Toward predictive first-principles simulations of warm dense matter

Michael Bonitz

Chair Statistical Physics
Institute for Theoretical Physics and Astrophysics, Kiel University

In collaboration with Christopher Makait, Linda Kordts, Paul Hamann,
Hanno Kählert, Daniels Krimans, Tobias Dornheim, Hanna Bellenbaum, and Jan Vorberger

„Warm Dense Matter“, Santa Fe, April 2025

pdf of talk at https://www.itap.uni-kiel.de//theo-physik/bonitz/talks.html

Motivation: Warm dense matter

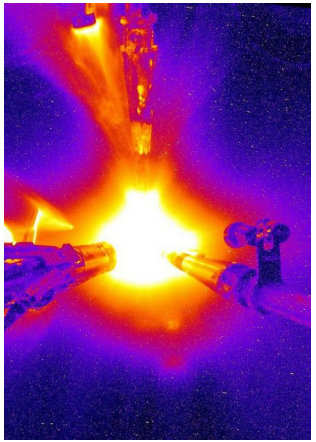
Astrophysics:

- gian planet interiors
- brown dwarfs
- newly discovered exoplanets
- Earth interior

Sci-News.com [Img4]

Lab experiments,
shock compression:

- lasers, FELs, Z-pinch, ion beams
- basic high compression studies at the NIF
- ICF



Don Jedlovec, LLNL

Motivation: Warm dense matter

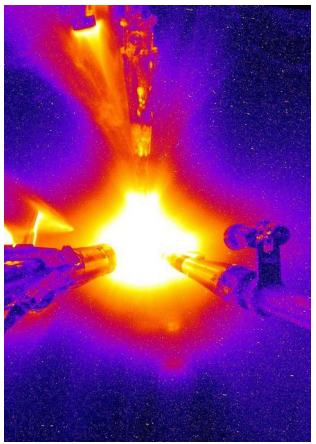
Astrophysics:

- gian planet interiors
- brown dwarfs
- newly discovered exoplanets
- Earth interior

Sci-News.com [Img4]

Lab experiments, shock compression:

- lasers, FELs, Z-pinch, ion beams
- basic high compression studies at the NIF
- ICF



Don Jedlovec, LLNL

Basic physics:

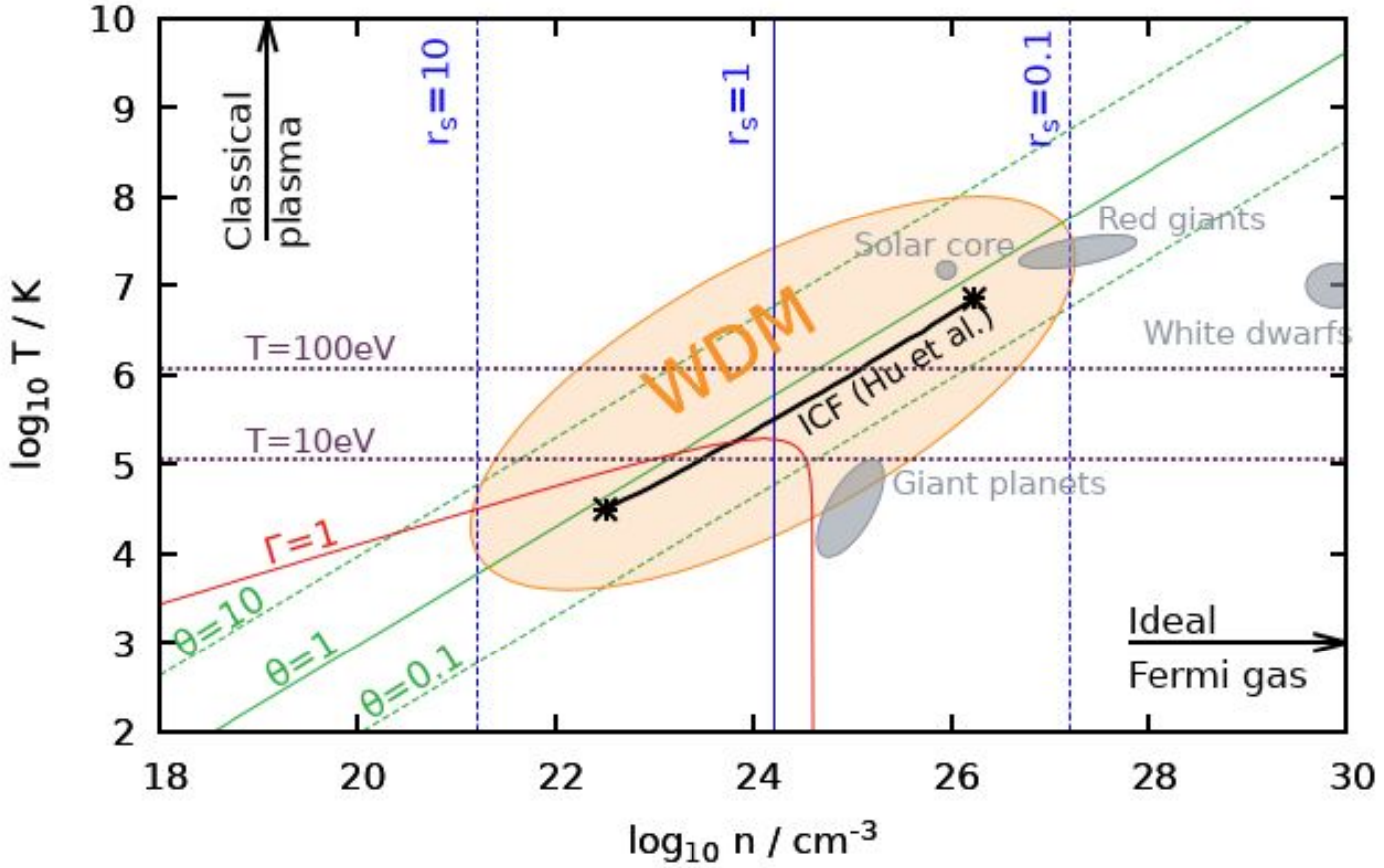
- behavior of matter under compression
- transition of condensed matter into plasma state
- properties under strong excitation

- behavior of atoms, molecules: Modification of bound states: IPD, continuum lowering

Simulation approaches:

- capture correlations, quantum and spin effects
- resolve relevant time scales

Motivation: Warm dense matter*



$$\Theta = \frac{k_B T}{E_F}$$ quantum degeneracy

$$r_s = \frac{d}{a_B}$$ coupling (T=0)

$$\Gamma = \frac{e^2}{d k_B T \frac{2\sigma+1}{n\Lambda^3} I_{3/2}(\beta\mu)}$$ generalized coupling par.

*M. Bonitz *et al.*, Phys. Plasmas 2024

Novel opportunities

1. dramatic recent progress in WDM experiments, in particular, at the NIF

2. progress in WDM diagnostics, accuracy, e.g., X-ray Thomson scattering (XRTS)

3. progress in theory and simulations: traditional (hydrodyamics, kinetic theory) and more recent methods – DFT, TDDFT, Green functions, quantum Monte Carlo

Novel opportunities

1. dramatic recent progress in WDM experiments, in particular, at the NIF

2. progress in WDM diagnostics, accuracy, e.g., X-ray Thomson scattering (XRTS)

3. progress in theory and simulations: traditional (hydrodyamics, kinetic theory) and More recent methods – DFT, TDDFT, Green functions, quantum Monte Carlo

4. breakthroughs in first-principles simulations for WDM in our group:
   - new fermionic PIMC simulations (PB-PIMC, CPIMC) for uniform electron gas benchmark data for thermodynamic functions (2011-2018)
   - dynamic structure factor, dielectric function (T. Dornheim *et al.*, 2018)
   - new first-principles hydrogen simulations (Filinov, Bonitz 2023)
   - new results by T. Dornheim *et al.:* imaginary time correlation functions, model-free analysis of XRTS experiments

Physics Reports **744**, 1-86 (2018)

The uniform electron gas at warm dense matter conditions

Tobias Dornheim [1], Simon Groth [1], Michael Bonitz *

Institut für Theoretische Physik und Astrophysik, Christian-Albrechts-Universität zu Kiel, Leibnizstr. 15, 24098 Kiel, Germany

PHYSICAL REVIEW LETTERS **121,** 255001 (2018)

Ab initio Path Integral Monte Carlo Results for the Dynamic Structure Factor of Correlated Electrons: From the Electron Liquid to Warm Dense Matter

T. Dornheim,[1] S. Groth,[1] J. Vorberger,[2] and M. Bonitz[1]

[1]*Institut für Theoretische Physik und Astrophysik, Christian-Albrechts-Universität zu Kiel, Leibnizstraße 15, D-24098 Kiel, Germany*

[2]*Helmholtz-Zentrum Dresden-Rossendorf, D-01328 Dresden, Germany*

PHYSICAL REVIEW E **108**, 055212 (2023)

Equation of state of partially ionized hydrogen and deuterium plasma revisited

A. V. Filinov * and M. Bonitz †

Institut für Theoretische Physik und Astrophysik, Christian-Albrechts-Universität zu Kiel, Leibnizstraße 15, 24098 Kiel, Germany

WDM enters a new era of high-precision research

Tremendous opportunities

Many challenges

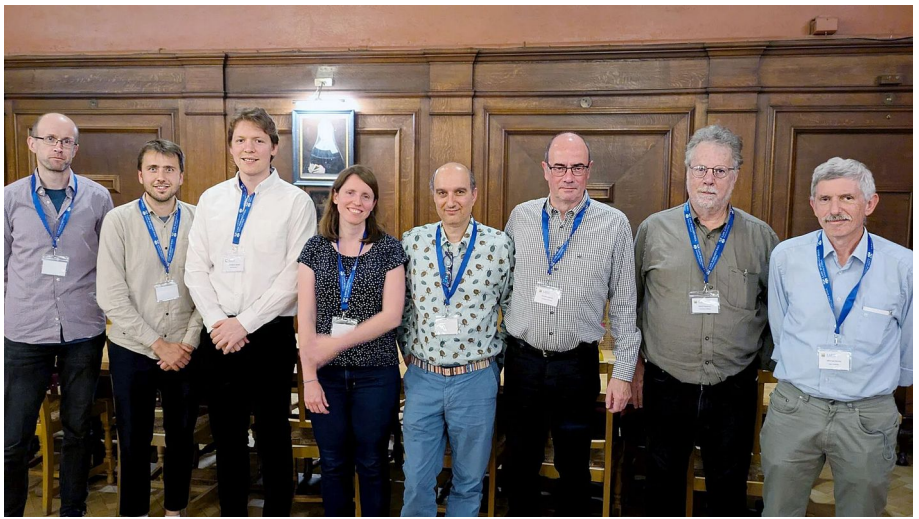
Recently: comprehensive analysis for warm dense hydrogen

REVIEW ARTICLE | NOVEMBER 13 2024

Toward first principles-based simulations of dense hydrogen

Special Collection: Papers from the 65th Annual Meeting of the APS Division of Plasma Physics , Reviews and Tutorials in Inertially Confined Plasmas, High-Energy Density Plasma Science, and Warm Dense Matter

Michael Bonitz ; Jan Vorberger ; Mandy Bethkenhagen ; Maximilian P. Böhme ; David M. Ceperley ; Alexey Filinov ; Thomas Gawne ; Frank Graziani ; Gianluca Gregori ; Paul Hamann ; Stephanie B. Hansen ; Markus Holzmann ; S. X. Hu ; Hanno Kählert ; Valentin V. Karasiev ; Uwe Kleinschmidt ; Linda Kordts ; Christopher Makait ; Burkhard Militzer ; Zhandos A. Moldabekov ; Carlo Pierleoni ; Martin Preising ; Kushal Ramakrishna ; Ronald Redmer ; Sebastian Schwalbe ; Pontus Svensson ; Tobias Dornheim



Phys. Plasmas **31**, 110501 (2024)
(86 pages, 800 Refs.)

at PNP conference,
Oxford Univ, LMH 2024

Abstract

- very large variety of models and simulations including path integral Monte Carlo (PIMC) simulations, density functional theory (DFT), chemical models, machine-learned models, and combinations thereof.

- each of these methods has fundamental limitations (fermion sign problem in PIMC, approximate exchange-correlation functionals of DFT, inconsistent interaction energy contributions in chemical models, etc.), so for some parameter ranges accurate predictions are difficult.

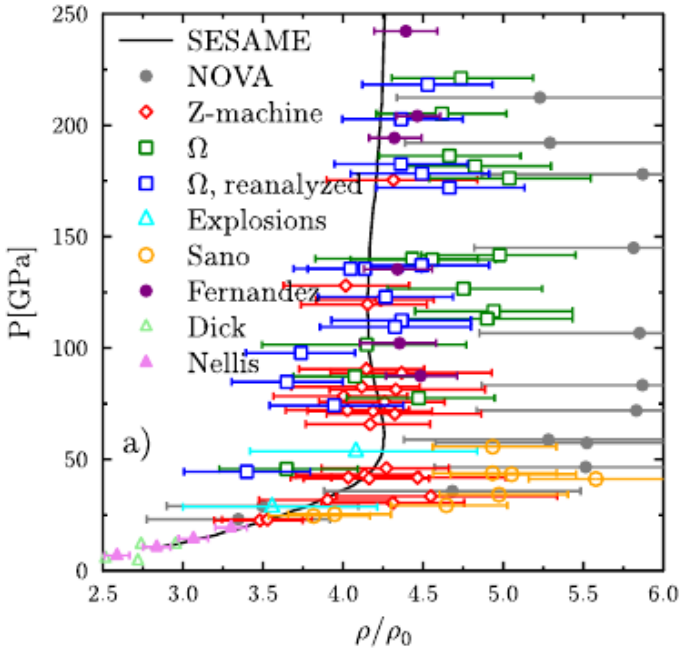
- Recently, a number of breakthroughs in first principle PIMC as well as in DFT simulations were achieved which are discussed in this review. Here we use these results to benchmark different simulation methods....

- strategies to combine different simulations to achieve accurate theoretical predictions that are based on first principles.

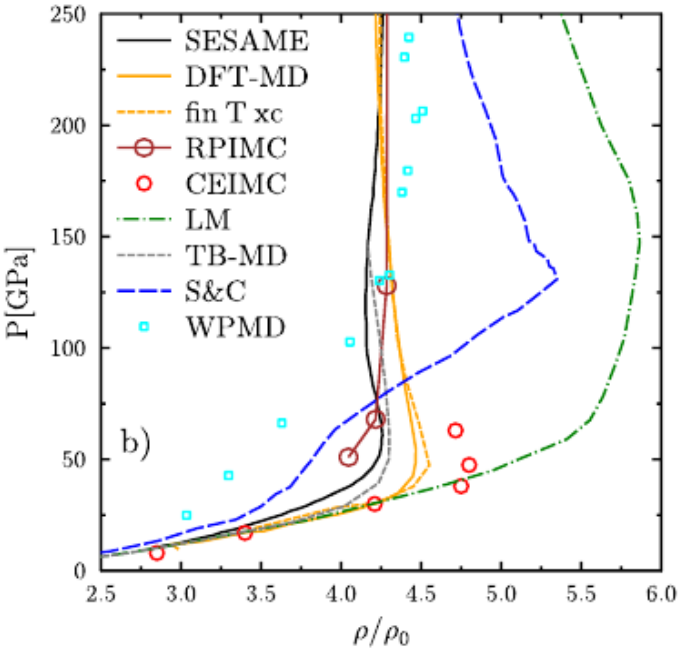
Difficult theory-experiment comparisons in WDM

Example: hydrogen hugoniot (1996-2000, unknown T, model input)

Experiment



Theory



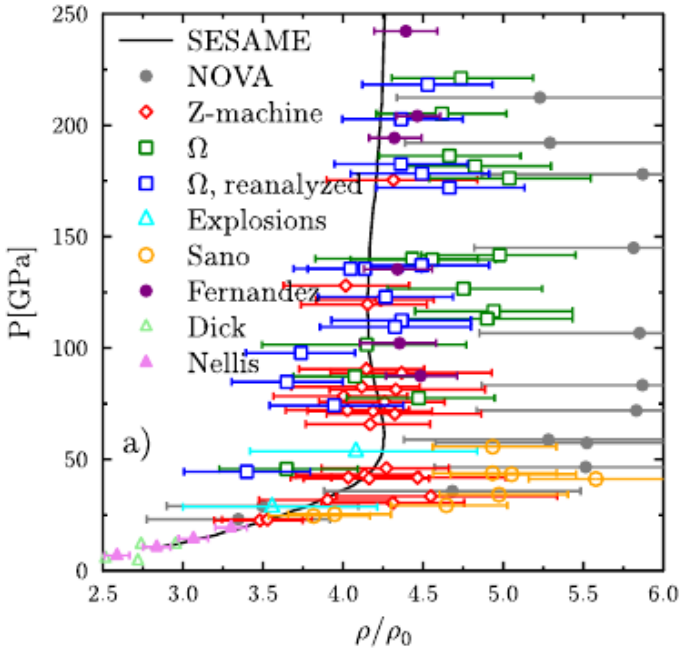
Experiment:
- large scatter of data
- large statistical errors
- poorly known system. errors

from M. Bonitz *et al.*, Phys. Plasmas 2024

Difficult theory-experiment comparisons in WDM

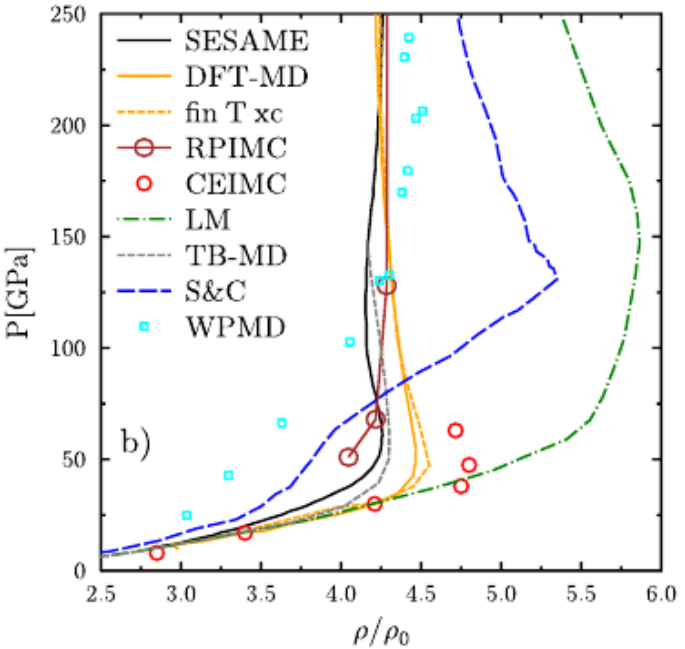
Example: hydrogen hugoniot (1996-2000, unknown T, model input)

Experiment



from M. Bonitz *et al.*, Phys. Plasmas 2024

Theory



RPIMC, CEIMC: quantum MC; LM: linear mixing, S&C: Saumon/Chabrier
WPMD: wave packet MD

Experiment:
- large scatter of data
- large statistical errors
- poorly known system. errors

Theory:
- general goal (of course): agreement with experiment
- here: agreement does not mean that theory is correct

needed:
- independent, reliable results

How to compare theory and experiment in WDM?

Given the large error in the experiments – does it matter at all which simulation to use?

Given the large scatter in simulation results – how to proceed?
Take the average of all simulations (weighted by the number of groups or papers)?

Given the large diversity of simulation approaches – can experiments help to „discriminate between models“?

How to compare theory and experiment in WDM?

Given the large error in the experiments – does it matter at all which simulation to use?

Of course

Given the large scatter in simulation results – how to proceed?
Take the average of all simulations (weighted by the number of groups or papers)?

Not a good idea

Given the large diversity of simulation approaches – can experiments help to „discriminate between models“?

Yes, for complex models. Not for simple observables.

How to compare theory and experiment in WDM?

Deuterium hugoniot: more recent experiments

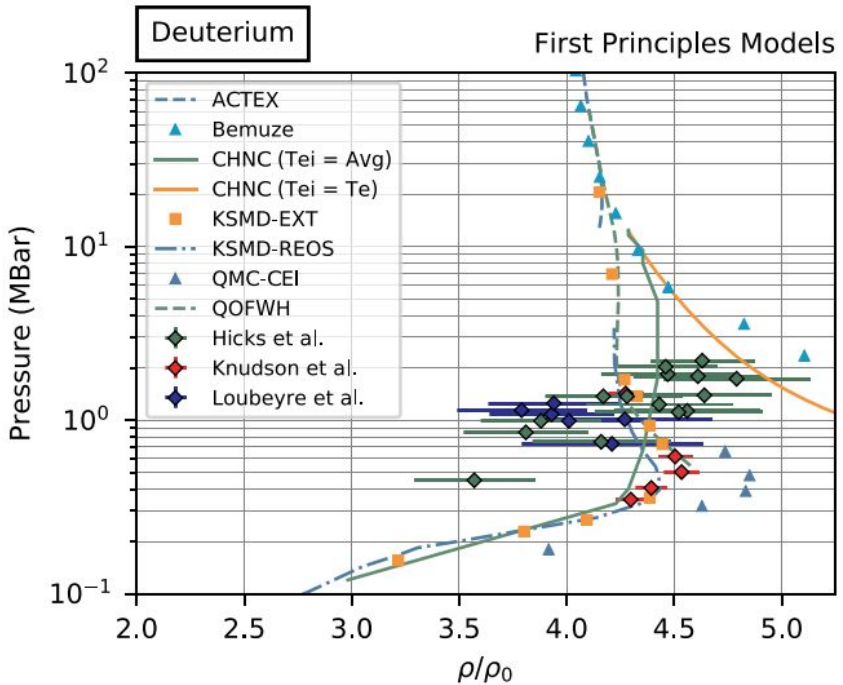


Fig. 2. Pressure along the principal Hugoniot for deuterium.

Qualitatively different simulation models*:

1. „first principles“: QMC and DFT (KSMD)
2. simpler models: ACTEX, CHNC

*from Gaffney *et al.*, HED Phys. (2018): first code comparison for ICF

How to compare theory and experiment in WDM?

Deuterium hugoniot: more recent experiments

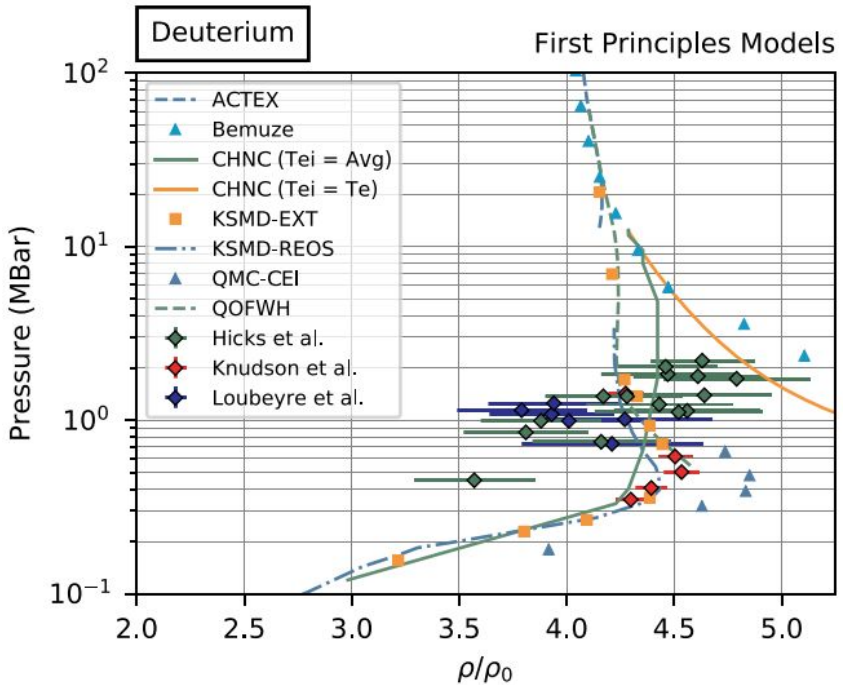


Fig. 2. Pressure along the principal Hugoniot for deuterium.

Qualitatively different simulation models*:

1. „first principles“: QMC and DFT (KSMD)
2. simpler models: ACTEX, CHNC

No experiment is needed (able) to „discriminate“ between QMC, DFT, average atom, CHNC, for simple observables, such as EOS

*from Gaffney *et al.*, HED Phys. (2018): first code comparison for ICF

How accurate are the simulations?

How large are the statistical errors in the simulation?

How large are the systematic errors?
What are the assumptions being made?
What are the validity limits (parameter range) of the simulation?

In practice: accuracy of most simulation results not known. No predictive capability

How accurate are the simulations?

How large are the statistical errors in the simulation?

How large are the systematic errors?
What are the assumptions being made?
What are the validity limits (parameter range) of the simulation?

In practice: accuracy of most simulation results not known. **No predictive capability**

Choice of approximations based on experience, intuition

Examples: E_{xc} of DFT, selfenergy of Green functions, Coulomb logarithm, IPD, screening parameters, hydrodynamic closures etc.

Novel opportunity: use accurate, predictive simulations as benchmark

Simulations with predictive capability

They exist!	**Even though, they have limitations**
- Solution of Schrödinger's equation (CI)	Only N=2...20, microcanonical ensemble
- DMRG simulations	Only 1D
- Quantum Monte Carlo simulations without nodal or other restrictions	only for parameters permitted by fermion sign problem, only WDM in equilibrium
Additionally: exact results for models: Ideal gas, uniform electron gas*, OCP etc.	No experimental realization, but useful special cases for simulations (match Θ, Γ, r_s)

Red: relevant for WDM, *Dornheim et al., Phys. Rep. **744**, 1-86 (2018)

Simulations with predictive capability

They exist!	**Even though, they have limitations**
- Solution of Schrödinger's equation (CI)	Only N=2...20, microcanonical ensemble
- DMRG simulations	Only 1D
- Quantum Monte Carlo simulations without nodal or other restrictions	only for parameters permitted by fermion sign problem, only WDM in equilibrium
Additionally: exact results for models: Ideal gas, uniform electron gas, OCP etc.	No experimental realization, but useful special cases for simulations (match Θ, Γ, r_s)

Computer experiments are source for benchmarks, complementing real experiments

Simulations relevant for warm dense matter (1)*

TABLE III. First-principles simulation methods for dense hydrogen in thermodynamic equilibrium at finite temperature ordered by their expected accuracy. Numbers I.–V. in the column "Observables" refer to the list in Sec. III A 2. $G^M(q,\tau)$: Matsubara Green function of "imaginary time" τ. The column "benchmarks" lists available relevant benchmarks of pressure (p), interaction energy (V), degree of ionization (α). For new benchmarks provided in this paper, the relevant figures are indicated, for details see footnotes and main text. For a more complete list of observables, see Tab. VI.

Method	Observables	approximations	main limitations	benchmarks	Section
Fermionic PIMC (FPIMC)	I., II. $G^M(q,\tau)$ nonlinear response	number of particles N number of high-T factors P	fermion sign problem $\Theta \gtrsim 0.5$ statistical errors	N/A	III E 1
Restricted PIMC (RPIMC)	I.	fixed node approximation	$\Theta \gtrsim 0.1$	p, α based on FPIMC Figs. 14, 23	III E 4
CEIMC	I., IV. DOS, energy gap	BO approximation Electrons in ground state using VMC or RQMC	$\Theta \lesssim 0.1$	N/A	III F
Green functions	I., II., III., IV. $A(q,\omega)$ DOS	Selfenergy Σ	moderate coupling strength	interaction energy based on FPIMC[a] model systems	III H 3
Kohn-Sham-DFT-MD	I., V. III.[b]	BO approx., XC functional		p for LDA, PBE and KDT16 Fig. 15	III C
Linear-response TDDFT	II.	BO approx., XC functional XC-kernel	no reliable dynamic XC-kernel	N/A[c]	III H 4
Orbital free DFT-MD	I., V.	BO approx., XC functional non-interacting free-energy functional	accurate only for relatively high T due to free-energy functional	N/A	III C

[a] Benchmarks of potential energy of Ref. 117 for jellium.
[b] Optical properties can be computed from a set of KS-orbitals via the Kubo-Greenwood relation that does, however, not include electron-electron collision, see Sec. IV E.
[c] Note that LR-TDDFT results for different XC-functionals and XC-kernels can be benchmarked against PIMC results for the ITCF $F(\mathbf{q},\tau)$ [cf. Eq. (121)] in future studies.

Observables

I. TD properties: p, E, g(r), S(q)

II. e-dynamic properties:
$S_{ee}(q,\omega)$, $F_{ee}(q,\tau)$

III. e-transport and opt. prop.s:
$\sigma(\omega)$, $\lambda(\omega)$, $\kappa(\omega)$

IV. e-spectral properties:
DOS, $A(q,\omega)$

V. Ion dynamic properties:
$S_{ii}(q,\omega)$, $D_i(\omega)$

*M. Bonitz *et al.*, Phys. Plasmas 2024

Quantum Monte Carlo simulations for dense H*

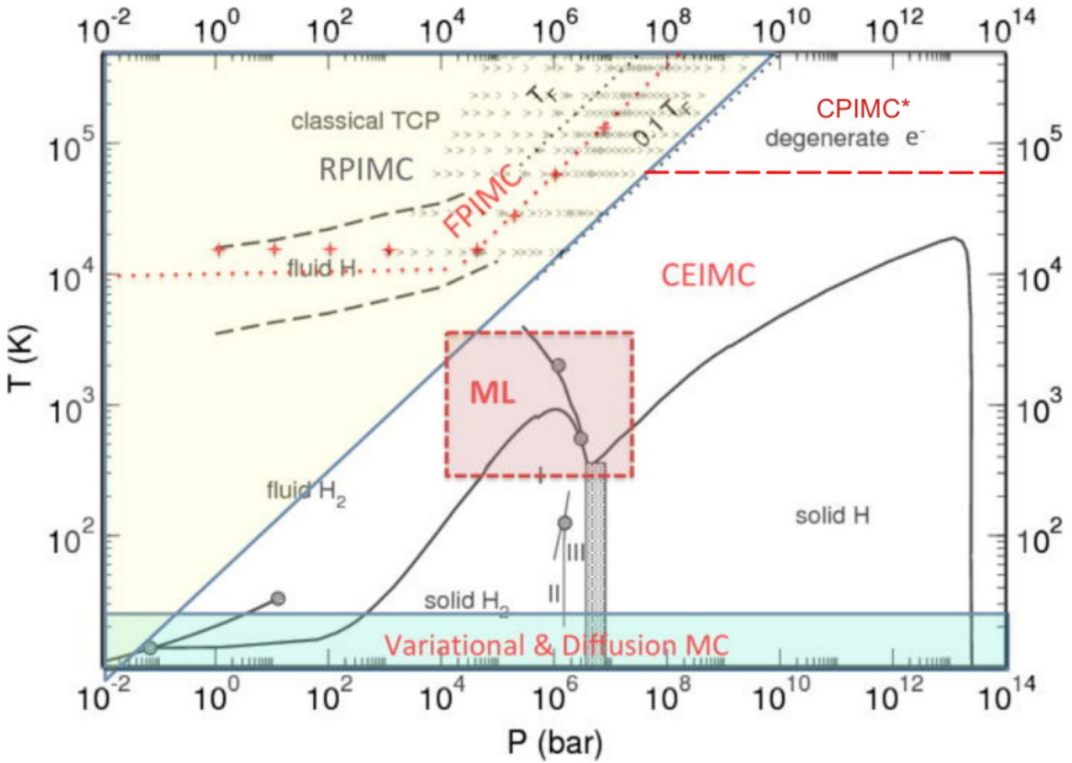


FIG. 8. Applicability range of different simulation methods across the hydrogen phase diagram. RPIMC: restricted PIMC, extends to about $0.1\,T_F$, Sec. III E 4, CEIMC: coupled electron-ion Monte Carlo, Sec. III F, FPIMC: fermionic PIMC, extends to about $0.5\,T_F$ and was applied to temperatures above 10 000 K, Sec. III E 5. ML denotes region fit by QMC-based machine learning force fields in Ref. 206 and also the region where CEIMC has been applied. Red pluses show FPIMC simulation points from Ref. 115 that mark the border of what is feasible today. Light black crosses show regions covered by the RPIMC database[114]. The other lines are introduced in Fig. 4.

CPIMC: high degeneracy, complementary to FPIMC

*M. Bonitz *et al.*, Phys. Plasmas 2024

Updated version from Graziani et al., WDM roadmap, 2025

Benchmarking RPIMC for dense H: EOS*

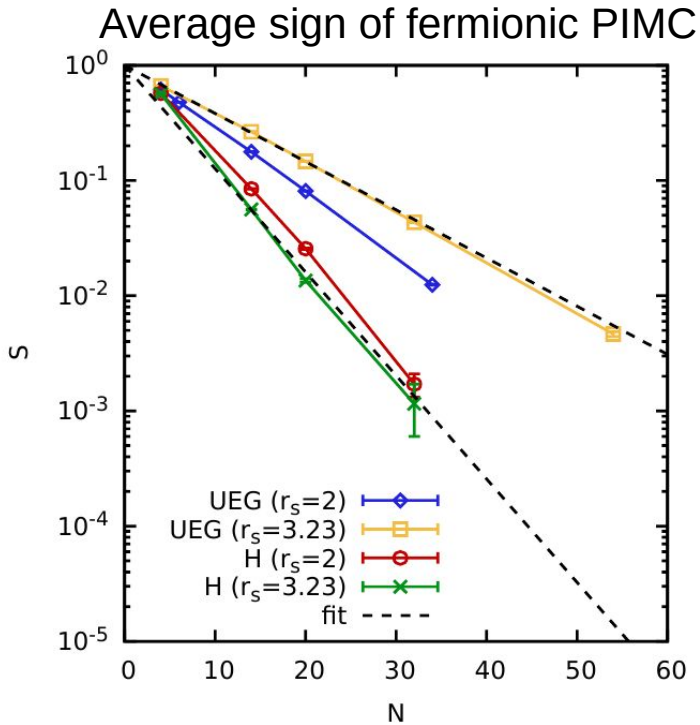
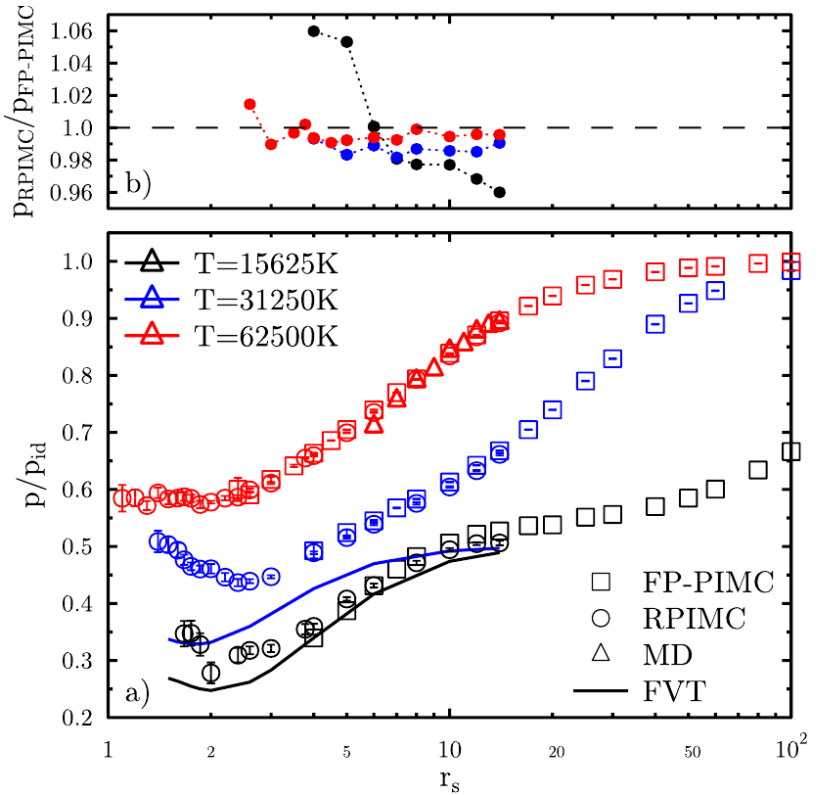


FIG. 10. Average sign S as a function of the number of electrons N at the electronic Fermi temperature $\Theta = 1$, for $r_s = 2$ ($\rho = 0.34\,\mathrm{g/cm^3}$, $T = 12.53\,\mathrm{eV}$) and $r_s = 3.23$ ($\rho = 0.08\,\mathrm{g/cm^3}$, $T = 4.80\,\mathrm{eV}$). The dashed black lines show exponential fits to the PIMC data, cf. Eq. (70). Reproduced from Ref. 149 with the permission of the authors.



RPIMC validated by fermionic PIMC

*A.Filinov and M. Bonitz, PRE **108**, 055212 (2023)
M. Bonitz *et al.*, Phys. Plasmas 2024

Simulations relevant for warm dense matter (2)*

TABLE III. First-principles simulation methods for dense hydrogen in thermodynamic equilibrium at finite temperature ordered by their expected accuracy. Numbers I.–V. in the column "Observables" refer to the list in Sec. III A 2. $G^M(q,\tau)$: Matsubara Green function of "imaginary time" τ. The column "benchmarks" lists available relevant benchmarks of pressure (p), interaction energy (V), degree of ionization (α). For new benchmarks provided in this paper, the relevant figures are indicated, for details see footnotes and main text. For a more complete list of observables, see Tab. VI.

Method	Observables	approximations	main limitations	benchmarks	Section
Fermionic PIMC (FPIMC)	I., II. $G^M(q,\tau)$ nonlinear response	number of particles N number of high-T factors P	fermion sign problem $\Theta \gtrsim 0.5$ statistical errors	N/A	III E 1
Restricted PIMC (RPIMC)	I.	fixed node approximation	$\Theta \gtrsim 0.1$	p, α based on FPIMC Figs. 14, 23	III E 4
CEIMC	I., IV. DOS, energy gap	BO approximation Electrons in ground state using VMC or RQMC	$\Theta \lesssim 0.1$	N/A	III F
Green functions	I., II., III., IV. $A(q,\omega)$ DOS	Selfenergy Σ	moderate coupling strength	interaction energy based on FPIMC[a] model systems	III H 3
Kohn-Sham-DFT-MD	I., V. III.[b]	BO approx., XC functional		p for LDA, PBE and KDT16 Fig. 15	III C
Linear-response TDDFT	II.	BO approx., XC functional XC-kernel	no reliable dynamic XC-kernel	N/A[c]	III H 4
Orbital free DFT-MD	I., V.	BO approx., XC functional non-interacting free-energy functional	accurate only for relatively high T due to free-energy functional	N/A	III C

[a] Benchmarks of potential energy of Ref. 117 for jellium.
[b] Optical properties can be computed from a set of KS-orbitals via the Kubo-Greenwood relation that does, however, not include electron-electron collision, see Sec. IV E.
[c] Note that LR-TDDFT results for different XC-functionals and XC-kernels can be benchmarked against PIMC results for the ITCF $F(\mathbf{q},\tau)$ [cf. Eq. (121)] in future studies.

Observables

I. TD properties: p, E, g(r), S(q)

II. e-dynamic properties:
$S_{ee}(q,\omega)$, $F_{ee}(q,\tau)$

III. e-transport and opt. prop.s:
$\sigma(\omega)$, $\lambda(\omega)$, $\kappa(\omega)$

IV. e-spectral properties:
DOS, $A(q,\omega)$

V. Ion dynamic properties:
$S_{ii}(q,\omega)$, $D_i(\omega)$

*M. Bonitz *et al.*, Phys. Plasmas 2024

What FPIMC-based benchmarks provide*

Green functions: - exact if selfenergy would be exactly known
- available: limited number of approximations (Feynman diagrams)
- only some selfenergies can be systematically improved (perturbation theory)
- *often not clear which of the choices is the most accurate*

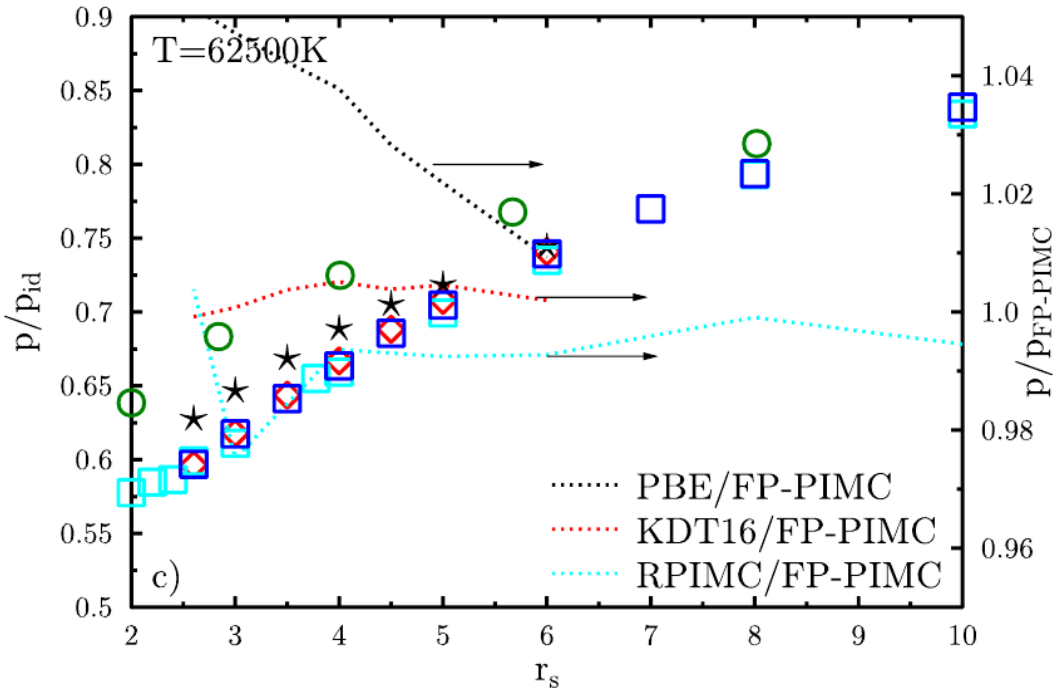
Schlünzen *et al.*, PRB **95**, 165139 (2017); J. Phys. Cond. Matt. **32**, 103001 (2020), Kas and Rehr, Phys. Rev. Lett. **119** , 176403 (2017)

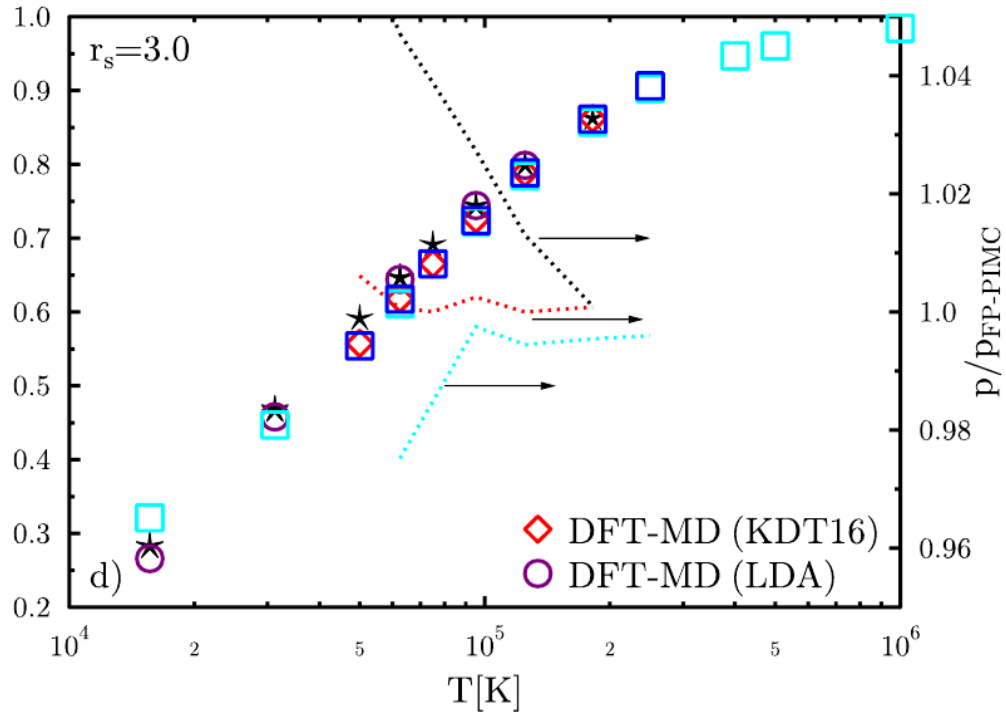
Kohn-Sham-DFT (KS-DFT, DFT-MD, based on Born-Oppenheimer approx.):
- exact if XC functional would be exactly known
- available: limited number of approximations („Jacob's ladder")
- in general, *unknown which of the choices is the most accurate*

FPIMC (or RPIMC) allows one to decide which selfenergy/XC functional is the most accurate, in the given case (and sometimes also why).

*M. Bonitz *et al.*, Phys. Plasmas 2024

Benchmarking DFT, AA simulations for dense H: EOS*

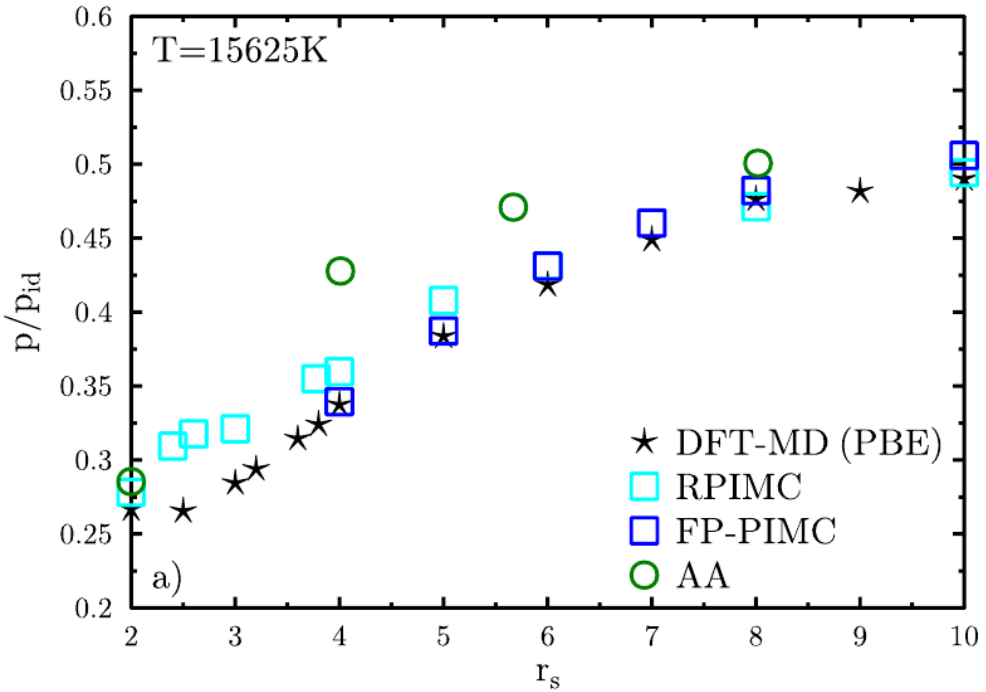


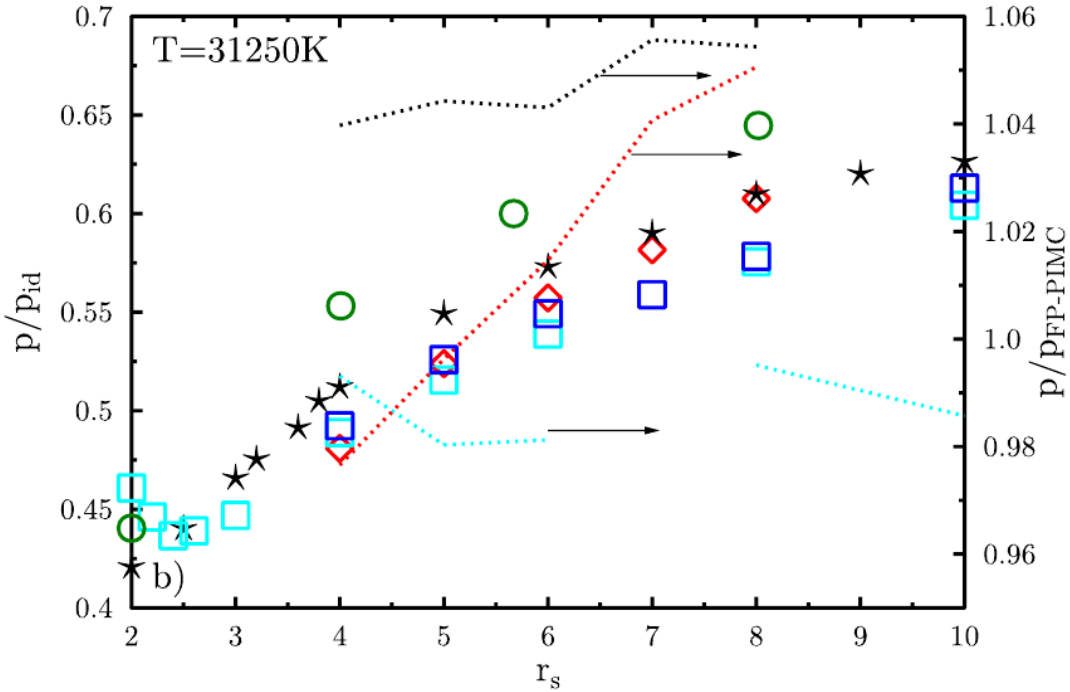


Dotted lines with arrows (right axes):
relative error compared to FPIMC

*A.Filinov and M. Bonitz, PRE **108**, 055212 (2023)
M. Bonitz *et al.*, Phys. Plasmas 2024

Benchmarking DFT, AA simulations for dense H: EOS*





*A.Filinov and M. Bonitz, PRE **108**, 055212 (2023)
M. Bonitz *et al.*, Phys. Plasmas 2024

Dotted lines with arrows (right axes): relative error compared to FPIMC

Simulations relevant for warm dense matter (3)*

TABLE IV. Further simulation methods for dense hydrogen in thermodynamic equilibrium at finite temperature, ordered by their expected accuracy. Numbers I.–VI. in the column "Observables" refer to the list in Sec. III A 2. α: degree of ionization, x_A: degree of dissociation (atom fraction). For a more complete list of observables, see Tab. VI.

Method	Observables	approximations	main limitations	benchmarks	Section
Average atom model	I., II., III. IV., V.	XC functional, single ion center	Surrounding atoms not treated explicitly	PDF based on FPIMC Fig. 18	III D
Wavepacket MD	I., II. V., VI.	classical dynamics pair potentials	wave function modeled by one or few Gaussians Approximations needed to prevent uncontrolled spreading	N/A	III G 1
Semiclassical MD	I., II. V., VI.	classical dynamics quantum pair potentials e.g., improved Kelbg pot.	$k_BT \gtrsim 0.5\,\mathrm{Ry}$ only pair exchange	pressure p, based on FPIMC Fig. 14	III G 2
Chemical models e.g., FVT	I. part of thermo-dynamic functions	equilibrium $n(k)$ α, x_A	approximations for interaction spatial homogeneity	p, x_A based on FPIMC Figs. 14, 22	III B

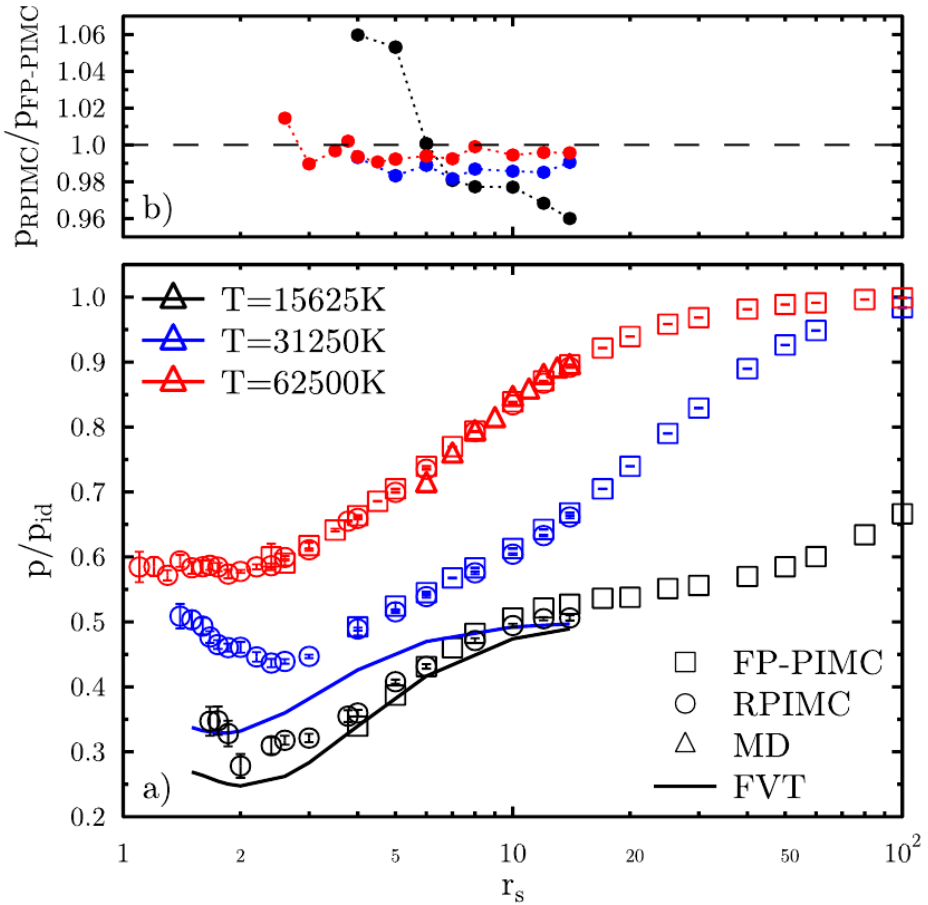
Properties: I: Thermodynamic; II: electron dynamic;
III: el. Transport; IV: el. Spectral, V: ion dynamic

*M. Bonitz *et al.*, Phys. Plasmas 2024

Benchmarking semiclassical MD and FVT for dense H: EOS*

Semiclassical MD simulations with improved Kelbg potential (Hanno Kählert)

IKP: A. Filinov et al., J. Phys. A (2003) and PRE 2004



*A.Filinov and M. Bonitz, PRE **108**, 055212 (2023)
M. Bonitz *et al.*, Phys. Plasmas 2024

Summary of thermodynamic benchmarks*

1. **RPIMC (free-particle nodes)**: very accurate in tested parameter region.
   - relative errors of the pressure do not exceed 2%, except for lowest temperature, $T = 15\,625$K.
   - accuracy of the results (and the quality of the nodes) at higher density, $r_s \lesssim 3$, remains to be tested.

2. **KS-DFT (PBE)**: moderately accurate in tested parameter region.
   - relative errors of the pressure reach 7%
   - KDT16 functional (finite T) substantially more accurate. For T= 60 000K, error ~ 2%.
     for T=30 000K error larger

3. **SC-MD (improved Kelbg)**: accuracy of 1...3%, for the pressure, for $T \gtrsim 60\,000$K and $r_s \gtrsim 7$,
   - density range quickly widens with increasing temperature. Suitable for ion-acoustic modes

4. DFT-AA: good agreement with FPIMC, for pair distributions and static structure factors,
   - with KS-DFT, for the conductivity.
   - for quantitative benchmarks, additional tests required.

*M. Bonitz *et al.*, Phys. Plasmas 2024

Which method to choose? There is no „silver bullet“*

- There is no single simulation that can reliably predict all relevant properties of warm dense matter, including ICF.

- first principles/predictive approaches do not reach the scales of interest

- simulations that do extend to large length / time scales have untested accuracy, no predictive power

* M. Bonitz *et al.*, Phys. Plasmas 2024

Which method to choose? There is no „silver bullet“*

- There is no single simulation that can reliably predict all relevant properties of warm dense matter, including ICF.

- first principles/predictive approaches do not reach the scales of interest

- simulations that do extend to large length / time scales have untested accuracy, no predictive power

The alternative: development of set of different methods and their combination.
Preserve predictive capability as much as possible

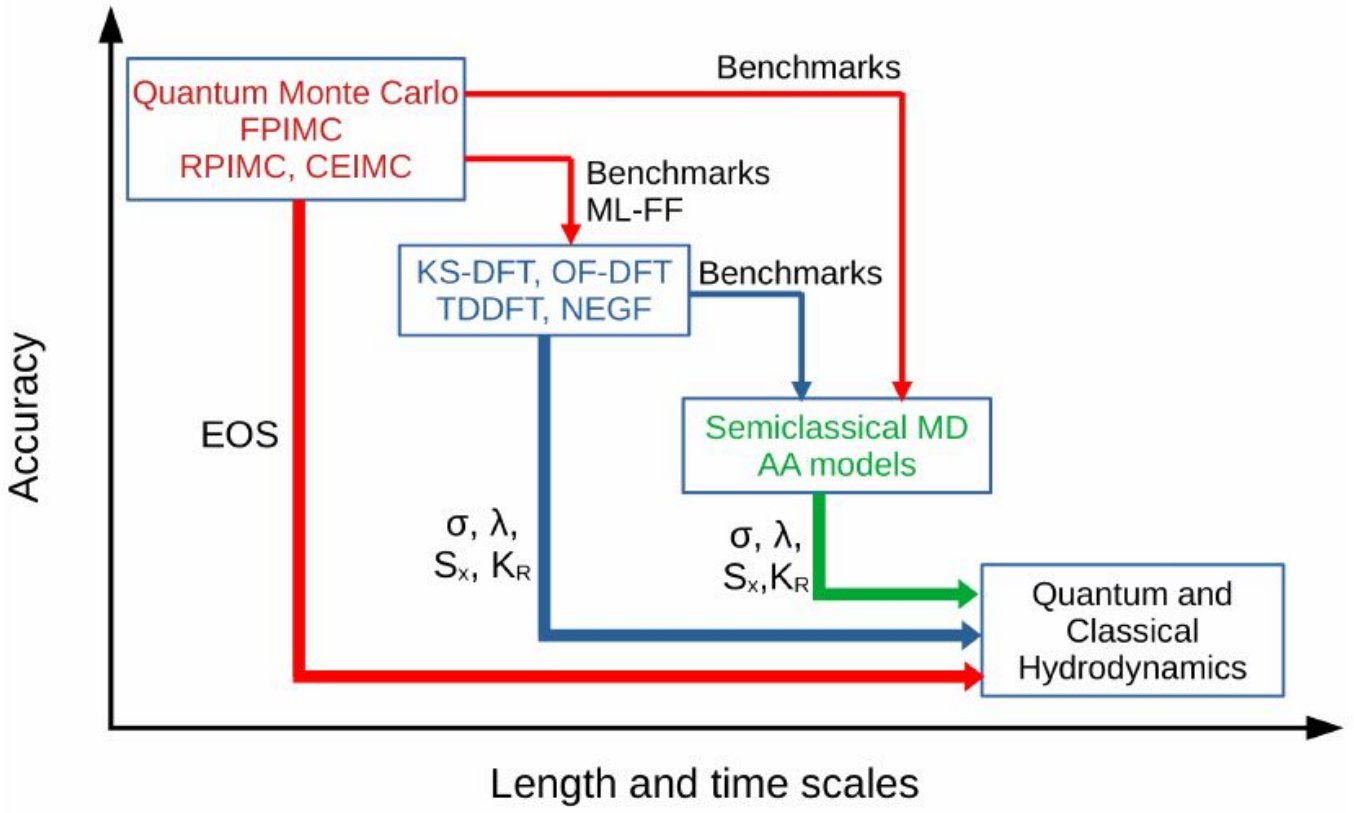
*M. Bonitz *et al.*, Phys. Plasmas 2024

Outlook: smart combination of methods

Starting from predictive simulations (Fermionic PIMC)

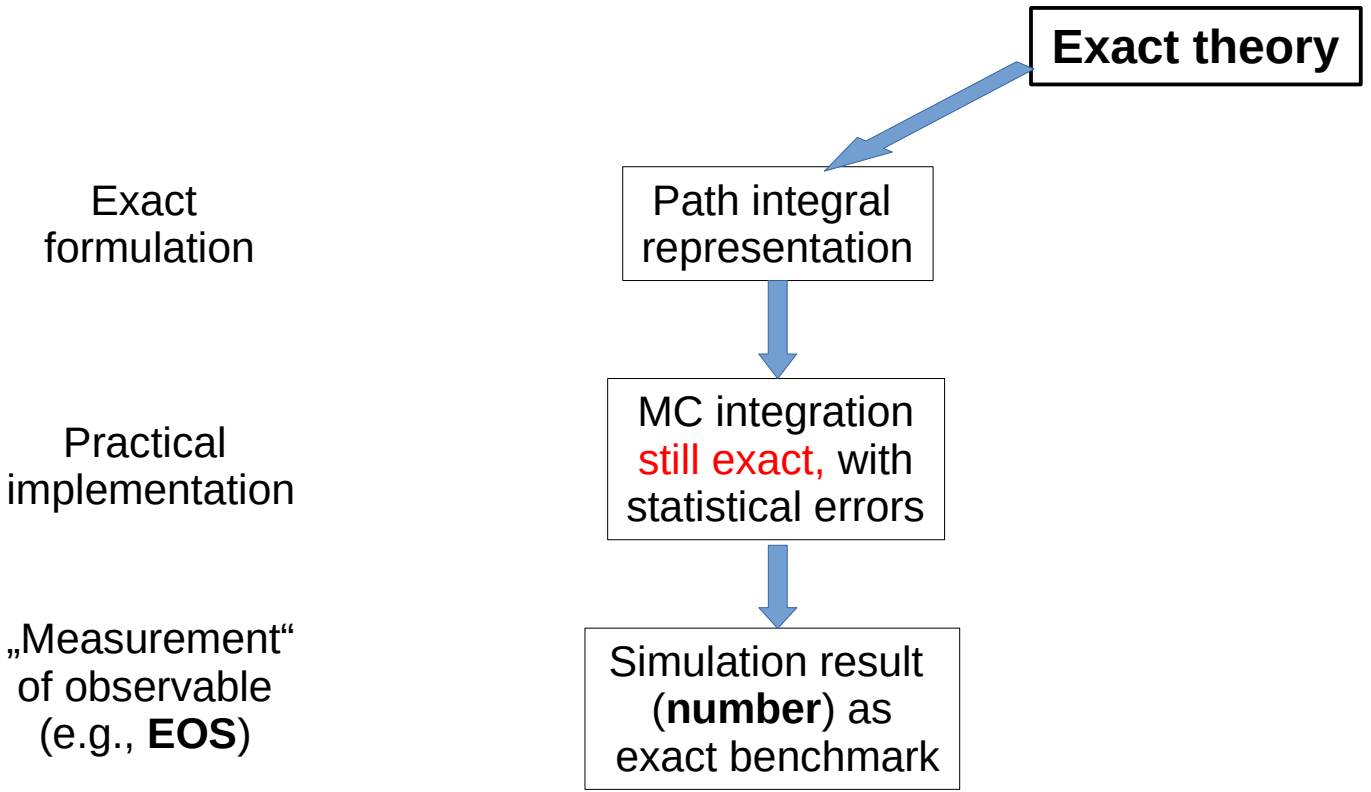
→ selection of most accurate approximations of lower level models

→ reduction of resolution + increase of accessible r- and t-scales + preservation of acceptable accuracy

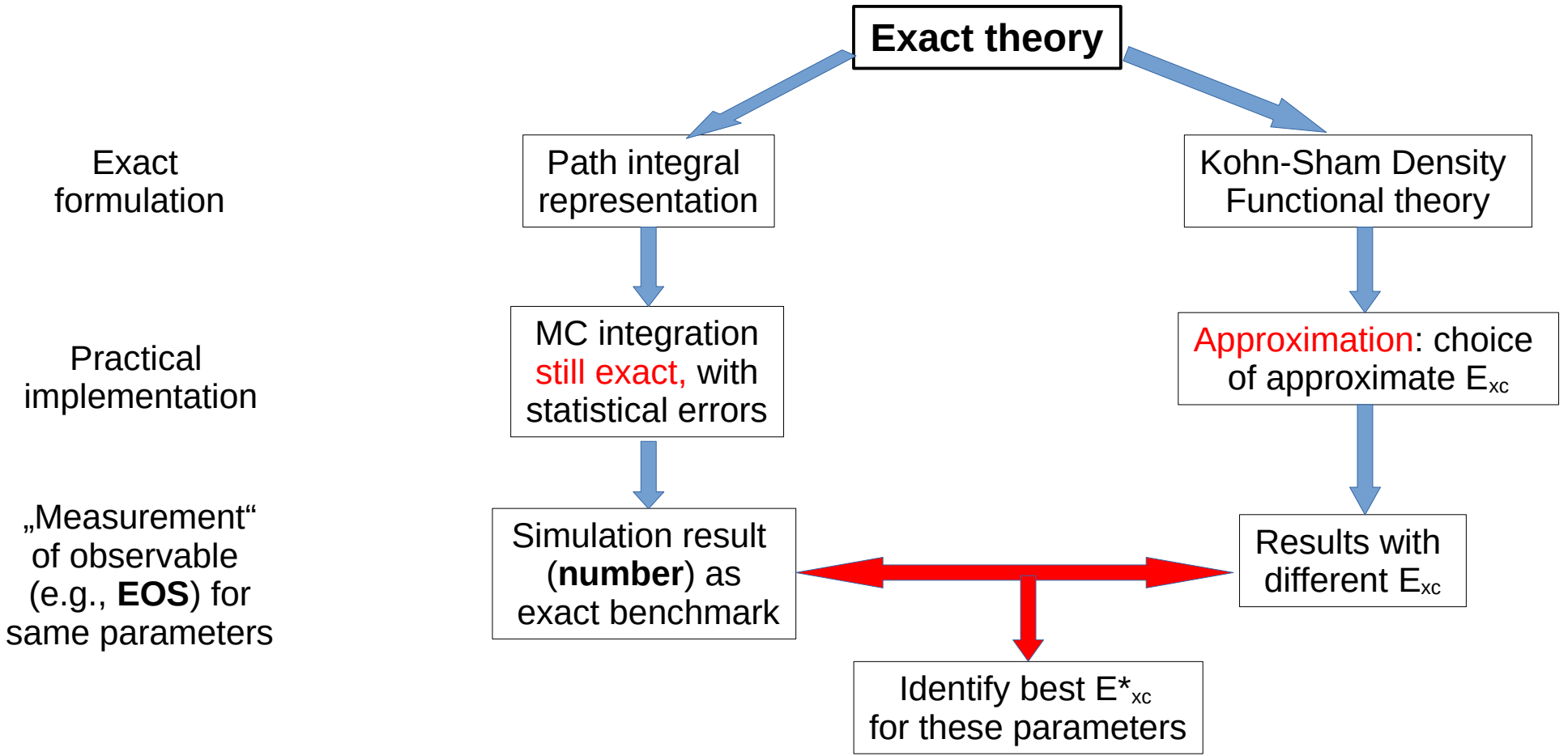


* M. Bonitz *et al.*, Phys. Plasmas 2024

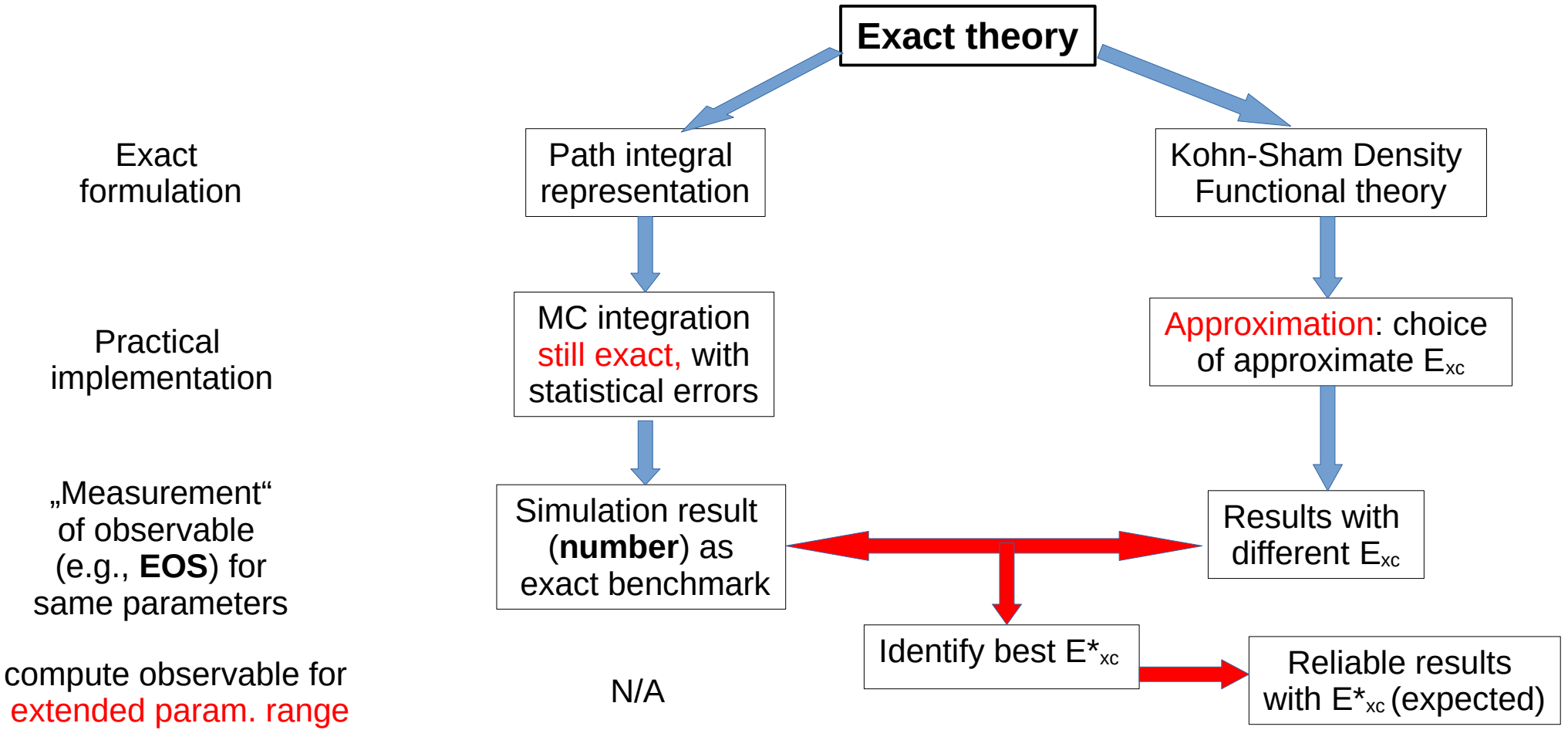
Example: FPIMC benchmarks for KS-DFT



Example: FPIMC benchmarks for KS-DFT



Example: FPIMC benchmarks for KS-DFT



Can we do better?

Can we go beyond simple benchmarks?

Can we go beyond deciding between known approximations?

Can we do better?

Can we go beyond simple benchmarks?

Can we go beyond deciding between known approximations?

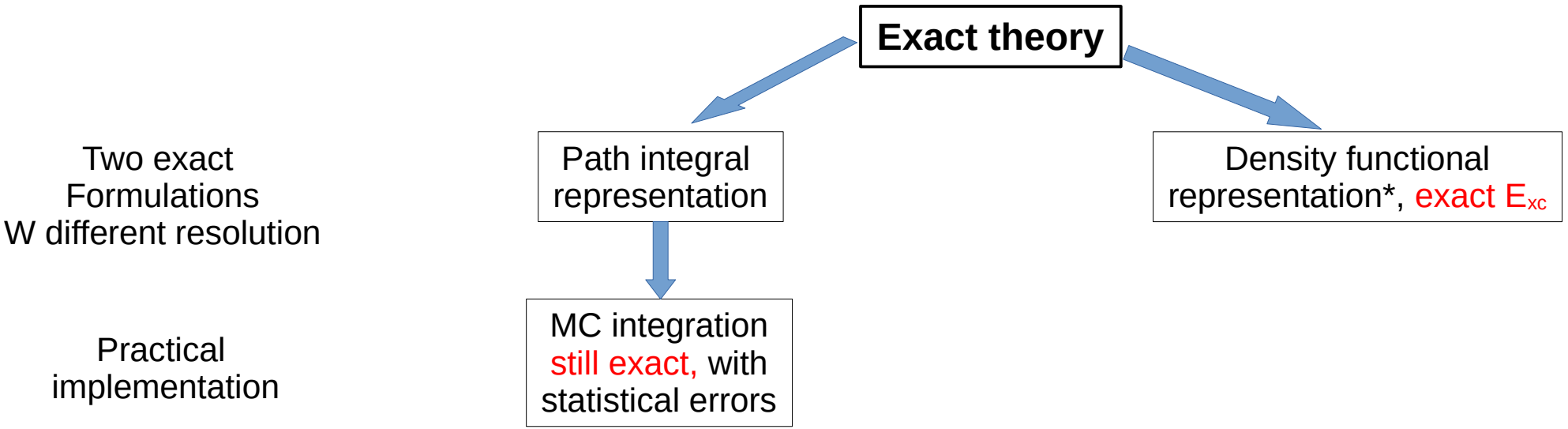
FPIMC „Downfolding“

→ use FPIMC to derive a simpler („coarse grained“) exact model

- potentially exact, with the appropriate input

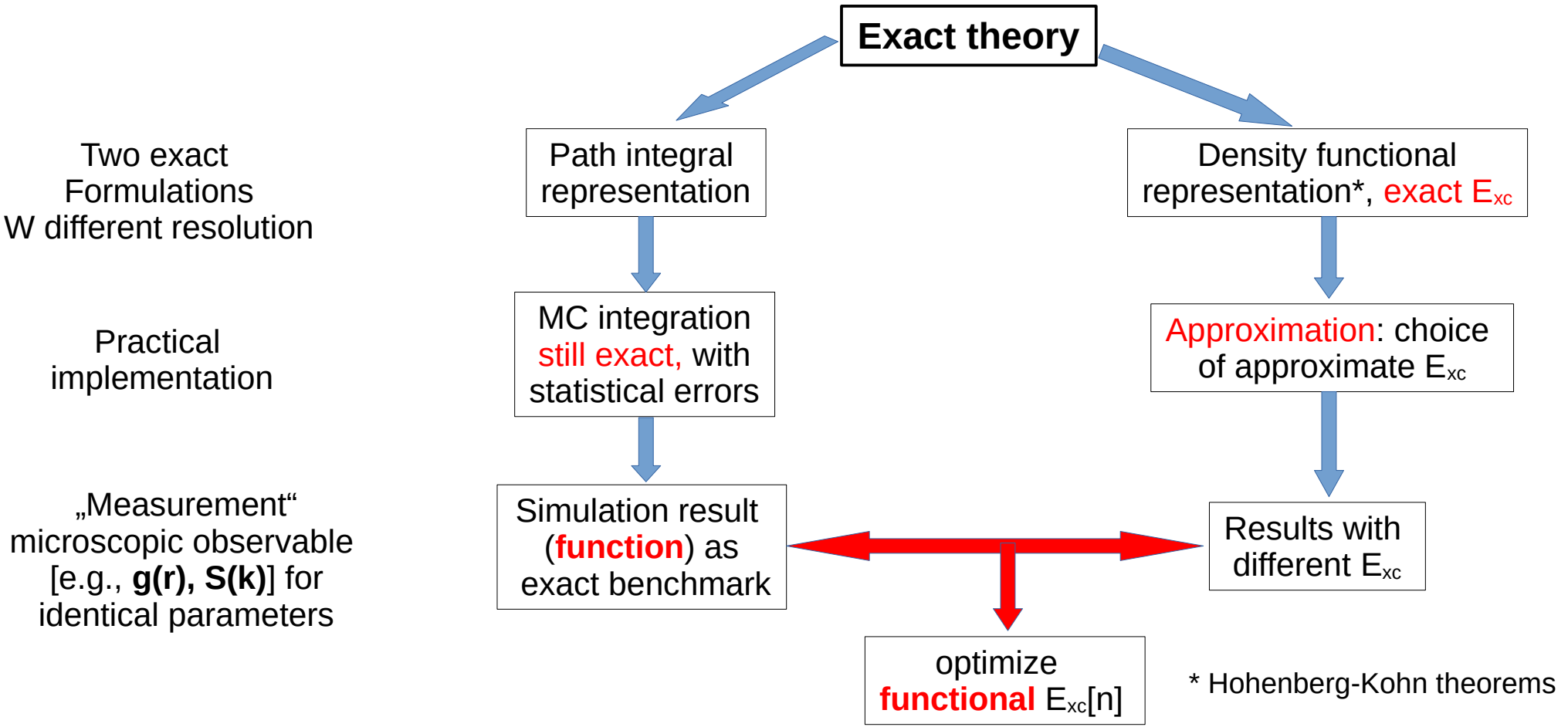
- this input (parameters) is provided by FPIMC

FPIMC downfolding (1), example KS-DFT



* Hohenberg-Kohn theorems

FPIMC downfolding (1), example KS-DFT

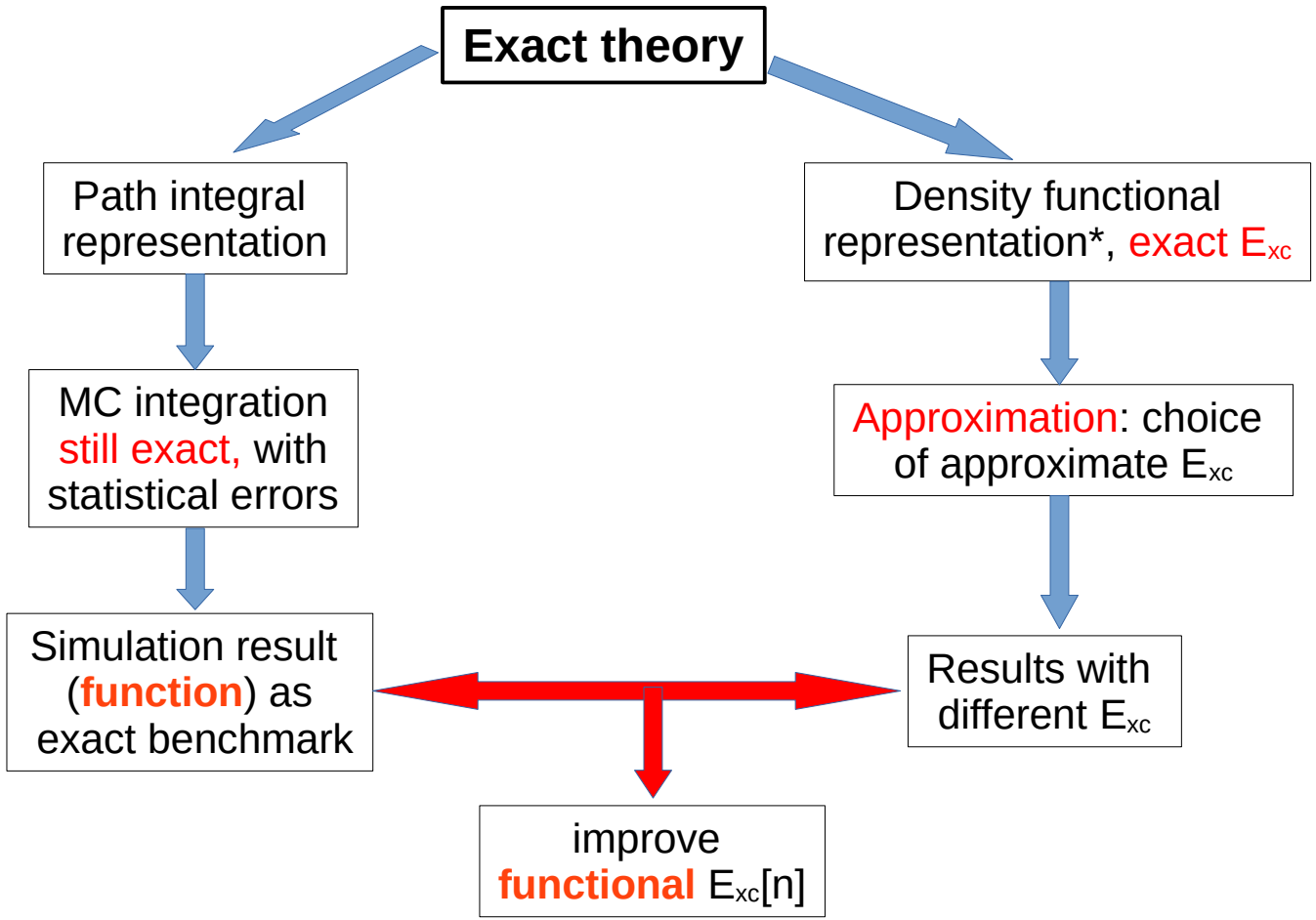


FPIMC downfolding (2), example KS-DFT

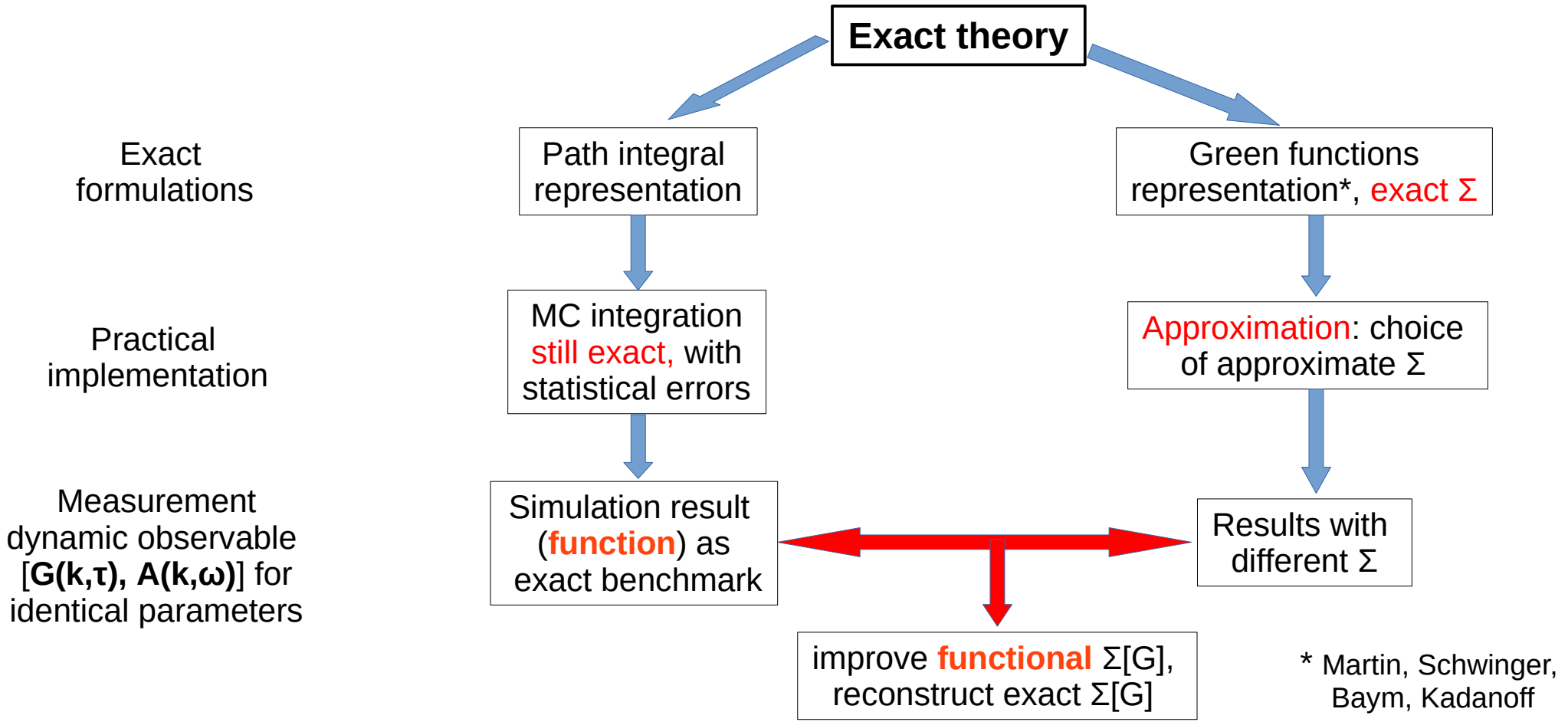
Two exact Formulations W different resolution

Practical implementation

„Measurement“ dynamic observable [**S(k,ω), F(k,τ)**] for identical parameters



FPIMC downfolding (3)→ Green functions theory



FPIMC downfolding results for Green function and spectral function

$$iG^{>}(k,\tau) = \int \frac{d\omega}{2\pi} e^{-\tau\omega} A(k,\omega)[1 - f(\omega)]$$

Imaginary time (Matsubara) Green function
Accessible in FPIMC in grand ensemble

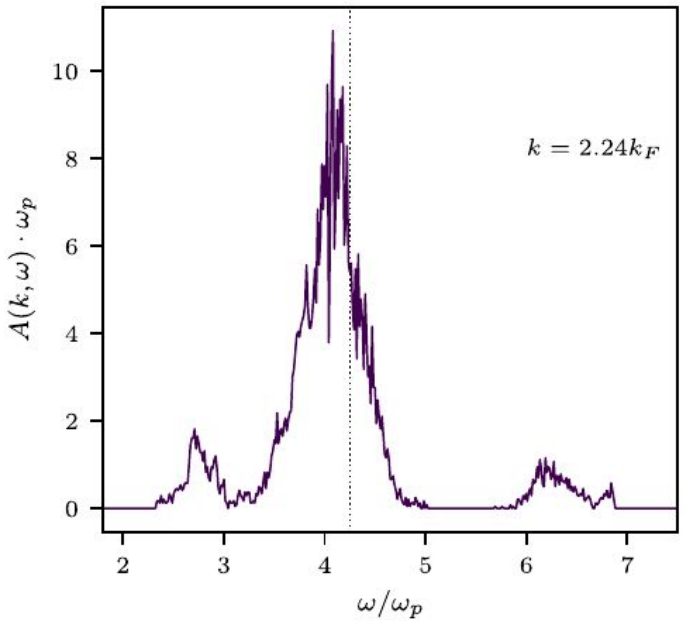
F: Fermi function of energy (known)
A: spectral function (DOS) – unknown

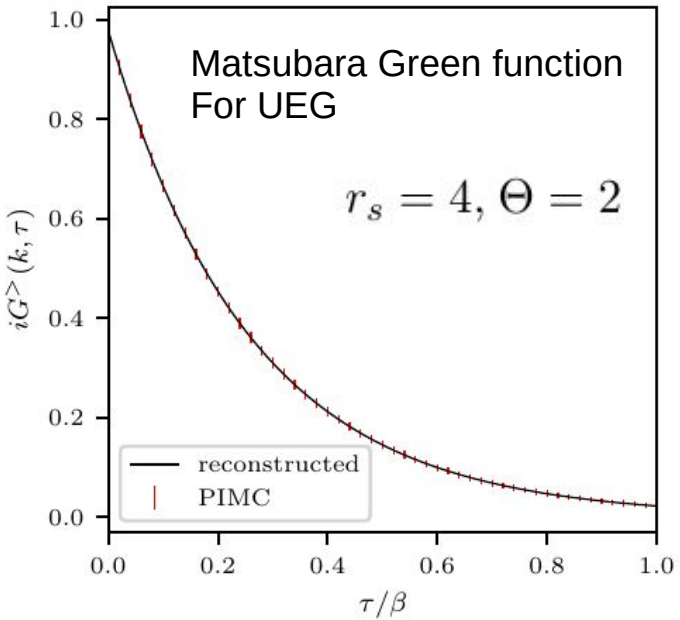
A yields selfenergy Σ

$$A(\mathbf{k},\omega) = \frac{-2\,\mathrm{Im}\,\Sigma(\mathbf{k},\omega)}{[\omega - \epsilon^0_{\mathbf{k}} - \mathrm{Re}\,\Sigma(\mathbf{k},\omega)]^2 + [\mathrm{Im}\,\Sigma(\mathbf{k},\omega)]^2}$$

M. Bonitz, „Quantum Kinetic Theory“, 2nd ed. Springer 2015; P. Hamann, Master thesis, Kiel 2025

FPIMC downfolding results for Green function and spectral function

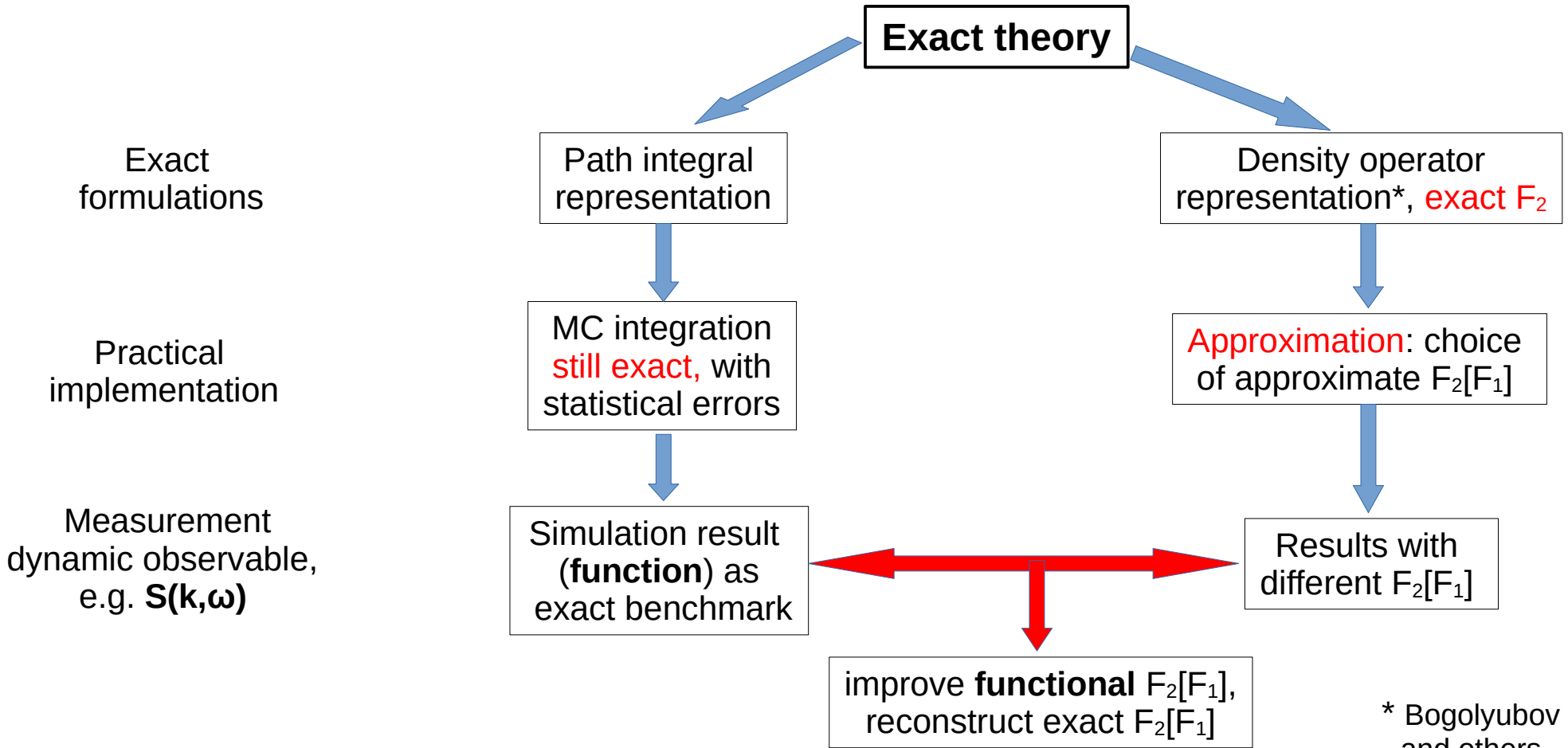




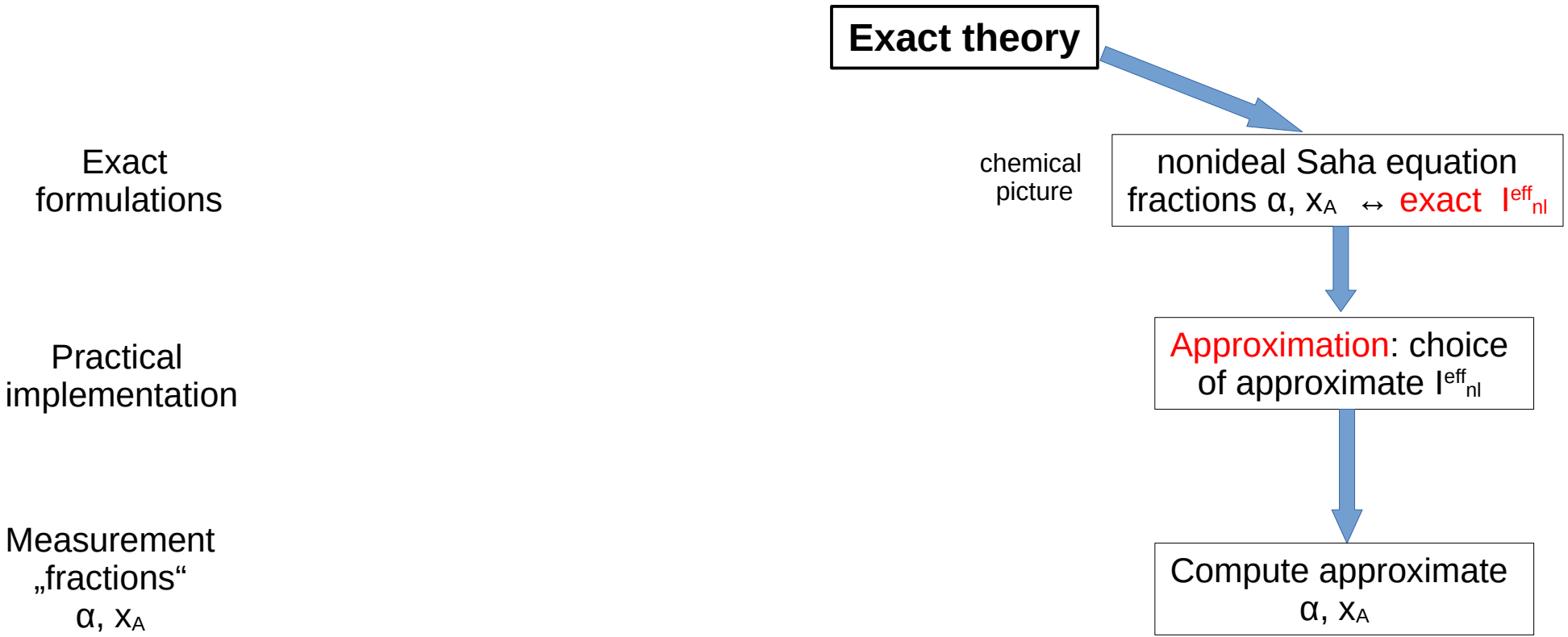
PIMC results for UEG spectral function
Quasiparticle peak and plasmon satellites.
Dotted line: ideal energy $k^2/2m$

P. Hamann, Master thesis, Kiel 2025

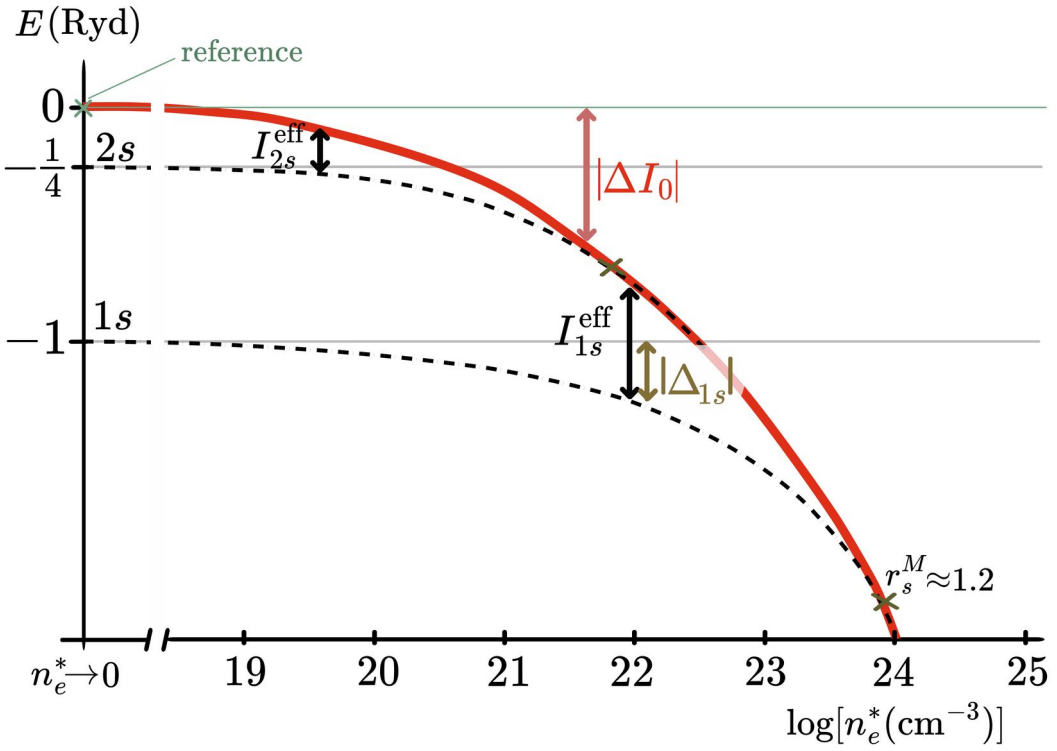
FPIMC downfolding (4)→ BBGKY-hierarchy



FPIMC downfolding (5) → Saha equation, IPD



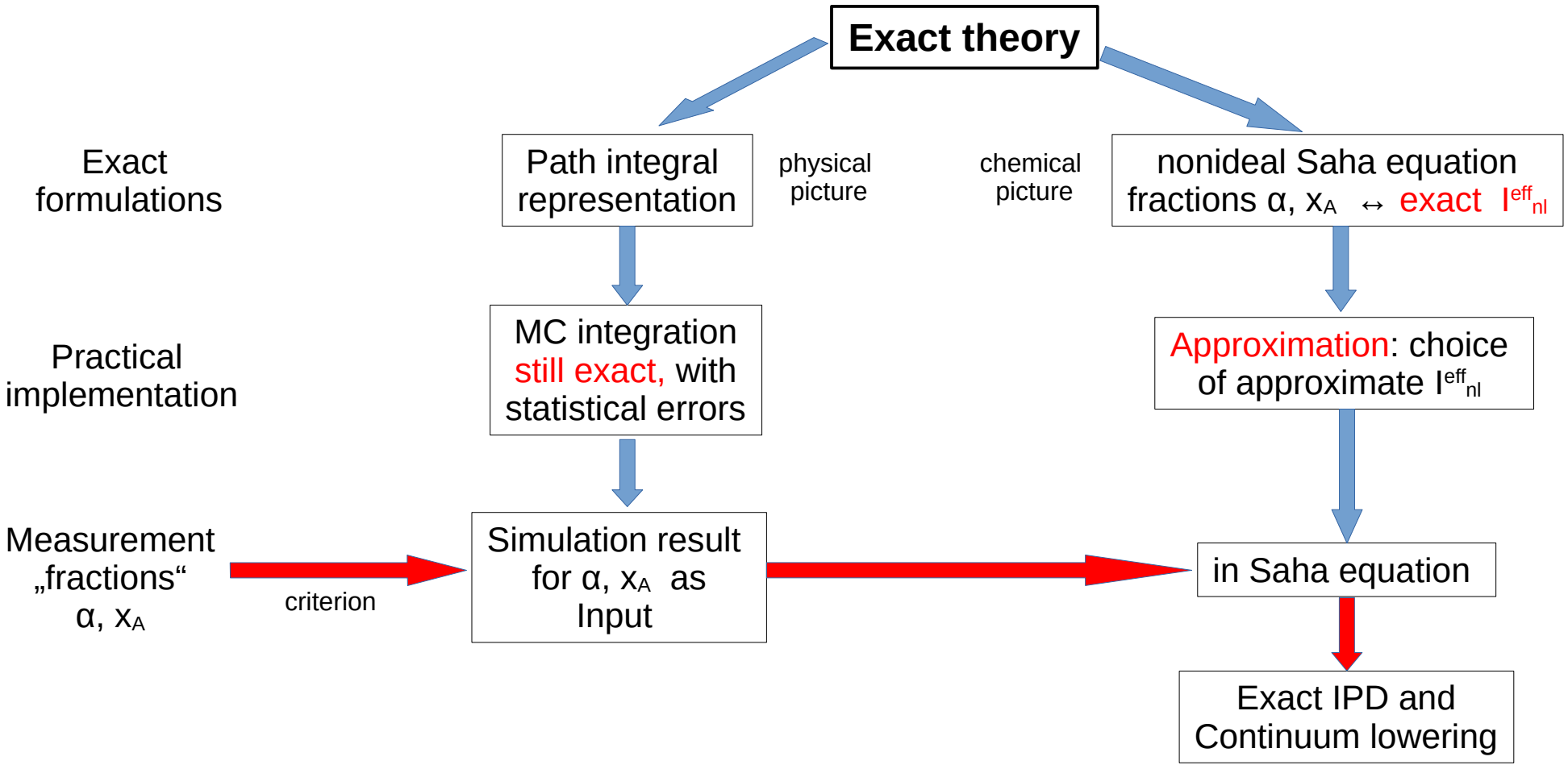
Standard picture of continuum lowering and Ionization potential depression (IPD)*



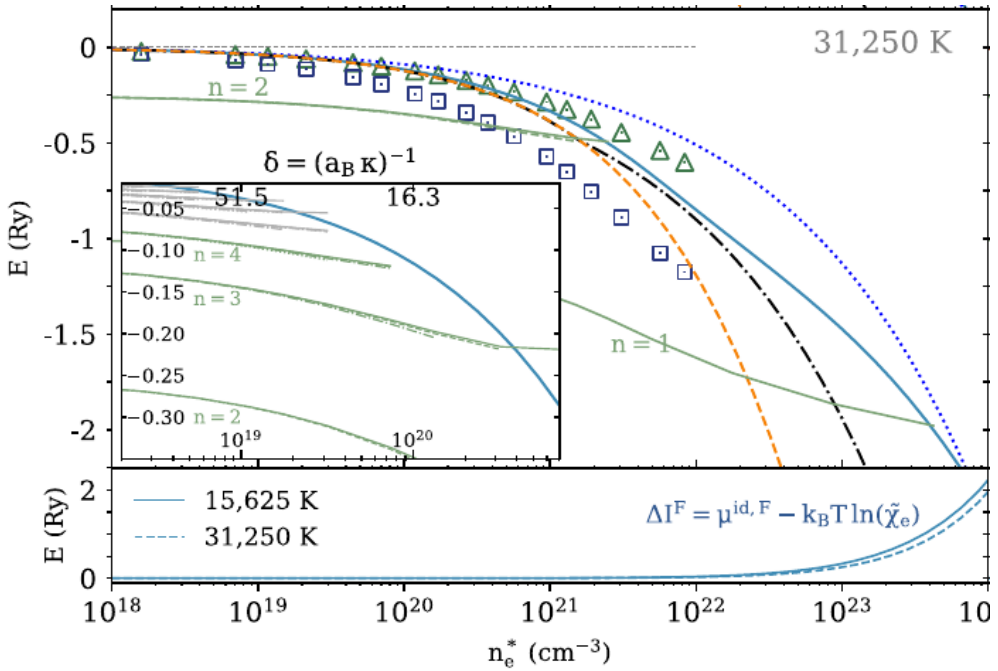
Model input for interaction contributions, yields IPD, Δ_{1s}
Solution of Saha equation yields α, x_A

*M. Bonitz and L. Kordts, CPP (2025)

FPIMC downfolding (5) → Saha equation, IPD



FPIMC downfolding result for hydrogen continuum lowering*



△ FPIMC continuum w/o level shifts

□ FPIMC continuum with level shifts§

Lines:
Debye model (dashes)
Stewart Pyatt (dots)
Ecker/Kröll (dash-dot)

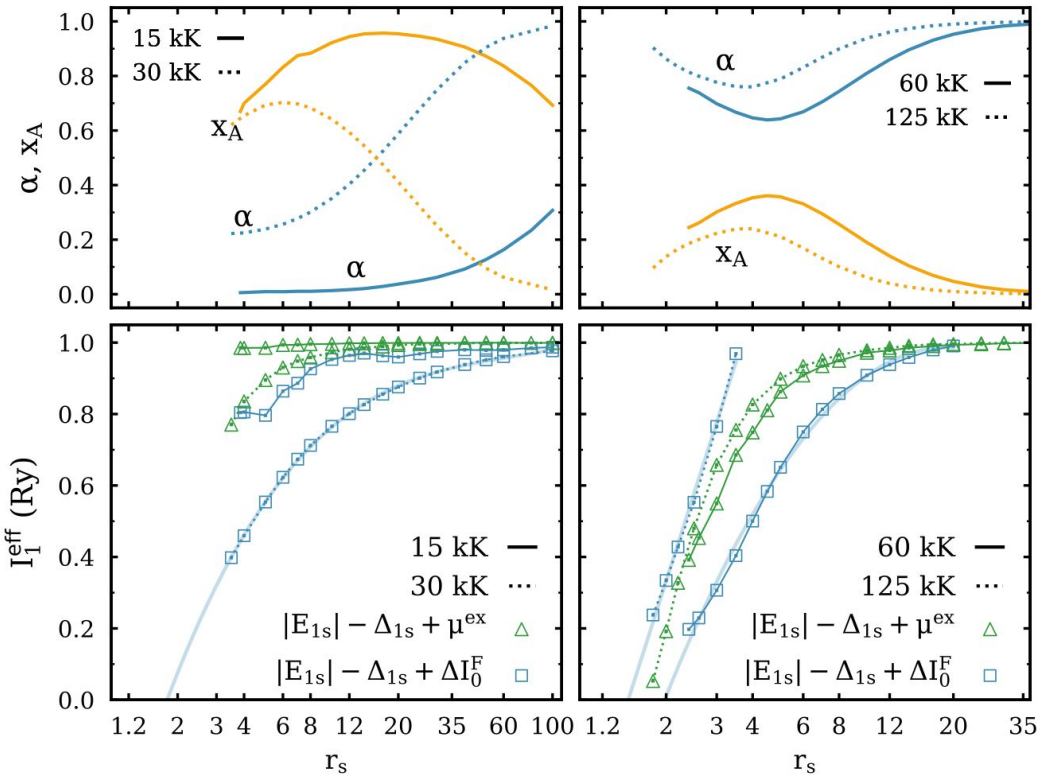
ΔI^F: Fermi barrier

* M. Bonitz *et al.*, Phys. Plasmas 2024
M. Bonitz and L. Kordts, CPP 2025

use FPIMC results for α as input to obtain continuum lowering

§Rogers et al., PRA 1970

FPIMC downfolding result for hydrogen ground state ionization potential*



* M. Bonitz and L. Kordts, CPP 2025

use FPIMC results for continuum lowering and fraction α

→ obtain effective ionization energy I^{eff}_{1s}

□ FPIMC with level shifts

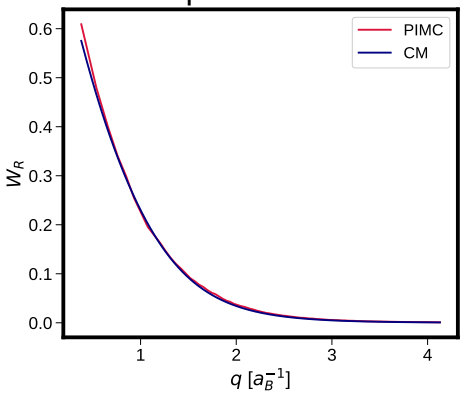
△ Analytical model (Pade formula)

Extrapolation yields Mott density

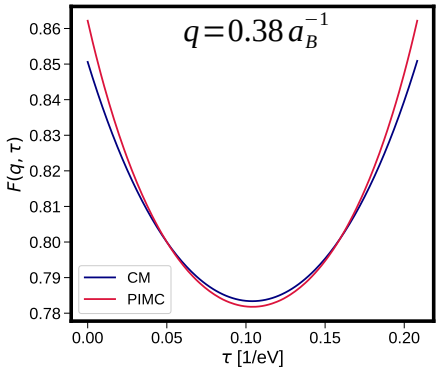
Sensitivity to criteria for α and x_A: 3...5%

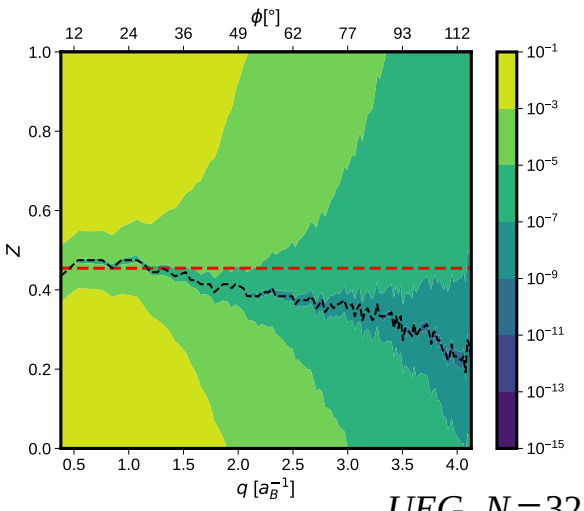
PIMC approach to ionization and IPD from XRTS via Chihara decomposition (Chemical Model)

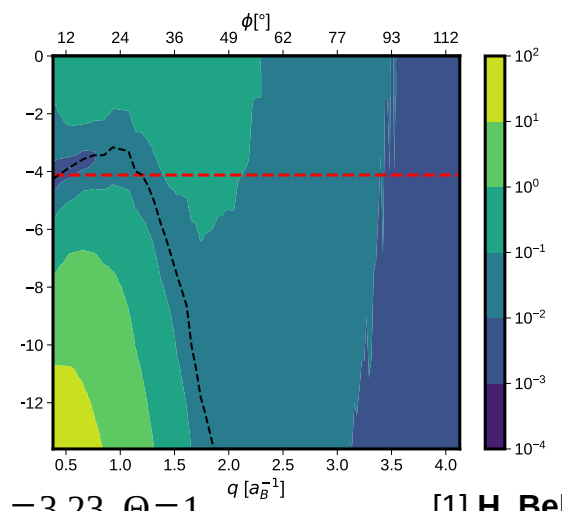
Elastic component



Inlastic component





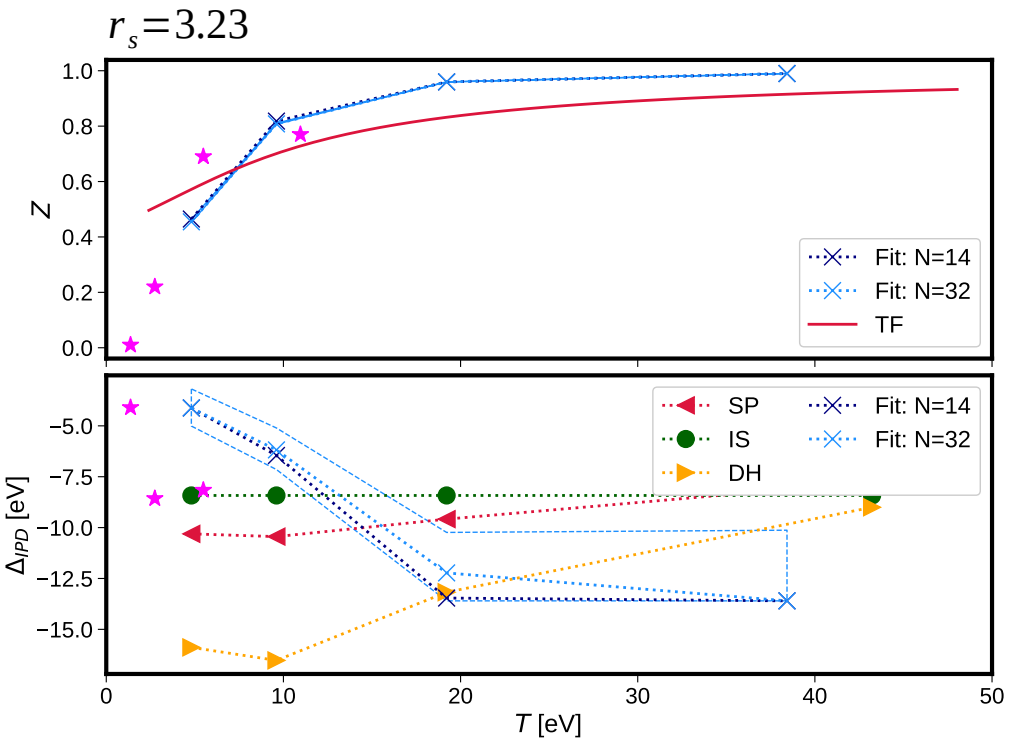


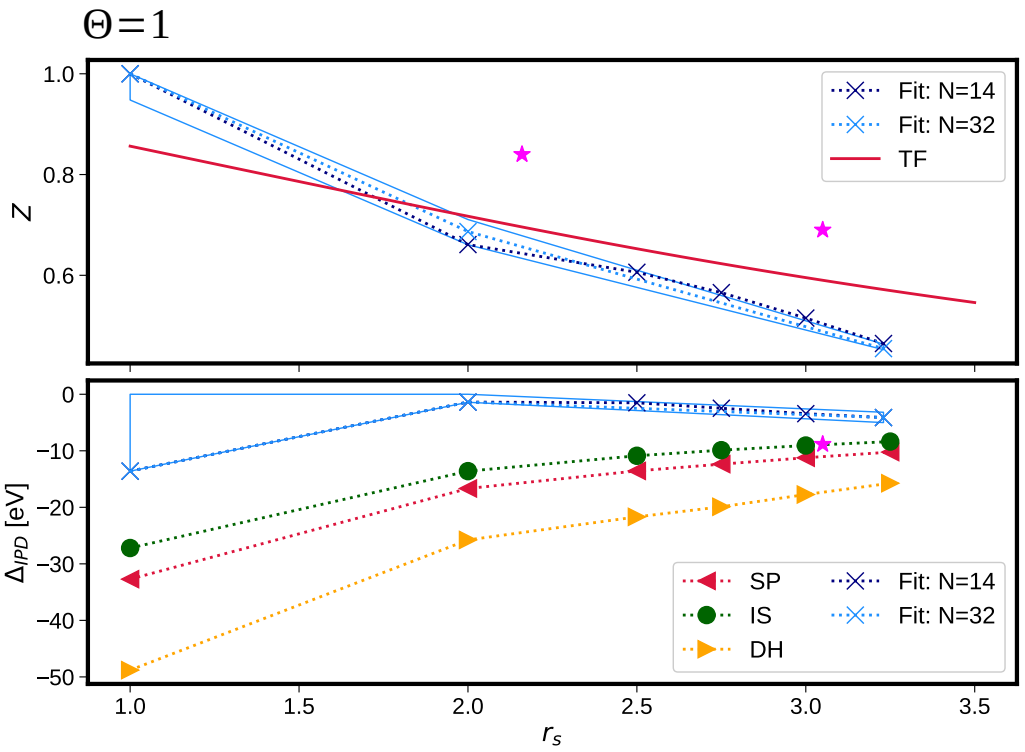
- We enforce a definition for the ionization degree and IPD using the Chihara decomposition (CM) for ab initio PIMC simulations of warm dense hydrogen [1]:
  - Separates ionic, free and bound contributions to the scattering
  - Allows direct comparison to the ITCF obtained from PIMC simulations
- Get ionization from elastic and IPD from inelastic scattering contributions
- Best fits are relatively robust with respect to wavenumber q
- No sensitivity to ionization/IPD at large wavenumbers (single-particle regime)

$UEG, N = 32, r_s = 3.23, \Theta = 1$

[1] **H. Bellenbaum** et al., arXiv:2503.14014 [physics.chem-ph] (2025)

Comparison of extracted estimates against commonly used ionization and IPD models (arXiv:2503.14014)





Top: Comparison of estimates for the ionization degree against the Thomas-Fermi model (TF).
Magenta points: from Filinov and Bonitz, Phys. Rev. E **108**, 055212 (2023).
Bottom: comparison against common IPD models – Stewart-Pyatt (SP), ion-sphere (IS) and Debye-Hückel (DH).
Magenta points: Bonitz and Kordts, arXiv:2502.10548 [physics.plasm-ph] (2025).
Error bars are estimated using a 10% increase of the error function around the minimum.

QMC-based benchmarks: benefit for ICF

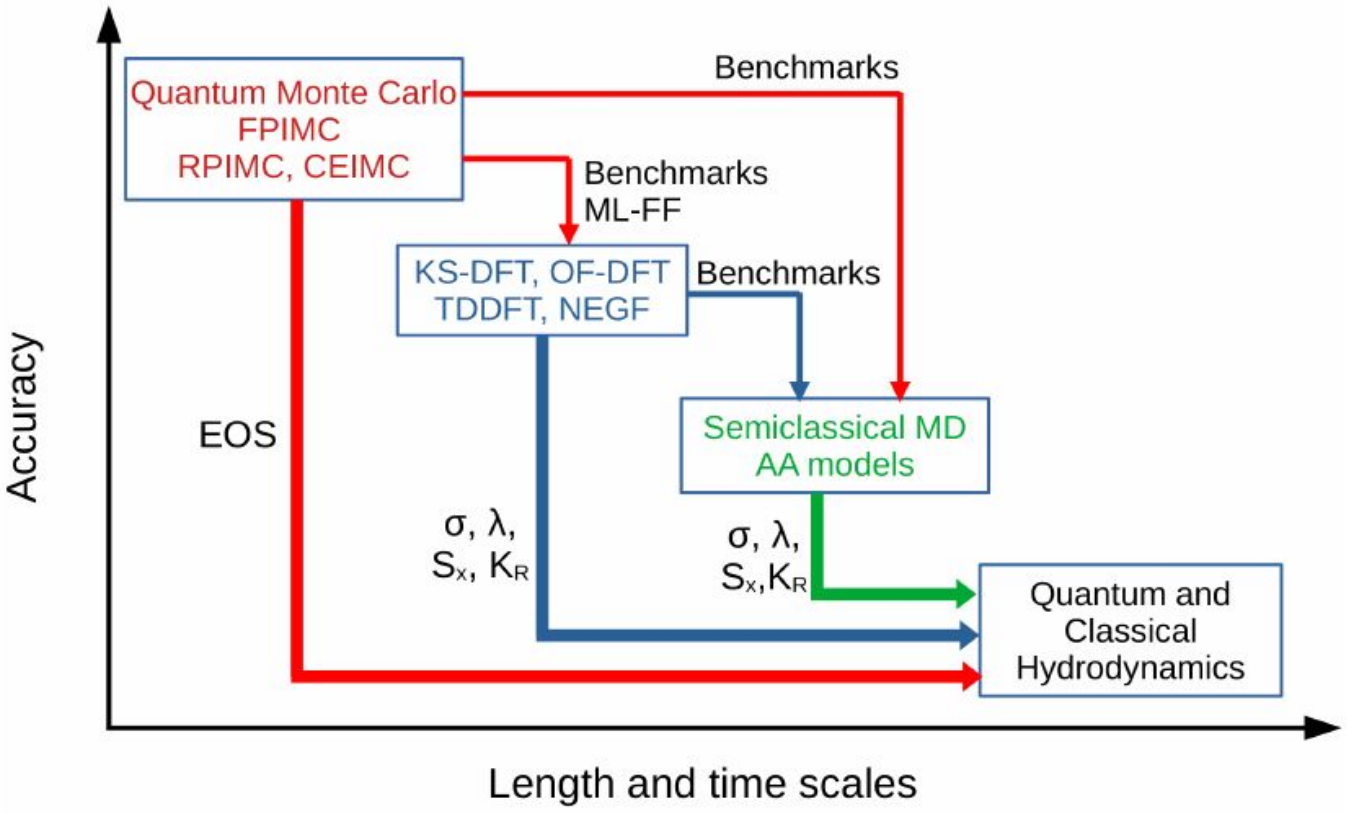
Hydro simulations indispensible to cover relevant scales and complexity

with present approach:

- needed input can be improved substantially

- limitations can be better quantified.

- **predictive simulations will become possible**



* M. Bonitz *et al.*, Phys. Plasmas 2024

Summary

new era of high-precision research in WDM

basis: first principles predictive fermionic PIMC simulations. Can be used
- for benchmarking other methods in limited parameter range (FSP)
- for optimizing choice of approximations for $E_{xc,}$ F_{xc}, Σ etc.
- for smart combination of methods, in particular RPIMC, DFT, Green functions

Summary

new era of high-precision research in WDM

basis: first principles predictive fermionic PIMC simulations. Can be used
- for benchmarking other methods in limited parameter range (FSP)
- for optimizing choice of approximations for $E_{xc,}$ F_{xc}, Σ etc.
- for smart combination of methods, in particular RPIMC, DFT, Green functions

New concept: FPIMC downfolding:

- formally exact methods can be made predictive via FPIMC-based reconstruction of exact $E_{xc,}$ F_{xc}, Σ etc., for a limited parameter range (beyond FPIMC)
- FPIMC results for IPD*
- extend to exact average atom models and exact hydrodynamics

- Needed: extend fermionic PIMC to broader parameter range and materials

*Bonitz, Kordts, CPP 2025; alternative idea: Bellenbaum et al., submitted, arXiv:2503.14014

Thank you for your attention