Theory of X-ray Scattering from Warm Dense Mixtures

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Motivation

Warm dense matter (WDM) is often characterized as exhibiting both strongly coupled ions and partially degenerate electrons. This combination leads to big challenges in the theoretically description as standard plasma and solid state theories lose their applicability in the WDM region. The description is further complicated by the fact that the matter in naturally occurring WDM, like planets and stars, as well as in technology, like ICF and laser ablation, usually consists of a mixture of several elements. Experiments applying x-ray scattering have been established as a reliable diagnostic tool for WDM strongly enhancing our understanding. However, these experiments rely on well-founded theoretical models for the (dynamic) electron and the ion structure in WDM and are presently limited to WDM with single components or in simple conditions.

In this contribution, we present the theoretical framework to analyze the x-ray scattering signal from complex WDM consisting of several ionic species [1]. All interactions between the ions of the same and different species are fully taken into account. This novel approach allows us to analyze recent experiments that have moved from simple elements, like Be and Li [2-4], to more complex, composite matter such as CH or LiH [5,6]. The free electron feature is not affected by the different ion species. Thus, we will mainly concentrating on the elastic scattering feature which is characterised by the microscopic ion structure and the electron density around the ion (bound electrons and screening cloud). We will demonstrate that the elastic scattering feature is very sensitive to the ratio of materials in the probe volume. Thus, x-ray scattering can be applied to probe the mixing properties of materials in the WDM region. This has important implications for ICF research (ablator-fuel mixing) and planetary physics (demixing of hydrogen-helium). Moreover, full quantum simulations show that the inter-ionic forces, and thus the ionic structure, are much more complex than described by the linearly screened Coulomb potential when matter is not fully ionized [7].

References

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Generalised electronic structure for multi-component plasmas

Consider a partially ionised plasma with bound and free electrons coupled with $\sum N_a$ nuclei of several ion species a.

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

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

- 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 [8,9], 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:

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

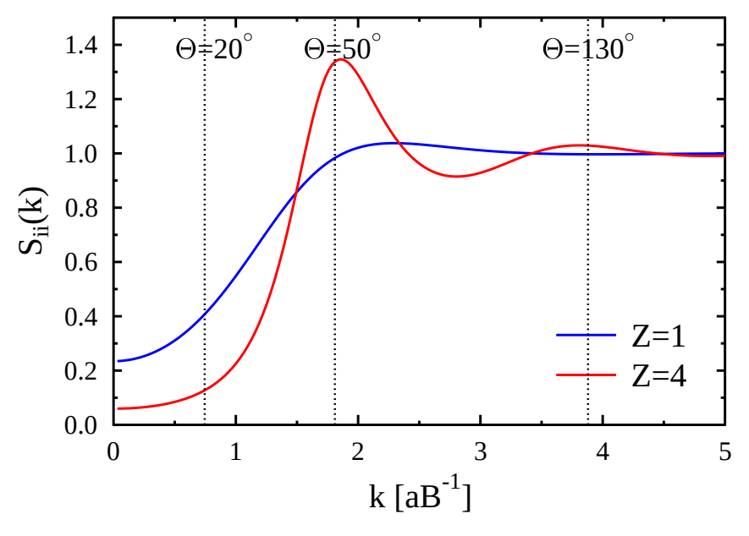
The generalisation to multiple ion species leads to:

- 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 to the ion species

Total electronic structure factor for multiple ion species [1]:

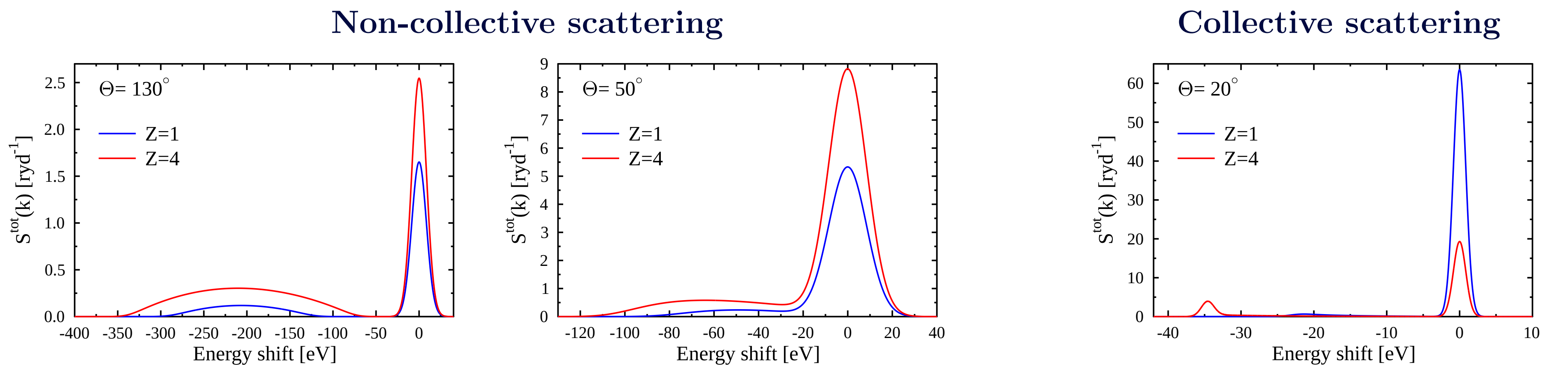
$$S_{ee}^{\text{tot}}(\mathbf{k},\omega) = \underbrace{\sum_{a,b}\frac{\sqrt{n_an_b}}{n}\,[f_a(\mathbf{k})+q_a(\mathbf{k})]\,[f_b(\mathbf{k})+q_b(\mathbf{k})]\,S_{ab}(\mathbf{k},\omega)}_{\text{ion feature}} + \underbrace{Z_f S_{ee}^0(\mathbf{k},\omega)}_{\text{electron feature}} + \underbrace{\sum_a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

Collective vs. non-collective spectrum of a Carbon plasma



Static structure factor of a carbon plasma with $T = 1eV$ and $\varrho = 2.2g/cm^3$ calculated with HNC method using a Yukawa model for two different ionization states.

Non-collective scattering	**Collective scattering**
inelastic Compton scattering → velocity distribution (n_e/T_e)	inelastic free electrons → plasma oscillations (n_e)
elastic Rayleigh Peak → bound electrons (Z_B)	elastic bound electrons → ionic structure (Z_B/T_e)



Theoretical x-ray scattering spectra for various scattering angles for the carbon plasma. In the non-collective scattering regime an energy bandwidth of $\Delta\omega = 20eV$ is used, whereas in forward scattering a bandwidth of $\Delta\omega = 2eV$ or less is required to resolve the plasmon peak.

In conclusion:

- higher precision measurements necessary for a more accurate spectrally resolved x-ray scattering spectrum and a better signal-to-noise ratio

⇒ future use of FEL/XFEL facilities will allow such high precision measurements to study ion acoustic modes, determine temperature equilibration and thermodynamic properties in high energy density plasmas

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 [10]. Furthermore, the uncertainty regarding the scattered angle and thus the wave number has 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:

$$S_{ee}^{\text{exp}}(k,\omega) = S_{ee}(k,\omega) * g(\omega) * g(k) \quad \text{with gaussian weighting function} \quad g(\omega) = \frac{1}{\sqrt{2\pi}\sigma}\exp\left(-\frac{(\omega-\omega')^2}{2\sigma^2}\right) .$$

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

Ion feature: strongly coupled ions + degenerate electrons [7,11]

a) *ab initio* simulations – DFT-MD

⇒ + full quantum description
  – high computational demand

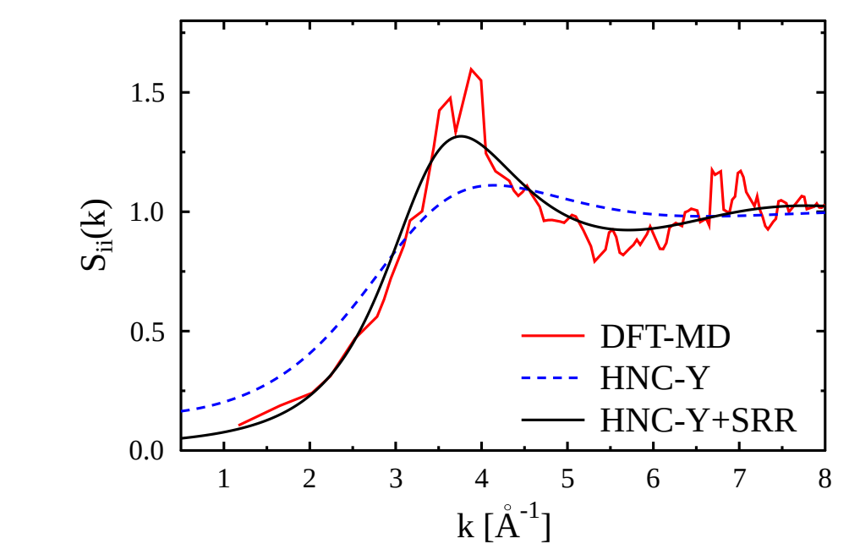
b) classical integral equations (HNC)
  Interaction potential: Yukawa with short range repulsion

$$V_{ab}^{Y+SRR}(r) = \frac{e_ae_b}{4\pi\epsilon_0 r}\mathrm{e}^{-\kappa r} + \left(\frac{c}{r}\right)^4$$

⇒ + high numerical efficiency
  – classical approach

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

Core excitations: negligible for low Z materials

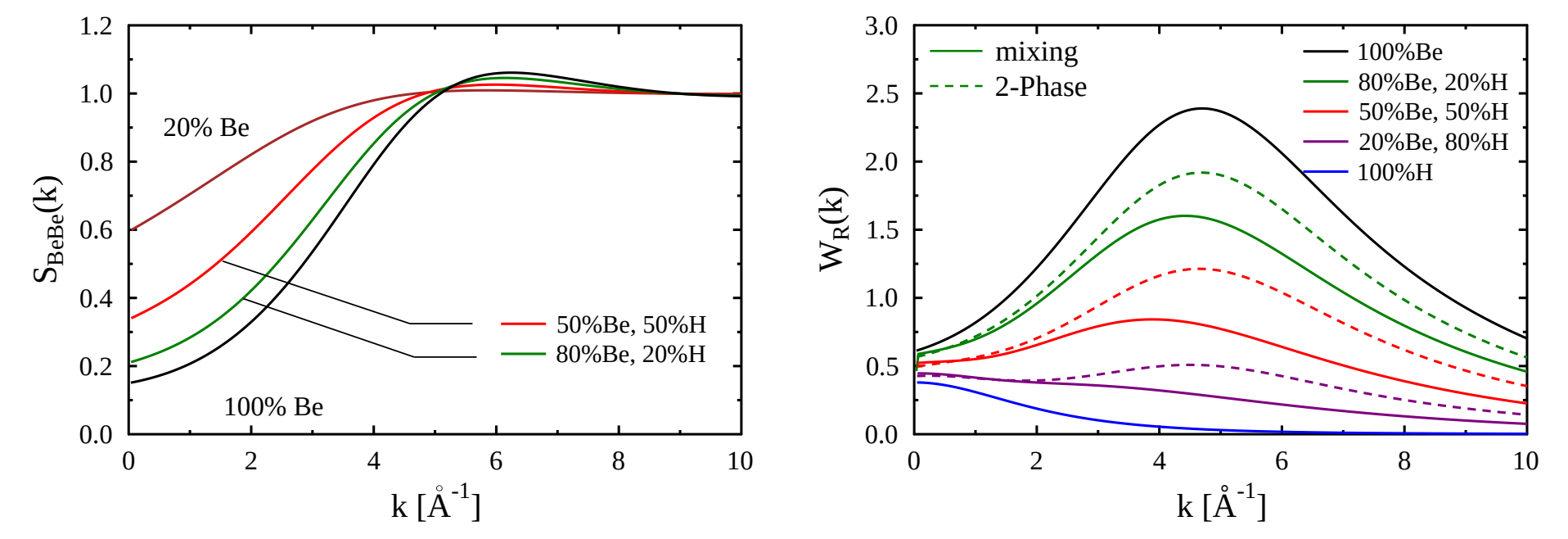


Benchmarking of the static structure factor from calculations applying HNC integral equations by DFT-MD simulations for a silicon plasma with $T = 4.7eV$, $\varrho = 2.33g/cm^3$ and $Z = 4$.

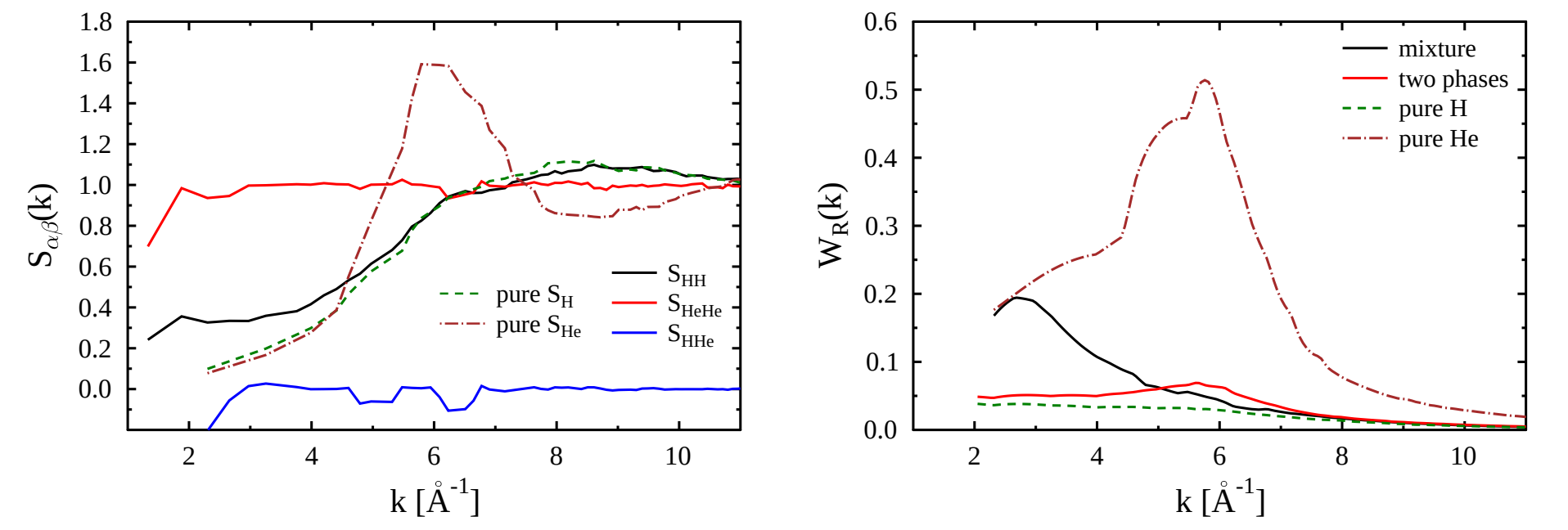
Our x-ray scattering code includes:

- various ionic structure theories, including HNC with different effective interaction potentials
- multi-component effects with consideration of all mutual correlations
- free electron contribution in RPA
- finite spectral bandwidth of detector

X-ray Thomson scattering as a probe for mixtures and mixing



The beryllium structure factors and 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 $n_e \approx 8\times10^{24}cm^{-3}$.



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 $n_e = 10^{24}cm^{-3}$).

BERYLLIUM-HYDROGEN-MIXTURES

- 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

- He-H mixtures naturally occur in astrophysical objects, e.g. giant gas planets (Jupiter)
- 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