Supporting ICF by quantum and orbital-free molecular dynamics

by F. Lambert

from CEA DAM DIF

at Computational challenges in warm dense matter, May 25$^{\text{th}}$

Quantum Physics makes me so happy.
It's like looking at the Universe naked.
S. Cooper

Contents

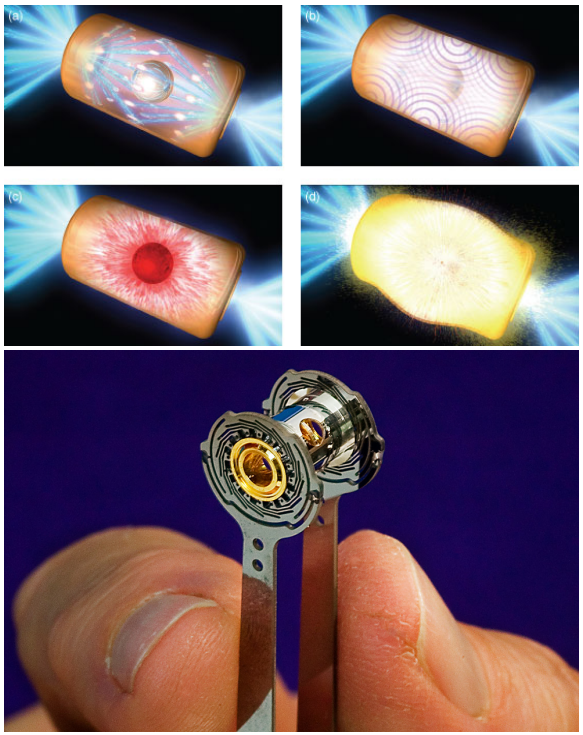
Contents

Inertial confinement fusion

- Compression of a Deuterium/Tritium layer to reach nuclear fusion;

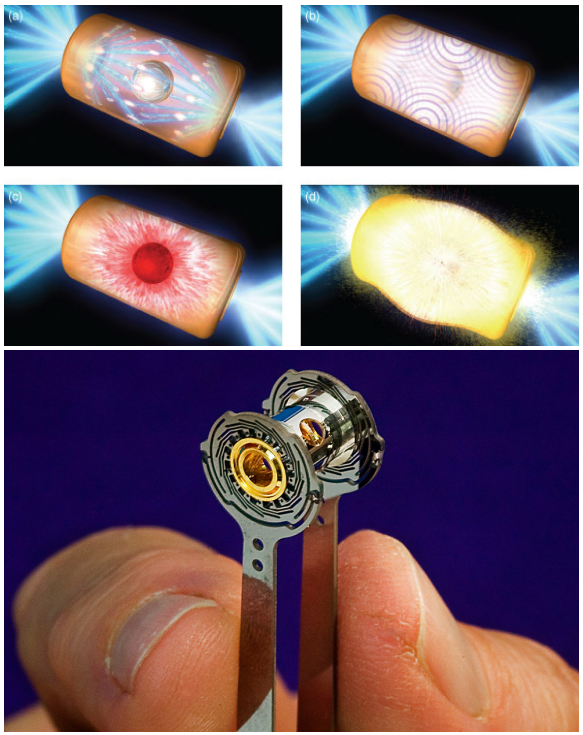
Inertial confinement fusion

- Compression of a Deuterium/Tritium layer to reach nuclear fusion;
- LASER heating of a gold cavity (indirect drive);



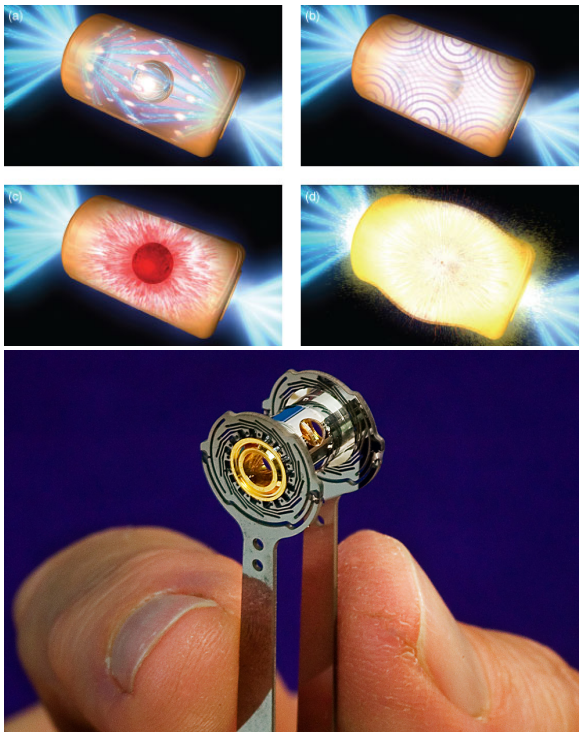
Inertial confinement fusion

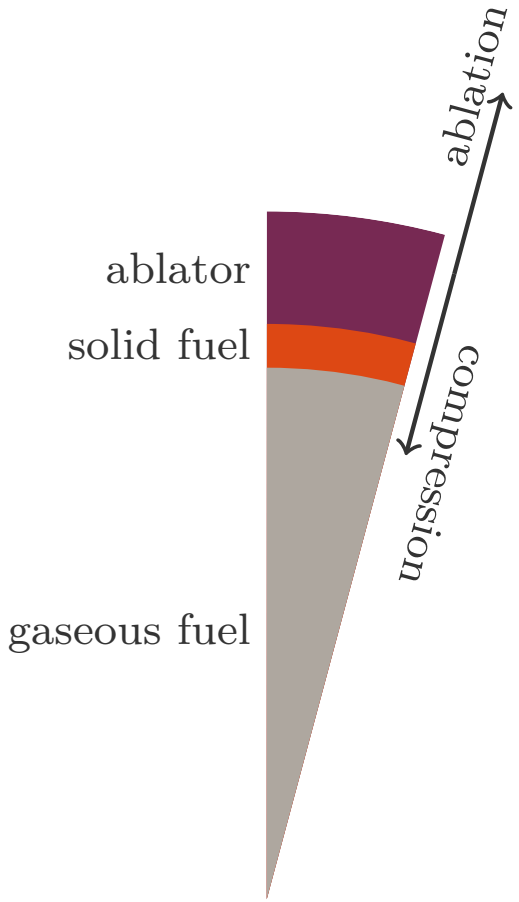
- Compression of a Deuterium/Tritium layer to reach nuclear fusion;
  - LASER heating of a gold cavity (indirect drive);
  - X-ray (black body) radiation (like an oven at 3.5 MK);



Inertial confinement fusion

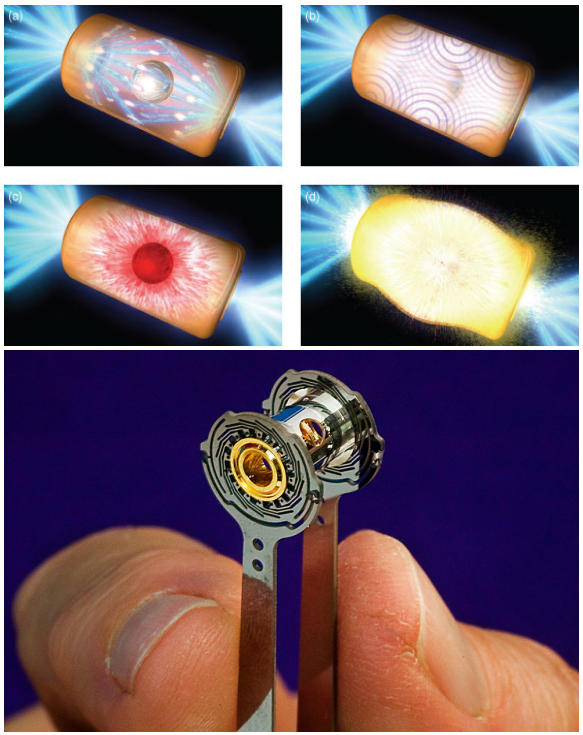
- Compression of a Deuterium/Tritium layer to reach nuclear fusion;
  - LASER heating of a gold cavity (indirect drive);
  - X-ray (black body) radiation (like an oven at 3.5 MK);
  - rocket effect: explosion/implosion of an outer plastic layer (generation of moderate shocks);

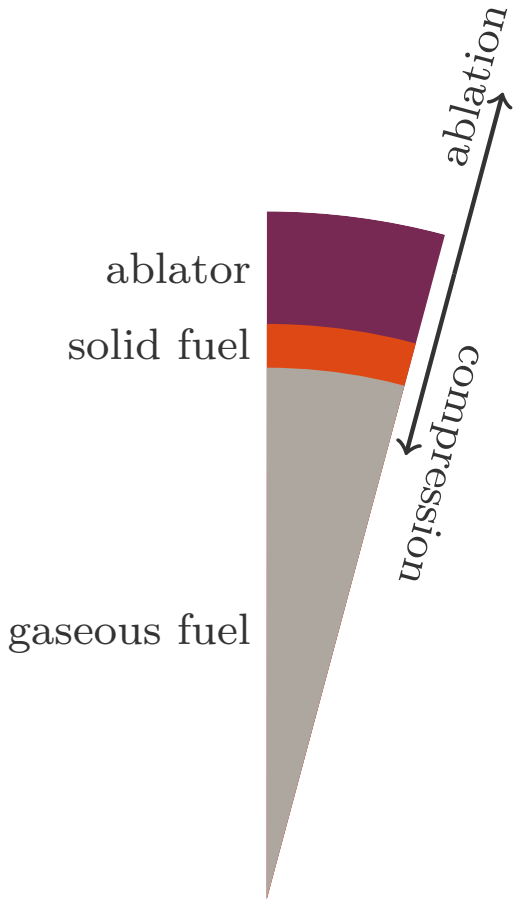




Inertial confinement fusion

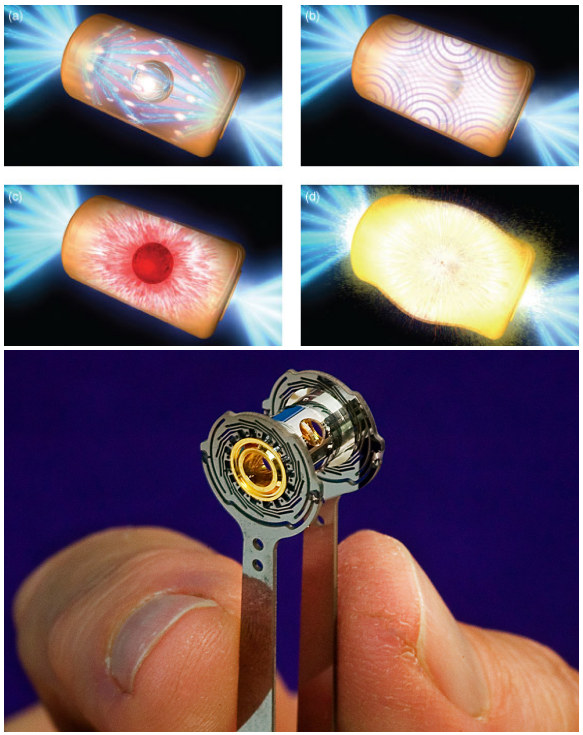
- Compression of a Deuterium/Tritium layer to reach nuclear fusion;
  - LASER heating of a gold cavity (indirect drive);
  - X-ray (black body) radiation (like an oven at 3.5 MK);
  - rocket effect: explosion/implosion of an outer plastic layer (generation of moderate shocks);
  - quasi-isentropic compression of the DT layer (shocks and final implosion);

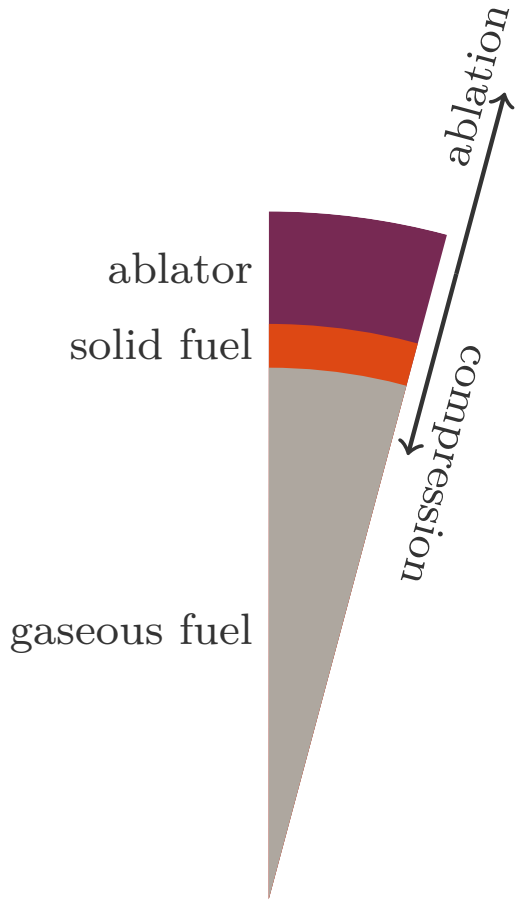




Inertial confinement fusion

- Compression of a Deuterium/Tritium layer to reach nuclear fusion;
  - LASER heating of a gold cavity (indirect drive);
  - X-ray (black body) radiation (like an oven at 3.5 MK);
  - rocket effect: explosion/implosion of an outer plastic layer (generation of moderate shocks);
  - quasi-isentropic compression of the DT layer (shocks and final implosion);
  - ignition: 100 low-fat-yogurt energy and 20 billions nuclear-power-plant power;





Inertial confinement fusion

- Compression of a Deuterium/Tritium layer to reach nuclear fusion;

- What about the underlying physics?
  - hydrodynamics of radiating flows, *e.g.* equation of energy conservation;
  - large thermodynamic domain: $kT \in [0 : 10\,\mathrm{keV}]$, $\rho \in \left[10^{-3} : 10^{3}\,\mathrm{g\,cm^{-3}}\right]$;

$$\rho \frac{\partial}{\partial t}\left(e + \frac{1}{\rho}\sigma T^4\right) + \left(p + \frac{1}{3}\sigma T^4\right)\boldsymbol{\nabla}\cdot\boldsymbol{u} = \boldsymbol{\nabla}\cdot\left(\kappa\boldsymbol{\nabla}T + \frac{\lambda_R c}{3}\sigma T^4\right)$$

Radiative hydrodynamics (1 cm, 20 ns)

target design: 300 simulations, from 30 min to 15 days

Inertial confinement fusion

- Compression of a Deuterium/Tritium layer to reach nuclear fusion;
- What about the underlying physics?
  - hydrodynamics of radiating flows, *e.g.* equation of energy conservation;
  - large thermodynamic domain: $kT \in [0 : 10\,\mathrm{keV}]$, $\rho \in \left[10^{-3} : 10^{3}\,\mathrm{g\,cm^{-3}}\right]$;
- What *must* be provided by electronic structure calculations?

$$\rho \frac{\partial}{\partial t}\left(e + \frac{1}{\rho}\sigma T^4\right) + \left(p + \frac{1}{3}\sigma T^4\right)\boldsymbol{\nabla}\cdot\boldsymbol{u} = \boldsymbol{\nabla}\cdot\left(\kappa\boldsymbol{\nabla}T + \frac{\lambda_R c}{3}\sigma T^4\right)$$

Radiative hydrodynamics (1 cm, 20 ns)

target design: 300 simulations, from 30 min to 15 days

Inertial confinement fusion

- Compression of a Deuterium/Tritium layer to reach nuclear fusion;
- What about the underlying physics?
  - hydrodynamics of radiating flows, *e.g.* equation of energy conservation;
  - large thermodynamic domain: $kT \in [0 : 10\,\mathrm{keV}]$, $\rho \in \left[10^{-3} : 10^{3}\,\mathrm{g\,cm^{-3}}\right]$;
- What *must* be provided by electronic structure calculations?
  - complex microscopic physics: low temperature solids or amorphous, partially electronic delocalization, strong correlations of electrons and nuclei, role of the radiation, *etc*;

$$\rho\frac{\partial}{\partial t}\left(e+\frac{1}{\rho}\sigma T^4\right)+\left(p+\frac{1}{3}\sigma T^4\right)\boldsymbol{\nabla}\cdot\boldsymbol{u}=\boldsymbol{\nabla}\cdot\left(\kappa\boldsymbol{\nabla}T+\frac{\lambda_R c}{3}\sigma T^4\right)$$

Radiative hydrodynamics (1 cm, 20 ns)

target design: 300 simulations, from 30 min to 15 days

routine call: 1 ms ↑ *fast* approximate models

Microscopic physics (10 nm, 1 ps)

thermodynamic equilibrium simulation: a few hours

What does it mean in terms of simulations?

- Several methods developed for solid, liquid or plasma physics:
  - one-center (average-atom like): quantum, computationally inexpensive;
  - multicenter:
    - quantum Monte-Carlo: electronic many-body;
    - molecular-dynamics based on either classical, quantum or semi-classical description.

What does it mean in terms of simulations?

- Several methods developed for solid, liquid or plasma physics:
  - one-center (average-atom like): quantum, computationally inexpensive;
  - multicenter:
    - quantum Monte-Carlo: electronic many-body;
    - molecular-dynamics based on either classical, quantum or semi-classical description.
- Material constraints in the high-energy-density regime:
  - low and high $\mathcal{Z}$ elements;
  - single or multi-specie;
  - low atomic fractions;
  - very different thermodynamic conditions;

What does it mean in terms of simulations?

- Several methods developed for solid, liquid or plasma physics:
  - one-center (average-atom like): quantum, computationally inexpensive;
  - multicenter:
    - quantum Monte-Carlo: electronic many-body;
    - molecular-dynamics based on either classical, quantum or semi-classical description.
- Material constraints in the high-energy-density regime:
  - low and high $\mathcal{Z}$ elements;
  - single or multi-specie;
  - low atomic fractions;
  - very different thermodynamic conditions;

What does it mean in terms of simulations?

- Several methods developed for solid, liquid or plasma physics:
  - one-center (average-atom like): quantum, computationally inexpensive;
  - multicenter:
    - quantum Monte-Carlo: electronic many-body;
    - molecular-dynamics based on either classical, quantum or semi-classical description.
- Material constraints in the high-energy-density regime:
  - low and high $\mathcal{Z}$ elements;
  - single or multi-specie;
  - low atomic fractions;
  - very different thermodynamic conditions;

What does it mean in terms of simulations?

- Several methods developed for solid, liquid or plasma physics:
  - one-center (average-atom like): quantum, computationally inexpensive;
  - multicenter:
    - quantum Monte-Carlo: electronic many-body;
    - **molecular-dynamics** based on either classical, **quantum or semi-classical description**.
- Material constraints in the high-energy-density regime:
  - low and high $\mathcal{Z}$ elements;
  - single or multi-specie;
  - low atomic fractions;
  - very different thermodynamic conditions;

Contents

Outline

Adiabatic approximation & molecular dynamics

- Adiabatic approximation:
  - instantaneous response of electronic fluid;
  - T-dependent electronic screening of nuclear coulomb interactions;
- Nuclei described as classical particles;

$$\mathcal{L}\left(\{\boldsymbol{R}\},\{\boldsymbol{P}\}\right) = \frac{1}{2}\sum_{\ell}\frac{\boldsymbol{P}_{\ell}^{2}}{\mathcal{M}_{\ell}} - \sum_{\ell\neq\ell'}\frac{\mathcal{Z}_{\ell}\mathcal{Z}_{\ell'}}{\left|\boldsymbol{R}_{\ell}-\boldsymbol{R}_{\ell'}\right|} - F^{e}\left[\{\boldsymbol{R}\}\right]$$

- Euler-Lagrange equations in the DFT framework;

$$\min_{n(\boldsymbol{r})} F^{e}\left[n(\boldsymbol{r}),\{\boldsymbol{R}\}\right] \text{ and } \frac{\mathrm{d}\boldsymbol{P}_{\ell}}{\mathrm{d}t} = \sum_{\ell'\neq\ell}\mathcal{Z}_{\ell}\mathcal{Z}_{\ell'}\frac{\boldsymbol{R}_{\ell}-\boldsymbol{R}_{\ell'}}{\left|\boldsymbol{R}_{\ell}-\boldsymbol{R}_{\ell'}\right|^{3}} - \boldsymbol{\nabla}_{\boldsymbol{R}_{\ell}}F^{e}\left[\{\boldsymbol{R}\}\right]$$

- Basic idea of molecular dynamics:

Adiabatic approximation & molecular dynamics

- Adiabatic approximation:
  - instantaneous response of electronic fluid;
  - T-dependent electronic screening of nuclear coulomb interactions;
- Nuclei described as classical particles;

$$\mathcal{L}(\{\boldsymbol{R}\},\{\boldsymbol{P}\}) = \frac{1}{2}\sum_{\ell}\frac{\boldsymbol{P}_{\ell}^{2}}{\mathcal{M}_{\ell}} - \sum_{\ell\neq\ell'}\frac{\mathcal{Z}_{\ell}\mathcal{Z}_{\ell'}}{\left|\boldsymbol{R}_{\ell}-\boldsymbol{R}_{\ell'}\right|} - F^{e}\left[\{\boldsymbol{R}\}\right]$$

- Euler-Lagrange equations in the DFT framework;

$$\min_{n(\boldsymbol{r})} F^{e}\left[n(\boldsymbol{r}),\{\boldsymbol{R}\}\right] \text{ and } \frac{\mathrm{d}\boldsymbol{P}_{\ell}}{\mathrm{d}t} = \sum_{\ell'\neq\ell}\mathcal{Z}_{\ell}\mathcal{Z}_{\ell'}\frac{\boldsymbol{R}_{\ell}-\boldsymbol{R}_{\ell'}}{\left|\boldsymbol{R}_{\ell}-\boldsymbol{R}_{\ell'}\right|^{3}} - \boldsymbol{\nabla}_{\boldsymbol{R}_{\ell}}F^{e}\left[\{\boldsymbol{R}\}\right]$$

- Basic idea of molecular dynamics:

$$\{\boldsymbol{R}(t)\} \longrightarrow$$

Adiabatic approximation & molecular dynamics

- Adiabatic approximation:
  - instantaneous response of electronic fluid;
  - T-dependent electronic screening of nuclear coulomb interactions;
- Nuclei described as classical particles;

$$\mathcal{L}(\{\boldsymbol{R}\},\{\boldsymbol{P}\}) = \frac{1}{2}\sum_{\ell}\frac{\boldsymbol{P}_{\ell}^{2}}{\mathcal{M}_{\ell}} - \sum_{\ell\neq\ell'}\frac{\mathcal{Z}_{\ell}\mathcal{Z}_{\ell'}}{\left|\boldsymbol{R}_{\ell}-\boldsymbol{R}_{\ell'}\right|} - F^{e}\left[\{\boldsymbol{R}\}\right]$$

- Euler-Lagrange equations in the DFT framework;

$$\min_{n(\boldsymbol{r})} F^{e}\left[n(\boldsymbol{r}),\{\boldsymbol{R}\}\right] \text{ and } \frac{\mathrm{d}\boldsymbol{P}_{\ell}}{\mathrm{d}t} = \sum_{\ell'\neq\ell}\mathcal{Z}_{\ell}\mathcal{Z}_{\ell'}\frac{\boldsymbol{R}_{\ell}-\boldsymbol{R}_{\ell'}}{\left|\boldsymbol{R}_{\ell}-\boldsymbol{R}_{\ell'}\right|^{3}} - \boldsymbol{\nabla}_{\boldsymbol{R}_{\ell}}F^{e}\left[\{\boldsymbol{R}\}\right]$$

- Basic idea of molecular dynamics:

$$\{\boldsymbol{R}(t)\} \longrightarrow$$

$$\text{Coulomb potential } V(\{\boldsymbol{R}(t)\}) \longrightarrow$$

Adiabatic approximation & molecular dynamics

- Adiabatic approximation:
  - instantaneous response of electronic fluid;
  - T-dependent electronic screening of nuclear coulomb interactions;
- Nuclei described as classical particles;

$$\mathcal{L}\big(\{\,\boldsymbol{R}\,\},\{\,\boldsymbol{P}\,\}\big) = \frac{1}{2}\sum_{\ell}\frac{\boldsymbol{P}_{\ell}^{2}}{\mathcal{M}_{\ell}} - \sum_{\ell\neq\ell'}\frac{\mathcal{Z}_{\ell}\mathcal{Z}_{\ell'}}{\big|\boldsymbol{R}_{\ell}-\boldsymbol{R}_{\ell'}\big|} - F^{e}\big[\{\,\boldsymbol{R}\,\}\big]$$

- Euler-Lagrange equations in the DFT framework;

$$\min_{n(\boldsymbol{r})} F^{e}\big[n(\boldsymbol{r}),\{\,\boldsymbol{R}\,\}\big] \text{ and } \frac{\mathrm{d}\boldsymbol{P}_{\ell}}{\mathrm{d}t} = \sum_{\ell'\neq\ell}\mathcal{Z}_{\ell}\mathcal{Z}_{\ell'}\frac{\boldsymbol{R}_{\ell}-\boldsymbol{R}_{\ell'}}{\big|\boldsymbol{R}_{\ell}-\boldsymbol{R}_{\ell'}\big|^{3}} - \boldsymbol{\nabla}_{\boldsymbol{R}_{\ell}}F^{e}\big[\{\,\boldsymbol{R}\,\}\big]$$

- Basic idea of molecular dynamics:

$$\{\,\boldsymbol{R}(t)\,\} \longrightarrow$$

Coulomb potential $V(\{\,\boldsymbol{R}(t)\,\}) \longrightarrow$

Free energy minimization $F^{e}\big[n(\boldsymbol{r})\big] \longrightarrow$

Adiabatic approximation & molecular dynamics

- Adiabatic approximation:
  - instantaneous response of electronic fluid;
  - T-dependent electronic screening of nuclear coulomb interactions;
- Nuclei described as classical particles;

$$\mathcal{L}(\{\boldsymbol{R}\},\{\boldsymbol{P}\}) = \frac{1}{2}\sum_{\ell}\frac{\boldsymbol{P}_{\ell}^{2}}{\mathcal{M}_{\ell}} - \sum_{\ell\neq\ell'}\frac{\mathcal{Z}_{\ell}\mathcal{Z}_{\ell'}}{\left|\boldsymbol{R}_{\ell}-\boldsymbol{R}_{\ell'}\right|} - F^{e}\left[\{\boldsymbol{R}\}\right]$$

- Euler-Lagrange equations in the DFT framework;

$$\min_{n(\boldsymbol{r})} F^{e}\left[n(\boldsymbol{r}),\{\boldsymbol{R}\}\right] \text{ and } \frac{\mathrm{d}\boldsymbol{P}_{\ell}}{\mathrm{d}t} = \sum_{\ell'\neq\ell}\mathcal{Z}_{\ell}\mathcal{Z}_{\ell'}\frac{\boldsymbol{R}_{\ell}-\boldsymbol{R}_{\ell'}}{\left|\boldsymbol{R}_{\ell}-\boldsymbol{R}_{\ell'}\right|^{3}} - \boldsymbol{\nabla}_{\boldsymbol{R}_{\ell}}F^{e}\left[\{\boldsymbol{R}\}\right]$$

- Basic idea of molecular dynamics:

$$\{\boldsymbol{R}(t)\} \longrightarrow \text{Coulomb potential } V(\{\boldsymbol{R}(t)\}) \longrightarrow \text{Free energy minimization } F^{e}\left[n(\boldsymbol{r})\right] \longrightarrow \text{Newton's equations } \{\boldsymbol{R}(t+\Delta t)\}$$

Outline

Quantum treatment

- Independent particle model: the Kohn-Sham scheme;

$$F^e\big[n(\boldsymbol{r})\big] = \sum_\ell \left(f_\ell \int \mathrm{d}\boldsymbol{r} \left|\boldsymbol{\nabla}\psi_\ell\right|^2 - \frac{1}{\beta}\Big[f_\ell \ln f_\ell + (1-f_\ell)\ln(1-f_\ell)\Big]\right)$$
$$+ \frac{1}{2}\iint \mathrm{d}\boldsymbol{r}\,\mathrm{d}\boldsymbol{r}'\,\frac{n(\boldsymbol{r})n(\boldsymbol{r}')}{|\boldsymbol{r}-\boldsymbol{r}'|} - \sum_{\ell=1}^{\mathrm{N_a}} \mathcal{Z}_\ell \int \mathrm{d}\boldsymbol{r}\,\frac{n(\boldsymbol{r})}{|\boldsymbol{R}_\ell - \boldsymbol{r}|}$$
$$+ \int \mathrm{d}\boldsymbol{r}\,\varepsilon_{xc}\big[n(\boldsymbol{r})\big]$$

$$\min_{n(\boldsymbol{r})} F^e\big[n(\boldsymbol{r})\big] \Leftrightarrow \left[\frac{p^2}{2} + V^H[n] + V^{xc}[n] + V^{ext}\right]|\psi_\ell\rangle = \varepsilon_\ell\,|\psi_\ell\rangle$$

$$n(\boldsymbol{r}) = \sum_\ell \frac{2}{1+\exp\beta(\varepsilon_\ell - \mu)}\left|\psi_\ell(\boldsymbol{r})\right|^2$$

High-energy -density and semi-classical treatment

- Limitation in temperature and density:
  - Fermi-Dirac distribution: flat when $kT \sim \mu$;
  - pseudo-potential and delocalization: frozen-core-electron approximation down (Mazevet *et al* PRE 2007);
- Only one term involving quantum states in LDA

$$F^e\big[n(\boldsymbol{r})\big] = \frac{1}{2}\iint \mathrm{d}\boldsymbol{r}\,\mathrm{d}\boldsymbol{r}'\,\frac{n(\boldsymbol{r})n(\boldsymbol{r}')}{|\boldsymbol{r}-\boldsymbol{r}'|} - \sum_{\ell=1}^{\mathrm{N_a}} \mathcal{Z}_\ell \int \mathrm{d}\boldsymbol{r}\,\frac{n(\boldsymbol{r})}{|\boldsymbol{R}_\ell - \boldsymbol{r}|} + \int \mathrm{d}\boldsymbol{r}\,\varepsilon_{xc}\big[n(\boldsymbol{r})\big] + \sum_\ell \left(f_\ell \int \mathrm{d}\boldsymbol{r} \big|\boldsymbol{\nabla}\psi_\ell\big|^2 - \frac{1}{\beta}\Big[f_\ell \ln f_\ell + (1-f_\ell)\ln(1-f_\ell)\Big]\right)$$

$$n(\boldsymbol{r}) = \sum_\ell \frac{2}{1+\exp\beta(\varepsilon_\ell - \mu)}\big|\psi_\ell(\boldsymbol{r})\big|^2$$

High-energy -density and semi-classical treatment

- Limitation in temperature and density:
  - Fermi-Dirac distribution: flat when $kT \sim \mu$;
  - pseudo-potential and delocalization: frozen-core-electron approximation down (Mazevet *et al* PRE 2007);
- Only one term involving quantum states in LDA: let's get rid of it!
- Semi-classical expansion: first term in $\hbar$ (Brack, 2007);

$$
\begin{aligned}
F^{e}\big[n(\boldsymbol{r})\big] = \frac{1}{2}\iint \mathrm{d}\boldsymbol{r}\,\mathrm{d}\boldsymbol{r}'\,\frac{n(\boldsymbol{r})n(\boldsymbol{r}')}{|\boldsymbol{r}-\boldsymbol{r}'|} - \sum_{\ell=1}^{\mathrm{N_a}} \mathcal{Z}_\ell \int \mathrm{d}\boldsymbol{r}\,\frac{n(\boldsymbol{r})}{|\boldsymbol{R}_\ell - \boldsymbol{r}|} \\
+ \int \mathrm{d}\boldsymbol{r}\,\varepsilon_{xc}\big[n(\boldsymbol{r})\big] \\
+ \int \mathrm{d}\boldsymbol{r}\left(n(\boldsymbol{r})\,\Phi\big[n(\boldsymbol{r})\big] - \frac{2\sqrt{2}}{3\pi^2\beta^{\frac{3}{2}}} I_{\frac{3}{2}}\Big(\Phi\big[n(\boldsymbol{r})\big]\Big)\right)
\end{aligned}
$$

$$
n(\boldsymbol{r}) = \frac{\sqrt{2}}{\pi^2\beta^{\frac{3}{2}}} I_{\frac{3}{2}}\Big(\Phi\big[n(\boldsymbol{r})\big]\Big)
$$

High-energy -density and semi-classical treatment

- Limitation in temperature and density:
  - Fermi-Dirac distribution: flat when $kT \sim \mu$;
  - pseudo-potential and delocalization: frozen-core-electron approximation down (Mazevet *et al* PRE 2007);
- Only one term involving quantum states in LDA
- Semi-classical expansion: first term in $\hbar$ (Brack, 2007);

$$E^e\big[n(\boldsymbol{r})\big] = \frac{3^{\frac{5}{3}}\pi^{\frac{4}{3}}}{10}\int \mathrm{d}\boldsymbol{r}\, n(\boldsymbol{r})^{\frac{5}{3}} + \frac{1}{2}\iint \mathrm{d}\boldsymbol{r}\mathrm{d}\boldsymbol{r}'\,\frac{n(\boldsymbol{r})n(\boldsymbol{r}')}{|\boldsymbol{r}-\boldsymbol{r}'|} - \sum_{\ell=1}^{\mathrm{N_a}} \mathcal{Z}_\ell \int \mathrm{d}\boldsymbol{r}\,\frac{n(\boldsymbol{r})}{|\boldsymbol{R}_\ell - \boldsymbol{r}|}$$

Contents

Outline

Orbital Free Molecular Dynamics

- Microcanonical $(N, \Omega, \mathcal{E})$ and isokinetic $(N, \Omega, \mathcal{K})$ statistical ensembles:
  - isokinetic: time reversal algorithm (Minary *et al*, JCP 2003);
- Periodic boundary conditions:
  - Ewald sums for the nuclei;
  - FFT for the electronic fluid: parallel FFTW 2.1.5 and FFTe;
- Functionals:
  - free energy from non-zero temperature Thomas-Fermi and gradient correction (Perrot, PRA 1979);
  - LDA exchange-correlation;
- Minimization of the electronic free energy: constrained conjugate gradient algorithm;
  - algorithm specially dedicated to orbital free methods (Jiang *et al*, JCP 2004): fake orbital;

$$n(\boldsymbol{r}) = \psi(\boldsymbol{r})^2$$

Outline

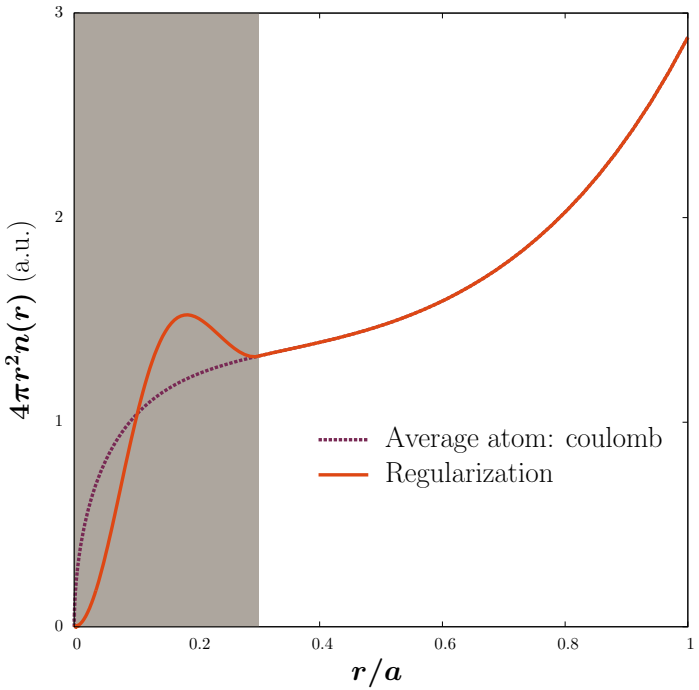
The regularization procedure

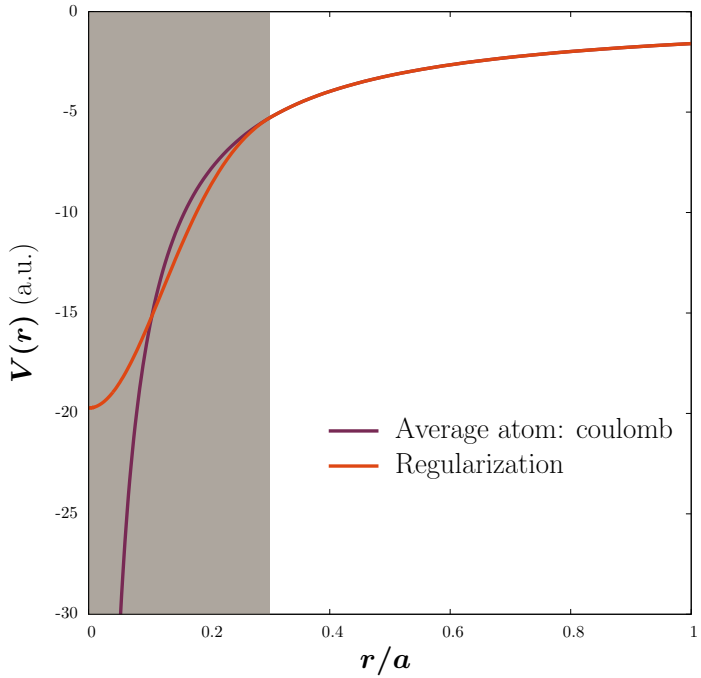
- r^{-1} divergence of the electron-*nucleus* potential: regularization (not a pseudo-potential);
- **all-electron calculation**!
- Numerical limitation: cut-off radius on the atomic scale, not on the nuclear one;

The regularization procedure

- Use of the OF average atom model (Feynman, Metropolis & Teller) and norm conserving pseudo-potential idea:
  - charge conservation is *mandatory*!
  - pressure and forces correctly evaluated (Gauss theorem);

$$\tilde{n}(r) = \begin{cases} \exp\left(a + br^2 + cr^4\right) \text{ if } r \leqslant r_c \\ n(r) \text{ if } r > r_c \end{cases} \quad = \alpha I_{\frac{1}{2}}\left[\beta\left(\mu - V_H(r) - \tilde{V}(r)\right)\right]$$





Outline

The parallelization paradigm

- Spread to efficiently work;
  - energy calculation, *i.e.* computation of integrals;

$$\Omega = \biguplus_{\ell} \Omega_\ell$$

$$\int_\Omega \mathrm{d}\boldsymbol{r}\, F[n(\boldsymbol{r})] = \sum_{\ell=1}^{N_p} \int_{\Omega_\ell} \mathrm{d}\boldsymbol{r}\, F[n(\boldsymbol{r})]$$

The parallelization paradigm

- Spread to efficiently work;
  - energy calculation, *i.e.* computation of integrals;
- Functionals' claim: "Let's be local!"
  - local functionals through Perrot's polynomial fit;

$$F^e[n] = \int \mathrm{d}\boldsymbol{r} \left(t\big[n(\boldsymbol{r})\big] - \beta^{-1}\sigma\big[n(\boldsymbol{r})\big] \right) + \int \mathrm{d}\boldsymbol{r}\, \varepsilon_{xc}\big[n(\boldsymbol{r})\big] + \int \mathrm{d}\boldsymbol{g}\, |n(\boldsymbol{g})|^2 \frac{4\pi}{\boldsymbol{g}^2} - \sum_{\ell=1}^{\mathrm{N_a}} \mathcal{Z}_\ell \int \mathrm{d}\boldsymbol{g}\, n(\boldsymbol{g}) \frac{4\pi}{\boldsymbol{g}^2} e^{-\imath \boldsymbol{g}\cdot\boldsymbol{R}_\ell}$$

The parallelization paradigm

- Spread to efficiently work;
  - energy calculation, *i.e.* computation of integrals;
- Functionals' claim: "Let's be local!"
  - local functionals through Perrot's polynomial fit;

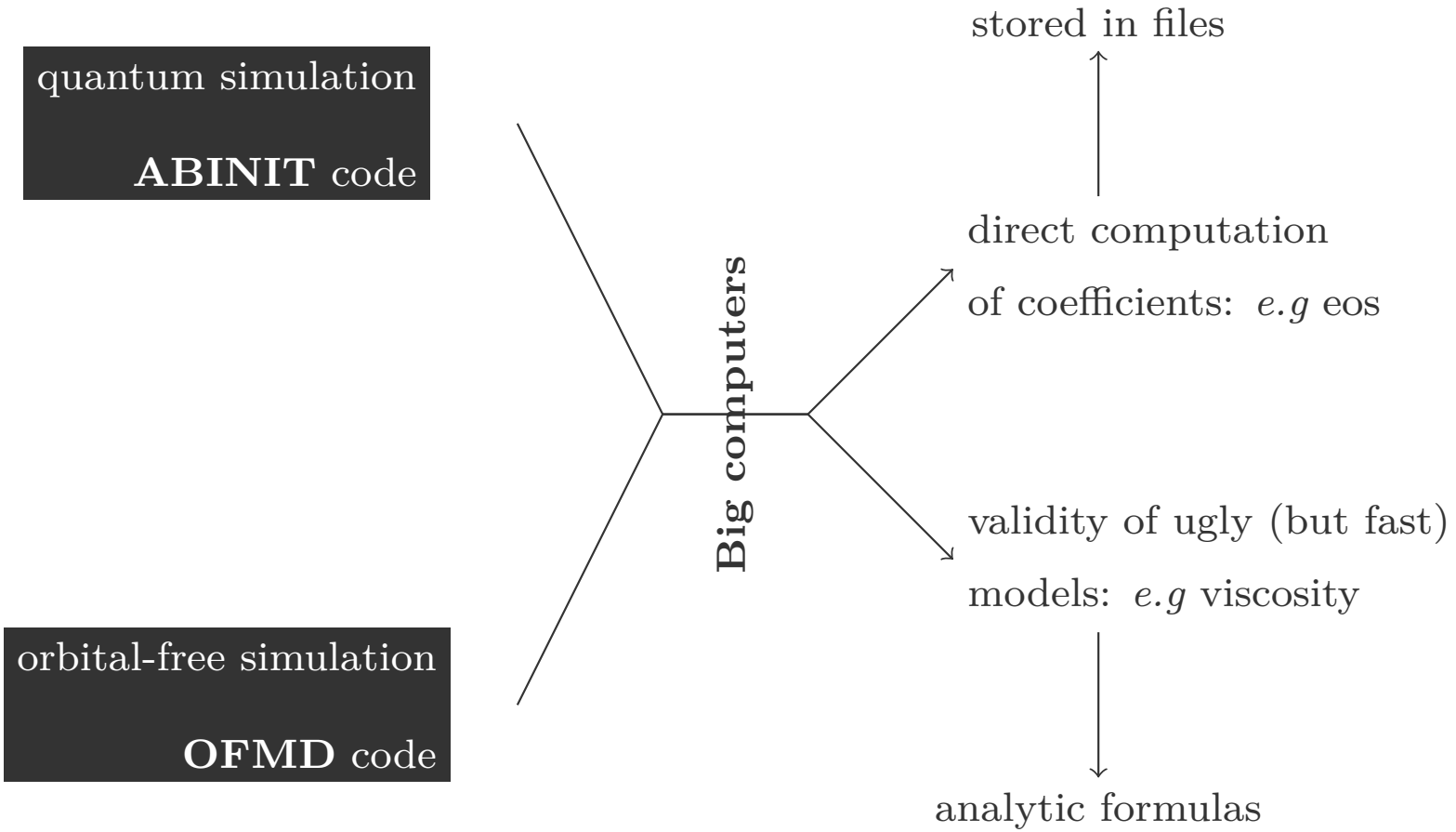
$$F^e[n] = \int \mathrm{d}\boldsymbol{r} \left(t[n(\boldsymbol{r})] - \beta^{-1} \sigma[n(\boldsymbol{r})] \right) + \int \mathrm{d}\boldsymbol{r}\, \varepsilon_{xc}[n(\boldsymbol{r})] + \int \mathrm{d}\boldsymbol{g}\, |n(\boldsymbol{g})|^2 \frac{4\pi}{\boldsymbol{g}^2} - \sum_{\ell=1}^{\mathrm{N_a}} \mathcal{Z}_\ell \int \mathrm{d}\boldsymbol{g}\, n(\boldsymbol{g}) \frac{4\pi}{\boldsymbol{g}^2} e^{-\imath \boldsymbol{g}\cdot\boldsymbol{R}_\ell}$$

Number of processors	$\langle \Delta t \rangle$ (s)	ratio (real/linear)
512	25.6	1/1
1024	13.5	1.9/2
2048	7.5	3.4/4
4096	5.6	4.6/8
8192	2.8	9.1/16

Average computation time per MD step, the electronic fluid being described on a 256^3 grid (performed by C. Ticknor on the Cielo machine at LANL).

Contents

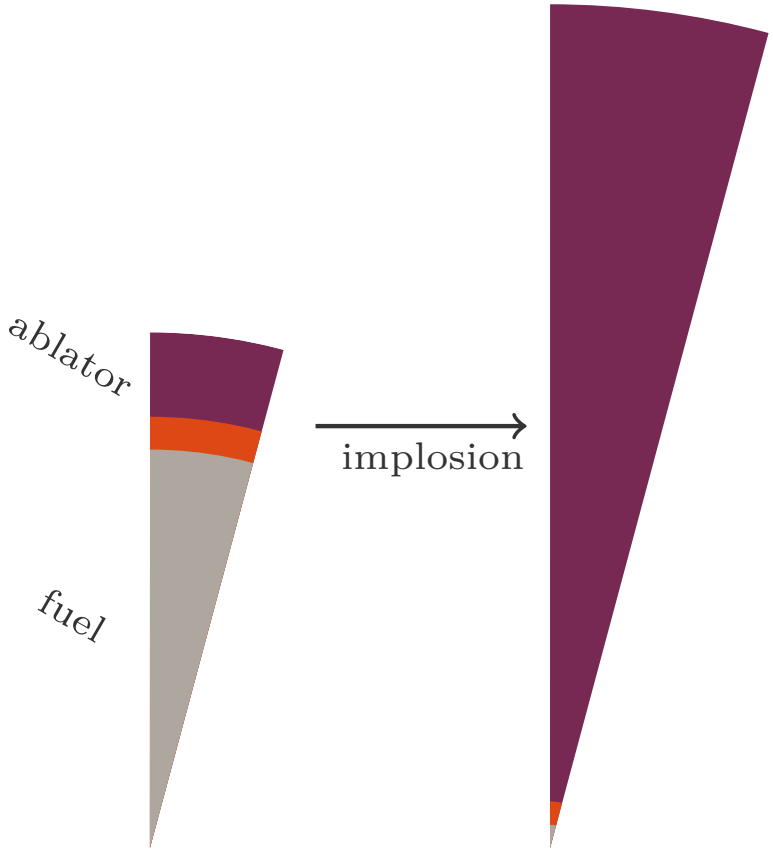
From microphysics to macrophysics

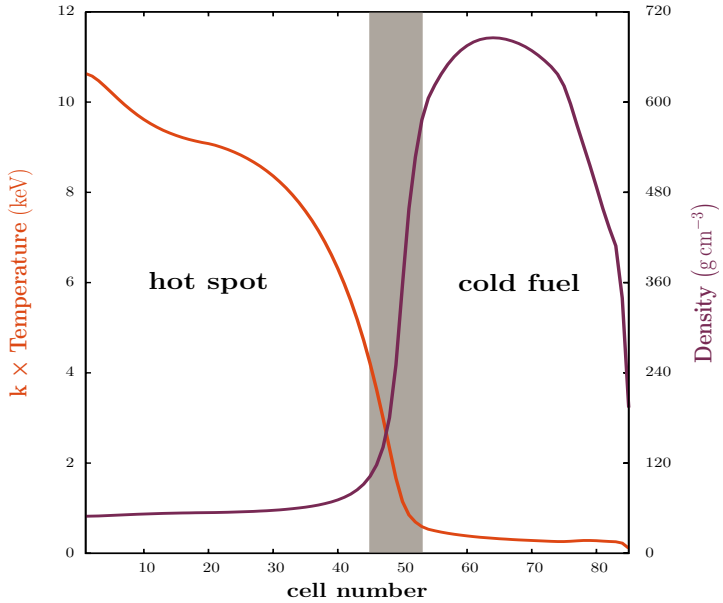


Outline

Physics of the hot-spot

- Ignition: no bulk combustion but propagation of a combustion wave;
- D and T burn for special ρ and kT, conditions reached with a specific design of two zones:
  - a central very hot ($kT \sim$ keV) mixture - "the hot spot": it's a match to launch a centrifugal combustion wave;
  - an outer very dense $\left(\rho \sim 500\,\mathrm{g\,cm^{-3}}\right)$ fuel: it releases most of the fusion energy!





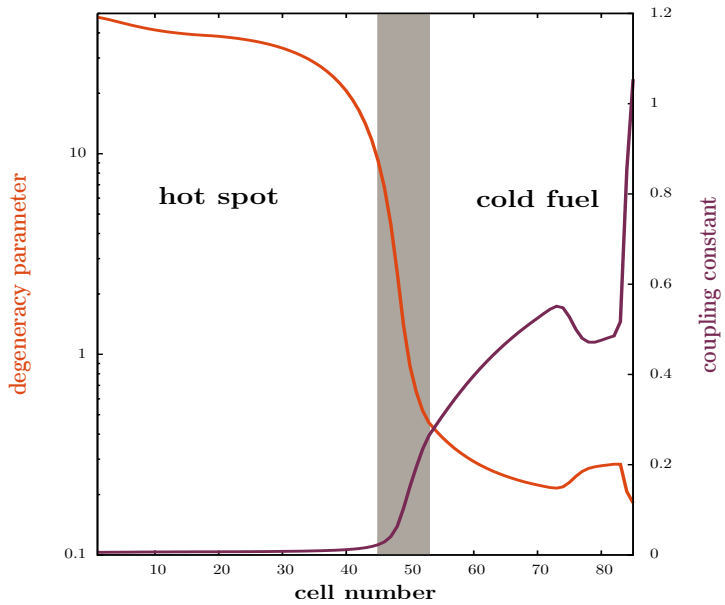
Physics of the hot-spot

- Ignition: no bulk combustion but propagation of a combustion wave;
- D and T burn for special ρ and kT, conditions reached with a specific design of two zones:
  - a central very hot ($kT \sim$ keV) mixture - "the hot spot": it's a match to launch a centrifugal combustion wave;
  - an outer very dense $\left(\rho \sim 500\,\mathrm{g\,cm^{-3}}\right)$ fuel: it releases most of the fusion energy!

- Wide range of plasma behavior:
  - from kinetic to partially degenerate and coupled;
  - conditions reached only in ICF!

$$\theta = \frac{kT}{\varepsilon_F} \sim 1$$

$$\Gamma = \frac{\langle U \rangle}{\langle K \rangle} = \frac{\mathcal{Z}^{\star 2}}{a\,kT} \sim 1$$



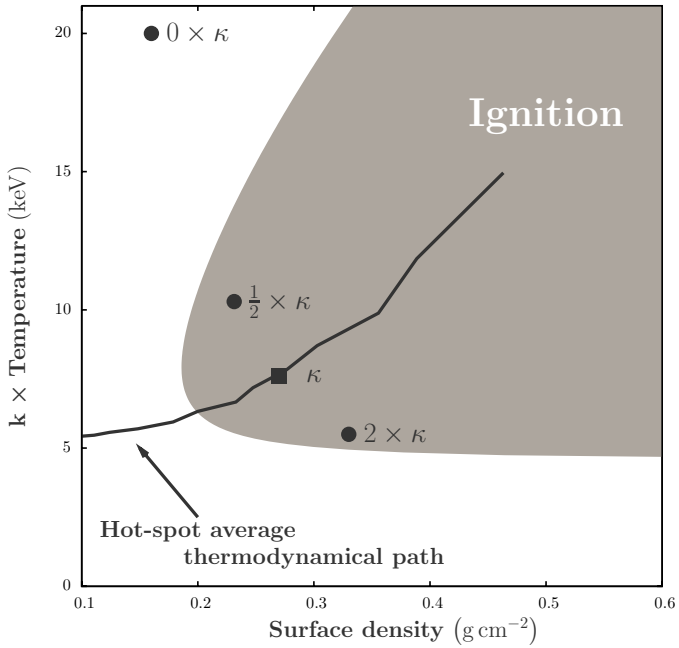
Physics of the hot-spot

- Ignition: no bulk combustion but propagation of a combustion wave;
- D and T burn for special ρ and kT, conditions reached with a specific design of two zones:
- What are the physical processes involved in ignition?
  - energy balance: (+) fusion energy (alpha particle deposition) and mechanical work, (-) radiative emission and electronic conduction;

Physics of the hot-spot

- Ignition: no bulk combustion but propagation of a combustion wave;
- D and T burn for special ρ and kT, conditions reached with a specific design of two zones:
- How confident are we in simulating such a process, *e.g.* thermal conduction?
  - very crude approximations of thermal conductivity;

- $\boldsymbol{\kappa}$ too low:
  - fusion energy confined in the center (no loss through conduction), cold fuel inert;
  - start but no propagation;
- $\boldsymbol{\kappa}$ too high:
  - too many energy loss, not enough heating of the "hot-spot";
  - no start at all.



Physics of the hot-spot

- Ignition: no bulk combustion but propagation of a combustion wave;
- D and T burn for special ρ and kT, conditions reached with a specific design of two zones:
- How confident are we in simulating such a process, *e.g.* thermal conduction?
- need for the verification of microscopic models in extreme conditions;

How can we compute thermal conductivity?

- Electronic transport coefficients: scattering, *i.e.* quantum states;
- Linear response formalism, the Kubo-Greenwood/Chester-Thellung formulation;

$$\kappa = \frac{1}{T}\left(\mathcal{L}_{22} - \frac{\mathcal{L}_{12} \times \mathcal{L}_{21}}{\mathcal{L}_{11}}\right)$$

$$\mathcal{L}_{ij} = (-1)^{i+j+1} \int \mathrm{d}\varepsilon \, \frac{\partial f(\varepsilon, \mu)}{\partial \varepsilon} (\varepsilon - \mu)^{i+j-2} s(\varepsilon)$$

$$s(\varepsilon) = \frac{1}{\Omega} \sum_{\ell,\ell'} \sum_{\alpha} \left| \langle \psi_{\ell'} | \, v_\alpha \, | \psi_\ell \rangle \right|^2 \delta(\varepsilon_{\ell'} - \varepsilon) \delta(\varepsilon_\ell - \varepsilon)$$

How can we compute thermal conductivity?

- Electronic transport coefficients: scattering, *i.e.* quantum states;
- Linear response formalism, the Kubo-Greenwood/Chester-Thellung formulation;
- We need orbitals...but at 500 eV!
  - computationally expensive!

$$\kappa = \frac{1}{T}\left(\mathcal{L}_{22} - \frac{\mathcal{L}_{12} \times \mathcal{L}_{21}}{\mathcal{L}_{11}}\right)$$

$$\mathcal{L}_{ij} = (-1)^{i+j+1} \int \mathrm{d}\varepsilon \, \frac{\partial f(\varepsilon, \mu)}{\partial \varepsilon} (\varepsilon - \mu)^{i+j-2} s(\varepsilon)$$

$$s(\varepsilon) = \frac{1}{\Omega} \sum_{\ell,\ell'} \sum_{\alpha} \left| \langle \psi_{\ell'} | \, v_\alpha \, | \psi_\ell \rangle \right|^2 \delta(\varepsilon_{\ell'} - \varepsilon) \delta(\varepsilon_\ell - \varepsilon)$$

- How can we do that?

How can we compute thermal conductivity?

- Electronic transport coefficients: scattering, *i.e.* quantum states;
- Linear response formalism, the Kubo-Greenwood/Chester-Thellung formulation;
- We need orbitals...but at 500 eV!
  - computationally expensive!

$$\kappa = \frac{1}{T}\left(\mathcal{L}_{22} - \frac{\mathcal{L}_{12} \times \mathcal{L}_{21}}{\mathcal{L}_{11}}\right)$$

$$\mathcal{L}_{ij} = (-1)^{i+j+1} \int \mathrm{d}\varepsilon \, \frac{\partial f(\varepsilon, \mu)}{\partial \varepsilon} (\varepsilon - \mu)^{i+j-2} s(\varepsilon)$$

$$s(\varepsilon) = \frac{1}{\Omega} \sum_{\ell,\ell'} \sum_{\alpha} \left| \langle \psi_{\ell'} | \, v_\alpha \, | \psi_\ell \rangle \right|^2 \delta(\varepsilon_{\ell'} - \varepsilon) \delta(\varepsilon_\ell - \varepsilon)$$

- How can we do that?
  - perform the MD with the orbital free (orbitals useless) scheme: nuclear structure;

How can we compute thermal conductivity?

- Electronic transport coefficients: scattering, *i.e.* quantum states;
- Linear response formalism, the Kubo-Greenwood/Chester-Thellung formulation;
- We need orbitals...but at 500 eV!
  - computationally expensive!

$$\kappa = \frac{1}{T}\left(\mathcal{L}_{22} - \frac{\mathcal{L}_{12} \times \mathcal{L}_{21}}{\mathcal{L}_{11}}\right)$$

$$\mathcal{L}_{ij} = (-1)^{i+j+1} \int \mathrm{d}\varepsilon \, \frac{\partial f(\varepsilon, \mu)}{\partial \varepsilon} (\varepsilon - \mu)^{i+j-2} s(\varepsilon)$$

$$s(\varepsilon) = \frac{1}{\Omega} \sum_{\ell,\ell'} \sum_{\alpha} \left| \langle \psi_{\ell'} | \, v_\alpha \, | \psi_\ell \rangle \right|^2 \delta(\varepsilon_{\ell'} - \varepsilon) \delta(\varepsilon_\ell - \varepsilon)$$

- How can we do that?
  - perform the MD with the orbital free (orbitals useless) scheme: nuclear structure;
  - extract a few nuclear configurations;

How can we compute thermal conductivity?

- Electronic transport coefficients: scattering, *i.e.* quantum states;
- Linear response formalism, the Kubo-Greenwood/Chester-Thellung formulation;
- We need orbitals...but at 500 eV!
  - computationally expensive!

$$\kappa = \frac{1}{T}\left(\mathcal{L}_{22} - \frac{\mathcal{L}_{12} \times \mathcal{L}_{21}}{\mathcal{L}_{11}}\right)$$

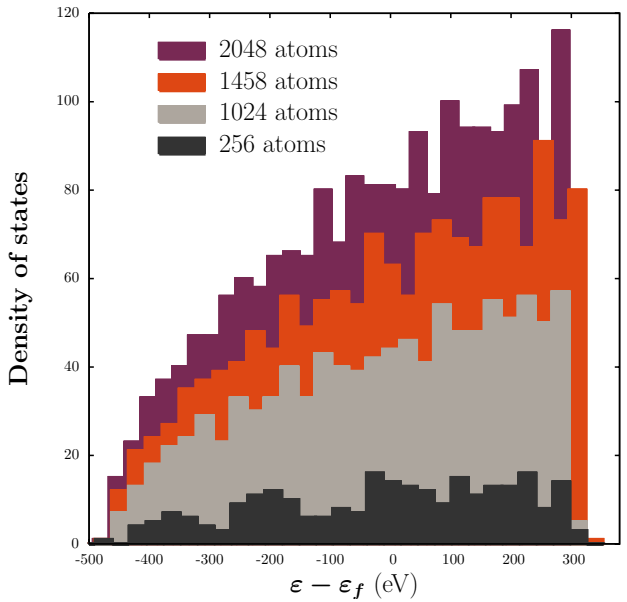
$$\mathcal{L}_{ij} = (-1)^{i+j+1} \int \mathrm{d}\varepsilon \, \frac{\partial f(\varepsilon, \mu)}{\partial \varepsilon} (\varepsilon - \mu)^{i+j-2} s(\varepsilon)$$

$$s(\varepsilon) = \frac{1}{\Omega} \sum_{\ell,\ell'} \sum_{\alpha} \left| \langle \psi_{\ell'} | \, v_\alpha \, | \psi_\ell \rangle \right|^2 \delta(\varepsilon_{\ell'} - \varepsilon) \delta(\varepsilon_\ell - \varepsilon)$$

- How can we do that?
  - perform the MD with the orbital free (orbitals useless) scheme: nuclear structure;
  - extract a few nuclear configurations;
  - compute the full quantum electronic structure on those configurations and average!

Comparison with models

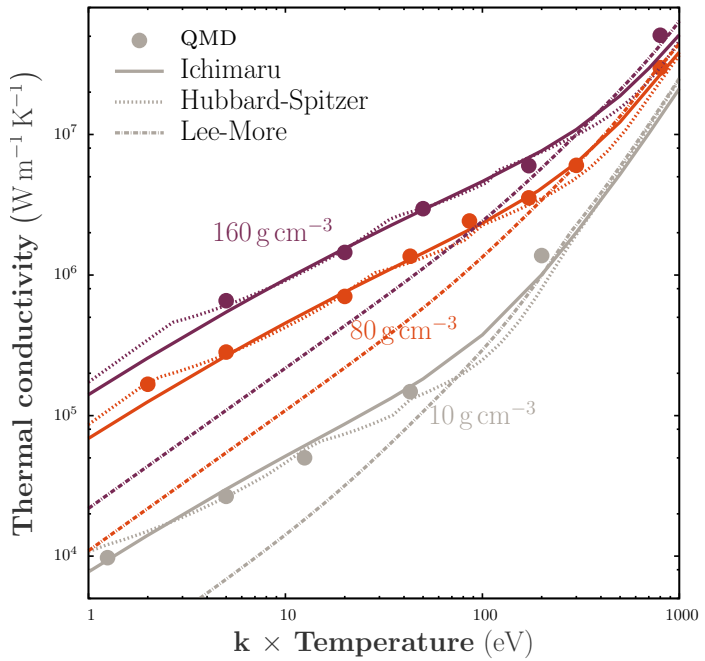
- Application to hydrogen at 10, 80 and 160 $\mathrm{g\,cm^{-3}}$ up to 800 eV!
- True challenge in terms of numerical simulation;
  - no pseudo-potential: purely coulombian;
  - competition between density and temperature effects: $kT \leqslant \varepsilon_f$;
  - electronic states are nearly plane waves;
  - at low temperatures need for 2000 atoms: numerical discretisation of the energy, sharp boundary of the Fermi distribution;



$\mathrm{dos}(\varepsilon) = \sum_\ell \delta(\varepsilon - \varepsilon_\ell) \propto \sqrt{\varepsilon}$ and $\Delta\varepsilon \propto \Omega^{-1}$ for a free fermion gas

Comparison with models

- Application to hydrogen at 10, 80 and 160 $\mathrm{g\,cm^{-3}}$ up to 800 eV!
- Comparison with models used in ICF or astrophysics codes:
  - **Hubbard-Spitzer**: kinetic Boltzmann theory at high kT and quantum perturbation theory at very low kT, interpolation inbetween;
  - **Ichimaru**: Ziman formulation plus a specific treatment of the dielectric function;
  - **Lee-More**: quantum Boltzmann equation with approximations on phase transitions;



Outline

Why do we need mixing rules?

- Material composed of elements ℓ of atomic mass $\mathcal{A}_\ell$ and atomic fraction x_ℓ, *e.g.* C_2H_3;
- EOS building based on using different models: average atom calculation, *i.e.* one element (QEOS, SESAME);
  - tabulated eos production;
- System involving a huge number of different materials: gravitational segregation in astrophysics;
  - on-line eos calculation;
- Molecular dynamics: rule of thumb for mixing laws;
  - put the right number of atoms in the right box, let the system live its life: direct calculation of mixture eos.

Isobaric-isothermal mixing rule

temperature matching

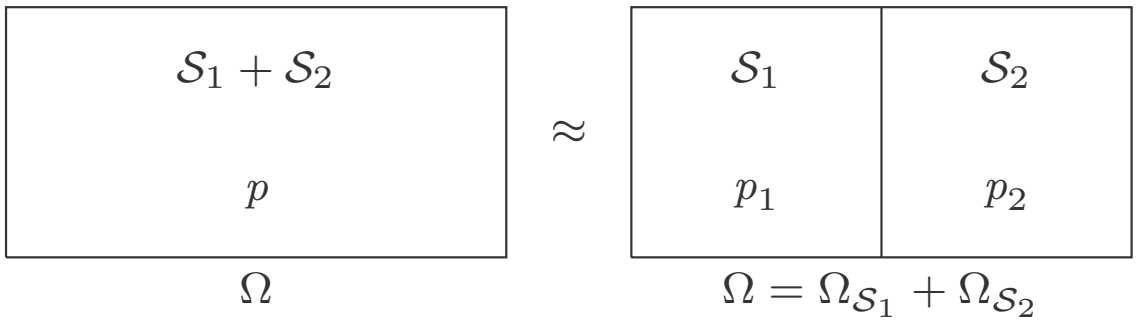
$$T_\ell = T, \, \forall \ell$$

additivity of partial volumes

$$\frac{1}{\rho} \sum_\ell x_\ell \mathcal{A}_\ell = \sum_\ell x_\ell \frac{\mathcal{A}_\ell}{\rho_\ell} \equiv \Omega = \sum_\ell \Omega_\ell$$

pressure matching

$$p_\ell(\rho_\ell, T) = p_{\ell'}(\rho_{\ell'}, T), \, \forall \ell \neq \ell'$$



Isobaric-isothermal mixing rule. The pressure matching between constituents provides the pressure of the mixture.

Isobaric-isothermal mixing rule

temperature matching

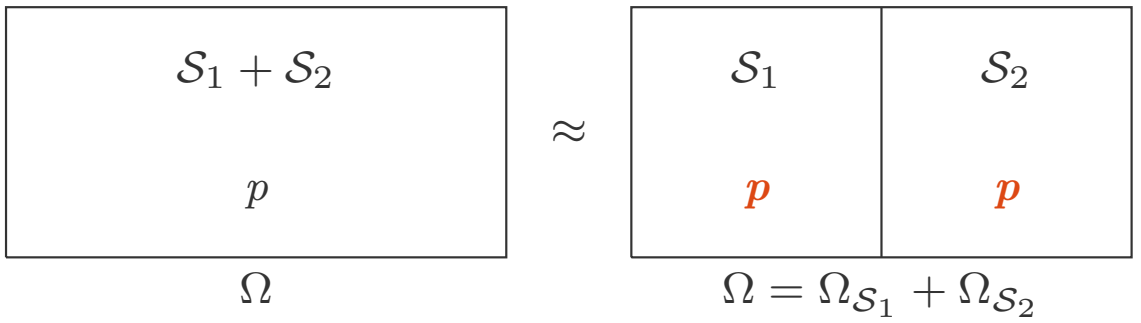
$$T_\ell = T, \, \forall \ell$$

additivity of partial volumes

$$\frac{1}{\rho} \sum_\ell x_\ell \mathcal{A}_\ell = \sum_\ell x_\ell \frac{\mathcal{A}_\ell}{\rho_\ell} \equiv \Omega = \sum_\ell \Omega_\ell$$

pressure matching

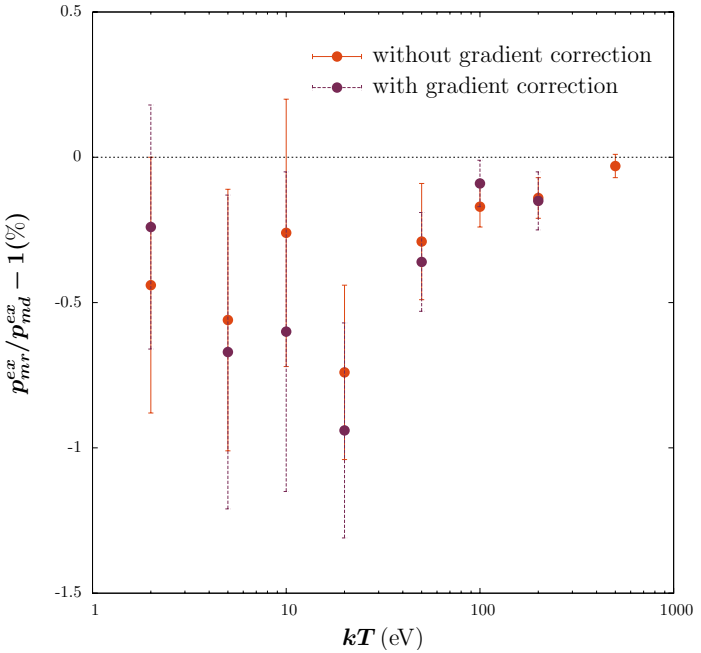
$$p_\ell(\rho_\ell, T) = p_{\ell'}(\rho_{\ell'}, T), \, \forall \ell \neq \ell' \Leftrightarrow F(\rho, T) = \sum_\ell x_\ell F_\ell(\rho_\ell, T)$$



Isobaric-isothermal mixing rule. The pressure matching between constituents provides the pressure of the mixture.

He/Fe example

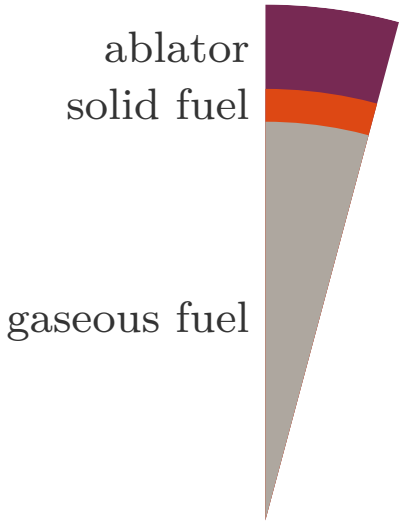
- Equimolar mixture of He and Fe at $10\,\mathrm{g\,cm^{-3}}$;
  - test of mixing law validity: agreement whatever the functional (Danel *et al*, PRE 79, 066408 (2009));



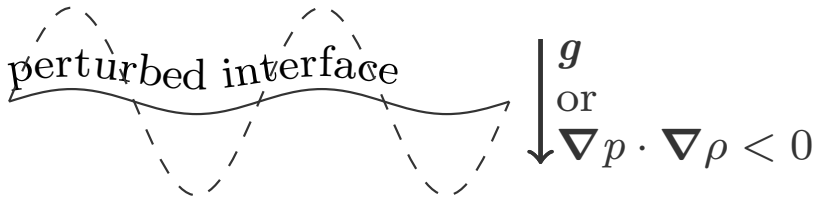
Outline

Hydrodynamic instability: the picture

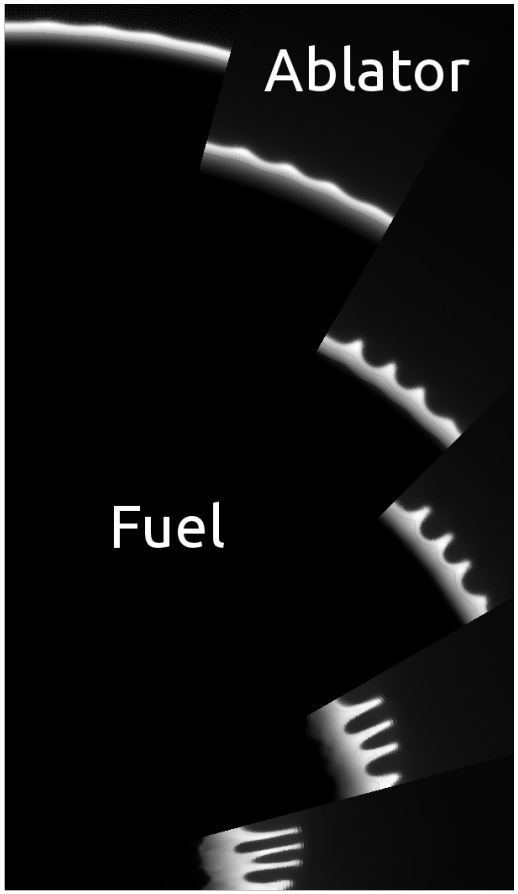
- Deleterious effects for ignition: hydrodynamic instabilities at material interfaces (fuel/ablator) or at the ablation front;
  - *e.g.* Rayleigh-Taylor instability: driven by ∇p and $\nabla \rho$;
  - precise knowledge of equation of state, thermal conductivity, viscosity or molecular diffusion;



heavy material (ablator)



light material (fuel, ablator)



Hydrodynamic instability: eos

- Hydrodynamic simulations: tabulated equation of state;
  - three contributions: cold curve, thermal electronic and thermal ionic (arbitrary);
  - often composed of several different models: average atom, Debye, *etc* but promising efforts (*e.g.* hydrogen by Kohn-Sham and PIMC for example);
  - approximations for mixtures: carbon and hydrogen for ablator;

PHYSICAL REVIEW B **83**, 094101 (2011)

Multiphase equation of state of hydrogen from *ab initio* calculations in the range 0.2 to 5 g/cc up to 10 eV

L. Caillabet, S. Mazevet, and P. Loubeyre
CEA, DAM, DIF, F-91287 Arpajon, France
(Received 10 September 2010; revised manuscript received 15 December 2010; published 2 March 2011)

PHYSICAL REVIEW B **84**, 224109 (2011)

First-principles equation-of-state table of deuterium for inertial confinement fusion applications

S. X. Hu (胡素兴),[1,*] B. Militzer,[2] V. N. Goncharov,[1] and S. Skupsky[1]
[1]*Laboratory for Laser Energetics, University of Rochester, Rochester, New York 14623, USA*
[2]*Department of Earth and Planetary Science and Department of Astronomy, University of California, Berkeley, California 94720, USA*
(Received 28 September 2011; revised manuscript received 21 November 2011; published 16 December 2011)

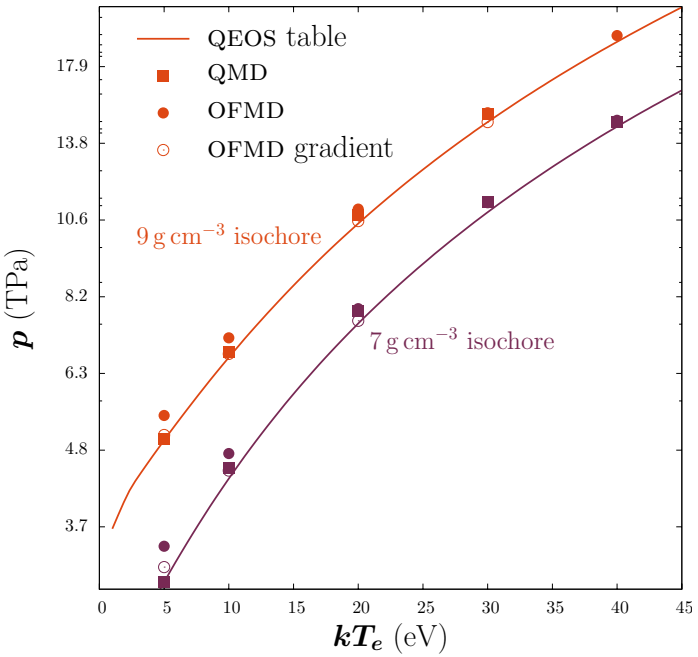
Ab initio calculations of the equation of state of hydrogen in a regime relevant for inertial fusion applications

Miguel A. Morales[a], Lorin X. Benedict[b,*], Daniel S. Clark[a], Eric Schwegler[a], Isaac Tamblyn[b], Stanimir A. Bonev[a,c], Alfredo A. Correa[a], Steven W. Haan[a]

[a] *Lawrence Livermore National Laboratory, Livermore, CA 94550, USA*
[b] *Molecular Foundry, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA*
[c] *Department of Physics, Dalhousie University, Halifax, Nova Scotia, Canada B3H*

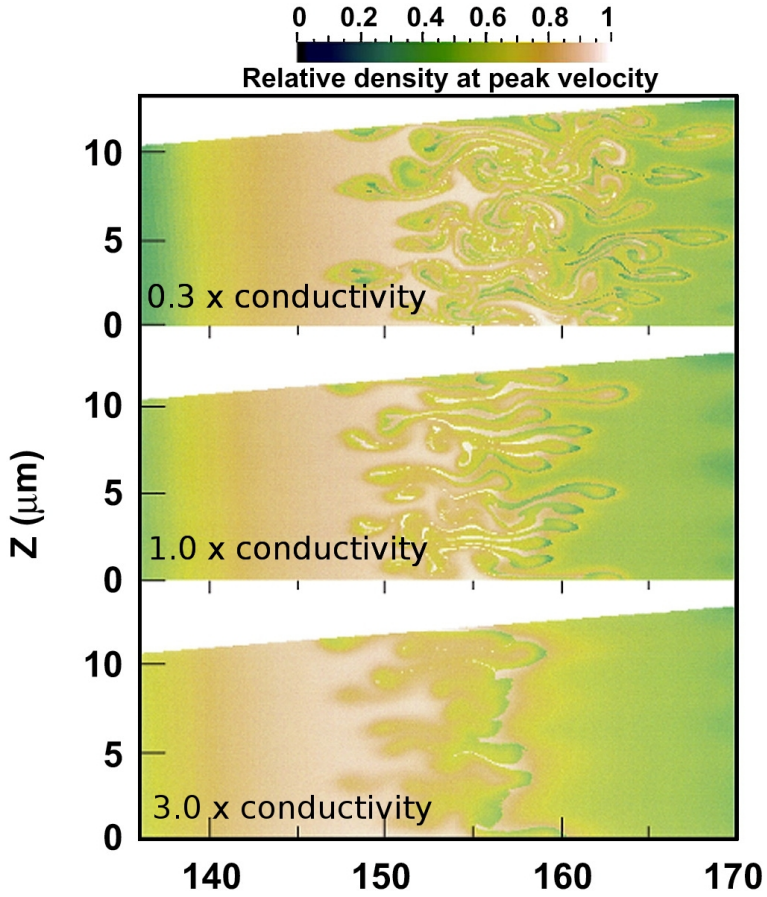
Hydrodynamic instability: eos

- Hydrodynamic simulations: tabulated equation of state;
- Need for comparisons with more fundamental calculations (quantum or orbital-free);
  - relevant conditions for instabilities: 7 and 9 g cm^{-3} isochores for C_2H_3;
  - OFMD features: 250 atoms propagated during 50000 time steps;



Hydrodynamic instability: thermal conductivity

- Thermal conduction: smoothing of temperature gradients but either stabilizing or destabilizing;
  - thermal conductivity: high impact on instability growth in hydrodynamics simulations (Hammel *et al*, HEDP 2010);



Hydrodynamic instability: thermal conductivity

- As for hot-spot: validity of approximate models but *new questions*;
  - models adapted for single element with a free parameter, the effective charge state;
  - ablator: multi-specie, different scattering centers;
    - several strategies: mix elements and compute conductivity (done in hydrodynamics code) or compute conductivities and mix them;

Hydrodynamic instability: thermal conductivity

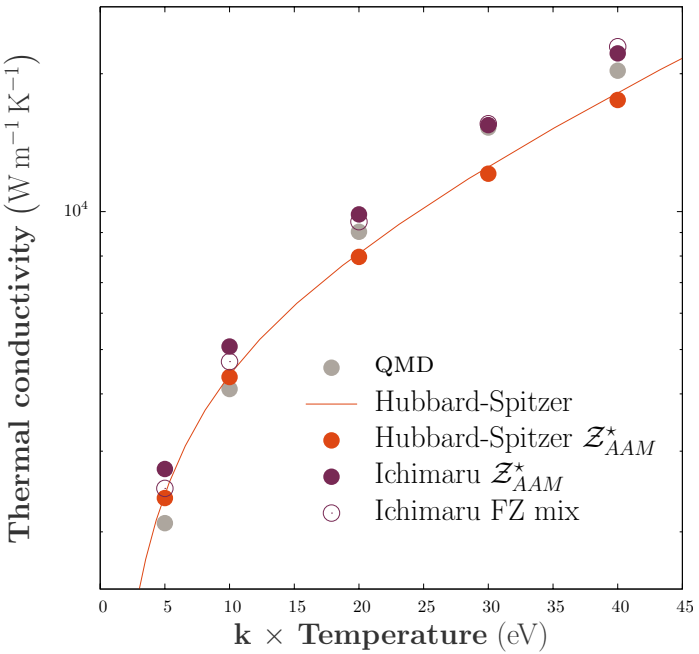
As for hot-spot: validity of approximate models but *new questions*;

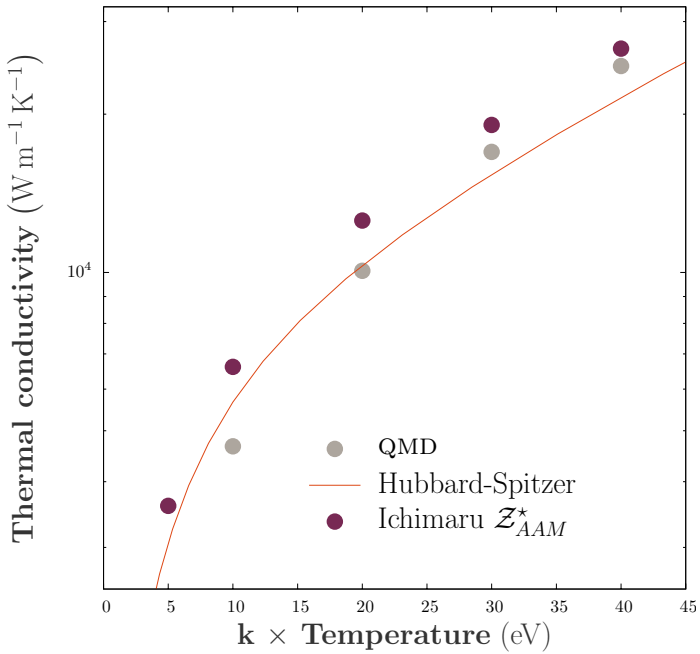
models adapted for single element with a free parameter, the effective charge state;

ablator: multi-specie, different scattering centers;

several strategies: *mix element and compute conductivity (done in hydrodynamics code)* or compute conductivities and mix them;

input parameters: average element ($\langle\mathcal{Z}\rangle$,$\langle\mathcal{A}\rangle$) or average ion ($\langle\mathcal{Z}^\star\rangle$,$\langle\mathcal{A}\rangle$);

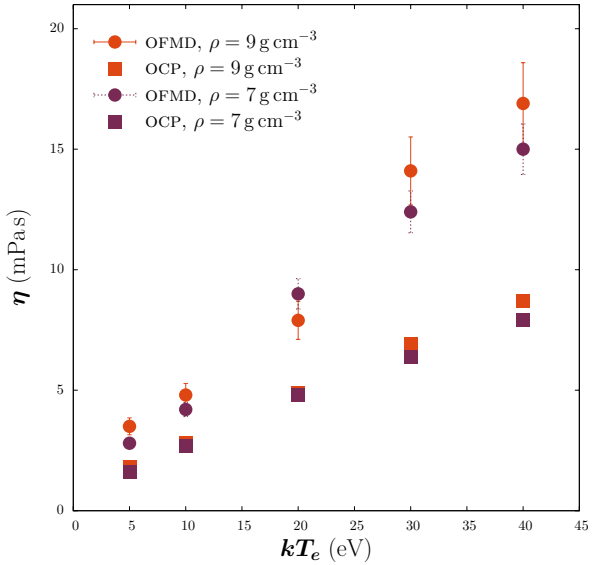




Hydrodynamic instability: viscosity

- Determination of the flow behavior: laminar, turbulent, *etc*;
  - Reynolds number: relevant length scale;
- Limitation of RT growth for small length scale (high modes);
- OFMD simulations: direct computation of viscosity for mixtures!
- Need for a quick evaluation: approximate parametrized model, *e.g* One component plasma with effective charge state;

$$\eta = \frac{1}{3\Omega kT} \sum_{\ell > \ell'} \int \left\langle \sigma^{\ell\ell'}(t) \sigma^{\ell\ell'}(0) \right\rangle \mathrm{d}t$$



Contents

Challenges?

- Equation of state: improvement of the moderate temperature part;
  - new orbital-free functionals: not $E_{xc}[n]$ but $T_s[n]$ and $\sigma_s[n]$;
- "Ionic" transport coefficients: increase system size, catch atomic traces, *e.g.* Si in plastic;
  - FFT, real space methods, minimization algorithms;
- "Electronic" transport coefficients:
  - semi-classical expansion of the Kubo-Greenwood formulation? Kubo?
  - time-dependent formulation of orbital-free DFT (Jérôme's talk, quantum hydrodynamics: Gao *et al* PRB 2008, Pittalis *et al* PRB 2011);
  - radiative coupling, out-of-equilibrium simulations (CIMARRON project, Graziani *et al* HEDP 2012);

Acknowledgments

V. Recoules (QMD), A. Decoster (Wisdom), G. Salin (QEOS), J. Clérouin, L. Kazandjian, J.-F. Danel and L. Masse (Instabilities)
Commissariat à l'Energie Atomique

C. Ticknor, J. Kress and L. Collins
Los Alamos National Laboratory

M. Desjarlais
Sandia National Labortory

More on orbital-free?

CECAM workshop, September 5^{th}-7^{th} in Paris