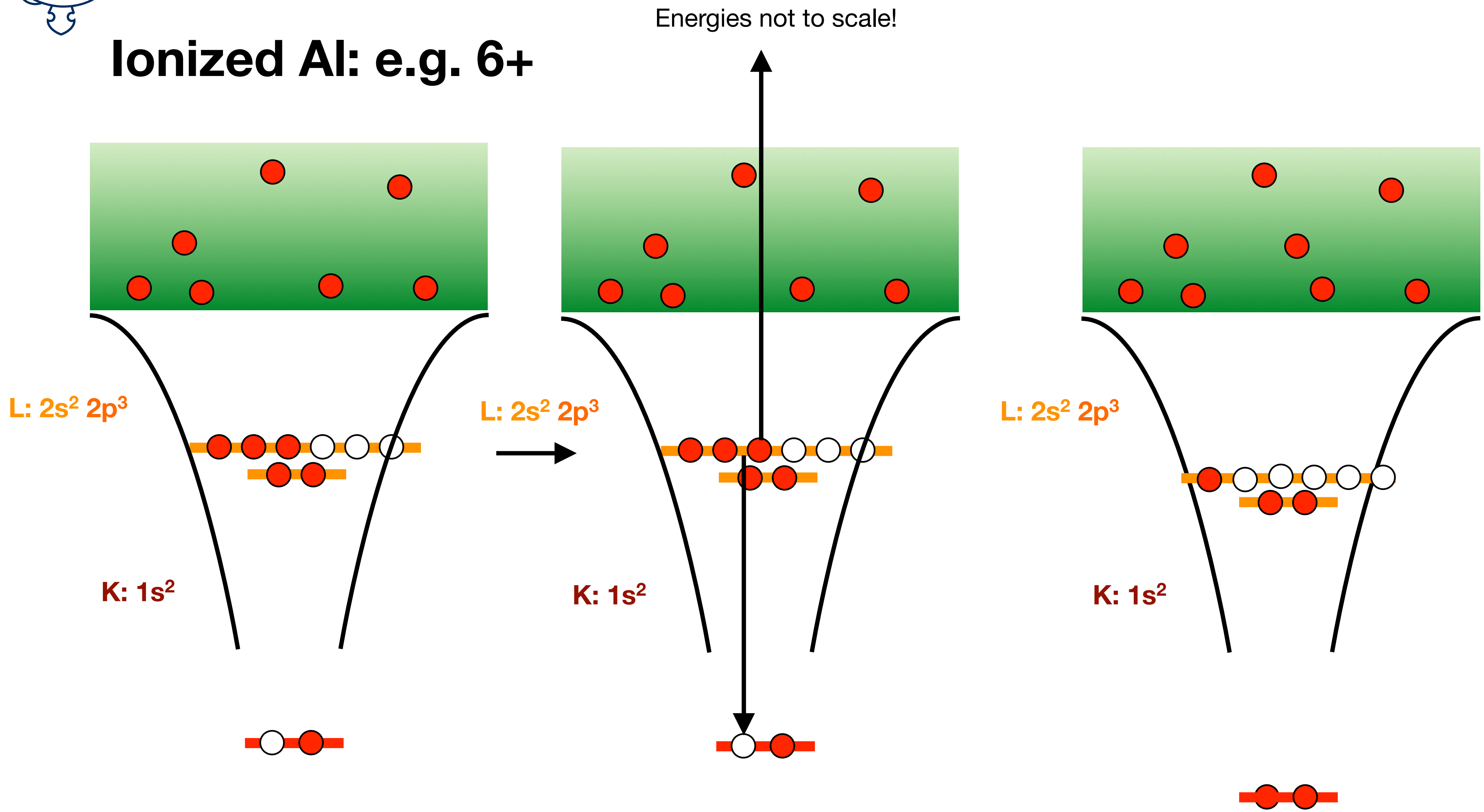
Neutral Al: **Ionized Al: e.g. 6+**



Note that both the K-edge energy, and the K-α energy increase with increasing charge state.

The FEL creates core holes, and the dominant heating is Auger

Ionized Al: e.g. 6+

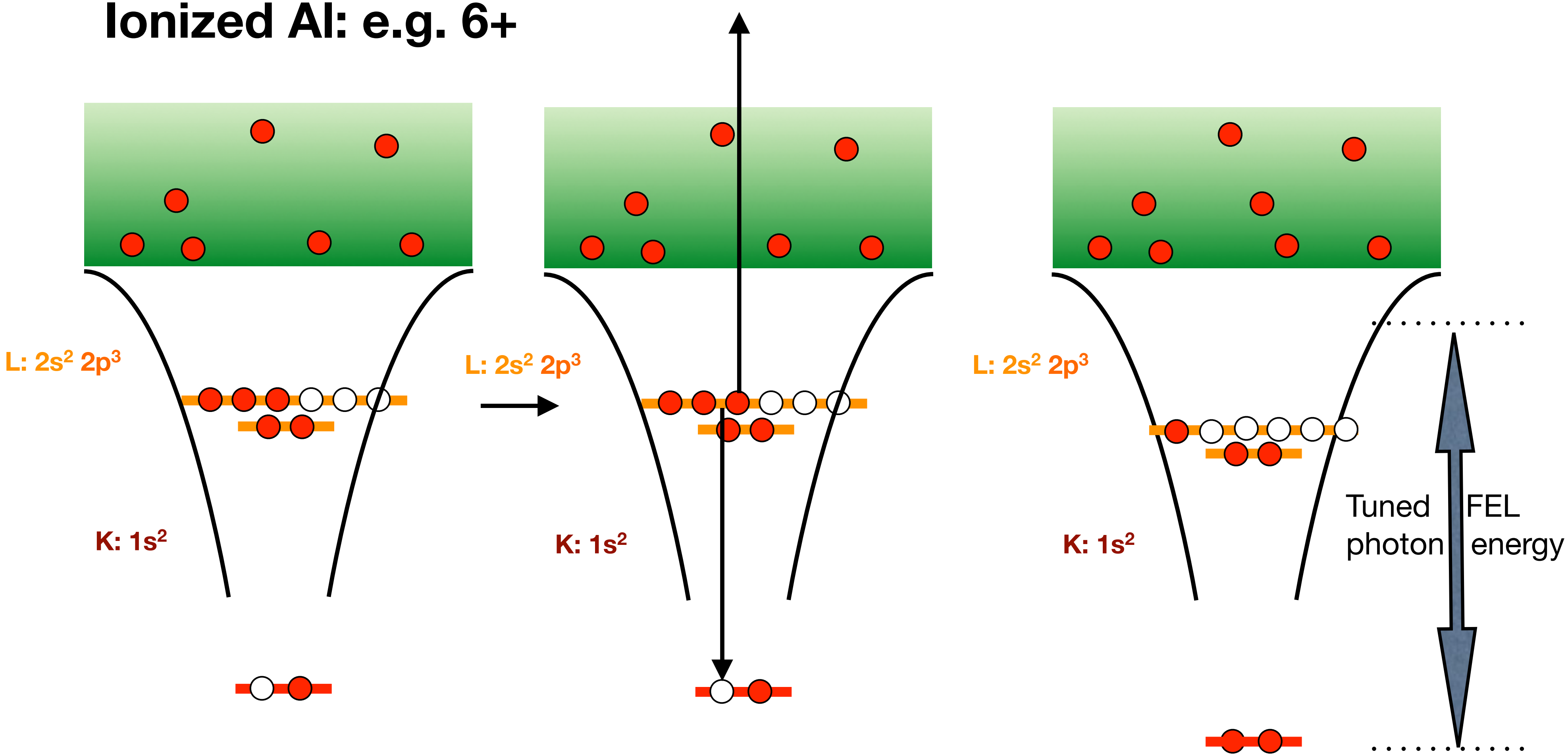


Important to note: the heated continuum - many tens of eV, can further collisionally ionise the system. No K-α will be seen if the K-edge energy of an ion exceeds the photon energy of the X-Ray Laser. Temperature of plasma insufficient to excite n=2. *We only see the K-shell emission while th x-ray laser is on.*

If the FEL photon energy is too low, no K-a generated

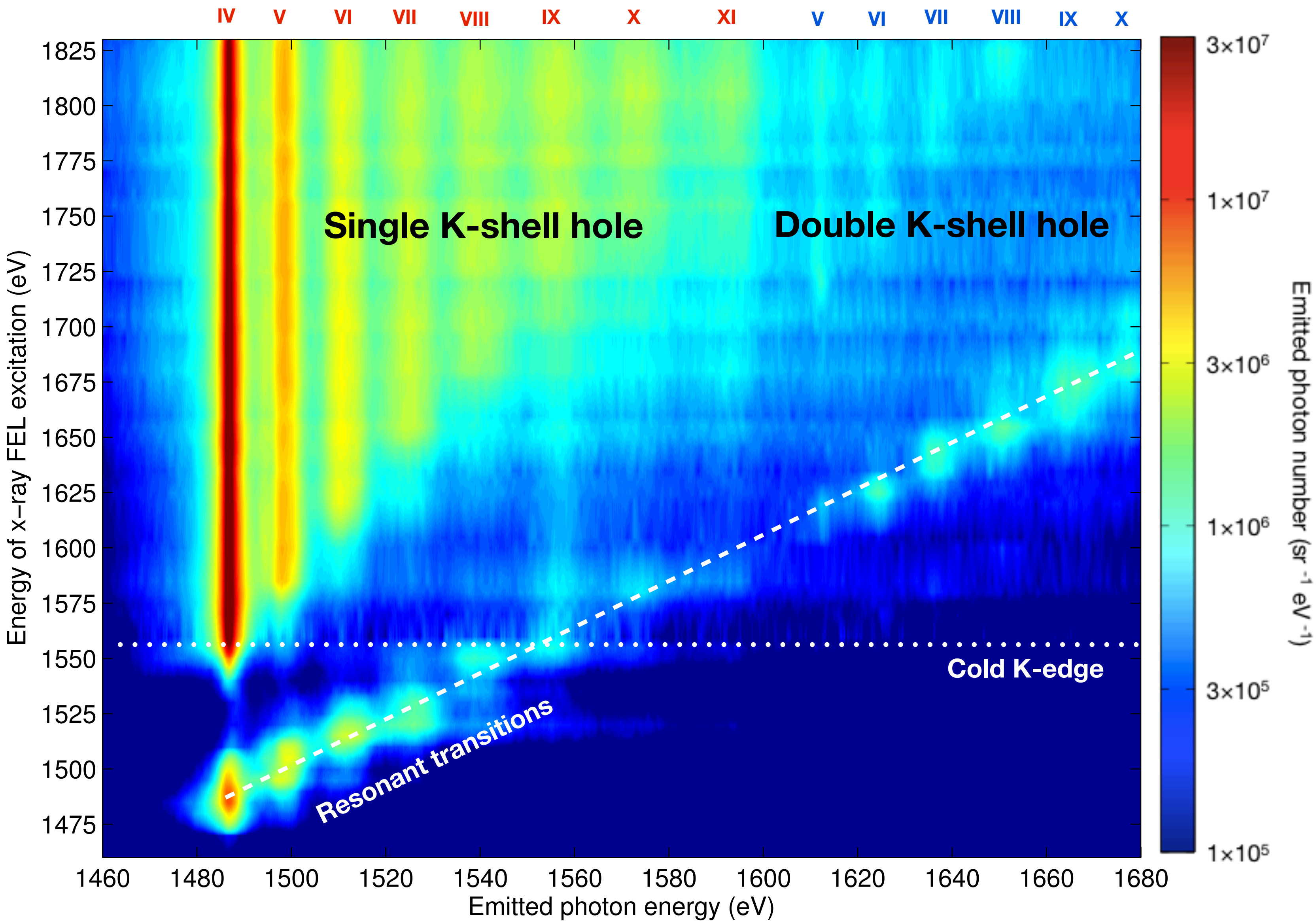
Energies not to scale!

Ionized Al: e.g. 6+



Important to note: the heated continuum - many tens of eV, can further collisionally ionise the system. No K-α will be seen if the K-edge energy of an ion exceeds the photon energy of the X-Ray Laser.

K-shell spectroscopy of Hot Dense Aluminium



Nature ***482***, *59 (2012)*

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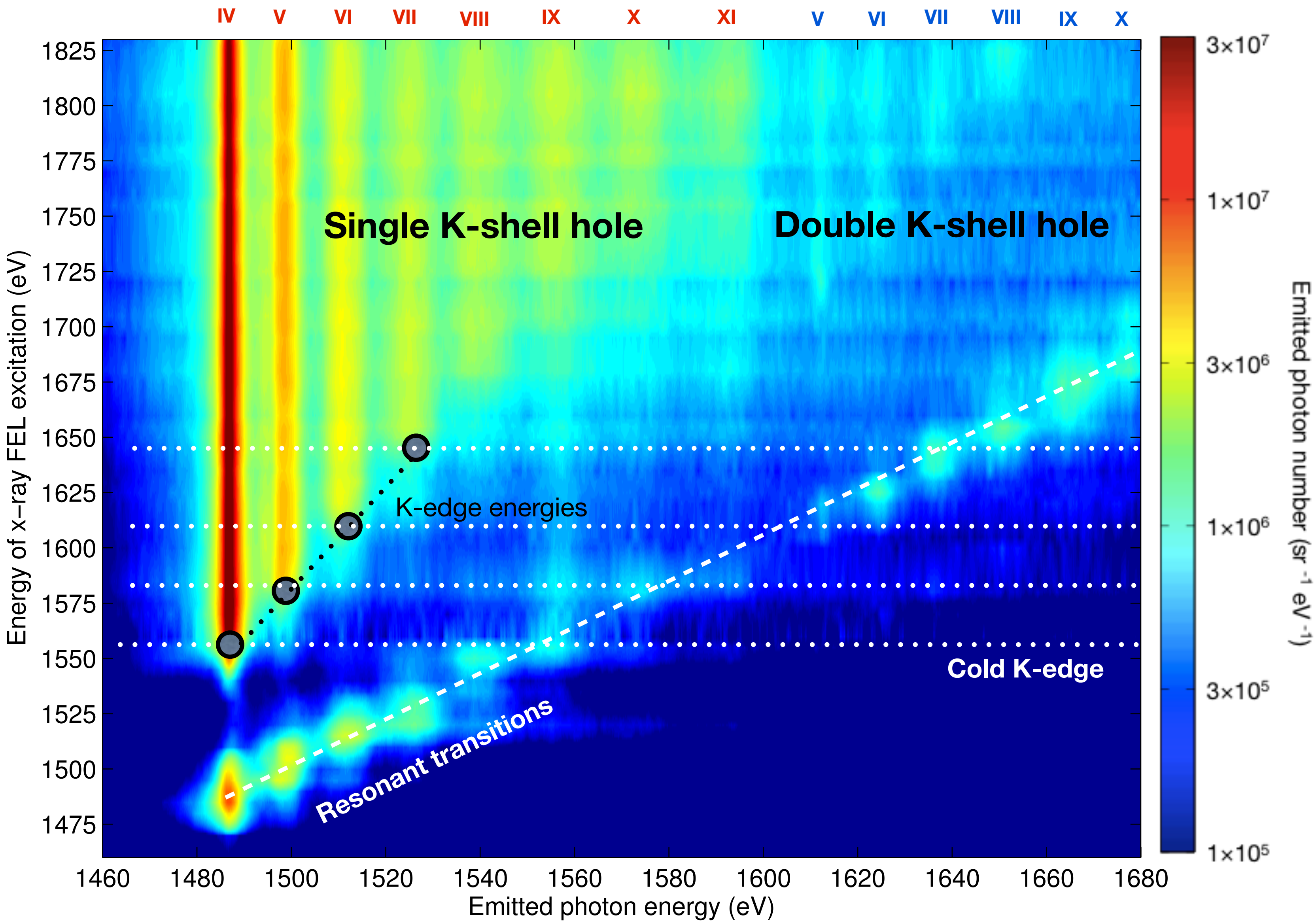
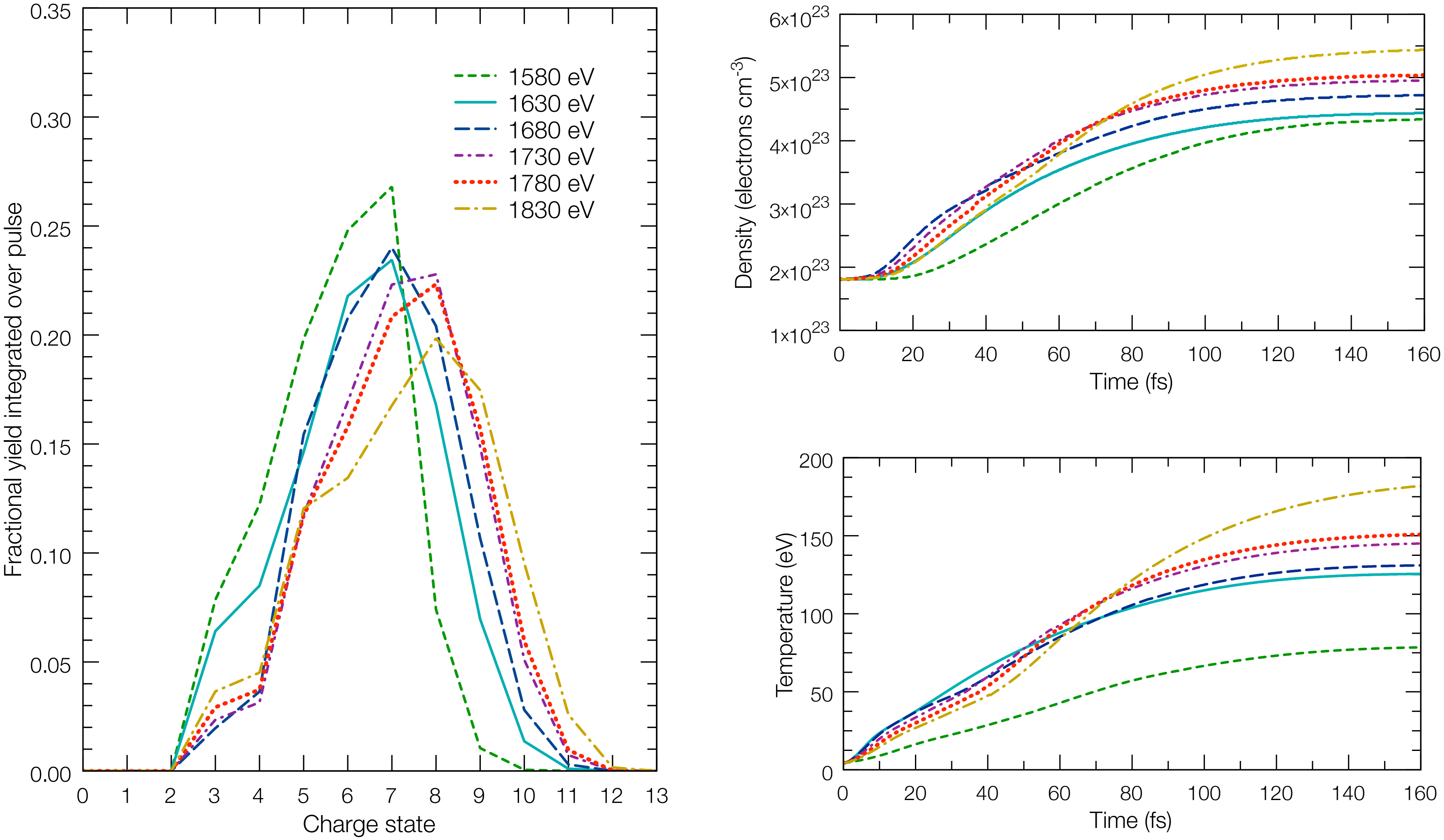


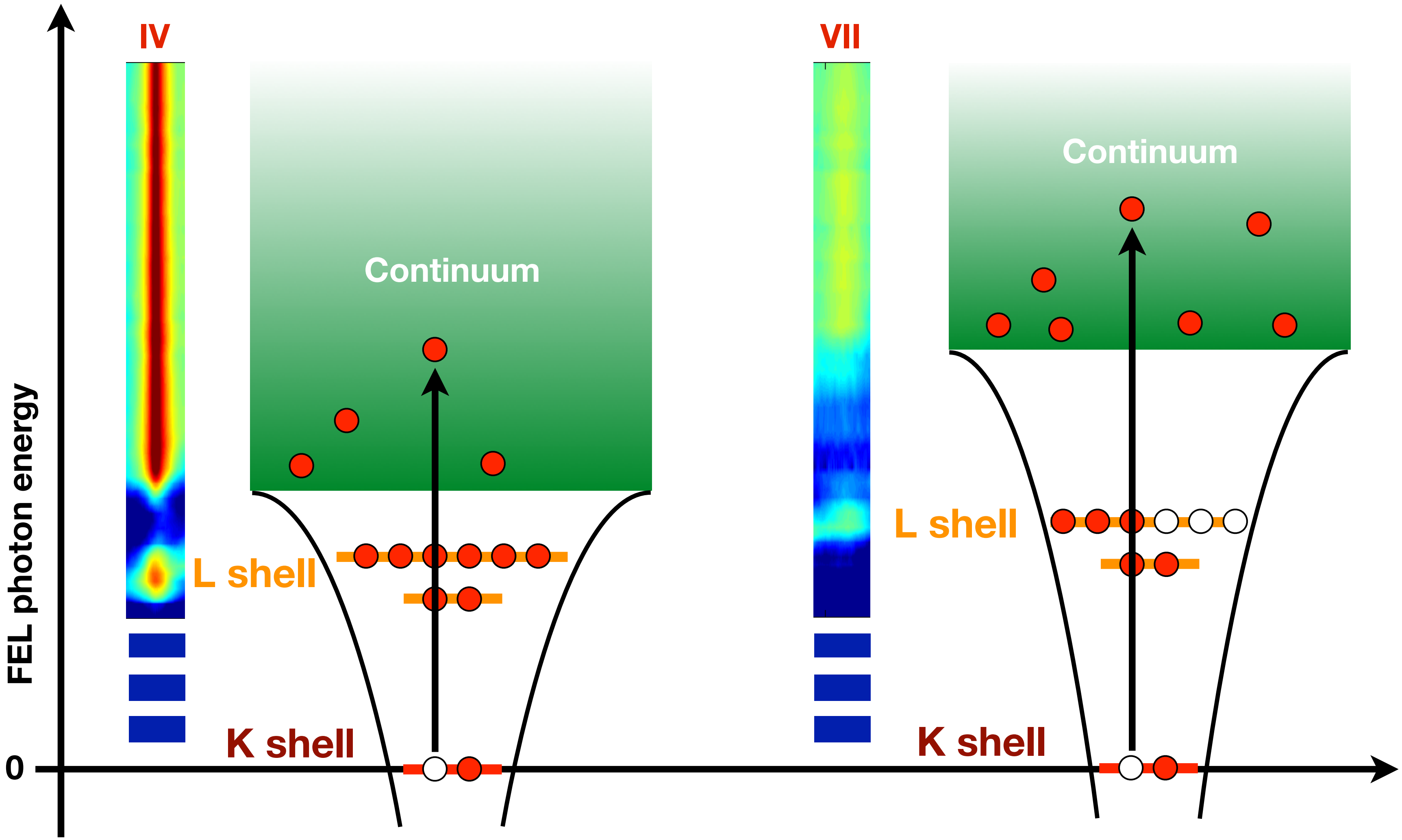
Nature ***482***, *59 (2012)*

Isochoric heating: plasma evolution



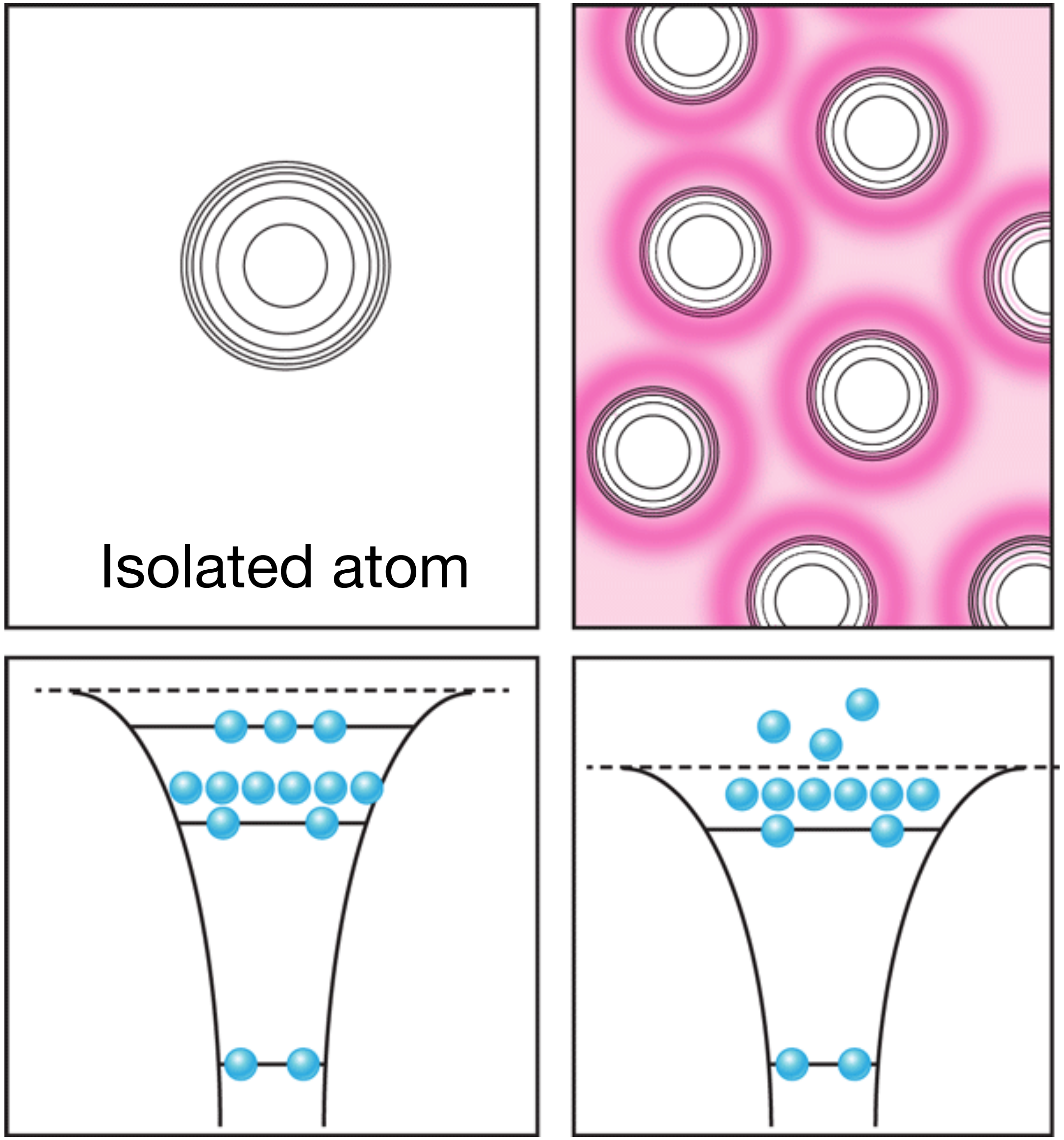
The FEL reveals charge states when its energy exceeds the K-edge - the charge state distribution itself does not vary that greatly with photon energy.

K-shell spectroscopy of Hot Dense Aluminium



We observe very clean absorption thresholds

Continuum lowering in dense plasmas



In dense systems at some radius outer orbitals may overlap – these can no longer be considered bound to a specific ion = ionised.

This means the energy required to ionise a bound state is reduced as the density increases:

Ionization Potential Depression (IPD)

Figure taken from
Umstadter, Physics **5**, 88 (2012)

Some simple Ionization Potential Depression models

- Analytical models used when fast calculations required (atomic kinetics, hydrodynamic codes, etc.):
  - Ion-sphere (compute total energy of free electron in Wiegner-Seitz ion sphere, originates from condensed matter theory):

$$\Delta I_{\mathrm{IS}} = \frac{3}{2}\frac{ze^2}{4\pi\epsilon_0 r_{\mathrm{SP}}}$$

$$\frac{4\pi}{3} r_{\mathrm{SP}}^3 = n_i^{-1}$$

  - Debye Hückel (calculate electrostatic energy of electron/ion + Debye cloud, works in weak-coupling):

$$\Delta I_{\mathrm{DH}} = \frac{ze^2}{4\pi\epsilon_0 \lambda_{\mathrm{D}}}$$

  - Stewart & Pyatt model, 1996, used in almost all atomic kinetics models (bridges between the two above):

$$\Delta I_{\mathrm{SP}} = \frac{k_{\mathrm{B}}T}{2(z^*+1)}\left\{\left(\frac{3(z^*+1)ze^2}{4\pi\epsilon_0\lambda_{\mathrm{D}}k_{\mathrm{B}}T}+1\right)^{2/3}-1\right\}$$

  - Ecker & Kröll model, 1963, (different Z scaling, matches LCLS data)

$$\Delta I_{\mathrm{EK}} = C\frac{ze^2}{4\pi\epsilon_0 r_{\mathrm{EK}}}$$

$$\frac{4\pi}{3} r_{\mathrm{EK}}^3 = \frac{1}{n_e + n_i}$$

Some simple Ionization Potential Depression models

- Analytical models used when fast calculations required (atomic kinetics, hydrodynamic codes, etc.):
  - Ion … es from co
  - De … eak- co
  - Ste

Based on Debye screening: at solid density there is less than 1 electron per Debye sphere!

$$\Delta I_{\mathrm{SP}} = \frac{k_{\mathrm{B}}T}{2(z^*+1)}\left\{\left(\frac{3(z^*+1)ze^2}{4\pi\epsilon_0\lambda_{\mathrm{D}}k_{\mathrm{B}}T}+1\right) - 1\right\}$$

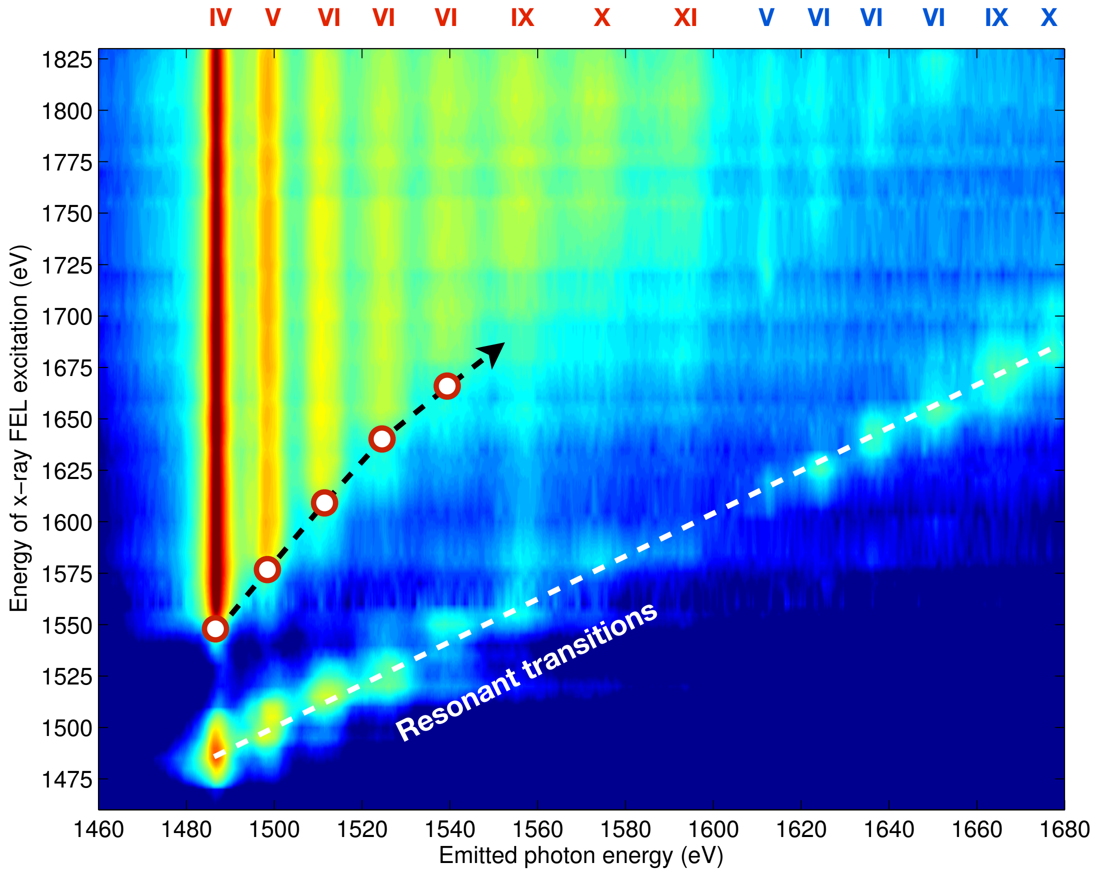
  - Ecker & Kröll model (different Z scaling, matches LCLS data)

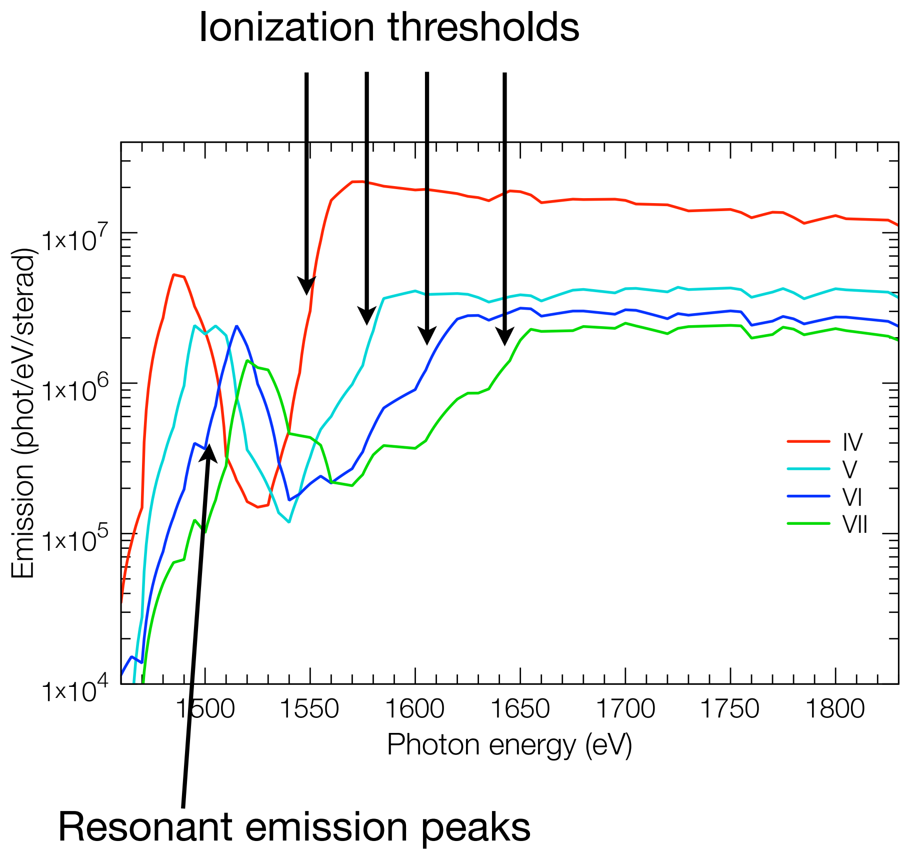
$$\Delta I_{\mathrm{EK}} = C\frac{ze^2}{4\pi\epsilon_0 r_{\mathrm{EK}}}$$

$$\frac{4\pi}{3}r_{\mathrm{EK}}^3 = \frac{1}{n_e+n_i}$$

Preston et al., HEDP **9**, 258 (2013)

Experimental measurement of IPD on LCLS



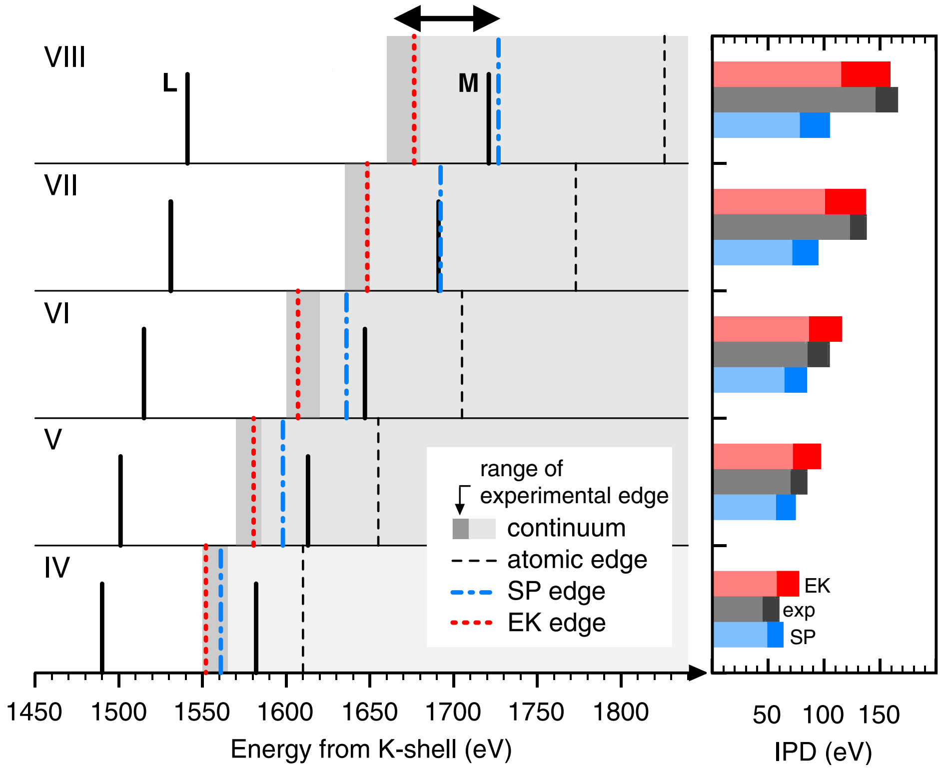


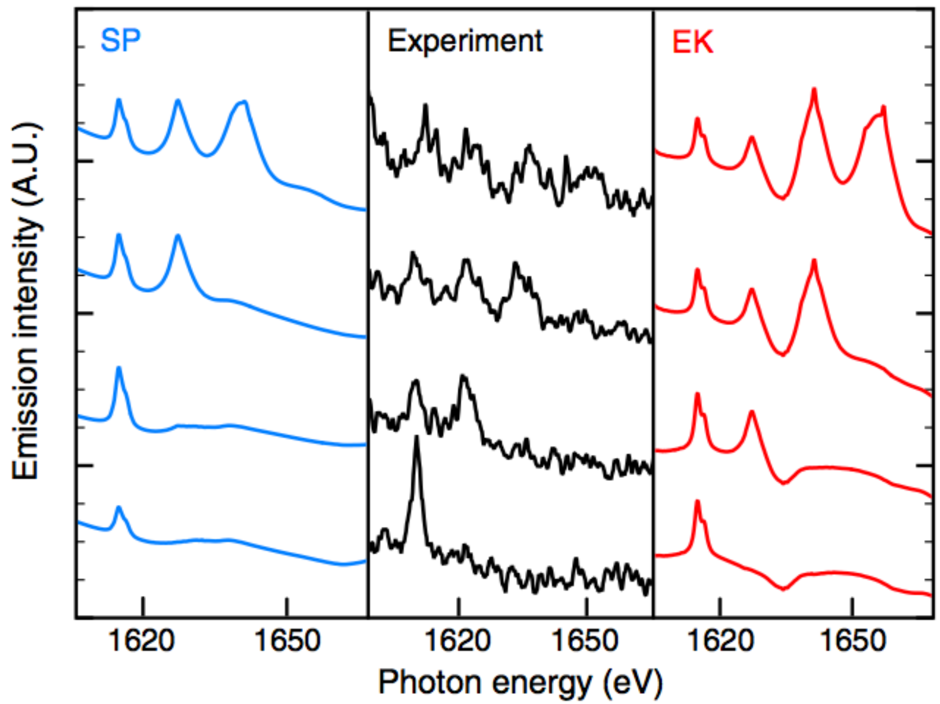
If we can identify these thresholds with K-edges, then:

Experimental IPD = Atomic edge - Measured edge

Experimental measurement of IPD on LCLS

~50 eV!





Single core holes

Double core holes

Note: M-shell always in continuum according to EK model.

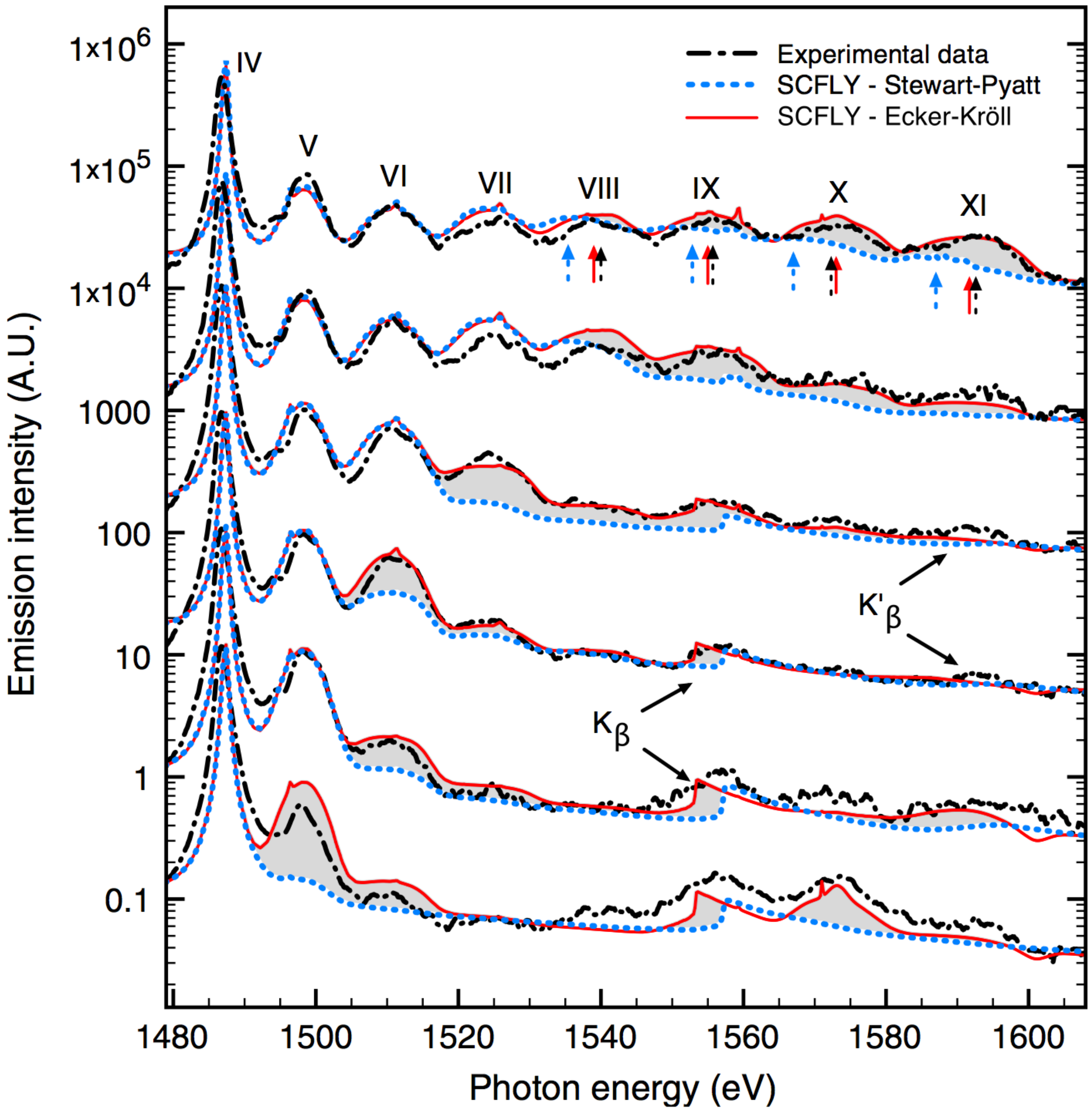
Note: we do NOT claim EK model is ‘correct’, only that it fits this case. It is also a crude semi-classical model.

Phys. Rev. Lett, ***109***, *065002 (2012)*

Modelling the date via collisional-radiative super-configuration code SCFLY

- Proven to work in modelling intense X-ray interaction with atoms in the collision-less regime
- **All** super-configurations up to n=3 included with appropriate degeneracy
- Self-consistent temperature calculated via X-ray deposition within duration of pulse
- Spectral synthesis via a super-configuration transition array model
- Included opacity via escape factor formalism, IPD via IS, SP and EK models
- We perform simulations at 24 intensities, and weight them according to experimentally measured profiles of the x-ray focal spot.
- There is no fit to the data - we simply run the code for the experimental photon energy and intensities, and simulate the spectrum.
- The only adjustable part is the model for ionisation potential depression

Continuum lowering results on LCLS



LCLS pump energy:

1830 eV

1720 eV

1650 eV

1630 eV

1600 eV

1580 eV

Ciricosta et al., PRL **109**, 065002 (2012)

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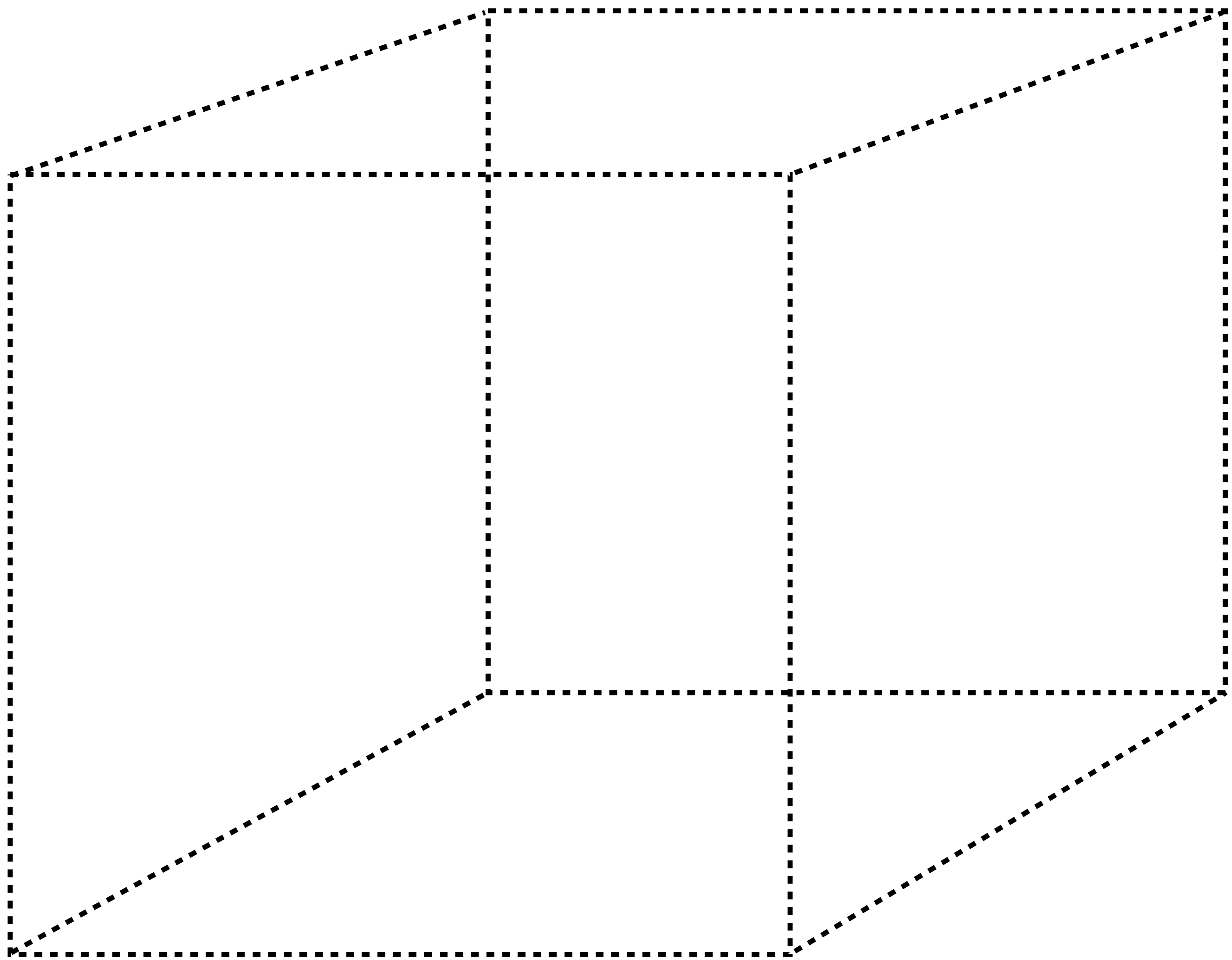
Continuum lowering via Density Functional Theory

- There is a fundamental problem with the way we think of continuum lowering
  - cannot explain the experimental observations: EK works only in some cases, SP only in others
  - we seem to be missing some physics

- Treatment of shells near continuum (M-shell) in analytical models is very poor
  - models need a sharp cutoff to what is physically a continuous process
  - states within a shell not always treated individually
  - we should not need to artificially separate states into ‘bound’ and ‘free’

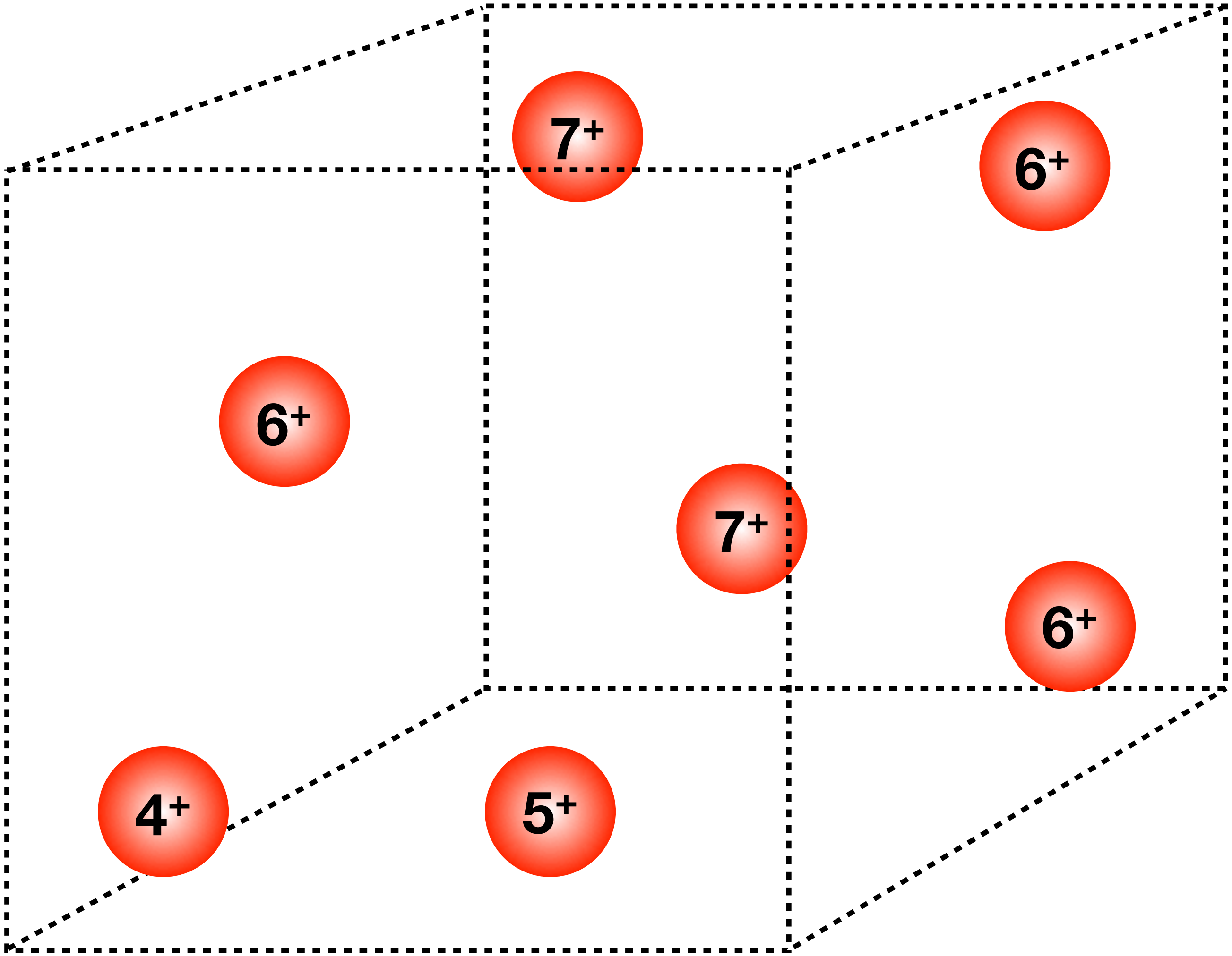
- Lets try a different approach that we know works for ground-state metals:
  - Calculate IPD independent of a specific analytical model
  - Want the results to be consistent with relevant atomic physics and thermodynamics
  - Applicable to the plasma conditions reached on LCLS
  - Extend beyond the average atom picture, and we do NOT assume spherical symmetry.

Continuum lowering via DFT - simulation box



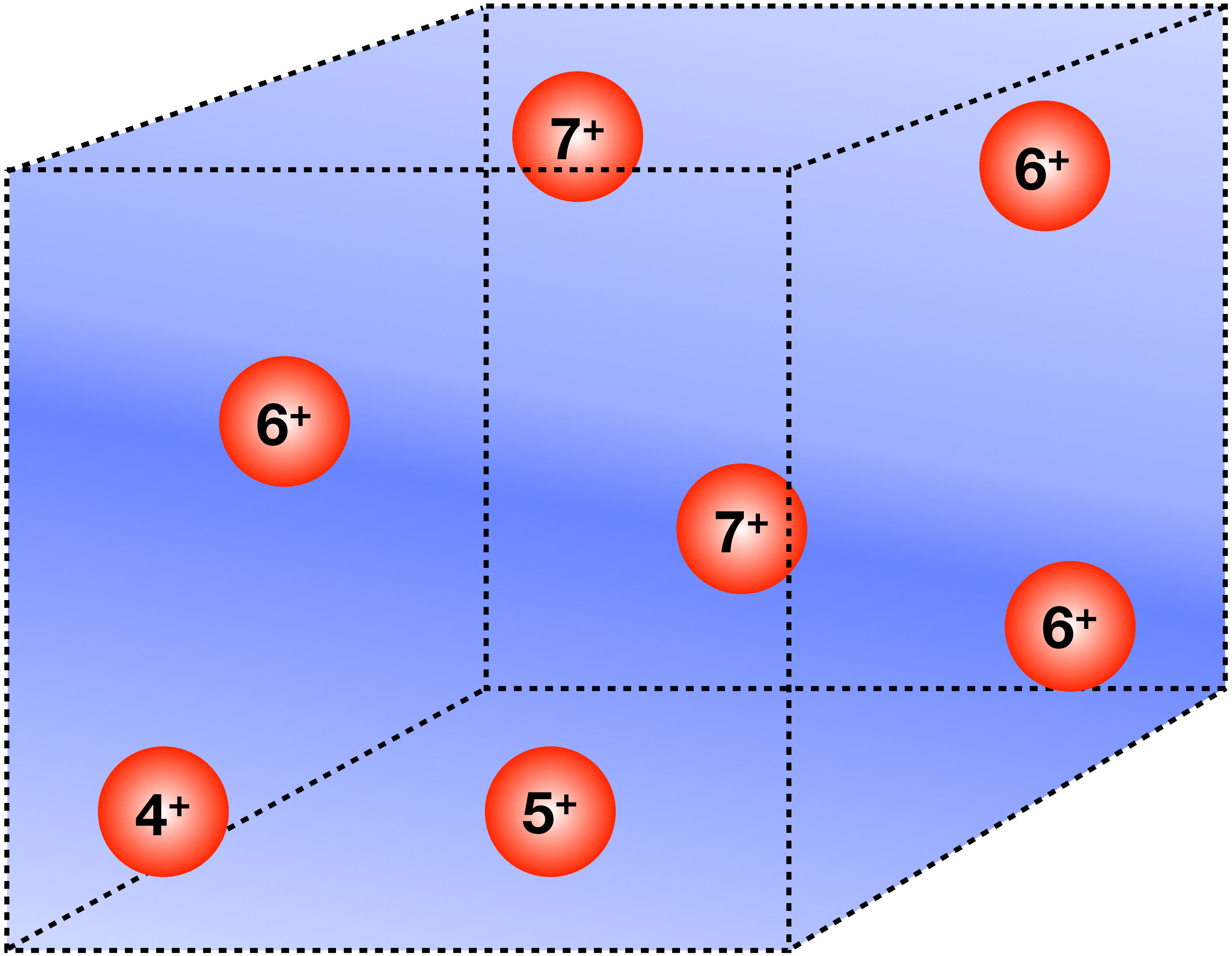
Continuum lowering via DFT - add the ions

PAW Potentials to simulate ion core + inner shell bound electron states



Continuum lowering via DFT - add electrons

Box should be charge neutral (here we need 41 electrons)



Find the free-electron density via finite temperature DFT

Excited state PAW potentials

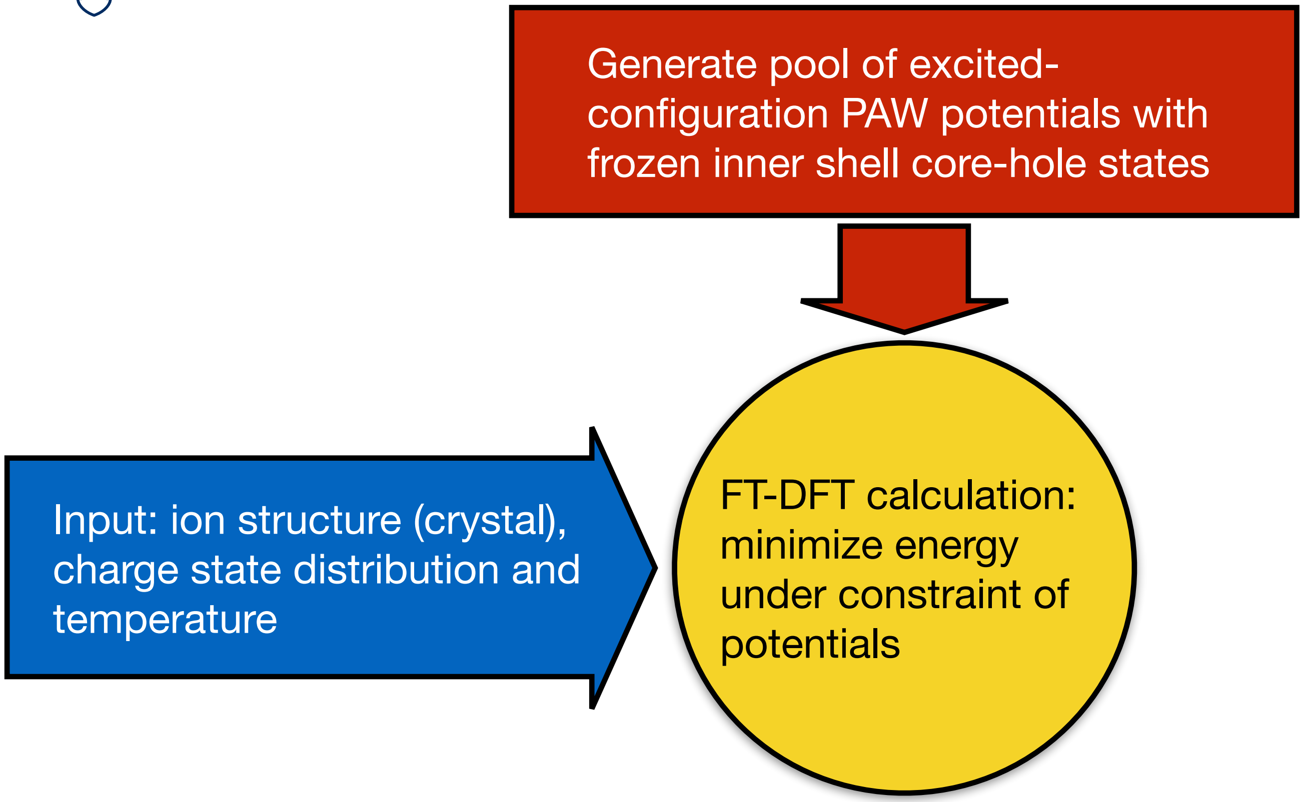
- Core-excited (rather than ionised) = global charge neutrality of the system

- PAW (projector augmented wave) formalism is key
  - allows for frozen-core, all-electron potentials
  - can calculate all core wave functions for excited atomic configurations to whatever accuracy required
  - can freeze holes in core states – allows for a fully 3D multi-centred approach of charge states which are **integers** (no average atom approximation for ionization)
  - charge state **independent** of temperature/density: can model **equilibrium** (pick the right temperature for the mean chosen ionization) or non-equilibrium systems (in terms of the ionization), includes some **fluctuations** (ensemble of integer charge states is simulated directly),
  - can reconstruct the "real" valence density everywhere, including on ionic cores, where overlap with core wave functions becomes relevant. No spherical symmetry assumed.

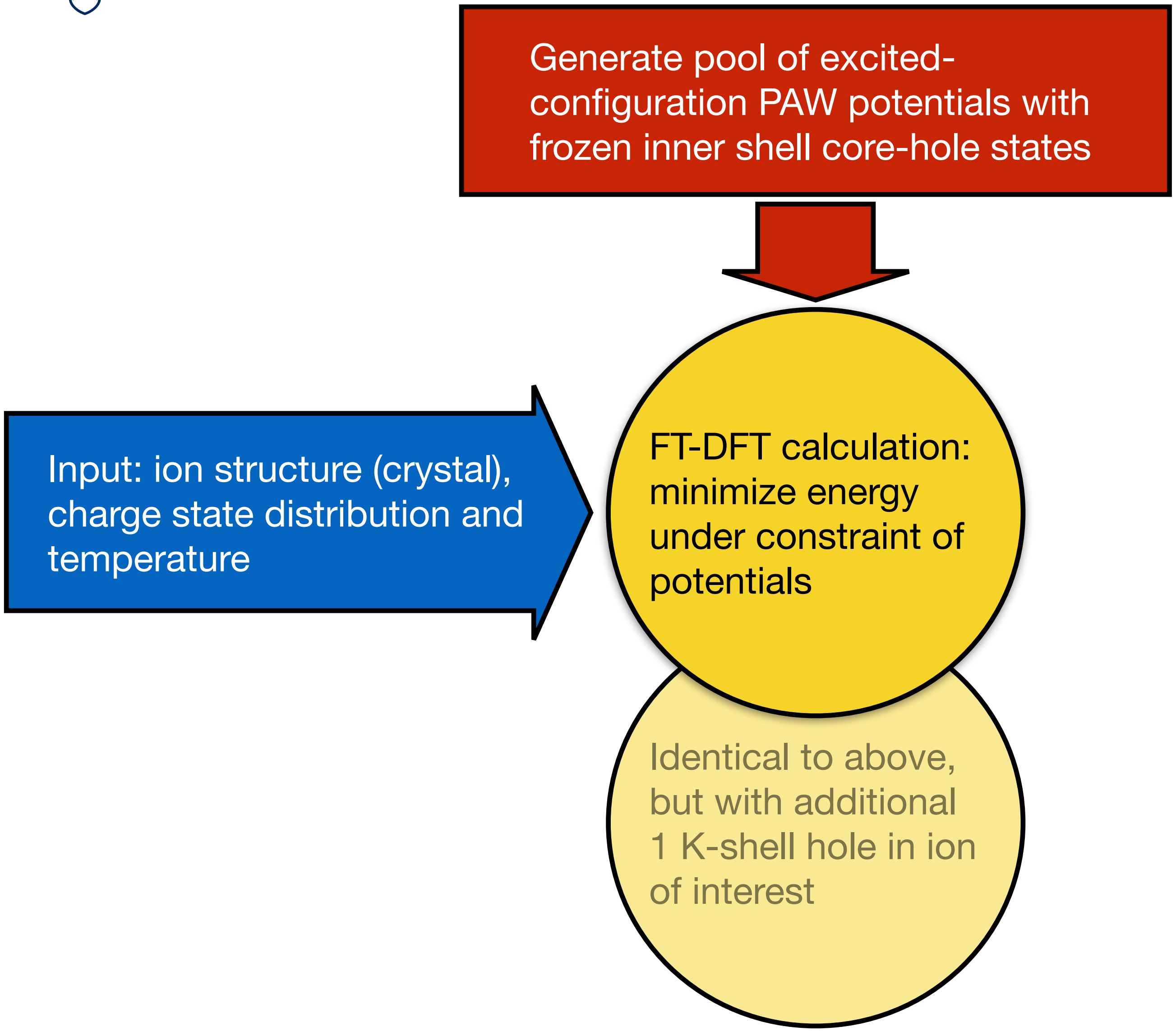
Calculate the IPD from first principles - Roadmap

Generate pool of excited-configuration PAW potentials with frozen inner shell core-hole states

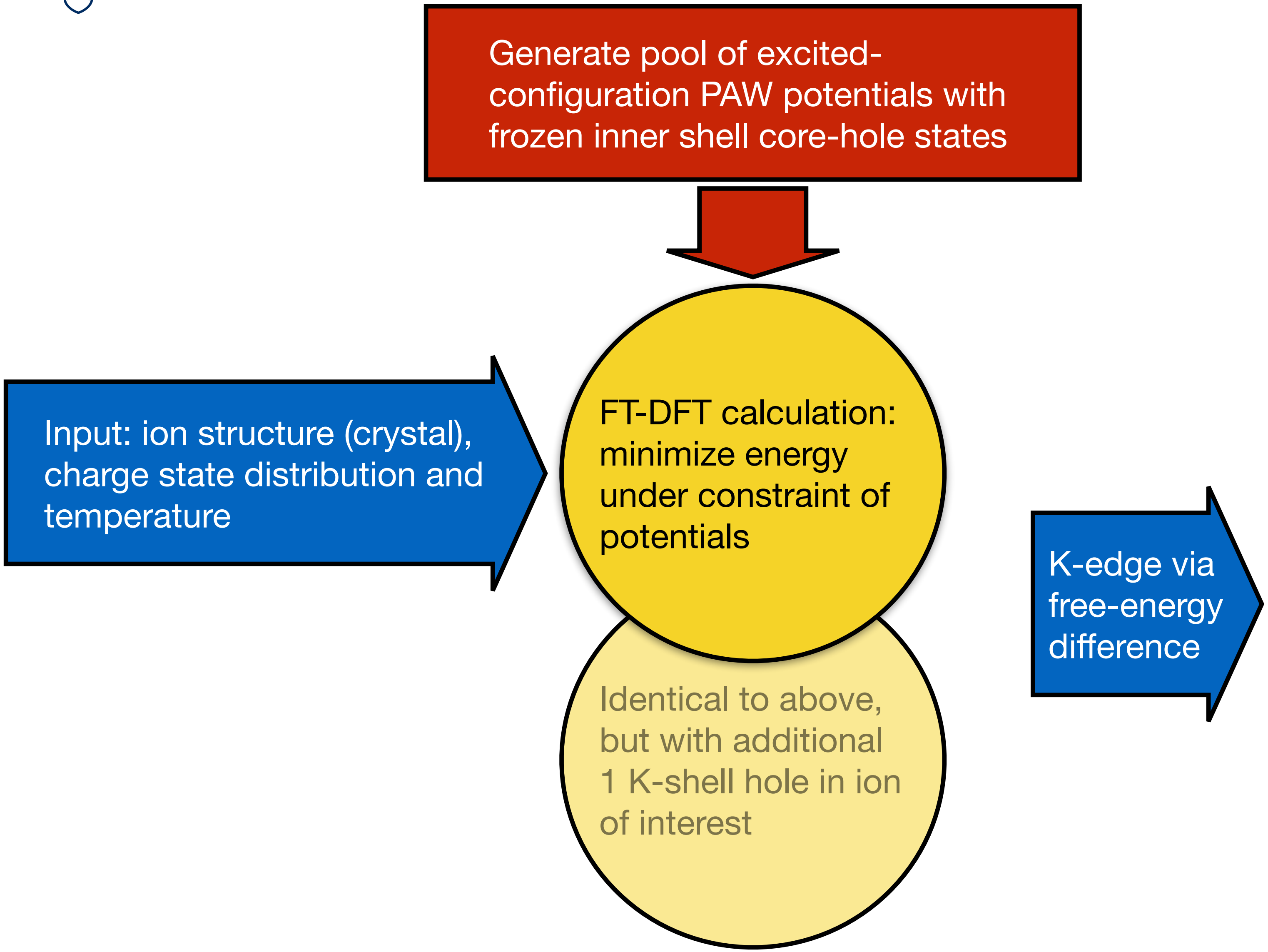
Calculate the IPD from first principles - Roadmap



Calculate the IPD from first principles - Roadmap

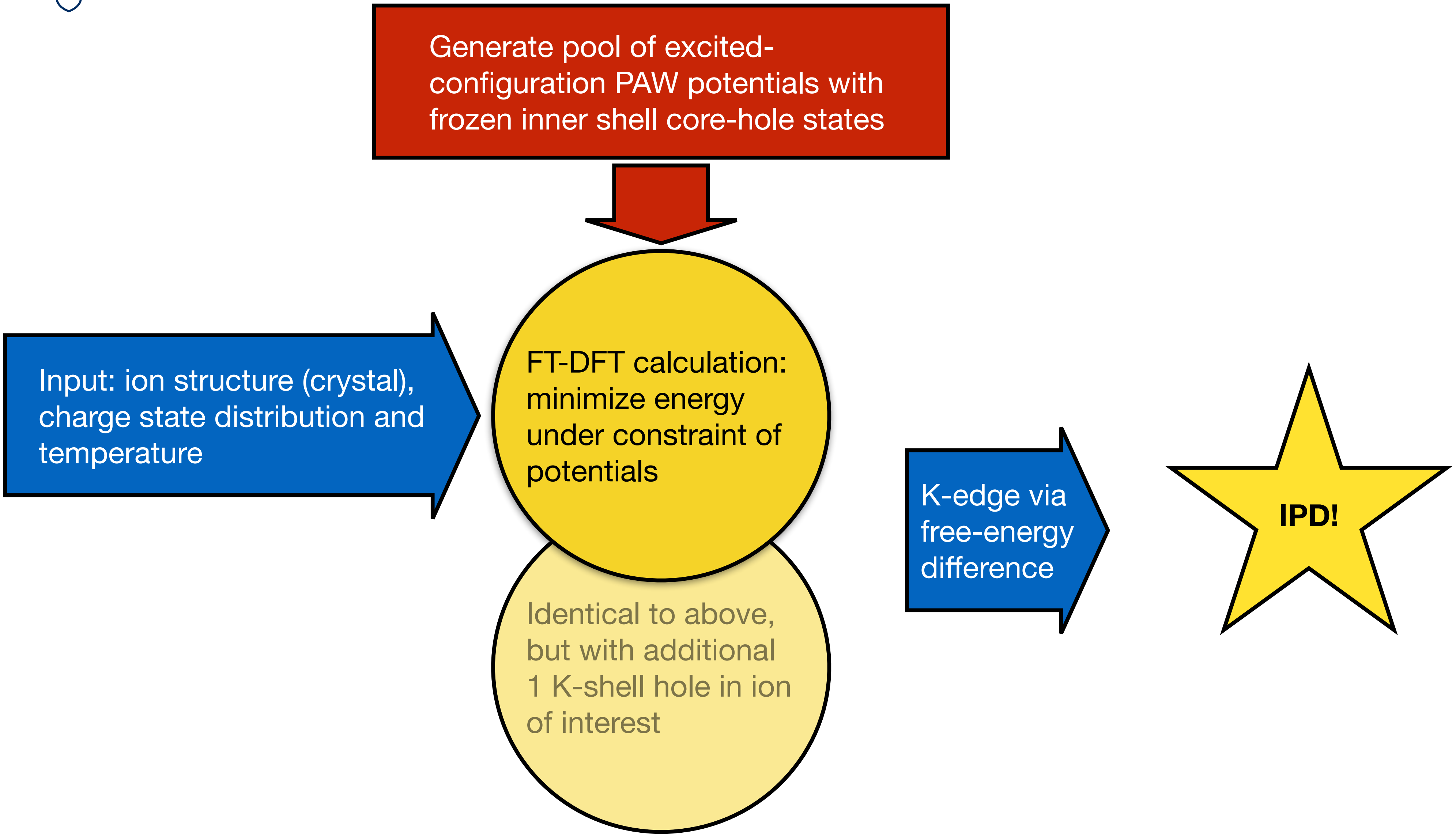


Calculate the IPD from first principles - Roadmap



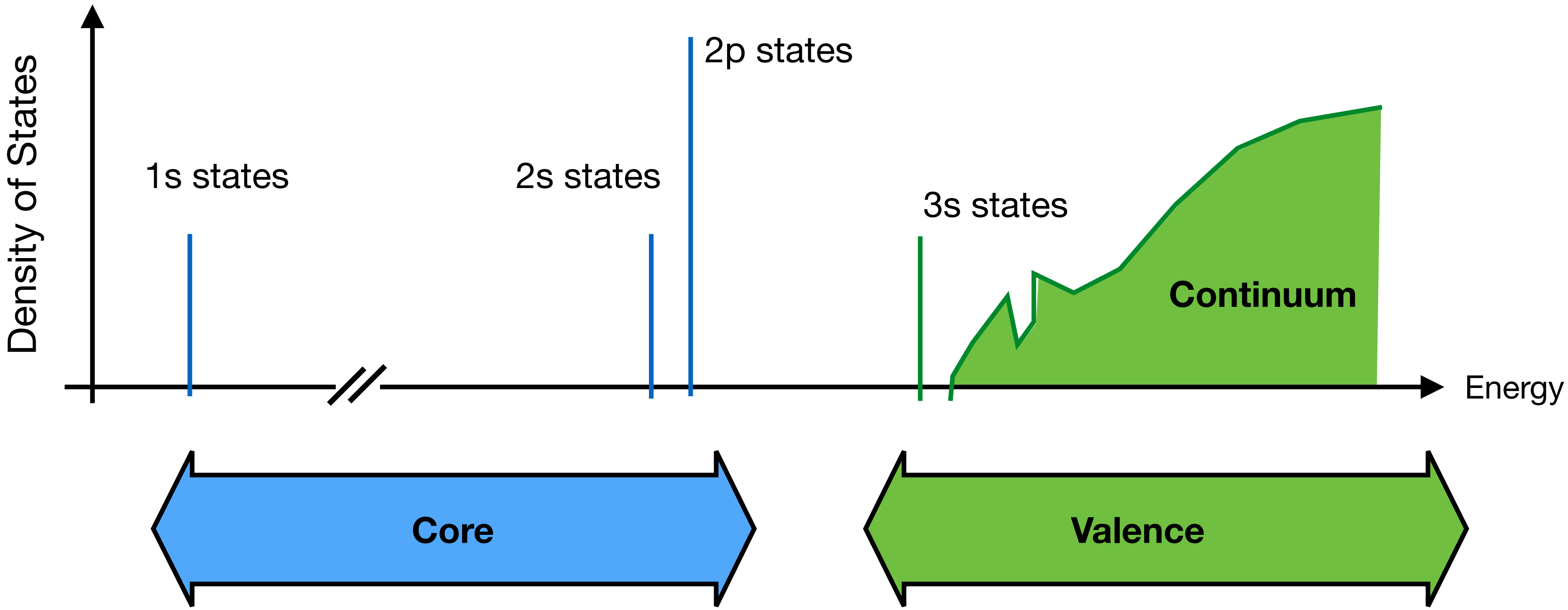
Vinko, Ciricosta & Wark, Nature Comm **5**, 3533 (2014)

Calculate the IPD from first principles - Roadmap



Vinko, Ciricosta & Wark, Nature Comm **5**, 3533 (2014)

What do we calculate?



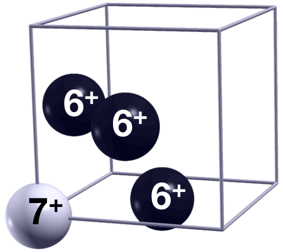
Populations selected in creation of the potential
Calculated in the single atom limit (radial DFT, Hartree-Fock-Dirac, etc.)

Determined from 3D DFT calculation: the lowest energy configuration possible given our choice of the core (still formally ground-state)

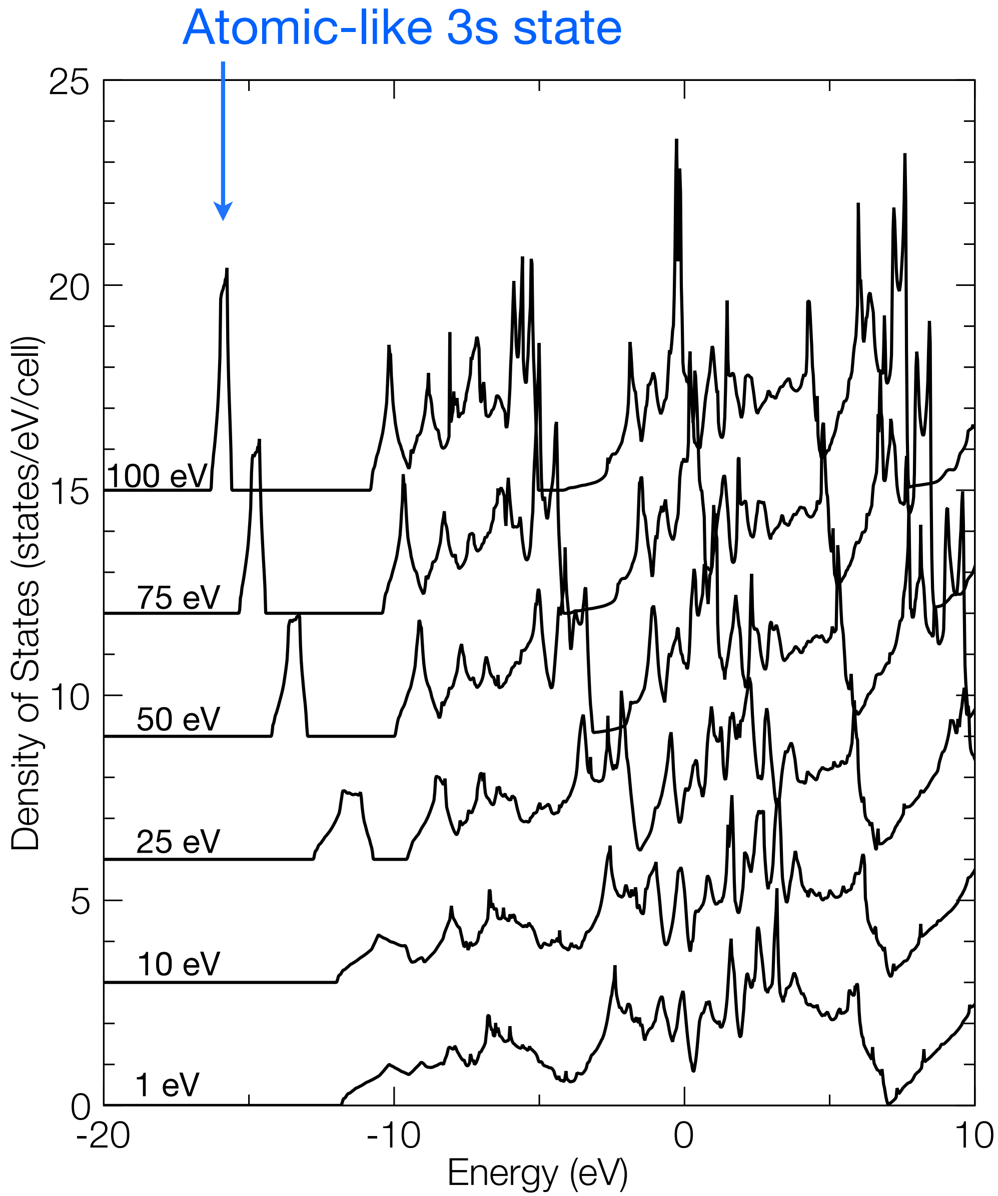
Populations determined by chosen temperature via the Fermi-Dirac distribution

Al continuum structure at different temperatures

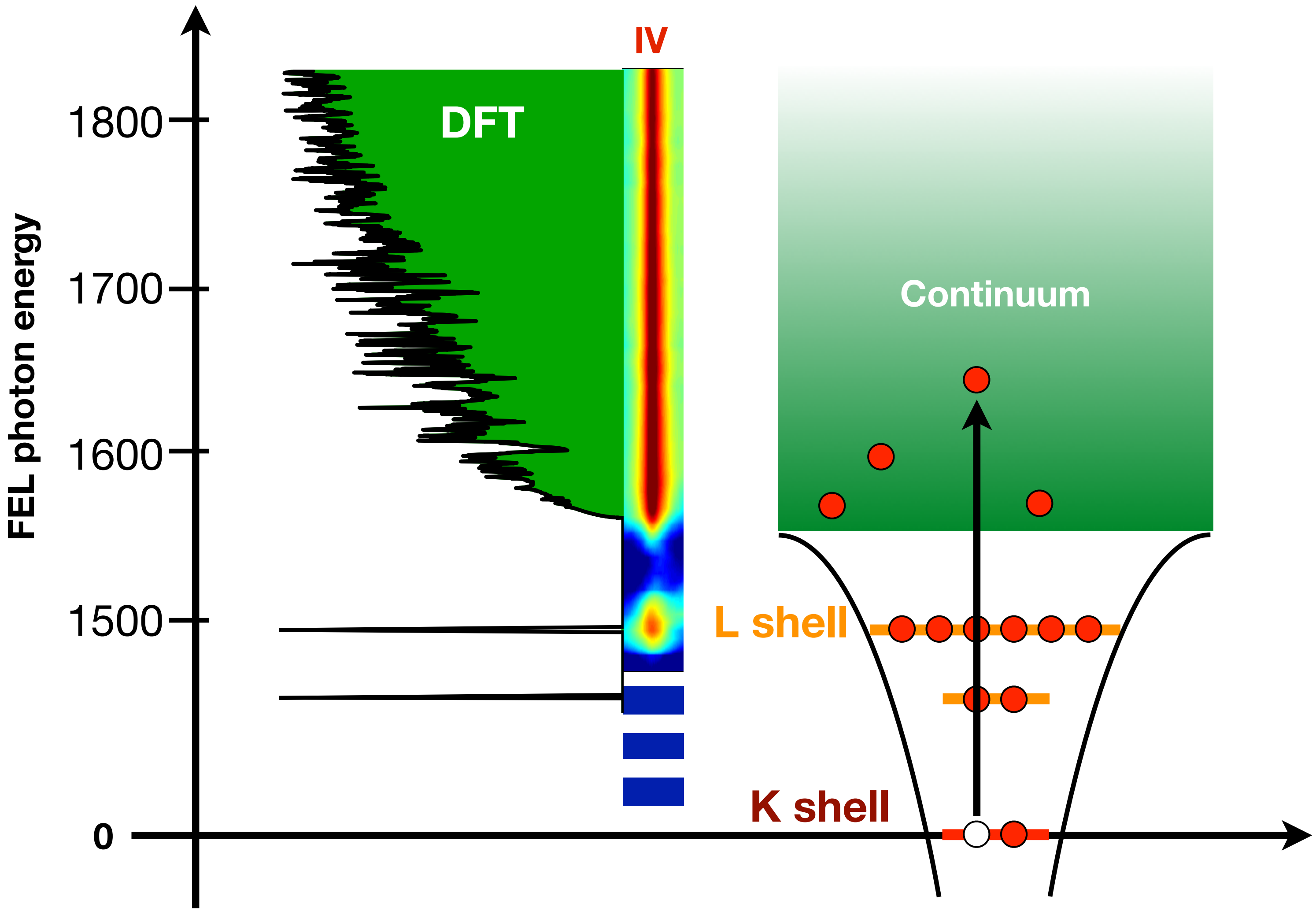
Calculation box:



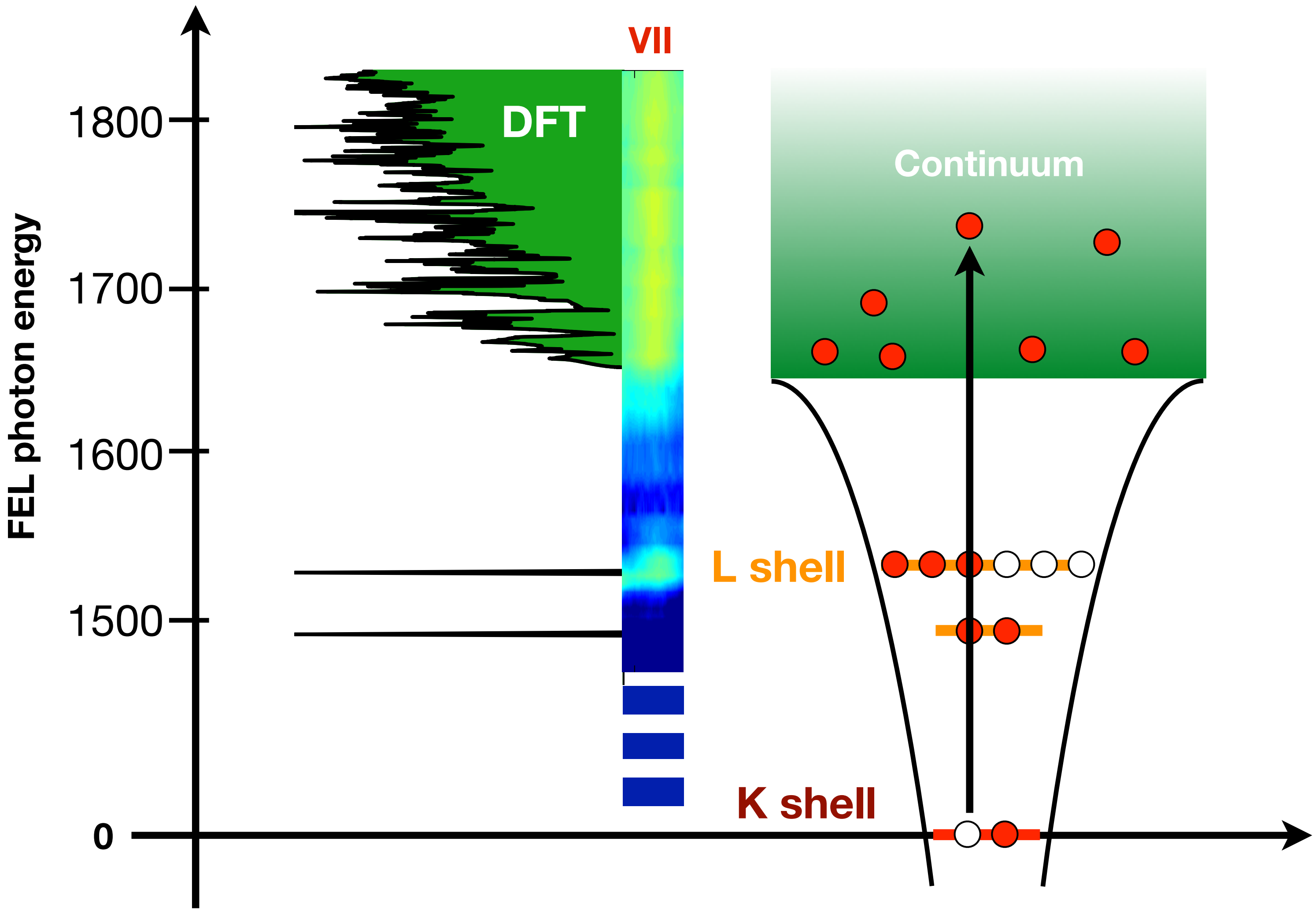
(Electron density 3.75 x10^{23} cm^{-3})



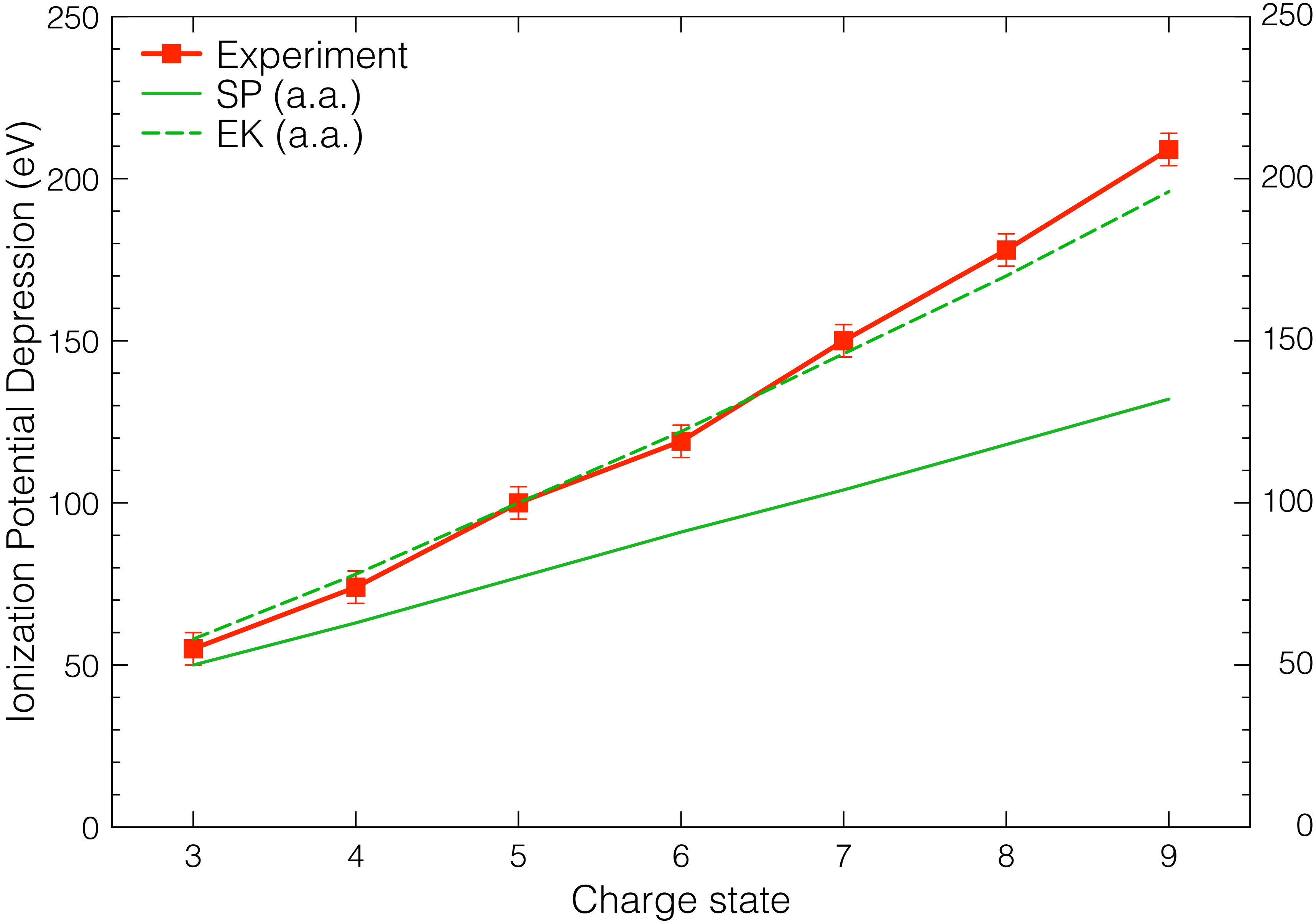
K-shell spectroscopy of Hot Dense Aluminium: **3+**



K-shell spectroscopy of Hot Dense Aluminium: **6+**



Comparison of calculations with experiment: Aluminium



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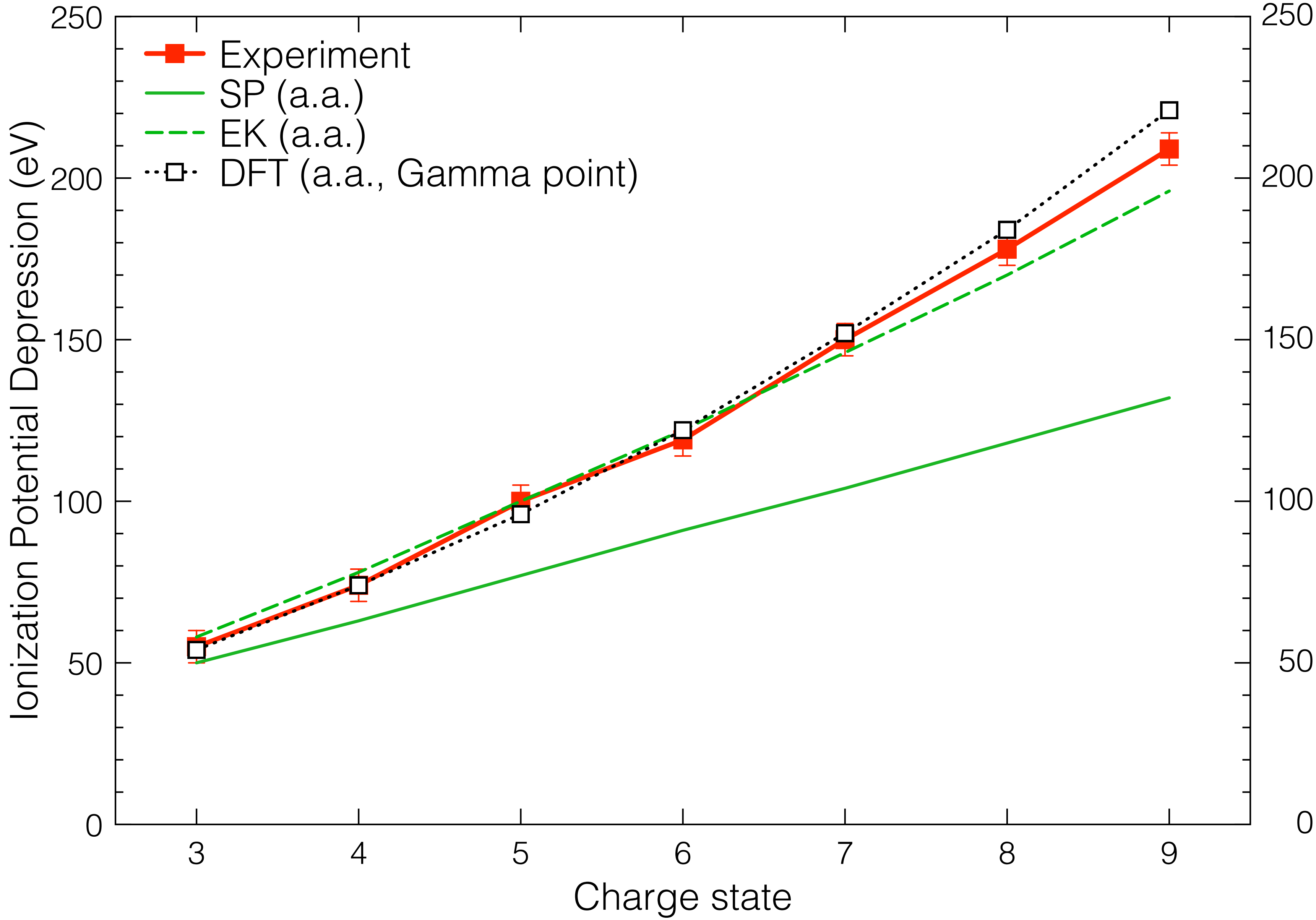

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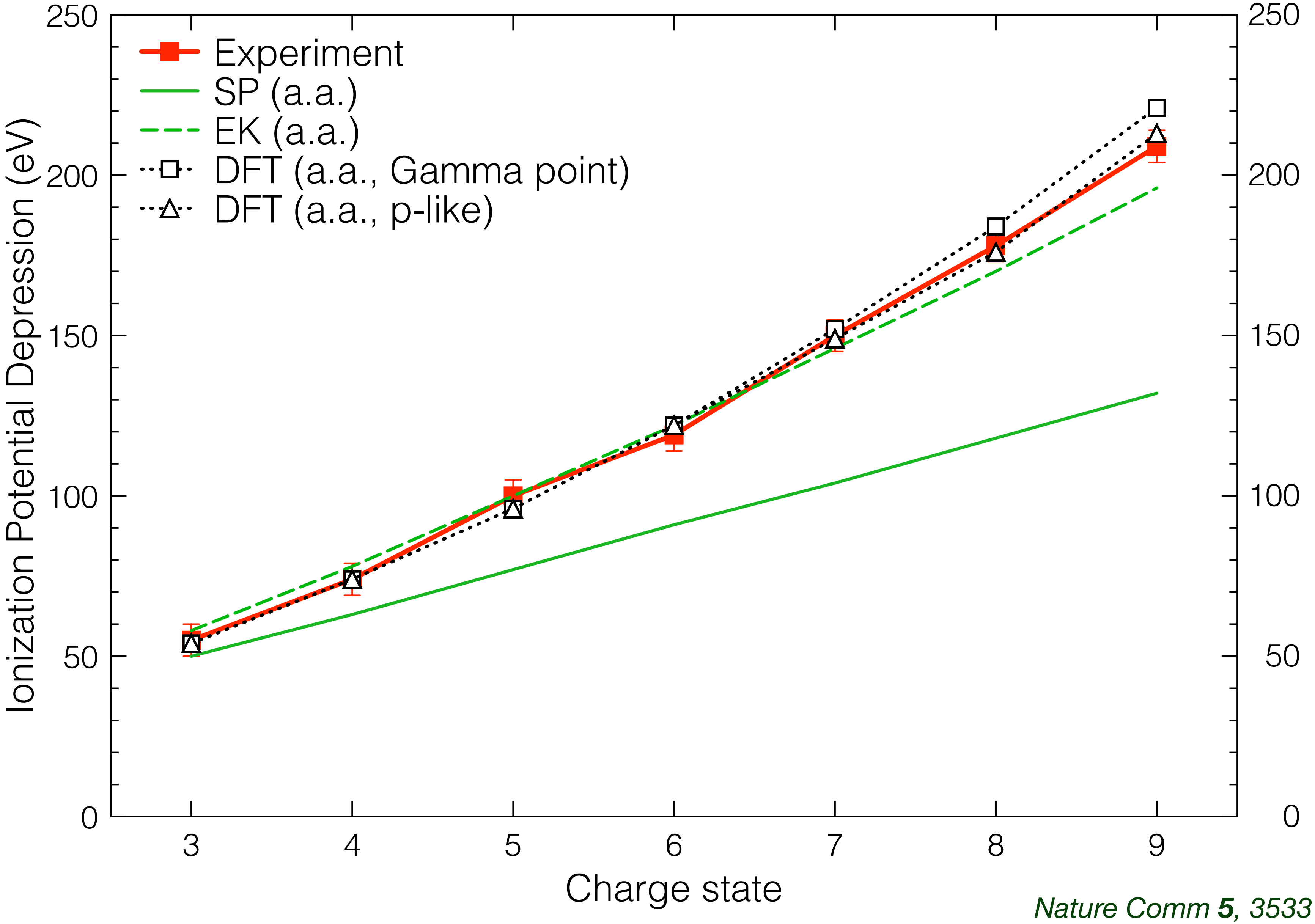
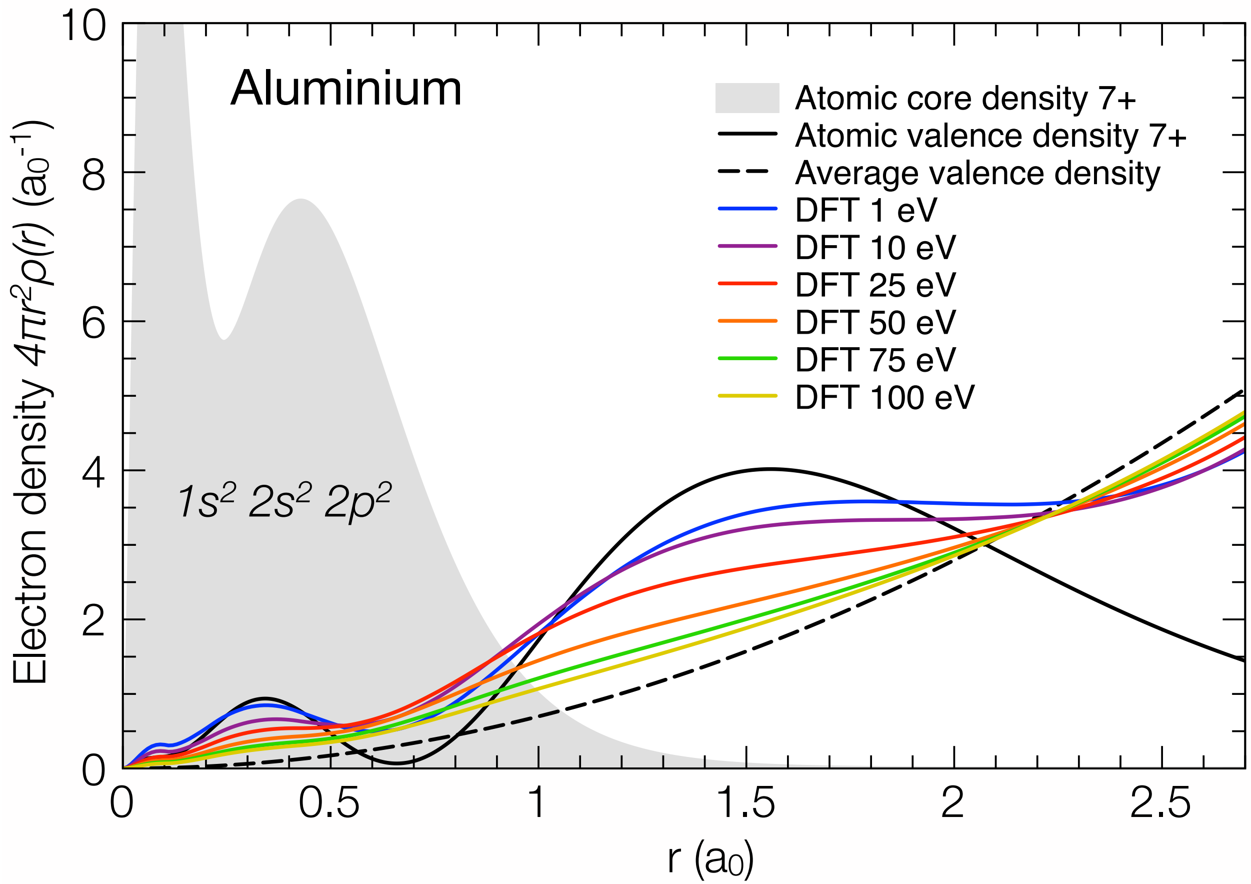


Nature Comm ***5****, 3533 (2014)*

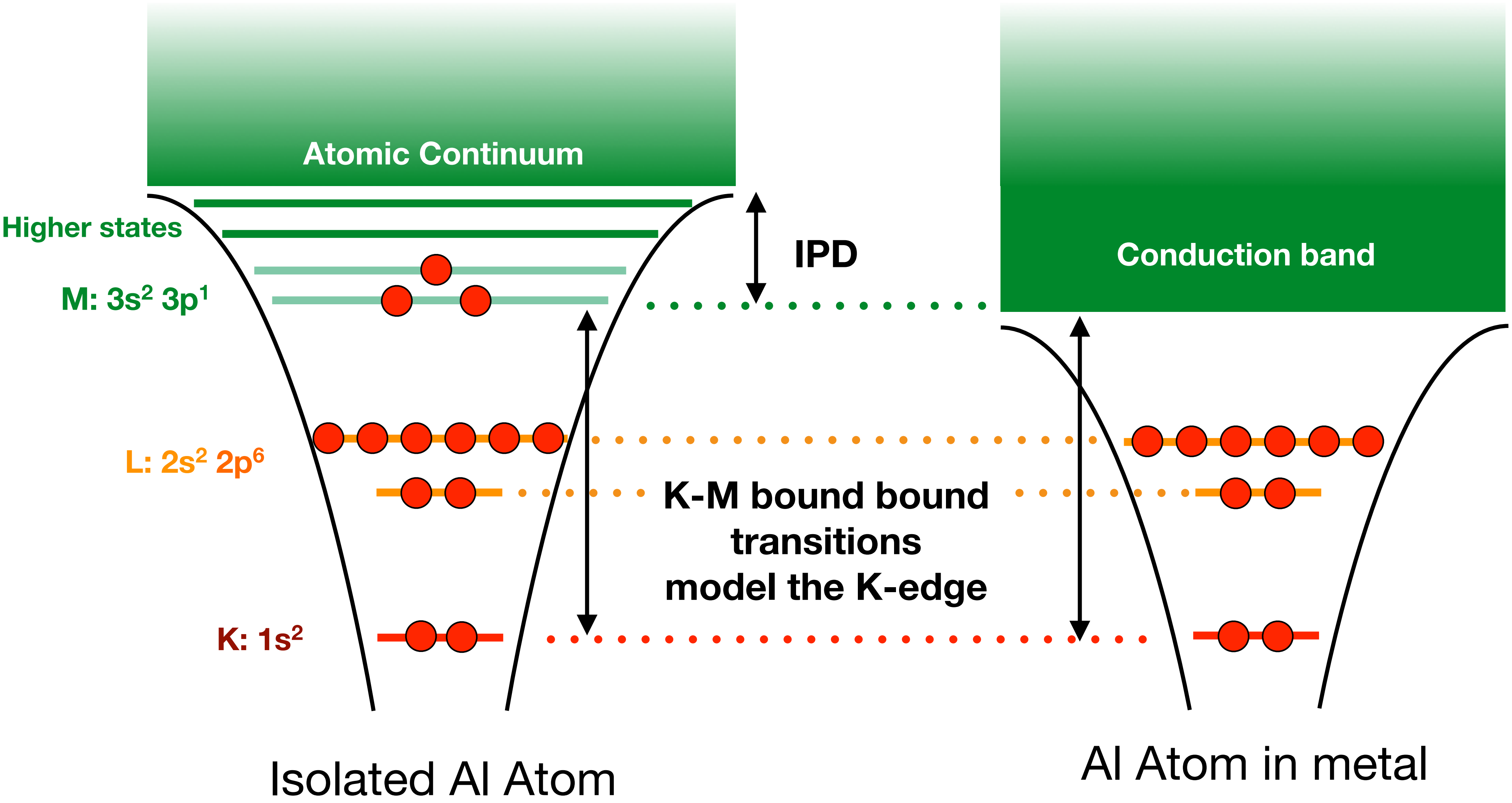
DFT method reproduces experimental IPDs

Electron interactions with ions and other electrons remains strong in the conditions generated on LCLS. **At solid density, continuum states are formed due to the interaction of M-shell states which form the conduction band (tight-binding picture).**



Vinko, Ciricosta & Wark, Nature Comm **5**, 3533 (2014)

Atomic bound-bound transitions can be a good approximation for bound-free edge



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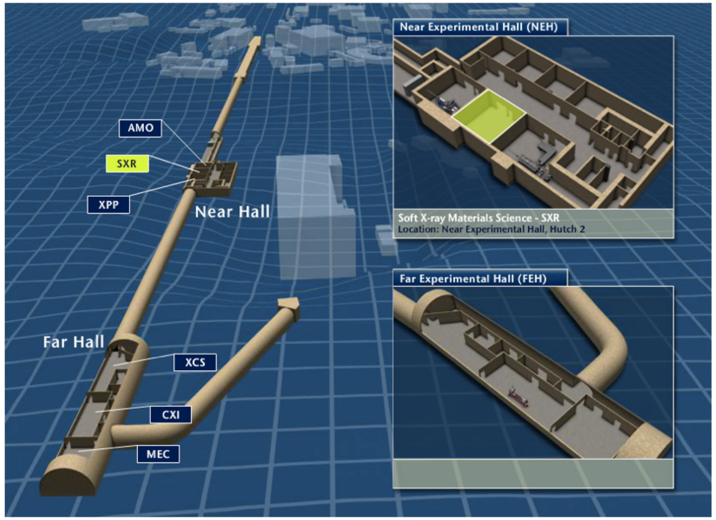
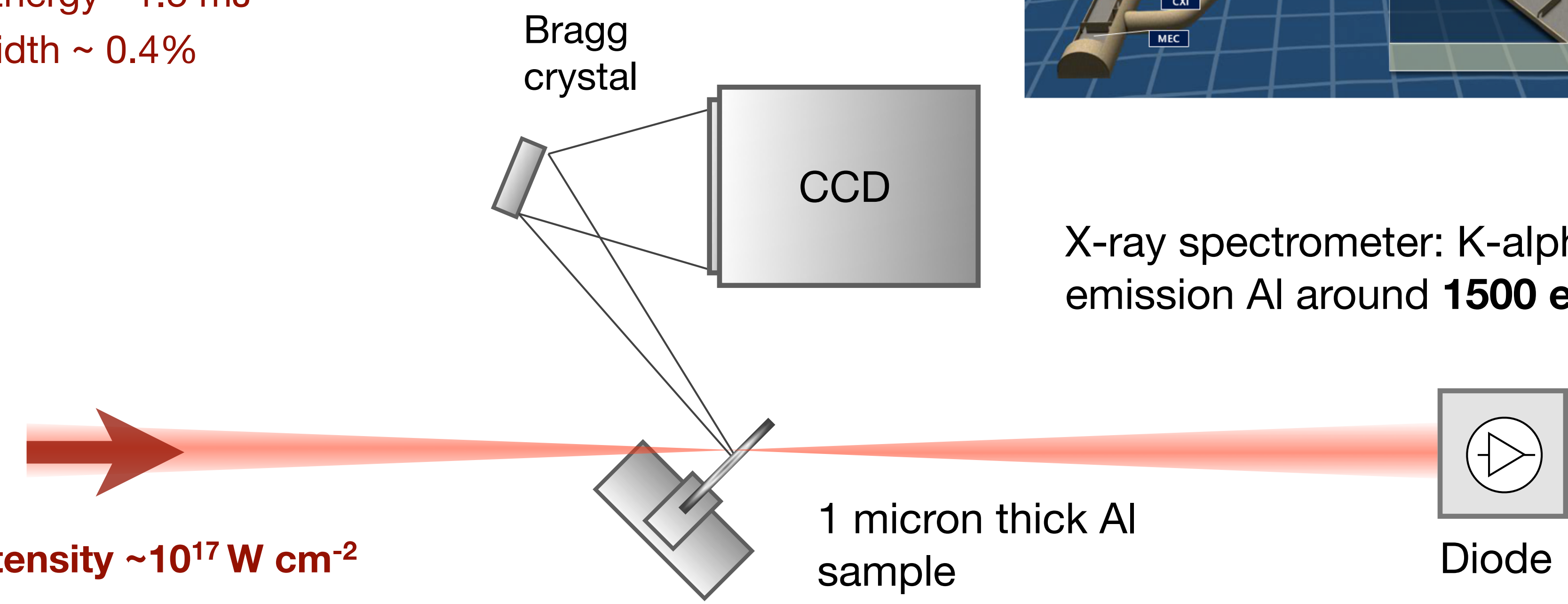
The experiment at the Linac Coherent Light Source X-ray Free-Electron Laser



LCLS pulse

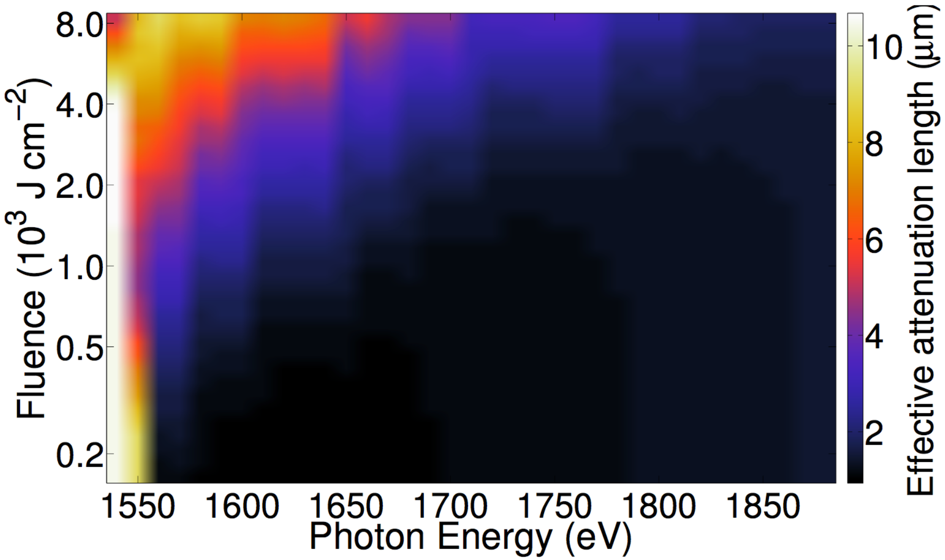
Photon energy: 1460–1830 eV
Pulse length < 60 fs
Pulse Energy ~1.5 mJ
Bandwidth ~ 0.4%

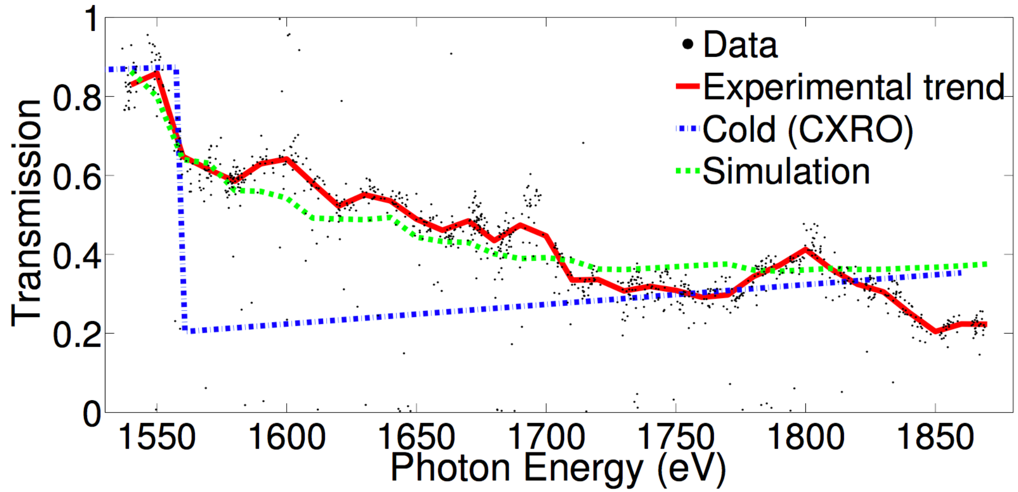


Peak Intensity ~10^{17} W cm^{-2}

Vinko et al., Nature **482**, 59 (2012)
Ciricosta et al., PRL **109**, 065002 (2012)

Saturable absorption at 1.5 - 1.8 keV ~ 10^{17} Wcm^{-2}





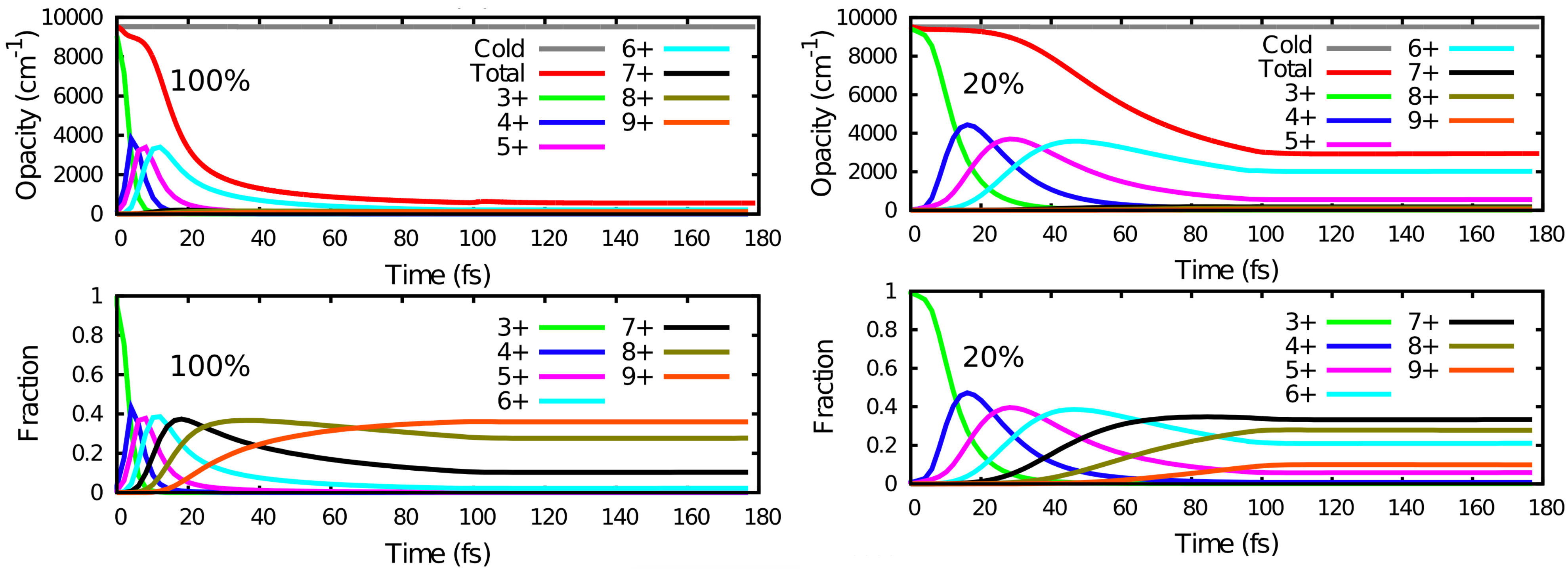
Saturable absorption occurs due to 'burning through' of charge states such that the K-edge of the highly charged ions lies above the FEL photon energy. Thus at the higher photon energies we need to wait until the end of the pulse to lower the absorption, and it is not so effective.

Note: the mechanism is <u>different</u> from that in the XUV, where we were 'beating' a longer (40-fsec) Auger rate.

Phys. Rev. Lett. ***114*** *015003 (2015)*

Evolution of charge states and opacity

FEL energy is 1670 eV, just below k-edge of 7+



Peak x-ray intensity

20% of peak x-ray intensity

Final absorption will be the integral over time and weighted intensities.

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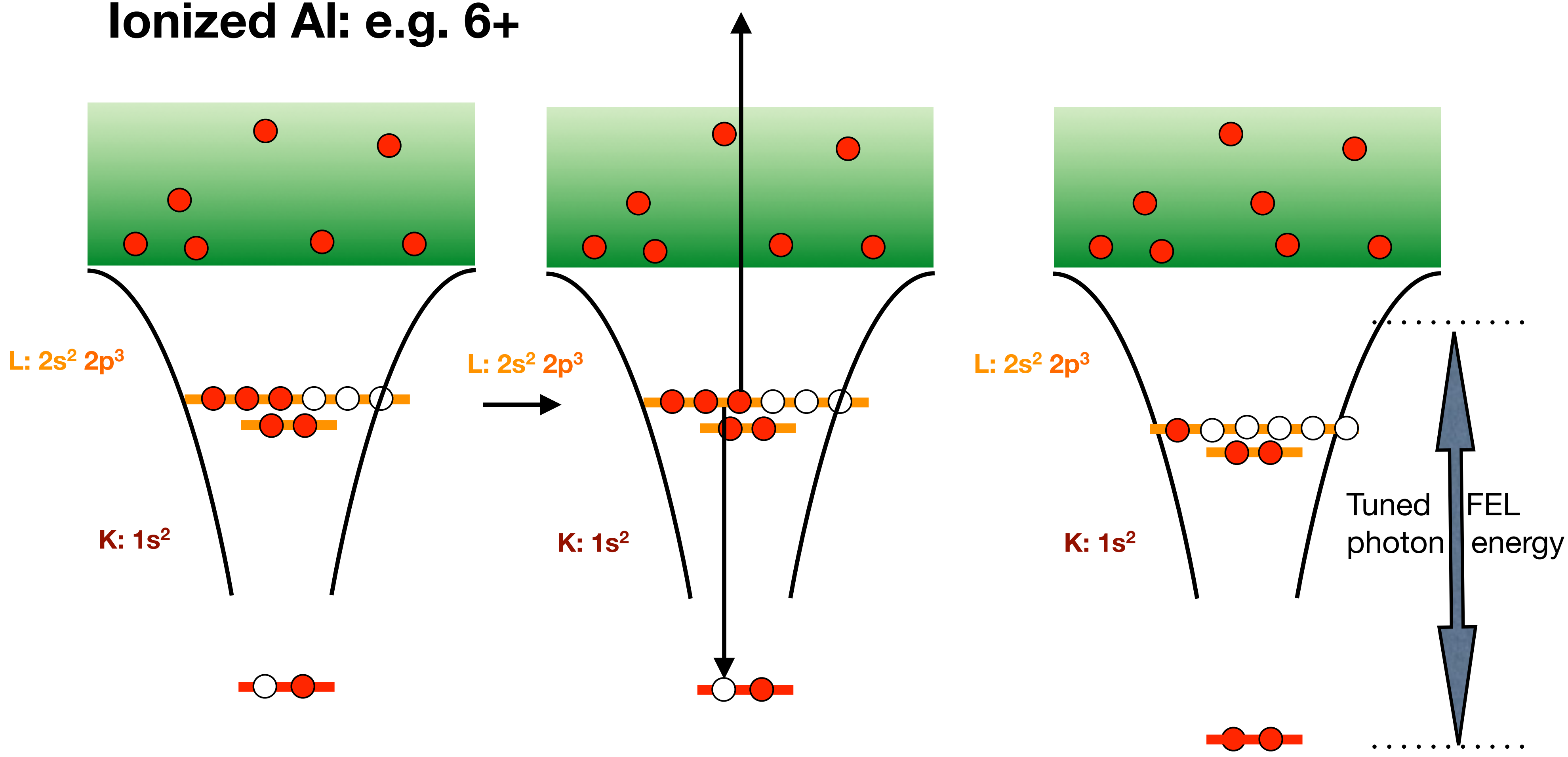
If the FEL photon energy is too low, no K-α generated

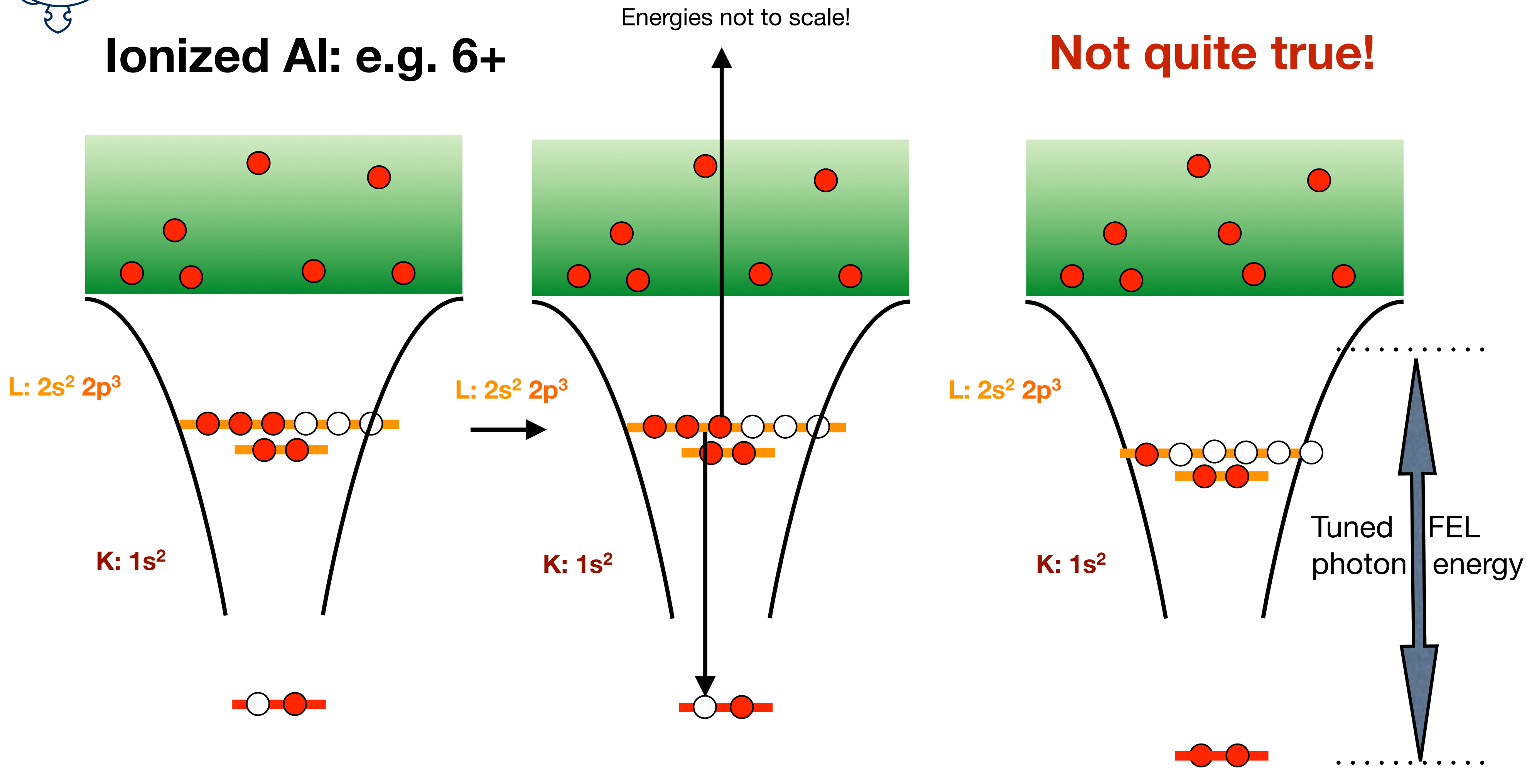
Energies not to scale!

Ionized Al: e.g. 6+



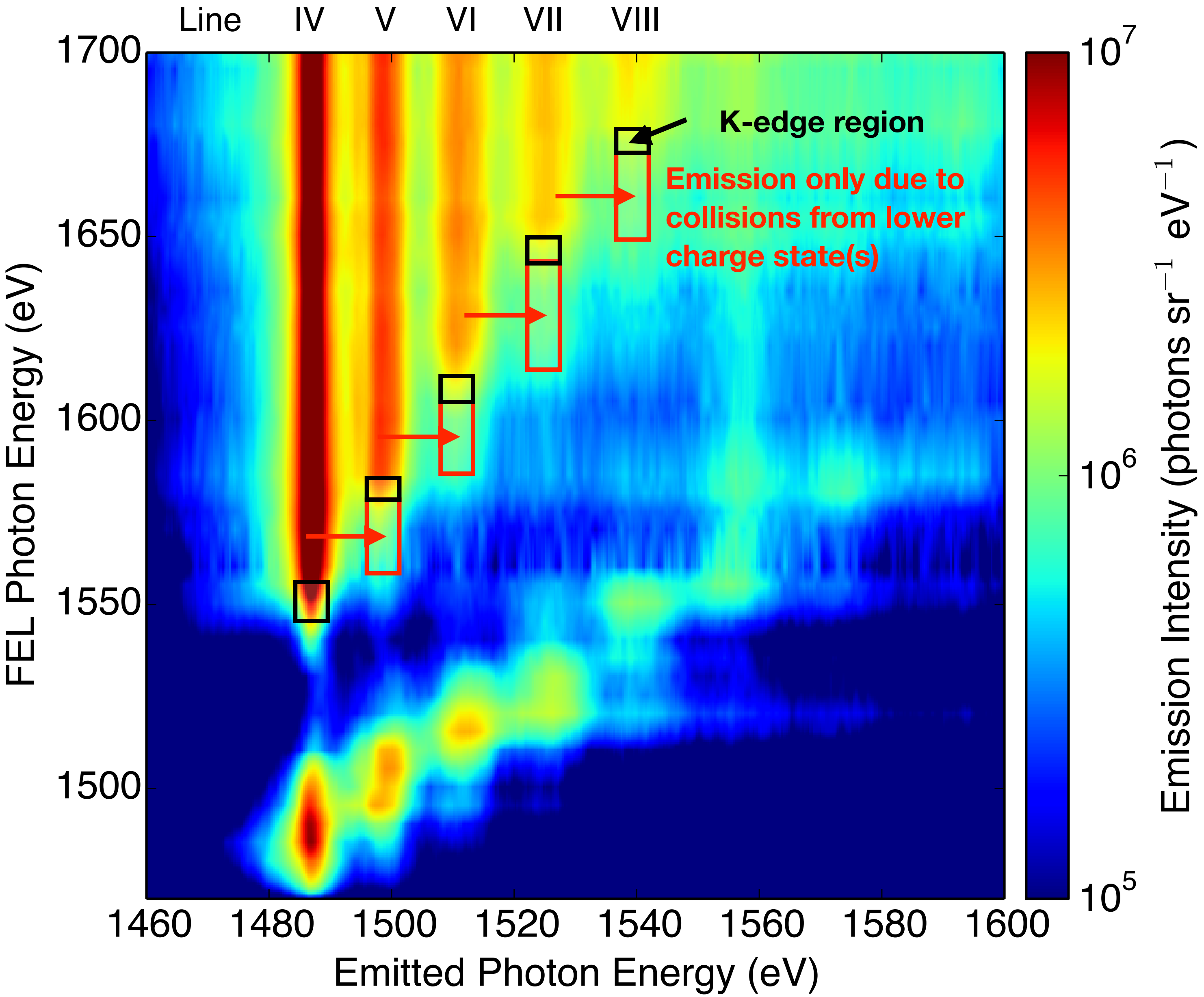
Important to note: the heated continuum - many tens of eV, can further collisionally ionise the system. No K-α will be seen if the K-edge energy of an ion exceeds the photon energy of the X-Ray Laser.

If the FEL photon energy is too low, no K-α generated

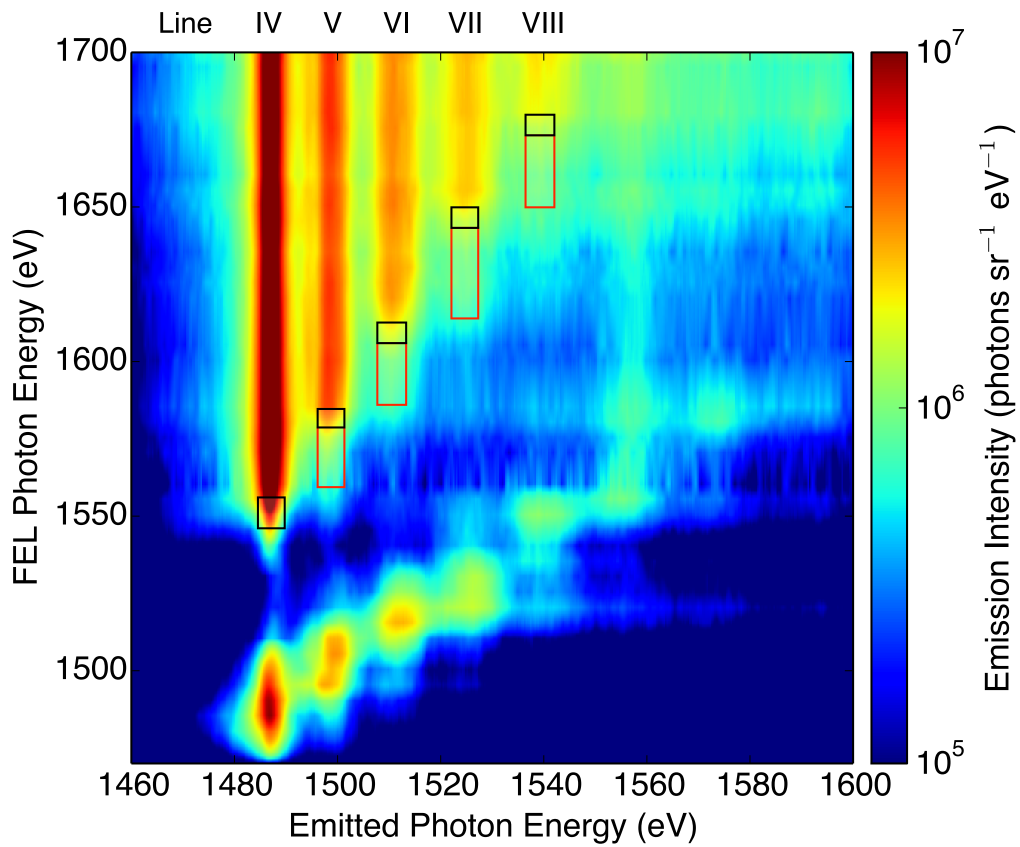


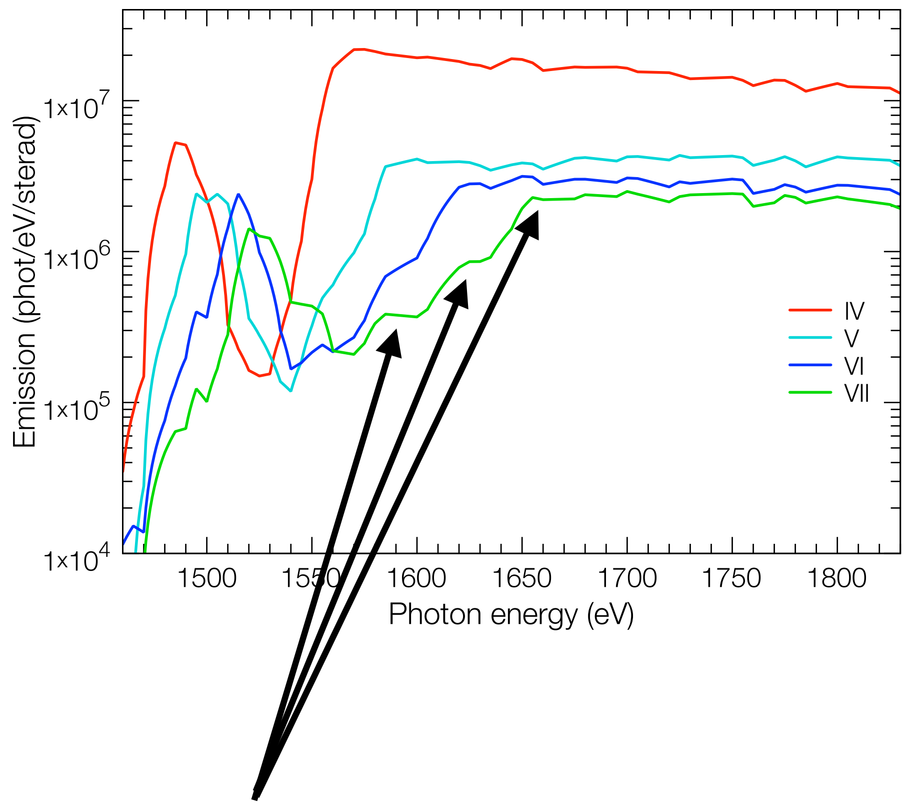
Important to note: the heated continuum - many tens of eV, can further collisionally ionise the system. No K-α will be seen if the K-edge energy of an ion exceeds the photon energy of the X-Ray Laser.

Experimental measurement of collisional ionization rates on LCLS



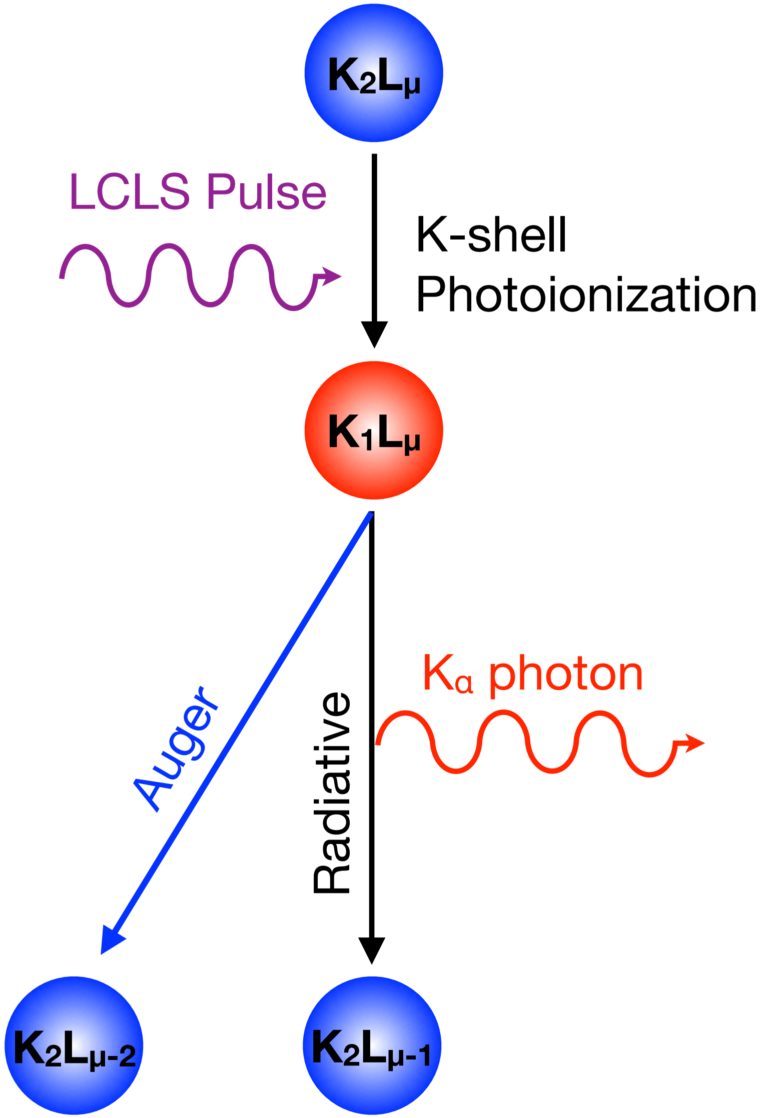
Experimental measurement of collisional ionization rates on LCLS





Clear steps in emission are observed below the K-edge of a given charge state!

Process that produces K_α emission



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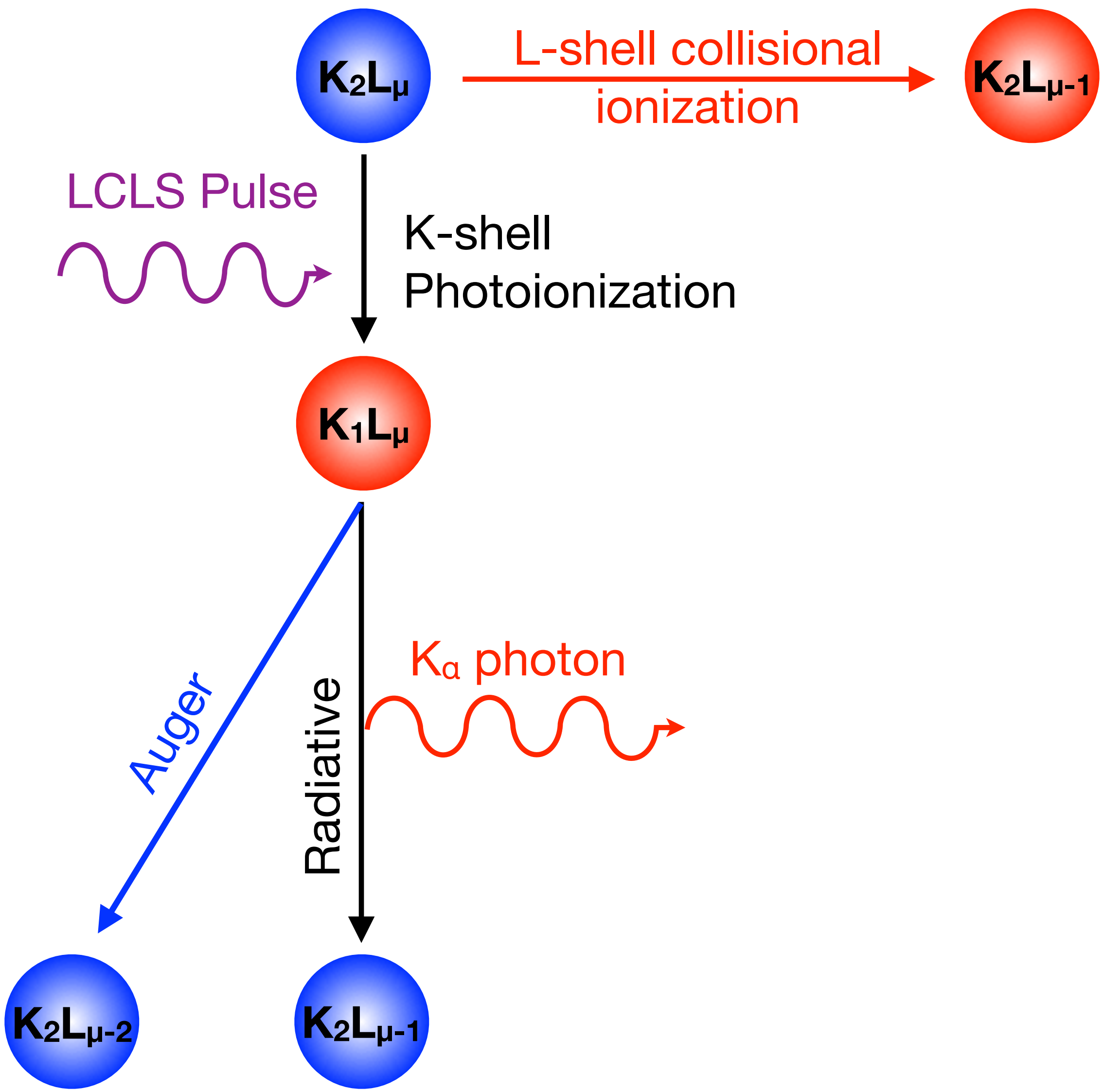
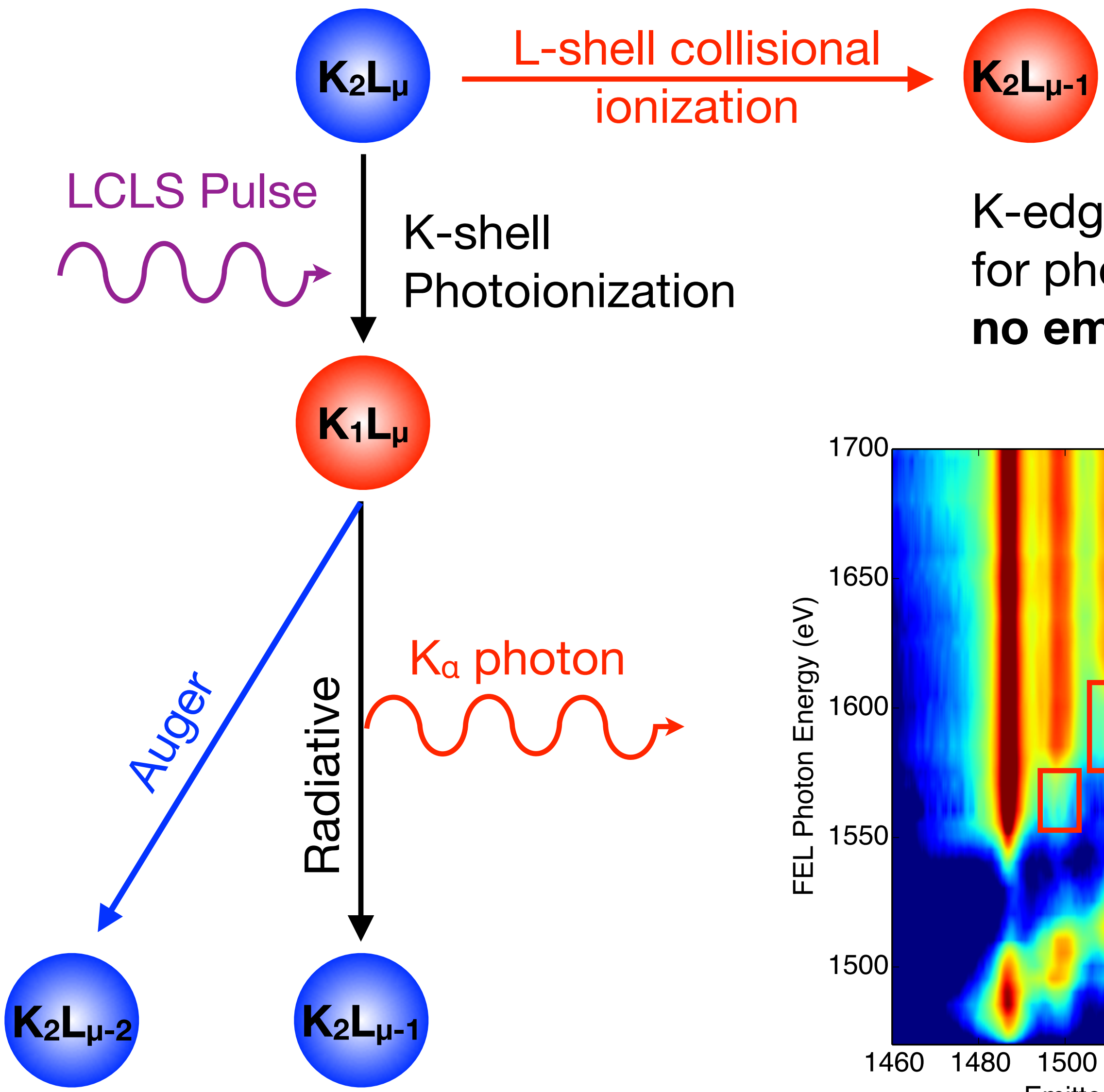
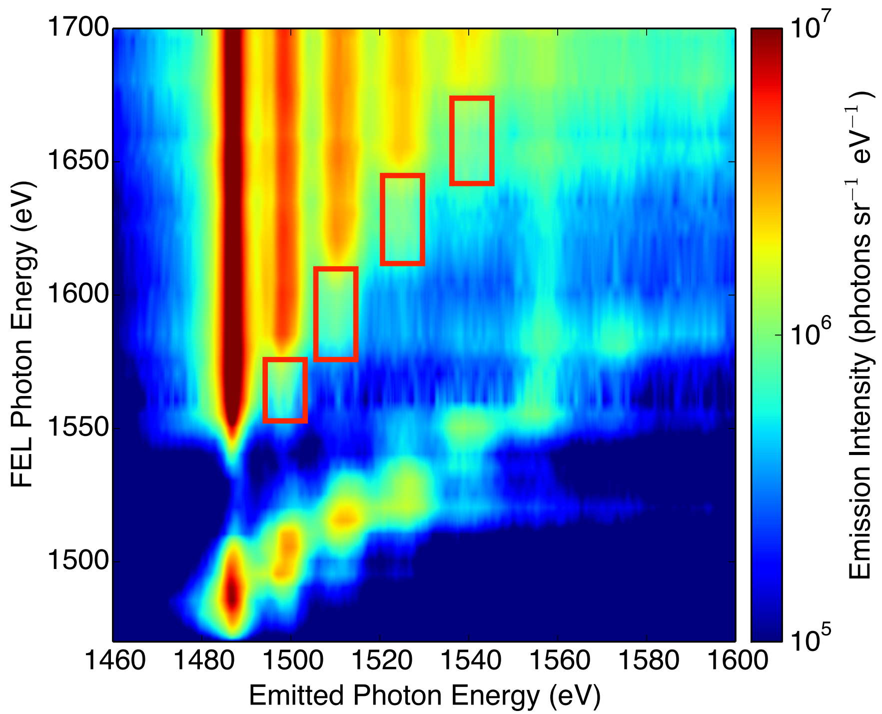


Process that produces K_α emission





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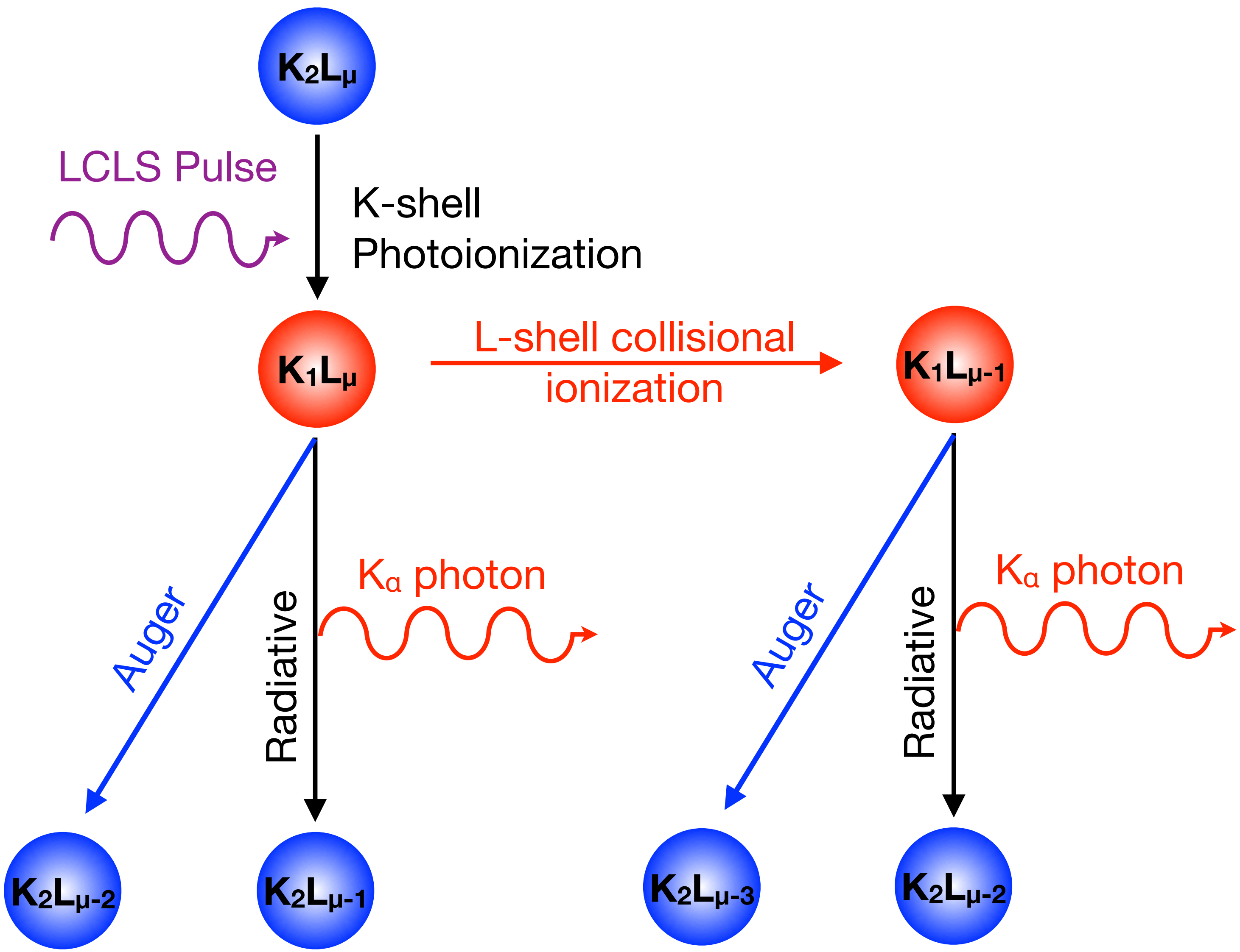

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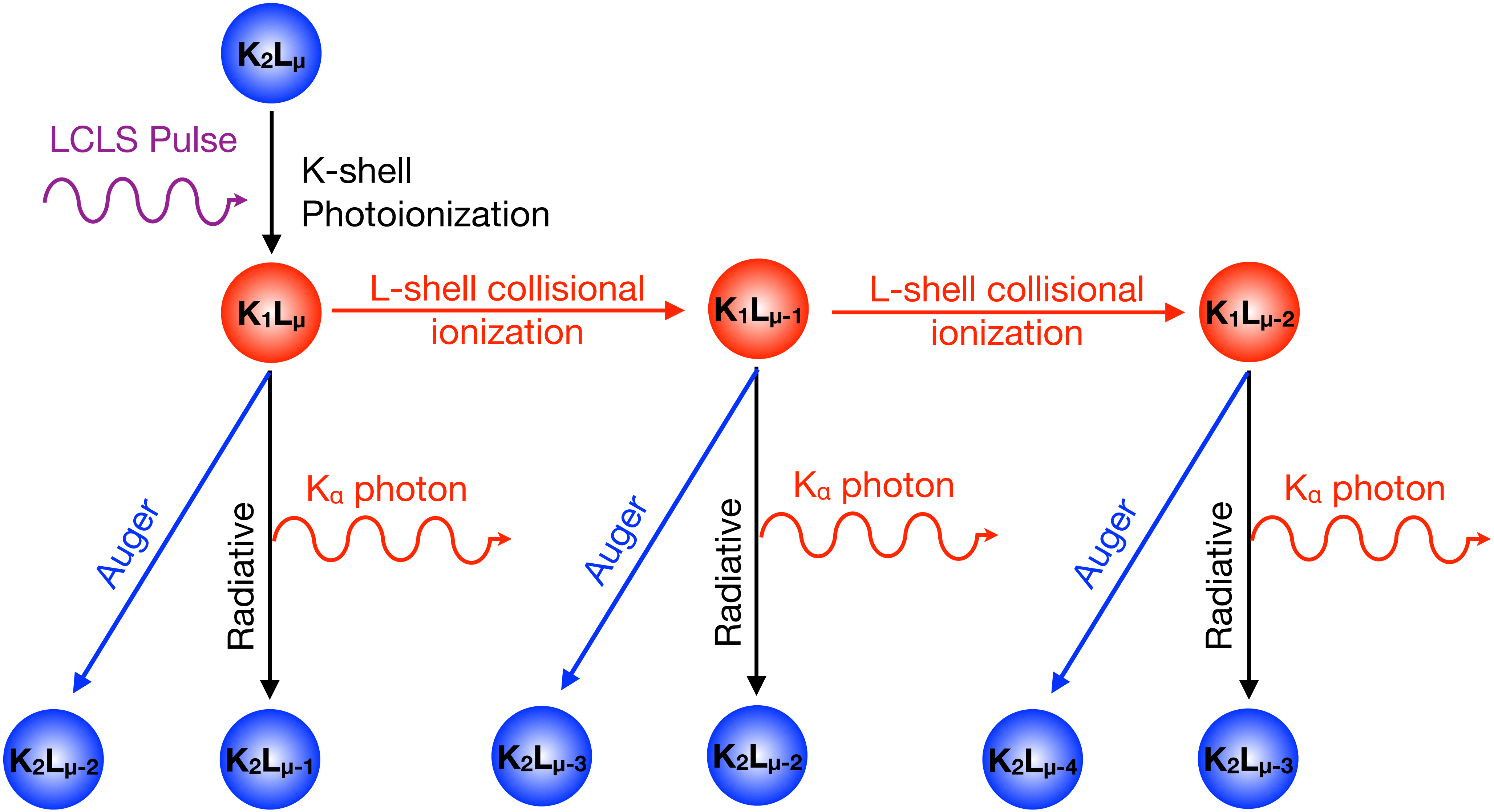
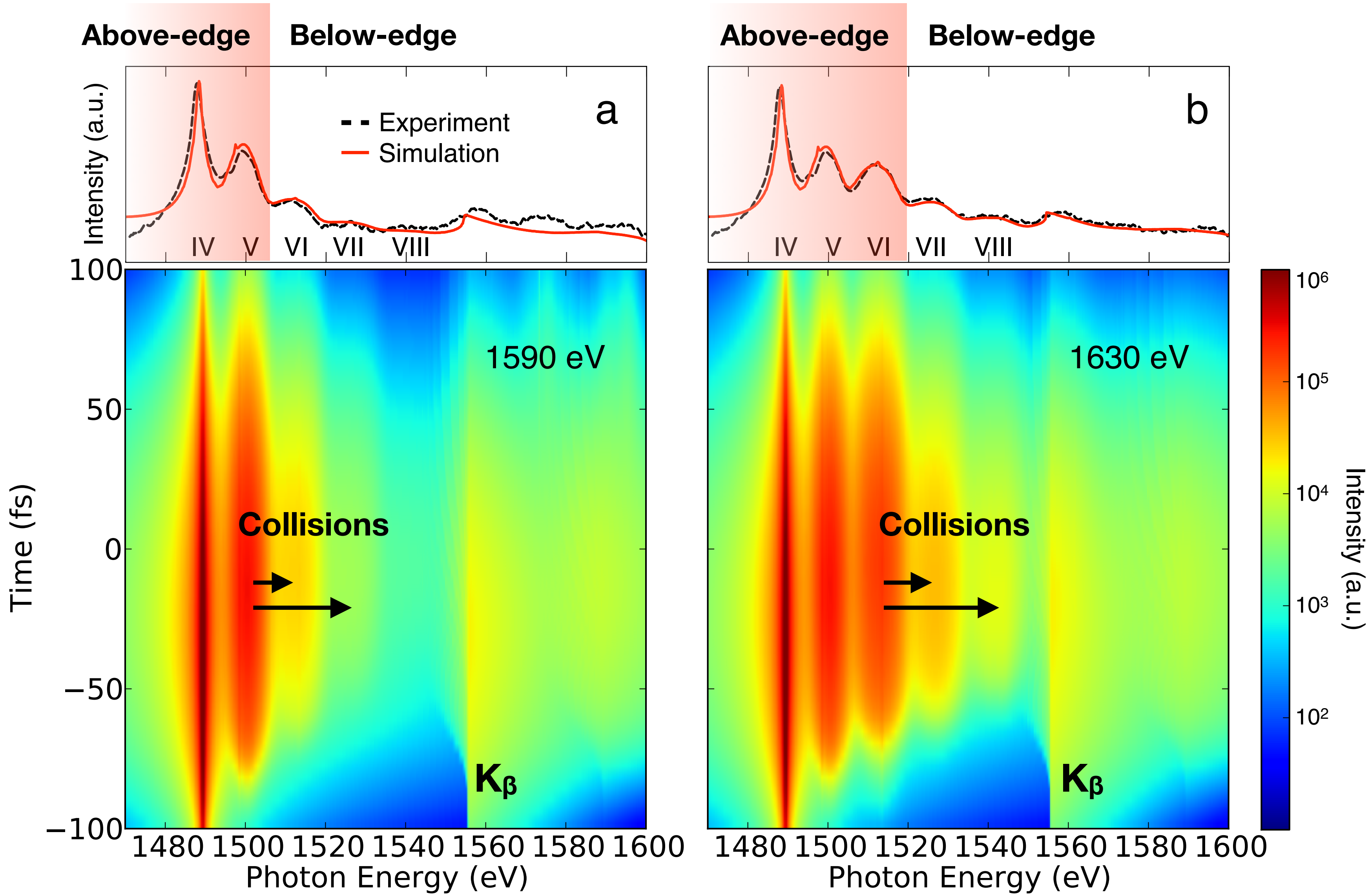
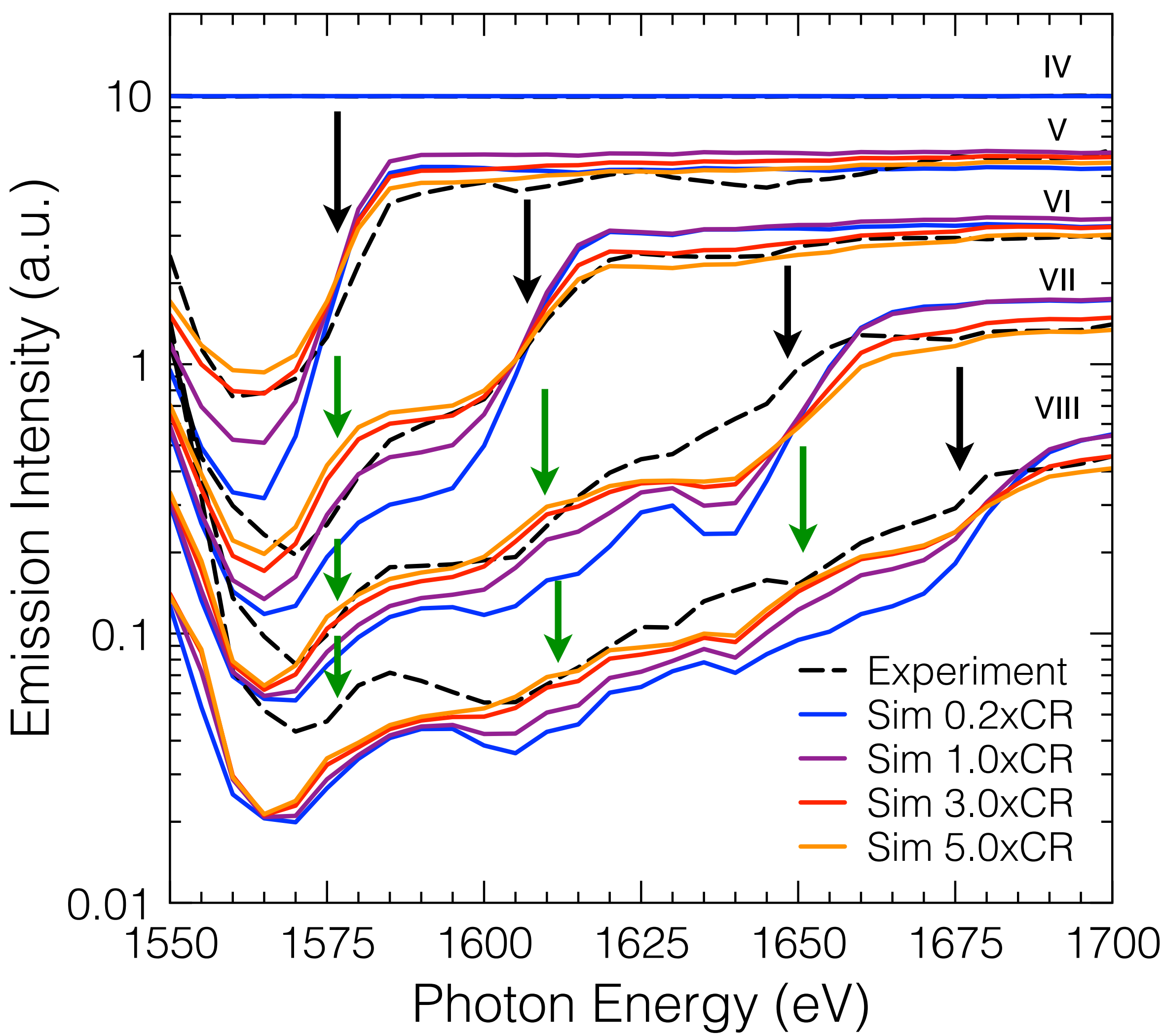


Collisions occur within Auger lifetime of K-shell



We can model the effect of stronger/weaker collisions



Rates are underestimated by a factor between 3 and 5!

First measure of collisional ionisation rate in a dense plasma.

*Nature Comm **6**, 6397 (2015)*

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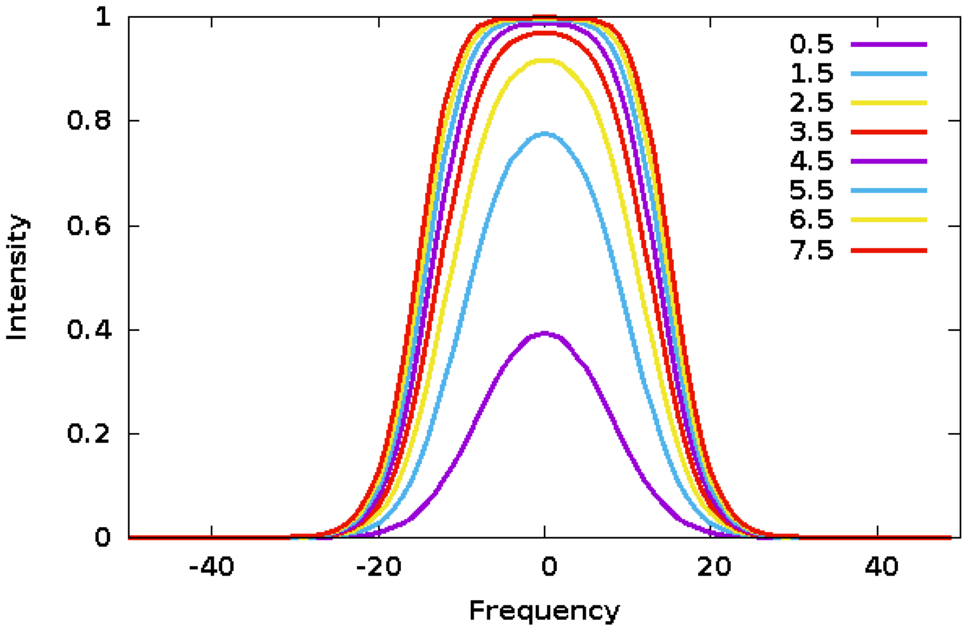
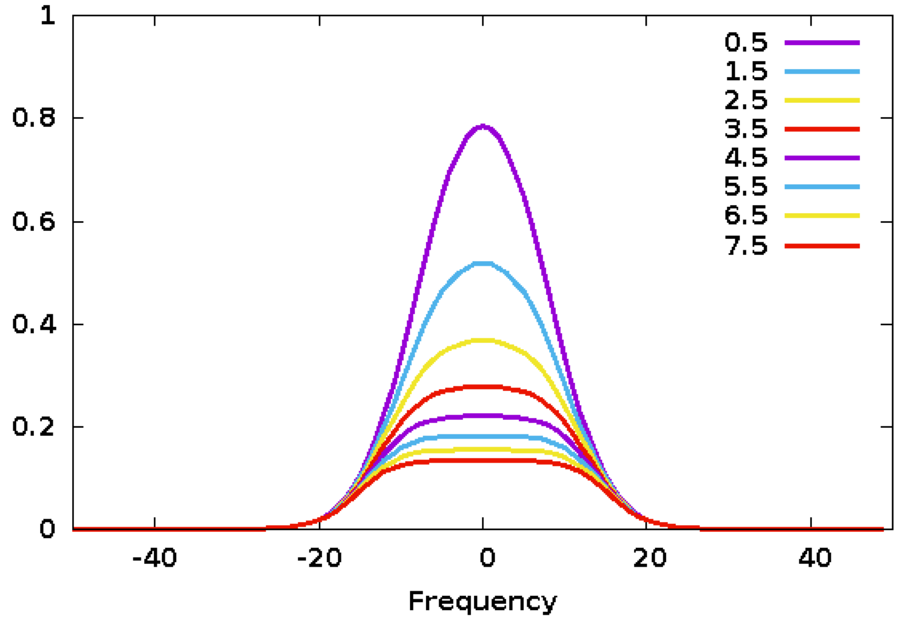
Opacity and Curves of Growth

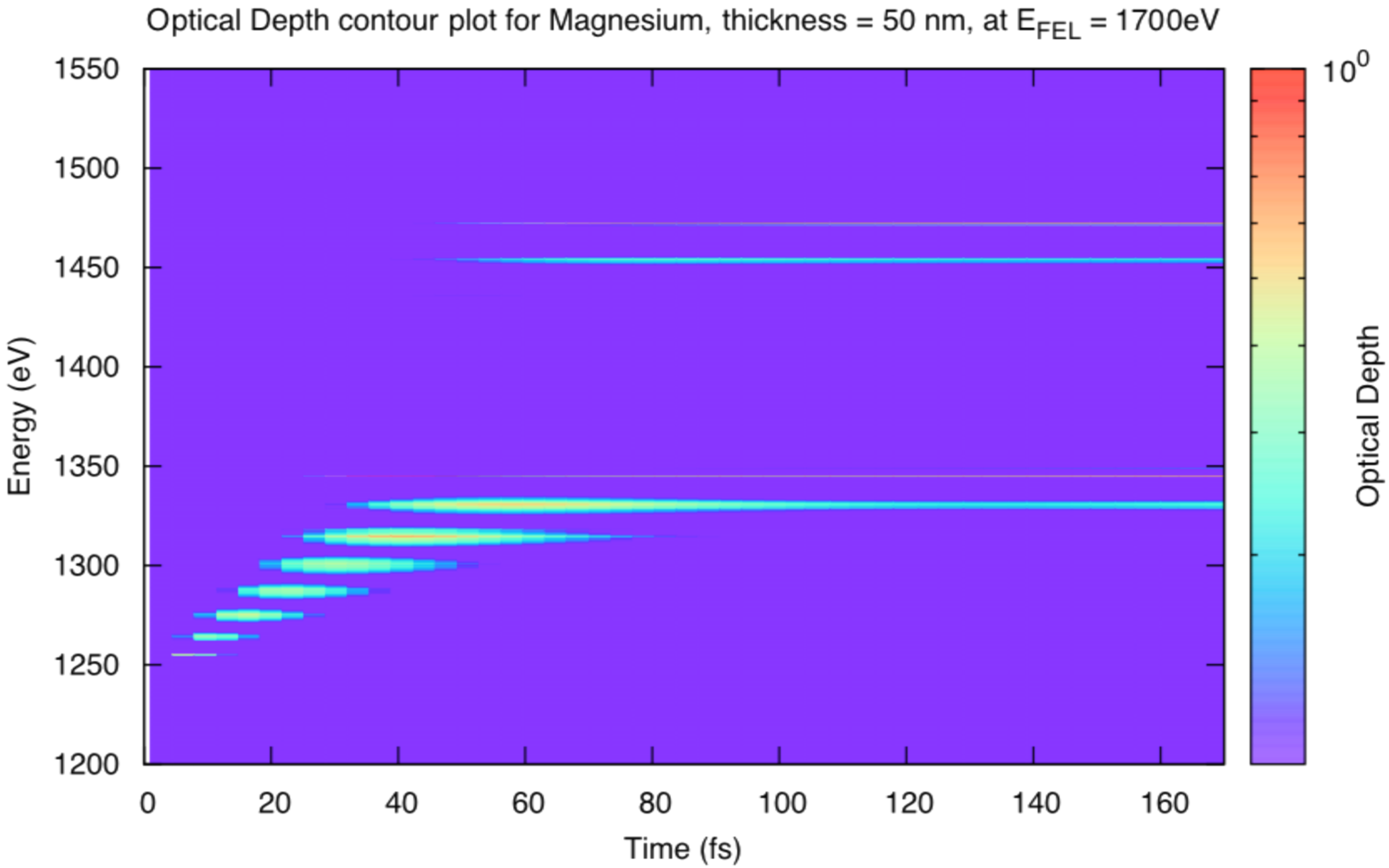




Intensity of Gaussian Line as a function of optical depth at line centre

Intensity of Gaussian Line divided by target thickness as a function of optical depth at line centre

Optical depth plot for 50nm Mg at FEL pump = 1700eV



We predict that we need a 25-50 nm target to have an optical depth less than 1 (i.e 120 - 240 atoms thick).

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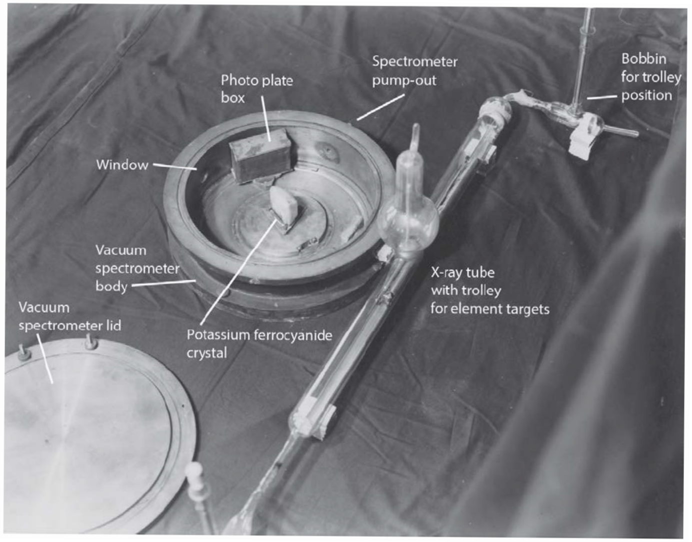
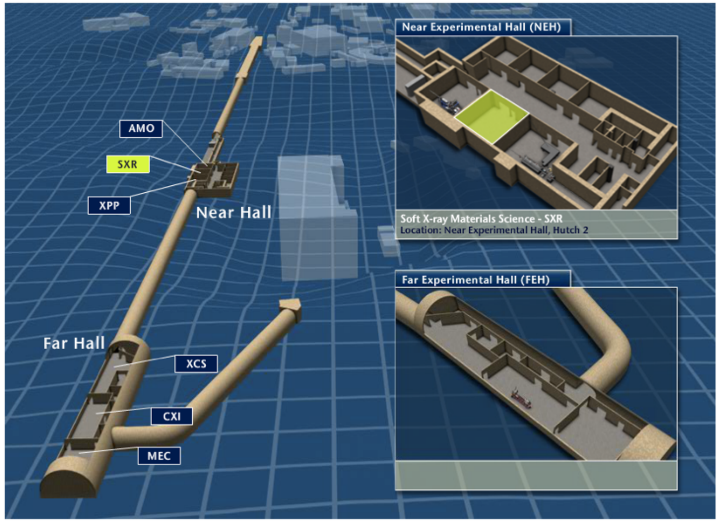
Reclimbing Moseley's Ladder





Moseley used the K-α emission from different elements to sort the elements in the periodic table (1913-1915).

A century later, 2015, we are using the K-α emission from a single element, with different ionisation stages, to determine the behaviour of elements under extreme conditions

In some ways it is as though we are climbing one step, the first step, of Moseley's ladder in multiple tiny steps.

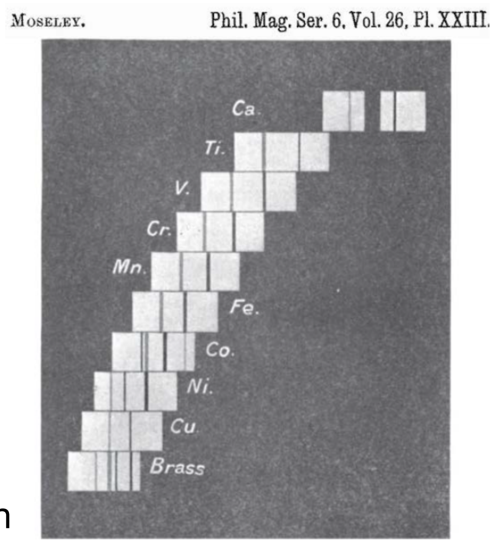
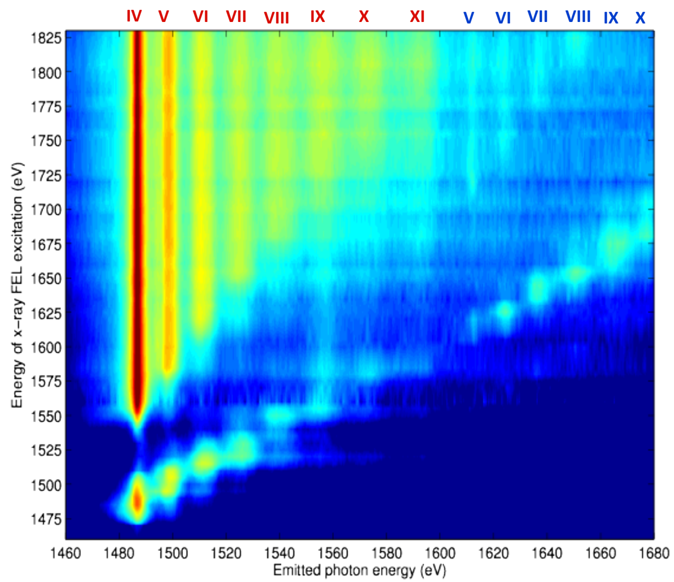


Figure 2 Moseley's plate from *The High-Frequency Spectra of the Elements.*



Reclimbing Moseley's Ladder

Henry ('Harry') Gwyn Jeffries Moseley, from Trinity College, Oxford used measurements of the wavelengths of K-α of the elements to put the periodic table in the correct order. He was shot in the head by a sniper in Gallipoli on 10th August 1915, aged just 27.

"In view of what he [Moseley] might still have accomplished … his death might well have been the most costly single death of the War to mankind generally", Isaac Asimov

Collaborators

U Oxford: Sam Vinko, Orlando Ciricosta, David Rackstraw, Thomas Preston, Patrick Hollebon

SLAC: Bob Nagler (formerly Oxford), Hae Ja Lee, Phillip Heimann

U Berkeley: Richard Lee

LBNL: Byoung-ick Cho, Kyle Engelhorn, Roger Falcone

IAEA: Hyun-Kyung Chung

Czech IOP: Jaromir Chalupsky, Tomas Burian, Vera Hajkova, Libor Juha

DESY: Sven Toleikis, Thomas Tschentscher

… and many others – see all authors of:

Nature Communications **6**, 6397 (2015)
Phys. Rev. Lett. **114**, 015003 (2015)
Nature Communications **5**, 3313 (2014)
Phys. Rev. Lett. **109**, 245003 (2012)
Phys. Rev. Lett. **109**, 065002 (2012)
Nature **482**, 59 (2012)
Phys. Rev. Lett. **106**, 164801 (2011)
Phys. Rev. Lett. **104**, 225001 (2010)
Nature Physics **5**, 693 (2009)

Conclusions

- X-Ray lasers have revolutionised our ability to create and diagnose solid-density plasmas with well controlled temperature and electron density distributions. They can uniformly heat matter before expansion.
- Basic optical phenomena, such as saturable absorption, previously confined to the optical regime, have been demonstrated in the XUV and X-Ray regions - though based on differing physical mechanisms in each case.
- The solid-density plasmas generated by LCLS only emit K-shell radiation during the <100fsec pulse, allowing accurate diagnosis well before hydrodynamic motion.
- The basic mechanisms and rates for ultra-intense X-Ray absorption are understood.
- Accurate measurements of the K-edges of ions in Al, Mg and Si plasmas show that the ionisation potential depression is far greater than standard semi-classical theory.
- Full ab initio finite-temperature multi-centre (non-spherically-symmetric) DFT calculations are in excellent agreement with measurements.
- We can gain a measure of femtosecond collisional ionisation rates by observing ‘leakage’ of K-a radiation from one charge state to the next.
- Optically-thin solid-density plasmas, just 120 atoms across, show potential for us to interrogate line-shapes and kinetics.