SAND2020-8763PE

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Sandia theory perspective: detail, completeness, and consistency

S. Hansen, T. Nagayama, T. Gomez, R. More*,
T. Hentschel**, C. Jennings, A. Baczewski, and a great community ***

Sandia National Laboratories (DOE/OFES early career & LDRD)

**private company*

***Cornell University*

**** LLNL, LANL, CEA…*

U.S. DEPARTMENT OF ENERGY

National Nuclear Security Administration

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High-precision measurements drive innovation and improvements in atomic models

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- **HEDP uses models for experimental design and diagnostics**
  - We can adjust model detail to match applications:
    - Low-resolution models are ok for rad-hydro design
    - High-resolution models are needed for spectroscopic diagnostics
  - For all applications, we care about accuracy, which requires completeness and consistency, and about tractability, which often requires sacrifices in detail

- **High-precision measurements from carefully prepared experiments set the standard:**
  - XFEL fluorescence (e.g. Vinko et al)
    - Continuum lowering & dense plasma effects
  - Opacity (Z, NIF, Omega) (e.g. Bailey et al)
    - One- and two-photon cross sections, line shapes
  - ICF: fluorescence and absorption (e.g. Hansen et al, Jiang et al)
    - Line shifts and dense plasma effects

- **Where would we like to see models go?**
  - Internally consistent, spectroscopically detailed, rigorously complete… and tractable

A brief tour of the atomic model zoo

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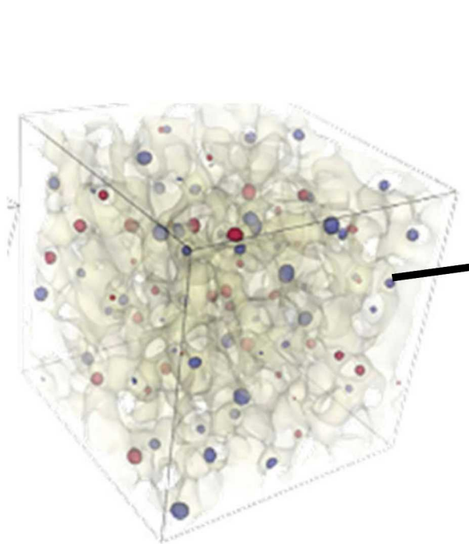
Density functional theory molecular dynamics (DFT-MD)
Accurate for EOS & transport
Quite expensive & undetailed
Natively consistent & complete
Limited to LTE

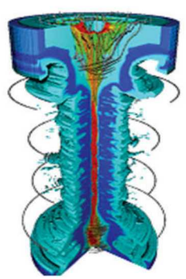
Density functional theory Average atom (DFT-AA)
Accurate-ish for EOS & transport
Quite cheap & undetailed
Natively consistent & complete
Limited to LTE

Configuration-based collisional-radiative models
Accurate for EOS & spectra
Variable cost & detail
Can be inconsistent, incomplete
Can handle non-LTE

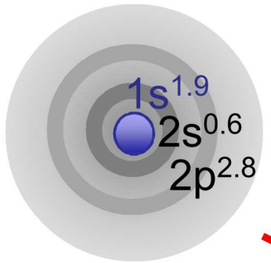


e.g. TD-DFT
A. Baczewski

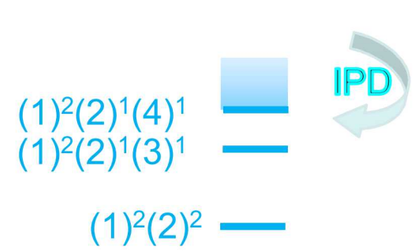
Tables



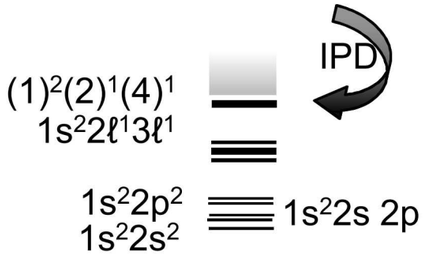
e.g. Gorgon
C. Jennings



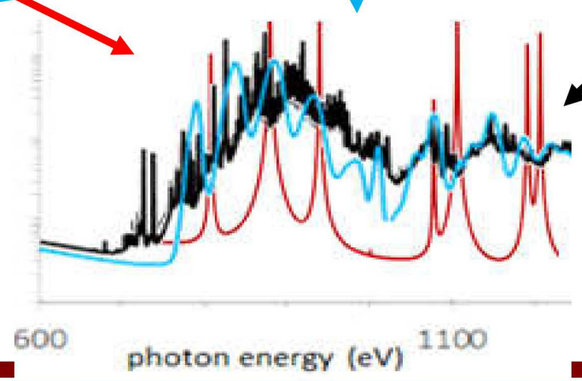
e.g. Purgatorio
B. Wilson, P. Sterne
or NPA (Starrett, Saumon)



e.g. DCA
H.Scott



e.g. SCRAM
S. Hansen

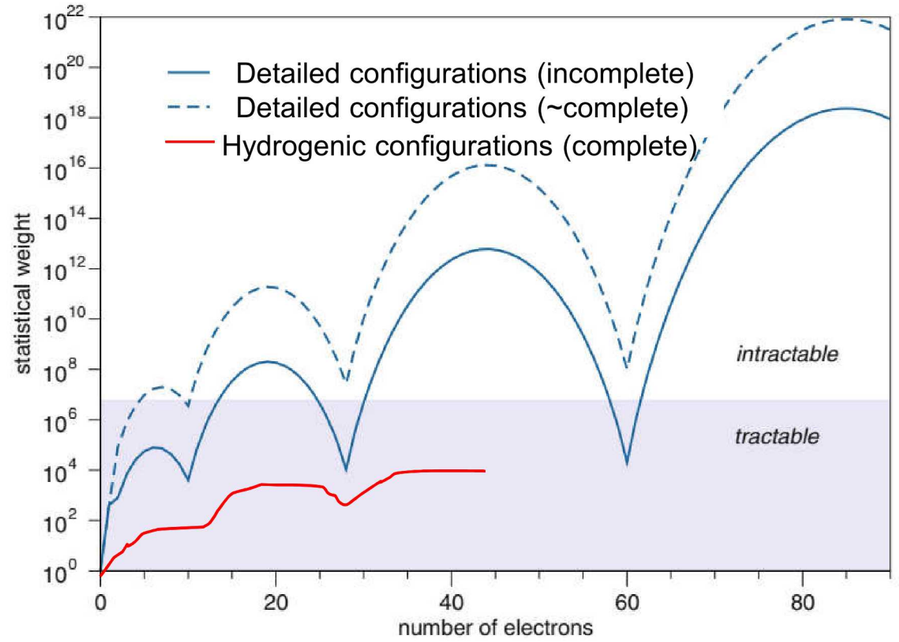


Detail and completeness compete with tractability; consistency is often an afterthought

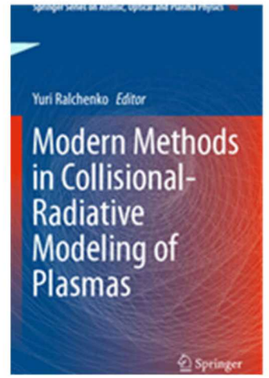
- Combinatorics of <u>completeness</u> are unforgiving:
  - Ion number and electronic states challenge DFT-MD at high Z, T
  - Electronic configurations & rates challenge collisional-radiative models at high Z
  - Sacrificing completeness degrades accuracy

- Sacrifices can be made in <u>detail/resolution</u>:
  - DFT-AA models access all Z, T easily
  - Hydrogenic collisional-radiative models are fast enough to run inline in simulations
  - Rough rate approximations can be ok

- <u>Consistency</u> also matters:
  - Density effects/structure/rates
  - Rates/fields/lineshapes
  - EOS/transport/opacity
  - Non-LTE & non-local effects



2016
Ed. Yu. Ralchenko

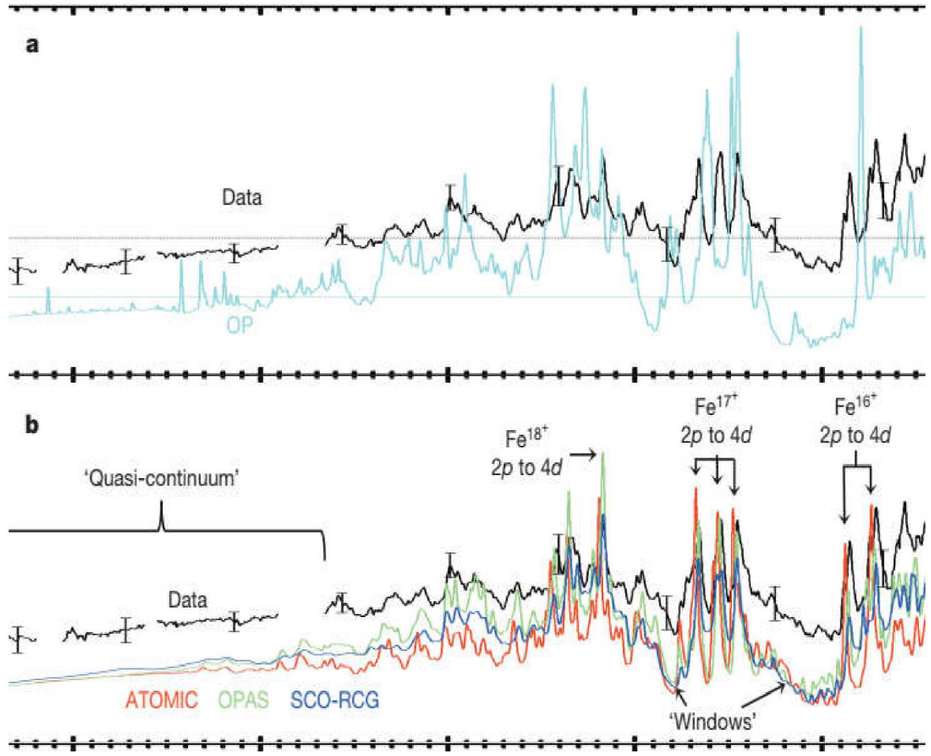


High-precision measurements help us know what's "good enough,"
but tend to require high resolution

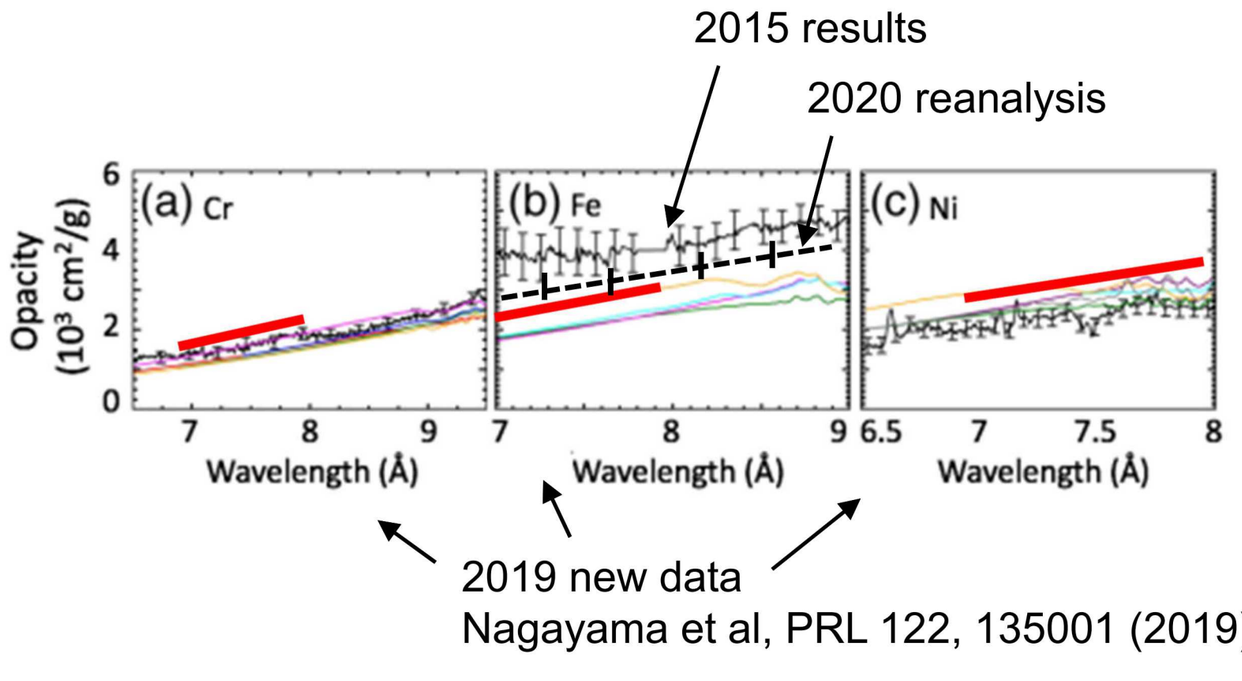
Bailey et al's 2015 iron opacity measurements stimulated extensive investigations

- Good agreement with detailed models at lower T, n_e
  → OP not sufficiently detailed

- Disagreement in widths, windows & continuum at higher T, n_e
  → Better cross sections? (Nahar& Pradhan)
  → More complete states? (Iglesias, Hansen)
  → More complete rates? (More, Pain, Kruse & Iglesias)
  → Better line shapes? (Gomez, Nagayama)
  → Refined analysis? (Nagayama)
  → More experiments? (Bailey, Loisel, Perry, Heeter…)



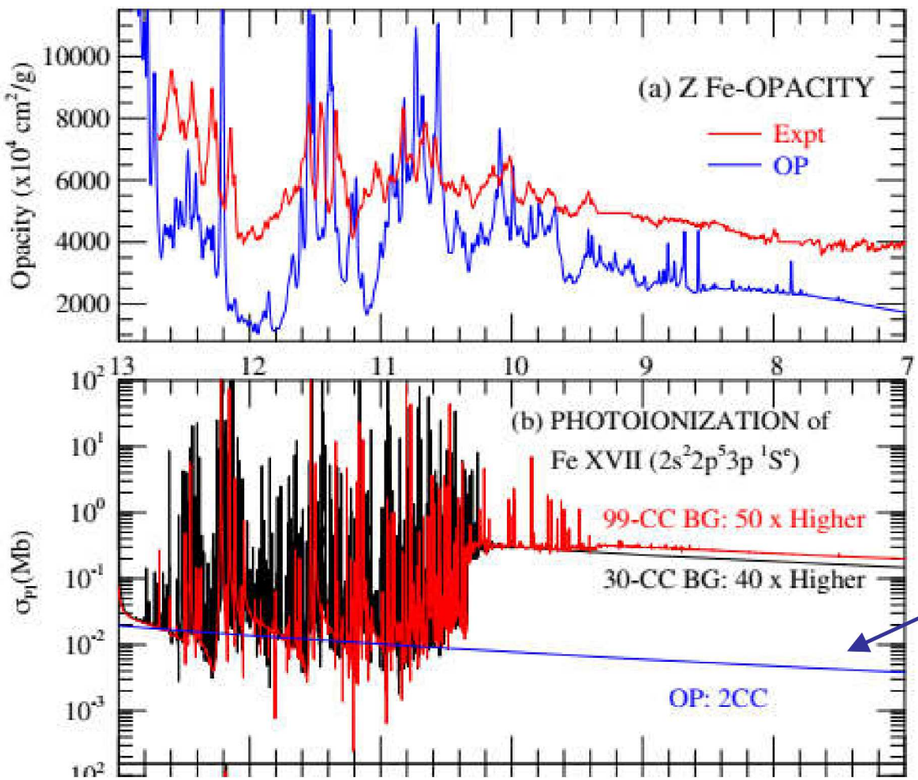
High iron continuum was especially worrisome



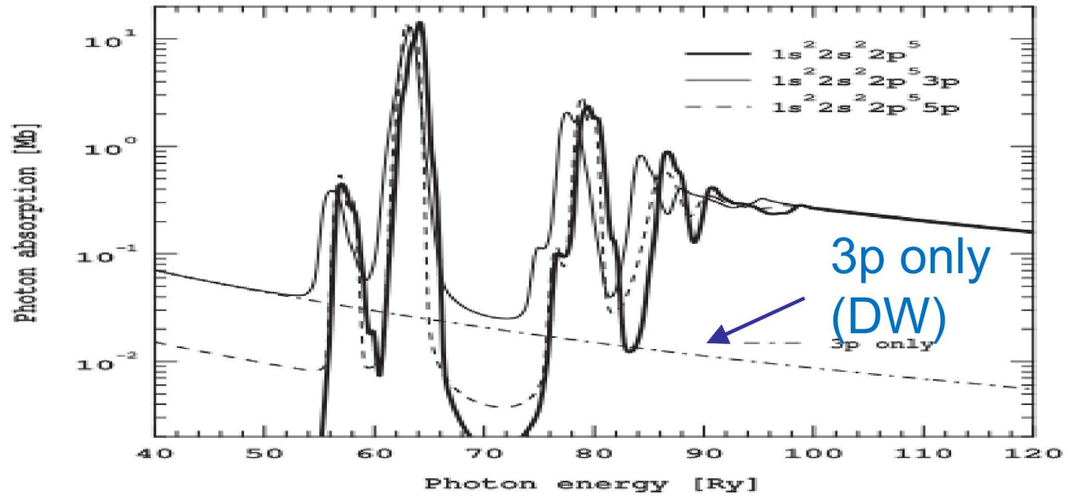
Nagayama et al, PRL 122, 135001 (2019)

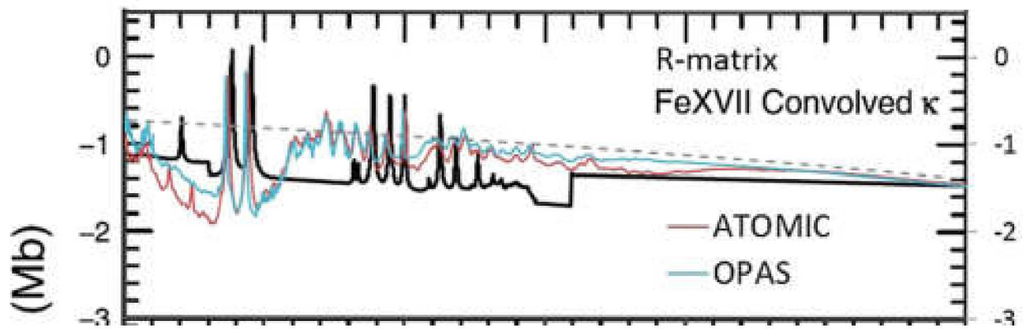
- Iglesias HEDP 15, 4 (2015): Sum rule violation?
  - Measurement range was insufficient for rigorous test
  - Absent other processes or redistribution, higher-than-cold σ^{PI} indicates $N_{L\text{-shell}} > 8$
- No similar discrepancy observed for Cr, Ni, or lower-T,n_e Fe – why?
- Reanalysis lowered Fe measurement, but discrepancy remains:
  - States: detail/accuracy? Completeness?
  - Rates: detail/accuracy? Completeness?

Rates: Do close-coupling/R-matrix calculations help?



3p only (R-matrix)



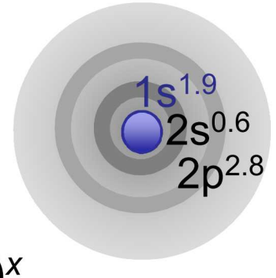


- Nahar and Pradhan, PRL 116 (2016) proposed that close-coupling/R-matrix cross sections might be more accurate than distorted wave (DW), thereby enhancing σ^{PI}

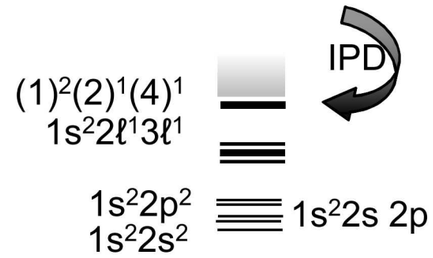
- Iglesias & Hansen (ApJ 835, 2017) showed that incompleteness in previous CC models accounted for the reported enhancement
- Blancard et al (PRL 117, 2016) showed that DW is adequate (& much easier to make complete!)

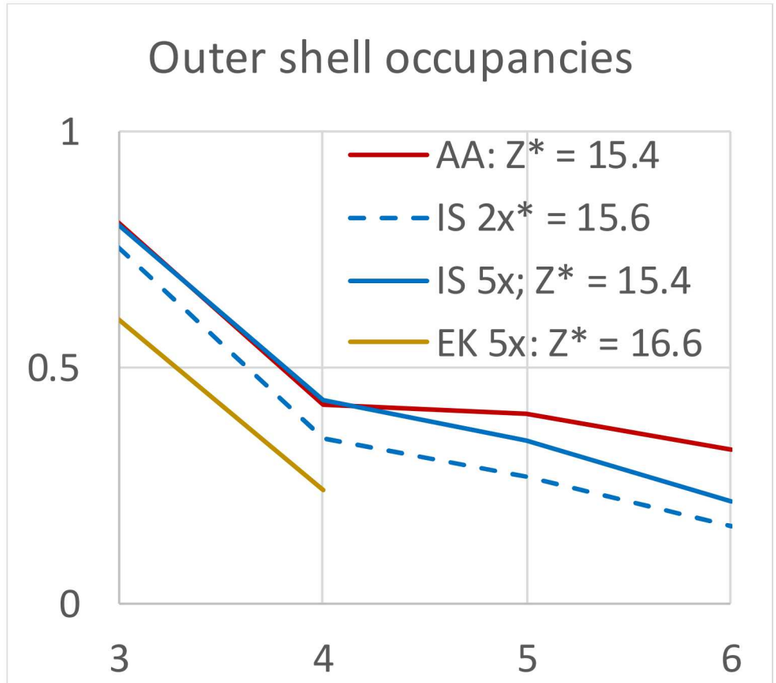
States: Are models including enough excitations?

- Average-atom models
  - Native and rigorous completeness
  - Native treatment of continuum lowering
  - Based on non-integer-occupied orbitals $(nl)^x$



- Configuration-based models $(nl)^N$
  - Externally imposed continuum lowering
    - SCRAM uses ion-sphere for consistency with average atom
- Completeness can be rigorously assessed
  - SCRAM uses systematic excitation of all electrons from all shells until $dN < \varepsilon$ (cf. James Colgan)
- Completeness is also needed at low density for dielectronic recombination (cf. Howard Scott)





Rates: are models missing an important process?
A two-photon interlude

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- Two-photon absorption had been discounted as negligible for K-shell (More & Rose, 1991)
- Increases in strength with n^8 *and with state complexity* so maybe not negligible here
- Has its own internal sum rule
- Multi-photon absorption/ionization/decay is well known for optical processes (NLO)
- X-ray two-photon ionization has been measured in XFEL experiments

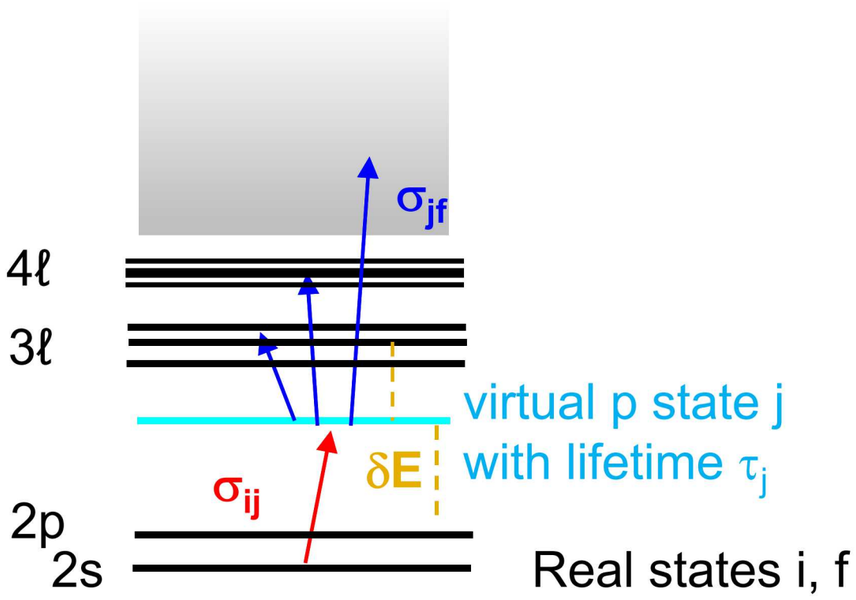
The 2015 iron results stimulated significant work:

- More et al HEDP 24, 44 (2017): initial calculations
- Pain HEDP 26, 23 (2018): hydrogenic, one-color
- Kruse & Iglesias HEDP 31, 28 (2019): total two-photon from 2s
- Colgan & Pindzola (private communication): TDSE cross sections
- More et al, HEDP 34, 100717 (2020): f-f matrix elements
- Baggot, Rose,& Mangles (2020): $\gamma + \gamma$ & $e + \gamma$; line shapes

Accuracy, detail, and completeness all matter, and including all together is challenging

- More, Kruse & Iglesias, and Pain all compute accurate QM two-photon cross sections
- I tried something reckless: the terrible-for-opacity-but-natively-complete average atom plus some stuff from nonlinear optics (Lambropolous, Adv. At. Mol. Phys. 12, 87 (1976))

$$\sigma^{TPA} \sim \sigma_{ij}(h\nu_1)\ \tau_j\ \sigma_{jf}\ (h\nu_2)\ [cm^4\ s].$$



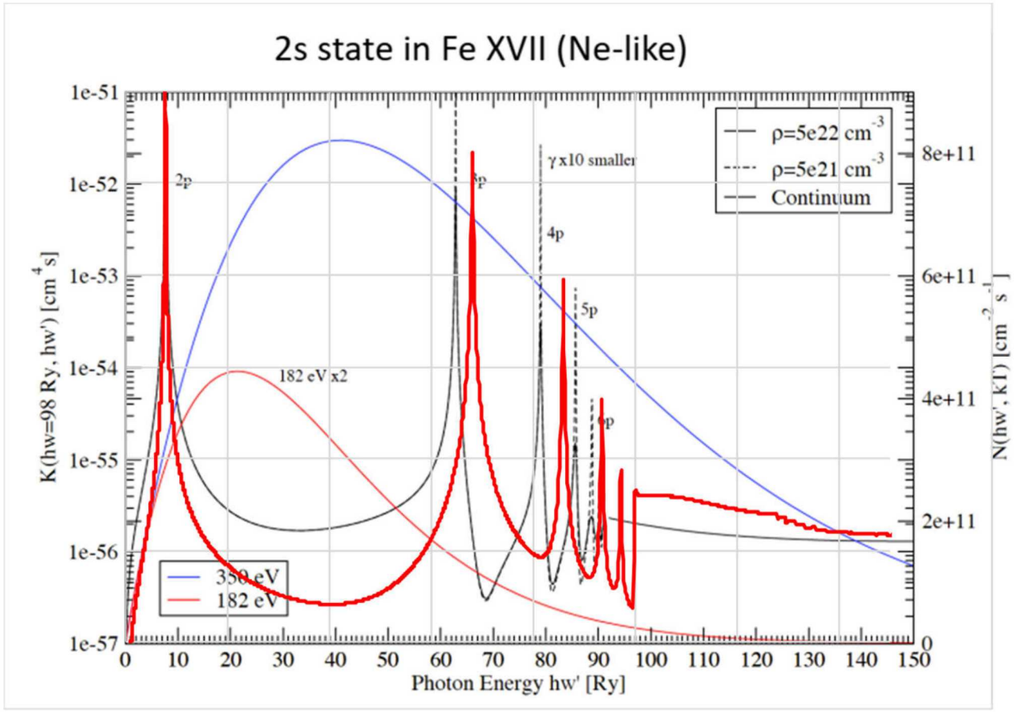
- σ are one-photon cross sections (~10^{-20} cm^2)
- τ are lifetimes of virtual states ($h/\delta \mathbf{E} + \tau_c$ ~10^{-14} s)
- → σ^{TPA} ~10^{-54} $cm^4 s$
- Must sum over all virtual and final states
- Backlighter photons can be either $h\nu_1$ or $h\nu_2$, so we must integrate *twice* over local rad field FdE (~eV/ eV-s-cm^2)

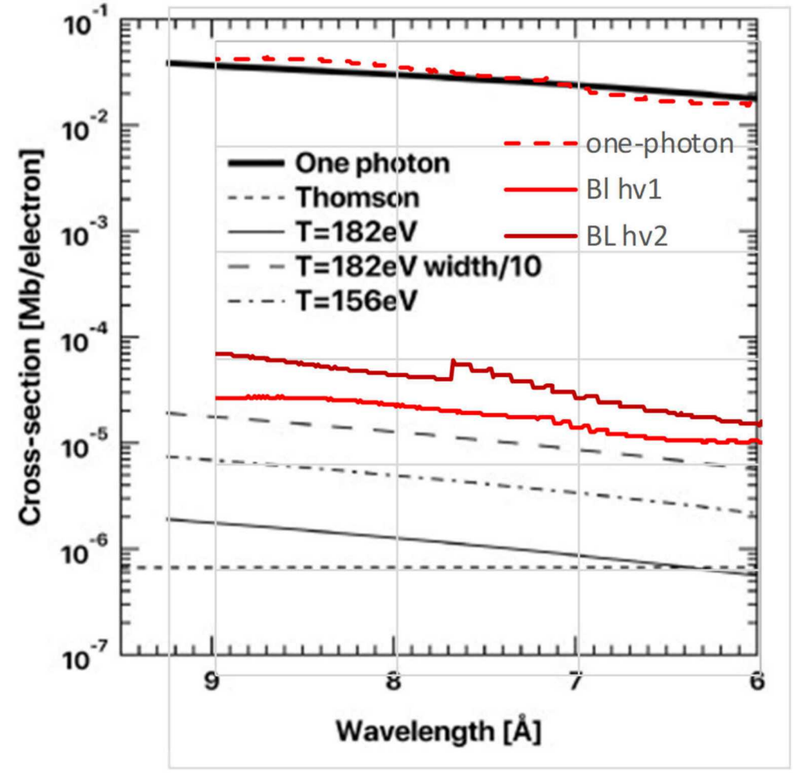
This approach tracks a *complete* set of states and i – j – f channels, but with much less detail than multiconfiguration one-photon models

Sparse states that maximize δE will minimize two-photon contributions

Test approximation for hv_1 = 1.3 keV backlighter photon from 2s against accurate $2s^2\,2p^6 - 2s\,2p^6\,nd$ calculations

This is the case considered by Kruse & Iglesias

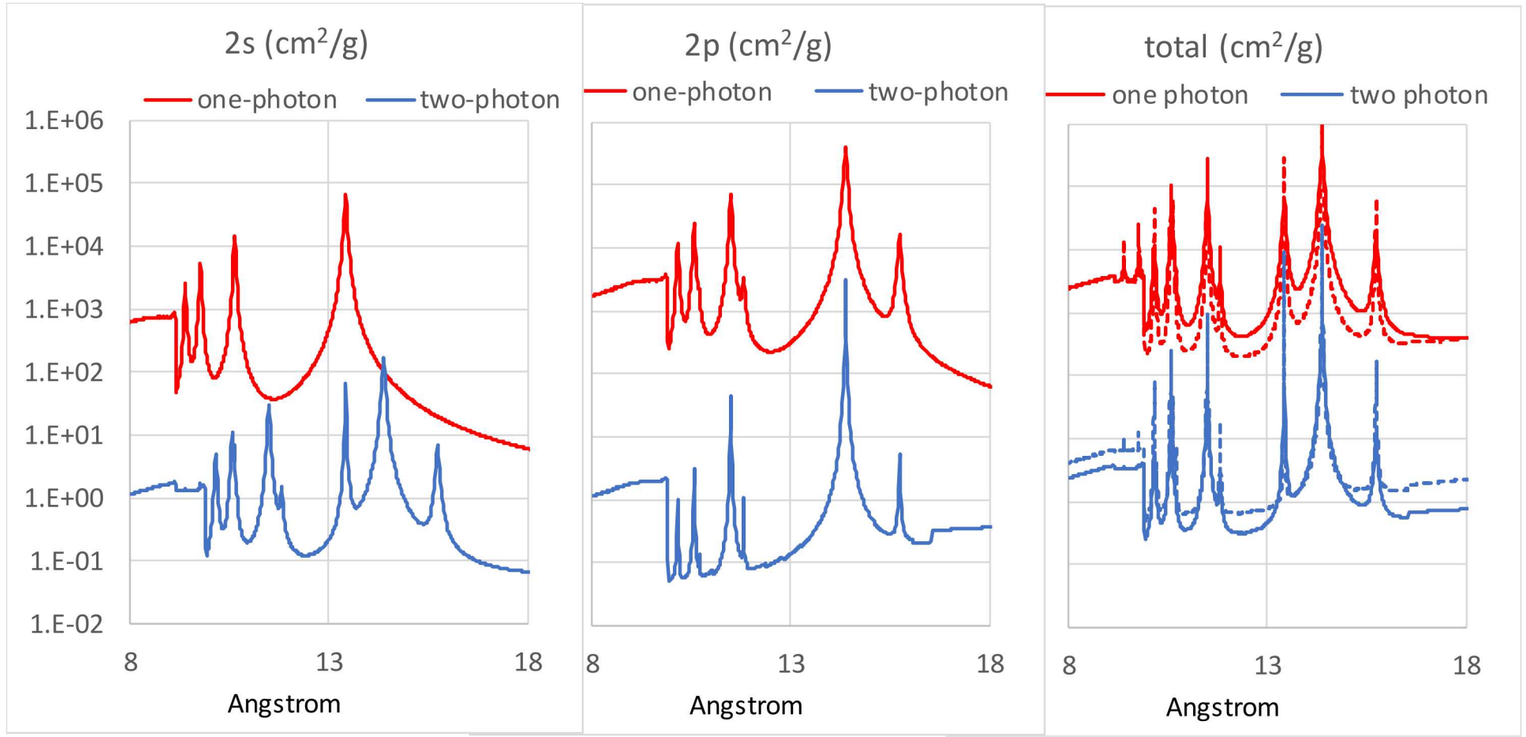




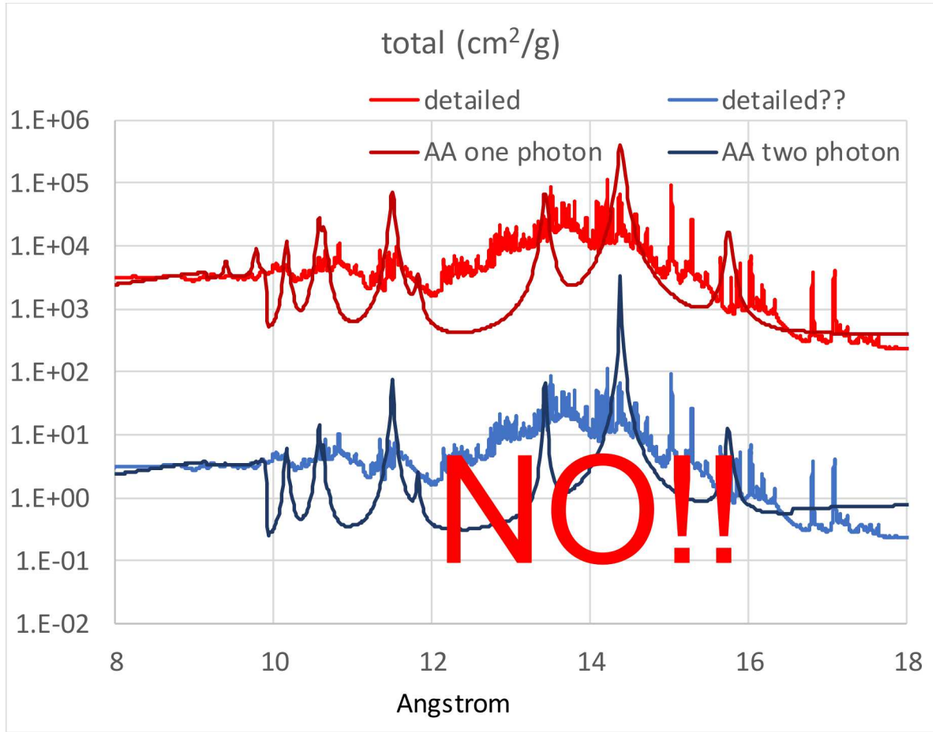
The approximate AA cross section is similar in character to Kruse & Iglesias' accurate calculation for detailed Ne-like Fe

First integral is similar to Kruse & Iglesias
Second integral (backlighter photon hv_2) is ~2x first
Add together

Approximate two-photon absorption from undetailed average atom is small, but detailed structure *matters*

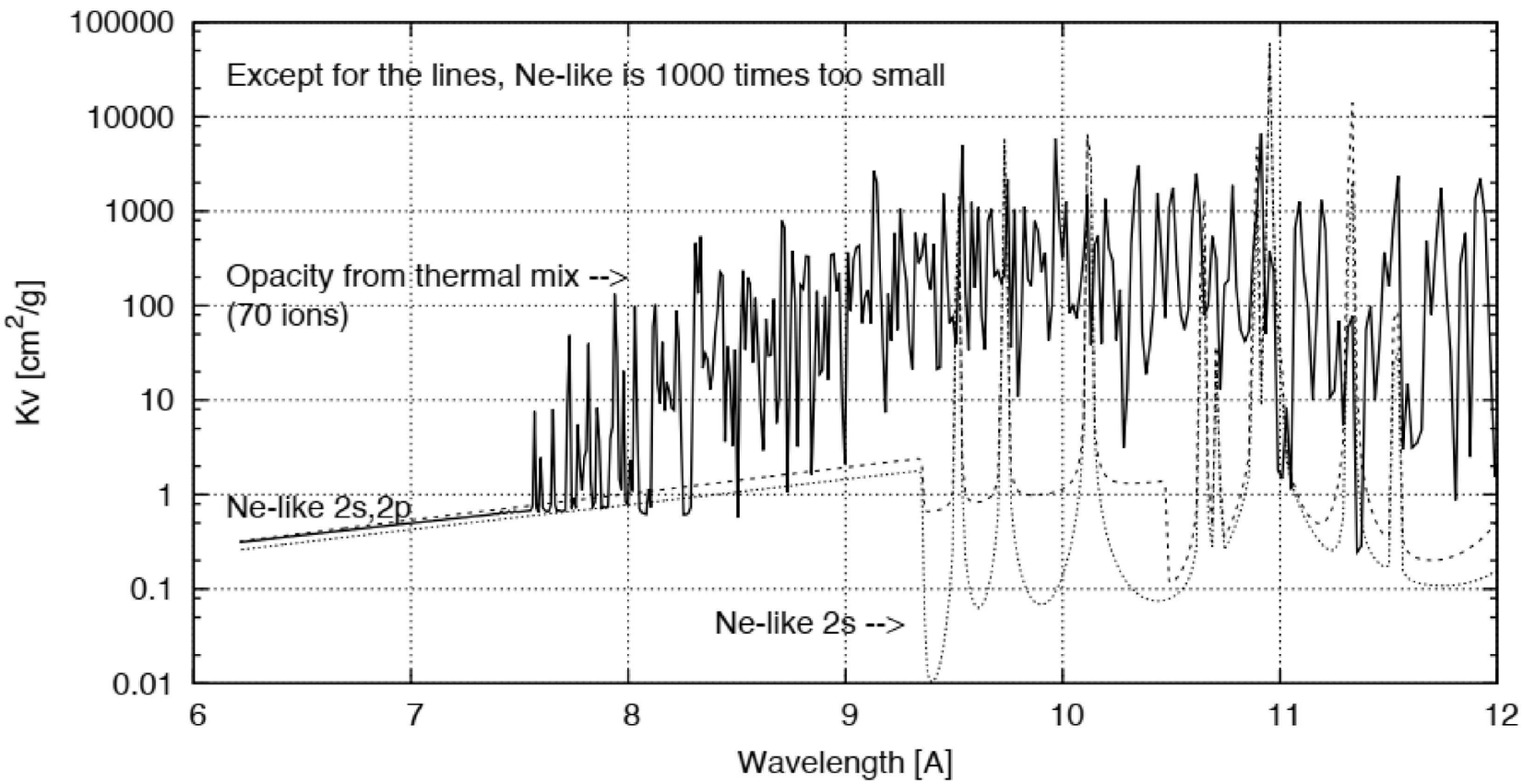


- 2p, 2s , & total two-photon cross sections from the average atom are all 1000x smaller than one-photon
- Like Kruse & Iglesias, AA results are dependent on line widths



While σ_{ij} and σ_{jf} both obey sum rules and preserve strength under splitting into detailed structure, the two-photon σ_{ij} $(h/\delta E)$ σ_{jf} cross section can get much larger with more detailed structure

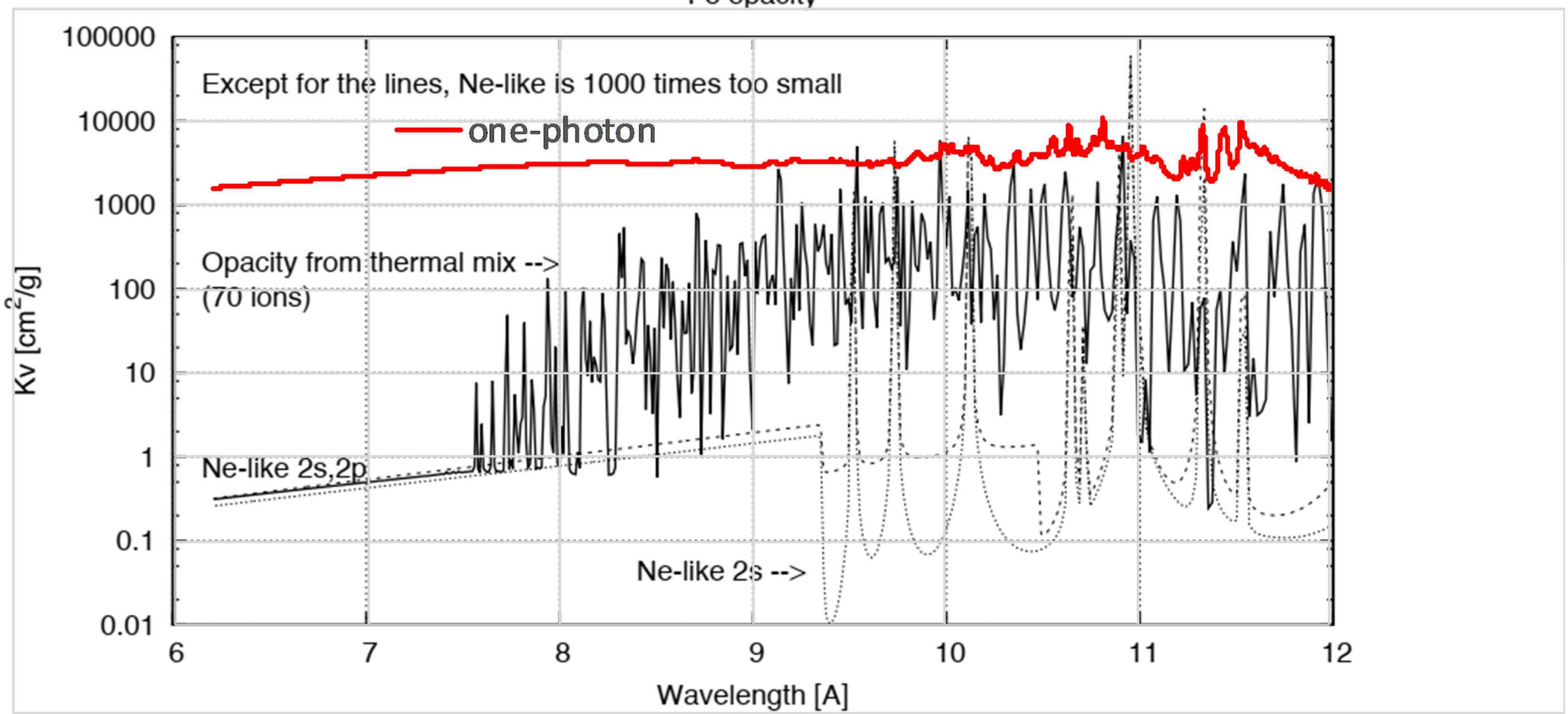
Fe opacity - two-photon and Raman effect



Calculations from Richard More

Ne-like ground-state opacity is hopelessly small (on this range).
Certain excited states of Ne-like ion have larger cross-sections.

Fe opacity - two-photon and Raman effect



Calculations from Richard More & SCRAM

Ne-like ground-state opacity is hopelessly small (on this range).
Certain excited states of Ne-like ion have larger cross-sections.

Line shapes matter for two-photon absorption, and completeness, detail, and consistency matter for line shapes

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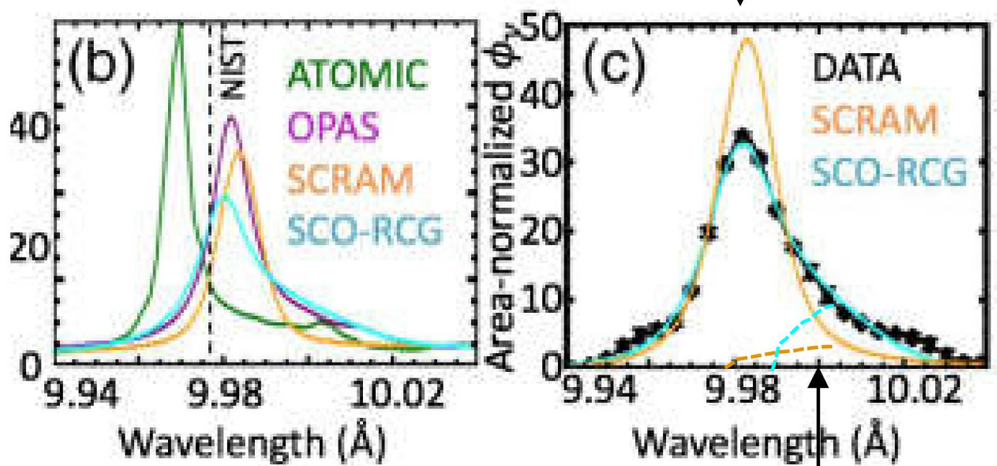
- Completeness & detail
  - Instrumental & thermal broadening (0.4, 0.1 eV)
  - Stark broadening from external fields (1-3 eV)
  - Natural broadening (0.7 eV)
  - Collisional broadening from collisional ex/dex (0.7 - 1 eV; must include $\Delta n=0$)
  - Additional collisional broadening from ion/rec? (~0.05 eV for $\Delta Z = 1$)
    Gomez et al PRL 214, 055043 (2020)
  - Complete set of spectator satellites
    Satellites seem best captured by SCO-RCG: SCRAM's hybrid-structure satellites are weak/broad and too close to resonant 2p-4d line, while ATOMIC's nlj satellites appear too far from line

- **Are collisions, fields, and structure consistent?**

Ne-like Ni 2p – 4d
(2.8 experimental width)



2p nl – 4d nl

Nagayama et al, PRL 122, 135001 (2019)
J-C Pain and F Gilleron: SCO-RCG

Consistency: what makes a consistent model, and why should we care?

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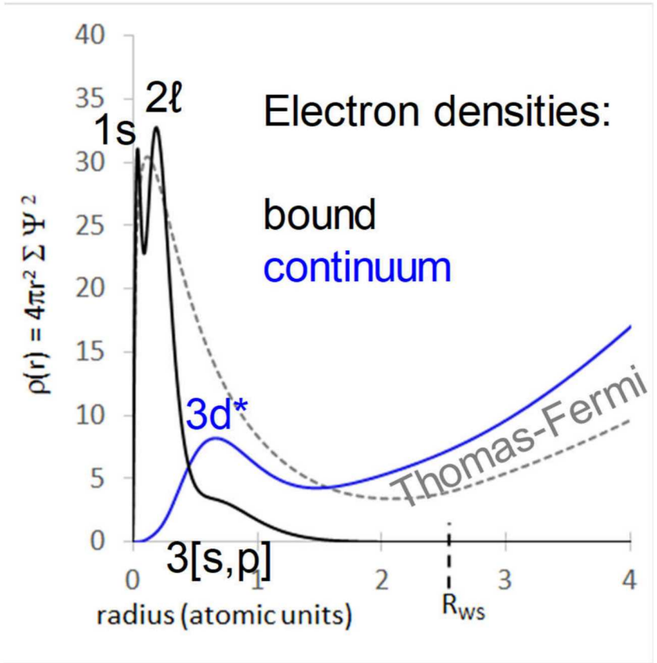
- <u>Consistency has multiple aspects</u>
  - Density effects on electronic structures/wavefunctions & rates
    → average atom/DFT/self-consistent-field models
  - Electronic screening effects on ion structure
    → Neutral pseudo atom (e.g. Starrett & Saumon)
  - Lines shapes consistent with electronic & ionic structure
    → rates from electronic structure, Stark fields from ionic structure
- Simulations often draw from separate models for EOS, transport, opacity
  → can we build a single model that generates everything?

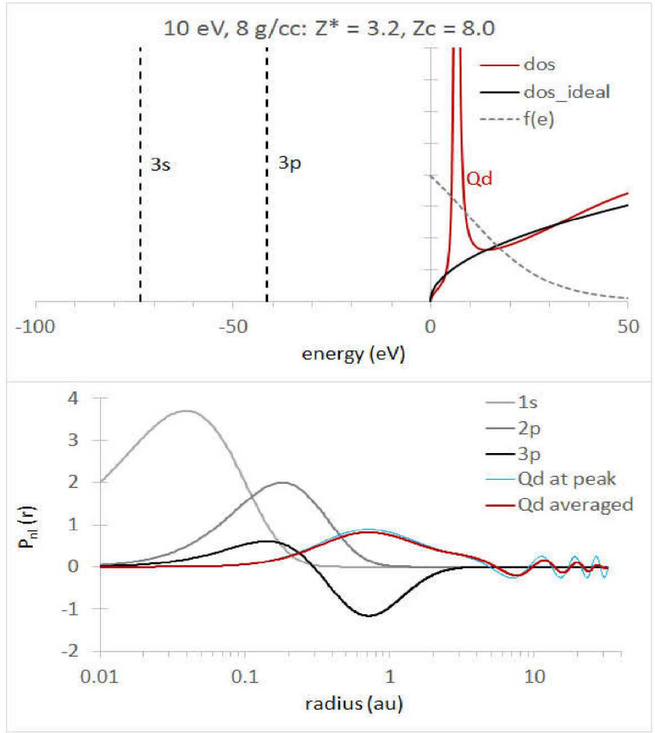
Rigorously enforcing consistency within models can help us constrain models even in regimes without high-precision measurements

Our approach: enforce consistency wherever possible

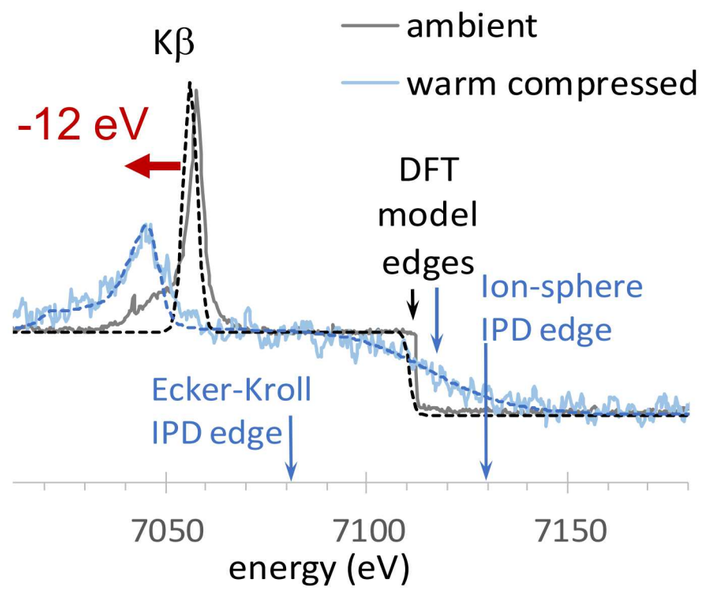
- In wavefunctions, atomic structure/EOS, and rates



Self-consistent field model lets electronic wavefunctions respond to changes in density and temperature



Continuum lowering is natively incorporated in structure & EOS (nothing ad-hoc)



Rates and transition energies respond to changes in the wavefunctions

Hansen et al HEDP 2017
Jiang et al PRE 2020

Liberman, *Phys Rev. B* **20**, 4981 (1981); Wilson, Sonnad, Sterne & Isaacs *JQSRT* **99**, 658 (2006)

Our approach: enforce consistency wherever possible

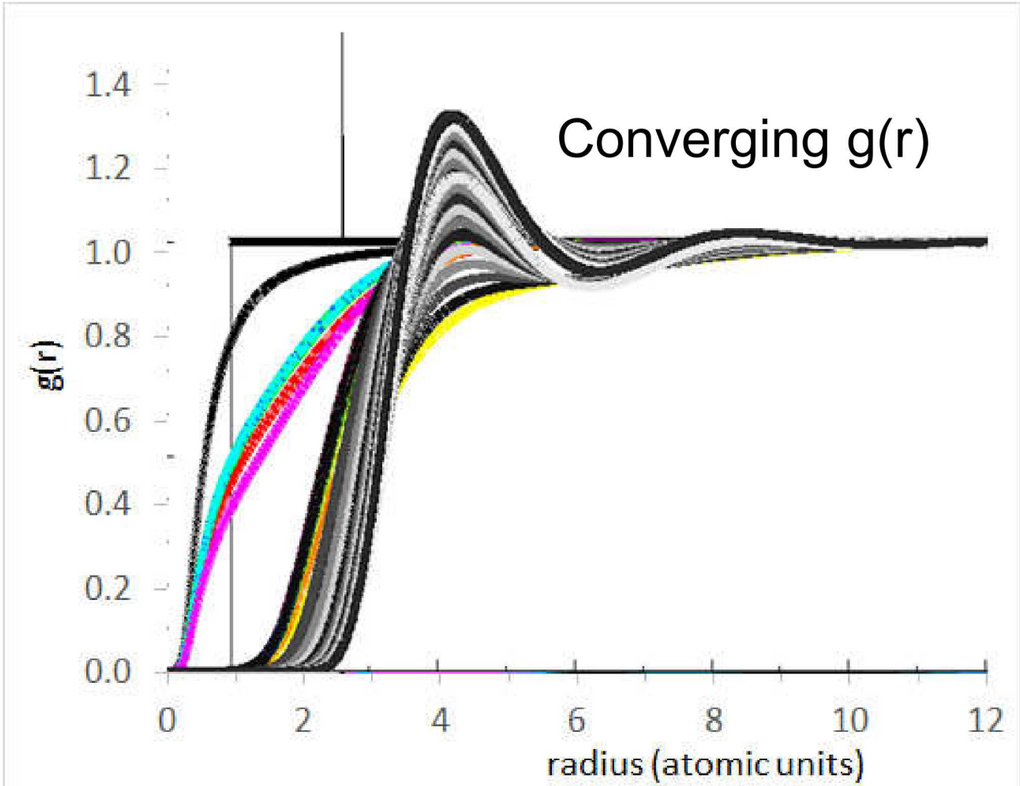
- Between electronic structure and ionic structure

The screening electron density determines the ion-ion interaction potential:

$$\beta V(k) = \frac{4\pi\beta\bar{Z}^2}{k^2} - n_e^{scr}(k)C_{le}(k)$$

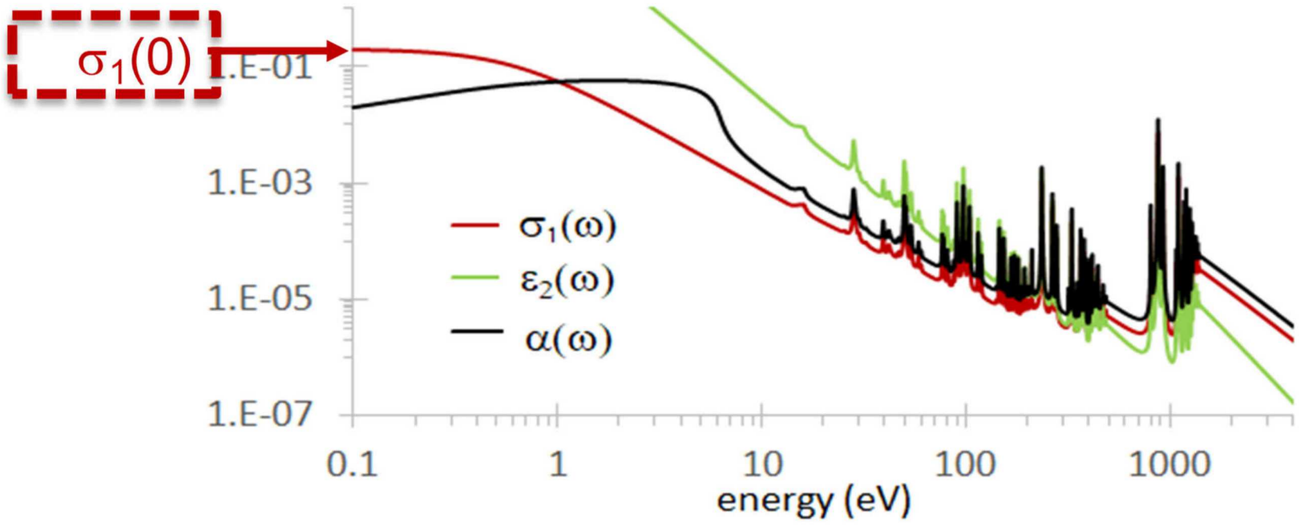
This potential constrains the ion distribution through the quantum Ornstein-Zernicke equations – or through classical MD simulations

Solid-density iron, 10 eV



Our approach: enforce consistency wherever possible

- Between electronic & ionic structure and transport quantities



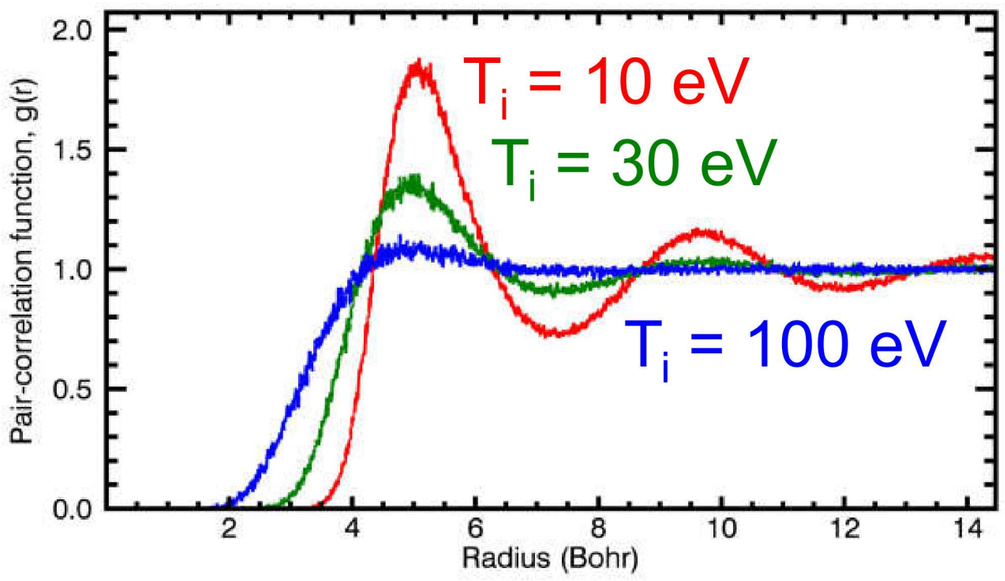
Response functions based on matrix elements of average-atom wavefunctions and ion g(r) constrain:

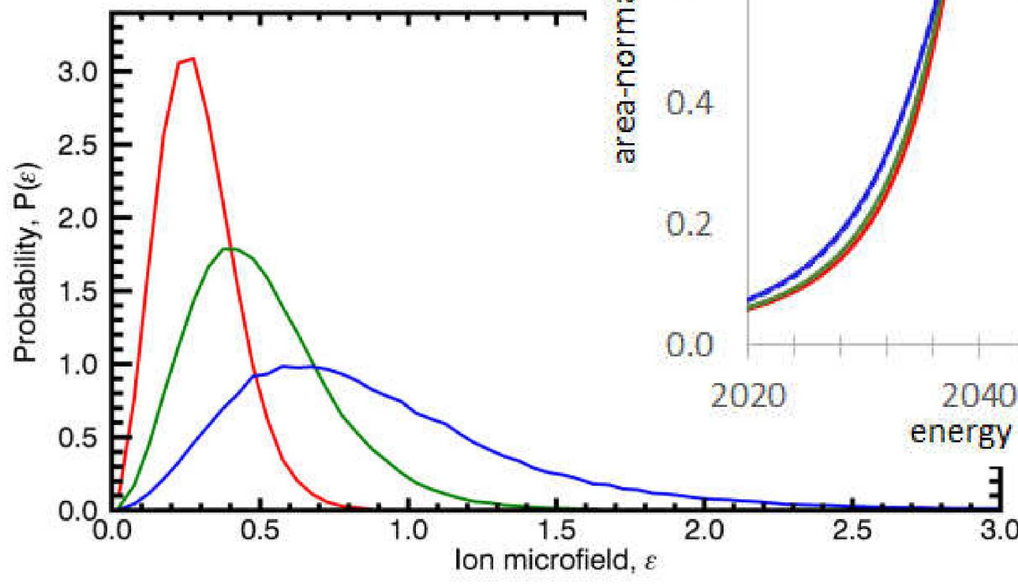
- Electrical and thermal conductivities (cf. M. Desjarlais)
- X-ray Thomson scattering signatures (A. Baczewski)
- Stopping powers (T. Hentschel)
- Collision frequencies

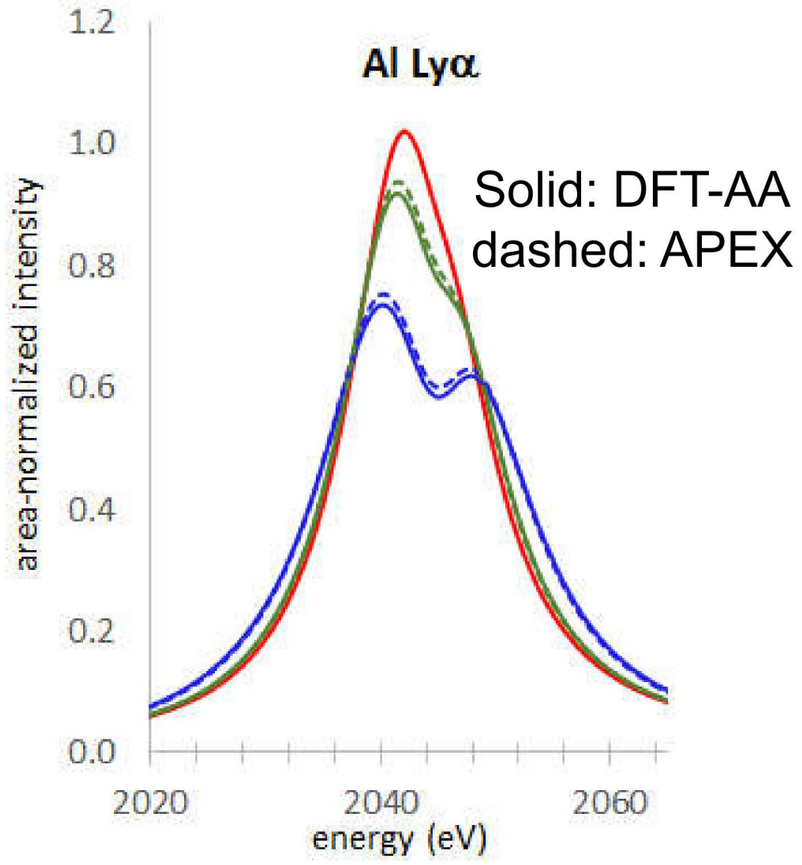
Our approach: enforce consistency wherever possible

- Between electronic & ionic structure and line shapes

Solid-density Al, T_e = 250 eV







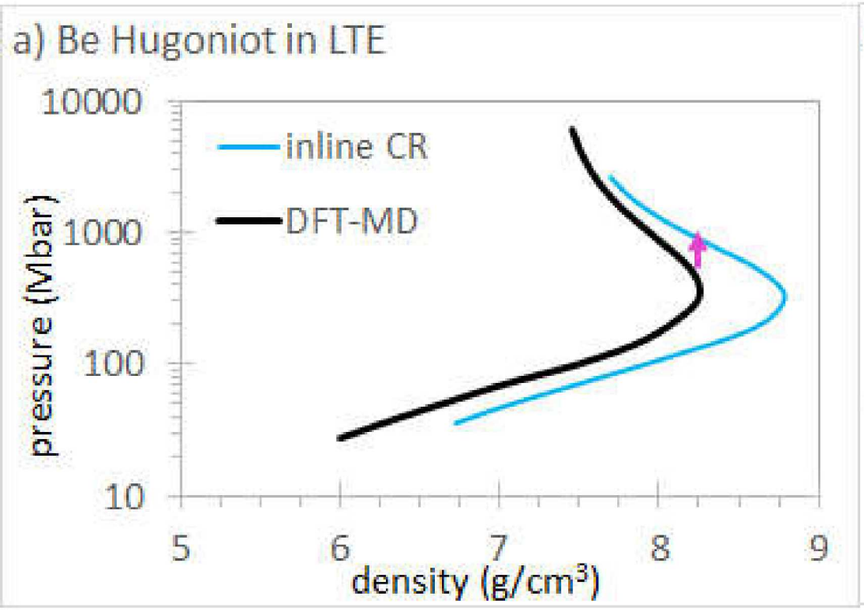
T. Gomez

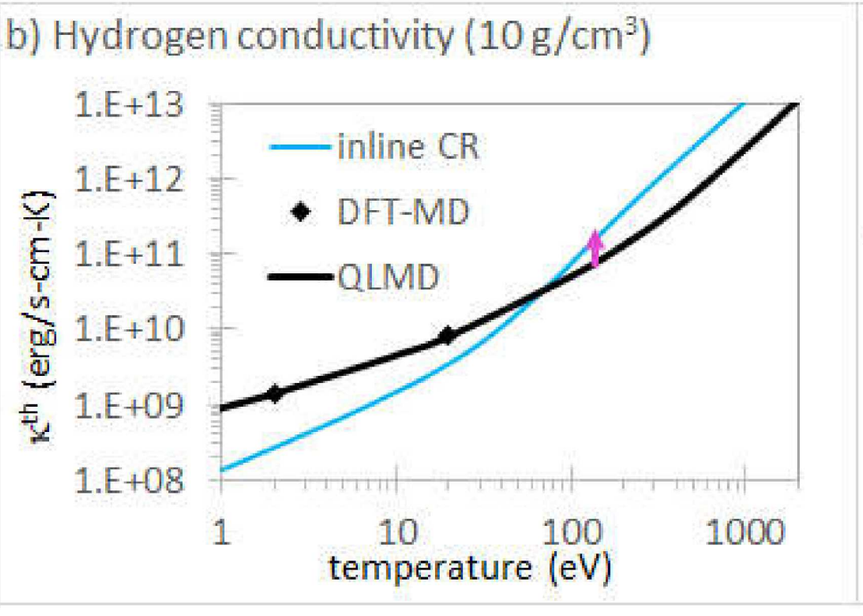
T. Nagayama, (M. Murillo, L. Stanek)
cf. Starrett's PAMD

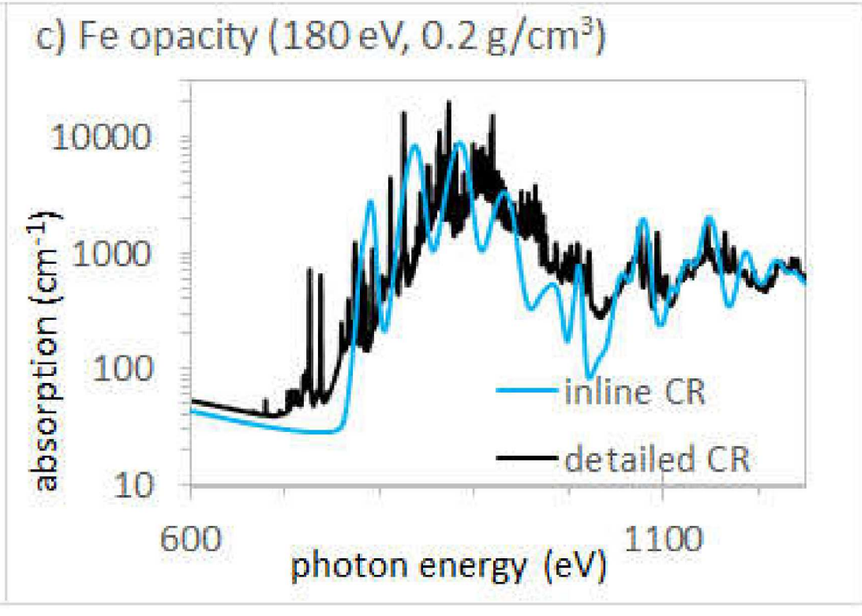
APEX: Iglesias *et al.*, *Phys Rev A* **31**, 1698 (1985)

Why we care: the effects of inconsistency on simulations is not well understood

- Simulations draw different quantities (EOS/transport/opacity) from different tables and models
- Parameterized quantities (or inline non-LTE) can increase consistency, at the expense of discontinuities and/or reduced resolution/detail







Our goal: a spectroscopically detailed, rigorously complete, and internally consistent model

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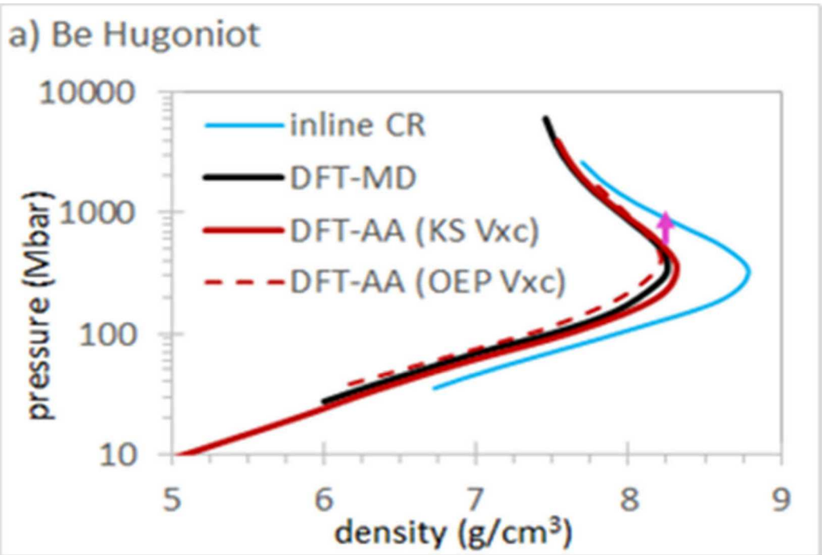
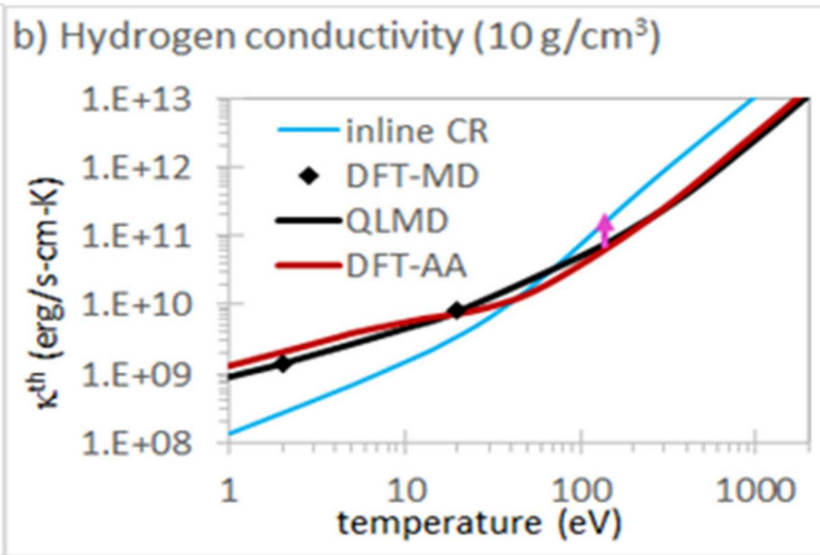
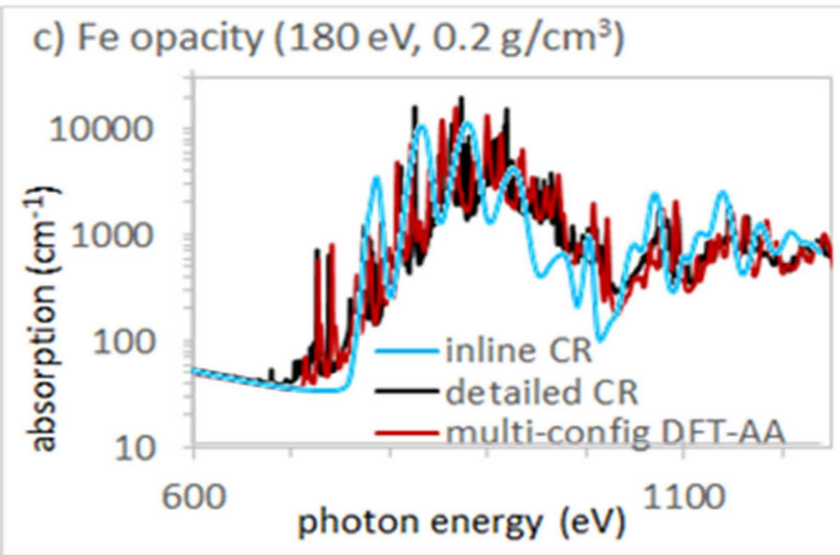
- Use AA wavefunctions to build multiconfiguration models
- Extensible to non-LTE + additional processes/quantities as needed (e.g. two-photon)
- Validated against DFT-MD, TD-DFT, and detailed CR models, where available
- Verified against high-precision measurements, where available
- Tractable enough for tabulation (cf. H. Scott)







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Thank you!

Questions?

Consistency: it is hard to use one model for everything

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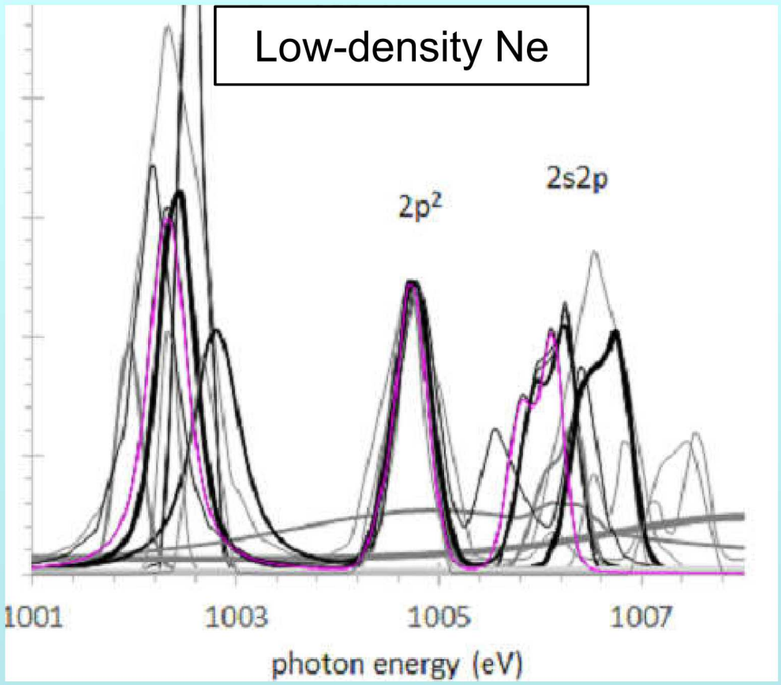
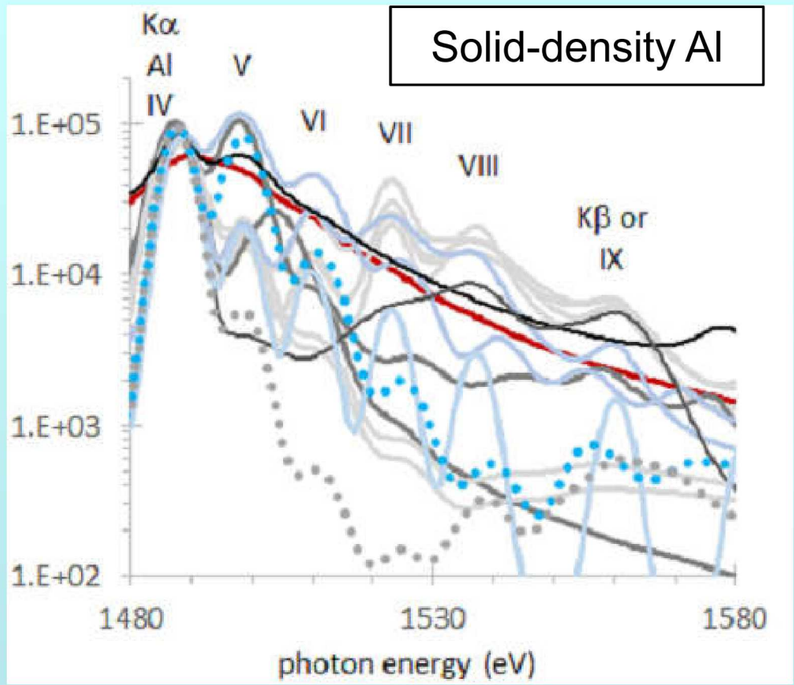
Detailed models enable precision diagnostics for plasmas at modest densities



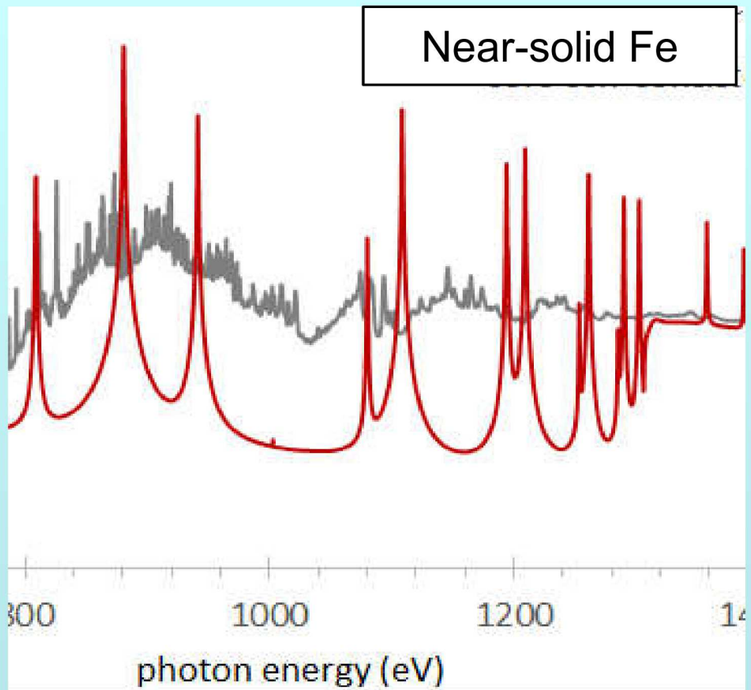
S. Hansen *et al,*
Review of the 10th NLTE code comparison workshop, HEDP (2019)

At high densities, *ad-hoc* corrections lead to incoherence



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

Models that natively include density effects tend to lack detail



cf. P. Sterne *et al,* HEDP **19**, 1 (2007)

Extending existing DFT-AA model to non-LTE will offer unprecedented consistency in material data

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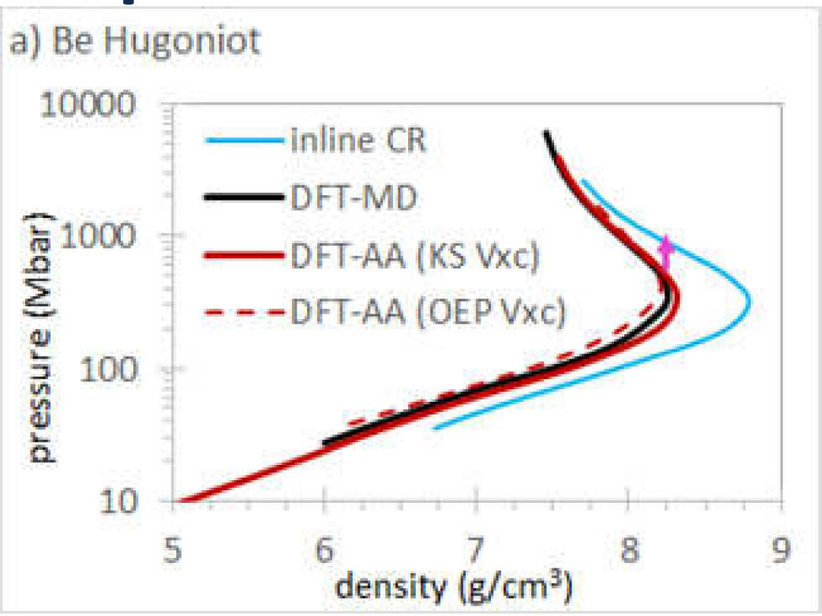

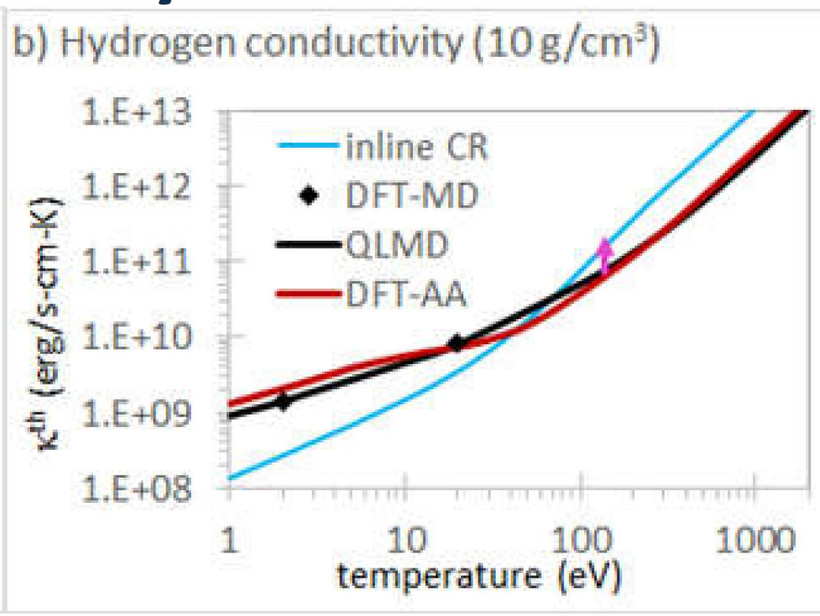

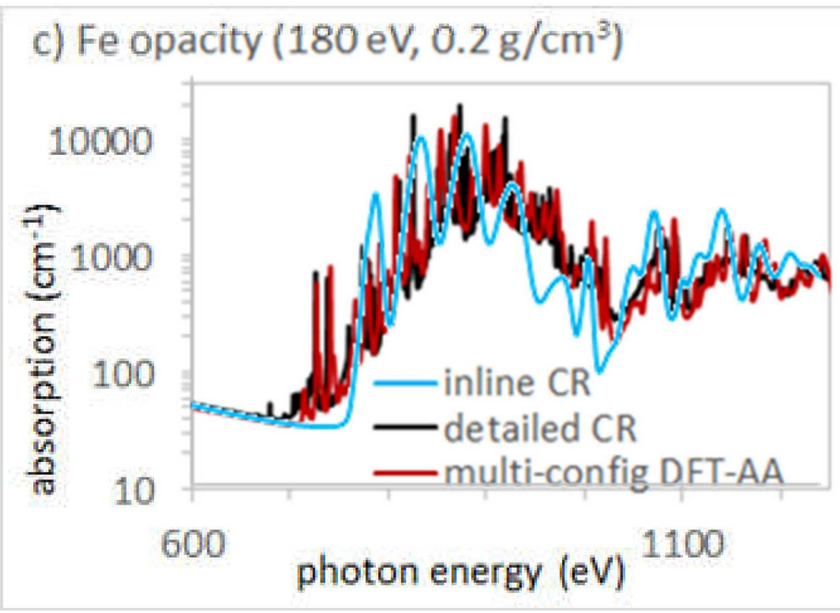

- Rigorous internal consistency/ DFT foundations eliminate “seams” & ill-quantified uncertainties (1)
- Detail could revolutionize design cycles, enabling routine generation quality synthetic diagnostics (3)
- UQ could be undertaken on *whole tables* rather than piecemeal quantities (outyears)

To be done:

Add rates to multi-configuration model → CR + new self-consistent electron & ion

Add configuration-interaction + implement line shapes in select levels for diagnostics

Find and enforce mutual constraints among properties (e.g. line shapes, $\partial E/\partial x$, σ)

Compare to existing tables / DFT-MD/ TD-DFT/ detailed CR models to benchmark

High-T and high-Z plasmas (RES, ICF mix/diagnostic) are particularly sensitive to non-LTE effects

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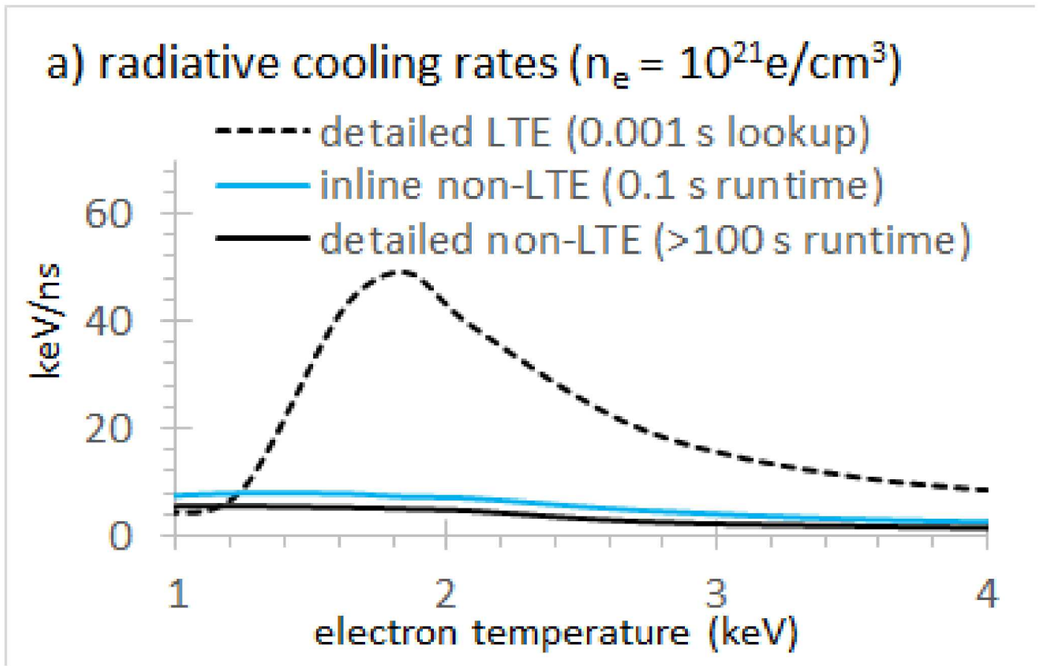
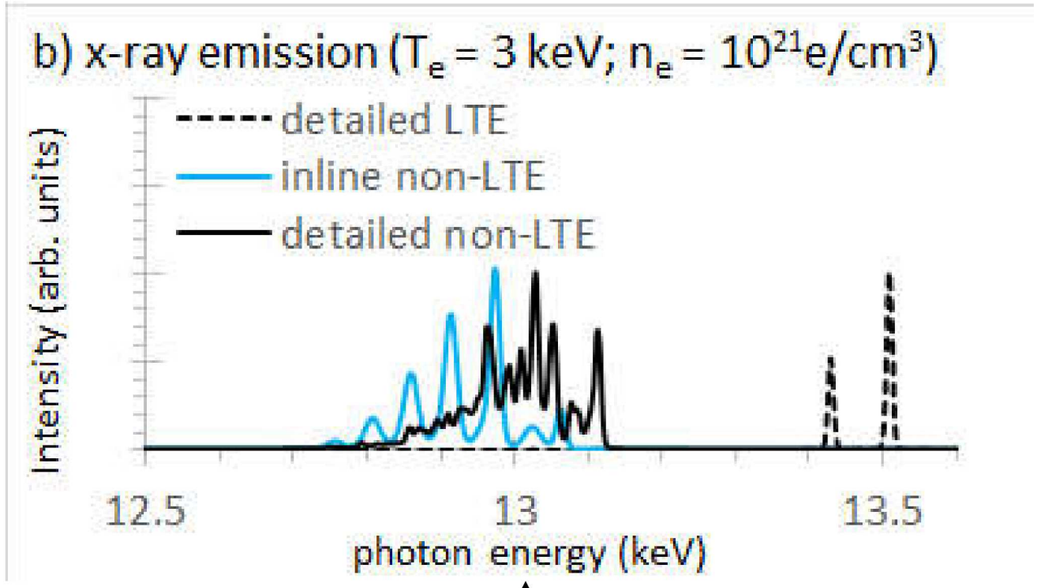
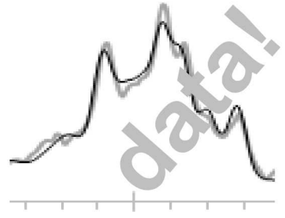
Exemplar: Kr-doped MagLIF
LTE (Tr =Te) v. non-LTE (Tr = 0)





Compared to the DFT models used for LTE tables, inline CR models are *slow* and *bad*



… and inform stopping powers (essential for self-heating)

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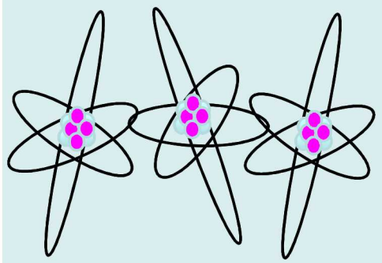
$$L(\rho,V)=\frac{i}{\pi\omega_0^2}\int_0^{\infty}\frac{dk}{k}\int_{-kV}^{kV}\omega dw\left(\frac{1}{\varepsilon(k,\omega)}-1\right)$$

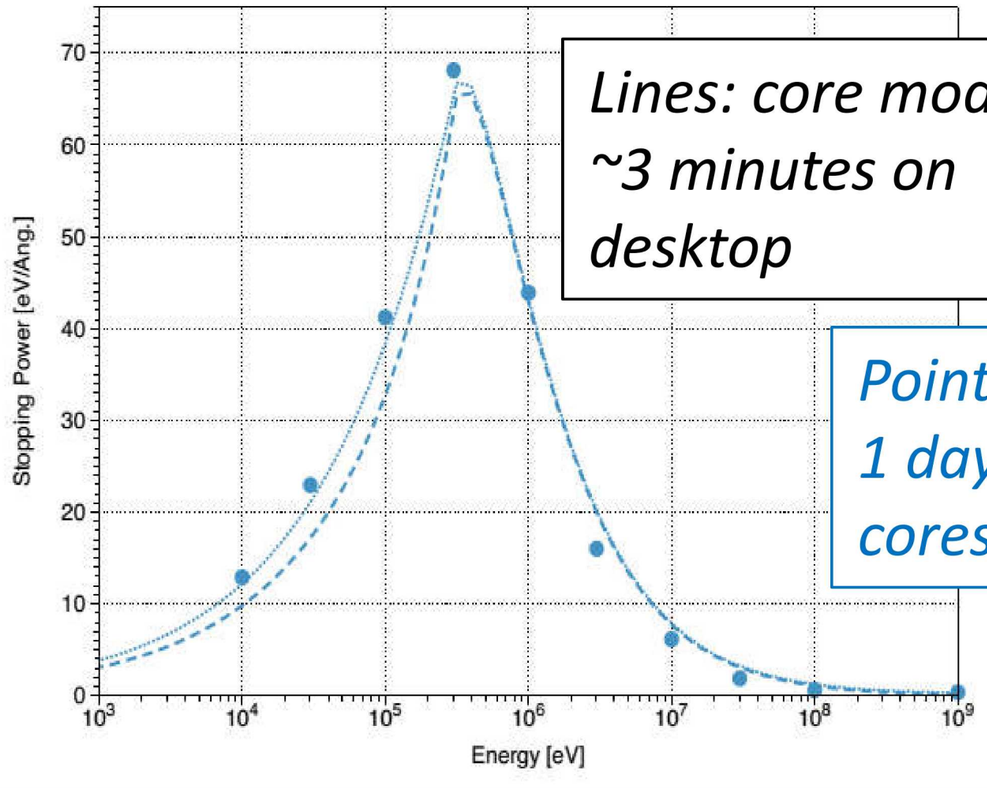


Core model: quantum average atom + ion correlations

$\sigma(\omega)$, $\varepsilon(\omega)$, κ^{th}, $\partial E/\partial x$

$$\left(\frac{dE}{dx}\right)=-\frac{4\pi}{m}\left(\frac{Ze^2}{V}\right)^2\int_0^{\infty}\rho(r)L(\rho,V)4\pi r^2dr$$

Protons stopping in 10 g/cc deuterium

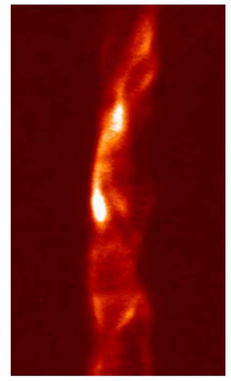


Lines: core model ~3 minutes on desktop

Points: TD-DFT 1 day on ~1000 cores

TD-DFT stopping: A. Cangi & A. Baczewski

Astrophysics & ICF



Wang *et al*., *Phys Plas* **5**, 2977 (1998); Magyar *et al*., *C. Plas Phys*. 56 459 (2016)

… and X-ray scattering (used to diagnose laboratory plasmas)

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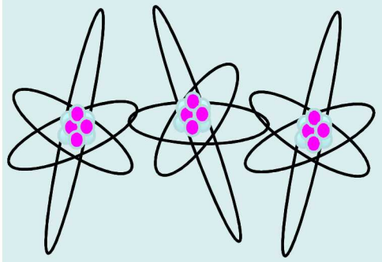
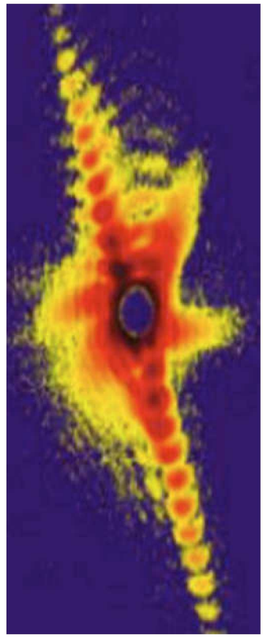
$$S(k,\omega) \propto \mathrm{Im}\left[\frac{1}{\varepsilon(k,\omega)}\right]$$



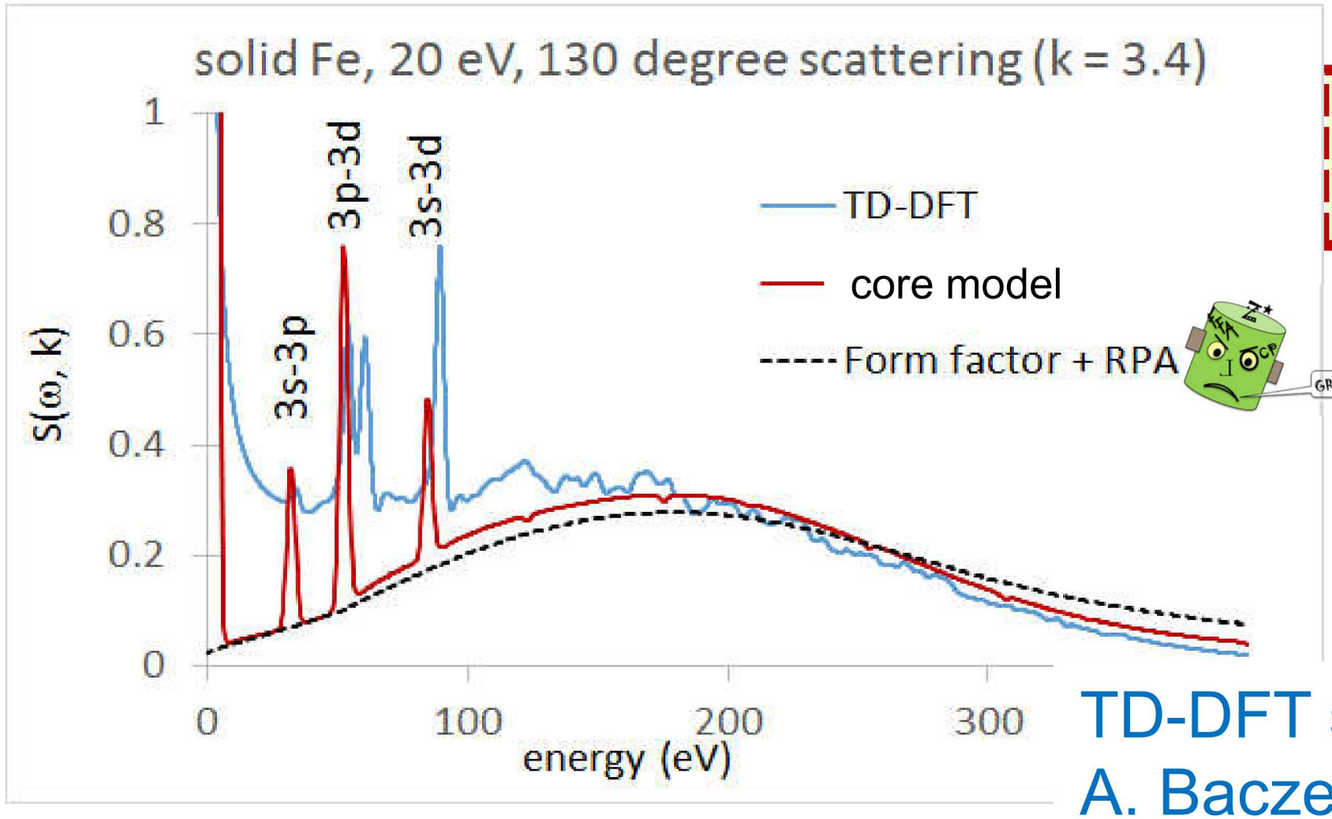
Core model: quantum average atom + ion correlations

$\sigma(\omega)$, $\varepsilon(\omega)$, κ^{th}, $\partial E/\partial x$

X-ray scattering



XFEL



TD-DFT scattering: A. Baczewski

Chihara decomposition:

$$S(k,\omega) = \underbrace{|f_I(k) + q(k)|^2 S_{ii}(k,\omega)}_{\text{elastic}} + \underbrace{\bar{Z} S_{ee}(k,\omega)}_{\text{free-free}} + \underbrace{S_{bf}(k,\omega)}_{\text{bound-free}} + \underbrace{S_{bb}(k,\omega)}_{\text{bound-bound}}$$

Johnson *et al*, *PRE* **86**, 036410 (2012); Baczewski et al PRL 116, 115004 (2016)