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On the optimal symmetric purification scheme of the one-particle density matrix

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ABSTRACT

Density matrix purification is an efficient way of avoiding the expensive cubic scaling diagonalization in self-consistent field calculations. Although there are a number of different algorithms suggested to reduce the number of matrix multiplications for purification, there is no rigorous mathematical proof which scheme is optimal. In this Letter, we show analytically that the repeated application of the fifth-order Holas polynomial throughout all iterations is an optimal scheme that reduces the error symmetrically for both occupied and virtual occupations, and either the use of lower/higher-order polynomials throughout or mixed use of polynomials of different degree at different iteration results in higher cost.

One-particle density matrix (**P**) is normally obtained by diagonalizing the Hamiltonian matrix (**H**) in self-consistent field (SCF) calculations. The cubic scaling of the matrix diagonalization, however, makes the conventional SCF calculations such as Hartree–Fock or density functional theory calculations difficult to treat large molecular systems. To address this diagonalization bottleneck in SCF, many methods [1–9] that scale linearly with system size (N) have been developed. Since there are only $O(N)$ number of significant matrix elements in **P** for large enough systems, direct minimization of the energy functional with respect to the density matrix elements led to a linear scaling algorithm [1].

Alternatively, one can form the Hamiltonian matrix, and from it computes the density matrix. In this, one essentially represents the density matrix as a Heaviside step function of the Hamiltonian matrix

$$\mathbf{P} = h(\mu\mathbf{I} - \mathbf{H}), \tag{1}$$

where $h(.)$ denotes Heaviside step function, **I** is a unit matrix, and μ is a chemical potential of the system. Eq. (1) can be computed by approximating the Heaviside step function as Fermi–Dirac [2] or complementary error functions [3] which can then be expanded and evaluated efficiently with the Chebyshev polynomials of the Hamiltonian matrix of finite order.

Instead of direct calculation of Eq. (1) using matrix polynomials of very high order, one can also determine **P** iteratively starting from an appropriately chosen initial guess for density matrix with the correct eigenfunctions. This latter category, the focus of the present Letter, is called the purification method [4] since the algorithm is designed to purify the initial density matrix to have the correct eigenvalues to 0s and 1s when converged. Both density matrix expansion methods or purification algorithms scale linearly with system size due to the locality and sparsity of **H** and **P** in the localized atomic orbital basis.

McWeeny's purification function [4] is the most widely used formula that purifies density matrix, where Eq. (2) is recursively used until the eigenvalues of **P** converge to 0s and 1s

$$\mathbf{P}_{n+1} = 3\mathbf{P}_n^2 - 2\mathbf{P}_n^3. \tag{2}$$

Although the original algorithm to use Eq. (2) for purification requires *a priori* knowledge of the chemical potential, Palser and Manolopolous [5] developed an algorithm that works without *a priori* knowledge of the chemical potential and only requires the total number of electrons, i.e., canonical purification. Other forms and degrees of polynomials have also been suggested to replace Eq. (2) for faster convergence and higher efficiency [6–9]. Kryachko [6] proposed a cubic polynomial that has a different form from that of McWeeny, and claimed that it shows a cubic convergence unlike the McWeeny's function that converges quadratically. The Kryachko's polynomial, however, later was correctly pointed out by Holas [7] that it only purifies the occupied occupation numbers. Holas [7] then suggested higher-order polynomials with more rapid convergence behavior than quadratic that purify both occupied and virtual occupation numbers. Niklasson [8] introduced a trace-correcting algorithm to switch between two different functions that purify only occupied ($\mathbf{P}_{n+1} = \mathbf{P}_n^2$ when the trace is too high) or virtual ($\mathbf{P}_{n+1} = 2\mathbf{P}_n - \mathbf{P}_n^2$ when the trace is too low) occupation numbers separately, namely, by using the latter asymmetric formulae selectively depending on the trace of the density matrix along the iteration. This use of asymmetric formulae was then extended to higher-order polynomials by Mazziotti [9] to enhance the convergence rate from quadratic to cubic. Despite the extra matrix multiplications needed to evaluate higher-order polynomials, it was concluded that the use of higher-order polynomials may overall be more efficient than lower-order purification functions due to a more rapid reduction of the error.

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In this Letter, we focus on purification functions that are symmetric in reducing the errors for the occupied and virtual occupation numbers equally, and derive an optimal purification scheme in terms of the degree and sequence of polynomials that are applied during iterations. If the polynomials used at all iterations are required to purify the density matrix, we show analytically that the repeated application of the fifth-order Holas polynomial throughout all iterations is an optimal algorithm, and either the use of lower/higher-order polynomials throughout or the mixed use of different-order polynomials at different iteration results in higher cost.

Theorem 1. *Let $\mathbf{P}_0 = \mathbf{P} + \epsilon$, where $\mathbf{P}$ is the exact density matrix and $\mathbf{P}_0$ is the initial density matrix. If $g_n(\mathbf{P}_0)$ denotes a polynomial that purifies $\mathbf{P}_0$ symmetrically for 0 and 1,*

$$\mathbf{P}_1 = g_n(\mathbf{P}_0) = \mathbf{P} + 0\ (\epsilon^n), \quad n \geqslant 2 \tag{3}$$

then its degree is at least $2n-1$ in general.

Proof. An exactly idempotent density matrix has eigenvalues 1s and 0s. If we take Taylor expansion of $g_n(\mathbf{P}+\epsilon)$ with respect to 1 and 0, the two stable points to be purified, four conditions must be met to purify the density matrix,

$$g_n(1) = 1, \tag{4a}$$
$$g_n(0) = 0, \tag{4b}$$
$$g_n^{(i)}(1) = 0, \tag{4c}$$
$$g_n^{(i)}(0) = 0, \tag{4d}$$

where $i = [1, n-1]$ and superscript (i) indicates the ith derivative. Every condition is linearly independent, and so to satisfy these four conditions, the polynomial should have at least $2n$ adjustable parameters. Therefore, the degree of polynomial is $2n-1$ at least. This completes the proof. □

For example, McWeeny's function is unique in that it is the only third-order polynomial of the same degree that can purify density matrix with an equal quadratic convergence for both occupied and virtual occupations, as also pointed out by many others [4–9]. By induction of the McWeeny's form, Holas gave explicit expressions for g_2, g_3 and g_4 [7], and later Mazziotti also derived the same formulae differently [9].

Theorem 2. *If the purification function is required to purify $\mathbf{P}$ at each iteration, the optimal purification scheme is when the polynomials at all iterations are of the same degree.*

Proof. We assume that the purification at each iteration is performed using different polynomials of different degrees, $2n_i - 1$, where i denotes the ith iteration. After m iterations, we then get

$$\mathbf{P}_m = \mathbf{P} + O\left(\epsilon^{\prod_{i=1}^{m} n_i}\right). \tag{5}$$

We stop the iteration if the exponent of the error $\prod_{i=1}^{m} n_i$ is close to k. In general, $2n-2$ matrix multiplications are needed to evaluate the $(2n-1)$th order general polynomial if the coefficients of *all* terms in each power of $\mathbf{P}$ are nonzero. For the Holas polynomial of degree $2n-1$, however, the coefficients of the 0th to $(n-1)$th terms are always zero to satisfy the four stability conditions in Theorem 1, and so not all $2n-2$ multiplications are needed. More precisely, the g_n, the polynomial of degree $2n-1$, takes the form $g_n = \mathbf{P}^n(a_0\mathbf{I} + a_1\mathbf{P} + a_2\mathbf{P}^2 + a_3\mathbf{P}^3 + \cdots + a_{n-1}\mathbf{P}^{n-1})$ where all $a_i (i = [0, n-1])$ are nonzero. To evaluate g_n, one thus needs to calculate all powers of $\mathbf{P}$ up to n and one extra matrix multiplication to compute the product, $\mathbf{P}^n$ times the term in parentheses, yielding the total n matrix multiplications. Therefore, the total computational cost (t) during all iterations is approximately proportional to

$$t(\{n_i\}) = \sum_{i=1}^{m} n_i \tag{6}$$

The objective is then to minimize t to achieve a target accuracy of ϵ^k with $k \sim \prod_{i=1}^{m} n_i$. To do so, we use the fact that the arithmetic mean is greater than or equal to the geometric mean

$$\frac{1}{m}\sum_{i=1}^{m} n_i \geqslant \sqrt[m]{\prod_{i=1}^{m} n_i}, \tag{7}$$

where the equality holds when all n_i have the same values. One can deduce

$$t(\{n_i\}) = \sum_{i=1}^{m} n_i \geqslant m\sqrt[m]{\prod_{i=1}^{m} n_i} \approx m\sqrt[m]{k}, \tag{8}$$

Therefore, the minimum in t occurs when all n_i are the same. This proves Theorem 2. In other words, applying the polynomials of the same order repeatedly is more efficient than combining polynomials of different degrees at different iterations. □

Theorem 3. *The fifth-order Holas polynomial is optimal if the purification function itself is required to purify a density matrix.*

We have shown in Theorem 2 that the application of polynomials of the same order at all iterations yields the optimal purification efficiency. If we use the same degree of polynomial n for all n_i, the total computational cost $t(\{n_i\}) = m\sqrt[m]{k} \approx \frac{\log(k)}{\log(n)} n$, which has a minimum at $n = 3$. Therefore, the optimal n is 3, yielding the fifth-order Holas polynomial, $g_3 = \mathbf{P}^3(10\mathbf{I} - 15\mathbf{P} + 6\mathbf{P}^2)$. This completes the proof for Theorem 3. Mazziotti suggested [9] that if the particle and hole density matrices are used and treated in a certain way to represent the symmetric purification functions, namely those of Holas [7], the number of matrix multiplications for higher-order polynomials can be reduced. For the latter case, the optimal degree of polynomial shown here to be five may change to higher-order. □

In summary, we have presented mathematical proofs that the repeated application of the same-order polynomial throughout all iterations is an optimal algorithm for the density matrix purification if the errors for the occupied and virtual occupation numbers are to be reduced equally and symmetrically, and any other combinations of polynomials at different iterations result in higher cost. We have shown that the fifth-order Holas polynomial, $g_3 = \mathbf{P}^3(10\mathbf{I} - 15\mathbf{P} + 6\mathbf{P}^2)$, is computationally optimal to purify the density matrix.

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