

# A Metastudy on Simulation Approaches for High Energy Density Matter

M. Marciante<sup>1</sup>, L.G. Stanton<sup>2</sup> and M.S. Murillo<sup>3</sup>

Transport Code Comparison Workshop – Sandia National Laboratories

*1 - Los Alamos National Laboratory*

*2 - Lawrence Livermore National Laboratory*

*3 - Michigan State University*

# **Outline**

1. Goals of this study
2. The Thomas-Fermi-Yukawa model
3. High Fidelity Physics Code Comparisons
4. Conclusion
5. Outlook

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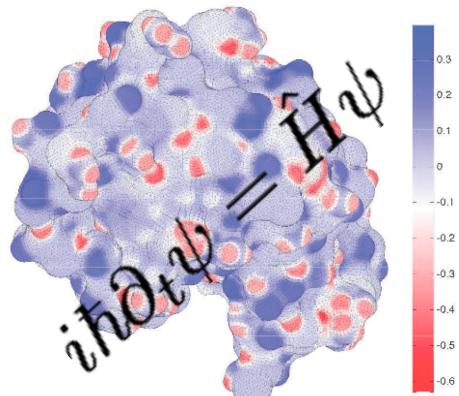
# Goals of this study

In the **High Energy Density Physics** world:

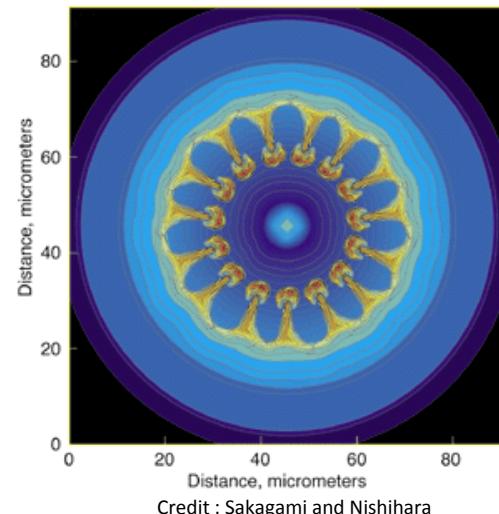
**High Fidelity Physics Codes (HFPC)** aim to provide an accurate description of the micro-scale physics to inform large-scale simulations:

- Computing equations of state
- Providing transport coefficients
- Providing relaxation rates

**HFPC** allow one to model complex many-body physics.



EOS  
Viscosity  
Thermal conductivity  
etc...



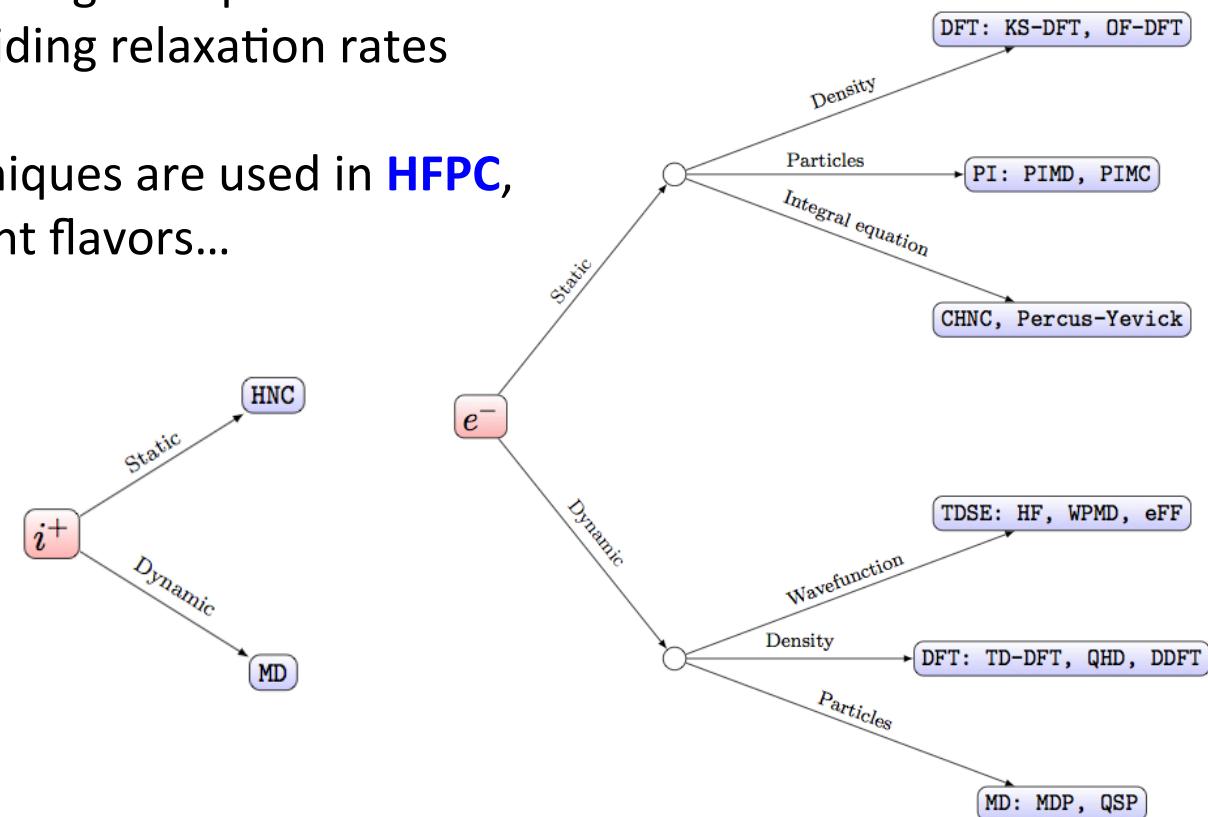
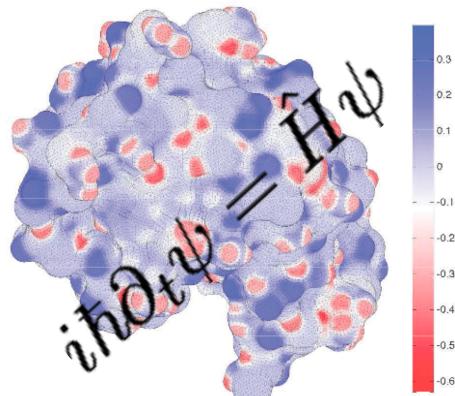
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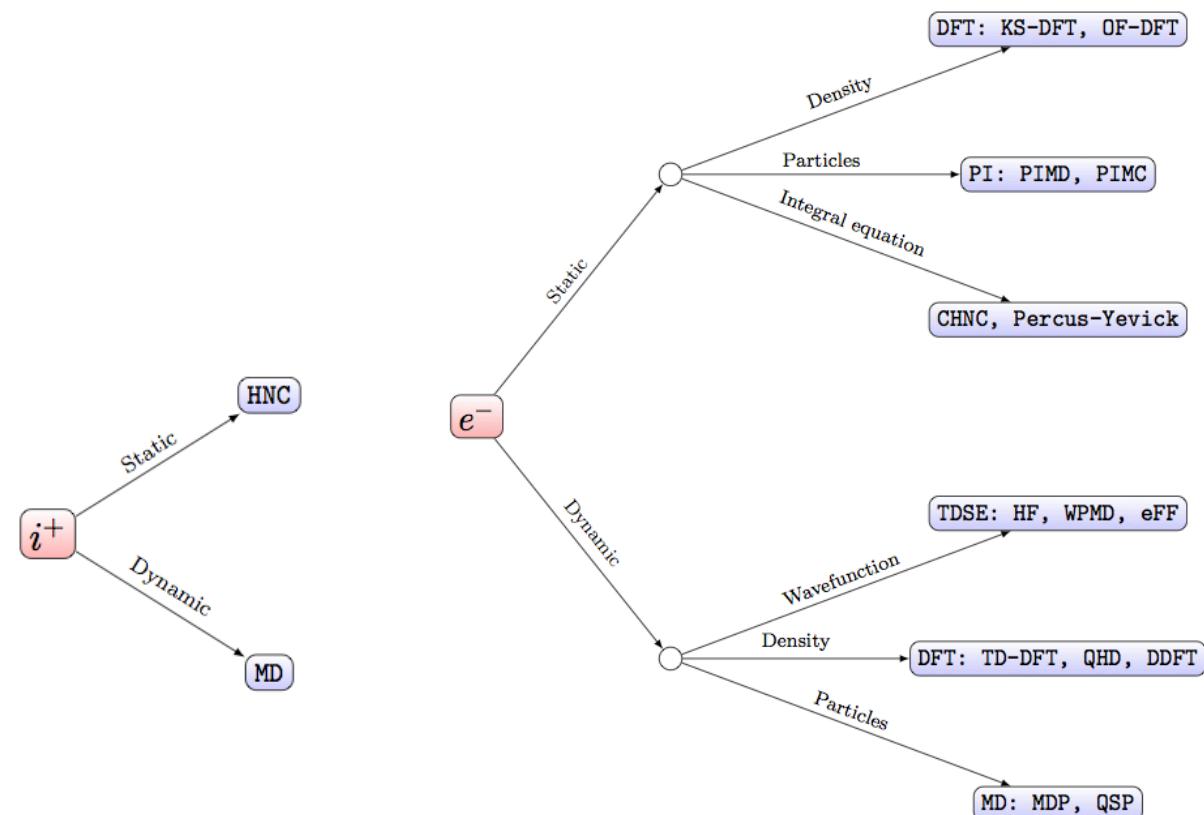
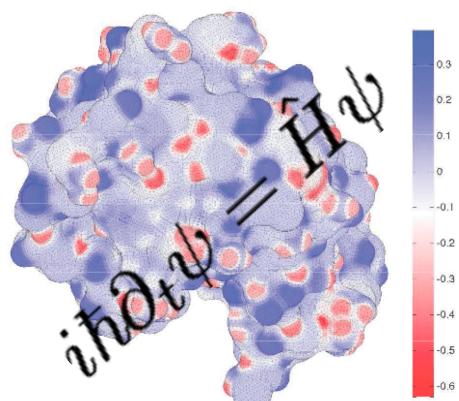
- Computing equations of state
- Providing transport coefficients
- Providing relaxation rates

A limited number of techniques are used in **HFPC**,  
but many different flavors...



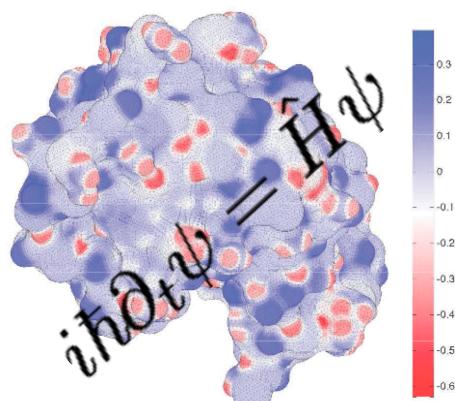
# Goals of this study

- Are we over-computing?  
Where should the computational effort be focused in the (T,n,Z)-space?

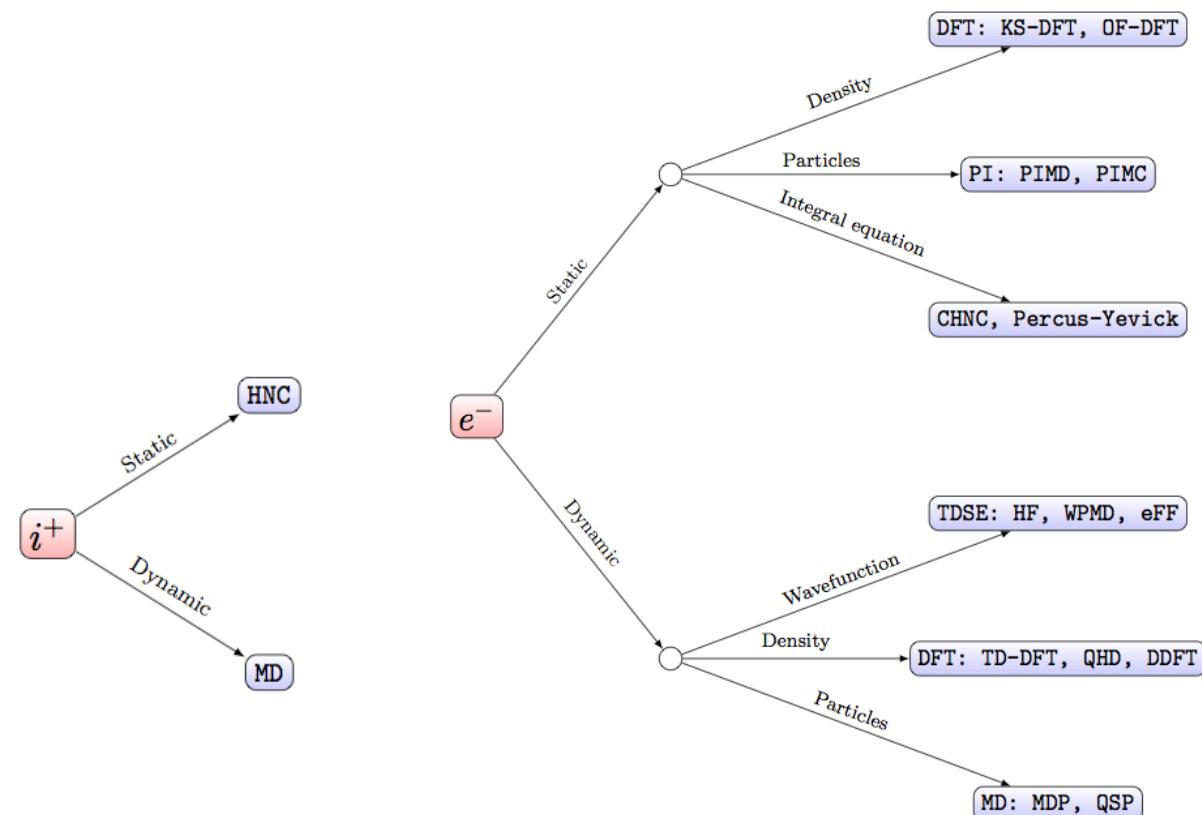


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- Are there methods more appropriate to access a given physics problem?  
larger length and time scales? (baro-diffusion, hydro-instabilities, shocks...)

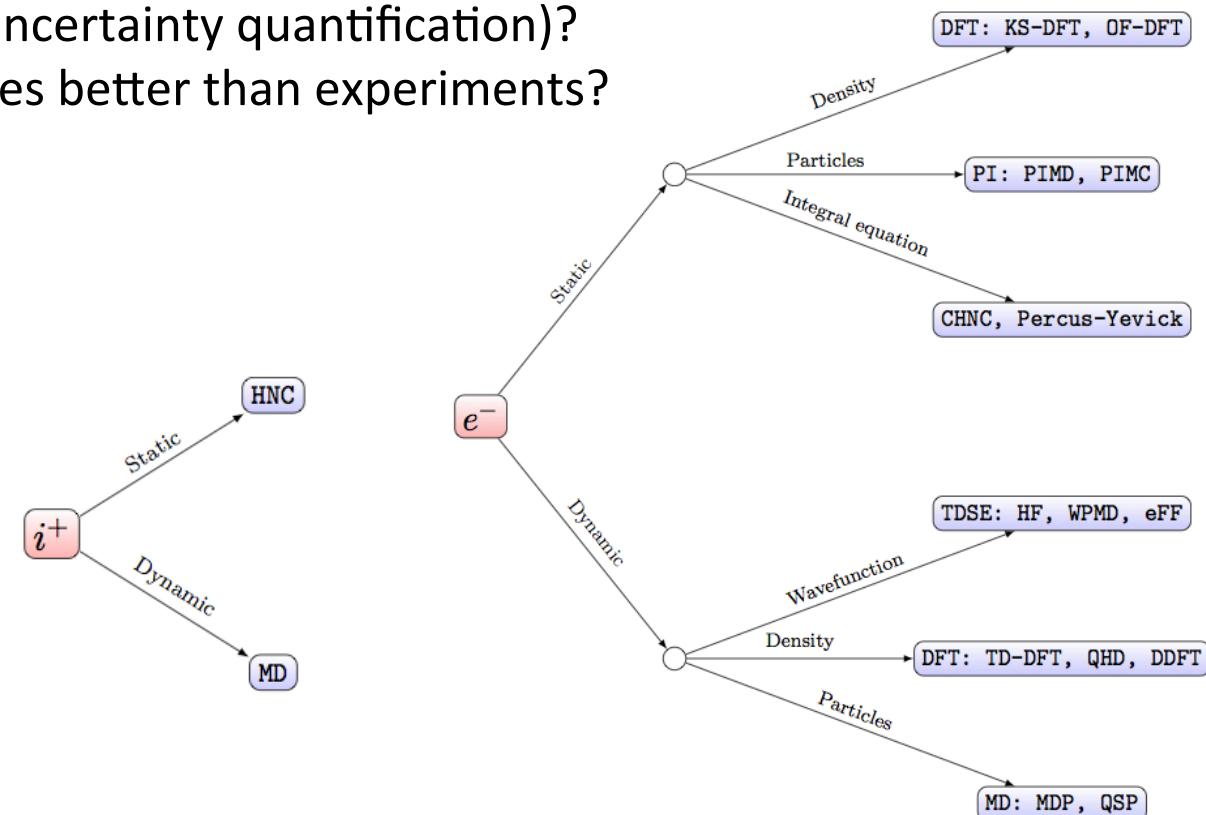
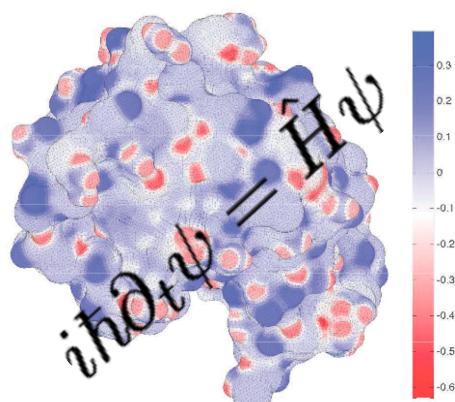


Credit : M. Knepley



# Goals of this study

- Are we over-computing?  
Where should the computational effort be focused in the (T,n,Z)-space?
- Are there methods more appropriate to access a given physics problem?  
larger length and time scales? (baro-diffusion, hydro-instabilities, shocks...)
- When is a cheap model sufficient for describing **HED** matter?  
better statistics (uncertainty quantification)?  
accessing quantities better than experiments?



# **Outline**

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## **2. The Thomas-Fermi-Yukawa model**

3. High Fidelity Physics Code Comparisons

4. Conclusion

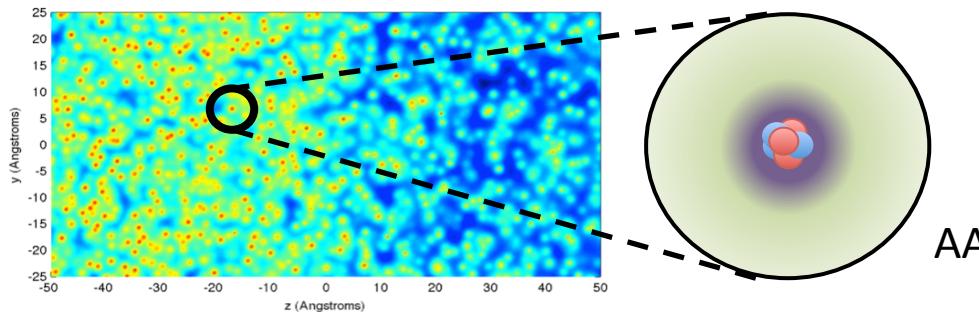
5. Outlook

# The Thomas-Fermi-Yukawa model

- 1) Average Atom
- 2) DFT treatment
- 3) Ionization state
- 4) Free e- lin. response

## 1) Average Atom approximation

The system is divided into spherical cells of radius  $a_i$ .



## 2) DFT is applied to the Average Atom

Grand potential for electrons: **Minimization determines the total electronic density**

Non-interacting e-  
Free energy functional

Hartree  
functional

Ionic  
potential

Chemical  
potential

Exchange-Correlation  
functional

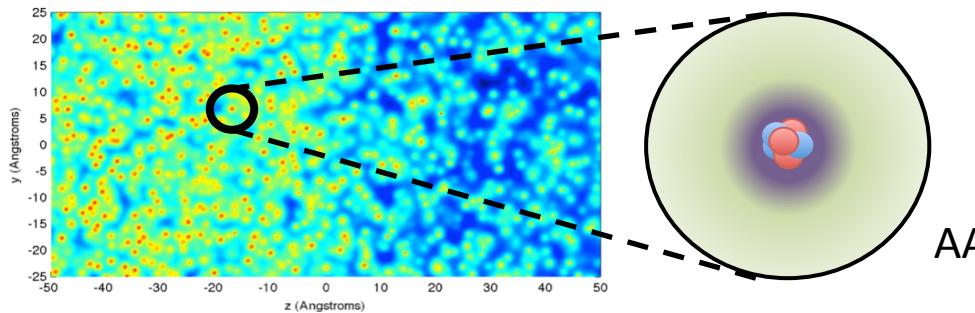
$$\Omega = \mathcal{T}[n] + \frac{1}{2} \int \int d\mathbf{r}' d\mathbf{r} \frac{n(\mathbf{r}) n(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} + \int d\mathbf{r} [v_{\text{ext}}(\mathbf{r}) - \mu] n(\mathbf{r}) + \mathcal{F}_{\text{xc}}[n]$$

# The Thomas-Fermi-Yukawa model

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## 1) Average Atom approximation

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Grand potential for electrons: **Minimization determines the total electronic density**

Non-interacting e- Free energy functional	Hartree functional	Ionic potential	Chemical potential
--	-----------------------	--------------------	-----------------------

$$\Omega = \mathcal{T}_{\text{TF}}[n] + \frac{1}{2} \int \int d\mathbf{r}' d\mathbf{r} \frac{n(\mathbf{r}) n(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} + \int d\mathbf{r} [v_{\text{ext}}(\mathbf{r}) - \mu] n(\mathbf{r}) + \cancel{\mathcal{F}_{\text{xc}}[n]}$$

For this simple model: The fully non-linear all-electron AA system is solved

- neglecting the exchange-correlation term
- using the Thomas-Fermi free energy functional:

$$\mathcal{T}_{\text{TF}}[n] = \frac{\sqrt{2}}{\pi^2 \beta^{5/2}} \int d\mathbf{r} \left[ \eta \mathcal{I}_{1/2}(\eta) - \frac{2}{3} \mathcal{I}_{3/2}(\eta) \right]$$

$$n(r) = \frac{\sqrt{2}}{\pi^2 \beta^{3/2}} \mathcal{I}_{1/2}[\eta(r)]$$

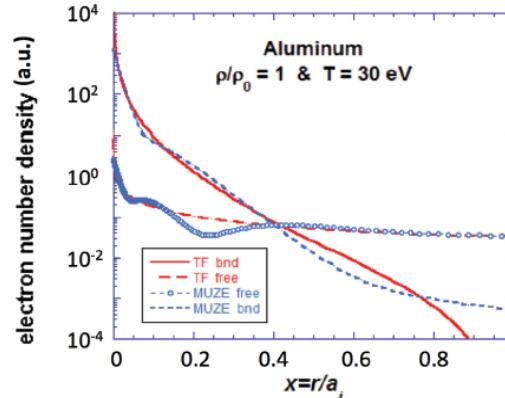
Fermi integral :

$$\mathcal{I}_p(\eta) = \int_0^\infty \frac{x^p}{1 + e^{x-\eta}} dx$$

# The Thomas-Fermi-Yukawa model

## 3) Mean ionization state

No distinction between free and bound electronic densities.



- 1) Average Atom
- 2) DFT treatment
- 3) Ionization state
- 4) Free e- lin. response

M.S. Murillo *et al.*  
PRE **87**, 063113 (2013)

- Symmetry consideration:  $\langle Z \rangle_{\text{TF}} = \frac{4}{3} \pi a_i^3 n_e(a_i)$

At the ion sphere radius, electrons don't feel any force : they are free.  
& Free electronic density is assumed to be homogeneous.

- Energy consideration:  $Z_{\text{TF}}^* = Z_{\text{nuc}} - \int 4\pi r^2 dr n_{\text{bd}}^{\text{TF}}(r)$

Negative energy contribution to the Fermi integral:

$$n_{\text{bd}}^{\text{TF}}(r) \propto \int_0^{-\frac{eU}{T}} dy \frac{y^{1/2}}{1 + \exp(y - \varepsilon(r))}$$

We chose for this study the first definition (symmetry consideration).  
The bound electronic density is collapsed to a point at the nucleus position.

# The Thomas-Fermi-Yukawa model

3) Mean ionization state       $\langle Z \rangle_{\text{TF}} = \frac{4}{3} \pi a_i^3 n_e(a_i)$

- 1) Average Atom
- 2) DFT treatment
- 3) Ionization state
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4) Free e- linear response       $n(r) = \frac{\sqrt{2}}{\pi^2 \beta^{3/2}} \mathcal{I}_{1/2}[\eta(r)]$

$$\mathcal{I}_p(\eta) = \int_0^\infty \frac{x^p}{1 + e^{x-\eta}} dx$$

$$\partial_\eta \mathcal{I}_p = p \mathcal{I}_{p-1}$$

$$\eta = \mu + \beta \langle Z \rangle e \Phi$$

$$n(\eta) = n(\mu) + \beta \langle Z \rangle e \Phi \frac{\sqrt{2}}{\pi^2 \beta^{3/2}} \partial_\eta \mathcal{I}_{1/2|\mu} + \mathcal{O}((\beta \langle Z \rangle e \Phi)^2)$$

Linearization of the Thomas-Fermi electron density response to the potential

$$\approx n(\mu) + \beta \langle Z \rangle e \Phi \frac{\sqrt{2}}{\pi^2 \beta^{3/2}} \frac{1}{2} \mathcal{I}_{-1/2|\mu}$$

Poisson equation:      
$$\Delta \Phi = -4\pi e \left\{ \langle Z \rangle_{\text{TF}} \delta(\vec{r}) - n_0 - \frac{\lambda_{\text{TF}}^{-2}}{4\pi e} \Phi \right\}$$

**Thomas – Fermi – Yukawa potential:**      
$$\Phi(r) = \frac{e \langle Z \rangle_{\text{TF}}}{r} e^{-r/\lambda_{\text{TF}}}$$

In practice for this study we used the interpolation:

$$E_F = \frac{\hbar^2 (3\pi^2 n_e)^{2/3}}{2m_e}$$

$$\lambda_{\text{TF}}^{-2} = \frac{4\pi e^2 \langle Z \rangle n_i}{\sqrt{T^2 + \left(\frac{2}{3} E_F\right)^2}}$$

Going beyond Thomas - Fermi:

Exact Gradient-corrected Screening (EGS)  
Stanton-Murillo PRE **91**, 033104 (2015)

# **Outline**

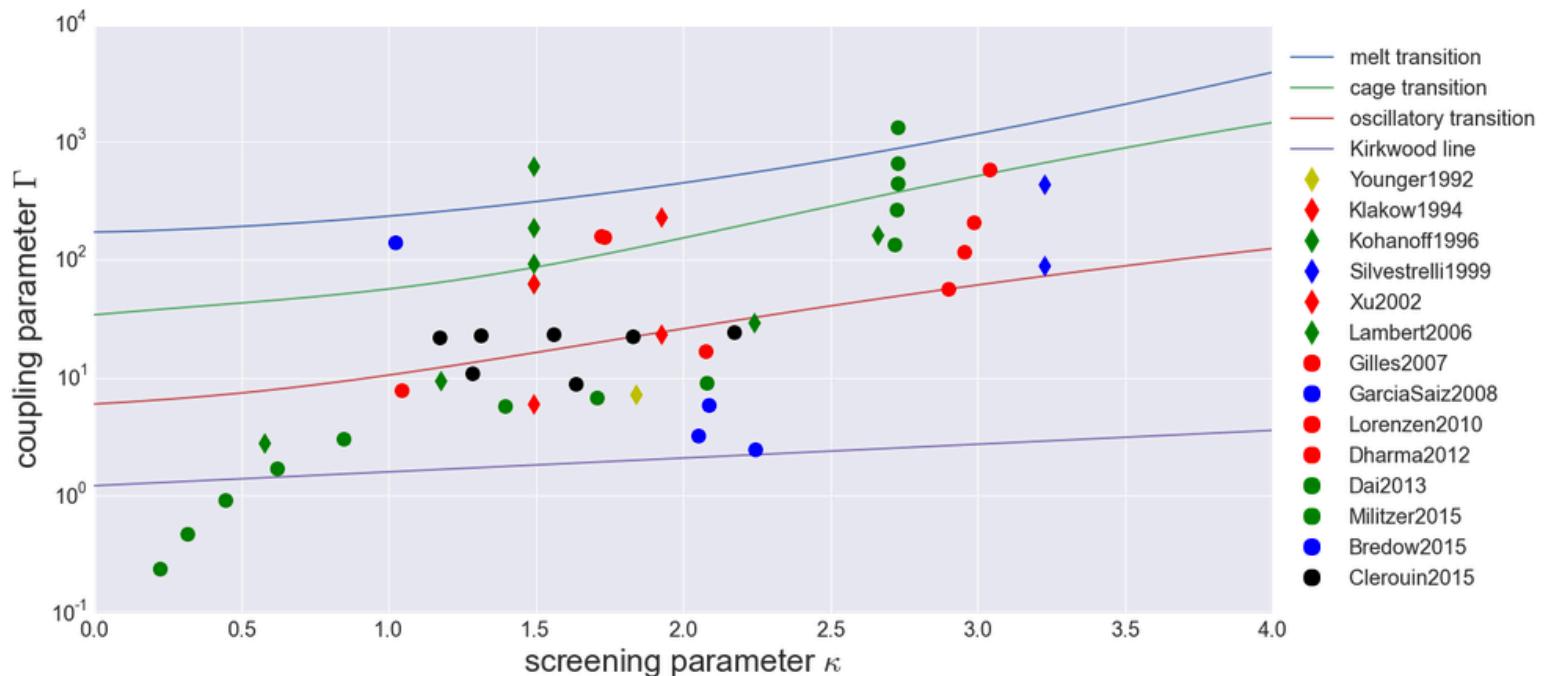
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# High Fidelity Physics Codes comparisons

$$\Gamma = \frac{\langle Z \rangle_{\text{TF}}^2 e^2}{a_i k_{\text{B}} T}$$
$$\kappa = a_i / \lambda_{\text{TF}}$$

We compared the **TFY** model to other **HFPC** results.

Summary of the TF-plasma states currently into the database.



In the following, lengths are in  $a_i$  units, times in  $\omega_p^{-1}$  units.

## Many-atom screening effects on diffusion in dense helium

Stephen M. Younger

*Los Alamos National Laboratory, Los Alamos, New Mexico 87545*

(Received 27 January 1992)

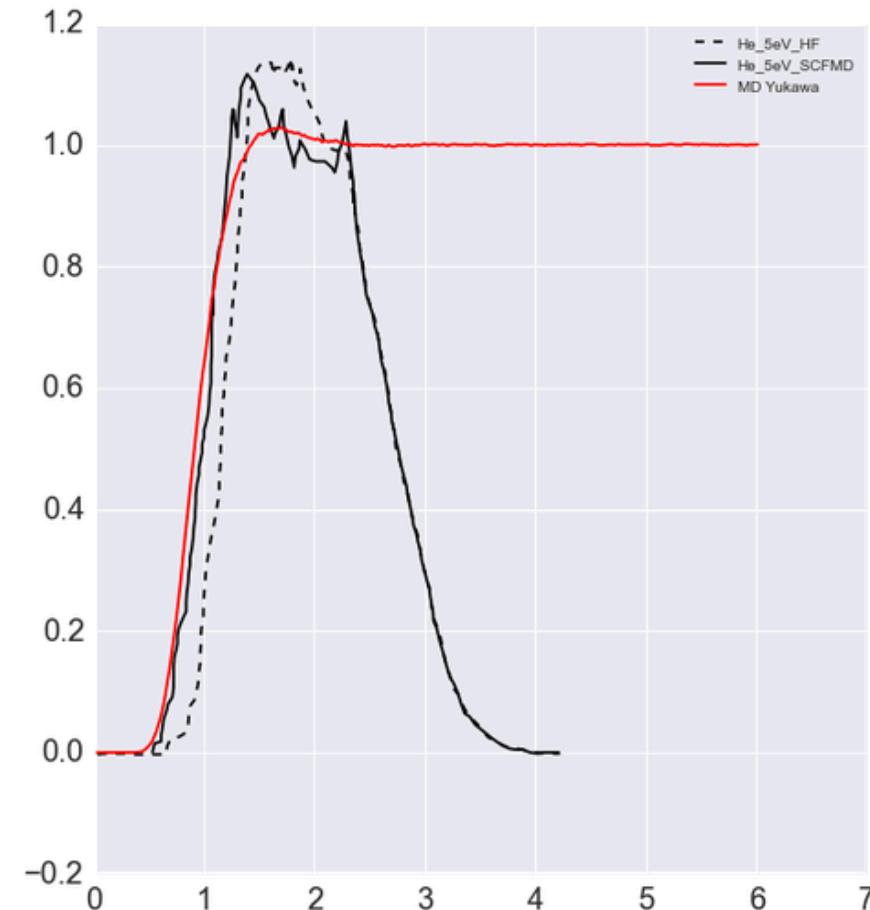
Helium, 5g/cc, 5eV

KS-DFT: SCFMD, 23 atoms

TF  $\langle Z \rangle / Z = 0.655$ 

$$\kappa = 1.84$$

$$\Gamma = 7.2$$



## Hydrogen under extreme conditions

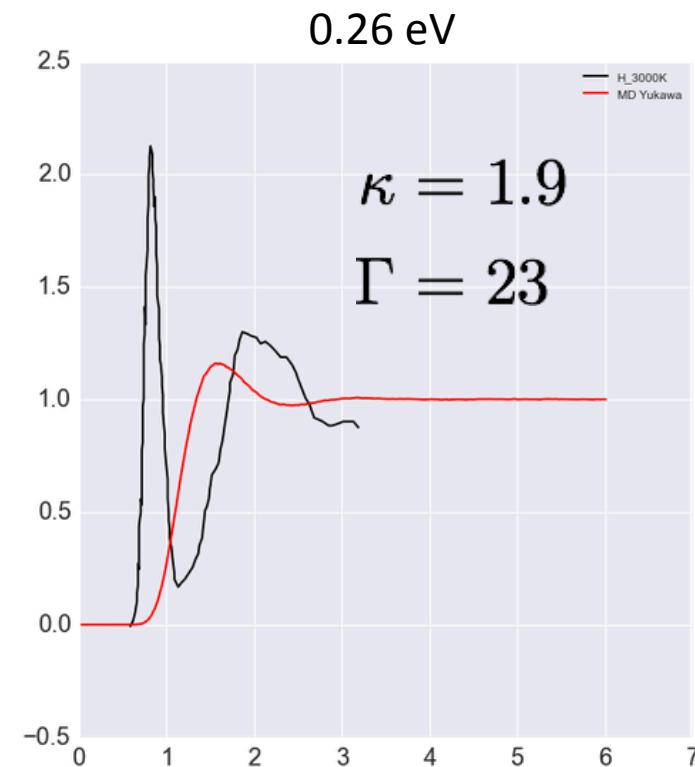
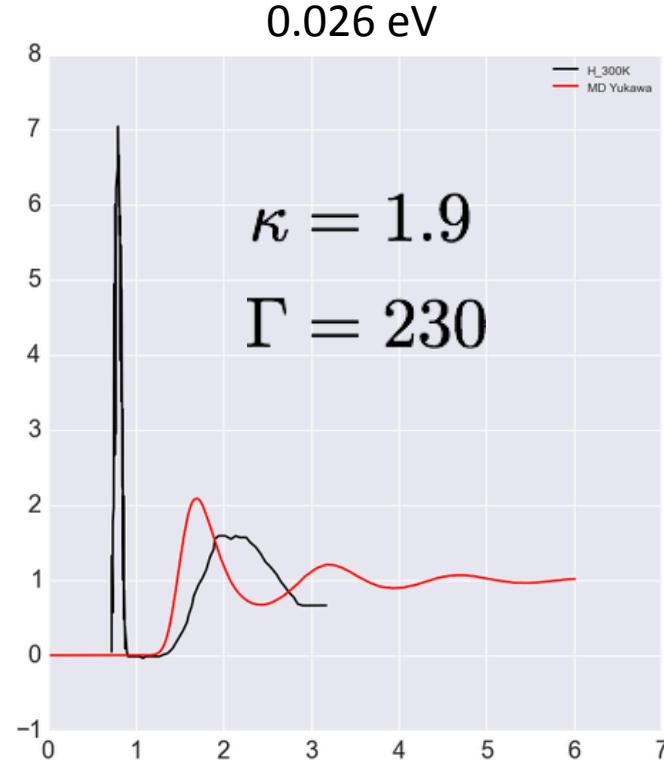
D. Klakow, C. Toepffer, P.-G. Reinhard

Institut für Theoretische Physik II, Staudtstrasse 7, Universität Erlangen-Nürnberg, D-91058 Erlangen, Germany

Hydrogen, 0.48g/cc

WPMD: 256 p+, 256 e-

TF  $\langle Z \rangle / Z = 0.62$



**Statistical properties of the dense hydrogen plasma:  
An *ab initio* molecular dynamics investigation**

Jorge Kohanoff<sup>1,2</sup> and Jean-Pierre Hansen<sup>2</sup>

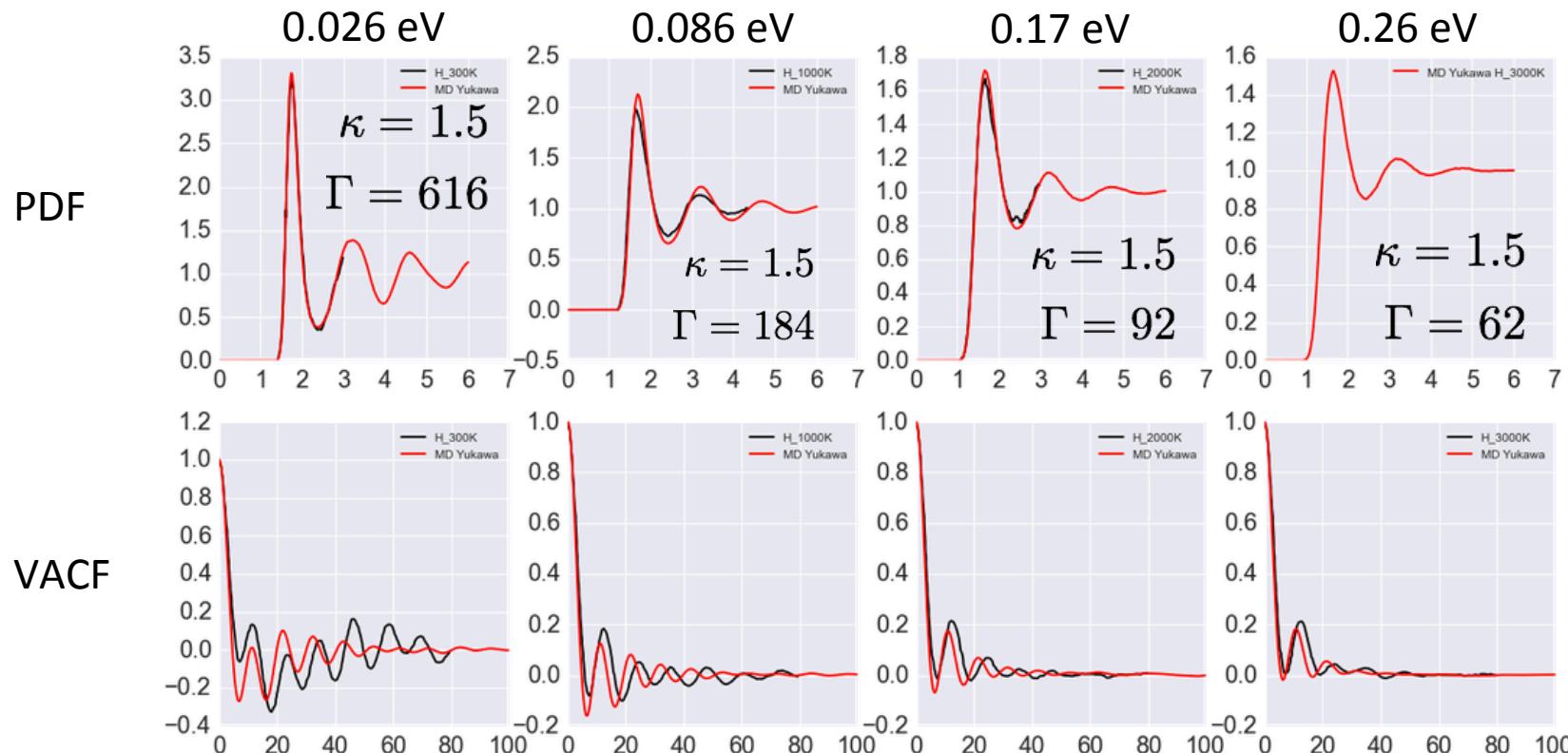
<sup>1</sup>*International Centre for Theoretical Physics, Strada Costiera 11, I-34014 Trieste, Italy*

<sup>2</sup>*Laboratoire de Physique, URA 1325 du CNRS, Ecole Normale Supérieure de Lyon, F-69364 Lyon Cedex 07, France*

Hydrogen, 71 g/cc

KS-DFT: CPMD from 54 to 162 atoms

TF  $\langle Z \rangle / Z = 0.77$



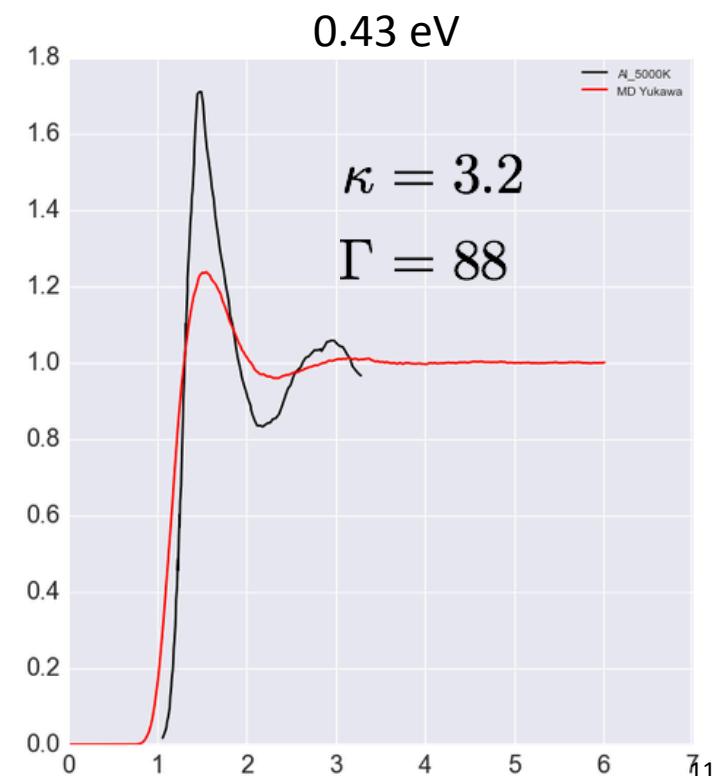
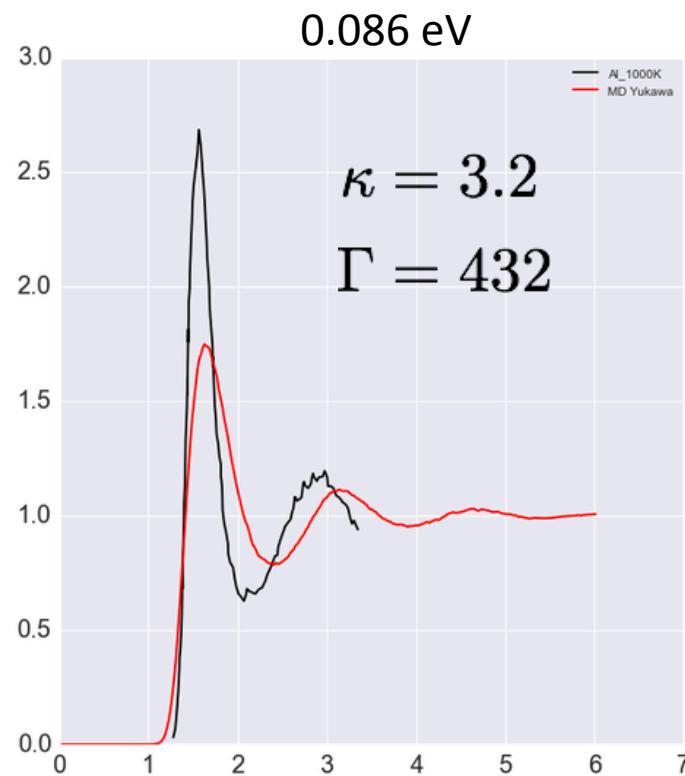
**No evidence of a metal-insulator transition in dense hot aluminum: A first-principles study**

Pier Luigi Silvestrelli

*Istituto Nazionale per la Fisica della Materia and Dipartimento di Fisica "G. Galilei," Università di Padova, via Marzolo 8,  
I-35131 Padova, Italy*

Aluminium, 2 g/cc

KS-DFT: 72 atoms

TF  $\langle Z \rangle / Z = 0.16$ 

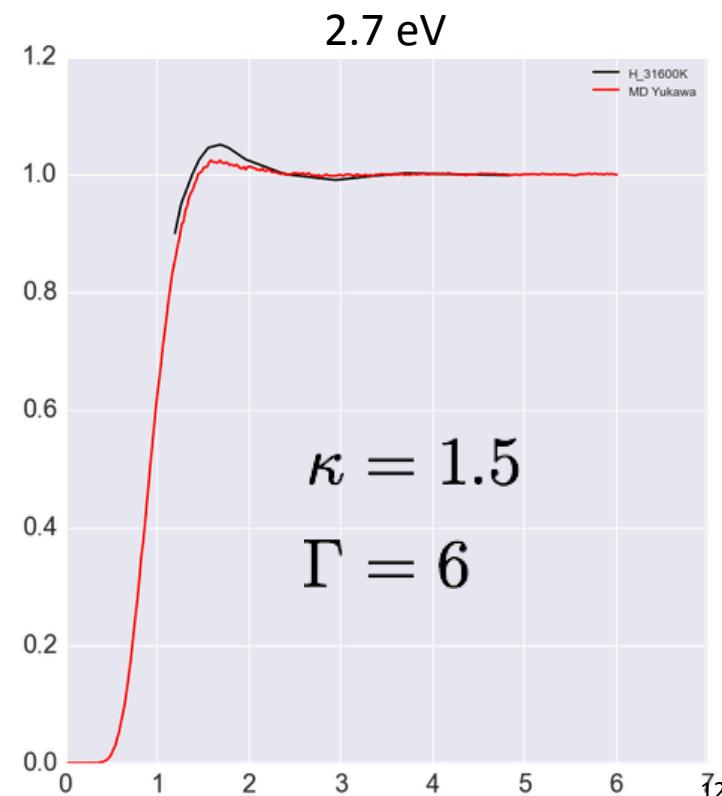
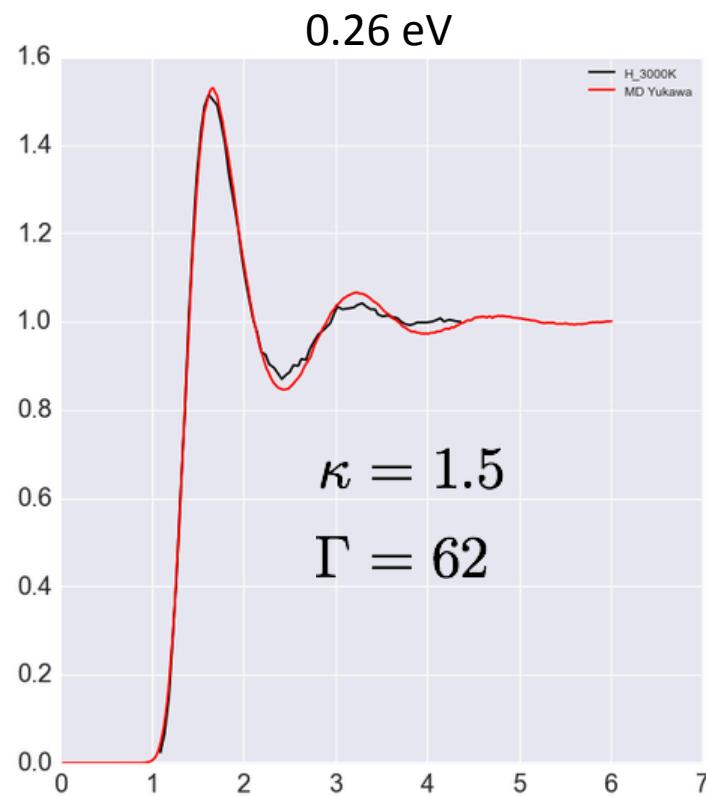
Hydrogen, 2.72 g/cc

OF-DFT

TF  $\langle Z \rangle / Z = 0.77$

## Density functional theory applied to metallic hydrogen: pair correlations and phase transitions

Hong Xu



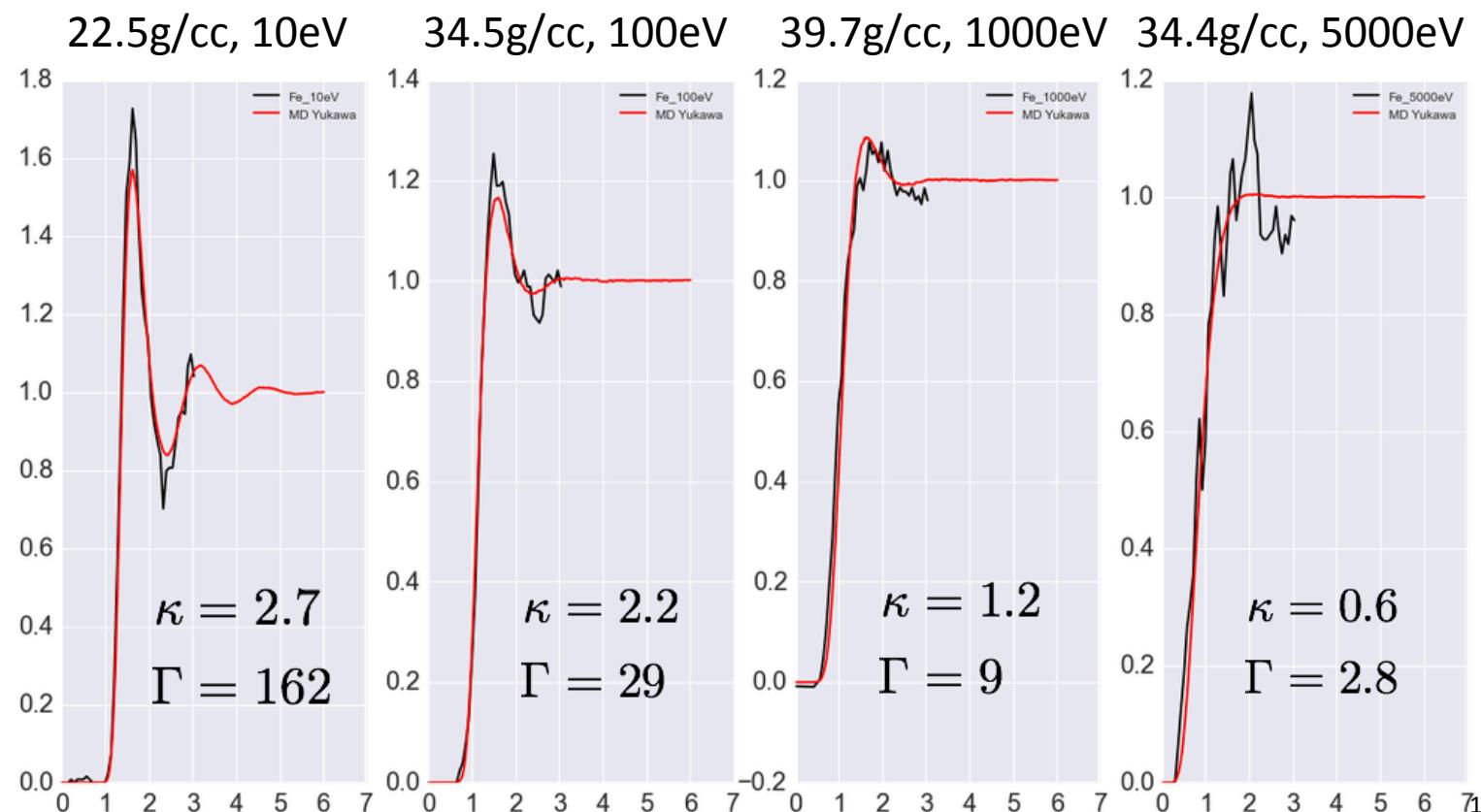
## Very-high-temperature molecular dynamics

Flavien Lambert, Jean Clérouin, and Gilles Zérah

*Département de Physique Théorique et Appliquée, CEA/DAM Île-de-France, BP12, 91680 Bruyères-le-Châtel Cedex, France*

Iron

KS-DFT: CPMD 54 atoms

TF  $\langle Z \rangle / Z = 0.35, 0.44, 0.77, 0.96$ 

# Yukawa Monte Carlo and Orbital Free Molecular Dynamics approaches for the equation of state and structural properties of hot dense matter

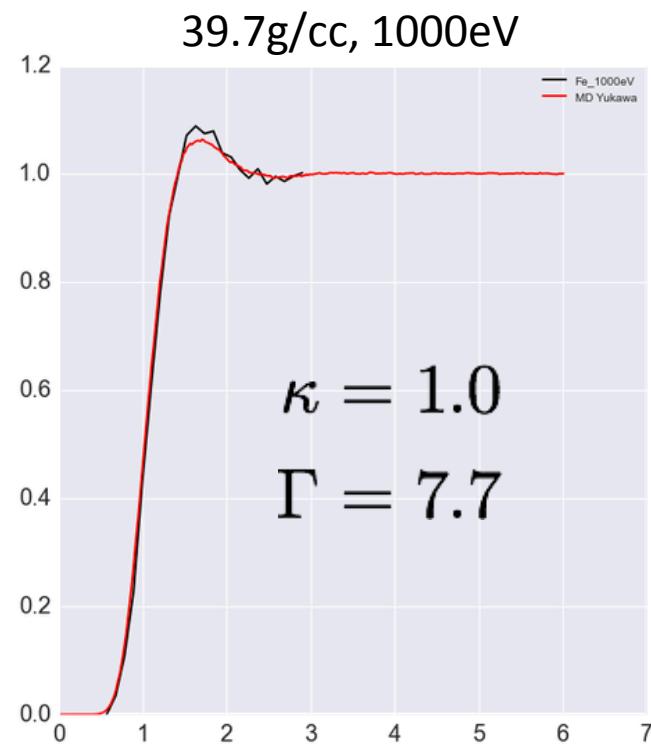
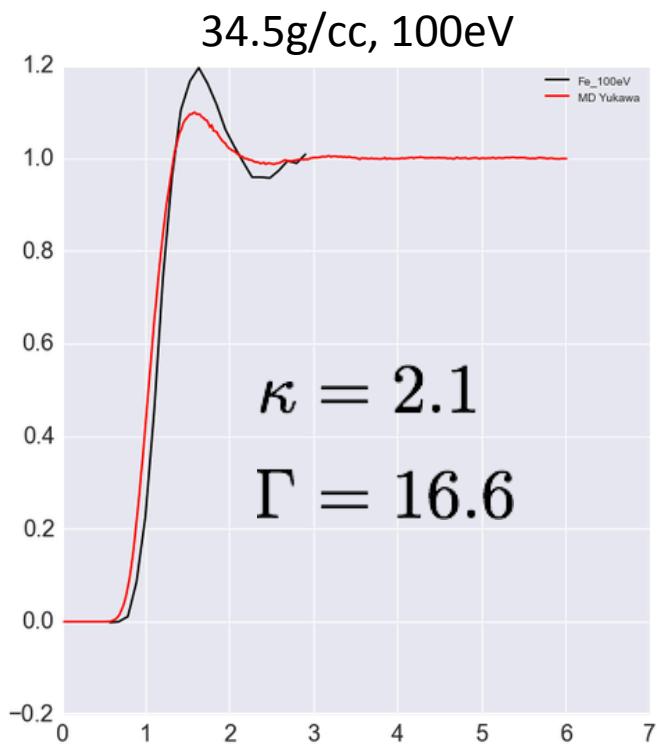
Iron

D. Gilles\*, F. Lambert, J. Clérouin, G. Salin

Commissariat à l'Energie Atomique, BP12, F-91680 Bruyères-le-Châtel, France

OF-DFT

TF  $\langle Z \rangle / Z = 0.38, 0.81$



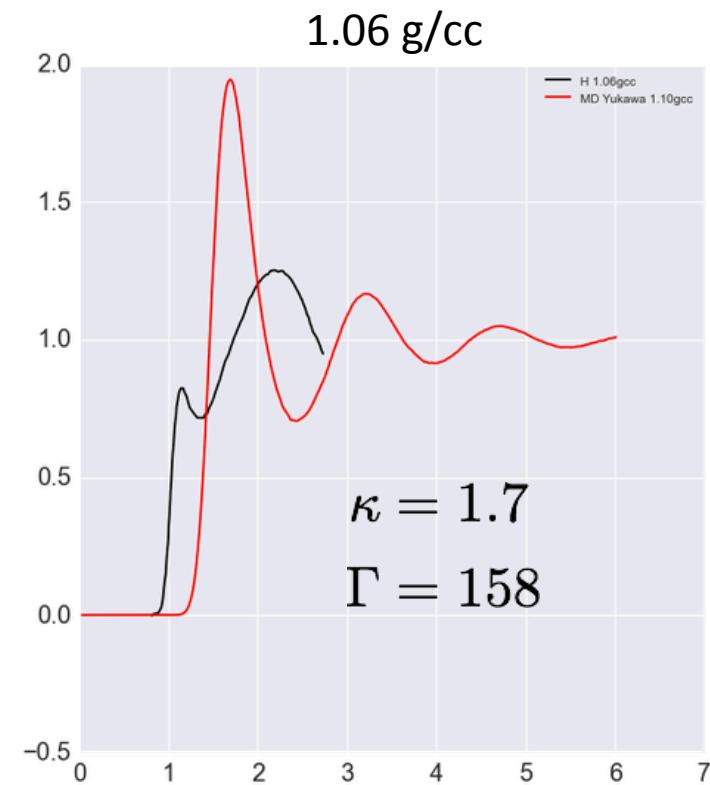
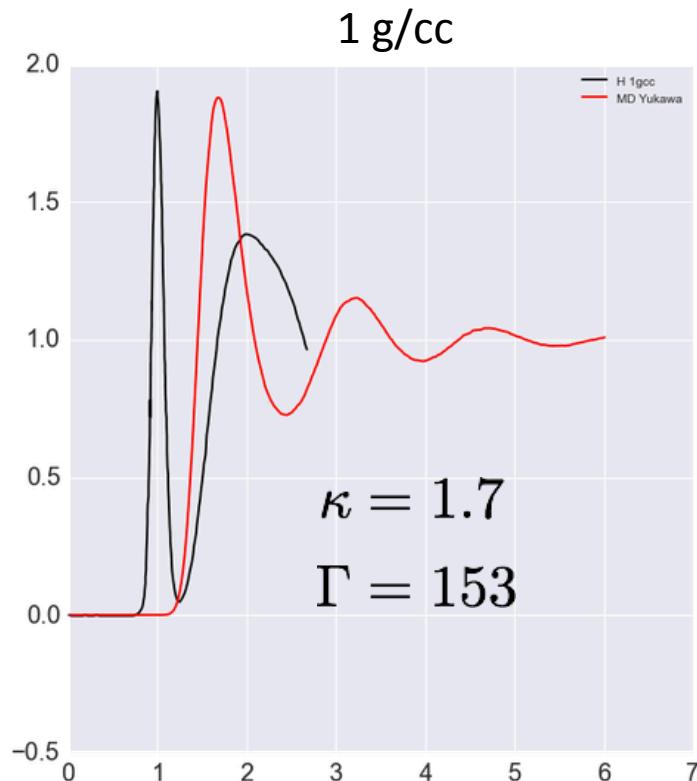
## First-order liquid-liquid phase transition in dense hydrogen

Winfried Lorenzen, Bastian Holst, and Ronald Redmer

*Institut für Physik, Universität Rostock, D-18051 Rostock, Germany*

Hydrogen at 0.06 eV

KS-DFT: 512 atoms FT-DFT (VASP)

TF  $\langle Z \rangle / Z = 0.7$ 

# Electron-ion and ion-ion potentials for modeling warm dense matter: Applications to laser-heated or shock-compressed Al and Si

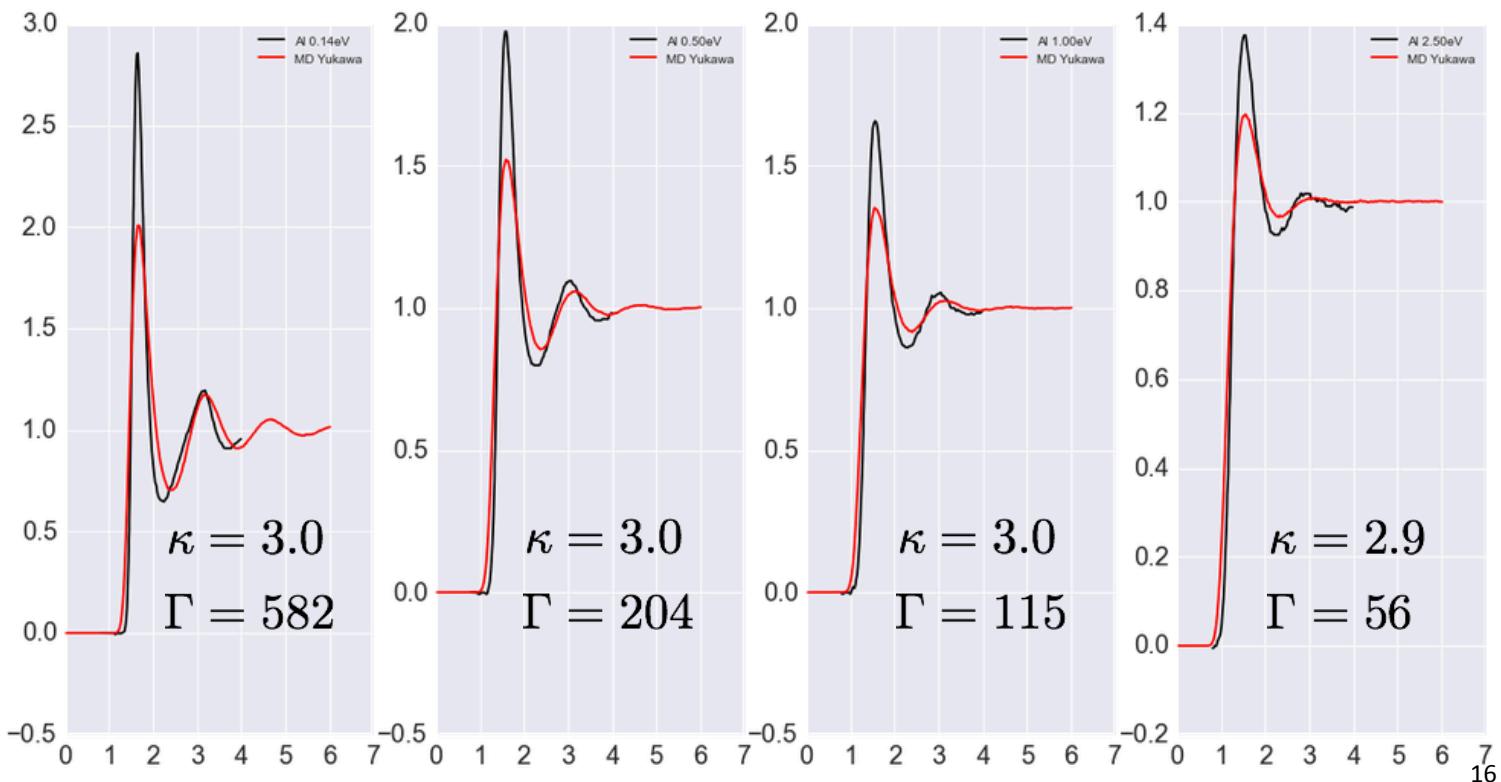
Aluminium  $T_e = 0.03$  eV

M. W. C. Dharma-wardana\*  
*National Research Council of Canada, Ottawa, Canada, K1A 0R6*

HNC

TF  $\langle Z \rangle / Z = 0.22, 0.24, 0.25, 0.26$

$3.78\text{g/cc}, 0.14\text{eV}$      $4.59\text{g/cc}, 0.5\text{eV}$      $5.1\text{g/cc}, 1\text{eV}$      $6.1\text{g/cc}, 2.5\text{eV}$



# Structure, equation of state, diffusion and viscosity of warm dense Fe under the conditions of a giant planet core

Fe 45 g/cc

KS-DFT: 216 atoms

TF  $\langle Z \rangle / Z = 0.33$

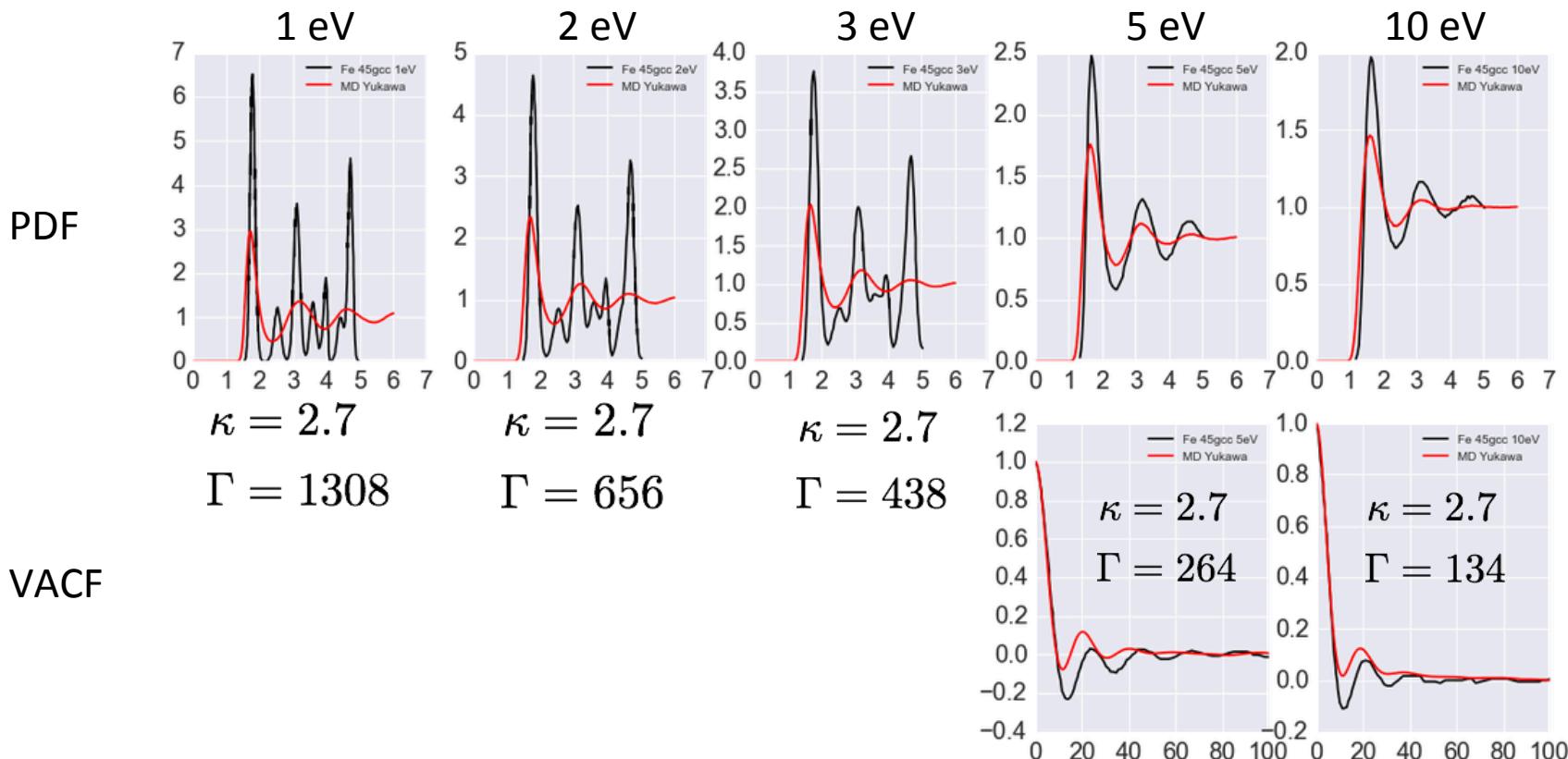
Jiayu Dai<sup>1,3</sup>, Yong Hou<sup>1</sup>, Dongdong Kang<sup>1</sup>, Huayang Sun<sup>1</sup>, Jianhua Wu<sup>1</sup> and Jianmin Yuan<sup>1,2,3</sup>

<sup>1</sup> Department of Physics, College of Science, National University of Defense Technology, Changsha 410073, People's Republic of China

<sup>2</sup> State Key Laboratory of High Performance Computing, National University of Defense Technology, Changsha 410073, People's Republic of China

E-mail: [jydai@nudt.edu.cn](mailto:jydai@nudt.edu.cn) and [jmyuan@nudt.edu.cn](mailto:jmyuan@nudt.edu.cn)

New Journal of Physics 15 (2013) 045003 (15pp)



# Classical-Map Hypernetted Chain Calculations for Dense Plasmas

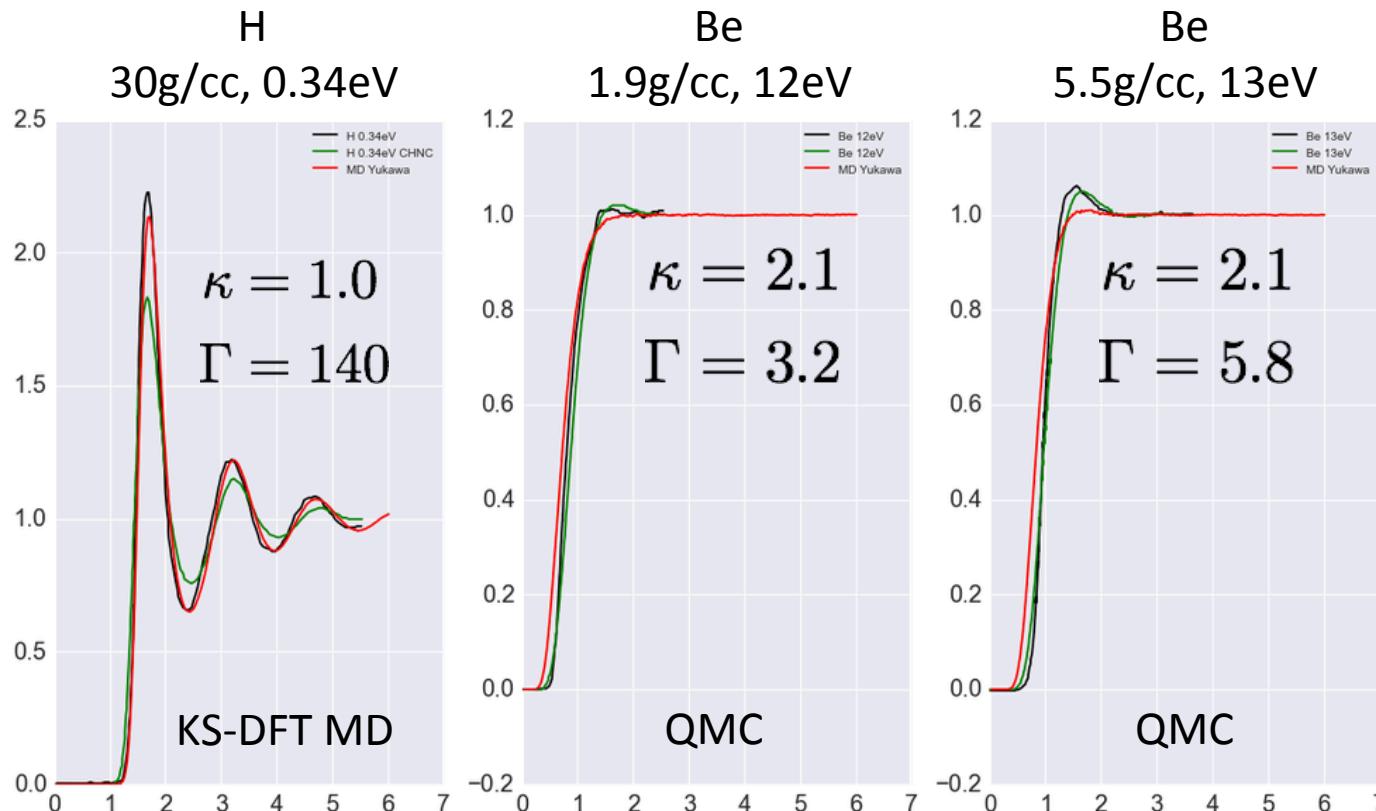
R. Bredow<sup>1\*</sup>, Th. Bornath<sup>1</sup>, W.-D. Kraeft<sup>1</sup>, M.W.C. Dharma-wardana<sup>2</sup>, and R. Redmer<sup>1</sup>

<sup>1</sup> Institut für Physik, Universität Rostock, 18051 Rostock, Germany

<sup>2</sup> National Research Council of Canada, Ottawa, Canada

CHNC

TF  $\langle Z \rangle/Z = 0.89, 0.46, 0.53$



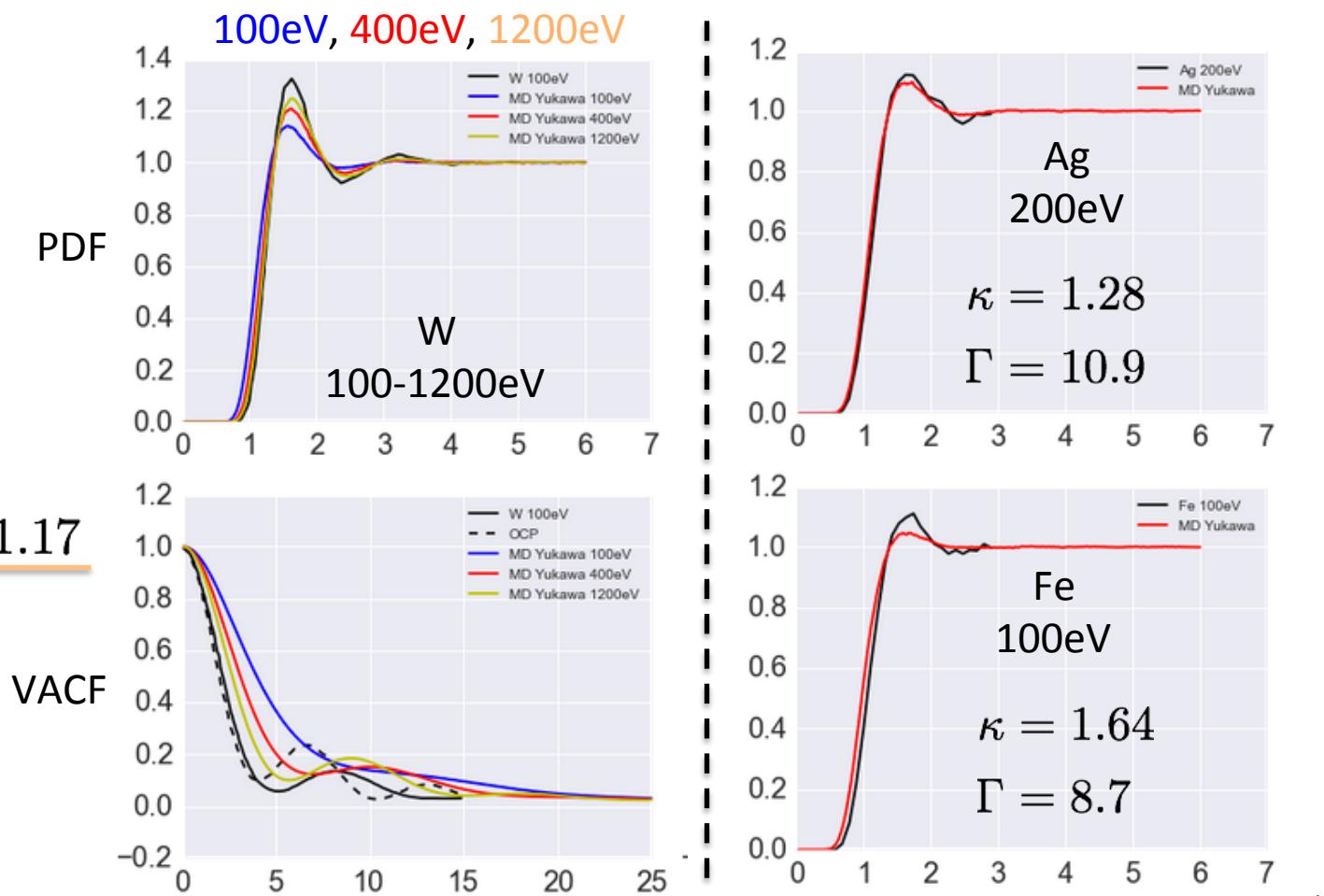
Tungsten	40g/cc
Silver	2.5g/cc
Iron	6.1g/cc

TF  $\langle Z \rangle / Z =$   
 W 0.19, 0.37, 0.63  
 Ag 0.42  
 Fe 0.37

$\kappa = 2.17, 1.56, 1.17$

$\Gamma = 23$

**INVITED ARTICLE**  
**Cooking strongly coupled plasmas**  
 Jean Clérouin\*  
*CEA, DAM, DIF, 91297 Arpajon, France*



# OF-MD and Integrated Yukawa Viscosity Model (iYVM)

PHYSICAL REVIEW E 93, 063208 (2016)

## Transport properties of an asymmetric mixture in the dense plasma regime

Christopher Ticknor, Joel D. Kress, and Lee A. Collins

Theoretical Division, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA

Jean Clérouin, Philippe Arnault, and Alain Decc

CEA, DAM, DIF, 91297 Arpajon, France

(Received 29 April 2016; published 23 June 2016)

- OFMD simulations
- Number of atoms : 64 ions for pure cases  
(up to 300 for mixtures)

A new viscosity data set is produced

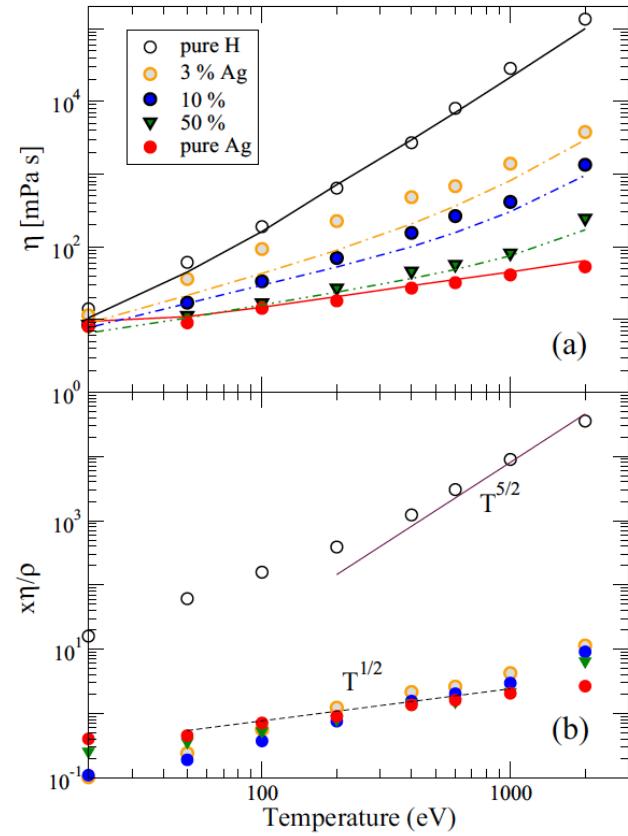


FIG. 9. (a) Viscosity as a function of temperature for both the PIJ model (lines) and OFMD simulations (symbols) at different concentrations: pure H (open circles), 3 (orange circles), 10 (blue circles), 50% (green down triangles), and pure Ag (red circles). (b)  $x\eta/\rho$  versus  $T$ . Lines represent the  $T^{5/2}$  and  $T^{1/2}$  scalings. 20

# OF-MD and Integrated Yukawa Viscosity Model (iYVM)

ELSEVIER

High Energy Density Physics 4 (2008) 49–57

[www.elsevier.com/locate/hephys](http://www.elsevier.com/locate/hephys)

## Viscosity estimates of liquid metals and warm dense matter using the Yukawa reference system

Michael S. Murillo\*

*Physics Division, MS D410, Los Alamos National Laboratory, Los Alamos, NM 87545, USA*

Received 26 September 2007; received in revised form 13 November 2007; accepted 28 November 2007  
Available online 14 December 2007

- New: iYVM
- Uses:
  - Ohta & Hamaguchi (MD)
  - +
  - Donko - Hartmann (MD)
  - +
  - Stanton - Murillo (EBE)

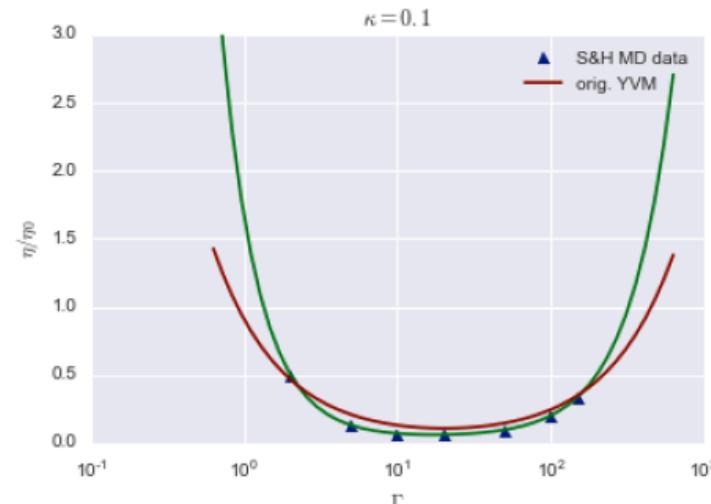
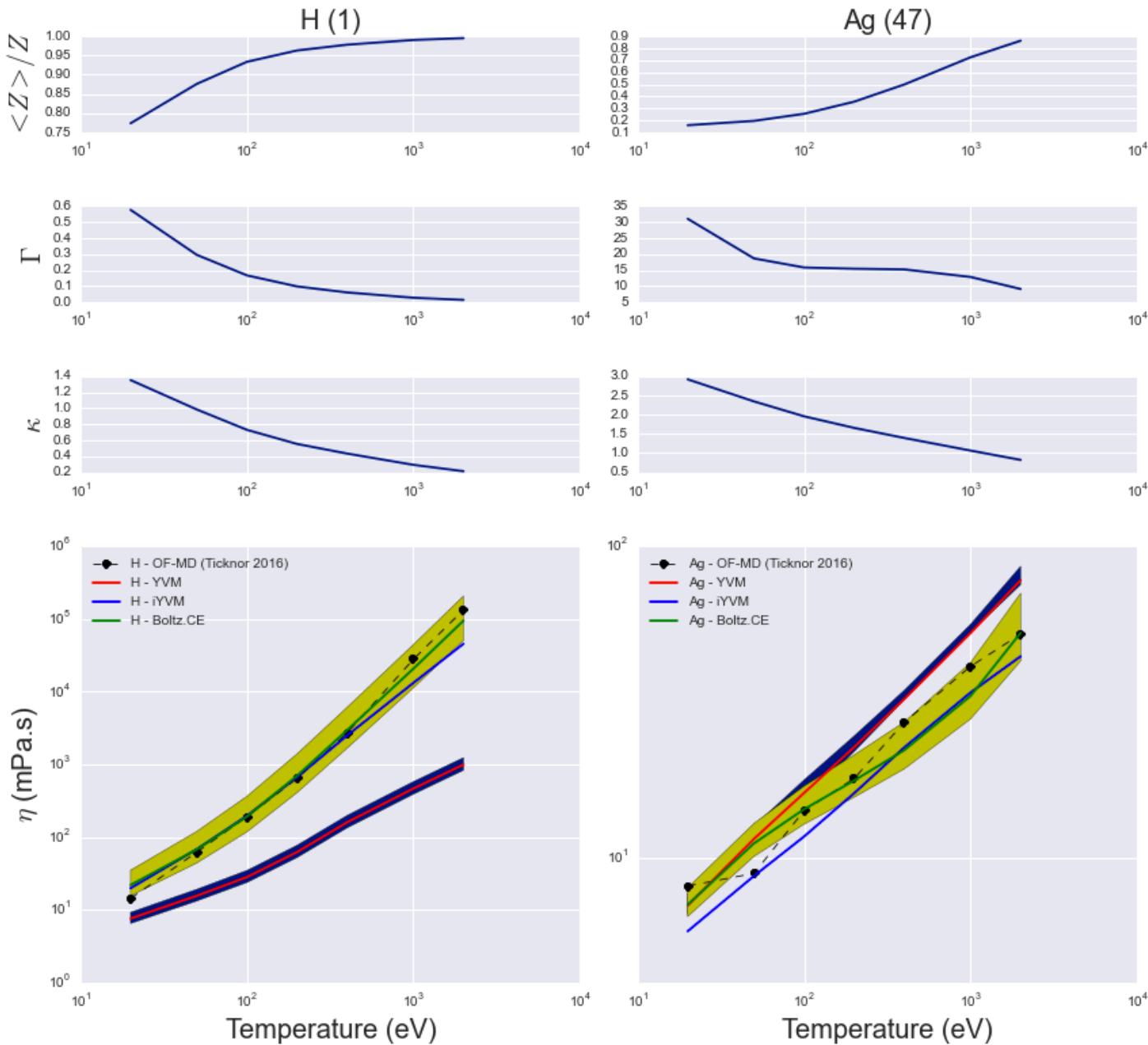


Fig. 1: For  $\kappa = 0.1$ , comparison of the SH MD data (black triangles), YVM (dark red line) and a new fit (green line) that does not include EBE data. Note that both fits are reasonable over the range of the data, but the difference can be very large outside of this range.

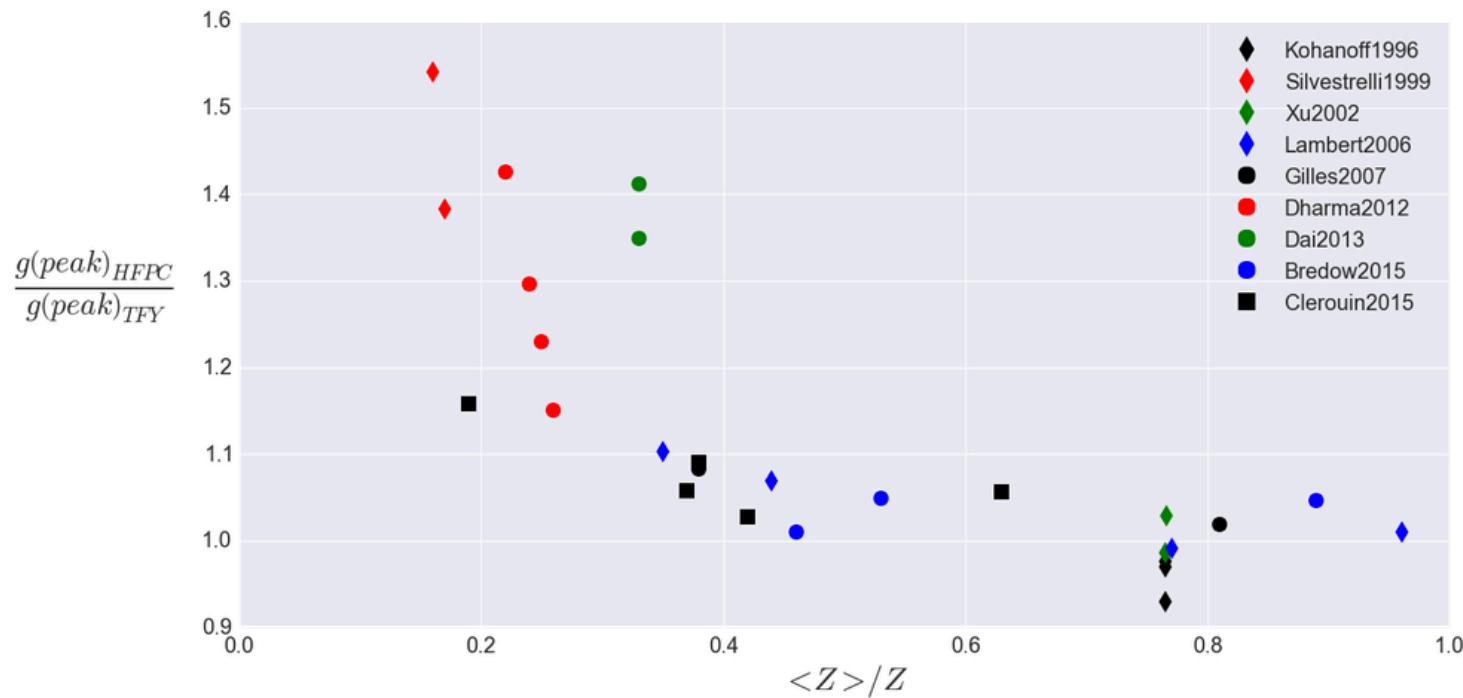
# OF-MD and Integrated Yukawa Viscosity Model (iYVM)



# High Fidelity Physics Codes comparisons

As a metric, we used the value of the pair distribution function at the first peak position.

Results show a correlation between the TF-ionization ratio  
and **HFPC** agreement for the PCF.



For  $\langle Z \rangle / Z > 0.5$  the agreement lies within the 5% difference.

Cases showing bond structures are not included.

# Summary

- Thomas-Fermi-Yukawa model for **HED** matter :

Good approximation of atomic physics when  $\langle Z \rangle / Z > 50\%$ .  
**however**

When the mean ionization state is lower, or molecular bonds are present  
higher complexity **HFPC** models than **TFY** are necessary.

- The iYVM extends the  $\Gamma - \kappa$  range of the YVM to low effective coupling  
and compares well with the plasma cases tested here.

- When **HED** systems meet  $\langle Z \rangle / Z > 50\%$   
the cheap computational cost of **TFY** allows to increase the simulated  
lengths and time scales by orders of magnitude.

**however**

Realistic systems may have significant density and temperature variations  
over time and space : **needs for a space-time resolved atomic physics.**

Computing the atomic physics locally on-the-fly : **MOD-MD**

Multi-scale Orbital-free DFT – **MD** model  
Murillo – Stanton - Glosli

# Outlook

- The **TFY** model: can we improve?

Empty core pair potential (finite extent of the bound density).

Going beyond the pair-potential approximation...

- A **database** is about to be released:

Gathering the available results in a user friendly database.

Will allow to compare the different **HPC** results ( currently  $g(r)$ , VACF ).

Understanding where the computational cost should be spent.

Contributions will be needed.