Ab initio PIMC approach to the dynamic properties of warm dense electrons

Workshop: Ab initio simulations of correlated fermions

Paul Hamann, Tobias Dornheim[†], Jan Vorberger[*], Zhandos Moldabekov[**], Michael Bonitz

Institute of Theoretical Physics and Astrophysics, Kiel University
[†] Center for Advanced Systems Understanding, [*] Helmholtz-Zentrum Dresden
[**] Al Farabi University, Almaty, Kazakhstan

July 8th, 2020

Introduction: warm dense matter

Warm dense matter (WDM):

- Nearly classical ions
- Degenerate non-ideal electrons
- Coupling parameter:

$$r_s = \frac{\bar{r}}{a_B} \sim 0.1 \ldots 10$$

- Degeneracy parameter:

$$\theta = T/T_F \sim 0.1 \ldots 10$$

- Temperature, degeneracy and coupling effects equally important
  → No small parameters

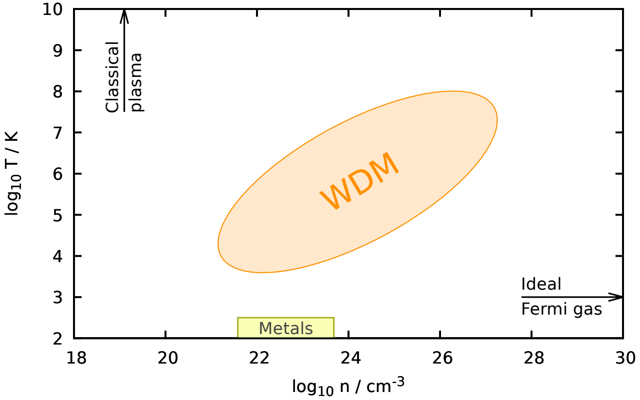


Figure: From T. Dornheim, S. Groth, and M. Bonitz, *Physics Reports* **744**, 1-86 (2018)

Perturbation theory and ground-state approaches fail

Ab initio dynamic (ω-dependent) results for the warm dense UEG

- **Key quantity:** dynamic structure factor

$$S(\mathbf{q},\omega) := \frac{1}{2\pi}\int_{-\infty}^{\infty} \mathrm{d}t\ \underbrace{\langle \hat{n}_{\mathbf{q}}(t)\hat{n}^{\dagger}_{\mathbf{q}}(0)\rangle}_{:=F(\mathbf{q},t)}\ e^{i\omega t}$$

$\rightarrow$ Directly measured in **scattering experiments**

- **Chihara decomposition** applies for non-collective scattering:

$$S(\mathbf{q},\omega) = S_{\text{b-b}}(\mathbf{q},\omega) + S_{\text{b-f}}(\mathbf{q},\omega) + S_{\text{f-f}}(\mathbf{q},\omega)$$

$$\rightarrow S_{\text{f-f}}(\mathbf{q},\omega) \sim S^{\text{UEG}}(\mathbf{q},\omega)$$

- **Practical example:** Fit model for $S(\mathbf{q},\omega;T_e)$ to spectrum to determine electron temperature T_e

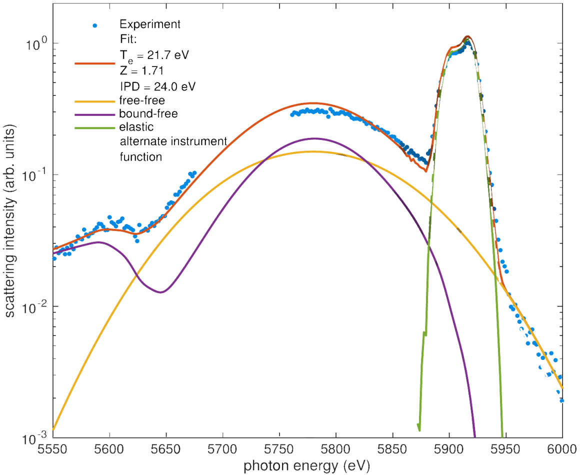


Figure: Scattering spectrum of isochorically heated graphite at LCLS. Taken from D. Kraus *et al.*, *Plasma Phys. Control. Fusion* (2019)

Investigation of dynamic quantities

$$S(\mathbf{q},\omega) := \frac{1}{2\pi}\int_{-\infty}^{\infty} \mathrm{d}t\; \underbrace{\langle \hat{n}_{\mathbf{q}}(t)\hat{n}_{\mathbf{q}}^{\dagger}(0)\rangle}_{:=F(\mathbf{q},t)}\; e^{i\omega t}$$

$\rightarrow$ requires **real time-dependent simulations**
$\rightarrow$ Rigorous treatment of correlations **in general not feasible**

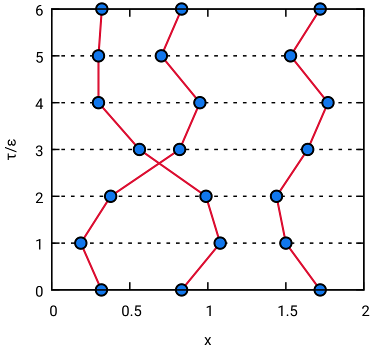
Investigation of dynamic quantities

$$S(\mathbf{q},\omega) := \frac{1}{2\pi}\int_{-\infty}^{\infty} \mathrm{d}t \underbrace{\langle \hat{n}_{\mathbf{q}}(t)\hat{n}^{\dagger}_{\mathbf{q}}(0)\rangle}_{:=F(\mathbf{q},t)} e^{i\omega t}$$

$\rightarrow$ requires **real time-dependent simulations**
$\rightarrow$ Rigorous treatment of correlations **in general not feasible**

Alternative: analytic continuation $t \rightarrow -i\tau$

$$F(\mathbf{q},\tau) = \int_{-\infty}^{\infty} \mathrm{d}\omega\ S(\mathbf{q},\omega)e^{-\tau\omega}\ \forall\ \tau \in [0,\beta]$$



Advantage:
$F(\mathbf{q},\tau)$ accessible within **PIMC** simulations
thermodynamic equilibrium $\rightarrow$ *ab initio* feasible

Investigation of dynamic quantities

$$S(\mathbf{q},\omega) := \frac{1}{2\pi}\int_{-\infty}^{\infty} \mathrm{d}t\ \underbrace{\langle \hat{n}_{\mathbf{q}}(t)\hat{n}^{\dagger}_{\mathbf{q}}(0)\rangle}_{:=F(\mathbf{q},t)}\ e^{i\omega t}$$

$\rightarrow$ requires **real time-dependent simulations**
$\rightarrow$ Rigorous treatment of correlations **in general not feasible**

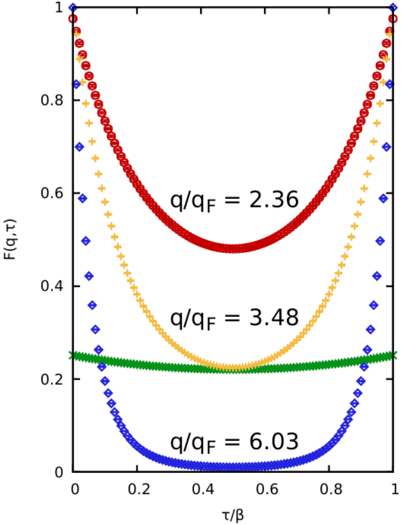
Alternative: analytic continuation $t \rightarrow -i\tau$

$$F(\mathbf{q},\tau) = \int_{-\infty}^{\infty} \mathrm{d}\omega\ S(\mathbf{q},\omega)e^{-\tau\omega}\ \forall\ \tau \in [0,\beta]$$

Advantage:
$F(\mathbf{q},\tau)$ accessible within **PIMC** simulations
thermodynamic equilibrium $\rightarrow$ *ab initio* feasible

Disadvantage:
$S(\mathbf{q},\omega)$ requires **inverse Laplace transform**
ill-posed problem $\rightarrow$ sensitive to statistical errors in F

Imaginary-time density-density correlation function:
($\theta = 1$, $r_s = 4$, $N = 66$)



The reconstruction problem

$$F(\mathbf{q},\tau) = \int_{-\infty}^{\infty} \mathrm{d}\omega\ S(\mathbf{q},\omega)e^{-\tau\omega}\ \forall\ \tau \in [0,\beta]$$

Known frequency moments: $\langle\omega^{-1}\rangle$, $\langle\omega^{0}\rangle$, $\langle\omega^{1}\rangle$, $\langle\omega^{3}\rangle$

$$\langle\omega^k\rangle = \int_{-\infty}^{\infty} \mathrm{d}\omega\ \omega^k S(\mathbf{q},\omega)$$

Spectrum which minimizes deviation from QMC results may not be the most desirable.

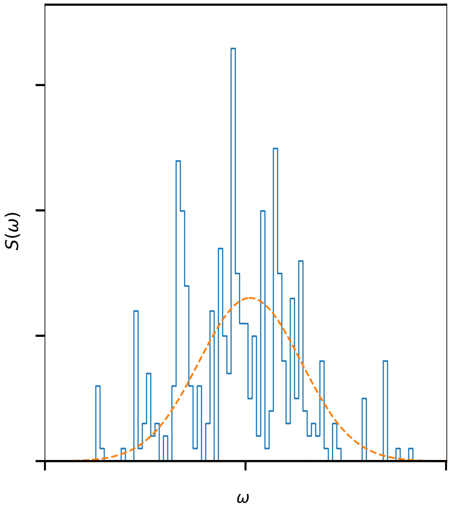
Two general approaches:

Introduce additional criteria, by which to select a most "physical" solution

$\rightarrow$ maximum entropy, ...

Stochastically sample set of all spectra reproducing $\{F(\tau), \langle\omega^k\rangle\}$ within accuracy limits, extract common features by averaging

$\rightarrow$ genetic algorithm (GIFT)[1], ...



[1] E. Vitali *et al.*, *Phys. Rev. B* **82**, 174510 (2010)

The reconstruction problem

$$F(\mathbf{q},\tau) = \int_{-\infty}^{\infty} \mathrm{d}\omega \; S(\mathbf{q},\omega) e^{-\tau\omega} \; \forall \; \tau \in [0,\beta]$$

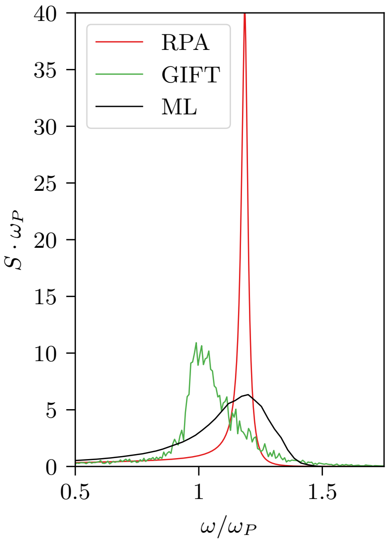
Known frequency moments: $\langle\omega^{-1}\rangle$, $\langle\omega^{0}\rangle$, $\langle\omega^{1}\rangle$, $\langle\omega^{3}\rangle$

$$\langle\omega^k\rangle = \int_{-\infty}^{\infty} \mathrm{d}\omega \; \omega^k S(\mathbf{q},\omega)$$

GIFT and ML spectra reproduce correct F and $\langle\omega^{-1}\rangle - \langle\omega^{3}\rangle$

Too much ambiguity $\Rightarrow$ Knowledge of F and $\langle\omega^{-1}\rangle - \langle\omega^{3}\rangle$ not sufficient to determine S

Dynamic structure factor of the UEG:
($\theta = 1$, $r_s = 10$, $N = 34$, $k = 0.63 k_F$)



[1] E. Vitali *et al.*, *Phys. Rev. B* **82**, 174510 (2010)

Incorporating additional information on $S(\mathbf{q},\omega)$ via dielectric formulation

- **Fluctuation-dissipation theorem:**

$$S(\mathbf{q},\omega) = -\frac{\mathrm{Im}\chi(\mathbf{q},\omega)}{\pi n(1-e^{-\beta\omega})}$$

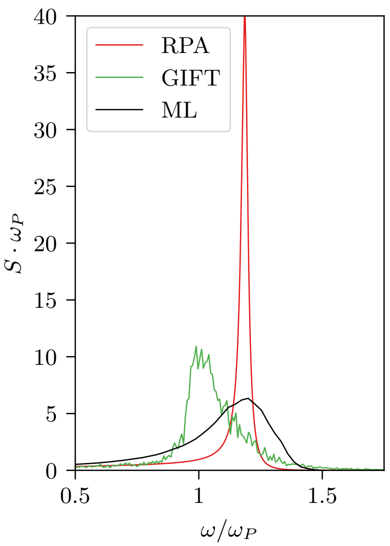
- Express response function χ via ideal response function χ_0 and **dynamic local field correction** G:

$$\chi(\mathbf{q},\omega) = \frac{\chi_0(\mathbf{q},\omega)}{1 - v_q\left[1 - G(\mathbf{q},\omega)\right]\chi_0(\mathbf{q},\omega)}$$

- **Random phase approximation (RPA):** $G \equiv 0$

Make ansatz and optimize $G(\mathbf{q},\omega)$ instead of $S(\mathbf{q},\omega)$

Dynamic structure factor of the UEG:
($\theta = 1$, $r_s = 10$, $N = 34$, $k = 0.63k_F$)

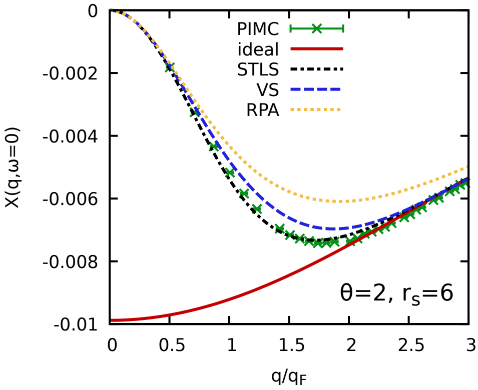


The Static Local Field Correction

- static limit $G(\mathbf{q}, \omega = 0)$ is connected to static density response $\chi(\mathbf{q})$:

$$G(\mathbf{q}) = 1 - \frac{1}{v_q}\left(\frac{1}{\chi_0(\mathbf{q},0)} - \frac{1}{\chi(\mathbf{q})}\right) \quad .$$

Source: S. Groth, et al., *Phys. Rev. B* **99**, 235122 (2019)



The Static Local Field Correction

- static limit $G(\mathbf{q}, \omega = 0)$ is connected to static density response $\chi(\mathbf{q})$:

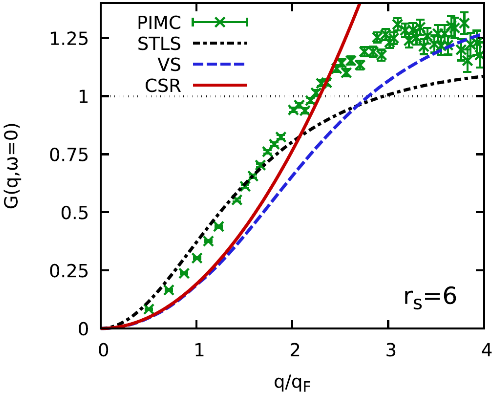
$$G(\mathbf{q}) = 1 - \frac{1}{v_q}\left(\frac{1}{\chi_0(\mathbf{q},0)} - \frac{1}{\chi(\mathbf{q})}\right) \quad .$$

- can be directly obtained from $F(\mathbf{q}, \tau)$:

$$\chi(\mathbf{q}) = -n \int_0^\beta \mathrm{d}\tau \; F(\mathbf{q}, \tau)$$

$\rightarrow$ Full $\mathbf{q}$-dependence from a single simulation of the **unperturbed UEG**

Source: S. Groth, et al., *Phys. Rev. B* **99**, 235122 (2019)



The Static Local Field Correction

- static limit $G(\mathbf{q}, \omega = 0)$ is connected to static density response $\chi(\mathbf{q})$:

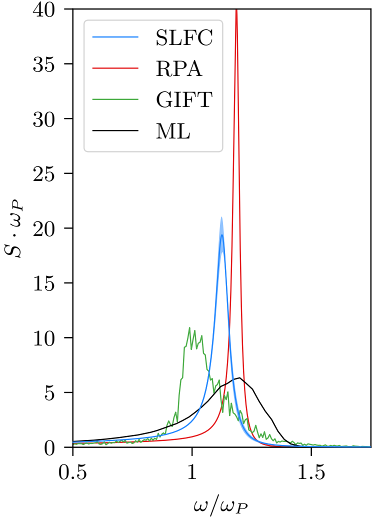
$$G(\mathbf{q}) = 1 - \frac{1}{v_q}\left(\frac{1}{\chi_0(\mathbf{q},0)} - \frac{1}{\chi(\mathbf{q})}\right) \quad .$$

- can be directly obtained from $F(\mathbf{q}, \tau)$:

$$\chi(\mathbf{q}) = -n \int_0^\beta \mathrm{d}\tau \; F(\mathbf{q}, \tau)$$

$\rightarrow$ Full $\mathbf{q}$-dependence from a single simulation of the **unperturbed UEG**

Dynamic structure factor of the UEG:
($\theta = 1$, $r_s = 10$, $N = 33$, $q = 0.63 q_F$)



Stochastic sampling of the **Dynamic** Local Field Correction

Source: S. Groth, et al., *Phys. Rev. B* **99**, 235122 (2019)

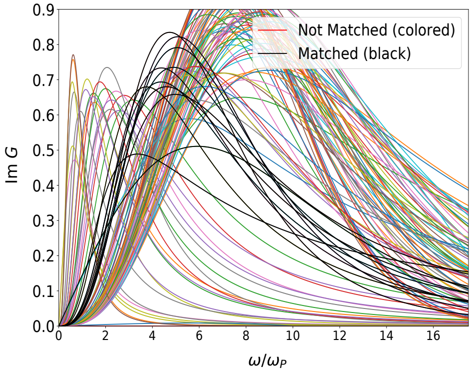
- Incorporating all knowledge about $G(\mathbf{q},\omega)$, make ansatz

$$\mathrm{Im}\, G(\mathbf{q},\omega) = \frac{a_0\omega + a_1\omega^3 + a_2\omega^5 + \cdots}{\left(b_0 + b_1\omega^2\right)^c}$$

Kramers-Kronig:

$$\mathrm{Re}\, G(\mathbf{q},\omega) - \mathrm{Re}\, G(\mathbf{q},\infty) = \frac{1}{\pi}\,\mathrm{P.V.}\int_{-\infty}^{\infty} \frac{\mathrm{Im}\, G(\mathbf{q},\omega')}{\omega'-\omega}\mathrm{d}\omega'$$

- randomly generate coefficients over many orders of magnitude
- store trial solutions that match $F(\mathbf{q},\tau)$ and $\langle\omega^k\rangle$



$q \approx 1.98 q_F$, $r_s = 6$, $\theta = 1$

Stochastic sampling of the **Dynamic** Local Field Correction

- Incorporating all knowledge about $G(\mathbf{q},\omega)$, make ansatz

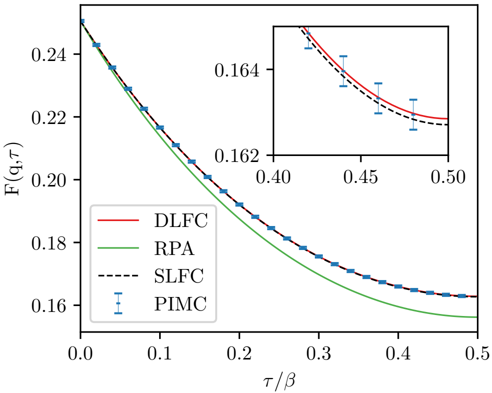
$$\mathrm{Im}\, G(\mathbf{q},\omega) = \frac{a_0\omega + a_1\omega^3 + a_2\omega^5 + \cdots}{\left(b_0 + b_1\omega^2\right)^c}$$

Kramers-Kronig:

$$\mathrm{Re}\, G(\mathbf{q},\omega) - \mathrm{Re}\, G(\mathbf{q},\infty) = \frac{1}{\pi}\,\mathrm{P.V.}\int_{-\infty}^{\infty} \frac{\mathrm{Im}\, G(\mathbf{q},\omega')}{\omega' - \omega}\mathrm{d}\omega'$$

Imaginary time density-density correlation function

$r_s = 4$, $\theta = 1$, $q/q_F \approx 0.63$

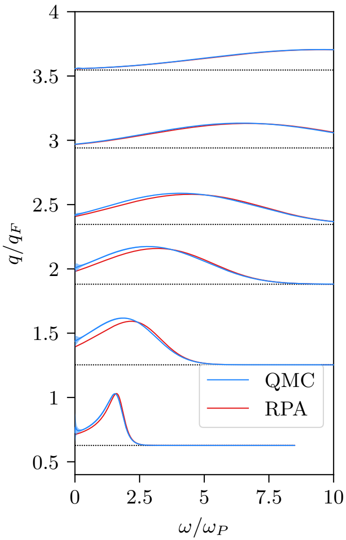


Correlation effects in the dispersion relation: $\theta = 1$, $r_s = 2$

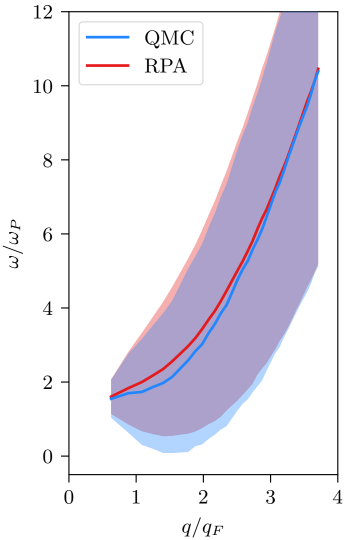
T. Dornheim, S. Groth, J. Vorberger, and M. Bonitz, *Phys. Rev. Lett.* **121**, 255001 (2018)

- Slight **correlation induced redshift** for intermediate q (at small r_s)

Dynamic structure factor of the UEG:



Peak position and FWHM:

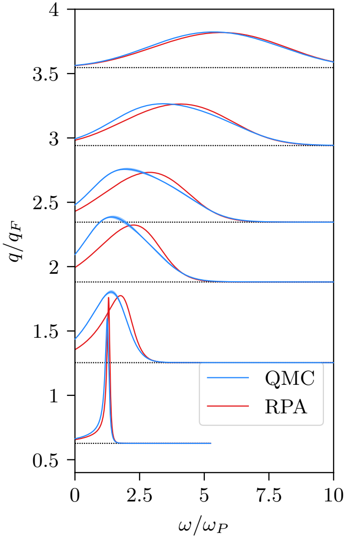


Correlation effects in the dispersion relation: $\theta = 1$, $r_s = 6$

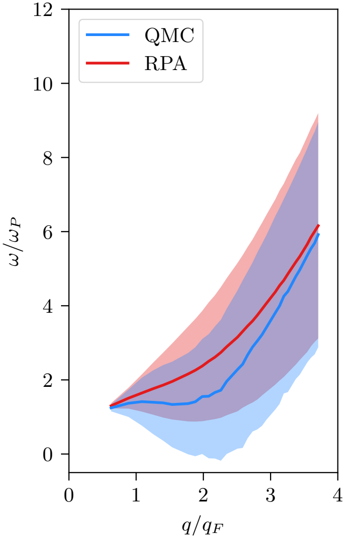
T. Dornheim, S. Groth, J. Vorberger, and M. Bonitz, *Phys. Rev. Lett.* **121**, 255001 (2018)

- Slight **correlation induced redshift** for intermediate q (at small r_s)
- **Pronounced redshift** and **broadening** with increasing r_s

Dynamic structure factor of the UEG:



Peak position and FWHM:

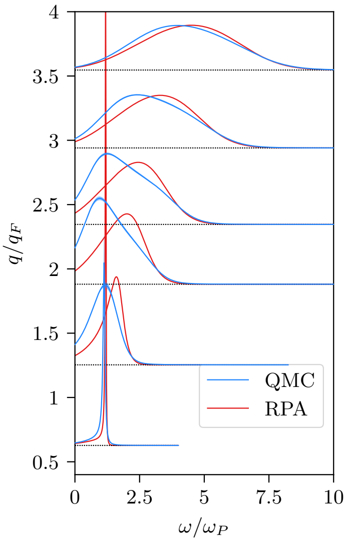


Correlation effects in the dispersion relation: $\theta = 1$, $r_s = 10$

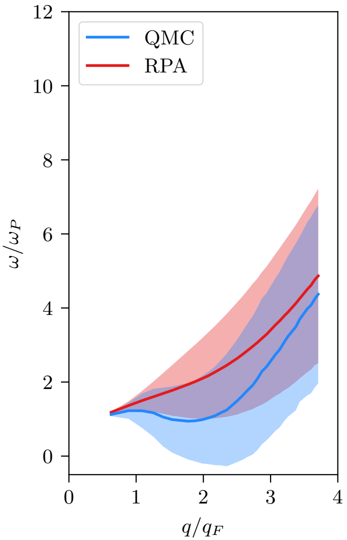
T. Dornheim, S. Groth, J. Vorberger, and M. Bonitz, *Phys. Rev. Lett.* **121**, 255001 (2018)

- Slight **correlation induced redshift** for intermediate q (at small r_s)
- **Pronounced redshift** and **broadening** with increasing r_s
- **Negative dispersion** for large r_s around $q = 2q_F$
- **dispersion** and $S(q,\omega)$ serve as rigorous benchmark for models

Dynamic structure factor of the UEG:

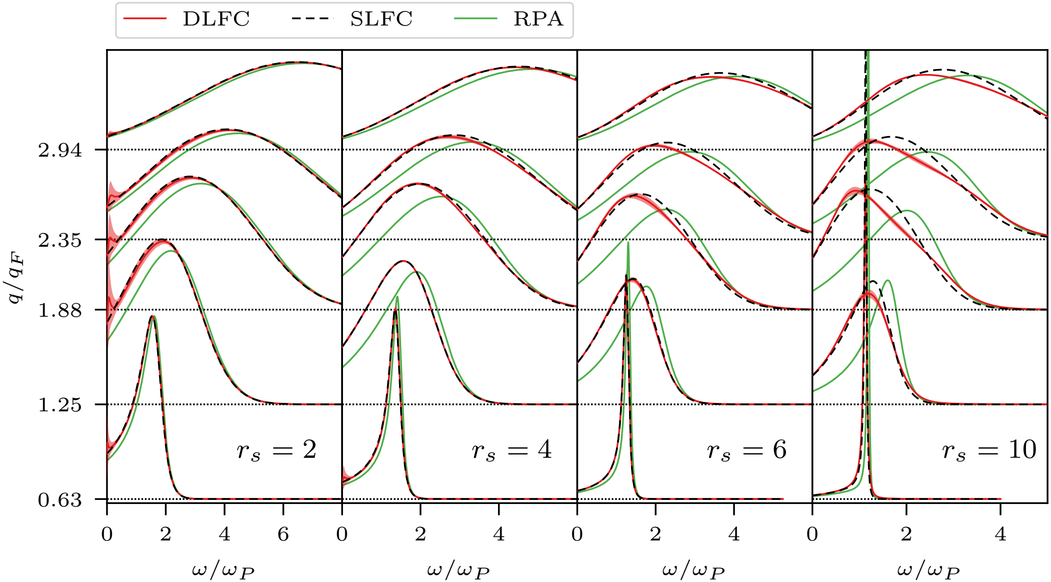


Peak position and FWHM:

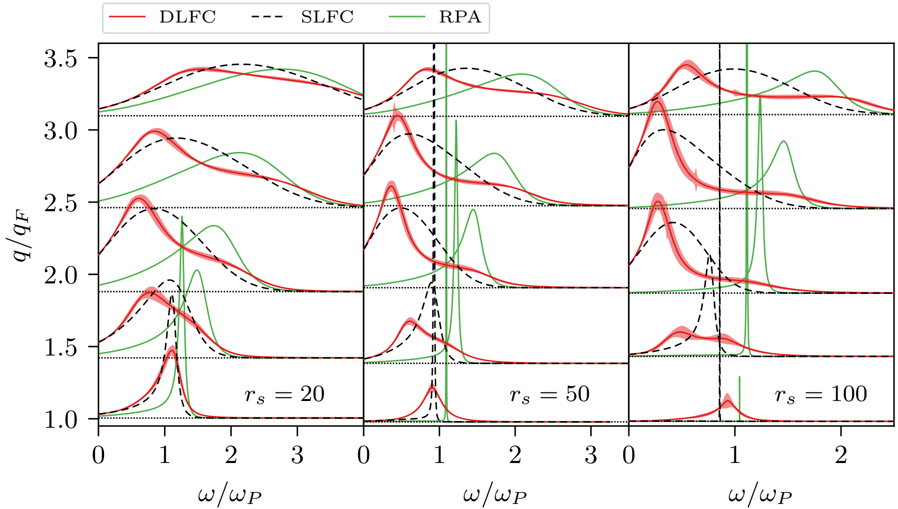


Dynamic structure factor of the UEG

T. Dornheim, S. Groth, J. Vorberger, and M. Bonitz, *Phys. Rev. Lett.* **121**, 255001 (2018)



Dynamic structure factor of the UEG



Dynamic density response function

- fluctuation-dissipation theorem

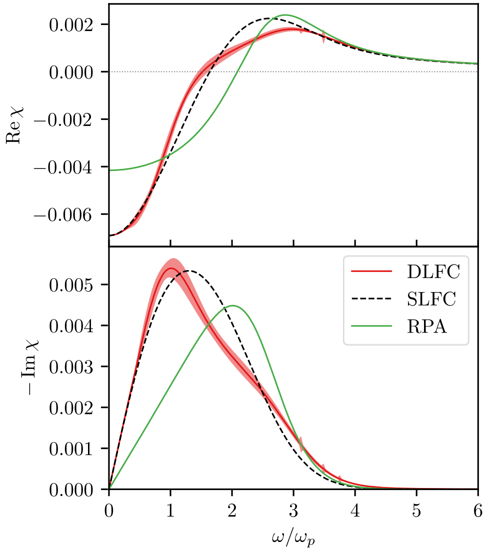
$$S(\mathbf{q},\omega) = -\frac{\text{Im}\chi(\mathbf{q},\omega)}{\pi n(1-e^{-\beta\omega})}$$

- obtain results for Re χ

$$\chi(\mathbf{q},\omega) = \frac{\chi_0(\mathbf{q},\omega)}{1 - v_q\left[1 - G(\mathbf{q},\omega)\right]\chi_0(\mathbf{q},\omega)}$$

- LFC leads to different low frequency behaviour

($r_s = 10$, $q/q_F \approx 1.88$)



Dynamic density response function

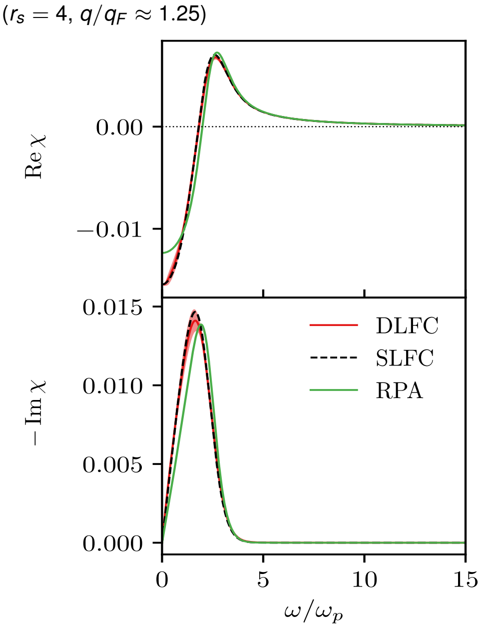
- fluctuation-dissipation theorem

$$S(\mathbf{q},\omega) = -\frac{\mathrm{Im}\chi(\mathbf{q},\omega)}{\pi n(1-e^{-\beta\omega})}$$

- obtain results for Re χ

$$\chi(\mathbf{q},\omega) = \frac{\chi_0(\mathbf{q},\omega)}{1 - v_q\left[1 - G(\mathbf{q},\omega)\right]\chi_0(\mathbf{q},\omega)}$$

- LFC leads to different low frequency behaviour
- deviations remain at weaker coupling



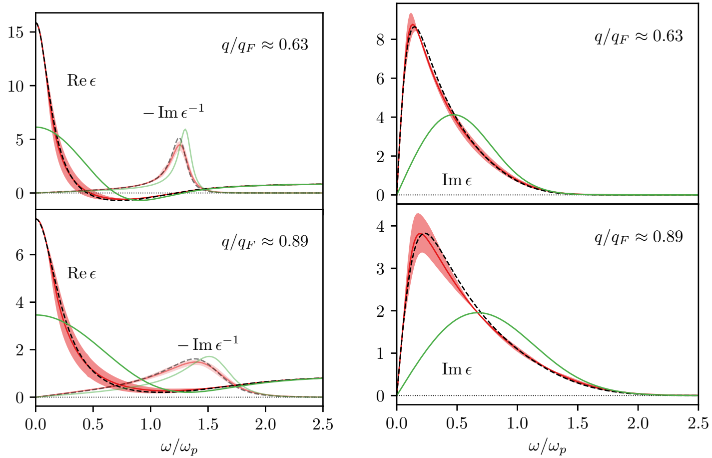
Dynamic dielectric function

- obtained from response function

$$\epsilon(\mathbf{q},\omega)^{-1} = 1 + v_q \chi(\mathbf{q},\omega)$$

- $r_s = 6$: agreement with RPA only for $\omega > \omega_P$

Dynamic dielectric function: ($\theta = 1$, $r_s = 6$)



Dynamic dielectric function

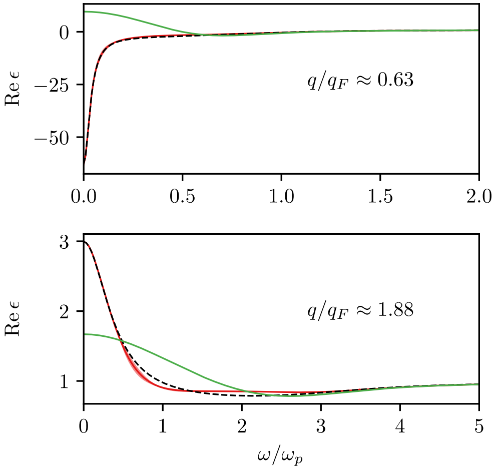
- obtained from response function

$$\epsilon(\mathbf{q},\omega)^{-1} = 1 + v_q \chi(\mathbf{q},\omega)$$

- $r_s = 6$: agreement with RPA only for $\omega > \omega_P$

- $r_s = 10$: negative static limit

Dynamic dielectric function: ($\theta = 1$, $r_s = 10$)



Dielectric function: plasmons

- existence of longitudinal plasma oscillations follows from zeroes of the dielectric function

$$\mathrm{Re}\,\epsilon = \mathrm{Im}\,\epsilon = 0$$

- undampened solutions only at $T = 0$
- poles of the response function at complex frequencies $z = \omega - i\gamma$

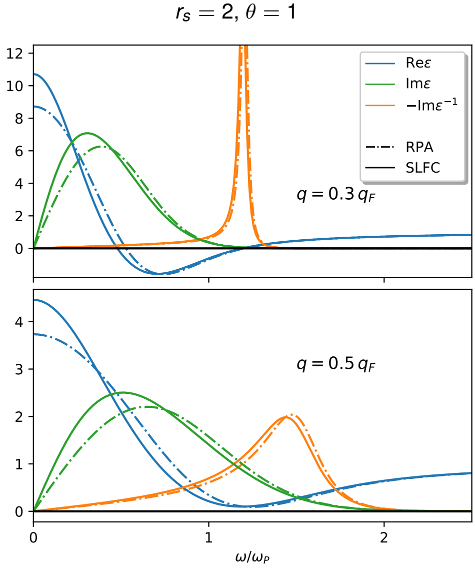
$$\delta(z - z') \leftrightarrow e^{i\omega t} e^{-\gamma t}$$

show up as peaks in $S(\mathbf{q}, \omega)$

- small damping approximation:

$$\mathrm{Re}\,\epsilon(\mathbf{q}, \omega) = 0$$

$$\gamma(q) = \frac{\mathrm{Im}\,\epsilon[\omega(q), q]}{\frac{\partial}{\partial\omega}\mathrm{Re}\,\epsilon[\omega(q), q]}\,, \quad |\gamma(q)| \ll \omega(q)$$



Dielectric function: plasmons

- existence of longitudinal plasma oscillations follows from zeroes of the dielectric function

$$\mathrm{Re}\,\epsilon = \mathrm{Im}\,\epsilon = 0$$

- undampened solutions only at $T = 0$
- poles of the respo
frequencies $z = $

$\delta(z -$

show up as peaks in $S(\mathbf{q}, \omega)$

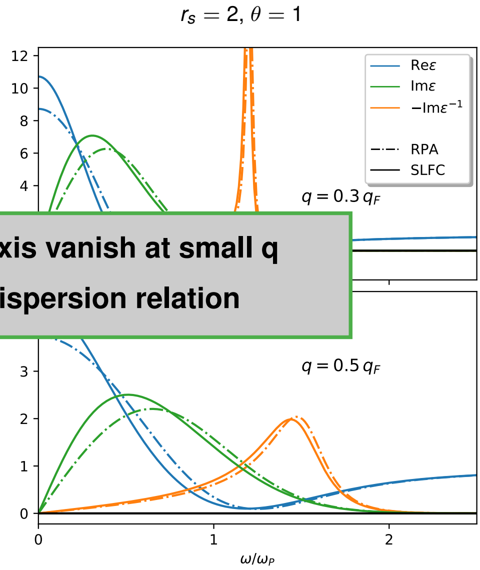
- small damping approximation:

$$\mathrm{Re}\,\epsilon(\mathbf{q}, \omega) = 0$$

$$\gamma(q) = \frac{\mathrm{Im}\,\epsilon[\omega(q), q]}{\frac{\partial}{\partial\omega}\mathrm{Re}\,\epsilon[\omega(q), q]}\,, \quad |\gamma(q)| \ll \omega(q)$$

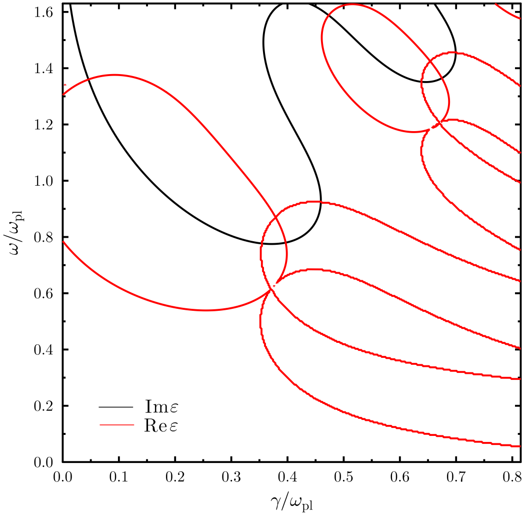
$T \gg 0$: solutions on real axis vanish at small q

$\rightarrow$ consider full complex dispersion relation

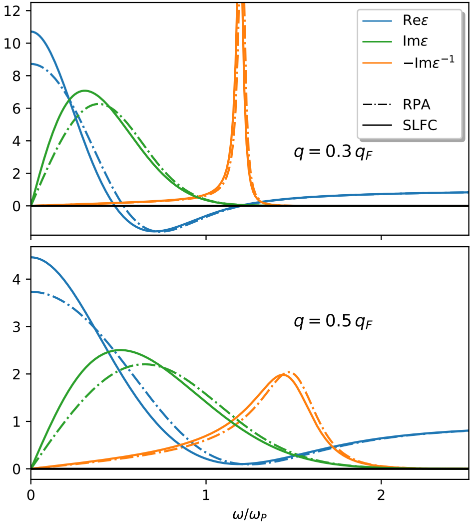


Dielectric function: plasmons

Retarded dielectric function (RPA) in the complex plane:



$r_s = 2$, $\theta = 1$



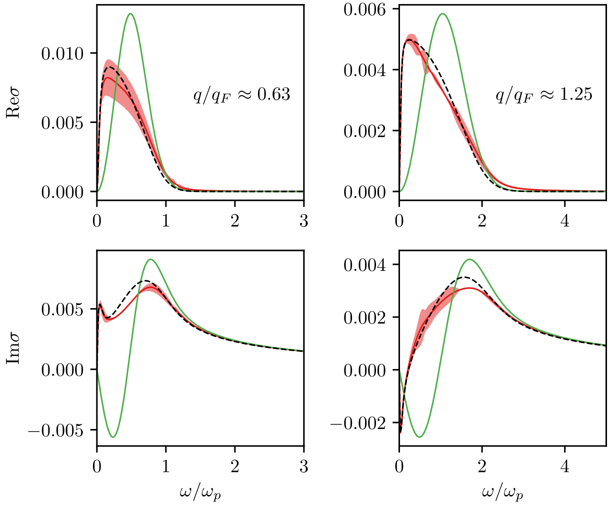
Dynamic Conductivity

- related to dielectric function

$$\epsilon(q,\omega) = 1 + \frac{4\pi i}{\omega}\sigma(q,\omega)\,.$$

- proof of concept for applications to two-component systems

Dynamic conductivity of the UEG: $\theta = 1$, $r_s = 10$



Summary

- ab-initio results for dynamic properties of the warm dense electron gas
- static LFC nearly exact for $r_s \lesssim 5$ (neural net parametrization available!)
- significant improvement compared to RPA
- apply method to QMC results for two-component systems
- investigate other imaginary-time correlation functions

Thank you for your attention!

Questions?