X-ray Scattering in Multicomponent Warm Dense Matter

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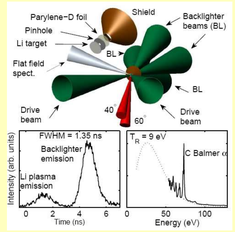
Motivation

A deeper understanding of warm dense matter (WDM) is of particular interest in order to advance inertial fusion research. For this purpose and to shed light on some fundamental issues of matter under extreme conditions, huge efforts have been made to create such a state of matter in a controlled laboratory environment over the last years. It involves compression and heating of material to solid state densities and beyond at several electron volts. This WDM state presents severe challenges which from a theory side comprise the simultaneous description of strongly coupled ions and quantum degenerate electrons.
Besides generation, diagnostics of WDM poses a challenge. X-ray scattering has emerged as a powerful tool, capable of delivering density, temperature, charge state and structural information at solid state densities due to the direct correlation of the power spectrum to the dynamic structure factor [1-4]

$$\frac{\mathrm{d}^2P(\mathbf{k},\omega)}{\mathrm{d}\Omega\,\mathrm{d}\omega} \propto S_{ee}^{tot}(\mathbf{k},\omega)\ .$$

A complete diagnostic does however rely on an excellent theoretical understanding of the structure of the WDM state under investigation. A first principle approach to electron and ion distribution seems favourable as it avoids the chemical picture. Recently, attention was brought to multicomponent WDM, where the interplay between different correlated ion species gives rise to new features in the scattering spectrum.



Schematic experimental setup for the investigation of warm dense lithium [4].

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Electronic structure for plasmas with multiple ion species

Let us consider a partially ionised plasma with bound and free electrons coupled with N_a nuclei of several ion species a.

- total electron density → core electrons ϱ_c & free-electrons ϱ_f

$$\varrho_e(\mathbf{k},t) = \varrho_c(\mathbf{k},t) + \varrho_f(\mathbf{k},t) = \sum_{\text{ions}} \varrho_c^a(\mathbf{k},t) + \varrho_f(\mathbf{k},t)$$

- intermediate scattering functions for multiple components

$$F_{ab} = \langle \varrho_a(\mathbf{k},t)\varrho_b(-\mathbf{k},0)\rangle$$

- total electronic dynamic structure factor

$$S_{ee}^{tot}(\mathbf{k},\omega) = \frac{1}{2\pi N_a}\int F_{ee}^{tot}(\mathbf{k},t)\mathrm{e}^{i\omega t}\mathrm{d}t$$

Following the steps as described by Chihara [5,6], we obtain the total structure factor.

Result for one ion species:
(for light elements neglecting the core excitations)

$$S_{ee}^{tot}(\mathbf{k},\omega) = \underbrace{Z_f S_{ee}^0(\mathbf{k},\omega)}_{\text{electron feature}} + \underbrace{|f_i(k)+q(k)|^2\, S_{ii}(\mathbf{k},\omega)}_{\text{ion feature}}$$

- *electron feature:* scattering contribution of free electrons
- *ion feature:* describes electrons co-moving with the ions:
  $f_i(k)$ - atomic form factor $q(k)$ - screening cloud

Result for two ion species [12]:

$$\begin{aligned} S_{ee}^{tot}(k,\omega) = {} & [f_1+q_1]^2\frac{n_1}{n}S_{11}(k,\omega) + [f_2+q_2]^2\frac{n_2}{n}S_{22}(k,\omega) \\ & + 2\left[f_1f_2 + q_1q_2 + f_1q_2 + f_2q_1\right]\frac{\sqrt{n_1n_2}}{n}S_{12}(k,\omega) \\ & + Z_f S_{ee}^0 + \text{core excitations} \end{aligned}$$

The generalisation to multiple ion species leads to:

- consideration of all mutual correlations of the several ion species, characterised by the partial structure factors $S_{ab}(k,\omega)$, weighted statistically by the densities
- occurrence of several atomic form factors and screening functions according the ion species

Total electronic structure factor for multiple ion species:

$$S_{ee}^{\mathrm{tot}}(\mathbf{k},\omega) = \underbrace{\sum_{a,b}\frac{\sqrt{n_an_b}}{n}\left[f_a(\mathbf{k})+q_a(\mathbf{k})\right]\left[f_b(\mathbf{k})+q_b(\mathbf{k})\right]S_{ab}(\mathbf{k},\omega)}_{\text{ion feature}} + \underbrace{Z_fS_{ee}^0(\mathbf{k},\omega)}_{\text{electron feature}} + \underbrace{\sum_a\frac{n_a}{n}\int Z_B^a\tilde{S}^{c_ae}(k,\omega-\omega')S_{S_a}(k,\omega')\mathrm{d}\omega'}_{\text{core excitations}}$$

Theoretical scattering spectrum

To generate a theoretical scattering spectrum for the x-ray scattering process comparable to experimental results, the finite bandwith of the beam as well as the finite resolution of the detector have to be taken into account [9]. Furthermore, the uncertain regarding the scattered angle and thus the wave number have to be considered. Therefore, the dynamic structure factor is convoluted with a weighting function, here a normalized gaussian, for the energy as well as the wavenumber:

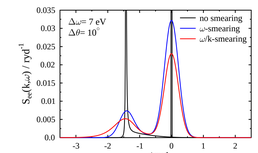
$$S_{ee}^{\mathrm{tot}}(k,\omega) = (S_{ee}(k,\omega) * g_\omega(\omega)) * g_k(k) = \frac{1}{2\pi\sigma_\omega\sigma_k}\int S_{ee}^{\mathrm{tot}}(k',\omega')\times\exp\left(-\frac{(\omega-\omega_0-\omega')^2}{2\sigma_\omega^2}\right)\times\exp\left(-\frac{(k-k')^2}{2\sigma_k^2}\right)\mathrm{d}\omega'\mathrm{d}k'\ .$$

Methods for the calculation of $S_{ee}^{\mathrm{tot}}(k,\omega)$:

Ion feature: strongly coupled ions + degenerate electrons [7,8]
- a) classical integral equations (hypernetted chain – HNC)
  → Yukawa potential with a short range repulsion for systems with full inner shells - Y+SRR
  → quantum pseudo-potentials, e.g. Klimontovich-Kraeft potential - KK (questionable in the WDM regime)
- b) density functional molecular dynamics simulations – DFT-MD

electron feature: weakly coupled degenerate electrons well described by random phase approximation – RPA

core excitations: negligible for low Z materials



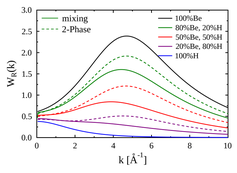
A theoretical x-ray scattering spectrum for a lithium plasma with $T = 1.7eV$, n_e=1.6×10$^{23}cm^{-3}$, Z=1 and the scattering vector $k = 1\AA$. Ion feature calculated with HNC method using a Yukawa model that is in agreement with DFT-MD simulations.

In conclusion:

- currently no experimental access to the frequency part of the ion feature → XFEL
- higher precision measurements necessary for a more accurate spectrally resolved x-ray scattering spectrum and a better signal-to-noise ratio

XRTS as a probe for mixtures and mixing

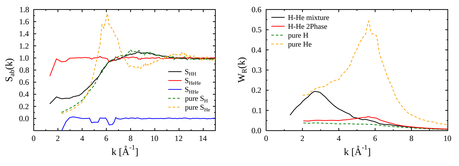
Beryllium-Hydrogen-Mixtures



The ion peak for a Be-H mixture comparing results from a 2-phase model with a completely mixed plasma at $T = 1.5\times10^5\,K$ and $n_e \approx 8\times10^{24}cm^{-3}$.

- beryllium, as a low-atomic-number material, is used as a capsule element in ICF
- the capsule performance depends on the Be-H mixing
- a warm dense matter state occurs during the compression of the fuel
- the strength of the ion feature varies with the Be/H content
- the strength of the ion feature varies between a 2-phase model and a completely mixed plasma consisting of Be and H ions

Helium-Hydrogen-Mixture



DFT-MD results for the partial structure factors $S_{\alpha\beta}(k)$ and the ion peaks $W_R(k)$ for a He-H mixture in comparison with results for pure He and H gas (T=10^4K and n_e=10$^{24}cm^{-3}$).

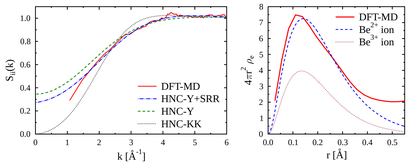
- He-H mixtures naturally occur in astrophysical objects, e.g. giant gas planets (Jupiter)
- presented mixture consists of 7.6 % atomic helium and 92.4% fully ionised hydrogen (Jupiter conditions)
- possibility of phase separation and additional layer boundary inside the planet
- the ion feature varies between model with two separate phases and a completely mixed plasma

In conclusion:

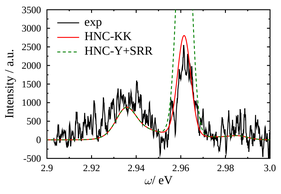
⇒ x-ray Thomson scattering is an applicable method to probe dense plasmas for mixing properties

Scattering spectrum of Beryllium

We compare the theoretical x-ray scattering spectrum of solid density beryllium with experimental results presented in [2]. The plasma parameters are given by $\varrho = 1.848\mathrm{g/cm}^3$, $T = 12\mathrm{eV}$ and $Z_f = 2$. The x-ray energy is $E_0 = 2.96\mathrm{keV}$ and the scattering angle is $\theta = 40°$, i.e. the collective scattering regime is considered.



Structure in beryllium for solid density. Left: the static structure factor comparing the different interaction potentials in HNC with results from DFT-MD. Right: the electron density around an ion.



Theoretical x-ray scattering spectrum in forward scattering ($k = 1\AA$). The Rayleigh peak was calculated using HNC with the KK-potential (red solid line) and the Yukawa+SRR-potential (green dashed line).

Static structure factor:

- HNC calculation using quantum-pseudo potential (KK) underestimates the electron-ion interaction
- HNC calculation using Yukawa model with an additional short range repulsion yields good agreement with DFT-MD results

Electron density around ions:

- DFT-MD results for the electron density compared with bound wave functions for isolated ions at two charge states

→ ions are doubly charged in the parameters considered

Free electron feature

- RPA treatment of the free electrons in the systems reflects the plasmon scattering very well

Contradiction:

- HNC-Y+SRR yields correct static ionic structure factor BUT highly overestimates the Rayleigh Peak
- correlation effects of the electron density still not well-understood; influence of the screening function?