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Dynamic collision frequencies, scattering, stopping, and spectra

S.B. Hansen, T.W. Hentschel, A. Kononov, A.D. Baczewski, K. Adler, and T. Gomez

Sandia National Labs
Cornell University

Radiative Proerties of Hot Dense Matter
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U.S. DEPARTMENT OF ENERGY | NNSA

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Overarching goal: Develop a complete and consistent atomic model for constitutive and observable data

1) Start with existing average-atom model based on density functional theory: **DFT-AA**

2) Use *ab-initio* multi-center DFT calculations to constrain DFT-AA model choices

3) Extend DFT-AA for dynamic properties (XRTS & line shapes)

4) Extend DFT-AA for multiconfiguration electronic structure (opacities)

5) Extend DFT- AA to non-equilibrium plasmas


SANDIA REPORT
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Improving Predictive Capability in REHEDS Simulations with Fast, Accurate, and Consistent Non-Equilibrium Material Properties

S.B. Hansen, A.D. Baczewski, T. Gomez, T.W. Hentschel, C.A. Jennings, A. Kononov, T. Nagayama, K. Adler, A. Cangi, K. Cochrane, B. Robinson, and A. Schleife

Rigorously validate against experimental data and reference models for XRTS, transport, EOS, non-LTE, and line shapes

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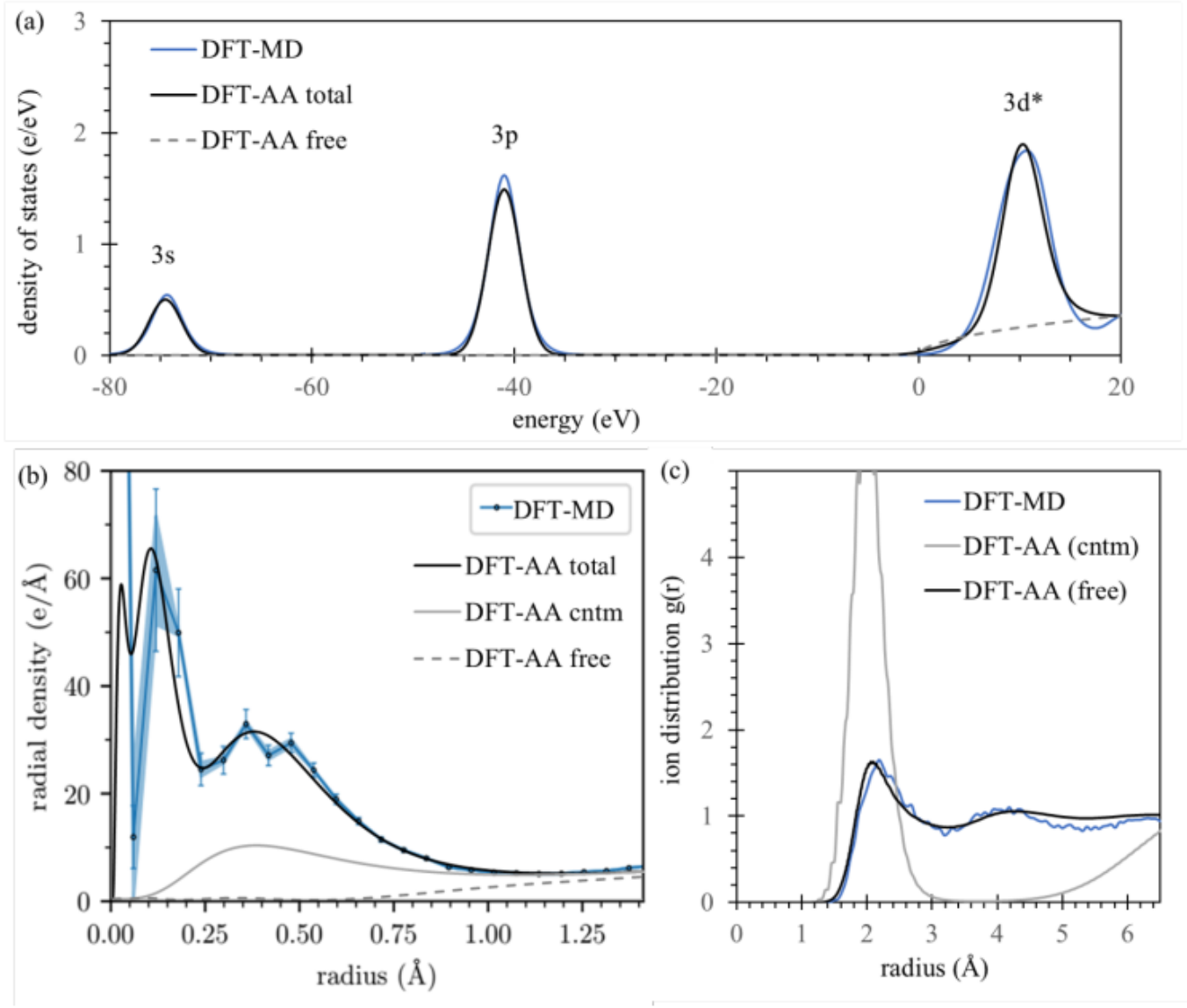
1) Start with existing average-atom model based on density functional theory: **DFT-AA**
→ self-consistent electronic and ionic structure, EOS, static transport coeff.

2) Use *ab-initio* multi-center DFT calculations to constrain choices
→ boundary conditions, Z*

Example: Solid iron at T = 1eV: electronic density of states (top) and radial distribution functions



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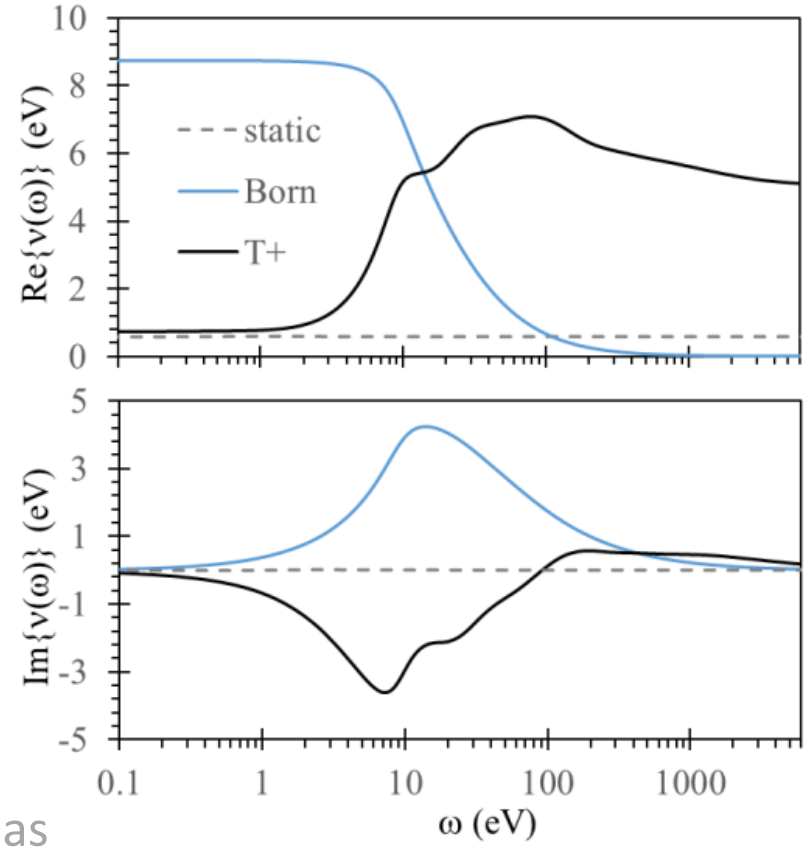
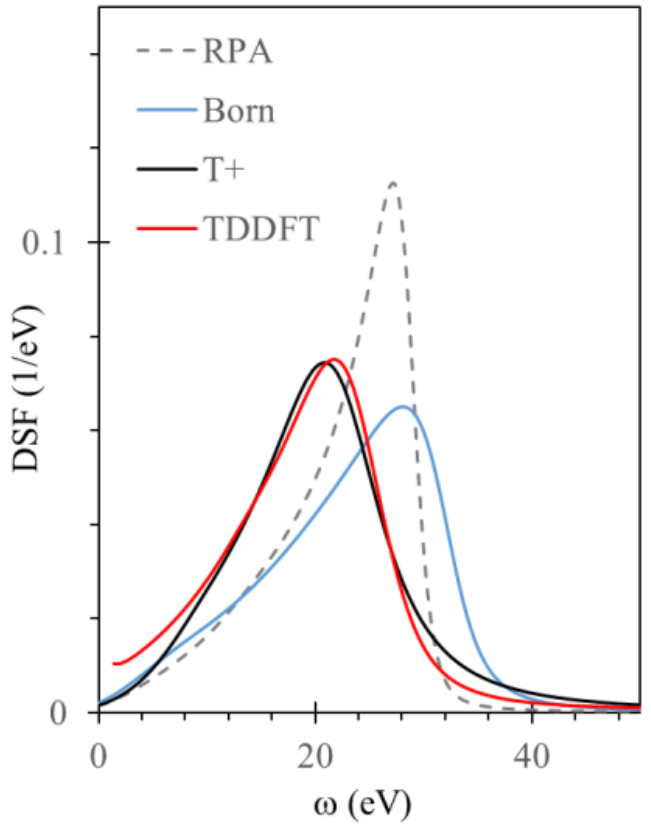




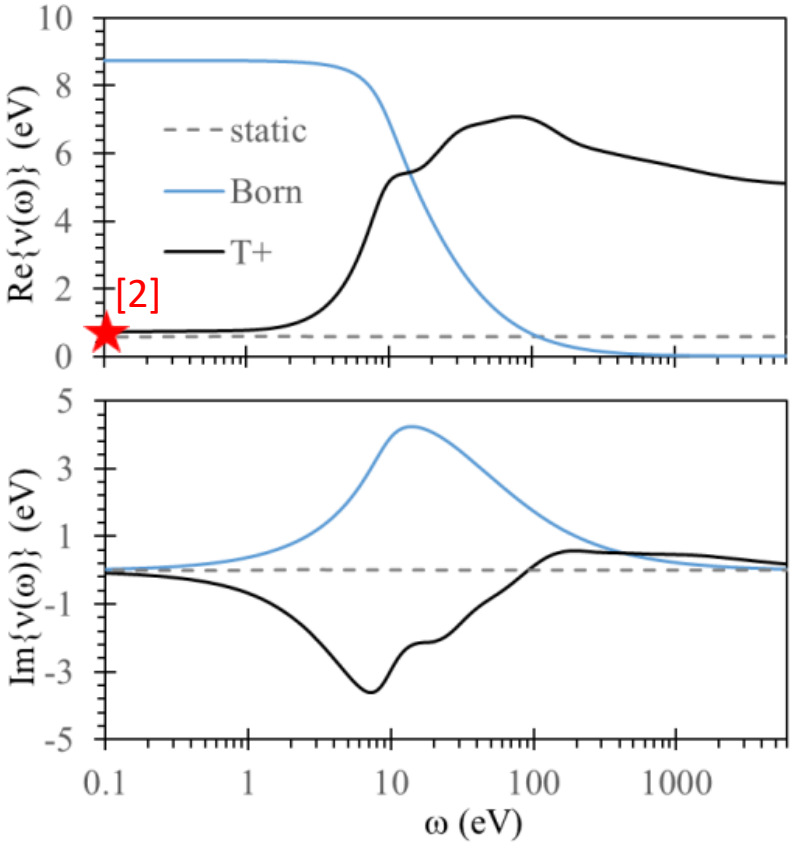
We will show that strong collisions and inelastic collisions improve DFT-AA agreement with TDDFT [1], static conductivities [2], and XRTS experiments [3]

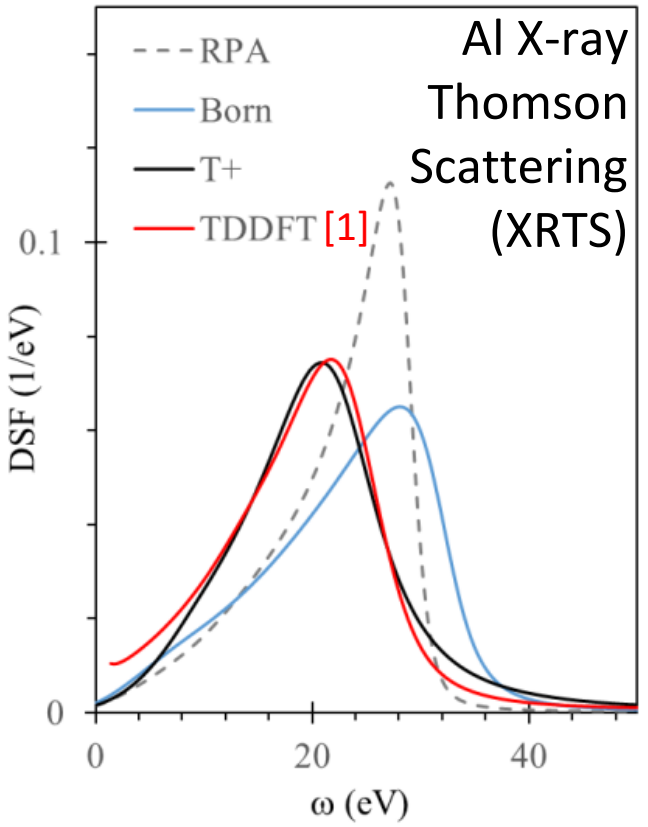
[1] Baczewski et al PRL 116, 115004 (2016); [2] Milchberg et al PRL 61 2364 (1988) ; [3] Sperling et al PRL 115, 115001 (2015)

The dynamic collision frequency modifies response functions and observables

- In a direct (zero-frequency) electric field, the static collision frequency $\nu(0)$ represents collisions that control DC conductivity $\sigma(0) = \frac{n_e}{\nu(0)}$
- In an oscillating field, the dynamic collision frequency $\nu(\omega)$ represents the damping of collective plasma oscillations with frequency ω
- The dynamic collision frequency informs the Mermin dielectric function $\epsilon^M(q,\omega)$ and observables like the dynamic structure factor (DSF) ~ $|\frac{1}{\epsilon(q,\omega)}|$

  → XRTS and stopping powers





The Born approximation for cross sections is commonly used in calculations of $v(\omega)$

$$v(\omega) \sim$$

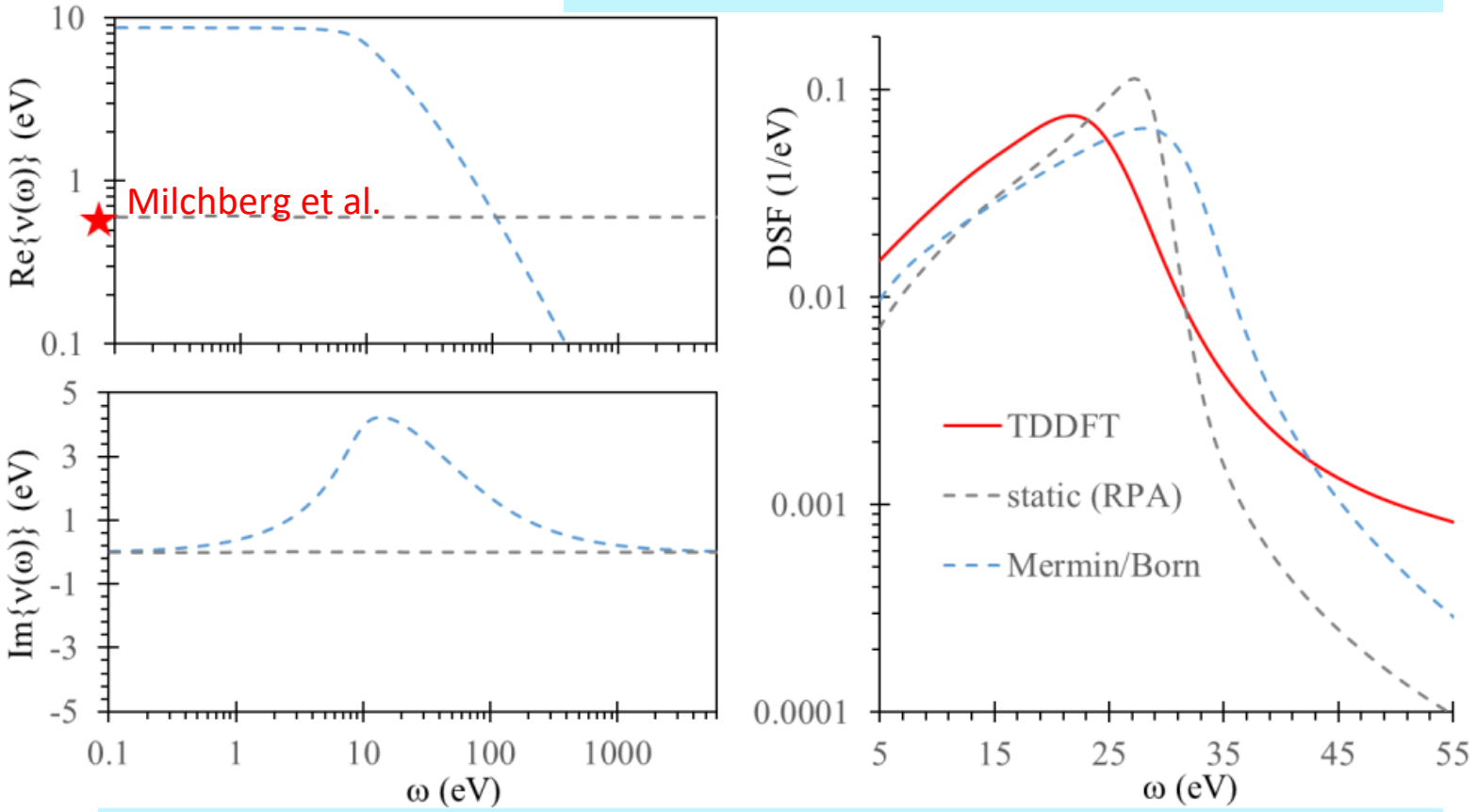
$$\int \frac{\epsilon(q,\omega) - \epsilon(q,0)}{\omega} q^6 S_{ii}(q) \frac{\partial \sigma^{tr}}{\partial \theta} dq$$

The Born cross section with a screened Yukawa potential is:

$$\frac{\partial \sigma^{tr}}{\partial \theta} = \frac{1}{4\pi^2} \left| \frac{4\pi Z^*}{q^2 + q_s^2} \right|^2$$

Whatever our choices for Z^* and q_s , the real and imaginary parts of $v(\omega)$ have a fixed functional form that moves $\epsilon^M(q,\omega)$ away from TDDFT

Example: Solid Al at T = 1eV

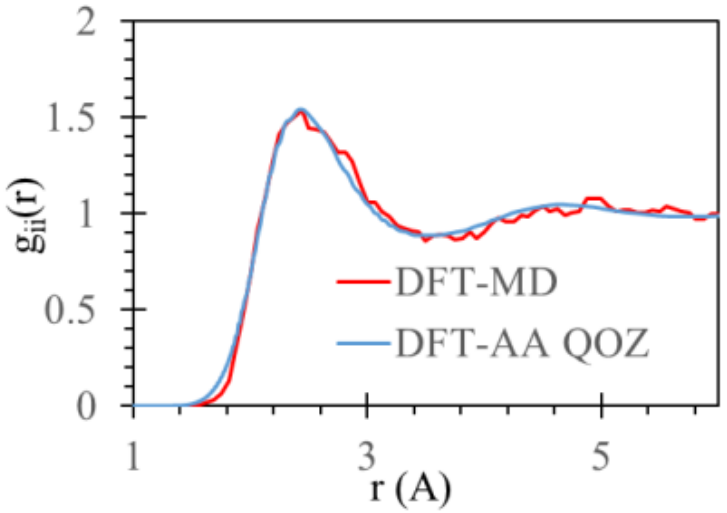


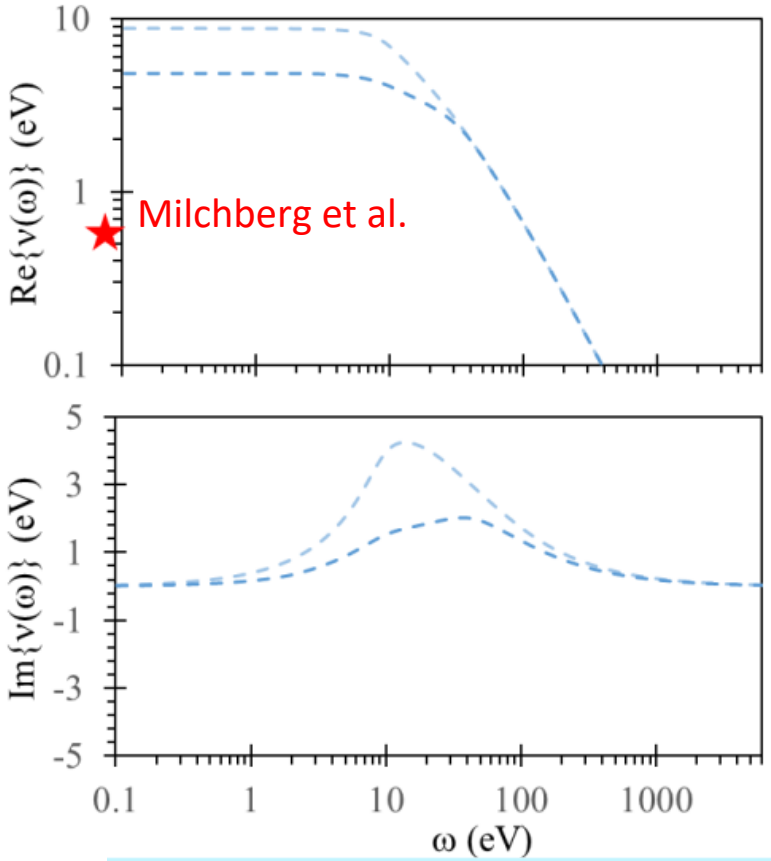
Weak scattering (Born) gives unreliable $v(0)$ and inevitable $\mathrm{Im}\{v(\omega)\} > 0$ shifts the RPA plasmon peak to higher energies

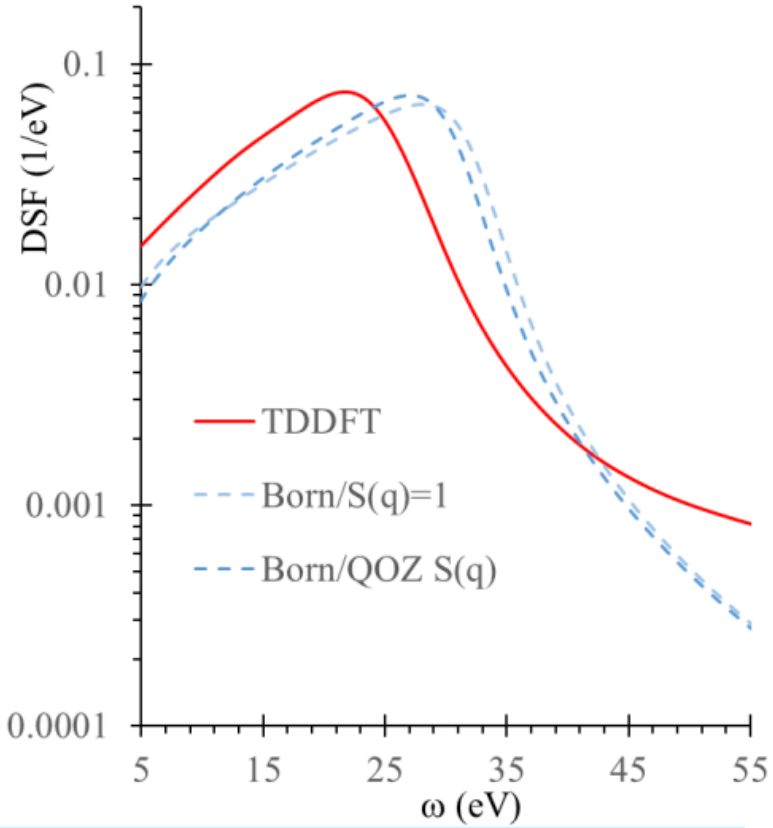
Modifying the ion-ion structure factor doesn't help much

$$v(\omega) \sim \int \frac{\epsilon(q,\omega) - \epsilon(q,0)}{\omega} q^6 S_{ii}(q) \frac{\partial \sigma^{tr}}{\partial \theta} dq$$

We can use a self-consistent $S_{ii}(q)$ from $g_{ii}(r)$ following Starrett & Saumon







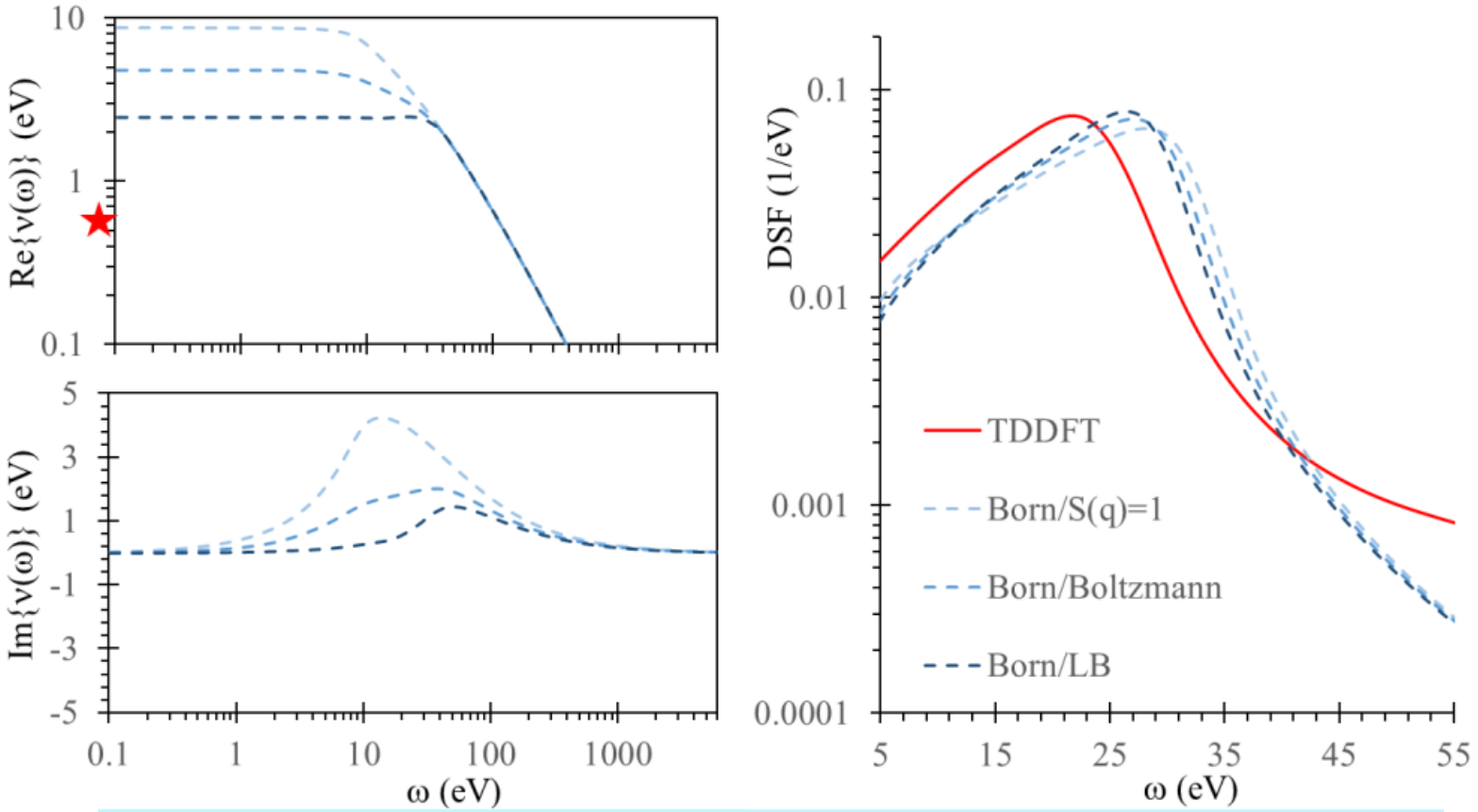
While we find good agreement with DFT-MD ionic structure, changing $S_{ii}(q)$ doesn't change the functional form of $v(\omega)$

Starrett & Saumon, HEDP 10, 35 (2014); *cf* Wetta and Pain, Phys Rev E 102, 053209 (2022)

Including dynamic screening via Lenard-Balescu integration [1,2] also has only a modest effect

$$v(\omega) \sim \int \frac{\epsilon^{-1}(q,\omega) - \epsilon^{-1}(q,0)}{\omega} q^6 S_{ii}(q) \frac{\partial \sigma^{tr}}{\partial \theta} dq$$

Replacing $\epsilon(q,\omega)$ with $\epsilon^{-1}(q,\omega)$ gives a better approximation for the collisional term in the generalized Boltzmann equation



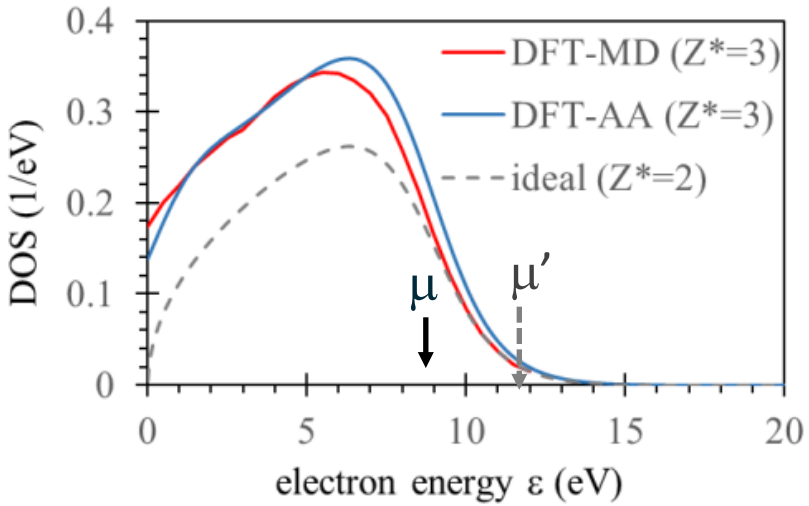
While this modification further decreases $v(0)$ and the plasmon shift, both remain far from trusted values

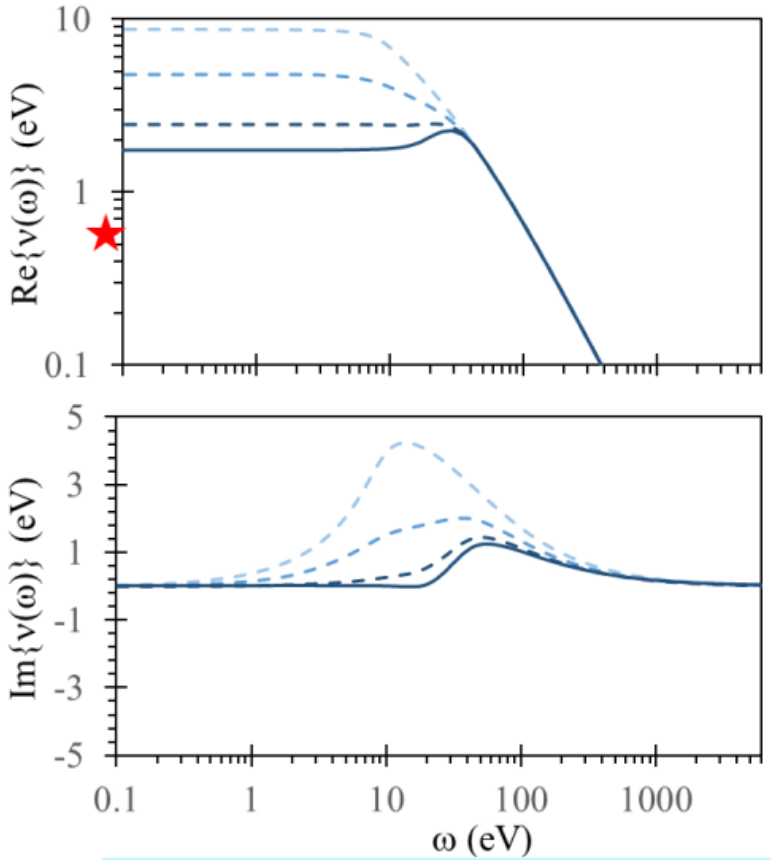
[1] Redmer, Physics Reports 282, 35 (1997) [2] Faussurier & Blancard, Physics of Plasmas 23, 012703 (2016)

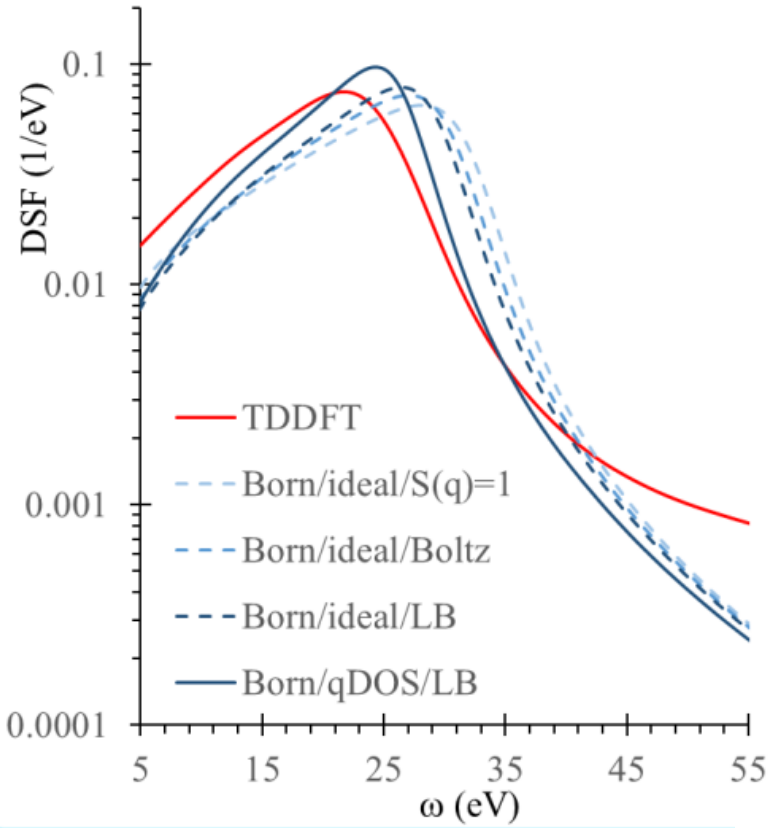
Using the quantum density of states gives a more reasonable DSF but still does not capture $v(0)$

$$v(\omega) \sim \int \frac{\epsilon_q^{-1}(q,\omega) - \epsilon_q^{-1}(q,0)}{\omega} q^6 S_{ii}(q) \frac{\partial \sigma^{tr}}{\partial \theta} dq$$

Using the quantum DOS modifies the integrals in $\epsilon(q,\omega)$ and makes Z* consistent with μ







The full quantum density of states better represents the continuum electron structure and eliminates ambiguity in Z*, but disagreement in $v(0)$ persists

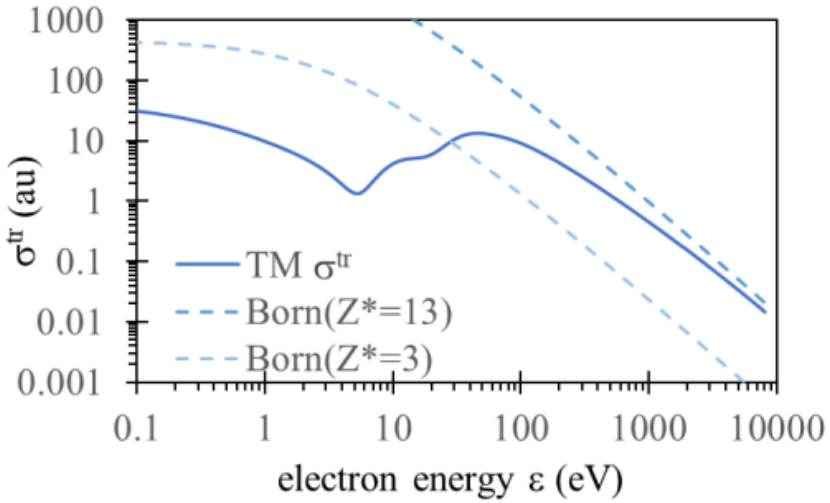
[1] Faussurier & Blancard, Physics of Plasmas 23, 012703 (2016)

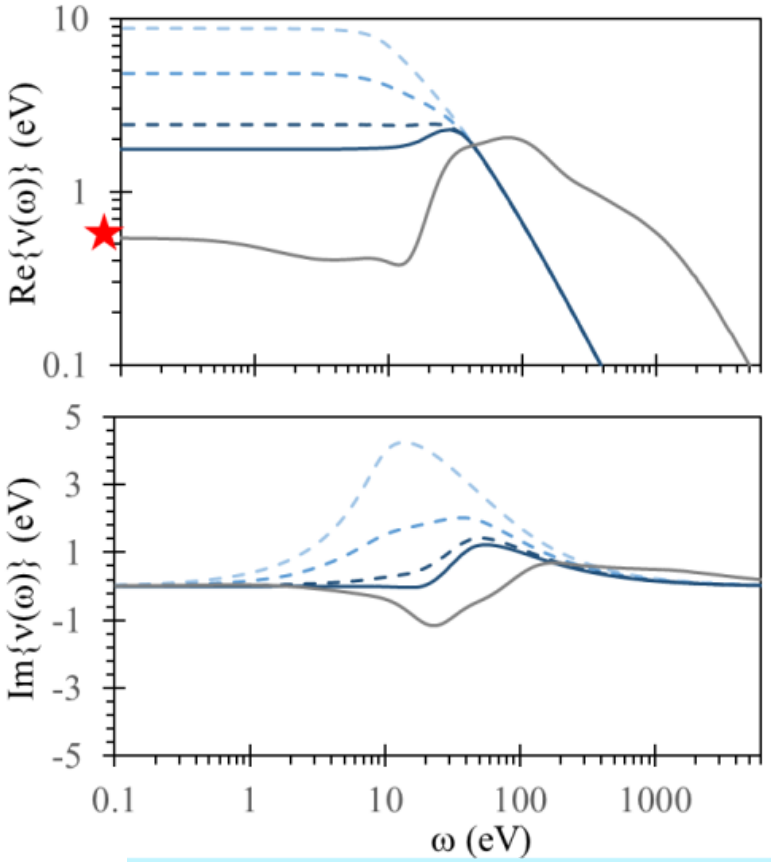
T-matrix cross sections capture strong collisions, significantly improving the $v(0)$ limit

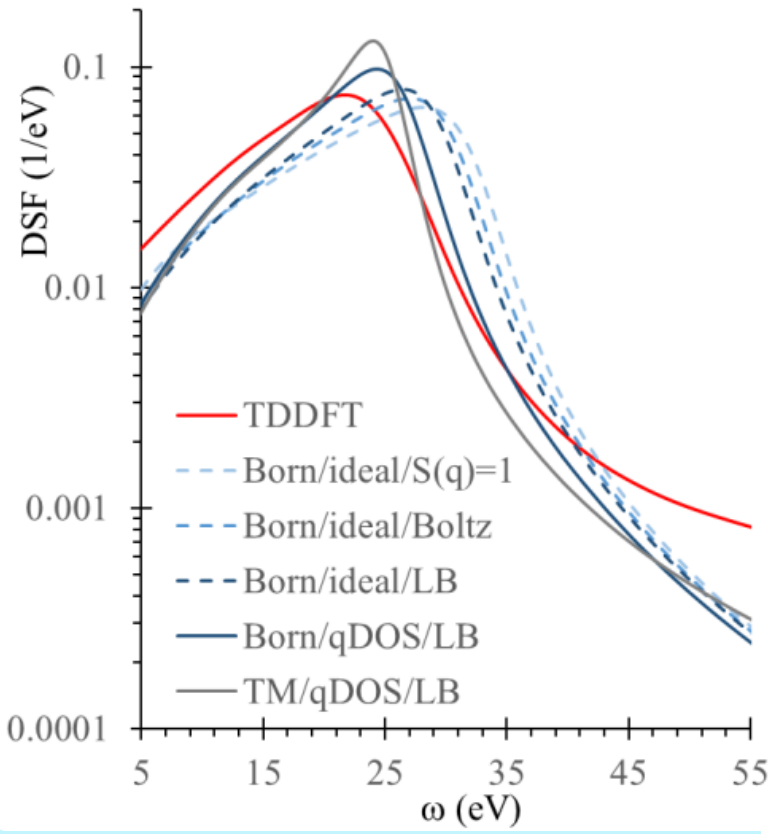
$$v(\omega) \sim$$

$$\int \frac{\epsilon(q,\omega) - \epsilon(q,0)}{\omega} q^6 S_{ii}(q) \frac{\partial \sigma^{tr}}{\partial \theta} dq$$

The T-matrix cross sections use phase shifts from the self-consistent continuum orbitals







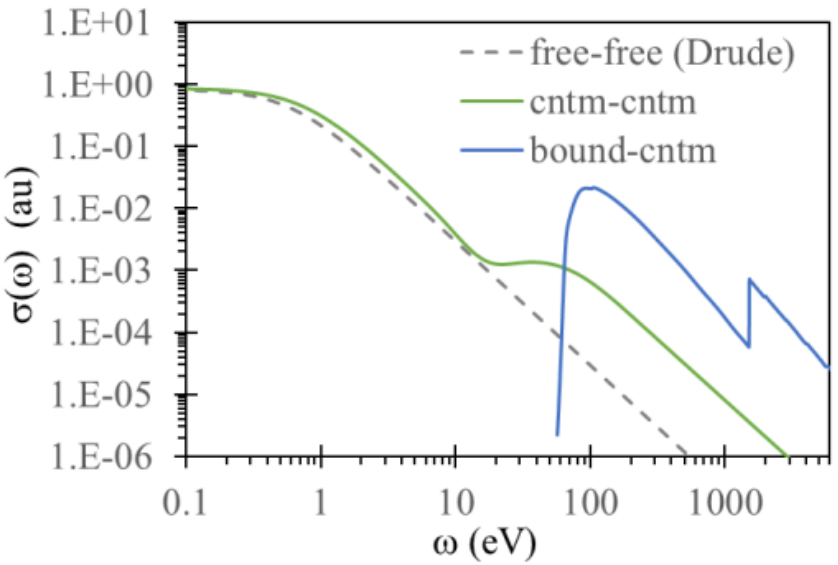
Strong scattering significantly improves $v(0)$ and departs significantly from the Born form, giving $\mathrm{Im}\{v(\omega)\} < 0$

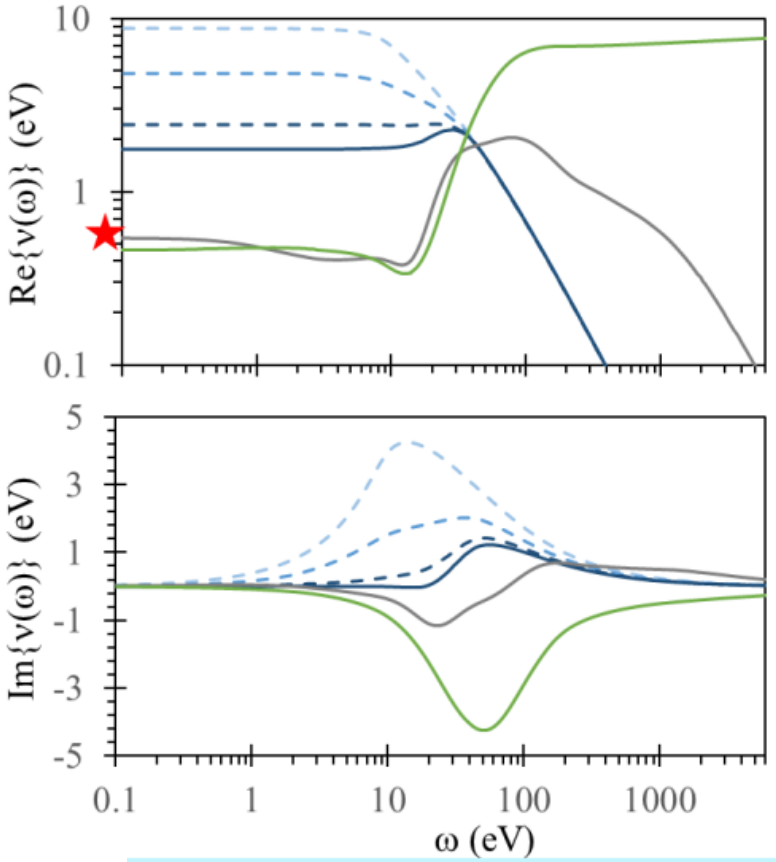
Independent Kubo-Greenwood calculations capture a similar $v(0)$ limit and frequency dependence

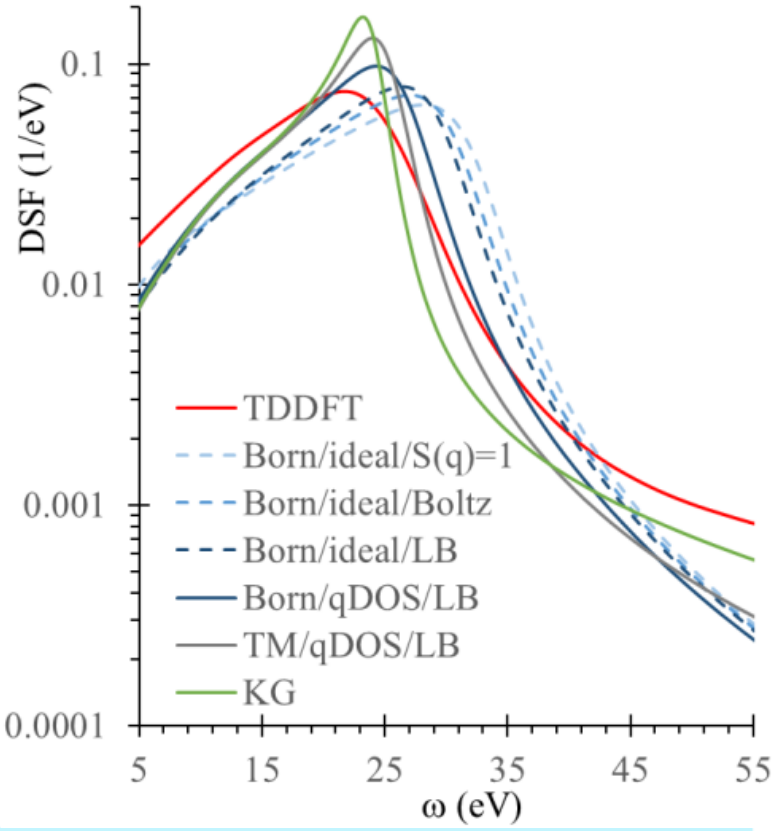
$$v(\omega) = \frac{n_e}{\sigma(\omega)} + i\omega$$

following Reinholz et al. [1]

The quantum Kubo-Greenwood dynamic conductivity $\sigma(\omega)$ adds a distinct feature to the Drude $\sigma(\omega)$







The K-G $v(0)$ limit enforces the conductivity sum rule; $\sigma(\omega)$ shows the non-Drude feature observed by Witte et al. [2] – but now the XRTS plasmon peak is too narrow

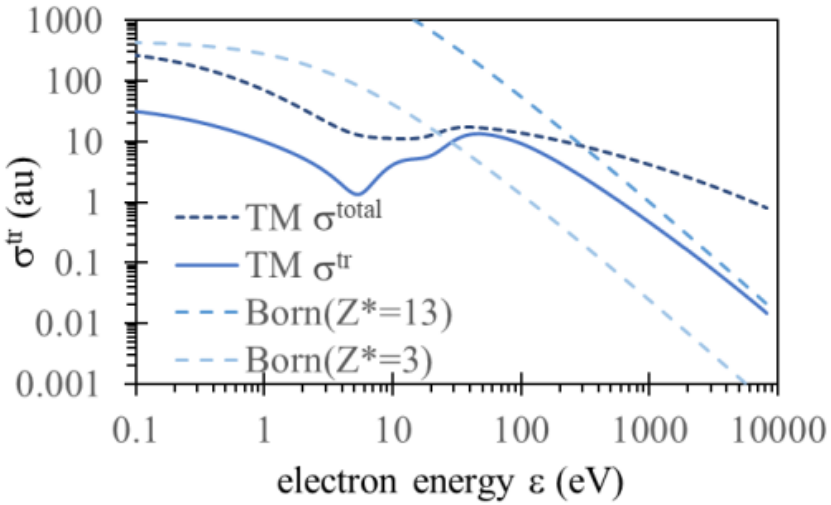
[1] Reinholz et al, Phys Rev E 62, 5648 (2000) [2] Witte et al., Physics of Plasmas 25, 056901 (2018)

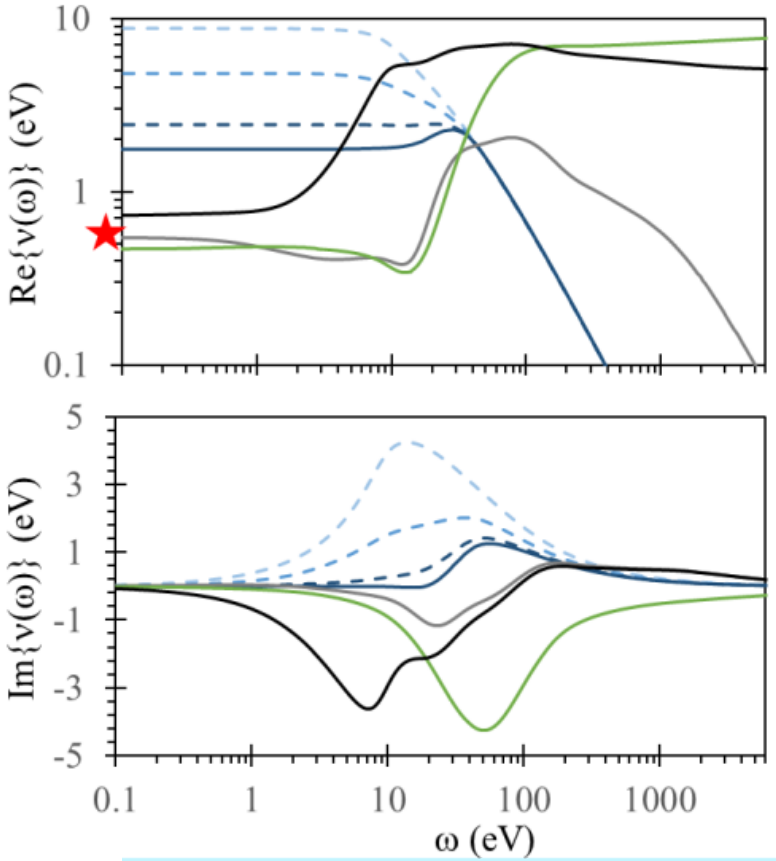
A trial estimate of inelastic processes brings our DFT-AA calculations into good agreement with reference data

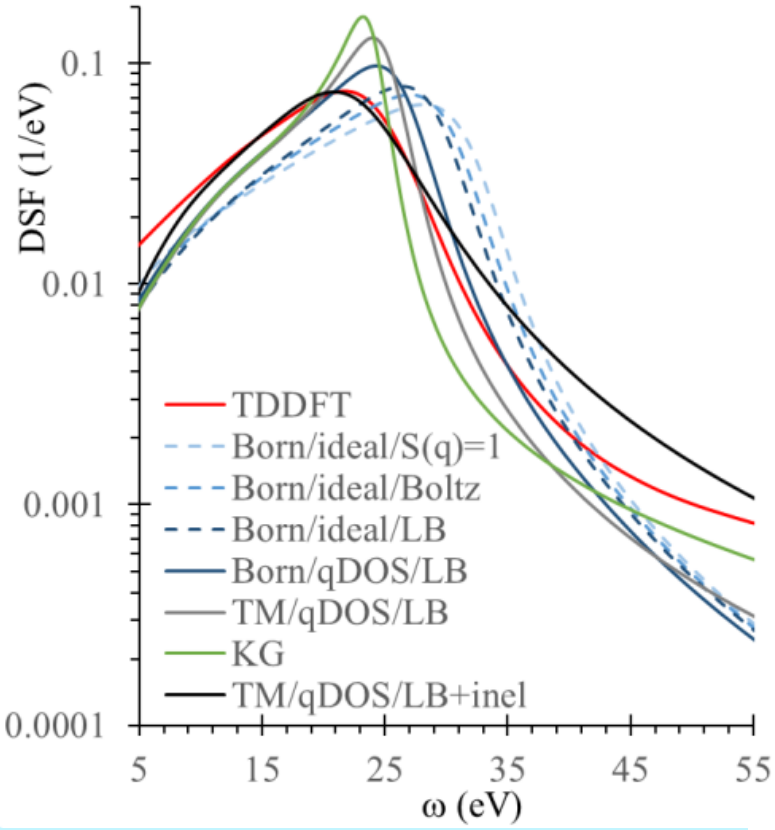
$$v^{inel}(\omega) \sim n_e \iint (\sigma^{tot} - \sigma^{tr})\, d\varepsilon_1 d\varepsilon_2$$

$$\times\ e^{(\mu-\varepsilon_f)/\tau}$$

Inelastic collisions include Pauli blocking and represent the relaxation time associated with a given excitation from equilibrium







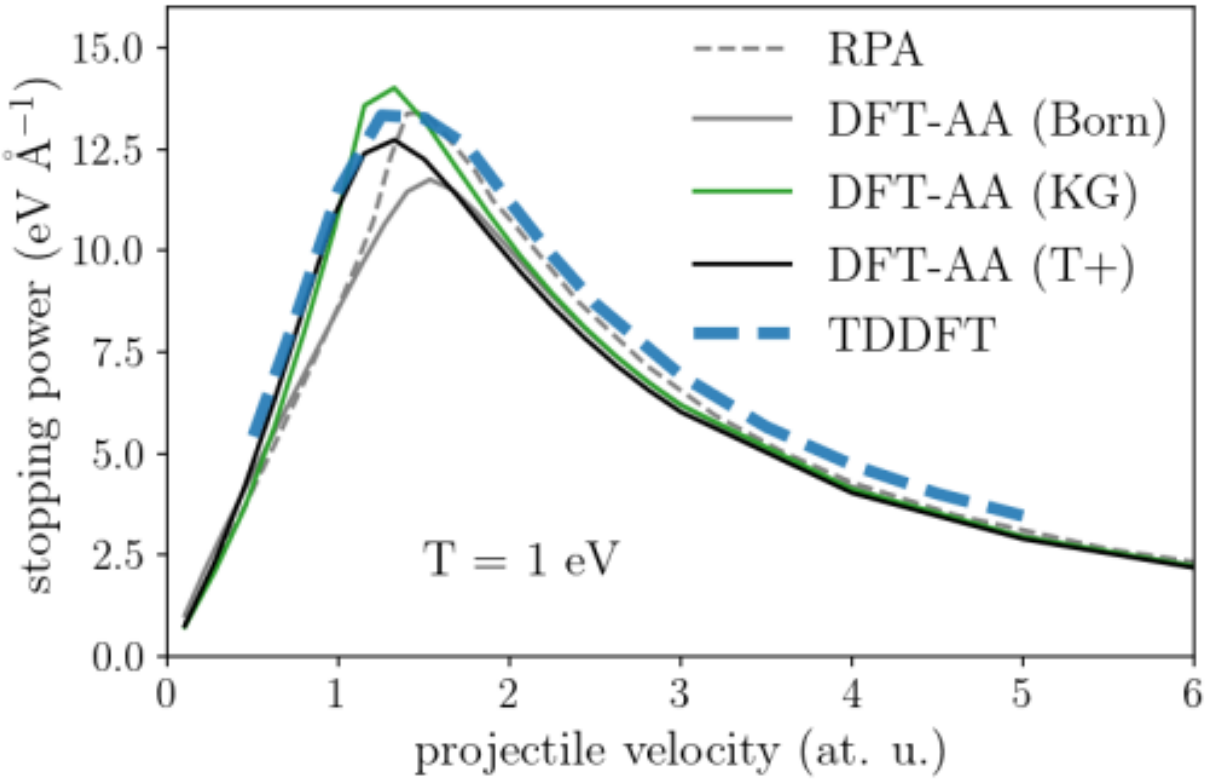
The impact of inelastic processes on the $v(0)$ limit and σ^{DC} will be investigated in future work

Papers in preparation: Baczewski et al, Hentschel et al

Our dynamic collision frequencies also improve DFT-AA stopping powers

$$\frac{dE}{dx} \sim \iint \left| \frac{1}{\epsilon(q,\omega)} \right| \omega \, d\omega \, dk$$

Modifications to the dielectric function impact charged-particle stopping powers.



Using $v(\omega)$ from both Kubo-Greenwood and T-matrix + inelastic collisions significantly improve low-velocity dE/dx

Papers in preparation: Kononov at al, Hentschel et al

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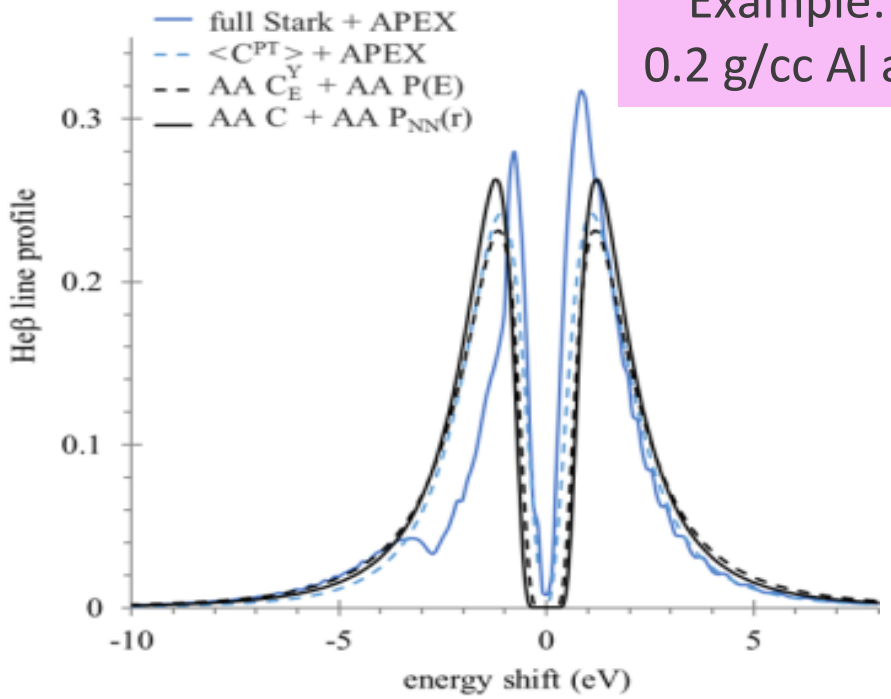
Example: Heβ of 0.2 g/cc Al at 250 eV



We have shown how DFT-AA can provide reasonable estimates for ion-Stark broadening
(see K. Adler's presentation)
With T. Gomez and C Iglesias, we are exploring whether dynamic collision frequencies with inelastic collisions can provide electron broadening terms

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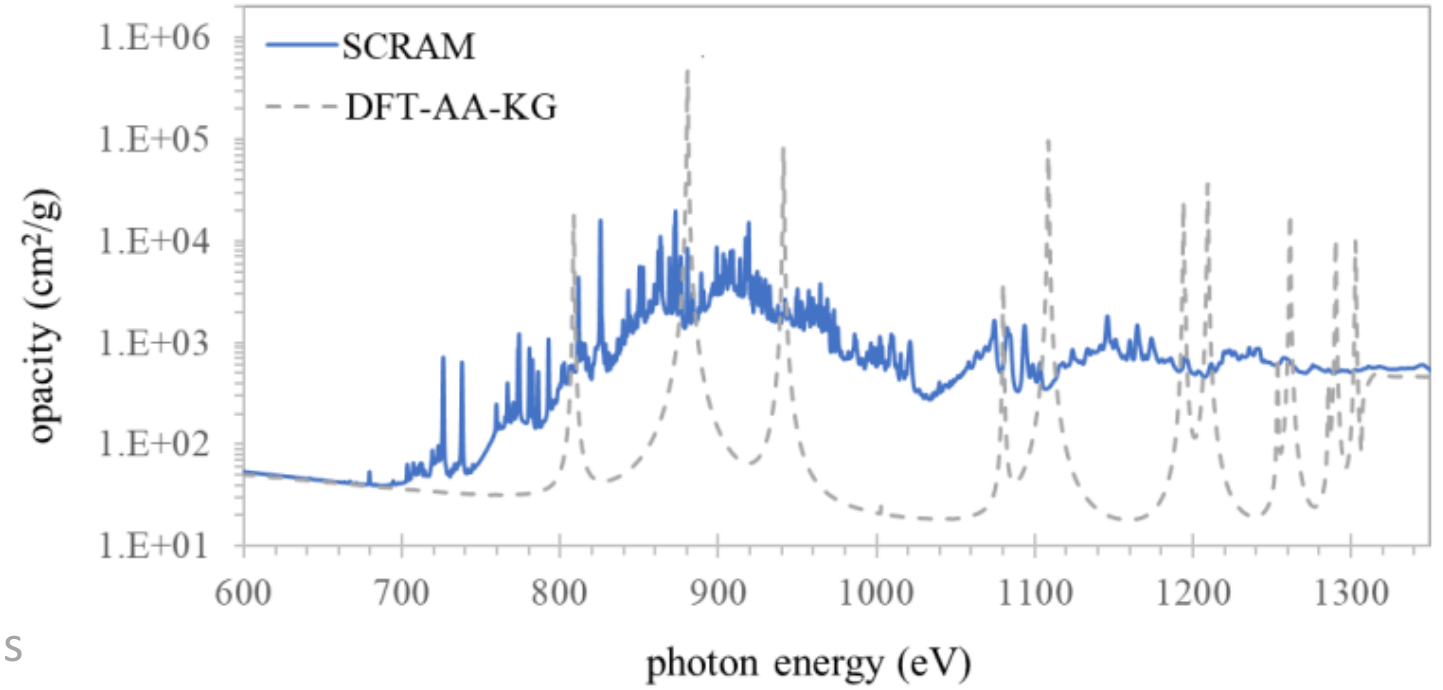
Example: 0.2 g/cc Fe at 180 eV

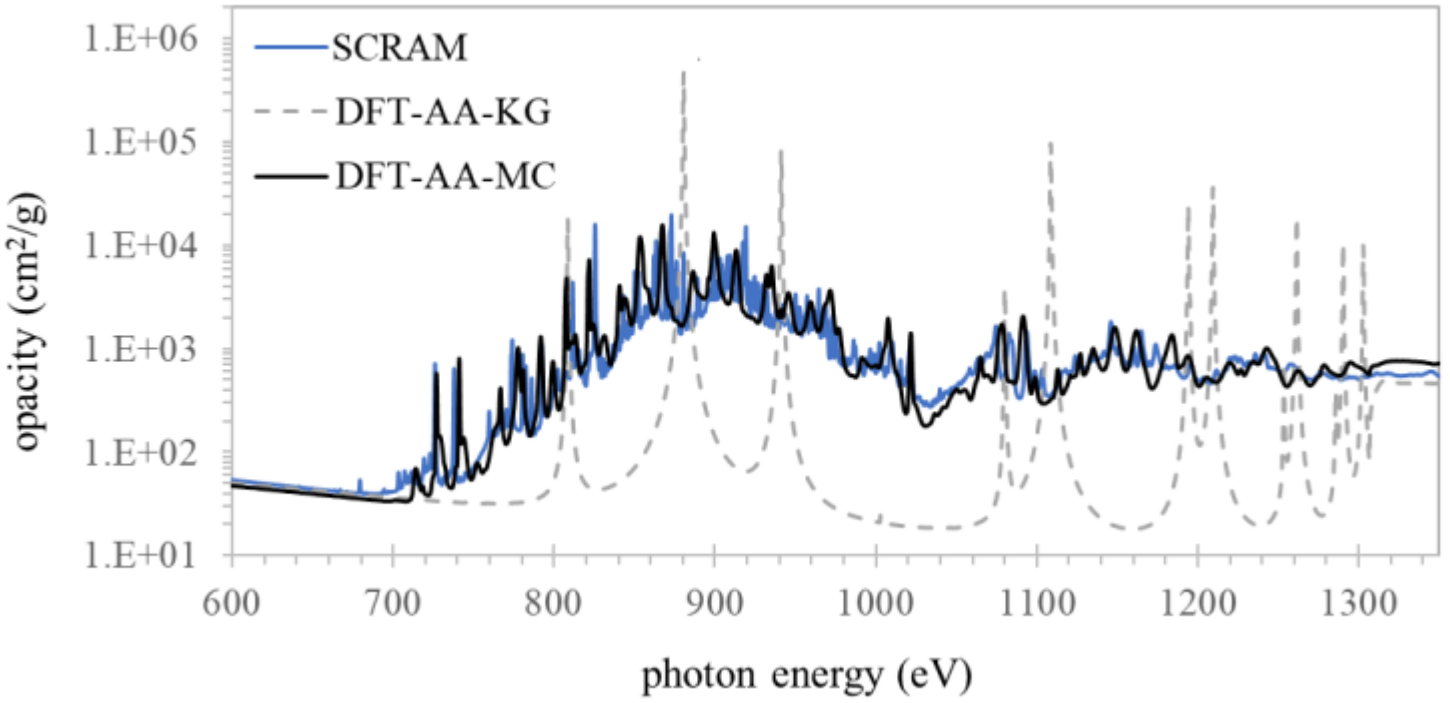


Good line shapes are only useful when combined with detailed multiconfiguration electronic structure and spectra

cf Bailey et al, Nature (2015)

We have developed an efficient approach to generate multiconfiguration atomic structure

- Using DFT-AA electronic $n\ell$ orbitals as a basis set, we compute Slater coefficients to generate real (integer-occupied) electronic configurations [1]
- Re-optimize all DFT-AA $n'\ell'$ orbitals under small changes in occupations in all $n\ell$ orbitals → new Slater coefficients
- Taylor-expand Slater coefficients on occupations to capture orbital relaxation
- Add spin-orbit effects to split $n\ell$ transitions into $n\ell j$



This approach generates very reasonable electronic structure and spectra without re-optimizing orbitals for every configuration

[1] Faussurier and Blancard, Phys Rev E 97, 023206 (2018); Paper in preparation: Hansen et al

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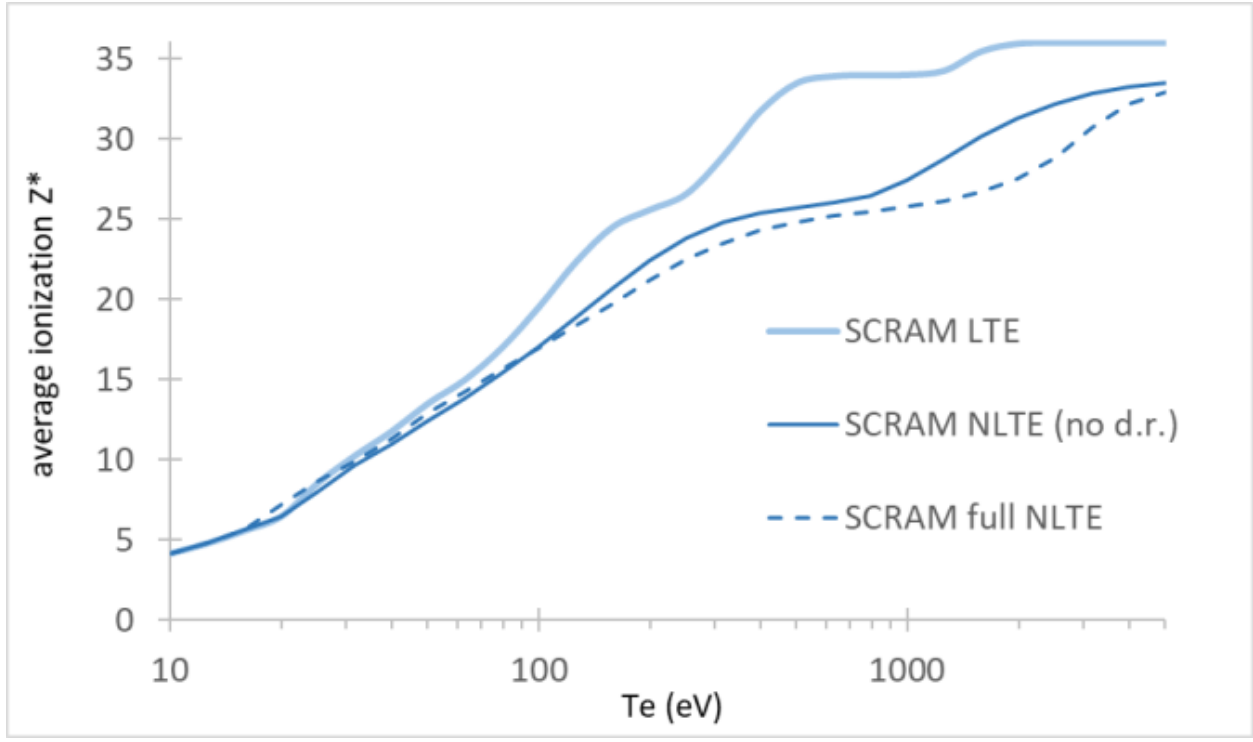
5) **Extend DFT- AA to non-equilibrium plasmas Tr << Te**

Example: 0.01 g/cc Kr



Non-LTE effects have profound impacts on charge state distributions, radiative losses, and emission and absorption spectra, especially for high Z and low densities

We have developed a simple modification to Fermi-Dirac occupation factors for a reasonable non-LTE Z*

Standard DFT-AA occupations:

$X_{n\ell} = 2(2\ell+1) / [1 + e^{(En\ell - m)/T}]$

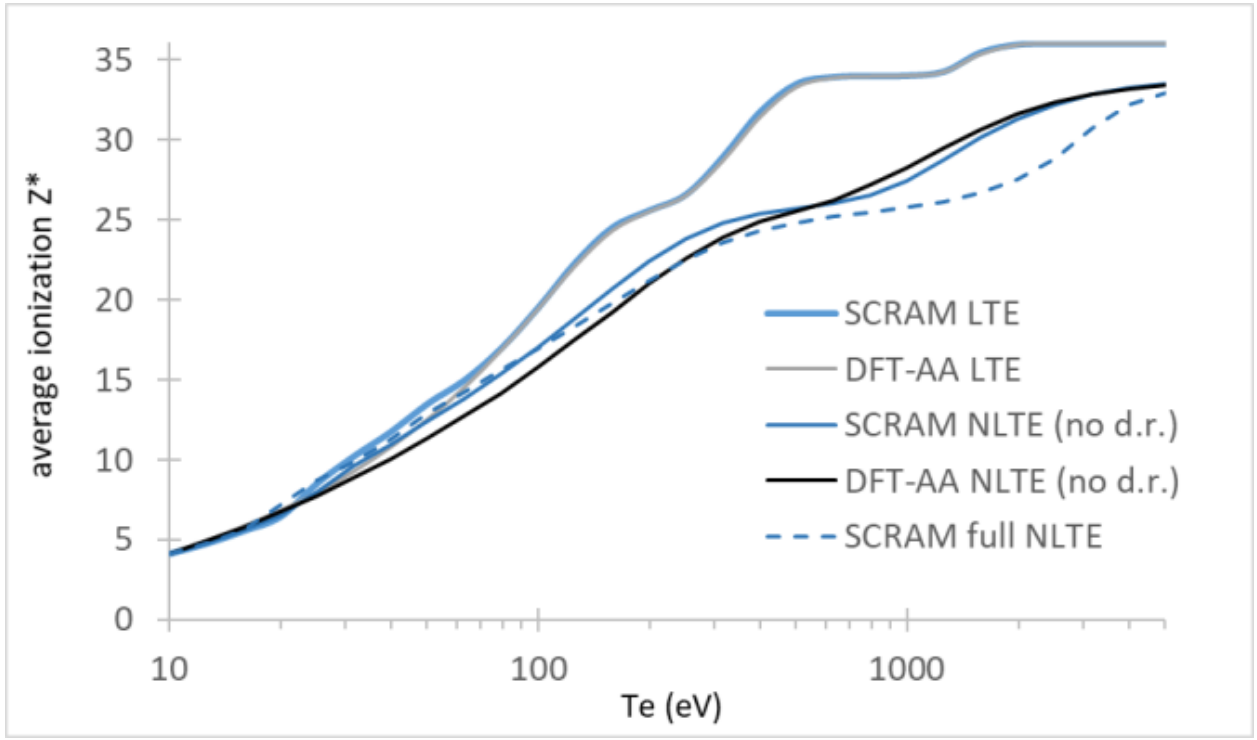
New DFT-AA occupations use collisional and photoionization (and recombination) rates $R_{n\ell \leftrightarrow cntm}$ that modify the occupations:

$X_{n\ell} = 2(2\ell+1) / [1 + (R^{ci} + R^{pi}) / (R^{cr} + R^{rr} + R^{dr})]$

→ Recovers LTE

→ Provides reasonable non-LTE Z*

→ Dielectronic recombination is tricky

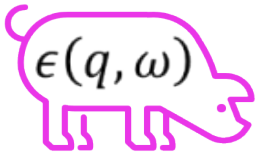


This simple modification gives a first approximation for Z* that is good enough to provide self-consistent orbitals suitable for full non-LTE collisional-radiative calculations

cf Faussurier et al, Physics of Plasmas 21, 112707 (2014); Paper in preparation: Hansen et al

Conclusion: Extensions of the DFT-AA model can produce reliable constitutive & observable properties

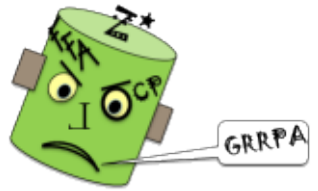
- Dynamic collision frequencies extend DFT-AA model predictions to include average plasma response as well as averaged ionic & electronic structure
  - → Conductivities
  - → X-ray Thomson Scattering (Hentschel, Baczewski)
  - → Stopping powers (Kononov, Hentschel)



Homer's magical animal

- We can split the DFT-AA electronic structure into real configurations and incorporate non-LTE effects
  - → Detailed spectra from self-consistent orbital basis set
  - → Self-consistent ion-Stark line shapes (Adler, Gomez)

All quantities come from a single, internally self-consistent DFT-AA model that agrees with available DFT-MD, TDDFT, CR, & line shape models



Rest well, Frankenmodels!

- Future: configuration-specific $\epsilon(q,\omega)$
  - → when can we average?
  - → Self-consistent dynamic electron-collisional broadening