SAND2022-2416C

Bound-bound features in x-ray Thomson scattering signals (arXiv:2109.09576)

Andrew Baczewski
Sandia National Laboratories

Thomas Hentschel
Cornell University

Alina Kononov
Sandia National Laboratories

Stephanie Hansen
Sandia National Laboratories

Sandia National Laboratories

APS March Meeting
2022-03-16

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology & Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.

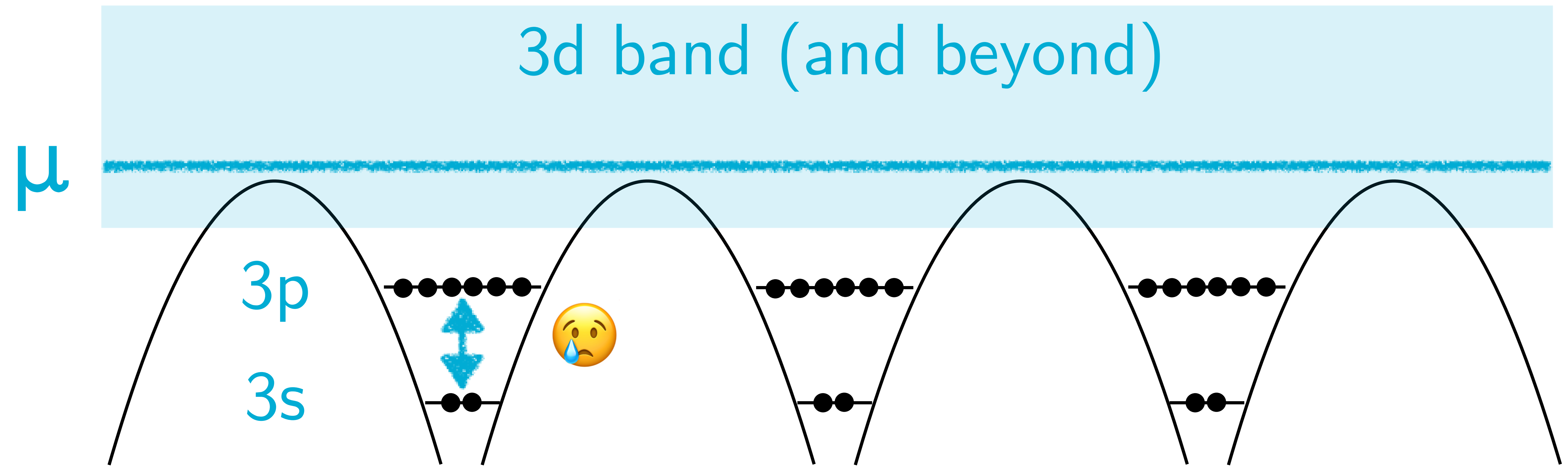
CCR
Center for Computing Research

Bound-bound transitions in warm dense matter

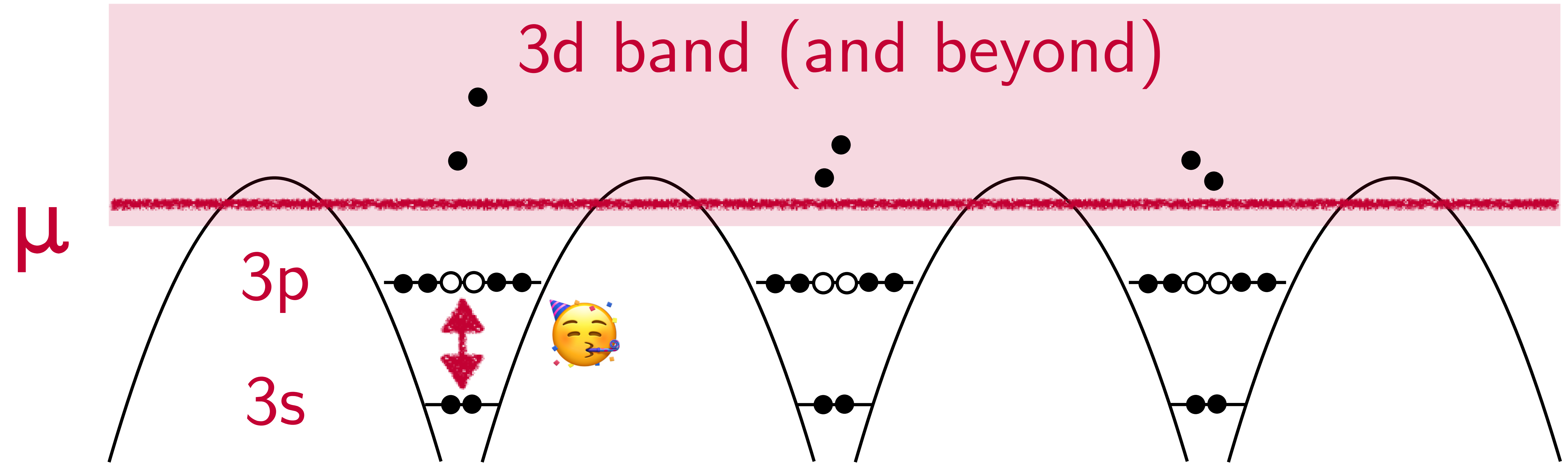
The bulk properties of degenerate matter are defined by the Pauli exclusion principle.

Exactly one electron is allowed to have the quantum numbers that it has...

Thermal excitation means that certain electronic rearrangements that would be forbidden are now allowed...



Solid density iron (8 g/cc)



Today, I'll show you how we're modeling these processes as they should appear in scattering experiments, using time-dependent DFT and average atom.

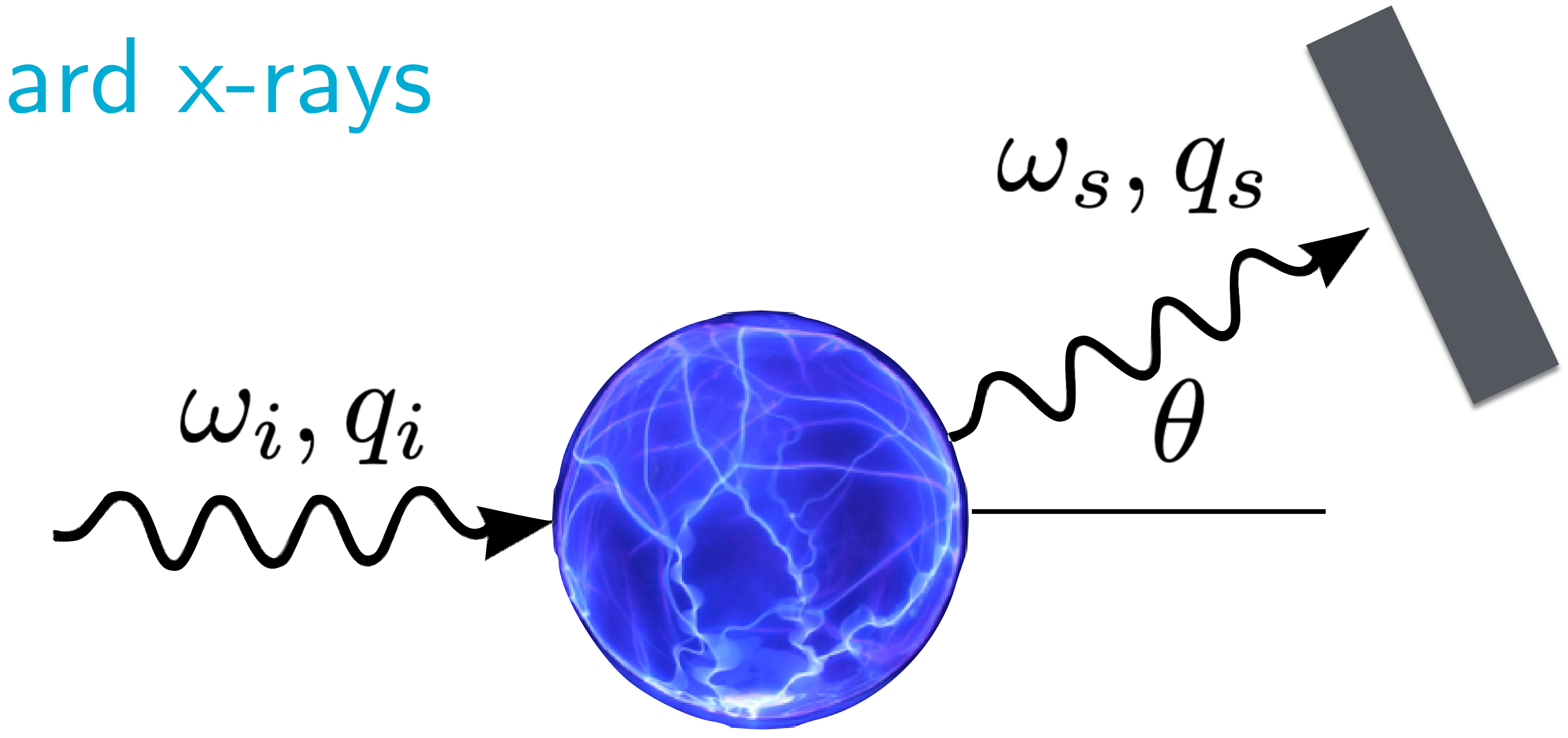
Basics of x-ray Thomson scattering (XRTS)

X-ray Thomson scattering in high energy density plasmas

Siegfried H. Glenzer and Ronald Redmer
Rev. Mod. Phys. **81**, 1625 – Published 1 December 2009

Measure inelastically scattered x-rays

Penetrate with hard x-rays



$$\omega = \omega_i - \omega_s$$
$$q = q_i - q_s$$

$$q = 2q_i \sin(\theta/2)$$

Sample of WDM
(opaque to optical probes)

$$\frac{d^2\sigma}{d\Omega d\omega} = \sigma_T \frac{q_s}{q_i} S(q,\omega)$$

Cross section proportional to dynamic structure factor (DSF)

Contains information about density, ionization state, structure, temperature, etc...

Dynamic structure factor in TDDFT

X-ray Thomson Scattering in Warm Dense Matter without the Chihara Decomposition

A. D. Baczewski, L. Shulenburger, M. P. Desjarlais, S. B. Hansen, and R. J. Magyar
Phys. Rev. Lett. **116**, 115004 – Published 18 March 2016

Probe system with x-ray*

$$v_{pert}(\mathbf{r}, t) = v_0 e^{i\mathbf{q}\cdot\mathbf{r}} f(t)$$

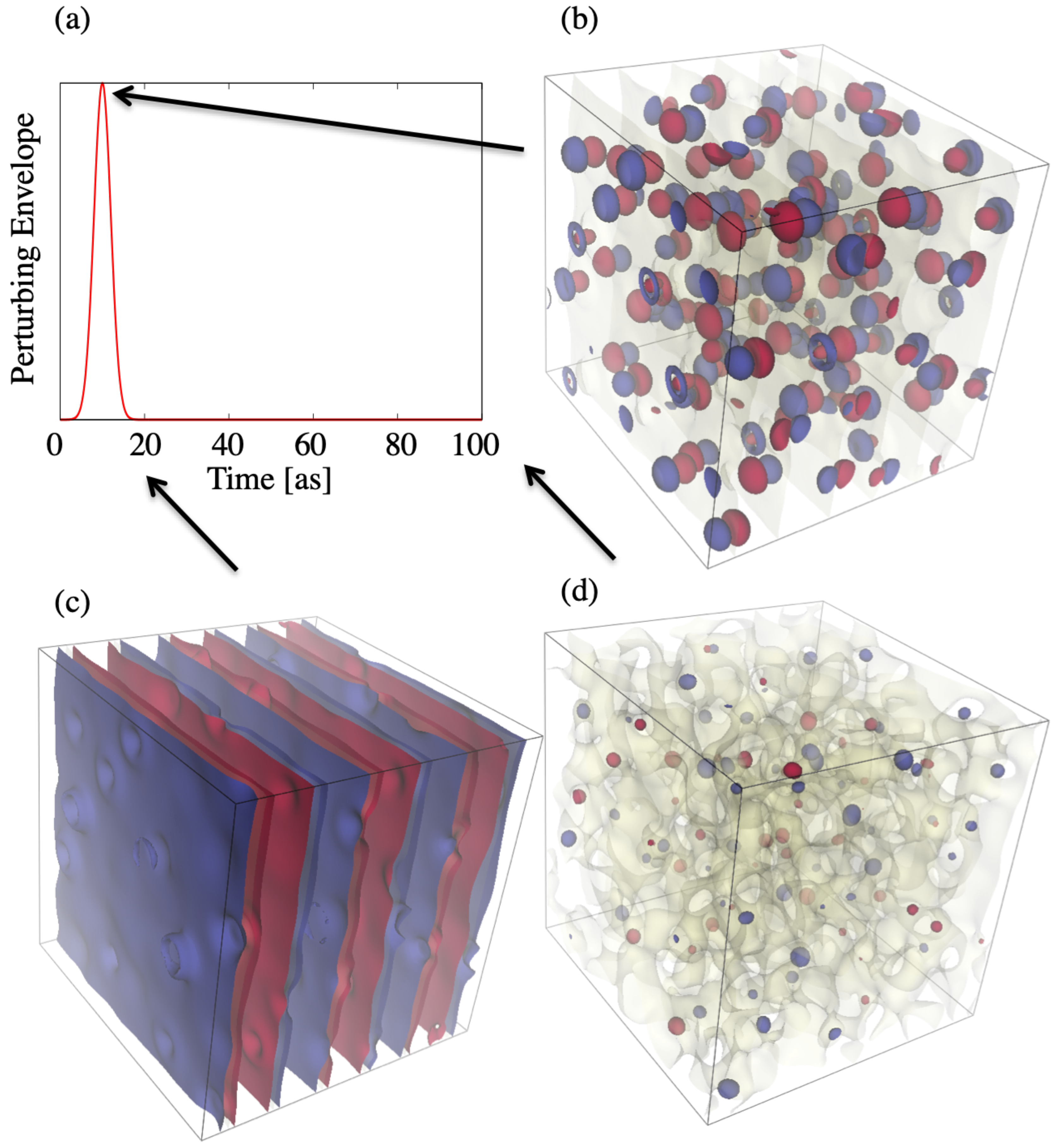
Record density response

$$\delta\rho(\mathbf{q}, t) = \int_0^\infty d\tau \chi_{\rho\rho}(\mathbf{q}, -\mathbf{q}, \tau) v_0 f(t-\tau)$$

Apply fluctuation-dissipation

$$\chi_{\rho\rho}(\mathbf{q}, -\mathbf{q}, \omega) = \frac{\delta\rho(\mathbf{q}, \omega)}{v_0 f(\omega)}$$

$$S(\mathbf{q}, \omega) = -\frac{1}{\pi} \frac{\mathrm{Im}\left[\chi_{\rho\rho}(\mathbf{q}, -\mathbf{q}, \omega)\right]}{1 - e^{-\omega/k_B T_e}}$$

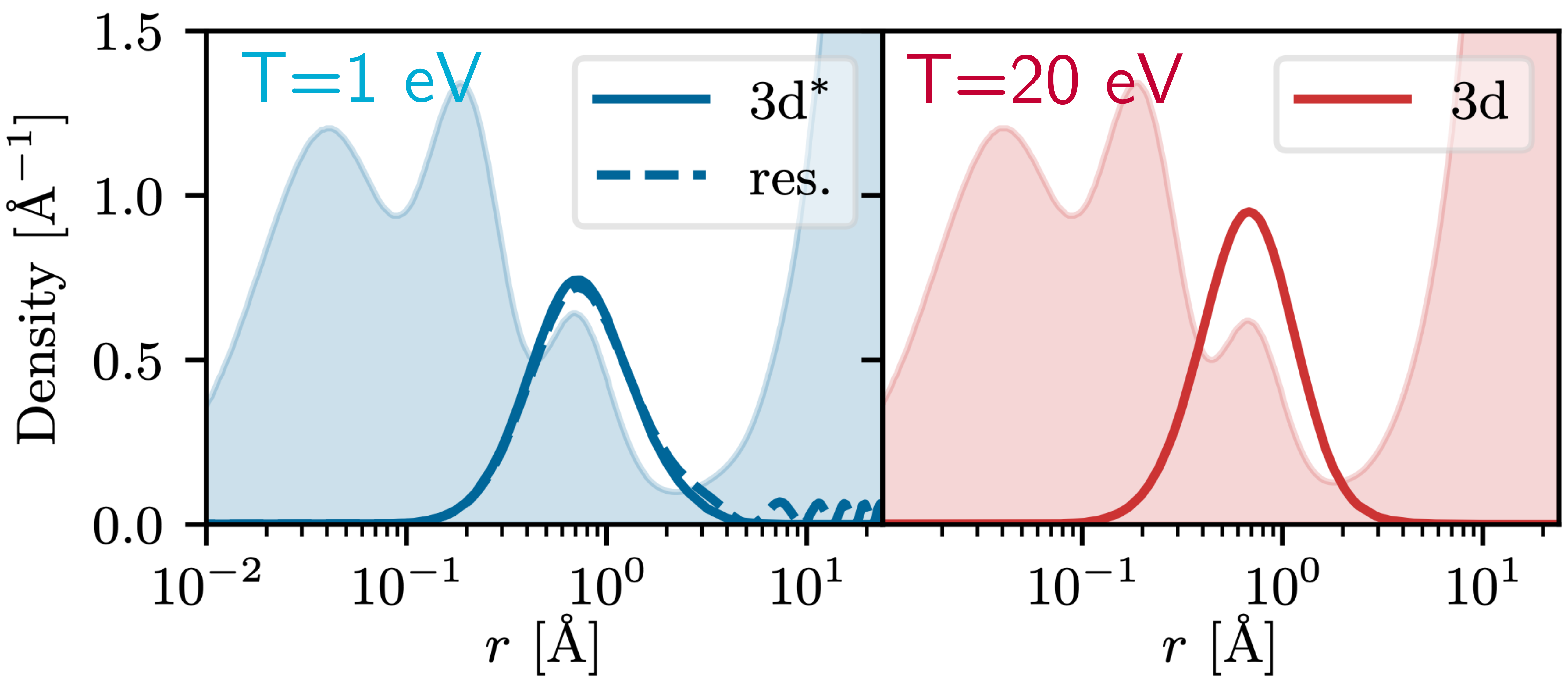


*energy/wave vector set by energy/momentum transfer of interest

Revising average atom theory

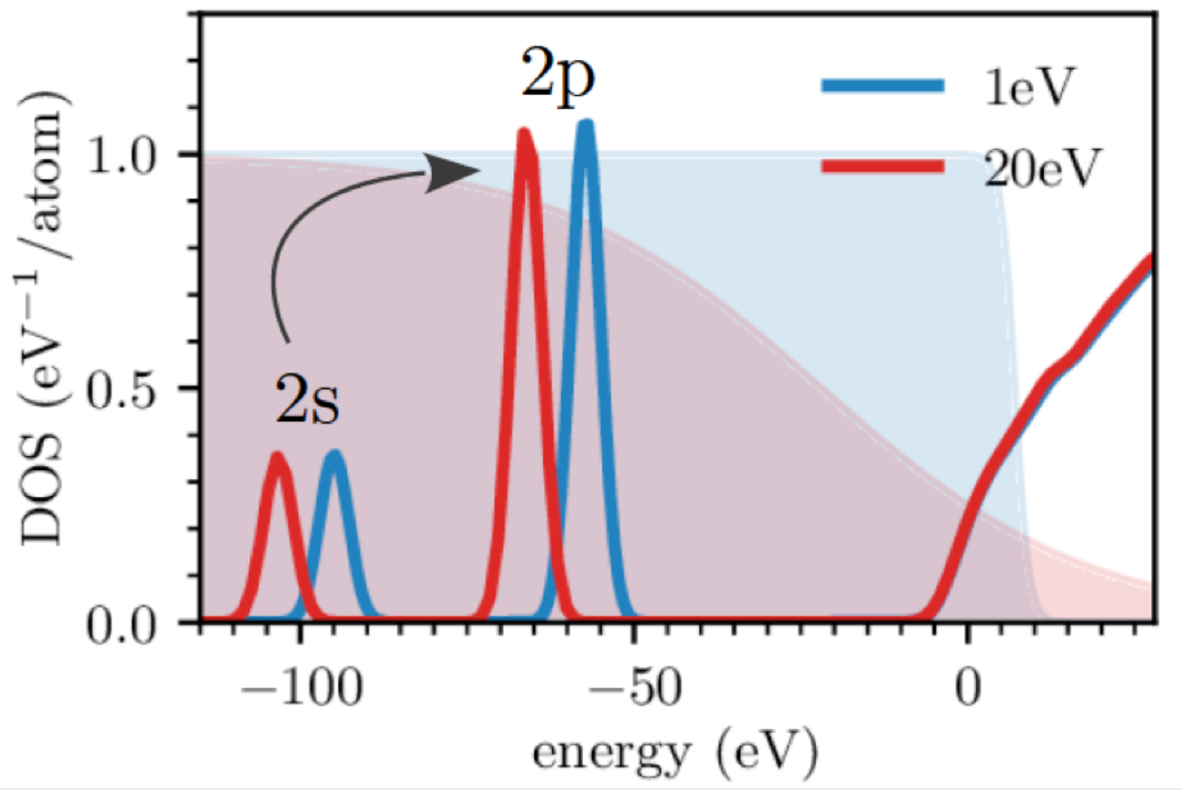
We've made 3 significant revisions to work in
[Starrett + Saumon, HEDP, 2014] and [Souza, et al. PRE, 2014]

1. The treatment of quasi-bound states, just above the chemical potential.
   Additional partition to ensure continuity of observables, going from bound to free (see below).
2. The addition of a bound-bound term to the Chihara decomposition.
   Rigorously justified, based on standard treatment in opacity.
3. An improved treatment of electron-ion collisions in the free-free term, using the Mermin formalism.
   Vastly improves treatment of plasmon, in general.



Example 1: Aluminum ("good" free-electron metal)

Thermal depletion of 2p states starts to appear above 10 eV [Witte, et al., PoP, 2018]

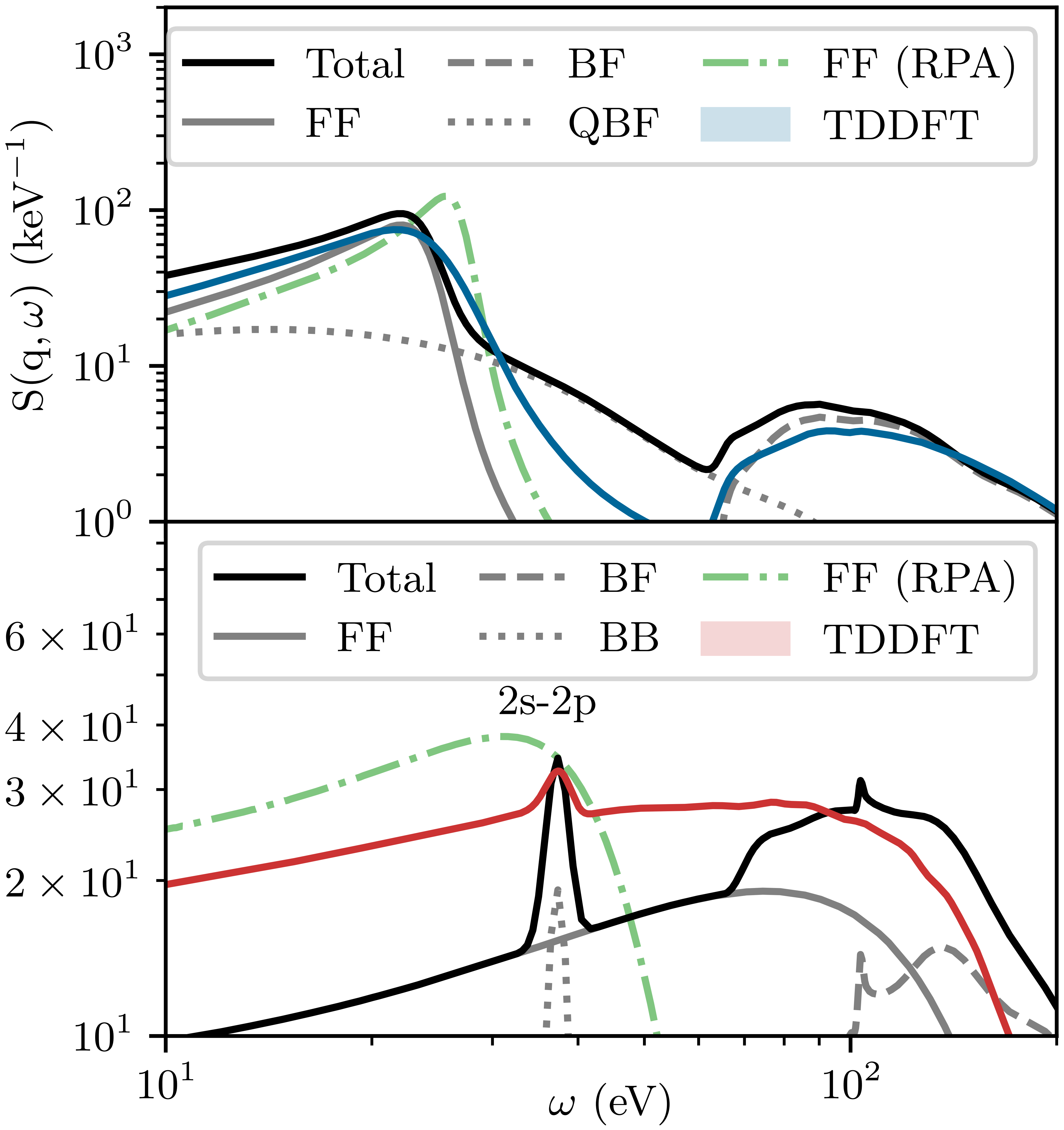


Results highlight the importance of revised Mermin treatment of the free-free contribution over RPA.

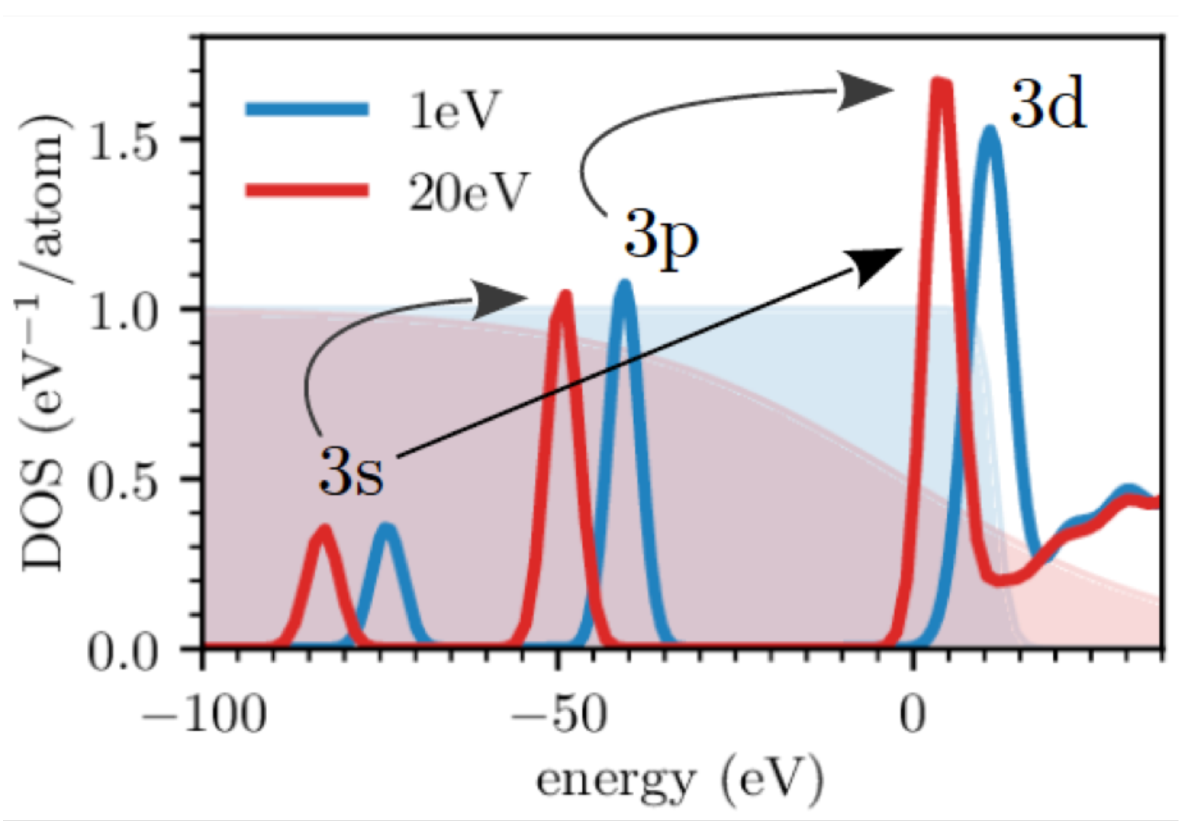
A 2s-2p bound-bound transition appears coincident with thermal depletion of 2p.

Low intensity, but if you see such a feature you have a smoking gun that you're above ~10 eV.

Bound-bound is weak, but this illustrates a significant improvement in AA collisional models.



Example 2: Iron (d-band near chemical potential)



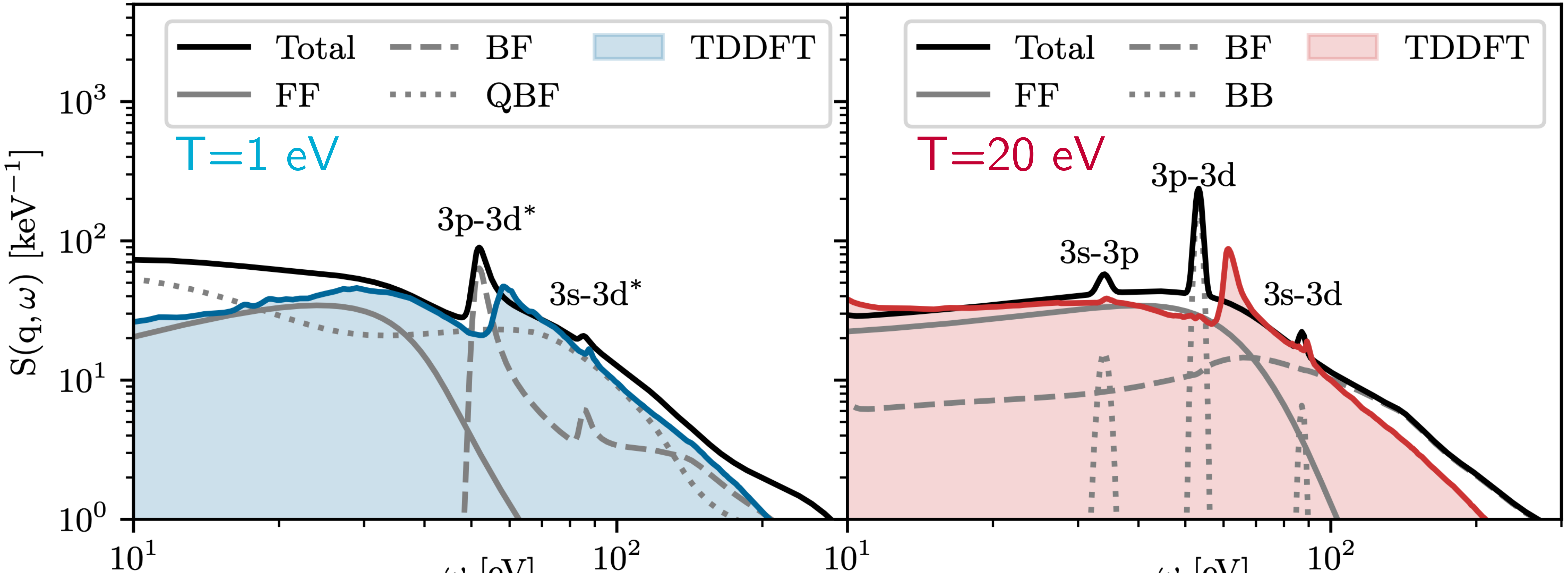
The spectrum of bound-bound transitions is richer yet in iron.

3p->3d @ 55 eV

3s->3d @ 85 eV

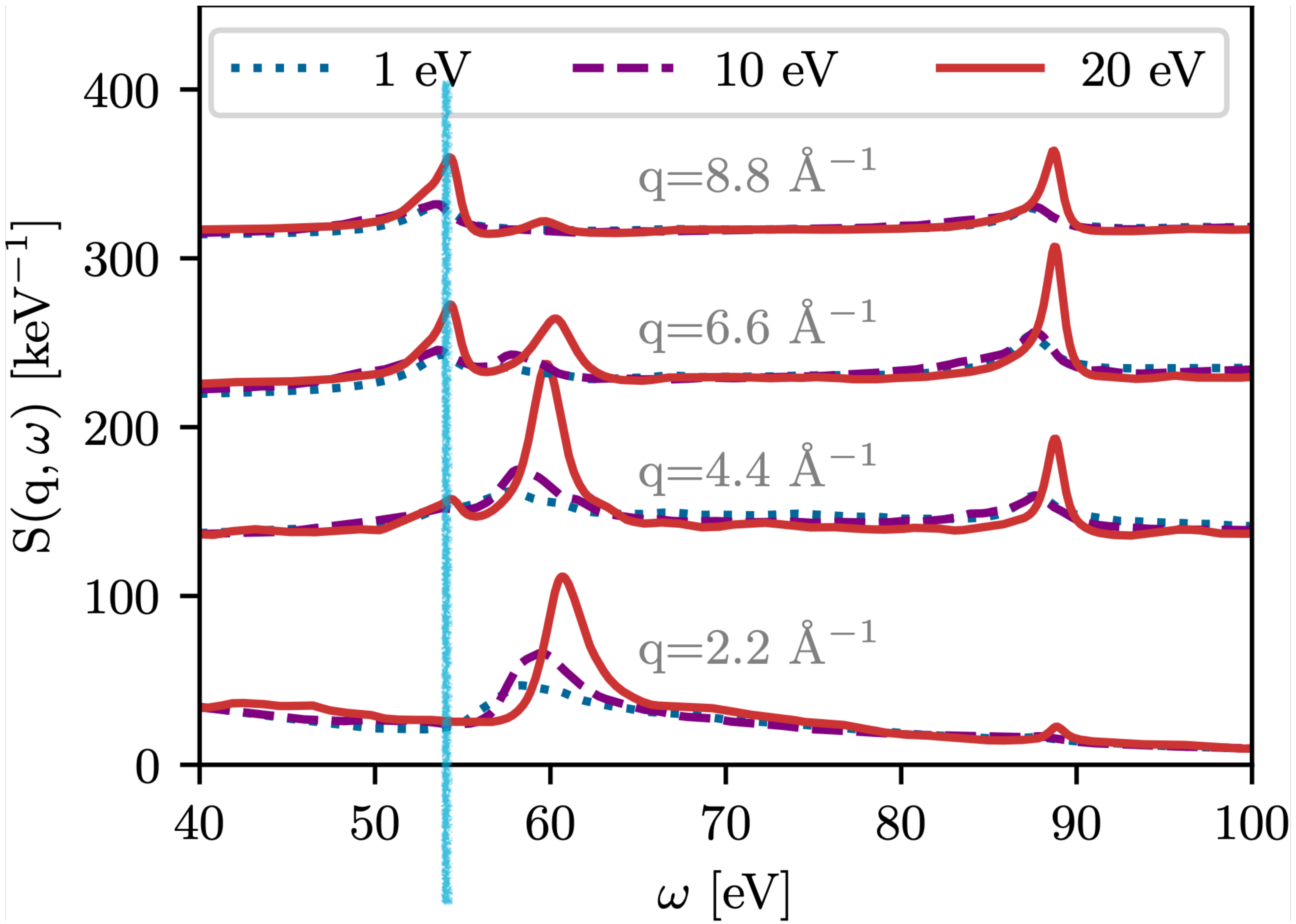
3s->3p @ 35 eV

There is a ~5 eV discrepancy between TDDFT and average atom for 3p-3d, worth further consideration...



Collective character of the iron 3p-3d feature

Solid density iron (8 g/cc)



Average atom predicts a non-dispersing bound-bound feature at 54 eV.

TDDFT predicts that a single-particle excitation around 54 eV will appear at large momentum transfers...

...but at smaller momentum transfers, this excitation has a collective character that gets stronger with temperature.

We have confirmed:
1) Not an exchange-correlation effect,
2) Kubo-Greenwood fails to reproduce.

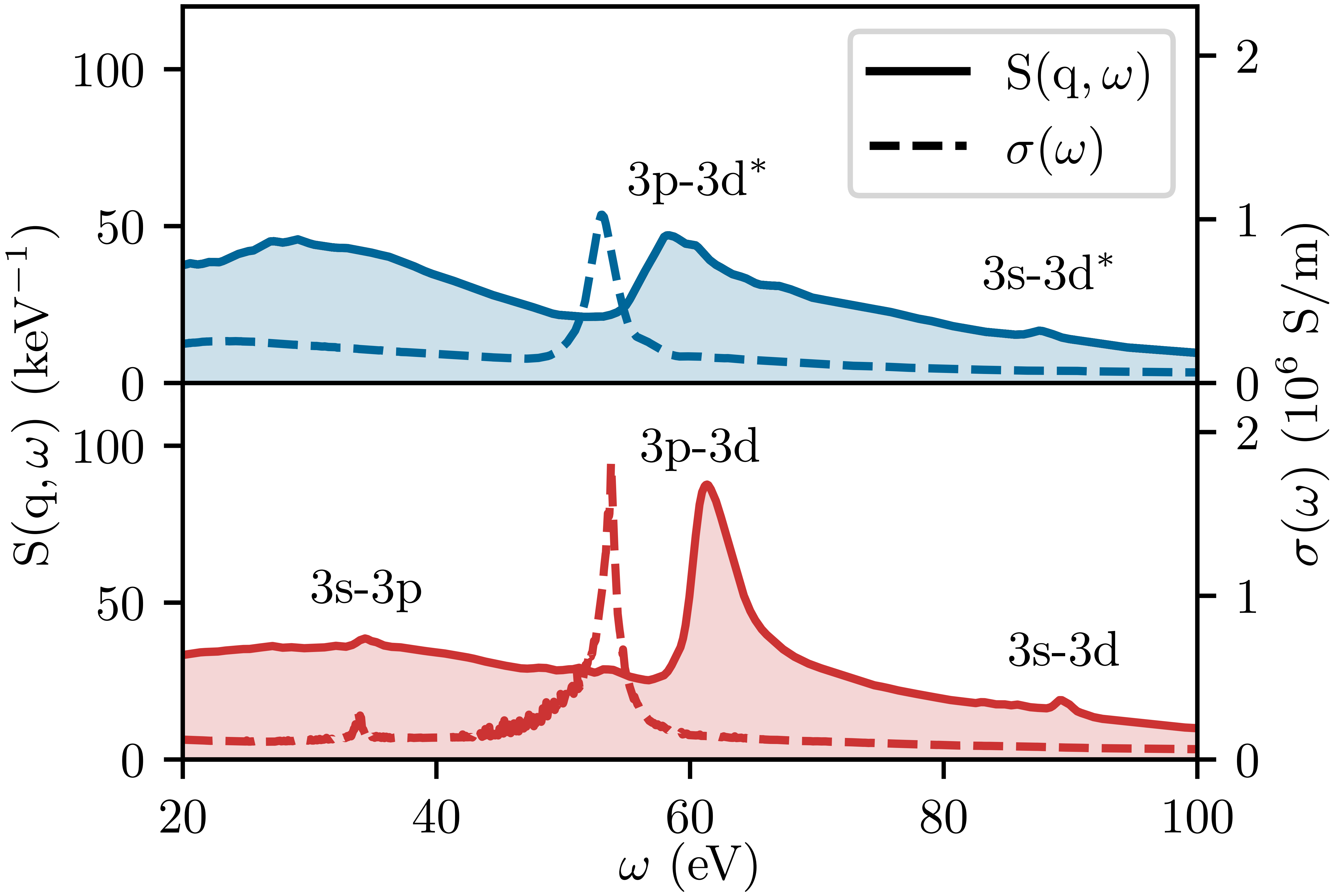
3d isn't *really* a bound state, it is a narrow band near the chemical potential.

Failure of Kubo-Greenwood*

Treatment of the Kubo-Greenwood* dielectric function common in our community is equivalent to a TDDFT calculation in which the Hartree+exchange-correlation kernel is zero.

Discrepancies between these treatments of the response function are thus entirely due to the neglect of collective effects in Kubo-Greenwood*.

Another way of putting this:
Kubo-Greenwood* is *only* capable of capturing single-particle (non-collective) excitations.



*Semantic distinction: I'm referring to the evaluation of the Kubo-Greenwood formula w/Kohn-Sham orbitals.
If you evaluated the Kubo-Greenwood formula with the exact wave function, this deficiency would not apply.

Inter-shell bound-bound processes in average atom

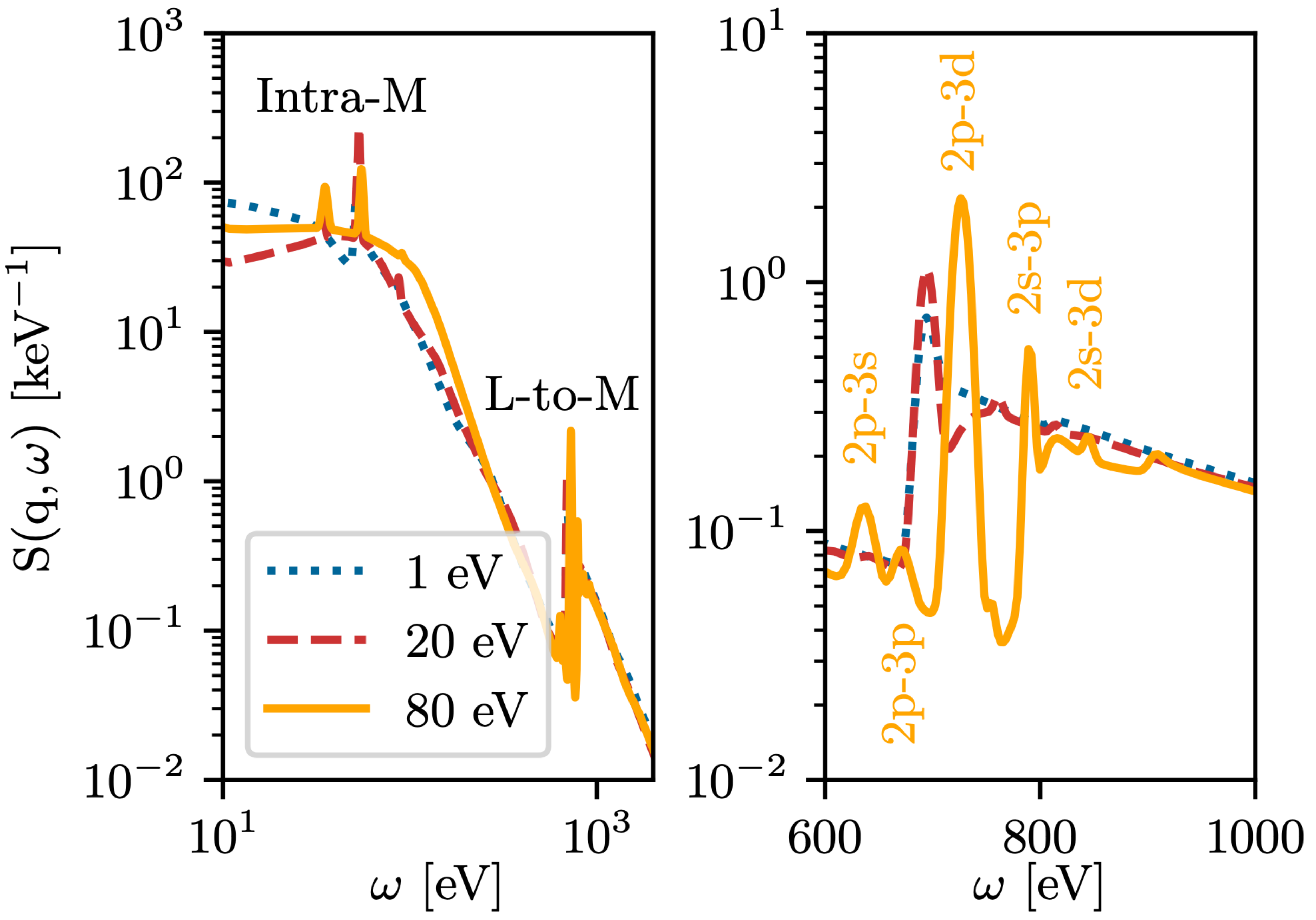
One benefit of average atom is being able to efficiently study conditions that are prohibitively expensive for TDDFT...

Looking at the L-shell in TDDFT would require (at least) O(100)x the CPU time!

We see that a rich set of inter-shell features around the L-edge at higher temperatures.

All of these features can be used in thermometry, better than plasmon shift for certain conditions.

Solid density iron (8 g/cc)



Conclusions

Check out arXiv:2109.09576* for the preprint, e-mail is adbacze@sandia.gov

Bound-bound transitions can contribute to x-ray Thomson scattering signals in warm dense matter.

Revised AA theory is corroborated by first principles TDDFT.

Even if bound-bound isn't visible, models should consistently account for phenomenology.

Opportunities to explore collective character of excitations previously considered to be 'single-particle' in systems w/narrow bands near the chemical potential.

More broadly, bound-bound transitions might be a useful new tool for thermometry in scattering experiments.

Under certain conditions, we should expect bound-bound transitions to dominate scattering response – as is the case in opacity.

*Expect v2 any day, now...

Sandia National Laboratories

Sandia National Laboratories is a multi-missions laboratory managed and operated by National Technology and Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International Inc., for DOE's National Nuclear Security Administration under contract DE-NA0003525.

CCR Center for Computing Research