Understanding electronic correlations in warm dense quantum plamas

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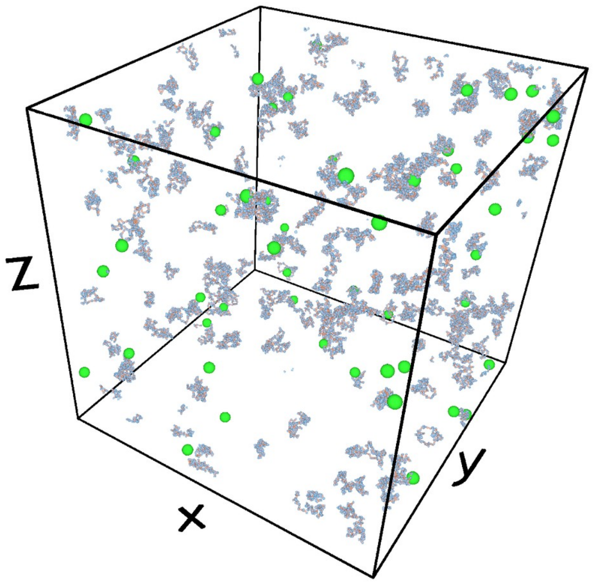
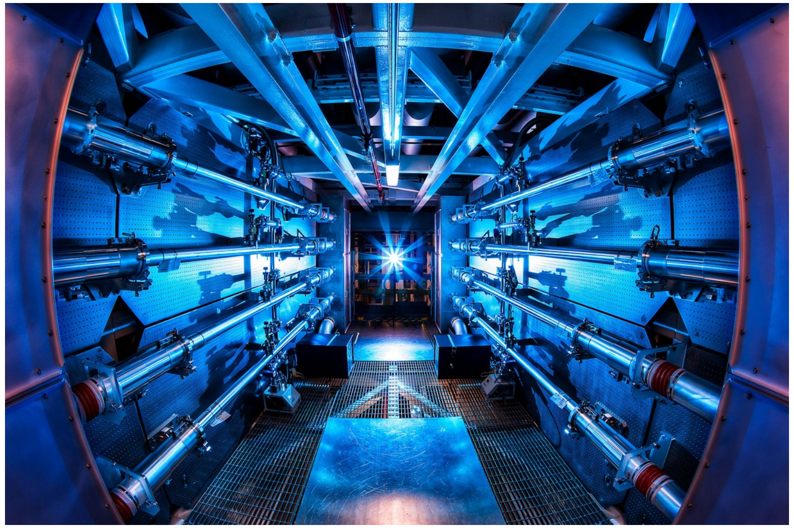
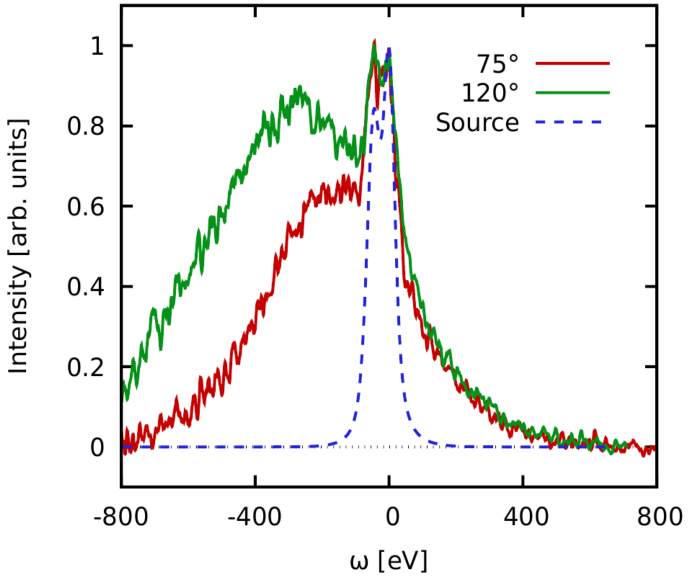






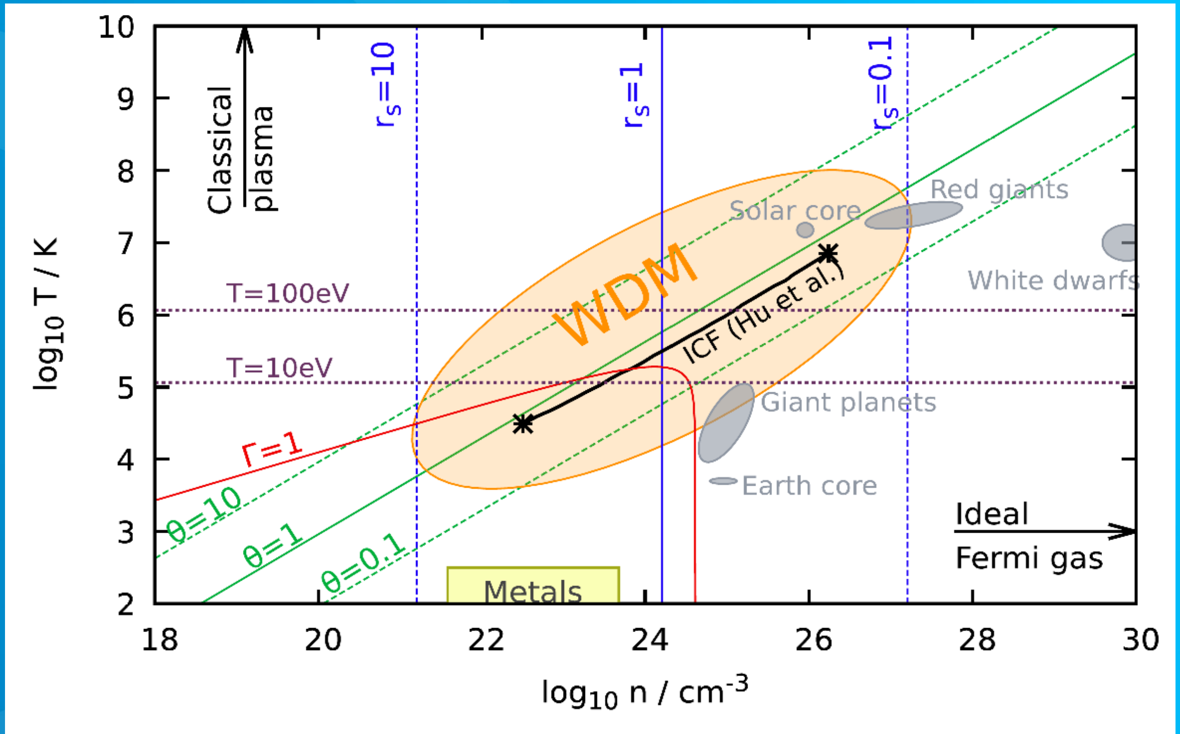
In collaboration with:

HZDR HELMHOLTZ ZENTRUM DRESDEN ROSSENDORF
CASUS CENTER FOR ADVANCED SYSTEMS UNDERSTANDING
Lawrence Livermore National Laboratory
European XFEL
KTH VETENSKAP OCH KONST
Universität Rostock Traditio et Innovatio

European Research Council Established by the European Commission

Outline

Part I: Introduction



Taken from: **T. Dornheim**, Zh. Moldabekov, K. Ramakrishna, P. Tolias, A. Baczewski, D. Kraus, Th. Preston, D. Chapman et al, Phys. Plasmas **30**, 032705 (2023)

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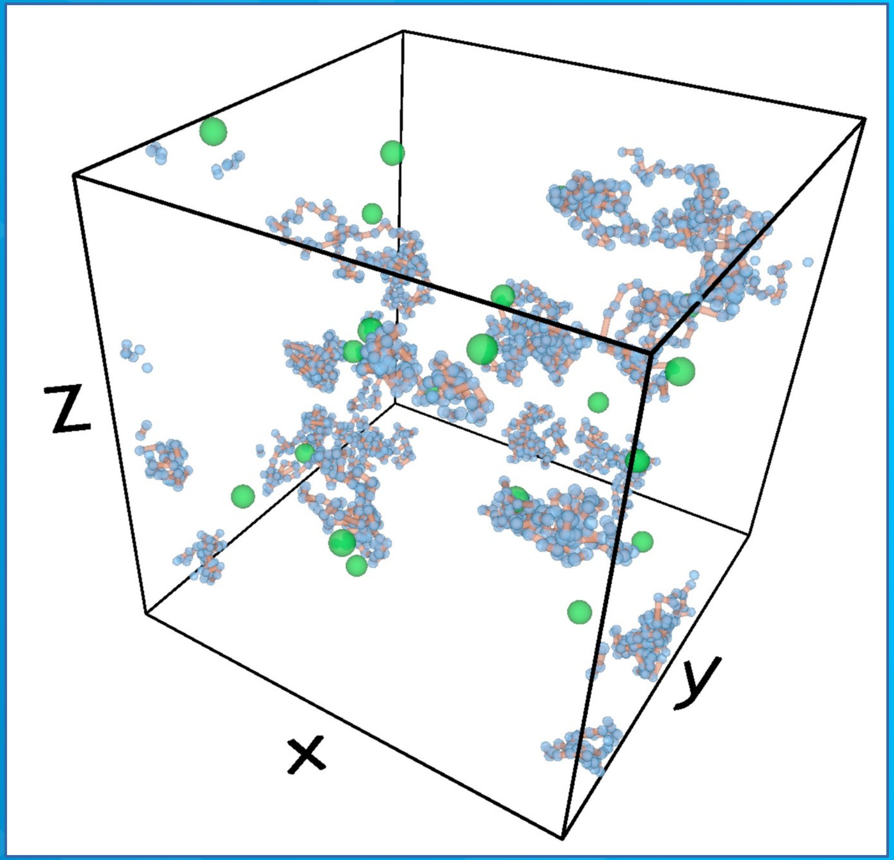
Part I: **Introduction**

Part II: **Electronic density response of warm dense matter**



Taken from: M. Böhme, Zh. Moldabekov, J. Vorberger, and **T. Dornheim**, Phys. Rev. Lett. **129**, 066402 (2022)

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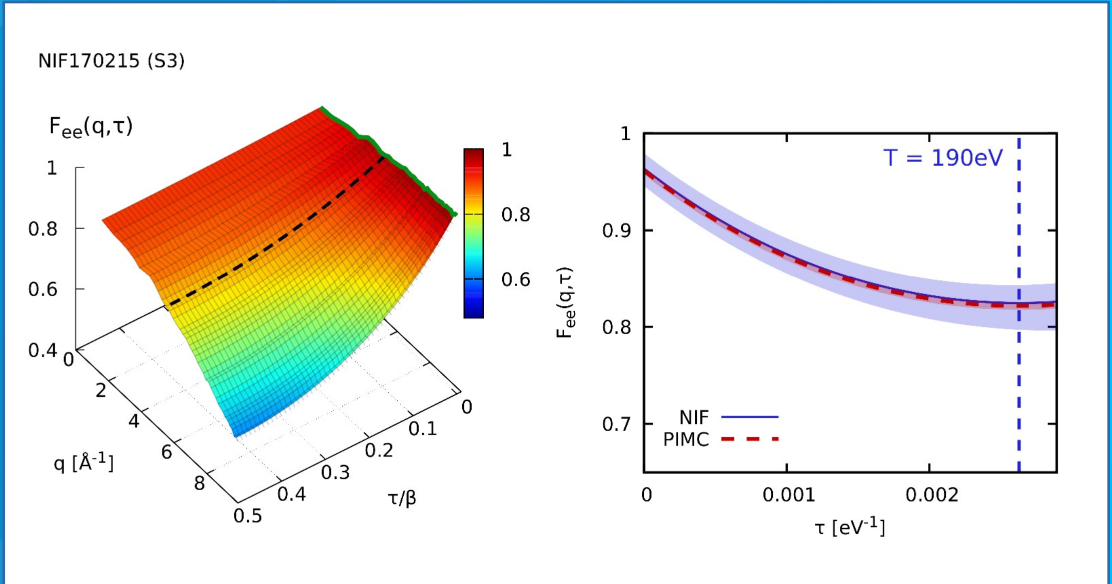
Part III: Imaginary-time correlation functions and XRTS



Taken from: **T. Dornheim**, Zh. Moldabekov, M. Böhme, J. Vorberger, P. Tolias, F. Graziani, and T. Döppner (in preparation)

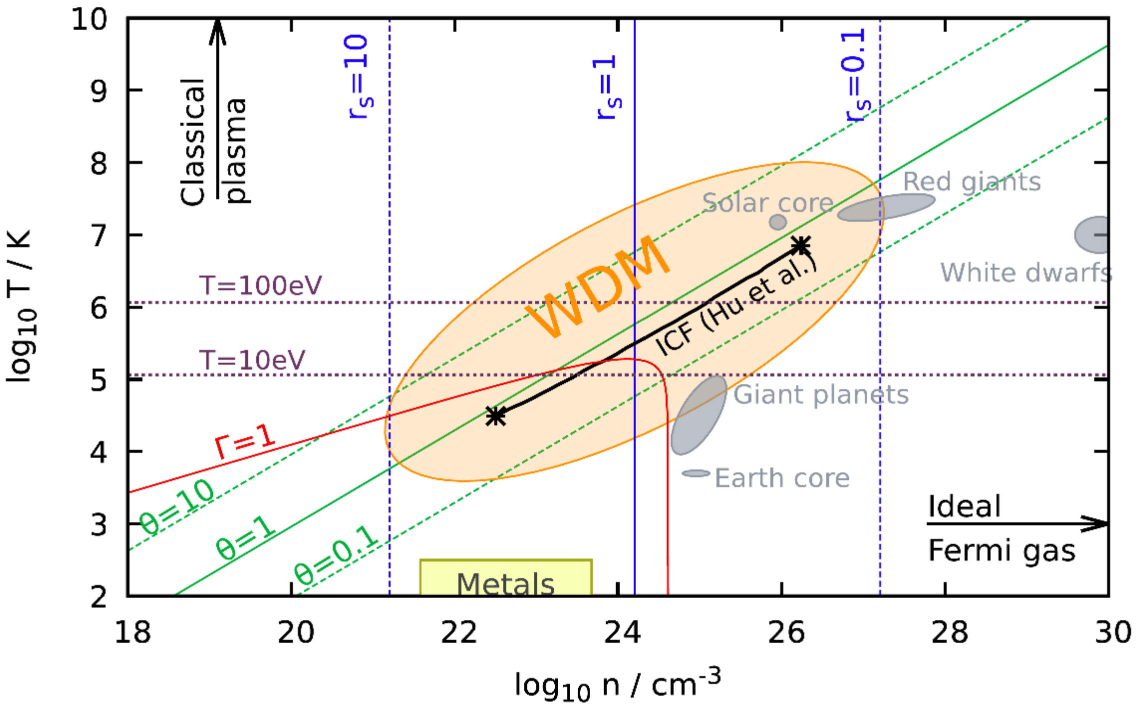
Part I: Introduction

Warm Dense Matter (WDM)

- Matter under extreme density / temperature ubiquitous throughout our universe

$r_S \sim \theta \sim \Gamma \sim 1$

→ $r_S = d/a_B$, density parameter, $\theta = k_B T/E_F$, $\Gamma = W/E_{kin}$



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SYSTEMS UNDERSTANDING

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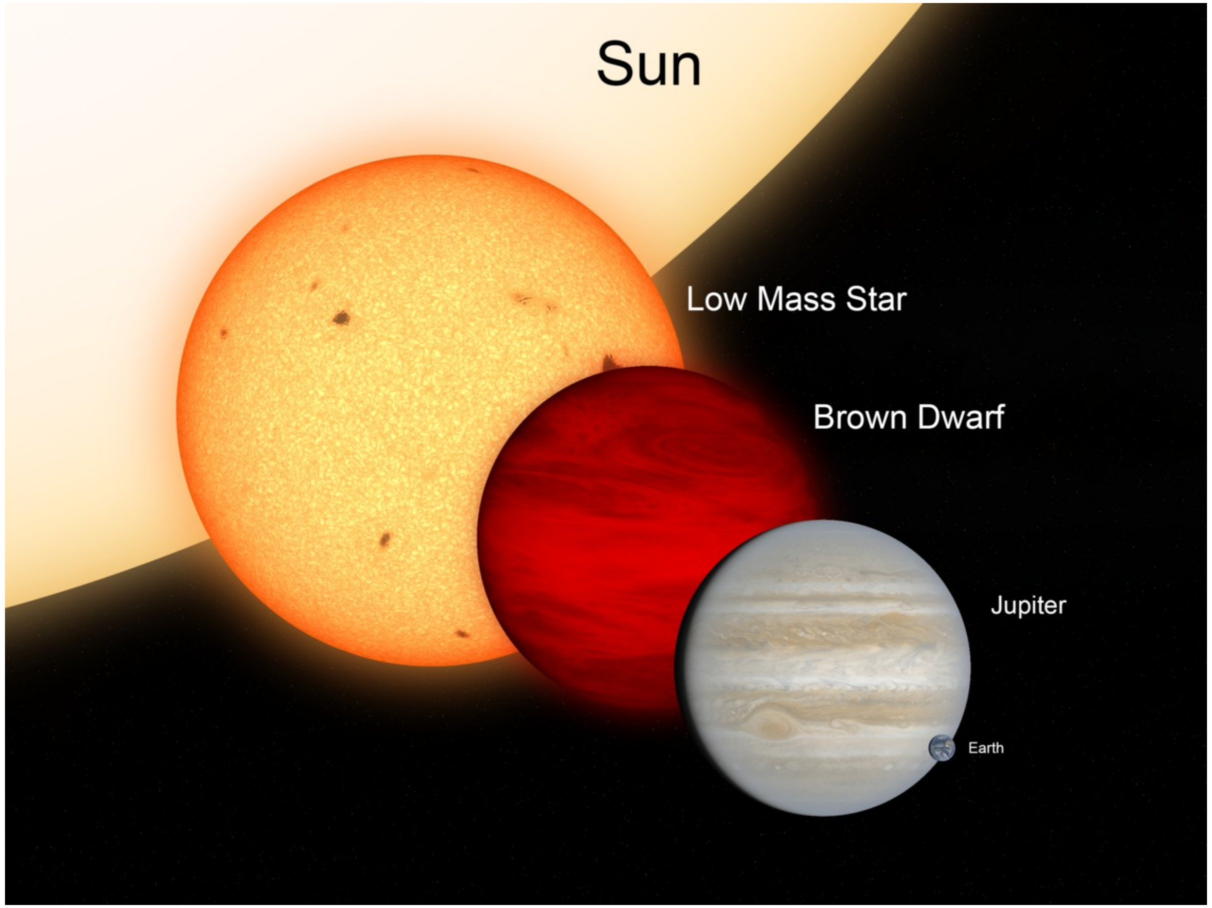
- Examples: giant planet interiors, brown dwarfs



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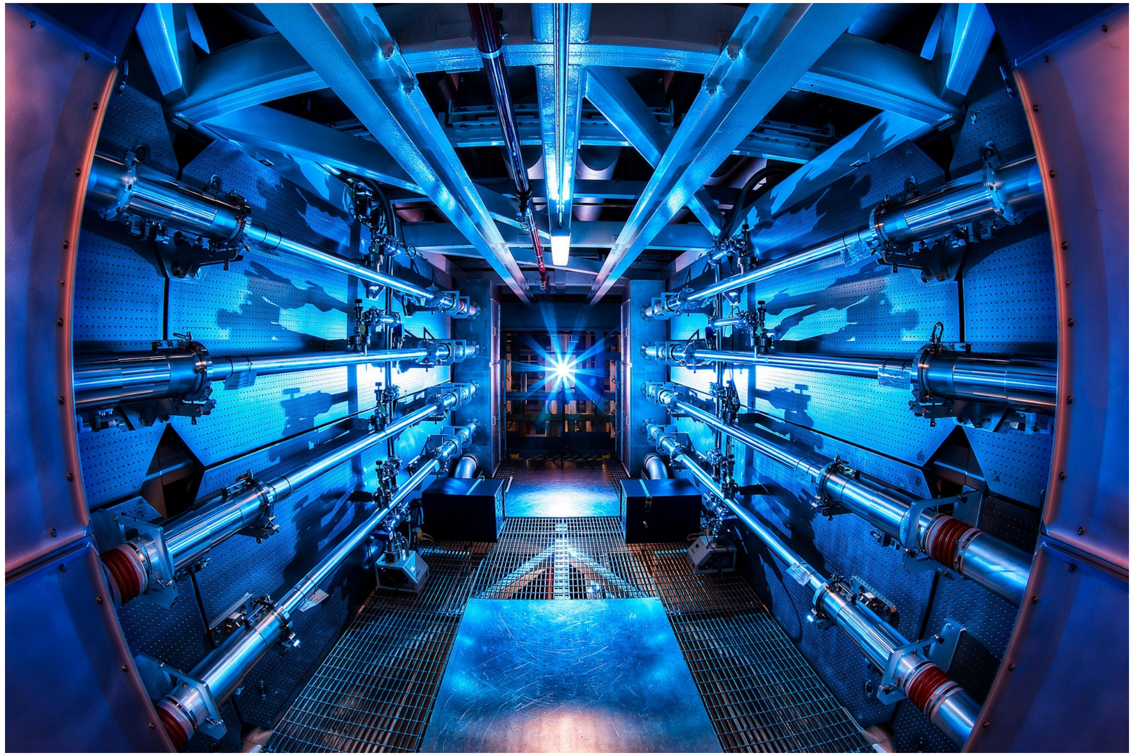
- WDM highly important for technological applications:

→ Inertial confinement fusion, etc.

National Ignition Facility (NIF)



Taken from: Lawrence Livermore National Laboratory

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WDM routinely realized in large research facilities around the globe!

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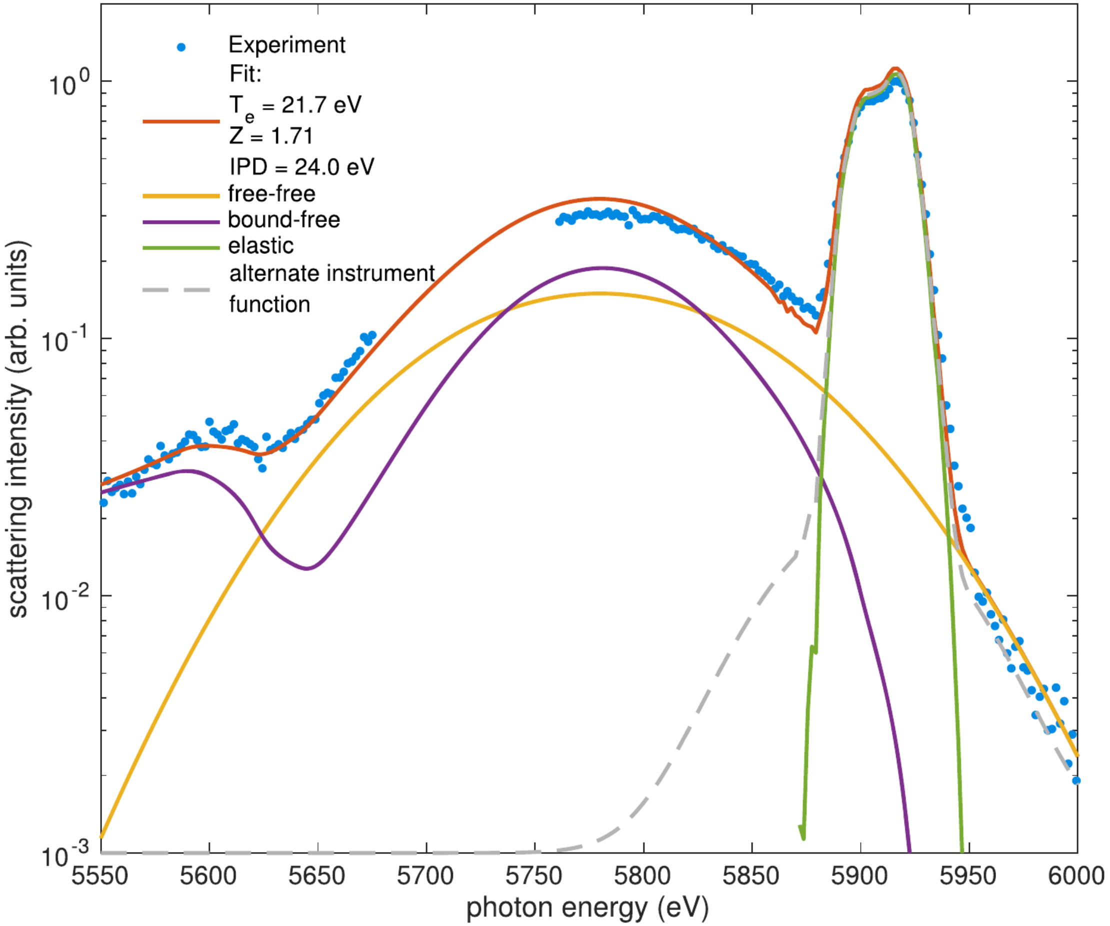
But: Rigorous WDM theory indispensable

- Diagnostics: parameters like *T*, *n*, *Z*, etc. cannot be measured and have to be inferred from theory

→ X-ray Thomson scattering (XRTS)

Isochorically heated graphite at LCLS (Stanford)



Taken from: D. Kraus *et al.*, *Plasma Phys. Control. Fusion* **61**, 014015 (2019)

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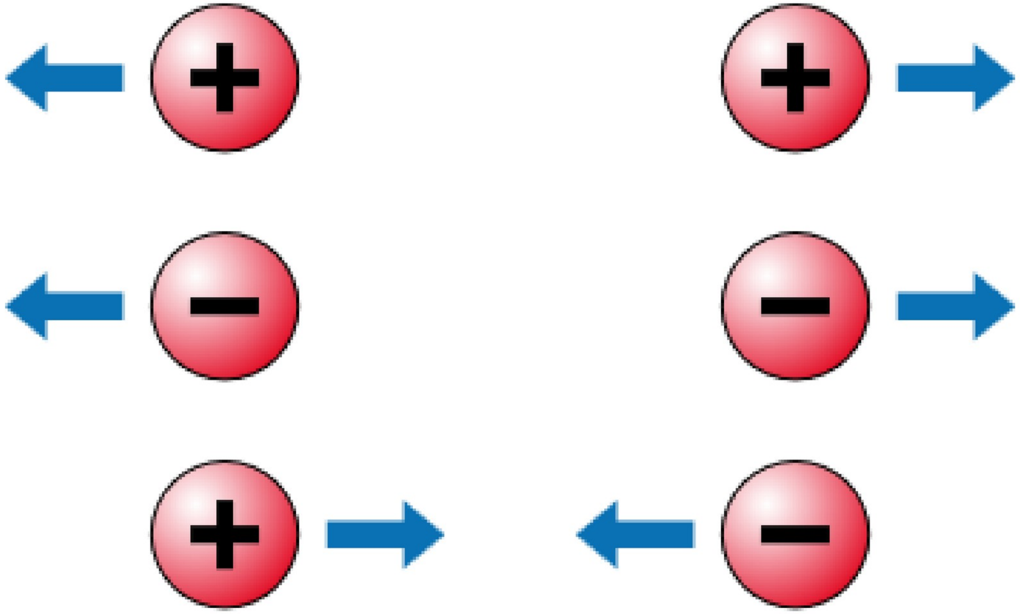
- **WDM theory notoriously challenging**

$r_s \sim \theta \sim \Gamma \sim 1$

→ intricate interplay of:

1) Coulomb coupling



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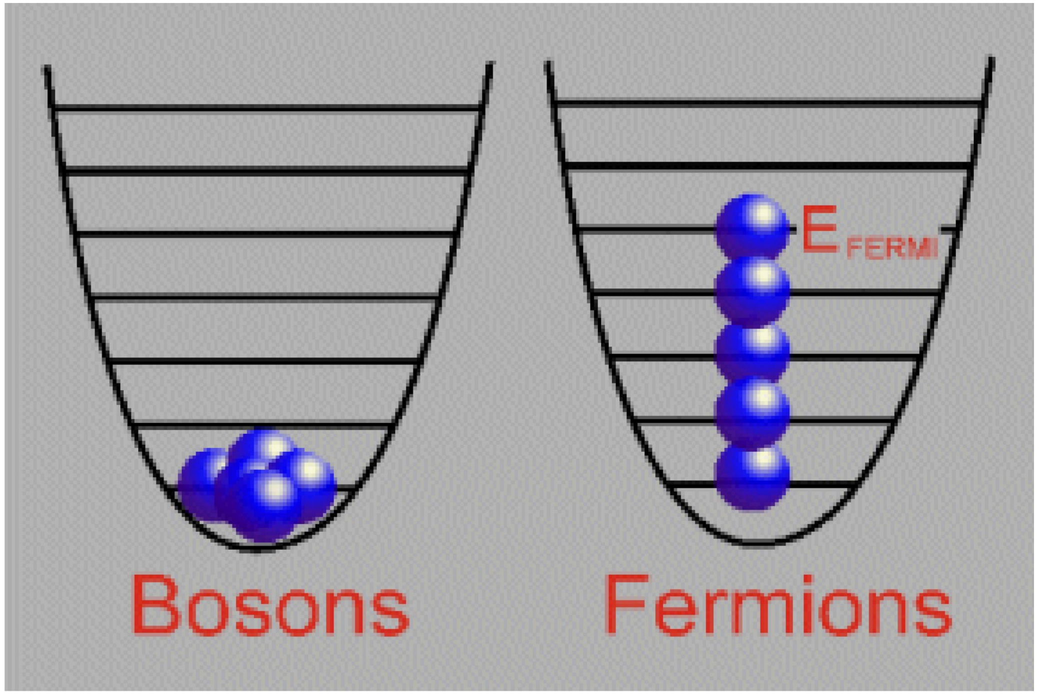

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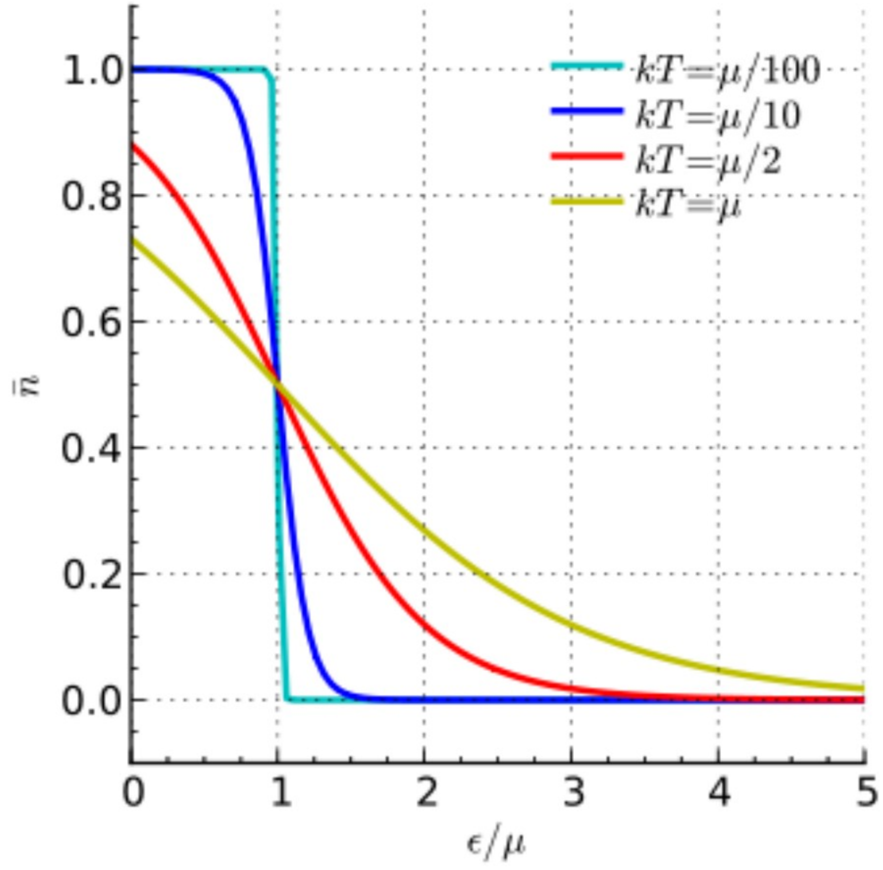
2) quantum degeneracy effects
3) thermal excitations



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How to achieve a real *ab initio* description of WDM?

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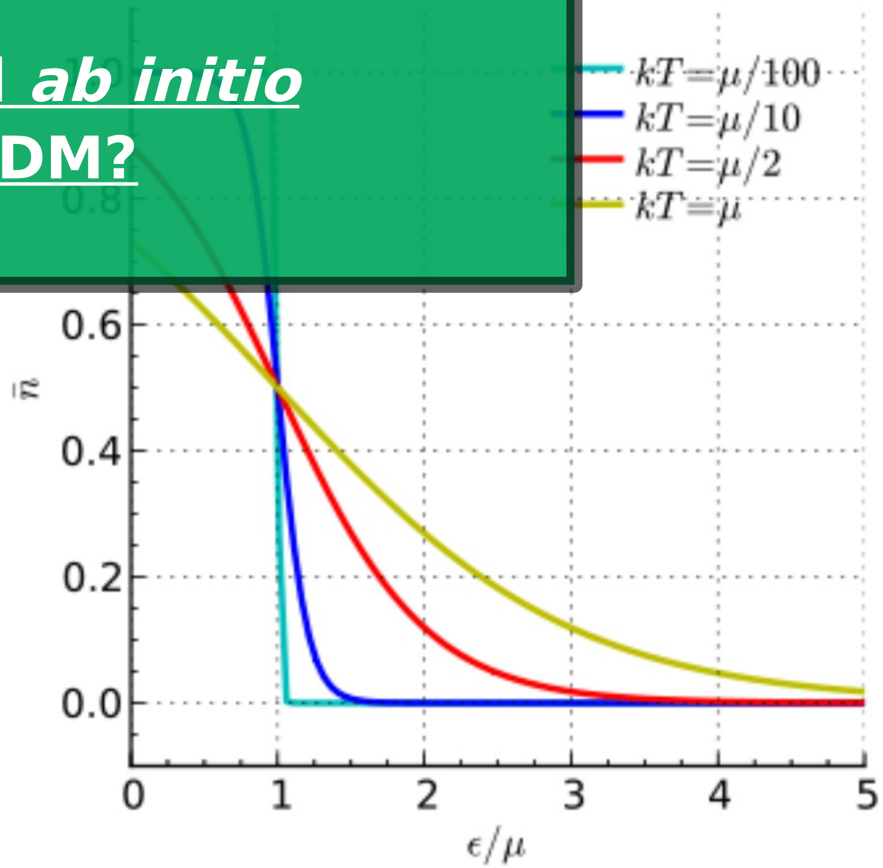

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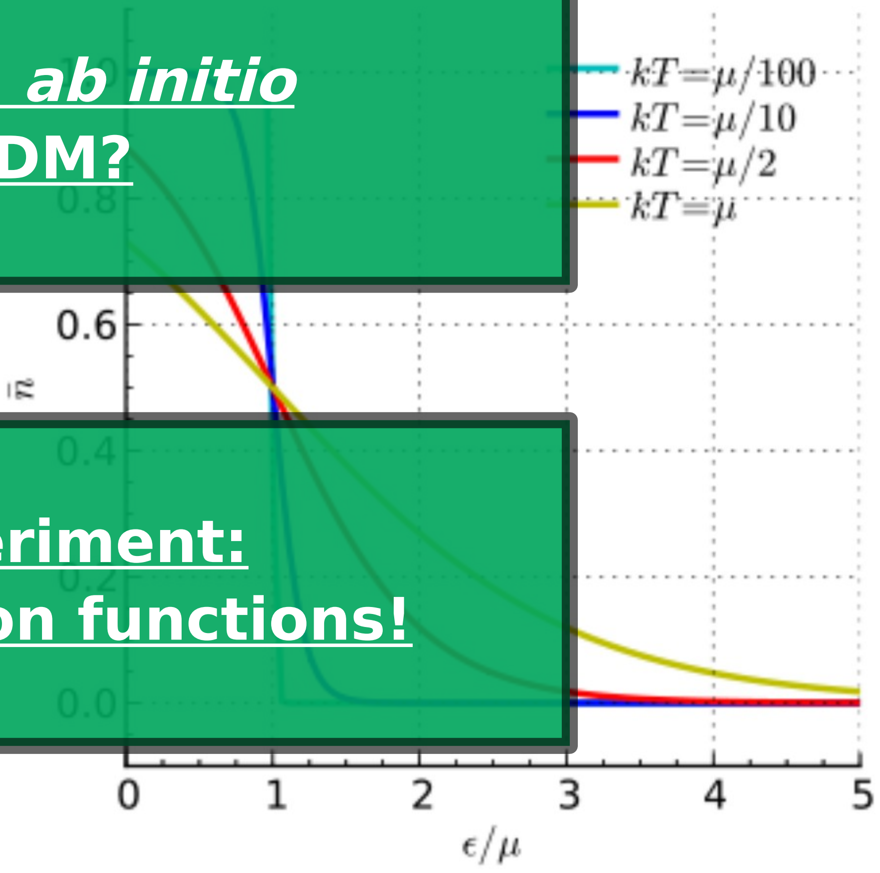

From theory to experiment: Imaginary-time correlation functions!

Ab-initio Quantum Monte Carlo (QMC) simulations

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Problem:

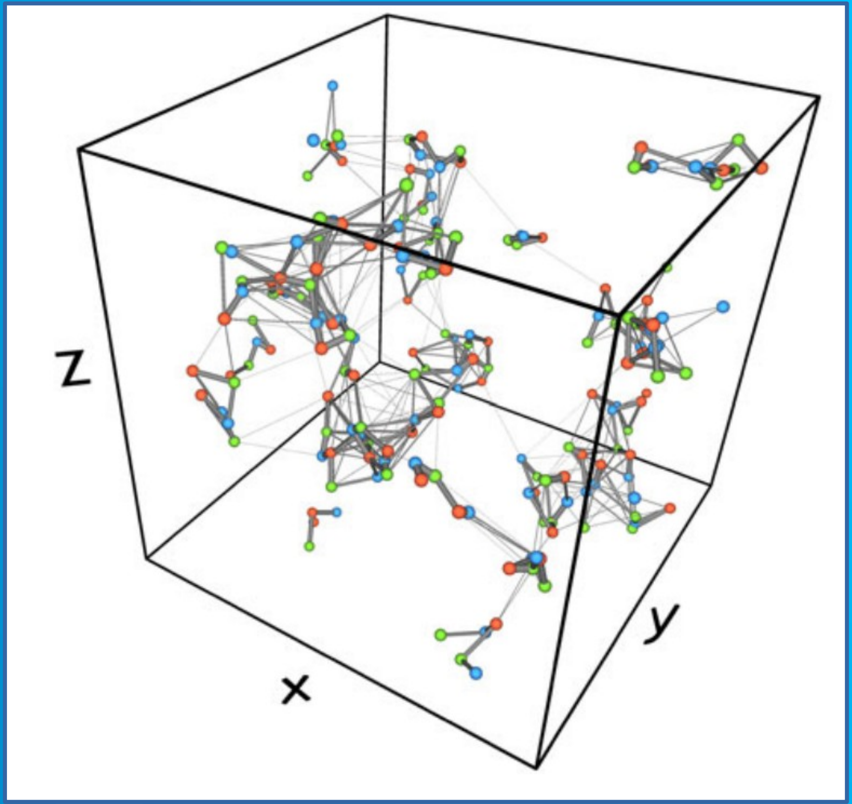
• **Density functional theory (DFT)** etc. require external input about XC-effects

→ finite *T*: XC-free energy f_{xc}

Solution:

• **Quantum Monte Carlo** methods in principle allow for exact solution of quantum many-body problems without any empirical input

• Finite *T*: **Path Integral Monte Carlo (PIMC)**



Taken from: **T. Dornheim**, S. Groth, and M. Bonitz, *Contrib. Plasma Phys.* **59**, e201800157 (2019)

Part I: XC-free energy of UEG

S. Groth, T. Dornheim, T. Sjostrom, F.D. Malone, W.M.C. Foulkes, and M. Bonitz, PRL 119, 135001 (2017)

Impact on thermal DFT simulation of warm dense hydrogen

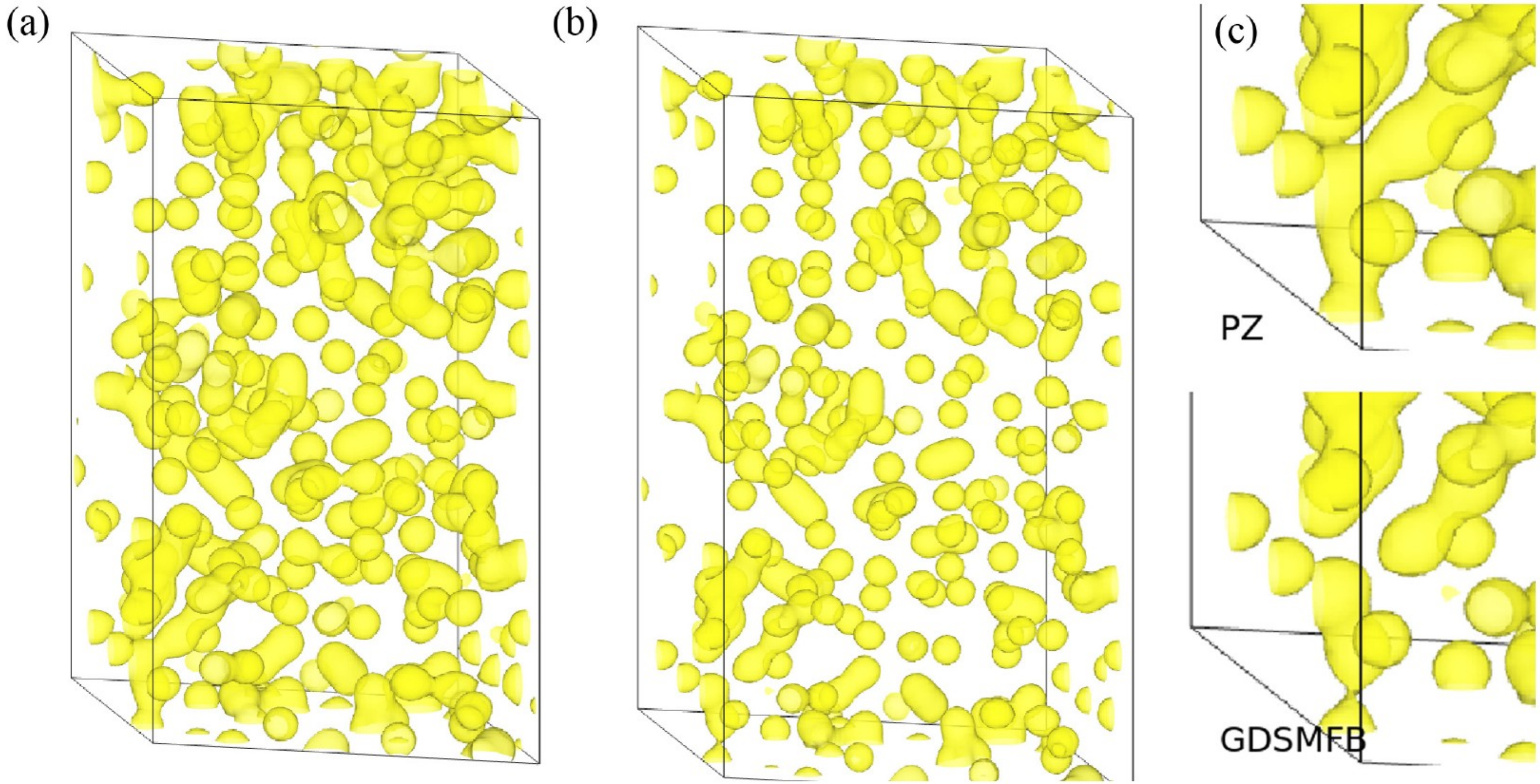
Example:

Hydrogen at T=65,000K

r_s = 2

(a) Ground-state LDA by Perdew And Zunger, PRB (1980) [PZ]

(b) our thermal LDA [GDSMFB]



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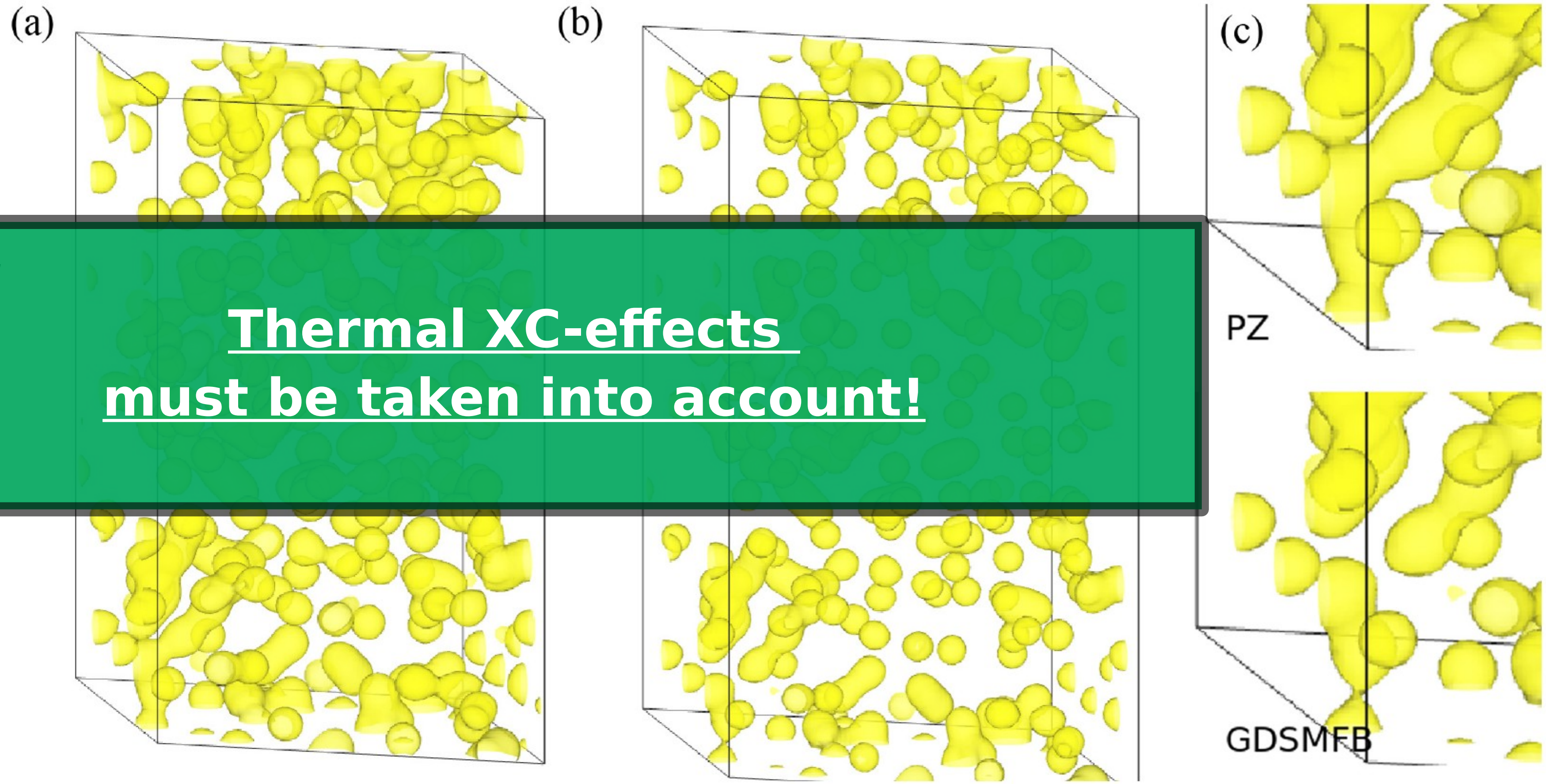

Thermal XC-effects must be taken into account!

Taken from: K. Ramakrishna, **T. Dornheim**, and J. Vorberger, *Phys. Rev. B* **101**, 195129 (2020)

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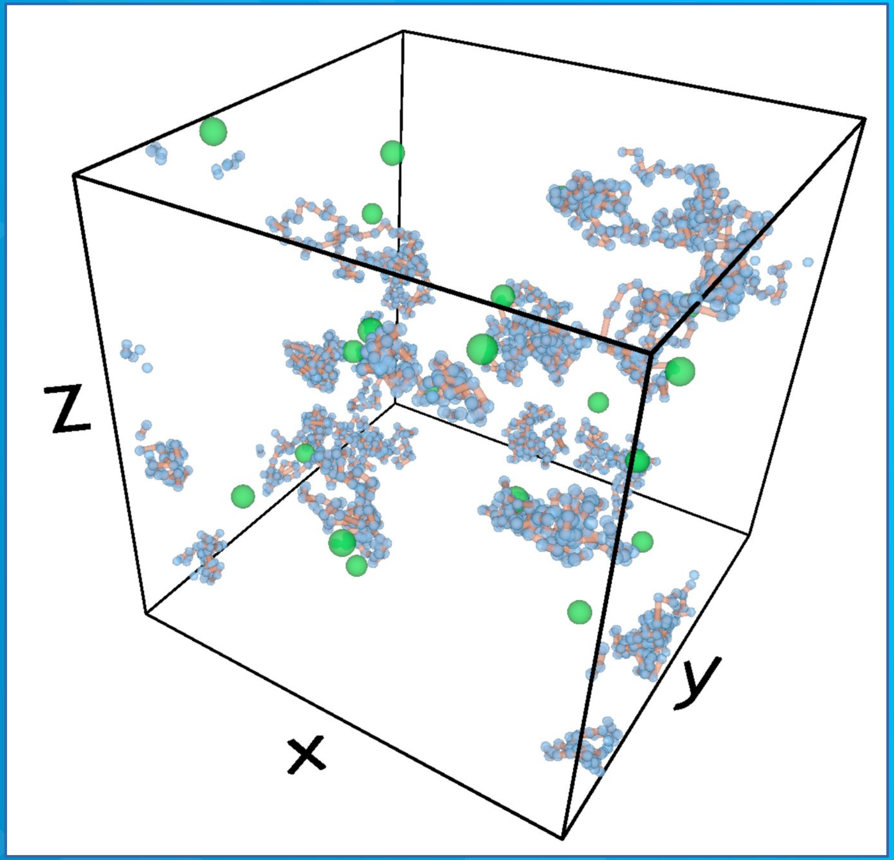


Taken from: M. Böhme, Zh. Moldabekov, J. Vorberger, and **T. Dornheim**, Phys. Rev. Lett. **129**, 066402 (2022)

Part II: Electronic density response of WDM

Density response functions, local field correction

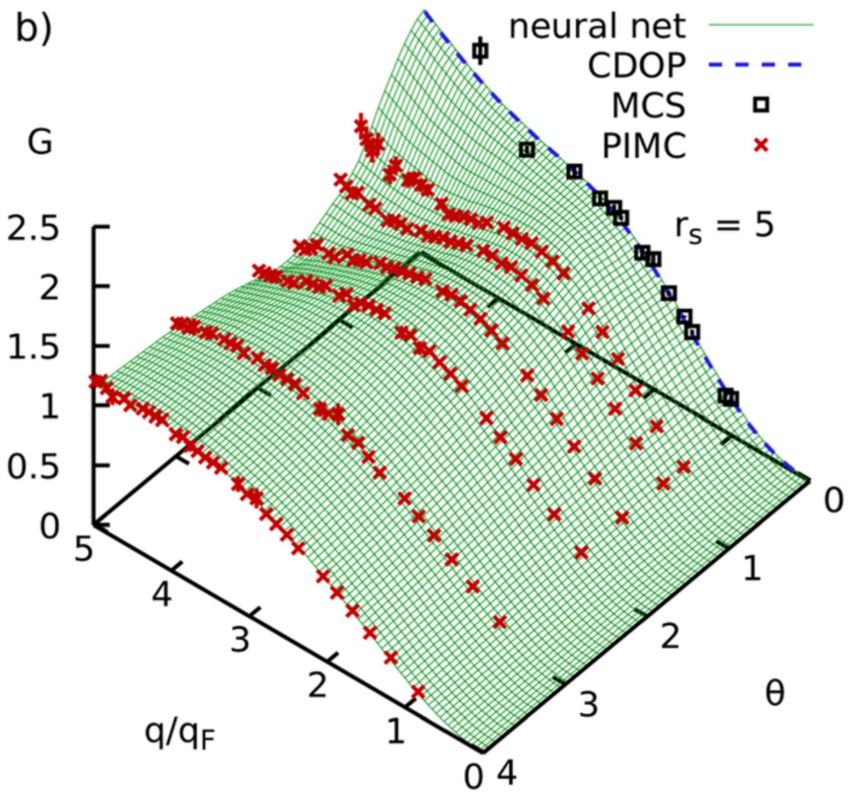
- Dynamic density response function

$$\chi(q,\omega) = \frac{\chi_0(q,\omega)}{1 - \frac{4\pi}{q^2}[1 - G(q,\omega)]\chi_0(q,\omega)}$$

→ $\chi_0(q,\omega)$ ideal density response function

→ $G(q,\omega)$ dynamic local field correction, **containing all electronic XC-effects**

$G(q,\omega) = -K_{xc}(q,\omega)/v(q)$



- Static limit: Exact QMC results for $\chi(q) := \chi(q,0)$, $G(q)$
- Extensive PIMC data for LFC $G(q)$ for ~50 r_s-θ combinations

Neural net representation covering full WDM regime.

Taken from: **T. Dornheim**, J. Vorberger, S. Groth, N. Hoffmann, Zh. Moldabekov, and M. Bonitz, *J. Chem. Phys.* **151**, 194104 (2019)

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- Dynamic density r

$$\chi(q,\omega) = \frac{}{1}$$

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→ $G(q,\omega)$ dynami
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- Static limit: Exact
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Physics of Plasmas ARTICLE scitation.org/journal/php

Electronic density response of warm dense matter

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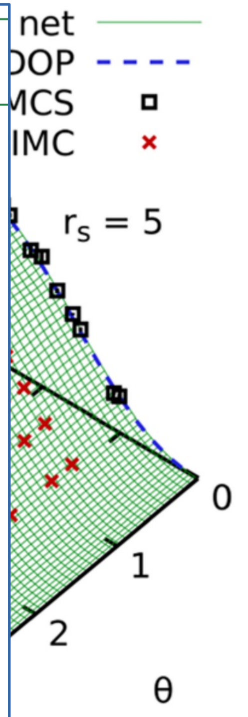

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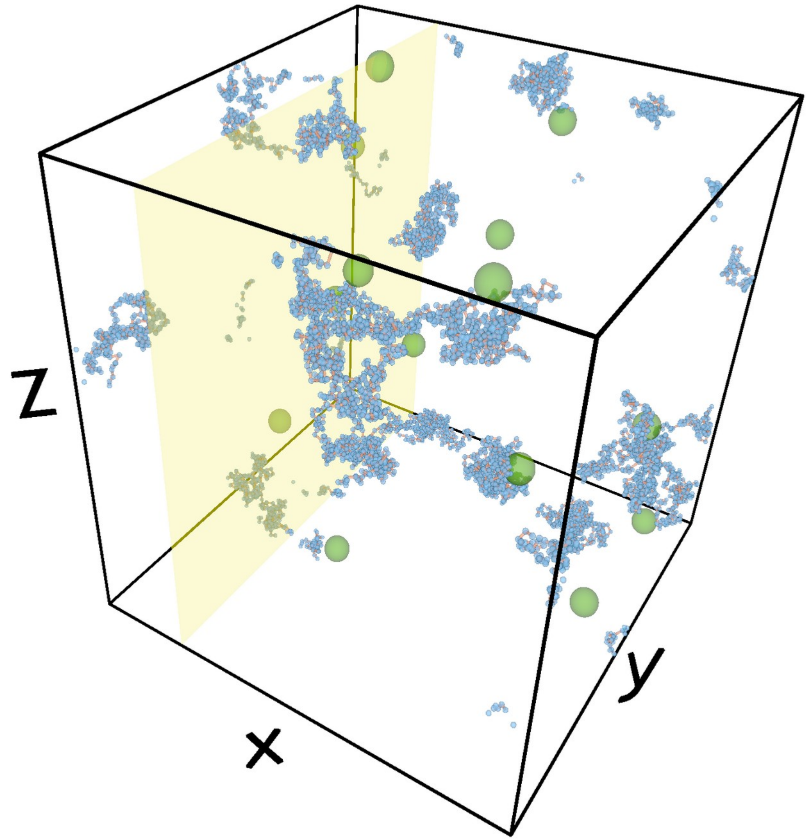
Part II: Electronic density response of WDM

Exact PIMC simulation of H snaphots

•Use PIMC to solve electronic problem in the potential of fixed protons

PIMC snapshot of hydrogen at rs=2, θ=1



Green orbs: protons
Blue paths: quantum degenerate electrons

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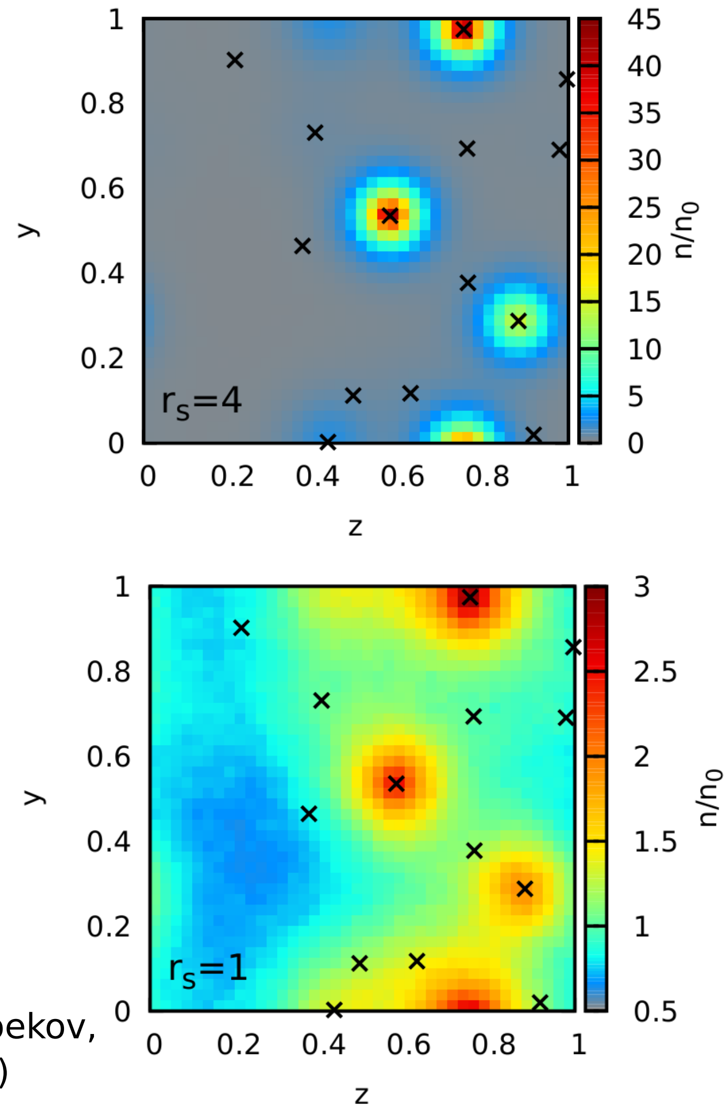
Advantage #1: nanostructure not averared out

→ study electronic localization around protons

Electronic density of H at θ=1



Taken from: **T. Dornheim**, M. Böhme, Zh . Moldabekov, and J. Vorberger, Phys. Rev. E **108**, 035204 (2023)

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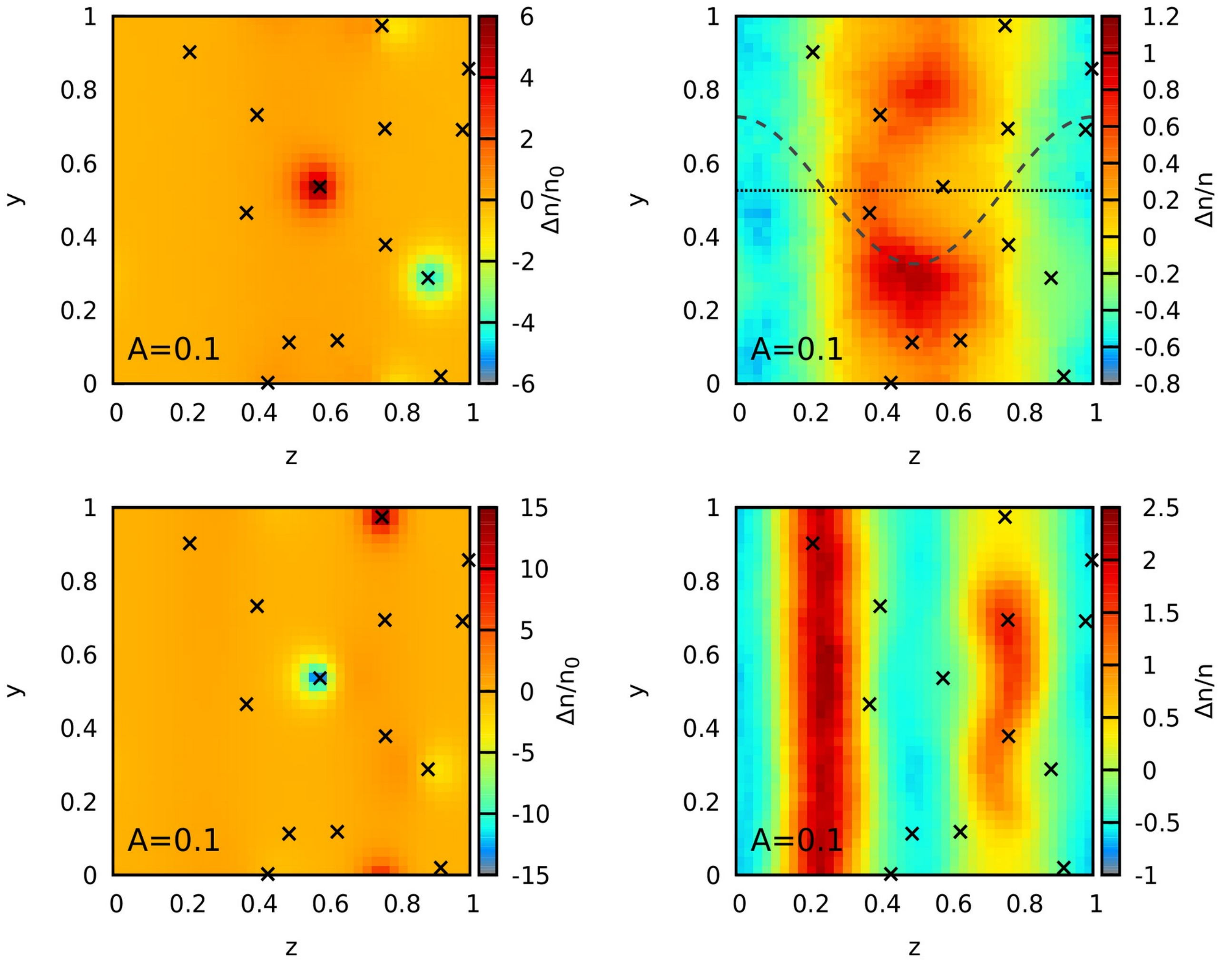
$$\hat{H}_{\mathbf{q},A} = \hat{H} + 2A\sum_{l=1}^{N}\cos\left(\mathbf{q}\cdot\hat{\mathbf{r}}_l\right)$$

→ study spatially resolved density response

Induced electronic density of H at rs=4, θ=1



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Advantage #2: Direct comparison to DFT

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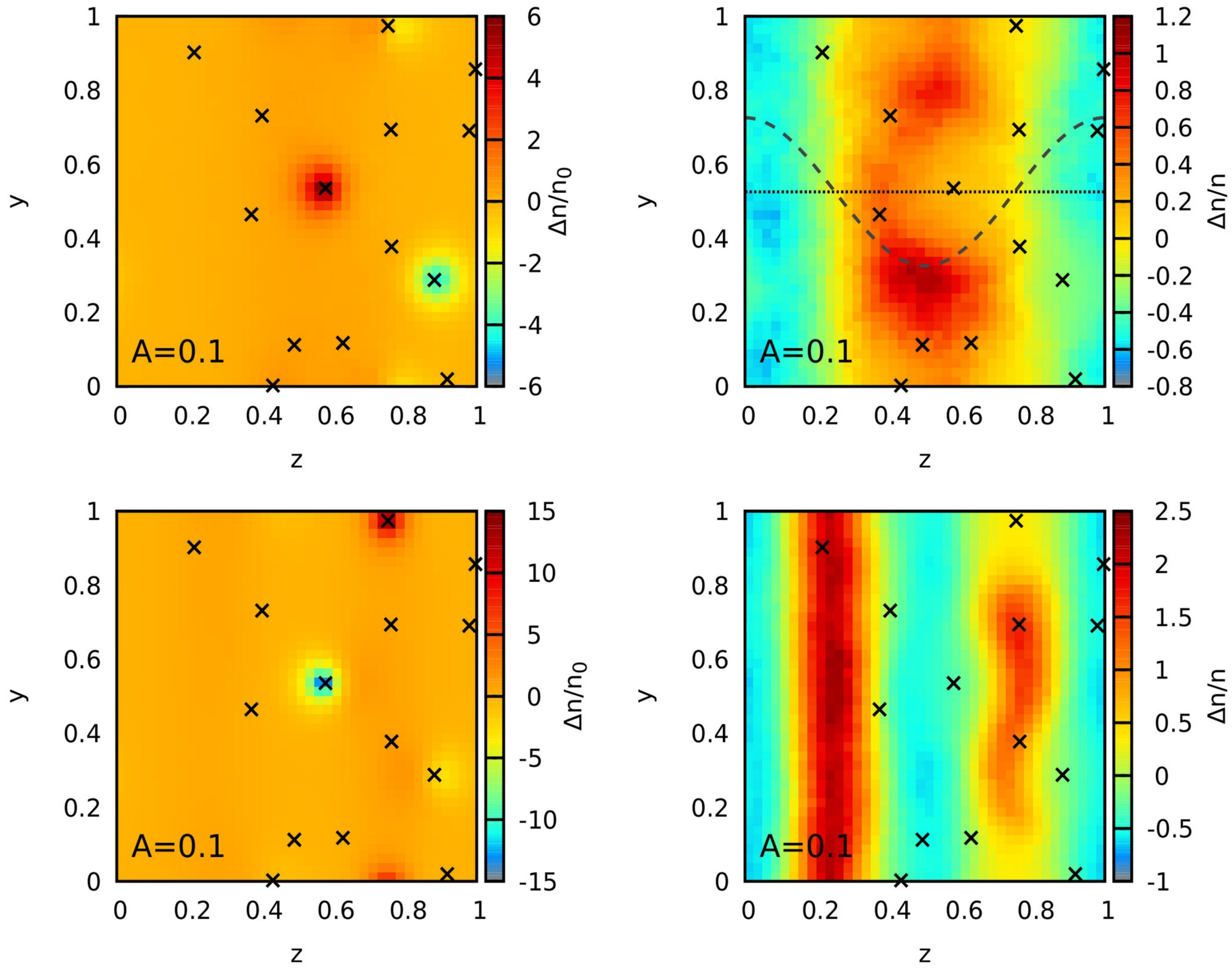

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Part II: Electronic density response of WDM

Exact PIMC results for XC kernel of H

Max Böhme
(PhD student)

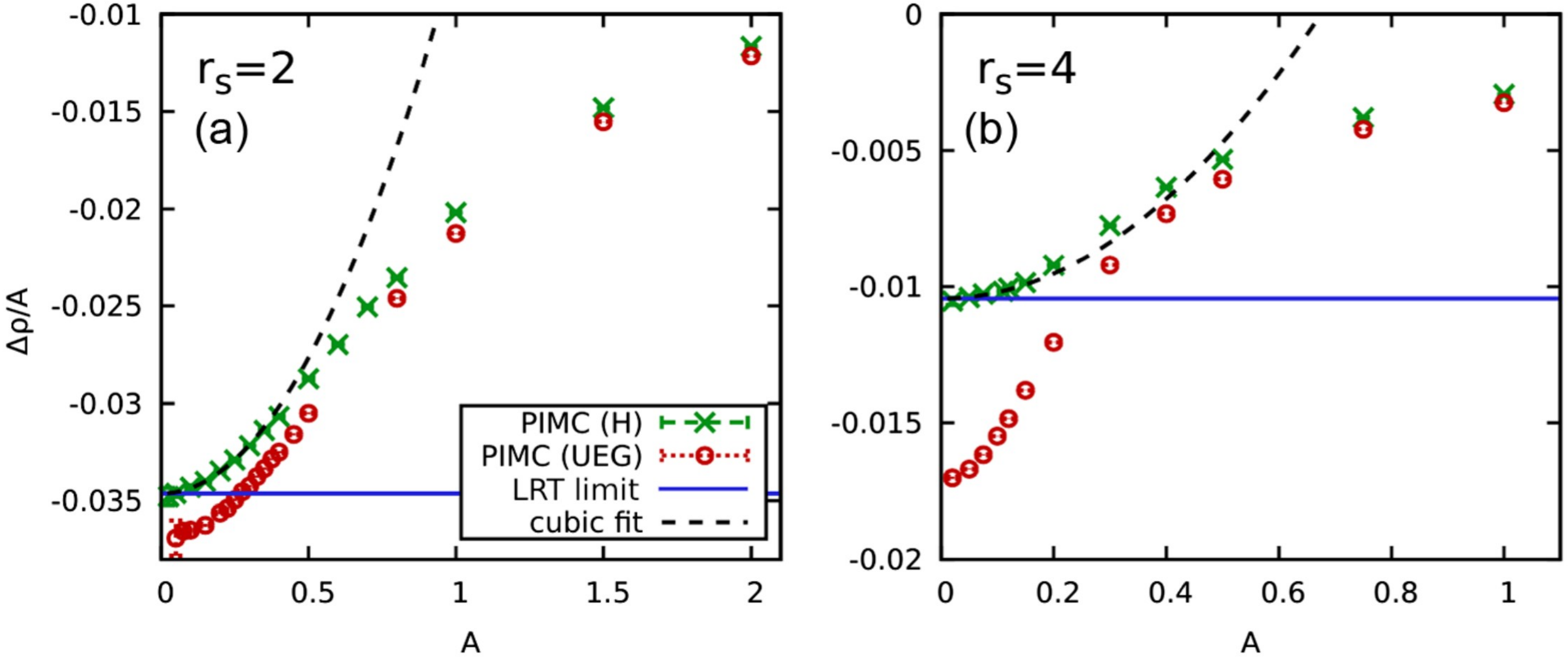
- Harmonically perturbed Hamiltonian:

$$\hat{H}_{\mathbf{q},A} = \hat{H} + 2A\sum_{l=1}^{N}\cos\left(\mathbf{q}\cdot\hat{\mathbf{r}}_l\right)$$

→ direct access to linear+nonlinear density response

$$\langle\hat{\rho}_{\mathbf{q}}\rangle_{q,A} = \chi^{(1)}(q)A + \chi^{(1,\mathrm{cubic})}(q)A^3$$

Electronic density response of H at θ=1



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$$G(q) = -\frac{4\pi}{q^2}K_{\text{xc}}(q)$$

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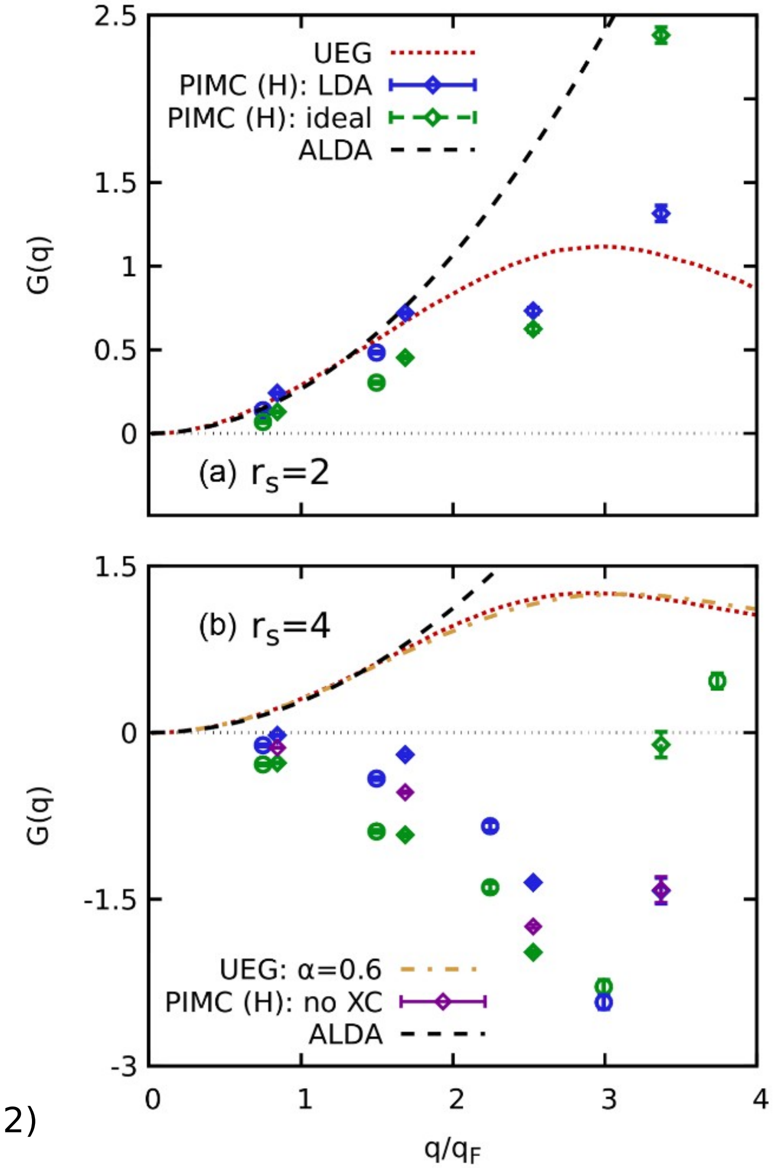

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Common UEG based XC models break down at low rs!

- Invert density response to get XC kernel / LFC

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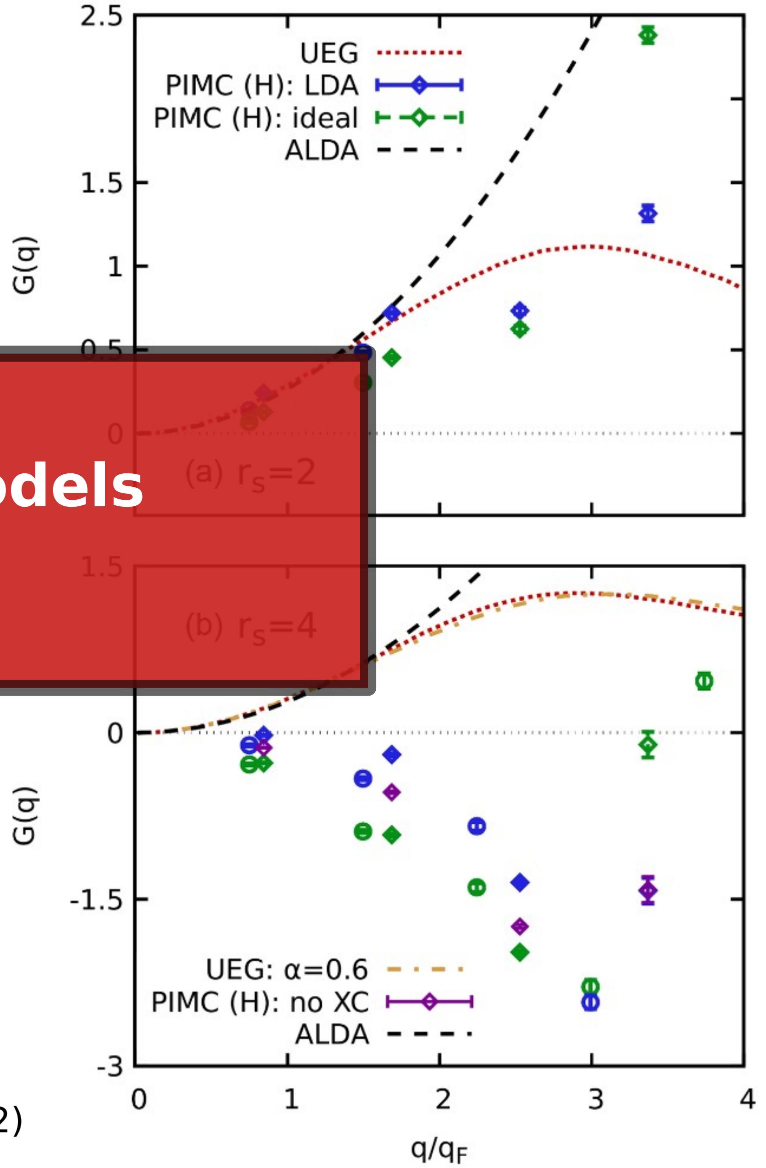

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Part II: Electronic density response of WDM

XC kernel from DFT without functional derivatives

Zh. Moldabekov (PostDoc)

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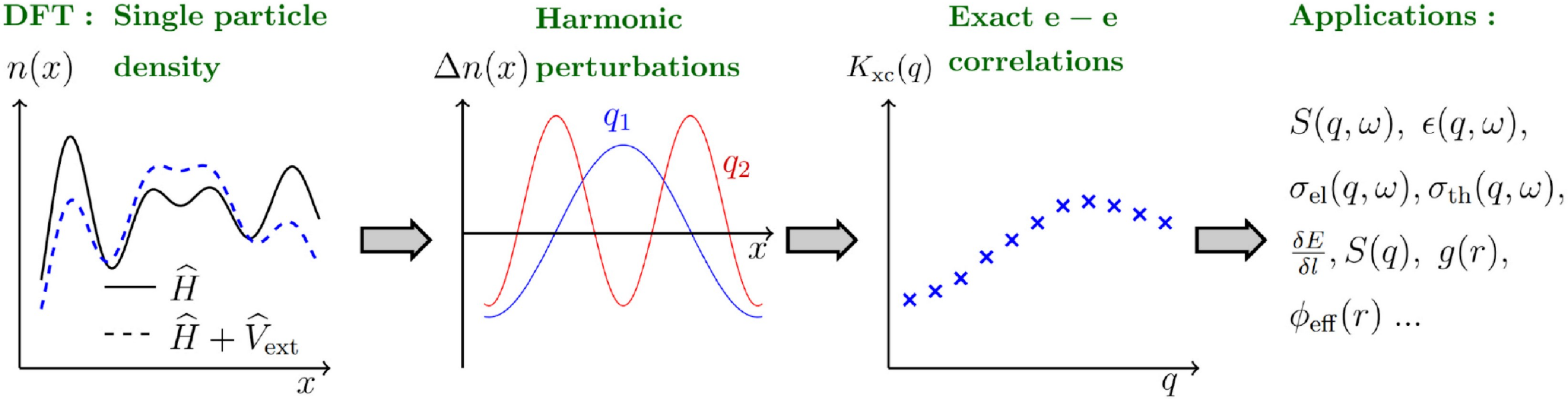
Taken from: Zh. Moldabekov, M. Böhme, J. Vorberger, D. Blaschke and **T. Dornheim**, JCTC **19**, 1286-1299 (2023)



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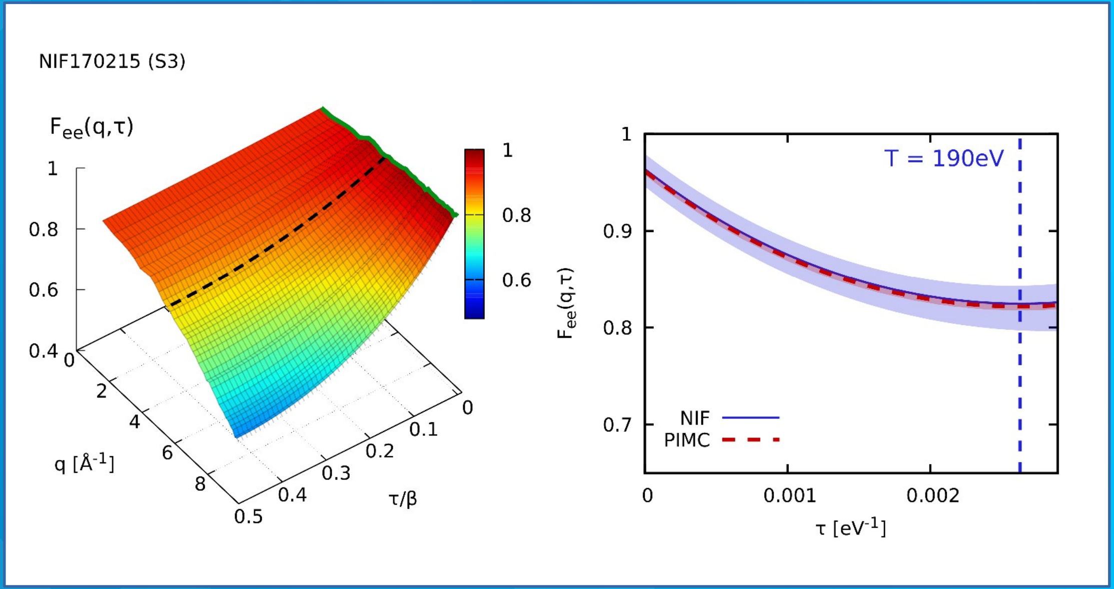
Part III: Imaginary-time correlation functions and XRTS



Taken from: **T. Dornheim**, Zh. Moldabekov, M. Böhme, J. Vorberger, P. Tolias, F. Graziani, and T. Döppner (in preparation)

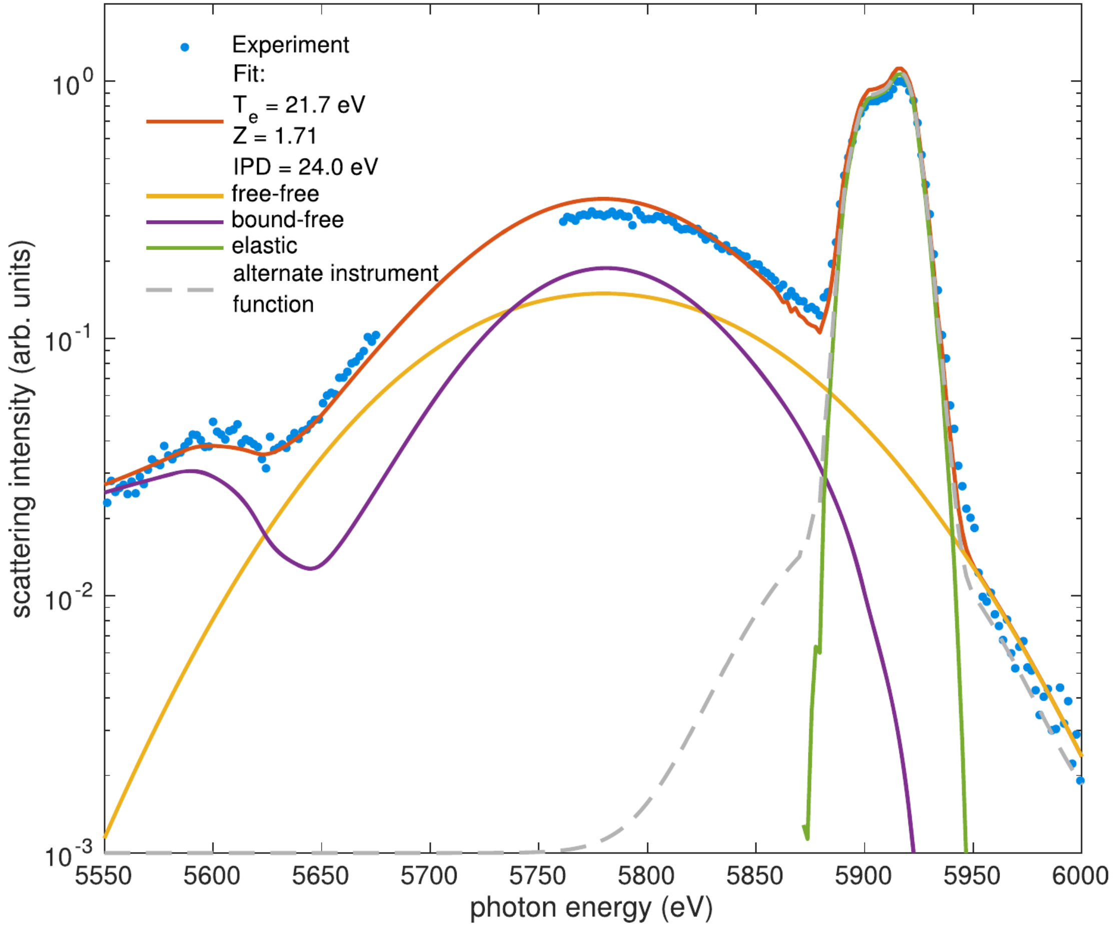
Part III: Imaginary-time correlation functions + XRTS

X-ray Thomson scattering (XRTS)

- Standard way: construct a model for S(q,ω), convolve with instrument function R(ω), fit to XRTS signal I(q,ω)

$$I(\mathbf{q},\omega) = S(\mathbf{q},\omega) \circledast R(\omega)$$

Isochorically heated graphite at LCLS (Stanford)



Taken from: D. Kraus *et al.*, *Plasma Phys. Control. Fusion* **61**, 014015 (2019)

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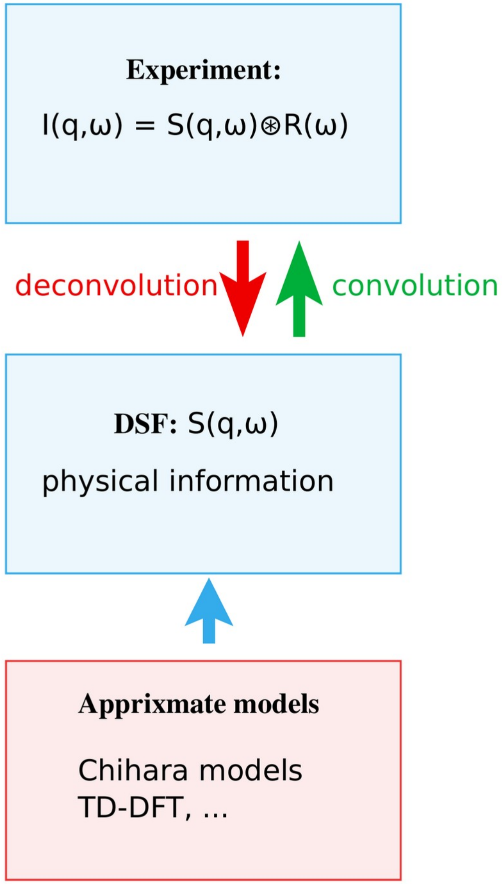
- **Frequency domain:**

→ no direct access to physical information

→ approximate theoretical models



Taken from: **T. Dornheim**, Zh. Moldabekov, P. Tolias, M. Böhme, and J. Vorberger, Matt. Rad. Extremes **8**, 056601 (2023)

Part III: Imaginary-time correlation functions + XRTS

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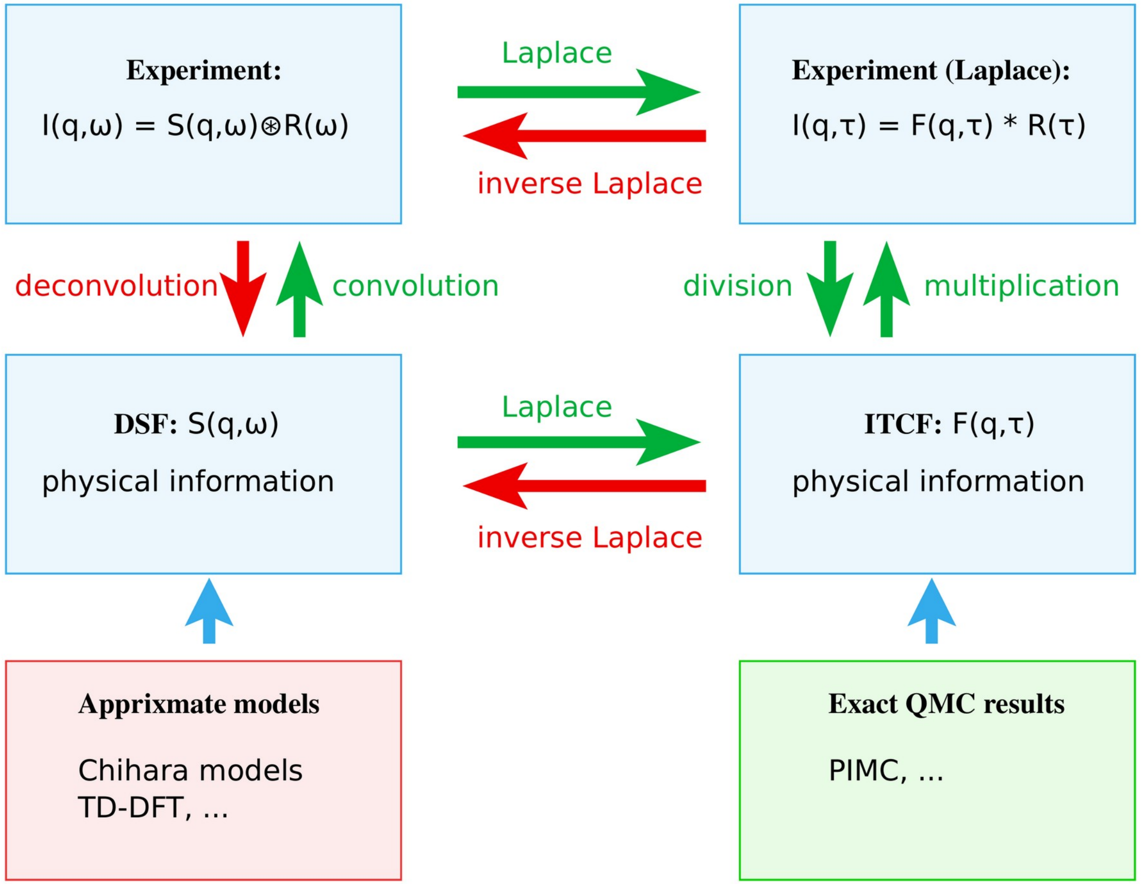
→ direct access to physics, e.g. T, ω_p

→ exact QMC simulations

$$\mathcal{L}\left[S(\mathbf{q},\omega)\right] = \int_{-\infty}^{\infty} \mathrm{d}\omega\ e^{-\tau\omega}\ S(\mathbf{q},\omega)$$



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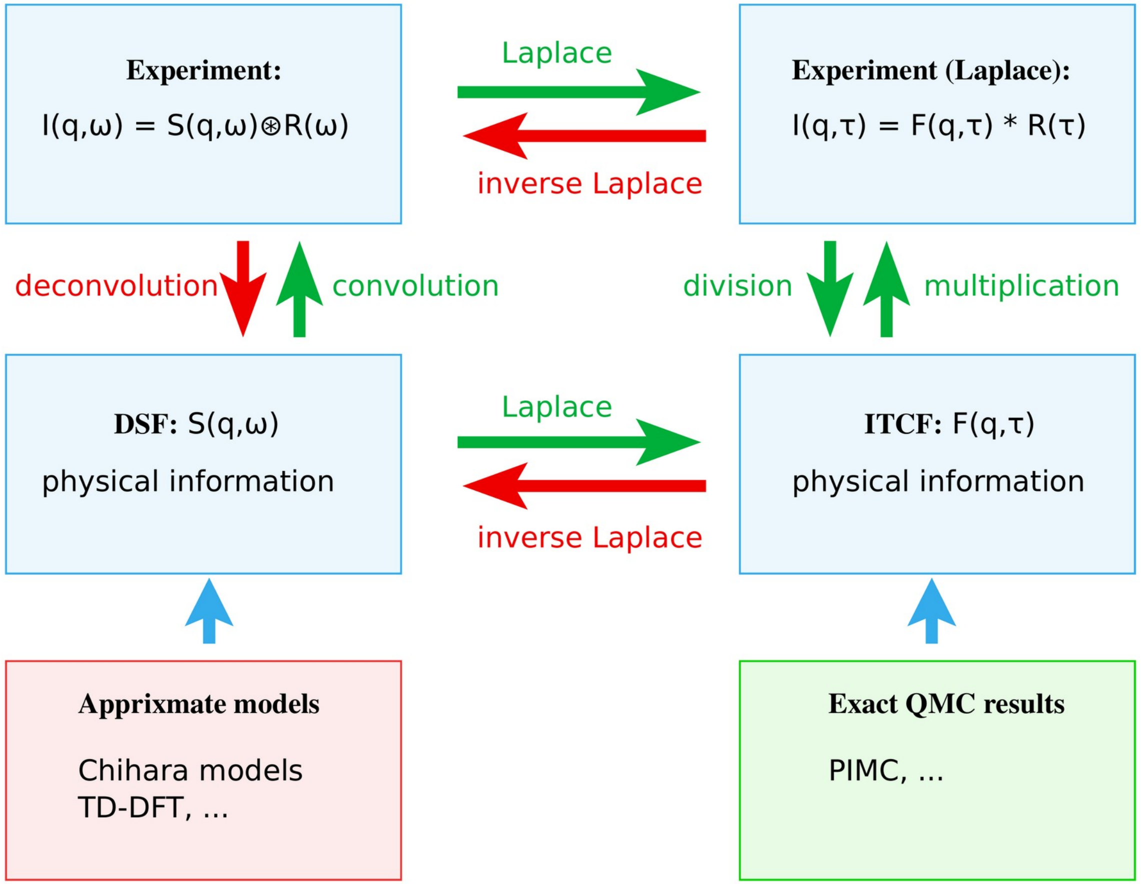

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Part III: Imaginary-time correlation functions + XRTS

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Application I: Model-free temperature diagnostics

- **Detailed balance in the τ-domain:**

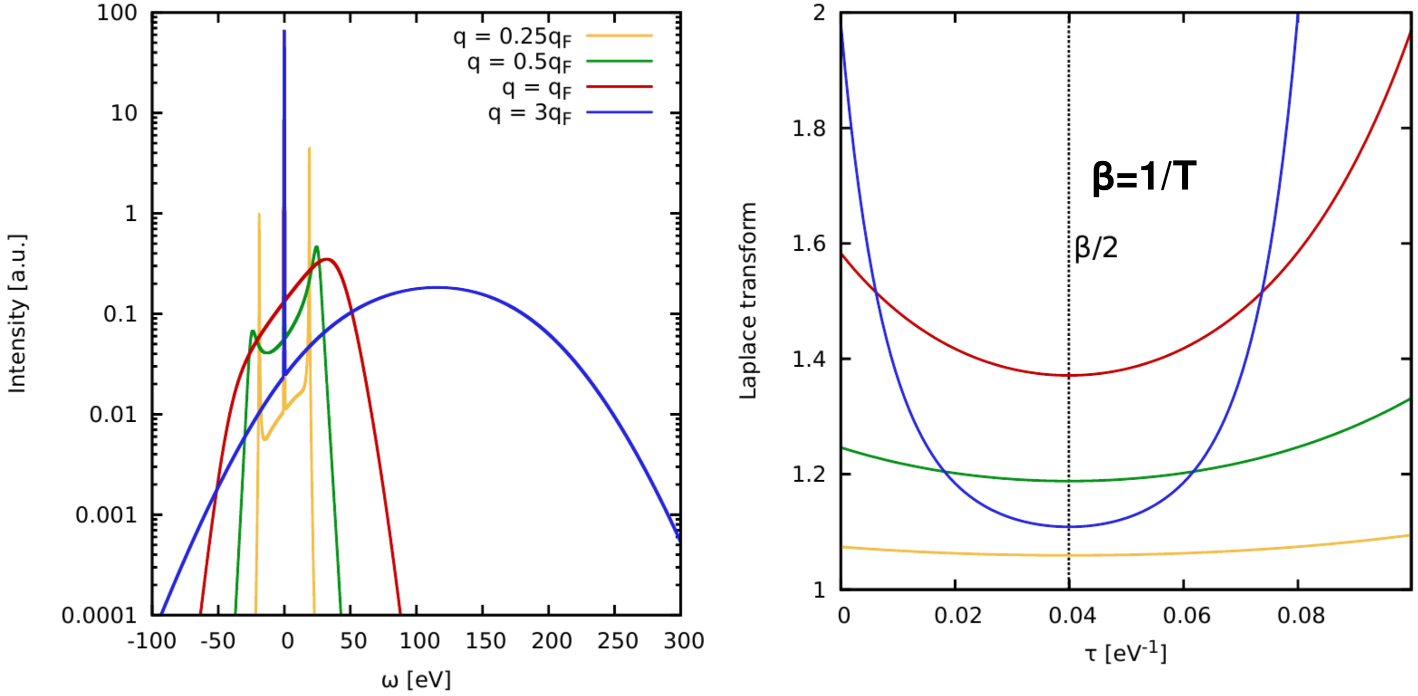
→ works for all wave numbers

→ no explicit resolution of plasmon required

$$S(\mathbf{q},-\omega) = S(\mathbf{q},\omega)e^{-\beta\omega}$$

→ symmetry around τ=(2T)$^{-1}$

Left: S(q,ω)   **Right:** Laplace transform



Taken from: **T. Dornheim** et al, Phys. Plasmas **30**, 042707 (2023)

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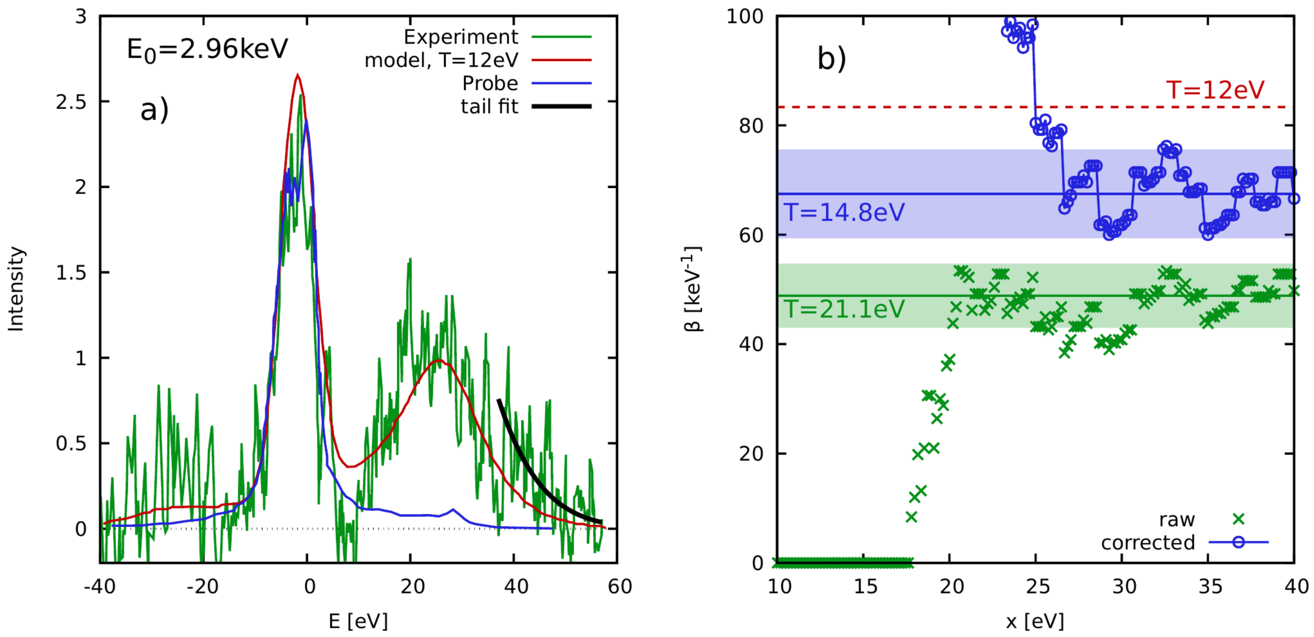
→ very robust with respect to noise

→ no models / simulations etc.

Temperature of warm dense Be [Glenzer (2007)]



Taken from: **T. Dornheim**, M. Böhme, D. Kraus, T. Döppner, T. Preston, Zh. Moldabekov, and J. Vorberger, Nature Comm. **13**, 7911 (2022)

Part III: Imaginary-time correlation functions + PIMC

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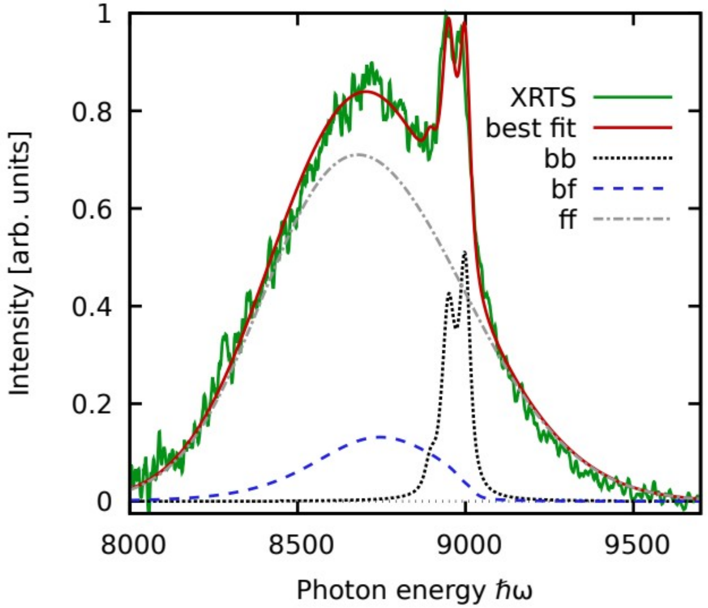
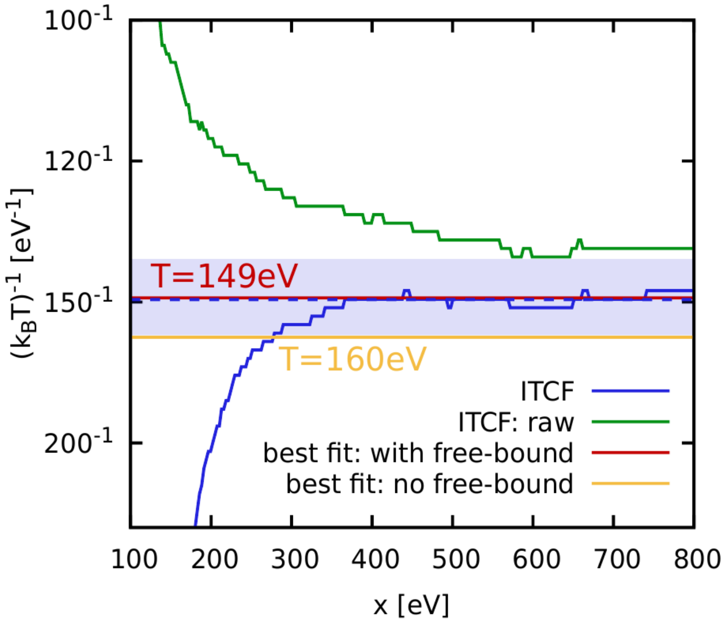
Temperature of strongly compressed Be@NIF [Döppner et al., (2023)]





Taken from: M. Böhme, L. Fletcher, T. Döppner, D. Kraus, A. Baczewski, Th. Preston, …, and **T. Dornheim**, arXiv:2306.17653 (submitted)

Part III: Imaginary-time correlation functions + PIMC

Model-free normalization of XRTS experiments:

- **Measured XRTS intensity is given by**

$$I(\mathbf{q},\omega) = A\, S_{ee}(\mathbf{q},\omega) \circledast R(\omega)$$

(with A being an a-priori unknown normalization factor)

Reference: **T. Dornheim**, T. Döppner, A. Baczewski, P. Tolias, M. Böhme, Zh. Moldabekov, et al., arXiv:2305.15305 (submitted)

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→ Laplace transform gives **unnormalized** ITCF

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- **Absolute knowledge of ITCF is required for practical applications, e.g.**

$$\chi(\mathbf{q},0) = -n\int_0^\beta \mathrm{d}\tau\ F_{ee}(\mathbf{q},\tau)$$

and

$$S_{ee}(\mathbf{q}) = F_{ee}(\mathbf{q},0)$$

Reference: **T. Dornheim**, T. Döppner, A. Baczewski, P. Tolias, M. Böhme, Zh. Moldabekov, et al., arXiv:2305.15305 (submitted)

Part III: Imaginary-time correlation functions + PIMC

Model-free normalization of XRTS experiments:

- **Solution: f-sum rule in the imaginary time** $M_1^S = \hbar q^2/2m_e$

$$M_\alpha^S = \int_{-\infty}^{\infty} \mathrm{d}\omega \ S_{ee}(\mathbf{q},\omega)\ \omega^\alpha$$

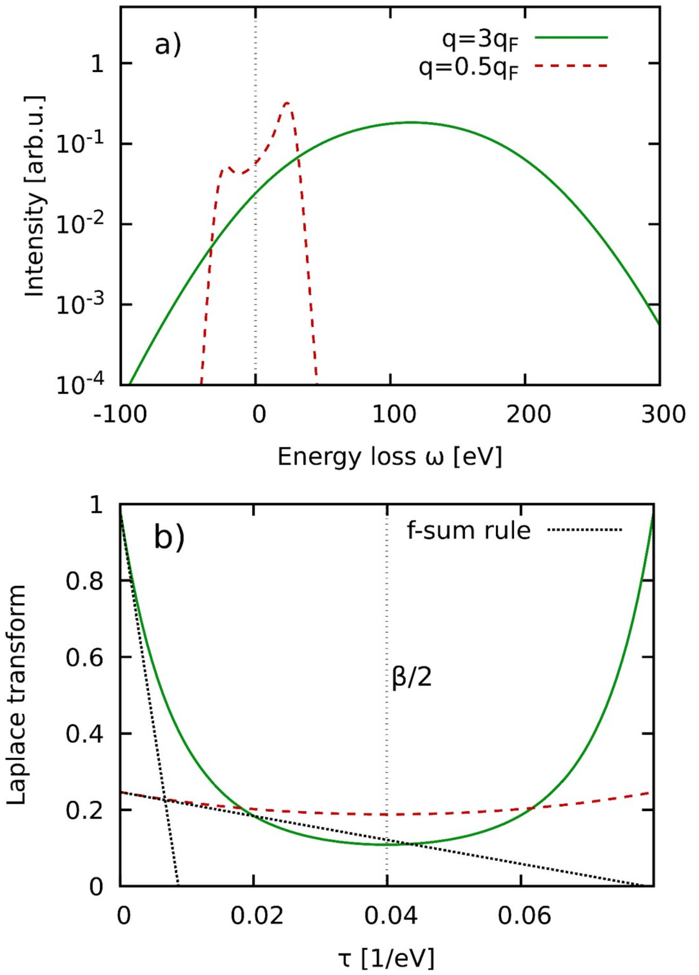
$$M_\alpha^S = \frac{(-1)^\alpha}{\hbar^\alpha} \left. \frac{\partial^\alpha}{\partial\tau^\alpha} F_{ee}(\mathbf{q},\tau) \right|_{\tau=0}$$

→ frequency moments of S(q,ω) are given by derivatives

Of the ITCF around τ=0

- **Determine the normalization from the first derivative of the ITCF**

Example: **uniform electron gas**



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Of the ITCF around τ=0

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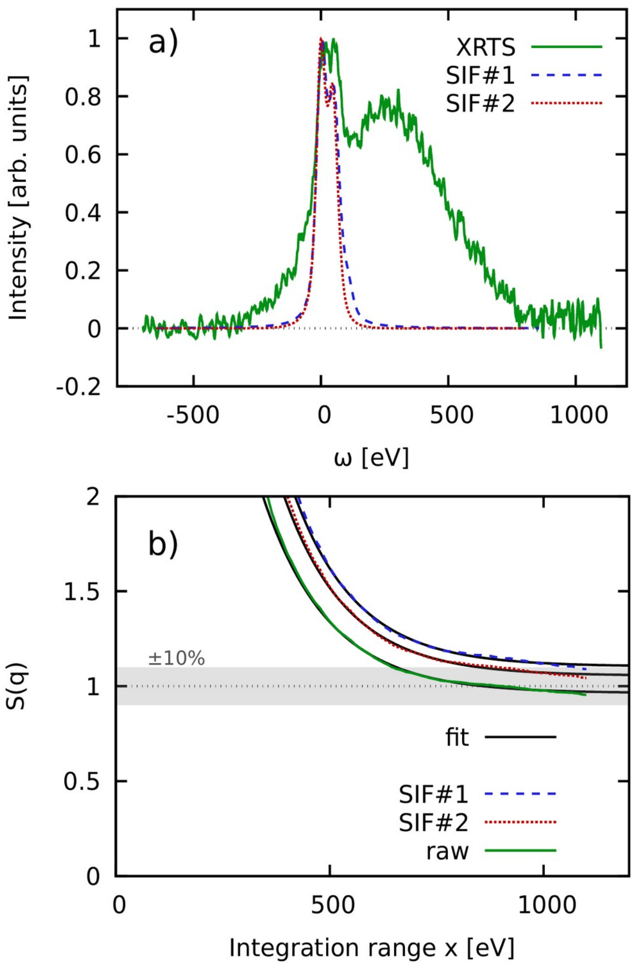
<u>Example:</u> Be@NIF (Döppner et al)



<u>Reference:</u> **T. Dornheim**, T. Döppner, A. Baczewski, P. Tolias, M. Böhme, Zh. Moldabekov, et al., arXiv:2305.15305 (submitted)

Part III: Imaginary-time correlation functions + PIMC

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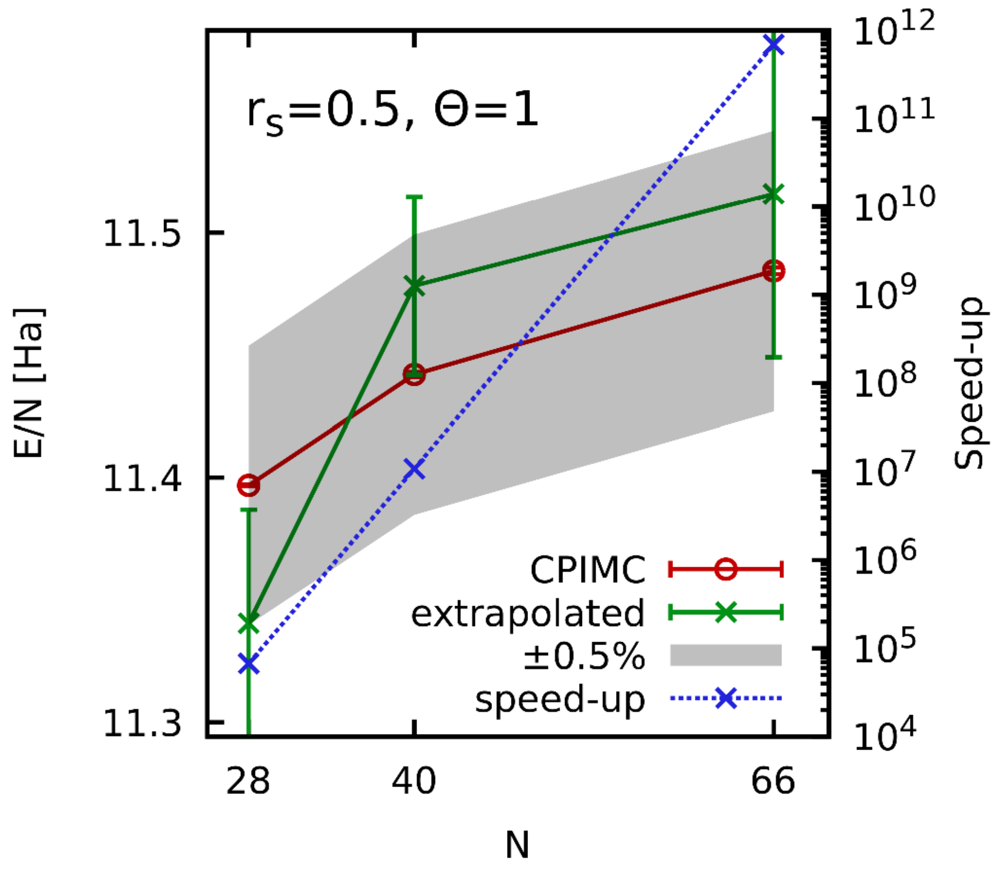
Work in progress: large PIMC simulations with exponential speed-up



Taken from: **T. Dornheim**, P. Tolias, S. Groth, Zh. Moldabekov, J. Vorberger, and B. Hirshberg, J. Chem. Phys. **159**, 164113 (2023)

→ Can do: large systems at moderate to weak quantum degeneracy

→ Can‘t do: low temperatures, strongly quantum degenerate regime

Perfectly suited for low-Z materials at the NIF!

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Part III: Imaginary-time correlation functions + PIMC

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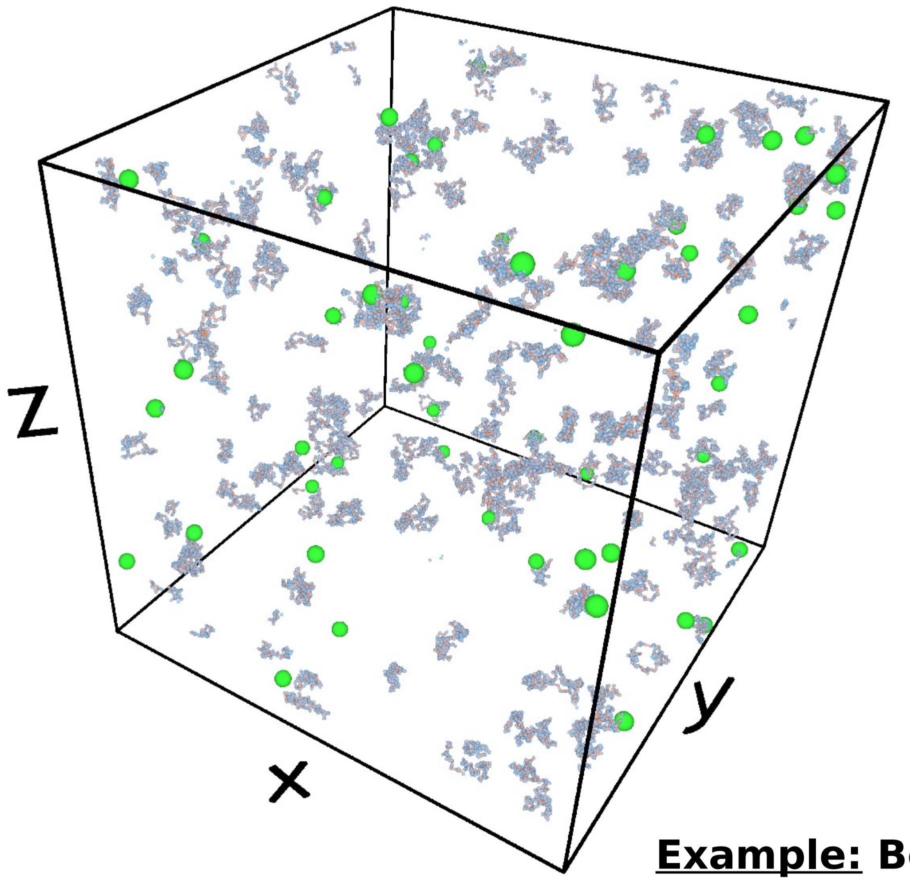

Example: Be at T=155eV

→ **Can do:** large systems at moderate to weak quantum degeneracy

→ **Can't do:** low temperatures, strongly quantum degenerate regime

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Reference: **T. Dornheim**, Zh. Moldabekov, M. Böhme, J. Vorberger, P. Tolias, D. Kraus, F. Graziani, and T. Döppner, in preparation

Part III: Imaginary-time correlation functions + PIMC

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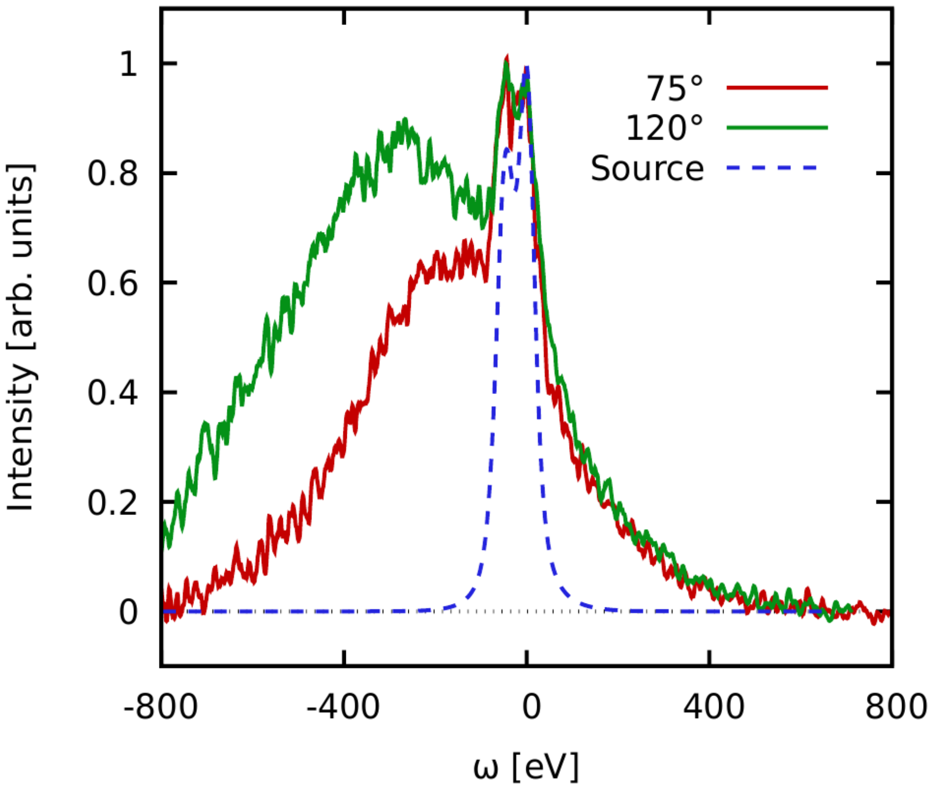

Example: Strongly compressed Be at NIF

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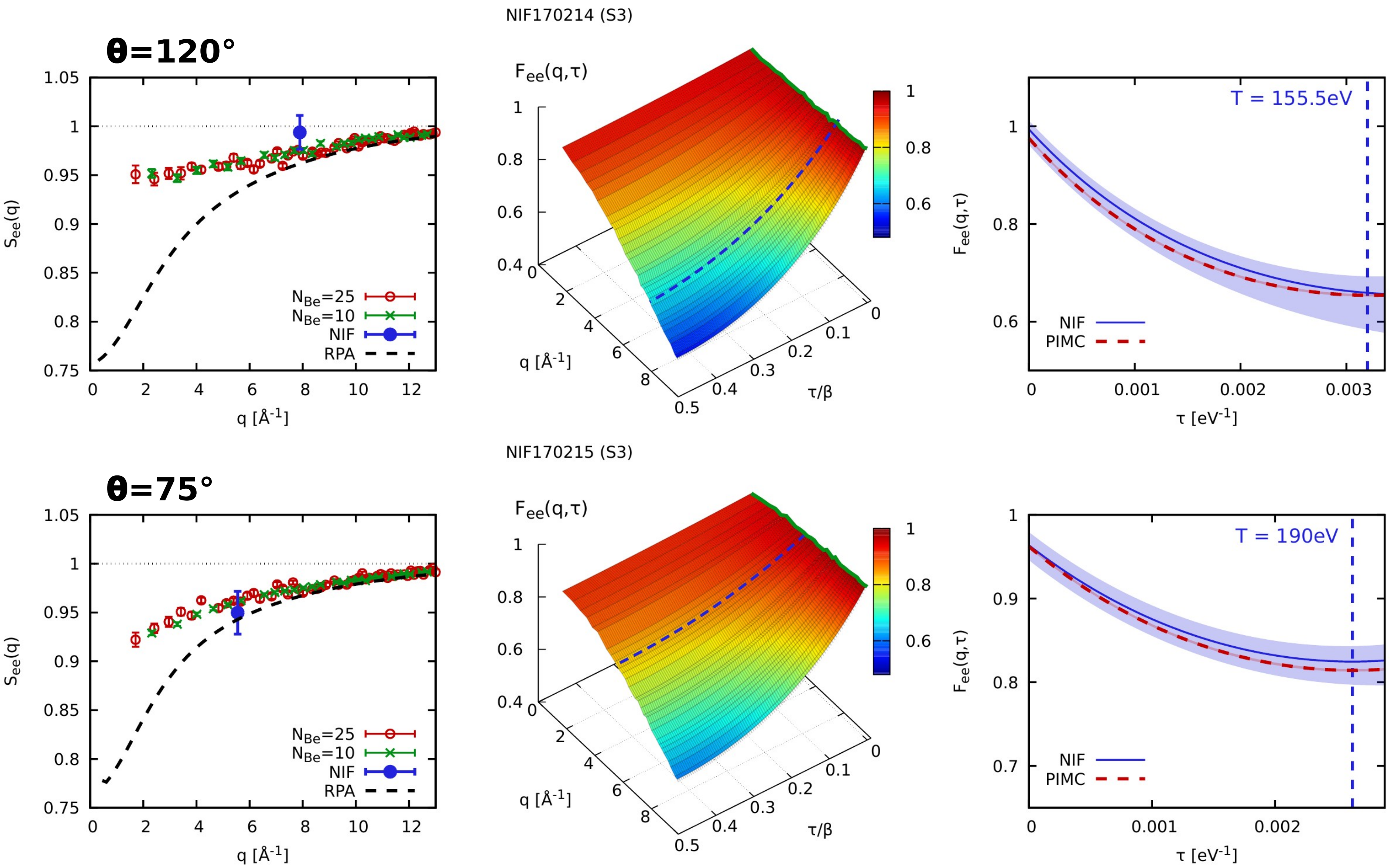


→ **exact PIMC results for NIF conditions**

→ **study e-e correlations (not possible with DFT)**

→ **all spectral information in the ITCF**

→ **predict experiments, guide developments**

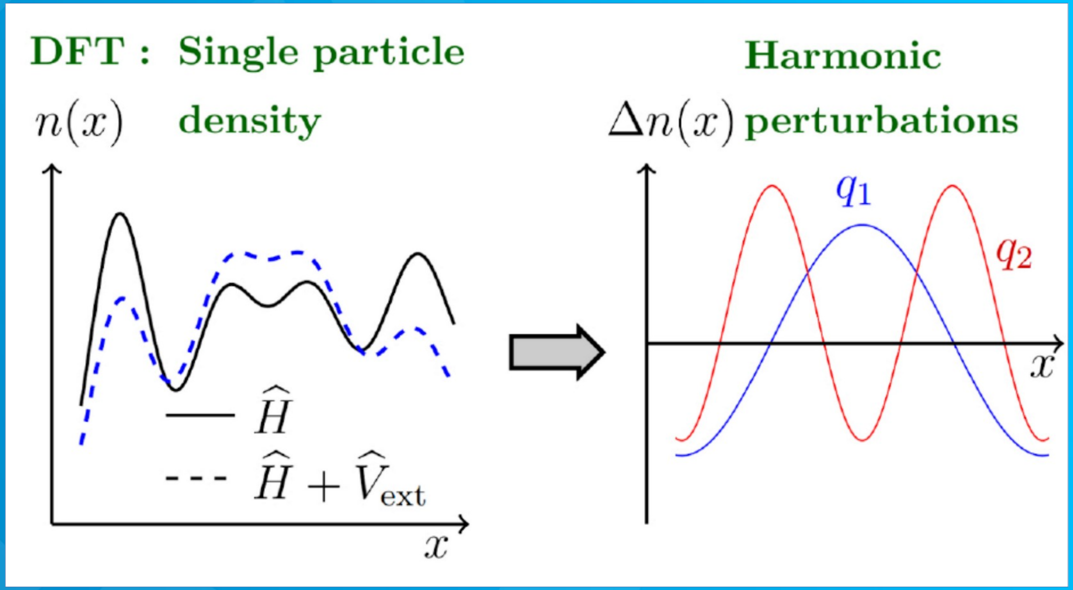
→ **benchmark for approximate methods etc**

→ **H, He, Li, LiH, Be, … ?**

Reference: **T. Dornheim**, Zh. Moldabekov, M. Böhme, J. Vorberger, P. Tolias, D. Kraus, F. Graziani, and T. Döppner (in preparation)

Summary and Outlook

- PIMC as a starting point to understand e-e correlations in WDM
- First exact PIMC results for XC-kernel of H
- XC-kernels within the framework of DFT



Taken from: Zh. Moldabekov, M. Böhme, J. Vorberger, D. Blaschke, and **T. Dornheim**, J. Chem. Theory Comput. **19**, 1286-1299 (2023)

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- PIMC as a starting point to understand e-e correlations in WDM
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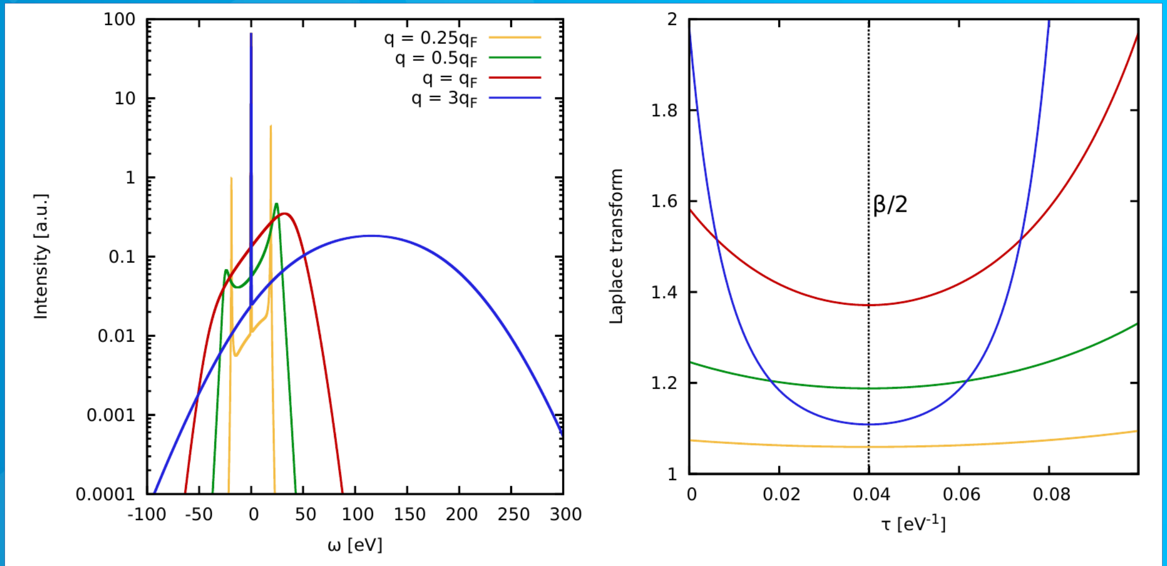
→ XRTS measurements: T, S(q), etc.



Taken from: **T. Dornheim**, M. Böhme, D. Chapman, D. Kraus, Th. Preston, Zh. Moldabekov, ..., and J. Vorberger, Phys. Plasmas **30**, 042707 (2023)

Summary and Outlook

- PIMC as a starting point to understand e-e correlations in WDM
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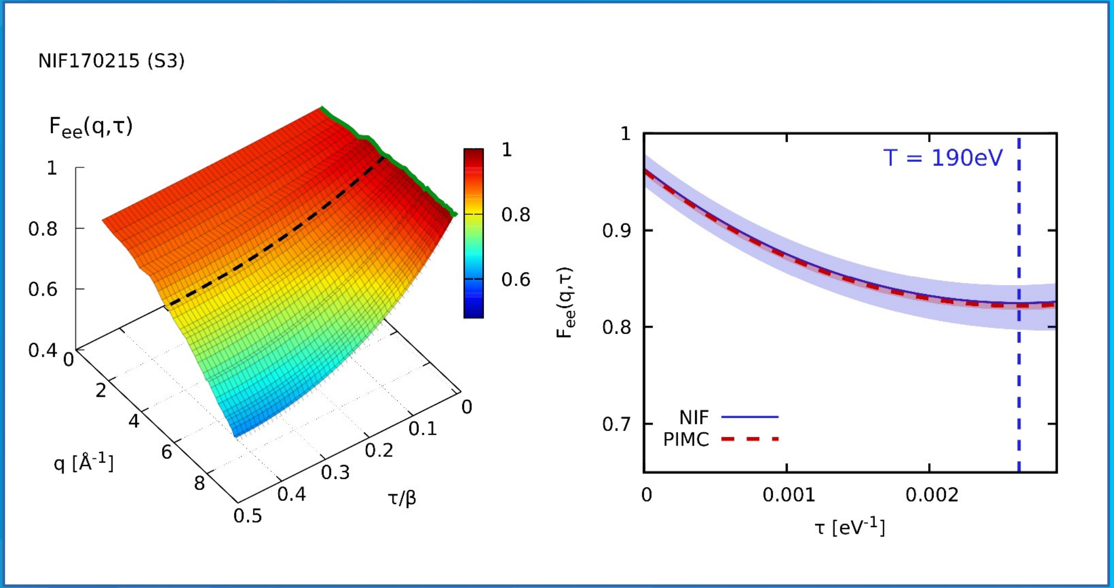
- Imaginary-time correlation functions as a framework to understand e-e correlations

→ XRTS measurements: T, S(q), etc.

→ *Ab initio* PIMC simulations

→ New experimental set-ups, ...

Thank you for your attention!



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Part III: Imaginary-time correlation functions + XRTS

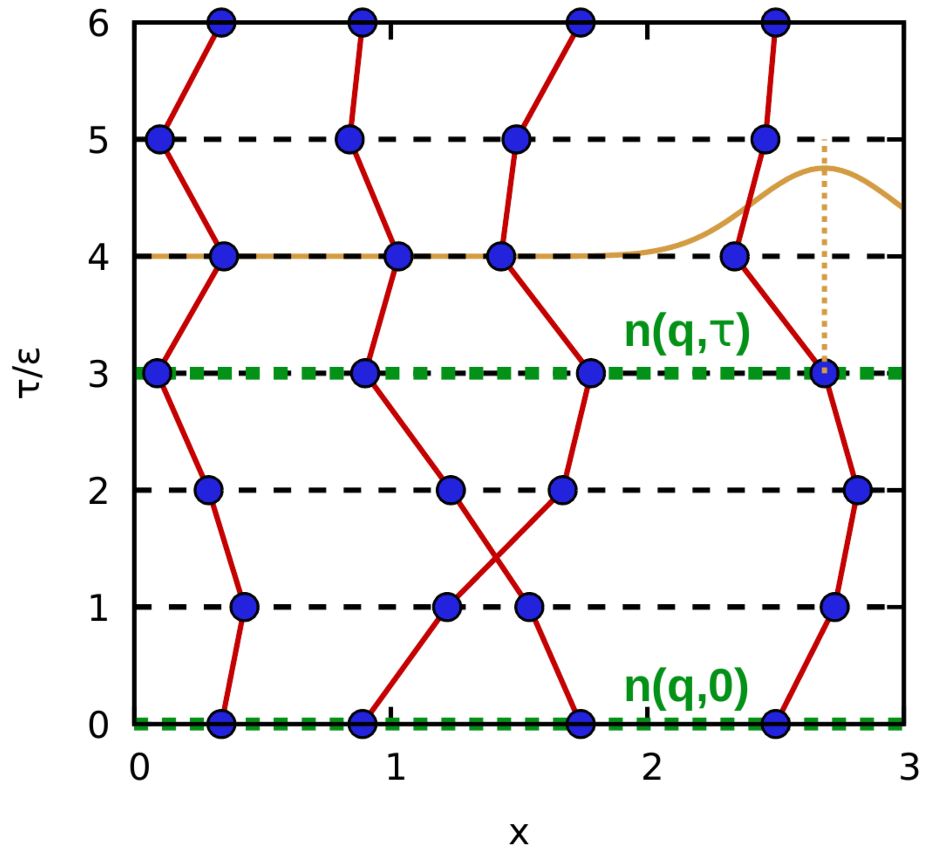
Imaginary-time correlation functions

- Density—density correlations:

$$F(\mathbf{q},\tau) = \langle \hat{n}(\mathbf{q},0)\hat{n}(-\mathbf{q},\tau)\rangle$$

→ measures stability / decay of correlations along τ

Imaginary-time path integral configuration



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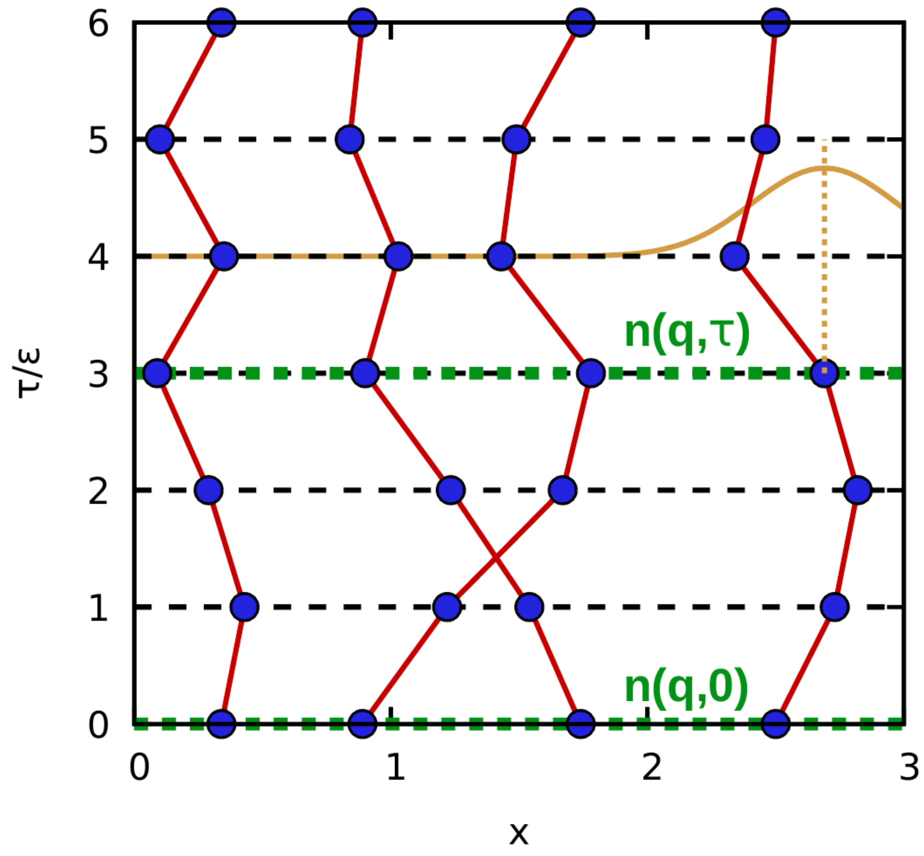

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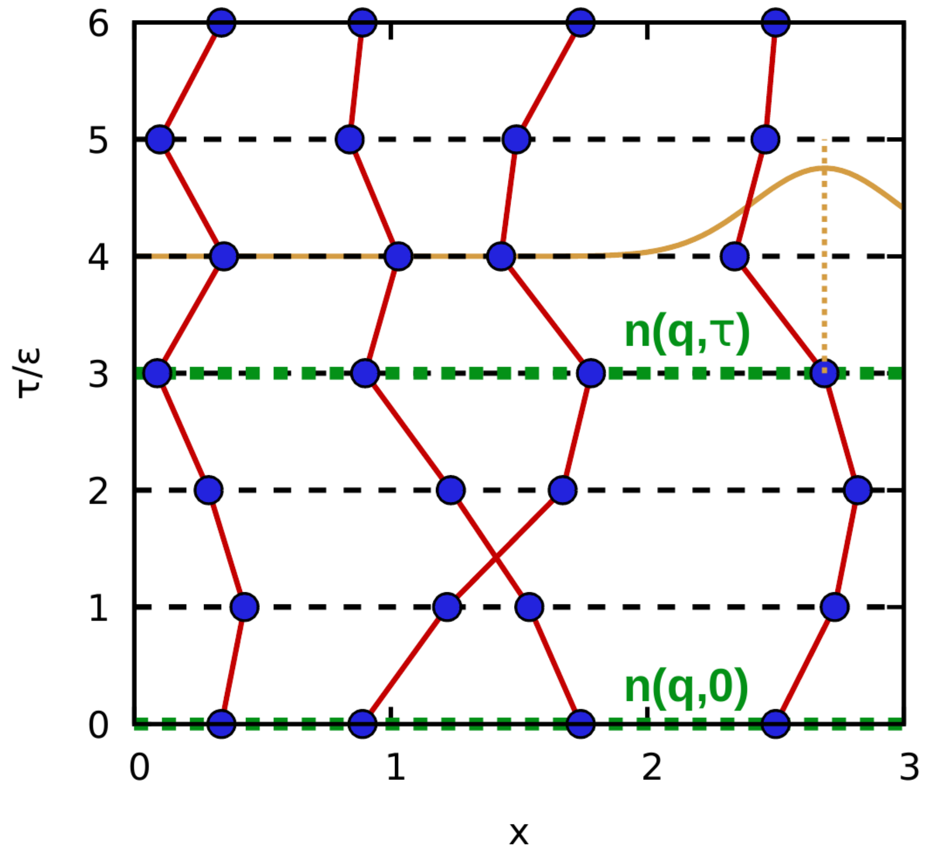
- Spectral representation:

$$S(\mathbf{q},\omega) = \sum_{m,l} P_m \left\|n_{ml}(\mathbf{q})\right\|^2 \delta(\omega - \omega_{lm})$$

$$F(\mathbf{q},\tau) = \sum_{m,l} P_m \left\|n_{ml}(\mathbf{q})\right\|^2 e^{-\tau\omega_{lm}}$$

Imaginary-time path integral configuration



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Appendix

Imaginary-time correlation functions

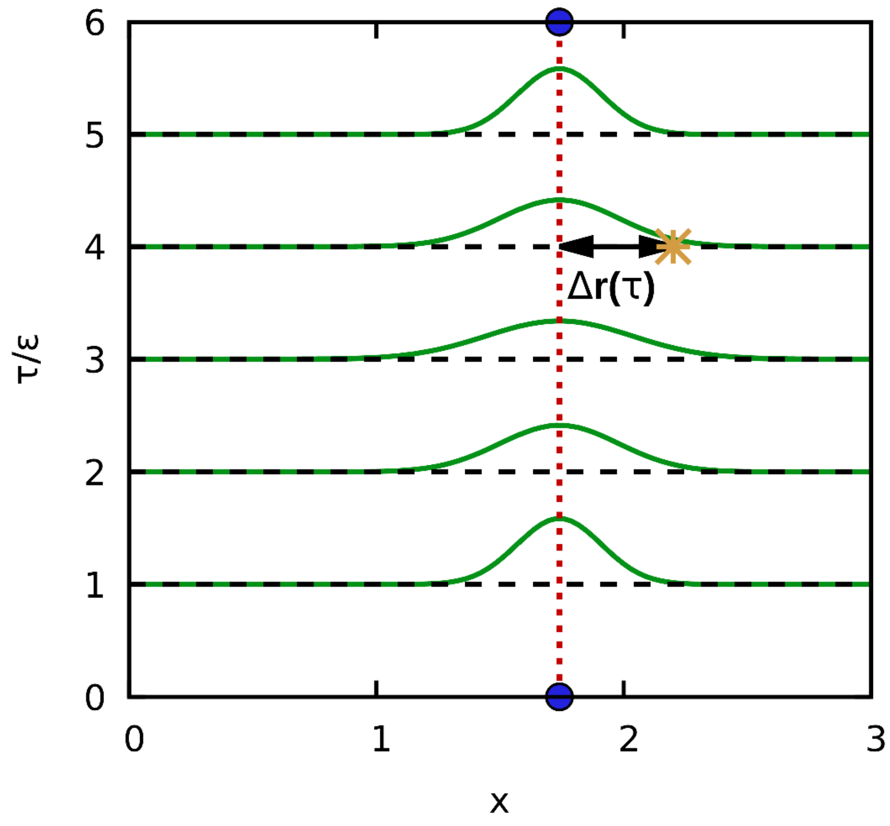
- Gaussian diffusion process:

$$\rho_0(\mathbf{r},\mathbf{r};\beta) = \int_\Omega \mathrm{d}\mathbf{r}' \ \rho_0(\mathbf{r},\mathbf{r}';\tau')\rho_0(\mathbf{r}',\mathbf{r};\beta-\tau')$$

→ Decay of elecronic correlations along τ:

$$F_{\mathrm{SP}}(\mathbf{q},\tau') = \int_\Omega \mathrm{d}\mathbf{\Delta r} \ P(\mathbf{\Delta r},\tau') \ \cos\left(\mathbf{q}\cdot\mathbf{\Delta r}\right)$$

Example: Imaginary-time diffusion of a single electron



Taken from: **T. Dornheim**, J. Vorberger, Zh. Moldabekov and M. Böhme , Phil. Trans. Royal. Soc. (in print), arXiv: 2211.00579

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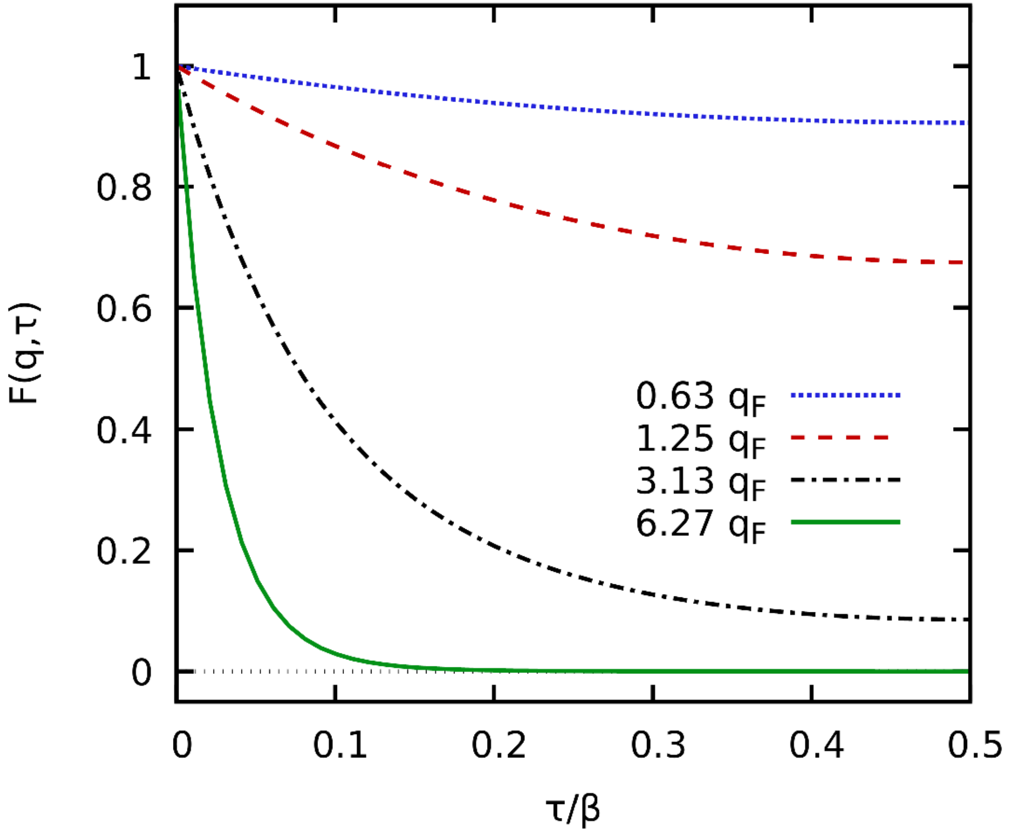
→ Increasing τ-decay with q accurately follows from single-particle model

Single-electron ITCF exhibits correct τ-dependence



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Excursion: Nonlinear electronic density response of WDM

Direct perturbation method gives access to nonlinear effects

- Harmonically perturbed electron gas

$$\hat{H} = \hat{H}_{\mathrm{UEG}} + 2A\sum_{k=1}^{N}\cos\left(\hat{\mathbf{r}}_k\cdot\mathbf{q}\right)$$

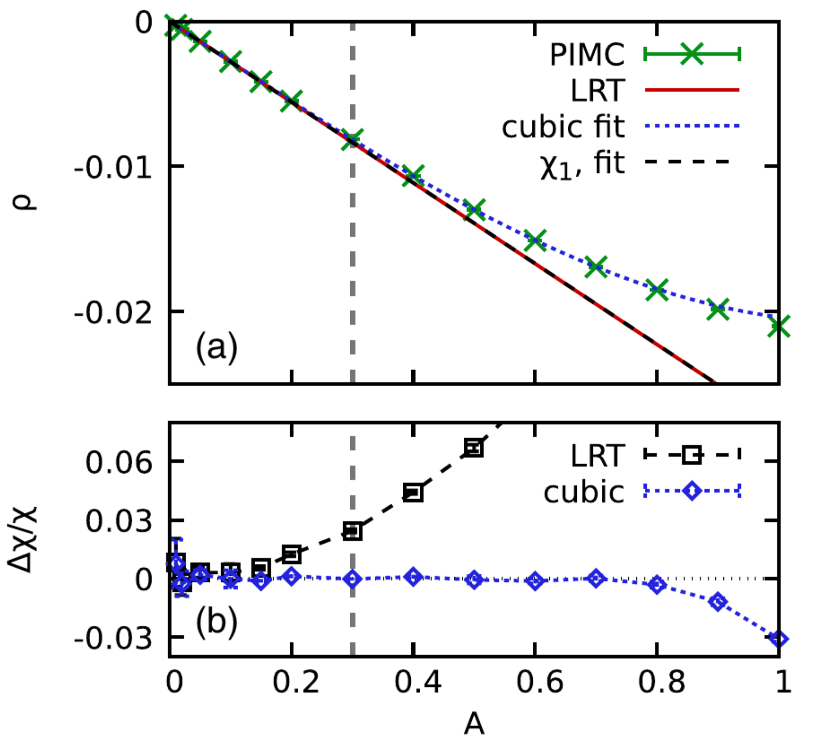
→ Expand nonlinear density response in powers of A

$$\langle\hat{\rho}_{\mathbf{q}}\rangle_{q,A} = \chi^{(1)}(q)A + \chi^{(1,\mathrm{cubic})}(q)A^3,$$

$$\langle\hat{\rho}_{2\mathbf{q}}\rangle_{q,A} = \chi^{(2)}(q)A^2,$$

$$\langle\hat{\rho}_{3\mathbf{q}}\rangle_{q,A} = \chi^{(3)}(q)A^3,$$

Density response at the first harmonic



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

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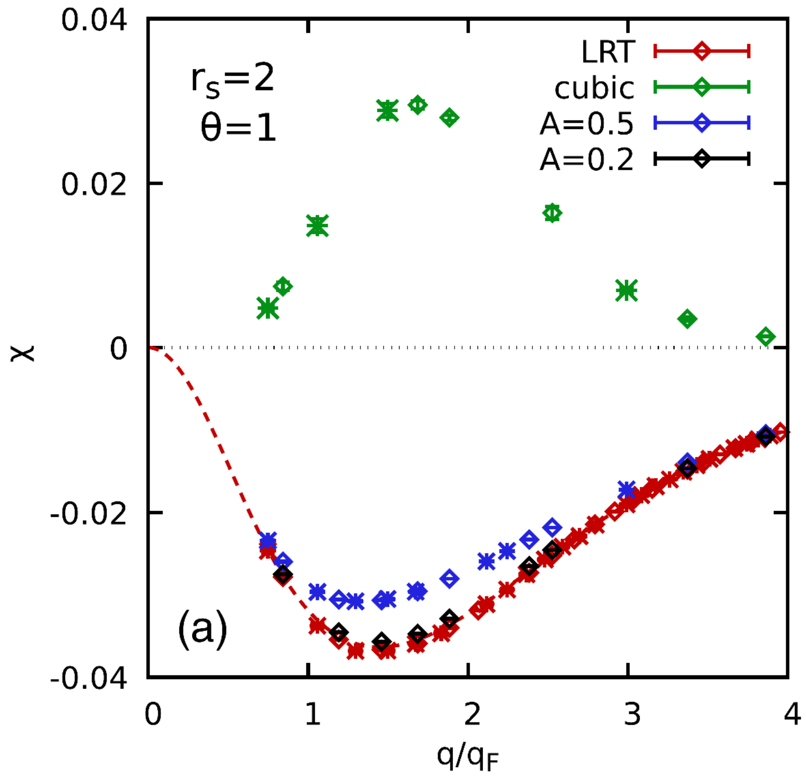

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$$\hat{H} = \hat{H}_{\text{UEG}} + 2A\sum_{k=1}^{N}\cos\left(\hat{\mathbf{r}}_k \cdot \mathbf{q}\right)$$

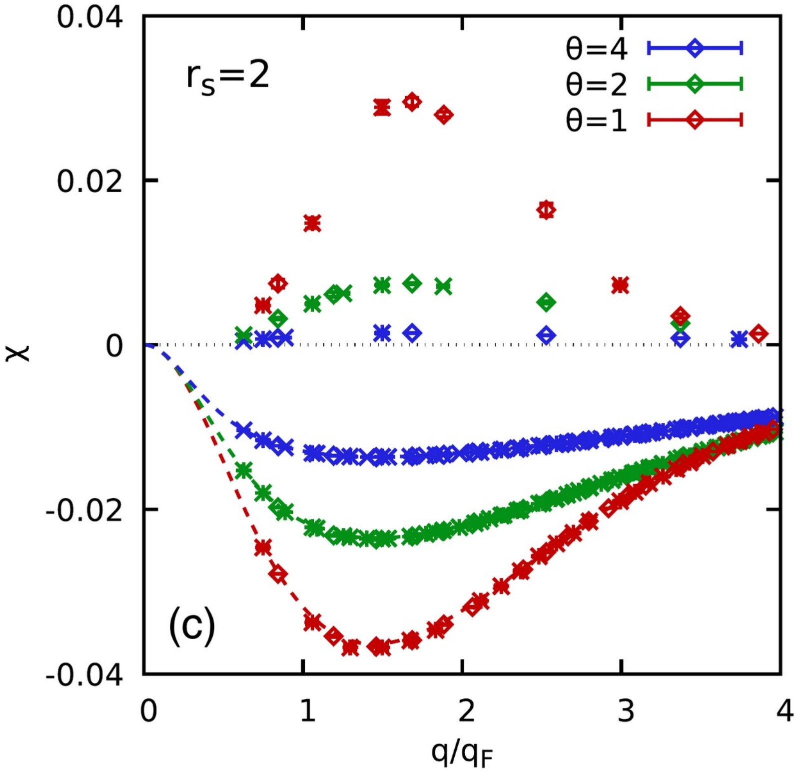
→ Expand nonlinear density response in powers of A

$$\langle\hat{\rho}_{\mathbf{q}}\rangle_{q,A} = \chi^{(1)}(q)A + \chi^{(1,\text{cubic})}(q)A^3,$$

$$\langle\hat{\rho}_{2\mathbf{q}}\rangle_{q,A} = \chi^{(2)}(q)A^2,$$

$$\langle\hat{\rho}_{3\mathbf{q}}\rangle_{q,A} = \chi^{(3)}(q)A^3,$$

Density response at the first harmonic



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Excursion: Nonlinear electronic density response of WDM

CASUS
CENTER FOR ADVANCED SYSTEMS UNDERSTANDING

Direct perturbation method gives access to nonlinear effects

- Harmonically pert

$$\hat{H} = \hat{H}_{\text{UEG}} +$$

→ Expand nonlinear

$$\langle \hat{\rho}_{\mathbf{q}} \rangle_{q,A} = \chi^{(1)}$$

$$\langle \hat{\rho}_{\mathbf{2q}} \rangle_{q,}$$

$$\langle \hat{\rho}_{\mathbf{3q}} \rangle_{q,}$$

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Electronic density response of warm dense matter

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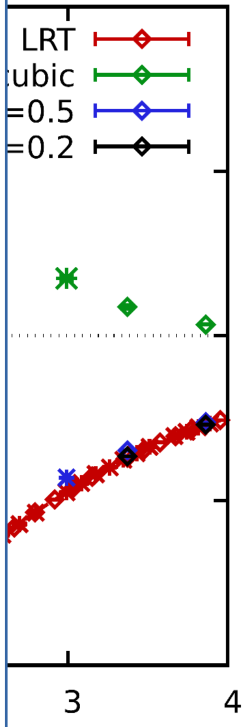
rst harmonic



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Various aspects of nonlinear density response theory

- Get nonlinear density response from higher-order imaginary-time correlation functions

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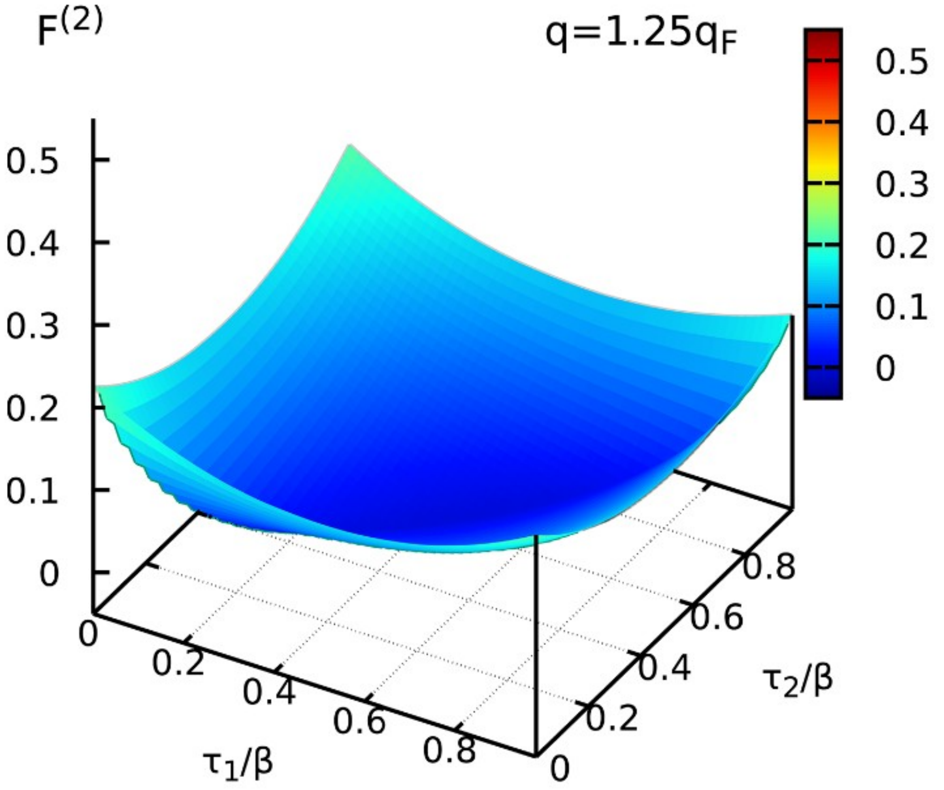

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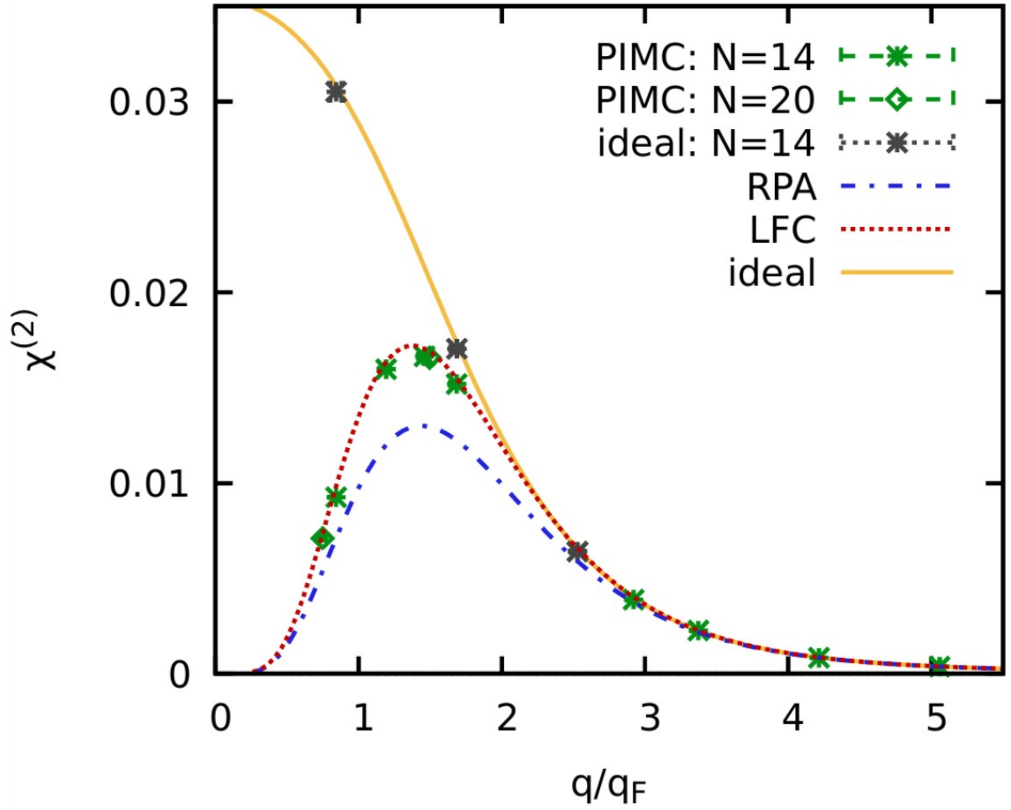

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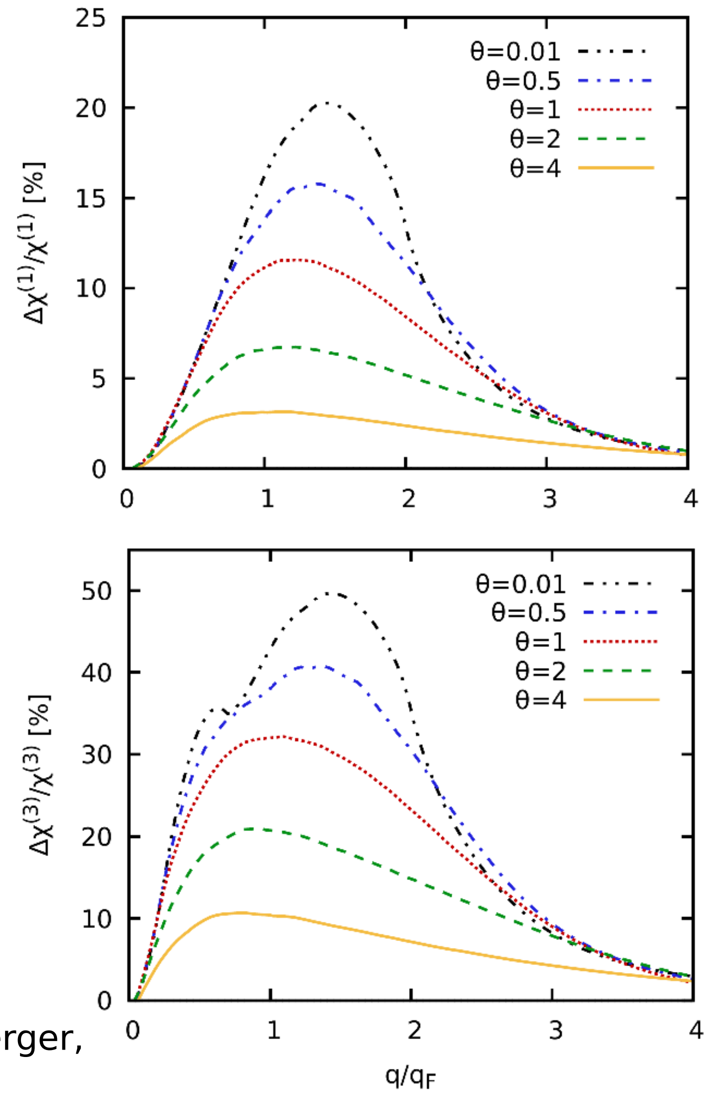

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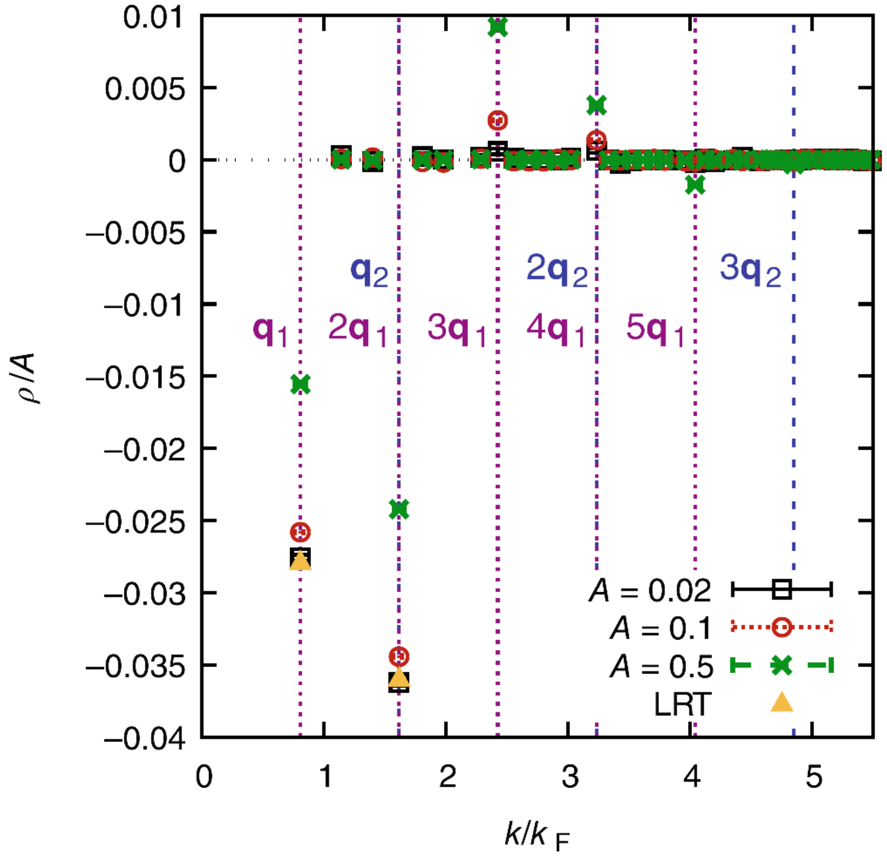

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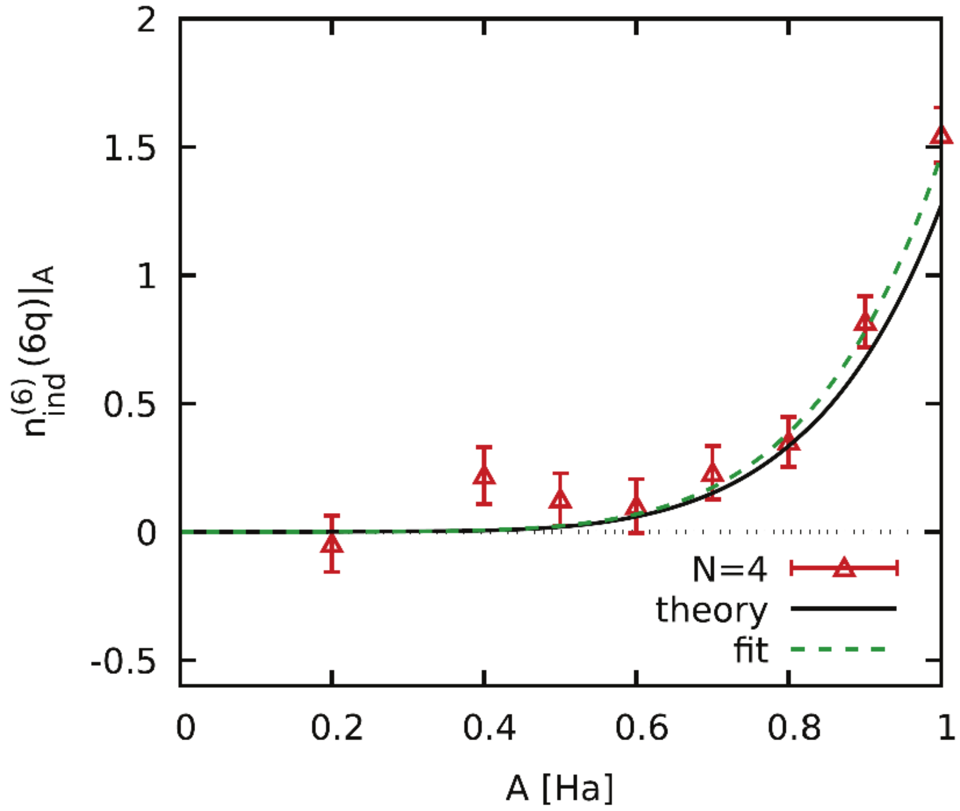
Sixth-order density response (!)



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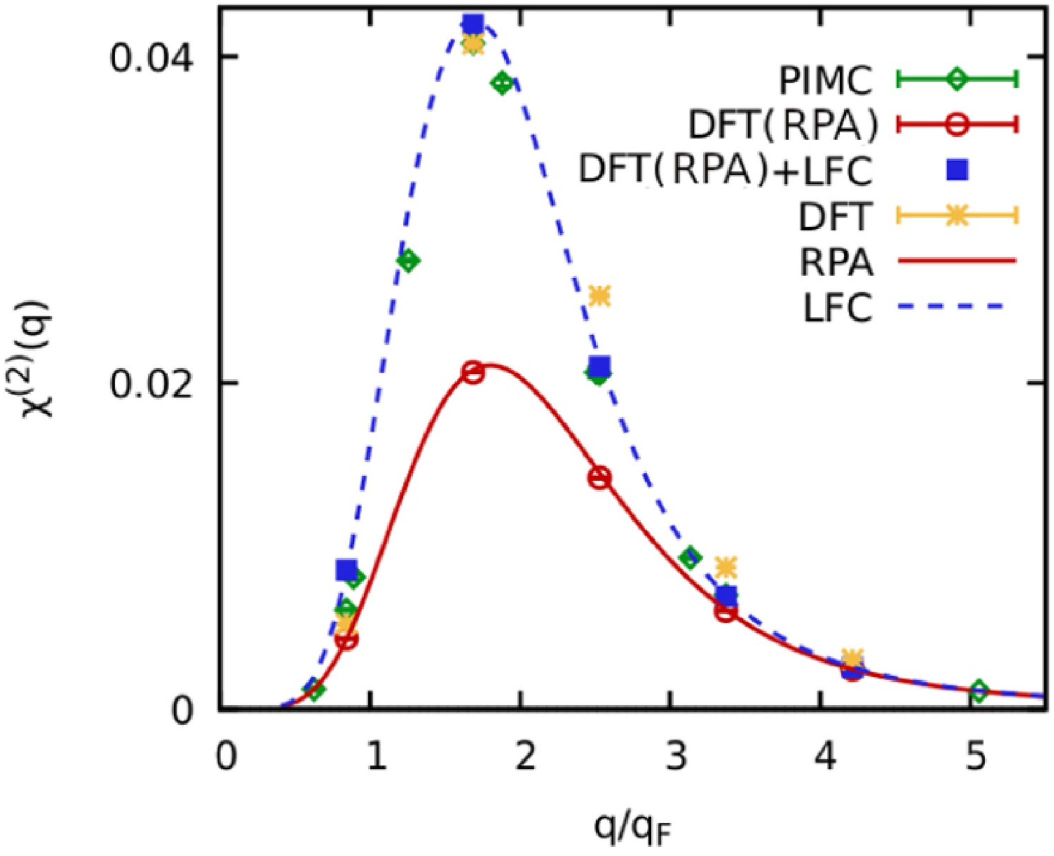

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