Perspectives of quantum plasma and warm dense matter theory[1]

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[3] Department of Physics, Imperial College London, UK

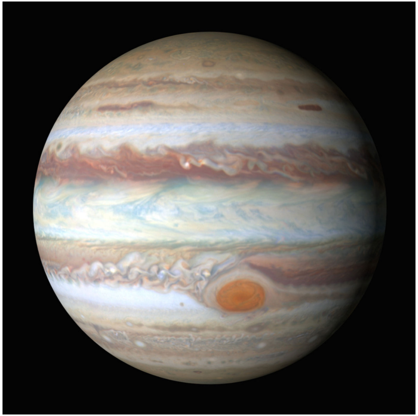
LAP Seminar, October 2020

DFG DAAD

[1] http://www.theo-physik.uni-kiel.de/bonitz/index.html ⇒ Research ⇒ Publications, Talks

Warm Dense Matter: Occurences and Applications

- **Astrophysics:**
  - Giant planet interiors (e.g. Jupiter)
  - Brown dwarfs
  - Meteor Impacts



Source: Sci-News.com [Img4]

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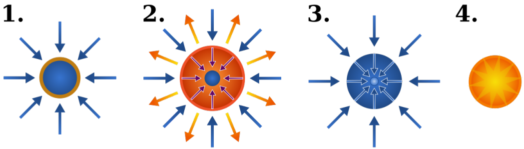
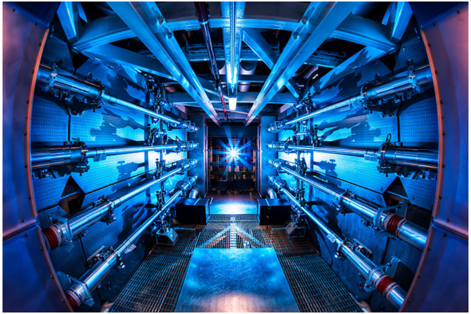
- **Experiments:**
  - Inertial confinement fusion

  Potential abundance of clean energy!





Source: en.wikipedia.org [Img5] and arstechnica.com [Img6]

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NIF, Omega (Rochester), LCLS (Stanford): Fundamental research into WDM properties: $\rightarrow$ Equation of state, $S(\mathbf{q},\omega)$, conductivity etc.

National Ignition Facility (Livermore, California)

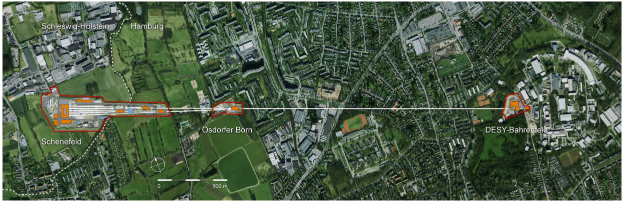
area: 70000m^2
cost: $\sim$1 billion Dollar
Source: C. Stolz, *Phil. Trans. R. Soc. A* **370**, 4115 (2012) [Img7]

Facilities for WDM experiments in Europe:

High intensity laser facilities: UK, France, ELI... hot (HED) matter

European XFEL:

- **E**uropean **X**-ray **F**ree-**E**lectron **L**aser, Hamburg – Schenefeld
- Total cost $\sim$ 1.2 billion Euro
- HIBEF Beamline and consortium (DiPOLE laser contributed from UK)



source: photon-science.desy.de

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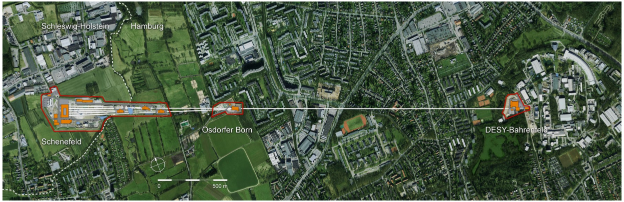

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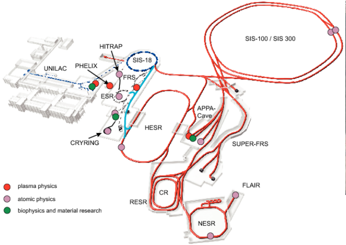
FAIR:

- **F**acility for **A**ntiproton and **I**on **R**esearch, Darmstadt
- Construction started in 2017
- Total cost $\sim$ 1.6 billion Euro
- Heavy ion beams: Isochoric heating up to $\sim 10^6 K$



source: inspirehep.net

source: dw.com

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Warm dense matter: indeed a HOT topic

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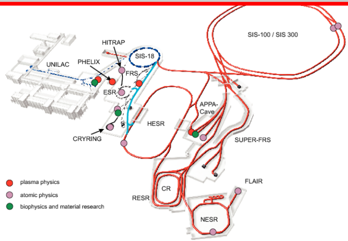

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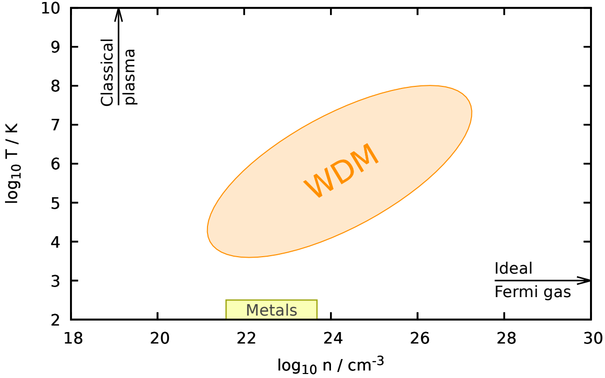
Warm Dense Matter and quantum plasmas: relevant parameters

- **Extreme and exotic state of matter:**
  $\rightarrow$ High temperature: $T \sim 10^3 - 10^8\ K$
  $\rightarrow$ Extreme density: $n \sim 10^{21} - 10^{27}\ cm^{-3}$

Source: T. Dornheim, S. Groth, and M. Bonitz, *Phys. Reports* **744**, 1-86 (2018)



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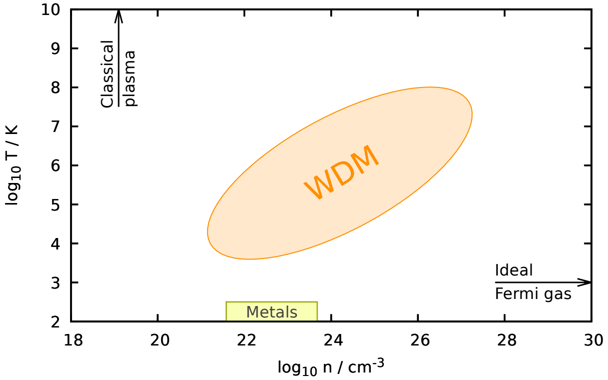

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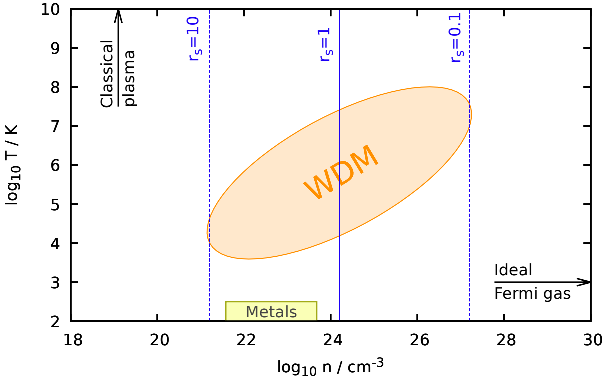

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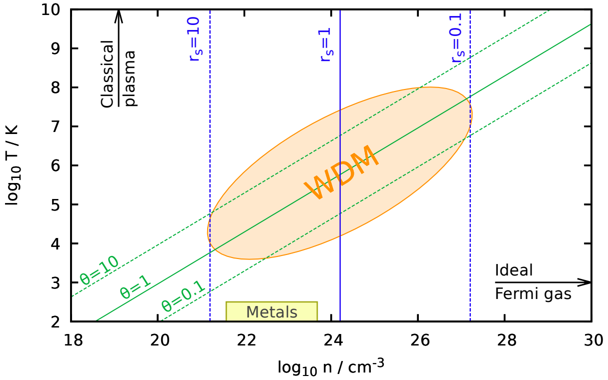

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    Classical coupling parameter $\Gamma = e^2 / r_s k_B T \sim 1$

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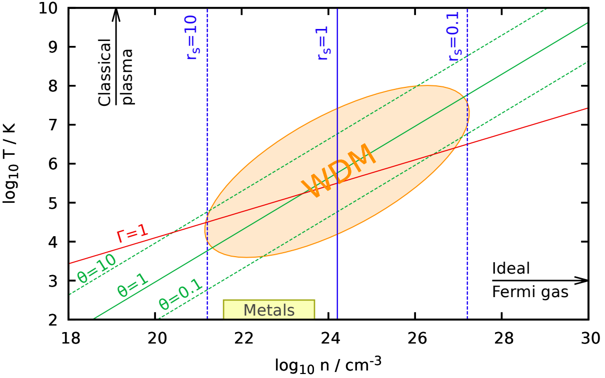

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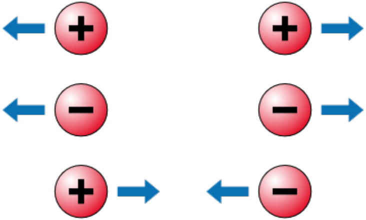

Source: bin-br.at [Img1]

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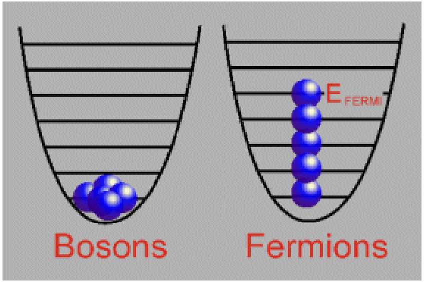

Source: cidehom.com [Img2]

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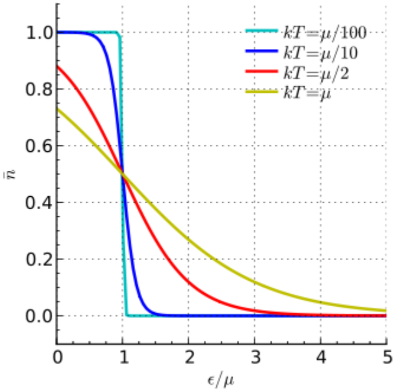
- **Nontrivial interplay of many effects:**
  - Coulomb coupling (non-ideality)
  - Fermionic exchange (anti-symmetry)
  - Thermal excitations (statistical description)



Source: en.wikipedia.org [Img3]

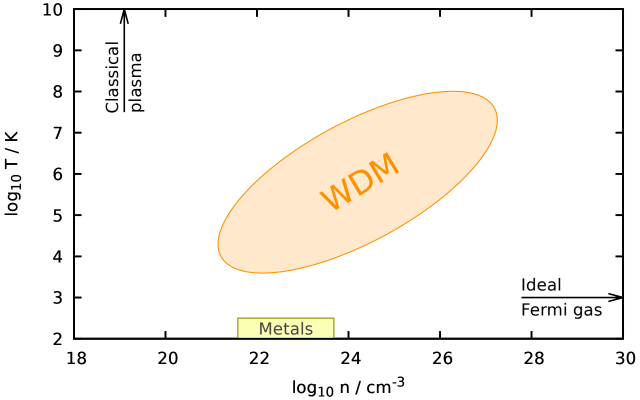
Introduction: how to theoretically approach warm dense matter?

**Warm dense matter (WDM) =
highly complex mix of ...**

- ... gas phase (atoms, molecules) and plasma: partial ionization, differently charged ions etc.
- ... condensed (crystalline or liquid) phase and gas (plasma) phase

WDM often subject to strong excitation ...

- ... mix of ground state and highly excited phases
- complex time evolution possible



Theoretical strategies: 1. Make a complex (but bad) model of the entire mess (standard), or

2. Perform an excellent description of one piece of it (our approach)
$\Rightarrow$ Series of recent breakthroughs:
from thermodynamic to dielectric and transport properties

Importance of the uniform electron gas (UEG)

Model system of Coulomb interacting quantum electrons in a uniform positive background

Ground state ($T = 0$):

- Simple model for conduction electrons in metals
- **Exchange-correlation (XC) energy**:

$$e_{xc}(r_s) = e_{tot}(r_s) - e_0(r_s)$$

$\rightarrow$ Input for density functional theory (DFT) simulations (in LDA and GGA)
$\rightarrow$ Parametrization[1] of $e_{xc}(r_s)$ from ground state quantum Monte Carlo data[2]
$\rightarrow$ this made DFT-MD the basis of modern atomic, molecular physics, chemistry, material science

[1] J.P. Perdew and A. Zunger, PRB **23**, 5048 (1981) [2] D.M. Ceperley and B. Alder, PRL **45**, 566 (1980) [3] N.D. Mermin, Phys. Rev **137**, A1441 (1965)
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Warm dense matter ($T \sim T_F$):

- **Thermal DFT**[3]: minimize free energy $F = E - TS$
  $\rightarrow$ Requires parametrization of XC **free** energy of UEG:

$$f_{xc}(r_s, \theta) = f_{tot}(r_s, \theta) - f_0(r_s, \theta)$$

- $f_{xc}(r_s, \theta)$ direct input for **EOS models** of astrophysical objects[4]
- $f_{xc}(r_s, \theta)$ contains **complete thermodynamic information** of UEG

[1] J.P. Perdew and A. Zunger, PRB **23**, 5048 (1981) [2] D.M. Ceperley and B. Alder, PRL **45**, 566 (1980) [3] N.D. Mermin, Phys. Rev **137**, A1441 (1965)
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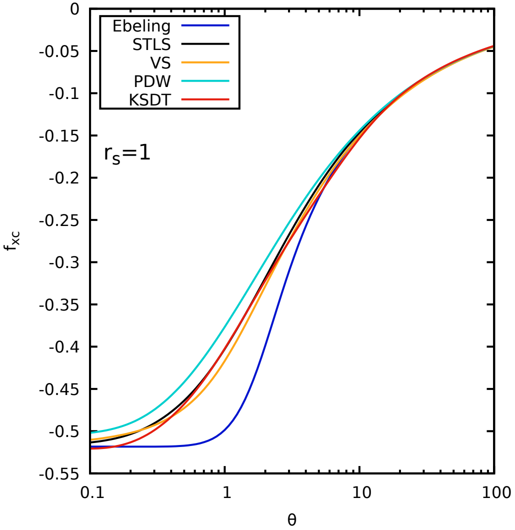
Many parametrizations for f_{xc} based on different approximate approaches:

- Semi-analytical approaches by **Ebeling**[1]
- Dielectric methods, e.g. Singwi-Tosi-Land-Sjölander[2] (**STLS**) and Vashista-Singwi[3] (**VS**)
- Quantum-classical mappings, e.g. Perrot and Dharma-wardana[4] (**PDW**)
- **Most recent:** Fit by Karasiev[5] *et al.* (**KSDT**) to Restricted Path Integral Monte Carlo (**RPIMC**) data[6]



[1] W. Ebeling and H. Lehmann, Ann. Phys. **45**, (1988)
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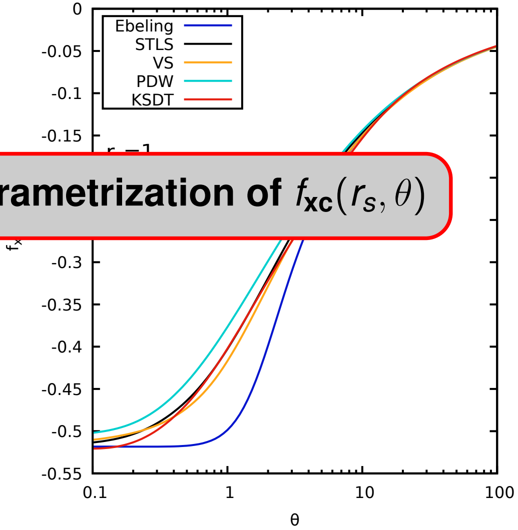
Goal 1: obtain *ab initio* parametrization of $f_{xc}(r_s, \theta)$



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Path Integral Monte Carlo (PIMC): Basic idea

- **Thermodynamic Equilibrium:** all properties can be computed from the (canonical) partition function Z

$$\hat{H} = \hat{K} + \hat{V}\,, \quad [\hat{K}, \hat{V}] \neq 0\,, \quad \beta = 1/k_{\mathrm{B}}T\,,$$
$$Z = (\mathrm{Tr}\,\hat{\rho})^{\pm}\,, \quad \hat{\rho} = e^{-\beta\hat{H}} = [e^{-\hat{H}/Pk_BT}]^P\,,$$

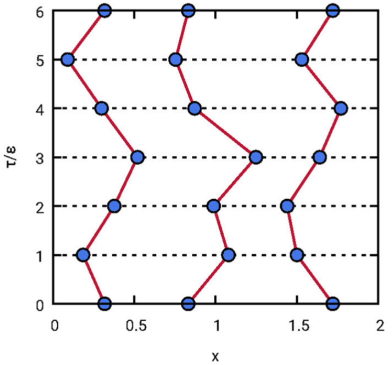
- N spin-polarized fermions in coordinate space, $\mathbf{R} = \{\mathbf{r}_1, \ldots, \mathbf{r}_N\}$

$$Z = \frac{1}{N!} \sum_{\sigma \in S_N} \mathrm{sgn}(\sigma) \int \mathrm{d}\mathbf{R}\ \langle \mathbf{R}|\, e^{-\beta\hat{H}}\, |\hat{\pi}_\sigma \mathbf{R}\rangle$$

- Express the density matrix as a path over P sets of particle coordinates at P times higher temperature

- The partition function is the sum over all closed paths $\mathbf{X} = \{\mathbf{R}_0, \ldots, \mathbf{R}_{P-1}\}$ in "imaginary time", with P "time slices"

$$Z = \sum_{\mathbf{X}} W(\mathbf{X}) \quad , \quad W(\mathbf{X})\text{: configuration weight of path } \mathbf{X}$$



PIMC configuration of $N = 3$ particles

Taken from: T. Dornheim, S. Groth, A. Filinov, and M. Bonitz, *J. Chem. Phys.* **151**, 014108 (2019)

Based on R. Feynman's path integral quantum mechanics

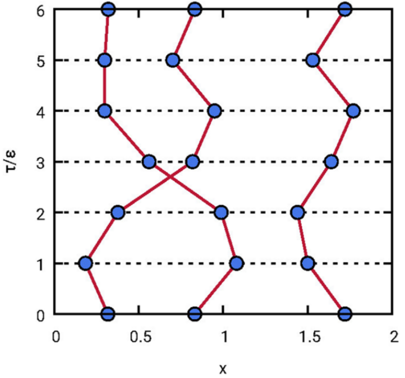
PIMC: parameter-free, potentially exact method!

Path Integral Monte Carlo (PIMC): Fermions

- **Fermionic antisymmetry:**

$$Z = \frac{1}{N!} \sum_{\sigma \in S_N} \mathrm{sgn}(\sigma) \int d\mathbf{R} \ \langle \mathbf{R} | \, e^{-\beta \hat{H}} \, | \hat{\pi}_\sigma \mathbf{R} \rangle$$

$\Rightarrow$ We must include **permutation-cycles**!



PIMC configuration of $N = 3$ particles, $W(\mathbf{X}) < 0$

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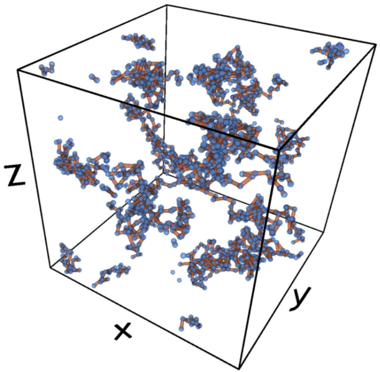
- Randomly generate all possible paths **X** using the **Metropolis algorithm**



Snapshot of PIMC simulation of UEG with $N = 19$, $r_s = 2$, $\theta = 0.5$

Taken from: T. Dornheim, S. Groth, A. Filinov, and M. Bonitz, *J. Chem. Phys.* **151**, 014108 (2019)

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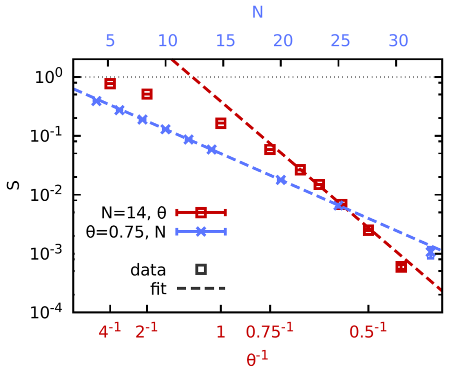
- Sign changes due to particle exchange lead to vanishing signal-to-noise ratio

$\Rightarrow$ **Fermion Sign Problem**



Exponential decrease of the average sign S with system size N and quantum degeneracy θ^{-1}

Taken from: T. Dornheim, *Phys. Rev. E* **100**, 023307 (2019)

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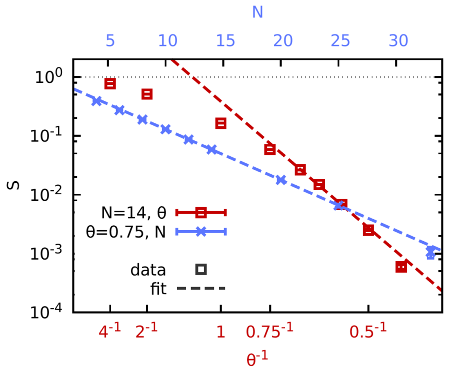

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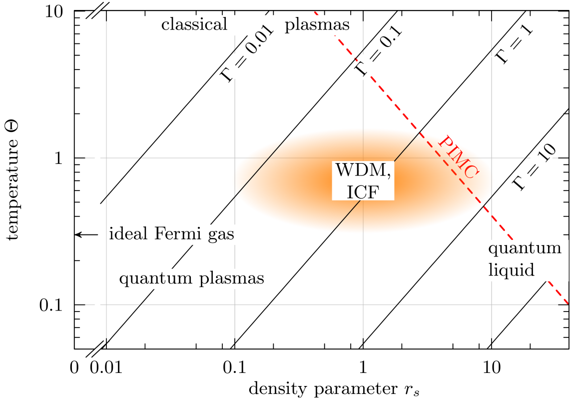
PIMC simulations of WDM very challenging!

Path integral Monte Carlo (PIMC) simulation of the warm dense UEG

- Standard PIMC in warm dense regime severely hampered by ***fermion sign problem***:



[1] E.W. Brown *et al.*, PRL **110**, 146405 (2013) [2] T. Schoof *et al.*, Phys. Rev. Lett **115**, 130402 (2015) [3] T. Schoof *et al.*, Contrib. Plasma Phys. **51**, 687 (2011)
[4] T. Dornheim *et al.*, New J. Phys. **17**, 073017 (2015) [5] T. Dornheim *et al.*, J. Chem. Phys. **143**, 204101 (2015)

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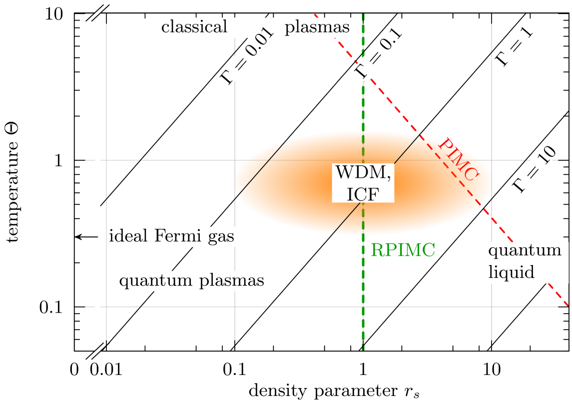
  - First results[1] by E. Brown, D. Ceperley *et al.* (2013) based on ***fixed node approximation*** (**RPIMC**)
  - Induces **systematic errors** of unknown magnitude
  - **RPIMC** limited to $r_s \gtrsim 1$



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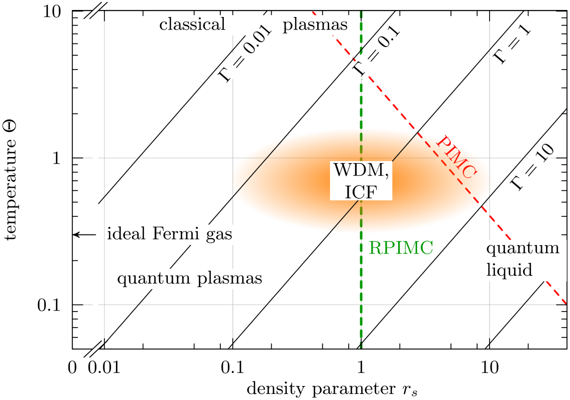

Our approach:

Avoid fermion sign problem by combining two exact and complementary QMC methods:

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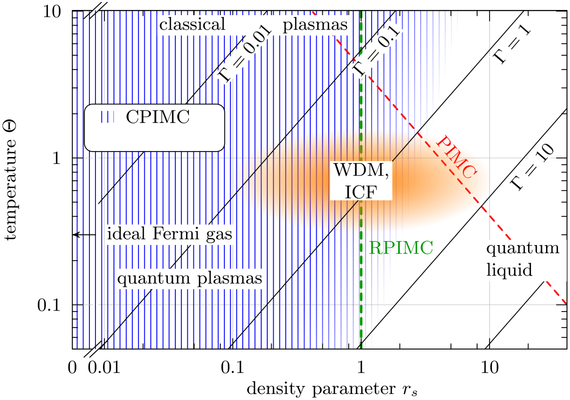

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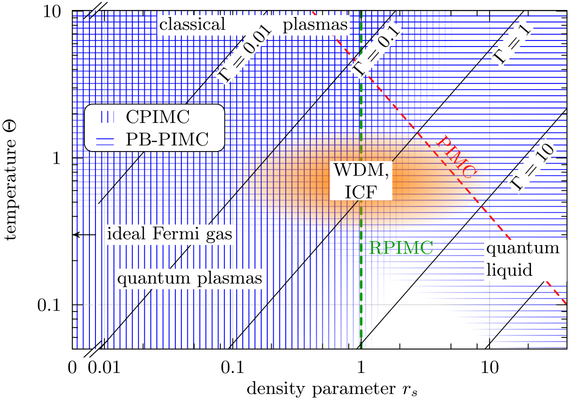

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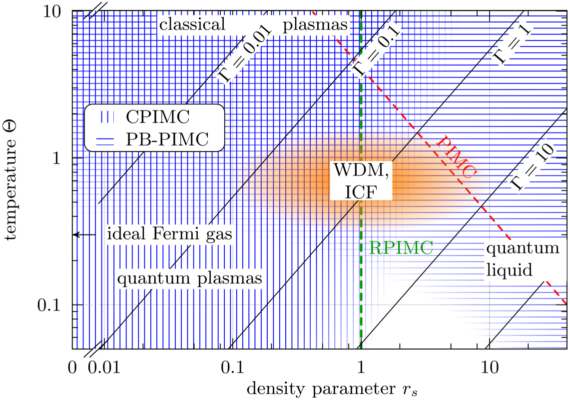

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***Ab initio* simulations over broad range of parameters possible**

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1. Exact exchange-correlation energy $E_{xc} = E - E_0$ (E_0: ideal energy)
($N = 33$ spin-polarized electrons, $\theta \geq 0.5$, $\forall\ r_s$)

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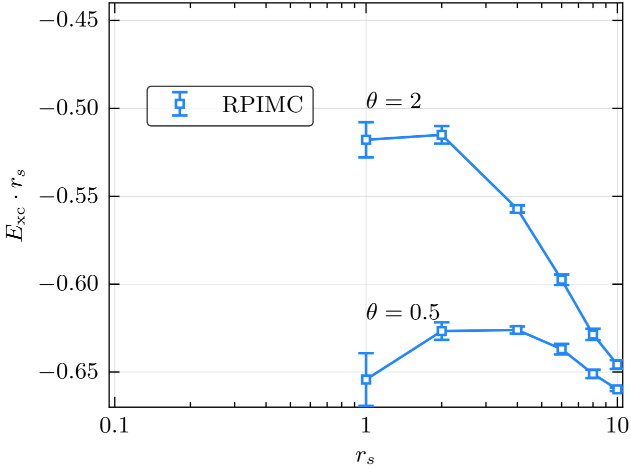

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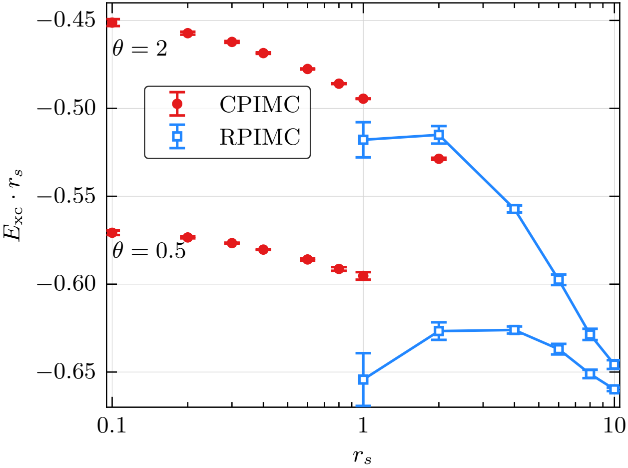

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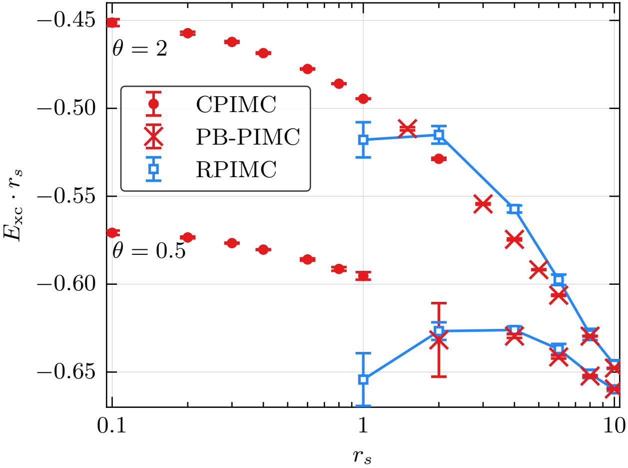

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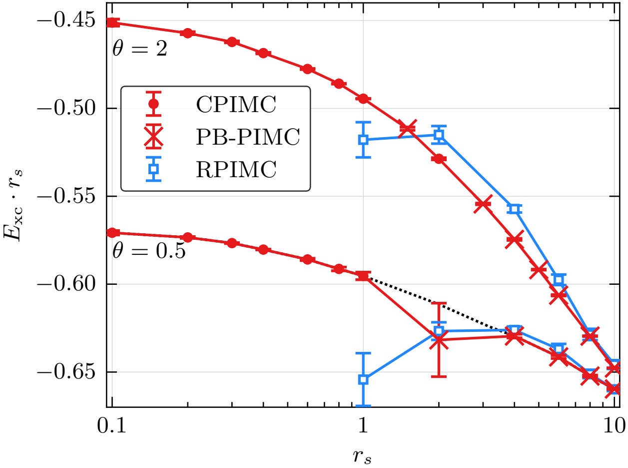
> **Combination[1] yields exact results over entire density range down to $\theta \sim 0.5$**

- Also applies to the **unpolarized** UEG[2]
- confirmed by independent **DMQMC** simulations[3]
- Extended to TD Limit[4] and to the ground state[5]
- Analytical parametrization of $f_{xc}(r_s, \theta, \xi)$, with error below 0.3%, Integrated in standard DFT libraries[5]



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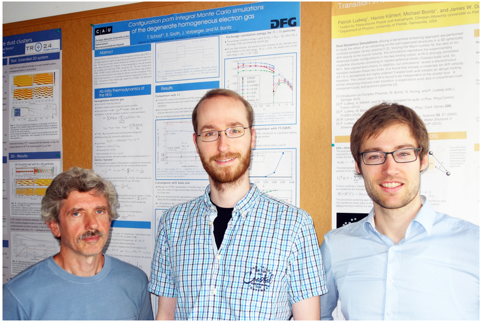
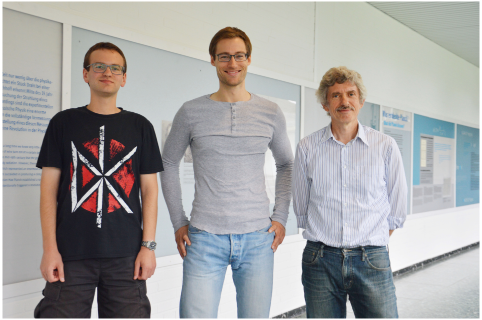
Acknowledgements to those who did most of the work...





Tim Schoof (PhD 2016), **Simon Groth** (PhD 2018):
CPIMC, finite size corrections etc.

Tobias Dornheim (PhD 2018): PB-PIMC
now at CASUS Görlitz and Helmholtz-Zentrum Dresden
Extension to static and dynamic response, transport, DFT, machine learning etc.

Recent review: T. Dornheim, S. Groth, and M. Bonitz, Physics Reports **744**, 1-86 (2018)
Photos: J. Siekmann

Ab Initio PIMC approach to equilibrium response and transport properties

Quantities accessible in PIMC:

all thermodynamic functions from $F(r_s, \theta)$; structural properties: $g(r)$, $S(q)$

fluctuations in response to excitation: $\delta\hat{H}(\mathbf{q}) \longrightarrow \delta\rho(\mathbf{q})$

correlation functions: e.g. $\langle\delta\rho(\mathbf{q}_1, \tau_1)\rho(\mathbf{q}_2, \tau_2)\rangle$ yield transport properties

Susceptibilites from linear response theory (LRT):

$\delta\rho(\mathbf{q}) = \chi(\mathbf{q})\delta H(\mathbf{q})$, χ: static density response $\longrightarrow$ comparison for PIMC to LRT/experiment

Correlation and exchange effects: encoded in “local field correction” $G(\mathbf{q}, \omega)$

straightforward connection to transport, optics etc.: $\chi(\mathbf{q}, \omega), S(\mathbf{q}, \omega), \epsilon(\mathbf{q}, \omega), \sigma(\mathbf{q}, \omega)$

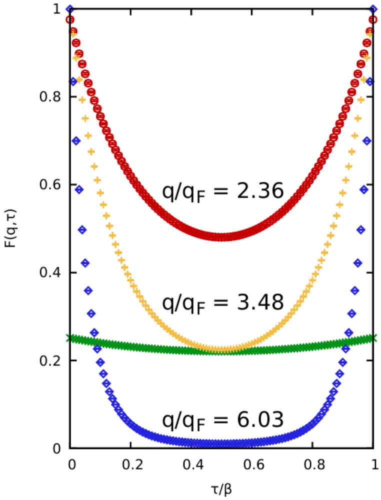
PIMC: susceptibilities beyond validity limits of LRT

2. The Static Local Field Correction: *Ab initio* PIMC Simulations

- PIMC gives direct access to imaginary-time density–density correlation function:

$$F(\mathbf{q}, \tau) = \frac{1}{N} \langle \rho(\mathbf{q}, \tau) \rho(-\mathbf{q}, 0) \rangle$$

S. Groth, T. Dornheim, and J. Vorberger, *Phys. Rev. B* **99**, 235122 (2019)



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

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

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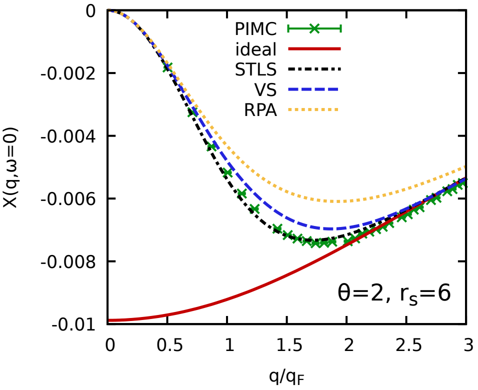

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- $G(q)$ can be obtained as the deviation from $\chi_0(q)$:

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

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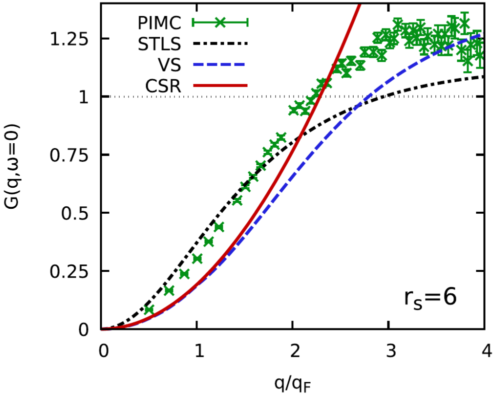
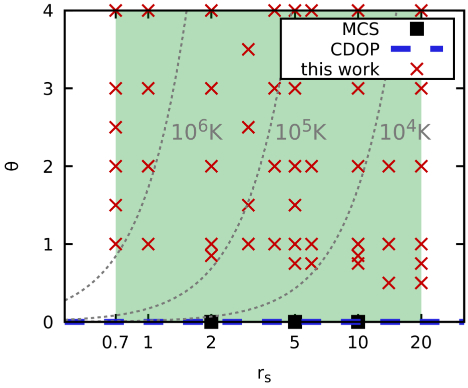


The Static Local Field Correction: Neural-net representation

Extensive set of new PIMC data

Source: T. Dornheim *et al.*, J. Chem. Phys. (2019)

- QMC data available at discrete grid (q; θ, r_s)



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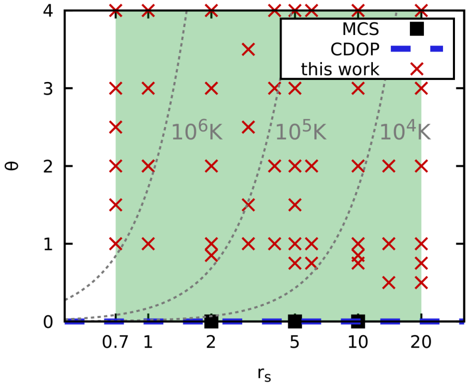

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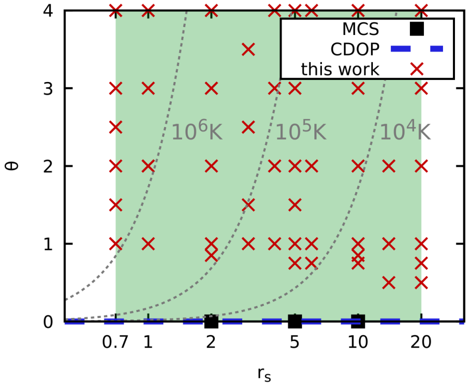

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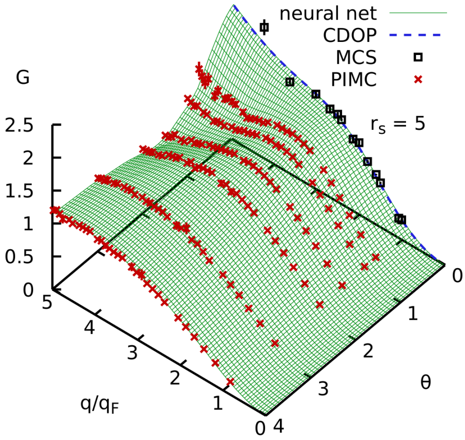

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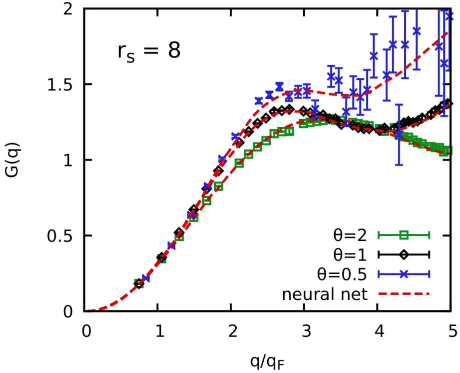

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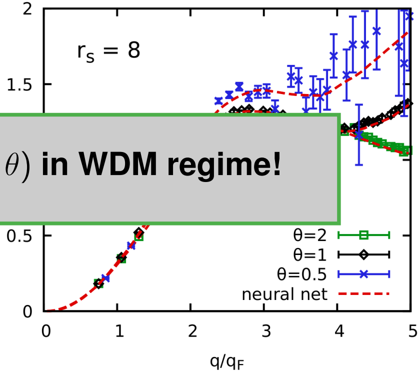

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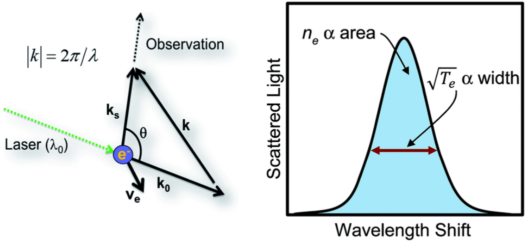
3. *Ab initio* dynamic (ω-dependent) results for the warm dense UEG

- **Key quantity:** dynamic structure factor

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

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



X-ray Thomson scattering experiments at free electron laser facilities (e.g. FLASH, X-FEL, LCLS)

yields the most accurate information on plasma density, ionic charge state, and temperature

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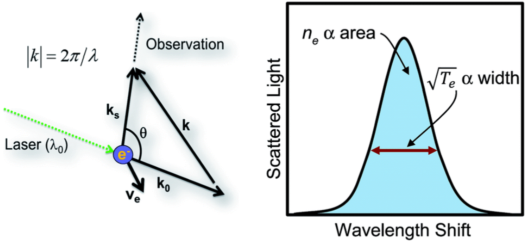

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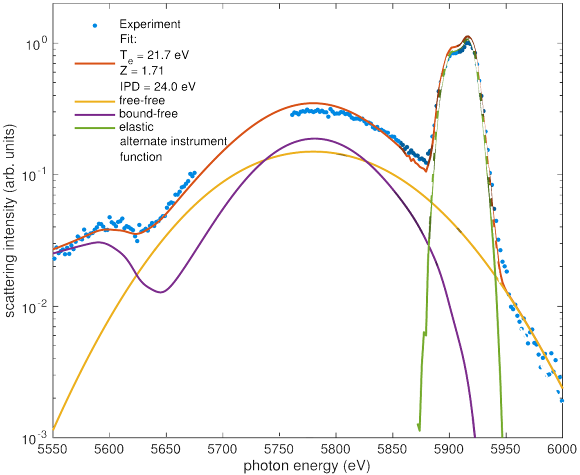
- **Practical example:** Fit model for $S(\mathbf{q},\omega;T_e)$ to spectrum to determine electron temperature T_e



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

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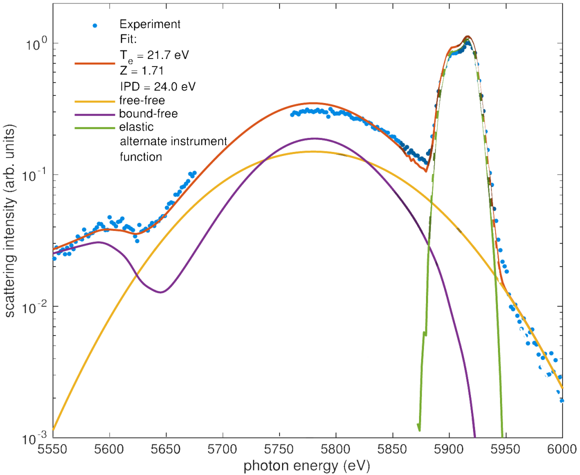
- **Practical example:** Fit model for $S(\mathbf{q},\omega;T_e)$ to spectrum to determine electron temperature T_e
- **Problem:**
  $F(\mathbf{q},t)$ requires **real time-dependent simulations**
  $\rightarrow$ with PIMC have to use analytic continuation, reconstruct $F(q,it)$ and 4 frequency moments, but: insufficient information



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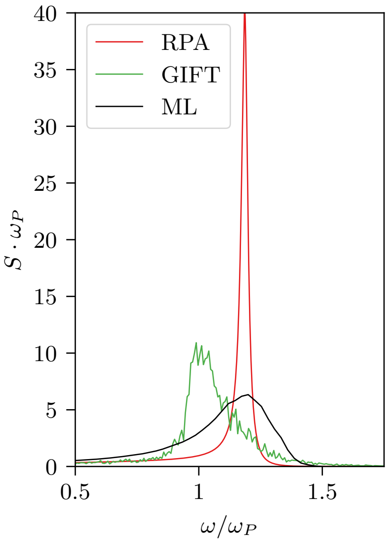
Incorporating additional information on $S(\mathbf{q},\omega)$ via dielectric formulation

- **Fluctuation-dissipation theorem:**

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

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



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- **Fluctuation-dissipation theorem:**

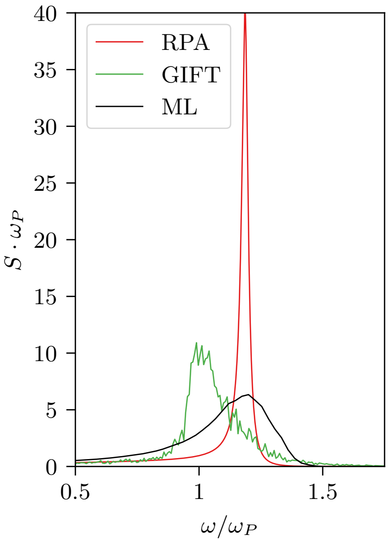
$$S(\mathbf{q}, \omega) = -\frac{\mathrm{Im}\chi(\mathbf{q}, \omega)}{\pi n(1 - e^{-\beta\omega})}$$

- 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):** $G \equiv 0$

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



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:

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

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

Advantages:

- Limits $G(\mathbf{q},0)$ and $G(\mathbf{q},\infty)$ known from PIMC simulation
- Other exact properties of G can be incorporated

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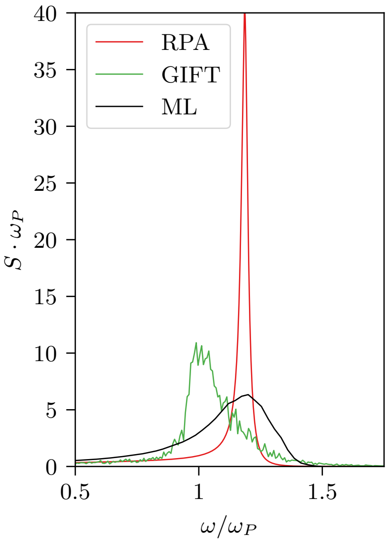

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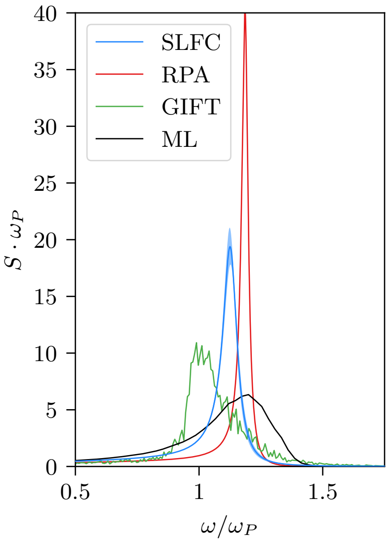

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- Ex… dy…

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GIFT and ML spectra not in agreement with exact properties of $G(\mathbf{q},\omega)$
$\rightarrow$ to be discarded as unphysical

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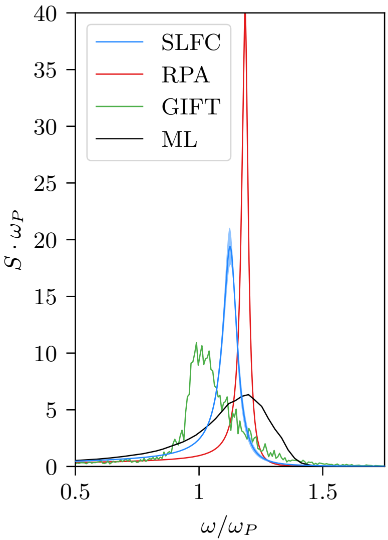

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dy…

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

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

3. Stochastic sampling of $G(\mathbf{q},\omega)$ accurately determines $S(\mathbf{q},\omega)$

Advantages:

- Limits $G(\mathbf{q},0)$ and $G(\mathbf{q},\infty)$ known from PIMC simulation
- Other exact properties of G can be incorporated

Dynamic structure factor of the UEG:
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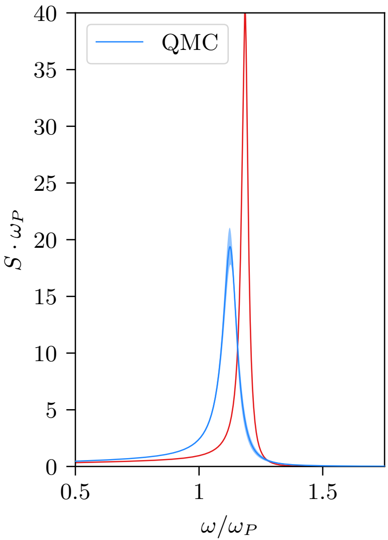
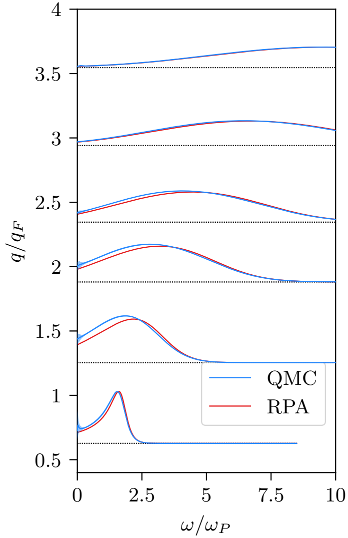


Correlation effects in the peak position of $S(q,\omega)$: $\theta = 1$, $r_s = 2$

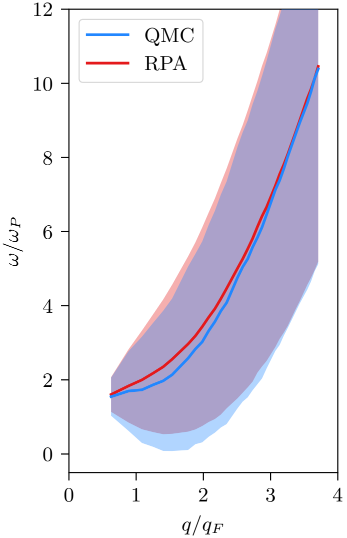
T. Dornheim, S. Groth, J. Vorberger, and M. Bonitz, *Phys. Rev. Lett.* **121**, 255001 (2018)

- *Ab initio* results for $G(q,0)$ available: Dornheim *et al.*, J. Chem. Phys. (2019)
- Slight **correlation induced redshift** of peak for intermediate q (at small r_s)

Dynamic structure factor of the UEG:



Peak position and FWHM:



Correlation effects in the peak position of $S(q,\omega)$: $\theta = 1$, $r_s = 6$

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

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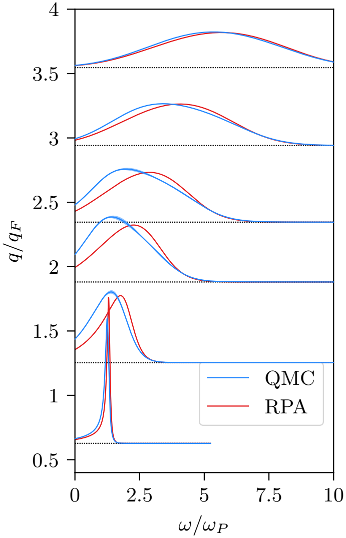
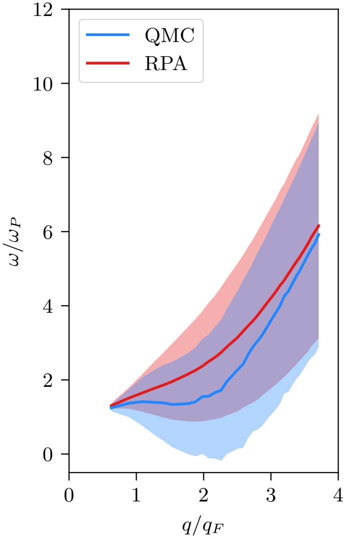
- Slight **correlation induced redshift** of peak for intermediate q (at small r_s)
- **Pronounced redshift** and **broadening** with increasing r_s

Dynamic structure factor of the UEG:



Peak position and FWHM:



Correlation effects in the peak position of $S(q,\omega)$: $\theta = 1$, $r_s = 10$

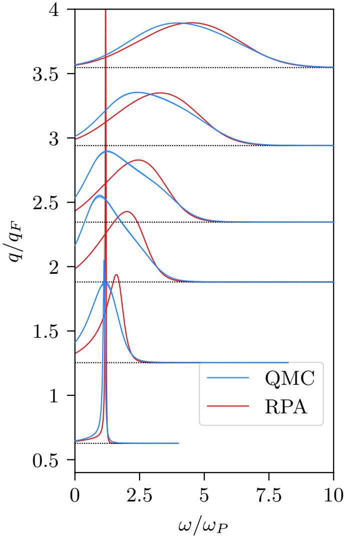
T. Dornheim, S. Groth, J. Vorberger, and M. Bonitz, *Phys. Rev. Lett.* **121**, 255001 (2018)

- *Ab initio* results for $G(q,0)$ available: Dornheim *et al.*, J. Chem. Phys. (2019)
- Slight **correlation induced redshift** of peak for intermediate q (at small r_s)
- **Pronounced redshift** and **broadening** with increasing r_s
- **Negative dispersion of peak** for large r_s around $q = 2q_F$ **predicted for dense hydrogen**
- **How is this related to plasmons?**

Requires dielectric function $\epsilon(q,\omega)$

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

Dynamic structure factor of the UEG:



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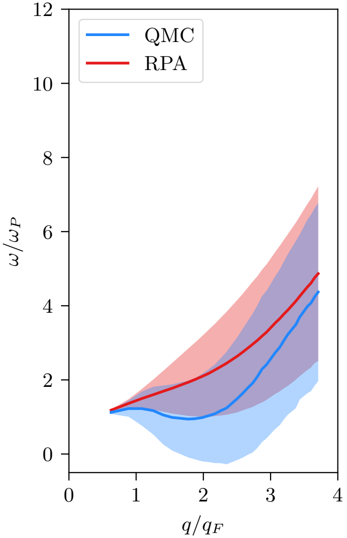

4. WDM Dielectric function: finite temperature, quantum and correlation effects

- Quantum hydrodynamics[2]: incorrect plasmon dispersion in 2D and 3D (factor $9/5$ in q^2 term)[3]
- Quantum Vlasov (Hartree, mean field or random phase) approximation (RPA) at finite T:

$$\epsilon(q,\omega;T) = 1 - \tilde{v}(q)\Pi(q,\omega;T)\,, \quad \Pi^{\mathrm{RPA}}(\vec{q},\omega;T) = \int \frac{d\vec{p}}{(2\pi)^3}\frac{f\left(E_{\vec{p}};T\right) - f\left(E_{\vec{p}+\vec{q}};T\right)}{E_{\vec{p}} - E_{\vec{p}+\vec{q}} + \omega + i\delta}\,, \qquad \delta \to 0^+\,.$$

- Mean field plus correlations: models for local field correction $G(q,\omega)$ or quantum kinetic theory:

$$\Pi^{\mathrm{RPA}} \to \Pi(q,\omega) = \frac{\Pi^{\mathrm{RPA}}(q,\omega)}{1+\tilde{v}(q)G(q,\omega)\Pi^{\mathrm{RPA}}(q,\omega)}\,.$$

- Exact results[4] : $G^{\mathrm{QMC}}(q,\omega) \to \Pi^{\mathrm{QMC}}(q,\omega) \to \epsilon^{\mathrm{QMC}}(q,\omega)$

  Accurate and efficient approximation: $G^{\mathrm{QMC}}(q,\omega) \to G^{\mathrm{QMC}}(q,0) = G(q)$, insert in $\Pi(q,\omega) \to \epsilon^{\mathrm{SLFC}}(q,\omega;T)$

- QHD with exchange-correlation corrections[5], but only: $T = 0$ and low accuracy xc effects (LDA)
- Improved QHD[6]: finite T, ω- and q-dependent coefficients, correlations via G and non-local effects

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[2] G. Manfredi and F. Haas, Phys. Rev. B (2001)
[3] M. Bonitz *et al.*, Phys. Plasmas (2019)
[4] P. Hamann *et al.*, Phys. Rev. B (2020), arXiv: 2007.15471
[5] N. Crouseilles *et al.*, Phys. Rev. B (2008)
[6] Zh. Moldabekov *et al.*, Phys. Plasmas (2018)

Parametrizations of the plasmon dispersion of the 3D electron gas (mean field)

- Bohm and Gross 1949, classical plasma[7]: $\frac{\omega^2(q)}{\omega_p^2} = 1 + \frac{v_{th}^2}{\omega_p^2} q^2, \quad v_{th}^2 = \frac{3k_B T}{m}$
- Bohm and Pines 1953, quantum plasma, $T = 0$ (RPA)[8]: $\frac{\omega^2(q)}{\omega_p^2} = 1 + \frac{3}{5}\frac{v_F^2}{\omega_p^2} q^2 + \frac{\hbar^2}{4m^2}\frac{q^4}{\omega_p^2},$
- Ferrell 1957, q^4 terms, $T = 0$[9]: $\frac{\omega^2(q)}{\omega_p^2} = 1 + \frac{3}{5}\frac{v_F^2}{\omega_p^2} q^2 + \left(\frac{(\Delta v_0^2)^2}{\omega_p^2} + \frac{\hbar^2}{4m^2}\right)\frac{q^4}{\omega_p^2}, \qquad \left(\Delta v_0^2\right)^2 = \langle v^4\rangle_0 - \langle v^2\rangle_0^2$
- Quantum hydrodynamics ($T = 0$)[10]: $\frac{\omega^2(q)}{\omega_p^2} = 1 + \frac{1}{3}\frac{v_F^2}{\omega_p^2} q^2 + \frac{\hbar^2}{4m^2}\frac{q^4}{\omega_p^2},$
- Hamann *et al.*[11] RPA, finite T : $\frac{\omega^2(q)}{\omega_p^2} = 1 + \frac{\langle v^2\rangle}{\omega_p^2} q^2 + \left(\frac{(\Delta v^2)^2}{\omega_p^2} + \frac{\hbar^2}{4m^2}\right)\frac{q^4}{\omega_p^2}, \quad \langle\ldots\rangle$ average with Fermi function

Analytical parametrization for WD UEG: $\frac{\omega^2(q)}{\omega_p^2} = 1 + B_2(r_s,\Theta)\frac{q^2}{q_F^2} + B_4(r_s,\Theta)\frac{q^4}{q_F^4}$

Note: finite q-range of plasmons to be accounted for separately

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[7] D. Bohm and E.P. Gross, Phys. Rev. (1949)
[8] D. Bohm and D. Pines, Phys. Rev. (1953), also: Lindhard, Klimontovich, Silin
[9] R.A. Ferrell, Phys. Rev. (1957)
[10] G. Manfredi and F. Haas, Phys. Rev. B (2001)
[11] P. Hamann et al., Contrib. Plasma Phys. (2020), arXiv: 2008.04605

5. Collective excitations. Plasmons

- Solution of Maxwell's equations: EM field modes, $E(q,t)$, in plasma (isotropic), from

$$\hat{\epsilon}(\vec{q}, \omega(q)) = 0$$

- contains collective excitations (plasmon)
- weak damping approximation (WDA):

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

- roots on real axis vanish for $q \geq q_{\mathrm{cr}}$, and damping, $|\mathrm{Im}\,\omega|$, becomes large
- drop WDA and find exact roots

$$\hat{\epsilon}(\vec{q}, z) = 0$$

at complex frequencies $z = \omega - i\gamma$:

$$E(q;t) \sim e^{i\omega(q)t} e^{-\gamma(q)t}, \quad \gamma > 0$$

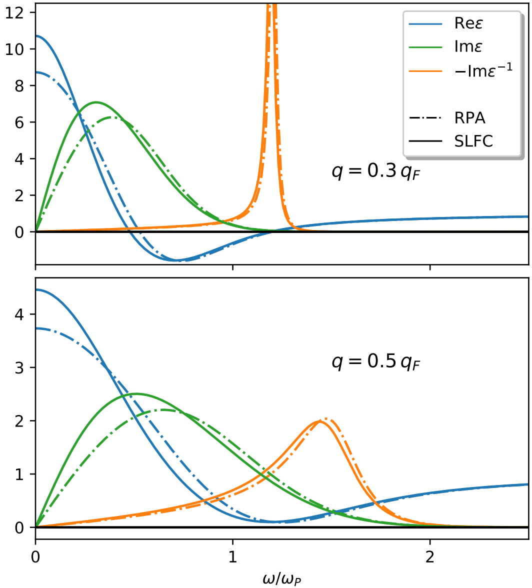
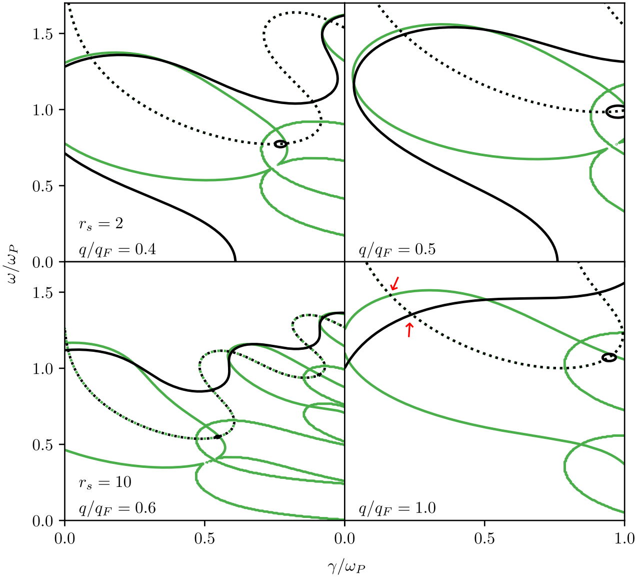


Figure: Moderately correlated electron gas, $\Theta = 1$, $r_s = 2$

Analytic continuation (AC) of the dielectric function[12]

- AC of the retarded DF into the lower frequency half plane, $\gamma > 0$.
- full lines: Re $\epsilon = 0$,
  dotted lines: Im $\epsilon = 0$,
  plasmon = intersection (arrows)
- green: mean field (RPA)
  black: correlated (static LFC)
- complex zeroes may exist, even if Re ϵ has no zeroes on real axis (top right).
- Finite temperature, $\Theta = 1$ ($k_B T = E_F$)



[12] M. Bonitz, Quantum Kinetic Theory, 2nd ed. Springer 2016; P. Hamann *et al.*, Contrib. Plasma Phys. (2020), arXiv:2008.04605

Correlation effects in plasmon dispersion and dynamic structure factor ($\Theta = 1$)

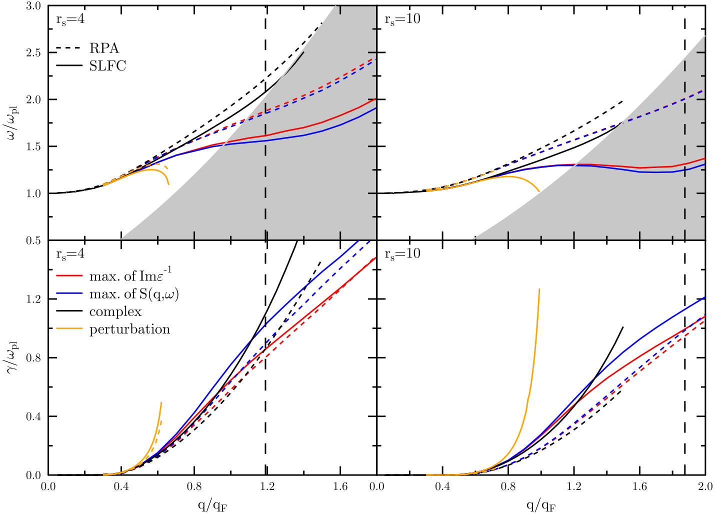
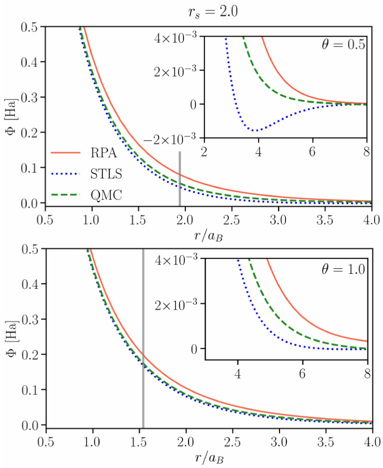


Figure: Plasmon dispersion (top) and damping (bottom) with (SLFC) and without (RPA) correlations. Orange: weak damping approximation. blue: peak of $S(q, \omega)$. Grey area: pair continuum. Vertical dashes: $2\pi/q = \lambda = \lambda_{\text{scr}}$.
From P. Hamann *et al.*, Contrib. Plasma Phys. (2020), arXiv:2008.04605

6. Nonlinear Electronic Density Response in WDM

T. Dornheim, J. Vorberger, and M. Bonitz, *Phys. Rev. Lett.* **125**, 085001 (2020)

- **Linear Response Theory (LRT)**
  implicitly assumed throughout WDM theory, including:
  - WDM diagnostics (e.g. XRTS)
  - Construction of effective potentials
  - Calculation of stopping power and conductivities
  - XC-functionals for DFT



Screened ion potential at $r_s = 2$ and $\theta = 0.5$ (top) and $\theta = 1$ (bottom). Vertical lines indicate where the impact of the ionic potential on the electrons is *small.*

Taken from: Zh. Moldabekov, T. Dornheim, and M. Bonitz, arxiv:2009.09180 (submitted)

[1] L. B. Fletcher *et al.*, *Nat. Photonics* **9**, 274 (2015)

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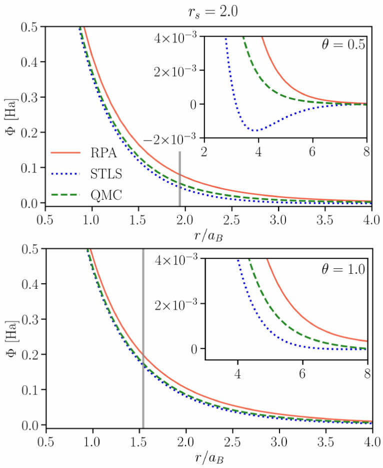
  - XC-functionals for DFT

- **Open question:** Cases of strong excitation:
  - Seeded FELs[1]: $I \sim 10^{22}\,\frac{W}{cm^2}$
  - THz lasers (high ponderomotive potential)

- **Consequences for XRTS signal, screened potentials, stopping power, etc?**



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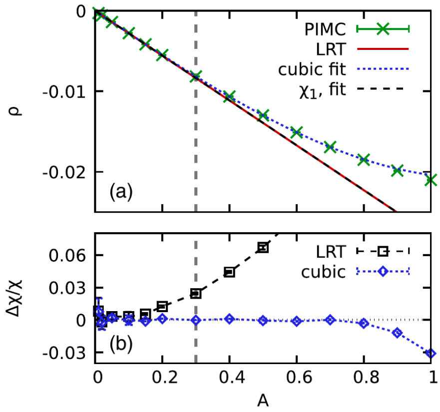
- **Simulation idea:** apply harmonic perturbation to UEG

$$\hat{H} = \hat{H}_{\text{UEG}} + 2A \sum_{j=1}^{N} \cos(\mathbf{q} \cdot \hat{\mathbf{r}}_j)$$

A: perturbation amplitude, $\mathbf{q}$: wave vector



Density response of the UEG at $r_s = 2$ and $\theta = 1$ for $q = 0.84 q_F$

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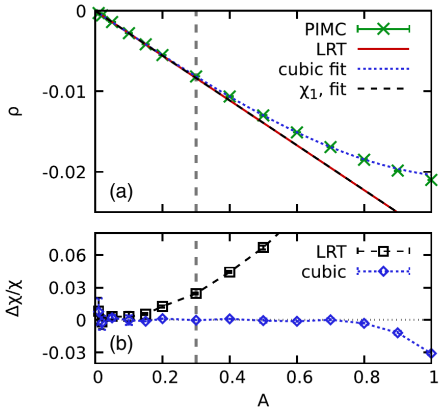
- Small perturbations: expansion of density resonse $\rho(q)$

$$\rho(q) = A\cdot\chi_{\text{LRT}}(q) + A^3\cdot\chi_3(q) + \dots$$

$\Rightarrow$ Unambiguous quantification of nonlinear effects



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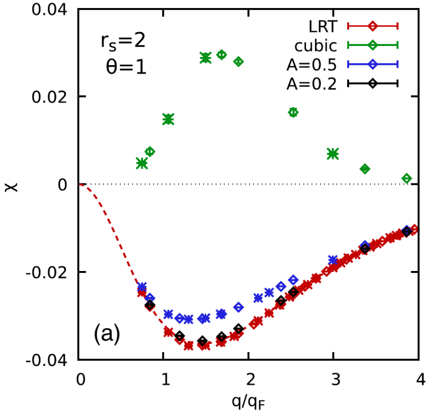
- Extensive *ab initio* PIMC results for **cubic response function** $\chi_3(q)$



Wave number dependence of linear and cubic density response function of the UEG at $r_s = 2$ and $\theta = 1$

Taken from: T. Dornheim, J. Vorberger, and M. Bonitz, *Phys. Rev. Lett.* **125**, 085001 (2020)

Nonlinear Electronic Density Response in WDM

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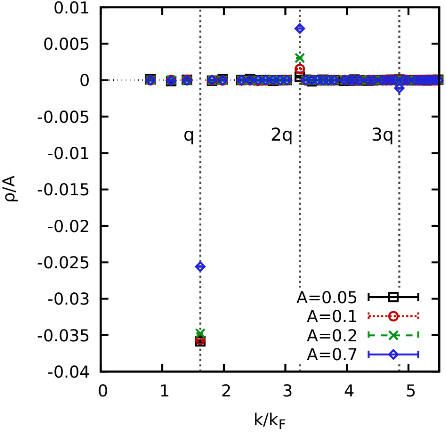
- **Work in progress [1]:** Excitation of harmonics of q
  - $\rightarrow$ Density response of harmonics in WDM open
  - $\rightarrow$ Evidence that second harmonic, $\rho(2q)$, constitutes dominant nonlinear effect
  - $\rightarrow$ Nontrivial generalized response functions are needed



Spectrum of the density response $\rho_q(k)$ of the UEG at $r_s = 2$ and $\theta = 1$ for $q = 1.69q_\mathrm{F}$

Taken from: T. Dornheim *et al.*, in preparation

[1] T. Dornheim, M. Böhme, J. Vorberger, Zh. Moldabekov, and M. Bonitz, in preparation

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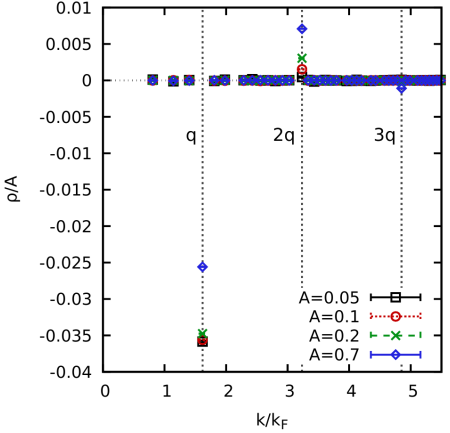
  - $\rightarrow$ Excitations of harmonics provide additional, nontrivial information about the sample, correlations



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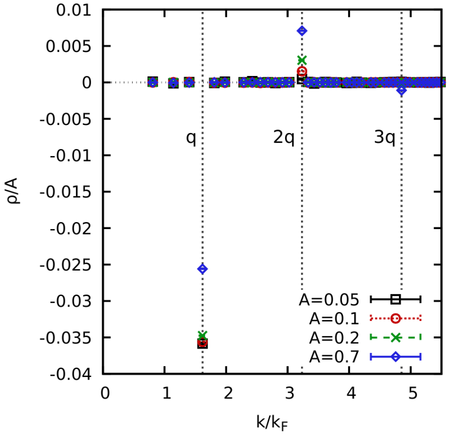
- **Investigation of nonlinear effects in...**
  - → Screened potentials
  - → Stopping power / energy loss
  - → . . .



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Summary[19]

- **WDM**: - crucial for astrophysics, materials properties, energy applications
  - remarkable progress in facilities and experimental diagnostics (XRTS)
  - but: complicated mix of phases, no small parameters
- **Our approach**: - highly accurate treatment of key component: warm dense equilibrium electrons
  - extension to WDM via hybrid schemes: DFT+MD, Mermin dielectric function etc.
  - Electrons treated via *ab initio* **QMC** simulations, combining CPIMC and PB-PIMC
- **Recent breakthroughs: benchmark data of unprecedented accuracy**

1. Thermodynamic functions for entire warm dense range[13]
2. accurate functional $f_{xc}(r_s, \Theta, \xi)$ input for finite-T LDA-DFT, implemented in Libxc (LDA_XC_GDSMFB)
3. *ab initio* data and machine learning representation static local field correction[14] $G(q)$
4. *ab initio* data for the dynamic structure factor $S(q, \omega)$ and XRTS signal [15];
5. *ab initio* data for the dynamic local field correction $G(\mathbf{q}, \omega)$, density response function, conductivity $\sigma(\mathbf{q}, \omega)$ [16]
6. first *ab initio* data for the dielectric function, plasmon dispersion $\omega(q)$, accurate parametrization[17]
7. Nonlinear density response[18] and harmonics generation

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[16]P. Hamann *et al.*, Phys. Rev. B (2020), arXiv:2007.15471
[17]P. Hamann *et al.*, Contrib. Plasma Phys. (2020), arXiv:2008.04605
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[19]http://www.theo-physik.uni-kiel.de/bonitz/index.html ⇒ Research ⇒ Publications, Talks