Some Progress in Modeling Warm and Hot Dense Matter

Charles Starrett

Collaborators, Postdocs and Students involved in this work:
Trinity Thelen, Nathaniel Shaffer, Simon Blouin, Didier Saumon, Crystal Ottoway, Michael Laraia, Matt Gill, Jerome Daligault, Scott Baalrud, Danny Rehn, Chris Fontes

The Problem

A number of independent experiments have revealed problems with our approach to opacity at high density

PHYSICAL REVIEW LETTERS **122,** 235001 (2019)

Editors' Suggestion | Featured in Physics

Systematic Study of *L*-Shell Opacity at Stellar Interior Temperatures

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ARTICLE

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Measurements of continuum lowering in solid-density plasmas created from elements and compounds

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LETTER

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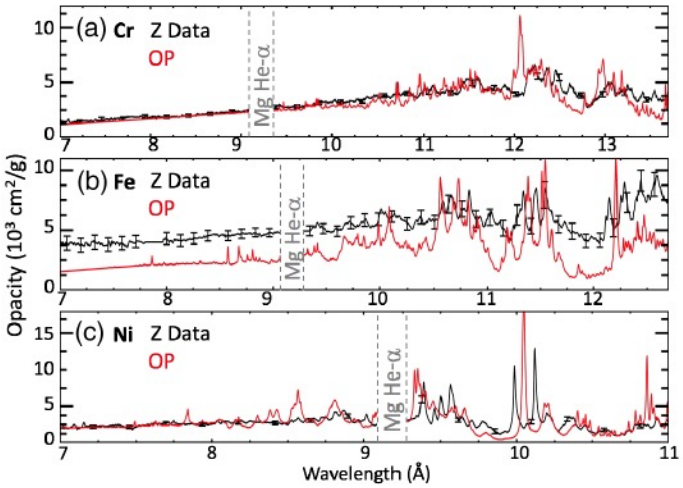
A higher-than-predicted measurement of iron opacity at solar interior temperatures

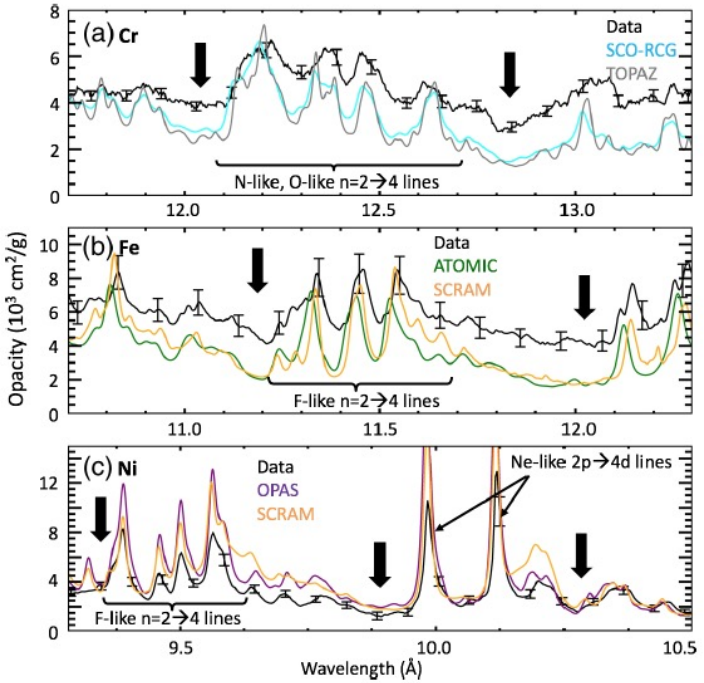
J. E. Bailey[1], T. Nagayama[1], G. P. Loisel[1], G. A. Rochau[1], C. Blancard[2], J. Colgan[3], Ph. Cosse[2], G. Faussurier[2], C. J. Fontes[3], F. Gilleron[2], I. Golovkin[4], S. B. Hansen[1], C. A. Iglesias[5], D. P. Kilcrease[3], J. J. MacFarlane[4], R. C. Mancini[6], S. N. Nahar[7], C. Orban[7], J.-C. Pain[2], A. K. Pradhan[7], M. Sherrill[3] & B. G. Wilson[5]

In particular, the iron opacity experiment, if true, is a major problem and efforts are underway to replicate it at the NIF

Our Best Models Fail…

From Nagayama et al (2019):
(180 eV, 3x10^{22} cm^{-3})





All these models start from an atomic perspective and plasma physics is treated crudely.

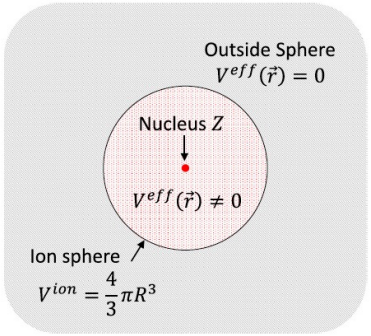
How About Using Our Best Dense Plasma Model? Density Functional Theory

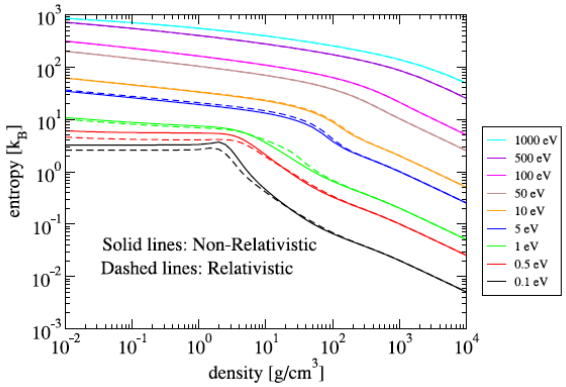
Need KS-DFT model that can access high temperatures and can calculate opacity at high T – a challenging problem

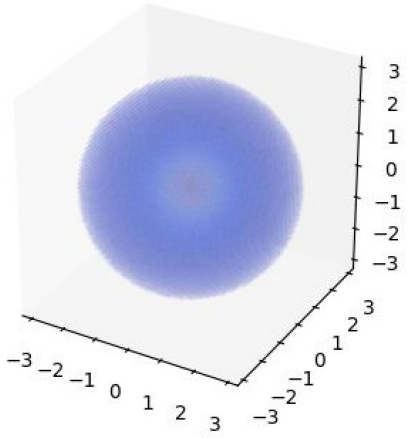
We start with the Average Atom model:

Use Mermin-Kohn-Sham density functional theory to solve for the properties of one atom in a plasma.

Usually used for EOS calculations





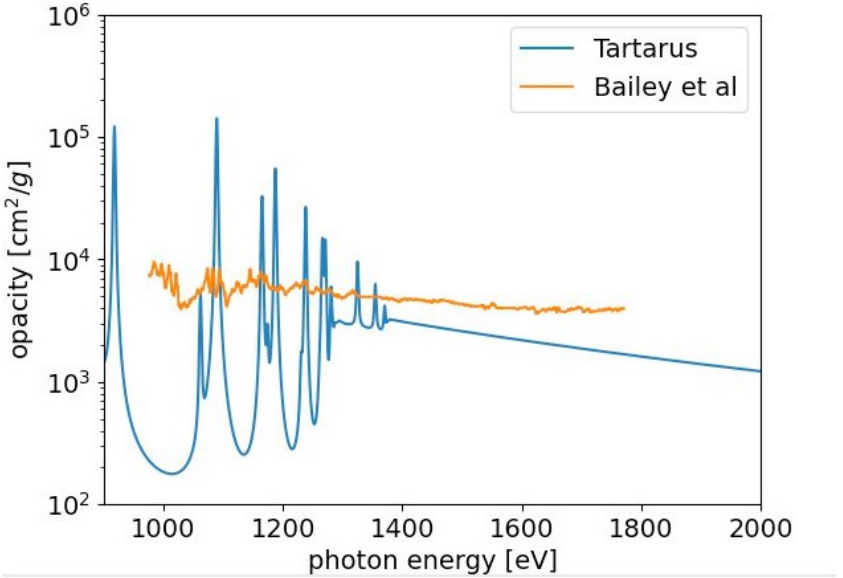


Average Atom Opacity

Computer Physics Communications
Volume 235, February 2019, Pages 50-62

Wide ranging equation of state with Tartarus: A hybrid Green's function/orbital based average atom code

C.E. Starrett [a], N.M. Gill [a b], T. Sjostrom [a], C.W. Greeff [a]



Why so poor?
-- DFT Averages over excited states → one "line" instead of many
-- Focus on the bound-free tail, that should be a reasonable prediction

Also, possibly
1) Independent particle approximation for opacity
2) Crude plasma physics (ion in sphere)

Improved Plasma Physics:
Mean Force Potential

High Energy Density Physics 31 (2019) 24–30

Contents lists available at ScienceDirect

High Energy Density Physics

journal homepage: www.elsevier.com/locate/hedp

Mean-force scattering potential for calculating optical properties of dense plasmas

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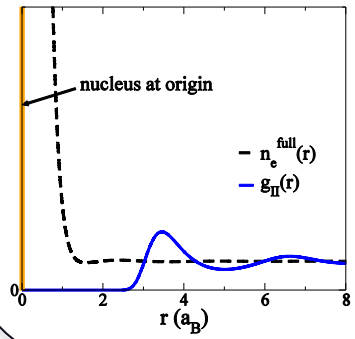
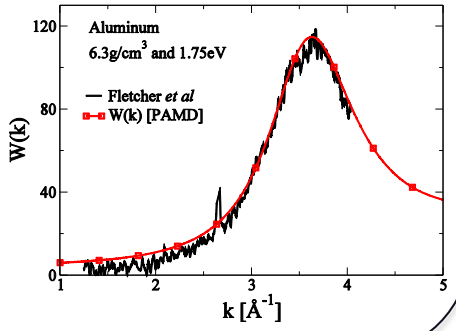
$$V^{MF}(r) = V_{ie}(r) + n_i^0 \int d\vec{r'} \frac{C_{ie}(|\vec{r} - \vec{r'}|)}{-\beta} h_{ii}(r')$$

$$+ n_e^0 \int d\vec{r'} \frac{C_{ee}(|\vec{r} - \vec{r'}|)}{-\beta} h_{ie}(r')$$

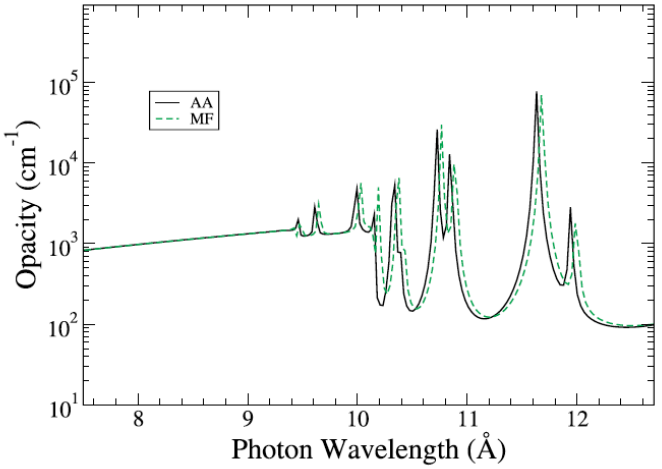




Following Krief et al's suggestion (ApJ 2018), we tested the effect of an improved treatment of ion correlations.

This did not solve the bound-free problem.

(But this potential is great for calculating electrical conductivities! Starrett, HEDP 2017)



TD-DFT

PHYSICAL REVIEW E **103**, 043206 (2021)

Time-dependent density functional theory applied to average atom opacity

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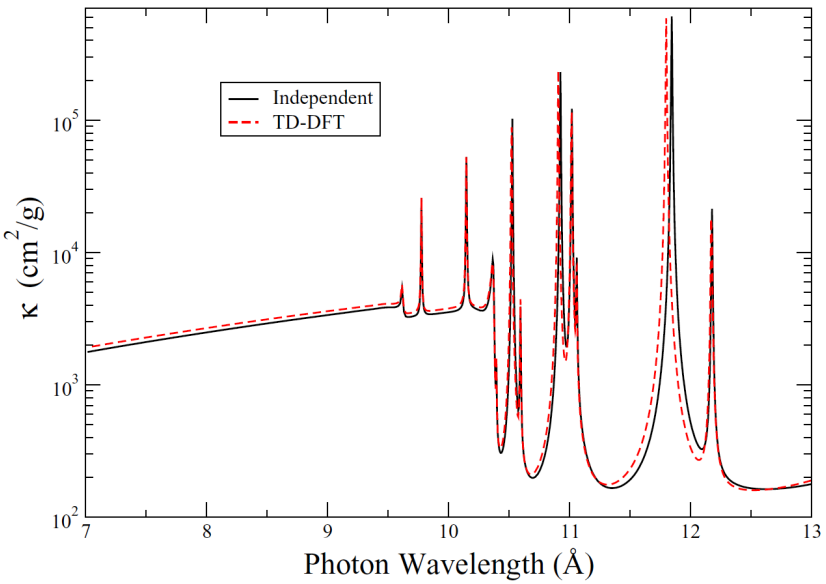
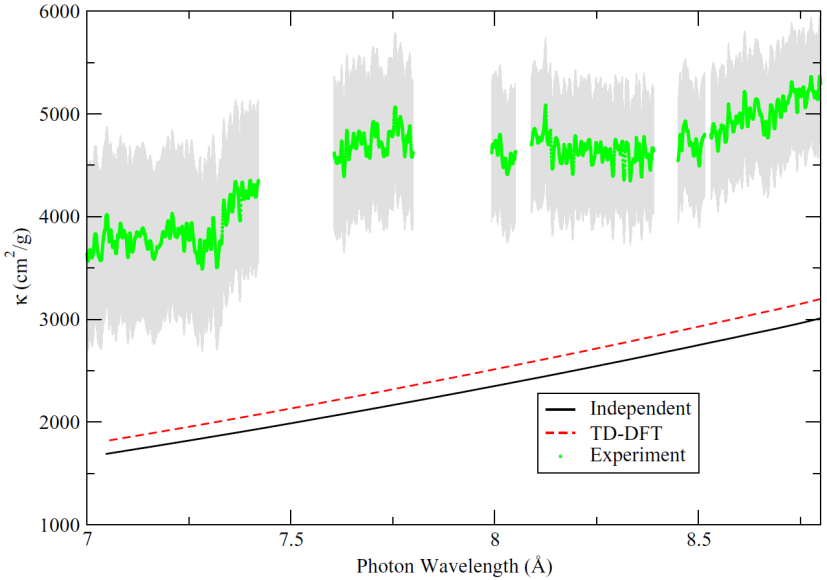
TD-DFT includes:

1) Mixing between excitation pathways
2) Response of atom to excitation (orbital relaxation, sort of)

$$\chi(\vec{r},\vec{r'},\omega) = \chi_0(\vec{r},\vec{r'},\omega) + \int d\vec{r}_1\,\chi_0(\vec{r},\vec{r}_1,\omega)\int d\vec{r}_2 \times \left[\frac{1}{|\vec{r'}-\vec{r}_2|} + \left.\frac{\delta V_{xc}(\vec{r'},\omega)}{\delta n(\vec{r}_2,\omega)}\right|_{n_0}\right]\chi(\vec{r}_2,\vec{r}_1,\omega)$$

TD-DFT





Result showed a smallish change in the bound-free (7%), not enough to see agreement with experiment.

Similar effect observed for Ni and Cr.

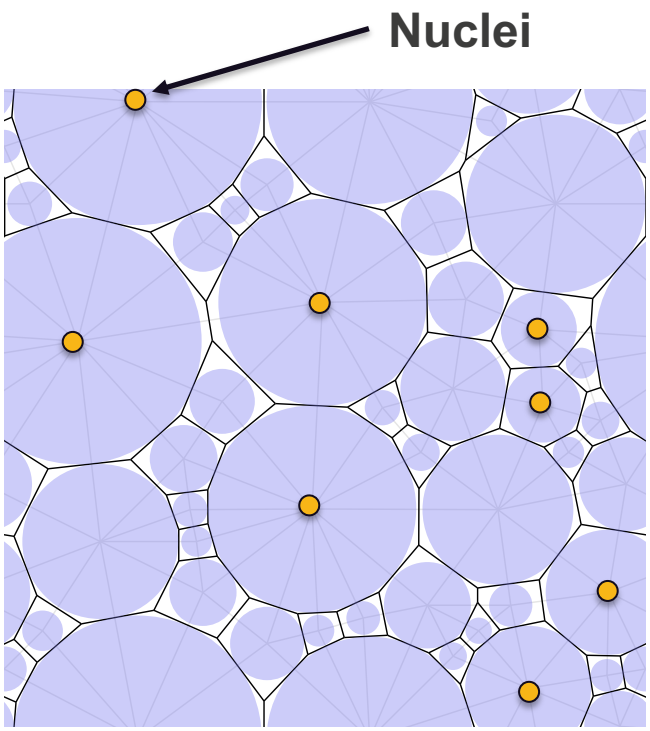
Maybe the problem is with the Average Atom model → Multiple Scattering Theory

Starrett & Shaffer, PRE 2020

Starting point: Divide space into cells

Next: solve local, single site (cell) problems using DFT and free-electron boundary condition

Final step: Use Dyson's equation for Green's function, to match the cell-solutions, correcting the boundary condition



$$G\left(\vec{r}+\overrightarrow{R^n},\overrightarrow{r'}+\overrightarrow{R^{n'}},z\right)=$$

$$\delta_{nn'}\sum_L H_L^{n\times}(\vec{r}_>,z)R_L^n(\vec{r}_<,z)$$

Single site term

$$+\sum_{LL'} R_L^n(\vec{r},z)\mathcal{G}_{LL'}^{nn'}(z)R_{L'}^{n'\times}(\overrightarrow{r'},z)$$

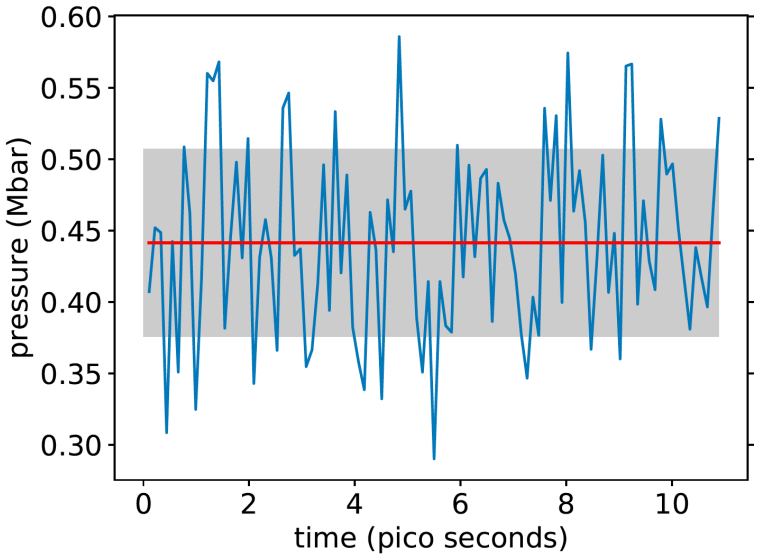
+ Multi-Center correction

Originally : Korringa ('47), Kohn-Rostoker ('54) for wfns, later Ham and Segall ('62) for GF

MST – captures ionic disorder, uses all-electron DFT, practical at high temperatures

MST uses molecular dynamics to get ion positions. EOS is from ensemble average.

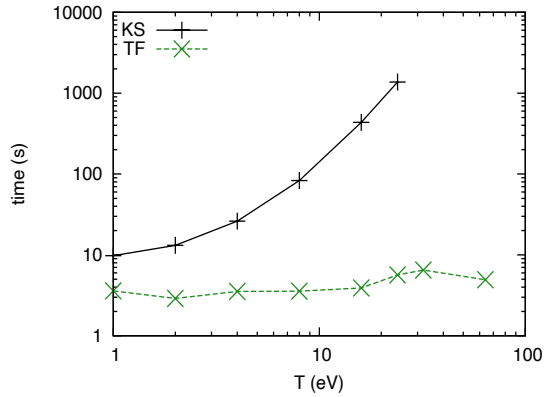
MD is currently not self-consistent – it comes from another model (PAMD, Starrett et al PRE 2015)



T [eV]	ρ [g/cm^3]	Time (mins)
10	2.7	15.7
10	0.027	11.4
100	2.7	14.4
100	0.027	9.5

Plane Wave

Wall-time $\propto T^3$



Wall-time does not significantly depend on temperature, density

Multiple Scattering Theory
Opacity via independent particle approximation

DFT averages over excited states – leads to very poor spectra

Does give a self-consistent treatment of the Stark effect.

(And is a good model for EOS, Hugoniots, Ottoway et al PRE 2021)

PHYSICAL REVIEW E **105**, 015203 (2022)

Dense plasma opacity via the multiple-scattering method

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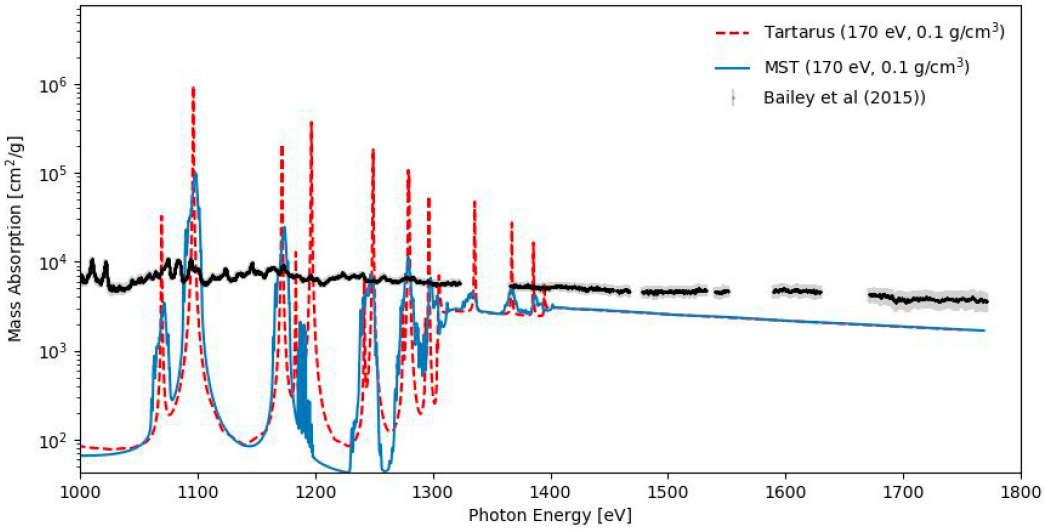

Maybe Mermin-KS-DFT isn't good enough
Can we introduce explicit excited states?

A notable attempt to do this is the super-configuration approach of Bar-Shalom (PRA 1989)

IOP Publishing Journal of Physics B: Atomic, Molecular and Optical Physics
J. Phys. B: At. Mol. Opt. Phys. **56** (2023) 015001 (12pp) https://doi.org/10.1088/1361-6455/acacd9

A superconfiguration calculation of opacity with consistent bound and continuum electron treatments using green's functions

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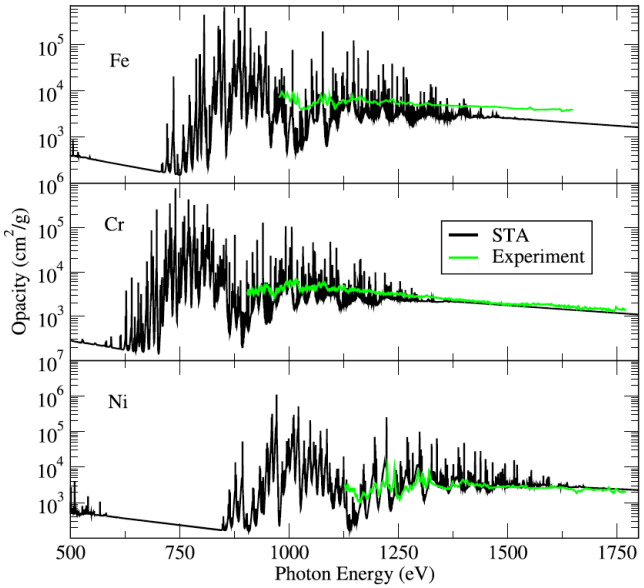

Much improved spectra over DFT, but Fe problem persists.

Super-Configurations, as implemented, is not variationally derived (DFT is), and is not used for EOS usually (DFT is). Could this weakness point to a way forward?

Variational, configurationally resolved model

ELSEVIER

Available online at www.sciencedirect.com

ScienceDirect

High Energy Density Physics 3 (2007) 34–47

High Energy Density Physics

www.elsevier.com/locate/hedp

Variational approach to the average-atom-in-jellium and superconfigurations-in-jellium models with all electrons treated quantum-mechanically

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Blenski et al have given a variational derivation of a super-configuration model.

However, it appears that this is not a practical, implementable model (no code exists).

Variational, configurationally resolved model

We are attempting to derive an alternative, simpler model.

What follows is some preliminary results from this effort.

Variational Model of configurations in a plasma

Model of the plasma: bound and free electrons in charge neutral spheres, connected though a common reference energy

The constrained Free energy is

$$\Omega = \sum_x W_x \left[\Delta F_x + f_0[n^0_{e,x}]V + T \ln W_x\right] - B\left[\sum_x W_x - 1\right]$$
$$+ \gamma \left[\sum_x W_x \left\{\int d^3r (n_{e,x}(\boldsymbol{r}) - n^0_e) + n^0_{e,x} V - Z\right\}\right]$$

Population

Free energy per ion

Charge Neutrality

Variational Model of Populations Minimization

$$\frac{d\Omega}{dn_{e,x}(\vec{r})} = 0$$ Leads to Effective potentials

$$\frac{d\Omega}{dW_x} = 0$$ Leads to configuration probabilities

$$P = -\frac{d\Omega}{dV}$$ Leads to EOS

Variational Model of Populations
Minimization gives EOS

Population: $$W_x = \exp\left(-\frac{1}{T}F_x\right)/\mathcal{Z}$$

Boltzmann factor comes out of model

$$P = -\frac{\partial\Omega}{\partial V} = -\sum_x W_x \left\{f_0[n^0_{e,x}] - \gamma n^0_{e,x}\right\}$$

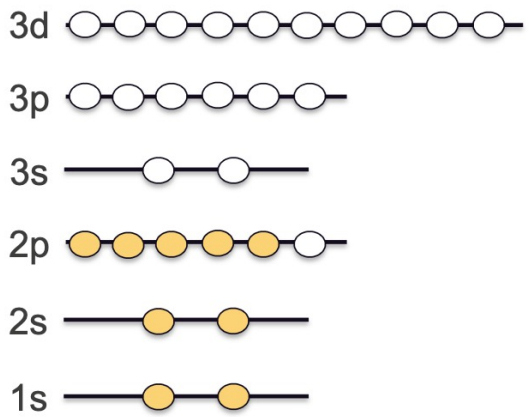
Self Consistent EOS

$$\gamma = \sum_x W_x \left[\mu^{xc}_x + \mu_x\right]$$

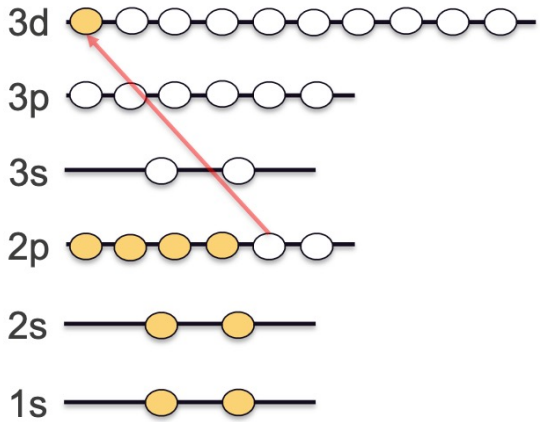
Connected by Muffin-Tin level

But how do we define a configuration?

Configuration: $1s^2\ 2s^2\ 2p^5$



Configuration: $1s^2\ 2s^2\ 2p^4\ 3d^1$



We can define a configuration by listing its bound state occupations

But what if a bound state does not exist (i.e., it has been pressure ionized)?

Then at one density the $1s^2 2s^2 2p^4 3d^1$ (for example) may exist and have significant probability, but on incrementally increasing the density, the configuration ceases to exist → EOS problems

Mixing configurationally resolved bound electrons with configurationally averaged free electrons

Electron density in ion

$$n(\vec{r}) = \sum_{all\ eigen\ states\ i} f_i |\phi_i(\vec{r})|^2$$

f_i is the occupation of eigenstate i

f_i =1 for a $1s^2$
(2 comes from spin degeneracy)

For self-consistent plasma effects, we need f_i for the free electrons too

$$n(\vec{r}) = \sum_{bound\ i} f_i |\phi(\vec{r})|^2 + \int d\epsilon\, f_\epsilon |\phi_i(\vec{r})|^2$$

It is sometimes argued that we can use the ensemble-average f_ϵ (the Fermi-Dirac distribution). However, this breaks consistency between bound and free electrons → EOS problems.

Let's start with a simple ansatz

We want to be able to choose a configuration, eg., $1s^2 2s^2 2p^4 3d^1$ irrespective of whether the eigenstates are bound or not.

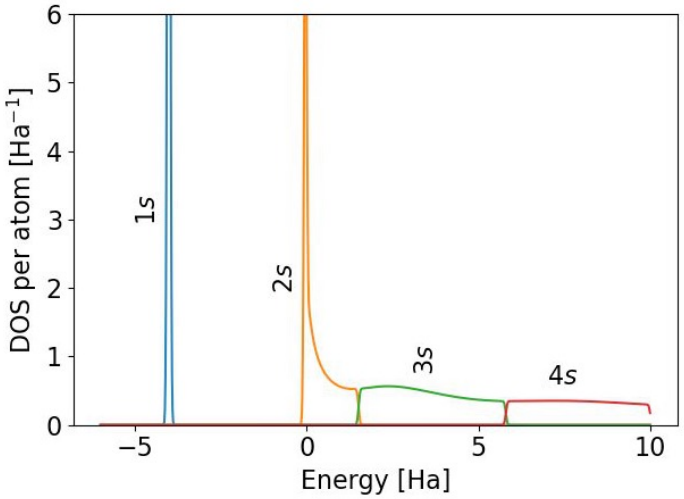
So, we extend the definition of the eigenstate into the continuum

$$2(2l+1) = \int_{\epsilon_{n,l}}^{\epsilon_{n+1,l}} d\epsilon \, \chi_l(\epsilon)$$

DOS for given l

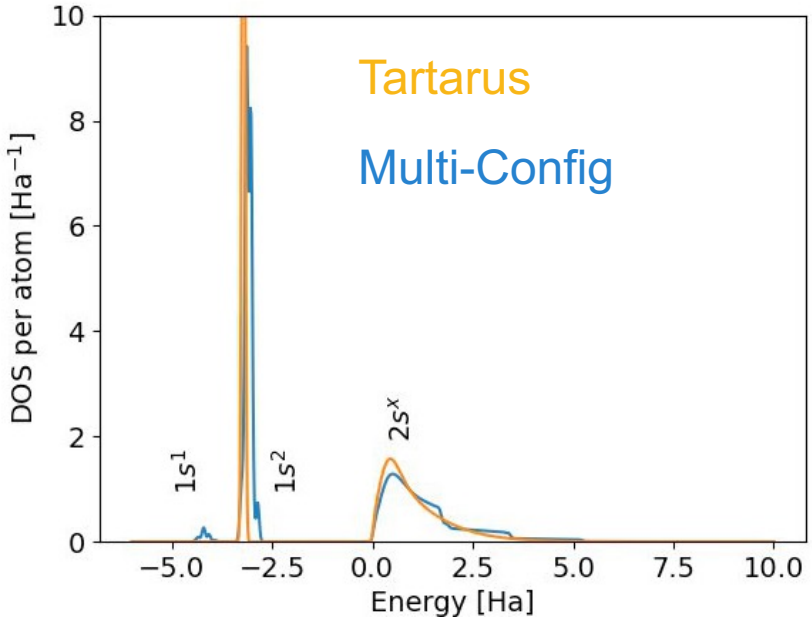
Solve for the energy boundaries.

This recovers the usual definition for bound states and gives an energy band for an eigenstate in the continuum.



Some Preliminary results
Density of States

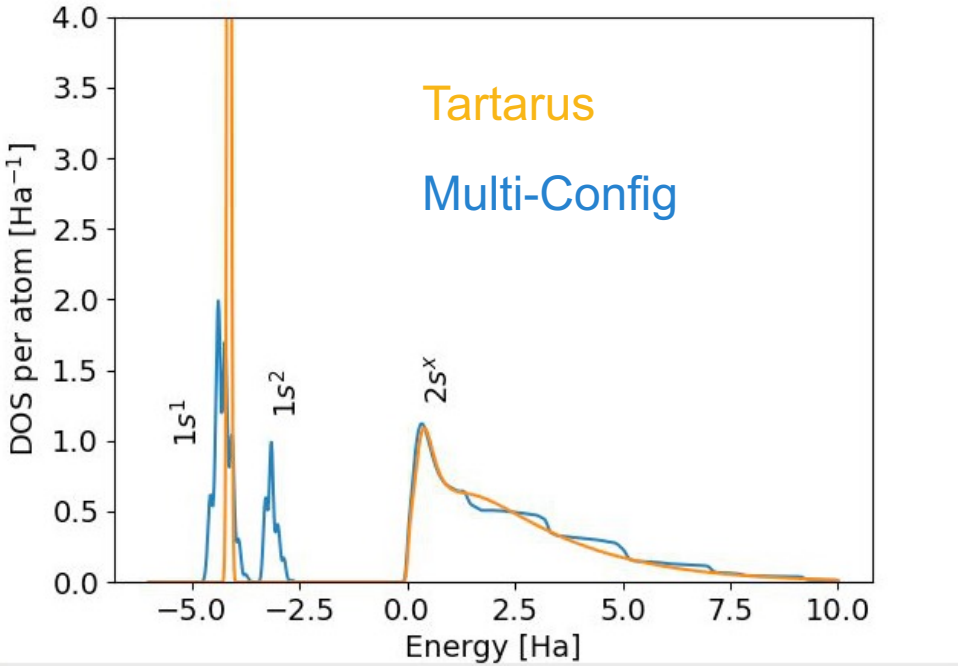
Beryllium at 20 eV and 1.85 g/cc



All possible (~10000) configs, up to n=6, considered

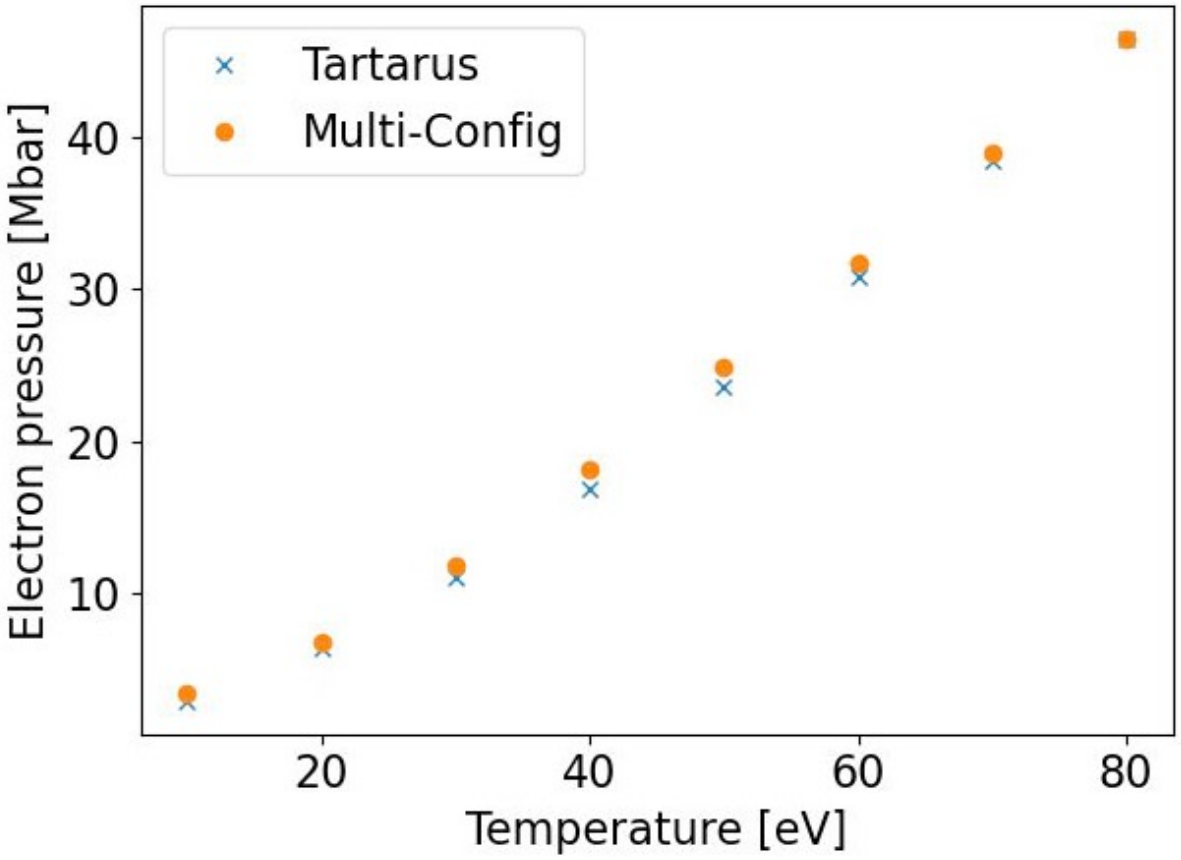
Some Preliminary results
Density of States

Beryllium at 50 eV and 1.85 g/cc



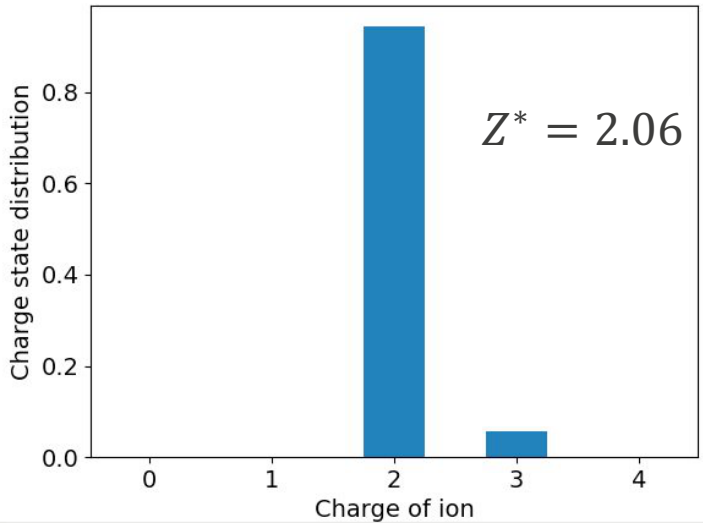
Some Preliminary results
Equation of State

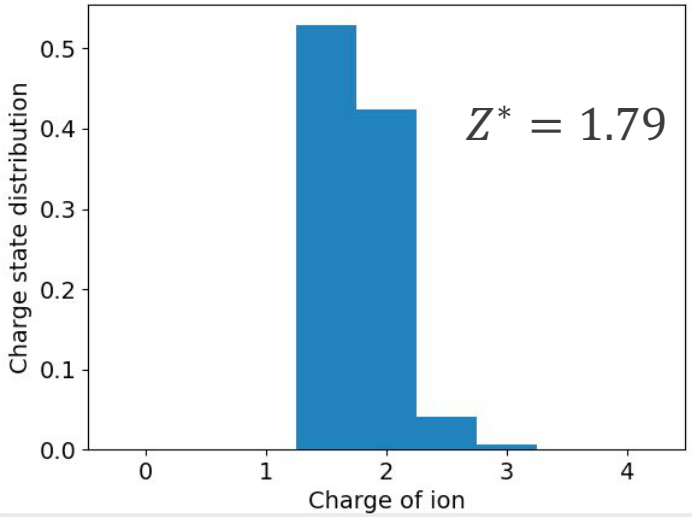
Beryllium at 1.85 g/cc



Some Preliminary results
Charge State Distribution

Beryllium at 20 eV and 1.85 g/cc





$\bar{Z}$ = nuclear charge
- # of bound electrons

Z^*= # of free electrons

Difference is electrons in quasi bound states (including resonances)

Average atom : $\bar{Z} = 2.06$

Average atom : $Z^* = 1.69$

Conclusions and Outlook

Systematic improvements to our physics models have so far not resolved the iron opacity problem (or the solar opacity problem).

Our model improvements have increased the fidelity of our conductivity and EOS models

We are working on a configurationally resolved variational model.

- We expect that this will lead to new insights into the EOS of dense plasmas as well as the opacity