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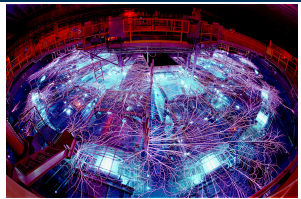

# Optical Conductivity of Magnetized Warm Dense Matter Using Time-dependent Density Functional Theory

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U.S. DEPARTMENT OF ENERGY | NNSA National Nuclear Security Administration

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# Optical Conductivity and Magnetized Warm Dense Matter

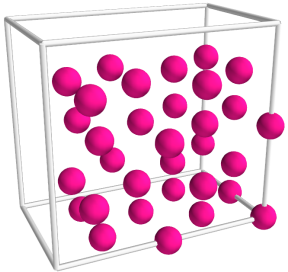
$\sigma$ and WDM

- Constitutive relations

$$j_i(\omega) = \sigma_{ij}(\omega) E_j(\omega)$$
$$D_i(\omega) = \varepsilon_{ij}(\omega) E_j(\omega)$$
$$H_i(\omega) = \mu_{ij}(\omega) B_j(\omega)$$

- Maxwell's equations

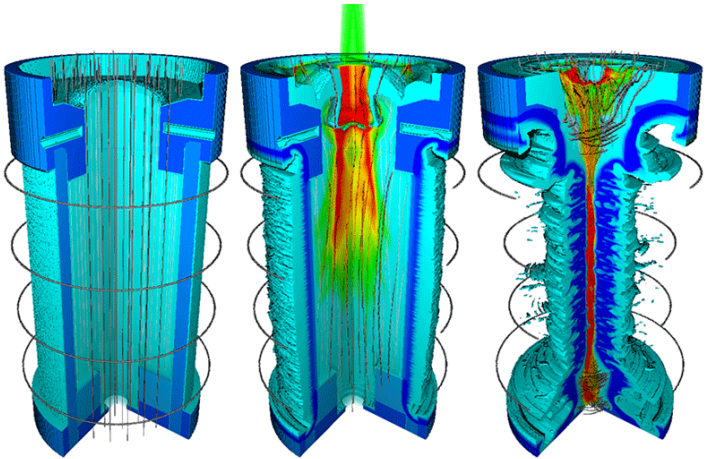
$$\nabla \cdot \mathbf{D} = \rho_{\text{ext}} \qquad \nabla \times \mathbf{H} - \frac{\partial \mathbf{D}}{\partial t} = \mathbf{j}_{\text{ext}}$$
$$\nabla \cdot \mathbf{B} = 0 \qquad \nabla \times \mathbf{E} + \frac{\partial \mathbf{B}}{\partial t} = 0$$



# Magnetized Liner Inertial Fusion (MagLIF)

Gomez et al. 2014, *Physical Review Letters*

1. $I_z = 19$ MA in shell produces $B_\phi$
2. $B_{z,\text{ext}} \approx 10$ T and preheat with laser
3. Increased fuel temperature during implosion



# Time-Dependent Density Functional Theory (TDDFT)

## Kohn-Sham Equations

- Time-dependent Kohn-Sham equations

$$\imath \frac{\partial}{\partial t} \phi_i (\mathbf{r}, t) = \left[ -\frac{1}{2} \boldsymbol{\nabla}^2 + v_{\mathrm{KS}} [n] (\mathbf{r}, t) \right] \phi_i (\mathbf{r}, t)$$

$$n (\mathbf{r}, t) = \sum_i |\phi_i (\mathbf{r}, t)|^2$$

$$v_{\mathrm{KS}} [n] (\mathbf{r}, t) = v_{\mathrm{ext}} (\mathbf{r}, t) + v_{\mathrm{H}} [n] (\mathbf{r}, t) + v_{\mathrm{xc}} [n] (\mathbf{r}, t)$$

$$v_{\mathrm{ext}}(\mathbf{r}) = -\sum_{A=1}^{K} \frac{Z_A}{|\mathbf{r} - \mathbf{R}_A|}, \qquad v_{\mathrm{H}}[n](\mathbf{r}, t) = \int \mathrm{d}\mathbf{r}' \, \frac{n(\mathbf{r}', t)}{|\mathbf{r} - \mathbf{r}'|}, \qquad v_{\mathrm{xc}}[n](\mathbf{r}, t) = \frac{\delta E_{\mathrm{xc}}}{\delta n(\mathbf{r}, t)}$$

- Runge-Gross Theorem (Runge and Gross 1984, *Physical Review Letters*)

$$v_{\mathrm{KS}} (\mathbf{r}, t) \overset{\Psi_0}{\longleftrightarrow} n (\mathbf{r}, t)$$

# Time-Dependent Density Functional Theory (TDDFT)

## Limitations and modifications

- Approximate exchange-correlation functionals $E_{\text{xc}}$
  - Adiabatic approximation

$$v_{\text{xc}}[n](\mathbf{r},t) \approx v_{\text{xc}}[n_0](\mathbf{r},t)\big|_{n_0 \to n(\mathbf{r},t)}$$

- Electronic temperature effects [1]

$$n(\mathbf{r}) = \sum_{j=1}^{N} \left\{1 + \exp\left[\left(\epsilon_j^{\tau} - \mu\right)/\tau\right]\right\}^{-1} |\phi_j(\mathbf{r})|^2$$

- Magnetic fields require a vector potential
  - $\mathbf{E} = -\nabla v - \partial \mathbf{A}/\partial t$ but $\mathbf{B} = \nabla \times \mathbf{A}$
- Periodic systems and Runge-Gross Theorem assumption[2]
  - Time-dependent current density functional theory (TDCDFT)
  - Magnetic field density functional theory (BDFT)

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[1] Pribram-Jones et al. 2013, *Frontiers and Challenges in Warm Dense Matter, edited by F. Graziani, et al. (Springer)*

[2] Maitra, Souza, and Burke 2003, *Physical Review B*

# Magnetic Field and Vector Potential

Previous *ab initio* work

- Kootstra, Boeij, and Snijders 2000, *The Journal of Chemical Physics* and Romaniello and Boeij 2005, *Physical Review B*
  - Dielectric function for metallic and nonmetallic crystals
  - TDCDFT
  - ADF code
- BYU Master's Thesis, Jensen 2010 - Real-space implementation of TDCDFT
- Yabana et al. 2012, *Physical Review B*
  - Couple Maxwell's equations with TDDFT
  - Neglect orbital magnetization
  - SALMON code
- Krieger et al. 2015, *Journal of Chemical Theory and Computation*
  - Noncollinear spin-dependent TDDFT in periodic solids
  - Laser-induced demagnetization
  - ELK code

# Implementation

## TDDFT-modified VASP

- Real-time TDDFT-Ehrenfest MD in VASP
  - Plane-wave basis
  - Projector-augmented wave (PAW) formalism
  - Crank-Nicolson propagator
  - GMRES iterative linear solver
- Applications
  - X-ray Thomson Scattering in WDM [3]
  - Stopping power
  - Optical conductivity


- More VASP features
  - Spin effects
    - Strong $\mathbf{B}_{ext} \Rightarrow$ collinear spin
  - Finite temperature

[3] Baczewski et al. 2016, *Physical Review Letters*

# Vector Potential

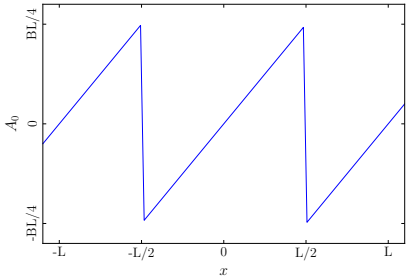
## Real-space representation

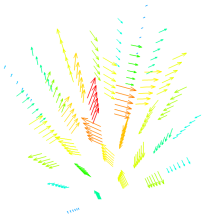
- Macrosopic constant $\mathbf{B}_{\text{ext}}$

$$\mathbf{A}_0(\mathbf{r}) = \frac{1}{2}\mathbf{B}_{\text{ext}} \times \mathbf{r}$$

  - Periodicity $\Rightarrow$ Saw-tooth $\Rightarrow$ discontinuity

- Gauge freedom



# Gauge-Including Projector Augmented Wave (GIPAW) Method

Pickard and Mauri 2001, *Physical Review B*

- Hamiltonian

$$H = \frac{1}{2}\left[-\imath\boldsymbol{\nabla} + \frac{1}{c}\mathbf{A}_{\mathrm{KS}}(\mathbf{r},t)\right]^2 + v_{\mathrm{KS}}(\mathbf{r},t)$$

- PAW = generalization of pseudopotential and LAPW methods
  - Avoid excessive Fourier components near ion cores
  - Eigenstates and phase factor after translation $\mathbf{r}'$

$$\tilde{\psi}_{n,\mathbf{k}}(\mathbf{r}) \rightarrow e^{(\imath/c)\mathbf{r}'\cdot\mathbf{A}(\mathbf{r})}\tilde{\psi}_{n,\mathbf{k}}(\mathbf{r}-\mathbf{r}')$$

- GIPAW $\rightarrow$ operators carry "phase twist"

$$\mathcal{T}_{\mathbf{B}} = \mathbf{1} + \sum_{\mathbf{R},n} e^{(\imath/c)\mathbf{R}\cdot\mathbf{A}(\mathbf{r})}\left[|\phi_{\mathbf{R},n}\rangle - |\tilde{\phi}_{\mathbf{R},n}\rangle\right]\langle\tilde{p}_{\mathbf{R},n}|\, e^{-(\imath/c)\mathbf{R}\cdot\mathbf{A}(\mathbf{r})}$$

- Similar to other NMR methods
  - GIAO
  - IGLO

# Current density

## Additional quantity

- Current density $\mathbf{j}(\mathbf{r}, t) \rightarrow \sigma$ and $\varepsilon$

$$j_i(\omega) = \sigma_{ij}(\omega) E_j(\omega)$$

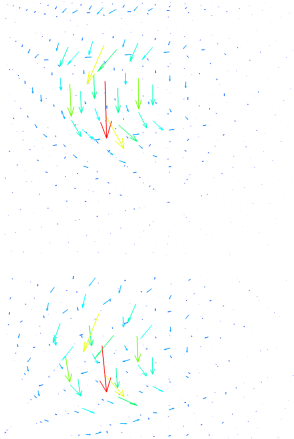
$$P_\alpha(\mathbf{r}, t) = -\frac{1}{\Omega_\mathbf{r}} \int^t \mathrm{d}t' \int \mathrm{d}\mathbf{r}' \mathbf{j}(\mathbf{r}', t')$$

$$P_\alpha(\mathbf{r}, \omega) = \chi_{ij}(\omega) E_j(\omega)$$

- Current operator

$$\mathbf{J}(\mathbf{r}') = -\frac{1}{2}\left[\mathbf{p}|\mathbf{r}'\rangle\langle\mathbf{r}'| + |\mathbf{r}'\rangle\langle\mathbf{r}'|\mathbf{p}\right] - \frac{\mathbf{A}(\mathbf{r}')}{c}|\mathbf{r}'\rangle\langle\mathbf{r}'|$$

  - GIPAW $\Rightarrow \bar{J} = \mathcal{T}_\mathbf{B}^\dagger \mathbf{J} \mathcal{T}_\mathbf{B}$



# Conclusions and Acknowledgements

## To-do list

- *Ab initio* calculation of optical conductivity
  - Not many knobs to turn
- Work in progress
  - Choice of gauge
  - Time dependence
  - Convergence analysis
    - Plane wave cutoff, time step, etc.
- Verification
  - Small test cases
  - Comparison with low temperature


- Collaborators
  - Andrew Baczewski
  - Attila Cangi
  - Stephanie Hansen

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