

Taylor series perspective on *ab initio* path integral Monte Carlo simulations with Fermi-Dirac statistics

Tobias Dornheim^{1,2,*} Alexander Benedix Robles^{2,1,3} Paul Hamann^{1,4,2} Thomas M. Chuna^{2,1} Pontus Svensson^{1,2}
 Sebastian Schwalbe^{1,2} Zhandos A. Moldabekov^{1,2} Panagiotis Tolias⁵ and Jan Vorberger¹

¹*Helmholtz-Zentrum Dresden-Rossendorf (HZDR), D-01328 Dresden, Germany*

²*Center for Advanced Systems Understanding (CASUS), D-02826 Görlitz, Germany*

³*Technische Universität Dresden, D-01062 Dresden, Germany*

⁴*Institut für Physik, Universität Rostock, D-18057 Rostock, Germany*

⁵*Space and Plasma Physics, Royal Institute of Technology (KTH), Stockholm SE-100 44, Sweden*



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The fermion sign problem constitutes a fundamental computational bottleneck across a plethora of research fields in physics, quantum chemistry, and related disciplines. Recently, it has been suggested to alleviate the sign problem in *ab initio* path integral molecular dynamics and path integral Monte Carlo (PIMC) calculations based on the simulation of fictitious identical particles that are represented by a continuous quantum statistics variable ξ [Xiong and Xiong, *J. Chem. Phys.* **157**, 094112 (2022)]. This idea facilitated a host of applications including the interpretation of an x-ray scattering experiment with strongly compressed beryllium at the National Ignition Facility [Dornheim *et al.*, *Nat. Commun.* **16**, 5103 (2025)]. In the present work, we express the original isothermal ξ extrapolation method as a special case of a truncated Taylor series expansion around the $\xi = 0$ limit of distinguishable particles. We derive PIMC estimators that allow us to evaluate the Taylor coefficients up to arbitrary order and we carry out extensive PIMC simulations of the warm dense electron gas to systematically analyze the sign problem from this perspective. This gives us important insights into the applicability of the ξ extrapolation method for different levels of quantum degeneracy in terms of the Taylor series radius of convergence. Moreover, the direct PIMC evaluation of the ξ derivatives, in principle, removes the necessity for simulations at different values of ξ and can facilitate more efficient simulations that are designed to maximize compute time in those regions of the full permutation space that contribute most to the final Taylor estimate of the fermionic expectation value of interest.

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I. INTRODUCTION

For over half a century, *ab initio* quantum Monte Carlo (QMC) methods [1–5] have been employed with great success to study a broad range of quantum many-body systems in both the zero-temperature limit [1,2,5–8] and thermal equilibrium [3,9–20]. The basic idea is usually to cast the expectation value of a given observable of interest $\hat{O}$ in terms of a high-dimensional (with $D \sim 10^{3-7}$ dimensions not being unusual) integral. The associated curse of dimensionality is then avoided with Monte Carlo importance sampling integration, often using a special purpose implementation of the celebrated Metropolis algorithm [21]. At finite temperatures, a particularly successful method is offered by the *ab initio* path integral Monte Carlo (PIMC) approach [22–24], which is based on the well-known *classical isomorphism* [25], where the quantum many-body system of interest is mapped onto an effectively classical system of interacting ring polymers.

Having originally been developed for the simulation of ultracold helium [26,27], PIMC has since given deep insights into important phenomena such as superfluidity [3,28–31] and potential supersolid behavior [32,33], collective and single-particle excitations [17,20,34–38], crystallization [39–42], and entanglement laws [43].

A decisive factor regarding the efficiency of the PIMC approach concerns the quantum statistics that are obeyed by the simulated particles. For bosons and boltzmannons (i.e., hypothetical distinguishable quantum particles), all contributions to the partition function are strictly non-negative. Thus, the corresponding path integral configuration space is ideally suited for Metropolis Monte Carlo sampling, and quasi-exact (within the given statistical Monte Carlo error bars) simulations of up to $N \sim 10^4$ particles have been reported [16,44]. The situation is dramatically changed for fermions, where the antisymmetry of the thermal density matrix under the exchange of particle coordinates induces sign changes in the respective contributions to both the partition function and the observables. The resulting cancellation of positive and negative terms is the root cause of the notorious *fermion sign problem* [45–47], which manifests itself as an exponential increase in the compute time required to attain a specific level of accuracy upon increasing the system size N or decreasing the temperature T . In practice, the sign problem limits

*Contact author: t.dornheim@hzdr.de

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direct PIMC simulations of fermions to moderate system sizes ($N \sim 10^0 - 10^2$) and weak-to-moderate levels of quantum degeneracy [47,48]. This is unfortunate, as degenerate Fermi systems potentially offer a wealth of interesting physics, such as the Bardeen Cooper Schrieffer-Bose Einstein condensate (BCS-BEC) transition in ultracold ^3He [49], roton-type collective modes in Fermi liquids [50–54], and the formation of Wigner molecules or crystals at low density [40,42,55–57]. A particularly important application is given by so-called *warm dense matter* [58], an extreme state that naturally occurs in a host of compact astrophysical objects [59–62], which is also highly relevant for material science [63–65] and inertial confinement fusion experiments [66–69]. This regime is characterized by the complex interplay of Coulomb correlations, thermal excitations, and partial electron degeneracy, making holistic descriptions using state-of-the-art *ab initio* methods such as PIMC paramount [70–72].

The rather pressing need to understand interacting quantum many-fermion systems has sparked a surge of developments in corresponding finite-temperature QMC methodologies [13–15,19,73–91]; see also Refs. [58,71,72] and references therein. A particularly interesting approach is offered by the simulation of fictitious identical particles guided by the continuous quantum statistics variable ξ [80,81,84,86,87,91–100], where the cases of $\xi = -1$, $\xi = 0$, and $\xi = 1$ correspond to the physically relevant cases of Fermi-Dirac, Maxwell-Boltzmann, and Bose-Einstein statistics, respectively. Specifically, Xiong and Xiong [80] have suggested to carry out simulations in the sign-problem free domain of $\xi \geq 0$ and then extrapolate to the fermionic limit of $\xi = -1$ using an empirical quadratic ansatz. Subsequent studies [84,86] have substantiated the high quality of the ξ extrapolation method for weak-to-moderate levels of quantum degeneracy. Importantly, when applicable, this method completely removes the fermionic cancellation problem, facilitating simulations of up to $N = 1000$ electrons [93] as well as the first direct comparison between *ab initio* PIMC simulations and an x-ray scattering measurement on warm dense beryllium performed at the National Ignition Facility (NIF) in Livermore [51,95,101]. Moreover, complete access to the full set of observables (including dynamic information as encoded into the variety of imaginary-time correlation functions [102–106]) is retained, in contrast to other methods (e.g., those relying on nodal restrictions [107]).

In the present work, we report a generalized Taylor series perspective on PIMC simulations of fictitious identical particles, which contains the original, empirical ξ extrapolation technique as a special case. To this end, we derive estimators that allow us to evaluate the Taylor extrapolation to the fermionic limit of $\xi = -1$ (and any other value of ξ) up to arbitrary order from a single PIMC simulation at any reasonable reference value $\xi_{\text{ref}} > 0$. Choosing the boltzmann case of $\xi = 0$ as the basis for the Taylor series, our findings seem to indicate that the physically meaningful cases of $\xi = 1$ and

$\xi = -1$ are indeed within its radius of convergence for the studied cases as far as it can be resolved within the given error bars. Furthermore, the convergence radius appears to approach unity from above upon decreasing the temperature. Crucially, access to the full Taylor series allows us to rigorously assess the convergence with the number of Taylor coefficients and, in this way, to choose appropriate levels of truncation. This, in turn, nicely verifies the quadratic empirical extrapolation that has been employed in previous works [80,84,86,94]. As a practical example, we consider the warm dense uniform electron gas (UEG) [108–110]—the quantum version of the classical one-component plasma—which has attracted a surge of interest over the last decade or so [19,36,77,79,108,111–128] owing to its relevance for a variety of warm dense matter applications, including thermal density functional theory simulations [129–135]. Nevertheless, our methodology and conclusions are very general and expected to be relevant for PIMC simulations of a gamut of interacting Fermi-Dirac systems, including ultracold atoms and real warm dense matter systems comprised of both electrons and ions.

The paper is organized as follows: In Sec. II, we introduce the required theoretical background, starting with the UEG model (Sec. II A), a brief discussion of PIMC (Sec. II B), and the simulation of fictitious identical particles (Sec. II C). This is followed by the introduction of the Taylor series perspective (Sec. II D) and the PIMC estimators for the ξ derivatives (Sec. II E). Section III contains the detailed presentation of our simulation results, including the analysis of the convergence radii (Sec. III A), the study of ξ derivatives (Sec. III B), and the investigation of the practical convergence of the Taylor series at different conditions (Sec. III C). The paper is concluded by a summary and outlook in Sec. IV.

II. THEORY

We assume Hartree atomic units throughout this work.

A. Uniform electron gas model

The UEG, also known as *jellium*, constitutes the quantum version of the classical one-component plasma [136,137]. In the context of this work, we simulate $N = N^\uparrow + N^\downarrow$ *spin-unpolarized* electrons in standard periodic boundary conditions, with $N^\uparrow$ and $N^\downarrow$ the equal numbers of spin-up and spin-down electrons. Following Fraser *et al.* [138], we express the corresponding N -body Hamiltonian as

$$\hat{H}_E = -\frac{1}{2} \sum_{l=1}^N \nabla_l^2 + \sum_{l < k}^N \phi_E(\mathbf{r}_l, \mathbf{r}_k) + \frac{N\xi_M}{2}, \quad (1)$$

where $\xi_M = -2.837297(3/4\pi)^{1/3}N^{-1/3}r_s^{-1}$ is the Madelung constant [12]. The Ewald pair potential $\phi_E(\mathbf{r}_a, \mathbf{r}_b)$ already includes the interaction between the electrons and the homogeneous neutralizing positive background and is given by

$$\phi_E(\mathbf{r}_a, \mathbf{r}_b) = \frac{1}{\Omega} \sum_{\mathbf{G} \neq 0} \frac{\exp(i2\pi \mathbf{G} \cdot (\mathbf{r}_a - \mathbf{r}_b) - \frac{\pi^2 \mathbf{G}^2}{\kappa^2})}{\pi \mathbf{G}^2} - \frac{\pi}{\kappa^2 \Omega} + \sum_{\mathbf{n}} \frac{\text{erfc}(\kappa |\mathbf{r}_a - \mathbf{r}_b + \mathbf{n}L|)}{|\mathbf{r}_a - \mathbf{r}_b + \mathbf{n}L|}, \quad (2)$$

with $\mathbf{n} = (n_x, n_y, n_z)^T$ being an integer vector and where the reciprocal lattice vectors $\mathbf{G}$ have been defined *without* the often included factor of 2π . For completeness, we note that other potentials have been explored in the literature [88,97,139–144], but using the standard Ewald method allows for direct comparisons with other PIMC variants, most notably configuration PIMC (CIPMC) [12,73,74,108,145] at high densities.

The unpolarized UEG is conveniently characterized by two dimensionless parameters [146]: The density parameter $r_s = (3/4\pi n)^{1/3} = d/a_B$ is given by the Wigner-Seitz radius in units of the Bohr radius and serves as the quantum coupling parameter; the reduced degeneracy temperature $\Theta = T/E_F$ is given by the ratio of the thermal energy to the Fermi energy and constitutes a straightforward measure for the degree of quantum degeneracy (with $\Theta \ll 1$ and $\Theta \gg 1$ the fully degenerate and semiclassical limits, respectively). In the warm dense matter regime, which is of considerable interest for astrophysics, planetary science, material science, and inertial fusion energy [58], we have $r_s \sim \Theta \sim 1$, indicating a complex interplay of thermal, quantum, and Coulomb coupling effects.

Originally developed as a model for conduction electrons in simple metals [110,147], the UEG has emerged as the archetypal model system of interacting electrons with a plethora of practical applications. For example, highly accurate parametrizations of UEG properties such as the exchange-correlation (free) energy [112,115,148–150] and linear density response function [117,151–154], based on state-of-the-art QMC simulations [6,108,114,155–160], constitute important input for other calculations, most notably density functional theory simulations of real materials [58,72,161].

In the warm dense matter regime, the extensive body of thermal QMC simulations includes results for the energy, free energy, and chemical potential [12,14,19,73,74,77,79,85,96,114,145,162–165], momentum distribution [120,166–168], linear [116,117,119,126,158,169–171] and nonlinear [102,118,172–178] density response, dynamic structure factor [36,53,179–181], and related dynamic properties [182,183].

B. Path integral Monte Carlo

In the following, we give a concise introduction to certain aspects of the PIMC method that are relevant for the present investigation. More detailed introductions to PIMC can be found, e.g., in Refs. [3,16,22,24,184].

Throughout this work, we consider N particles in a cubic simulation cell of volume $\Omega = L^3$ in the canonical ensemble, meaning that N , Ω , and the inverse temperature $\beta = 1/T$ are fixed. On the most abstract level, the path integral form of the canonical partition function Z can be expressed as

$$Z_\xi(\beta, N, \Omega) = \int d\mathbf{X} \underbrace{W(\mathbf{X})^\xi}_{W_\xi(\mathbf{X})} \xi^{N_{pp}(\mathbf{X})}, \quad (3)$$

where the metavariable $\mathbf{X} = (\mathbf{R}_0, \dots, \mathbf{R}_{P-1})^T$ contains the coordinates of all particles on all P imaginary time slices, with $\mathbf{R}_\alpha = (\mathbf{r}_1, \dots, \mathbf{r}_N)^T$ being the N -particle coordinate on

time slice α at $\tau = -i\alpha\epsilon$ where $\epsilon = \beta/P$ is the imaginary time step. We note that it holds $\mathbf{R}_0 \equiv \mathbf{R}_P$; the paths are, hence, closed, giving rise to their interpretation as ring polymers [25], with $W(\mathbf{X})$ being their associated configuration weight containing both kinetic and potential terms; it is a function that can straightforwardly be evaluated in practice. The notation $\int d\mathbf{X}$ in Eq. (3) implies that we have to integrate over all $3NP$ dimensions of the metavariable $\mathbf{X}$ and, in addition, also includes the sum over all possible permutations of particle coordinates of particles of the same spin orientation (i.e., spin-up and spin-down electrons for the unpolarized UEG). Finally, the aforementioned variable ξ takes into account quantum statistics, with $\xi = -1, 0, 1$ corresponding to fermions, boltzmannions, and bosons, respectively, and $N_{pp}(\mathbf{X})$ being the corresponding number of pair permutations. For the purposes of the present work, any $\xi \in \mathbb{R}$ constitutes a valid option. The basic idea of the PIMC method is to use a modern implementation of the Metropolis algorithm [16,167,185] to stochastically generate a Markov chain $\{\mathbf{X}\}_i$ that is distributed according to $P_\xi(\mathbf{X}) = W_\xi(\mathbf{X})/Z_\xi$; we note that the arguments (β, N, Ω) will be suppressed for simplicity throughout the remainder of this work. The corresponding Monte Carlo estimate for the thermodynamic equilibrium expectation value of an arbitrary observable $\hat{O}$ is

$$\langle \hat{O} \rangle_\xi = \frac{1}{Z_\xi} \int d\mathbf{X} W_\xi(\mathbf{X}) O(\mathbf{X}), \quad (4)$$

where $O(\mathbf{X})$ denotes the corresponding Monte Carlo estimator.

Unfortunately, both $P_\xi(\mathbf{X})$ and $W_\xi(\mathbf{X})$ can be either positive or negative for $\xi < 0$, which is, respectively, the case when N_{pp} is even or odd. This precludes their interpretation as a probability distribution, requiring a further intermediate step. As a practical workaround, we sample path configurations $\mathbf{X}$ according to the modified probability distribution $P'_\xi(\mathbf{X}) = |W_\xi(\mathbf{X})|/Z'_\xi = P_{|\xi|}(\mathbf{X})$, and the exact signful expectation value is then given by

$$\langle \hat{O} \rangle_{\xi < 0} = \frac{\langle \hat{O} \hat{S} \rangle_{|\xi|}}{\langle \hat{S} \rangle_{|\xi|}}, \quad (5)$$

where the numerator and denominator are computed as a Monte Carlo average with respect to the absolute value of ξ . We note that $S \equiv \langle \hat{S} \rangle_{|\xi|}$, with $S(\mathbf{X}) = |W_\xi(\mathbf{X})|/W_\xi(\mathbf{X})$, is known as the *average sign* in the literature and constitutes a direct measure for the degree of cancellations within the PIMC simulation for fermionic observables. Fermionic PIMC simulations are generally feasible for $S \gtrsim 10^{-2} - 10^{-3}$ [47].

C. Fictitious identical particles and reweighting

In their seminal work, Xiong and Xiong [80] have proposed to carry out path integral molecular dynamics simulations [82,186] for continuous non-negative values of ξ and then subsequently extrapolate to the fermionic limit of $\xi = -1$ using the empirical parabolic ansatz

$$O_\xi = a_0 + b_0 \xi + c_0 \xi^2, \quad (6)$$

with a_0 , b_0 , and c_0 free fit parameters. Usually, simulations have been performed on a grid with $\xi \in [0, 1]$ although

alternative intervals $\xi \in [\xi_{\min}, \xi_{\max}]$ are certainly possible. While choosing $\xi_{\min} > 0$ makes no practical sense, it seems worthwhile to attempt to choose $\xi_{\min}$ as close to $\xi = -1$ as possible to explicitly capture more of the fermionic sector; this can be possible even in situations where the full fermion sign problem is rather severe as the average sign increases exponentially upon decreasing $|\xi|$ in the fermionic sector of $\xi < 0$ [84,96]. Similarly, one might choose $\xi_{\max}$ larger or smaller than unity. A more systematic answer to this question can be given in terms of the radius of convergence of the Taylor series [see Eqs. (14) and (15)].

Clearly, PIMC simulations for $\xi \geq 0$ are not subject to the exponential computation bottleneck inherent to the

fermion sign problem. Having to perform $N_\xi = 5\text{--}20$ independent PIMC simulations for different values of $\xi \geq 0$ thus appears to be a reasonable trade-off to extend the capabilities of standard PIMC to larger system sizes. At the same time, simulations of $N \sim \mathcal{O}(10^2)$ particles on $P \sim \mathcal{O}(10^2)$ imaginary-time slices can be computationally demanding even without the sign problem. To avoid this obstacle, Dornheim *et al.* [91] have recently presented a reweighting estimator, which, in principle, allows the evaluation of the full ξ dependence from a single PIMC simulation at an arbitrary reference value ξ_{ref} . Specifically, we can express the expectation value of an observable $\hat{O}$ at any ξ in terms of its expectation values at ξ_{ref} ,

$$\langle \hat{O} \rangle_\xi = \frac{Z_{\xi_{\text{ref}}}}{Z_\xi} \frac{1}{Z_{\xi_{\text{ref}}}} \sum_{\sigma \in S_N} \int d\mathbf{X} W_{\xi_{\text{ref}}}(\mathbf{X}) \underbrace{\frac{W_\xi(\mathbf{X})}{W_{\xi_{\text{ref}}}(\mathbf{X})} O(\mathbf{X})}_{O_{\xi_{\text{ref}},\xi}(\mathbf{X})} = \frac{Z_{\xi_{\text{ref}}}}{Z_\xi} \langle \hat{O}_{\xi_{\text{ref}},\xi} \rangle_{\xi_{\text{ref}}}, \quad (7)$$

$$\frac{Z_\xi}{Z_{\xi_{\text{ref}}}} = \frac{1}{Z_{\xi_{\text{ref}}}} \sum_{\sigma \in S_N} \int d\mathbf{X} W_{\xi_{\text{ref}}}(\mathbf{X}) \frac{W_\xi(\mathbf{X})}{W_{\xi_{\text{ref}}}(\mathbf{X})} = \left\langle \frac{W_\xi(\mathbf{X})}{W_{\xi_{\text{ref}}}(\mathbf{X})} \right\rangle_{\xi_{\text{ref}}}, \quad (8)$$

leading to the estimator

$$\langle \hat{O} \rangle_\xi = \frac{\langle \hat{O}_{\xi_{\text{ref}},\xi} \rangle_{\xi_{\text{ref}}}}{\left\langle \frac{W_\xi(\mathbf{X})}{W_{\xi_{\text{ref}}}(\mathbf{X})} \right\rangle_{\xi_{\text{ref}}}}, \quad (9)$$

with the ratio of configuration weights given by

$$\frac{W_\xi(\mathbf{X})}{W_{\xi_{\text{ref}}}(\mathbf{X})} = \left(\frac{\xi}{\xi_{\text{ref}}} \right)^{N_{\text{pp}}}. \quad (10)$$

In essence, Eq. (9) constitutes a reweighting estimator, where measurements of the estimator $O(\mathbf{X})$ are weighted by Eq. (10) and then renormalized by Eq. (8).

D. Taylor series and extrapolation

Within the reasonable assumption that the O_ξ expectation value of an observable $\hat{O}$ constitutes a smooth, continuous and infinitely differentiable function with respect to ξ at a reference real number ξ_T , as it has been verified empirically in recent PIMC investigations [80,84,91], we can express O_ξ as a Taylor series around the reference value ξ_T ,

$$O(\xi) = O_{\infty,\xi_T}(\xi) = \sum_{v=0}^{\infty} \frac{(\xi - \xi_T)^v}{v!} \frac{\partial^v}{\partial \xi^v} O_\xi \bigg|_{\xi=\xi_T}. \quad (11)$$

In practice, the Taylor series would need to be truncated at a polynomial degree p ,

$$O_{p,\xi_T}(\xi) = \sum_{v=0}^p \frac{(\xi - \xi_T)^v}{v!} \frac{\partial^v}{\partial \xi^v} O_\xi \bigg|_{\xi=\xi_T}. \quad (12)$$

It is apparent that the standard ξ extrapolation method, based on the empirical parabolic fit formula Eq. (6), corresponds to the special case of $O_{2,0}(\xi)$.

Heuristically, it makes sense to choose $\xi_T = 0$, which minimizes the extrapolation interval from the bosonic sector to the fermionic limit of interest. Then, the truncated version $O_{p,0}(\xi)$

is automatically expressed as a canonical polynomial,

$$O_{p,0}(\xi) = \sum_{v=0}^p c_v \xi^v, \quad (13)$$

which, among other things, lends itself for a fit to PIMC results for O_ξ in the sign-problem free domain of $\xi \geq 0$. Naively, it might seem beneficial to consider a ξ interval with $\xi_{\max} \gg 1$ in order to extract as many coefficients c_v as possible. In this regard, however, it is important to recall the *a priori* unknown radius of convergence r of Eq. (11). In general, a Taylor series converges within the radius r if one of two limits exists [187]

$$r = \lim_{v \rightarrow \infty} \frac{1}{\sqrt[v]{|c_v|}}, \quad (14)$$

$$r = \lim_{v \rightarrow \infty} \left| \frac{c_v}{c_{v+1}} \right|. \quad (15)$$

In practice, the root test of Eq. (14) will be considered throughout this investigation, since the ratio test of Eq. (15) is rendered ineffective by the oscillating nature of the c_v coefficients at strong degeneracy. A polynomial fit according to Eqs. (12) and (13) outside of $\xi \in [0, r)$ (where we again limit ourselves to the bosonic sector) will thus not yield any information that is pertinent to the attempted extrapolation to $\xi = -1$. In fact, it is not obvious to the present authors that the circle of convergence of $O_{p,0}(\xi)$ must include the physical limits of $\xi = \pm 1$. However, we empirically verify this based on extensive PIMC results in Sec. III.

E. PIMC estimation of ξ derivatives

Aiming to derive estimators for the ξ derivatives of $\langle \hat{O} \rangle_\xi$, we introduce the convenient shorthand notation

$$\langle \hat{O} \rangle_\xi = \frac{1}{Z_\xi} \int d\mathbf{X} W(\mathbf{X}) O(\mathbf{X}) \xi^{N_{\text{pp}}} = \frac{O_\xi}{Z_\xi}, \quad (16)$$

where O_ξ denotes the un-normalized $\hat{O}$ expectation value at ξ . We first consider the case of $\xi \neq 0$, for which

$$\frac{\partial O_\xi}{\partial \xi} = \oint W(\mathbf{X})_\xi^{N_{pp}(\mathbf{X})} \frac{O(\mathbf{X})N_{pp}(\mathbf{X})}{\xi}, \quad (17)$$

$$\frac{\partial Z_\xi}{\partial \xi} = \oint W(\mathbf{X})_\xi^{N_{pp}(\mathbf{X})} \frac{N_{pp}(\mathbf{X})}{\xi}. \quad (18)$$

Combining Eqs. (17) and (18) with the quotient rule yields

$$\frac{\partial \langle \hat{O} \rangle_\xi}{\partial \xi} = \frac{\langle \hat{O} \hat{N}_{pp} \rangle_\xi - \langle \hat{O} \rangle_\xi \langle \hat{N}_{pp} \rangle_\xi}{\xi}. \quad (19)$$

Evidently, Eq. (19) can directly be used to compute the ξ derivatives of any expectation value, including $\langle \hat{O} \hat{N}_{pp} \rangle_\xi$ and $\langle \hat{N}_{pp} \rangle_\xi$ that are needed for the computation of the higher-order derivatives. In this way, Eq. (19) serves as a generator of derivatives of arbitrary order. We find

$$\frac{\partial^2 \langle \hat{O} \rangle_\xi}{\partial \xi^2} = \frac{1}{\xi^2} \{ \langle \hat{O} \rangle_\xi (\langle \hat{N}_{pp} \rangle_\xi + 2 \langle \hat{N}_{pp}^2 \rangle_\xi - \langle \hat{N}_{pp}^2 \rangle_\xi) - \langle \hat{O} \hat{N}_{pp} \rangle_\xi (2 \langle \hat{N}_{pp} \rangle_\xi + 1) + \langle \hat{O} \hat{N}_{pp}^2 \rangle_\xi \}, \quad (20)$$

$$\begin{aligned} \frac{\partial^3 \langle \hat{O} \rangle_\xi}{\partial \xi^3} = \frac{1}{\xi^3} \{ & \langle \hat{O} \rangle_\xi (2 \langle \hat{N}_{pp} \rangle_\xi + 6 \langle \hat{N}_{pp}^2 \rangle_\xi - 3 \langle \hat{N}_{pp}^2 \rangle_\xi + 6 \langle \hat{N}_{pp}^3 \rangle_\xi - 6 \langle \hat{N}_{pp} \rangle_\xi \langle \hat{N}_{pp}^2 \rangle_\xi + \langle \hat{N}_{pp}^3 \rangle_\xi) \\ & - \langle \hat{O} \hat{N}_{pp} \rangle_\xi (2 + 6 \langle \hat{N}_{pp} \rangle_\xi + 6 \langle \hat{N}_{pp}^2 \rangle_\xi - 3 \langle \hat{N}_{pp}^2 \rangle_\xi) + \langle \hat{O} \hat{N}_{pp}^2 \rangle_\xi 3(1 + \langle \hat{N}_{pp} \rangle_\xi) - \langle \hat{O} \hat{N}_{pp}^3 \rangle_\xi \}. \end{aligned} \quad (21)$$

We then consider the case where $\xi = 0$, which is of the highest interest for the Taylor extrapolation to the fermionic limit of $\xi = -1$ discussed above. It is straightforward to obtain

$$\left. \frac{\partial^\nu}{\partial \xi^\nu} Z_\xi \right|_{\xi=0} = \nu! \oint_{N_{pp} \equiv \nu} d\mathbf{X} W(\mathbf{X}) =: Z(\nu) \nu!, \quad (22)$$

$$\left. \frac{\partial^\nu}{\partial \xi^\nu} O_\xi \right|_{\xi=0} = \nu! \oint_{N_{pp} \equiv \nu} d\mathbf{X} W(\mathbf{X}) O(\mathbf{X}) = Z(\nu) \langle \hat{O} \rangle_{N_{pp} \equiv \nu} \nu!, \quad (23)$$

where $\oint_{N_{pp} \equiv \nu} d\mathbf{X}$ implies that only path-integral configurations with $N_{pp} = \nu$ contribute. Equation (16) lends itself to an implementation of the generalized quotient rule [188]

$$\frac{\partial^\nu}{\partial \xi^\nu} \left(\frac{O_\xi}{Z_\xi} \right) = \sum_{\kappa=0}^{\nu} \binom{\nu}{\kappa} O_\xi^{(\kappa)} \frac{(-1)^{\nu-\kappa}}{Z_\xi^{(\nu-\kappa+1)}} B_{\nu-\kappa}(Z_\xi^{(1)}, Z_\xi^{(2)}, \dots, Z_\xi^{(\nu-\kappa)}), \quad (24)$$

with $B_{\nu-\kappa}(\dots)$ the complete Bell polynomials and $Z_\xi^{(a)}$ being the a th derivative with respect to ξ . In practice, we can estimate $Z(\nu)$ and $\langle \hat{O} \rangle_{N_{pp} \equiv \nu}$, up to a proportionality factor that cancels in view of Eq. (24), by only measuring observables in our PIMC simulation when $N_{pp} = \nu$,

$$\frac{Z(\nu)}{Z_{\text{tot}}} = \langle \hat{\delta}_{N_{pp}, \nu} \rangle_{\text{tot}} \langle \hat{O} \rangle_{N_{pp} \equiv \nu} = \frac{Z_{\text{tot}}}{Z(\nu)} \langle \hat{O} \hat{\delta}_{N_{pp}, \nu} \rangle_{\text{tot}}, \quad (25)$$

with $\langle \dots \rangle_{\text{tot}}$ and Z_{tot} indicating the full configuration space that can contain paths with any value of N_{pp} . For the first three orders, Eq. (24) gives

$$\left. \frac{\partial \langle \hat{O} \rangle_\xi}{\partial \xi} \right|_{\xi=0} = \frac{Z_{N_{pp} \equiv 1}}{Z_{N_{pp} \equiv 0}} \{ \langle \hat{O} \rangle_{N_{pp} \equiv 1} - \langle \hat{O} \rangle_{N_{pp} \equiv 0} \}, \quad (26)$$

$$\left. \frac{\partial^2 \langle \hat{O} \rangle_\xi}{\partial \xi^2} \right|_{\xi=0} = 2 \left\{ \frac{Z_{N_{pp} \equiv 2}}{Z_{N_{pp} \equiv 0}} (\langle \hat{O} \rangle_{N_{pp} \equiv 2} - \langle \hat{O} \rangle_{N_{pp} \equiv 0}) - \left(\frac{Z_{N_{pp} \equiv 1}}{Z_{N_{pp} \equiv 0}} \right)^2 (\langle \hat{O} \rangle_{N_{pp} \equiv 1} - \langle \hat{O} \rangle_{N_{pp} \equiv 0}) \right\}, \quad (27)$$

$$\begin{aligned} \left. \frac{\partial^3 \langle \hat{O} \rangle_\xi}{\partial \xi^3} \right|_{\xi=0} = & 6 \left\{ \frac{Z_{N_{pp} \equiv 3}}{Z_{N_{pp} \equiv 0}} (\langle \hat{O} \rangle_{N_{pp} \equiv 3} - \langle \hat{O} \rangle_{N_{pp} \equiv 0}) + \left(\frac{Z_{N_{pp} \equiv 1}}{Z_{N_{pp} \equiv 0}} \right)^3 (\langle \hat{O} \rangle_{N_{pp} \equiv 1} - \langle \hat{O} \rangle_{N_{pp} \equiv 0}) \right\} \\ & - \left(\frac{Z_{N_{pp} \equiv 2}}{Z_{N_{pp} \equiv 0}} \right) \left(\frac{Z_{N_{pp} \equiv 1}}{Z_{N_{pp} \equiv 0}} \right) \{ 6 \langle \hat{O} \rangle_{N_{pp} \equiv 2} + 6 \langle \hat{O} \rangle_{N_{pp} \equiv 1} - 12 \langle \hat{O} \rangle_{N_{pp} \equiv 0} \}. \end{aligned} \quad (28)$$

We conclude the theoretical background with a concise summary of the structure of the resulting Taylor extrapolation around $\xi_T = 0$ to the fermionic limit of $\xi = -1$. First, evaluating the Taylor series up to an order p involves path

configurations with up to $N_{pp} = p$ pair permutations. These path configurations can, in general, be comprised of a variety of combinations of permutation cycles and topologies [189–191]. For a spin-unpolarized system, the maximum

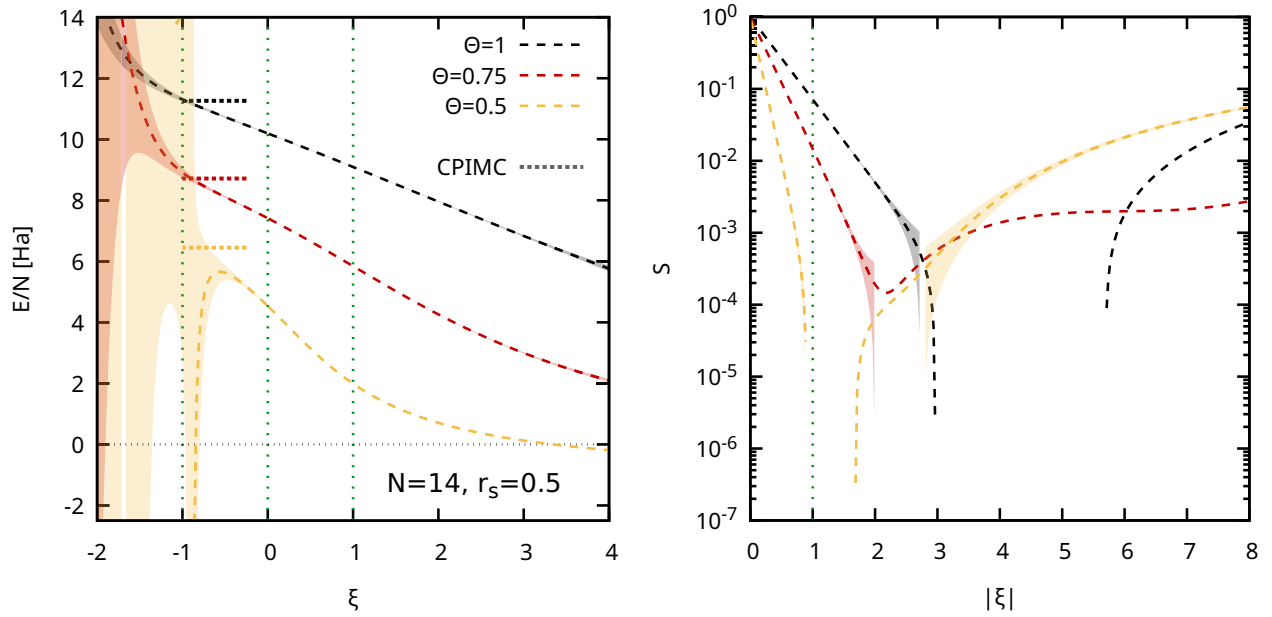


FIG. 1. Dependence of the total energy (per particle) (left) and the average sign S (right) on the fictitious quantum statistics variable ξ for the unpolarized UEG at $r_s = 0.5$ with $N = 14$ electrons. Results are shown for $\Theta = 1$ (black), $\Theta = 0.75$ (red), and $\Theta = 0.5$ (gold). The shaded areas indicate the corresponding uncertainty, the horizontal dotted lines correspond to highly accurate CPIMC reference results for $\xi = -1$, and the vertical dotted green lines indicate the physically meaningful cases of $\xi = -1$ (Fermi-Dirac), $\xi = 0$ (Maxwell-Boltzmann), and $\xi = 1$ (Bose-Einstein). For each quantum degeneracy, the entire depicted ξ dependence has been obtained from a single PIMC simulation at a reference value of $\xi_{\text{ref}} = 1$ using the reweighting estimator [Eq. (9)] [91].

possible number of pair exchanges is given by $N_{\text{pp}}^{\text{max}} = 2(N/2 - 1)$; nevertheless, derivatives of the order $\nu > N_{\text{pp}}^{\text{max}}$ do not vanish, since terms with $N_{\text{pp}} < \nu$ contribute to Eq. (24). Finally, we note that the evaluation of Eqs. (22) and (23), which is the only PIMC expectation value required to evaluate the Taylor series, does not involve any fermionic cancellation and is, thus, formally sign-problem free. Unfortunately, the generalized quotient rule [Eq. (24)] does involve cancellations between these expectation values [this can be discerned already in Eqs. (26)–(28)]. As we shall see in Sec. III, this reintroduces the original fermion sign problem in the limit of $p \rightarrow \infty$. Yet, a reasonable and often well justified truncation of the Taylor series at a finite $p \lesssim 3$ can produce reliable results without the full cancellation problem as we will demonstrate empirically in the following.

III. RESULTS

We use a canonical adaption [167] of the worm algorithm by Boninsegni *et al.* [16,44] as it has been implemented into the open-source ISHTAR code [192]. A repository containing all PIMC results is available online [193]. CPIMC [12,73] results have been obtained using the open-source Julia implementation CPIMC.jl [194].

A. General results: dependence on ξ and radius of convergence

As a starting point, we investigate the dependence of different observables on the quantum statistics variable ξ for the UEG at $r_s = 0.5$ in Fig. 1. These conditions can be realized in compression experiments, e.g., at the NIF [195,196], and play an important role in inertial fusion energy applications

[58]. Furthermore, due to the relatively weak coupling, highly accurate CPIMC [12,74] reference results are available as a rigorous benchmark for any attempted Taylor series extrapolation. Finally, these conditions were also considered by Dornheim *et al.* [84], who found that the original ξ extrapolation guided by Eq. (6) breaks down for $\Theta = 0.5$. Below, we will analyze this breakdown from the perspective of the full Taylor series expansion introduced in Sec. II D.

All results shown in Fig. 1 have been obtained for $N = 14$ particles, and the black, red, and golden curves correspond to $\Theta = 1$, $\Theta = 0.75$, and $\Theta = 0.5$, respectively. Specifically, we use the reweighting estimator from Eq. (9) with a reference value of $\xi_{\text{ref}} = 1$ [91]. The shaded areas represent the statistical uncertainty that we estimate from the ratio estimate given by Hatano [197]; we have compared these error estimates with a standard jackknife procedure [198] for selected cases and found them to be indistinguishable. The vertical dotted green lines indicate the physically relevant cases of fermions, boltzmannons, and bosons.

The left panel shows results for the ξ dependence of the total energy per particle, and the horizontal dotted lines represent quasi-exact CPIMC data (with the associated statistical errors being smaller than the width of the lines). For $\Theta = 1$ and $\Theta = 0.75$, the direct PIMC method is capable of providing accurate results for the fermionic limit of $\xi = -1$, which nicely agree with the CPIMC results within the error intervals. In stark contrast, the error interval diverges for $\xi \rightarrow -1$ for $\Theta = 0.5$. This is a direct consequence of the average sign S vanishing within the given Monte Carlo error bars (see the right panel of Fig. 1). At a first glance, the dependence of S on ξ over the full depicted ξ range appears to be counterintuitive and nontrivial. Starting at $|\xi| = 0$, we find the usual

monotonic decay of the sign that appears to follow the expected exponential behavior [96] for all three temperatures. At some point, the sign vanishes within error bars; unfortunately, this happens before the fermionic limit of $\xi = -1$ for $\Theta = 0.5$ and for somewhat larger $|\xi|$ for the two higher values of Θ . For even larger $|\xi|$, the sign starts to increase again. This is a direct consequence of the function of the ξ term as an N_{pp} -dependent weight in the partition function [cf. Eq. (3)]. For $|\xi| \gg 1$, configurations with large N_{pp} are actually being favored in the PIMC generated Markov chain, eventually leading to an exclusive sampling of paths with the maximum possible number $N_{pp}^{\max}$. In this limit (which is always associated with a positive sign for spin-unpolarized systems), there is no more cancellation between adjacent permutation sectors, and the simulation will be sign-problem free for both positive and negative values of ξ .

Returning to the ξ dependence of the total energy per particle in the left panel of Fig. 1, we observe an increasingly steep dependence of $E(\xi)$ on ξ for $\xi \in [-1, 1]$ with decreasing temperatures, reflecting the increasing dissimilarity between the corresponding bosonic and fermionic limits as the impact of quantum statistics becomes more important. Moreover, the dependence on ξ appears to be almost linear in this range for $\Theta = 1$ and still relatively simple for $\Theta = 0.75$. In contrast, the exact CPIMC result for $\xi = -1$ implies the presence of an inflection point around $\xi \approx -0.7$ for $\Theta = 0.5$, thus resulting in a substantially more complicated dependence of E on ξ . In the context of the present work, a crucial question is thus if it is even theoretically possible to use a Taylor series to go beyond the inflection point and reach $\xi = -1$ even in the limit of $p \rightarrow \infty$.

To answer this question, we have repeated the above simulations for $N = 8$ electrons, for which the fermionic limit of $\xi = -1$ can be reached within the limit of feasible computational effort, and we find an average sign of $S = 0.00359(2)$ for $\Theta = 0.5$. The results are shown in Fig. 2, and the top panel shows the interaction energy per particle as a function of ξ with the same color code as in Fig. 1. Equivalent conclusions can be drawn from the total energy and kinetic energy, but the interaction energy is particularly convenient for our purposes as its Monte Carlo estimator has an intrinsically lower variance [199]. Interestingly, we find a polelike phenomenon for all three curves for different values of $|\xi|$. These “pseudodivergencies” can be directly traced back to the average sign vanishing within error bars (see the bottom panel of Fig. 2). For completeness, we note that the existence of an actual pole, or, equivalently, of a truly vanishing sign fundamentally cannot be resolved with PIMC: The only conclusion that we can draw is that the average sign is definitely smaller than our statistical error bars; resolving an actual pole with $S \equiv 0$ would, by definition, require an infinite amount of compute time. Since the average sign in direct PIMC is just the ratio of fermionic and bosonic partition functions, $S = Z_\xi/Z_{|\xi|}$ [47], a vanishing sign would be connected with a zero in Z_ξ as $Z_{|\xi|}$ is always larger than zero. This phenomenon has recently been explored by He *et al.* [200]. In particular, these authors extended the domain of definition of ξ from the real axis to the complex plane and studied the distribution of the complex roots of Z_ξ , which translate to complex poles of $\langle \hat{O} \rangle_\xi$. When these zeros approach the fermionic sector

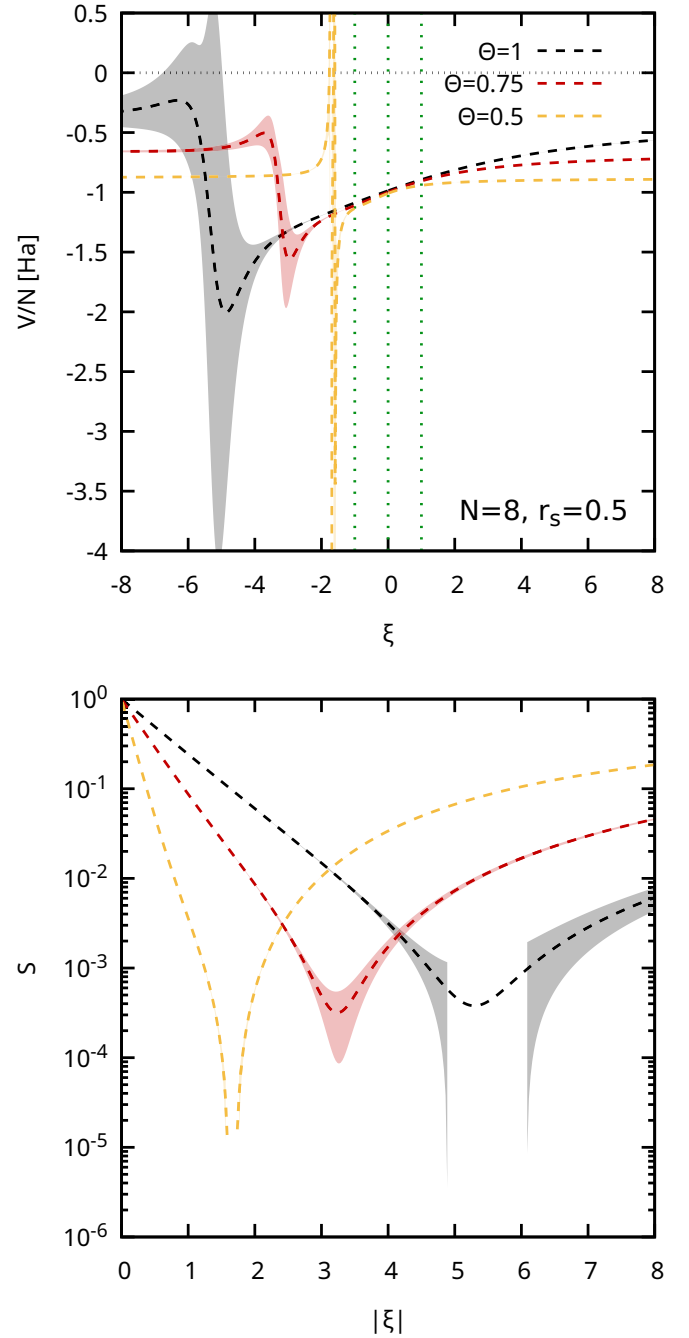


FIG. 2. Dependence of the interaction energy per particle (top) and the average sign (bottom) on the fictitious quantum statistics variable ξ for the unpolarized UEG with $N = 8$ particles at $r_s = 0.5$ and $\Theta = 1$ (black), $\Theta = 0.75$ (red), and $\Theta = 0.5$ (gold).

$\xi \in [-1, 0]$ of the real axis, any polynomial extrapolation is bound to fail. In particular, in the zero-temperature limit, these authors report the occurrence of such Z_ξ zeros for $\xi = -1, -1/2, -1/3, \dots, -1/(N-1)$, thus fundamentally preventing bosonic to fermionic extrapolations along the real axis. This conclusion is somewhat expected as it has been known that PIMC cannot directly reach the limit of $\beta \rightarrow \infty$ [201]. Fortunately, we do not find any zeros in Z_ξ for $|\xi| \leq 1$. At the same time, our PIMC simulations clearly show that the observed “pseudopoles”/“pseudozeros” move closer to

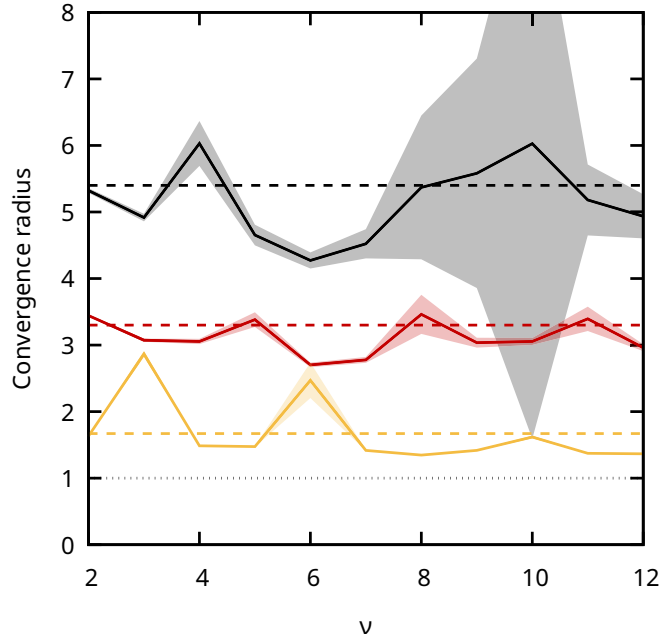


FIG. 3. Dependence of the radius of convergence estimate of the Taylor series around $\xi_T = 0$ on the polynomial order ν [cf. Eq. (14)] for the unpolarized UEG with $N = 8$ particles at $r_s = 0.5$ and $\Theta = 1$ (black), $\Theta = 0.75$ (red), and $\Theta = 0.5$ (gold). The horizontal dashed lines indicate the position of the “pseudo-poles” shown in the top panel of Fig. 2. The error estimates (shaded areas) have been obtained using a standard jackknife resampling scheme [198].

$|\xi| = 1$ with decreasing Θ for the investigated temperatures. Despite the fact that we constrained ourselves along the real axis, this is qualitatively consistent with Ref. [200].

In Fig. 3, we show the radius of convergence r computed via Eq. (14) as a function of the degree of the coefficients ν (solid lines) evaluated from the same PIMC simulations as Fig. 2 for the expansion point $\xi_T = 0$, i.e., computed directly from the ξ derivatives. Interestingly, these curves are in very good agreement with the position of the “pseudopoles,” which are included as the horizontal dashed lines. Importantly, the radius of convergence includes the physically relevant limit of $|\xi| = 1$ in all cases, leaving the door open to a controlled Taylor extrapolation to the fermionic limit.

Let us next consider the dependence of these findings on the number of simulated electrons N . In the top panel of Fig. 4, we show the ξ dependence of the potential energy per particle, with a particular focus on the location of the “pseudo-poles.” Interestingly, the latter only appears in our PIMC results for $N = 4$ (dashed gold) and $N = 8$ (solid black), but not for $N = 6$ (dotted red). This is further substantiated by the average sign S shown in the bottom panel of Fig. 4. Evidently, S vanishes for all depicted N within the given Monte Carlo error bars around $|\xi| = 1.5$, except for $N = 6$. This is likely a consequence of some special symmetry for this particular case; otherwise, the occurrence and the location of the “pseudo-pole” appear to be a general feature of the UEG, and likely of Fermi-Dirac systems overall. In Fig. 5, we show the corresponding convergence radii as a function of the

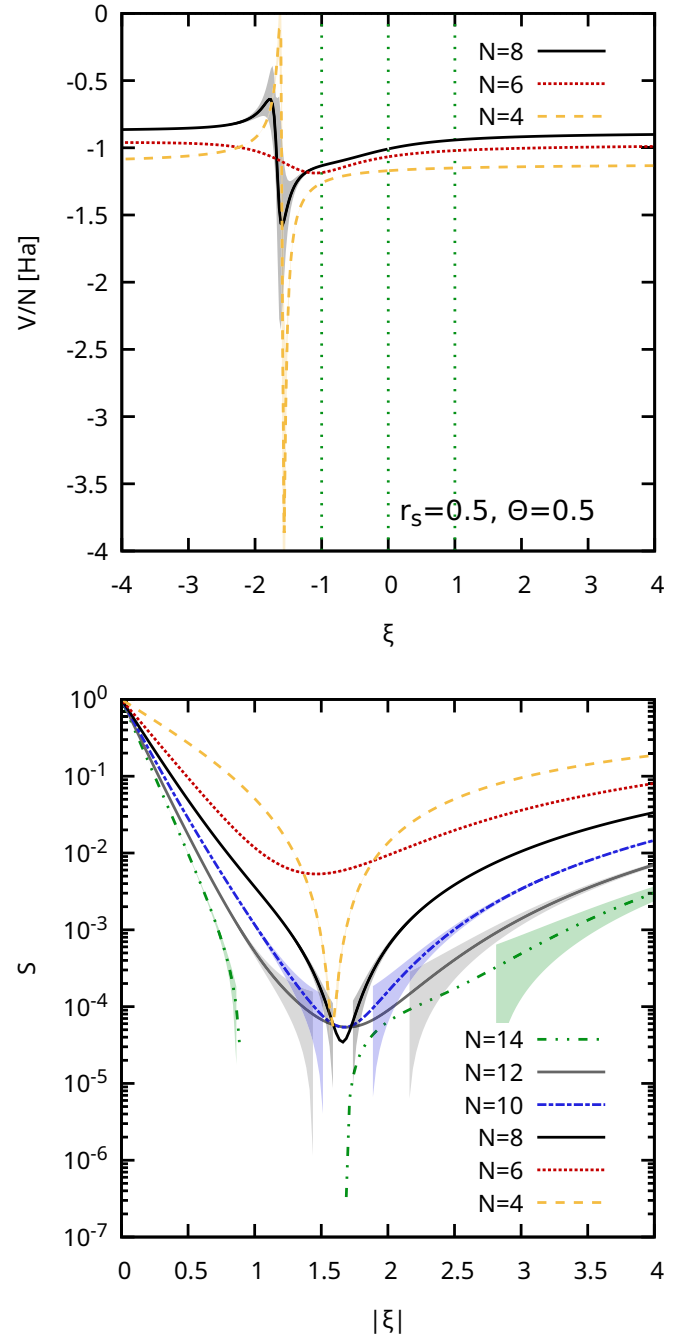


FIG. 4. Dependence of the total energy (per particle) (top) and average sign S (bottom) on the fictitious quantum statistics variable ξ for the unpolarized UEG at $r_s = 0.5$ and $\Theta = 0.5$. Results for $N = 8$ (solid black), $N = 6$ (dotted red), and $N = 4$ (dashed golden). For the average sign, results are also shown for $N = 14$ (dashed-double-dotted green), $N = 12$ (dashed-triple-dotted gray), and $N = 10$ (dash-dotted blue). Shaded areas indicate the statistical uncertainty.

polynomial degree ν . These curves are generally above unity, substantiating the principal possibility of a Taylor extrapolation to the fermionic limit; nevertheless, results for $N = 14$ are consistent with unity, which already indicates the difficulty of an extrapolation to the fermionic limit of $\xi = -1$.

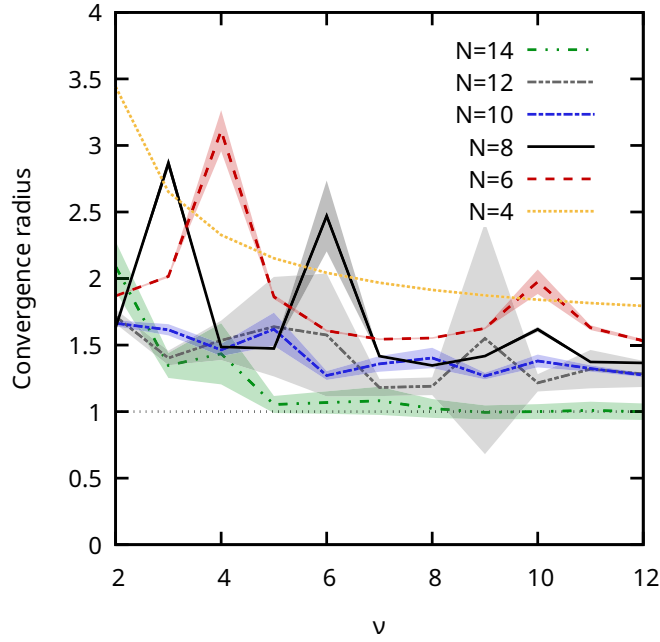


FIG. 5. Dependence of the radius of convergence estimate of the Taylor series around $\xi_T = 0$ on the polynomial order ν [cf. Eq. (14)] for the unpolarized UEG at $r_s = 0.5$ and $\Theta = 0.5$. Results for $N = 14$ (dashed-double-dotted green), $N = 12$ (dashed-triple-dotted gray), $N = 10$ (dash-dotted blue), $N = 8$ (solid black), $N = 6$ (dotted red), and $N = 4$ (dashed golden).

As the final question, we investigate the dependence of these findings on the density parameter in Fig. 6 for $N = 8$ and $\Theta = 0.5$. The black curve has been obtained for $r_s = 10$, which is on the margin of the strongly coupled electron liquid regime [36,110,169,202], and the red curve corresponds to $r_s = 3.23$, i.e., the electron number density of solid density hydrogen that is realized, e.g., in experiments with hydrogen jets [52,72,203,204]; the yellow curve represents $r_s = 0.5$, the weakly coupled high-density regime that we have considered so far. Interestingly, we only find the “pseudo-poles” for $r_s = 3.23$ and $r_s = 0.5$, whereas it is absent for $r_s = 10$. Indeed, the sign remains finite and can be resolved within the given error bars over the entire ξ range. Second, we find that the position of the “pseudo-poles” moves toward smaller $|\xi|$ with decreasing coupling strength and, thus, with increasing degree of quantum degeneracy. This is also directly reflected by the convergence radii of the Taylor expansion around $\xi_T = 0$ shown in Fig. 7.

Let us conclude this section with an attempted practical interpretation of the reported observations. Empirically, we find that the radius of convergence of the Taylor expansion around the Maxwell-Boltzmann system is directly connected to the (negative) value of ξ at which the average sign and, hence, the fermionic partition function vanishes. For the physically relevant domain of $\xi \in [-1, 1]$, the sign is expected to vanish only in the ground-state limit of $\beta \rightarrow \infty$ ($\Theta = 0$). We, therefore, expect Taylor extrapolations to the fermionic limit to converge at any finite temperature, even though, as we shall see below, the estimation of all significant coefficients might be unfeasible in practice.

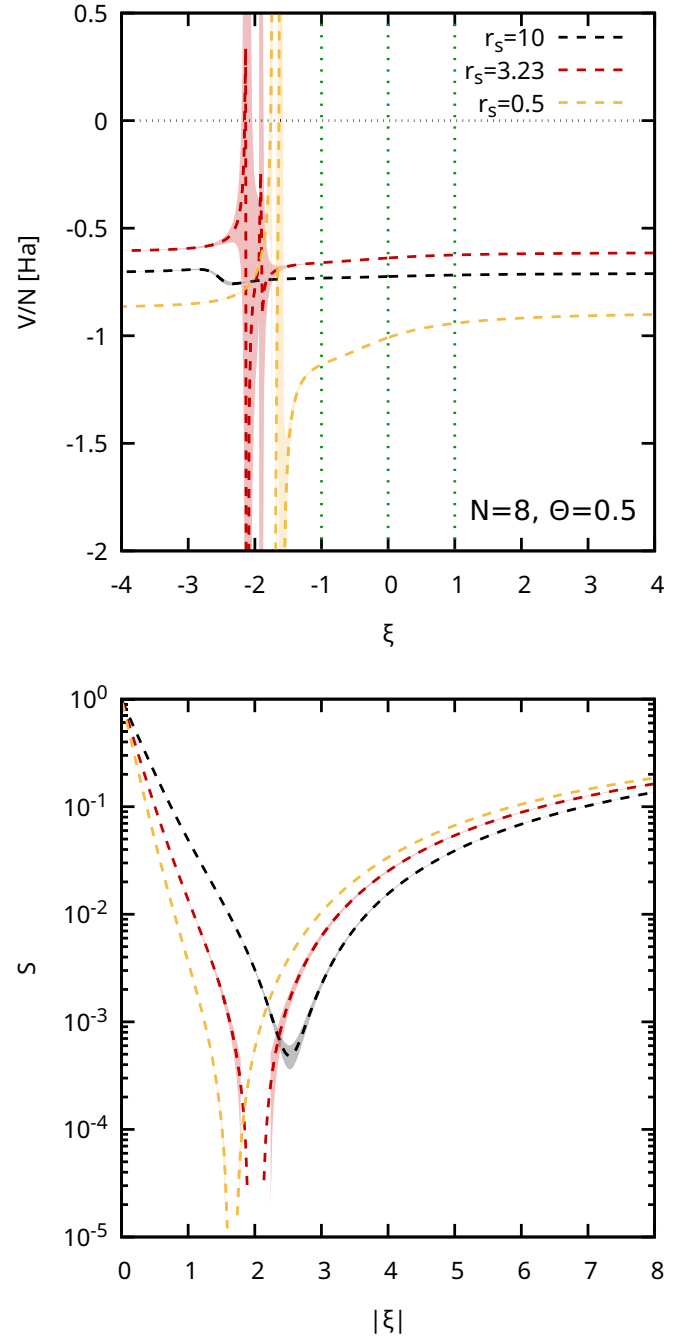


FIG. 6. Dependence of the interaction energy per particle (top) and the average sign (bottom) on the fictitious quantum statistics variable ξ for the unpolarized UEG with $N = 8$ particles at $\Theta = 0.5$. Results for $r_s = 10$ (dashed black), $r_s = 3.23$ (dashed red), and $r_s = 0.5$ (dashed golden).

B. PIMC results for ξ derivatives

Let us next turn our attention to the derivatives of expectation values with respect to ξ , which play a central role for our Taylor series perspective. As an example, we consider the interaction energy per particle V/N , which is shown in Fig. 8 for $N = 8$ at $r_s = 0.5$ and three temperatures. In addition, we have performed polynomial fits of the order $p = 2$ according to Eq. (13) for $\xi \in [0, 1]$, corresponding to the

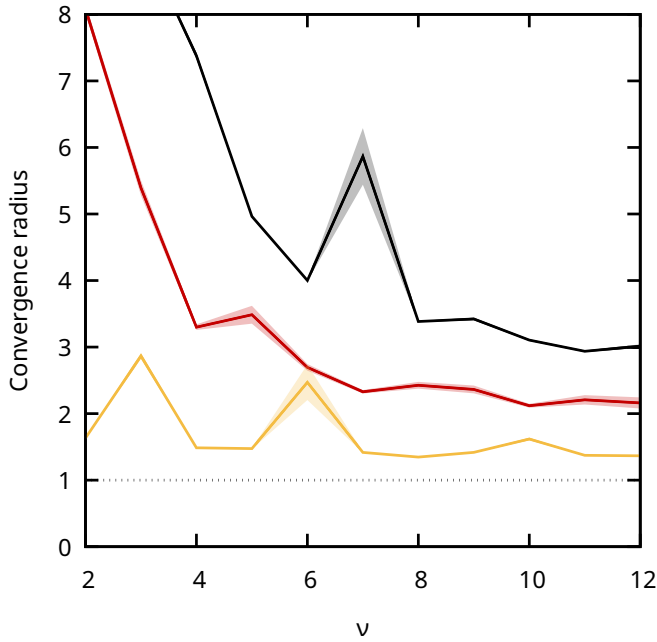


FIG. 7. Dependence of the radius of convergence estimate of a Taylor series around $\xi_T = 0$ as a function of the polynomial order ν [cf. Eq. (14)] for the unpolarized UEG with $N = 8$ particles at $\Theta = 0.5$. Results for $r_s = 10$ (dashed black), $r_s = 3.23$ (dashed red), and $r_s = 0.5$ (dashed golden).

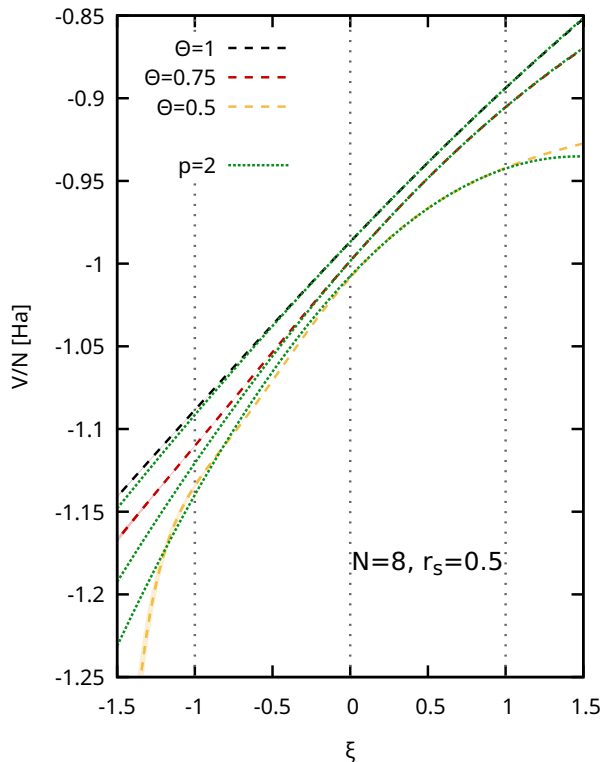


FIG. 8. Dependence of the interaction energy per particle on the fictitious quantum statistics variable ξ for the unpolarized UEG with $N = 8$ particles at $r_s = 0.5$ and $\Theta = 1$ (black), $\Theta = 0.75$ (red), and $\Theta = 0.5$ (gold). The green dots correspond to bosonic sector (within the fitting range of $\xi \in [0, 1]$) polynomial fits of the order of $p = 2$ [cf. Eq. (13)].

usual isothermal ξ extrapolation carried out in previous works [80,84,86,93]. For $\Theta = 1$ (black curve), the dependence of V on ξ is almost linear for $\xi \in [-1, 1]$ and the fermionic limit of $\xi = -1$ is accurately reproduced by the parabolic extrapolation. For $\Theta = 0.75$ (red curve), the dependence of V on ξ is noticeably less linear. As a result, the parabolic polynomial fit does reproduce its input data in the fitting interval of $\xi \in [0, 1]$, but the gap to the true fermionic limit widens compared to the higher temperature. Finally, we find a substantially less trivial dependence of V on ξ for $\Theta = 0.5$ (golden curve), with a pronounced curvature for $\xi \in [0, 1]$ and a significantly more linear progression for $\xi \in [-1, 0]$. The parabolic extrapolation is not capable of reproducing this intricate ξ dependence in the signful domain, and the relatively good agreement at $\xi = -1$ has to be viewed as coincidental.

A more systematic analysis of the polynomial fitting and extrapolation is presented in Fig. 9 for different polynomial degrees p . For $\Theta = 1$ (top panel), only the linear fit ($p = 1$, black) is incapable of reproducing the interaction energy in the fitting range of $\xi \in [0, 1]$ with deviations of $\Delta V/V \lesssim 0.1\%$, resulting in an extrapolation to the fermionic limit with an error of $\sim 0.8\%$. Interestingly, this error can be reduced with increasing p beyond $p = 2$, although this strategy is only expected to work when the accuracy of the underlying PIMC data is high enough to directly resolve the fermionic limit. The parabolic fit gives a systematic error of $\lesssim 0.3\%$ for $\xi = -1$, which is consistent with previous works [84]. Let us next turn to $\Theta = 0.75$, for which an analogous analysis is shown in the center panel of Fig. 9. Overall, we observe the same qualitative trends as for $\Theta = 1$, albeit with somewhat larger systematic errors compared to $\Theta = 1$. Specifically, we find a systematic error of 1% for the standard ξ extrapolation with $p = 2$ and even for $p = 6$ a small yet significant deviation of $\sim 0.1\%$ can be resolved with the given accuracy. The bottom panel of Fig. 9 shows results for $\Theta = 0.5$, for which the situation again becomes noticeably more complex. First, we note that $p = 2$ is no longer sufficient to accurately reproduce the interaction energy in the fitting range of $\xi \in [0, 1]$; $p \geq 3$ is required. Second, none of the depicted polynomial degrees is sufficient to reproduce the fermionic limit and the good agreement at $p = 2$ is decisively confirmed as being coincidental. For completeness, we have also included a comparison to highly accurate CPIMC reference data for $\xi = -1$ as the gray symbols for all three temperatures in Fig. 9. We find excellent agreement within the given error bars for all three cases, amounting to an agreement on the level of $\sim 0.01\%$ for $\Theta = 1$ and $\Theta = 0.75$ and $\sim 0.1\%$ for $\Theta = 0.5$ due to the more severe sign problem in our direct PIMC calculations.

Let us proceed with the topic at hand, which is the investigation of the derivatives of V_ξ with respect to ξ shown in Fig. 10. The left panel corresponds to the first derivative and the dashed curves show our direct PIMC results for $\xi \neq 0$ evaluated from Eq. (19). The blue dots have been computed for $\xi = 0$ via Eq. (24) and are in perfect agreement with the former datasets for all temperatures. The intersection of the results for $\Theta = 0.5$ and $\Theta = 0.75$ for $\xi = 0$ is likely a coincidence. Finally, the dotted green curves have been obtained by taking the derivative of the polynomial fits with $p = 10$ (cf. Fig. 9). They are in perfect agreement with the direct PIMC results within the fitting range of $\xi \in [0, 1]$, which

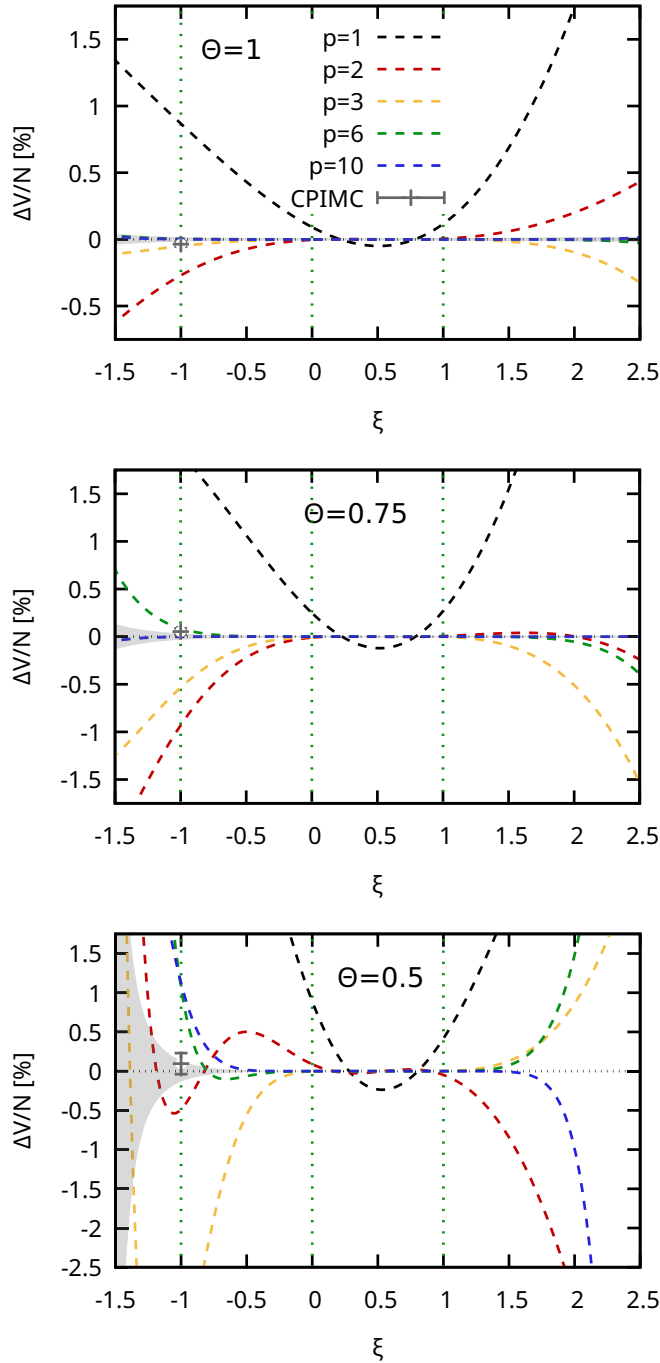


FIG. 9. Relative deviation of polynomial fits of different order p [cf. Eq. (13)] for the interaction energy per particle shown in Fig. 8. The shaded gray areas show the statistical error in the PIMC estimates for V_ξ . Also shown are comparisons to the CPIMC reference data in the fermionic limit of $\xi = -1$ (gray symbols).

further substantiates the correctness of our implementation. For $\Theta = 1$, we find a smooth curve without any marked features, and which is reproduced well by the polynomial fit over the entire depicted ξ range. For $\Theta = 0.75$, the first derivative with respect to ξ is substantially more complex than it was evident from V itself (cf. Fig. 8) and we find a shallow local maximum followed by a shallow local minimum around $\xi = -0.5$ and $\xi = -1$, respectively. Both features are well

reproduced by the polynomial fit with $p = 10$. For $\Theta = 0.5$, these features become substantially more pronounced, indicating the onset of strong quantum degeneracy effects. The tenth-order polynomial fit qualitatively captures the existence of both maximum and minimum, but fails to describe in particular the latter accurately.

The center and right panels of Fig. 10 show analogous investigations of the second and third derivatives of V with respect to ξ , with overall similar trends. Most importantly, we again find excellent agreement between the different PIMC estimators and the polynomials (within their fitting interval). In addition, our PIMC results reveal further intricacies of the impact of quantum statistics in both bosonic and fermionic sectors. While the present study is focused on the Taylor series estimation of the fermionic limit, future works might apply similar techniques also to gain further insights into the role and manifestation of Bose-Einstein statistics for a variety of systems, e.g., in the context of superfluidity [17,20,28,33,205–208].

C. Taylor series extrapolation

As the capstone of our work, we utilize our PIMC results for the ξ derivatives around $\xi = 0$ for a controlled Taylor series extrapolation to the fermionic limit of $\xi = -1$ and the bosonic limit of $\xi = 1$. In the left panel of Fig. 11, we analyze the convergence of Eq. (13) with respect to the Taylor series order p at $r_s = 0.5$, $\Theta = 1$ with $N = 14$ particles; the coefficients have been computed directly via Eq. (24). The green and blue symbols correspond to fermions and bosons, respectively. CPIMC results are employed as a reference for fermions and direct PIMC results are employed as a reference for bosons. As evident from Fig. 8, the bosonic and fermionic limits are nearly equidistant from the boltzmann results that correspond to $p = 0$. Both datasets rapidly converge with p . This can be discerned particularly well in the inset that shows deviations from the respective exact limits. Interestingly, truncating the Taylor series at $p = 1$ works better for fermions ($\Delta V \sim 0.1\%$) than bosons ($\Delta V \sim 0.3\%$), although both errors are small and of the same order of magnitude.

In practice, a successful application of the original ξ extrapolation method has been its utilization for larger systems, whose PIMC simulations are otherwise computationally unfeasible (even in case of a moderate degree of quantum degeneracy) due to the exponential increase in compute time with N that is inherent to the sign problem [84]. As an example, we consider $N = 66$ at the same density and temperature in the right panel of Fig. 11. The green and blue horizontal lines still correspond to the correct fermionic (CPIMC) and bosonic (direct PIMC) limits, while the colored symbols now depict the PIMC results for different values of ξ_{ref} [cf. Eq. (9)]. It can be deduced from Eqs. (24) and (25) [see Eqs. (26)–(28) for an explicit form for $\nu = 1, 2, 3$] that the Taylor series around $\xi_T = 0$ truncated at an order p only contains measurements with a number of pair permutations up to $N_{\text{pp}} \leq p$. However, such configurations are rare for larger systems even for weak to moderate degrees of quantum degeneracy. It is straightforward that even if the number of pair permutations per particle is low, there will be more such exchanges when there are more particles. Consequently,

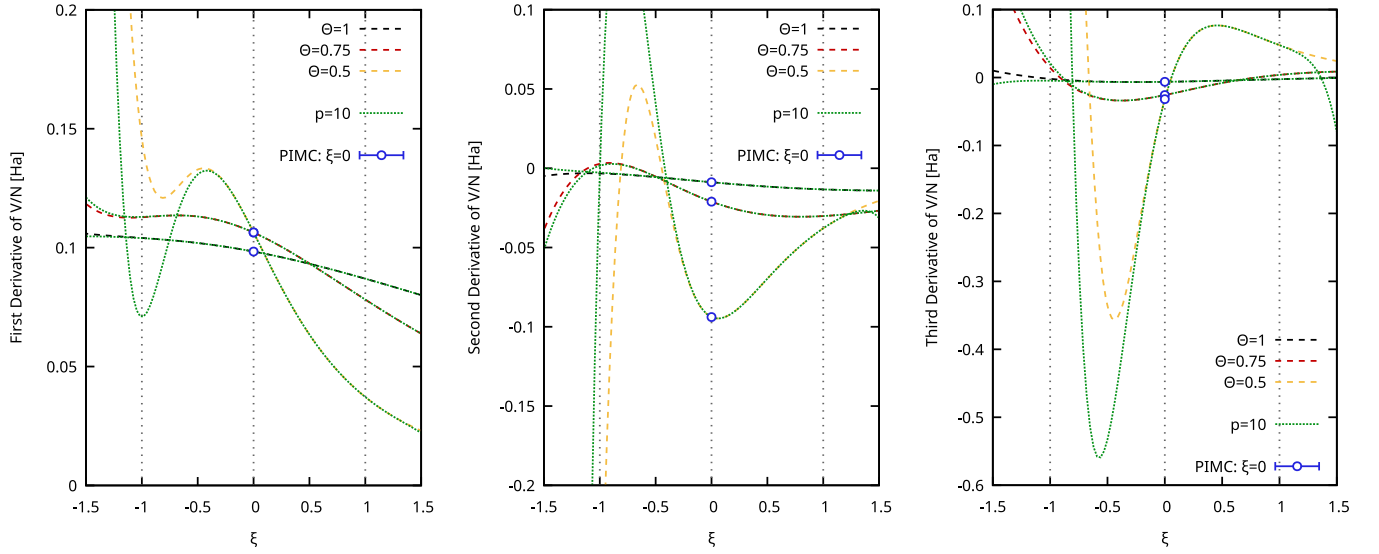


FIG. 10. Dependence of the first (left), second (center), and third (right) order ξ derivatives of the interaction energy per particle on the fictitious quantum statistics variable ξ for the unpolarized UEG with $N = 8$ particles at $r_s = 0.5$. The dashed black, red, and golden curves have been computed using Eqs. (19), (20), and (21) for $\Theta = 1$, $\Theta = 0.75$, and $\Theta = 0.5$, respectively. The dotted green curves show the corresponding derivatives of the polynomial fits to V_ξ with $p = 10$ (see Figs. 8 and 9). The blue dots correspond to PIMC results for the ξ derivatives at $\xi = 0$ computed via Eq. (24).

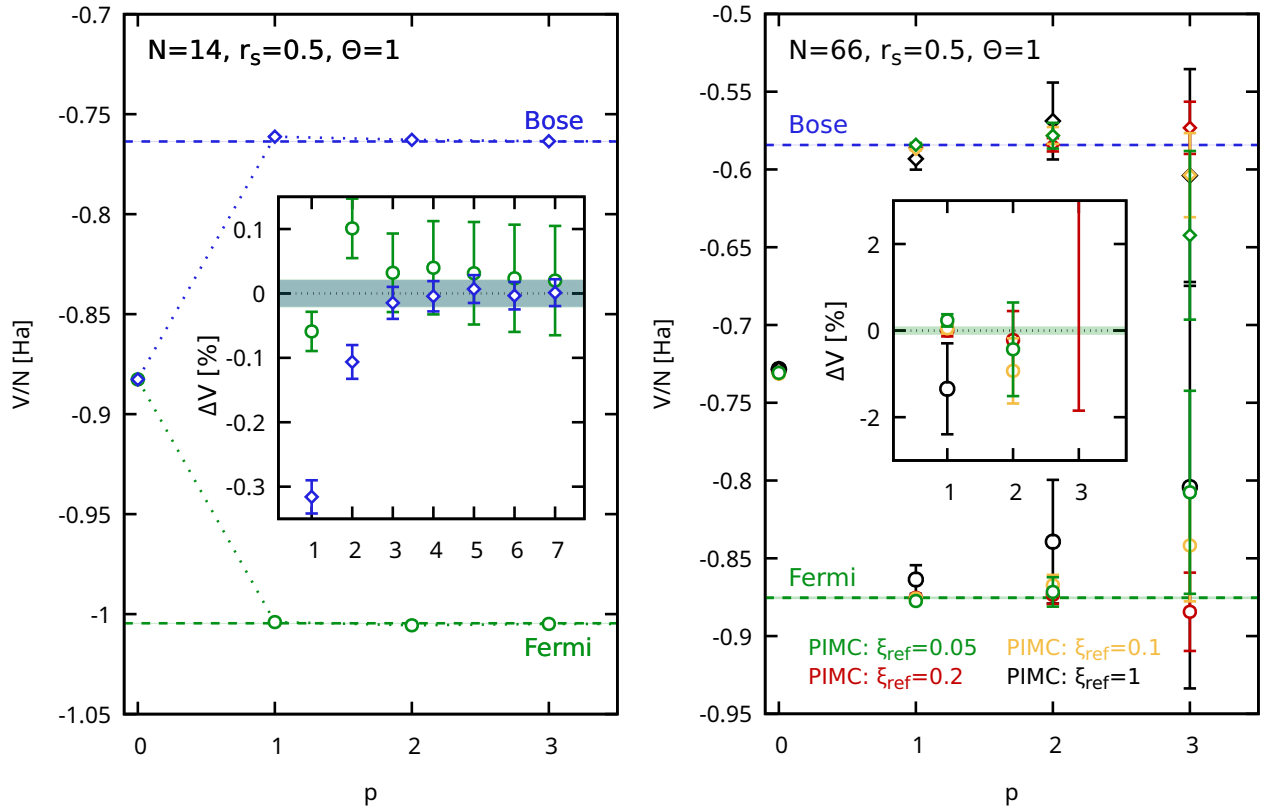


FIG. 11. Taylor series extrapolation of the interaction energy per particle for the unpolarized UEG at $r_s = 0.5$, $\Theta = 1$ to the bosonic limit of $\xi = 1$ (blue) and the fermionic limit of $\xi = -1$ (green) as a function of the Taylor order p [cf. Eq. (12)]. Results for $N = 14$ (left) and $N = 66$ (right). The fermionic reference data (horizontal dashed green) have been obtained with CPIMC. For $N = 14$, all results have been obtained for $\xi_{\text{ref}} = 1$. For $N = 66$, we compare results for different ξ_{ref} (see the color code). The insets show the relative deviation to the correct bosonic and fermionic limits ($N = 14$) and the relative deviation to the correct fermionic limit for different ξ_{ref} ($N = 66$).

configurations with $N_{pp} \ll N$ will be underexplored and the corresponding ξ derivatives around $\xi = 0$ will suffer from comparably bad statistics for $\xi_{\text{ref}} = 1$. This causes the relatively large error bars of the black symbols in the right panel of Fig. 11. As a simple workaround, we have also performed PIMC calculations with $\xi_{\text{ref}} = 0.2$ (red), $\xi_{\text{ref}} = 0.1$ (gold), and $\xi_{\text{ref}} = 0.05$ (green). In these calculations, the number of pair permutations is artificially suppressed (see also the discussion in Ref. [91]), giving us a higher accuracy in the relevant sectors. Indeed, we obtain error bars that are smaller by 2 orders of magnitude, in particular for $\xi_{\text{ref}} = 0.1$ and $\xi_{\text{ref}} = 0.2$. In both cases, we find perfect agreement with the true fermionic limit within the associated statistical error bars of $<0.15\%$ for $p = 1$, which surpasses the accuracy attained in previous studies for the same parameters using the standard ξ extrapolation method. We stress that a direct PIMC simulation would give an average sign of $S \sim 10^{-6}$ for $\xi = -1$, making this fundamentally impossible. The current scheme gives us a speedup by 12 orders of magnitude *from a single PIMC simulation*, e.g., at $\xi_{\text{ref}} = 0.2$.

The final question concerns how to decide if the Taylor series has indeed converged at a given p and with what accuracy. The easiest option would be to rely on PIMC simulations of a smaller system. Indeed, it has been argued in earlier works [84,86,93] that the effects of quantum statistics, which are at the root of the ξ dependence, are predominantly short ranged and, hence, should not strongly depend on the system size. This was indeed verified empirically based on PIMC simulations. In the present work, the case of $N = 14$ depicted in the left panel of Fig. 11 indicates that the systematic error should be $\lesssim 0.1\%$ for $p = 1$. Alternatively, we can check a larger p for the larger system of interest. For the present case, considering $p = 2$ would then confirm convergence within an error bar of $\sim 0.6\%$, comparable to the accuracy level in the two previous investigations [84,91], where such a size-consistent check was not carried out.

In Fig. 12, we analyze the convergence of the truncated Taylor series with $N = 14$, at $r_s = 0.5$ and the somewhat lower temperature of $\Theta = 0.75$ (for $\xi_{\text{ref}} = 1$). Overall, we find the same trends as for $\Theta = 1$, although there appear systematic errors of $\sim 0.2\%$ for $p = 1$ and $p = 2$ in the extrapolation to the fermionic limit. This is often sufficient for practical applications [58,108,115,209], in particular when it, at the same time, facilitates the simulation of larger systems and thus helps mitigate possible finite-size errors [210–213]. Interestingly, we again observe a slower convergence for bosons compared to fermions.

As the final and most difficult example, we consider the lower temperature of $\Theta = 0.5$ in Fig. 13. Since for $N = 14$ we cannot reach the fermionic limit of $\xi = -1$ due to the fermion sign problem, we consider a range of particle numbers $N = 4, \dots, 14$, and the results are depicted by the differently colored curves. The top row corresponds to the interaction energy per particle and the left panel shows the convergence for $\xi = -1$ with respect to the Taylor order p . Overall, we find a smooth and monotonic convergence for $N = 4$ (gold), whereas the curves start to increasingly oscillate for larger numbers of electrons. These oscillations have amplitudes of the order of a few percent, and the resulting

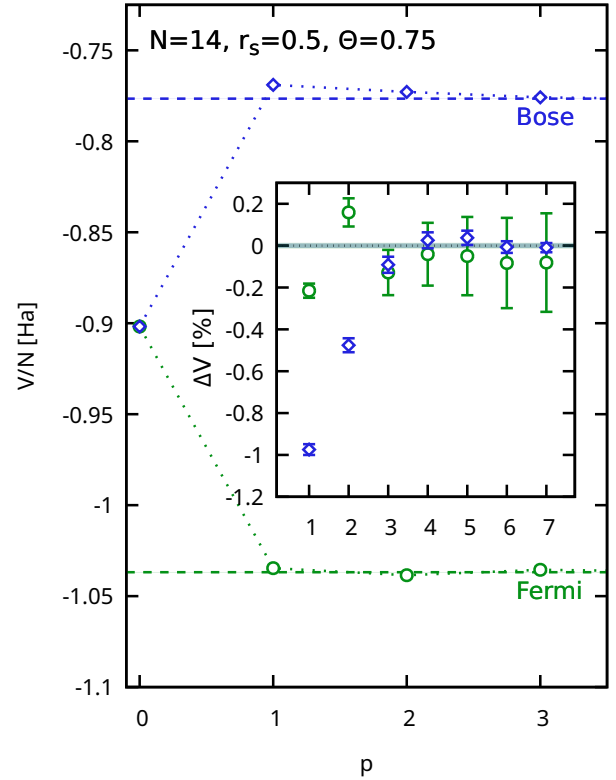


FIG. 12. Taylor series extrapolation of the interaction energy per particle for the unpolarized UEG with $N = 14$ at $r_s = 0.5$ and $\Theta = 0.75$ to the bosonic limit of $\xi = 1$ (blue) and the fermionic limit of $\xi = -1$ (green) as a function of the Taylor order p [cf. Eq. (12)]. The fermionic reference data (horizontal dashed green) have been obtained with CPIMC. All results have been obtained for $\xi_{\text{ref}} = 1$. The inset shows the relative deviation to the correct bosonic and fermionic limits.

nonmonotonicity makes any kind of controlled truncation or extrapolation nontrivial in practice.

The kinetic energy per particle shown in the bottom row of Fig. 13 overall exhibits the same qualitative trends, albeit with larger statistical error bars due to the thermodynamic PIMC estimator [199]. A second difference with respect to the interaction energy is that the true kinetic energy is underestimated in the limit of $p = 0$, whereas the (negative) interaction is overestimated in the boltzmann systems compared to proper Fermi-Dirac statistics.

Let us next analyze the behavior of the Taylor expansion coefficients c_ν , which are computed from Eq. (24) and are shown in the central column of Fig. 13. In this representation, it is easy to see that (1) the amplitude of the oscillations in the c_ν decays with increasing ν , which is expected as $\xi = \pm 1$ lies within the radius of convergence (cf. Fig. 5); and (2) the wavelength of the oscillations increases with the number of particles. Empirically, we find that a rescaling of the expansion order ν by a factor of $1/\sqrt{N}$ leads to a remarkable invariance of the oscillations in the kinetic and potential energy coefficients with respect to N (see the right column of Fig. 13). In practice, the ν dependence of the Taylor

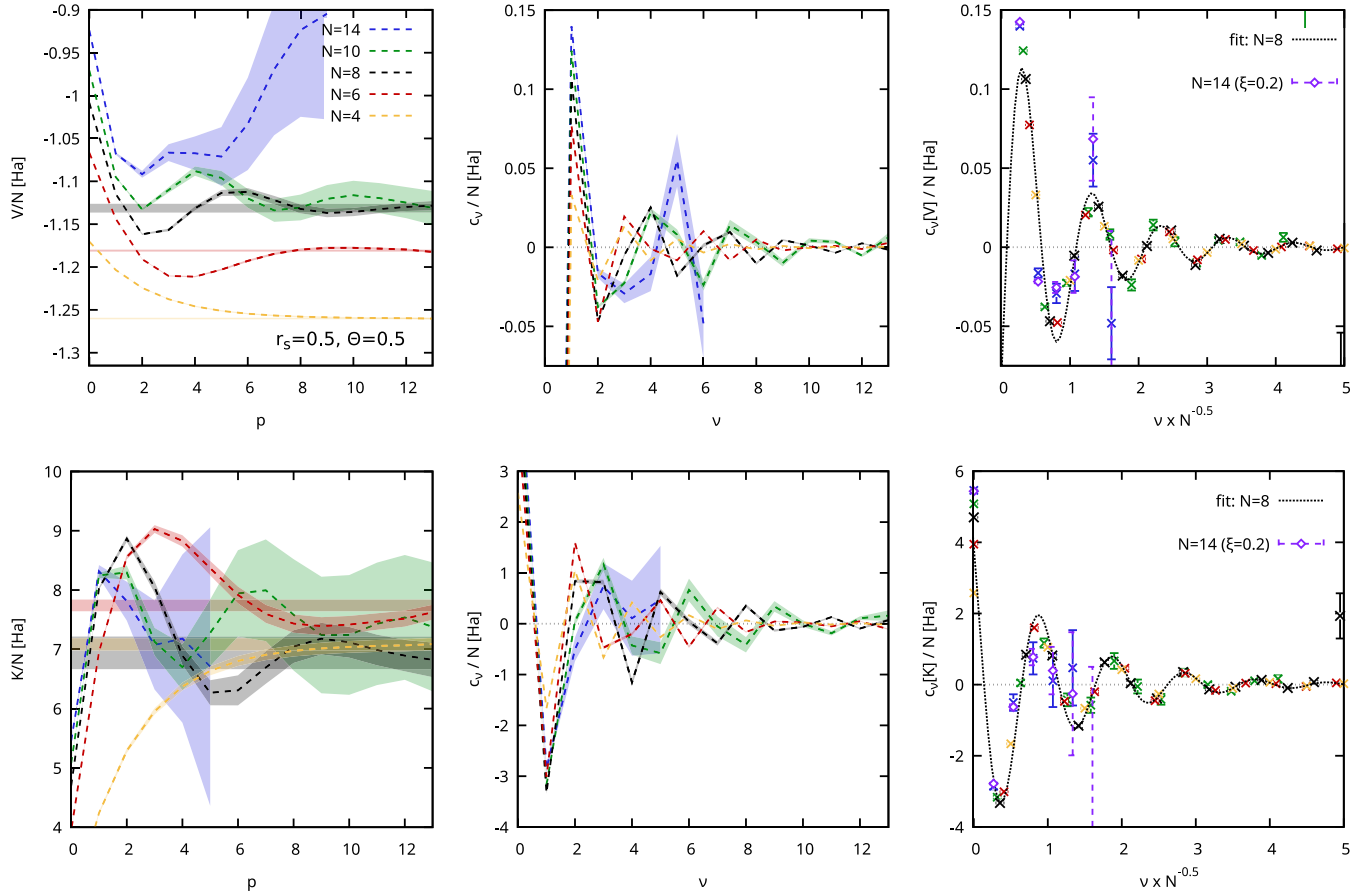


FIG. 13. Left: convergence of the Taylor series extrapolation to the fermionic limit of $\xi = -1$ around $\xi_T = 0$ for the potential (top row) and kinetic (bottom row) energy per particle for the unpolarized UEG at $r_s = 0.5$ and $\Theta = 0.5$ with different N . The horizontal shaded areas indicate the full direct PIMC results for $\xi = -1$ with the corresponding uncertainty interval for $N = 4, 6, 8$. Center: convergence of the extracted expansion coefficients c_v [cf. Eq. (13)]. Right: same as center panel, but the x axis has been rescaled by a factor of $1/\sqrt{N}$. The dotted black lines show empirical fits according to Eq. (29) to the $N = 8$ dataset obtained within the fitting interval of $v \in [1, 11]$. All results have been obtained for $\xi_{\text{ref}} = 1$; the single exception is given by the lilac diamonds corresponding to results for $\xi_{\text{ref}} = 0.2$.

coefficients of an observable $O \in \{V, K\}$ is well reproduced by

$$c_v[\hat{O}] = a_O e^{-b_O v} \sin(2\pi c_O v) + d_O e^{-b_O v/2} \sin(2\pi c_O v + \pi/2), \quad (29)$$

with a_O , b_O , c_O , and d_O the four free parameters. The results are shown, as the dotted black lines in the right column of Fig. 13 and fit all datasets remarkably well. A possible exception is given by $N = 14$, which is afflicted with substantially higher error bars. It is also pointed out that the rescaling of the x axis by $1/\sqrt{N}$ means that we need larger v for larger systems to reach the same characteristic oscillation decay on the rescaled scale compared to smaller systems. For completeness, we have also performed additional PIMC simulations with $\xi_{\text{ref}} = 0.2$, resulting in a higher accuracy for small v ; see the lilac diamonds in the right column. While indeed offering an improvement for small v , this advantage is not decisive.

In Fig. 14, we show the convergence of the Taylor series extrapolation to $\xi = -1$, but with the x axis again being rescaled by a factor of $1/\sqrt{N}$. In this representation, it appears that all curves converge toward the true fermionic limit around $p/\sqrt{N} \approx 5$. Unfortunately, this limit cannot be reached for $N = 14$ at these conditions, as discussed above. In light of

the remarkable N invariance observed in Fig. 13, it makes sense to ponder whether it can be exploited to model the convergence of a given larger N based on a fit following Eq. (29) for a smaller N ; here, we again choose $N = 8$ for the latter. Following this procedure to obtain the Taylor coefficients for $v > 2$ with $N = 14$ results in the solid blue curve in Fig. 14. Evidently, these data exhibit even faster oscillations compared to the other datasets. This is a consequence of the denser v grid on this rescaled representation. In addition, the blue curve shows a nice and controlled convergence; yet, its $p \rightarrow \infty$ limit differs from the true fermionic interaction energy (dotted blue line obtained from CPIMC) by $\sim 3\%$, which is even worse than a simple truncation at $p = 2$ for this case. Thus, we have to conclude that there presently does not exist a reliable strategy to converge the Taylor series to the true fermionic limit for strong degrees of quantum degeneracy.

Let us finish our investigation with an analysis of the capability of the Taylor series expansion around $\xi_T = 0$ to describe the actual ξ dependence of an observable over the entire relevant ξ range. To this end, we show the total energy per particle for $N = 8$, $r_s = 0.5$, and $\Theta = 0.5$ in Fig. 15. The dashed black line shows our direct PIMC results, which exhibit a

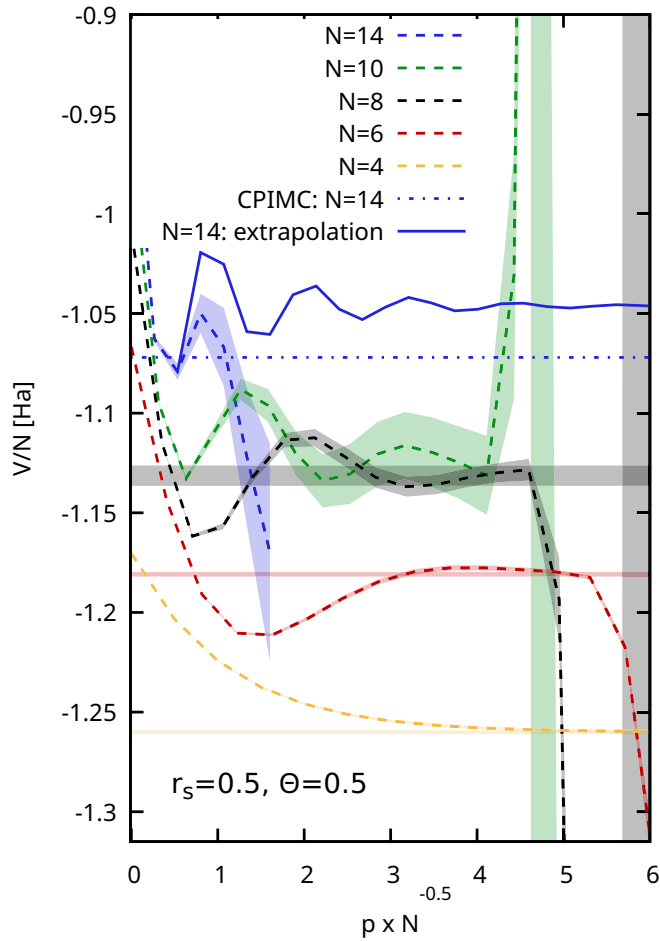


FIG. 14. Convergence of the Taylor series extrapolation around $\xi_T = 0$ to the fermionic limit of $\xi = -1$ for the interaction energy per particle for the same conditions as in Fig. 13, but with the x axis having been rescaled by a factor of $1/\sqrt{N}$. The horizontal shaded areas show direct PIMC results for $\xi = -1$ with the associated uncertainty interval for $N = 4, 6, 8$. The dashed blue line shows CPIMC reference data for $N = 14$ and the solid blue line has been obtained by recomputing the Taylor coefficients for $\nu > 2$ with the fit function [Eq. (29)] determined for $N = 8$.

complicated behavior in particular around $\xi \sim -0.5$ already hinted at in Ref. [84]. The dash-dotted horizontal gray bar indicates a highly accurate CPIMC reference result for the fermionic limit, which agrees with our direct PIMC calculations within the given error interval (shaded gray area), as it is expected. We note that we find an average sign of $S \approx 0.0036$, making our simulations computationally very demanding despite the comparably small system size. The dotted colored curves correspond to Taylor series expansions around $\xi_T = 0$ evaluated via Eq. (24) and truncated at different polynomial degrees p . At a first glance, the extrapolation to the fermionic limit appears to be more challenging than to the bosonic limit. This is somewhat unsurprising as the ξ dependence in the bosonic sector of $\xi > 0$ seems less complicated than in the fermionic domain of $\xi < 0$. Then again, $p = 6$ leads to worse results for $\xi = 1$ compared to $p = 2$, indicating a nonmonotonic convergence. A key question is if the Taylor series around $\xi_T = 0$ is capable of capturing the full complex

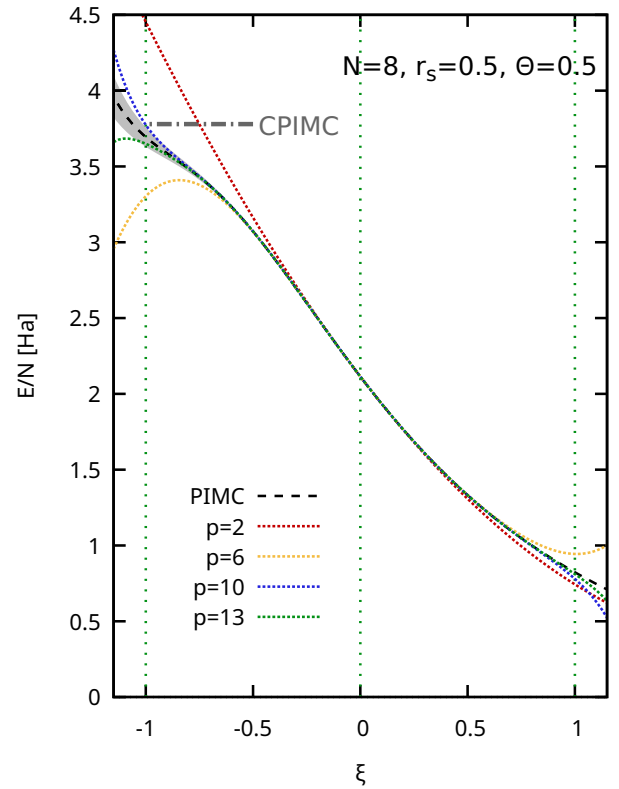


FIG. 15. Dependence of the total energy per particle on the fictitious quantum statistics variable ξ for $N = 8$, $r_s = 0.5$, and $\Theta = 0.5$ (computed for $\xi_{\text{ref}} = 1$). Dashed black: full PIMC results obtained from a single simulation at $\xi_{\text{ref}} = 1$ via Eq. (9). The shaded gray area indicates the associated uncertainty interval. The horizontal dash-dotted gray line indicates the highly accurate CPIMC result at the $\xi = -1$ fermionic limit. The dotted curves have been computed by combining the ξ derivatives around $\xi = 0$ [Eq. (24)] with the corresponding Taylor series expansion [Eq. (13)] truncated at different polynomial degrees p .

ξ dependence in the fermionic sector. This is indeed the case for $p = 10$ (blue) and $p = 13$ (green), leading us to the following conclusion: Empirically, the polynomial ansatz [Eq. (13)] is well justified by the present Taylor series perspective even for strong levels of quantum degeneracy. The practical challenge will be to find suitable truncation schemes that avoid the evaluation of coefficients involving larger numbers of pair exchanges N_{pp} , or to find alternative analytical simplifications.

IV. SUMMARY AND OUTLOOK

In this work, we investigated the *ab initio* PIMC simulation of quantum degenerate fermions ($\xi = -1$) [and bosons ($\xi = 1$)] from the perspective of a Taylor expansion around the limit of distinguishable quantum particles, i.e., boltzmannons ($\xi = 0$). To this end, we derived PIMC estimators to evaluate the arbitrary-order derivatives of arbitrary observables $\hat{O}$ with respect to the continuous quantum statistics variable ξ for finite values of ξ and the particularly important case of $\xi = 0$. This allowed us to evaluate a Taylor series representation of O_ξ up to, in principle, arbitrary order p , which includes

the previous empirical parabolic isothermal ξ extrapolation method [80,84,86,93] as a special case.

In the first part of our investigation, we analyzed the radius of convergence of the Taylor series, which, in all considered cases, includes the bosonic and fermionic limits of $\xi = \pm 1$. Interestingly, for some parameters, we observed “pseudo-poles” for $\xi < -1$, which would be consistent with the recent study of Lee-Yang zeros by He *et al.* [200]. As the next step, we explicitly investigated the derivatives of an observable with respect to ξ , which are difficult to capture within polynomial extrapolation ansätze based on fits in a limiting fitting range such as $\xi \in [0, 1]$. Finally, we investigated the convergence of the Taylor extrapolation to the fermionic and bosonic limits for a range of conditions. Remarkably, we find that accurate estimates of fermionic properties with an accuracy of $\Delta O \sim 0.1\%$ are possible even after truncating at $p = 1$ for $\Theta \geq 0.75$. This indicates that, while not necessarily small, the influence of quantum statistics remains approximately linear in this regime. In practice, this has allowed us to compute highly accurate results for the UEG with $N = 66$, $r_s = 0.5$ and $\Theta = 1$ from a single PIMC simulation with, e.g., $\xi_{\text{ref}} = 0.2$. For $\Theta = 0.5$, on the other hand, the situation is considerably more complex. In particular, the convergence of the Taylor series is nonmonotonic, with some characteristic damped oscillations of the Taylor coefficients amenable to near N invariance after rescaling the coefficient order ν by an empirical factor of $1/\sqrt{N}$. Unfortunately, these commonalities between different N do not allow us to estimate the converged fermionic limit for, say, $N = 14$ without a systematic error of $\sim 3\%$. Conversely, we found that more than ten Taylor coefficients are required to converge to the fermionic limit for $\Theta = 0.5$, which is challenging in practice.

We expect our results to be useful for a variety of PIMC simulations over a range of research fields. First and foremost, the fermion sign problem remains a central bottleneck in physics, quantum chemistry, and related disciplines. The present Taylor series perspective put the empirical isothermal ξ extrapolation technique in a more firm theoretical ground and makes it more controlled by providing the possibility for convergence analysis. In addition, we have shown that one can evaluate, in principle, the entire Taylor series from a single simulation at a given reference value ξ_{ref} (also, without any reweighting [91]), which is computationally efficient. We note that the estimation of the derivatives around $\xi = 0$ simply involves the estimation of expectation values for different numbers of pair permutations N_{pp} , which can be easily incorporated into existing PIMC codes.

Future works might focus on the dedicated analysis of the Taylor coefficients for strong quantum degeneracy, and on the development of improved truncation schemes. In addition, we note that the presented estimators for the ξ derivatives might be used to improve polynomial fitting schemes, and thus improve previously used ξ -extrapolation ansätze. A particularly important application will be the Taylor series analysis of real warm dense matter systems that include both electrons and positively charged nuclei [72,86,87,94,143,214,215], which will be pursued in a dedicated future study. Finally, we note that the gradual activation of quantum statistics in terms of the fictitious quantum statistics parameter ξ is interesting in itself, and might also open up perspectives onto important physical effects in Bose-Einstein systems such as superfluidity.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [193].

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