

Application of a spherically averaged pair potential in *ab initio* path integral Monte Carlo simulations of a warm dense electron gas

Tobias Dornheim ^{1,2,*} Thomas M. Chuna ^{1,2} Hannah M. Bellenbaum ^{1,2,3} Zhandos A. Moldabekov ^{1,2}
Panagiotis Tolias ⁴ and Jan Vorberger ²

¹*Center for Advanced Systems Understanding (CASUS), D-02826 Görlitz, Germany*

²*Helmholtz-Zentrum Dresden-Rossendorf (HZDR), D-01328 Dresden, Germany*

³*Institut für Physik, Universität Rostock, D-18057 Rostock, Germany*

⁴*Space and Plasma Physics, Royal Institute of Technology (KTH), Stockholm, SE-100 44, Sweden*



(Received 1 April 2025; accepted 7 August 2025; published 8 September 2025)

Spherically averaged periodic pair potentials offer the enticing promise to provide accurate results at a drastically reduced computational cost compared to the traditional Ewald sum. In this work, we employ the pair potential by Yakub and Ronchi [*J. Chem. Phys.* **119**, 11556 (2003)] in *ab initio* path integral Monte Carlo (PIMC) simulations of the warm dense uniform electron gas. Overall, we find very accurate results with respect to Ewald reference data for integrated properties such as the kinetic and potential energy, whereas wavenumber-resolved properties such as the static structure factor $S(\mathbf{q})$, the static linear density response $\chi(\mathbf{q})$, and the static quadratic density response $\chi^{(2)}(\mathbf{q}, 0)$ fluctuate for small q . In addition, we perform an analytic continuation to compute the dynamic structure factor $S(\mathbf{q}, \omega)$ from PIMC results of the imaginary-time density-density correlation function $F(\mathbf{q}, \tau)$ for both pair potentials. Our results have important implications for future PIMC calculations, which can be sped up significantly using the Yakub and Ronchi potential for the estimation of equation-of-state properties or q -resolved observables in the noncollective regime, whereas a full Ewald treatment is mandatory to accurately resolve physical effects manifesting for smaller q , including the evaluation of compressibility sum rules, the interpretation of x-ray scattering experiments at small scattering angles, and the estimation of optical and transport properties.

DOI: [10.1103/lj9c-bh48](https://doi.org/10.1103/lj9c-bh48)

I. INTRODUCTION

Warm dense matter (WDM) comprises a complex state of matter that is often characterized by at least two dimensionless parameters simultaneously of the order of one [1,2]: the density parameter—also coined as Wigner-Seitz radius— $r_s = (3/4\pi n)^{1/3}$, with n being the electronic number density, and the degeneracy temperature $\Theta = (\beta E_F)^{-1}$, where $\beta = 1/k_B T$ is the inverse temperature in energy units and E_F the usual Fermi energy of the electrons [3]. In nature, such seemingly extreme conditions abound in astrophysical objects such as giant planet interiors [1,4,5] and white dwarf atmospheres [6,7]. In addition, extreme states of matter become increasingly important for cutting-edge technological applications, with inertial confinement fusion (ICF) [5,8,9] being a prime example. Indeed, recent spectacular achievements at the National Ignition Facility [10,11] and the OMEGA laser facility [12] have demonstrated the principal feasibility of using ICF to produce net energy, but the pathway towards a commercial utilization of this technology will require a further

optimization of current energy gains by several orders of magnitude [13]. This, in turn, will require integrated simulations with real predictive capability, which must also cover the initial stages of the compression path, where both the fusion fuel and its surrounding ablator material have to traverse the WDM regime in a controlled way [14].

From a theoretical perspective, the accurate description of WDM poses a formidable challenge as it must treat holistically a plethora of physical effects. Specifically, the moderate to strong coupling of the Coulomb interacting electrons in nuclei often rules out weak-coupling expansions such as equilibrium or nonequilibrium Green's functions [15–17]. In addition, quantum degeneracy and quantum diffraction effects prevent the application of semiclassical methods such as molecular dynamics simulations with effective quantum pair potentials between the electrons except at very high temperatures [15,18]. On the other hand, the strong thermal excitations that occur when $\Theta \sim 1$ generally prevent a straightforward application of the well-stocked arsenal of ground-state simulation tools, e.g., from quantum chemistry and material science [19–21]. Finally, WDM states often feature partial ionization rendering them substantially more complex than either the well-defined orbitals of bound electrons at ambient conditions or the fully ionized semiclassical plasma.

In this situation, *ab initio* thermal density-functional theory (DFT) [22] has emerged as the de-facto workhorse of WDM theory as it often balances a reasonable level of accuracy with an acceptable computational effort. Specifically, the

*Contact author: t.dornheim@hzdr.de

Published by the American Physical Society under the terms of the [Creative Commons Attribution 4.0 International](https://creativecommons.org/licenses/by/4.0/) license. Further distribution of this work must maintain attribution to the author(s) and the published article's title, journal citation, and DOI.

combination of a DFT treatment of the electrons with a classical or quantum molecular dynamics propagation of the ions within the widely assumed Born-Oppenheimer approximation [1] allows for the estimation of a broad range of material properties such as the equation of state, ionic correlation functions, and electronic transport properties; see the review article by Bonitz *et al.* [15] for a topical overview. The accuracy of a given DFT simulation decisively relies on the employed electronic exchange-correlation (XC) functional, which has to be supplied as an external input. At ambient conditions, where electrons can generally be assumed to be in their respective ground state, the applicability of the zoo of existing functionals [23] for different materials and properties is relatively well understood. However, it is clear that thermal DFT simulations of real WDM systems require a thermal XC functional that explicitly depends on the temperature. Here, the situation is significantly less clear than in the extensively explored ground-state limit, as available XC functionals are substantially less numerous [21,24–27], and their respective accuracy has been benchmarked in far fewer situations [28–32].

In practice, both the construction of thermal XC functionals by itself as well as their rigorous benchmarking require the availability of highly accurate results that have been obtained without the need for any empirical external input. In principle, experimental observations would constitute a possible route, but their respective interpretation is notoriously difficult [33] and often depends on a number of uncontrolled model assumptions such as the decomposition of the electrons into effectively bound and free states [34–39]. A more realistic option is given by first-principles quantum Monte Carlo (QMC) methods [40], with the *ab initio* path integral Monte Carlo (PIMC) technique [41] being the method of choice at finite temperatures. Being exact within the given Monte Carlo error bars, the central limitation of PIMC simulations is the infamous fermion sign problem [42–44] that emerges from the antisymmetry of the fermionic density matrix under the exchange of particle coordinates. The sign problem manifests as an exponentially decreasing signal-to-noise ratio in PIMC simulations when the system size N or inverse temperature β are being increased.

Due to the pressing need to understand the properties of WDM—as well as a host of other interesting thermal Fermi-Dirac systems such as ultracold ^3He [45–47] or electrons in quantum dots [48–50]—there has been a remarkable surge of progress in fermionic QMC simulations over the last 15 years or so [16,51–65]. In fact, recent methodological developments based on path integral calculations of fictitious identical particles [63,64,66,67] allow for the simulation of up to $N = 1000$ electrons at weak to moderate levels of quantum degeneracy [68].

These developments open up both promising opportunities and new challenges. On the one hand, large simulations reduce finite-size effects, i.e., the difference in simulation results between a finite N and the thermodynamic limit (TDL) of $N \rightarrow \infty$ at a constant density parameter r_s [69–71]. This is particularly important for multicomponent systems consisting of both electrons and nuclei, for which finite-size corrections are substantially less developed compared to the more simple uniform electron gas (UEG) [72–77]. In addition, a large box length L (assuming a cubical simulation cell of volume $\Omega =$

L^3) allows to access small wavenumbers q , since the minimum wavenumber is given by $q_{\min} = 2\pi/L$ due to the momentum discretization. This is crucial for the description of x-ray scattering experiments in a forward scattering geometry [78–81] that probe collective modes such as the plasmon oscillation [82], and might eventually even facilitate the estimation of optical properties that are defined in the limit of $q \rightarrow 0$ [15]. On the other hand, PIMC simulations of quantum many-body systems with $N = 1000$ (and $P \sim 10^2$ replicas of each particle on different imaginary-time slices; see below) and beyond become computationally demanding even without the exponential bottleneck due to the sign problem. Specifically, the main computational expense is given by the evaluation of the long-range pair interaction, which is typically treated with the usual Ewald sum over all periodic images [69].

A vastly cheaper alternative is given by spherically averaged pair potentials, which have been averaged over different angular orientations of the periodic array of images of the particles in the main simulation cell [70,83–86]; see also the review by Fukuda and Nakamura [87] for an overview of non-Ewald methods. Here, we explore the spherically averaged pair potential introduced by Yakub and Ronchi (YR) [83,84]. Demyanov *et al.* [70] applied the YR potential to Monte Carlo simulations of the classical one-component plasma that consists of negative unit charges in a homogeneous neutralizing background and found a comparable convergence of the internal energy to the standard Ewald potential. In addition, V. Filinov *et al.* [88–90] used the YR potential to compute the internal energy (and its correlation contribution) in PIMC simulations of the UEG; the quantum analog of the classical one-component plasma. Very recently, A. Filinov and Bonitz [91] tested the Ewald and the YR potentials in PIMC simulations of warm dense hydrogen and “... found that, for the simulation parameters used in this paper, the results for both are practically indistinguishable.” However, these references were limited to integrated properties such as energies and pressure, sometimes extending to spatial observables such as cluster analyses. The effect of the pair potential onto wavenumber-resolved properties has, thus, remained unclear. This is unfortunate for (at least) two reasons: (i) the invariance of the electronic static structure factor $S(\mathbf{q})$ with respect to the system size even for $N \sim 10$ is of key importance for finite-size corrections, e.g., of the interaction energy W [73,75] and has been pivotal for the construction of accurate parametrizations of the UEG in the TDL [24,26,72,75]; (ii) the invariance with respect to N of species-resolved static structure factors $S_{ab}(\mathbf{q})$ and related imaginary-time correlation functions (ITCFs) $F_{ab}(\mathbf{q}, \tau)$, where $t = -i\hbar\tau$ is the imaginary time with $\tau \in [0, \beta]$, in PIMC simulations of real WDM systems such as hydrogen [92,93] and beryllium [35,94] facilitates the comparison with experimental observations, most notably x-ray Thomson scattering (XRTS) measurements [37,94].

In the present work, we remedy this situation by presenting a rigorous and detailed analysis of the effect of the YR potential compared to the standard Ewald sum onto different properties of the UEG, including different energies, static structure factor, and linear/quadratic static density response functions [95,96]. Finally, we carry out an analytic continuation [97–99] to compute the dynamic structure factor $S(\mathbf{q}, \omega)$ for selected wavenumbers. As a general trend, we find that

the standard Ewald and YR potentials do indeed give comparable results for integrated properties such as the kinetic and potential energy. However, use of the YR potential breaks the N invariance of q -resolved properties (in particular at small wavenumbers), which constitutes a serious limitation for comparisons with experiments and finite-size corrections.

The paper is organized as follows: In Sec. II, we provide the theoretical background with a focus on the Ewald and YR pair potentials. In Sec. III, we present our PIMC simulation results on the warm dense UEG at $r_s = 3.23$ (Sec. III A), the electron liquid at $r_s = 10$ (Sec. III B), the analytic continuation to obtain the dynamic structure factor (Sec. III C), and the static quadratic density response $\chi^{(2)}(\mathbf{q}, 0)$ (Sec. III D). The paper is concluded by a summary and outlook in Sec. IV.

II. THEORY

The *ab initio* PIMC method [41] is based on the celebrated quantum-to-classical mapping [100] and has been described extensively in the literature [41,101,102]. We limit ourselves to a brief summary of our implementation and its features. We assume hartree atomic units.

The main advantage of PIMC is its capability to provide quasixact (i.e., exact within the given Monte Carlo error bars) results for any thermodynamic observable for the specified quantum N -body problem. In practice, an important convergence parameter is given by the number of imaginary-time propagators P . Here, we use the primitive high-temperature factorization $e^{-\epsilon\hat{H}} \approx e^{-\epsilon\hat{K}} e^{-\epsilon\hat{V}}$, with $\epsilon = \beta/P$ and $\hat{K}, \hat{V}$ the kinetic and potential contributions to the full Hamiltonian $\hat{H}$, respectively. The convergence with P is discussed in Sec. III. To ensure ergodic sampling of all relevant path configurations, we use the canonical extended ensemble adaption of the worm algorithm by Boninsegni *et al.* [101,103] presented in Ref. [104], which is implemented in the open-source *ishtar* code [105]. Finally, we consider electrons, which obey Fermi-Dirac statistics. The antisymmetry of the thermal density matrix under the exchange of particle coordinates is taken into account by sampling the permutation structure [102] without any nodal restrictions. Hence, our simulations are computationally costly due to the fermion sign problem [44], but exact within error bars. Moreover, we retain full access to equilibrium system dynamics in the form of various imaginary-time correlation functions [96].

We consider $N = N^\uparrow + N^\downarrow$ (with $N^\uparrow = N^\downarrow$) electrons in a cubic simulation cell of volume $\Omega = L^3$ and standard periodic boundary conditions. To take into account the long-range nature of the Coulomb interaction, we consider the usual infinite array of periodic images; the corresponding solution to Poisson's equation is then given by the Ewald sum [69]. Following the notation by Fraser *et al.* [69], we express the Ewald pair interaction as

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

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* including the customary factor of 2π . We note that κ —often coined as the Ewald parameter—can be chosen freely to shift weights between the sums over $\mathbf{G}$ and $\mathbf{n}$, while the fully converged result is invariant. In practice, both sums have to be truncated, which affects the computation cost. For completeness, we note that a vast literature exists on efficient numerical implementation of Eq. (1) for Monte Carlo simulations; see, e.g., Refs. [106–109] and references therein. The full Hamiltonian of the UEG with Ewald interaction is given by

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

with $\xi_M = -2.837\,297(3/4\pi)^{1/3}N^{-1/3}r_s^{-1}$ the Madelung constant [16] that takes into account the interaction between a charge and its own background and periodic array of self-images.

Angular averaging over all possible orientations of the infinite periodic array of images leads to a considerable simplification of Eq. (1). For the one-component plasma, the Hamiltonian is given by [70]

$$\hat{H}_{\text{YR}} = -\frac{1}{2} \sum_{l=1}^N \nabla_l^2 + \sum_{l < k}^N \sum_{\mathbf{n}} \phi_{\text{YR}}(\hat{\mathbf{r}}_l, \hat{\mathbf{r}}_k + \mathbf{n}L) - \frac{3}{4r_{\text{cut}}} \left(1 + \frac{N}{5}\right), \quad (3)$$

with the definition of the YR potential [83,84]

$$\phi_{\text{YR}}(\mathbf{r}_a, \mathbf{r}_b) = \begin{cases} \frac{1}{r_{ab}} \left\{ 1 + \frac{1}{2} \left(\frac{r_{ab}}{r_{\text{cut}}} \right) \left[\left(\frac{r_{ab}}{r_{\text{cut}}} \right)^2 - 3 \right] \right\}, & \text{if } r_{ab} < r_{\text{cut}} \\ 0, & \text{otherwise,} \end{cases} \quad (4)$$

where $r_{ab} = |\mathbf{r}_b - \mathbf{r}_a|$. The cutoff radius follows from the volume-equivalent sphere of the main cell, $r_{\text{cut}} = L(3/4\pi)^{1/3}$. We note that, by definition, the sum over periodic images $\mathbf{n}$ in Eq. (3) can be truncated for $|\mathbf{n}| \geq 1.62$; still, it is possible that the YR pair potential [Eq. (4)] does give contributions for more than one image (cf. the blue areas in Fig. 1); see also the discussion in Ref. [83].

Demyanov *et al.* [70] report an acceleration by 2 orders of magnitude of the YR over the Ewald potential in their classical MC simulations; we find a similar acceleration of $\mathcal{O}(10^1 - 10^2)$ for PIMC. We further point out that typical PIMC simulations deal with $N \sim 10 - 100$ particles, with replicas on $P \sim 100$ imaginary-time slices. This often limits the utility of sophisticated multipole expansions of the Ewald

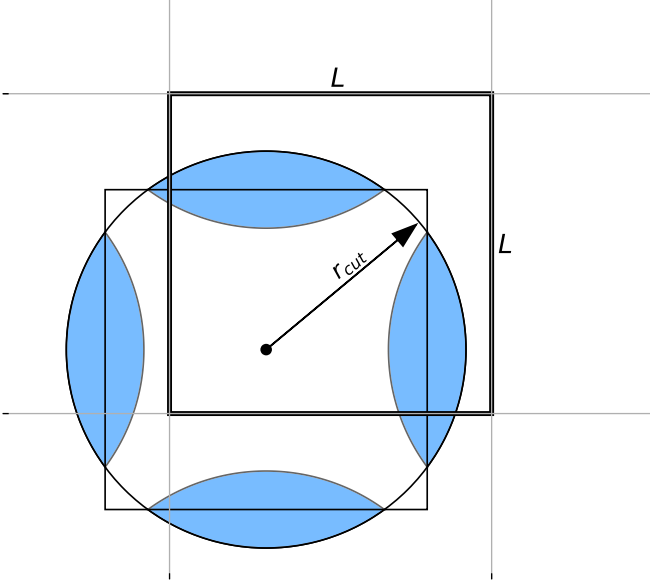


FIG. 1. Schematic illustration of the YR potential, Eq. (4). A random particle in the main simulation cell of length L interacts with all particles and their periodic images within the volume equivalent sphere of radius r_{cut} . This leads to a mandated double counting of both an original particle and its image in the shaded blue areas. Adapted from Ref. [83].

potential, which, despite having a favorable scaling with N , often start becoming more efficient for $N \sim 1000$. At the same time, the numerical evaluation of Eq. (1) is usually responsible for the main fraction of the compute time in PIMC simulations of periodic Coulomb systems such as the UEG, which makes any possible acceleration highly desirable.

For completeness, we note that the application of spherically averaged potentials is well justified for disordered systems such as warm dense matter, but will likely break down when long-range correlations are being introduced into the system; this limits their application to the UEG to $r_s \lesssim 50$ at $\Theta = 1$, and their applicability should be checked carefully even in the strongly coupled electron liquid regime.

III. RESULTS

All PIMC results presented in this work have been obtained using the open-source *ishtar* code [105], and are freely available in an online repository [110].

A. Warm dense matter

As the first set of relevant conditions, we consider the UEG at $r_s = 3.23$ and $\Theta = 1$ (i.e., $T = 4.8$ eV). These fall into the WDM regime and can be realized, e.g., in experiments with optically pumped hydrogen jets [113,114]. We note that Hamann *et al.* [115] have predicted the emergence of a roton-type feature in the electronic dynamic structure factor of hydrogen at these parameters. This is a consequence of the partial ionization of hydrogen in this regime [116] leading to an effectively lower free-electron density, whereas the roton-type feature is absent in the UEG at $r_s = 3.23$.

In the top panel of Fig. 2, we study the convergence of the static structure factor $S(\mathbf{q})$ (computed via the YR potential)

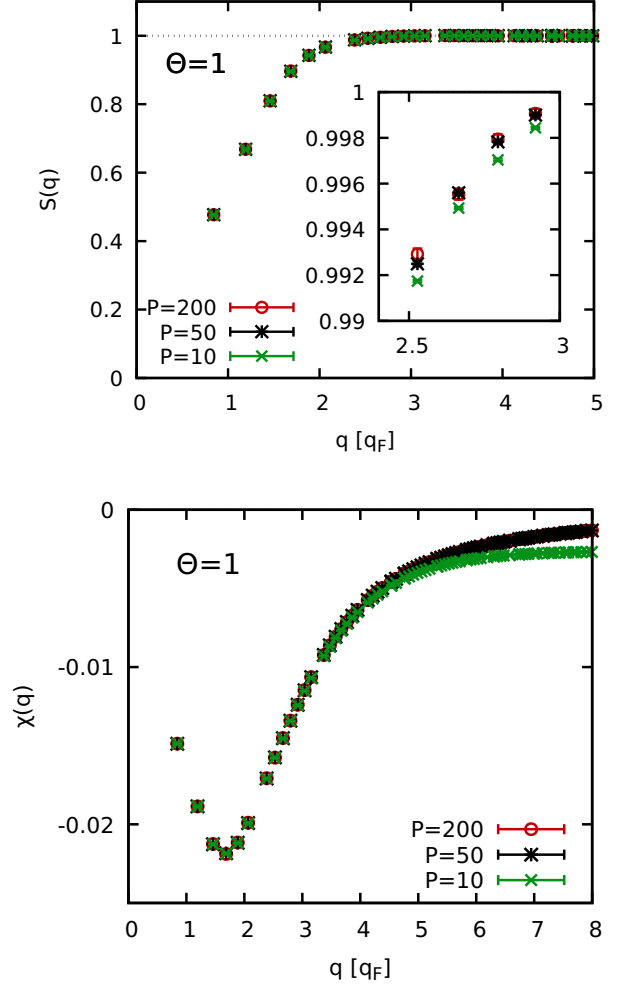


FIG. 2. PIMC results for the static structure factor $S(\mathbf{q})$ (top) and the static linear density response function $\chi(\mathbf{q})$ [bottom, cf. Eq. (5)] of the unpolarized UEG at $r_s = 3.23$, $\Theta = 1$, with $N = 14$ and the YR pair potential. Different symbols and colors distinguish different numbers of imaginary-time propagators P . The top panel inset shows a magnified segment.

with the number of imaginary-time propagators P . We only find systematic bias for $P = 10$ (green crosses), whereas, e.g., $P = 50$ (black stars) and $P = 200$ (red circles) cannot be distinguished within the given error bars. This can be discerned particularly well in the inset that shows a magnified segment. Indeed, even for $P = 10$ the convergence error is of the order of $\sim 0.1\%$. In the bottom panel of Fig. 2, we repeat this analysis for the static linear density response function $\chi(\mathbf{q})$. In principle, the latter can be obtained by applying a small harmonic perturbation to the system and measuring its density response [117–124]. However, this procedure requires individual simulations for multiple perturbation amplitudes for each value of q , which rules out extensive parameter scans. A more convenient and more elegant alternative is given by the imaginary-time version of the fluctuation-dissipation theorem [125]:

$$\chi(\mathbf{q}) = -n \int_0^\beta d\tau F(\mathbf{q}, \tau), \quad (5)$$

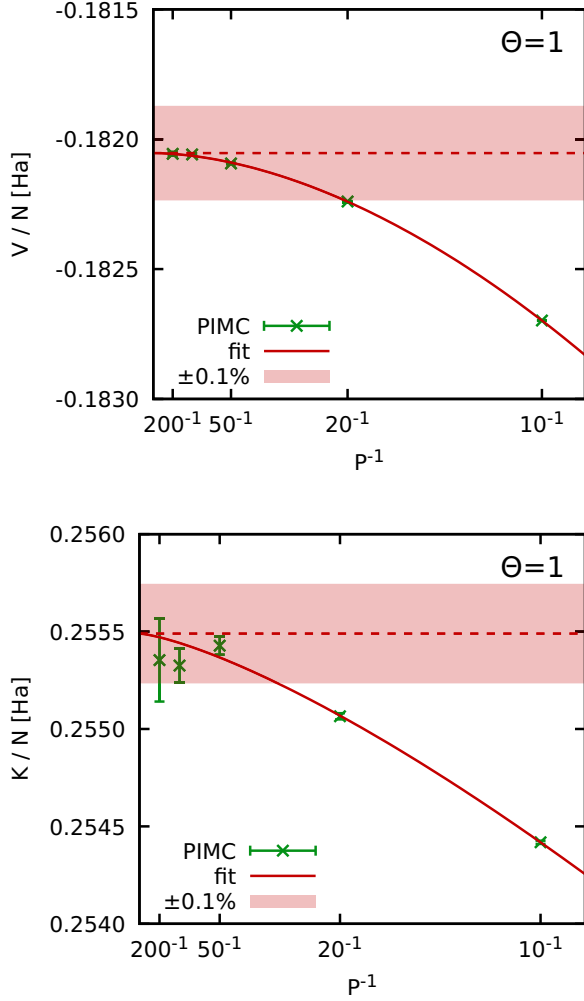


FIG. 3. The convergence of the potential energy V (top) and kinetic energy K (bottom), for $N = 14$ unpolarized electrons interacting with the YR potential at $r_s = 3.23$, $\Theta = 1$, with the number of imaginary-time propagators P . The solid red lines show empirical fits of the form $f(x) = a + bx^c$ that have been included as a guide to the eye.

which implies that one can obtain the full q dependence from a single simulation of the unperturbed system from the imaginary-time density-density correlation function $F(\mathbf{q}, \tau) = \langle \hat{n}(\mathbf{q}, \tau) \hat{n}(-\mathbf{q}, 0) \rangle$. We find the equivalent convergence behavior with respect to P for $\chi(\mathbf{q})$ as for $S(\mathbf{q})$, except in the limit of large q ; here, the $P = 10$ datasets drastically deviate from the rest. This, however, is not primarily a factorization error of the density operator $\hat{\rho} = e^{-\beta \hat{H}}$, but rather a quadrature error in the evaluation of the integral of Eq. (5), since $F(\mathbf{q}, \tau)$ can only be estimated on the given P imaginary-time slices. In Fig. 3, we present a similar convergence analysis for the potential (top) and kinetic energy (bottom). In both cases, the present choice of $P = 200$ primitive imaginary-time propagators is fully sufficient to even resolve small differences in the respective observables. We thus safely conclude that $P = 200$ can faithfully resolve even small effects due to the considered pair potentials, Eqs. (1) and (4).

Let us next come to the topic at hand, which is the comparison of simulation results between the Ewald potential [Eq. (1)] and the YR potential [Eq. (4)]. In the top row of Fig. 4, we compare the static structure factors, with the left and right panels corresponding to the former and the latter, respectively. The solid red curve has been computed within the *effective static approximation* (ESA) [111,112], which is known to perform well at these conditions, and has been included as a reference. The green crosses, black stars, and blue diamonds have been obtained from PIMC simulations of $N = 14$, $N = 20$, and $N = 30$ unpolarized electrons, respectively. For completeness, we mention that we find an average sign of $S \approx 0.05$ for $N = 30$, precluding the simulation of much larger systems due to the fermion sign problem [44]. There are no apparent finite-size effects in the Ewald data (see also the magnified segment in the inset), except for the different q grids; this is a well-known consequence of momentum quantization in the finite simulation cell [75]. This is expected based on previous investigations of q -resolved properties [73–77]. In stark contrast, the results for $S(\mathbf{q})$ that have been obtained using the spherically averaged YR potential exhibit significant oscillations around the red ESA reference curve; this can again be discerned particularly well in the magnified inset. The reason for these oscillations directly follows from the definition of the YR potential, Eq. (4). Recall that particles around a distance of r_{cut} interact twice with a reference particle; cf. Fig. 1. Consequently, the probability of finding two particles within such a distance gets reduced due to the additional potential-energy penalty. The corresponding wavenumber $q_{\text{cut}} = 2\pi/r_{\text{cut}}$ for $N = 30$ electrons is indicated by the blue arrow at the bottom and by the vertical blue line in the inset of the top right panel of Fig. 4. As it is expected, the static structure factor is reduced in this segment (shaded blue area in the inset). This reduction is then compensated by the somewhat higher values of $S(\mathbf{q})$ around this segment, which explains the observed oscillations. Finally, we point out that the Ewald and YR potentials lead to indistinguishable results for $q \gg q_{\text{cut}}$.

In the bottom row of Fig. 4, we repeat this analysis for the static linear density response function $\chi(\mathbf{q})$; cf. Eq. (5). Overall, we find the same qualitative trends as for $S(\mathbf{q})$, i.e., no finite-size effects for Ewald and oscillations around the ESA reference curve for YR at $q \sim q_{\text{cut}}$. Indeed, Eq. (5) implies a direct connection between the static structure factor and the linear static density response function as it holds $F(\mathbf{q}, 0) = S(\mathbf{q})$. The observed oscillations in $\chi(\mathbf{q})$ thus imply that the reduction in correlations around q_{cut} persist throughout the imaginary-time diffusion that is encoded in $F(\mathbf{q}, \tau)$.

To further elucidate this observation, we show the full τ dependence of the ITCF in Fig. 5 for $N = 14$ and $N = 30$ electrons. In particular, the different colors correspond to different wavenumbers q , and the solid and dashed lines have been computed using the Ewald and YR potential, respectively; the associated Monte Carlo error bars are shown as shaded areas around these curves, but are barely visible due to the high data quality. We note that, in addition to its utility for the estimation of $\chi(\mathbf{q})$ via Eq. (5), the ITCF has attracted considerable recent attention. For example, it constitutes the starting point for the analytic continuation to compute the dynamic structure factor $S(\mathbf{q}, \omega)$ (see Sec. III C

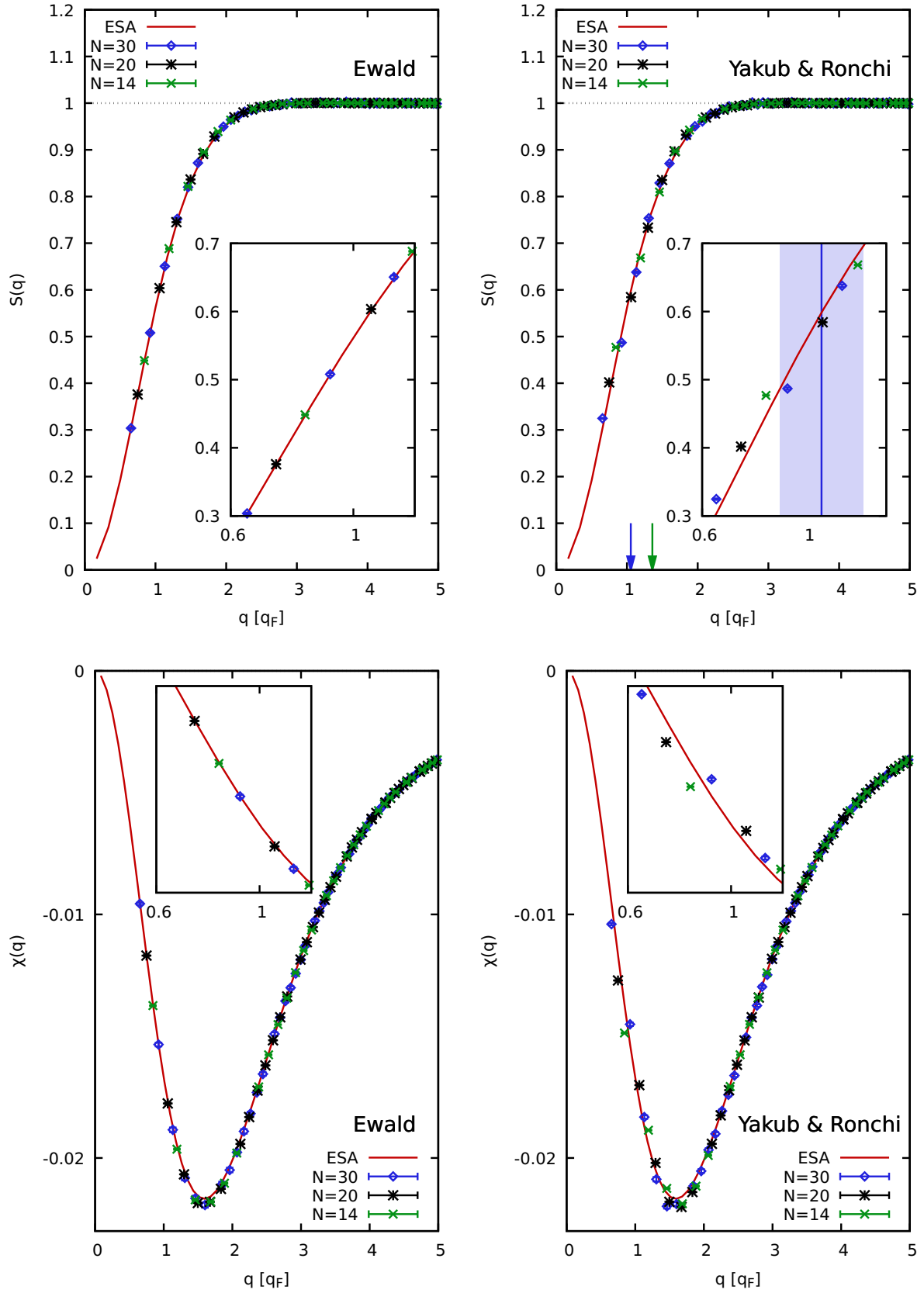


FIG. 4. PIMC results for the static structure factor $S(\mathbf{q})$ (top row) and the static linear density response function $\chi(\mathbf{q})$ [cf. Eq. (5), bottom row] at $r_s = 3.23$ and $\Theta = 1$. Symbols: PIMC data for different N computed with the Ewald potential (left) and the YR potential (right). Solid red: Effective static approximation (ESA) [111,112], included as a reference. The insets show magnified segments.

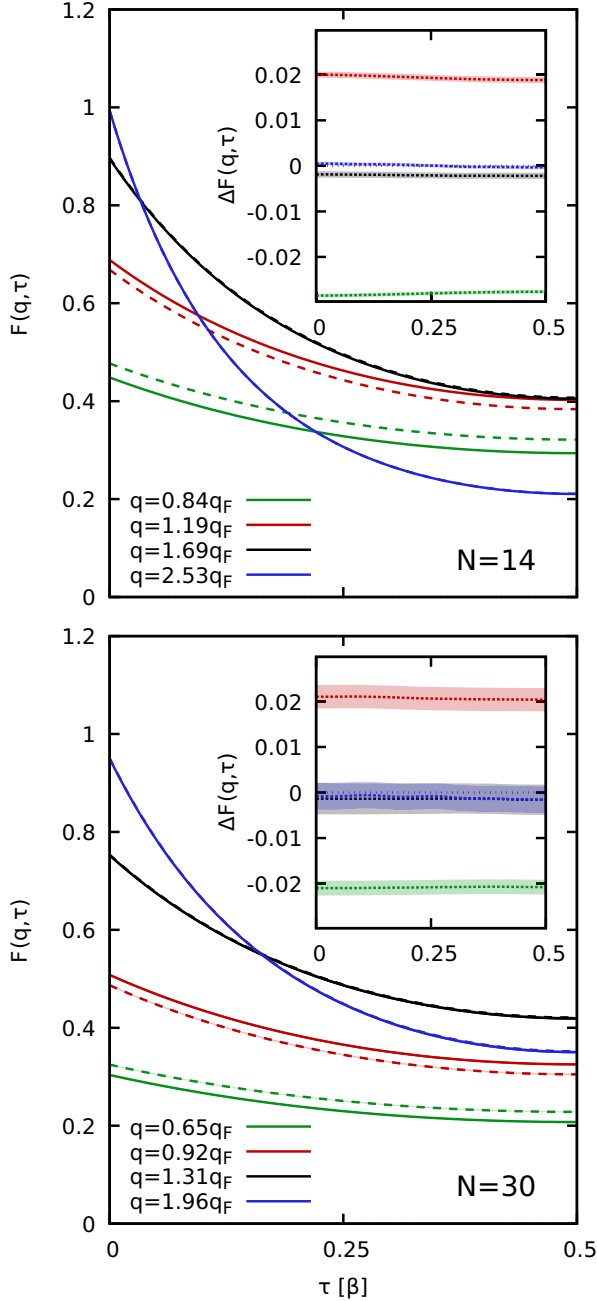


FIG. 5. PIMC results for the density-density ITCF $F(\mathbf{q}, \tau)$ of the UEG at $r_s = 3.23$, $\Theta = 1$, with $N = 14$ (top) and $N = 30$ (bottom). The solid and dashed lines have been computed using the Ewald and YR pair potentials, respectively. The insets show the absolute difference between the two, i.e., $\Delta F(\mathbf{q}, \tau) = F_E(\mathbf{q}, \tau) - F_{YR}(\mathbf{q}, \tau)$. The shaded areas indicate the given statistical uncertainty.

below), and contains rich information, e.g., about quasiparticle excitations [125], frequency moments of $S(\mathbf{q}, \omega)$ [126], and quantum delocalization [127]. Moreover, the ITCF has emerged as a standard tool for the interpretation of XRTS experiments and allows for the model-free estimation of the temperature [36,81,128,129], the absolute intensity [130], and the Rayleigh weight [35]. Extensive discussions of its physical meaning have been presented in Refs. [124,125,127] and need not be repeated here.

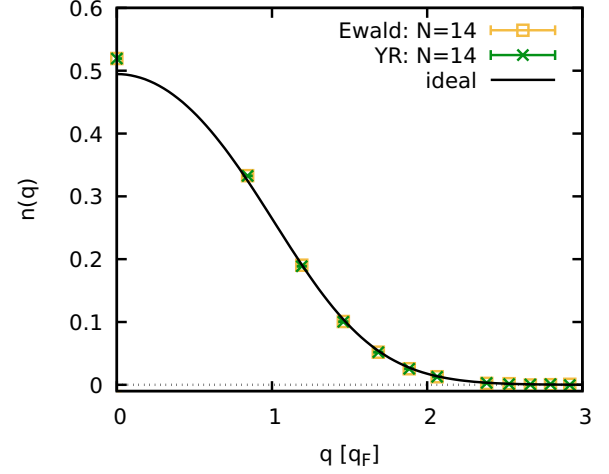


FIG. 6. PIMC results for the momentum distribution $n(\mathbf{q})$ of the UEG with $N = 14$ at $r_s = 3.23$, $\Theta = 1$. The yellow squares and green crosses have been computed using the Ewald and YR pair potentials, respectively. The solid black line corresponds to the Fermi distribution that describes the ideal Fermi gas at the same conditions.

Overall, we find that the effect of the YR potential onto $F(\mathbf{q}, \tau)$ is nearly independent from τ at these conditions and manifests as a constant shift. This can be discerned particularly well in the insets, which illustrate the absolute difference $\Delta F(\mathbf{q}, \tau) = F_E(\mathbf{q}, \tau) - F_{YR}(\mathbf{q}, \tau)$; they are constant within the given uncertainty intervals (shaded areas). The effect of these differences onto the analytic continuation is discussed in Sec. III C below.

An additional interesting question concerns the effect of the selected pair potential on the momentum distribution function $n(\mathbf{q})$. The latter is an off-diagonal observable within the PIMC formalism [41,131] and can be conveniently estimated (including its proper normalization) using the extended ensemble approach introduced in Ref. [104]. The corresponding PIMC simulation results for $N = 14$ are shown in Fig. 6, with the yellow squares and green crosses obtained with the Ewald and YR pair potential, respectively. In addition, we have included the Fermi distribution function that describes the momentum distribution of the ideal Fermi gas (solid black curve) as a reference. Interestingly, we find an increase in the occupation of the zero-momentum state with respect to the ideal result; this is connected with a potential XC-induced lowering of the kinetic energy at finite temperatures for some densities, and has been discussed in detail in Refs. [104,132–134]. In the context of the present work, the key question concerns the effect of the pair potential; no differences between the Ewald and YR results can be resolved within the Monte Carlo error bars, even for $N = 14$. In fact, this observation is not unexpected: $S(\mathbf{q})$ and $F(\mathbf{q}, \tau)$ [from which we also estimate $\chi(\mathbf{q})$] are two-body observables and, thus, subject to the double-counting effect discussed above. In contrast, $n(\mathbf{q})$ is obtained from the Fourier transform of the single-particle density matrix, which is computed from correlations between the ends (usually denoted as *head* and *tail*) of an open imaginary-time trajectory. In particular, the head and tail do not interact with each other and are thus not directly affected by the double counting for any system size.

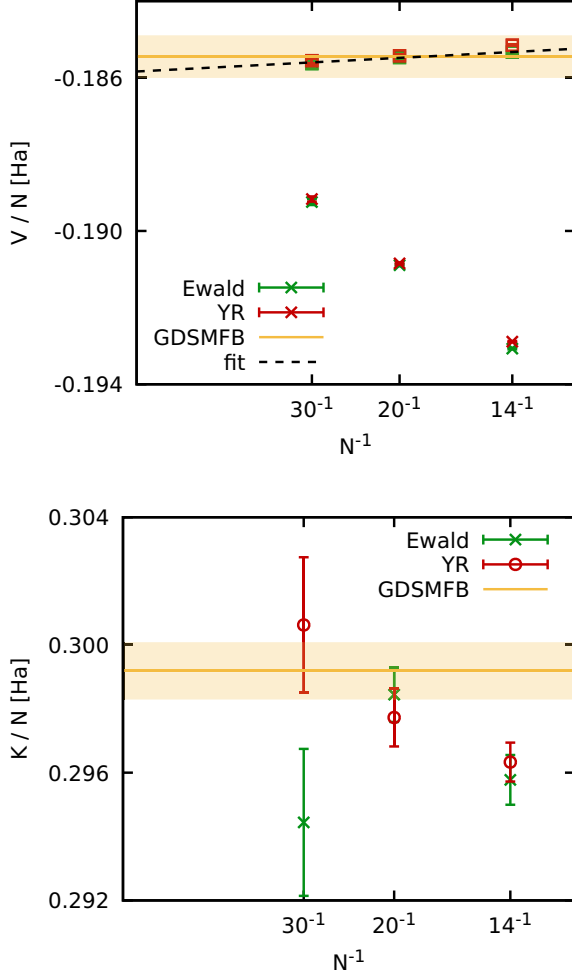


FIG. 7. (Top) Potential energy per electron V/N computed with the Ewald (green) and YR pair potentials (red); crosses and squares show raw PIMC results, and finite-size corrected points via Eq. (6); the dashed black lines correspond to an empirical linear fit. (Bottom) Kinetic energy per electron K/N . All results are for the UEG at $r_s = 3.23$, $\Theta = 1$. The horizontal yellow lines and shaded yellow areas correspond to the parametrization by Groth *et al.* [25] (GDSMFB) and its nominal uncertainty of $\pm 0.3\%$.

Let us conclude the analysis of the warm dense UEG at $r_s = 3.23$ and $\Theta = 1$ by investigating the effect of the pair potential onto integrated properties. As two representative examples, we consider the potential and kinetic contributions to the energy in the top and bottom panels of Fig. 7, respectively. Both datasets (green and red crosses) exhibit the same qualitative behavior and we can only resolve small differences for $N = 14$. This is unsurprising, as the potential energy is given by a sum over $S(\mathbf{q})$ and, hence, the oscillations in the latter cancel to a large degree. The green and red squares have been obtained by adding to the raw PIMC results the first-order finite-size correction that was first presented by Chiesa *et al.* [73] at $T = 0$ and subsequently generalized by Brown *et al.* [52] to finite temperatures:

$$\Delta V(N) = \frac{\omega_p}{4N} \coth\left(\frac{\beta\omega_p}{2}\right); \quad (6)$$

the plasma frequency is given by $\omega_p = \sqrt{3/r_s^3}$. Evidently, Eq. (6) works well at the present conditions and removes most of the N dependence from both datasets; see also the extensive discussion in Ref. [72]. We perform an empirical linear fit to the green squares (see the dashed black curve), which works well. Moreover, the extrapolation to the thermodynamic limit of $N^{-1} \rightarrow 0$ lies within the $\pm 0.3\%$ nominal uncertainty range (shaded yellow area) of the parametrization by Groth *et al.* [25] (GDSMFB).

For the kinetic energy K/N (Fig. 7, bottom panel), no deviations between the two potentials can be resolved. This might be a consequence of either the reduced sensitivity of single-particle operators to two-body correlations (see Fig. 6 above), or simply be masked by the comparably larger error bars—a well-known issue in PIMC simulations that originates from the large number of imaginary-time propagators P [135]. We note that the error bars further increase with N due to the fermion sign problem [44]; the same holds for the interaction energy, where this trend is, however, less obvious due to the overall much lower variance of the estimator. Overall, we observe that our PIMC results for K/N are consistent with the GDSMFB result; this indeed also holds for the Ewald result for $N = 30$, which is within the 2σ range with respect to the parametrization. At the same time, a conclusive quantitative statement is ruled out by the statistical uncertainty.

B. Electron liquid

As a second example, we consider the UEG at $r_s = 10$ and $\Theta = 1$. These conditions are located at the boundary of the strongly coupled electron liquid regime [3,136], which gives rise to a roton-type feature in the dynamic structure factor $S(\mathbf{q}, \omega)$ that has attracted significant recent attention [97,137–141].

In the top and bottom rows of Fig. 8, we analyze the effect of the potential onto $S(\mathbf{q})$ and $\chi(\mathbf{q})$, respectively, and find the same qualitative trends as for $r_s = 3.23$. In Fig. 9, we show a magnified segment around the maximum in $S(\mathbf{q})$. On this scale, we can clearly resolve small yet significant systematic errors in the ESA result. The YR potential is generally capable of resolving both the position and the height of the maximum, although small deviations from the Ewald results are visible even for $N = 34$; the gray triangles showing the YR results for $N = 54$, on the other hand, cannot be distinguished from the Ewald results for these q values. In practice, the convergence of any YR data set with respect to N thus has to be carefully checked for any given q range of interest.

Interestingly, the ITCF $F(\mathbf{q}, \tau)$, which is analyzed in Fig. 10, is more strongly affected by the YR potential than at $r_s = 3.23$ (cf. Fig. 5 above). In particular, the deviations $\Delta F(\mathbf{q}, \tau)$ between the two results, which are shown in the insets, are no longer constant with respect to τ , in particular for $N = 14$. The implications of these findings for the analytic continuation to the dynamic structure factor $S(\mathbf{q}, \omega)$ are investigated in Sec. III C below.

Finally, we analyze the potential and kinetic energies per particle in the top and bottom panels of Fig. 11. In the former, we find systematic deviations of $\sim 0.1\%$ for $N \lesssim 34$, which is still acceptable for many practical applications. In the latter, again, no systematic differences between the two

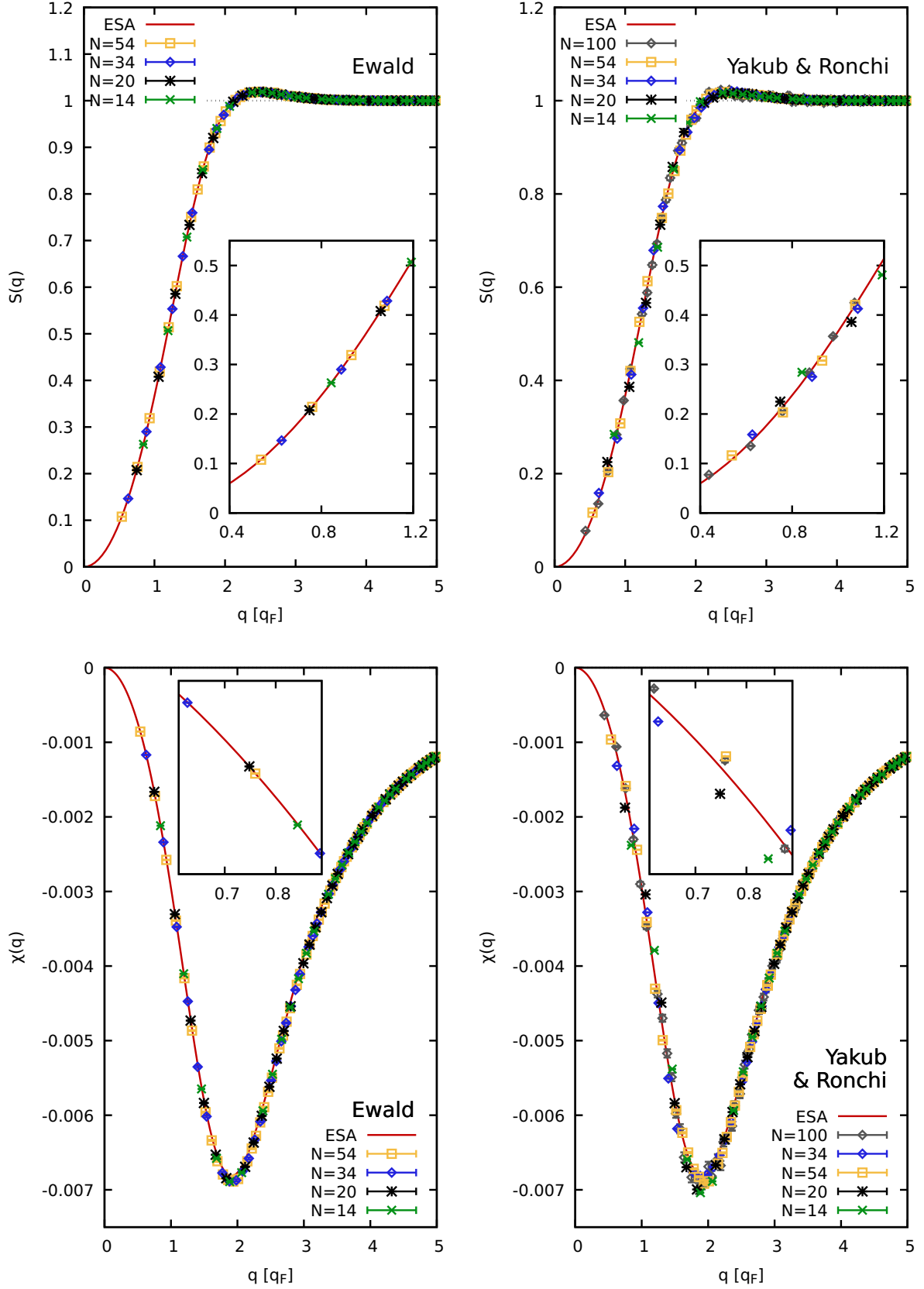


FIG. 8. PIMC results for the static structure factor $S(\mathbf{q})$ (top row) and the static linear density response function $\chi(\mathbf{q})$ [cf. Eq. (5), bottom row] at $r_s = 10$, $\Theta = 1$. Symbols: PIMC data for different N computed with the Ewald (left) and the YR potential (right). Solid red: ESA [111,112], included as a reference. The insets show magnified segments.

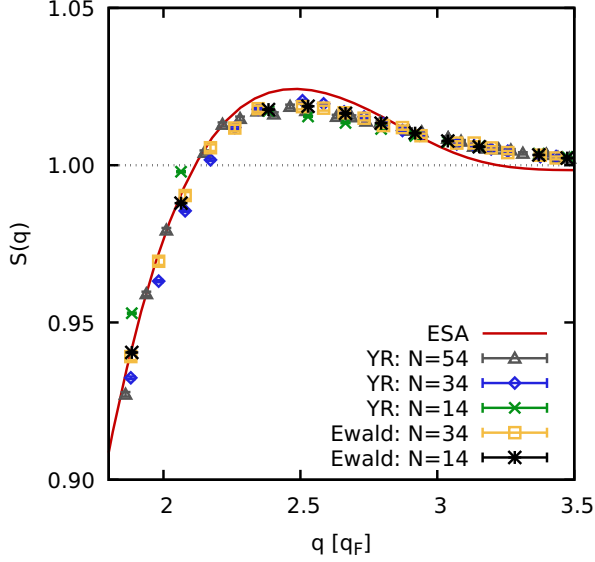


FIG. 9. Magnified segment from Fig. 8 in the vicinity of the static structure factor maximum $S(q)$ at $r_s = 10$ and $\Theta = 1$.

pair potentials can be resolved. For completeness, we have performed an empirical linear fit of the Ewald result; see the dashed black line. The thus extrapolated result for the thermodynamic limit is shown by the bold blue point, and is consistent with GDSMF within the given uncertainty.

C. Sensitivity of analytic continuation

In what follows, we analyze the effect of the pair potential onto analytic continuation results for the dynamic structure factor $S(\mathbf{q}, \omega)$. The latter is connected to the ITCF via a two-sided Laplace transform [125],

$$F(\mathbf{q}, \tau) = \int_{-\infty}^{\infty} d\omega S(\mathbf{q}, \omega) e^{-\tau\omega}, \quad (7)$$

which needs to be inverted numerically; a notoriously difficult, exponentially ill-posed problem [99]. For the UEG, Dornheim and co-workers [97,142,143] have presented highly accurate results for $S(\mathbf{q}, \omega)$ by incorporating a number of additional exact constraints [144], many of which, however, are known only for the UEG but not for realistic WDM systems. Very recently, Chuna *et al.* [98,145] have applied the all-purpose maximum entropy method (MEM) to the UEG and found that it gives very accurate results for $S(\mathbf{q}, \omega)$ with high-quality PIMC input data for $F(\mathbf{q}, \tau)$ and an appropriate choice for the Bayesian prior $\mu(\omega)$. Here, we consider $r_s = 10$ and $\Theta = 1$, for which we attain Monte Carlo error bars of an absolute magnitude of $\delta F(\mathbf{q}, \tau) \sim 10^{-5}$. Moreover, we choose the random-phase approximation (RPA) for $\mu(\omega)$, which is (a) an appropriate choice for $r_s = 10$ [145] and (b) constitutes an equal prior for Ewald and YR potentials, allowing for a clear comparison between the two pair potentials. The analytic continuation for realistic electron-ion systems such as warm dense hydrogen will require careful dedicated future works.

In Fig. 12, we show the thus reconstructed dynamic structure factors for six relevant wavenumbers q , with the blue

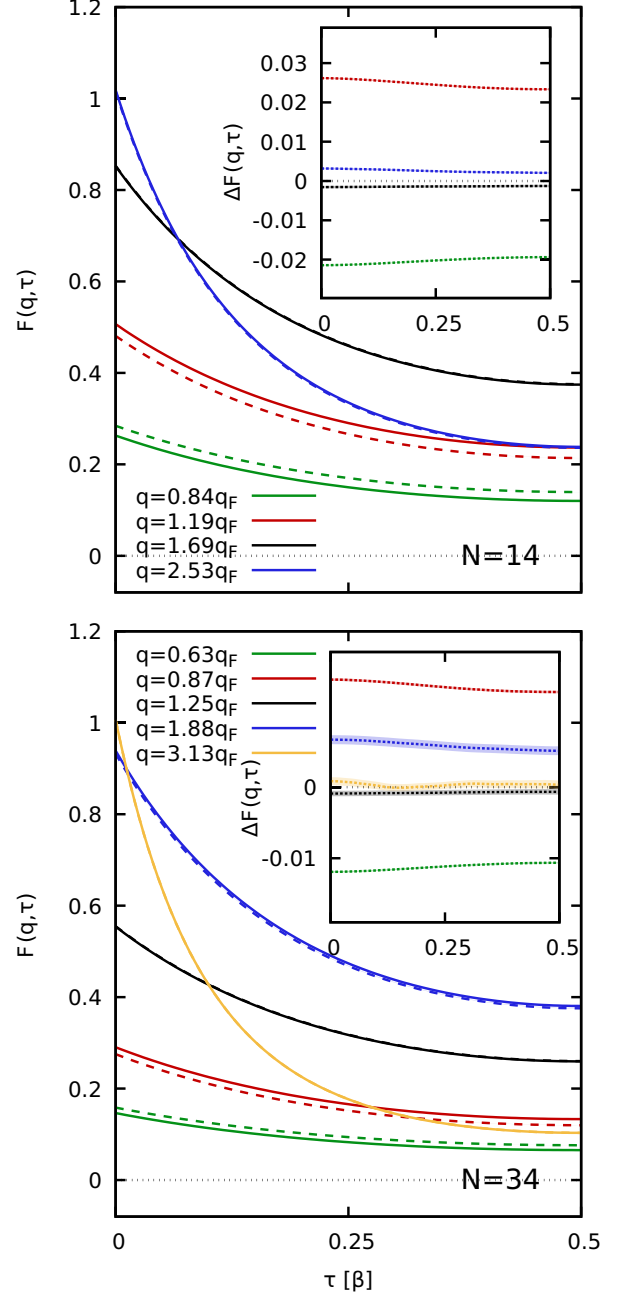


FIG. 10. PIMC results for the density-density ITCF $F(\mathbf{q}, \tau)$ of the UEG at $r_s = 10$, $\Theta = 1$, with $N = 14$ (top) and $N = 34$ (bottom). The solid and dashed lines have been computed using the Ewald and the YR pair potentials, respectively. The insets show the absolute difference between the two, i.e., $\Delta F(\mathbf{q}, \tau) = F_E(\mathbf{q}, \tau) - F_{YR}(\mathbf{q}, \tau)$. The shaded areas indicate the given statistical uncertainty.

and red curves corresponding to the Ewald and YR potentials, respectively. We note that the shaded areas around these curves show the uncertainty that is inherent in the inversion of Eq. (7), which takes into account both the statistical noise in the PIMC data for $F(\mathbf{q}, \tau)$ via a leave-one-out method and the effect of averaging over regularization parameters in the entropy term that relates the reconstructed $S(\mathbf{q}, \omega)$ to the default model $\mu(\omega)$; see Ref. [145] for an extensive discussion

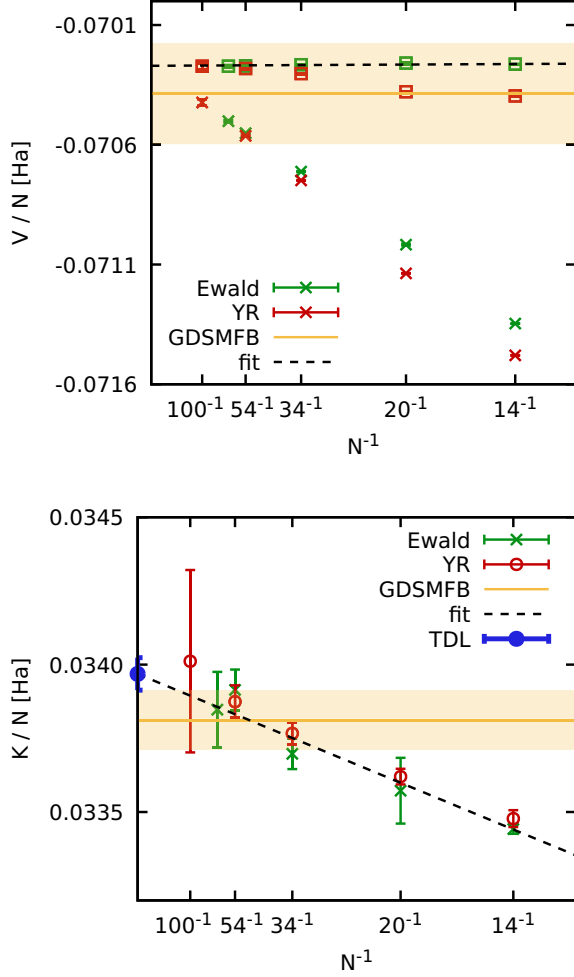


FIG. 11. (Top) Potential energy per electron V/N computed with the Ewald (green) and the YR pair potential (red); crosses and squares show raw PIMC results, and finite-size corrected points via Eq. (6); the dashed black lines correspond to an empirical linear fit. (Bottom) Kinetic energy per electron K/N . All results are for the UEG at $r_s = 10$, $\Theta = 1$. The horizontal yellow lines and shaded yellow areas correspond to the parametrization by Groth *et al.* [25] (GDSMFB) and its nominal uncertainty of $\pm 0.3\%$.

of this procedure. Finally, the purple curves show the RPA default model.

As a sanity check, we demonstrate that our MEM implementation perfectly reproduces the PIMC input data for four of the considered wavenumbers. This is illustrated in Fig. 13, where the solid and dashed lines correspond to the PIMC ITCF with Ewald and YR potentials, while the deep-purple squares and crosses correspond to the ITCF as obtained by plugging the reconstructed dynamic structure factors into Eq. (7).

It is evident that there are significant differences in the MEM results for $S(\mathbf{q}, \omega)$ itself. For $q = 0.63q_F$ (top left), we find that the utilization of the YR potential further dampens the plasmon, which is located at around $q \approx 1.2\omega_p$, and which is already damped with the Ewald potential compared to RPA. Moreover, YR leads to a substantial shift of spectral weight to lower frequencies. This is a direct consequence of the

up-shift in the YR ITCF compared to the Ewald ITCF at this wavenumber; see Fig. 10. In fact, it is easy to see that a constant up-shift in the ITCF is associated with a contribution to the dynamic structure factor at $\omega = 0$; this is highly relevant for two-component systems where the Rayleigh weight that describes the spectral weight of the ion feature manifests as such a shift [93,116]. In the present case, the difference in the ITCF between the Ewald and YR calculations exhibits a weak dependence on τ , which then leads to a shift of spectral densities to smaller, but still finite, frequencies.

For $q = 0.87q_F$ (top right), we find the opposite trend. In this case, the YR plasmon is less damped than the Ewald result, and spectral weight is shifted to higher frequency. Again, this trend directly follows from the ITCF, in which the YR potential manifests as a down-shift in this case. In the middle row of Fig. 12, we show results for $q = 1.25q_F$ (left) and $q = 1.88q_F$ (right), for which the impact of the pair potential is, however, less clear. First, both wavenumbers are in the vicinity of the roton-type feature in the dispersion of the dynamic structure factor, and we observe a substantial exchange-correlation induced redshift of spectral weight compared to the RPA default model for both Ewald and YR. For $q = 1.25q_F$, the PIMC results for the ITCF are very similar. Interestingly, we still observe quantitative differences in $S(\mathbf{q}, \omega)$, which highlights the high sensitivity of the analytic continuation to any change in $F(\mathbf{q}, \tau)$. For $q = 1.88q_F$, $q = 2.51q_F$, and $q = 3.13q_F$, the effect of the pair potential is smaller and we can hardly resolve any significant differences within the given error bars.

D. Quadratic density response

While linear response theory is ubiquitous in WDM theory [124] and constitutes the basis for most diagnostics [147–149], there has emerged a recent interest in the nonlinear response properties of WDM [96,122,146,150–154] (and beyond [155,156]). Such effects are important, e.g., for stopping-power calculations [157–160] and for the construction of effective forces and potentials [138,161]. Reliable results for the nonlinear density response of the warm dense UEG have been presented in Ref. [122] based on exact PIMC simulations of the harmonically perturbed system. Subsequently, Dornheim *et al.* [96] have generalized Eq. (5) to the nonlinear density response by relating the latter to higher-order ITCFs. For example, the static quadratic density response function $\chi^{(2)}(\mathbf{q}, 0)$ that describes the density response at the second harmonic of the original perturbation is given by

$$\chi^{(2)}(\mathbf{q}, 0) = \frac{\Omega^2}{2} \int_0^\beta d\tau_1 \int_0^\beta d\tau_2 F^{(2)}(\mathbf{q}, \tau_1, \tau_2), \quad (8)$$

with the definition of the diagonal three-body ITCF,

$$F^{(2)}(\mathbf{q}, \tau_1, \tau_2) = \langle \hat{n}(2\mathbf{q}, 0) \hat{n}(-\mathbf{q}, \tau_1) \hat{n}(-\mathbf{q}, \tau_2) \rangle. \quad (9)$$

In Fig. 14, we analyze the effect of the pair potential onto PIMC results for $\chi^{(2)}(\mathbf{q}, 0)$. The solid red and dotted black curves have been obtained using the analytical framework introduced in Ref. [146], which approximates screening effects on a linear level, using as input the ESA local-field correction and random-phase approximation (i.e., no

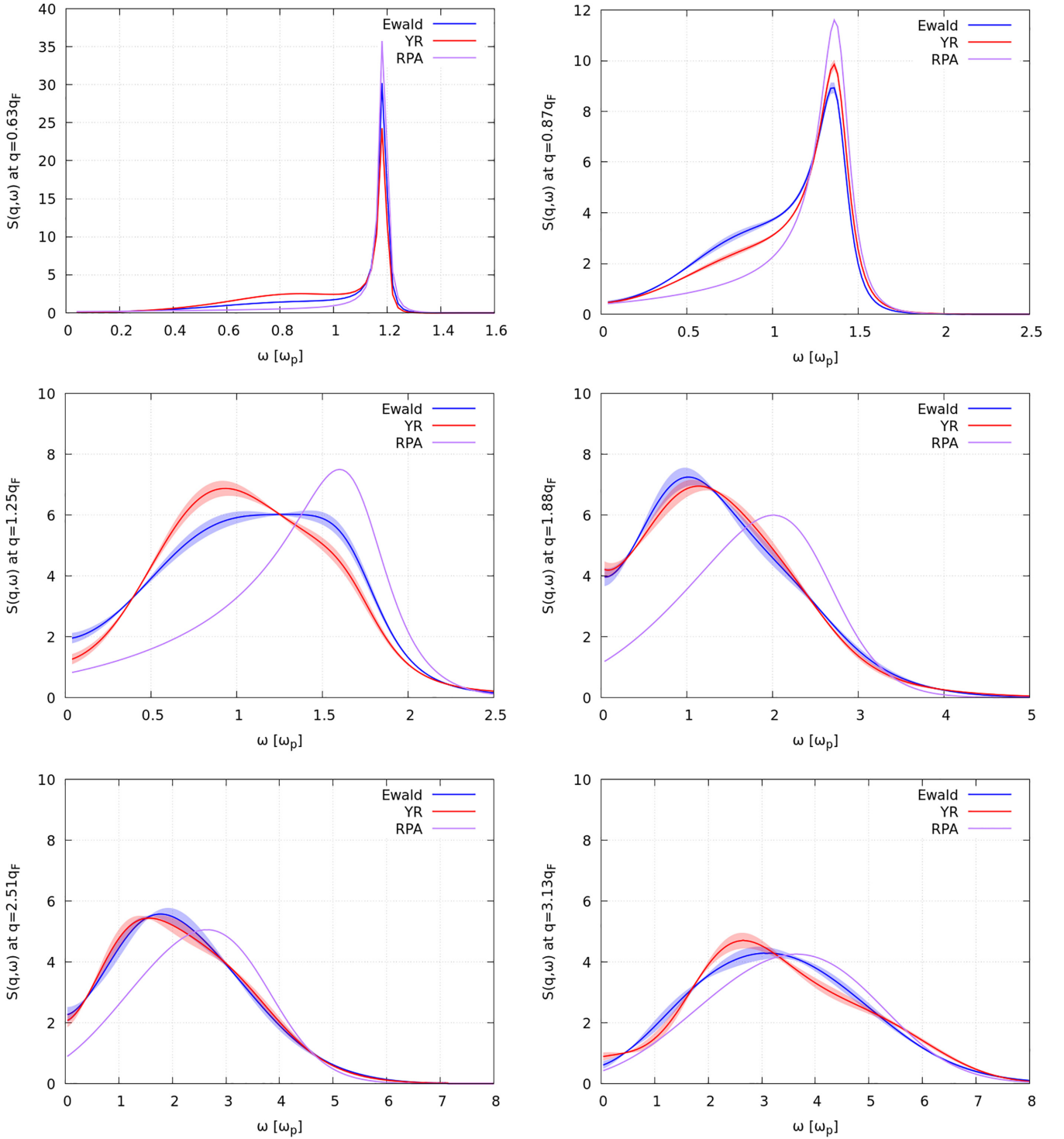


FIG. 12. MEM estimates of $S(\mathbf{q}, \omega)$, where q values are indicated in the y label. The MEM was conducted on ITCF data with either Ewald interaction (red) or YR interaction (blue). The RPA (purple) was used as the Bayesian prior in either case. At small q , there was statistically significant disagreement between the $S(\mathbf{q}, \omega)$ obtained from Ewald and YR ITCF data.

local-field correction), respectively. Empirically, this framework works well for $q \gtrsim 2q_F$, but underestimates the true magnitude of screening effects for $q \rightarrow 0$; in the context of the present work, its main purpose is as a guide to the eye. The black stars and yellow triangles have been obtained using the Ewald pair potential and no dependence on the system size can be resolved within the given error bars. In

contrast, the YR results (green crosses and blue diamonds) deviate from the Ewald results, and from each other for small to intermediate q . First, we note that the particular dependence of these oscillations in $\chi^{(2)}(\mathbf{q}, 0)$ is less trivial compared to $S(\mathbf{q})$ and $\chi(\mathbf{q}, 0)$ investigated above, as it involves correlations between triples of particles instead of pairs; see Eq. (9). Second, we find significant inaccuracies in the vicinity of the

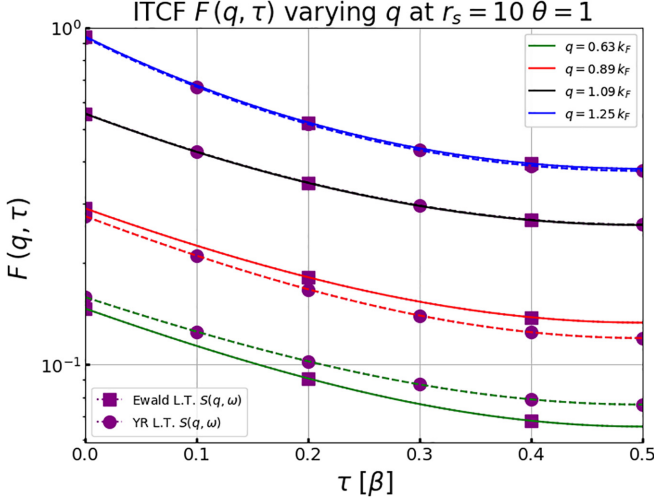


FIG. 13. The ITCF data and the associated Laplace-transformed MEM $S(\mathbf{q}, \omega)$ estimate. The solid and dashed lines indicate ITCFs that have been computed using the Ewald and YR pair potentials, respectively. The MEM estimates associated with each ITCF (squares and circles) are indistinguishable from the respective data. For any MEM estimate, the χ^2 value is of order 10^{-6} or less.

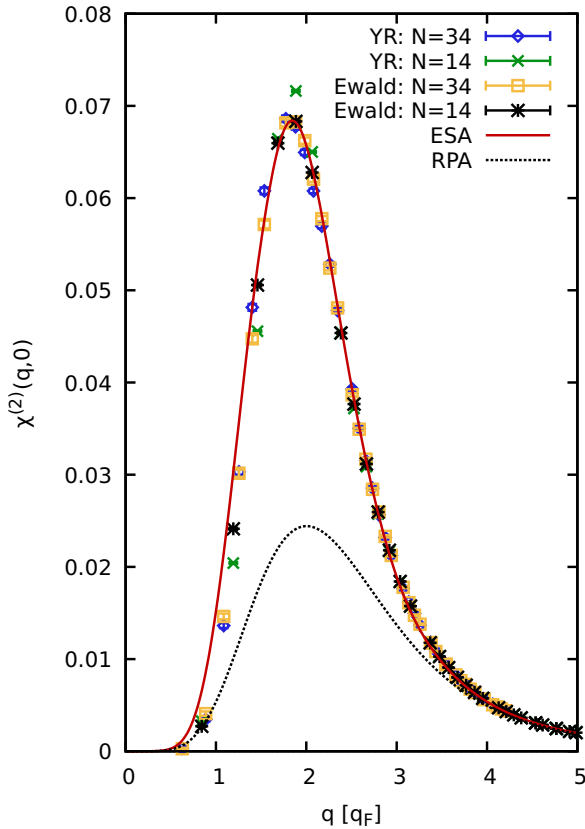


FIG. 14. Static quadratic density response function [at the second harmonic of the original perturbation] $\chi^{(2)}(\mathbf{q}, 0)$ at $r_s = 10$ and $\Theta = 1$. Solid red (dotted black): Analytical expressions from Ref. [146] using as input the ESA local-field correction [111,112] (within the random-phase approximation). The symbols distinguish the Ewald and YR pair potentials, as well as $N = 14$ and $N = 34$ electrons.

peak in particular for $N = 14$, which further substantiates our earlier recommendation for thorough N comparisons in YR PIMC calculations.

IV. SUMMARY AND OUTLOOK

In this work, we have investigated in detail the effect of using the spherically averaged periodic potential by Yakub and Ronchi [83,84] onto *ab initio* PIMC simulations of the warm dense UEG. Overall, the YR potential reduces the computational effort by 1 to 2 orders of magnitude, which is particularly important for emerging capabilities to perform PIMC simulations with large numbers of electrons [67,68].

For integrated properties such as the kinetic- and potential-energy contributions, we find that the YR potential gives results in very close agreement with the more expensive Ewald potentials even at relatively strong coupling, $r_s = 10$. This agrees with previous investigations [70,86,91] and makes the YR potential the method of choice for such applications.

The situation is, however, more complex for q -resolved properties such as the static structure factor $S(\mathbf{q})$ and the static linear density response function $\chi(\mathbf{q})$. In particular, the YR potential effectively penalizes certain pair distances due to a double counting procedure, which introduces fluctuations in q -resolved properties in the vicinity of the cutoff wavenumber $q_{\text{cut}} = 2\pi/r_{\text{cut}}$, where $r_{\text{cut}} \approx 0.6L$ is of the same order as the length of the simulation cell. In practice, this means that the YR potential does not allow us to reliably study the small- q limit, which is important for the description of XRTS experiments in a forward scattering geometry [79–81,147], the evaluation of compressibility sum rules [3,39,162], and the estimation of optical and transport properties [163].

In addition, we have applied the recent maximum entropy method setup by Chuna *et al.* [98,145] to PIMC results for the ITCF $F(\mathbf{q}, \tau)$ with both Ewald and YR potentials. The thus obtained dynamic structure factors $S(\mathbf{q}, \omega)$ exhibit significant deviations at small q , which further emphasizes the limitations of YR in this regime. On the other hand, the deviations vanish for $q \gtrsim 2q_F$, which might facilitate future YR calculations to describe and interpret XRTS measurements at large scattering angles that effectively probe the noncollective or even the single-particle regime [94,116].

Finally, we have analyzed the effect of the pair potential on the static quadratic density response at the second harmonic. This involves the PIMC estimation of an imaginary-time triple-correlation function, leading to a less obvious effect of the YR potential. Most notably, we find a larger effect of the latter onto $\chi^{(2)}(\mathbf{q}, 0)$ in the vicinity of its maximum compared to the static linear response function $\chi(\mathbf{q})$.

Future efforts might include the application of the YR potential to PIMC simulations of real WDM systems that consist of both electrons and ions [51,91–94,164–166], combined with a dedicated treatment of the diverging Coulomb attraction at small distances [167–169]. Moreover, the YR potential can be easily combined with other optimization schemes such as path contraction [170] or an improved Metropolis treatment of long-range potentials [109].

ACKNOWLEDGMENTS

This work was partially supported by the Center for Advanced Systems Understanding (CASUS), financed by Germany's Federal Ministry of Education and Research and the Saxon state government out of the State budget approved by the Saxon State Parliament. This work has received funding from the European Union's Just Transition Fund (JTF) within the project *Röntgenlaser-Optimierung der Laserfusion* (ROLF), Contract No. 5086999001, cofinanced by the Saxon state government out of the State budget approved by the Saxon State Parliament. This work has received funding from the European Research Council (ERC) under the European Union's Horizon 2022 research and innovation programme (Grant Agreement No. 101076233, "PREXTREME"). Computations were performed on a

Bull Cluster at the Center for Information Services and High-Performance Computing (ZIH) at Technische Universität Dresden and at the Norddeutscher Verbund für Hoch- und Höchstleistungsrechnen (HLRN) under Grant No. mvp00024.

Views and opinions expressed are, however, those of the authors only and do not necessarily reflect those of the European Union or the European Research Council Executive Agency.

DATA AVAILABILITY

The data that support the findings of this article are openly available [110].

-
- [1] *Frontiers and Challenges in Warm Dense Matter*, edited by F. Graziani, M. P. Desjarlais, R. Redmer, and S. B. Trickey (Springer International Publishing, Switzerland, 2014).
 - [2] T. Ott, H. Thomsen, J. W. Abraham, T. Dornheim, and M. Bonitz, Recent progress in the theory and simulation of strongly correlated plasmas: Phase transitions, transport, quantum, and magnetic field effects, *Eur. Phys. J. D* **72**, 84 (2018).
 - [3] G. Giuliani and G. Vignale, *Quantum Theory of the Electron Liquid* (Cambridge University Press, Cambridge, 2008).
 - [4] A. Benuzzi-Mounaix, S. Mazevet, A. Ravasio, T. Vinci, A. Denoeud, M. Koenig, N. Amadou, E. Brambrink, F. Festa, A. Levy, M. Harmand, S. Brygoo, G. Huser, V. Recoules, J. Bouchet, G. Morard, F. Guyot, T. de Resseguier, K. Myanishi, N. Ozaki *et al.*, Progress in warm dense matter study with applications to planetology, *Phys. Scr.* **T161**, 014060 (2014).
 - [5] R.P. Drake, *High-Energy-Density Physics: Foundation of Inertial Fusion and Experimental Astrophysics*, Graduate Texts in Physics (Springer International Publishing, Berlin, 2018).
 - [6] A. L. Kritcher, D. C. Swift, T. Döppner, B. Bachmann, L. X. Benedict, G. W. Collins, J. L. DuBois, F. Elsner, G. Fontaine, J. A. Gaffney, S. Hamel, A. Lazicki, W. R. Johnson, N. Kostinski, D. Kraus, M. J. MacDonald, B. Maddox, M. E. Martin, P. Neumayer, A. Nikroo *et al.*, A measurement of the equation of state of carbon envelopes of white dwarfs, *Nature (London)* **584**, 51 (2020).
 - [7] D. Saumon, S. Blouin, and P.-E. Tremblay, Current challenges in the physics of white dwarf stars, *Phys. Rep.* **988**, 1 (2022).
 - [8] R. Betti and O. A. Hurricane, Inertial-confinement fusion with lasers, *Nat. Phys.* **12**, 435 (2016).
 - [9] O. A. Hurricane, P. K. Patel, R. Betti, D. H. Froula, S. P. Regan, S. A. Slutz, M. R. Gomez, and M. A. Sweeney, Physics principles of inertial confinement fusion and U.S. program overview, *Rev. Mod. Phys.* **95**, 025005 (2023).
 - [10] H. Abu-Shawareb, R. Acree, P. Adams, J. Adams, B. Addis, R. Aden, P. Adrian, B. B. Afeyan, M. Aggleton, L. Aghaian, A. Aguirre, D. Aikens, J. Akre, F. Albert *et al.* (The Indirect Drive ICF Collaboration), Achievement of target gain larger than unity in an inertial fusion experiment, *Phys. Rev. Lett.* **132**, 065102 (2024).
 - [11] A. B. Zylstra, O. A. Hurricane, D. A. Callahan, A. L. Kritcher, J. E. Ralph, H. F. Robey, J. S. Ross, C. V. Young, K. L. Baker, D. T. Casey, T. Döppner, L. Divol, M. Hohenberger, S. Le Pape, A. Pak, P. K. Patel, R. Tommasini, S. J. Ali, P. A. Amendt, L. J. Atherton *et al.*, Burning plasma achieved in inertial fusion, *Nature (London)* **601**, 542 (2022).
 - [12] V. Gopalaswamy, C. A. Williams, R. Betti, D. Patel, J. P. Knauer, A. Lees, D. Cao, E. M. Campbell, P. Farmakis, R. Ejaz, K. S. Anderson, R. Epstein, J. Carroll-Nellenbeck, I. V. Igumenshchev, J. A. Marozas, P. B. Radha, A. A. Solodov, C. A. Thomas, K. M. Woo, T. J. B. Collins *et al.*, Demonstration of a hydrodynamically equivalent burning plasma in direct-drive inertial confinement fusion, *Nat. Phys.* **20**, 751 (2024).
 - [13] D. Batani, A. Colaitis, F. Consoli, C. N. Danson, L. A. Gizzi, J. Honrubia, T. Köhl, S. Le Pape, J.-L. Miquel, J. M. Perlado, *et al.*, Future for inertial-fusion energy in europe: A roadmap, *High Power Laser Sci. Eng.* **11**, e83 (2023).
 - [14] S. X. Hu, B. Militzer, V. N. Goncharov, and S. Skupsky, First-principles equation-of-state table of deuterium for inertial confinement fusion applications, *Phys. Rev. B* **84**, 224109 (2011).
 - [15] M. Bonitz, J. Vorberger, M. Bethkenhagen, M. P. Böhme, D. M. Ceperley, A. Filinov, T. Gawne, F. Graziani, G. Gregori, P. Hamann, S. B. Hansen, M. Holzmann, S. X. Hu, H. Kählert, V. V. Karasiev, U. Kleinschmidt, L. Kordts, C. Makait, B. Militzer, Z. A. Moldabekov *et al.*, Toward first principles-based simulations of dense hydrogen, *Phys. Plasmas* **31**, 110501 (2024).
 - [16] T. Schoof, S. Groth, J. Vorberger, and M. Bonitz, *Ab initio* thermodynamic results for the degenerate electron gas at finite temperature, *Phys. Rev. Lett.* **115**, 130402 (2015).
 - [17] N.-H. Kwong and M. Bonitz, Real-time Kadanoff-Baym approach to plasma oscillations in a correlated electron gas, *Phys. Rev. Lett.* **84**, 1768 (2000).
 - [18] V. Golubnychiy, M. Bonitz, D. Kremp, and M. Schlanges, Plasmon dispersion of a weakly degenerate nonideal one-component plasma, *Contrib. Plasma Phys.* **42**, 37 (2002).
 - [19] V. V. Karasiev, L. Calderin, and S. B. Trickey, Importance of finite-temperature exchange correlation for warm dense matter calculations, *Phys. Rev. E* **93**, 063207 (2016).
 - [20] K. Ramakrishna, T. Dornheim, and J. Vorberger, Influence of finite temperature exchange-correlation effects in hydrogen, *Phys. Rev. B* **101**, 195129 (2020).

- [21] T. Sjöström and J. Daligault, Gradient corrections to the exchange-correlation free energy, *Phys. Rev. B* **90**, 155109 (2014).
- [22] N. D. Mermin, Thermal properties of the inhomogeneous electron gas, *Phys. Rev.* **137**, A1441 (1965).
- [23] L. Goerigk, A. Hansen, C. Bauer, S. Ehrlich, A. Najibi, and S. Grimme, A look at the density functional theory zoo with the advanced GMTKN55 database for general main group thermochemistry, kinetics and noncovalent interactions, *Phys. Chem. Chem. Phys.* **19**, 32184 (2017).
- [24] V. V. Karasiev, T. Sjöström, J. Dufty, and S. B. Trickey, Accurate homogeneous electron gas exchange-correlation free energy for local spin-density calculations, *Phys. Rev. Lett.* **112**, 076403 (2014).
- [25] S. Groth, T. Dornheim, T. Sjöström, F. D. Malone, W. M. C. Foulkes, and M. Bonitz, *Ab initio* exchange–correlation free energy of the uniform electron gas at warm dense matter conditions, *Phys. Rev. Lett.* **119**, 135001 (2017).
- [26] V. V. Karasiev, J. W. Dufty, and S. B. Trickey, Nonempirical semilocal free-energy density functional for matter under extreme conditions, *Phys. Rev. Lett.* **120**, 076401 (2018).
- [27] V. V. Karasiev, D. I. Mihaylov, and S. X. Hu, Meta-GGA exchange-correlation free energy density functional to increase the accuracy of warm dense matter simulations, *Phys. Rev. B* **105**, L081109 (2022).
- [28] Z. Moldabekov, S. Schwalbe, M. P. Böhme, J. Vorberger, X. Shao, M. Pavanello, F. R. Graziani, and T. Dornheim, Bound-state breaking and the importance of thermal exchange–correlation effects in warm dense hydrogen, *J. Chem. Theory Comput.* **20**, 68 (2024).
- [29] Z. Moldabekov, J. Vorberger, and T. Dornheim, From density response to energy functionals and back: An *ab initio* perspective on matter under extreme conditions, *Prog. Part. Nucl. Phys.* **140**, 104144 (2025).
- [30] Z. A. Moldabekov, M. Lokamani, J. Vorberger, A. Cangi, and T. Dornheim, Non-empirical mixing coefficient for hybrid xc functionals from analysis of the XC kernel, *J. Phys. Chem. Lett.* **14**, 1326 (2023).
- [31] Z. A. Moldabekov, M. Lokamani, J. Vorberger, A. Cangi, and T. Dornheim, Assessing the accuracy of hybrid exchange-correlation functionals for the density response of warm dense electrons, *J. Chem. Phys.* **158**, 094105 (2023).
- [32] Z. Moldabekov, M. Böhme, J. Vorberger, D. Blaschke, and T. Dornheim, *Ab initio* static exchange–correlation kernel across Jacob’s ladder without functional derivatives, *J. Chem. Theory Comput.* **19**, 1286 (2023).
- [33] M. F. Kasim, T. P. Galligan, J. Topp-Mugglestone, G. Gregori, and S. M. Vinko, Inverse problem instabilities in large-scale modeling of matter in extreme conditions, *Phys. Plasmas* **26**, 112706 (2019).
- [34] G. Gregori, S. H. Glenzer, W. Rozmus, R. W. Lee, and O. L. Landen, Theoretical model of x-ray scattering as a dense matter probe, *Phys. Rev. E* **67**, 026412 (2003).
- [35] T. Dornheim, H. M. Bellenbaum, M. Bethkenhagen, S. B. Hansen, M. P. Böhme, T. Döppner, L. B. Fletcher, T. Gawne, D. O. Gericke, S. Hamel, D. Kraus, M. J. MacDonald, Zh. A. Moldabekov, T. R. Preston, R. Redmer, M. Schörner, S. Schwalbe, P. Talias, and J. Vorberger, Model-free Rayleigh weight from x-ray Thomson scattering measurements, *Phys. Plasmas* **32**, 052712 (2025).
- [36] T. Dornheim, M. Böhme, D. Kraus, T. Döppner, T. R. Preston, Z. A. Moldabekov, and J. Vorberger, Accurate temperature diagnostics for matter under extreme conditions, *Nat. Commun.* **13**, 7911 (2022).
- [37] T. Döppner, M. Bethkenhagen, D. Kraus, P. Neumayer, D. A. Chapman, B. Bachmann, R. A. Baggott, M. P. Böhme, L. Divol, R. W. Falcone, L. B. Fletcher, O. L. Landen, M. J. MacDonald, A. M. Saunders, M. Schörner, P. A. Sterne, J. Vorberger, B. B. L. Witte, A. Yi, R. Redmer *et al.*, Observing the onset of pressure-driven k-shell delocalization, *Nature (London)* **618**, 270 (2023).
- [38] M. P. Böhme, L. B. Fletcher, T. Döppner, D. Kraus, A. D. Baczewski, T. R. Preston, M. J. MacDonald, F. R. Graziani, Z. A. Moldabekov, J. Vorberger, and T. Dornheim, Evidence of free-bound transitions in warm dense matter and their impact on equation-of-state measurements, [arXiv:2306.17653](https://arxiv.org/abs/2306.17653).
- [39] M. W. C. Dharma-wardana and D. D. Klug, X-ray Thomson scattering studies on spin-singlet stabilization of highly compressed h-like be ions heated to two million degrees kelvin, [arXiv:2502.16693](https://arxiv.org/abs/2502.16693).
- [40] J. B. Anderson, *Quantum Monte Carlo: Origins, Development, Applications* (Oxford University Press, New York, 2007).
- [41] D. M. Ceperley, Path integrals in the theory of condensed helium, *Rev. Mod. Phys.* **67**, 279 (1995).
- [42] M. Troyer and U. J. Wiese, Computational complexity and fundamental limitations to fermionic quantum Monte Carlo simulations, *Phys. Rev. Lett.* **94**, 170201 (2005).
- [43] E. Y. Loh, J. E. Gubernatis, R. T. Scalettar, S. R. White, D. J. Scalapino, and R. L. Sugar, Sign problem in the numerical simulation of many-electron systems, *Phys. Rev. B* **41**, 9301 (1990).
- [44] T. Dornheim, Fermion sign problem in path integral Monte Carlo simulations: Quantum dots, ultracold atoms, and warm dense matter, *Phys. Rev. E* **100**, 023307 (2019).
- [45] D. M. Ceperley, Path-integral calculations of normal liquid ^3He , *Phys. Rev. Lett.* **69**, 331 (1992).
- [46] T. Dornheim, Z. A. Moldabekov, J. Vorberger, and B. Militzer, Path integral Monte Carlo approach to the structural properties and collective excitations of liquid ^3He without fixed nodes, *Sci. Rep.* **12**, 708 (2022).
- [47] T. Morresi and G. Garberoglio, Normal liquid ^3He studied by path-integral Monte Carlo with a parametrized partition function, *Phys. Rev. B* **111**, 014521 (2025).
- [48] R. Egger, W. Häusler, C. H. Mak, and H. Grabert, Crossover from fermi liquid to wigner molecule behavior in quantum dots, *Phys. Rev. Lett.* **82**, 3320 (1999).
- [49] S. M. Reimann and M. Manninen, Electronic structure of quantum dots, *Rev. Mod. Phys.* **74**, 1283 (2002).
- [50] T. Dornheim and Y. Yan, Abnormal quantum moment of inertia and structural properties of electrons in 2d and 3d quantum dots: An *ab initio* path-integral Monte Carlo study, *New J. Phys.* **24**, 113024 (2022).
- [51] K. P. Driver and B. Militzer, All-electron path integral Monte Carlo simulations of warm dense matter: Application to water and carbon plasmas, *Phys. Rev. Lett.* **108**, 115502 (2012).
- [52] E. W. Brown, B. K. Clark, J. L. DuBois, and D. M. Ceperley, Path-integral Monte Carlo simulation of the warm dense homogeneous electron gas, *Phys. Rev. Lett.* **110**, 146405 (2013).

- [53] N. S. Blunt, T. W. Rogers, J. S. Spencer, and W. M. C. Foulkes, Density-matrix quantum Monte Carlo method, *Phys. Rev. B* **89**, 245124 (2014).
- [54] F. D. Malone, N. S. Blunt, J. J. Shepherd, D. K. K. Lee, J. S. Spencer, and W. M. C. Foulkes, Interaction picture density matrix quantum Monte Carlo, *J. Chem. Phys.* **143**, 044116 (2015).
- [55] T. Dornheim, S. Groth, A. Filinov, and M. Bonitz, Permutation blocking path integral Monte Carlo: A highly efficient approach to the simulation of strongly degenerate non-ideal fermions, *New J. Phys.* **17**, 073017 (2015).
- [56] S. Groth, T. Schoof, T. Dornheim, and M. Bonitz, *Ab initio* quantum Monte Carlo simulations of the uniform electron gas without fixed nodes, *Phys. Rev. B* **93**, 085102 (2016).
- [57] F. D. Malone, N. S. Blunt, E. W. Brown, D. K. K. Lee, J. S. Spencer, W. M. C. Foulkes, and J. J. Shepherd, Accurate exchange-correlation energies for the warm dense electron gas, *Phys. Rev. Lett.* **117**, 115701 (2016).
- [58] A. Yilmaz, K. Hunger, T. Dornheim, S. Groth, and M. Bonitz, Restricted configuration path integral Monte Carlo, *J. Chem. Phys.* **153**, 124114 (2020).
- [59] B. Hirshberg, M. Invernizzi, and M. Parrinello, Path integral molecular dynamics for fermions: Alleviating the sign problem with the Bogoliubov inequality, *J. Chem. Phys.* **152**, 171102 (2020).
- [60] T. Dornheim, M. Invernizzi, J. Vorberger, and B. Hirshberg, Attenuating the fermion sign problem in path integral Monte Carlo simulations using the Bogoliubov inequality and thermodynamic integration, *J. Chem. Phys.* **153**, 234104 (2020).
- [61] J. Lee, M. A. Morales, and F. D. Malone, A phaseless auxiliary-field quantum Monte Carlo perspective on the uniform electron gas at finite temperatures: Issues, observations, and benchmark study, *J. Chem. Phys.* **154**, 064109 (2021).
- [62] P.-C. Hou, B.-Z. Wang, K. Haule, Y. Deng, and K. Chen, Exchange-correlation effect in the charge response of a warm dense electron gas, *Phys. Rev. B* **106**, L081126 (2022).
- [63] Y. Xiong and H. Xiong, On the thermodynamic properties of fictitious identical particles and the application to fermion sign problem, *J. Chem. Phys.* **157**, 094112 (2022).
- [64] T. Dornheim, P. Tolias, S. Groth, Z. A. Moldabekov, J. Vorberger, and B. Hirshberg, Fermionic physics from *ab initio* path integral Monte Carlo simulations of fictitious identical particles, *J. Chem. Phys.* **159**, 164113 (2023).
- [65] T. Dornheim, Z. A. Moldabekov, S. Schwalbe, and J. Vorberger, Direct free energy calculation from *ab initio* path integral Monte Carlo simulations of warm dense matter, *Phys. Rev. B* **111**, L041114 (2025).
- [66] Y. Xiong, GPU acceleration of *ab initio* simulations of large-scale identical particles based on path integral molecular dynamics, [arXiv:2404.02628](https://arxiv.org/abs/2404.02628).
- [67] T. Dornheim, Z. Moldabekov, S. Schwalbe, P. Tolias, and J. Vorberger, Fermionic free energies from *ab initio* path integral monte carlo simulations of fictitious identical particles, *J. Chem. Theory Comput.* **21**, 7290 (2025).
- [68] T. Dornheim, S. Schwalbe, Z. A. Moldabekov, J. Vorberger, and P. Tolias, *Ab initio* path integral Monte Carlo simulations of the uniform electron gas on large length scales, *J. Phys. Chem. Lett.* **15**, 1305 (2024).
- [69] L. M. Fraser, W. M. C. Foulkes, G. Rajagopal, R. J. Needs, S. D. Kenny, and A. J. Williamson, Finite-size effects and Coulomb interactions in quantum Monte Carlo calculations for homogeneous systems with periodic boundary conditions, *Phys. Rev. B* **53**, 1814 (1996).
- [70] G. S. Demyanov, A. S. Onegin, and P. R. Levashov, N-convergence in one-component plasma: Comparison of Coulomb, Ewald, and angular-averaged Ewald potentials, *Contrib. Plasma Phys.* **64**, e202300164 (2024).
- [71] T. Dornheim, M. Bonitz, Z. A. Moldabekov, S. Schwalbe, P. Tolias, and J. Vorberger, Chemical potential of the warm dense electron gas from *ab initio* path integral Monte Carlo simulations, *Phys. Rev. B* **111**, 115149 (2025).
- [72] T. Dornheim, S. Groth, and M. Bonitz, The uniform electron gas at warm dense matter conditions, *Phys. Rep.* **744**, 1 (2018).
- [73] S. Chiesa, D. M. Ceperley, R. M. Martin, and M. Holzmann, Finite-size error in many-body simulations with long-range interactions, *Phys. Rev. Lett.* **97**, 076404 (2006).
- [74] N. D. Drummond, R. J. Needs, A. Sorouri, and W. M. C. Foulkes, Finite-size errors in continuum quantum Monte Carlo calculations, *Phys. Rev. B* **78**, 125106 (2008).
- [75] T. Dornheim, S. Groth, T. Sjostrom, F. D. Malone, W. M. C. Foulkes, and M. Bonitz, *Ab initio* quantum Monte Carlo simulation of the warm dense electron gas in the thermodynamic limit, *Phys. Rev. Lett.* **117**, 156403 (2016).
- [76] T. Dornheim and J. Vorberger, Overcoming finite-size effects in electronic structure simulations at extreme conditions, *J. Chem. Phys.* **154**, 144103 (2021).
- [77] M. Holzmann, R. C. Clay, M. A. Morales, N. M. Tubman, D. M. Ceperley, and C. Pierleoni, Theory of finite size effects for electronic quantum Monte Carlo calculations of liquids and solids, *Phys. Rev. B* **94**, 035126 (2016).
- [78] S. H. Glenzer, O. L. Landen, P. Neumayer, R. W. Lee, K. Widmann, S. W. Pollaine, R. J. Wallace, G. Gregori, A. Höll, T. Bornath, R. Thiele, V. Schwarz, W.-D. Kraeft, and R. Redmer, Observations of plasmons in warm dense matter, *Phys. Rev. Lett.* **98**, 065002 (2007).
- [79] T. Gawne, Z. A. Moldabekov, O. S. Humphries, K. Appel, C. Baecht, V. Bouffetier, E. Brambrink, A. Cangi, S. Göde, Z. Konôpková, M. Makita, M. Mishchenko, M. Nakatsutsumi, K. Ramakrishna, L. Randolph, S. Schwalbe, J. Vorberger, L. Wollenweber, U. Zastrau, T. Dornheim *et al.*, Ultrahigh resolution x-ray Thomson scattering measurements at the European x-ray free electron laser, *Phys. Rev. B* **109**, L241112 (2024).
- [80] P. Sperling, E. J. Gamboa, H. J. Lee, H. K. Chung, E. Galtier, Y. Omarbakiyeva, H. Reinholz, G. Röpke, U. Zastrau, J. Hastings, L. B. Fletcher, and S. H. Glenzer, Free-electron x-ray laser measurements of collisional-damped plasmons in isochorically heated warm dense matter, *Phys. Rev. Lett.* **115**, 115001 (2015).
- [81] H. M. Bellenbaum, B. Bachmann, D. Kraus, Th. Gawne, M. P. Böhme, T. Döppner, L. B. Fletcher, M. J. MacDonald, Zh. A. Moldabekov, T. R. Preston, J. Vorberger, and T. Dornheim, Toward model-free temperature diagnostics of warm dense matter from multiple scattering angles, *Appl. Phys. Lett.* **126**, 044104 (2025).
- [82] P. Hamann, J. Vorberger, T. Dornheim, Z. A. Moldabekov, and M. Bonitz, *Ab initio* results for the plasmon dispersion and damping of the warm dense electron gas, *Contrib. Plasma Phys.* **60**, e202000147 (2020).
- [83] E. Yakub and C. Ronchi, An efficient method for computation of long-ranged Coulomb forces in computer

- simulation of ionic fluids, *J. Chem. Phys.* **119**, 11556 (2003).
- [84] E. Yakub and C. Ronchi, A new method for computation of long ranged Coulomb forces in computer simulation of disordered systems, *J. Low Temp. Phys.* **139**, 633 (2005).
- [85] G. Vernizzi, G. I. Guerrero-García, and M. Olvera de la Cruz, Coulomb interactions in charged fluids, *Phys. Rev. E* **84**, 016707 (2011).
- [86] G. S. Demyanov and P. R. Levashov, Systematic derivation of angular-averaged Ewald potential, *J. Phys. A: Math. Theor.* **55**, 385202 (2022).
- [87] I. Fukuda and H. Nakamura, Non-Ewald methods: theory and applications to molecular systems, *Biophys. Rev.* **4**, 161 (2012).
- [88] V. S. Filinov, V. E. Fortov, M. Bonitz, and Zh. Moldabekov, Fermionic path-integral Monte Carlo results for the uniform electron gas at finite temperature, *Phys. Rev. E* **91**, 033108 (2015).
- [89] V. S. Filinov, A. S. Larkin, and P. R. Levashov, Uniform electron gas at finite temperature by fermionic-path-integral Monte Carlo simulations, *Phys. Rev. E* **102**, 033203 (2020).
- [90] V. Filinov, P. Levashov, and A. Larkin, Monte Carlo simulations of the electron short-range quantum ordering in Coulomb systems and the ‘fermionic sign problem’, *J. Phys. A: Math. Theor.* **55**, 035001 (2022).
- [91] A. V. Filinov and M. Bonitz, Equation of state of partially ionized hydrogen and deuterium plasma revisited, *Phys. Rev. E* **108**, 055212 (2023).
- [92] T. Dornheim, S. Schwalbe, M. P. Böhme, Z. A. Moldabekov, J. Vorberger, and P. Tolias, *Ab initio* path integral Monte Carlo simulations of warm dense two-component systems without fixed nodes: Structural properties, *J. Chem. Phys.* **160**, 164111 (2024).
- [93] T. Dornheim, S. Schwalbe, P. Tolias, M. P. Böhme, Z. A. Moldabekov, and J. Vorberger, *Ab initio* density response and local field factor of warm dense hydrogen, *Matter Radiat. Extrem.* **9**, 057401 (2024).
- [94] T. Dornheim, T. Döppner, P. Tolias, M. P. Böhme, L. B. Fletcher, T. Gawne, F. R. Graziani, D. Kraus, M. J. MacDonald, Z. A. Moldabekov, S. Schwalbe, D. O. Gericke, and J. Vorberger, Unraveling electronic correlations in warm dense quantum plasmas, *Nat. Commun.* **16**, 5103 (2025).
- [95] T. Dornheim, J. Vorberger, S. Groth, N. Hoffmann, Zh. A. Moldabekov, and M. Bonitz, The static local field correction of the warm dense electron gas: An *ab initio* path integral Monte Carlo study and machine learning representation, *J. Chem. Phys.* **151**, 194104 (2019).
- [96] T. Dornheim, Z. A. Moldabekov, and J. Vorberger, Nonlinear density response from imaginary-time correlation functions: *Ab initio* path integral Monte Carlo simulations of the warm dense electron gas, *J. Chem. Phys.* **155**, 054110 (2021).
- [97] T. Dornheim, S. Groth, J. Vorberger, and M. Bonitz, *Ab initio* path integral Monte Carlo results for the dynamic structure factor of correlated electrons: From the electron liquid to warm dense matter, *Phys. Rev. Lett.* **121**, 255001 (2018).
- [98] T. Chuna, N. Barnfield, T. Dornheim, M. P. Friedlander, and T. Hoheisel, Dual formulation of the maximum entropy method applied to analytic continuation of quantum Monte Carlo data, [arXiv:2501.01869](https://arxiv.org/abs/2501.01869).
- [99] M. Jarrell and J. E. Gubernatis, Bayesian inference and the analytic continuation of imaginary-time quantum Monte Carlo data, *Phys. Rep.* **269**, 133 (1996).
- [100] D. Chandler and P. G. Wolynes, Exploiting the isomorphism between quantum theory and classical statistical mechanics of polyatomic fluids, *J. Chem. Phys.* **74**, 4078 (1981).
- [101] M. Boninsegni, N. Prokof'ev, and B. Svistunov, Worm algorithm for continuous-space path integral Monte Carlo simulations, *Phys. Rev. Lett.* **96**, 070601 (2006).
- [102] T. Dornheim, S. Groth, A. V. Filinov, and M. Bonitz, Path integral Monte Carlo simulation of degenerate electrons: Permutation-cycle properties, *J. Chem. Phys.* **151**, 014108 (2019).
- [103] M. Boninsegni, N. V. Prokofev, and B. V. Svistunov, Worm algorithm and diagrammatic Monte Carlo: A new approach to continuous-space path integral Monte Carlo simulations, *Phys. Rev. E* **74**, 036701 (2006).
- [104] T. Dornheim, M. Böhme, B. Militzer, and J. Vorberger, *Ab initio* path integral Monte Carlo approach to the momentum distribution of the uniform electron gas at finite temperature without fixed nodes, *Phys. Rev. B* **103**, 205142 (2021).
- [105] T. Dornheim, M. Böhme, and S. Schwalbe, ISHTAR - imaginary-time stochastic high- performance tool for *Ab initio* Research, Zenodo (2024), <https://doi.org/10.5281/zenodo.10497098>.
- [106] A. Y. Toukmaji and J. A. Board, Ewald summation techniques in perspective: A survey, *Comput. Phys. Commun.* **95**, 73 (1996).
- [107] G. Rajagopal and R. J. Needs, An optimized Ewald method for long-ranged potentials, *J. Comput. Phys.* **115**, 399 (1994).
- [108] I. Kylänpää and E. Räsänen, Extended Ewald summation technique, *Comput. Phys. Commun.* **206**, 64 (2016).
- [109] F. Müller, H. Christiansen, S. Schnabel, and W. Janke, Fast, hierarchical, and adaptive algorithm for metropolis Monte Carlo simulations of long-range interacting systems, *Phys. Rev. X* **13**, 031006 (2023).
- [110] T. Dornheim, T. M. Chuna, H. M. Bellenbaum, Z. A. Moldabekov, P. Tolias, and J. Vorberger, Data publication Application of a spherically averaged pair potential in *ab initio* path integral Monte Carlo simulations of the warm dense electron gas [Data set], Rodare (2025), <https://doi.org/10.14278/rodare.3959>.
- [111] T. Dornheim, A. Cangi, K. Ramakrishna, M. Böhme, S. Tanaka, and J. Vorberger, Effective static approximation: A fast and reliable tool for warm-dense matter theory, *Phys. Rev. Lett.* **125**, 235001 (2020).
- [112] T. Dornheim, Z. A. Moldabekov, and P. Tolias, Analytical representation of the local field correction of the uniform electron gas within the effective static approximation, *Phys. Rev. B* **103**, 165102 (2021).
- [113] U. Zastrau, P. Sperling, M. Harmand, A. Becker, T. Bornath, R. Bredow, S. Dzierzhytski, T. Fennel, L. B. Fletcher, E. Förster, S. Göde, G. Gregori, V. Hilbert, D. Hochhaus, B. Holst, T. Laarmann, H. J. Lee, T. Ma, J. P. Mithen, R. Mitzner *et al.*, Resolving ultrafast heating of dense cryogenic hydrogen, *Phys. Rev. Lett.* **112**, 105002 (2014).
- [114] L. B. Fletcher, J. Vorberger, W. Schumaker, C. Ruyer, S. Goede, E. Galtier, U. Zastrau, E. P. Alves, S. D. Baalrud, R. A. Baggott, B. Barbel, Z. Chen, T. Döppner, M.

- Gauthier, E. Granados, J. B. Kim, D. Kraus, H. J. Lee, M. J. MacDonald, R. Mishra *et al.*, Electron-ion temperature relaxation in warm dense hydrogen observed with picosecond resolved x-ray scattering, *Front. Phys.* **10**, 838524 (2022).
- [115] P. Hamann, L. Kordts, A. Filinov, M. Bonitz, T. Dornheim, and J. Vorberger, Prediction of a roton-type feature in warm dense hydrogen, *Phys. Rev. Res.* **5**, 033039 (2023).
- [116] H. M. Bellenbaum, M. P. Böhme, M. Bonitz, T. Döppner, L. B. Fletcher, T. Gawne, D. Kraus, Z. A. Moldabekov, S. Schwalbe, J. Vorberger, and T. Dornheim, Estimating ionization states and continuum lowering from *ab initio* path integral Monte Carlo simulations for warm dense hydrogen, *Phys. Rev. Res.* **7**, 033016 (2025).
- [117] S. Moroni, D. M. Ceperley, and G. Senatore, Static response from quantum Monte Carlo calculations, *Phys. Rev. Lett.* **69**, 1837 (1992).
- [118] S. Moroni, D. M. Ceperley, and G. Senatore, Static response and local field factor of the electron gas, *Phys. Rev. Lett.* **75**, 689 (1995).
- [119] C. Bowen, G. Sugiyama, and B. J. Alder, Static dielectric response of the electron gas, *Phys. Rev. B* **50**, 14838 (1994).
- [120] T. Dornheim, S. Groth, J. Vorberger, and M. Bonitz, Permutation blocking path integral Monte Carlo approach to the static density response of the warm dense electron gas, *Phys. Rev. E* **96**, 023203 (2017).
- [121] S. Groth, T. Dornheim, and M. Bonitz, Configuration path integral Monte Carlo approach to the static density response of the warm dense electron gas, *J. Chem. Phys.* **147**, 164108 (2017).
- [122] T. Dornheim, J. Vorberger, and M. Bonitz, Nonlinear electronic density response in warm dense matter, *Phys. Rev. Lett.* **125**, 085001 (2020).
- [123] Z. Moldabekov, J. Vorberger, and T. Dornheim, Density functional theory perspective on the nonlinear response of correlated electrons across temperature regimes, *J. Chem. Theory Comput.* **18**, 2900 (2022).
- [124] T. Dornheim, Z. A. Moldabekov, K. Ramakrishna, P. Tolias, A. D. Baczewski, D. Kraus, T. R. Preston, D. A. Chapman, M. P. Böhme, T. Döppner, F. Graziani, M. Bonitz, A. Cangi, and J. Vorberger, Electronic density response of warm dense matter, *Phys. Plasmas* **30**, 032705 (2023).
- [125] T. Dornheim, Z. Moldabekov, P. Tolias, M. Böhme, and J. Vorberger, Physical insights from imaginary-time density–density correlation functions, *Matter Radiat. Extremes* **8**, 056601 (2023).
- [126] T. Dornheim, D. C. Wicaksono, J. E. Suarez-Cardona, P. Tolias, M. P. Böhme, Z. A. Moldabekov, M. Hecht, and J. Vorberger, Extraction of the frequency moments of spectral densities from imaginary-time correlation function data, *Phys. Rev. B* **107**, 155148 (2023).
- [127] T. Dornheim, J. Vorberger, Z. A. Moldabekov, and M. Böhme, Analysing the dynamic structure of warm dense matter in the imaginary-time domain: Theoretical models and simulations, *Philos. Trans. R. Soc. A* **381**, 20220217 (2023).
- [128] T. Dornheim, M. P. Böhme, D. A. Chapman, D. Kraus, T. R. Preston, Z. A. Moldabekov, N. Schlünzen, A. Cangi, T. Döppner, and J. Vorberger, Imaginary-time correlation function thermometry: A new, high-accuracy and model-free temperature analysis technique for x-ray Thomson scattering data, *Phys. Plasmas* **30**, 042707 (2023).
- [129] M. Schörner, M. Bethkenhagen, T. Döppner, D. Kraus, L. B. Fletcher, S. H. Glenzer, and R. Redmer, X-ray Thomson scattering spectra from density functional theory molecular dynamics simulations based on a modified Chihara formula, *Phys. Rev. E* **107**, 065207 (2023).
- [130] T. Dornheim, T. Döppner, A. D. Baczewski, P. Tolias, M. P. Böhme, Zh. A. Moldabekov, Th. Gawne, D. Ranjan, D. A. Chapman, M. J. MacDonald, Th. R. Preston, D. Kraus, and J. Vorberger, X-ray Thomson scattering absolute intensity from the f-sum rule in the imaginary-time domain, *Sci. Rep.* **14**, 14377 (2024).
- [131] B. Militzer, E. L. Pollock, and D. M. Ceperley, Path integral Monte Carlo calculation of the momentum distribution of the homogeneous electron gas at finite temperature, *High Energy Density Phys.* **30**, 13 (2019).
- [132] B. Militzer and E. L. Pollock, Lowering of the kinetic energy in interacting quantum systems, *Phys. Rev. Lett.* **89**, 280401 (2002).
- [133] K. Hunger, T. Schoof, T. Dornheim, M. Bonitz, and A. Filinov, Momentum distribution function and short-range correlations of the warm dense electron gas: *Ab initio* quantum Monte Carlo results, *Phys. Rev. E* **103**, 053204 (2021).
- [134] T. Dornheim, J. Vorberger, B. Militzer, and Z. A. Moldabekov, Momentum distribution of the uniform electron gas at finite temperature: Effects of spin polarization, *Phys. Rev. E* **104**, 055206 (2021).
- [135] W. Janke and T. Sauer, Optimal energy estimation in path-integral Monte Carlo simulations, *J. Chem. Phys.* **107**, 5821 (1997).
- [136] T. Dornheim, T. Sjöström, S. Tanaka, and J. Vorberger, Strongly coupled electron liquid: *Ab initio* path integral Monte Carlo simulations and dielectric theories, *Phys. Rev. B* **101**, 045129 (2020).
- [137] T. Dornheim, Z. Moldabekov, J. Vorberger, H. Kählert, and M. Bonitz, Electronic pair alignment and roton feature in the warm dense electron gas, *Commun. Phys.* **5**, 304 (2022).
- [138] T. Dornheim, P. Tolias, Z. A. Moldabekov, A. Cangi, and J. Vorberger, Effective electronic forces and potentials from *ab initio* path integral Monte Carlo simulations, *J. Chem. Phys.* **156**, 244113 (2022).
- [139] J. Koskela, L. Reining, and M. Gatti, Short-range excitonic phenomena in low-density metals, *Phys. Rev. Lett.* **134**, 046402 (2025).
- [140] Y. Takada, Emergence of an excitonic collective mode in the dilute electron gas, *Phys. Rev. B* **94**, 245106 (2016).
- [141] Y. Takada, Low-energy peak in the one-particle spectral function of the electron gas at metallic densities, *Phys. Rev. B* **110**, 085132 (2024).
- [142] S. Groth, T. Dornheim, and J. Vorberger, *Ab initio* path integral Monte Carlo approach to the static and dynamic density response of the uniform electron gas, *Phys. Rev. B* **99**, 235122 (2019).
- [143] T. Dornheim and J. Vorberger, Finite-size effects in the reconstruction of dynamic properties from *ab initio* path integral Monte Carlo simulations, *Phys. Rev. E* **102**, 063301 (2020).

- [144] B. Dabrowski, Dynamical local-field factor in the response function of an electron gas, *Phys. Rev. B* **34**, 4989 (1986).
- [145] T. Chuna, N. Barnfield, J. Vorberger, M. P. Friedlander, T. Hoheisel, and T. Dornheim, Estimates of the dynamic structure factor for the finite temperature electron liquid via analytic continuation of path integral Monte Carlo data, [arXiv:2503.20433](https://arxiv.org/abs/2503.20433).
- [146] T. Dornheim, M. Böhme, Z. A. Moldabekov, J. Vorberger, and M. Bonitz, Density response of the warm dense electron gas beyond linear response theory: Excitation of harmonics, *Phys. Rev. Res.* **3**, 033231 (2021).
- [147] S. H. Glenzer and R. Redmer, X-ray Thomson scattering in high energy density plasmas, *Rev. Mod. Phys.* **81**, 1625 (2009).
- [148] J. Sheffield, D. Froula, S.H. Glenzer, and N.C. Luhmann, *Plasma Scattering of Electromagnetic Radiation: Theory and Measurement Techniques* (Elsevier Science, Burlington, USA, 2010).
- [149] W. Nolting and W. D. Brewer, *Fundamentals of Many-body Physics: Principles and Methods* (Springer, Heidelberg, 2009).
- [150] T. Dornheim, J. Vorberger, and Z. A. Moldabekov, Nonlinear density response and higher order correlation functions in warm dense matter, *J. Phys. Soc. Jpn.* **90**, 104002 (2021).
- [151] T. Dornheim, Z. A. Moldabekov, and J. Vorberger, Nonlinear electronic density response of the ferromagnetic uniform electron gas at warm dense matter conditions, *Contrib. Plasma Phys.* **61**, e202100098 (2021).
- [152] T. Dornheim, J. Vorberger, Z. A. Moldabekov, and M. Bonitz, Nonlinear interaction of external perturbations in warm dense matter, *Contrib. Plasma Phys.* **62**, e202100247 (2022).
- [153] P. Tolias, T. Dornheim, Z. A. Moldabekov, and J. Vorberger, Unravelling the nonlinear ideal density response of many-body systems, *Europhys. Lett.* **142**, 44001 (2023).
- [154] J. Vorberger, T. Dornheim, M. P. Böhme, Z. A. Moldabekov, and P. Tolias, Green's function perspective on the nonlinear density response of quantum many-body systems, *J. Stat. Phys.* **192**, 75 (2025).
- [155] S.A. Mikhailov, Second-order response of a uniform three-dimensional electron gas to a longitudinal electric field, *Annalen der Physik* **524**, 182 (2012).
- [156] S. A. Mikhailov, Nonlinear electromagnetic response of a uniform electron gas, *Phys. Rev. Lett.* **113**, 027405 (2014).
- [157] C. D. Hu and E. Zaremba, Z^3 correction to the stopping power of ions in an electron gas, *Phys. Rev. B* **37**, 9268 (1988).
- [158] J. A. Frenje, R. Florido, R. Mancini, T. Nagayama, P. E. Grabowski, H. Rinderknecht, H. Sio, A. Zylstra, M. Gatu Johnson, C. K. Li, F. H. Séguin, R. D. Petrasso, V. Yu Glebov, and S. P. Regan, Experimental validation of low- z ion-stopping formalisms around the Bragg peak in high-energy-density plasmas, *Phys. Rev. Lett.* **122**, 015002 (2019).
- [159] A. Bergara, I. Campillo, J. M. Pitarke, and P. M. Echenique, Quadratic induced polarization by an external heavy charge in an electron gas, *Phys. Rev. B* **56**, 15654 (1997).
- [160] A. Kononov and A. Schleife, Pre-equilibrium stopping and charge capture in proton-irradiated aluminum sheets, *Phys. Rev. B* **102**, 165401 (2020).
- [161] S. Gravel and N. W. Ashcroft, Nonlinear response theories and effective pair potentials, *Phys. Rev. B* **76**, 144103 (2007).
- [162] K.-U. Plagemann, H. R. Rüter, T. Bornath, M. Shihab, M. P. Desjarlais, C. Fortmann, S. H. Glenzer, and R. Redmer, *Ab initio* calculation of the ion feature in x-ray Thomson scattering, *Phys. Rev. E* **92**, 013103 (2015).
- [163] P. E. Grabowski, S. B. Hansen, M. S. Murillo, L. G. Stanton, F. R. Graziani, A. B. Zylstra, S. D. Baalrud, P. Arnault, A. D. Baczewski, L. X. Benedict, C. Blancard, O. Čertík, J. Cléroutin, L. A. Collins, S. Copeland, A. A. Correa, J. Dai, J. Daligault, M. P. Desjarlais, M. W. C. Dharma-wardana *et al.*, Review of the first charged-particle transport coefficient comparison workshop, *High Energy Density Phys.* **37**, 100905 (2020).
- [164] B. Militzer, F. González-Cataldo, S. Zhang, K. P. Driver, and F. Soubiran, First-principles equation of state database for warm dense matter computation, *Phys. Rev. E* **103**, 013203 (2021).
- [165] K. P. Driver, F. Soubiran, and B. Militzer, Path integral Monte Carlo simulations of warm dense aluminum, *Phys. Rev. E* **97**, 063207 (2018).
- [166] M. Böhme, Z. A. Moldabekov, J. Vorberger, and T. Dornheim, Static electronic density response of warm dense hydrogen: *Ab initio* path integral Monte Carlo simulations, *Phys. Rev. Lett.* **129**, 066402 (2022).
- [167] B. Militzer, Computation of the high temperature Coulomb density matrix in periodic boundary conditions, *Comput. Phys. Commun.* **204**, 88 (2016).
- [168] M. Böhme, Z. A. Moldabekov, J. Vorberger, and T. Dornheim, *Ab initio* path integral Monte Carlo simulations of hydrogen snapshots at warm dense matter conditions, *Phys. Rev. E* **107**, 015206 (2023).
- [169] G. S. Demyanov and P. R. Levashov, Kelbglib: Program implementation of the high-temperature kelbg density matrix for path integral and molecular dynamics simulations with long-range Coulomb interaction, *Comput. Phys. Commun.* **305**, 109326 (2024).
- [170] C. John, T. Spura, S. Habershon, and T. D. Kühne, Quantum ring-polymer contraction method: Including nuclear quantum effects at no additional computational cost in comparison to *ab initio* molecular dynamics, *Phys. Rev. E* **93**, 043305 (2016).