

Electronic Transport Properties of Warm Dense Matter from Time-Dependent Density Functional Theory



PRESENTED BY

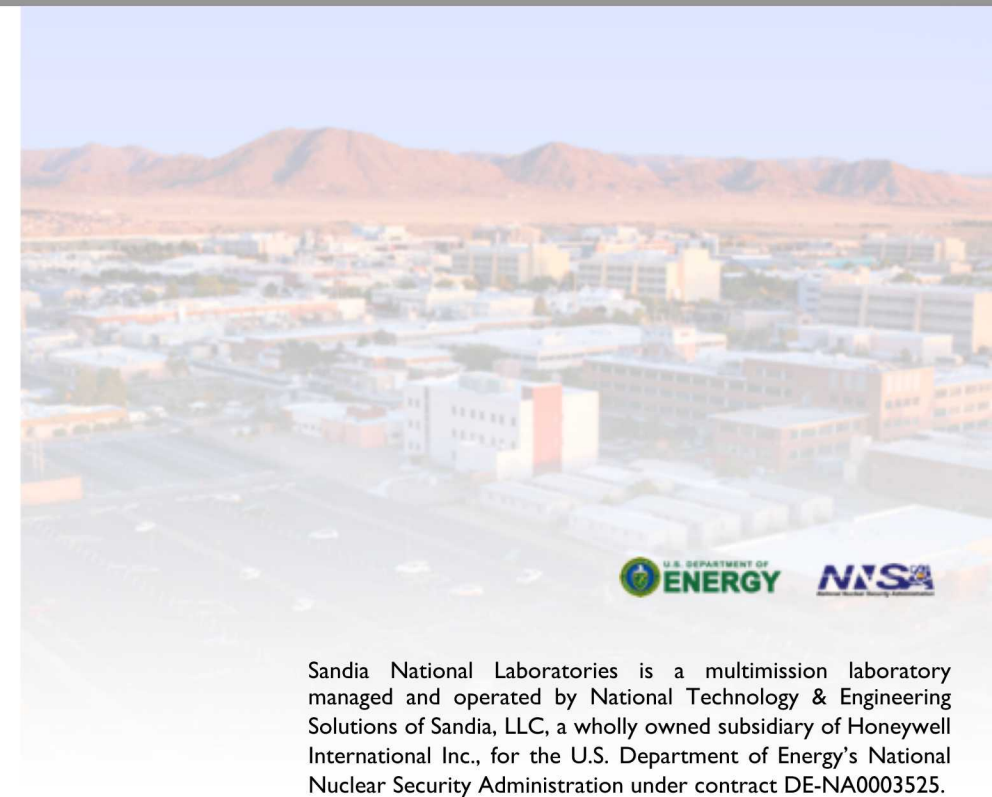
Attila Cangi

2019 Z Fundamental Science Workshop, Albuquerque, NM

Collaborators

Andrew. D. Baczewski, Stephanie. B. Hansen

Monday, August 12, 2019



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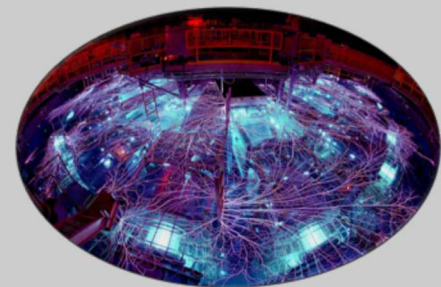
Characterization of HED phenomena is challenging.

Modeling landscape is a blend of **various disciplines** (plasma, atomic, condensed matter physics) and **methods** (magnetohydrodynamics, particle-in cell, molecular dynamics, average-atom, DFT-MD, TDDFT).

Persistence of electron correlation and non-LTE conditions are the greatest challenges for numerical modeling.

Ultimately want to avoid empiricism and a **self-consistent, first-principles method** for the prediction of WDM/HED phenomena.

Compare prediction of HED **transport phenomena** within various methods (average atom, Kubo-Greenwood) with TDDFT.



Stopping power

$$\frac{\partial E}{\partial x} = \frac{1}{v} \frac{\partial}{\partial t} \langle E \rangle$$

Electrical conductivity

$$\sigma(\omega) = \frac{J(\omega)}{E(\omega)}$$

Dynamic structure factor

$$S(\mathbf{q}, \omega) = -\frac{1}{\pi} \frac{\Im[\chi(\mathbf{q}, -\mathbf{q}, \omega)]}{1 - e^{-\omega/k_B T}}$$

Coupled Electron-Ion Many-Body Problem

$$i\hbar \frac{\partial}{\partial t} \Psi(\{\mathbf{R}\}, \{\mathbf{r}\}, t) = \hat{H} \Psi(\{\mathbf{R}\}, \{\mathbf{r}\}, t)$$

$$\hat{H} = \hat{T}^n + \hat{W}^{nn} + \boxed{\hat{T}^e + \hat{W}^{ee} + \hat{W}^{en}} + \hat{V}^n + \boxed{\hat{V}^e}$$

$$\hat{T}^n = \sum_{\alpha}^{N^n} -\frac{1}{2M_{\alpha}} \nabla_{\alpha}^2 \quad \hat{W}^{nn} = \frac{1}{2} \sum_{\alpha, \beta, \alpha \neq \beta}^{N^n} \frac{Z_{\alpha} Z_{\beta}}{|\mathbf{R}_{\alpha} - \mathbf{R}_{\beta}|}$$

$$\hat{T}^e = \sum_{\alpha}^{N^e} -\frac{1}{2} \nabla^2 \quad \boxed{\hat{W}^{ee} = \frac{1}{2} \sum_{i, j, i \neq j}^{N^e} \frac{1}{|\mathbf{r}_i - \mathbf{r}_j|}}$$

$$\hat{W}^{ne} = - \sum_i^{N^e} \sum_{\alpha}^{N^n} \frac{Z_{\alpha}}{|\mathbf{r}_i - \mathbf{R}_{\alpha}|}$$

$$\hat{V}^n = \sum_{\alpha}^{N^n} v^n(\mathbf{R}_{\alpha}) \quad \hat{V}^e = \sum_i^{N^e} v^e(\mathbf{r}_i)$$

Time-dependent (non-relativistic) Schrödinger equation for the many-particle (molecular) Hamiltonian.

Kinetic energy and interaction of ions.

Kinetic energy and interaction of electrons.

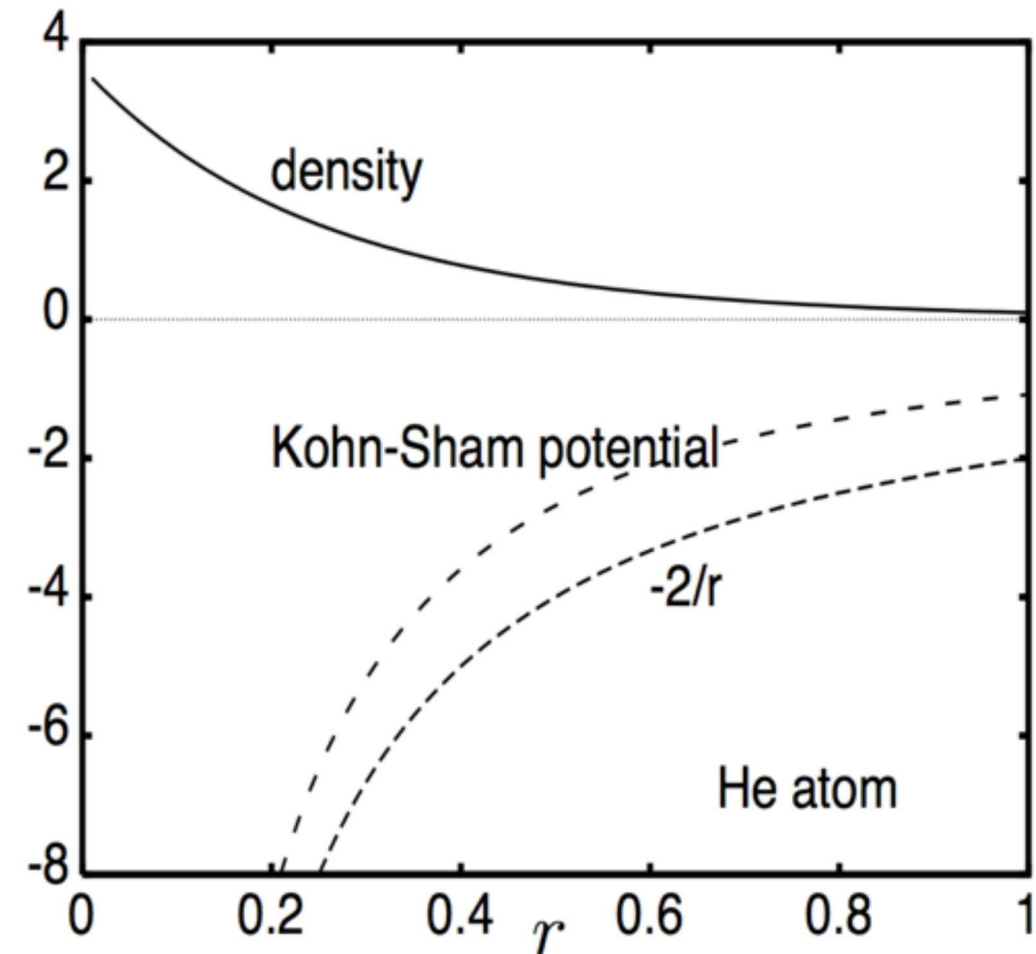
Electron-ion interaction.

External potential.

$$\left[-\frac{\nabla^2}{2} + v_s(\mathbf{r}) \right] \phi_j(\mathbf{r}) = \epsilon_j \phi_j(\mathbf{r}) \quad (\text{Kohn-Sham equations})$$

$$v_s(\mathbf{r}) = v(\mathbf{r}) + \frac{\delta U}{\delta n(\mathbf{r})} + \boxed{\frac{\delta E_{\text{xc}}}{\delta n(\mathbf{r})}} \quad (\text{Kohn-Sham potential})$$

$$n(\mathbf{r}, t) = \sum_i f_i \phi_i^*(\mathbf{r}, t) \phi_i(\mathbf{r}, t) \quad (\text{Electronic density})$$



The ABC of DFT (dft.uci.edu/doc/gl.pdf).

Introduction to Time-dependent DFT

Real-time propagation



1. Prepare initial state from a static DFT calculation

$$\left[-\frac{\nabla^2}{2} + v_S(\mathbf{r}) \right] \phi_j(\mathbf{r}) = \epsilon_j \phi_j(\mathbf{r})$$

2. Solve the TD Kohn-Sham equations

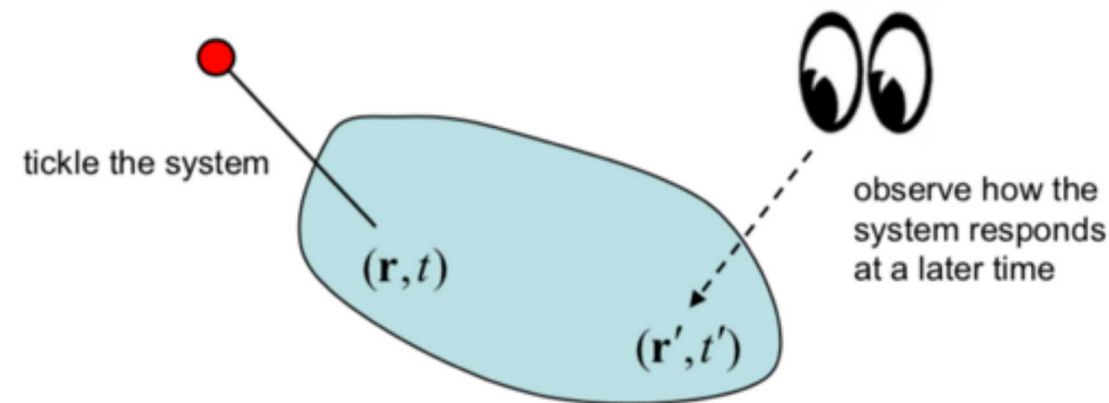
$$i \frac{d\phi_j(\mathbf{r})}{dt} = \left[-\frac{\nabla^2}{2} + v_S(\mathbf{r}) \right] \phi_j(\mathbf{r})$$

$$v_S(\mathbf{r}, t) = v(\mathbf{r}, t) + v_H(\mathbf{r}, t) + v_{XC}(\mathbf{r}, t) \approx v_S^{gs}[n(\mathbf{r}, t)](\mathbf{r})$$

3. Compute observables in terms of time-dependent density

$$n(\mathbf{r}, t) = \sum_i f_i \phi_i^*(\mathbf{r}, t) \phi_i(\mathbf{r}, t)$$

Linear response



Courtesy of N. Maitra and C. Ullrich (2018).

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

$$\epsilon^{-1}(\mathbf{r}, \mathbf{r}', \omega) = \delta(\mathbf{r} - \mathbf{r}') + \int d\mathbf{r}'' \frac{\chi(\mathbf{r}'', \mathbf{r}', \omega)}{|\mathbf{r} - \mathbf{r}''|}$$

Compute various transport properties (stopping power, conductivity, dynamical structure factor).

Implementation of TDDFT-Ehrenfest MD in VASP

- Andrew D. Baczewski et al., PRL **116**, 115004 (2016)
- Plane wave basis
- Projector-augmented wave (PAW) formalism
- Crank-Nicolson time integration (unitary)
- Generalized minimal residual method

Scales well on DOE machines

- Typically 100s of cores, a few hours
- No “free” parameters
- takes mass density
- # of electrons
- exchange-correlation functional



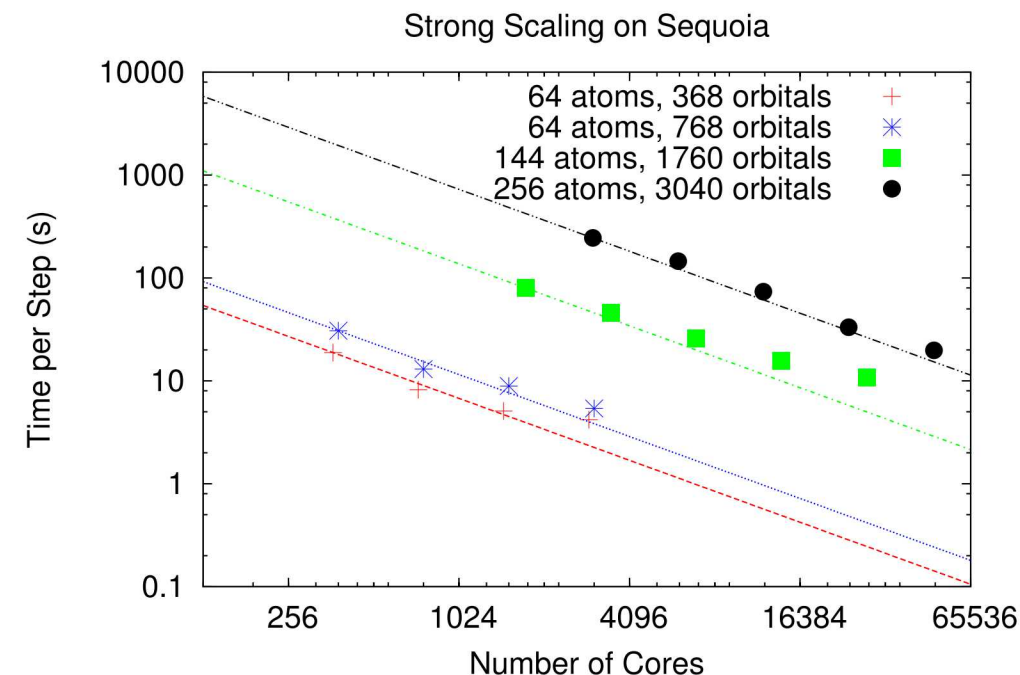
Elk code



Coupled electron-ion equations of motion

$$i \frac{\partial}{\partial t} \phi_i(\mathbf{r}, t) = \left\{ -\frac{1}{2} \nabla^2 + v_s(\mathbf{r}, t) \right\} \phi_i(\mathbf{r}, t)$$

$$M_\alpha \frac{\partial^2 \mathbf{R}_\alpha}{\partial t^2} = -\nabla_{\mathbf{R}_\alpha} E[\mathbf{R}_\alpha, n(\mathbf{r}, t)]$$



Stopping Power in Warm Dense Targets

Example: Hydrogen moving through cold, bulk aluminum in a channeling trajectory

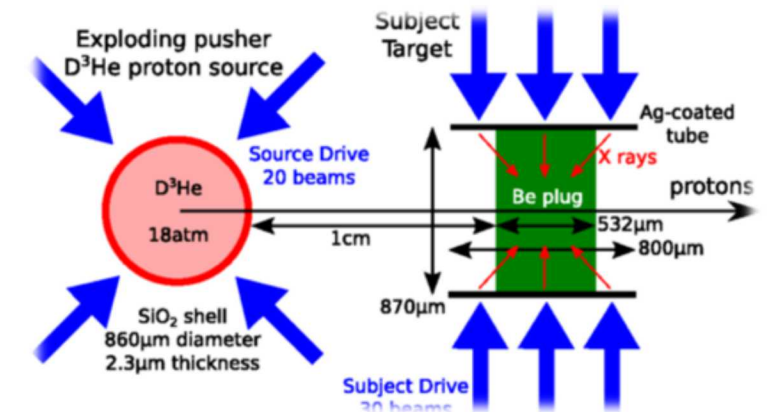
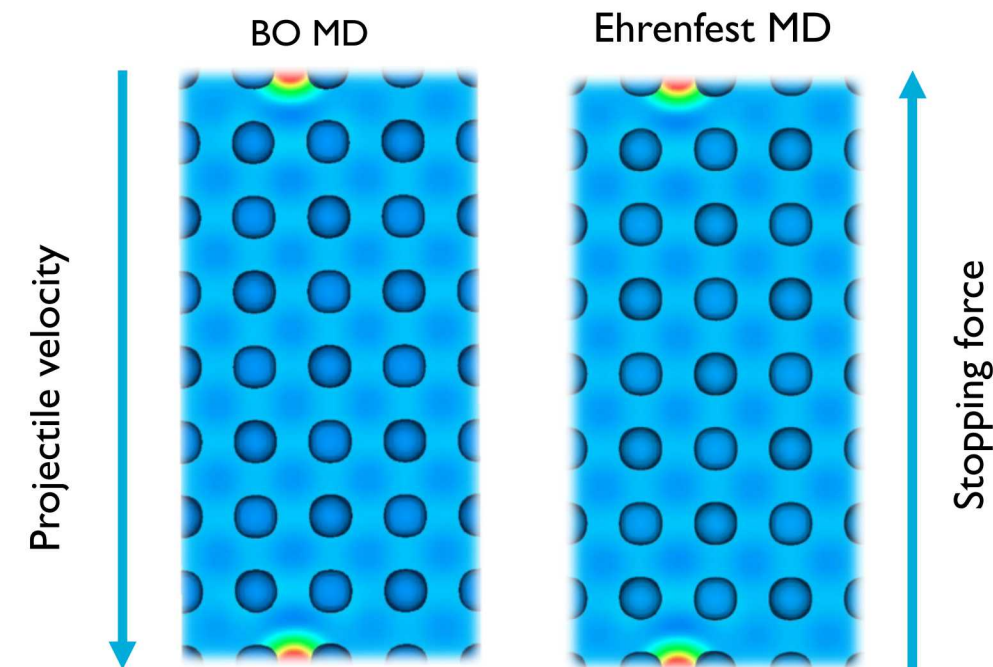
$$\frac{\partial E}{\partial x} = \frac{1}{v} \frac{\partial}{\partial t} \langle E \rangle$$

Stopping mechanisms

- Nuclear stopping (lattice vibrations)
- Electronic stopping (electronic excitations)

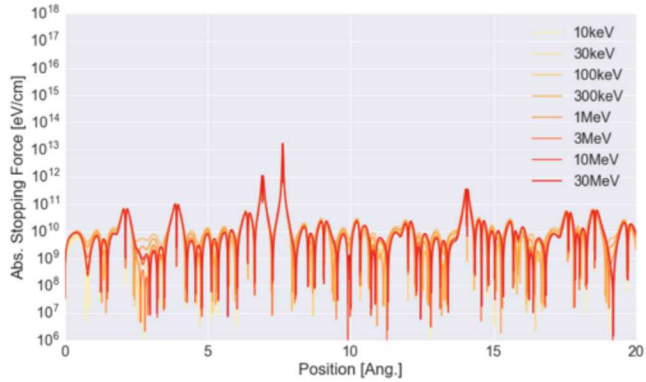
Large body of literature for cold targets

- Empirical approximations (Rutherford, Thomson, Bohr, Bethe)
- Parameter-free atomistic simulations
- Electronic structure coupled to molecular dynamics
- Cold stopping power (Echenique, Correa, Artacho, Schleife)

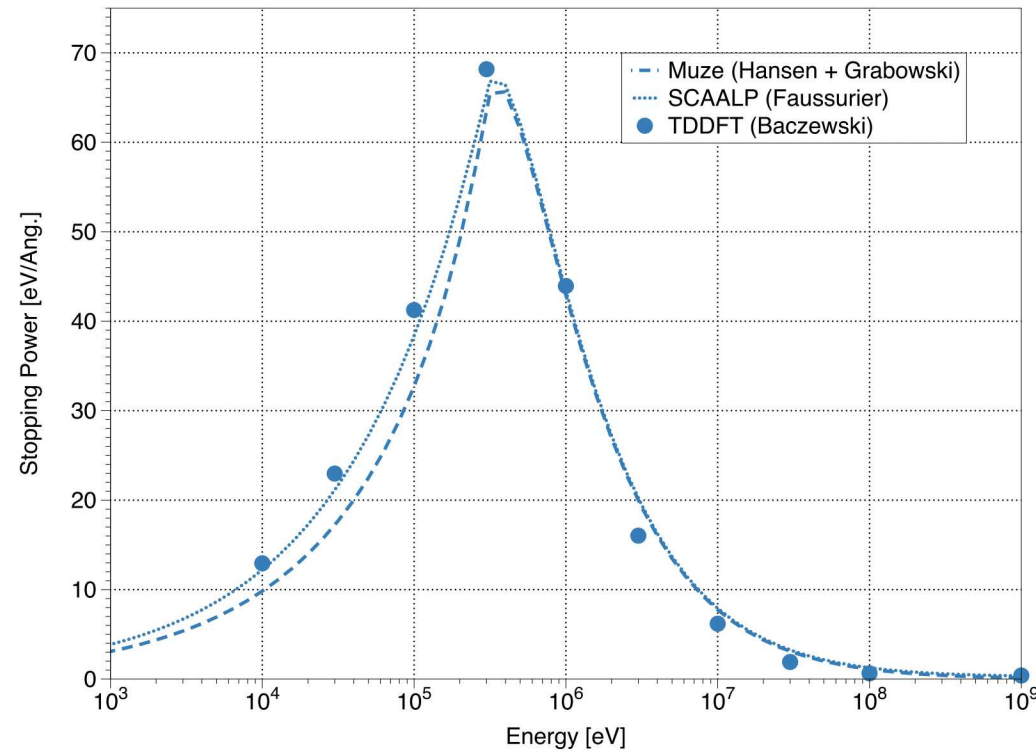
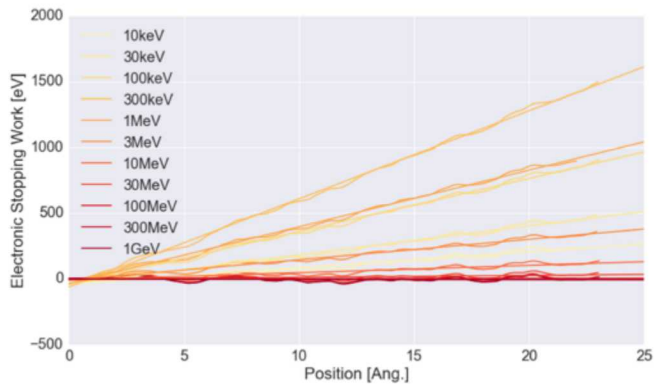
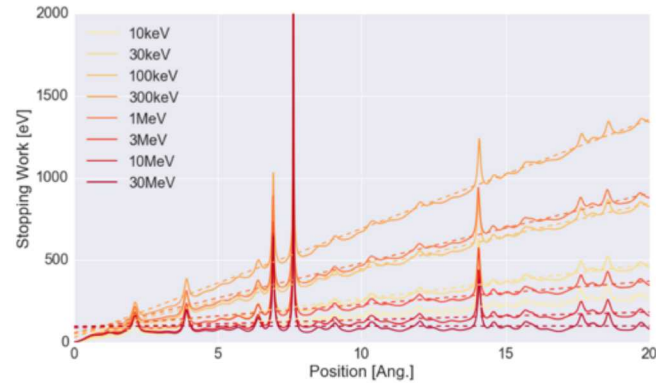


Zylstra et al., Phys. Rev. Lett. 114, 215002 (2015).

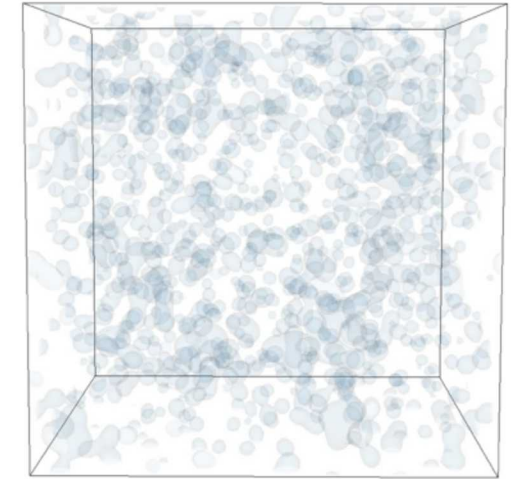
Stopping Power in Warm Dense Targets



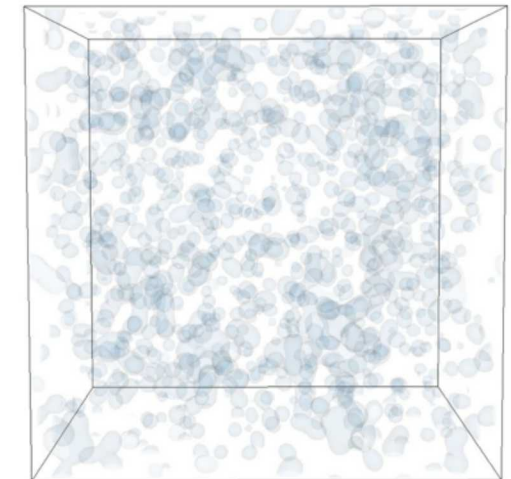
- Stopping at 10 g/cc (mass density) and 2 eV (temperature)
- **Force vs. projectile distance:** Similar across velocities
- **Work vs. projectile distance:** Spikes represent ions
- **Electronic work vs. projectile distance:** Slope represents stopping power



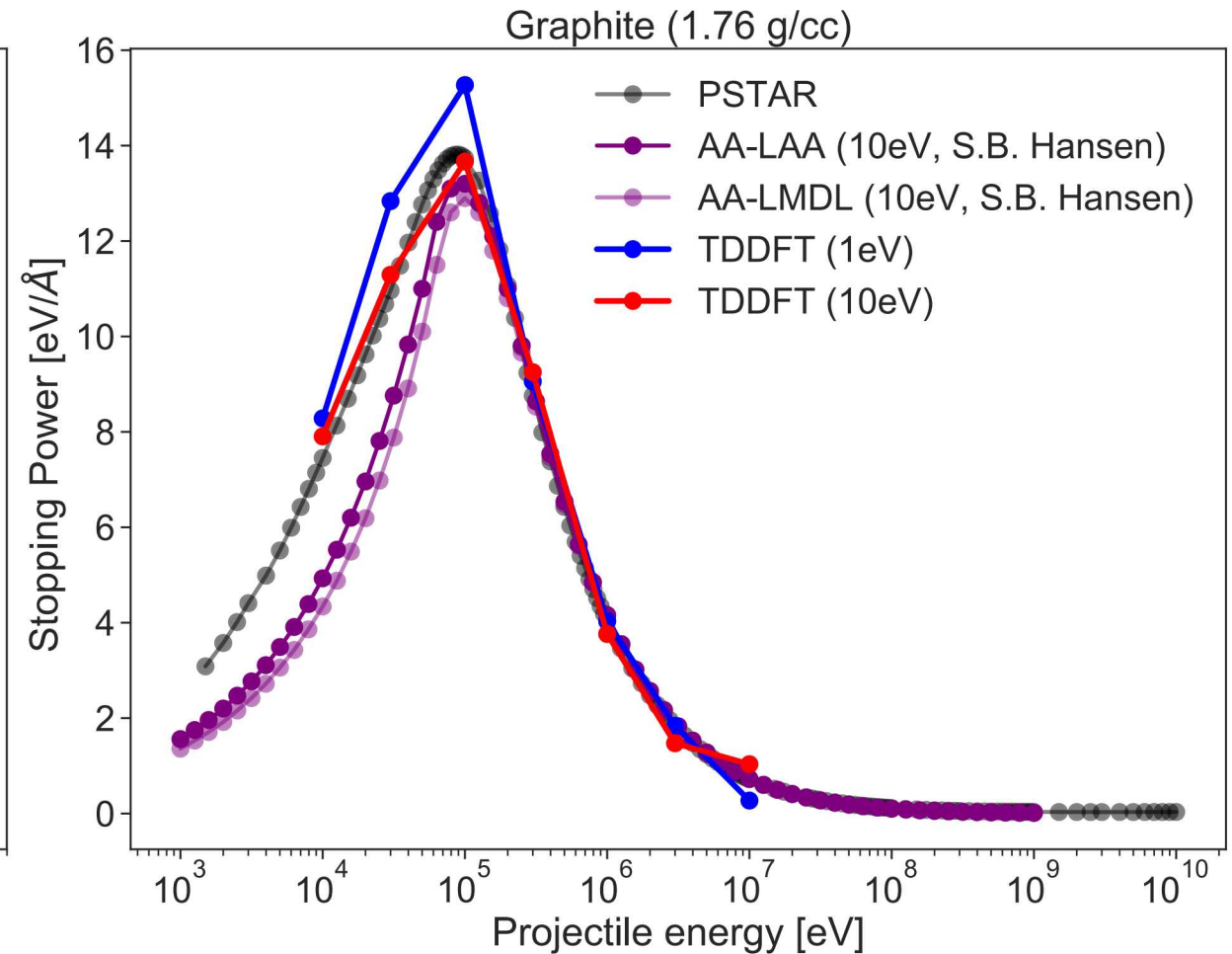
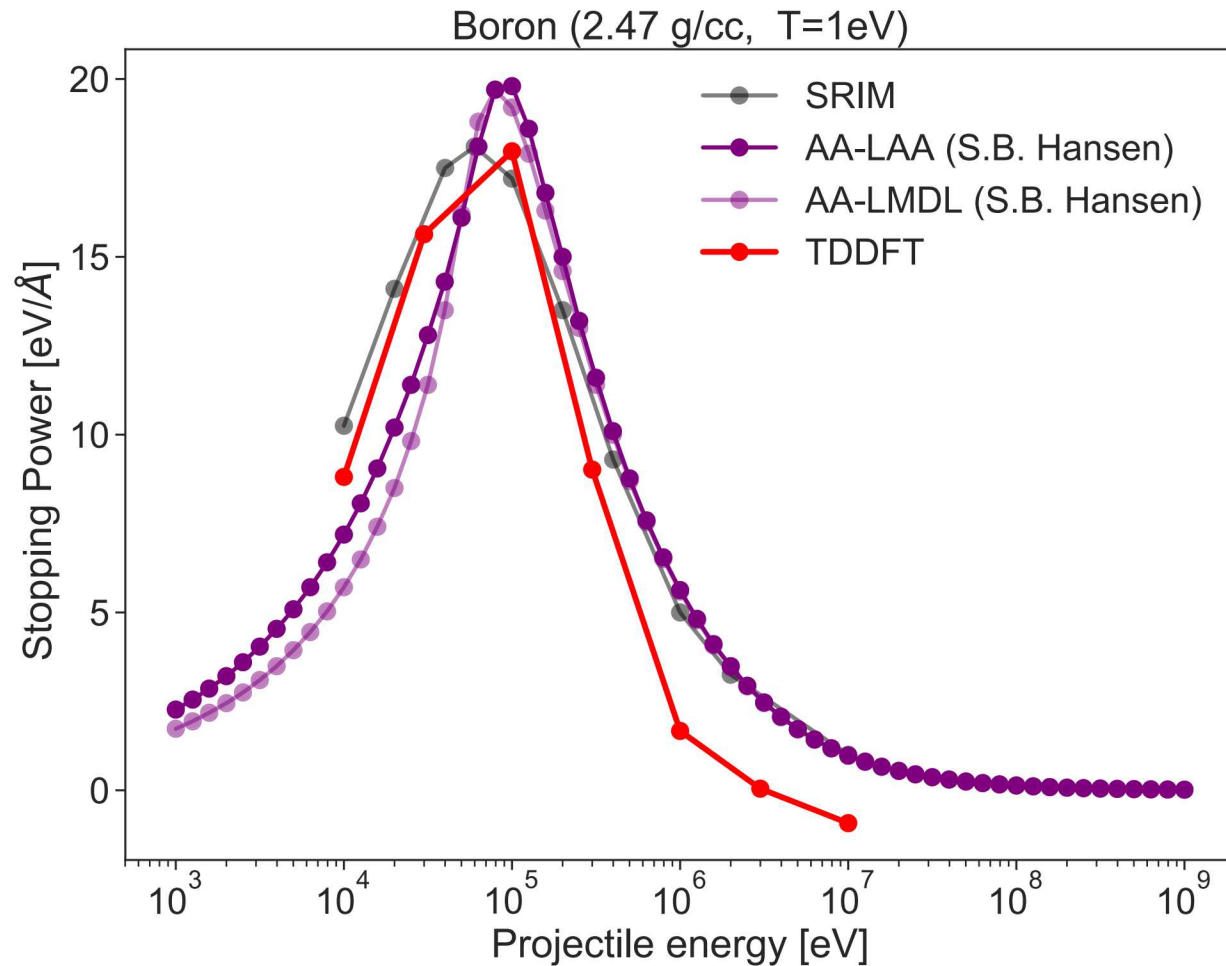
Projectile at 300 keV



Projectile at 30 MeV

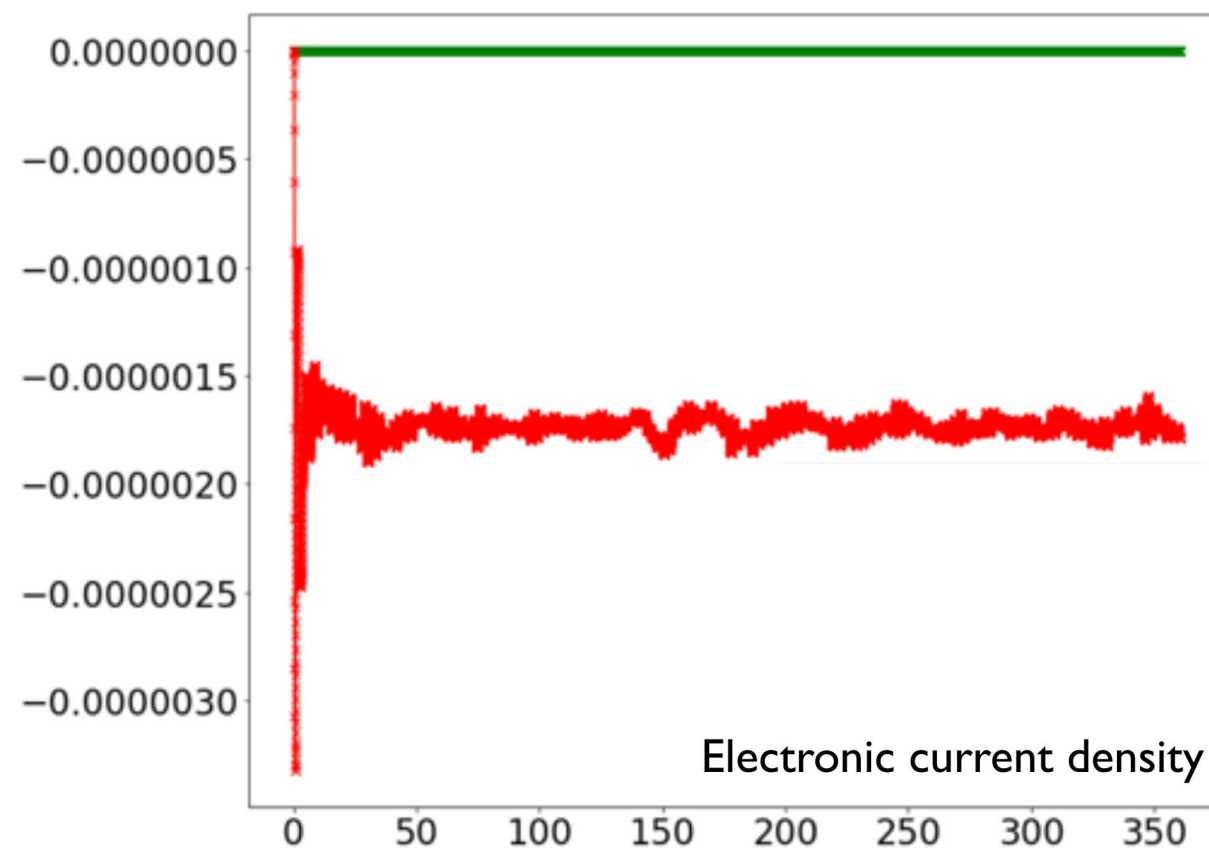
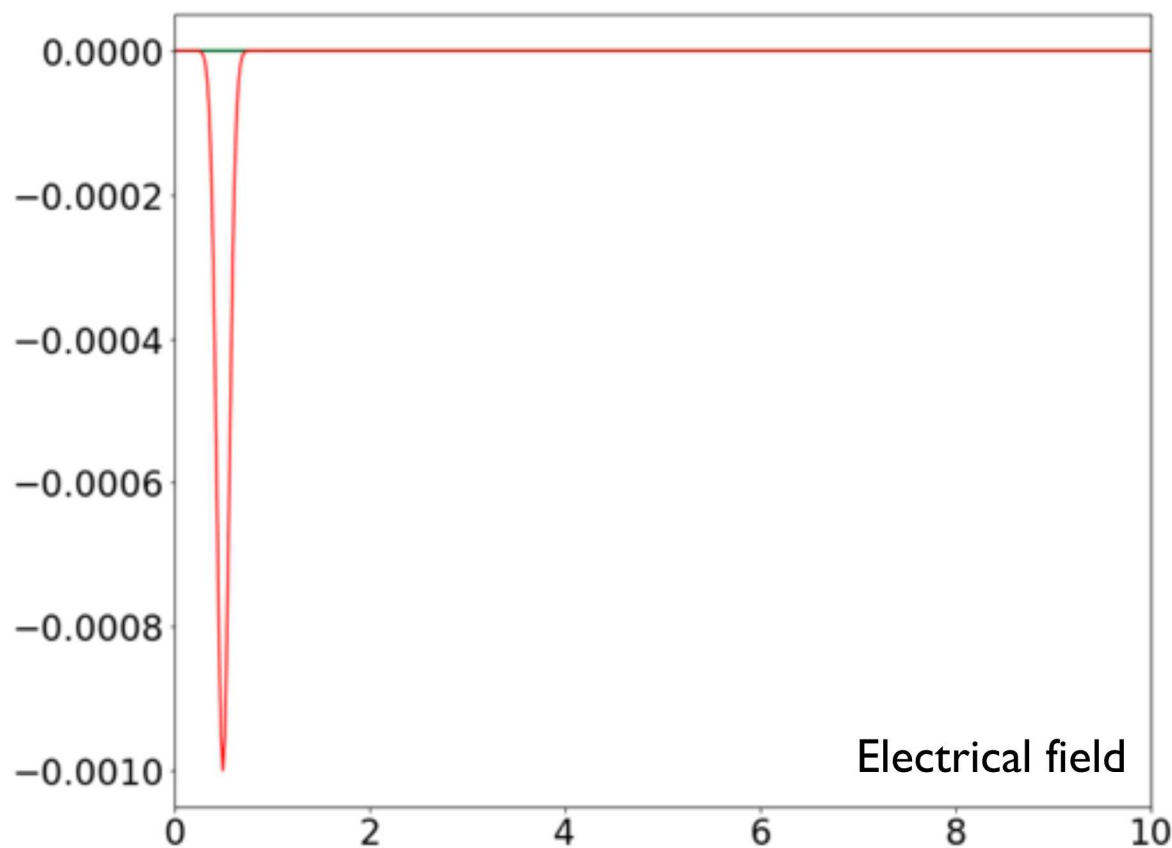


- Stopping power calculations with TDDFT are now routinely available
- We support other simulations tools, such as help in constraining parameters in average-atom models



Linear response: $\epsilon(\omega) = 1 + 4\pi i \frac{\sigma(\omega)}{\omega}$

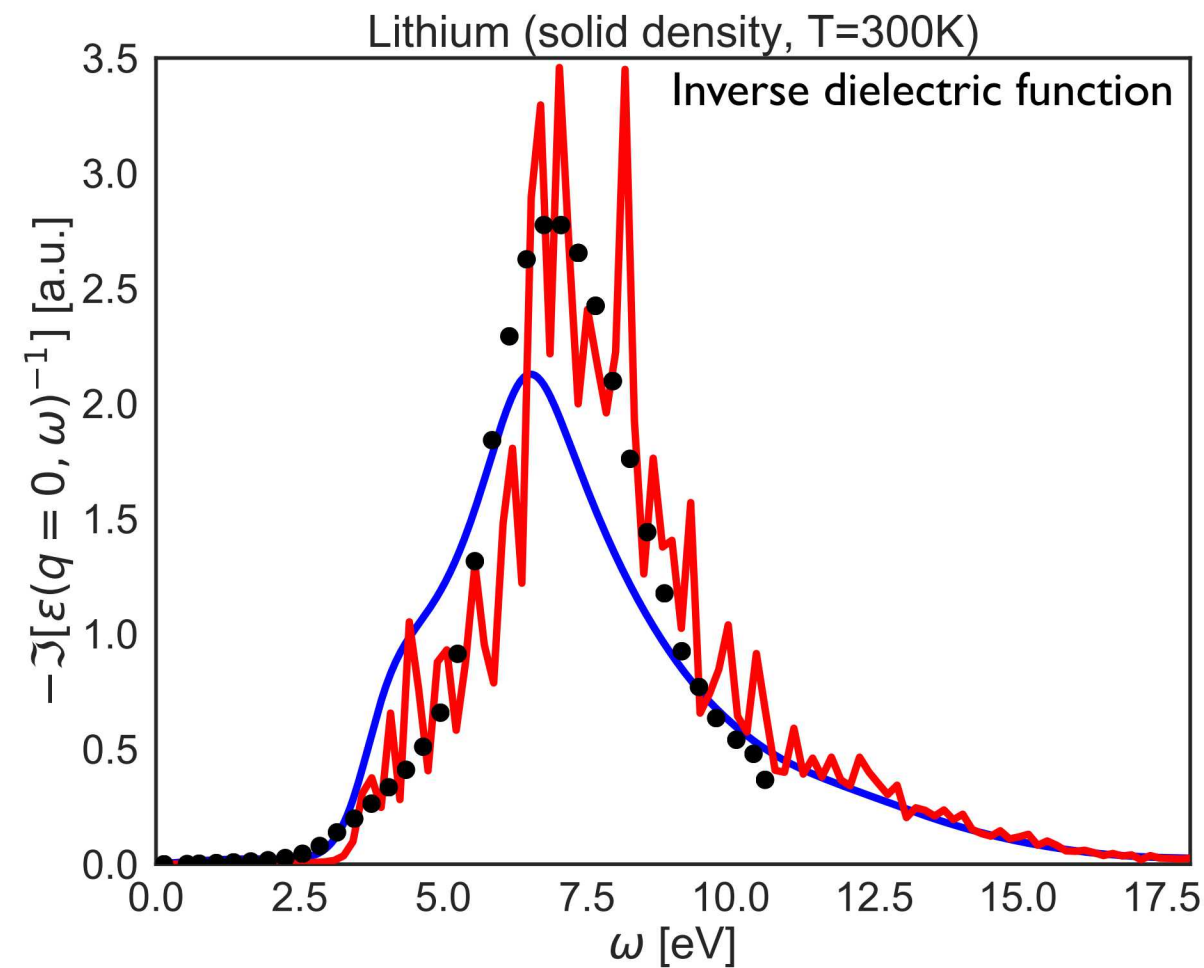
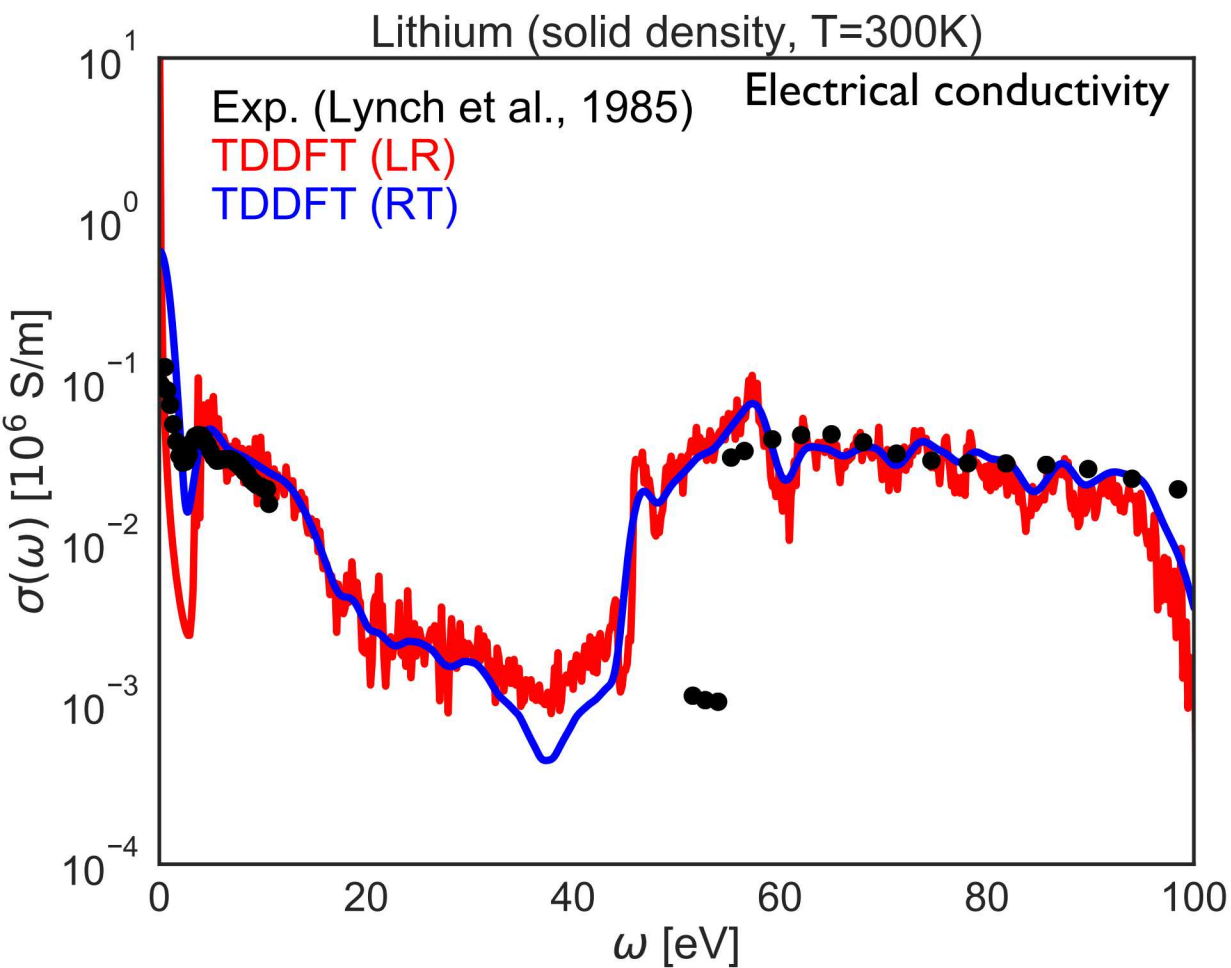
Real-time: $\mathbf{E}(t) = -\frac{1}{c} \frac{\mathbf{A}(t)}{dt}$, $\mathbf{J}(t) = -\frac{i}{\Omega} \int d\mathbf{r} \sum_j \phi_j^*(\mathbf{r}) [\hat{H}(t), \hat{\mathbf{r}}] \phi_j(\mathbf{r})$, $\mathbf{J}(\omega) = \sigma(\omega) \mathbf{E}(\omega)$



Baseline (simple metal)

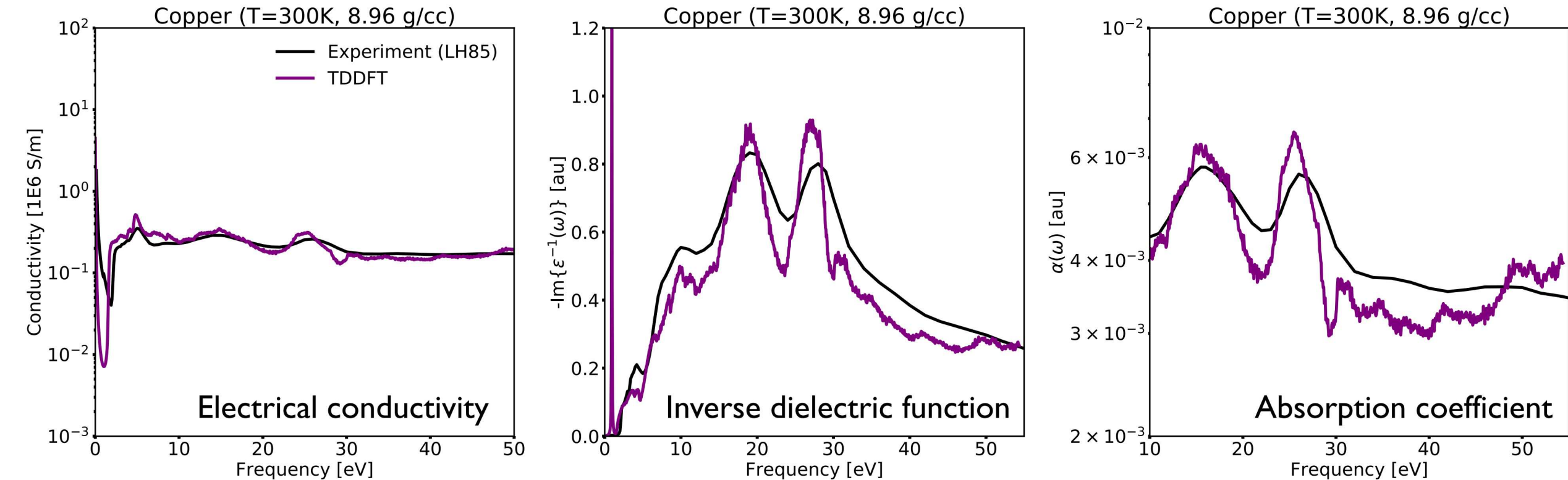
Reference data:

Handbook of Optical Constants of Solids, Academic Press (1998)



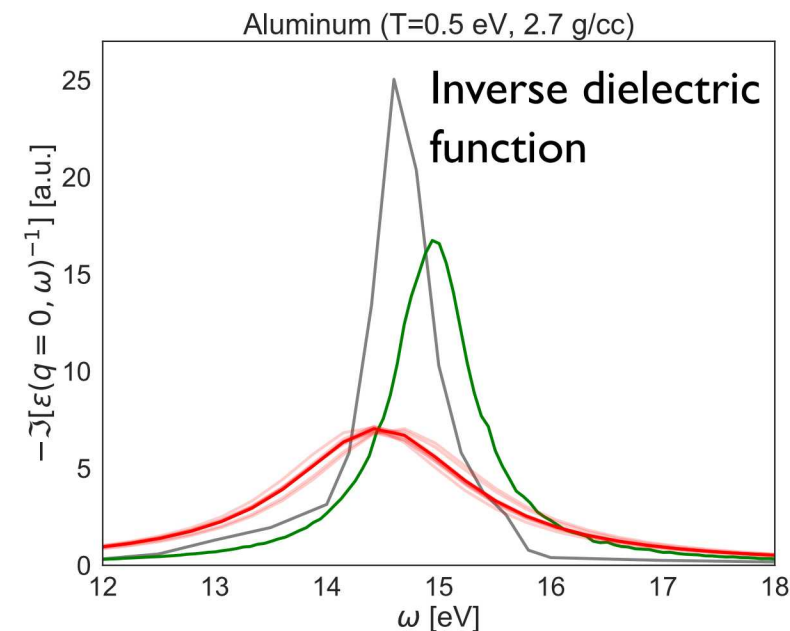
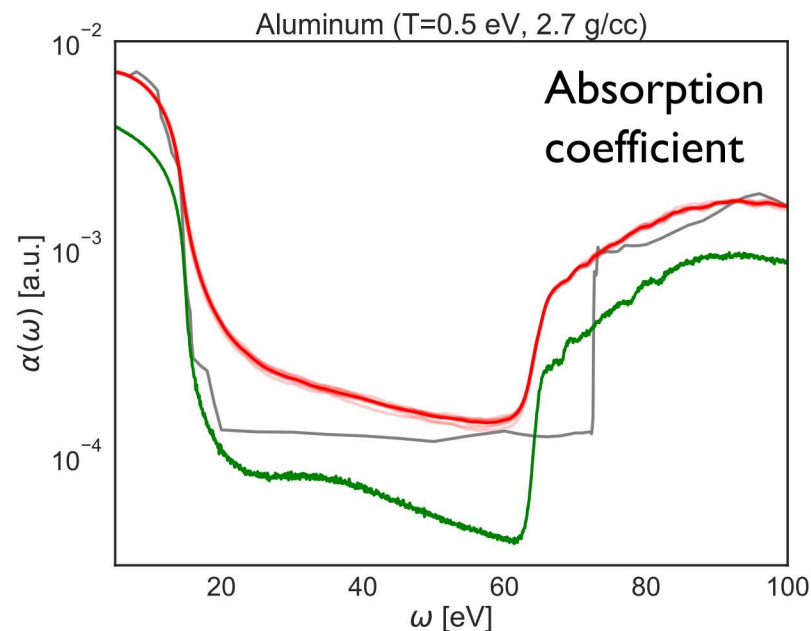
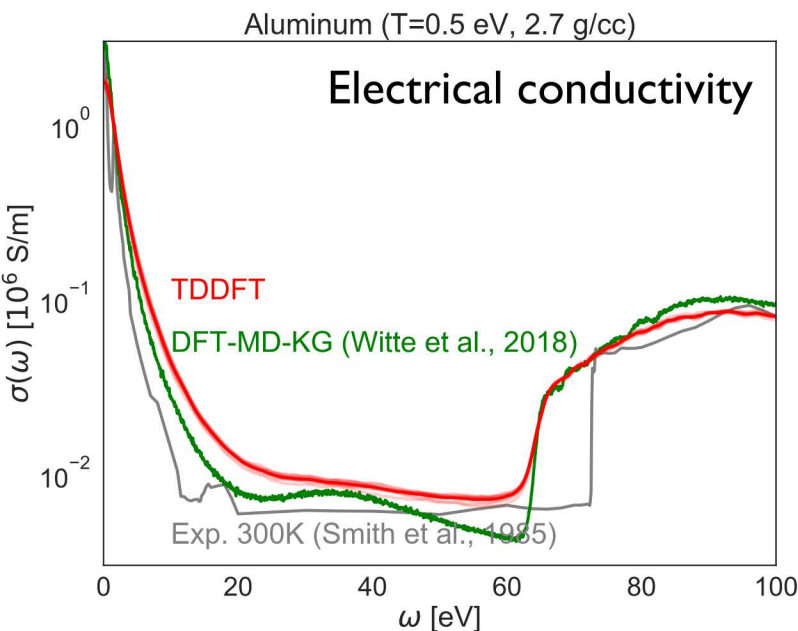
Copper under ambient conditions

Baseline (a somewhat more complex metal)



$$\lim_{q \rightarrow 0} \int_0^\infty d\omega \frac{1}{\omega} \Im \left[\frac{1}{\epsilon(q, \omega)} \right] = -\frac{\pi}{2}$$

	Sum rule	Error	Relative error
Experiment	-1.50	-0.07	0.04
TDDFT (I)	-1.41	-0.15	0.10



$$\lim_{q \rightarrow 0} \int_0^\infty d\omega \frac{1}{\omega} \Im \left[\frac{1}{\epsilon(q, \omega)} \right] = -\frac{\pi}{2}$$

	Sum rule	Error	Relative error
Experiment	-1.577	0.007	-0.004
DFT-MD-KG	-1.568	-0.003	0.002
TDDFT	-1.563	-0.007	0.005

Avoid Chihara decomposition:

$$S(\mathbf{q}, \omega) = \underbrace{|f_I(\mathbf{q}) + \rho(\mathbf{q})|^2 S_{ii}(\mathbf{q}, \omega)}_{\text{Bound electrons following ions}} + \underbrace{Z_f S_{ee}(\mathbf{q}, \omega)}_{\text{Free electrons}} + \underbrace{S_{bf}(\mathbf{q}, \omega)}_{\text{Photoionized electrons}}$$

Probe system with x-ray:

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

$$|\mathbf{q}| = \frac{4\pi \sin(\theta/2)}{\lambda_0}$$

λ_0 : probe wavelength (2Å)

Record density response:

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

Apply dissipation-fluctuation theorem:

$$\chi(\mathbf{q}, -\mathbf{q}, \omega) = \frac{\delta n(\mathbf{q}, \omega)}{v_0 f(\omega)} \quad S(\mathbf{q}, \omega) = -\frac{1}{\pi} \frac{\Im[\chi(\mathbf{q}, -\mathbf{q}, \omega)]}{1 - e^{-\omega/T}}$$

PRL 116, 115004 (2016)

PHYSICAL REVIEW LETTERS

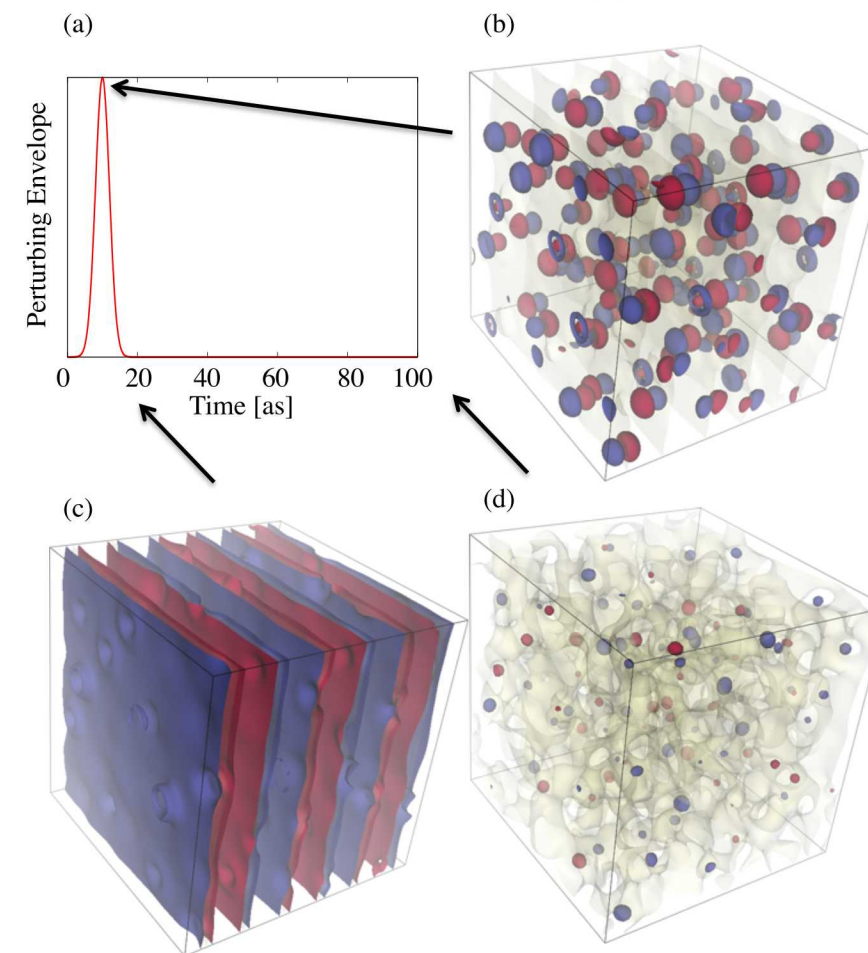
week ending
18 MARCH 2016

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

A. D. Baczewski,^{1,*} L. Shulenburger,² M. P. Desjarlais,² S. B. Hansen,² and R. J. Magyar¹

¹Center for Computing Research, Sandia National Laboratories, Albuquerque, New Mexico 87185, USA

²Pulsed Power Sciences Center, Sandia National Laboratories, Albuquerque, New Mexico 87185, USA



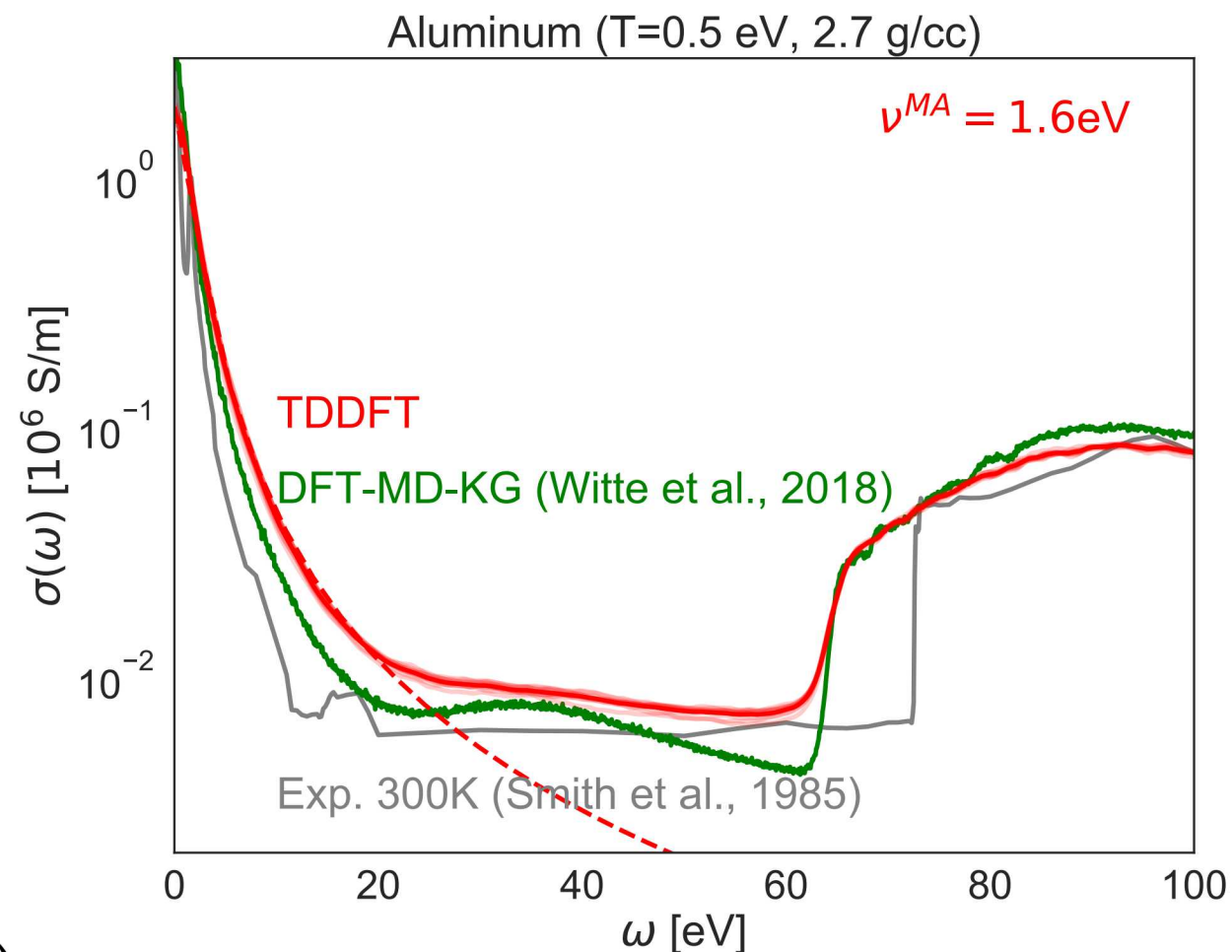
Mermin approximation to the dielectric function:

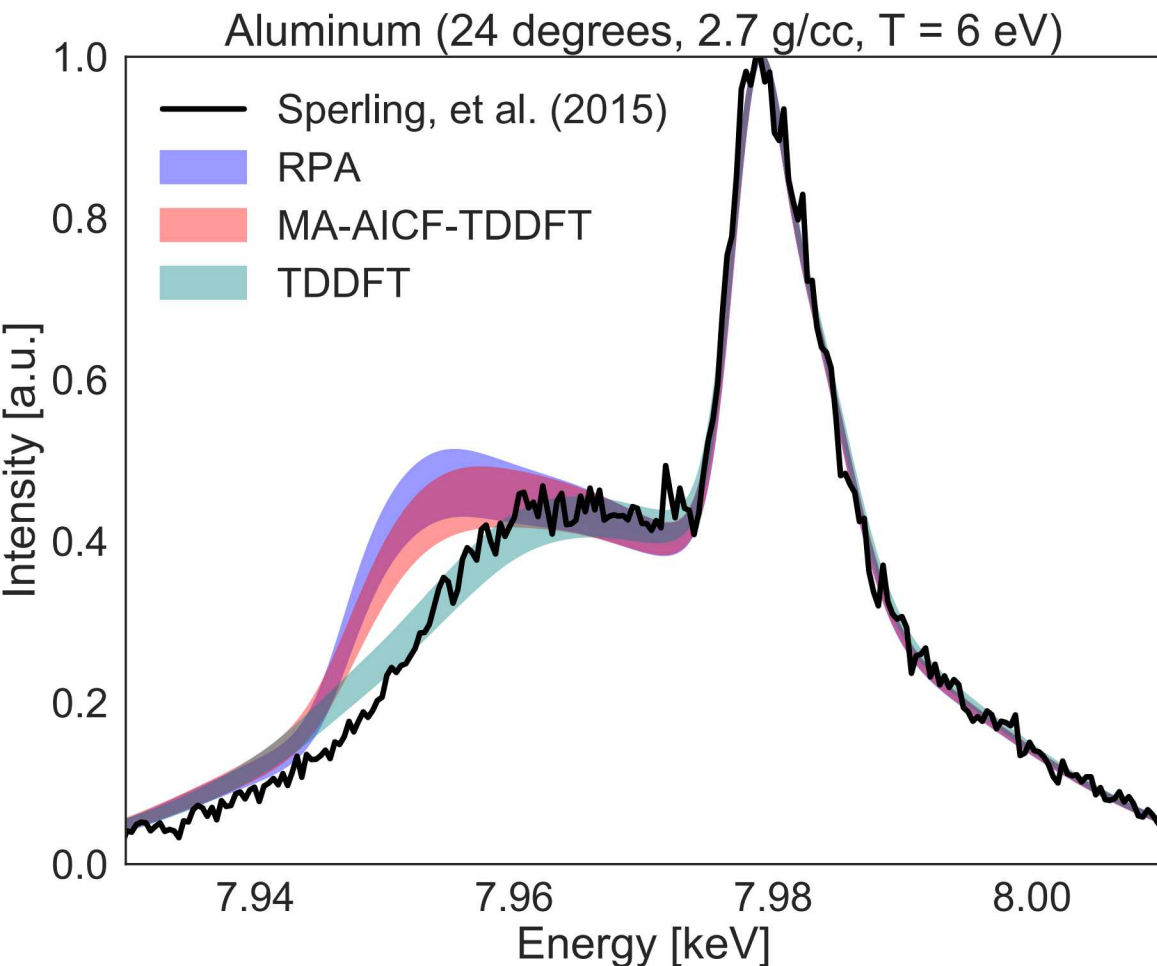
$$\epsilon^M(\mathbf{q}, \omega) = 1 + \frac{\left(1 + i\frac{\nu(\omega)}{\omega}\right) [\epsilon^{RPA}(\mathbf{q}, \omega + i\nu(\omega)) - 1]}{1 + \frac{\nu(\omega)}{\omega} \frac{\epsilon^{RPA}(\mathbf{q}, \omega + i\nu(\omega)) - 1}{\epsilon^{RPA}(\mathbf{q}, 0) - 1}}$$

Ab-initio collision frequencies from Drude-fit to TDDFT electrical conductivity.

$$\Re(\sigma^D) = \frac{\lim_{\omega \rightarrow 0} \sigma(\omega)}{1 + \omega^2 \tau^2}$$

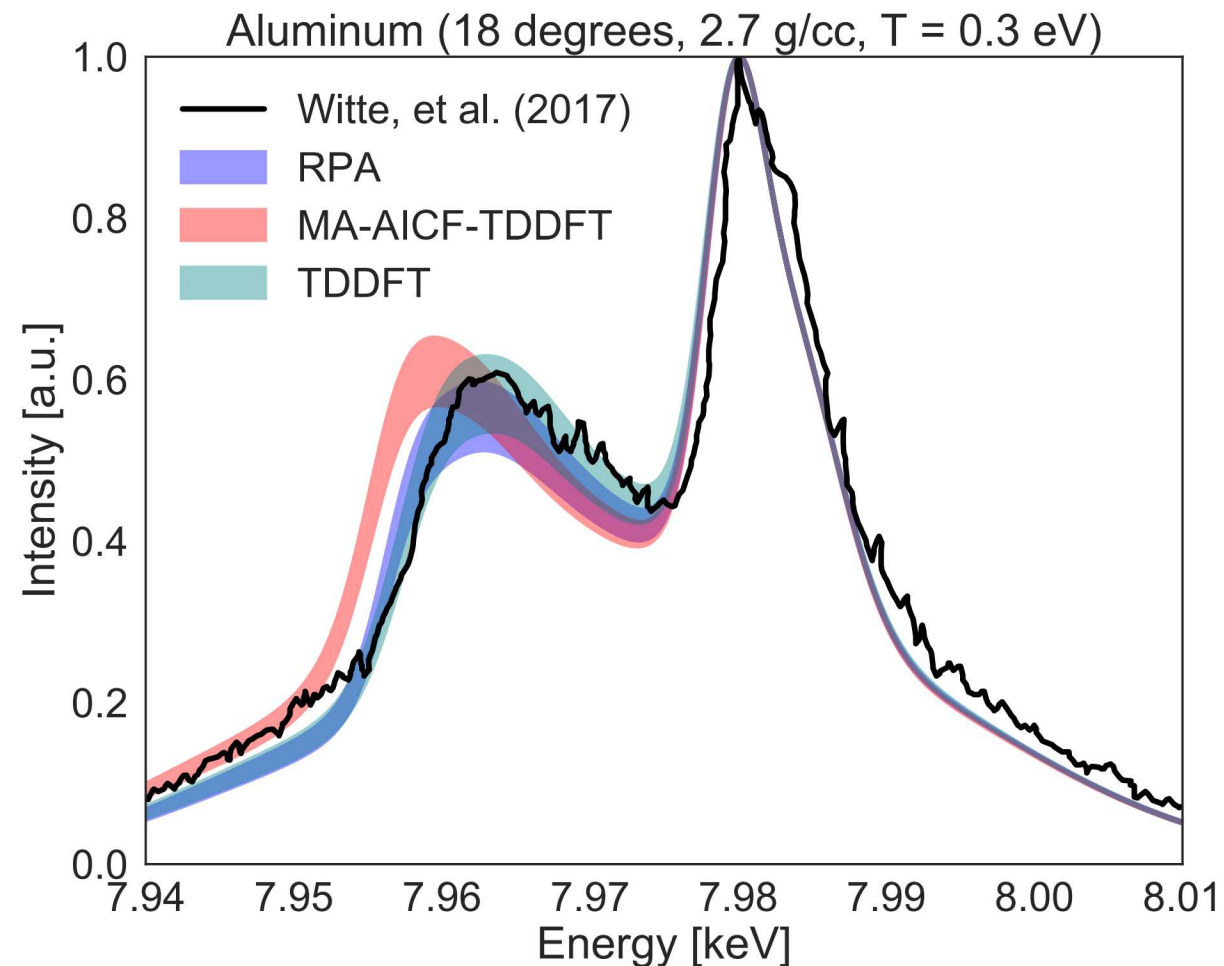
Mermin approximation with ab-initio collision frequencies from TDDFT (**MA-AICF-TDDFT**).





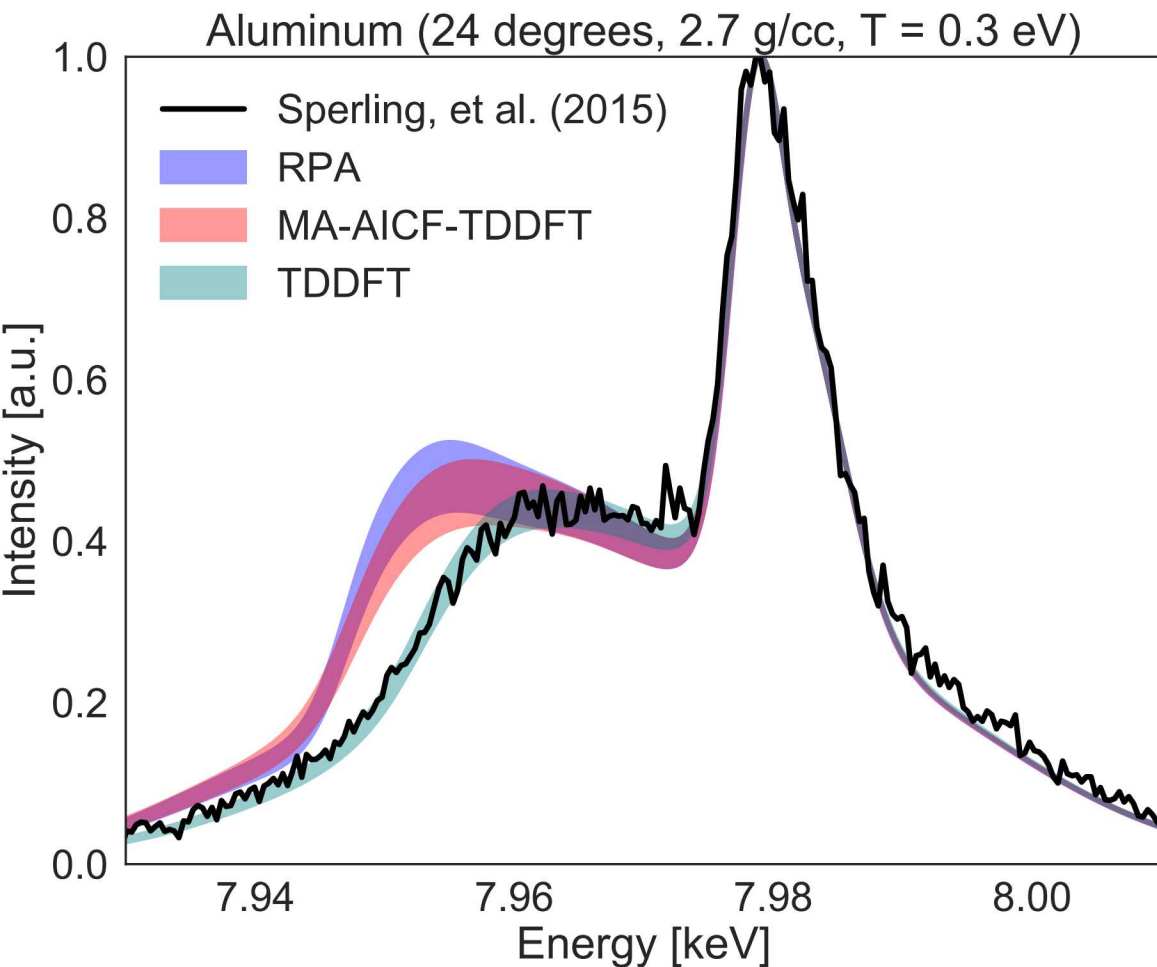
Free-Electron X-Ray Laser Measurements of Collisional-Damped Plasmons in Isochorically Heated Warm Dense Matter

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



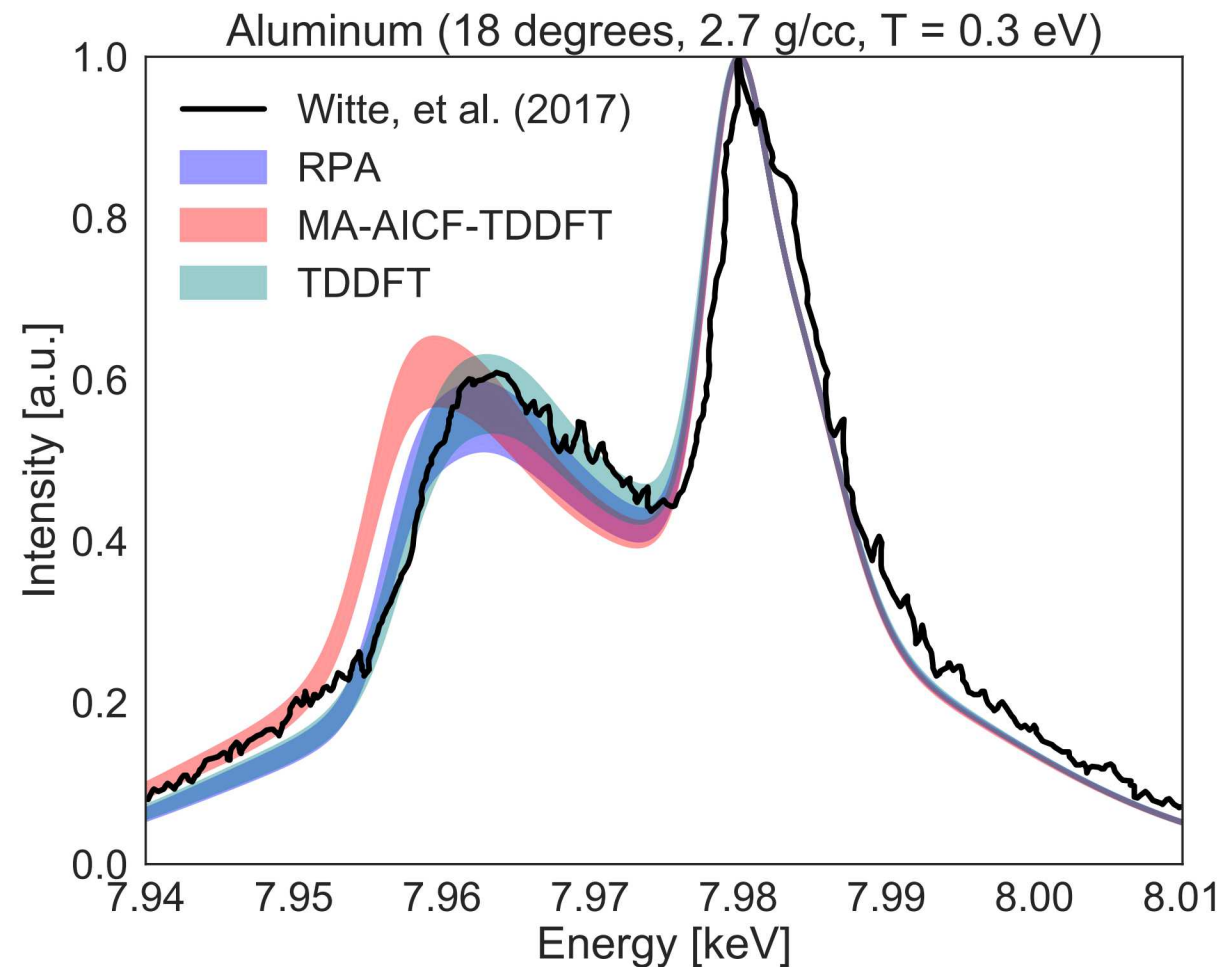
Warm Dense Matter Demonstrating Non-Drude Conductivity from Observations of Nonlinear Plasmon Damping

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



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 Phys. Rev. Lett. **118**, 225001 – Published 31 May 2017

Capability to predict first-principles transport properties of HED from TDDFT

- Systematic approach with minimal need for post-processing
- Better scaling in real time than in energy domain
- Nonlinear effects

Support other simulation tools

- Constrain parameters in average-atom models
- Validate DFT-MD Kubo-Greenwood results
- Input for resistive magneto-hydrodynamics

Support interpretation of experiments

- Combine TDDFT tools to provide consistent predictions
- Recent experiments at LCLS
- Isochorically heated and shock-compressed materials (such as Al and Cu)

Collaborators

- Andrew D. Baczewski (Sandia National Laboratories)
- Stephanie B. Hansen (Sandia National Laboratories)

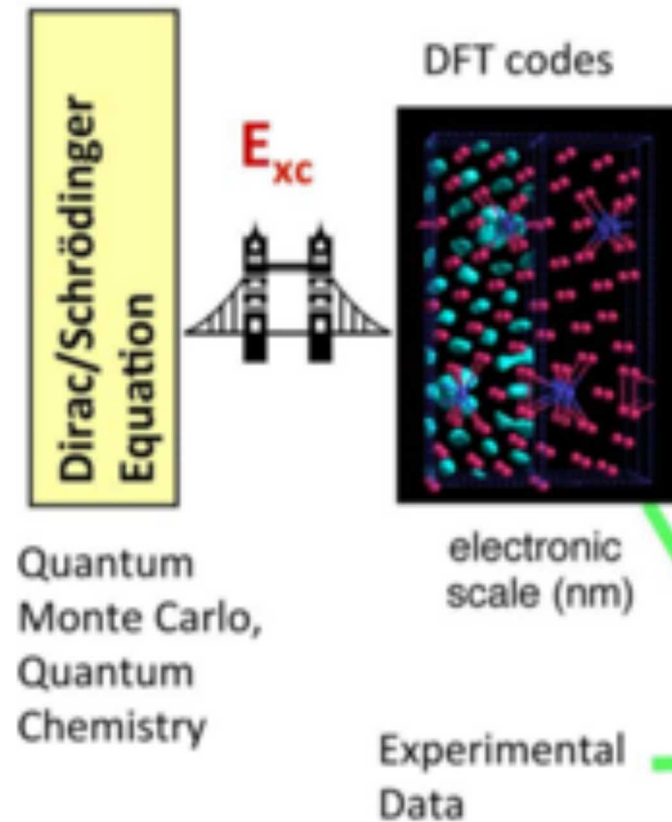
Upcoming

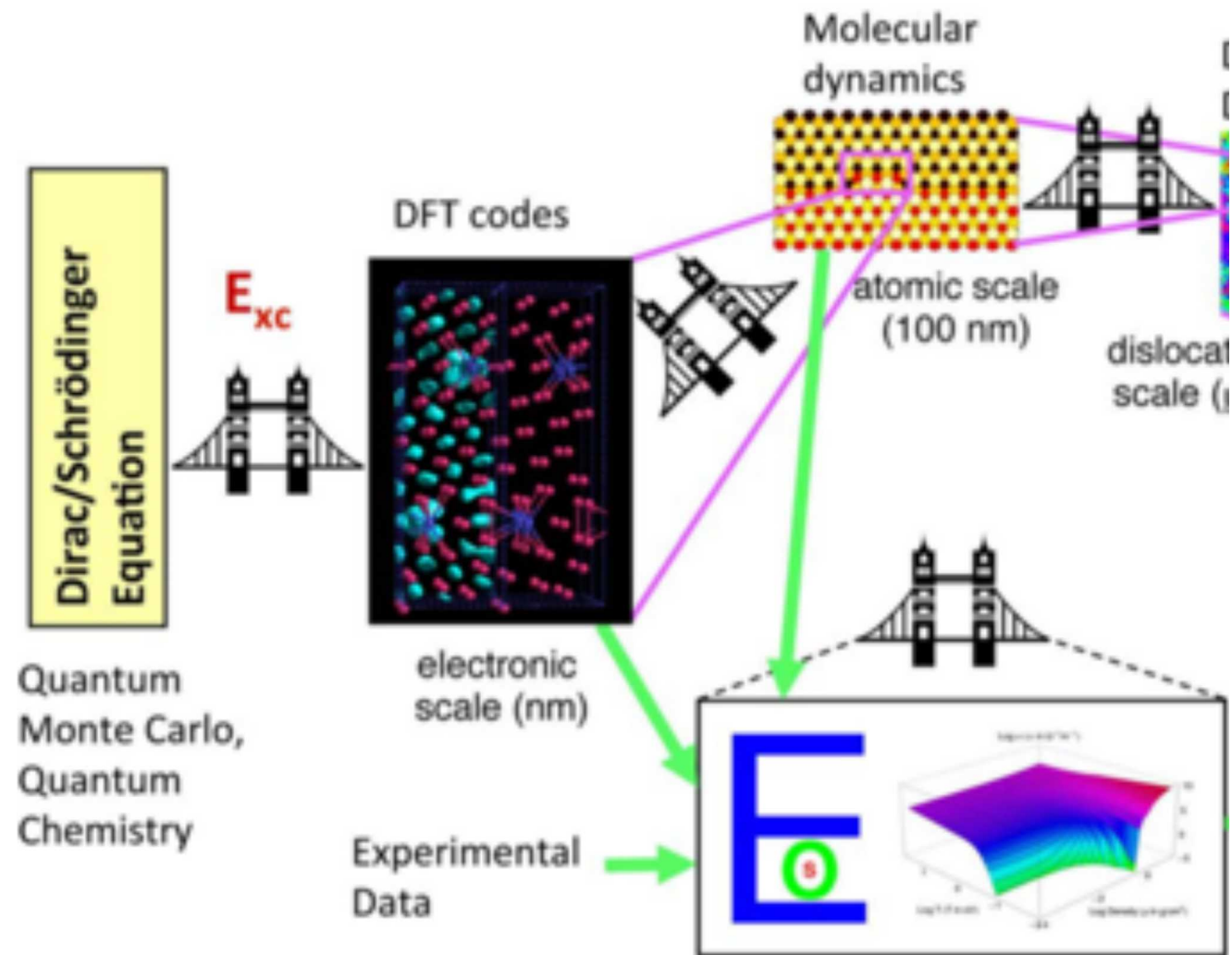


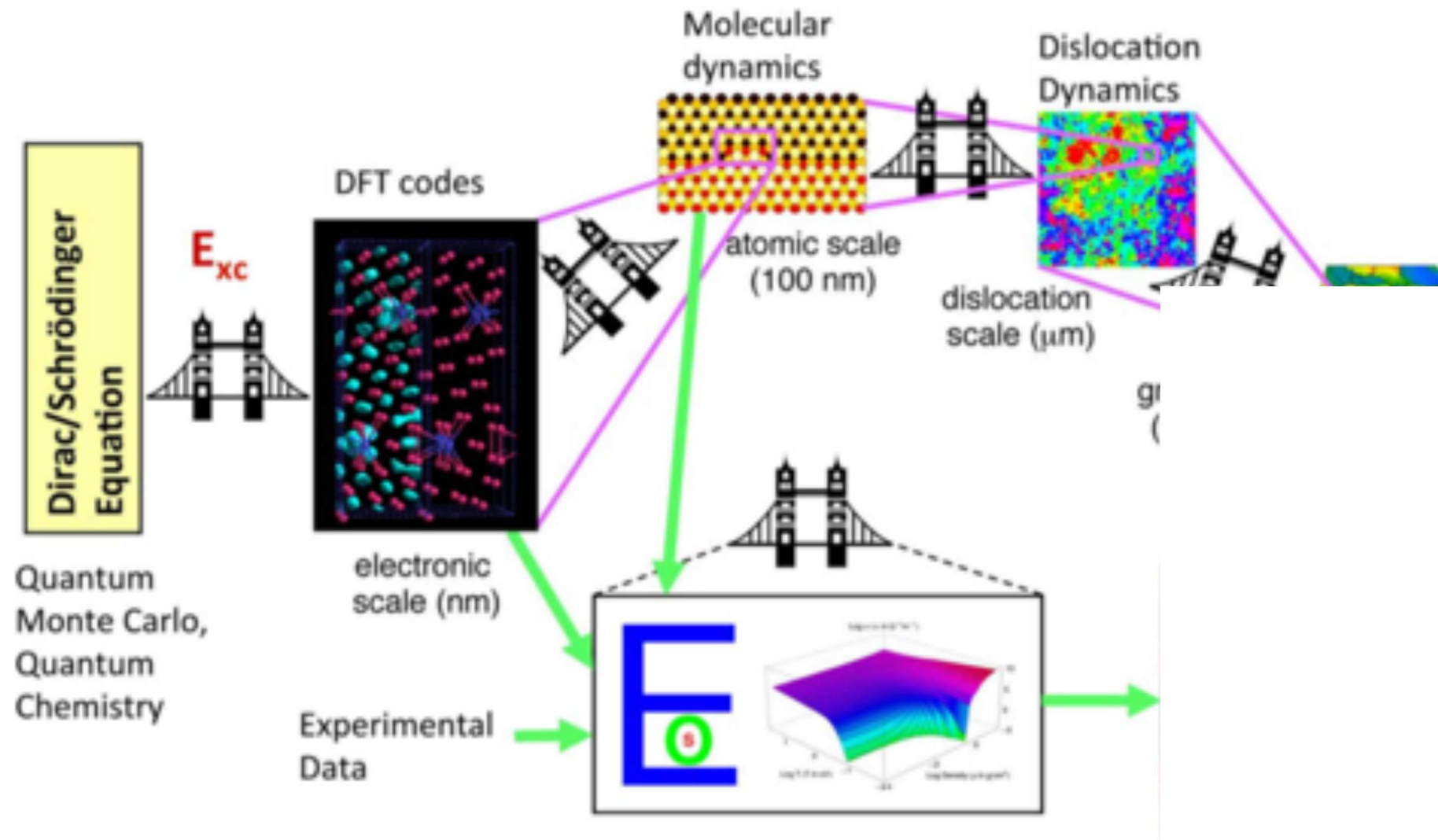
Quantum-Accurate Multiscale Modeling of Shock Hugoniot, Ramp Compression Paths, Structural and Magnetic Phase Transitions, and Transport Properties in Highly Compressed Metals

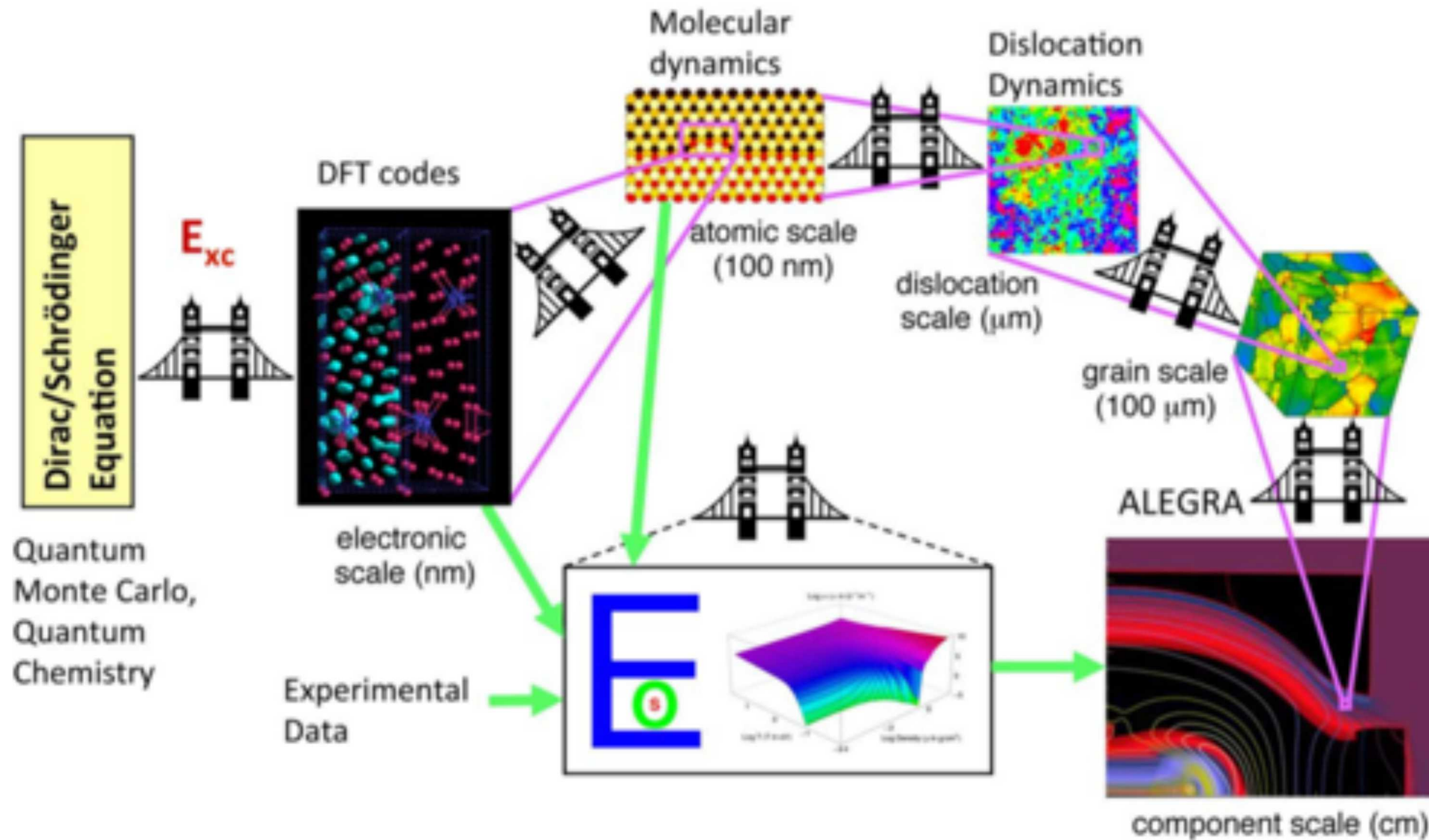
Thank you for your attention!

Supplemental Material

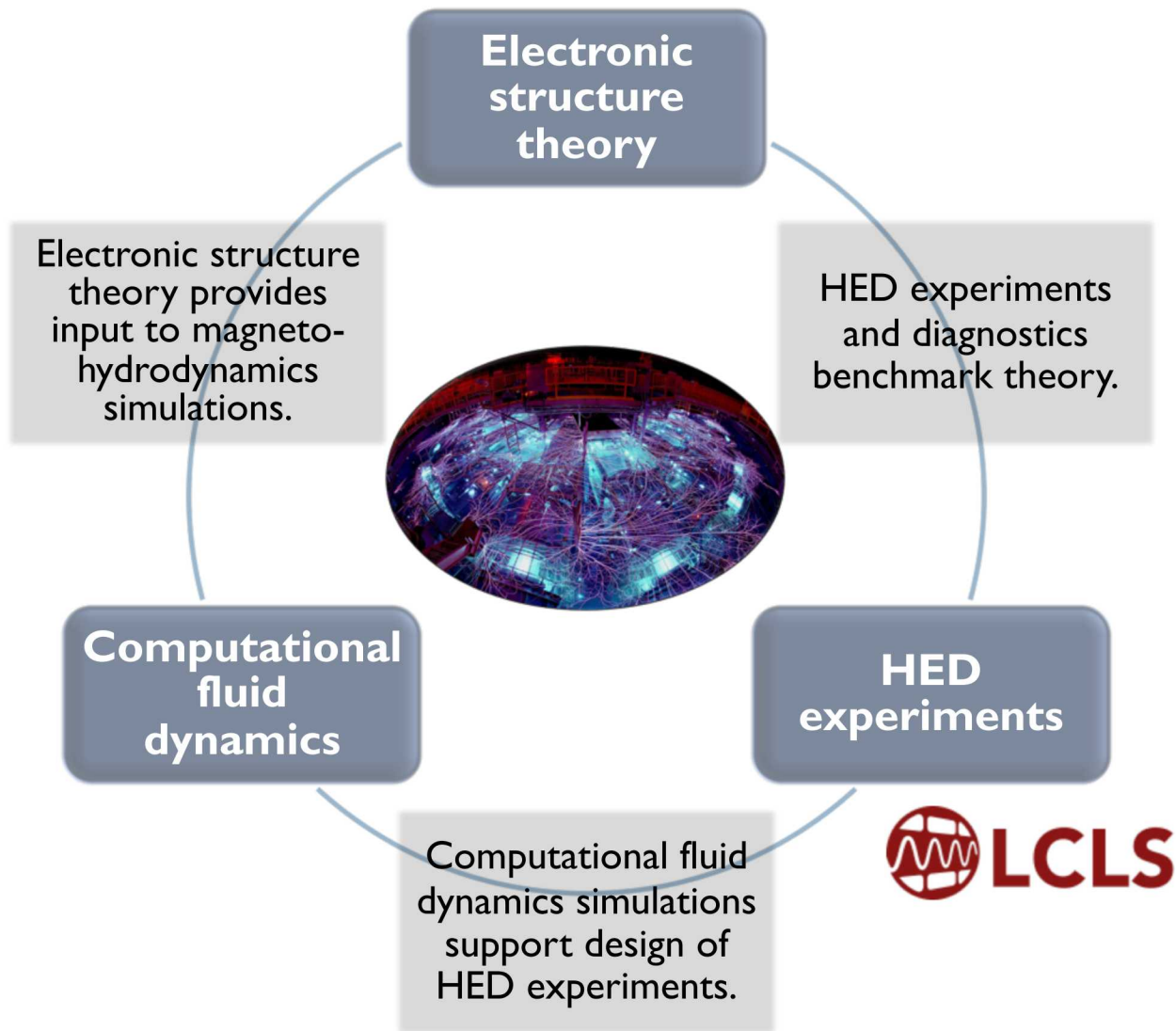








Ann E. Mattson et al., International Journal of Quantum Chemistry (2016), DOI: 10.1002/qua.25097.



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

$$\frac{\partial E}{\partial x} = \frac{1}{v} \frac{\partial}{\partial t} \langle E \rangle$$

Electrical conductivity

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

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Phys. Rev. Lett. **115**, 115001 – Published 9 September 2015

$$\sigma(\omega) = \frac{\mathbf{J}(\omega)}{\mathbf{E}(\omega)}$$

Dynamic structure factor

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, G. A. Rochau¹, C. Blancard², J. Colgan³, Ph. Coe², G. Faussurier², C. J. Fontes³, F. Gilleney⁴, I. Golovkin⁴, S. B. Hansen¹, C. A. Iglesias⁴, D. P. Kikrease¹, J. J. MacFarlane⁵, R. C. Mancini⁶, S. N. Nahar⁷, C. Orban⁸, J.-C. Pain⁹, A. K. Pradhan¹, M. Sherrill¹⁰ & B. G. Wilson¹

$$S(\mathbf{q}, \omega) = -\frac{1}{\pi} \frac{\Im[\chi(\mathbf{q}, -\mathbf{q}, \omega)]}{1 - e^{-\omega/k_B T}}$$

Coulomb coupling parameter

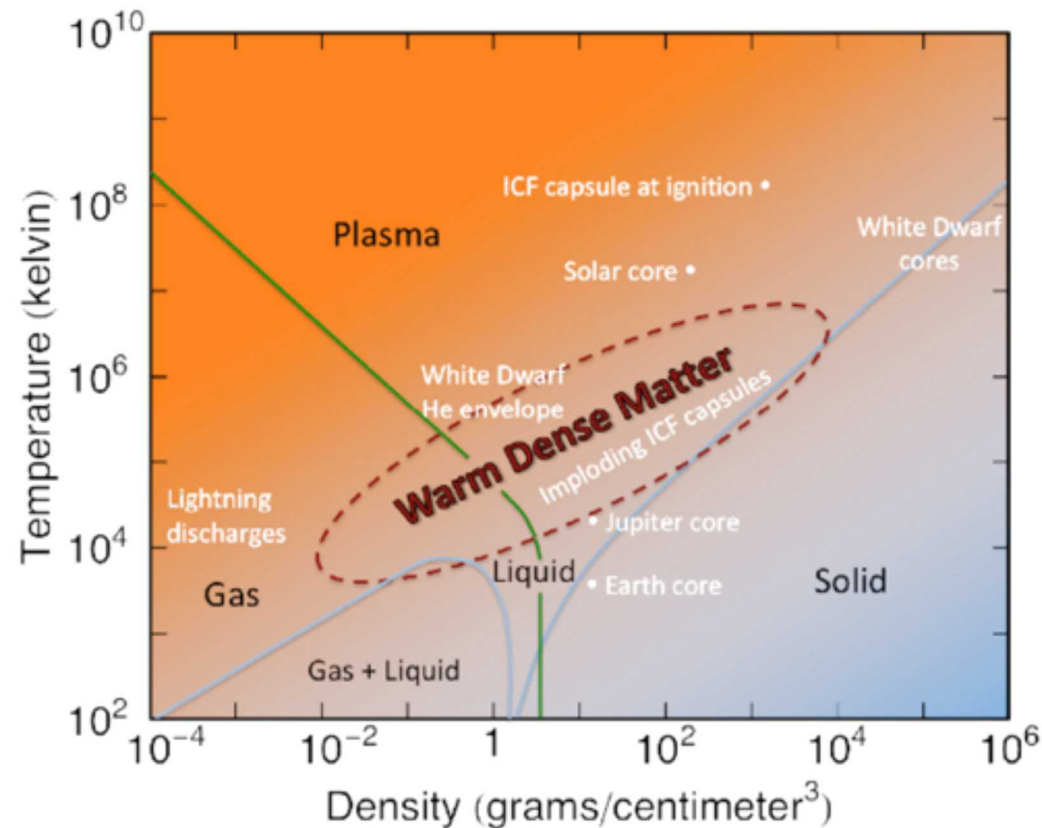
$$\Gamma = \frac{V}{T} = \frac{Ze^2}{r_S k_B \tau}$$

Electron degeneracy parameter

$$\Theta = \frac{k_B \tau}{E_F}$$

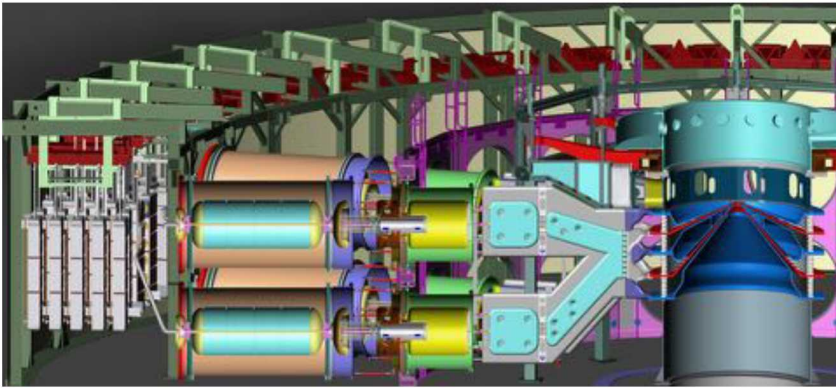
Warm dense matter regime

$$\Gamma \approx 1, \quad \Theta \approx 1$$



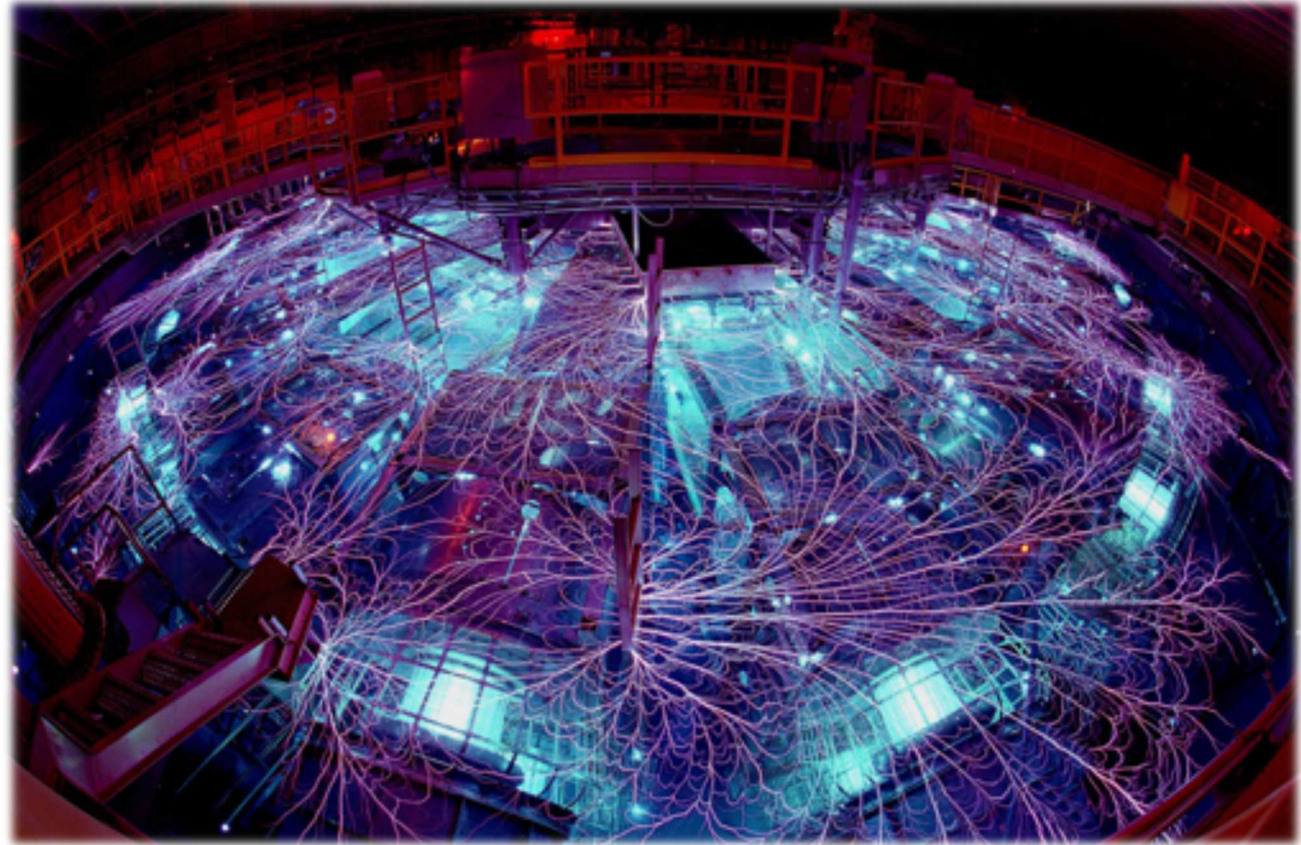
Basic Research Needs for HEDLP, Report of the
Workshop on HEDLP Research Needs, DOE (2008).

Creating HED conditions requires transferring an enormous amount of energy to a target in a very short period of time.

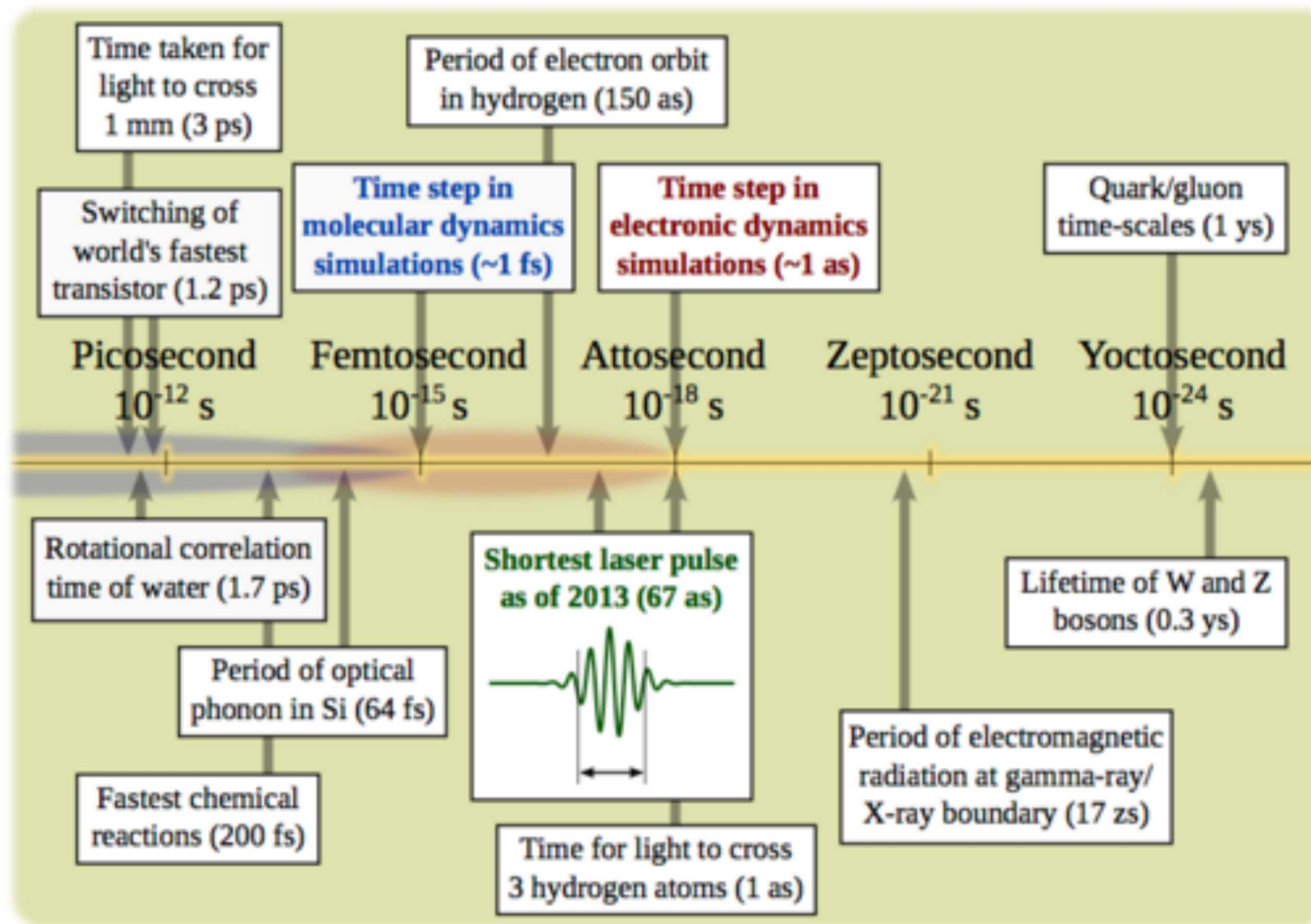


Z Machine, Sandia National Laboratories.
(https://www.sandia.gov/z-machine/about_z/how-z-works.html)

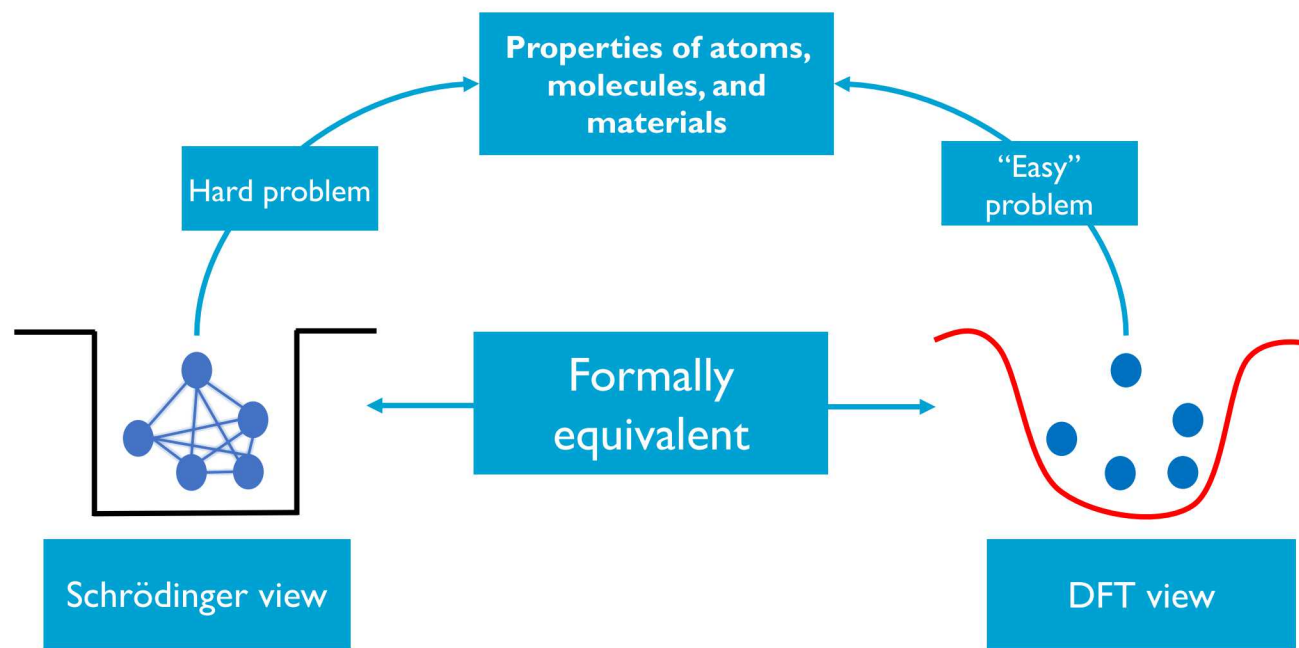
Compression of energy in time and space in pulsed power facilities (Z machine) enables exciting science (astrophysics, planetary science, inertial confinement fusion).



Z Machine, Sandia National Laboratories.



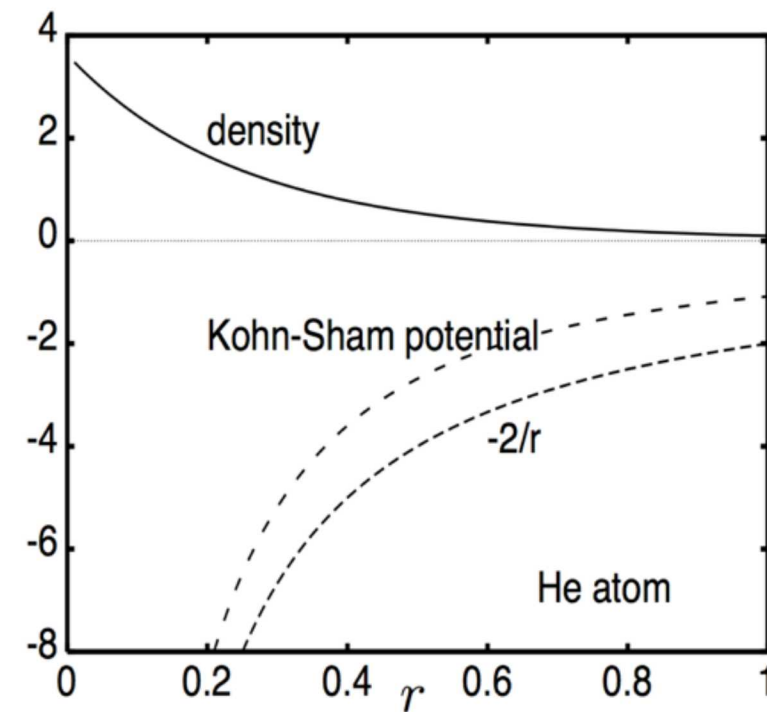
Courtesy of K. Dewhurst, Max Planck Institute of Microstructure Physics (2015).



$$\left[-\frac{\nabla^2}{2} + v_S(\mathbf{r}) \right] \phi_j(\mathbf{r}) = \epsilon_j \phi_j(\mathbf{r}) \quad (\text{Kohn-Sham equations})$$

$$v_S(\mathbf{r}) = v(\mathbf{r}) + \frac{\delta U}{\delta n(\mathbf{r})} + \frac{\delta E_{xc}}{\delta n(\mathbf{r})} \quad (\text{Kohn-Sham potential})$$

$$n(\mathbf{r}) = \sum_i \phi_i^*(\mathbf{r}) \phi_i(\mathbf{r}) \quad (\text{Electronic density})$$



The ABC of DFT (dft.uci.edu/doc/gl.pdf).

$$\chi(\omega) = \chi_1(\omega) + i \chi_2(\omega)$$

$$\chi_1(\omega) = \frac{1}{\pi} \mathcal{P} \int_{-\infty}^{\infty} d\omega' \frac{\chi_2(\omega')}{\omega' - \omega}$$

$$\chi_2(\omega) = \frac{1}{\pi} \mathcal{P} \int_{-\infty}^{\infty} d\omega' \frac{\chi_1(\omega')}{\omega' - \omega}$$

$$\epsilon(\omega) = 1 + 4\pi i \frac{\sigma}{\omega}$$

$$\alpha(\omega) = \frac{2}{c} \omega \Im \left[\sqrt{\epsilon(\omega)} \right]$$

$$\mathbf{E}(t) = -\frac{1}{c} \frac{d\mathbf{A}(t)}{dt}$$

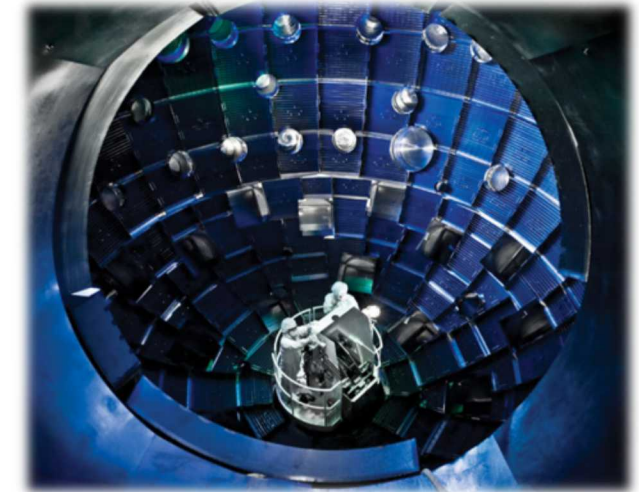
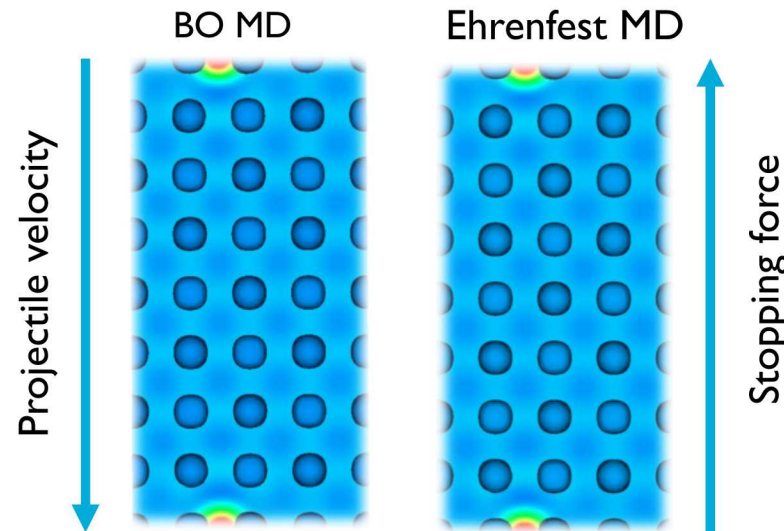
$$\mathbf{J}(t) = -\frac{i}{\Omega} \int d\mathbf{r} \sum_j \phi_j^*(\mathbf{r}) [\hat{H}(t), \hat{\mathbf{r}}] \phi_j(\mathbf{r})$$

$$\mathbf{J}(\omega) = \sigma(\omega) \mathbf{E}(\omega)$$

$$\begin{aligned} \sigma_{\mathbf{k}}(\omega) = & \frac{2\pi}{3\omega\Omega} \sum_{i,j} [f(\epsilon_{i,\mathbf{k}}) - f(\epsilon_{j,\mathbf{k}})] \\ & \times |\langle \phi_{j,\mathbf{k}} | \nabla | \phi_{j,\mathbf{k}} \rangle|^2 \delta(\epsilon_{j,\mathbf{k}} - \epsilon_{i,\mathbf{k}} - \omega) \end{aligned}$$

Example: Hydrogen moving through cold, bulk aluminum in a channeling trajectory

$$\frac{\partial E}{\partial x} = \frac{1}{v} \frac{\partial}{\partial t} \langle E \rangle$$



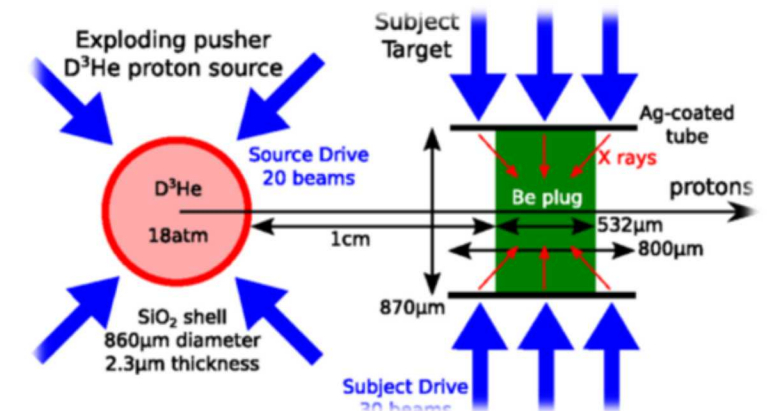
Target chamber, National Ignition Facility, Lawrence Livermore National Laboratories.

Stopping mechanisms

- Nuclear stopping (lattice vibrations)
- Electronic stopping (electronic excitations)

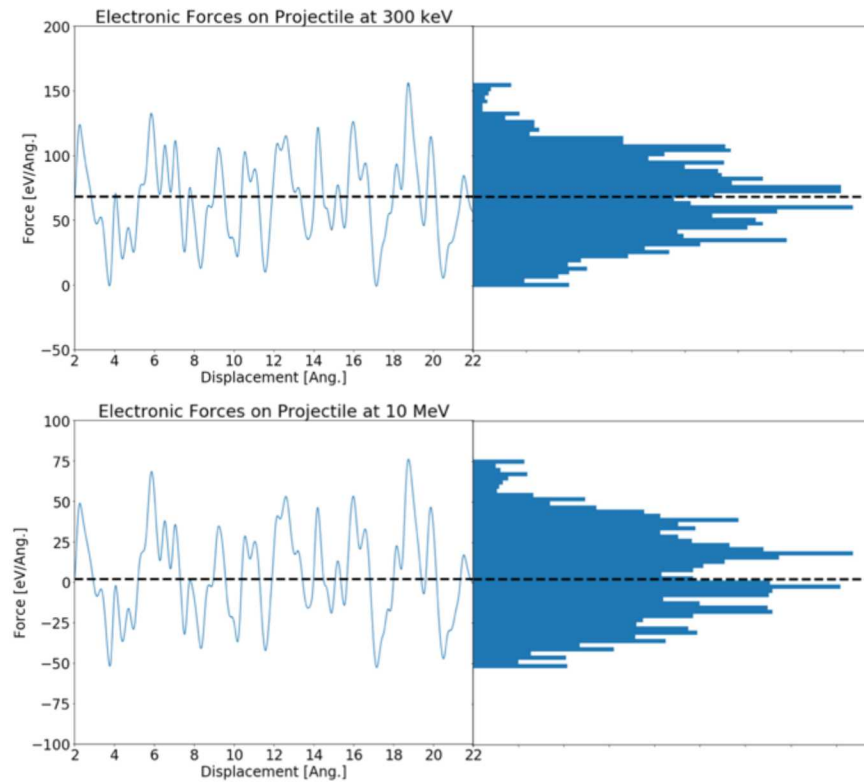
Large body of literature for cold targets

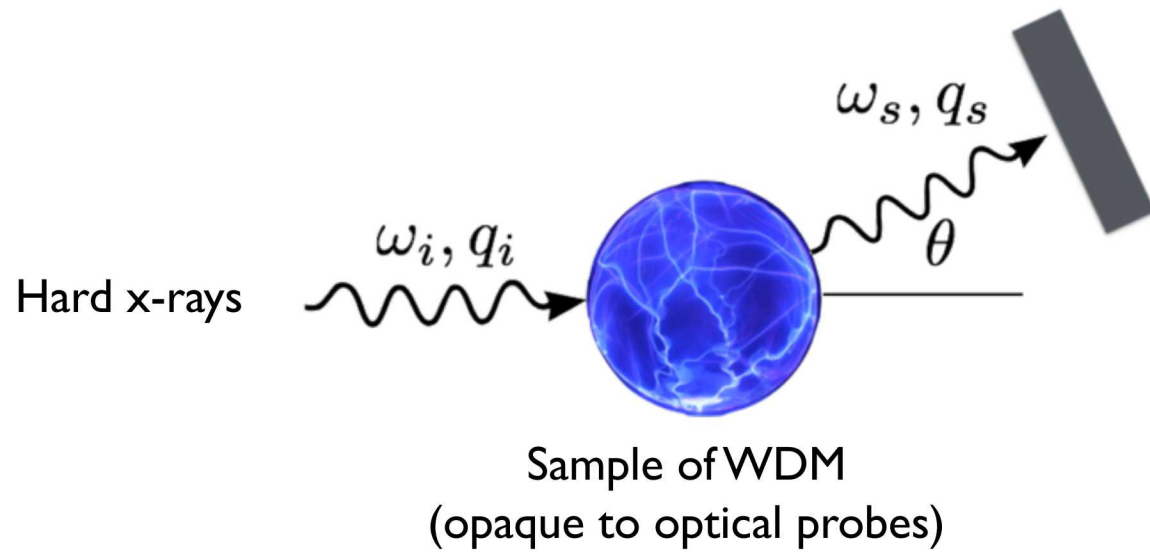
- Empirical approximations (Rutherford, Thomson, Bohr, Bethe)
- Parameter-free atomistic simulations
- Electronic structure coupled to molecular dynamics
- Cold stopping power (Echenique, Correa, Artacho, Schleife)



Zylstra et al., Phys. Rev. Lett. 114, 215002 (2015).

Distribution of microscopic forces in warm dense beryllium





$$\omega = \omega_i - \omega_s$$

$$q = q_i - q_s$$

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

- X-ray Thomson scattering probes density, ionization state, structure, temperature, etc.

X-ray Thomson scattering in high energy density plasmas

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

- Cross section is proportional to dynamic structure factor

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

Probe system with x-ray:

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

$$|\mathbf{q}| = \frac{4\pi \sin(\theta/2)}{\lambda_0}$$

λ_0 : probe wavelength (2Å)

Record density response:

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

Apply dissipation-fluctuation theorem:

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

$$S(\mathbf{q}, \omega) = -\frac{1}{\pi} \frac{\Im [\chi(\mathbf{q}, -\mathbf{q}, \omega)]}{1 - e^{-\omega/k_B T}}$$

X-ray Thomson scattering in high energy density plasmas

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PRL **116**, 115004 (2016)

PHYSICAL REVIEW LETTERS

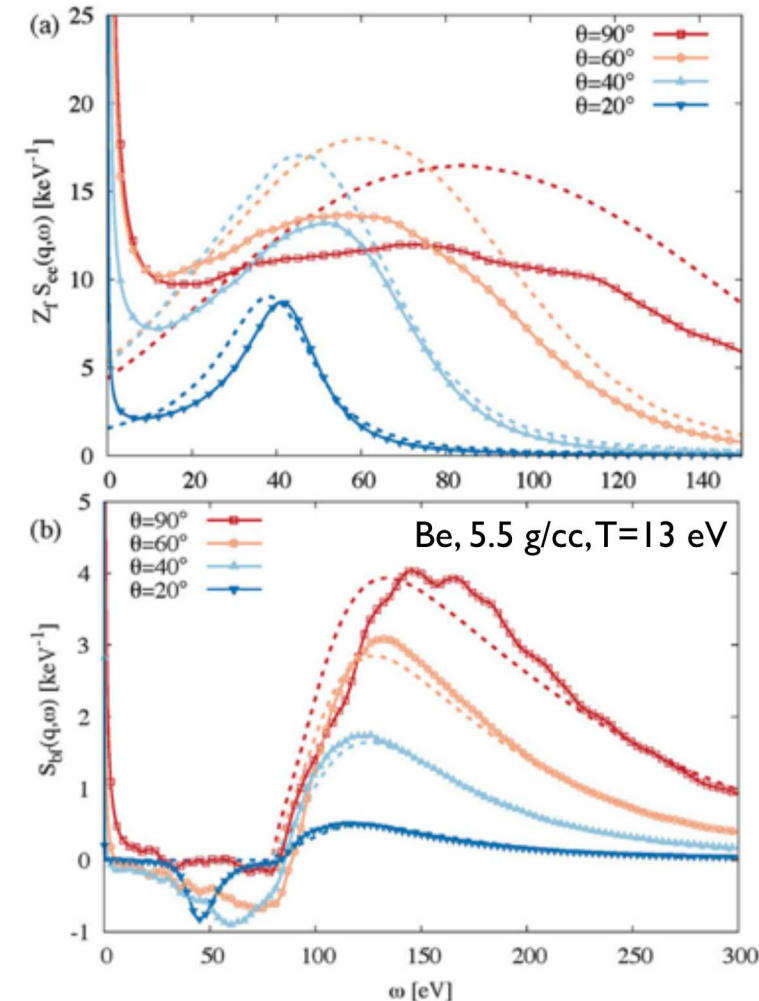
week ending
18 MARCH 2016

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

A. D. Baczewski,^{1,*} L. Shulenburger,² M. P. Desjarlais,² S. B. Hansen,² and R. J. Magyar¹

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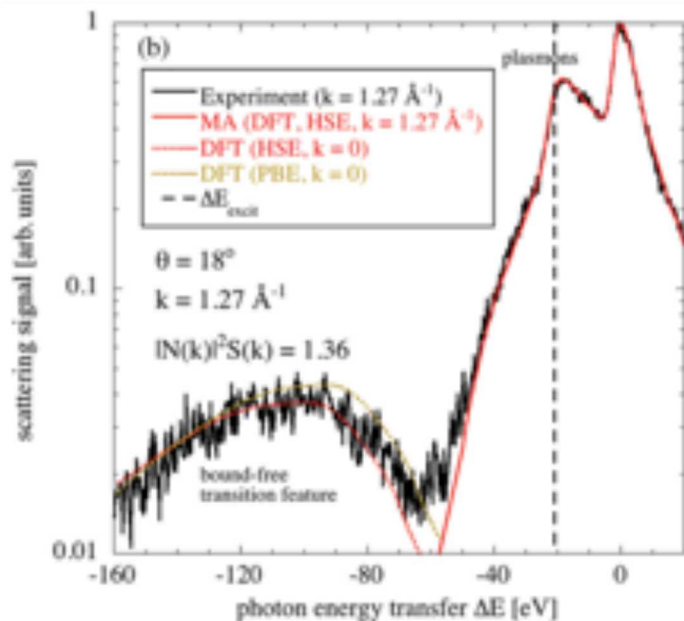
Warm Dense Matter Demonstrating Non-Drude Conductivity from Observations of Nonlinear Plasmon Damping

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

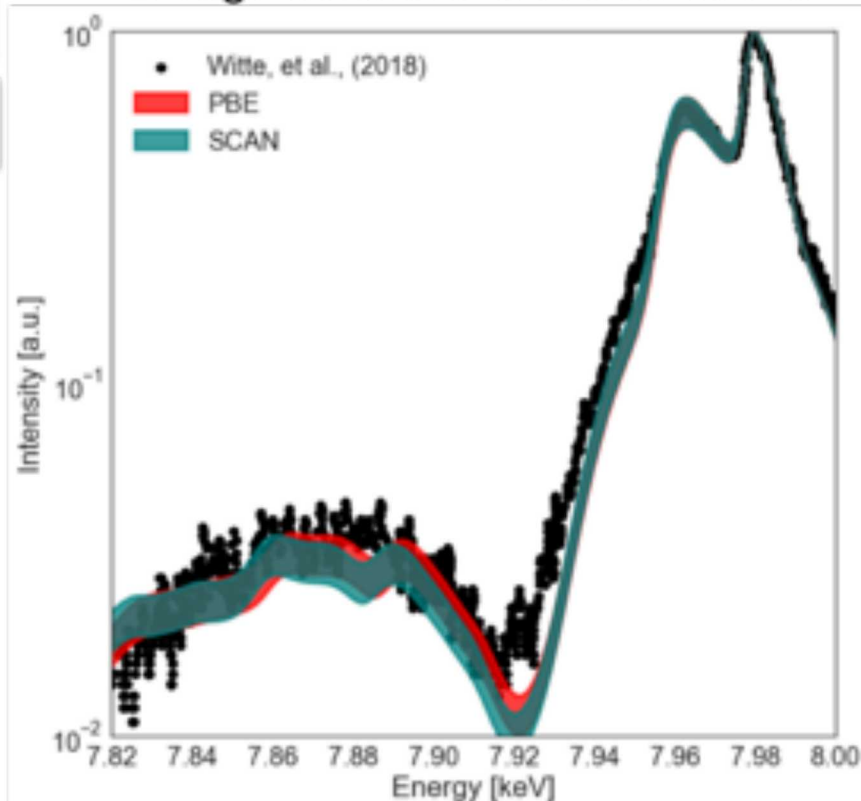
Observations of non-linear plasmon damping in dense plasmas

Physics of Plasmas **25**, 055901 (2018); <https://doi.org/10.1063/1.5017669>

B. B. L. Witte^{1,2,3,4}, P. Sperling^{1,2,3}, M. Frenkel¹, U. Zastrow¹, S. H. Glenzer¹, and R. Redmer¹



L-edge sensitive to XC functional



Angle-resolved
DSF bound-free feature

