Properties and $N$-representability conditions of the two-body density matrix

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A summary is presented of the properties of the coefficient matrices formed by expanding the two-body reduced density matrix in a complete set of two-electron wave functions. Calculating the relationship between the many electron wave function and these so-called two-body density matrices enables the derivation of necessary N-representability conditions that ensure a given matrix obeys the postulates of many-body quantum mechanics.

I. INTRODUCTION

In 1955, Lowdin [1–3], and Mayer [4] presented similar methods to express the ground state energy of a many-body reduced density quantum system as a functional of the two-body reduced density matrix (2-RDM). Their articles inspired great interest in the possibility of the exact determination of many-electron quantum properties by an effective two-particle analysis [5]. As summarized by Mazzinoti [6], modern analysis of the 2-RDM usually proceeds along one of two paths: minimization of the energy as functional of the 2-RDM or the solution of contracted Schrodinger equations (CSE) [7, 8] that depend on higher order reduced densities. The principal difficulty in either approach is to sufficiently constrain the 2-RDM so that it is guaranteed to correspond to a true many-electron wave function. This problem of determining the so-called N-representability conditions has remained open since Coleman [9] coined the term in 1963.

While significant strides have been made to constrain the 2-RDM in the context of both CSE [10, 11] and direct minimization schemes [6, 12, 13], they come with the cost of increasingly sophisticated mathematical structure. The added complexity is unsurprising because, despite its name, the 2-RDM is a function of four positions including spin with no inherent matrix structure. However, the 2-RDM can be expanded in a two-electron general basis [14] to generate a true matrix representation.

Here we focus on such an expansion, first performed by Bopp [15, 16], and argue that the resulting two-body density matrix provides a conceptually simple foundation upon which we can construct solutions to the many body problem. A formula is derived to compute the matrix elements for an arbitrary many-electron state, allowing the compilation of a set of necessary N-representability conditions. Valid matrices are presented for a non-interacting Hamiltonian and remarks are made about a generalization to find solution of fully-interacting systems. The follow-up work, presented in Ref. [17], depends on an extension of the present work to a time-dependent analysis.

The paper is organized as follows. Section II introduces the two-body density matrix as the fundamental quantity used to compute the expectation value of a many body quantum operator. In section III we derive the N-representability conditions along with a formula for the conversion of a many-body wave function into a two-body density matrix. Finally in Section IV we derive the matrices for example states and propose a path toward computing fully-correlated eigenfunctions.

II. THE TWO-BODY DENSITY MATRIX

We now present the two-body density matrix as the basic quantity used to calculate the observable properties of a many-electron quantum system. We begin with the exchange-antisymmetric N-body wave function $\Psi(r_1 \sigma_1, \ldots, r_N \sigma_N)$, with $\sigma_i$ denoting the $i$-th spin coordinates. For simplicity, we combine spatial and spin degrees of freedom into the variable $x_i = r_i \sigma_i$.

Observables are represented by exchange-symmetric N-body linear operators which we assume to have one and two-body components

$$A(x_1, \ldots, x_N) = \sum_k A_1(x_k) + \sum_{i,j>i} A_2(x_i, x_j). \quad (1)$$

Eq. 1 contains a sum over coordinates for the one body contribution $A_1$ and another over electron pairs for the two-body component $A_2$. We may combine both into the sum over pairs

$$A(x_1, \ldots, x_N) = \sum_{i,j>i} A_1(x_i, x_j) + A_2(x_i, x_j) \quad (2)$$

by defining the effective one-body operator

$$A_1(x_i, x_j) = \frac{A_1(x_i) + A_1(x_j)}{N-1}. \quad (3)$$

The equivalence of Eqs. 1 and 2 is apparent by noting the pair sum in Eq. 3 overcounts each one-body term by a factor of $N-1$.
We proceed to compute the expectation value of $A$ by the usual formula

$$
\langle A \rangle = \sum_{i<j=1}^{N} \int \Psi^*(x_1, x_2, \ldots, x_N) \times A(x_i, x_j) \Psi(x_1, x_2, \ldots, x_N) \prod_{i=1}^{N} dx_i,
$$

where the differentials $dx_i$ are understood to include a sum over spin coordinates. The integrals for each pair $(i, j)$ in the sum are found to be equivalent by the following procedure: swap the names $x_i \leftrightarrow x_1$ and $x_j \leftrightarrow x_2$ then permute the same pairs within the argument lists of $\Psi$ and $\Psi^*$. The even number of permutations leaves the total integral unchanged, yielding the final result

$$
\langle A \rangle = \left( \frac{N}{2} \right) \int \Psi^*(X, Y) A(X) \Psi(X, Y) dX dY.
$$

(5)

We have introduced in Eq. 5 the shorthand notation $X = x_1, x_2$ and $Y = x_3, \ldots, x_N$. The symbols $dX$ and $dY$ are similarly defined as the product of differentials for each set of coordinates in the corresponding list. The prefactor, equal to $N(N-1)/2$, is the number of terms in the pair sum. The operator $A(X)$ is

$$
A(X) = A_1(X) + A_2(X) = \frac{A_1(x_1) + A_2(x_2)}{N-1} + A_2(X),
$$

(6)

with $A_1(X)$ re-scaled as in Eq. 3.

Next, remove $A(X)$ from the integral over $Y$ by introducing a set of primed coordinates $X'$ upon which the operator $A(X')$ acts. This permits the re-formulation of Eq. 5 as

$$
\langle A \rangle = \left( \frac{N}{2} \right) \int dX dX' \delta(X' - X) \times A(X') \int \Psi^*(X, Y) \Psi(X', Y) dY.
$$

(7)

The term $\delta(X - X')$, understood to include a discrete delta function for spin degrees of freedom, ensures that primed and unprimed values are taken to be equal after applying the operator to the wave function.

We simplify Eq. 7 by defining the 2-RDM

$$
\rho(X, X') = \left( \frac{N}{2} \right) \int \Psi^*(X, Y) \Psi(X', Y) dY,
$$

(8)

so that the expectation value is computed as

$$
\langle A \rangle = \int dX dX' \delta(X' - X) A(X') \rho(X, X').
$$

(9)

Next, express the 2-RDM as the position representation of a non-local two-body linear operator $\hat{D}$:

$$
\rho(X, X') = \langle X' | \hat{D} | X \rangle.
$$

(10)

Introducing a complete set of two-electron basis functions $|i\rangle$ with $\psi_i(X) = \langle X|i\rangle$, we expand $\hat{D}$ by inserting two resolutions of the identity $1 = \sum_i |i\rangle \langle i|$ to find

$$
\hat{D} = \sum_{mn} |m\rangle \langle m| \hat{D}|n\rangle \langle n|.
$$

(11)

Defining $D_{mn} = \langle m| \hat{D}|n\rangle$ and taking the position representation by pre-multiplying $|X\rangle$ and post-multiplying $|X\rangle$, the two-body density as expressed in Eq. 10 takes the form

$$
\rho(X, X') = \sum_{mn} D_{mn} \psi_n^*(X) \psi_m(X').
$$

(12)

Inserting Eq. 12 into Eq. 9 yields

$$
\langle A \rangle = \sum_{mn} D_{mn} \int \psi_n^*(X) A(X) \psi_m(X) dX.
$$

(13)

Defining the matrix $D$ with coefficients $D_{mn}$ and $A$ with coefficients $A_{nm}$ given by the integral in Eq. 13, we finally discover the simple relation

$$
\langle A \rangle = \text{Tr}[DA].
$$

(14)

Thus, we can calculate the expectation value of an arbitrary many body linear operator provided we know the coefficient matrix in some orthonormal basis. Since all measurable properties of a system correspond to such an operator, the two-body density matrix $D$ contains all relevant information of the $N$-electron wave function.

### III. MATRIX CONSTRAINTS

Having demonstrated the utility of the two-body density matrix $D$, we shift focus to imposing constraints on its form. These $N$-representability conditions will serve to ensure that there exists a valid $N$-electron wave function that is represented by a given density matrix. The simplest such rule arises from a symmetry of Eq. 8 wherein $\rho(X', X) = \rho^*(X, X')$. Applying this transformation directly to the expansion of Eq. 12, we see that

$$
\sum_{mn} D_{mn} \psi_n^*(X) \psi_m(X) = \sum_{mn} D_{mn}^* \psi_n(X) \psi_m^*(X').
$$

After swapping the sum index labels on the right hand side, we find the symmetry to be satisfied when $D_{mn} = D_{nm}$. This relationship implies the matrix identity

$$
D^\dagger = D.
$$

(15)

We uncover another $N$-representability requirement by forcing $X' = X$ in Eq. 8 and integrating both sides over the remaining free coordinates $X$. The integral on the right-hand side reduces to unity by the normalization condition of the wave function. Choosing a convenient representation for the left hand side gives

$$
\int \delta(X - X') \rho(X, X') dX dX' = \left( \frac{N}{2} \right).
$$

(16)
which we note by comparison with Eq. 14 is the expectation value of the identity operator 1. From Eq. 14 it is therefore evident that

$$\text{Tr} [D] = \frac{N}{2}. \quad (17)$$

We also require antisymmetry under exchange $x_1 \leftrightarrow x_2$ (or $x'_1 \leftrightarrow x'_2$), so that $\rho(x_2, x_1, X') = -\rho(X, X')$. Swapping these coordinates in the two-body expansion of Eq. 12 reveals

$$\rho(x_2, x_1, X') = \sum_{mn} D_{mn} \psi_m(x_2, x_1) \psi_m(X') = -\rho(X, X'), \quad (18)$$

indicating that the property is inherited from the anti-symmetry of the basis functions and does not further restrict $D$.

Unfortunately, at this point we have extracted all the information contained in Eq. 8 alone. To proceed examining the $N$-representability problem we must choose an explicit basis and derive expressions for the matrix elements $D_{mn}$. We do so by pre-multiplying Eq. 12 by $\psi_n(x) \psi^*_m(x')$ and integrating over $dX$ and $dX'$ to find

$$D_{mn} = \int \psi_n(x) \psi^*_m(x') \rho(x, x') dX dX'. \quad (19)$$

Subsequently substituting Eq. 8 into Eq. 19 yields the simple equation

$$D_{mn} = \int \Theta^*_n(Y) \Theta_m(Y) dY, \quad (20)$$

with overlap functions $\Theta_m(Y)$ defined to be

$$\Theta_m(Y) = \int \psi^*_m(X) \psi(X, Y) dX. \quad (21)$$

We choose the basis functions $\psi_{ij}(X)$ to be those formed by the anti-symmetrized product of two single particle spinors. Grouping the two index labels into the tuple $n = \{n_1, n_2\}$, the basis functions take the form

$$\psi_n(X) = \frac{1}{\sqrt{2}} [\chi_{n_1}(x_1) \chi_{n_2}(x_2) - \chi_{n_2}(x_1) \chi_{n_1}(x_2)]. \quad (22)$$

We define a configuration $\{\alpha\}$ for the overlap $\Theta$ is a possibly infinite linear superposition of $N$-body Slater determinants

$$\Psi(X, Y) = \sum_{\{\alpha\}} C_{\{\alpha\}} \Psi_{\{\alpha\}}(X, Y). \quad (23)$$

In Eq. 23 we defined a configuration $\{\alpha\}$ to be an ordered list of single-particle spinors in a given product of states. The properly anti-symmetrized wave function of a single configuration has the determinant form

$$\Psi_{\{\alpha\}}(X, Y) = \frac{1}{\sqrt{N!}} \begin{vmatrix} \chi_{\alpha_1}(x_1) & \chi_{\alpha_2}(x_1) & \ldots & \chi_{\alpha_N}(x_1) \\ \chi_{\alpha_1}(x_2) & \chi_{\alpha_2}(x_2) & \ldots & \chi_{\alpha_N}(x_2) \\ \vdots & \vdots & \ddots & \vdots \\ \chi_{\alpha_1}(x_N) & \chi_{\alpha_2}(x_N) & \ldots & \chi_{\alpha_N}(x_N) \end{vmatrix}. \quad (24)$$

We now expand each $\Psi_{\{\alpha\}}(X)$ in Eq. 23 along minors of the top two rows of Eq. 24 to isolate the $x_1$ and $x_2$ dependence. The end result is.

$$\Psi(X, Y) = \frac{1}{\sqrt{N(N-1)}} \sum_{\{\alpha\}} C_{\{\alpha\}} \sum_{i,j>i} (-1)^{i+j-1} \left[\chi_{\alpha_i}(x_1)\chi_{\alpha_j}(x_2) - \chi_{\alpha_j}(x_1)\chi_{\alpha_i}(x_2) \right] \Psi_{\{\alpha\}_{ij}}(Y), \quad (25)$$

where the reduced configuration $\{\alpha\}_{ij} = \{\alpha\} \setminus \{\alpha_i, \alpha_j\}$ is the set subtraction of $\alpha_i$ and $\alpha_j$ from the original list of states. The state $\Psi_{\{\alpha\}_{ij}}(Y)$ is the determinant of the matrix formed by removing rows 1 and 2 and columns $i$ and $j$ from Eq. 24. As the normalization of this $N - 2$ electron state requires the prefactor $1/\sqrt{(N-2)!}$, we multiplied by its inverse which partially canceled with the $1/\sqrt{N!}$ prefactor.

Continuing to normalize the $x_1$ and $x_2$ dependence into a two-electron Slater determinant $\psi_{\alpha_i, \alpha_j}(X)$, we finally have

$$\Psi(X, Y) = \sqrt{\frac{2}{N(N-1)}} \sum_{\{\alpha\}} C_{\{\alpha\}} \sum_{i,j>i} (-1)^{i+j-1} \psi_{\alpha_i, \alpha_j}(X) \Psi_{\{\alpha\}_{ij}}(Y). \quad (26)$$

Plugging Eq. 26 into Eq. 21 for the overlap $\Theta_m(Y)$, the integration over $dX$ reduces the two-electron wave functions to $\delta_{\alpha_i, m_1} \delta_{\alpha_j, m_2}$ by orthonormality. Thus, a given configuration that does not contain $m = \{m_1, m_2\}$ will not contribute to $D_{mn}$. Consequently, we may reduce the $\{\alpha\}$ (configuration) sum into one over $\{\alpha\} \ni$
\{m_1, m_2\}$. The remaining sum is reduced to the single term with $(i, j) = \{m_1, m_2\}$ so that

$$\Theta_m(Y) = \sqrt{\frac{2}{N(N-1)}} \sum_{\{\alpha\} \ni m} C_{\{\alpha\}} S_\alpha[m] |\Psi_{\{\alpha\}} m(Y)\rangle,$$

(27)

where $\{\alpha\}_m = \{\alpha\}_{m_1, m_2} = \{\alpha\} \setminus \{m_1, m_2\}$. The symbol $S_\alpha[m]$ is the sign function

$$S_\alpha[m] = (-1)^{I_{\alpha}[m_1]+I_{\alpha}[m_2]-1}$$

(28)

with $I_{\alpha}[p]$ the index of basis function $p$ in configuration $\{\alpha\}$. We absorb this sign into the expansion coefficient by defining $C_{\{\alpha\}} = C_{\{\alpha\}} S_\alpha[m]$.

Finally, using that $\Theta_n^* (Y)$ is the complex conjugate of Eq. 27, we compute $D_{mn}$ by Eq. 20:

$$D_{mn} = \sum_{\{\alpha\} \ni n} C_{\{\beta\}}^* C_{\{\alpha\}} \int \Psi_{\{\beta\}} n (Y) \Psi_{\{\alpha\}} m (Y) dY.$$  

(29)

The integral, being the inner product between orthonormal $N-2$ electron Slater determinants, equals one when $\{\alpha\}_n = \{\beta\}_m$ and zero otherwise. Therefore,

$$D_{mn} = \sum_{\{\alpha\} \ni n} C_{\{\beta\}}^* C_{\{\alpha\}} \delta_{\{\alpha\}_n, \{\beta\}_m}.$$  

(30)

The diagonal matrix elements are then found from Eq. 30 to take the simple form

$$D_{nn} = \sum_{\{\alpha\} \ni n} |C_{\{\alpha\}}|^2.$$  

(31)

Because Eq. 31 is a sum over the magnitude squared of all expansion coefficients of configurations containing $n$, the overall normalization condition $\sum_{\{\alpha\}} |C_{\{\alpha\}}|^2 = 1$ implies that

$$0 \leq D_{nn} \leq 1.$$  

(32)

The maximum diagonal value, $D_{nn} = 1$, occurs when every configuration $\{\alpha\}$ contains $n$. In this case we encounter the additional rule that

$$D_{mn} = 1 \implies \forall m \neq n, D_{mn} = D_{nm} = 0,$$  

(33)

meaning that 1 on the diagonal in position $n$ forces all other elements in the column and row $n$ to zero.

The proof of Eq. 33 proceeds as follows. Per Eq. 31, any non-zero term must simultaneously satisfy the conditions $\{\alpha\} \ni \{m_1, m_2\}$, $\{\beta\} \ni \{n_1, n_2\}$ and $\{\alpha\} \setminus \{m_1, m_2\} = \{\beta\} \setminus \{n_1, n_2\}$. By this equality, $\{\alpha\} \setminus \{m_1, m_2\}$ does not contain $\{n_1, n_2\}$. Since $\{\alpha\}$ is formed by the set addition of some $\{m_1, m_2\} \neq \{n_1, n_2\}$, we have that $\{\alpha\} \neq \{n_1, n_2\}$. Supposing now that $D_{mn} \neq 0$ implies existence of some $\{\alpha\}$ in the state expansion that does not contain $\{n_1, n_2\}$. This contradicts the requirement that must be met for $D_{nn} = 1$ so we conclude that the existence of 1 on a diagonal implies all other elements in that row and column are 0.

By the Hermiticity (Eq. 15) of $D$, it can always be transformed into diagonal form by a unitary basis transformation (see Appendix A). The resulting diagonal matrix obeying Eq. 32 has the property

$$0 \leq \text{Tr}[D^2] \leq \left(\frac{N}{2}\right),$$  

(34)

which follows trivially from the fact that $a^2 \leq a$ for a number $a \leq 1$. $\text{Tr}[D^2]$ is a basis-independent quantity as it is invariant under unitary transformation by the cyclic property of the trace.

We finally summarize the necessary $N$-representability constraints on the matrix $D$:

$$D = D^\dagger$$  

(35a)

$$0 \leq D_{nn} \leq 1$$  

(35b)

$$\text{Tr}[D] = \left(\frac{N}{2}\right)$$  

(35c)

$$0 \leq \text{Tr}[D^2] \leq \left(\frac{N}{2}\right).$$  

(35d)

Condition (35a) expresses the Hermiticity of the two-body density matrix. We may interpret Rules (35b) and (35c) as the maximum occupation number for a given two-electron state and the total number of electron pairs present in a system, respectively. The final expression (35d) provides a simple way to distinguish between states which are not connected by some unitary transformation.

While the constraints in Eq. 35 are necessary, they are not sufficiently restrictive to ensure a given matrix is $N$-representable. After providing some concrete examples of valid matrices, we are able to propose a path toward the calculation of two-body density matrices representing the ground state of highly correlated systems.

### IV. MATRICES REPRESENTING SLATER DETERMINANTS

We begin this section by studying the density matrix in a basis comprising states which diagonalize an effective two-body non-interacting Hamiltonian. We discover an expression for the simplest matrices, those which correspond to a single antisymmetrized product state. We continue to address the interacting case in the second subsection; while the solution of this problem is outside the scope of this work, we uncover hints that it may be solved by a more thorough analysis.
A. Non-interacting Hamiltonian

As discussed in Section III, it is necessary to specify an orthonormal basis to define an explicit form for $D$. Here, we choose the set of wave functions that satisfy the eigenvalue relation $H(X)\psi_n(X) = E_n\psi_n(X)$ for a Hamiltonian $H(X) = H_1(X)$ containing only one-body terms.

A matrix composed entirely of $N(N-1)/2$ ones along the diagonal satisfies the necessary conditions in Eq. 35. These states have the special properties

\[
\text{Tr}[D^2] = \left(\frac{N}{2}\right) \quad (36a)
\]

\[
D^2 = D, \quad (36b)
\]

with the second line representing that $D$ is an idempotent projection.

Since $D_{nn} = 1$ implies that each configuration contains $n = \{n_1, n_2\}$, the wave function expansion must contain all represented pair in every configuration. This requirement is only possible for a single configuration, meaning the proposed matrix corresponds to a Slater determinant. However, as we now begin to show, the occupied diagonals may not be chosen arbitrarily due to the paired nature of the basis.

It was mentioned in Section III that the tuples $n$ must be flattened to an ordered list indexed by a single integer to exploit the matrix properties of $D$. Here we provide an example of this ordering and observe the effects on the two-body density matrix.

We select 15 example functions from the basis of two-electron spinors that diagonalize the non-interacting Hamiltonian

\[
\begin{align*}
(1 \uparrow, 1 \downarrow) \\
(1 \uparrow, 2 \uparrow), (1 \uparrow, 2 \downarrow), (1 \downarrow, 2 \uparrow), (1 \downarrow, 2 \downarrow) \\
(2 \uparrow, 2 \downarrow) \\
(1 \uparrow, 3 \uparrow), (1 \uparrow, 3 \downarrow), (1 \downarrow, 3 \uparrow), (1 \downarrow, 3 \downarrow) \\
(2 \uparrow, 3 \uparrow), (2 \uparrow, 3 \downarrow), (2 \downarrow, 3 \uparrow), (2 \downarrow, 3 \downarrow) \\
(3 \uparrow, 3 \downarrow), \\
\end{align*}
\]

labeled by an energy-ordered principal quantum number and an arrow representing the spin. We take the basis functions in Eq. 37 to be numbered 1 to 15 in the order written and construct valid $N$-body states for various values of $N$.

1. Two electrons

This case is trivial because the basis functions are already two-electron wave functions; a general two-electron state may be represented by $D_{nn} = 1$ for any integer $n$.

2. Three electrons

We know that a Slater determinant state will be an antisymmetrized product $\hat{S}_-\chi_{\alpha_1}(x_1)\chi_{\alpha_2}(x_2)\chi_{\alpha_3}(x_3)$ (with $\hat{S}_-$ the determinant operator of Eq. 24) for some configuration $\alpha = \{\alpha_1, \alpha_2, \alpha_3\}$. Thus, a valid matrix must represent a wave function that contains 3 single particle eigenstates. Some care must be taken to satisfy this requirement because the basis is labeled by two single-particle spinors. For example, we immediately notice that $D_{nn} = 1$ for $n \in \{1, 2, 3\}$ is invalid as it involves four single particle basis functions: $|1 \uparrow>, |1 \downarrow>, |2 \uparrow>, \text{ and } |2 \downarrow>$.

On the other hand, matrices with $D_{nn} = 1$ are valid for $n$ belonging to the sets $\{1, 2, 4\}$, $\{1, 3, 5\}$, $\{1, 2, 4\}$, $\{1, 7, 9\}$, $\{1, 8, 10\}$, $\{6, 11, 13\}$, $\{6, 12, 14\}$. Each of these examples is seen to arise from a three-particle Slater determinant.

3. 4+ electrons

The matrix representing a 4-electron determinant state must have 6 ones along the diagonal. The simple choice of $D_{nn} = 1$ for $n \leq 6$ is valid as it contains exactly four one-particle basis functions. For completeness, we will also calculate the total energy of this state. From Eq. 14

\[
E = \text{Tr}[DH] = \sum_n D_{nn}E_n, \quad (38)
\]

where $E_n = (E_{n_1} + E_{n_3})/3$ by Eq. 13 and the one-body expression of Eq. 6. Adding terms, we find that $E = E_1 + E_2 + E_3 + E_4$ as expected. As the number of electrons increases, the principles remain the same but it becomes more cumbersome to build valid states.

4. Example of a non-determinant state

We can also construct matrices for states that are not described by a single determinant. We choose for example an $N$-electron state given by the constant superposition of $M$ disjoint antisymmetrized configurations. That is,

\[
\Psi(X) = \frac{1}{\sqrt{M}} \hat{S}_- \left[ \left( \chi_1(x_1) \cdots \chi_N(x_N) \right) + \left( \chi_{N+1}(x_1) \cdots \chi_{2N}(x_N) \right) + \cdots + \left( \chi_{M-1}(x_1) \cdots \chi_{MN}(x_N) \right) \right]. \quad (39)
\]

By Eq. 30, all off-diagonal matrix elements are zero because no two configurations share $N - 2$ basis functions. The diagonal terms are similarly simple: each pair contained in the expansion occurs in a single configuration with coefficient $1/\sqrt{M}$ so that the diagonal elements have $D_{nn} = 1/M$ by Eq. 31.
It is straightforward to show that $\text{Tr}[D^2] = N(N - 1)/(2M^2)$, which decreases as the number $M$ of disjoint configurations increases. Comparing to the squared trace of Eq. 36a, we find that this property serves as a measure of the amount of non-unitary transformation needed to evolve from a state with maximal squared trace.

### B. Interacting Hamiltonian

The effective two-particle Hamiltonian with Coulomb interaction is given in Hartree atomic units ($\hbar = |e| = m_0 = 1/(4\pi\epsilon_0) = 1$) by

$$H(X) = H_1(X) + \frac{1}{|r_1 - r_2|}$$

and gives rise to its own set of orthonormal eigenvectors $\psi_n(X)$. If we were to transform the previously-discussed Slater determinant states into the basis which diagonalizes Eq. 40, the resulting matrix would no longer be diagonal.

The important question that remains is: Is there some way to transform the matrix $D$ itself the lowest energy valid state while remaining in the fully-correlated basis? First, we note that a change in $D$ with the basis fixed corresponds to an evolution of the state. This evolution, which we can take to be that in time, may proceed by the influence of some time-dependent Hamiltonian term. If temporal evolution is dictated by a unitary transformation, then we furthermore would have that $\text{Tr}[D^2] = N(N - 1)/2$. This simplifies the problem immensely because it implies that the ground state may be written as a diagonal matrix with coefficients of only 1 or 0.

### V. CONCLUSIONS

We have summarized the most important properties of the two-body density matrix that contains all relevant information of the many-body wave function. As usual for 2-RDM-based analysis, ensuring $N$-representability serves as the major roadblock in the practicality of the formalism. A set of four necessary conditions were presented along with a formula for the diagonal and off-diagonal elements of the two-body density matrix.

The approach presented here is remarkable due to its simplicity; the 2-RDM can be analyzed strictly by matrix operations without requiring higher order tensors. Examples of valid $N$-representable states have been presented along with hints that fully-correlated ground states may be determined by unitary evolution from these most simple examples. This idea is explored in Ref. [17].

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### Appendix A: Change of basis

The formula for a change of basis is identical to that for any density matrix, but we re-derive it here for clarity. We begin with the two-body density matrix

$$D = \sum_{mn} D_{mn} \psi_n^*(X)\psi_m(X)$$

and introduce a new orthonormal basis with wave functions $\phi_i(X)$. Expanding the initial states $\psi_n(X)$ in terms of the new, we find

$$D = \sum_{ij} \left( \sum_{mn} U_{jm}^\dagger \phi_j^*(X) \right) \left( \sum_{im} U_{im} \phi_i(X') \right)$$

$$= \sum_{ij} \left( \sum_{mn} U_{im} D_{mn} U_{jm}^\dagger \right) \phi_j^*(X)\phi_i(X').$$

The parenthetical term gives the expression for $D_{ij}$, which we take to be the coefficients of matrix $D'$. We finally find the matrix form for the change of basis

$$D' = UDU^\dagger,$$

where $U$ is the unitary matrix of coefficients $U_{ij}$.

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