

Towards an ab initio real-time treatment of warm dense matter

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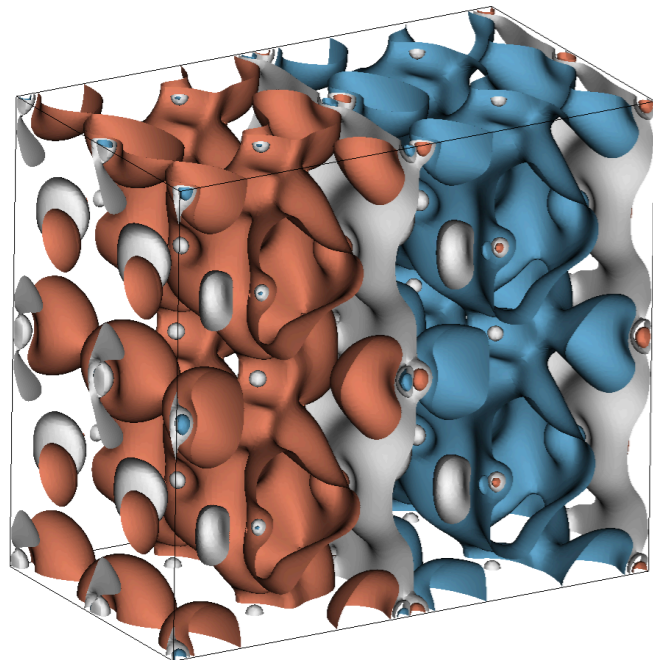
- Collaborators (Sandia National Laboratories)
 - Attila Cangi - **PO5.00008 (~3:30PM Wednesday)**
 - Daniel Jensen - **PO5.00009 (~3:30PM Wednesday)**
 - Stephanie Hansen - **KI2.00001 (3PM today)**
 - Mike Desjarlais, Luke Shulenburger, and Rudy Magyar

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

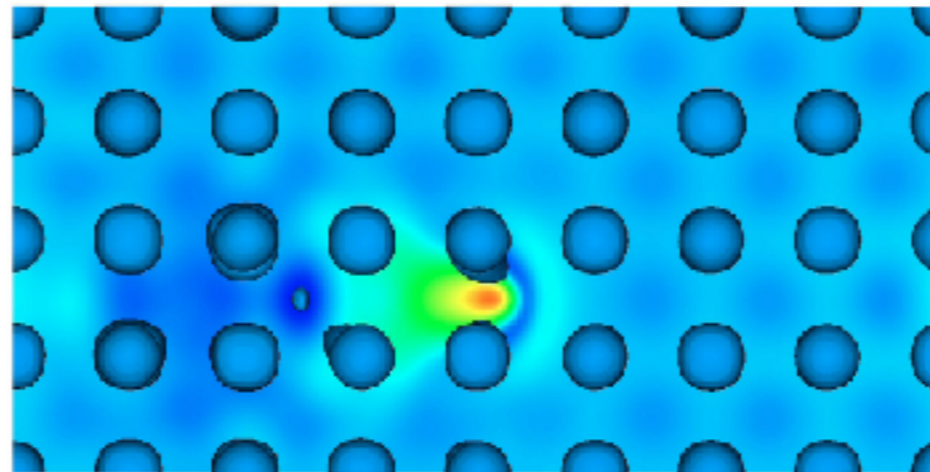
A. D. Baczewski, L. Shulenburger, M. P. Desjarlais, S. B. Hansen, and R. J. Magyar
Phys. Rev. Lett. **116**, 115004 – Published 18 March 2016

- Useful conversations with
 - Ronald Redmer, Philipp Sperling, and Bastian Witte

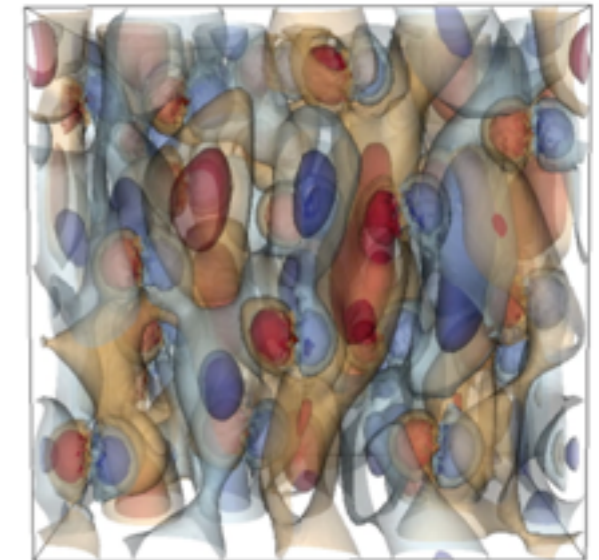
We already have such a theory, TDDFT...



Density response due to hard x-rays in solid Be



Wake of a keV proton in Al



Current response in electride liquid Li

...and it reliably provides us with a wealth of information

But, we *know* it has deficiencies

- Orbital-based - aufbau baked in, but cost prohibitive above 100 eV
- Excited state properties beyond DFT, but far from perfect
- Deficiencies are difficult to remediate
- These problems have proved difficult enough in molecules/solids

- Density built from **TD** orbitals with **fixed** Mermin weights

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

- Orbitals verify the **TD Kohn-Sham equation**

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

- Potential = Coulomb field of atoms + driving potential + **Hxc**

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

- **Fluctuation-dissipation** gives us an **exact** DSF functional

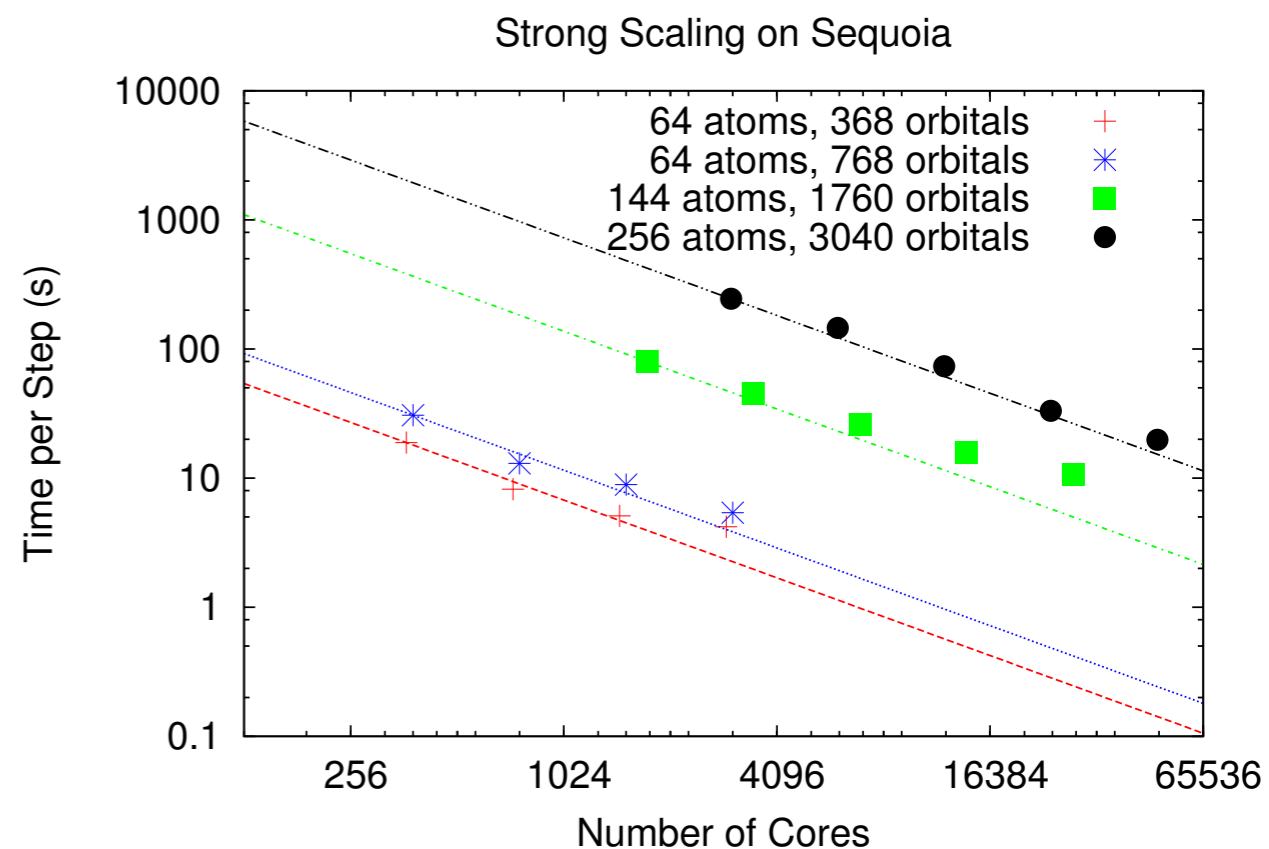
$$\chi_{\rho, \rho}(\mathbf{q}, -\mathbf{q}, \omega) = \frac{\delta \rho(\mathbf{q}, \omega)}{\delta v_{ext}(\mathbf{q}, \omega)} \longrightarrow S(\mathbf{q}, \omega) = -\frac{1}{\pi} \frac{\Im [\chi_{\rho, \rho}(\mathbf{q}, -\mathbf{q}, \omega)]}{1 - e^{-\omega/k_B T_e}}$$

- **TDDFT (+ Ehrenfest dynamics)** in VASP
 - Georg Kresse's group (www.vasp.at)
 - Commercial code (most used in mat. sci.)
 - Many SNL successes with
 - DFT-MD for **thermodynamics**
 - Kubo-Greenwood for **transport**



TDDFT parallelizes very well
over bands - a **huge
advantage for WDM!**

The most expensive
calculations I will show run
in ~1 day on ~1000 CPUs



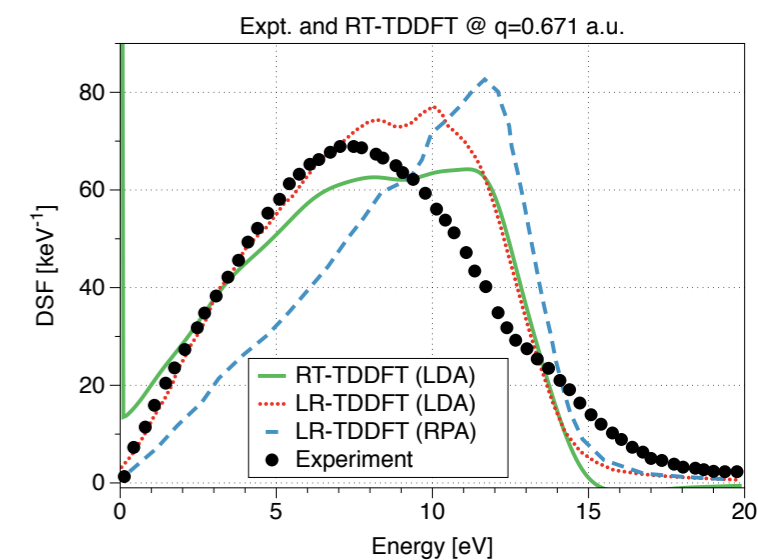
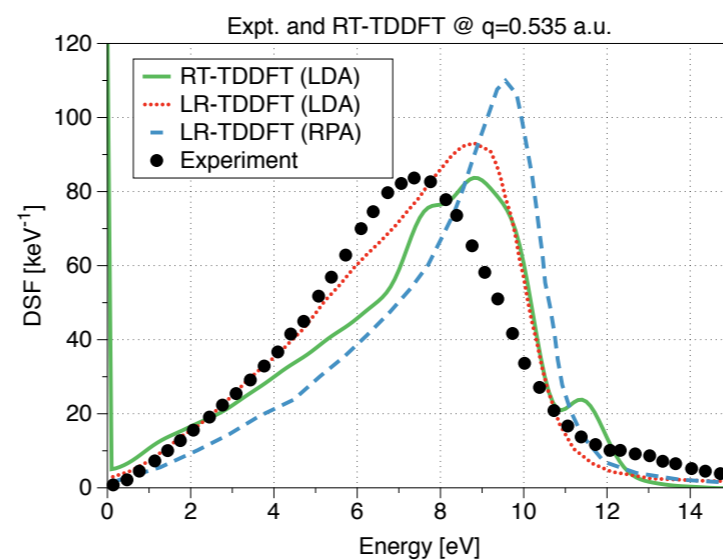
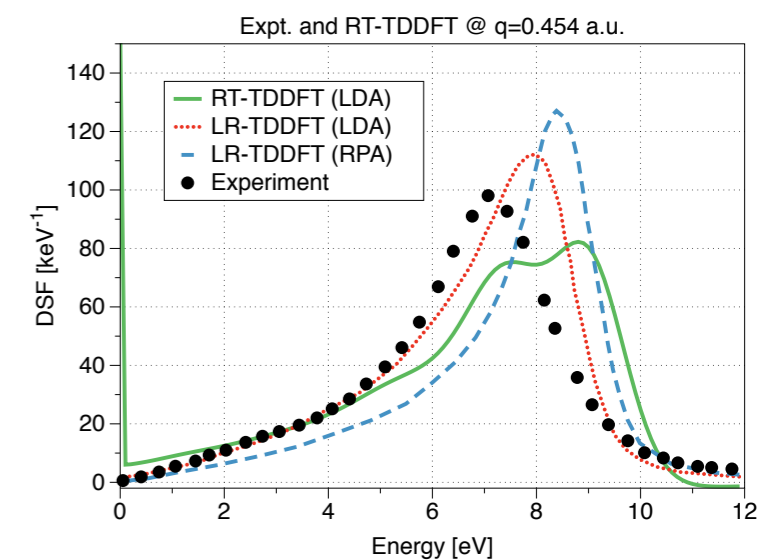
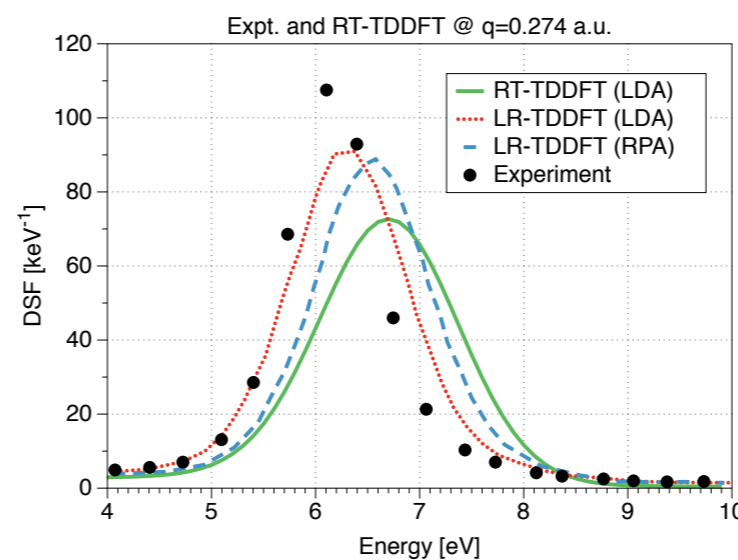
(Test system = shock compressed Xe)

...but what should we expect from *ab initio* methods?

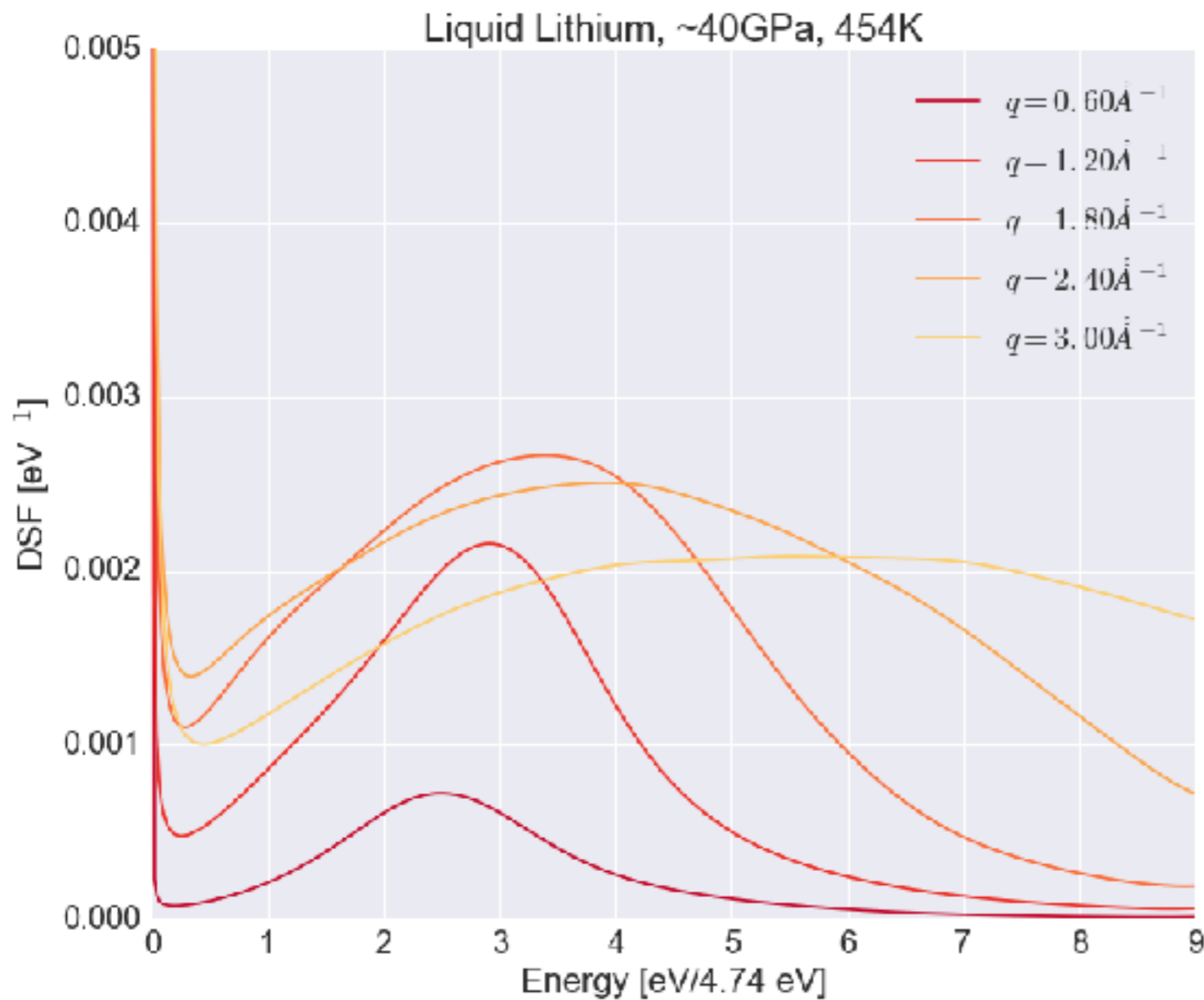
High resolution data from condensed matter **calibrates expectations**

Compare to energy domain TDDFT and experiment from
Cazzaniga, et al., PRB (2011)

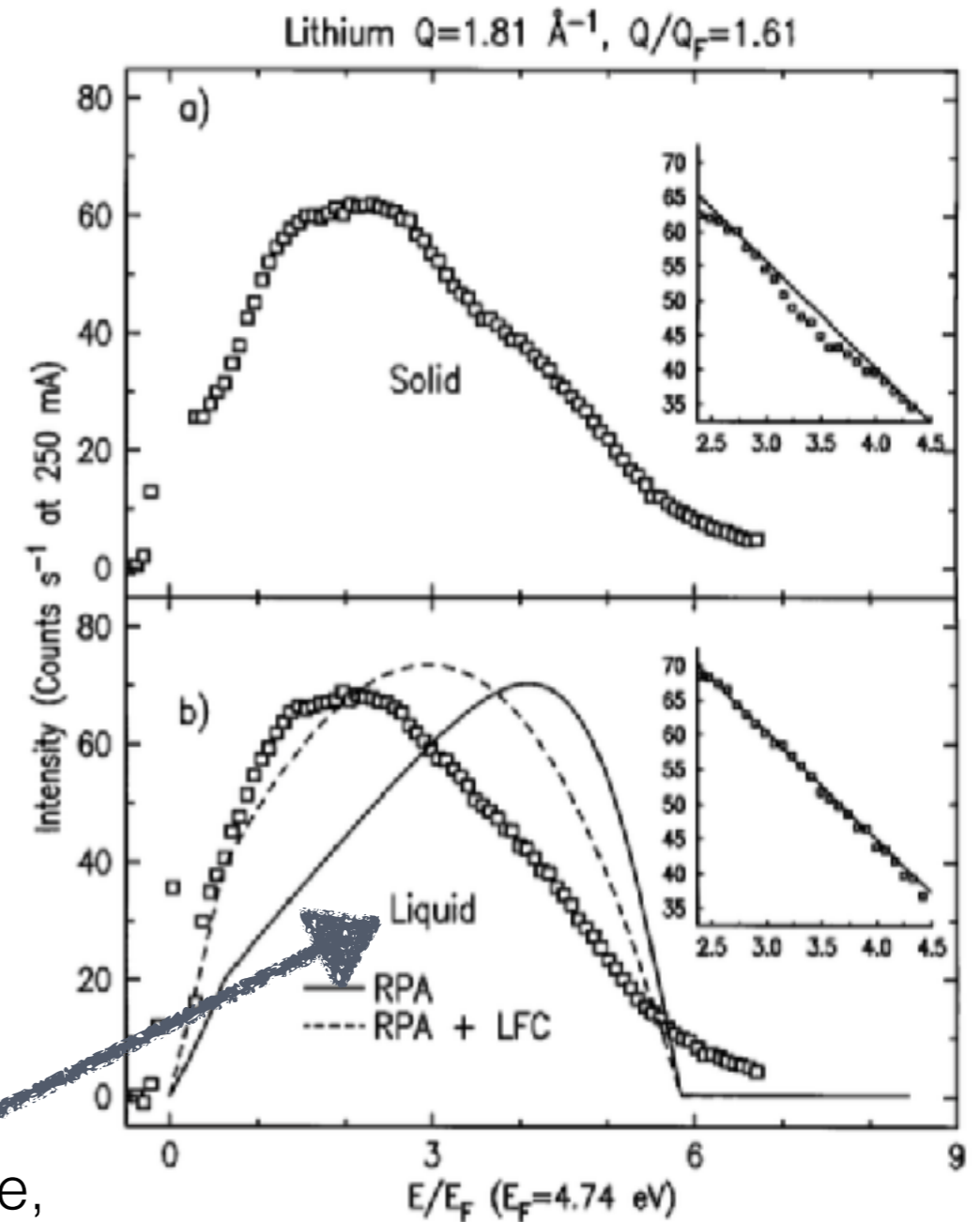
RPA = time-dependent Hartree approximation, but **not** applied to jellium



454 K + ~40 GPa, just above ambient melt line



Same temperature,
ambient density

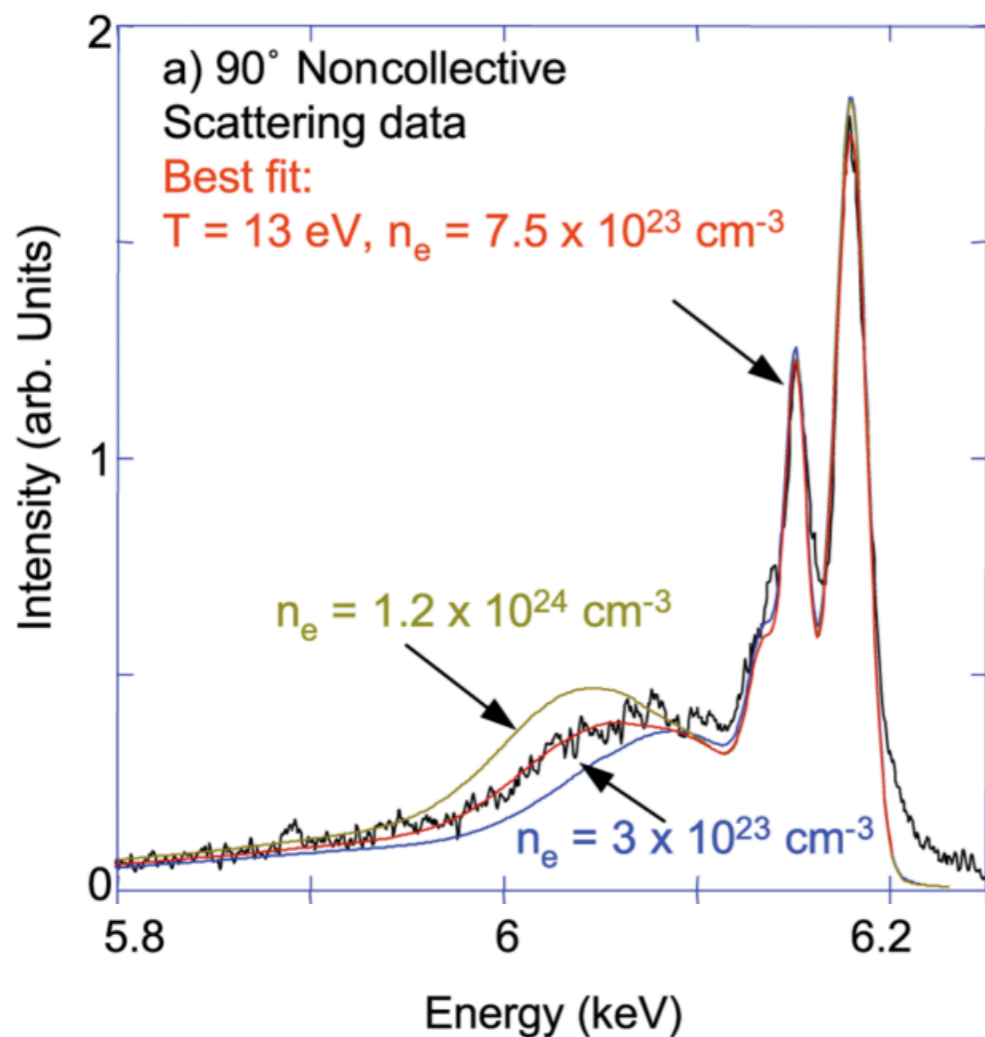


Hill, et al., PRL 1996

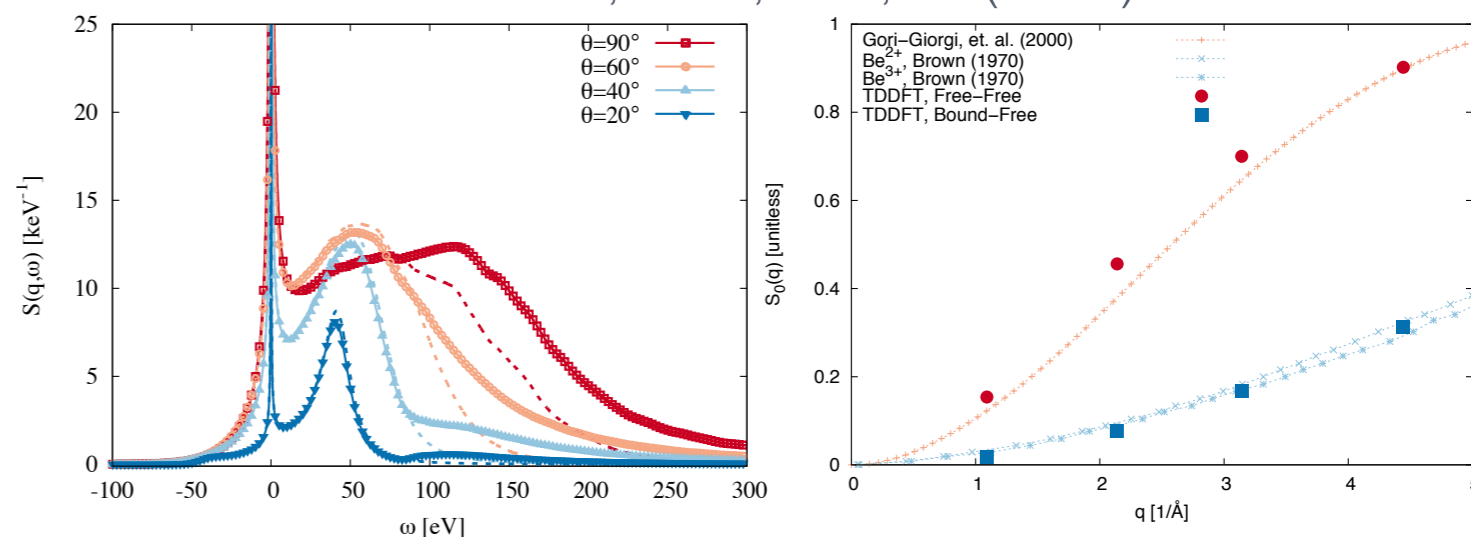
XRTS of shock-compressed beryllium

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 15 March 2009

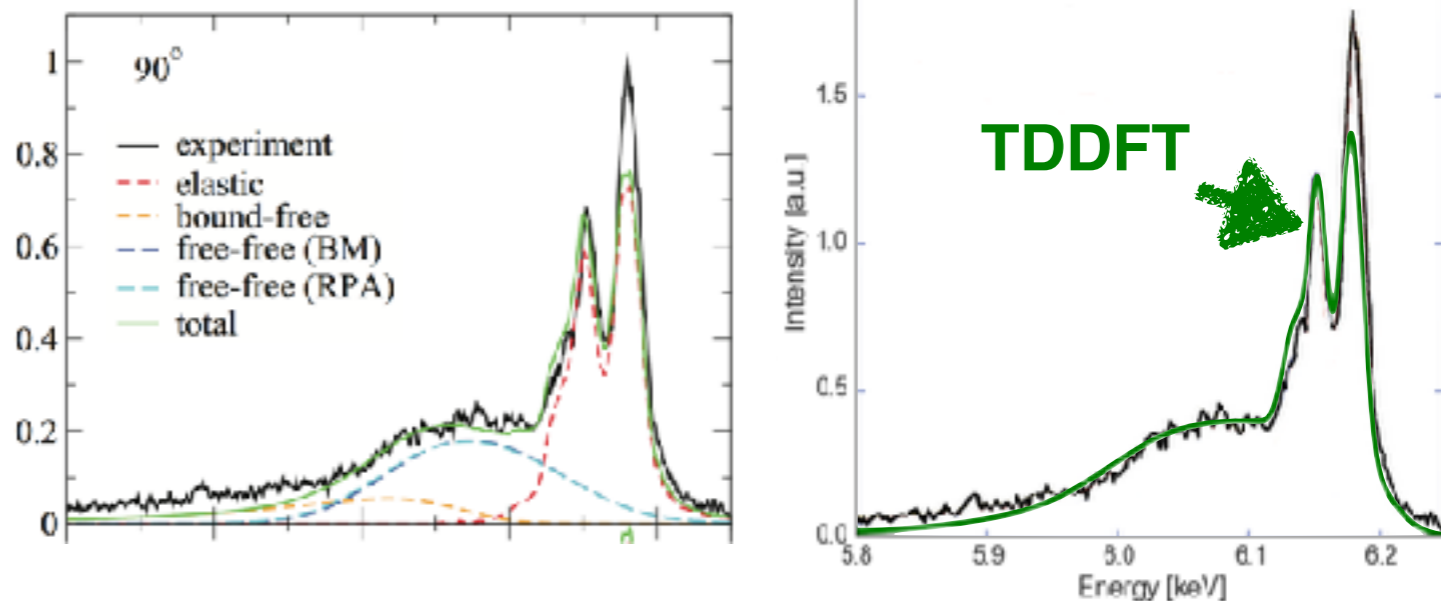


Baczewski, et al., PRL, **89** (2016)



TDDFT picks up electron correlation effects that DFT fails to pick up

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



TDDFT agrees w/experiment as well as more empirical theories

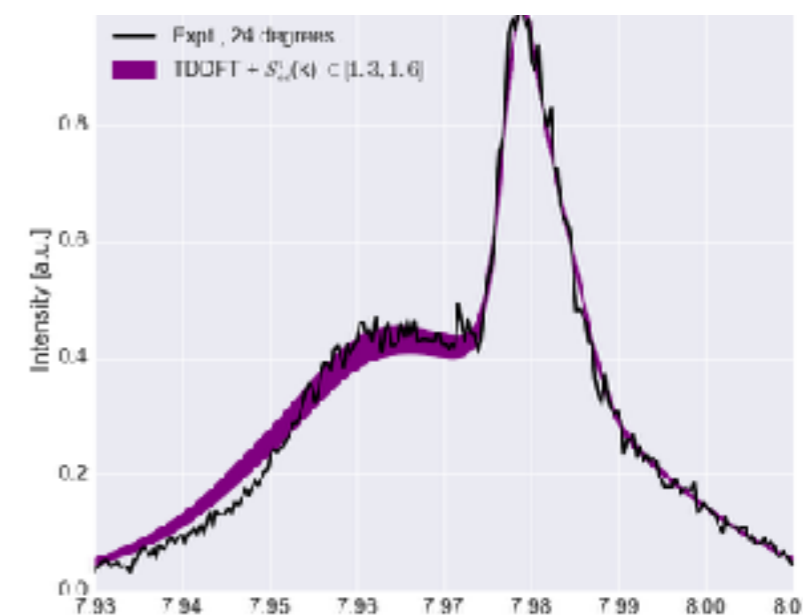
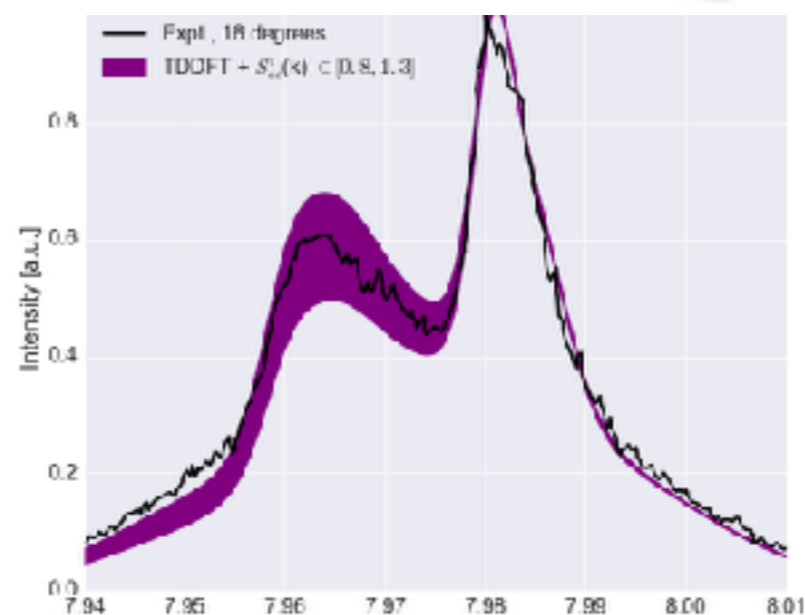
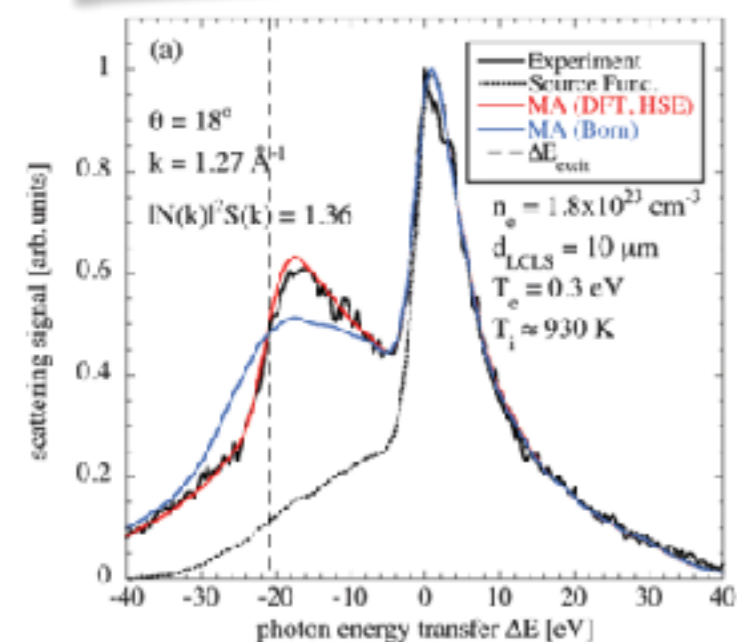
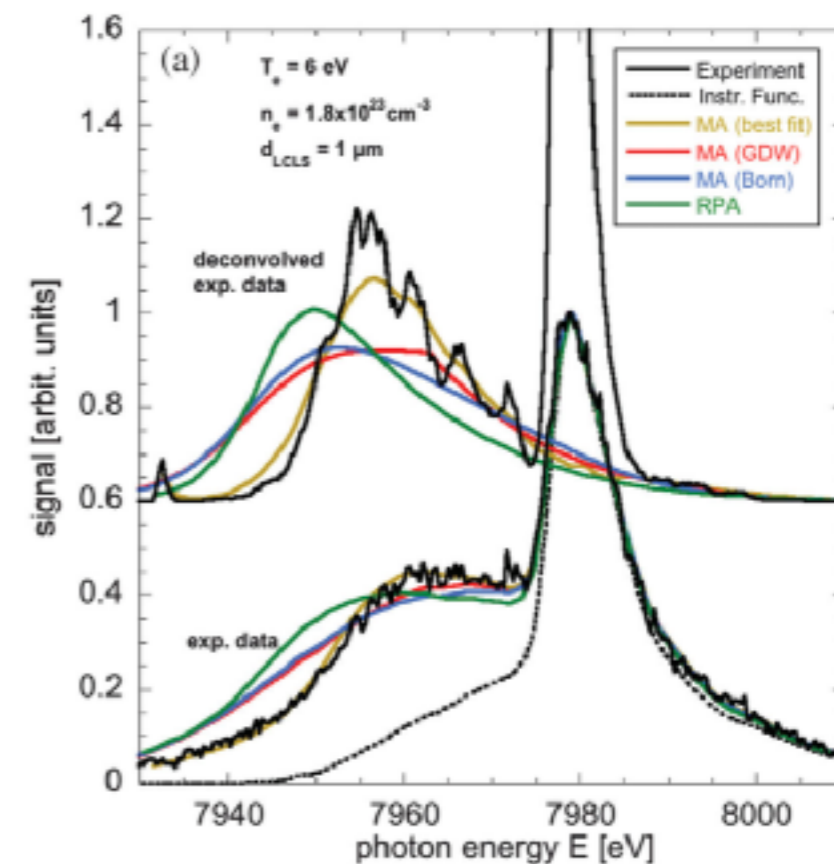
Free-Electron X-Ray Laser Measurements of Collisional-Damped Plasmons in Isochorically Heated Warm Dense Matter

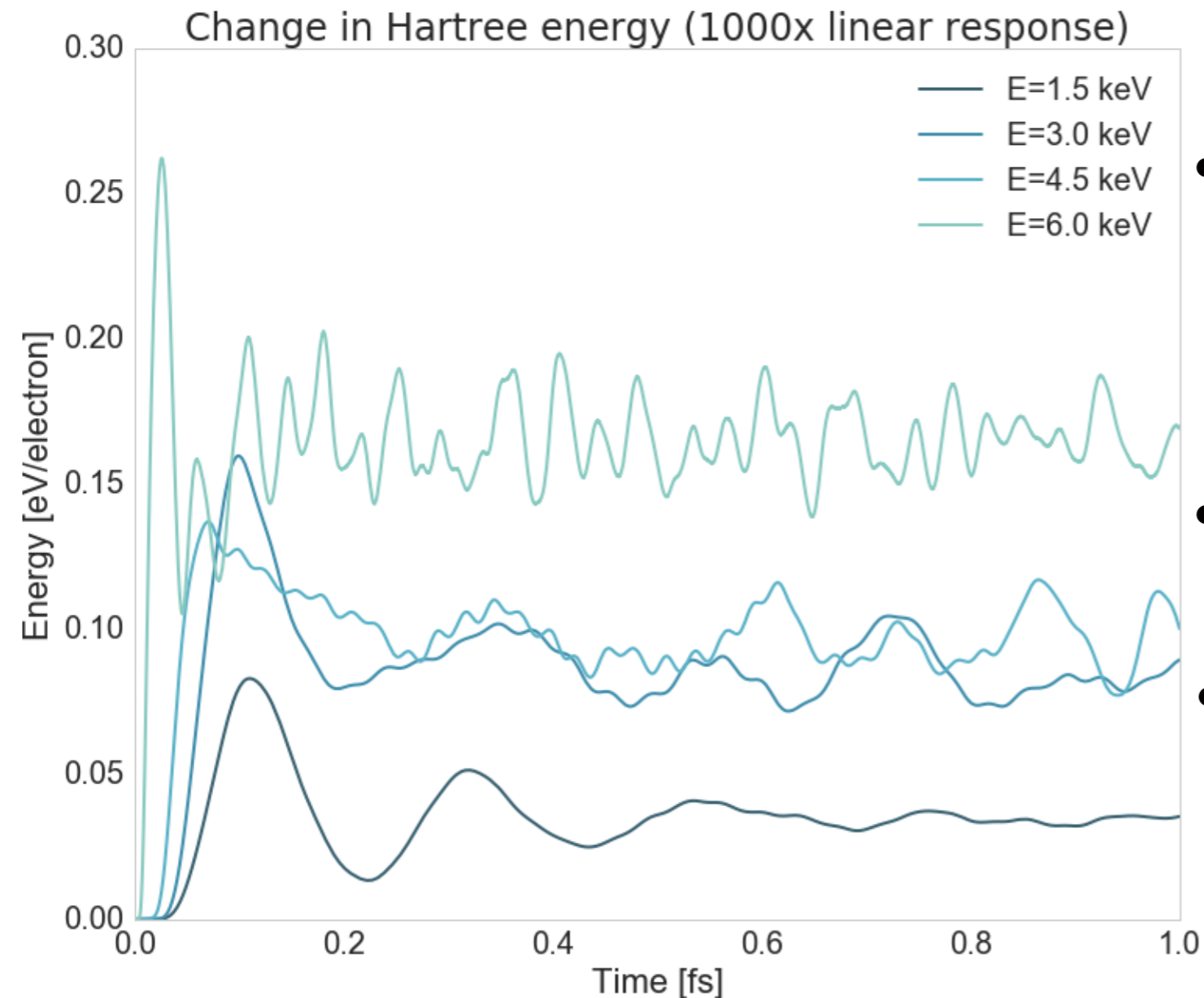
P. Sperling, E. J. Gamboa, H. J. Lee, H. K. Chung, E. Galtier, Y. Omarbaklyeva, H. Reinholz, G. Röpke, U. Zastrau, J. Hastings, L. B. Fletcher, and S. H. Glenzer
 Phys. Rev. Lett. **115**, 115001 – Published 9 September 2015

- LCLS beam used to isochorically heat and probe **Al to 6 eV**
- Collision models **beyond Born approximation** needed for fit

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





- Drive amplitude is 1000x larger than amplitude that gives **linear response DSF**
- **Hartree energy** is observable
- Increases for < 1 fs and **saturates** for all energies

1.) Kohn-Sham occupancies **don't know how to change!**

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

2.) Even if they could, **we don't have high (~keV) energy states** into which we can directly drive core electrons...

3.) Nor do we have the possibility of capturing **Auger processes...**

4.) Not only do we need to treat these excitations, but we need a reliable method for capturing their **equilibration...**

TDDFT is a great start for ab initio simulations of warm dense matter, in- and out-of-equilibrium, but it isn't perfect...

1. TDDFT captures physics that DFT does not
2. TDDFT's accuracy is decent, but difficult to improve upon
3. There are clearly defined deficiencies that need to be resolved, and we are working on going *beyond* TDDFT at SNL

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Consider the optical absorption spectrum of Na_2

1. Start from the DFT ground state
2. Apply an electric field along dimer axis
3. Record the dipole moment in time

