X-ray Thomson Scattering in Non-Equilibrium

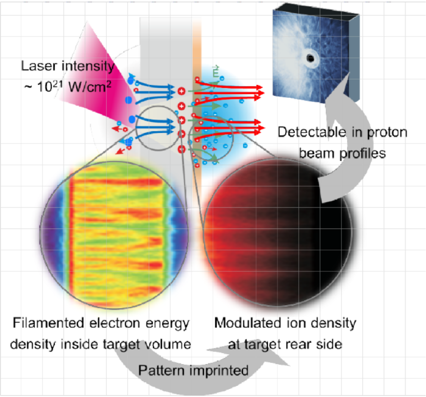
J. Vorberger

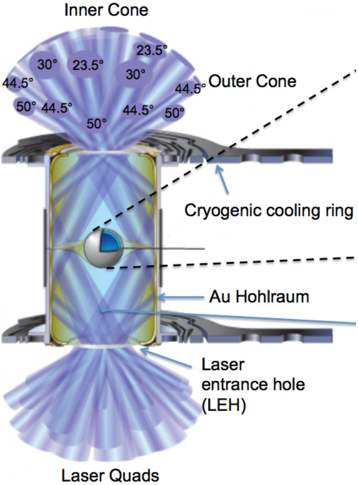
Institute of Radiation Physics, Helmholtz-Zentrum Dresden-Rossendorf e.V.

EUCALL Joint Foresight Topical Workshop on Theory and Simulation of Photon-Matter Interaction

2nd July 2018

Laser-matter interaction





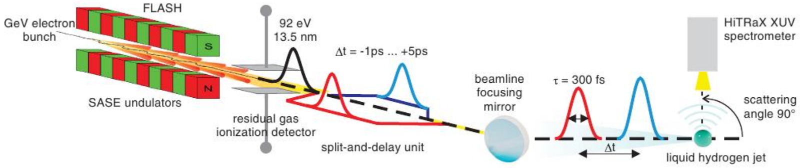
high power lasers

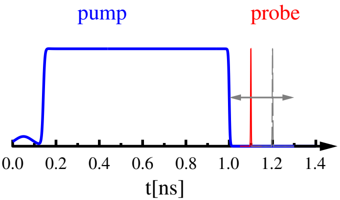
high energy lasers

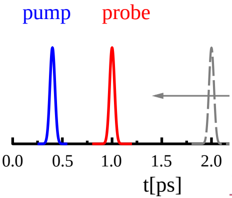
copyright Th. Kluge, HZDR

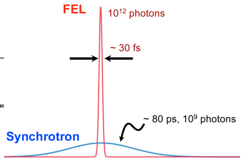
copyright NIF, LLNL

Ultrafast excitation and diagnostics







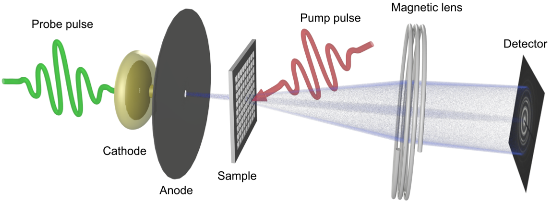


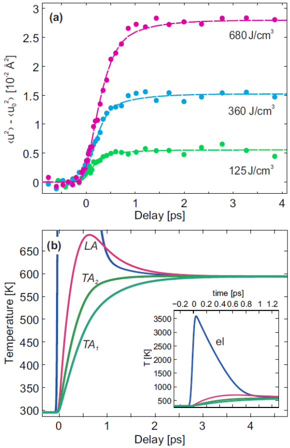
high-energy laser for compression or laser generated ions for heating

XFEL pulses are $\leq$ 10 fs long & are able to probe on these time scales

Zastrau et al. PRL (2014), PRE (2014), R. Neutze

Measuring temperature relaxation in laser heated aluminium by electron scattering



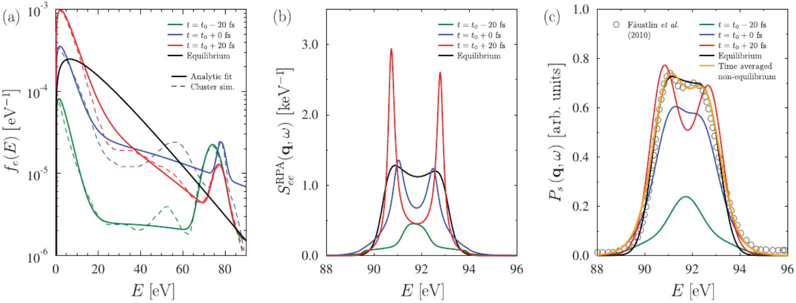


- **Cannot** use Born-Oppenheimer for energy transfer rates!
- How about Born-Oppenheimer in the modelling of the scattering signal (Chihara)?

L. Waldecker, R. Bertoni, R. Ernstorfer, J. Vorberger, PRX (2016)

HZDR

Evolution of the structure of hydrogen under XFEL radiation



- Femto-second lasers and XFELs allow to study initial electron relaxation and subsequent temperature & charge relaxation
- X-ray scattering theory (for diagnostics) needs to take into account **non-equilibrium** and **correlations**!

D.A. Chapman et al., PRL (2011), N. Medvedev et al., PRL (2011), R.R. Fäustlin et al., PRL (2010), Sperling et al. PRL (2015)

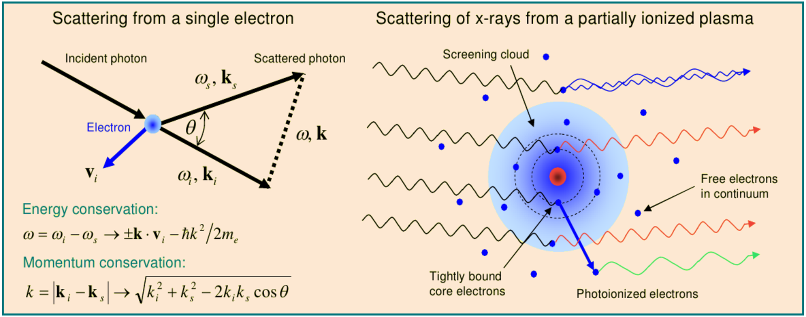
Goals & Overview

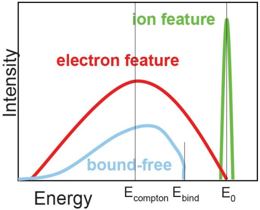
We want to detect and diagnose

- inhomogeneities
- anisotropies
- non-equilibrium distribution functions
- two-temperature systems
- excitation & relaxation phenomena

To do this, we need a theory for x-ray scattering (the electronic structure) including different species in non-equilibrium featuring correlations and quantum effects.

Calculating the x-ray scattering signal in equilibrium



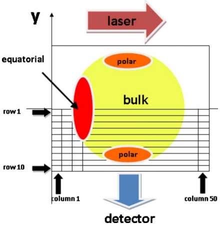


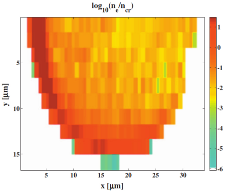
Chihara J Phys F (1987), Chihara J Phys Cond Matt (2000), Wünsch et al. EPL (2011)

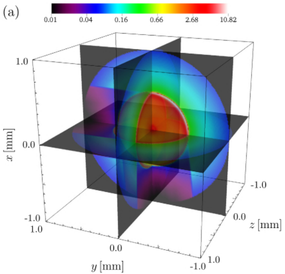
$$\text{scattered intensity} \sim S_{ee}^{tot}(k,\omega) = \bar{Z} S_{ee}^{0}(k,\omega)$$
$$+ \sum_{ab} \sqrt{x_a x_b}[f_a(k)+q_a(k)][f_b(k)+q_b(k)]S_{ab}(k,\omega)$$
$$+ \sum_{a} Z_a^c x_a \int \tilde{S}_{ce}(k,\omega-\omega')S_s(k,\omega')d\omega'$$

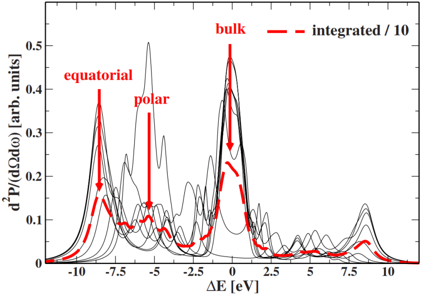
$$S_{ea}(k,\omega) = [f_a(k)+q_a(k)]S_{aa}(k,\omega) + [f_b(k)+q_b(k)]S_{ab}(k,\omega)$$

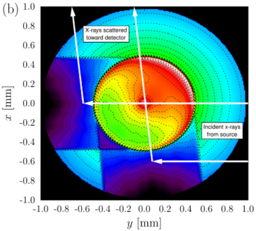
Inhomogeneous systems – hydrogen









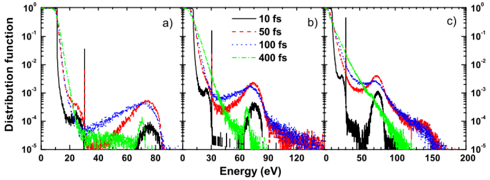


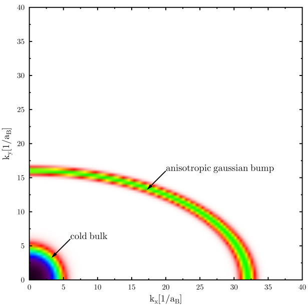
add up small volumes of different *n* and *T*, account for attenuation

Thiele et al. PRE (2010), Chapman et al. PoP (2014)

Non-equilibrium distribution functions

tails of fast thermalized electrons, Auger electrons, laser accelerated electrons giving anisotropies...





top: *electron distribution in laser illuminated aluminium*

right: *model anisotropic electron distribution*

there is no viable first principle quantum simulation to use for such situations.

Fäustlin et al. PRL (2011), Medvedev et al. PRL (2011), Sperling et al. PRL (2015)

Non-equilibrium structure theory

$$S_{ee}(\mathrm{k},\omega;t)=\frac{i}{2\pi}L^{>}_{ee}(\mathrm{k},\omega;t)=\frac{1}{2\pi}\frac{1}{\Omega}\int_{-\infty}^{\infty}d\tau\langle\delta\rho(\mathrm{k},\tau,t)\delta\rho(\mathrm{k},0,t)\rangle e^{i\omega\tau}$$

This becomes $S_{ee}(\mathrm{k},\omega)=1/\pi[1+n_B(\omega)]\mathrm{Im}L^{R}_{ee}(\mathrm{k},\omega)$ in equilibrium

$$L_{ee}(t_1,t_2)=\Pi_{ee}(t_1,t_2)+\sum_{cd}\int_{\mathcal{C}}d\bar{t}\,\Pi_{ec}(t_1,\bar{t})V_{cd}L_{de}(\bar{t},t_2)$$

real time Green's functions approach, local approximation, two–fluid picture

$$L^{>}_{ee}(\mathbf{k},\omega;t)=\frac{\mathcal{L}^{>}_{e}(\mathbf{k},\omega;t)+|\mathcal{L}^{R}_{e}(\mathbf{k},\omega;t)|^2V^2_{ei}(k)\,\mathcal{L}^{>}_{i}(\mathbf{k},\omega;t)}{|1-V_{ie}(k)\,\mathcal{L}^{R}_{e}(\mathbf{k},\omega;t)V_{ei}(k)\,\mathcal{L}^{R}_{i}(\mathbf{k},\omega;t)|^2}$$

$$\mathcal{L}^{>}_{a}(\mathbf{k},\omega;t)=\frac{\Pi^{>}_{a}(\mathbf{k},\omega;t)}{|1-V_{aa}\Pi^{R}_{a}(\mathbf{k},\omega;t)\,|^2}\,,\;\mathcal{L}^{R}_{a}(\mathbf{k},\omega;t)=\frac{\Pi^{R}_{a}(\mathbf{k},\omega;t)}{1-V_{aa}\Pi^{R}_{a}(\mathbf{k},\omega;t)}$$

Chapman et al. PRL (2011)), Vorberger & Chapman PRE (2018)

Dynamic local field corrections in non-equilibrium

Exact connection of density response and polarisation function

$$\mathcal{L}_a(t_1, t_2) = \Pi_a(t_1, t_2) + \int_{\mathcal{C}} d\bar{t}\; \Pi_a(t_1, \bar{t}) V_{aa} \mathcal{L}_a(\bar{t}, t_2)\,.$$

Ansatz = definition of non-equilibrium LFCs

$$\mathcal{L}_a(t_1, t_2) = \mathcal{L}^0_a(t_1, t_2) + \int_{\mathcal{C}} d\bar{t} d\bar{\bar{t}}\; \mathcal{L}^0_a(t_1, \bar{t}) V_{aa} [1 - G_{aa}(\bar{t}, \bar{\bar{t}})] \mathcal{L}_a(\bar{\bar{t}}, t_2)\,.$$

Two independent LFC quantities needed

$$G^R_{aa}(\mathbf{k}, \omega; t) = \frac{1}{V_{aa}(k)} \left\{ \frac{1}{\Pi^R_a(\mathbf{k}, \omega; t)} - \frac{1}{\mathcal{L}^{0\,R}_a(\mathbf{k}, \omega; t)} \right\}$$

$$G^>_{aa}(\mathbf{k}, \omega; t) = \frac{1}{V_{aa}(k)} \left\{ \frac{\mathcal{L}^{0\,>}_a(\mathbf{k}, \omega; t)}{|\mathcal{L}^{0\,R}_a(\mathbf{k}, \omega; t)|^2} - \frac{\Pi_a{}^>(\mathbf{k}, \omega; t)}{|\Pi_a{}^R(\mathbf{k}, \omega; t)|^2} \right\}$$

HZDR

Chihara decomposition in non-equilibrium I

sketch of formulas after going from Keldysh contour to physical time axis

connection of electron-ion and ion-ion structure

$$L_{ei}^{R/A}(\mathbf{k},\omega;t) = \rho^{R/A}(\mathbf{k},\omega;t) L_{ii}^{R/A}(\mathbf{k},\omega;t)$$

$$S_{ei}(\mathbf{k},\omega;t) = \frac{i}{2\pi}\rho^{>}(\mathbf{k},\omega;t) L_{ii}^{A}(\mathbf{k},\omega;t) + \rho^{R}(\mathbf{k},\omega;t) S_{ii}(\mathbf{k},\omega;t)$$

Chihara decomposition

$$L_{ee}^{R/A}(\mathbf{k},\omega;t) = L_{ee}^{free\,R/A}(\mathbf{k},\omega;t) + \rho^{R/A}(\mathbf{k},\omega;t) L_{ii}^{R/A}(\mathbf{k},\omega;t)\, \rho^{R/A}(\mathbf{k},\omega;t)$$

$$L_{ee}^{>} \quad = \quad L_{ee}^{free>} + (L_{ei}^{A} + L_{ei}^{R})\rho^{>} + |\rho^{R}|^2 L_{ii}^{>}$$

independent generalized screening clouds ρ^R and $\rho^>$ due to non-equilibrium conditions!

Member of the Helmholtz Association
J. Vorberger | Institute of Radiation Physics | http://www.hzdr.de

Chihara decomposition in non-equilibrium II $(\Pi_{ei} = 0)$

$$S_{ee}(\mathbf{k},\omega;t) = S_{ee}^{free}(\mathbf{k},\omega;t) + \frac{i}{2\pi}\left[L_{ei}^{A}(\mathbf{k},\omega;t)+L_{ei}^{R}(\mathbf{k},\omega;t)\right]\rho^{>}(\mathbf{k},\omega;t) + |\rho^{R}(\mathbf{k},\omega;t)|^{2} S_{ii}(\mathbf{k},\omega;t)$$

total electron structure = free electron structure + non-Born-Oppenheimer part + screening cloud × ion structure

$$\rho^{R/A}(\mathbf{k},\omega;t) = V_{ei}(k)\,\mathcal{L}_{e}^{R/A}(\mathbf{k},\omega;t) \qquad \rho^{>}(\mathbf{k},\omega;t) = V_{ei}(k)\,\mathcal{L}_{e}^{>}(\mathbf{k},\omega;t)$$

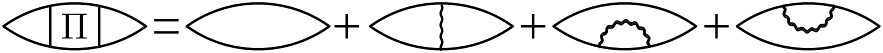
$$L_{ee}^{free>}(\mathbf{k},\omega;t) = \mathcal{L}_{e}^{>}(\mathbf{k},\omega;t) = \frac{\Pi_{e}^{>}(\mathbf{k},\omega;t)}{|1 - V_{ee}(k)\,\Pi_{e}^{R}(\mathbf{k},\omega;t)\,|^{2}}$$

A fully generalized version including all Π_{ei} terms is available!

Exchange and correlation in non-equilibrium

only pure species contributions for polarization function in first order

$$\Pi_a(\mathbf{k},\omega;t) = \Pi_a^{RPA}(\mathbf{k},\omega;t) + \Pi_a^{V}(\mathbf{k},\omega;t) + \Pi_a^{S_1}(\mathbf{k},\omega;t) + \Pi_a^{S_2}(\mathbf{k},\omega;t)$$



- evaluate correlation functions $\Pi^{\gtrless}$ for Vertex & Self-energy term
- use Kramers-Kronig relation for retarded quantities

$$\Pi_a^R(p,\omega;t) = i\int_{-\infty}^{\infty}\frac{d\omega'}{2\pi}\,\frac{\Pi_a^{>}(p,\omega;t') - \Pi_a^{<}(p,\omega;t')}{\omega-\omega'+i\varepsilon}\,.$$

Evaluation of the self-energy & vertex terms

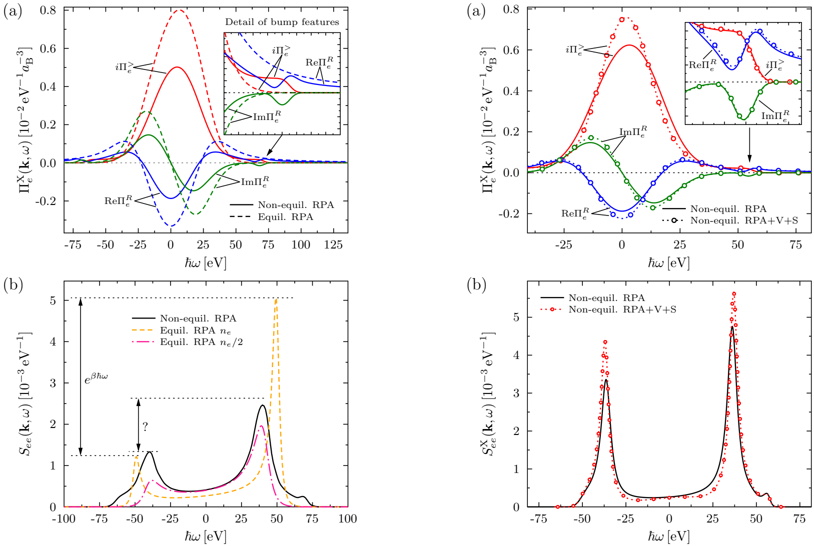
static screening + application of Langreth-Wilkins rules gives

$$
\begin{aligned}
i\Pi_a^{V\gtrless}(\mathbf{k},\omega;t) &= 4\pi(i\hbar)^2\,\mathcal{P}\int\frac{d\mathbf{p}d\mathbf{q}}{(2\pi\hbar)^6}\,V_{aa}^S(\mathbf{p}-\mathbf{q}) \\
&\quad\times f_a^{\lessgtr}(\mathbf{p},t)f_a^{\gtrless}(\mathbf{p}+\mathbf{k},t)\Big[f_a(\mathbf{q},t)-f_a(\mathbf{q}+\mathbf{k},t)\Big] \\
&\quad\times\frac{\delta\Big(\hbar\omega+E_a(\mathbf{p})-E_a(\mathbf{p}+\mathbf{k})\Big)}{\hbar\omega+E_a(\mathbf{q})-E_a(\mathbf{q}+\mathbf{k})}\,,
\end{aligned}
$$

$$
\begin{aligned}
i\Pi_a^{S\gtrless}(\mathbf{k},\omega;t) &= 2\pi\hbar\int\frac{d\mathbf{p}}{(2\pi\hbar)^3}\,\Sigma_a(p)f_a^{\gtrless}(\hbar\omega+E_a(\mathbf{p}-\mathbf{k}),t)f_a^{\lessgtr}(E_a(\mathbf{p}-\mathbf{k}),t) \\
&\quad\times\delta'\Big(\hbar\omega+E_a(\mathbf{p}-\mathbf{k})-E_a(p,t)\Big) \\
&+\;2\pi\hbar\int\frac{d\mathbf{p}}{(2\pi\hbar)^3}\,\Sigma_a(p)f_a^{\gtrless}(E_a(\mathbf{p}+\mathbf{k}),t)f_a^{\lessgtr}(E_a(\mathbf{p}+\mathbf{k})-\hbar\omega,t) \\
&\quad\times\delta'\Big(\hbar\omega+E_a(p,t)-E_a(\mathbf{p}+\mathbf{k})\Big)
\end{aligned}
$$

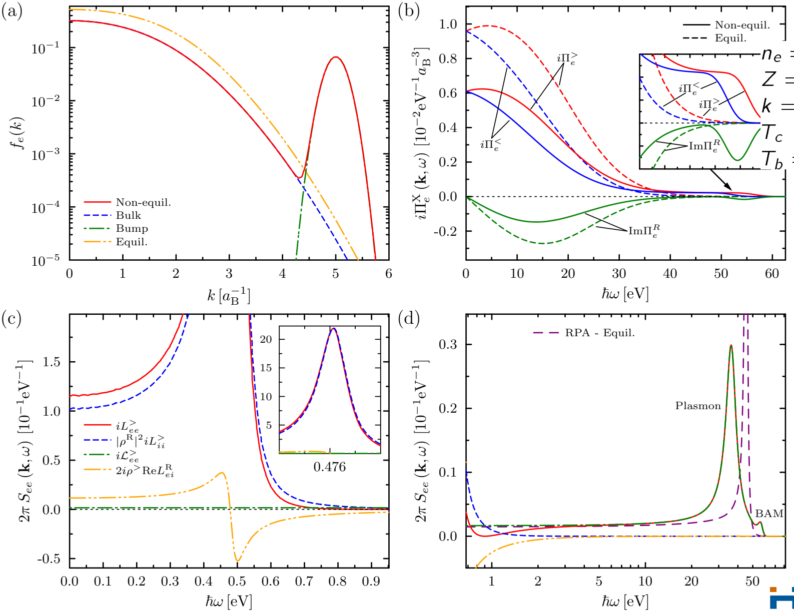
$$
\begin{aligned}
E_a(p) &= \hbar^2p^2/2m_a \\
\Sigma_a(p,t) &= -\hbar\int\frac{d\mathbf{q}}{(2\pi\hbar)^3}V_{aa}^S(\mathbf{p}+\mathbf{q})f_a(\mathbf{q},t)
\end{aligned}
\qquad
\begin{aligned}
f_a^> &= f_a-1 \\
f_a^< &= f_a
\end{aligned}
$$

Structure in a non-equilibrium electron gas



$n_e = 10^{24}\mathrm{cm}^{-3}$, $n_e/2$ @ $T_c = 4\times10^5\,\mathrm{K}$, $n_e/2$ in bump around $p_b = 5\hbar/a_B$, $k = 0.5/a_B$ (left), $k = 0.4/a_B$ (right)

Structure in laser pumped hydrogen



$n_e = 10^{24}\,cm^{-3}$

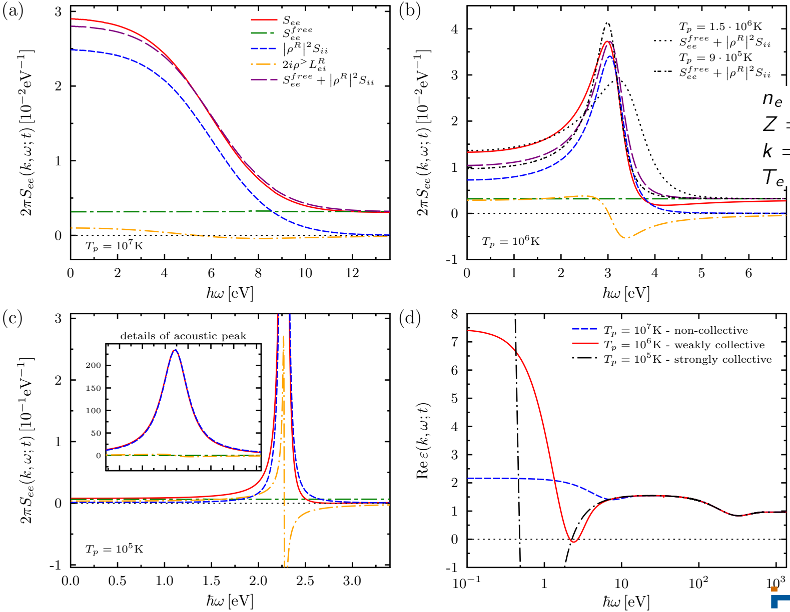
$Z = 1$

$k = 0.4/a_B$

$T_c = 4\times 10^5\,\mathrm{K}$

$T_b = 10^4\,\mathrm{K}$

Structure in two-temperature hydrogen



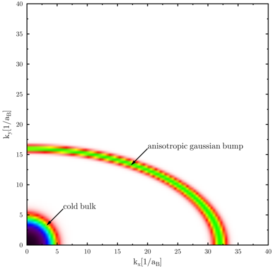
$n_e = 10^{25}\,cm^{-3}$

$Z = 1$

$k = 1/a_B$

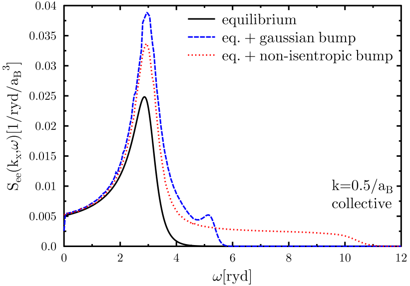
$T_e = 10^7$K

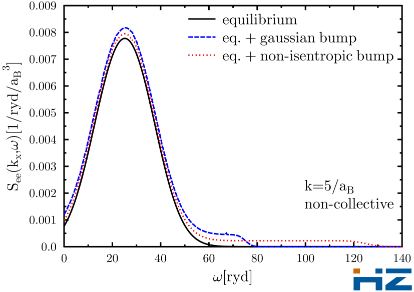
Non-equilibrium & anisotropy in an electron gas



$$f(k_x,k_y,k_z) = \frac{1}{e^{(k_x^2+k_y^2+k_z^2-\mu)/k_B/T}+1} + Ae^{-B\left(\sqrt{(k_x/c_x)^2+(k_y/c_y)^2+(k_z/c_z)^2}-D\right)^2}$$

anisotropic features of the electron distribution should be very well visible in scattering spectrum





Summary & Outlook

- exact formalism to calculate structure in non-equilibrium on the basis of the Wigner distribution function
- definition of local field corrections in non-equilibrium
- account of exchange & correlation in non-equilibrium (in weak coupling approximation)
- decomposition of total electron structure factor similarly to equilibrium (Chihara)

- same formalism gives exact energy transfer rate in full non-equilibrium or two-temperature systems

HZDR