Probing the Transient Properties of Warm Dense Matter

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centre for fusion, space and astrophysics

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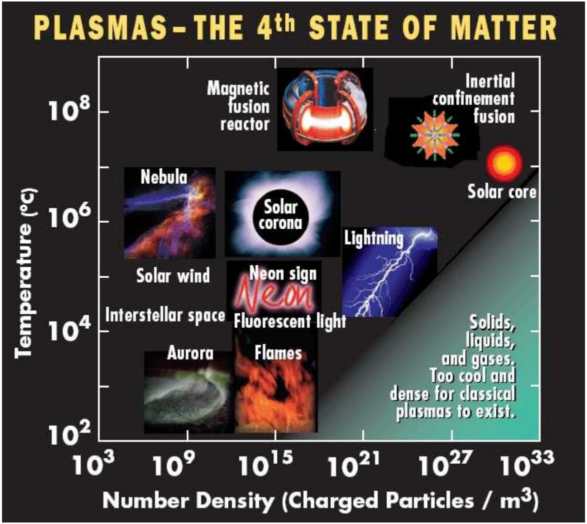
Introduction

to Warm Dense Matter

A Universe of Plasmas – only a few Dense Systems?

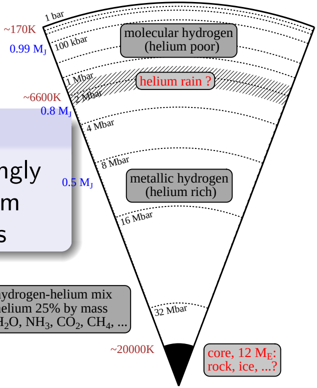


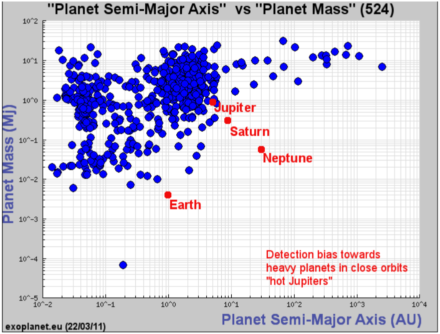
from NRC, *Frontiers in High Energy Density Physics: The X-Games of Contemporary Physics*

Motivation: Solar and Extrasolar Giant Gas Planets

> 520 Planets are known

inner structure and evolution strongly depend on EOS of hydrogen-helium mixtures under extreme conditions





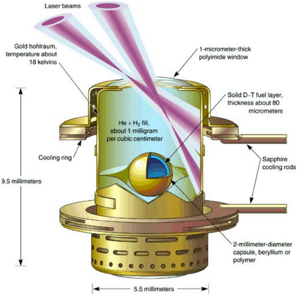
Physics to be included:

- Gas clouds to high-density plasmas
- Ionization equilibrium
- Mixing behavior of H, He, metals
- High pressure chemistry

$\Rightarrow$ **Test planetary evolution models**

Physics for Indirectly Driven ICF-Targets

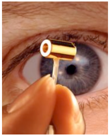
Indirect drive target



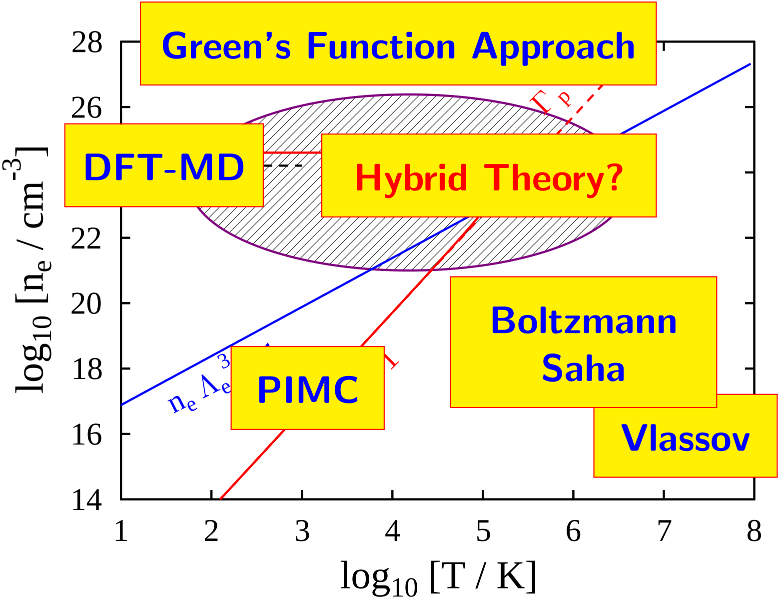
Related exciting physics

- Energy coupling into capsule (scattering)
- Energy absorption by the walls
- Equation of state for various materials
- Radiation transport and hydrodynamics
- Burn physics: kinetics of α-particles
- Probing the HED states created

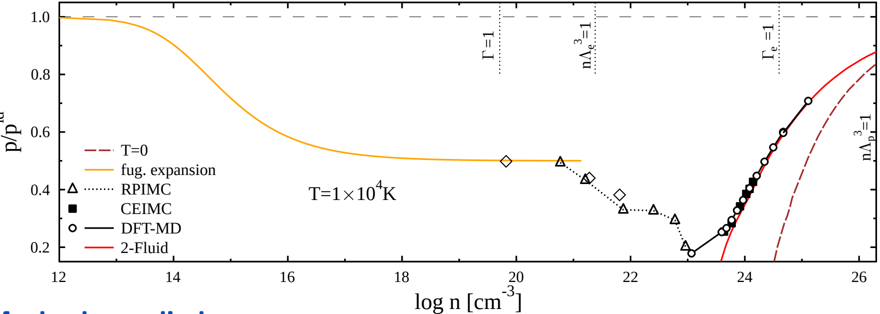
Full ignition campaign is running since 2010 making progress but still facing problems as well



Theoretical Approaches for Warm Dense Matter



Mixture of Approaches required for WDM



Methods applied:

- Fugacity expansion for weakly coupled plasmas and gases
- Density functional molecular dynamics simulations with hydrogen basis set
- Path-integral Monte Carlo simulations for weakly degenerate plasmas
- Density functional molecular dynamics simulations and coupled electron-ion Monte Carlo simulations for high degeneracy
- 2-Fluid Model and $T = 0$ limiting law for very high densities

Riemann et al. Physica 1995, Militzer PRE 2001/2002, Morales PRE 2010, Vorberger et al. PRB 2007, 2011

Important Relaxation Processes in Warm Dense Matter

Relaxation Processes after Strong Excitations

momentum relaxation electrons: $\sim 1\,\mathrm{fs}$

ionisation equilibrium; rate equations: $\sim 1\,\mathrm{ps}$

momentum relaxation ions: $\sim 100\,\mathrm{fs}$

temperature relaxation $\sim 10\,\mathrm{ps}$

- Sequence of subsequent relaxation processes towards equilibrium
- **Relaxation stages might overlap and influence each other**
- **There are no first principle methods available**
- All relaxation times are only approximate and under discussion
- Strong indications of much slower temperature relaxation than predicted
  Celliers et al. (1992), Riley et al. (2000), White et al. (submitted)

Probing Electron Dynamics by X-Ray Thomson Scattering

Standard Theory for X-Ray Scattering

Light scattered from strongly coupled, partially ionized plasmas

$$P(\theta,\omega) \sim S_{ee}^{tot}(k,\omega) = |f_i(k)+q(k)|^2\, S_{ii}(k,\omega) + Z_f\, S_{ee}^0(k,\omega) + Z_b \int d\omega'\, \tilde{S}^{ce}(k,\omega-\omega') S_s(k,\omega')$$

Chihara (1987), (2000); multiple ion species: Wünsch et al., (2011)

1^{st} term: **Ion feature** (electrons co-moving with the ions)

2^{nd} term: **Electron feature** (free electrons)
calculated via the density-density response function (FDT)

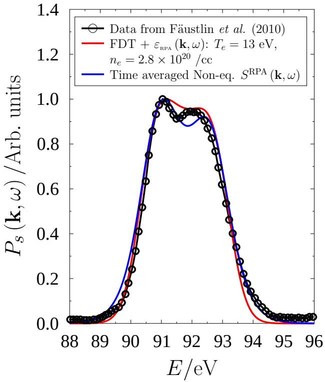
$$S_{ee}(k,\omega) = \frac{\hbar}{\pi n_e} \frac{1}{1-\exp(-\beta_e \hbar\omega)} \,\mathrm{Im}\chi_{ee}(k,\omega)$$

3^{rd} term: **Inelastic Raman scattering** (unimportant for light elements)

Extension to Systems with Nonequilibrium Electrons

Motivation for Nonequilibrium Physics

- Study relaxation processes directly (collisionality, collective response)
- Ultra fast pumping and probing is possible with news sources (FEL)
- Persistent heating might be ongoing in "equilibrium" experiments



Power spectrum from FEL heated H.

Example for scattering of VUV radiation from FLASH

- Systems is heated and probed by 92 eV photons from FEL (Fäustlin et al., PRL (2010))
- Strongly driven system due to high photon numbers
- Data much less noisy due to averaging over 1500 shots

⇒ **Nonequilibrium analysis needed?**

Electron Feature for Nonequilibrium Plasmas

Analysis using nonequilibrium FDT and electron distributions

- Fluctuation-dissipation theorem used for equilibrium fits

$$S_{ee}(k,\omega) = \frac{\hbar}{\pi n_e} \frac{1}{1-\exp(-\beta_e \hbar\omega)} \, \mathrm{Im}\chi_{ee}(k,\omega)$$

- Fluctuation-dissipation theorem in nonequilibrium

$$S_{ee}(k,\omega) = \frac{i\hbar}{2\pi n_e} \frac{\Pi_{ee}^{>}(k,\omega)}{|\varepsilon(k,\omega)|^2} \overset{\mathrm{RPA}}{=} \frac{S_{ee}^0(k,\omega)}{|\varepsilon(k,\omega)|^2}$$

- Ideal structure factor given by distribution functions

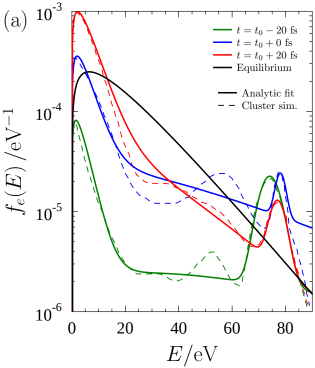
$$S_{ee}^0 = \frac{2\hbar}{n_e} \int \frac{d\mathbf{p}}{(2\pi\hbar)^3} \, \delta\Big(E(\mathbf{p}+\mathbf{q} - E(p) - \hbar\omega \Big) \Big[1 - f_e(\mathbf{p}+\mathbf{q}) \Big] f_e(p)$$

- Screening function in RPA is also given by distribution functions
- Mode spectrum is modified for nonequilibrium situations

Application to FLASH-Data from Self-Scattering

Analysis using nonequilibrium FDT and distributions

- Distributions functions from cluster simulations (courtesy B. Ziaja)
- Distributions modeled by analytical form of a bump on hot tail



Bump on hot tail model (BOHT)

$$f_e(p) = \alpha f(p, T_c) + \beta f(p, T_h) + \gamma e^{-\frac{(p-p_0)^2}{p_b^2}}$$

Model describes:

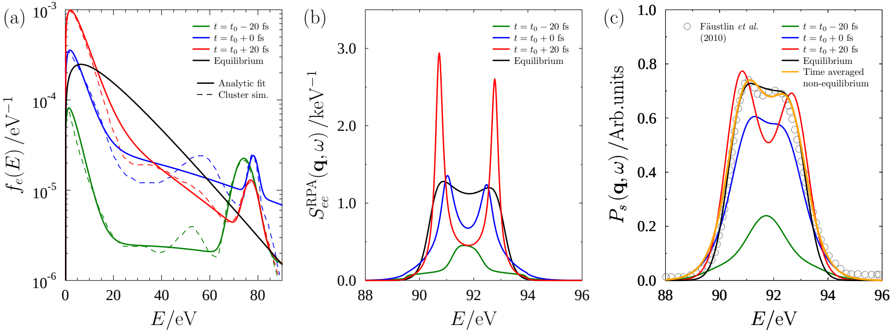
- Bulk electrons with "cold" distribution (Fermi distribution)
- Hot tail of energetic electrons (Boltzmann distribution)
- Electrons pumped to specific energy (Gaussian peak around excitation)

⇒ **Essential electron dynamics captured by BOHT model**

⇒ **2 stage relaxation from state excited by the FEL radiation**

Power Spectrum for Self-Scattering Experiment

Analysis using nonequilibrium FDT and BOHT model



Chapman & Gericke, PRL (2011)

$\Rightarrow$ Nonequilibrium analysis yield good agreement with experiments,
BUT plasma parameters evolve + differ from equilibrium fit

$\Rightarrow$ **Nonequilibrium dynamics can, in principle, being tested by XRTS**

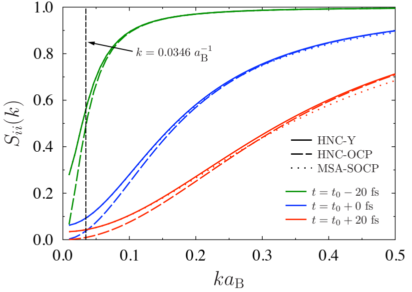
“Missing” Elastic (Ion) Feature in the Exp. Data

Full spectrum should contain an elastic scattering peak

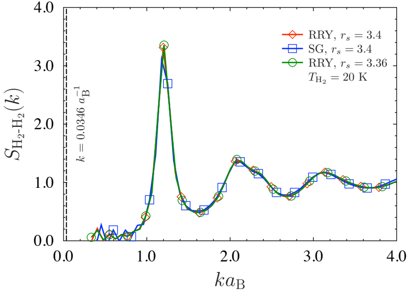
$$P(\theta,\omega) \sim S_{ee}^{tot}(k,\omega) = |f_i(k) + q(k)|^2 \, S_{ii}(k)\delta(\omega) + Z_f S_{ee}^0(k,\omega)$$

⇒ **What is the static ion structure in the system?**

Coulomb interactions (T=20 K)



Original hydrogen liquid



⇒ **Ions retain their structure from the cold liquid during the pulse**

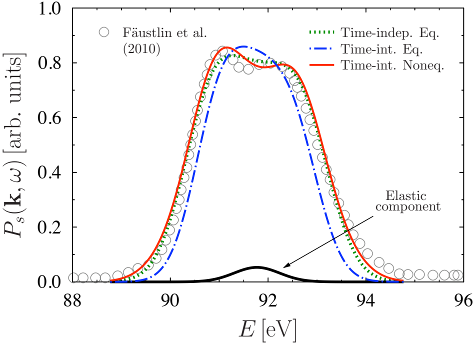
⇒ **The ion-ion structure factor for the k-value probed is very small**

"Missing" Elastic (Ion) Feature in the Exp. Data

Full spectrum should contain an elastic scattering peak

$$P(\theta,\omega) \sim S_{ee}^{tot}(k,\omega) = |f_i(k) + q(k)|^2 \, S_{ii}(k)\delta(\omega) + Z_f \, S_{ee}^0(k,\omega)$$

Applying initial hydrogen structure to scattering spectrum



Full power spectrum for FEL driven hydrogen (self-scattering)

- **Excellent agreement with experimental data**
- Best fit with data for:
  $S_{ii}(k_{\mathrm{probed}}) = 0.04$
- Ionic correlations are not present on fs-time scales
- We need to consider yet another relaxation process: **Build up of Ion Correlations**

Ion Dynamics in Dense Matter

Build-up of Ionic Correlations & Structure

Models for a theoretical description

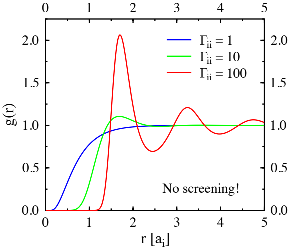
- Quantum kinetic approach: generalized Kadanoff-Baym equations
  Semkat *et al.*, PRE (1999)
- Direct (classical) molecular dynamics simulation Murillo, PRL (2001)
- Use of energy conservation ⇒ final state
  Gericke *et al.*, J.Phys.A (2003)

$$\begin{aligned} E_i^{total}(0^+) &= E_i^{kin}(0^+) + U_{ii}^{corr}(0^+) \\ &= E_i^{kin}(\infty) + U_{ii}^{corr}(\infty) \\ &= E_i^{total}(\infty) \end{aligned}$$

$$\text{with } U_{ii}^{corr}(t) = \frac{n_i}{2}\int d^3\mathbf{r}\, g(r,t)\, V_{ii}(r)$$

$$g(r,t) = \frac{1}{n\,N}\sum_{i\neq j}\langle\delta(r - r_{ij}(t))\rangle$$

Pair distribution function



Effects of Build-up of Correlations & Structure

Know phase changes due to new ion-ion forces

- Structural transitions between 2 solid phases
- Ultra fast, nonthermal melting of semiconductors
  Break of bonds or ionisation driven transition?
- Gas-liquid or gas-plasma transition
  $\Rightarrow$ Creation of structure due to strong forces

Heating in ultra-cold plasmas

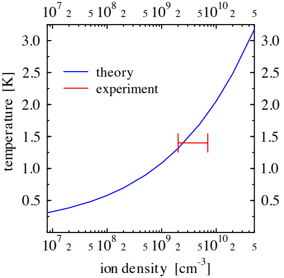
- No initial correlations (gas)
- Almost no kinetic energy at $t = 0$
  (gas temperature $\approx 1\,\mu$K)

$\Rightarrow$ $U_{ii}^{corr}(0) = 0$ and $E_i^{kin}(0) = 0$

$\Rightarrow$ Effective coupling strength:

$$\Gamma_{ii}^{eff}(t) = \frac{\left|U_{ii}^{corr}(t)\right|}{E_i^{kin}(t)} = \frac{\left|U_{ii}^{corr}(t)\right|}{E_i^{kin}(0) + \left|U_{ii}^{corr}(t)\right|} \rightarrow 1$$

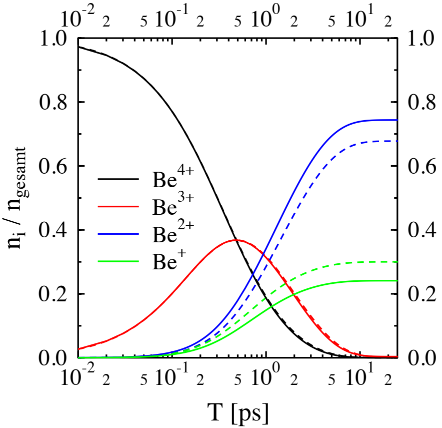
Final ion temperature



Exp: Killian et al., PRL (2005)

Ionisation Kinetics

Time-Dependent Charge State Distribution



Relaxation of a beryllium plasma
(courtesy of G.K. Grubert)

Technique applied:

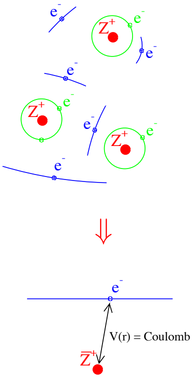
- Numerical solution of a system of rate equations
- Correlation effects in effective ionisation energy $I_{bound} = |E_0| + \Delta_e + \Delta_p$

Problems:

- Effective ionisation energy
- Inner shell ionisations
- Rate coefficients assume equilibrium distributions neglecting possible hot tails $\Rightarrow \alpha = \alpha(T_c) + \alpha(T_h)$?

Electron-Ion Energy Equilibration and 2-Temperature EOS

Electron-Ion Energy Transfer: LS Approach



Approximations applied:

- Binary collisions with straight line trajectories
- Pure Coulomb interactions with hard cut-offs

$$\dot{T}_e = (T_i - T_e)\frac{8\sqrt{2\pi}Z_i^2 e^4 \ln\lambda_c}{3m_e m_i}\left(\frac{T_e}{m_e}+\frac{T_i}{m_i}\right)^{-3/2}$$

Landau (1936), Spitzer (1967)

- Corrections due to hyperbolic orbits yield: $\ln\lambda_c \rightarrow \frac{1}{2}\ln(1+\lambda_c^2)$

- Brysk formula (ext. to degenerate plasmas)

What is 'λ_C'?

$$\dot{T}_e = \frac{8m_e Z_i^2 e^4 \ln\lambda_c}{3\pi m_i \hbar^3}\left[\exp(-\mu_e/k_B T_e + 1)\right]$$

Brysk, Plasma Phys. (1974)

. . . more Advanced Theoretical Models

- Strong binary collision within quantum kinetic theory

$$E_{e\to i}^{trans} = \frac{1}{2\pi\hbar^3} \frac{n_e \Lambda_e^3}{m_i m_r} \int_0^\infty dk\, k^5\, Q^T(k) \exp\left(-\frac{k^2}{2m_e k_B T_e}\right)$$

Gericke *et al.*, PRE (2002)

- Energy transfer through coupled collective modes

$$E_{e\to i}^{trans} = 4\hbar \int_0^\infty \frac{d\omega}{2\pi}\,\omega \int \frac{d^3\mathbf{k}}{(2\pi)^3} \left|U_{ei}^S(k)\right|^2 \frac{\Delta N_{ei}\, \chi_e''(\omega,k)\chi_i''(\omega,k)}{1 - V_{ei}(k)\chi_e(\omega,k)\chi_i(\omega,k)}$$

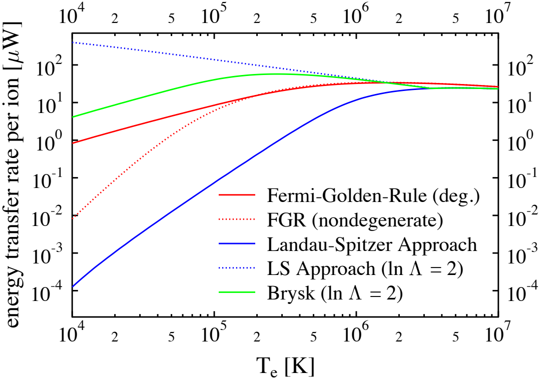
Dharma-wardana & Perrot, PRE (1998)

- Fermi's-Golden-Rule approach (simplest model with collective modes)

$$E_{e\to i}^{trans} = 4\hbar \int_0^\infty \frac{d\omega}{2\pi}\,\omega \int \frac{d^3\mathbf{k}}{(2\pi)^3} \left|U_{ei}^S(k)\right|^2 \Delta N_{ei}\, \chi_e''(\omega,k)\, \chi_i''(\omega,k)$$

Electron-Ion Energy Transfer: Degenerate Electrons

Results for the energy transfer rates



Energy transfer rates for silicon plasmas with $Z_i = 4$, $n_i = 1.17 \times 10^{23}\,\mathrm{cm}^{-3}$, and $T_i = 10^3\,\mathrm{K}$. Parameters like Celliers et al., PRL (1992)

Insights gained

- LS approach fails for degenerate plasmas
- Brysk formula describe the rates only qualitatively
- Full **FGR gives longer relaxation times** than Brysk formula
- **USE FGR** as the easiest theory!

Temperature Relaxation: Defining the Subsystems

How can one define an ion and an electron subsystems?

- We want to solve a system of equations like
$$\frac{\partial}{\partial t}E_e \stackrel{\text{Def}}{=} \frac{\partial}{\partial t}\left(E_e^{kin} + U_e^{cor}\right) = \mathbf{Z_{ei}} = -\frac{\partial}{\partial t}E_i \stackrel{\text{Def}}{=} -\frac{\partial}{\partial t}\left(E_i^{kin} + U_i^{cor}\right)$$
- However, the total energy of the system is given by
$$E = E_e^{kin} + U_{ee}^{cor} + E_i^{kin} + U_{ii}^{cor} + U_{ei}^{cor}$$

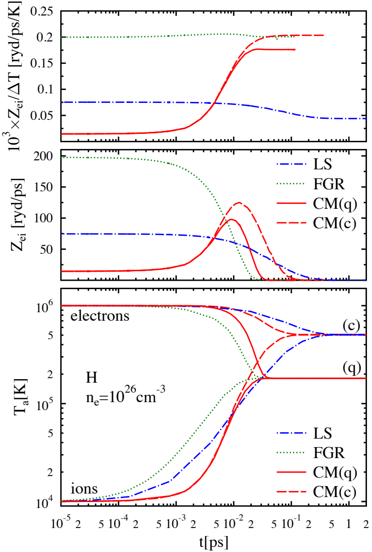
How should we to treat the cross term?

$\Rightarrow$ Split total energy according to (Gericke et al., J. Phys. A (2006)):
$$E_a = E_a^{kin} + U_a^{cor} \qquad \text{with} \qquad U_a^{cor} = U_{aa}^{cor} + \frac{1}{2}U_{ab}^{cor}$$

Questions remaining for temperature relaxation approach:

1. What is the (quasi)-equation of state for a 2-temperature system?
2. How can the energy transfer rates be obtained? Correlation effects?

Temperature Relaxation in Nonideal Plasmas



Time-dependent T_e, T_i and Z_{ei}.

Insights gained

- Rates (CM effects) and heat capacities are time-dependent
- Condition for CM effects: $T_i < 0.27\, Z\, T_e$ or $T_i < 0.27\, Z\, T_F$ (usually not fulfilled at $t = \infty$)
- Quantum effects are important
- Correlations can be added via LFC; heat capacities from DFT-MD

$\Rightarrow$ **Electron-ion coupling constant cannot be used in many cases!**

$\Rightarrow$ **Electron-ion energy relaxation can be tested wit x-ray scattering**

… as a Summary

momentum relaxation electrons: ~ 1 **fs**

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

momentum relaxation ions: ~ 100 **fs**

temperature relaxation ~ 10 **ps**

Transient processes offer a window to rich & interesting physics and FELs, combined with high-energy laser, are a perfect tool to investigated the different relaxation stages toward equilibrium.

One has to resist the "equilibrium trap" when analysing the data!

Thank you!

… and **many thanks** for fruitful collaborations
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