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Contraction relations for Grassmann products of reduced density matrices and implications for density matrix reconstruction

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We consider, for systems of indistinguishable fermions, approximate reconstruction of the three- and four-particle reduced density matrices (RDMs) from the one- and two-particle RDMs, $\boldsymbol{\gamma}$ and $\boldsymbol{\Gamma}$. Our ansatz for reconstructing the four-particle RDM is the linear combination $a(\boldsymbol{\Gamma}\wedge\boldsymbol{\Gamma})+b(\boldsymbol{\gamma}\wedge\boldsymbol{\gamma}\wedge\boldsymbol{\Gamma})+c(\boldsymbol{\gamma}\wedge\boldsymbol{\gamma}\wedge\boldsymbol{\gamma}\wedge\boldsymbol{\gamma})$, where "$\wedge$" denotes the antisymmetrized (Grassmann) product. This is a generalization of reconstruction functionals employed recently to perform direct RDM calculations without wave functions via the contracted Schrödinger equation. Here we consider relationships between the parameters a, b, and c that are required in order for the reconstruction functionals to respect the hierarchy of contraction relations between RDMs. To this end we establish several general theorems concerning contractions of antisymmetrized tensor products of $\boldsymbol{\gamma}$, $\boldsymbol{\Gamma}$, and various products thereof. The accuracy of proposed reconstruction functionals is evaluated using accurate density matrices for the ground state of Be.

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I. INTRODUCTION

"Direct" density matrix methods, in which the two-electron reduced density matrix (2-RDM or 2-matrix) is calculated without recourse to an electronic wave function, are hampered by an incomplete characterization of the set of N-representable RDMs [1,2]. Recently, several methods that employ approximate N-representability constraints have achieved some success. Variational calculation of the 2-matrix, constrained by several known N-representability requirements [3–9], has been carried out recently for atoms and for molecules containing no more than one heavy atom [10]. At the same time, several groups [11–20] have developed an alternative scheme for direct calculation of the 2-RDM based upon the contracted Schrödinger equation (CSE) [21–23]. In these latter methods, approximate N-representability constraints are imposed in the form of *reconstruction functionals*—prescriptions for building higher-order RDMs from lower-order ones.

The CSE involves the two-, three-, and four-electron RDMs and the two-electron reduced Hamiltonian. It is equivalent (in a necessary and sufficient sense) to the electronic Schrödinger equation but does not involve the wave function explicitly. The shadow of the wave function is manifest, however, in N-representability requirements [1–9] that RDMs must satisfy if they are to derive from physically admissible N-electron states. These requirements are the density-matrix formulation of the Pauli principle, and in their absence the CSE possesses spurious, unphysical solutions for the 2-RDM [24]. At least for nondegenerate ground states, however, the N-representable 2-RDMs map one-to-one onto the N-representable p-RDMs for each $p\geqslant 2$ [25,26]. This (unknown) map is precisely the reconstruction functional for the p-RDM, and any approximation for this functional serves as an approximate N-representability requirement in an iterative, self-consistent solution of the CSE.

In this paper we examine a particular ansatz for the 4-RDM reconstruction functional and compare it with previously suggested reconstructions. We consider a reconstructed 4-RDM formed from antisymmetrized (Grassmann) products of the 1-RDM $\boldsymbol{\gamma}$ and the 2-RDM $\boldsymbol{\Gamma}$,

$$\boldsymbol{\Gamma}_{\mathrm{G}}^{(4)}[\boldsymbol{\Gamma}] \overset{\mathrm{def}}{=} a(\boldsymbol{\Gamma}\wedge\boldsymbol{\Gamma})+b(\boldsymbol{\Gamma}\wedge\boldsymbol{\gamma}\wedge\boldsymbol{\gamma})+c(\boldsymbol{\gamma}\wedge\boldsymbol{\gamma}\wedge\boldsymbol{\gamma}\wedge\boldsymbol{\gamma}). \tag{1.1}$$

Here $\overset{\mathrm{def}}{=}$ indicates a definition, a, b, and c are parameters, and "$\wedge$" denotes the Grassmann product [19,27], for which a precise definition is given in Sec. III. This reconstruction ansatz is a generalized, density-matrix analog of decoupling approximations introduced in order to solve the hierarchy of Green's function equations of motion, and each such decoupling scheme is equivalent to a partial summation of the perturbation expansion for the relevant Green's function [28]. However, unlike certain decoupling strategies (such as the random-phase approximation) employed frequently in condensed-matter physics, the reconstruction scheme of Eq. (1.1) is necessarily antisymmetric. We consider the antisymmetry requirement to be of paramount importance in electronic structure.

Reconstruction of the 3-RDM $\boldsymbol{\Gamma}^{(3)}$ may proceed through a separate functional, or $\boldsymbol{\Gamma}^{(3)}$ may be obtained as the one-particle contraction (trace over the coordinates of one particle) of the reconstructed 4-matrix, as in the algorithms of Valdemoro and co-workers [12–14] and Mazziotti [19,20]. As such, there exist several plausible iteration schemes that might be used to achieve a self-consistent solution of the CSE; three such schemes are discussed in this paper.

Given any approximate reconstruction functional $\boldsymbol{\Gamma}^{(4)}[\boldsymbol{\Gamma}]$, there is always the question of whether it is *contraction-consistent*. That is, if we trace over the coordinates of two particles in $\boldsymbol{\Gamma}^{(4)}[\boldsymbol{\Gamma}]$, do we recover the input $\boldsymbol{\Gamma}$ used to generate this matrix? This question is especially germane if one's iteration scheme employs the contractions of $\boldsymbol{\Gamma}^{(4)}[\boldsymbol{\Gamma}]$.

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Within our Grassmann product ansatz, the contraction-consistency requirement leads to a set of relations between *a*, *b*, and *c*, which we derive. Furthermore, we demonstrate that neither the two- nor three-particle contraction of $\boldsymbol{\Gamma}_{\mathrm{G}}^{(4)}$ can be written in a simple Grassmann product form; for example, the one-particle contraction of $\boldsymbol{\Gamma}_{\mathrm{G}}^{(4)}$ cannot be expressed as a linear combination of $\boldsymbol{\gamma}\wedge\boldsymbol{\Gamma}$ and $\boldsymbol{\gamma}\wedge\boldsymbol{\gamma}\wedge\boldsymbol{\gamma}$. It is therefore inconsistent to assume a Grassmann product form for both $\boldsymbol{\Gamma}^{(3)}$ and $\boldsymbol{\Gamma}^{(4)}$. This conclusion is implicit in recent work by Mazziotti [29], although he does not provide an explicit form for the contractions of Grassmann product functionals.

One note concerning semantics is in order. The matrix $\boldsymbol{\Gamma}_{\mathrm{G}}^{(4)}$ is, by construction, antisymmetric and self-adjoint. There is no reason, however, to expect that it is positive in general. (Indeed, a recurring theme in the work of Mazziotti [19,20,30] has been solution of the CSE supplemented with positivity constraints for the reconstructed 4-RDM.) As such it is disingenuous to refer to $\boldsymbol{\Gamma}_{\mathrm{G}}^{(4)}$ as a density matrix or 4-RDM. We shall refer to $\boldsymbol{\Gamma}_{\mathrm{G}}^{(4)}$ (or any four-particle matrix obtained as a functional of $\boldsymbol{\Gamma}$) as a *reconstructed* 4-matrix, with similar language for 3-matrices. Implicit in this idiom is the understanding that the reconstruction is not exact, so $\boldsymbol{\Gamma}_{\mathrm{G}}^{(4)}$ is not *a priori* positive. Other than the work of Mazziotti cited above, positivity within the reconstruction process has been handled numerically (by equating to zero any negative diagonal elements [11,14,31]) but not analytically in the formal derivation of reconstruction functionals.

The remainder of this article is structured as follows. In Sec. II we present a unified discussion of a class of ‘‘reduced eigenvalue equations,’’ of which the CSE is the most important example. We formulate these both as integral equations (in which the RDMs are integral kernels) and as matrix equations in a spin-orbital basis. In Sec. III we discuss the proposed reconstruction functionals of Valdemoro [32,31] and Nakatsuji [15–17], as well as three possible iterative schemes for solving the CSE. Section IV consists of several general theorems concerning the contraction of antisymmetrized products of one- and two-particle density matrices for fermions, and in Sec. V we apply these theorems to $\boldsymbol{\Gamma}_{\mathrm{G}}^{(4)}$ to obtain the 1-, 2-, and 3-RDMs to which it corresponds. In Sec. VI we employ an accurate 2-RDM for the ground state of Be to compare the reconstructed density matrices obtained from $\boldsymbol{\Gamma}_{\mathrm{G}}^{(4)}$, for several sets of parameters *a*, *b*, and *c*.

II. REDUCED EIGENVALUE EQUATIONS

Given a wave function $\Psi(1,2,\ldots,N)$ for a system of N indistinguishable fermions, the pth-order reduced density matrix (p-RDM or p-matrix) $\boldsymbol{\Gamma}^{(p)}$ for the state described by Ψ is the integral kernel

$$\begin{aligned}\Gamma^{(p)}(1,\ldots,p;1',\ldots,p') &\overset{\mathrm{def}}{=} \binom{N}{p}\int \Psi(1,\ldots,N)\Psi^{*}(1',\ldots,p',p+1,\ldots,N)\,d\mathbf{x}_{p+1}\cdots d\mathbf{x}_{N}\\ &= \sum_{\substack{i_1,\ldots,i_p\\ j_1,\ldots,j_p}} (\boldsymbol{\Gamma}^{(p)})^{i_1,\ldots,i_p}_{j_1,\ldots,j_p}\,\phi^{*}_{i_1}(1')\,\phi_{j_1}(1)\cdots\phi^{*}_{i_p}(p')\,\phi_{j_p}(p). \end{aligned} \tag{2.1}$$

Here ‘‘1’’ represents the spatial and spin coordinates $\mathbf{x}_1=(\mathbf{r}_1,\xi_1)$ of particle 1, etc., and $\{\phi_k\}$ is a basis of orthonormal spin orbitals. We assume that this basis is specified and fixed, so that the p-RDM is completely determined by the tensor $\boldsymbol{\Gamma}^{(p)}$ of expansion coefficients. In order to make our results more amenable to numerical implementation, we shall formulate what follows in terms of tensor arithmetic rather than integral kernels.

The elements of $\boldsymbol{\Gamma}^{(p)}$ can be expressed succinctly in second-quantized notation,

$$(\boldsymbol{\Gamma}^{(p)})^{i_1,\ldots,i_p}_{j_1,\ldots,j_p}=\langle\Psi|\hat{a}^{\dagger}_{i_1}\cdots\hat{a}^{\dagger}_{i_p}\hat{a}_{j_p}\cdots\hat{a}_{j_1}|\Psi\rangle/p!, \tag{2.2}$$

where $\{\hat{a}^{\dagger}_k\}$ and $\{\hat{a}_k\}$ are, respectively, the creation and annihilation operators for the basis $\{\phi_k\}$. The tensor $\boldsymbol{\Gamma}^{(p)}$ is (separately) antisymmetric in both its upper and lower indices. It is also self-adjoint, $(\boldsymbol{\Gamma}^{(p)})^{i_1,\ldots,i_p}_{j_1,\ldots,j_p}=[(\boldsymbol{\Gamma}^{(p)})^{j_1,\ldots,j_p}_{i_1,\ldots,i_p}]^{*}$, and is normalized to $\binom{N}{p}$:

$$\mathrm{tr}\,\boldsymbol{\Gamma}^{(p)}=\sum_{k_1,\ldots,k_p}(\boldsymbol{\Gamma}^{(p)})^{k_1,\ldots,k_p}_{k_1,\ldots,k_p}=\binom{N}{p}. \tag{2.3}$$

For convenience let $\boldsymbol{\gamma}\overset{\mathrm{def}}{=}\boldsymbol{\Gamma}^{(1)}$ and $\boldsymbol{\Gamma}\overset{\mathrm{def}}{=}\boldsymbol{\Gamma}^{(2)}$.

The $(p-n)$-RDM is obtained from the p-RDM via a linear map ${}^{\;p}_{p-n}\!\downarrow$ that is proportional to the n-trace $\mathrm{tr}_{p-n+1,p-n+2,\ldots,p}$. This operation is known as *contraction* and applied to $\boldsymbol{\Gamma}^{(p)}$ it gives

$$\boldsymbol{\Gamma}^{(p-n)}={}^{\;p}_{p-n}\!\downarrow\boldsymbol{\Gamma}^{(p)}=\left[\binom{N}{p-n}\Big/\binom{N}{p}\right]\mathrm{tr}_{p-n+1,\ldots,p}\boldsymbol{\Gamma}^{(p)}. \tag{2.4}$$

For tensors the n-trace is the usual diagonal sum, as in Eq. (2.3), but taken over only the final (rightmost) n indices. For integral kernels the corresponding operation is

$$\begin{aligned}[\mathrm{tr}_{p-n+1,\ldots,p}\boldsymbol{\Gamma}^{(p)}](1,\ldots,p-n;1',\ldots,(p-n)')\\ =\int\boldsymbol{\Gamma}^{(p)}(1,\ldots,p;1',\ldots,(p-n)',p-n+1,\ldots,p)\\ \times d\mathbf{x}_{p-n+1}\cdots d\mathbf{x}_{p}.\end{aligned} \tag{2.5}$$

The goal of so-called direct density matrix calculations is to compute RDMs without calculating wave functions; however, the definitions in Eqs. (2.1) and (2.2) both involve Ψ explicitly. Alternatively, without specifying any particular wave function one may define a *p*-particle density matrix as simply a positive (semidefinite), self-adjoint tensor of rank $2p$. Not all density matrices defined in this way are *reduced* density matrices for real physical systems and one must distinguish the set of physically admissible RDMs. An RDM for a system of indistinguishable fermions is said to be *pure-state N-representable* [1] if it is a contraction of $\Psi\Psi^*$ for some *N*-particle wave function Ψ that is antisymmetric with respect to permutations of particle coordinates.

For statistical ensembles of states $\{\Psi_i\}$ one needs to consider *ensemble N-representable* RDMs, the contractions of convex linear combinations $c_1\Psi_1\Psi_1^* + \cdots + c_k\Psi_k\Psi_k^*$. We will not consider such ensemble states, except to note that each component $\Psi_j\Psi_j^*$ is itself a pure-state *N*-representable density matrix, and therefore its contractions are pure-state *N*-representable RDMs. Consequently, if the $\{\Psi_i\}$ are eigenfunctions of a self-adjoint operator $\hat{\Lambda}$ then each reduced component satisfies a reduced eigenvalue equation (introduced below) with respect to $\hat{\Lambda}$. Furthermore, if $\hat{\Lambda}\Psi_j = \lambda\Psi_j$ for each j then the contractions of $c_1\Psi_1\Psi_1^* + \cdots + c_k\Psi_k\Psi_k^*$ satisfy a reduced eigenvalue equation as well.

We now restrict our attention to pure-state *N*-representable RDMs. Suppose that the *N*-fermion wave function Ψ is an eigenfunction of a self-adjoint operator $\hat{\Lambda}$,

$$\hat{\Lambda}\Psi = \lambda\Psi, \tag{2.6}$$

where $\hat{\Lambda}$ is a sum of one- and two-body operators,

$$\hat{\Lambda} = \sum_i \hat{f}(i) + \sum_{i<j} \hat{g}(i,j). \tag{2.7}$$

For indistinguishable particles, all established quantum-mechanical observables have this form. Clearly

$$\langle\Psi|\hat{a}_p^\dagger\hat{a}_q^\dagger\hat{a}_n\hat{a}_m(\hat{\Lambda}-\lambda)|\Psi\rangle = 0 \tag{2.8}$$

for each *p*, *q*, *n*, and *m*. In this second-quantized formalism we should replace the operator $\hat{\Lambda}$ with the second-quantized expansion of the corresponding two-particle reduced operator $\hat{\Lambda}_{1,2}(1,2) = \hat{g}(1,2) + [\hat{f}(1) + \hat{f}(2)]/(N-1)$. The corresponding operator in second quantization is

$$\hat{\Lambda}_{1,2} \;\leftrightarrow\; \frac{1}{2}\sum_{ijrs} \mathbf{\Lambda}_{ij}^{rs}\hat{a}_i^\dagger\hat{a}_j^\dagger\hat{a}_s\hat{a}_r \tag{2.9}$$

and the expansion coefficients are the integrals

$$\mathbf{\Lambda}_{ij}^{rs} = \langle\phi_i(1)\phi_j(2)|\hat{g}(1,2)|\phi_r(1)\phi_s(2)\rangle + \frac{\langle\phi_i(1)|\hat{f}(1)|\phi_r(1)\rangle\delta_{js} + \langle\phi_j(1)|\hat{f}(1)|\phi_s(1)\rangle\delta_{ir}}{(N-1)}. \tag{2.10}$$

Note that $\hat{\Lambda}_{1,2}$ and $\mathbf{\Lambda}_{ij}^{rs}$ depend upon the number of particles, *N*. $\mathbf{\Lambda}$ is self-adjoint but not antisymmetric, although $\mathbf{\Lambda}_{ij}^{rs} = \mathbf{\Lambda}_{ji}^{sr}$. It is possible to antisymmetrize the integrals to make $\mathbf{\Lambda}_{ij}^{rs} = -\mathbf{\Lambda}_{ji}^{rs} = -\mathbf{\Lambda}_{ij}^{sr} = \mathbf{\Lambda}_{ji}^{sr}$, and for some purposes this may be convenient. Here, however, $\mathbf{\Lambda}$ is *not* antisymmetric. The advantage we gain with definition (2.10) is that for a spin-free operator $\hat{\Lambda}$, the tensor $\mathbf{\Lambda}$ is diagonal in the spin variables of both particles. This block structure is lost if $\mathbf{\Lambda}$ is antisymmetrized.

Inserting Eq. (2.9) into Eq. (2.8) we have

$$\sum_{ijrs} \mathbf{\Lambda}_{ij}^{rs}\langle\Psi|\hat{a}_p^\dagger\hat{a}_q^\dagger\hat{a}_n\hat{a}_m\hat{a}_i^\dagger\hat{a}_j^\dagger\hat{a}_s\hat{a}_r|\Psi\rangle = 2\lambda\langle\Psi|\hat{a}_p^\dagger\hat{a}_q^\dagger\hat{a}_n\hat{a}_m|\Psi\rangle. \tag{2.11}$$

To obtain the *reduced eigenvalue equation* for the operator $\hat{\Lambda}$, we reorder products of creation and annihilation operators in Eq. (2.11) using $\hat{a}_i\hat{a}_j^\dagger + \hat{a}_j^\dagger\hat{a}_i = \delta_{ij}$ and express the result in terms of density matrix elements via Eq. (2.2). This results in a set of equations, which we may write in matrix form as

$$\lambda\mathbf{\Gamma} = \mathbf{\Lambda\Gamma} + 6(\hat{\mathcal{A}}_2\otimes\hat{\mathcal{A}}_2)\mathrm{tr}_3\mathbf{\Lambda\Gamma}^{(3)} + 6\;\;\mathrm{tr}_{3,4}\mathbf{\Lambda\Gamma}^{(4)}. \tag{2.12}$$

An equivalent equation can also be derived in which the order of each matrix product is reversed. The eigenvalue λ is given by

$$\lambda = \langle\Psi|\hat{\Lambda}\Psi\rangle = \mathrm{tr}\,(\mathbf{\Lambda\Gamma}) = \sum_{mnpq}\mathbf{\Lambda}_{pq}^{mn}\;\;\mathbf{\Gamma}_{mn}^{pq}. \tag{2.13}$$

The product tensors in Eq. (2.12) have elements

$$(\mathbf{\Lambda\Gamma})_{rs}^{ij} = \sum_{pq}\mathbf{\Lambda}_{rs}^{pq}\;\;\mathbf{\Gamma}_{pq}^{ij}, \tag{2.14a}$$

$$(\mathbf{\Lambda\Gamma}^{(3)})_{rst}^{ijk} = \sum_{pq}\mathbf{\Lambda}_{st}^{pq}\;\;(\mathbf{\Gamma}^{(3)})_{rpq}^{ijk}, \tag{2.14b}$$

and

$$(\mathbf{\Lambda\Gamma}^{(4)})_{rstu}^{ijk\ell} = \sum_{pq}\mathbf{\Lambda}_{tu}^{pq}\,(\mathbf{\Gamma}^{(4)})_{rspq}^{ijk\ell}. \tag{2.14c}$$

Note that $\mathrm{tr}_{3,4}(\mathbf{\Lambda\Gamma}^{(4)}) = \mathrm{tr}_{3,4}(\mathbf{\Gamma}^{(4)}\mathbf{\Lambda})$ but in general $\mathrm{tr}_3(\mathbf{\Lambda\Gamma}^{(3)}) \neq \mathrm{tr}_3(\mathbf{\Gamma}^{(3)}\mathbf{\Lambda})$ since tr_3 is a sum over one index while the product involves a sum over two.

The operator $\hat{\mathcal{A}}_2$ in Eq. (2.12) is the antisymmetrizing projection operator for two-particle functions, while $\hat{\mathcal{A}}_2\otimes\hat{\mathcal{A}}_2$ is the direct product of $\hat{\mathcal{A}}_2$ for the vector space and $\hat{\mathcal{A}}_2$ for the dual space [see Eq. (3.1) for a rigorous definition]. $\hat{\mathcal{A}}_2\otimes\hat{\mathcal{A}}_2$ thus antisymmetrizes two-particle tensors (i.e., tensors with two upper and two lower indices). Although we consider only fermion density matrices here, by employing the boson commutation rule $\hat{a}_i^\dagger\hat{a}_j - \hat{a}_j\hat{a}_i^\dagger = \delta_{ij}$ one can derive an analog of Eq. (2.12) for (symmetric) boson density matrices; the only difference is that $\hat{\mathcal{A}}_2$ is replaced by the *symmetrizing* projection operator.

In this paper we work exclusively with tensors, in order to derive formulas that are directly applicable to numerical calculations in a finite basis set. Much of the density matrix literature, however, is written in terms of integral kernels. In order to connect with this formalism, we translate the reduced eigenvalue equation (2.12) into this integral kernel language:

$$\begin{aligned}\lambda\Gamma(\mathbf{x}_1,\mathbf{x}_2;\mathbf{x}_1',\mathbf{x}_2') = {}&\int \Lambda_{1,2}(\mathbf{p},\mathbf{q};\mathbf{x}_1',\mathbf{x}_2')\Gamma(\mathbf{x}_1,\mathbf{x}_2;\mathbf{p},\mathbf{q})\,d\mathbf{p}\,d\mathbf{q}\\ &+3\int \Lambda_{1,2}(\mathbf{p},\mathbf{q};\mathbf{x}_2',\mathbf{r})\,\Gamma^{(3)}(\mathbf{x}_1,\mathbf{x}_2,\mathbf{r};\mathbf{x}_1',\mathbf{p},\mathbf{q})\,d\mathbf{p}\,d\mathbf{q}\,d\mathbf{r}\\ &-3\int \Lambda_{1,2}(\mathbf{p},\mathbf{q};\mathbf{x}_1',\mathbf{r})\,\Gamma^{(3)}(\mathbf{x}_1,\mathbf{x}_2,\mathbf{r};\mathbf{x}_2',\mathbf{p},\mathbf{q})\,d\mathbf{p}\,d\mathbf{q}\,d\mathbf{r}\\ &+6\int \Lambda_{1,2}(\mathbf{p},\mathbf{q};\mathbf{r},\mathbf{s})\,\Gamma^{(4)}(\mathbf{x}_1,\mathbf{x}_2,\mathbf{r},\mathbf{s};\mathbf{x}_1',\mathbf{x}_2',\mathbf{p},\mathbf{q})d\mathbf{p}\,d\mathbf{q}\,d\mathbf{r}\,d\mathbf{s}.\end{aligned} \tag{2.15}$$

Here $\mathbf{p},\mathbf{q},\ldots$ each represent the space- and spin-coordinates of a single particle.

The reduced operator $\hat{\Lambda}_{1,2}(1,2)$ has kernel

$$\Lambda_{1,2}(\mathbf{x}_1,\mathbf{x}_2;\mathbf{x}_1',\mathbf{x}_2') = \delta(\mathbf{x}_1-\mathbf{x}_1')\;\delta(\mathbf{x}_2-\mathbf{x}_2')[g(\mathbf{x}_1,\mathbf{x}_2)+\{f(\mathbf{x}_1)+f(\mathbf{x}_2)\}/(N-1)]. \tag{2.16}$$

Substituting this expression into Eq. (2.15), evaluating the δ function integrals, and making use of the contraction relation (2.5) between density matrix kernels yields

$$\begin{aligned}\lambda\Gamma(1,2;1',2') = {}&[f(1')+f(2')+g(1',2')]\Gamma(1,2;1',2')\\ &+3\int \{[f(3')+g(1',3')+g(2',3')]\Gamma^{(3)}(1,2,3;1',2',3')\}_{3'=3}d\mathbf{x}_3\\ &+6\int \{g(3',4')\,\Gamma^{(4)}(1,2,3,4;1',2',3',4')\}_{\substack{3'=3\\4'=4}}d\mathbf{x}_3 d\mathbf{x}_4.\end{aligned} \tag{2.17}$$

This is the integral kernel version of the reduced eigenvalue equation for $\hat{\Lambda}$ [15,16,22]. The notation $\{\cdots\}_{3'=3}$ means that $\mathbf{x}_3'$ is set equal to $\mathbf{x}_3$ in the integrand *after* the operators $\hat{f}$ and $\hat{g}$ have acted on the primed coordinates. [The tensor expansion of density matrix kernels in Eq. (2.1) forces us into the somewhat unconventional situation of having operators act on *primed* coordinates.] For Hamiltonian operators, $\hat{\Lambda}=\hat{H}$, the reduced eigenvalue equation (2.12) is known as CSE; the corresponding integral kernel equation (2.17) is sometimes called the density equation [44].

We have shown above how the CSE follows from the usual Schrödinger equation, but in fact the two are equivalent (in a necessary and sufficient sense) for *N*-representable density matrices [19,22]. In other words, given some antisymmetric, normalized (but otherwise arbitrary) function $\Phi(1,\ldots,N)$, suppose we define $\mathbf{\Gamma}$, $\mathbf{\Gamma}^{(3)}$, and $\mathbf{\Gamma}^{(4)}$ as the appropriate contractions of $\Phi\Phi^*$. Then $\mathbf{\Gamma}$, $\mathbf{\Gamma}^{(3)}$, and $\mathbf{\Gamma}^{(4)}$ satisfy Eq. (2.12) if and only if $\hat{\Lambda}\Phi=\lambda\Phi$, where $\lambda=\langle\Phi|\hat{\Lambda}\Phi\rangle$. Our derivation of Eq. (2.12) demonstrates how this equation follows from Eq. (2.6); the proof of the converse for *N*-representable RDMs is analogous to the proof that the CSE implies the ordinary Schrödinger equation. This was originally demonstrated by Nakatsuji [22], then later proved in second quantization by Mazziotti [19] and will not be repeated here. The crucial point is that Eq. (2.12) implies the zero-dispersion condition $\langle\Phi|\hat{\Lambda}\Phi\rangle^2=\langle\Phi|\hat{\Lambda}^2\Phi\rangle$, which for self-adjoint operators is equivalent to the eigenvalue equation $\hat{\Lambda}\Phi=\langle\Phi|\hat{\Lambda}\Phi\rangle\Phi$. The presence of $\hat{\Lambda}^2$, a four-particle operator, in the zero-dispersion relation indicates why the 3- and 4-RDMs must appear in the reduced equation even though $\mathbf{\Gamma}$ alone determines the eigenvalue.

The reduced eigenvalue equation for the total spin operator $\hat{S}^2$ has been discussed by Valdemoro *et al.* [13], although its formal similarity to the CSE has not been emphasized. To our knowledge all other work has focused exclusively on the CSE and in what follows we, too, shall take $\hat{\Lambda}=\hat{H}$ as a Hamiltonian. In keeping with standard notation, we will use $\mathbf{K}=\mathbf{\Lambda}$ for the matrix of the two-electron reduced Hamiltonian.

III. RECONSTRUCTION FUNCTIONALS

Since $\mathbf{\Gamma}$ contains the part of Ψ needed to compute expectation values of one- and two-electron operators, we would like to determine the 2-RDM directly from Eq. (2.12). The equivalence of the Schrödinger equation and the CSE implies that *within the set of N-representable 2-RDMs* such a unique solution of the CSE is possible (up to degeneracy). However, necessary and sufficient N-representability conditions for the 2-RDM are unknown [2], and outside the set of N-representable 2-RDMs the solution of the system of equations in Eq. (2.12) is not unique [24].

As in the Green's function literature, one means to sidestep this indeterminacy is to decouple the higher-order RDMs into products of lower-order density matrices. Thus we seek *reconstruction functionals* $\mathbf{\Gamma}^{(3)}[\mathbf{\Gamma}]$ and $\mathbf{\Gamma}^{(4)}[\mathbf{\Gamma}]$ that we may substitute into Eq. (2.12) to produce a nonlinear equation with $\mathbf{\Gamma}$ as the only unknown. This equation can then be solved self-consistently for $\mathbf{\Gamma}$.

Grassmann product ansatz

Several approximate reconstruction functionals $\mathbf{\Gamma}^{(3)}[\mathbf{\Gamma}]$ and $\mathbf{\Gamma}^{(4)}[\mathbf{\Gamma}]$ have been proposed, and Mazziotti [19] has shown how the content of these reconstruction approximations is described conveniently using antisymmetrized (Grassmann) products. If $\mathbf{A}=(A^{i_1,\ldots,i_n}_{j_1,\ldots,j_n})$ and $\mathbf{B}=(B^{i_1,\ldots,i_m}_{j_1,\ldots,j_m})$ are tensors, then their antisymmetrized product $\mathbf{A}\wedge\mathbf{B}$ is the tensor with elements [19,27]

$$\begin{aligned}(\mathbf{A}\wedge\mathbf{B})^{i_1,\ldots,i_{m+n}}_{j_1,\ldots,j_{m+n}} &\overset{\text{def}}{=} (\hat{\mathcal{A}}_{m+n}\otimes\hat{\mathcal{A}}_{m+n})A^{i_1,\ldots,i_n}_{j_1,\ldots,j_n}B^{i_{n+1},\ldots,i_{m+n}}_{j_{n+1},\ldots,j_{m+n}}\\ &=[(m+n)!]^{-2}\sum_{\sigma,\pi\in\mathbb{S}_{m+n}}\epsilon(\sigma)\,\epsilon(\pi)A^{\sigma(i_1),\ldots,\sigma(i_n)}_{\pi(j_1),\ldots,\pi(j_n)}\\ &\quad\times B^{\sigma(i_{n+1}),\ldots,\sigma(i_{m+n})}_{\pi(j_{n+1}),\ldots,\pi(j_{m+n})}. \end{aligned} \tag{3.1}$$

Here $\mathbb{S}_{m+n}$ denotes the symmetric group of order $(m+n)!$ and $\epsilon(\sigma)=\pm 1$ is the parity of $\sigma\in\mathbb{S}_{m+n}$. The product $\wedge$ is commutative, distributive, and associative, and $\mathbf{A}\wedge\mathbf{B}$ is a linear operator on the $(m+n)$-particle space. If $\mathbf{A}$ and $\mathbf{B}$ are self-adjoint then so is $\mathbf{A}\wedge\mathbf{B}$. For convenience we also define

$$\mathbf{A}^{\wedge n} \overset{\text{def}}{=} \underbrace{\mathbf{A}\wedge\mathbf{A}\wedge\cdots\wedge\mathbf{A}}_{n\ \text{factors}}. \tag{3.2}$$

Such "wedge powers" must be distinguished from matrix products; we denote the latter by simple exponents.

The 3- and 4-RDM reconstruction functionals proposed by Valdemoro and co-workers [31,32] take the form of certain Grassmann products of $\boldsymbol{\gamma}$ and $\mathbf{\Gamma}$, and are derived by considering exact relations between particle- and hole-RDMs. By positing an approximate separation between the particle and hole matrices, one may equate $\mathbf{\Gamma}^{(p)}$ to a sum of Grassmann products of lower-order RDMs [19,32]. For $p=3$ the result is Valdemoro's $\mathbf{\Gamma}^{(3)}[\mathbf{\Gamma}]$ functional,

$$\mathbf{\Gamma}^{(3)}_{\text{Val}}[\mathbf{\Gamma}] \overset{\text{def}}{=} 3\,\boldsymbol{\gamma}\wedge\mathbf{\Gamma}-2\,\boldsymbol{\gamma}^{\wedge 3}, \tag{3.3}$$

while for $p=4$ the approximate particle-hole separation yields

$$\mathbf{\Gamma}^{(4)}\approx 4\;\boldsymbol{\gamma}\wedge\mathbf{\Gamma}^{(3)}-6\;\boldsymbol{\gamma}\wedge\boldsymbol{\gamma}\wedge\mathbf{\Gamma}+3\;\boldsymbol{\gamma}^{\wedge 4}. \tag{3.4}$$

Substituting $\mathbf{\Gamma}^{(3)}_{\text{Val}}[\mathbf{\Gamma}]$ for $\mathbf{\Gamma}^{(3)}$ above results in the reconstruction functional

$$\mathbf{\Gamma}^{(4)}_{\text{Val}}[\mathbf{\Gamma}] \overset{\text{def}}{=} 6\;\boldsymbol{\gamma}\wedge\boldsymbol{\gamma}\wedge\mathbf{\Gamma}-5\;\boldsymbol{\gamma}^{\wedge 4}. \tag{3.5}$$

If $\mathbf{\Gamma}$ derives from a single determinant of spin orbitals then $\mathbf{\Gamma}^{(p)}=\boldsymbol{\gamma}^{\wedge p}$ for each p; in this case $\mathbf{\Gamma}^{(3)}_{\text{Val}}[\mathbf{\Gamma}]$ and $\mathbf{\Gamma}^{(4)}_{\text{Val}}[\mathbf{\Gamma}]$ are exact. Furthermore, using diagrammatic expansions of the three- and four-electron Green's functions, Nakatsuji and Yasuda [15,16] have shown that Valdemoro's reconstruction functionals give $\mathbf{\Gamma}^{(3)}$ and $\mathbf{\Gamma}^{(4)}$ to first order in the correlation potential.

Nakatsuji and Yasuda [15,16] carry the perturbation expansion for $\mathbf{\Gamma}^{(4)}$ to second order in the correlation potential to obtain a correction to Eq. (3.4):

$$\begin{aligned}\mathbf{\Gamma}^{(4)}&\approx 6\;\boldsymbol{\gamma}^{\wedge 4}+4\;\boldsymbol{\gamma}\wedge\mathbf{\Gamma}^{(3)}-12\;\boldsymbol{\gamma}\wedge\boldsymbol{\gamma}\wedge\mathbf{\Gamma}+3\;\mathbf{\Gamma}\wedge\mathbf{\Gamma}\\ &=\mathbf{\Gamma}^{(4)}_{\text{Val}}+3(\mathbf{\Gamma}-\boldsymbol{\gamma}\wedge\boldsymbol{\gamma})\wedge(\mathbf{\Gamma}-\boldsymbol{\gamma}\wedge\boldsymbol{\gamma}).\end{aligned} \tag{3.6}$$

These authors also perform a diagrammatic expansion of $\mathbf{\Gamma}^{(3)}$ but do not sum all of the second-order diagrams. The resulting partial second-order expression for $\mathbf{\Gamma}^{(3)}$ defines the Nakatsuji-Yasuda 3-RDM reconstruction functional,

$$\mathbf{\Gamma}^{(3)}_{\text{NY}}[\mathbf{\Gamma}] \overset{\text{def}}{=} 3\;\boldsymbol{\gamma}\wedge\mathbf{\Gamma}-2\;\boldsymbol{\gamma}^{\wedge 3}+\mathbf{\Gamma}^{(3)}_{\text{UV}}. \tag{3.7}$$

$\mathbf{\Gamma}^{(3)}_{\text{UV}}$ represents the second-order contributions [cf. Eq. (3.3)] but cannot be written in terms of Grassmann products of $\boldsymbol{\gamma}$ and $\mathbf{\Gamma}$. Consequently, the 4-matrix reconstruction functional of Nakatsuji and Yasuda, which is obtained from Eq. (3.6) by substituting $\mathbf{\Gamma}^{(3)}_{\text{NY}}$ in place of the exact $\mathbf{\Gamma}^{(3)}$, does not have the Grassmann product form. In addition, evaluation of the Nakatsuji-Yasuda 4-matrix reconstruction functional is much more computationally demanding than reconstruction via $\mathbf{\Gamma}^{(4)}_{\text{G}}$, and we will not consider the Nakatsuji-Yasuda reconstruction functionals in this paper.

Instead, we focus our attention on the possible Grassmann product reconstruction functionals $\mathbf{\Gamma}^{(4)}_{\text{G}}$. In Sec. V we derive expressions for the contractions of $\mathbf{\Gamma}^{(4)}_{\text{G}}$. These formulas demonstrate that a Grassmann product form for $\mathbf{\Gamma}^{(4)}$ is inconsistent with a Grassmann product form for the lower-order RDMs. Nevertheless, our expressions for ${}^{4}_{3}{\downarrow}\mathbf{\Gamma}^{(4)}_{\text{G}}$ and ${}^{4}_{2}{\downarrow}\mathbf{\Gamma}^{(4)}_{\text{G}}$ allow us to obtain a self-consistent solution to the CSE; moreover, these expressions also allow for a *contraction-consistent* solution in the sense that all density matrices on the right side of the CSE [Eq. (2.12)] are related by contraction. (Note that this relationship is utilized explicitly in deriving the CSE.) Specifically, if $\mathbf{\Gamma}_n$ is the (approximate) 2-matrix after n iterations, we might construct an updated 2-matrix $\mathbf{\Gamma}_{n+1}$ according to

$$\mathbf{\Gamma}_{n+1}=\frac{1}{E_n}[\mathbf{K}(\,{}^{4}_{2}\!\downarrow\mathbf{\Gamma}^{(4)}[\mathbf{\Gamma}_n])+6(\hat{\mathcal{A}}_2\otimes\hat{\mathcal{A}}_2)\times\mathrm{tr}_3\{\mathbf{K}(\,{}^{4}_{3}\!\downarrow\mathbf{\Gamma}^{(4)}[\mathbf{\Gamma}_n])\}+6\ \mathrm{tr}_{3,4}\{\mathbf{K}(\mathbf{\Gamma}^{(4)}[\mathbf{\Gamma}_n])\}], \tag{3.8}$$

with $E_n=\mathrm{tr}(\mathbf{K\Gamma}_n)$ the approximate energy after n iterations. Given a reconstruction functional for $\mathbf{\Gamma}^{(4)}$, $\mathbf{\Gamma}_n$ is the only unknown on the right side of Eq. (3.8), so there is no need to construct (or store) any three- or four-electron tensors in order to obtain $\mathbf{\Gamma}_{n+1}$. The advantages of this are further discussed and quantified in the Appendix.

The self-consistent iteration formula in Eq. (3.8) enforces the contraction relations at each iteration, and is the iteration algorithm currently employed by Valdemoro and co-workers [12–14], with $\mathbf{\Gamma}^{(4)}[\mathbf{\Gamma}]$ given by Eq. (3.5). [In truth, Valdemoro's procedure also includes adjustments to correct for normalization, positivity, and $\hat{S}^2$ eigenvalue, but once these adjustments are made iteration proceeds according to Eq. (3.8).] This would seem to be a reasonable manner in which to proceed, since contraction consistency is an exact requirement of the physical density matrices. One can, however, envision other reasonable procedures as well.

Since both Valdemoro and Nakatsuji derive reconstruction functionals for the 3-RDM, we might consider employing separate reconstructions for $\mathbf{\Gamma}^{(3)}$ and $\mathbf{\Gamma}^{(4)}$. This corresponds to

$$\mathbf{\Gamma}_{n+1}=\frac{1}{E_n}[\mathbf{K\Gamma}_n+6(\hat{\mathcal{A}}_2\otimes\hat{\mathcal{A}}_2)\mathrm{tr}_3\{\mathbf{K\Gamma}^{(3)}[\mathbf{\Gamma}_n]\}+6\ \mathrm{tr}_{3,4}\{\mathbf{K}(\mathbf{\Gamma}^{(4)}[\mathbf{\Gamma}_n])\}], \tag{3.9}$$

which was the iteration formula originally used by Colmenero and Valdemoro [11]. Finally, Mazziotti's iteration formula [19,20] is an intermediate case in which $\mathbf{\Gamma}^{(3)}$ is obtained by contraction but the input matrix $\mathbf{\Gamma}_n$ at the nth step is used as the 2-RDM:

$$\mathbf{\Gamma}_{n+1}=\frac{1}{E_n}[\mathbf{K\Gamma}_n+6(\hat{\mathcal{A}}_2\otimes\hat{\mathcal{A}}_2)\mathrm{tr}_3\{\mathbf{K}(\,{}^{4}_{3}\!\downarrow\mathbf{\Gamma}^{(4)}[\mathbf{\Gamma}_n])\}+6\ \mathrm{tr}_{3,4}\{\mathbf{K}(\mathbf{\Gamma}^{(4)}[\mathbf{\Gamma}_n])\}]. \tag{3.10}$$

Mazziotti's "ensemble representability method" [19] consists in Eq. (3.10) supplemented by positivity conditions.

In Sec. VI we provide a numerical comparison of these three iteration schemes using $\mathbf{\Gamma}^{(3)}_{\mathrm{Val}}$ and $\mathbf{\Gamma}^{(4)}_{\mathrm{Val}}$ along with two other reconstruction functionals in the class $\mathbf{\Gamma}^{(4)}_{\mathrm{G}}$.

IV. CONTRACTION THEOREMS

Constraints imposed upon RDM reconstruction functionals by the contraction relations in Eq. (2.4) have been discussed by several authors. In particular, Kutzelnigg and Mukherjee [33] and Mazziotti [29] have used the contraction relations in deriving cumulant expansions of RDMs. Valdemoro, Tel, and Pérez-Romero [34] derive an explicit formula for $\mathbf{I}^{(p)}\wedge\mathbf{\Gamma}^{(q)}$ as a linear combination of the Grassmann products $\mathbf{I}^{(p)}\wedge\mathbf{\Gamma}^{(q-1)}$ and $\mathbf{I}^{(p-1)}\wedge\mathbf{\Gamma}^{(q)}$, where $\mathbf{I}^{(n)}$ denotes the identity operator on the n-particle space. Coleman and Absar [27] have also consider decomposition of RDMs into certain Grassmann products involving identity matrices, although not in the context of density matrix reconstruction.

None of the aforementioned work provides explicit expressions for the contractions of the Grassmann product 4-matrix $\mathbf{\Gamma}^{(4)}_{\mathrm{G}}$; the derivation of such expressions occupies this section and the next. In this section we establish several theorems concerning the one-particle contractions of antisymmetrized products of $\boldsymbol{\gamma}$ and $\mathbf{\Gamma}$ that will facilitate contraction of $\mathbf{\Gamma}^{(4)}_{\mathrm{G}}$. The techniques used in each proof are similar, so we provide detailed proofs only for the first two theorems in this section. Because the notation used in the theorems (not to mention the proofs) is somewhat involved, following several of the theorems we present examples of their application prior to embarking upon a formal proof.

We first define a deletion operator $\ominus$. If $\mathbf{T}=\mathbf{T}_1\wedge\mathbf{T}_2\wedge\cdots\wedge\mathbf{T}_n$ is the antisymmetrized product of a set of tensors $\mathcal{T}=\{\mathbf{T}_i|i=1,\ldots,n\}$ and $\mathcal{S}\subseteq\mathcal{T}$, then

$$\mathbf{T}\ominus\mathcal{S}\overset{\mathrm{def}}{=}\bigwedge_{\mathbf{T}_i\in\mathcal{T}\setminus\mathcal{S}}\mathbf{T}_i. \tag{4.1}$$

For example, $\mathbf{T}\ominus\{\mathbf{T}_1,\mathbf{T}_2\}=\mathbf{T}_3\wedge\cdots\wedge\mathbf{T}_n$.

Theorem 1. Let n be a positive integer and $p_1,\ldots,p_n$ non-negative integers with $p_1+\cdots+p_n=P$. Define the P-particle tensor

$$\mathbf{T}=\boldsymbol{\gamma}^{\wedge p_1}\wedge(\boldsymbol{\gamma}^2)^{\wedge p_2}\wedge\cdots\wedge(\boldsymbol{\gamma}^n)^{\wedge p_n}.$$

Then the one-particle contraction of $\mathbf{T}$ is given by

$$\begin{aligned}\mathrm{tr}_P(\mathbf{T})=&(1/P^2)\sum_{m=1}^{n}p_m\,\mathrm{tr}(\boldsymbol{\gamma}^m)[\mathbf{T}\ominus\{\boldsymbol{\gamma}^m\}]\\&-(1/P^2)\sum_{m=1}^{n}p_m(p_m-1)[\mathbf{T}\ominus\{\boldsymbol{\gamma}^m,\boldsymbol{\gamma}^m\}]\wedge\boldsymbol{\gamma}^{2m}\\&-(2/P^2)\sum_{m=1}^{n}\sum_{\ell=m+1}^{n}p_mp_\ell[\mathbf{T}\ominus\{\boldsymbol{\gamma}^m,\boldsymbol{\gamma}^\ell\}]\wedge\boldsymbol{\gamma}^{\ell+m}.\end{aligned}$$

Remark. Deletions such as $\mathbf{T}\ominus\{\boldsymbol{\gamma}^m,\boldsymbol{\gamma}^m\}$ make sense only if $p_m\geqslant 2$. If $p_m<2$, however, this term does not appear as a result of the multiplicative factor $p_m(p_m-1)$.

Example 1. Since tr $\boldsymbol{\gamma}=N$,

$$\mathrm{tr}_p(\boldsymbol{\gamma}^{\wedge p})=\frac{N}{p}\boldsymbol{\gamma}^{\wedge(p-1)}-\frac{(p-1)}{p}[\boldsymbol{\gamma}^{\wedge(p-2)}\wedge\boldsymbol{\gamma}^2] \tag{4.2}$$

for $p\geqslant 2$. This illustrates the case $n=1$ and $p_1=p$.

Example 2. The case $n=3$, $p_1=2$, $p_2=0$, and $p_3=1$ (so $P=3$) corresponds to

$$\mathrm{tr}_3(\boldsymbol{\gamma}\wedge\boldsymbol{\gamma}\wedge\boldsymbol{\gamma}^3)=\frac{1}{9}[2N(\boldsymbol{\gamma}\wedge\boldsymbol{\gamma}^3)+(\mathrm{tr}\,\boldsymbol{\gamma}^3)(\gamma\wedge\gamma) -2(\boldsymbol{\gamma}^2\wedge\boldsymbol{\gamma}^3)-4(\boldsymbol{\gamma}\wedge\boldsymbol{\gamma}^4)]. \tag{4.3}$$

Proof of Theorem 1. The elements of **T** are given by

$$T^{i_1,\ldots,i_P}_{j_1,\ldots,j_P} =(P!)^{-2}\sum_{\sigma,\pi\in\mathbb{S}_P}\epsilon(\sigma)\,\epsilon(\pi)\,(\boldsymbol{\gamma}^{\omega})^{\sigma(i_1)}_{\pi(j_1)}\cdots(\boldsymbol{\gamma}^{\upsilon})^{\sigma(i_P)}_{\pi(j_P)}, \tag{4.4}$$

where ω and υ are, respectively, the smallest and largest values of m such that $p_m>0$. For simplicity, we define two ordered P tuples,

$$I_P \overset{\mathrm{def}}{=} \{i_1,\ldots,i_P\},$$

$$J_P \overset{\mathrm{def}}{=} \{j_1,\ldots,j_P\}, \tag{4.5}$$

which are reordered by permutations $\sigma,\pi\in\mathbb{S}_P$ according to

$$\sigma(I_P)=\{\sigma(i_1),\ldots,\sigma(i_P)\},$$

$$\pi(J_P)=\{\pi(j_1),\ldots,\pi(j_P)\}. \tag{4.6}$$

Furthermore, define products

$$[\gamma(\vec{x})]^{I_P}_{J_P} \overset{\mathrm{def}}{=} (\boldsymbol{\gamma}^{\omega})^{i_1}_{j_1}\cdots(\boldsymbol{\gamma}^{\upsilon})^{i_P}_{j_P}, \tag{4.7}$$

in which $\vec{x}=\{\omega,\ldots,\upsilon\}$ is a P-tuple whose rth component x_r is the exponent for the rth term in the product. We can express the elements of $\mathrm{tr}_P(\mathbf{T})$ as

$$(\mathrm{tr}_P\,\mathbf{T})^{i_1,\ldots,i_{P-1}}_{j_1,\ldots,j_{P-1}}=(P!)^{-2}\mathrm{tr}_P(T^{I_P}_{J_P}), \tag{4.8}$$

where

$$T^{I_P}_{J_P} \overset{\mathrm{def}}{=} \sum_{\sigma,\pi\in\mathbb{S}_P}\epsilon(\sigma)\quad\epsilon(\pi)\quad[\gamma(\vec{x})]^{\sigma(I_P)}_{\pi(J_P)}. \tag{4.9}$$

Because the P-trace operator can be written as

$$\mathrm{tr}_P(\cdots)=\sum_k\ \delta_{i_P,k}\delta_{j_P,k}(\cdots), \tag{4.10}$$

we will group the permutations in Eq. (4.9) according to where they place i_P and j_P. There are three possibilities: i_P and j_P can index the same matrix, they can index different matrices having the same exponent, or they can index different matrices with different exponents. This leads to the partition

$$T^{I_P}_{J_P}=\sum_r\ (T^{I_P}_{J_P})_{rr}+\sum_{r,s\neq r}(T^{I_P}_{J_P})_{rs}+\sum_{r,s}(T^{I_P}_{J_P})'_{rs}, \tag{4.11}$$

where each summand represents a restricted sum over permutations in Eq. (4.9). $(T^{I_P}_{J_P})_{rr}$ includes those $\sigma,\pi\in\mathbb{S}_P$ for which $\sigma(r)=P=\pi(r)$. Included in $(T^{I_P}_{J_P})_{rs}$ are permutations for which $x_r=x_s$, $r\neq s$, and $\sigma(r)=P=\pi(s)$. Finally, $(T^{I_P}_{J_P})'_{rs}$ is restricted to σ and π with $\sigma(r)=P=\pi(s)$ and $x_r\neq x_s$. Together the three summations in Eq. (4.11) exhaust all pairs of permutations in $\mathbb{S}_P\otimes\mathbb{S}_P$. We evaluate each sum separately, then apply tr_P to each and thus obtain the three terms in $\mathrm{tr}_P(\mathbf{T})$.

A. Evaluation of the first sum in Eq. (4.11)

To treat terms in the first sum, consider a permutation $\tau^{(P)}_r\in\mathbb{S}_P$ with the effect

$$\begin{aligned}\tau^{(P)}_r\sigma(I_P)&=\{\sigma(i_1),\ldots,\sigma(i_{r-1}),\sigma(i_{r+1}),\ldots,\sigma(i_P),\sigma(i_r)\}\\&=\{\sigma'(I^{(P-1)}_r),\sigma(i_r)\}.\end{aligned} \tag{4.12}$$

$I^{(P-1)}_r \overset{\mathrm{def}}{=} I_P\backslash\{\sigma(i_r)\}$ and $\sigma'\in\mathbb{S}_{P-1}$ puts these $P-1$ indices in the same order that they have in $\sigma(I_P)$. Similarly,

$$\tau^{(P)}_r\pi(J_P)=\{\pi'(J^{(P-1)}_r),\pi(j_r)\} \tag{4.13}$$

for some $\pi'\in\mathbb{S}_{P-1}$. Observe that $\epsilon(\sigma')=\epsilon(\tau^{(P)}_r)\,\epsilon(\sigma)$ and $\epsilon(\pi')=\epsilon(\tau^{(P)}_r)\,\epsilon(\pi)$ so $\epsilon(\sigma')\,\epsilon(\pi')=\epsilon(\sigma)\,\epsilon(\pi)$.

Recall that i_r and j_r index the matrix $\boldsymbol{\gamma}^{x_r}$ in the product $[\gamma(\vec{x})]^{I_P}_{J_P}$. This of course implies that $p_{x_r}>0$; in fact there are exactly p_{x_r} choices of r with the same exponent x_r. Thus

$$\begin{aligned}(T^{I_P}_{J_P})_{rr}&=\sideset{}{'}\sum_{\substack{\sigma,\pi\in\mathbb{S}_P\\ \sigma(r)=P=\pi(r)}}\epsilon(\sigma')\,\epsilon(\pi')\,(\boldsymbol{\gamma}^{x_r})^{i_P}_{j_P}([\gamma(\vec{x})\backslash\{\boldsymbol{\gamma}^{x_r}\}]^{\sigma'(I^{(P-1)}_r)}_{\pi'(J^{(P-1)}_r)})\\&=[(P-1)!]^2([\mathbf{T}\ominus\{\boldsymbol{\gamma}^{x_r}\}]^{I_{P-1}}_{J_{P-1}})(\boldsymbol{\gamma}^{x_r})^{i_P}_{j_P}.\end{aligned} \tag{4.14}$$

$[\mathbf{T}\ominus\{\boldsymbol{\gamma}^{x_r}\}]^{I_{P-1}}_{J_{P-1}}$ is, by definition, a Grassmann product on $P-1$ indices, and includes a factor of $[(P-1)!]^{-2}$.

Application of tr_P to Eq. (4.14) gives $\mathrm{tr}\,\boldsymbol{\gamma}^{x_r}$ in place of the last term. There are p_{x_r} (possibly zero) values of r for a given x_r, and it follows that

$$\mathrm{tr}_P\left[(P!)^{-2}\sum_r (T^{I_P}_{J_P})_{rr}\right]=P^{-2}\sum_{m=1}^{n} p_m(\mathrm{tr}\,\boldsymbol{\gamma}^m)[\mathbf{T}\ominus\{\boldsymbol{\gamma}^m\}]. \tag{4.15}$$

This is precisely the first term in the statement of the theorem.

B. Evaluation of the second and third sums in Eq. (4.11)

Initially the evaluation of both $(T^{I_P}_{J_P})_{rs}$ and $(T^{I_P}_{J_P})'_{rs}$ in Eq. (4.11) proceeds in the same fashion; we first evaluate $(T^{I_P}_{J_P})_{rs}$, $r\neq s$, then make appropriate modifications to obtain $(T^{I_P}_{J_P})'_{rs}$. Fix $r\neq s$ and consider the action of permutations $\tau_r^{(P)}\in\mathbb{S}_P$ and $\tau_s^{(P-1)}\in\mathbb{S}_{P-1}$ as defined in Eq. (4.12):

$$\begin{aligned}\tau_s^{(P-1)}\tau_r^{(P)}\sigma(I_P)&=\tau_s^{(P-1)}\{\sigma'(I_r^{(P-1)}),\sigma(i_r)\}\\&=\{\sigma''(I_{r,s}^{(P-2)}),\sigma(i_s),\sigma(i_r)\},\end{aligned} \tag{4.16}$$

where

$$\sigma'(I_r^{(P-1)})=\{\sigma''(I_{r,s}^{(P-2)}),\sigma(i_s)\}. \tag{4.17}$$

$\sigma''\in\mathbb{S}_{P-2}$ orders the indices in $I_{r,s}^{(P-2)}\overset{\mathrm{def}}{=}I_P\backslash\{\sigma(i_r),\sigma(i_s)\}$ in the same way as they appear in $\sigma(I_P)$. Similarly,

$$\tau_s^{(P-1)}\tau_r^{(P)}\pi(J_P)=\{\pi''(J_{r,s}^{(P-2)}),\pi(j_s),\pi(j_r)\} \tag{4.18}$$

for some $\pi''\in\mathbb{S}_{P-2}$. Then $\epsilon(\sigma'')=\epsilon(\tau_s^{(P-1)}\tau_r^{(P)})\,\epsilon(\sigma)$ and $\epsilon(\pi'')=\epsilon(\tau_s^{(P-1)}\tau_r^{(P)})\,\epsilon(\pi)$ and consequently $\epsilon(\sigma'')\,\epsilon(\pi'')=\epsilon(\sigma)\,\epsilon(\pi)$.

Thus we have

$$(T^{I_P}_{J_P})_{rs}=\sideset{}{'}\sum_{\substack{\sigma,\pi\in\mathbb{S}_P\\ \sigma(r)=P=\pi(s)}}\epsilon(\sigma'')\,\epsilon(\pi'')\,[\gamma(\vec{x})\backslash\{\boldsymbol{\gamma}^{x_r},\boldsymbol{\gamma}^{x_s}\}]^{\sigma''(I_{r,s}^{(P-2)})}_{\pi''(I_{r,s}^{(P-2)})}(\boldsymbol{\gamma}^{x_s})^{\sigma(i_s)}_{\pi(j_s)}(\boldsymbol{\gamma}^{x_r})^{\sigma(i_r)}_{\pi(j_r)} \tag{4.19}$$

so that

$$\mathrm{tr}_P\left[\sum_{r,s\neq r}(T^{I_P}_{J_P})_{rs}\right]=\sum_{r,s\neq r}\sideset{}{'}\sum_{\substack{\sigma,\pi\in\mathbb{S}_P\\ \sigma(r)=P=\pi(s)}}\epsilon(\sigma'')\,\epsilon(\pi'')\,[\gamma(\vec{x})\backslash\{\boldsymbol{\gamma}^{x_r},\boldsymbol{\gamma}^{x_s}\}]^{\sigma''(I_{r,s}^{(P-2)})}_{\pi''(I_{r,s}^{(P-2)})}(\boldsymbol{\gamma}^{x_r+x_s})^{\sigma(i_s)}_{\pi(j_r)}. \tag{4.20}$$

To cast this as a Grassmann product consider the permutation $\sigma'\in\mathbb{S}_{P-1}$ defined in Eq. (4.17), along with the analogous permutation $\pi'\in\mathbb{S}_{P-1}$,

$$\pi'(J_s^{(P-1)})=\{\pi''(J_{r,s}^{(P-2)}),\pi(j_r)\}. \tag{4.21}$$

Comparing Eqs. (4.16), (4.17), and (4.21) we find that $\epsilon(\sigma'')\,\epsilon(\pi'')=-\epsilon(\sigma')\,\epsilon(\pi')$. Thus

$$\begin{aligned}\mathrm{tr}_P\left[\sum_{r,s\neq r}(T^{I_P}_{J_P})_{rs}\right]&=-\sum_{r,s\neq r}\sum_{\sigma',\pi'\in\mathbb{S}_{P-1}}\epsilon(\sigma')\,\epsilon(\pi')([\gamma(\vec{x})\backslash\{\boldsymbol{\gamma}^{x_r},\boldsymbol{\gamma}^{x_s}\}]\boldsymbol{\gamma}^{x_r+x_s})^{\sigma'(I_r^{(P-1)})}_{\pi'(J_s^{(P-1)})}\\&=-[(P-1)!]^2\sum_{r,s\neq r}([\mathbf{T}\ominus\{\boldsymbol{\gamma}^{x_r},\boldsymbol{\gamma}^{x_s}\}]\wedge\boldsymbol{\gamma}^{x_r+x_s})^{I_{P-1}}_{J_{P-1}}.\end{aligned} \tag{4.22}$$

So far we have not used the fact that $x_r=x_s$; consequently Eq. (4.22) remains valid if we replace $(T^{I_P}_{J_P})_{rs}$ with $(T^{I_P}_{J_P})'_{rs}$. To evaluate Eq. (4.22) with $x_r=x_s$, observe that there are p_{x_r} choices for r and $(p_{x_r}-1)$ choices for $s\neq r$ such that $x_r=x_s$. Hence

$$\begin{aligned}(P!)^{-2}\,\mathrm{tr}_P\left[\sum_{r,s\neq r}(T^{I_P}_{J_P})_{rs}\right]&=-P^{-2}\sum_m p_m(p_m-1)\\&\quad\times[\mathbf{T}\ominus\{\boldsymbol{\gamma}^{x_m},\boldsymbol{\gamma}^{x_m}\}]\wedge\boldsymbol{\gamma}^{2m}.\end{aligned} \tag{4.23}$$

To obtain $\mathrm{tr}_P\Sigma_{r,s}(T^{I_P}_{J_P})'_{rs}$, consider the aforementioned analog of Eq. (4.22), assuming $x_r \neq x_s$. Then there are $p_{x_r}p_{x_s}$ choices (x_r, x_s) for the exponents with $r>s$, and another $p_{x_r}p_{x_s}$ choices with $r<s$. Both cases give the same result in Eq. (4.22) so

$$(P!)^{-2}\,\mathrm{tr}_P\left[\sum_{r,s}(T^{I_P}_{J_P})'_{rs}\right]$$
$$=-2P^{-2}\sum_{l=1}^{n}\sum_{m=l+1}^{n}p_l p_m[\mathbf{T}\ominus\{\boldsymbol{\gamma}^{x_m},\boldsymbol{\gamma}^{x_l}\}]\wedge\boldsymbol{\gamma}^{m+l}. \tag{4.24}$$

Equations (4.23) and (4.24) are, respectively, the second and third terms in the statement of the theorem. ■

In order to state the next theorem succinctly we define one-index products $\boldsymbol{\gamma}\boldsymbol{\Gamma}$ and $\boldsymbol{\Gamma}\boldsymbol{\gamma}$,

$$(\boldsymbol{\gamma}\boldsymbol{\Gamma})^{ij}_{rs}\overset{\text{def}}{=}\sum_m \boldsymbol{\gamma}^m_s\,\boldsymbol{\Gamma}^{ij}_{rm}, \tag{4.25}$$

$$(\boldsymbol{\Gamma}\boldsymbol{\gamma})^{ij}_{rs}\overset{\text{def}}{=}\sum_m \boldsymbol{\Gamma}^{im}_{rs}\,\boldsymbol{\gamma}^j_m.$$

If we view $\boldsymbol{\Gamma}$ as a matrix of matrices, then these tensors result from either right- or left-multiplication of each block of $\boldsymbol{\Gamma}$ by the matrix $\boldsymbol{\gamma}$. Note that $\boldsymbol{\Gamma}\boldsymbol{\gamma}$ is antisymmetric in its lower indices only, $\boldsymbol{\gamma}\boldsymbol{\Gamma}$ is antisymmetric only in its upper indices, and $(\boldsymbol{\gamma}\boldsymbol{\Gamma})^\dagger=\boldsymbol{\Gamma}\boldsymbol{\gamma}$. Retaining from the preceding proof the notation I_{p+q} and J_{p+q} to denote ordered $(p+q)$-tuples that provide a carrier space for $\mathbb{S}_{p+q}$, we further define

$$[\underline{\mathbf{AB}}]^{I_{p+q}}_{J_{p+q}}\overset{\text{def}}{=}A^{i_1,\ldots,i_p}_{j_1,\ldots,j_p}B^{i_{p+1},\ldots,i_q}_{j_{p+1},\ldots,j_q} \tag{4.26}$$

for tensors $\mathbf{A}=(A^{i_1,\ldots,i_p}_{j_1,\ldots,j_p})$ and $\mathbf{B}=(B^{i_1,\ldots,i_q}_{j_1,\ldots,j_q})$. Finally,

$$[\underline{\boldsymbol{\gamma}^{(p)}}]^{I_p}_{J_p}\overset{\text{def}}{=}\underbrace{\boldsymbol{\gamma}^{i_1}_{j_1}\cdots\boldsymbol{\gamma}^{i_p}_{j_p}}_{p\ \text{factors}}. \tag{4.27}$$

Lemma. Let $\mathbf{A}=(A^{i_1,\ldots,i_p}_{j_1,\ldots,j_p})$ and put

$$\mathbf{T}=\sum_{\sigma,\pi\in\mathbb{S}_{p+2}}\epsilon(\sigma)\,\epsilon(\pi)\,[\underline{\mathbf{A}\boldsymbol{\Gamma}}]^{\sigma(I_{p+2})}_{\pi(J_{p+2})}$$

in which $\boldsymbol{\Gamma}$, as always, is antisymmetric. Let $\vartheta_1=(i_{p+1},i_{p+2})$ and $\vartheta_2=(j_{p+1},j_{p+2})$ be transpositions. Then

$$\vartheta_1\mathbf{T}=\mathbf{T}=\vartheta_2\mathbf{T}.$$

Proof. For any $\sigma\in\mathbb{S}_{p+2}$, $\vartheta_1\sigma(I_{p+2})=\{\sigma'(I_p),\sigma(i_{p+2}),\sigma(i_{p+1})\}$ for some $\sigma'\in\mathbb{S}_p$. Clearly $\epsilon(\vartheta_1\sigma)=-\epsilon(\sigma)$, so for any pair of permutations $\sigma,\pi\in\mathbb{S}_{p+2}$,

$$\epsilon(\vartheta_1\sigma)\,\epsilon(\pi)\,[\underline{\mathbf{A}\boldsymbol{\Gamma}}]^{\vartheta_1\sigma(I_{p+2})}_{\pi(J_{p+2})}$$
$$=-\epsilon(\sigma)\,\epsilon(\pi)\,A^{\sigma'(I_p)}_{\pi'(J_p)}\,\boldsymbol{\Gamma}^{\sigma(i_{p+2}),\sigma(i_{p+1})}_{\pi(j_{p+1}),\pi(j_{p+2})}$$
$$=\epsilon(\sigma)\,\epsilon(\pi)[\underline{\mathbf{A}\boldsymbol{\Gamma}}]^{\sigma(I_{p+2})}_{\pi(J_{p+2})}. \tag{4.28}$$

The last equality follows since $\boldsymbol{\Gamma}$ is antisymmetric. Summing Eq. (4.28) over all $\sigma,\pi\in\mathbb{S}_{p+2}$, we obtain $\vartheta_1\mathbf{T}=\mathbf{T}$. The remaining claim is similarly dispatched. ■

We can now establish the following.

Theorem 2. For $p\geqslant 2$,

$$\mathrm{tr}_{p+2}(\boldsymbol{\gamma}^{\wedge p}\wedge\boldsymbol{\Gamma})=[Np\,(\boldsymbol{\gamma}^{\wedge(p-1)}\wedge\boldsymbol{\Gamma})-p\,(p-1)(\boldsymbol{\gamma}^{\wedge(p-2)}$$
$$\wedge\boldsymbol{\Gamma}\wedge\boldsymbol{\gamma}^2)+2(N-1)(\boldsymbol{\gamma}^{\wedge(p+1)})$$
$$-2p\,(\boldsymbol{\gamma}^{\wedge(p-1)}\wedge\boldsymbol{\gamma}\boldsymbol{\Gamma})$$
$$-2p\,(\boldsymbol{\gamma}^{\wedge(p-1)}\wedge\boldsymbol{\Gamma}\boldsymbol{\gamma})]/(p+2)^2$$

with $N=\mathrm{tr}\,\boldsymbol{\gamma}$.

Example 3. For $p=2$ Theorem 2 reads

$$\mathrm{tr}_4(\boldsymbol{\gamma}\wedge\boldsymbol{\gamma}\wedge\boldsymbol{\Gamma})=\frac{1}{8}[N(\boldsymbol{\gamma}\wedge\boldsymbol{\Gamma})-(\boldsymbol{\gamma}^2\wedge\boldsymbol{\Gamma})$$
$$+(N-1)(\boldsymbol{\gamma}\wedge\boldsymbol{\gamma}\wedge\boldsymbol{\gamma})-2\,\boldsymbol{\gamma}\wedge(\boldsymbol{\gamma}\boldsymbol{\Gamma}+\boldsymbol{\Gamma}\boldsymbol{\gamma})]. \tag{4.29}$$

Example 4. On the right side of the equality in Theorem 2, the symbol $\wedge$ stands in for the the antisymmetrizer $\hat{\mathcal{A}}_{p+1}\otimes\hat{\mathcal{A}}_{p+1}$ [see Eq. 3.1], since $\mathrm{tr}_{p+2}(\boldsymbol{\gamma}^{\wedge p}\wedge\boldsymbol{\Gamma})$ is a $(p+1)$-particle tensor. In fact, the proof of Theorem 2 presented below does not require $p>1$, but for $p=1$ we must replace $\boldsymbol{\gamma}\boldsymbol{\Gamma}$ and $\boldsymbol{\Gamma}\boldsymbol{\gamma}$ by $(\hat{\mathcal{A}}_2\otimes\hat{\mathcal{A}}_2)\boldsymbol{\gamma}\boldsymbol{\Gamma}$ and $(\hat{\mathcal{A}}_2\otimes\hat{\mathcal{A}}_2)\boldsymbol{\Gamma}\boldsymbol{\gamma}$, since in this case there is no place to put the $\wedge$ symbol. Otherwise, the result stated in Theorem 2 holds for $p=1$ and is given explicitly by

$$\mathrm{tr}_3(\boldsymbol{\gamma}\wedge\boldsymbol{\Gamma})=\frac{1}{9}[N\boldsymbol{\Gamma}+2(N-1)(\boldsymbol{\gamma}\wedge\boldsymbol{\gamma})$$
$$-2(\hat{\mathcal{A}}_2\otimes\hat{\mathcal{A}}_2)(\boldsymbol{\gamma}\boldsymbol{\Gamma}+\boldsymbol{\Gamma}\boldsymbol{\gamma})]. \tag{4.30}$$

Proof of Theorem 2. The tensor $(\boldsymbol{\gamma}^{\wedge p}\wedge\boldsymbol{\Gamma})$ has elements

$$(\boldsymbol{\gamma}^{\wedge p}\wedge\boldsymbol{\Gamma})^{I_{p+2}}_{J_{p+2}}=[(p+2)!]^{-2}$$
$$\times\sum_{\sigma,\pi\in\mathbb{S}_{p+2}}\epsilon(\sigma)\,\epsilon(\pi)\,[\underline{\boldsymbol{\gamma}\boldsymbol{\Gamma}}]^{\sigma(I_{p+2})}_{\pi(J_{p+2})}. \tag{4.31}$$

Since i_{p+2} and j_{p+2} will become summation indices when we apply tr_{p+2}, we partition the above sum according to where σ and π permute these two indices:

$$(\boldsymbol{\gamma}^{\wedge p}\wedge\boldsymbol{\Gamma})^{I_{p+2}}_{J_{p+2}}=[(p+2)!]^{-2}\Bigg[(T^{\Gamma}_{\Gamma})^{I_{p+2}}_{J_{p+2}}+(T^{\gamma}_{\Gamma})^{I_{p+2}}_{J_{p+2}}+(T^{\Gamma}_{\gamma})^{I_{p+2}}_{J_{p+2}}+\sum_{r,s=1}^{p}(T_{rs})^{I_{p+2}}_{J_{p+2}}\Bigg]. \tag{4.32}$$

The notation in Eq. (4.32) means the following. For $r,s\leqslant p$, $(T_{rs})^{I_{p+2}}_{J_{p+2}}$ includes the permutations in Eq. (4.31) for which $\sigma(r)=p+2=\pi(s)$. That is, the indices i_{p+2} and j_{p+2} both appear in $[\underline{\boldsymbol{\gamma}}]^{\sigma(I_p)}_{\pi(J_p)}$. The remaining terms in Eq. (4.32) are defined as

$$(T^{\Gamma}_{\Gamma})^{I_{p+2}}_{J_{p+2}}\overset{\text{def}}{=}\sum_{r,s=p+1}^{p+2}\ \sideset{}{'}\sum_{\substack{\sigma,\pi\in\mathbb{S}_{p+2}\\ \sigma(r)=p+2=\pi(s)}}\epsilon(\sigma)\epsilon(\pi)[\underline{\boldsymbol{\gamma}^{(p)}\boldsymbol{\Gamma}}]^{\sigma(I_{p+2})}_{\pi(J_{p+2})}, \tag{4.33a}$$

$$(T^{\gamma}_{\Gamma})^{I_{p+2}}_{J_{p+2}}\overset{\text{def}}{=}\sum_{r=1}^{p}\ \sum_{s=p+1}^{p+2}\ \sideset{}{'}\sum_{\substack{\sigma,\pi\in\mathbb{S}_{p+2}\\ \sigma(r)=p+2=\pi(s)}}\epsilon(\sigma)\epsilon(\pi)[\underline{\boldsymbol{\gamma}^{(p)}\boldsymbol{\Gamma}}]^{\sigma(I_{p+2})}_{\pi(J_{p+2})}, \tag{4.33b}$$

and

$$(T^{\Gamma}_{\gamma})^{I_{p+2}}_{J_{p+2}}\overset{\text{def}}{=}\sum_{r=p+1}^{p+2}\ \sum_{s=1}^{p}\ \sideset{}{'}\sum_{\substack{\sigma,\pi\in\mathbb{S}_{p+2}\\ \sigma(r)=p+2=\pi(s)}}\epsilon(\sigma)\epsilon(\pi)[\underline{\boldsymbol{\gamma}^{(p)}\boldsymbol{\Gamma}}]^{\sigma(I_{p+2})}_{\pi(J_{p+2})}. \tag{4.33c}$$

The following mnemonic is helpful: the superscript "Γ" and subscript and "γ" in T^{Γ}_{γ} indicate that T^{Γ}_{γ} includes all permutations for which i_{p+2} is an index of $\boldsymbol{\Gamma}$ and j_{p+2} indexes $\boldsymbol{\gamma}$.

The final term in Eq. (4.32), $\Sigma_{r,s}(T_{rs})^{I_{p+2}}_{J_{p+2}}$, may itself be partitioned as in the proof of Theorem 1 [cf. Eq. (4.11)]. Its one-particle trace is then evaluated by the same techniques used to establish Theorem 1, and the result is the first two terms in the statement of Theorem 2.

Now consider $(T^{\Gamma}_{\Gamma})^{I_{p+2}}_{J_{p+2}}$. The sums over r and s in Eq. (4.33a) give rise to four terms that are related to one another by the transpositions (i_{p+1},i_{p+2}) and (j_{p+1},j_{p+2}). According to our lemma (with $\mathbf{A}=[\underline{\boldsymbol{\gamma}^{(p)}}]$), all four terms are thus identical so we have

$$\begin{aligned}
[(p+2)!]^{-2}(\mathrm{tr}_{p+2}T^{\Gamma}_{\Gamma})^{I_{p+1}}_{J_{p+1}}&=4[(p+2)!]^{-2}\sum_{k}\ \sideset{}{'}\sum_{\substack{\sigma,\pi\in\mathbb{S}_{p+2}\\ \sigma(r)=p+2=\pi(s)}}\epsilon(\sigma)\epsilon(\pi)[\underline{\boldsymbol{\gamma}^{(p)}}]^{\sigma(I_p)}_{\pi(J_p)}\,\boldsymbol{\Gamma}^{\sigma(i_{p+1}),k}_{\pi(j_{p+1}),k}\\
&=2[(N-1)/(p+2)!^{2}]\sum_{\sigma',\pi'\in\mathbb{S}_{p+1}}\epsilon(\sigma')\epsilon(\pi')[\underline{\boldsymbol{\gamma}^{(p+1)}}]^{\sigma'(I_{p+1})}_{\pi'(J_{p+1})}\\
&=2[(N-1)/(p+2)^{2}]\boldsymbol{\gamma}^{\wedge(p+1)}.
\end{aligned} \tag{4.34}$$

In the second line we have used $\mathrm{tr}_2\boldsymbol{\Gamma}=[(N-1)/2]\boldsymbol{\gamma}$ and in the third line we introduce a factor of $[(p+1)!]^2$ to account for the normalization of the resulting Grassmann product. Equation (4.34) is the third term in the formula for $\mathrm{tr}_{p+2}(\boldsymbol{\gamma}^{\wedge p}\wedge\boldsymbol{\Gamma})$.

By definition, $(T^{\gamma}_{\Gamma})^{I_{p+2}}_{J_{p+2}}$ is a sum over permutations $\sigma,\pi\in\mathbb{S}_{p+2}$ such that $\sigma(i_r)=i_{p+2}$ for some $r\leqslant p$ and $\pi(j_s)=j_{p+2}$ for $s=p+1$ or $s=p+2$. For such permutations, consider the action of $\tau^{(p+2)}_r\in\mathbb{S}_{p+2}$ as defined in Eq. (4.12):

$$\begin{aligned}
\tau^{(p+2)}_r\,\sigma(I_{p+2})&=\{\sigma'(I^{(p+1)}_r),\sigma(i_r)\}\\
&=\{\sigma'(I^{(p-1)}_r),\sigma'(i_{p+1}),\sigma'(i_{p+2}),\sigma(i_r)\}
\end{aligned} \tag{4.35}$$

for $r\leqslant p$. As usual, $\sigma'\in\mathbb{S}_{p+1}$ orders $I^{(p+1)}_r$ in the same way that σ orders the corresponding indices of I_{p+2}. We can write an analogous equation for $\pi\in\mathbb{S}_{p+2}$, and it should be obvious by now that $\epsilon(\sigma)\ \epsilon(\pi)=\epsilon(\sigma')\ \epsilon(\pi')$. Applying the transposition $(p+1,p+2)$ of the last two indices to the aforementioned analog of Eq. (4.35) we obtain

$$\begin{aligned}(p+1,p+2)\;\tau_r^{(p+2)}\;\pi(J_{p+2})\\ =\{\pi'(J_r^{(p-1)}),\pi'(j_{p+1}),\pi(j_r),\pi'(j_{p+2})\},\end{aligned}\tag{4.36}$$

with $\pi' \in \mathbb{S}_{p+1}$.

We invoke the lemma once again to show, in Eq. (4.33b), that the two terms in the sum over s are identical. Using Eq. (4.35) and the analogous equation for $\pi \in \mathbb{S}_{p+2}$, we then have

$$\begin{aligned}(T_\Gamma^\gamma)_{J_{p+2}}^{I_{p+2}}=2\sum_{r=1}^{p}\;\sideset{}{'}\sum_{\substack{\sigma,\pi\in\mathbb{S}_{p+2}\\ \sigma(r)=p+2=\pi(p+2)}}\epsilon(\sigma')\,\epsilon(\pi')\\ \times[\underline{\boldsymbol{\gamma}^{(p-1)}}]_{\pi'(J_r^{(p-1)})}^{\sigma'(I_r^{(p-1)})}\boldsymbol{\Gamma}_{\pi'(j_{p+1}),\pi'(j_{p+2})}^{\sigma'(i_{p+1}),\sigma'(i_{p+2})}\;\boldsymbol{\gamma}_{\pi(j_r)}^{\sigma(i_r)}.\end{aligned}\tag{4.37}$$

It follows from Eq. (4.36),

$$\begin{aligned}(\mathrm{tr}_{p+2}T_\Gamma^\Gamma)_{J_{p+2}}^{I_{p+2}}=-2\sum_{r=1}^{p}\;\sideset{}{'}\sum_{\substack{\sigma,\pi\in\mathbb{S}_{p+2}\\ \sigma(r)=p+2=\pi(p+2)}}\epsilon(\sigma')\epsilon(\pi')\\ \times[\underline{\boldsymbol{\gamma}^{(p-1)}}]_{\pi'(J_r^{(p-1)})}^{\sigma'(I_r^{(p-1)})}(\boldsymbol{\gamma}\boldsymbol{\Gamma})_{\pi'(j_{p+1}),\pi(j_r)}^{\sigma'(i_{p+1}),\sigma'(i_{p+2})}.\end{aligned}\tag{4.38}$$

The sum over r is performed trivially, since the summand is the same for each r. Thus

$$(\mathrm{tr}_{p+2}T_\Gamma^\Gamma)_{J_{p+2}}^{I_{p+2}}=-2p(p+1)!^2[\boldsymbol{\gamma}^{\wedge(p-1)}\wedge(\boldsymbol{\gamma}\boldsymbol{\Gamma})]_{J_{p+1}}^{I_{p+1}}.\tag{4.39}$$

The factor of $(p+1)!^2$ cancels the corresponding factor introduced by the Grassmann product. We pick up a factor of $(p+2)!^{-2}$ from Eq. (4.32), and the net result is the fourth term in the statement of Theorem 2.

The final term in $\mathrm{tr}_{p+2}(\boldsymbol{\gamma}^{\wedge p}\wedge\boldsymbol{\Gamma})$ arises from T_γ^Γ in Eq. (4.32), but its evaluation is quite similar to that of T_Γ^γ and is omitted. ■

Many other variations on this theme are possible. We present two additional results without proof, as the proofs are quite similar to the preceding two.

Theorem 3. For any $p \geqslant 2$,

$$\begin{aligned}\mathrm{tr}_{2p}(\boldsymbol{\Gamma}^{\wedge p})=[(N-1)/2p](\boldsymbol{\gamma}\wedge\boldsymbol{\Gamma}^{\wedge(p-1)})\\ -(1/p)(\hat{\mathcal{A}}_{2p-1}\otimes\hat{\mathcal{A}}_{2p-1})\mathcal{J}^{(p)},\end{aligned}$$

where $\mathcal{J}^{(p)}$ is the tensor with elements

$$(\mathcal{J}^{(p)})_{j_1,\ldots,j_{2p-1}}^{i_1,\ldots,i_{2p-1}}=\left(\sum_m \boldsymbol{\Gamma}_{j_1,j_3}^{i_1,m}\boldsymbol{\Gamma}_{j_2,m}^{i_2,i_3}\right)\boldsymbol{\Gamma}_{j_4,j_5}^{i_4,i_5}\cdots\boldsymbol{\Gamma}_{j_{2p-2},j_{2p-1}}^{i_{2p-2},i_{2p-1}}.$$

In this paper we shall only require the case $p=2$; the elements of $\mathcal{J}^{(2)}$ are given explicitly in the next section. The final result of this section is a generalization of Theorem 2:

Theorem 4. For $p \geqslant 2$ and $k \geqslant 1$,

$$\begin{aligned}\mathrm{tr}_{p+2}[(\boldsymbol{\gamma}^k)^{\wedge p}\wedge\boldsymbol{\Gamma}]=\{p\,(\mathrm{tr}\,\boldsymbol{\gamma}^k)[\boldsymbol{\gamma}^{\wedge(p-1)}\wedge\boldsymbol{\Gamma}]-p\,(p-1)\\ \times[(\boldsymbol{\gamma}^k)^{\wedge(p-2)}\wedge\boldsymbol{\Gamma}\wedge\boldsymbol{\gamma}^{2k}]+2(N-1)\\ \times[\boldsymbol{\gamma}\wedge(\boldsymbol{\gamma}^k)^{\wedge p}]-2p\,[(\boldsymbol{\gamma}^k)^{\wedge(p-1)}\\ \wedge(\boldsymbol{\gamma}^k\boldsymbol{\Gamma}+\boldsymbol{\Gamma}\boldsymbol{\gamma}^k)]\}/(p+2)^2.\end{aligned}$$

V. CONTRACTION OF $\boldsymbol{\Gamma}_{\mathrm{G}}^{(4)}$

Applying the theorems in the previous section to $\boldsymbol{\Gamma}_{\mathrm{G}}^{(4)}$, one readily obtains

$$\begin{aligned}{}_3^4\!\downarrow\boldsymbol{\Gamma}_{\mathrm{G}}^{(4)}\\ =\frac{1}{2}\{[2a(N-1)+bN](\boldsymbol{\gamma}\wedge\boldsymbol{\Gamma})-b(\boldsymbol{\gamma}^2\wedge\boldsymbol{\Gamma})\\ +[b(N-1)+2cN](\boldsymbol{\gamma}\wedge\boldsymbol{\gamma}\wedge\boldsymbol{\gamma})-6c(\boldsymbol{\gamma}\wedge\boldsymbol{\gamma}\wedge\boldsymbol{\gamma}^2)\\ -2b[\boldsymbol{\gamma}\wedge(\boldsymbol{\gamma}\boldsymbol{\Gamma}+\boldsymbol{\Gamma}\boldsymbol{\gamma})]-4a(\hat{\mathcal{A}}_3\otimes\hat{\mathcal{A}}_3)\mathcal{J}^{(2)}\}/(N-3).\end{aligned}\tag{5.1}$$

$\mathcal{J}^{(2)}$ in Eq. (5.1) is the tensor defined in Theorem 3 of the previous section. Let $\boldsymbol{\Gamma}_r^i=(\boldsymbol{\Gamma}_{rs}^{ij})$ denote the matrix comprising the (i,r)th block of $\boldsymbol{\Gamma}$. Then the tensor element $(\mathcal{J}^{(2)})_{rst}^{ijk}$ is the (k,t)th entry of the product matrix $(\boldsymbol{\Gamma}_r^i)(\boldsymbol{\Gamma}_s^j)$,

$$(\mathcal{J}^{(2)})_{rst}^{ijk}=[(\boldsymbol{\Gamma}_r^i)(\boldsymbol{\Gamma}_s^j)]_t^k=\sum_m \boldsymbol{\Gamma}_{rt}^{im}\,\boldsymbol{\Gamma}_{sm}^{jk}.\tag{5.2}$$

We also define a tensor $\mathcal{J}$ whose elements are traces of such matrix products:

$$\mathcal{J}_{rs}^{ij}\overset{\mathrm{def}}{=}\mathrm{tr}[(\boldsymbol{\Gamma}_r^i)(\boldsymbol{\Gamma}_s^j)]=\mathcal{J}_{sr}^{ji}.\tag{5.3}$$

For tensors $\mathbf{A}=(A_{j_1,\ldots,j_n}^{i_1,\ldots,i_n})$ and $\mathbf{B}=(B_{j_1,\ldots,j_m}^{i_1,\ldots,i_m})$, define a tensor $\mathbf{A}*\mathbf{B}$ with elements

$$(\mathbf{A}*\mathbf{B})_{j_1,\ldots,j_m}^{i_1,\ldots,i_m}\overset{\mathrm{def}}{=}\sum_{k_1,\ldots,k_n}A_{j_1,\ldots,j_n}^{k_1,\ldots,k_n}B_{k_1,\ldots,k_n,j_{n+1},\ldots,j_m}^{i_1,\ldots,i_n,i_{n+1},\ldots,i_m},\tag{5.4a}$$

if $n<m$, and

$$(\mathbf{A}*\mathbf{B})_{j_1,\ldots,j_m}^{i_1,\ldots,i_m}\overset{\mathrm{def}}{=}\sum_{k_1,\ldots,k_m}A_{j_1,\ldots,j_m,j_{m+1},\ldots,j_n}^{k_1,\ldots,k_m,i_{m+1},\ldots,i_n}B_{k_1,\ldots,k_m}^{i_1,\ldots,i_m},\tag{5.4b}$$

if $n \geqslant m$. (If $n=m$, $\mathbf{A}*\mathbf{B}=\mathbf{AB}$.) Using this notation and the results of the previous section one can show that

$$
\begin{aligned}
{}^{4}_{2}{\downarrow}\boldsymbol{\Gamma}_{\mathrm{G}}^{(4)} = \Bigg(& \frac{1}{6}[2aN(N-1)+b(N^2-\mathrm{tr}\,\boldsymbol{\gamma}^2)]\boldsymbol{\Gamma}+\frac{2}{3}a\boldsymbol{\Gamma}^2-\frac{4}{3}b(\boldsymbol{\gamma} \\
& \wedge \mathrm{tr}_2\,\boldsymbol{\gamma}\boldsymbol{\Gamma})-2[b(N-1)+2cN](\boldsymbol{\gamma}\wedge\boldsymbol{\gamma}^2) \\
& +\frac{1}{6}[4a(N-1)^2+5bN(N-1)+6c(N^2-\mathrm{tr}\,\boldsymbol{\gamma}^2)] \\
& \times(\boldsymbol{\gamma}\wedge\boldsymbol{\gamma})+4c(\boldsymbol{\gamma}\wedge\boldsymbol{\gamma}^3)+2c(\boldsymbol{\gamma}^2\wedge\boldsymbol{\gamma}^2) \\
& +(\hat{\mathcal{A}}_2\otimes\hat{\mathcal{A}}_2)\bigg\{\frac{4}{3}b\,(\boldsymbol{\gamma}\boldsymbol{\Gamma}\boldsymbol{\gamma})-\frac{2}{3}[2a(N-1)+bN](\boldsymbol{\gamma}\boldsymbol{\Gamma} \\
& +\boldsymbol{\Gamma}\boldsymbol{\gamma})+\frac{2}{3}b(\boldsymbol{\gamma}^2\boldsymbol{\Gamma}+\boldsymbol{\Gamma}\boldsymbol{\gamma}^2)+\frac{1}{3}b(\boldsymbol{\gamma}*(\boldsymbol{\gamma}\boldsymbol{\Gamma})+(\boldsymbol{\Gamma}\boldsymbol{\gamma})*\boldsymbol{\gamma}) \\
& -\frac{8}{3}a\,\mathcal{J}\bigg\}\Bigg) \Bigg/ [(N-3)(N-2)].
\end{aligned}
\tag{5.5}
$$

Finally, we contract once again to obtain the reconstructed 1-matrix,

$$
\begin{aligned}
{}^{4}_{1}{\downarrow}\boldsymbol{\Gamma}_{\mathrm{G}}^{(4)} = \Bigg\{ & 4a\,\mathrm{tr}_2(\boldsymbol{\Gamma}^2)+\bigg[aN(N-1)^2+b(N-1) \\
& \times\left(N^2-\frac{3}{2}\mathrm{tr}\,\boldsymbol{\gamma}^2\right)+c(N^3-3N\,\mathrm{tr}\,\boldsymbol{\gamma}^2+2\,\mathrm{tr}\,\boldsymbol{\gamma}^3)\bigg]\boldsymbol{\gamma} \\
& -\bigg[2a(N-1)^2+\frac{15}{6}bN(N-1) \\
& +3c(N^2-\mathrm{tr}\,\boldsymbol{\gamma}^2)\bigg]\boldsymbol{\gamma}^2+3[b(N-1)+2cN]\boldsymbol{\gamma}^3 \\
& -6c\boldsymbol{\gamma}^4-\frac{2}{3}b\mathbf{G}-2[2a(N-1)+bN]\,\mathrm{tr}_2(\boldsymbol{\gamma}\boldsymbol{\Gamma}) \\
& +2b\,\mathrm{tr}_2(\boldsymbol{\gamma}^2\boldsymbol{\Gamma})+\frac{4}{3}b[\boldsymbol{\gamma}\cdot(\mathrm{tr}_2\,\boldsymbol{\gamma}\boldsymbol{\Gamma}) \\
& +(\mathrm{tr}_2\,\boldsymbol{\gamma}\boldsymbol{\Gamma})\cdot\boldsymbol{\gamma}]\Bigg\} \Bigg/ [(N-3)(N-2)(N-1)],
\end{aligned}
\tag{5.6}
$$

where **G** has elements

$$
\mathbf{G}^{i}_{r} \overset{\mathrm{def}}{=} \sum_{k}\,[(\boldsymbol{\gamma}\boldsymbol{\Gamma}\boldsymbol{\gamma})^{ik}_{kr}+(\boldsymbol{\gamma}\boldsymbol{\Gamma}\boldsymbol{\gamma})^{ki}_{rk}].
\tag{5.7}
$$

Although the reconstruction functional $\boldsymbol{\Gamma}_{\mathrm{G}}^{(4)}$ ostensibly contains three adjustable parameters, as a result of normalization only two of these are independent. The normalization is obtained as a function of *a*, *b*, and *c* by contracting Eq. (5.6) once more. The normalization thus obtained is

$$
\begin{aligned}
\mathrm{tr}\,\boldsymbol{\Gamma}_{\mathrm{G}}^{(4)} = {} & \frac{a}{24}[N^2(N-1)^2-4(N-1)^2\,\mathrm{tr}\,\boldsymbol{\gamma}^2+4\,\mathrm{tr}\,(\boldsymbol{\Gamma}^2)] \\
& +\frac{c}{24}[N^4-6N^2\,\mathrm{tr}\,\boldsymbol{\gamma}^2+8N\,\mathrm{tr}\,\boldsymbol{\gamma}^3+3(\mathrm{tr}\,\boldsymbol{\gamma}^2)^2-6\,\mathrm{tr}\,\boldsymbol{\gamma}^4] \\
& +\frac{b}{24}\bigg[N^3(N-1)-5N(N-1)\,\mathrm{tr}\,\boldsymbol{\gamma}^2+4(N-1)\mathrm{tr}\,\boldsymbol{\gamma}^3 \\
& +4\sum_{klmn}\,\boldsymbol{\gamma}^m_n\boldsymbol{\gamma}^k_l\,\boldsymbol{\Gamma}^{nl}_{mk}\bigg].
\end{aligned}
\tag{5.8}
$$

Setting $\mathrm{tr}\,\boldsymbol{\Gamma}_{\mathrm{G}}^{(4)}=\binom{N}{4}$ fixes one of the parameters in terms of the other two.

Natural spin orbitals

Up to this point the spin-orbital basis $\{\phi_k\}$ has been orthonormal but otherwise arbitrary. Now let us introduce the basis of "natural" spin orbitals (NSOs) that diagonalize $\boldsymbol{\gamma}$ and put $\boldsymbol{\gamma}^i_j=\lambda_j\delta_{ij}$. For an exact 1-RDM $\boldsymbol{\gamma}$, it follows that $0\leqslant\lambda_k\leqslant 1$ for each k [1].

The expressions for the reconstructed 1- and 2-RDMs, ${}^{4}_{2}{\downarrow}\boldsymbol{\Gamma}_{\mathrm{G}}^{(4)}$ and ${}^{4}_{1}{\downarrow}\boldsymbol{\Gamma}_{\mathrm{G}}^{(4)}$, are especially simple in the NSO basis. In particular,

$$
[(\hat{\mathcal{A}}_2\otimes\hat{\mathcal{A}}_2)\,\boldsymbol{\gamma}\boldsymbol{\Gamma}\boldsymbol{\gamma}]^{ij}_{rs} \overset{\mathrm{NSO}}{=} \frac{1}{4}(\lambda_i+\lambda_j)(\lambda_r+\lambda_s)\boldsymbol{\Gamma}^{ij}_{rs},
\tag{5.9a}
$$

$$
[(\hat{\mathcal{A}}_2\otimes\hat{\mathcal{A}}_2)(\boldsymbol{\gamma}^n\boldsymbol{\Gamma}+\boldsymbol{\Gamma}\boldsymbol{\gamma}^n)]^{ij}_{rs} \overset{\mathrm{NSO}}{=} \frac{1}{2}(\lambda^n_i+\lambda^n_j+\lambda^n_r+\lambda^n_s)\boldsymbol{\Gamma}^{ij}_{rs},
\tag{5.9b}
$$

$$
[(\hat{\mathcal{A}}_2\otimes\hat{\mathcal{A}}_2)(\boldsymbol{\gamma}*(\boldsymbol{\gamma}\boldsymbol{\Gamma})+(\boldsymbol{\Gamma}\boldsymbol{\gamma})*\boldsymbol{\gamma})]^{ij}_{rs} \overset{\mathrm{NSO}}{=} (\lambda_i\lambda_j+\lambda_r\lambda_s)\boldsymbol{\Gamma}^{ij}_{rs},
\tag{5.9c}
$$

and

$$
\begin{aligned}
[\boldsymbol{\gamma}\wedge(\mathrm{tr}_2\,\boldsymbol{\gamma}\boldsymbol{\Gamma})]^{ij}_{rs} \overset{\mathrm{NSO}}{=} {} & \frac{1}{4}\lambda_r\sum_k\,\lambda_k(\delta_{ir}\boldsymbol{\Gamma}^{jk}_{sk}-\delta_{jr}\boldsymbol{\Gamma}^{ik}_{sk}) \\
& +\frac{1}{4}\lambda_s\sum_k\,\lambda_k(\delta_{js}\boldsymbol{\Gamma}^{ik}_{rk}-\delta_{is}\boldsymbol{\Gamma}^{jk}_{rk}).
\end{aligned}
\tag{5.9d}
$$

Expressions for the relevant components of ${}^{4}_{1}{\downarrow}\boldsymbol{\Gamma}_{\mathrm{G}}^{(4)}$ follow readily from these equations. One finds that, in general, the reconstructed 1-matrix may possess nonvanishing off-diagonal elements, even when the input $\boldsymbol{\gamma}={}^{2}_{1}{\downarrow}\boldsymbol{\Gamma}$ is diagonal. As such we must diagonalize ${}^{4}_{1}{\downarrow}\boldsymbol{\Gamma}_{\mathrm{G}}^{(4)}$ in order to investigate whether it violates Coleman's (ensemble) *N*-representability conditions for the 1-matrix [1]. We are currently investigating under what circumstances ${}^{4}_{2}{\downarrow}\boldsymbol{\Gamma}_{\mathrm{G}}^{(4)}$ and ${}^{4}_{1}{\downarrow}\boldsymbol{\Gamma}_{\mathrm{G}}^{(4)}$ satisfy known *N*-representability requirements [3–9].

TABLE I. Largest elements of the CI and reconstructed 2-RDMs in the NSO basis, for the ground state of Be. None of the reconstructions is properly normalized: tr$({}^{4}_{2}{\downarrow}\mathbf{\Gamma}^{(4)}_{\mathrm{Val}})=6.164$, tr$({}^{4}_{2}{\downarrow}\mathbf{\Gamma}^{(4)}_{\mathrm{G}})=9.828$ for the fit to $\boldsymbol{\gamma}_{\mathrm{CI}}$, and tr$({}^{4}_{2}{\downarrow}\mathbf{\Gamma}^{(4)}_{\mathrm{G}})=6.918$ for the fit to $\mathbf{\Gamma}_{\mathrm{CI}}$. Natural spin orbitals are labeled by the Hartree-Fock component of greatest magnitude (absolute value of coefficient >0.95 in each case). The degeneracy $|\mathbf{\Gamma}^{3s\alpha,3s\beta}_{2s\alpha,2s\beta}|\approx|\mathbf{\Gamma}^{4s\alpha,5s\beta}_{2s\alpha,2s\beta}|$ is accidental and disappears with additional significant digits.

2-RDM element				${}^{4}_{2}{\downarrow}\mathbf{\Gamma}^{(4)}_{\mathrm{G}}$ with fitted parameters *a*, *b*, and *c*			
(NSO basis)	$\mathbf{\Gamma}_{\mathrm{CI}}$	${}^{4}_{2}{\downarrow}\mathbf{\Gamma}^{(4)}_{\mathrm{Val}}$	Error	Fit to $\boldsymbol{\gamma}_{\mathrm{CI}}$	Error	Fit to $\mathbf{\Gamma}_{\mathrm{CI}}$	Error
$\mathbf{\Gamma}^{1s\alpha,1s\beta}_{1s\alpha,1s\beta}$	0.498965	0.496461	−0.002504	0.653711	0.154746	0.506778	0.007813
			−0.5%		31.0%		1.6%
$\mathbf{\Gamma}^{2s\alpha,2s\beta}_{2s\alpha,2s\beta}$	0.455695	0.416092	−0.039603	0.593240	0.137545	0.400071	−0.055624
			−8.7%		30.2%		−12.2%
$\mathbf{\Gamma}^{1s\alpha,2s\beta}_{1s\alpha,2s\beta}$	0.455013	0.416093	−0.038920	0.673567	0.218554	0.463285	0.008272
			−8.6%		48.0%		1.8%
$\mathbf{\Gamma}^{1s\alpha,2s\alpha}_{1s\alpha,2s\alpha}$	0.454976	0.338698	−0.116278	0.673633	0.218657	0.463299	0.008323
			−25.6		48.1%		1.8%
$\mathbf{\Gamma}^{3s\alpha,3s\beta}_{2s\alpha,2s\beta}$	−0.080902	−0.100940	−0.020038	0.053747	0.134649	−0.047162	0.033740
			24.8%		−166.4%		−41.7%
$\mathbf{\Gamma}^{4s\alpha,5s\beta}_{2s\alpha,2s\beta}$	0.080902	0.100940	0.020038	−0.053747	−0.134649	0.047162	−0.033740
			24.8%		−166.4%		−41.7%
$\mathbf{\Gamma}^{6s\alpha,6s\beta}_{2s\alpha,2s\beta}$	−0.018827	−0.025651	−0.006824	0.014940	0.033767	−0.011710	0.007117
			36.2%		−179.4%		−37.8%

VI. NUMERICAL EXAMPLE: Be ATOM

In this section we use Valdemoro's Grassmann product reconstruction functional $\mathbf{\Gamma}^{(4)}_{\mathrm{Val}}$ to reconstruct the 4-matrix for the ground state of atomic Be from a highly accurate input matrix $\mathbf{\Gamma}_{\mathrm{CI}}$, obtained from a configuration interaction (CI) calculation. We will evaluate the contraction consistency of $\mathbf{\Gamma}^{(4)}_{\mathrm{Val}}[\mathbf{\Gamma}_{\mathrm{CI}}]$ and compare it with two reconstruction functionals of the type $\mathbf{\Gamma}^{(4)}_{\mathrm{G}}$, where the parameters *a*, *b*, and *c* are determined by fits to the CI density matrices. Furthermore, we shall compare the three self-consistent iteration schemes discussed at the end of Sec. III.

We choose Be for this example due to the availability of high-quality ground-state wave functions for this system [35,36]. The accuracy of 3- and 4-matrices reconstructed using Valdemoro's prescription has been tested in the past [31] by comparison to full CI density matrices, but only for tiny basis sets (a few basis functions beyond the minimal basis). The calculations in this section are the first reported reconstruction in a large basis set. The CSE as a quantum-chemical methodology is still young, however, and at the present time the size of this basis set (96 spin orbitals) outstrips our ability to effect a complete, self-consistent solution to the CSE. Despite this limitation we have been able to accomplish the comparisons and evaluations outlined in the preceding paragraph.

We employ 1- and 2-RDMs obtained from Bunge's CI wave function [35]. This 180-configuration wave function is represented in a Hartree-Fock optimized basis [37] consisting of 96 *s*, *p*, and *d* Slater-type spin orbitals, and the resulting energy bound $E_{\mathrm{CI}}=-14.664\,193$ a.u. recovers 96.7% of the estimated (nonrelativistic) correlation energy of ground-state Be. The total nonrelativistic energy estimated from experiment is $-14.667\,328\pm25$ a.u. [38,39]. Although more accurate wave functions exist for this system [36], the humongous basis sets employed in these superior calculations make storage and manipulation of the 2-RDM troublesome; for our purposes the slightly less accurate wave function suffices.

${}^{4}_{2}{\downarrow}\mathbf{\Gamma}^{(4)}_{\mathrm{Val}}$ was constructed in the basis of NSOs of $\boldsymbol{\gamma}_{\mathrm{CI}}$. Relative to the Hartree-Fock basis, $\mathbf{\Gamma}_{\mathrm{CI}}$ in the NSO basis has fewer large elements, which facilitates presentation of the data. In Table I we have listed all independent elements of $\mathbf{\Gamma}_{\mathrm{CI}}$ greater than 0.015 in magnitude; juxtaposed with these are the corresponding elements of ${}^{4}_{2}{\downarrow}\mathbf{\Gamma}^{(4)}_{\mathrm{G}}$ for three choices of the parameters *a*, *b*, and *c*. ${}^{4}_{2}{\downarrow}\mathbf{\Gamma}^{(4)}_{\mathrm{Val}}$ indicates that $a=0$, $b=6$, and $c=-5$, as in Eq. (3.5). For the column labeled "fit to $\boldsymbol{\gamma}_{\mathrm{CI}}$," *a*, *b*, and *c* were chosen by least-squares fit of ${}^{4}_{1}{\downarrow}\mathbf{\Gamma}^{(4)}_{\mathrm{G}}$ to $\boldsymbol{\gamma}_{\mathrm{CI}}$, the CI 1-RDM. Similarly, "fit to $\mathbf{\Gamma}_{\mathrm{CI}}$" indicates that *a*, *b*, and *c* were chosen by fitting ${}^{4}_{2}{\downarrow}\mathbf{\Gamma}^{(4)}_{\mathrm{G}}$ to $\mathbf{\Gamma}_{\mathrm{CI}}$. The fitted parameters in both cases are listed in Table II.

Valdemoro's reconstruction reproduces the largest matrix element quite accurately but for the other elements in Table I, ${}^{4}_{2}{\downarrow}\mathbf{\Gamma}^{(4)}_{\mathrm{Val}}$ differs from $\mathbf{\Gamma}_{\mathrm{CI}}$ by 10–40%. These errors are far larger than those reported by Colmenero and Valdemoro [31] for the same system with a double-ζ basis set (six spin orbitals). However, for the elements in Table I Valdemoro's

TABLE II. Parameters employed in reconstruction of $\mathbf{\Gamma}^{(4)}_{\mathrm{G}}$. For each reconstruction, the top set of parameters has not been renormalized, while the bottom set is rescaled so that tr $\mathbf{\Gamma}^{(4)}=\binom{N}{4}=1$ for the reconstructed 4-RDM.

Approximation	*a*	*b*	*c*
$\mathbf{\Gamma}^{(4)}_{\mathrm{Val}}$ [Eq. (3.5)]	0.000000	6.000000	−5.000000
	0.000000	5.840070	−4.866725
$\mathbf{\Gamma}^{(4)}_{\mathrm{G}}$, fit to $\boldsymbol{\gamma}_{\mathrm{CI}}$	−5.470984	8.709500	2.174761
	−3.340039	5.317155	−1.327693
$\mathbf{\Gamma}^{(4)}_{\mathrm{G}}$, fit to $\mathbf{\Gamma}_{\mathrm{CI}}$	1.171816	5.353079	−3.238447
	−1.016319	4.642740	−2.808714

TABLE III. Comparison of CI and reconstructed 2-RDMs, following renormalization.

2-RDM element				${}^{4}_{2}{\downarrow}\mathbf{\Gamma}^{(4)}_{\mathrm{G}}$ with fitted parameters *a*, *b*, and *c*			
(NSO basis)	$\mathbf{\Gamma}_{\mathrm{CI}}$	${}^{4}_{2}{\downarrow}\mathbf{\Gamma}^{(4)}_{\mathrm{Val}}$	Error	Fit to $\boldsymbol{\gamma}_{\mathrm{CI}}$	Error	Fit to $\mathbf{\Gamma}_{\mathrm{CI}}$	Error
$\mathbf{\Gamma}^{1s\alpha,1s\beta}_{1s\alpha,1s\beta}$	0.498965	0.483228	-0.015467 -3.2%	0.399078	-0.099887 -20.0%	0.439531	-0.059434 -11.9%
$\mathbf{\Gamma}^{2s\alpha,2s\beta}_{2s\alpha,2s\beta}$	0.455695	0.329670	-0.126025 -27.7%	0.362162	-0.093533 -20.5%	0.346983	-0.108712 -23.9%
$\mathbf{\Gamma}^{1s\alpha,2s\beta}_{1s\alpha,2s\beta}$	0.455013	0.405002	-0.050011 -11.0%	0.411200	-0.043813 -9.6%	0.401809	-0.053204 -11.7%
$\mathbf{\Gamma}^{1s\alpha,2s\alpha}_{1s\alpha,2s\alpha}$	0.454976	0.405001	-0.049975 -11.0%	0.411240	-0.043736 -9.6%	0.401821	-0.053155 -11.7%
$\mathbf{\Gamma}^{3s\alpha,3s\beta}_{2s\alpha,2s\beta}$	-0.080902	-0.098249	-0.017347 21.4%	0.032811	0.113713 -140.6%	-0.040903	0.39999 -49.4%
$\mathbf{\Gamma}^{4s\alpha,5s\beta}_{2s\alpha,2s\beta}$	0.080902	0.098249	0.017347 21.4%	-0.032811	-0.113713 -140.6%	0.040903	-0.39999 -49.4%
$\mathbf{\Gamma}^{6s\alpha,6s\beta}_{2s\alpha,2s\beta}$	-0.018827	-0.024967	-0.006140 32.6%	0.009120	0.027947 -148.4%	-0.010156	0.008671 -46.1%

approximation is no worse than the fit of ${}^{4}_{2}{\downarrow}\mathbf{\Gamma}^{(4)}_{\mathrm{G}}$ to $\mathbf{\Gamma}_{\mathrm{CI}}$. Of course, such a fit is not a useful paradigm for solving the CSE but does provide a benchmark against which we may compare other reconstruction functionals of the Grassmann product form.

Each of the three aforementioned reconstructions overestimates the trace of the reconstructed 2-RDM, so we also compared the RDMs obtained with rescaled parameters, such that $\mathrm{tr}\,\mathbf{\Gamma}^{(4)}_{\mathrm{G}}=\binom{N}{4}$ in each case. The rescaled parameters are also listed in Table II, and in Table III we compare the matrix elements of ${}^{4}_{2}{\downarrow}\mathbf{\Gamma}^{(4)}_{\mathrm{G}}$ for these renormalized reconstructions. It is understandable why rescaling worsens the original fits, but notable that upon renormalization Valdemoro's reconstructed matrix elements also show greater deviation from the CI values. The energy calculated using ${}^{4}_{2}{\downarrow}\mathbf{\Gamma}^{(4)}_{\mathrm{Val}}$ also becomes less accurate when we impose proper normalization, increasing from $E=-14.4813$ a.u. (not normalized) to $E=-14.0953$ a.u. (normalized). In contrast, the energy obtained from the fit to $\mathbf{\Gamma}_{\mathrm{CI}}$ is significantly worse when we do *not* impose proper normalization: $E=-16.1355$ a.u. (not normalized), while $E=-13.9944$ a.u. (normalized).

Since $E=\Sigma_{ijmn}\mathbf{K}^{ij}_{mn}\mathbf{\Gamma}^{mn}_{ij}$, an error $\Delta\mathbf{\Gamma}^{mn}_{ij}$ in $\mathbf{\Gamma}^{mn}_{ij}$ corresponds to an error $\Delta E^{ij}_{mn}=\mathbf{K}^{ij}_{mn}\Delta\mathbf{\Gamma}^{mn}_{ij}$ in the energy. The ΔE^{ij}_{mn} are thus an importance sampling of the errors in the reconstructed 2-matrices. In Table IV we tabulate these ΔE values for each of the largest elements of the reconstructed, renormalized 2-matrices. Again Valdemoro's approximation fares better than a simple fit to $\mathbf{\Gamma}_{\mathrm{CI}}$.

We next examine the eigenvalues of the reconstructed, renormalized matrices ${}^{4}_{2}{\downarrow}\mathbf{\Gamma}^{(4)}_{\mathrm{G}}[\mathbf{\Gamma}_{\mathrm{CI}}]$ and ${}^{4}_{1}{\downarrow}\mathbf{\Gamma}^{(4)}_{\mathrm{G}}[\mathbf{\Gamma}_{\mathrm{CI}}]$. To diagonalize a two-particle matrix such as $\mathbf{\Gamma}$ it is easiest to work in a basis of two-particle functions. Thus, we construct from our orthonormal spin orbitals $\{\phi_i\}$ a set of orthonormal spin geminals $\{\Phi_{jk}\}$, where for each $j\leqslant k$

$$\Phi_{jk}(1,2)\overset{\mathrm{def}}{=}\frac{1}{\sqrt{2}}[\phi_j(1)\phi_k(2)-\phi_k(1)\phi_j(2)]. \qquad (6.1)$$

Denoting $J=(j_1,j_2)$, we may express $\mathbf{\Gamma}$ as a two-index matrix in the $\{\Phi_{jk}\}$ basis,

$$\begin{aligned}\Gamma(1,2;1',2')&=2\sum_{i<j}\sum_{r<s}\mathbf{\Gamma}^{ij}_{rs}\Phi^{*}_{ij}(1',2')\,\Phi_{rs}(1,2)\\&=\sum_{J,K=1}^{\binom{2\nu}{2}}\mathbf{\Gamma}_{JK}\Phi^{*}_{J}(1',2')\Phi_{K}(1,2). \qquad (6.2)\end{aligned}$$

Here ν is the number of orbitals, so there are 2ν spin orbitals and $\binom{2\nu}{2}$ spin geminals. Comparing Eq. (2.1) to Eq. (6.2) we see that $\mathbf{\Gamma}_{JK}=2\,\mathbf{\Gamma}^{j_1j_2}_{k_1k_2}$. We obtain the ''natural'' spin

TABLE IV. Energy differences (atomic units) for the largest magnitude elements of reconstructed, renormalized 2-RDMs in the NSO basis, for the ground state of Be. $\Delta E^{ij}_{mn}=\mathbf{K}^{ij}_{mn}\Delta\mathbf{\Gamma}^{mn}_{ij}$ (see text). The total energies calculated with these 2-matrices are $E_{\mathrm{CI}}=-14.664$ and $E_{\mathrm{Val}}=-14.095$ a.u. The fit to $\boldsymbol{\gamma}_{\mathrm{CI}}$ gives an energy of -14.209 a.u. and the fit to $\mathbf{\Gamma}_{\mathrm{CI}}$ yields an energy of -13.994 a.u.

$\mathbf{K}^{ij}_{mn}$ (NSO basis)	ΔE^{ij}_{mn} (Valdemoro)	ΔE^{ij}_{mn} Fit to ($\boldsymbol{\gamma}_{\mathrm{CI}}$)	ΔE^{ij}_{mn} Fit to ($\mathbf{\Gamma}_{\mathrm{CI}}$)
$\mathbf{K}^{1s\alpha,1s\beta}_{1s\alpha,1s\beta}=-3.015813$	-0.046646	-0.301241	-0.179242
$\mathbf{K}^{2s\alpha,2s\beta}_{2s\alpha,2s\beta}=-0.721147$	-0.090883	-0.067451	-0.078397
$\mathbf{K}^{1s\alpha,2s\beta}_{1s\alpha,2s\beta}=-2.697228$	-0.134891	-0.118174	-0.143503
$\mathbf{K}^{1s\alpha,2s\alpha}_{1s\alpha,2s\alpha}=-2.697228$	-0.134794	-0.117966	-0.143371
$\mathbf{K}^{2s\alpha,2s\beta}_{3s\alpha,3s\beta}=+0.357902$	0.006209	-0.040698	-0.014316
$\mathbf{K}^{2s\alpha,2s\beta}_{4s\alpha,5s\beta}=-0.078553$	0.001363	-0.008932	-0.003142
$\mathbf{K}^{2s\alpha,2s\beta}_{6s\alpha,6s\beta}=+0.056642$	-0.000348	0.001583	0.000491

TABLE V. Largest and smallest eigenvalues of $\mathbf{\Gamma}^{\alpha\alpha}_{\alpha\alpha}$ and their reconstructed, renormalized approximations, for the Be ground state.

		$({}^{4}_{2}{\downarrow}\mathbf{\Gamma}^{(4)}_{\mathrm{G}})^{\alpha\alpha}_{\alpha\alpha}$	
$(\mathbf{\Gamma}_{\mathrm{CI}})^{\alpha\alpha}_{\alpha\alpha}$	$({}^{4}_{2}{\downarrow}\mathbf{\Gamma}^{(4)}_{\mathrm{Val}})^{\alpha\alpha}_{\alpha\alpha}$	Fit to $\boldsymbol{\gamma}_{\mathrm{CI}}$	Fit to $\mathbf{\Gamma}_{\mathrm{CI}}$
0.910016	0.810105	0.502110	0.697023
0.028718	0.047260	0.040686	0.047736
0.028718	0.047260	0.040686	0.047736
0.028718	0.047260	0.040686	0.047736
0.001715	0.007099	0.023537	0.021982
⋮	⋮	⋮	⋮
0.000000	-0.003440[a]	-0.000076	0.000000

[a]Not listed are another 41 eigenvalues more negative than -1×10^{-6}.

geminals and their occupation numbers (eigenvalues of $\mathbf{\Gamma}$) by diagonalizing the matrix $(\mathbf{\Gamma}_{JK})$. More precisely, we diagonalize the two independent blocks of this matrix, which correspond to the spin components $\mathbf{\Gamma}^{\alpha\alpha}_{\alpha\alpha}$ and $\mathbf{\Gamma}^{\alpha\beta}_{\alpha\beta}$ (see the Appendix).

The largest and smallest eigenvalues of $\mathbf{\Gamma}$ and the reconstructed, renormalized approximations thereof are displayed in Tables V and VI; eigenvalues of the corresponding 1-matrices are presented in Table VII. For the large eigenvalues we find that Valdemoro's reconstruction is more accurate than the fitted Grassmann product functionals. Valdemoro's approximation breaks down at the other end of the eigenvalue spectrum, however: of 4560 eigenvalues, 153 are more negative than -1×10^{-6}, although none is more negative than $-0.003\,440$. The fit to $\mathbf{\Gamma}_{\mathrm{CI}}$ produces no negative eigenvalues.

Although our primary interest in this paper is the Grassmann product ansatz, as indicated in Sec. III the reconstruction functionals considered by Nakatsuji, Yasuda, and coworkers [15–17] include corrections to Valdemoro's reconstruction that are not of the Grassmann product form. Since these correction terms are second order in the correlation potential, while Valdemoro's reconstruction is first order, we expect that the numerical results presented above would improve if these correction terms were included. Unfortunately, however, computation of the Nakatsuji-Yasuda correction terms requires two additional summations over spin-orbital indices and for the large basis set considered here we are unable to carry out this calculation using our present algorithms. Nevertheless, the calculations that we have presented are significant in that for the first time the reconstruction has been carried out in a large basis set.

TABLE VI. Largest and smallest eigenvalues of $\mathbf{\Gamma}^{\alpha\beta}_{\alpha\beta}$ and reconstructed, renormalized approximations, for the Be ground state.

		$({}^{4}_{2}{\downarrow}\mathbf{\Gamma}^{(4)}_{\mathrm{G}})^{\alpha\beta}_{\alpha\beta}$	
$(\mathbf{\Gamma}_{\mathrm{CI}})^{\alpha\beta}_{\alpha\beta}$	$({}^{4}_{2}{\downarrow}\mathbf{\Gamma}^{(4)}_{\mathrm{Val}})^{\alpha\beta}_{\alpha\beta}$	Fit to $\boldsymbol{\gamma}_{\mathrm{CI}}$	Fit to $\mathbf{\Gamma}_{\mathrm{CI}}$
1.000664	0.968718	0.502110	0.762930
0.998484	0.929264	0.502016	0.697030
0.910307	0.810352	0.487271	0.697030
0.910016	0.810105	0.450850	0.634978
0.028718	0.218379	0.040686	0.133495
0.028718	0.047260	0.040686	0.047736
0.028718	0.047260	0.040686	0.047736
⋮	⋮	⋮	⋮
0.000000	-0.003440[a]	-0.131858[b]	0.000000

[a]Not listed are another 110 eigenvalues more negative than -1×10^{-6}.

[b]Not listed are another three eigenvalues more negative than -1×10^{-6}.

TABLE VII. Largest and smallest eigenvalues of the CI 1-RDM for ground-state Be, and the reconstructed, renormalized approximations thereof. Each eigenvalue corresponds to a set of exactly degenerate values arising from m_s and $|m_l|$ degeneracies. Those degeneracies appearing in the table are accidental.

		${}^{4}_{1}{\downarrow}\mathbf{\Gamma}^{(4)}_{\mathrm{G}}$	
$\boldsymbol{\gamma}_{\mathrm{CI}}$	${}^{4}_{1}{\downarrow}\mathbf{\Gamma}^{(4)}_{\mathrm{Val}}$	Fit to $\boldsymbol{\gamma}_{\mathrm{CI}}$	Fit to $\mathbf{\Gamma}_{\mathrm{CI}}$
0.998136	0.959886	0.952621	0.943006
0.911596	0.776082	0.871583	0.821229
0.028778	0.083757	0.055979	0.074902
0.028778	0.083757	0.055979	0.074902
0.001856	0.083757	0.055979	0.074902
0.000484	0.005575	0.003903	0.005039
0.000352	0.001699	0.000903	0.001414
0.000352	0.001277	0.000646	0.001043
0.000055	0.001277	0.000646	0.001043
0.000055	0.001277	0.000646	0.001043
0.000055	0.000160	0.000119	0.000148
⋮	⋮	⋮	⋮
0.000000	0.000001	0.000000	0.000000

Iteration of the CSE

Finally in this section we return to an earlier issue: the nature of the appropriate iteration algorithm for self-consistent solution of the CSE. Three possibilities were introduced in Sec. III. In the fully contraction-consistent scheme of Eq. (3.8), the approximate 2-RDM after n iterations, $\mathbf{\Gamma}_n$, is used to construct $\mathbf{\Gamma}^{(4)}_n$ (the approximate 4-RDM used in the next iteration), while the 2- and 3-RDMs employed at the $(n+1)$th step are the contractions of $\mathbf{\Gamma}^{(4)}_n$. The scheme represented by Eq. (3.9) employs separate reconstruction functionals for the 3- and 4-RDMs to construct $\mathbf{\Gamma}^{(3)}_n$ and $\mathbf{\Gamma}^{(4)}_n$ from $\mathbf{\Gamma}_n$. Lastly, according to the procedure of Eq. (3.10), $\mathbf{\Gamma}^{(4)}_n$ is reconstructed from $\mathbf{\Gamma}_n$ and $\mathbf{\Gamma}^{(3)}_n$ is obtained by contraction of $\mathbf{\Gamma}^{(4)}_n$, but the 2-RDM employed in this method is the input matrix $\mathbf{\Gamma}_n$ rather than a contraction of $\mathbf{\Gamma}^{(4)}_n$.

As indicated previously, the combination of a large basis set and poor scaling prevents us from carrying out a complete self-consistent solution to the CSE at this time. Nevertheless, we are able to carry out a single update of a small number of matrix elements (since we cannot calculate all the matrix elements, the iterative procedure cannot be extended beyond a single step). Thus, starting from $\mathbf{\Gamma}_0=\mathbf{\Gamma}_{\mathrm{CI}}$, we cal-

TABLE VIII. First iterative correction $\boldsymbol{\Gamma}_1$ to the CI 2-RDM for the ground state of Be. $\boldsymbol{\Gamma}_1$ is obtained from a single iteration of the CSE by using three different iteration schemes, as described in the text. Beneath each matrix element is listed its deviation from the accurate CI starting value, $\boldsymbol{\Gamma}_0$.

Matrix element	$\boldsymbol{\Gamma}_0$	$\boldsymbol{\Gamma}_1$ Eq. (3.8)	$\boldsymbol{\Gamma}_1$ Eq. (3.9)	$\boldsymbol{\Gamma}_1$ Eq. (3.10)
$\boldsymbol{\Gamma}^{1s\alpha,1s\beta}_{1s\alpha,1s\beta}$	0.498965	0.120794	0.496747	0.495529
		−0.378171	−0.002218	−0.003436
$\boldsymbol{\Gamma}^{2s\alpha,2s\beta}_{2s\alpha,2s\beta}$	0.455695	0.043428	0.426295	0.374638
		−0.412267	−0.029400	−0.081057
$\boldsymbol{\Gamma}^{1s\alpha,2s\beta}_{1s\alpha,2s\beta}$	0.455013	0.124256	0.451807	0.433891
		−0.330757	−0.003206	−0.021122
$\boldsymbol{\Gamma}^{1s\alpha,2s\alpha}_{1s\alpha,2s\alpha}$	0.454976	0.124185	0.451779	0.433869
		−0.330791	−0.003197	−0.021107
$\boldsymbol{\Gamma}^{3s\alpha,3s\beta}_{2s\alpha,2s\beta}$	−0.080902	−0.040809	−0.084309	−0.000960
		0.040093	−0.003407	0.079942
$\boldsymbol{\Gamma}^{4s\alpha,5s\beta}_{2s\alpha,2s\beta}$	0.080902	0.040812	0.084310	0.091503
		−0.040090	0.003408	0.010601
$\boldsymbol{\Gamma}^{6s\alpha,6s\beta}_{2s\alpha,2s\beta}$	−0.018827	−0.011907	−0.020068	−0.006087
		0.006920	−0.001241	0.012740

culate an updated value for each of the largest elements of the 2-RDM by using each of the aforementioned iteration schemes. The results are presented in Table VIII.

Among the three iteration schemes, the fully contraction-consistent scheme is comically bad, while the procedure that employs separate reconstruction functionals [Eq. (3.9)] stands apart in accuracy, with relative errors that are an order of magnitude smaller than the third iteration strategy, Eq. (3.10). Note in particular that iterated matrix elements in the second scheme lie closer to the CI values than do the elements of ${}^{4}_{2}{\downarrow}\boldsymbol{\Gamma}^{(4)}_{\mathrm{Val}}[\boldsymbol{\Gamma}_{\mathrm{CI}}]$ in Table I. Insofar as we may draw conclusions on the basis of only one iteration, the self-consistent procedures of Eqs. (3.9) and (3.10) appear to be heading in the right direction, i.e., back toward the initial 2-RDM. However, this behavior may be an artifact of the fact that, at the first iteration, Eqs. (3.9) and (3.10) employ $\boldsymbol{\Gamma}_0$, the accurate 2-RDM, while Eq. (3.8) uses the approximate 2-matrix ${}^{4}_{2}{\downarrow}\boldsymbol{\Gamma}^{(4)}_{\mathrm{G}}[\boldsymbol{\Gamma}_0]$.

VII. CONCLUSION

In this paper we have derived contraction formulas for the most general 4-RDM reconstruction functional that can be formed from a linear combination of Grassmann products of the 1-RDM $\boldsymbol{\gamma}$ and the 2-RDM $\boldsymbol{\Gamma}$. We have used accurate RDMs for Be atom to test and compare various reconstruction functionals and iteration schemes that have been proposed for solving the contracted Schrödinger equation. This represents the first test of density matrix reconstruction formulas in a large basis set.

It is known, both from the cumulant expansions pursued by Kutzelnigg and Mukherjee [33,40] and by Mazziotti [29], as well as from the Green's function expansions of Nakatsuji and Yasuda [15] and Yasuda and Nakatsuji [16], that reconstruction of $\boldsymbol{\Gamma}^{(4)}$ by Grassmann products of $\boldsymbol{\gamma}$ and $\boldsymbol{\Gamma}$ is necessarily approximate (see also Ref. [41]). These authors point out that the Grassmann products $\boldsymbol{\gamma}\wedge\boldsymbol{\gamma}\wedge\boldsymbol{\gamma}\wedge\boldsymbol{\gamma}$, $\boldsymbol{\Gamma}\wedge\boldsymbol{\gamma}\wedge\boldsymbol{\gamma}$, and $\boldsymbol{\Gamma}\wedge\boldsymbol{\Gamma}$ cannot by themselves incorporate correlations ("collision terms," in the language of diagrammatic perturbation theory) between three or more particles. It is possible, however, that the parameters a, b, and c appearing in the ansatz of Eq. (1.1) could be chosen so as to incorporate (possibly in an indirect or a semiempirical way) some of these higher-order correlation effects, much as an uncorrelated Hückel calculation can recover certain correlation effects if the parameters are chosen appropriately. It is worth pursuing whether the parameters here can be related to the aforementioned cumulant expansions.

It is also known that the Grassmann product ansatz is not contraction-consistent except in the single-determinant limit, so that in general ${}^{4}_{2}{\downarrow}\boldsymbol{\Gamma}^{(4)}_{\mathrm{G}}\neq\boldsymbol{\Gamma}$, where $\boldsymbol{\Gamma}$ and $\boldsymbol{\gamma}={}^{2}_{1}{\downarrow}\boldsymbol{\Gamma}$ are the density matrices used in the reconstruction. The Grassmann product form for $\boldsymbol{\Gamma}^{(4)}$ is thus inconsistent with a Grassmann product form for $\boldsymbol{\Gamma}^{(3)}$, as our results in Sec. V demonstrate explicitly. A related question is the stability of the contracted Schrödinger equation: if an accurate 2-RDM is introduced as the starting point, will it be returned following reconstruction and iteration of the CSE?

The 4-matrix reconstruction functional proposed by Valdemoro and co-workers [32] produces an approximate $\boldsymbol{\Gamma}^{(4)}$ that yields, upon contraction, a 2-matrix that is a good approximation to the input $\boldsymbol{\Gamma}$. For ground-state Be, this approximate 2-RDM better reproduces the large matrix elements of $\boldsymbol{\Gamma}$ than does the general Grassmann reconstruction functional optimized by least-squares fit to $\boldsymbol{\Gamma}$ itself. Moreover, Valdemoro's approximate 2-RDM is even closer to the accurate $\boldsymbol{\Gamma}$ following a single iteration of the contracted Schrödinger equation. The energy, as computed using $E=\mathrm{tr}\,\mathbf{K}\boldsymbol{\Gamma}$, is also somewhat better for the Valdemoro reconstruction than for the contracted best-fit reconstruction.

Finally, for the same numerical example, we have compared the three self-consistent iteration schemes that have been published in the literature. Of these we find that the method proposed by Colmenero and Valdemoro [11] gives the best results after a single iteration, although Mazziotti's procedure [19] is only slightly less accurate and should not be discounted on the basis of this calculation alone. Interestingly, the procedure that fares worst is the one currently employed by Valdemoro's group [12–14], in lieu of their earlier procedure.

In closing we should mention that although the Grassmann product ansatz is compact and elegant for formal manipulations, a great deal of redundancy is hidden within the sums over permutations that define the antisymmetrizers. For example, naive application of $\hat{\mathcal{A}}_4\otimes\hat{\mathcal{A}}_4$ to $\boldsymbol{\Gamma}^{i_1i_2}_{j_1j_2}\boldsymbol{\Gamma}^{k_1k_2}_{l_1l_2}$ results in $(4!)^2=576$ terms for each element of $\boldsymbol{\Gamma}\wedge\boldsymbol{\Gamma}$, but taking into account permutational antisymmetry of $\boldsymbol{\Gamma}$ this number can be reduced to a mere 18. Elements of $\boldsymbol{\gamma}^{\wedge 4}$ involve only 24 independent terms (not 576) and elements of $\boldsymbol{\gamma}\wedge\boldsymbol{\gamma}\wedge\boldsymbol{\gamma}^2$ can be expressed using 18 rather than $(3!)^2=36$ terms. The list goes on. Moreover, since each of these tensors is antisymmetric and also self-adjoint, many of its elements are not required. An example of this (further elaborated in the Appendix) is $\boldsymbol{\Gamma}$ itself: the only independent elements of $\boldsymbol{\Gamma}^{ij}_{kl}$ are those for which $i<j$, $k<l$, and $i\leqslant k$, and furthermore when $i=k$ only those elements with $j\leqslant l$ are independent.

To some extent we have incorporated these simplifying symmetries into our algorithms, although not in a systematic or exhaustive fashion. It is possible that in the future these or other symmetries may be applied systematically to obtain efficient algorithms not only for accomplishing the reconstruction but also for iterative solution of the CSE. In particular, Valdemoro's group [12–14] has spent a great deal of time developing such algorithms.

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APPENDIX: STORAGE REQUIREMENTS

Using the contraction formulas to avoid storage of 3- and 4-RDMs reduces storage requirements by the fourth power of the basis size, for a basis of spin orbitals. In fact we can be more explicit about memory requirements. Let us first introduce a basis set of orthonormal orbitals $\{\chi_1,\ldots,\chi_\nu\}$ and construct from them 2ν spin orbitals $\{\phi_1,\ldots,\phi_{2\nu}\}$, ordered such that $\phi_\kappa(\mathbf{r},\xi)=\chi_\kappa(\mathbf{r})\alpha(\xi)$ if $\kappa\leqslant\nu$ and $\phi_\kappa(\mathbf{r},\xi)=\chi_{\kappa-\nu}(\mathbf{r})\beta(\xi)$ for $\kappa>\nu$. Expanding the 2-RDM by spin components, we have

$$\begin{aligned}\Gamma(\mathbf{r}_1,\xi_1,\mathbf{r}_2,\xi_2;\mathbf{r}_1',\xi_1',\mathbf{r}_2',\xi_2')&=(\Gamma^{\alpha\alpha}_{\alpha\alpha})\alpha^*\alpha^*\alpha\alpha+(\Gamma^{\alpha\beta}_{\alpha\beta})\alpha^*\beta^*\alpha\beta\\&\quad+(\Gamma^{\beta\alpha}_{\beta\alpha})\beta^*\alpha^*\beta\alpha+(\Gamma^{\alpha\beta}_{\beta\alpha})\alpha^*\beta^*\beta\alpha\\&\quad+(\Gamma^{\beta\alpha}_{\alpha\beta})\beta^*\alpha^*\alpha\beta+(\Gamma^{\beta\beta}_{\beta\beta})\beta^*\beta^*\beta\beta,\end{aligned}\tag{A1}$$

where $\Gamma^{\alpha\beta}_{\alpha\beta}=\Gamma^{\alpha\beta}_{\alpha\beta}(\mathbf{r}_1,\mathbf{r}_2;\mathbf{r}_1',\mathbf{r}_2')$, for example, depends only on spatial coordinates. The notation for the two-electron spin functions is $\alpha^*\beta^*\alpha\beta=\alpha^*(\xi_1')\beta^*(\xi_2')\alpha(\xi_1)\beta(\xi_2)$, etc. There are 16 functions of this type, but as a result of the requirement $m_s(1)+m_s(2)=m_s(1')+m_s(2')$ for $\hat{S}_z$ eigenstates, only the six spin components listed above are nonzero. In fact only three of these are independent [42,43] and we may take the independent components to be $\Gamma^{\alpha\alpha}_{\alpha\alpha}$, $\Gamma^{\alpha\beta}_{\alpha\beta}$, and $\Gamma^{\beta\beta}_{\beta\beta}$. This follows readily from the permutational antisymmetry of $\boldsymbol{\Gamma}$, as we now demonstrate.

Expanding $\Gamma^{\alpha\beta}_{\alpha\beta}(\mathbf{r}_1,\mathbf{r}_2;\mathbf{r}_1',\mathbf{r}_2')$ in the $\{\chi_k\}$ basis we may represent this spatial kernel by its tensor of expansion coefficients, which we denote by $\boldsymbol{\Gamma}^{\alpha\beta}_{\alpha\beta}$. The tensor $\boldsymbol{\Gamma}^{\alpha\beta}_{\alpha\beta}$ has elements $(\boldsymbol{\Gamma}^{\alpha\beta}_{\alpha\beta})^{ij}_{kl}=\boldsymbol{\Gamma}^{i,j+\nu}_{k,l+\nu}$ for $i,j,k,l\leqslant\nu$, so, for example, we have

$$(\boldsymbol{\Gamma}^{\alpha\beta}_{\alpha\beta})^{ij}_{kl}=\boldsymbol{\Gamma}^{i,j+\nu}_{k,l+\nu}=-\boldsymbol{\Gamma}^{i,j+\nu}_{l+\nu,k}=-(\boldsymbol{\Gamma}^{\alpha\beta}_{\beta\alpha})^{ij}_{lk}.\tag{A2}$$

Similar relations exist among other spin components.

We wish to determine the number of independent elements in the three independent spin component tensors and thereby derive an explicit expression for storage requirements as a function of basis size. Consider first a typical element $(\boldsymbol{\Gamma}^{\alpha\alpha}_{\alpha\alpha})^{ij}_{kl}$, with $i,j,k,l\leqslant\nu$. Since $\boldsymbol{\Gamma}^{\alpha\alpha}_{\alpha\alpha}$ is self-adjoint and antisymmetric, we require only those elements with $i<j$, $k<l$, and $i\leqslant k$. Furthermore, when $i=k$ we need only the elements with $j\leqslant l$. The number of elements satisfying these requirements is

$$\begin{aligned}N_{\alpha\alpha\alpha\alpha}&=\sum_{i=1}^{\nu-1}\sum_{j=i+1}^{\nu}\left[\sum_{k=i}^{\nu-1}\sum_{l=k+1}^{\nu}1-\sum_{l=i+1}^{j-1}1\right]\\&=\frac{1}{8}\nu(\nu^3-2\nu^2+3\nu-2).\end{aligned}\tag{A3}$$

The second of the terms in brackets subtracts the number of elements with $i=k$ and $j>l$. Clearly $N_{\beta\beta\beta\beta}=N_{\alpha\alpha\alpha\alpha}$, and a similar exercise demonstrates that $N_{\alpha\beta\alpha\beta}=\nu^3(\nu-1)/2$. Adding these together we get the total number of independent elements for an arbitrary 2-RDM expressed in a basis set of 2ν spin orbitals:

$$N_{\mathrm{ind}}(\nu)=\frac{1}{4}\nu(\nu-1)(3\nu^2-\nu+2).\tag{A4}$$

This is the number of matrix elements that must be stored in order to solve Eq. (2.12) self-consistently. The tensor $\boldsymbol{\Gamma}$ contains a total of $16\nu^4$ elements, so as $\nu\to\infty$ the ratio of N_{ind} to the total number of elements in $\boldsymbol{\Gamma}$ approaches 0.046 875. For singlet states $\boldsymbol{\Gamma}^{\alpha\alpha}_{\alpha\alpha}=\boldsymbol{\Gamma}^{\beta\beta}_{\beta\beta}$, which further decreases the number of independent elements of $\boldsymbol{\Gamma}$.

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[1] A.J. Coleman, Rev. Mod. Phys. **35**, 668 (1963).
[2] A.J. Coleman and V.I. Yukalov, *Reduced Density Matrices: Coulson's Challenge*, Vol. 72 of Lecture Notes in Chemistry (Springer, New York, 2000).
[3] C. Garrod and J. Percus, J. Math. Phys. **5**, 1756 (1964).
[4] H. Kummer, J. Math. Phys. **8**, 2063 (1967).
[5] M.B. Ruskai, Phys. Rev. **183**, 129 (1969).
[6] E.R. Davidson, J. Math. Phys. **10**, 725 (1969).
[7] R.M. Erdahl, J. Math. Phys. **13**, 1608 (1972).
[8] T.L. Gilbert, Phys. Rev. B **12**, 2111 (1975).
[9] H. Kummer, Int. J. Quantum Chem. **12**, 1033 (1977).
[10] M. Nakata, H. Nakatsuji, M. Ehara, M. Fukuda, K. Nakata, and K. Fujisawa, J. Chem. Phys. **114**, 8282 (2001).
[11] F. Colmenero and C. Valdemoro, Int. J. Quantum Chem. **51**, 369 (1994).
[12] C. Valdemoro, Int. J. Quantum Chem. **60**, 131 (1996).
[13] C. Valdemoro, L.M. Tel, and E. Pérez-Romero, Adv. Quantum Chem. **28**, 33 (1997).
[14] C. Valdemoro, L.M. Tel, E. Pérez-Romero, and A. Torre, J. Mol. Struct.: THEOCHEM **537**, 1 (2001).
[15] H. Nakatsuji and K. Yasuda, Phys. Rev. Lett. **76**, 1039 (1996).
[16] K. Yasuda and H. Nakatsuji, Phys. Rev. A **56**, 2648 (1997).

[17] M. Nakata, M. Ehara, K. Yasuda, and H. Nakatsuji, J. Chem. Phys. **112**, 8772 (2000).

[18] K. Yasuda, Phys. Rev. A **59**, 4133 (1999).

[19] D.A. Mazziotti, Phys. Rev. A **57**, 4219 (1998).

[20] D.A. Mazziotti, Phys. Rev. A **60**, 4396 (1999).

[21] L. Cohen and C. Frishberg, Phys. Rev. A **13**, 927 (1976).

[22] H. Nakatsuji, Phys. Rev. A **14**, 41 (1976).

[23] H. Nakatsuji, Theor. Chem. Acc. **102**, 97 (1999).

[24] J.E. Harriman, Phys. Rev. A **19**, 1893 (1979).

[25] M. Rosina, in *Reduced Density Matrices with Applications to Physical and Chemical Systems* edited by A.J. Coleman and R.M. Erdahl, Vol. 11 of Queen’s Papers on Pure and Applied Mathematics (Queen’s University Press, New York, 1968), p. 369.

[26] M. Rosina, in *Many-Electron Densities and Reduced Density Matrices*, edited by J. Cioslowski (Plenum, New York, 2000), p. 19.

[27] A.J. Coleman and I. Absar, Int. J. Quantum Chem. **18**, 1279 (1980).

[28] R.D. Mattuck and A. Theumann, Adv. Phys. **20**, 721 (1971).

[29] D.A. Mazziotti, Chem. Phys. Lett. **326**, 212 (2000).

[30] D.A. Mazziotti and R.M. Erdahl, Phys. Rev. A **63**, 042113 (2001).

[31] F. Colmenero and C. Valdemoro, Phys. Rev. A **47**, 979 (1993).

[32] F. Colmenero, C. Pérez del Valle, and C. Valdemoro, Phys. Rev. A **47**, 971 (1993).

[33] W. Kutzelnigg and D. Mukherjee, J. Chem. Phys. **110**, 2800 (1999).

[34] C. Valdemoro, L.M. Tel, and E. Pérez-Romero, Phys. Rev. A **61**, 032507 (2000).

[35] C.F. Bunge, Phys. Rev. **168**, 92 (1968).

[36] O. Jitrik and C.F. Bunge, Phys. Rev. A **56**, 2614 (1997).

[37] R.E. Watson, Phys. Rev. **119**, 170 (1960).

[38] C.F. Bunge, Phys. Rev. A **14**, 1965 (1976).

[39] C.F. Bunge, Phys. Rev. A **17**, 486 (1978).

[40] W. Kutzelnigg and D. Mukherjee, Chem. Phys. Lett. **317**, 567 (2000).

[41] L.M. Tel, E. Pérez-Romero, C. Valdemoro, and F.J. Casquero, Int. J. Quantum Chem. **82**, 131 (2001).

[42] R. McWeeny and Y. Mizuno, Proc. R. Soc. London, Ser. A **259**, 554 (1961).

[43] J.E. Harriman, Int. J. Quantum Chem. **15**, 611 (1979).

[44] Nakatsuji and Yasuda [15] attribute the earliest derivation of the density equation to S. Cho, Sci. Rep. Gumma Univ. **11**, 1 (1962).