Legendre-spectral Dyson equation solver with super-exponential convergence

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Quantum many-body systems in thermal equilibrium can be described by the imaginary time Green’s function formalism. However, the treatment of large molecular or solid ab initio problems with a fully realistic Hamiltonian in large basis sets is hampered by the storage of the Green’s function and the precision of the solution of the Dyson equation. We present a Legendre-spectral algorithm for solving the Dyson equation that addresses both of these issues. By formulating the algorithm in Legendre coefficient space, our method inherits the known faster-than-exponential convergence of the Green’s function’s Legendre series expansion. In this basis, the fast recursive method for Legendre polynomial convolution, enables us to develop a Dyson equation solver with quadratic scaling. We present benchmarks of the algorithm by computing the dissociation energy of the helium dimer He$_2$ within dressed second-order perturbation theory. For this system, the application of the Legendre spectral algorithm allows us to achieve an energy accuracy of $10^{-9}E_h$ with only a few hundred expansion coefficients.

I. INTRODUCTION

The equilibrium properties of many-body quantum systems can be described by the finite temperature imaginary-time Green’s function formalism, which is widely applicable to condensed matter physics, quantum chemistry, and material science. Applications include numerical methods for low energy effective model Hamiltonians such as lattice Monte Carlo, dynamical mean field theory and its extensions, and diagrammatic Monte Carlo. Ab initio calculations using the random phase approximation, self-consistent second order perturbation theory, Hedin’s GW approach, and self energy embedding theory can also be formulated in imaginary time.

While the finite temperature Green’s function formalism is very successful in applications to model Hamiltonians, its applicability to quantum chemistry and materials science remains limited to simple molecular and periodic problems. This is due to the necessity of simultaneously describing both the core and valence orbitals, which results in an energy scale that is difficult to describe by a single imaginary time/frequency grid. A simple equidistant Matsubara grid would contain millions of points, thus making the storage and manipulation of the Green’s functions computationally costly. In contrast, a grid with only a small number of equidistant points will result in a poorly converged energy or density matrix, making calculations with Hartree precision challenging. Such precision is necessary in applications where the evaluation of interaction energies, energies of conformers, or energies of high-lying excited states is needed. Consequently, it is important to develop a compact representation that yield highly converged properties.

With the standard approach using equidistant Matsubara frequency grids with finite frequency cut-off, the imaginary time Green’s function only converges to the analytical result linearly in the number of Matsubara frequencies. Amending the representation with a low order high frequency expansion results in polynomial convergence. In practice, this is problematic, since for systems with a wide range of energy scales, the number of coefficients is controlled by the largest energy scale. Alternatives such as uniform power meshes have had some success. However, the most compact representations are achieved using a set of (orthogonal) continuous basis functions directly in imaginary time, such as orthogonal polynomials or numerical basis functions. The convergence of such a representation is faster than exponential and asymptotically superior to any polynomially converging representation.

In all imaginary time methods a central step is the solution of the Dyson equation for the single particle Green’s function. In the Matsubara frequency representation, the Dyson equation is diagonal and can be readily solved. However, the solution is plagued by the polynomial convergence with respect to the number of frequency coefficients used. In imaginary time the Dyson equation is a non-trivial integro-differential equation with a mixed boundary condition. Recently an algorithm for solving the Dyson equation in imaginary time using the Chebyshev polynomials has been presented. This algorithm preserves the exponential convergence of the orthogonal polynomial expansions. However, the central convolution step has a cubic scaling in the expansion order $N_L \sim O(N^3_L)$, which limits the applicability of the algorithm.

The development of compact representations and algorithms for solving the Dyson equation is an active field of research, see Tab. for an overview of the state-of-the-art methods. For a recent development see Ref.

In this paper, we present a Legendre spectral method...
for solving the Dyson equation with super exponential convergence and a convolution that scales quadratically \( \sim O(N^2) \), one order better than previous formulations. The super exponential convergence allows us to achieve an energy accuracy of \( 10^{-9}E_h \) in a realistic quantum chemistry system with a few hundred expansion coefficients. We show this in a proof-of-concept benchmark: computing the dissociation energy of He\(_2\) using self-consistent second order perturbation theory, taking both the zero temperature and the complete basis limit.

This paper is organized as follows. In section II, we introduce the Dyson equation. In section III, we present our Legendre spectral method. In section IV and V, we apply our method to a realistic quantum chemistry problem, the dissociation energy of the noble gas He\(_2\). In section VI we present conclusions.

II. THEORY

The imaginary time single particle Green’s function \( G \) is defined on the interval \( \tau \in [-\beta, \beta] \), \( G(\tau) \equiv G(\beta - \tau) \), with \( \xi = +1 \) \((-1)\) for bosons (fermions), making it an (anti-)periodic function with a step discontinuity at \( \tau = 0 \), see Fig. 1. The imaginary time Dyson equation for \( G(\tau) \) is given by

\[
[-\partial_\tau - h]G(\tau) - \Sigma * G = 0,
\]

where \( h \) is the single particle energy, and \( \Sigma \) the self-energy, which accounts for all many-body interactions. We note in passing that \( \Sigma(\tau) \) has the same periodicity as \( G(\tau) \). The boundary condition for Eq. (1) is \( G(0) - \xi G(\beta) = -1 \), and the Fredholm type imaginary time convolution is defined as \( \Sigma * G \equiv \int_0^\beta d\tilde{\tau} \Sigma(\tau - \tilde{\tau})G(\tilde{\tau}) \).

Analytically the Dyson equation (Eq. 1) can be solved using the Fourier series expansion

\[
G(\tau) = \frac{1}{\beta} \sum_{n=-\infty}^{\infty} e^{-i\omega_n \tau} G(i\omega_n), \quad G(i\omega_n) = \int_0^\beta d\tau e^{i\omega_n \tau} G(\tau),
\]

where the Matsubara frequencies \( i\omega_n \) are given by \( i\omega_n = \frac{\pi}{\beta} (2n + \eta) \) with \( \eta = (1 - \xi)/2 \), and \( n \) integers. In Matsubara frequency space the Dyson equation (Eq. 1) is diagonal

\[
[i\omega_n - h - \Sigma(i\omega_n)]G(i\omega_n) = 1.
\]

Numerically, however, the discontinuity at \( \tau = 0 \) results in a slow asymptotic decay \( G(i\omega_n) \sim (i\omega_n)^{-1} \), see Fig. 1. This prevents a naive finite frequency \( |n| < N_\omega \) approximation \( G(\tau) \approx \frac{1}{\beta} \sum_{|n| < N_\omega} e^{-i\omega_n \tau} G(i\omega_n) \) from converging in \( N_\omega \) (the maximal error in \( G(\tau) \) scales as \( \sim O(N_\omega) = O(1) \)). The standard solution to this problem is to use a finite number \( p \) of high-frequency "tail" coefficients \( G_k \) to approximate \( G(i\omega_n) \approx \sum_k G_k/(i\omega_n)^k \) for \( |n| > N_\omega \), where the known asymptotic decay implies \( G_1 = 1 \). This type of tail correction procedure gives polynomial convergence in \( G(\tau) \) with the power determined by the order \( p \) of the tail expansion \( \sim O(N_\omega^{-p}) \), see e.g. Ref. [42–44]. In Fig. 2 this is shown for the case of \( p = 3 \) using the TRIQS library [61].

Since \( G(\tau) \) is infinitely derivable on \( \tau \in [0, \beta] \), it can be much more efficiently represented by a finite orthogonal polynomial expansion

\[
G(\tau) \approx \sum_{n=0}^{N_L} G_n L_n[x(\tau)],
\]

where \( L_n[x] \) are Legendre polynomials defined on \( x \in [-1, 1] \) and \( x(\tau) = \frac{\tau}{\beta} - 1 \). The Legendre coefficients \( G_n \) have a faster than exponential asymptotic decay, see Fig. 1. This also causes the finite \( N_L \) expansion at the right hand side of Eq. (3) to converge faster than exponential \( \lesssim O(e^{-N_L}) \) to the analytical \( G(\tau) \).

III. LEGENDRE SPECTRAL METHOD

Here we develop a Legendre spectral method for solving the Dyson equation (Eq. 1), reformulating the
integro-differential equation in the space of Legendre coefficients $G_n$ (Eq. 1). In the space of a finite Legendre expansion of order $N_L$, Eq. 1 is cast to a linear equation

$$\partial_{\tau} L_n[x(\tau)] = \frac{2}{\beta} \partial_x L_n(x)$$

$$= \frac{2}{\beta} \sum_{k=0, \text{odd}}^{n-1} (2k+1) L_k(x) = \sum_k D_{kn} L_k(x).$$

Hence the derivative matrix $D_{kn}$ in Eq. 4 is given by

$$\frac{\beta}{2} D_{kn} \equiv \begin{cases} 2k+1, & 0 \leq k \leq n, k+n \text{ odd} \\ 0, & \text{elsewhere} \end{cases},$$

and is upper triangular, see Fig. 3. Using $L_n(\pm1) = (\pm1)^n$ the Dyson equation boundary condition can be written as

$$-1 = G(0) - \xi G(\beta) = \sum_n ((-1)^n - \xi) G_n. \quad (7)$$

A. Spectral convolution

The imaginary time convolution $[\Sigma * G]$ in the Dyson equation (Eq. 1) can be separated into the two terms of Volterra type

$$[\Sigma * G](\tau) = \int_0^\beta d\tau' \Sigma(\tau - \tau') G(\tau') \quad (8)$$

$$= \int_0^\tau d\tau' \Sigma(\tau - \tau') G(\tau') + \int_\tau^\beta d\tau' \xi \Sigma(\beta + \tau - \tau') G(\tau'),$$

FIG. 1. Single particle Green’s function in a) imaginary time $G(\tau)$, b) Matsubara frequency $G(i\omega_n)$ (with $(i\omega_n)^{-1}$ black line), and c) Legendre expansion coefficients $G_n$, for site one in the fermionic two level system with the second quantization Hamiltonian $H = -\mu c_i^c c_i + V(c_i^c c_2 + c_2^c c_1) + V_0 c_i^c c_2$ at inverse temperature $\beta = 1$, where $c_i^c$ creates and $c_i$ annihilates a fermion at site $i$ and $\mu = -3, \epsilon = 3.3, V = 4$.

FIG. 2. Error in density $\Delta n$ as a function of Legendre expansion order $N_L$ and number of Matsubara frequencies $N_\omega$, for the same system as in Fig. 1.

FIG. 3. (Color online) Matrix structure of the spectral derivative operator $D_{kn}$ and the convolution operator $[\Sigma * G]$ for $\epsilon = 1$ and $\Sigma(\tau) = e^{-\tau\beta} (\xi e^{-\beta} - 1)^{-1}$ at $\beta = 1$ and $\xi = -1$ (fermions).
using the periodicity property \( \Sigma(-\tau) = \xi \Sigma(\beta - \tau) \). In
Eq. 8, \( \Sigma(\tau) \) is only evaluated for \( \tau \in [0, \beta] \), avoiding the
discontinuity at \( \tau = 0 \).

In Legendre space the convolution operator \([\Sigma \ast] \) can be written as a sum of two matrices \( B_{kn}^{\leq} \) rep-resenting the two Volterra terms Eq. 8
\[
[\Sigma \ast]_{kn} \equiv B_{kn}^{\leq} + \xi B_{kn}^{\geq} .
\]
(9)

Stable recursion relations for \( B_{kn}^{\leq} \) have been derived by
Hale and Townsend\(^\text{63} \) using the Fourier connection be-tween Legendre polynomials and spherical Bessel functions. Since the derivation is detailed in Ref. 63 we only state the result specialized to to the imaginary time con-
version in Eq. 8 here, and provide a derivation in Appendix A.

The coefficients are related by the recursion relation
\[
B_{k,n+1}^{\leq} = -\frac{n+1}{2k+3} B_{k+1,n}^{\leq} + \frac{2n+1}{2k-1} B_{k-1,n}^{\leq} + B_{k,n-1}^{\leq}
\]
(10)
which for each column require two previous columns to be
known. The recursion is only for the lower triangular coef-
ficients in \( B_{kn}^{\leq} \). The upper triangular coefficients are
computed using the transpose relation
\[
B_{k,n}^{\leq} = (-1)^{n+k} \frac{2k+1}{2n+1} B_{n,k}^{\leq} .
\]
(11)
The two first columns are given by the starting relations
\[
B_{k,0}^{\leq} = \begin{cases} 
\Sigma_0 \pm \frac{\Sigma_1}{3}, & k = 0 \\
\pm \frac{\Sigma_{k-1} - \Sigma_{k+1}}{2k+3}, & k \geq 1
\end{cases},
\]
(12)
with the special case for \( k = 0 \), \( B_{0,1}^{\leq} = \pm B_{1,0}^{\geq} / 3 \), using
the Legendre coefficients \( \Sigma_n \) of the self-energy \( \Sigma \), c.f. Eq. 3.

B. Convergence and scaling

Since each coefficient in \( B_{kn}^{\leq} \) can be computed in \( O(1) \)
operations, the scaling of the convolution matrix con-
struction is \( \sim O(N_L^2) \). The self-energy \( \Sigma(\tau) \) is a smooth
function with asymptotic exponentially decaying Legendre coefficients which causes the entries of the dominantly
diagonal spectral convolution operator \([\Sigma \ast]_{kn} \) to decay exponentially both along and away from the diagonal, see Fig. 3.

The numerical solution of \( G(\tau) \) from the Dyson equation
constructed in terms of the linear system in Eq. 14 converges faster than exponentially to the analytical
solution, with increased number of Legendre coefficients \( N_L \), see Fig. 2. This is in stark contrast to the polynomial con-
vergence of the standard Matsubara tail approach\(^{14} \), also shown in Fig. 2.

C. Imaginary time transform

To retain the high accuracy of the Legendre spectral
Dyson solver the method has to be complemented with stable transforms between Legendre coefficients and imaginary time
\[
G_n = \sum_{i=0}^{N_L} S_{ni} G(\tau_i), \quad G(\tau_i) = \sum_{n=0}^{N_L} L_{in} G_n .
\]
(13)

To construct the well-conditioned transform matrices \( S_{ni} \) and \( L_{in} \) we employ Legendre quadrature and the Legendre-Gauss-Lobatto points \( x_i \in \{ x : (1 - x^2)L_{N_L}(x) = 0 \} \), \( x_0 = -1, x_N = 1 \), re-scaled to the imaginary time interval \([0, \beta] \), \( \tau_i = \beta x_i + 1 \). Using \( x_i \), the matrices \( S_{ni} \) and \( L_{in} \) can be directly constructed (avoiding
matrix inversion)
\[
L_{in} = L_n(x(\tau_i)) , \quad S_{ni} = \frac{\beta}{2W_n} \omega_i L_n(x(\tau_i)),
\]
(14)
where \( \int_{-1}^{1} dx L_n(x)L_m(x) = \delta_{nm} \frac{2}{2n+1} \equiv \delta_{nm} W_n \) and
\( \omega_i = \frac{2}{N(N+1)} L_{N_L}(x_i) \), see e.g. Refs. 62 and 64.

IV. APPLICATION (GF2)

As a proof of concept application of the Legendre
spectral Dyson solver developed in this paper we em-
ploy the solver in a quantum chemistry setting using a
Gaussian basis set. We will employ self-consistent sec-
dorder order perturbation theory, also known as GF2\(^3 \),
which has seen a revival in recent years, both in ab initio condensed matter applications\(^6 \) and in quantum chemistry\(^1 \) in combination with embedding method.\(^2 \) Our implementation is built on the Coulomb-
integrals of the pyscf library\(^6 \).

In the resulting non-orthogonal basis set the Dyson
equation takes the form
\[
\sum_j [ S_{ij}(\partial_\tau - \mu) + F_{ij} + \Sigma_{ij}^s ] G_{jk}(\tau) = 0
\]
(15)
in which \( i,j,k \) are orbital indices, \( S_{ij} \) is the overlap
matrix, and \( F_{ij} \) is the so called Fock matrix, \( F_{ij} \equiv h_{ij} + \Sigma_{ij}^{\text{(HF)}} \). The boundary condition for this equation is \( \sum_j (G_{ij}(0) - \xi G_{ij}(\beta)) \cdot S_{jk} = -1_{ik} \). Here, the single
particle term \( h_{ij} \) accounts for electronic kinetic and
electrostatic matrix elements and the Hartree-Fock
self energy \( \Sigma_{ij}^{\text{(HF)}} \) is given by
\[
\Sigma_{ij}^{\text{(HF)}} = \sum_{kl} P_{kl}(v_{ijkl} - v_{iklj}/2) ,
\]
(16)
were \( P_{ij} \) the density matrix \( P_{ij} = -2G_{ij}(\beta) \), and \( v_{ijkl} \)
the electron-electron Coulomb repulsion integral.

In GF2 the imaginary-time-dependent part of the self
energy \( \Sigma(\tau) \) is approximated with the second order self
energy diagram using the full electron Greens function $G$, $\Sigma \approx \Sigma^{(\text{GF2})}[G]$ where

$$
\Sigma^{(\text{GF2})}_{ij}(\tau) = \sum_{klmpq} G_{kl}(\tau) G_{mn}(\tau) G_{pq}(\beta - \tau) \times v_{impk}(2v_{jnlq} - v_{jlnq}).
$$

The evaluation of $\Sigma^{(\text{GF2})}_{ij}(\tau)$ for fixed $\tau$ scales as $\sim O(N^5)$ where $N$ is the number of atomic orbitals.

Solving for the GF2 Greens function $G$ amounts to solving Eqs. (15), (16), and (17) which is a highly nonlinear problem. To find the solution we perform self-consistent iterations, see Fig. 4 for a schematic picture. The inner loop solves the Dyson equation [Eq. (15)] and updates the Hartree-Fock self energy $\Sigma^{(\text{HF})}$ [Eq. (16)] until convergence (in the Fock-matrix $F$). At convergence in $F$, one step of the outer loop is performed by re-evaluating the GF2 self energy $\Sigma^{(\text{GF2})}$ [Eq. (17)] and computing the relative change in total energy $E$. If the change is above a fixed threshold, the inner loop is started again. To compute the inter molecular energies, which is an energy difference, we need a threshold of $10^{-10}$.

The total energy $E$ of the system is given by

$$
E = \frac{1}{2} \text{Tr}[(h + F) P] + \text{Tr}[\Sigma \ast G] + E_{\text{nuc}},
$$

where $E_{\text{nuc}}$ is the nuclei-nuclei Coulomb energy. The imaginary time trace $\text{Tr}[:]$ is defined as $\text{Tr}[A] \equiv -\sum_{\alpha} A_{\alpha}(\beta)$, and the $\Sigma \ast G$ convolution is computed with the spectral Legendre convolution as in Eq. (4).

V. RESULTS

The faster than exponential convergence of the Legendre spectral Dyson solver Eq. (4) is particularly suited for high precision calculations. A prime example is the computation of the binding energy $D_e$ in noble-gas dimers, where the weak bonding requires high precision calculations of total energies. The binding energy $D_e$ is obtained from the minimum of the interaction energy $E_{\text{int}}(r)$ as a function of atomic separation $r$

$$
D_e \equiv -E_{\text{int}}(r_e) \equiv -\min_r E_{\text{int}}(r),
$$

where $r_e$ is the equilibrium atomic distance. The interaction energy $E_{\text{int}}$ is in turn given by

$$
E_{\text{int}}(r) = E_{A_2}(r) - 2E_A(r),
$$

where $E_{A_2}$ is the total energy of the dimer and $E_A$ is the total energy of the single atom (the monomer) evaluated using the standard counterpoise correction. In noble gases the total energies $E_A$ and $E_{A_2}$ are of the order of Hartrees ($\sim E_h \equiv 1$ Ha) while the binding energy $D_e$ is of the order of tens of micro Hartrees ($\sim 10^{-4}$ eV), hence requiring high precision calculation of the total energies.

We use He$_2$ as a prototype system since there exist published reference results for the binding energy $D_e$ and equilibrium distance $r_e$ calculated with Hartree-Fock (HF), second-order Moller-Plesset perturbation theory (MP2), coupled cluster single double (CCSD) theory and coupled cluster single double triple (CCSD(T)) theory. The MP2 method is closely related to GF2 and uses the second order self energy [Eq. (17)] evaluated at the HF Green’s function $G^{(\text{HF})}$, $\Sigma^{(\text{MP2})} = \Sigma^{(\text{GF2})}G^{(\text{HF})}$. Note however that the prefactors in the total energy differ.

Fig. 5 shows $E_{\text{int}}(r)$ (and $-D_e$) of He$_2$ computed with HF, MP2, and GF2 in the aug-cc-pvqz basis together with CCSD and CCSD(T) reference results on $D_e$. The GF2 results are obtained by fitting a 4th order polynomial to 21 $r$-points of $E_{\text{int}}(r)$ computed in a 0.1 Bohr range centered around the minimum at $r_e$. The GF2 results are obtained using the Legendre spectral Dyson solver while HF and MP2 are computed using pyscf. As seen in Fig. 5 He$_2$ does not bind within the Hartree-Fock approximation which gives a strictly positive interaction energy. Compared to MP2 our GF2 results are a considerable improvement, using the coupled cluster CCSD and CCSD(T) as reference.
TABLE II. Dissociation energies $D_e$ (top) and equilibrium distances $r_e$ computed by MP2, CCSD, CCSD(T), and GF2 with the basis sets aug-cc-pv$\{d,t,q,5\}$, with $n = d, t, q, 5$. The MP2, CCSD and CCSD(T) results are from Ref. 71.

| $D_e$ [$\mu E_h$] | MP2 | CCSD | CCSD(T) | GF2 |
|-------------------|-----|------|---------|-----|
| aug-cc-pv$dz$     | 12.69 | 16.78 | 18.57   | 18.17 |
| aug-cc-pv$tz$     | 17.97 | 23.77 | 27.10   | 24.63 |
| aug-cc-pv$qz$     | 19.66 | 25.79 | 29.64   | 26.59 |
| aug-cc-pv$5z$     | 20.71 | 27.09 | 31.25   | 27.79 |
| CBS               | 22.98 | 30.06 | 34.70   | 29.67 |

| $r_e$ [Bohr] | MP2 | CCSD | CCSD(T) | GF2 |
|------------|-----|------|---------|-----|
| aug-cc-pv$dz$ | 6.1680 | 6.0580 | 6.0086 | 6.0547 |
| aug-cc-pv$tz$ | 5.9175 | 5.8060 | 5.7452 | 5.8244 |
| aug-cc-pv$qz$ | 5.8606 | 5.7546 | 5.6891 | 5.7272 |
| aug-cc-pv$5z$ | 5.8244 | 5.7210 | 5.6537 | 5.7388 |
| CBS         | 5.769 | 5.672 | 5.607   | 5.680  |

A. Complete basis set limit

In order to extrapolate the results to the complete basis set (CBS) limit\cite{13},\cite{14},\cite{15}\cite{16}, we repeat the calculations using the augmented correlation consistent (aug-cc-pv$\{d,t,q,5\}$) basis set series with $n = d, t, q, 5$ (i.e., $n = 2, 3, 4, 5$). This series has been shown to enable accurate extrapolation of a number of properties due to its systematic convergence\cite{17} with the basis sets aug-cc-pv$\{d,t,q,5\}$, with $n = d, t, q, 5$. The MP2, CCSD, and CCSD(T) results are from Ref. 71.

In Table II, we summarize the binding energy $D_e$ and equilibrium distance $r_e$ of He$_2$ computed by MP2, CCSD, CCSD(T), and GF2 using the aug-cc-pv$\{d,t,q,5\}$ basis sets. The aug-cc-pv$\{d,t,q,5\}$ basis sets are of the same order as the number of atomic orbitals $N$. Hence, the scaling of GF2, $O(1)$, is comparable to the scaling of CCSD, $O(N^6)$. As seen in Table II, the GF2 result for $D_e$ is closer to CCSD(T) than CCSD, while the CCSD result for $r_e$ is closer to CCSD(T) result than GF2, making GF2 competitive to CCSD and a considerable improvement compared to MP2.

With the systematic convergence of $D_e$ and $r_e$ as a function of basis set size $n$, it is possible to extrapolate to the complete basis set limit $n \to \infty$. We extrapolate $D_e$ and $r_e$ using our GF2 aug-cc-pv$\{d,t,q,5\}$ results by fitting the exponential model: $A \cdot e^{-B(n-2)} + C$, proposed in Ref. 71, where $A$, $B$, and $C$ are parameters. The applicability of the model is checked by a logarithmic plot, see Fig. 6. The resulting CBS limit of our GF2 results are $D_e \approx 29.67 \mu E_h$ and $r_e \approx 5.680 a_0$, see also Table II.

VI. CONCLUSION AND OUTLOOK

We introduce a Legendre-spectral algorithm for solving the Dyson equation in Legendre coefficient space. By staying in Legendre-coefficient space the algorithm converges super exponentially with respect to the number of Legendre coefficients $N_L$ used to represent the imaginary time Green’s function\cite{18}. This is in stark contrast to the Matsubara frequency space based approach with only polynomial convergence\cite{19},\cite{20}. The exponential convergence is shared with a recently presented Chebyshev polynomial based algorithm\cite{21} where the convolution scales as $O(N_L^2)$. Currently there is no known algorithm for Chebyshev series that can compute the convolution term with the same efficiency as in the Legendre series\cite{22}. Our work goes beyond this, employing a Legendre convolution with $O(N_L^2)$ scaling, enabling the application to larger $ab$ initio systems.

To benchmark the algorithm we apply it to the quantum chemistry computation of the dissociation energy of the noble gas He$_2$ using self-consistent second order perturbation theory (GF2). The exponential convergence of our algorithm allows us to reach the required 10$^{-10}$ $E_h$ zero temperature total-energy precision using only 100 – 200 Legendre coefficients in the Dunning basis series aug-cc-pv$\{d,t,q,5\}$.

The algorithm is also relevant for condensed matter $ab$ initio applications in periodic systems that require high precision, such as GF2\cite{23} and Hedin’s GW\cite{24}. This is a promising venue for future research.

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Appendix A: Convolution matrix

In this appendix, we derive Eqs. \[10\] \[11\] \[12\] in the main text. The derivation follows Ref. \[63\] but with more details for both integrals in Eq. \[8\]

1. Convolution and Fourier transform

The convolution of two continuous integrable functions is defined in Ref. \[63\]

\[
h(x) = (f * g)(x) \equiv \int_{-\infty}^{\infty} dt \, f(t)g(x - t). \tag{A1}
\]

With the assumption \(f\) and \(g\) are periodic functions, their Fourier transform can be written as

\[
\mathcal{F}\{f\}(\omega) = \int_{-\infty}^{\infty} dx \, e^{-i\omega x}f(x), \tag{A2}
\]

\[
\mathcal{F}^{-1}\{f\}(x) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dx \, e^{i\omega x}f(x), \tag{A3}
\]

which satisfy the Fourier inversion theorem \(\mathcal{F}^{-1}\{\mathcal{F}\{f\}\} = f\) and convolution theorem \[63\]

\[
\mathcal{F}\{f * g\} = \mathcal{F}\{f\} \cdot \mathcal{F}\{g\}. \tag{A4}
\]

2. Legendre polynomials

The Legendre polynomials \(P_n(x)\) can be defined recursively using the three term recurrence relation

\[
P_0(x) = 1, \quad P_1(x) = x, \quad (n + 1)P_{n+1}(x) = (2n + 1)xP_n(x) - nP_{n-1}(x). \tag{A5}
\]

They are orthogonal on \([-1, 1]\)

\[
\int_{-1}^{1} dx \, P_m(x)P_n(x) = \delta_{m,n} \frac{2}{2n + 1} \tag{A6}
\]

and the derivatives satisfy the recurrence relation

\[
(2n + 1)P_n(x) = \frac{d}{dx} [P_{n+1}(x) - P_{n-1}(x)] \tag{A7}
\]

The Fourier transform and inverse Fourier transform of the Legendre polynomials can be expressed in terms of Bessel functions

\[
\mathcal{F}\{P_n\} = \int_{-1}^{1} dx \, e^{-i\omega x}P_n(x) = 2(-i)^n j_n(\omega) \tag{A8}
\]

\[
\mathcal{F}^{-1}\{P_n\} = \int_{-1}^{1} dx \, e^{i\omega x}P_n(x) = 2i^n j_n(\omega) \tag{A9}
\]

where \(j_n(z)\) is the \(n\)th spherical Bessel function, and \(P_n = 0\) outside \([-1, 1]\).

By combining Eq. \[A4\] and Eq. \[A8\], the convolution of Legendre polynomials can be expressed in terms of Bessel functions

\[
(P_m * P_n)(x) = \frac{2(-i)^{m+n}}{\pi} \int_{-\infty}^{\infty} d\omega \, e^{i\omega x} j_m(\omega)j_n(\omega) \tag{A10}
\]

This is the central observation of Ref. \[63\] that enables the derivation of recursion relations for the Legendre polynomial convolution.

The main property of Spherical Bessel functions used is the three term recurrence relation

\[
j_{n-1}(z) = \frac{\cos z}{z} j_n(z) - \frac{1}{z} j_n(z), \quad n \geq 0 \tag{A11}
\]

The convolution equation Eq. \[A1\] can be computed by replacing the two continuous functions \(f(x)\) and \(g(x)\) on bounded interval with polynomial approximates \(f_M(x)\) and \(g_N(x)\) of sufficiently high degree. With two Legendre series \(f_M(x)\) and \(g_N(x)\) supported on \(x \in [-1, 1]\)

\[
f_M(x) = \sum_{m=0}^{M} a_m P_m(x), \quad g_N(x) = \sum_{n=0}^{N} b_n P_n(x), \tag{A12}
\]

Eq. \[A1\] becomes

\[
h(x) = (f_M * g_N)(x) = \int_{\min(1,x+1)}^{\max(-1,x-1)} dt \, f_M(t)g_N(x - t)
\]

\[
= \int_{-1}^{x+1} dt \, f_M(t)g_N(x - t) + \int_{x-1}^{1} dt \, f_M(t)g_N(x - t), \tag{A13}
\]

which can be computed separately in two integration domains \(x \in [-2, 0]\) and \(x \in [0, 2]\) (see Fig. 4.1 in Ref. \[63\].)
a. **First interval** \( x \in [-2, 0] \)

For \( x \in [-2, 0] \) we have \( h(x) = h^<(x) \) where

\[
h^<(x) = \int_{-2}^{x+1} dt f_M(t) g_N(x-t) = \sum_{k=0}^{m+n+1} \gamma_k P_k(x+1).
\]  

Using the orthogonality of Legendre polynomials Eq. \((A10)\), we have

\[
\gamma_k = \frac{2k+1}{2} \sum_{m=0}^{N} \alpha_m \left[ \int_{-2}^{0} dx P_k(x+1) \int_{-1}^{x+1} dt f_M(t) g_N(x-t) \right] + \frac{N}{2} \beta_n \sum_{m=0}^{M} \alpha_m \left[ \int_{-2}^{0} dx P_k(x+1) \int_{-1}^{x+1} dt P_m(t) P_n(x-t) \right]
\]  

collecting terms we can write \( \gamma_k = \sum_{n=0}^{N} B_{k,n}^\gamma \beta_n \) where

\[
B_{k,n}^\gamma = \frac{2k+1}{2} \sum_{m=0}^{M} \alpha_m \left[ \int_{-2}^{0} dx P_k(x+1) \int_{-1}^{x+1} dt P_m(t) P_n(x-t) \right] = \frac{2k+1}{2} \sum_{m=0}^{M} \alpha_m \int_{-1}^{1} ds P_k(s) (P_m * P_n)(s-1).
\]  

Applying the recursion relation of the spherical Bessel functions (Eq. \((A11)\)) on \( n \) and \( k \), we have

\[
(-i)^{m+n+1} i^k j_k(\omega) j_m(x)(\omega) = (-i)^{m+n+1} i^k j_k(\omega) j_m(\omega) \left( \frac{2n+1}{\omega} j_n(x) - j_{n-1}(x) \right)
\]  

\[
= \frac{2n+1}{2k+1} \left[ (-i)^{m+n+1} i^k (j_k(\omega) + j_{k-1}(\omega)) j_m(\omega) j_n(x) + (-i)^{m+n+1} i^k (j_k(\omega) j_m(\omega)) j_{n-1}(\omega) \right]
\]  

Back insert in Eq. \((A18)\) and simplifying prefactors in \( k \) gives

\[
B_{k,n+1}^\gamma = -\frac{2n+1}{2k+3} B_{k+1,n}^\gamma + \frac{2n+1}{2k-1} B_{k-1,n}^\gamma + B_{k,n-1}^\gamma
\]  

b. **Second interval** \( x \in [0, 2] \)

For \( x \in [0, 2] \) we have \( h(x) = h^>(x) \) where

\[
h^>(x) = \int_{x-1}^{1} dt f_M(t) g_N(x-t) = \sum_{k=0}^{m+n+1} \gamma_k^> P_k(x-1).
\]  

\( \gamma_k^> \) can be computed in the same way as \( \gamma_k^< \), see Eq. \((A15)\)

\[
\gamma_k^> = \frac{2k+1}{2} \sum_{m=0}^{N} \alpha_m \left[ \int_{-2}^{0} dx P_k(x-1) \int_{x-1}^{1} dt f_M(t) g_N(x-t) \right] = \frac{2k+1}{2} \sum_{m=0}^{N} \alpha_m \int_{-1}^{1} ds P_k(s) (P_m * P_n)(s-1).
\]  

collecting terms we can write \( \gamma_k^> = \sum_{n=0}^{N} B_{k,n}^\gamma \beta_n \) where

\[
B_{k,n}^\gamma = \frac{2k+1}{2} \sum_{m=0}^{M} \alpha_m \left[ \int_{-2}^{0} dx P_k(x-1) \int_{x-1}^{1} dt P_m(t) P_n(x-t) \right] = \frac{2k+1}{2} \sum_{m=0}^{M} \alpha_m \int_{-1}^{1} ds P_k(s) (P_m * P_n)(s+1).
\]  

Consider the \( B_{k,n+1}^< \) term, changing the order of integration and Fourier transforming the remaining Legendre polynomial gives

\[
B_{k,n+1}^< = \frac{2(2k+1)}{\pi} \sum_{m=0}^{M} (-i)^{m+n+1} i^k \alpha_m \int_{-\infty}^{\infty} d\omega j_k(\omega) j_m(\omega) j_{n+1}(\omega) e^{-i\omega}.
\]  

\[
B_{k,n}^> = \frac{2k+1}{\pi} \sum_{m=0}^{M} (-i)^{m+n} \alpha_m \int_{-\infty}^{\infty} d\omega e^{i\omega(s+1)} j_m(\omega) j_n(\omega)
\]
Since the exponent in the integral is unchanged when applying the recursion relations of the spherical Bessel functions we conclude that \( B^< \) obeys the same recursion relation as \( B^< \), albeit with a different starting point since the “seeding” integrals have a different sign in the exponent.

c. Summary

The convolution matrices for both the intervals can be expressed as the integral sums

\[
B_{k,n}^\leq = \frac{2(2k + 1)}{\pi} \sum_{m=0}^{M} (-i)^{m+n} i^k a_m \\
\times \int_{-\infty}^{\infty} d\omega \, j_k(\omega) j_m(\omega) j_n(\omega) e^{\pm i\omega} \quad (A25)
\]
differing only in the sign in the exponent. The coefficients are related by the recursion relation

\[
B_{k,n+1}^\leq = \frac{2n + 1}{2k + 3} B_{k+1,n}^\leq + \frac{2n + 1}{2k + 1} B_{k-1,n}^\leq + B_{k,n-1}^\leq \quad (A26)
\]

In practice this recursion relation is only stable below the diagonal with \( k > n \). To get entries above diagonal, the transpose relation, that can be derived from the integral expression Eq. [A18] is used

\[
B_{k,n}^\leq = (-1)^{n+k} \frac{2k + 1}{2n + 1} B_{n,k}^\leq \quad (A27)
\]

3. Initial values \( B_{k,0}^\leq \) and \( B_{k,1}^\leq \)

To start the recursion, the initial values for \( n = 0 \) and \( 1 \) are needed. To derive explicit expressions for these terms we repeatedly use the Volterra integral formula for Legendre polynomials from Ref. [11]

\[
S_{a,n}(x) = \int_0^x dt \, P_n(t) , \quad (A28)
\]

\[
S_{a,0}(x) = x - a , \quad (A29)
\]

\[
S_{a,n}(x) = \frac{1}{2n + 1} \left[ P_{n+1}(t) - P_{n-1}(t) \right]_a \quad (A30)
\]

for \( a = \pm 1 \) we get

\[
S_{\pm1,0}(x) = x \mp 1 = P_1(x) \mp P_0(x) , \quad (A31)
\]

\[
S_{\pm1,n}(x) = \frac{1}{2n + 1} \left[ P_{n+1}(x) - P_{n-1}(x) \right] , \quad (A32)
\]

where we have used \( P_n(\pm1) = (\pm1)^n \) to cancel the constant terms.

Returning to the convolution matrices we have for \( B_{k,n}^\leq \) and \( n = 0 \), using \( P_0(x) = 1 \),

\[
B_{k,0}^\leq = \pm \frac{2k + 1}{2} \sum_{m=0}^{M} \alpha_m \int_{-1}^{1} dx \, P_k(x) \int_{-1}^{x} dt \, P_m(t)
\]

\[
= \pm \frac{2k + 1}{2} \sum_{m=0}^{M} \alpha_m \int_{-1}^{1} dx \, P_k(x) S_{\mp1,m}(x)
\]

\[
= \pm \frac{2k + 1}{2} \sum_{m=0}^{M} \alpha_m \frac{2m + 1}{2m + 1} \int_{-1}^{1} dx \, P_k(x) \left[ P_{m+1}(x) - P_{m-1}(x) \right] \quad (A33)
\]

repeatedly using the Legendre orthogonality relation [Eq. (A6)] gives

\[
B_{k,0}^\leq = \begin{cases} 
\alpha_0 \pm \frac{\alpha_1}{3} , & \quad k = 0 \\
\pm \frac{\alpha_k}{2k + 1} + \frac{\alpha_{k+1}}{2k + 3} , & \quad k \geq 1 
\end{cases} \quad (A34)
\]

For the second column with \( n = 1 \) we detail the derivation of \( B_{k,1}^\leq \); the other case \( B_{k,1}^\leq \) can be done analogously. Using \( F_1(x) = x \) we get

\[
B_{k,1}^\leq = \frac{2k + 1}{2} \sum_{m=0}^{M} \alpha_m \\
\times \int_{-2}^{0} dx \, P_k(x + 1) \int_{-1}^{x+1} dt \, P_m(t) P_1(x-t)
\]

\[
= \frac{2k + 1}{2} \sum_{m=0}^{M} \alpha_m \\
\times \int_{-1}^{1} dx \, P_k(x) \int_{-1}^{x} dt \, P_m(t)(x-t-1)
\]

\[
= -B_{k,0}^\leq + \frac{2k + 1}{2} \sum_{m=0}^{M} \alpha_m \int_{-1}^{1} dx \, P_k(x) \int_{-1}^{x} dt \, P_m(t) \int_{t}^{x} ds
\]

\[
= -B_{k,0}^\leq + \frac{2k + 1}{2} \sum_{m=0}^{M} \alpha_m \int_{-1}^{1} dx \, P_k(x) \int_{-1}^{x} ds \int_{-1}^{s} dt \, P_m(t) \quad (A35)
\]

where the last step is obtained by changing the order of integration. The last integral relation is a double Volterra integral and can hence be written using \( S_{-1,m}(x) \) as

\[
B_{k,1}^\leq = -B_{k,0}^\leq + \frac{2k + 1}{2} \sum_{m=0}^{M} \alpha_m \int_{-1}^{1} dx \, P_k(x) \int_{-1}^{x} ds \, S_{-1,m}(s)
\]

\[
= -B_{k,0}^\leq + \frac{1}{2} \sum_{m=0}^{M} \alpha_m \int_{-1}^{1} dx \, [P_{k-1}(x) - P_{k+1}(x)] S_{-1,m}(x) \quad (A36)
\]
where we in the second step have used partial integration and the Legendre derivative relation, Eq. (A7).

For the second case \( B_{k,1} \), the only difference is when we change the integration variable, we get \((x - t + 1)\) instead of \((x - t - 1)\) in Eq. (A35), so the sign before \( B_{k,0} \) is changed to \(+1\). By using Eq. (A33) we obtain the recursion relation

\[
B_{k,1} = \mp B_{k,0} + \frac{B_{k-1,0}}{2k-1} - \frac{B_{k+1,0}}{2k+3}, \quad k \geq 1 \quad (A37)
\]

with the special case for \( k = 0 \), \( B_{0,1} = \mp B_{1,0}/3 \).
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