Density effects on plasma spectroscopy

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2023 Atomic Processes in Plasmas

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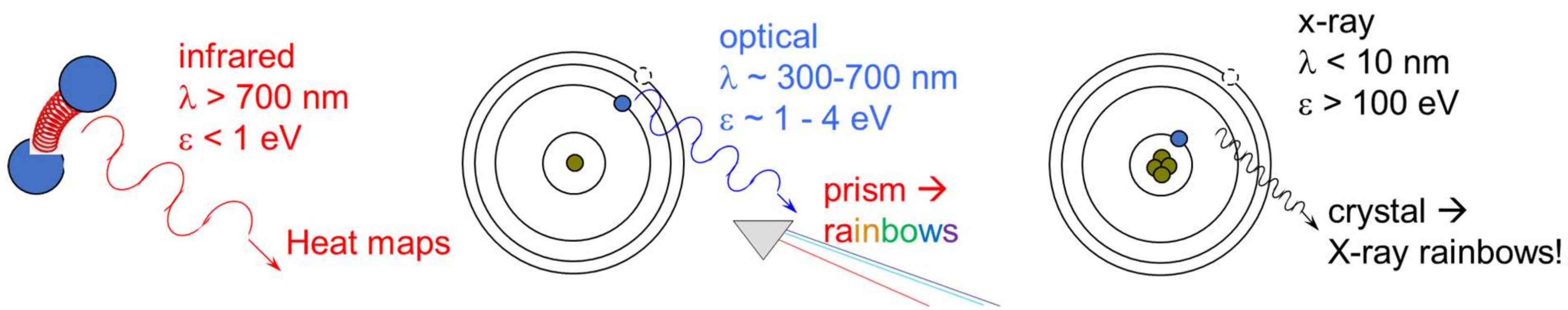
Spectroscopy tells us about the basic structure of matter

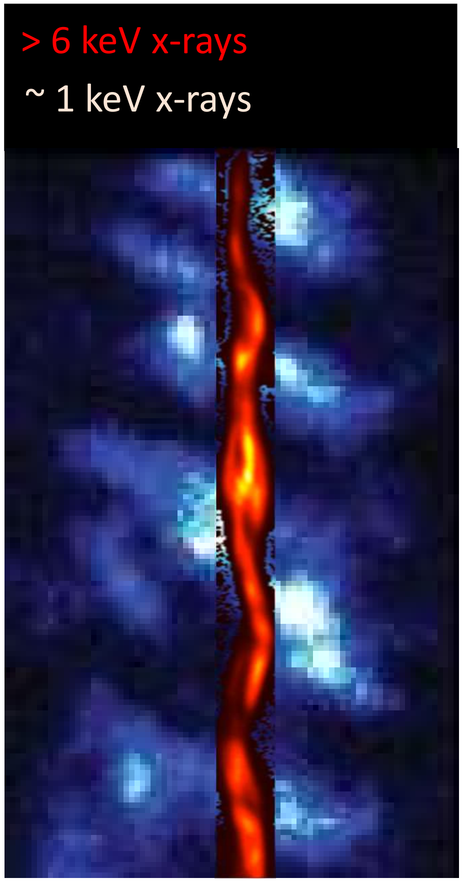


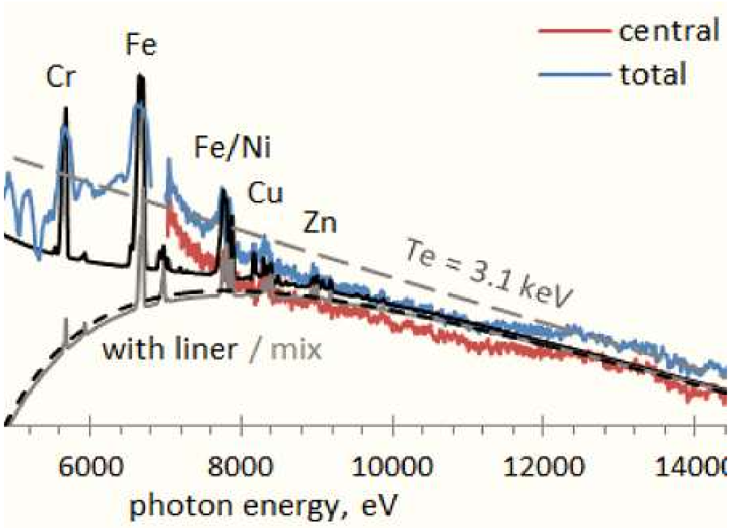
Probing material at different wavelengths/ energies allows us to probe different physical phenomena, from thermal motion to electronic structure

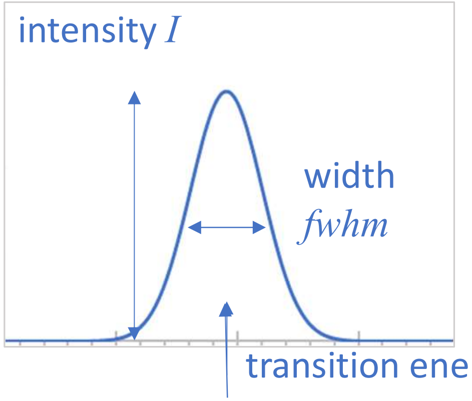
For highly ionized plasmas, low-energy emission is often dominated by collective effects and/or optically thick, so X-rays are particularly useful

For many HED plasmas, a spectrum is worth a thousand pictures

- Coarse energy resolution in imaging can reveal plasma gradients
- Fine energy resolution (spectroscopy) can reveal details of plasma composition, temperature, and <u>density</u>
- Each emission or absorption line is characterized by its wavelength (energy), intensity, and width (or shape)
- Interpreting spectra requires understanding atomic-scale structure and response







There are two categories of density effects we can see with spectroscopy

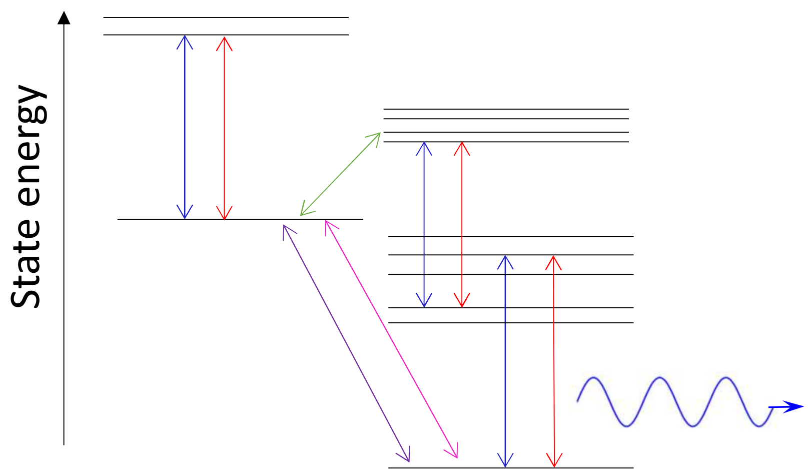
1. Atomic kinetic effects – Collisional radiative models
    a. Ladder ionization
    b. Metastable states
    c. Degenerate electron distributions

2. Atomic structure effects – Density functional theory models
    a. Plasma screening
    b. Pressure ionization
    c. Line broadening

Collisional-radiative models are foundational to x-ray spectroscopy

Collisional–radiative models (Ralchenko) are built from <u>states</u> (Gu) and <u>rates</u> (Fontes, Ballance, Gu)

Collisional-radiative models balance rates into and out of each state: $dX_i/dt = \Sigma_j X_j R_{ji} - X_i \Sigma_j R_{ij}$



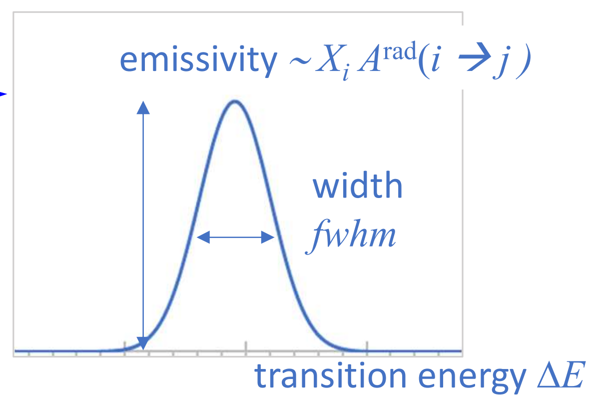
Collisional excitation & de-excitation

Photo-excitation, stimulated emission, & radiative decay

Collisional ionization & **three-body recombination**

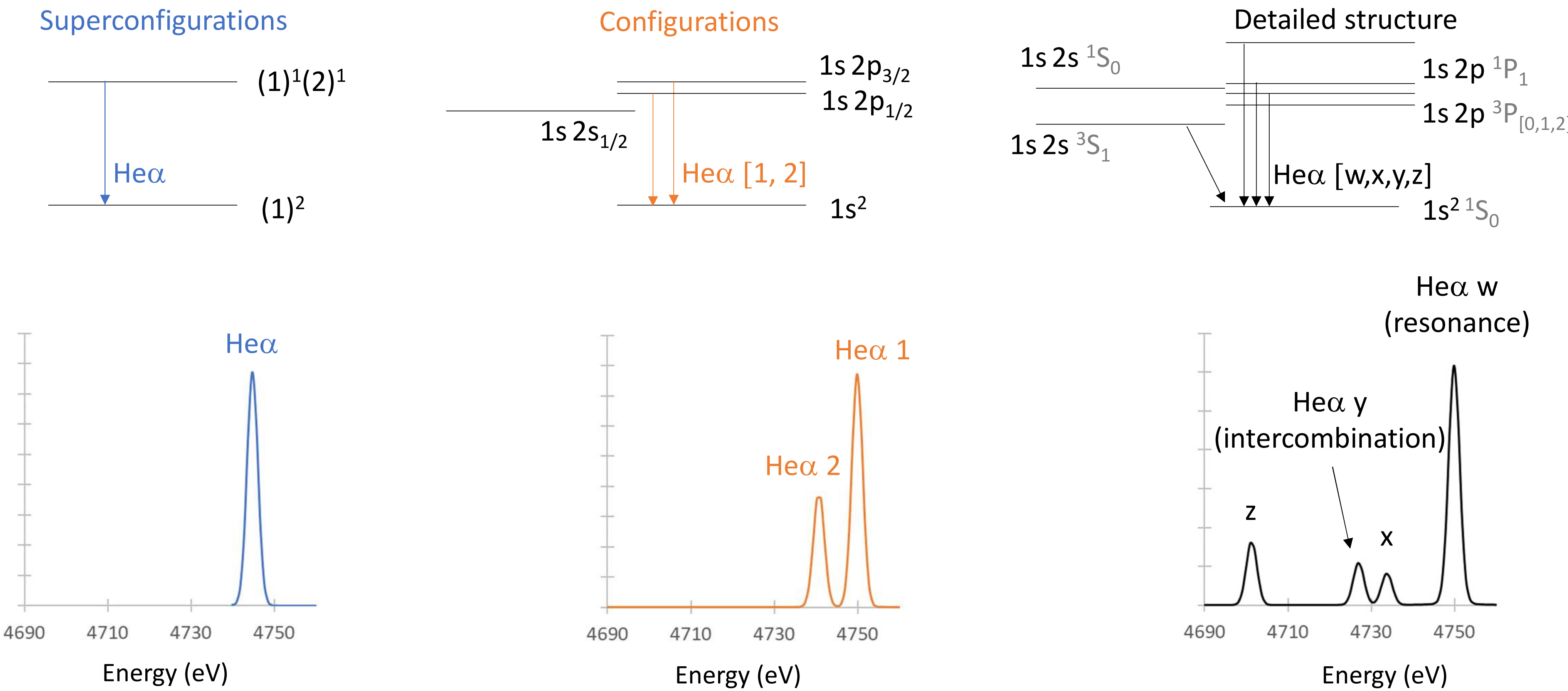
Photoionization & **radiative recombination**

Autoionization & **dielectronic recombination**



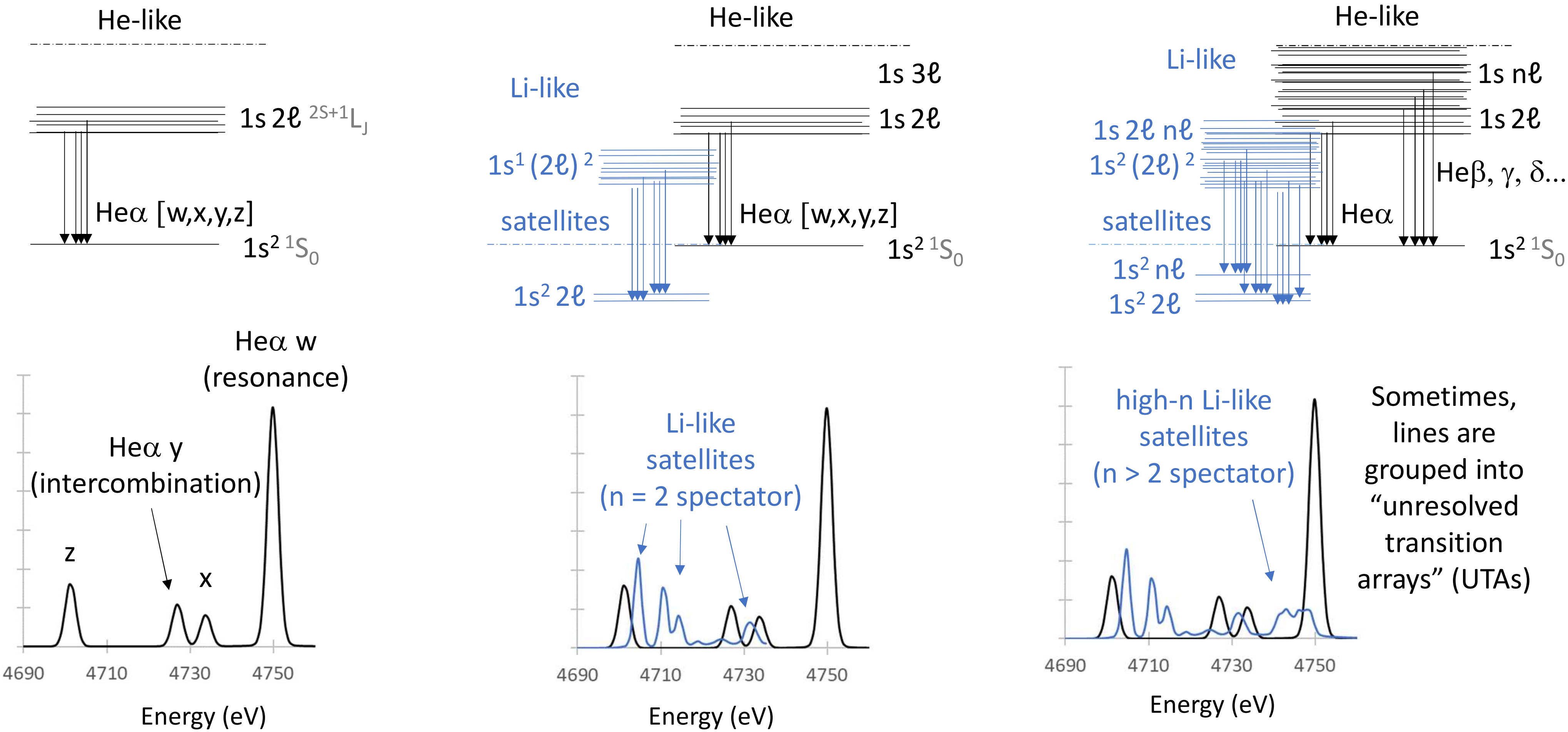
If you know state populations X_i, radiative decay rates A^{rad}, and line profiles, you can compute a spectrum!

Electronic states can be treated with varying levels of detail



Generally, we need fairly detailed state structure to reliably diagnose experiments

Different degrees of completeness also modify modeled spectra



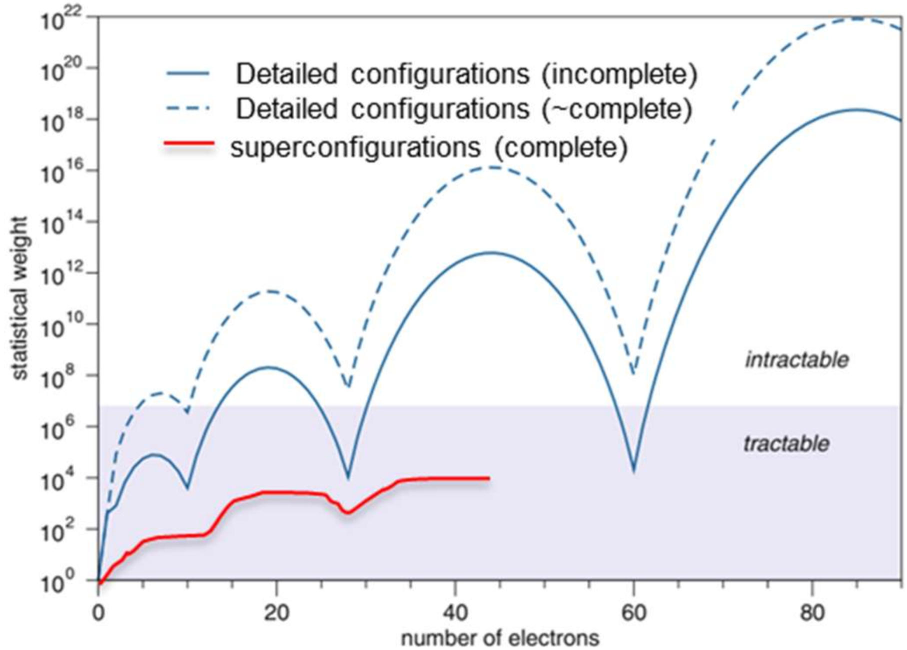
Sometimes, lines are grouped into "unresolved transition arrays" (UTAs)

Unfortunately, complete & detailed CR models can become intractable

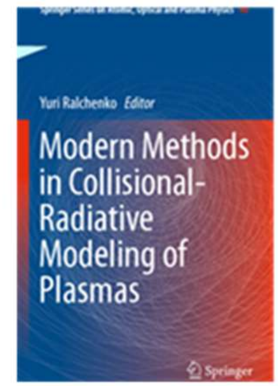
The combinatorics of detailed electronic structure can be daunting: the most detailed models (and real ions) have $N \sim \Sigma_i g_i$

CR models must calculate all rates among all states and invert the rate matrix to find the occupations X_i needed to compute line intensities

Thus, many CR models use less detailed states (configurations or superconfigurations) – or hybrid structure representations

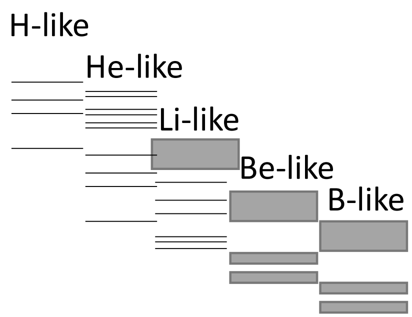


2016
Ed. Yu. Ralchenko

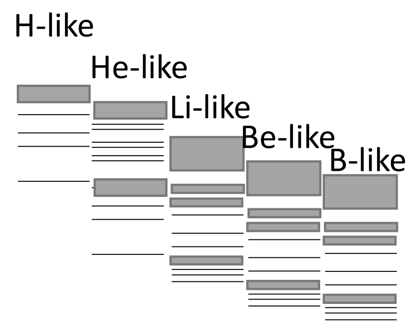


Different CR models take different approaches to state structure

<u>No</u> CR model can be fully detailed, statistically complete, AND computationally tractable.
Different modelers make different choices to balance detail, completeness, and speed

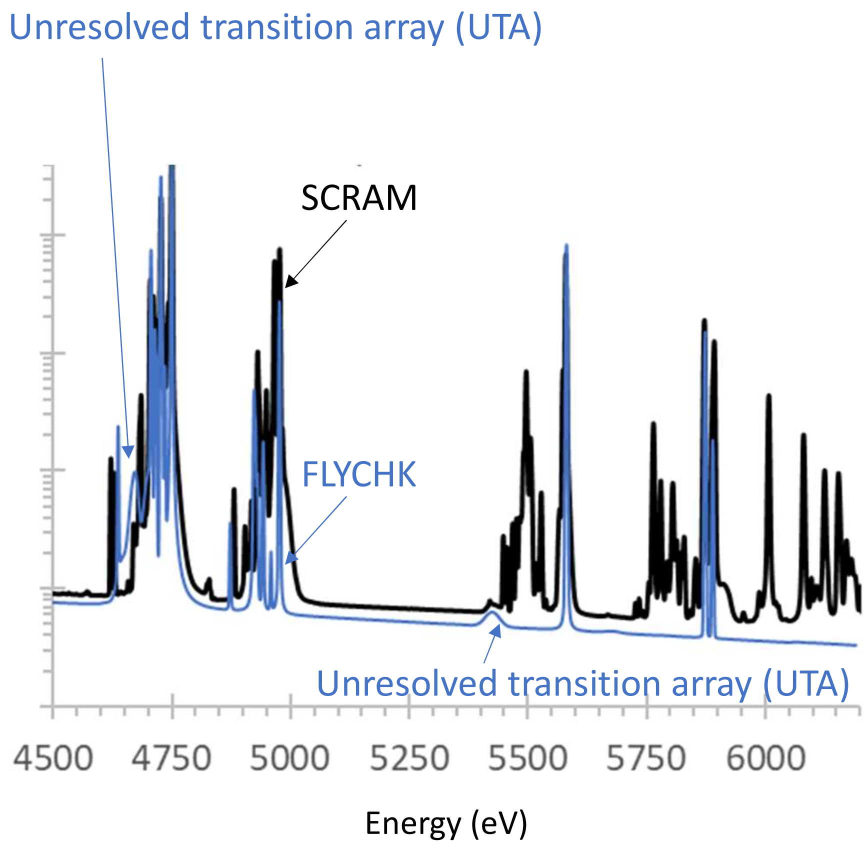


FLYCHK uses mostly superconfigurations, limiting detail to some K-shell ions
→ runs in seconds



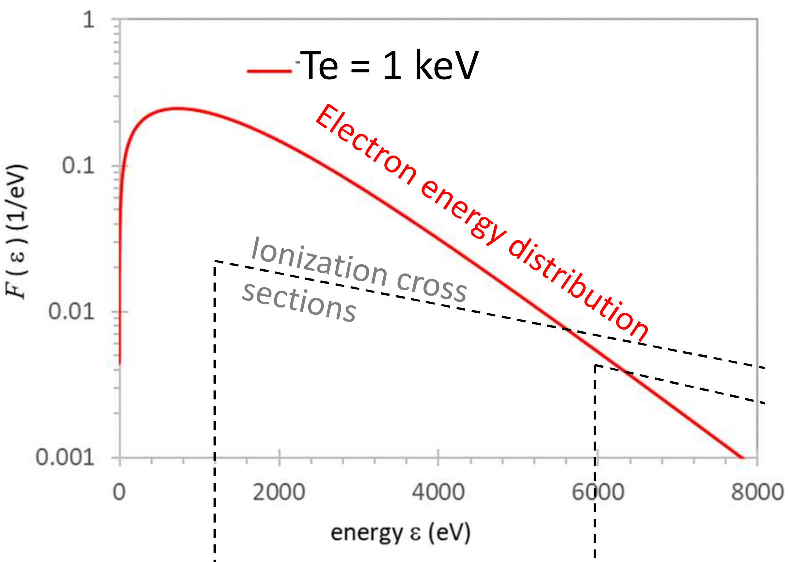
SCRAM mixes detailed states with configurations & superconfigurations, retaining some detail for every ion
→ runs in minutes

Differences in modeled structure are reflected in modeled spectra



No model is perfect:
understanding model limitations &
uncertainties is a critical part of
spectroscopic diagnostics

Rates are an additional source of model differences



ΔE ~ 1.4 keV to ionize 2s in Li-like Ti

ΔE ~ 6 keV to ionize $1s^2$ in He-like Ti

Collisional rates are calculated by integrating cross sections over the electron energy distribution $F(\varepsilon)$

Cross sections can be calculated in a wide variety of ways (see Fontes, Ballance, Gu)

In classical (non-degenerate) plasmas, the electron energy distribution is Maxwellian:
$F_M(\varepsilon) = 2/T_e^{3/2}\ (\varepsilon/\pi)^{1/2}\ e^{-\varepsilon/Te}$

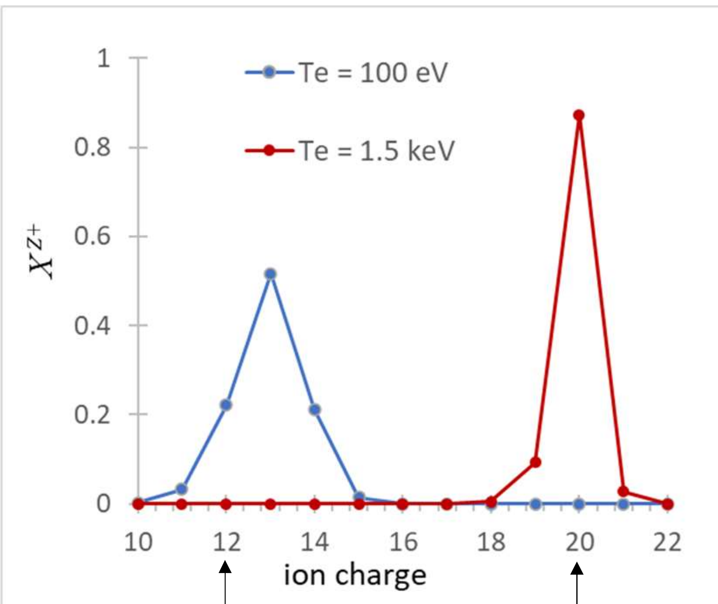
While downward rates sample all of the electrons, upward rates can only happen when impact electron energies are above some threshold (e.g. the ionization potential)

State and ion populations are thus strongly dependent on temperature

We can sum up the state occupation probabilities X_i in different ways:

Charge state distributions (CSDs)

$X^{Z+} = \Sigma_i X_i$ for all i in ion with charge $Z+$



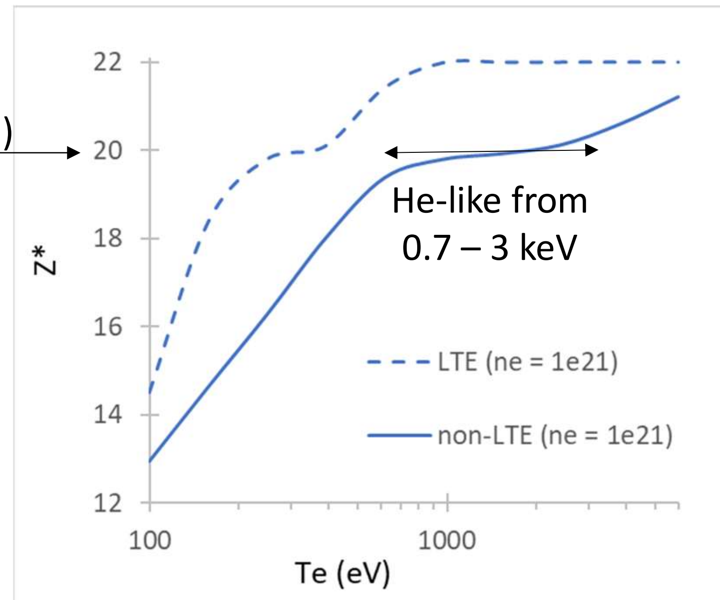
Ne-like Ti
(10 electrons)

He-like Ti
(2 electrons)

Average ionization Z^* (Zbar, < Z >)

$Z^* = \Sigma\ (Z+)\ X^{Z+}$

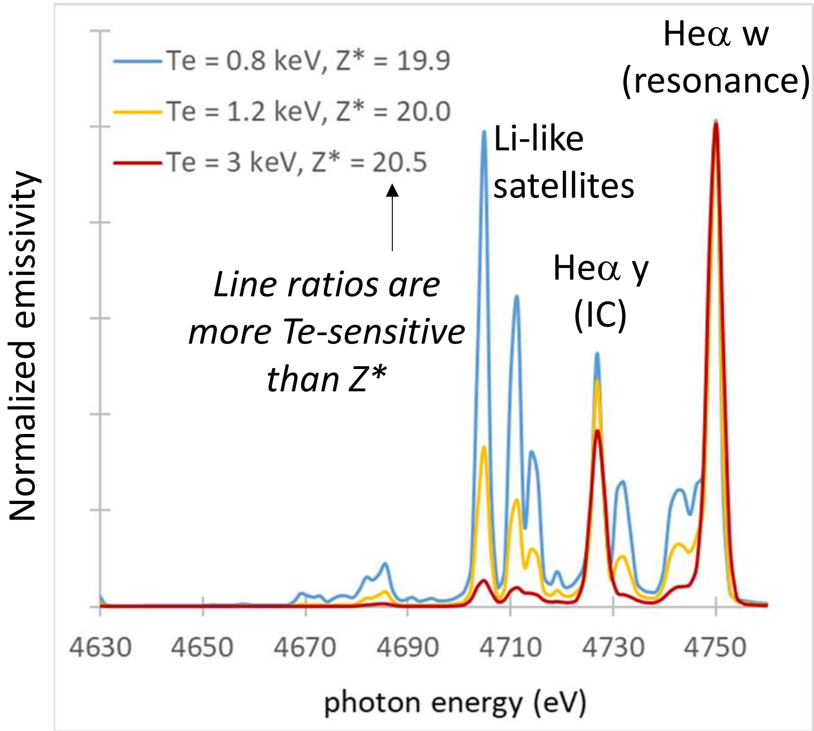
He-like Ti
(2 electrons)
closed shell

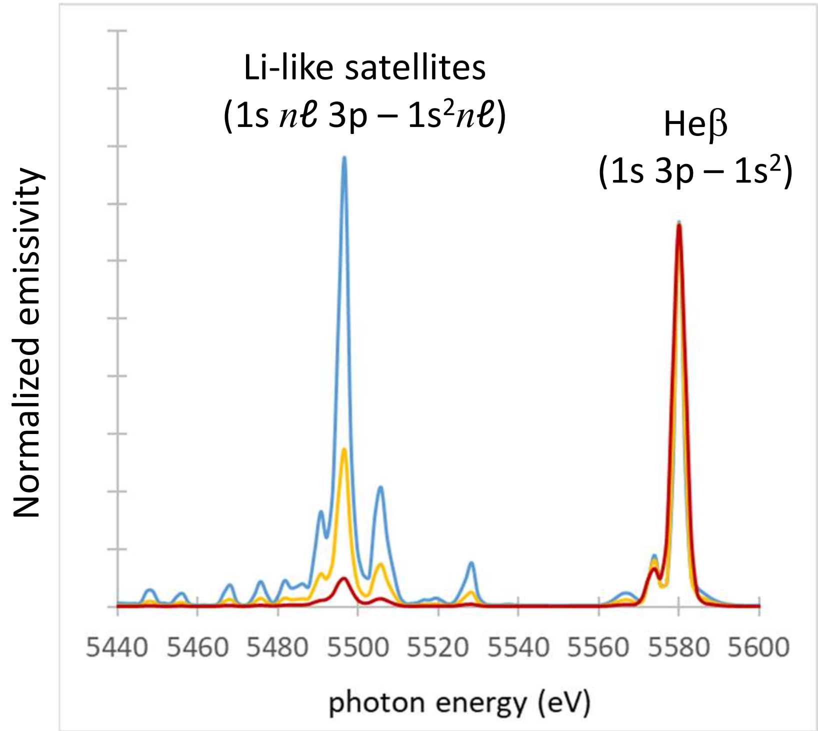


The average ion charge is most sensitive to temperature away from closed shells

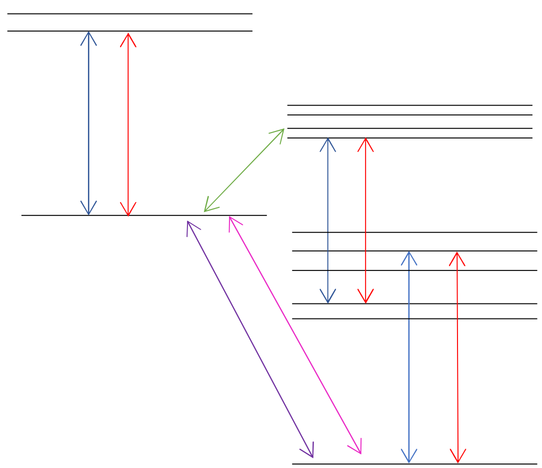
This temperature sensitivity is also reflected in emission spectra

For a given electron density, ratios of Li-like satellites to Heα and Heβ lines are good "thermometers"





Rates also depend on the electron density



Collisional excitation & de-excitation ~ n_e^1

Photo-excitation, stimulated emission, & radiative decay ~ n_e^0

Collisional ionization ~ n_e^1 & **three-body recombination ~ n_e^2**

Photoionization ~ n_e^0 & **radiative recombination ~ n_e^1**

Autoionization ~ n_e^0 & **dielectronic recombination ~ n_e^1**

$R^{ion}/R^{rec} \sim (1/n_e)\,\exp(-\Delta E/T_e)$ *for all ionization rate pairs*.

For $T_r = 0$,

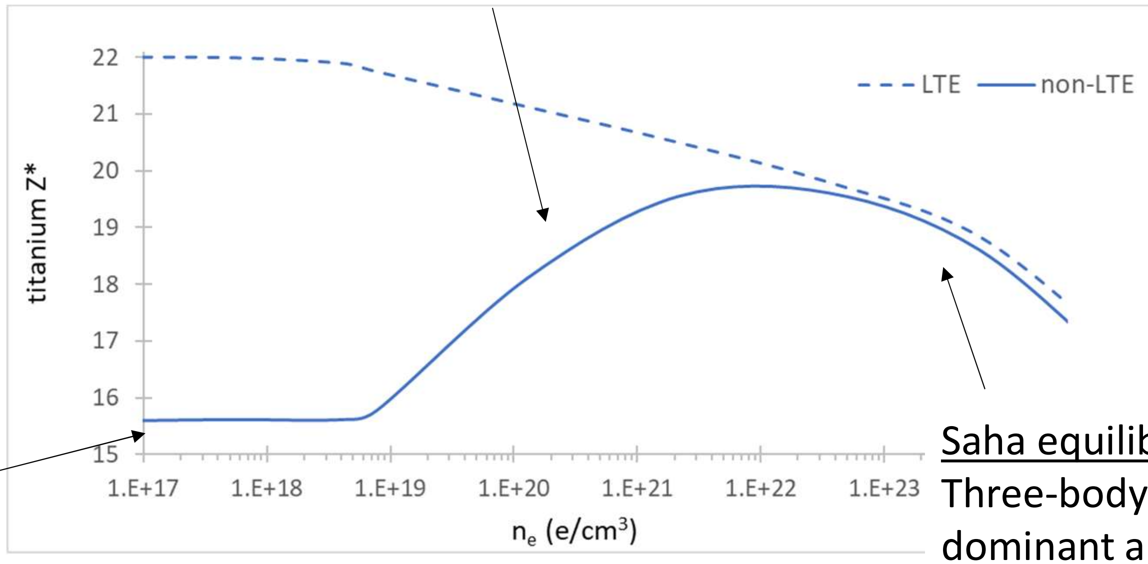
<u>At low densities</u>, collisional ionization (~n_e^1) balances with radiative & dielectronic recombination (~n_e^1) : $X^{Z+1}/X^Z \sim \exp(-\Delta E/T_e)$ → coronal

<u>At high densities</u>, collisional ionization (~n_e^1) balances with three-body recombination (~n_e^2): $X^{Z+1}/X^Z \sim (1/n_e)\,\exp(-\Delta E/T_e)$ → Saha/LTE

Balancing rates leads to complex density dependence in Z^*

<u>Ladder ionization</u>

Excitation-ionization processes boost ionization; lead to increasing Z^* with n_e



<u>Coronal equilibrium</u>

Balance of collisional ionization with radiative & dielectronic recombination is independent of n_e

<u>Saha equilibrium</u>

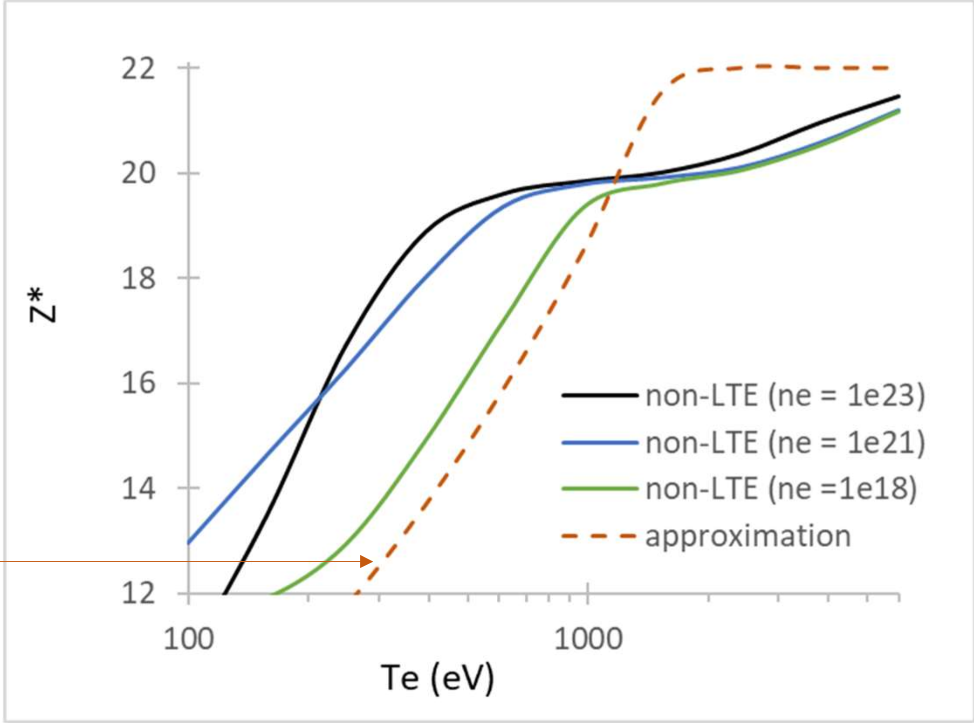
Three-body recombination becomes dominant and enforces detailed balance with collisional ionization; Z^* decreases with n_e

Z^* thus depends on both temperature and density

If you have at least a rough idea of density, obtaining Z^* from spectroscopy is a good thermometer. Otherwise, use caution.

In the low density/ coronal regime (e.g. tokamak, EBIT, solar corona), Z^* can give a first estimate of temperature by inverting the simple (*very rough!!*) approximation:

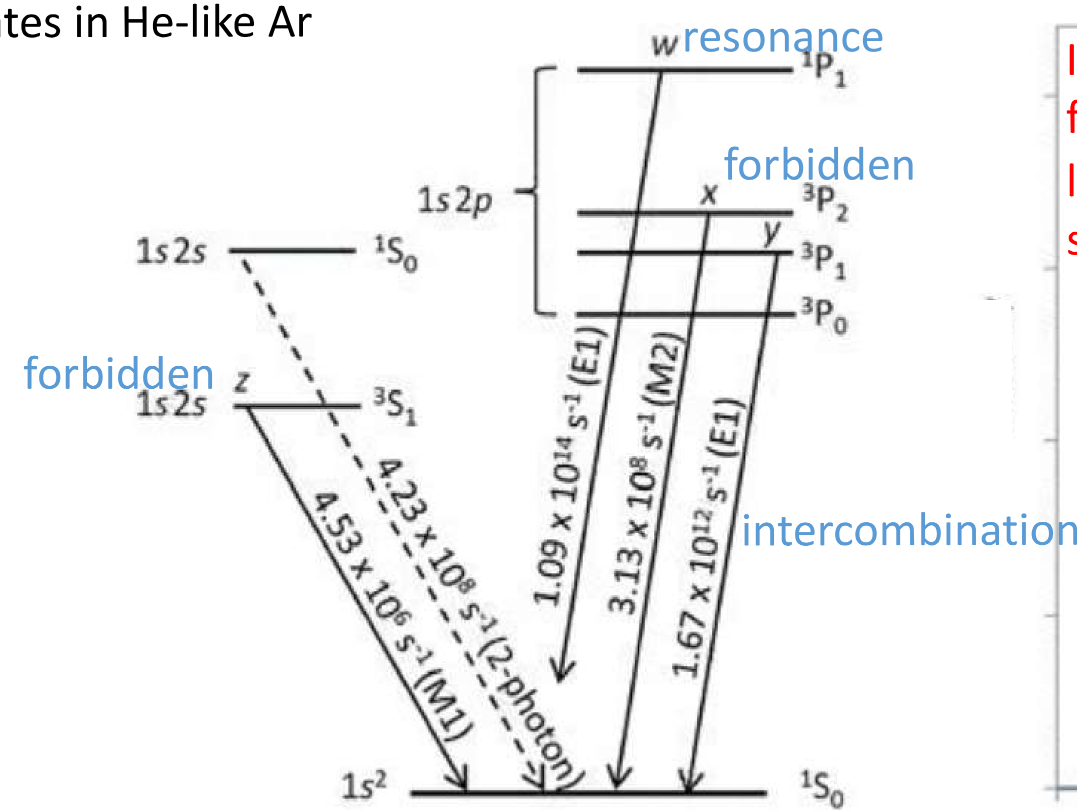
$$Z^* \sim (2/3)\,[Z_{nuc} T_e(\mathrm{eV})]^{1/3}$$

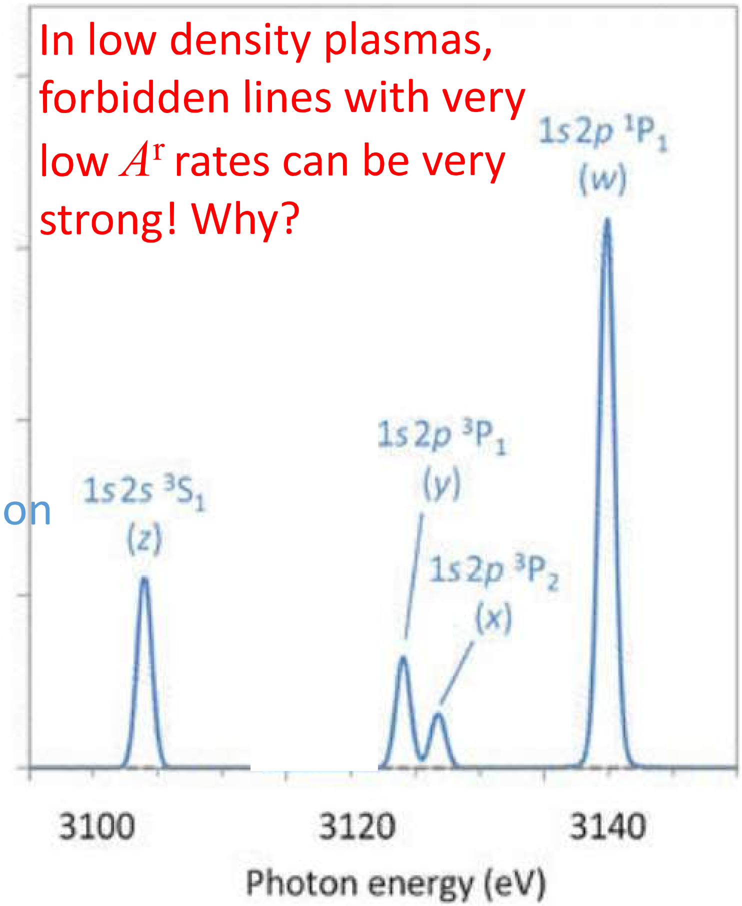


But it's better to <u>directly diagnose density</u> and use a trusted CR model

Sometimes, we can use line ratios to estimate density

States in He-like Ar





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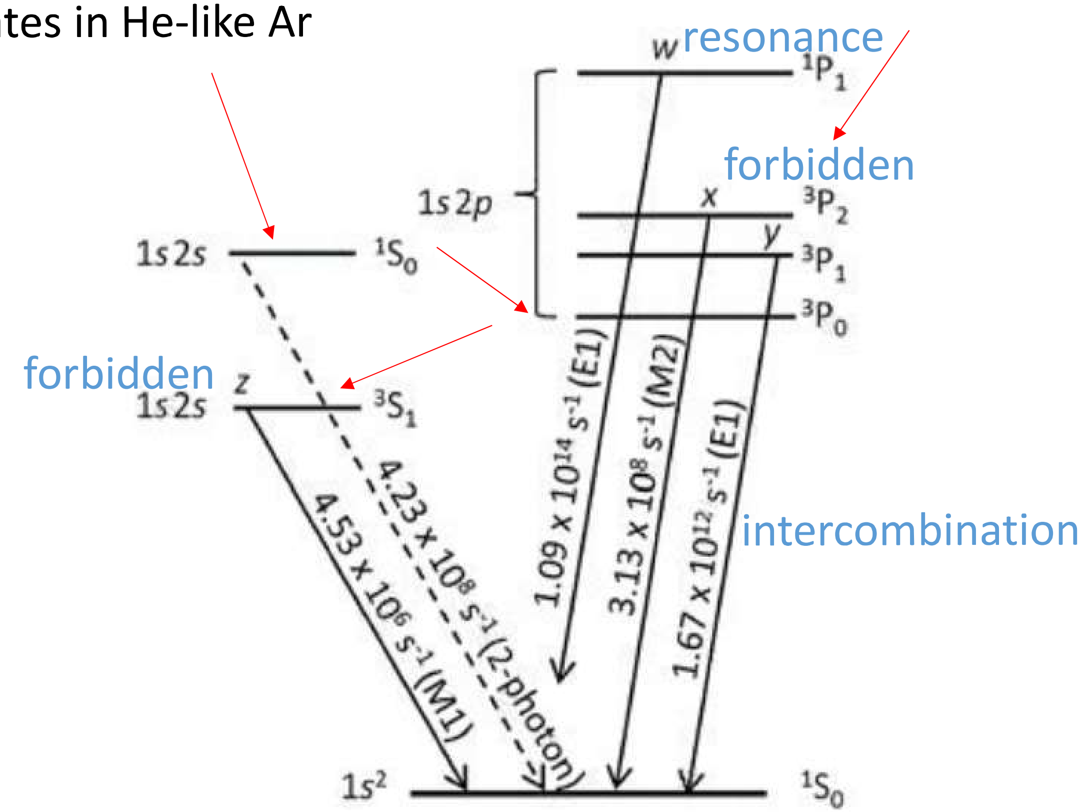

Recall that emissivity $\eta_{i0} = X_i A^r_{i0}$

The only way we can get a strong signal from a "forbidden" transition with a very small A^r_{i0} is to have a very large X_i

In the low density/coronal limit, low-lying "metastable" states with small A^r_{i0} can collect a significant fraction of the total population from radiative cascades

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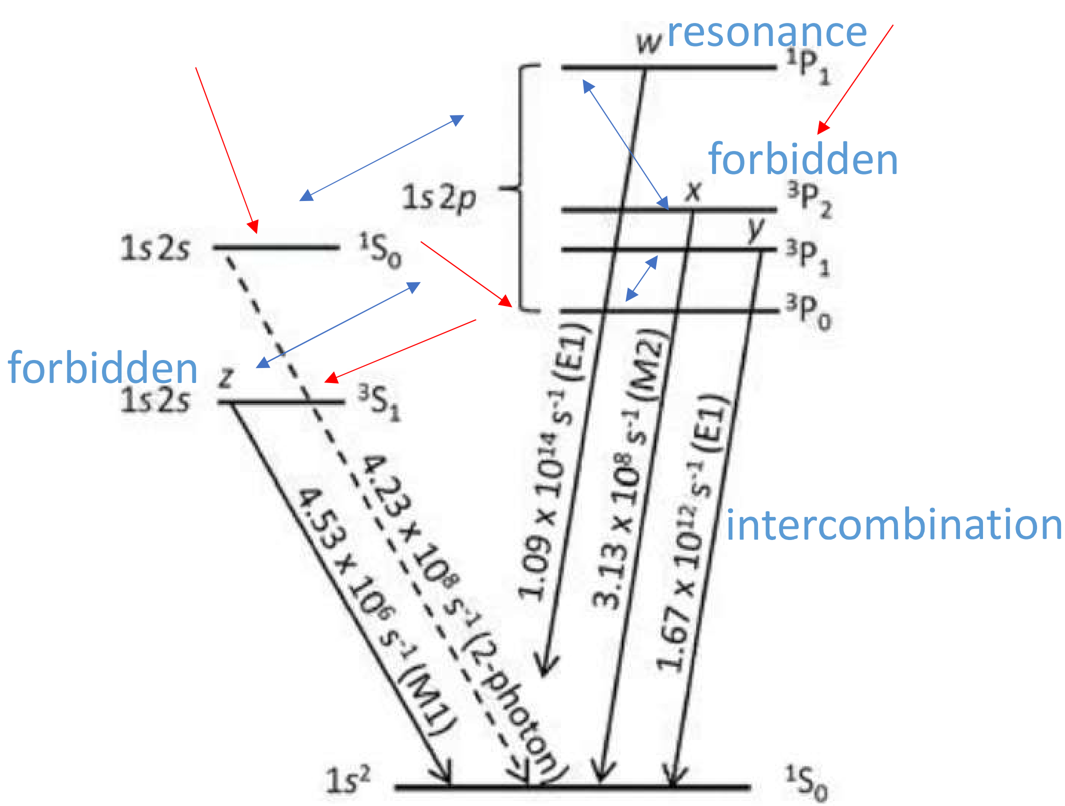

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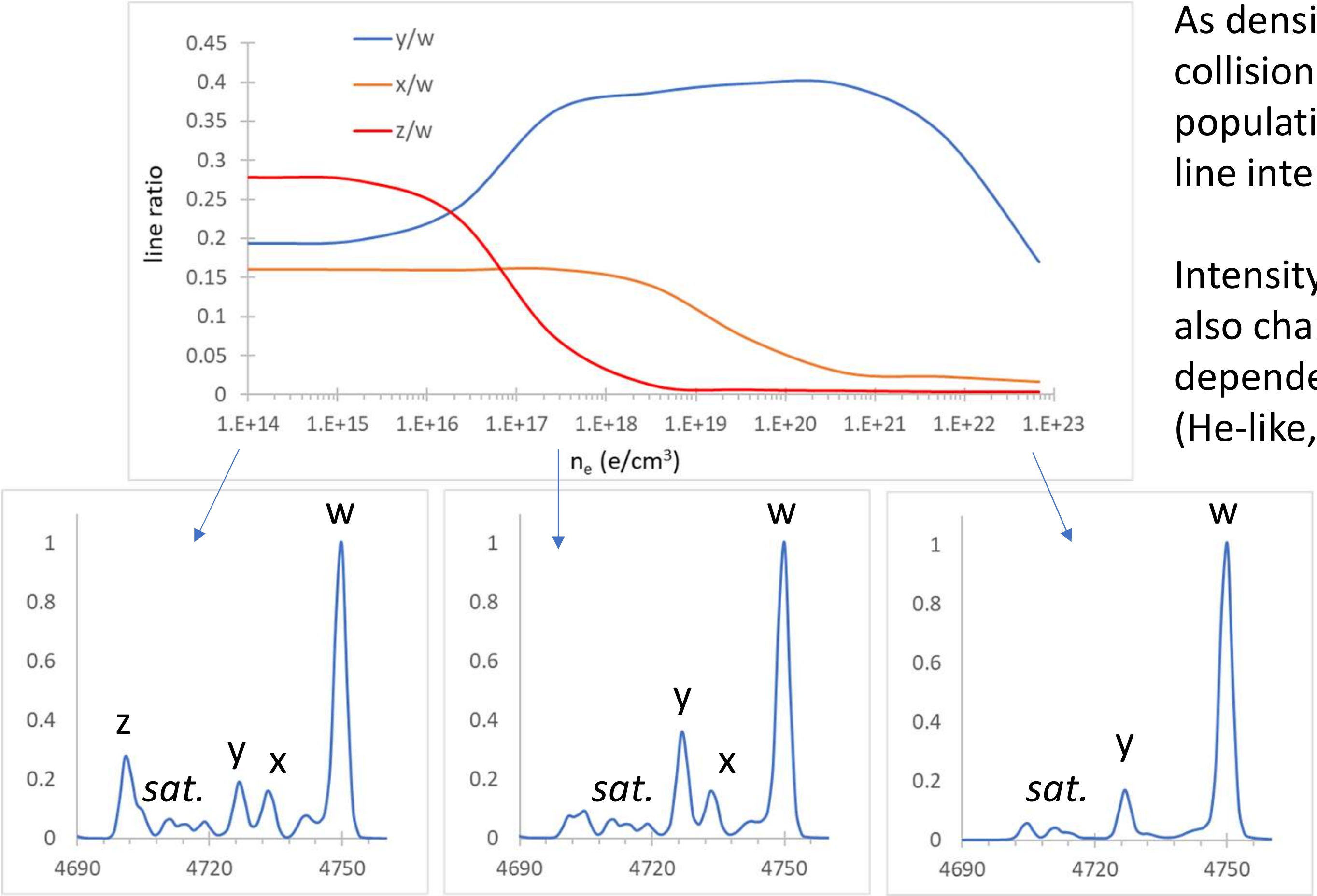
As density increases, collisional excitation and de-excitation move populations towards their LTE limit: $X_i = (g_i / g_0) \exp(- \Delta E_{i0} / T_e)$

Line ratios within ions are sensitive to the electron density

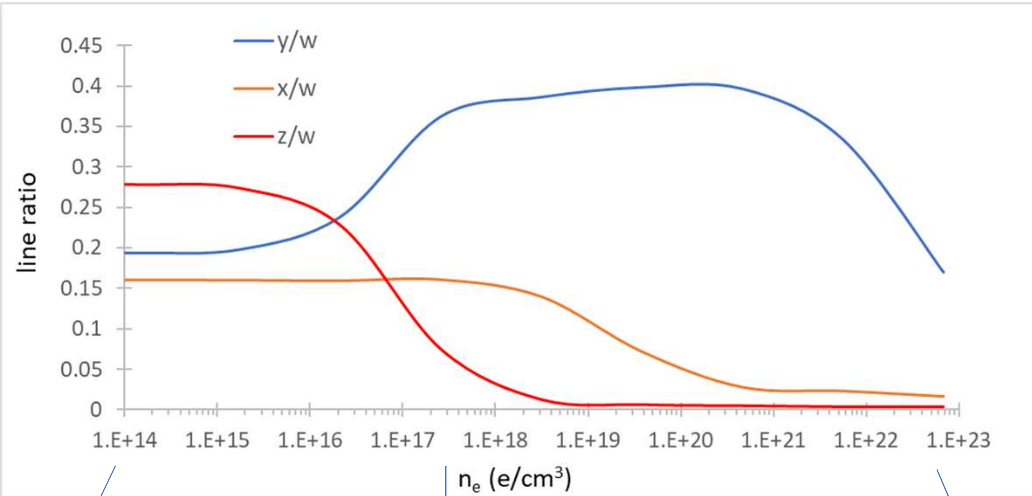


As density modifies the balance of collisional and radiative rates, state populations are redistributed and relative line intensities change significantly.

Intensity ratios within satellite complexes also change, but the most pronounced dependence is in closed-shell ions (He-like, Ne-like, Ni-like)

Density-dependent line ratios can be highly model dependent

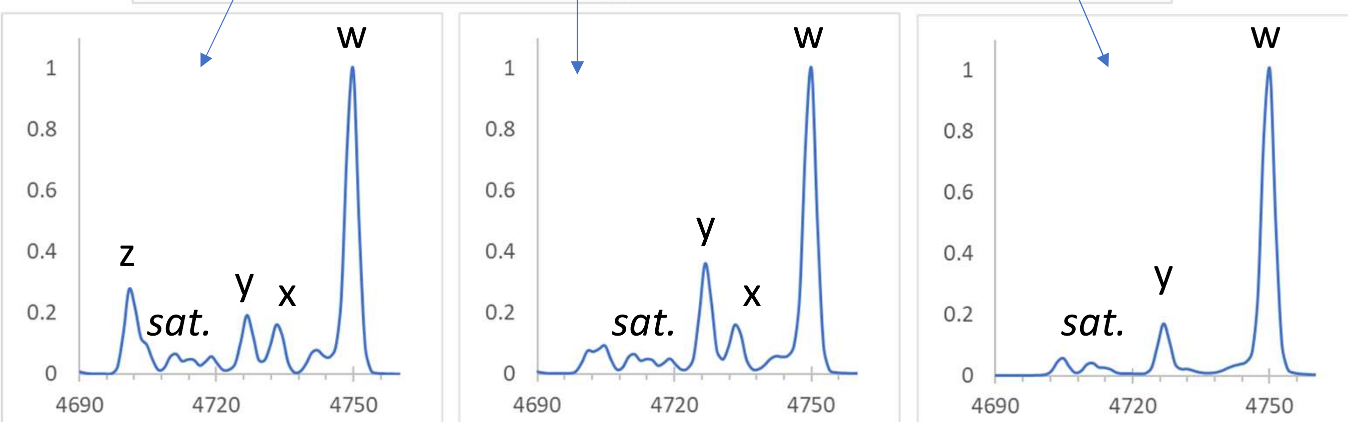
SCRAM



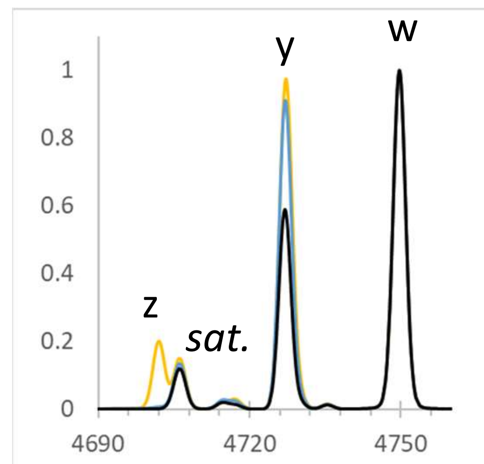
Why?

- Cascades depend on model completeness
- Accurate balancing of collisional and radiative rates requires detailed states and high-order transitions

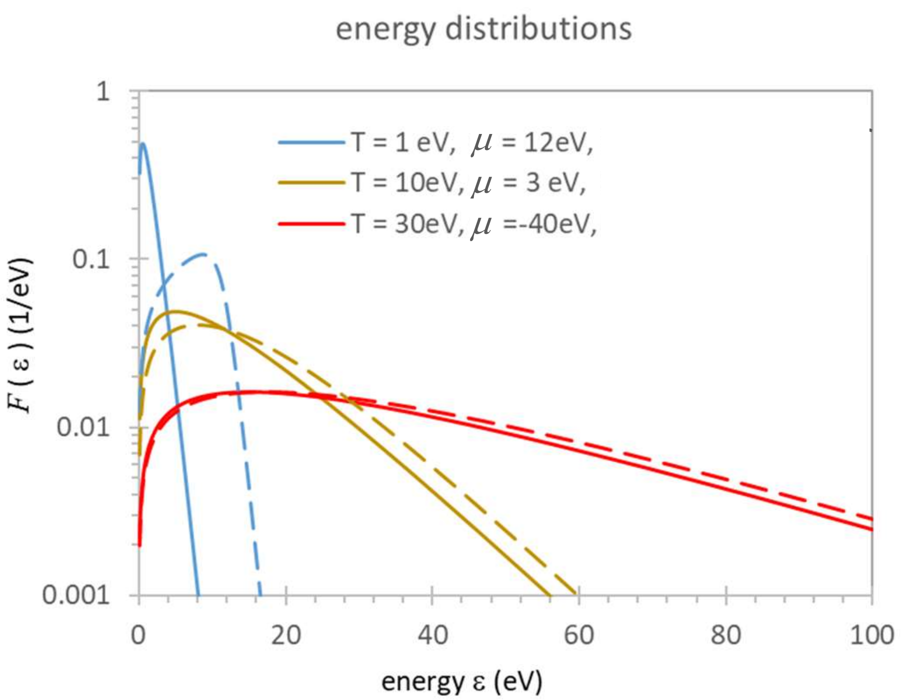
→ Use these ratios with care!



FLYCHK



At high density, degeneracy modifies free-electron distributions



Example: aluminum at 2.7 g/cm^3 (ni = 6x10^{22} cm^{-3})

In classical plasmas, the electron energy distribution is Maxwellian:

$$F_M(\varepsilon) = 2/T_e^{3/2}\,(\varepsilon/\pi)^{1/2}\,e^{-\varepsilon/Te}$$

At low T_e and high n_e, even free electrons are forced into close proximity. Since identical fermions cannot occupy identical states, the distribution function becomes:

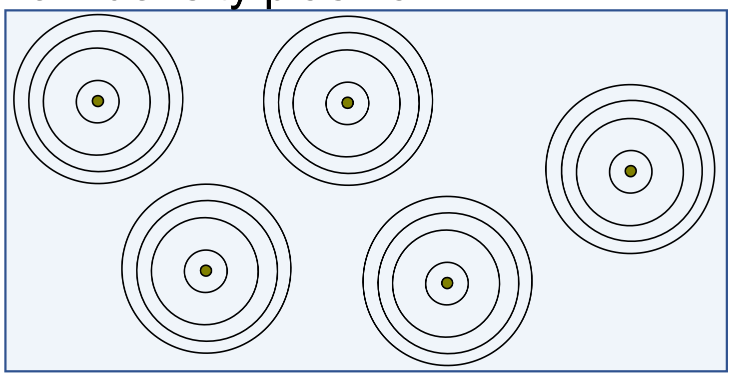
$$F_{FD}(\varepsilon) = (2\varepsilon)^{1/2} / (n_i Z^* \pi^2)[\,1 + e^{(\varepsilon-\mu)/Te}\,]^{-1}$$

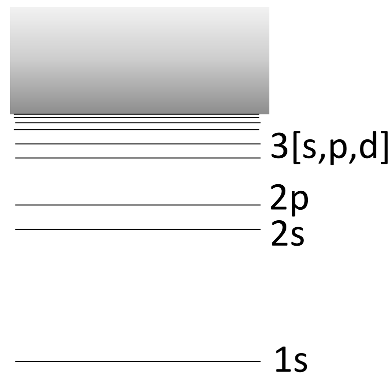
Note that we have introduced a new state variable, μ, which is directly related to the electron density.

These changes (and associated Pauli blocking) affect collisional rates – but not all CR models account for degeneracy effects

Very high plasma densities modify electronic structure

low-density plasma





In low-density plasmas, ions are isolated from each other and interact with a uniform background of ideal (uniform) free electrons

A puzzle:
statistical weights $g_n = 2n^2 \rightarrow \infty$ for high n
binding energies $\varepsilon_n = -13.6$ eV $(Z_{eff}/n)^2 \rightarrow 0$ for high n
then occupations $X_n = g_n \exp(-\varepsilon_n/T_e) \rightarrow \infty$ for high n
…. But infinities are always problematic

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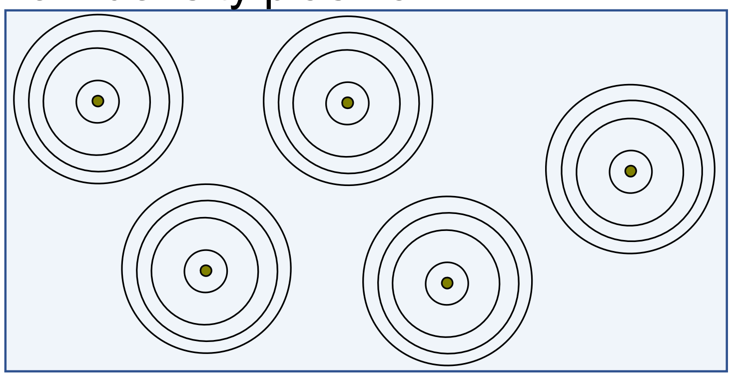

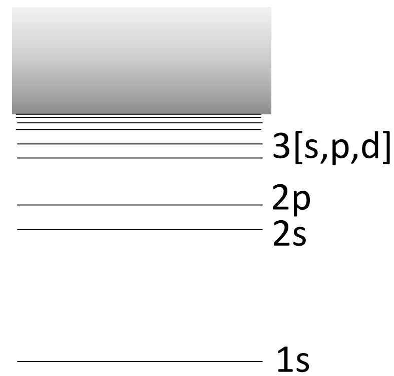

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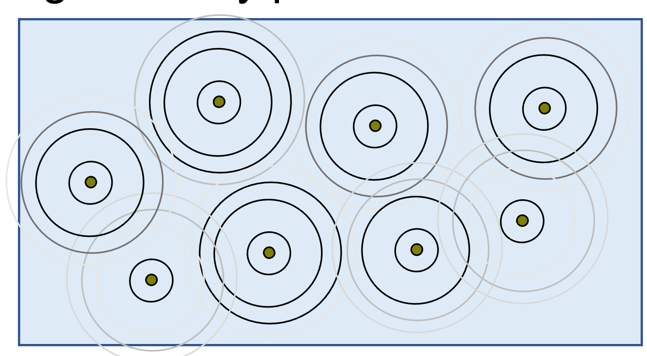
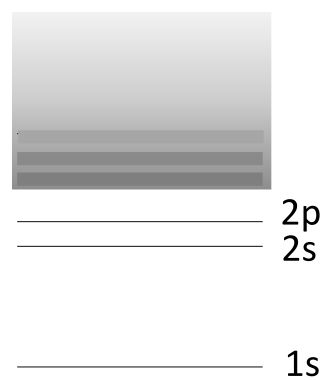
high-density plasma

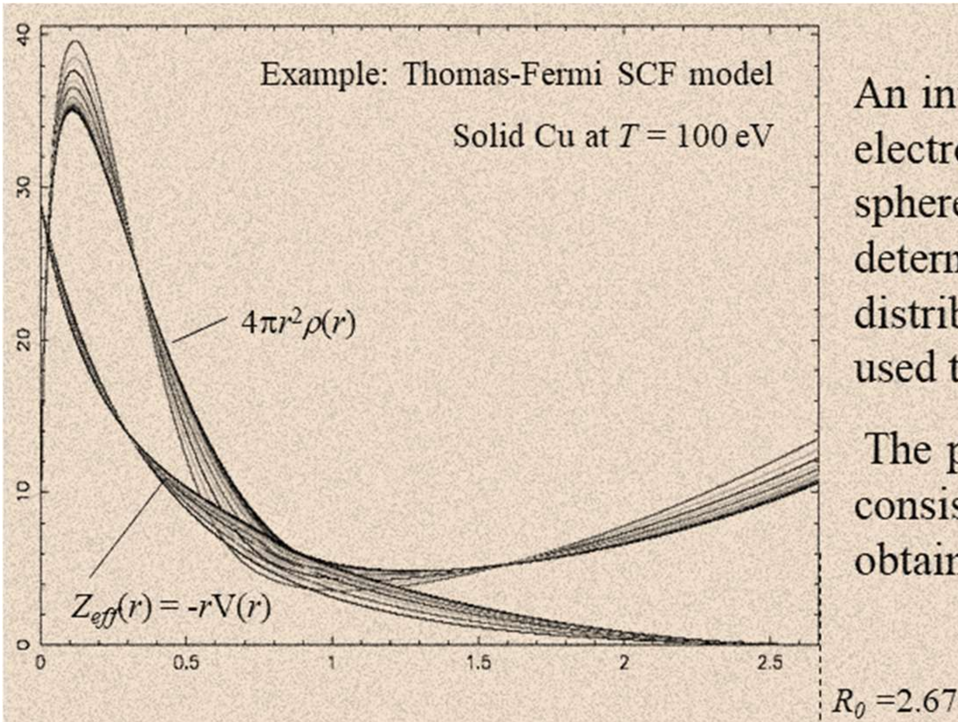




In high-density plasmas, neighboring ions perturb each other and respond to increasing screening from background free electrons: bound states “dissolve” into the continuum (related to band structure in solids) and their effective statistical weight decays (no infinities!)

A brief excursion into density functional theory

Self-consistent-field (SCF) Thomas-Fermi models developed in the 1920s treat electrons as a degenerate fluid that screens the nuclear charge, giving μ, Z*, pressure & energy



An initial guess is made for the electrostatic potential $V(r)$ in the ion sphere. This potential is used to determine an electron density distribution $\rho(r)$, which is in turn used to generate a new potential.

The procedure is iterated until self-consistent $V(r)$ and $\rho(r)$ are obtained, giving also Z_i and μ.

$$\rho \sim \int \frac{\sqrt{\varepsilon}}{1 + e^{(\varepsilon + V - \mu)/\tau}} d\varepsilon$$

$$\int_0^{Rws} 4\pi r^2 \rho \, dr = Z_{nuc}$$

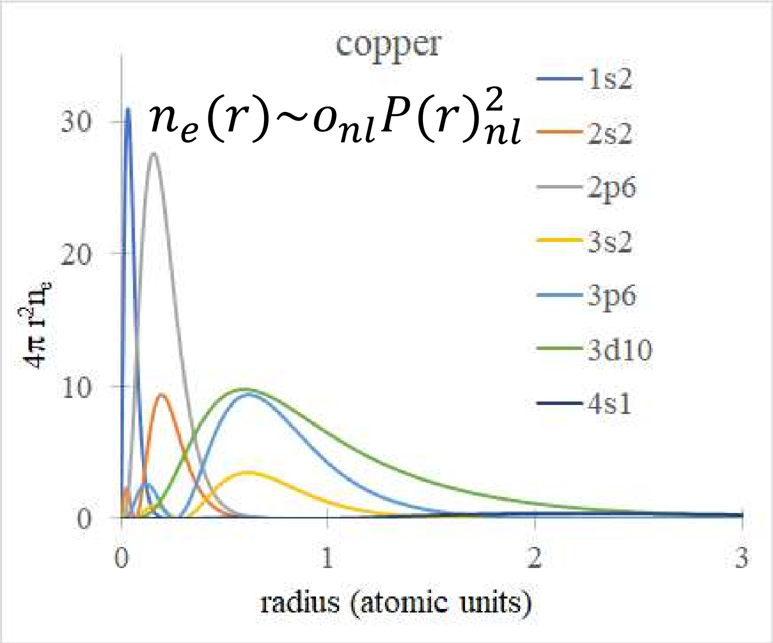
Neutrality in the Wigner-Seitz sphere is enforced by the chemical potential μ

$$\nabla^2 V = -4\pi\rho$$

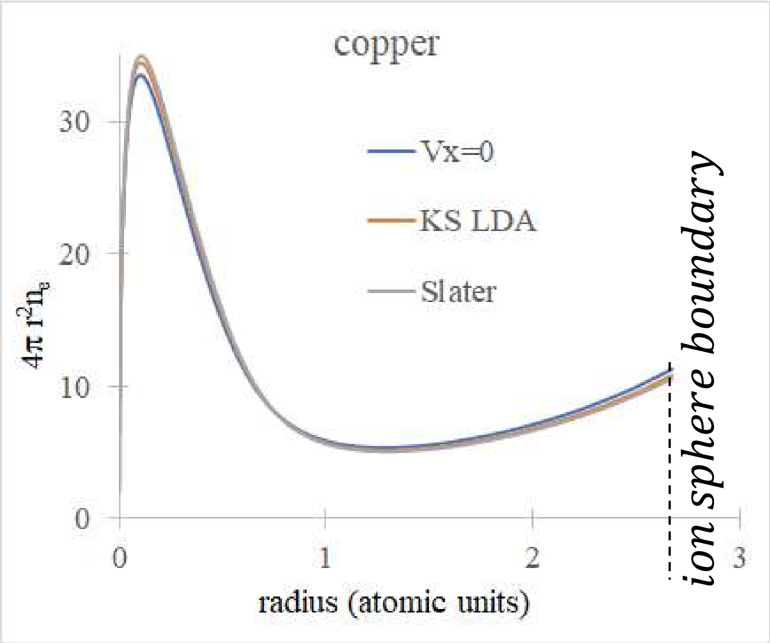
L Thomas, Proc Camb Phil Soc 23, 542 (1927); E Fermi, Z Angew Phys 48, 73 (1928)

The first average-atom models were the first DFT models

Through the 1930s and 1950s, development of Hartree-Fock-Slater methods in atomic physics (for isolated atomic structure calculations) led to improved exchange-correlation functionals for self-consistent field (SCF) models, and density functional theory (DFT) took off as its own field



Isolated, 0-T atomic Hartree-Fock:
quantum, for atoms & fixed configurations



High-T, ρ Thomas-Fermi SCF AA:
fluid adjusts to electron temperature & density

Slater, Phys Rev 81, 385 (1951); cf Cowan, Kohn & Sham Phys Rev 140, 1133 (1965)

In 1979, a fully quantum average-atom model was born

"ABANDON ALL HOPE YE WHO ENTER HERE"

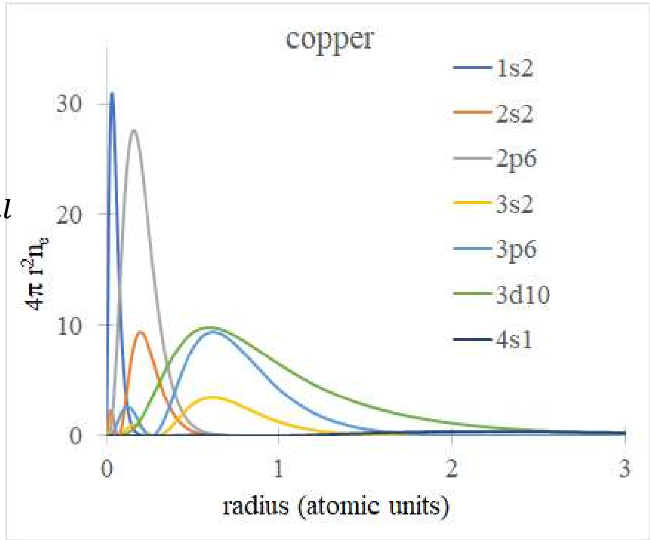
There have been many models of atoms in plasmas which have been proposed in the past fifty or sixty years. Perhaps my main advantage in undertaking the present work is that I know very little about them. This means going back to the fundamental principles and devising a model which makes the best possible use of our present day knowledge of electronic structure and of the electronic computers now available. I have called it INFERNO. **David Liberman**

J. Quant. Spectrosc. Radiat. Transfer Vol. 27, No. 3, pp. 335–339, 1982

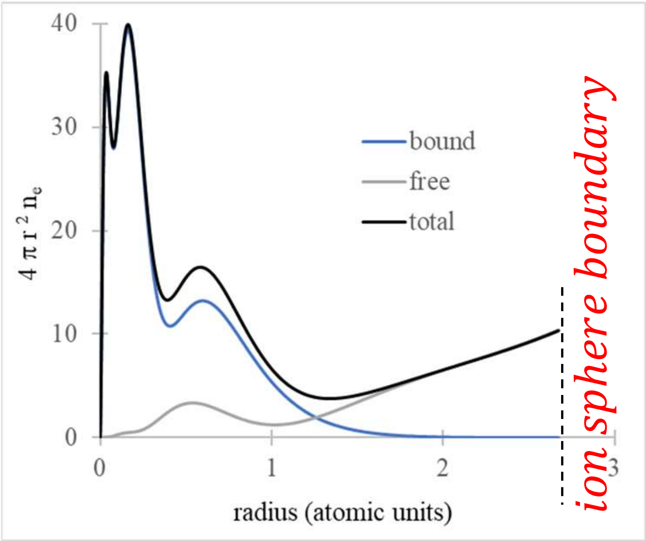
$$n_e(r) \sim \sum_{\varepsilon l} \int \frac{P_{\varepsilon l}^2(r)}{1+e^{(\varepsilon-\mu)/\tau}} dr$$

Isolated, 0-T

$n_e(r) \sim o_{nl} P(r)_{nl}^2$



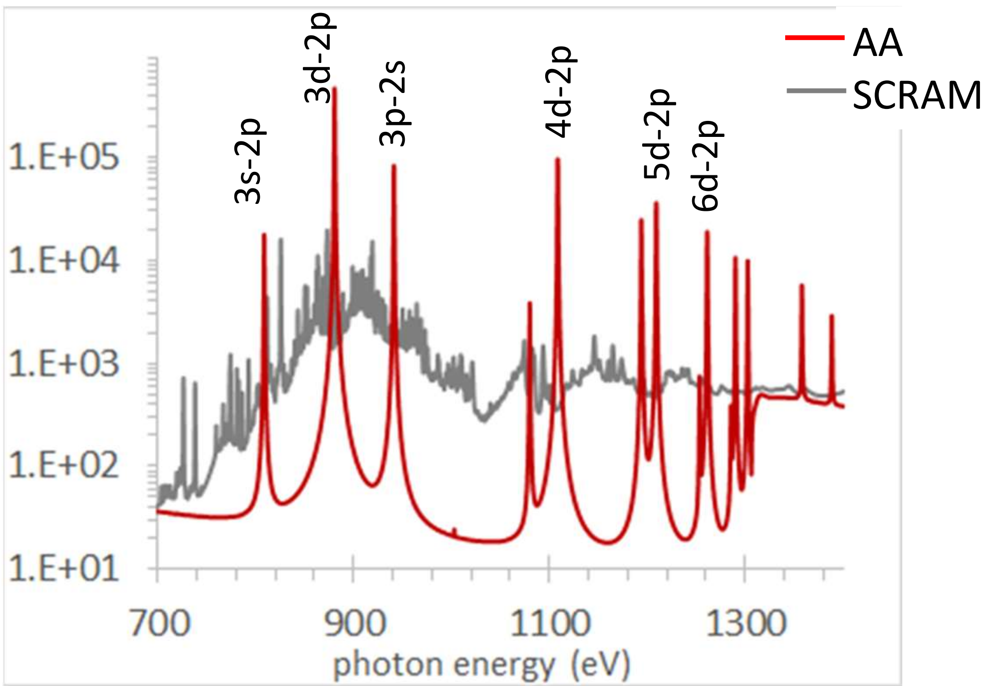
Low-T solid



Inferno *confines* the self-consistent potential in an ion sphere with Fermi-Dirac occupations for quantum electron orbitals: it natively accounts for density & degeneracy effects on atomic structure

Average-atom models oversimplify atomic structure & spectra

Example: L-shell iron



Average atom models have a single transition where detailed models have many

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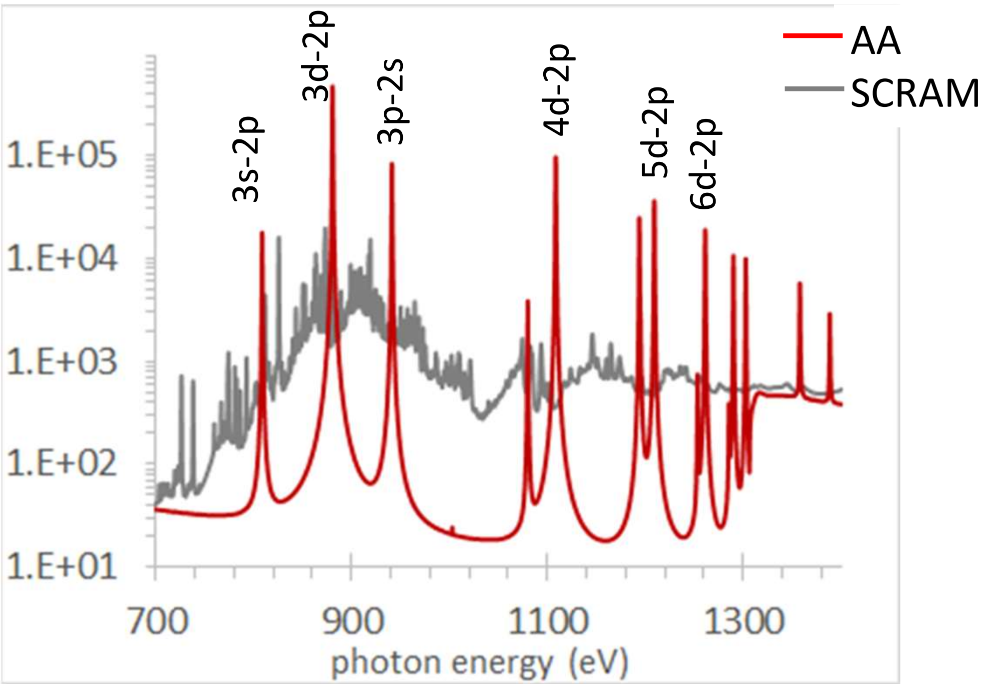
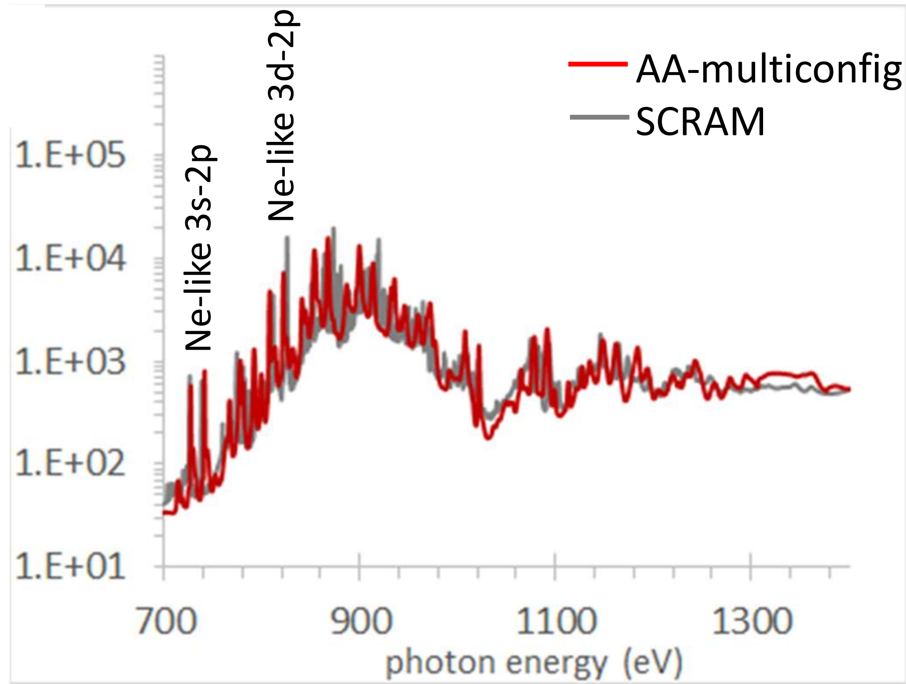


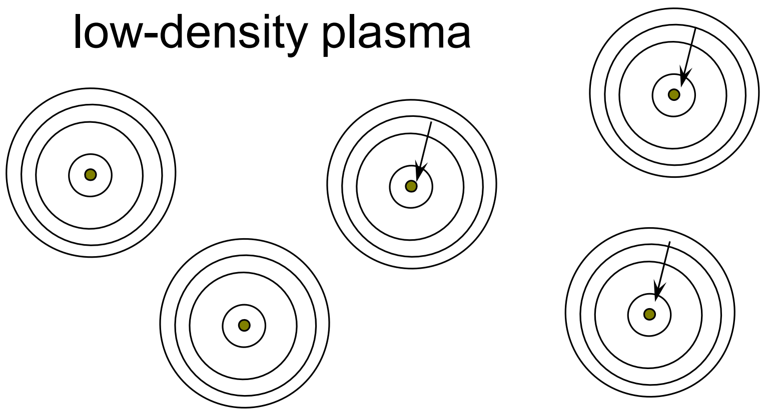


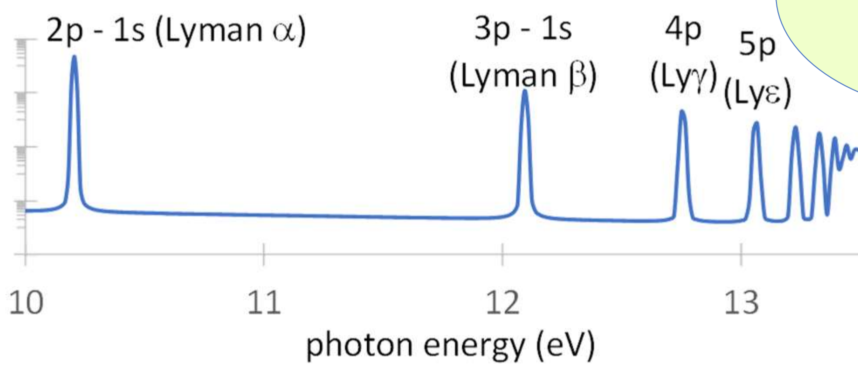
Average atom models have a single transition where detailed models have many

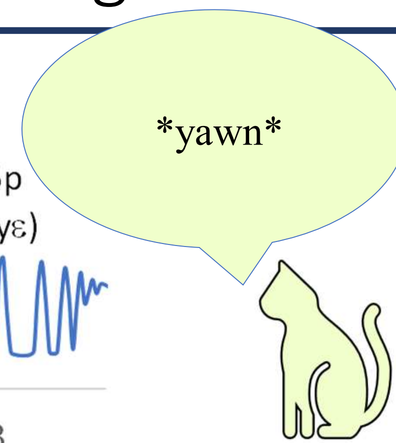
We can apply standard atomic physics methods to AA orbitals to refine spectra (see Rozsnyai, Hansen)

High-density environments also modify spectroscopic signatures

low-density plasma

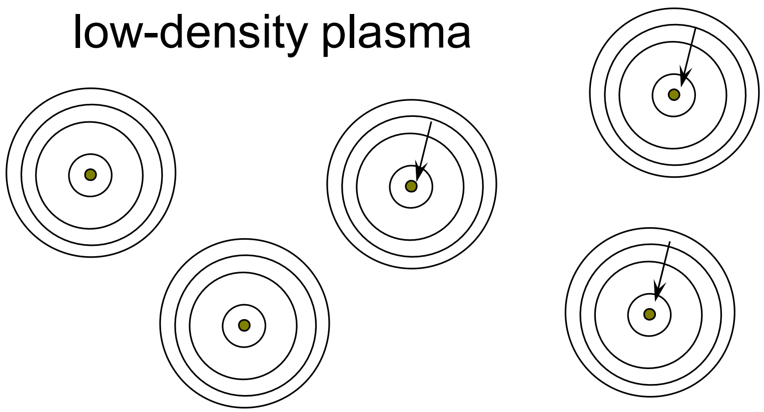


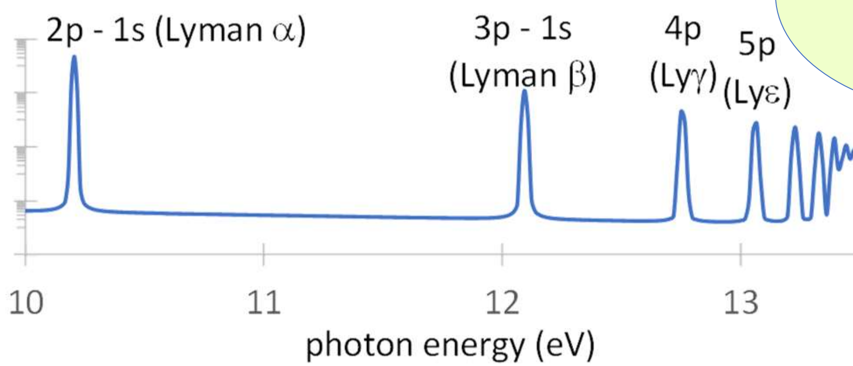


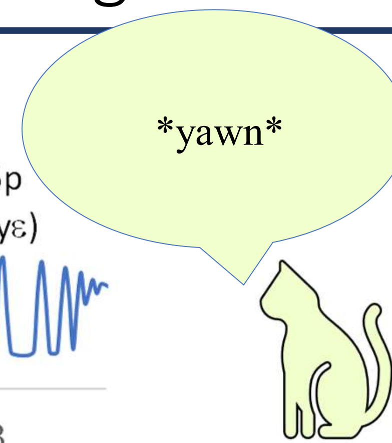


High-density environments also modify spectroscopic signatures

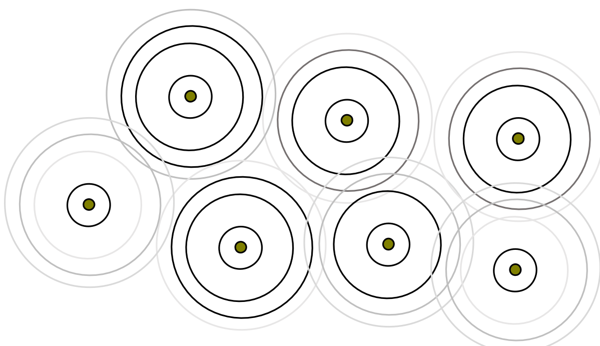
low-density plasma

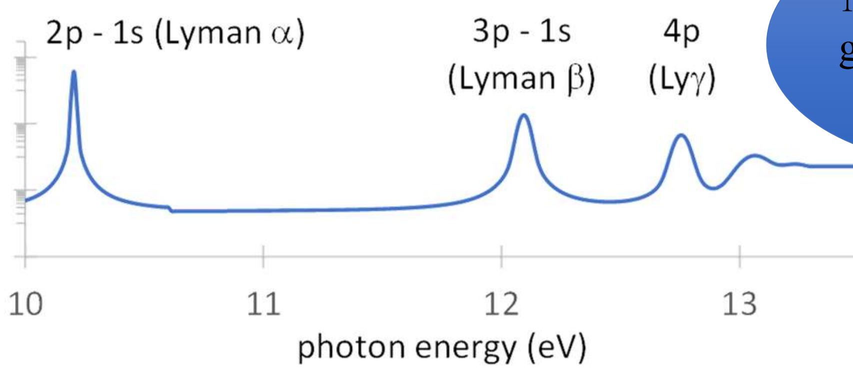


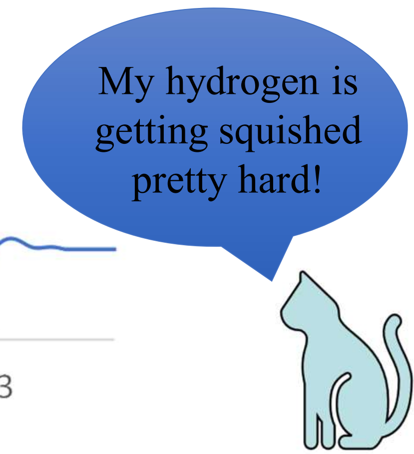




high-density plasma

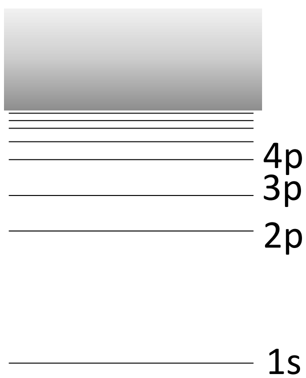


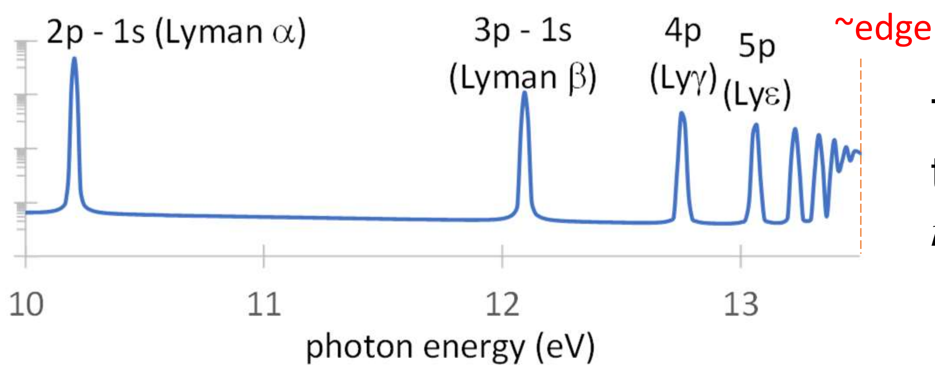




Free-electron screening leads to continuum lowering

lower density

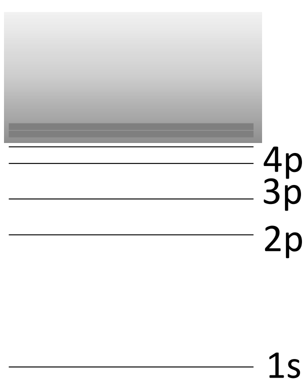


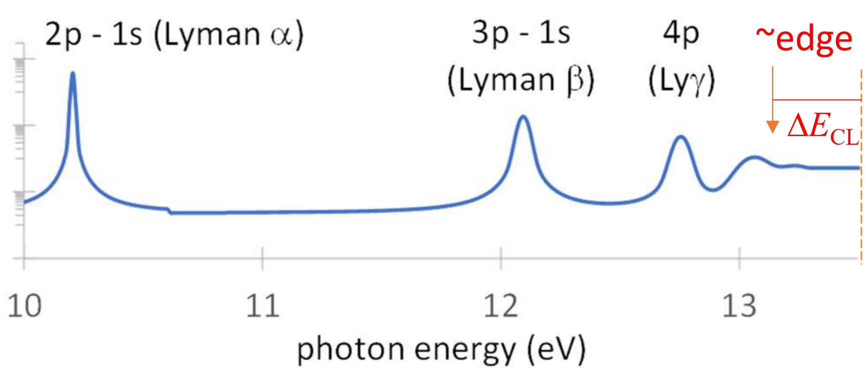


The Ingliss-Teller limit is the highest distinct n-line:
$n \sim 1.7(10^{22} / n_{\text{ion}})^{0.133}(Z^*)^{0.4}$

Continuum lowering is sometimes called ionization potential depression (IPD)

higher density

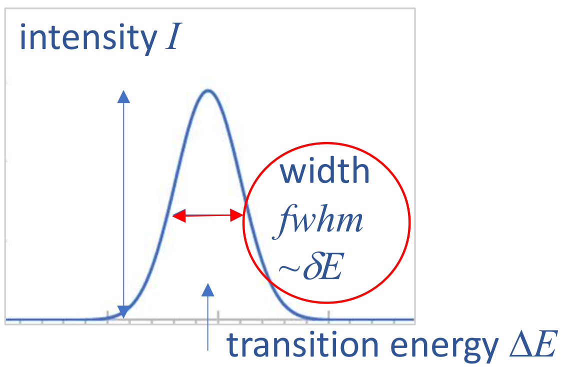




The continuum lowering energy is:
$\Delta E_{\text{CL}}\,(\text{eV}) \sim$
$7.5\,(Z^*+1)(n_{\text{ion}}/10^{22})^{1/3}$

Note that changes to ionization energies can significantly affect rates

Increasing collisional rates contribute to line broadening

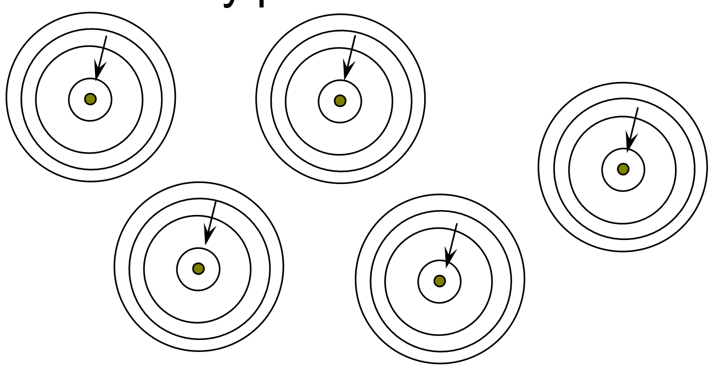


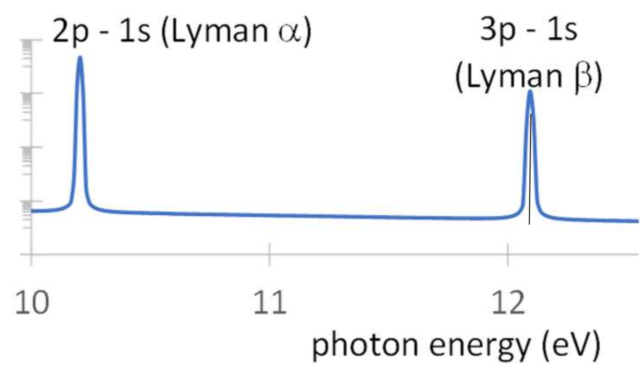
In cold low-density plasmas, lines have characteristic "natural" broadening from the uncertainty principle: $\delta E\ \delta\tau = \frac{1}{2}\hbar$
with state lifetimes $\delta\tau = 1/A^r \rightarrow \delta E \sim \frac{1}{2}\ \hbar\ A^r$

In dense plasmas, collisional rates decrease the state lifetimes, leading to a compensatory increase in the uncertainty of the transition energy (i.e. line width) $\delta E \sim \frac{1}{2}\ \hbar\ (A^r + C)$

Microfields from nearby ions also broaden lines: Stark effect

low-density plasma

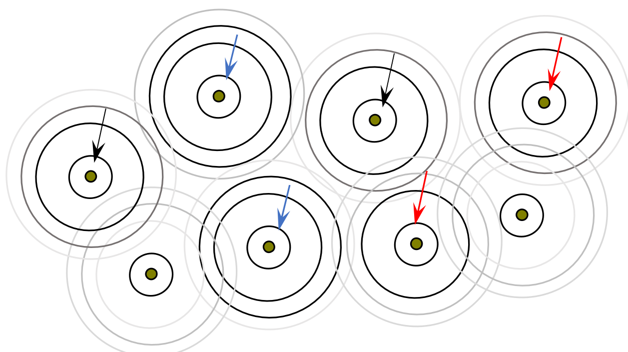


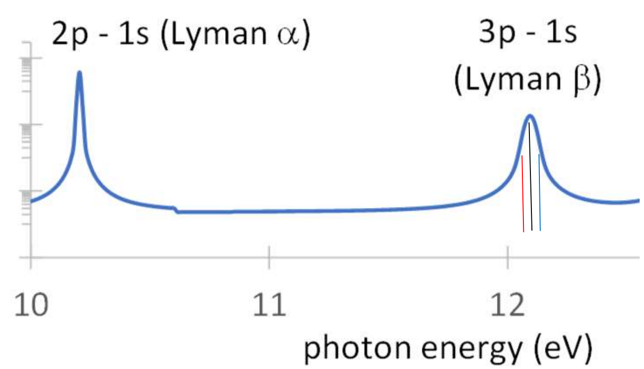


Changes to local potentials lead to Stark broadening:

Stark *fwhm* (eV)
$\sim 4.3\, n_{up}^{2}/Z_{nuc}\,(n_e/10^{22})^{0.58}$
for even $n_{up} - n_{down}$

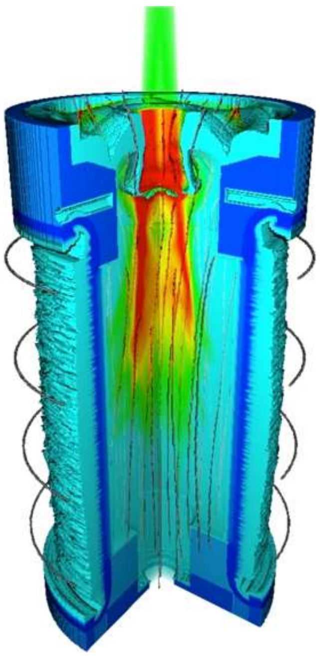
high-density plasma



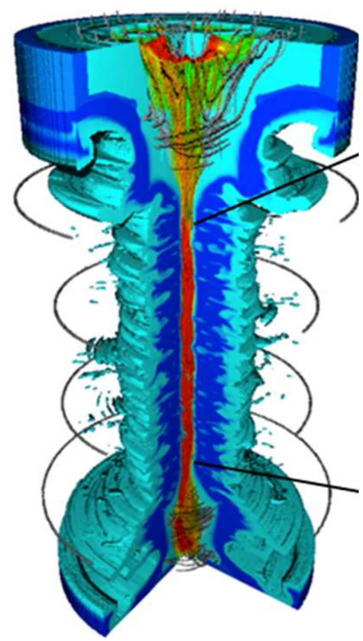


Application: a fusion plasma with mix and large gradients

Magnetized liner fusion (MagLIF) is a Be liner with ~100 ppm Fe impurities surrounding a pure-D_2 fuel core



The target is pre-magnetized with an axial B field and the fuel is preheated to ~100 eV with a kJ-class laser

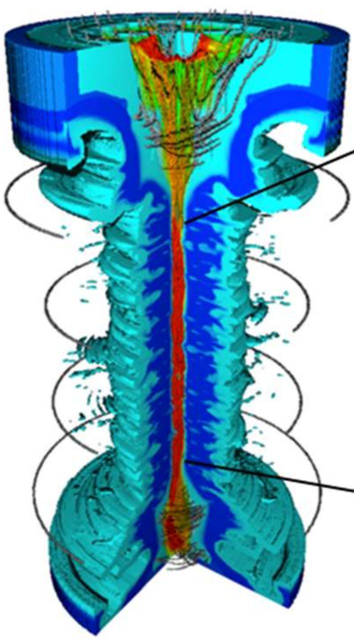


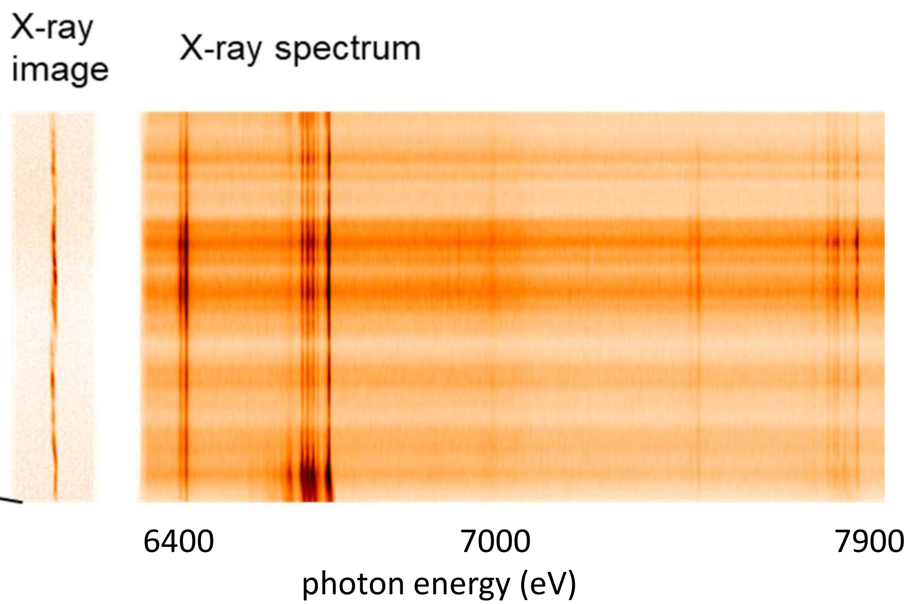
The target is imploded with the SNL Z machine's ~20 MA of axial current, which compresses and heats the fuel to ~ 3 keV temperatures

Simulation images courtesy C. Jennings

See Gomez et al, PRL 113, 155003 (2014)

Application: a fusion plasma with mix and large gradients



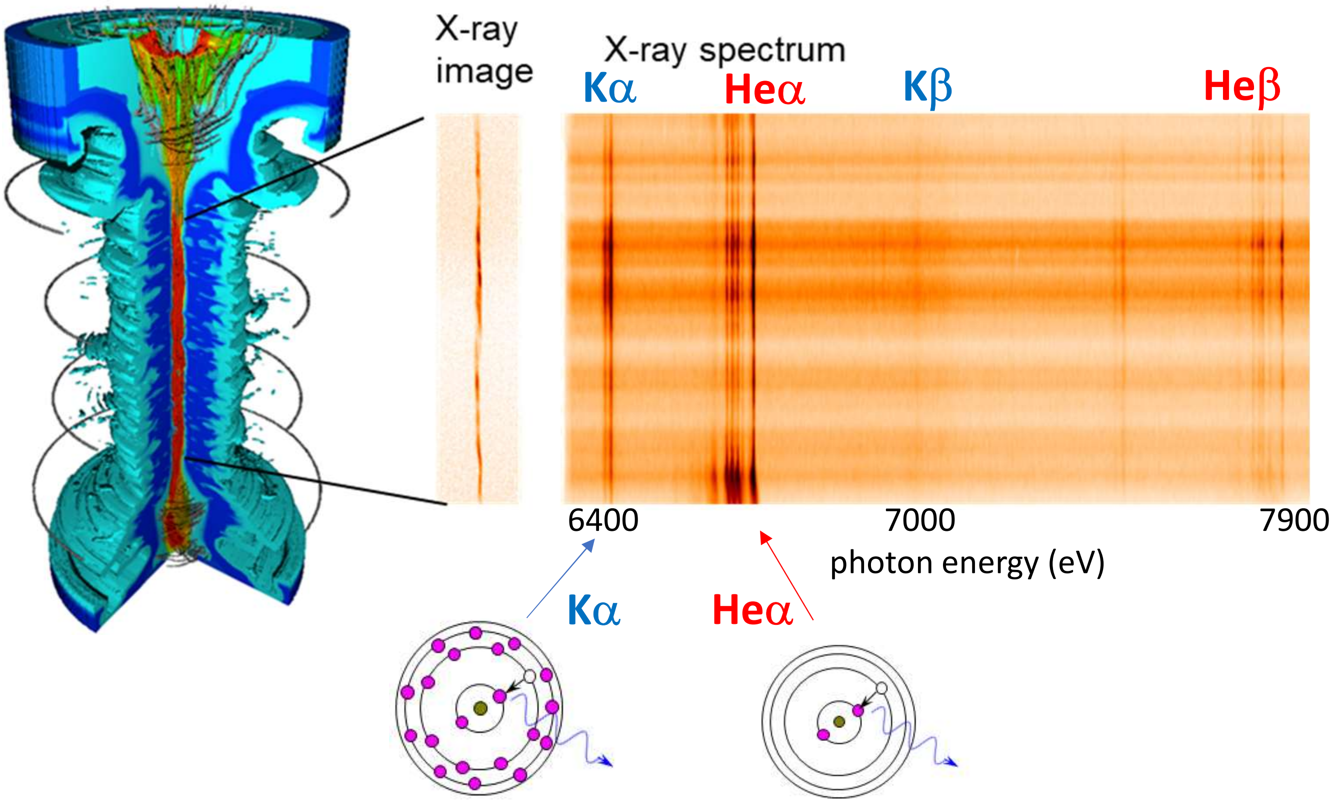


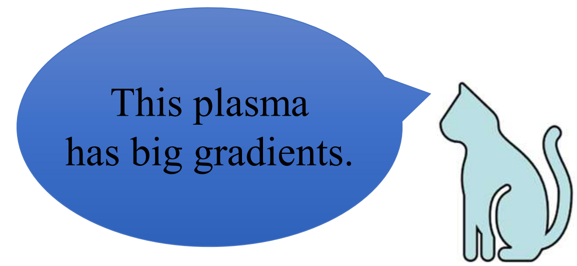
At stagnation, MagLIF targets produce fusion yields of ~1 kJ equivalent DT neutrons and copious x-rays

What can we learn from a high-resolution spectrum of the stagnating MagLIF plasma?

Spectrum courtesy E. Harding: absolutely calibrated 10 within 1 eV in energy, resolution E/ΔE ~ 4000 (< 2 eV width)

What can we learn from a high-resolution MagLIF spectrum?



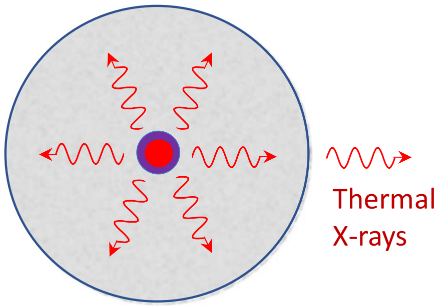


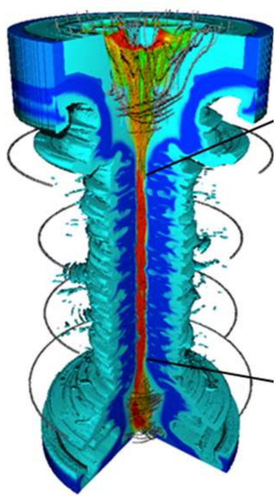
He-like iron K-shell lines:
some of the liner, with its 100 ppm iron impurities, must reach ~ keV temperatures: mix?

Neutral iron K-shell lines:
Some of the liner stays cold (~10 eV) and is photoionized by radiation from the hot core, producing fluorescence

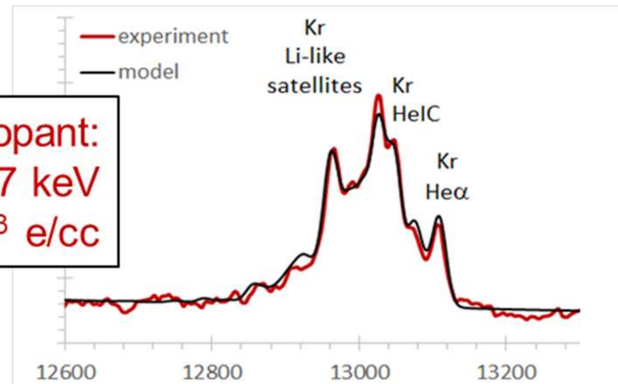
A closer look at the thermal lines gives details of gradients in the fuel

Top-down view



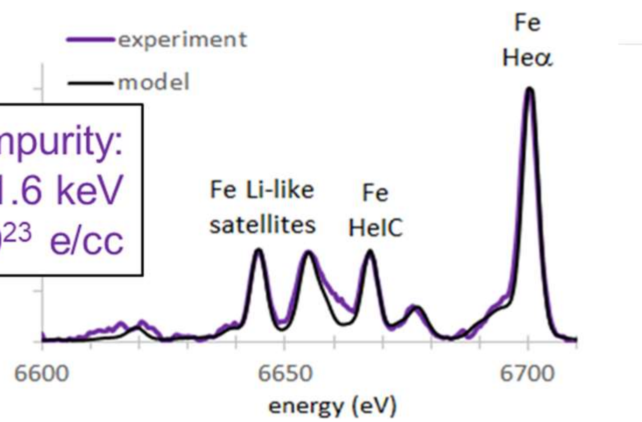


Kr fuel dopant:
T_e^{core} = 2.7 keV
n_e^{core} = 1×10^{23} e/cc



Line ratios from Kr and Co co-mixed with the bulk fuel indicate core conditions

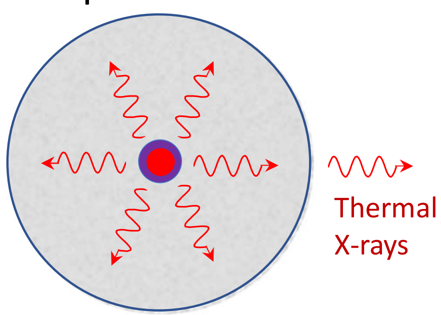
Fe liner impurity:
T_e^{mix} = 1.6 keV
n_e^{mix} = 2×10^{23} e/cc

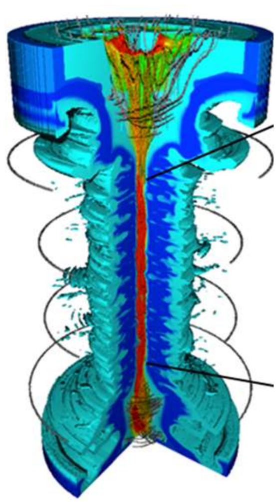


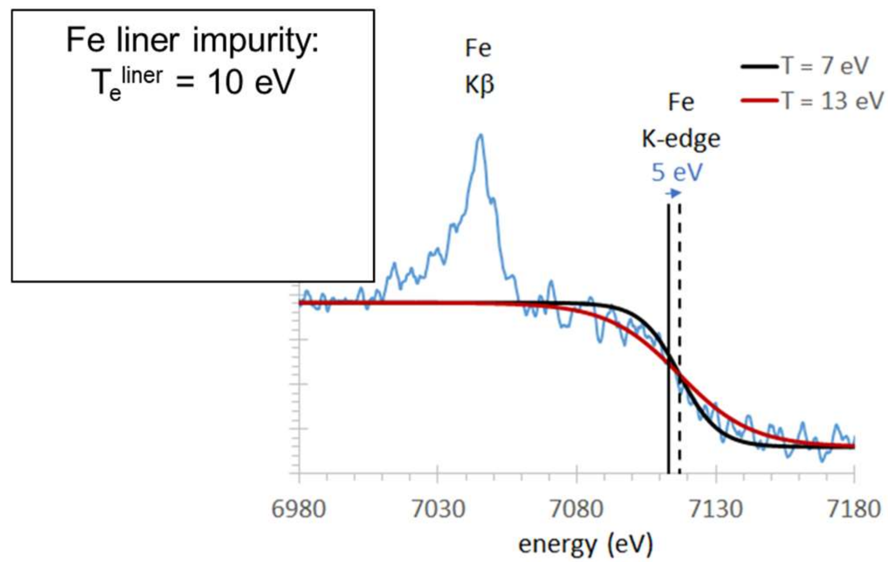
Line ratios from Fe and Ni liner impurities indicate a cooler, denser mixed layer

A closer look at fluorescence lines & edges tells us about the liner

Top-down view



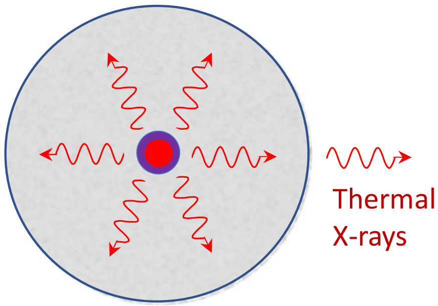


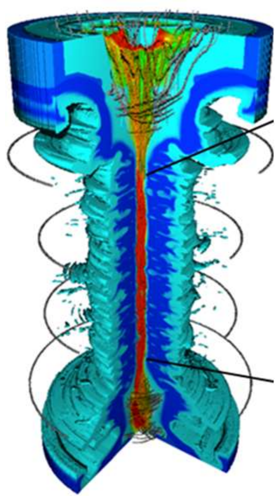


The shape of the absorption edge follows the degenerate Fermi electron distribution, indicating temperature

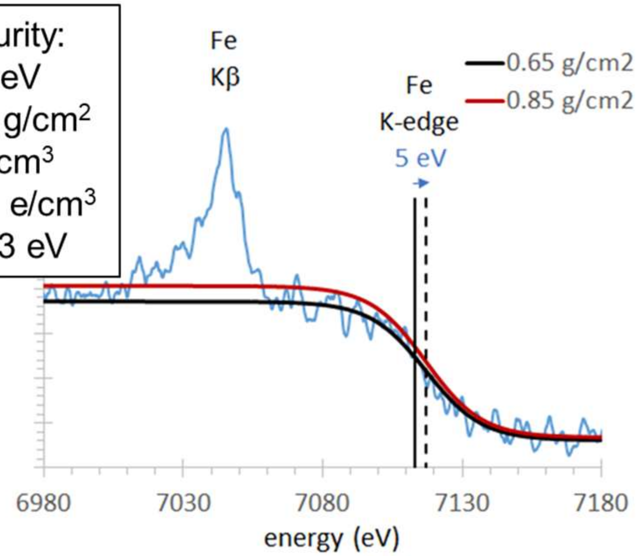
A closer look at fluorescence lines & edges tells us about the liner

Top-down view





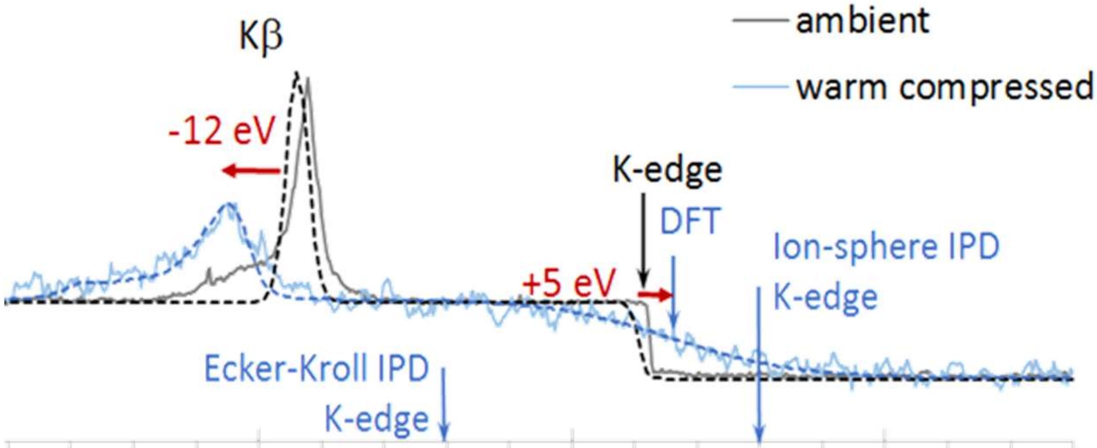
Fe liner impurity:
$T_e^{liner} = 10$ eV
$\rho R^{liner} = 0.75$ g/cm^2
$\rho^{liner} = 15$ g/cm^3
$n_e^{liner} = 2\text{x}10^{24}$ e/cm^3
$\rightarrow E_{Fermi} = 53$ eV



The <u>depth</u> of the absorption edge gives us the areal density of the liner; mass conservation and imaging constrain the density

An intriguing red shift in Kβ tells us about fundamental atomic physics

Modeled spectra (dashed) vs. measured (solid)



Looking closely at the Kβ line, we see a significant shift to lower energy in the MagLIF spectrum (blue) compared to reference data (black)

But Kβ *always only* shifts to higher energies under ionization

Roughly, ionization reduces screening of the core while compression (to 8x solid!) increases screening from excess free electrons → leading to the red shift

This result required a high-resolution, exquisitely calibrated spectrometer and enabled us to test self-consistent atomic models and *ad-hoc* continuum lowering models

S. Hansen, E. Harding *et al, HEDP* **24**, 39 (2017)

Spectroscopy is a “Rosetta Stone*” for the universe

- Spectroscopy unites the very small (quantum) with the very large (astrophysics) and very strange (extreme environments in HEDS)

- Spectroscopy tells us:
  - what elements compose a plasma
  - what conditions a plasma is in: densities and temperatures
  - can inform our understanding of atomic structure in extreme plasma environments

- Frontiers of spectroscopy include:
  - Self-consistent treatment of dense plasma effects (e.g. ICF) on atomic structure, free-electron densities of states, rates, and line shapes
  - Highly non-equilibrium systems (e.g. XFEL-pumped plasmas) where bound and free-electron occupations are non-thermal and highly transient

*Peter Beiersdorfer