

Warm Dense Matter:

What theory is telling experiment,
what theory could be telling experiment,
and why method developers should care

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Non-Conventional Computing Technologies
Center for Computer Research
Sandia National Laboratories

Joint ICTP-IAEA School and Workshop on Fundamental Methods for Atomic, Molecular
and Materials Properties in Plasma Environments

April 17, 2018



Acknowledgements

Thanks to the organizers
and for contributions from/conversations with:

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Michele Pavanello (Rutgers)

Ronald Redmer (Rostock)

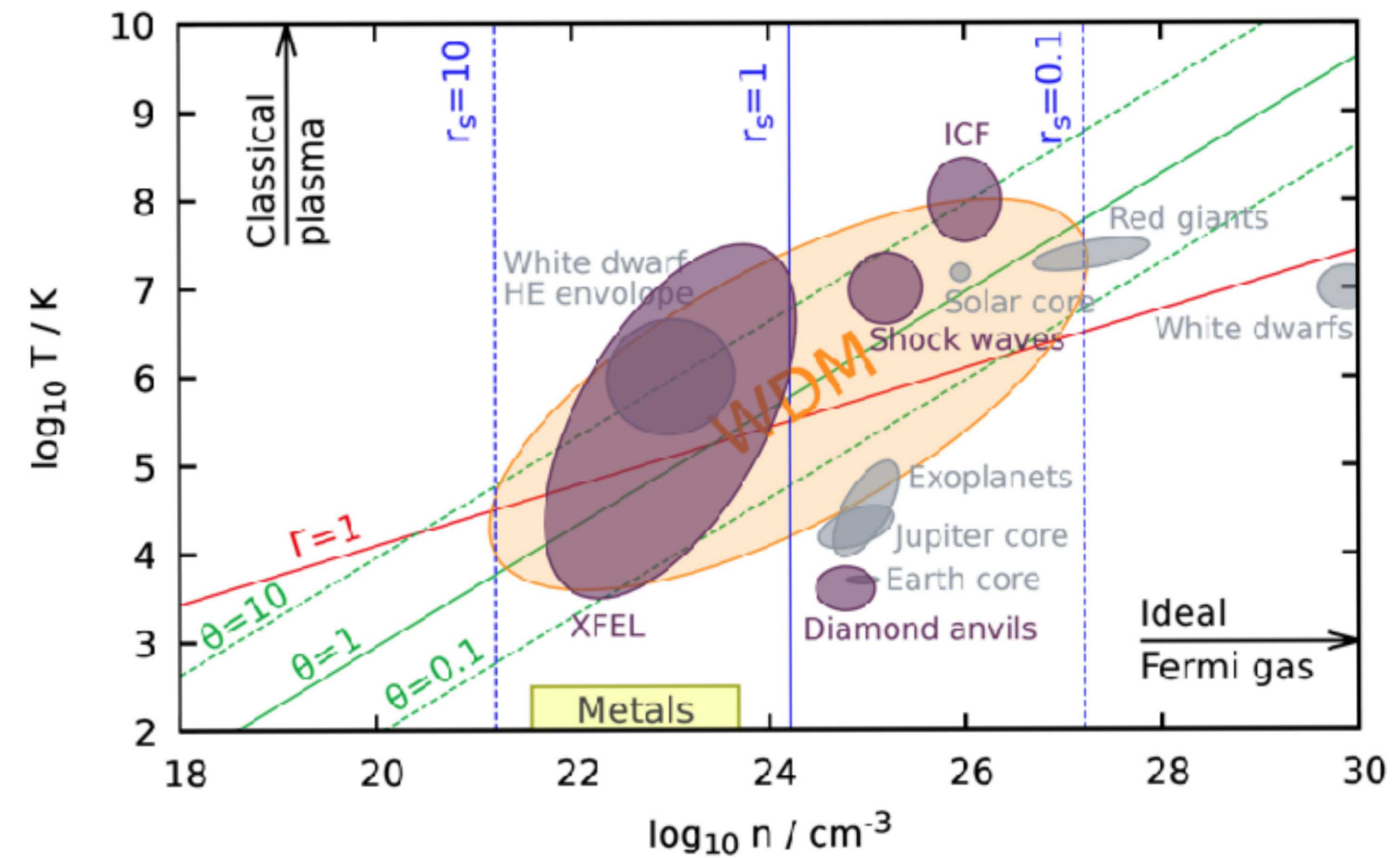
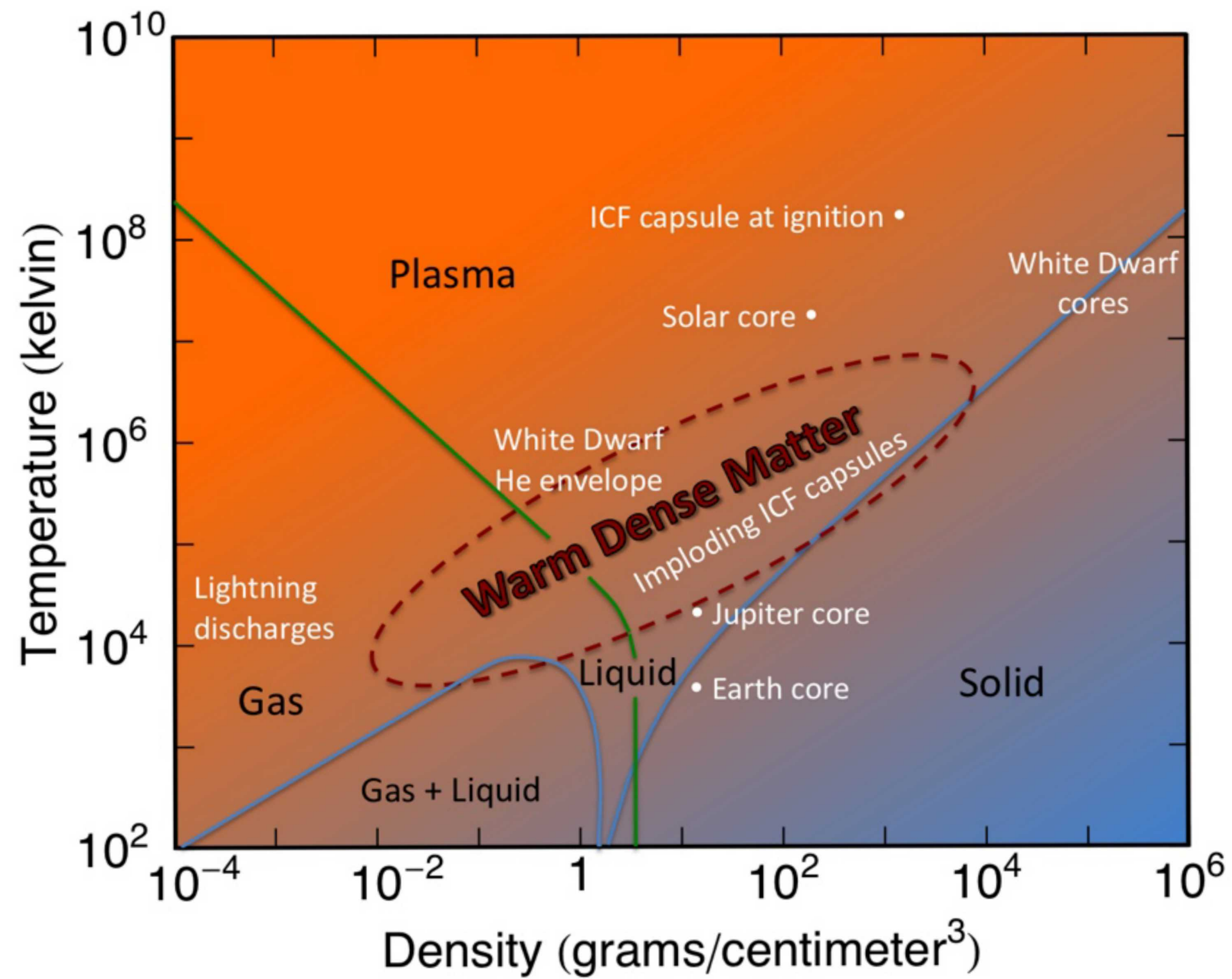
Siegfried Glenzer (SLAC)

Kai-Uwe Plagemann (Rostock)

Philipp Sperling (Rostock/SLAC)

Bastian Witte (Rostock/SLAC)

Warm Dense Matter



Dornheim, Groth, and Bonitz,
[arXiv:1801.05783](https://arxiv.org/abs/1801.05783)

Choose your favorite units of density

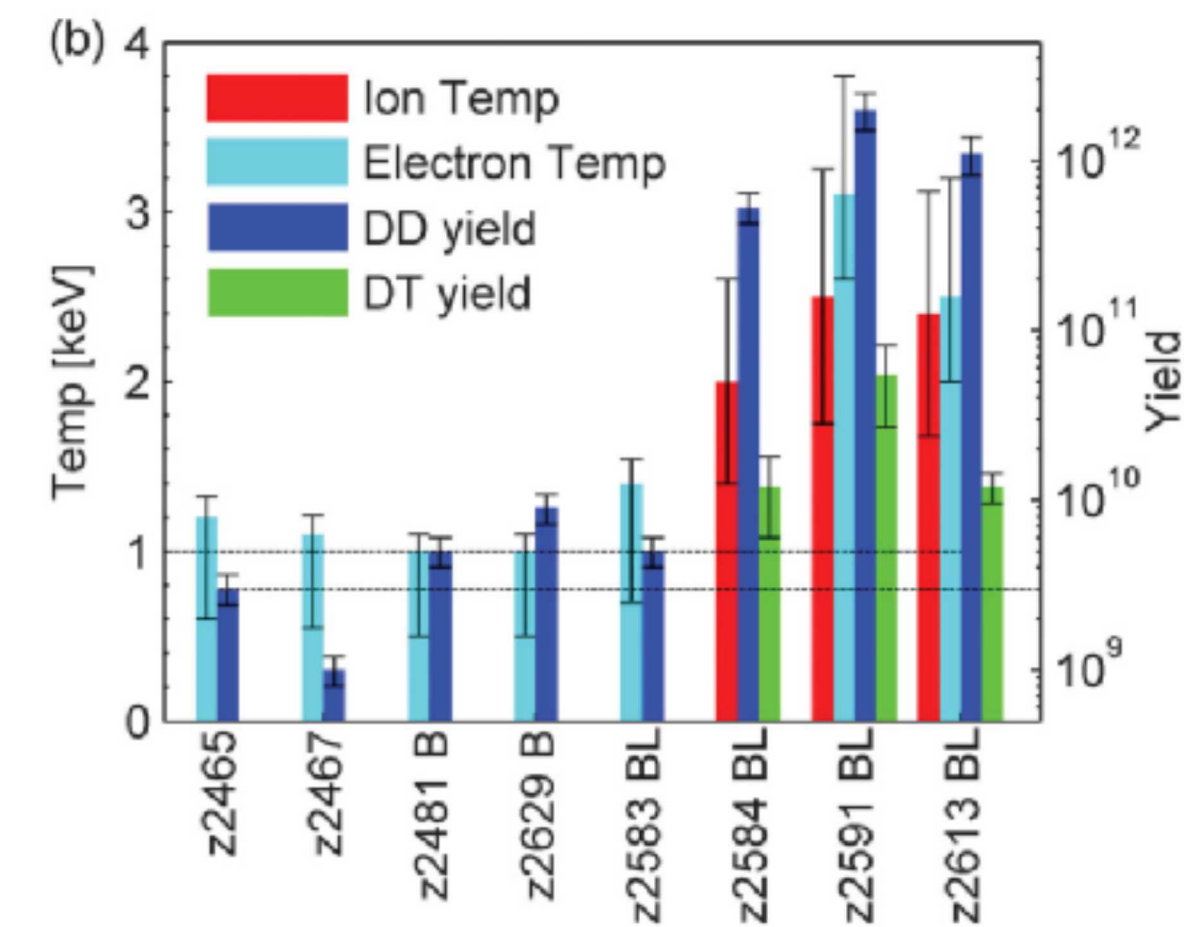
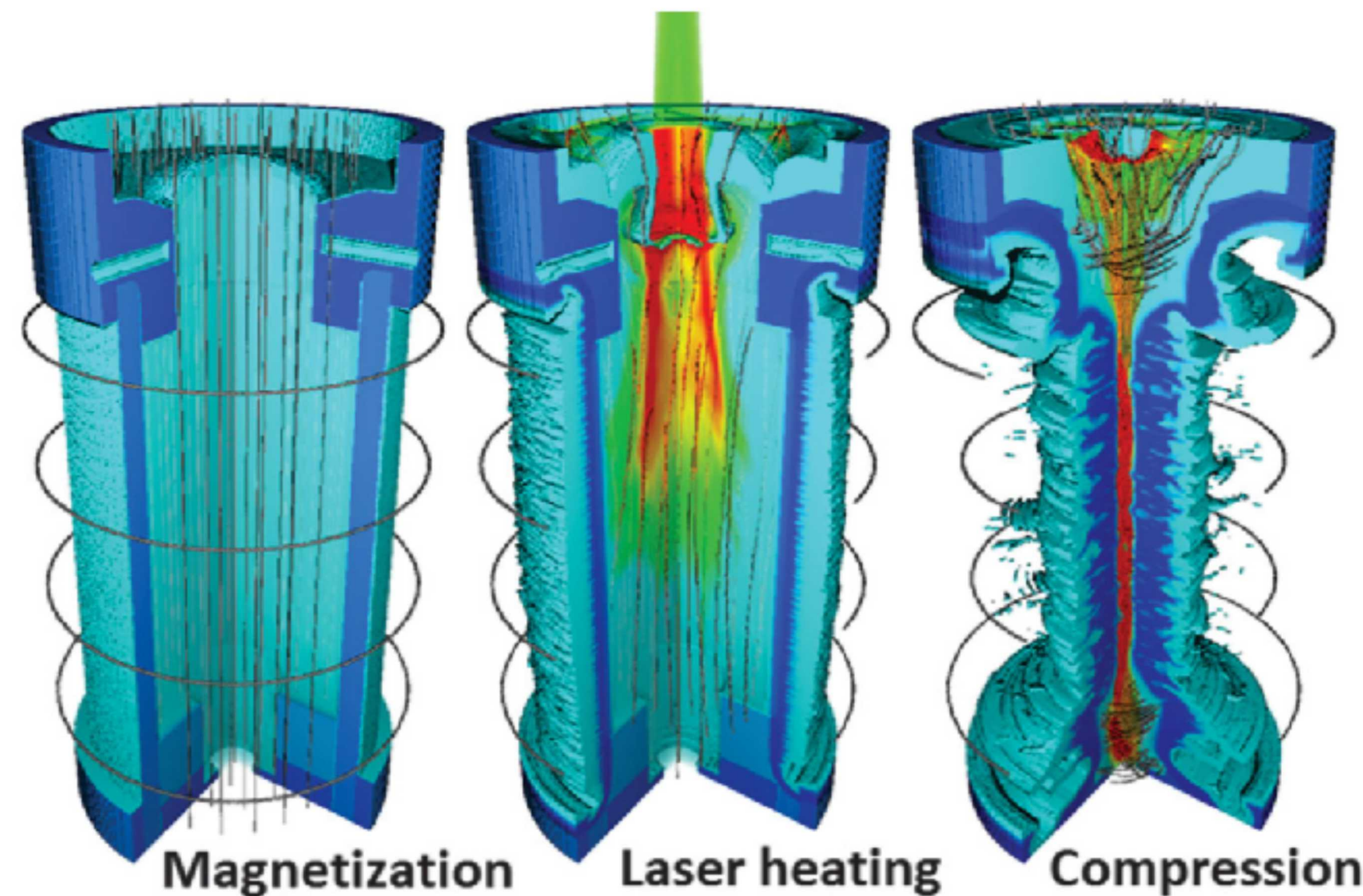
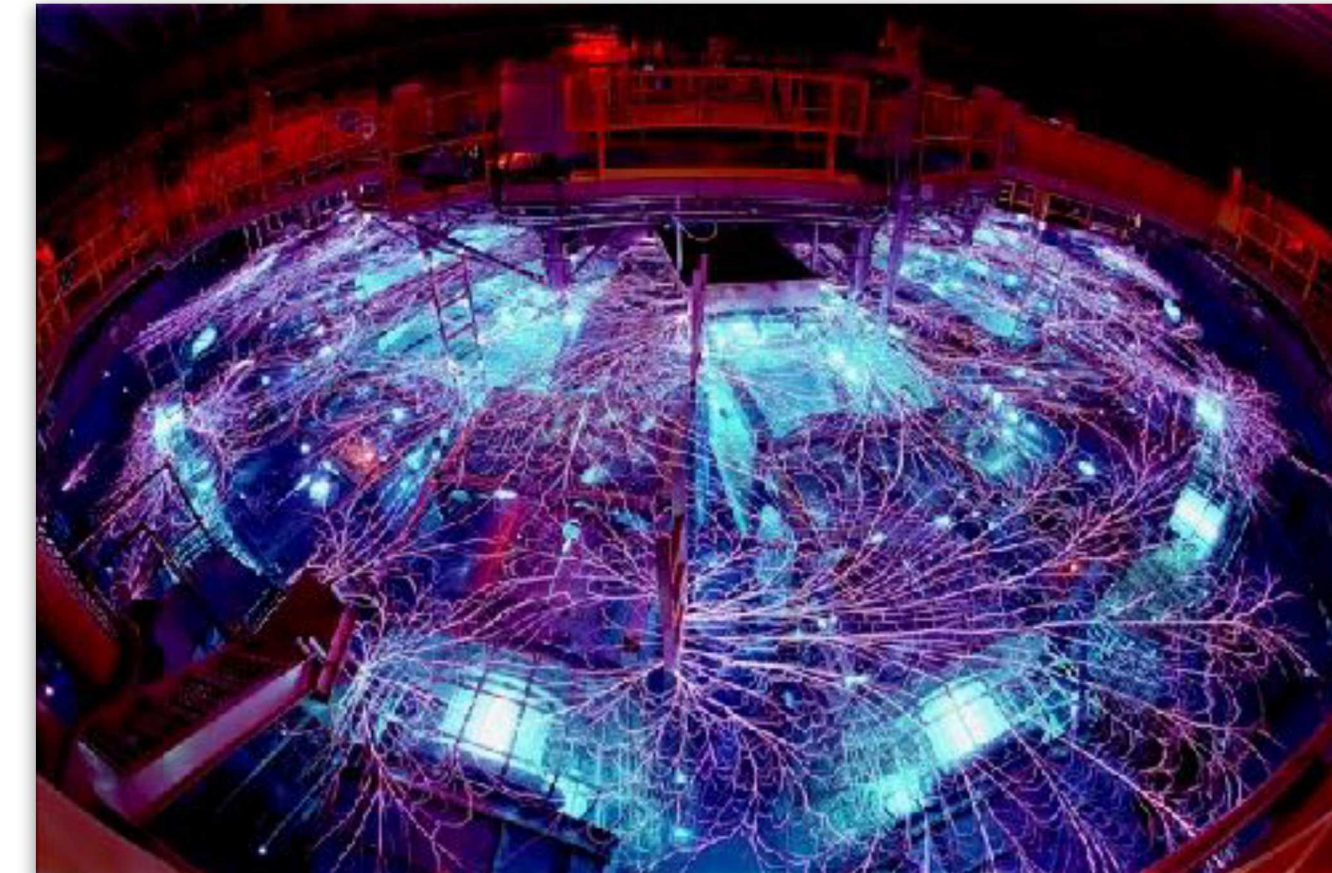
What is *theory* telling *experiment*?

PRL 113, 155003 (2014) PHYSICAL REVIEW LETTERS week ending 10 OCTOBER 2014

Experimental Demonstration of Fusion-Relevant Conditions in Magnetized Liner Inertial Fusion

M. R. Gomez, S. A. Slutz, A. B. Sefkow, D. B. Sinars, K. D. Hahn, S. B. Hansen, E. C. Harding, P. F. Knapp, P. F. Schmit, C. A. Jennings, T. J. Awe, M. Geissel, D. C. Rovang, G. A. Chandler, G. W. Cooper, M. E. Cuneo, A. J. Harvey-Thompson, M. C. Herrmann, M. H. Hess, O. Johns, D. C. Lamppa, M. R. Martin, R. D. McBride, K. J. Peterson, J. L. Porter, G. K. Robertson, G. A. Rochau, C. L. Ruiz, M. E. Savage, I. C. Smith, W. A. Stygar, and R. A. Vesey

Sandia National Laboratories, P.O. Box 5800, Albuquerque, New Mexico 87185, USA
(Received 18 June 2014; published 6 October 2014)

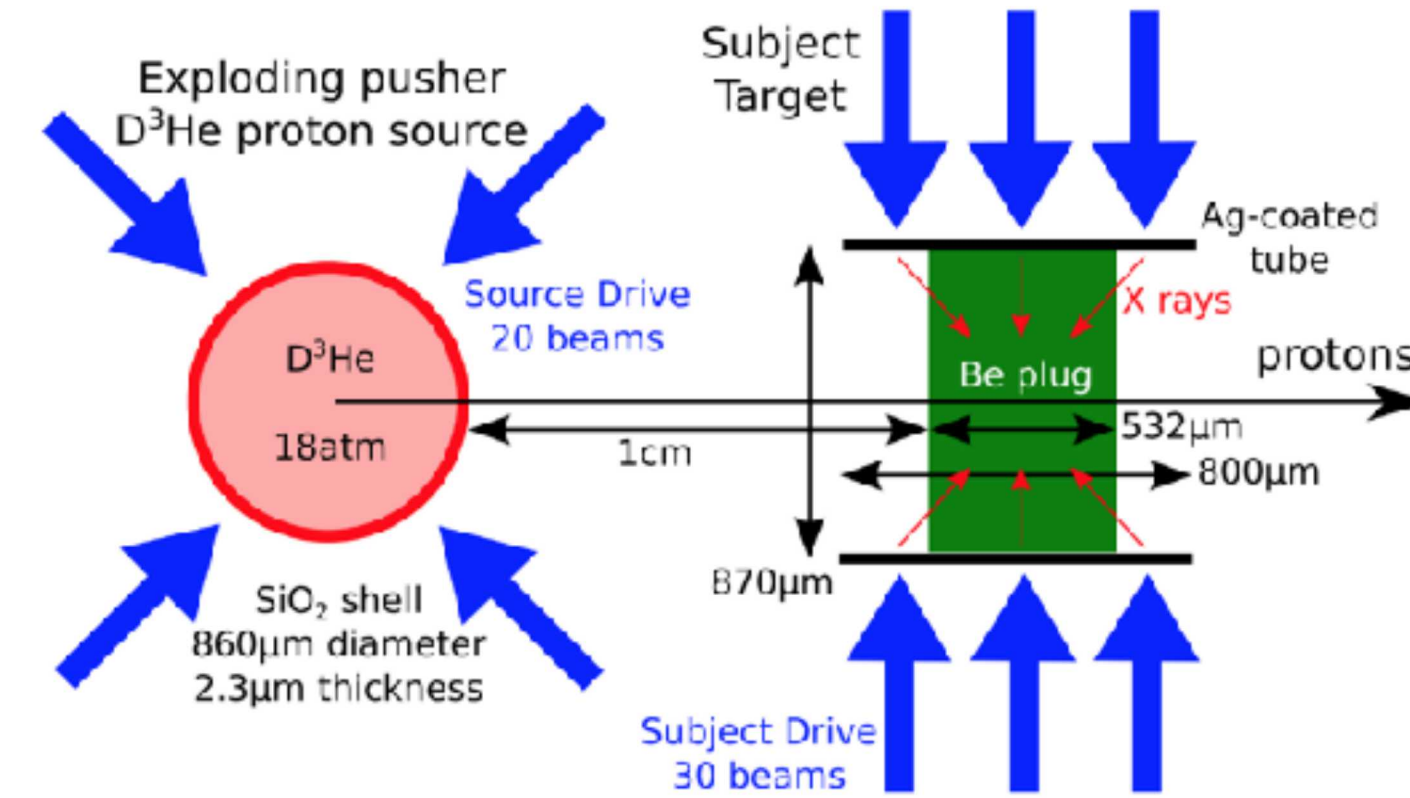
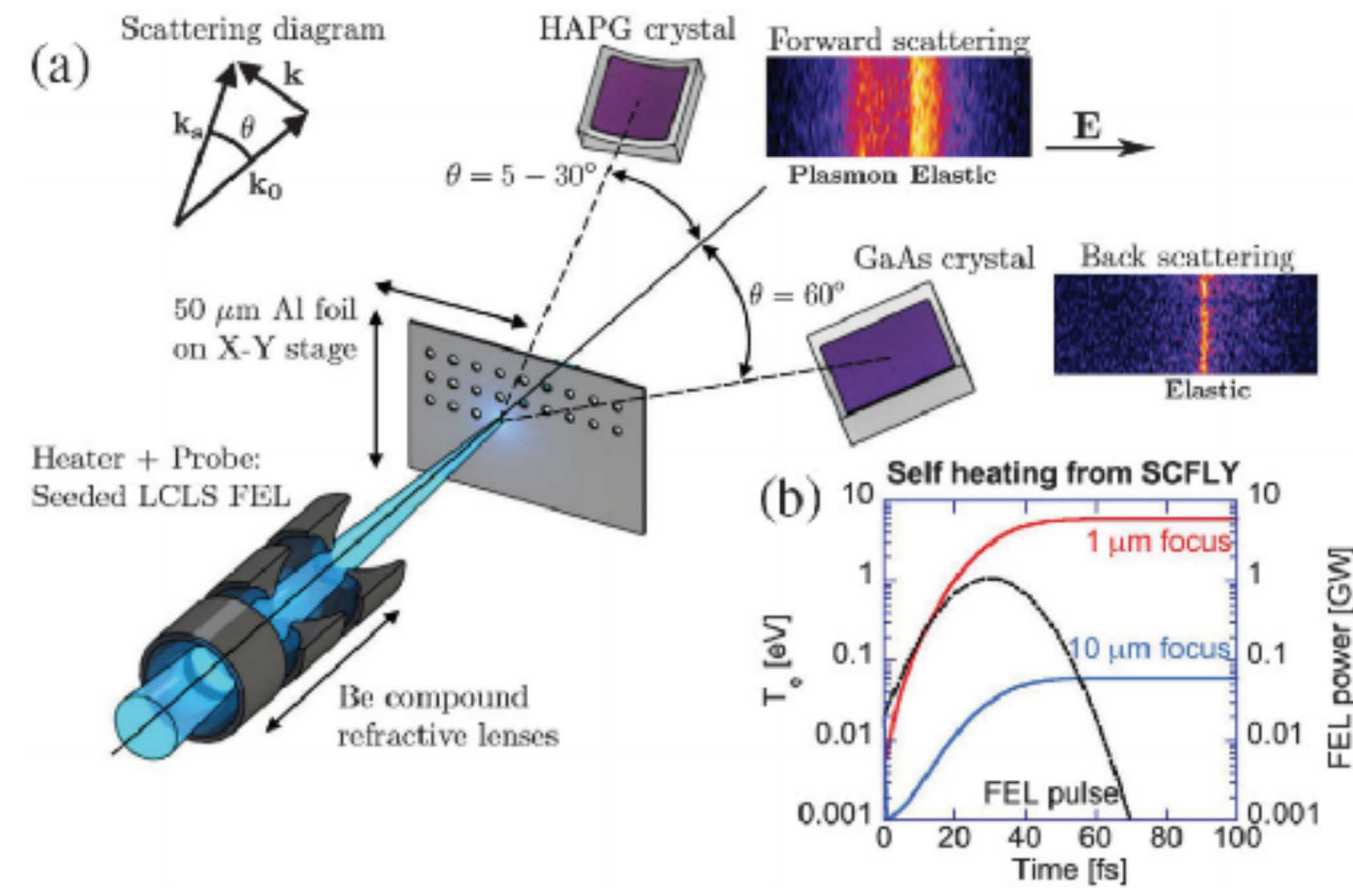


Experimental design requires **equation-of-state** and **transport properties**
(among many other things)

What is *experiment* telling *experiment*?

Measurement of Charged-Particle Stopping in Warm Dense Plasma

A. B. Zylstra, J. A. Frenje, P. E. Grabowski, C. K. Li, G. W. Collins, P. Fitzsimmons, S. Glenzer, F. Graziani, S. B. Hansen, S. X. Hu, M. Gatu Johnson, P. Keiter, H. Reynolds, J. R. Rygg, F. H. Séguin, and R. D. Petrasso
 Phys. Rev. Lett. **114**, 215002 – Published 27 May 2015



Free-Electron X-Ray Laser Measurements of Collisional-Damped Plasmons in Isochorically Heated Warm Dense Matter

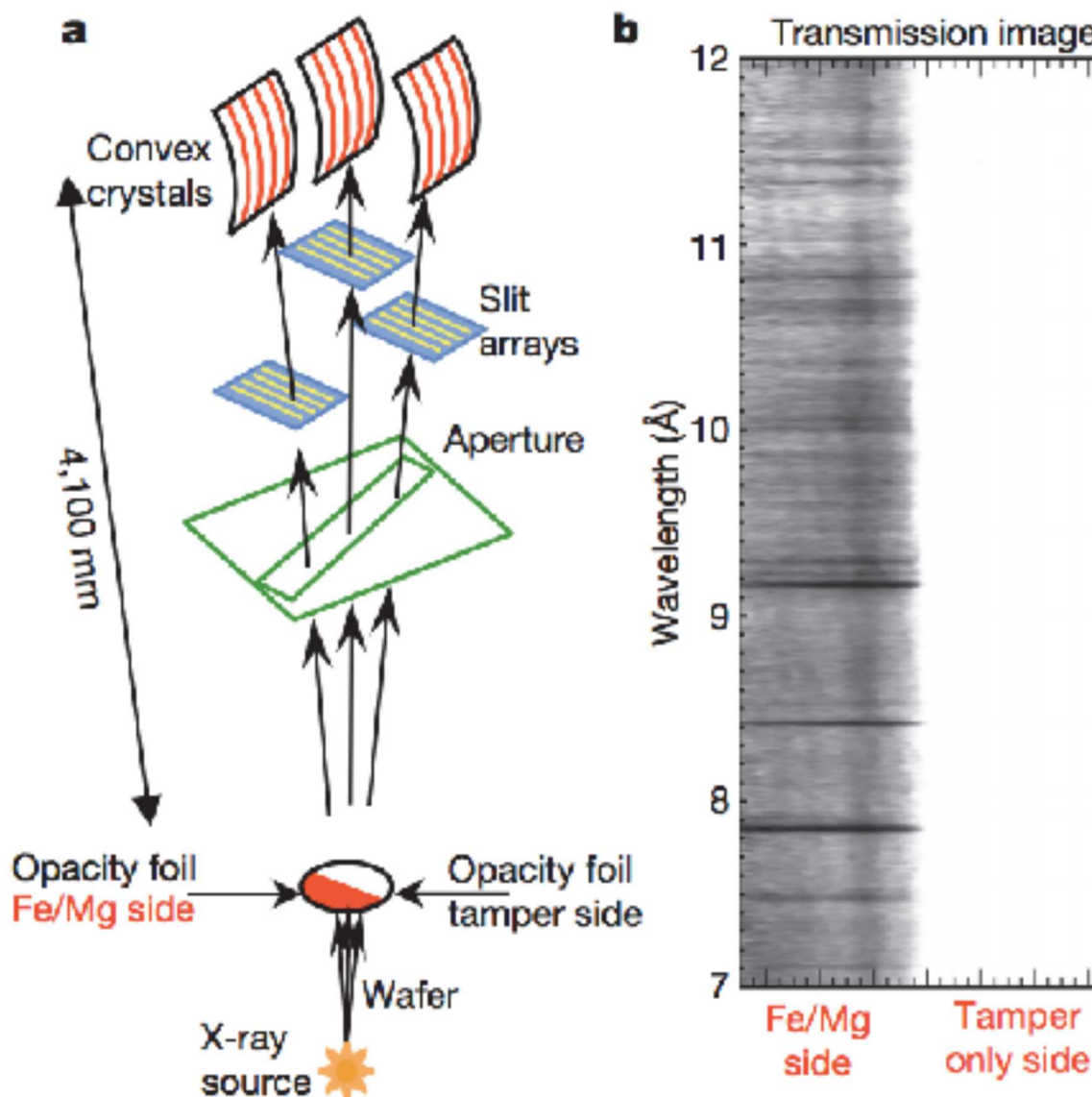
P. Sperling, E. J. Gamboa, H. J. Lee, H. K. Chung, E. Galtier, Y. Omarbakiyeva, H. Reinholz, G. Röpke, U. Zastra, J. Hastings, L. B. Fletcher, and S. H. Glenzer
 Phys. Rev. Lett. **115**, 115001 – Published 9 September 2015

LETTER

doi:10.1038/nature14048

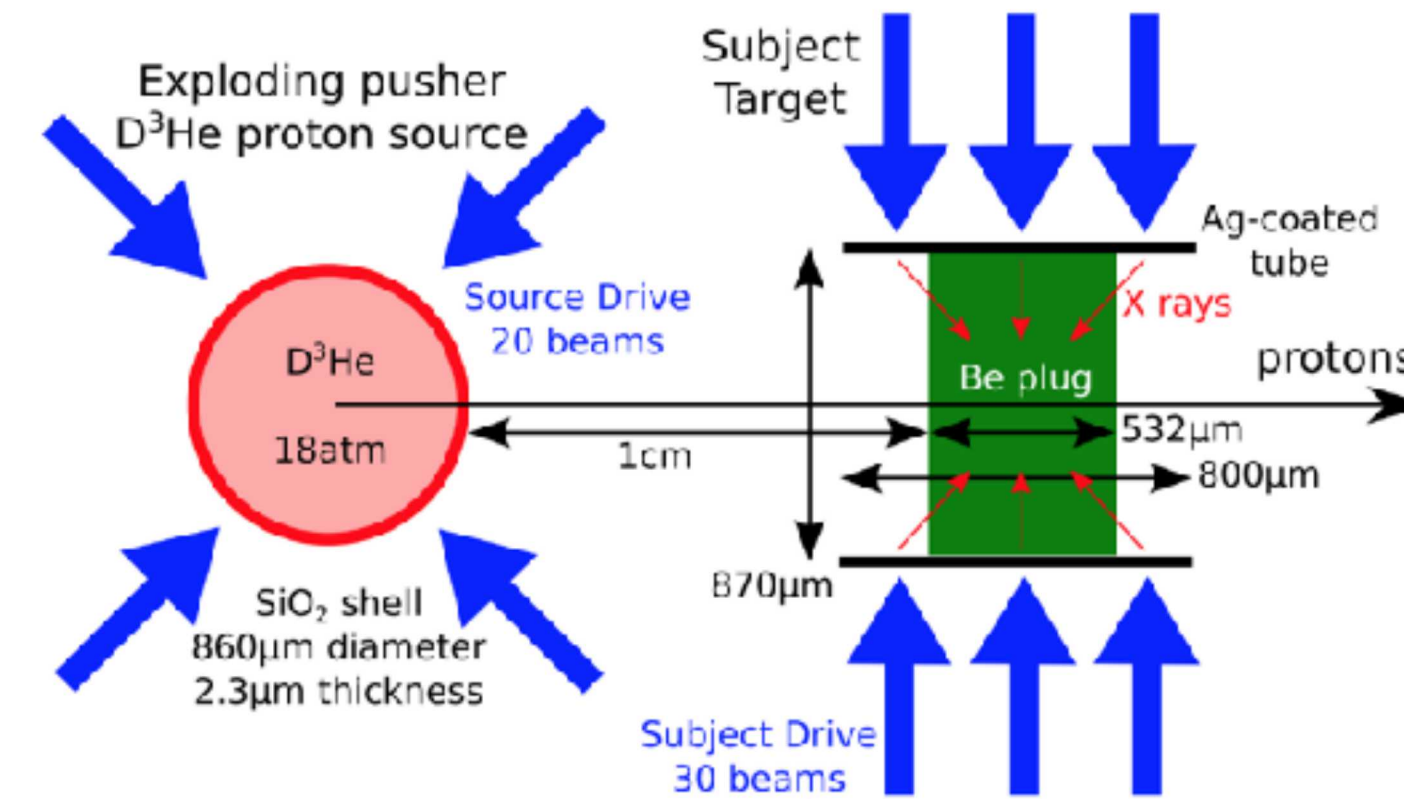
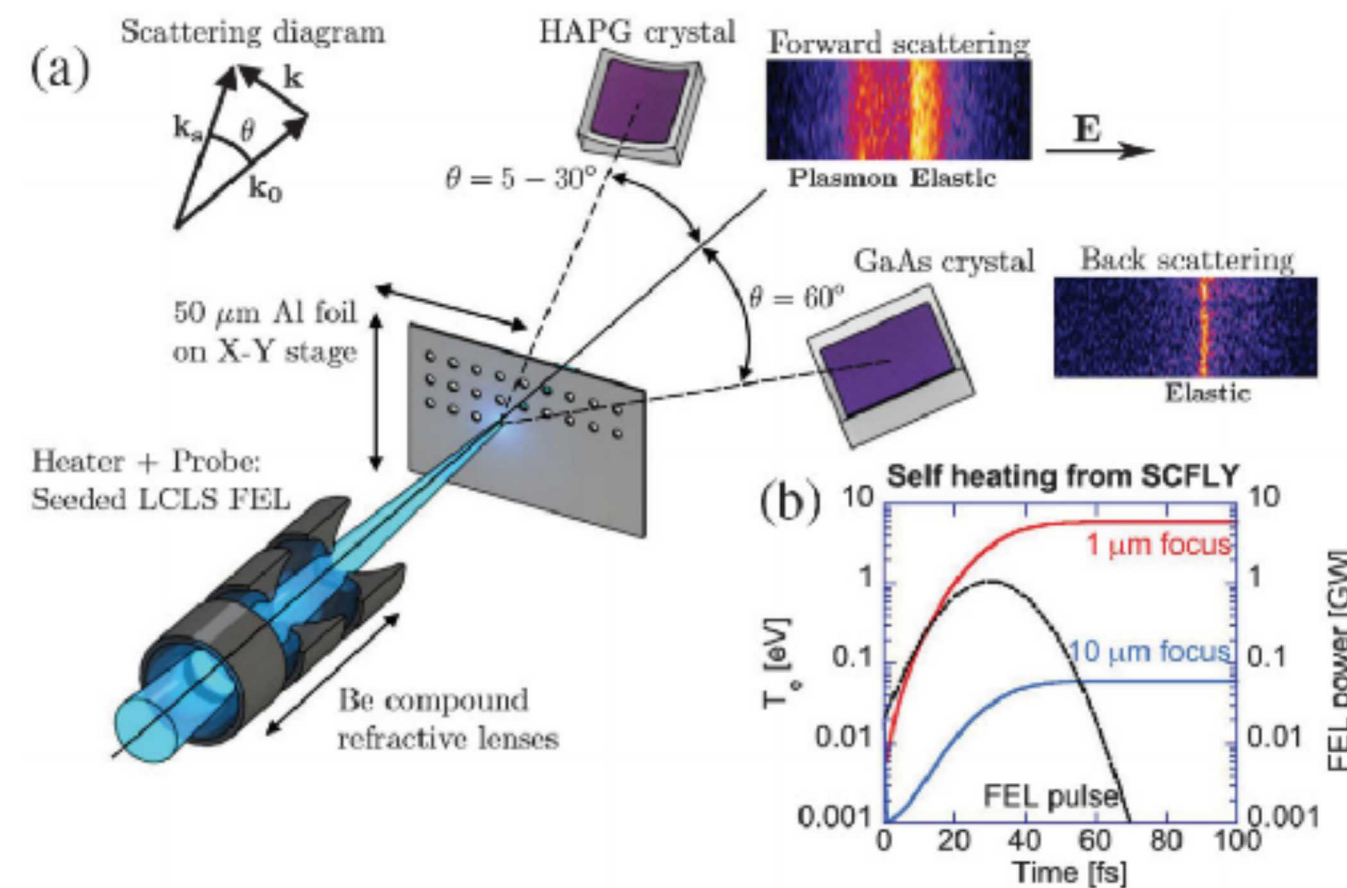
A higher-than-predicted measurement of iron opacity at solar interior temperatures

J. E. Bailey¹, T. Nagayama¹, G. P. Loisel¹, C. A. Rochau¹, C. Blancard², J. Colgan³, Ph. Cossé², G. Faussurier², C. J. Fontes³, F. Gillieron², I. Golovkin⁴, S. B. Hansen¹, C. A. Iglesias⁵, D. P. Kilcrease³, J. J. MacFarlane⁴, R. C. Mancini⁶, S. N. Nahar⁷, C. Orban⁷, J.-C. Pain², A. K. Pradhan⁷, M. Sherrill⁸ & B. G. Wilson⁹



What theory *could* tell experiment?

Stopping power:
Force that electrons in target apply to
projectile



Conductivity:
Linear response of current to applied
homogeneous vector potential

Dynamic structure factor:
Linear response of density to applied
inhomogeneous scalar potential

Real-Time TDDFT

...one such tool for computing response functions

What?

Density from Kohn-Sham orbitals

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

Non-interacting auxiliary system

$$i \frac{\partial}{\partial t} \psi_{n, \mathbf{k}}(\mathbf{r}, t) = \left[-\frac{\nabla^2}{2} + v_{KS}(\mathbf{r}, t) + v_{pert}(\mathbf{r}, t) \right] \psi_{n, \mathbf{k}}(\mathbf{r}, t)$$

Observables = density functionals

$$\langle \Psi(\mathbf{r}, t) | \hat{O} | \Psi(\mathbf{r}, t) \rangle = O[\rho(\mathbf{r}, t)]$$

Why?

No abuse of Kohn-Sham orbitals

(à la Kubo-Greenwood)

Better scaling than energy domain

(parallel and algorithmic)

Beyond linear response possible

(stopping power and more)

Our Implementation

TDDFT + Ehrenfest, initial implementation in VASP*

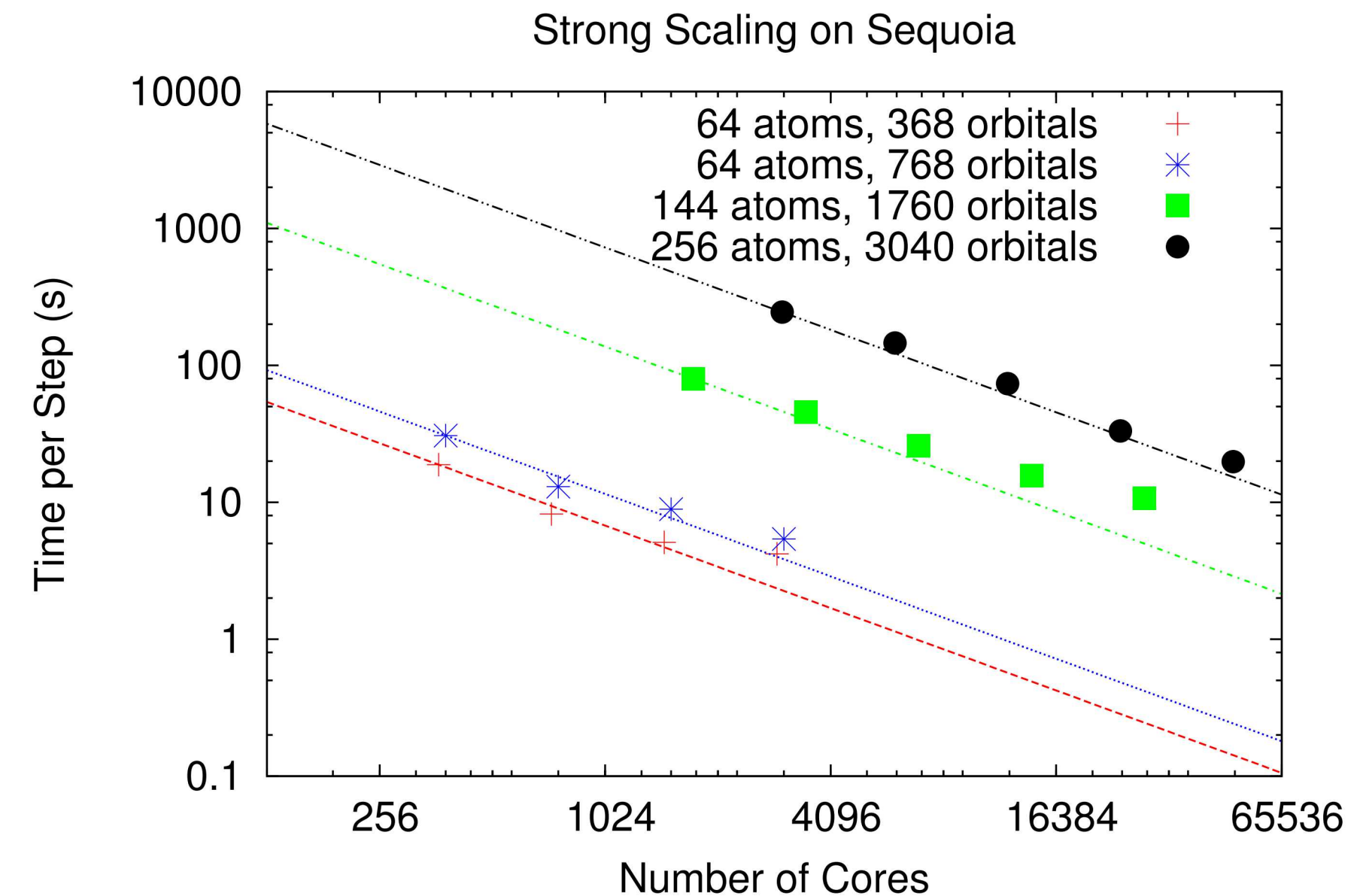
Recently teamed up with Michele Pavanello's QE fork, eQE**

- Self-contained code
- Crank-Nicolson time integrator
 - CGS and GMRES solvers
 - Non-Hermitian corrections for charge conservation
- Correct Ehrenfest forces for PAW

Nonadiabatic Ehrenfest molecular dynamics within the projector augmented-wave method

The Journal of Chemical Physics **136**, 144103 (2012); <https://doi.org/10.1063/1.3700800>

Ari Ojanpera¹, Ville Havu¹, Lauri Lehtovaara², and Martti Puska¹



Scaling for Xe (pathological)

Shock Compression of a Fifth Period Element: Liquid Xenon to 840 GPa

Seth Root, Rudolph J. Magyar, John H. Carpenter, David L. Hanson, and Thomas R. Mattsson
Phys. Rev. Lett. **105**, 085501 – Published 17 August 2010

*so, we can't share our source...

**...but we're working on it

Scenic Interlude I

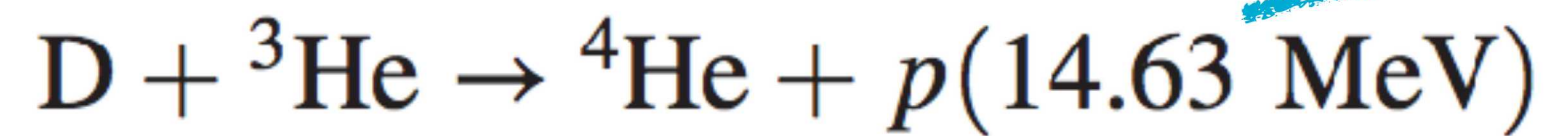
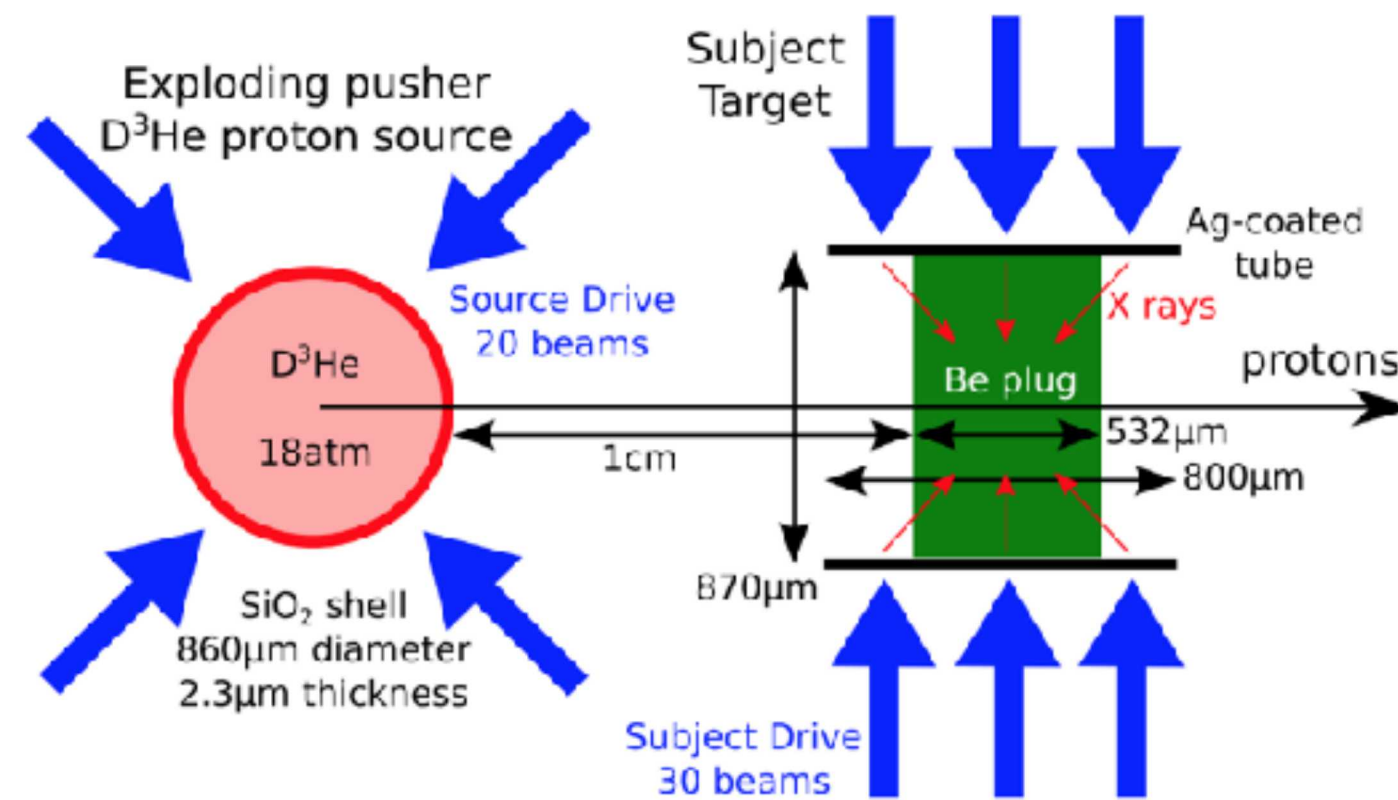


El Diente (Colorado, USA)

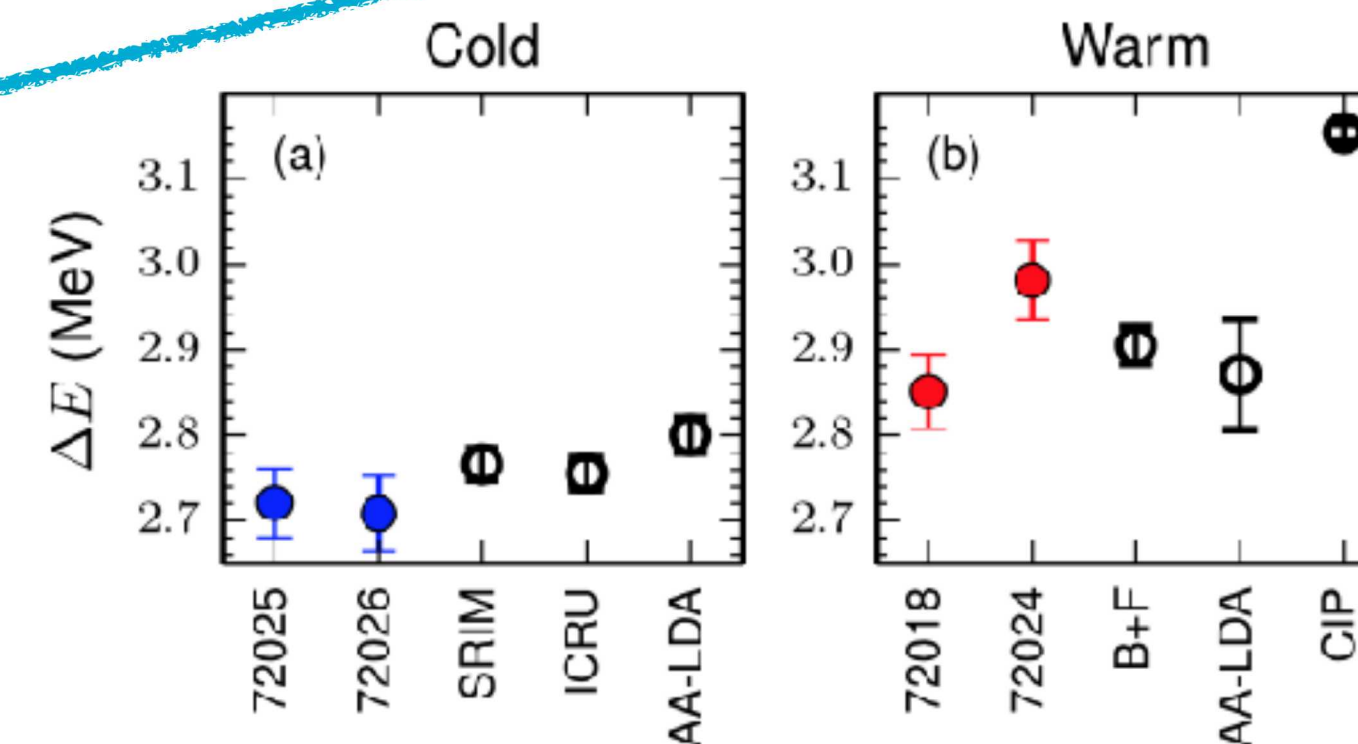
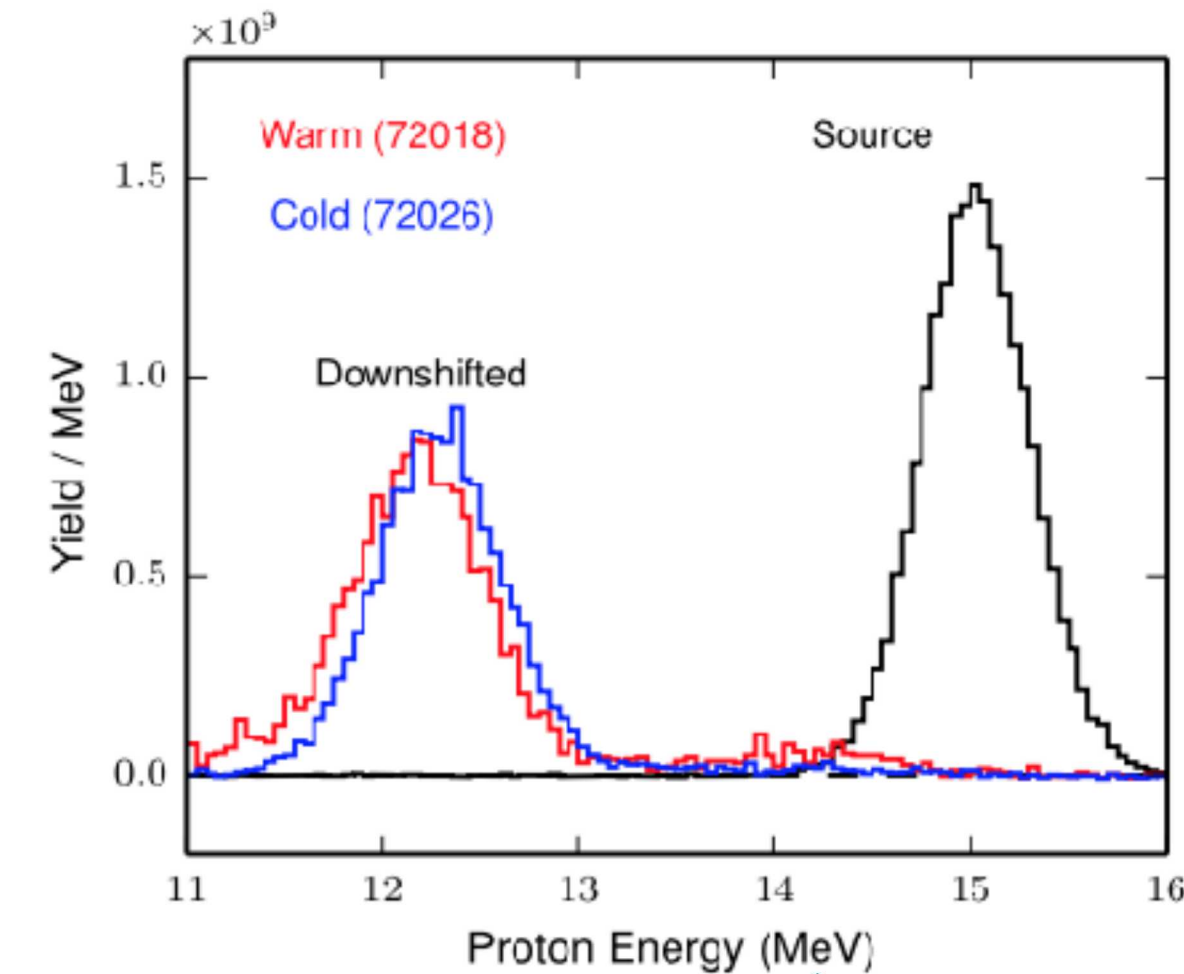
Electronic Stopping Power

Measurement of Charged-Particle Stopping in Warm Dense Plasma

A. B. Zylstra, J. A. Frenje, P. E. Grabowski, C. K. Li, G. W. Collins, P. Fitzsimmons, S. Glenzer, F. Graziani, S. B. Hansen, S. X. Hu, M. Gatu Johnson, P. Keiter, H. Reynolds, J. R. Rygg, F. H. Séguin, and R. D. Petrasso
 Phys. Rev. Lett. **114**, 215002 – Published 27 May 2015



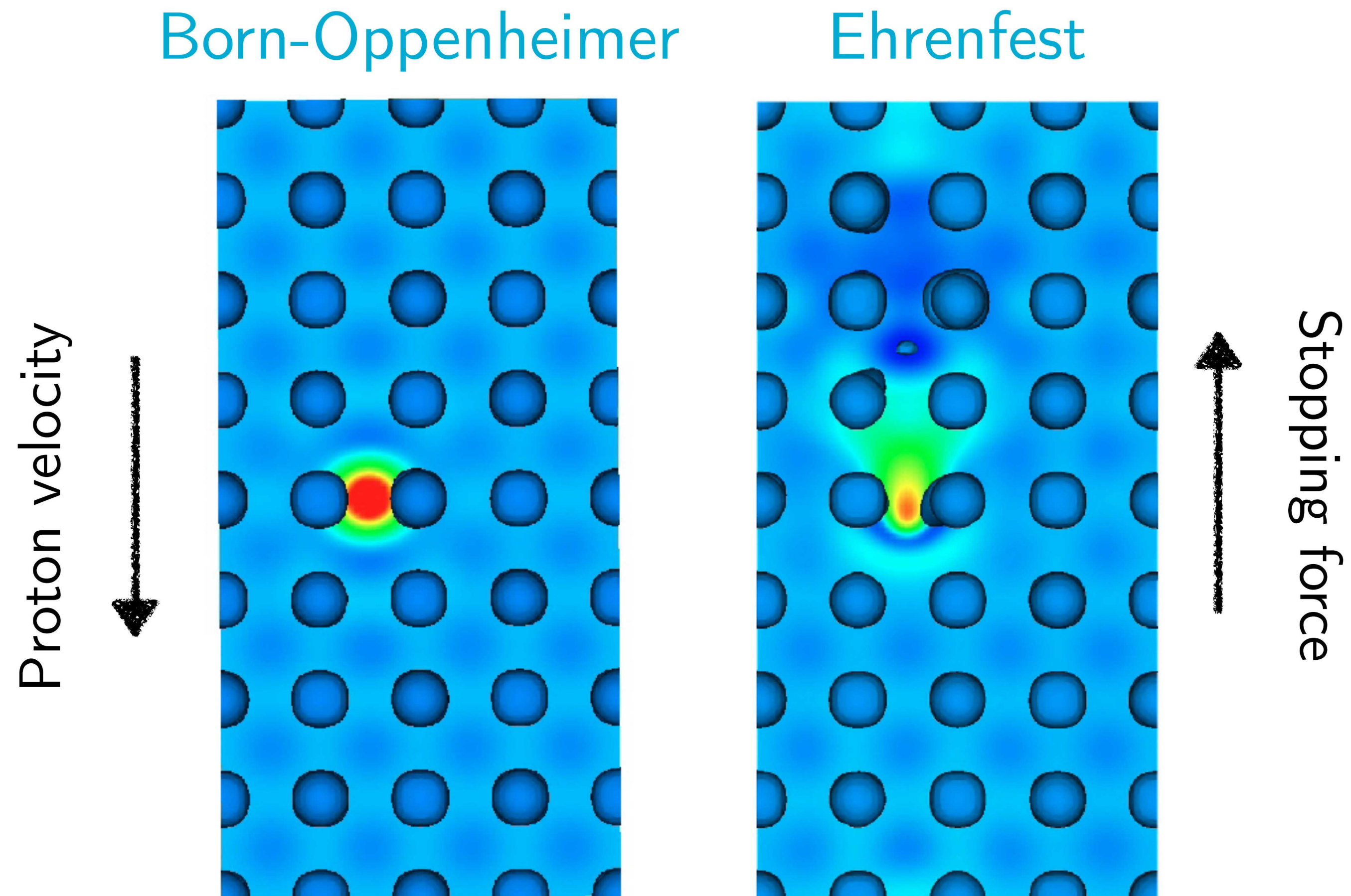
What is the **energy-dependent force** on a **proton** traversing an **isochorically heated Be plasma near 32 eV**?



approx.
 0.5 eV/Ang.
 or 0.01 a.u.

TDDFT: 10 hours on 160,000 CPUs my error bar is bigger than the y-axis

(At least) 240,000 Words*

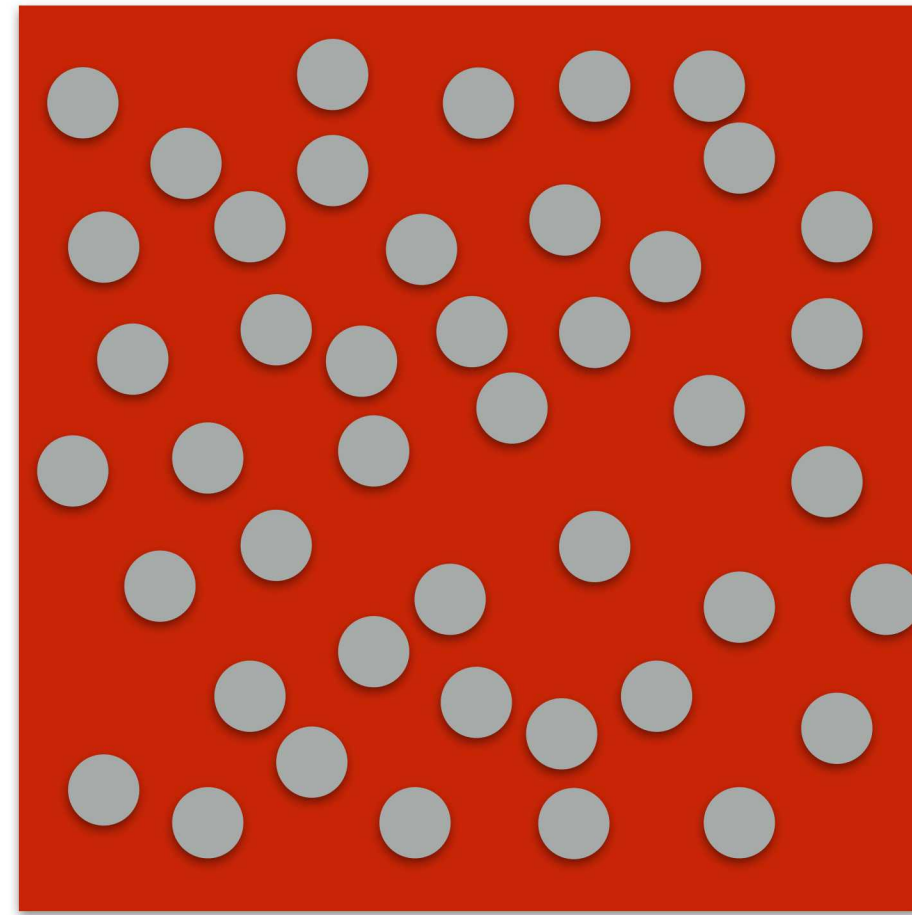


Average force is zero on the left
Average force agrees well with SRIM on the right

*20 pictures per second x 6 seconds x 2 movies x 1,000 words/picture

Ehrenfest-TDDFT

Start in a Mermin-Kohn-Sham equilibrium state

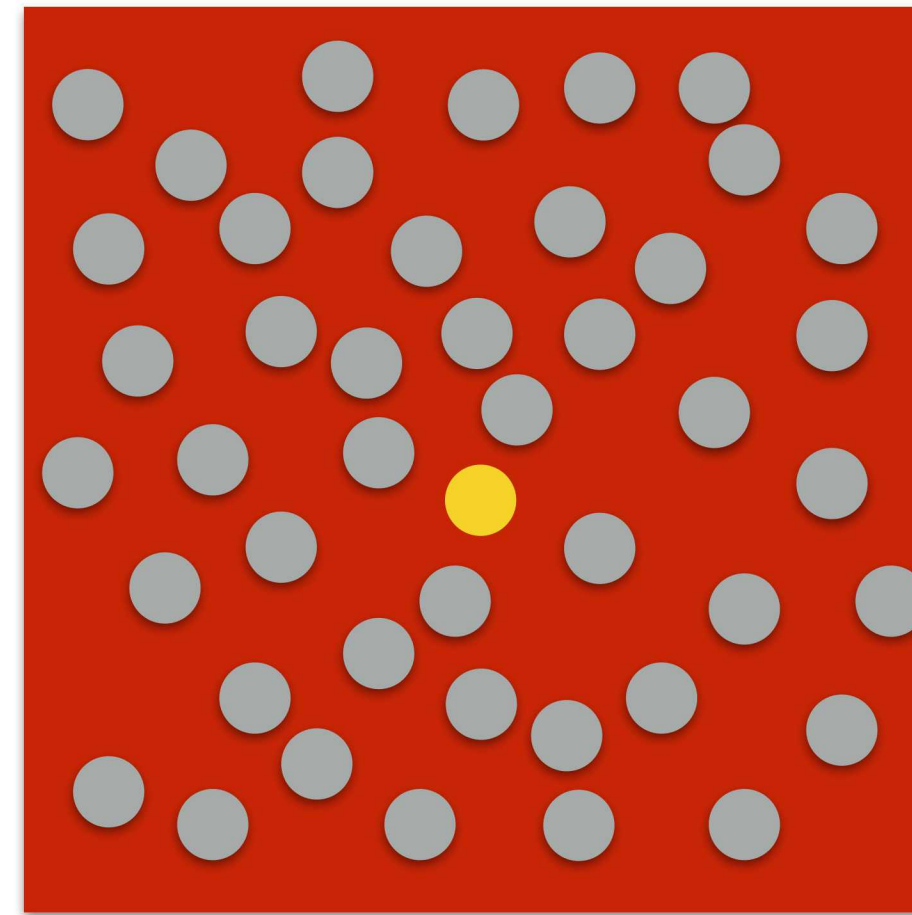


$$\rho_0(\mathbf{r}) = \sum_{n,\mathbf{k}} f_{n,\mathbf{k}}(T_e) |\psi_{n,\mathbf{k}}(\mathbf{r})|^2$$

Ehrenfest-TDDFT

Add a **projectile**, re-compute MKS state

This is the initial condition for **time-dependent Kohn-Sham system**

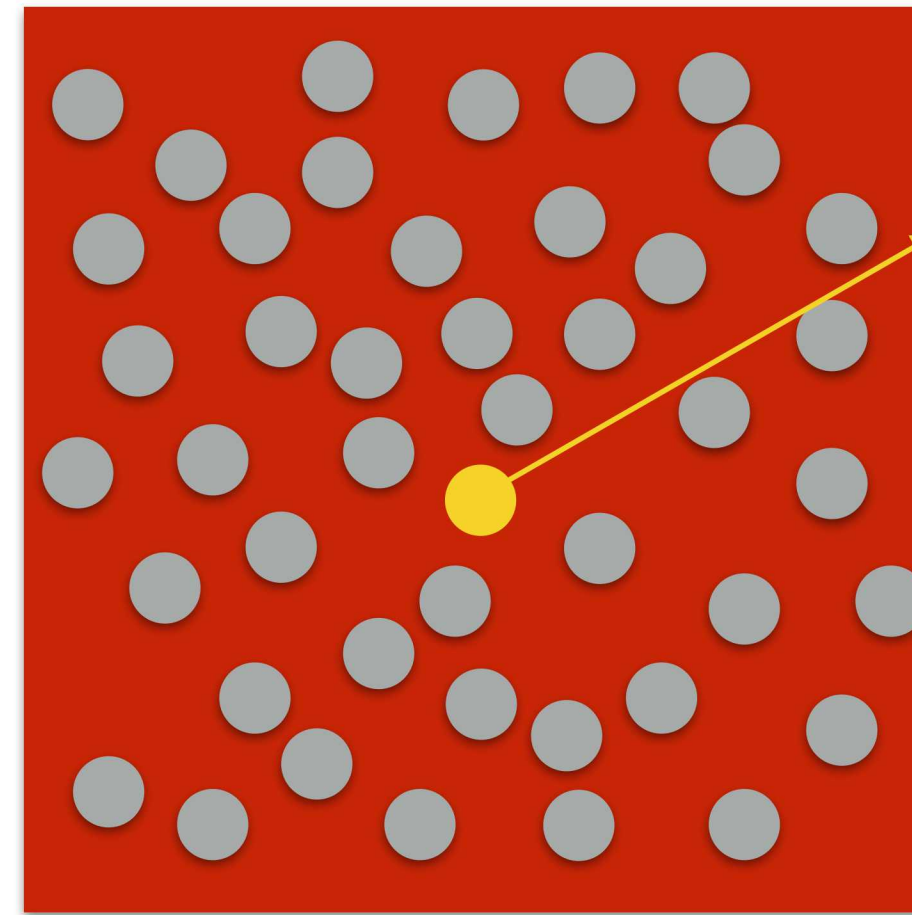


$$i\frac{\partial}{\partial t}\psi_{n,\mathbf{k}}(\mathbf{r},t) = \left[-\frac{\nabla^2}{2} + v_{KS}(\mathbf{r},t) + v_{pert}(\mathbf{r},t) \right] \psi_{n,\mathbf{k}}(\mathbf{r},t)$$

Ehrenfest-TDDFT

Drag the projectile along at the **velocity of interest**

Compute the force retarding the projectile

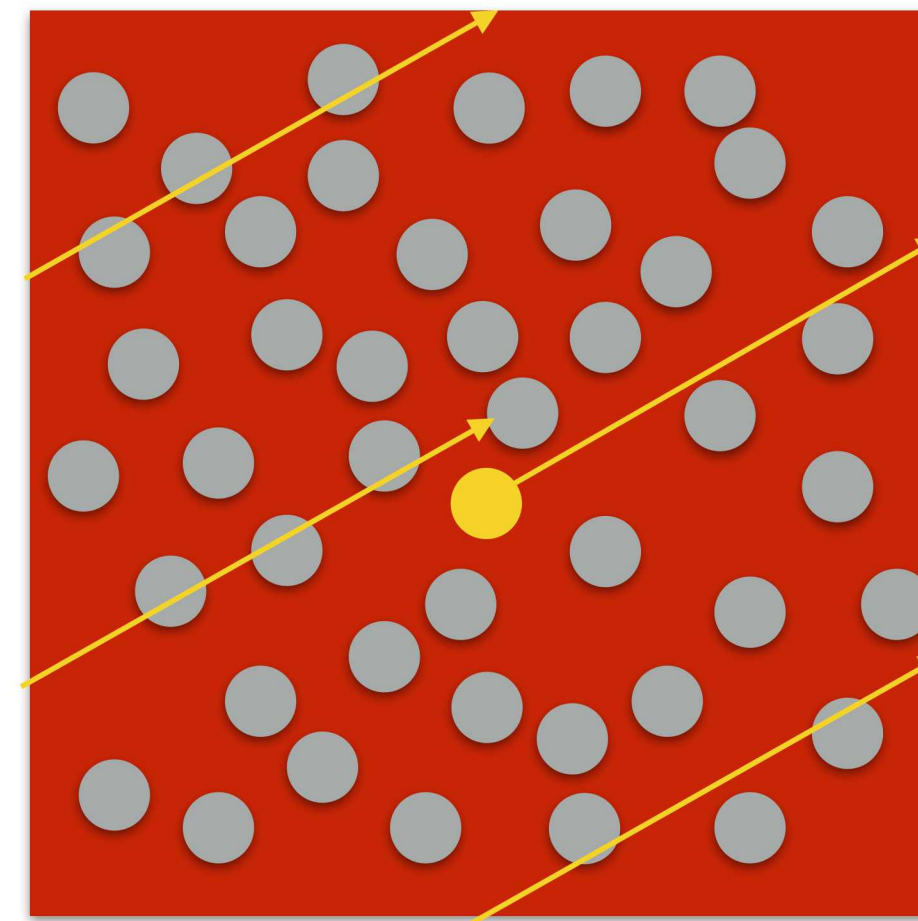


$$\frac{\delta \mathcal{A}}{\delta \mathbf{R}_i} = 0 \rightarrow M_i \frac{d^2 \mathbf{R}_i}{dt^2} = \frac{\partial \mathcal{L}_q}{\partial \mathbf{R}_i} - \frac{d}{dt} \frac{\partial \mathcal{L}_q}{\partial \frac{d\mathbf{R}_i}{dt}}$$

$$\mathcal{L}_q = \sum_{n, \mathbf{k}} f_{n, \mathbf{k}}(T_e) \langle \psi_{n, \mathbf{k}} | i \frac{d}{dt} - \frac{\nabla^2}{2} | \psi_{n, \mathbf{k}} \rangle - \mathcal{L}_{a, Hxc} \{ \rho(\mathbf{r}, t) \}$$

Ehrenfest-TDDFT

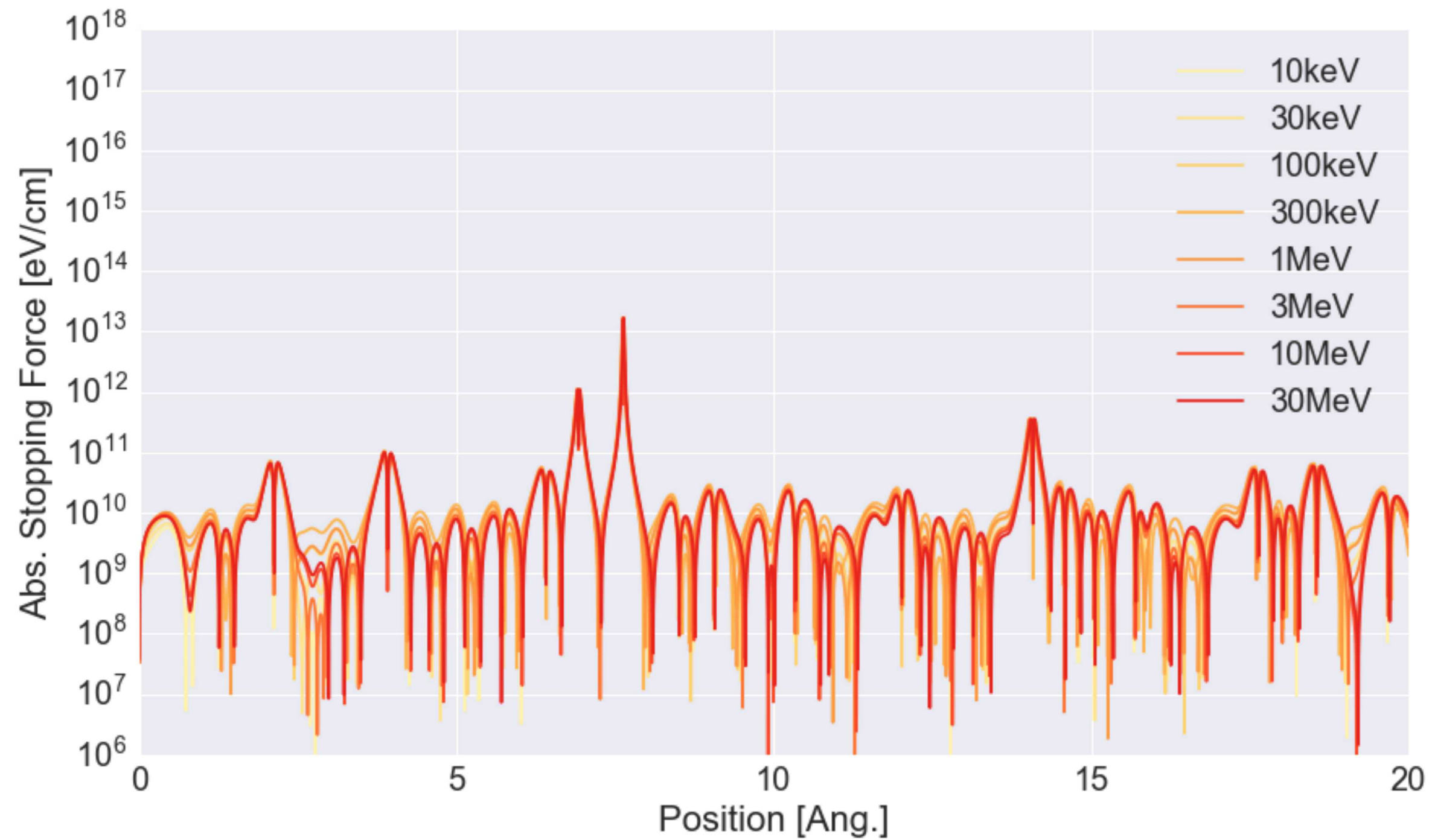
Collect as much data as you can
(100 Angstrom trajectories typically feasible)



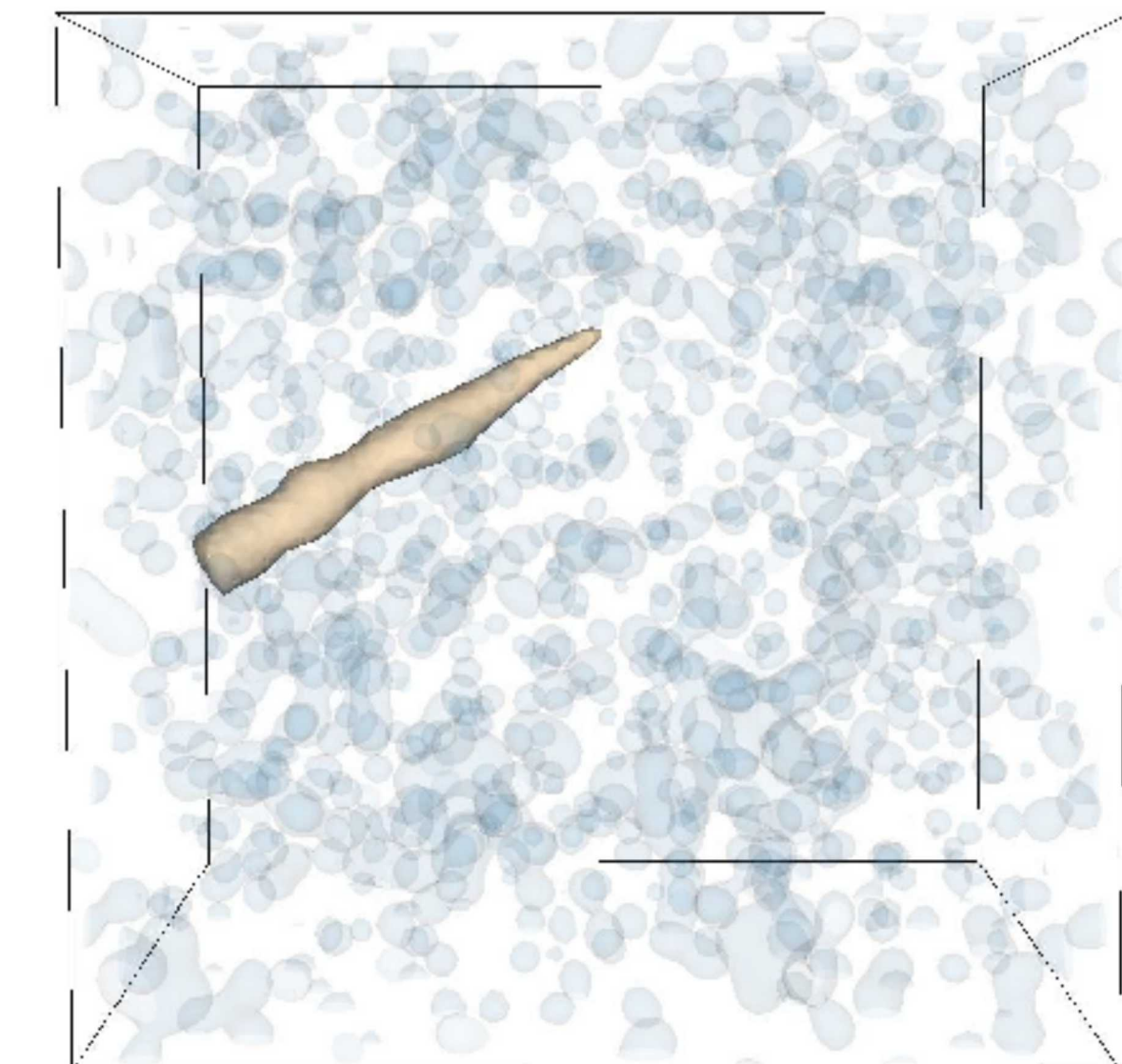
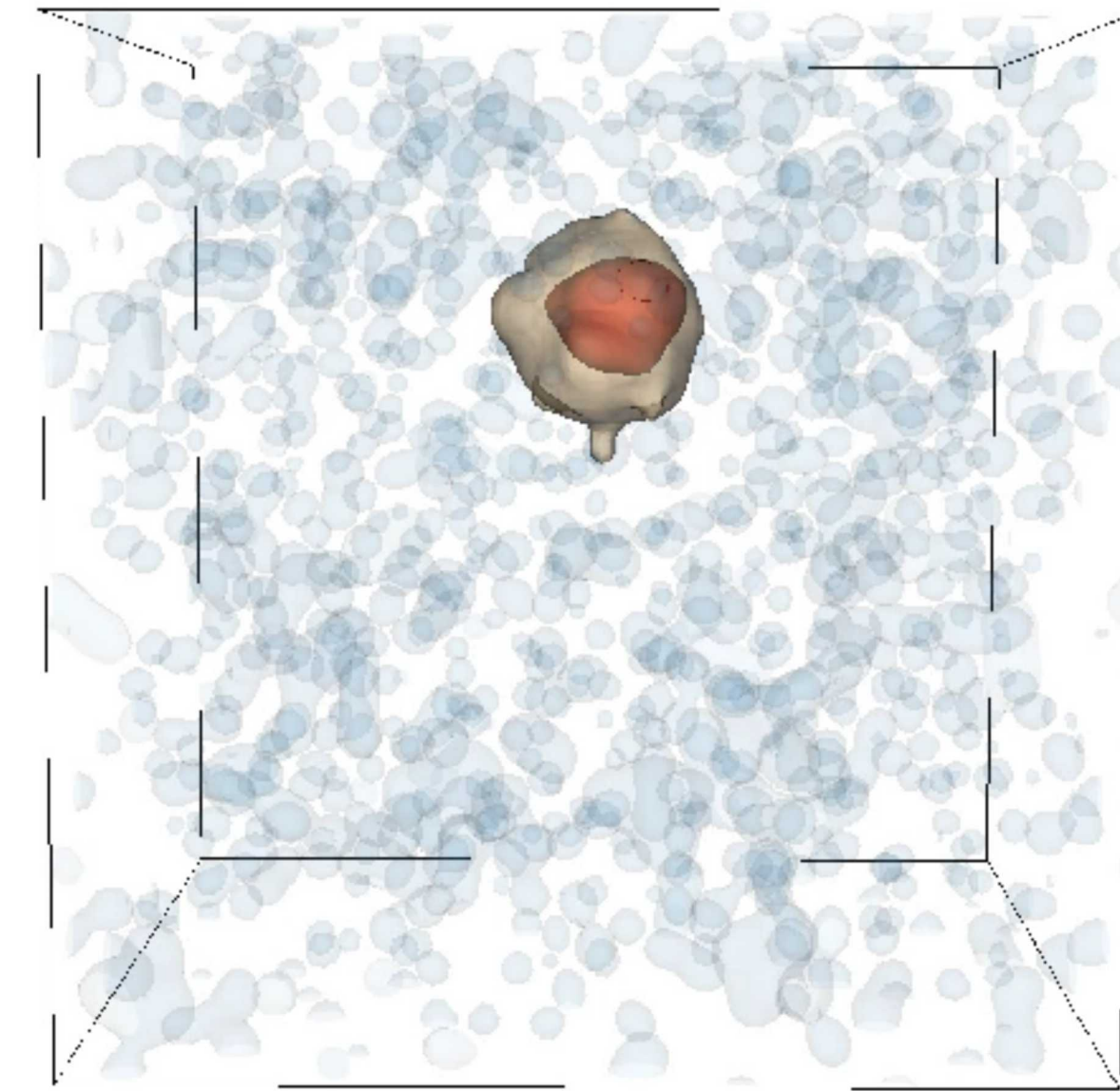
Others have done something similar in condensed matter,
Artacho (Cambridge), Correa (LLNL), Kohanoff (Belfast), Schleife (UIUC), and many more
We are looking at higher energies and temperatures

Warm Dense Deuterium

Stopping at **10 g/cc** and **2 eV**
(dead center of the DOE/Bonitz diagram)

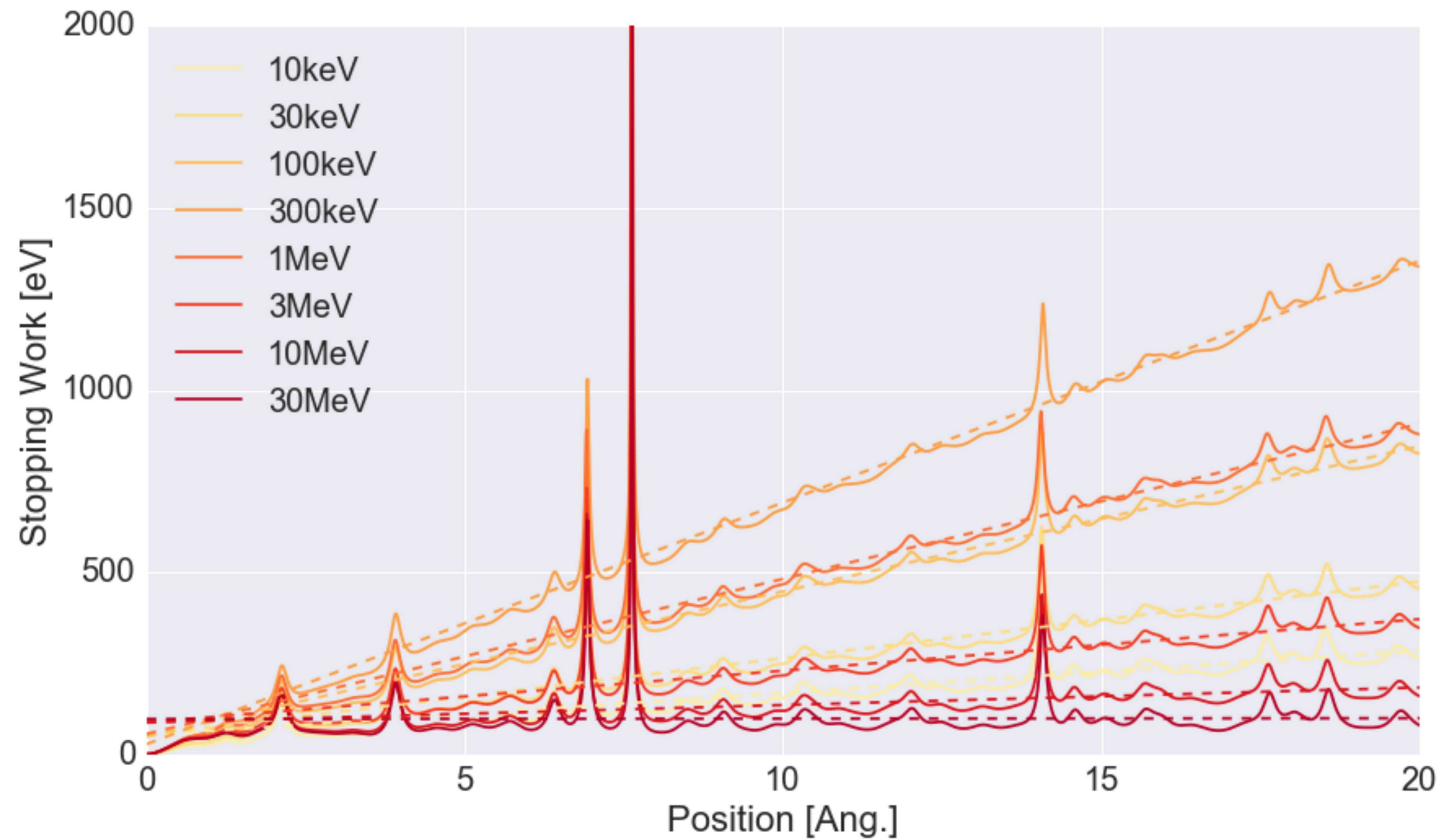


Force on projectile vs. distance traversed
Hard to tell the difference between
300 keV (top) and 30 MeV (bottom)



Warm Dense Deuterium

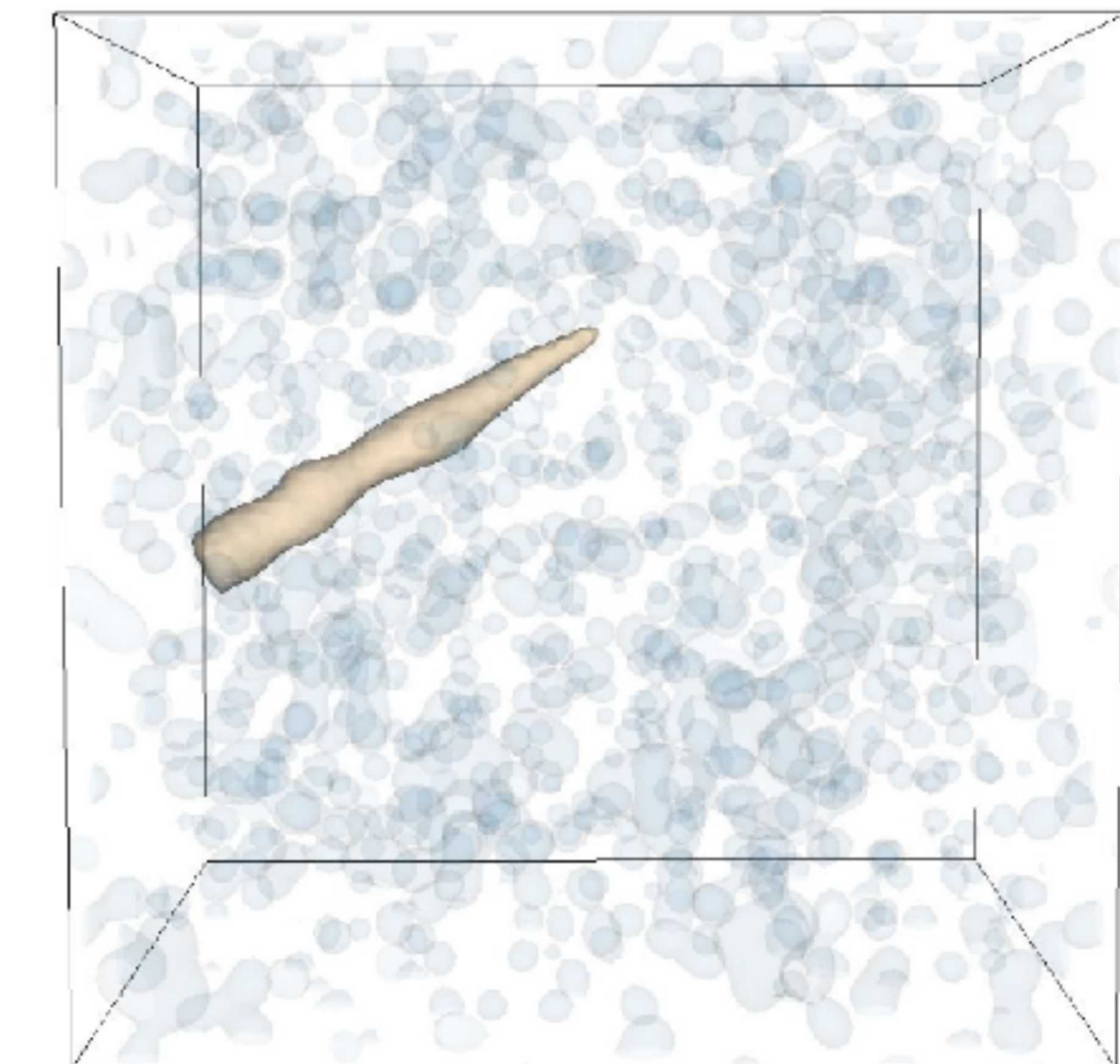
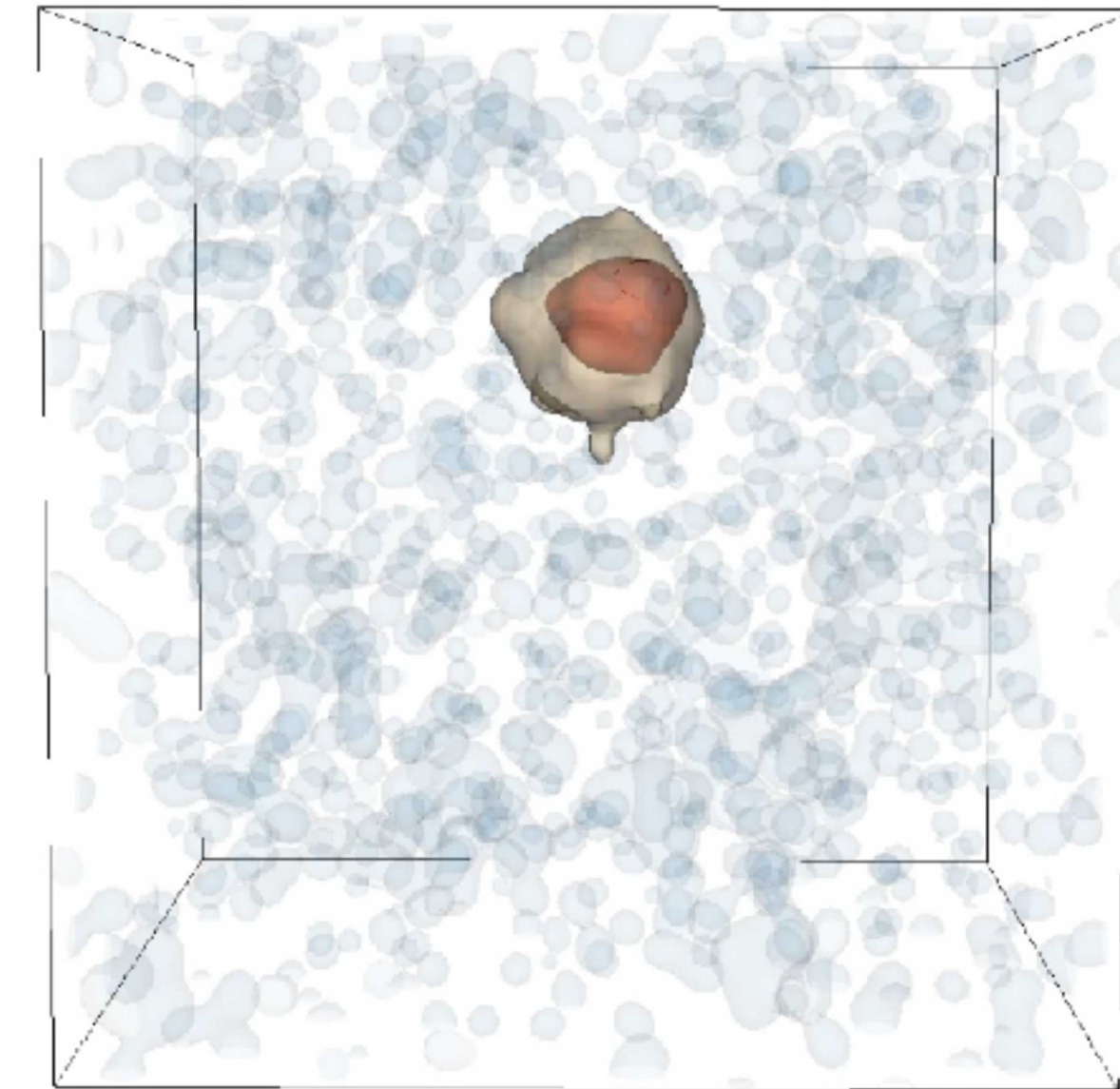
Stopping at 10 g/cc and 2 eV
(dead center of the DOE/Bonitz diagram)



Work on projectile vs. distance traversed

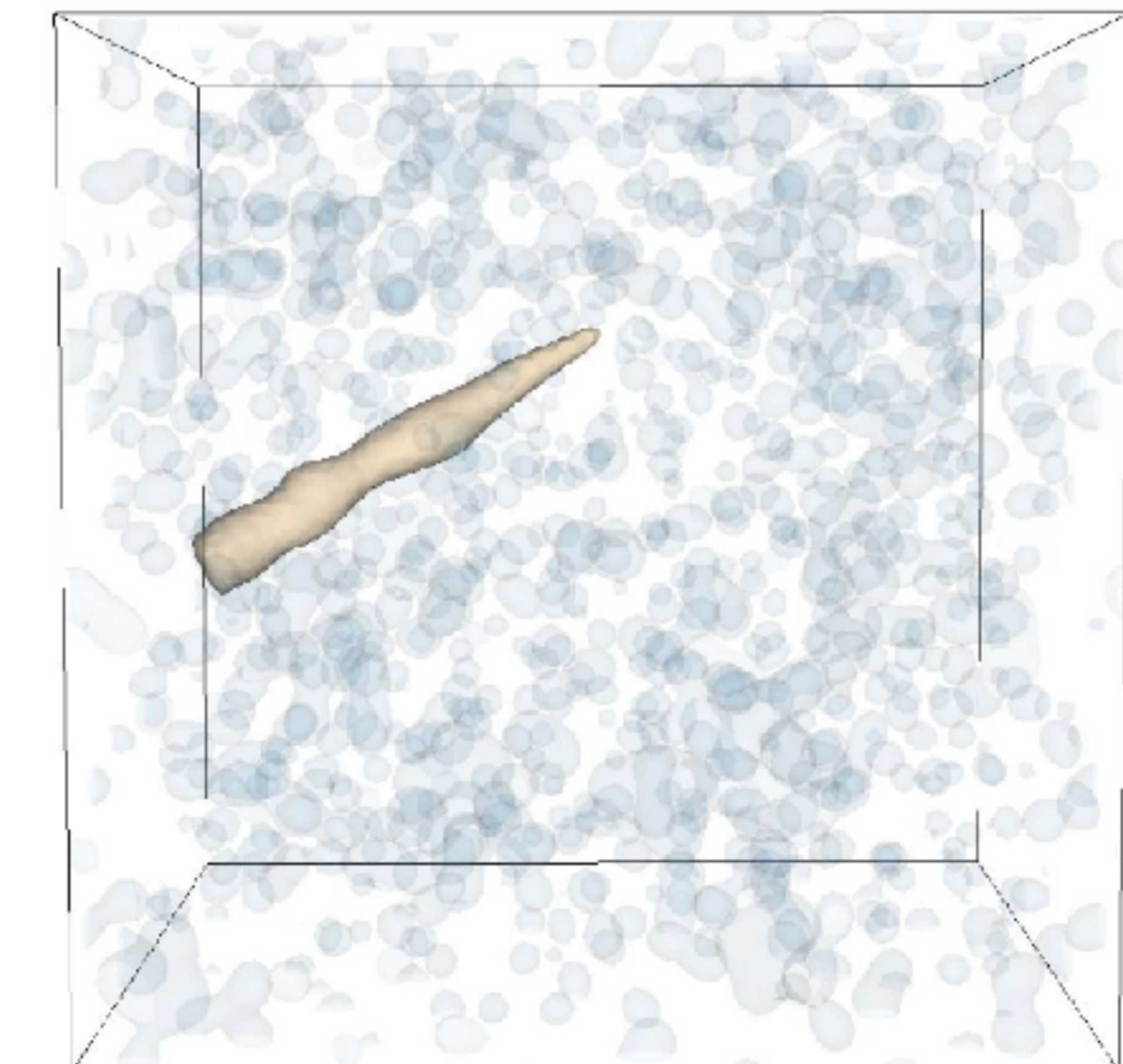
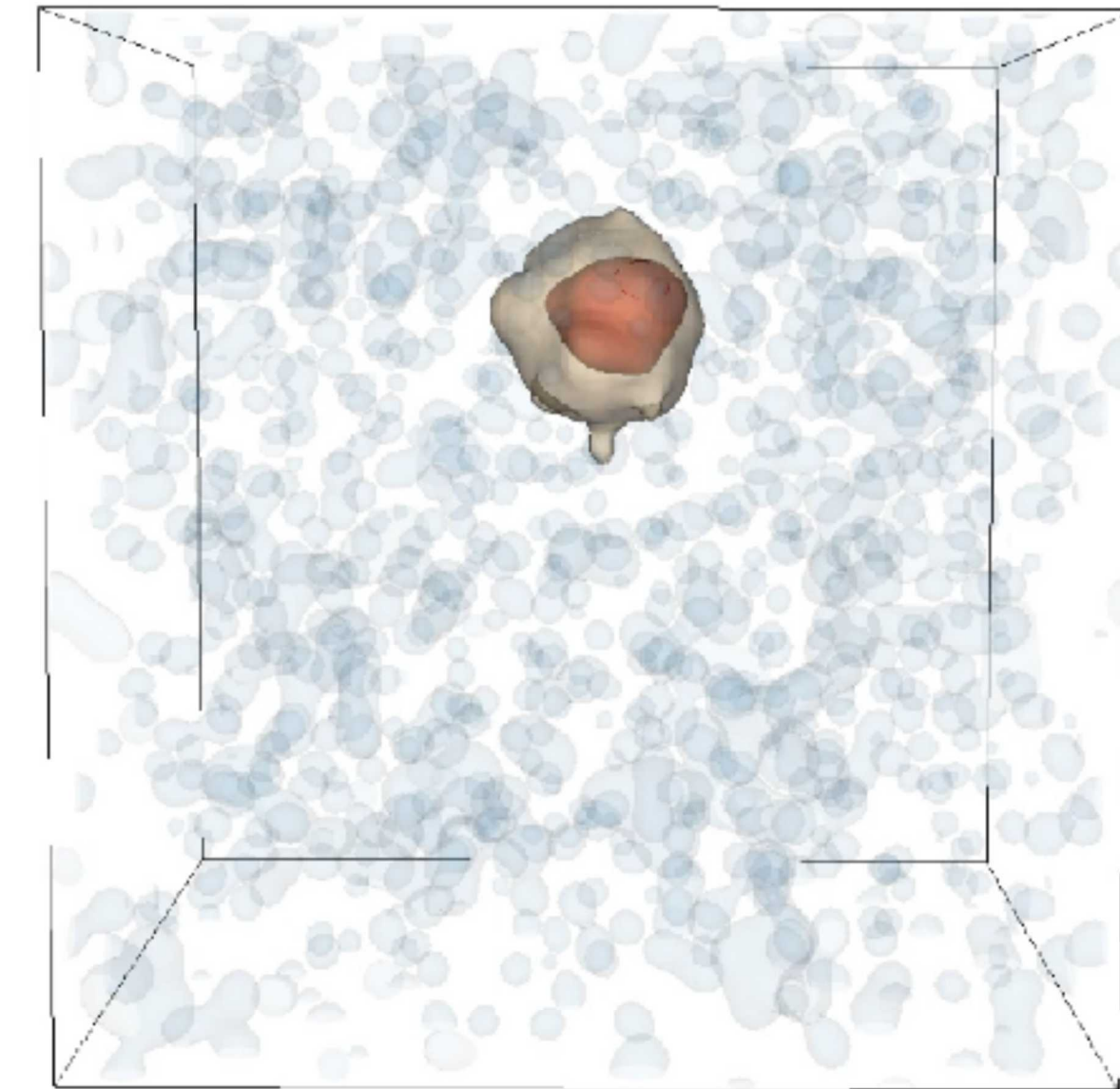
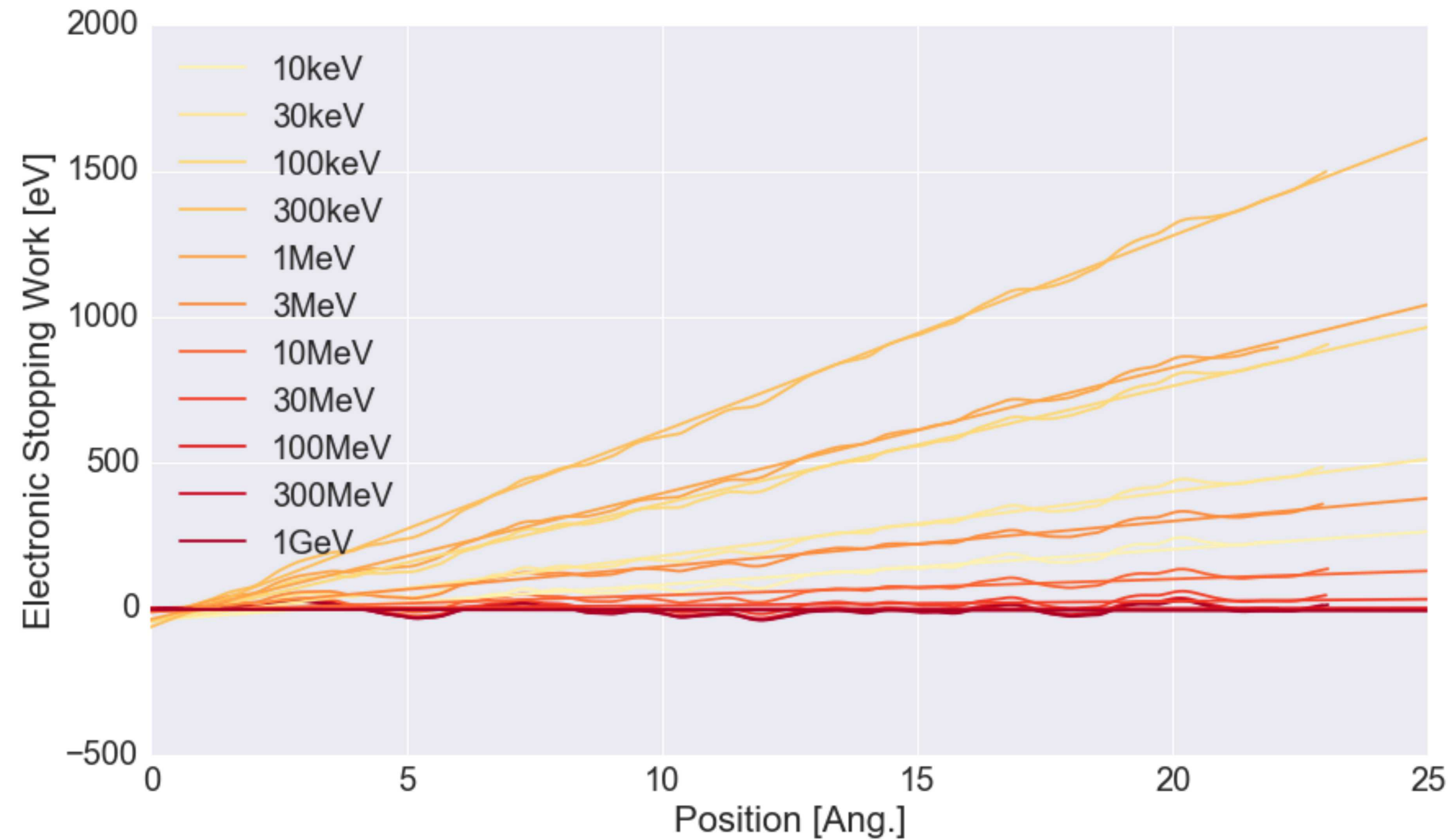
Spikes = close passage to ions

Linear slope = stopping power



Warm Dense Deuterium

Stopping at 10 g/cc and 2 eV
(dead center of the DOE/Bonitz diagram)

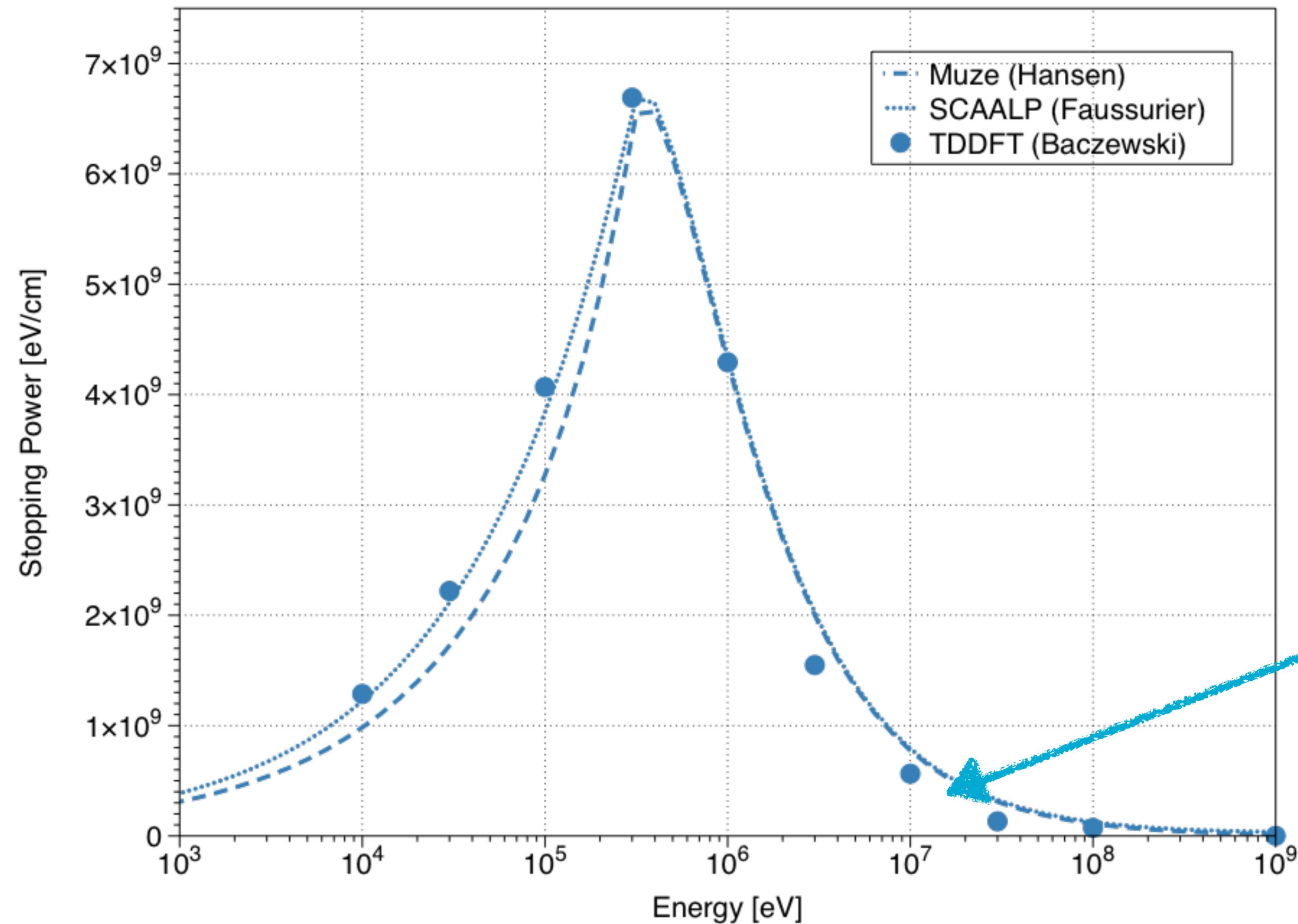


Work on projectile vs. distance traversed
Subtracting out the ion-ion force gives us a
purely electronic stopping power

Average Atom vs. Ehrenfest-TDDFT

Results from transport code comparison workshop at SNL

(note: still deuterium at 10 g/cc, 2 eV)



fusion products
used by Zylstra
style experiments

High energies: standard deviation of fore close to the mean, distribution is wide, non-Gaussian

Whither Ehrenfest-TDDFT for WDM?

il buono

For homogeneous systems accessible by average atom, TDDFT can corroborate empirical choices

We can look at inhomogeneous/mixed systems at keV energies (and we have)

There may be something interesting about force distributions, too*

il brutto

Fusion-relevant stopping power will be a challenge in Ehrenfest-TDDFT

il cattivo

This might also just be the wrong tool to answer this specific question

*I've been chatting with some plasma physicists working on 2-fluid models

Scenic Interlude II

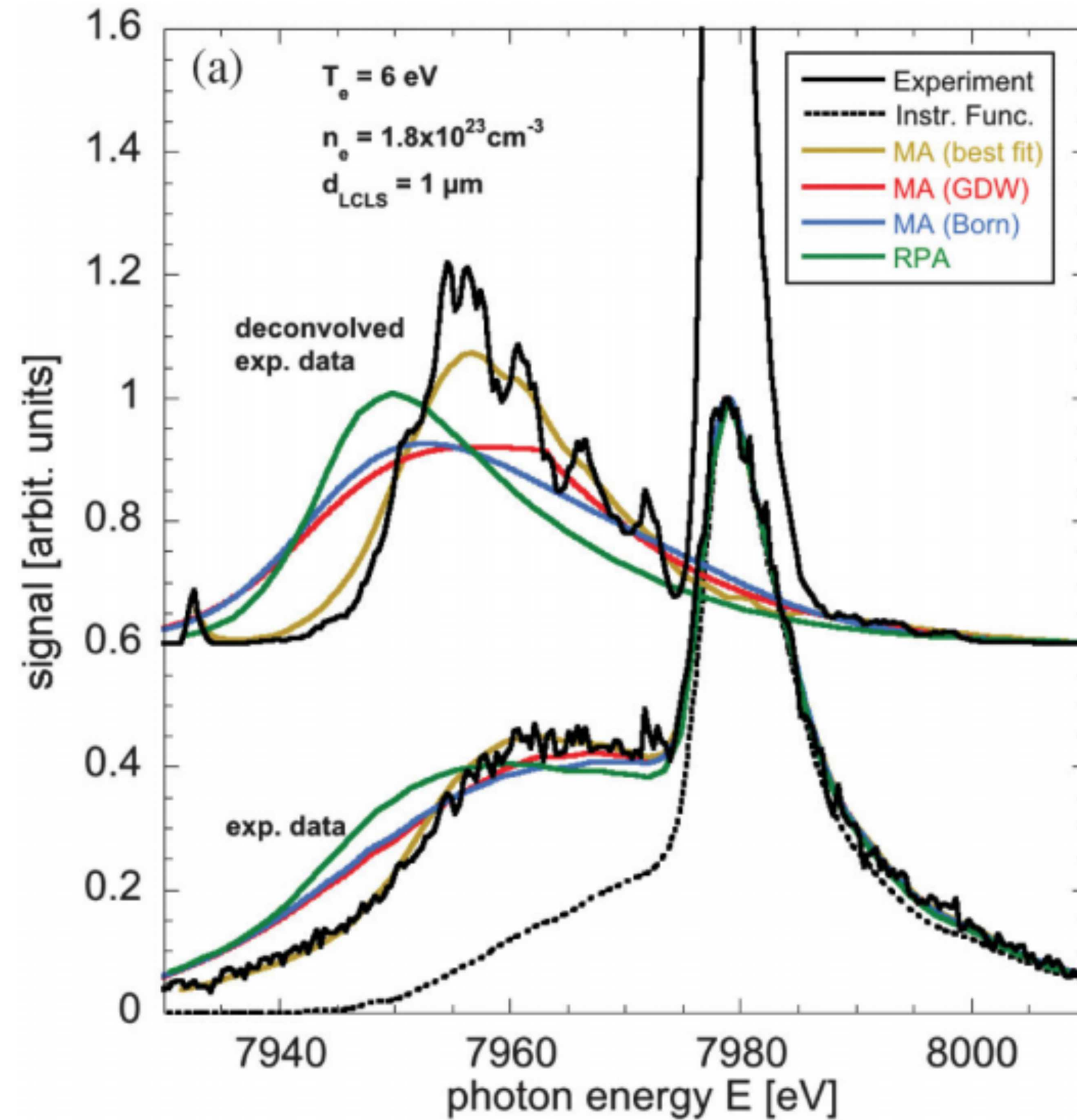


The Maroon Bells (Colorado, USA)

DC Conductivity*

Free-Electron X-Ray Laser Measurements of Collisional-Damped Plasmons in Isochorically Heated Warm Dense Matter

P. Sperling, E. J. Gamboa, H. J. Lee, H. K. Chung, E. Galtier, Y. Omarbakiyeva, H. Reinholz, G. Röpke, U. Zastra, J. Hastings, L. B. Fletcher, and S. H. Glenzer
 Phys. Rev. Lett. **115**, 115001 – Published 9 September 2015



X-ray Thomson scattering in high energy density plasmas

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

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

$$S(q, \omega) = -\frac{k^2}{4\pi^2 n_e} \frac{\text{Im} [\epsilon^{-1}(q, \omega)]}{1 - \exp(-\omega/T_e)}$$

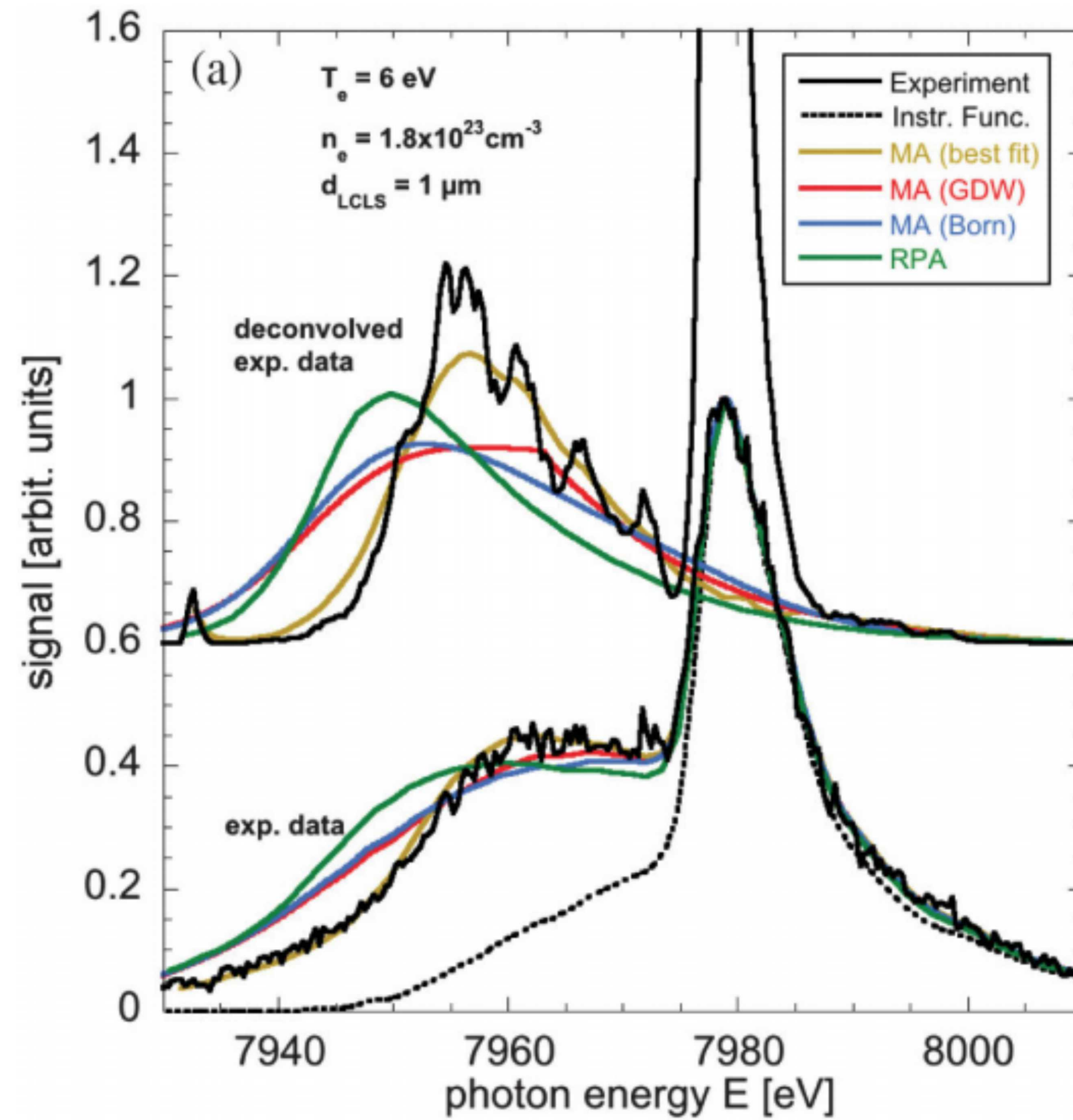
$$\epsilon^M(q, \omega) - 1 = \frac{\left(1 + i\frac{\nu(\omega)}{\omega}\right) [\epsilon^{RPA}(q, \omega + i\nu(\omega)) - 1]}{1 + i\frac{\nu(\omega)}{\omega} \frac{\epsilon^{RPA}(q, \omega + i\nu(\omega)) - 1}{\epsilon^{RPA}(q, 0) - 1}}$$

*by way of the dynamic structure factor

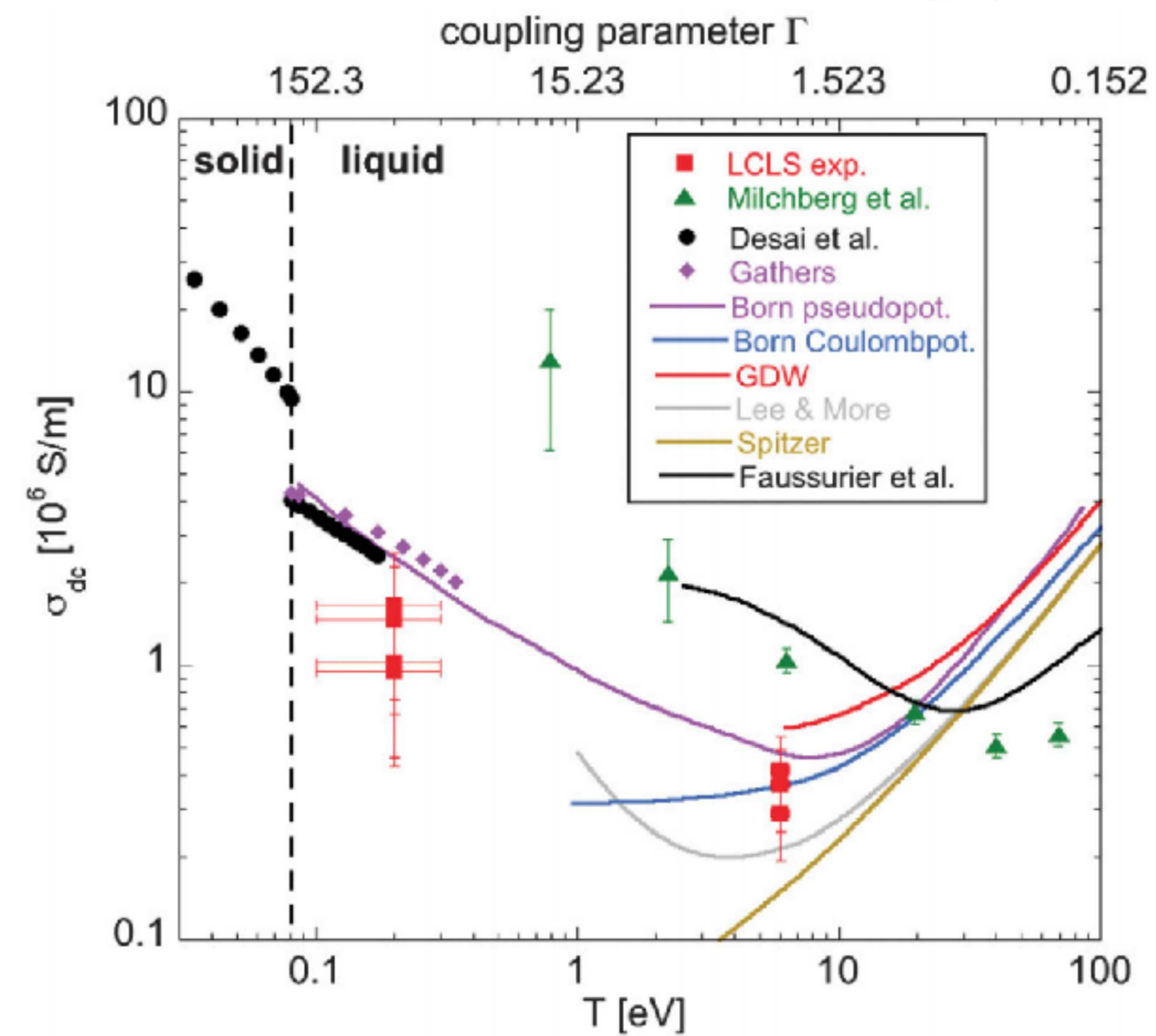
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$$\epsilon^M(q, \omega) - 1 = \frac{\left(1 + i \frac{\nu(\omega)}{\omega}\right) [\epsilon^{RPA}(q, \omega + i\nu(\omega)) - 1]}{1 + i \frac{\nu(\omega)}{\omega} \frac{\epsilon^{RPA}(q, \omega + i\nu(\omega)) - 1}{\epsilon^{RPA}(q, 0) - 1}}$$



*by way of the dynamic structure factor

Dynamic Structure Factor

Probe system with **x-ray***

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

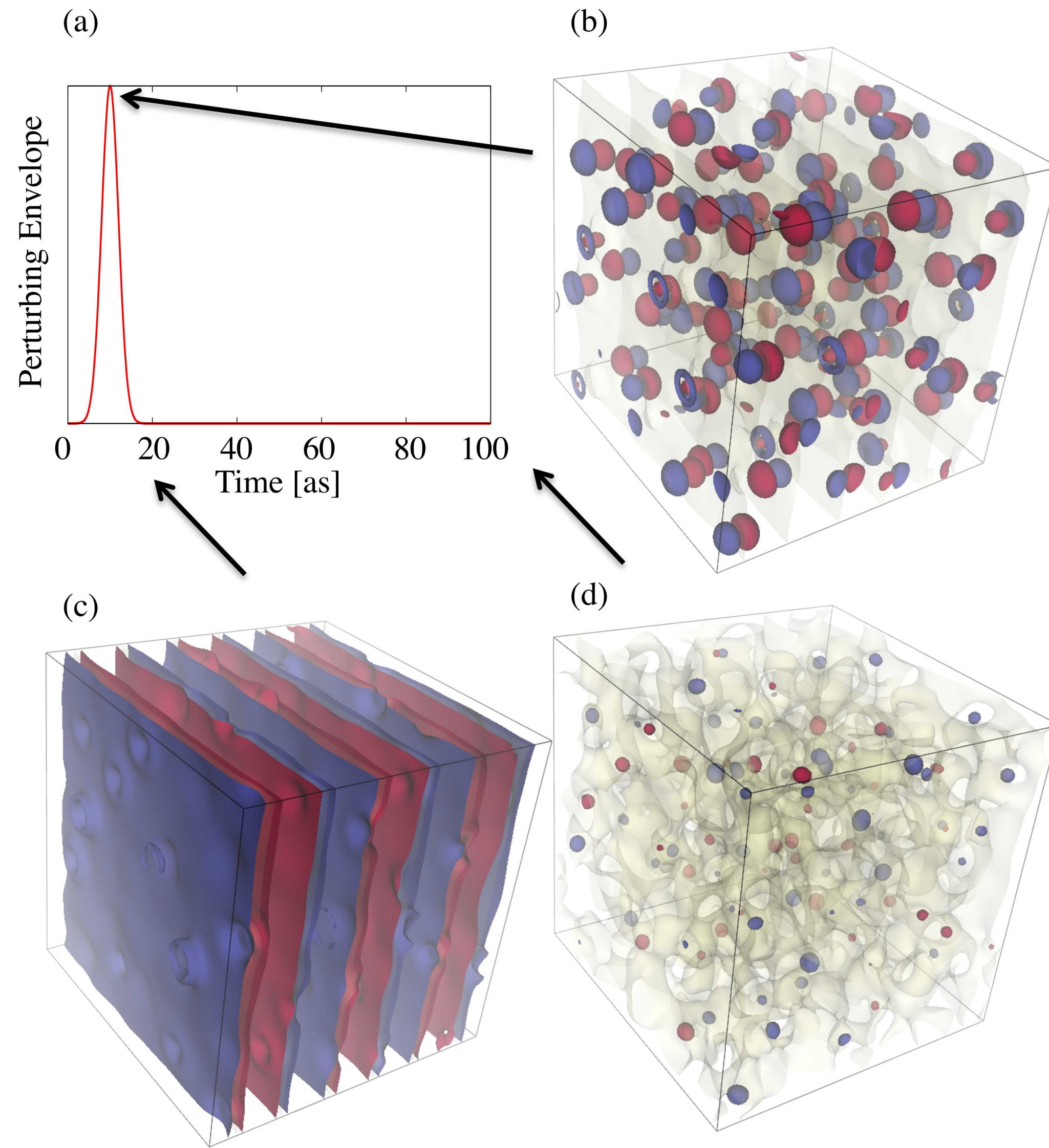
Record **density response**

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

Apply **fluctuation-dissipation**

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

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



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

**you can download the movie used to create this figure with our 2016 PRL

Cold Dense Matter*

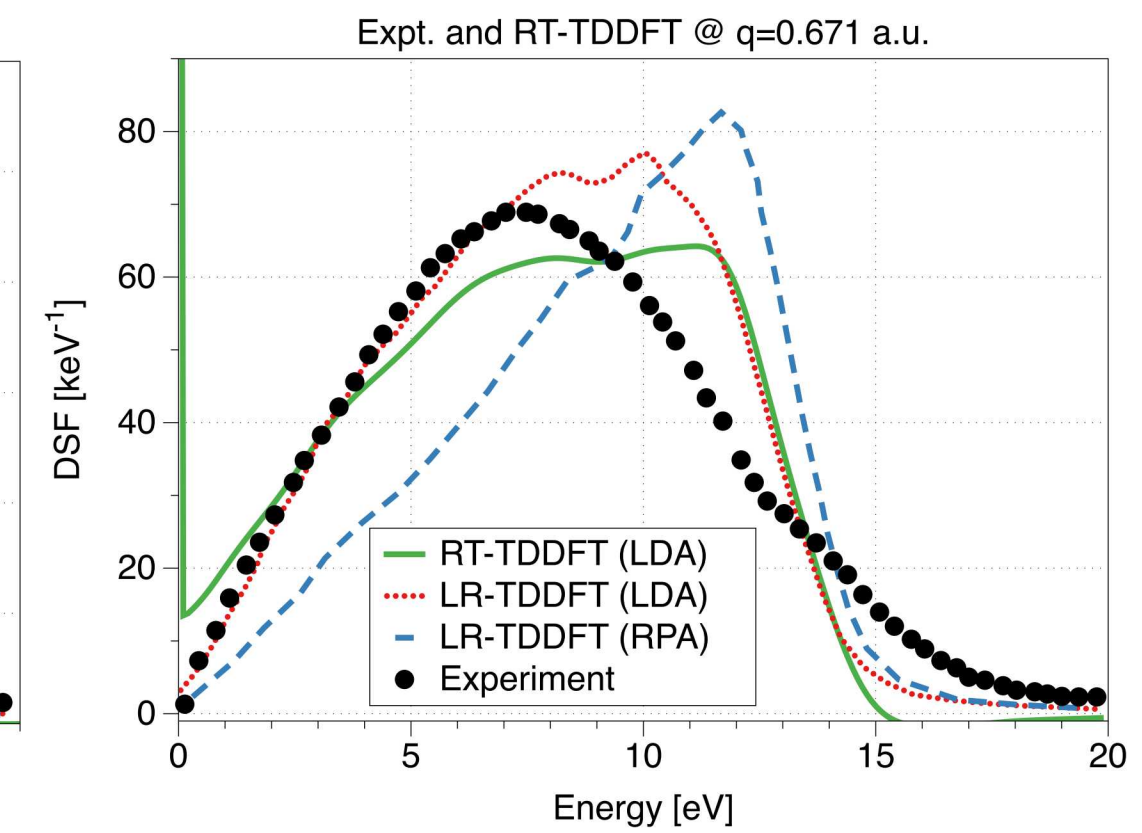
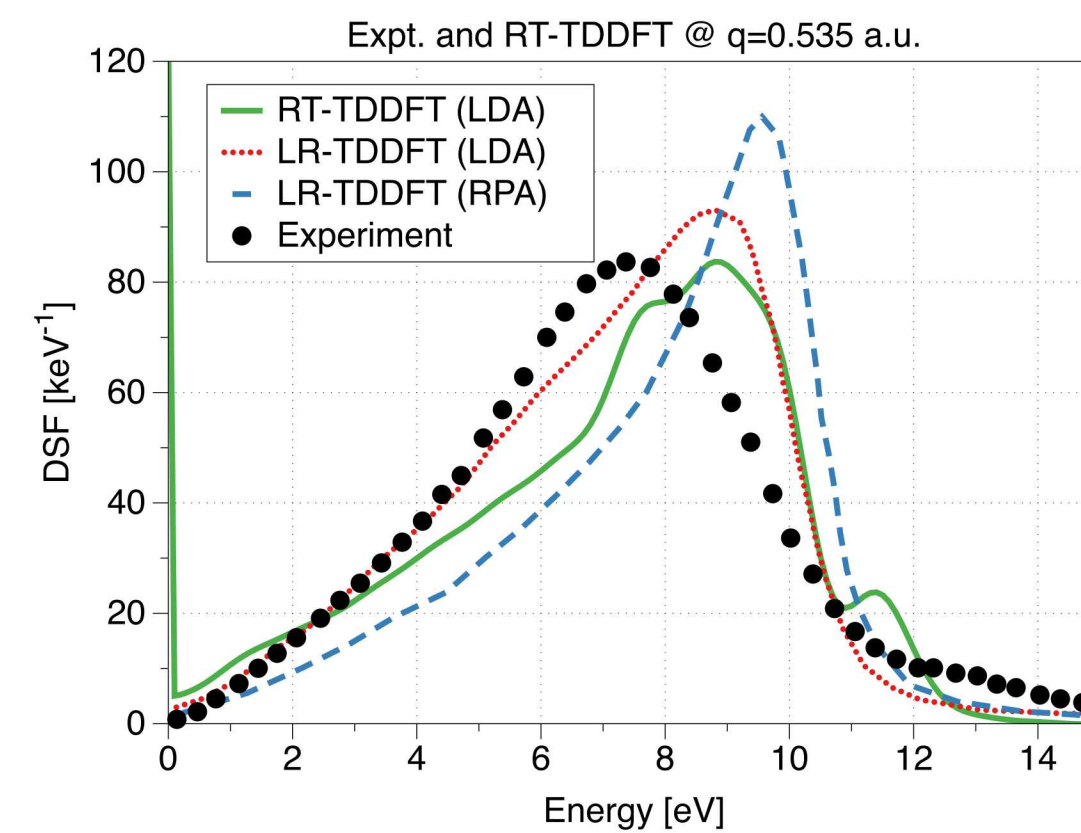
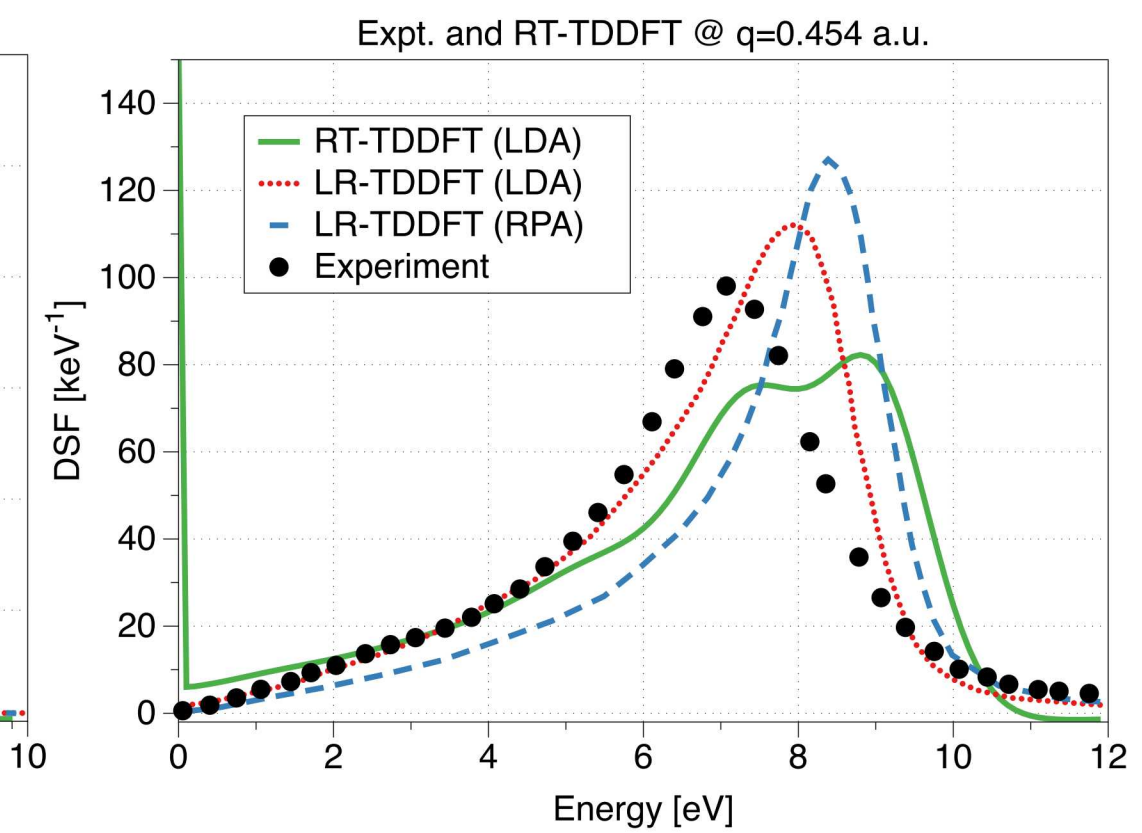
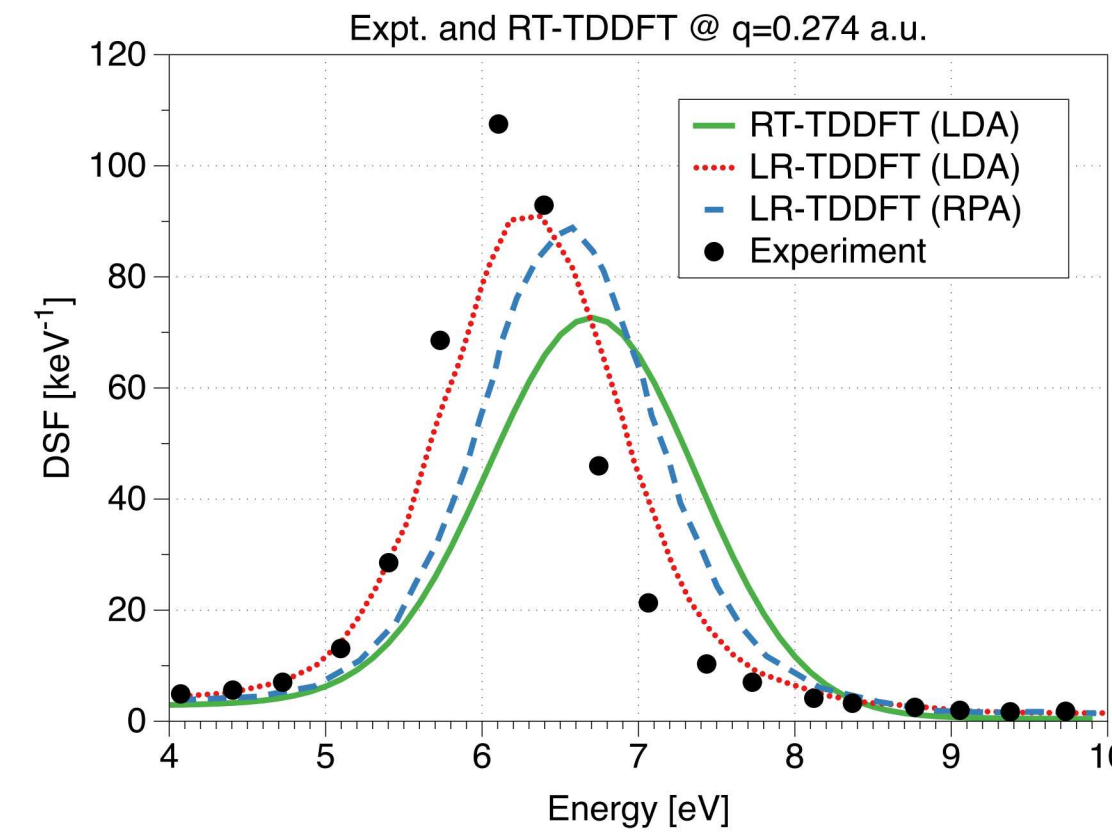
High resolution IXS data **calibrates expectations**

Comparison to Na IXS data

Dynamical response function in sodium and aluminum from time-dependent density-functional theory

Marco Carzani, Hans-Christian Weissker, Simo Huotari, Tuomas Pykkänen, Paolo Salvestrini, Giulio Monaco, Giovanni Onida, and Lucia Reining
Phys. Rev. B **84**, 075109 – Published 5 August 2011

- Adiabatic LDA isn't perfect
- Smearing / signal processing *really* matters
- ...good enough for XRTS



*condensed matter?

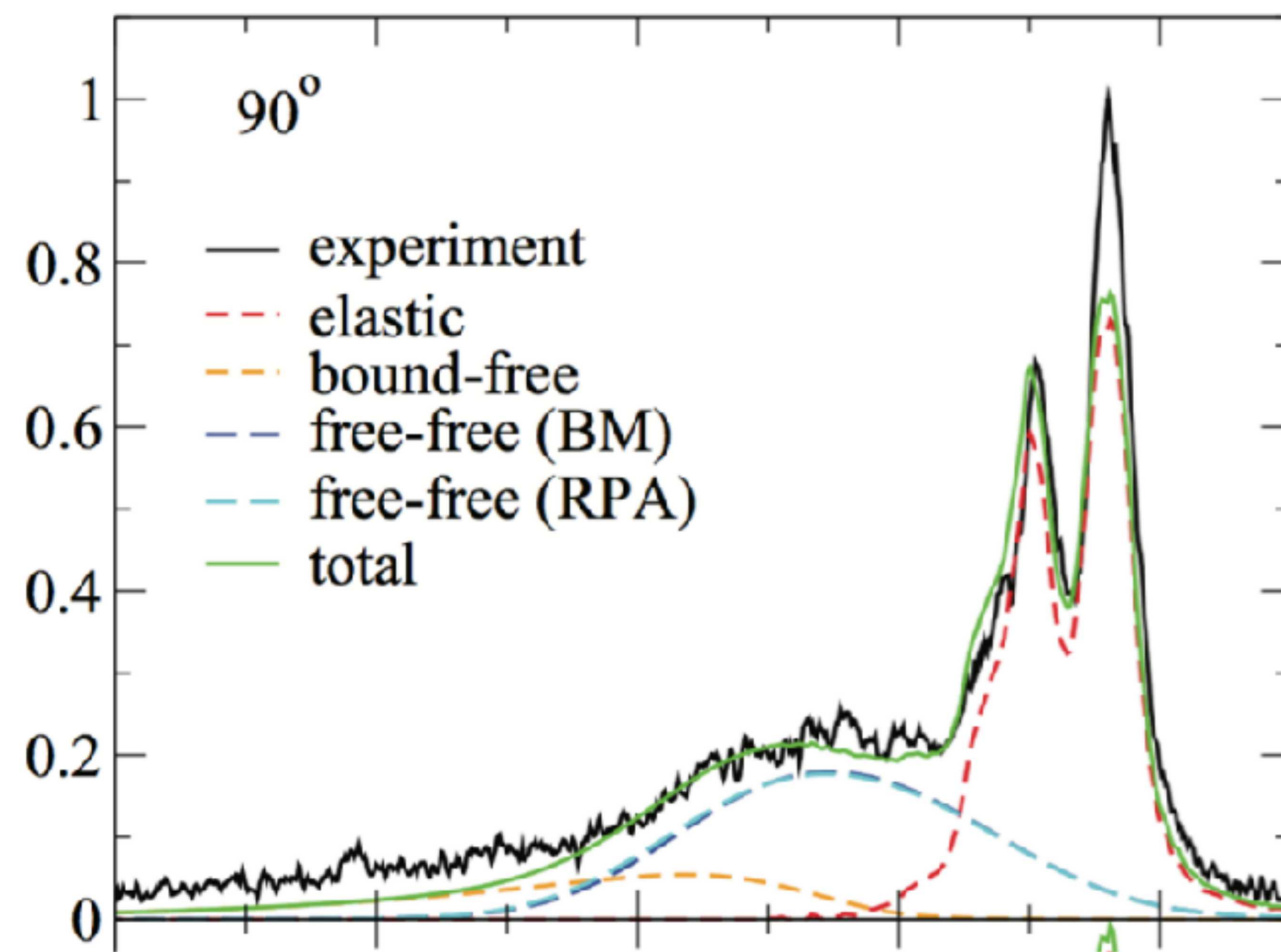
Warm Dense Matter

X-Ray Thomson-Scattering Measurements of Density and Temperature in Shock-Compressed Beryllium

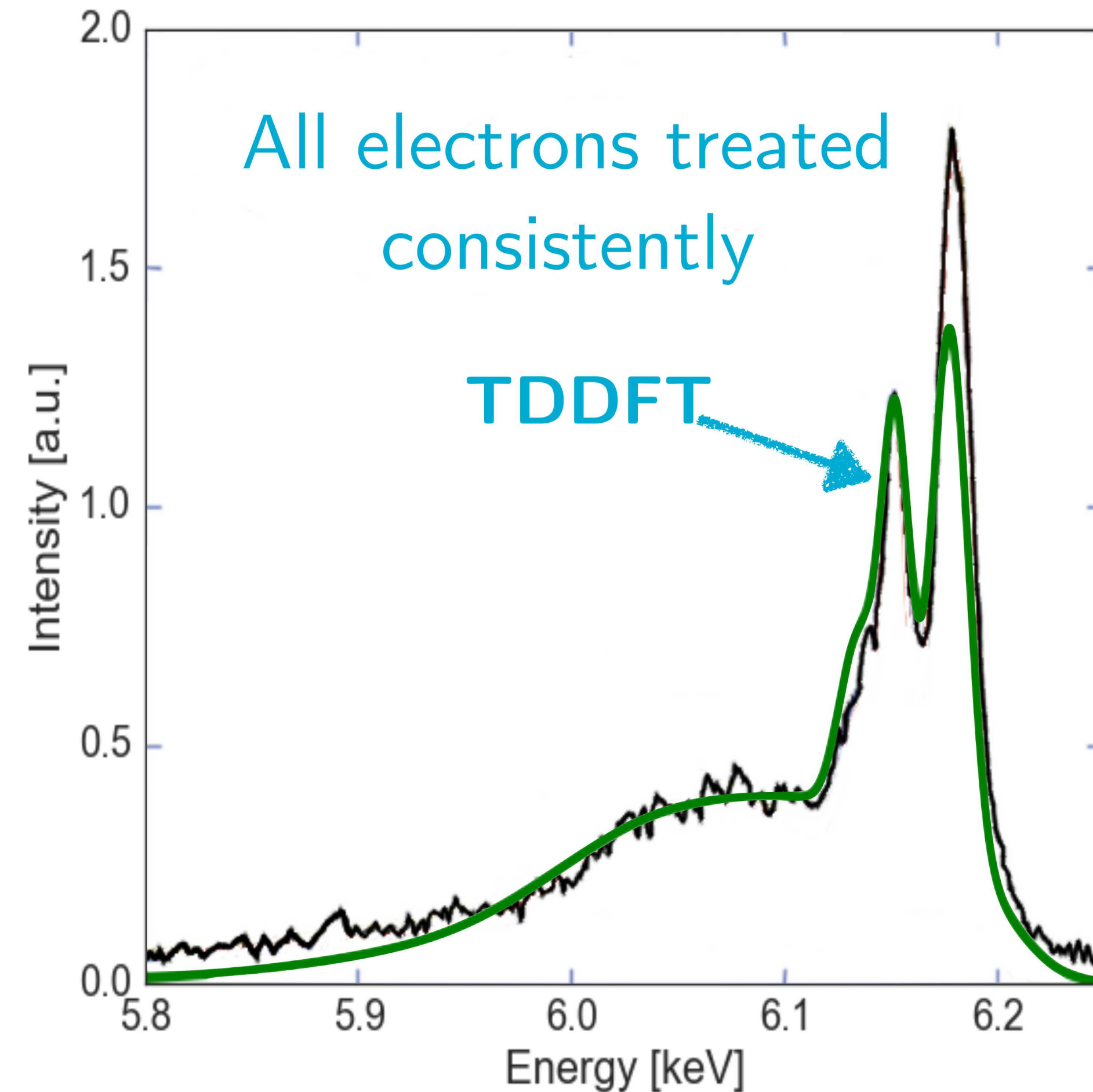
H. J. Lee, P. Neumayer, J. Castor, T. Döppner, R. W. Falcone, C. Fortmann, B. A. Hammel, A. L. Kritcher, O. L. Landen, R. W. Lee, D. D. Meyerhofer, D. H. Munro, R. Redmer, S. P. Regan, S. Weber, and S. H. Glenzer
Phys. Rev. Lett. **102**, 115001 – Published 16 March 2009

3x-compressed (+/-7%) Be
Ti=Te=13 eV (+/- 3 eV)

TDDFT does about as well
as average atom



Souza, et al., PRE, **89** (2014)



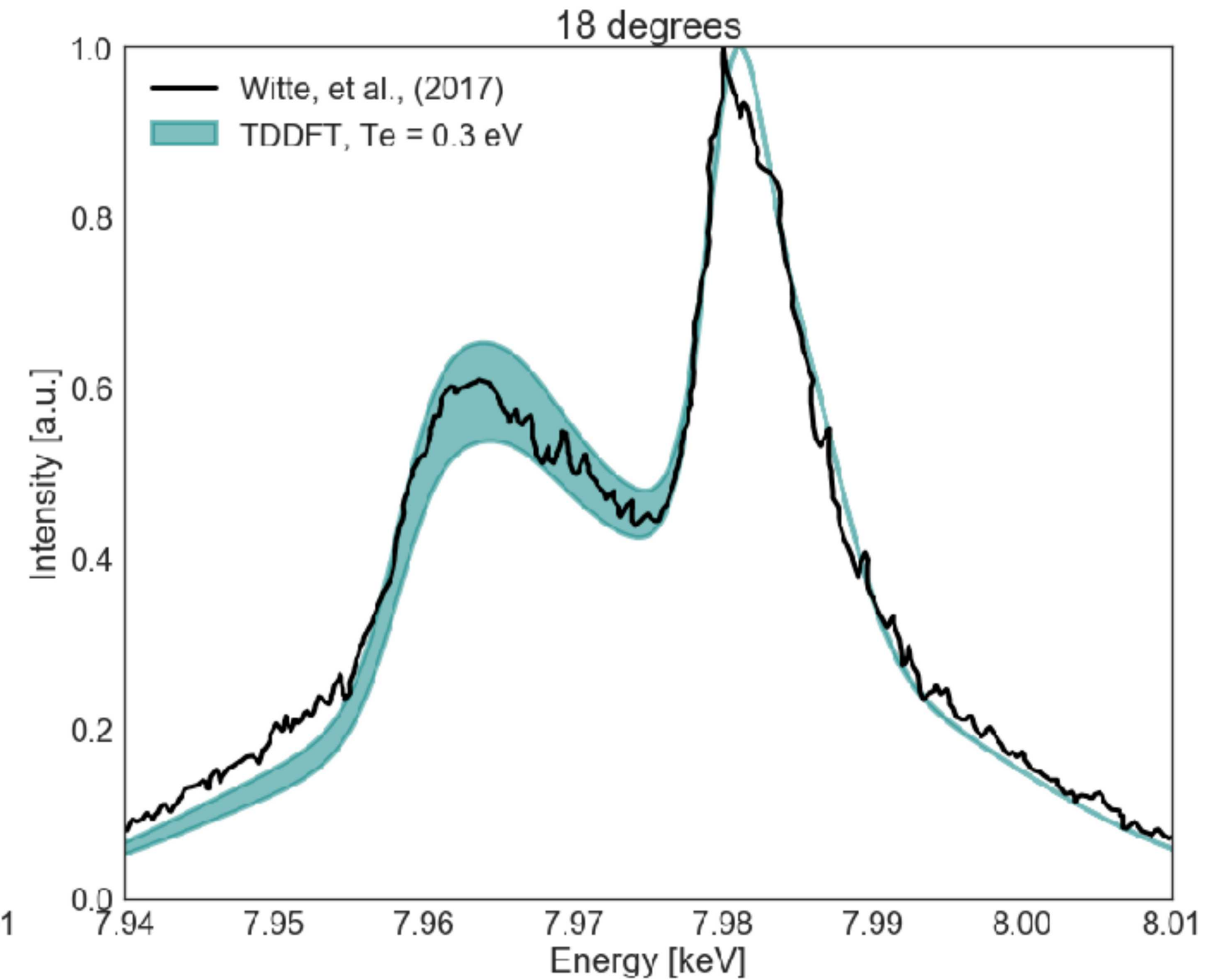
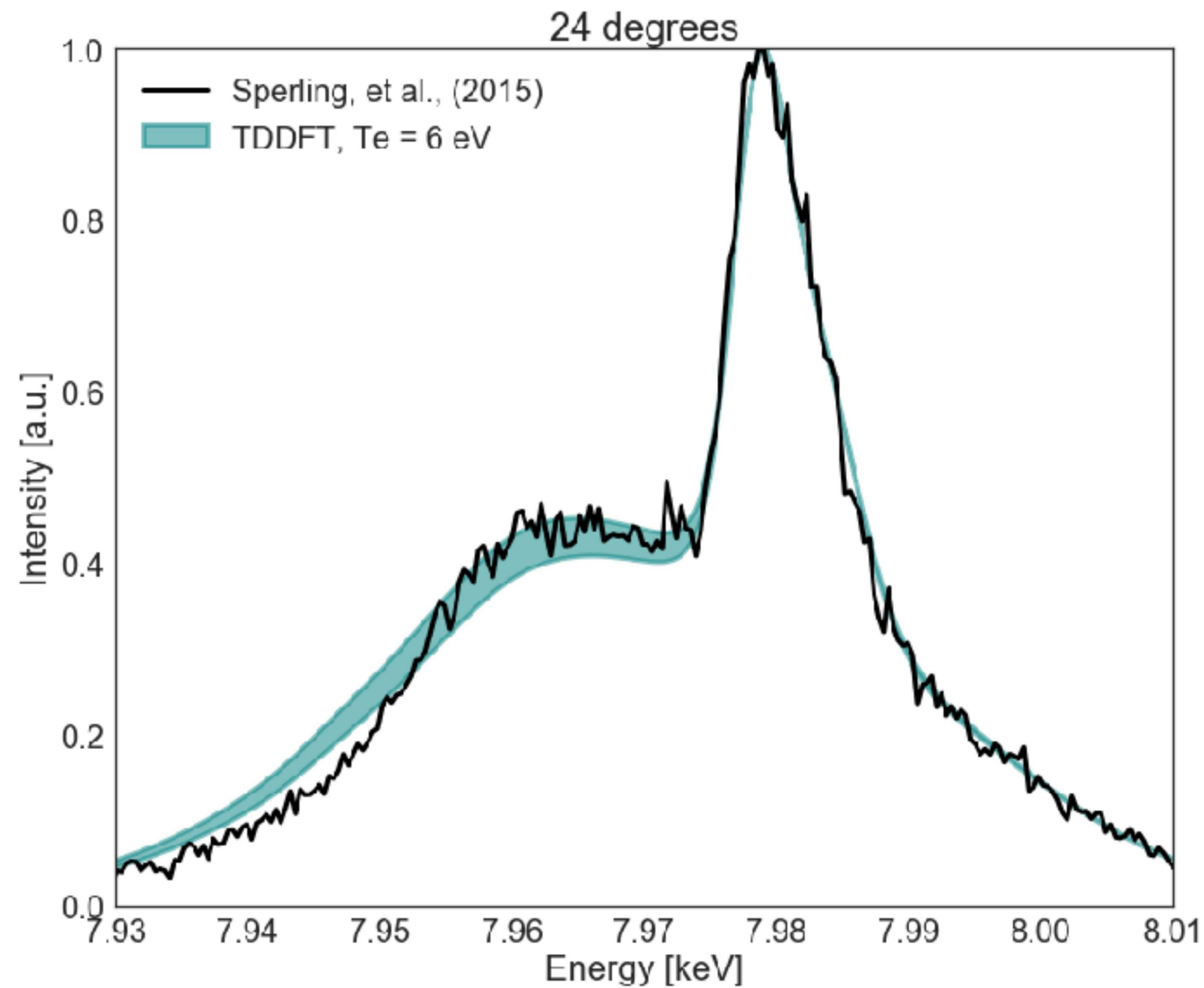
Isochorically Heated Aluminum

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P. Sperling, E. J. Gamboa, H. J. Lee, H. K. Chung, E. Galtier, Y. Omarbakiyeva, H. Reinholz, G. Röpke, U. Zastrau, J. Hastings, L. B. Fletcher, and S. H. Glenzer
Phys. Rev. Lett. **115**, 115001 – Published 9 September 2015

Warm Dense Matter Demonstrating Non-Drude Conductivity from Observations of Nonlinear Plasmon Damping

B. B. L. Witte, L. B. Fletcher, E. Galtier, E. Gamboa, H. J. Lee, U. Zastrau, R. Redmer, S. H. Glenzer, and P. Sperling
Phys. Rev. Lett. **118**, 225001 – Published 31 May 2017



Spread represents (very rough) uncertainty in elastic feature

Isochorically Heated Aluminum

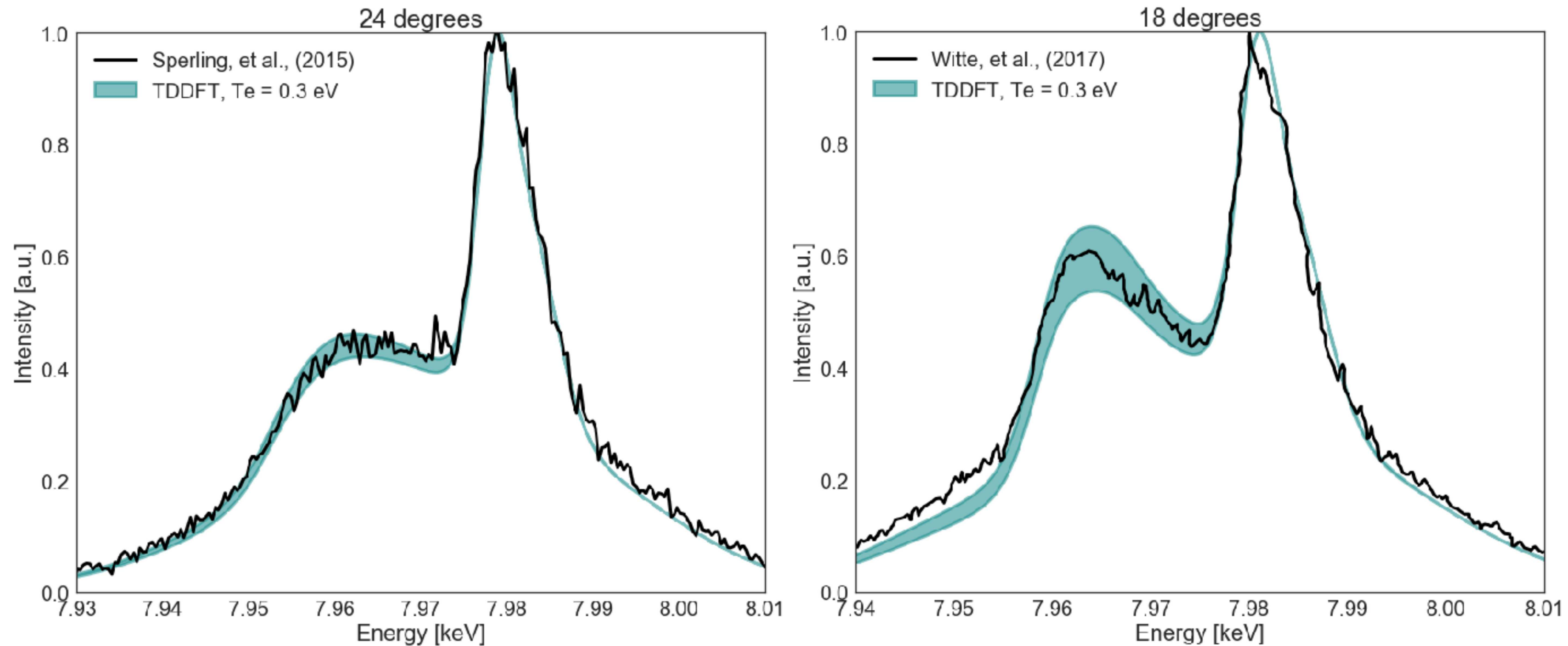
*similar observation by Chongjie Mo at March Meeting 2018

Free-Electron X-Ray Laser Measurements of Collisional-Damped Plasmons in Isochorically Heated Warm Dense Matter

P. Sperling, E. J. Gamboa, H. J. Lee, H. K. Chung, E. Galtier, Y. Omarbakiyeva, H. Reinholz, G. Röpke, U. Zastrau, J. Hastings, L. B. Fletcher, and S. H. Glenzer
Phys. Rev. Lett. **115**, 115001 – Published 9 September 2015

Warm Dense Matter Demonstrating Non-Drude Conductivity from Observations of Nonlinear Plasmon Damping

B. B. L. Witte, L. B. Fletcher, E. Galtier, E. Gamboa, H. J. Lee, U. Zastrau, R. Redmer, S. H. Glenzer, and P. Sperling
Phys. Rev. Lett. **118**, 225001 – Published 31 May 2017



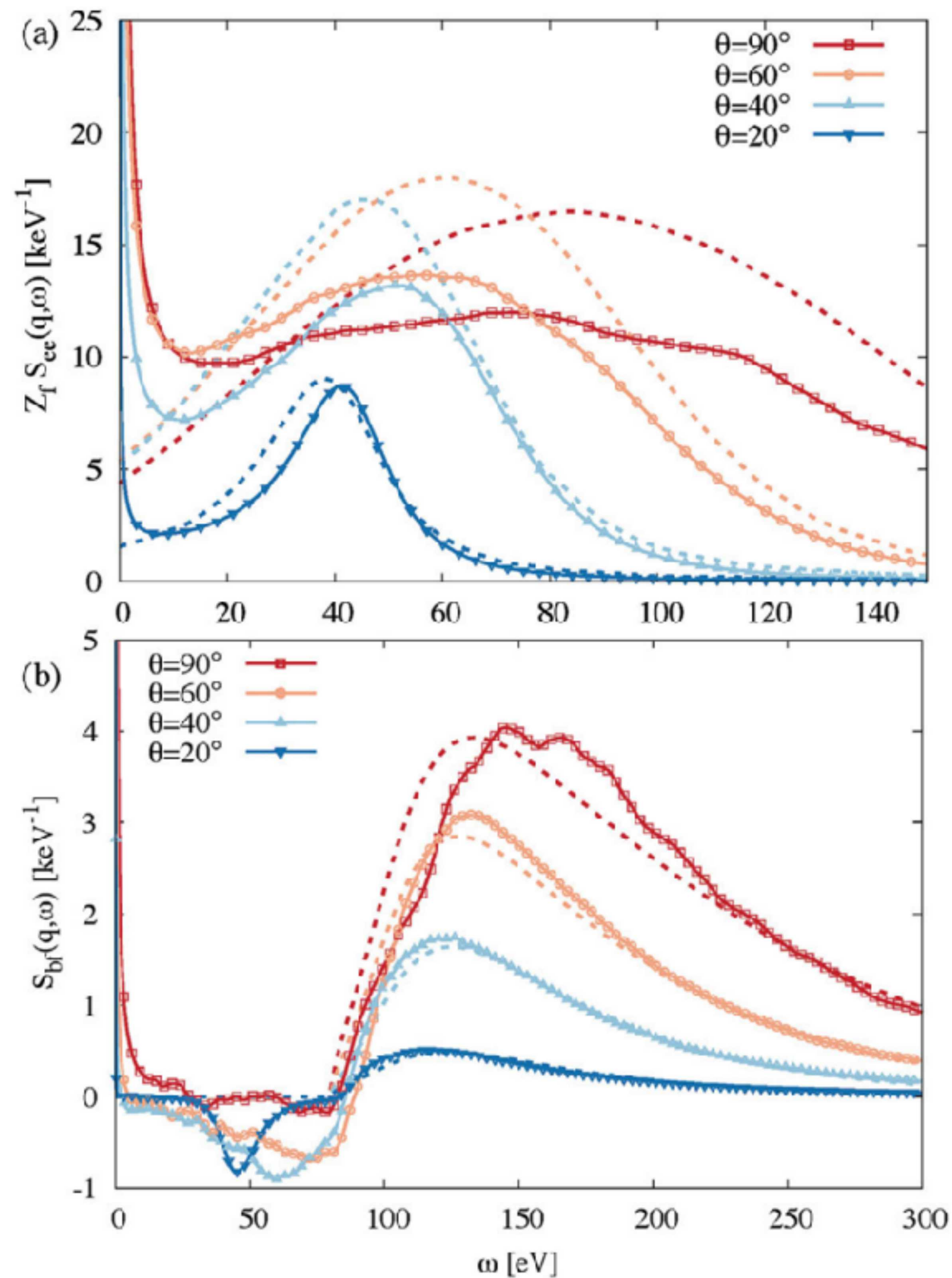
What this means depends on how much you believe TDDFT*
relative to detailed balance

What is *theory* telling *theory*?

3x-compressed (+/-7%) Be
Ti=Te=13 eV (+/- 3 eV)

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



Solid = TDDFT

Dashed (top) = Mermin w/DFT v(w)

Dashed (bot) = Avg. atom (Slater-A)

- Mermin degrades w/increasing q
- Average atom generally does well...
- ...missing core-valence polarization?

Functionals?

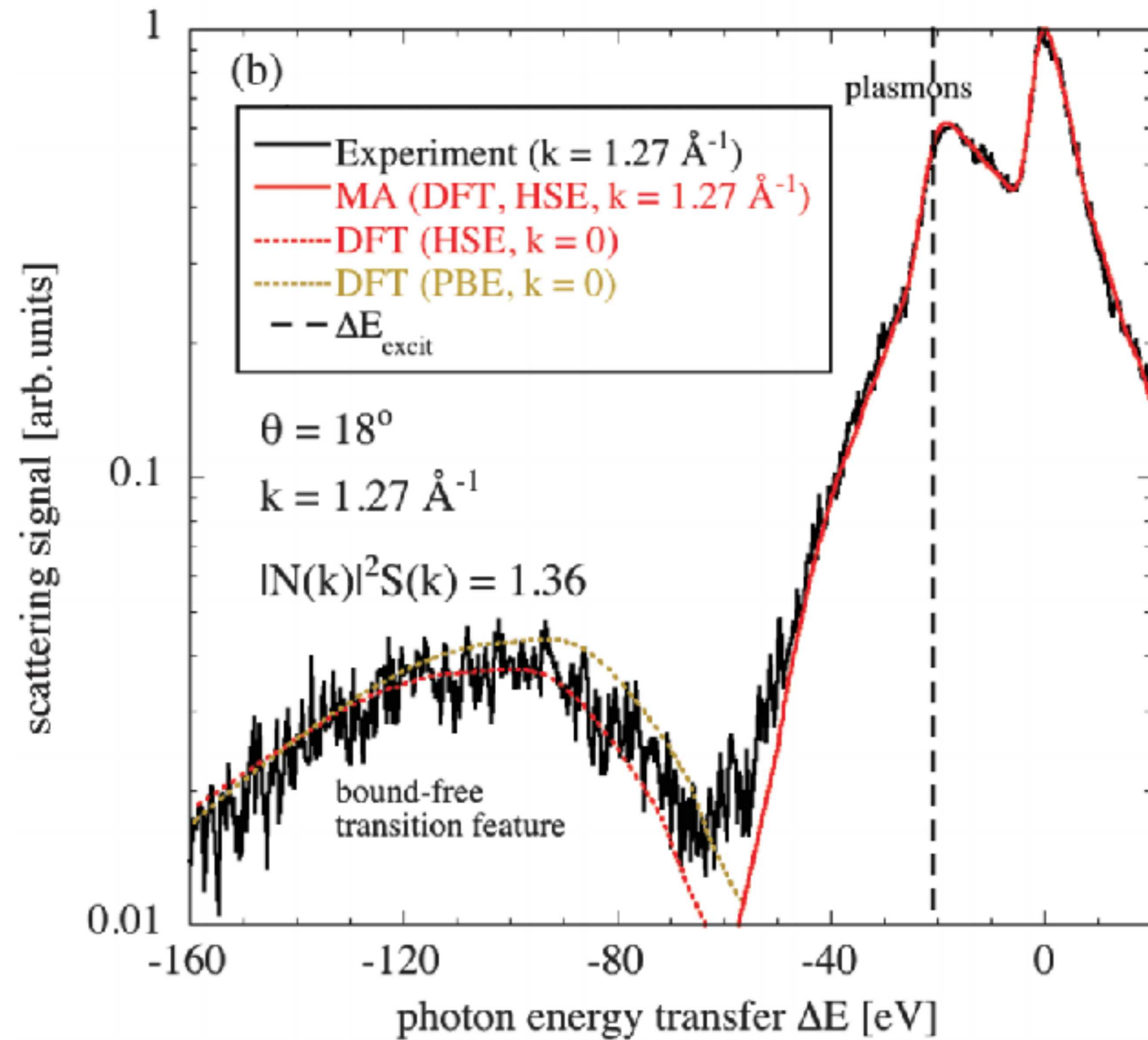
Warm Dense Matter Demonstrating Non-Drude Conductivity from Observations of Nonlinear Plasmon Damping

B. B. L. Witte, L. B. Fletcher, E. Galtier, E. Gamboa, H. J. Lee, U. Zastra, R. Redmer, S. H. Glenzer, and P. Sperling
Phys. Rev. Lett. **118**, 225001 – Published 31 May 2017

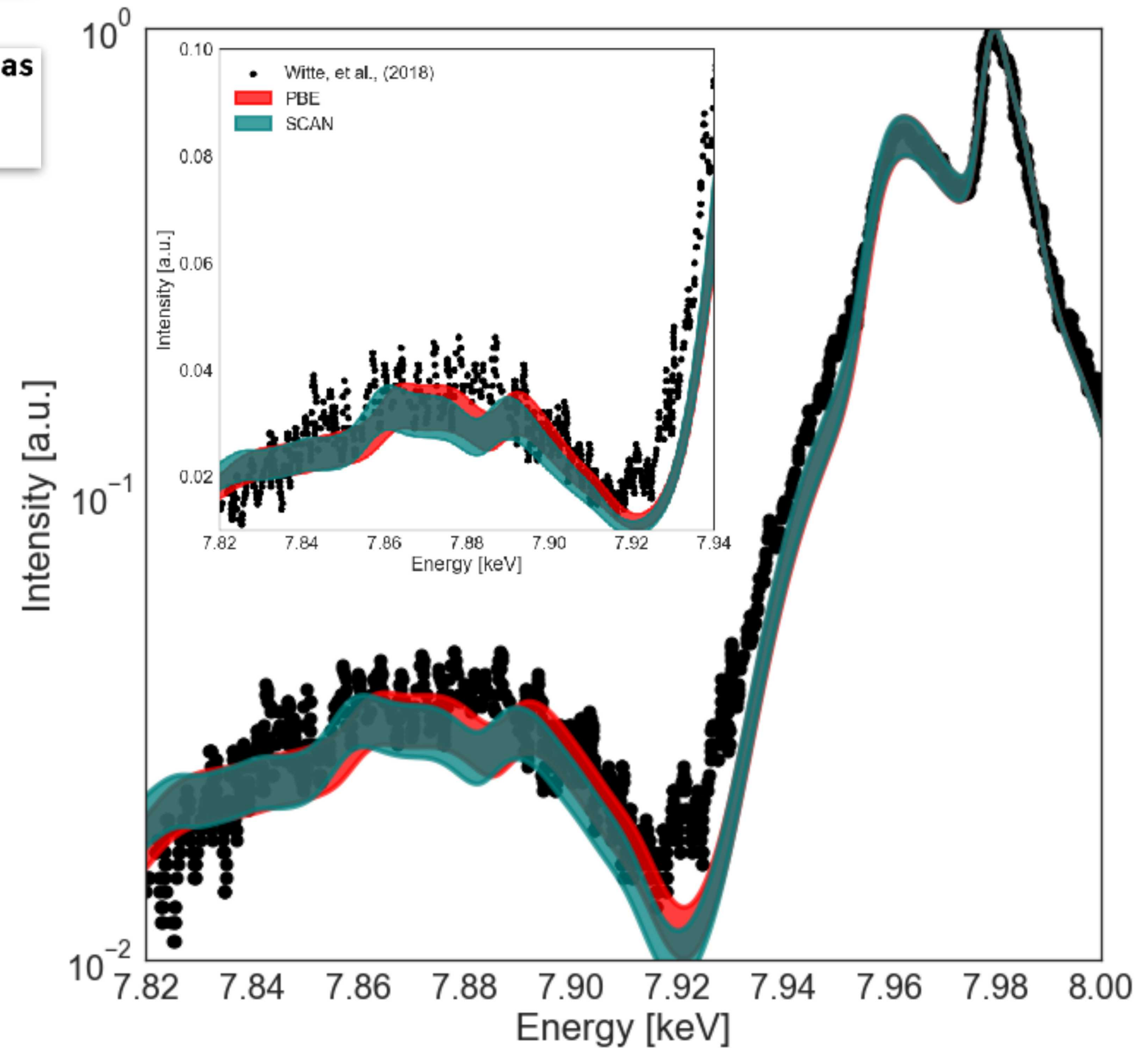
Observations of non-linear plasmon damping in dense plasmas

Physics of Plasmas **25**, 056901 (2018); <https://doi.org/10.1063/1.5017889>

B. B. L. Witte^{1,2,a}, P. Sperling^{1,3,4}, M. French², V. Recoules⁵, S. H. Glenzer¹, and R. Redmer²

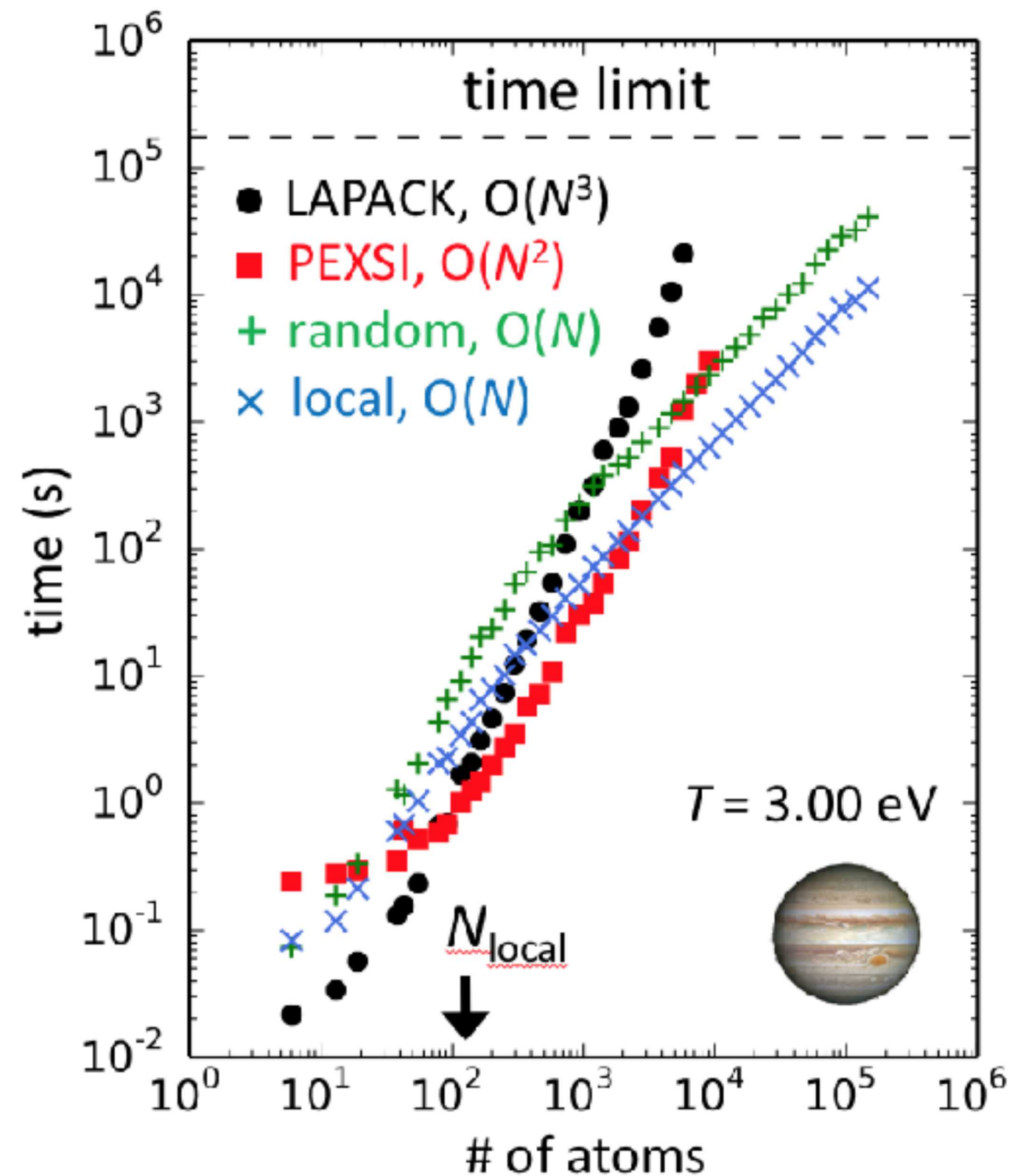


L-edge sensitive to XC functional



Higher Temperatures, Bigger Systems

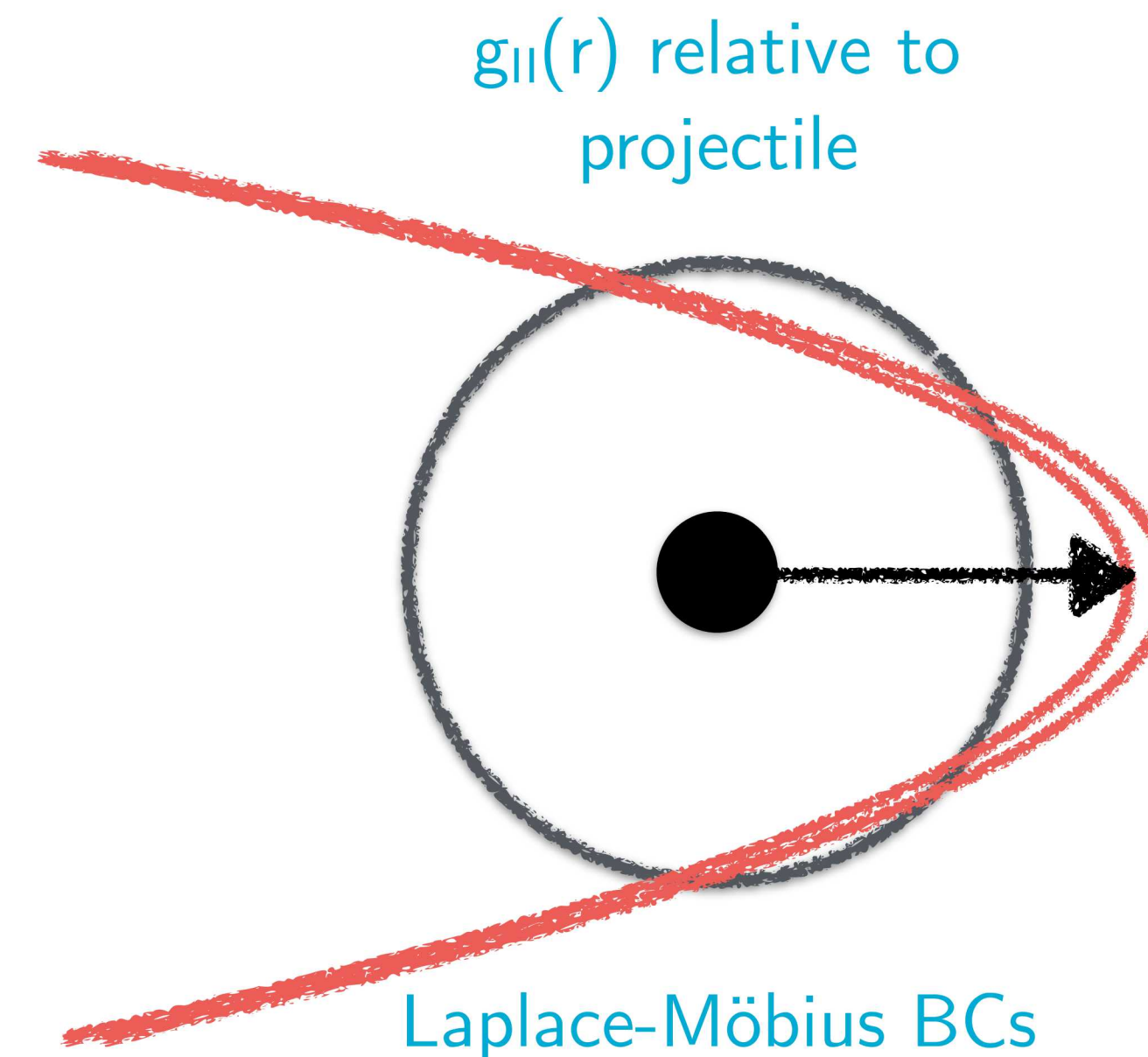
New ideas for linear scaling



Work w/ Jonathan Moussa

Minimax rational approximation of the Fermi-Dirac distribution
 The Journal of Chemical Physics 145, 164108 (2016); <https://doi.org/10.1063/1.4965886>
 Jonathan E. Moussa

Ehrenfest for average atom



Work w/ Attila Cangi

Efficient formalism for warm dense matter simulations
 Attila Cangi and Aurora Pribram-Jones
 Phys. Rev. B 92, 161113(R) – Published 16 October 2015

Conclusion

ICF target designers, planetary scientists, and others don't just want equation-of-state (energetics)

Transport models are increasingly important, and experiments are coming online that will test them

This is an excellent opportunity for theorists to engage, and to start developing methods to target problem points

The views expressed in this presentation do not necessarily represent the views of the U.S. Department of Energy or the United States Government.