

# Perspectives of quantum plasma and warm dense matter theory<sup>1</sup>

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<sup>4</sup> IVTAN, Moscow

PLTP 2020, Kazan, November 2020

**DFG**

**DAAD**

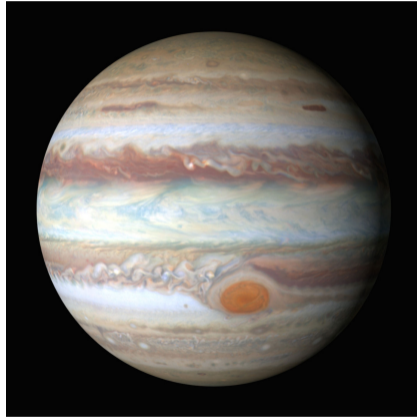


<sup>1</sup> <http://www.theo-physik.uni-kiel.de/bonitz/index.html> ⇒ Research ⇒ Publications, Talks

# Warm Dense Matter: Occurrences and Applications

## ▶ **Astrophysics:**

- ▶ Giant planet interiors (e.g. Jupiter)
- ▶ Brown dwarfs
- ▶ Meteor Impacts



[Source: Sci-News.com \[Img4\]](#)

# Warm Dense Matter: Occurrences and Applications

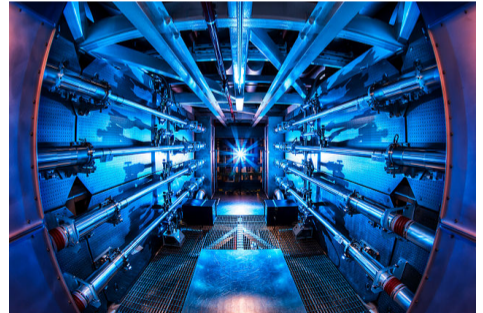
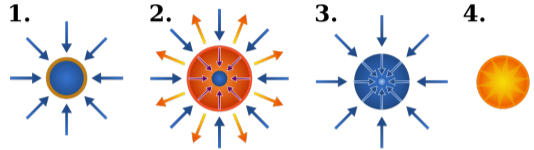
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## ▶ Experiments:

- ▶ Inertial confinement fusion

**Potential abundance of clean energy!**



Source: [en.wikipedia.org](https://en.wikipedia.org) [Img5] and [arstechnica.com](https://arstechnica.com) [Img6]

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**Potential abundance of clean energy!**

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

## National Ignition Facility (Livermore, California)



area:  $70000m^2$

cost:  $\sim 1$  billion Dollar

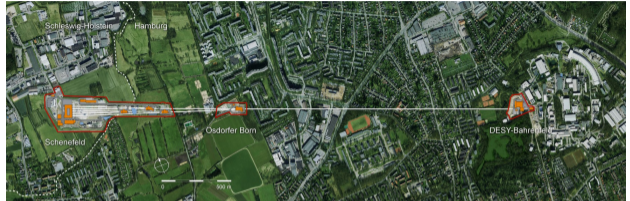
Source: C. Stolz, *Phil. Trans. R. Soc. A* **370**, 4115 (2012) [Img7]

# Facilities for WDM experiments in Europe:

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

## **European XFEL:**

- ▶ **European X-ray Free-Electron Laser**, Hamburg – Schenefeld
- ▶ Total cost  $\sim$  **1.2 billion Euro**
- ▶ **HIBEF Beamline and consortium** (DiPOLE laser contributed from UK)



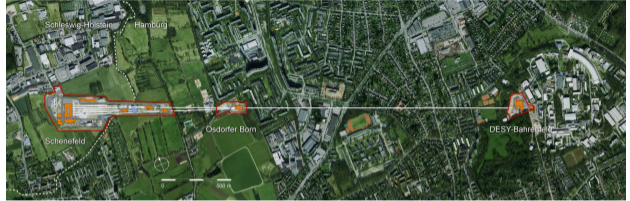
source: [photon-science.desy.de](http://photon-science.desy.de)

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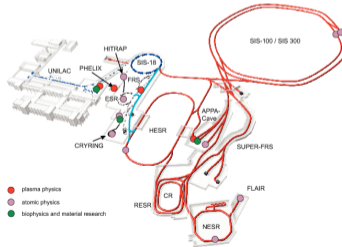
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## FAIR:

- ▶ Facility for Antiproton and Ion Research, Darmstadt
- ▶ Construction started in 2017
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- ▶ Heavy ion beams: Isochoric heating up to  $\sim 10^6 K$



source: inspirehep.net



source: dw.com

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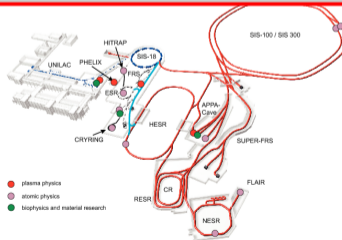
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Warm dense matter: indeed a **HOT** topic

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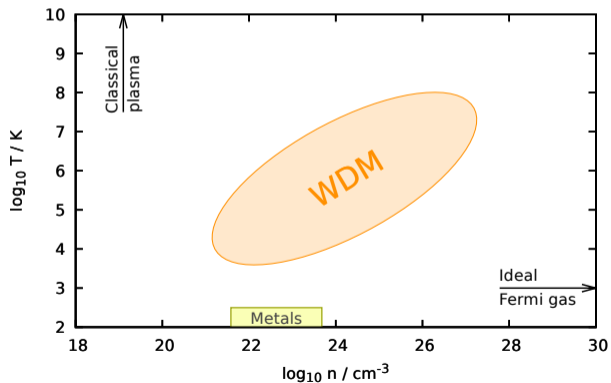
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## Warm Dense Matter and quantum plasmas: relevant parameters

### ► Extreme and exotic state of matter:

- High temperature:  $T \sim 10^3 - 10^8 \text{ K}$
- Extreme density:  $n \sim 10^{21} - 10^{27} \text{ cm}^{-3}$

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





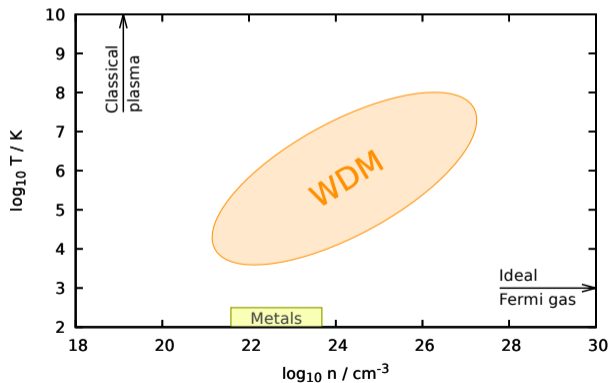
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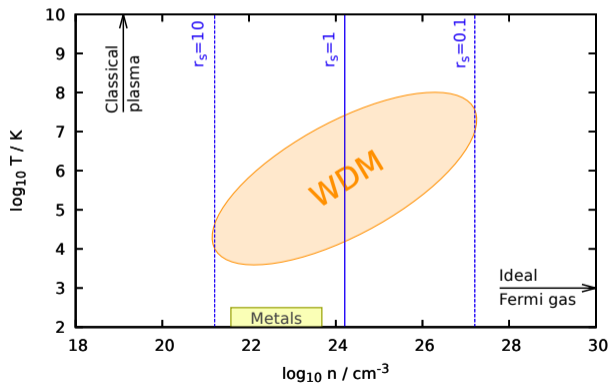
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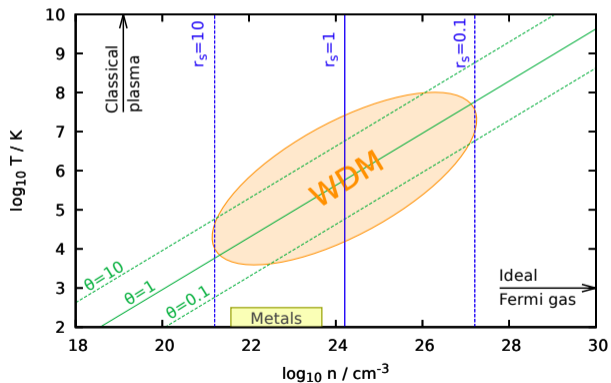
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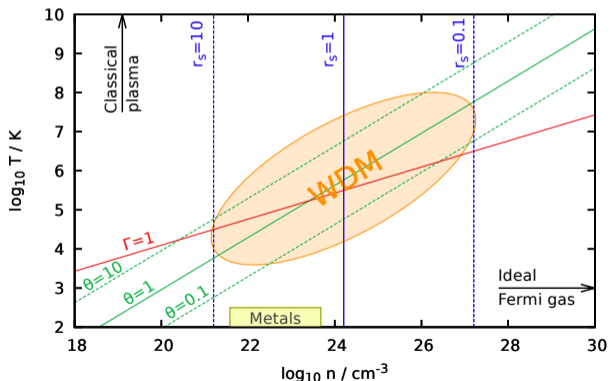
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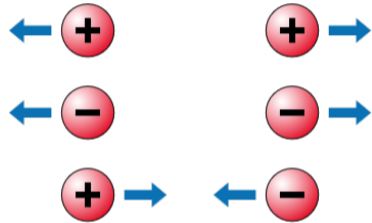
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## ▶ Nontrivial interplay of many effects:

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[Source: bin-br.at](http://bin-br.at) [Img1]

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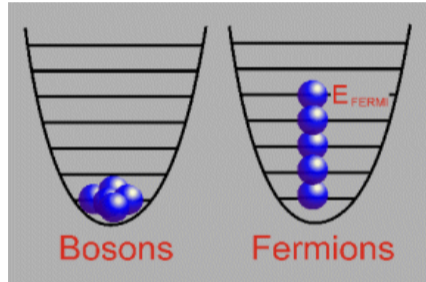
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- ▶ Coulomb coupling (non-ideality)
- ▶ Fermionic exchange (anti-symmetry)



Source: [cidehom.com](http://cidehom.com) [Img2]

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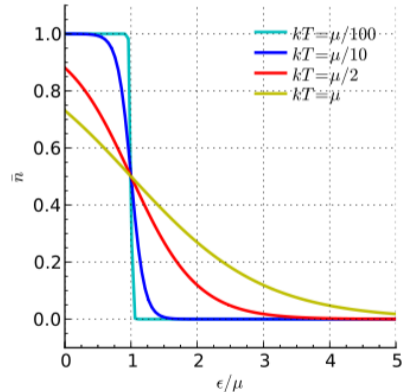
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## ► Nontrivial interplay of many effects:

- Coulomb coupling (non-ideality)
- Fermionic exchange (anti-symmetry)
- Thermal excitations (statistical description)



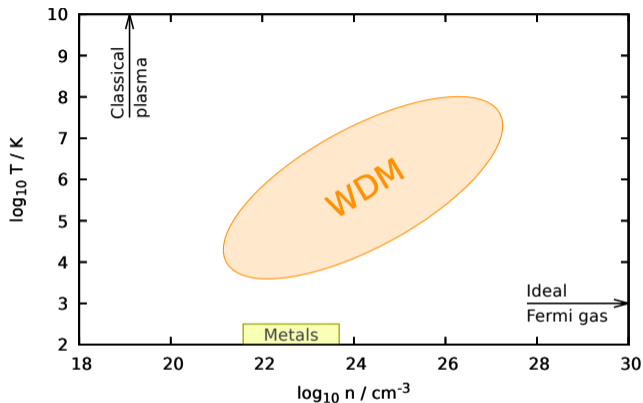
## Introduction: how to theoretically approach warm dense matter?

**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)

⇒ 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:**

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

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

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<sup>1</sup> J.P. Perdew and A. Zunger, PRB **23**, 5048 (1981)    <sup>2</sup> D.M. Ceperley and B. Alder, PRL **45**, 566 (1980)    <sup>3</sup> N.D. Mermin, Phys. Rev **137**, A1441 (1965)

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## Warm dense matter ( $T \sim T_F$ ):

- ▶ **Thermal DFT<sup>3</sup>:** minimize free energy  $F = E - TS$
- **Requires parametrization of XC free energy of UEG:**

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

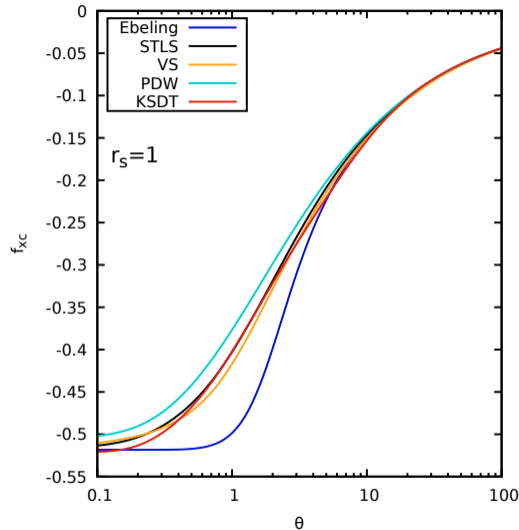
- ▶  $f_{xc}(r_s, \theta)$  direct input for **EOS models** of astrophysical objects<sup>4</sup>
- ▶  $f_{xc}(r_s, \theta)$  contains **complete thermodynamic information** of UEG

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## Many parametrizations for $f_{xc}$ based on different approximate approaches:

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



<sup>1</sup> W. Ebeling and H. Lehmann, Ann. Phys. **45**, (1988)

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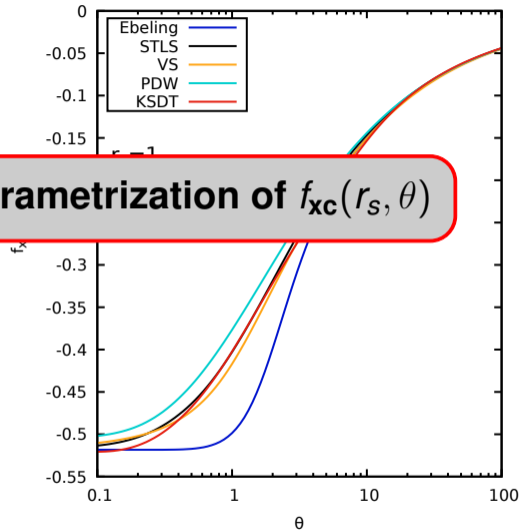
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- ▶ Semi-analytic
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- ▶ Quantum-classical mappings, e.g. Perrot and Dharma-wardana<sup>4</sup> (PDW)
- ▶ **Most recent:** Fit by Karasiev<sup>5</sup> *et al.* (KSDT) to Restricted Path Integral Monte Carlo (RPIMC) data<sup>6</sup>

**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  $Z$

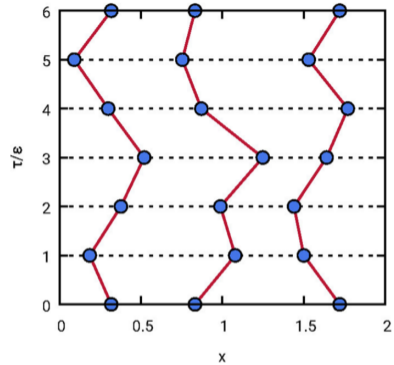
$$\hat{H} = \hat{K} + \hat{V}, \quad [\hat{K}, \hat{V}] \neq 0, \quad \beta = 1/k_B T,$$
$$Z = (\text{Tr } \hat{\rho})^\pm, \quad \hat{\rho} = e^{-\beta \hat{H}} = [e^{-\hat{H}/Pk_B T}]^P,$$

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

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

- ▶ 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  $\mathbf{X} = \{\mathbf{R}_0, \dots, \mathbf{R}_{P-1}\}$  in “imaginary time”, with  $P$  “time slices”

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



PIMC configuration of  $N = 3$  particles

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

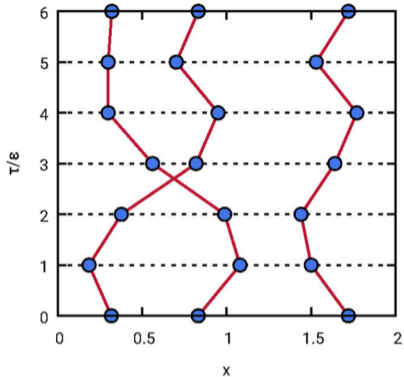
Based on R. Feynman's path integral quantum mechanics  
**PIMC: parameter-free, potentially exact method!**

# Path Integral Monte Carlo (PIMC): Fermions

## ► Fermionic antisymmetry:

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

⇒ We must include **permutation-cycles!**



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

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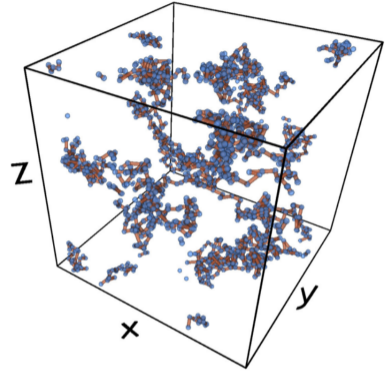
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► Randomly generate all possible paths **X** using the **Metropolis algorithm**



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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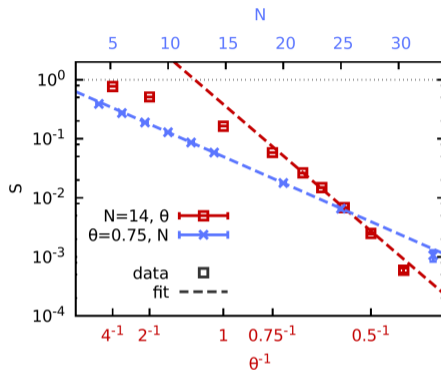
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## ▶ Randomly generate all possible paths $\mathbf{X}$ using the **Metropolis algorithm**

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

⇒ Fermion Sign Problem



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

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



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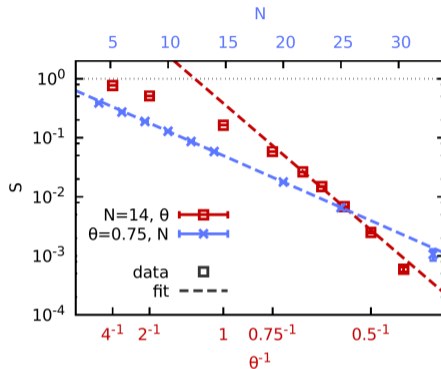
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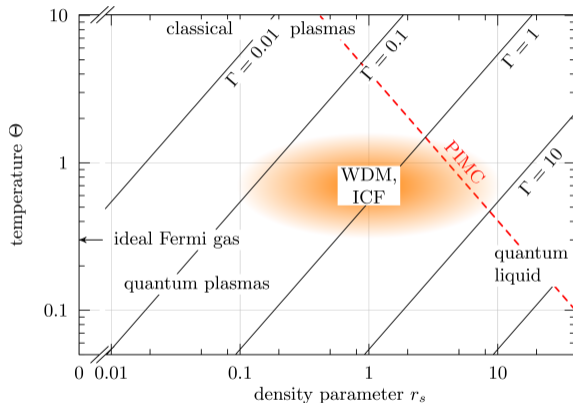
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**PIMC simulations of WDM very challenging!**

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

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



<sup>1</sup> E.W. Brown *et al.*, PRL **110**, 146405 (2013)

<sup>3</sup> T. Schoof *et al.*, Phys. Rev. Lett **115**, 130402 (2015)

<sup>4</sup> T. Schoof *et al.*, Contrib. Plasma Phys. **51**, 687 (2011)

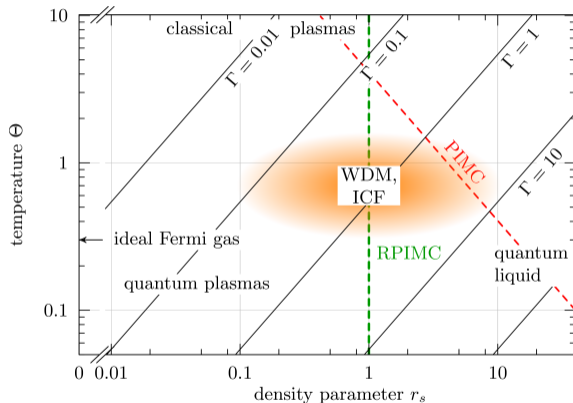
<sup>2</sup> V. Filinov *et al.*, Phys. Rev. E **91**, 033108 (2015)

<sup>5</sup> T. Dornheim *et al.*, New J. Phys. **17**, 073017 (2015)

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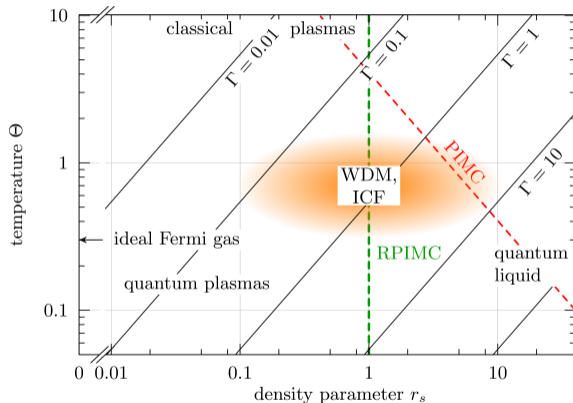
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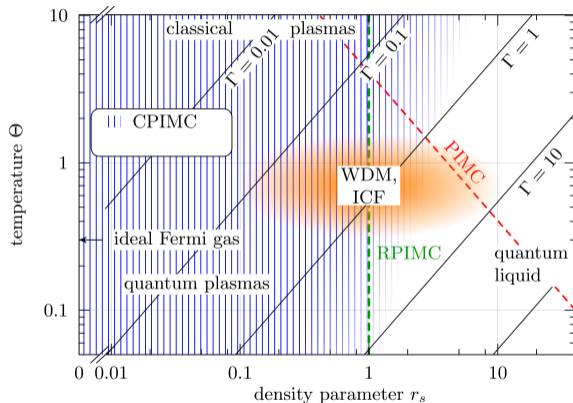
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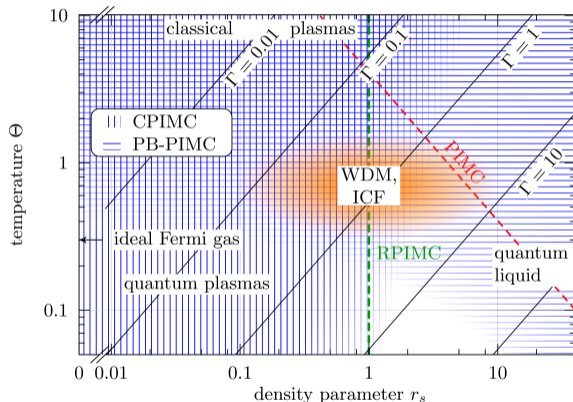
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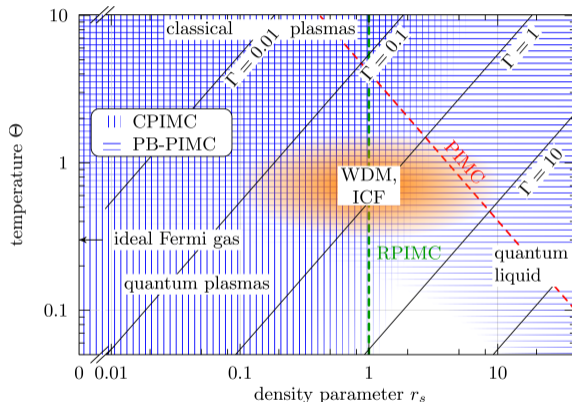
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**Ab initio simulations over broad range of parameters possible**

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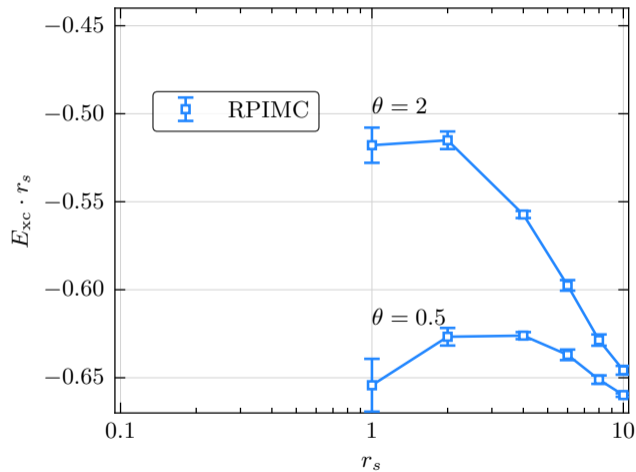
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1. Exact exchange-correlation energy  $E_{xc} = E - E_0$  ( $E_0$ : ideal energy)  
( $N = 33$  spin-polarized electrons,  $\theta \geq 0.5, \forall r_s$ )

► **RPIMC** limited to  $r_s \geq 1$



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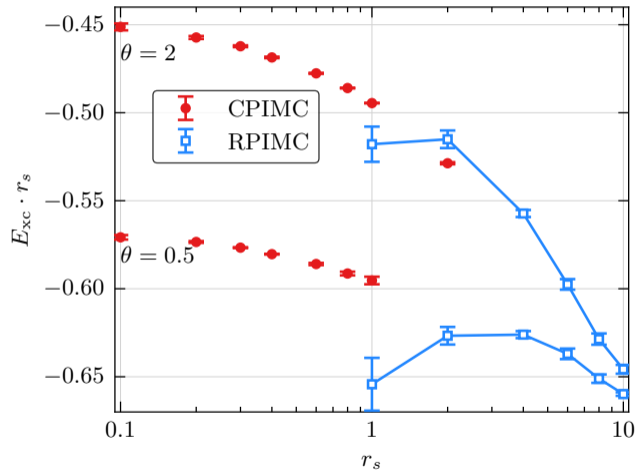
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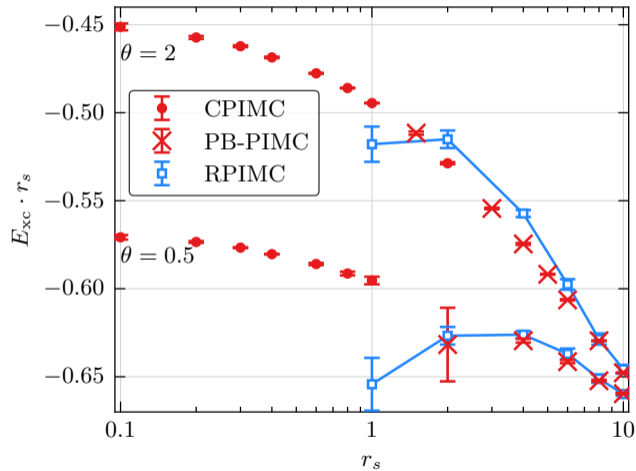
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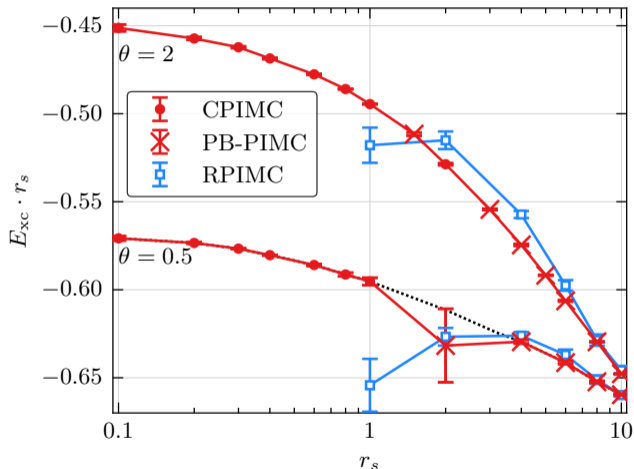
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**Combination<sup>1</sup> yields exact results over entire density range down to  $\theta \sim 0.5$**

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



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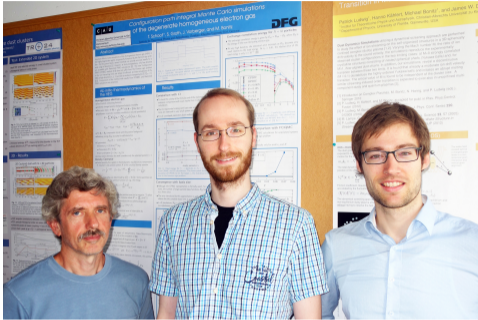
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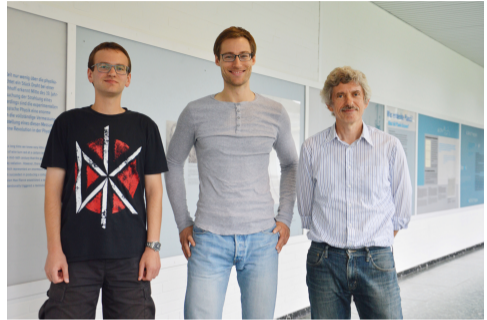
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## Acknowledgements to those who did most of the work...



**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:  $\delta \hat{H}(\mathbf{q}) \rightarrow \delta \rho(\mathbf{q})$

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

### **Susceptibilities from linear response theory (LRT):**

$\delta \rho(\mathbf{q}) = \chi(\mathbf{q}) \delta H(\mathbf{q})$ ,  $\chi$ : static density response  $\rightarrow$  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.:  $\chi(\mathbf{q}, \omega)$ ,  $S(\mathbf{q}, \omega)$ ,  $\epsilon(\mathbf{q}, \omega)$ ,  $\sigma(\mathbf{q}, \omega)$

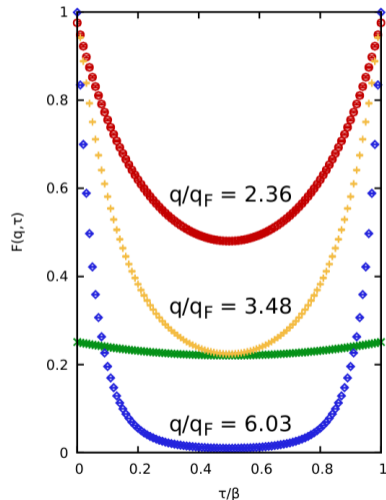
**PIMC: susceptibilities beyond validity limits of LRT**

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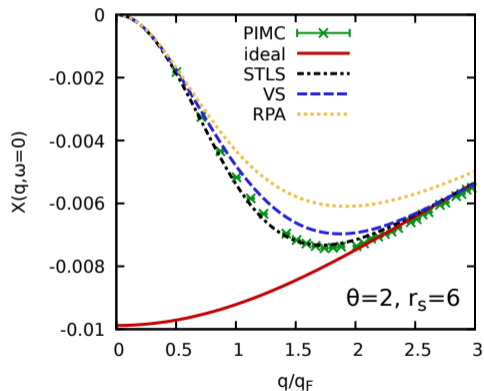
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$$\chi(\mathbf{q}) = -n \int_0^\beta d\tau F(\mathbf{q}, \tau)$$

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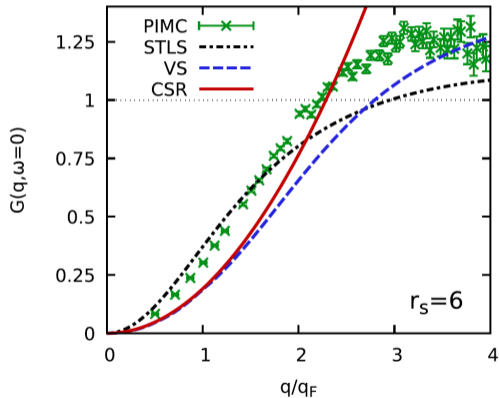
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- ▶  $G(q)$  can be obtained as the deviation from  $\chi_0(q)$ :

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

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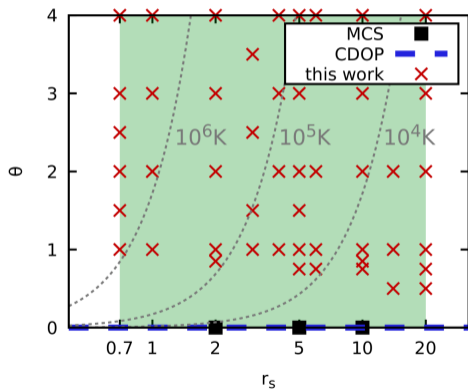


# The Static Local Field Correction: Neural-net representation

## Extensive set of new PIMC data

- ▶ QMC data available at discrete grid  $(q; \theta, r_s)$

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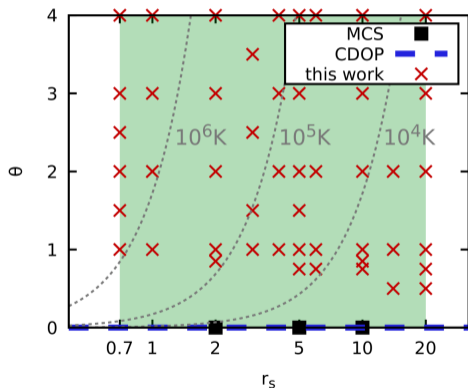


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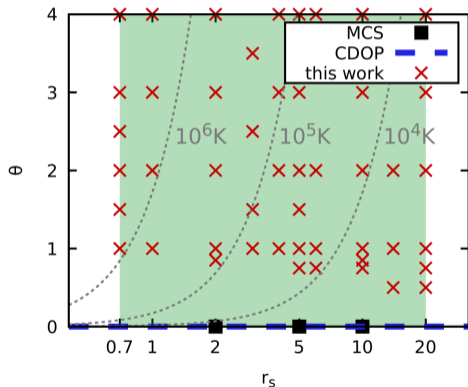


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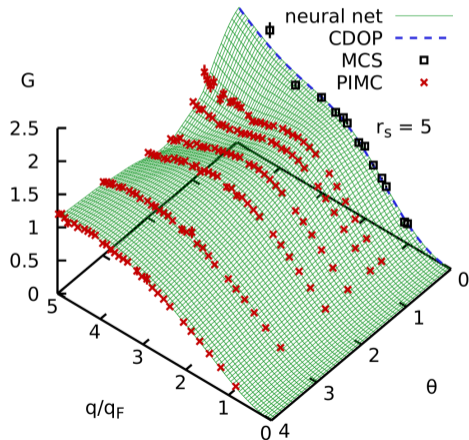


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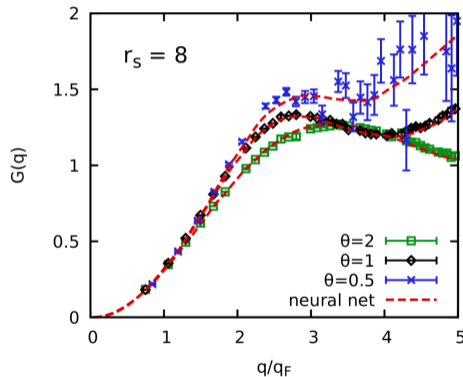


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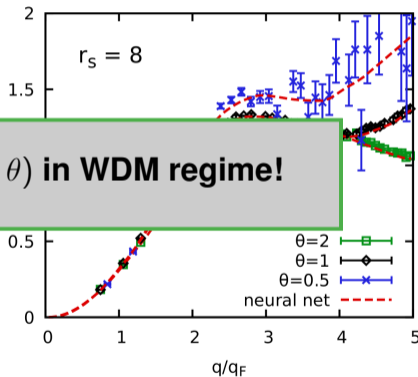
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- ▶ Computational analytical
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**2. Complete *ab initio* data for  $G(q; r_s, \theta)$  in WDM regime!**

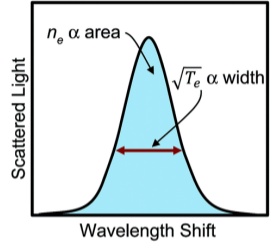
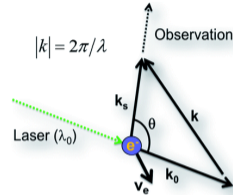


### 3. *Ab initio* dynamic ( $\omega$ -dependent) results for the warm dense UEG

► **Key quantity:** dynamic structure factor

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→ directly measured in **scattering experiments**



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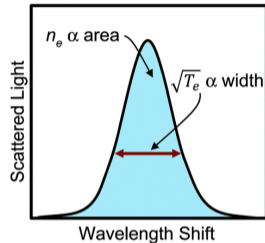
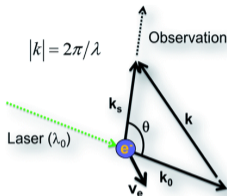
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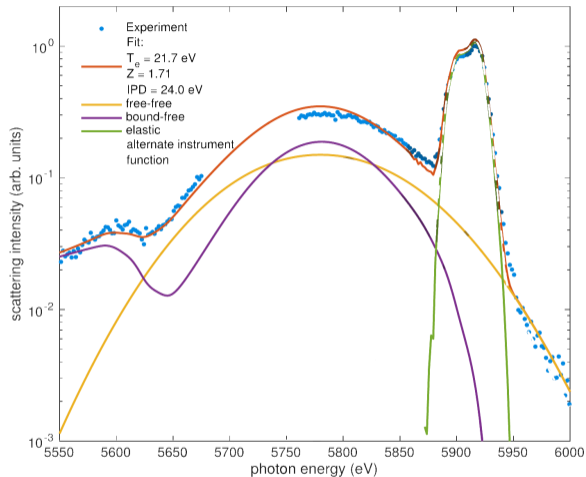
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- ▶ **Practical example:** Fit model for  $S(\mathbf{q}, \omega; T_e)$  to spectrum to determine electron temperature  $T_e$



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

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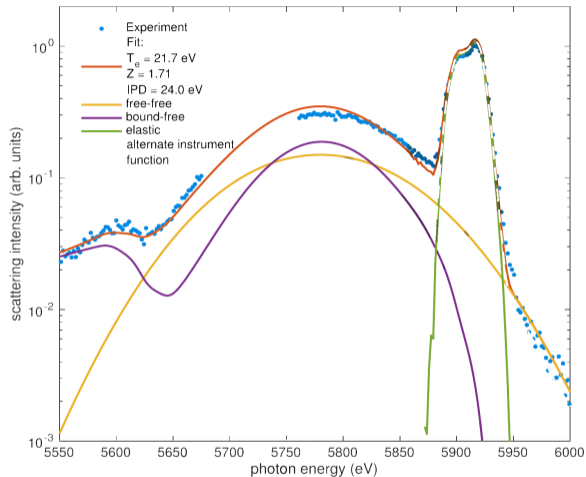
$$\rightarrow S_{f-f}(\mathbf{q}, \omega) \sim S^{\text{UEG}}(\mathbf{q}, \omega)$$

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

- ▶ **Problem:**

$F(\mathbf{q}, t)$  requires **real time-dependent simulations**

→ with PIMC have to use analytic continuation, reconstruct  $F(q, it)$  and 4 frequency moments, but: insufficient information



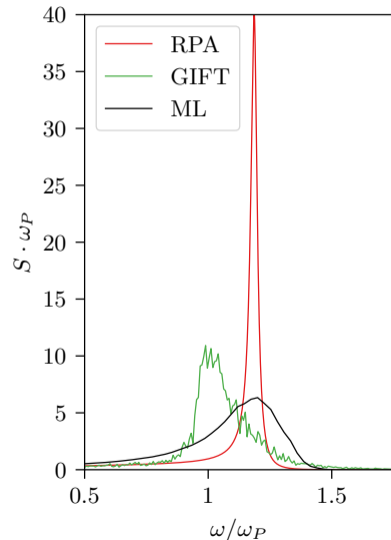
Scattering spectrum of isochorically heated graphite at LCLS.  
From D. Kraus *et al.*, *Plasma Phys. Control. Fusion* (2019)

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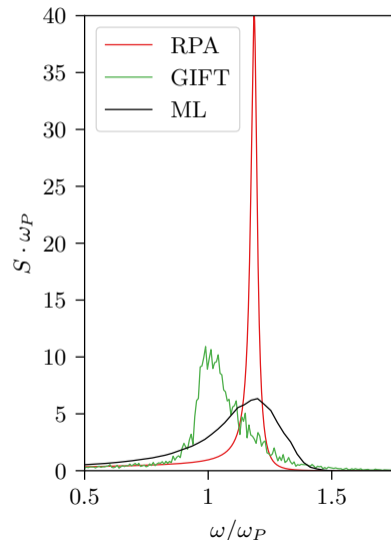
$$S(\mathbf{q}, \omega) = -\frac{\text{Im}\chi(\mathbf{q}, \omega)}{\pi n(1 - e^{-\beta\omega})}$$

- ▶ Express response function  $\chi$  via ideal response function  $\chi_0$  and **dynamic local field correction  $G$** :

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

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

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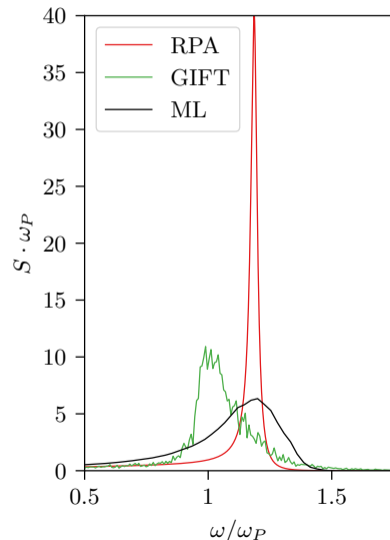
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**Make ansatz and optimize  $G(\mathbf{q}, \omega)$  instead of  $S(\mathbf{q}, \omega)$**

### 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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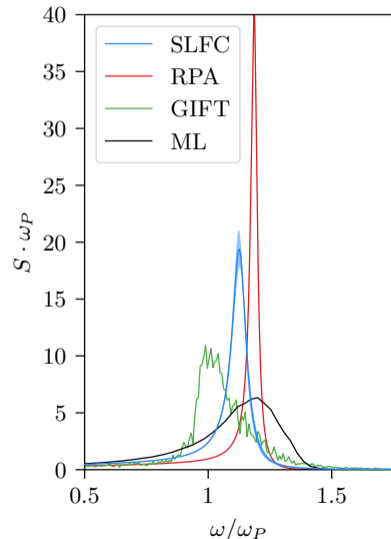
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▶ Ex  
dy

**GIFT and ML spectra not in agreement  
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→ to be discarded as unphysical**

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

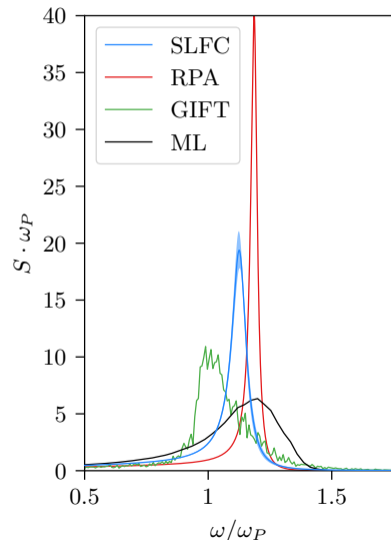
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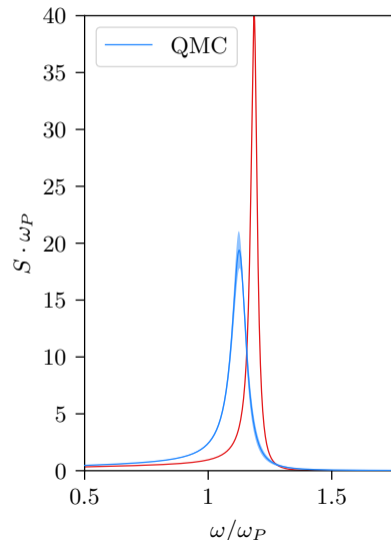
- ▶ **Random phase approximation (RPA):  $G \equiv 0$**

**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
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( $\theta = 1, r_s = 10, N = 33, q = 0.63q_F$ )



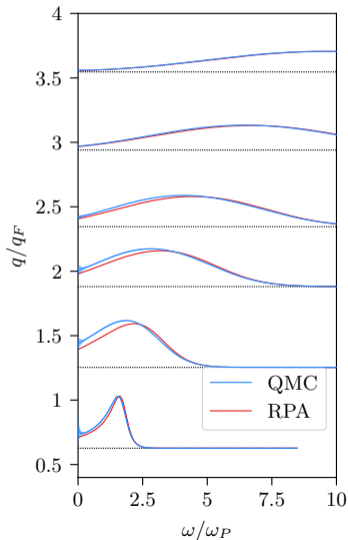


# Correlation effects in the peak position of $S(q, \omega)$ : $\theta = 1$ , $r_s = 2$

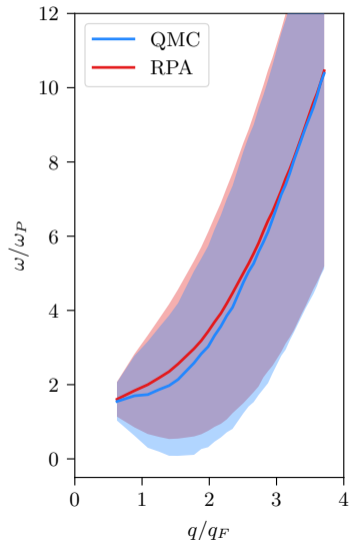
T. Dornheim, S. Groth, J. Vorberger, and M. Bonitz, *Phys. Rev. Lett.* **121**, 255001 (2018)

- ▶ *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:

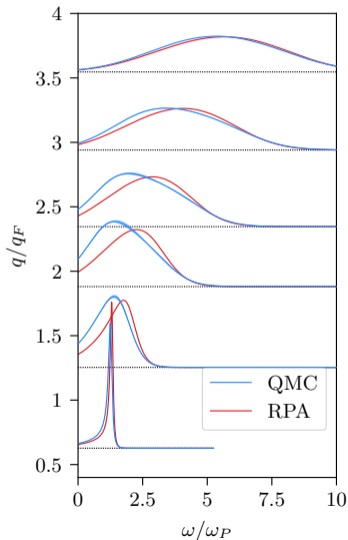


# Correlation effects in the peak position of $S(q, \omega)$ : $\theta = 1$ , $r_s = 6$

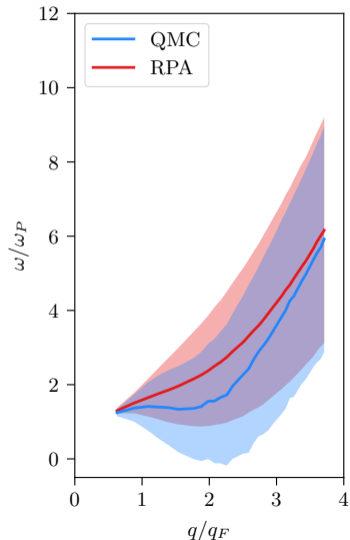
T. Dornheim, S. Groth, J. Vorberger, and M. Bonitz, *Phys. Rev. Lett.* **121**, 255001 (2018)

- ▶ *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  $r_s$

Dynamic structure factor of the UEG:



Peak position and FWHM:



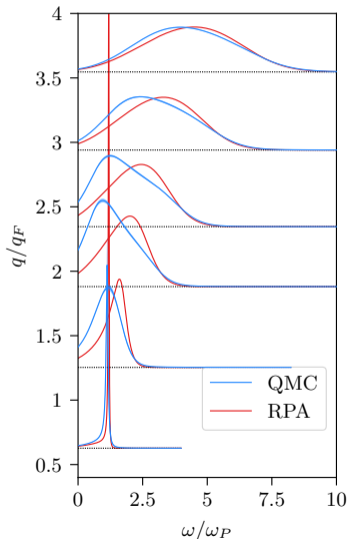
# Correlation effects in the peak position of $S(\mathbf{q}, \omega)$ : $\theta = 1$ , $r_s = 10$

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

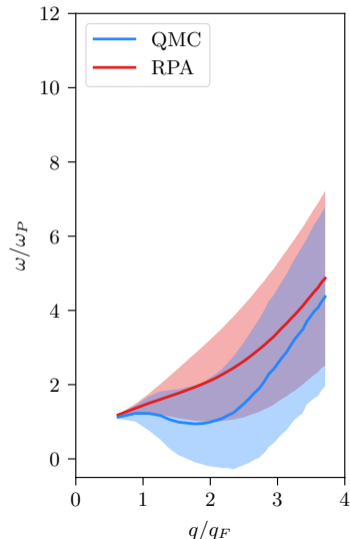
- ▶ *Ab initio* results for  $G(q, 0)$  available: Dornheim *et al.*, J. Chem. Phys. (2019)
- ▶ Slight **correlation induced redshift** of peak for intermediate  $q$  (at small  $r_s$ )
- ▶ **Pronounced redshift and broadening** with increasing  $r_s$
- ▶ **Negative dispersion of peak** for large  $r_s$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

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

Dynamic structure factor of the UEG:



Peak position and FWHM:



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

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

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

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

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

- ▶ **Exact results**<sup>4</sup> :  $G^{\text{QMC}}(q, \omega) \rightarrow \Pi^{\text{QMC}}(q, \omega) \rightarrow \epsilon^{\text{QMC}}(q, \omega)$   
**Accurate and efficient approximation**:  $G^{\text{QMC}}(q, \omega) \rightarrow G^{\text{QMC}}(q, 0) = G(q)$ , insert in  $\Pi(q, \omega) \rightarrow \epsilon^{\text{SLFC}}(q, \omega; T)$
- ▶ QHD with exchange-correlation corrections<sup>5</sup>, but only:  $T = 0$  and low accuracy xc effects (LDA)
- ▶ Improved QHD<sup>6</sup>: finite  $T$ ,  $\omega$ - and  $q$ -dependent coefficients, correlations via  $G$  and non-local effects

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<sup>2</sup>G. Manfredi and F. Haas, Phys. Rev. B (2001)

<sup>3</sup>M. Bonitz *et al.*, Phys. Plasmas (2019)

<sup>4</sup>P. Hamann *et al.*, Phys. Rev. B (2020), arXiv: 2007.15471

<sup>5</sup>N. Crouseilles *et al.*, Phys. Rev. B (2008)

<sup>6</sup>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<sup>7</sup>:  $\frac{\omega^2(q)}{\omega_p^2} = 1 + \frac{v_{th}^2}{\omega_p^2} q^2$ ,  $v_{th}^2 = \frac{3k_B T}{m}$
- ▶ Bohm and Pines 1953, quantum plasma,  $T = 0$  (RPA)<sup>8</sup>:  $\frac{\omega^2(q)}{\omega_p^2} = 1 + \frac{3}{5} \frac{v_F^2}{\omega_p^2} q^2 + \frac{\hbar^2}{4m^2} \frac{q^4}{\omega_p^2}$ ,
- ▶ Ferrell 1957,  $q^4$  terms,  $T = 0$ <sup>9</sup>:  $\frac{\omega^2(q)}{\omega_p^2} = 1 + \frac{3}{5} \frac{v_F^2}{\omega_p^2} q^2 + \left( \frac{(\Delta v_0^2)^2}{\omega_p^2} + \frac{\hbar^2}{4m^2} \right) \frac{q^4}{\omega_p^2}$ ,  $(\Delta v_0^2)^2 = \langle v^4 \rangle_0 - \langle v^2 \rangle_0^2$
- ▶ Quantum hydrodynamics ( $T = 0$ )<sup>10</sup>:  $\frac{\omega^2(q)}{\omega_p^2} = 1 + \frac{1}{3} \frac{v_F^2}{\omega_p^2} q^2 + \frac{\hbar^2}{4m^2} \frac{q^4}{\omega_p^2}$ ,
- ▶ Hamann *et al.*<sup>11</sup> RPA, finite  $T$  :  $\frac{\omega^2(q)}{\omega_p^2} = 1 + \frac{\langle v^2 \rangle}{\omega_p^2} q^2 + \left( \frac{(\Delta v^2)^2}{\omega_p^2} + \frac{\hbar^2}{4m^2} \right) \frac{q^4}{\omega_p^2}$ ,  $\langle \dots \rangle$  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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<sup>7</sup>D. Bohm and E.P. Gross, Phys. Rev. (1949)

<sup>8</sup>D. Bohm and D. Pines, Phys. Rev. (1953), also: Lindhard, Klimontovich, Silin

<sup>9</sup>R.A. Ferrell, Phys. Rev. (1957)

<sup>10</sup>G. Manfredi and F. Haas, Phys. Rev. B (2001)

<sup>11</sup>P. Hamann et al., Contrib. Plasma Phys. (2020), arXiv: 2008.04605

## 5. Collective excitations. Plasmons

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

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

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

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

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

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

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

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

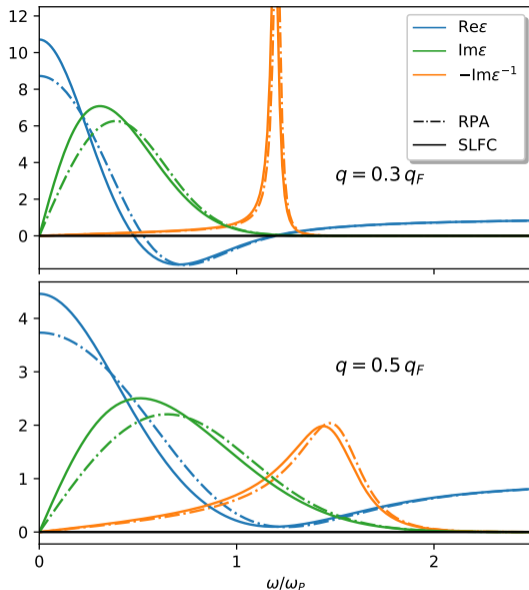
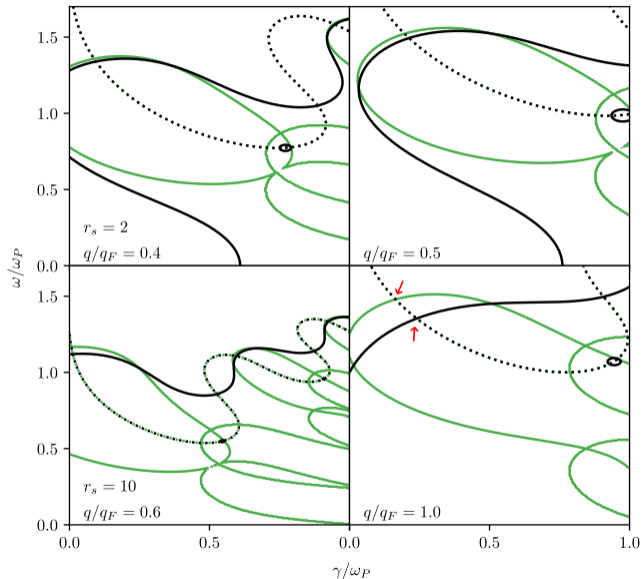


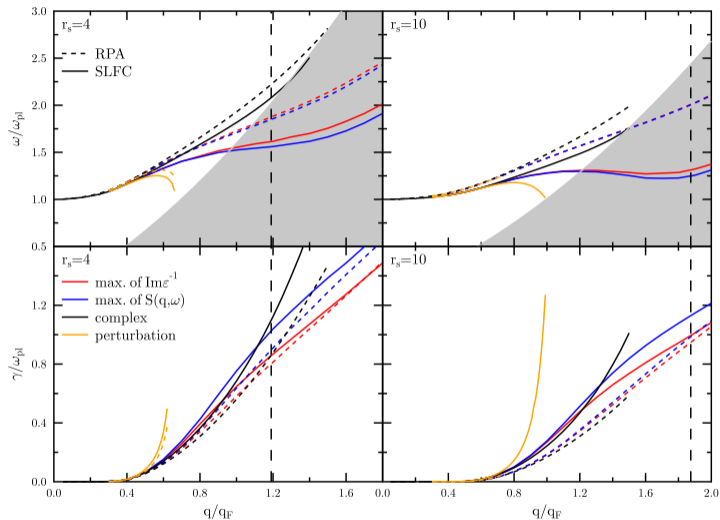
Figure: Moderately correlated electron gas,  $\Theta = 1$ ,  $r_s = 2$

## Analytic continuation (AC) of the dielectric function<sup>12</sup>

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



# 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:  $2\pi/q = \lambda = \lambda_{scr}$ .  
 From P. Hamann *et al.*, Contrib. Plasma Phys. (2020), arXiv:2008.04605



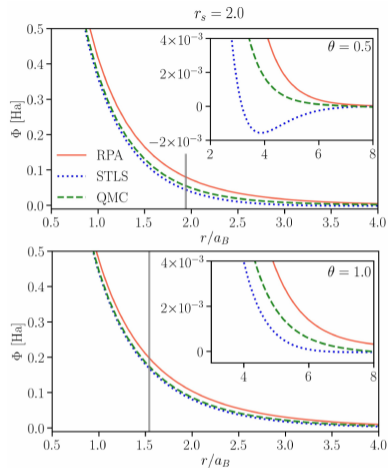
## 6. Nonlinear Electronic Density Response in WDM

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*.

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

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

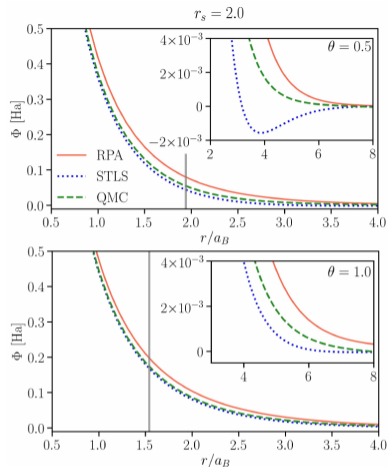
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- ▶ **Linear Response Theory (LRT)** implicitly assumed throughout WDM theory, including:
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  - ▶ Construction of effective potentials
  - ▶ Calculation of stopping power and conductivities
  - ▶ XC-functionals for DFT
- ▶ **Open question:** Cases of strong excitation:
  - ▶ Seeded FELs<sup>[1]</sup>:  $I \sim 10^{22} \frac{W}{cm^2}$
  - ▶ THz lasers (high ponderomotive potential)

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

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



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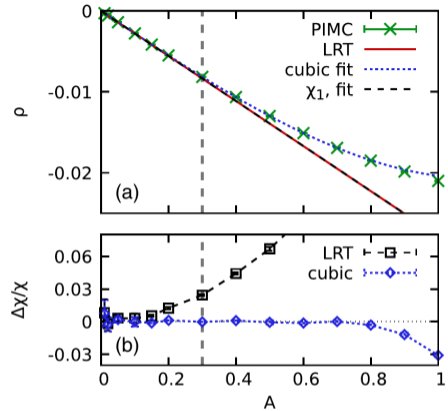
# Nonlinear Electronic Density Response in WDM

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

- **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  $q = 0.84q_F$

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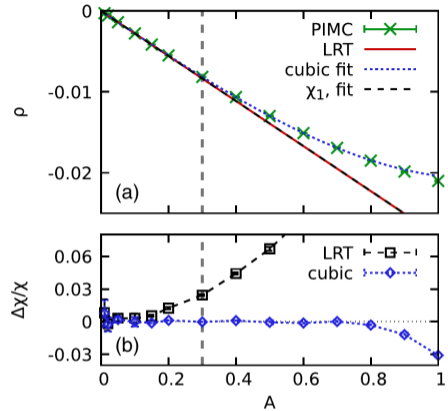
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$$\rho(q) = A \cdot \chi_{\text{LRT}}(q) + A^3 \cdot \chi_3(q) + \dots$$

⇒ Unambiguous quantification of nonlinear effects



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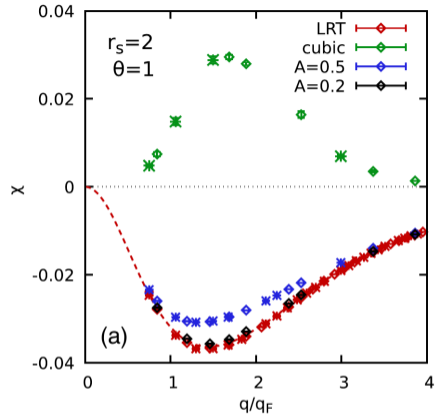
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- ▶ Extensive *ab initio* PIMC results for **cubic response function**  $\chi_3(q)$



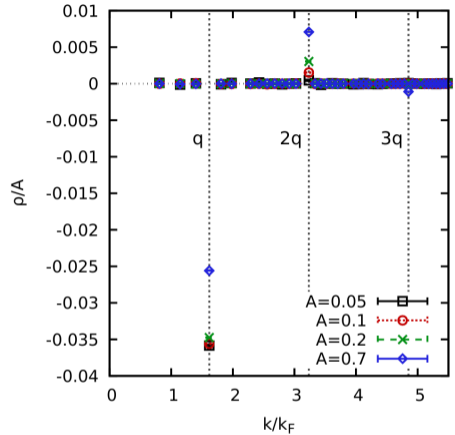
Wave number dependence of linear and cubic density response function of the UEG at  $r_s = 2$  and  $\theta = 1$

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

# Nonlinear Electronic Density Response in WDM

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

- **Work in progress [1]:** Excitation of harmonics of  $q$ 
  - Density response of harmonics in WDM open
  - Evidence that second harmonic,  $\rho(2q)$ , constitutes dominant nonlinear effect
  - 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  $q = 1.69q_F$

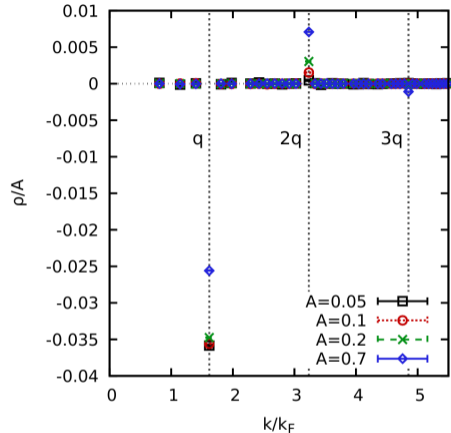
[1] T. Dornheim, M. Böhme, J. Vorberger, Zh. Moldabekov, and M. Bonitz, in preparation

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

# Nonlinear Electronic Density Response in WDM

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

- ▶ **Work in progress [1]:** Excitation of harmonics of  $q$ 
  - Density response of harmonics in WDM open
  - Evidence that second harmonic,  $\rho(2q)$ , constitutes dominant nonlinear effect
  - Nontrivial generalized response functions are needed
- ▶ **Idea: Nonlinear diagnostics of WDM**
  - Excitations of harmonics provide additional, nontrivial information about the sample, correlations



Spectrum of the density response  $\rho_q(k)$  of the UEG at  $r_s = 2$  and  $\theta = 1$  for  $q = 1.69q_F$

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

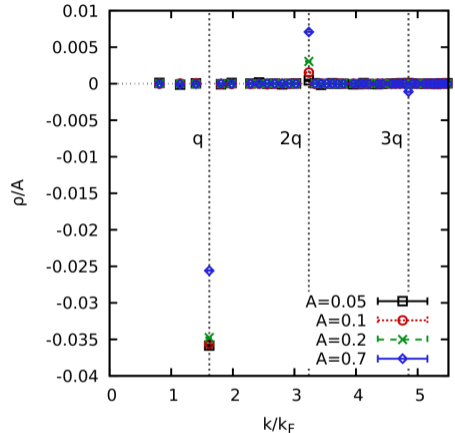
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- ▶ **Investigation of nonlinear effects in...**
  - Screened potentials
  - Stopping power / energy loss
  - ...

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- ▶ **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

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- ▶ **Recent breakthroughs: benchmark data of unprecedented accuracy**
  1. Thermodynamic functions for entire warm dense range<sup>14</sup>
  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<sup>15</sup>  $G(q)$
  4. *ab initio* data for the dynamic structure factor  $S(q, \omega)$  and XRTS signal<sup>16</sup>;
  5. *ab initio* data for the dynamic local field correction  $G(\mathbf{q}, \omega)$ , density response function, conductivity  $\sigma(\mathbf{q}, \omega)$ <sup>17</sup>
  6. first *ab initio* data for the dielectric function, plasmon dispersion  $\omega(q)$ , accurate parametrization<sup>18</sup>
  7. Nonlinear density response<sup>19</sup> and harmonics generation

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<sup>14</sup>T. Dornheim *et al.*, Phys. Rev. Lett. (2016), Phys. Reports (2018), Groth *et al.*, Phys. Rev. Lett. (2017)

<sup>15</sup>T. Dornheim *et al.*, J. Chem. Phys. (2019)

<sup>16</sup>T. Dornheim *et al.*, Phys. Rev. Lett. (2018)

<sup>17</sup>P. Hamann *et al.*, Phys. Rev. B (2020), arXiv:2007.15471

<sup>18</sup>P. Hamann *et al.*, Contrib. Plasma Phys. (2020), arXiv:2008.04605

<sup>19</sup>T. Dornheim *et al.*, Phys. Rev. Lett. (2020)

<sup>20</sup><http://www.theo-physik.uni-kiel.de/bonitz/index.html> ⇒ Research ⇒ Publications, Talks