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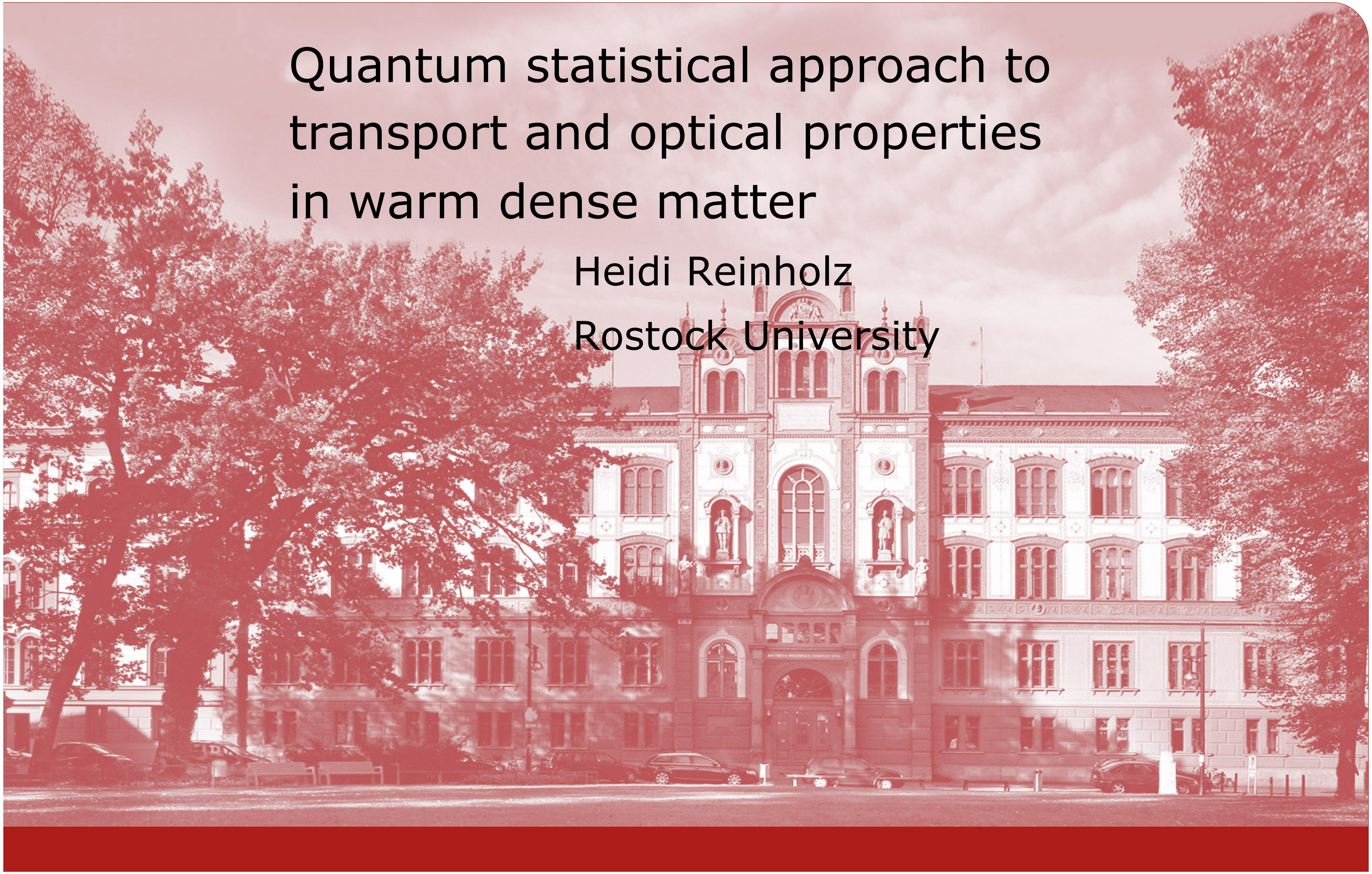
Quantum statistical approach to transport and optical properties in warm dense matter

Heidi Reinholz

Rostock University



800 years Rostock (1218-2018)
600 years University (1419-2019)
60 years Prof. Dr. David Blaschke
>40 years of friendship (since 1978)

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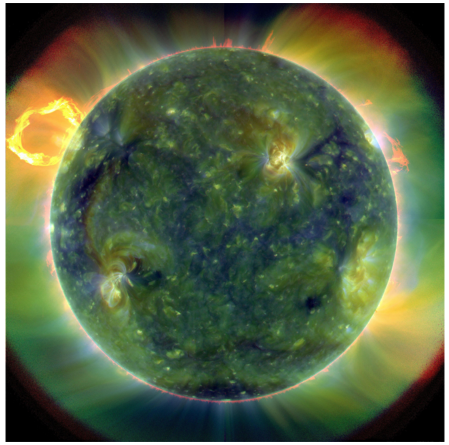
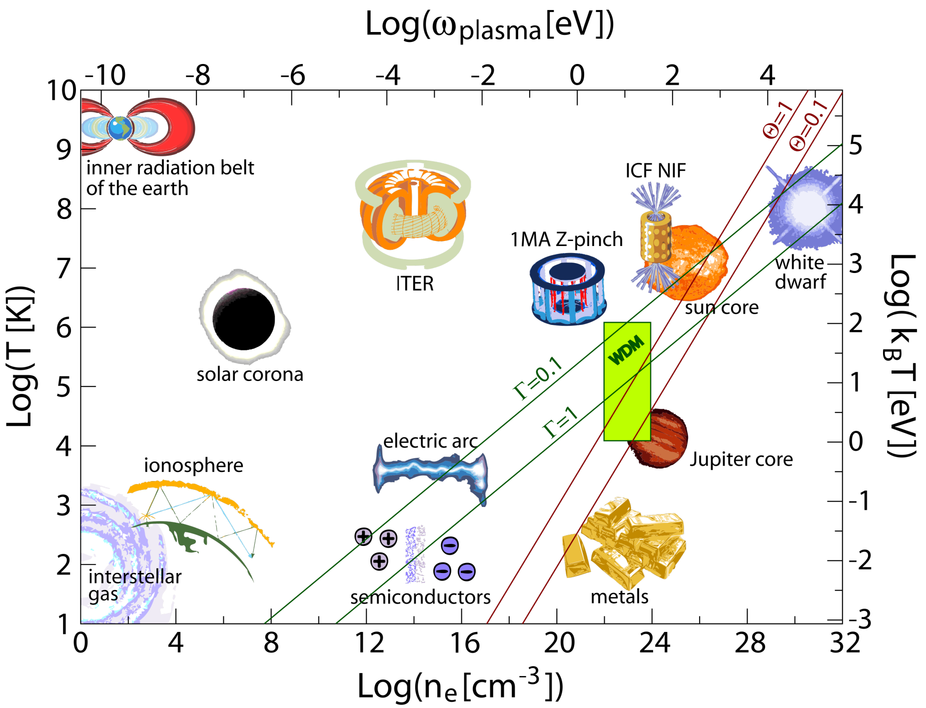
Outline

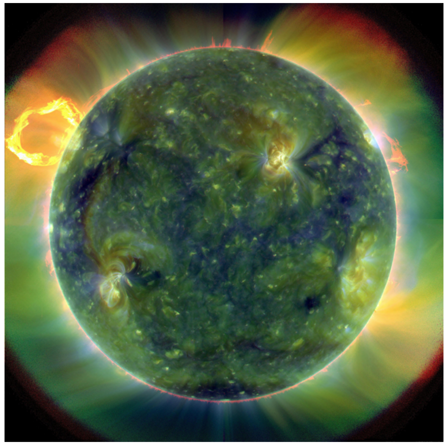
- Motivation
- Diagnostics – some examples
- Dielectric function &
  dynamical collision frequency –
  using the generalized Zubarev formalism
- Diagnostics – more on examples
- Outlook

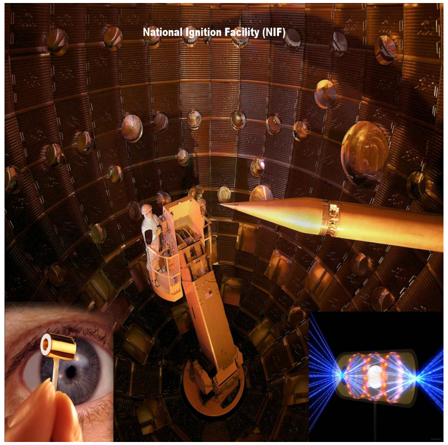


Motivation

- Dense plasma & warm dense matter





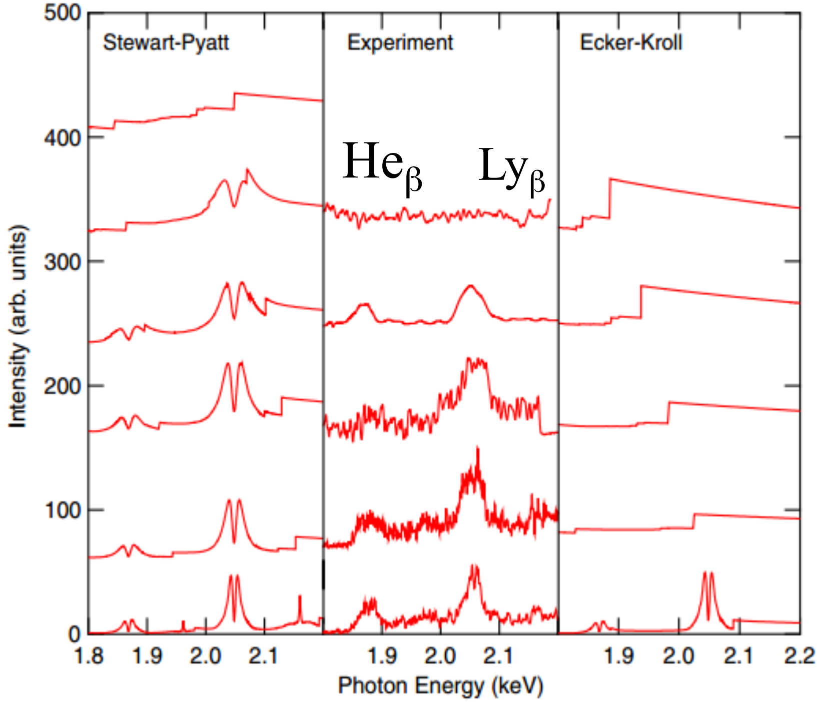


ICF @ NIF, Livermore

Diagnostics

- Dense plasma & warm dense matter
- Diagnostics – some examples
  - ✓ K_α fluorescence (IPD)

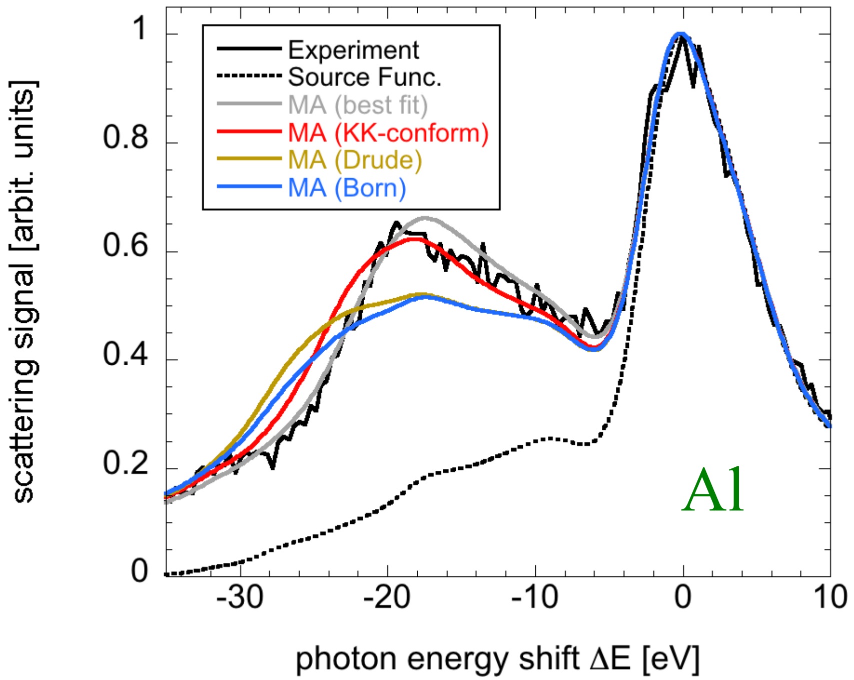
ionization potential depression



shock compressed aluminum

D.J. Hoarty et al., PRL **110**, 265003 (2013)

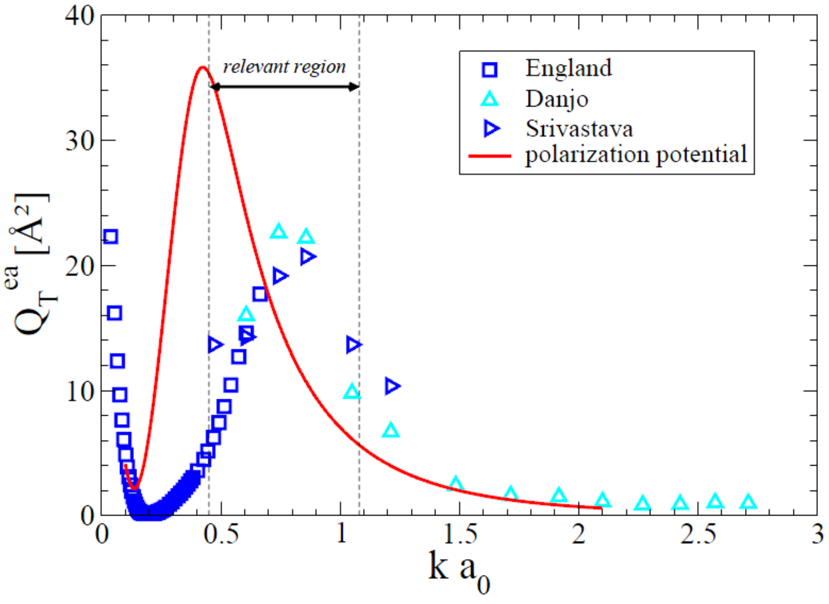
- Dense plasma & warm dense matter
- Diagnostics – some examples
  - ✓ K_{α} fluorescence (IPD)
  - ✓ Thomson scattering in Al



Diagnostics

- Dense plasma & warm dense matter
- Diagnostics – some examples
  - ✓ K_α fluorescence (IPD)
  - ✓ Thomson scattering in Al
  - ✓ Inert gases (dc conductivity, reflectivity)

electron-atom cross section



Outline

Tschernogolovka/Russia
shock waves

- Dense plasma & warm dense matter
- Diagnostics – some examples
  - ✓ K_α fluorescence (IPD)
  - ✓ Thomson scattering in Al
  - ✓ Inert gases (dc conductivity)
- Theory - generalized Zubarev formalism

FLASH @ DESY Hamburg
& European XFEL @ Schenefeld

Dense plasma

correlated Coulomb systems

- collisions
- medium effects: dynamical screening, collective excitations
- bound states, continuum correlations

$$\hat{H} = \sum_p E_p \hat{a}_p^\dagger \hat{a}_p + \sum_{pq} V_{\rm ei}(q) \hat{a}_{p+q}^\dagger \hat{a}_p + \frac{1}{2} \sum_{p_1 p_2 q} V_{\rm ee}(q) \hat{a}_{p_1+q}^\dagger \hat{a}_{p_2-q}^\dagger \hat{a}_{p_2} \hat{a}_{p_1}$$

many particle theories

- (linear) response theory $\hat{\rho}(t)$
- kinetic equations $f(\vec{p}, t) = {\rm Tr}\left\{\hat{\rho}(t)\, \delta\hat{n}_p\right\}$

$$\vec{j}(t) = \frac{e}{m\Omega_0} \sum_p \hbar\vec{p} f(\vec{p}, t) = \frac{e}{m\Omega_0} \vec{P}_1(t) = \frac{e}{m\Omega_0} {\rm Tr}\left\{\hbar\vec{p}\delta\hat{n}_p\right\}$$

Dielectric and optical response

$$\epsilon(\vec{k},\omega) = 1 - \frac{1}{\epsilon_0 k^2}\,\Pi(\vec{k},\omega) \qquad \Pi = \Pi_1 + \Pi_2 + \dots$$

Polarization function Π from many-particle theory, cluster expansion

- Optical information: refraction index & absorption coefficient

  Π_1 – Bremsstrahlung, Π_2 – spectral line profiles

$$\lim_{k\to 0} \epsilon_t(\vec{k},\omega) = \left(n(\omega) + \frac{ic}{2\omega}\alpha(\omega)\right)^2$$

- Dynamical structure factor → Thomson scattering, IPD

$$S(\vec{k},\omega) = \frac{1}{\pi\, V(k)}\,\frac{1}{\mathrm{e}^{-\beta\hbar\omega}-1}\,\mathrm{Im}\epsilon_l^{-1}(\vec{k},\omega)$$

- for one-particle properties:

$$\Pi_1(\vec{k},\omega) = \frac{k^2}{i\omega}\sigma(\vec{k},\omega) = \frac{\epsilon_0 k^2 \omega_{\rm pl}^2}{\omega(\omega - i\nu(\vec{k},\omega))}$$

Dynamical conductivity

$$\lim_{k\to 0} \epsilon(\vec{k},\omega) = 1 + \frac{i}{\epsilon_0\omega}\sigma(\omega) = 1 - \frac{\omega_{\rm pl}^2}{\omega(\omega - i\nu(\omega))}$$

! dynamical conductivity ←→ static conductivity

Drude formula $\sigma(\omega) = \frac{\omega_{\rm pl}^2}{i\omega + 1/\tau}$ with relaxation time $\tau = \frac{1}{\nu} = \frac{\sigma_{\rm dc}}{\epsilon_0\omega_{\rm pl}^2}$

here: generalized Zubarev formalism in linear response

Relevant observables B_m

generalized grand canonical ensemble with respect to a set of relevant observables $\{B_n\}$ from principle of maximum of the entropy leading to generalized Boltzmann equation with response parameters F_n(in linear response)

$$\langle B_m; \dot{\vec{R}}_c \rangle_z e_c \vec{E} = \sum_n \left\{ \langle B_m; \dot{B}_n \rangle_z - i\omega \langle B_m; B_n \rangle_z \right\} F_n$$

$$\mathrm{Tr}\,\{\rho B_m\} = \sum_n (B_m; B_n)\, F_n$$

$$\frac{\mathrm{d}S(t)}{\mathrm{d}t} = \sum_n F_n(t) \mathrm{Tr}\left\{\rho_{\mathrm{irrel}}(t) \dot{B}_n\right\}$$

choice of relevant observables is arbitary but determines

- which initial conditions/correlations are taken into account
- dynamical build up of remaining correlations
- explicit solutions after specific approximations (variational principle, perturbation theory, Green function techniques)

Relevant observables B_m

$$\langle B_m; \dot{\vec{R}}_c \rangle_z e_c \vec{E} = \sum_n \left\{ \langle B_m; \dot{B}_n \rangle_z - i\omega \langle B_m; B_n \rangle_z \right\} F_n$$

$$\mathrm{Tr}\,\{\rho B_m\} = \sum_n (B_m; B_n)\, F_n$$

$$\frac{\mathrm{d}S(t)}{\mathrm{d}t} = \sum_n F_n(t) \mathrm{Tr}\left\{ \rho_{\mathrm{irrel}}(t) \dot{B}_n \right\}$$

- ✓ Kubo: $\rho_{\mathrm{rel}}=\rho_0$ (curent-current correlations functions)
- ✓ B_n: linear momentum -> force-force correlation function (dynamical collision frequency)
- ✓ B_n: one-particle distribution function -> kinetic theory
- ✓ B_n: two-particle distribution function -> bound states
- ✓ B_n: fluctuations $\{\delta n_p\}$ of single-particle occupation number -> kinetic equation

$$\sigma(\omega) \propto \langle \vec{j}; \vec{j} \rangle_{\omega+i\eta}$$

$$\nu(\omega) = \frac{1}{\tau(\omega)} \propto \langle \vec{F}; \vec{F} \rangle_{\omega+i\eta}$$

$$F = F_{\mathrm{ei}} + F_{\mathrm{ee}} + F_{\mathrm{ea}}$$

Kinetic equation

- choose fluctuations $\{\delta n_p\}$ of single-particle occupation number as relevant observables $\{B_n\}$ and introduce response parameters $F_p(t)$

$$\delta f(\vec{p},t) = \sum_{p'} (\delta\hat{n}_p, \delta\hat{n}_{p'})\, F_{p'}(t)$$

$$\delta S(t) = -k_B \sum_p F_p(t)\ \delta f(\vec{p},t) \leq 0$$

- terms in kinetic equation can be identified

$$\frac{\partial}{\partial t} f(\vec{p},t) = \mathrm{D}\left[f(\vec{p},t)\right] + \mathrm{C}\left[f(\vec{p},t)\right]$$

$$-i\omega\delta\tilde{f}(\vec{p},\omega) = \frac{e\hbar}{m}\beta\tilde{\vec{E}}(\omega)\cdot\vec{p}\, f_p(1-f_p) + \mathrm{C}\left[\delta\tilde{f}(\vec{p},\omega)\right]$$

- Kohler variational principle is generalized for arbitrary frequency:
  internal entropy production as functional of an arbitrary function G_p - is a maximum if G_p=F_p is solution of the linear Boltzmann equation [HR, Röpke, PRE (2012)]

Relaxation time vs Drude

- collision term in Boltzmann equation $\mathrm{C}\left[\delta\tilde{f}(\vec{p},\omega)\right]$

- for relaxation time approximation, taken from static case C = - δf (p) / τ_p for Lorentz plasma
  leading to dynamical conductivity (isotropic system)

$$\sigma_{\mathrm{KT}}(\omega) = \frac{2}{3}\frac{e^2\hbar^2\beta}{m^2}\int\frac{d^3\vec{p}}{(2\pi)^3}\frac{p^2 f_p(1-f_p)}{-i\omega+1/\tau_p}$$

  - energy dependent static collision time
  - Drude type expression is not obtained, no e-e collisions, sum rules not fulfilled

- from linear response theory: generalized Drude expression with collision frequency $\nu(\omega)=1/\tau(\omega)$

$$\mathrm{C}_p[\delta\tilde{f}(\vec{p},\omega)] = -\sum_{p'}\mathcal{L}_{pp'}(\omega)\tilde{F}_{p'}$$

$$\mathcal{L}_{pp'}(\omega) = \mathcal{L}^{\mathrm{ei}}_{pp'}(\omega) + \mathcal{L}^{\mathrm{ee}}_{pp'}(\omega)$$

$$\sigma_{\mathrm{LRT}}(\omega) = \frac{\epsilon_0\omega_{\mathrm{pl}}^2}{-i\omega+\nu(\omega)}$$

Landau,Lifshitz X; HR, Röpke PRE 2012

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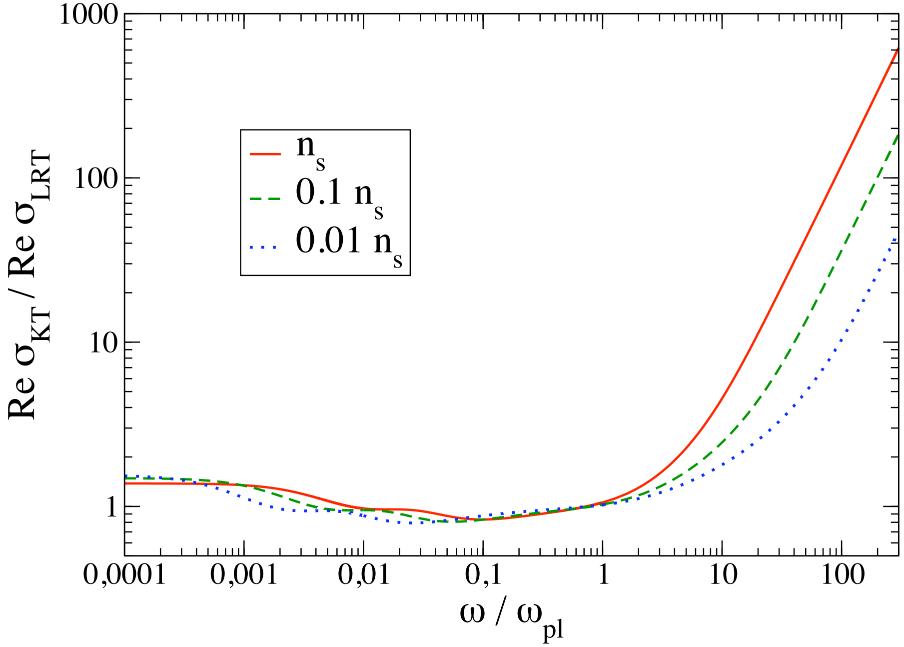
Relaxation time vs Drude

$$\nu_{\mathrm{LRT}}(\omega) = r^{(2)}(\omega) \frac{\beta}{m\, n\, \Omega_0} \langle \hat{\dot{P}}_1; \hat{\dot{P}}_1 \rangle_{\omega+i\epsilon}$$

solar core conditions
T=573eV
n_s=1.5 10^{25} cm^{-3}

HR, Röpke PRE 2012



Correction factor $r^{(2)}(\omega)$ due to higher moments, including e-e-correlations, for static case see *S. Rosmej PhD thesis 2018, CPP 56 (2016) 327*

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Tschernogolovka/Russia
shock waves

FLASH (XUV)
& European XFEL

Thomson scattering from Al foil

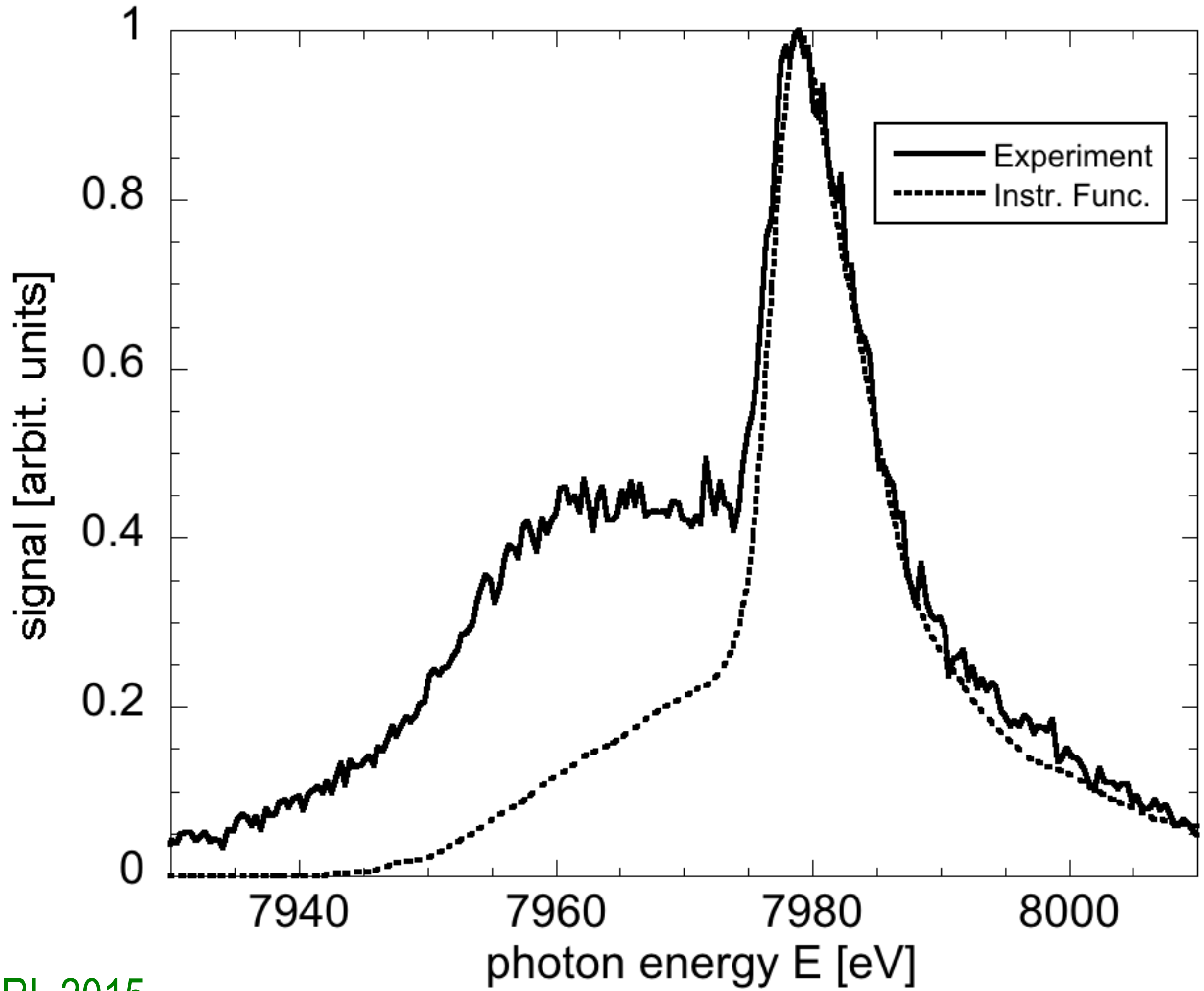
SLAC Stanford

n_e= 1.8 10^{22} cm^{-3}

laser @ 7.98 keV

1 μm focal spot

Θ = 24^0 scattering angle



Sperling, HR, Röpke et al. PRL 2015

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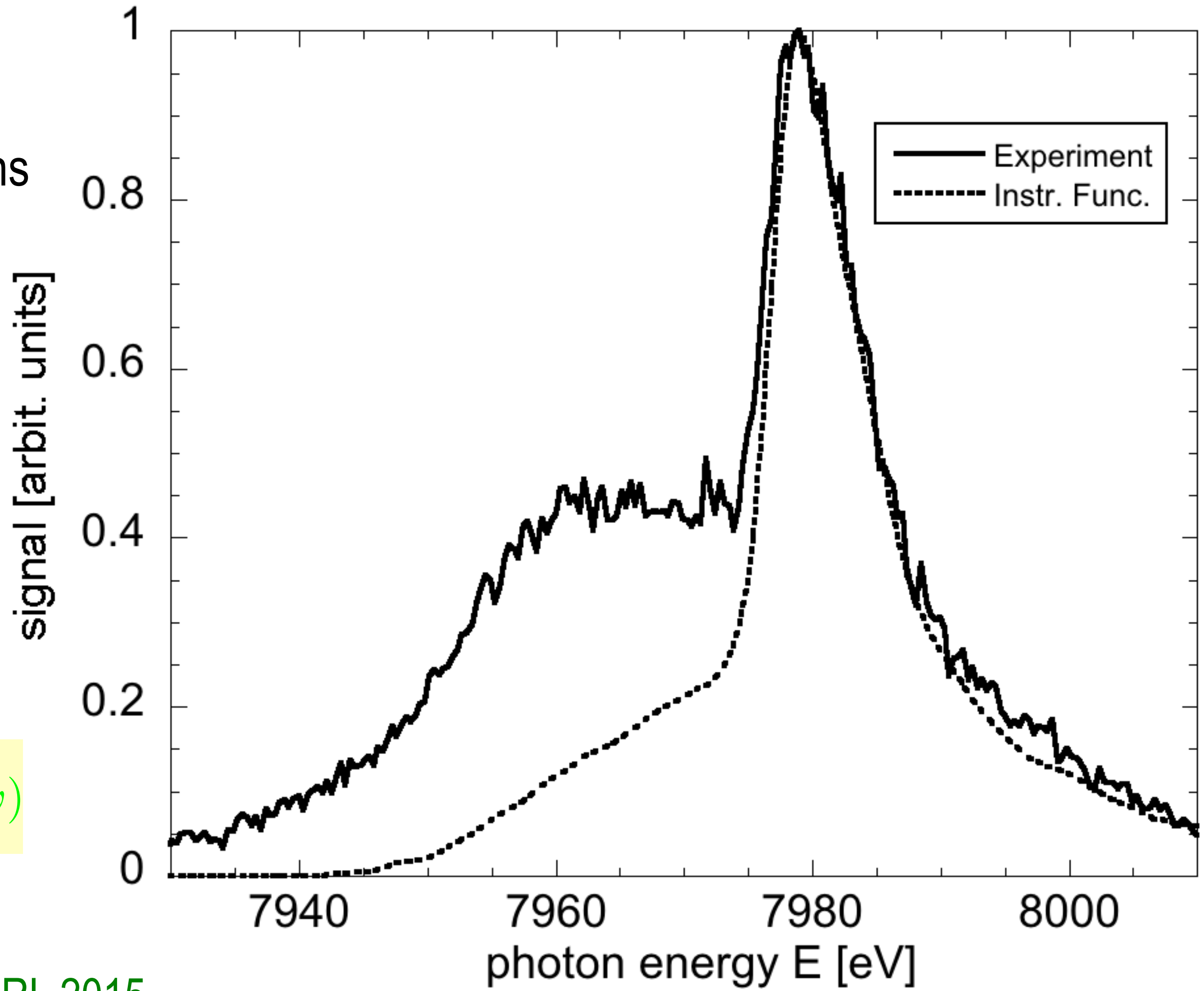
Thomson scattering from Al foil

intensity *I*
of signal from free electrons determined by
Thomson scattering cross section σ_T
and
strukturfactor S

$$I \propto \frac{d^2\sigma}{d\Omega\, d\omega} = \sigma_T \frac{k_1}{k_0} S(k,\omega)$$



Sperling, HR, Röpke et al. PRL 2015

Dynamical structure factor

$$S(\vec{k},\omega) = \frac{1}{\pi V(k)} \frac{1}{e^{-\beta\hbar\omega}-1} \mathrm{Im}\epsilon_l^{-1}(\vec{k},\omega)$$

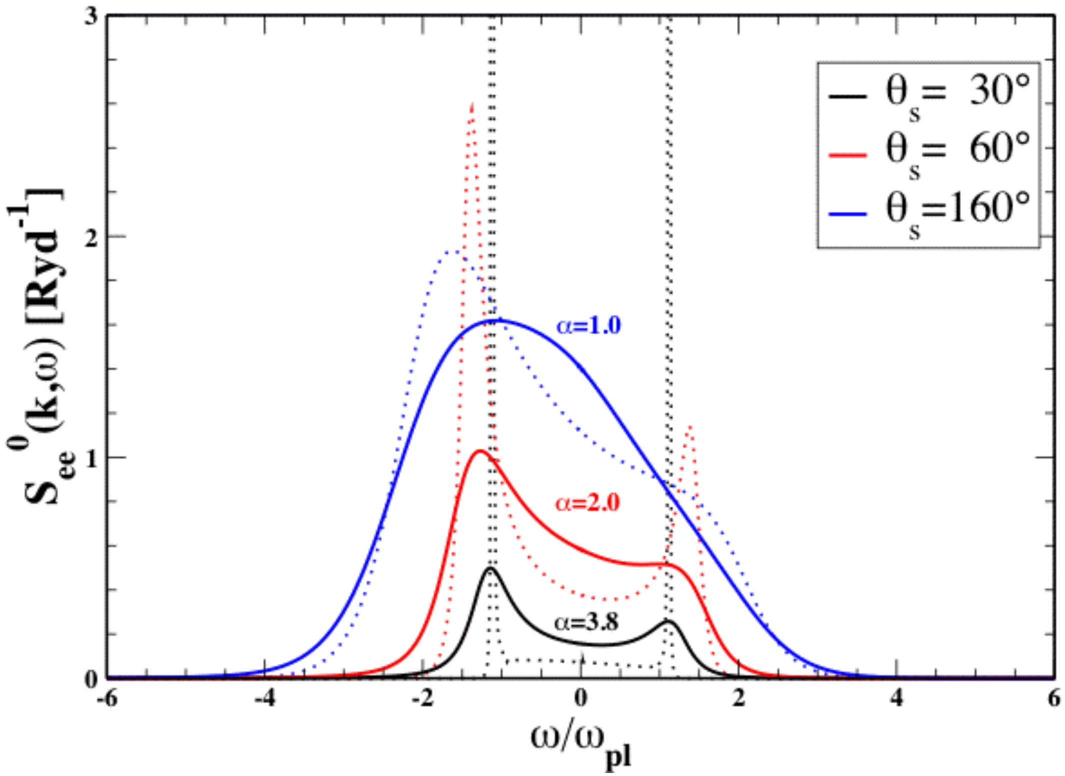
Calculation of Thomson signal for (nearly) free electrons @ small wavevectors via Mermin approach, thus going beyond RPA (collisionless plasma)

$$\epsilon^{\mathrm{Mermin}}(k,\omega) = 1 + \frac{1}{\epsilon_0 k^2} \frac{\Pi^{\mathrm{RPA}}(k,\omega+i\nu(\omega))}{1 - \frac{1}{1-i\omega/\nu(\omega)}\left[1 - \frac{\Pi^{\mathrm{RPA}}(k,\omega+i\nu(\omega))}{\Pi^{\mathrm{RPA}}(k,0)}\right]}$$

Linear response theory: introducing a complex collision frequency ν(w) in RPA
expression in order to take into account collisions consistently
via dynamical collision frequency ν(ω) from generalized Drude type expression

Thomson scattering

R. Thiele et al, PRE 2007



hydrogen:
$n_e = 10^{21}$ cm^{-3}
T = 2 eV
λ_L = 4.13 nm

•••••• ideal plasma
(RPA – random phase approximation)

——— plasma with collisions
(Born-Mermin approximation)

scattering parameter: $\alpha(n_e, T, \lambda_L, \Theta_s) > 1$ collective scattering on plasmons

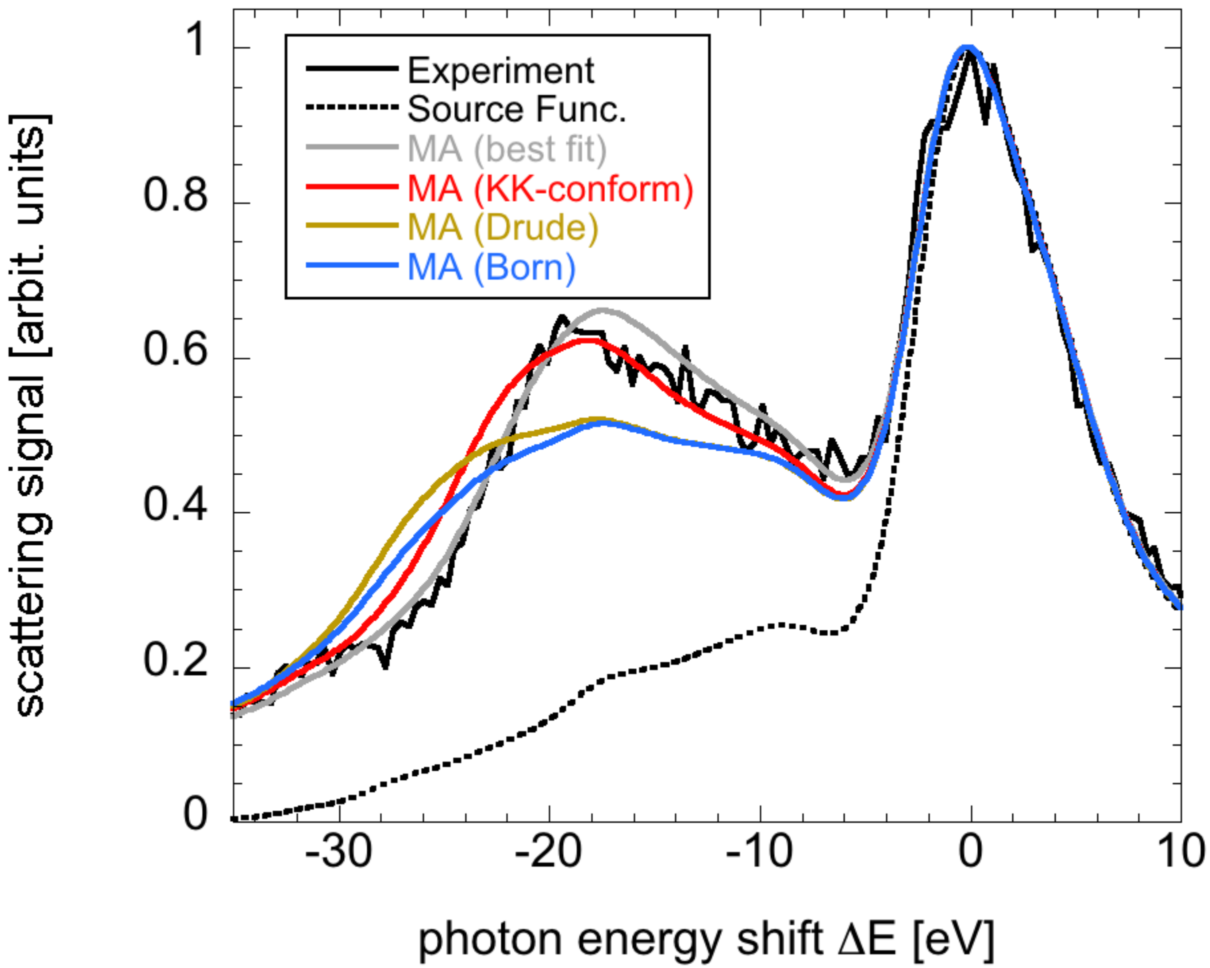
Thomson scattering from Al foil

N_e= 1.8 10^{22} cm^{-3}
laser @ 7.98 keV
10 μm focal spot, T=0.5eV
Θ = 24^0 scattering angle

temperature via

- SCFLY simulation code
- detailed balance

KK – Kramers-Kronig relation
(sum rule)



Sperling, Rosmej et al.
PRL 2015, JPB 50 (2017)

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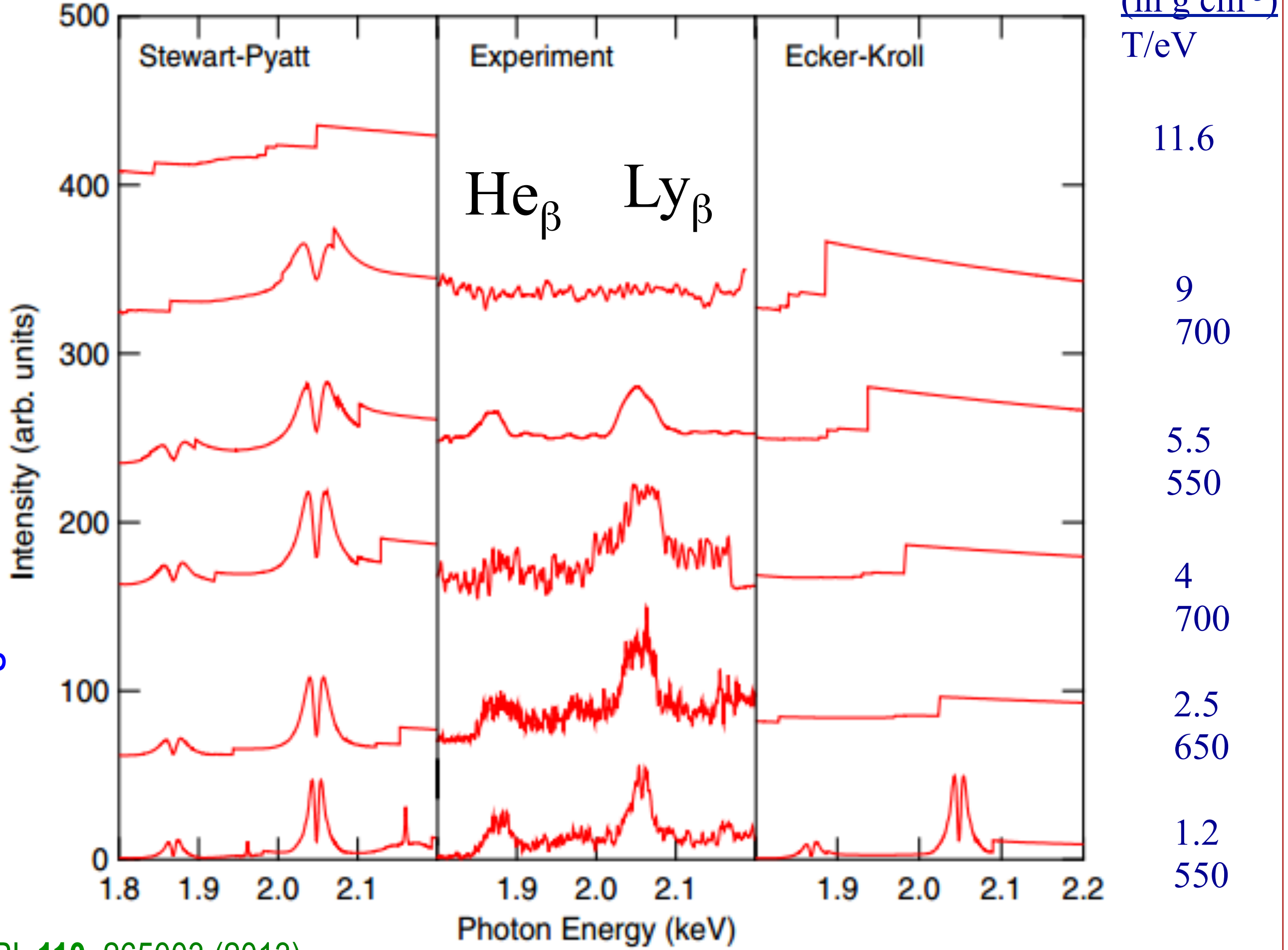
FLASH (XUV)
& European XFEL

IPD in shock commpressed Al

Shock wave compressed Al at different densities

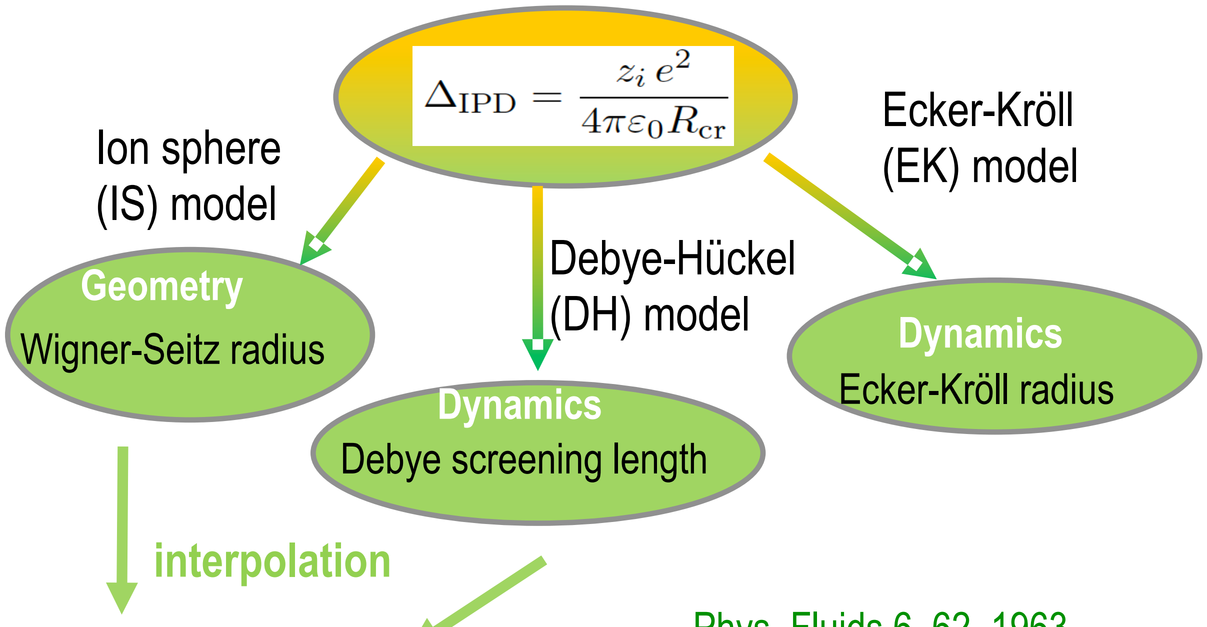


FLYCHK (@ T=700 eV) with SP (Stewart-Pyatt) model agrees better with experiment

D.J. Hoarty et al., PRL **110**, 265003 (2013)

Continuum lowering models

analytic approaches



Stewart-Pyatt (SP) model

finite-temperature
Thomas-Fermi theory

Phys. Fluids 6, 62, 1963

Astrophys. J. 144, 1203, 1966

G. Zimmerman and R. More,
JQSRT 23, 517, 1980

numerical approaches

two-step Hatree-Fock calculations
S.-K. Son et al.,
Phys. Rev. X 4, 031004 (2014)

density functional theory
S. M. Vinko et al,
Nat. Commun. 5, 3533 (2014)

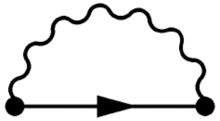
classical molecular dynamical simulations
A .Calisti, S. Ferri, and B. Talin,
J. Phys. B 48, 224003 (2015)

Monte-Carlo simulations
M. Stransky,
Phys. Plasmas 23, 012708 (2016)

Lin, 2018

IPD: quantumstatistical approach

- Self-energy in Montroll-Ward approximation

$$\Sigma_1^{\mathrm{MW}}(\mathbf{p}, z) =$$  $$= \sum_{\mathbf{q},\omega} G_1^0(\mathbf{p}-\mathbf{q}, z-\omega)\cdot V^{\mathrm{scr}}(\mathbf{q},\omega)$$

real part: energy shift

imaginary part: broadening

with the screened potential

$$V_{ab}^{\mathrm{scr}}(\mathbf{q},\omega) = V_{ab}(\mathbf{q})\cdot\left\{1+\int_{-\infty}^{\infty}\frac{d\omega'}{\pi}\cdot\frac{\mathrm{Im}\,\varepsilon^{-1}(\mathbf{q},\omega'-i\eta)}{\omega-\omega'}\right\}$$

dielectric function: influence of the plasma

- Fluctuation-dissipation theorem for the structure factor (SF)

$$\mathrm{Im}\,\frac{1}{\varepsilon(\mathbf{q},\omega+i0)} = \frac{\pi\, e^2}{\hbar\varepsilon_0\, q^2\,(1+n_{\mathrm{B}}(\omega))}\cdot\left[z_i^2\, n_i S_{\mathrm{ii}}(\mathbf{q},\omega) - 2z_i\sqrt{n_e n_i}S_{\mathrm{ei}}(\mathbf{q},\omega) + n_e S_{\mathrm{ee}}(\mathbf{q},\omega)\right]$$

ionic contribution

screening cloud

electronic contribution

$\Delta_c^{ion-ion}$ effective ionic contribution to the shift

Lin, 2018

- Change of the self-energy during an ionization process

$$i_{Z_i} \rightarrow e + i_{Z_i+1}$$

$$\Delta_{\mathrm{IPD}}^{\mathrm{ion-ion}} = \Delta_i^{\mathrm{ion-ion}} - (\Delta_e^{\mathrm{ion-ion}} + \Delta_{i+1}^{\mathrm{ion-ion}})$$

- Expression for IPD

$$\Delta_{IPD}^{ion-ion} \approx -\frac{(Z_i+1)e^2}{2\pi^2\epsilon_0 r_{\mathrm{WS}}} f(\Gamma_i) \int_0^\infty \frac{dq}{q^2} S_{ii}^{ZZ}(q)$$

Parameter function $F(\Gamma_i)$ via charge neutrality

$$f(\Gamma_i) = \frac{3\Gamma_i}{\sqrt{(9\pi/4)^{2/3} + 3\Gamma_i}}$$

① low density (weak coupling): DH limit

② high density (strongly coupled): IS model

③ transition from SP model to EK model with increasing coupling parameter

Lin, Röpke, Kraeft, HR, PRE 96 (2017)

IPD in shock commpressed Al

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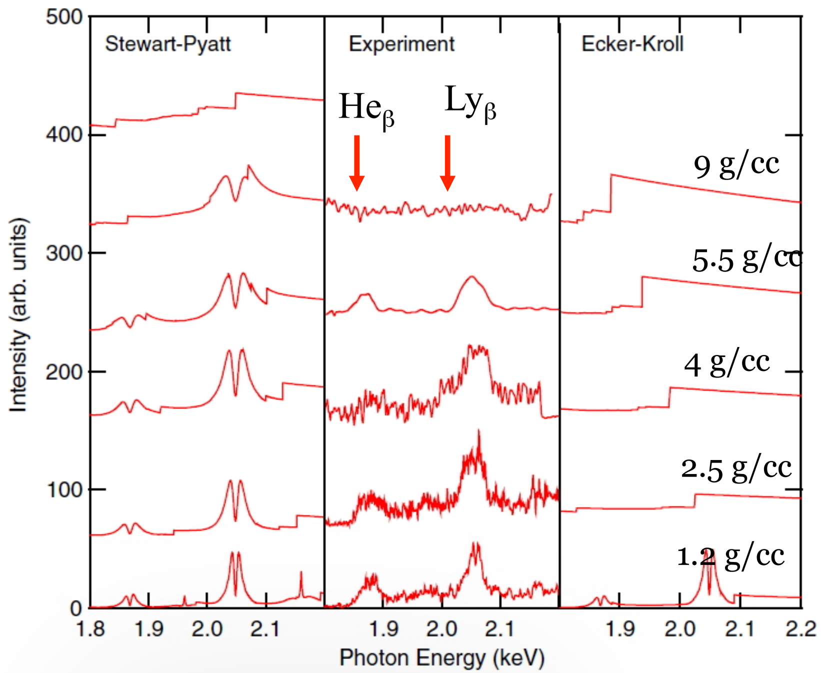
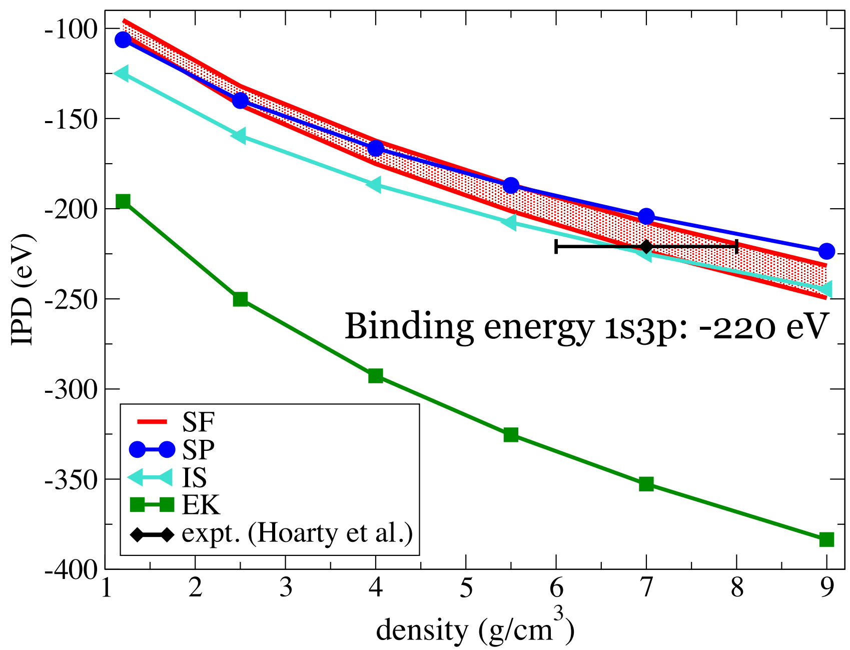
disappearance of spectral lines



- intermediate coupling
- relevant density range: 5.5 – 9 g/cc

Hoarty et al., PRL 110 (2013)

predictions by different approaches



- SF in good agreement with IS/SP

Lin et al., PRE 96 (2017)

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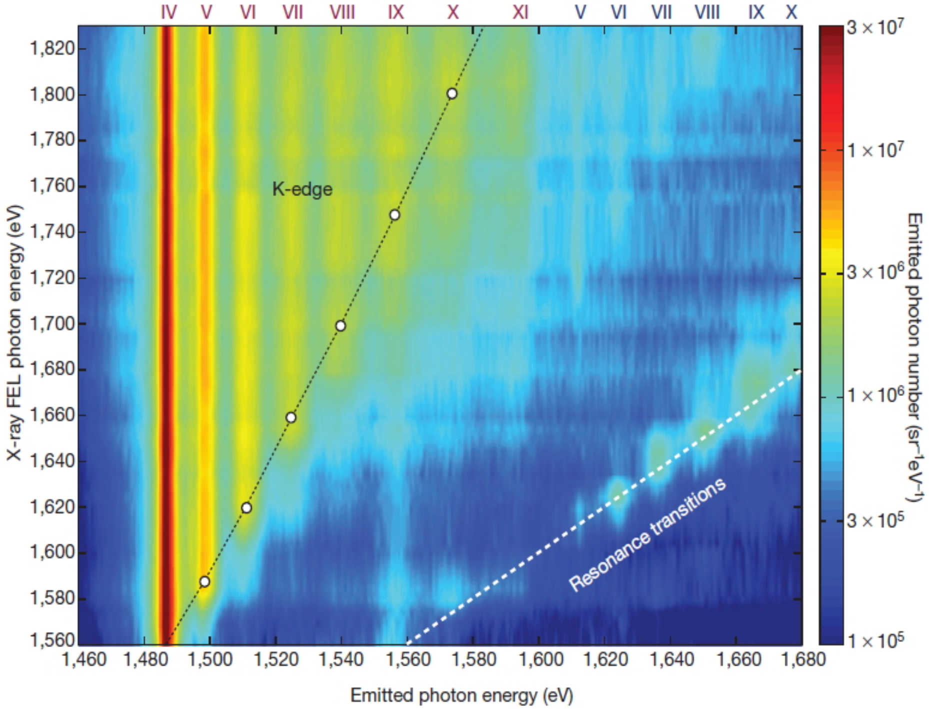
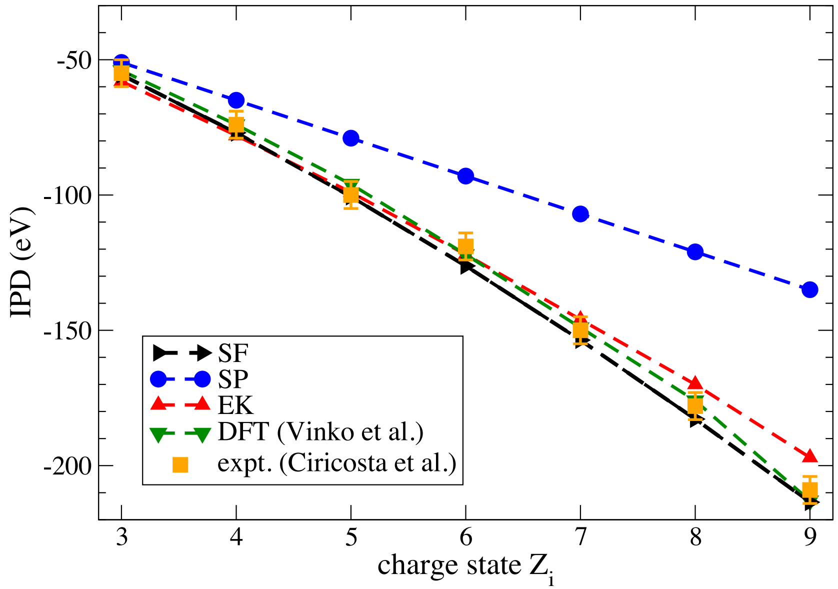
IPD for aluminum





- strong coupling
- SF in good agreement with EK model and DFT (Vinko et al.)

[2] Ciricosta *et al.*, PRL **109** (2012), Natl. Commun. 7(2016)
S. M. Vinko *et al.*, Nature **482** (2012); Lin et al., PRE 96 (2017)

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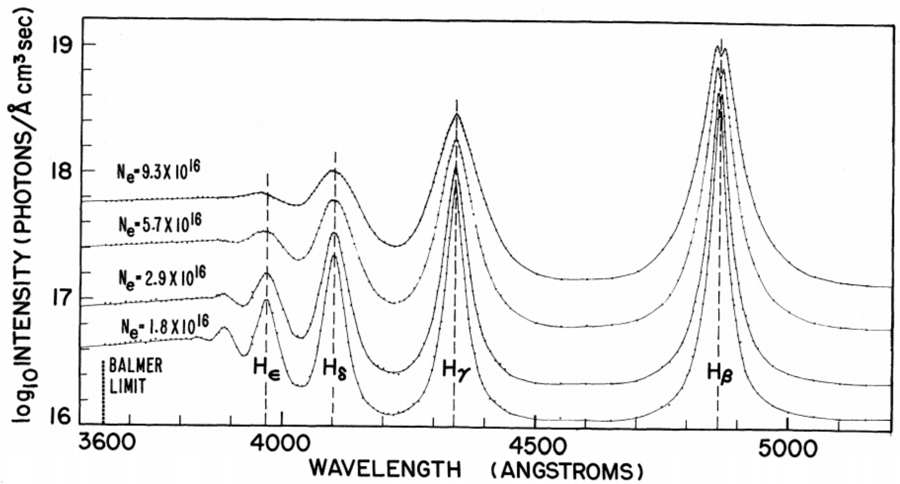
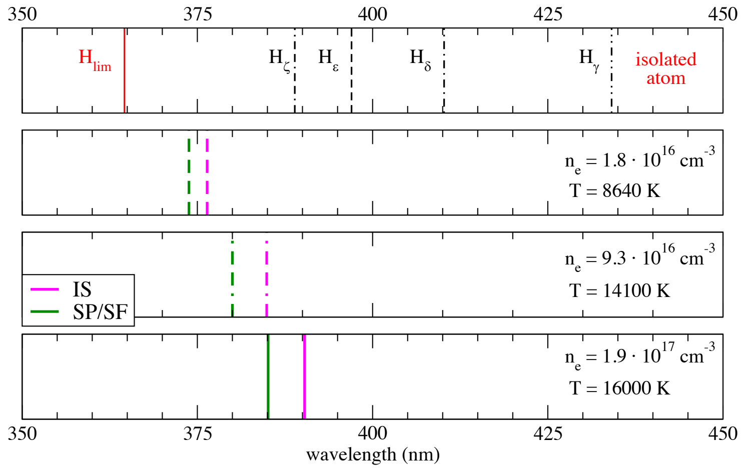
IPD vs. Inglis-Teller effect



Continuum Lowering (Lin PhD 2018)



Dissolution of spectral lines due to:

① Inglis-Teller effect

W. Wiese et al.,PRA 6, 1132 (1972)

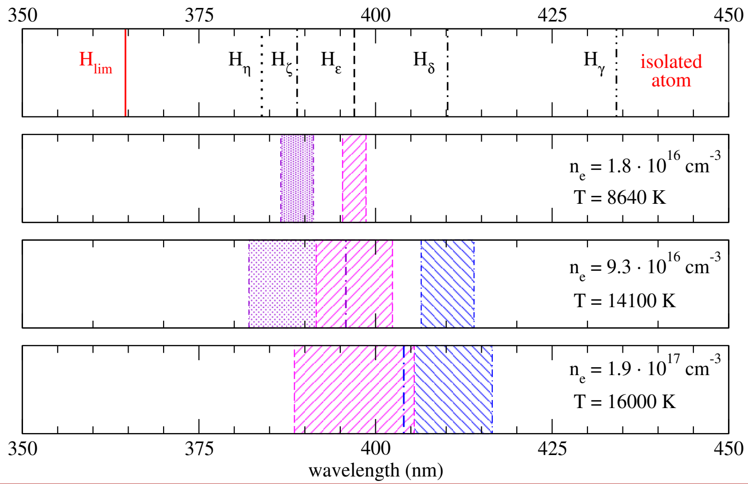
② IPD

D. J. Hoarty et al., PRL 110, 265003 (2013)

③ Inglis-Teller effect and IPD

B. Omar et al.,Contrib. Plasma Phys. 47, 315 (2007)

Inglis-Teller effect (Lin PhD 2018)



In-medium wave equation

Consistent treatment of two-particle problem:

$$\frac{p^2}{2m_e}\psi_n(p)+\sum_q V(q)\psi_n(p+q)-E_n\psi_n(p)=\sum_q V(q)\left[\psi_n(p+q)f_e(p)-\psi_n(p)f_e(p+q)\right]$$

Pauli blocking, Fock self-energy shift, Fermi function f_e

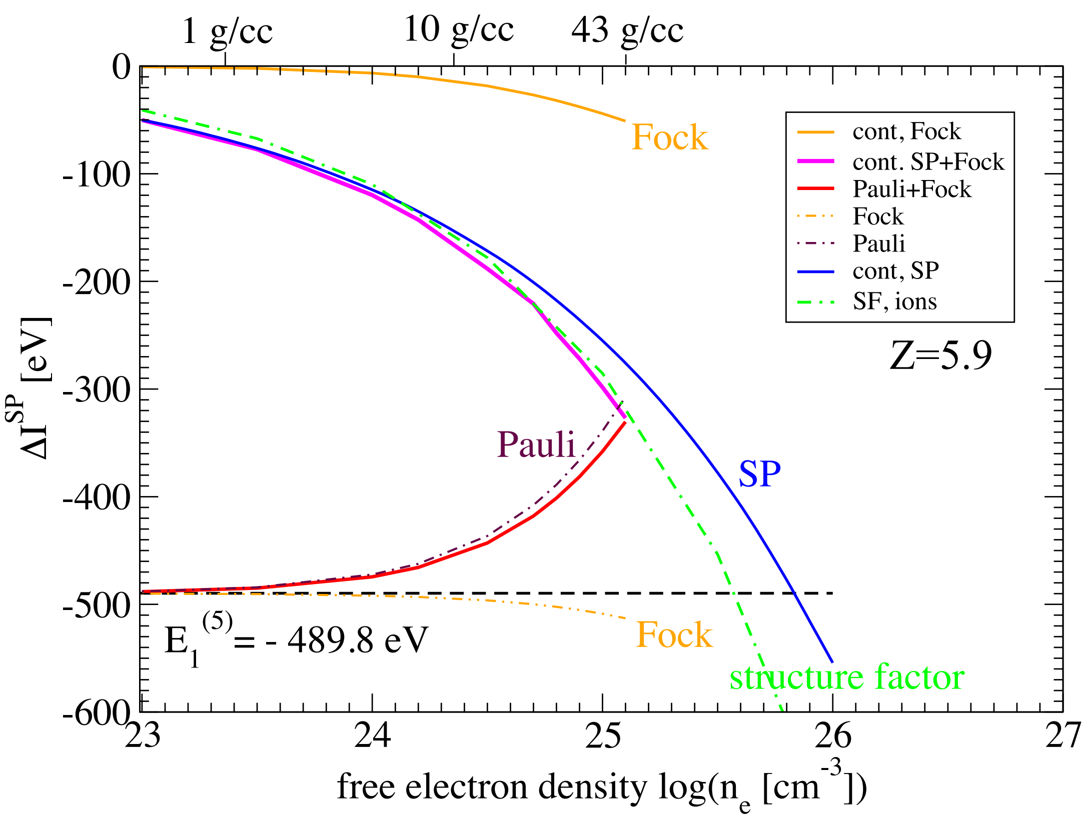
V(q) → dynamically screened Coulomb potential (dielectric function)

→ dynamical screening leads to dynamical self-energy shift (DH in low density limit)

Röpke, DB, Lin, Kraeft, HR, Redmer, HR, PRE 99 (2019)

Carbon ionization @ 100 eV

IPD of $C^{5+} <=> e + C^{6+}$



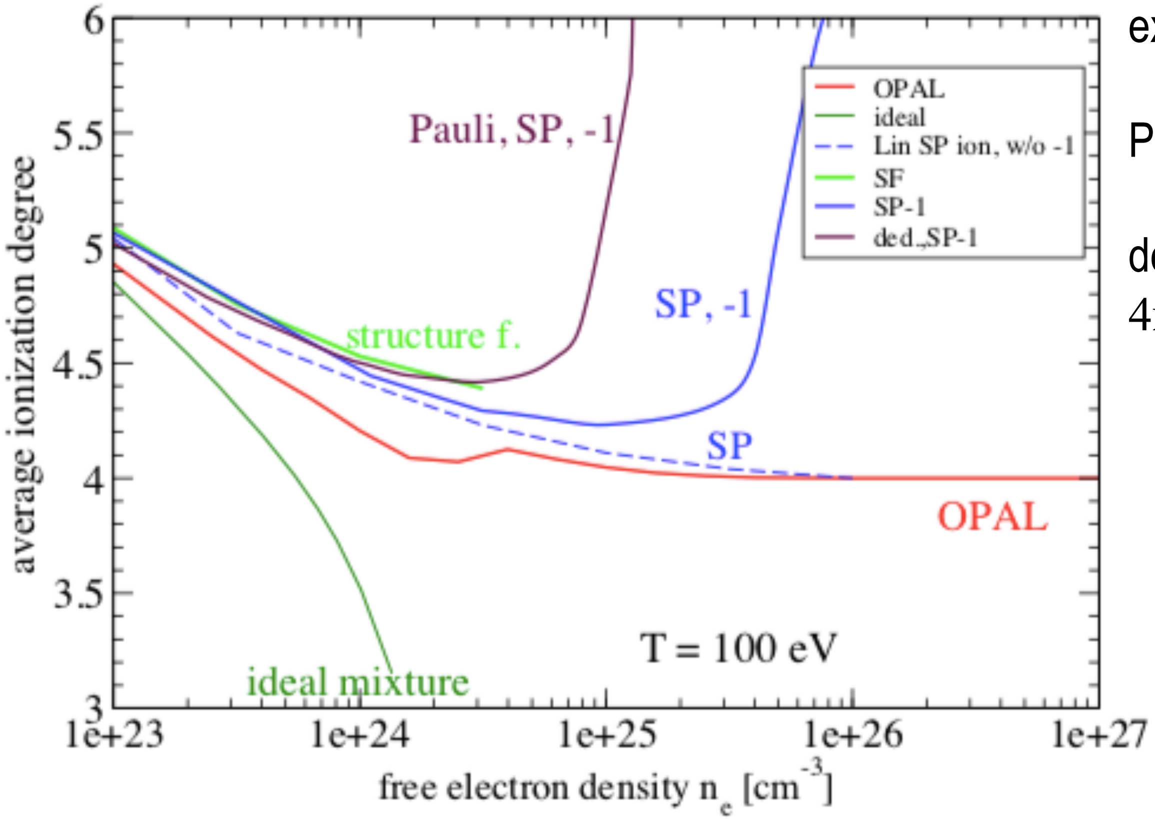
Composition and ionization degree
Potekhin et al., PRE 72 (2005)
$Z=5.9$

degeneracy: $T=T_F$ @
4×10^{24} cm^{-3} = 20 g cm^{-3}

Röpke, DB, Lin, Kraeft, HR, Redmer, HR, PRE 99 (2019)

Carbon ionization @ 100 eV

IPD of C^{5+} <=> e + C^{6+}



Saha equations including all ions and excited states (NIST)

Planck-Larkin partition function (-1)

degeneracy: $T=T_F$ @ 4×10^{24} cm^{-3} = 20 g cm^{-3}

Röpke, DB, Lin, Kraeft, HR, Redmer, HR, PRE 99 (2019)

summary and outlook

- A quantum statistical approach is worked out for WDM at arbitrary degeneracy with well defined concepts: spectral function, density of states, frequency dependent absorption coefficient, collision frequency, screening
- IPD - low-density (Debye-Hueckel) approximations are improved. EK and SP approximations are improved including dynamical screening and pair distribution functions.
- At high densities, the electrons become degenerate, and exchange effects must be taken into account. In particular, bound states are destroyed as a result of Pauli blocking.
- Future work
  - ➢ Optical properties (continuum edge, Inglis-Teller-effect)
  - ➢ Comparison with DFT–MD calculations and other approaches
  - ➢ Input for codes

Collaboraters

Rostock:	**C. Lin, S. Rosmej, A. Sengebusch, G. Röpke**, R. Redmer group
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