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Sandia National Laboratories

HED, DFT, and QMC – Connecting electronic structure methods to multi-physics simulations in HED/ICF

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Sandia National Laboratories

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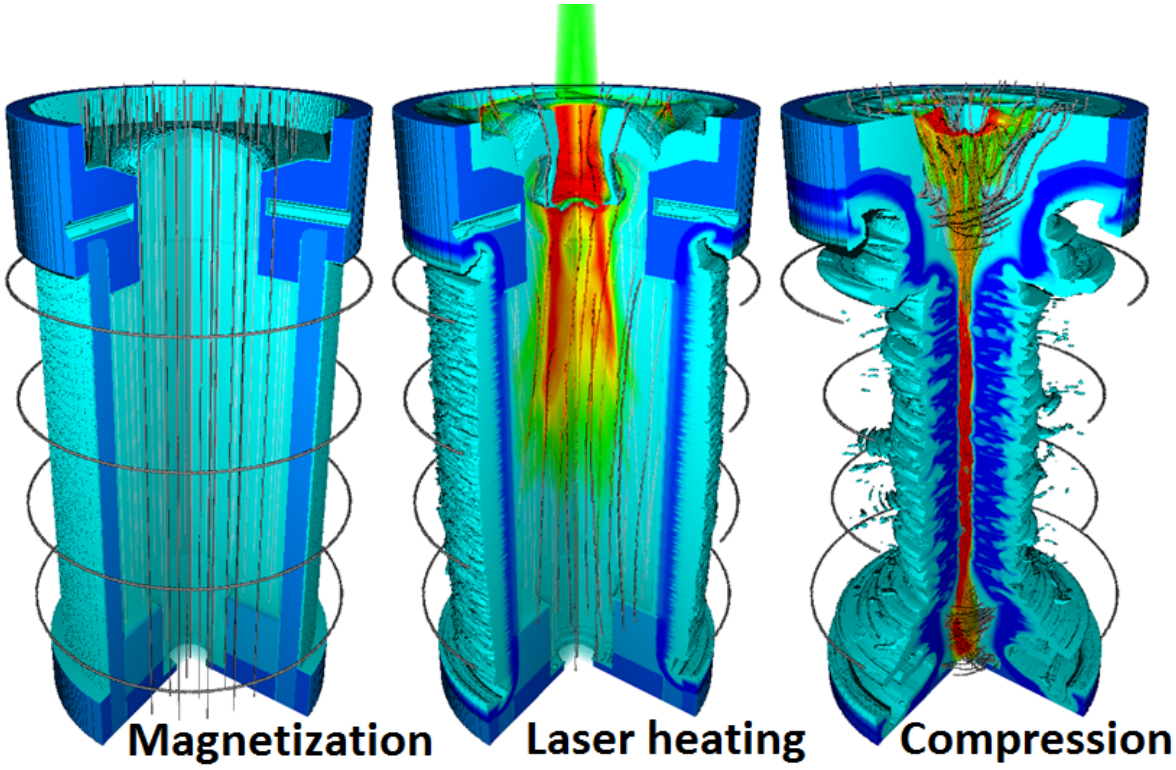
Multiphysics simulations are essential to design, optimize, and understand experiments and processes in HED physics



Sandia ICF Concept MagLIF: Slutz, Gomez, Harvey-Thompson, Ruiz, and others [2013-2023].

HED experiments across the world using many different platforms (short- and long pulse lasers, pulsed power, gas driven launchers) are designed, optimized, and analyzed/ understood using radiation-magneto-hydrodynamics simulation codes.

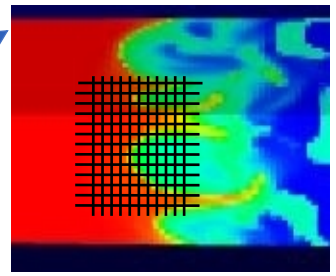
Simulations can be mesmerizing in resolving details and illustrating phenomena

What does is take to do it well?

Material models are an integral component of radiation and magneto-hydrodynamic simulations

The hydrodynamics moves material based on pressure gradients and body forces.

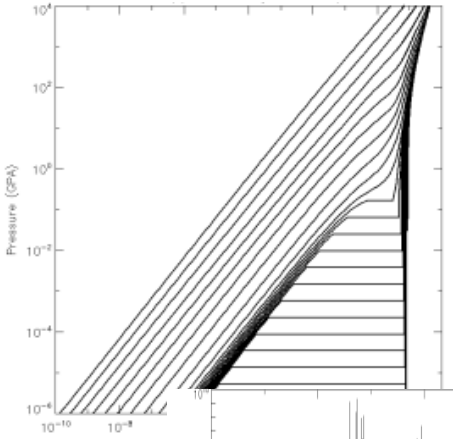
Hydrodynamics



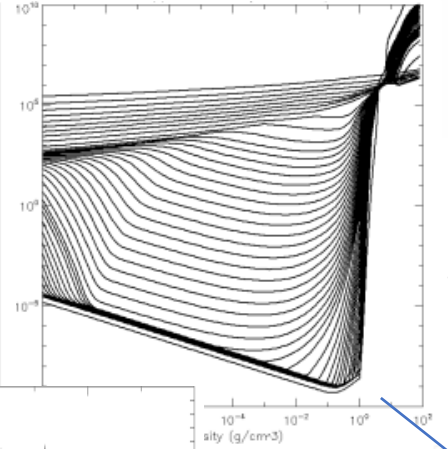
Tom Haill, SNL

Hydrodynamic evolution – change in temperature and volume/ pressure/ density

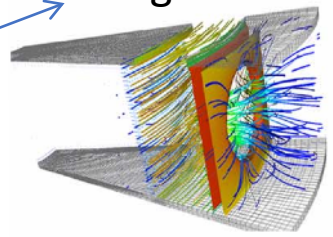
Equation of State



Conductivity



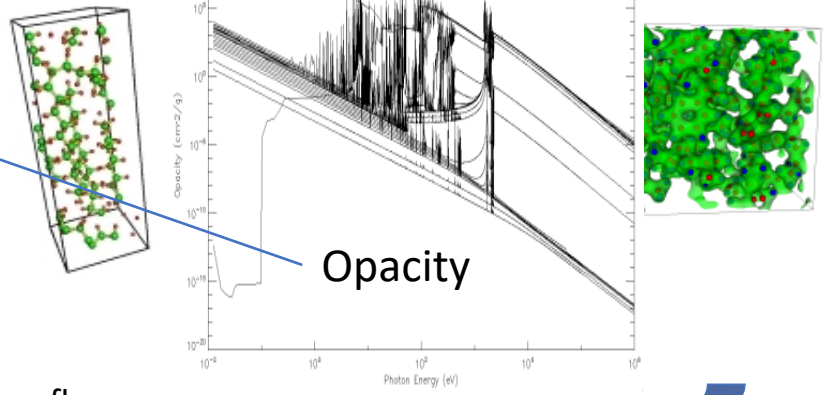
Magnetics



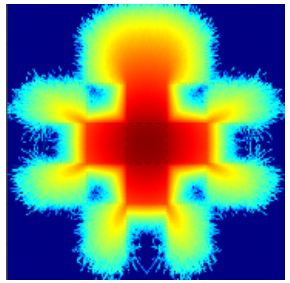
Chris Garasi, SNL

Magnetic field evolution – joule heating rate and magnetic pressure gradients pushing plasmas

Opacity



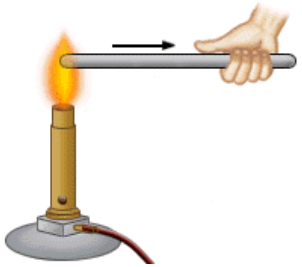
Radiation



Tom Brunner, LLNL

Radiation flow

Conduction



Thermal conduction – equilibrating temperature

High fidelity simulations in HED require expertise across a broad range of fields

Chain of capabilities – where is the weak link for your application?

Other things equal – Material models often make the difference between qualitative and quantitative

The Equation of State (EOS) governs the hydrodynamic and thermal evolution in the simulation

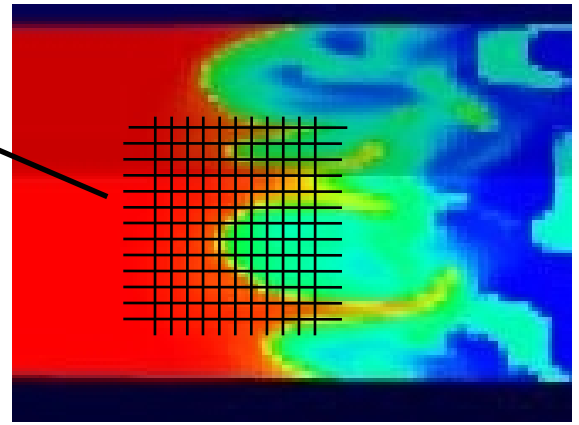
The EOS connects density, temperature, internal energy and pressure: $E(\rho,T)$, $P(\rho,T)$ for each specific material

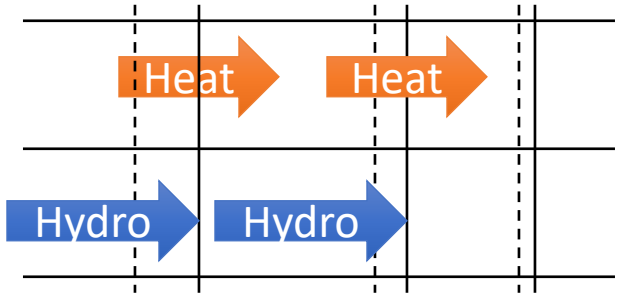
ρ_1,E_1	ρ_2,E_2	ρ_3,E_3
ρ_4,E_4	ρ_5,E_5	ρ_6,E_6
ρ_7,E_7	ρ_8,E_8	ρ_9,E_9

P_4,T_4	P_5,T_5	P_6,T_6
P_7,T_7	P_8,T_8	P_9,T_9

Solve the multi-physics evolution of the system, heat conduction and hydrodynamics

Simulation of a shock wave in a foam
(Tom Haill, SNL)





Chain of capabilities – where is the weak link for your application?

Defects in material models can never be removed at higher levels by modifications in discretization algorithms, mesh, or time-integrators.

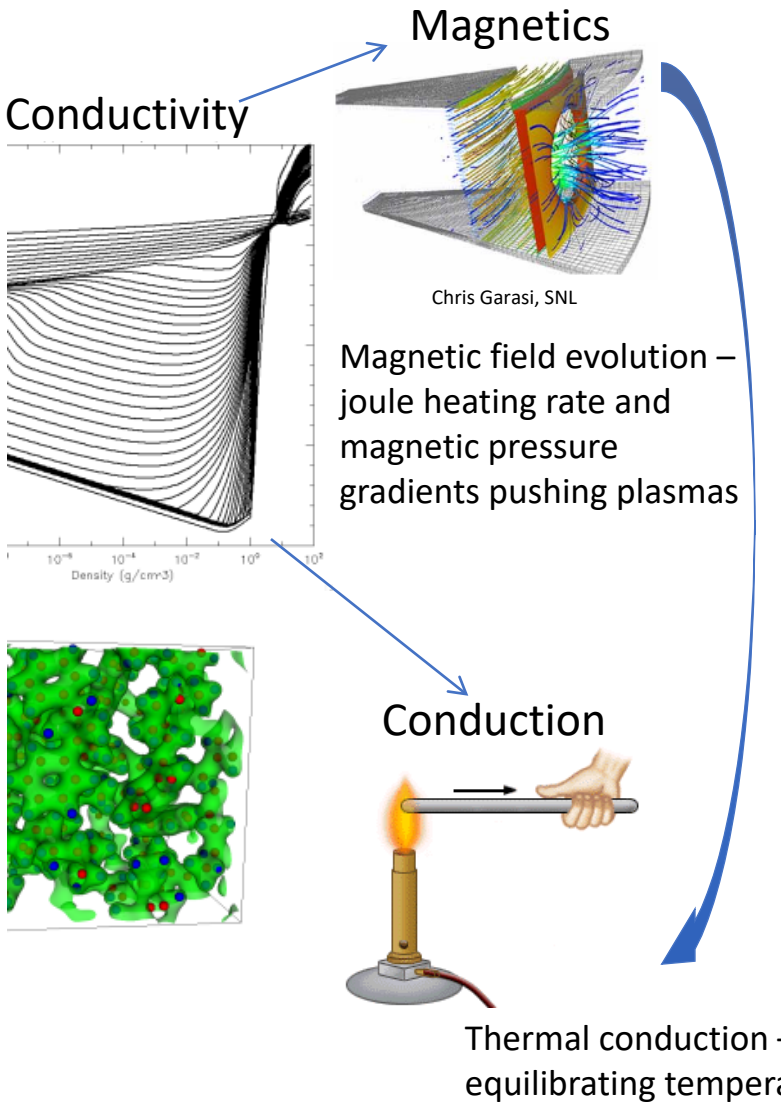
On the other hand – even the best material models won't help you if the multi-physics code has problems with discretization algorithms, meshing, energy conservation, or an outright lack of physics.

Density Functional Theory has emerged as an indispensable component of developing Equation of State models.

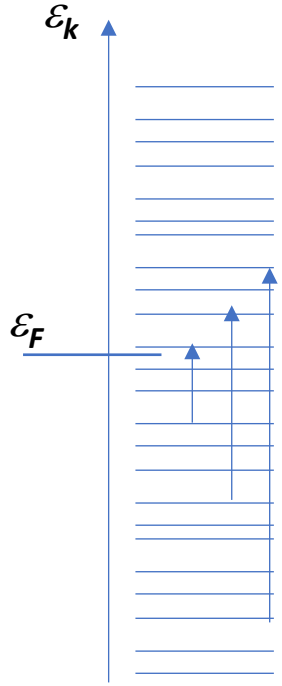
DFT Theory and Cold curves

Nathan Argaman

Electrical conductivity plays a key role for magnetized plasma physics at pulsed power facilities like Sandia's Z-Machine



- We calculate electronic and thermal conductivity using DFT
- Imaginary part of the dielectric function using the Kubo-Greenwood relation
  - Linear response
  - Matrix elements of single particle wavefunctions – Kohn-Sham orbitals
- Our experience is that the approach works well when going from solid into liquid into dense plasma – Warm Dense Matter
- *Potential issues when the band gap is not described well*



All possible transitions – weighted with Fermi occupancy

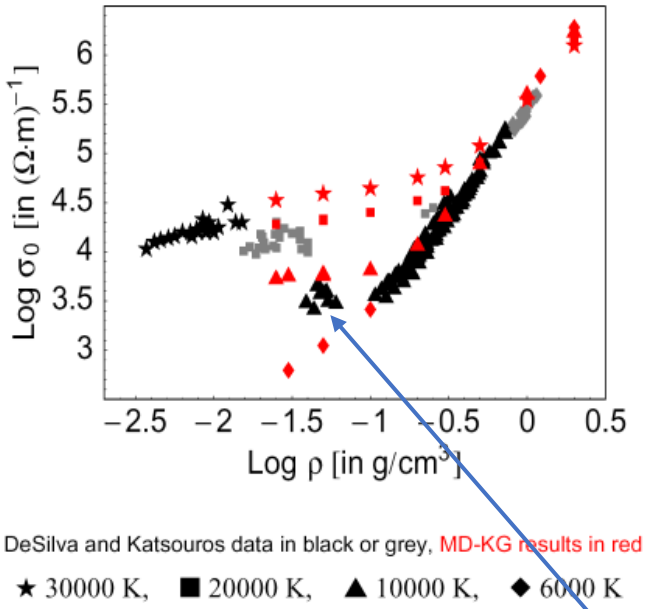
Contributions from partially occupied bands at the Fermi level

$$\sigma_e(\omega) = \frac{2\pi e^2}{3m^2\omega V}\sum_{\mathbf{k}} w_{\mathbf{k}} \sum_{j=1}^{N_b}\sum_{i=1}^{N_b}\sum_{\alpha=1}^{3}\left[f(\epsilon_{j,\mathbf{k}}) - f(\epsilon_{i,\mathbf{k}})\right]$$

$$\times \left|\langle \Psi_{j,\mathbf{k}}|\hat{p}_\alpha|\Psi_{i,\mathbf{k}}\rangle\right|^2 \delta(\epsilon_{i,\mathbf{k}} - \epsilon_{j,\mathbf{k}} - \hbar\omega),$$

DFT – linear response / Kubo-Greenwood is a powerful way to calculate electrical conductivity for materials

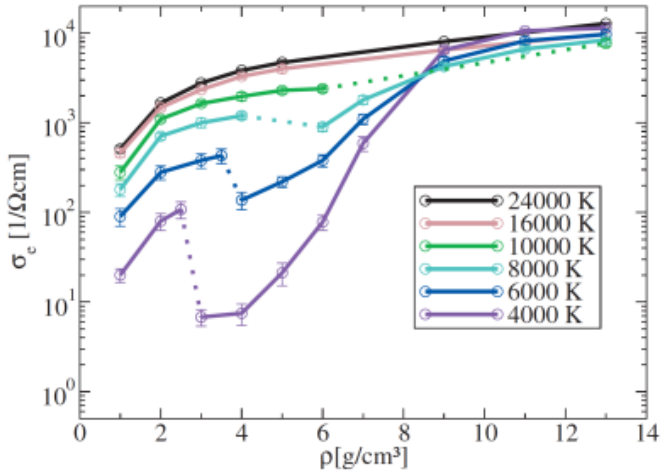
Aluminum electrical conductivity



Desjarlais, Kress, and Collins, Phys. Rev. E **66**, 025401 (2002).

Enabled predictive simulations of dynamic materials experiments on Sandia's Z-Machine – we predict flyer plate impact velocities to a few % !

Water electrical conductivity

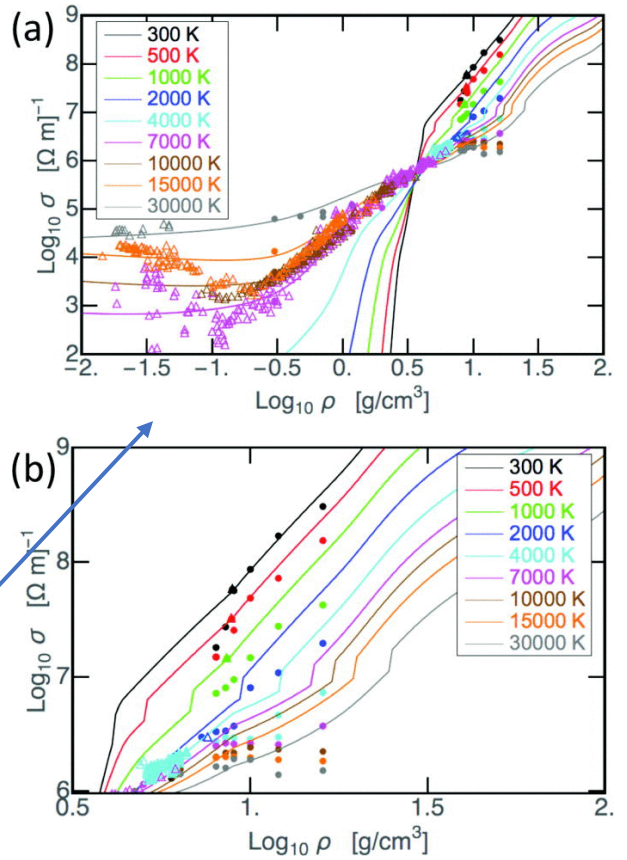


French, Mattsson, and Redmer, Phys Rev. B **82,** 174108 (2010).

Understand the structure of giant planets: magnetic fields and layers

Data from exploding wire experiments – probing warm dense matter conditions: A. W. Da Silva and team

Copper electrical conductivity



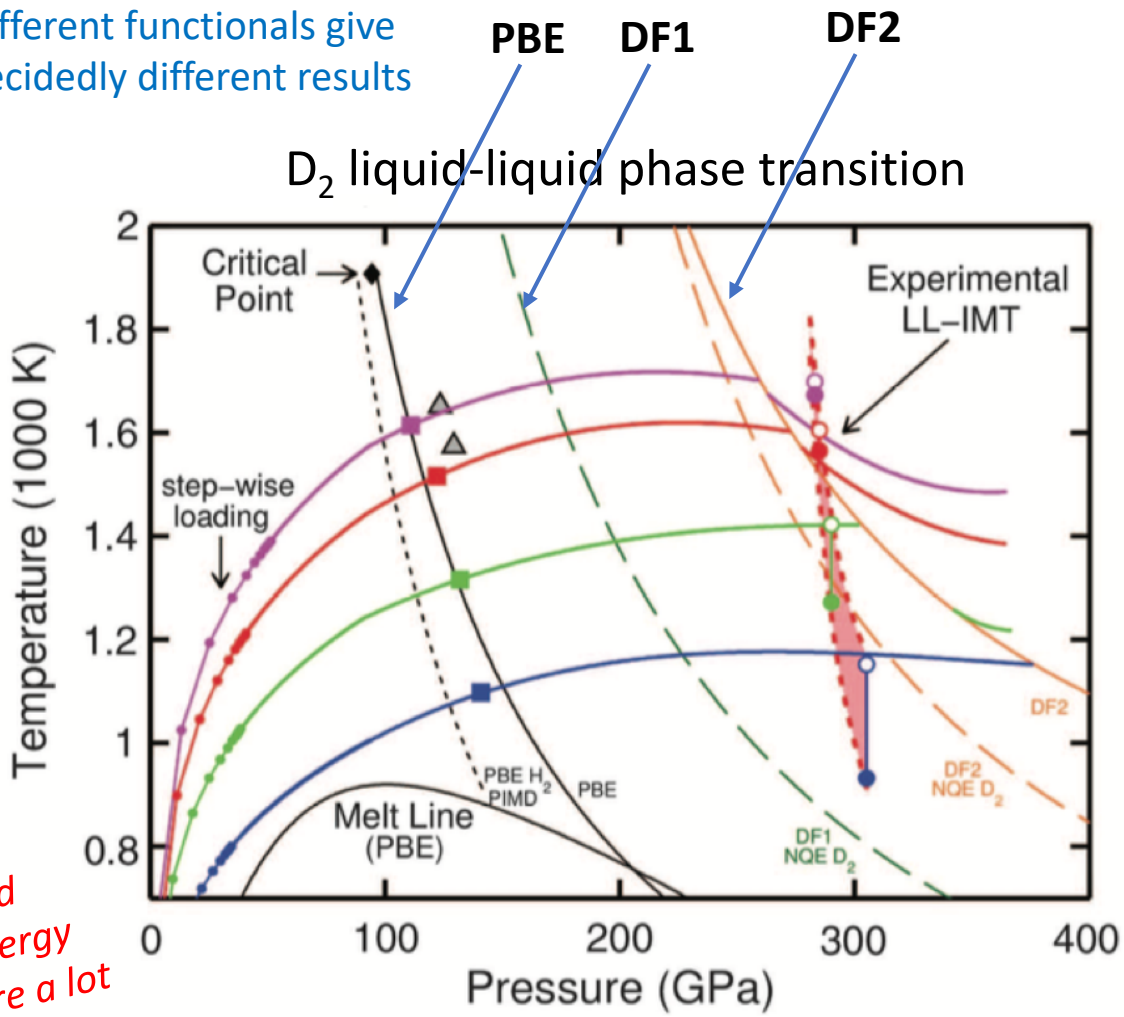
Cochrane, Lemke, Riford, and Carpenter, J. of Applied Phys. **119**, 105902 (2016).

We now use copper as a standard drive plate for dynamic materials experiments

DFT is a powerful method – but not without limitations

- Scaling with temperature
  - Memory ~ T^3
  - CPU time ~ $T^{4.5}$
- Approximations are notoriously difficult to improve
- *These shortcomings are not curiosities – they put practical limitations on the materials and conditions we can model with confidence*
  - *Challenges for strongly correlated systems – like transition metal oxides*
  - *Challenges at high temperatures – albeit not yet asymptotic ideal plasma temperatures*
- *There is no a priori way of knowing how accurate any given calculation will be*

Different functionals give decidedly different results



To be fair, D_2 dissociation is a VERY hard problem to get right and also small energy differences shift the transition pressure a lot

Knudson, Desjarlais, Becker, Lemke, Cochrane, Savage, Bliss, Mattsson and Redmer, Science **348**, 1455 (2015).

Quantum Monte Carlo Calculations offer a different approach – one without the functional dilemma of DFT

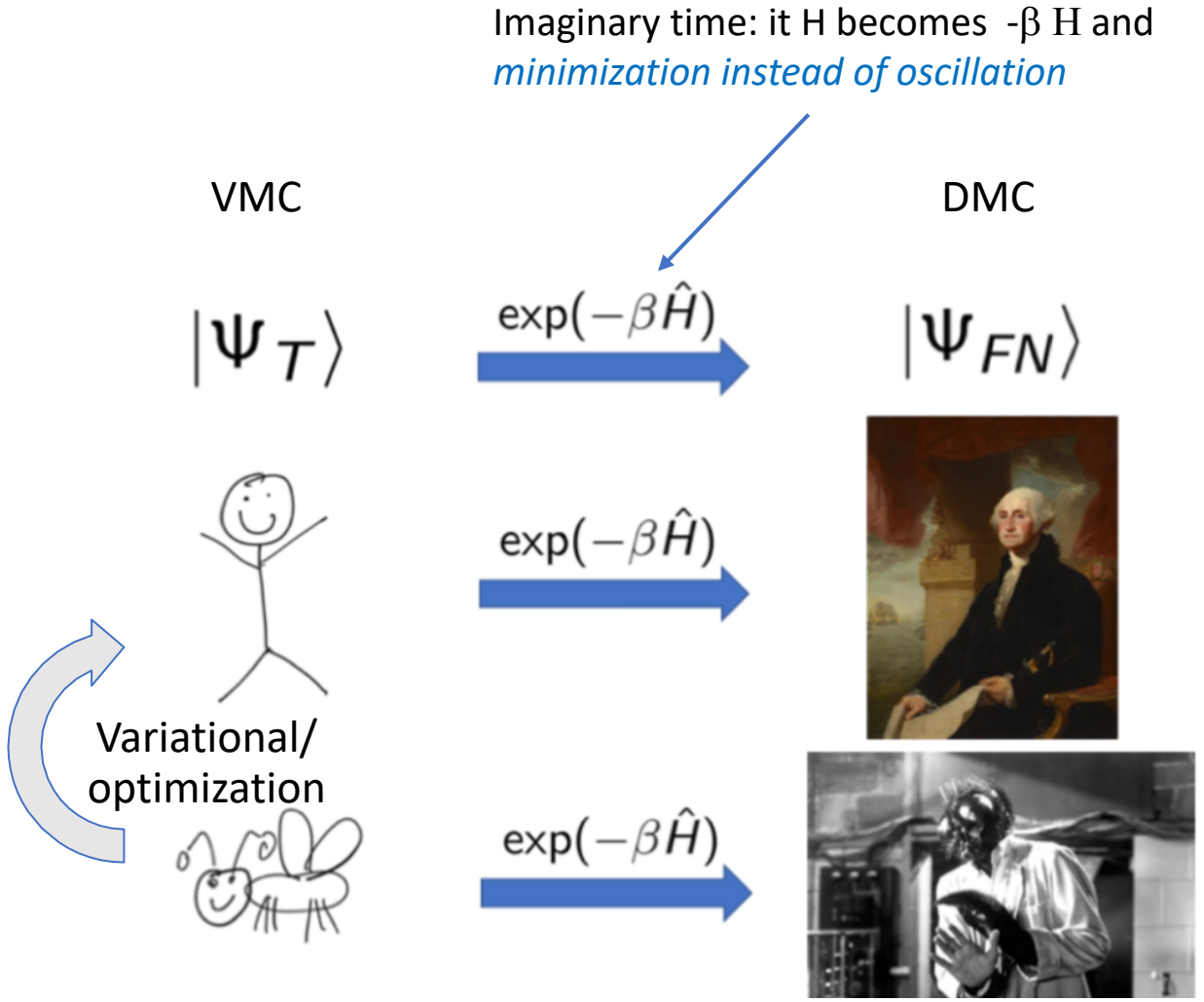
- Recast Schrodinger equation as an integral problem in 3N dimensions

$$\langle \hat{H} \rangle = \frac{\int \Psi^*(\mathbf{R}) \hat{H}(\mathbf{R}) \Psi(\mathbf{R}) d\mathbf{R}}{\int \Psi^*(\mathbf{R}) \Psi(\mathbf{R}) d\mathbf{R}}$$

- Massive parallelism available, each point can be calculated independently
- *Variational principle lets you know when your approximation is improving*
- Poor scaling if nontrivial trial wavefunction
  - 3 dimensions per electron
  - 20 points in each direction
  - $20^9 \approx 512$ billion points for 3 electrons
    - 3.8 TB just to store!
- Stochastic Methods scale much better for multidimensional integrals
- Effort for constant error scales as $1/\sqrt{N}$ regardless of dimensionality

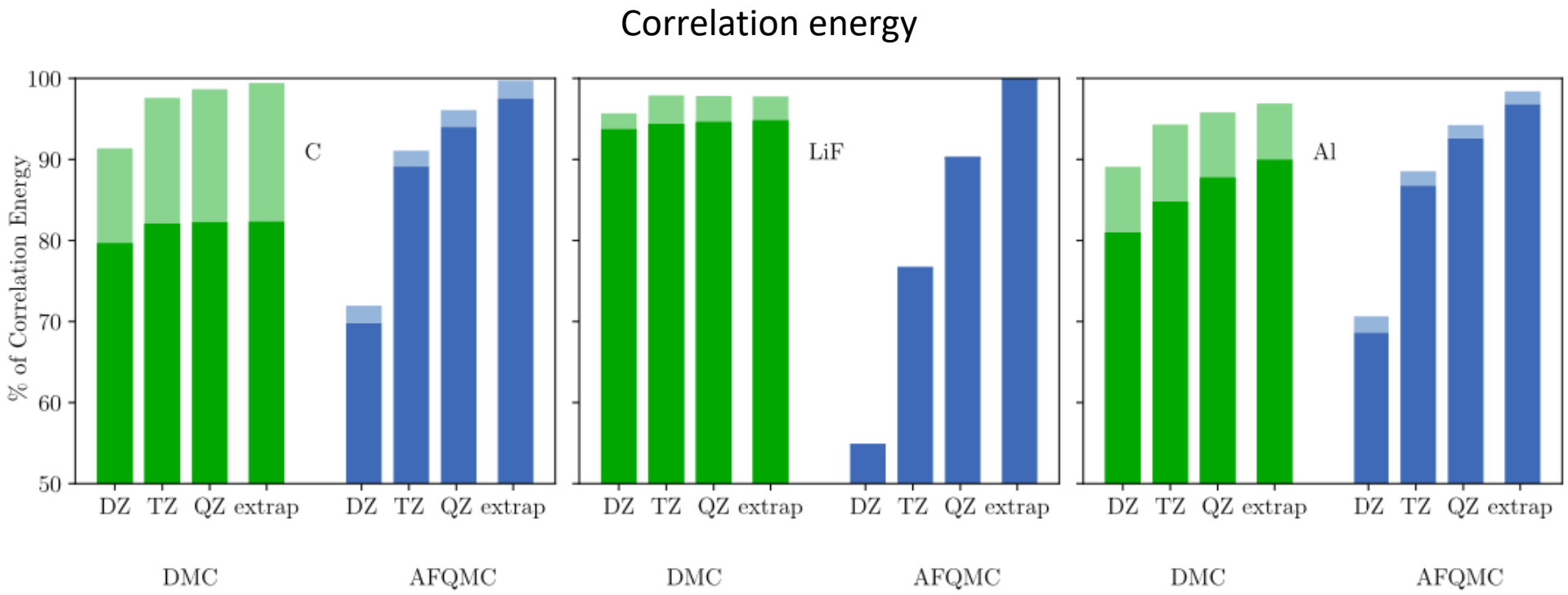
How is this used in practice?

- If you knew the many body wavefunction, you'd be done
- Variational Monte Carlo
- In practice, you build a guess from DFT and use the variational principle to optimize parameters in it
- Then you can use imaginary time projection (imaginary time Schrodinger equation looks like a diffusion equation) to improve things



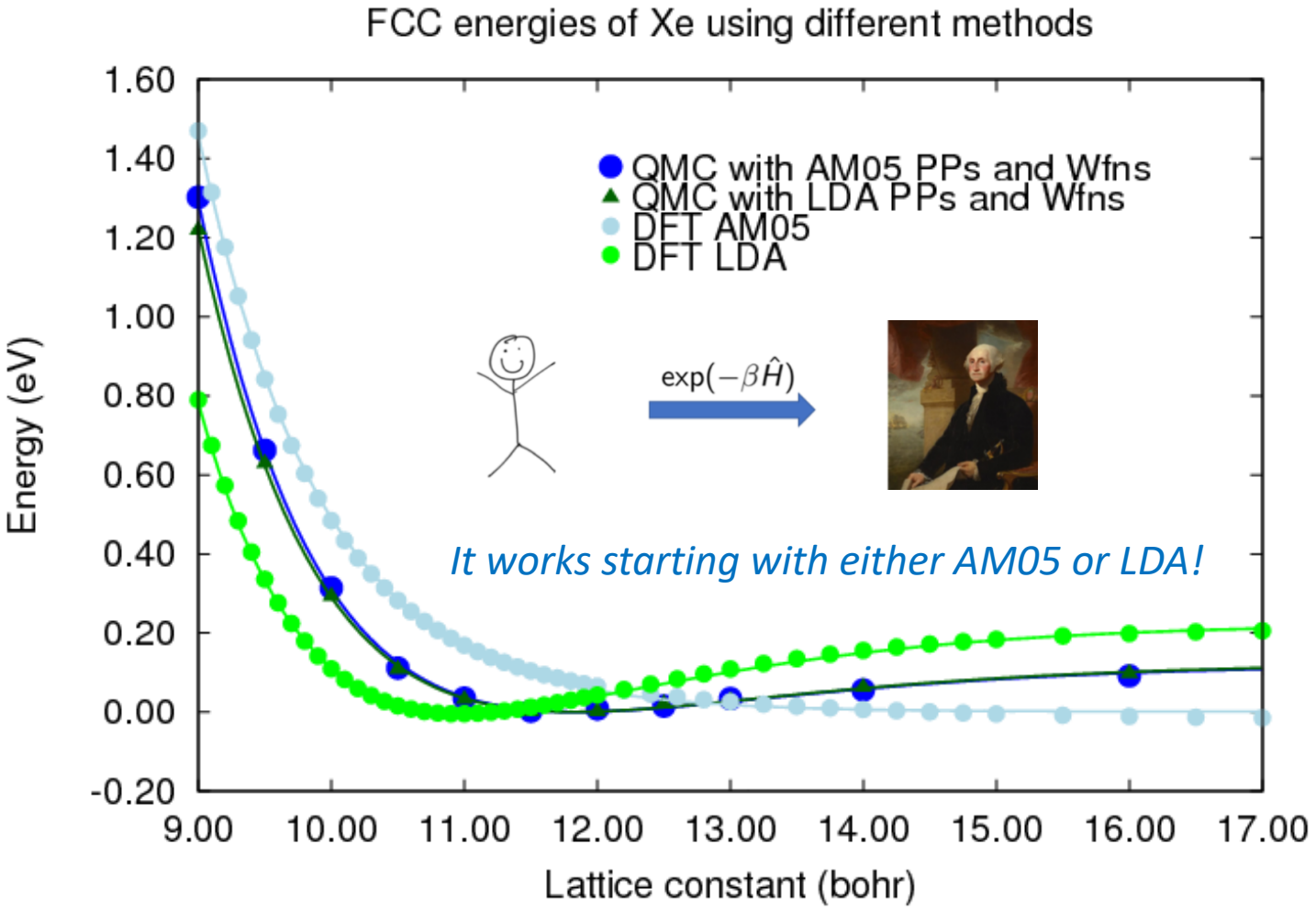
How is this different from DFT?

- It is possible to quantify errors
  - Strategy: Understand properties of errors, search for most impactful route to scalable improvements



Case study for Xe compression and melting at high pressure

- The two functionals LDA and AM05 give vastly different results for solid Xe due to van der Waals interactions
  - *LDA overbinds, AM05 is repulsive – no bond*
  - *DMC is expected to accurately treat van der Waals interactions*
  - *Large molecules, closed shell atoms, etc.*
- Validate approximations for known phase
- Compare E(V) curve of FCC xenon to experiment
  - Starting from two different points results in very similar answers
  - Both answers compare well to experiment



The triangles correspond to DFT or DMC simulations based on the LDA and the circles to DFT or DMC based on AM05.

Theory of melting at high pressures: Amending density functional theory with quantum Monte Carlo
L. Shulenburger, M. P. Desjarlais, and T. R. Mattsson
Phys. Rev. B **90**, 140104(R) (2014)

Thermodynamic integration can capture the difference between DFT and QMC – giving higher fidelity results

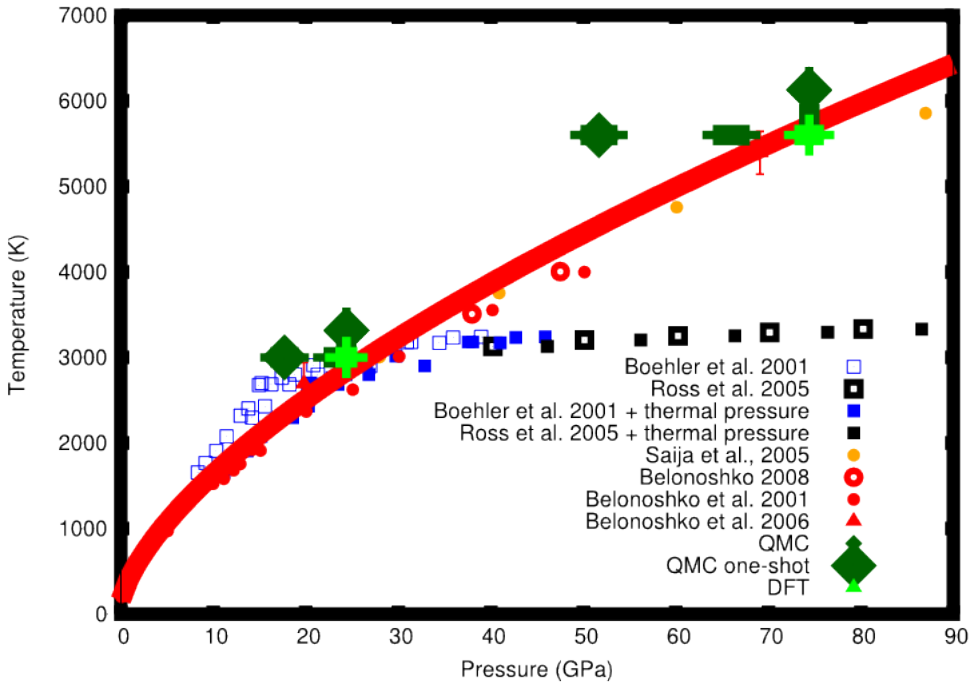
- Thermodynamic integration also allows relation of free energies from one interaction to another

- Use abstract parameter to tune from DFT interaction to DMC

$$\Delta F = \int_0^1 d\lambda \langle \Delta U \rangle_\lambda \approx \langle \Delta U \rangle_{\lambda=0} - \frac{1}{2k_B T} \langle (\Delta U - \langle \Delta U \rangle_{\lambda=0})^2 \rangle_{\lambda=0}$$

- Terms on right assume that difference in dynamics between DFT and DMC is small (fluctuation terms above are small)

Melting of Xe under pressure



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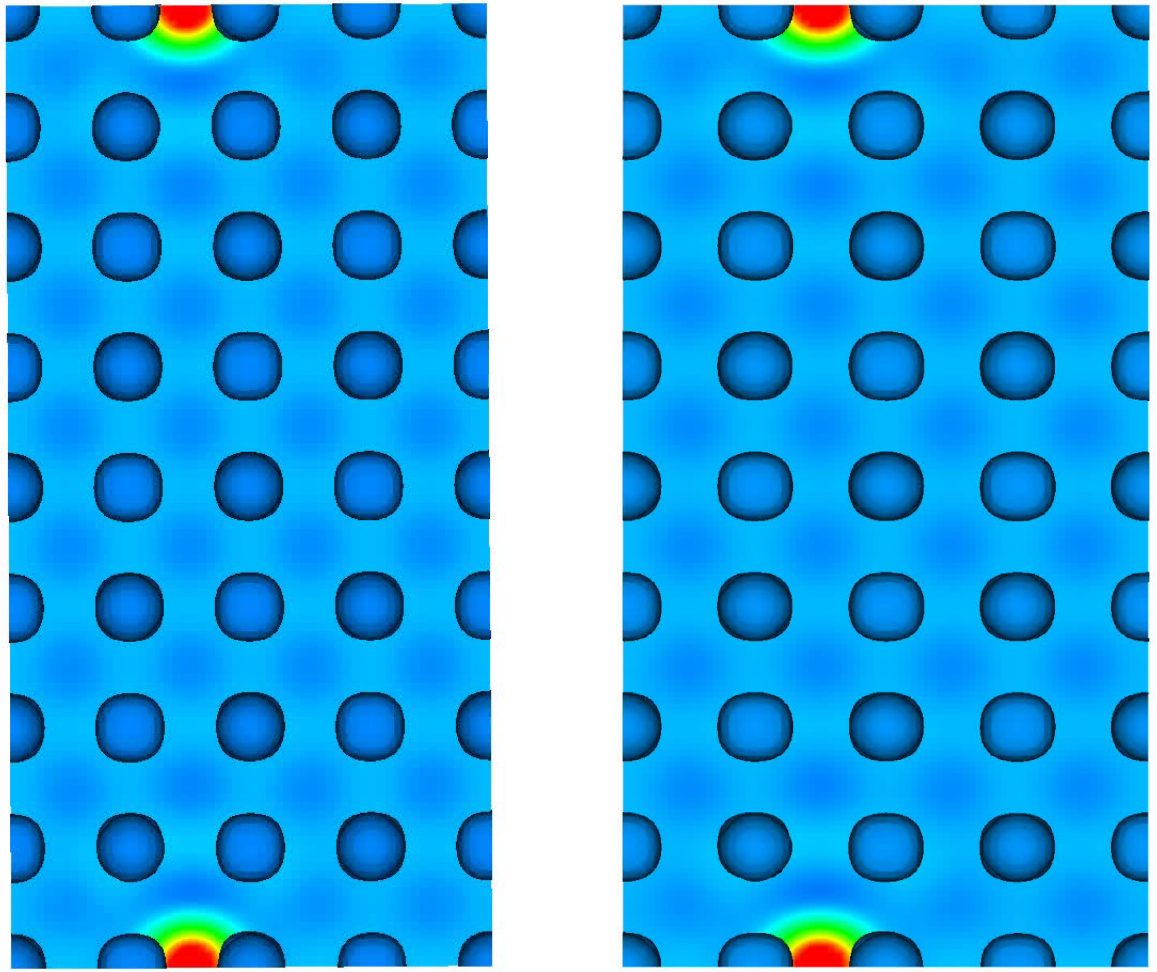
Going beyond linear response and Born-Oppenheimer approximation – Time-Dependent DFT for transport properties

- Dense disordered material, so periodic not cluster/ molecules
- Real-time evolution at finite temperature for non-harmonic nuclear motion to couple
- Coupled-electron-ion motion
- Electron-ion energy transfer
- Extended system optical or small-**q** response

Born-Oppenheimer DFT-MD

Ehrenfest TDDFT-MD



Send a proton into an Al crystal

TDDFT describes excitations of plasmons

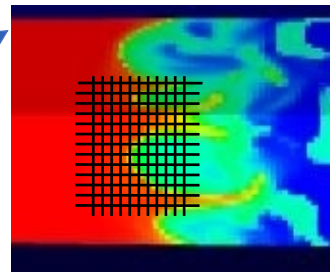
Stopping of Deuterium in Warm Dense Deuterium from Ehrenfest Time-Dependent Density Functional Theory, Magyar, Shulenburger, and Baczewski, Contributions to Plasma Physics **56**, 456-466 (2016).

Electrical conductivity of iron in Earth's core from microscopic Ohm's law, Ramakrishna, Lokamani, Baczewski, Vorberger, and Cangi, Phys. Rev. B **107**, 115131 (2023).

Material models are an integral component of radiation and magneto-hydrodynamic simulations

The hydrodynamics moves material based on pressure gradients and body forces.

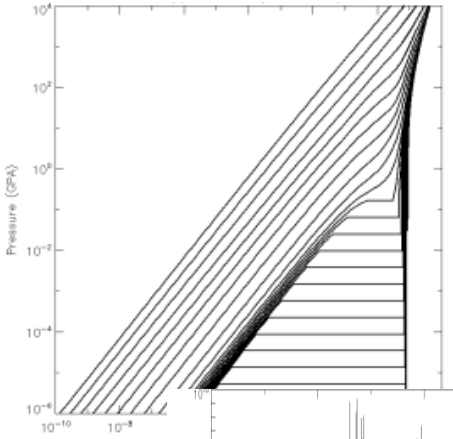
Hydrodynamics



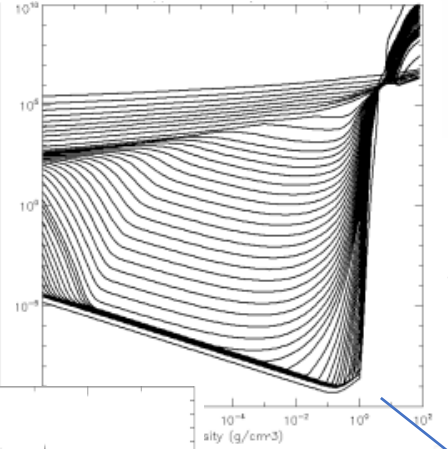
Tom Haill, SNL

Hydrodynamic evolution – change in temperature and volume/ pressure/ density

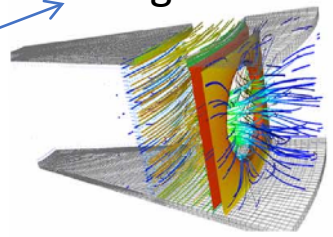
Equation of State



Conductivity



Magnetics



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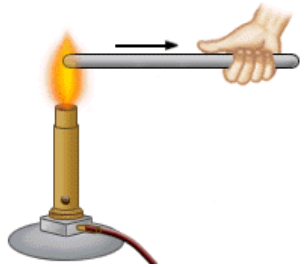
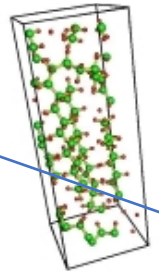
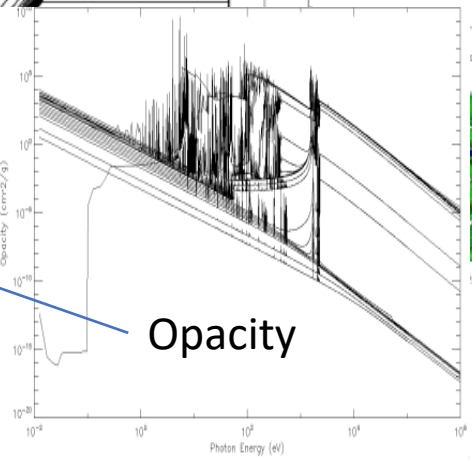
Magnetic field evolution – joule heating rate and magnetic pressure gradients pushing plasmas

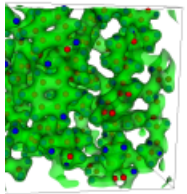
Conduction



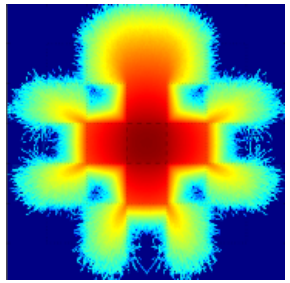
Opacity







Radiation



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Radiation flow

Material models often make the difference between qualitative and quantitative. results

Chain of capabilities – where is the weak link for your application?

What does is take to do it well?