Theory of Warm Dense Matter

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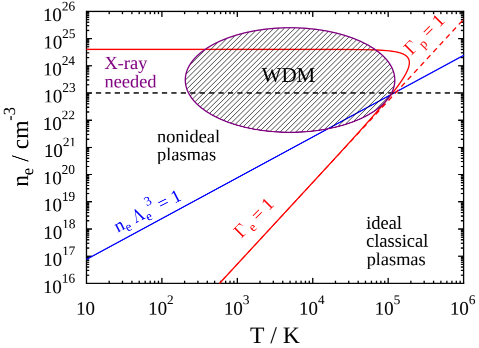
Introduction – Important Parameters for WDM Theory



Useful parameters to characterise dense plasmas / warm dense matter

- Classical coupling parameter

$$\Gamma_{aa} = \frac{Z_a^2 e^2}{a_a k_B T_a} \approx \frac{\langle E_a^{pot} \rangle}{\langle E_a^{kin} \rangle}$$

$\Gamma_{aa} > 1$ and $\Gamma_{ii} < 172$

⇒**no correlation expansion**

- Degeneracy parameter

$$n_e \Lambda_e^3 = n_e \left(\frac{2\pi\hbar^2}{m_e k_B T_e} \right)^{3/2}$$

$n_e \Lambda_e^3 > 0.1$

⇒**no quasi-classical theory**

$n_e \Lambda_e^3 < 10$

⇒**no perfect electron fluid**

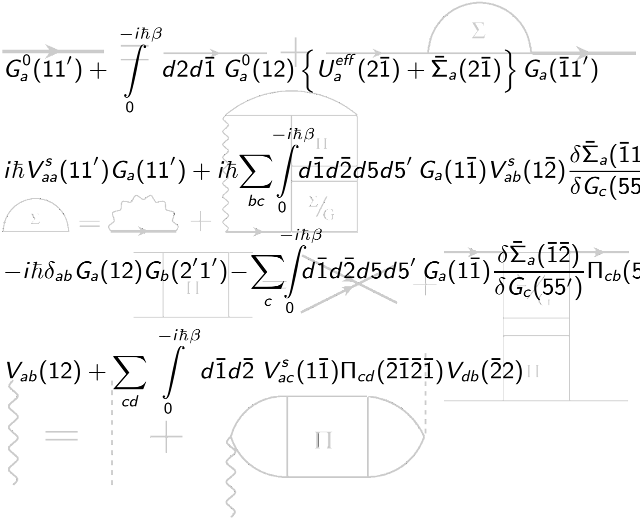
Quantum Statistical Description of WDM

$$G_a(11') = G_a^0(11') + \int_0^{-i\hbar\beta} d2\, d\bar{1}\; G_a^0(12)\left\{ U_a^{eff}(2\bar{1}) + \bar{\Sigma}_a(2\bar{1}) \right\} G_a(\bar{1}1')$$

$$\bar{\Sigma}_a(11') = i\hbar V^s_{aa}(11') G_a(11') + i\hbar \sum_{bc} \int_0^{-i\hbar\beta} d\bar{1}\, d\bar{2}\, d5\, d5'\; G_a(1\bar{1}) V^s_{ab}(1\bar{2}) \frac{\delta \bar{\Sigma}_a(\bar{1}1')}{\delta G_c(55')} \Pi_{cb}(5\bar{2}5'\bar{2})$$

$$\Pi_{ab}(12'1'2) = -i\hbar \delta_{ab} G_a(12) G_b(2'1') - \sum_c \int_0^{-i\hbar\beta} d\bar{1}\, d\bar{2}\, d5\, d5'\; G_a(1\bar{1}) \frac{\delta \bar{\Sigma}_a(\bar{1}\bar{2})}{\delta G_c(55')} \Pi_{cb}(525'2') G_a(\bar{2}1')$$

$$V^s_{ab}(12) = V_{ab}(12) + \sum_{cd} \int_0^{-i\hbar\beta} d\bar{1}\, d\bar{2}\; V^s_{ac}(1\bar{1}) \Pi_{cd}(\bar{2}\bar{1}\bar{2}\bar{1}) V_{db}(\bar{2}2)$$



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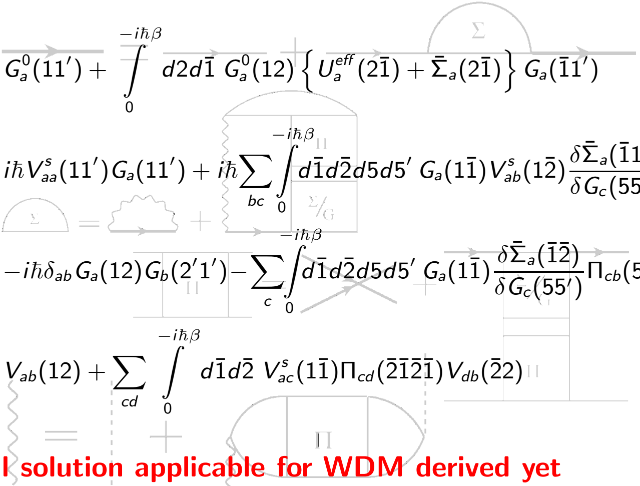

$$G_a(11') = G_a^0(11') + \int_0^{-i\hbar\beta} d2\,d\bar{1}\; G_a^0(12)\left\{ U_a^{eff}(2\bar{1}) + \bar{\Sigma}_a(2\bar{1}) \right\} G_a(\bar{1}1')$$

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$$V_{ab}^s(12) = V_{ab}(12) + \sum_{cd} \int_0^{-i\hbar\beta} d\bar{1}\,d\bar{2}\; V_{ac}^s(1\bar{1}) \Pi_{cd}(\bar{2}\bar{1}\bar{2}\bar{1}) V_{db}(\bar{2}2)$$

No general solution applicable for WDM derived yet
Approximate quantum solutions use expansion in correlations (RPA)

Basics for Dynamic Quantum Simulation of WDM

many particle
system - exact

$$\begin{tikzcd}
V_{ext}(\mathbf{r}) \arrow[d] & n_0(\mathbf{r}) \arrow[l, "\mathbf{H-K}"'] \\
\Psi_i(\{\mathbf{r}\}) \arrow[r] & \Psi_0(\{\mathbf{r}\}) \arrow[u]
\end{tikzcd}$$

Hohenberg&Kohn
proof

Assumption

$$\begin{tikzcd}
{} \arrow[r, leftrightarrow, "K-S"] & {}
\end{tikzcd}$$

Kohn&Sham
Ansatz

many particle
system - noninteracting

$$\begin{tikzcd}
n_0(\mathbf{r}) \arrow[r, leftrightarrow, "\mathbf{H-K_0}"] & V_{KS}(\mathbf{r}) \arrow[d, leftrightarrow] \\
\Psi_{i=1\ldots N}(\mathbf{r}) \arrow[u] \arrow[r, leftrightarrow] & \Psi_0(\mathbf{r})
\end{tikzcd}$$

Hohenberg&Kohn
proof

$$\left[-\frac{1}{2}\nabla^2 + V_{KS}^{\sigma}(r)\right]\psi_i^{\sigma}(r) = \varepsilon_i^{\sigma}\psi_i^{\sigma}(r)$$

$$V_{KS}^{\sigma}(r) = V_{ext}(r) + V_H[n] + V_{xc}^{\sigma}[n^{\uparrow}, n^{\downarrow}]$$

DFT → forces → classical molecular dynamics for ions (DFT-MD)

Practical (fast) Theory Applicable for WDM

Practical (fast) Theory Applicable for WDM

Present Choices:

- Approximate theory neglecting important physics;
  importance / magnitude of error even partially unknown
- Extensive computer simulations running for weeks;
  moreover, simulations restricted to lower temperatures

- Very few data points for comparison with more approximate theories

⇒ **Every additional experimental data point is very welcome!**

Ionic Structure for Strong Coupling – HNC

Ornstein-Zernicke relation (exact)

$$h_{ab}(\mathbf{r}) = c_{ab}(\mathbf{r}) + \sum_c n_c \int d\bar{\mathbf{r}}\, c_{ac}(\bar{\mathbf{r}})\, h_{cb}(|\mathbf{r} - \bar{\mathbf{r}}|)$$

Convolution! ⇒ transformation in Fourier space (matrix notation)

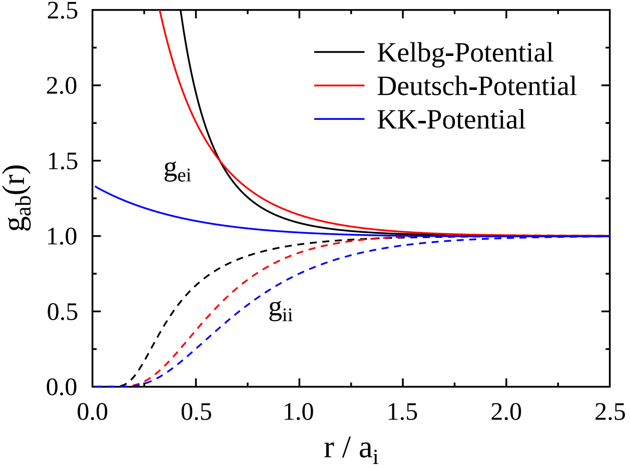
$$\tilde{H}(k) = \tilde{C}(k) + \tilde{C}(k)\, \delta_{ab} n_a\, \tilde{H}(k)$$

Hypernetted Chain (HNC) closure relation (approximation)

$$g_{ab}(r) = \exp(-\beta V_{ab}(r) + h_{ab}(r) - c_{ab}(r))$$

- Approach uses **classical mechanics** only
- Approach **works well** for ions up to coupling strength of $\Gamma \leq 100$

Results for Electron-Ion Systems – Quantum Potentials



Electron-ion and ion-ion pair distributions for a plasma with $n = 10^{22}\,\mathrm{cm}^{-3}$, $Z = 1$, and $T = 4.5 \times 10^4\,\mathrm{K}$.

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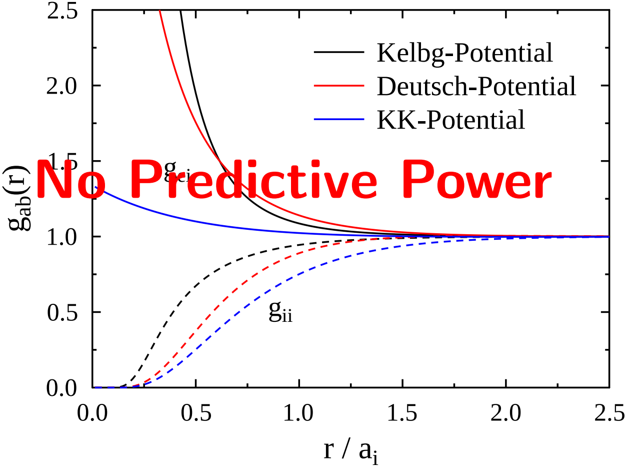
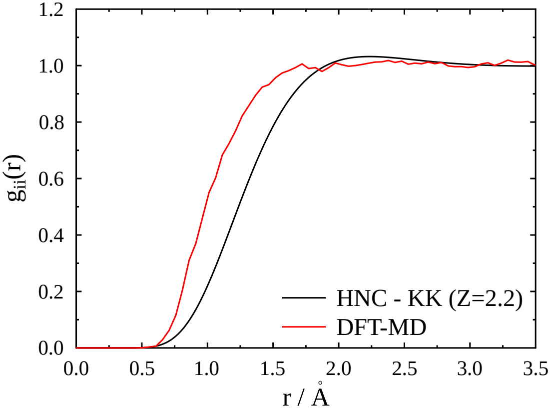


Electron-ion and ion-ion pair distributions for a plasma with $n = 10^{22}\,\mathrm{cm}^{-3}$, $Z = 1$, and $T = 4.5 \times 10^4\,\mathrm{K}$.

Hypernetted Chain Calculations vs. Quantum Simulations



Beryllium plasma with:

$n_i = 1.23 \times 10^{23}\,\mathrm{cm}^{-3}$

$T = 1.39 \times 10^{5}\,\mathrm{K}$

(isochorically heated)

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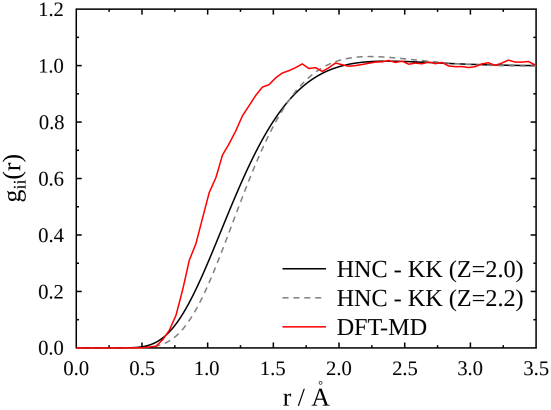

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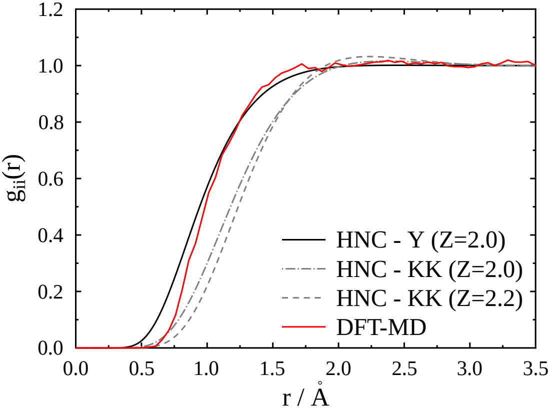

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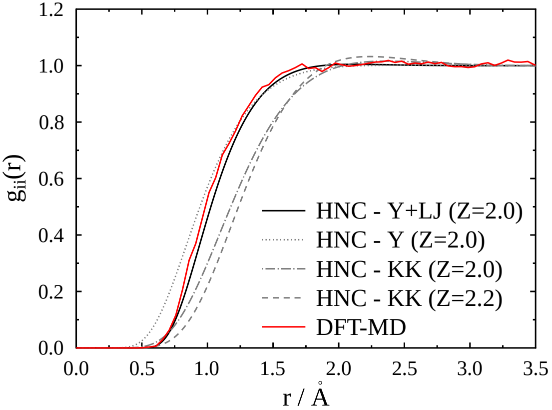

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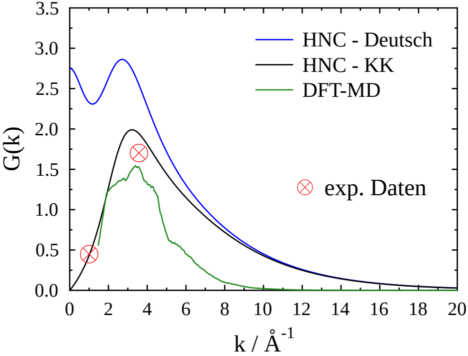
(isochorically heated)

⇒ **good agreement with Yukawa+LJ model**

- Yukawa model: linearly screened interaction potential (deg. electrons)
- 1s shell results additional repulsion at small distances $\sim 1/r^4$ (LJ part)
- 1s shell must be intact! ⇒ Possibility to obtain charge state: $\bar{Z} = 2$

HNC Results, Quantum Simulations and Experiments

Weight of the Ion Feature in the Thomson Scattering Signal



Weight of the ion peak (HNC)

$$G(k) \quad = \left|f_i(k) + q(k)\right|^2 S_{ii}(k)$$

Weight using DFT-MD

$$G(k) \quad = n_e^2(k) S_{ii}(k)$$

plasma parameters:
$n_i = 1.23 \times 10^{23}\,\mathrm{cm}^{-3}$,
$T = 12\,\mathrm{eV}$, $Z = 2$

⇒ Good agreement: DFT-MD simulations and experiments

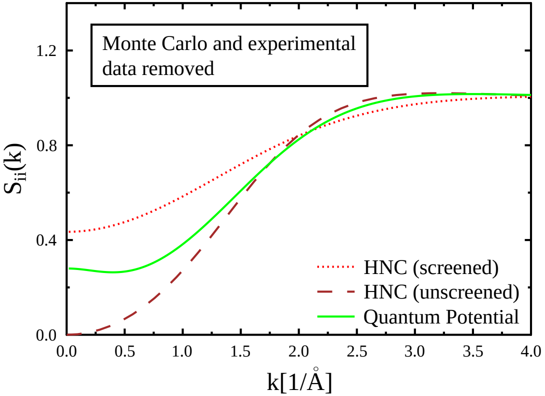
⇒ Good agreement: DFT-MD and HNC using KK-potential

⇒ Need to measure at higher k values using much harder x-rays

Experimental data from: Glenzer et al., PRL (2003) and Glenzer et al., PRL (2007)

HNC Results, Quantum Simulations and Experiments

Ion Structure in Compressed Lithium



Static structure factor for a lithium plasma with $n_i = 5.2 \times 10^{22}\,\mathrm{cm}^{-3}$, $T = 4.5\,\mathrm{eV}$, $\bar{Z} = 1.35$.

Experimental data points: García Saiz et al. (submitted)

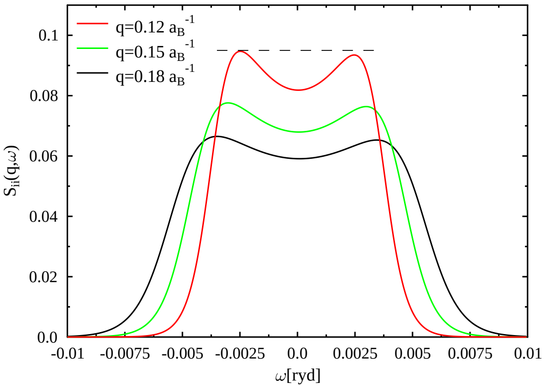
Insights gained:

- Good agreement: DFT-MD simulations, linearly screened HNC and experiments
- HNC with quantum potentials yields too strongly coupled ions (screening too weak)
- Strange feature at smaller k values

⇒ Need for a strong **point source**

Dynamics of the Ions from X-ray Scattering

Dynamic Ion Structure Factor

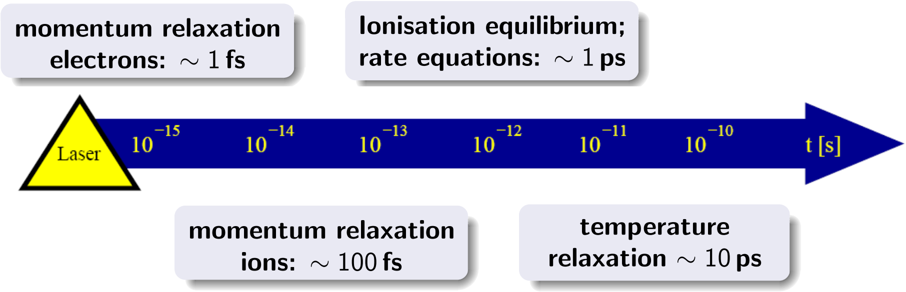


Dynamic ion structure factor for an hydrogen plasmas with $n_i = 10^{22}\,\mathrm{cm}^{-3}$ and $T = 3 \times 10^4\,\mathrm{K}$.

Experimental demands:

- Well defined x-ray source; spectral width $\ll 0.01\,\mathrm{eV}$
- Very small scattering angle; point source
- High energy laser for sample creation
- Different means of diagnostics of plasma parameters or highly reproducible experiments
- Better theoretical models

Time Scales: Relaxation in Warm Dense Matter



momentum relaxation electrons: ~ 1 **fs**

Ionisation equilibrium; rate equations: ~ 1 **ps**

momentum relaxation ions: ~ 100 **fs**

temperature relaxation ~ 10 **ps**

Relaxation processes

- are intrinsically connected to the creation of WDM
- define **any** equilibrium measurement
- reveal many WDM properties more pronounced

Goals defines times structure needed for the probing light source

Summary and Demands for Light Source

Demands on the x-ray source

- Bright x-ray source for diagnostics of WDM
- Source must be tunable over two orders of magnitude
- X-rays must be focused enough to allow for very small scattering angle
- Spectral width of the x-rays should be smaller than 0.01 eV
- Pulses should be shorter than 100 fs; shorter: more applications

Additional sources and beams

- High energy source to create WDM: electron beam, multiple lasers
- Independent diagnostics for cross validation is needed
- Laser-produced ion beams could yield additional information
- For relaxation studies, a high energy short pulse laser is needed

⇒ **There is a good scientific case for several optical lasers**