Correlation energies of the high-density spin-polarized electron gas to meV accuracy

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We present a novel combination of quantum Monte Carlo methods and a finite-size extrapolation framework with which we calculate the thermodynamic limit of the correlation energy of the polarized electron gas at high densities to meV accuracy. By independently extrapolating the fixed node diffusion Monte Carlo correlation energy and the fixed node error with respect to full configuration-interaction quantum Monte Carlo, we find correlation energies of $-40.44(5)$ and $-31.70(4)$ mHa at $r_s = 0.5$ and 1, respectively, improving the precision of existing parametrizations by an order of magnitude.

The uniform (or homogeneous) electron gas (UEG) [1] is a system consisting of electrons in a neutralizing uniform background which models the behavior of electrons in metals [2]. This system is of crucial importance in understanding the nature of electronic correlation, and is of huge practical relevance since knowledge of the correlation energy of the UEG as a function of its homogeneous density can be used as a key ingredient in the description of the behavior of electrons in real systems [3–5]. The release-node Green’s function quantum Monte Carlo (RN-GFMC) calculations of Ceperley and Alder [6] provided data connecting the analytic high-density [7, 8] and low-density [9] limits of the correlation energy, and enabled the development of parametrizations over the entire density range [10–12] which are routinely used in density functional theory calculations.

Despite its seeming simplicity, the complex correlations caused by the long-ranged character of the Coulomb interaction require the use of explicit many-body methods to accurately characterize the UEG. The diffusion quantum Monte Carlo (DMC) method [13, 14] has been extensively used to study the UEG [15–18], but this method suffers from a sign problem when used to study Fermi systems [6, 19], requiring a fixed node approximation which introduces a bias. Full configuration-interaction quantum Monte Carlo (FCIQMC) is a stochastic projection technique that explicitly operates in a basis of antisymmetric functions, thus avoiding the need for a fixed node approximation [20, 21]. FCIQMC has been successfully used to study several systems of interest in quantum chemistry and condensed matter physics [22–27], including the unpolarized UEG [28–30].

The Perdew-Wang parametrization of the correlation energy of the electron gas [12] has become a cornerstone in Hartree atomic units ($\hbar = m_e = |e| = 4 \pi \epsilon_0 = 1$),

$$\hat{H} = \frac{1}{2} \sum_i \nabla_i^2 + \sum_{i<j} \frac{1}{|\mathbf{r}_i - \mathbf{r}_j|},$$

where $\mathbf{r}_i$ is the position vector of the $i$th electron, and the system is characterized by its uniform number density $n$, usually specified via $r_s = (4 \pi n/3)^{-1/3}$. The second-quantized Hamiltonian of the infinite UEG is

$$\hat{H} = \frac{1}{2} \sum_k k^2 a_k^\dagger a_k + \sum_{p,q} \sum_{k \neq 0} \frac{4 \pi}{k^2} q^\dagger p q^\dagger q a_p^\dagger a_q^\dagger a_k a_p,$$

where $k$, $p$, and $q$ are reciprocal-space vectors, and $a_k^\dagger$ and $a_k$ are the creation and annihilation operators for the single-electron state of wave vector $k$, respectively. The Fermi wave vector, $k_F = (6 \pi^2 n)^{1/3}$ at full spin polarization, characterizes the system. The kinetic energy term is diagonal, and the interaction term only connects states with equal total momentum $k_T$. The Hilbert space of the system thus consists of disjoint subspaces corresponding to different $k_T$, and the ground state

![Figure 1](image-url)
is the solution of the Schrödinger equation in the subspace for which the total energy is minimized.

We simulate a finite version of this system consisting of \( N \) electrons in a cubic simulation cell of side \( L = (n/N)^{1/3} \) subject to periodic boundary conditions. This requires replacing the Coulomb interaction in Eq. 1 with an Ewald summation [35], restricting the summations in Eq. 2 to reciprocal lattice vectors, \( \mathbf{G} = 2\pi (i_x, i_y, i_z) \), where \( i_x, i_y, \) and \( i_z \) are integers, and adding a self-interaction constant to both Hamiltonians. Throughout this Letter we discuss and report energies per electron.

In the high density regime the UEG behaves as a Fermi liquid, for which a plane-wave basis is a natural choice. The configuration interaction (CI) expansion of the ground-state wave function is \( \Psi_0 = \sum \{ C_I \} D_I \), where \( \{ C_I \} \) are the CI coefficients, \( D_I = \det(e^{i \mathbf{G} \cdot \mathbf{r}_i}) \) are determinants of plane-wave orbitals, and \( \mu_{IJ} \) is the index of the \( I \)th wave vector occupied in the \( J \)th determinant. We label the Hartree-Fock (HF) determinant, which corresponds to the choice of \( I \) that minimizes \( \langle D_I | H | D_I \rangle \), as \( I = 1 \).

The VMC method [13, 36] requires a trial wave function \( \Psi_T \) to evaluate \( \langle \Psi_T | H | \Psi_T \rangle \) by direct Monte Carlo integration in real space, and provides a framework for optimizing wave function parameters [37, 38]. In the DMC method [13, 19] the wave function is represented by a set of real-space walkers which evolves according to a small time-step approximation [39] to the Green’s function associated with the imaginary-time Schrödinger equation. The fixed node approximation prevents this process from collapsing onto the bosonic ground state by requiring the DMC wave function to have the same nodes as \( \Psi_T \). The positive bias in the energy incurred by the fixed node approximation is referred to as the fixed node error, \( \varepsilon_{FN} \).

The Slater-Jastrow form is a common choice of trial wave function for electronic systems, and consists of the HF determinant multiplied by a Jastrow correlation factor [40, 41]. Backflow transformations [15–17, 42, 43] offer the ability to modify the nodes of the Slater-Jastrow wave function and give significantly lower DMC energies. Further details can be found in the Supplemental Material [44].

The FCIQMC method [20, 21, 26, 27] obtains the CI coefficients by evolution of a population of random walkers, each associated with a determinant in Hilbert space, in imaginary time via diagonal death/cloning and off-diagonal spawning processes. An annihilation step is carried out at each time step to cancel walkers of opposite signs on the same determinant, which is crucial for sign coherence [20]. The initial set of walkers in an FCIQMC calculation is usually placed on the HF determinant, and after an equilibration stage the occupation of each determinant is on average proportional to its exact CI coefficient.

The initiator approximation modifies the dynamics of the random walk to allow a substantial reduction in the number of walkers \( W \) required for convergence, but is a source of bias [21, 26, 27]. The initiator error vanishes as \( W \to \infty \), and in practice we increase the walker population until energy changes become negligible. The number of walkers required to overcome the initiator error increases with the size of the Hilbert space of the system, which grows very quickly with system size, and has also been observed to increase with \( \rho_s \) [28, 29].

Basis sets for the UEG consist of the \( M \) plane waves with the smallest wave vectors. This finite basis set provides access to a finite portion of the Hilbert space of the system, resulting in a positive energy bias. The infinite basis set limit can be estimated by extrapolation, as is standard practice in quantum chemistry [45]. The basis-set size dependence of the correlation energy of the polarized UEG is well-described by a quadratic polynomial in \( \rho_s^{-1} \) [44], in contrast with the linear dependence found for the unpolarized UEG [28].

We assess the quality of our FCIQMC energies by comparison with VMC and DMC energies for increasingly accurate trial wave functions. We construct multi-determinantal wave functions for the 19-electron gas at \( \rho_s = 1 \) by truncating the FCIQMC wave function to the \( \rho_s \) leading determinants, with symmetry-equivalent determinants grouped together. Determinant coefficients are re-optimized in VMC together with other wave function parameters. The results, obtained using the CASINO code [46], are plotted in Fig. 2 against \( \rho_s \).

The variational convergence of our VMC and DMC energies towards the FCIQMC energy is consistent with FCIQMC being exact for this system. The best backflow DMC energy is only 0.027(5) mHa higher than the FCIQMC energy, and is to our knowledge the most accurate DMC energy for this system reported to date.

For our main calculations we use twist averaging in the canonical ensemble to reduce finite-size errors [31]. The translational invariance of the wave function of a periodic system is defined up to a phase factor, \( \Psi(r_1, ..., r_i + \mathbf{R}, ..., r_N) = e^{i \theta} \Psi(r_1, ..., r_i, ..., r_N) \), where \( \mathbf{R} \) is a simulation cell lattice vector, \( \mathbf{R} = L (i_x, i_y, i_z) \), and \( i_x, i_y, \) and \( i_z \) are integers. This phase factor can be obtained by shifting the reciprocal lattice by a certain \( \mathbf{k}_R \) in the Brillouin zone such that \( \theta = \mathbf{k}_R \cdot \mathbf{R} \).

We note that the total momentum \( \mathbf{k}_T = \sum_i \mathbf{G}_{\mu_i} \) of the

![Figure 2. VMC and DMC energies of the polarized 19-electron gas at \( \rho_s = 1 \) (at \( \Gamma \)) relative to FCIQMC, as a function of the number of determinants in the wave function, both without and with backflow transformations.](image-url)
ground-state wave function changes discretely with \( k_\pi \), dividing the Brillouin zone into \( Z \) regions associated with different \( k_\pi \). Since it is not trivial to determine \textit{a priori} which \( k_\pi \) yields the lowest energy at a given \( k_s \), \( k_\pi \) is usually chosen so as to minimize the energy of the non-interacting system, resulting in convex polyhedral regions bounded by Bragg planes [31, 44].

Averaging expectation values over \( k_s \) in the Brillouin zone is referred to as twist averaging, and has the effect of reducing quasirandom fluctuations of expectation values with system size [31]. The integration over \( k_s \) is usually performed stochastically or using a grid in the Brillouin zone [31, 33]. However, inspection of the second-quantized Hamiltonian of Eq. 2 reveals that, for a fixed \( k_\pi \), shifting the reciprocal lattice by \( k_s \) adds a constant to the diagonal kinetic energy term and leaves the interaction term unchanged, since it only depends on differences between reciprocal lattice vectors. This suggests a twist-averaging scheme which is more efficient than other approaches at small system sizes [47]. By expressing the total energy as the HF energy plus the correlation energy, \( E_{\text{tot}}(k_s) = E_{\text{HF}}(k_s) + E_{\text{corr}}(k_s) \), the HF energy absorbs the continuous variation of the kinetic energy with \( k_s \), while the correlation energy is constant within each region. Evaluating the average correlation energy weighted by the region volumes, which can be obtained exactly for \( N \lesssim 100 \) [44], yields the twist-averaged correlation energy. We use this scheme to twist-average our FCIQMC energies, and we use random sampling to twist-average our DMC energies. The basis-set dependence of the FCIQMC correlation energies is found to depend weakly on the region, and in selected cases we perform the FCIQMC basis-set extrapolation in a single region, for efficiency [44]. In what follows we discuss and report twist-averaged energies only.

The thermodynamic limit of the energy of the UEG is usually obtained by extrapolation of \( E_{\text{tot}}(N) - \Delta K(N) \), where \( \Delta K(N) = K(N) - K(\infty) \) is the finite-size error in the HF kinetic energy [6, 33]. However in our calculations we find that the correlation energy exhibits significantly smaller quasirandom fluctuations. Chiesa et al. [32] showed that the leading-order contribution to the finite size error in the DMC energy of an electronic system is \( t_3 N^{-1} \), where \( t_3 = -\sqrt{2} \tau_{\lambda}^{-3/2} \) for the polarized UEG. Drummond et al. [33] found that the leading-order contribution to the finite size error in the HF energy of an electron gas is \( h_2 N^{-2/3} \), where \( h_2 = -\frac{577}{600} r_s^{-1} \) for the polarized UEG and \( \epsilon_1 = 5.6745946 \) for simple cubic simulation cells [33, 44]. Since beyond-leading-order contributions to the HF energy are proportional to \( N^{-4/3} \) [31, 33], the DMC correlation energy satisfies

\[
E_{\text{corr}}^{\text{FN}}(N) = c_0 - h_2 N^{-2/3} + t_3 N^{-1} + c_4 N^{-4/3} + c_5 N^{-5/3} + c_6 N^{-2} + \ldots ,
\]

where \( \{ c_n \} \) are density-dependent parameters.

We evaluate the DMC correlation energy of the polarized UEG using the Slater-Jastrow wave function at system sizes \( 15 \leq N \leq 515 \) at \( r_s = 0.5 \) and \( 15 \leq N \leq 1021 \) at \( r_s = 1 \), and we use Eq. 3 to obtain the thermodynamic limit of the fixed node correlation energy, setting \( h_2 \) and \( t_3 \) to their analytic values and treating \( c_0, c_4, c_5 \), and \( c_6 \) as fit parameters. We do not use backflow or multi-determinants to avoid introducing optimization noise in our DMC energies. The magnitude of quasirandom fluctuations has been observed to decay as \( N^{-1} \) [31], so we use \( N^2 \) as weights in our fits.

In Fig. 3 we plot the DMC correlation energies as a function of \( N^{-1} \). These results numerically confirm the absence of additional contributions to Eq. 3 at order \( N^{-1} \) or slower. The inset of Fig. 3 shows the magnitude of quasirandom fluctuations at \( r_s = 0.5 \), which decay as \( N^{-1} \) as expected. The correlation energy is 4 times less noisy than \( E_{\text{tot}} - \Delta K \).

Figure 3. Fixed node correlation energies of the polarized UEG at \( r_s = 0.5 \) and 1 as a function of \( N^{-1} \). The lines are fits of the data to Eq. 3. The inset shows the absolute fit error at \( r_s = 0.5 \).

We turn our attention to the density dependence of Eq. 3, which we re-express as

\[
E_{\text{corr}}^{\text{FN}}(\xi) = c_0 - h_2 \xi^2 + t_3 \xi^3 + c_4 \xi^4 + c_5 \xi^5 + c_6 \xi^6 + \ldots ,
\]

where \( \xi = r_s^{-1/2} N^{-1/3} \). We find that assuming tilded coefficients to be density-independent, in line with leading-order extrapolation formulas proposed in the literature [49], incurs a negligible error at high densities. In Fig. 4 we plot \( E_{\text{corr}}^{\text{FN}} \) as a function of \( \xi^3 \), and we perform a combined fit of the data at \( r_s = 0.5 \) and 1 to Eq. 4, which we find to fit the data extremely well [44]. We also plot fixed node energies at \( r_s = 5 \) to demonstrate the breakdown of this approximation at low densities.

We compute the exact energy of the system using FCIQMC at system sizes \( N = 15, 19, \) and \( 27 \) at \( r_s = 1 \) and \( N = 15, 19, 27, \) and \( 33 \) at \( r_s = 0.5 \), and we evaluate the fixed node error as the difference between the fixed node and exact correlation energies, which we give in Table I.

Holzmann et al. [34] found that the use of backflow contributes to the finite-size error in the energy of the UEG at order \( N^{-1} \). This has the subtle consequence that the coefficient of \( N^{-1} \) in the finite-size error of the exact energy must
II we plot the fixed node error and the resulting fit curves, to obtain the thermo-
density-independent coefficients. We perform a combined fit
estimate proportionality between these two quantities $[\xi^2 \varepsilon]$. The line represents a combined fit of the data at $r_s = 0.5$ and 1
to Eq. 4, with density-dependent $c_0$ and density-independent $c_4$, $c_5$, and $c_6$ coefficients.

Table I. Fixed node and exact correlation energies and fixed node
error for the polarized UEG at $r_s = 0.5$ and 1 for different system
sizes, in mHa.

| $r_s$ | $N$ | $E_{\text{corr}}^{\text{FN}}$ | $E_{\text{corr}}$ | $\varepsilon_{\text{FN}}$ |
|-------|-----|-----------------|-----------------|-----------------|
| 0.5   | 15  | -12.885(6)      | -13.5203(7)     | 0.638(6)        |
| 19    |     | -14.636(10)     | -15.404(3)      | 0.769(10)       |
| 27    |     | -16.953(9)      | -17.918(7)      | 0.965(12)       |
| 33    |     | -18.455(9)      | -19.516(13)     | 1.061(15)       |
| 1.0   | 15  | -12.144(3)      | -12.6926(4)     | 0.549(3)        |
| 19    |     | -13.665(8)      | -14.313(4)      | 0.648(9)        |
| 27    |     | -15.654(9)      | -16.395(1)      | 0.741(9)        |

Table II. Thermodynamic limit of the fixed node correlation energy,
of the fixed node error, and of the exact correlation energy of the
polarized UEG at $r_s = 0.5$ and 1, in mHa. Values of the Perdew-
Wang fit [12] excluding and including our data are also shown.

| $r_s$ | $E_{\text{corr}}^{\text{FN}}$ | $E_{\text{corr}}$ | $\varepsilon_{\text{FN}}$ |
|-------|-----------------|-----------------|-----------------|
| 0.5   | -38.778(10)     | -30.650(3)     |                 |
| 1.0   |                 |                 | 1.67(5)         |
|       | -40.44(5)       | -31.70(4)      | 1.05(4)         |
| PW fit|                 | -31.8(4)       |                 |
| PW fit incl. present data| -40.38(6) | -31.77(8) | |

We expect the variation of $\varepsilon_{\text{FN}}$ with $N$ to be smoother than
that of the fixed node energy, and thus we model it using a lower-order expression. Under the assumption that, like $E_{\text{corr}}^{\text{FN}}$,
the exact correlation energy is accurately represented at high
densities by a polynomial in $\xi$, we write

$$\varepsilon_{\text{FN}}(\xi) = f_0 + \tilde{f}_3 \xi^3 + \tilde{f}_4 \xi^4 + \ldots,$$

where $f_0$ is a density-dependent parameter and $\tilde{f}_3$ and $\tilde{f}_4$ are
density-independent coefficients. We perform a combined fit
of our data at $r_s = 0.5$ and 1 to Eq. 5 to obtain the thermo-
dynamic limit of the fixed node error at both densities. In
Fig. 5 we plot the fixed node error and the resulting fit curves,
and in the inset we show the same data as a function of $\xi^3$.
The results obtained with this procedure are given in Table II
along with values of the Perdew-Wang parametrization of the
correlation energy [12].

Our study of the polarized UEG in the high density regime
using a combination of FCIQMC and DMC offers a fresh
look at the system and new insight into the methodology for
treating extended electronic systems. Our results greatly
improve the precision of existing parametrizations of the correlation
energy of the polarized UEG. We have characterized the
monotonic increase of the fixed node error with system size,
and we expect that knowledge of its magnitude and asymptotic behavior will be extremely useful in assessing finite-size errors in DMC calculations. Crucially, our present work
demonstrates the great potential of FCIQMC, which, along
with similar methods [50], we expect to become an increasingly common tool for extended systems as continued developments broaden its applicability [30].

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See the Supplemental Material at [URL] for discussion of twist averaging, Brillouin zone division, trial wave functions, and finite size errors, and additional results.

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**TWIST AVERAGING**

Twist averaging is a technique to reduce quasirandom fluctuations in expectation values as a function of system size \(N\) [S1] which amounts to averaging an expectation value \(A\) over reciprocal lattice shifts \(k_s\) in the Brillouin zone of the reciprocal lattice of the simulation cell,

\[
A_{TA} = \frac{1}{\Omega_{BZ}} \int_{BZ} A(k_s) \, dk_s , \tag{S1}
\]

where \(\Omega_{BZ}\) is the volume of the Brillouin zone. The correlation energy only depends on the total momentum \(k_T\), which changes discretely with \(\Omega_{BZ}\) specified by \(k_s\), and therefore the integral reduces to a sum over the \(Z\) regions in which the total momentum is constant,

\[
E_{corr}^{TA} = \frac{1}{\Omega_{BZ}} \int_{BZ} E_{corr}(k_s) \, dk_s = \sum_z \frac{\Omega_z^2}{\Omega_{BZ}} E_{corr}(k_s^z) , \tag{S2}
\]

where \(\Omega_z\) is the volume of the \(z\)th region and \(k_s^z\) is an arbitrary reciprocal lattice shift in the \(z\)th region. Below we provide further details of the division of the Brillouin zone.

**Exact division of the Brillouin zone**

The energy (per electron) of the non-interacting system equals the HF kinetic energy,

\[
E_{NI}(k_s; k_T) = K_1(k_s; k_T)
\]

where \(\mu_{1i}\) are the indices of the reciprocal lattice vectors occupied in the HF determinant. These indices determine \(k_T\) and vice versa. The energy of the non-interacting system at fixed \(k_T\) is a paraboloid centred at \(k_s = -\frac{1}{N}k_T\). Since the total momentum at shift \(k_s\) is that which minimizes \(E_{NI}(k_s; k_T)\), \(k_T\) changes discretely at the intersection of two such paraboloids. If \(k_T^1\) and \(k_T^2\) are the total momenta of two adjacent regions, this intersection is given by

\[
\frac{1}{N} (k_T^2 - k_T^1) \cdot k_s = E_{NI}(0; k_T^2) - E_{NI}(0; k_T^1) , \tag{S4}
\]

which is the equation of a plane. The Brillouin zone regions of constant total momentum are therefore convex polyhedra.

In practice we work in the irreducible Brillouin zone (IBZ), which for a simple cubic simulation cell is the tetrahedron given by \(0 \leq z \leq y \leq x \leq \pi/L\), where \(x\), \(y\), and \(z\) are the Cartesian components of \(k_s\). Consequently the total momentum \(k_T = \frac{2\pi}{L}(i_x, i_y, i_z)\), where \(i_x\), \(i_y\), and \(i_z\) are integers, satisfies \(0 \leq -i_z \leq -i_y \leq -i_x \leq N/2\).

The problem of dividing the IBZ reduces to locating the vertices of the polyhedral regions. Note that Eq. S4 represents a Bragg plane, which can be defined in terms of integers, and the region vertices are the intersections of three or more inter-region and/or IBZ planes, and are therefore proportional to vectors of rational numbers. The use of integer arithmetic enables solving the IBZ division problem exactly for moderate system sizes.

Given a shift \(k_s\), finding the \(N\) reciprocal lattice vectors with the smallest \(|G_j + k_s|\) yields the indices of the occupied orbitals \(\{\mu_{1i}\}\), which determines \(k_T\). However, at points on inter-region planes the set of occupied orbitals is not unique, and multiple total momenta give the same, degenerate kinetic energy. The allowed values of the total momentum at a vertex can be obtained by considering all possible occupations, and the equations of the inter-region planes passing through the vertex are given by Eq. S4 for each pair of allowed total momenta. In turn, each pair of planes intersect at a line corresponding to a polyhedral edge which points to an adjacent vertex.

It is thus possible to find the vertices of all polyhedral regions in the IBZ by successively moving between adjacent vertices along region edges. We illustrate our algorithm using the particularly simple case of the 7-electron gas, which we do not consider in our main results. The IBZ division for this example is shown in Fig. S1, where we have labelled the high-symmetry points \(\Gamma, X, M,\) and \(R\) at the corners of the IBZ and the additional vertices \(\alpha, \beta, \gamma, \delta, \) and \(\epsilon\). We start at \(\Gamma\), where we find that \(k_T = 0\) is the only allowed value of the total momentum. We then perform a line search between \(\Gamma\) and \(M\), corresponding to the intersection between two of the three IBZ planes passing through \(\Gamma\), to find the point furthest from \(\Gamma\) at which any of the allowed values of the total momentum at \(\Gamma\) is also an allowed value. This is done by bisection using floating-point arithmetic, and upon locating vertex \(\beta = \frac{2\pi}{L}(2, 1, 0)\) we revert to using integer arithmetic. Inspecting the degenerate occupations at \(\beta\) yields two possible total momenta, \(k_T = 0\) and \(-\frac{2\pi}{L}(2, 1, 0)\), defining an inter-region plane of normal \((2, 1, 0)\). The intersections between this plane and/or the two IBZ planes passing through \(\beta\) provide search directions to find adjacent vertices \(\alpha, \gamma, \), and \(\delta,\) and this process continues until we exhaust the lists of edges radiating from all vertices.

The resulting vertex locations characterize the regions and, using the divergence theorem, we obtain their volume, center, and contributions to the HF kinetic energy (the HF exchange energy is constant within each region). These integrals can
be carried out accurately using floating-point arithmetic; the use of integers is however crucial for the location of vertices, since for $N \gtrsim 30$ the proximity of some of the vertices can cause incorrect IBZ division under floating-point arithmetic.

In Tables S1, S2, and S3 we give the exact region volumes and centers (the latter truncated to four decimals for conciseness) of the IBZ regions corresponding to $N = 15, 19, 27$, and 33, for which we have run FCIQMC calculations, as well as those for the 7-electron system of Fig. S1. In Fig. S2 we plot the number of regions $Z$ as a function of $N$, showing that $Z \sim N^2$. Therefore the number of evaluations of an expectation value required for twist averaging increases quadratically with system size.

The need to enumerate all possible occupations of partially-filled shells causes a computational bottleneck in our exact division algorithm, which we are able to use in practice for $N \lesssim 100$. Computing twist-averaged DMC correlation energies requires knowledge of the corresponding twist-averaged HF energy components, which we obtain using random sampling for $N \gtrsim 100$. Our twist-averaged HF energies are given in Table S4.

### FITTING METHODOLOGY AND STATISTICAL UNCERTAINTY

In our work we use least-squares fits of energy data to perform extrapolations with respect to basis-set size and system size. We avoid the use of “chi squared” fits in which each datum is weighed by the inverse of its squared standard error, since this practice cannot handle datasets simultaneously containing zero and non-zero uncertainties, as is the case of our HF energies, and underestimates the uncertainty in the fit parameters due to a double-counting effect. Instead we perform our least squares fits without these weights, and we obtain the uncertainty in the fit parameters by a stochastic process in which we replace each energy datum with a random number drawn from a normal distribution centred at the expected value of the energy of variance its standard error. The standard error in each fit parameter is then obtained as the square root of the variance of the values of the parameter in 10,000

| $N$ | $z$ | $\frac{\Omega_z}{\Omega_{BZ}}$ | $\frac{\Omega_z}{\Omega_{BZ}}$ |
|-----|-----|-----------------------------|-----------------------------|
| 7   | 0   | 1/18                        | 0.1458, 0.0833, 0.0417     |
|     | 1   | 1/9                         | 0.2708, 0.1146, 0.0417     |
|     | 2   | 7/18                        | 0.3899, 0.2173, 0.0655     |
|     | 3   | 4/9                         | 0.4167, 0.3333, 0.2083     |
| 15  | 0   | 13/252                      | 0.1513, 0.1036, 0.0499     |
|     | 1   | 1/28                        | 0.2411, 0.0491, 0.0179     |
|     | 2   | 4/21                        | 0.3452, 0.1845, 0.1533     |
|     | 3   | 7/72                        | 0.2798, 0.2485, 0.1488     |
|     | 4   | 5/8                         | 0.4250, 0.2937, 0.1250     |
| 19  | 0   | 1/40                        | 0.1188, 0.0562, 0.0250     |
|     | 1   | 11/360                      | 0.1680, 0.1055, 0.0553     |
|     | 2   | 1/7                         | 0.2606, 0.1728, 0.0558     |
|     | 3   | 83/252                      | 0.3354, 0.1143, 0.0497     |
|     | 4   | 19/105                      | 0.3392, 0.2646, 0.2070     |
|     | 5   | 4/55                        | 0.3856, 0.3391, 0.1614     |
|     | 6   | 205/5544                    | 0.3920, 0.2406, 0.0216     |
|     | 7   | 1/56                        | 0.3705, 0.0491, 0.0179     |
|     | 8   | 146/693                     | 0.4487, 0.2780, 0.1765     |
|     | 9   | 43/504                      | 0.4479, 0.1271, 0.0540     |
|     | 10  | 19/792                      | 0.4098, 0.3794, 0.1298     |
|     | 11  | 17/396                      | 0.4711, 0.3528, 0.1290     |
|     | 12  | 7/72                        | 0.4658, 0.4211, 0.1250     |
Throughout our work we use fitting functions with more bias. While this is not a rigorous approach, we expect our estimated using the $\Omega_{z/\Omega_BZ}$, $\Omega_{z/\Omega_BZ}$, and $\Omega_{z/\Omega_BZ}$.

Table S2. Index $z$, total momentum $k_B$, exact weight $\Omega_{z/\Omega_BZ}$, and center $k_z$ of the IBZ regions for $N = 27$.

Table S3. Index $z$, total momentum $k_B$, exact weight $\Omega_{z/\Omega_BZ}$, and center $k_z$ of the IBZ regions for $N = 33$.

Details of the Variational and Diffusion Quantum Monte Carlo Calculations

All of our VMC and DMC calculations have been performed using the CASINO code [S2]. The Slater-Jastrow trial wave function is of the form $\Psi_T(R) = e^{i\mathcal{H}(R)}\Psi_{HF}(R)$, where...
Table S4. Twist-averaged HF kinetic and exchange energies for the polarized UEG at several system sizes, in Ha. Energies for N ≤ 93 are exact, and energies for N ≥ 123 have been estimated using random sampling. The analytic N → ∞ limit is also shown, for reference.

| N  | r_s^2/K | r_sX       |
|----|---------|------------|
| 7  | 1.77110059 | −0.663751377 |
| 15 | 1.75971498 | −0.630999714 |
| 19 | 1.75843687 | −0.623184756 |
| 27 | 1.75774258 | −0.613700247 |
| 33 | 1.75826227 | −0.608534546 |
| 40 | 1.75615221 | −0.605364222 |
| 57 | 1.75545710 | −0.599501435 |
| 81 | 1.75453662 | −0.595096762 |
| 93 | 1.75476609 | −0.593397250 |
| 123| 1.7545311(2)| −0.59069020(3) |
| 147| 1.7542302(2)| −0.58928659(4) |
| 171| 1.7544501(1)| −0.58801682(4) |
| 179| 1.7544764(2)| −0.58767775(4) |
| 203| 1.7541746(2)| −0.58696564(3) |
| 251| 1.7542188(1)| −0.58565017(4) |
| 305| 1.7541073(2)| −0.58465873(4) |
| 515| 1.7541090(1)| −0.58245938(5) |
| 1021| 1.7540160(2)| −0.58058038(5) |
| 2007| 1.7540110(2)| −0.57937257(5) |
| ∞  | 1.75399969 | −0.577252097 |

\[ e^{J(R)} \] is the Jastrow correlation factor, which we parametrize as [S3, S4]

\[
J(R) = \sum_{i<j} (1 - r_{ij}/L_u)^3 \Theta (r_{ij} - L_u) \sum_{l=0}^{8} \alpha_l r_{ij}^l \\
+ \sum_{i<j} \sum_{s=1}^{8} a_s \sum_{G \in \text{th star}} \cos (G \cdot r_{ij}) ,
\]

(S5)

where \( \Theta \) is the Heaviside step function, \( \{ G \in \text{sth star} \} \) are the reciprocal lattice vectors of the simulation cell in the sth star of symmetry-equivalent vectors, and \( \{ \alpha_l \}, \{ a_s \} \) and \( L_u \) are optimizable parameters.

In our multideterminant benchmark of FCIQMC we replace the HF determinant with a selected-CI expansion extracted from FCIQMC. Backflow transformations replace the argument \( R \) of the Slater determinants with transformed coordinates \( X(R) \) of the form [S5]

\[
x_i = r_i + \sum_{j \neq i} (1 - r_{ij}/L_\eta)^3 \Theta (r_{ij} - L_\eta) \sum_{m=1}^{8} c_m r_{ij}^m r_{ij} ,
\]

(S6)

where \( \{ c_l \} \) and \( L_\eta \) are optimizable parameters.

Each of our DMC energies is obtained by linear extrapolation of the results of a DMC calculation consisting of \( M_1 \) steps with a time step of \( \tau_1 \) and a target walker population of \( P_1 \), and a second DMC calculation consisting of \( M_2 = M_1/2 \) steps with a time step of \( \tau_2 = 4\tau_1 \) and a target walker population of \( P_2 = P_1/4 \). We set \( \tau_1 = 0.01 r_s^2 \), \( P_1 = 2048 \), and adjust \( M_1 \) to obtain the desired statistical accuracy.

For our TA-VMC calculations we have used 6400 random values of \( k_s \), and for our TA-DMC calculations we have used up to 3200 values for the system sizes at which we compute the fixed node error, and 32 values for other system sizes.

**DETAILS OF THE FULL CONFIGURATION INTERACTION QUANTUM MONTE CARLO CALCULATIONS**

At each basis-set size \( M \) and IBZ region \( z \) we run an initiator-FCIQMC calculation using the NECI development code. The initiator approximation constrains the random walk so that spawning new walkers on unpopulated determinants from sites that contain less than \( n_{\text{init}} \) walkers is forbidden, where \( n_{\text{init}} \) is a tunable parameter which we set to 3 in our calculations. We start our FCIQMC calculations by placing 100 walkers on the HF determinant, which we then let grow up to a target population \( W \). We gradually increase \( W \) until energy changes become negligible. Our largest calculations use up to \( W = 1.5 \times 10^8 \) walkers. Figure S3 represents the equilibrated walker population on the leading determinants of the CI wave function for one of the systems reported in our work.

![Figure S3. Walker population \( N_I \) on Slater determinant \( D_I \) as a function of \( I \), sorted by decreasing \( N_I \), for the 19-electron gas at \( r_s = 1 \) and \( k_s = 0 \) using a 341-plane-wave basis and \( 10^7 \) walkers. The first peak corresponds to the HF determinant.](image)

**Basis-set extrapolation**

We extrapolate our FCIQMC correlation energies at each IBZ region \( z \) to the complete basis set limit using the fitting
function

$$E_{\text{corr}}^z(M) = E_{\text{corr}}^z(\infty) + a_z M^{-1} + b_z M^{-2},$$  \tag{S7}$$

where $E_{\text{corr}}^z(\infty)$, $a_z$, and $b_z$ are fit parameters. Setting $a_z = 0$ yields a very good fit to the energy data, but we keep $a_z$ as a fit parameter to account for the parametrization bias.

We find that the basis-set error is roughly independent of $z$, as shown in Fig. S4 for the 19-electron gas at $r_s = 1$. To reduce the cost of our FCIQMC calculations for selected systems ($N = 19$, 27, and 33 at $r_s = 0.5$) we perform the basis set extrapolation at the $\Gamma$-point region, $z = 0$, only. For other regions we evaluate the correlation energy at a single basis set size $M$ ($\simeq 1050$, 830, and 830, respectively) and we obtain $E_{\text{corr}}^z(\infty)$ from Eq. S7 by setting $a_z = a_0$ and $b_z = b_0$.

![Figure S4. Finite basis set error in the FCIQMC correlation energy as a function of inverse basis-set size $M^{-1}$ for the 19-electron gas at $r_s = 1$. The data correspond to calculations at multiple basis-set sizes in each of the 13 IBZ regions.](image)

We investigate the bias incurred by this approximation by comparing the value of $b_0$ with its average $b_{\text{ave}} = \sum_z \left( \Omega_z/\Omega_{\text{BZ}} \right) b_z$ in fits of our data at $r_s = 1$ to Eq. S7 with $a_z = 0$. The largest deviation occurs for the 27-electron gas, for which $|1 - b_0/b_{\text{ave}}| = 0.153$. Therefore a contribution of $0.153 \times b_0 M^{-2}$ is added (in quadrature) to the uncertainty of the twist-averaged energy for the systems for which we use the $\Gamma$-point extrapolation scheme, which we expect to overestimate the corresponding bias. This correction represents an increase in the uncertainty of the twist-averaged correlation energy by up to 75%, but despite this, the $\Gamma$-point extrapolation method provides a net reduction in the computational cost of the FCIQMC calculations.

**FINITE-SYSTEM RESULTS**

In Table S5 we give the twist-averaged DMC correlation energies omitted from in Table I of our manuscript. We plot our full set of DMC and FCIQMC correlation energies in Fig. S5.

![Table S5. Twist-averaged DMC correlation energies of the polarized UEG omitted from Table I of our manuscript, in mHa.](image)

**FINITE SIZE ERRORS AND EXTRAPOLATION**

Evaluation of integration errors at $k = 0$

As shown in Refs. S6 and S7, some of the leading-order finite-size errors in DMC energies can be ascribed to integration errors which are effectively due to the inability to sample $k = 0$ at finite $N$. We define the object

$$\epsilon_n(\alpha) = \Omega^{-1} \left[ (2\pi)^{-3} \int \frac{4\pi}{k^2} k^n e^{-\alpha k^2} \, dk \right] - \frac{1}{\Omega} \sum_{G \neq 0} \frac{4\pi}{G^2} G^n e^{-\alpha G^2},$$  \tag{S8}$$
where $\Omega$ is the simulation cell volume, whose limit $\epsilon_n = \lim_{r_n \to 0} \epsilon_n(\alpha)$ represents the error in the discretization of the reciprocal-space convolution of the interaction potential $4\pi p^2/k^2$ and a power of the wave vector $k^n$.

The finite size error in the HF exchange energy of the polarized UEG due to integrations errors at $k = 0$ (given in Eq. S1 of Ref. S7 for the unpolarized UEG) can be written in terms of $\epsilon_n$ as

$$X(N) = X(\infty) - \frac{3\epsilon_1}{16\pi} r_s^{-1} N^{-2/3} + \frac{\epsilon_3}{(4\pi)^3} \left( \frac{\pi}{6} \right)^{2/3} r_s^{-1} N^{-4/3} + \ldots . \quad (S9)$$

Note that, in the notation of Ref. S7, $\epsilon_1 = 2C_{HF}$. The finite size error in the DMC kinetic energy of the polarized UEG due to integrations errors at $k = 0$ (given in Eq. S6 of Ref. S7) can also be expressed in terms of $\epsilon_n$,

$$T(N) = T(\infty) - \frac{\sqrt{3}}{4} r_s^{-3/2} N^{-1} + \frac{\epsilon_3}{16\pi} r_s^{-2} N^{-4/3} + \ldots . \quad (S10)$$

Note that, in the notation of Ref. S7, $\epsilon_3 = 4C_{3D}$.

By manipulating Eq. S8 we arrive at a computable expression for $\epsilon_n(\alpha)$,

$$\epsilon_n(\alpha) = \Omega \frac{n^{1/2}}{\pi\alpha^{n/2}} \left[ \frac{\Gamma \left( \frac{n+1}{2} \right)}{\Gamma \left( \frac{n+1}{2} \right)} - \frac{4\pi}{\Omega} \sum_{\alpha \neq 0} G^{n-2} e^{-\alpha G^2} \right] , \quad (S11)$$

where $\Gamma$ is the Gamma function. The numerical evaluation of $\epsilon_n$ requires computing $\epsilon_n(\alpha)$ at increasingly small values of $\alpha$ until a convergence criterion is met. As can be gathered from Eq. S11, $\epsilon_n(\alpha)$ at $\alpha \to 0$ is the difference of increasingly large numbers, one of which is itself a sum of many terms which needs to be converged independently. This is numerically delicate, and we find that rounding errors prevent obtaining more than 4–5 decimal places of precision in the value of $\epsilon_n$ with this procedure.

However, inspection of the behavior of $\epsilon_n(\alpha)$ with $\alpha$ reveals an exponential convergence pattern, which can be exploited to produce much more accurate estimates of $\epsilon_n$ at values of $\alpha$ at which rounding errors are not problematic. Using the model $\epsilon_n(\alpha) = \epsilon_n e^{-p_1 \alpha}$ we find a two-point extrapolation formula,

$$\epsilon_n \approx \epsilon_n^2(\alpha) e^{-1}(2\alpha) , \quad (S12)$$

and a higher-order model $\epsilon_n(\alpha) = \epsilon_n e^{-p_1 \alpha - p_2 \alpha^2}$ yields a three-point extrapolation formula,

$$\epsilon_n \approx \epsilon_n^3(\alpha) e^{-2}(2\alpha) e^{1/3}(4\alpha) \epsilon_n, \quad (S13)$$

We plot the values of $\epsilon_3(\alpha)$ and the results from the two- and three-point extrapolation formulae in Fig. S6. This technique significantly accelerates convergence: Eq. S13 gives $\epsilon_3$ to 10-digit precision at a value of $\alpha$ at which $\epsilon_3(\alpha)$ is only accurate to 4 decimal places. We note that we have used 128-bit floating-point arithmetic ("quad" precision) to further enhance numerics. With this approach we obtain the values $\epsilon_1 = 5.67459496$ and $\epsilon_3 = 21.04959844$ for our simple cubic simulation cell (but note that we do not use $\epsilon_3$ in our work).

**Other sources of finite-size errors**

Besides integration errors and quasirandom fluctuations, there is a third source of finite-size errors in twist-averaged energies. As reported in Table I of Ref. S1, the twist-averaged HF kinetic energy exhibits finite-size errors that scale as $N^{-4/3}$ to leading order. These finite size errors arise due to the use of the canonical ensemble, i.e., keeping $N$ fixed as $k_F$ is varied during twist-averaging, and is associated with the mismatch between the Fermi vector at size $N$ and in the thermodynamic limit [S1]. In other words, these are integration errors at $k = k_F$ due to the smearing of the Fermi surface as an artifact of twist-averaging in the canonical ensemble.

We observe in our data that the estimate of the finite-size error at order $N^{-4/3}$ in the HF exchange energy given by Eq. S9 and in the DMC kinetic energy given by Eq. S10 do not completely account for the finite-size error at order $N^{-4/3}$ in either of these energy components or in the DMC correlation energy. We hypothesize that integration errors at $k = k_F$ from the various energy components enter the DMC correlation energy at order $N^{-4/3}$ and would need to be fully accounted for in order to determine the $c_4$ coefficient in Eq. 3 of our manuscript. For this reason we treat all coefficients beyond order $N^{-1}$ as fit parameters in our analysis of the correlation energies.

Our DMC correlation energies are well described by Eq. 3 of our manuscript with $c_6 = 0$, but we keep $c_6$ as a fit parameter to account for the parametrization error.
In our manuscript we find that correlation energies at high densities are accurately described by polynomials in \( \xi = r_s^{-1/2} N^{-1/3} \) with density-independent coefficients. The fit shown in Fig. 4 of our manuscript yields fixed node correlation energies in the thermodynamic limit of \(-38.722(8)\) and \(-30.658(2)\) mHa at \( r_s = 0.5 \) and 1, respectively, which differ by 4.3 and 2.0 standard deviations from the values obtained by independent extrapolation at each density given in Table II of our manuscript.

The thermodynamic limit of the fixed node correlation energy at \( r_s = 5.0 \) obtained in our auxiliary calculations is \(-15.270(4)\) mHa.

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