Recent Progress in simulations of dense quantum plasmas and warm dense matter

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DPG Meeting, Mainz, March 30 2022

Symposium *Plasmas in the Universe*

pdf at www.theo-physik.uni-kiel.de/bonitz/research.html

Plasmas in the Universe: equilibrium phase diagram

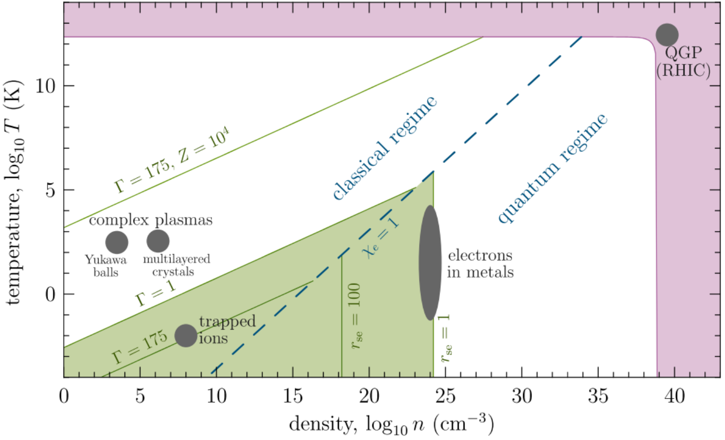
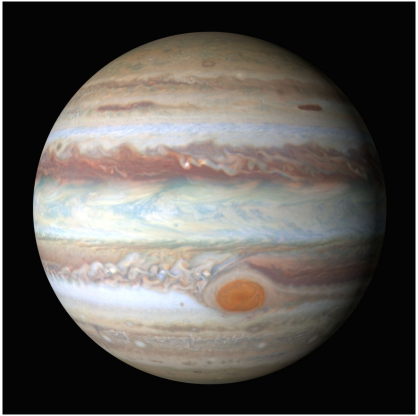


Figure 1. Examples of strongly correlated systems in thermodynamic equilibrium include complex plasmas, trapped ions and the QGP extending along the outer (pink) area, dot shows the conditions at RHIC). Prominent properties of all systems can be quantified by a few dimensionless parameters: the coupling parameter Γ, equation (2), the degeneracy parameter χ, equation (3), and the Brueckner parameter r_s, equation (4).

V. Filinov et al., *Color path-integral Monte-Carlo simulations of quark-gluon plasma: Thermodynamic and transport properties*, Phys. Rev. C **87**, 035207 (2013). Similar physics and methods.

Warm Dense Matter: Occurences and Applications [Andrew NG (2000): “missing link between CM, plasmas”]

- **Astrophysics:**
  - Giant planet interiors (e.g. Jupiter)
  - Brown dwarfs
  - Earth interior, Meteor Impacts
  - Recently discovered planets



Source: Sci-News.com [Img4]

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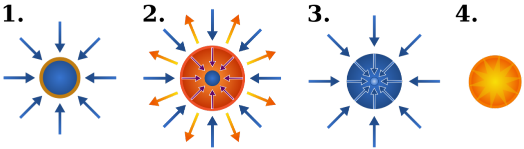
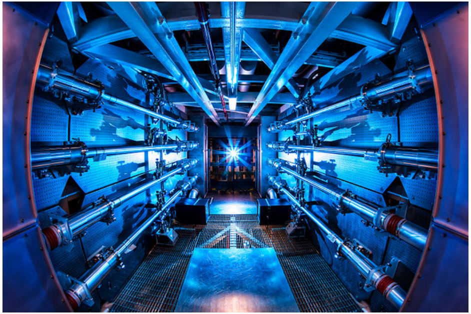
- **Laboratory Experiments, shock compression:**
  - Lasers, FELs, Z-pinch, ion beams
  - Properties of matter under extreme conditions
  - major driver: Inertial confinement fusion

  Potential abundance of clean energy!





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

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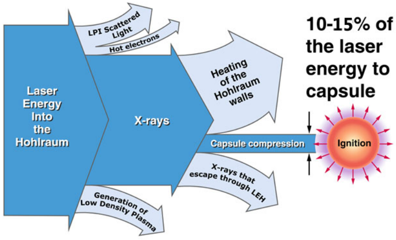
US: NIF, Omega (Rochester), LCLS (Stanford): Fundamental research into WDM properties: $\rightarrow$ Equation of state, $S(\mathbf{q}, \omega)$, conductivity etc.

National Ignition Facility (Livermore, California)

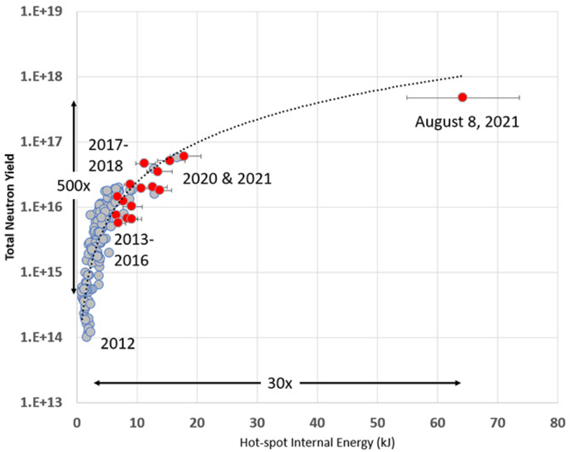
area: $70000 m^2$
cost: $\sim$1 billion Dollar
Source: C. Stolz, *Phil. Trans. R. Soc. A* **370**, 4115 (2012) [Img7]

Progress in Inertial Confinement Fusion



continuous optimization of target design, pulse shape etc.

Record shot on August 8 2021: 1.92 MJ UV laser energy
source: https://lasers.llnl.gov/news/hybrid-experiments-drive-nif-toward-ignition

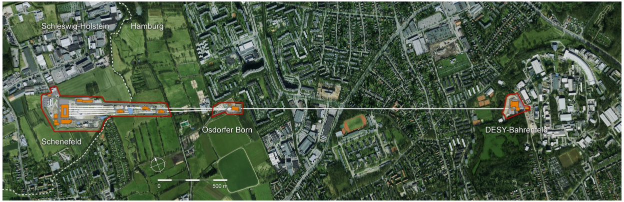


The “Hybrid-E experiment” on Aug. 8 achieved a hot-spot absorbed energy of about 65 kJ—about 20 kJ from the implosion, and the rest from “self-heating” from the fusion reactions (self-sustained burn). 1.35 MJ fusion energy yield, corresponds to 70% of ignition threshold (NAS criterion).

Facilities for WDM experiments in Europe and Asia:

Free electron lasers:

- **FLASH** (DESY, Hamburg)
- **E**uropean **X**-ray **F**ree-**E**lectron **L**aser, Hamburg – Schenefeld
- HIBEF Beamline and consortium. 2021 first successful experiments
- **Fermi** (Triest, Italy)
- **SACLA** (Riken, Japan)



source: photon-science.desy.de

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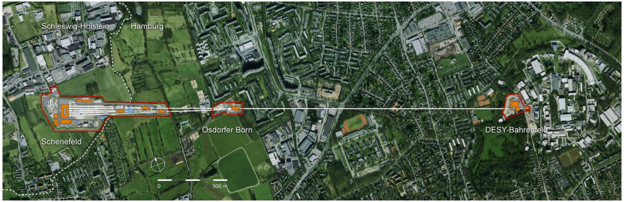

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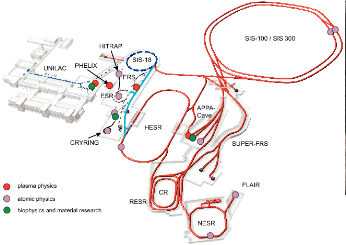
Heavy ion beams:

- **F**acility for **A**ntiproton and **I**on **R**esearch, Darmstadt
- Construction started in 2017
- 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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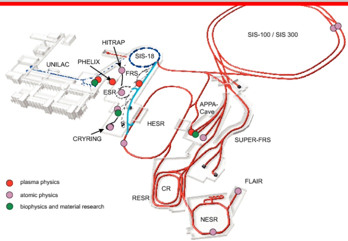

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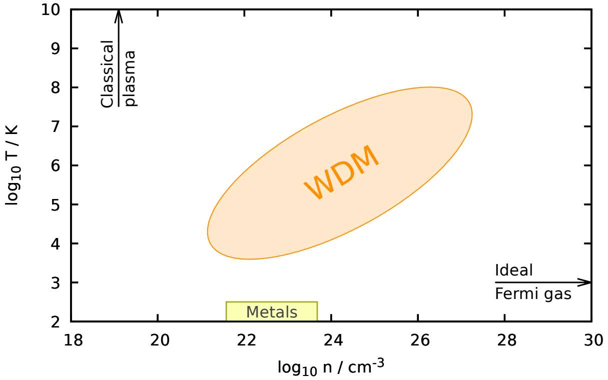
Warm Dense Matter and quantum plasmas: relevant parameters

- **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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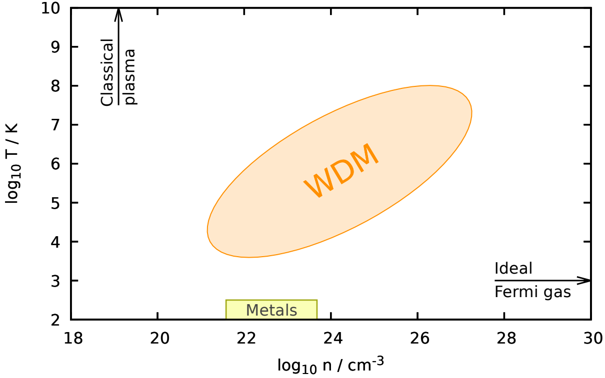

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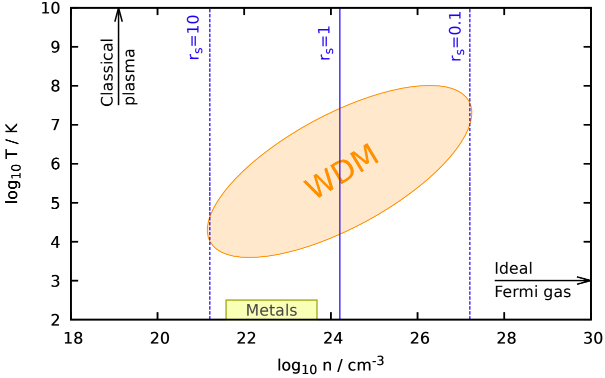

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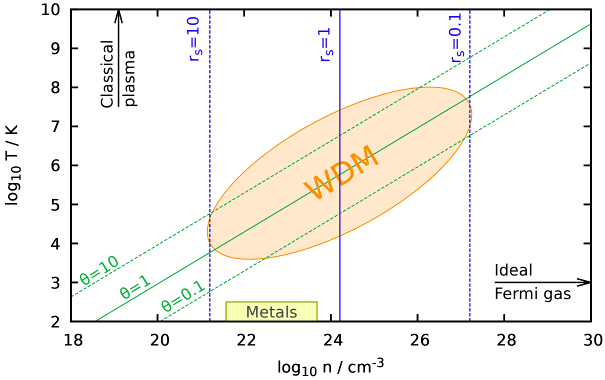

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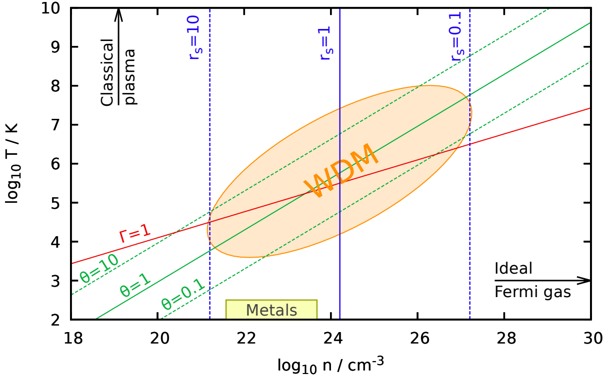

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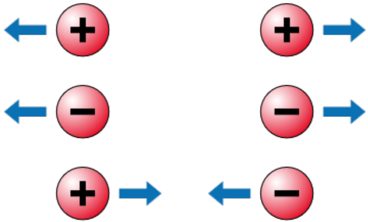
  - Coulomb coupling (non-ideality)



Source: bin-br.at [Img1]

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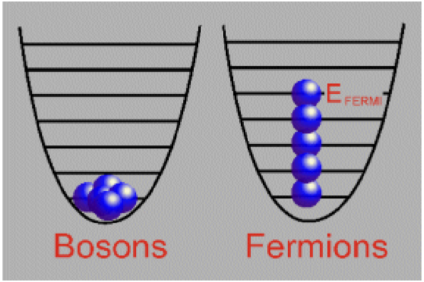

Source: cidehom.com [Img2]

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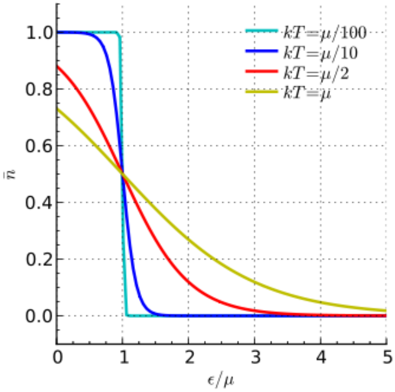
- **Nontrivial interplay of many effects:**
  - Coulomb coupling (non-ideality)
  - Fermionic exchange (anti-symmetry)
  - Thermal excitations (statistical description)



Source: en.wikipedia.org [Img3]

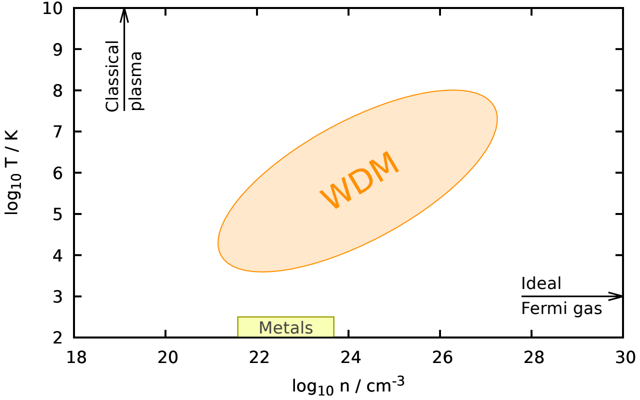
How to experimentally diagnose warm dense matter?

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
- Nonequilibrium: complex time evolution



Experimental strategies: 1. X-ray diffraction: morphology of solid and liquid state,

2. Transport (conductivity) and optics (e.g. X-ray absorption)

3. Recent breakthroughs: light scattering (X-ray Thomson scattering) indirect access to temperature, density, charge state, plasmon dispersion/damping...

How to theoretically approach warm dense matter?

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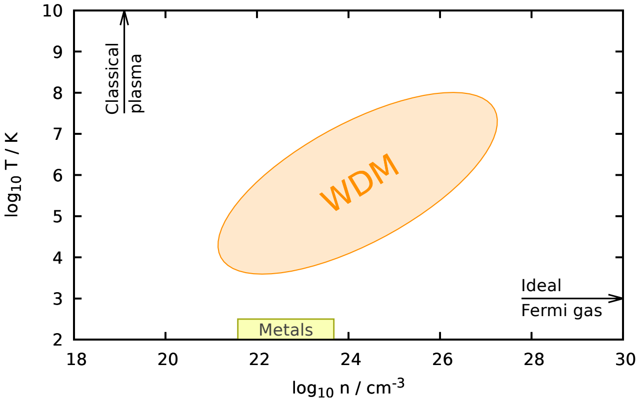

Theoretical strategies: **1. Make a complex (but poor) model of the entire "mess", e.g. phenomenology, hydrodynamics, DFT, or**

2. Perform an excellent description of key component: electrons

⇒ Series of recent breakthroughs: exact quantum Monte Carlo approach: 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**:

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

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

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Warm dense matter ($T \sim T_{\mathrm{F}}$):

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

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

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

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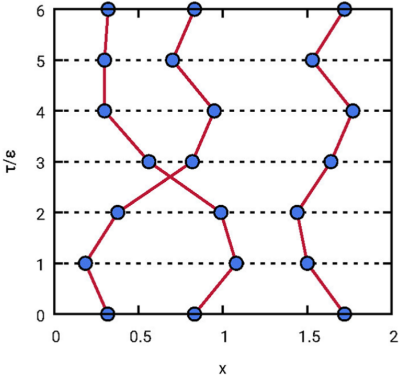
Path Integral Monte Carlo (PIMC): Fermions

- **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, $W(\mathbf{X}) < 0$

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

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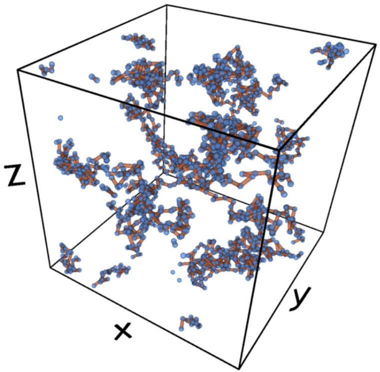
- Randomly generate all possible paths **X** using the **Metropolis algorithm**



Snapshot of PIMC simulation of UEG with $N = 19$, $r_s = 2$, $\theta = 0.5$ (fluctuating probability density)

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

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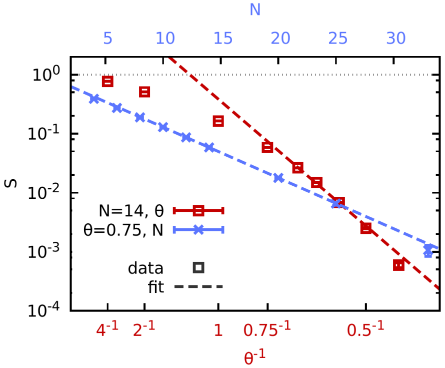
- Sign changes due to particle exchange lead to vanishing signal-to-noise ratio

$\Rightarrow$ **Fermion Sign Problem (unsolved!)**

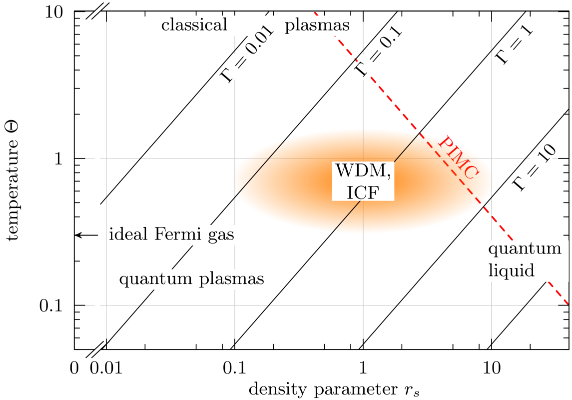


Exponential decrease of the average sign S with system size N and quantum degeneracy θ^{-1}

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

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

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



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[5] T. Dornheim *et al.*, New J. Phys. **17**, 073017 (2015)
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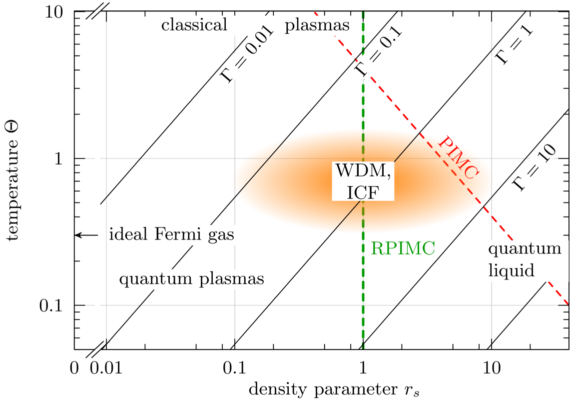
  - First results[1] by E. Brown, D. Ceperley *et al.* (2013) based on ***fixed node approximation*** (**RPIMC**)
  - Induces **systematic errors** of unknown magnitude
  - **RPIMC** limited to $r_s \gtrsim 1$
  - Fermionic **PIMC**: Filinov et al.[2] limited to $r_s \gtrsim 1$



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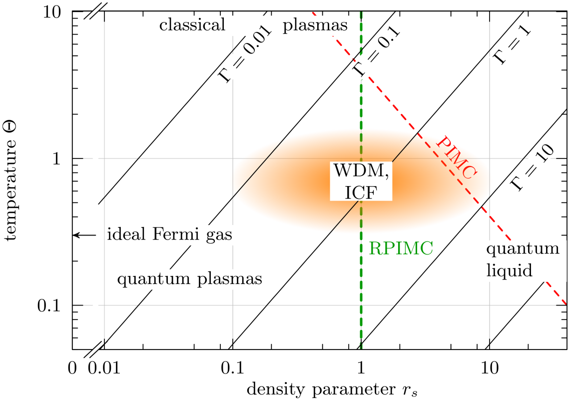
Our approach:

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



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[5] T. Dornheim *et al.*, New J. Phys. **17**, 073017 (2015)
[6] T. Dornheim *et al.*, J. Chem. Phys. **143**, 204101 (2015)

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

- Standard PIMC in warm dense regime severely hampered by ***fermion sign problem***:
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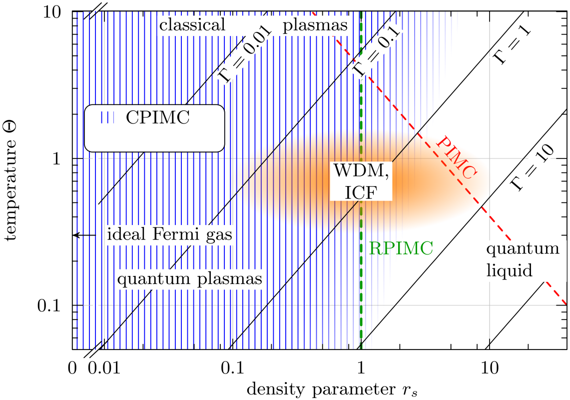
$\rightarrow$ Excels at high density $r_s \lesssim 1$ and strong degeneracy



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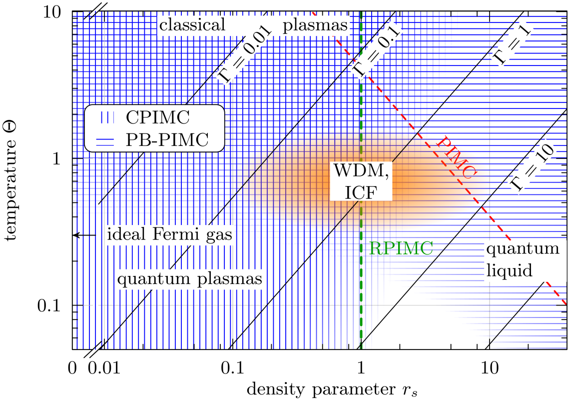

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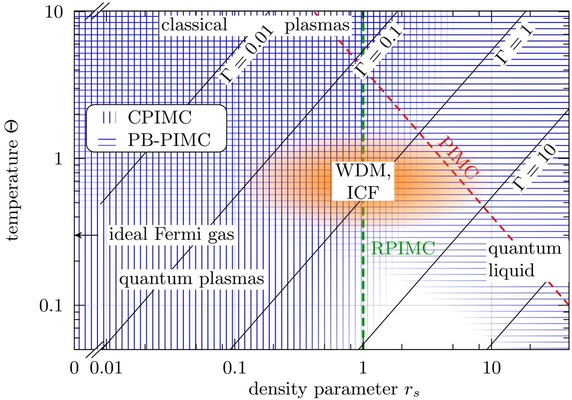

Ab initio simulations over broad range of parameters possible

[1] E.W. Brown *et al.*, PRL **110**, 146405 (2013)
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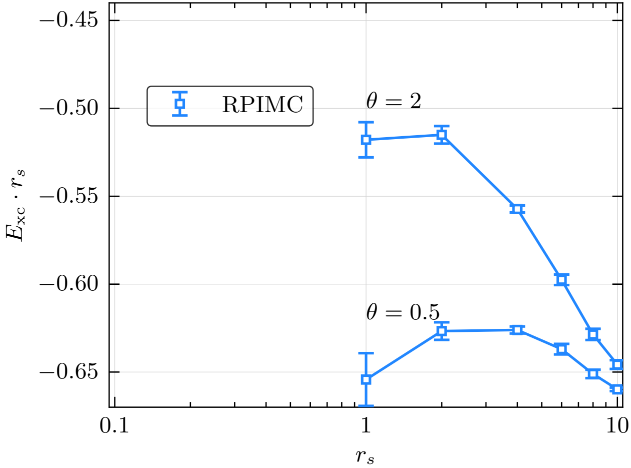
1. Exact exchange-correlation energy $E_{xc} = E - E_0$ (E_0: ideal energy)
($N = 33$ spin-polarized electrons, $\theta \geq 0.5$, $\forall\ r_s$)

- **RPIMC** limited to $r_s \geq 1$



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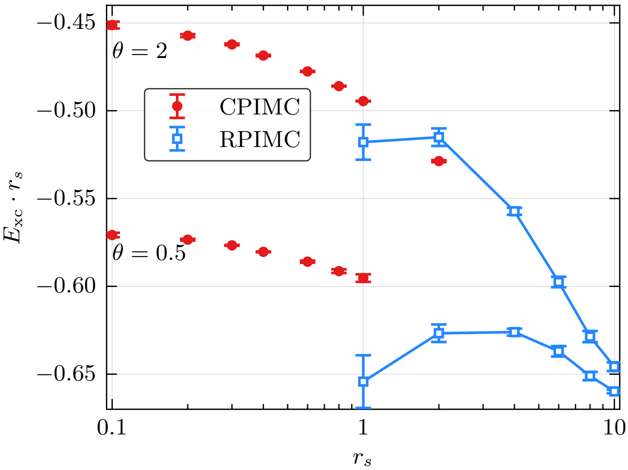

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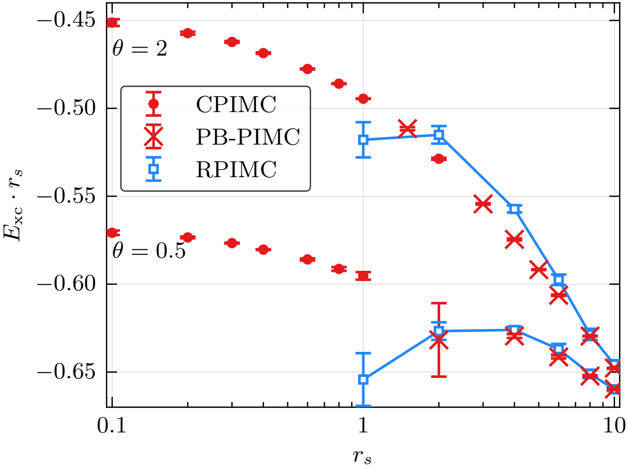

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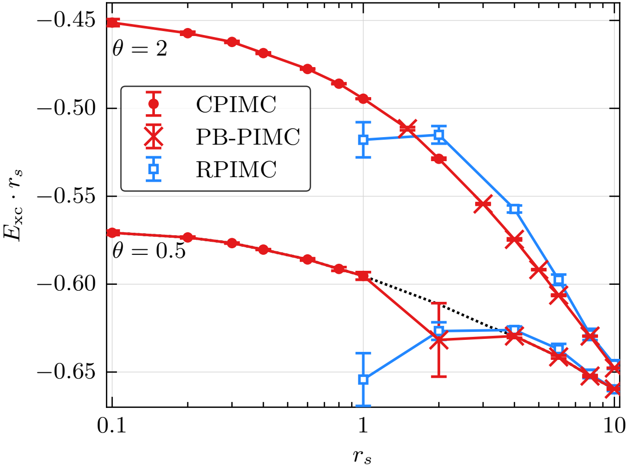
Combination[1] yields exact results over entire density range down to $\theta \sim 0.5$

- 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 $f_{xc}(r_s, \theta, \xi)$, "GDSMFB", with error below 0.3%, Integrated in standard DFT libraries[5]



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Recognition for our work...

APS John Dawson Award 2021

Kiel group:

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

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

UK and US collaborators:
F. Malone, M. Foulkes and T. Sjostrom

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[1], optics etc.: $\chi(\mathbf{q}, \omega)$, $S(\mathbf{q}, \omega)$[2], $\epsilon(\mathbf{q}, \omega)$, $\sigma(\mathbf{q}, \omega)$, plasmon dispersion[3]

PIMC: susceptibilities beyond validity limits of LRT[4]

***Ab initio* spectral properties, momentum distribution $n(p)$**

---

[1] Hamann et al., Phys. Rev. B (2020)
[2] Dornheim et al., Phys. Rev. Lett. 2018
[3] Hamann et al., Contrib. Plasma Phys. 2020
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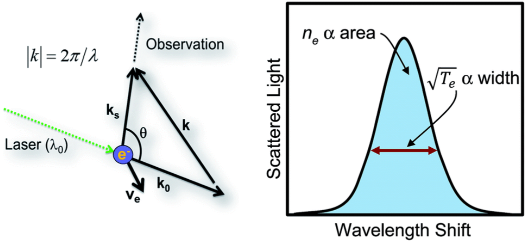
Ab initio dynamic (ω-dependent) results for the warm dense UEG

- **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

data analysis requires model input

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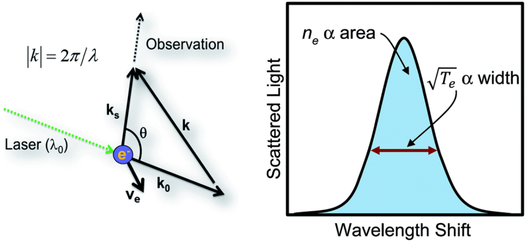
- **Chihara decomposition** applies for non-collective scattering:

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

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



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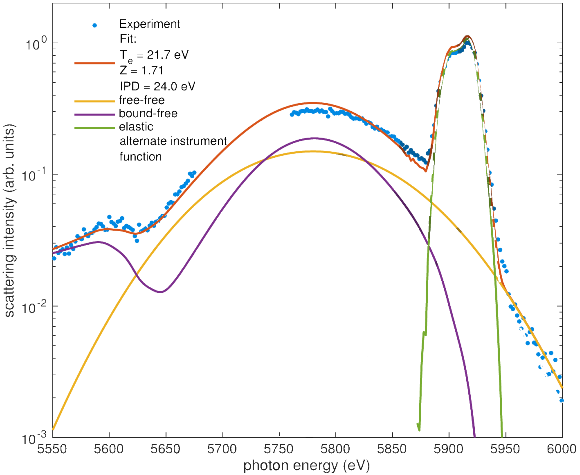
- **Practical example:** Fit model for $S(\mathbf{q},\omega;T_{\text{e}})$ to spectrum to determine electron temperature T_{e}
- **Problem:**
  $F(\mathbf{q},t)$ requires **real time-dependent simulations**
  $\rightarrow$ with PIMC have to use analytic continuation, reconstruct $F(q,it)$ and 4 frequency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



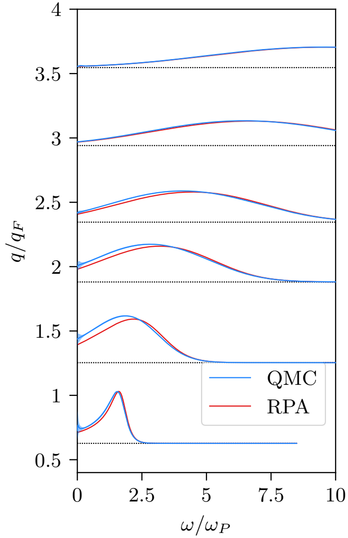
Scattering spectrum of isochorically heated graphite at LCLS.
From D. Kraus *et al.*, *Plasma Phys. Control. Fusion* (2019)

Correlation effects, Landau and collisional damping in $S(q, \omega)$: $\theta = 1$, $r_s = 2$

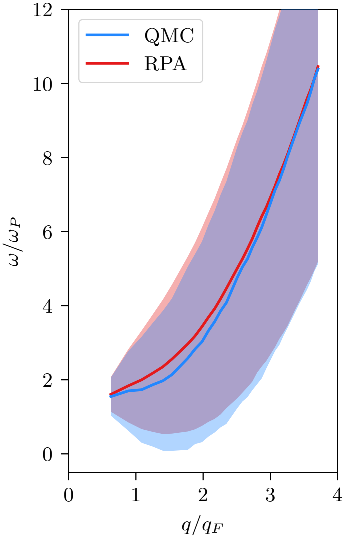
T. Dornheim, S. Groth, J. Vorberger, and M. Bonitz, *Phys. Rev. Lett.* **121**, 255001 (2018)

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

Dynamic structure factor of the UEG:



Peak position and FWHM:



[a]Explanation in Dornheim et al., arXiv:2203.12288

Correlation effects, Landau and collisional damping in $S(q,\omega)$: $\theta = 1$, $r_s = 10$

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

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Pronounced redshift and **broadening** with increasing r_s

Negative dispersion of peak[a] for large r_s around $q = 2q_F$ **predicted for dense hydrogen**

Closely related to plasmons

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

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

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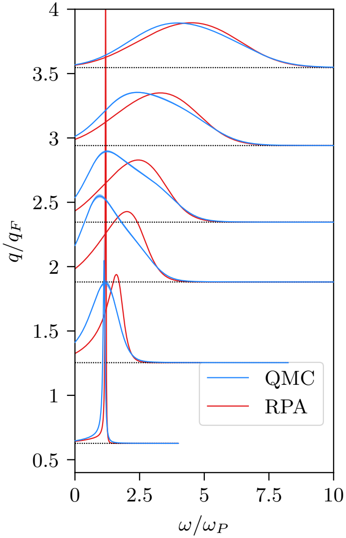

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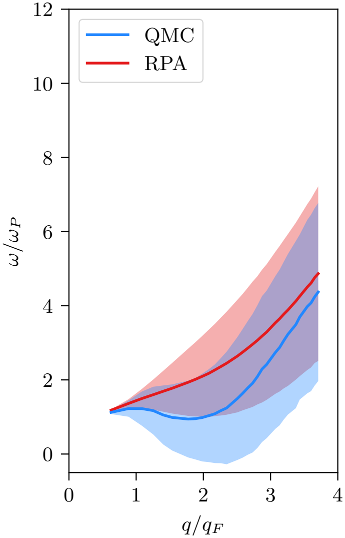
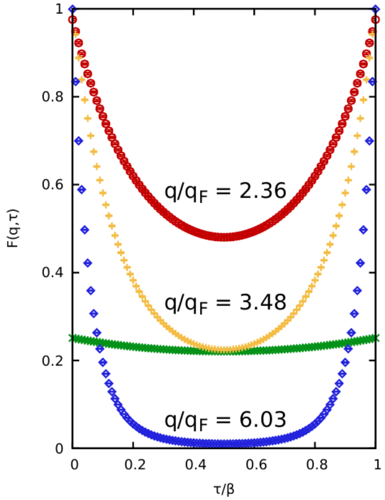


The Static Local Field Correction: *Ab initio* PIMC Simulations

- 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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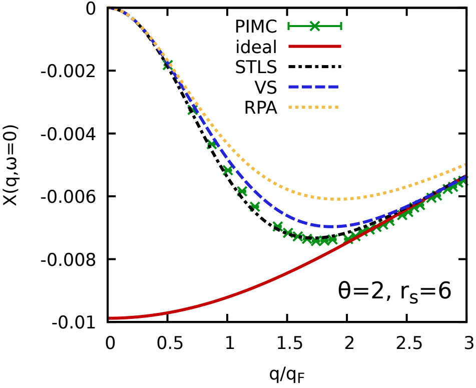
→ full **q**-dependence from a single simulation of the **unperturbed UEG**

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



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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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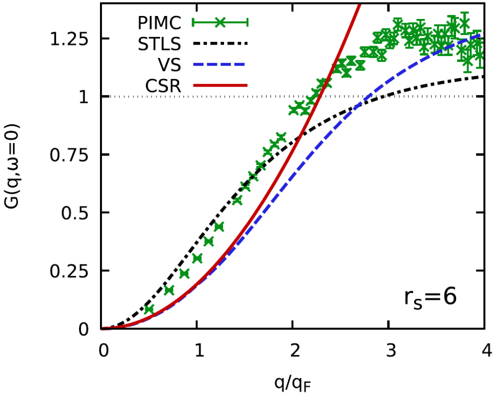
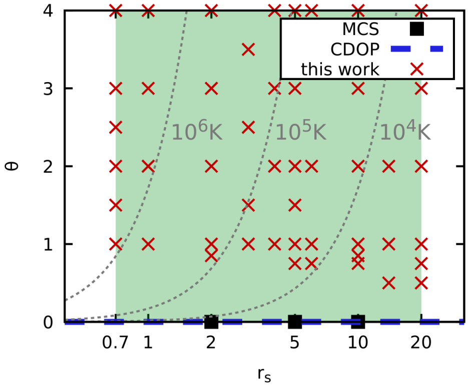


The Static Local Field Correction: Neural-net representation

Extensive set of new PIMC data

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

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



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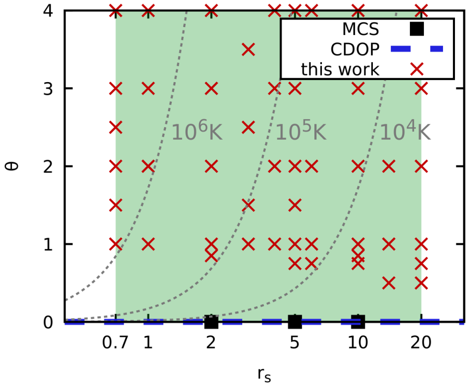

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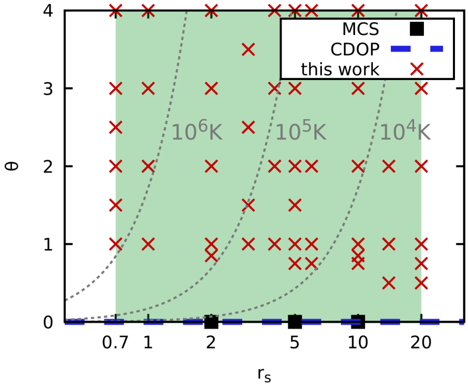

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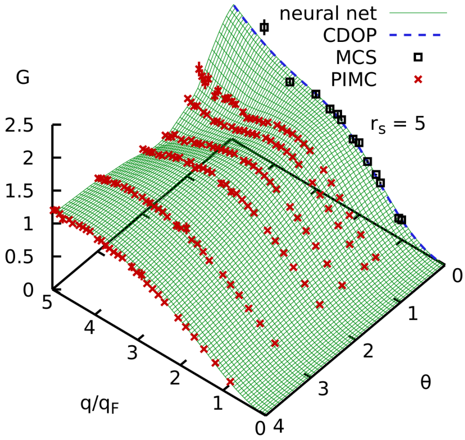

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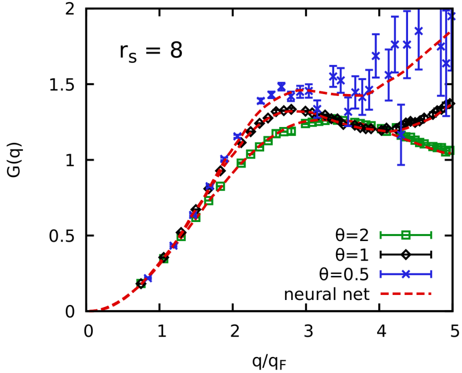
- **Basis for transport quantities, screened ion potential Benchmarks for models and simulations**

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



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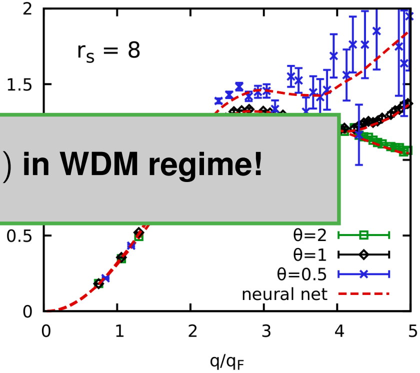
Complete *ab initio* data for $G(q; r_s, \theta)$ in WDM regime!

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



The momentum distribution function (thermodynamic equilibrium)

- **Classical plasma**
  - ideal plasma: Maxwell distribution
  - interacting plasma: Maxwell distribution

  $\Rightarrow$ exponential decay for large momenta

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- **What about nonideal Quantum plasmas?**
  - slower non-exponential decay, $\sim p^{-8}$, predicted[5]
  - relevant for threshold processes: dramatic increase of reaction rates, fusion rates could be possible
  - important for electrons under warm dense matter (WDM) conditions or ions in dense stars
  - First *ab initio* Quantum Monte Carlo results for WDM available:
    K. Hunger *et al.*, Phys. Rev. E **103**, 053204 (2021), arXiv:2101.00842
    T. Dornheim *et al.*, Phys. Rev. B **103**, 205142 (2021)

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[5] Daniel, Vosko (1960); Galitskii, Migdal (1967)

Ab initio CPIMC-results for the momentum distribution[6]

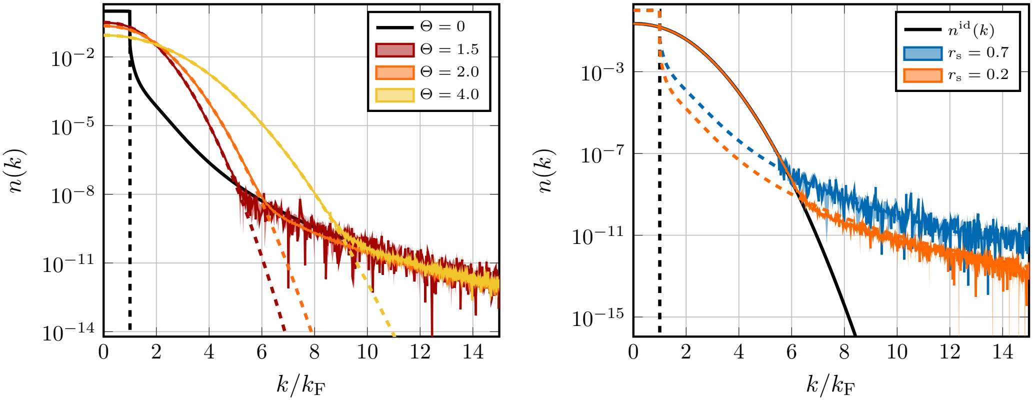
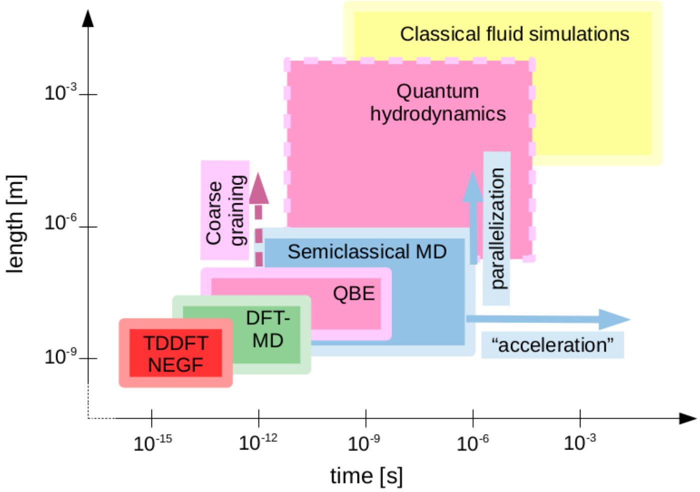


Figure: **Left**: Temperature dependence at $r_s = 0.5$. Full lines: CPIMC, dashed: Fermi function n^{id}.
Right: Density dependence at $\Theta = 2$. Full lines: CPIMC, dashed: ground state, black: n^{id}.

Unprecedented accuracy (11 digits, large k-range), confirm p^{-8} asymptotic, accurate n- and T-dependence
Crucial for reaction rates of threshold processes

[6]K. Hunger *et al.*, Phys. Rev. E **103**, 053204 (2021); T. Dornheim *et al.*, Phys. Rev. B **103**, 205142 (2021)
ground state results: Gori-Giorgi *et al.* (2001)

Nonequilibrium simulations of warm dense matter



- QBE: Quantum Boltzmann equation
- NEGF: Nonequilibrium Green Functions
- TDDFT: time-dependent DFT
- additional methods for classical plasmas: PIC/MCC, Boltzmann equation

Figure: Approximate range of applicability of different methods; from Bonitz *et al.*, Phys. Plasmas **27** (4), 042710 (2020)

Quantum kinetic theory simulations[7]

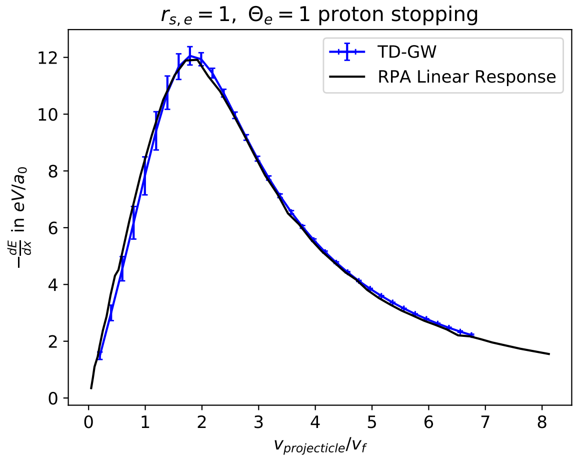


Figure: Stopping power of protons in a dense electron plasma. Time-dependent solution of quantum kinetic equation with the generalized Kadanoff-Baym ansatz, compared to linear response calculations involving *ab initio* QMC-input (C. Makait, Z. Moldabekov, and M. Bonitz, to be published)

- NEGF are the most accurate approach to nonequilibrium quantum plasmas, but very CPU time costly

- recently we achieved a dramatic acceleration [Schlünzen et al., PRL 2020]

⇒ NEGF results will provide benchmarks for real-time TDDFT and deliver improved xc-functionals

⇒ TDDFT results will provide benchmarks for QHD and improved Bohm potential V_B

⇒ Basis for accurate time-dependent quantum simulations over large time and length scales

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[7] M. Bonitz, *Quantum Kinetic Theory*, 2nd ed., Springer 2016

Quantum hydrodynamics for shock propagation

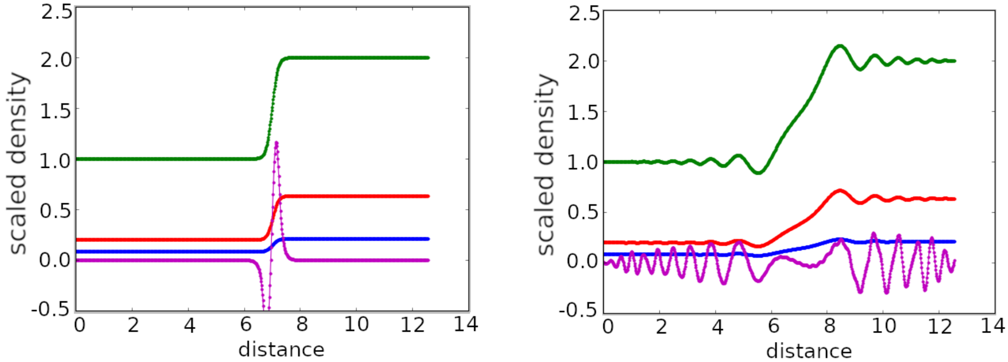


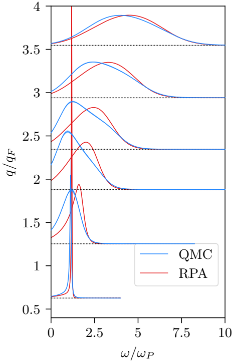
Figure: Influence of th Bohm potential (pink) on the density profile (green) of a running shock. Dense plasma of $r_s = 2$ and $\Theta \approx 0$. Left: initial state, right: $t = 0.62 a_B / c_s$. Red: Thomas-Fermi pressure, blue: exchange pressure.
From: Graziani *et al.*, Contrib. Plasma Phys. (2022), arXiv:2109.09081

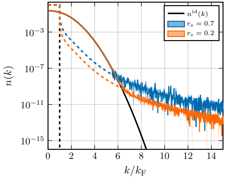
The Bohm potential V_B causes a shear force (stretching) of the shock front. Accurate form of V_B is crucial.[8]

[8]Moldabekov *et al.*, Phys. Plasmas **25**, 031903 (2018); Bonitz *et al.*, Phys. Plasmas **26**, 090601 (2019)

Warm dense matter and quantum plasmas – Summary and outlook[9]

- **Crucial for astrophysics and laboratory experiments**:
  - complex state of matter, between condensed matter and plasmas
  - New facilities: accurate experimental results (e.g. X-ray Thomson scattering)
- **Accurate simulation results now available**:
  - *Ab initio* QMC results for the electron component, avoid sign problem[a].
  - Benchmarks, input for analytical models and for DFT and QHD
  - *Ab initio* results for transport and dielectric properties, momentum distrib.[b]
- **Outlook: accurate multiscale nonequilibrium simulations**[c]:
  - combination of Green functions, TDDFT, and QHD[d]
  - further improvement of CPIMC, extension to two-component plasmas





[a] Dornheim *et al.*, Phys. Reports (2018)
[b] Dornheim *et al.*, PRL (2018); Hamann *et al.*, PRB (2020); Hunger *et al.*, PRE (2021)
[c] Bonitz *et al.*, Phys. Plasmas (2020)
[d] Bonitz *et al.*, Phys. Plasmas (2019); Moldabekov *et al.*, SciPost Phys. (2022) arXiv:2103.08523

[9] references and talk at http://www.theo-physik.uni-kiel.de/bonitz/research.html