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Title: Superconfiguration Calculations Using Green's Functions

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Superconfiguration Calculations Using Green's Functions

A New STA Opacity Capability at Los Alamos

Matt Gill, XCP-5
Chris Fontes, Charlie Starrett
09/06/2022

Los Alamos NATIONAL LABORATORY EST.1943

NNSA National Nuclear Security Administration

Outline

- Overview of Superconfigurations and STA Opacity
- Superconfiguration electronic structure calculation
  - SCF Procedure
  - **Green's function approach**
- Influence of different continuum electron treatments
  - Challenges for high density plasmas
- Comparisons to Z experiments

Superconfigurations

- Group together atomic subshells into supershells
- Superconfiguration (SC) determined by supershell occupation
- SCs can represent many different configurations

Configurations:

$C = \prod_{s \in C} s^{q_s}$, e.g. $C = 1s^2 2p^1$

Supershells:

$$\sigma = \prod_{s \in \sigma} s$$

Superconfigurations:

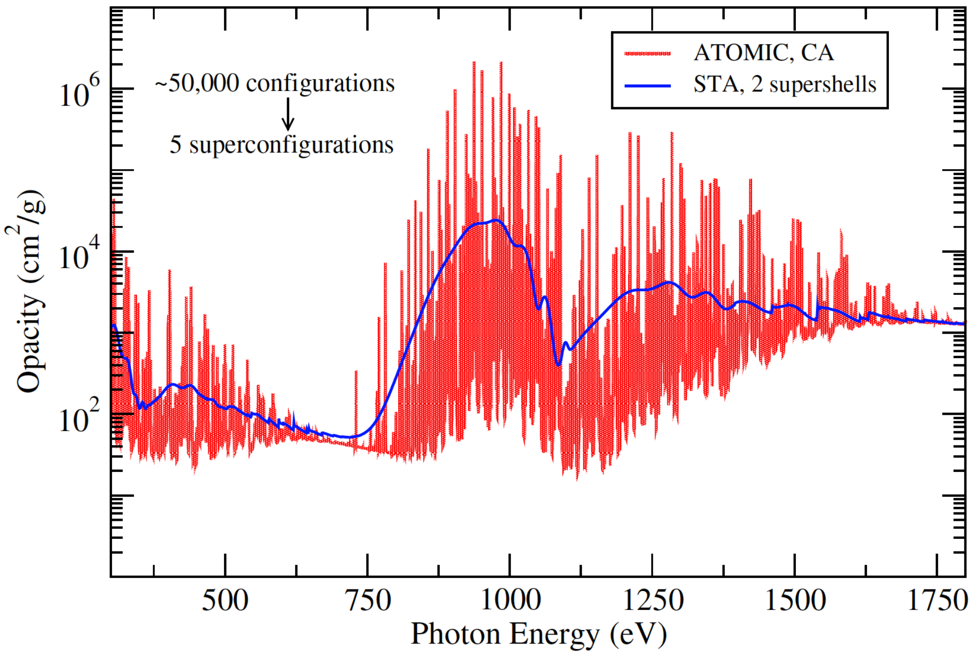
$$\Xi = \prod_{\sigma} \sigma^{q_\sigma}$$

e.g. $\Xi = (1s\ 2s\ 2p)^3 (3s\ 3p\ 3d)^2 (4s \ldots 10k)^0$

Bar-Shalom et al, Phys. Rev. A, **40** (6), 1989

STA Opacity

Iron Opacity, 175 eV, 0.001 g/cm^3



- SC gives one "average" electronic structure to represent all constituent configurations
- Super Transition Array (STA) formalism used to generate representative opacities from SCs
- STA spectra replace many transition arrays with fewer, broader STAs
- Statistical moments (average opacity, transition energies, variance of arrays) approximately the same as that of "true" spectra
- Refine SCs → Resolve Opacity

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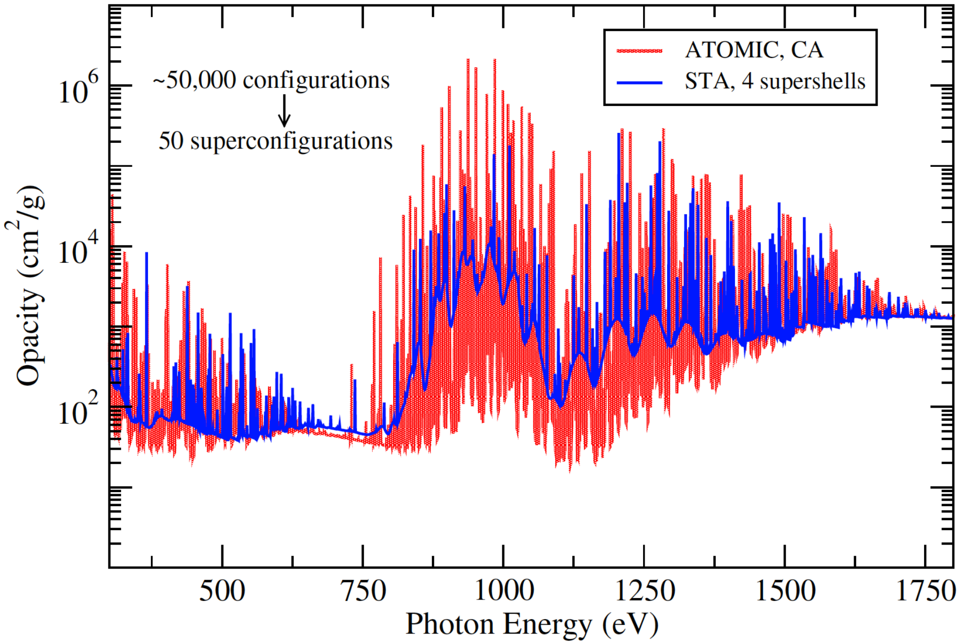

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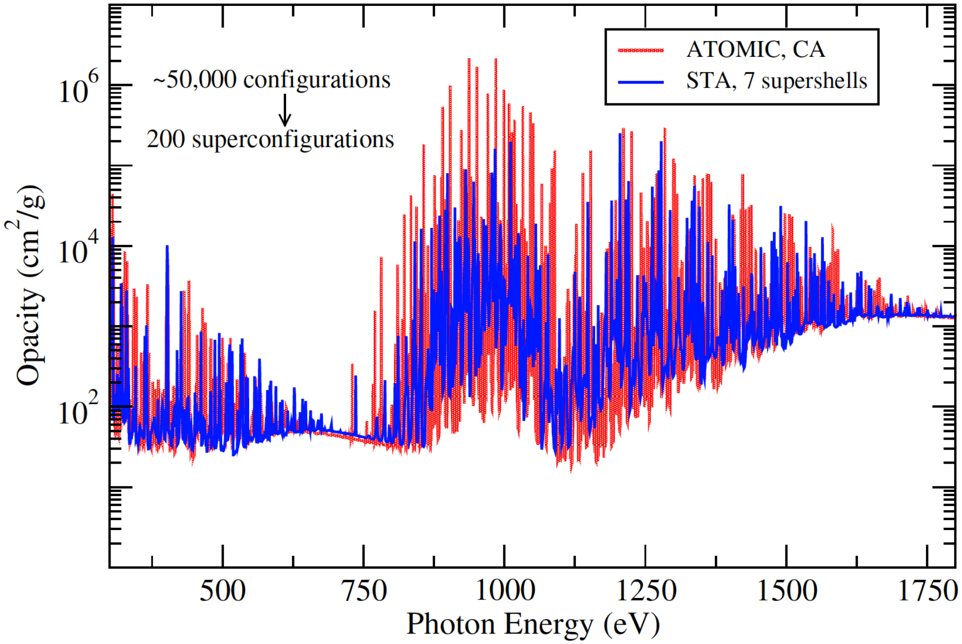

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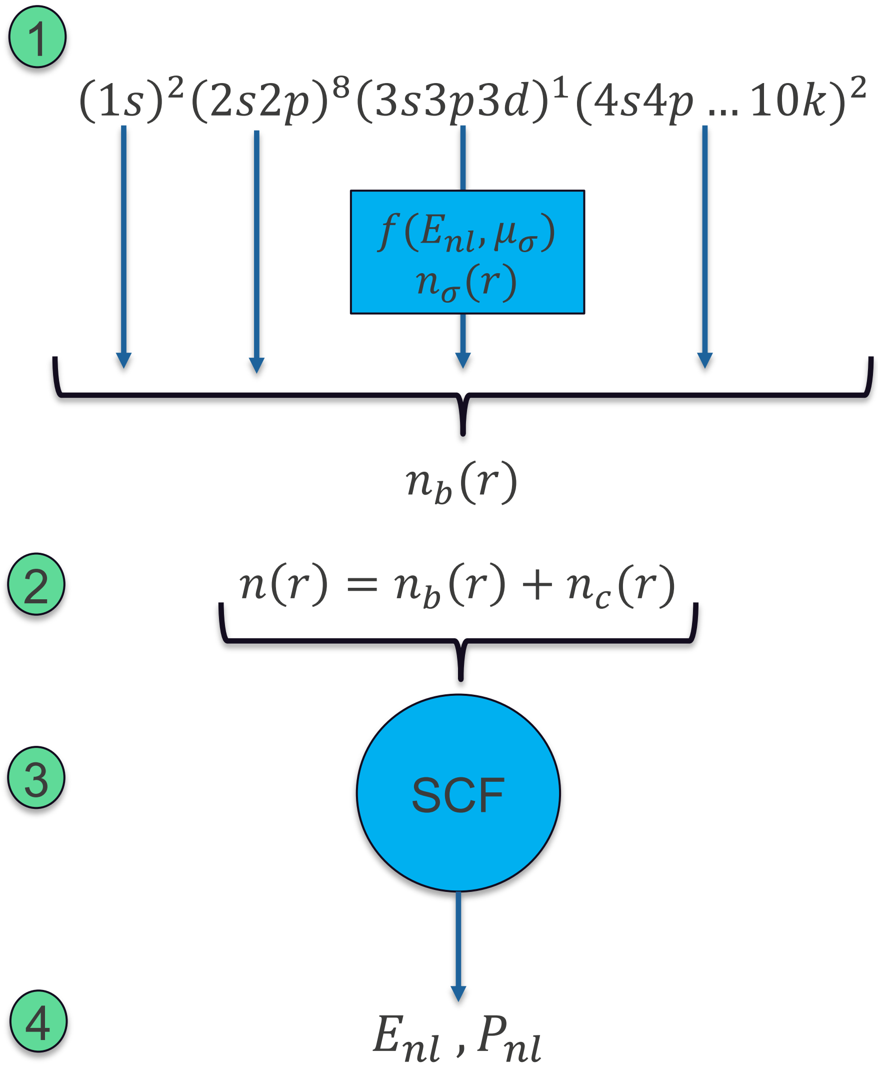
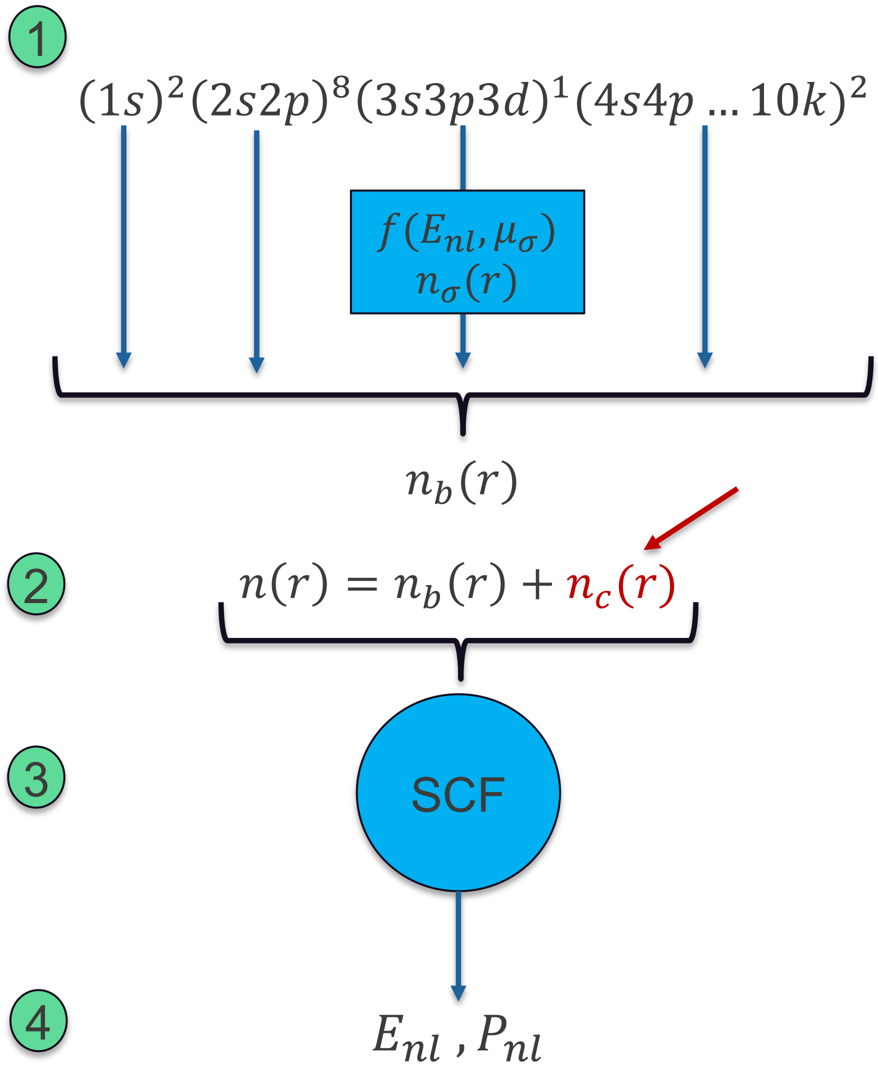
Superconfiguration Electronic Structure



- Ion-Sphere Model
  - Finite sphere size based on plasma mass density (cuts off high n orbitals)
  - Plasma screening through boundary condition (fields perfectly screened outside sphere)
  - Sphere charge neutral
- Solve self-consistent field (SCF) equations
  - Nonrelativistic and fully relativistic modes
  - Orbitals and occupations used to construct electron density

Superconfiguration Electronic Structure



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Continuum Electron Density

$$n_c(\vec{r}) = \int_{E_{min}}^{\infty} dEf(E)|\psi_E(\vec{r})|^2 = -\frac{1}{\pi}\Im \int_{E_{min}}^{\infty} dEf(E)TrG(\vec{r},E)$$

$$= 2k_B T \sum_i \Re\, TrG(\vec{r},z_i) + \frac{1}{\pi}\Im \int_C dzf(z)TrG(\vec{r},z)$$

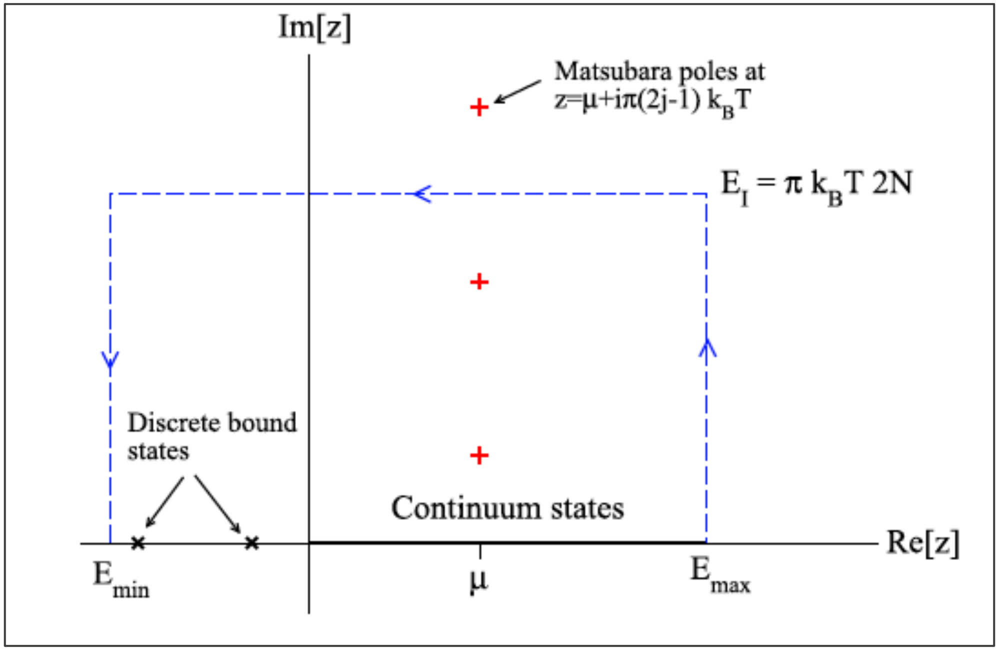
- Integration over real energy changed to integration over complex energy
- Integrand is smoother the larger the imaginary part of the energy, $\Im(z)$
- No approximation – Cauchy's integral theorem

Starrett et al, Comp. Phys. Comm., **235** (50-62), 2019

Complex Contour

Cauchy's integral theorem: $\int_{CC} f(z)dz = 2\pi i \sum_i Res\, f(z_i) = \int_C f(z)dz + \int_{-\infty}^{\infty} f(z)dz$

$$n(\vec{r}) = 2k_B T \sum_i \Re\, TrG(\vec{r}, z_i) + \frac{1}{\pi}\Im \int_C dz f(z) TrG(\vec{r}, z)$$

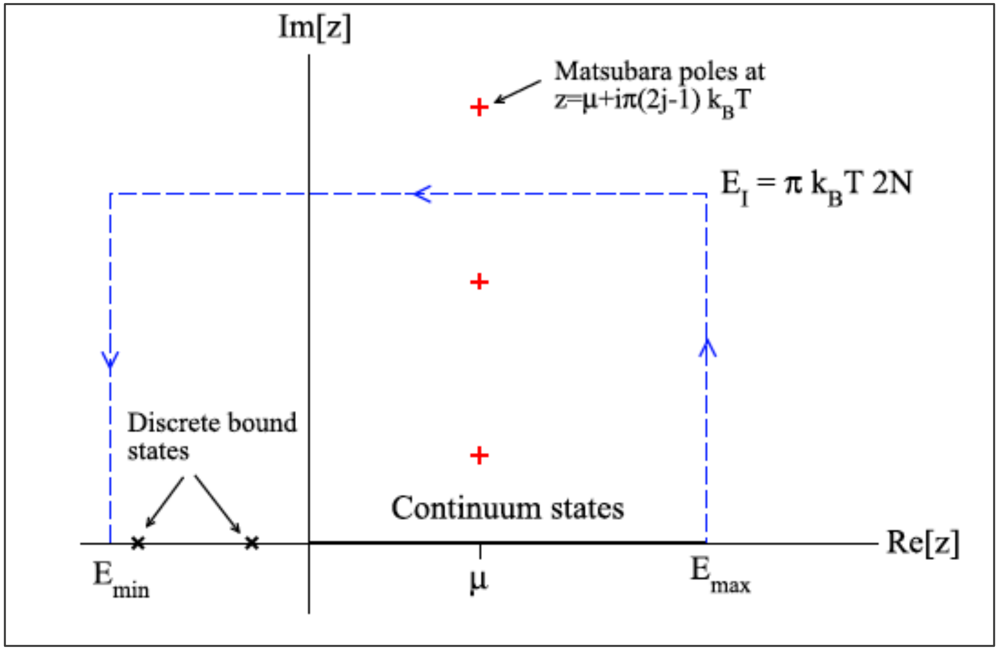


Starrett, HEDP, **16** (18-22), 2015

Complex Contour

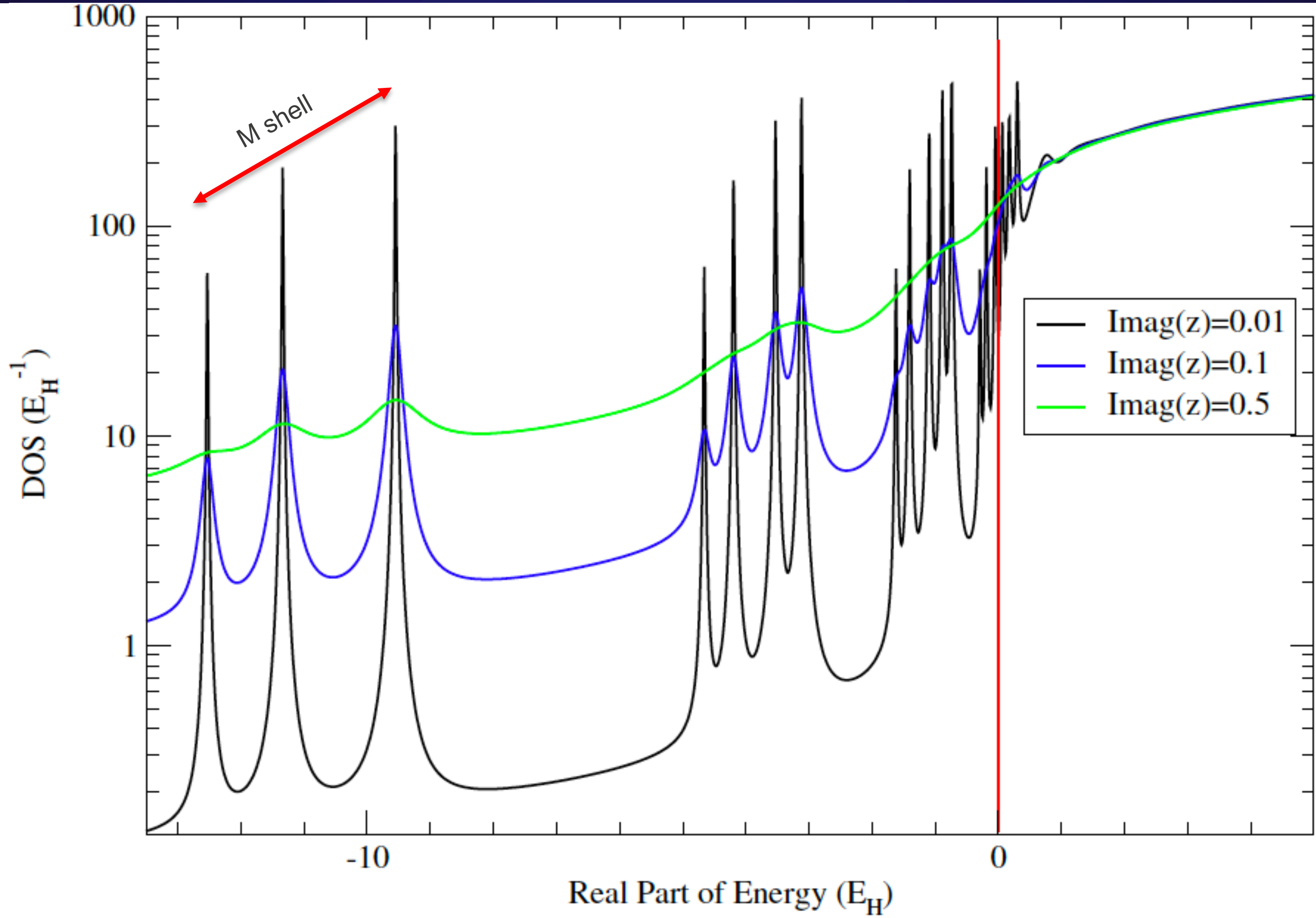
Density of States (DOS) along contour broadened by Lorentzian: FWHM proportional to imaginary part of energy

$$\chi(z) = \frac{1}{\pi}\int_{-\infty}^{\infty} d\varepsilon' \frac{\Im(z)}{(\varepsilon-\varepsilon')^2 + [\Im(z)]^2}\chi(\varepsilon')$$



Starrett, HEDP, **16** (18-22), 2015

Density of States Along Contour
Iron 120 eV, 0.5 g/cm^3



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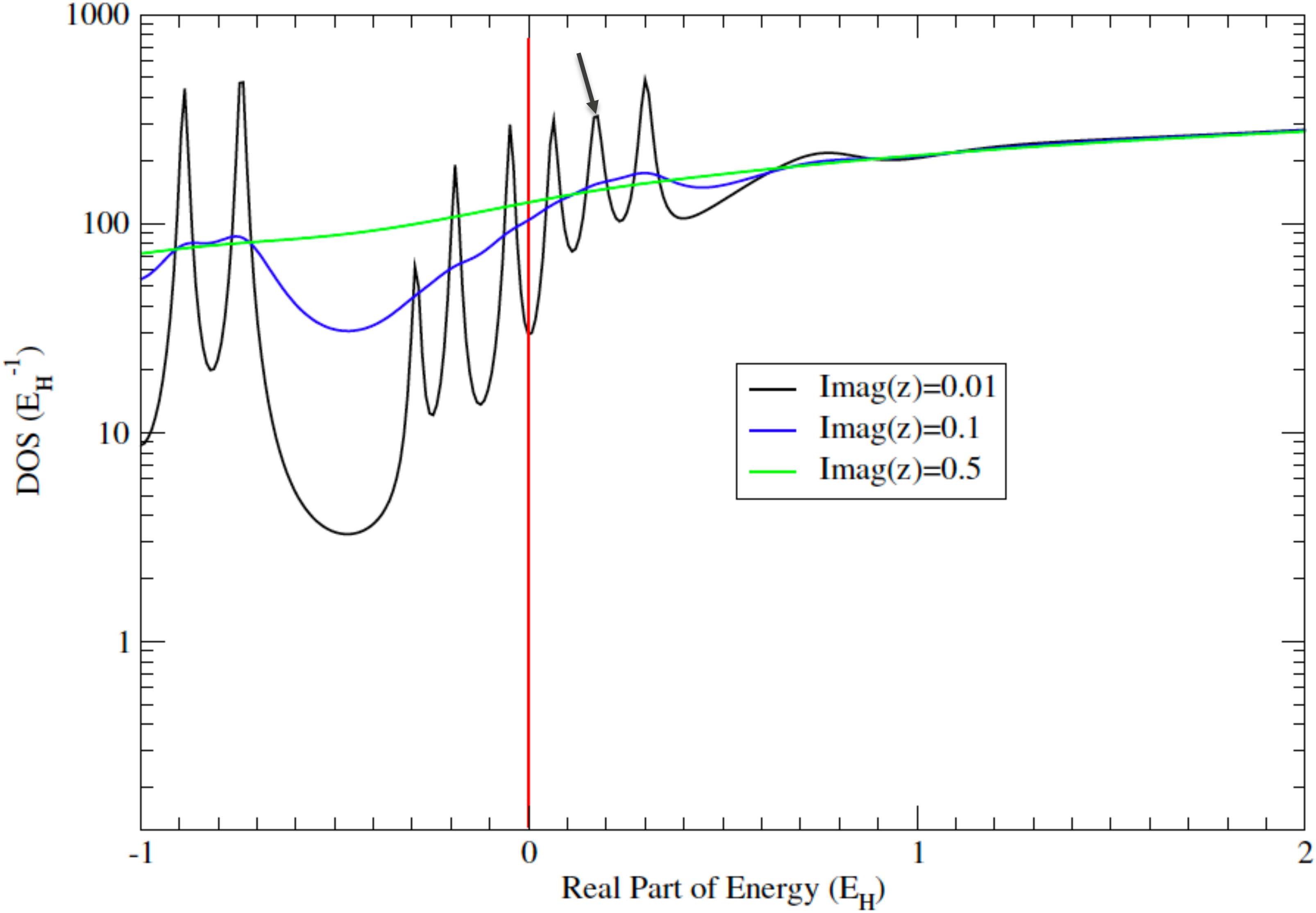

Continuum Supershell

For Example, Iron Z=26

$$\Xi = (1s)^2\ (2s\ 2p)^8\ (3s\ 3p\ 3d)^2\ (4s \ldots 6f)^2$$

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Highest Orbital Predicted by average atom calculation

Continuum Supershell

For Example, Iron Z=26

$$\Xi = (1s)^2\,(2s\,2p)^8\,(3s\,3p\,3d)^2\,(4s\,\ldots 6f\,)^2$$

Highest Orbital Predicted by average atom calculation

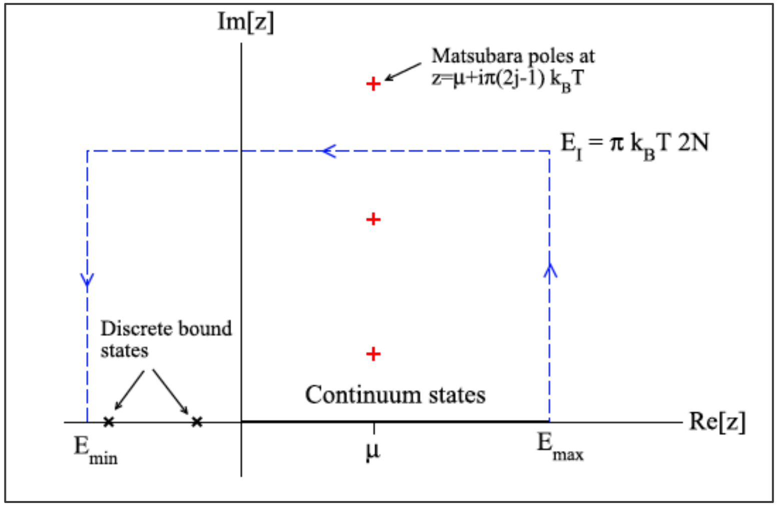
$$\Xi = (1s)^2\,(2s\,2p)^8\,(3s\,3p\,3d)^2\,(4s\,\ldots 6f\,)^2(\varepsilon = 0, \ldots, \infty)^{12}$$

Continuum Supershell

Continuum Supershell, Iron Example

Different SCs have different bound states and therefore (possibly) different pressure ionization thresholds

$$\Xi = (1s)^2\,(2s\;2p)^8\,(3s\;3p\;3d)^2\,(4s\ldots 6f)^2(6g\;6h\,,\varepsilon = 0,\ldots,\infty)^{12}$$

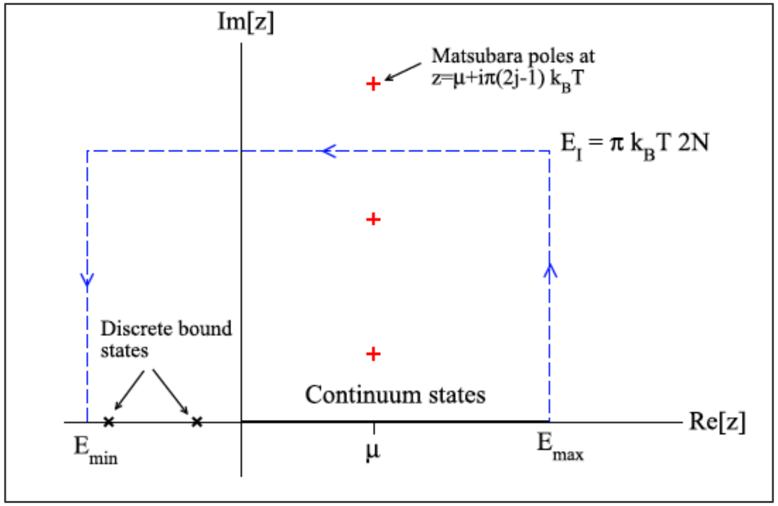


$$E_{min} = E_{6f} + \delta$$

“Unexpected” bound states now part of continuum, occupied according to Fermi-Dirac with continuum chemical potential

Continuum Supershell, Iron Example

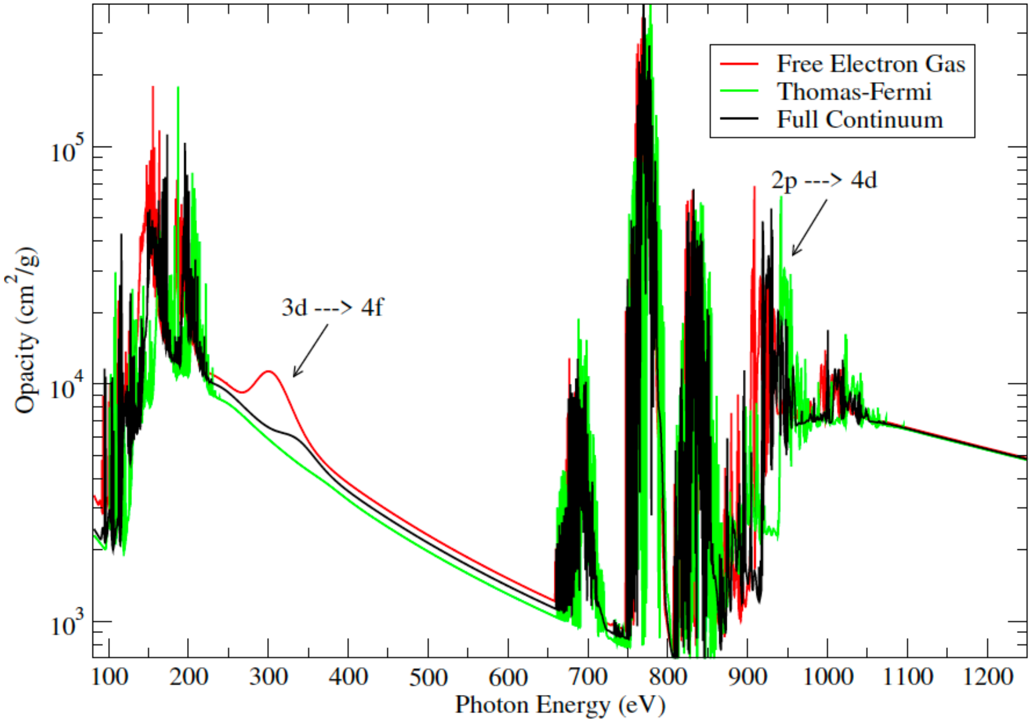
$$\Xi = (1s)^2\,(2s\;2p)^8\,(3s\;3p\;3d)^2\,(4s\ldots 6f)^2(6g\;6h\,,\varepsilon = 0,\ldots,\infty)^{12}$$



- Stabilizes SCF procedure at high plasma densities
- If predicted orbitals become pressure ionized, still accounts for them as part of continuum (resonances)
  - -Breaks model consistency, but less than other methods

Opacity Near Pressure Ionization

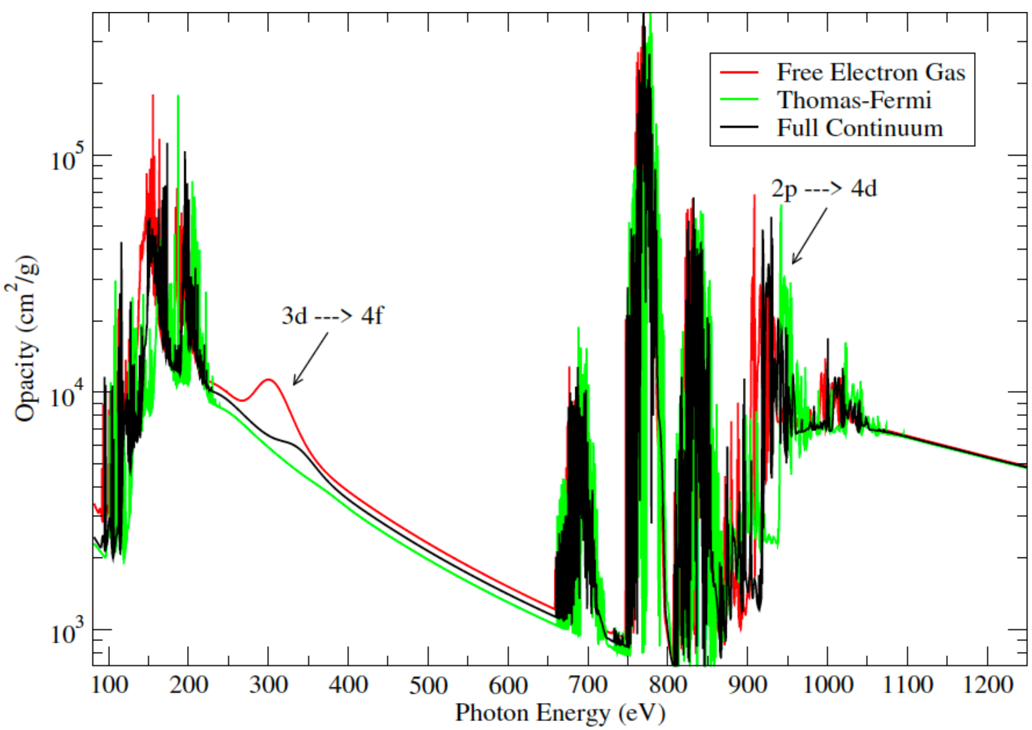
Iron 120 eV, 3.6 g/cm^3



- Most 4f electrons near pressure ionization
  - Exist as bound states for some SCs
- Free electron ---> Weaker plasma screening
- Thomas-Fermi ---> Stronger plasma screening
- Differences disappear for high temperatures, extreme densities, near-neutral

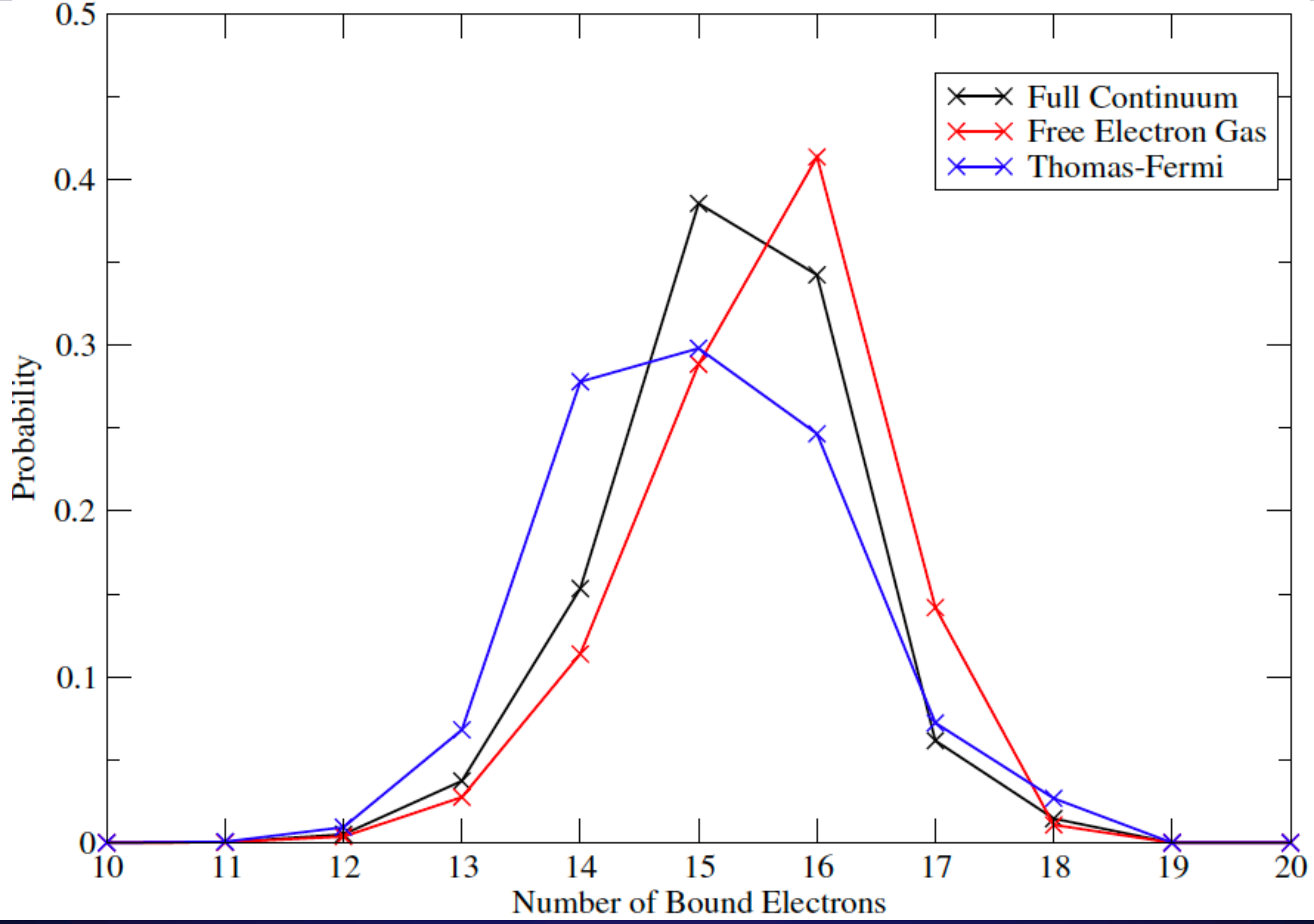
Opacity Near Pressure Ionization

Iron 120 eV, 3.6 g/cm³

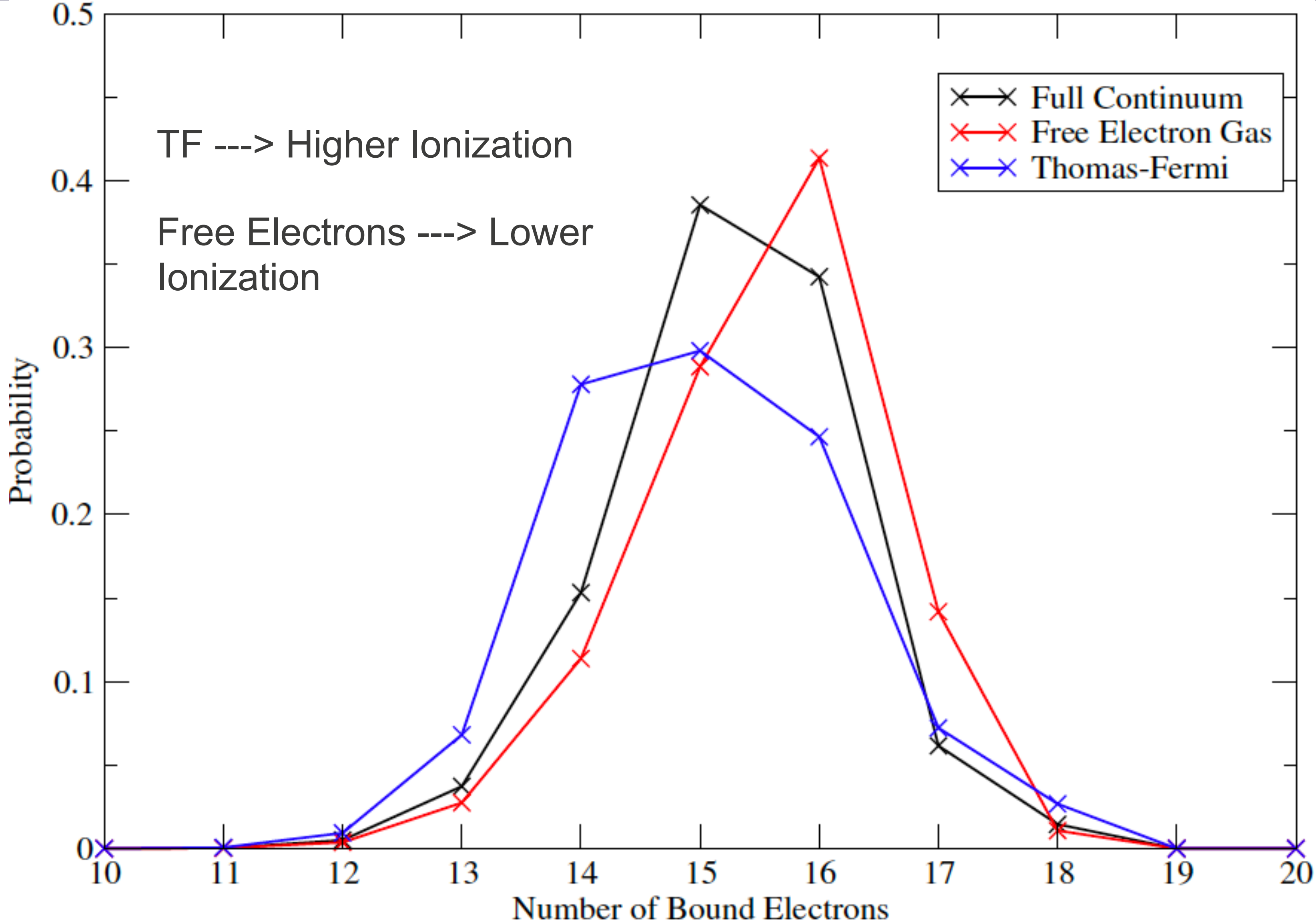


- Electronic structure has consistent continuum treatment
- EOS from SB using only bound electrons
  - Need Free Energy of continuum to improve EOS
- Free electron/TF shift peak of charge state distribution (CSD) around proper continuum treatment

CSD, Iron 120 eV, 3.6 g/cm^3

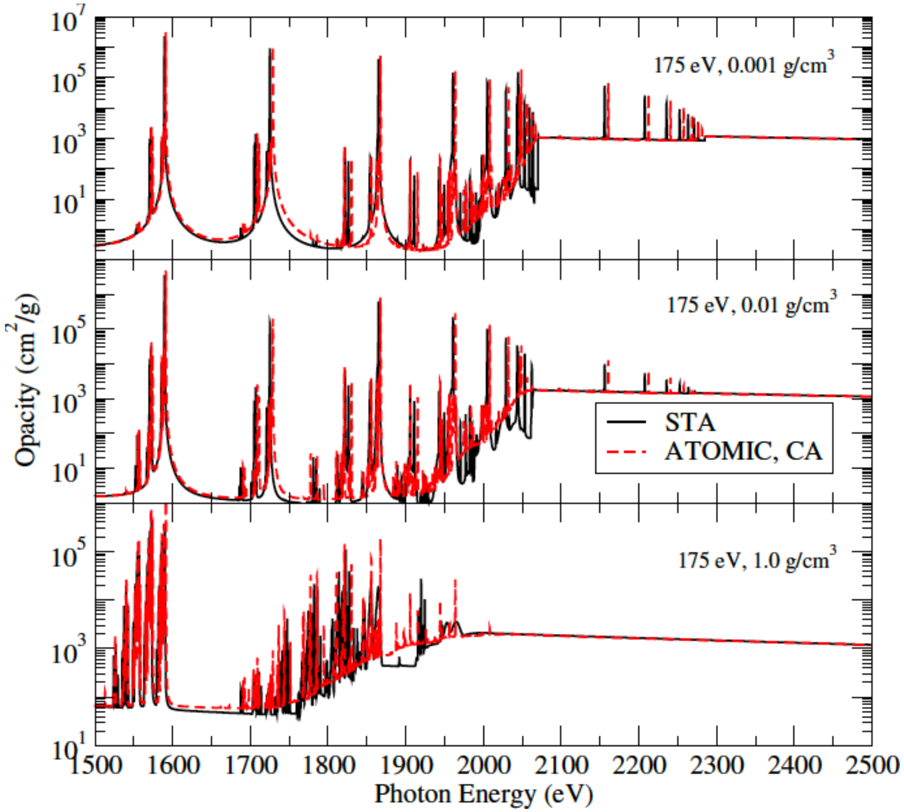


CSD, Iron 120 eV, 3.6 g/cm^3



Correction to Transition Energies

$$E_C^{(1)} = \sum_{\phi_C} \langle \phi_c | H_0 + H_1 | \phi_C \rangle / g_c$$
$$= \sum_{s \in C} q_s \langle s \rangle + \sum_{r,s \in C} q_s (q_r - \delta_{r,s}) \langle r,s \rangle \ .$$



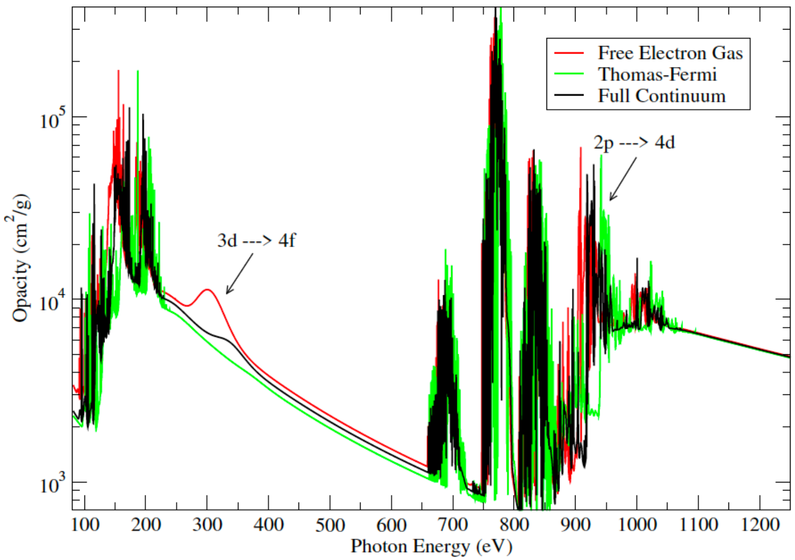
- Need two-electron corrections to get accurate transition energies
- Corrections inaccurate when pressure ionization is significant, especially for bound-free correlations

Bar-Shalom et al, Phys. Rev. A, **40** (6), 1989

Bound-Free Interaction

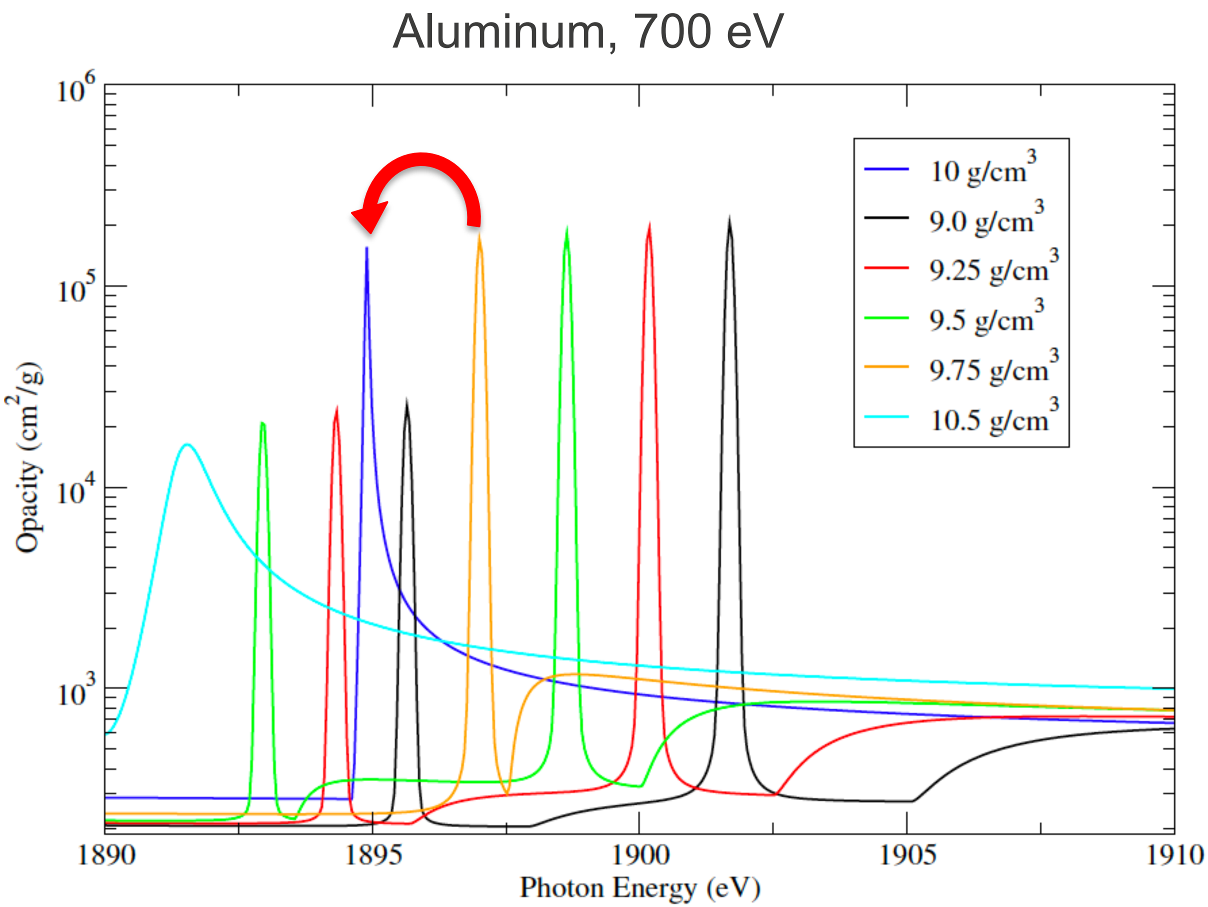
$$< r, s > \neq 0$$

Iron 120 eV, 3.6 g/cm^3



- Bound-Free resonances not apparent because of edge energy corrections failing
- TF 4f-resonance fully "dissolved" in continuum states
- When electronic structure is dominated by electrons occupying continuum resonances, too much information lost by throwing away bound-continuum interaction

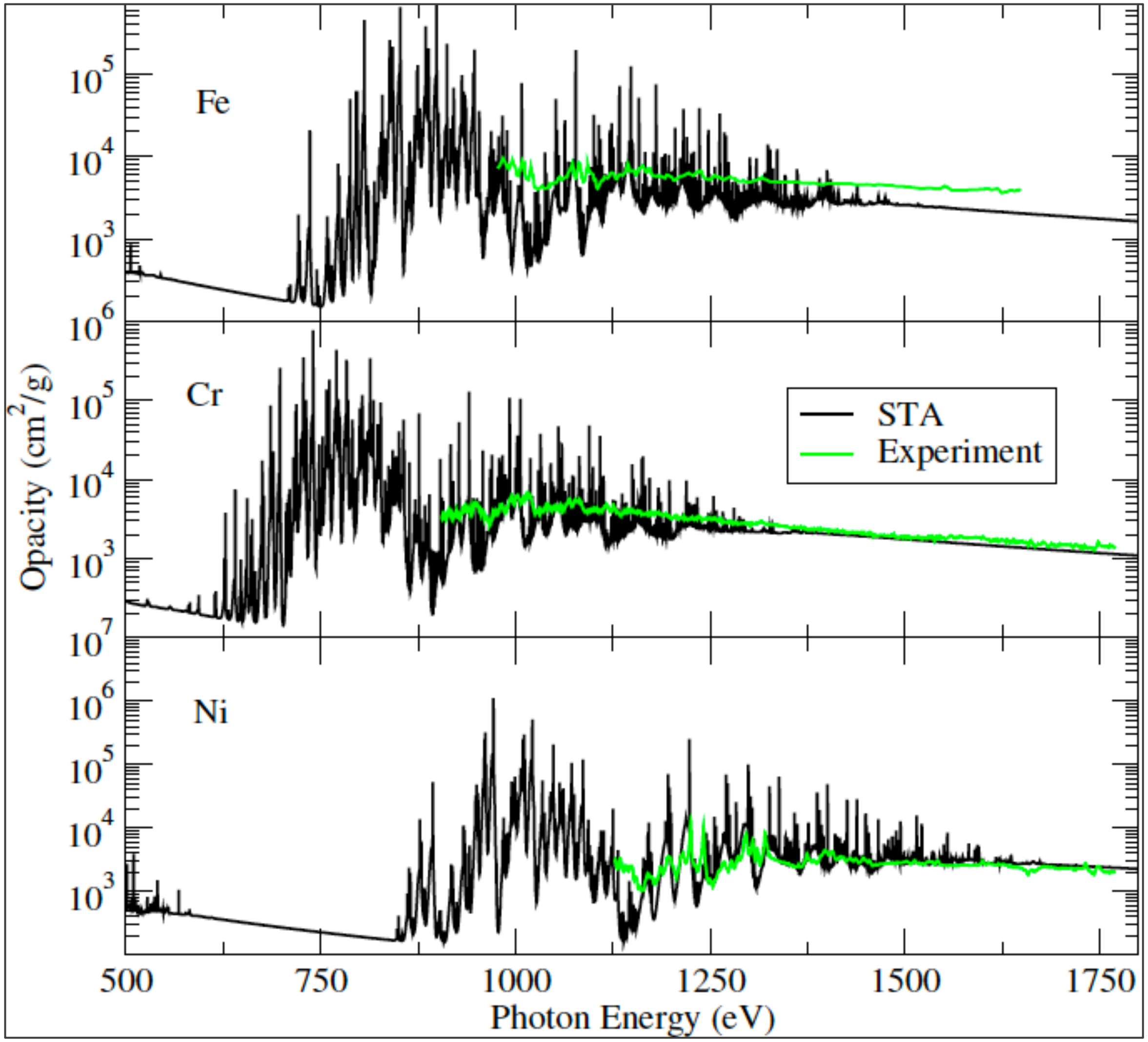
Pressure Ionization of 3p States in Aluminum



D. J. Hoarty et al., Phys. Rev. Lett. 110, 265003 (2013)

- Independent particle transition energies
  - Still need to improve description of bound-continuum interaction under photo-absorption
- Bound-bound features smoothly turn into bound-free features after pressure ionization event

Sandia Z Experiments



- Nonrel, natural and Doppler broadening
- Different continuum treatments not important for structure or EOS
- No change in discrepancies when comparing to experiments

Fe (180 eV, 0.16 g/cm^3)

Cr (180 eV, 0.16 g/cm^3)

Ni (180 eV, 0.17 g/cm^3)

Conclusions

- Developed superconfiguration and STA opacity codes
  - Focus on robustness over wide range of plasma conditions
  - Includes consistent treatment of continuum electrons using Green's functions and complex contour integration
- Comparisons to configuration-average calculations excellent in the isolated atom limit and for highly charged systems
- Comparison between various treatments of electron continuum highlights challenges of modeling dense plasmas
  - EOS may need consistent level of approximation to have confidence in CSDs
  - Transition energies need reasonable inclusion of bound-continuum Coulomb interaction