SAND2022-15128C

Real-time methods in the warm dense regime:

Recent progress & future prospects

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Theory Meets XFELs
2022-11-04

CCR Center for Computing Research

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.

Acknowledgements.

Tommy Hentschel, Cornell - AA

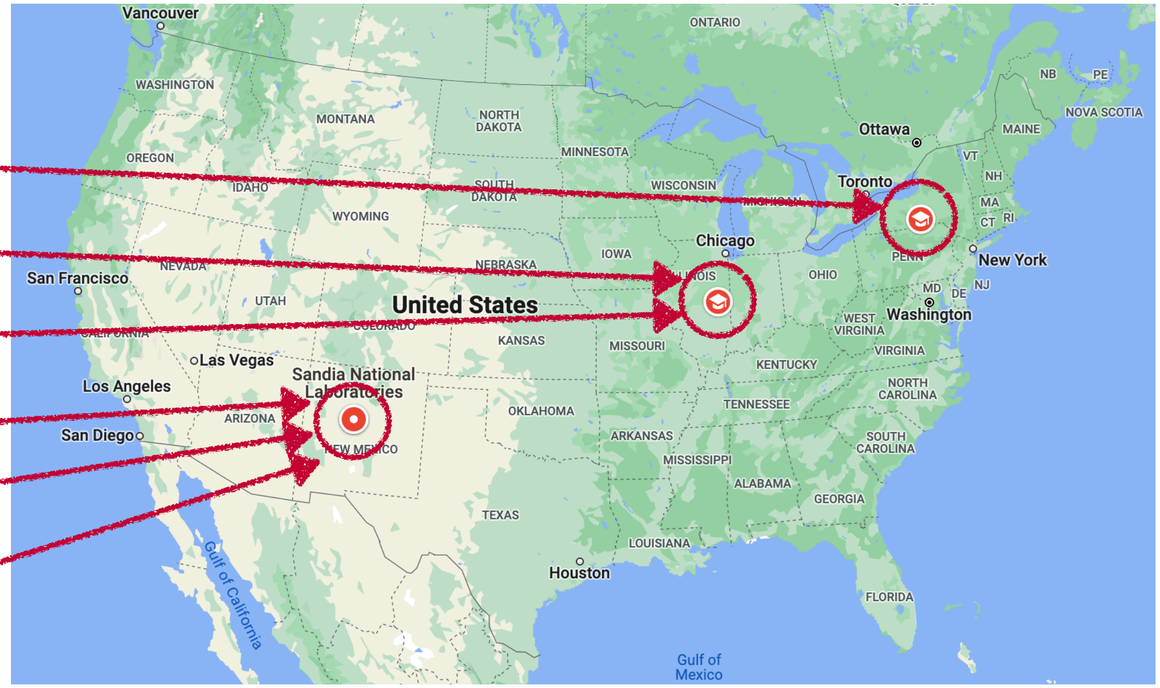
Brian Robinson, UIUC - DFT/GW/BTE

Andre Schleife, UIUC - DFT/GW/BTE

Alina Kononov, Sandia - TDDFT

Stephanie Hansen, Sandia - AA

Alexandra Olmstead, Sandia - TDDFT



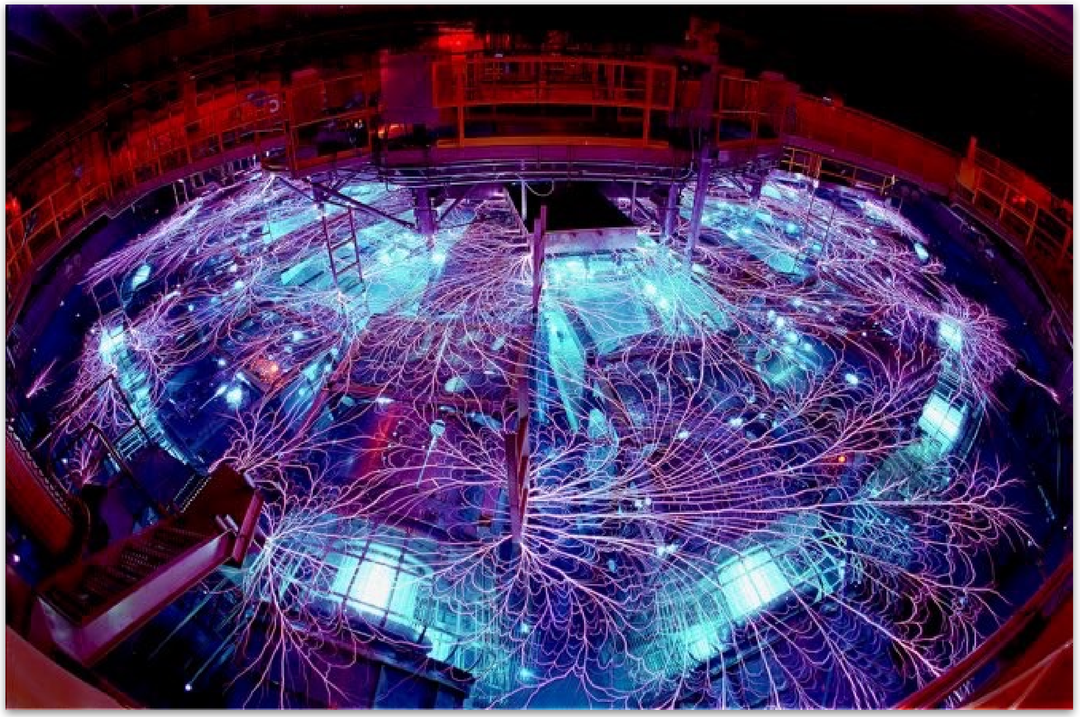
Funding from:

National Nuclear Security Administration

Sandia Laboratory Directed Research and Development

Two different HED science facilities.

Z Pulsed Power Facility, Sandia National Labs



O(10 MA) currents in O(10 ns)

European XFEL, here :)

O(1 trillion) x-ray photons in <100 fs

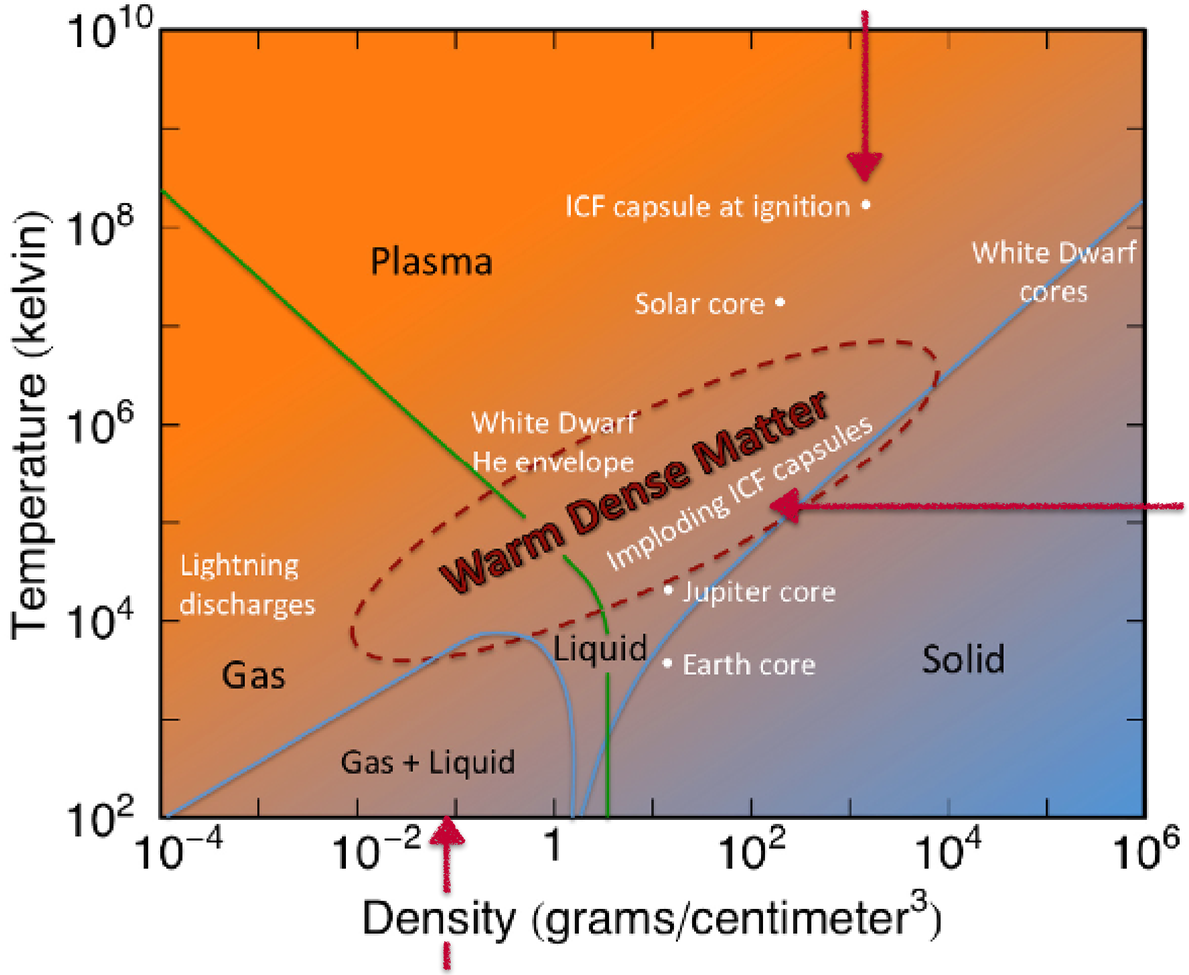
Both facilities create and probe extreme conditions,
real-time methods impact science at both.

Z figure credit: Randy Montoya

EuXFEL figure credit: Andrew Baczewski

Materials in extreme conditions.

Fusion happens here



Hard to constrain materials models here

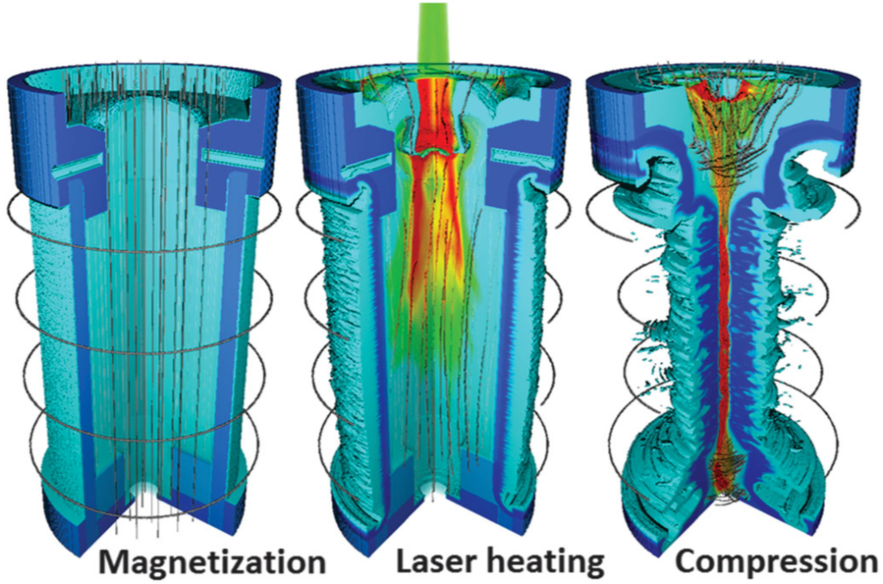
(And interesting basic science!)

But the fuel starts here

Figure credit: Mike Desjarlais

Challenges of multiphysics modeling.

What physics needs to be captured for ICF?
(e.g., MagLIF - M.R. Gomez *et al.*, PRL (2014))



Current in walls drives implosion via Lorentz force

Laser coupling to fuel and window

Magnetic inhibition of transport

Alpha particle self-heating,
balance against radiation and conduction loss

X-rays generated in plasma propagate through surrounding matter

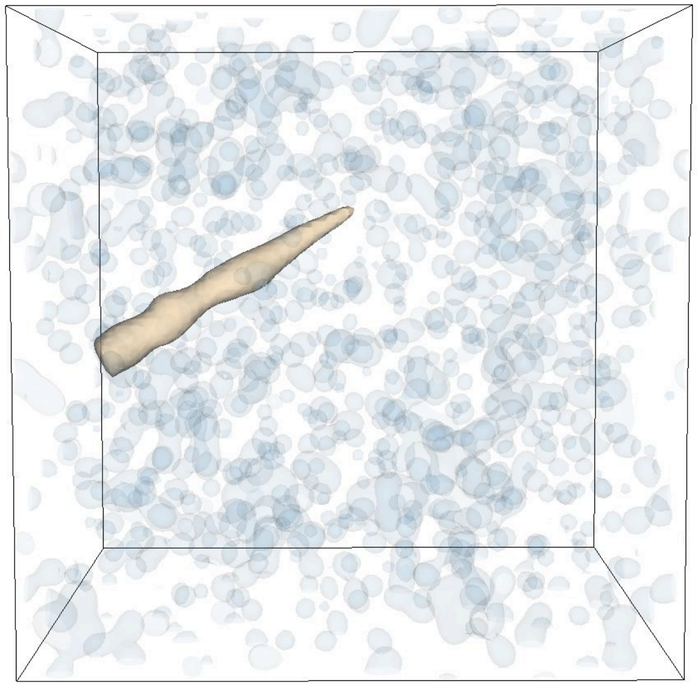
Vast multidisciplinary expertise dedicated to capturing all of this in a large system of coupled partial differential equations (PDEs).

We want high-fidelity models of the *coefficients* of those PDEs.

Figure credit: M.R. Gomez *et al.*, Phys. Rev. Lett. **113** (2014)

Many-atom models are expensive.

Say that we want to compute electronic stopping power in the fuel...



This calculation consisted of 1024 deuterium atoms, 10 g/cc and 2 eV.

Movie created for the charged-particle transport coefficient comparison workshop, Grabowski, et al., HEDP, 2020

Each projectile velocity took ~1 day on a moderately large HPC system (~10k cores).

Higher Z, higher T, larger cells - all require more time.

Our biggest stopping calculations, 1 curve = 1 machine-week on a million-core system

Average-atom models are inexpensive.

We need to cut the cost of these calculations down by 6-8 orders of magnitude* if we want to use them for tabulation of materials models - particularly non-LTE!

AA models fit this description,
but they're necessarily making more severe approximations than many-atom models.

What does TDDFT tell us about the quality of those approximations?

At Sandia, we've been undertaking a comparison between AA and (TD)DFT.

The rest of this talk...

1.) Why we think real-time methods are great.

2.) Some exemplary results from our recent work.

3.) What's next?

*Notably, there are tabular DFT models that take O(months) to develop on O(10k cores), but there are good reasons to do it faster and/or save the CO_2 emissions.

Quick course in real-time time-dependent DFT

Simple, integrate the time-dependent Kohn-Sham equations

$$i\frac{\partial}{\partial t}\phi_{n,\mathbf{k}}(\mathbf{r},t) = \left(-\frac{\nabla^2}{2} + v_S\left[\rho\right](\mathbf{r},t)\right)\phi_{n,\mathbf{k}}(\mathbf{r},t)$$

$$v_S\left[\rho\right](\mathbf{r},t) = v_{ext}(\mathbf{r},t) + v_H\left[\rho\right](\mathbf{r},t) + v_{xc}\left[\rho\right](\mathbf{r},t)$$

$$\rho(\mathbf{r},t) = \sum_{n,\mathbf{k}} f_{n,\mathbf{k}}(T_e)|\phi_{n,\mathbf{k}}(\mathbf{r},t)|^2$$

Noteworthy approximations:

1.) Finite basis set and pseudization of electron-ion interaction

2.) Exchange-correlation model

3.) We don't *really* know what we're doing with temperature

Nevertheless, we can model a wide range of perturbations and compute many observables that are density functionals. Also, *extremely* scalable.

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*

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

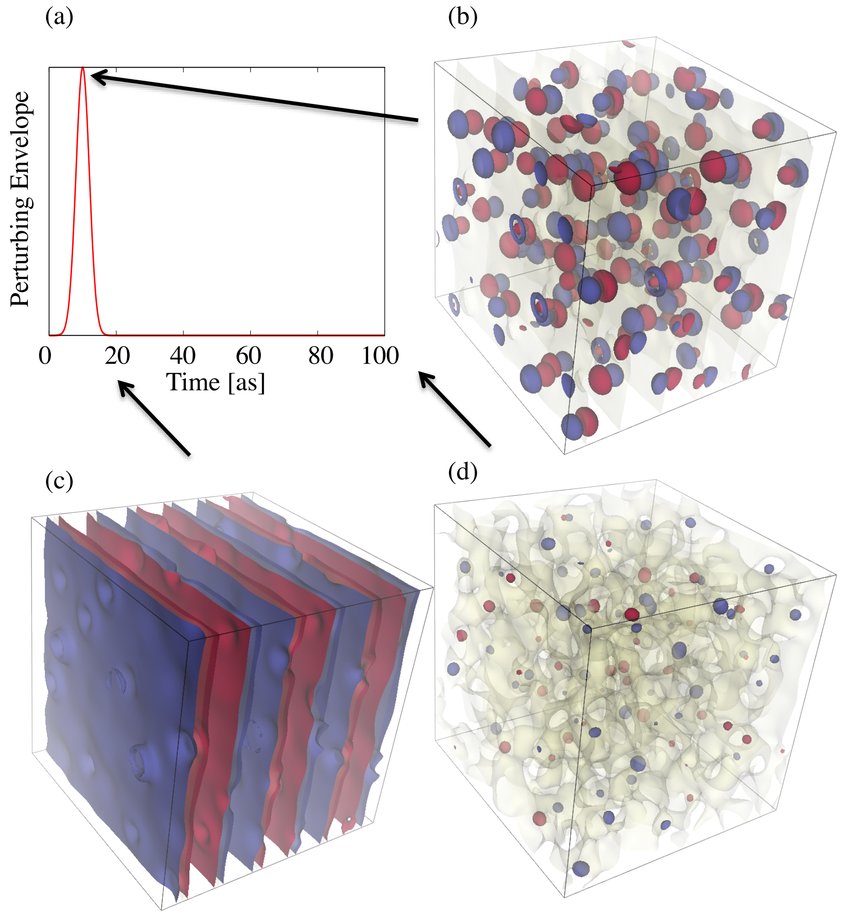
Record density response

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

Apply fluctuation-dissipation

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

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



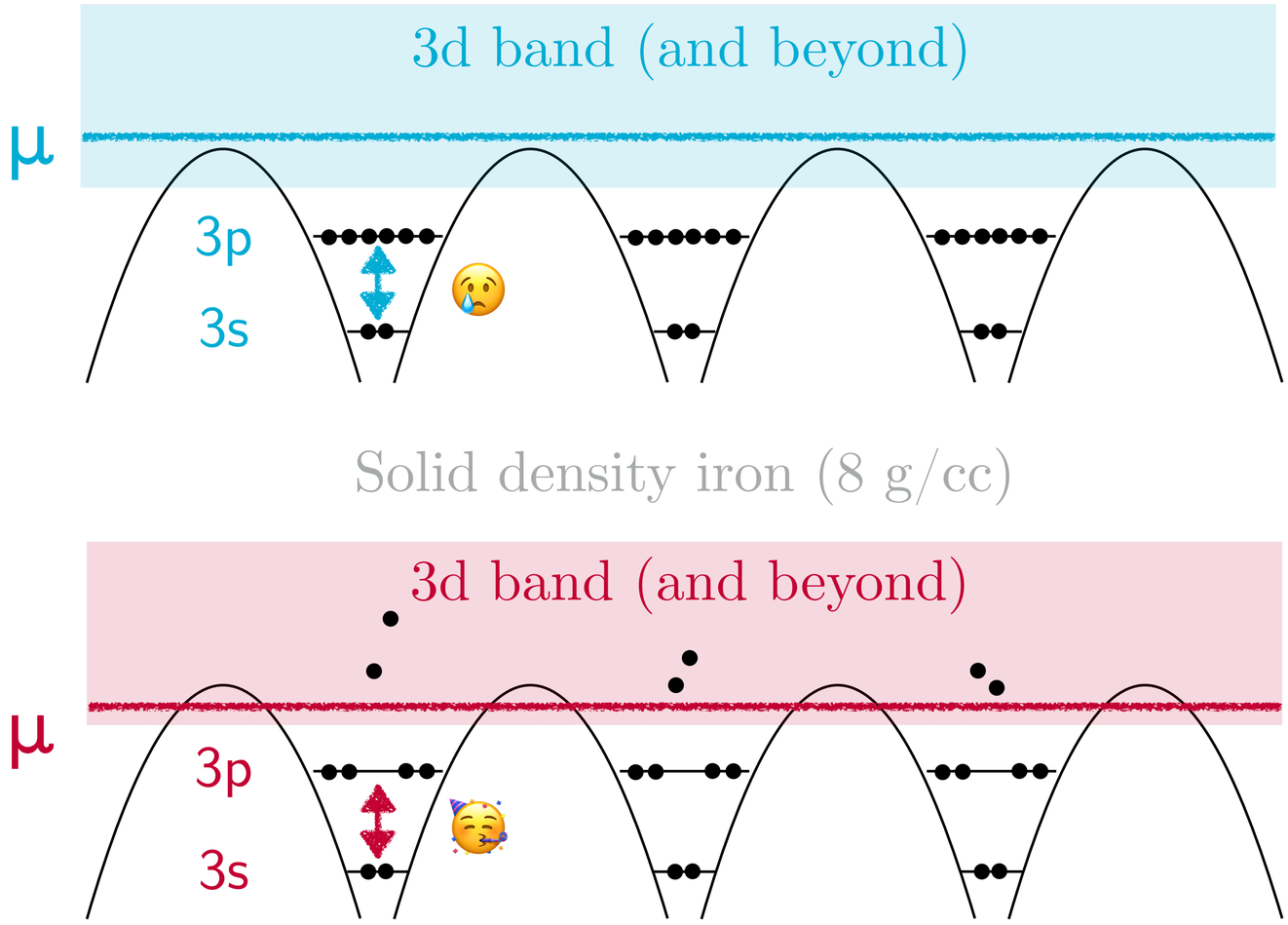
*energy/wave vector set by energy/momentum transfer of interest

Bound-bound transitions in warm dense matter

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

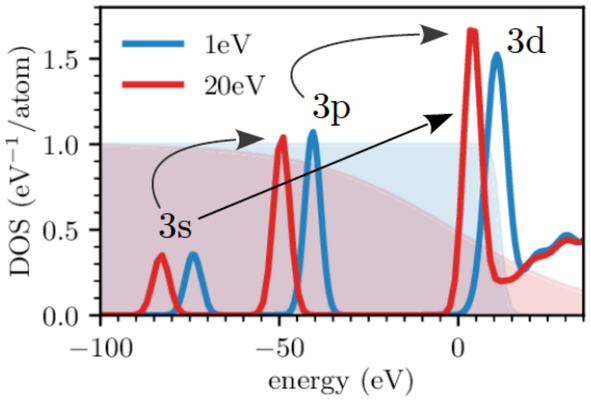
Exactly one electron is allowed to have the quantum numbers that it has...

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



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.

Iron (d-band near chemical potential)



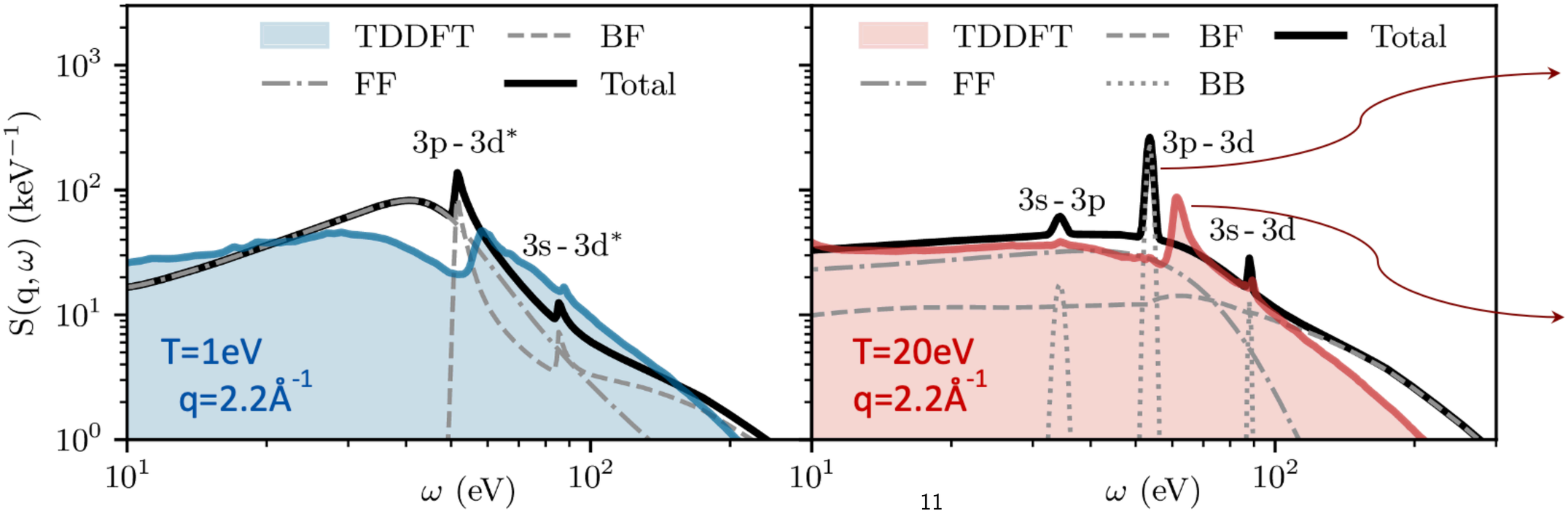
The spectrum of bound-bound transitions is richer yet in iron.

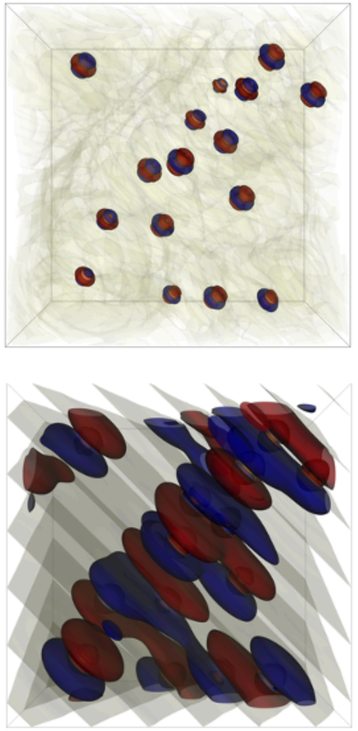
3p->3d @ 55 eV

3s->3d @ 85 eV

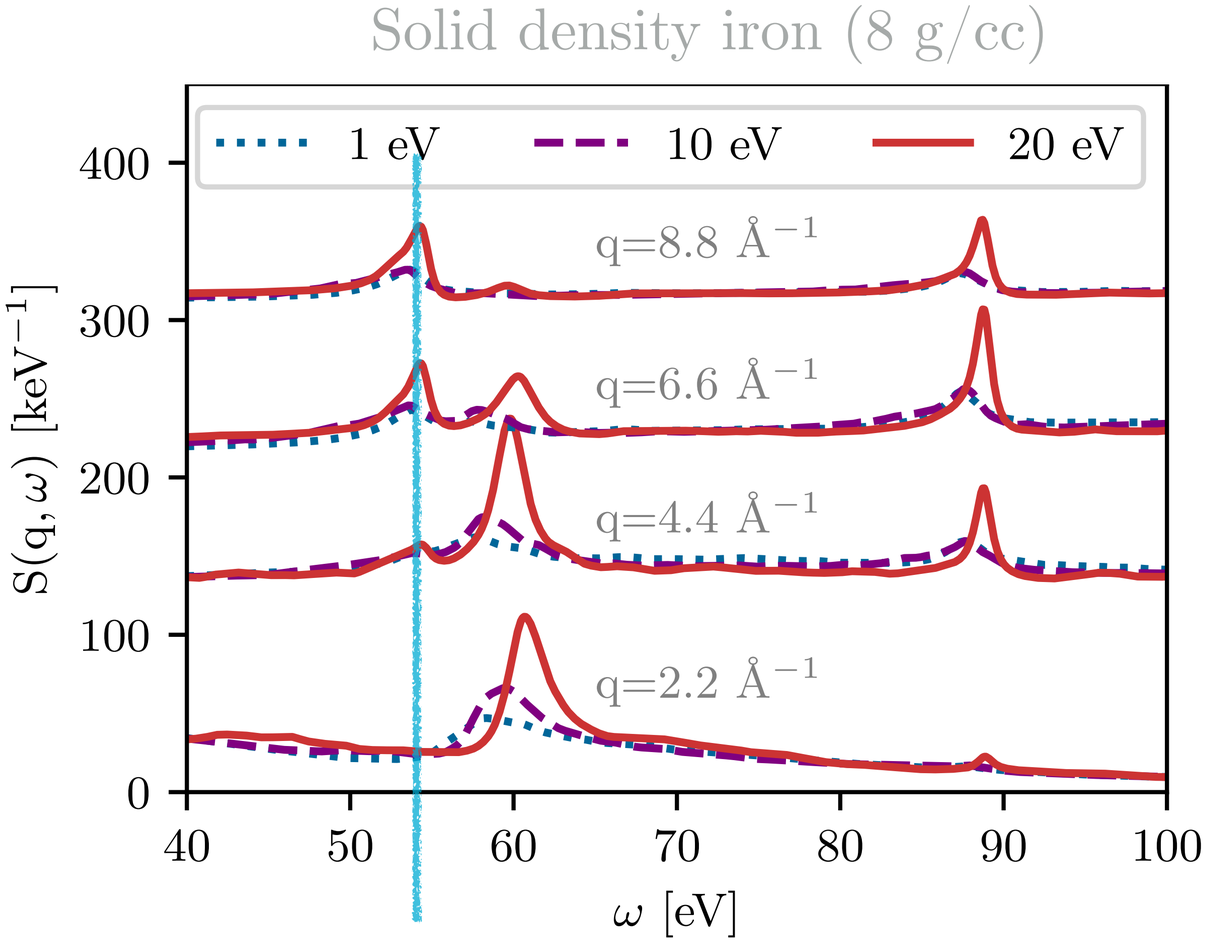
3s->3p @ 35 eV

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



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.

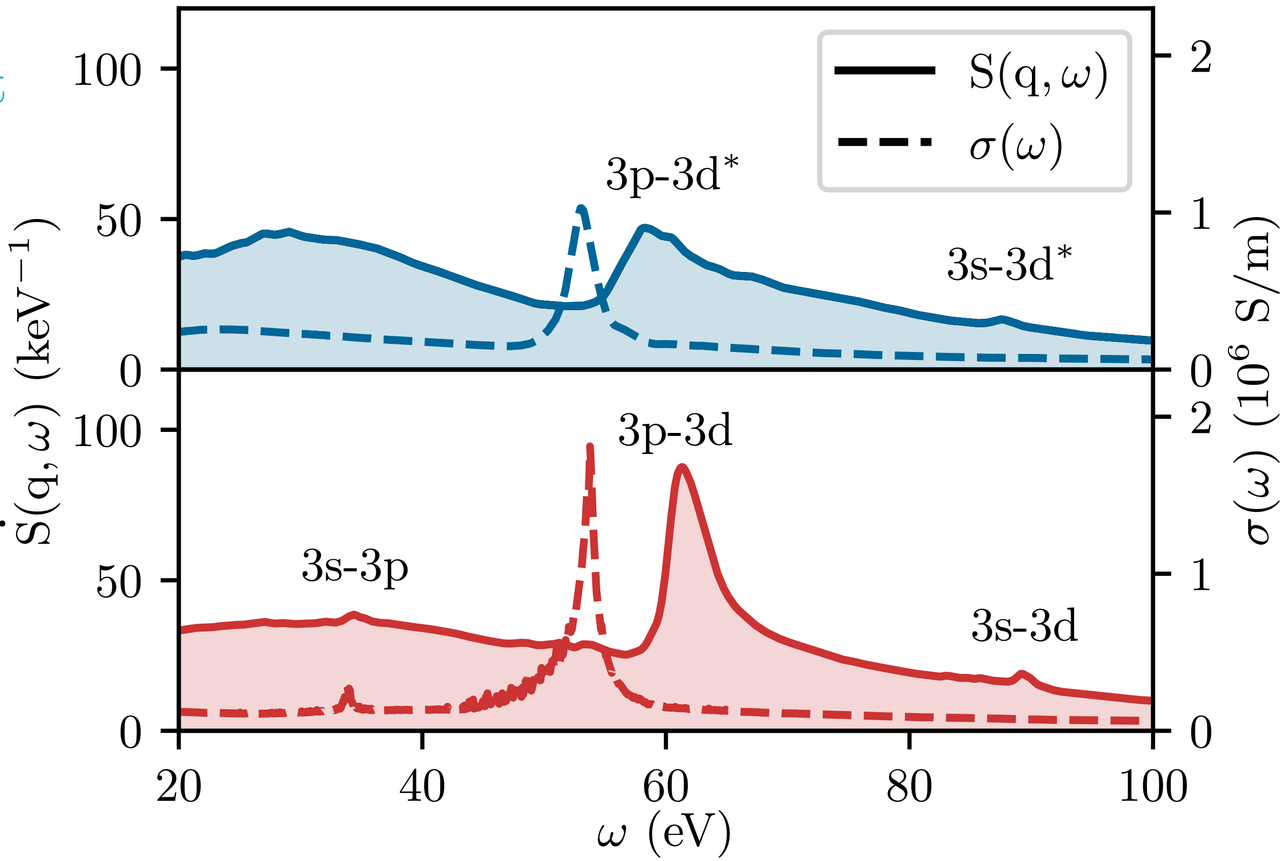
3d isn't *really* a bound state, it is a narrow band near the chemical potential.

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.



*Important semantic distinction: I'm referring 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.

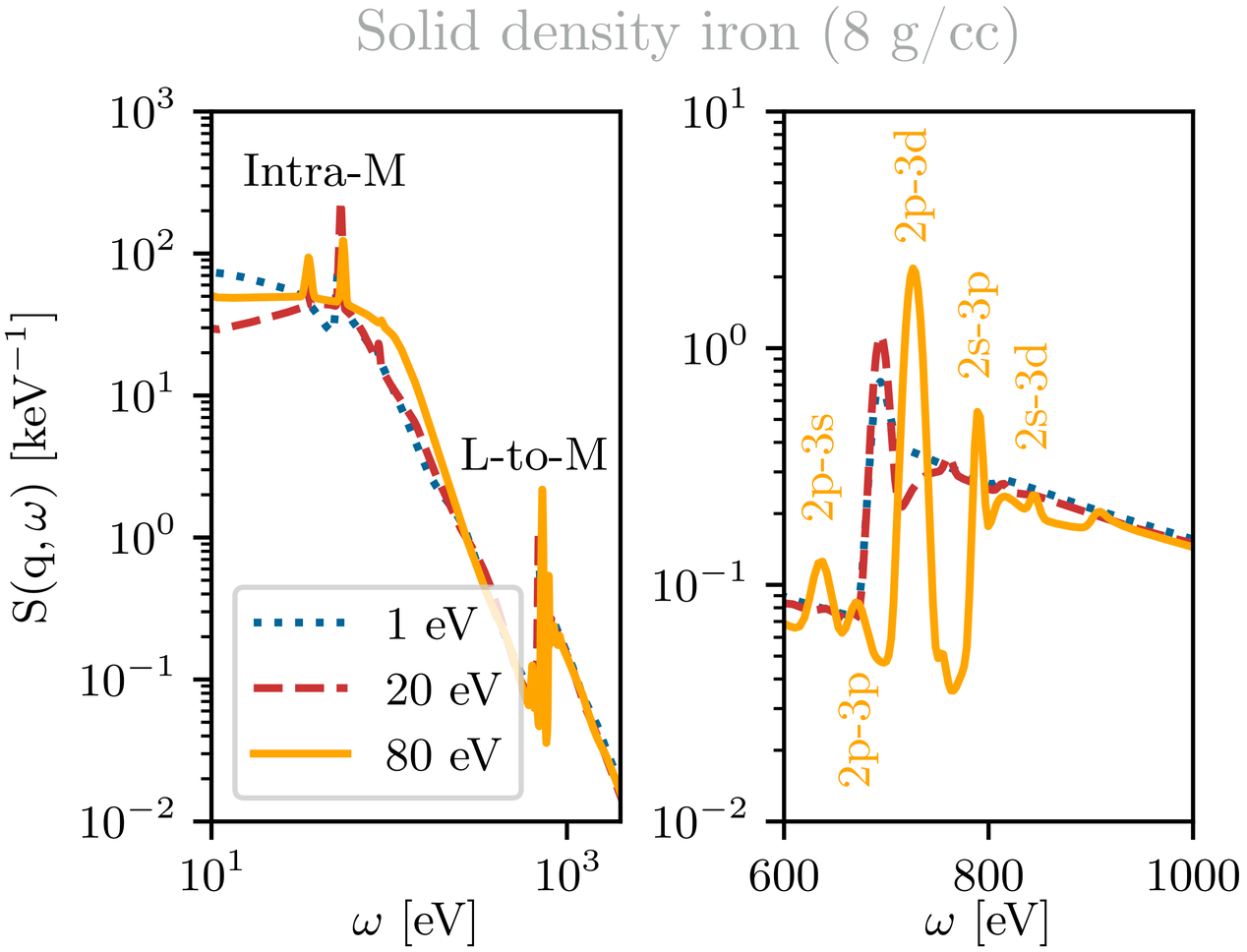
Inter-shell bound-bound processes

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.

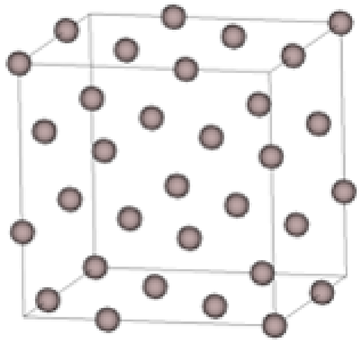
All of these features can be used in thermometry, better than plasmon shift for certain conditions.

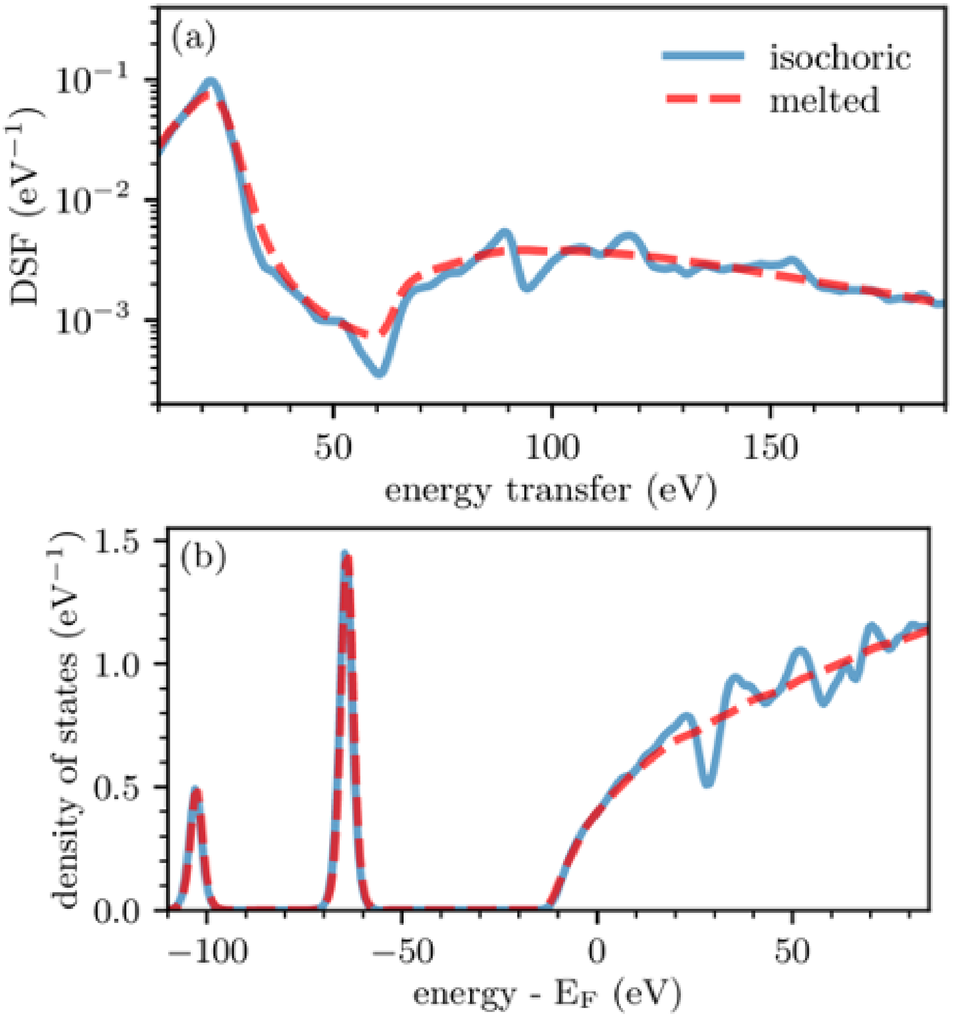


Signatures of non-equilibrium in XRTS

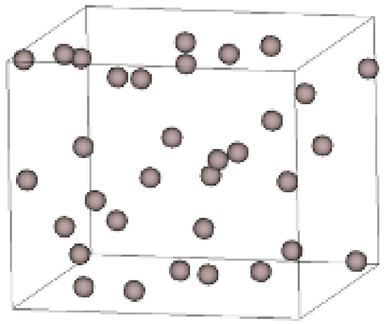
Band structure effects persist for hot electrons that aren't equilibrated with the underlying ions...

Isochoric





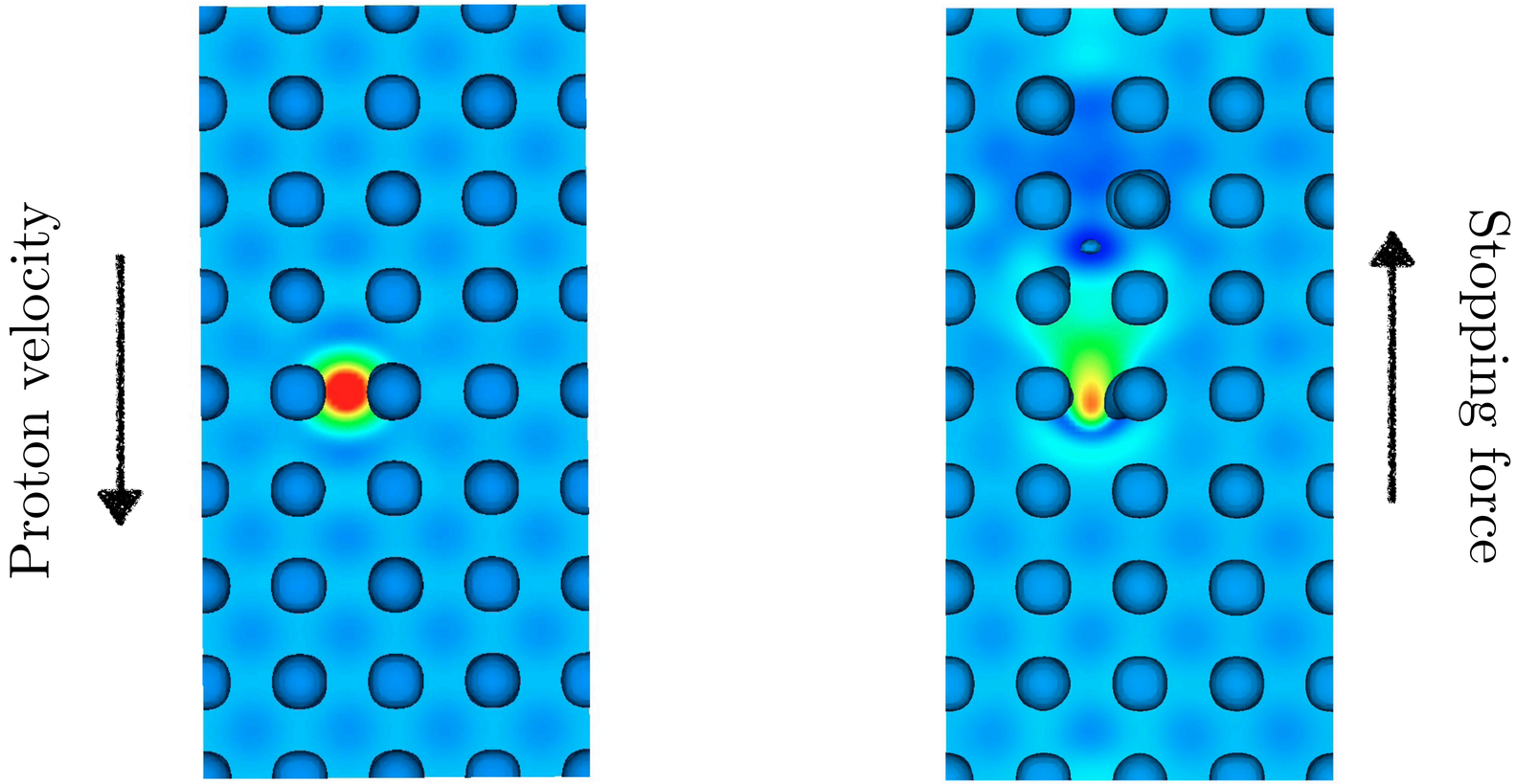
Melted



Stopping power from real-time TDDFT

Born-Oppenheimer + DFT

"Ehrenfest"+ TDDFT



(Left) Average force is identically zero

(Right) Average force is *not* zero, even agrees well with experiment

Stopping power in solids

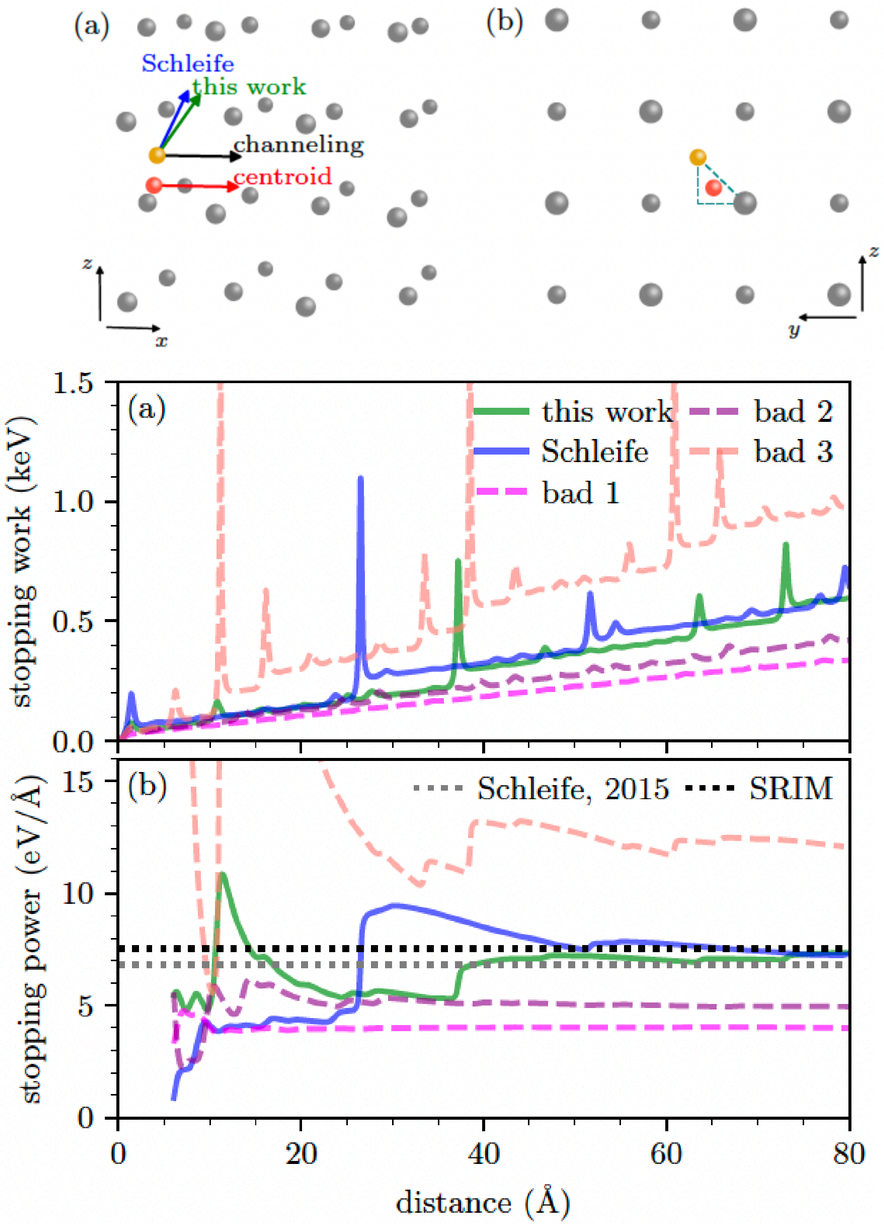
Stopping power is another property of interest for materials in extreme conditions.

These calculations are among the most expensive that we do, O(100 MCPU-hour/year).

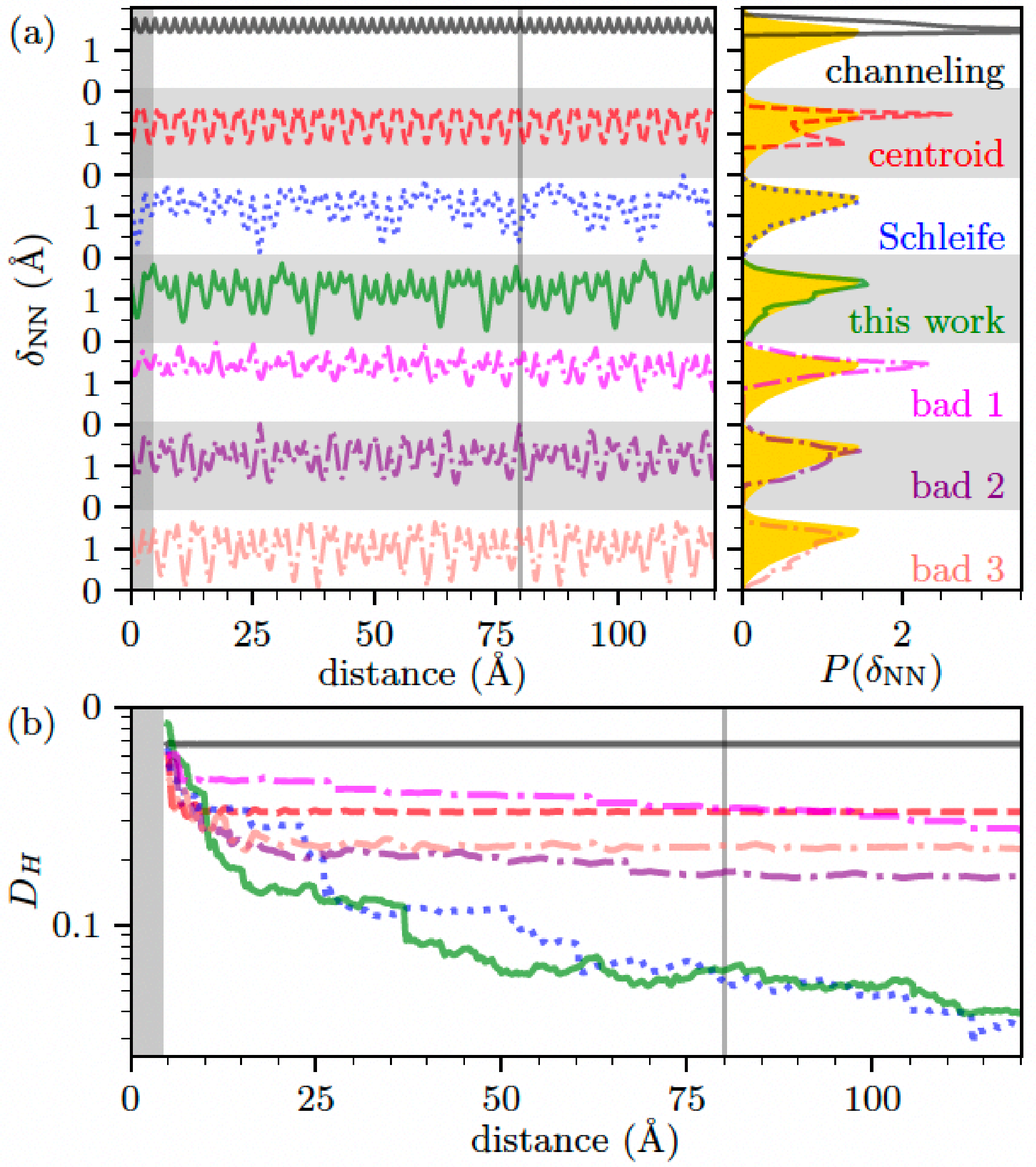
"Warm dense" calculations dominate our "budget".

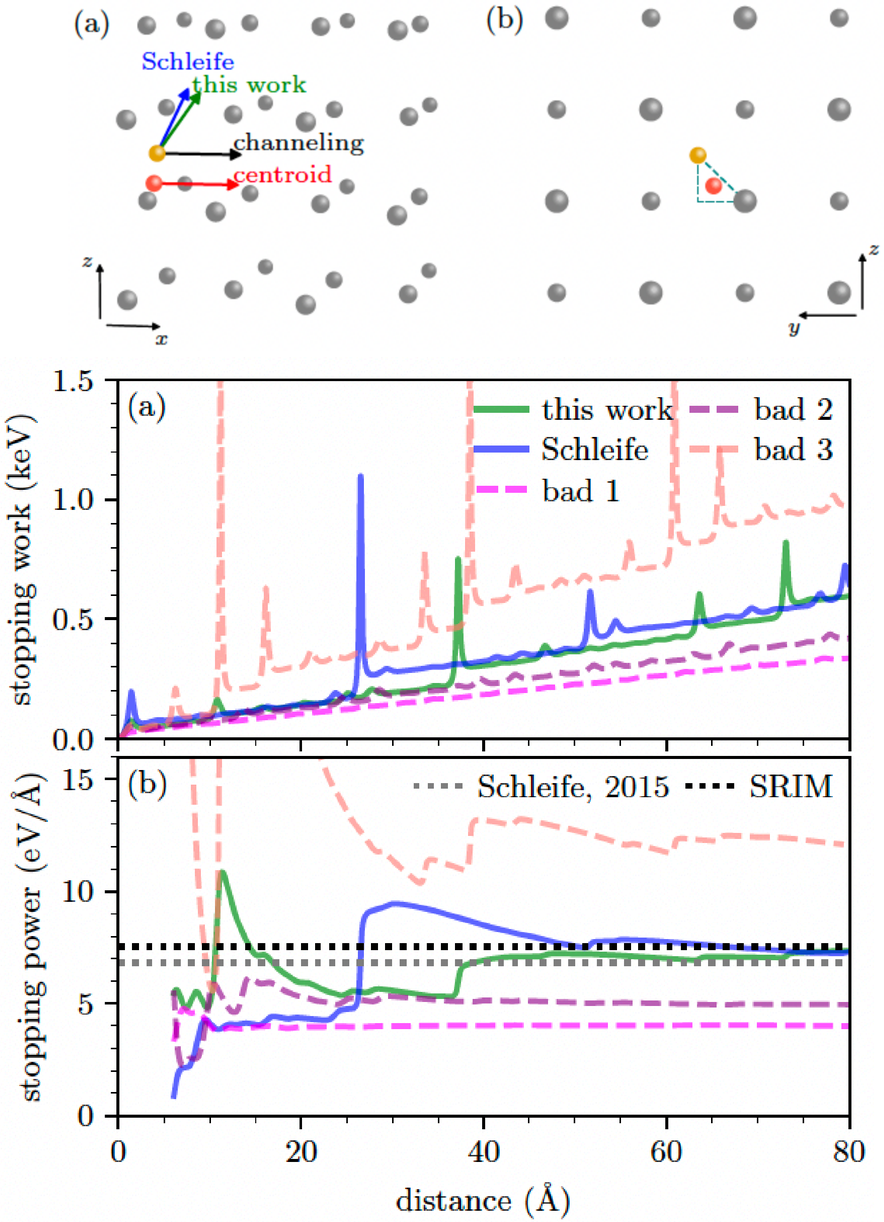
Because of their expense, we set out to determine the most efficient way to select trajectories.

Crystalline solids present a simple starting point.

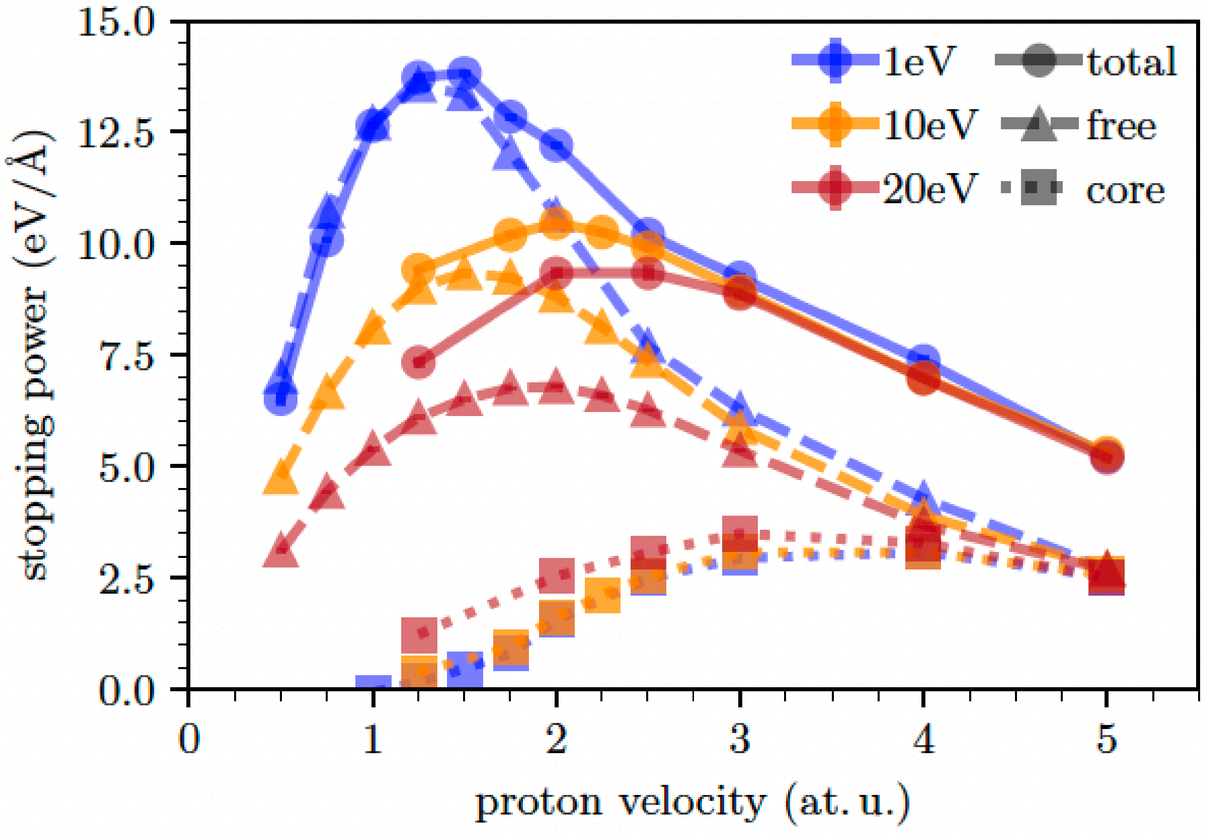


Stopping power in solids





Stopping power in warm dense aluminum



We are able to select trajectories that enable us to determine representative stopping powers for warm dense aluminum.

Calculations took 250 MCPU-hours over 1.5 years, "just averaging" over a few more trajectories isn't really feasible.

Aim is to inform AA model calibration, but...

Integrated stopping + XRTS experiments would be invaluable for reconciling whether our model is worth the cost of generating calibration data

Conclusions

arXiv:2109.09576, preprint on bound-bound results. E-mail - adbacze@sandia.gov.

Experimental design and macroscopic simulations require wide-ranging materials models.

Where multi-atom models are too expensive, average-atom models can step in.

We have inclinations about where AA models need refinement and TDDFT corroborates these.

Augmented AA theory w/rigorous extension of established scattering theory and partitioning techniques common in opacity to account for bound-bound scattering, consistent w/TDDFT.

We proposed a metric for determining typicality of trajectories for many-atom stopping, prospectively enormous savings in CPU time.

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