SAND2019-2909C

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

Sandia National Laboratories

PRESENTED BY

Attila Cangi

CUNY Graduate Center

"Computational Modeling for High Energy Density Science and Complex Systems"

Friday, March 15, 2019

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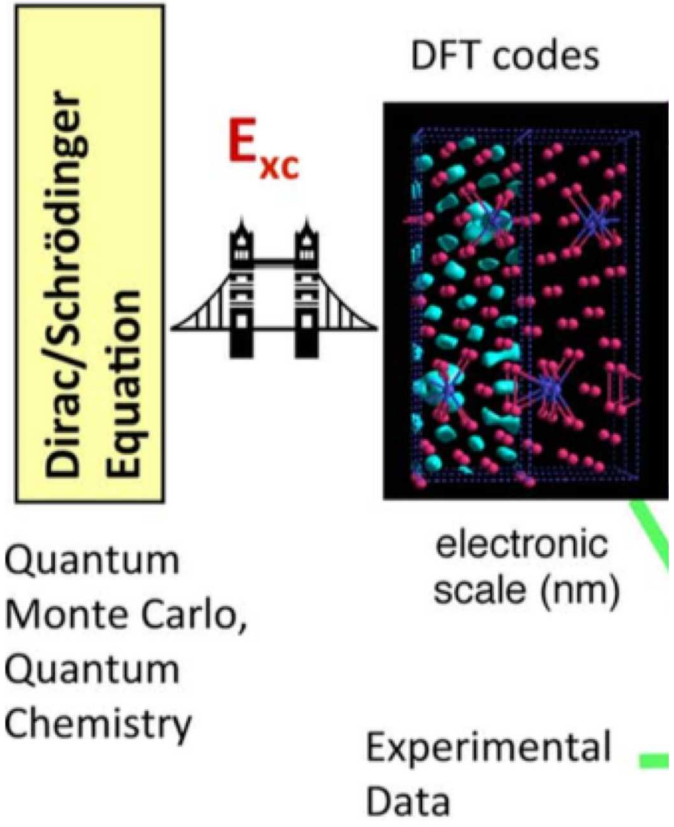
Multiscale Science



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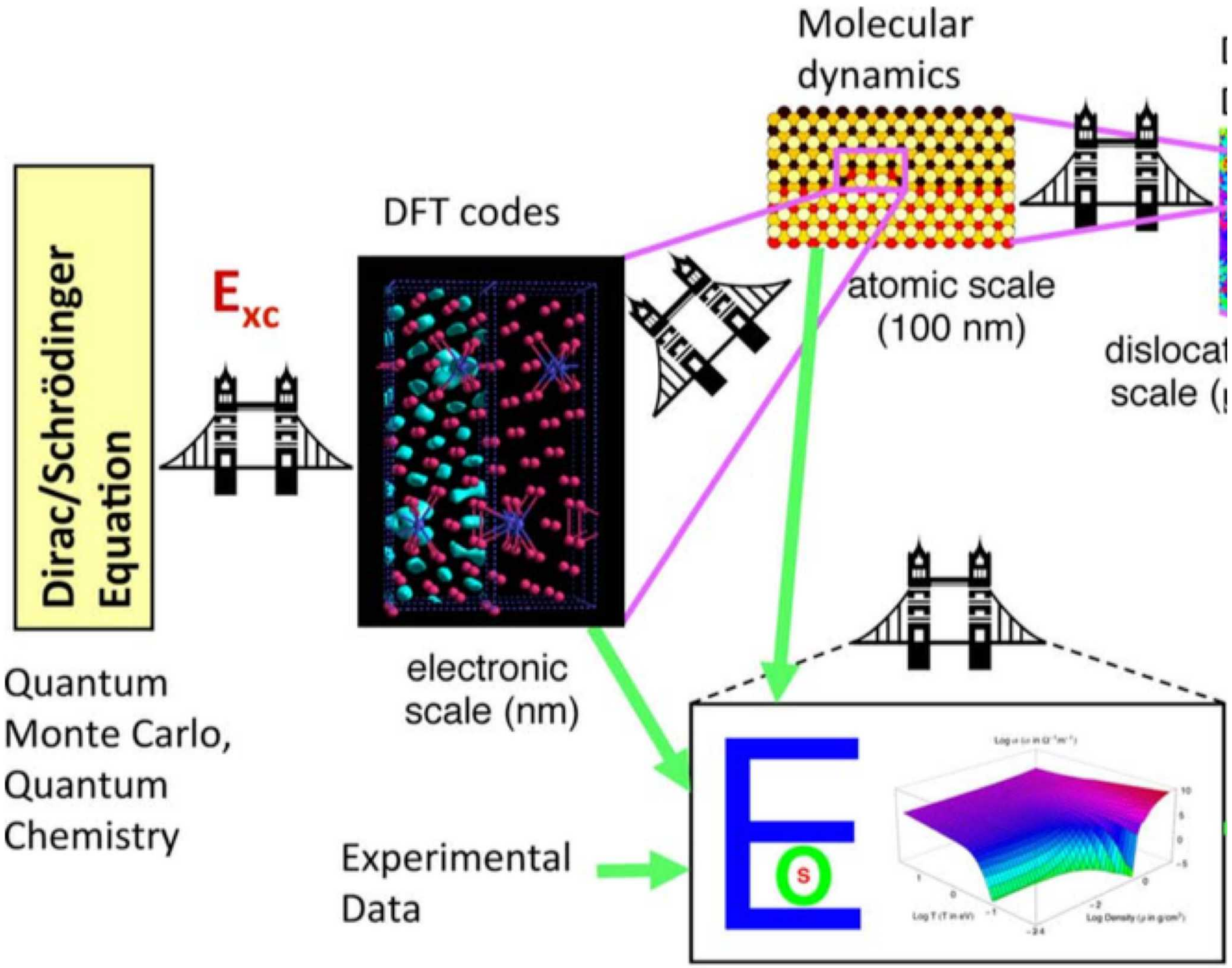
Molecular dynamics
DFT codes
Dirac/Schrödinger Equation
E_{xc}
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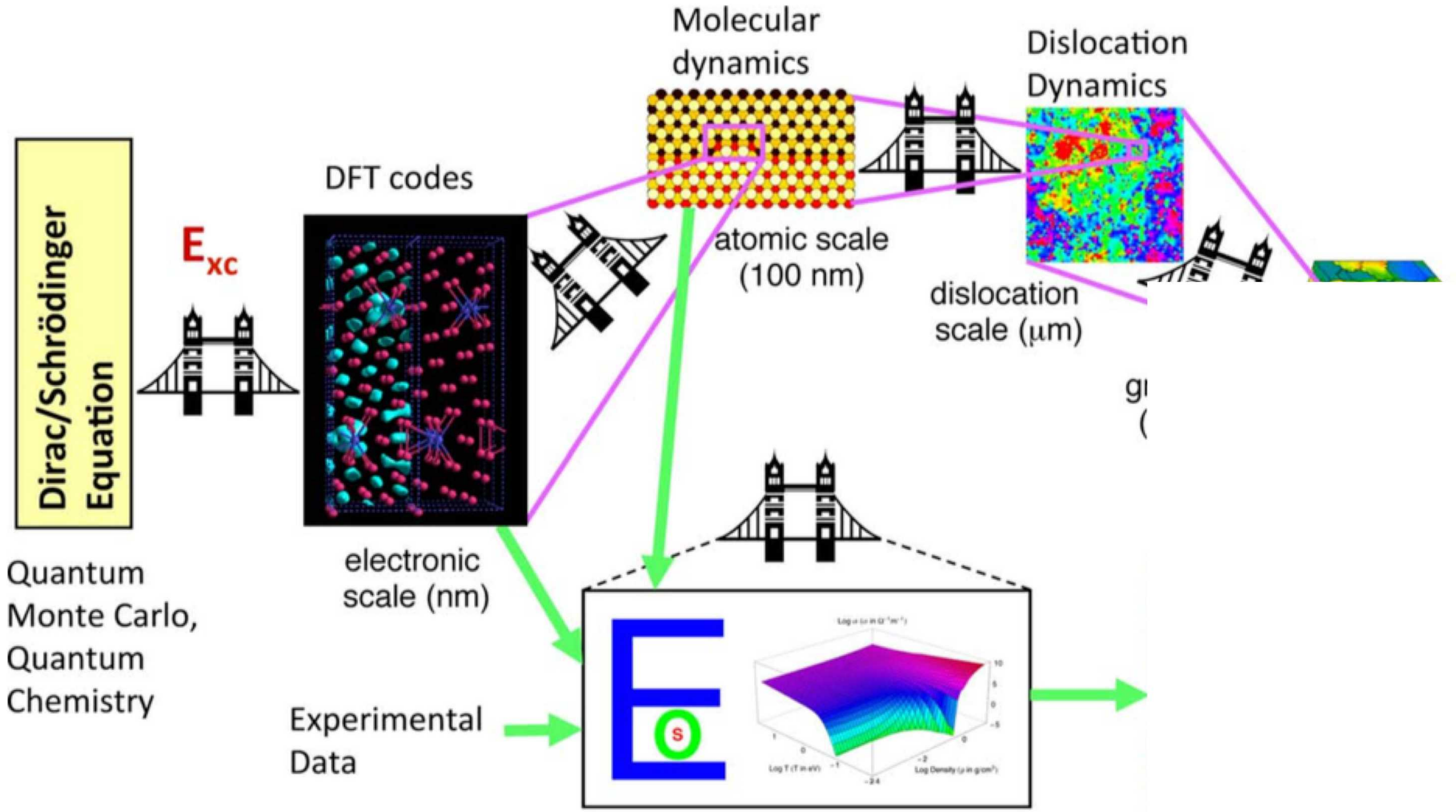
Quantum Monte Carlo, Quantum Chemistry
Experimental Data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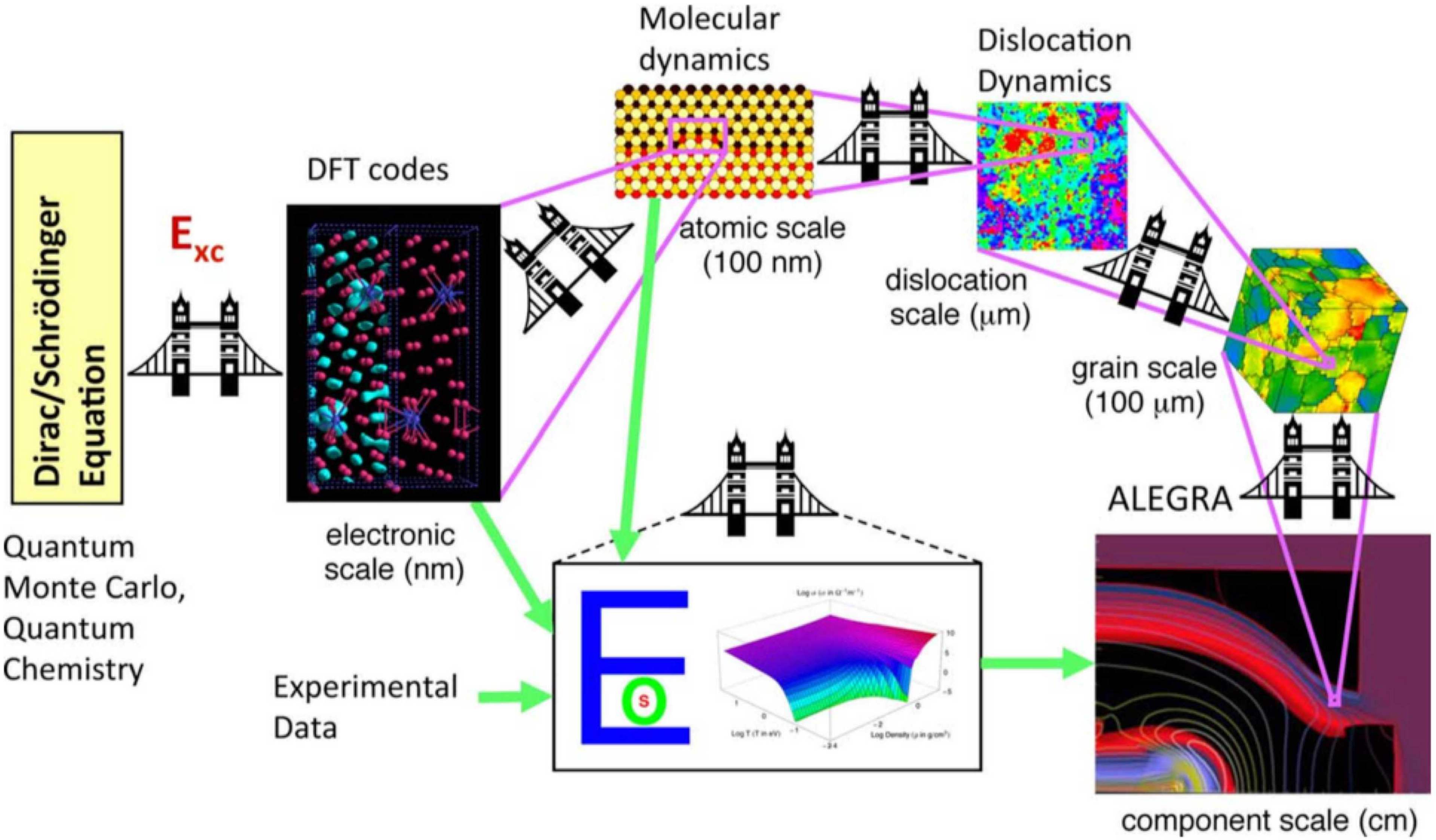

Ann E. Mattson *et al.*, International Journal of Quantum Chemistry (2016), DOI: 10.1002/qua.25097..

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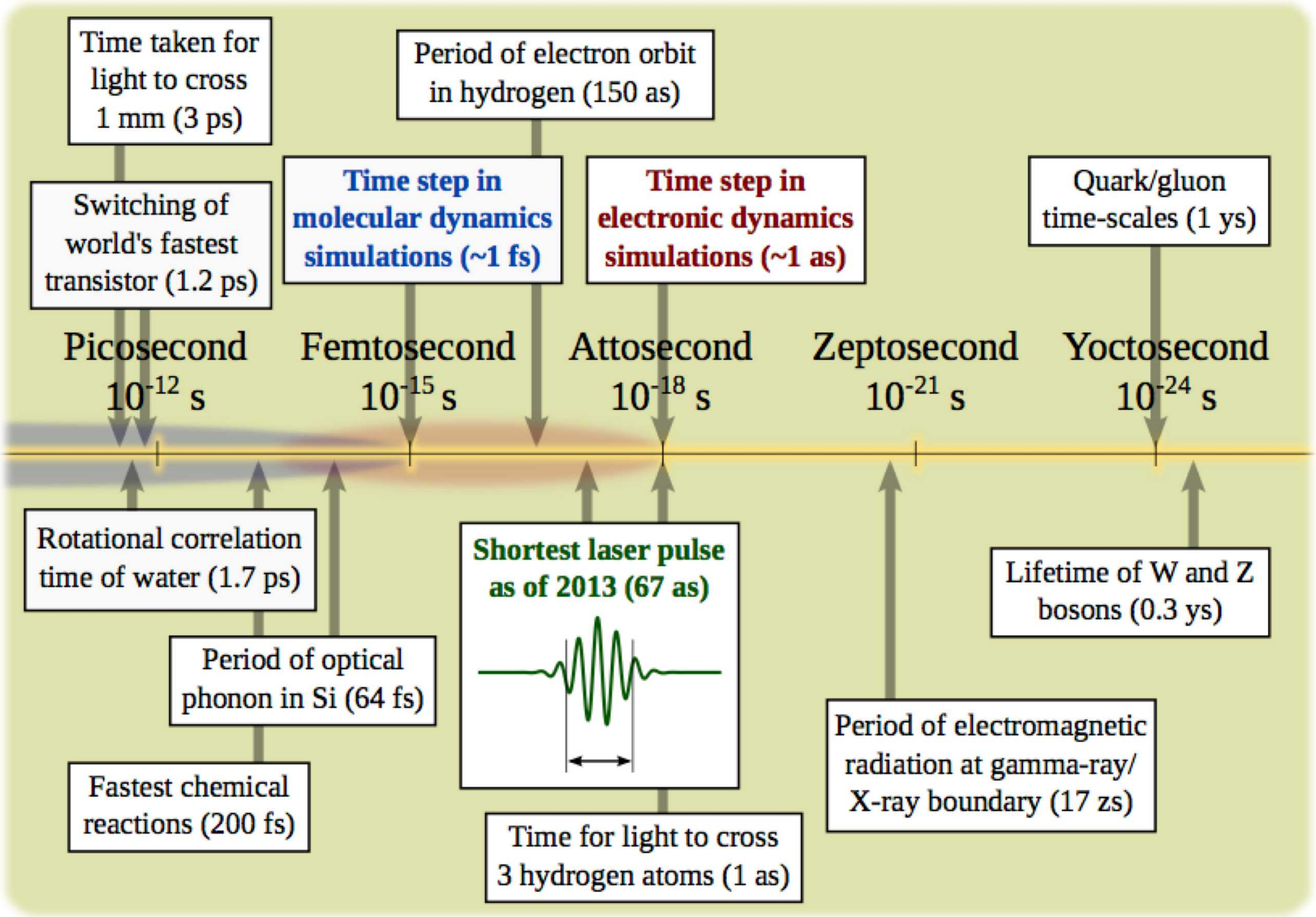

Courtesy of K. Dewhurst, Max Planck Institute of Microstructure Physics (2015).

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Coulomb coupling parameter

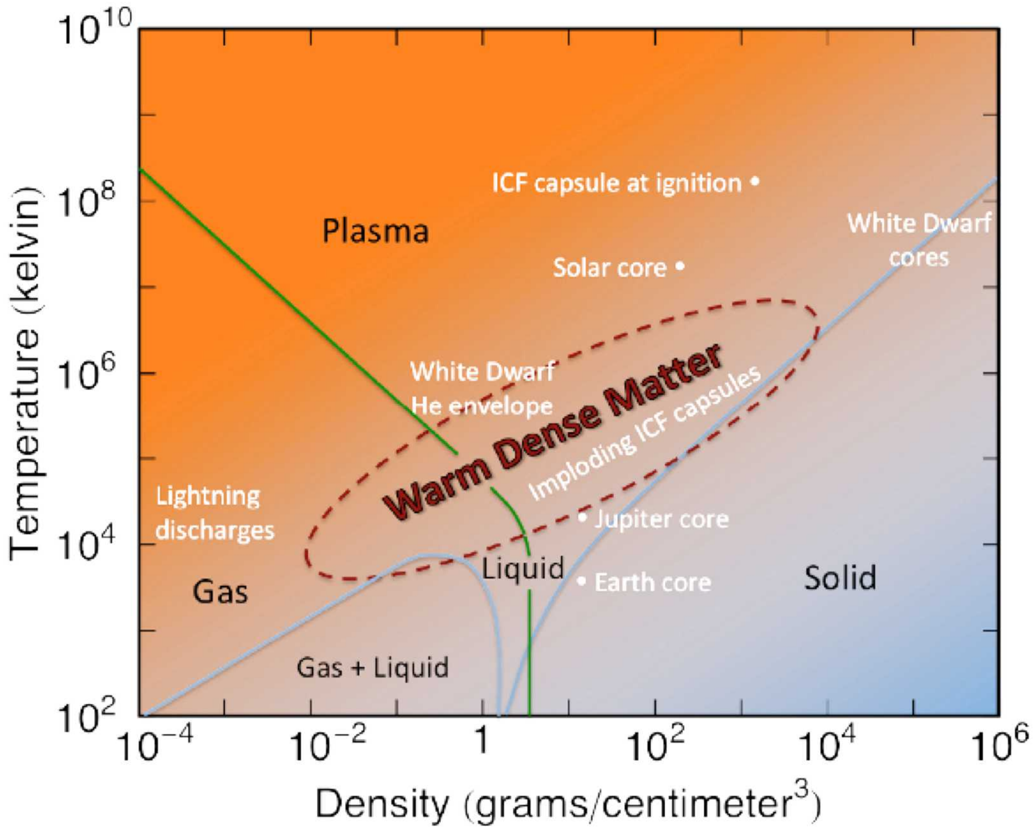
$$\Gamma = \frac{V}{T} = \frac{Ze^2}{r_\mathrm{S} k_\mathrm{B} \tau}$$

Electron degeneracy parameter

$$\Theta = \frac{k_\mathrm{B}\tau}{E_\mathrm{F}}$$

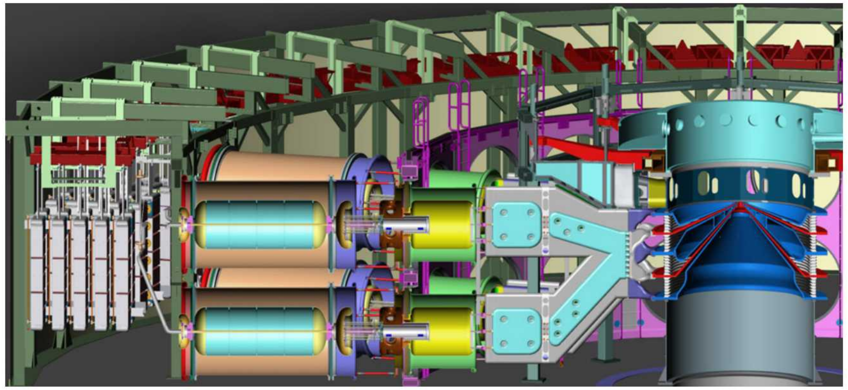
Warm dense matter regime

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



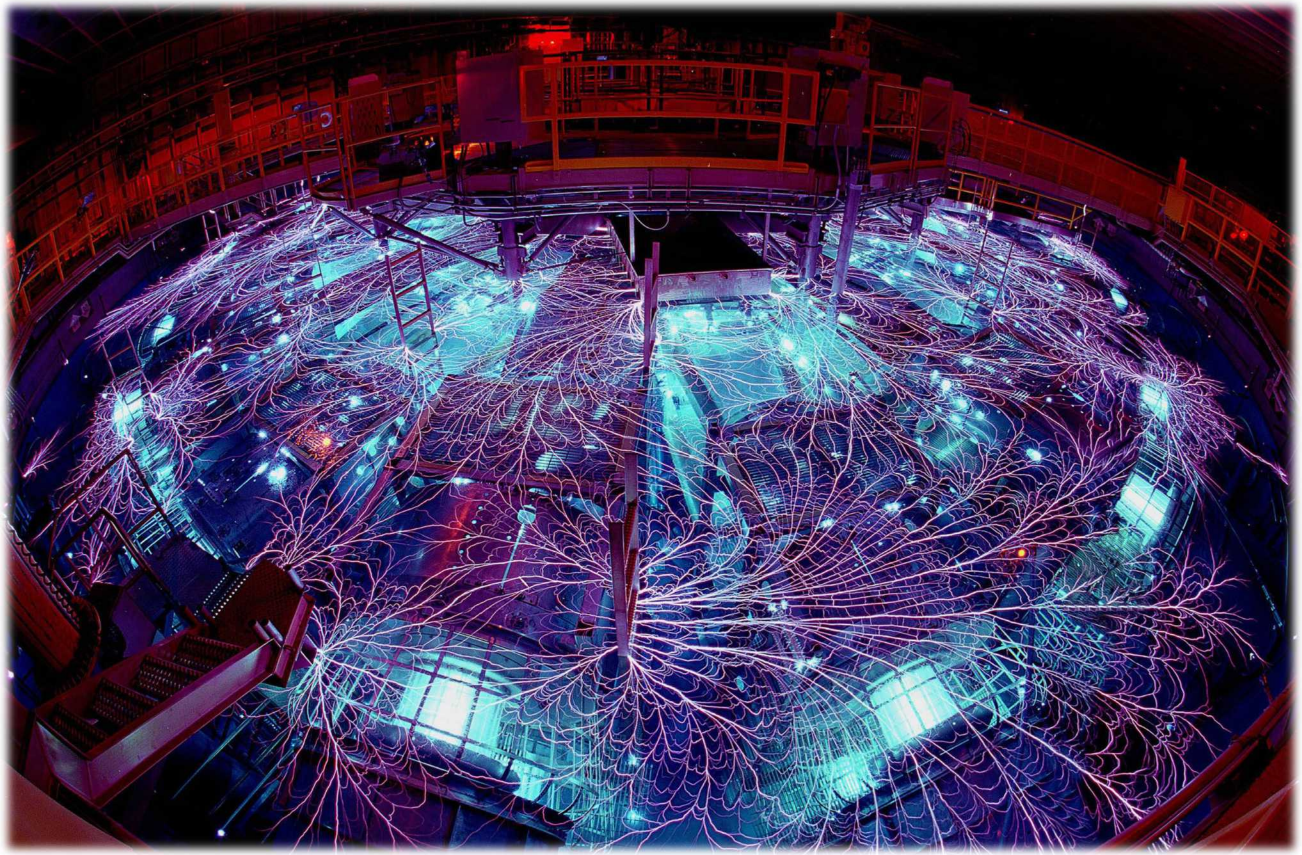
Basic Research Needs for HEDLP, Report of the Workshop on HEDLP Research Needs, DOE (2008).

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)

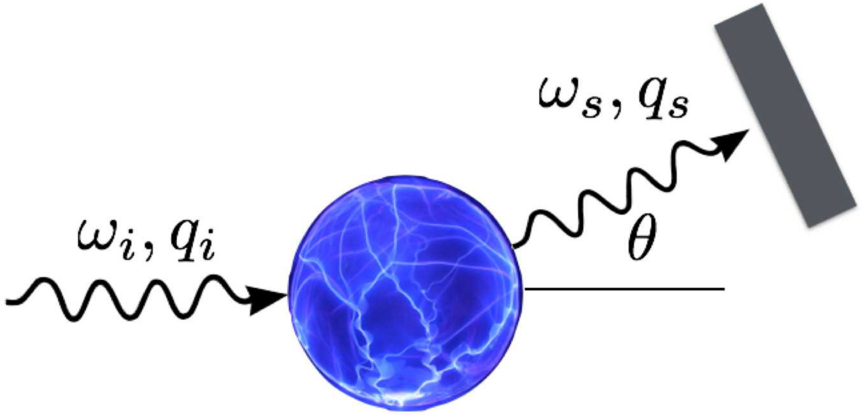
Compression of energy in time and space in pulsed power facilities (Z machine) enables exciting science (astrophysics, planetary science, inertial confinement fusion).



Z Machine, Sandia National Laboratories.

X-Ray Thomson Scattering

Hard x-rays



Measure inelastically scattered x-rays

$$\omega = \omega_i - \omega_s$$
$$q = q_i - q_s$$
$$q_s = 2q_i \sin(\theta/2)$$

Sample of WDM
(opaque to optical probes)

- X-ray Thompson 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

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

Interplay between Theory, Simulations, and Experiment

Electronic structure theory

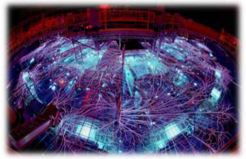
Electronic structure theory provides input to magneto-hydrodynamics simulations.

Computational fluid dynamics

Computational fluid dynamics simulations support design of HED experiments.

HED experiments

HED experiments and diagnostics benchmark theory.



LCLS

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

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

Dynamic structure factor

LETTER

doi:10.1038/nature14048

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

J. E. Bailey[1], T. Nagayama[1], G. P. Loisel[1], G. A. Rochau[1], C. Blancard[2], J. Colgan[3], Ph. Cosse[2], G. Faussurier[2], C. J. Fontes[3], F. Gilleron[2], I. Golovkin[4], S. B. Hansen[1], C. A. Iglesias[5], D. P. Kilcrease[3], J. J. MacFarlane[4], R. C. Mancini[6], S. N. Nahar[7], C. Orban[7], J.-C. Pain[2], A. K. Pradhan[7], M. Sherrill[3] & B. G. Wilson[5]

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

Electrical 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. Zastrau, J. Hastings, L. B. Fletcher, and S. H. Glenzer

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

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

Coupled Electron-Ion Many-Body Problem

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

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

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

"The fundamental laws necessary for (…) the whole of chemistry [and materials science] are thus completely known, and the difficulty lies only in the (…) equations that are too complex to be solved."

Paul Dirac

Kinetic, interaction, and external potential terms.

$$\hat{T}^n=\sum_{\alpha}^{N^n}-\frac{\hbar^2}{2M_\alpha}\nabla_\alpha^2 \qquad \hat{T}^e=\sum_{i}^{N^e}-\frac{\hbar^2}{2m}\nabla_i^2$$

$$\hat{W}^{nn}=\frac{1}{2}\sum_{\alpha,\beta,\alpha\neq\beta}^{N^n}\frac{e^2\,Z_\alpha\,Z_\beta}{4\pi\epsilon_0\,|\mathbf{R}_\alpha-\mathbf{R}_\beta|} \qquad \boxed{\hat{W}^{ee}=\frac{1}{2}\sum_{i,j,i\neq j}^{N^e}\frac{e^2}{4\pi\epsilon_0\,|\mathbf{r}_i-\mathbf{r}_j|}} \qquad \hat{W}^{en}=-\sum_{i}^{N^e}\sum_{\alpha}^{N^n}\frac{e^2\,Z_\alpha}{4\pi\epsilon_0\,|\mathbf{r}_i-\mathbf{R}_\alpha|}$$

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

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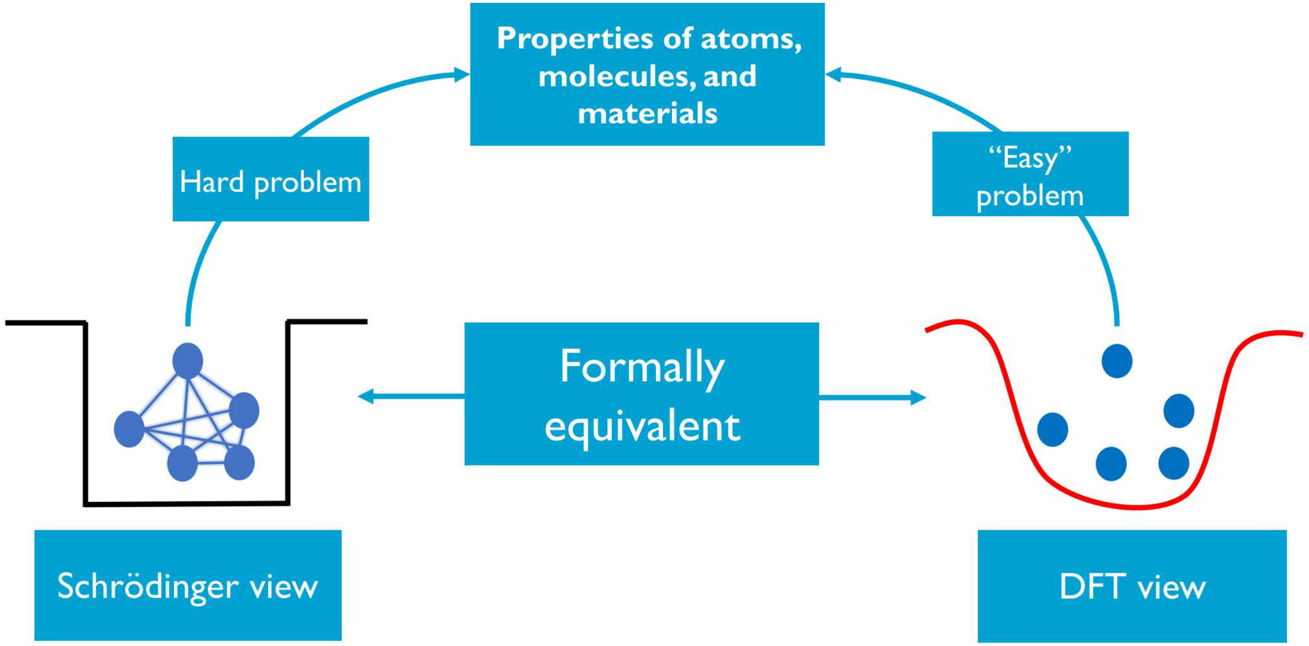
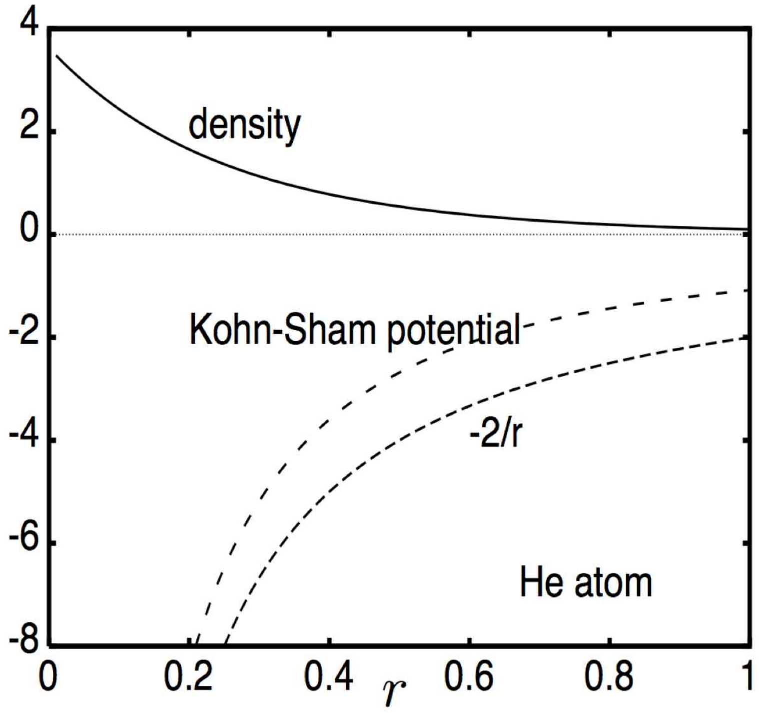
(Time-Dependent) Density Functional Theory in a Nutshell



$$\left[-\frac{\nabla^2}{2}+\boxed{v_{\rm S}(\mathbf{r})}\right]\phi_j(\mathbf{r})=\epsilon_j\phi_j(\mathbf{r}) \qquad \text{(Kohn-Sham equations)}$$

$$v_{\rm S}(\mathbf{r})=v(\mathbf{r})+\frac{\delta U}{\delta n(\mathbf{r})}+\frac{\delta E_{\rm XC}}{\delta n(\mathbf{r})} \qquad \text{(Kohn-Sham potential)}$$

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



The ABC of DFT (dft.uci.edu/doc/g1.pdf).

Real-time and Linear Response TDDFT

1. Prepare initial state from a static DFT calculation

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

2. Solve the TD Kohn-Sham equations

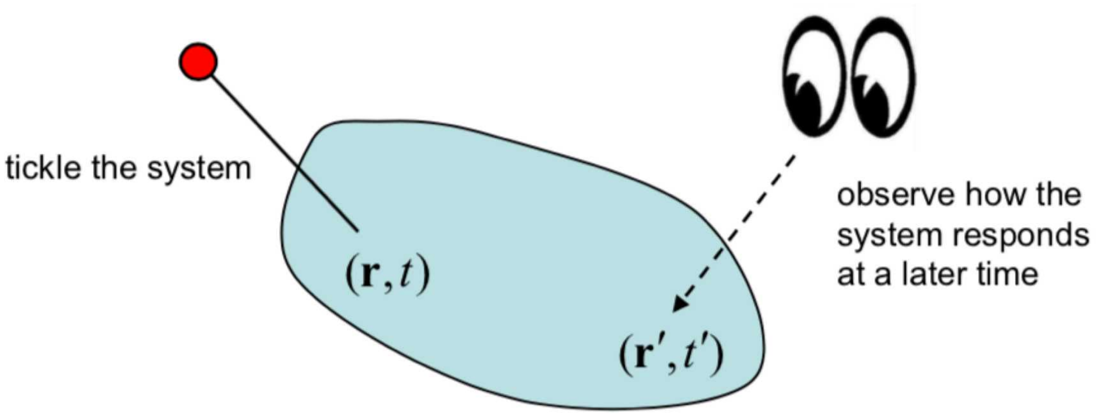
$$i\frac{d\phi_j(\mathbf{r})}{dt}=\left[-\frac{\nabla^2}{2}+v_{\mathrm{S}}(\mathbf{r})\right]\phi_j(\mathbf{r})$$

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

3. Compute observables as a functional of time-dependent density

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

Alternatively, use linear response theory



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

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

Sandia's Implementation of TDDFT

Sandia National Laboratories

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

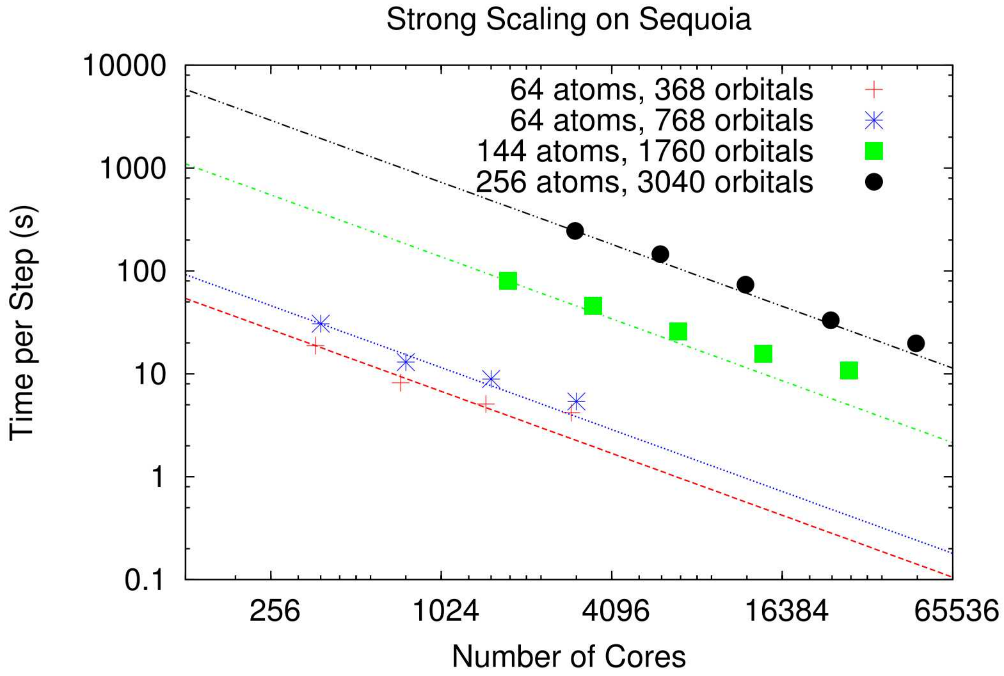
Coupled electron-ion equations of motion

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

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

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

QUANTUM ESPRESSO

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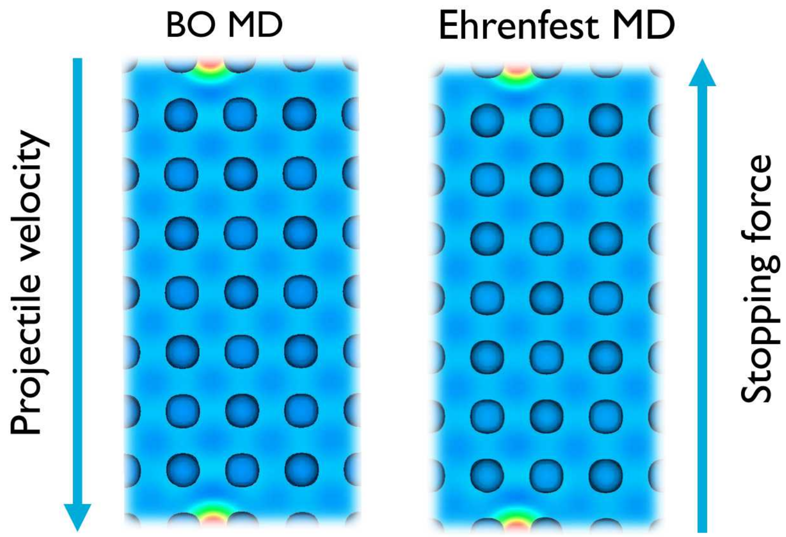
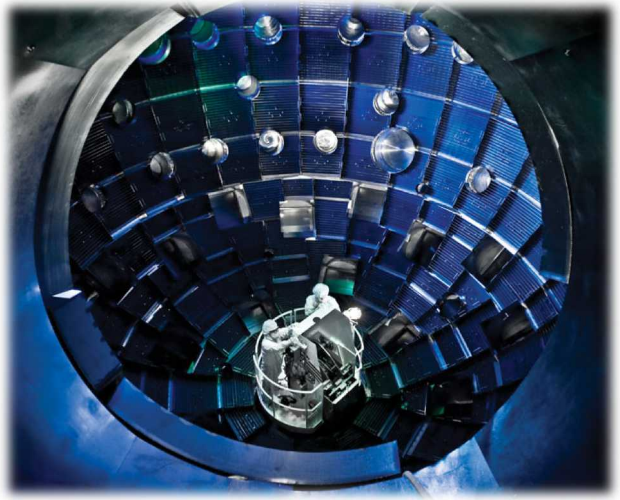
Stopping Power in Warm Dense Targets

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

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





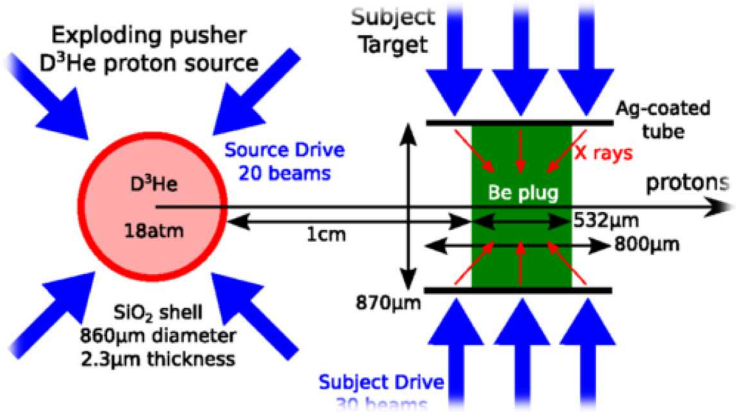
Target chamber, National Ignition Facility, Lawrence Livermore National Laboratories.

Stopping mechanisms

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

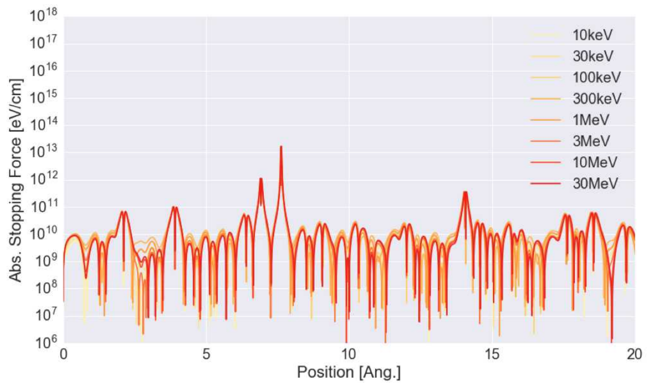
Large body of literature for cold targets

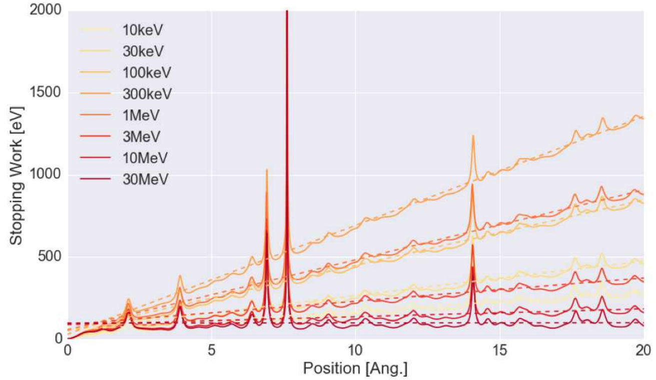
- Empirical approximations (Rutherford, Thomson, Bohr, Bethe)
- Parameter-free atomistic simulations
- Electronic structure coupled to molecular dynamics
- Cold stopping power (Echenique, Correa, Artacho, Schleife)

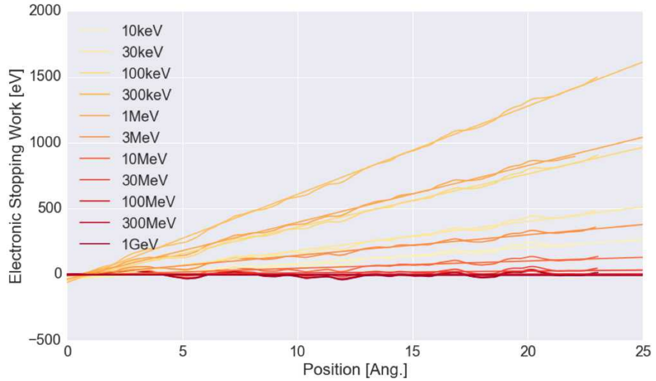


Zylstra et al., Phys. Rev. Lett. 114, 215002 (2015).

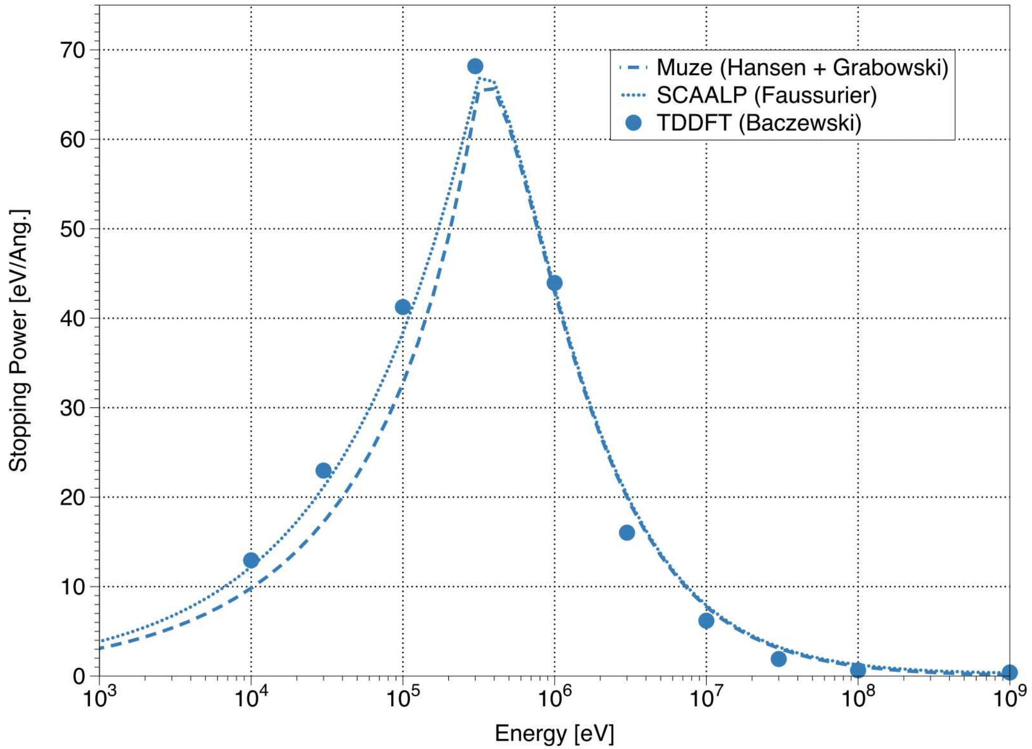
Stopping Power in Warm Dense Targets



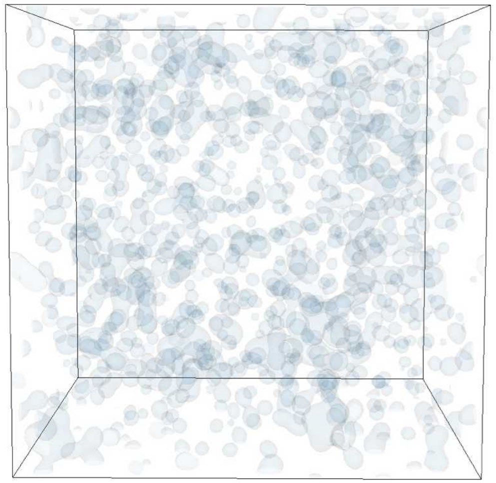




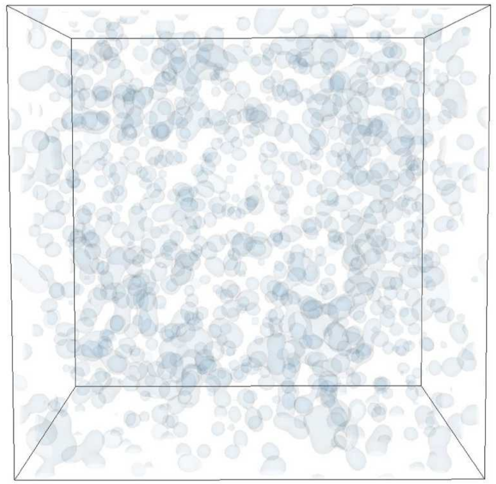
- Stopping at 10 g/cc (mass density) and 2 eV (temperature)
- **Force vs. projectile distance:** Similar across velocities
- **Work vs. projectile distance:** Spikes represent ions
- **Electronic work vs. projectile distance:** Slope represents stopping power



Projectile at 300 keV



Projectile at 30 MeV



Dynamic Structure Factor

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)

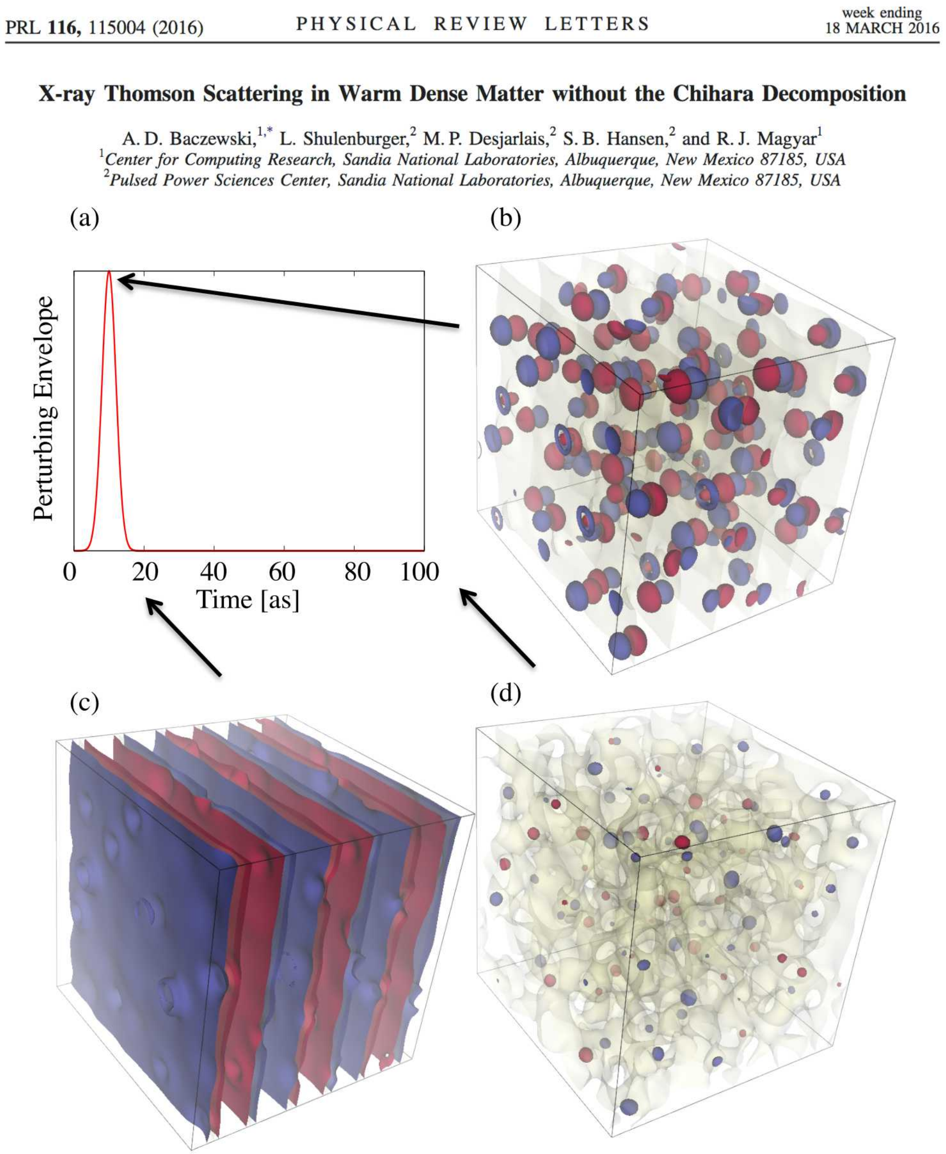
Record density response:

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

Apply dissipation-fluctuation theorem:

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

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


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. Magyar[1]
[1]*Center for Computing Research, Sandia National Laboratories, Albuquerque, New Mexico 87185, USA*
[2]*Pulsed Power Sciences Center, Sandia National Laboratories, Albuquerque, New Mexico 87185, USA*

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X-ray Thomson scattering in high energy density plasmas

Siegfried H. Glenzer and Ronald Redmer

Rev. Mod. Phys. **81**, 1625 – Published 1 December 2009

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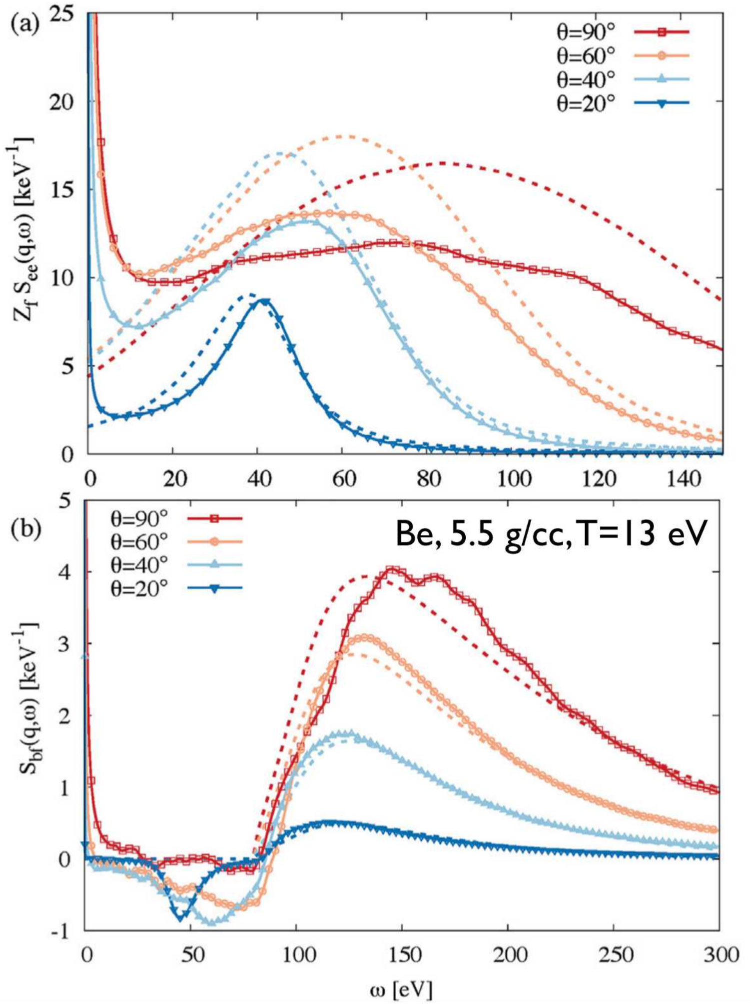

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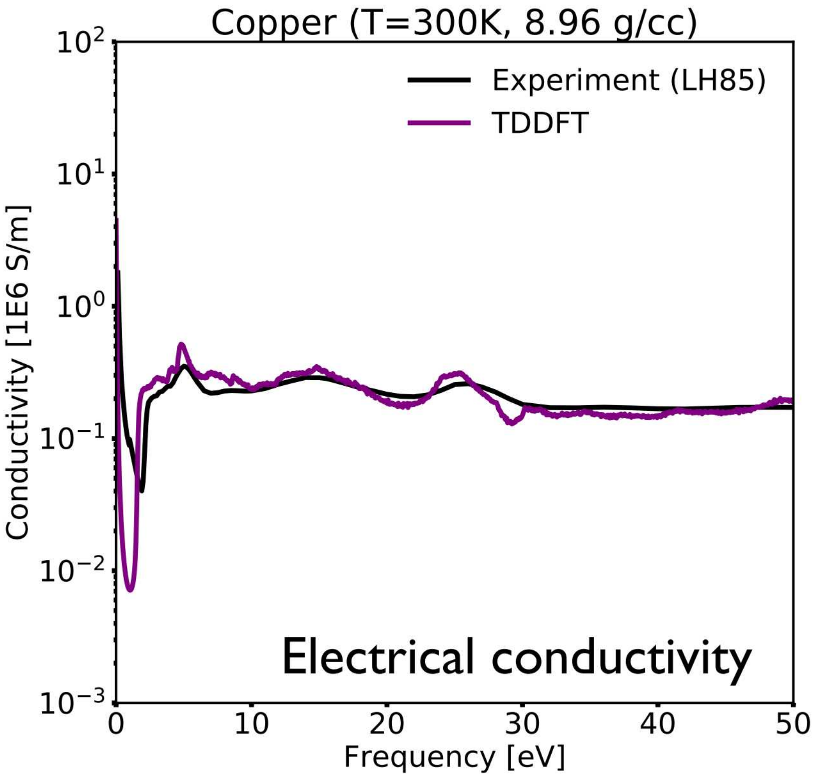
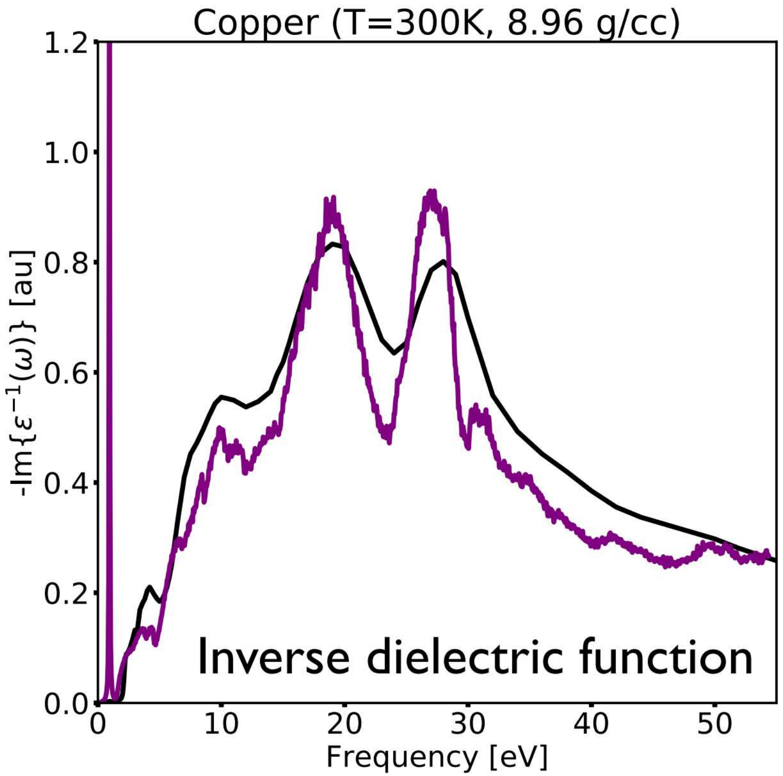
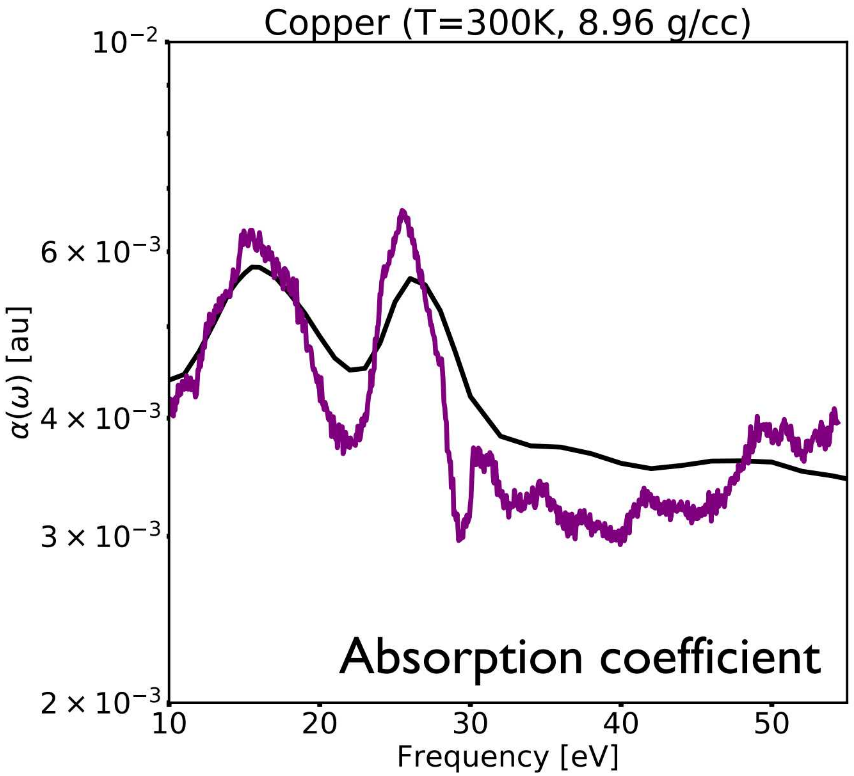
Copper under ambient conditions







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

	Sum rule	Error	Relative error
Experiment	-1.50	-0.07	0.04
TDDFT (I)	-1.41	-0.15	0.10
TDDFT (II)	-1.36	-0.21	0.14

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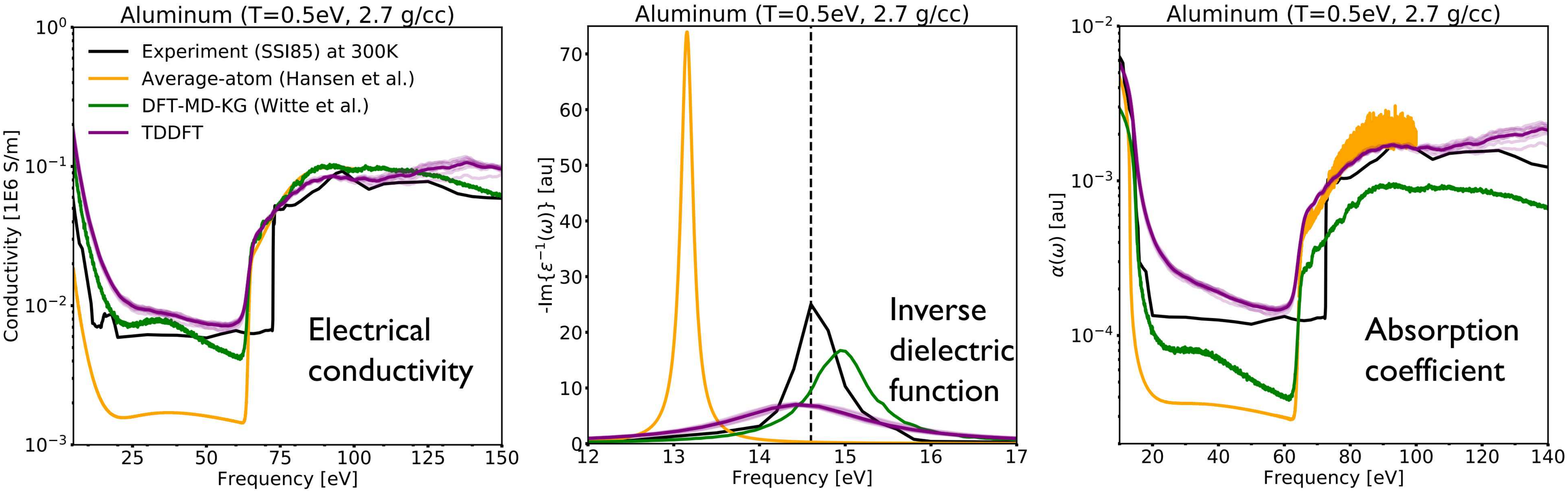
Isochorically heated Aluminum



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

	Sum rule	Error	Relative error
Experiment	-1.577	0.007	-0.004
DFT-MD-KG	-1.568	-0.003	0.002
TDDFT (11)	-1.563	-0.007	0.005

Summary

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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)
- Taisuke Nagayama (Sandia National Laboratories)
- Thomas Gomez (Sandia National Laboratories)
- Stephanie B. Hansen (Sandia National Laboratories)

Thanks for your attention!

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Supplemental Material

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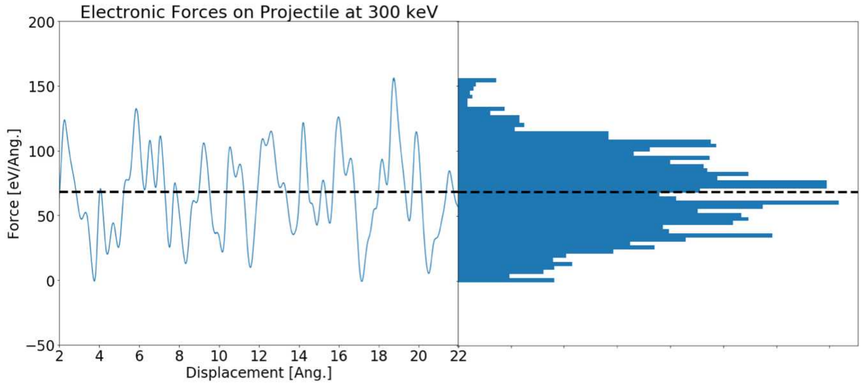
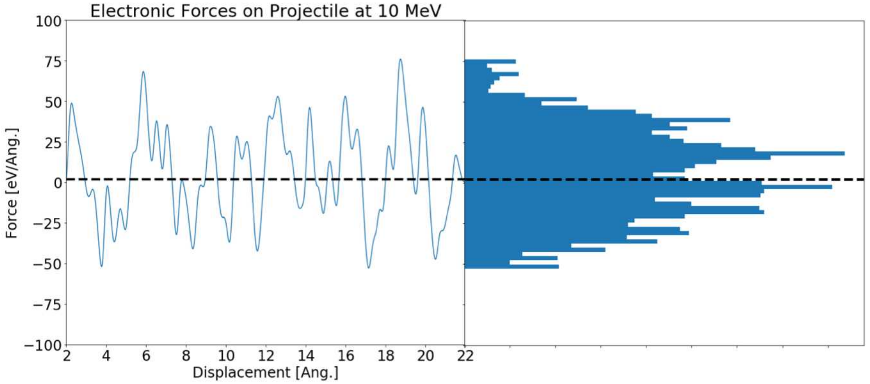
Distribution of microscopic forces in warm dense beryllium





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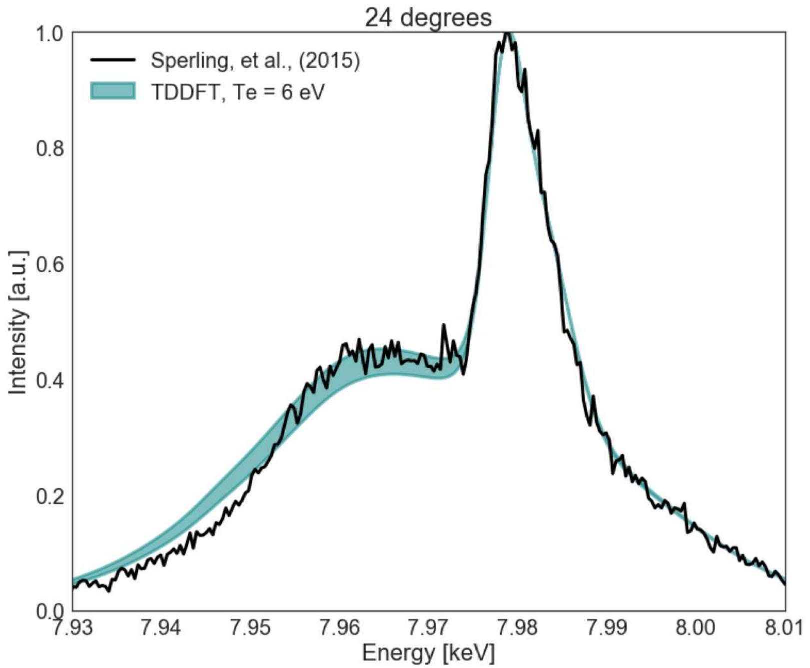
Supporting diagnostics at LCLS with TDDFT



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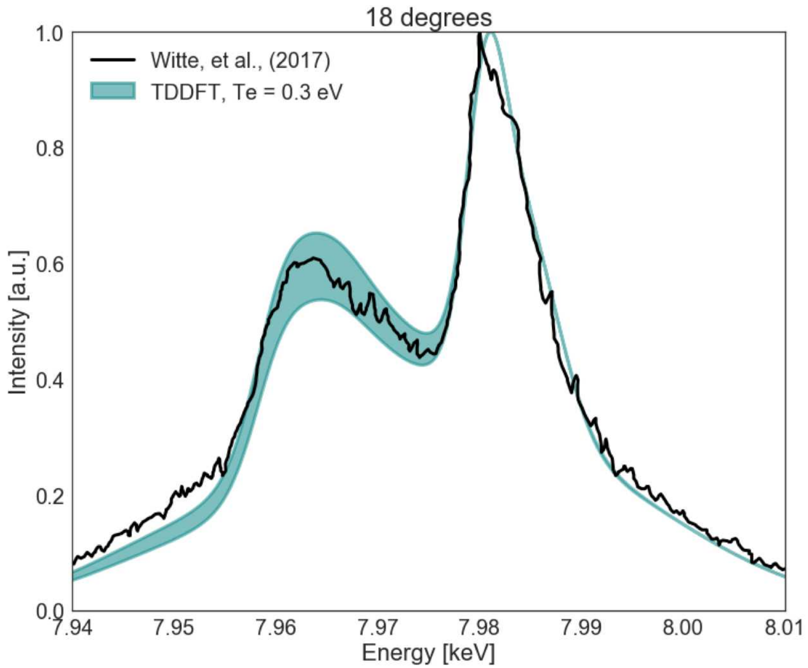
Phys. Rev. Lett. **115**, 115001 – Published 9 September 2015



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

Definitions of Transport Properties

$$\chi(\omega) = \chi_1(\omega) + \mathrm{i}\,\chi_2(\omega)$$

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

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

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

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

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

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

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

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

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