

Transcorrelated approach to the thermodynamic limit

Hongjun Luo 

Max-Planck-Institute for Solid State Research, Heisenbergstraße 1, 70569 Stuttgart, Germany

Ali Alavi 

*Max-Planck-Institute for Solid State Research, Heisenbergstraße 1, 70569 Stuttgart, Germany
and Department of Chemistry, University of Cambridge, Lensfield Road, Cambridge CB2 1EW, United Kingdom*



(Received 19 January 2026; revised 23 March 2026; accepted 9 April 2026; published 27 April 2026)

For electronic structure calculations of periodic extended systems based on many-body wave functions, we investigate the utility of a similarity transformation of the Hamiltonian in the thermodynamic limit, followed by a treatment of the resulting transcorrelated Hamiltonian within a supercell approximation. Using a correlation factor fulfilling the long-range asymptotic condition of the uniform electron gas, the effective potential of the transcorrelated Hamiltonian decays much faster than the original Coulomb potential. This is found to significantly speed up the convergence of the supercell energies towards the thermodynamic limit.

DOI: [10.1103/jwz7-f23z](https://doi.org/10.1103/jwz7-f23z)

I. INTRODUCTION

In modeling extended systems, electronic structure calculations based on many-body wave functions often employ the periodic supercell method, whereby the infinite system is approximated with a finite system (the supercell), periodically arranged to fill the whole space. In this approximation, a charged particle (electron or nucleus) located in one supercell interacts with all charged particles in all other supercells, while the periodic nature of the arrangement imposes severe constraints on the form of the many-body wave function. Exchange and correlation effects are significantly affected by the artificial periodicity imposed by this construction. Due to the slow decay of the electronic Coulomb potential ($\propto r_{ij}^{-1}$), this leads to a finite-size error (the approximation error) in the energy, which converges only very slowly with respect to the supercell size [1]. Since the cost of many-body wave-function calculations increases very rapidly with the system size, it is often impossible to converge the calculation satisfactorily with respect to the supercell size. It is thus extremely important to try to reduce the finite-size error in the most efficient way. Several methods based on truncation of the Coulomb potential or invoking auxiliary compensating functions have been suggested to remove the long-range singularity of the potential [1–5], in order to speed up the convergence of the exact exchange energy contributions to the thermodynamic

limit. On the other hand, for the correlation contribution, which is generally assumed to be more short ranged, there exists no specific treatment. In this work we explore how to speed up the convergence of the supercell method by applying transcorrelation techniques.

The Transcorrelated (TC) method was originally suggested as a method to deal with the short-range singularity of the Coulomb potential [6,7], which induces electronic cusps in the many-body wave function. The pairwise cusp cannot be efficiently factorized into products of single-particle functions, and this leads to a slow convergence in the conventional configuration interaction (CI) expansion of the many-body wave functions. Instead of a CI expansion, a much more compact product form to factor out the cusp from the many-body wave function is the Jastrow ansatz [8]

$$\Psi(\mathbf{r}_1, \dots, \mathbf{r}_N) = e^{\tau(\mathbf{r}_1, \dots, \mathbf{r}_N)} \Phi(\mathbf{r}_1, \dots, \mathbf{r}_N), \quad (1)$$

where the reference function Φ may still be described by a CI expansion, while τ is a two-body factor (the correlation factor)

$$\tau(\mathbf{r}_1, \dots, \mathbf{r}_N) = \frac{1}{2} \sum_{ij} u(\mathbf{r}_i, \mathbf{r}_j). \quad (2)$$

With a properly chosen correlation factor, the cusp singularity is removed from the reference function, which intensively speeds up the convergence of the CI expansion. Recently, TC methods have been successfully implemented in various quantum chemistry calculations [9–23].

Up to now, most TC calculations have focused only on short- and intermediate-range correlations. For long-range correlation, there have been preliminary TC calculations of uniform electron gases (UEG) based on a single determinant ansatz [24,25], and more recently a perturbation treatment [26] in the thermodynamic limit. However, the TC method has not been used to handle the slow convergence of the finite-size

*Contact author: h.luo@fkf.mpg.de

†Contact author: a.alavi@fkf.mpg.de

error of the supercell approach. In this work, we suggest a transcorrelated method for long-range correlation (ℓ -TC). The basic idea is to first perform a similarity transformation in the thermodynamic limit to remove the leading order term in the long-range Coulomb potential, and only then treat the resulting TC Hamiltonian with the supercell approach. Since the effective electronic TC potential decays much faster than the Coulomb potential, in this way we can expect this supercell approach to have a much faster convergence to the thermodynamic limit. In the next section, we will present the details of the ℓ -TC method, and in Sec. III we test this method on the uniform electron gas, which will be followed by some concluding remarks in Sec. IV.

II. TC METHOD FOR THE LONG-RANGE CORRELATION

By using the Jastrow ansatz (1) in electronic structure calculations, we try to solve the Schrödinger equation

$$\hat{H}e^\tau\Phi = Ee^\tau\Phi, \quad (3)$$

where $\hat{H}$ is the electronic Hamiltonian

$$\hat{H} = \sum_i \left(-\frac{1}{2} \nabla_i^2 + U(\mathbf{r}_i) \right) + \frac{1}{2} \sum_{i \neq j} \frac{1}{r_{ij}}. \quad (4)$$

The exponential form of the factor guarantees size consistency, but makes the ansatz highly nonlinear, such that any variational treatment of Jastrow ansatz leads to extremely high-dimensional integrals. To remove this troublesome exponential factor, Boys and Handy [6,27] suggested to multiply both sides of Eq. (3) with $e^{-\tau}$ to change it to a non-Hermitian eigenvalue problem

$$\tilde{H}_{\text{TC}}\Phi = E\Phi, \quad (5)$$

where the TC Hamiltonian has a finite expansion in powers of τ ,

$$\begin{aligned} \tilde{H}_{\text{TC}} &= e^{-\tau} \hat{H} e^\tau \\ &= \hat{H} + [\hat{H}, \tau] + \frac{1}{2} [[\hat{H}, \tau], \tau]. \end{aligned} \quad (6)$$

This Hamiltonian contains only up to three-body effective interactions. The short-range Coulomb singularity of $\hat{H}$ is canceled by the leading short-range term in $[\hat{H}, \tau]$, and the solution Φ is much smoother than the original wave function Ψ . In fact, using a variational quantum Monte Carlo (VMC) optimized correlation factor τ , we can obtain most of the dynamic correlation with a single determinant reference function Φ . The remaining static correlation can then be treated more efficiently with a configuration expansion of Φ [11,12,14].

As has been mentioned in the previous section, in the study of extended systems, it is also difficult to capture the long-range correlation due to the slow decay of the Coulomb potential. The finite-size error converges very slowly with supercell size. To improve the convergence, we suggest to perform a similarity transformation in the thermodynamic limit with a factor $u_0(\mathbf{r}_{ij})$ which fulfills the asymptotic conditions

$$\frac{du_0}{dr_{ij}} = \frac{1}{2}, \quad \text{when } r_{ij} \rightarrow 0, \quad (7)$$

$$u_0 \sim -\frac{1}{\omega_p r_{ij}}, \quad \text{when } r_{ij} \rightarrow \infty, \quad (8)$$

where $\omega_p = \sqrt{4\pi\rho}$ is the plasma frequency and ρ is the (averaged) electron charge density. This leads to a transcorrelated Hamiltonian in the thermodynamic limit

$$\begin{aligned} \hat{H}_{\text{TC0}} &= \hat{H} - \frac{1}{2} \sum_{ij} \{ \nabla_i^2 u_0(\mathbf{r}_{ij}) + 2[\nabla_i u_0(\mathbf{r}_{ij})] \cdot \nabla_i \\ &\quad + [\nabla_i u_0(\mathbf{r}_{ij})]^2 \} - \frac{1}{2} \sum_{ijk} [\nabla_i u_0(\mathbf{r}_{ij})] \cdot [\nabla_i u_0(\mathbf{r}_{ik})] \end{aligned} \quad (9)$$

In this expression, the short-range singularity of $1/r_{ij}$ in $\hat{H}$ is canceled by the term $\nabla_i^2 u_0(\mathbf{r}_{ij})$ as long as condition (7) is fulfilled. On the other hand, the long-range tail of $1/r_{ij}$ is canceled by the three-body term, which can be expressed in terms of second quantization with a plane-wave basis as

$$-\frac{1}{2\Omega_0^2} \sum_{\mathbf{p}\mathbf{q}\mathbf{k}\mathbf{k}'} \mathbf{k} \cdot \mathbf{k}' \tilde{u}_0(\mathbf{k}) \tilde{u}_0(\mathbf{k}') a_{\mathbf{p}-\mathbf{k}}^\dagger a_{\mathbf{q}+\mathbf{k}}^\dagger a_{\mathbf{s}+\mathbf{k}-\mathbf{k}'}^\dagger a_{\mathbf{s}} a_{\mathbf{q}} a_{\mathbf{p}}, \quad (10)$$

where Ω_0 is the formal volume of the infinite space and $\tilde{u}_0$ is the Fourier transformation of u_0 . For simplicity, here and in the following, we ignore the spin indices. It is found that the leading long-range contribution is given by a special contraction of the three-body term [25,28] by taking only $\mathbf{k}' = \mathbf{k}$ [a random phase approximation (RPA) contraction]

$$-\frac{\rho}{2\Omega_0} \sum_{\mathbf{p}\mathbf{q}\mathbf{k}} \mathbf{k}^2 \tilde{u}_0^2(\mathbf{k}) a_{\mathbf{p}-\mathbf{k}}^\dagger a_{\mathbf{q}+\mathbf{k}}^\dagger a_{\mathbf{q}} a_{\mathbf{p}}. \quad (11)$$

In order to cancel the long-range Coulomb tail with this term, we need asymptotically

$$\rho \mathbf{k}^2 \tilde{u}_0^2(\mathbf{k}) \sim \frac{4\pi}{k^2}, \quad (12)$$

and this leads to the condition (8).

With the leading long-range tail of the Coulomb potential removed, the TC Hamiltonian $\hat{H}_{\text{TC0}}$ can then be treated by the supercell method

$$\begin{aligned} \hat{H}_{\text{TC0-Cell}}^{(N)} &= \hat{H}_{\text{Cell}}^{(N)} - \frac{1}{2} \sum_{ij\mathbf{R}_m} \{ \nabla_i^2 u_0(\mathbf{r}_{ij} + \mathbf{R}_m) \\ &\quad + 2[\nabla_i u_0(\mathbf{r}_{ij} + \mathbf{R}_m)] \cdot \nabla_i + [\nabla_i u_0(\mathbf{r}_{ij} + \mathbf{R}_m)]^2 \} \\ &\quad - \frac{1}{2} \sum_{ijk\mathbf{R}_m\mathbf{R}_n} [\nabla_i u_0(\mathbf{r}_{ij} + \mathbf{R}_m)] \cdot [\nabla_i u_0(\mathbf{r}_{ik} + \mathbf{R}_n)] \\ &\quad + \frac{N}{2} \xi_{\text{Cell}}, \end{aligned} \quad (13)$$

where $\mathbf{R}_m, \mathbf{R}_n$ are the lattice vectors of the supercells and N is the number of particles in the supercell. ξ_{Cell} is a self-energy correction term, the Madelung constant, which will be discussed in detail in the next section. Since u_0 is determined in the thermodynamic limit, it cannot be optimized for a given supercell. In order to benefit further from a correlation factor, i.e., to speed up the basis convergence at each supercell level, a second similarity transformation can be performed on the supercell Hamiltonian,

$$\hat{H}_{\text{TC-Cell}}^{(N)} = e^{-\tau_1} \hat{H}_{\text{TC0-Cell}}^{(N)} e^{\tau_1}, \quad (14)$$

with a periodic factor

$$\tau_1 = \frac{1}{2} \sum_{ij\mathbf{R}_m} u_1(\mathbf{r}_i, \mathbf{r}_j + \mathbf{R}_m) \quad (15)$$

optimized for $\hat{H}_{\text{TC-Cell}}$. The final expression is obtained as

$$\begin{aligned} \hat{H}_{\text{TC-Cell}}^{(N)} = & \hat{H}_{\text{Cell}}^{(N)} - \frac{1}{2} \sum_{ij\mathbf{m}} \{ \nabla_i^2 v(\mathbf{r}_i, \mathbf{r}_j + \mathbf{R}_m) + 2[\nabla_i v(\mathbf{r}_i, \mathbf{r}_j + \mathbf{R}_m)] \cdot \nabla_i \} - \frac{1}{2} \sum_{ijk\mathbf{mn}} [\nabla_i v(\mathbf{r}_i, \mathbf{r}_j + \mathbf{R}_m)] \cdot [\nabla_i v(\mathbf{r}_i, \mathbf{r}_k + \mathbf{R}_n)] \\ & - \frac{1}{2} \sum_{ij\mathbf{m}} [\nabla_i u_0(\mathbf{r}_{ij} + \mathbf{R}_m)]^2 - \frac{1}{2} \sum_{ij\mathbf{mn}} [\nabla_i u_1(\mathbf{r}_i, \mathbf{r}_j + \mathbf{R}_m)] \cdot [\nabla_i u_1(\mathbf{r}_i, \mathbf{r}_j + \mathbf{R}_n)] + \frac{N}{2} \xi_{\text{Cell}}, \end{aligned} \quad (16)$$

where $v(\mathbf{r}, \mathbf{r}') = u_0(\mathbf{r} - \mathbf{r}') + u_1(\mathbf{r}, \mathbf{r}')$. We find that in the above expression, the two factors can be combined into one factor for the first-order terms and the three-body term. Crucially, however, they have different expressions for the second-order two-body term.

III. ℓ -TC CALCULATIONS OF THE UNIFORM ELECTRON GAS

For the UEG, the local factor u_1 should also have the translational symmetry $u_1(\mathbf{r}_i, \mathbf{r}_j) = u_1(\mathbf{r}_i - \mathbf{r}_j)$, and so $v(\mathbf{r}_i, \mathbf{r}_j) = v(\mathbf{r}_i - \mathbf{r}_j)$. The second quantization formalism of the Hamiltonian can be obtained for plane-wave orbitals [i.e., $\phi_{\mathbf{k}}(\mathbf{r}) = \frac{1}{\sqrt{\Omega_0}} e^{i\mathbf{k} \cdot \mathbf{r}}$, where Ω_0 is the formal volume of the whole space]:

$$\begin{aligned} \hat{H}_{\text{TC-Cell}}^{(N)} = & \hat{H}_{\text{Cell}}^{(N)} + \frac{1}{2\Omega} \sum_{\sigma\sigma'} \sum_{\mathbf{k}\mathbf{p}\mathbf{q}} \{ k^2 \tilde{v}(\mathbf{k}) - (\mathbf{p} - \mathbf{q}) \cdot \mathbf{k} \tilde{v}(\mathbf{k}) - F_0[u_0](\mathbf{k}) - F_1[u_1](\mathbf{k}) \} a_{\mathbf{p}-\mathbf{k},\sigma}^\dagger a_{\mathbf{q}+\mathbf{k},\sigma'}^\dagger a_{\mathbf{q},\sigma'} a_{\mathbf{p},\sigma} \\ & - \frac{1}{2\Omega^2} \sum_{\sigma\sigma'\sigma''} \sum_{\mathbf{k}\mathbf{k}'\mathbf{p}\mathbf{q}\mathbf{s}} \tilde{v}(\mathbf{k}) \tilde{v}(\mathbf{k}') \mathbf{k}' \cdot \mathbf{k} a_{\mathbf{p}-\mathbf{k},\sigma}^\dagger a_{\mathbf{q}+\mathbf{k}',\sigma'}^\dagger a_{\mathbf{s}+\mathbf{k}-\mathbf{k}',\sigma''}^\dagger a_{\mathbf{s},\sigma''} a_{\mathbf{q},\sigma'} a_{\mathbf{p},\sigma} + \frac{N}{2} \xi_{\text{Cell}}, \end{aligned} \quad (17)$$

where Ω is the volume of the supercell. F_0 and F_1 are Fourier transformations of the $(\nabla u)^2$ terms, which become convolutions in reciprocal space:

$$\begin{aligned} F_0[u_0](\mathbf{k}) &= \frac{\Omega}{\Omega_0} \int d\mathbf{r}^3 \left\{ \sum_{\mathbf{m}} [\nabla u_0(\mathbf{r} + \mathbf{R}_m)]^2 e^{i\mathbf{k} \cdot \mathbf{r}} \right\} \\ &= \int d\mathbf{r}^3 \{ [\nabla u_0(\mathbf{r})]^2 e^{i\mathbf{k} \cdot \mathbf{r}} \} \\ &= -\frac{1}{(2\pi)^3} \int (\mathbf{k} - \mathbf{k}') \cdot \mathbf{k}' \tilde{u}_0(\mathbf{k} - \mathbf{k}') \tilde{u}_0(\mathbf{k}') d^3\mathbf{k}', \end{aligned} \quad (18)$$

$$\begin{aligned} F_1[u_1](\mathbf{k}) &= \frac{\Omega}{\Omega_0} \int d\mathbf{r}^3 \left\{ \sum_{\mathbf{m}} [\nabla u_1(\mathbf{r} + \mathbf{R}_m) \cdot \nabla u_1(\mathbf{r} + \mathbf{R}_n)] e^{i\mathbf{k} \cdot \mathbf{r}} \right\} \\ &= \int d\mathbf{r}^3 \left\{ \sum_{\mathbf{m}} [\nabla u_1(\mathbf{r}) \cdot \nabla u_1(\mathbf{r} + \mathbf{R}_m)] e^{i\mathbf{k} \cdot \mathbf{r}} \right\} \\ &= -\frac{1}{(2\pi)^3} \sum_{\mathbf{m}} \int (\mathbf{k} - \mathbf{k}') \cdot \mathbf{k}' \tilde{u}_1(\mathbf{k} - \mathbf{k}') \tilde{u}_1(\mathbf{k}') e^{-i\mathbf{k}' \cdot \mathbf{R}_m} d^3\mathbf{k}' \\ &= -\frac{1}{\Omega} \sum_{\mathbf{k}'} (\mathbf{k} - \mathbf{k}') \cdot \mathbf{k}' \tilde{u}_1(\mathbf{k} - \mathbf{k}') \tilde{u}_1(\mathbf{k}'), \end{aligned} \quad (19)$$

where we have used the fact that for the discretized $\mathbf{k}$ point $e^{-i\mathbf{k} \cdot \mathbf{R}_m} = 1$, and for the continuous $\mathbf{k}'$ vector the sum $\frac{1}{(2\pi)^3} \sum_{\mathbf{m}} e^{-i\mathbf{k}' \cdot \mathbf{R}_m}$ leads to a sum of delta functions. We find that $F_0[u_0](\mathbf{k})$ is given by a continuous convolution, while $F_1[u_1](\mathbf{k})$ is given by a discretized convolution. Therefore the long-range ($k \sim 0$) information of $\tilde{u}_0$ is kept in F_0 , but that of $\tilde{u}_1$ is lost in F_1 . In this work for the UEG, we will focus on the

convergence to the thermodynamic limit and only use u_0 . u_1 will always be set to zero.

A. Determination of a u_0 factor

In the effective TC Hamiltonian, the long-range tail of the Coulomb potential will be canceled as long as the two

asymptotic conditions (7) and (8) are fulfilled. In the momentum space this amounts to

$$\tilde{u}_0(\mathbf{k}) \sim -\frac{4\pi}{\omega_p k^2}, \quad \text{when } k \sim 0, \quad (20)$$

$$\tilde{u}_0(\mathbf{k}) \sim -\frac{4\pi}{k^4}, \quad \text{when } k \sim \infty. \quad (21)$$

A simple form of such a factor can be obtained by the following interpolation,

$$\tilde{u}_{\text{CY}}(k) = -\frac{4\pi}{\omega_p} \left(\frac{1}{k^2} - \frac{1}{k^2 + \omega_p} \right), \quad (22)$$

which is known as the Coulomb-Yukawa (CY) factor and is usually used in its real-space form

$$u_{\text{CY}}(r) = -\frac{1}{\omega_p r} (1 - e^{-\sqrt{\omega_p} r}). \quad (23)$$

The CY factor takes good care of the two asymptotic regions $r_{ij} \rightarrow 0$ and $r_{ij} \rightarrow \infty$, but not of the intermediate region. In order to get a better factor, we can try to optimize the factor based on the single determinant ansatz, by solving a projected Schrödinger equation in the thermodynamic limit [25]

$$\langle \Phi_{\text{HF}} | \delta \tau (\hat{H}_{\text{TC0}}[\tau] - E) | \Phi_{\text{HF}} \rangle = 0, \quad (24)$$

which is a highly nonlinear equation and its numerical solution $\tilde{u}_{\text{TC}}(k)$ will be later referred as the TC factor. This complicated equation can be approximated by taking only its three leading terms

$$\rho k^2 T_2(k) \tilde{u}^2(k) - k^2 \tilde{u}(k) - \frac{4\pi}{k^2} T_2(k) \approx 0, \quad (25)$$

where $T_2(k)$ is a volume factor,

$$T_2(k) = \begin{cases} 1, & k > 2, \\ \frac{3}{4}k - \frac{1}{16}k^3, & k \leq 2. \end{cases} \quad (26)$$

Equation (25) can also be derived by an RPA approximation of a variational treatment of the Jastrow ansatz [29,30] and the solution will be referred to as the RPA factor,

$$\tilde{u}_{\text{RPA}}(k) = \frac{k^2 - \sqrt{k^4 + 16\pi \rho T_2^2(k)}}{2\rho k^2 T_2(k)}. \quad (27)$$

With these factors, the effective electronic potential should decay faster than the Coulomb potential. For a better understanding of this screening effect, it is helpful to examine the effective potential in $\hat{H}_{\text{TC0}}$ induced by u_0 . It contains four terms:

$$\begin{aligned} & -\frac{1}{2} \sum_{ij} \{ \nabla_i^2 u_0(\mathbf{r}_{ij}) + 2[\nabla_i u_0(\mathbf{r}_{ij})] \cdot \nabla_i + [\nabla_i u_0(\mathbf{r}_{ij})]^2 \} \\ & -\frac{1}{2} \sum_{ijk} [\nabla_i u_0(\mathbf{r}_{ij})] \cdot [\nabla_i u_0(\mathbf{r}_{ik})]. \end{aligned} \quad (28)$$

Taking the Coulomb-Yukawa factor (23) as an example, the effective potential arising from the first term is easily evaluated as

$$V_{\text{eff}}^{(1)} = -\nabla^2 u(r) = -\frac{1}{r} e^{-\sqrt{\omega_p} r}. \quad (29)$$

This term is short ranged and it removes the short-range Coulomb singularity of the electronic potential. From the third term of (28) we get a second local potential, which for the CY factor can be calculated as

$$\begin{aligned} V_{\text{eff}}^{(2)}(r) &= -[\nabla u(r)]^2 \\ &= -\left[-\frac{1}{\omega_p r^2} + \left(\frac{1}{\omega_p r^2} + \frac{1}{\sqrt{\omega_p} r} \right) e^{-\sqrt{\omega_p} r} \right]^2. \end{aligned} \quad (30)$$

This term is finite at $r = 0$ and has a fourth-power decay in the long range. As for the three-body term in (28), we consider here its leading long-range RPA contribution, which can also be calculated explicitly for the CY factor,

$$V_{\text{eff}}^{(3)}(r_{ij}) = -\int d^3 r_k [\nabla_k u(r_{ik})] \cdot [\nabla_k u(r_{jk})] \rho \quad (31)$$

$$= -\frac{1}{r_{ij}} + \frac{(\sqrt{\omega_p} r_{ij} + 2)e^{-\sqrt{\omega_p} r_{ij}}}{2r_{ij}}. \quad (32)$$

This term cancels the long-range Coulomb potential, and its remaining part decays exponentially. All other three-body contributions as well as the second term in expression (28) cannot be translated into simple local effective potentials. However, some preliminary analysis of these terms indicates that their leading long-range contribution decays as $\propto r^{-2}$.

The TC factor $u_{\text{TC}}(r)$ should have the same long-range tail as the RPA factor $u_{\text{RPA}}(r)$. An inverse Fourier transformation of $\tilde{u}_{\text{RPA}}(k)$ in Eq. (27) does not lead to a compact analytic expression, but we can still get the long-range expansion

$$u_{\text{RPA}}(r) = -\frac{1}{\omega_p r} + \frac{1}{k_F^2 r^2} + \dots \quad (33)$$

Comparing with Eq. (23), we find that RPA factor has the same long-range leading-order term as the CY factor, but its second long-range leading term decays as r^{-2} which is missing in the CY factor. This makes the CY factor fall slightly below the RPA one in the large r region. Based on expression (33), the long-range leading-order term of the effective potential $V_{\text{eff}}^{(1)}$ and $V_{\text{eff}}^{(2)}$ for the RPA factor can be obtained as

$$V_{\text{eff}}^{(1)}(r) \approx \frac{2}{k_F^2 r^4}, \quad (34)$$

$$V_{\text{eff}}^{(2)}(r) \approx -\frac{1}{\omega_p^2 r^4}. \quad (35)$$

The Evaluation of $V_{\text{eff}}^{(3)}(r)$ for the RPA factor is somewhat tedious, and the final long-range expansion looks like

$$V_{\text{eff}}^{(3)}(r) = -\frac{1}{r} + \frac{4}{\sqrt{3\pi} k_F r^2} + \dots \quad (36)$$

These three factors ($\tilde{u}_{\text{CY}}$, $\tilde{u}_{\text{RPA}}$, and $\tilde{u}_{\text{TC}}$) will be tested in the following study to reveal their effect of the convergence of the TC supercell approach.

B. Convergence of the mean-field energy

As a starting point, we consider the Hartree-Fock determinant as the reference wave function, and calculate the mean-field (MF) energy of the TC Hamiltonian in the

thermodynamic limit:

$$\epsilon_{\infty, \text{MF}}^{(\text{TC})} = \lim_{N \rightarrow \infty} \epsilon_{\text{MF}}^{(\text{TC})}(N), \quad \epsilon_{\text{MF}}^{(\text{TC})}(N) = \frac{\langle \Phi_{\text{HF}}^{(N)} | \hat{H}_{\text{TC-Cell}}^{(N)} | \Phi_{\text{HF}}^{(N)} \rangle}{N}. \quad (37)$$

For the UEG, $\epsilon_{\infty, \text{MF}}^{(\text{TC})}$ can be calculated directly in the thermodynamic limit [24,25]. For nonuniform systems, however, supercell approximations might be required to obtain this value. Since the effective potential in the TC Hamiltonian decays much faster than the Coulomb potential, it is expected that the convergence of the TC mean-field energy should be faster than that of the Hartree-Fock energy in the supercell approximation.

To calculate the mean-field energy for a supercell with N , electrons, we need first to obtain the Madlung constant, which is the interaction of an electron with all its images as well as the positive background

$$\xi_{\text{Cell}} = -\frac{1}{\Omega} \int V_{\text{eff}}(r) d^3r + \sum_{\mathbf{R}_n \neq 0} V_{\text{eff}}(\mathbf{R}_n), \quad (38)$$

where V_{eff} is the total effective potential. As an approximation, we include in V_{eff} only the original Coulomb potential and the several local potentials discussed in the previous section,

$$V_{\text{eff}}(r) = \frac{1}{r} + V_{\text{eff}}^{(2)}(r) + V_{\text{eff}}^{(3)}(r). \quad (39)$$

$V_{\text{eff}}^{(1)}$ is not included, since it arises from the single commutator, and the single commutator does not contribute to the mean-field energy. Equation (38) is quite formal, since each of the two terms may diverge, but their sum remains finite. A simple way to evaluate ξ_{Cell} is to take a cutoff at a large R_{cut} and approximate ξ_{Cell} as

$$\xi_{\text{Cell}} \approx -\frac{1}{\Omega} \int_{r < R_{\text{cut}}} V_{\text{eff}}(r) d^3r + \sum_{\mathbf{R}_n \neq 0, R_n < R_{\text{cut}}} V_{\text{eff}}(\mathbf{R}_n). \quad (40)$$

Take the RPA factor for example: $V_{\text{eff}}(r)$ decays as r^{-2} and thus the leading contribution of the two terms in expression (40) are both $\propto R_{\text{cut}}$, which will then cancel each other. In our numerical calculations, we take mostly $R_{\text{cut}} = 200L_{\text{Cell}}$, where L_{Cell} is the supercell width. With this choice we get usually three effective digits of ξ_{Cell} , which is sufficient for our current calculations.

In the supercell approximation, the Fermi sphere is sampled with a finite number of k points, and this sampling usually leads to severe oscillations of the function $\epsilon_{\text{MF}}^{(\text{TC})}(N)$. To reduce the oscillation, we use twist-averaged boundary conditions [31], where on each dimension we use nine twist angles and so the final results is averaged over 9^3 twist samples. Besides, since the oscillations mainly come from the kinetic energy, which is irrelevant to our current comparison of performances of potentials, we therefore measure only the convergence of the finite-size error of the potential part of $\epsilon_{\text{MF}}^{(\text{TC})}(N)$. In Fig. 1, finite-size errors of the potential part of the transcorrelated mean-field energy calculated for the three different factors at $r_s = 2$ are plotted in comparison with the finite-size error of the Hartree-Fock exchange energy. We find that the slow power-law decay of the finite-size error of the Hartree-Fock exchange energy [32,33] is substantially improved by

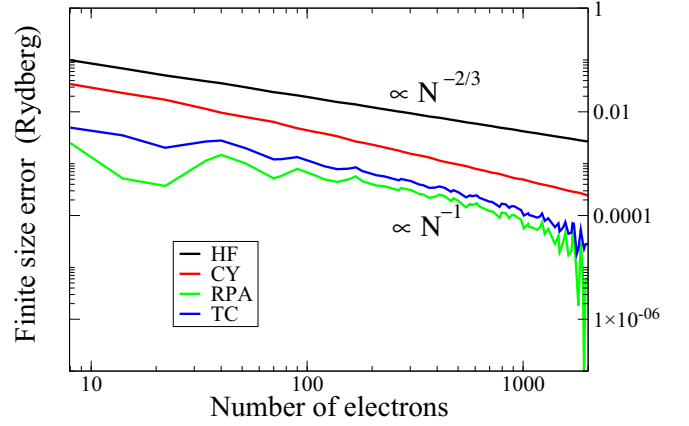


FIG. 1. Finite-size errors of the potential part of the transcorrelated mean-field energy (in the unit of Rydberg) calculated for three different factors at $r_s = 2$, plotted in comparison with the finite-size error of the Hartree-Fock (HF) exchange energy. All three long-ranged correlation factors display a faster decay ($\sim N^{-1}$) of the finite-size error compared to the $\sim N^{-2/3}$ decay of the error of HF exchange. However, the RPA and TC factors also show order-of-magnitude reduced absolute error compared to the CY correlation factor.

the long-range transcorrelated method. Not only is the decay power improved from $N^{-2/3}$ to N^{-1} , but in addition, the prefactor of the power-law decay is reduced by one order of magnitude. With a simulation cell containing only ~ 100 electrons, one can already reach millihartree level accuracy. The performance of the TC factor and the RPA factor are similar and they are obviously better than that of the CY factor.

C. Convergence of the full CI energy

The mean-field energy of the ℓ -TC Hamiltonian contains already most of the long-range dynamic correlations and the missing correlations are mainly static ones and should be even more short ranged. Based on this, in order to achieve a fast convergence, the ℓ -TC correlation energy (per particle) in the supercell approach should be evaluated as

$$\epsilon_{\text{Corr}}^{(\text{TC})}(N) = \frac{\langle \Psi^{(N)} | \hat{H}_{\text{TC-Cell}}^{(N)} | \Psi^{(N)} \rangle}{N} - \epsilon_{\text{MF}}^{(\text{TC})}(N) + \epsilon_{\text{MF}}^{(\text{TC})} - \epsilon_{\text{HF}}. \quad (41)$$

We test this idea by performing full-CI quantum Monte Carlo (FCIQMC) calculations on UEG.

FCIQMC calculations are numerically very expensive for the larger systems and we have limited the calculations to the Baldereschi k points [34] to sample the Brillouin zone. Calculations are performed for $r_s = 1, 2$, and 5.

In a complete transcorrelated FCIQMC calculation, the pure three-body excitation makes the calculations very expensive. On the other hand, it has been found [15,16] in molecular calculations that the pure three-body excitation make negligible contributions to the final results. We have tested this approximation for the ℓ -TC calculations on a small supercell with $N = 14$, and the error due to this approximation is found to be even smaller than the error bar. We therefore ignore

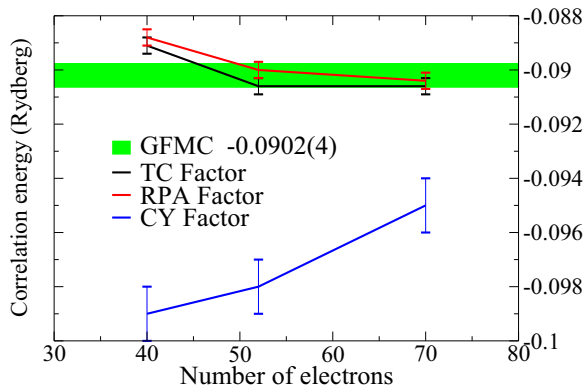


FIG. 2. Correlation energies at $r_s = 2$ calculated according to Eq. (41) with the three correlation factors are plotted as functions of supercell sizes, in comparison with the benchmark GPMC result [35].

the pure three-body excitations in most of our calculations. We have managed to calculate supercells containing up to 70 electrons. Since the correlation factors also describes the short-range cusps correctly, the convergences with respect to basis size are also speeded up. On the largest supercell, most of the calculations converged already at the basis size $M = 2838$ and some of the calculations are checked with $M = 4218$, where M is the number of spin orbitals contained in the basis.

In Fig. 2, correlation energies at $r_s = 2$ calculated according to Eq. (41) with the TC, RPA, and CY factors are plotted as functions of supercell sizes, in comparison with the benchmark Green's function Monte Carlo (GPMC) result [35]. Results of the TC and RPA factors converge very fast to the thermodynamic limit. The performance of the TC factor in particular is especially good, with convergence achieved already at $N = 52$. The result agrees very well with the GPMC result. The performance of the CY factor is relatively poor, where the error at $N = 70$ is still larger than 5% of the total correlation energy. The CY correlation factor also leads to nonvariational total energies, which we consider to be undesirable. The CY factor therefore should be viewed primarily as a model factor in which to illustrate qualitative properties of the TC hamiltonian, but should not be used for quantitative calculations, where both RPA and TC factors fare significantly better.

For $r_s = 1$ and 5, calculations have been performed with the TC factor, and the final results together with $r_s = 2$ are

TABLE I. Correlation energies (in Rydberg unit) calculated with the ℓ -TC method with the TC correlation factor, in comparison with the GPMC results.

r_s	1	2	5
ℓ -TC	-0.121(1)	-0.0906(3)	-0.0560(2)
GPMC	-0.120(1)	-0.0902(4)	-0.0563(1)

presented in Table I, where we find the ℓ -TC results agree very well with the GPMC results.

IV. CONCLUSIONS AND REMARKS

In this work we propose a transcorrelated method to deal with the long-range correlation. By using a properly chosen correlation factor, a similarity transformation is implemented and the resulting transcorrelated Hamiltonian is then treated with the supercell approach. The performance of this method has been examined on three-dimensional uniform electron gas models, where three different correlation factors have been tested. In calculations of both the mean-field energies and the FCIQMC correlation energies, we find this method can greatly improve the speed of convergence to the thermodynamic limit in the supercell approach, both in terms of the scaling (from $N^{-2/3}$ to N^{-1}) as well as the magnitude of the error. The best performing correlation factors are the RPA and TC factors, ahead of the CY correlator.

To extend the application to nonuniform systems, we believe that the RPA or TC factor (based on the averaged charge density) may still be used, since the nonuniform part of the Hamiltonian is largely local and short ranged, and can be handled by short-ranged correlation factors. In this respect, the experience with using TC methods in molecular systems, in which efficient methods to approximate the three-body contributions of the TC hamiltonian via the xTC approximation [16], the calculation of the TC matrix elements using the TCHINT package [12,36], and the application of TC coupled-cluster methods [14,15,17] will be extended to periodic systems and reported in due course.

DATA AVAILABILITY

The data that support the findings of this article are openly available [37], embargo periods may apply.

- [1] 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).
- [2] F. Gygi and A. Baldereschi, Self-consistent Hartree-Fock and screened-exchange calculations in solids: Application to silicon, *Phys. Rev. B* **34**, 4405 (1986).

- [3] J. Spencer and A. Alavi, Efficient calculation of the exact exchange energy in periodic systems using a truncated Coulomb potential, *Phys. Rev. B* **77**, 193110 (2008).
- [4] N. A. W. Holzwarth and X. Xu, Analysis of numerical methods for evaluating the Fock exchange integral in a plane-wave basis, *Phys. Rev. B* **84**, 113102 (2011).
- [5] R. Sundararaman and T. A. Arias, Regularization of the Coulomb singularity in exact exchange by Wigner-Seitz

- truncated interactions: Towards chemical accuracy in nontrivial systems, *Phys. Rev. B* **87**, 165122 (2013).
- [6] S. F. Boys and N. C. Handy, A condition to remove the indeterminacy in interelectronic correlation functions, *Proc. R. Soc. A* **309**, 209 (1969).
- [7] S. F. Boys and N. C. Handy, The determination of energies and wavefunctions with full electronic correlation, *Proc. R. Soc. A* **310**, 43 (1969).
- [8] R. Jastrow, Many-body problems with strong forces, *Phys. Rev.* **98**, 1479 (1955).
- [9] S. Ten-no, A feasible transcorrelated method for treating electronic cusps using a frozen Gaussian geminal, *Chem. Phys. Lett.* **330**, 169 (2000).
- [10] O. Hino, Y. Tanimura, and S. Ten-no, Biorthogonal approach for explicitly correlated calculations using the transcorrelated Hamiltonian, *J. Chem. Phys.* **115**, 7865 (2001).
- [11] H. Luo and A. Alavi, Combining the transcorrelated method with full configuration interaction quantum Monte Carlo: Application to the homogeneous electron gas, *J. Chem. Theory Comput.* **14**, 1403 (2018).
- [12] A. J. Cohen, H. Luo, K. Guthrie, W. Dobrutz, D. P. Tew, and A. Alavi, Similarity transformation of the electronic Schrödinger equation via Jastrow factorization, *J. Chem. Phys.* **151**, 061101 (2019).
- [13] A. Baiardi and M. Reiher, Transcorrelated density matrix renormalization group, *J. Chem. Phys.* **153**, 164115 (2020).
- [14] K. Liao, T. Schraivogel, H. Luo, D. Kats, and A. Alavi, Towards efficient and accurate *ab initio* solutions to periodic systems via transcorrelation and coupled cluster theory, *Phys. Rev. Res.* **3**, 033072 (2021).
- [15] T. Schraivogel, A. J. Cohen, A. Alavi, and D. Kats, Transcorrelated coupled cluster methods, *J. Chem. Phys.* **155**, 191101 (2021).
- [16] E. M. Christlmaier, T. Schraivogel, P. López Ríos, A. Alavi, and D. Kats, xTC: An efficient treatment of three-body interactions in transcorrelated methods, *J. Chem. Phys.* **159**, 014113 (2023).
- [17] T. Schraivogel, E. M. Christlmaier, P. López Ríos, A. Alavi, and D. Kats, Transcorrelated coupled cluster methods. II. Molecular systems, *J. Chem. Phys.* **158**, 214106 (2023).
- [18] K. Liao, H. Zhai, E. M. Christlmaier, T. Schraivogel, P. López Ríos, D. Kats, and A. Alavi, Density matrix renormalization group for transcorrelated Hamiltonians: Ground and excited states in molecules, *J. Chem. Theory Comput.* **19**, 1734 (2023).
- [19] J. P. Haupt, E. M. Corvid Christlmaier, P. L. Ríos, N. A. Bogdanov, D. Kats, and A. Alavi, Transcorrelated methods for multireference problems, *J. Chem. Phys.* **163**, 144113 (2025).
- [20] M. Mörchen, A. Baiardi, M. Lesiuk, and M. Reiher, Non-iterative triples for transcorrelated coupled cluster theory, *J. Chem. Theory Comput.* **21**, 1588 (2025).
- [21] K. Simula, M.-A. Filip, and A. Alavi, Transcorrelated theory for transition-metal atoms, *Phys. Rev. A* **112**, 032805 (2025).
- [22] K. Simula, E. M. Corvid Christlmaier, M.-A. Filip, J. P. Haupt, D. Kats, P. Lopez-Rios, and A. Alavi, Transcorrelated theory with pseudopotentials, *J. Chem. Theory Comput.* **21**, 5155 (2025).
- [23] M.-A. Filip, P. L. Ríos, J. P. Haupt, E. M. C. Christlmaier, D. Kats, and A. Alavi, Transcorrelated methods applied to second row elements, *J. Chem. Phys.* **162**, 064110 (2025).
- [24] N. Umezawa and S. Tsuneyuki, Ground-state correlation energy for the homogeneous electron gas calculated by the transcorrelated method, *Phys. Rev. B* **69**, 165102 (2004).
- [25] H. Luo, Transcorrelated calculations of homogeneous electron gases, *J. Chem. Phys.* **136**, 224111 (2012).
- [26] H. Luo and A. Alavi, Perturbation calculation of the uniform electron gas with a transcorrelated Hamiltonian, *J. Chem. Phys.* **157**, 074105 (2022).
- [27] S. F. Boys, Some bilinear convergence characteristics of the solutions of dissymmetric secular equations, *Proc. R. Soc. A* **309**, 195 (1969).
- [28] E. A. G. Armour, The calculation of the ground-state energy of the free-electron gas by the transcorrelated method, *J. Phys. C: Solid State Phys.* **13**, 343 (1980).
- [29] T. Gaskell, The collective treatment of a Fermi gas: II, *Proc. Phys. Soc.* **77**, 1182 (1961).
- [30] T. Gaskell, The collective treatment of many-body systems: III, *Proc. Phys. Soc.* **80**, 1091 (1962).
- [31] C. Lin, F. H. Zong, and D. M. Ceperley, Twist-averaged boundary conditions in continuum quantum Monte Carlo algorithms, *Phys. Rev. E* **64**, 016702 (2001).
- [32] 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).
- [33] M. Ruggeri, P. López Ríos, and A. Alavi, Correlation energies of the high-density spin-polarized electron gas to meV accuracy, *Phys. Rev. B* **98**, 161105(R) (2018).
- [34] A. Baldereschi, Mean-value point in the Brillouin zone, *Phys. Rev. B* **7**, 5212 (1973).
- [35] D. M. Ceperley and B. J. Alder, Ground state of the electron gas by a stochastic method, *Phys. Rev. Lett.* **45**, 566 (1980).
- [36] J. P. Haupt, S. M. Hosseini, P. López Ríos, W. Dobrutz, A. Cohen, and A. Alavi, Optimizing Jastrow factors for the transcorrelated method, *J. Chem. Phys.* **158**, 224105 (2023).
- [37] H. Luo, Raw data to accompany the paper “Transcorrelated Approach to the Thermodynamic Limit” [Dataset] version 1, Edmond, (2026), <https://doi.org/10.17617/3.VCJFZV>.